INTRODUCTION

A plethora of multiscale astrophysical and cosmological evidence suggests that roughly 85% of the matter in our universe is unlike the matter we see around us. Despite almost a century of research, the evidence for this so-called dark matter (DM) is purely gravitational. As a result, its entire particle identity including its mass, spin, and interactions with itself and with other particles remains unknown.

Various theoretical models propose an abundance of candidates that would explain the physical nature of DM. A well-motivated one is a postulated particle named the axion (1, 2), originally introduced to solve the strong CP problem (3). Over the years, many generalizations to the axion have been postulated, and they are collectively known as axion-like particles (ALPs) (4). These ALPs can be produced in the early universe and can account for the observed phenomena associated with DM. While the uncertainty for the ALP mass spans many orders of magnitudes, a particularly interesting range is that of ultralight masses (5). In this regime, the ALP De-Broglie wavelength is considerably longer than the length of the detector, and in addition, the number of particles within a single De-Broglie wavelength cubed (roughly the classical volume a single particle occupies) is much larger than 1. This implies that any interaction of ultralight ALPs with other particles such as protons, neutrons, electrons, and photons would be coherently enhanced and more easily detected (6–14). Moreover, the coupling between ultralight ALP DM with electron and nuclear spins can be manifest in the form of anomalous magnetic fields that induce an oscillatory energy shift at a characteristic frequency that depends on the ALP mass (15).

Various groups search for cosmological DM using astronomical observations and terrestrial detectors. Comagnetometers and nuclear magnetic resonance (NMR) sensors, in particular, are compact sensors that feature enhanced sensitivity to regular and anomalous magnetic fields. These sensors are composed of a dense ensemble of spin-polarized nuclei in a gaseous, liquid, or solid phase, whose collective response to regular or anomalous fields is measured by a precision magnetometer (6, 9–13, 16–25).

While these sensors have long been applied in various disciplines including medicine (26), chemistry (27, 28), geology (29), physics, and engineering (30), their application for the search of cosmological DM is only at its infancy, with potential unprecedented sensitivities (31–33). Recently, these sensors have set new terrestrial constraints on the coupling of neutrons to ALP DM with masses $m_{\text{DM}} < 4 \times 10^{-15}$ eV/c$^2$ and improve upon previous terrestrial bounds by up to 1000-fold for masses above $4 \times 10^{-13}$ eV/c$^2$. We also set bounds on pseudoscalar dark matter models with quadratic coupling.

New constraints on axion-like dark matter using a Floquet quantum detector

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Dark matter is one of the greatest mysteries in physics. It interacts via gravity and composes most of our universe, but its elementary composition is unknown. We search for nongravitational interactions of axion-like dark matter with atomic spins using a precision quantum detector. The detector is composed of spin-polarized xenon gas that can coherently interact with a background dark matter field as it traverses through the galactic dark matter halo. Conducting a 5-month-long search, we report on the first results of the Noble and Alkali Spin Detectors for Ultralight Coherent dark matter (NASDUCK) collaboration. We limit ALP-neutron interactions in the mass range of $4 \times 10^{-15}$ to $4 \times 10^{-12}$ eV/c$^2$ and improve upon previous terrestrial bounds by up to 1000-fold for masses above $4 \times 10^{-13}$ eV/c$^2$. We also set bounds on pseudoscalar dark matter models with quadratic coupling.

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for neutron-DM quadratic interactions. Last, we also cast additional model-dependent bounds on the coupling of ALPs to protons and discuss their model uncertainty.

RESULTS

Interaction of ALP DM with spins

ALPs are pseudoscalars that can couple to neutrons in the form of an oscillatory magnetic-like field. The field amplitude $b_{DM}$ is related to the DM local energy density $\rho_{DM}$, whereas its frequency $f_{DM}$ is related to the ALP DM mass $m_{DM}$. The coupling of ALPs to spins is described by the interaction Hamiltonian

