Self-Compensating Co-Magnetometer vs. Spin-Exchange Relaxation-Free Magnetometer: Sensitivity To Nonmagnetic Spin Couplings

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Self-compensating co-magnetometer vs. spin-exchange relaxation-free magnetometer: sensitivity to nonmagnetic spin couplings

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ABSTRACT  

Searches for pseudo-magnetic spin couplings require implementation of techniques capable of sensitive detection of such interactions. While Spin-Exchange Relaxation Free (SERF) magnetometry is one of the most powerful approaches enabling the searches, it suffers from a strong magnetic coupling, deteriorating the pseudo-magnetic coupling sensitivity. To address this problem, here, we compare, via numerical simulations, the performance of SERF magnetometer and noble-gas-alkali-metal co-magnetometer, operating in a so-called self-compensating regime. We demonstrate that the co-magnetometer allows reduction of the sensitivity to low-frequency magnetic fields without loss of the sensitivity to nonmagnetic couplings. Based on that we investigate the responses of both systems to the oscillating and transient spin perturbations. Our simulations reveal about five orders of magnitude stronger response to the neutron pseudo-magnetic coupling and about three orders of magnitude stronger response to the proton pseudo-magnetic coupling of the co-magnetometer than those the SERF magnetometer. Different frequency responses of the co-magnetometer to magnetic and nonmagnetic perturbations enables differentiation between these two types of interactions. This outlines the ability to implement the co-magnetometer as an advanced sensor for the Global Network of Optical Magnetometer for Exotic Physics searches ( GNOME), aiming at detection of ultra-light bosons (e.g., axion-like particles).

Introduction  

In optically pumped magnetometers (OPMs), measurements of external magnetic fields rely on detection of energy-level shifts/spin precession arising from Zeeman interaction1. However, if nonmagnetic interactions similarly affect the energy levels, OPMs can also be used to detect those interactions. This enables the application of optical magnetometry to searches for anomalous spin-dependent interactions.  

Currently, OPMs are used to search for physics beyond the Standard Model in a variety of experiments (see, Ref.2 and references therein). A particular example of such OPMs application is the search for microscopic-range spin-dependent interactions, indicating a possibility of existence of axion-like particles (ALPs), which are one of prime candidates for the dark matter2. OPMs are also used to search for transient nonmagnetic spin couplings, which could arise due to interaction with macroscopic objects made of ALPs, in particular, Q-balls3, topological defects (e.g., domain walls) of ALP field4, or ALP-field pulses generated in cataclysmic astrophysical events (e.g., black-hole mergers)5. These transient couplings are targeted by the Global Network of Optical Magnetometers for Exotic physics searches ( GNOME)6–8. Heretofore, the GNOME consists of various OPMs, originally developed for ultra-sensitive magnetometry in globally distributed locations. This leads to several challenges when searching the data for global transient signals. First, due to a different nuclear-spin content of atoms used in specific sensors, they are characterised with different sensitivity to exotic spin couplings9 (coupling to protons and neutrons could be, in general, different). Second, the implemented OPMs are characterised with different bandwidths, sensitivities, and local noise floors, which complicates data analysis10. Third, the magnetometers were designed to maximise the sensitivity to magnetic fields. Uncontrolled and uncompensated magnetic-field perturbations are detrimental to the sensitivity to other couplings. These issues triggered work to upgrade conventional OPMs in the GNOME with a sensor less sensitive to magnetic fields but highly sensitive to nonmagnetic spin couplings.

A specific example of a sensor, being predominantly sensitive to nonmagnetic spin couplings, hence well suited for searches
for physics beyond the Standard Model, is an alkali-metal-noble-gas co-magnetometer originally developed by Romalis and coworkers\cite{11-13} and later studied extensively by other researchers\cite{14-18}. Such a system operates based on coupled evolution of the magnetizations of noble gas (NG) and alkali-metal (AM) vapour. Both can achieve a high percentage of polarization as AM can be optically pumped, whereas polarisation of the NG can be generated via spin-exchange collisions with the optically polarized AM\cite{19}. In the co-magnetometer, the vapour cell is heated above 150°C to achieve a sufficiently high AM density, such that relaxation due to spin-exchange collisions, which is one of the main mechanisms of AM polarisation relaxation and hence one of a limiting factor to spin-coupling sensitivity, is suppressed. This is the so-called Spin-Exchange Relaxation Free (SERF) regime\cite{20}. Furthermore, for the alkali-metal-noble-gas co-magnetometer the effect of low-frequency magnetic drifts can be suppressed by application of a carefully chosen bias magnetic field. If the bias field is approximately equal to the sum of AM and NG magnetization fields, the system retains high sensitivity to both electron and nuclear nonmagnetic spin couplings, but becomes insensitive to low-frequency magnetic-field changes. Such self-compensating co-magnetometers have already been used for tests of the Lorentz symmetry\cite{21-23}, setting limit on the neutron coupling to light pseudoscalar particles\cite{24}, and spin-mass interaction of fermions\cite{25}. In all of those applications, however, the signals of interests had low frequencies (typically below a few Hz), for which the response to magnetic fields is almost entirely suppressed and the system is only sensitive to nonmagnetic spin couplings. Since the GNOME targets transient signals, a question of the co-magnetometer’s applicability to such searches is important and well motivated. Additional interest in co-magnetometer systems, in the context of searches for transient effects, arises from the possibility for quantitative distinguishing between magnetic and nonmagnetic transients. This was originally considered for precise rotation sensing with NG-AM co-magnetometers\cite{12} but can be extended for other scenarios.

