

WIDE-FREQUENCY-RANGE ATOMIC COMAGNETOMETRY TO SEARCH FOR SPIN-DEPENDENT INTERACTIONS BEYOND THE STANDARD MODEL

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by

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Oświadczenie

Ja niżej podpisany **Mikhail Padniuk** (nr indeksu: **1098895**), doktorant Wydziału Fizyki, Astronomii i Informatyki Stosowanej Uniwersytetu Jagiellońskiego oświadczam, że przedłożona przeze mnie rozprawa doktorska pt. "*Wide-frequency-range atomic magnetometry to search for spin-dependent interactions beyond the standard model*" jest oryginalna i przedstawia wyniki badań wykonanych przeze mnie osobiście, pod kierunkiem prof. dr. hab. Szymona Pustelnego. Pracę napisałem samodzielnie. Oświadczam, że moja rozprawa doktorska została opracowana zgodnie z Ustawą o prawie autorskim i prawach pokrewnych z dnia 4 lutego 1994 r. (Dziennik Ustaw 1994 nr 24 poz. 83 wraz z późniejszymi zmianami). Jestem świadom, że niezgodność niniejszego oświadczenia z prawdą ujawniona w dowolnym czasie, niezależnie od skutków prawnych wynikających z ww. ustawy, może spowodować unieważnienie stopnia nabytego na podstawie tej rozprawy.

Kraków, ...

.
(podpis doktoranta)

*One accurate measurement is worth
a thousand expert opinions*

Unknown author¹

¹This quote have been used by many historical figures of 20-th century, however, the author of this quote was searched by an inquiry in the Asked and Answered section of Book News, vol. XV, number 175, Philadelphia, March 1897. Therefore, it had been known long before.

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ABSTRACT

The problem of the dark matter remains unresolved for nearly a century. In recent years, the searches for possible interactions between dark matter and ordinary matter frequently involve techniques relying on atomic and molecular physics. In this context, special interest lies in optical magnetometry and related techniques, as nowadays, they provide the highest sensitivity to exotic spin couplings in a perturbation spectrum below 100 Hz.

The Global Network of Optical Magnetometers for Exotic physics searches (GNOME) is a distinct example of a worldwide experiment searching for exotic spin couplings with a help of optical magnetometry. Seeking for consistent noise pattern in the observed signals, the network is capable of detecting time-dependent interactions between ultralight dark matter and spins of ordinary matter, which may be, for example, induced by hypothetical astrophysical phenomena contested by axion dark matter.

This thesis presents the work on the development of a next-generation sensor for GNOME, which, over the last couple of years, has been built at the Marian Smoluchowski Institute of Physics at the Jagiellonian University in Kraków. The sensor is a self-compensating comagnetometer, which is based on the polarised mixture of alkali metals and ^3He atoms. For the dissertation, the comagnetometer was run in two-sensitivity-axis mode, providing more complete searches for exotic spin couplings, and was optimised for continuous measurements over extended periods of time (multiple months). As the result of the research, the system reached a sensitivity of 1.5×10^{-21} eV/ $\sqrt{\text{Hz}}$ in the case of the exotic couplings to neutron spins and 1.5×10^{-19} eV/ $\sqrt{\text{Hz}}$ in the case of the exotic couplings to proton spins. In addition to the research on the sensor, the thesis also presents the development of the novel optimisation-calibration procedure of the sensor operation. This procedure allows the sensor to be operated continuously for 24 hours, interrupted then with only relatively brief (250 s) calibration procedures. In turn, this procedure provides means for continuous multi-month operation with a duty cycle (ratio of up time to calibration-down time) exceeding previous experiments by at least an order of magnitude. Finally, the work presents a novel method for calibration of the comagnetometer, which enables an interpretation of the measurements results in arbitrary frequency range. As previously the constant frequency response of the comagnetometer was assumed, this new capability provides a quantitative change in the searches for exotic spin couplings, including ultralight-dark-matter searches. This may also alter some of the exiting limits on the exotic spin couplings, where constant response was assumed. The developed method was successfully tested on the rotation sensing during the author's visit to Helmholtz Institute in Mainz, where another next-generation GNOME sensor of GNOME was built.

STRESZCZENIE

Problem istnienia ciemnej materii pozostaje nierozwiązany od niemal stulecia. Współcześnie, liczne poszukiwania oddziaływań między ciemną, a zwykłą materią barionową wykorzystują metody oparte na technikach bazujących na fizyce atomowej i molekularnej. W tym kontekście szczególne zainteresowanie budzi optyczna magnetometria oraz komagnetometria atomowa, ponieważ obecnie to te techniki pozwalają na najdokładniejsze pomiary egzotycznych sprzężeń spinowych w paśmie częstotliwości poniżej 100 Hz.

Szczególnym przykładem eksperymentu wykorzystującego magnetometrię optyczną do poszukiwania oddziaływań z ciemną materią jest Globalna Sieć Magnetometrów Optycznych do Poszukiwań Fizyki Egzotycznej (ang. Global Network of Optical Magnetometers for Exotic physics search – GNOME). Sieć GNOME pozwala na detekcję zależności od czasu sprzężeń spinowych, które mogą być wygenerowane przez hipotetyczne obiekty astronomiczne składające się z ultralekkiej ciemnej materii, w tym w szczególności materii aksjonowej.

Niniejsza praca przedstawia wyniki badań nad rozwojem sensora nowej generacji dedykowanego dla eksperymentu GNOME, który był budowany w Instytucie Fizyki im. Mariana Smoluchowskiego Uniwersytetu Jagiellońskiego w Krakowie. Działanie tego czujnika, będącego samo-kompensującym się komagnetometrem atomowym, oparte jest o mieszaninę spolaryzowanych par metali alkalicznych oraz spolaryzowanego jądro ^3He . W ramach niniejszego doktoratu zbudowany komagnetometer pracujący w reżimie dwóch osi czułości, co umożliwia uzyskanie bardziej kompletnej informacji o potencjalnych egzotycznych sprzężeniach spinowych. Optymalizacja pracy tego urządzenia pozwoliła na pomiary egzotycznych sprzężeń do spinów neutronu z czułością na poziomie $1,5 \times 10^{-21} \text{ eV}/\sqrt{\text{Hz}}$ oraz $1,5 \times 10^{-19} \text{ eV}/\sqrt{\text{Hz}}$ w przypadku sprzężeń do spinów protonowych. W ramach doktoratu opracowano również nową procedurę zoptymalizowanej kalibracji urządzenia. Dzięki niej układ może być kalibrowany rzadziej niż co 24 godziny, z martwym czasem, podczas którego kalibrowane jest urządzenie, rzędu 250-s. Sprawia to, że nowa procedura umożliwia wielomiesięczne pomiary z martwym czasem aktywności urządzenia co najmniej o rząd wielkości krótszym od wcześniej stosowanych metod. Warto na koniec zauważyć, że przedstawiona praca opisuje nowatorską metodę kalibracji samo-kompensujących się komagnetometrów optycznych, która pozwala na interpretację wyników pomiarów w szerokim paśmie częstotliwości. Ponieważ we wcześniejszych badaniach zakładana była spektralnie niezmienna odpowiedź sensorów, to uwzględnienie charakterystyki spektralnej urządzenia stanowi jakościową zmianę w stosunku do prowadzonych wcześniej poszukiwań, a także może spowodować konieczność modyfikacji wcześniejszych limitów na niemagnetyczne sprzężenia spinowe. W celu potwierdzenia poprawności metody, została ona zweryfikowana przy użyciu pomiarów rotacji na układzie komagnetometru optycznego w Moguncji (Niemcy) podczas wizyty naukowej autora niniejszej pracy doktorskiej.

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STRUCTURE OF THE DISSERTATION

The presented dissertation provides a summary of the results of the authors' PhD studies along with the introduction into the topic of searches for exotic spin couplings with spin-based sensors in general and self-compensating comagnetometers in particular. It is also author's attempt to address multiple questions related to the topic that was born in discussions within the GNOME collaboration.

This dissertation contains six main chapters and a conclusion chapter. Chapter 1 provides a brief introduction to the topic of the axion dark matter that might generate interactions of exotic spin couplings and its searches with spin-based sensors. This is followed by Chapter 2, where the theory underlying the operation of spin-exchange-relaxation-free magnetometers and self-compensating comagnetometers. Chapter 3 provides a description of experimental setups of self-compensating comagnetometers built in Krakow and Mainz. Chapter 4 describes a collection of practical techniques for the optimisation and characterisation of self-compensating comagnetometers performance. The detailed procedure of the wide-bandwidth calibration of the comagnetometer is provided in Chapter 5, which also includes results of the experiments conducted during author visit at Prof. Budker's group at Helmholtz institute in Mainz. Chapter 6 discusses the performance of the Krakow comagnetometer. The thesis is summarised in the Conclusion (Chapter 7).

1

INTRODUCTION

1.1. BEYOND THE STANDARD MODEL

The last hundred years of modern physics development brought a great understanding of the Universe we live in. The **Standard Model of particle physics** (SM) is among the greatest successes of this development. This quantum field theory describes the whole “visible” matter in the Universe [1], dividing it into groups consisting of elementary particles: fermions and bosons. The particles may form more complex structures by four fundamental interactions: electromagnetic, weak, strong, and gravity.¹ The Standard Model was verified in multiple experiments involving accelerators of elementary particles, as well as multiple low-energy experiments and astronomical observations. The peak of this experimental survey was the observation of the Higgs boson in 2012 by the ATLAS collaboration in the Large Hadron Collider (LHC) at CERN [2, 3].

Despite the whole glory of mathematical consistency and experimental justification, SM has several problems. They imply the existence of yet undiscovered physics, often called **physics beyond the Standard Model**. The first significant “problem” of SM is **matter-antimatter asymmetry** observed in the Universe[4]. Within SM, each particle has its antiparticle, which possesses identical properties except for an opposite electric charge. The particle-antiparticle pair production is a process inevitably arising in the early Universe. However, astronomical observations show that the visible Universe consists entirely of ordinary matter. This suggests the existence of a **symmetry-breaking mechanism**, which is responsible for a different interaction with matter and antimatter. This asymmetry eventually led to the excess of ordinary matter in the later Universe, which is also known as **the problem of baryogenesis**.

Violation of the combined charge-conjugation-parity-reversal (CP) symmetry is thought to be responsible for the matter-antimatter asymmetry. Such a violation is observed in many processes (e.g., particle decays) governed by the weak interaction and is

¹The Standard Model does not explain gravity, however, there is a hypothesis of graviton – an elementary particle responsible for the gravitational interaction. It should be noted though that gravity does not have an important role in microscopic systems due to its very small strength and it is important and taken into account mostly at a cosmic scale.

consistent with SM. However, the scale of CP violation in SM is not sufficient to explain the observable difference in the matter-antimatter abundance. Moreover, SM predicts violation of the CP symmetry not only by weak but also by strong interactions.² This contradicts experimental evidence that consistently returns an immeasurably small CP violation (zero within the error bars) in processes governed by the strong interaction. This so-called **strong CP problem** is the second major issue of SM and one of the unsolved mysteries of modern physics.

The most popular proposal to solve the strong CP problem is based on **violating the so-called Peccei-Quinn symmetry**, an anomalous symmetry of Nature [5], which is related to the existence of a hypothetical particle called **axion** [6]. Such a **quantum electrodynamics (QCD) axion**³ is a spin-zero pseudoscalar particle whose mass is not set by theory and must be identified experimentally. The idea of the QCD axion was further extended to the so-called **axion-like particles (ALPs)** [7–9], which do not serve the purpose of solving the strong CP problem, but arise naturally in most string-theory extensions of SM.

The indications of physics beyond SM do not come only from the SM problems. Multiple astronomical observations suggest that the Universe contains much more matter than is “visible”, i.e., that absorbs, emits, or scatters electromagnetic radiation. For example, inconsistency between the mass of visible matter and the velocity dispersion of stars’ rotation in galaxies, the so-called **galaxy-rotation problem**, implies that the galaxy should contain more gravitationally interacting, yet electromagnetically noninteracting matter [10–14]. This matter of unknown nature is called **dark matter (DM)**.

Another indication of such a dark matter comes from the gravitational lensing of electromagnetic radiation in seemingly empty space [15]. The distinct pattern of the gravitational lensing phenomenon allows conclusive recognition of the gravitational deflection of the light propagation trajectory. However, there is a lack of any other evidence for the presence of known astronomical objects in the place where a significant mass of matter is expected. Therefore, the origin of this massive matter remains unknown, which is consistent with the dark matter hypothesis.

Another hint of dark-matter existence comes from theoretical considerations of the formation of galaxies, which suggest that these structures could not arise without dark-matter density fluctuation [15, 16]. The agreement between theoretical simulations of the galaxy evolution and astronomical observations requires the assumption of the dark matter existence from the very early stages of the Universe evolution.

Despite the absence of direct evidence of the DM existence, it is possible to infer some of its generic properties based on astronomical observations. For example, the analysis of Cosmic Microwave Background (CMB) radiation enables the estimation of the DM density, which accounts for 26% of the total energy in the Universe, while ordinary matter makes up only 6% (the remaining 68% is in the form of **dark energy**) [17]. The motion of stars in the Milky Way allows the estimation of the local density of dark matter, which is expected to be 0.3-0.4 GeV/cm³. This means that the DM density is

²In fact, the violation of the CP symmetry in the strong interaction is expected to be significantly larger than in the case of the weak interaction.

³The QCD (quantum chromodynamic) axion is the boson that is introduced to solve the strong CP problem. In contrast to axion-like particles described later, it has a fixed relation between its mass and coupling strength.

about five orders of magnitude larger than the average density of the electromagnetically interacting matter in the Universe. Moreover, the clustering and evolution of galaxies suggest that dark matter is “cold” (nonrelativistic), since, otherwise, it would escape the galactic gravitation pull. Finally, the observation of the Bullet Cluster (1E 0657-56) shows that DM lacks any significant nongravitational interaction with itself or ordinary matter [18]. These properties of DM provide a framework for theoretical investigations of possible explanations of the matter and suggest experimental searches for its direct detection.

Historically, DM was expected to be composed of **weakly interacting massive particles** (WIMP)[1, 4]. However, over three decades of a search for experimental evidence of WIMPs with both high-energy accelerators and low-energy experiments did not lead to any positive results [19–21]. This triggered interest in alternative candidates. Nowadays, a great variety of new exotic particles are under meticulous consideration from both theoretical and experimental perspectives. A partial list of the candidates includes the superWIMPS [22], which are hypothetical particles being a product of the WIMPS decay; sterile neutrinos[22–24], the hypothetical type of the neutrinos, which interact much weaker than known types of the neutrinos with the normal matter; dark photons [25], the ultralight particle, carrying the interaction between dark matter particles in a way similar to the photon in case of the electromagnetic interaction; massive (astronomical) compact objects (MACHOs)[24, 26, 27], a group of hypothetical astronomical objects, which are invisible in the electromagnetic spectrum of astronomical observations, which might be represented by known phenomena such as small mass black holes or neutron stars as well as by exotic objects consisted of non-barionic dark matter. However, recently, the QCD axion and ALPs⁴ are considered as prime candidates for DM particles [16]. This is so as they fit well in some DM concepts, considering DM to be composed of low-mass bosons.⁵ The search for such DM can be performed by seeking effects that the matter would impose on ordinary matter. The scope of this thesis is related to a particular subgroup of such searches where one uses spin-based sensors [28].

1.2. SEARCHES FOR AXION DM

Assuming axions to be a significant component of DM, they should be present throughout our galaxy. Their direct detection, however, requires the axions interaction with ordinary matter. Assuming that they are pseudoscalar (spin-0) particles,⁶ there are several ways in which they can interact with ordinary matter. These interactions can be described by the following phenomenological Hamiltonian [16]

$$H = \chi_{a\gamma} \int a \mathbf{E} \cdot \mathbf{B} dV + \sum_{\eta} \chi_{\eta EDM} a \hat{\mathbf{S}}_{\eta} \cdot \mathbf{E} + \sum_f \chi_f \nabla a \cdot \hat{\mathbf{S}}_f, \quad (1.1)$$

where a is the axion field in energy units, $\chi_{a\gamma}$ is the axion coupling to electromagnetic field, $\mathbf{E}$ and $\mathbf{B}$ are electric and magnetic fields, respectively, χ_f is the axion coupling

⁴QCD axions and ALPs have a similar mechanism of interactions with ordinary matter, wherefore below both types of particles will be referred to as axions and specify the axion type when necessary.

⁵Due to its mass, the ultralight matter manifests as coherent fields called **ultralight bosonic fields**.

⁶There are models that include axions with vector (spin-1) and scalar properties [16], but their searches require experimental strategies, reaching beyond the scope of this dissertation.

strength to the fermions, with the index f denoting a fermion type, $\hat{\mathbf{S}}_f$ is the spin of the fermion, $\chi_{\eta EDM}$ is a strength of a permanent electric dipole moment (EDM) induced in a hadron η by the QCD axion and $\hat{\mathbf{S}}_\eta$ is the spin of hadron η . The form of this Hamiltonian implies possible axions-detection strategies. Specifically, the interaction between axions and electromagnetic field, as described by the first term of the Hamiltonian (1.1), may result in the conversion of axions into photons. This could lead to an unexplained excess of electromagnetic radiation in the Universe. Such an excess is being searched for in experiments that look for photons inside cavities placed in a strong magnetic field [29, 30], astronomical observations searching for anomalous electromagnetic radiation [31, 32], and “shining through the wall” experiments [33]. As the frequency of the anomalous photons corresponds to the Compton frequency, hence the mass of the axions [16], the range of detectable electromagnetic fields determines the mass range of the sought-after axions.

The second term of the Hamiltonian (1.1) describes the interaction of the electric field with EDM of hadrons which is induced by the interaction with QCD axions. The **axion field** can generate a time-varying EDM in nucleons with an oscillating frequency, corresponding to the Compton frequency of the axion field [34]. The direct searches for such interactions are realised with nuclear magnetic resonance (NMR) techniques by Cosmic Axion Spin Precession Experiment (CASPER) [35] and with the cold neutrons-mercury comagnetometry in the nEDM experiment [36].

The last term of the Hamiltonian (1.1) is responsible for the interaction of the axion field with spins of fermions. Specifically, the spins are expected to couple to the spatial gradient of axion fields. From an experimental standpoint, such interactions have a form of anomalous pseudomagnetic field perturbing the spin state of fermions and can be detected in atomic and molecular systems as anomaly energy-level shifts generated by the field. Such a coupling is considered in this thesis and hence is discussed in details in Sec. 1.3.

The weakness of the coupling to ordinary matter may not be the only challenge in the direct observation of axion DM. Although astronomical observations suggest the average local density of DM in the Milky Way, the DM density on Earth may differ significantly from this value [37, 38]. In fact, the distribution of axion DM may be influenced by many factors that cannot be calculated theoretically, including the axion generation mechanism, the mass of the particles, or their self-interaction [16]. Moreover, some models predict that axion may form stable topological defects (e.g., domain walls) that have been preserved until now [39]. In this case, the effects of axions on ordinary matter may vary over time, and coupling is observed only during the interaction with the defect (**transient coupling**).⁷ The transient character of such exotic spin perturbations significantly complicates detection of axion DM.

Based on astronomical observation and theoretical considerations, the axion mass is constrained between 10^{-22} and 10^{-2} eV [16]. In the case of the QCD axion, the connection to the strong interaction narrows the constraint to the range between 0.025 and 0.5 meV. However, as the mass of ALPs and their coupling strength to ordinary matter are highly model-dependent, the parameters must be determined experimentally.

⁷The spatial orientation of the wall and Earth motion (axions are being assumed to be motionless in galactic frame) determine the spatio-temporal gradient of the field and hence its interaction with spins.

The small mass of axions results in de Broglie's waves with the wavelength at macroscopic and even cosmic scales. As axions are bosons, such a long wavelength leads to a huge occupation number of each field mode, which makes it more suitable to treat the axions as a classical field rather than an ensemble of particles. The wavy nature of axions may lead to coherent and incoherent interference of the axion field, manifesting as their clustering of DM, which may result in highly inhomogeneous distributions at the Earth scale [16]. Although interaction with axions within such structures could be detected with laboratory experiments, their identification would be challenging due to the time-dependent character of the signal. This problem can be addressed using a network of spatially separated sensors whose measurements are synchronised in time. An example of such an experiment is the Global Network of Optical Magnetometers for Exotic physics searches (GNOME) [40, 41]. This thesis describes the research performed within the framework of GNOME activities. More on the subject, is described in in Sec. 1.3.

The inhomogeneous density distribution of the axion field is not the only possible source of the axion-field gradients. For example, the axion field may exhibit stochastic fluctuations, leading to **stochastic exotic spin couplings** [42]. Such interactions would then manifest as an excess of noise registered by a detector (more details in Sec. 1.4.3). The stochastic fluctuations, however, can be identified with a network of synchronised sensors (e.g., GNOME) where they would manifest as cross-correlations between the sensor readouts [43].

Time-dependent interactions with axion DM may also be generated by bursts of the axions **produced in cataclysmic astrophysical events** (e.g., binary black-hole merger) [44]. In that case, a jet of particles may be generated, and it may propagate through space with ultrarelativistic velocity. Reaching Earth, it may couple to spins inducing pulsed oscillating signals in atomic magnetometers (more details in Sec. 1.4.3). Even though the signals are recorded in individual sensors, a set of sensors may enhance the possibility of detecting the pulse by recording a consistent signal (specific spatiotemporal pattern) across the whole network. Moreover, synchronising the exotic signal with traditional astronomical observation channels opens new capabilities of multi-messenger astronomy.

1.3. OPTICAL MAGNETOMETRY FOR FUNDAMENTAL PHYSICS

1.3.1. OPTICAL MAGNETOMETERS

Optical magnetometry refers to a collection of techniques that utilises the interaction between light and atoms (or atom-like systems) to measure magnetic fields [45]. In **optical magnetometers**, also known as **optically-pumped magnetometers (OPMs)**, light first polarises atoms, which spin polarisation next evolves due to external magnetic field(s). This evolution modifies the optical properties of the atoms, which then affect the light propagation. This way measurements of such light parameters as its intensity or polarisation provide quantitative information about the magnetic field. Nowadays, optical magnetometers offer the most sensitive magnetic-field measurements in a frequency range between DC and 100 Hz.⁸

The operation of the optical magnetometers is fundamentally based on the interaction of a spin with the magnetic field. The interaction of a non-zero spin $\mathbf{S}$ with a

⁸Here, comagnetometers operating at low-magnetic field environments are under consideration.

magnetic field, $\mathbf{B}$ is governed by the **Zeeman Hamiltonian**

$$\hat{H}_z = -\mu_s \mathbf{B} \cdot \hat{\mathbf{S}}, \quad (1.2)$$

where μ_s is a proportionally constant between the spin and the magnetic moment μ .

Comparison of the third term of Eq. (1.1) with the form of the Zeeman Hamiltonian [Eq. (1.2)] reveals that the fermion-spin coupling with the axion-field gradient is similar to the coupling of the spin to the magnetic field. Hence, the exotic spin couplings from the axions may be considered as an additional pseudomagnetic field $\mathbf{b}_f$ and the Hamiltonian describing the interaction may be written as

$$\hat{H}_{af} = -\mu_s \left(-\frac{\chi_s \hbar c}{\mu_s} \nabla a \right) \cdot \hat{\mathbf{S}} = -\mu_s \mathbf{b}_f \cdot \hat{\mathbf{S}}, \quad (1.3)$$

where χ_s considers both the spin content of $\hat{\mathbf{S}}$ and the coupling strength of the axions to fermions, which spins impact $\hat{\mathbf{S}}$. Naturally, if the spin $\hat{\mathbf{S}}$ is a net spin arising as a combination of spins of other particles, including electrons, protons, or neutrons, such a spin is sensitive to the gradient of the axion field

In the case of ALPs and their interaction with spins, higher orders of interaction are also possible. In particular, the quadratic coupling between the axions and spins can be considered [39].⁹ It should be noted that in this case, the exotic spin coupling raises due to the coupling of the spins to the gradient of the square of the axion field. This coupling can be also presented in the form of the pseudomagnetic field $\mathbf{b}_{qf}$

$$\hat{H}_{aaf} = -\mu_s \left(-\frac{\chi_{qs} \hbar c}{\mu_s} \nabla a^2 \right) \cdot \hat{\mathbf{S}} = -\mu_s \mathbf{b}_{qf} \cdot \hat{\mathbf{S}}, \quad (1.4)$$

where χ_{qs} stands for the quadratic coupling constant.

The analogy between pseudoscalar spin coupling of the magnetic field and axion-field gradient shows that optical magnetometers can be used to search for both linear and quadratic exotic spin couplings. However, unlike the magnetic field, axion gradients only couple to neutron, proton, and electron spins and do not couple to any orbital angular momentum [28]. Therefore, predicting the response of the magnetometer to a given type of exotic spin coupling requires careful consideration of the spin structure of utilised atoms. This aspect of the couplings is discussed in more detail in Sec. 2.1.

1.3.2. COMAGNETOMETERS

Although OPMs proven their usefulness in searches for exotic spin couplings [46], their high magnetic-field sensitivity results in practical limitations on the sensitivity to non-magnetic couplings (magnetic noise spoils the sensitivity to possible exotic spin interactions). To alleviate this problem, one can implement a technique of **comagnetometry**. Such a technique is based on simultaneous magnetic-field measurements, using multiple spin ensembles and successive comparisons of the readouts coming from the systems. The comagnetometers generally use at least two atomic species of different spin constitutions (e.g., alkali-metal vapour and noble gas) to distinguish the known magnetic perturbation from non-magnetic ones. This ability of distinguishing different types

⁹This is an alternative, usually more speculative, way of coupling (portal) of the spins to axion DM.

of couplings is responsible for optical comagnetometry application in many tests of fundamental physics, searches for spins beyond SM, or rotation sensing [47].

The simplest example of a comagnetometer is a **gradiometer**.¹⁰ The subtraction of the readouts of two magnetometers placed at a close distance gives information about the gradient of the magnetic field and allows the suppression of a common magnetic background. Optically-pumped gradiometers have a wide range of applications in geophysics, biomagnetism, defects identification, low-field NMR, etc. [45].

An example of a comagnetometer with spatially separated spin samples is a system exploiting ^{199}Hg to search for EDM [48]. In this experiment, four chambers filled with the optically pumped ^{199}Hg vapour were put in the same magnetic but opposite electric fields. Comparison of the Larmor frequencies of ^{199}Hg from four adjoining magnetometers allow one to maximise sensitivity to EDM by suppressing the influence of the magnetic field and its gradients. This comagnetometer provided the most stringent limit on permanent EDM of ^{199}Hg [49–51].

In comagnetometry, the utilisation of overlapping spin ensembles allows one to improve the compensation of effects related to the magnetic field (gases occupy the same volume, so in the ideal case, they can completely reject the magnetic contribution from the comagnetometer readout). A specific example of such a comagnetometer is based on a mixture of two noble gases [52]. The gases have different nuclear spins (they could be two isotopes of the same element or two different atomic species), so their magnetic precession frequency in a given field is different. To observe such precession, both nuclear spin species have to be polarised, which may be achieved through spin-exchange collisions with optically polarised alkali atoms that are also present in a container hosting the noble gases. Besides the polarisation of the noble gases, the alkali atoms may be also used to detect their nuclear-spin precession. In this case, one explores good shielding of the nuclear spins of the noble gases from the external environment, i.e., the alkali atoms only weakly perturb the noble-gas spin state, and hence their Larmor precession can be measured without significant effect on the noble gas.¹¹ The ratio between the Larmor frequencies of the two noble gases is determined by their well-known gyromagnetic ratios so that any deviation from the known ratio indicates the presence of a perturbation that couples differently to the magnetic moments of the noble gas spins.¹² Thereby, the **noble-gas comagnetometry** can be used to search for exotic spin interactions.

Interestingly, different species or isotopes are not needed to perform comagnetometric measurements. There are implementations in which one species is used for the measurements. In that case, however, transitions among the states associated with different spin constitutions are used [53–55]. For example, in a so-called **liquid-state nuclear comagnetometer**, exploiting ultra-low-field nuclear magnetic resonance measurements of ^{13}C -labelled acetonitrile ($^{13}\text{CH}_3\text{CN}$), two reference frequencies are observed [56]. Because the signals originate from the same molecule, this approach is extremely efficient

¹⁰Usually, optically pumped gradiometers are not considered as comagnetometers, but they fit well into the heuristic definition of the comagnetometers given above.

¹¹Note that while the alkali-metal atoms work as an interface for the magnetometry of the noble gases, in principle, the noble-gas Larmor precession can be measured by an external magnetic-field sensor (see, for example, Ref. [47]).

¹²In real experiments, the difference could be also caused by systematic errors and some higher-order corrections to the spin evolution.

in cancelling various systematic effects (e.g., those related to the magnetic-field gradients). In particular, this comagnetometer was used to set a limit for nuclear coupling strength of ALPs of masses between 10^{-22} and 10^{-17} eV [56].

Utilizing a strongly coupled system of two different spin species is also a viable option for comagnetometry. A specific example of such a comagnetometer is the **noble-gas-alkali-metal comagnetometer** [57]. Similarly to noble-gas comagnetometers, in this system, the noble gas is polarised via spin-exchange collisions with optically pumped alkali-metal vapour and alkali atoms are also used for monitoring the system state. However, in contrast to the comagnetometer with two noble gases, here the alkali-metal and noble-gas polarisations are strongly coupled via the Fermi contact interaction[57]. This strong coupling is achieved through a careful adjustment of an external magnetic field, i.e., the field is adjusted to minimise the difference in the Larmor frequencies of the two species [57]. It turns out that under such conditions, the noble-gas polarisation can follow low-frequency changes of the magnetic field and hence compensate for the torque generated in electron spins by magnetic field interaction, making it effectively insensitive to the low-frequency transverse magnetic field. This mechanism is called the **self-compensation**. Importantly, the self-compensation does not work for perturbations, which have a different than the magnetic-field coupling strength to nuclear and electron spins. Such interactions (both electron and nuclear) result in non-zero torque generated on the electron spins, changing their orientation, which is then detected optically, leading to the high response amplitude in the case of the non-magnetic spin couplings. Suppression of the sensitivity to magnetic fields is not the only advantage of the comagnetometers in the context of searches for exotic spin couplings. The possibility of employing a high-density hyper-polarised nuclear sample such as noble gases, with a readout through optically accessible alkali atoms, allows one to achieve an extremely high sensitivity to the nuclear spin couplings. The high response amplitude to non-magnetic couplings with simultaneous suppression of the low-frequency magnetic field leads to the gain of the sensitivity to exotic spin couplings. This system has been used for searches of the fifth force, preferred frames and exotic spin couplings due to the dark matter particles [47]. In the scope of this work, the applications of the self-compensating comagnetometers in searches for transient and time-dependent manifestations of axion DM are explored. The theory of the self-compensating comagnetometer is discussed in details in Sec. 2.2.

1.3.3. MAGNETIC SHIELDING OF SPIN ENSEMBLES

To achieve high sensitivity to non-magnetic couplings, OPMs and comagnetometers should be shielded from ambient magnetic fields. Primarily, this is achieved by placing the devices inside passive **magnetic shields**. There are two main types of the magnetic shields, classified based on the shielding process: superconducting and ferromagnetic. In the first class, the magnetic field inside the shield is compensated due to the Meissner effect [45, 58]. In the case of ferromagnetic shields, the shielding mechanism depends on the frequency of the magnetic field. For quasi-stationary magnetic fields (below 10 Hz), the magnetic field is shielded due to the high permeability of shield material, while higher-frequency fields are shielded predominantly due to the skin effect [45].

Searches for exotic spin couplings with OPMs or optical comagnetometers require understanding the interaction between exotic couplings and magnetic-shield material. As electrons are responsible for shielding the exotic fields (nuclear couplings do not affect by the shielding), coupling of the fields to the particles needs to be only considered. More specifically, the ferromagnetic shields will not affect the measurements of AC exotic field since the skin effect involves the interaction of the field with the electric charge of the electrons. The superconducting shields would not affect measurements at all frequencies since the Meissner effect does not involve the interaction with electron spins.

The situation is more complicated in the case of ferromagnetic shields in measurements of low-frequency couplings since shielding of the slow magnetic fields relies on the ferromagnetic nature of the materials, which arises from the self-ordering of the electron spins of the atoms in the material. [59]. Hence, in this regime, the interaction with the electron spins plays a key role in the shielding of the magnetic field. Exotic couplings to electron spins of the shielding material will lead to the induction of the magnetic field inside of the shield. The induced magnetic field will have the orientation opposite to the orientation of the pseudomagnetic field generated by the exotic coupling. The induced magnetic field must be taken into account in the measurements performed with the magnetometers since it will compensate for the exotic perturbations of the sensor similarly as for the magnetic field, and therefore, it might compromise the result of the measurements. This is not so much of a problem for searches with comagnetometers since, in that case, the response of the system to the magnetic field and non-magnetic perturbations are different, and the magnetic field response is suppressed. This is an additional argument in favour of using the comagnetometers for searches of exotic spin-dependent phenomena.

1.3.4. THE SCOPE OF THIS WORK

This work mainly focuses on implementing self-compensating magnetometers into the GNOME experiment (which will be discussed in more detail in the subsequent section). Along with the self-compensating (SERF) comagnetometers, it also discusses the spin-exchange-relaxation-free magnetometers as a closely related topic. The SERF magnetometer is interesting within the scope of this work for two main reasons. First, several GNOME stations are equipped with this type of a sensor, which is the most sensitive among all OPMs (both in magnetic and non-magnetic field sensing). Therefore, discussing the sensor is a good opportunity to demonstrate the capabilities of the current GNOME experiment. Second, the SERF mechanism is also used in noble gas-alkali atom comagnetometer considered in this work (alkali atoms are subjected to the same regime of fast spin-exchange collisions as in the conventional SERF magnetometer).

The self-compensating comagnetometer, which will be used in the next generation of the GNOME experiment (Advanced GNOME), is an advanced optical sensor, that will allow increasing capabilities of the exotic-interaction searches. Specifically, it is capable of gaining 6-8 orders of magnitude improvement in sensitivity to nuclear exotic spin couplings. Moreover, it allows overcoming the problem of the suppressed sensitivity to electron exotic spin couplings due to the interactions with the magnetic-shield electrons (see Sec. 1.3.3), and therefore extend the range of targets, which might be searched with GNOME. Unlike ordinary magnetometers, self-compensating comagnetometers have

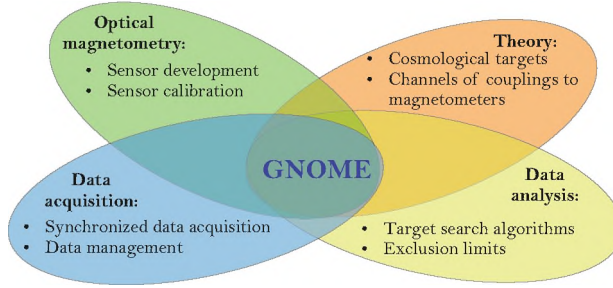


Figure 1.1: The main activities realised within GNOME.

the outstanding feature of having qualitatively different responses to magnetic and non-magnetic spin couplings, which opens a potential possibility for improving the effectiveness of GNOME data analysis methods, and, as a result of that, the overall sensitivity of the network to various forms of axion DM.

1.4. DARK MATTER SEARCHES WITH GNOME

The Global Network of Optical Magnetometers for Exotic physics searches (GNOME) is a worldwide collaboration whose prime goal is to search for ultralight dark matter (e.g., axions and axion-like particles). The collaboration activities revolve around the development of theoretical models of exotic spin couplings that can be searched with GNOME, sensors with enhanced sensitivity to non-magnetic spin couplings, and means of data analysis. These activities are summarised in Fig. 1.1.

A network of sensors, such as GNOME, has two main advantages over a single sensor. The spatial separation of the sensors allows one to distinguish local noise from the effects of global perturbations and, in such a way, to identify transient signals that may be associated with momentarily exotic spin couplings at a global scale. Moreover, information about the precise locations of the sensors and the timing of signals recorded by each sensor enables the identification of the direction and speed of propagation of the perturbation. This may be used to test various models predicting the structuring of dark matter.

In the following sections, various theoretical models concerning the spatial distribution of ultralight dark matter that can be searched with GNOME are discussed. The status of current (GNOME) and future (Advanced GNOME) searches are also presented.

1.4.1. CURRENT STATUS OF GNOME

Current GNOME consists of more than ten optical magnetometers located in North America (Berkeley, Hayward, Los Angeles, Oberlin, Lewisburg), Europe (Mainz, Moxa, Kraków, Belgrade), Asia (Hefei, Beijing, Daejeon), and Australia (Canberra) (Fig. 1.4). Different types of alkali-metal magnetometers, including AM NMOR, M_x , and SERF magnetometers, are used in the network. Aside from their types, the magnetometers also differ in atomic species (Rb, K, and Cs) used to search for exotic spin couplings. The sensitivity of sensors varies between 30 and 100 $\text{fT}/\sqrt{\text{Hz}}$ in a bandwidth between DC and 100 Hz.

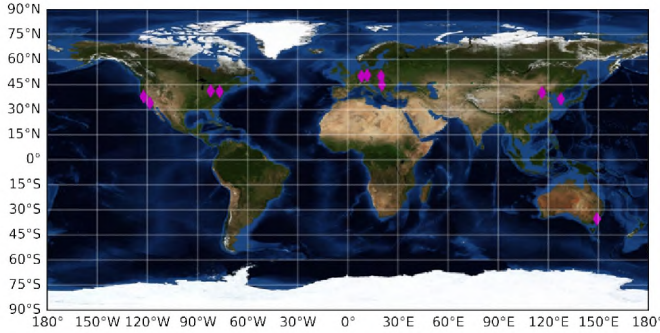


Figure 1.2: Geographical distribution of GNOME stations. Currently, the GNOME network covers four continents: North America (Berkeley, Hayward, Los Angeles, Oberlin, Lewisburg), Europe (Mainz, Moxa, Kraków, Belgrade), Asia (Hefei, Beijing, Daejeon), and Australia (Canberra) .

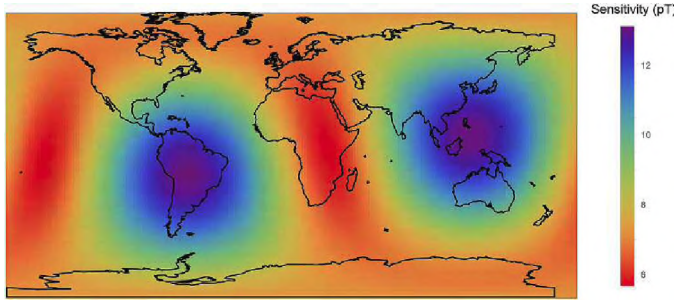


Figure 1.3: Simulated directional sensitivity of first-generation GNOME. Based on Ref. [61].

All the devices are vector sensors with their sensitivity direction aligned along an applied external magnetic field. It should be stressed that the distribution of these sensitive directions translates into an angular sensitivity of the network [60]. In fact, the network does not homogeneously cover the whole celestial sphere, and up to 50% of sensitivity modulation is observed depending on the direction (Fig. 1.3).

All GNOME magnetometers are placed inside multi-layer (ferromagnetic) magnetic shields, which are used to isolate the sensors from the ambient fields. As discussed in Sec. 1.3.3, this deteriorates the capabilities of the magnetometers to search for electronic non-magnetic spin couplings. Therefore GNOME data collected so far can be used only for tests of nuclear exotic spin couplings. Specifically, in alkali atoms, the nuclear spin is determined by a valence proton, therefore, current GNOME using optical magnetometers is predominantly sensitive to proton exotic spin couplings.

In addition to the optical magnetometer, each station is equipped with a system monitoring various physical parameters (the so-called Sanity Box). It is based on Arduino microcontroller and its aim is to flag data corrupted by some conventional perturbations. Specifically, the system monitors external sensors (e.g., flux magnetometer and accelerometer, and gyroscope) placed next to the magnetic shield. It is also equipped with additional inputs, which can be used to monitor user-defined signals (e.g., laser er-

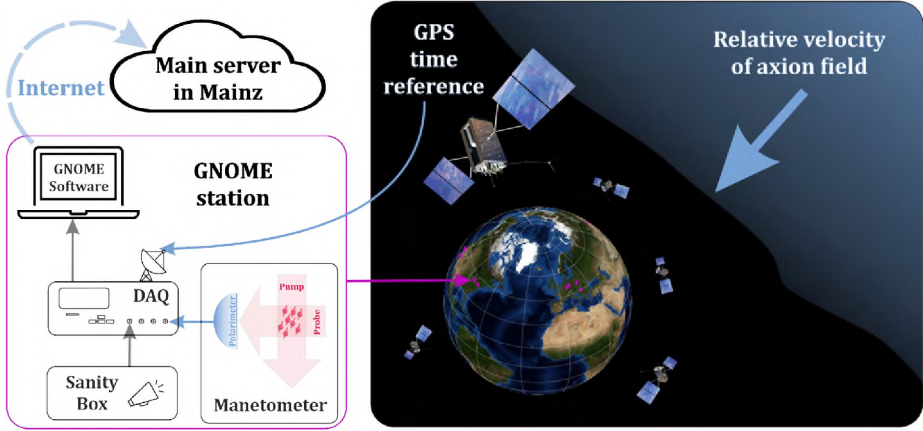


Figure 1.4: Schematic of the GNOME infrastructure. The output of each magnetometer is time-stamped with the GPS time and recorded using the GPS data acquisition system (DAQ). The signals are measured along with the signals recorded by the “sanity box” monitoring conventional experimental parameters (temperature, acceleration, external magnetic field). All data are stored at a local computer but also uploaded to a GNOME main server in Mainz, where signals from all stations can be accessed and processed.

ror signal, which provides information about any mode jumps). If anything unexpected happens to the experiment (abrupt motion of an optical table, a laser mode jump, spike in an external magnetic field, etc.), the magnetometer data sent to a local computer is flagged as suspicious (insane).

The main sensor output, which may contain information about possible exotic spin couplings, measured along the sanity signals, is sent to a GNOME data acquisition system (the so-called GPS box) [62]. It allows one to record timestamped signals using Global Positioning System (GPS) time, i.e., the only time reference broadly available at whole Earth. This data acquisition system ensures a time precision of the samples at a level of 10^{-7} s, though its sampling rate ranges between 1 S/s to 1024 S/s (in multiplies of two). To avoid losing of the data (in a typical measurement the GPS box can work independently for about 24 hours and then it overrides the data), the data recorded by the acquisition system is automatically transferred to a local computer, where it is stored using and HDF5 data format. This data is then sent to a main GNOME server located in Mainz, Germany, where data from all stations can be accessed and used for further analysis.

In order to calibrate the sensors, every hour a train of magnetic-field pulses of 100-pT amplitude and frequencies ranging between 1 and 100 Hz is applied to the magnetometers (field generated inside the magnetic shields). The analysis of the magnetometers’ responses enables monitoring the sensitivity and bandwidth of the devices and allows one to counteract any changes in the performance (by adjusting system parameters), which is crucial for long-term operation of the network.

Heretofore, GNOME had a series of a scientific runs, which lasted between 24h and 11 months (Fig. 1.5). The term **scientific run** is used for the periods of continuous data acquisition with a whole network. It separates scientific measurements from test runs

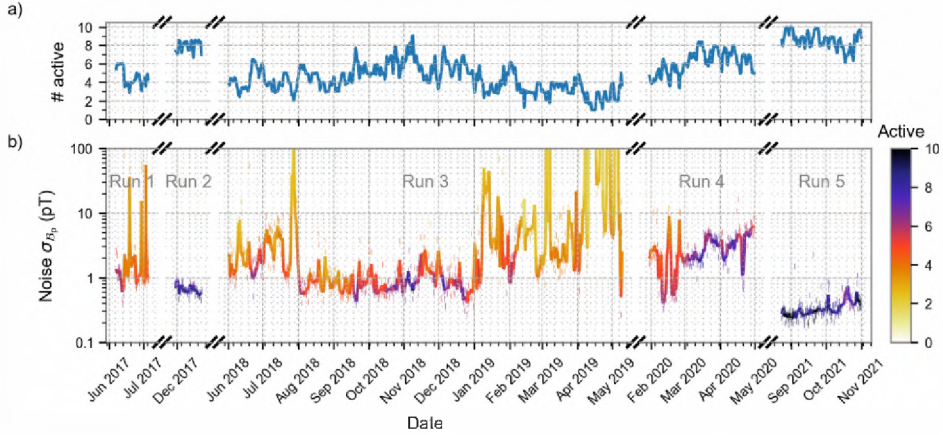


Figure 1.5: Sensitivity of GNOME over five scientific runs conducted to date. The top plot presents a number of active stations, and the bottom plot presents the sensitivity of the network [41].

and maintenance periods which are necessary between science runs.

1.4.2. ADVANCED GNOME

In the first incarnation of the GNOME project, optical magnetometers were chosen as sensors to search for dark matter because of their simplicity and accessibility. In fact, they allowed the setting up of the infrastructure for the measurements and data analysis protocols. Nonetheless, their replacement with sensors better suited for searches for the non-magnetic spin couplings, such as comagnetometers, were envisioned right from the beginning of GNOME [40]. The main motivation for the implementation of such sensors comes from the desire to limit magnetic noise and hence improve the sensitivity to non-magnetic spin perturbations. Specifically, the noble-gas-alkali metal comagnetometers, which have been described briefly in Sec. 1.3.2 and will be discussed in more detail in the next chapter (see Sec. 2.2), will be used in the next generation of the experiment called Advanced GNOME.

Although the noble-gas comagnetometers fit well in the concept of the GNOME studies, the actual implementation of the devices requires addressing a range of important technical problems. For example, the operation of the GNOME station requires a periodic calibration routine. Such a routine has to be quick and give information about the frequency response of the system to non-magnetic spin couplings. In the case of ordinary magnetometers, such validation can be done by application of a magnetic field as its frequency response to the magnetic and non-magnetic perturbations differ by the constant scaling factor. The comagnetometer responses to magnetic, non-magnetic nuclear and non-magnetic electron perturbations are qualitatively and quantitatively different, and direct application of the non-magnetic perturbation is much more challenging than the application of the magnetic field. Therefore, there is a need for a suitable calibration routine. Another challenge comes from the requirement of long-term con-

tinuous measurements. Fundamentally, the noble-gas comagnetometer is run in a continuous measurement mode, however, in practice, the measurements are interrupted by the necessity to reset the compensation point. This stems from the fact that the compensation point may drift over time of the measurement. One source of such a drift is the changes in noble-gas polarisation. For the reliable performance of the network, the GNOME measurements require minimisation of time of such zeroing. In addition to those two issues, the overall comagnetometer operation should be stable over multiple months (preferably years) with minimal time required for the maintenance of the system. Those challenges are addressed by the main content of this work.

Currently, two Advanced GNOME stations are being developed (Kraków and Mainz). In both stations, ^3He -K-Rb mixture is used, where two alkali-metal vapours have significantly different concentrations. This solution is used to optimised using the so-called **hybrid pumping** [63]. The comagnetometers are operated in a two-axis mode, where they are sensitive to perturbation in a plane perpendicular to the pump-light propagation direction. This is in contrast to the first-generation GNOME stations, where each station is sensitive along a single axis. The main part of this dissertation is dedicated to describing the Advanced GNOME development and demonstration of its novel capabilities. The operating stations of advanced GNOME are already used for the collection of the data, which will allow for improving the previous results of the GNOME searches. In the near future, the building of a few more Advanced GNOME stations is about to start in Moxa, Oberlin, and Hayward.

1.4.3. THE CURRENT SCIENTIFIC TARGETS

As discussed in Sec. 1.2, optical magnetometers and comagnetometers are potentially sensitive to the gradients of the axion field, and hence GNOME is capable of detecting such gradients on a global scale. Interestingly, different axion-field astrophysical structures that can be searched for by GNOME would have different signal patterns in the GNOME data. The parameters of the axion field spacial gradient and its velocity relative to the Earth depend on the specific model of axion DM. Here, a few examples of the possible GNOME targets, which manifest themselves with different spatio-temporal patterns are discussed. The presented examples are just a part of a considered set of possible targets, and for a complete discussion of currently considered GNOME targets, the reader is referenced to Ref. [41].

DOMAIN WALLS

The first hypothesis of axion dark-matter objects tested by GNOME concerned **domain walls of the axion-like field**. In this model, the gradient of the axion field arises in a domain wall that separates two neighbouring axion-field regions (domains). Since inside the domain, the ALP field is homogeneous, it does not exert any torque on spins, and hence, only transition through the wall can be detected with (co)magnetometers. As in this model the size of the domain is large with respect to Earth, a “collision” with the domain wall observed from Earth’s perspective is a plane perturbation propagating with a constant velocity in a certain direction. Crossing through such a wall would appear in the GNOME data as a series of transient perturbations, fulfilling a special spatio-temporal pattern across the network. The timing of the domain wall “hitting” a given sensor de-

depends on the direction and velocity of its propagation. Moreover, as the axion-field gradient is perpendicular to the plane of the wall, the spatial sensitivity of the sensors would imprint an additional pattern in the signal. It can be assumed that the signal from the single sensor generated by the domain wall crossing in the case of the linear coupling to the fermions can be approximated with a Lorentzian profile [39]. The thickness of the wall depends on the axion mass. The propagation direction, velocity, and thickness of the wall are generally unknown and have to be tested experimentally along with the coupling strength of the axion field.

If the domain wall propagates along the $\mathbf{x}$ axis, the solution for the axion field has the following form [41]

$$a(x) = a_0 \arcsin[\tanh(x/\lambda_c)], \quad (1.5)$$

where a_0 is a constant proportional to the spontaneous symmetry-breaking scale associated with the axion field, and $\lambda_c = \hbar/(m_a c)$ is the Compton wavelength of the axion, which is determined by its mass m_a . The transition through the axion domain wall will generate the following pseudomagnetic field

$$\mathbf{b}_{\text{DW}} \sim \frac{a_0}{\lambda_c} \text{sech}(x/\lambda_c) \mathbf{x}. \quad (1.6)$$

Data collected in Science Run 2 (see Fig. 1.5) have been analysed with a routine developed for the domain-wall search [61]. This allowed to set a new constraint for the parameters of ALPs domain walls [46].

AXION STARS

It was shown in Ref. [64–67], that axion DM may exist in the form of compact objects called **axion stars**. They would arise due to a gravitational or non-gravitational self-interaction between axions. These interactions could lead to the formation of stable (spherical) objects within which the axion field a oscillates and is described with the following equation [41]

$$a(\mathbf{r}, t) = \Psi(r) \cos(\mathbf{k} \cdot \mathbf{r} - \omega t + \theta), \quad (1.7)$$

where $\mathbf{r}$ is the distance to the centre of the axion star, $\mathbf{k}$ is the wave vector of the axion star and θ is a random phase, and $\Psi(r)$ is the spherically symmetric wave function of the axion field¹³ [41]

$$\Phi(r) = \sqrt{\frac{N\kappa^3}{7\pi R^3}} \left(1 + \frac{\kappa r}{R}\right) e^{-\kappa r/R}, \quad (1.8)$$

where R is the characteristic radius of the axion star determining a sphere which contains 99% of the axion star mass, κ is a numerical parameter depending on the model of the axion field. The interaction with the oscillating axion field inside the axion star manifests itself in the GNOME in the form of the oscillating transient spin coupling

$$\mathbf{b}_f \sim \frac{\partial \Phi}{\partial r} \cos(\mathbf{k} \cdot \mathbf{r} - \omega t) \frac{\mathbf{r}}{r} - \frac{m_a \theta}{\hbar} \Phi(r) \sin(\mathbf{k} \cdot \mathbf{r} - \omega t) \frac{\mathbf{k}}{k}, \quad (1.9)$$

¹³There are different forms of the wave functions, which are considered to be a viable description of the axion field. Here, the most probable form of the wave function is used (see Ref [41] and references therein).

where ϑ is the relative velocity of the axion star with respect to Earth. The frequency of the oscillations would be determined by the axion mass or, more precisely, the Compton frequency of the axions. The duration of the oscillating transient depends on the size of the axion star.

Some of the current GNOME data analyses search for transient oscillating signals that may be associated with the Earth collision with much larger objects

$$10R_{\oplus} \lesssim R \lesssim 10^6 R_{\oplus}, \quad (1.10)$$

where $R_{\oplus}$ is the Earth radius. The Earth-DM star collision would produce a given imprint in a single-sensor signal and a specific spatiotemporal pattern across the whole network. The specific propagation time of the signal across the network would be given by

$$T \approx \frac{2R_E}{\vartheta_c}, \quad (1.11)$$

where ϑ_c is the relative velocity of the axion star with respect to the Earth. These parameters set some limitations on the properties of the signals associated with the axion-star collisions.

If a substantial fraction of dark matter exists in the form of axion stars of relatively small masses, their presence would neither produce detectable small-scale gravitational microlensing nor create observable effects in the Solar system dynamics [68]. However, such structures could occasionally go through Earth, potentially generating effects detectable with terrestrial experiments.

Assuming at least one encounter of Earth with an axion star a year, the mass of such objects would be upperbounded by [69]

$$M \lesssim 10^{-16} M_{\odot}, \quad (1.12)$$

where $M_{\odot}$ is the mass of the Sun. Despite being extremely small on the astrophysical scale, collisions with objects of such small masses would produce effects detectable with GNOME.

AXION BURSTS AND MULTI-MESSENGER ASTRONOMY

It is speculated in Ref. [44] that axions can be generated in high-energy astronomical events, such as supernovae and black-hole or neutron-star merges. In these cases, a coherent pulse of ultra-relativistic axions, manifested as an exotic-field pulse of a certain central frequency and bandwidth, could be generated. Propagating through space with nearly a vacuum speed of light c , the pulse would experience dispersion, manifesting as lengthening and chirping of the pulse. Thereby, the axion-field pulse detected on the Earth would have a specific spectral character and would propagate through a network from the direction of the event. The propagation time of the pulse through Earth would be several orders of magnitude faster than in previously considered cases

$$T_{BR} \approx \frac{2R_{\oplus}}{c}, \quad (1.13)$$

and its length and chirp would depend on the axion mass and the Earth-event distance.

In principle, signals associated with the coherent emission of axions during cataclysmic astrophysical events could be detected with GNOME. However, the detection requires the axion-field pulse central frequency to lie within the bandwidth of the magnetometers used in the network. The search for the signal potentially induced by axion-field bursts adds a new channel for multi-messenger astronomy. This provides a strong motivation to search for signals correlated with observations in classical multi-messenger astronomical channels such as electromagnetic radiation, gravitational waves, or neutrinos. The stream of massive particles will always reach the Earth later than the gravitational waves, while the time of arrival time photons and neutrinos with respect to axions may depend on conditions of the production and propagation of the particles. Therefore, the detection of the signal in conventional astronomy can be used as a trigger for searches of the axion signal within the GNOME data. Moreover, the information about the position of the source in the sky allows the narrowing of the search angle and simplifies the analysis of the GNOME data.

STOCHASTIC FLUCTUATIONS AND OSCILLATIONS OF THE AXION FIELD

The astronomical structures with naturally inhomogeneous distribution of the axion DM, which were discussed above, are the most obvious targets for GNOME searches. However, they are just a fraction of all possible theoretical models of axion cosmology. In contrast to topological or coherent cases, a wide range of axion models leads to the homogeneous distribution of axions. For instance, the standard halo model predicts the existence of virialized axions [70–72]. In such a case, there is no gradient of the axion field on the scale of Earth, which could be detected as transient perturbation propagating through the network. Nevertheless, even in this case, GNOME can be used for testing the properties of the axion field.

The virialized axion field can be considered a superposition of independent axion waves with velocities $f_{lab}(\boldsymbol{\vartheta})$ given by the Maxwell-Boltzmann distribution

$$f_{lab}(\boldsymbol{\vartheta}) \approx \frac{1}{\pi^{3/2} \vartheta_0^3} \exp\left(-\frac{(\boldsymbol{\vartheta} - \boldsymbol{\vartheta}_{lab})^2}{\vartheta_0^2}\right), \quad (1.14)$$

where $|\boldsymbol{\vartheta}_{lab}| \approx 233$ km/s is the lab frame velocity dominated by the motion of Sun in the galactic frame and $\vartheta_0 \approx 220$ km/s is the characteristic viral velocity. The equation above suggests that for a terrestrial observer the axion field has a form of the incoherent stream coming from the direction of the Solar system motion in the Galactic frame. This phenomenon is called the **axion wind** [42].

The relative motion of the oscillating axion field produces the time-dependent gradient of the field. This will produce an oscillating perturbation, which can be detected with optical magnetometers and comagnetometers. Considering the stochastic nature of the axion field, the signal corresponding to the interaction with the axions will have a form of additional noise in the system with parameters (central frequency, bandwidth, etc.) determined by the parameters of the axion field. The change of the angle between the axion speed observed in the laboratory frame and the sensitivity axis of the sensor appearing due to the rotation of the Earth also leads to the daily modulation of the signal when a sensor has directional sensitivity.

