

Zero-field atomic magnetometer to extract longitudinal magnetic field

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(Received 5 December 2024; revised 14 April 2025; accepted 18 July 2025; published 1 August 2025)

Zero-field atomic ensembles enable the development of ultrahigh-sensitivity magnetometry. However, their intrinsic insensitivity to longitudinal magnetic fields along the pumping direction poses a fundamental limitation, restricting precise weak-field quantification and complex system decoupling. Herein, we propose a novel zero-field weak parametric modulation scheme capable of ultrahigh-sensitivity extraction of longitudinal magnetic fields. Transverse modulation is employed to manifest the electron spin polarization projection induced by the longitudinal fields. Based on the perturbation model established, first-order parametric resonance associated with the longitudinal magnetic field yields significant signal enhancement under low-frequency and weak modulation conditions, while simultaneously suppressing spin-exchange relaxation caused by magnetic field modulation. We experimentally achieved a longitudinal measurement sensitivity of $16 \text{ fT/Hz}^{1/2}$, more than an order of magnitude improved over traditional schemes, with a bandwidth of 70 Hz. Benefiting from low spin relaxation, the noise floor of the transverse magnetic field measurement is preserved at $4 \text{ fT/Hz}^{1/2}$. The proposed scheme pushes the boundaries of existing weak magnetic field measurement, providing critical capabilities for state-of-the-art precision experiments including new physical quantity detection.

DOI: [10.1103/yjc6-b26g](https://doi.org/10.1103/yjc6-b26g)

Introduction. Optically pumped atomic ensembles detect weak magnetic fields encountered by means of Larmor precession, giving rise to various types of atomic magnetometers. Especially for zero-field atomic ensembles operated in the spin-exchange relaxation-free (SERF) regime, it has the potential for ultrahigh $\text{aT/Hz}^{1/2}$ -level measurement sensitivity due to the resolution of the contradiction between atomic density and spin-exchange relaxation, becoming a new generation magnetometer that can replace the superconducting quantum interferometer device [1–3]. In recent years, the ability of atomic magnetometers to precisely measure weak magnetic fields has been further expanded [4–7]. Zero-field atomic magnetometers have been used for multichannel magnetocardiography and magnetoencephalography systems, providing a trending method for detecting organ and tissue abnormalities [8–10]. In addition, atomic magnetometers have become a powerful way to explore new physical quantities such as the fifth force, as well as to detect extremely weak magnetism and magnetic moments of materials [11–13].

By absorbing photons in the light field, the alkali metal exhibits a longitudinal electron spin polarization direction

parallel to the pump beam [14,15]. Under the premise of consistent polarization orientation, when the atomic ensemble is exposed to an external magnetic field $\mathbf{B}$, it precesses around $\mathbf{B}$ under the action of the torque [16,17]. However, since a parallel magnetic field exerts no torque on the spin, the system is inherently insensitive to fields aligned with the initial spin polarization [18–20]. This defect in longitudinal magnetic field extraction has become a constraint on the ability of zero-field atomic magnetometers and remains an urgent issue to solve across various quantum measurement applications. Embodied in zero-field atomic magnetometers operating in the longitudinal open-loop mode, significant low-frequency scale factor drifts occur. These drifts introduce errors in weak magnetic field quantization due to asynchronous frequency responses during calibration and testing; furthermore, for co-magnetometers employing multiple spin species, deviations from the longitudinal field compensation point induce coupled measurements of physical quantities [21,22] and result in dynamic drifts linked to transverse magnetic fields arising from the weakened electron-nucleus spin correlation [23–25].

Several research groups have identified this challenge and are actively investigating approaches for longitudinal magnetic field extraction in zero-field atomic ensembles [26,27]. The single nuclear-species co-magnetometer achieves self-compensation through electron resonance locking, thereby extending the spin coherence time [25,28–30]. For enhanced measurement accuracy, bias magnetic fields are employed to rotate the initial electron spin polarization, obtaining a mapping of longitudinal magnetic fields on the spin polarization projection [31], but this causes serious aliasing in

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real-time measurements. Additionally, a series of methods classified as coordinate transformation have been successively proposed [27,32] to convert the initial longitudinal magnetic field into a relative transverse component perpendicular to another independent pumping system. These indirect approaches demonstrate limitations in mechanistically resolving longitudinal magnetic field responses within atomic ensembles, while inducing substantial coherence loss through the requisite application of high-amplitude multiaxis modulation [33–36]. Currently, two distinct schemes are employed to validate the direct longitudinal magnetic field detection capability of zero-field atomic magnetometers [37–40]. However, their sensitivity stagnates at pT/Hz^{1/2} or hundreds of fT/Hz^{1/2}. This substantial gap in sensitivity compared to conventional modes originates from spin-exchange relaxation induced by modulation fields and pronounced signal attenuation, greatly limiting the application of longitudinal magnetic field measurements.

