

New Constraints on Axion-Mediated Spin Interactions Using Magnetic Amplification

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Axions are highly motivated hypothetical particles beyond the standard model that can be dark matter candidates and address the strong CP problem. Here we search for axion-mediated interactions generated between two separated ^{129}Xe gas ensembles, monitoring and polarizing the ^{129}Xe nuclear spins through spin-exchange interactions with Rb vapor. Our method exploits the magnetic amplification through effective fields from Rb-Xe collisions, increasing the sensitivity for axion-mediated interactions by up to 145-fold relative to conventional methods. Moreover, we employ template filtering to extract exotic interactions with a maximum signal-to-noise ratio. By combining two techniques, axion-mediated interactions are constrained to be less than 10^{-5} of normal magnetic interactions on a length scale of 60 mm. We establish new constraints on the neutron-neutron pseudoscalar couplings for a mass range that expands into the well-motivated “axion window” (10 μeV –1 meV), improving previous constraints by up to 50-fold within it and 118-fold outside it. We further discuss promising applications in searches for other axion-nucleon interactions, including axion dark matter and black hole axion bursts with sensitivity well beyond astrophysical limits by several orders of magnitude.

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Introduction—Axion and axionlike particles predicted by numerous theories embed the standard model of particle physics into a more general, unified framework [1–8]. The main challenge encountered in the search for axions stems from their wide-ranging mass. Several theories (e.g., high-temperature lattice QCD [9], the standard model axion seesaw Higgs portal inflation model [10], and axion-string networks [11–15]) propose the axion window (10 μeV –1 meV) as one of the most likely regions of parameter space where axions may exist. Previous constraints from laboratory searches (e.g., ADMX) and astrophysical observations (e.g., SN1987A) mostly explored mass ranges outside the window [16,17,17,18,18–25]. Recently, this mass range has been explored in both haloscope and “fifth-force” experiments, while other experiments are in preparation. The haloscope experiments include HAYSTAC [26], CERN axion solar telescope [27], ORGAN [28,29], MADMAX [30], and the Orpheus experiment [31]. Fifth-force experiments include ARIADNE [32] and SAPPHIRE [25].

Axions are predicted to mediate exotic interactions by a variety of theories beyond the standard model [1,2,33–36]. The basic experimental scheme to search for exotic interactions is to employ a sensitive detector of torques or forces on particles from a source on a certain length scale [13,20,32,37–53]. Searches for axion-mediated interactions have the advantage of covering many orders of magnitude in the axion mass, including the axion window, without scanning. Specifically, the pseudoscalar coupling to spin-polarized fermions is of particular theoretical interest because it is predicted by numerous theories with a broken high-energy symmetry [54] and is similar to the propagating torsion field predicted by several extensions of general relativity [55]. However, normal (i.e., standard model) magnetic interactions hinder searching for the exotic interactions. Previous experiments, respectively, constrained exotic interactions mediated by light axions ($<1 \mu\text{eV}$) to be less than 10^{-5} and 10^{-8} of their normal magnetic interactions on the length scale of 400 and 500 mm between two polarized neutron ensembles [20,21]. The constraints fall off exponentially with increasing axion mass and thus become less stringent within the axion window. A pioneering experiment by Ramsey [56] constrained the exotic interactions in this range to be less than 10^{-3} of normal

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magnetic interactions. Therefore, it is imperative to develop novel techniques in searches for exotic spin-dependent interactions.

In this Letter, we demonstrate magnetic amplification to search for exotic spin-dependent interactions between two separated ^{129}Xe spin ensembles that are mediated by axions within the axion window. Tailoring a spin-monitoring technique assisted by an *in situ* ^{87}Rb magnetometer, our technique demonstrates remarkable amplification for pseudomagnetic fields induced by the exotic interactions by up to 145-fold. Through the combination of the template filtering and high sensitive spin-monitoring technique, our Letter maximizes the signal-to-noise ratio to extract the sought-after signal from exotic interactions. As a first application, we constrain the exotic interactions between polarized ^{129}Xe spins to be less than 10^{-5} of normal magnetic interactions on a length scale of 60 mm. New constraints on neutron-neutron spin coupling are placed in the mass range (3.2–24.3 μeV) expanding into the axion window and improve previous experimental constraints by up to 50-fold within the window and 118-fold outside it. Although demonstrated for the exotic spin-dependent interactions, the magnetic amplification can be applied to various axion-nucleon interactions, including axion dark matter and axion bursts from astrophysical events [57,58]. With current experiments, the sensitivity should exceed astrophysical limits [59,60] by several orders of magnitude.