$$H = \gamma_{Xe} (\cos (2\pi f_{DM} t + \phi_{DM}) b_{DM}) \gamma_{Xe}$$

where $I_{Xe}$ denotes the nuclear spin-1/2 operator of xenon and $\gamma_{Xe}$ denotes its gyromagnetic ratio. The DM field oscillates at a frequency $f_{DM} = m_{DM}c^2/v^2$ (and with a random initial phase $\phi_{DM}$), with $v$ being its velocity. The DM’s wave function is a narrow wave packet in the frequency domain with a center slightly above $m_{DM}c^2/\hbar$ and a spread (as well as the offset from $m_{DM}c^2/\hbar$) that depends on the velocity distribution of the DM, which is moving in the galaxy with respect to Earth [see (41) for further details]. According to the standard halo model, the mean and the SD of $v$ are both of order the virial velocity $v_{vir} \approx 220$ km/s (42), and as a result, the field remains coherent for a considerably long time $\hbar/(m_{DM}v_{vir})$, which corresponds to about $2 \times 10^6$ oscillations. In Eq. 1, $\langle \gamma_{Xe} \rangle$ denotes the averaging over the velocity and phase distributions [see (41) for further details], while $b_{DM}$ is the anomalous magnetic field that is defined by

$$b_{DM} = \epsilon_{N}g_{N\text{-Quad}}\sqrt{2\rho_{DM}c^2v/\gamma_{Xe}}$$

Here, $g_{N\text{-Quad}}$ is the ALP-neutron coupling coefficient and $\rho_{DM} = 0.4$ GeV/(c$^2$cm$^3$) (42). $\epsilon_{N}$ is the fractional contribution of neutrons to the nuclear spin. Because the $^{129}$Xe nucleus has a valence neutron, the model uncertainty of $\epsilon_{N}$ is relatively small, and contribution from coupling to its protons is about two orders of magnitude smaller. We adapt $\epsilon_{N} = 0.63$, corresponding to the smallest estimation in (43), using the $\Delta NLO_{GG}(394)$ model of the nuclear interactions. $b_{DM}$ is considered an anomalous magnetic field because the Hamiltonian in Eq. 1 is independent of the gyromagnetic ratio (note that $b_{DM} \propto 1/\gamma_{Xe}$), which is associated with the spin coupling of fermions to regular magnetic fields and is thus unrelated to the interaction with ALPs.

In addition to ALP models, we also cast bounds on a pseudoscalar DM model, which interacts quadratically with neutrons (13). In this model, the anomalous field is given by

$$b_{DM} = \epsilon_{N}g_{N\text{-Quad}}\frac{\sqrt{2\rho_{DM}c^2v/\gamma_{Xe}}}{m_{DM}}$$

Here, the DM oscillates at $f_{DM}^{\text{Quad}}(m_{DM}) = f_{DM}^{\text{ALP}}(2m_{DM})$, implying that bounds on quadratic-type interactions are derived in the mass range $2 \times 10^{-15}$ eV/$c^2 < m_{DM} < 2 \times 10^{-12}$ eV/$c^2$.

Experimental setup and detection mechanism

The heart of our sensor consists of spin-polarized $^{129}$Xe atoms, whose collective response to time-varying fields is detected via an in situ precision optical magnetometer made of rubidium vapor. The atoms are encapsulated in a small cubical glass cell that is maintained
at 150°C and surrounded by magnetic shields as shown in Fig. 1. We continuously polarize the Rb spins at their electronic ground state via optical pumping. The Rb polarizes, in turn, the nuclear spins of $^{129}$Xe via spin-exchange collisions along the $\hat{z}$ axis, effectively maintaining $\sim 3 \times 10^{15}$ fully spin-polarized nuclei. Further details on the experimental configuration are described in Materials and Methods and (41).

An anomalous DM field pointing in the $xy$ plane of the detector and interacting with the $^{129}$Xe nucleons would collectively tilt them off the $\hat{z}$ axis by an angle $\theta_{\text{Xe}}$ and force their precession at a frequency $f_{\text{DM}}$. In the presence of an axial magnetic field, this tilt is suppressed outside of a narrow frequency band with width $\Gamma_{\text{Xe}}$ centered around the NMR frequency $f_{\text{Xe}}$ (corresponding to the precession frequency around the axial field) [We note that, in our setup, $f_{\text{Xe}} = \gamma_{\text{Xe}} (B_{\text{ext}}^z + b_{\text{Rb}})$, where $B_{\text{ext}}^z$ is the external field in the $z$ direction and $b_{\text{Rb}}$ is the effective magnetic field induced by the Rb atoms via the spin-exchange interactions]. Thus, for $f = f_{\text{DM}}$

$$\theta_{\text{Xe}}(f = f_{\text{DM}}) = \frac{\gamma_{\text{Xe}} b_{\text{DM}}}{2 |i \Gamma_{\text{Xe}} + f_{\text{Xe}} - f_{\text{DM}}|}$$  \hspace{1cm} (4)

Here, $\gamma_{\text{Xe}} \approx -1.18 \text{ kHz/G}$ is the gyromagnetic ratio of xenon, and $\Gamma_{\text{Xe}} \approx 0.3 \text{ Hz}$ is the measured decoherence rate of the xenon spins. From Eq. 4, we thus learn that the noble-gas spins efficiently respond to the anomalous ALP field if it oscillates at a frequency $f_{\text{DM}}$ that resonates with the NMR frequency $f_{\text{Xe}}$. Our setup is capable of efficiently sensing $f_{\text{DM}}$ in the 1- to 1000-Hz range.

