Ultrasensitive atomic comagnetometer with enhanced nuclear spin coherence

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Achieving high energy resolution in spin systems is important for fundamental physics research and precision measurements, with alkali-noble-gas comagnetometers being among the best available sensors. We found a new relaxation mechanism in such devices, the gradient of the Fermi-contact-interaction field that dominates the relaxation of hyperpolarized nuclear spins. We report on precise control over spin distribution, demonstrating a tenfold increase of nuclear spin hyperpolarization and transverse coherence time with optimal hybrid optical pumping. We propose to use this comagnetometer to search for exotic spin-dependent interactions involving proton and neutron spins. The projected sensitivity surpasses the previous experimental and astrophysical limits by more than four orders of magnitude.

Coherent control of electron and nuclear spins via light-matter interactions is an important platform for fundamental physics research [1], and an essential tool for quantum sensors [2, 3] and quantum information processing [5]. Dense mixture of vapors of polarized alkali-metal atoms and noble gases with hyperpolarized nuclei have found prominent use in quantum-technology devices such as atomic magnetometers and comagnetometers. Particularly, atomic comagnetometers with hybrid spin ensembles are used to search for “new physics”, including fifth forces [6-7], axion-like particles [8-9], permanent electric dipole moments [10-11], and to test the combined charge-parity-time (CPT) and Lorentz symmetries [12-13].

These applications have long been limited by systematic errors and spurious signals due to magnetic field from ambient environment or interactions between atoms [14-15]. A typical approach for addressing this problem is to isolate the magnetic-field effect by using two species with different gyromagnetic ratio, for example, 129Xe and 131Xe [8-10], 3He and 129Xe [11], 85Rb and 87Rb [17], different nuclear spins in the same molecule [15] or different hyperfine levels of single-species atoms [18]. Another approach is operating the alkali-noble-gas atomic comagnetometer in the self-compensation (SC) regime [4, 19], where noble gas nuclear spins interact with alkali electron spins by spin-exchange (SE) interactions and adiabatically cancel slowly changing magnetic fields. Another advantage of SC comagnetometer is that the alkali atoms are in the spin-exchange relaxation free (SERF) regime, one can achieve sub-femtotesla magnetic sensitivity [20].

The combination of the SC and SERF regimes enables ultrasensitive measurements of non-magnetic fields and rotations. However, the SC mechanism for different alkali-noble-gas pairs which are used for numerous new-physics searches and inertial navigation [1] varies significantly and is not fully explored. The long-coherence-time 3He-alkali pair is used to achieve the highest sensitivity in “fifth force” measurements [7-21], whose SC regime breaks down at higher frequencies. In order to improve the sensitivity for non-magnetic field or to measure exotic interactions coupling to spin-3/2 nuclei, 21Ne atoms are used instead of the $I = 1/2$ 3He [13-15]. Due to the stronger Fermi contact interaction (FCI) between 21Ne atoms and alkali atoms where the FCI field experienced by 21Ne due to Rb is two orders of magnitude higher than that for the 3He-K pair [22-23], the SC regime is complicated by the strong FCI field from alkali atoms and the quadrupole relaxation as compared to the “simpler” 3He atoms. The SC regime of heaviest stable-noble-gas 129Xe, promising for electric dipole moment measurements [24] and quick-start gyroscope, is significantly influenced by the larger FCI factor $\kappa_0$ (two orders of magnitude larger than that for 3He) and shorter coherence time. Moreover, the optimal sensitivity in SERF regime is achieved when the collective spin polarization of alkali atoms is half of full polarization [20-23]. This special condition results in a significant polarization gradient of alkali atoms due to strong absorption of pump light [26-27], which further complicates the mechanism of SC suppression.