Axion-mediated spin-dependent interactions—The setup contains two cells: a spin-sensor cell I with 0.026 amg ^{129}Xe , 0.33 amg N_2 , and a droplet of ^{87}Rb ; and a spin-source cell II with 3 amg ^{129}Xe , 0.066 amg N_2 buffer gas, and a droplet of ^{87}Rb metal. Each cell is shielded by a magnetic shield and placed within a five-layer magnetic shield, as shown in Fig. 1(a). The centers of the two cells are ≈ 60.5 mm apart from each other. In each cell, ^{87}Rb spins are polarized using a circularly polarized beam of 795 nm D1 light and ^{129}Xe spins are polarized via spin-exchange collisions with polarized ^{87}Rb spins [16,61,62]. The fractional contribution of neutrons in ^{129}Xe spins is 0.73 [63]. The axion-mediated interactions between polarized neutrons are ($c = \hbar = 1$) [34–36]

$$V_{\text{ps-ps}} = \frac{g_{\text{ps}}^n g_{\text{ps}}^n}{16\pi m_n m_n} \left[\hat{\sigma}_{\text{I}} \cdot \hat{\sigma}_{\text{II}} \left(\frac{m_a}{r^2} + \frac{1}{r^3} \right) - (\hat{\sigma}_{\text{I}} \cdot \hat{r})(\hat{\sigma}_{\text{II}} \cdot \hat{r}) \left(\frac{m_a^2}{r} + \frac{3m_a}{r^2} + \frac{3}{r^3} \right) \right] e^{-m_a r}, \quad (1)$$

where g_{ps}^n is the neutron pseudoscalar coupling constant, m_n is the neutron mass, m_a is the axion mass, the spin vector $\hat{\sigma}_{\text{I,II}}$ is labeled with subscript I for sensor and II for source (the same for other parameters), and r (\hat{r}) is the distance (unit vector) between the two interacting neutrons. The pseudomagnetic fields \mathbf{B}_p experienced by the nuclei in the sensor cell can be deduced from

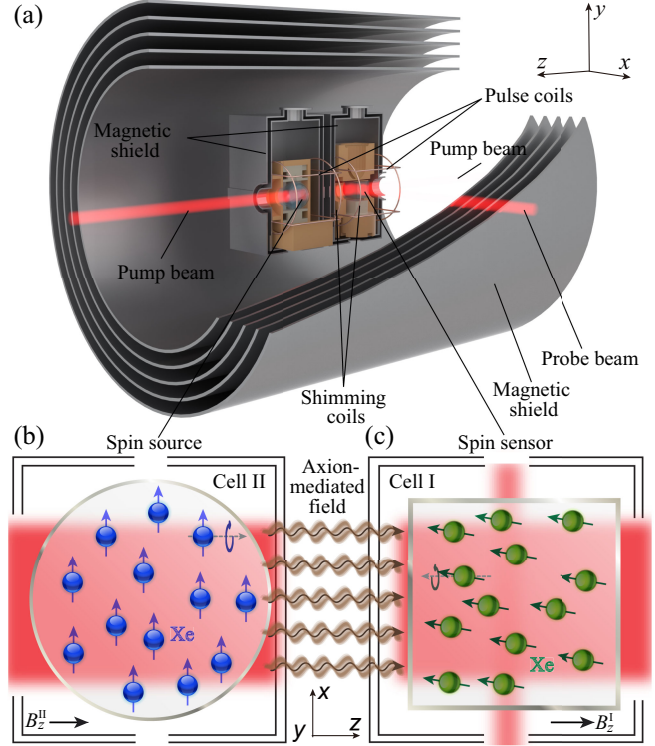


FIG. 1. Schematic of the experiment. Polarized ^{129}Xe spins in the source cell I are tilted into the x - y plane via a $\pi/2$ pulse and precess about the z axis, which generates free-decay pseudomagnetic fields on ^{129}Xe spins in the sensor cell II. (a) A three-dimensional layout. (b) A 795 nm pump beam tuned to the ^{87}Rb D1 transition delivers maximum 1.0 W of power to the source cell for optical pumping ^{87}Rb spins along \hat{z} . (c) In the sensor cell, ^{87}Rb spins are polarized with a circularly polarized beam of 795 nm D1 light and probed via optical rotation of a linearly polarized probe beam, which is detuned by 110 GHz to higher frequency from the D2 resonance.

