

^3He absolute magnetometer at geomagnetic fieldYuefeng Lu , Ekrem Abdukerim , Baichuan Li, Liwei Lei , Teng Wu *, and Hong Guo [†]*State Key Laboratory of Photonics and Communications, School of Electronics, and Center for Quantum Information Technology, Peking University, Beijing 100871, China*

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The high accuracy of the nuclear spin g factor provides a feasible way to construct absolute magnetometry. However, at geomagnetic field, the sensitivity and accuracy of the nuclear spin magnetometer are limited by the ratio of nuclear-spin polarization and the detection efficiency of low-frequency signal. We propose an absolute magnetometer scheme based on the nuclear spin of ^3He , demonstrating an order-of-magnitude improved sensitivity and onefold improved accuracy compared to a proton magnetometer for the current geomagnetic field standard. We present systematic estimations on the sources of errors within the ^3He magnetometer. Through adopting multipass-probe scheme and radio-frequency power stabilization, it is promising to further improve the accuracy of the ^3He magnetometer to 100 ppb at geomagnetic field, paving the way to the next generation of geomagnetic field standard.

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Absolute magnetometer provides traceable determination of magnetic field strength to the international standard [1–3], which enables unified estimations of parameters in magnetic field-related applications, making it extremely important in metrology [4–8] and measurements of fundamental constants [9–16]. With the reform of the International System of Units from 2019, the method of magnetic field standard is on the path of transitioning from coil-based to gyromagnetic-ratio-based techniques, linking magnetic field to the gyromagnetic ratio (γ) of valence electron spin (e.g., K, Rb [17], Cs [18], ^4He [5,19,20]) or nuclear spin (e.g., ^3He [9,12,21], proton in water) and the frequency [22,23].

Achieving high-accuracy gyromagnetic-ratio-based absolute magnetometry demands not only stringent control of systematic errors inherent to atomic magnetometers, but also accurate knowledge of the atomic gyromagnetic ratio itself, both theoretically and experimentally. Electron-spin-based magnetometers, leveraging efficient optical pumping and detection, have reached sensitivities on the order of tens of $\text{fT}/\text{Hz}^{1/2}$ at geomagnetic fields [24,25]. Although the gyromagnetic ratio of free electron is a well-established physical constant [26], the gyromagnetic ratio of the valence electron, e.g., in alkali-metal and ^4He atoms, remains insufficiently determined experimentally due to the scarcity of high-accuracy measurements [19,27–31]. Realizing

electron-spin-based magnetic field standards entails careful mitigation of multiple systematic effects, such as nonlinear Zeeman shifts [24,32], light shifts [5,17–20], and spin-exchange frequency shifts [23], increasing the complexity of systematic error analysis and correction.

Nuclear gyromagnetic ratios (^3He and water protons) are fundamental constants listed in CODATA [26] and distinguished by minimal systematic errors, with values confirmed through long-term, cross-institutional measurements achieving sub-ppb uncertainties. To date, absolute magnetic field measurements based on nuclear spin have demonstrated accuracy at tens of ppb level in both low ($\sim 1\ \mu\text{T}$) and high ($\sim 1.45\ \text{T}$) magnetic fields, through utilizing optical pumping magnetometers (OPMs, at low field) [9,21] or pickup coils (at high field) for measuring the nuclear spin-precession signals [12]. However, the precession frequency of the nuclear spin at geomagnetic field (from 10–100 μT) is typically on the order of kHz, which is not efficient for both OPM-based and coil-based detection, leading to limited sensitivity to tens of $\text{pT}/\text{Hz}^{1/2}$ [33] and accuracy to hundreds of pT. Further improvements are urgent for applications demanding high-accuracy geomagnetic field measurements.