GNOME allows the search for the linear coupling of fermions to the virialized axion field within the bandwidth of the sensors, which limits the detection range to masses of

the axions smaller than 10^{-13} eV. Single-sensor searches of such features have been performed with other types of spin-based sensors, however, the network of sensors brings additional advantages. First, it allows gaining the sensitivity by a factor $1/\sqrt{N_S}$, where N_S is the number of sensors in the network. Moreover, the cross-correlation of the output of the sensor gives strong proof for the global nature of the observed phenomena.

Studying the quadratic coupling of the axion field to fermions allows extending the range of the tested axion masses through a measure of the axion field intensity. The finite width of the spectrum of the axion field leads to spectral components of the intensity spectrum. One is the high-frequency component, which is not visible to the sensor with limited bandwidth, and another is a low-frequency component, which describes the changes in the oscillation envelope. The low-frequency changes in the axion field intensity can be detected by GNOME, which effectively extends the mass range of detectable virtualized axion field. The data analysis for this signal pattern would rely on the cross-correlation of the signal from two different stations.

1.5. SUMMARY OF THE CHAPTER

This chapter aimed to present the broad context of the thesis. It includes the topic of dark matter (Sec. 1.1), with particular attention paid to the axion dark matter (Sec. 1.2). In the context of this work, the axion dark matter is of the main interest due to the possibility of its detection or exclusion with techniques of optical magnetometry, as there is a possibility of coupling the axions to the spins of the atoms used for sensing in optical magnetometers. Hence, this chapter also presents the foundations of the search for exotic spin couplings with optical magnetometers (Sec. 1.3). The discussion briefly covers the first principles of optical magnetometry, its sensitivity to exotic spin couplings, and the key challenges related to axion dark matter searches with optical magnetometers. The first fundamental challenge is the potential time dependence and uncertainty of the time characteristics of the signals expected from the interaction with the axion dark matter. The solution for this problem is provided by the GNOME experiment, which is also discussed in Sec. 1.4. The second challenge is related to the high sensitivity of the optical magnetometers to the magnetic fields since the magnetic field noise limits the sensitivity to the non-magnetic spin couplings. This chapter explains how this problem could be addressed with self-compensating comagnetometers, which are the main topic of this thesis. The next chapter provides a detailed theoretical description of the SERF optical magnetometers and the self-compensating comagnetometers, which will provide a more rigorous discussion of their sensitivity and working principles.

2

THEORETICAL BACKGROUND

In this chapter, the theoretical framework necessary for understanding the operation of spin-exchange-relaxation-free (SERF) magnetometers and noble-gas-alkali-metal co-magnetometers is presented. Semi-classical models based on the Bloch equations are employed for the description of both systems.

2.1. SERF MAGNETOMETRY

The **SERF magnetometers** are the magnetic-field sensors using atomic vapours of alkali metals. The measurement of the magnetic and non-magnetic spin perturbations relies on the monitoring of the ground-state spin polarisation. In these devices, the spins are optically-oriented with circularly polarised (pumping) light. In the presence of the magnetic field, the polarised spins precess at a rate proportional to the amplitude of the field. Such precession is then detected by probing light, usually by monitoring the polarisation rotation of the linearly polarised light after the medium. The rotation is proportional to the electron spin-polarisation projection on the propagation axis of the probe light. In a standard arrangement of the SERF magnetometer, the probe and pump beams are perpendicular to each other, hence the system is primarily sensitive to the magnetic fields applied along the **y** axis that is transverse to the propagation directions of both beams.

The fundamental sensitivity of the optical magnetometers to the magnetic field, for the measurement time τ much longer than the spin coherence time $T_2 = R_{tot}^{-1}$, where R_{tot} is the total spin-relaxation rate, is determined by the atomic shot noise and given by [45]

$$\delta B = \frac{1}{\gamma} \sqrt{\frac{R_{tot}}{N_a \tau}}, \quad (2.1)$$

where N_a is the number of atoms taking part in the measurement, and γ is the gyromagnetic ratio of the atoms.

The distinct feature of the SERF magnetometer is its high sensitivity. It stems from a small relaxation rate, which, at high concentrations, is suppressed by the efficient **spin-**

exchange collisions. Operation under such suppressed spin relaxation is known as a **spin-exchange relaxation-free regime**.

This section, providing a theoretical description of the SERF magnetometry, starts with basic thermodynamic considerations of an field-sensing atomic vapour contained a glass cell (Sec. 2.1.1). Next, the energy-level structure of the alkali atoms is described. The section is followed by a discussion of key collisional processes which strongly affect the spin polarisation of the system (Sec. 2.1.2). The spin dynamics in the presence of the external magnetic field and collisional evolution, along with the origin of the SERF regime, is discussed in Secs. 2.1.3 and 2.1.4, respectively. The details of the light-atom interactions, including optical absorption, resultant optical pumping and polarisation rotation, AC Stark effect, are discussed in Sec. 2.1.5. In Sec. 2.1.6, various methods of magnetic-field measurements using a SERF system are described. The last section (Sec. 2.1.7) provides a description of the parametric modulation of the alkali-metal polarisation, which can be used for the optimisation of the alkali-metal polarisation detection.

2.1.1. ATOMS IN VAPOUR CELL

The heart of all optical magnetometers is an evacuated glass cell containing a droplet of liquid alkali metal. While optical magnetometers operate based on various elements, in this work, potassium and rubidium are of prime importance and will be the main focus. The concentration of the vapours of the elements is determined by their saturation concentration in the presence of the liquid metal droplet at a given temperature, which can be described with the following empirical expressions [73]

$$\begin{aligned} N_K(T_a) &= \frac{10^{26.2681-4453/T_a}}{T_a} \text{ atoms} \cdot \text{cm}^{-3}, \\ N_{\text{Rb}}(T_a) &= \frac{10^{26.178-4040/T_a}}{T_a} \text{ atoms} \cdot \text{cm}^{-3}, \end{aligned} \quad (2.2)$$

where T_a is the vapour temperature in kelvin.

The SERF magnetometers typically operate in the temperature range between 160 and 200°C, which results in a concentration on the order of $\sim 10^{13} - 10^{15}$ atoms/cm³ (Fig. 2.1).

2.1.2. ENERGY LEVEL STRUCTURE OF ALKALI-METAL ATOMS

The alkali-metal atoms are often used in atomic-physics experiments. Utilisation of the elements is mainly driven by a simple, hydrogen-like energy-level structure of alkalies (Fig. 2.2). In their case, a valence electron has a relatively weak bound to the atomic core, consisting of the nucleus surrounded by closed electronic shells. In this case, the energy-level structure is determined by the interaction of valence-electron spin $\hat{\mathbf{S}}$, orbital angular momentum $\hat{\mathbf{L}}$, and nuclear spin of the atom $\hat{\mathbf{I}}$. Coupling between $\hat{\mathbf{S}}$ and $\hat{\mathbf{L}}$ results in a fine structure of the atom described with the total angular momentum of the electron

$$\hat{\mathbf{J}} = \hat{\mathbf{S}} + \hat{\mathbf{L}}, \quad (2.3)$$

where the hat symbol $\hat{}$ indicates operator.

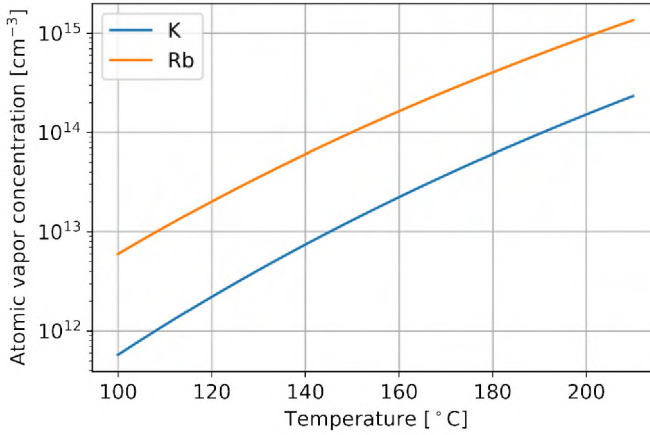


Figure 2.1: Comparison of the K and Rb atomic vapour concentrations versus temperature. Based on Eqs. (2.2).

For all alkalis, optical transitions from the electronic ground state $^2S_{1/2}$ ($L = 0$, $J = 1/2$) to the first two excited states, $^2P_{1/2}$ ($L' = 1$, $J' = 1/2$) and $^2P_{3/2}$ ($L' = 1$, $J' = 3/2$) lay in a near-infrared spectral range. This makes them accessible with inexpensive, commercially available diode lasers, providing convenient optical means of manipulating and probing the state of the atoms.

The hyperfine structure of the atoms is a result of the interaction between the total angular momentum of electron $\mathbf{\hat{J}}$ and the nuclear spin $\mathbf{\hat{I}}$, and it is described with the total angular momentum of atom $\mathbf{\hat{F}}$ given by

$$\mathbf{\hat{F}} = \mathbf{\hat{J}} + \mathbf{\hat{I}}. \quad (2.4)$$

Since in the ground state of alkali atoms $L = 0$, the hyperfine interaction leads to two hyperfine levels with $F = I \pm 1/2$, and the transition frequencies between ground-state hyperfine levels lie in the microwave range. Different alkalis and their isotopes have nuclear spins with different spin quantum numbers (for ^{85}Rb $I = 5/2$, and for ^{87}Rb and ^{39}K $I = 3/2$), which results in a different number of Zeeman sublevels in each hyperfine level.

Each hyperfine level consists of the $2F + 1$ sublevels, which are degenerate in the absence of external magnetic (or pseudomagnetic) fields. The Zeeman sublevels are labelled by the magnetic quantum number m , and hence the quantum state can be described by $|F, m\rangle$.¹ The application of the magnetic (or pseudomagnetic) fields splits the Zeeman sublevels. The energy-level splitting is determined by the magnetic-field strength and direction and gyromagnetic ratio of atoms.

As shown by Eq. (2.1), the long-lived ground states are of particular interest for magnetometry. The Hamiltonian governing the evolution of the alkali-atom ground state has

¹Note that for unambiguous determination of the level, one should also include S , L , and J , so that the state is $|F, J, L, S, m\rangle$. Here, for the sake of the notation compactness, J and L are omitted, as well as S ($S = 1/2$ in all cases).

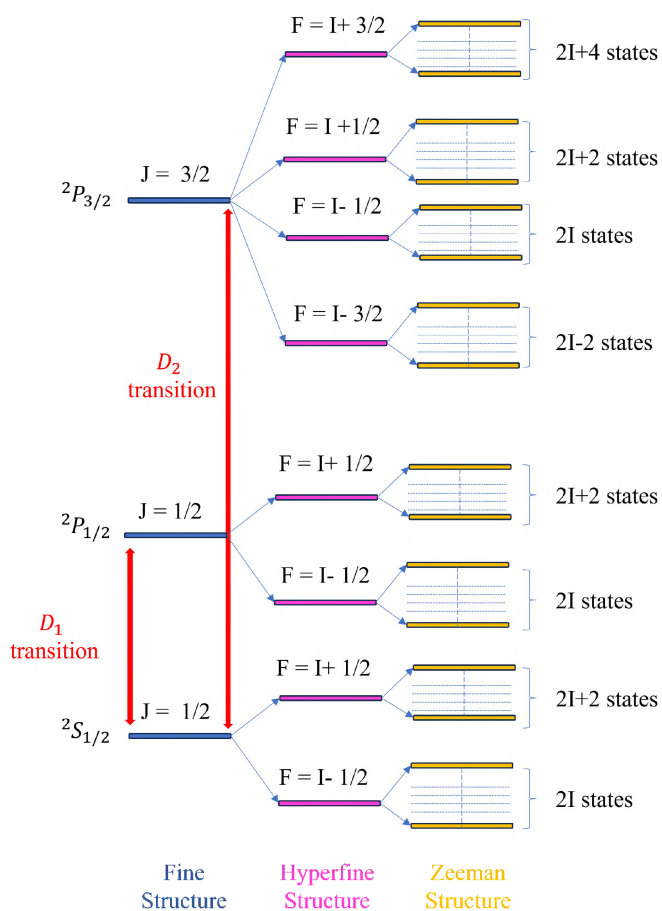


Figure 2.2: Generic energy-level structure of the alkali-metal atoms for the optical transitions from the ground to the first two excited states (D_1 and D_2 transitions). The energy-level splittings are not in scale.

the following form

$$\hat{H}_g = A\hat{\mathbf{I}} \cdot \hat{\mathbf{S}} - g_s \mu_B \hat{\mathbf{S}} \cdot \mathbf{B} + \frac{\mu_I}{\hbar} \hat{\mathbf{I}} \cdot \mathbf{B} + \frac{1}{S} \chi_e \Xi \cdot \hat{\mathbf{S}} + \frac{1}{I} \chi_I \Xi \cdot \hat{\mathbf{I}}, \quad (2.5)$$

where A is the hyperfine coupling constant, g_s is a Landé factor of the atoms, μ_B is the Bohr magneton, $\mathbf{B}$ is applied magnetic field, μ_I is the magnetic moment of the nuclear spin, χ_e and χ_I are the electronic and nuclear couplings constants, which characterise couplings strength of spins to a generic exotic spin coupling Ξ .²

The ratio between nuclear and electron magnetic moments exceeds 100. Therefore, in optical magnetometry, the Zeeman interaction of the magnetic field with the nuclear spin is often neglected. However, in the case of exotic interactions, there is no information about the strength of the coupling, and therefore, the interaction with the nuclear spin cannot be similarly neglected.

In a similar way as in Eq. (1.3), one may define the pseudomagnetic fields b_e and b_I corresponding to the exotic couplings to electron and nuclear spins

$$\mathbf{b}_e = -\frac{\chi_e}{g_s \mu_B S} \Xi, \quad (2.6)$$

$$\mathbf{b}_I = \frac{\chi_I}{\mu_I} \Xi, \quad (2.7)$$

which leads to the following form of the interaction Hamiltonian

$$\hat{H}_g = A\hat{\mathbf{I}} \cdot \hat{\mathbf{S}} - g_s \mu_B \hat{\mathbf{S}} \cdot (\mathbf{B} + \mathbf{b}_e) + \frac{\mu_I}{\hbar} \hat{\mathbf{I}} \cdot \mathbf{b}_I, \quad (2.8)$$

where the nuclear magnetic interaction was neglected.

The microscopic evolution of the atomic ensembles in optical magnetometry is usually described with the density-matrix formalism, where the density matrix $\hat{\rho}$ is defined in the following way

$$\hat{\rho} = \sum_{i,j} \rho_{ij} |i\rangle \langle j|, \quad (2.9)$$

with i and j running over all basis vectors and $\rho_{ij} = \langle i | \hat{\rho} | j \rangle$ are non-negative numerical coefficients. Within the formalism, the time evolution of the quantum state of the system is described by the Liouville–von Neumann equation

$$\frac{d\hat{\rho}_0}{dt} = \frac{1}{i\hbar} [\hat{H}_g, \hat{\rho}_0], \quad (2.10)$$

where $\hat{\rho}_0$ is the density matrix of the alkali-metal atom exposed to interactions given by the Hamiltonian $\hat{H}_0$.

2.1.3. COLLISIONAL EVOLUTION OF ALKALIES GROUND STATE

Atoms in the vapour are moving and collide with one another as well as walls of their container (cell). These two processes have a significant impact on the evolution of the ground-state spin polarisation of the atoms. This section considers the physical grounds of the processes.

²Although this work is mostly related to searches of spin couplings to axion field described in Eq. (1.1), here a generic spin coupling, not connected to any particular theoretical framework, is introduced.

COLLISIONS WITH THE WALLS

Since collisions with the walls randomise the orientation of spin polarisation, they are considered as the depolarisation process. In fact, if no measures are undertaken to reduce the frequency of such collisions or reduce the cross-section for the depolarisation, these collisions dominate relaxation of the atomic polarisation.

There are two methods that enable the suppression of wall relaxation. The first is coating of the inner walls of the cell with special (antirelaxation) layer. The layer decreases the probability of the atomic-state change during the collision. This method is widely used in room-temperature magnetometers, where paraffin coating of the walls is used to prolong spin relaxation. However, the SERF magnetometers operate at temperatures much above the melting temperature of the paraffin (the reasoning for the temperature conditions will be clarified in the subsequent section), which does not allow the application of the approach in the magnetometers. The high-temperature coatings for the SERF magnetometry have been used in Ref. [74], however, the coating material (octadecyl trichlorosilane) is stable in operation temperatures below 160°C and for higher temperatures, it starts, relatively slow, degradation process, which limited the application of this solution.

Adding a buffer gas (usually ^4He , Ne , or N_2) into the cell decreases diffusion of the atoms toward the cell walls and hence is another process to reduce wall relaxation. The motion of the atoms in the atmosphere of a dense inert gas is characterised by a much shorter mean free path, and therefore, the travel between the walls takes longer, prolonging the polarisation lifetime. The evolution of the density matrix in the presence of a buffer gas is determined by the following equation [75]

$$\frac{d\hat{\rho}_D}{dt} = D\nabla^2\hat{\rho}_D, \quad (2.11)$$

where D is the diffusion coefficient

$$D = D_0 \left(\frac{T_a}{T_{a0}} \right)^{3/2} \frac{p_0}{p}, \quad (2.12)$$

which depends on both the temperature T_a and pressure of the buffer gas p , with D_0 being a diffusion constant at the temperature T_{a0} and pressure p_0 . To slow down the diffusion and hence limit the relaxation due to wall collisions, it is favourable to operate at a higher concentration of the buffer gas.

SPIN-EXCHANGE COLLISIONS BETWEEN ALKALI ATOMS

After the suppression of the relaxation due to the wall collisions with either antirelaxation coating or buffer gas, alkali-atom spin relaxation is dominated by spin-exchange collisions. These collisions preserve the total angular momentum of two colliding atoms but lead to the redistribution of the angular momentum between two hyperfine levels. Due to the opposite precession of spin polarisation in the levels, at a nonzero magnetic field, the collisions lead to the deterioration of the transverse polarisation of atoms, hence being the source of relaxation. The relaxation rate due to the spin-exchange collisions is given by the following expression [76]

$$R_{SE} = \frac{1}{T_{se}} = N_a \langle \sigma_{se} \tilde{\theta} \rangle, \quad (2.13)$$

where N_a is the concentration of alkali metal, σ_{se} is the cross-section for the spin exchange, and $\bar{v}$ is the mean relative velocity of the colliding atoms

$$\bar{v} = \sqrt{\frac{8k_B T_a}{\pi\mu}}, \quad (2.14)$$

where μ stands for the reduced mass of two colliding atoms.³

The spin-exchange process during a collision arises from the interaction between electron spins of colliding atoms

$$\hat{V}_{ex} = J(R)\hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j, \quad (2.15)$$

where $J(R)$ is the coupling coefficient, which depends on the distance R between the colliding atoms [76]. In the ensemble of a large number of atoms, all possible trajectories of the collisions, hence all possible values of $J(R)$, are realised, which leads to effective averaging over such a process. Since the room- or higher-temperature alkali-metal atoms undergo mostly sudden (compared to the characteristic time of the hyperfine interaction) two-atom collisions, in atomic evolution, the effect of such collisions can be considered as an instant change of the electron quantum (spin) state. Simultaneously, the spin-exchange process has no direct effect on the nuclear spins of the atoms. For this reason, it is convenient to separate the alkali-metal density matrix $\hat{\rho}_{se}$, associated with the collisions, into purely nuclear part $\hat{\phi}$, which is unaffected by the spin-exchange, and electron-polarised part $\hat{\Theta} \cdot \hat{\mathbf{S}}$ [76]

$$\frac{d\hat{\rho}_{se}}{dt} = \hat{\phi} + \hat{\Theta} \cdot \hat{\mathbf{S}}, \quad (2.16)$$

$$\hat{\phi} = \frac{1}{4}\hat{\rho}_{se} - \hat{\mathbf{S}} \cdot \hat{\rho}_{se}\hat{\mathbf{S}}, \quad (2.17)$$

$$\hat{\Theta} \cdot \hat{\mathbf{S}} = \frac{3}{4}\hat{\rho}_{se} - \hat{\mathbf{S}} \cdot \hat{\rho}_{se}\hat{\mathbf{S}}. \quad (2.18)$$

Then, the evolution of the atomic density matrix for such a system has the following form [76]

$$\frac{d\hat{\rho}_{se}}{dt} = \frac{1}{T_{se}}[\hat{\phi}(1 + 4\langle\hat{\mathbf{S}}\rangle \cdot \hat{\mathbf{S}}) - \hat{\rho}_{se}] + \frac{2\kappa}{iT_{ex}}[\langle\hat{\mathbf{S}}\rangle \cdot \hat{\mathbf{S}}, \hat{\rho}_{ex}], \quad (2.19)$$

where the first term accounts for the incoherent part of spin-exchange interaction [Eq. (2.15)], which leads to the relaxation of atomic states. The second term takes into account the coherent part of spin-exchange interaction [Eq. (2.15)], which leads to the energy shift. The unitless parameter κ characterises the interaction shifting the energy and is usually at the level of a few percent [76, 80]. This small self-interaction term [the second term on the right side of Eq. (2.19)] is usually neglected in consideration of the alkali-atom dynamics in optical magnetometry because it does not modify the measurement process in any significant way.

A more detailed discussion of the spin-exchange collisions can be found in Refs. [75, 76, 80]. Moreover, Ref. [81] provides a review of the experimental values of the spin exchange rates, which can be found in the literature. Table 2.1 provides the known spin-exchange relaxation cross-sections for rubidium and potassium atoms used in this work.

³Due to the same mass of the colliding atoms, here the reduced mass equals half of the alkali-atom mass.

Species	$\sigma_{se} [10^{-14} \text{ cm}^2]$
$^{39}\text{K}-^{39}\text{K}$	1.5 ± 0.2 [77]
$^{85}\text{Rb}-^{85}\text{Rb}$	1.9 ± 0.3 [77]
$^{85}\text{Rb}-^{87}\text{Rb}$	1.7 ± 0.3 [78]
$^{87}\text{Rb}-^{87}\text{Rb}$	1.9 ± 0.2 [77, 79]

Table 2.1: Experimentally-determined spin-exchange relaxation cross-sections of alkali-metal atoms of the elements used in this thesis.

SPIN-DESTRUCTION COLLISIONS

The spin-exchange interaction is not the only process appearing during the collision of the alkalis [75]. There is a range of processes which do not preserve the total angular momentum and hence lead to complete depolarisation of the colliding atoms. These spin-dissipative processes are commonly referenced as the **spin-destruction collisions**. They arise both in alkali interatomic collisions and in the collisions between alkali-metal atoms and buffer-gas atoms. The relaxation rate corresponding to the spin-destruction collisions is determined by the concentration of depolarising collision partners N_j , the mean relative velocity of the colliding partners $\bar{v}_j$, and the cross-section for the spin-destruction collisions σ_j^{SD}

$$R_{SD} = \sum_j N_j \langle \sigma_j^{SD} \bar{v}_j \rangle, \quad (2.20)$$

where j denotes various species of colliding partners. Since the sudden collisions change only the electron spin state, the collisional evolution of the density matrix is determined by the following equation [76]

$$\frac{d\hat{\rho}_{SD}}{dt} = \frac{1}{T_{SD}} (\hat{\phi} - \hat{\rho}_{SD}). \quad (2.21)$$

The spin-destruction processes in interatomic collisions between alkali-metal atoms are not fully understood yet. Currently, it is known that the spin-destruction process between atoms appears due to spin-axis interactions, spin-rotation interactions, and the formation of alkali-metal dimers [75, 80, 82, 83]. The impact of the various channels of spin-destruction relaxation strongly depends on the conditions in the cell [82]. In the context of this work, the focus is on the case of the cells with rather high concentrations of the buffer gas ($\sim 10^{19} \text{ atoms}\cdot\text{cm}^{-3}$) and high concentrations of the alkali metal atoms ($\sim 10^{14} \text{ atoms}\cdot\text{cm}^{-3}$), which operate in a near-zero magnetic field environment. Then, relaxation is dominated by spin-axis collisions between alkali metals and spin-rotation collisions with noble-gas atoms. The experimental values of the cross-section for spin-destruction collisions are presented in Table 2.2.

2.1.4. SPIN-EXCHANGE-RELAXATION-FREE REGIME

In many cases, spin-exchange collisions are the limiting factor for the sensitivity of the optical magnetometers operating at room and alleviated temperatures. This stems from

	K	Rb
σ_{self}^{SD}	$1 \times 10^{-18} \text{ cm}^2$ [84]	$9 \times 10^{-18} \text{ cm}^2$ [85]
σ_{He}^{SD}	$8 \times 10^{-25} \text{ cm}^2$ [85, 86]	$9 \times 10^{-24} \text{ cm}^2$ [85]
σ_{Ne}^{SD}	$1 \times 10^{-23} \text{ cm}^2$ [86]	$1.9 \times 10^{-23} \text{ cm}^2$ [87]
$\sigma_{N_2}^{SD}$	$1.1 \times 10^{-23} \text{ cm}^2$ [87]	$6 \times 10^{-22} \text{ cm}^2$ [88]

Table 2.2: Experimental values of spin-destruction cross sections for alkali metals and noble gasses.

the broadening of the magnetic resonances used for the measurement of the magnetic fields with the growing temperature. Specifically, as follows from Eqs. (2.2), (2.13), and (2.14), the spin-exchange relaxation rate grows with the temperature and one may expect a continuous broadening of the magnetic resonances with growing temperature. However, this is not always true. For not-too-large magnetic fields, $\omega_a \ll 1/T_{se}$, the trend first saturates and then starts to decrease with the temperature. This ultimately leads to the magnetic-resonance line narrowing and stabilising at a value determined by other sources of relaxation, as the spin-exchange relaxation is eliminated (eventually, the resonance width starts to grow due to different temperature-dependent relaxation processes). This effect was first observed experimentally [89] and then explained theoretically by W. Happer and coworkers in the 1970's [90]. In the early 2000s, the narrowing of the resonance lines at higher temperatures ($\approx 180^\circ\text{C}$) was employed for the precise magnetometry and comagnetometry by M. Romalis and coworkers [91]. The regime in which the relaxation due to spin-exchange collision is eliminated is called the **spin-exchange-relaxation-free (SERF)** regime.

To understand the origins of the elimination of the relaxation due to spin-exchange collisions, one can consider the evolution of the atomic density matrix in the presence of a magnetic field and collisions. Taking into account Eqs. (2.10), (2.11), (2.19), and (2.21), the time evolution of the density matrix is given by

$$\begin{aligned} \frac{d\hat{\rho}}{dt} = & \frac{A}{i\hbar} [\hat{\mathbf{I}} \cdot \hat{\mathbf{S}}, \hat{\rho}] + D\nabla^2 \hat{\rho} - \frac{g_s \mu_B}{i\hbar} [\hat{\mathbf{S}} \cdot (\mathbf{B} + \mathbf{b}_e), \hat{\rho}] + \frac{\mu_I}{i\hbar} [\hat{\mathbf{I}} \cdot \mathbf{b}_I] \\ & + \frac{1}{T_{se}} [\hat{\phi}(1 + 4\langle \hat{\mathbf{S}} \rangle \cdot \hat{\mathbf{S}}) - \hat{\rho}] + \frac{2\kappa}{iT_{ex}} [\langle \hat{\mathbf{S}} \rangle \cdot \hat{\mathbf{S}}, \hat{\rho}] + \frac{1}{T_{SD}} (\hat{\phi} - \hat{\rho}). \end{aligned} \quad (2.22)$$

For the discussion, the $|F, m\rangle$ eigenbasis of the density matrix was chosen

$$\hat{\rho} = \sum_{F, F', m, m'} \rho_{Fm, F'm'} |F, m\rangle \langle F', m'|, \quad (2.23)$$

with the quantisation axis oriented along $\mathbf{z}$

$$\hat{F}_z |F, m\rangle = m |F, m\rangle. \quad (2.24)$$

The hyperfine term, i.e., the first term of Eq. (2.22) is considered at the start. As $|F, m\rangle$

are the eigenstates of the $\hat{\mathbf{I}} \cdot \hat{\mathbf{S}}$ operator,

$$\hat{\mathbf{I}} \cdot \hat{\mathbf{S}} |F, m\rangle = \frac{F(F+1) - I(I+1) - S(S+1)}{2} |F, m\rangle, \quad (2.25)$$

the term takes the following form

$$\frac{A}{i\hbar} [\hat{\mathbf{I}} \cdot \hat{\mathbf{S}}, \rho] = \frac{\Delta E_{hf}}{i\hbar} (F - F') \rho_{Fm, F'm'}, \quad (2.26)$$

where ΔE_{hf} stands for the energy difference between hyperfine levels with different F

$$\Delta E_{hf} = \frac{A}{2} (F + F' + 1). \quad (2.27)$$

Equation (2.26) shows that the hyperfine interaction drives fast oscillations between hyperfine states, and it does not affect the evolution of the atomic state within a given manifold, i.e. when $F = F'$.

This thesis considers optical magnetometers that do not employ microwave radiation, operating at low magnetic fields (the linear Zeeman-effect regime). In such a case, there are no coherences between hyperfine states. Hence, the density matrix describing ground states of the alkali-metal elements takes a block-diagonal form

$$\hat{\rho} = \begin{pmatrix} \hat{\rho}_a & 0 \\ 0 & \hat{\rho}_b \end{pmatrix}, \quad (2.28)$$

where the $\hat{\rho}_a$ and $\hat{\rho}_b$ correspond to the submatrices of the Zeeman-sublevel manifolds of $F = I \pm 1/2$, respectively. For convenience, the following definitions are used

$$\begin{aligned} \hat{\zeta}_a &= \begin{pmatrix} \hat{\rho}_a & 0 \\ 0 & 0 \end{pmatrix}, \\ \hat{\zeta}_b &= \begin{pmatrix} 0 & 0 \\ 0 & \hat{\rho}_b \end{pmatrix}. \end{aligned} \quad (2.29)$$

Presenting the density matrix in such a form allows one to independently consider its evolution in each hyperfine level.

In a typical SERF magnetometer, apart from the alkali-metal atoms, a vapor cell is additionally filled with a high-pressure buffer gas, which slows down atomic diffusion. This leads to small values of the diffusion coefficient D , which, in practice, allows one to neglect the diffusion

$$D \nabla^2 \hat{\rho} \xrightarrow{D \rightarrow 0} 0. \quad (2.30)$$

This simplifies Eq. (2.22) by neglecting its second term.

The high concentration of the alkali-metal atoms makes the spin-exchange collisions the fastest process in the system. To describe the evolution of the system, one can consider two time scales. One corresponds to slower processes and hence being described by “long” time τ_l and the other determines the “fast” processes and is characterised by

the time τ_s . There are following relations between the coefficients and other characteristic time constants in the experiment

$$T_{SD} \lesssim \frac{1}{|\omega_L|} \lesssim \tau_l, \quad (2.31)$$

$$T_{se} \ll \tau_s \ll \tau_l. \quad (2.32)$$

where $|\omega_L|$ is the absolute value of Larmor frequency of a given atomic spin F . Here, it is also assumed that within the short time τ_s the system is able to reach the steady state determined by the spin-exchange collisions [Eq. (2.19)]. As discussed in Sec. 2.1.3, the collisions redistribute angular momentum over hyperfine sublevels while preserving the total angular momentum of the colliding atoms. This leads to the following steady-state solution of Eq. (2.22) [92]

$$\hat{\rho}_{se} = \frac{e^{\boldsymbol{\beta} \cdot \hat{\mathbf{F}}}}{Z}, \quad (2.33)$$

where Z is the partition function

$$Z = \text{Tr}(e^{\boldsymbol{\beta} \cdot \hat{\mathbf{F}}}), \quad (2.34)$$

and $\boldsymbol{\beta}$ is the spin-temperature parameter,⁴ which is parallel to the spin expectation value $\langle \hat{\mathbf{F}} \rangle$. This density-matrix distribution is called the **spin-temperature distribution**. The orientation of $\boldsymbol{\beta}$ determines the direction of the total atomic spin expectation value $\langle \hat{\mathbf{F}} \rangle$. It is convenient to choose the coordinate frame such that $\boldsymbol{\beta}$ is collinear with the quantisation axis, i.e., $\boldsymbol{\beta} \times \mathbf{z} = \mathbf{0}$. In such a case, the state of the ensemble is determined by a single scalar parameter $\beta = |\boldsymbol{\beta}|$, called the **spin temperature**, and the density matrix in the $|F, m\rangle$ basis is diagonal

$$\hat{\rho}_{se} = \sum_{F,m} \frac{e^{\beta m}}{Z} |F, m\rangle \langle F, m|, \quad (2.35)$$

with $Z = \sum_m \exp(\beta m)$. At this point, one can note that β carries the information about the polarisation of the ensemble. Zero temperature ($\beta = 0$) corresponds to a fully unpolarised state (all sublevels are equally occupied). Fully polarised states, where the whole ensemble is in one of the stretched states $m = \pm F_{max}$, where F_{max} is the highest total angular momentum in the considered manifold, correspond to the infinite spin temperature ($\beta = \pm\infty$). The strict mathematical correspondence between spin temperature and atomic polarisation is discussed later in this section.

The dominance of the spin-exchange collision rate over other relaxation processes and Larmor precession at low magnetic fields results in the preservation of the spin-temperature distribution [90]. This effectively cancels out the effect of the spin-exchange collisions on the relaxation and hence broadening of the magnetic-field resonances and reduces the overall relaxation rate in the system.⁵

Now the focus is shifted to the determination of the total angular momentum $\langle \hat{\mathbf{F}} \rangle$. This expectation value can be written as a sum of the angular momentum from the

⁴For a more rigorous relation between $\boldsymbol{\beta}$ and $\langle \hat{\mathbf{F}} \rangle$ see Eq. (2.49)

⁵The dependence of the relaxation rate on the applied magnetic field is discussed in more detail later in this section.

atoms in two hyperfine states

$$\langle \hat{\mathbf{F}} \rangle = \langle \hat{\mathbf{F}}_{\mathbf{a}} \rangle + \langle \hat{\mathbf{F}}_{\mathbf{b}} \rangle, \quad (2.36)$$

where

$$\langle \hat{\mathbf{F}}_{a,b} \rangle = \text{Tr}(\hat{\mathbf{F}} \hat{\zeta}_{a,b}). \quad (2.37)$$

Then, by applying the angular momentum operator to both sides of Eq. (2.22) and tracing it over, one gets the time evolution of the expectation value of the angular momentum correspondent to each hyperfine state⁶

$$\frac{d\langle \hat{\mathbf{F}}_{a,b} \rangle}{dt} = \gamma_e (\mathbf{B} + \mathbf{b}_e) \times \langle \hat{\mathbf{S}}_{a,b} \rangle + \gamma_I \mathbf{b}_I \times \langle \hat{\mathbf{I}}_{a,b} \rangle - R_{SD} \langle \hat{\mathbf{S}}_{a,b} \rangle, \quad (2.38)$$

where $\gamma_e = \mu_B g_e / \hbar$ and $\gamma_I = \mu_I / (\hbar I)$ are the electron and nuclear gyromagnetic ratios, respectively. For further calculations, one can express all terms on the right side of the equation using $\langle \hat{\mathbf{F}}_{a,b} \rangle$. For $\langle \hat{\mathbf{S}}_{a,b} \rangle$, the special case of Wigner–Eckart theorem is used, which is also known as the projection theorem [93]

$$\langle \hat{\mathbf{S}}_{a,b} \rangle = \frac{\langle \hat{\mathbf{S}}_{a,b} \cdot \hat{\mathbf{F}}_{a,b} \rangle}{F(F+1)} \langle \hat{\mathbf{F}}_{a,b} \rangle = \pm \frac{1}{2I+1} \langle \hat{\mathbf{F}}_{a,b} \rangle. \quad (2.39)$$

Using this equation, one can also express $\langle \hat{\mathbf{I}}_{a,b} \rangle$

$$\langle \hat{\mathbf{I}}_{a,b} \rangle = \langle \hat{\mathbf{F}}_{a,b} \rangle - \langle \hat{\mathbf{S}}_{a,b} \rangle = \left(1 \mp \frac{1}{2I+1} \right) \langle \hat{\mathbf{F}}_{a,b} \rangle. \quad (2.40)$$

Incorporating Eqs. (2.39) and (2.40) into Eq. (2.38), the time evolution of $\langle \hat{\mathbf{F}}_{a,b} \rangle$ can be written as

$$\frac{d\langle \hat{\mathbf{F}}_{a,b} \rangle}{dt} = \pm \frac{\gamma_e}{2I+1} (\mathbf{B} + \mathbf{b}_e) \times \langle \hat{\mathbf{F}}_{a,b} \rangle + \gamma_I \left(1 \mp \frac{1}{2I+1} \right) \mathbf{b}_I \times \langle \hat{\mathbf{F}}_{a,b} \rangle \mp \frac{R_{SD}}{2I+1} \langle \hat{\mathbf{F}}_{a,b} \rangle. \quad (2.41)$$

The spin-temperature distribution given in Eq. (2.35) allows one to calculate the fraction of the total angular momentum stored in each of the hyperfine levels

$$\kappa_a = \frac{\langle \hat{\mathbf{F}}_{\mathbf{a}} \rangle}{\langle \hat{\mathbf{F}} \rangle} = \frac{\sum_{m_F=-F_a}^{F_a} m_F e^{\beta m_F}}{\sum_{m_F=-F_a}^{F_a} m_F e^{\beta m_F} + \sum_{m_F=-F_b}^{F_b} m_F e^{\beta m_F}}, \quad (2.42)$$

$$\kappa_b = \frac{\langle \hat{\mathbf{F}}_{\mathbf{b}} \rangle}{\langle \hat{\mathbf{F}} \rangle} = \frac{\sum_{m_F=-F_b}^{F_b} m_F e^{\beta m_F}}{\sum_{m_F=-F_a}^{F_a} m_F e^{\beta m_F} + \sum_{m_F=-F_b}^{F_b} m_F e^{\beta m_F}}. \quad (2.43)$$

⁶Note that here the evolution is considered under assumption of the system preserving the spin temperature distribution at all times, hence there is no cross-term correspondent to the spin-exchange collisions.

Here, the assumption of collinearity between the spin polarisation and the quantisation axis is also used. However, in the presence of the magnetic field, the expectation value of the spin is subjected to the Larmor precession and, therefore, changes its direction. Hence, for consistency with the spin-temperature distribution, it is assumed that the quantisation axis used to describe the ensemble follows the atomic polarisation movement.

To get the time evolution of the $\langle \hat{\mathbf{F}} \rangle$, one can combine equations of motion for $\langle \hat{\mathbf{F}}_a \rangle$ and $\langle \hat{\mathbf{F}}_b \rangle$, which gives

$$\begin{aligned} (\kappa_a + \kappa_b) \frac{d\langle \hat{\mathbf{F}} \rangle}{dt} = & (\kappa_a - \kappa_b) \frac{\gamma_e}{2I+1} (\mathbf{B} + \mathbf{b}_e) \times \langle \hat{\mathbf{F}} \rangle \\ & + \gamma_I \left((\kappa_a + \kappa_b) - \frac{(\kappa_a - \kappa_b)}{2I+1} \right) \mathbf{b}_I \times \langle \hat{\mathbf{F}} \rangle \\ & - \frac{(\kappa_a - \kappa_b) R_{SD}}{2I+1} \langle \hat{\mathbf{F}} \rangle. \end{aligned} \quad (2.44)$$

This brings the final equation of motion for $\langle \hat{\mathbf{F}} \rangle$

$$\frac{d\langle \hat{\mathbf{F}} \rangle}{dt} = \frac{1}{q(\beta, I)} \left[\gamma_e (\mathbf{B} + \mathbf{b}_e) \times \langle \hat{\mathbf{F}} \rangle - R_{SD} \langle \hat{\mathbf{F}} \rangle \right] + \gamma_I \left(1 - \frac{1}{q(\beta, I)} \right) \mathbf{b}_I \times \langle \hat{\mathbf{F}} \rangle, \quad (2.45)$$

where the slowing-down factor q takes into account both the effect of the hyperfine interaction and the averaging of the angular momentum over hyperfine states as a result of the spin-exchange collisions

$$q(\beta, I) = (2I+1) \frac{\kappa_a(\beta) + \kappa_b(\beta)}{\kappa_a(\beta) - \kappa_b(\beta)} = \frac{2I+1}{Q(\beta, I)}. \quad (2.46)$$

The slowing-down factor strongly depends on the nuclear spin I not only due to the hyperfine term but also due to different multiplicity of the hyperfine states, which affects $\kappa_{a,b}$ [see Eq. (2.42)].

As will be shown later in this work, in optical magnetometers, the probe light is used to directly measure the expectation value of the electron spin. Hence, it is more convenient to use expectation value of electron spin in the description. From the magnetic-field term, i.e., the first term in Eq. (2.45), assuming that in the considered model, the magnetic field interacts only with the electron spin (due to the difference between the electron and nuclear gyromagnetic ratios), one gets

$$\frac{d\langle \hat{\mathbf{F}} \rangle}{dt} \approx \gamma_e \mathbf{B} \times \langle \hat{\mathbf{S}} \rangle, \quad (2.47)$$

one can relate $\langle \hat{\mathbf{F}} \rangle$ with $\langle \hat{\mathbf{S}} \rangle$, which, in the SERF regime, is given by

$$\langle \hat{\mathbf{S}} \rangle = \frac{1}{q(\gamma, I)} \langle \hat{\mathbf{F}} \rangle. \quad (2.48)$$

For practical applications, it is convenient to express the expectation value of electron spin in terms of the electron polarisation Υ [76, 81]

$$\Upsilon \equiv 2\langle \hat{\mathbf{S}} \rangle = \frac{\beta}{\beta} \tanh \frac{\beta}{2}. \quad (2.49)$$

This leads to the following expressions for the slowing-down factors for two hyperfine states of rubidium isotopes [76]

$$Q(\Upsilon, I = 3/2) = 2 - \frac{4}{3 + \Upsilon^2}, \quad (2.50)$$

$$Q(\Upsilon, I = 5/2) = 3 - \frac{48(1 + \Upsilon^2)}{19 + 26\Upsilon^2 + 3\Upsilon^4}, \quad (2.51)$$

where $\Upsilon = |\Upsilon|$. One can also express the spin temperature in terms of the [76]

$$\beta = \ln \left(\frac{1 + \Upsilon}{1 - \Upsilon} \right). \quad (2.52)$$

This allows one to rewrite Eq. (2.45) using the electron polarisation Υ

$$\frac{d\Upsilon}{dt} = \frac{1}{q(\Upsilon, I)} \left[\gamma_e(\mathbf{B} + \mathbf{b}_e) \times \Upsilon - R_{SD}\Upsilon \right] + \gamma_I \left(1 - \frac{1}{q(\Upsilon, I)} \right) \mathbf{b}_I \times \Upsilon, \quad (2.53)$$

As a final result [Eq. (2.53)], the standard Bloch equation for the polarisation of the electron spin in the SERF magnetometer has been obtained. It needs to be stressed that the SERF regime is the result of the total angular momentum conservation in the spin-exchange collisions and the high spin-exchange rate in the system. At the same time, the spin-exchange collisions lead to the redistribution of the angular momentum in hyperfine levels, which ensures the preservation of the spin-temperature distribution. This is, however, an idealisation which is fulfilled only at near-zero magnetic fields. With a growing magnetic field, spin-exchange collisions start to contribute to the transverse relaxation at a rate determined by the amplitude of the applied magnetic field and the polarisation of the alkalis.

In the general case (for arbitrary polarisation and magnetic field), the dependence of the spin-exchange relaxation is highly nontrivial due to two different processes responsible for the suppression of the spin-exchange relaxation at high and low polarisation limits. At a low polarisation limit (approximating the density-matrix evolution up to the first order in $\hat{\rho}$), the spin-exchange relaxation rate grows with the increasing magnetic field (from zero at zero field). This originates from the fact that under such conditions the spin-temperature approximation starts to fail. In this case, the transverse relaxation rate can be obtained analytically [90]

$$\frac{1}{T_2} = \frac{(2I+1)^2 + 2}{3T_{se}(2I+1)^2} - \text{Re} \left\{ \sqrt{-\gamma_e^2 B^2 - \frac{2i\gamma_e B}{T_{se}(2I+1)} + \left[\frac{(2I+1)^2 + 2}{3T_{se}(2I+1)^2} \right]^2} \right\}. \quad (2.54)$$

This dependence is shown in Fig. 2.3. When the magnetic field is low such that the spin-exchange collision rate dominates over the Larmor precession, $\gamma_e B T_{se} \ll 1$ and, to first order, the corresponding relaxation rate grows proportionally to a square of the magnetic-field amplitude

$$\frac{1}{T_2} \xrightarrow{\gamma_e B \ll T_{se}^{-1}} \frac{T_{se}(2I+1)(1 - C^{-2})}{2C} (\gamma_e B)^2, \quad (2.55)$$

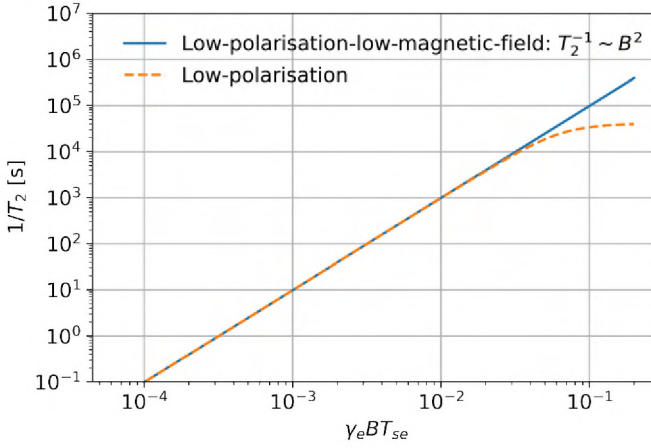


Figure 2.3: Transverse relaxation rate of alkali-metal atoms driven by the spin-exchange collisions estimated for the low-polarisation limit with Eq. (2.54) (the solid blue line) and its low-field approximation estimated with Eq. (2.55) (the dashed orange line). Both estimations are made assuming $I = 3/2$ and $T_{se} = 10^{-6}$ s.

where the constant C is given by

$$C \equiv \frac{(2I+1)^2 + 2}{3(2I+1)}. \quad (2.56)$$

At a high polarisation limit, $F^e \rightarrow 1$, the spin-exchange collisions do not affect the transverse coherence time since the whole population of atoms is at the same stretched state, and therefore the spin exchange is not possible. In this case, the magnetic field does not affect the spin-exchange relaxation. A precise description of the relaxation in the case of intermediate polarisation requires considering the nonlinear evolution of the atomic density matrix, which is much more complicated than the low-polarisation case. However, in most practical applications of SERF magnetometry, the nonlinear effects are rather small and will lead to a slight improvement of the low-polarisation case [Eq.(2.55)], and therefore it is less relevant to the topic of this work. More information on higher-order relaxation effects can be found in Refs. [94–96].

2.1.5. LIGHT-ATOMS INTERACTION

So far it was assumed that the alkali atoms are “in the dark”, ignoring their possible interaction with light. However, the interaction of alkali-metal atoms with light is crucial for the operation of the SERF magnetometer. The light is used for pumping the system and measuring its state. It is also a source of systematic effects, which may affect the performance of the magnetometer. This section discusses the key light-atom interaction processes associated with the SERF magnetometers.

LIGHT ABSORPTION

The interactions between light and atoms depend on the light frequency detuning from the atomic transitions. Alkali-metal-based optical magnetometers usually employ light

tuned to either D_1 or D_2 lines (see Fig. 2.2) of the elements for both pumping and probing the ground state employed for the measurement of the magnetic field. The simplest process which may arise during light across the medium is absorption. The cross-section for absorption of monochromatic light with frequency ν propagating through the unpolarised atomic vapour is given by the following expression[80]

$$\sigma(\nu) = cr_e f \mathcal{L}(\nu), \quad (2.57)$$

where c is the speed of light, r_e is the classical electron radius, and f is the oscillator strength of the transition. $\mathcal{L}(\nu)$ is the spectral profile of the corresponding transition. The intensity of the near-resonant light traversing the medium is given by

$$I_T(\nu, l) = I_0 e^{-N_a \sigma(\nu) l}, \quad (2.58)$$

where I_0 is the initial intensity of the light, N_a is the concentration of atoms absorbing light, and l is the optical path of the light in the medium.

The cross-section obeys the following summation rule [80]

$$\int \sigma_i(\nu) d\nu = \pi cr_e f_i, \quad (2.59)$$

which might be useful for the cell characterisation purposes, while in combination with Eq. (2.58) can be used to determine the alkali-metal concentration in the given cell.

In this work, optical magnetometers with a buffer gas are under consideration (see Sec. 2.1.3). In this case, spectral profiles of optical transitions are predominantly determined by the collisions with the buffer-gas atoms. Such collisions have three main effects on the spectral lines: spectral-line broadening, shifting their central frequency, and generating asymmetry of the lines. In this case, the spectral line profile can be described by the modified Lorentzian profile [97]

$$\mathcal{L}(\nu) = \frac{\frac{\Gamma}{2} (1 + 0.664 \frac{2\pi T_d \Delta\nu}{\Gamma})}{\Delta\nu^2 + (\frac{\Gamma}{2})^2}, \quad (2.60)$$

where Γ is the full width at the half maximum of the pressure-broadened spectral line, $\Delta\nu$ is the detuning from the centre of the collisionally shifted line, while T_d is the approximate collisional duration, which plays a role in the dispersion asymmetry parameter in the line profile.⁷

The width of the line spectral profile is determined by the noncollisional rate Γ_N , given by its natural line width and Doppler broadening of the transition, and the collisional rate Γ_c ,

$$\Gamma = \Gamma_N + \Gamma_c. \quad (2.61)$$

The collisional broadening is proportional to the number density of buffer-gas atoms and depends on the temperature of the cell. Following Ref. [97], this dependence can be parameterised in the following way

$$\Gamma_c(n_{bg}, T) = \alpha n_{bg} \left(\frac{T}{T_0} \right)^q + \varrho, \quad (2.62)$$

⁷This work is focused on collisions between alkalis and He, which are rather sudden $T_d \sim 10^{-12}$ s.

Transition	Collisional broadening			Collisional shift		
	α	q	ϱ	α'	q'	ϱ'
	[GHz/amg]		[GHz]	[GHz/amg]		[GHz]
$^{Rb}D_1$	18.31±0.07	0.26±0.04	-0.19±0.13	5.46±0.05	0.38±0.06	0.24±0.13
$^{Rb}D_2$	20.51±0.08	0.39±0.04	-0.35±0.15	0.63±0.04	1.42±0.43	0.20±0.12
$^K D_1$	14.26±0.09	0.44±0.06	0.04± 0.11	1.36±0.06	- 0.14±0.36	0.28±0.12
$^K D_2$	19.59±0.10	0.39±0.05	0.11±0.13	0.69 ±0.06	- 2.04±0.72	0.02± 0.12

Table 2.3: Experimental parameters for the pressure broadening [Eq. (2.62)] and shift [Eq. (2.63)] of the first two optical transitions of potassium and rubidium in atmosphere of ^3He . The parameters taken from Ref. [97].

where T is the temperature of the cell in kelvins, n_{bg} is the concentration of the buffer gas measured in amagats⁸ $T_0 = 353$ K, and α , q and ϱ are the experimental parameters. The experimental parameters for potassium and rubidium D lines relevant to this work are presented in Table 2.3. For the SERF magnetometers, the spectral width of the optical transitions is dominated by collisions broadening $\Gamma_N \ll \Gamma_c$, and therefore, the measurement of the spectral width may be used for practical determination of the buffer-gas concentration.

Detuning from the centre of the spectral line Δ_ν is determined by the central frequency of unperturbed transition ν_0 and collisional frequency shift ν_c and the frequency of the light wave interacting with atoms ν

$$\Delta_\nu = \nu - \nu_0 - \nu_c. \quad (2.63)$$

The collisional frequency shift has a similar dependence on the concentration of the buffer gas and temperature as the broadening [97]

$$\nu_c(n_{bg}, T) = \alpha' n_{bg} \left(\frac{T}{T_0} \right)^{q'} + \varrho', \quad (2.64)$$

where α' , q' and ϱ' are the experimental parameters. The relevant values of experimental parameters are presented in Table 2.3.

OPTICAL PUMPING

From the perspective of (co)magnetometry, the control over atomic polarisation is of paramount importance. In general, the optical polarisation of atoms requires their interaction with appropriately polarised (pumping) light. This section discusses optical pumping under the SERF regime using light tuned to the D_1 and D_2 lines.

⁸Amgamat (amg), is the unit of the concentration equivalent to $2.6868 \cdot 10^{19} \text{ cm}^{-3}$ [92]. It corresponds to the concentration of the ideal gas at a pressure of 1 atm at temperature 0°C.

At a most fundamental level, optical pumping can be considered as a process of transferring energy and angular momentum from light to atoms. Therefore, the state of the atoms pumped with continuous-wave (CW) pumping light for a sufficiently long time (the steady state is reached) is an outcome of the “competition” between the energy and momentum transfer and dissipation processes.⁹

Since the pumping light couples directly to the electrons of the alkali metal, one can ignore the nuclear spin in the considerations and study only the electron-state evolution during the process. For the upcoming discussion, it is assumed that the pumping light is σ^+ -polarised and propagates along the quantisation axis, i.e., along the z -axis. This simplifies the description of the pumping process, which leads to the redistribution of the atomic populations among the Zeeman sublevels of the ground and excited states and does not generate any coherences between the sublevels within a given state. When such symmetry is not present, or in the case of a magnetic field applied at some axis with respect to the propagation direction, the description would need to take into account coherences arising between magnetic sublevels. The analysis also assumes that the excitation rate is much smaller than the excited-state relaxation rate. Thereby, as the atoms reside in the ground state for most of the time, the role of the excited state can be reduced to an intermediate step in the transition between ground-state sublevels, and hence, the state can be excluded from the description of the pumping process.

The assumptions given above allow one to describe the pumping process using rate equations for populations of the ground-state sublevels, where the dynamics related to the excited state are taken into account in the rates of repopulation between pairs of the atomic sublevels. Before the start of a discussion of the specific dynamics of the atomic populations in the process of pumping at the D_1 and D_2 optical transitions, the excitation and relaxation processes determining specific repopulation rates are considered, which are generic for both cases.

Absorption of a photon from the σ^+ -polarised pumping beam leads to the transition of the atom from the ground to an excited state with the magnetic quantum number increased by 1 (-1 for the σ^- -polarised light). After some time in the excited state, the atom returns to the ground state. In the simplest scenario, when atoms do not collide with one another when in the excited state, the decay originates from the spontaneous emission, and therefore, the atom initially present in the excited state of a magnetic quantum number m' can only relax into the states with magnetic quantum numbers m either equal or different by ± 1 from m' . The probability of the relaxation into a certain ground state is governed by the Fermi golden rule and branching coefficients and can be calculated using the Clebsch-Gordan coefficients. However, the presence of the buffer gas modifies the branching coefficients [98]. Specifically, collisions with the buffer gas mix the populations of the excited-state Zeeman sublevels, leading to the equalisation of the excited-state populations. As in considered conditions, this process is much faster than the spontaneous emission, it is assumed that the excited-state sublevels are always equally populated.

The secondary photons reemitted during the spontaneous emission have random polarisation and propagation direction. Due to these random properties, reabsorption of the photons by the alkali atoms is a source of ground-state relaxation. This limits the

⁹In this context, the Larmor precession is considered as the dissipation process.

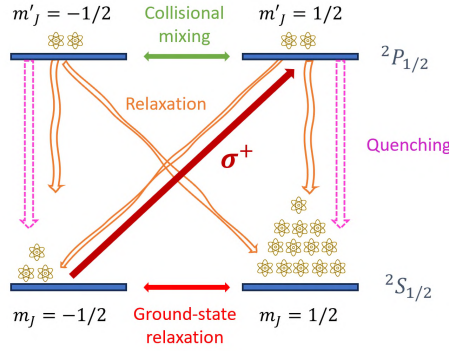


Figure 2.4: Scheme of the optical pumping of the alkali metal atoms with the circularly polarised (σ^+) light resonant with D_1 transition. The arrows on the scheme demonstrate the key processes responsible for the transitions between energy levels of atoms determining the efficiency of the pumping: the optical transition between the ground and excited states (the dark red arrow), relaxation of the ground state (the red arrow), relaxation of the excited state due to the spontaneous emission (the orange arrows), the photons transition from the excited state to the ground due to the collisions with the ground state (the magenta arrow), the collisional mixing of the excited state (the green line).

efficiency of optical pumping. To suppress the effect, a small¹⁰ amount of molecular gas, e.g., N_2 , is added to the vapor cell. Collisions with the molecules enable nonradiative decay of the alkali atoms to the ground state, as the energy and orbital angular momentum of excited alkali-metal atoms are transferred to internal molecular degrees of freedom of molecular gas. Importantly, such a photonless transition does not change the magnetic quantum number. The process of nonradiative decay due to the molecular gas is called **quenching**, and the gas itself is often called a quenching gas.

PUMPING AT THE D_1 LINE

One can consider the specific example of the pumping with the light resonant with the D_1 transition in the presence of both buffer and quenching gases (Fig. 2.4). In this case, both excited and ground states have $J = S = 1/2$, therefore, there are only two sublevels in each manifold. The pumping light with σ^+ -polarisation drives transition $m_J = -1/2 \rightarrow m'_J = 1/2$, and the sublevel $m_J = 1/2$ does not interact with the light, i.e., it is the dark state. After the excitation, the populations of the excited-state sublevels are equalised by collisions with the buffer-gas atoms. Then, the quenching process transfers populations to the ground state. The combination of these two processes makes equally probable the transition from the excited state to both sublevels of the ground state.