In this work, we demonstrate an ultrahigh non-magnetic field sensitivity of $3 \times 10^{-8}$ rad/s/Hz$^{1/2}$ in the low frequency range in a SC 21Ne-Rb-K comagnetometer, which is achieved by investigating new mechanism of SC regime. To overcome the newly found relaxation mechanism of
noble-gas nuclear spins that significantly shortens the coherence time and deteriorates the SC performance, the influences of hybrid alkali atoms (Rb-K) on spin-polarization homogeneity, hyperpolarization efficiency, and relaxation of noble-gas nuclear spins have been theoretically modelled and experimentally optimized, yielding a tenfold increase of coherence time of nuclear spins and the SC suppression ability of the hybrid comagnetometer. The energy sensitivity of this device for exotic field coupling to nuclear spins is on the order of $10^{-23}$ eV/Hz$^{1/2}$, which is six orders of magnitude better than state-of-art comagnetometers based on Rb atoms. This work will boost the experiments to search for exotic spin-dependent forces coupled to proton and neutron spins, whose sensitivity could be more than four orders of magnitude higher than previous experimental results and astrophysical limits.

**Hybrid Spin Ensembles Interactions** Hybrid SC comagnetometer consists of gaseous mixture of alkali-metal atoms and noble-gas atoms occupying the same glass cell as illustrated in Fig. 1(a). Using hybrid spin-exchange optical pumping (HSEOP), the lower-density alkali species is optically pumped and is used to polarize the higher-density alkali species via SE collisions. Due to the sufficiently large cross section between the hybrid alkali atom pair, hybrid alkali spins are strongly coupled and are in spin-temperature equilibrium with the same polarization. Simultaneously, electron-spin polarization of the alkali atoms is transferred to noble-gas nuclear spins through SE collisions between the alkali atoms and the noble-gas atoms, resulting in hyperpolarization of the noble-gas nuclear spins. Under a small external magnetic field, alkali atoms work in the SERF regime. The spin ensembles are pumped along $\hat{z}$. The transverse polarization of higher-density alkali spins induced by the excitation signals, coupled to noble-gas spins or alkali spins, is measured by detecting optical rotation of the linearly polarized probe light propagating along $\hat{x}$.

The SE interaction between alkali electron spins and noble-gas nuclear spins couples them together, which can be described by the FCI field seen by one spin species due to the magnetization of the other:

$$\vec{B}^{n/e} = \frac{2}{3} n^{e-n} M_0^{n/e} p^{n/e},$$

where the superscripts “e” and “n” denote electron and nuclear spins, respectively; $n^{e-n}$ is the FCI enhancement factor. $p^{n/e} = <S_n> / S_e$ is the collective polarization of alkali electron spins, $p^{n/e} = <K_n> / K_e$ for noble-gas nuclear spins; $M_0^{n}$ and $M_0^{e}$ are the magnetizations of alkali electrons and noble-gas nuclei for the case of full polarization.

**Self-compensation degradation mechanism.** For different alkali-noble-gas SC comagnetometer, the working regimes are significantly different. To realize an ultra-high sensitivity of the SC comagnetometer, it is essential to characterize the transverse magnetic field suppression in the SC regime. We define a suppression factor $SF_x (SF_y)$ as the ratio of the scale factors for the response to magnetic field $B_x (B_y)$ and the response to a pseudo-magnetic signal (e.g. inertial rotation $\Omega_x$). The explicit expressions can be found in the Supplemental Information of Ref. 31:

$$SF_x = \frac{R_0^n + \omega / 2 + \omega^2 \omega_0^n / (R_2^n \omega_0^n)}{\sqrt{(\omega_0^n)^2 + (\omega_0^n)^2 \omega^2 / R_2^n}},$$

$$SF_y = \frac{(R_0^n)^2 + \omega^2}{\omega_0^n \sqrt{(\omega_0^n)^2 + (\omega_0^n)^2 \omega^2 / R_2^n}}.$$  

where $\omega$ is the angular frequency of the external magnetic field. $R_0^n$ and $R_2^n$ are the transverse relaxation rates of noble-gas nuclear spins and alkali electron spins respectively. $\omega_0^n = \gamma_\omega B_z^n$ is the electron-spin Larmor precession frequency in the FCI field $B_z^n$, while $\omega_0^n = \gamma_\omega B_z^n$ is the nuclear-spin Larmor precession frequency in the FCI field $B_z^n$. The precession frequencies of coupled electron and noble-gas nuclear spins are combinations of $\omega_0^n$ and $\omega_0^n$.