This study achieved for the first time the highly sensitive real-time measurement of the longitudinal magnetic field by zero-field atomic magnetometers. We utilize a transverse weak modulation scheme to actively promote dynamic deviations of electron spin polarization from the optical pumping axis, enabling effective response separation of the longitudinal magnetic field in the polarization projection. Then, a complete analytical model is derived through systematic iteration of perturbation response terms. Under the combined effect of parametric modulation, the method achieves optimal signal gain in response to longitudinal magnetic fields at low modulation frequency, while the spin-exchange relaxation is almost eliminated. The effectiveness of the method has been verified both theoretically and experimentally. Compared with conventional approaches, the proposed scheme demonstrates a remarkable enhancement exceeding 1 order of magnitude in longitudinal field measurement sensitivity. This breakthrough methodology, based on zero-field atomic magnetometers, represents a significant advancement in weak magnetic detection. Furthermore, it establishes a groundbreaking paradigm for physical decoupling in quantum sensing applications.

Theoretical model. For the atomic ensemble in the SERF regime, alkali-metal atoms with their single valence electron can be effectively modeled as spin-1/2 particles, and the dynamic evolution model of the electron spin polarization $\mathbf{S}$ can be described by the Bloch equation. Assume that the pump beam is along the z axis and the probe beam is along the x axis. To decouple the longitudinal magnetic field, we apply a modulation magnetic field $B_{\text{mod}} \cos(\omega t)$ with a frequency of ω on the x axis. The model is transformed into

$$\begin{aligned}\dot{S}_x(t) &= \omega_y S_z(t) - \omega_z S_y(t) - \Gamma S_x(t), \\ \dot{S}_y(t) &= \omega_z S_x(t) - [\omega_x + \omega_{\text{mod}} \cos(\omega t)] S_z(t) - \Gamma S_y(t), \\ \dot{S}_z(t) &= [\omega_x + \omega_{\text{mod}} \cos(\omega t)] S_y(t) - \omega_y S_x(t) + R - \Gamma S_z(t),\end{aligned}\quad (1)$$

where $\omega_j = \gamma_e B_{0j}/q$; γ_e is the gyromagnetic ratio of the bare electron; B_{0j} is the magnetic field component to be measured, $j = x, y$, and z ; q is the nuclear slowing-down factor; $\omega_{\text{mod}} = \gamma_e B_{\text{mod}}/q$; B_{mod} is the amplitude of the modulation magnetic field; $\beta = \gamma_e B_{\text{mod}}/q\omega$ is the modulation factor, $\Gamma = (R_{\text{op}} +$

$R_{\text{rel}})/q$, $R = R_{\text{op}}/2q$; and R_{op} and R_{rel} are the pumping rate and the relaxation rate of the atomic ensemble, respectively. The relaxation rate R_{rel} incorporates contributions from the spin-destruction rate and the wall-collision rate.

Assuming the spin-polarization projections S_x , S_y , and S_z as independent variables, let $S_{\pm}(t) = S_z(t) \pm iS_y(t)$ and $\omega_{\pm} = (\omega_z \pm i\omega_y)/2$; then Eq. (1) is further transformed into integral form as

$$\begin{aligned}S_x(t) &= ie^{-\Gamma t} \int e^{\Gamma t} [\omega_- S_+(t) - \omega_+ S_-(t)] dt, \\ S_+(t) &= e^{-[i\omega_x t + i\beta \sin(\omega t) + \Gamma t]} \\ &\times \int e^{[i\omega_x t + i\beta \sin(\omega t) + \Gamma t]} [2i\omega_+ S_x(t) + R] dt.\end{aligned}\quad (2)$$