$\mathbf{B}_p \hat{\sigma}_{\text{I}} = -V_{\text{ps-ps}} / \mu_{\text{Xe}}$, where μ_{Xe} is the magnetic moment of ^{129}Xe nuclei. Figure 1(b) shows that free-decay pseudomagnetic fields are generated by precessing ^{129}Xe source spins in the x - y plane after a $\pi/2$ pulse

$$\mathbf{B}(t) = |\mathbf{B}_p| N_{\text{II}}^* e^{-t/T_{\text{II}}^*} [\cos(2\pi\nu_{\text{II}} t) \hat{x} + \sin(2\pi\nu_{\text{II}} t) \hat{y}], \quad (2)$$

where N_{II}^* is the total number of polarized ^{129}Xe spins, T_{II}^* is the effective coherence time, and ν_{II} is the ^{129}Xe Larmor frequency of source spins. The pseudomagnetic fields have both x and y components [64].

Magnetic amplification—Magnetic and pseudomagnetic fields $\mathbf{B}(t)$ collectively excite ensembles of polarized ^{129}Xe -sensor spins, as shown in Fig. 1(c). Because of Fermi-contact interactions [66–69], the transverse magnetization generated by these spins produces effective fields $\tilde{\mathbf{B}}(t)$ on ^{87}Rb spins [64],

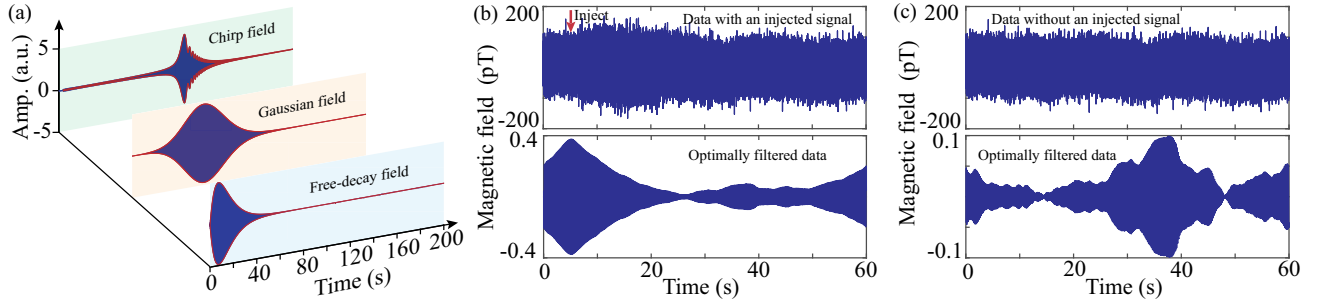


FIG. 2. Demonstration of the magnetic amplification and template filtering. (a) ^{129}Xe effective fields excited by various external fields. The envelope of experimental data (blue line) shows good agreement with the theoretical fit (red line). (b) Calibration of an injected signal with the template filter. The amplitude and arrival time of the injected signal can be exactly extracted (see main text). (c) Calibration of sensitivity.

$$\begin{bmatrix} \tilde{B}_x(\nu) \\ \tilde{B}_y(\nu) \end{bmatrix} \approx \eta \frac{1/T_I^*}{i2\pi(\nu - \nu_1) + 1/T_I^*} \begin{bmatrix} 1 & 1 \\ -1 & 1 \end{bmatrix} \begin{bmatrix} B_x(\nu) \\ B_y(\nu) \end{bmatrix}, \quad (3)$$