From Eq. (2) and (3), we can find three sub-regimes considering the critical parameters $R_2^n$ and $\omega$:

$$SF_x \approx \begin{cases} \omega / \omega_0^n, & R_2^n \ll \omega_0^n, \\ R_0^n / \omega_0^n, & R_2^n \ll \omega_0^n, \omega < R_0^n, \\ (R_2^n + \omega) / \omega_0^n, & R_2^n \ll \omega_0^n, \omega > R_0^n \end{cases}$$

The suppression factor $SF_x$ can be derived similarly, which is approximately the square of $SF_x$. $SF_x$ is about one order of magnitude worse than $SF_y$. Improving the $SF_x$ is more important for the overall performance of the device as a detector of nonmagnetic signals (rotations...
or exotic fields), hence we focus on $SF_x$. Previous work [1, 27] described the SC regime in case (4), because the $R_2^o$ was considered to be small as for $K^3$He. Here, we find that for the Rb-21Ne system and also for Cs(Rb)-129Xe system, case (5) primarily relevant, in significant difference with the $K^3$He system.

The case (5) can be divided into two sub-cases. In case (5a), the suppression factors $SF_x$ is limited by the term $R_2^o/\omega_0^e$. This can be understood from the fact that magnetic noise $B_\perp$ is compensated by the transverse component of noble-gas nuclear magnetization $B_\perp^n$ whose amplitude is determined by $R_2^n/\omega_0^n$. Case (5a) is $\omega$-independent, which is contrary to the case (4) for $^3$He system. In case (5b), the $SF_x$ is limited by the term $(R_2^n + \omega)/\omega_0^n$, which can be interpreted as that the higher the frequency of magnetic noise $\omega$ is, the harder the transverse component of $P_z^n$ to follow and compensate the magnetic noise $B_\perp$, especially for $\omega$ higher than the intrinsic resonance frequency of noble-gas atoms $\omega_0^n$. The ways to improve the suppression ability for both two sub-cases are to increase $\omega_0^n$, i.e. the polarization of noble gas $F_z^n$, and reduce $R_2^n$. In the following, we define two parameters $SF_{x,low}$ and $SF_{x,mid}$, which are the values of $SF_x$ in cases (5a) and (5b) respectively.

The SC model is applied to different operation conditions in 21Ne-Rb-K comagnetometer. In the bottom coordinate of Fig.1(b), the nuclear spin polarization $P_z^n$ is improved by increase the pump light intensity. $SF_{x,low}$ and $SF_{x,mid}$ decrease with $P_z^n$, in agreement with the dependence of $SF_x$ on $\omega_0^n \propto P_z^n$. In the top coordinate of Fig.1(b), when increasing the cell temperature, the $P_z^n$ and the nuclear spin transverse relaxation rate $R_2^n$ all increase. The $R_2^n$ affects the $SF_x$ in the low-frequency range more significantly than the $P_z^n$, leading to that $SF_{x,low}$ deteriorates with $R_2^n$ regardless of the corresponding increment of $P_z^n$.

The relaxation rate $R_2^n$ is an important parameter for the low-frequency magnetic noise suppression. To explain the observed values of $R_2^n$, we estimated the partial rates from several known relaxation mechanisms [22-32], including spin-exchange and spin-destruction collisions and magnetic field gradients, and found that their sum about $110^{-3}$s$^{-1}$ is significantly smaller than the value measured. We find that the observed relaxation rate is, in fact, dominated by the Fermi-contact-interaction field gradient $\nabla B_z^n$ coming from the polarization gradient $\nabla P_z^n$ of alkali spins in the SC regime, see Eq. (1). The value of $\nabla B_z^n$ is calculated to be tens of nT/cm, much higher than the real magnetic field gradient of $\nabla B_z \approx 2$nT/cm. Adding this contribution to the gradient-related relaxation [33] brings the calculated value of $R_2^n$ to agreement with the measurement.