Since Eq. (2) is more than complicated and exists coupling between three-axis polarization projections, it cannot be directly solved in closed form. To separate the projection of the electron spin polarization along the x axis, we consider $\omega_{\pm}$ as the perturbation component and expand $S_x(t)$ and $S_{\pm}(t)$ in a perturbation series as follows:

$$\begin{aligned}S_x(t) &= S_x^{(0)}(t) + \dots + S_x^{(N)}(t) + \dots, \\ S_{\pm}(t) &= S_{\pm}^{(0)}(t) + \dots + S_{\pm}^{(N)}(t) + \dots,\end{aligned}\quad (3)$$

where the superscript N represents the order of expansions. Referring to the Dupont-Roc method [37,41], substituting the expanded form into the integral equation, the recursive relationships are obtained as

$$\begin{aligned}S_x^{(N)}(t) &= ie^{-\Gamma t} \int e^{\Gamma t} [\omega_- S_+^{(N-1)}(t) - \omega_+ S_-^{(N-1)}(t)] dt, \\ S_+^{(N)}(t) &= e^{-[i\omega_x t + i\beta \sin(\omega t) + \Gamma t]} \\ &\times \int e^{[i\omega_x t + i\beta \sin(\omega t) + \Gamma t]} 2i\omega_+ S_x^{(N-1)}(t) dt.\end{aligned}\quad (4)$$

For solvable undisturbed parts, i.e., $\omega_{\pm} = 0$, $S_x^{(0)}(t)$ and $S_{\pm}^{(0)}(t)$ have definite analytical solutions. Introducing the Jacobi-Anger expansion and considering the zero-order resonance as the dominant term, their expressions are

$$\begin{aligned}S_x^{(0)}(t) &= 0, \\ S_+^{(0)}(t) &= \frac{RJ_0(\beta)}{\Gamma + i\omega_x} e^{-i\beta \sin(\omega t)} = M^{(0)} e^{-i\beta \sin(\omega t)}, \\ S_-^{(0)}(t) &= S_+^{(0)*}(t) = M^{(0)*} e^{i\beta \sin(\omega t)},\end{aligned}\quad (5)$$

where J_k is the k th-order Bessel function of the first kind.

Based on solvable terms, the spin-polarization projection can be corrected step by step according to established recursive relationships until it approaches actual situations. The first-order correction term of $S_+(t)$ is

$$\begin{aligned}S_+^{(1)}(t) &= e^{-[i\omega_x t + i\beta \sin(\omega t) + \Gamma t]} \\ &\times \int e^{[i\omega_x t + i\beta \sin(\omega t) + \Gamma t]} 2i\omega_+ S_x^{(0)}(t) dt = 0\end{aligned}\quad (6)$$

and the corresponding correction of $S_x(t)$ is

$$\begin{aligned}S_x^{(1)}(t) &= ie^{-\Gamma t} \int e^{\Gamma t} [\omega_- M^{(0)} e^{-i\beta \sin(\omega t)} - \omega_+ M^{(0)*} e^{i\beta \sin(\omega t)}] dt \\ &= \sum_{n=-\infty}^{\infty} J_n(\beta) \left[i\omega_- M^{(0)} \frac{e^{-in\omega t}}{\Gamma - in\omega} - i\omega_+ M^{(0)*} \frac{e^{in\omega t}}{\Gamma + in\omega} \right].\end{aligned}\quad (7)$$