where T_I^* is the effective coherence time (subscript I for sensor) and ν_1 is the ^{129}Xe Larmor frequency. The ^{87}Rb magnetometer can simultaneously measure the external magnetic fields and the ^{129}Xe effective fields excited by external fields [64]. In the resonant case ($\nu \approx \nu_1$), the amplitude of the ^{129}Xe effective fields $\tilde{\mathbf{B}}(\nu_1)$ is much greater than that of the external fields $\mathbf{B}(\nu_1)$, improving the sensitivity of the ^{87}Rb magnetometer. Such enhancement can be characterized with an amplification factor $\eta = |\tilde{\mathbf{B}}(\nu_1)|/|\mathbf{B}(\nu_1)| = 4\pi\kappa_0 M_1 P_1^0 \gamma_n T_I^*/3$ [64], where M_1 is the ^{129}Xe maximum magnetization associated with full spin polarization, P_1^0 is the ^{129}Xe equilibrium polarization, and γ_n is the ^{129}Xe gyromagnetic ratio. The Fermi-contact enhancement factor κ_0 that characterizes the frequency shifts for Rb- ^{129}Xe is measured to be 493 ± 31 [66–69]. Tailoring spin-monitoring experiments for high sensitivity using the effective fields was also demonstrated for K-He collisions [70], a system which was used to produce limits on pseudoscalar interactions [20] and Lorentz- and *CPT*-violating spin-spin interactions [71]. Recently, Rb-Xe effective fields have been used with large enhancements in the sensitivity to Xe spin for a transverse nuclear magnetic resonance gyroscope [72,73].

Equation (3) provides the ^{129}Xe effective fields in both frequency and time domains. Both x and y components of pseudomagnetic fields can induce rotating ^{129}Xe effective magnetic fields in the sensor. The resonant effective fields induced by free-decay pseudomagnetic fields in Eq. (2) are [64]

$$\tilde{B}_y(t) = 2\eta |\mathbf{B}_p| N_{\text{II}}^* \frac{e^{-t/T_I^*} - e^{-t/T_{\text{II}}^*}}{1 - T_I^*/T_{\text{II}}^*} \cos(2\pi\nu_1 t + \phi), \quad (4)$$

where ϕ is the phase from rectangular $\pi/2$ pulses [64]. The response for $-\pi/2$ pulses includes a negative sign. The parameters of spin sensor, for example, $\nu_1 \approx 5.360$ Hz and $T_I^* \approx 7.63$ s, are obtained by measuring the free-decay signal of the ^{129}Xe spins. $\eta \approx 145$ is calibrated by applying auxiliary continuous fields, one at the resonance frequency ν_1 and the other—far from resonance—at 70 Hz. Subsequently, Eq. (4) is experimentally verified by applying a synthetic “free-decay” resonant field with a strength of ≈ 10.0 pT and a coherence time of $T_{\text{II}}^* \approx 7.04$ s along y [see Fig. 2(a)].

To demonstrate the characteristics of noble-gas magnetic amplification, Fig. 2(a) also shows the experimental response to a Gaussian field and a chirp field with linearly modulated frequency. The theoretical fitting based on Eq. (3) is in good accordance with experimental results [64]. More examples are presented in Supplemental Material [64].

We have significantly enhanced the sensitivity to weak fields in our sensor cell by integrating two experimental techniques. First, resonant interactions between pseudo-magnetic fields and the sensor’s spins maximize the ^{129}Xe transverse polarization in the sensor cell for resonant source fields. Second, the ^{129}Xe transverse polarization in the sensor cell via Rb- ^{129}Xe Fermi-contact interactions substantially amplifies the detected ^{129}Xe signal [66–69]. To realize our full sensitivity gain, a meticulous selection of several key physical parameters $\{\kappa_0, M_1, P_1^0, T_I^*\}$ is required (see Supplemental Material [64] for details). This combined signal enhancement can be considered an effective amplification of the weak fields, which we refer to as “magnetic amplification,” and is linear for the weak fields considered here.