In this work, we analyse the alkali-metal-noble-gas co-magnetometer in the context of its response to time-dependent electron and nuclear spin perturbations, and we compare the results with the response of the AM SERF magnetometer. Our theoretical analysis is based on numerical solution of differential equations describing the coupled evolution of the AM and NG. We use Ordinary Differential Equations (ODE) to simulate the response of atoms to spin perturbations in the absence of noise. The discussions concern the responses of both, SERF magnetometer and co-magnetometer, to magnetic fields and pseudo-magnetic spin perturbations, where in the latter case we independently consider the effects of electron, proton, and neutron spin perturbations. These allow us to determine the frequency and phase responses of both devices. Finally, we compare the response of the co-magnetometer and SERF magnetometer to transient effects of both magnetic and nonmagnetic nature. We show that differences between the co-magnetometer frequency responses for different spin perturbations allows identification of nonmagnetic transient effects even with a single sensor. These additional signatures of the nonmagnetic transient effects can be utilised to decrease the false-positive event rate in searches with the GNOME network.

Methods

Numerical models
In this section, we describe the theoretical models used to simulate the response of the SERF and co-magnetometers to magnetic and nonmagnetic spin perturbations.

SERF magnetometer model
In a conventional SERF magnetometer, a spin-zero NG can be used as a buffer gas. The buffer gas limits diffusion of the atoms towards the cell walls (the AM atoms are depolarised in wall collisions), which increases the polarisation lifetime but, it does not produce any magnetisation. Thereby, to simulate a response of such a magnetometer to a spin perturbation, we implement an approach based on the solutions of the Bloch equation\cite{26} with inclusion of nonmagnetic spin couplings (for more details see Supplementary Information (SI)). The equation describing the AM polarisation $P$ of atoms subjected to an external magnetic field $B$ and circularly polarised light can be written as

$$\frac{dP}{dt} = \frac{1}{q} \left[ \gamma_e (B + b_e) \times P + (q - 1) \gamma_e b_{N}^{AM} \times P + (s - P) R_p - RP \right]$$

(1)

where $\gamma_e$ is the electron gyromagnetic ratio, $s$ is the optical pumping vector, $R_p$ is the pumping rate, $R$ is the polarisation-relaxation rate, $q$ is the slowing-down factor, which is a function of the nuclear spin of the AM and its polarisation $P$. The vectors $b_e$ and $b_{N}^{AM}$ are the nonmagnetic electron and nuclear spin perturbations, respectively, given the magnetic units (pseudo-magnetic field).

Co-magnetometer model
In the co-magnetometer, a polarised AM and NG (in this case NG with nonzero nuclear spin is used, so its nuclei can be polarised) occupy the same volume inside of a spherical glass cell. Then, the response of the co-magnetometer to magnetic and
nonmagnetic perturbations is determined by a set of coupled Bloch equations (the so-called Bloch-Hasegawa equations)\textsuperscript{11} with inclusion of nonmagnetic spin perturbations (for more details see the SI)

\[
\begin{align*}
\frac{d\mathbf{P}^e}{dt} &= \frac{1}{\tau} \left[ \gamma_e (\mathbf{B} + \mathbf{b}_e + \lambda M^P \mathbf{P}^n) \times \mathbf{P}^e + (q - 1) \gamma_e b_{N}^{AM} \times \mathbf{P}^e + (s - \mathbf{P}^e) R_P - R_e \mathbf{P}^e \right], \\
\frac{d\mathbf{P}^n}{dt} &= \gamma_n (\mathbf{B} + \mathbf{b}_{NG}^N + \lambda M^P \mathbf{P}^e) \times \mathbf{P}^n + (\mathbf{P}^n_0 - \mathbf{P}^n) R^n,
\end{align*}
\]

where \(\mathbf{P}^e\) and \(\mathbf{P}^n\) stand for the electron polarisation of the AM and nuclear polarisation of the NG, respectively, \(\lambda\) is the coupling-strength factor for interaction between the two polarisations, \(M^P\) and \(M^n\) are the maximal possible magnetisation of AM and NG, and \(\gamma\) is the nuclear gyromagnetic ratio of NG. \(R^e\) and \(R^n\) are the electron and nuclear polarisation-relaxation rates, respectively. The nonmagnetic nuclear perturbation of the NG spins is denoted \(\mathbf{b}_{NG}\).

In order to fully capitalise on the co-magnetometric capabilities (self-compensation of slow magnetic fields), here we consider the operation in the self-compensating regime, which is achieved when a static magnetic field \(\mathbf{B}_c\)

\[
\mathbf{B}_c = - (\lambda M^P P^p_0 + \lambda M^n P^n_0) z,
\]

is applied to the system\textsuperscript{11} (here we assumed that the initial AM and NG polarisations are oriented along the \(z\) axis).