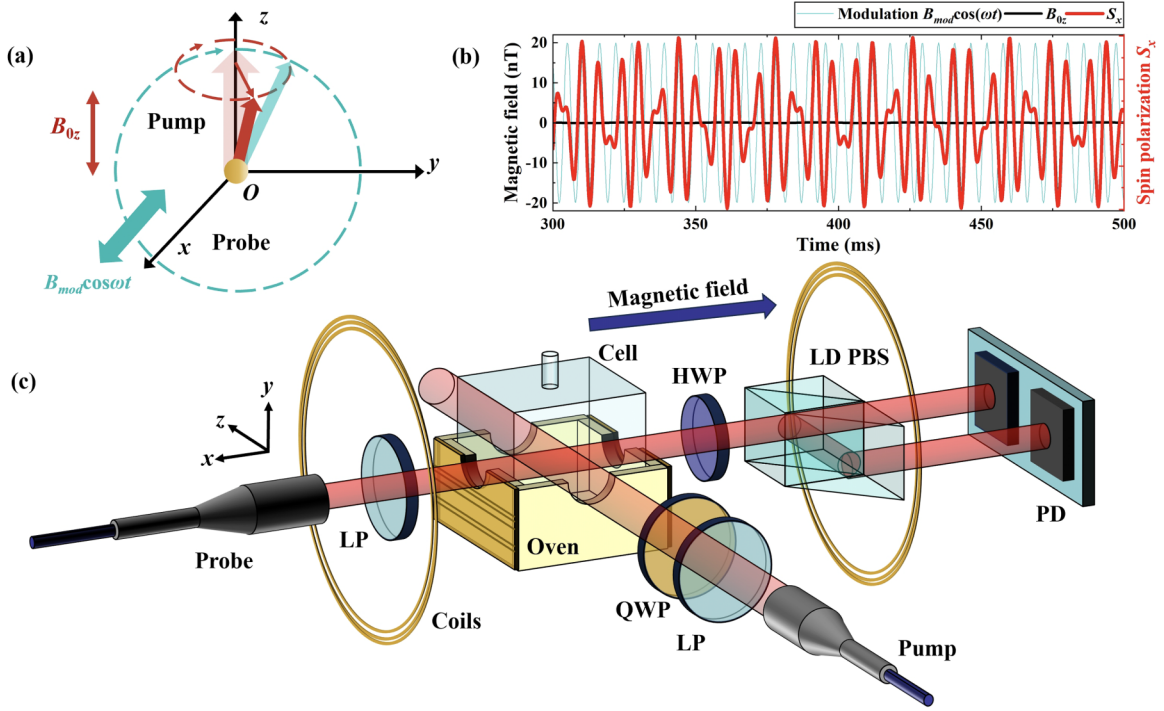


FIG. 1. Schematic of the longitudinal magnetic field extraction in a zero-field atomic magnetometer. (a) The initial spin polarization of the atomic ensemble is along the z axis upon absorption of pump photons. By applying a modulation magnetic field on the transverse x axis, the electron spin polarization is dynamically deviated from the z axis, so that the response of the atomic magnetometer to the longitudinal magnetic field B_z to be measured is manifested on the x -axis projection. (b) The time-domain simulation of spin-polarization projection S_x under the modulation magnetic field $B_{\text{mod}} \cos(\omega t)$ and the measured magnetic field B_{0z} . (c) Experimental setup for the measurements. LP, linear polarizer; HWP, half-wave plate; QWP, quarter-wave plate; LD PBS, lateral displacement polarized beam splitter; PD, photodetector.

Among the multiple resonance components constituting the correction term, their amplitudes decrease successively with increasing resonance order n . Therefore, by further iterating the spin polarization via the zero-order resonance, the second- and third-order correction terms are obtained as Eqs. (8) and (9), respectively:

$$S_x^{(2)}(t) = ie^{-\Gamma t} \int e^{\Gamma t} [\omega_- S_+^{(1)}(t) - \omega_+ S_-^{(1)}(t)] dt = 0, \quad (8)$$

$$S_x^{(3)}(t) = -\frac{2i\omega_+ \omega_- J_0^2(\beta)}{\Gamma} (\omega_- M^{(0)} - \omega_+ M^{(0)*}) \times \sum_{n=-\infty}^{\infty} J_n(\beta) \left[\frac{e^{-in\omega t}}{(\Gamma + i\omega_x)(\Gamma - in\omega)} + \frac{e^{in\omega t}}{(\Gamma - i\omega_x)(\Gamma + in\omega)} \right]. \quad (9)$$