Noise reduction using a signal template—The main objective of our experiment is to extract the unknown amplitude of the sought-after signal with the known template [see Eq. (4)] by maximizing signal-to-noise ratio. To achieve this, we use a template filtering technique akin

to that used by the Laser Interferometer Gravitational Wave Observatory in gravitational wave detection [74]. The template filter $\Omega(\nu) = \alpha \tilde{B}_y^*(\nu)/S_n(\nu)$ is designed based on the power spectral density of noise $S_n(\nu)$ including color noise and technical noise peak [75] and functional form of \tilde{B}_y , where α is the normalization factor. The extracted field $B_y^{\text{exp}}(t)$ is [64]

$$B_y^{\text{exp}}(t) = \int_{-\infty}^{\infty} e^{i2\pi\nu t} \Omega(\nu) \tilde{B}_y(\nu) d\nu. \quad (5)$$

The signal contributes coherently to the cross-correlation with the template, while the noise contributes incoherently and thus is reduced relative to the signal. The filter is proven to be the optimal linear filter for maximizing the signal-to-noise ratio. Additionally, while not relevant for the present exotic-force search, template filtering can be used to extract signals with unknown arrival times, which is difficult to achieve using conventional spectral analysis [64]. For white noise in our case, the template filter is reduced to the matched filter.

The parameters of template are calibrated by a series of experiments [64]. Former experiments calibrate parameters of the sensor cell (ν_I , η , and T_I^*) and the rest of the source cell (N_{II}^* , T_{II}^* , ν_{II} , and ϕ) are calibrated using the atomic magnetometer itself. A small-angle pulse is applied to measure the free-decay signal through the ^{87}Rb magnetometer, and the coherence time T_{II}^* and the Larmor frequency ν_{II} can be directly obtained by fitting the signal. To measure the polarization, an auxiliary continuous field is applied to obtain the conversion coefficient from magnetic field to voltage. Then the amplitude of the decay signal is transformed into the field strength and compared to the effective magnetic field of 3 amg of fully polarized ^{129}Xe to obtain the polarization [66–69].

With the calibrated template, we apply the template filter to the data with or without an injected signal. The injected signal with the same template and a field strength of ≈ 0.4 pT arrives at unknown times. As shown in Fig. 2(b), the injected signal is almost hidden inside the noise because of the 50-Hz power line interference. With a remarkable improvement of the signal-to-noise ratio, the strength and arrival time (5 s) is exactly extracted. We further demonstrate the improved magnetic sensitivity with template sensing for a 60-s single-shot experiment. Figure 2(c) shows a time-domain sensitivity of 28.1 pT for the ^{87}Rb magnetometer. A further improvement is achieved by applying the template filter. The amplitude of the filtered noise at 5 s represents the extracted field strength for a single-shot experiment. By applying the template filter to 1000 such experiments, the standard deviation of extracted strength corresponds to a sensitivity of ≈ 30 fT, which improves the sensitivity by at least 2 orders of magnitude.

Search experiments and data analysis—The ^{129}Xe response $\tilde{B}_y(\nu)$ can be maximized by optimizing the

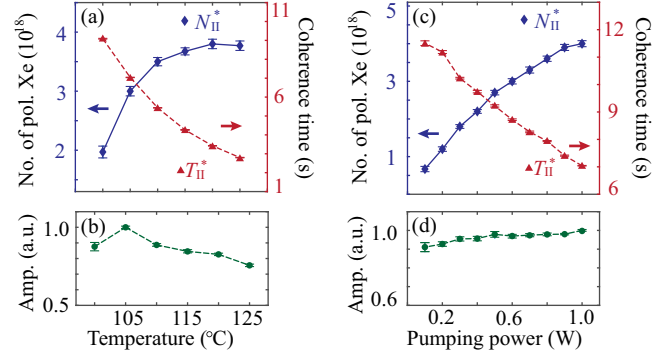


FIG. 3. Calibration of spin-source parameters and optimization of them for maximizing \tilde{B}_y . (a) The coherence time (red line) is reduced but the polarization (blue line) is increased with increasing temperature. (b) The amplitude of $\tilde{B}_y(\nu)$ reaches maximum at 105 °C. (c) The coherence time (red line) is reduced but the polarization (blue line) is increased with increasing pumping power. (d) The amplitude of $\tilde{B}_y(\nu)$ shows marginal increase up to the maximum available laser power (1.0 W) emitted into the source cell.