**Nuclear spin content and sensitivity to neutron and proton spin perturbations**

Due to the composite nature of atomic nuclei, the nuclear response may arise due to coupling to protons, neutrons, or a combination of both. Therefore, the effective pseudo-magnetic fields \(\mathbf{b}_{N}^{AM}\) and \(\mathbf{b}_{N}^{NG}\) can be divided into parts: \(\mathbf{b}_p\) affecting the protons and \(\mathbf{b}_n\) acting on the neutrons\textsuperscript{9}

\[
\mathbf{b}_N^i = \mathbf{b}_n^i + \mathbf{b}_p^i,
\]

where \(i\) may stand for either AM or NG. These pseudo-magnetic fields are determined by the nonmagnetic field \(\Xi\) and coupling constants \(\chi_n\) and \(\chi_p\) characterising the coupling to neutrons and protons, respectively

\[
\begin{align*}
\mathbf{b}_N^{AM} &= - \frac{\sigma_j^{AM}}{\mu_B g_S} \chi_j \Xi, \\
\mathbf{b}_N^{NG} &= \frac{\sigma_j^{NG}}{\mu_N g_K} \chi_j \Xi,
\end{align*}
\]

where \(j\) indicates either proton or neutron, \(\sigma_j\) corresponds to the proton or neutron fraction of the nuclear spin polarisation of AM and NG (denoted with upper indices), \(\mu_B\) is the Bohr magneton, \(\mu_N\) is the nuclear magneton, \(g_S\) is the AM Landé factor, and \(g_K\) is the NG nuclear spin \(g\)-factor. Equations (5) show that the effective pseudo-magnetic fields for the NG are generally different from those for the AM. Thus, for the simulations or interpretation of results, it is convenient to introduce scaling factors \(\eta_j\) which allows comparison between the response of the SERF magnetometer and the co-magnetometer to nonmagnetic spin couplings of the same strength

\[
\mathbf{b}_N^{AM} = \eta_j b_j^{NG},
\]

where the scaling factors are

\[
\eta_j = - \frac{\sigma_j^{AM}}{\sigma_j^{NG}} \frac{\mu_N g_K}{\mu_B g_S}.
\]

In the simulations presented in this paper, we consider the responses of the magnetometers to the perturbation of the same coupling strength. This approach takes into account the scaling factors defined in Eq. (7). One can find a more detailed discussion of the nonmagnetic spin couplings in the SERF and the co-magnetometer in the SI.

**Simulation parameters**

In the case of both, the SERF and co-magnetometer, the spin polarisation is monitored through measurements of the AM-polarisation projection on a given direction (here it is the \(x\) axis). As in both systems the atomic species are initially polarised along the \(x\) axis, both magnetometers are primarily sensitive to perturbations along \(y\) (the sensitive direction is determined by the torque generated by the external fields, rotating the spins around the sensitive direction). Specifically, it can be shown from Eqs. (1) and (2) that the magnetic or pseudo-magnetic field applied along \(y\) rotates the initial polarisation in the \(xz\) plane, which
results in a change of the polarisation projection on the x axis. At the same time, a field applied along the x axis generates rotation in the yz plane, therefore the projection of the polarisation on the x axis remains unchanged.

In the simulations, we assume that the SERF magnetometer operates using ³⁹K atoms and the co-magnetometer operates using a ³⁹K-³He mixture. Parameters of potassium vapour are chosen to be exactly the same for both systems, so we can properly compare the sensitivity. The concentration of the alkali metal is equal to 10¹⁴ cm⁻³, which corresponds to saturated atomic K vapour at 190°C. The concentration of ³He is 10²⁰ cm⁻³, which corresponds to 3.5 amg. The assumed relaxation rates 600 s⁻¹ for potassium and 5·10⁻⁵ s⁻¹ helium, corresponding to a lifetime of about 53 min, well reproduce the experimental conditions. The steady-state polarisation of the AM is 0.5, which ensures the highest amplitude of the co-magnetometer response, and also corresponds to typical experimental conditions. Polarisation of the NG is chosen to be 0.05, which corresponds to typical experimental conditions. These parameters lead to the compensation-field value of −131 nT. The other simulation parameters (a complete list) is given in the SI.

In case of ³⁹K and ³He the relation between the effective magnetic fields for the AM and the NG generated by the same nonmagnetic perturbation defined in Eq. (7) leads to the following scaling factors

\[ b_{p}^{³⁹K} = \eta_{b} b_{p}^{³He} \approx 10^{-3} b_{p}^{³He}, \]

\[ b_{n}^{³⁹K} = \eta_{b} b_{n}^{³He} \approx 10^{-5} b_{n}^{³He}, \]

where the nuclear spin content information was taken from Ref. ⁹.

**Results and discussion**

**Frequency response to different spin perturbations**

In this section, we compare responses of the SERF and co-magnetometer devices to various spin perturbations. We analyse the response of the devices by investigating their signals when perturbed with an either magnetic or nonmagnetic periodic y-oriented field A

\[ A = A_{0} \sin(2\pi vt) y, \]

where A₀ is the amplitude and ν is the frequency of the field. For the simulations, we assume that the amplitude of the perturbation is low enough so that the co-magnetometer continuously operates in the self-compensating regime. To analyse the response, the simulated data are fitted with the function

\[ S = S_{0} \sin(2\pi ft + \phi), \]

where S₀, ϕ and f are, respectively, the amplitude, phase, and frequency of the fitted signal. To avoid distortions in the fit, we ignore transient phenomena at the beginning of the simulations and just fit the dynamical steady-state data.

The fitted amplitude and phase of the signals arising due to magnetic, electron nonmagnetic, neutron nonmagnetic, and proton nonmagnetic perturbations are shown in Fig. 1.