It can be concluded that the even-order corrections of $S_x(t)$ are 0, while the amplitudes of odd-order terms decrease with the order. Taking into account the response attenuation of high-order parametric resonance, the DC and low-order harmonic components containing the main magnetic field information are decomposed. $S_x(t)$ is therefore modified by these recursive resonance terms according to Eq. (3). Ignoring

the irrelevant minor terms, the analytical expressions of $S_x(t)$ after third-order correction are

$$S_x(t) \Big|_{n=0} = R\Gamma J_0^2(\beta) \frac{\Gamma\omega_y + \omega_x\omega_z}{(\Gamma^2 + \omega_x^2)^2}, \quad (10)$$

$$S_x(t) \Big|_{n=\pm 1} = \frac{2R\Gamma^2}{\Gamma^2 + \omega^2} J_0(\beta) J_1(\beta) \frac{\Gamma\omega_z - \omega_x\omega_y}{(\Gamma^2 + \omega_x^2)^2} \times [\Gamma \sin(\omega t) - \omega \cos(\omega t)], \quad (11)$$

$$S_x(t) \Big|_{n=\pm 2} = \frac{2R}{\Gamma^2 + 4\omega^2} J_0(\beta) J_2(\beta) \frac{\Gamma\omega_y + \omega_x\omega_z}{(\Gamma^2 + \omega_x^2)^2} \times [\Gamma^3 \cos(2\omega t) + 2\Gamma^2 \omega \sin(2\omega t)]. \quad (12)$$

Special for the proposed modulation scheme, the resonance orders correspond to the harmonics. Among them, since the magnetic field measured by the zero-field atomic magnetometer is extremely weak, i.e., $\omega_j/\Gamma \ll 1$, its higher-order terms become safely negligible. Consequently, the DC component exhibits sensitivity to the transverse magnetic field along the y axis, as described by Eq. (10), while the primary oscillation term responds primarily to the longitudinal magnetic field B_{0z} , as expressed in Eq. (11). The relative response amplitude between the two is closely related to the total relaxation rate and the modulation parameters. Therefore, it is completely feasible to achieve independent measurement of the longitudinal magnetic field with ultrahigh sensitivity while enabling real-time dual-axis detection by means of simultaneous

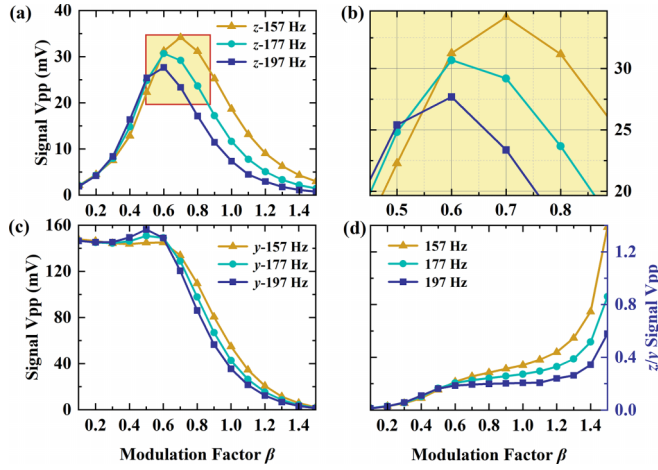


FIG. 2. Relationship between the response intensity to magnetic fields and the modulation factor β at different modulation magnetic field frequencies ω . The frequencies were set to 157, 177, and 197 Hz, respectively, represented by yellow, green, and blue lines. Figure (b) shows a partial enlargement of panel (a). The modulation factor varies accordingly by the modulation amplitude at a certain frequency. (a) The response intensity to the longitudinal magnetic field exhibits a peak, which is related to the product of Bessel functions, i.e., $J_0 J_1$, and (b) the enhanced output response relies on the low modulation frequency. Similarly, (c) the relationship between the response of transverse magnetic fields and the modulation factor is affected by the square of the zero-order Bessel function, showing a decreasing trend. The distortion of curves stems from the frequency response characteristic of the low-pass filter. (d) The relative ratio of the output response to longitudinal and transverse magnetic fields with the same magnitude. Under the appropriate modulation frequency ω and factor β , close signal gain and optimal longitudinal response can be obtained.

decoupling of the DC and primary oscillation terms in zero-field atomic magnetometers.