We attempted compensate $\nabla B_z^n$ by using coils to generate a uniform gradient $\nabla B_z$; however, application of such a gradient always increased the relaxation rate. We believe that this is because the effective field gradient has high-order nonuniform in HSEOP, which is caused by the combination of nonlinear absorption of pump photons, density gradients of the alkali atoms and diffusion.

Improvement of SC. In HSEOP, the polarization gradient $\nabla P_z^n$ is mainly determined by $\xi = n_{RB}/n_K$, the ratio of alkali number densities. Therefore, it is necessary to characterize the relationship between the polarization distribution of electron and nuclear spins and the density ratio. We take into account the diffusion of alkali and noble-gas atoms, the inhomogeneity and attenuation of pump light, cell geometry and wall relaxation to simulate the spin-polarization distribution using finite-element analysis [34-35].

In the simulation, $P_z^n$ at the cell center is normalized to 0.5 at 190°C, where the sensitivity of the SERF magnetometer is optimal. The pump light beam has a Gaussian profile with 18 mm beam diameter to cover the 12 mm diameter spherical vapor cell. The cell is filled with 2280 torr (3 amg) 21Ne and 50 torr N2. Other parameters of the 21Ne-Rb-K spin ensembles are the same as in Refs. [29, 32]. As shown in Fig.2(a), for a small $\xi$, $P_z^n$ decreases significantly along $\hat{z}$ (along the pump-light propagation direction), while for a larger $\xi$, $P_z^n$ becomes more uniform. In comparison, $P_z^o$ is always spatially homogeneous, due to the fact that the diffusion rate of noble gas is faster than its relaxation rate. We use $\eta^o = P_z^o/P_z^n_{Max}$, the ratio of the volume-averaged value to the maximum value to characterize the homogeneity of the polarization. Figure2(b) shows the dependence of $\eta^o$ and the volume-averaged nuclear spin polarization $P_z^n$ on the density ratio $\xi$. The polarization $P_z^n$ saturates at $\xi \approx 100$ while homogeneity $\eta^o$ continues to increases.
Five $^{21}$Ne-Rb-K cells with different $\xi$ were tested, with the values of $\xi = 6, 25, 83, 138$ and 163, respectively. Apart from $\xi$, other parameters were kept nearly the same, i.e. $^{21}$Ne density about 2.67 $\times$ 3.24 amag and $N_2$ pressure about 35 $\sim$ 53 torr. As shown in Fig. 2(c), the $P^n_x$ for each cell increases with $\xi$ but reaches a maximum at approximately $\xi = 83$ and then decreases, which is different from the simulation result in Fig. 2(b). The difference is due to the fact that $P^n_z$ in the simulations of $P^n_z$ is set to 0.5 for different $\xi$, while in the experiment, the available pump-light intensity is insufficient for the cell with larger $\xi$ to achieve high $P^n_z$, leading to a smaller $P^n_z$. With the increase of the pump-light intensity, $P^n_z$ can increase for larger $\xi$. However, this increase breaks down for large $\xi$, because the SE between K and Rb atoms is too slow to transfer the polarization of photon spin efficiently. Because $P^n_z$ is small for the cell with $\xi = 6$, we focus on the cells with $\xi$ ranging from 25 to 163. Figure 2(c) shows the relationship between $R^n_x$ and $\xi$ for a range of pump-light intensities. $R^n_x$ decreases with $\xi$ due to the reduction of FCI gradient with $\xi$, which agrees with the theoretical expectation in Fig. 2(b).