A key feature of the co-magnetometer in searches for pseudo-magnetic spin couplings is its suppressed sensitivity to low-frequency magnetic-field perturbations. This is clearly visible in the data presented in Fig. 1(a). At the lowest frequencies, the response amplitude of the co-magnetometer to the magnetic field is roughly four orders of magnitude lower than the response of the SERF magnetometer. This difference decreases at higher frequencies reducing to zero at about 4 Hz. Since compensation is provided by the NG, which adiabatically follows the field changes, and, at the compensation point, the atoms are only experiencing a field proportional to their own magnetisation, \[ B' \approx B_{t} + \lambda M_{t}' \overline{P}_{0} = \lambda M_{t}' \overline{P}_{0}, \] the frequency at which the SERF and co-magnetometer magnetic responses are equal is determined by the NG Larmor frequency. Shifting the compensation point towards higher frequencies requires increasing the NG magnetisation, which can be achieved by either increasing the NG concentration or the polarisation. Both may be challenging experimentally. For higher frequencies the response of both systems has the same amplitude, since outside of the self-compensating regime the co-magnetometer response is predominantly determined by the AM. Thereby, the AM polarisation in the co-magnetometer starts to behave in the same way as the free-AM polarisation in a usual SERF magnetometer.

There is a remarkable difference in the phase response of the two devices [Fig. 1(a)]. For magnetic field frequencies below the NG Larmor frequency (4 Hz), the response of the co-magnetometer is phase shifted by about π, and it decreases sharply above that frequency, eventually becoming similar for both devices. For lower frequencies, the difference is due to the NG that compensates the magnetic field and no such compensation is present in the SERF magnetometer. For higher field frequencies the magnetic response of both devices is determined by the AM, so the observed dependencies are similar.
Figure 1. Simulated amplitude and phase of the SERF (dashed lines) and co-magnetometer (solid lines) responses (normalised polarisation along x) for magnetic (a), electron nonmagnetic (b) neutron nonmagnetic (c), and proton nonmagnetic (d) perturbations.

When analysing the response of the co-magnetometer to nonmagnetic nuclear perturbation, it should be first noted that, unlike in the case of magnetic perturbation, the nonmagnetic nuclear perturbations are not compensated. Therefore, in that case, there is no reduction of the response amplitude at lower frequencies. To the contrary, the amplitude response to pseudo-magnetic nuclear perturbations of the co-magnetometer is significantly stronger than the response of the SERF magnetometer [Fig. 1(c) & (d)]. In particular, for frequencies below 4 Hz, the co-magnetometer response is roughly five orders of magnitude stronger for the neutron nonmagnetic perturbation [Fig. 1(c)], about three orders of magnitude stronger for the proton perturbation [Fig. 1(d)] and even though it deteriorates for higher frequencies, it still remains significantly larger than for the SERF system. Because of the high concentration of the NG, the response of the co-magnetometer to the nonmagnetic nuclear perturbation is predominantly determined by the gas. In turn, the large concentration difference between the NG and AM concentration (about six order of magnitude) is responsible for much higher sensitivity of the former to the nonmagnetic nuclear couplings. Moreover, the high magnetisation of the NG atoms in the co-magnetometer ensures efficient transfer of the NG-spins perturbation to AM polarisation through the strong interaction between two species. Therefore, the mediation of the nuclear coupling by the NG magnetisation for the co-magnetometer leads to a significant increase of the amplitude of the response to nonmagnetic nuclear perturbations. An additional cause of the difference in the response to nuclear perturbations stems from the different nuclear spin contents of the $^{39}$K and the $^{3}$He nuclei. Contribution of proton polarisation in $^{39}$K is roughly four times larger than in the case of $^{3}$He. In contrast, the neutron polarisation has an about 24 times stronger effect on the nuclear polarisation of $^{3}$He, than on the atomic polarisation of $^{39}$K. The difference in neutron and proton fraction in $^{3}$He also leads to different response amplitudes of the co-magnetometer for proton and neutron perturbations.

The phase response of the co-magnetometer is similar for both the neutron and proton pseudo-magnetic perturbations. Specifically, below 4 Hz, the phase shift between perturbation and response is close to $\pi$ and it drops to about zero for
higher frequencies. While for frequencies above 100 Hz the two responses differ, it should be noted that this frequency range is well beyond the bandwidth of the co-magnetometer, where amplitude of the response drops by several orders of magnitude. At the same time, the phase response of the SERF magnetometer is the same for magnetic and nuclear nonmagnetic perturbations, being zero at lower frequencies and monotonically shifting toward $-\pi/2$ for frequencies beyond the bandwidth of the magnetometer.

The response of both devices to the electron nonmagnetic perturbation is similar at most of the frequencies with a distinct exception of the frequency corresponding to the NG Larmor frequency (4 Hz) [Fig. 1(b)]. Such a behaviour is not surprising since, in both cases, the electron coupling perturbs the AM electron spins. On the other hand, differences in the response of the systems at the NG Larmor frequency arise due to the coupling between the perturbed AM and the NG atoms.

Co-magnetometer response to transients

It was shown in the previous section that the response of the co-magnetometer to nonmagnetic nuclear couplings is much stronger than that of the SERF magnetometer. Therefore, below we only focus on analysis of the response of the co-magnetometer to transient magnetic and nonmagnetic perturbations.

As a generic example, we take a temporal Lorentzian perturbation of the amplitude $\Lambda_0$ and half-width $\Delta t$, centred at the time $t_0$, which is directed along the $y$ axis (this can be easily generalised for any pulse shape)

$$\Lambda_t = y \frac{\Lambda_0 \Delta t^2}{(t - t_0)^2 + \Delta t^2}. \quad (11)$$

Such a definition allows keeping the amplitude of the pulse constant while varying its width (note that here the energy of the pulse is not preserved). Since here we are only interested in temporal parameters of the response, the proton and nuclear couplings are not considered independently but they are treated in as a generic nuclear coupling.