Results. A schematic of the experimental setup is shown in Fig. 1(c). A cubic glass cell with a side length of 14 mm and a wall thickness of 1 mm is used as a sensitive element to confine alkali-metal ^{87}Rb atoms and approximately 500 Torr N_2 . This vapor cell is placed in a boron nitride oven with a pair of flexible heating films attached to two side surfaces. The oven and the vapor cell are heated to 160°C by applying an alternating current with a frequency of 180 kHz in heating films. Both pumping and detection for the alkali-metal atoms use distributed feedback diode lasers with a spot size of 7.8 mm. The pump beam is tuned to the D1 line of ^{87}Rb atoms, with a power of 6.3 mW; the probe beam is right-detuned by 70 GHz, with a power of 4.8 mW. The entire experimental setup is enclosed in a magnetic shielding system comprising four permalloy layers supplemented with low-noise ferrite to suppress magnetic noise below $1 \text{ fT/Hz}^{1/2}$. The uniform magnetic field coils inside the magnetic shield are used to compensate and regulate three-axis magnetic field components, driven by the signal generators.

The magneto-optical rotation angle of the probe beam transmitted through the vapor cell, expressed as Eq. (13), is measured via balanced polarimetry, which can suppress

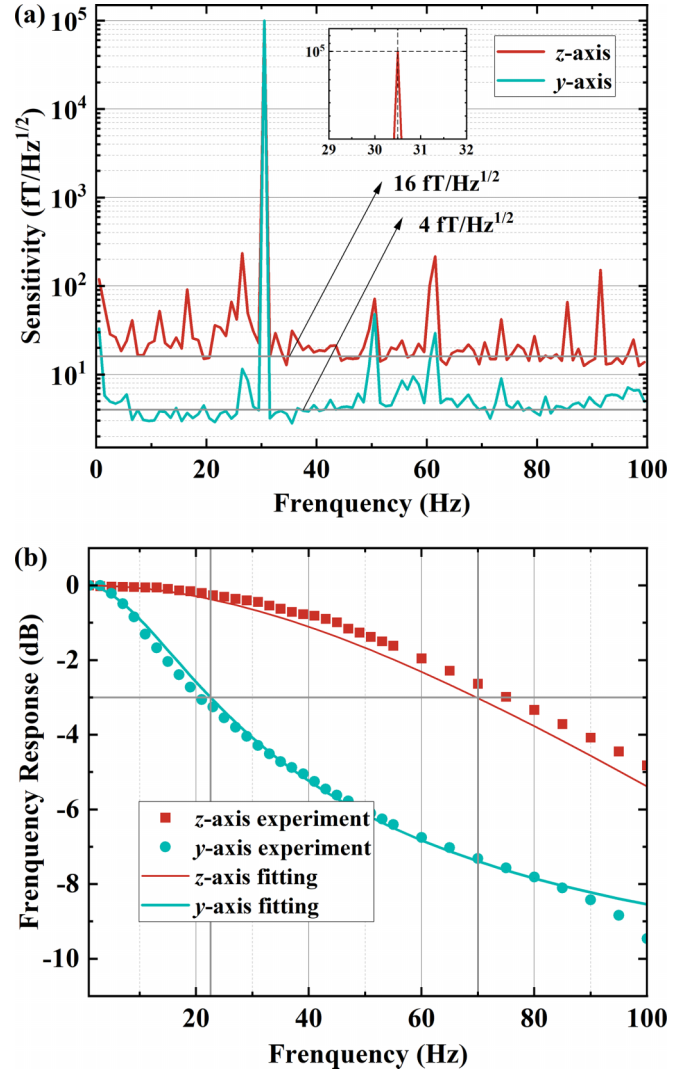


FIG. 3. (a) Under the proposed scheme of transverse weak modulation, the z -axis magnetic field detection sensitivity is improved to $16 \text{ fT/Hz}^{1/2}$, while the sensitivity of the y -axis magnetic field in the DC component is $4 \text{ fT/Hz}^{1/2}$. The sensitivity is characterized by the power spectral density, averaged over 1-Hz intervals. The results include responses to calibrated magnetic fields with an amplitude of 100 pT rms and a frequency of 30.5 Hz and their harmonics, as well as additional peaks introduced by industrial frequency interference. (b) Frequency response of the zero-field atomic magnetometer to the magnetic field on the y and z axes. The longitudinal magnetic field measurement bandwidth is increased to 70 Hz.

common-mode detection noise:

$$\theta = \frac{1}{2} l n_{\text{Rb}} r_e c f_{\text{D1}} \frac{\nu - \nu_{\text{D1}}}{(\nu - \nu_{\text{D1}})^2 + (\Gamma_{\text{D1}}/2)^2} S_x, \quad (13)$$

where l represents the optical path length through the vapor cell, n_{Rb} denotes the number density of rubidium atoms, r_e is the electron radius, f_{D1} is the transition frequency of the D1 line, $(\nu - \nu_{\text{D1}})$ is the detuning of the probe light frequency from the D1 resonance, and Γ_{D1} is the pressure broadening fullwidth of the D1 transition. In addition, the employed vapor cell exhibits a linewidth Γ_{D1} of approximately 13 GHz, so

far-detuned linearly polarized light enables precise measurements of atomic spin states.