Spin-exchange interactions in ^3He not only enable efficient nuclear spin polarization but also provide a mechanism for transducing the nuclear spin precession signal into optically detectable channels [34]. Two established techniques, i.e., metastability-exchange optical pumping (MEOP) [35–41] and spin-exchange optical pumping [41–45], are widely employed for polarizing and probing ^3He nuclear spins. Among them, MEOP offers superior polarization efficiency and faster buildup times at low pressures, and is based solely on ^3He , and in turn has fewer sources of systematic errors in affecting the nuclear spin evolution under magnetic field. However, both approaches inevitably introduce spin-exchange frequency shift-induced systematic error. A quantitative method of such a shift is thus

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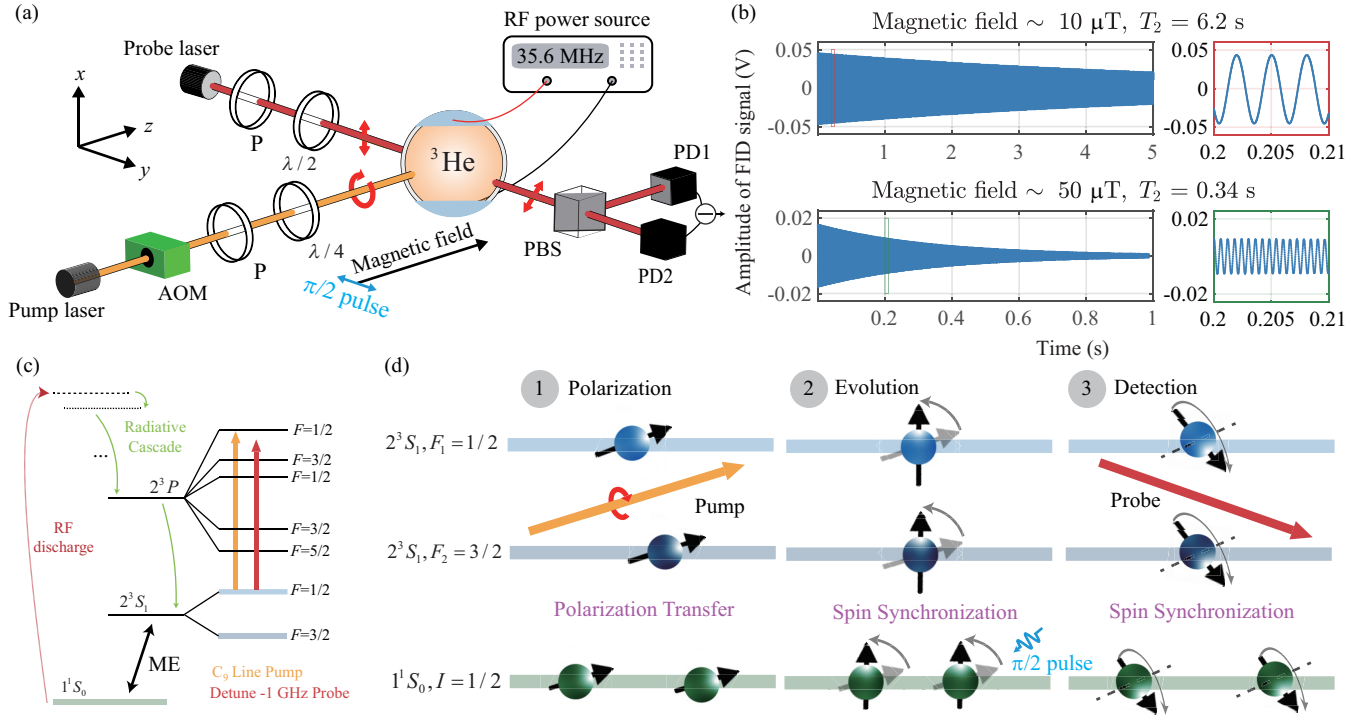


FIG. 1. Structure and principle of the ^3He magnetometer. (a) The structure of ^3He magnetometer. AOM, acousto-optic modulator. P, polarizer. $\lambda/4$, quarter-wave plate. $\lambda/2$, half-wave plate. PBS, polarizing beam splitter. PD1 and PD2 are balanced differential photodiodes for measuring the polarization rotation of probe laser. RF discharge power source is used to excite and generate metastable atoms in ^3He cell. The optical components and ^3He cell are integrated as a compact structure. (b) The measurement results of the ^3He FID signal at ~ 10 and $\sim 50 \mu\text{T}$. T_2 is transverse relaxation time of ^3He FID signal. (c) ^3He energy levels and corresponding optical frequencies relevant to the proposed ^3He magnetometer. ME is metastability exchange. (d) The spin dynamics of ^3He atoms in the laser-detected ^3He FID magnetometer.

desirable to be developed for realizing ^3He -based absolute magnetometry.