The presence of the dark state enables full polarisation of the atomic ensemble (by transferring an entire population of atoms into the state). In such a case of maximal saturation, one may expect near-zero absorption of the pumping light. To describe the polarisation process for intermediate polarisation, one should consider the probability of the excitation from all hyperfine sublevels involved in the considered optical transition. In this work, optical pumping with laser light is considered. The spectral width of the semiconductor lasers, which are typically used for the optical magnetometers, is at

¹⁰Concentration of the quenching gas is usually smaller than concentration of the buffer gas.

least three orders of magnitude narrower than the pressure-broadened spectral lines of the alkali metal, and for this reason, it is safe to assume that the pumping light is monochromatic. The average absorption (pumping) rate for an unpolarised atom is determined by the sum of the absorption rate for each individual transition

$$R_p = \Phi(\nu) \sum_i \sigma_i = c r_e f \Phi(\nu) \sum_i A_i \mathcal{L}(\Delta_i), \quad (2.65)$$

where $\Phi(\nu)$ is the photon flux, Δ_i is the detuning from the transition between two specific hyperfine levels, and i goes over all possible transitions. A_i is a relative strength of the transition, which is given by the following expression [99]

$$A_i = \frac{(2F+1)(2F'+1)}{2I+1} \left\{ \begin{matrix} J & J' & 1 \\ F' & F & I \end{matrix} \right\}^2, \quad (2.66)$$

where the expression in the curly bracket is the Wigner 6-j symbol, and the prime was used to identify quantum numbers of the excited state. Note that relative strengths for a given transition add up to unity

$$\sum_i A_i = 1. \quad (2.67)$$

The pressure broadening in the high-pressure cells (with a buffer-gas concentration above 1 atm) is at least an order of magnitude larger than the splitting between the hyperfine levels. This allows approximating of the superposition of the multiple hyperfine transitions with a single spectral profile centred at the average frequency and which line shape is given by Eq. (2.60). Taking into account Eq. (2.67), this also simplifies Eq. (2.65) for the pumping rate

$$R_p = c r_e f \Phi(\nu) \mathcal{L}(\nu). \quad (2.68)$$

Since this is the average pumping rate for the unpolarised atomic ensemble and only one sublevel interacts with the pumping light, the state $m_J = -1/2$ is depopulated at rate $2R_p$. This leads to the following rate equation for the populations of the ground-state sublevels

$$\begin{cases} \frac{d|\downarrow\rangle}{dt} &= -2R_p |\downarrow\rangle + \frac{1}{2} \cdot 2R_p |\downarrow\rangle \\ \frac{d|\uparrow\rangle}{dt} &= \frac{1}{2} \cdot 2R_p |\downarrow\rangle \\ 1 &= |\downarrow\rangle + |\uparrow\rangle \end{cases}, \quad (2.69)$$

where $|\downarrow\rangle$ and $|\uparrow\rangle$ denote normalised populations of the magnetic states with $m_J = -1/2$ and $m_J = 1/2$ respectively, and the last equations embody the assumption of the weak pumping coupling, according to which the entire population of atoms is at the ground state.

In Sec. 2.1.4, it was shown that in the context of SERF magnetometry, an ultimate interest lies upon the dynamics of electron polarisation [Eq. (2.49)], which has the following form in terms of the populations which was discussed above

$$\Upsilon = |\uparrow\rangle - |\downarrow\rangle. \quad (2.70)$$

Using Eq. (2.69) one obtains

$$\frac{dY}{dt} = 2R_p |\downarrow\rangle = R_p(1 - Y). \quad (2.71)$$

Considering relaxation of the electron polarisation (see Sec. 2.1.3)

$$\frac{dY}{dt} = 2R_p |\downarrow\rangle = R_p(1 - Y) - R_{SD}Y, \quad (2.72)$$

the polarisation change over time (here, it is assumed that the pumping starts with the unpolarised ensemble) is given by

$$Y(t) = Y_{D1} [1 - e^{-(R_{SD} + R_p)t}], \quad (2.73)$$

where Y_{D1} is the equilibrium polarisation

$$Y_{D1} = \frac{R_p}{R_p + R_{SD}}. \quad (2.74)$$

Note that from the equation above, it is possible to fully polarise the ensemble

$$\lim_{R_p \rightarrow \infty} Y_{D1} = 1, \quad (2.75)$$

which is the result of the existence of the dark state in the system.

So far, the problem was considered in a single dimension, i.e., the polarisation was building up along the pump beam in absence of the external fields. The discussion, however, can be easily generalised to the three-dimensional case

$$\frac{d\mathbf{Y}}{dt} = R_p \left(\frac{\mathbf{s}}{s} - \mathbf{Y} \right) - R_{SD} \mathbf{Y}, \quad (2.76)$$

where $\mathbf{s}$ is the pumping vector, defining the direction and degree of circular polarisation of the pumping light.

Equation (2.76) was obtained based on the discussion of the electron-spin evolution. However, the electron spin is coupled to the nuclear spin via the hyperfine interaction, which leads to the transfer of the polarisation to the nuclear spin as well. Moreover, the spin-exchange collisions lead to the spin-temperature distribution with a non-zero value of the spin temperature due to the optical pumping. It is noteworthy that the provided model is fully accurate only when the applied magnetic or pseudomagnetic fields are applied along the pumping beam. When the fields are not colinear, there is no single symmetry axis in the problem, and therefore it is not possible to choose the quantisation axis such, that the state of the atoms can be accurately described by the populations. However, the consideration is limited to the case of low magnetic fields, where the SERF regime is observed, and the spin-temperature distribution is assumed to be approximately conserved. This allows adding the pumping term into Eq. (2.53), which gives a complete model for the SERF magnetometer.

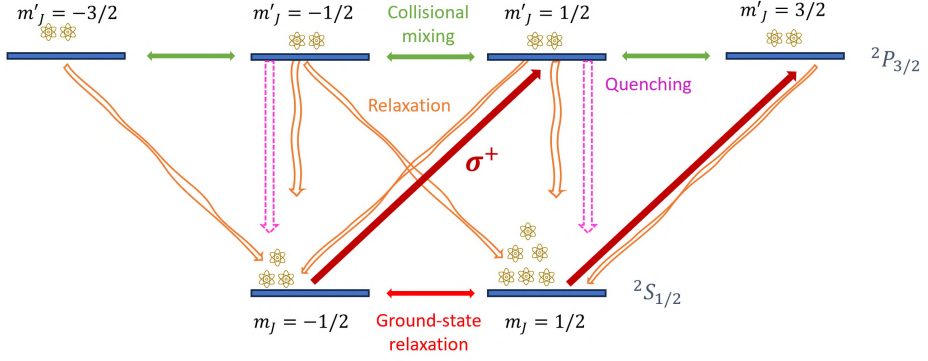


Figure 2.5: Scheme of the optical pumping of the alkali metal atoms with the circularly polarised (σ^+) light resonant with D_2 transition. The key processes involved in the optical pumping are presented in a way similar to Fig. 2.4.

PUMPING AT THE D_2 LINE

This section considers a similar situation as described above, with the exception of pumping with light tuned to the D_2 line (see Fig. 2.5). Similar to the previous discussion, the considerations start with the electron polarisation, ignoring the hyperfine interaction.

The D_2 transition occurs between the ground state of $J = 1/2$ and the excited state of $J = 3/2$. The higher multiplicity of the excited state results in the lack of the dark state, i.e., all sublevels of the ground state interact with the light. Hence, in the presence of the collisional mixing of the excited state and quenching gas, it is not possible to fully polarise the ensemble with the D_2 tuned light.

To get the specifics of the pumping process, the rate equations similar to those presented in the previous section are formulated. Using the Wigner-Eckart theorem, one can show that the transition driven by circularly polarised (σ^+) light from the ground sublevel of $m_J = -1/2$ is three times weaker than the similar transition from the sublevel $m_J = 1/2$ [99], which gives the depopulation rates $R_p/4$ and $3R_p/4$, respectively. The presence of the collisional mixing equalises the populations of the excited states; therefore, $1/4$ of the excited atoms will be in the states $m'_J = \pm 1/2$. In turn, the collisions with atoms of the quenching gas suppress the radiative transitions to the ground state. Hence, one can assume that the excited atoms undergo the transition to the ground state only via quenching. Thus, transitions from excited with $m'_J = \pm 1/2$ to the ground state are equally probable with a transition rate equal to $1/2$. Therefore, the combination of collisional mixing of excited states and quenching led to the repopulation rates for both ground sublevels being equal to $1/8$ of the total excitation rates. This can be summarised in the following rate equations

$$\begin{cases} \frac{d|\downarrow\rangle}{dt} = -\frac{1}{4}R_p|\downarrow\rangle + \frac{1}{8}\left(\frac{1}{4}R_p|\downarrow\rangle + \frac{3}{4}R_p|\uparrow\rangle\right) \\ \frac{d|\uparrow\rangle}{dt} = -\frac{3}{4}R_p|\uparrow\rangle + \frac{1}{8}\left(\frac{1}{4}R_p|\downarrow\rangle + \frac{3}{4}R_p|\uparrow\rangle\right) \\ 1 = |\downarrow\rangle + |\uparrow\rangle \end{cases}, \quad (2.77)$$

which then leads to the following equation for the evolution of the electron polarisation

$$\frac{dY}{dt} = -\frac{1}{4}R_p(1 + 2Y). \quad (2.78)$$

After including the relaxation, the polarisation dynamics (with the assumption of the zero polarisation at $t = 0$) is described by

$$Y(t) = Y_{D_2} \left[1 - e^{-(R_{SD} + R_p/2)t} \right], \quad (2.79)$$

where the equilibrium polarisation for pumping at D_2 transition is

$$Y_{D_2} = -\frac{R_p}{4 \left(R_{SD} + \frac{R_p}{2} \right)}. \quad (2.80)$$

One can note that Y_{D_2} has the opposite sign compared to the Y_{D_1} . Also, the equation above reveals the limitation of the achievable polarisation in the case of optical pumping with light tuned to the D_2 line

$$\lim_{R_p \rightarrow \infty} (Y_{D_2}) = \frac{1}{2}. \quad (2.81)$$

The generalisation of the result for the three-dimensional polarisation problem leads to the following equation

$$\frac{dY}{dt} = -\frac{1}{4}R_p \left(\frac{s}{s} + 2Y \right) - R_{SD}Y. \quad (2.82)$$

The polarisation process using light tuned to the D_2 line is (in general) less efficient. This can be illustrated by a comparison of the equilibrium polarisations for the same values of the relaxation and pumping rates presented in Fig. 2.6(top). One can note that the maximum polarisation, which can be obtained with D_2 -tuned light is twice smaller than for the D_1 tuning. Moreover, for the unsaturated case, the pumping rate necessary for obtaining the same polarisation level must be twice larger for the D_2 pumping than for the D_1 pumping. Using Eqs. (2.73) and (2.79), one can also compare the characteristic time scale of reaching the equilibrium polarisation in both cases

$$\tau_{D_1} = \frac{1}{R_{SD} + R_p}, \quad (2.83)$$

$$\tau_{D_2} = \frac{1}{R_{SD} + R_p/2}. \quad (2.84)$$

The dependence of these two quantities on the pumping rate is shown in Fig. 2.6(bot-tom). One can note that for pumping with the D_2 transition, the system reaches equilibrium much longer than in the case of the D_1 pumping. The difference in the efficiency of the pumping makes the pumping with D_1 transition a more preferable choice for the optical magnetometers.

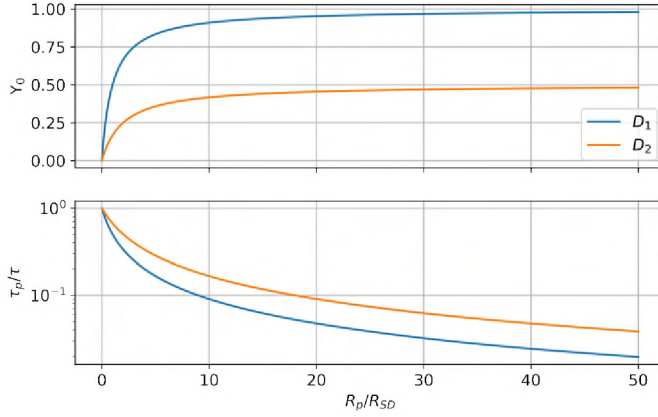


Figure 2.6: Efficiency of the optical pumping with light tuned to the D_1 and D_2 lines. The plot on the top demonstrates equilibrium polarisation for different values of the pumping rate normalised to the relaxation rate. The plot on the bottom demonstrates time of pumping normalised to the lifetime of the polarisation in the dark $\tau = R_{SD}^{-1}$ for different values of the pumping rate normalised to the relaxation rate.

DETECTION OF THE SPIN STATE - OPTICAL ROTATION

To utilise atomic spins for any kind of measurement, one has to gain information about their state. In alkali-metal-based magnetometers, this may be done by observation of the optical rotation of linearly-polarised probe light (Fig. 2.7). The observed optical rotation originates from circular birefringence associated with spin orientation. The following discussion presents the processes responsible for the optical rotation in polarised alkali-metal atoms and provides an estimation of the angle of polarisation rotation for the probe light tune to the D_1 and D_2 lines.

One can consider the propagation of the linearly polarised probe laser beam through the cell in the SERF magnetometer. The probe light is considered classically, i.e., it is assumed that the light is a monochromatic linearly-polarised electromagnetic wave propagating along the x -axis.

$$\mathbf{E}(x, t) = \frac{E_0}{\sqrt{4}} \left(e^{i(k_+ x - \omega t + \phi_0)} \mathbf{e}_+ + e^{i(k_- x - \omega t)} \mathbf{e}_- \right) + c.c., \quad (2.85)$$

where E_0 is the amplitude of the electric field, $k_{\pm}$ are the wave vectors for the $\sigma^{\pm}$ polarisation components of light, respectively, $\omega = 2\pi\nu$ is the light angular frequency, and ϕ_0 stands for the relative phase between the polarisation components. $\mathbf{e}_+$ and $\mathbf{e}_-$ are the unit vectors, corresponding respectively to the σ^+ and σ^- polarisation components, and defined as

$$\begin{aligned} \mathbf{e}_+ &= \mathbf{z} + i\mathbf{y}, \\ \mathbf{e}_- &= \mathbf{z} - i\mathbf{y}. \end{aligned} \quad (2.86)$$

When k_+ and k_- are the same (the absence of the circular birefringence), the relative phase ϕ_0 determines the angle between the y -axis and the polarisation of the light

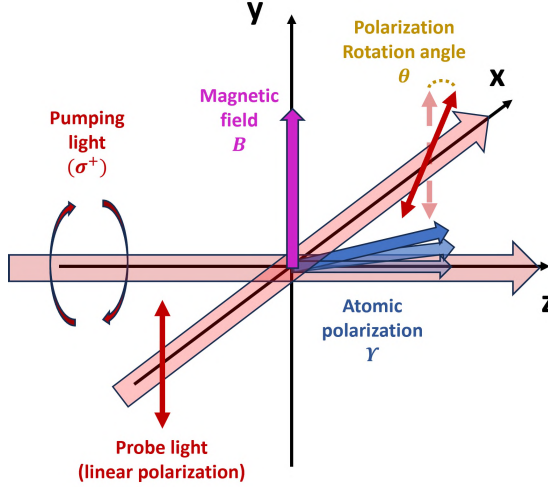


Figure 2.7: Schematic representation of the polarisation probing in the SERF magnetometer. The detection of the spin polarisation state is realised via measurement of the polarisation rotation of the linear polarised probe light. The rotation is proportional to the projection of the atomic polarisation at the probe-light propagation direction. In the scheme, the circularly polarised pump light is used to initially polarise the atomic ensemble in the direction perpendicular to the probe-beam axis. The atomic-polarisation projection on the probe axis is changing under influence of the magnetic field, hence its detection provided measurement of the magnetic field applied to the system.

through the relation

$$\theta = \frac{\phi_0}{2}. \quad (2.87)$$

In the optically active medium $k_+ \neq k_-$, the phase difference between the two circular polarisation components is a function of the light propagation path in the medium. If one assumes that the probe-light polarisation before the vapour cell is colinear with the y -axis ($\phi_0 = 0$) and the propagation length in the cell is l , the rotation angle after the cell can be easily calculated from the phase difference

$$\phi = \text{Re}(k_+ - k_-)l, \quad (2.88)$$

where the wave vector components are given by

$$k_{\pm} = \frac{\omega}{c} n_{\pm}, \quad (2.89)$$

with $n_{\pm}$ being the refractive indices for each polarisation. Taking into account Eqs. (2.87) and (2.88), one can write the polarisation rotation of light after the medium

$$\theta = \frac{\omega l}{2c} \text{Re}(n_+ - n_-). \quad (2.90)$$

As discussed in the subsection about light absorption, the atomic transitions in the cells with a high-pressure buffer gas are significantly modified by the collisions with the

buffer-gas atoms. In particular, the spectral lines for the absorption are shifted, broadened, and deformed into an asymmetric Lorentzian shape [Eq. (2.60)]. Similar effects are expected to appear in the spectrum of the optical rotation since the absorption coefficient $a(\nu)$ is determined by the imaginary part of the refractive index

$$a_{\pm}(\nu) = \text{Im}(k_{\pm}) = \frac{2\pi\nu}{c} \text{Im}(n_{\pm}) = \frac{1}{2} N_{\pm} \sigma_{\pm}(\nu), \quad (2.91)$$

where the last equality comes from Eq. (2.58) and the fact that $I \sim |\mathbf{E}|^2$. Here, $N_{\pm}$ denotes the number of atoms which can interact with $\sigma^{\pm}$ -polarised light.

Equation (2.91) establishes the connection between pressure broadened absorption spectrum [Eqs. (2.57) and (2.60)] and the refractive indices of the medium. This connection allows on to use the experimentally measured absorption line shapes (the imaginary part of the refractive index) for the estimation of the optical rotation as a function of the detuning from the centre of the pressure-broadened optical transition (the real part of the refractive index). To solve this problem, one can parameterise the refractive index with the electric susceptibility $\chi_{\pm}$ for the $\sigma^{\pm}$ polarisations

$$n_{\pm} = \sqrt{1 + \chi_{\pm}} \approx 1 + \frac{1}{2} \chi_{\pm}, \quad (2.92)$$

where it is assumed that $\chi_{\pm} \ll 1$, which is an adequate assumption for the alkali-metal vapours. The electric susceptibility obeys the Kramers-Kronig relations [100]

$$\begin{aligned} \text{Re}[\chi_{\pm}(\nu)] &= \frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\text{Im}[\chi_{\pm}(\nu')]}{\nu' - \nu} d\nu', \\ \text{Im}[\chi_{\pm}(\nu)] &= -\frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\text{Re}[\chi_{\pm}(\nu')]}{\nu' - \nu} d\nu', \end{aligned} \quad (2.93)$$

where $\mathcal{P}$ stands for the Cauchy principal value. The Kramers-Kronig relations, combined with Eq. (2.92), lead to the following relation between real and imaginary parts of the refractive index

$$\begin{aligned} \text{Re}[n_{\pm}(\nu)] &= 1 - \frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\text{Im}[n_{\pm}(\nu')]}{\nu' - \nu} d\nu' \\ &= 1 - \frac{cN_{\pm}}{4\nu\pi^2} \mathcal{P} \int_{-\infty}^{\infty} \frac{\sigma_{\pm}(\nu')}{\nu' - \nu} d\nu' \end{aligned} \quad (2.94)$$

where Eq. (2.91) was used in the second part of the relation.

Until now the discussion was general, but hereafter the considerations of probing at the D_1 and D_2 lines start to differ due to differences in transition to specific hyperfine levels.

For further discussion, it is useful to express the population of the ground-state levels through the projection of the electron polarisation on the probe-light propagation

direction¹¹

$$\begin{aligned} N_{|\downarrow\rangle} &= \frac{1 - Y_x}{2} N, \\ N_{|\uparrow\rangle} &= \frac{1 + Y_x}{2} N. \end{aligned} \quad (2.95)$$

In the case of the D_1 excitation (Fig. 2.4), each component of the circular polarisation can interact with a single energy level. The absorption cross-section profile is assumed to be the same for both component and

$$N_{\pm}^{D_1} \sigma_{\pm}^{D_1} = 2(1 \mp Y_x) \sigma^{D_1}(\nu), \quad (2.96)$$

where the relative strength of the transition has been taken into account, and where σ_{D_1} is the absorption cross section for D_1 transition calculated according to Eq. (2.57). This leads to the following expression for the optical-rotation angle

$$\theta_{D_1} = -\frac{1}{2} c r_e N l f_{D_1} \mathcal{D}_{D_1}(\nu) Y_x, \quad (2.97)$$

where the spectral profile of the optical rotation given by Eq. (2.60) is described with the following expression

$$\mathcal{D}(\nu) = -\frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\mathcal{L}(\nu')}{\nu' - \nu} d\nu' = \frac{\Delta_\nu + 0.664 \cdot 2\pi T_d (\Gamma/2)^2}{\Delta_\nu^2 + (\Gamma/2)^2}, \quad (2.98)$$

and the subscript in Eq. (2.97) denotes the specific transition.

In the case of the D_2 transition, there are two allowed transitions for each circular-polarisation component, which have different relative strengths. Therefore, in this case, the total absorption of the specific polarisation component depends on the populations of both atomic transitions (Fig. 2.5). Therefore, considering both polarisation components, one obtains

$$N_{\pm}^{D_2} \sigma_{\pm}^{D_2}(\nu) = \frac{1}{2} \left(1 \pm \frac{Y}{2} \right) \sigma^{D_2}(\nu), \quad (2.99)$$

where σ_{D_2} is the absorption cross section for D_2 transition calculated according to Eq. (2.57). This leads to the following expression for the rotation angle

$$\theta_{D_2} = \frac{1}{4} c r_e N l f_{D_2} \mathcal{D}_{D_2}(\nu) Y_x. \quad (2.100)$$

The presented considerations demonstrated that for the light tuned to either of the D transitions, the rotation angle for the optical polarisation is proportional to the projection of the atomic polarisation on the probe-light propagation axis. Hence, the optical rotation provides an efficient interface for determining the atomic spin state. The efficiency of the monitoring for both transitions is comparable since the difference in the

¹¹Note that here the quantisation axis is chosen to be parallel to the probe light propagation direction, which differs from the quantisation axis, which was chosen earlier for the description of the optical pumping. This is indicated by the subscript x in Y_x .

numeric factors in Eqs. (2.97) and (2.100) is compensated by the difference in the transition strength $f_{D_2}/f_{D_1} \approx 2$. Although there are no solid arguments for the choice of any particular transition for polarisation detection, as was pointed out in Ref. [101], for better stability of the signal, one can choose the probe laser frequency between resonant frequency for D_1 and D_2 transitions, since in this regime the signal is basically independent of the laser frequency drifts.

LIGHT SHIFT

The optical excitation and polarisation change are not the only effects induced by light on the alkali-metal atoms. Specifically, an oscillating electric field of light may generate the **AC Stark effect**, in which the light changes the energies of the ground-state sublevels. This affects the electron spins in a similar way as the magnetic field applied along the propagation axis of the light. Thereby uncontrolled light shifts produced by the pumping or probe beam can be misinterpreted as the magnetic field. This is why, in the context of optical magnetometry, the light shift is often considered as a source of systematic errors. But the light shift may also be used in controlled manipulation of the electron spins of the alkali-metal atoms. For example, it may be used to induce electron non-magnetic couplings as this perturbation does not other spins in the system. The light shifts, from both (negative and positive) perspectives, are considered in this work.

The light shift is closely related to the absorption of the circularly polarised light. These processes can be considered as a result of the unitary and dissipative evolution of the alkali-metal ground state driven by the electric field of light. For this interaction, the effective Hamiltonian for the ground state, in the presence of the high-pressure buffer gas, has the following form [92]

$$H_{em} = \left(\delta \mathcal{E} - \frac{i\hbar}{2} \delta \Gamma \right) (1 - 4K \mathbf{s} \cdot \hat{\mathbf{S}}), \quad (2.101)$$

where

$$K = \frac{(-1)^{(J'-S)}}{2J'+1}, \quad (2.102)$$

and the unitary part $\delta \mathcal{E}$ is the light shift amplitude, Γ is the dissipation rate, and $\mathbf{s}$ is the average spin of a photon determined by the versor of light polarisation $\mathbf{e}$ through

$$\mathbf{s} = i\mathbf{e} \times \mathbf{e}^*. \quad (2.103)$$

The scalar coupling between the average spin of the photon and the electron spin allows consideration of the unitary part of light-atom interaction [Eq. (2.101)] in a similar way as the magnetic-field interaction [Eq. (2.5)]. This enables to interpret the perturbation due to the light shift as the effective magnetic field generated

$$\mathbf{B}_{LS} = \frac{4K\delta \mathcal{E}}{\gamma_e} \mathbf{s}. \quad (2.104)$$

The dissipative part of Eq. (2.101) corresponds to the absorption coefficient [Eq. (2.68)]

$$\langle \delta \Gamma \rangle = \text{Tr}(\delta \Gamma \rho) = R_p(\nu) = \Phi \sigma(\nu). \quad (2.105)$$

Since the process of the photon absorption is causal (no excitation is possible before the application of resonant light), $\delta\Gamma$ and $\delta\mathcal{E}$ must obey the Kramers-Kroninger relations defined in Eq. (2.93) [102]. This allows us to get the latest quantity based on the experimental line shape of the absorption Eq. (2.60)

$$\delta\mathcal{E} = \frac{\hbar}{2\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\delta\Gamma}{v' - v} dv' = \frac{\hbar c r_e f \Phi}{2} \mathcal{D}(v), \quad (2.106)$$

where Eq. (2.98) was used to determine the spectral profile of the absorption line. In such a case, the effective magnetic field, corresponding to the light shift, is given by

$$\mathbf{B}_{\text{LS}} = \frac{4K\hbar c r_e f \Phi}{2\gamma_e} \mathcal{D}(v) \mathbf{s}. \quad (2.107)$$

For a specific example, one can consider the light shift generated by the σ^+ -polarised pumping beam, which propagates along the z -axis, i.e., the same configuration as described in the subsection dedicated to optical pumping. In such a case, $\mathbf{s} = \mathbf{z}$, thus the pumping light can generate the pseudomagnetic field along the light-propagation direction. However, the light shift from the pumping laser can be avoided if it is tuned to the centre of the optical transition. This is so because the dispersion-like shape of the spectral profile $\mathcal{D}(v)$. When the light shift is introduced in the system to manipulate the electron spin, the laser light is far detuned. This is crucial for avoiding the unwanted pumping from the light used for light-shift generation. Fortunately, $\mathcal{L}(v)$ decays much faster on the wings than $\mathcal{D}(v)$ and it is possible to optimise the power and detuning of the light-shift beam so that the pumping rate is genially eliminated with the desirable pseudomagnetic field still present.

2.1.6. MAGNETIC FIELD MEASUREMENTS

To conclude this section, the described system is considered in real-world measurements of magnetic or pseudomagnetic fields. It is natural to start with the case of the SERF magnetometer placed in a near-zero magnetic field (e.g., inside a magnetic shield). In this case, the alkali-metal atoms experience only the fields of interest. The atoms are pumped with σ^+ -polarised pumping light tuned to the D_1 line, which propagates along the z -axis and generates initial spin polarisation along the axis. The polarisation is probed by the detection of polarisation rotation of the linearly-polarised probe light, blue-detuned with respect to the D_1 line and propagating along the x -axis. From Eq. (2.97), it is clear that this allows to probe Y_x spin-polarisation component. To maximise the amplitude of the optical rotation, light is detuned from the centre frequency by half of the optical transition width, $\Delta\nu = \Gamma/2$. The configuration is shown in Fig. 2.7. It should be stressed that in all cases described in this section, it is assumed that the polarisation is pumped to its steady state, $Y_z = Y_0$, prior to the measurements.

The evolution of electron polarisation in the SERF magnetometer is described with Eqs. (2.53) and (2.73). For convenience, the equation for the polarisation evolution is rewritten with the introduction of the steady-state polarisation Y_0 , defined in Eq. (2.74), and total relaxation rate $R_{\text{tot}} = R_p + R_{SD}$

$$\frac{d\mathbf{Y}}{dt} = \frac{1}{q_0} \left[\gamma_e \mathbf{B} \times \mathbf{Y} - R_{\text{tot}} (Y_0 \mathbf{z} - \mathbf{Y}) \right], \quad (2.108)$$

where the slowing-down factor is approximated with a constant value q_0 . This is possible because the alkali-metal polarisation does not change significantly during the measurements.

2

MEASUREMENTS OF SLOW-VARYING FIELDS

First, the response of the system to slow changes in the magnetic field is under consideration.¹² In that case, the system evolves adiabatically, remaining in the steady state at all times. From Eq. (2.108), the atoms are primarily sensitive to the perturbations applied along $\mathbf{y}$ which is perpendicular to both the probe and pump propagation directions

$$\mathbf{Y}_x \sim \mathbf{B} \times \mathbf{Y} \sim B_y \mathbf{Y}_0. \quad (2.109)$$

The steady-state solution to Eq. (2.108) reveals the dispersion-like magnetic resonance

$$\mathbf{Y}_x(B_y) = \frac{R_{tot} \gamma_e (B_y + B_x B_z)}{R_{tot}^2 + \gamma_e^2 (B_x^2 + B_y^2 + B_z^2)} \mathbf{Y}_0. \quad (2.110)$$

For the two remaining axes, the magnetometer has only second-order sensitivity. In turn, in most cases, these directions are neither considered nor used for the measurements (the contribution from the fields is often treated as the potential source of the systematic errors). Due to the small contribution from the directions, the discussion focuses mostly on the most sensitive direction. The width of the resonance is determined by the relaxation rate of the atomic polarisation, and its amplitude depends on the polarisation level.

For a near-zero field, close to the centre of the resonance, the response of the system is linearly proportional to the applied magnetic field

$$\mathbf{Y}_x(B_y) \approx \frac{\gamma_e \mathbf{Y}_0}{R_{tot}} B_y = \alpha_a B_y, \quad (2.111)$$

and the proportionality coefficient α_a can be treated as an atomic calibration factor. Such an approximation is valid when $B_y \ll R_{tot}/\gamma_e$.

MEASUREMENTS OF THE TIME-VARYING FIELDS

Atomic polarisation requires finite time to respond to changes in the measured magnetic field. Therefore, the magnetometer is capable of following the magnetic field only within the limited frequency bandwidth. To estimate the bandwidth of the magnetometer, the aim is to obtain the frequency response of the SERF magnetometer. For that, one can consider the magnetometer, which is perturbed by the harmonic monochromatic magnetic field of the frequency $f_m = \omega_m/(2\pi)$ and the amplitude B_{0y}

$$B_y(t) = B_{0y} \sin(\omega_m t) = \frac{B_{0y}}{2i} \left(e^{i\omega_m t} - e^{-i\omega_m t} \right). \quad (2.112)$$

¹²Although here the response of the SERF magnetometer to the magnetic field is under consideration, the discussion can be easily generalised to the non-magnetic fields because the qualitative properties of the response are the same for both types of perturbation [see Eq. (2.53)].

For small perturbations, such that

$$\frac{\gamma_e B_y}{q_0 \omega_m} \ll 1, \quad (2.113)$$

the relative changes of the longitudinal ($\mathbf{z}$) component of the polarisation is small. Thereby, it can approximate the longitudinal polarisation to be constant and equal to its equilibrium value. Under the approximation, the probed component of the polarisation driven by Eq. (2.112) has the following form¹³

$$\begin{aligned} Y_x(t, \omega_m) &= \frac{\gamma_e Y_0 B_{0y}}{\omega_m^2 + \left(\frac{R_{tot}}{q}\right)^2} \left[\frac{R_{tot}}{q} \sin(\omega_m t) - \omega_m \cos(\omega_m t) \right] \\ &= Y_s \sin(\omega_m t) + Y_c \cos(\omega_m t), \end{aligned} \quad (2.114)$$

where Y_s and Y_c are the in-phase and out-of-phase components of the output signal, respectively.

To get the frequency response of the system, one can present the result in the following manner

$$Y_x = Y_R \sin(\omega_m t + \phi), \quad (2.115)$$

where Y_R is the amplitude of the magnetometer response and ϕ is the phase

$$\begin{aligned} Y_R &= \sqrt{Y_c^2 + Y_s^2}, \\ \phi &= \arccos\left(\frac{Y_s}{Y_R}\right) \operatorname{sgn}\left[\arcsin\left(\frac{Y_s}{Y_R}\right)\right]. \end{aligned} \quad (2.116)$$

From Eq. (2.114) the amplitude of the response for the considered case takes the following form

$$Y_R = \gamma_e Y_0 B_{0y} \frac{R_{tot}}{q_0 \sqrt{\omega_m^2 + \left(\frac{R_{tot}}{q_0}\right)^2}}. \quad (2.117)$$

The normalised frequency response for the SERF magnetometer is plotted in Fig. 2.8.

The bandwidth of the magnetometer is defined as a cut-off frequency for which the amplitude of the response is attenuated to a certain value given by the definition. One can define the attenuation of the magnetometer response $\mathcal{A}$ as

$$\mathcal{A} = \frac{Y_R(\omega_B)}{Y_R(0)}, \quad (2.118)$$

where $f_B = \omega_B / (2\pi)$ is the system bandwidth corresponding to a given attenuation $\mathcal{A}$. One can also define the bandwidth by reversing the definition of the attenuation

$$f_B = \frac{R_{tot}}{q_0} \sqrt{\frac{1}{\mathcal{A}^2} - 1}. \quad (2.119)$$

For instance, for a commonly used definition of bandwidth associated with the -3 -dB cut-off, $\mathcal{A}_{3dB} = 0.708$, one gets

$$f_{-3dB} = 0.9974 \cdot \frac{R_{tot}}{q_0} \approx \frac{R_{tot}}{q_0}. \quad (2.120)$$

¹³Note that for $\omega_m = 0$ (constant perturbation) the result reduces to the case of slow-varying fields [given by Eq. (2.111)].

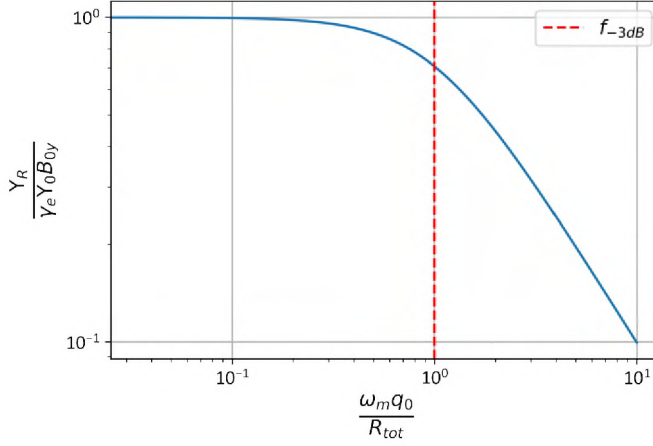


Figure 2.8: Normalised response amplitude of the SERF magnetometer obtained from Eq. (2.117). The red dashed line shows the -3-dB bandwidth defined in Eq. (2.120).

MAGNETIC RESONANCE IN THE NON-ZERO LEADING FIELD

Although the SERF magnetometer has the best performance in near-zero fields, for this work it is useful to consider detection of the magnetic resonance with a non-zero leading field. Here, the detected magnetic field is considered as a deviation from the leading-field value.

One can solve Eq. (2.108) assuming a constant, non-zero B_z , and oscillating B_y described by Eq. (2.112). Making the same assumption of the small perturbations as for Eq. (2.113), one can derive the expression for the response of the magnetometer

$$\begin{aligned} Y_x(t, \omega_m) &= Y_s \sin(\omega_m t) + Y_c \cos(\omega_m t) \\ &= \text{Im}[Y_- - Y_+] \sin(\omega_m t) + \text{Re}[Y_- + Y_+] \cos(\omega_m t), \end{aligned} \quad (2.121)$$

where $Y_{\pm}$ have a form of the complex Lorentzians centred on the frequency $|\Omega| = \gamma_e |B_z| / q_0$

$$Y_{\pm} = -\frac{\gamma_e B_0 Y_0}{2 \left[(\omega_m \mp \Omega)^2 + (R_{tot} / q_0)^2 \right]} \left(\omega_m \mp \Omega \pm i \frac{R_{tot}}{q_0} \right). \quad (2.122)$$

This configuration (combination of constant leading and oscillating transverse fields) can be used for measurements of either the leading field or oscillating perpendicular fields (RF mode of the magnetometer operation). Note, however, that in the case of SERF magnetometry, this regime has a limited applications, as the system falls out of the SERF regime with a growing magnetic field [Eq. (2.55)] and hence measurement sensitivity deteriorates.

SYNCHRONOUS PUMPING

Observation of the magnetic resonance at the SERF regime in non-zero leading field is possible not only with modulation of the magnetic field (Sec. 2.1.6), but also with modulation of the pumping rate. Such a technique is known in the literature as **synchronous pumping** [101].

One can consider Eq. (2.108) in the case of the constant non-zero leading field B_z , no transverse fields, and the pumping rate modulated in the following manner

$$R_p = \frac{R_{op}}{2} [1 + \sin(\omega_{op} t)]. \quad (2.123)$$

Here, ω_{op} is the frequency of the modulation, and R_{op} is the amplitude of the pumping-rate modulation. In practice, such modulation can be realised by varying the pumping-light intensity with a sinusoidal wave of a 100% modulation depth. One can show that in the technique, the response of the magnetometer at the first harmonic of the modulation frequency is similar to the response of the magnetometer to the oscillating magnetic field [Eq. (2.1.6)] [103]. However, in this case, the complex Lorentzians defining solution have a different form

$$\Upsilon_{\pm} = \pm \frac{1}{4} \frac{R_{op} [(R_{sd} + R_{op}/2) + i(\Omega \pm \omega_{op})]}{(R_{sd} + R_{op}/2)^2 + (\Omega \pm \omega_{op})^2}, \quad (2.124)$$

where Ω stands for the alkali-metal Larmor frequency corresponding to the leading field [as in Eq. (2.121)]. Similarly to the oscillating-field-induced magnetic resonance, the frequency response to the modulation of the pumping rate takes the form of the complex Lorentzian centred at the Larmor frequency of the magnetic field. Hence, synchronous pumping provides a means for the all-optical measurements of the non-zero magnetic field with the SERF system.

2.1.7. PARAMETRIC MODULATION

Many applications of optical magnetometers, including the searches for exotic spin couplings, may benefit from the simultaneous measurement of the magnetic field along multiple axes. The first-order sensitivity to magnetic fields along two perpendicular axes may be achieved by applying the magnetic field of the high [compared to the bandwidth of the magnetometer from Eq. (2.120)] frequency ω_{pm} , and the amplitude B_{pm} , applied along the pumping-beam propagation direction

$$\mathbf{B}_{pm} = B_{pm} \cos(\omega_{pm} t) \mathbf{z}. \quad (2.125)$$

The first-harmonic response of the magnetometer under such conditions is proportional to the field component colinear with the probe beam, while the second-harmonic response is proportional to the field component perpendicular to both probe and pump beams [104]

$$\Upsilon_x \approx \frac{\gamma_e B_x Y_0}{R_{tot}} 2J_0(u) J_1(u) \sin(\omega_{pm} t) + \frac{\gamma_e B_y Y_0}{R_{tot}} 2J_0(u) J_2(u) \cos(2\omega_{pm} t), \quad (2.126)$$

where J_i is the Bessel function of the i -th order and u is the unitless parameter, describing the dependence of the response amplitude on the frequency and the amplitude of the modulation

$$u = \frac{\gamma_e B_{pm}}{q \omega_{pm}}. \quad (2.127)$$

The expression for the response of the magnetometer to the parametric modulation [Eq. (2.126)] is obtained under the assumption of the strongest torque in the system being generated by the modulating field. The demodulation of the signal at the first and

second harmonics allows the simultaneous detection of the perturbations along $\mathbf{x}$ and $\mathbf{y}$. Importantly, the parametric modulation does not affect the original bandwidth of the SERF magnetometer.

In addition to the two-axis sensitivity, the parametric modulation may also suppress low-frequency noise present in the magnetometer and originating, for example, from airflow in beam paths. Such noise is often one of the main limitations of the SERF-magnetometer performance, hence the parametric modulation may allow to increase the sensitivity of the magnetometer. It needs to be stressed that the amplitudes of the response (and therefore the calibration factors) to the perturbation along two sensitive axes, in general, are not the same. Moreover, from Eq. (2.126) and Eq. (2.127), one can note that the response amplitude in both cases is the function of the modulation-field amplitude B_{pm} and frequency ω_{pm} . Therefore, the amplitude and frequency of the modulating field have to be optimised depending on the aims of the parametric modulation application.

The downside of the parametric modulation is the possible cross-talk between the two sensitivity axes. In Ref. [104], the observed cross-talk is reported at a level of about 9% for both axes. Nevertheless, the ability to measure perturbation in two directions broadens the application of both SERF magnetometers and comagnetometers.

2.2. NOBLE-GAS-ALKALI-METAL COMAGNETOMETER

Efficient techniques of polarisation and detection of the nuclear-spin state open numerous experimental possibilities in science and industry. They include comagnetometry for fundamental physics studies [47], precise magnetometry [105–107], rotation sensing [105, 108–111], magnetic resonance imaging of lungs [112, 113] and (potentially) long-lived quantum memory [114–116], etc. The alkali-metal-noble-gas comagnetometer system has gained particular interest in the context of the searches for exotic spin couplings due to the extremely high sensitivity ensured by the high concentration and polarisation of the noble-gas nuclear spin in the system.

From a technical point of view, the comagnetometers are quite similar to the SERF magnetometers. The alkali metal atoms in the comagnetometer are operated in the SERF regime, therefore most of the alkali-metal properties discussed in the context of the magnetometry are inherited by the comagnetometer with some minor exceptions. The most significant difference between those systems arises from the cell content. The comagnetometer cell contains the noble gas with a non-zero nuclear spin instead of the spin zero buffer gas that is used in magnetometry. This leads to an additional degree of freedom in the system, which significantly modifies the response of the alkali-metal atoms to external perturbations.

The evolution of the noble-gas nuclear spin is determined by three main processes: the interaction with external (magnetic and non-magnetic) perturbations, the spin exchange interaction with alkali atoms, and the relaxation of the spin states. In fact, the spin-exchange interaction between the electron spin of alkali metal and the nuclear spin of the noble gas plays a key role in the comagnetometry. First, it is used for the polarisation of the noble-gas spins without the necessity to directly optically polarise the atoms (the ground-state excitation of the noble gas requires the application of ultraviolet radiation). This allows for the use of high densities of noble gas. In turn, the high density

of the noble gas and the presence of the coherent part in the spin-exchange interaction provide a strong coupling between the polarisations, which ultimately leads to the self-compensation effect (Sec. 1.3.2), and allows the continuous monitoring of the coupled system state via observation of the alkali-metal atoms.

As the spin evolution of alkali-metal atoms was already discussed in the previous section, later in this chapter, the main focus is put on the evolution of the noble-gas spins and successively their coupled evolution with alkali-metal atoms. Particularly, the interaction with the external perturbations is discussed along with spin-exchange coupling, and relaxation of the noble gases. The discussion is narrowed down to the case of ^3He , which is a common choice for fundamental physics experiments due to its long coherence time. This atomic species is also used in Advanced GNOME comagnetometers, which are the main topic of this work.

2.2.1. NOBLE-GAS SPINS RESPONSE TO THE EXTERNAL PERTURBATIONS

The Hamiltonian for the interaction of the noble-gas nuclear spin G with the magnetic field $\mathbf{B}$ and pseudomagnetic field $\mathbf{b}_G$ placed in the non-inertial frame rotating with the frequency $\mathbf{\Omega}$ has the following form

$$H_{0,NG} = \gamma_n \hbar (\mathbf{B} + \mathbf{b}_G) \cdot \mathbf{G} + \hbar \mathbf{\Omega} \cdot \mathbf{G}, \quad (2.128)$$

where $\mathbf{b}_G$, similarly to the alkali-metal case [Eq. (2.5)], may be defined by the generic exotic perturbation Ξ and the coupling strength χ_G

$$\mathbf{b}_G = \frac{\chi_G}{\gamma_n G} \Xi. \quad (2.129)$$

The rotation term $\mathbf{\Omega}$ acts as an effective change of the Larmor frequency in the system.

The nuclear spins of the noble gas in the comagnetometers are commonly described in terms of its microscopic polarisations, which are defined by the expectation value of the nuclear spin under the assumption of the spin-temperature equilibrium [76]. This description is similar to the alkali-metal spins in the SERF magnetometer, which was discussed in the previous section. However, unlike the SERF regime, the noble-gas spins can be described much simpler because of the lack of the hyperfine structure (absence of total angular momentum of the electrons).

The consideration of the Hamiltonian given by Eq. (2.128) leads to the following equation of motion for the expectation value of the noble-gas spin

$$\frac{d\langle \hat{\mathbf{G}} \rangle}{dt} = \gamma_n (\mathbf{B} + \mathbf{b}_G) \times \langle \hat{\mathbf{G}} \rangle + \mathbf{\Omega} \times \langle \hat{\mathbf{G}} \rangle, \quad (2.130)$$

The nuclear polarisation for $G = 1/2$ has the following definition

$$\boldsymbol{\zeta} = 2\langle \hat{\mathbf{G}} \rangle. \quad (2.131)$$

Subsequently, the equation for the evolution of nuclear polarisation-driven external perturbations has the following form

$$\frac{d\boldsymbol{\zeta}}{dt} = \gamma_n (\mathbf{B} + \mathbf{b}_G) \times \boldsymbol{\zeta} + \mathbf{\Omega} \times \boldsymbol{\zeta}, \quad (2.132)$$

which is used later in this work to describe the interaction of the noble-gas spins with external perturbations of the comagnetometer. However, for the complete picture of the noble-gas spin evolution, one should also consider the relaxation of the noble gas spins and the interaction between the alkali-metal and noble-gas polarisations. Those aspects of the magnetometer evolution are discussed in subsequent sections.

2.2.2. RELAXATION OF THE NOBLE GAS SPINS

Long coherence times of the nuclear states in the noble gases are mainly the result of an efficient shielding of their nuclei by closed-shell electrons. This makes the nuclear spins largely insensitive to the spin-exchange collisions between noble-gas atoms. Therefore, in contrast to the alkali-metal atoms, spin relaxation is not limited by spin-exchange collisions. In turn, the main relaxation mechanisms may be associated with the motion of noble-gas atoms in inhomogeneous magnetic fields.

This work considers the relaxation of the noble-gas spins at high pressure.¹⁴ In such a case, when the constant magnetic field is applied along the z -axis, the longitudinal relaxation, induced by the perpendicular gradient $|\nabla B_\perp|$, is inversely proportional to the square of the applied field amplitude [117]

$$\frac{1}{T_1^n} = D_n \frac{|\nabla B_\perp|^2}{B_z^2}, \quad (2.133)$$

where D_n is the self-diffusion coefficient of the noble gas, which depends on the temperature Θ and the concentration of the noble gas N_n , and can be approximated with the following expression [117]

$$D_n = D_0 \frac{\sqrt{\Theta/273\text{ K}}}{N_n/1\text{ amg}}, \quad (2.134)$$

with $D_0^{3\text{He}} = 1.65\text{ cm}^2/\text{s}$ for ^3He .

The transverse relaxation rate is proportional to the longitudinal gradient $|\nabla B_z|$ [117]

$$\frac{1}{T_2^n} = \frac{8a^4\gamma_n^2|\nabla B_z|^2}{175D_n}, \quad (2.135)$$

where γ_n is the gyromagnetic ratio of the noble-gas nuclei and a is the radius of the cell.

The atoms placed in the vapour cell are exposed to various sources of magnetic-field gradients. The gradients may be generated by residual magnetisation of the magnetic shield and ferromagnetic impurities in elements placed inside the shield, including a glass of the vapour cell. Therefore, to avoid the gradients, the magnetic shield needs to be carefully demagnetised along with all elements of the experimental setup placed inside the magnetic shield. As it is difficult to passively eliminate all these field inhomogeneities through degaussing, magnetic-field gradient coils are often used in precise experiments. Moreover, the magnetic-field coils, used for compensation of residual fields

¹⁴Following Ref. [117], the high pressure is defined as the pressure for which the time required for the atom to diffuse across the cell is much longer than the period of the Larmor precession corresponding to the applied magnetic field.

or generation of well-controllable fields inside the shield, may also produce magnetic-field gradients.

Besides the magnetised elements, the polarised sample of atoms also generates a magnetic field. This may be an internal source of the magnetic-field gradient, which could also cause depolarisation. To avoid self-generated magnetic relaxation, the vapour cell used in the comagnetometer has a spherical shape. As the homogeneously polarised sphere generates a homogeneous magnetic field inside the sphere, the sphericity of the cell provides the disappearance of the self-interaction. For the noble gases with a nuclear spin larger than $1/2$, an additional relaxation is generated by the presence of a quadrupole moment. The quadrupole moment is sensitive to the electric field gradient generated in binary interatomic collisions [118]. For this reason, the polarisation lifetime of ^{21}Ne , possessing spin $3/2$ is usually about an order of magnitude shorter than in the case of ^3He spins [87].

Taking into account the relaxation due to both longitudinal and transverse magnetic-field gradients [Eqs. (2.133) and (2.135)], the evolution of nuclear polarisation due to the relaxation processes can be described in a following way

$$\frac{d\zeta}{dt} = -\hat{\Gamma}_n \zeta, \quad (2.136)$$

where $\hat{\Gamma}_n$ stands for the relaxation operator

$$\hat{\Gamma}_n = \begin{pmatrix} \frac{1}{T_2^n} & 0 & 0 \\ 0 & \frac{1}{T_2^n} & 0 \\ 0 & 0 & \frac{1}{T_1^n} \end{pmatrix}. \quad (2.137)$$

The presented relaxation mechanisms are the main limiting factors in the polarisation of the noble-gas spins in the comagnetometer. The polarisation level achieved in the comagnetometer results from the balance of the relaxation and efficiency of the polarisation transfer from the optically polarised alkali metal atoms via spin-exchange collisions. The latter issue is described in more detail in the next section.

2.2.3. SPIN-EXCHANGE INTERACTION BETWEEN AN ALKALI METAL AND NOBLE GAS

The key properties of the noble-gas-alkali-metal comagnetometer arise from the fact that both atomic species occupy the same volume. In such a case, the long-range interaction between the spins is enhanced by collisional (Fermi contact) interaction [119]. The interaction is the result of a spatial overlapping of wave functions of the electron in the alkali-metal atom and noble-gas nucleus. Since the spin-exchange interaction couples the electron spin directly to the nuclear spin, in this work the noble-gas-alkali-metal system is considered as the nuclear polarisation coupled to the atomic polarisation of the alkali atoms. The interaction of the alkali-metal atoms with electrons of the noble gas (e.g., van der Waals interaction) is important mostly for the precise description of the collision event, which, in this work, is treated phenomenologically and included in a form of the resulting parameters for the spin-exchange process.

The collisional interaction between alkali-metal electron spin and noble-gas nuclear spin is dominated by isotropic interaction of the following form [92]

$$V_{se} = \alpha \hat{\mathbf{S}} \cdot \hat{\mathbf{G}}, \quad (2.138)$$

where the coupling coefficient α is given by the Fermi-Sergé formula [92]

$$\alpha = \frac{8\pi g_s \mu_B \mu_G}{2G} |\psi_e(0)|^2, \quad (2.139)$$

with $|\psi_e(0)|^2$ being the probability of the alkali-metal valence electron overlap with the noble-gas nucleus and μ_G is the magnetic moment of the nuclear spin $\hat{\mathbf{G}}$.

The spin-exchange coupling defined in Eq. (2.138) preserves the total angular momentum of the colliding atoms. The effect of spin-exchange collisions on the individual spin species can be separated into two parts. The first, coherent, part does not lead to a change in the spin states of the colliding atoms but only shifts the energy levels of the interacting spins. The second, incoherent, part leads to a change of the states for both interacting spins. This part of the interaction is particularly important for the spin-exchange pumping of the noble gases since it allows the transfer of the polarisation from optically-pumped alkali-metal atoms to the nuclear spins of the noble-gas atoms. Below, one can find a review of both interactions.

COHERENT EVOLUTION

An ensemble-averaged coherent part of the spin-exchange interaction [Eq. (2.138)] leads to the precession of either of the interacting spins around the expectation value of the second spin. This acts as an effective magnetic field $\delta \mathbf{B}_{ij}$ [120]

$$\delta \mathbf{B}_{ij} = \frac{8\pi}{3} \kappa_{ij} n_j \frac{\mu_j}{S_j} \langle \hat{\mathbf{S}}_j \rangle, \quad (2.140)$$

where i denotes the perturbed and j perturbing spin species, n_j is the concentration of the perturbing species, μ_j is the magnetic moment of the j th atom, and S_j is the main quantum number of the spin $\hat{\mathbf{S}}_j$. The unitless coefficients κ_{ij} account for the collisional enhancement of the interaction between two species and are defined as the ratio between the effective magnetic field inside of the spherical cell and the classical magnetic field from the homogeneously polarised sphere with magnetisation defined by the magnetic moments correspondent to the spins. Note that the definition of the effective field, and therefore the enhancement factors, are determined by the geometry of the cell. This work is limited to the case of spherical cells.

For high-pressure noble-gas systems, which are of interest for this work, the enhancement factors for both polarisations are the same

$$\kappa_0 = \kappa_{ij} = \kappa_{ji}. \quad (2.141)$$

As shown in Table 2.4, the spin-exchange enhancement is stronger for the heavier noble gases. At the same time, the enhancement factors depend on the temperature T according to [121]

$$\kappa_0^{ij}(T) = \kappa_0^{ij}(T_{ref}) + \kappa'_0(T - T_{ref}), \quad (2.142)$$

Species	κ_0	T_{ref} [°C]	κ'_0	Reference
K- ³ He	5.99 ± 0.11	200	0.0086 ± 0.0020	[121]
Rb- ³ He	6.15 ± 0.09	175	0.00934 ± 0.00014	[122]

Table 2.4: Experimental values for the enhancement factors κ_0 . T_{ref} stands for the reference temperature at which κ_0 was measured. The provided values are relevant to the operational conditions of the comagnetometer (150-200°C).

where T_{ref} is the reference temperature (in Centigrades) at which the the enhancement factor was measured and κ'_0 is the temperature coefficient. The experimentally determined enhancement parameters, relevant for this work, are given in Table 2.4.

The maximum possible magnetisation M_j created by the perturbing spin has the following definition

$$M_j = n_j \mu_j, \quad (2.143)$$

which allows the transformation of the relation for the effective magnetic field (2.140) to

$$\delta \mathbf{B}_{ij} = \lambda M_j \frac{\langle \hat{\mathbf{S}}_j \rangle}{S_j}, \quad (2.144)$$

where $\lambda = \frac{8\pi\kappa_0}{3}$ is the coupling constant.

Taking into account Eqs. (2.108), (2.132), and (2.140), the evolution of the nuclear- and electron-spin polarisations are coupled and should be described with the set of two equations¹⁵

$$\begin{cases} \frac{d\mathbf{Y}}{dt} = \frac{\gamma_e}{q(\mathbf{Y}, t)} \lambda M_n \boldsymbol{\zeta} \times \mathbf{Y} \\ \frac{d\boldsymbol{\zeta}}{dt} = \gamma_n \lambda M_e \mathbf{Y} \times \boldsymbol{\zeta} \end{cases}. \quad (2.145)$$

From the obtained system of equations, one can note that the coupling strength is not symmetrical, as the constants in front of the vector products are different. The difference appears due to the different gyromagnetic ratios of the alkali-metal spins and the noble gas spins, as well as due to the different maximal magnetisations of those spin species. As the maximum magnetisation depends on the concentration of the atoms [Eq. (2.143)], the coupling strength between the polarisations can be adjusted via control of the concentrations of the atoms used for the operation of the comagnetometer. In particular, one can make a cell with a significantly higher concentration of the noble gas, such that $M_n \gg M_e$. This creates a situation in which the noble gas generates a large frequency shift in the alkali-metal polarisation while its own frequency remains relatively unchanged. As will be shown later in this chapter, this condition plays a key role in the achievement of the self-compensating regime.

¹⁵For the moment, it is assumed that all external fields are equal to zero and there is no relaxation in the system.