To measure the precession of the noble-gas spins, we use the rubidium as an optical magnetometer. Using a linearly polarized optical probe beam, we measure the collective spin of the rubidium along the $\hat{z}$ axis via its imprint on the polarization of the probe beam, which rotates after traversing the alkali medium. The polarization rotation is subsequently measured with a set of differential photodiodes in a homodyne configuration (44–46). While the polarized rubidium spins are initially oriented along the $\hat{z}$ axis, they are tilted by an angle

$$\theta_{\text{Rb}}(f_{\text{DM}}) = \frac{\gamma_{\text{Xe}} b_{\text{Xe}}}{|i \Gamma_{\text{Rb}} + f_{\text{Rb}} - f_{\text{DM}}|}$$  \hspace{1cm} (5)

Here, $b_{\text{Xe}}\theta_{\text{Xe}}$ is the transverse spin-exchange field. $b_{\text{Xe}}$ is proportional to the magnetic field produced by the spin-polarized xenon atoms and is enhanced by a large factor of $k_0 = 518$ owing to the Fermi contact interaction with the rubidium (47). We stress that this factor is a virtue of having an in situ magnetometer and allows for an improved sensitivity of the detector. $f_{\text{Rb}}$ is the electron paramagnetic

![Fig. 2. Constraints on ALP-neutron couplings.](https://www.science.org) In this work, we derive the constraints using the Floquet quantum detector by the NASUCK collaboration as a function of the ALP mass. All presented constraints calculated in this paper correspond to 95% C.L. of the bound. Because of the finite resolution of the figure and given the dense set of measurements, the limits appear as a bright blue band. The width of this band denotes the strongest and weakest values around each mass point. The precise tabulated bounds can be found in (48). The bright blue solid line shows a binned average of the bound, while its 1σ variation is shown in dark blue band (both calculated in log space at a binning resolution of 1% of the mass). The light transparent blue region shows the exclusion region for the ALP-neutron couplings. The dashed dark blue line shows the projected sensitivity of this experiment, as discussed in the text. The olive green regions show other terrestrial constraints, including the CASPEr ZULF experiments (12, 49), $^{3}$He comagnetometer bounds (6, 18, 50), and long-range constraints on ALP-neutron (18) couplings. In beige, the agreed astrophysical excluded region from solar ALPs unobserved in the SNO (51) and from NS cooling (52, 66–68) is shown. The region above the gray dashed line is excluded by SN cooling considerations and neutrino flux measurements (4, 52, 54). The SN cooling constraint strongly relies on the unknown collapse mechanism, and hence, the limits should be taken with a grain of salt (55).

![Fig. 3. Constraints on neutron-DM couplings of quadratic type.](https://www.science.org) The quadratic constraints (13) are derived as a function of the ALP mass, using the Floquet quantum detector by the NASUCK collaboration in this work. All presented constraints calculated in this paper correspond to 95% C.L. of the bound. As in Fig. 2, because of the finite resolution of the figure and given the dense set of measurements, the limits appear as a bright blue band. The width of this band denotes the strongest and weakest values around each mass point. The precise tabulated bounds can be found in (48). The bright blue solid line shows a binned average of the bound, while its 1σ variation is shown in dark blue band (both calculated in log space at a binning resolution of 1% of the mass). The light transparent blue region shows the exclusion region for this model. The dashed dark blue line shows the projected sensitivity of this experiment, as discussed in the text. The olive green regions show other terrestrial constraints, including the CASPEr ZULF experiments (12, 49) and the recasted $^{3}$He comagnetometer bounds (6, 18, 50). The region above the dashed gray line is excluded by SN cooling considerations (13). We stress that the SN cooling constraint strongly relies on the unknown collapse mechanism, and hence, the limits should be taken with a grain of salt (55).
We search for anomalous magnetic fields oscillating in the range of $f_{\text{DM}} = 1 \text{ to } 1000 \text{ Hz}$ (corresponding to $m_{\text{DM}}$ in the range of $4 \times 10^{-15}$ to $4 \times 10^{-12} \text{ eV/c}^2$). To maximize the realized sensitivity, for each searched frequency, we tuned the axial magnetic field to bring the NMR frequency $f_{\text{Xe}}(B_{\text{ext}}^z)$ near resonance with $f_{\text{DM}}$ and typically recorded the sensor response for $(2 - 20) \times 10^5$ oscillations. To cover all frequencies within the search range, we scanned the magnetic field in fine steps of about 0.2 mG to maintain overlap in the NMR frequencies of neighboring measurements. We have scanned the entire range of frequencies several times by slightly shifting the NMR frequencies to get ample measurements of any given frequency at different sensitivities. As a result, the search consisted of almost 3000 measurements, taken during a period of 5 months. To characterize the sensor response and validate its stability during the long search period, each measurement was preceded and followed by a set of calibration measurements. The calibration measurements automatically tuned the parameters of the oscillatory Floquet field and characterized the response function of the sensor. Data processing is described in Materials and Methods, and detailed search and calibration protocols are given in the Supplementary Materials (41).