In this paper, a laser-detected ^3He free induction decay (FID) magnetometer based on MEOP is presented. The experimental results reveal clearly that the dominant systematic error is metastability-exchange (ME)-induced frequency shift. We thus develop a theoretical approach, including a detailed list of experimental procedures, to quantitatively evaluate the ME-induced frequency shift. We also carefully investigate the other sources of systematic errors within the ^3He magnetometer. The performance of the proposed ^3He magnetometer is validated by comparing with the proton magnetometer from the National Institute of Metrology, China.

II. EXPERIMENTAL SYSTEM CONSTRUCTION AND SYSTEMATIC ERROR CORRECTION

A schematic of the ^3He FID magnetometer is shown in Fig. 1(a), and the laser-detected FID signals at two values of geomagnetic field, e.g., ~ 10 and $\sim 50 \mu\text{T}$, are shown in Fig. 1(b). The relevant energy-level structure of ^3He is shown in Fig. 1(c). A circularly polarized pump laser (Toptica DL pro, 1083.327 nm, 300 mW, 2 mm diameter), resonant with the C_9 transition line of ^3He [46], propagates along the magnetic field axis to induce nuclear spin polarization in a 10 torr, 3-cm-diameter spherical cell via MEOP. After polarization, the pump laser is switched off, and a $\pi/2$ magnetic field pulse [47] is used to manipulate the nuclear spins, inducing

transverse polarization in metastable-state spins through ME. The rapid ME process locks the precession of the metastable-state spins with the ground-state spins. A linearly polarized probe beam (NKT Y10, 1083.331 nm, 1 mW, 2 mm diameter), red detuned 1.2 GHz from the C_9 line and orthogonal to the pump beam, interacts with the ^3He metastable states to measure the nuclear spin precession signal. The spin dynamics of the metastable states, including the polarization, evolution, and detection of the ground state (nuclear spin), are illustrated in Fig. 1(d).

The Cramér-Rao lower bound [48,49] yields a magnetometer sensitivity of $4.8 \text{ pT/Hz}^{1/2}$ at $\sim 50 \mu\text{T}$ and $0.18 \text{ pT/Hz}^{1/2}$ at $\sim 10 \mu\text{T}$, surpassing the previous benchmark of $21 \text{ pT/Hz}^{1/2}$ [33]. The sensitivity dependence on magnetic field is governed by ME-induced relaxation, which increases with field strength owing to the gyromagnetic ratio mismatch between the metastable and ground states. The spin-projection-noise limit [23,50] is $86 \text{ fT/Hz}^{1/2}$, indicating 2 orders of magnitude potential improvement toward the fundamental quantum limit. An order-of-magnitude sensitivity improvement is promised with a multipass optical detection [51], which is well suited to ^3He because its ground-state relaxation is nearly independent of probe power, enabling stronger light-atom interaction without loss of coherence.

To enable absolute magnetic field measurements, the systematic errors in the MEOP-based ^3He magnetometer are analyzed. The sources of the systematic errors arise from laser, radio frequency (RF) discharge, residual magnetism of