INCOHERENT EVOLUTION

The spin-exchange interaction is also responsible for the redistribution of the angular momentum between alkali-metal and noble-gas polarisations atoms. The efficiency of the spin-exchange process can be described with the spin-exchange rate coefficient [92]

$$k_{SE} = \frac{R_{ji}}{n_j}, \quad (2.146)$$

where j and i denote the perturbing and perturbed atomic species, respectively, and R_{ji} is the spin-exchange rate between two polarisations. Similarly to the case of alkali-metal polarisation, the comagnetometer system, in the presence of spin-exchange collisions, can be described by the spin-temperature distribution [92]. The exchange rates for ^3He -Rb and ^3He -K systems have the following experimental values [123, 124]

$$\begin{aligned} k_{SE}^{Rb} &= 6.8 \cdot 10^{-20} \text{ cm}^3/\text{s}, \\ k_{SE}^K &= 5.5 \cdot 10^{-20} \text{ cm}^3/\text{s}. \end{aligned} \quad (2.147)$$

It is assumed that the whole system of the comagnetometer remains in the spin-temperature at all times. Then, the incoherent spin-exchange evolution couples the expectation values for electron and nuclear spins [118]

$$\begin{cases} \frac{d\langle\hat{\mathbf{F}}\rangle}{dt} &= -R_{ne}(\epsilon(G, \beta)\langle\hat{\mathbf{S}}\rangle - \langle\hat{\mathbf{G}}\rangle) \\ \frac{d\langle\hat{\mathbf{G}}\rangle}{dt} &= R_{en}(\epsilon(G, \beta)\langle\hat{\mathbf{S}}\rangle - \langle\hat{\mathbf{G}}\rangle) \end{cases}, \quad (2.148)$$

where $\epsilon(G, \beta)$ is the paramagnetic coefficient, which, in a general case, depends on the spin-temperature parameter β . Assuming $\langle\hat{\mathbf{G}}\rangle$ being oriented along $\mathbf{z}$ [118]

$$\epsilon(G, \beta) = 2\langle G(G+1) - G_z^2 \rangle. \quad (2.149)$$

For noble gasses with the nuclear spin $G = 1/2$, the paramagnetic coefficient is constant, $\epsilon(G, \beta) = 1/2$ [118]. For $G > 1/2$, the paramagnetic coefficient depends on the polarisation of the sample and varies from $4G(G+1)/3$ at the limit of the low polarisation to $2K$ for the large polarisations [118].

This work primarily considers the comagnetometers utilising the ^3He atoms. Then, from Eq. (2.148), the polarisation transfer between two species in the comagnetometer is given by the following system of equations

$$\begin{cases} \frac{d\Upsilon}{dt} &= -\frac{R_{ne}}{q}(\Upsilon - \zeta) \\ \frac{d\zeta}{dt} &= R_{en}(\Upsilon - \zeta) \end{cases}. \quad (2.150)$$

The spin-exchange transfer of the polarisation from optically-polarised alkali-metal atoms allows for the effective pumping of the noble-gas atoms, while it can be treated as the additional source of relaxation for alkali-metal atoms. In such an approach, the spin-exchange rate R_{en} acts as the pumping rate for the noble-gas atoms. The pumping efficiency depends on the balance between the spin exchange and relaxation in the system. After taking into account the relaxation of the noble gas [Eq. (2.133)] along with the relaxation and pumping of the alkali-metal polarisation, the incoherent evolution of the

longitudinal component of the comagnetometer polarisations system from Eq. (2.150) has the following form¹⁶

$$\begin{cases} \frac{dY_z}{dt} = \frac{R_{ne}}{q} \zeta_z + \frac{R_{tot}^e}{q} (Y_0 - Y_z) \\ \frac{d\zeta_z}{dt} = R_{en} (Y_z - \zeta_z) - \frac{1}{T_1^n} \zeta_z \end{cases}, \quad (2.151)$$

where Y_0 is defined depending on the optical transition chosen for the pumping of the alkali metal polarisation by Eq. (2.74) or Eq. (2.80) and the alkali-metal relaxation rate R_{tot}^e accounts for all channel of the alkali-metal spin relaxation, including the spin-exchange with the noble-gas polarisation. This results in the following expression of the steady state of the alkali-metal polarisation (the case when all derivatives in Eq. (2.151) are equal to zero)

$$Y'_0 = Y_0 + \frac{R_{ne}}{R_{tot}^e} \zeta_0, \quad (2.152)$$

where ζ_0 is the steady-state polarisation of the noble gas. The second term on the right side of the equation is responsible for the back polarisation.¹⁷ The back polarisation process is usually neglected due to the domination of the alkali-metal relaxation over the spin-exchange relaxation rate $R_{ne}/R_{tot}^e \ll 1$, which leads to

$$Y'_0 \approx Y_0. \quad (2.153)$$

This result allows one to use the second equation in relations (2.151) for derivation of the steady-state polarisation of the noble-gas ζ_0

$$\zeta_0 = \frac{R_{en}}{R_{en} + 1/T_1^n} Y_0. \quad (2.154)$$

This shows that in the comagnetometer, the noble-gas polarisation level is ultimately determined by the equilibrium polarisation of the alkali-metal atoms. Using the analogy between conventional optical pumping alkali-metal polarisation [Eq. (2.74)] and the spin-exchange pumping of the noble gas [Eq. (2.154)], the alkali-metal polarisation plays a role of the pumping vector, while the spin-exchange rate R_{en} plays a role of the pumping rate.

2.2.4. SIMULTANEOUS EVOLUTION OF COUPLED POLARISATIONS

The discussed aspects of the polarisation evolution of the alkali-metal atomic spins and the noble-gas nuclear spins allow a comprehensive description of the comagnetometer evolution. To get the complete description of spin-system evolution in the comagnetometer, one has to combine the results for the evolution of the alkali-metal polarisation [Eq.(2.53)], the nuclear-polarisation evolution in the external fields [Eq. (2.132)], and the relaxation of the noble gas [Eq. (2.136)], the coherent [Eq. (2.145)] and incoherent

¹⁶Here it is assumed that the pumping beam propagation axis parallel to z axis, which determines the longitudinal axis.

¹⁷The back polarisation is the process in which the spin orientation (and hence angular momentum), initially transferred from alkali metal atoms to the noble gas, is transferred back from the noble gas polarisation to the alkali metal polarisation due to the spin-exchange collisions.

[Eq. (2.150)] couplings between the polarisations, which leads to the following system of equations

$$\left\{ \begin{array}{l} \frac{d\mathbf{Y}}{dt} = \frac{1}{q(\mathbf{Y}, I)} \left[\gamma_e (\mathbf{B} + \mathbf{b}_e + \lambda M_n \boldsymbol{\zeta}) \times \mathbf{Y} - R_{tot}^e (\mathbf{Y} - \mathbf{Y}_0) \right] + \gamma_I \left(1 - \frac{1}{q(\mathbf{Y}, I)} \right) \mathbf{b}_I \times \mathbf{Y} \\ \frac{d\boldsymbol{\zeta}}{dt} = \gamma_n \left(\mathbf{B} + \mathbf{b}_G + \lambda M_e \mathbf{Y} + \frac{\boldsymbol{\Omega}}{\gamma_n} \right) \times \boldsymbol{\zeta} - (\hat{\Gamma}_n + R_{en}) (\boldsymbol{\zeta} - \boldsymbol{\zeta}_0) \end{array} \right., \quad (2.155)$$

where $\mathbf{Y}_0$ is the steady state of the alkali-metal polarisation, which was defined in Eq. (2.74) or Eq. (2.80) (the definition for the equilibrium polarisation may differ depending on the D line used for the pumping), and $\boldsymbol{\zeta}_0$ is the steady state of the noble-gas polarisation, which was defined in Eq. (2.154). The obtained set of equations is also known as the **Bloch-Hasegawa equations** (BHE) [125], which are a standard tool for the description of the comagnetometer spin dynamics.

2.2.5. COMPENSATION POINT

The comagnetometer is operated in the self-compensation regime where the leading field is tuned to the compensation point B_c [57]

$$\mathbf{B}_c \equiv -\lambda M_e \mathbf{Y}_0 - \lambda M_n \boldsymbol{\zeta}_0. \quad (2.156)$$

The compensation field is predominantly determined by the noble-gas polarisation. This is due to the significant difference in the concentration of the alkali-metal atoms and noble-gas atoms. Specifically, in the K- ^3He comagnetometer, containing 3 amg of the noble gas, the helium concentration is $8 \cdot 10^{19} \text{ cm}^{-3}$, and the potassium concentration, when operated at 180°C , is $6 \cdot 10^{13} \text{ cm}^{-3}$. Under such conditions, the ratio between maximal magnetisations of the atoms [Eq. (2.143)] is

$$\frac{M_n}{M_e} = \frac{n_n \mu_{^3\text{He}}}{n_e \mu_e} \approx 1.5 \times 10^3. \quad (2.157)$$

Thereby, even if the nuclear polarisation is an order of magnitude lower than the electron polarisation (which is a typical situation in the comagnetometer), the effect of the nuclear polarisation on the electron polarisation remains dominant. In the considered example, assuming $Y_0 = 0.5$ and $\zeta = 0.03$, the effective magnetic fields generated by the polarised atoms are

$$\begin{aligned} \lambda M_e Y_0 &\approx -2.7 \text{ nT}, \\ \lambda M_n \zeta_0 &\approx -250 \text{ nT}. \end{aligned} \quad (2.158)$$

The comagnetometers consistently achieve a remarkable sensitivity of less than 1 fT (in pseudomagnetic units) for non-magnetic spin couplings. [see Eq. (2.129)]. From Eqs. (2.155) and (2.156), one notes that in the compensation point, each polarisation is exposed to the magnetic field equal to the effective magnetic field created by itself [Eq. (2.158)]. These longitudinal fields are much larger than the exotic perturbations targeted for the measurements. Therefore, the system can be considered under a small-angle approximation, where the longitudinal polarisations are assumed to be constant

and equal to their equilibrium values

$$\begin{aligned} Y_z &\approx Y_0 = \text{const}, \\ \zeta_z &\approx \zeta_0 = \text{const}. \end{aligned} \quad (2.159)$$

This also allows the approximation of the slowing-down factor with the constant value, corresponding to the equilibrium polarisation

$$q(Y, I) \approx q(Y_0, I) = q_0 = \text{const}. \quad (2.160)$$

Within the small-angle approximation, it is convenient to separate the equations of the polarisation evolution into components transverse and longitudinal to the pump-beam propagation direction. Such an approach requires the separation of all vectors in Eq. (2.155)

$$\mathbf{v} = v_z \mathbf{z} + \text{Im}[v_\perp] \mathbf{y} + \text{Re}[v_\perp] \mathbf{x}, \quad (2.161)$$

where $\mathbf{v}$ stands to any vector quantity and $v_\perp$ is the transverse component of the vector in the complex form

$$v_\perp = v_x + i v_y. \quad (2.162)$$

In such a case, Eq. (2.155) for the transverse components of the polarisations take the following form

$$\begin{cases} \frac{dY_\perp}{dt} = -i\gamma_F \left[(B_\perp + b_{e\perp} + \lambda M_n \zeta_\perp) Y_z - (\Delta_{B_z} + \lambda M_e Y_z) Y_\perp \right] - R_e Y_\perp - i\gamma_F (q_0 - 1) b_{I\perp} \times Y_z, \\ \frac{d\zeta_\perp}{dt} = -i\gamma_n \left[(B_\perp + b_{G\perp} + \lambda M_e Y_\perp - \frac{\Omega_\perp}{\gamma_n}) \zeta_z - (\Delta_{B_z} + \lambda M_n \zeta_z) \zeta_\perp \right] - R_n \zeta_\perp \end{cases}, \quad (2.163)$$

where the non-magnetic perturbations along the z -axis are dropped out as they are much smaller than the leading field. To formulate the equations, the detuning from the compensation point was introduced

$$\Delta_{B_z} = B_c - B_z. \quad (2.164)$$

The relaxation rates for the transverse polarisations and gyromagnetic ratio of the electron polarisation are defined in the following way

$$\bar{R}_e = \frac{R_{tot}^e}{q_0}, \quad (2.165)$$

$$R_n = \frac{1}{T_2} + R_{en}, \quad (2.166)$$

$$\gamma_F = \frac{\gamma_e}{q_0}, \quad (2.167)$$

such that Eq. (2.163) obtains more simplistic form.

2.2.6. NON-MAGNETIC PERTURBATION OF PROTONS AND NEUTRONS

Heretofore, in the discussion of the comagnetometer polarisation dynamics the non-magnetic nuclear couplings to alkali-metal $\mathbf{b}_I$ and noble-gas $\mathbf{b}_G$ spins were considered independently. However, in the context of the search for the exotic spin couplings, it is

Species	σ_n	σ_p
^3He	0.87	-0.03
^{39}K	0	-0.2
^{87}Rb	0	0.31

Table 2.5: Fractional impact parameter [Eq. (2.169)] of the nucleon spin in the total nuclear spins of the atoms considered in this work. The presented values follow Ref. [126].

important to specify couplings for the nuclear spin of noble-gas and alkali-metal atoms [126]

$$b_{i\perp} = \sum_{j=n,p} \sigma_{ji} b_{j\perp}, \quad (2.168)$$

where i denotes either noble-gas or alkali-metal nuclear spin, and $\sigma_{n,p\ i}$ is the so-called fractional impact parameter for the nucleon spin in the considered nuclear spin [126]

$$\frac{\langle \mathbf{S}_{n,p} \rangle}{\langle \mathbf{S}_i \rangle} = \sigma_{n,p\ i}. \quad (2.169)$$

The values for σ_{ij} , relevant to this work, are collected in Table 2.5.

2.2.7. GENERIC SPIN PERTURBATION

For further discussion of the system response to various perturbations, it is convenient to introduce a generic perturbation $\mathbf{b}_\perp$, which is measured in effective magnetic-field units and couples to the electron and nuclear polarisations with the respective strengths α_e and α_n . The relation between $\mathbf{b}_\perp$ and several known and postulated spin interactions, along with the related coupling constants, are presented in Table 2.6. This allows one to introduce a more compact form of BHEs, without losing the generality of the discussion

$$\begin{cases} \frac{dY_\perp}{dt} &= -i\gamma_F \left[(\alpha_e \mathbf{b}_\perp + \lambda M_n \zeta_\perp) Y_z - (\Delta_{B_z} - \lambda M_e Y_z) Y_\perp \right] - R_e Y_\perp \\ \frac{d\zeta_\perp}{dt} &= -i\gamma_n \left[(\alpha_n \mathbf{b}_\perp + \lambda M_e Y_\perp) \zeta_z - (\Delta_{B_z} - \lambda M_n \zeta_z) \zeta_\perp \right] - R_n \zeta_\perp \end{cases}. \quad (2.170)$$

Such a form of BHE allows one to obtain the general solution, which then can be interpreted for various types of coupling by simple substitution of the generic perturbation and coupling constants by appropriate expression from Table 2.6.

2.2.8. RESPONSE TO ROTATING FIELDS

For effective GNOME searches, it is important to understand the response of the comagnetometer over the broad band of frequencies. With this in mind, the frequency response of the comagnetometer is studied in the following subsections.

The discussion starts with the response of the comagnetometer to a generic rotating field with frequency $\pm\omega$ and amplitude b_0

$$\mathbf{b}_\perp(\pm\omega) = b_0 e^{\pm i\omega t}. \quad (2.171)$$

Perturbation type	$\mathbf{b}_\perp$	α_e	α_G
Magnetic field	$B_\perp$	1	1
Electron perturbation	$\frac{\hbar \gamma_e}{\gamma_n} \Xi_\perp$	1	0
Proton perturbation	$\frac{\hbar \sigma_{pG} \chi_p}{\gamma_n} \Xi_\perp$	$(q_0 - 1) \frac{\gamma_n}{\gamma_e} \frac{\sigma_{pl}}{\sigma_{pG}}$	1
Neutron perturbation	$\frac{\hbar \sigma_{nG} \chi_n}{\gamma_n} \Xi_\perp$	$(q_0 - 1) \frac{\gamma_n}{\gamma_e} \frac{\sigma_{nl}}{\sigma_{nG}}$	1
Rotation	$\Omega_\perp / \gamma_n$	$\frac{\gamma_n}{\gamma_F}$	1

Table 2.6: Coupling strengths of various known and postulated spin interactions.

In this case, the dynamic steady-state solution of Eq. (2.170) takes the form

$$\begin{aligned} Y_\perp^\pm(t, \omega) &= \mathcal{F}_{Y_\perp^\pm}(\omega) \mathbf{b}_0 e^{\pm \omega t}, \\ \zeta_\perp^\pm(t, \omega) &= \mathcal{F}_{\zeta_\perp^\pm}(\omega) \mathbf{b}_0 e^{\pm \omega t}, \end{aligned} \quad (2.172)$$

where the frequency responses are

$$\mathcal{F}_{Y_\perp^\pm}(\omega) = -\gamma_F Y_z \frac{\omega_n(\alpha_e - \alpha_n) + (\pm\omega + \gamma_n \Delta_{B_z} - iR_n)\alpha_e}{(\pm\omega + \omega_e + \gamma_F \Delta_{B_z} - iR_e)(\pm\omega + \omega_n + \gamma_n \Delta_{B_z} - iR_n) - \omega_e \omega_n}, \quad (2.173)$$

$$\mathcal{F}_{\zeta_\perp^\pm}(\omega) = -\gamma_n \zeta_z \frac{\omega_e(\alpha_n - \alpha_e) + (\pm\omega + \gamma_F \Delta_{B_z} - iR_e)\alpha_n}{(\pm\omega + \omega_e + \gamma_F \Delta_{B_z} - iR_e)(\pm\omega + \omega_n + \gamma_n \Delta_{B_z} - iR_n) - \omega_e \omega_n}, \quad (2.174)$$

where the Larmor frequencies of the electron and nucleus in the compensation point are introduced according to the following definition

$$\begin{aligned} \omega_e &= \gamma_F \lambda M_e Y_z, \\ \omega_n &= \gamma_n \lambda M_n \zeta_z. \end{aligned} \quad (2.175)$$

Although, in general, BHE lead to a nonlinear response of the system, under the small angles approximation [Eq. (2.159)], the response is linear. This linearity provides a practical measure of the regime where the response of the system can be predicted with BHE under the small-angles approximation [Eq.(2.170)], when the presented analytical results are valid.

The denominator of the frequency response of the comagnetometer¹⁸ [Eq. (2.173)] is presented by the second-order polynomial in ω . Hence, this system can be interpreted as a combination of the second-order filters with different attenuation for the various types of perturbations. As the numerator of the frequency response presented in Eq. (2.173) is the first order polynomial in ω , such a combination would consist of two filters

$$\mathcal{F}_{Y_\perp^\pm}(\omega) = \mathcal{F}_{LP}(\pm\omega) + \mathcal{F}_{BP}(\pm\omega), \quad (2.176)$$

¹⁸Since now on, whenever there is a reference to the response of the comagnetometer, it refers to the evolution of the alkali-metal polarisation $Y_\perp$.

where the general expressions for the frequency response of the second-order low-pass filter $\mathcal{F}_{LP}$ and band-pass filter $\mathcal{F}_{BP}$ have the following form [127]

$$\mathcal{F}_{LP}(\omega) = \frac{A_{LP}}{\omega^2 + B_{LP}\omega + C_{LP}}, \quad (2.177)$$

$$\mathcal{F}_{BP}(\omega) = \frac{A_{BP}\omega}{\omega^2 + B_{BP}\omega + C_{BP}}. \quad (2.178)$$

The filter-like parameters for the comagnetometer can be derived from Eq. (2.173)

$$A_{LP} \equiv \gamma_F \Upsilon_z [\omega_n (\alpha_n - \alpha_e) + (\gamma_n \Delta_{B_z} + i R_n) \alpha_e], \quad (2.179)$$

$$A_{BP} \equiv \gamma_F \Upsilon_z \alpha_e, \quad (2.180)$$

$$B_{LP} = B_{BP} \equiv \omega_n + \omega_e - (\gamma_n + \gamma_F) \Delta_{B_z} - i(R_n + R_e), \quad (2.181)$$

$$C_{LP} = C_{BP} \equiv \gamma_n \gamma_F \Delta_{B_z}^2 - [\gamma_F (\omega_n - i R_n) + \gamma_n (\omega_e - i R_e)] \Delta_{B_z} - (R_e R_n + i R_e \omega_n + i R_n \omega_e). \quad (2.182)$$

The analogy between the comagnetometer response and the response of the low- and band-pass filters provides a convenient and intuitive model for the understanding and prediction of the comagnetometer response to various perturbations. This analogy is illustrated with the results of the frequency-response simulation plotted for the representative experimental conditions at the compensation point (Fig. 2.9). Notably, from Eq. (2.173), the response of the comagnetometer to purely nuclear perturbation ($\alpha_n \neq 0$, $\alpha_e = 0$) is determined by the low-pass-like frequency response, while for the magnetic field ($\alpha_n = \alpha_e \neq 0$) the response is determined by the band-pass-like frequency response, which has straight-forward correspondence to the presented plots. As for purely electron perturbations ($\alpha_e \neq 0$, $\alpha_n = 0$), the predicted response is given by the combination of both low- and band-pass filter responses. In the case of the response to the perturbation with the positive frequency, $\mathcal{F}_\perp^+$, such a combination leads to the significant suppression of the comagnetometer response around $\omega = \omega_n$, which is the result of the phase difference between low- and band-pass components of the response. This can be explained by the opposite sign in front of α_e in Eq. (2.179) and Eq. (2.180).

The filter-like analogy also provides a simple explanation for the highest suppression of the magnetic-field response at the compensation point. This originates from the fact that the detuning from the compensation point adds additional low-pass contribution to the band-pass component of the response [Eq. (2.173)].

2.2.9. RESPONSE TO DC PERTURBATION

The DC response of the comagnetometer can be considered by putting $\omega = 0$ into Eq. (2.176). For this, one obtains

$$\mathcal{F}_{Y_\perp^\pm}(0) = \frac{A_{LP}}{C_{LP}} = \frac{\gamma_F \Upsilon_z [\omega_n (\alpha_n - \alpha_e) + (\gamma_n \Delta_{B_z} + i R_n) \alpha_e]}{\gamma_n \gamma_F \Delta_{B_z}^2 - [\gamma_F (\omega_n - i R_n) + \gamma_n (\omega_e - i R_e)] \Delta_{B_z} - (R_e R_n + i R_e \omega_n + i R_n \omega_e)}. \quad (2.183)$$

At the compensation point and for the measurement time much shorter than the relaxation time of the nuclear polarisation, the system response is given by

$$\mathcal{F}_{Y_\perp^\pm}(0) \xrightarrow[R_n \approx 0]{\Delta_{B_z} = 0} -i \frac{\gamma_F \Upsilon_z}{R_e} (\alpha_n - \alpha_e), \quad (2.184)$$

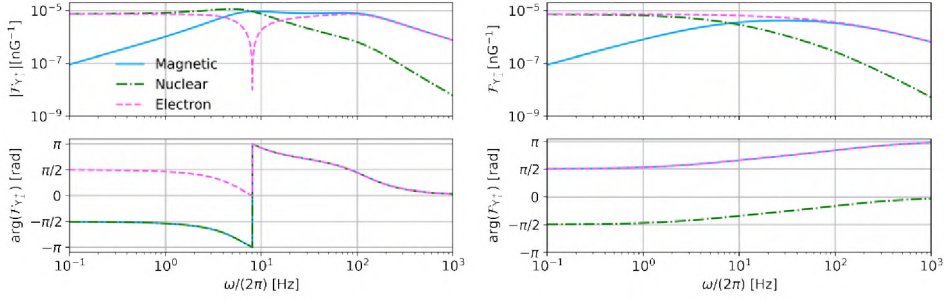


Figure 2.9: Analytical solutions of the comagnetometer frequency response to the rotating perturbations [Eq. (2.173)]. The left column shows the solution for $\mathcal{P}_\perp^+$, and the right column presents the solution for $\mathcal{P}_\perp^-$. The complete list of parameters used for the simulations is given in Appendix A.1. The obtained magnetic field response is similar to the response of the band-pass filter in both presented cases. Such characteristics arise from the low-frequency response being suppressed by the self-compensating mechanism and the limited bandwidth of the alkali-metal response at high frequencies. This is in contrast to the response to non-magnetic perturbations, where the absence of the self-compensating mechanism at low frequencies results in a frequency response similar to the low-pass filter.

which means that the system is sensitive only to perturbations with $\alpha_n \neq \alpha_e$. This is in agreement with the results of Ref. [57]. In practice, it means that in such a regime, the system is insensitive to the external magnetic fields while remaining sensitive to the non-magnetic perturbations. It is also worth noticing the similarity between the DC response of the SERF magnetometer [Eq. (2.111)] and the comagnetometer [Eq. (2.184)]. This similarity shows that for the low-frequency perturbations and at the compensation point, the comagnetometer acts as the SERF magnetometer, which is shielded from the magnetic-field perturbations by the noble-gas polarisation, and it senses the difference between the perturbations of the noble-gas and alkali-metal polarisation, which is proportional to $(\alpha_n - \alpha_e)$.

The finite relaxation rate of the noble polarisation, $R_n \neq 0$, leads to the non-zero response of the system to the low-frequency magnetic field, as pointed out in Ref. [128]. Hence, the finite nuclear-polarisation relaxation rate limits the suppression of the magnetic fields in the compensation point and may lead to deterioration of the sensitivity to the low-frequency non-magnetic couplings. This effect may be more prominent in the comagnetometers, which are based on ^{21}Ne due higher relaxation rate of the nuclear polarisation. However, in practice, this effect remains rather small since the noble-gas relaxation rate is still much lower than any other parameter of the system.

It is clear from Eq.(2.183) that the sensitivity of the system strongly depends on the detuning from the compensation point Δ_{B_z} . The detuning suppresses the response to the non-magnetic perturbations but also as increases the sensitivity to the magnetic field. For this reason, it is crucial to keep the Δ_{B_z} as low as possible over the whole time of the measurement.

2.2.10. RESPONSE TO THE PERIODIC PERTURBATIONS

This section analyses the response of the comagnetometer to the periodic perturbations applied along the y -axis

$$b_{\perp} = i b_0 \sin(\omega t) = \frac{b_0}{2} (e^{i\omega t} - e^{-i\omega t}), \quad (2.185)$$

where it was assumed that b_0 is real.

The periodic perturbation [Eq. (2.185)] can be considered as the superposition of two counter-rotating fields also considered in Eq. (2.171). Since BHE [Eq. (2.170)] is the first-order ordinary equations, the response of the comagnetometer to the periodic (harmonic) perturbations $\Upsilon_{\perp}^h$ can be expressed in terms of the response to the rotating perturbations

$$\Upsilon_{\perp}^h = \frac{1}{2} (\Upsilon_{\perp}^+ e^{i\omega t} - \Upsilon_{\perp}^- e^{-i\omega t}). \quad (2.186)$$

Then, from Eq. (2.162), the polarisation components along $\mathbf{x}$ and $\mathbf{y}$ can be expressed in a following way

$$\Upsilon_x^h = \text{Re}[\Upsilon_{\perp}^h] = \text{Re}[\Upsilon_{\perp}^+ - \Upsilon_{\perp}^-] \cos(\omega t) - \text{Im}[\Upsilon_{\perp}^- + \Upsilon_{\perp}^+] \sin(\omega t) = \Upsilon_x^c \cos(\omega t) + \Upsilon_x^s \sin(\omega t), \quad (2.187)$$

$$\Upsilon_y^h = \text{Im}[\Upsilon_{\perp}^h] = \text{Im}[\Upsilon_{\perp}^+ - \Upsilon_{\perp}^-] \cos(\omega t) + \text{Re}[\Upsilon_{\perp}^+ + \Upsilon_{\perp}^-] \sin(\omega t) = \Upsilon_y^c \cos(\omega t) + \Upsilon_y^s \sin(\omega t), \quad (2.188)$$

where Υ_i^c and Υ_i^s are the out- and in-phase components of the response, respectively. Alternatively, these results can also be presented in terms of the amplitude $|\Upsilon_i^h(\omega)|$ and phase $\arg[\Upsilon_i^h(\omega)]$ of the comagnetometer response

$$\Upsilon_i^h = |\Upsilon_i^h(\omega)| \sin(\omega t + \arg[\Upsilon_i^h(\omega)]). \quad (2.189)$$

The amplitude and phase responses then can be defined through the complex polarisation response [Eq. (2.187) or Eq. (2.188)]

$$|\Upsilon_i^h(\omega)| = \sqrt{(\Upsilon_i^c)^2 + (\Upsilon_i^s)^2}, \quad (2.190)$$

$$\arg[\Upsilon_i^h(\omega)] = \arccos\left(\frac{\Upsilon_i^c}{|\Upsilon_i^h(\omega)|}\right). \quad (2.191)$$

Although the connection between previously considered frequency responses to the rotating perturbation is not trivial, the qualitative behaviour of the comagnetometer can still be predicted based on the filter-like model. Figure 2.10 presents the calculated frequency responses to the harmonic perturbations applied along the y -axis, plotted for the same parameters as for the rotating perturbation in Fig. 2.9. In both cases, the response to the non-magnetic nuclear perturbation is determined by the low-pass filter functions and the response to the periodic non-magnetic nuclear perturbations reveals the same low-pass filter character. The same is true for the magnetic-field response, i.e., the response character remains that of a band-pass filter. The response to purely electron perturbation is given by the combination of both low- and band-pass filters.

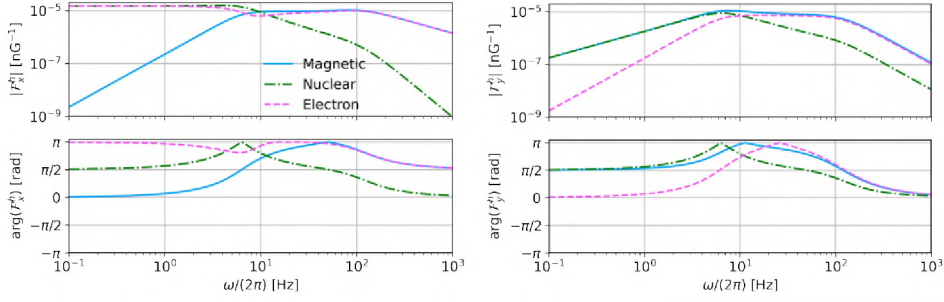


Figure 2.10: Analytical solutions for the frequency response of the comagnetometer at the compensation point to the harmonic perturbation applied along the y -axis. The left column shows $\mathcal{F}_x^h$ and right $\mathcal{F}_y^h$. The parameters used for the plots are the same as in Fig. 2.9, and they are presented in Appendix A.1.

2.2.11. DIRECTIONAL SENSITIVITY OF THE COMAGNETOMETER

Application of the comagnetometer for searches of the exotic-spin couplings generated by astrophysical sources requires a good understanding of the directional sensitivity of the sensor. In particular, this is important in determination of expected-signal patterns, generated in the model under consideration.

In general, there are two main directional characteristics of the comagnetometer. The first is the ability to detect the perturbation from various directions, while the second is the ability to identify the direction of the detected perturbation. The results of the previous section [Eq. (2.190) and Fig. 2.10] can be used to characterise the considered comagnetometer in these categories.

This section considers the comagnetometer exposed to the generic perpendicular perturbation $\mathbf{b}_\perp$. Such perturbation can be expressed through its Cartesian components according to Eq. (2.162)

$$\mathbf{b}_\perp = \mathbf{b}_x + i\mathbf{b}_y. \quad (2.192)$$

Equation (2.190) provides the solution for the response of the system to the generic field applied along $\mathbf{y}$. The rotating symmetry of the system (the leading field is colinear with the pump beam, and therefore initial polarisation) allows the derivation of the result for the perturbation along $\mathbf{x}$ (along the probe beam) swapping the indices for x - and y -axes and taking into account the complex phase change related to such a change.¹⁹

From the frequency response presented in Fig. 2.10, one can note that for the low-frequency non-magnetic perturbations, the system response to the perturbations perpendicular to the probe beam is by multiple orders of magnitude stronger than to the field along the probe beam. This indicates the possibility of only detecting the perturbations along the single axis. The difference in the response decreases with the frequency and equalises when $\omega \approx \omega_n$. For the frequencies above ω_n , the responses for both axes have approximately the same amplitude. Such a situation creates two distinct regimes in terms of directional sensitivity for low- ($\omega \ll \omega_n$) and high- ($\omega \gtrsim \omega_n$) frequency perturbations. In this case, the comagnetometer is sensitive to any perturbation in the xy -plane.

¹⁹The swapping of $\mathbf{x}$ - and $\mathbf{y}$ -axes is equal to rotation in the complex plane, hence the complex phase change by $\pi/2$ must be taken into account.

Therefore, instead of the sensitivity axis, it is more appropriate to consider the sensitivity plane, which is perpendicular to the pump beam. In a high-frequency regime, the comagnetometer is sensitive to the projection of the perturbation on the sensitivity plane, and the determination of the perturbation direction based on the amplitude of the response is not possible anymore. However, the difference in the phase response to the perturbations applied along the perpendicular axes may potentially provide a way for a more precise determination of the perturbation direction.

The frequency response to the magnetic field (Fig. 2.10) in the low-frequency regime is suppressed due to the self-compensation, with the much stronger suppression in the direction perpendicular to the probe beam.²⁰ However, in the high-frequency regime, the response to the magnetic field along both axes has the same amplitude, which leads to sensitivity in the xy -plane. Such ambiguity in the comagnetometer response to the perturbations applied along the x - and y -axes may have particular importance in searches where the directional sensitivity plays an important role in the determination of the expected signal pattern. These are effects modulated by the Earth rotation [129–131] or the axion domain walls [39, 46, 61], where the expected perturbation has a certain propagation direction.

2.2.12. RESPONSE TO THE TRANSIENTS

The detection of transient spin perturbations is one of the crucial scientific targets for the GNOME project. In the context of the searches with the comagnetometers, the main interest is in the transients with the substantial part of the perturbation spectrum laying within the bandwidth of the self-compensation.

For the current discussion, one can consider the Lorentzian-shaped perturbation applied along the y -axis

$$b^p(t) = i b_0 \mathcal{L}_E(t), \quad (2.193)$$

where $\mathcal{L}_E(t)$ is the Lorentzian curve

$$\mathcal{L}_E(t) = \sqrt{\tau} \frac{\tau}{\tau^2 + (t - t_0)^2}, \quad (2.194)$$

where t_0 is the time of the pulse centre and τ is the pulse temporal width (HWHM). With such a pulse, the total energy of the pulse, corresponding to the square of $\mathcal{L}_E(t)$, remains unchanged with modification of the pulse width.

Since the comagnetometer is considered in the linear regime [Eq. (2.170)], the spectral response to the transient perturbation can be obtained with the frequency response of the comagnetometer [Eq. (2.189)] and the Fourier transform of the pulse $b^p(\omega)$

$$Y_x^p(\omega) = \sqrt{2\pi} b^p(\omega) Y_x^h(\omega) = \sqrt{2\pi} b_0 \mathcal{L}_E(\omega) Y_x^h(\omega), \quad (2.195)$$

where the Fourier transform of the Lorentzian is

$$\mathcal{L}_E(\omega) = \mathcal{F}[\mathcal{L}_E(t)] = \sqrt{\frac{\pi\tau}{2}} e^{-i t_0 \omega - \tau |\omega|/2}. \quad (2.196)$$

²⁰This is a result of the finite relaxation rate, which adds the low-pass filter-like component to the comagnetometer frequency response [Eq. (2.177)].

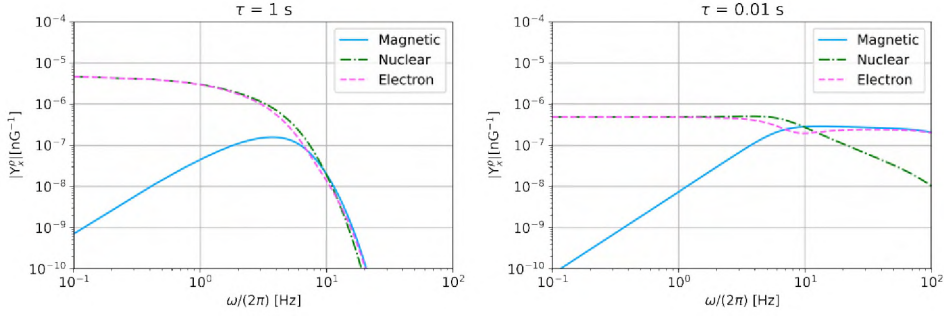


Figure 2.11: Simulated amplitude response simulated [Eq. (2.197)] to the 1-s-long (**left**) and 0.01-s-long (**right**) Lorentzian pulse [Eq. (2.193)]. The complete set of parameters used for the comagnetometer simulations is presented in Appendix A.1.

Because the time shift of the pulse leads to an additional phase in the response, but it does not change its amplitude, without loss of generality, in further considerations, it is assumed that $t_0 = 0$. Then, from the last two equations, one gets the explicit expression for the spectrum of the response to the Lorentzian pulse

$$Y_x^p(\omega) = b^p(\omega) Y_x^h(\omega) = b_0 \pi \sqrt{\tau} Y_x^h(\omega) e^{-\tau|\omega|/2}. \quad (2.197)$$

Most of the data-analysis methods to search for transient effects exploit the **access power analysis**, which detect potentially valuable signals by tracking the moments when the response signal energy,

$$\mathcal{E}_S = \int |Y_x^p(\omega)|^2 d\omega = \int |Y_x^p(t)|^2 dt, \quad (2.198)$$

surpasses a certain threshold. For the considered transients [Eq. (2.194)], the duration of the pulse τ determines its spectral width.²¹ Therefore, for the long pulses when $\tau^{-1} \leq \omega_n$, the vast majority of the perturbation energy lies within the self-compensation bandwidth, and therefore, the magnetic-field response is significantly suppressed for the non-magnetic perturbation. This is in contrast to the non-magnetic response, where the low-frequency components are not suppressed [see Fig. 2.11 (left)]. For shorter pulses, i.e. for the perturbation with a wider spectrum, the unsuppressed high-frequency components of the response spectrum dominate, hence, there is no such a difference in the amplitude response [see Fig. 2.11 (right)].

The relation between the total energy of the response [Eq. (2.198)] and the pulse width τ is plotted in Fig. 2.12 (left). The plot reveals again two regimes of comagnetometer operation, i.e., the low- and high-frequency regimes, determined by the noble-gas Larmor frequency. For the low-frequency perturbations (long pulses), the energy of the response to magnetic pulses and, therefore, its visibility is suppressed, which is not the case for all considered non-magnetic perturbations. Hence, the search for the outlying energy events in this case makes most probable the detection of the non-magnetic perturbations. However, the situation changes for the high-frequency perturbations (short

²¹The response decreases e -times for frequency $\omega = \tau^{-1}$.

pulses). In this case, the energy of the response to magnetic and non-magnetic pulses is generally the same for electron coupling, and for the nuclear couplings, the energy starts to be weaker due to the limited bandwidth of the nuclear polarisation response. This change appears due to the dominance of high-frequency components in the perturbation spectrum, where self-compensation is not observed. Hence, for the short pulses, it is much more challenging to unambiguously confirm the origin of the detected event, which may lead to a much higher rate of false positives in the comagnetometer output.

The difference between the comagnetometer response to the non-magnetic and magnetic perturbations also provides another benchmark for the identification of the non-magnetic perturbations. This is possible with the application of time-averaging of the signal

$$\langle Y_x^p(t) \rangle_t = \frac{1}{\Delta_T} \int_{t-\Delta_T}^t Y_x^p(t') dt', \quad (2.199)$$

where Δ_T is the time window used for averaging. Here, the previous assumption of $t_0 = 0$ is kept for the pulse perturbation. Then, one can put $Y_x^p(t \ll 0) \approx 0$ and assume that the width of the averaging window is much wider than the width of the pulse ($\Delta_T \ll \tau$), which allows extending the integration limit to minus infinity

$$\langle Y_x^p(t) \rangle_t = \frac{1}{\Delta_T} \int_{-\infty}^t Y_x^p(t') dt'. \quad (2.200)$$

Such a form of the time-averaged signal allows one to find the analytic expression for its spectrum

$$\mathcal{F}[\langle Y_x^p(t) \rangle_t] = \frac{Y_x^p(\omega)}{i\omega} + \pi Y_x^p(\omega) \delta(\omega), \quad (2.201)$$

where $\delta(\omega)$ is the Dirac delta function. This expression allows calculating the spectrum of the averaged response using the result of Eq. (2.197). Then, as a measure of the detectability of the pulses, one can calculate the energy of the averaged signal

$$\mathcal{E}_{\langle S \rangle_t} = \int |\langle Y_x^p(\omega) \rangle_t|^2 d\omega. \quad (2.202)$$

The results of the simulations of the energy and the averaged signal are presented in Fig. 2.12 (right). Qualitatively, the results of the time-averaged response to magnetic and non-magnetic perturbations follow the same pattern as the results for the raw comagnetometer response, namely, the magnetic response to the long pulses is sufficiently suppressed compared to both non-magnetic response and high-frequency magnetic response. However, in this case, the magnetic response is much weaker compared to the non-magnetic over the whole range of the pulse widths. This effect appears due to the averaging of the high-frequency components from the signal as a result of the time averaging, which leads to the energy of the averaged signal being determined by the low-frequency components of the response. This results in much better detectability of the non-magnetic transients compared to the transients of the magnetic origin, which leads to a much lower rate of false positives in the comagnetometer response.

The implementation of the averaging in the GNOME data analysis pipeline requires a more sophisticated treatment of the expected signals, with particular attention to its

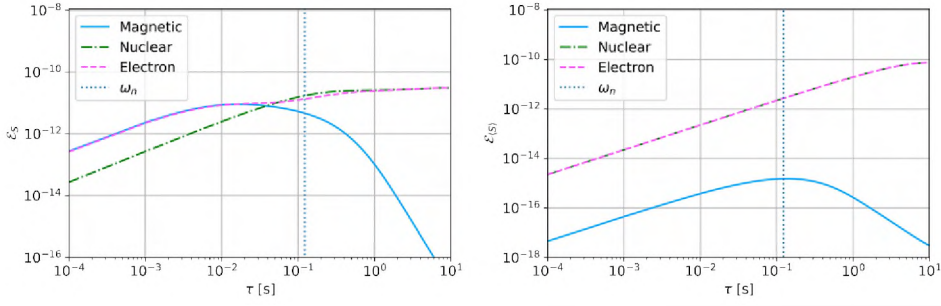


Figure 2.12: The parameters of the comagnetometer response [Eq. (2.197)] to the Lorentzian pulses [Eq. (2.193)] with different pulse duration τ . **Left:** the total energy of the response [Eq. (2.198)]. **Right:** the total energy of the response average [Eq. (2.202)].

spectral distribution. In practice, the gain of the pulse detectability is determined by the parameters of the system, defining the efficiency of the self-compensation²² and by the effects of the averaging on the noise of the comagnetometer. These two parameters, as well as the width of the targeted pulses, will determine the optimal width of the averaging window. Although the results of this section are provided for the Lorentzian pulses, they can be easily extended to the case of any “non-oscillating” transient perturbation, whose spectrum is centred at zero frequency.

2.2.13. HYBRID OPTICAL PUMPING

Over the course of the discussion presented in this chapter, the self-compensating comagnetometer was considered as a coupled system of polarised alkali-metal and noble-gas atoms. However, in practice (and later in this work), the self-compensating comagnetometer frequently employs two alkali-metal species instead of one. A mixture of two different alkali metals is used for the implementation of so-called **hybrid optical pumping**. This is a process in which one of the alkali metals (the donor) is optically pumped with light. The second alkali metal (the receiver) then obtains the angular momentum from the donor via spin-exchange collisions (Fig. 2.13). The spin-exchange cross-section between the alkali metals of different species is comparable to the cross-section for the spin-exchange between the atoms of the same species and is on the order of $\sigma_{se} \sim 10^{-14} \text{ cm}^2$. Such a high spin-exchange rate leads to a strong coupling between alkali-metal polarisations, which results in its coherent evolution in low magnetic fields [132].

Hybrid optical pumping is used to improve the homogeneity of the alkali-metal polarisation, which is crucial to achieve the identical compensation-point condition across the whole cell. In the direct pumping scheme, the inhomogeneity of the polarisation is primarily caused by the strong absorption of pumping light in the dense vapour. For hybrid pumping, the concentration of the donor is kept at a much lower level than the concentration of the receiver, which results in a much lower optical density of the former.

²²In this regard, the magnetisation of the noble gas is particularly important since it extends the frequency range where the self-compensation is observed and enhances the overall suppression of the magnetic response.

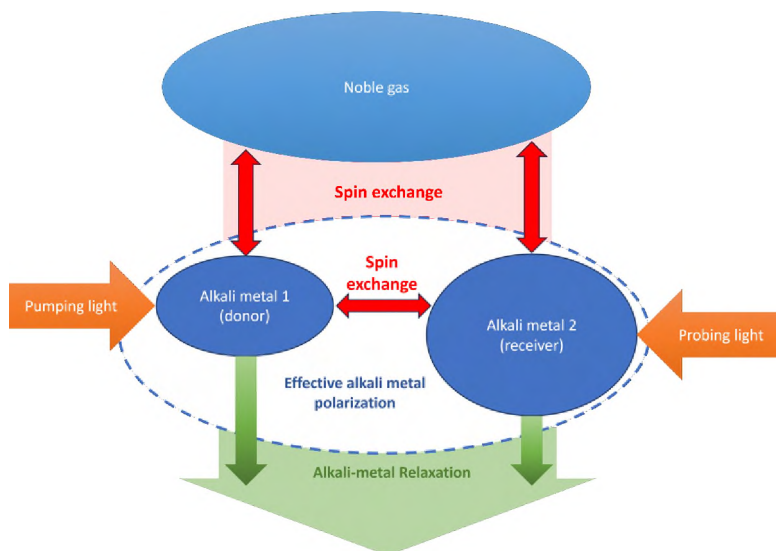


Figure 2.13: Scheme demonstrating the main processes related to the angular-momentum transfer arising in the hybrid optical pumping. The angular momentum is added to the system through optical pumping of the lower-density alkali metal (the donor). The donor polarisation is transferred to the alkali metal of a higher density (the receiver). Both the donor and receiver are used for spin-exchange pumping of the noble gas and are subjected to relaxation through spin-distraction collisions. The fast spin exchange between the donor and receiver allows consideration of both alkali metals as hybridised alkali-metal system, described by the effective parameters determined by the underlining microscopic dynamics of alkali metals. The state of the hybridised alkali-metal system is detected with the probe light resonant with the receiver.

Thereby, much smaller polarisation gradients can be achieved with optical pumping of donor, which is next transferred to the receiver, leading to homogeneous spin polarisation of this much denser species.

The strong coupling between the alkali-metal polarisations leads to hybridisation of its common evolution, i.e., they have the same Larmor frequencies and the relaxation rates [81, 132]. The limit of the fast spin exchange allows one to consider the two-alkali-metal system as an effective hybridised alkali-metal polarisation whose parameters are determined by the common evolution of both polarisations. The comagnetometer with such a hybrid system is described with the same set of the coupled equations as discussed earlier in this work [see Eq. (2.155)]. However, in this case, the hybridised polarisation of the alkali-metal has modified values of the pumping and spin-destruction rates [81]

$$\tilde{R}_p = \frac{R_p}{2q_d}(1 + \alpha), \quad (2.203)$$

$$\tilde{R}_{sd}^e = \frac{R_{sd}^r}{2q_r}(1 - \alpha) + \frac{R_{sd}^d}{2q_d}(1 + \alpha), \quad (2.204)$$

where $q_{d,r}$ are the slowing down factors of the donor and receiver, $R_{sd}^{d,r}$ are the spin-destruction rates of the donor and receiver, and the coupling parameter α is defined in a following way

$$\alpha \equiv \frac{q_d n_d - q_r n_r}{q_d n_d + q_r n_r}, \quad (2.205)$$

with $n_{d,r}$ being the concentrations of the donor and receiver. The state of the hybridised alkali-metal system is typically probed with the light resonant with the acceptor atoms, but detuned from the center of their absorption lines.

The scope of this work does not include the microscopic dynamics of the alkali-metal polarisations, and it considers only their effective polarisation. In such an approach, the spin-exchange pumping of the noble-gas nuclear spins is also considered in terms of the overall effects that both alkali-metal species impose on the nuclear polarisation of the noble gas. The schematic summary of the spin-exchange and relaxation mechanisms presented in the system with hybrid optical pumping are presented in Fig. 2.13.

2.2.14. PARAMETRIC MODULATION OF THE ALKALI-METAL POLARISATION

The sensitivity of the comagnetometer is fundamentally limited by the spin-projection noise of the probed alkali-metal atoms and the photon shot noise of the probe light. However, in practice, the main limitation of the comagnetometer sensitivity comes from the air motion induced, e.g., by the temperature gradients around the oven heating the vapour cell. This noise scales inversely with the frequency, hence mostly affecting the sensitivity at lower frequencies.

In order to mitigate the noise, in high-performance comagnetometers, one can use an evacuated oven or even the entire optical apparatus [133]. Clearly, reducing the air pressure along the optical path of the probe beam markedly diminishes air turbulence and associated effects. Nevertheless, the integration of a vacuum system considerably enhances the complexity of both the experimental configuration and the operation of the comagnetometer.

Alternatively, the problem can be addressed with the implementation of the parametric modulation of the alkali-metal polarisation [104, 134]. This can be achieved with the modulation of the leading field. The idea of using the parametric modulation in the comagnetometer is, in fact, an extension of the parametric modulation of the SERF magnetometer concept described in Sec. 2.1.7.

The parametric modulation of the comagnetometer requires the application of the strong oscillating magnetic field along the polarisation axis. The frequency of the modulation has to be way above the bandwidth of the nuclear Larmor frequency (see Fig. 2.10). This condition allows the modulation to mostly perturb electron polarisation without significant effect on the nuclear polarisation. In such a case, the parametric modulation can be treated as a measure for more efficient detection of the electron polarisation state and can be neglected in consideration of the coupled dynamics of the electron polarisation of the alkali metal and the nuclear polarisation of the noble gas.

Implementation of the parametric modulation enables simultaneous observation of both transverse components of the electron polarisation. This ensures the extension of double-axis sensitivity by the low-frequency regime (see Sec. 2.2.11). The response at the first harmonics [see Eq. (2.126)] provides the information about the perturbations along the probe-beam propagation direction, while the second harmonic provides the information about the perturbations along the axis perpendicular to both pump- and probe-beam propagation directions. Therefore, one can consider x as the primary sensitive axis when the first harmonic response is considered as the output signal. If the second harmonic response is considered the output signal, the prime sensitivity axis is oriented along the y -axis.

From Eq. (2.126), one can see that the amplitude of the response to the transverse perturbations depends on the frequency and amplitude of the applied modulating field and the dependence is defined by the unitless parameter u [Eq. (2.127)]. Moreover, the first- and second-harmonic responses scale differently with u (see Fig. 2.14). The optimisation of the modulation parameters would require setting of the amplitude and frequency of the modulation such that the response amplitude is maximal. However, for the two-axis operation, the simultaneous optimisation of both response channels is impossible since the maximum response amplitude for the first- and second-harmonic signals is observed at two different values of u . Hence, the optimal value for u has to be checked experimentally. From Fig. 2.14, the u value is expected to be between the first local maxima of the responses at the first and second harmonics, which appears to be around $u = 1.2$.

The modulation frequency has to be such that it allows the effective lock-in detection of the output signal without limitation of the targeted output bandwidth. Hence, in practice, the targeted output bandwidth sets the requirement for the time constant and the filter of the lock-in amplifier, which ultimately leads to the lower limit for the modulation frequency. Then, the frequency of the modulation determines the optimal amplitude [Eq. (2.14)]. This connection effectively sets the higher limit for the frequency modulation since the increase of the modulation frequency leads to the nonlinear effects response of the order where Eq. (2.14) become invalid.

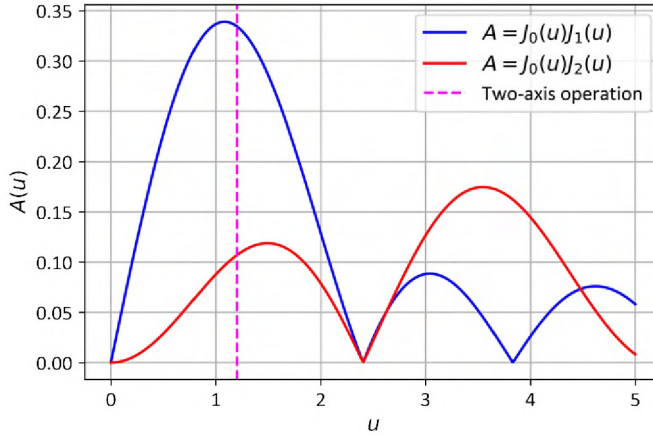


Figure 2.14: Bessel function defining the scaling factors for the first- and second-harmonic response to the parametric modulation of the alkali-metal atoms described with Eq. (2.126). The unitless parameter u is defined in Eq. (2.127). The dashed line indicates the value of u , which is optimal for two-sensitivity axis operation.

MODULATION OF THE ATOMS VS PROBE MODULATION

Parametric modulation is not the only scheme exploited to increase the sensitivity of the comagnetometer. Another frequently used technique is based on modulation of the probe light and successive demodulation of the light (detection of either intensity or polarisation state) after it traverses the medium using a lock-in amplifier. This method limits the detection bandwidth and hence improves the signal-to-noise ratio of the signal. As such, it is qualitatively different from the parametric-modulation method discussed above. Specifically, modulation of light properties is capable of transmitting the noise related directly to the probe light. In the case of the atomic modulation with the magnetic field, the low-frequency probe-light-related noise is not modulated, and therefore, the lock-in detection allows a limitation of the low-frequency noise related to the probe light parameters.

The signal detected with the photodiode S can be considered for the illustration of this discussion

$$S = S_a + \xi_{ph}, \quad (2.206)$$

where the S_a is the signal corresponding to the atomic polarisation, while the ξ_{ph} is the total noise related to the probe light (photon shot noise, $1/f$, etc.) The modulation of the probe light leads to a similar modulation of the signal from atoms and light noise

$$S^{LM} = (S_a + \xi_{ph}) \sin(\omega_m t), \quad (2.207)$$

so the demodulation of the output signal on the first harmonic of the modulation frequency will lead to the signal proportional to the original noise

$$S_{out}^{LM} \sim S'_a + \xi'_{ph}, \quad (2.208)$$

where $\sim S'_a$ and ξ'_{ph} are the low-pass filtered input atomic signal and the probe noise, respectively.

In the case of parametric modulation, only the atomic signal is modulated

$$S^{PM} = S_a \sin(\omega_m t) + \xi_{ph}, \quad (2.209)$$

therefore the signal demodulated on the first harmonic of ω_m is

$$S_{out}^{PM} \sim S'_a + \xi''_{ph}, \quad (2.210)$$

where ξ''_{ph} is the probe-related noise filtered with the window centred at the modulation frequency with a width two times wider than the output bandwidth of the lock-in.

The difference between the probe-light noise terms in Eqs. (2.208) and (2.210) lies in the central frequency of the noise passing through the detection scheme. For the white noise contributions (e.g., photon shot noise) both cases will lead to the same output noise since the noise spectrum is flat over all frequencies. However, for the frequency-dependent noise sources, such as $1/f$ noise related, e.g., to the air turbulence, the higher frequency window results in substantial suppression of the output noise.

2.3. SUMMARY OF THEORETICAL CONSIDERATIONS

This chapter presented the fundamentals of the optical magnetometry necessary for understanding the SERF magnetometry and self-compensation comagnetometry. This includes the spin dynamics inside of the atomic cell, the optical pumping and probing of the alkali-metal atoms and the spin-exchange optical pumping of the noble-gas atoms. It allowed tracking of the derivation of the models used for the simulation of the real-world sensors, starting from the first principles. The obtained model of the self-compensating comagnetometer [Eq. (2.163)] has been used for the derivation of the analytical solutions for the comagnetometer response to magnetic and non-magnetic spin couplings.

3

ADVANCED GNOME SENSORS

This chapter continues the discussion of the sensors used in the next generation of Global Network of Optical Magnetometers for Exotic Physics searches (GNOME), which is also known as Advanced GNOME (Sec. 1.4). The current Advanced GNOME network consists of two sensors located in Kraków (Poland) and Mainz (Germany). The majority of the experimental results provided in this work were measured with the Kraków Advanced GNOME magnetometer, as its development was a main part of the author's PhD project (see Chapters 4 and 6). The author also visited Helmholtz Institute in Mainz, where he participated in experiments involving the rotation of the Mainz Advanced GNOME sensor (see Chapter 5). To support the discussion of the experimental results, this chapter provides the details of the experimental setups for both stations, focusing mostly on the technical aspects of the experiments. The optimisation and characterisation techniques implemented in these systems are discussed in Chapter 4.