In the previous section, we have shown that the co-magnetometer frequency responses for magnetic and nonmagnetic spin couplings are different. In particular, the compensation of low-frequency magnetic fields, the mechanism present in the co-magnetometer, results in a suppression of low-frequency components of the pulse. This may affect the shape of the response of the co-magnetometer to magnetic pulses and is well visible in Fig. 2(a), where the response of the co-magnetometer to 0.05-ms pulses of different nature is shown.

![Figure 2](image_url)

In particular, the results show that the response significantly deviates from the Lorentzian shape of the magnetic pulse; the pulse is slightly longer and its shape is significantly distorted. At the same time the distortion is much smaller both in the case of electron and nuclear perturbations, which is a manifestation of absence of such compensation for pseudo-magnetic spin interaction. This is shown in Fig. 3, where pulse power and magnitude of the pulse integral is presented versus the pulse length.

Comparing the frequency response of the co-magnetometer [Fig. 2(a)] with the spectrum of the Lorentzian pulses [Fig. 2(b)],
one may expect a weaker response of the device to the magnetic pulse which spectrum is within the self-compensation band. To the contrary, no such behaviour is expected for nonmagnetic pulses, since the frequency response for nonmagnetic couplings have high amplitudes at low frequencies. These expectations are confirmed with the simulations; the longer the pulse, the more prominent is the divergence in energy between the response to magnetic and nonmagnetic perturbations [Fig. 3]. For 1-s long pulses, the difference is more than five orders of magnitude, and it grows for longer pulses. As for the pulse widths around 0.01 s, the response energy is comparable for all types of perturbations, since for the short pulses a significant fraction of the pulse spectrum is at higher frequencies, where response is comparable and large for both magnetic and nonmagnetic perturbations.

The cut-off of the low-frequency spectral magnetic components in the co-magnetometer provides another remarkable feature of the system; the integral over the co-magnetometer signal is significantly suppressed for magnetic field perturbations when integrated over time intervals significantly longer than the pulse width (for the presented results 200-s-long integration window have been used). In contrast, the integral is finite for nonmagnetic spin perturbations. For the simulated co-magnetometer system, assuming that the value of the effective pseudo-magnetic field is the same as the magnetic field, the difference between integrals over detected magnetic and nonmagnetic transients is about seven orders of magnitude for pulse widths between 0.01 s to 1 s [Fig. 3].

Conclusions

Numerical simulations of SERF and AM-NG co-magnetometers show a significantly stronger response of the latter to nuclear nonmagnetic spin perturbations. While the proton-coupling enhancement stems from the high sensitivity of the co-magnetometer to the nuclear spin perturbations, a larger contribution of the neutron polarisation to the NG polarisation provides an additional enhancement. At the same time, the response of both devices to the electron nonmagnetic spin perturbations is similar.

Our results demonstrate benefits of the co-magnetometer in searches for transient pseudo-magnetic spin couplings. On one hand, there is a suppressed response to low-frequency magnetic fields, which reduces noise of the device, on the other, due to “high-pass” magnetic-filter nature of the co-magnetometer, the device allows to differentiate between the magnetic and nonmagnetic transient responses, enabling a new way of identification of the observed signal nature. Specifically, the integral over time series signal for magnetic pulses has very small value, while it remains finite for nonmagnetic pulses. The features of the co-magnetometer presented at this work demonstrate the capabilities of the co-magnetometer in searches for transient nonmagnetic spin couplings, i.e., the signals that are being searched by the GNOME.

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Author contributions statement

M.P. defined a numerical models, conducted simulations, analysed the results and wrote the manuscript, M.K. participated into definition of a numerical models and edited manuscript, S.P. conceived and supervised the project, D.B., R.C., S.P., and A.W. analysed the results, discussed their interpretation and edited the manuscript. All authors reviewed the manuscript.

Additional information

Nonmagnetic interactions in SERF and AM-NG co-magnetometers

SERF magnetometer

The interaction-picture Hamiltonian, describing the evolution of AM electron spin $S$ and AM nuclear spin $I$ exposed to magnetic field $B$ and generic nonmagnetic spin perturbation $\Xi$, is given by

$$H = \mu_B g_{S} B \cdot S - \mu_B g_{I} B \cdot I + \chi_{e} \Xi \cdot S + \chi_{I} \Xi \cdot I,$$

(12)

where $\mu_B$ is the Bohr magneton, $g_S$ and $g_I$ are the electron and nuclear Landé factors, respectively, and $\chi_e$ and $\chi_I$ are the electron and nuclear constants associated with nonmagnetic coupling strength. Since $|g_I/g_S| \lesssim 10^{-2}$, we can simplify the Hamiltonian by neglecting the second term in Eq. (12).