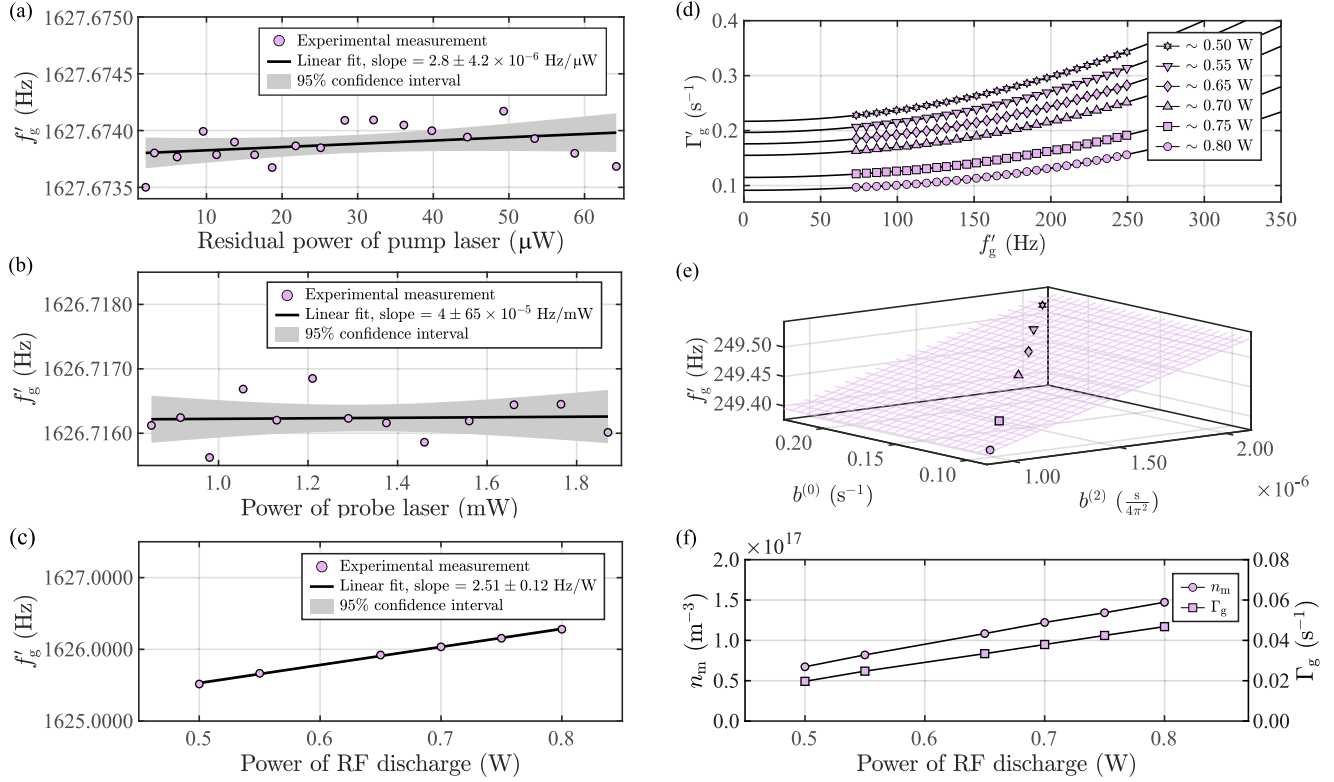


FIG. 2. Systematic errors analysis and correction. The relations between ³He FID frequency and residual power of pump laser (a), the power of probe laser (b), and the RF power (c). (d) FID relaxation rates vs frequencies at varying RF powers. (e) FID frequencies vs ME-induced relaxation coefficients $b^{(0)}$, $b^{(2)}$ at varying RF powers. (f) Measured metastable-state densities n_m and ground-state relaxation rates Γ_g at varying RF powers. All markers are larger than the associated error bars.

the sensor, magnetic field gradient, and the clock frequency deviation. Beneficially, the ³He ground state is not optically coupled, rendering negligible light shift [see Figs. 2(a) and 2(b)] [47]. The dominant contribution of the systematic error arises from RF power, as shown in Fig. 2(c), since RF power affects the metastable-state density and relaxation rates, which modify ground-state precession via ME.

We thus propose a theoretical model describing ³He spin dynamics, along with detailed experimental procedures for correcting the ME-induced frequency shift. The expectation values of the transverse components of the nuclear spin $\langle I \rangle$ and the metastable-state spins $\langle F_1 \rangle$ and $\langle F_2 \rangle$ [see Fig. 1(c)] evolve according to

$$\frac{\partial}{\partial t} \langle I \rangle = k_{ME} n_m \left(-\langle I \rangle - \frac{1}{3} \langle F_1 \rangle + \frac{1}{3} \langle F_2 \rangle \right) + 2\pi i f_g \langle I \rangle - \Gamma_g \langle I \rangle, \quad (1)$$

$$\frac{\partial}{\partial t} \langle F_1 \rangle = k_{ME} n_g \left(-\frac{7}{9} \langle F_1 \rangle + \frac{1}{9} \langle F_2 \rangle - \frac{1}{9} \langle I \rangle \right) + 2\pi i \frac{\gamma_{F_1}}{\gamma_g} f_g \langle F_1 \rangle - \frac{n_g}{n_m} \Gamma_g \langle F_1 \rangle, \quad (2)$$

$$\frac{\partial}{\partial t} \langle F_2 \rangle = k_{ME} n_g \left(-\frac{4}{9} \langle F_2 \rangle + \frac{10}{9} \langle F_1 \rangle + \frac{10}{9} \langle I \rangle \right) + 2\pi i \frac{\gamma_{F_2}}{\gamma_g} f_g \langle F_2 \rangle - \frac{n_g}{n_m} \Gamma_g \langle F_2 \rangle. \quad (3)$$