The current signal is converted into the voltage by a transimpedance amplifier and then transmitted to a low-pass filter and a lock-in amplifier to decouple the DC and first-harmonic components, respectively. The reference end of the lock-in amplifier is connected to the synchronous output of the modulation signal generator and its output end is connected to the signal acquisition system to evaluate the noise floor through power spectral density (PSD) analysis. The experimental responses of the magnetometer to magnetic fields on the y and z axes are shown in Fig. 2. It can be seen that the signal amplitude is closely related to the modulation factor associated with the Bessel functions [42,43]. In addition, since the first harmonic component exhibits an approximately quadratic decay with respect to the modulation frequency following from Eq. (11), the signal amplitude related to the longitudinal magnetic field obtains the optimized gain at low modulation frequency. Therefore, in contrast to conventional high-amplitude magnetic field modulation, this study introduces a weak modulation scheme. This scheme significantly enhances the longitudinal field response in zero-field atomic magnetometers while effectively circumventing the issue of aggravated spin-exchange relaxation.

To prevent the applied low-frequency modulation from raising the overall noise floor, we adopted a modulation frequency of 177 Hz in the experiment to verify the longitudinal measurement sensitivity under the corresponding optimal modulation factor. The amplitude of the modulation magnetic field was set to about 20 nT. The corresponding frequency response curves of the y - and z -axis magnetic fields, as well as the sensitivity of the magnetometer, were measured. Besides, we additionally applied a calibration magnetic field at 30.5 Hz with an amplitude of 100 pT rms for verification purposes. As shown in Fig. 3, we have achieved a longitudinal measurement sensitivity of $16 \text{ fT/Hz}^{1/2}$ while maintaining a noise floor of $4 \text{ fT/Hz}^{1/2}$ in the y axis. Experimental verification confirms that our real-time measurement scheme greatly improves the sensitivity of zero-field atomic magnetometers in measuring longitudinal magnetic fields and

exhibits low-frequency response characteristics comparable to those of transverse magnetic fields, which is of great significance for various quantum physics researches. In addition, this scheme improves the working bandwidth of the dual-beam zero-field atomic magnetometer. The measured -3 dB bandwidth reaches approximately 70 Hz on the z axis, representing a threefold improvement over the 23-Hz bandwidth achieved on the y axis.

Conclusion. In this Letter, we present a novel scheme for ultrahigh-sensitivity measurements of the longitudinal magnetic field in a zero-field atomic magnetometer and provide a complete perturbative analytical model. Eliminating conventional requirements for strong modulations or auxiliary optics, this methodology enables direct extraction of the first-harmonic response associated with longitudinal magnetic fields through optimized weak modulation with nearly eliminated spin-exchange relaxation. Experiments have verified that the longitudinal measurement sensitivity and bandwidth have reached $16 \text{ fT/Hz}^{1/2}$ and 70 Hz, respectively. This parametric resonance scheme breaks through the limitation of extracting longitudinal magnetic field by zero-field atomic magnetometers, which achieves more than an order of magnitude improvement in sensitivity. This research creates feasible conditions for weak magnetic field measurements of atomic magnetometers in the field of quantum metrology; this also paves the way for the real-time control of the longitudinal self-compensation points of multispin species ensembles, which can significantly increase the coherence and stability in systems such as Lorentz invariance testing, searches for anomalous spin-dependent force, and inertial rotation sensing [29,44,45], providing broader prospects for applications in various related fields.

Acknowledgments. This work is supported by the Innovation Program for Quantum Science and Technology (Grant No. 2021ZD0300502), the National Natural Science Foundation of China (Grant No. 62473018), and the Academic Excellence Foundation of BUAA for Ph.D. Students.

Data Availability. The data that support the findings of this article are openly available [46].

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