The suppression factors $SF_x$ for these four cells are shown in Fig. 2(d). $SF_x^{\text{low}}$ is mostly dominated by $R^n_x$, while the behaviour of $SF_x^{\text{Mid}}$ appears to be more complicated as it depends on factors including $R^n_x$, $\omega$, $P^n_x$ and the pump-light intensity. Although $\xi = 83$ achieves the highest $P^n_x$, its suppression ability is not the optimal, indicating that improving the noble-gas polarization alone is insufficient to optimize the suppression of magnetic noise. The reduction of relaxation rate is relatively more important. The optimal suppression ability occurs when $\xi = 163$, which is one order of magnitude higher than that of $\xi = 25$. The low-frequency suppression factor for the $\xi = 163$ cell is smaller than 0.01, which means the magnetic noise in the magnetic shield (on the order of $10^{-15}$) can be suppressed by two orders of magnitude.

The amplitude spectral density of comagnetometer signal is shown in Fig. 3. The calibration of the SC comagnetometer sensitivity is done by measuring the rotation of the Earth [31]. The peak around 5.0 Hz is related to the vibration resonance of the vibration-isolation platform, which is confirmed with a seismometer installed on the platform. Below 0.2 Hz, the comagnetometer noise is dominated by the 1/f noise caused by various drifts such as those of temperature and pump-light alignment. The measured noise spectrum reflects the ability of the device to suppress magnetic noise. The noise from the inner ferrite shield is calculated based on finite element analysis using the measured complex permeability $\mu'/\mu_0 = 6308(14)$, $\mu''/\mu_0 = 45(3)$ and geometric parameters [56]. The magnetic noise is calculated to be about $2.5f^{-1/2}$ rad/s/Hz (f in unit of Hz) and converted to the corresponding noise with unit of rad/s/Hz$^{1/2}$ by multiplying the gyromagnetic ratio of $^{21}$Ne [4]. Except for the frequency range dominated by the 1/f noise, the noise from the ferrite shield exceeds the comagnetometer noise in the low-frequency range, indicating that the magnetic noise is effectively suppressed by the SC effect. The probe-light noise is measured to be smaller than the comagnetometer noise. Above 1.0 Hz, the polarimetry noise of probe light based on optical rotation is lower than $2 \times 10^{-8}$ rad/Hz$^{1/2}$, approaching the limit of photon shot noise [37].

The averaged noise is $3 \times 10^{-8}$ rad/s/Hz$^{1/2}$ in the frequency range from 0.2 to 1.0 Hz, corresponding to an effective pseudomagnetic field sensitivity of $\delta B = 1.5 \text{fT}/\text{Hz}^{1/2}$. For the exotic-field Zeeman-like pseudomagnetic coupling to $^{21}$Ne nuclear spin, the energy is $E = \mu Ne \cdot \mathbf{b}^n$, yielding an energy resolution of $\delta E_{Ne} = 3.1 \times 10^{-23}$ eV/Hz$^{1/2}$.

The energy sensitivity of the exotic field coupling to neutron and proton spins are determined by $\delta E_{n/p} = \delta E_{Ne}/\eta_{n/p}$, where $\eta_n = 0.58$ and $\eta_p = 0.04$ are the neutron and proton fraction of spin polarization in $^{21}$Ne atoms [55] respectively. Therefore, the energy sensitivity of our setup are $\delta E_{n} = 5.4 \times 10^{-23}$ eV/Hz$^{1/2}$ and $\delta E_{p} = 7.8 \times 10^{-22}$ eV/Hz$^{1/2}$ respectively.

Based on the demonstrated performance of the device, we propose an experiment to search for exotic interaction between the comagnetometer spins and the Earth gravitational field, using the geometry similar to that of Ref. [17, 39]. The spin-dependent force could be mediated, for example, by an ultralight spin-0 boson, such as axion or axionlike particle (ALP) [40, 42]:

$$V = \frac{g_{s} g_{p} h^2}{8 \pi m} (\hat{\sigma} \cdot \hat{r}) \left( \frac{1}{r \lambda} + \frac{1}{r^2} \right) e^{-r/\lambda},$$