**Detection capability**

As DM ALP signals feature extremely long coherence time compared to the sensor backgrounds, they can potentially be directly detected and distinguished from the background. In the frequency domain, a signal centered at $f_{\text{DM}}$ would have an ultranarrow bandwidth with a quality factor of about $\sim 2 \times 10^7$, thus being distinguishable from the noise that is approximately white within that bandwidth [see (41)]. Sideband analysis further enables to differentiate between the signal and background. We use measurements at magnetic fields in which the NMR frequency is off-resonant and the sensitivity to anomalous fields is negligible as control measurements. These measurements enable us to identify frequencies in which the background has coherent properties that require specialized analysis procedures that rely on the different responses instead of the coherence times of signal and background. Last, multiple repetitions of measurements enable to exclude transient noise and differentiate it from the coherent ALP signal.

**Search results**

We use the log-likelihood ratio test to constrain the presence of ALP DM with 95% confidence level (C.L.) bounds, presented on
**DISCUSSION**

In summary, we presented new constraints on the ALP DM couplings to neutrons, substantially improving previous bounds. Our detector used dense xenon spins and in situ Floquet magnetometer, which enabled the extension of ALP masses to higher values. In addition, new bounds on neutron-quadratic type interactions were also cast.

NMR detectors are frontier technology to search for new physics, which, for ALPs, can potentially reach the limits of QCD axions in the standard quantum limit (33). Practically, however, these sensors are often limited by the magnetic noise floor and the realized detector sensitivity that nonetheless are expected to exceed astrophysical bounds for coupling with neutron spins. In addition, similar searches using nuclei whose proton spin component is known with higher certainty could extend such searches and cast reliable bounds on the coupling of ALPs with protons.

**Note added**

Toward the end of our search and during the late stages of the analysis of the recorded data, we became aware of the study of Jiang et al. (60),...
which uses an NMR detector to search for nucleon-ALP coupling. Jiang et al.’s (60) study finds similar sensitivity, but it does not use Floquet, it does not account for $\varepsilon_N$, and it does not account for the stochastic effects of the ALPs. In addition, Gramolin et al. (61) have recently independently shown how to account for the stochastic properties of the ALPs, in a method that is similar to our detailed analysis in (41).

**MATERIALS AND METHODS**

**The Floquet bandwidth enhancement**

The Floquet field $B_F \cos(2\pi f_F t) \hat{z}$ is aligned with the previously described axial field, $B_F^{\text{ext}}$. The strong field modulates the ground-state energy of the rubidium spins, thereby dressing their energy levels with the field induced by the radio frequency (RF) photons (62). This modulation is spectrally manifested as a series of resonance bands that appear at discrete harmonics of the driving field in the magnetic spectrum of the rubidium, and via multiphoton processes, it encodes the response of the spins to low-frequency fields. The resulting tilt of the measured rubidium is thus obtained from Eq. 5 by shifting the spectrum by $nf_F$ with an integer $n$ and multiplying the response with a bandwidth enhancement factor $\eta_F^{(n)}$ such that around the harmonics of the Floquet frequency $\theta_F^{\text{Floquet}}(f = f_{DM} + nf_F) = \eta_F^{(n)} \theta_F(f = f_{DM})$.