The key elements of the experimental setup are discussed on a generic scheme of the Advanced GNOME station presented in Fig. 3.1. The heart of such a system is an atomic cell containing atoms used to search for the interaction of interest. The cell is placed inside an oven, which allows precise control over the temperature and, hence, concentration/pressure of the atoms. Along with the cell, the oven is mounted inside a magnetic shield, which, in combination with a set of magnetic-field and magnetic-gradient coils, is used for precise control over the magnetic field in the volume of the cell. The state of the atoms necessary for the operation of the comagnetometer is created with the pump light, allowing optical polarisation of the alkali-metal atoms. The pump beam is circularly polarised and propagates along the z -axis. The state of the atoms is probed through the detection of the alkali-metal polarisation with the probe-laser light, which polarisation is detected with a polarimeter. The probe beam is linearly polarised and propagates along the x -axis. Since the alkali-metal polarisation state is detected via a parametric modulation technique (see Sec. 2.2.14), the polarimeter signal is demodulated at the parametric-modulation frequency. In the case of the GNOME synchronous measurements (GNOME science runs), the demodulated signal is sent to the **GNOME data acquisition system** [62] or, in the case of characterisation measure-



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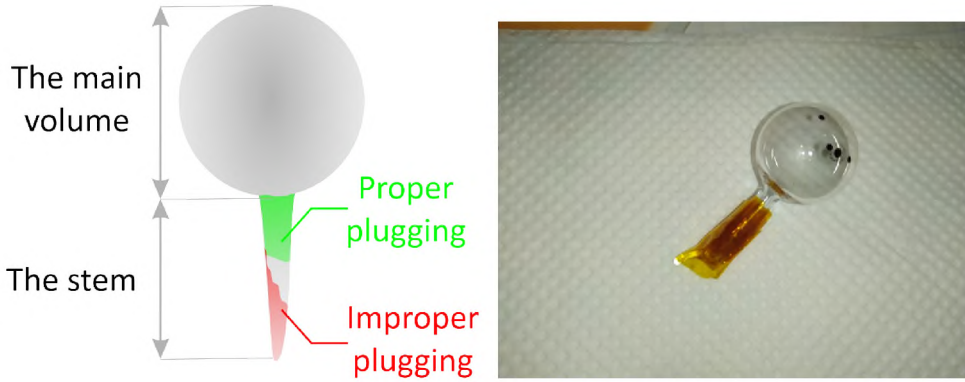


Figure 3.2: Schematic drawing of the atomic cell employed in the Advanced GNOME sensors (left) and a photograph of the cell used in Kraków station (right). The green area in the schematic drawing represents the proper plugging of the cell stem with the alkali-metal droplet, which prevents the diffusion of the polarised noble gas into the stem volume (the case of the Mainz setup). The red area represents improper plugging of the stem (the case of the Kraków setup). On the right picture is the photo of the cell similar to the one used in the Advanced GNOME comagnetometers.

point (for more details, see Sec. 4.4). Due to this reason, it is important to keep the volume of the stem as small as possible. The simple solution for this problem is to plug the stem with the alkali-metal droplet present in the cell. This prevents the diffusion of the polarised noble gas into the stem volume and hence improves the sphericity of the cell.

In order to plug the stem with the alkali-metal droplet, in both systems, the cell is placed with the stem pointing downwards. In such an arrangement, the droplet is kept in the stem by its own weight. This solution works well for the Mainz station, however, in the Kraków setup the metal droplet is too small to plug the stem properly (see Fig. 3.2). This leads to about 30% smaller polarisation of the ^3He obtained in the Kraków setup (see Sec. 4.4 for more details).

Although the entire cell is made of non-magnetic materials (except for the cell contents with non-zero atomic spins), the ferromagnetic impurities in the glass may produce the local gradients of the magnetic field near the walls of the cell, providing sufficient decoherence of noble-gas spins.¹ To alleviate this problem, the cell may be demagnetised prior to the installation of the setup.

3.2. HEATING SYSTEM

The operation of the comagnetometer requires a high concentration of the alkali-metal gas, which is ensured by the heating up of the atomic cell. In both Advanced GNOME stations, the comagnetometers operate at 180°C , which is maintained by the oven manufactured by Twinleaf. In the Kraków setup, the cell is installed on the cell holder made of Teflon (see Fig. 3.3), and the cell is attached to the holder with the Kapton tape. The holder is surrounded by a layer of thermal insulation placed between the stem and the tape for two reasons. First, it creates the temperature gradient between the cell volume

¹This is a serious problem that hold the development of the Kraków station for many months.



Figure 3.3: Atomic cell and a cell holder used in Kraków magnetometer. The cell is mounted with the Kapton tape. Between the tape and the stem, there is a piece of thermal insulation material.

and the stem (especially when increasing the temperature of the cell), which prevents the condensation of the alkali metals on the wall of the main part of the cell. Second, the elasticity of the thermal insulation prevents tensions due to the thermal expansion of the glass.

The cell with its holder is mounded inside the heat distributor made of Boron Nitride (see Fig. 3.4). The heat distributor surrounding the cell leads to its more homogeneous heating. The optical access to the cell is provided by five windows consisting of transparent vacuum tubes made of optical glass with antireflection coating (the semitransparent blue-coloured elements in Fig. 3.4). The vacuum tubes are used the air motion induced by the temperature gradient. They also improve the thermal insulation of the oven in the area surrounding the optical accesses. As the windows naturally tend to be the coolest area in the oven, it may result in the condensation of the alkalis on the walls of the cell. This risk is partially mitigated by the placement of the heating elements, which take the shape of an o-ring attached to the heat distributor (the dark-green elements in Fig. 3.4), such that the heating element surrounds the optical window of the oven. This provides a reduction of the temperature gradient near the windows.

The heaters are powered with a high-frequency (150 kHz) AC current, which limits the magnetic-field perturbation of the comagnetometer (the frequency of the current is far beyond the bandwidth of the comagnetometer). The temperature of the system is monitored with the PT1000 thermistor placed within the heat distributor. The temperature control and the heater powering are provided by the driver for the heating system (Twinleaf TCHF-50V).

The heat distributor is enclosed in the external housing made of Teflon. The space between the housing and the heater distributor is filled with glass fibre for better thermal insulation of the heating system. Outside, the oven housing is covered with an additional

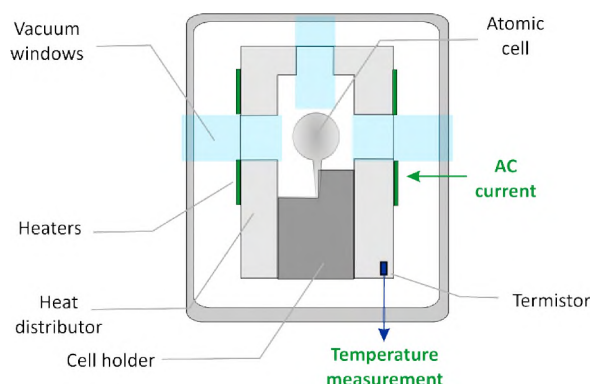


Figure 3.4: The scheme of the oven used for heating the atomic cell.

4-mm layer of thermal insulation based on the Aerogel [136], which provides additional thermal insulation between the oven and the magnetic shield.

The oven is placed on the Teflon stage inside the magnetic shield. The whole heating system is produced with non-magnetic materials and demagnetised prior to the installation inside of the magnetic shield. This limits any potential magnetic-field gradients, originating from the contamination of the materials.

3.3. CONTROL OVER MAGNETIC FIELDS

As was discussed in Chapter 2, the operation of the comagnetometer requires precise control over the magnetic field in the volume of the atomic cell. In practice, it requires the suppression of the ambient magnetic fields, which is provided by the mu-metal magnetic shields. However, the magnetic shields themselves can produce magnetic fields due to the residual magnetisation of the material they are made from. These fields may be suppressed by a set of magnetic-field coils placed inside the shield, which is also used for the application of controlled magnetic fields necessary for the operation of the system.

3.3.1. KRAKÓW SETUP

The Kraków and Mainz Advanced GNOME stations use two different models of magnetic shields. The Kraków setup utilises the Twinleaf MS-2 magnetic shield, which consists of the four cylindrical layers of mu-metal. The shield provides the suppression of the external magnetic field by a factor of 10^6 . The innermost layer of the shield has a length of 36 cm and a diameter of 18 cm. The shield has three optical access ports on each base of the cylinder. Five more optical access ports are available at the sides of the shield. The apertures of all ports are 1.8 cm in diameter except for the aperture along the main axis of the shield, which is 4.19 cm in diameter. The shield is equipped with a set of magnetic-field coils, which allow controlling magnetic fields in all three directions, and gradient coils, which allow one to control all five independent first-order gradients of the magnetic field.

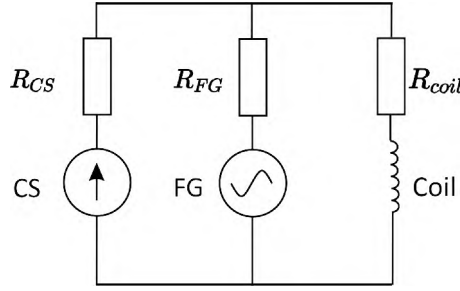


Figure 3.5: Electric circuit of the voltage divider used for combining static current from MCS and modulated current from a generator. CS stands for the current source, FG stands for function generator, R_{CS} is the resistor for the current source connection, R_{FG} is the resistor for the functional generator connection, R_{coil} denotes the resistance of the coil.

The coils are powered with the DM Technologies Multichannel Current Source (MCS) for the generation of the constant fields. The modulation of the field is provided by the voltage signal from the external generators, which is applied to the coils. The currents from MCS and generator are mixed using a voltage divider (see Fig. 3.5). The resistance of the additional resistors (R_{CS} and R_{FG}) has to be much larger than the resistance of the coil to avoid interference between the sources of the signal sent into the coil. The whole system is connected to the shielded coaxial cables, and the additional elements are mounted inside the grounded metal housing to avoid picking up of the laboratory electromagnetic noise.

3.3.2. MAINZ SETUP

The magnetic shield in the Mainz setup is the Twinleaf MS-1LF magnetic shield, which consists of three cylindrical layers of mu-metal and the ferrite innermost layer. The estimated shielding factor of this shield is about 10^6 . The shield is smaller than the Kraków setup shield dimensions, and the innermost layer is 12 cm in diameter and 14 cm in length. Optical access to the magnetic shield is possible through the apertures present on all sides of the shield. The apertures are 1.8 cm in diameter for all apertures except for the main axis, where the aperture has a diameter of 3 cm.

The shield is equipped with a magnetic-field and gradient coils, which provide the same functionality as the coils in the Kraków setup. The magnetic field coils are powered with the Twileaf CSB-20 current source, while the gradient coils are powered by the voltage output channels of the lock-in amplifier (ZI MFLI 5MHz) connected to the coils with an additional high-precision resistor. All connections are made with coaxial shielded cables, and the additional components are enclosed in the metal housing for shielding from the pick up of the external electromagnetic perturbations.

3.4. LASER SYSTEM

The current GNOME comagnetometers employ the experimental arrangement with two separate laser beams, one of which is used for probing, and the other is used for pumping of the alkali-metal atoms. The Mainz system also has an additional laser beam pointing

along the sensitivity axis, which is used for the generation of the AC Stark effect and calibration of the comagnetometer. The details of the implemented laser systems are discussed in the successive subsections.

3.4.1. KRAKÓW SYSTEM

PUMP LASER

In the Kraków system, the pumping light is provided by the Toptica TA Pro tapered amplifier driven with the Toptica DLC Pro laser controller. The laser is tuned to the centre of the rubidium D_2 line and is not locked to any external frequency reference, considering the pressure broadening of spectral lines induced by ^3He , which leads to the linewidth of about 60 GHz (for more details, see Sec. 2.1.5). The right-handed circular polarisation is ensured by a crystal polariser and quarter-wave plate. The pump beam is guided through free space (no optical fibres) and is expanded before the magnetic shield such that the coverage of the cell is maximized (the Gaussian beam profile diameter after the extension of the laser beam is about 1.8 cm). Such a beam expansion is used to maximise the transverse homogeneity of the beam used for optical pumping. After passing the cell, the pump light falls onto the beam dump, where it is absorbed.

The optimal operation of the Kraków comagnetometer requires about 150 mW/cm^2 of the laser power going into the magnetic shield. The pump laser is operated on the exceeding output power (the total output power is about 3.5 W), and then the power of the pumping beam is decreased by the subsequent polariser. Such a regime of operation was found to be optimal for the homogeneity of the pump beam.

PROBE LASER

In Kraków, probing of the potassium polarisation is done with linearly polarised light generated by the Toptica DL Pro laser system driven with the Toptica DC 110 laser driver system. The laser frequency is blue-detuned by about 0.6 THz from the centre of the potassium D_1 line and its power is 1 mW (before entering the shield). The diameter of the Gaussian beam profile of the probe beam is equal to about 3 mm. The linear polarisation of the beam is ensured by the crystal polariser placed in the magnetic shield. After the cell, the transmitted light polarisation is measured with the polarimeter. The polarimeter consists of the Wollaston prism and the differential photodetector [see Fig. 3.6 (right)]. The Wollaston prism splits the beam into two orthogonal polarisation components, which orientation is determined by the axis of the prism. In the measurements, the axis of the prism is oriented in such a way, that for the unpolarised atoms (no optical rotation) intensities of the components are equal. The components are detected by two channels of the differential photodetector. The output signal of the differential photodetector is proportional to the difference between the intensities of the light detected by its two inputs.

3.4.2. MAINZ SETUP

PUMP LASER

The Mainz comagnetometer is optically pumped with circularly polarised light tuned to the centre of the rubidium D_1 line. The pump beam has a diameter of about 1 cm and the power of 70 mW. The pumping light is generated with the Toptica TA Pro tapered am-

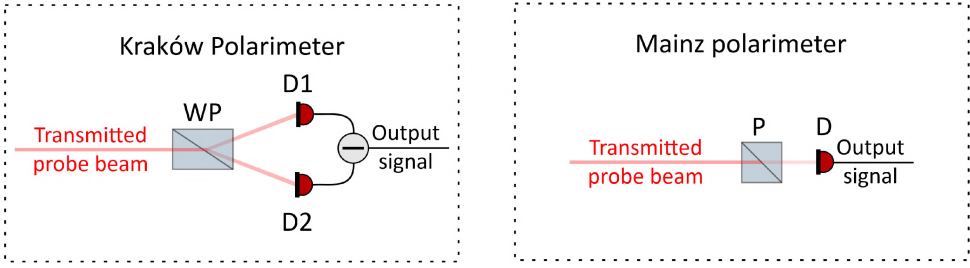


Figure 3.6: Schematic representation of the polarimeter employed in the Kraków (left) and Mainz (right) setups. WP is the Wollaston prism, P is the polariser, and D stands for the photodetectors.

plifier controlled with the DLC Pro digital laser driver. An important difference between the Kraków and Mainz pump-laser systems is that in the latter, the laser light is coupled to polarisation-maintaining (PM) fibre and is transmitted next to the shield within the fibre. This solution allows for the reduction of intensity noise present in the free-space configuration and originating mostly from air motion and thermal drifts in optomechanical components of the setup. The pump light is decoupled from the fibre using a mount attached to the shield using the cage system. This system is also used for the mounting of remaining optical elements, such as the beam, the expander, and the quarter-wave plate.

PROBE LASER

The state of the Mainz comagnetometer is probed with the linearly polarised light blue detuned by about 0.5 THz from the potassium D_1 line. The probe light is generated by the Toptica DL Pro laser system controlled with the Toptica DLC Pro laser driver. Similarly, as in the pump-light case, the probe light is guided through a PM fibre and leaves the fibre close to the shield using a coupler mounted to the shield with the cage system, which also holds the crystal polariser and half-waveplate used for control of the pump-beam intensity and polarisation.

The probe light transmitted through the atomic cell goes to the cage-mounted polarimeter. This polarimeter consists of the crystal polariser (playing a role of an analyser in this case) and a single photodetector [Fig. 3.6 (right)]. The main axis of the polariser is oriented in such a way that it is nearly crossed with the initial polarisation of the probe light. The polarisation rotation of the probe beam then causes a change in the intensity of the light detected by the photodetector which is proportional to the angle of the rotation.

3.4.3. LIGHT-SHIFT GENERATING LASER

The Mainz system is equipped with an additional laser used for the generation of the AC Stark effect, employed for testing and calibrating the comagnetometer.² To do so, the circularly polarised light is applied along the x -axis. The laser is blue detuned by about

²No such laser is used in the Kraków station, thus the setup does not offer the capability of testing non-magnetic spin couplings based on the AC Stark effect.

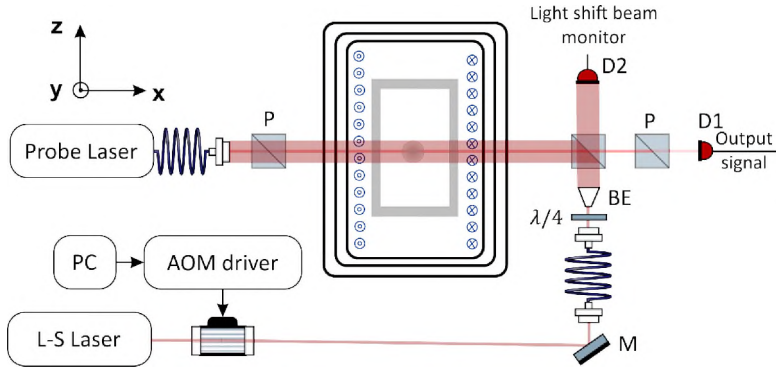


Figure 3.7: Simplified scheme of the Mainz setup, presenting the elements used for controllable generation of the light shift and probing of the alkali-metal polarisation. PC stands for the personal computer controlling the system, P is the polariser, D denotes photodetectors, L-S laser stands for the laser used for the generation of the light shift, M is a mirror, $\lambda/4$ is the quarter wave plate.

1.5 THz from the potassium D_2 line. The amplitude of the applied light shift is controlled through the power of the applied light.

The light-shift beam is generated by the MSquared Ti:Sa laser. This free-space beam is sent through the acousto-optical modulator (AOM) operating in the first order of diffraction, which is used for modulation of light intensity. This solution enables shaping the light pulses and hence the time dependence of the perturbation. The light directed into the first order of AOM diffraction is sent to the optical coupler attached to the high-power PM fibre. The optical fibre is used for guiding the light to the output coupler attached to magnetic shield with the cage system [see Fig. 3.7]. To limit the cross-talk between the beams, the light-shift beam propagates in the opposite direction with respect to the probe beam. The overlapping of those two laser beams is achieved with the 70/30 beam splitter placed in the crossing of the beams.

3.5. OUTPUT SIGNAL

Both currently operating Advanced GNOME comagnetometers employ parametric modulation for the efficient detection of the alkali-metal polarisation (for more details, see Sec. 4.1). This requires the generation of high-frequency modulation of the leading field, which in the systems is provided by an internal generator of the lock-in amplifier (see Fig. 3.1). In that case, the polarimeter signal is sent to the input of the lock-in amplifier and demodulated at the modulation frequency. The signal demodulated at the first and second harmonics of the frequency provides information about the perturbations along x and y , respectively. In the Kraków setup, demodulation on the first two harmonics is provided by two separate lock-in amplifiers (SRS 830), while in the case of the Mainz setup, a single lock-in amplifier (ZI MFLI 5 MHz) allows the use of two separate demodulators for the same input signal.

The demodulated output signals are recorded with the GNOME DAQ system and/or directly with a computer. The GNOME DAQ system time-stamps the measured data points with respect to the GPS time reference and sends them to the computer for long-

term storage and further analysis. The standard sampling rate utilised in the GNOME searches is 512 samples/s.³

The Kraków setup is also equipped with the GNOME sanity system [135]. This is an Arduino-based system using a flux magnetometer, accelerometer, and gyroscope. The sensors of this system are placed near the magnetic shield, which allows for flagging of the laboratory environment anomalies unrelated to the exotic spin couplings. The sanity system sends the digital signal to the GNOME DAQ system, which, in case of classical perturbations exceeding the thresholds, flags the comagnetometer data (the readouts of the sanity-system sensors are independently stored on a local computer).

3.6. CALIBRATION PULSE

The Kraków setup employs a periodic magnetic-field pulse for monitoring the comagnetometer and its precise calibration⁴ (for more information see Chapter 5). For this purpose, the magnetic-field pulse can be applied along the x - or y -axes. The signal with the magnetic-field pulse is generated with the NI 6221 PCI DAQ card, which is connected to the modulation input of the magnetic coil (Fig. 3.5). The connection is realised via the relay, which is controlled through a digital signal generated by the same DAQ card. The relay allows physical disconnection of the DAQ card from the magnetic coil when it is not used. This enables the avoidance of contamination of the comagnetometer signal with additional noise.

The time of the pulse is precisely synchronised with the GPS reference provided by the GNOME DAQ system. This system provides a pulse-per-second (PPS) signal precisely synchronised with the GPS time reference. The PPS signal is sent to one of the inputs of the AND gate used to trigger the calibration pulse. The second input of the AND gate is connected to the DAQ card programmed to send a 1-s long digital signal centred at the time of pulse (see Fig. 3.8). The time of pulse is defined with respect to Universal Time Coordinated (UTC) based on the local computer time, which is synchronised with the local Internet time server.⁵ The synchronisation of the pulse sent from the DAQ card with the GPS-synchronised PPS signal is necessary for the compensation of the delay acquired by the local computer clock, which was measured at a level of 2 ms/day for the Kraków computer.

3.7. LOCATION OF THE EXPERIMENTAL SETUPS

3.7.1. KRAKÓW STATION

The Kraków comagnetometer is located at the second floor of the Marian Smoluchowski Institute of Physics of the Jagiellonian University in Kraków. The geolocation coordinates of the experimental setup are: latitude: 50.03° , longitude: 19.91° . The azimuth angle of the prime sensitivity axis ($\mathbf{x}$ in the laboratory frame) is 156° , while for the pump beam orientation ($\mathbf{z}$ axis in the laboratory frame) is 246° . The altitude of both axes is equal to 0° (they are horizontal with respect to the Earth's surface). Thereby, the sensitivity

³The sampling rates in the range between 1 S/s to 1024 S/s, in steps of two, are possible.

⁴At the point of writing this dissertation, the Mainz system is not equipped with such a capability.

⁵The local in this context means the servers located in Poland.

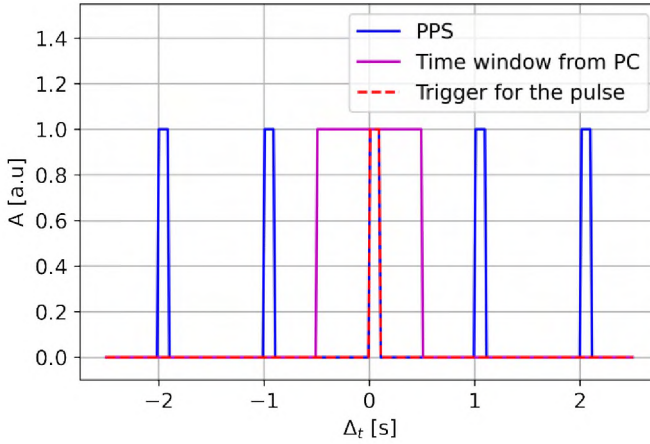


Figure 3.8: Time equence triggering the calibration pulse. The PPS signal (blue) and the time-window signal (computer DAQ card) are sent to the AND gate. The output of the gate is used for triggering of the calibration pulse. The triggering pulse is synchronisation with the PPS signal reference the GPS time. The time axis Δ_t is measured in time delay from the time of an actual pulse.

plane is vertical with respect to the Earth's surface and is oriented at the 156° , along the azimuth angle.

3.7.2. MAINZ STATION

The Mainz station is located on the underground floor of the Helmholtz Institute at Mainz. The geolocation coordinates of the experimental setup are latitude: 50.0° and longitude: 8.23° . The azimuth angle of the prime sensitivity axis (x in the laboratory frame) is oriented at 162° , and it is parallel to the Earth's surface (the altitude angle = 0°). The pump-beam orientation (the z -axis in the laboratory frame) is vertical with respect to the Earth's surface (altitude angle = 90°).

3.7.3. RELATIVE ORIENTATION OF THE SENSITIVITY PLANES

The prime sensitivity axes of both stations are near parallel and they are near-parallel to the Earth's rotation axis. Hence, the orientation of the prime sensitivity axis of the Advanced GNOME stations is weakly dependent on the daily rotation of the Earth. This should be taken into account in the experiments, where the daily modulation of the low-frequency response is expected. In a high-frequency regime, one should take into account the sensitivity planes. The common-mode response of the Mainz and Kraków stations is determined by the intersection of the sensitivity axes from the two stations, which practically overlaps. However, the near-perpendicular orientation of the pump beams for the stations leads to the sensitivity planes being near-perpendicular, therefore eventual daily modulation effects on the station's signal will be shifted in phase by nearly a quarter of a day.

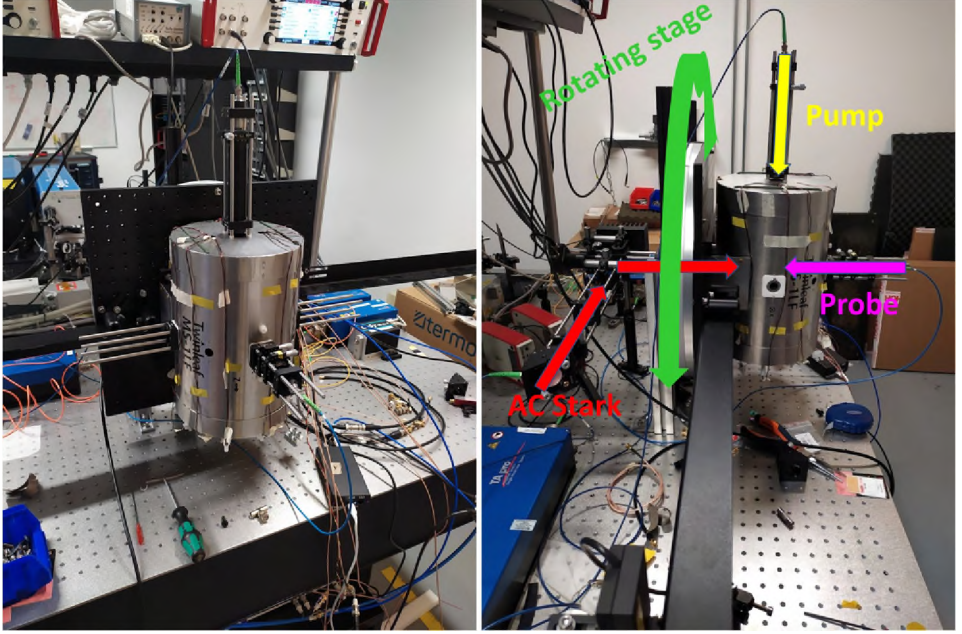


Figure 3.9: Photos of the Mainz comagnetometer. The magnetic shield is mounted on the rotating stage, which allows the rotation of the whole system. The fibre-guided laser light, which output couplers mounted at the side of the magnetic shield, allows the rotation of the system without a change in the relative orientation between the applied magnetic fields and laser beams.

3.8. THE ROTATION SYSTEM

The Mainz comagnetometer is mounted on the rotating stage (Fig. 3.9), which allows the controllable rotation of the system around the prime sensitivity axis. This is used for the direct calibration of the system. The output couplers of the fibre-guided pump, probe, and light shift laser beams are mounted on the side of the magnetic shield and rotation stage. This allows the rotation of the system without relative change of the orientation between the laser beams and the magnetic fields applied to the atomic sample.

The rotation of the stage is driven by the Thorlabs MTS50-Z8 step motor, which is mounted to the long mechanical arm attached to the stage (Fig. 3.10). The step motor moves the end of the arm in a vertical direction, which translates into the rotation movement of the stage. The motor is capable of the movement with the maximum linear acceleration of $a_{max} = 4.5 \text{ mm/s}^{-2}$ and the linear velocity of $\dot{\theta}_{max}^r = 2.5 \text{ mm/s}$. The mechanical-arm length (the distance between the rotation axis and the point of the connection with the step motor) is $L = 70 \text{ cm}$. This leads to the maximal possible angular frequency of the rotation ω_{max}^r

$$\omega_{max}^r = \frac{\dot{\theta}_{max}^r}{L} \approx 0.0036 \text{ rad/s.} \quad (3.1)$$

This corresponds to the effective nuclear perturbation amplitude equal to $b_{max}^r = 17.43 \text{ pT}$.

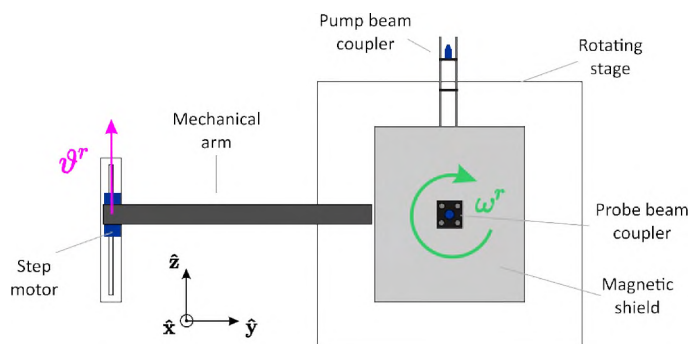


Figure 3.10: Scheme of the rotation system implemented in the Mainz system. The system is mounted on the rotating stage of the rotation axis overlapping with the primarily sensitive axis of the comagnetometer. The linear motion of the step motor is translated into the rotation motion through the mechanical arm attached to the rotating stage.

3.9. FINAL REMARKS ON THE ADVANCED GNOME SENSORS

In this chapter, the key technical details of the Advanced GNOME comagnetometers developed in Kraków and Mainz were described. Although both systems realise similar measurement protocols and are built with the vapour cells with the same isotopic contents, there are a number of differences between implemented solutions. The difference is notable even visually when the photograph of the Mainz system (Fig. 3.9) is compared with the photo of the Kraków setup (Fig. 3.11). Such a variation in the technical solutions utilised in Advanced GNOME comagnetometers leads to the requirement for standardization of the comagnetometer calibration and maintenance procedures, which allow meaningful comparison of the measurements from different stations. Specifically, this is discussed in details in Chapter 5. In the next Chapter, the Kraków comagnetometer is used as an example in a discussion of the techniques which can be used for the optimisation of the GNOME comagnetometers.

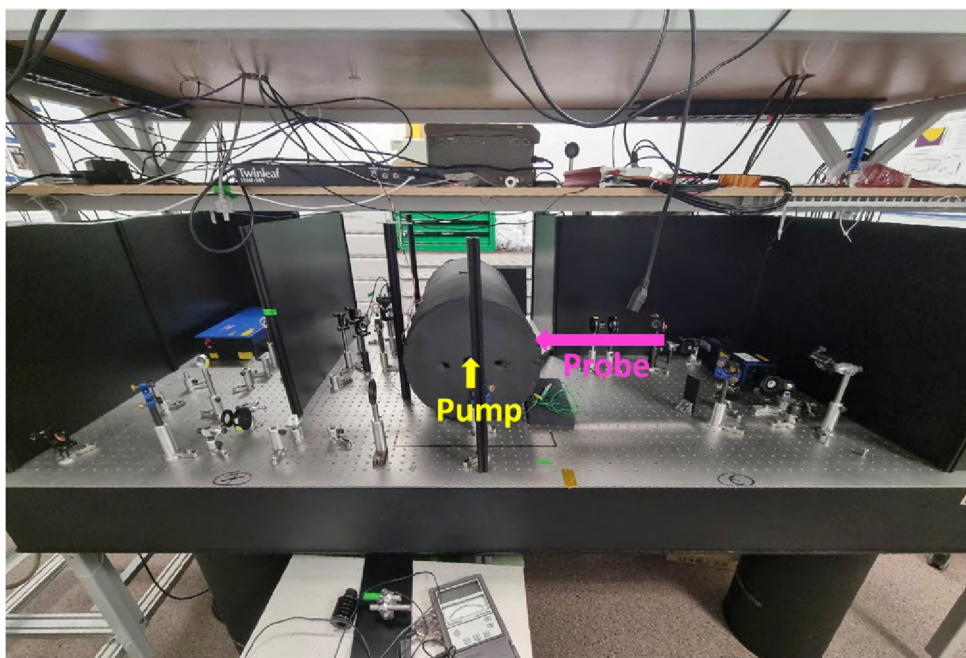


Figure 3.11: Photo of the Kraków Advanced GNOME comagnetometer. The arrows depict the orientations of the probe (magenta) and the pump (yellow) beam propagation directions.

4

CHARACTERISATION AND OPTIMISATION OF THE SENSOR

4.1. OPTIMISATION OF THE PARAMETRIC MODULATION

To perform ultrasensitive searches for yet undiscovered exotic spin coupling, optimisation of the comagnetometer performance is one of the most important tasks to be realised. A specific approach to achieve this goal involves optimisation of parametric modulation, specifically such parameters of the modulation as its frequency and amplitude (principles of the parametric-modulation technique are described in Sec. 2.1.7). This chapter describes the optimisation process based on the example of the Kraków Advanced GNOME comagnetometer.

4.1.1. THE OUTPUT BANDWIDTH OF THE DETECTION SYSTEM

The bandwidth requirement for the comagnetometer to be used in GNOME searches determines an upper limit on the time constant used for the lock-in detection of the modulation. In other words, the time constant sets a lower limit on the frequency that can be used for the modulation, as the carrier frequency has to be filtered out by the low-pass filter of the lock-in amplifier. Thereby, the parametric modulation frequency is predetermined and the optimisation process falls into the search for the optimal value of the modulation amplitude. This task is tackled with the measurement of the amplitude of the comagnetometer signal as a function of parametric-modulation amplitude. In fact, this is an experimental determination of the dependence presented in Fig. 2.14.

In our searches, the desired bandwidth of the comagnetometer searching for transient or oscillatory non-magnetic perturbations is 100 Hz. In the Kraków station, this is achieved by detecting the parametrically modulated signal with a lock-in amplifier of the 1-ms time constant and -12 dB/oct filter. In this case, the frequency of the sine-wave modulation of the leading field is set to 2 kHz.

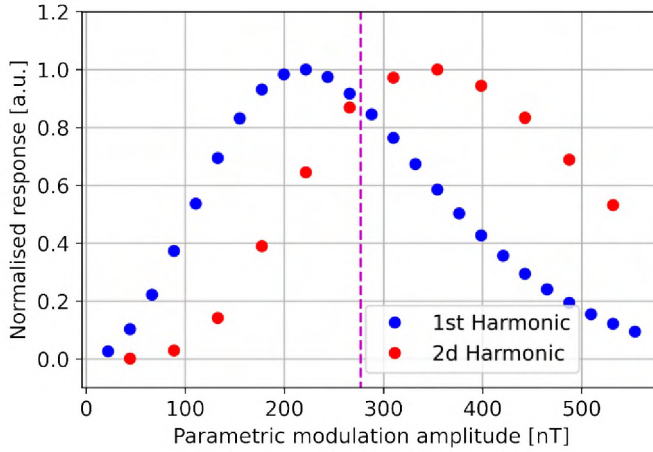


Figure 4.1: Normalised first- and second-harmonic response of the comagnetometer versus parametric-modulation amplitude. The probing sinusoidal modulation of the magnetic field with a constant amplitude of 1 pT and a constant frequency of 40 Hz have been applied along the x -axis for the measurements of the first harmonic response and along the y -axis for the second-harmonic response. The dashed magenta line denotes the modulation amplitude optimal for the two-axis operation of the magnetometer.

4.1.2. MODULATION AMPLITUDE

The parametric modulation tests are conducted with the comagnetometer operated at the compensation point (for more details, see 2.2.4). In that case, to analyse the first harmonic response, the response amplitude is probed by applying the test sinusoidal magnetic field along the primary sensitivity axis x . In turn, the sinusoidal modulation along the y -axis is used to analyse the second-harmonic response. The fields used for the analysis of the response later in this work will be called the **probing field** for differentiation from the field used for parametric modulation. The amplitude of the probing magnetic field is 1 pT, and its frequency is 40 Hz. The probing-modulation frequency is chosen to be above the helium Larmor frequency but still within the bandwidth for the alkali-metal response. The amplitude of the response is determined using the Fast Fourier Transform (FFT) of the output signal, in particular, the amplitude of a 40-Hz component observed in the spectrum.

The results of the measurement are presented in Fig. 4.1. As shown, two maxima (one at the first harmonic and the other at the second harmonic) arise at different modulation amplitudes. As the Advanced GNOME sensors have to be operated in a two-axis mode, the compromise modulation amplitude is chosen such that the response at both harmonics is at a level of 90% of their maximums.

Although experimental data follows qualitatively the predictions of the theoretical model (see Fig. 2.14), one can also note its slight deviation from the predictions based on the Bessel functions. This is particularly visible at higher modulation amplitudes. This is due to the nonlinear response of the alkali-metal polarisation to the perturbation, which becomes significant at these frequencies. It should be stressed that the same behaviour is also observed in the SERF magnetometers [104].

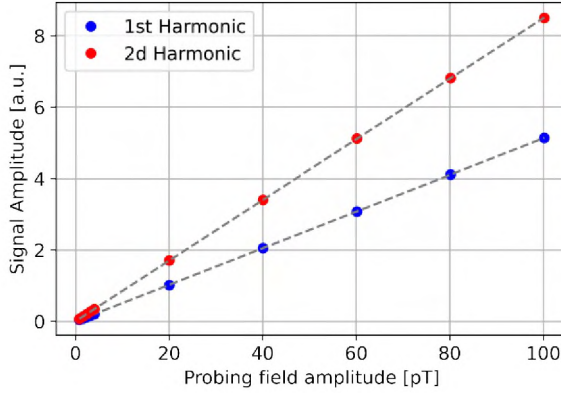


Figure 4.2: Amplitude of the signal detected at the first and second harmonic of the modulation frequency versus the modulation amplitude along with the linear fits. The provided results present the dependence of the comagnetometer response to sinusoidal perturbation with a frequency of 40 Hz applied along the x -axis in the case of the first harmonic response and the y -axis in the case of the second harmonic response.

4.1.3. LINEARITY OF THE RESPONSE

The theoretical predictions for the comagnetometer response presented in Sec. 2.2.4 are derived under the approximations of small angles, which is equivalent to the linearisation of alkali-metal polarisation response within the coupled comagnetometer system. Therefore, the linearity of the response indicates the system operation under conditions described in the theory section.

The linearity of the comagnetometer response has been studied with a periodic probing perturbation similar to the one used for the optimisation of the parametric modulation. However, in that case, the amplitude of the parametric modulation was constant, and the amplitude of the probing modulation have been varied. The results of the measurements are presented in Fig. 4.2. The measured response of the comagnetometer is linear, which confirms the agreement between the theoretical predictions and the experiment within the perturbation range of the interest.

4.2. OPTIMISATION OF THE PROBE LIGHT PARAMETERS

Maximisation of the comagnetometer sensitivity requires optimisation of the probe light. The probe-light parameters, mainly its power and frequency, can be optimised with respect to the signal-to-noise ratio (SNR) of the detected signal. These two parameters can affect both the amplitude and noise of the detected signal, so it is crucial to optimise them with respect to the SNR value.

Similar to the case of the amplitude optimisation discussed in Sec. 4, the direct application of the non-magnetic perturbation to the spins is challenging and experimentally impractical. Therefore, same as before, the system is probed with the sinusoidal modulation along the x -axis with an amplitude of 1 pT and a frequency of 40 Hz, and the noise floor for the SNR analysis has been estimated with the average noise in the 10 Hz wide window centred at 40 Hz.

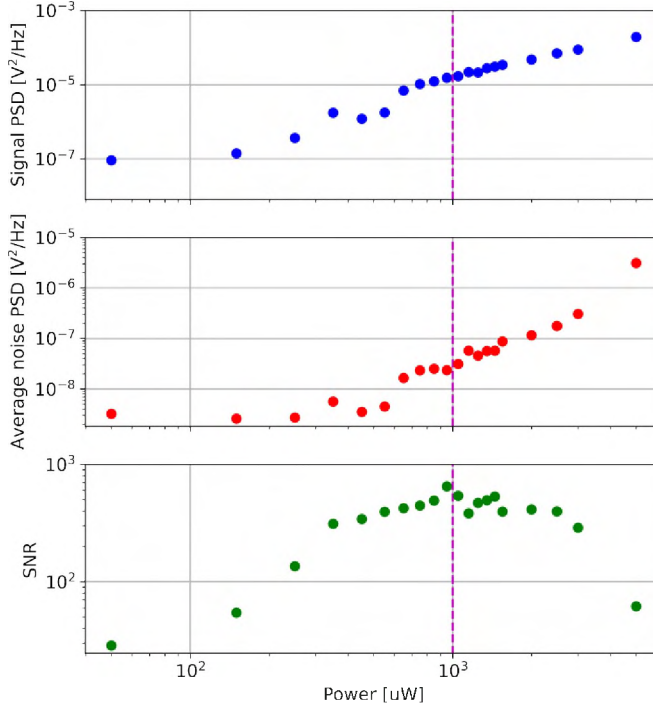


Figure 4.3: Experimental optimisation of the probe beam power in the Kraków comagnetometer. The presented results are obtained with the probe modulation of the system with the sine-wave magnetic field. The modulation frequency is set to 40 Hz, while the amplitude of the modulation is 1 pT. The presented average noise floor is obtained with a 10 Hz window centred at the modulation frequency (with the exception of the 50-Hz point). The dashed magenta line denotes the probe power chosen for the operation of the comagnetometer.

The results of the probe-power optimisation are presented in Fig. 4.3. As shown, the signal amplitude is proportional to the power of the probe beam, with the exception of the probed powers below $200 \mu\text{W}$, where its amplitude is strongly affected by absorption of the light in the atomic vapour. At the same time, the noise floor does not change up to $\approx 400 \mu\text{W}$, where it eventually starts to rise. For the lower end of the probe power, the noise is dominated by the sources unrelated to the probe light (e.g., electronic noise). For higher power, it starts to depend on the power of the probe light, which indicates that the probe-light polarisation noise becomes dominant. For our experimental conditions, the optimal value of SNR is observed at a probe-power of about 1 mW. The fact that at such a power there is no strong dependence on SNR on light intensity is beneficial in the scope of the long-term stability of the system.

The dependence of SNR on the detuning of the probe light from the rubidium D_1 transition is presented in Fig. 4.4. The detuning mostly affects the amplitude of the response. Similarly to the case of power optimisation, the results for the detuning reveal a plateau of SNR close to the point of maximum SNR. The optimal value of the detuning

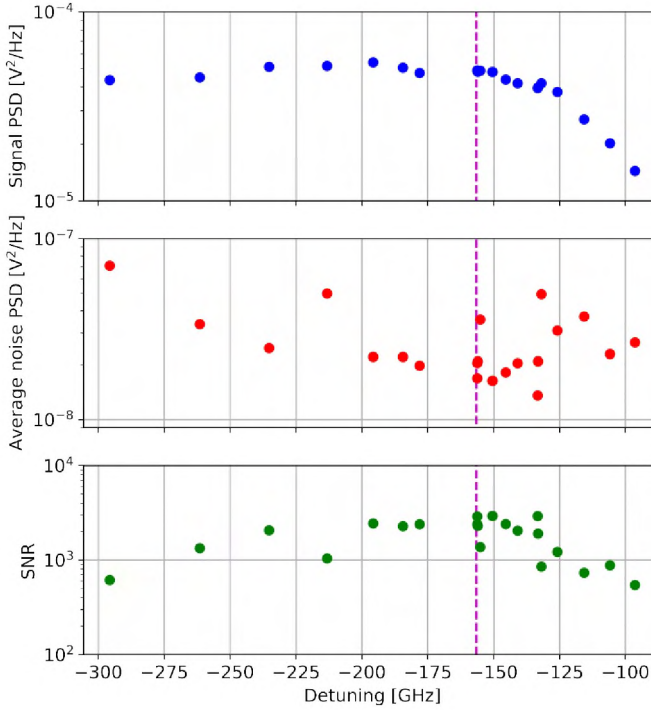


Figure 4.4: Experimental optimisation of the probe-light frequency in the Kraków comagnetometer. The presented results are obtained with the probe modulation identical to the case described in Fig. 4.3. The dashed magenta line denotes the frequency of the probe beam chosen for the operation of the comagnetometer.

is determined by the approximate centre of the flat area in the SNR results, which allows for improving both the sensitivity and the long-term stability of the comagnetometer system.

4.3. DIRECT MEASUREMENTS OF ^3He MAGNETISATION

Monitoring of the noble-gas polarisation is an important element in the development and characterisation of the noble-gas-alkali-metal comagnetometers. There are two techniques of monitoring the noble-gas polarisation used in the development of the Advanced GNOME sensors. The first method is based on the single-beam measurement of the synchronously pumped SERF magnetic resonance (Sec. 2.1.6). The second involves the observation of the comagnetometer response to the magnetic-field modulation in various leading fields. Both methods exploit the original idea of using the shift in the Larmor frequency of the alkali-metal atoms generated by the interaction with the polarised noble-gas atoms [122]. However, the different experimental realisation of the measurements leads to complementary results and reveals various aspects of the system behaviour. The details of the experimental realisation of these methods and the experimental results obtained with the Kraków comagnetometer are presented in the following

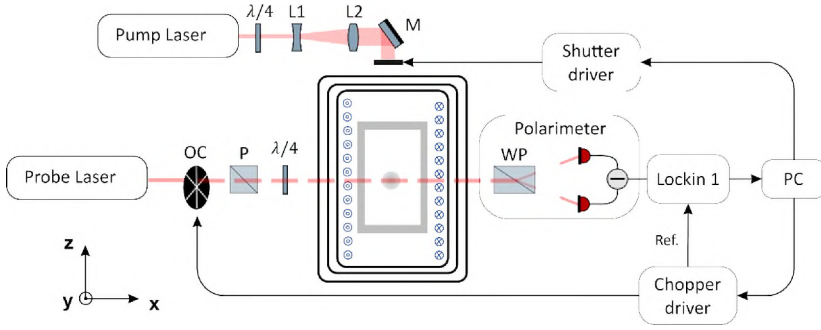


Figure 4.5: Experimental setup for the noble-gas polarisation measurements exploiting the measurements of synchronously pumped alkali-metal atoms.

4

subsections.

4.3.1. SYNCHRONOUSLY PUMPED SERF RESONANCE

In this section, the method for a non-perturbing measurement of the noble-gas polarisation is proposed. The method uses the alkali-metal magneto-optical resonance observed with the synchronous pumping. In the comagnetometers system, observation of the magnetic resonances is similar to the measurement of the magnetic resonances in the SERF magnetometer, which is described in Sec. 2.1.6. However, in this system, in addition to the leading magnetic field, the alkali-metal polarisation is exposed to the frequency shift generated by the interaction with the polarised noble gas.

SERF RESONANCES IN THE COMAGNETOMETER SYSTEM

The measurement is realised in a single-beam configuration, in which the probe-beam polarisation is changed from linear to elliptical by adding a quarter-wave plate before the light goes inside the shield (Fig. 4.5). This allows one to simultaneously pump and probe the alkali-metal atoms. The modulation of the light intensity, necessary for the synchronous pumping, is provided by the mechanical chopper placed in the beam path before the light enters the shield. The measurement is performed at the constant leading field applied along the z -axis, whose amplitude is far from the compensation point. The synchronously pumped resonances are recorded by scanning the chopper/modulation frequency and demodulating the polarimeter signal at the first harmonic of the modulation frequency. When the magneto-optical resonance is measured, the pump beam is blocked by the mechanical shutter to avoid perturbation of atoms from the pump beam. Figure 4.6 presents a representative magnetic resonance measured with the technique.

Measured signals are fitted using the results of Sec. 2.1.6, where the formula for the magnetic resonance is derived, and the resonance parameters are obtained. To adapt the results of the theoretical section to the comagnetometer case, one has to take into account the difference in the orientation of the pump-beam axis. The theory section considered the arrangement with perpendicular pump and probe beams, while here, a single beam is used for pumping and probing. This difference results in a change of the global phase of the complex Lorentzian profile [Eq. (2.124)] without modification of other

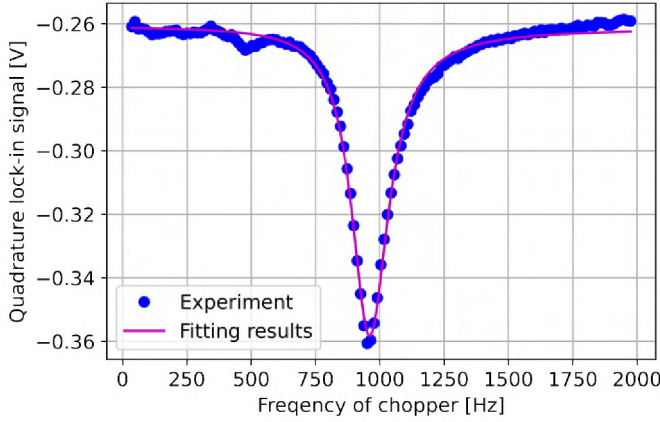


Figure 4.6: SERF magneto-optical? resonance measured using synchronous pumping. The experimental data (blue dots) is the quadrature response at the chopper modulation frequency obtained with the lock-in amplifier. The solid line provides the results of the experimental-data fitting with the model from Eq. (4.1).

parameters. The second difference to be considered lies in the pumping-rate modulation pattern. In the discussed experiment, the mechanical chopper provided square-wave modulation of the pumping rate, while the theoretical model assumes sinusoidal modulation. However, the first-order approximation of the square wave is represented by the sine function, and hence Eq. (2.123) provides a valuable approximation of the pumping rate modulation in the considered experiment.

The solution given in Eq. (2.124) leads to the following fitting model for the quadrature signal obtained with the lock-in detection

$$S_q = \text{Im}[(S_- - S_+)\exp(i\phi)] + c, \quad (4.1)$$

$$S_{\pm} = \pm a \frac{f_0 - f + i\delta_f}{(f \pm f_0)^2 + \delta_f^2},$$

where ϕ , c , a , f_0 and δ_f are fitting parameters, while f is the frequency of the chopper. The fitting parameters have the following interpretation: ϕ is the relative phase shift, c is the offset signal, which is present due to the non-zero time-averaged pumping rate; f_0 is the position of the resonance, while δ_f is the width of the resonance and a is the amplitude of the resonance.

INSIGHTS FROM THE NUCLEAR POLARISATION DYNAMICS

Relaxation or buildup of the ^3He polarisation can be observed by repetitive observation of the magnetic resonances.¹ The change in the noble-gas polarisation amplitude leads to changes in the SERF resonance frequency. Figure 4.7 presents an example of the ^3He relaxation observed in the Kraków comagnetometer.

¹In case of the pumping and buildup of the polarisation, the pumping light is blocked only for the time of the SERF resonance measurement. To minimise a systematic effect of the periodically blocked pump beam, the repetition time of the resonance measurement has to be much longer than the time of scanning the resonance.

The presented method is particularly useful for the direct determination of the longitudinal relaxation time of the noble-gas polarisation T_1^n . Unlike many conventional methods, this technique does not destroy the noble-gas polarisation. This not only simplifies the measurements of T_1^n but also makes it possible to apply the technique in the cases when the longitudinal relaxation time is comparable with the transverse relaxation time $T_2^n \approx T_1^n$. Moreover, the method may be further improved by adding the feedback system controlling the chopper frequency to keep the modulation in resonance with the Larmor frequency of the alkali-metal atoms. This enables continuous monitoring of the level of the noble-gas polarisation, making it even more suitable for cases when $T_2^n \approx T_1^n$.

The measurement of the noble-gas relaxation can be a powerful tool in the analysis of the magnetic-field conditions to which the noble-gas polarisation is exposed. In particular, for the ^3He cells, the scaling of T_1^n with the applied magnetic field may provide information about the dominating source of relaxation. Specifically, when the magnetic-field gradients are the dominating source of ^3He relaxation, T_1 is inversely proportional to the amplitude of the applied magnetic field [Eq. (2.133)]. However, if the atomic cell is magnetised due to the ferromagnetic contamination of the cell material, the relaxation time scales linearly with the applied leading field.² Therefore, observation of the relaxation time versus the magnetic field provides information about the dominant relaxation source and may imply the necessity for demagnetisation of the atomic cell.

The analysis of the noble-gas relaxation may also provide information about the self-interaction of the noble gas. If such interaction occurs, the noble-gas magnetisation leads to the deviation of the noble-gas polarisation relaxation from the expected exponential decay. Such a situation is observed in the Kraków comagnetometer (Fig. 4.7). It is believed that the effect is associated with the asphericity of the atomic cell (for more details, see Fig. 3.2 and Sec. 3.1). Hence, in addition to diagnostics of the comagnetometer system at the development stage, the presented method can also be employed for the systematic studies of the atomic cell quality and tests of novel shapes and designs of atomic cells.

THE RESULTS FROM THE KRAKOW COMAGNETOMETER

As was discussed at the beginning of this section, the resonance frequency of the SERF resonance [Eq. (4.1)] in the comagnetometer depends on the polarisation level

$$f_0 \sim \zeta_z. \quad (4.2)$$

Hence, the changes in the f_0 can be related to the changes of the noble-gas polarisation. For example, in the absence of the self-interaction, the relaxation rate of the noble-gas polarisation is expected to be independent of the polarisation amplitude, and therefore, the change of the SERF-resonance frequency due to the decaying noble-gas polarisation [Eq. (2.136)] is described with the following equation

$$\frac{df_0}{dt} = -\frac{1}{T_1^n} f_0. \quad (4.3)$$

²It should be stressed that until now the mechanism of the cell magnetisation is not fully understood. Although the atomic cells are not exposed to the magnetic field strong enough for the magnetisation of the microscopic ferromagnetic contamination agents, sometimes they reveal non-zero magnetisation, which, in the case of the comagnetometers, may result in the lack of a stable compensation point. Such cases were reported in the literature [137] and a similar phenomenon have been observed in the Kraków comagnetometer.

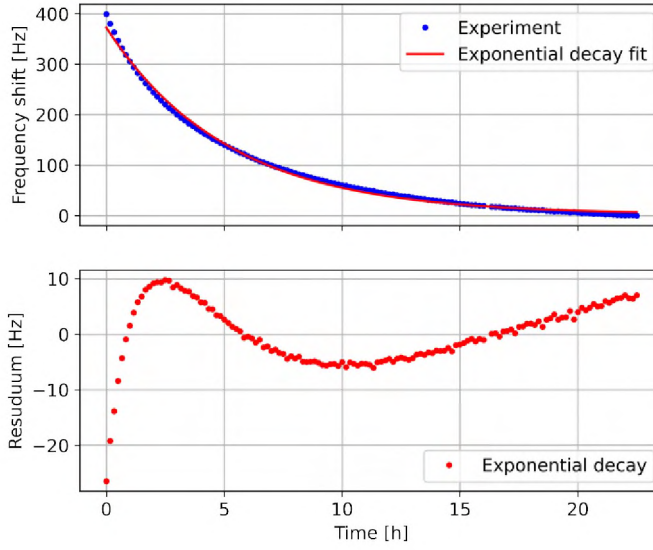


Figure 4.7: Decay of ^3He polarisation over time measured using the synchronously pumped SERF resonance. **Top:** The experimental data (blue dots) with results of the fitting with an exponential function (red line) defined in Eq. (4.4) and **Bottom:** residua of the fitting. The presented results are measured at 150 nT of the leading field. The noble-gas polarisation is expressed in terms of the frequency shift of the SERF resonance, which is measured with respect to the resonance observed in the measurement series.

This results in the exponential decay of the collisional frequency shift in time

$$f_0 = f_a e^{-\frac{1}{T_1} t} + f_c, \quad (4.4)$$

where f_a and f_c are constant parameters which depend on the initial conditions. However, the experimental results from the Kraków comagnetometer presented in Fig. 4.7 reveal non-exponential decay. The deviation from the exponential behaviour is clearly visible in the residua of the exponential fit Fig. 4.7 (bottom). The nature of this non-exponential decay can be identified by analysing the time derivative of the polarisation decay, which is presented in Fig. 4.8. According to Eq. (4.3), the time derivative of exponentially decaying polarisation has to be linear with respect to the polarisation amplitude. The experimental results reveal, however, a quadratic dependence of the derivative on the polarisation amplitude, which indicates the presence of self-induced relaxation of the noble-gas polarisation. In order to quantify this effect, the experimental results obtained for the derivative are fitted with the following phenomenological function

$$\frac{df}{dt} = -\frac{1}{T_1} [1 - \epsilon_f f] f, \quad (4.5)$$

where the ϵ_f is the parameter, which can be considered a characteristic of the self-interaction of the noble-gas polarisation in the system. The fitting results are presented in Fig. 4.8.

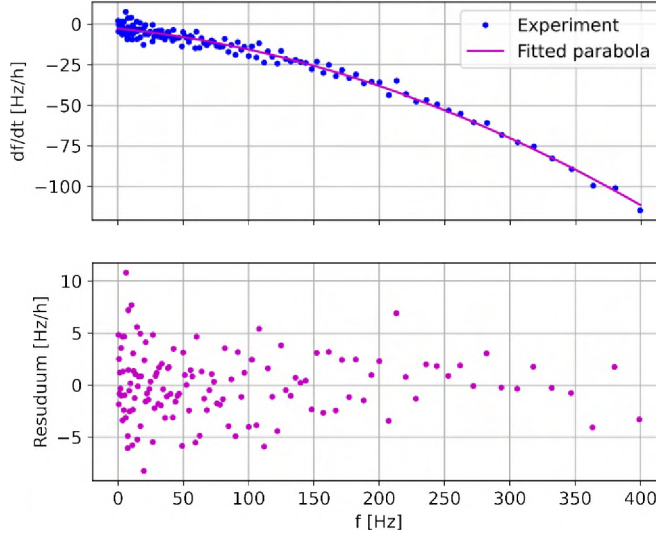


Figure 4.8: The time derivative of the alkali-metal Larmor-frequency shift induced by collisions with polarised ^3He atoms as a function of the collisional frequency shift. The results are obtained numerically from experimental data for the ^3He polarisation decay (Fig. 4.7). The solid line presents the result of fitting with Eq. (4.5).