The first term on the right-hand side of the equations represents the ME effect [35–41], while the second term accounts for the effect of magnetic field, and the third term describes relaxation. n_g and n_m are the atomic densities of ground state and metastable states, and must be determined experimentally. The gyromagnetic ratios of ³He ground state and metastable state are γ_g , γ_{F_1} , and γ_{F_2} , respectively, and are constants [19,46,47,52]. The ME coefficient ($k_{ME} = 154 \times (T/300)^{1.09} \times 10^{-18}$ m³/s [41]) defines the ME rates $k_{ME} n_m$ for nuclear spin and $k_{ME} n_g$ for metastable-state spin. $f_g = \gamma_g B$ is the Larmor frequency of the nuclear spin without the influence of ME. Γ_g and $n_g \Gamma_g / n_m$ are the relaxation rates of ground state and metastable states, respectively [47]. The metastable state exchanges its precession phase with ground state at ME rate $k_{ME} n_g$, which is $\sim 10^7$ Hz in a 10 torr ³He cell, one order of magnitude higher than the metastable-state Larmor frequency ($\sim 10^6$ Hz) at geomagnetic field.

The fast ME dominates the spin dynamics of the metastable state, synchronizing its precession frequency and relaxation rate with those of the ground state under geomagnetic conditions. As a result, the solution of Eqs. (1)–(3) takes the form $\langle I \rangle = k_1 \langle F_1 \rangle = k_2 \langle F_2 \rangle = A \exp(-\Gamma'_g t) \sin(2\pi f'_g t + \phi)$. f'_g and Γ'_g are the frequency and relaxation rate of ³He FID signal under the influence of the ME effect, which are distinguished from the intrinsic Larmor frequency f_g and the intrinsic relaxation rate Γ_g . k_1 and k_2 are the amplitude ratios of $\langle F_1 \rangle$ and $\langle F_2 \rangle$ compared to $\langle I \rangle$, respectively. A is the amplitude and

TABLE I. Systematic errors correction of ^3He magnetometer at $\sim 50 \mu\text{T}$. All uncertainties reported in this work represent one standard deviation (1σ) of the measured values.

Source	Correction	Uncertainty
RF discharge	-19.384 nT	25 pT
Residual magnetism	$+2.607 \text{ nT}$	46 pT
Clock deviation	-0.34463 nT	0.27 pT
Total correction	-17.122 nT	52 pT (1 ppm)

ϕ is the phase. From the above theory, expressions for the ME-induced relaxation and frequency shift are obtained

$$\Gamma'_g = b^{(0)} + b^{(2)} f_g^2 + O(f_g^4), \quad (4)$$

$$f'_g = f_g + \Delta f_g(f_g, n_g, b^{(0)}, b^{(2)}), \quad (5)$$

where $b^{(0)}$ and $b^{(2)}$ are functions of n_g , n_m , and Γ_g , and are independent of the external magnetic field. These effects arise from the mismatch between the gyromagnetic ratios of the metastable and ground states, which reduce the total spin magnitude and alter the precession frequency of the ground state during ME. Equation (4) describes the ME-induced relaxation, showing that the ^3He FID relaxation rate increases quadratically with frequency. Equation (5) describes the corresponding frequency shift. Given the measurable parameters $b^{(0)}$, $b^{(2)}$, and n_g , a quantitative relation between f'_g and f_g can be established, enabling correction of the ME-induced shift.

Specifically, the $\Gamma'_g - f'_g$ dependence is measured at different RF powers under external fields below 8000 nT [see Fig. 2(d)], and is fitted with Eq. (4), yielding $\{b^{(0)}, b^{(2)}\}$ for different RF powers. Then, f'_g is plotted versus $\{b^{(0)}, b^{(2)}\}$ at different RF powers [see Fig. 2(e)], from which the ground-state atomic density n_g is fitted via Eq. (5). This value is $3.5956(28) \times 10^{23} \text{ m}^{-3}$ and is consistent with the expected value for a 10 torr cell [41].