In general, the anomalous field $\Xi$ couples to protons and neutrons, thus we can factorise the nuclear coupling constant $\chi_I$ into neutron and proton contributions

$$\chi_I = \sigma_n \chi_n + \sigma_p \chi_p,$$

(13)

where $\sigma_n$ and $\sigma_p$ define the neutron and proton fractional contributions to the total nuclear spin of the AM and $\chi_n$ and $\chi_p$ are the neutron and proton anomalous (dipole) coupling constants, respectively. For the comparison of the response of the spins to the magnetic and anomalous couplings, it is convenient to redefine exotic spin couplings in terms of effective pseudo-magnetic field, which, for all perturbations yield

$$b_e = \frac{\chi_e \Xi}{g_S \mu_B},$$

(14a)

$$b_{n}^{AM} = \frac{\sigma_n \chi_n \Xi}{g_S \mu_B},$$

(14b)

$$b_{p}^{AM} = \frac{\sigma_p \chi_p \Xi}{g_S \mu_B}.$$  

(14c)

Substituting the effective pseudo-magnetic field into Eq. (12) allows one to write the interaction Hamiltonian $H$ as

$$H = \mu_B g_{S} (B + b_e) \cdot S + \mu_B g_{S} (b_{p}^{AM} + b_{n}^{AM}) \cdot I.$$  

(15)
Using the Liouville equation
\[
\frac{d\rho}{dt} = \frac{1}{\hbar} [H, \rho],
\]  
where \([\cdot]\) denotes the commutator, to describe the evolution of the density matrix due to the Hamiltonian \(H\) [Eq. (15)] allows us to calculate temporal evolution of the AM density matrix in the considered case
\[
\frac{d\rho}{dt} = \frac{\mu B g S}{\hbar} \left\{ \left[ (B + b_e) \cdot S, \rho \right] + \left[ (b_0^A + b_p^A) \cdot I, \rho \right] \right\}.
\]  
(17)

At low fields and in the absence of hyperfine excitations, we can neglect hyperfine coherences in the AM atoms, which leads to a block-diagonal form of the AM density matrix
\[
\rho = \begin{pmatrix}
\rho_a & 0 \\
0 & \rho_b
\end{pmatrix},
\]  
(18)
where \(\rho_{a,b}\) corresponds to the \(F_{a,b} = \pm 1/2\) hyperfine state of the AM atoms\(^26\). The expectation values of AM electron spin \((S_{a,b})\) and AM total atomic spin \((F_{a,b})\) in the hyperfine states \(a\) and \(b\) are given by
\[
\langle S_{a,b} \rangle = \text{Tr}(S_{a,b} \rho_{a,b}),
\]
\[
\langle F_{a,b} \rangle = \text{Tr}(F_{a,b} \rho_{a,b}).
\]  
(19a, 19b)
Based on these definitions, evolution of the expectation value of the total atomic spin in the two hyperfine states is given by
\[
\frac{d\langle F_{a,b} \rangle}{dt} = \gamma_e (B + b_e) \times \langle S_{a,b} \rangle + \gamma_e (b_0^A + b_p^A) \times \langle I_{a,b} \rangle,
\]  
(20)
where \(\gamma_e = \mu B g S/\hbar\) denotes the electron gyromagnetic ratio. We can further use the projection theorem\(^28\)
\[
\langle S_{a,b} \rangle = \frac{\langle S_{a,b} \cdot F_{a,b} \rangle}{F(F + 1)} \langle F_{a,b} \rangle = \pm \frac{1}{2F + 1} \langle F_{a,b} \rangle,
\]  
(21)
and the relation between total angular momentum \(F_{a,b}\) and the nuclear spin \(I_{a,b}\) and electron spin \(S_{a,b}\)
\[
\langle I_{a,b} \rangle = \langle F_{a,b} \rangle - \langle S_{a,b} \rangle = \left( 1 \mp \frac{1}{2F + 1} \right) \langle F_{a,b} \rangle,
\]  
(22)
to rewrite Eq. (20) into the form
\[
\frac{d\langle F_{a,b} \rangle}{dt} = \pm \gamma_e \frac{1}{2F + 1} (B + b_e) \times \langle F_{a,b} \rangle + \gamma_e \left( \frac{1 + 1}{2F + 1} \right) (b_0^A + b_p^A) \times \langle F_{a,b} \rangle.
\]  
(23)
In the SERF regime, the expectation values of the projections of the total atomic angular momentum with the magnetic quantum number \(m\) are described with spin-temperature distribution\(^1\)
\[
\langle F_{a,b}, m | F_{a,b}, m \rangle = \frac{m \beta}{Z},
\]  
(24)
where \(\beta\) is the spin-temperature parameter, which can be expressed as a function of the AM polarisation \(P\)
\[
\beta = \ln \left( \frac{1 + P}{1 - P} \right),
\]  
(25)
and \(Z\) is the partition function
\[
Z = \sum_{m_a, m_b} e^{m \beta},
\]  
(26)
where indexes \(m_a\) and \(m_b\) denote the summation over all micro-states of the \(F_{a,b}\) hyperfine states.
In the considered case, the hyperfine components of the total atomic angular momentum \( \mathbf{F}_{a,b} \) can be expressed through ensemble-averaged total atomic spin \( \langle \mathbf{F} \rangle \)

\[
\langle \mathbf{F}_{a,b} \rangle = k_{a,b} \langle \mathbf{F} \rangle,
\]

where \( k_{a,b} \) are defined based on the spin-temperature distribution

\[
k_a = \frac{\sum_n m_n e^{\alpha n}}{\sum_m m_m e^{\beta m}},
\]

\[
k_b = \frac{\sum_n m_n e^{\alpha n}}{\sum_m m_m e^{\beta m}}.
\]