where $g_s$ and $g_p$ are scalar and pseudoscalar coupling constants; $h$ is the reduced Planck constant; $\hat{\sigma}$ is the Pauli spin-matrix vector of one fermion and $m$ is its mass; $\lambda$ is the force range, which is inversely proportional to the mass of the force-mediating boson; $r$ is the relative distance about 35 21 torr. As shown in Fig. 2(c), the noise spectrum is dominated by various 1/f noises. The peak in the noise spectrum is due to the vibration which is confirmed with precision seismometer. The probe noise, which is measured with unpolarized spin ensembles by blocking the pump light, is smaller than the comagnetometer noise and approaches the photon shot noise limit.

![FIG. 3. The noise spectrum of the SC comagnetometer. In the frequency range from 0.2 to 1.0 Hz, the averaged noise is $3 \times 10^{-8}$ rad/s/Hz$^{1/2}$. Below 0.2 Hz, the noise spectrum is dominated by various 1/f noises. The peak in the noise spectrum is due to the vibration which is confirmed with precision seismometer. The probe noise, which is measured with unpolarized spin ensembles by blocking the pump light, is smaller than the comagnetometer noise and approaches the photon shot noise limit.](Image 326x604 to 553x740)
distance between two fermions and \( \mathbf{r} \) is the unit vector directed from the one fermion to the other. If it exists, this exotic force violates parity (P) and time-reversal invariance (T).

The state-of-the-art experiments for the proton spin-gravity coupling include the \(^{85}\text{Rb}\) and \(^{87}\text{Rb}\) comagnetometer [17] or the \(^{87}\text{Rb}\) comagnetometer using two hyperfine levels of \(^{87}\text{Rb}\) atoms [18]. Both these experiment realized energy resolutions on the order of \(10^{-18}\) eV for an integration time of more than one hundred hours. The best experimental result for the neutron-spin coupling to Earth gravity was obtained with a \(^{199}\text{Hg}^{201}\text{Hg}\) comagnetometer and realized an energy resolution on the order of \(10^{-21}\) eV [39]. We estimate the sensitivity of our experiment using Earth as a source and integrating for about 100 hours using a similar approach to that work of Refs. [13, 17, 39]. The estimated exotic magnetic field sensitivity is \(\delta B\lesssim 0.01\) fT, and energy sensitivity as \(\delta E_n \lesssim 4 \times 10^{-25}\) eV and \(\delta E_p \lesssim 5 \times 10^{-23}\) eV respectively. The estimated sensitivity to the coupling constants and a comparison with the previous work is shown in Fig. 4. The sensitivity of this proposal can surpass the direct experimental limits and the astrophysical limits on the exotic interactions coupling to proton and neutron spins by more than four orders of magnitude.

Taking advantage of the ultrahigh sensitivity, this comagnetometer can also be used to explore new spin-dependent physics, including directly searching for axion and axion-like particles (ALPs) [9], and local Lorentz invariance (related to the CPT symmetry) [12, 13].

**Conclusion** We establish an analytical model to describe the SC effect and find that the relaxation and polarization of noble-gas nuclear spins are the key factors determining the SC performance. The relaxation of noble-gas nuclear spins is found to be dominated by a new relaxation mechanism, i.e., the Fermi-contact-interaction field gradient resulting from nonuniform alkali spin polarization. The degree of spin polarization and its relaxation time for the noble-gas atoms are both improved by one order of magnitude over the earlier work [14] by optimizing the density ratio between the two alkali species. An average sensitivity of \(3 \times 10^{-5}\) rad/s/Hz\(^{1/2}\) in the frequency range from 0.2 Hz to 2.0 Hz has been achieved. This sensitivity represents six orders of magnitude better energy resolution compared to comagnetometers using Rb isotopes that were used to search for exotic gravity coupling to proton spins [17]. The improvement of nuclear spin coherence time and polarization is also beneficial for NMR gyroscopes, noble-gas-spins-based quantum memory, coherent bidirectional coupling between light and noble-gas spins [5], as well as for neutron spin filters [55].

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