Finally, by substituting the known $\{b^{(0)}, b^{(2)}\}$ and the fitted n_g into Eq. (5), the relation between the measured FID frequency f'_g and the intrinsic ground-state Larmor frequency f_g at a typical RF power is obtained. For instance, at the operating RF power of $\sim 0.55 \text{ W}$, neglecting terms smaller than 10^{-5} Hz ($\sim 0.3 \text{ pT} \times \gamma_g$), we obtain

$$f'_g = 1.00051507(43)f_g - 6.875(58) \times 10^{-11} f_g^3 + 1.076(25) \times 10^{-17} f_g^5 - \dots + O(10^{-5}). \quad (6)$$

For measurements at $\sim 50 \mu\text{T}$, the correction of the ME-induced frequency shift is presented in Table I. The associated uncertainty arises from RF power fluctuations, which induce variations in n_m and relaxation rate Γ_g during calibration. In the present setup, RF power noise with an RMS of $\sim 0.1 \text{ mW}$ contributes an uncertainty of 25 pT to the ME-induced frequency shift correction. Improving RF noise to a level of $20 \mu\text{W}$ would reduce this to 5 pT (0.1 ppm). In addition, if discrete RF power jumps or drifts are observed by the power meter during magnetometer operation (e.g., due to RF circuit disturbances), the corresponding metastable-state atomic density should be recalibrated using the same procedure.

Moreover, since n_m and Γ_g are functions of n_g , $b^{(0)}$, $b^{(2)}$, the obtained $\{b^{(0)}, b^{(2)}\}$ and n_g allow direct calculation of n_m

and Γ_g . Figure 2(f) shows that both n_m and Γ_g increase with RF power, consistent with earlier results [37]. Detailed derivations, including the experimental procedures, are summarized in the Supplemental Material [47].

Additionally, residual magnetism is corrected using the vector relation among the measured magnetic field, the applied field, and the residual magnetism, as detailed in Appendix A. Clock deviation is corrected by comparing the data acquisition clock with a frequency standard, also described in Appendix A. The correction results are summarized in Table I. Considering the finite cell size and uniform atomic diffusion, the magnetometer effectively measures the magnetic field at the cell center [53]. In our experiment, the first-order field gradient produces a shift relative to the center value of less than 22 aT , which is negligible, as detailed in Appendix B. The method used to analyze the measurement uncertainty is given in Appendix C.

It should be noted that, since the calibrated quantities, i.e., atomic densities, residual magnetization, and sampling frequency, are independent of the external magnetic field, the correction procedures above are directly applicable across a broader range of magnetic field.

The performance of the ^3He magnetometer is evaluated through a direct comparison with the proton magnetometer from the National Institute of Metrology, China. For geomagnetic field, the uncorrected readout of the ^3He magnetometer is $50\,117.414(5) \text{ nT}$, and is corrected as $50\,100.292(52) \text{ nT}$, in close agreement with the proton magnetometer measurement of $50\,100.840(100) \text{ nT}$. Residual discrepancies may arise from external magnetic field fluctuations and alignment errors between the sensing regions of the two magnetometers.

III. CONCLUSION

In conclusion, an absolute geomagnetic field measurement scheme using an optically detected ^3He FID magnetometer is proposed. Optical detection improves the estimated sensitivity of nuclear spin magnetometers at geomagnetic fields, reaching $4.8 \text{ pT/Hz}^{1/2}$ at $\sim 50 \mu\text{T}$ and $0.18 \text{ pT/Hz}^{1/2}$ at $\sim 10 \mu\text{T}$. ME-induced frequency shift caused by RF discharge is measured and corrected. After systematic error correction, a nuclear spin magnetometer achieving an absolute geomagnetic field measurement uncertainty of 52 pT is demonstrated, with measurement results consistent with the standard proton magnetometer. By implementing multipass configuration, nonmagnetic probe construction, and stabilizing RF power, a measurement accuracy at 100 ppb level at geomagnetic field is promising. The ME-induced frequency shift correction method developed here enables *in situ* atomic density measurements and provides a general framework for quantitative studies of spin-exchange-coupled atomic systems. Most importantly, this work establishes a promising and solid approach for next-generation geomagnetic field standards, covering from zero to geomagnetic field, with wide-reaching implications, including accurate experimental determinations of alkali-metal gyromagnetic ratios.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX A: CORRECTION OF RESIDUAL MAGNETISM AND CLOCK DEVIATION