After substitution of Eq. (27) into Eq. (23) and adding the equations corresponding to the hyperfine levels, one obtains

\[
(k_{a} + k_{b}) \frac{d\langle \mathbf{F} \rangle}{dt} = \frac{(k_{a} - k_{b})\gamma e}{2I + 1} (\mathbf{B} + \mathbf{b}_e) \times \langle \mathbf{F} \rangle + \gamma e \left( (k_{a} + k_{b}) - \frac{k_{a} - k_{b}}{2I + 1} \right) (\mathbf{b}_n^{\text{AM}} + \mathbf{b}_p^{\text{AM}}) \times \langle \mathbf{F} \rangle,
\]

which leads to

\[
\frac{d\langle \mathbf{F} \rangle}{dt} = \gamma e \frac{Q}{2I + 1} (\mathbf{B} + \mathbf{b}_e) \times \langle \mathbf{F} \rangle + \gamma e \left( 1 - \frac{Q}{2I + 1} \right) (\mathbf{b}_n^{\text{AM}} + \mathbf{b}_p^{\text{AM}}) \times \langle \mathbf{F} \rangle,
\]

where \( Q \) denotes a dimensionless gyromagnetic ratio

\[
Q = \frac{k_{a} - k_{b}}{k_{a} + k_{b}}.
\]

Using Eq. (24), one can express the dimensionless gyromagnetic ratio \( Q \) of AM atoms with a given nuclear spin \( I \) using the polarisation

\[
Q(P, I = 3/2) = 2 - \frac{4}{3 + P^2}, \quad \text{(32a)}
\]

\[
Q(P, I = 5/2) = 3 - \frac{48(1 + P^2)}{19 + 26P^2 + 3P^4}, \quad \text{(32b)}
\]

\[
Q(P, I = 7/2) = \frac{4(1 + 7P^2 + 7P^4 + P^6)}{11 + 35P^2 + 17P^4 + P^6}. \quad \text{(32c)}
\]

In the description, it is convenient to introduce the slowing-down factor of the AM electron-spin expectation value, which takes into account both hyperfine interaction and averaging over hyperfine levels

\[
q = \frac{2I + 1}{Q}.
\]

Applying the projection theorem (21) in the Eq. (30) and taking into account Eq. (33) we can get the evolution of the ensemble-averaged electron spin expectation value \( \langle S \rangle \), which is directly measured in SERF magnetometers

\[
\frac{d\langle S \rangle}{dt} = \frac{1}{q} \left[ \gamma_e (\mathbf{B} + \mathbf{b}_e) \times \langle S \rangle + \gamma e \left( q - 1 \right) (\mathbf{b}_n^{\text{AM}} + \mathbf{b}_p^{\text{AM}}) \times \langle S \rangle \right]. \quad \text{(34)}
\]

Expressing the electron-spin expectation value \( \langle S \rangle \) with the electron polarisation \( \mathbf{P}^e \)

\[
\mathbf{P}^e = \frac{\langle S \rangle}{S},
\]

where \( S \) is an electron spin quantum number, and introducing the relaxation rate \( R^e \) and pumping rate \( R_p \) of the electron polarisation, we can write the equation fully describing evolution of the electron polarisation in the SERF magnetometer

\[
\frac{d\mathbf{P}^e}{dt} = \frac{1}{q} \left[ \gamma_e (\mathbf{B} + \mathbf{b}_e) \times \mathbf{P}^e + \gamma e \left( q - 1 \right) (\mathbf{b}_n^{\text{AM}} + \mathbf{b}_p^{\text{AM}}) \times \mathbf{P}^e + (s - P^e)R_p - R^e\mathbf{P}^e \right]. \quad \text{(36)}
\]

This equation is used in the main text for simulation of the SERF-magnetometer response to magnetic and nonmagnetic spin perturbations.
Co-magnetometer

To describe the coupled spin dynamics of AM and NG atoms subjected to the magnetic and nonmagnetic spin perturbations, we implement a similar approach to that described above. Since we consider the AM-NG co-magnetometer, we can use the result of the previous section to describe the AM electron spin evolution in the presence of magnetic and nonmagnetic fields and only consider the spin dynamics of NG atoms.

The evolution of the NG nuclear spins $\mathbf{K}$ in the presence of the magnetic field $\mathbf{B}$ and nonmagnetic perturbation $\Xi$ is governed by the Hamiltonian $H_{NG}$

$$H_{NG} = -\mu_{NG} \mathbf{B} \cdot \mathbf{K} - \lambda_{K} \Xi \cdot \mathbf{K},$$

where $\mu_{N}$ is the nuclear magneton, $g_{NG}$ is the NG $g$-factor, and $\lambda_{K}$ is the NG nuclear coupling to the anomalous field. In an approach similar to that derived for the AM nuclear anomalous coupling, we divide $\lambda_{K}$ into a part describing the neutron coupling constant $\lambda_{n}$ and proton coupling constant $\lambda_{p}$

$$H_{NG} = -\mu_{NG} \mathbf{B} \cdot \mathbf{K} - (\lambda_{n} \sigma_{n}^{K} + \lambda_{p} \sigma_{p}^{K}) \Xi \cdot \mathbf{K},$$

where $\sigma_{n}^{K}$ and $\sigma_{p}^{K}$ are the neutron and proton fractional contributions to the total NG nuclear spin $K$. It is also convenient to introduce an effective pseudo-magnetic field for the NG spin

$$b_{n}^{NG} = \frac{\lambda_{n} \sigma_{n}^{K}}{\mu_{NG}} \Xi, \quad b_{p}^{NG} = \frac{\lambda_{p} \sigma_{p}^{K}}{\mu_{NG}} \Xi.$$

In such a case, the effective pseudo-magnetic field, affecting the NG nuclear spin $b_{N}^{NG}$, has the following form

$$b_{N}^{NG} = b_{n}^{NG} + b_{p}^{NG}.$$

The Hamiltonian (38) leads to the following evolution of the NG nuclear spin expectation value

$$\frac{d \langle K \rangle}{dt} = \gamma_{e} (\mathbf{B} + b_{n}^{NG} + b_{p}^{NG}) \times \langle K \rangle,$$

where $\gamma_{e} = -\mu_{NG} / h$ denotes nuclear gyromagnetic ratio of the NG.