4.3.2. LOCKING TO THE COMPENSATION POINT

Although the observation of the noble-gas polarisation in the constant magnetic field allows for gaining valuable information about the system, it is not always an optimal technique for characterising the comagnetometer system. In the case when the relaxation of the noble-gas polarisation is dominated by magnetic-field gradients, the applied magnetic field determines the longitudinal lifetime [Eq. (2.133)], and hence, the equilibrium polarisation of the noble gas [Eq. (2.154)]. In turn, the steady-state polarisation determines the compensation point [Eq. 2.156]. Ideally, one would like to know the value of the leading field, which provides the relaxation rate, such that the system is at the compensation point after it reaches equilibrium. However, in practice, prior knowledge of such a field is genuinely impossible. Therefore, a mismatch between the leading field used for pumping of the system and the compensation point leads to the instability of the latter (for more details see Sec. 4.4). Gaining information relevant to the conditions necessary for the achieving of the self-compensating regime requires measurement of the noble-gas polarisation while the system remains at the compensation point. This requires adjustments of the applied magnetic field with respect to the changes in the noble-gas polarisation level.

This section presents a convenient measurement technique allowing for the direct measurement of the effective magnetic field experienced by the alkali-metal atoms due to the noble-gas polarisation. Such a measurement provides the reference for a feedback loop to lock the system to the compensation point and hence operation of the comagnetometer in the self-compensating regime.

The presented method is based on the measurement of the comagnetometer re-

sponse to the harmonically modulated magnetic field applied along the secondary sensitivity axis of the device³ for different “detunings” from the compensation point. As shown in Ref. [138], the amplitude of the response to such a modulated field reaches its minimum when the external compensating field cancels the effective field generated by the noble-gas polarisation. Moreover, in this particular field, both in- and out-of-phase responses cross zero, which might be used as an error signal for the feedback system and allowing its locking to the point. As the compensation field [Eq. (2.156)] is mostly determined by the noble-gas polarisation, the measured zero crossing provides an approximate value of the compensation field, and therefore, the lock of the characteristic point provides a possibility for the online observation of the compensation-point changes [139]. Reference [138] presents the experimental tests of the technique along with a numerical analysis of the theoretical model for the system. Here, a complementary analytical analysis is provided. In particular, the results presented in Sec. 2.2.4 are analysed to get the analytical solution. The theory is later confirmed by the measurements results from Kraków Advanced GNOME comagnetometer. The experimental results include the implementation of the feedback-loop-based technique, allowing real-time tracking of the compensation point with time-dependent noble-gas polarisation, e.g., while the noble-gas polarisation builds up over time.

THEORETICAL BACKGROUND OF THE POLARISATION MEASUREMENT

This subsection considers the comagnetometer system in an arrangement similar to the one used in Sec. 2.2.4, where the atoms are polarised along the z -axis and probed along the x -axis. In such a case, the secondary sensitivity axis is parallel to the probed polarisation axis. In Sec. 2.2.4, the response of the comagnetometer to harmonic perturbations applied along the primary sensitivity axis (the y -axis) is theoretically derived [Eq. (2.187)]. However, the rotational symmetry of the problem allows obtaining of the result for the response to the perturbations applied along the insensitive axis by simply swapping the x - and y -axes in the solution. Hence, Eq. (2.188) leads to the following form of the comagnetometer output signal

$$S(t) = S_{in}(\omega)\sin(\omega t) + S_{out}(\omega)\cos(\omega t), \quad (4.6)$$

where S_{in} and S_{out} are in- and out-of-phase components of the comagnetometer signals. These components can be directly measured with the lock-in amplifier using the modulation frequency as the reference. From Eq. (2.188), one gets

$$S_{in}(\omega, \Delta_{B_z}) = g \operatorname{Re} \left[Y_{\perp}^{+}(\omega, \Delta_{B_z}) + Y_{\perp}^{-}(\omega, \Delta_{B_z}) \right], \quad (4.7)$$

$$S_{out}(\omega, \Delta_{B_z}) = g \operatorname{Im} \left[Y_{\perp}^{+}(\omega, \Delta_{B_z}) - Y_{\perp}^{-}(\omega, \Delta_{B_z}) \right], \quad (4.8)$$

where g denotes the calibration factor characterising the efficiency of the polarisation detection. This factor allows recalculation of the polarisation rotation into the electronic signal.

One can analyse the response of the comagnetometer with respect to the detuning from the compensation point Δ_{B_z} . While the frequency response to the rotating field at

³Here the definitions of the primary and secondary sensitivity axes introduced in Sec. 2.1.7 are followed.

different field detuning is quite complicated [Eq. (2.173)], since the main interest is in the measurements around the compensation point, one can utilise a number of approximations to simplify the expression. In particular, in the considered comagnetometer model, the relaxation rate of the noble gas is the smallest comagnetometer parameter given in frequency units [Eq. (2.170)]. This is especially the case for the ^3He -based system which possess long relaxation time (multiple hours). Hence, the approximation $R_n \approx 0$ leads only to insignificant relative deviations of the frequency response amplitude from the exact solution (estimated at a level below 1%). Noting that the field experienced by the nuclear polarisation is much larger than the detuning from the compensation point leads to the second approximation

$$\gamma_n \Delta_{B_z} + \omega_n \approx \omega_n, \quad (4.9)$$

which simplifies the denominator in Eq. (2.173). In turn, the frequency response to the rotating magnetic field is given by⁴

$$\mathcal{F}_{Y_\perp}^\pm(\omega) = -\gamma_F Y_z \frac{(\pm\omega + \gamma_n \Delta_{B_z})}{(\pm\omega + \omega_e + \gamma_F \Delta_{B_z} - i R_e)(\pm\omega + \omega_n) - \omega_e \omega_n}. \quad (4.10)$$

To consider the in- and out-of-phase components of the response [Eqs. (4.7) and (4.8)], it is convenient to split this expression into real and imaginary parts. For this, the above equation is rewritten in the following form

$$\begin{aligned} \mathcal{F}_{Y_\perp}^\pm(\omega) &= -Y_z \frac{\pm\omega + \gamma_n \Delta_{B_z}}{\pm\omega + \omega_n} \frac{\Delta_{B_z} + \frac{\omega^2 \pm \omega(\omega_e + \omega_n)}{\gamma_F(\pm\omega + \omega_n)} + i \frac{R_e}{\gamma_F}}{\left(\frac{R_e}{\gamma_F}\right)^2 + \left(\Delta_{B_z} + \frac{\omega^2 \pm \omega(\omega_e + \omega_n)}{\gamma_F(\pm\omega + \omega_n)}\right)^2} \\ &= -Y_z \mathcal{M}(\omega, \Delta_{B_z}) \mathcal{L}(\omega, \Delta_{B_z}). \end{aligned} \quad (4.11)$$

One can note that the field-detuning solution is defined with the linear function $\mathcal{M}(\omega, \Delta_{B_z})$, which is multiplied by a complex Lorentzian $\mathcal{L}(\omega, \Delta_{B_z})$ with a width R_e/γ_F centred at

$$\Delta_{B_z}^\pm = \frac{\omega^2 \pm \omega(\omega_e + \omega_n)}{\gamma_F(\pm\omega + \omega_n)}. \quad (4.12)$$

When the modulation frequency is much higher than the noble-gas Larmor frequency, $|\omega| \gg \omega_n$, one can simplify the function $\mathcal{M}(\omega, \Delta_{B_z})$ with a constant

$$\mathcal{M}(\omega, \Delta_{B_z}) = \frac{\pm\omega + \gamma_n \Delta_{B_z}}{\pm\omega + \omega_n} \xrightarrow{\omega \gg \omega_n} 1. \quad (4.13)$$

Then, the simplified expression for the frequency response of the comagnetometer has the following form

$$\mathcal{F}_{Y_\perp}^\pm(\omega) = -Y_z \frac{\Delta_{B_z} + \frac{\omega^2 \pm \omega(\omega_e + \omega_n)}{\gamma_F(\pm\omega + \omega_n)} + i \frac{R_e}{\gamma_F}}{\left(\frac{R_e}{\gamma_F}\right)^2 + \left(\Delta_{B_z} + \frac{\omega^2 \pm \omega(\omega_e + \omega_n)}{\gamma_F(\pm\omega + \omega_n)}\right)^2}. \quad (4.14)$$

⁴As this section considers the response to the magnetic field, one can set $\alpha_n = \alpha_e = 1$.

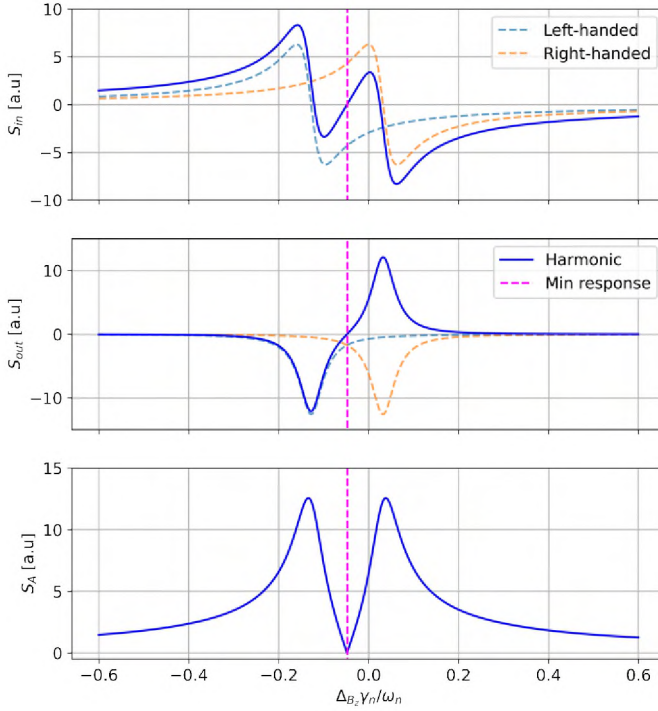


Figure 4.9: Magnetic-field response of the comagnetometer system as a function of the detuning from the compensation point plotted based on the analytical solution. The in- and out-of-phase components of the signal, calculated for the right- and left-handed circular perturbations, are obtained by taking a real and imaginary part of Eq. (4.14) respectively, while the responses to harmonic oscillations are obtained in Eqs. (4.7) and (4.8). The amplitude of the response has been calculated as $S_A = \sqrt{S_{in}^2 + S_{out}^2}$. The calculations are performed using a modulation frequency significantly exceeding the noble-gas Larmor frequency.

The analytical results for the response of the comagnetometer are presented in Fig. 4.9. As shown, they reproduce the main results of the original work, while the minimum of the response amplitude is close to the compensation point, and both in- and out-of-phase responses have a zero crossing at the same leading field. The presented plots also contain the curves for the Lorentzians presented in Eq. (4.14), which provide insights into the origin of the observed curves, as well as into their dependence on the parameters of the system. In particular, the broadening of the obtained curve with an increase in the modulation frequency can be explained by the change in the central detuning quantified in Eq. (4.20).

The solution for the comagnetometer response presented in Eq. (4.14) is used to obtain the field value at which the in- and out-of-phase components of the comagnetometer response cross zero, $S_{in} = 0$. Then, from Eq. (4.7), one gets the condition for zeroing the in-phase signal

$$\text{Re} \left[\Upsilon_{\perp}^{+}(\omega, \Delta B_z) \right] = -\text{Re} \left[\Upsilon_{\perp}^{-}(\omega, \Delta B_z) \right]. \quad (4.15)$$

This leads to the following solution for the detuning field

$$\Delta_0^{in} = -\frac{\omega^2 \omega_e}{\gamma_F(\omega^2 - \omega_n)} \xrightarrow{\omega \gg \omega_n} -\frac{\omega_e}{\gamma_F} = -\lambda M^e \Upsilon_z. \quad (4.16)$$

From Eq. (4.8), one gets the condition for zeroing of the out-of-phase response

$$\text{Im} \left[\Upsilon_{\perp}^+(\omega, \Delta_{B_z}) \right] = \text{Im} \left[\Upsilon_{\perp}^-(\omega, \Delta_{B_z}) \right], \quad (4.17)$$

which leads to

$$\Delta_0^{out} = -\frac{\omega_e + \omega_n}{\gamma} \approx -\frac{\omega_e}{\gamma_F} = -\lambda M^e \Upsilon_z, \quad (4.18)$$

where it was assumed that $\omega_e \gg \omega_n$. This is a reasonable assumption as, in usual experimental conditions, the alkali-metal spins Larmor frequency at the compensation point is at least an order of magnitude higher than in the case of the noble gas.

Taking into account the definition of the detuning from the compensation point [Eq. (2.156)], one gets the value of the magnetic field necessary for zeroing the comagnetometer response

$$B_0 = B_c + \lambda M^e \Upsilon_z = -\lambda M^n \zeta_z. \quad (4.19)$$

This confirms the prediction of the original work as, according to the analytical solution, the signal is zeroed when the applied leading field compensates for the field generated by the nuclear polarisation. However, the analytical solution is more insightful since it gives a clue on the potential sources of the systematic effects, which may affect the precision of the noble-gas polarisation measurements. The most obvious source of the systematic effect comes from the choice of the modulation frequency. If it is too low then, due to the linear background arising from Eq. (4.13), the curve presented in Fig. 4.9 becomes asymmetric. Moreover, if the modulation frequency is not high enough, the zero-crossing is shifted, as shown in Eq. (4.16).

4.3.3. EXPERIMENTAL MEASUREMENT OF THE NOBLE-GAS POLARISATION

The theoretically predicted magnetic-field response curves (Fig. 4.20), which allow the measurement of the magnetisation of the noble gas in the Kraków Advanced GNOME comagnetometer, are measured using a double-stage lock-in detection. The first demodulation is performed on the first harmonic of the parametric modulation (Sec. 2.2.14). The output of the device is demodulated for the second time on the frequency of the magnetic-field modulation applied along the insensitive y -axis.

The experimental setup used for this measurement is identical to the main setup used for the double-sensitivity-axis operation of the comagnetometer (Fig. 3.1). The only difference is that in this case, the second lock-in amplifier is used independently from the first one, and its internal generator has been used for the modulation of the magnetic field along y . The field was modulated with the 40-Hz sine wave of an amplitude of 1 pT. The detuning from the compensation point was changed by the scanning of the current generated by the current supply. For each value of the scanned magnetic field, the readout from the second lock-in has been recorded after the signal reached the steady state.

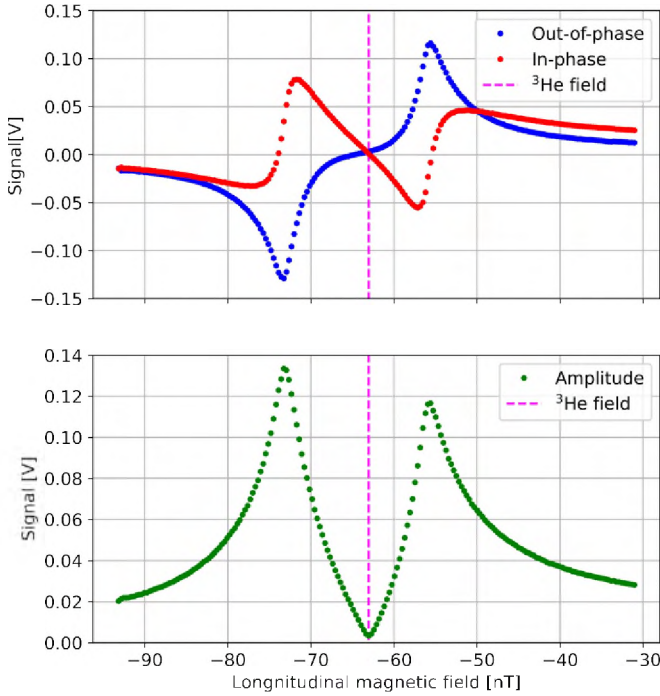


Figure 4.10: Demodulated response of the Advanced Kraków comagnetometer to the sine wave modulation as a function of the leading field. The top plot shows the in-phase (blue) and out-of-phase (red) components of the signal. The bottom plot shows the amplitude of the response. The vertical dashed lines on the plots mark the field for which the minimum amplitude of the response is observed and indicate the field generated by the noble-gas polarisation. The results are measured for a 1-pT sine-wave modulated field of a frequency of 40 Hz.

The representative measurement results are presented in Fig. 4.10. These results are in agreement with the theoretical predictions from Sec. 4.3.2. Specifically, the in- and out-of-phase components of the response reproduce a distinct pattern arising from a superposition of two complex Lorentzians. The difference between the theoretical in-phase response from Fig. 4.9 and the experimental curve can be attributed to the additional phase shift introduced by the lock-in detection and/or the direction of the compensating field⁵. The slight asymmetry of the curve visible in the result is probably caused by the misalignment between the probe-beam propagation direction and the direction of the B_y magnetic field.

Worth noticing is that both in-phase and out-of-phase components are linear near the zero crossing. This allows the use of one of these curves as an error signal for locking the leading field to the noble-gas polarisation with the feedback-loop system. Such a feedback loop allows monitoring of the noble-gas polarisation in real-time while the system is operated at conditions which are close to the compensation point. This can be

⁵The change of the sign in the compensating field leads to the similar effect as $\pi/2$ change in phase of the signal.

used for the *in situ* compensation of the magnetic-field gradients in the comagnetometer system [139] (for more detail also see Sec. 4.4). The choice of the component/curve for the feedback system is arbitrary and the suitability of the particular curve depends on the system and modulation parameters.

The scheme of the feedback loop used for tracking the noble-gas polarisation in the Advanced GNOME comagnetometer is present in Fig. 4.11. In this case, one can use the in-phase signal as the error signal for the feedback system. The error signal is sent to the NI 6221 DAQ card, which works as an input interface for the software PID system programmed in the LabView software environment. The output of the virtual PID system controls the current source such that the error signal remains at zero. In such a configuration, the value of the leading field is, in a sense, equal to the value of the effective field from the ^3He polarisation at any moment of time when the feedback loop is active.

It is worth noting that under typical operation conditions of the comagnetometer with continuous pumping, the polarisation of the alkali-metal atoms remains at a constant level. Moreover, the shape of the curves does not depend significantly on the nuclear polarisation, as from Eq. (4.12)

$$\Delta_{B_z}^{\pm} \xrightarrow{\omega \gg \omega_n} \frac{1}{\gamma_F} (\pm\omega + \omega_e + \omega_n) \xrightarrow{\omega_e \gg \omega_n} \frac{1}{\gamma_F} (\pm\omega + \omega_e), \quad (4.20)$$

and therefore the frequency response to the rotating fields [Eq. (4.14)] and inherently the in- and out-of-phase components of the response [Eqs. (4.7) and (4.8)] are approximately independent of the noble-gas polarisation level. Since the shape of the error signal does not change significantly with the leading field, it is possible to lock the system at an arbitrary point of the error curve using the constant offset to the set point of the PID system. In particular, one can take into account the magnetic field corresponding to the effective field from the electron polarisation and lock the system at the compensation point. Thereby, this method allows for real-time tracking of the compensation point. The results of the long-term measurement of the compensation point during the build-up of the nuclear polarisation are presented in Fig. 4.12. One can note that the polarisation increases for about 10 h from the start of the measurements up to the point where it reaches its steady level.

The presented technique is useful in both developments of the comagnetometer setup and the long-term stabilisation of the compensation point, as the system can detect non-magnetic couplings even when the compensation point is locked. However, in practice, the feedback loop may add additional noise to the system, which may deteriorate its sensitivity, and when improperly tuned, the PID feedback may interfere with the non-magnetic response of the system. Therefore, the trade-off between sensitivity and the stability of the system related to the feedback loop should be carefully considered for a particular application.

4.4. OBSERVATION OF STABLE COMPENSATION POINT

One of the main issues with the long-term continuous operation of the comagnetometer arises from the fact that most of the comagnetometer systems require frequent re-setting of the compensation point and sometimes repumping of the noble-gas polarisation. Both of these operations interrupt the operation of the comagnetometer. The

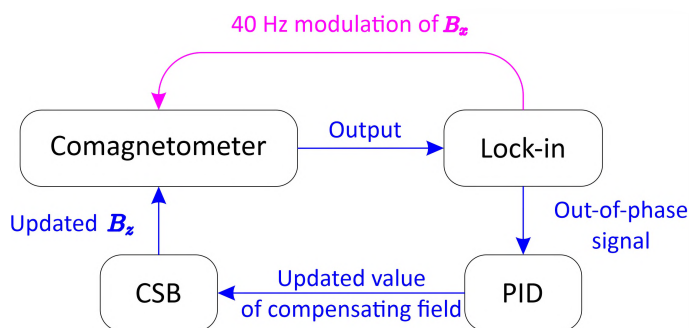


Figure 4.11: Schematics of the feedback-loop system used for locking to the compensation point in the Advanced Kraków comagnetometer. The comagnetometer output signal is obtained from the lock-in amplifier demodulating the photodetector signal at the frequency of the parametric modulation (Fig. 4.10). The signal detected via parametric modulation is then sent to the second lock-in amplifier, demodulating the signal at the frequency of the insensitive axis modulation. This modulating signal is generated with the internal generator of the second lock-in amplifier. The out-of-phase signal of the lock-in amplifier is sent to the virtual PID system (programmed using Labview) via a DAQ card. The output signal of the PID system controls the current source, which drives the coil, generating the leading magnetic field.

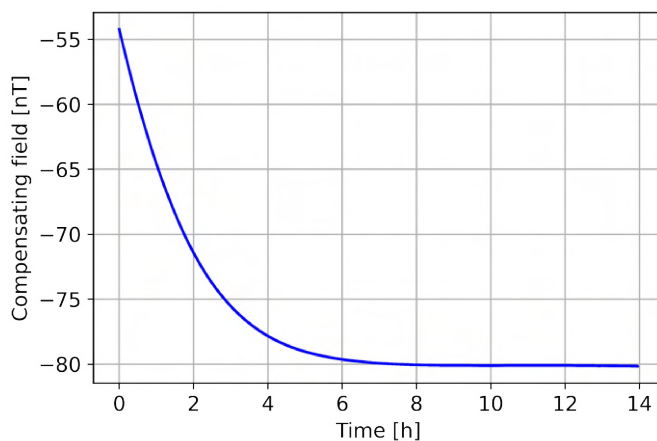


Figure 4.12: Time dependence of the compensation point observed during the noble-gas polarisation build-up. The measurement was performed by locking of the compensation point using the feedback loop presented in Fig. 4.11. The signal was measured in Kraków magnetometer.

necessity of resetting the compensation field can be partially alleviated by locking the compensation point with the method described in Sec. 4.3.2, which keeps the system operational while tracking the compensation point. However, it might not be sufficient in the case when the compensation point is fundamentally unstable. Such a situation is possible when relaxation of the noble-gas polarisation is dominated by relaxation due to the magnetic-field gradients. It is shown in Eq. (2.133) that this relaxation channel depends on the applied magnetic field, which is a fundamental reason for the instability of the compensation point. The issue was studied in Ref. [139]. This section discusses the main results of the work and presents the experimental results of magnetic-field-gradient compensation in the Advanced Kraków comagnetometer.

4.4.1. CONDITIONS FOR THE STABLE COMPENSATION POINT

For a better understanding of the problem, one can consider the comagnetometer, which is exposed to the constant magnetic field much larger than the compensation field

$$B_z(t=0) \gg B_c(t=0). \quad (4.21)$$

When the system is in a steady state, the nuclear polarisation is described with Eq. (2.154). In order to operate the comagnetometer in the self-compensation regime, the leading field is first set to the actual compensation point and then one should wait until the system reaches the steady state again. However, since the magnetic field decreases, the relaxation rate increases, and the nuclear polarisation becomes smaller than initially, which results in a smaller compensation-field value. Thereby, the system is again outside of the compensation point. Resetting the system repeats the whole cycle. Ultimately, this process leads to the complete decay of the noble-gas polarisation. The presented example shows that the operation of the system requires either periodic repumping of the noble-gas polarisation at a higher magnetic field or optimisation of the system to the degree where, in a given magnetic field, it is possible to create such a nuclear polarisation that the system operates at the compensation point. As the first option is not suitable for the GNOME measurements, the second approach to the problem is taken in this work.

The stability of the compensation point is a result of the interplay between three main factors: the pumping efficiency of the noble gas, the relaxation of the noble gas due to the magnetic-field gradients, and the amplitude of the effective field created by the noble-gas polarisation. The effective addressing of the stability issue requires a deeper understanding of the relation between these three factors. One can get an insight into this relation by combining Eqs. (2.133), (2.154), and (2.156). One starts by setting the magnetic field to the compensation point

$$B_z = B_c. \quad (4.22)$$

From Eq. (2.133), the relaxation rate of the noble gas polarisation is

$$\frac{1}{T_1} = D_n \frac{|\nabla B_\perp|^2}{B_c^2}. \quad (4.23)$$

Next, the compensation point [Eq. (2.156)] is approximated with the effective magnetic

field from the noble-gas polarisation⁶

$$B_c \approx -\lambda M^n \zeta_z, \quad (4.24)$$

which allows the definition of the longitudinal polarisation through the observed compensation point

$$\zeta_z = -\frac{B_c}{\lambda M^n}. \quad (4.25)$$

Substitution of Eqs. (4.23) and (4.25) into Eq. (2.154) leads to

$$-\frac{B_c}{\lambda M^n} = \frac{R_{en} \Upsilon_z}{R_{en} + D_n \frac{|\nabla B_\perp|^2}{B_c^2}}, \quad (4.26)$$

which relates to all of the considered parameters. One can rewrite the equation in a different form

$$B_c^2 + \lambda M^n \Upsilon_z B_c + \frac{D_n}{R_{en}} |\nabla B_z|^2 = 0, \quad (4.27)$$

which is a quadratic equation with respect to B_c . The solution of this equation, and therefore the condition for a stable compensation point, is achieved when

$$(\lambda M^n \Upsilon_z)^2 - 4 \frac{D_n}{R_{en}} |\nabla B_\perp|^2 \geq 0, \quad (4.28)$$

which sets the following limit on the gradient of the magnetic field

$$|\nabla B_\perp| \leq \lambda M^n \Upsilon_z \sqrt{\frac{R_{en}}{4 D_n}}. \quad (4.29)$$

The maximum value of the transverse gradient is effectively set by the polarisation of the noble-gas atoms, the concentration of the noble gas,⁷ and the rate of spin-exchange between the alkali-metal polarisation and the noble-gas polarisation. In principle, this enables optimisation of the experimental conditions to improve the comagnetometer tolerance to the transverse magnetic-field gradients. Specifically, it is beneficial to have a high concentration of the noble gas, as the relatively low concentration of the noble gas leads to a more stringent limit of the tolerated magnetic-field gradient. The spin-exchange rate can be increased by raising the alkali-metal concentration, e.g., by increasing the temperature of the atomic cell. However, for a given set of experimental parameters, the gradient has to be small enough for the stable compensation point to exist. This fact gains the importance of the precise demagnetisation of the experimental setup. However, it is not always possible to demagnetise the experimental setup at a level sufficient for the observation of the stable compensation point. In such a case, the magnetic-field gradients have to be cancelled precisely with a set of dedicated gradient coils. The method for such compensation is described in the next section.

⁶At normal operation of the comagnetometer, the noble-gas magnetisation is much larger than the alkali-metal magnetisation.

⁷The limit for the gradient scales as $n_n^{\frac{3}{2}}$ since $M^n \sim n_n$ and $D_n \sim n_n^{-1}$.

4.4.2. PRECISE CANCELLATION OF THE MAGNETIC-FIELD GRADIENTS

In the research presented in this thesis, the gradient cancellation method, originally proposed in Ref. [139], is used. The method is based on the continuous observation of noble-gas polarisation decay with the lock of the compensation point (Sec. 4.3.2) under different external magnetic-field gradients. To cancel the gradients, various gradients are applied by changing the current in the gradient coils and finding the conditions for the slowest decay of the polarisation.⁸

The gradient optimisation utilises the identical experimental setup as described in Sec. 4.3.2. Before the cancellation of the gradients, the atoms are polarised at some relatively high constant magnetic field.⁹ The optimisation of the gradient generated by a given coil starts with the lock to the compensation point (Sec. 4.3.2). With the lock to the compensation point, the current on one of the gradient coils is scanned, and each value is on for 90 s (Fig. 4.13). The value of the compensation point is observed over time, and each 90 s long segment is analysed to get the slope of the decay curve, indicating the time of the decay or pumping.

The slope analysis is performed by calculating the time derivative of the polarisation-decay signal [Fig. 4.13 (bottom)]. Within a time sequence corresponding to a current value, one can notice two distinct parts of the system response. At the beginning of each time period, the time derivative suddenly changes due to the change of the field gradient, and then it stabilises at the constant value indicating a linear decay of the polarisation. The sudden change of the polarisation is related to the delayed response of the feedback loop, while the linear decay is the result of the short-time observation of exponentially decaying signal. The latter relation can be obtained from Eq. (2.150) considering the evolution of the longitudinal component of the noble-gas polarisation

$$\frac{d\zeta_z}{dt} = -\left(R_{en} + \frac{1}{T_1}\right)\zeta_z. \quad (4.30)$$

Since $\zeta_z \sim \zeta_0 e^{-(R_{en}+1/T_1)t}$, for $t \ll T_1$, one gets

$$\frac{d\zeta_z}{dt} \approx -\left(R_{en} + \frac{1}{T_1}\right)e^{-(R_{en}+1/T_1)t} \approx -\left(R_{en} + \frac{1}{T_1}\right), \quad (4.31)$$

which shows that in the first order, the decay signal is linear. Equation 4.31 sets the experimental requirement that, over the observation period, the leading-field change is small, such that the relaxation rate $1/T_1$ can be considered to be a constant.

The slope for each gradient-coil current is estimated by averaging the derivative over the second half of each measured period, such that the gradient within the considered time period is constant. An example of such estimation is presented in Fig. 4.14. The presented result indicates a second-order dependence of the slope on the applied gradient, which is consistent with the theoretical predictions of Eq. (4.31) [the relaxation rate depends on the square of the linear-gradient amplitude, Eq. (2.133)]. To identify the

⁸Similar measurements may be performed by pumping the system and finding the conditions corresponding to the fastest buildup of the polarisation.

⁹The choice of the field is arbitrary, however, nonzero polarisation is needed for the measurements. Since higher leading fields allow for a higher polarisation level (due to the suppression of the relaxation related to the magnetic-field gradients [Eq. (2.133)]) stronger fields are preferential for the measurements.

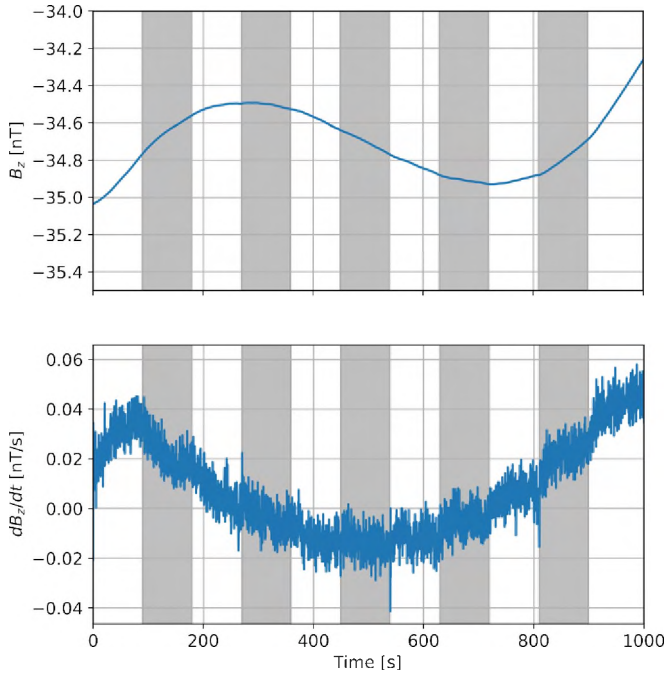


Figure 4.13: Experimental measurements of the compensation point changes in time (top) for the different values of the current applied for the dB_z/dz gradient coil. Periods with different values of the gradient are denoted with white and grey colours of the plot background. (Bottom) The time derivative of the data from the top plot.

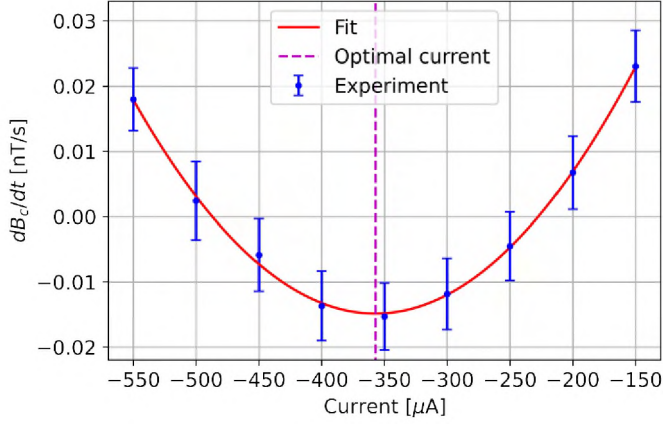


Figure 4.14: Experimental relation between the current in the dB_z/dz coil and the slope of the compensation-point change. The blue dots present the experimental data obtained by averaging the time derivative of the compensation point change in time (Fig. 4.13). The error bars are estimated with a standard deviation calculated from the averaged data. The red line denotes the curve fitted to the experimental data [with a parabola parameterised in Eq. (4.32)]. The vertical dashed line denotes the optimal value of the current determined from the fitting result.

current for which the minimal decay has been observed, the experimental data are fitted with the following model

$$S(J) = a(J - J_0)^2 + c, \quad (4.32)$$

where a , J_0 and c are the fitting parameter. J_0 corresponds to the current, for which the optimal behaviour (the smallest slope of the decay curve) is observed.

The Advanced GNOME comagnetometers are equipped with a set of gradient coils, enabling the control over five independent components of the first-order magnetic-field gradients (Sec. 3.3). The optimisation of the gradient is done subsequently for all first-order gradient coils available in the system. After each measurement, the current in the tested coil is set to the optimal value and the next gradient is scanned.¹⁰

4.5. ZEROING OF THE RESIDUAL MAGNETIC FIELD

In the discussed experiment, the control over the magnetic-field environment includes precise cancellation of the residual magnetic fields, particularly the ones originating from the residual magnetisation of the magnetic shield. For this purpose, the Kraków comagnetometer utilises the alkali-metal magnetometer realised in the comagnetometer cell under the low ^3He polarisation limit. This limit is determined by decreasing the pump-beam power to the lowest value that still allows observation of the alkali-metal magnetic resonances. As residual fields have a large effect on the performance of the comagnetometer, it should be stressed that the residual-field zeroing procedure must be

¹⁰From the author's experience, for better stability of the compensation point, the gradient optimisation has to be performed after any significant change in the system (e.g., opening of the magnetic shield, cooling and heating of the atomic cell, etc.).

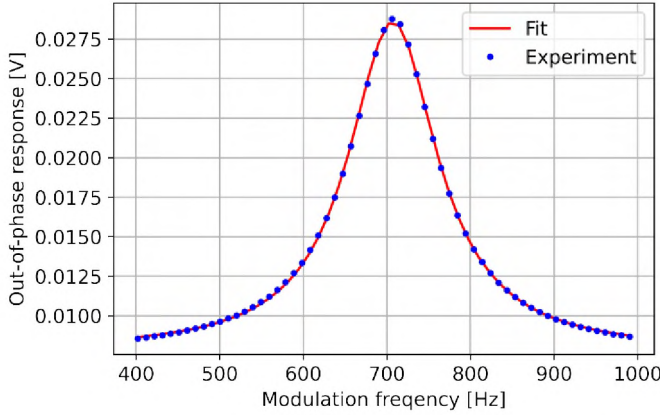


Figure 4.15: SERF resonance observed in a finite magnetic field versus the modulation frequency. The blue dots represent the experimentally measured out-of-phase component of the comagnetometer signal, which is fitted with the function from Eq. (4.33) (red line). The signal is used to zero the residual magnetic field.

performed prior to the polarisation of the noble gas and actual comagnetometer measurements.

The zeroing procedure is based on the measurements of the SERF resonances in a finite leading field.¹¹ The resonances are used to determine offsets to the applied field produced by the magnetic-field coils generating homogeneous fields. The compensation is based on the fact that in the case of the applied field cancelling the residual field in a given direction, the resonance position in a frequency domain is minimal.

In the experiment, the magnetic resonances are measured with the constant magnetic field applied along the z -axis and with the modulation of the magnetic field along the sensitivity axis x . The frequency of the modulated field is scanned over a certain range around the Larmor frequency of the alkali-metal polarisation. The modulation signal is generated with an internal generator of the lock-in amplifier successively used for demodulation of the detected signal (Lockin 1 in Fig. 3.1).

The representative out-of-phase signal measured in the experiment is presented in Fig. 4.15. The obtained resonance curve is fitted with the function based on Eq. (2.122)

$$S_{out}(f) = a \operatorname{Im} \left[\frac{f - f_0 + i\delta_f}{(f - f_0)^2 + \delta_f^2} \right] + c, \quad (4.33)$$

where a , f_0 , δ_f , and c are fitting parameters, f is the modulation frequency, and $S_{out}(f)$ is the out-of-phase signal from the lock-in amplifier. The parameter f_0 corresponds to the resonance frequency of the alkali-metal polarisation. The results of the fitting for the resonance are presented in Fig. 4.15.

The dependence of the observed Larmor frequency on the current applied to the coils is present in Fig. 4.16. The B_z results follow the dependence predicted by Eq. (2.122).

¹¹Such measurements are discussed in Sec. 2.1.6.

The observed Larmor frequency is linearly proportional to the absolute value of the applied current J_z . Hence, the measurement is insensitive to the spin precession direction. To identify the residual field value in the z -direction, the experimental results are fitted with the following function

$$f_0(J_z) = a|J_z - J_{0z}| + c, \quad (4.34)$$

where a , J_{0z} , and c are fitting parameters and J_{0z} corresponds to the current generating the compensating field. The results of the fitting are presented in Fig. 4.16 (top). Further measurements for the transverse magnetic fields are conducted in the regime where the transverse magnetic fields are much smaller than the leading field. Hence, one can consider the transverse magnetic field as a second-order correction to the value of the total magnetic field.¹² Therefore, the observed Larmor frequency for the B_x and B_y coils are fitted with the following function

$$f_0(J_{x,y}) = a\sqrt{(J_{x,y} - J_{0\ x,y})^2 + c}, \quad (4.35)$$

where a , $J_{0\ x,y}$, and c are fitting parameters, and $J_{0\ x,y}$ corresponds to the current generating compensating magnetic field in a given direction.

The presented method provides results similar to the ones obtained with a conventional method of zeroing perpendicular magnetic fields [101]. The conventional method is based on the observation of the steady-state response of the comagnetometer, operated at high noble-gas polarisation near the compensation point, under different transverse and leading fields. The main difference between the methods lies in the presence of the high compensating field and the application of the high-power pump light in the conventional method. It should be stressed that the high-power pump light may affect the resonance position when there is a misalignment between the probe and pump beams (via the AC Stark effect). Hence, zeroing the residual fields with the presented method allows at least partial separation of the beam-misalignment problem from the field zeroing. On the other hand, zeroing the transverse fields near the compensation point enables compensation for the possible misalignment in the orientation of the coils, e.g., when the additional transverse field is generated with the B_z coil. The compensation for the misalignment of the magnetic-field coils with the method proposed in this work, the approximate value of the target compensation point has to be known prior to the measurements. This allows the performance of the zeroing procedure with a leading field value close to the target compensation point, which was the case for the Kraków Advanced GNOME comagnetometer.

4.6. SUMMARY OF THE CHAPTER

This chapter discussed a collection of various measurement techniques and tests, which, in the opinion of the author of this work, were particularly useful over the course of the development of the Kraków Advanced GNOME comagnetometer. The selected topics for the discussion cover the elements of the author's experimental work, such that either employed approaches which are not typical in the context of the self-compensating comagnetometers or which, from the author's experience, might be problematic for exper-

¹²More information on this regime one can find in Sec. 5.4.

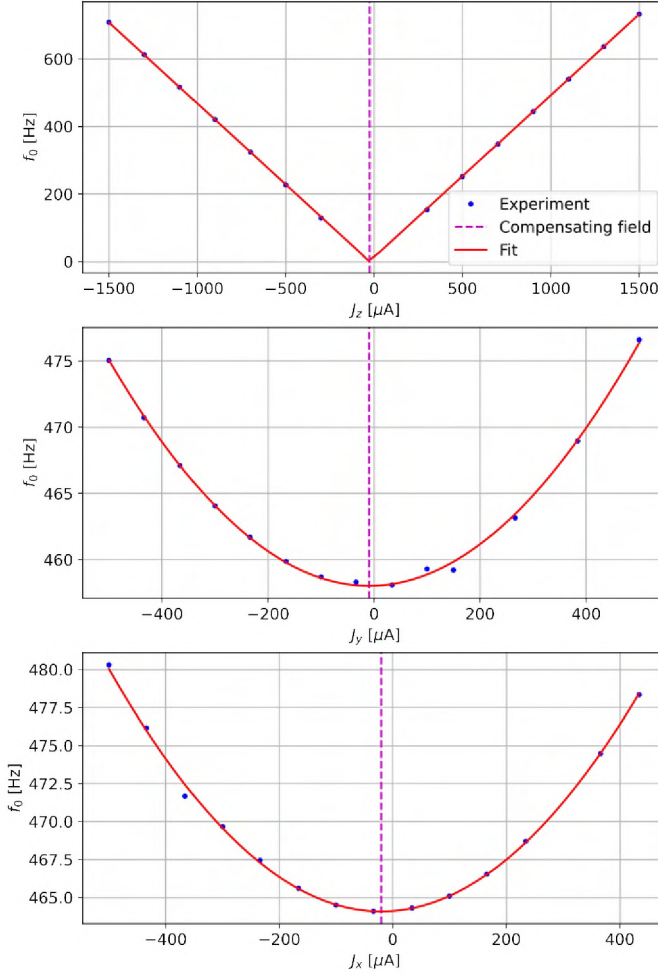


Figure 4.16: Resonance frequency of the SERF resonance f_0 observed in alkali-metal atoms versus the electric current applied to the magnetic-field coils, generating a homogenous magnetic field. The top plot presents results for the B_z coil (blue dots) fitted with the model from Eq. (4.34) (red solid line). The middle and bottom plots present similar results for the B_x and B_y coils, which are fitted with the model from Eq. (4.35).

imentalists with a lack of previous experience in the comagnetometry. Hence, the discussion presented in this chapter is far from being comprehensive. For a more thorough discussion of the technical aspects of the comagnetometer development, the following references are recommended [81, 101, 140, 141].

5

CALIBRATION OF THE COMAGNETOMETERS

Precise calibration of the comagnetometer has principle importance for its applications in navigation (gyroscope) and fundamental-physics tests. In the latter case, the calibration of the sensor is complicated by the unavailability of a controllable application of the targeted perturbation for a direct study of the system response. This naturally raises the question about possible techniques that allow for the indirect calibration of the system response, e.g., by analysis of its response to other known controllable perturbations.

This chapter provides a detailed discussion of the methods for the comagnetometer calibration in the context of the GNOME experiment. Section 5.1 discusses the choice of the perturbation used for the calibration reference. Then, a conventionally used method for the calibration of the comagnetometer is considered in Sec. 5.2. The Advanced GNOME calibration method details are presented in Sec. 5.3. Next, the results of the comparison of the proposed indirect calibration method with the response to the rotation of the system are presented (Sec. 5.3.4). This is followed by the description of the magnetic-coils calibration with respect to the gyromagnetic ratio of ^3He presented in Sec. 5.4. The concluding section (Sec. 5.5) provides a summary of the presented calibration routine in the context of its application in the GNOME experiment.

5.1. CHOOSING THE CALIBRATION REFERENCE

The indirect technique employed for the comagnetometer calibration utilises the system response to a well-defined and properly controlled perturbation, which is distinct from the perturbation under investigation. Such a method is instrumental in acquiring valuable insights about the system, enabling accurate predictions of its response to such perturbation as exotic spin couplings. For the comagnetometer, there are three primary channels of controllable perturbation: light (through the AC Stark effect), rotation (through the gyroscopic effect), and magnetic field (through the Zeeman effect).

The **AC Stark effect** perturbs only the electron polarisation of atoms. Therefore, it has

a similar effect on the system as a pseudomagnetic electron perturbation. This similarity can be utilised for the calibration of the comagnetometer response to the electron perturbation. However, the main targets of the GNOME experiment are the exotic nuclear spin couplings [41].

Rotation is another effect that can be used for calibration. As shown in Table 2.6, rotation mostly perturbs nuclear spins, and hence it imposes a similar effect on the comagnetometer as hypothesized non-magnetic nuclear perturbations. With this respect, rotation allows one to gain insight into its response to non-magnetic nuclear perturbation. However, the implementation of the high-quality rotation system significantly complicates the experimental setup and increases its cost. One can also use the Earth's rotation as the source of perturbation [133]. The change in the relative orientation between the Earth's rotation axis and the comagnetometer sensitivity axis allows one to control the effect of the Earth's rotation on spins. Since the rotation period of the Earth is well known, it makes an excellent calibration reference. However, the rotation calibration of the comagnetometer is not ideal in searches for exotic coupling nucleon spins, as it couples to the total nuclear spin, and hence, it does not take into account the nuclear spin content of the atoms utilised in the sensor. Finally, the calibration with respect to the Earth's rotation is only valid for the low-frequency perturbations.

The calibration methods based on the AC Stark effect or gyroscopic effect possess an important feature of independent addressing of electron and nuclear polarisations. This is in contrast to the **magnetic-field perturbation** since, in this case, the interaction arises simultaneously with electron and nuclear polarisations. In this context, utilisation of the magnetic field for the comagnetometer calibration requires a good theoretical understanding of the system response to a targeted exotic spin coupling. At the same time, the application of a magnetic field for calibration has a number of practical advantages. The most important one is the simplicity of its implementation, as the magnetic-field coils are an essential part of the experimental setup, and hence using them for the calibration does not require any modifications of the comagnetometer setup. The simplicity of the magnetic-field manipulation provides flexibility in designing calibration routines and their optimisation for various regimes of the comagnetometer operation (e.g., double-sensitivity-axis operation) without the need for changes in the experimental setup. Moreover, the magnetic-field calibration allows the use of a single calibration routine to calibrate the response to all-type non-magnetic spin-couplings applied along an arbitrary direction. In addition, it provides information about actual detuning from the compensation point (one can find more details in Sec. 5.3), hence minimising the time necessary for the maintenance of the magnetometer. The completeness and minimal duration of the calibration routine with a magnetic field makes it an optimal choice for the GNOME experiment.

5.2. STANDARD APPROACH

This section presents the conventional method for calibrating the comagnetometer, originally proposed in Ref. [101]. It provides a determination of a calibration factor for the DC response of the comagnetometer, which is denoted as k_{DC} . This factor is used for the conversion of the low-frequency component of the output voltage signal S_{out} into

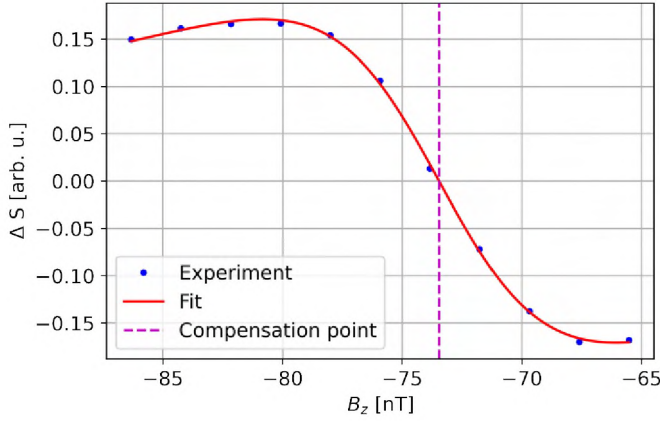


Figure 5.1: Measurements of the difference in the steady-state signal for the alternating magnetic field applied along the sensitivity axis $\mathbf{x}$ as a function of the leading field B_z obtained with the Kraków comagnetometer. The observed output signal is a first-harmonic signal measured at the parametric-modulation frequency. The experimental data (blue dots) are fitted (red solid line) with the model from Eq. (5.7), which allowed the determination of the compensation point (magenta dashed line) and the calibration factor for a low-frequency response of the comagnetometer [Eq. (5.8)].

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the signal measured in the effective magnetic-field units b_G

$$b_G = k_{DC} S_{out}. \quad (5.1)$$

The conventional calibration relies on measurements of the difference between the steady-state response of the comagnetometer to a magnetic field applied along the considered sensitivity axis in two opposite directions. Such comparison is repeated for different detuning from the compensation point. One can show using Eq. (2.183) that, for a small detuning from the compensation point, alternating the magnetic-field orientation would lead to the difference in the measured projection of the alkali-metal polarisation, which can be approximated with the following expression

$$\begin{aligned} \Delta Y &\approx -\frac{\gamma_n Y_z B_m}{\omega_n} \operatorname{Im} \left[\frac{\Delta B_z}{\Delta B_z - i R_e / \gamma_F} \right] \\ &= -\frac{\gamma_n Y_z B_m}{\omega_n} \frac{\Delta B_z R_e / \gamma_F}{\Delta B_z^2 + (R_e / \gamma_F)^2}, \end{aligned} \quad (5.2)$$

where B_m is the total change of the modulated magnetic field between two compared

steady states. The solution is obtained based on the following assumptions¹

$$\Delta B_z \ll 1, \quad (5.3)$$

$$R_n \approx 0, \quad (5.4)$$

$$\omega_n \ll \omega_e, \quad (5.5)$$

$$\gamma_F \gg \gamma_n. \quad (5.6)$$

From Eq. (5.2), the difference between the steady states when alternating the transverse field, measured as a function of the leading field, is described with the dispersive Lorentzian centred at the compensation point. Hence, this approach is often used to experimentally determine the compensation point.

The result from Eq. (5.2) is in agreement with the experimental data obtained with the Kraków comagnetometer (Fig. 5.1) and fitted with the following model

$$\Delta S(B_z) = a \frac{(B_c - B_z)\Gamma_B}{(B_c - B_z)^2 + \Gamma_B^2} + c, \quad (5.7)$$

where a , B_c , c , and Γ_B are fitting parameters and B_c corresponds to the compensation point.

Comparison of Eq. (2.184) with Eq. (5.2) and with the model given by Eq. (5.7) allows one to define the experimental value of the calibration factor given in Eq. (5.1)

$$k_{DC} = \frac{\Gamma_B}{a} \frac{\gamma_n}{\omega_n} B_m \approx \frac{\Gamma_B}{a} \frac{B_m}{B_c}, \quad (5.8)$$

where Eqs. (2.175) and (2.175) were used and the approximation $\lambda M^n \zeta_z \approx B_c$ was applied.

The conventional calibration method yields reliable data in a low-frequency range, i.e., for the frequency lower than the noble-gas Larmor frequency ω_L at the compensation point [see Eq. (2.175)]. This is in agreement with the theoretical result for the response of the comagnetometer to non-magnetic nuclear perturbation [Eq. (2.189)], which is plotted in Fig. 5.2. One can note that the conventional calibration provides a valid lower-bound approximation of the comagnetometer response at the considered frequency range. However, it may underestimate the response amplitude at frequencies comparable to ω_L . Such an underestimation ultimately leads to the underestimation of the comagnetometer sensitivity at frequencies near ω_L . The scale of the underestimation would depend on parameters of the system defining the frequency response [Eq. (2.173)]. The theoretical result also confirms that the conventional calibration factor quickly overestimates response amplitude for frequencies above ω_L , hence making the calibration invalid for this frequency range.

It is noteworthy that the conventional calibration method may not be suitable for the recalibration of the comagnetometer in long-term measurements when the relaxation of the noble-gas polarisation is dominated by the magnetic-field gradients. The

¹The presented assumptions are typically valid for the ³He-based comagnetometers due to their long coherence time. It can have limited applicability for systems based on ²¹Ne and Xe atoms due to their shorter coherence times.

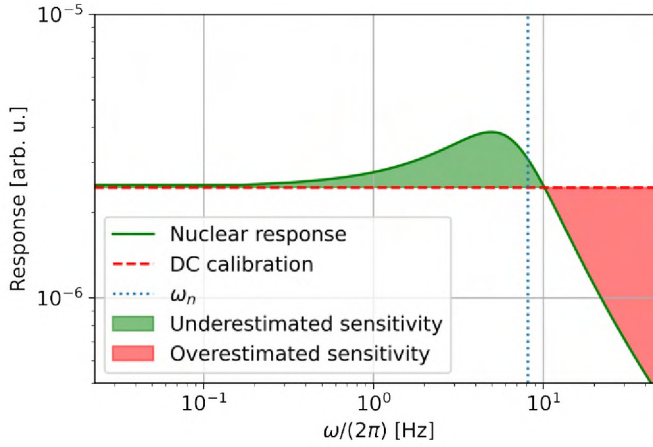


Figure 5.2: The comparison of the conventional calibration with the frequency response to non-magnetic nuclear perturbations ($\alpha_e = 0$) obtained from the theoretical simulation of the comagnetometer response. The response of the comagnetometer predicted by Eq. (2.189) is denoted with a solid green line. The dashed red line denotes the DC response, which is equivalent to the information provided by the conventional calibration method [Eq. (5.8)]. The dotted blue line indicates the Larmor frequency of the nuclear polarisation. The semi-transparent green area highlights the frequency range for which the conventional calibration underestimates the sensitivity, while the red area highlights the frequency range where the conventional calibration overestimates the sensitivity.

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sweep over the leading field that is necessary for the calibration [Eq. (5.7)], leads to the instability of the compensation point due to the change of the nuclear relaxation rate associated with a change of the leading field [Sec. 4.4]. Specifically, significant changes in operational conditions make this method not optimal for the GNOME experiment, as it affects long term-stability of the system due to significant interruptions in the continuity of the observation results due to the comagnetometer being not operational during the calibration routine.

Notably, there is a version of the conventional calibration method that does not require the change of the leading field. This allows one to avoid the issue with the stability of the comagnetometer system generated by the relaxation-rate changes, corresponding to the magnetic-field change. The method employs the low-frequency magnetic-field modulation applied along the primary sensitivity axis [133]. From Eq. (2.187), one can show that, when the modulation frequency ω_m is much smaller than the Larmor frequency of the noble gas $\omega_m \ll \omega_L$, the out-of-phase response of the comagnetometer is proportional to the frequency of the applied magnetic field

$$\Upsilon_x^s(\omega_m) \xrightarrow{\omega_m \ll \omega_n} \frac{\gamma_e \Upsilon_z}{R_e \omega_n} B_m \omega_m, \quad (5.9)$$

where B_m is the amplitude of the sinusoidal modulation. Hence, taking into account Eqs. (2.183) and (5.1), the low-frequency calibration factor is

$$k_{DC} = \frac{S_{out}(\omega_m)}{B_m} \frac{\omega_n}{\omega_m}. \quad (5.10)$$

Although such a version of the calibration method is more suitable for the long-term measurements within the GNOME experiment, it is not optimal because it provides the calibration only for a limited frequency range. For example, it does not provide information on the detuning from the compensation point, while under non-zero detuning from the compensation point, it will lead to an inaccurate calibration factor.

5.3. DETERMINATION OF THE COMPLETE FREQUENCY RESPONSE

The limitations of the conventional calibration method discussed in the previous section necessitate the development of an alternative calibration procedure, which can be used in long-term measurements, e.g., those conducted with GNOME. In Ref. [142], such a new method was proposed. This method is based on an analysis of the magnetic-field frequency response of the comagnetometer followed by a reconstruction of the comagnetometer response to the non-magnetic spin couplings. This method provides the estimation of the frequency response of the system over an arbitrary frequency range for all perturbation types, including both conventional (rotation, magnetic, etc.) and exotic (hypothetical interaction with dark matter) interactions. Hence, in contrast to the conventional calibration method, the proposed method allows for the calibration of the system response for the frequencies above the Larmor frequency of the noble gas. This effectively leads to the extension of the useful comagnetometer bandwidth beyond the noble-gas Larmor frequency. A specific advantage of the method is related to the fact that the magnetic-field frequency response can be measured without scanning the leading field, which makes the method significantly less invasive for the comagnetometer system. The proposed procedure works well under small² detunings from the compensation point and it provides the actual value of the detuning. The combination of these factors makes the proposed method better suited for addressing the requirements of the GNOME experiment.