operation parameters of the spin source such as temperature, pumping power, pressure, etc. We first fix the pump power at 1.0 W, change the temperature to obtain number of tilted polarized ^{129}Xe N_{II}^* and coherence time T_{II}^* . It is found that the polarization increases but the relaxation time reduces with increasing temperature, as shown in Fig. 3(a). The amplitude of $\tilde{B}_y(\nu)$ is calculated and reaches maximum around 105 °C, as shown in Fig. 3(b). Subsequently, we set the operation temperature at 105 °C and change the pumping power. Figure 3(c) shows that the polarization increases but the relaxation time reduces with the increasing power. The decrease of the coherence time can be caused by the gradient field of nonuniform ^{87}Rb polarization along the radial direction, because of the inhomogeneity of light intensity of pump beam and edge of the vapor cell that is not illuminated. The amplitude of $\tilde{B}_y(\nu)$ shows marginal increase up to the highest power used of 1.0 W, as shown in Fig. 3(d). At 105 °C and 1.0 W, the experimental parameters are $\nu_{II} \approx 5.360$ Hz, $N_{II}^* \approx 4.0 \times 10^{18}$, and $T_{II}^* \approx 7.04$ s.

To induce the pseudomagnetic fields, $\pm\pi/2$ pulses are alternately applied every 60 s to initiate a single-shot experiment. Subsequently, the single-shot experiment is repeated 1200 times over a 20-h data collection period to generate a single dataset (see Supplemental Material [64] for details). Then the sensitivity of the spin sensor is optimized and recorded. The experimental conditions (e.g., temperature, pump power, laser frequency) of the spin source are monitored and adjusted to maintain stable operation. The data collection is repeated 36 times over a 720-h data collection period, yielding a total of 36 datasets. The raw spin-sensor data are segmented into ≈ 60 s segments based on the pulse sequence, because the response of ^{129}Xe spins spans approximately 60 s [see Eq. (4)], as shown

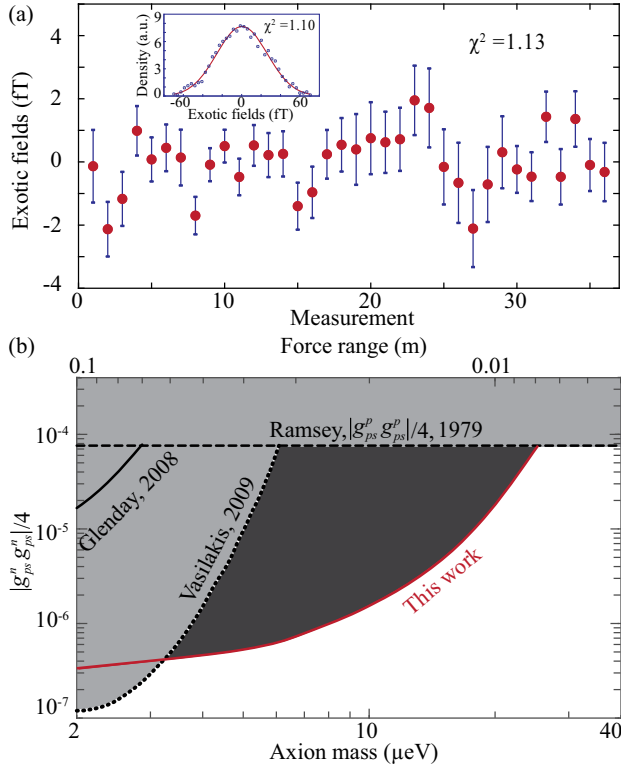


FIG. 4. Experimental results. (a) Each point represents the average and statistical error of exotic fields for a 20-h dataset. Inset: histogram of a 20-h dataset with a Gaussian fit in red line. (b) Constraints on $|g_{ps}^n g_{ps}^n|/4$. The black line represents the results from previous experiments of Ramsey [56], Glenday *et al.* [21], and Vasilakis *et al.* [20]. The red line represents the new constraints, improving former constraints by a factor about 118 at 6.04 μeV and 50 at 10 μeV .

in Fig. 2(a). The normalized template filter is then applied to each segment. The first data point of the resulting filtered signal is extracted as the measured exotic field. A typical distribution of the exotic fields deduced from 20-h data shows a standard deviation of ≈ 30 fT, as shown in the inset of Fig. 4(a).