For the complete description of the AM-NG co-magnetometer, we need to take into account interaction between the AM and NG polarisations. Such interaction manifests as an effective magnetic field originating from by polarisation of one group atoms, which affects the polarisation the other group\(^{11}\)

$$\frac{d \mathbf{P}^{e}}{dt} = \frac{\gamma_{e}}{q} \mathbf{B}^{e} \times \mathbf{P}^{e} = \frac{\gamma_{e}}{q} \lambda M^{e} \mathbf{P}^{n} \times \mathbf{P}^{e},$$

$$\frac{d \mathbf{P}^{n}}{dt} = \gamma_{n} \mathbf{B}^{n} \times \mathbf{P}^{n} = \gamma_{n} \lambda M^{e} \mathbf{P}^{e} \times \mathbf{P}^{n},$$

where $\mathbf{B}^{e}$ and $\mathbf{B}^{n}$ stand for the fields stemming from the AM and NG polarisations, respectively, $\lambda$ is the coupling parameter\(^{11}\), $M^{e}$ and $M^{n}$ are the maximal magnetisations of the AM and NG, respectively, and $\mathbf{P}^{n}$ is the NG nuclear polarisation (here $K$ is NG nuclear spin quantum number)

$$\mathbf{P}^{n} = \frac{\langle K \rangle}{K}.$$

Rewriting Eq. (41) in terms of the polarisation and adding the interaction between the polarisation along with the relaxation, we get the complete description of the co-magnetometer in external fields by combining this result with Eq. (36)

$$\begin{cases}
\frac{d \mathbf{P}^{e}}{dt} = \frac{1}{q} \left[ \gamma_{e} (\mathbf{B} + b_{n}^{NG} + \lambda M^{e} \mathbf{P}^{n}) \times \mathbf{P}^{e} + (q - 1) \gamma_{e} b_{N}^{AM} \times \mathbf{P}^{e} + (s - \mathbf{P}^{e}) R_{p} - R_{e} \mathbf{P}^{e} \right], \\
\frac{d \mathbf{P}^{n}}{dt} = \gamma_{n} (\mathbf{B} + b_{n}^{NG} + \lambda M^{e} \mathbf{P}^{e}) \times \mathbf{P}^{n} + (\mathbf{P}^{n} - \mathbf{P}^{e}) R_{p}.
\end{cases}$$

This set of equations is used in our main manuscript to analyse the response of the co-magnetometer to magnetic and pseudo-magnetic spin perturbations.
Simulation parameters

For the numerical simulations presented in the main text, the parameters, given in Table 1, were used. The maximum effective fields associated with interaction between the AM and NG polarisations were calculated based on

\[
\lambda^e_M = \frac{8\pi \kappa}{3} n_{AM} \mu_{AM} S, \\
\lambda^n_M = \frac{8\pi \kappa}{3} N_{NG} \mu_{NG} K, 
\]

(45)

where \( \kappa \) is the enhancement factor\(^{11} \) (see Table 1), \( n_{AM} \) and \( n_{NG} \) are concentrations of the AM and NG atoms, \( \mu_{AM} \) and \( \mu_{NG} \) are the magnetic moments of the AM and NG atoms. We estimate the concentration of \(^{39}K\) at 190\(^\circ\)C using the phenomenological formula based on Ref.\(^29\)

\[
n_K = \frac{1}{k_B T} 10^{3.4077-4453/T} \text{cm}^{-3}.
\]

(46)

**Table 1.** Complete list of parameters used for the simulations.

| Parameter                                | Value   | Units          |
|------------------------------------------|---------|----------------|
| Electron gyromagnetic ratio \( \gamma_e \)  | 2\( \pi \cdot 2802.5 \) | s\(^{-1}\) mG\(^{-1}\) |
| AM pumping rate \( R_P \)                 | 600     | s\(^{-1}\)     |
| AM relaxation rate                       | 600     | s\(^{-1}\)     |
| AM steady state polarisation \( P^e_0 \)  | 0.5     |                |
| AM nuclear spin \( I \)                  | 3/2     | \( \hbar \)    |
| System temperature                       | 190     | \(^\circ\)C    |
| AM concentration                         | 10\(^{14}\) | cm\(^{-3}\)   |
| AM-NG enhancement factor \( \kappa \)     | 6       |                |
| NG concentration                         | 3.5     | 1 amg \( \approx 2.69 \cdot 10^{19} \) cm\(^{-3}\) |
| NG gyromagnetic ratio \( \gamma_n \)     | 2\( \pi \cdot 3.24 \) | s\(^{-1}\) mG\(^{-1}\) |
| NG relaxation rate \( R^n \)              | 5 \cdot 10\(^{-5}\) | s\(^{-1}\)     |
| NG steady state polarisation \( P^n_0 \)  | 0.05    |                |
| NG nuclear spin \( K \)                  | 1/2     | \( \hbar \)    |
| Maximum AM electron effective field \( \lambda^e_M \) | 0.067   | mG             |
| Maximum NG effective field \( \lambda^n_M \) | 25      | mG             |
| Compensation magnetic field \( B_c \)    | -1.31   | mG             |