5.3.1. FROM MAGNETIC TO NON-MAGNETIC FREQUENCY RESPONSE

The theoretical background for a proposed calibration method can be derived from the results presented in Secs. 2.2.8 and 2.2.10. From Eq. (2.173), one can note that the frequency response of the comagnetometer to magnetic and non-magnetic perturbations is determined with the same parameters. Hence, it should be, in principle, possible to estimate the frequency response to non-magnetic perturbations based on the directly measured spectral response to magnetic fields. This explains why the proposed calibration procedure is based on the measurement of the magnetic-field frequency response of the system to a perturbation applied along the prime sensitivity axis (Sec. 5.3.2). Obtained experimental results for in- and out-of-phase response can be described with the experimental model following from Eq. (2.187)

$$\mathcal{F}_{in}^m(\omega) = -\text{Im}[\mathcal{F}_+^m(\omega) + \mathcal{F}_-^m(\omega)], \quad (5.11)$$

$$\mathcal{F}_{out}^m(\omega) = \text{Re}[\mathcal{F}_+^m(\omega) - \mathcal{F}_-^m(\omega)], \quad (5.12)$$

²Its performance was tested for detunings up to $\approx 10\%$ of the compensation-point value. For more details, see Sec. 5.3.4.

where $\mathcal{F}_{\pm}^m$ are get from Eq. (2.173) and have the following parametrisation

$$\mathcal{F}_{\pm}^m(\omega) = -a \frac{\pm\omega + \gamma_n \Delta_{B_z} - i\sqrt{R_n^2}}{(\pm\omega + \omega_e + \Delta_{B_z} \gamma_e / q - \sqrt{R_e^2})(\pm\omega + \omega_n + \gamma_n \Delta_{B_z} - i\sqrt{R_n^2}) - \omega_e \omega_n}, \quad (5.13)$$

with the parameters in the model having the same interpretation as in Eq. (2.173). Here, the fitting parameters are marked with blue and predefined constant parameters are marked with violet. The slowing-down factor has to be approximated with a constant value based on the characterisation of the system prior to the presented calibration. In general, q depends on the alkali-metal polarisation [Eq. (2.46)], however, in a properly operating setup it is not expected to change significantly. In turn, the approximation $q \approx \text{const}$ works well (more details further in this section and in Sec. 5.3.4). Any significant change in the alkali-metal polarisation level inevitably leads to changes in other parameters of the model, hence the proposed calibration method allows detection of the system malfunctioning related to the alkali-polarisation level, even under the assumption of the constant slowing-down factor.

In practice, the in- and out-of-phase components of the signal are fitted simultaneously in the form of the complex signal

$$\begin{aligned} \mathcal{F}^m &= (\mathcal{F}_{in}^m + i\mathcal{F}_{out}^m)e^{i\theta} \\ &= i(\mathcal{F}_+^m - \mathcal{F}_-^{m*})e^{i\theta}, \end{aligned} \quad (5.14)$$

where the star operator stands for the complex conjugate and the phase fitting parameter θ takes into account all potential phase shifts that may arise in the system.

The parameters obtained in the complex-function fit [Eq. (5.14)] allow for the reconstruction of the frequency response by substitution of the response to rotating perturbations estimated with the fit parameters, which have the following form

$$\mathcal{F}_{\pm}^r = -a \frac{\omega_n(\alpha_e - \alpha_n) + (\pm\omega + \gamma_n \Delta_{B_z} - i|R_n|)\alpha_e}{(\pm\omega + \omega_e + \Delta_{B_z} \gamma_e / q - i|R_e|)(\pm\omega + \omega_n + \gamma_n \Delta_{B_z} - i|R_n|) - \omega_e \omega_n}, \quad (5.15)$$

where the coupling parameters marked with the magenta come from Table 2.6. This result can be used for the estimation of the system response to perturbation of arbitrary type and direction by utilising Eq. (2.187) in case of the perturbations applied along the primary sensitivity axis and Eq. (2.188) in case of the perturbations applied along the secondary sensitivity axis. This leads to the following expressions for the frequency responses to perturbations along the primary sensitivity axis ($\mathcal{F}_{pr}^r$) and the secondary ($\mathcal{F}_{sec}^r$)

$$\mathcal{F}_{pr}^r(\omega) = (\mathcal{F}_+^r - \mathcal{F}_-^{r*})e^{i(\theta+\pi/2)}, \quad (5.16)$$

$$\mathcal{F}_{sec}^r(\omega) = (\mathcal{F}_+^r + \mathcal{F}_-^{r*})e^{i(\theta+\pi)}. \quad (5.17)$$

The complex frequency response then allows calculation of the in- and out-of-phase responses as well as the amplitude response

$$\begin{aligned}\mathcal{F}_{in}(\omega) &= \text{Re}[\mathcal{F}_i^r], \\ \mathcal{F}_{out}(\omega) &= \text{Im}[\mathcal{F}_i^r], \\ \mathcal{F}_a(\omega) &= |\mathcal{F}_i^r|,\end{aligned}\tag{5.18}$$

where $\mathcal{F}_i^r$ can stand either for $\mathcal{F}_{pr}^r$ or $\mathcal{F}_{sec}^r$, depending on the considered axis.

5.3.2. MEASUREMENT OF THE FREQUENCY RESPONSE TO MAGNETIC FIELDS

The directly applied set of harmonic perturbations³ with different frequencies allows explicit determination of the frequency response to the perturbations aligned with the direction of the testing perturbations. Such a test perturbation for each considered frequency ω_{test} can be parameterised in the following way

$$B_{test} = B_0 \sin(2\pi\omega_{test}t).\tag{5.19}$$

After reaching the dynamic steady state, the comagnetometer response is fitted with the following function

$$S_{test}(\omega_{test}) = S_0(\omega_{test}) \sin[2\pi\omega_{test}t + \theta(\omega_{test})],\tag{5.20}$$

where $S_0(\omega_{test})$ and $\theta(\omega_{test})$ are the fitting parameters related to the amplitude and phase response of the system, respectively. One can get the amplitude response by dividing the amplitude of the output signal by the amplitude of the applied magnetic-field perturbation

$$|\mathcal{F}_{test}(\omega_{test})| = \frac{S_0(\omega_{test})}{B_0}.\tag{5.21}$$

The results of such measurements, for both sensitivity axes, measured at the first and second harmonic of the modulation frequency are presented in Fig. 5.3.

Fitting experimental data with the theoretical model (Sec. 5.3.1) requires a sufficient number of measurement points over the considered frequency range, particularly in a low-frequency range, i.e., up to the resonant frequency of the nuclear polarisation. As a result, the direct calibration performed by scanning the frequency ω_{test} may take a sufficient amount of time as the measurement for each frequency has to be long enough for the system to reach the dynamic steady state.

The duration of the frequency-response determination can be reduced by the application of pulse-based perturbation [142]. This is why, for more efficient calibration of the Advanced GNOME comagnetometers, it is better to utilise the response of the comagnetometer system to the step-change of the magnetic field applied along the primary sensitivity axis

$$B_{test} = B_0\Theta(t_0 - t),\tag{5.22}$$

where $\Theta(t)$ is the Heaviside theta function and t_0 indicates the time of the change of the magnetic field. To determine the frequency response, it is convenient to consider the

³Although in this case one considers the response to the magnetic field, perturbations of other origins can be considered within a similar framework by replacing the magnetic field with generic perturbation [Eq. (2.162)].

response of the system in the frequency domain, as from Eq. (2.195). The spectrum of the comagnetometer response is determined by multiplication of the spectrum of the comagnetometer and perturbation (here, a Heaviside theta function)

$$S_{test}(\omega) = \sqrt{2\pi} B_0 \Theta(\omega) \mathcal{F}_{test}(\omega), \quad (5.23)$$

where the spectrum of the Heaviside Θ function is

$$\Theta(\omega) = \frac{1}{\sqrt{2\pi}} \frac{1}{i\omega} + \sqrt{\frac{\pi}{2}} \delta(\omega). \quad (5.24)$$

In practice, the frequency-response determination is performed with the application of a square magnetic-field pulse. The pulse ought to be long enough to allow the system to reach a new steady state. The spectrum of the system response can be obtained by performing the FFT of the signal recorded right after switching off the magnetic field. However, for testing with a step function, it is more convenient to consider the derivative of the comagnetometer response

$$\mathfrak{F} \left[\frac{dS_{test}(t)}{dt} \right] = i\omega S_{test}(\omega) = B_0 \mathcal{F}_{test}(\omega), \quad (5.25)$$

which leads to the following form of the experimental frequency response

$$\boxed{\mathcal{F}_{test}(\omega) = \frac{1}{B_0} \mathfrak{F} \left[\frac{dS_{test}(t)}{dt} \right]}. \quad (5.26)$$

The experimental demonstration of the frequency response determination is realised in Kraków comagnetometer by applying a 20-s long square pulse with 100-pT amplitude oriented along the x -axis. The frequency response determined via the derivative of the response to the step change agrees well with the direct measurement of the frequency response for the prime sensitivity axis over the whole considered frequency range (Fig. 5.3). In the case of the secondary sensitivity axis, both results are in agreement for a low-frequency range (up to 10 Hz), while the high-frequency part of the response is dominated by noise in the case of the step-function method. Such behaviour is explained by the inhomogeneous probing of the response on different frequencies by the step change of the magnetic field due to Eq. (5.24), according to which the amplitude spectrum of such perturbation is intentionally proportional to the frequency. Hence, the amplitude of the magnetic-field change used for the calibration has to be large enough for efficient probing of the frequency response over the aimed frequency range.

The frequency range probed by the step-change method is determined by the duration T and sampling rate f_s of the time series used for the calibration of the magnetic-field response

$$\frac{1}{T} < \frac{\omega_{test}}{2\pi} < \frac{f_s}{2}. \quad (5.27)$$

In contrast to the direct measurement, the step-change method provides information about the whole frequency range in a single measurement, which requires the amount of time equivalent to the direct measurement of a single point with the lowest considered

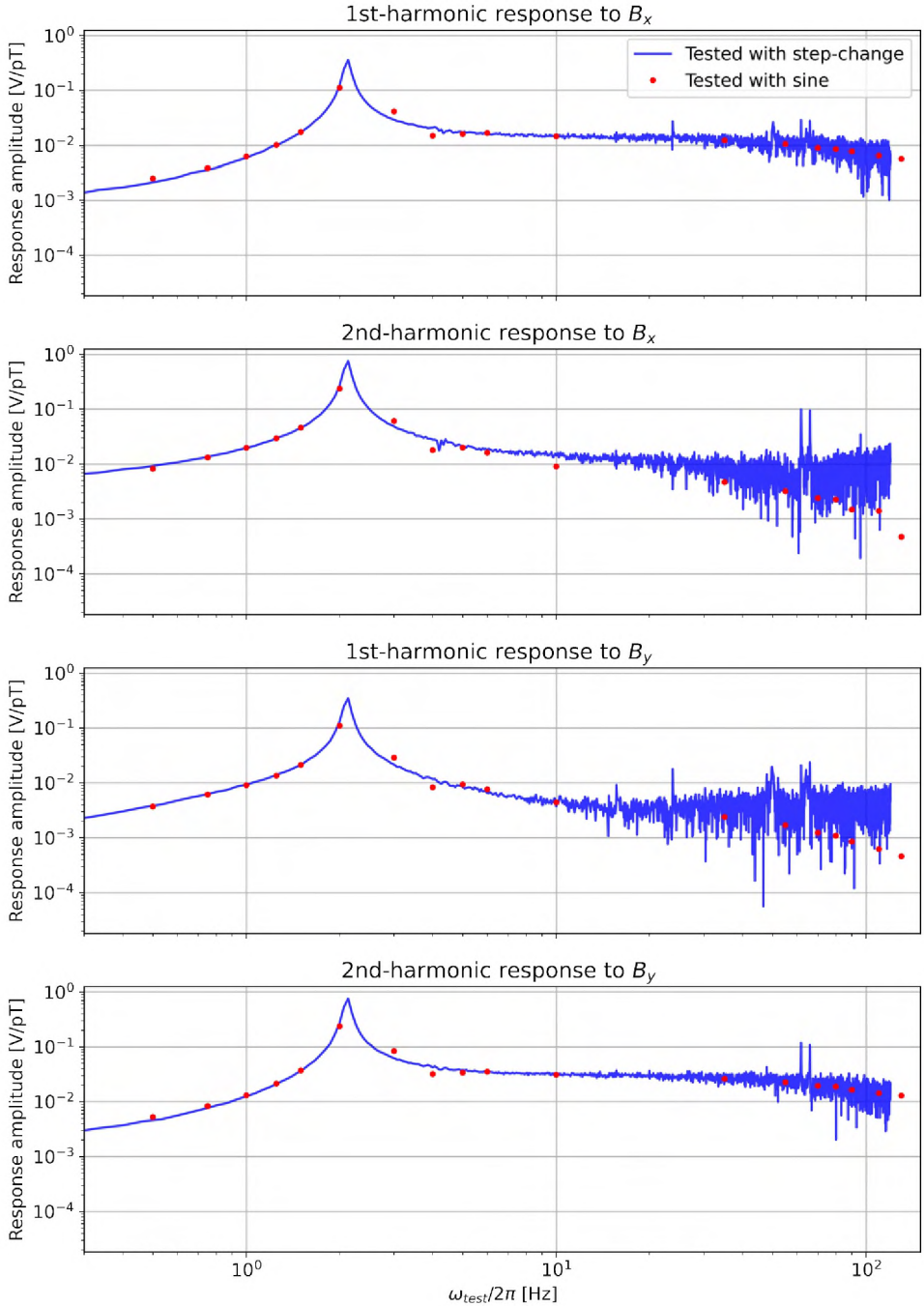


Figure 5.3: Spectral response to the magnetic field measured with scanned sine perturbation [Eq. (5.19)] (red dots) and step-wise change (blue lines). The plots present (from top to bottom): the response to the B_x field measured at the first harmonics of the parametric modulation frequency, the response to the B_x field measured at the second harmonics, the response to the B_y field measured at the first harmonics of the parametric modulation frequency, the response to the B_y field measured with the second harmonics of the parametric modulation frequency. The presented data sets are notch-filtered at the power-line frequency (50 Hz).

frequency. This leads to a higher measurement duty cycle compared with the use of direct measurement.

Due to the rotational symmetry of the system (Sec. 2.2.4), the frequency response to perturbations applied along the prime and secondary sensitivity axes and measured at the first and second harmonic of the parametric modulation (Sec. 2.2.14) are described with the same functions up to the phase factor related to the rotation of the coordinate frame. Since the comagnetometer is operated in a linear regime (Sec. 4.1.3), the experimentally determined frequency response, measured on the first and second harmonic of the modulation, may differ only by a constant factor. This factor allows the normalisation of one output comagnetometer channel with respect to another. Such a normalisation factor is derived based on the directly measured primary-sensitivity-axis responses measured at the first and the second harmonics

$$\eta_{12} = \frac{1}{N_\omega} \sum_i^{N_\omega} \frac{|S_2(\omega_i)|}{|S_1(\omega_i)|}, \quad (5.28)$$

where N_i is the number of tested frequencies, i runs over the tested frequencies, and $|S_{1,2}|$ is the amplitude of the response on a first and second harmonic of the parametric modulation. The comparison between the first-harmonic and the normalised second-harmonic responses is presented in Fig. 5.4. The results for both channels are in good agreement, hence, the normalisation factor combined with the frequency response measured for one of the magnetic-field directions provides complete information about the magnetic-field response of the system.

5

5.3.3. WIDE-BANDWIDTH ESTIMATION OF THE COMAGNETOMETER RESPONSE BASED ON MAGNETIC STEP RESPONSE

Real and imaginary parts of the magnetic response of the comagnetometer [Eq. (5.26)], measured for the primary sensitivity axis, are fitted with the complex-function model defined in Eq. (5.14). The results of the fitting are presented in Fig. 5.5. It was found empirically that the fitting routine works better when the relaxation of the noble-gas polarisation is neglected (it is fixed to $R_n = 0$). This originates from the fact that the noble-gas relaxation rate is much smaller than other characteristic rates of the system. Hence damping of the noble-gas polarisation is determined by its interaction with the electron polarisation.

The reconstructed spectral response to the non-magnetic nuclear spin couplings, determined based on the magnetic-field data, is shown in Fig. 5.6 (top). The results are normalised according to the parametrisation defined in Table 2.6. The results are presented along with the fitted magnetic-field response for comparison. The shape of the amplitude response to neutron and proton non-magnetic couplings and rotation are generally the same for the low-frequencies, while they start to differ at frequencies above 20 Hz. This difference stems from the different coupling constants for the proton and neutron couplings to the alkali-metal polarisation due to the specifics of the nuclear spin content of the alkali-metal atoms and the fact that the rotation couples to the total nuclear spins.

The comparison between the estimation of the non-magnetic and the magnetic-field

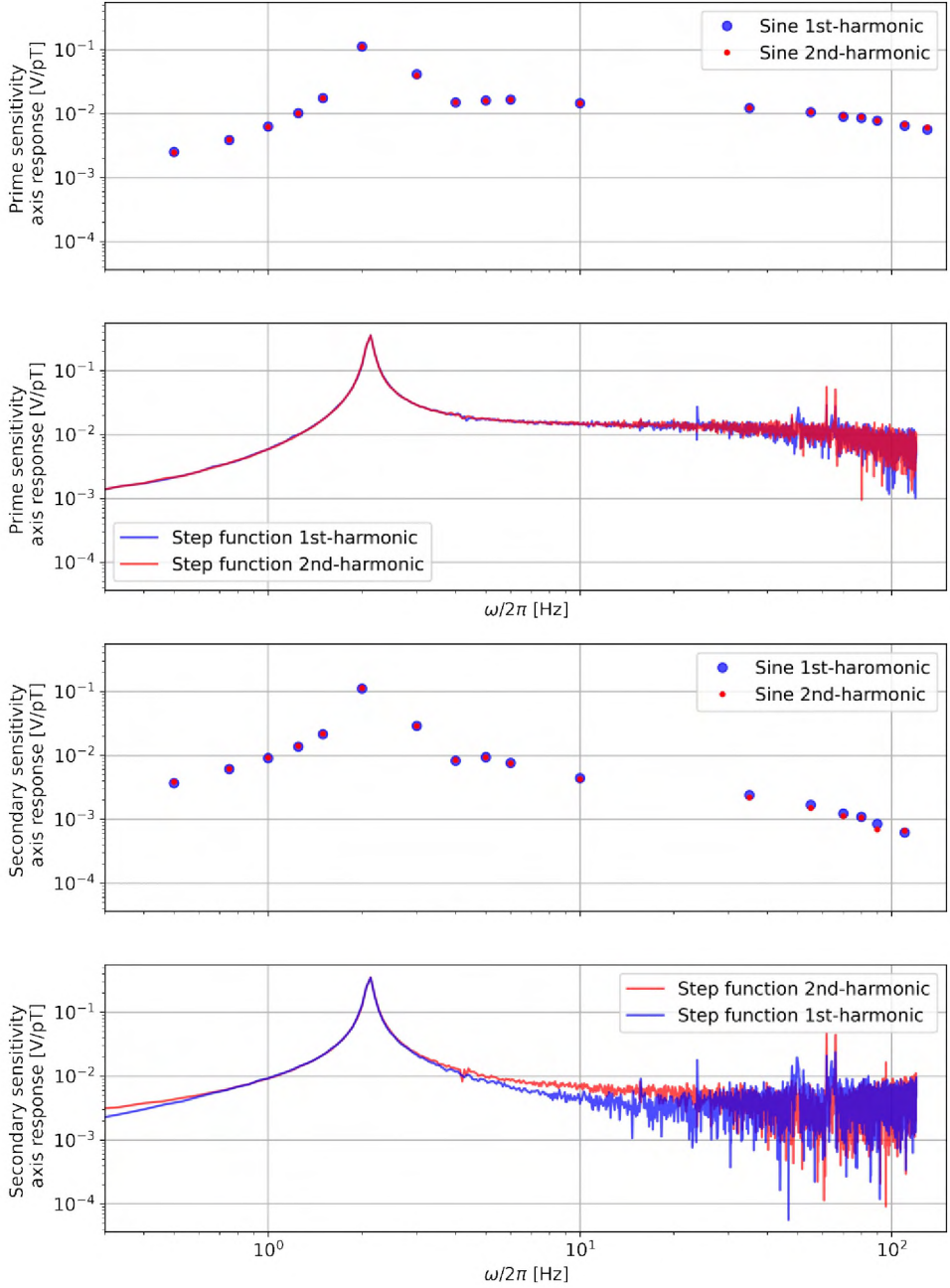


Figure 5.4: Comparison of the spectral response to the perturbations applied along the primary and secondary sensitivity axes, measured at the first and second harmonics of the modulation frequency. The plots show (from top to bottom): the directly measured primary sensitivity axis response; the primary sensitivity axis response measured with the step function; the directly measured secondary sensitivity axis response; the secondary sensitivity axis response measured with the step function. The response on the second harmonic is normalised with the factor defined in Eq. (5.28).

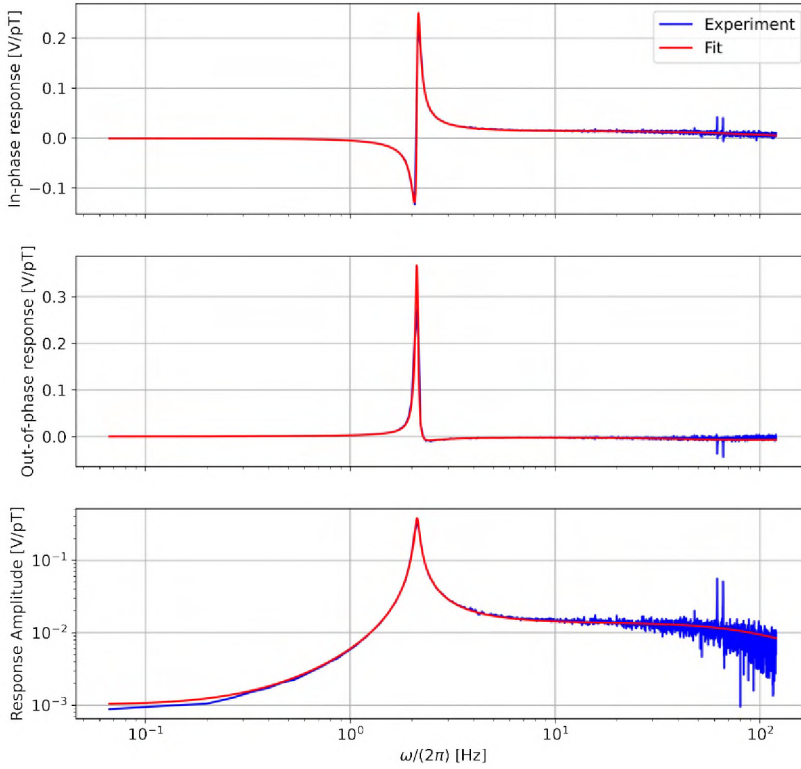


Figure 5.5: Experimental data (in-phase, out-of-phase, and magnitude of the signal) and fitting results for the magnetic-field response for the primary sensitivity axis. The frequency-response determination process is presented in Sec. 5.3.1. The fitting model is presented in Eqs. (5.14).

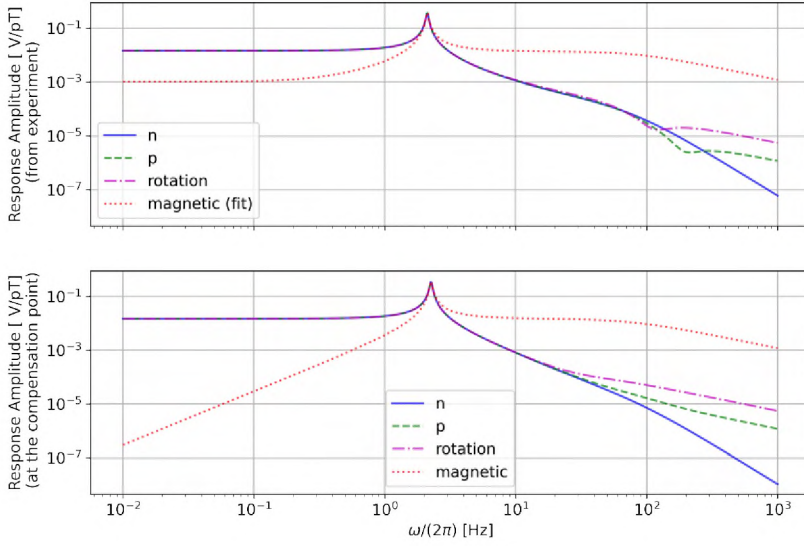


Figure 5.6: Estimation of the frequency response to the neutron (blue solid line) and proton (green dashed line) non-magnetic spin couplings, along with a response to the rotation (magenta dot-dashed line). The estimation is obtained with the proposed calibration method based on the fitting of the magnetic field response (dotted red line) measured in the Kraków comagnetometer. The estimation model is presented in Eq. (5.16). The top plot presents the result of the estimation for the experimental results presented in Fig. 5.5, where non-zero detuning from the compensation point has been observed. The bottom plot presents a similar estimation for the detuning from the compensation point set to zero. All presented results are normalised to the nuclear polarisation coupling constant α_n (see Table 2.6).

responses indicates that the suppression of the response to a low-frequency magnetic field is less efficient than predicted in Sec. 2.2.10. This may be explained by the operation outside of the compensation point at the moment of calibration. As the presented method provides an estimation for the detuning of the compensation point, in addition to the calibration of the system, it can be used for the monitoring of the detuning from the compensation point. One can find more details related to this topic in Sec. 5.3.4. For presented experimental data, the detuning is estimated to be at the level of 4 nT, which is about 5% of the compensation point ($B_c \approx 70$ nT at the moment of the calibration). Accounting for the eventual detuning from the compensation point is particularly important when testing high-frequency perturbations, as, in this frequency range, the response amplitude is strongly affected by the detuning. This is noticeable in Fig. 5.6, which presents the comparison of the obtained result with the estimation of the system response at the compensation point. While the low-frequency response to non-magnetic perturbations is relatively insensitive to a small detuning from the compensation point, the difference in the high-frequency response quickly grows for the frequencies above 20 Hz.

The proposed calibration method (Sec. 5.3.1) not only allows for the calibration of the non-magnetic response but also provides the estimation of the magnetic-field response along the secondary sensitivity axis. This enables cross-referencing the estima-

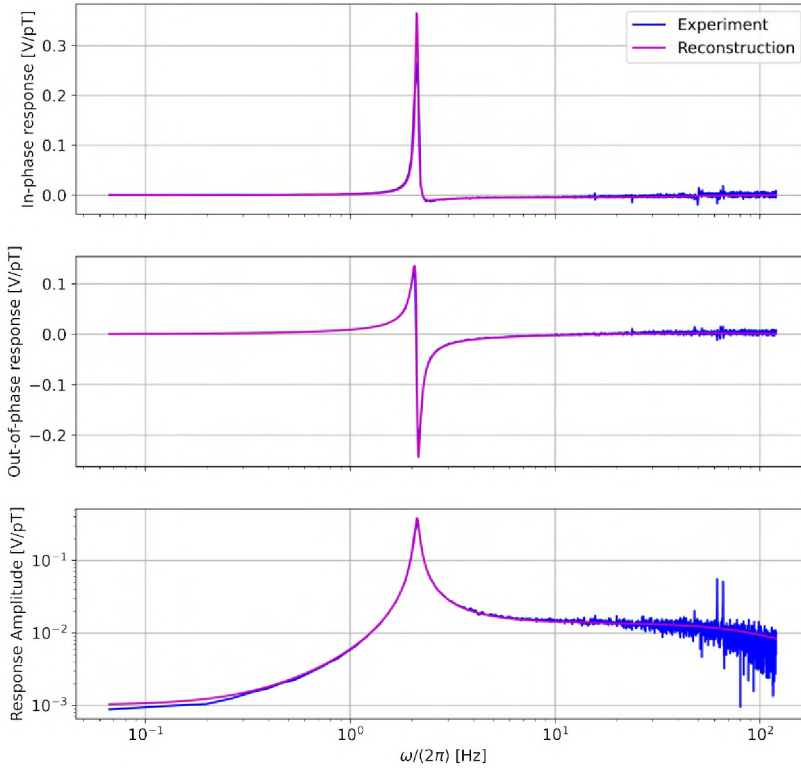


Figure 5.7: Comparison of the experimentally measured response to the magnetic field applied along the secondary sensitivity axis (blue lines) with the estimation based on fitting to the primary-axis data (Fig. 5.5) and the reconstruction model from Eq. (5.17) (magenta lines). The experimental data is notch-filtered at the power-line frequency (50 Hz).

tions with the actual magnetic measurements along the secondary sensitivity axis. The results of such a comparison are presented in Fig. 5.7. The response estimation for the “insensitive” axis might be used as the validation method in the calibration routine for the GNOME station maintenance protocol or in the case of a single-probe-axis operation of the comagnetometer.

The directional sensitivity of the comagnetometer was discussed in Sec. 2.2.11. The implications of this theoretical discussion are in agreement with the experimental data for both primary and secondary sensitivity axes (Fig. 5.8). The experimental results reveal a stronger response at ultra-low frequencies in the case of a field applied along the secondary sensitivity axis, and the responses for both sensitivity axes obtain the same amplitude around the ^3He Larmor frequency. For the higher frequencies, the response to the magnetic field applied along the secondary sensitivity axis is dominated by the noise of the system. The experimental results confirm the directional ambiguity of the magnetic-field response, which is especially prominent at the frequencies close to the noble-gas Larmor frequency. The direction of the perturbation observed at these fre-

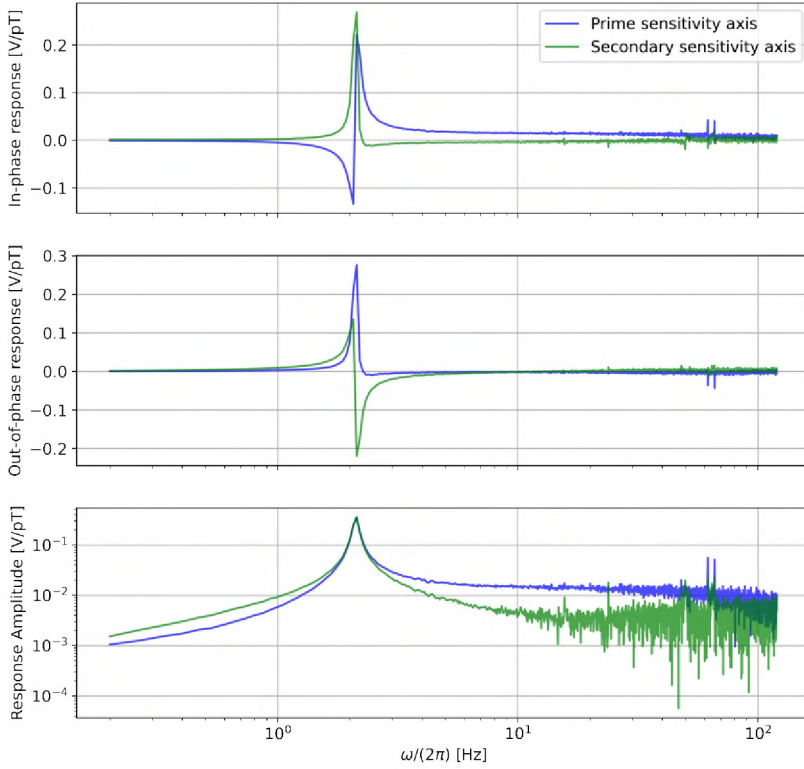


Figure 5.8: Experimental comparison of the magnetic-field response to the perturbation along the primary and secondary sensitivity axes (same data as in Fig. 5.3). The results take into account the normalisation defined in Eq. (5.28).

quencies can be identified only with the analysis of the phase of the response; there is a phase shift between the responses along the primary and secondary axes. A similar situation is expected for the non-magnetic response, which makes the proposed calibration method even more useful, as it allows the estimation of the total frequency response, which includes both sensitivity axes.

5.3.4. RECONSTRUCTED FREQUENCY RESPONSE VS. DIRECTLY MEASURED RESPONSE TO THE ROTATION

Since the gyromagnetic ratio of the noble gas is much smaller than in the case of the alkali-metal atoms, the effective magnetic field, corresponding to a rotation of the comagnetometer, is much larger for the nuclear polarisation than it is for the alkali-metal polarisation (Table 2.6). Therefore, the rotation of the comagnetometer mostly affects the nuclear polarisation. This makes the rotation a natural choice for the imitation of nuclear non-magnetic spin couplings, and it was selected to test the proposed calibration method. In this experiment, which was conducted in Mainz, the comagnetometer was mounted on the rotating stage driven with an electrical step motor (more details

in Sec. 3.8). A specific aim of the experiment was the investigation of the effect for different detunings from the compensation point. For this, the estimated response to the rotation obtained from the calibration routine was compared with the directly measured frequency response to the rotation. In the research, the detuning of ± 10 nT from a compensation point of ≈ 120 nT was investigated.

The experimental sequence for each detuning consisted of two subsequent square pulses: the magnetic-field pulse and the rotation pulse, both of the same length (10 s).⁴ The magnetic-field pulse with a 80-nT amplitude was applied along the x -axis. The rotation pulse consisted of a short acceleration period followed by the constant-angular velocity motion around the x -axis with a frequency of 0.5 mHz (equivalent to a 14 pT magnetic field applied to the nuclear polarisation), which was instantaneously halted. As the rotation stopped fast, the nuclear spins could not react, so the falling edge of the rotation pulse could be considered as a square step change. This is similar to the case of testing perturbation considered in Eq. (5.22). This allows the determination of the frequency response to the rotation with the analysis presented in Sec. 5.3.3.

The left column in Fig. 5.9 shows the spectral response of the comagnetometer to the magnetic field and the fitting result based on Eq. (5.14). The fittings are being used to reconstruct the spectral response to the rotation (solid line in the right-column plots). These results are superimpose on the directly measured spectral responses to the rotation. As shown, the reconstructed rotation response is in good agreement with the directly measured response up to ≈ 20 Hz for all three detunings from the compensation points. For higher frequencies the directly measured data is dominated by the mechanical noise associated with imperfections of the rotating system.

The fitting model used for the calibration of the comagnetometer provides a value for the actual detuning from the compensation point [Eq. (5.14)]. As shown in Eq. (2.164), the detuning Δ_{B_z} is a linear function of B_z as long as the longitudinal polarisation of the system is constant. The estimated values of $\Delta_{B_z}(B_z)$ are presented in Fig. 5.10. The observed dependence agrees well with the linear function for $B_z > -125$ nT, and the zero crossing of this dependence agrees with the compensation point observed experimentally prior to the measurements presented in this section. Hence, the subpart of the presented data set, for which $|\Delta_{B_z}| \leq 4$ nT was fitted with a linear function of the following form

$$\Delta_{B_z}(B_z) = a_B(B_z - B_c), \quad (5.29)$$

where a_B and B_c are the fitting parameters. The fitting results are included in Fig. 5.10. The obtained slope of the linear function has the following value

$$a_B = -1.001(18), \quad (5.30)$$

which is in agreement with the theoretical definition of the detuning from the compensation point [Eq. (2.164)]. For $B_z < -125$ nT, the observed $\Delta_{B_z}(B_z)$ significantly deviates from the linear dependence, which stems from the change of the equilibrium polarisation of the noble gas. The polarisation change results from suppressing the magnetic-gradient relaxation of the noble gas with increasing $|B_z|$ (for more details see Sec. 4.4).

⁴The pulses were long enough for the system to reach the steady state, which allowed the utilisation of the falling edge of the pulse for determination of the system spectral response (Sec. 5.3.2).

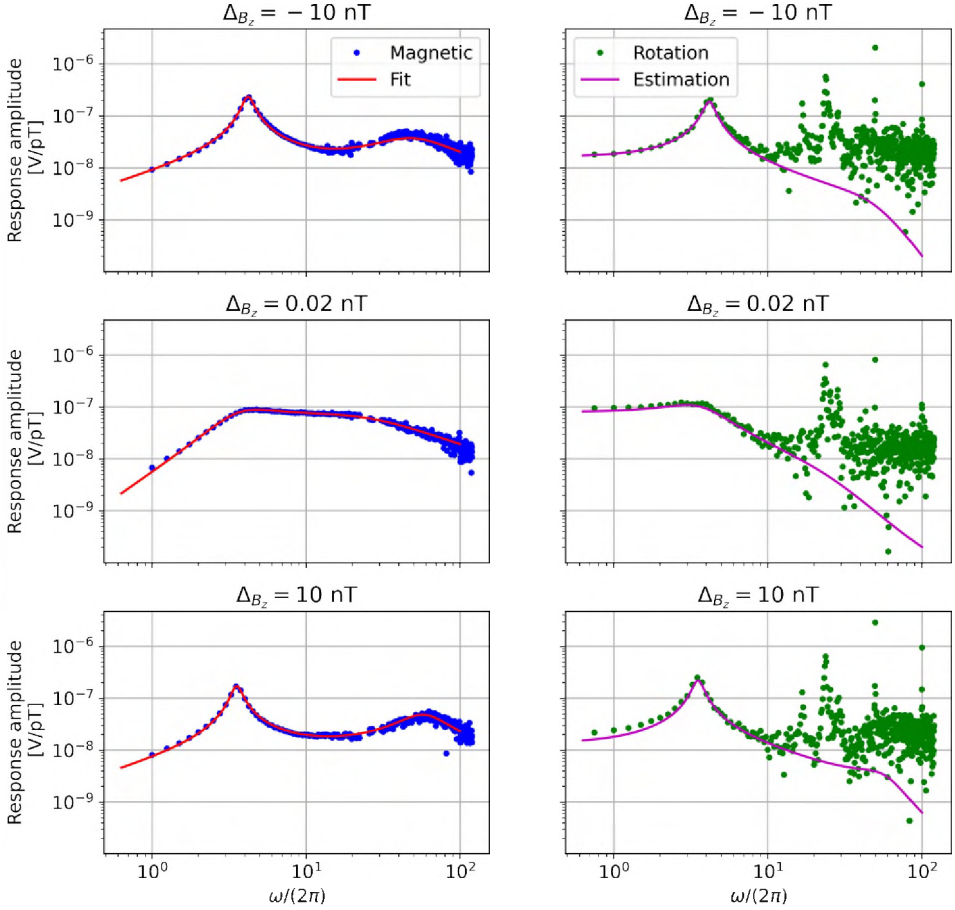


Figure 5.9: Comparison of the spectral response to rotation estimated based on the complete calibration method presented in Sec. 5.3.1 and the result of the direct measurements performed with the Mainz comagnetometer. Each row presents the results for a given detuning from the compensation point (top to bottom: -10 nT, 0.02 nT, and 10 nT). The left column presents the experimentally measured response to the magnetic field applied along the primary sensitivity axis (blue dots) with results of a fitting the model from Eq. (5.14). The right column presents the directly measured frequency response to rotation around the primary sensitivity axis (green dots) along with the sensitivity reconstructed based on the magnetic-field response and the model given in Eq. (5.16).

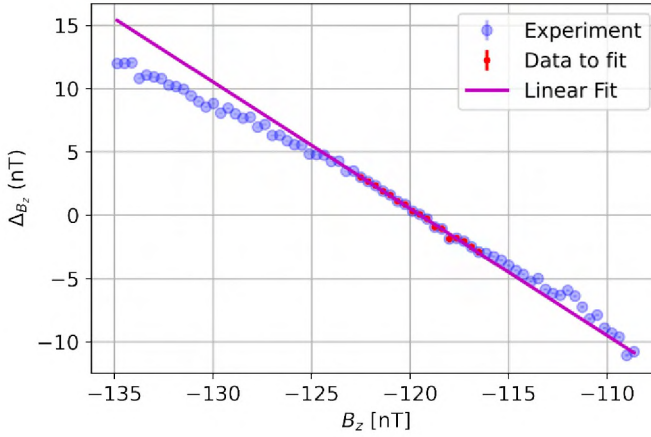


Figure 5.10: Determined detuning from the compensation point ΔB_z as a function of the applied (constant) magnetic field B_z (blue dots). The estimation is obtained with the calibration routine described in Sec. 5.3.1. The results for the small detunings $|\Delta B_z| \leq 4$ nT (red dots) are fitted with the linear model form Eq. (5.29) (magenta line).

5.4. CALIBRATION OF THE MAGNETIC-FIELD COILS

As shown earlier in this chapter, the calibration of the system response to non-magnetic interaction relies on applying the magnetic field. In this scope, the precision of the calibration depends on the precision of the control over the magnetic field. For this reason, accurate calibration of the magnetic-field coils has crucial importance for the overall precision of the comagnetometer calibration.

In the Advanced GNOME comagnetometers, the magnetic-field coils are calibrated with respect to the precession of ^3He . Specifically, in the coil calibration procedure, the comagnetometer operates in a magnetic field much higher than the compensation point ($B_z \gg B_c$). In contrast to the operation at the compensation point, such a high detuning allows the transverse noble-gas polarisation to precess freely without being damped by interaction with the alkali-metal polarisation. As the gyromagnetic ratio of noble gas is known to have extremely high accuracy, its precession frequency provides an accurate reference for the total magnetic field generated by the calibrated magnetic-field coil.

To better understand the operation at conditions utilised for the coil calibration, one can consider Eq. (2.173). The operation far from the compensation point leads to the significant difference between Larmor frequencies of the electronic and nuclear polarisations

$$\omega_e + \gamma_F \Delta B_z \gg \omega_n + \gamma_n \Delta B_z. \quad (5.31)$$

This provides decoupling of the nuclear polarisation evolution from the electron polarisation since the nuclear-polarisation resonant spectrum remains at the low frequencies $\omega \sim \omega_n + \gamma_n \Delta B_z$. In such a regime, the frequency response to rotating magnetic fields [Eq. (2.173)] can be approximated with

$$\mathcal{F}_{\chi_{\pm}}(\omega) \approx A \frac{\pm[\omega \pm (\omega_n + \gamma_n \Delta B_z)] + iR_n}{[\omega \pm (\omega_n + \gamma_n \Delta B_z)]^2 + R_n^2} + C, \quad (5.32)$$

where A and C are complex, frequency-independent parameters, and plus and minus signs in the superscript denote the response to the left- and right-handed rotating perturbation, respectively. Hence, in the considered (spin decoupled) regime, the frequency response to the rotating perturbation is presented by the superposition of two complex Lorentzian, which are centred on the positive and negative resonance frequency with its absolute value equal to the Larmor frequency of the noble-gas spins

$$\omega_{0n} = |\omega_n + \gamma_n \Delta B_z|. \quad (5.33)$$

Moreover, since $\gamma_n \Delta B_z \gg R_n$, the resonances for the left- and right-handed components of the magnetic field are well separated, and therefore, the response to the harmonic perturbation [Eq. (2.186)] can be approximated with a single Lorentzian

$$\mathcal{F}_{Y_\perp^h}(\omega) \approx A \frac{\omega_{0n} - \omega + iR_n}{(\omega_{0n} - \omega)^2 + R_n^2} + C. \quad (5.34)$$

Such a response of the comagnetometer is, in fact, similar to the response of the free noble-gas polarisation, and therefore, it allows the use of the gyromagnetic ratio of ^3He spins for the direct *in situ* calibration of the applied magnetic field.

For the measurements of the ^3He precession, a short yet strong magnetic-field pulse of an amplitude B_x was applied along the x -axis, which induced the free induction decay (FID) of the nuclear polarisation [Fig. 5.11 (top)] visible on the first harmonic of the parametric modulation signal. To get the observed Larmor frequency of the ^3He , the FFT of the FID signal was calculated and fitted with model given in Eq. (5.34) [Fig. 5.11 (bottom)]. The observation of the FID signal allows the determination of the absolute value of the magnetic field experienced by ^3He nuclei since the decaying transverse polarisation oscillates around the total magnetic field.

To obtain the calibration factor for the B_z coil, the ^3He precession measurements were repeated for different values of the current applied to the coil [Fig. 5.12 (top)]. The Larmor frequency was then recalculated into the applied field amplitude using the gyromagnetic ratio of the ^3He [143]

$$\gamma_{^3\text{He}} = 32.43409942(38) \text{ MHz/T}. \quad (5.35)$$

The obtained results are in good agreement with Eq. (5.33) since the observed Larmor frequency scales linearly with the applied magnetic field. The calibration factor for the B_z coil corresponds to the slope of $|B(J_z)|$ dependence, where J_z is the current applied to the magnetic coil and B is the total magnetic field experienced by the noble-gas spins. The calibration factor k_z is obtained with the linear fit to the experimental results [Fig. 5.12 (top)] with the following model

$$|B| = k_z J_z + c, \quad (5.36)$$

where k_z and c are the fitting parameters. This results in the following numerical value of the calibration factor

$$k_z = 0.1034404(44) \text{ nT}/\mu\text{A}. \quad (5.37)$$

For calibration of the transverse magnetic coils (B_x and B_y coils), a constant leading magnetic field is applied along the z -axis, such that the system is operated far from the

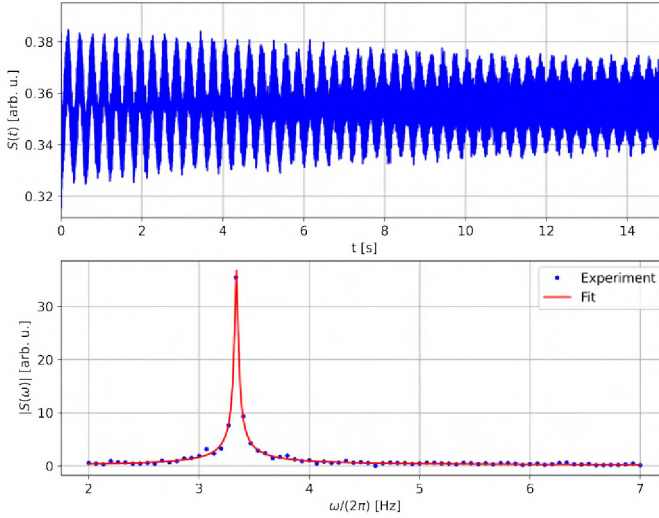


Figure 5.11: Representative FID measurement used for determination of the ^3He Larmor frequency during the calibration of the Advanced Kraków comagnetometer. The top plot presents the time series of the output signal recorded right after application of the B_x pulse used for the excitation of the FID. The bottom plot presents the FFT of the measured time series (blue dots) with the fitted model based on Eq. (5.34) (red line).

5

compensation point. Then, the measurements of the Larmor frequency with the FID technique are repeated for different values of the current in the calibrated transverse-magnetic-field coil. The range of the current is kept small enough such that the leading field has the main impact on the value of the total magnetic field. For small values of the perpendicular magnetic field, the absolute value of the magnetic field is the second-order function of the transverse magnetic field

$$|B| = \sqrt{B_z^2 + B_\perp^2} \approx |B_z| + \frac{B_\perp^2}{|B_z|}, \quad (5.38)$$

which explains the shape of the experimental dependence shown in Fig. 5.12 (middle and bottom plots). The calibration factors for the transverse coils are obtained by fitting the experimental results with the following model

$$|B_i(I_i)| = k \sqrt{J_i^2 + c}, \quad (5.39)$$

where k and c are the fitting parameters and k corresponds to the calibration factor of the studied coil. The fitting dependences are presented in Fig. 5.12 (middle and bottom), and the corresponding calibration parameters are

$$\begin{aligned} k_y &= 0.0554589(19) \text{ nT}/\mu\text{A}, \\ k_x &= 0.056366(12) \text{ nT}/\mu\text{A}. \end{aligned} \quad (5.40)$$

One drawback of the proposed calibration scheme is its inability to determine the direction of the field (the precession frequency for two equal but oppositely oriented fields

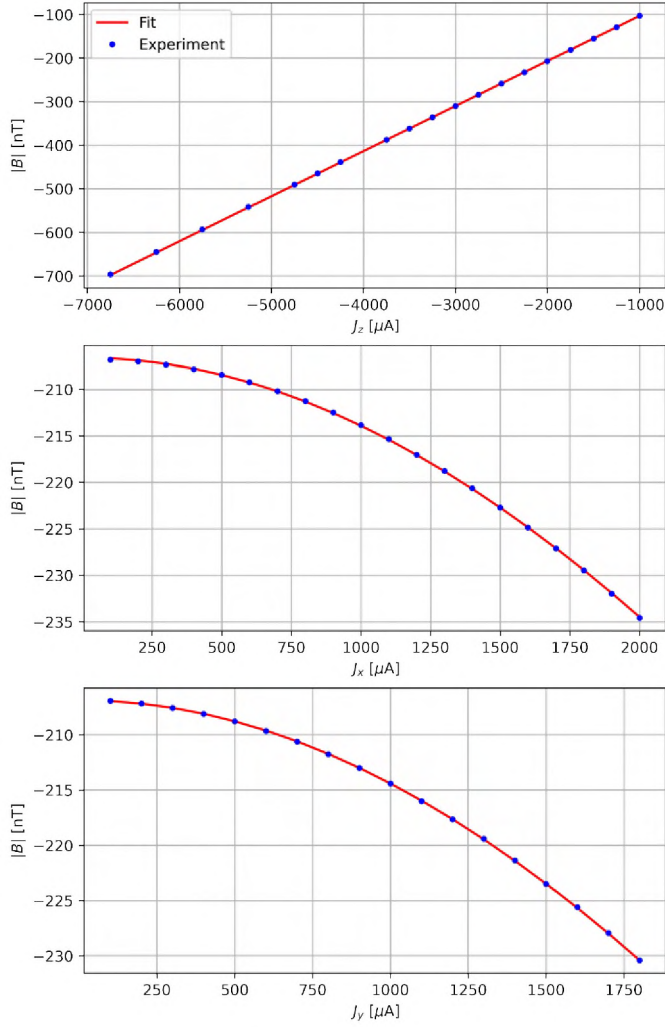


Figure 5.12: Measured values of the total magnetic field, recalculated from the ^3He spin precession frequency, versus the current applied with the magnetic-field coils in all three directions. The top plot presents the results for z-coil, fitted with the model given in Eq. (5.36). The remaining plots present the results obtained for the current applied in the x-middle) and y-coils (bottom). These results were fitted with the model given in Eq. (5.39).

are identical). Therefore, to determine the field orientation, a flux-gate magnetometer was used with a constant DC current to supply the coils. These measurements allowed the orientation of the coordinate system used in Fig. 3.1 with the actual direction of the field.

5.5. CALIBRATION FOR THE GNOME EXPERIMENT

It should be stressed that the presented discussion of the calibration method does not provide a recipe to recalculate the comagnetometer output signal into the signal, which can be processed using the GNOME data-analysis pipeline. In general, various data-analysis approaches may be used to classify signals in the time domain (time-series analysis) and frequency-domain analysis (spectral analysis).

One can consider an output signal $S(t)$ for the system characterised by the frequency response estimated with the presented calibration routine $\mathcal{F}^r(\omega, \alpha_n, \alpha_e)$ [Eqs. (5.16) and (5.17)]. The calibrated spectral response of the comagnetometer can be obtained from Eq. (2.195) by dividing the spectrum of the measured time series $S(\omega)$ by the frequency response $\mathcal{F}^r(\omega, \alpha_n, \alpha_e)$

$$\mathfrak{b}(\omega, \alpha_n, \alpha_e) = \frac{1}{\sqrt{2\pi}} \frac{S(\omega)}{\mathcal{F}^r(\omega, \alpha_n, \alpha_e)}. \quad (5.41)$$

This can then be used for the spectral analysis of the signal output. In the case when the demanded GNOME data analysis method operates based on the time series, a properly calibrated time series can be obtained with the inverse Fourier transform of the calibrated spectrum

$$\mathfrak{b}(t, \alpha_n, \alpha_e) = \frac{1}{\sqrt{2\pi}} \mathfrak{F}^{-1} \left[\frac{S(\omega)}{\mathcal{F}^r(\omega, \alpha_n, \alpha_e)} \right], \quad (5.42)$$

which then can be used in the data analysis routine conducted in the time domain.

5.6. SUMMARY OF THE CHAPTER

The calibration procedure described in this chapter allows for the interpretation of the comagnetometer output signal in terms of magnetic and non-magnetic (particularly exotic) spin perturbations. The important feature of the presented framework comes from the fact that the amplitude of the output signal is defined with respect to the physical constant represented by the magnetic moment of the ^3He nucleus, which is ensured by a precise calibration of the magnetic-field coils. Moreover, the high damping rate of the response to magnetic-field transient perturbation near the compensation point minimises the time necessary for the measurement used in the calibration. Specifically, it was shown for the Mainz comagnetometer that this time may be as short as 4-s long calibration.⁵ It was shown that such a short calibration routine leads to the complete characterisation of the system response to all types of perturbations independent of its

⁵With respect to the Kraków comagnetometer, the Mainz device provides a higher noble-gas polarisation due to better sphericity of the cell, which limits the additional polarisation relaxation. As a result, the Mainz comagnetometer calibration may be faster than the one in Kraków.

frequency for both sensitivity axes without an effect on the long-term stability of the system. Hence, the presented calibration routine is well-optimised for long-term continuous measurements conducted within the GNOME experiment.

6

PERFORMANCE OF THE ADVANCED GNOME KRAKÓW STATION

This chapter considers a representative experimental data set to demonstrate the capabilities of the Kraków Advanced GNOME comagnetometer. In particular, its sensitivity and long-term stability are under investigation. The information provided in the chapter can be used to estimate the sensitivity of the comagnetometer and further of the whole GNOME network.

As many GNOME analysis routines require the calibrated data streams [41], the chapter starts with the consideration of the calibrated time series (Sec. 6.1). The quality of the comagnetometer performance in the context of the GNOME experiment is determined by its sensitivity, long-term stability, and duty cycle. Therefore, Sec. 6.2 investigates the sensitivity and the long-term stability of the comagnetometer via analysis of its noise spectrum, calibrated with the method described in Sec. 5.3. The noise analysis is followed by an investigation of the comagnetometer calibration stability (Sec. 6.3). On one hand, the consistency of the comagnetometer calibration has an impact on the long-term stability of the comagnetometer signal, but on the other hand, it directly affects the duty cycle of the comagnetometer, as it sets the upper limit on the re-calibration period. The regular daily variations of the sensitivity observed in the comagnetometer signal are analysed in Sec. 6.4. These variations are identified to be of technical origins.

6.1. THE CALIBRATED TIME SERIES

The procedure for the calibration of the comagnetometer is described in Sec. 5.5. It starts with the calculation of FFT of the uncalibrated comagnetometer output signal. Then the signal is divided by the transfer function obtained based on the pulse-based calibration (Sec. 5.3) and the lockin-amplifier transfer function.¹ The inverse FFT of such a

¹As a lock-in amplifier is used for demodulation of the signal at the parametric-modulation frequency, its low-pass filter may affect the comagnetometer output signal.

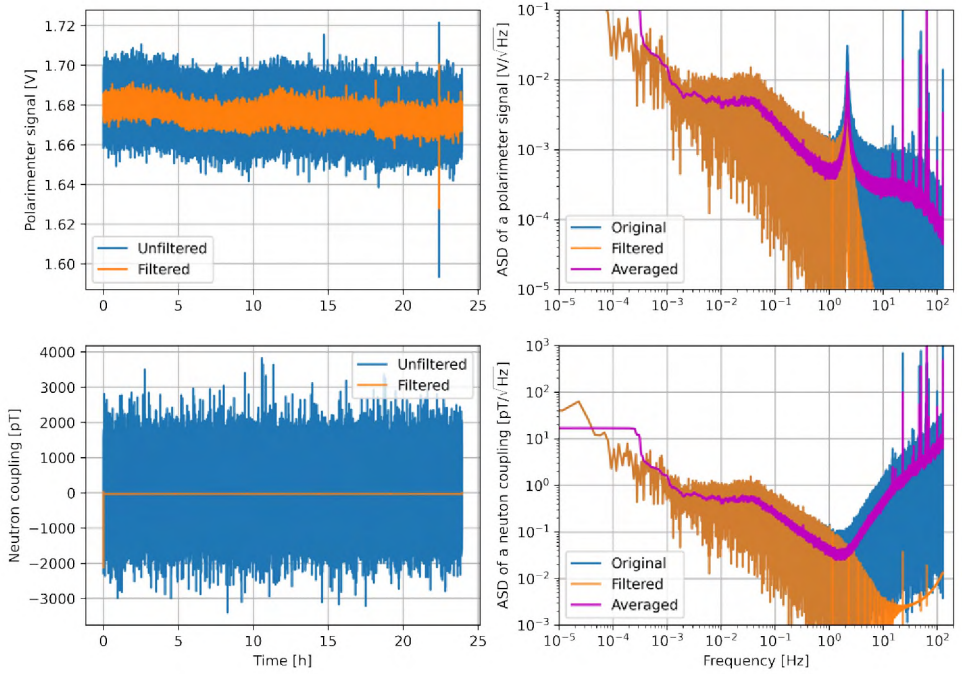


Figure 6.1: Raw and filtered signal measured with the comagnetometer (top) and the signal after calibration with respect to the response to neutron exotic spin coupling (bottom). The left column presents the time series, while the right column presents the amplitude spectral density (ASD) of the signal. Each time series and ASD are presented in two forms: unfiltered (blue) and after application of the first-order low-pass filter with the 3-dB cutoff at 2 Hz (orange). The moving averaged spectra (magenta) are provided for illustration.

calibrated spectrum provides the time-dependent calibrated signal that can be used for further analysis.

In the discussion below, only the primary sensitive axis is considered for simplicity. However, the approach can be easily generalised for the secondary sensitivity axis by changing the frequency-response function used in the calibration.