Using all the 720 h of data, the exotic fields are measured to be $(-86 \pm 140_{\text{stat}})$ aT between the two separated neutron ensembles, which is less than 10^{-5} of their magnetic interactions on a length scale of 60 mm. The spurious magnetic fields generated by the polarized ^{129}Xe spins and magnetic pulses are suppressed by the magnetic screening effect originating from the attenuation due to distance and the use of magnetic shields. The screening factor is over 10^{10} through cross-check from theoretical calculations, numerical simulations, and experimental calibrations [64,76]. The ^{129}Xe classical dipole fields are reduced to ≈ 0.2 aT and magnetic pulses are reduced to 50 aT with 20 ms duration, both of which are negligible.

We include possible systematic errors in the calibration that come from variations in number of polarized ^{129}Xe N_{II}^* ,

coherence time T_{I}^* and T_{II}^* , position, resonance frequency ν_{II} , the phase ϕ , and the amplification factor η . By measuring all these factors and combining the corresponding uncertainties in quadrature, which are assumed to be independent of each other, the total uncertainty in the systematic error is ≈ 45 aT [64]. We thus find no evidence for the axions and quote the final total coupling strength $|g_{ps}^n g_{ps}^n|/4$ as $(2.0 \pm 3.2_{\text{stat}} \pm 1.1_{\text{sys}}) \times 10^{-7}$ at $m_a = 4.94 \mu\text{eV}$.

Figure 4(b) shows the obtained constraints together with those from previous experiments [20,21,56]. The red line represents the new constraints with 1σ limit, part of which reaches into the unexplored parameter space within the axion window. The dashed line represents the first constraint of exotic spin-spin interactions derived by Ramsey [56]. Then the maser [21] (the solid line) and the comagnetometer [20] (the dotted line) experiment placed constraints on $|g_{ps}^n g_{ps}^n|/4$ outside the axion window. For the mass range from 3.2 to 24.3 μeV , we set the most stringent constraints on $|g_{ps}^n g_{ps}^n|/4$, improving previous constraints by a factor about 118 at 6.04 μeV and 50 at 10 μeV within the axion window. We can also place constraints on proton-proton pseudoscalar couplings $|g_{ps}^p g_{ps}^p|/4$, which improve on those from Ramsey’s measurements [20,56] by up to 16-fold.

Conclusion and discussion—In this Letter, we report the magnetic amplification and template filtering and apply them to search for axion-mediated spin-dependent interactions. These techniques extract the template signal with a spin amplification of at least 2 orders of magnitude and a maximum signal-to-noise ratio. Using our technique, new constraints on pseudoscalar couplings are set within the axion window.

Our technique can be applied to probe axion dark matter directly coupling to the ^{129}Xe neutron spins (no spin source is needed). For “axion wind,” axions generate oscillating exotic fields proportional to the coupling constant g_{aNN} [77]. The spin sensor is sensitive to the axions within a narrow bandwidth centered at 5.36 Hz. The current sensitivity $g_{\text{aNN}} \approx 10^{-9} \text{ GeV}^{-1}$ could exceed the astrophysical limits [59,60] in a frequency range of $\approx 5.33\text{--}5.39$ Hz. Our technique can also be employed to search for topological defect dark matter consisting of axions [57]. The pseudo-magnetic fields of it are similar to magnetic pulses, which could induce free-decay ^{129}Xe effective magnetic fields. The functional form of these free-decay fields is the template for topological defect dark matter searches. By combining the magnetic amplification and template filtering, the 720-h data correspond to a sensitivity to the coupling of topological defect dark matter and nuclear spins $\approx 10^{-10} \text{ GeV}^{-1}$, exceeding the astrophysical limit [59,60] with the mass range of $10^{-10}\text{--}10^{-7} \text{ eV}$ and a fixed symmetry-breaking scale $\approx 10^{11} \text{ eV}$ [57].

Our technique could also be used to constrain bursts of axion fields produced from binary black holes [58]. Axion

bursts lag behind the emitted gravitational waves that act as triggers for detection. Assuming isotropic propagation of exotic fields, the field strength decreases with the increasing distance from its source, reducing the sensitivity to the coupling strength. Using the functional form of axions bursts in Ref. [57] as template, our sensor could achieve an energy resolution of 10^{-22} eV/Hz^{1/2}. For the prior astrophysical limits on linear coupling $\approx 10^8$ GeV and quadratic coupling 10^4 GeV [58], our sensor could have an astrophysical reach of 10^8 light years.

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