We notice that the laser collimator exhibits a residual magnetism. Therefore, the measurement result (B_{read}) reflects the magnitude of the vector sum of the residual magnetism of the sensor (B_r) and the external field (B_0). Rotating the sensor alters the angle θ between B_r and B_0 . This relation is described by $B_{\text{read}}^2 = |B_0|^2 + |B_r|^2 - 2|B_0||B_r|\cos(\pi - \theta)$. Thus, the FID signal frequencies at different sensor orientation angles are measured to determine the magnitude and direction of the residual magnetism of the sensor. To address this, we are developing nonmagnetic fiber collimators for integration into the next-generation ³He sensor, which will directly eliminate this source of systematic error. In the absence of residual magnetization, the magnetometer proposed would achieve an uncertainty of 25 pT (0.5 ppm). The sampling frequency of the data acquisition (DAQ) system is referenced to a frequency standard. In this work, the DAQ sampling rate is calibrated against a known standard to ensure traceability. The resulting corrections are listed in Table I, with a detailed analysis of the associated systematic errors provided in the Supplemental Material [47].

APPENDIX B: MAGNETIC FIELD GRADIENT-INDUCED SHIFTS AND RELAXATION

For a single magnetometer, the measurement reflects the magnetic field at the geometric center of its sensitive element (i.e., the cell) [53]. However, magnetic field gradients can cause a frequency shift relative to the cell center and induce gradient-induced relaxation. Considering the finite cell size and uniform atomic diffusion mode [53], the frequency shift

and relaxation induced by a first-order magnetic field gradient in a spherical cell are

$$\delta f_g = \frac{\gamma_g R^2}{10B_z} \sum_{i=x,y} |\nabla B_i|^2, \quad \delta \Gamma_g = \frac{8\gamma_g^2 R^4}{175D} |\nabla B_z|^2, \quad (\text{B1})$$

where $R \approx 0.015$ m is the radius of the spherical ³He cell, $D = 0.0137$ m²/s is the diffusion coefficient of ³He at 10 torr [54], and $|\nabla B_z| = (\sum_{i=x,y,z} (\partial_i B_z)^2)^{1/2}$ is magnitude of the gradient of B_z . These expressions hold under the following conditions: The mean free path of atoms is much smaller than the cell radius R ; the relaxation time is sufficiently long such that $(DT_2)^{1/2} \gg R$; and the magnetic field gradient is small compared to the main field, i.e., $R|\nabla B| \ll B_0$ [54]. Experimentally, the field gradients are measured below 5 nT/m transversely ($|\nabla B_{x,y}|$) and 50 nT/m longitudinally ($|\nabla B_z|$), under an external field B_0 of ~ 50 μ T along the z axis. Substituting these values into Eq. (B1), the gradient-induced frequency shift and relaxation are estimated as

$$\delta f_g < 7.3 \times 10^{-10} \text{ Hz}, \quad \delta \Gamma_g < 1.8 \times 10^{-5} / \text{s}. \quad (\text{B2})$$

The gradient-induced frequency shift is 7 orders of magnitude smaller than the current magnetometer absolute uncertainty and the gradient-induced relaxation is at least 4 orders of magnitude lower than the FID relaxation rate. Therefore, under the present field conditions, both effects are negligible, and the magnetometer measurement reflects the field at the cell's geometric center.

Additionally, magnetic field gradients also introduce systematic errors in intercomparison measurements between magnetometers, necessitating careful consideration of the geometric center of each sensor's sensitive element in subsequent analyses to correct this error.

APPENDIX C: UNCERTAINTY ANALYSIS

The uncertainties of the fitted parameters are evaluated using a parametric Monte Carlo method based on bootstrap resampling [16,47,55–58]. This approach simultaneously incorporates contributions from the uncertainty in the measured FID frequency, the uncertainty in the relaxation rate, and the fitting error arising from RF discharge noise.

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