In the considerations below, the output signal calibrated with respect to the neutron coupling is used as an example. A 24-hour sub-set of data, starting at 20:04:10 on August 4, 2023 (CEST), is analysed. Note that the last 250 s of a data set is excluded from the analysis, as it contains the data with the pulse response utilised for the calibration.

The time series for uncalibrated comagnetometer output and the signal-based calibrated neutron-coupling response along with amplitude spectral density (ASD) of both signals are presented in Fig. 6.1. The neutron-coupling time series reveals a substantial noise level [Fig. 6.1 (bottom)], as the standard deviation of the presented neutron-coupling data reaches a 0.7-nT level. This is due to the enhanced impact of the high-frequency noise component, which is seen in ASD of the observed neutron coupling signal. Such an enhancement is related to the decreasing response amplitude for the higher perturbation frequency (Fig. 5.6). This may affect the sensitivity of the time-domain data

analysis in the context of the GNOME searches, however, when the time scale of the signals does not require consideration of the high-frequency components of the spectrum (e.g., when searching for relatively long transients), the low-frequency filter can be applied to the signal. This allows a significant reduction of the observed noise level. For illustration purposes, in Fig. 6.1 (bottom), the low-pass filtered time series was for both raw and calibrated signals. In the case of the calibrated signal, applying the first-order low-pass filter with a 2 Hz cut-off frequency leads to a reduction of the standard deviation to 3.6 fT. The application of the low-pass filter, when it is possible, will lead to the significant improvement of the time-domain SNR and, hence, to better sensitivity of the time-domain data analysis routines.

Although the compensation of the low-pass filter of the lock-in amplifier and then the application of another low-pass filter in post-processing may seem counter-intuitive, such an approach has a significant advantage. The signal calibrated with the presented method reflects the correct response of the comagnetometer over an arbitrary frequency range. Further analysis of the comagnetometer signal becomes independent of the specific system parameters since all the physical information about the system response is present in the signal, and there is no need for any metadata for the analysis. This means that the resulting calibrated signal can be treated as the detected projection of the exotic spin coupling on the sensitivity axis of the comagnetometer in the bandwidth determined by the Nyquist frequency for the considered time series. Of course, obtaining truly generic data requires information about the isotopes used for the particular system and the convention used for the definition of effective magnetic fields.² Expressing the signal in terms of the energy-level splitting of a fermion in question would make the considered data generic and independent of the system type (not only system parameters). In principle, such an approach allows analysis of the data from the network consisting of different types of sensors, e.g., alkali-metal magnetometers and self-compensating comagnetometers.

6.2. SENSITIVITY AND STABILITY OF THE COMAGNETOMETER

It is important to emphasise that the results regarding sensitivity to different spin couplings presented in this section have been derived using the same experimental data. As sensitivity reflects SNR, the differences in the sensitivity level are related to different amplitude responses to the perturbation of different origins, which is determined by the frequency-response calibration.

The presented data illustrate one of the most prominent results of the full-frequency-range calibration, which is visible in the difference in the raw and neutron-coupling calibrated signals ASDs (the right column in Fig. 6.1). While the ASD signal has a distinct noise peak at around 2 Hz, determined by the noble-gas Larmor frequency, there is no such feature in the calibrated signal. This is because of the correction of the frequency response of the comagnetometer. A similar situation is observed for the magnetic field and rotation sensitivity (Fig. 6.2), as well as for the sensitivity to the other non-magnetic spin couplings (Fig. 6.3). This results in the system having the highest sensitivity at

²In this case, all effective fields are defined with respect to the field experiences by the nuclear polarisation, as was defined in Table 2.6.

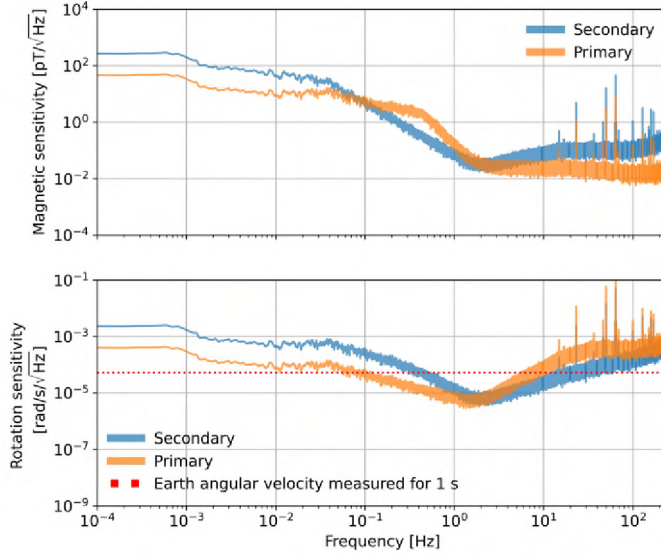


Figure 6.2: Experimentally measured comagnetometer sensitivity to known spin couplings. The top plot presents the sensitivity to the magnetic field applied along the primary sensitivity axis (orange) and the secondary sensitivity axis (the blue curve). The bottom plot presents the sensitivity to rotation around the primary sensitivity axis (the orange curve) and the secondary sensitivity axis (blue). The red dotted horizontal line shows the response amplitude equivalent to the Earth rotation rate (for the details, see Sec. 6.2.2).

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the frequency between 1 and 2 Hz for all non-magnetic spin couplings. In turn, the low-frequency sensitivity to the non-magnetic spin couplings is deteriorated by non-magnetic noise sources (e.g., vibrations, pump-beam-generated light shifts, technical noise) and in some cases by the lower amplitude of the response (Fig. 5.6). The effect of the magnetic-field noise on the low-frequency sensitivity is assumed to be small due to the self-compensation mechanism resulting in a significantly lower magnetic-field sensitivity in this frequency range (Fig. 6.2). The low-frequency part of the noise spectrum reveals an interesting feature, which is also present in the calibrated sensitivity. For the frequency range between 0.04 and 1 Hz, the noise floor is scaled as f^{-1} , and the same trend is observed for the frequencies below 0.001 Hz. However, the noise floor remains flat for the frequency range between 0.001 and 0.04 Hz. Such behaviour may suggest the existence of two separate sources of the $1/f$ noise in the system [144]. This issue is investigated further in Sec. 6.2.4.

At the high-frequency limit, the sensitivity to non-magnetic spin couplings drops due to the decrease in the response amplitude [Eq. (5.15)]. The specifics of the frequency-dependent sensitivity trends for various spin couplings are discussed in the following subsections. The last section (Sec. 6.2.4) provides the noise characteristics along with the analysis of the long-term stability of the output signal.

6.2.1. SENSITIVITY TO THE MAGNETIC FIELD

As one can expect, due to the self-compensation mechanism, the comagnetometer has reduced sensitivity to the low-frequency magnetic field for both sensitivity axes. This effect is visible in the experimental results presented in Fig. 6.2 (top). In the case of the primary sensitivity axis, the plateau at frequencies between 0.001 and 0.04 Hz is present, and the noise amplitude is at the 1-nT level, while the magnetic-field sensitivity at frequencies between 0.04 and 2 Hz is approximately scaled as f^{-3} . In the case of the secondary sensitivity axis, the same frequency region reveals the f^{-2} scaling, while at the frequency range where for the primary sensitivity axis a plateau is observed, the scaling is close to f^{-1} . These trends are changing at ≈ 2 Hz, where the sensitivity along both axes reaches its maximum of ≈ 30 fT. For higher frequencies, the system is more sensitive to the magnetic field parallel to the primary sensitivity axis since, in this case, the sensitivity remains at a constant level up to 100 Hz, where it starts to decrease due to the limited bandwidth of the alkali-metal response. In the case of the secondary sensitivity axis, the sensitivity constantly decreases for the frequency above 2 Hz due to the decrease of the response amplitude (Fig. 5.3).

6.2.2. SENSITIVITY TO THE ROTATION

The calibration results reveal the high sensitivity of the comagnetometer to rotation [Fig. 6.2 (bottom)]. To put the obtained result into perspective, the signal amplitude, corresponding to the Earth's rotation angular velocity Ω_E , is added to the plot in addition to the rotation sensitivity. The comagnetometer is sensitive to the angular velocity of the system rotation around the sensitivity axis. Hence, the rotational modulation of the system can be understood as the harmonic oscillation ("shaking") of the system around the considered sensitivity axis such that the rotation angle α is described in the following way

$$\alpha = \alpha_0 \sin(2\pi f t), \quad (6.1)$$

where α_0 and f are the modulation amplitude and frequency, respectively. The rotation rate, playing the role of rotational perturbation Ω , can be determined with the time derivative of the rotation angle

$$\frac{d\alpha}{dt} = \Omega = 2\pi f \alpha_0 \cos(2\pi f t), \quad (6.2)$$

hence, the angle amplitude, corresponding to the perturbation equivalent to the angular velocity of the Earth's rotation, is

$$\alpha_0 = \frac{\Omega_E}{2\pi f}. \quad (6.3)$$

Since the length of the shorter edge of the optical table,³ at which the comagnetometer rests is $l = 1.2$ m, the amplitude of the table edge oscillation at 1 Hz is equal to $7 \mu\text{m}$. Moreover, considering the same framework, the actual comagnetometer sensitivity at 1 Hz corresponds to the table edge translation of $1 \mu\text{m}/\sqrt{\text{Hz}}$. Such a high sensitivity to the rotation makes the magnetometer system susceptible to vibration noise sources.

³The shorter edge of the optical table is parallel to the z -axis and is perpendicular to the x -axis.

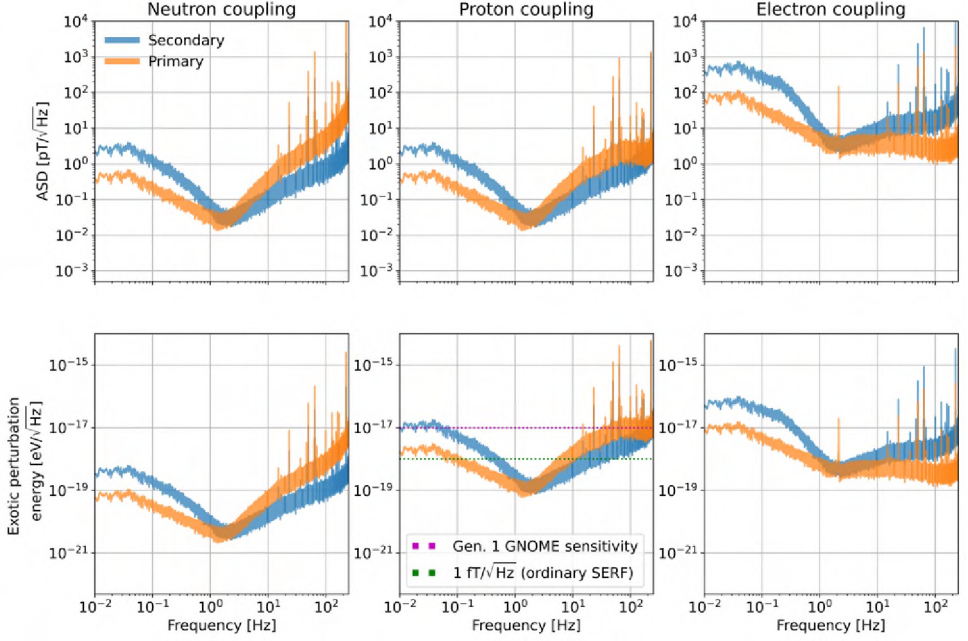


Figure 6.3: Kraków magnetometer sensitivity to the exotic spin couplings presented in effective magnetic-field units (top row) and in the energy units (bottom row). The results for exotic spin couplings to neutron, proton, and electron are shown in the first, second, and third columns, respectively. Each plot presents the sensitivity along the primary sensitivity axis (orange curve) and the secondary sensitivity axis (blue curve). The plot displaying the proton results in energy units (second column in bottom row) presents both the average sensitivity of the first-generation GNOME network (represented by the magenta dotted line) and the sensitivity level equivalent to the typical sensitivity of a ^{39}K SERF magnetometer, which corresponds to a 1 fT effective magnetic-field sensitivity.

6.2.3. SENSITIVITY TO THE EXOTIC SPIN COUPLINGS

The sensitivity of the calibrated comagnetometer to exotic spin couplings is shown in Fig. 6.3. The experimental results are presented in both the effective magnetic-field units (defined according to Tab. 2.6) and the exotic-coupling energy units.

In the case of the neutron spin coupling [Fig. 6.3 (left column)], the highest sensitivity is observed at a frequency of around 1.5 Hz, and it reaches $5 \cdot 10^{-21}$ eV/ $\sqrt{\text{Hz}}$ for both sensitivity axes. For the frequency between 0.04 Hz and 1.5 Hz, the sensitivity along the primary sensitivity axis scales approximately as $f^{-1/2}$, while for the secondary axis approximately as $f^{-3/2}$. For the frequencies below 0.04 Hz, the sensitivity reaches plateau up to 0.001 Hz, where the sensitivity along the primary axis levels up a 10^{-19} eV/ $\sqrt{\text{Hz}}$ and along the secondary axis at 10^{-17} eV/ $\sqrt{\text{Hz}}$. For the frequencies below 0.001 Hz, the sensitivity starts to deteriorate again in a f^{-1} manner, and for the frequencies above 2 Hz, the sensitivity to neutron coupling scales as f^2 in the case of the primary sensitivity axis and as f in the case of the secondary sensitivity axis. These trends are kept until ≈ 100 Hz where the sensitivities start a faster drop due to the limited response of the alkali-metal polarisation to the perturbation (bandwidth limit of the comagnetometer).

The low-frequency sensitivity to the proton coupling follows the same pattern as the neutron sensitivity (up to ≈ 11 Hz), however, in terms of the coupling energy, it is more than an order of magnitude worse than for the neutron coupling. The highest sensitivity to the proton coupling is observed at about 1.5 Hz and is equal to $1.5 \cdot 10^{-19}$ eV/ $\sqrt{\text{Hz}}$. In this frequency range, the difference in the coupling sensitivity to either nucleon mostly arises due to the difference between the proton and neutron spin contributions in the ^3He nuclear spin. In contrast to the neutron coupling, the proton-coupling sensitivity remains approximately constant at 10^{-16} eV/ $\sqrt{\text{Hz}}$ for frequencies between 11 and 100 Hz. Such a difference between the sensitivity to the neutron coupling with respect to the proton coupling sensitivity stems from the fact that the proton coupling perturbs both noble-gas and alkali-metal polarisations, while the neutron coupling perturbs only the noble-gas polarisation. Specifically, the proton coupling trend is changing when the coupled-spin system starts to be predominantly perturbed through the alkali-metal polarisation. Hence, for the frequencies above 11 Hz, the proton-coupling sensitivity is mostly determined by the response of the electron polarisation (similar to the high-frequency response to the magnetic field) and therefore, it starts to drop again at frequencies above 100 Hz due to the limited bandwidth of the alkali-metal response. This demonstrates the importance of accounting for the proton coupling to the alkali-metal polarisation when the comagnetometer is used for the high-frequency searches for exotic spin couplings. It is also worth noting that the comagnetometer at higher frequencies has better sensitivity to nuclear exotic spin couplings along the secondary axis, while at lower frequencies, the situation reverses. This fact must be taken into account in the analysis of the directional sensitivity of the Advanced GNOME sensors.

As for the electron coupling sensitivity, it has near flat characteristics at frequencies between 1 and 100 Hz, where the sensitivity reaches approximately 10^{-18} eV/ $\sqrt{\text{Hz}}$. At the lower frequencies, the sensitivity scales similarly to the nuclear coupling. For frequencies above 100 Hz, the sensitivity starts to decrease due to the limited bandwidth of the alkali-metal response. Finally, for the exotic spin couplings to the electron, the comagnetometer is mostly sensitive along the primary sensitivity axis [Fig. 6.3 (third column)].

One should note that the presented results for the electron spin couplings do not take into account the effect of the potential magnetic field induced inside of the magnetic shield due to the interaction of the exotic field with electrons inside of the shield (for more details, see Sec. 1.3.3). The application of the self-compensating comagnetometer to search for such couplings requires a precise consideration of the comagnetometer-shield system, which is beyond the scope of this work.

The current sensitivity of the Kraków comagnetometer is about an order of magnitude worse than in the case of the state-of-art system, which reached sub-fT/ $\sqrt{\text{Hz}}$ sensitivity [137]. Such a difference is a result of several factors. First, as discussed in more detail in Sec. 6.4, the sensitivity of the Kraków comagnetometer is limited by mechanical noise (vibrations). Second, the state-of-art comagnetometer utilised cells with about three times higher concentrations of noble-gas atoms, which allowed for stronger noble-gas magnetisation and, therefore, stronger suppression of the magnetic-field response, as well as stronger response to the non-magnetic field [Eq. (2.173)]. The third aspect that explains the deteriorated ultra-low-frequency sensitivity can be related to the fact that the parametric modulation provides the less efficient suppression of the pump-beam-

related light shift generated by the air motion in the pump-beam path compared to the vacuum systems surrounding the whole experimental setup implemented in the other work [81, 137].

In the context of the GNOME searches, the Kraków comagnetometer, in addition to the obvious extension of the GNOME capabilities by probing the neutron couplings, also opens a possibility for the improvement of the proton coupling sensitivity. Therefore, in Fig. 6.3 (second column, bottom), the average sensitivity of the first-generation GNOME is presented along with the proton coupling sensitivity to facilitate the comparison. Even though the current sensitivity of the Kraków comagnetometer is far from the best shown in the literature, it surpasses the sensitivity of the Generation 1 GNOME sensors over about 80 Hz bandwidth (when both magnetometer axes are considered), and in the best case scenario, it has about two orders of magnitude better sensitivity. Moreover, the same plot presents the sensitivity corresponding to the state-of-art SERF magnetometer based on the ^{39}K , assuming it has $1\text{-fT}/\sqrt{\text{Hz}}$ sensitivity,⁴ and the Kraków comagnetometer at current state surpasses its sensitivity at frequency up to 10 Hz. In the best case, the difference is equal to approximately an order of magnitude, effectively reaching the fundamental limit for the conventional SERF magnetometers. This demonstrates that even a sub-optimal comagnetometer setup allows one to obtain a better sensitivity to the proton spin coupling compared to the best possible SERF magnetometers sensitivity.

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6.2.4. NOISE CHARACTERISATION AND SYSTEM STABILITY

For a better insight into the noise characteristics of the comagnetometer, one can consider the two-point standard Allan deviation of the calibrated neutron-coupling output (Fig. 6.4).⁵ The Allan deviation obtained for a short observation time revealed fast oscillations, indicating high-frequency deterministic oscillations in the comagnetometer output. These oscillations are associated with the technical noise observed in the system. The result also reveals several distinct trends in the Allan deviation. For the short measurement time τ , the Allan deviation scales as $\tau^{-1/2}$, which suggests a dominance of white noise at this time scale. This trend is kept up to $\tau \approx 200$ s, where the minimum of the observed Allan standard deviation is reached. After this point, the Allan deviation starts to grow up until $\tau \approx 1000$ s, and then it slightly deteriorates, reaching a local minimum. For the longer τ the Allan deviation starts to follow $\sim \tau^{1/4}$ trend. The existence of two minima can be an indication of two different sources of $1/f$ noise affecting the system stability.

The Allan deviation plot allows the determination of the time scale for which the output signal of the comagnetometer can be considered as the stationary process. This also predicts the frequency range in which the comagnetometer output signal is expected to scale inversely with measurement time. Such a scaling can be used for the increase of the SNR in searches for the phenomena presented by the deterministic constant oscillation of the non-magnetic spin couplings or by the stochastic signal with a power spectrum, which does not scale with the observation time. This observation is consistent with the experimental results presented in Fig. 6.5, where the amplitude spectrum obtained for

⁴It is rather challenging to achieve such a performance in DC measurements for a single device.

⁵As mentioned in the introduction to this section, due to the nature of the calibration process, the key statistical properties of the output signal are shared among all types of calibrated outputs.

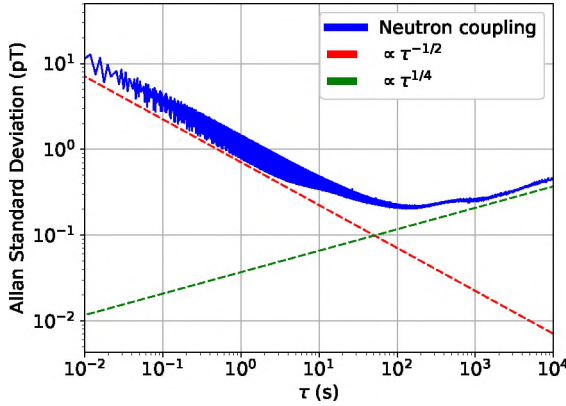


Figure 6.4: Standard Allan deviation of the Kraków comagnetometer output calibrated for the neutron exotic spin coupling. The red dashed line shows $\tau^{-1/2}$ asymptotic behaviour, while the green dashed line corresponds to $\tau^{1/4}$ asymptotic.

different observation times is shown. For the considered observation time (1 day) and frequency range above 10^{-3} Hz, the average noise value scales with the observation time τ as τ^{-1} . Such scaling is characteristic of the weakly stationary noise processes⁶. This is in agreement with the stationary bandwidth estimated by the second local minimum of the Allan standard deviation, which has been observed at $\tau \approx 1000$ s.

The increase of the Allan deviation for $\tau > 1000$ s indicates the non-stationarity of the output signal. This is consistent with the lack of the scaling of the amplitude spectrum for the $1/f$ noise observed below 10^{-3} Hz (Fig. 6.5). This supports the initial hypothesis about the presence of two independent $1/f$ noise sources. The noise source, whose impact is dominant at the frequency range between 0.001 and 0.04 Hz is an example of $1/f$ noise with the characteristic time scale $T_c = 25$ s. The characteristic time scale determines the limiting frequency below which the power spectrum of $1/f$ noise becomes constant. This is the so-called “ageing” feature of noise [144]. In the presence of noise with such characteristics, the output signal of the comagnetometer remains stationary. This is in contrast to the noise. The $1/f$ noise observed for the frequencies below 0.001 Hz, which spectrum does not scale with the observation time, and hence is an example of unlimited Brownian noise. This makes the output of the comagnetometer non-stationary on long-time scales, as shown in Fig. 6.6.

6.3. STABILITY OF THE COMAGNETOMETER CALIBRATION

In this section, the stability of the comagnetometer calibration is under investigation. Changes in the system parameters lead to inevitable modifications of its response to external perturbations. Hence, calibration stability is a particularly important factor af-

⁶The observed behaviour of the system output noise might be consistent with the moving average process [145]. Such behaviour can stem from white noise (atomic and/or magnetic) perturbing the comagnetometer spin system and being “filtered” by the system response function. Formal proof of this hypothesis requires deeper statistical analysis of the comagnetometer output, which goes beyond the scope of this work.

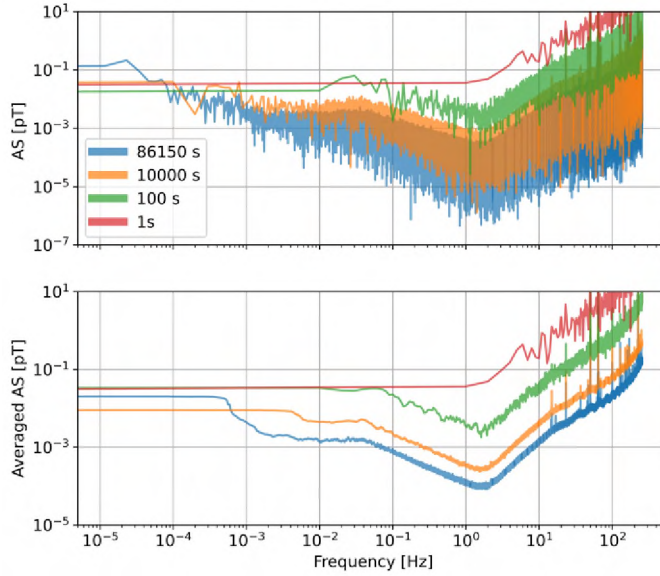


Figure 6.5: Amplitude-spectrum scaling for the calibrated neutron exotic coupling signal. The data sets are presented for four observation times: 1 s (red line), 100 s (green line), 10000 s (orange line), and 86150 s (blue line). Both the top and the bottom plots present the same sets of data. However, the data on the bottom plot for the observation times above 1 s are moving-averaged over ten points for better visibility of the average noise floor. For the frequencies above 10^{-3} Hz, the noise level scales as τ^{-1} and below 10^{-3} Hz, it does not depend on the observation time.

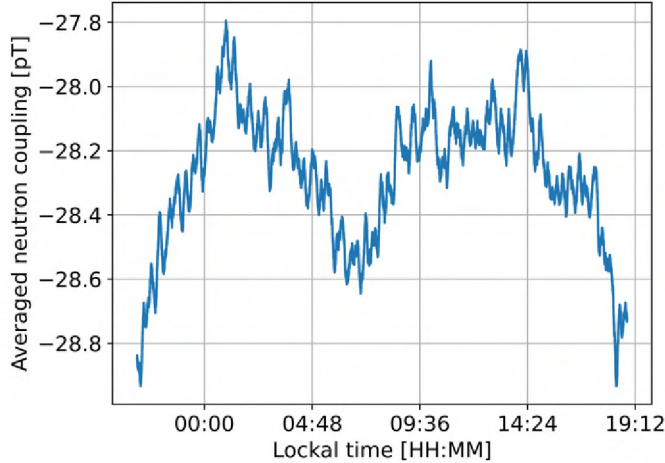


Figure 6.6: Time-averaged (350 s) signal given in effective magnetic units for neutron perturbation. The result reveals the non-stationary nature of the output signal observed on time scales longer than 350 s. In the long-time scales, the signal is dominated by $1/f$ noise.

fecting the precision of long-term measurements. As the Advanced GNOME calibration is based on the interpretation of the frequency response to the magnetic field (Sec. 5.3), here, particular attention is paid to the stability of the system response to the harmonic magnetic field perturbations at several different frequencies. To avoid the potential effects of the calibration routine on the stability analysis, this section focuses on the analysis of a raw comagnetometer signal with minimal post-processing. The post-processing of the data for this section only compensates for the limited bandwidth of the lock-in amplifier used in the measurements of parametric resonance. The test perturbation is applied along the primary sensitivity axis. The response of the comagnetometer, i.e., the amplitude of the signal at a given frequency, is estimated based on the spectrum of the output signal.

In practice, the stability of the calibration of a self-compensating comagnetometer is often limited by the stability of the detuning from the compensation point. Hence, the period of resetting the compensating field may have a direct effect on the stability of the calibration. To address this problem, the system response is considered with hourly and daily correction of the magnetometer detuning from the compensation point for comparison. In both cases, 20-day-long data sets, measured in two consecutive measurement campaigns, are under consideration.

The regular corrections to the compensation point were made by the application of 12-s-long 40-Hz modulation along the secondary sensitivity axis and measurements of an error signal used for setting the compensating field (see Sec. 4.3.2 for more details). Before each resetting of the compensation point, the frequency responses were measured by sending the train of sinusoidal pulses applied along the primary sensitivity axis. This results in 480 measurements for hourly calibrations (20 frequencies measured 24 times a day) or 20 results for the daily correction. The amplitude of the applied modulation was 100 pT in the case of the hourly correction and 25 pT in the case of the daily corrections.⁷ The data sets for hourly and daily corrected compensating fields are shown in Fig. 6.7.

The comparison of the results measured for different frequencies of the applied modulation in the case of the daily and hourly corrected compensation point is realised by consideration of the time progression of the normalised deviation of the response amplitude from its initial value $S(t, f)$, which is defined in the following way

$$S(t, f) = N \frac{a(t, f) - a(0, f)}{\sum_{t'} a(t', f)}, \quad (6.4)$$

where $a(t, f)$ denotes the amplitude of the response for the testing perturbation with frequency f at time t and N is the total number of measured data points. The experimental results for the normalised deviations are presented in Fig. 6.8.

The normalised deviations obtained for low-frequency perturbations are more noisy (compared to the high-frequency) due to a lower SNR resulting from the self-compensation regime. Hence, for better visibility, both data sets are split into “low” and “high” frequency parts, i.e., below and above an approximate value of the noble-gas Larmor frequency at the compensation point. One can note a significant linear trend (more visible in the high-frequency data, but also present for the data below 2 Hz), indicating a slow

⁷The change of the modulation amplitude was unrelated to the described measurement.

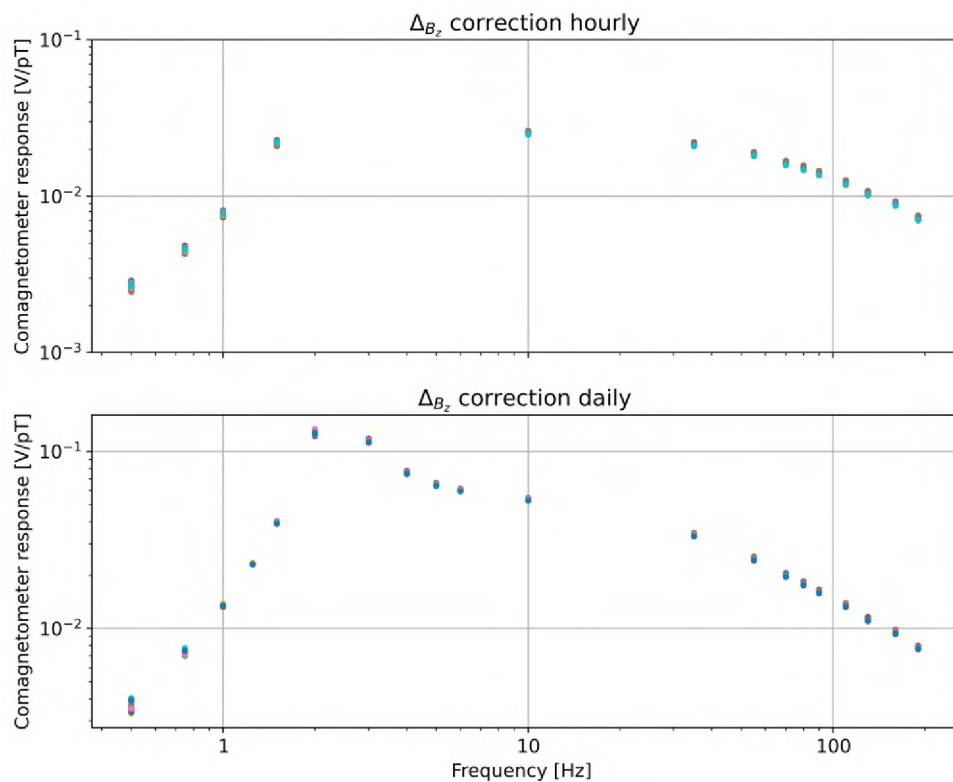


Figure 6.7: Data sets used for the analysis of the calibration stability. Each frequency response is presented with a different colour; small differences between subsequent measurement results lead to almost entire overlapping of the data points from different data sets. The top plot presents the results of 480 frequency-response measurements made hourly before resetting the compensating field to the new value. The bottom plot presents the results of 20 frequency response measurements made daily before resetting the compensation point.

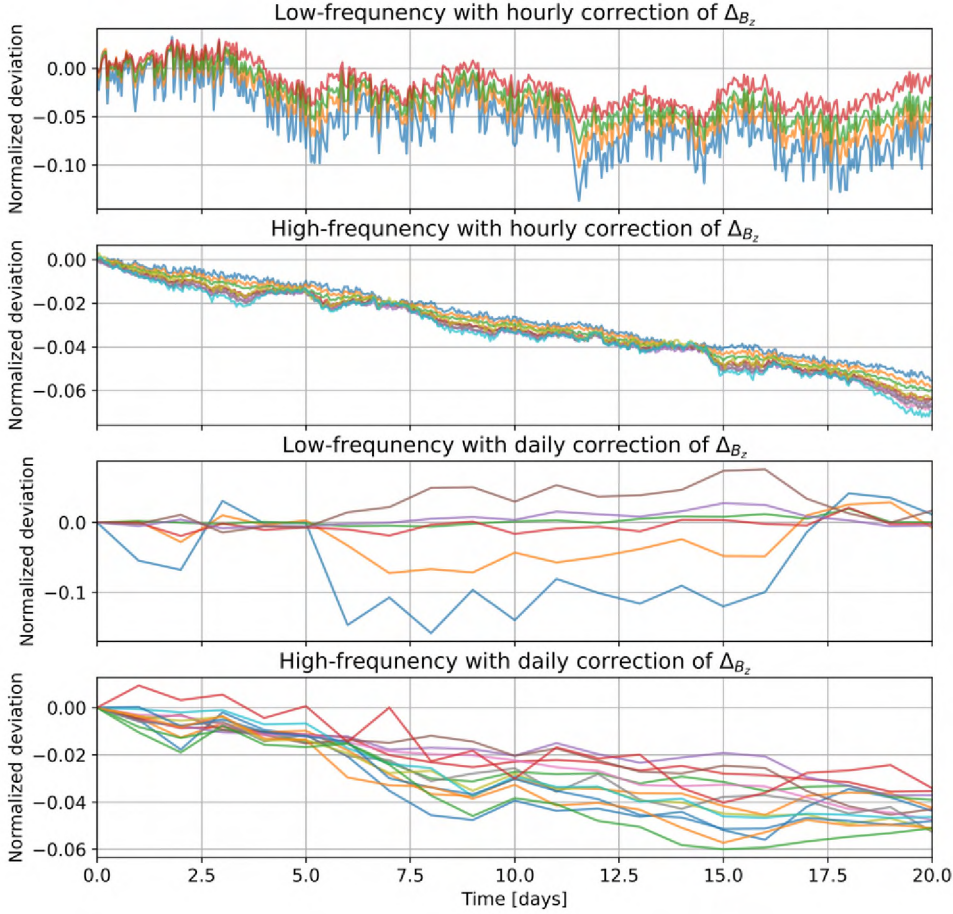


Figure 6.8: Normalised deviation from the initial amplitudes at various frequencies [Eq. (6.4)] determined based on the experimental data. Different colours of the curves represent data sets for different frequencies. The top plot presents results for perturbation frequencies below 2 Hz, while the second from the top plot shows results for the same period of resetting the compensating field but for frequencies above 2 Hz. The second from the bottom plot displays the results for the perturbation frequencies below 2 Hz and the bottom-most above 2 Hz for the compensating field, which is reset on a daily basis.

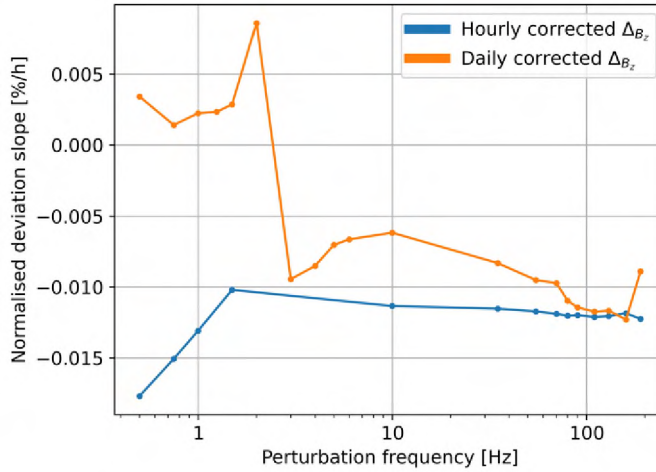


Figure 6.9: Slopes of the linear trends observed in the normalised deviations from the initial amplitude obtained with the linear fitting of the experimental results presented Fig. 6.8. The blue line presents results for the hourly corrected value of the compensation point, while the orange line presents the results for the daily corrected value of the point.

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decrease in the response amplitude. Most of the tested data sets reveal drifted by 4-8% in 20 days. For further investigation of this effect, the linear function was fitted to data sets corresponding to each tested perturbation frequency. The obtained slopes of the linear trends as a function of the perturbation frequency are presented in Fig. 6.9. In all cases, the change in the response amplitude due to the linear drift does not exceed 0.02%/h. One may also notice that low- and high-frequency response results reveal different trends. The low-frequency response slope for the hourly corrected compensation field grows linearly while it decreases at a much smaller rate for higher frequencies. In the case of the daily corrected compensation field, the contrast is even more dramatic since the observed slope for low- and high-frequency changes sign. Such a change is most probably related to the drifting alkali-metal and/or noble gas polarisation, which is qualitatively consistent with the numerical simulations based on the theoretical results from Sec. 2.2.10.

For further analysis, the experimental results from Fig. 6.8 are detrended by subtracting the linear fit from the data. The procedure allows for the distinction of the linear trend from the stochastic component of the response. The resulting time series obtained for a few representative perturbation frequencies are presented in Fig. 6.10 along with ASD of the signals. The stochastic components for both considered frequency ranges are quite similar, i.e., ASD in both cases has the same values for the comparable frequency range. This leads to the conclusion that both data sets are predominately affected by the same source of noise. The better resolution in the case of the results obtained for the hourly corrected compensating field allows distinguishing between two types of noise affecting the response. For the frequencies above about 10^{-5} Hz, the noise component is dominated by the white noise corresponding to the accuracy of the response-amplitude

determination method. At the frequency range below 10^{-5} Hz, the response is affected by $1/f$ technical noise.

6.4. OBSERVATION OF THE DAILY SENSITIVITY CHANGE

Despite the stability of the comagnetometer calibration, the output sensitivity of the comagnetometer reveals regular daily changes in the frequency between 0.1 and 2 Hz (Fig. 6.11). Specifically, between 23:00 and 5:00, the sensitivity improves by 10-30% depending on a day (Fig. 6.12). Noticeably, within the considered anomaly window, the observed sensitivity is at the same level.

In principle, such an anomaly could be related to the axion dark matter. However, a much more likely reason behind this, anticipated from the daily pattern, are trams passing the Kraków comagnetometer at a distance of about 160 m (Fig. 6.13). The time when the anomaly starts coincides with the time of the last daily tram, while the end of the anomaly coincides with the first daily tram (see vertical lines in Fig. 6.12). Moreover, the noise floor is notably higher during the daytime, and the local maxima are observed near the times typical for the high traffic in Kraków. Hence, the observed noise floor changes can be related to the vibrations of the Institute of Physics building produced by the local trams and traffic. This also allows the identification of the vibrations as the current limitation for the Kraków comagnetometer sensitivity.

6.5. SUMMARY OF THE CHAPTER

This chapter presented a detailed discussion of the sensitivity and long-term stability of the Advanced GNOME Kraków comagnetometer. The presented results reveal significant improvement in the sensitivity to the exotic proton spin couplings when compared to the first-generation GNOME stations consisting of the alkali-metal-based atomic magnetometers. The peak sensitivity to the exotic spin couplings to proton reaches $1.5 \cdot 10^{-19}$ eV/ $\sqrt{\text{Hz}}$, which is observed around 1.5 Hz. This is in addition to even higher sensitivity to exotic spin couplings to the neutron spins, which reaches $1.5 \cdot 10^{-21}$ eV/ $\sqrt{\text{Hz}}$ around the same frequency. It is worth noticing that neutron spin couplings were inaccessible with regular magnetometers used for the first-generation GNOME stations. The current sensitivity of the Advanced GNOME comagnetometer is mostly limited by the vibrations of the building hosting the experimental setup, which was proved by the observation of the noise-level change correlated with the schedule of the trams passing the nearest to the experimental setup tram stop.

The consideration of the statistical properties of the comagnetometer output signal reveals that the noise of the system can be considered weakly stationary for the observation times of about 1000 s. The system noise is dominated by a non-stationary $1/f$ noise for longer observation times. While the optimal observation time, for which the absolute minimum of the Allan standard deviation is equal to about 200 s.

The long-term stability of the system allows uninterrupted operation of the comagnetometer for 24-h period. After that period, the compensating field must be reset in order to avoid systematic effects related to the changes in the noble-gas polarisation level. This sets the current upper limit on the duty cycle of the Kraków Advanced GNOME comagnetometer.

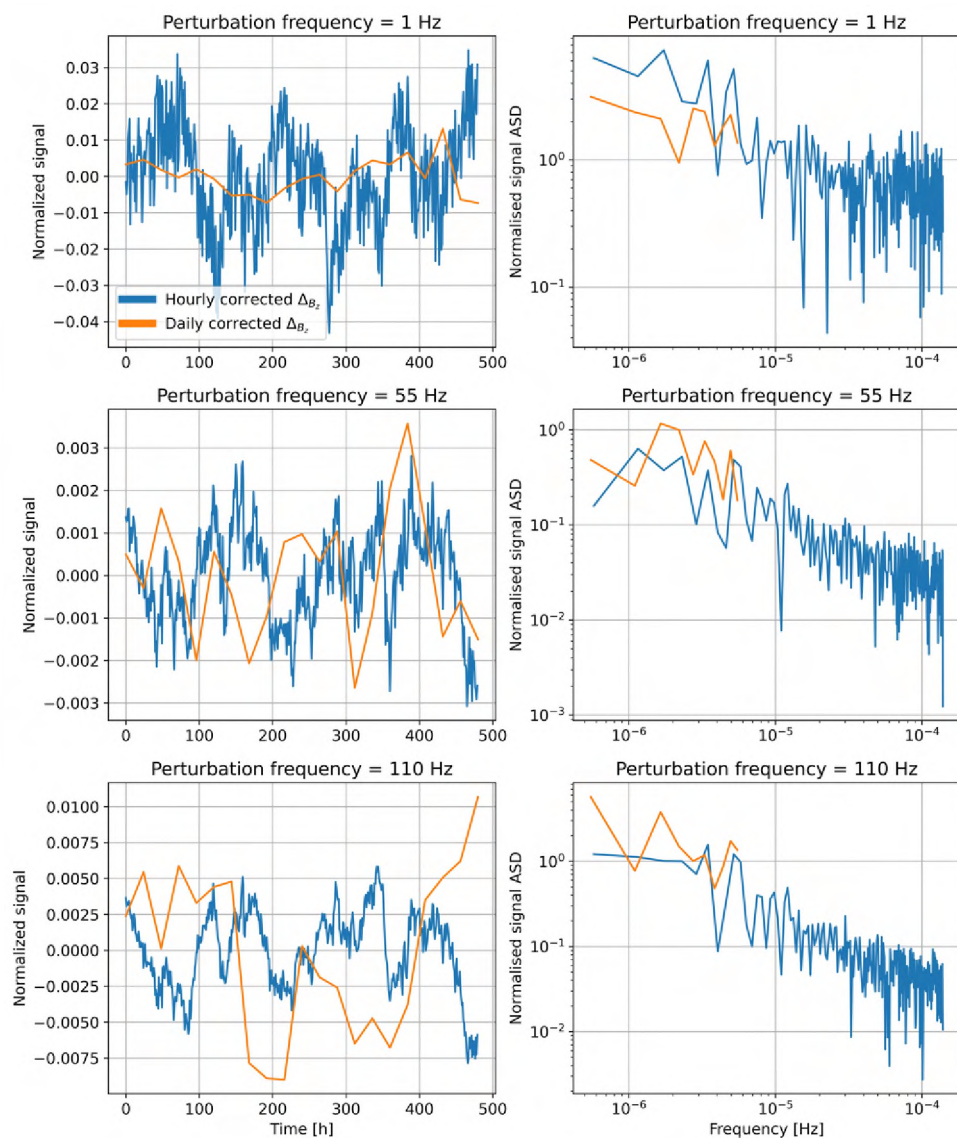


Figure 6.10: Left column shows examples of detrended normalised deviations of the response amplitude observed for the perturbations with a frequency of 1 Hz (top), 55 Hz (middle), and 110 Hz (bottom). The right column presents ASD for each deviation time series. Each plot contains data for both hourly (blue lines) and daily (orange lines) corrected leading fields.

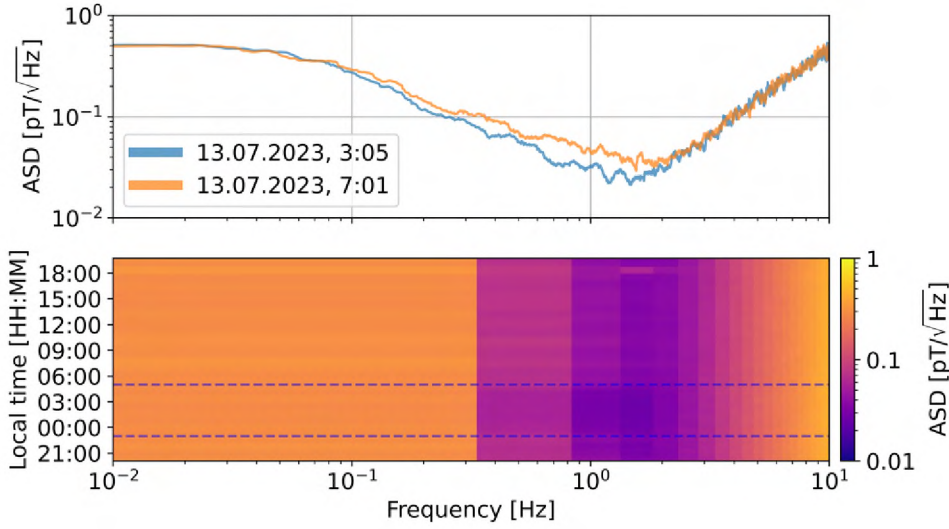


Figure 6.11: Example of the Kraków-comagnetometer sensitivity changes at the frequency range between 0.1 and 2 Hz over a single day. The top plot presents the sensitivity estimated from 100-s-long data sets recorded at 3:05 (the blue line) and at 7:01 (the orange line) on July 13, 2023. The sensitivity curves have been subjected to the moving average over 50 data points for illustration purposes. The bottom plot shows the spectrogram with the sensitivity, which covers the 24-h cycle, which starts at 20:00 on July 12, 2023. Each presented data bin represents the average amplitude spectrum density of the signal. The blue dashed lines show the time period when the change of sensitivity improves.

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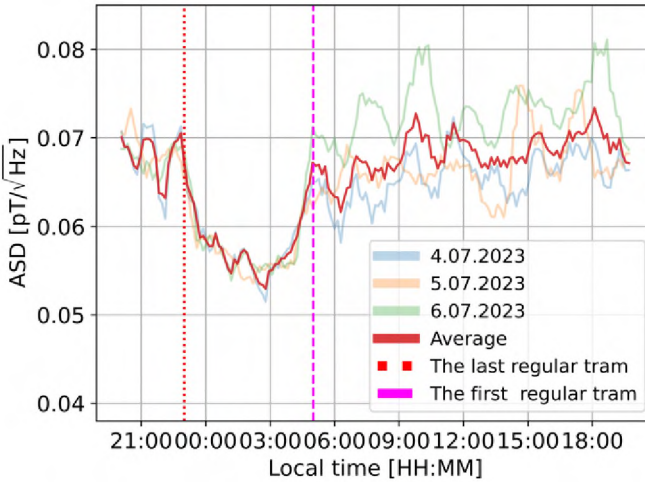


Figure 6.12: Comparison of daily cycles for the average ASD of the neutron-coupling sensitivity observed within 0.5-Hz window centred at 0.5 Hz for the data sets starting at 20:00 of July 12-14, 2023 (blue, orange, and green lines, respectively). The red solid line shows the average over all three data sets. The dotted red line shows the scheduled time of the last regular daily tram, while the dashed magenta line shows the time of the first regular daily tram passing the nearest to the Kraków GNOME station tram stop.

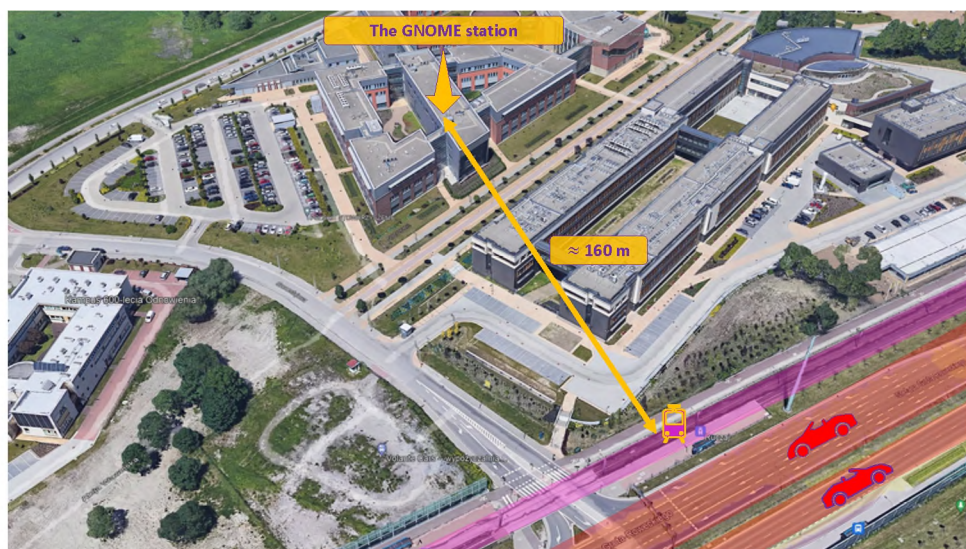


Figure 6.13: Schematics illustrating the position of the Krakow Advanced GNOME station with respect to the tram lines and four-lane road located near the Campus of the Jagiellonian University in Kraków. The distance between the experimental setup and the nearest tram station is approximately 160 m. The vibrations produced by the tram line and heavy traffic on the road are the main limitations of the peak sensitivity for the Kraków comagnetometer. The variation of the noise level observed in the experiment is in good agreement with the tram schedule (Fig. 6.12).

7

CONCLUSIONS

7.1. THESIS SUMMARY

This work presented a comprehensive study of a new type of sensor to search for time-dependent exotic spin couplings. This device was based on a concept of a self-compensating comagnetometer proposed in Ref. [57], which was previously used for investigating time-independent spin couplings. In this work, however, the emphasis was placed on the utilisation of a similar device for measurements of transient and oscillatory couplings. This required the development of an independent theoretical description of the device operation, as well as its construction and optimisation. Due to these reasons, the thesis thoroughly discussed both the theoretical aspects of the dynamics of coupled spin systems, which forms the basis of the constructed sensor and the experimental aspect of the work in which the device was built, and its operation was analysed versus various experimental parameters.

7.1.1. INVESTIGATIONS OF TIME-DEPENDENT SPIN COUPLINGS

A significant part of this dissertation was dedicated to the formulation and discussion of the theoretical description of the self-compensating comagnetometers in the generic searches for time-dependent spin couplings. The presented framework was based on the linearised response of the coupled spin system, which allowed the definition of convenient formalism. The formalism enabled the development of suitable data-analysis routines, which were later used to process experimental signals. In particular, it was used to perform a full spectral reconstruction of the system response to non-magnetic spin couplings based on the sensor's magnetic response. Here, the strong spectral dependence of the sensitivity was demonstrated, which was in contrast to the previous studies where a flat frequency response of the comagnetometer was assumed. Accounting for frequency-dependent sensitivity could be particularly important when the perturbation frequency is close to the Larmor frequency of the nuclear polarisation. In this respect, this may be crucial when such global transient signals as the ones induced by axion domain wall, axion wind, Q-balls, etc., are considered. It is noteworthy that this new ap-

proach is suitable for long-term searches for time-dependent exotic spin couplings as the wide-spectrum reconstruction could be performed with a single square pulse of the magnetic field, which can last for a couple of seconds and can be easily implemented experimentally. The method provided an estimation of the comagnetometer response at the compensation point but its validity was also demonstrated for non-zero detuning from the compensation point, simultaneously allowing for estimation of the actual detuning from the point. This can be used for re-tuning the compensation field to the compensation value without additional manipulations of the system. Such a reduction in the time required for the maintenance of the system improved the continuity of the measurements.

7.1.2. SENSOR FOR THE ADVANCED GNOME

In addition to general theoretical and experimental investigations of the comagnetometer, this thesis also provided a description of the experimental systems that were used in the first Advanced GNOME stations located in Kraków and Mainz. One important feature of the constructed devices was their ability to operate in a two-axis-sensitivity mode. This was possible due to the application of the parametric modulation of the atoms. It is noteworthy that operation in such a mode means that a single Advanced GNOME sensor is effectively equivalent to four first-generation GNOME sensors. Moreover, the detection of the alkali-metal polarisation with the parametric modulation also enabled an effective suppression of the $1/f$ noise, which stems, for example, from air motion and turbulence in the optical path of the probe beam.

Using the results obtained during the realisation of the dissertation, it was demonstrated that, up to the uncertainty of the estimation, the system response remained stable within a 24-h run (Sec. 6.3). This proved that for the Advanced GNOME science runs, the resetting of the compensation point and the calibration check can be repeated only once a day. This significantly improves the continuity of the measurements, which is of paramount importance for the analysis of various theoretical models.

The analysis of the Kraków comagnetometer and successive analysis of its sensitivity performed within this dissertation showed the significant (by many orders of magnitudes) improvement of the sensitivity of the GNOME searches to the proton exotic spin couplings with respect to the first generation GNOME sensors (magnetometers). Moreover, as the comagnetometer was based on the ^3He polarisation, it was also sensitive to the neutron spin couplings. This allowed for the extension of the GNOME search to the neutron coupling, which was not previously possible.

The analysis also revealed directions for further sensitivity improvement of the searches in the Kraków comagnetometer. Specifically, the measurements showed the daily changes in the noise level. These changes eventually enabled the identification of the main contribution to the noise, which originated from vibrations induced by the nearby traffic. This indicated that the sensitivity can be enhanced through the improvement of the anti-vibration system and its better isolation from the floor.

Another important aspect of the comagnetometer developed was related to its ability to determine and hence compensate magnetic-field gradients. As the gradients may affect the operation of the comagnetometer, compensation of the gradients enabled a stable operation of the systems without the necessity of frequent resetting the compen-

sation point. This led to the significant improvement of the system duty cycle, making it suitable for long-term continuous searches.

7.2. FUTURE WORK

The natural continuation of the work presented in this thesis lies in the long-term measurements aiming at the detection of the global exotic spin couplings with the Advanced GNOME network. The first measurements with a “network” consisting of two advanced sensors (Kraków and Mainz) are ongoing at the time of preparation of this thesis. Moreover, the extension of the network with more self-compensating comagnetometers is envisioned in the near future.

The uncertainty and great versatility of the theoretical predictions of the exotic spin couplings make it difficult to identify the optimal observation time and the ideal configuration of the network. Because of that, further development of the GNOME network may prioritise long-term autonomous measurements, in which data will be collected over multiple years or even a decade. Ideally, such measurements should ensure sensitivity in all-direction with minimal requirements for intervention in the system operation. This is a shift in the experimental paradigm from an atomic-physics style experiment, where the experimental endeavour is planned in the time span of months and single years (scientific runs), towards an astronomical-observations approach, where the data are collected continuously for extended periods of time for the future evaluation. In turn, this will allow the utilisation of GNOME data as another source of information for multi-messenger astronomy with potentially useful information for cosmology/astronomy and particle physics. Such an approach requires proper data collection and storage procedures, along with standardised protocols for data validation, station maintenance, and sensor calibration. The experience of the GNOME collaboration and the results of this work opens a direct path for the realisation of such an approach with the Advance GNOME network.

The focus of future work on the Advanced GNOME comagnetometers should also be directed towards the improvement of the sensitivity of each individual sensor, as the current sensitivity of both the Kraków and Mainz stations is at least an order of magnitude worse than the best results published for the self-compensating comagnetometers. As mentioned above, the sensitivity can be improved by reducing of technical noise (e.g., vibration), but also by the implementation of atomic cells with a higher concentration of the noble gas¹ and improved sphericity.

Finally, further investigation is required on the issue of the multi-axes sensitivity and directional ambiguity of the self-compensating comagnetometer measurements, which have been highlighted in this work. This work treated the readouts for two sensitivity axes independently. However, a comprehensive approach for the comagnetometer readout requires the development of joined two-channel signal processing for the readouts corresponding to both sensitivity axes. Such signal processing must allow generic treatment of the two-component signal from the comagnetometer in a way which ensures precise identification of the direction of the measured perturbation.

¹Notably, the system with the best-known sensitivity of the self-compensating comagnetometers has four times higher concentration of ³He atoms in the atomic cell.

7.2.1. FINAL REMARKS

All the aspects of the comagnetometric technique developed within the dissertation contribute to a qualitative change in the application of the devices for long-term operations and the exploration of both transient and oscillatory spin couplings. Specifically, the presented knowledge can be used in the construction of the new sensors in other GNOME stations, but it also can be a basis for further research. With this respect, this dissertation constructively contributes to the development of atomic comagnetometry, and, more generally, to the searches for ultralight bosonic dark matter.

EPILOGUE

This is an optional epilogue.

A

APPENDIX

A.1. THE PARAMETERS FOR THE SIMULATED DATA

The list of the parameter values used for illustration of the solutions for the comagnetometer response presented in Chapter 2 is provided in Table A.1.

Parameter	value	units
γ_F	$-2\pi \cdot 467$	rad/s
γ_n	$-2\pi \cdot 3.24$	rad/s
Y_z	0.5	
ζ_z	0.03	
ω_e	$-2\pi \cdot 76$	rad/s
ω_n	$-2\pi \cdot 8$	rad/s
ΔB_z	0	mG
R_e	600	1/s
R_n	10^{-5}	1/s

Table A.1: The list of the parameters used for the illustration of the analytical solutions for the comagnetometer response presented in Chapter 2.

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