In Fig. 4A, we exemplify the Floquet spectrum via typical measurement of the spectrum, $\theta_{Rb}(f)$, in response to a low-frequency transverse magnetic noise, whose variance is white up to a cutoff frequency set at $f_c = 10$ kHz (light green shade), while setting $f_{Rb} = 105$ kHz. In the absence of the Floquet field, the rubidium response appears at the same frequencies of the drive (green line), and the response is suppressed because the driving field is tuned away from resonance $|f_{Rb} - f_c| \approx 14\Gamma_{Rb}$. By applying a strong and resonant Floquet field $(f_F = f_{Rb}, B_F = 0.4 \text{G})$, the low-frequency spectrum is mapped to discrete harmonics of the Floquet field at integer multiples $n$ for which efficient coupling is realized. We observe an $\eta_F^{(1)} = 5.9$-fold enhancement of the magnetic response near the first Floquet band with respect to the low-frequency response of the unmodulated sensor. Notably, routine calibrations of the Floquet parameters during the search did not inject magnetic noise but used coherent sinusoidal signals, following the protocol that is described in detail in (41).

The resonant nature of the Floquet modulation is encoded in $\eta_F^{(n)}$, which depends on the external magnetic field and the Floquet amplitude, $B_F$. In Fig. 4B, we show the enhancement, $\eta_F \equiv \eta_F^{(1)}$, as a function of the axial magnetic field. At each measured value, we optimized for the frequency and amplitude of the Floquet field, following the theoretical analysis detailed in (41).

**Background and signal sensitivity**

The sensor is susceptible not only to the anomalous fields but also to various sources of noise that limit its detection sensitivity. In this section, we present and characterize the noise model for the detector, whereas the protocol that routinely monitored and calibrated for variations of the model parameters during the search is described in (41). The dominant sources of noise are the magnetic field noise, $\delta B$, and optical polarization noise due to the shot noise of the probe beam $W$. In the presence of the former, Eqs. 4 and 5 are modified by taking $b_{DM} \rightarrow b_{DM} + \delta B$ and $b_{Xe} \theta_{Xe} \rightarrow b_{Xe} \theta_{Xe} + \delta B$. To study the response of the system to the above noise, we combine Eqs. 4 and 5 and rewrite the Floquet-demodulated output of the optical magnetometer $S + N$, decomposed to the noise contribution

$$N(f) = \xi_1 \frac{\delta B(f)}{1 + i(f_{Xe} - f)/\Gamma_{Xe}} + \delta B(f) + W(f)$$

and the coherent signal contribution of the ALP at $f = f_{DM}$

$$S(f) = \xi_2 \frac{b_{DM}}{1 + i(f_{Xe} - f)/\Gamma_{Xe}}$$

The detector reading, $S + N$, is measured around the first Floquet band and is given here in units of magnetic field. The calibration protocol of the magnetometer, which determines the proportionality constant that converts the measured optical signal to magnetic field units, is described in section S3 in (41). Here, $W$ denotes the optical probe noise obtained after demodulation. $\xi = \gamma_{Xe} b_{Xe} / 2 \Gamma_{Xe}$ is the overall dimensionless factor that encodes the enhancement of the rubidium response to the xenon precession over the direct impact of magnetic noise. This is in line with the view of the xenon precession as the signal (also affected by regular and anomalous magnetic fields) measured via the optical magnetometer. $\xi$ is calibrated routinely, and during the entire search, its value ranged from 1 to 3 (see (41) for further details). For $\xi \gg 1$ and $|\delta B | \gg | W |$, the experimental setup reaches a maximal sensitivity to the ALP couplings ($\xi_{NN}$) and is limited solely by the magnetic field noise and the gyro-magnetic ratio of the nuclei. In the absence of a noise-cancellation mechanism, this limit is universal to all types of NMR sensors, independent of the number of polarized nuclear spins, the performance of the used magnetometer, or the coherence times of the atoms.

To exemplify the noise characteristics of the detector, in Fig. 5, we present the square root of the spectral density of a noise measurement for a single recording at $B_F^{\text{ext}} = 0.1$ G. In this measurement, no coherent calibration signals (other than the Floquet drive) are present, and the spectral density of the recorded noise realization PSD($N$) is calculated using Welch’s method. The noise spectrum has a typical square root of spectral density of $100 \text{fT} / \sqrt{\text{Hz}}$ (dark blue shaded region) and is dominated by noise of the probe beam $W$, which we show in (41) to be governed by photon shot noise. The estimated contribution of the magnetic noise is $\approx 10 \text{fT} / \sqrt{\text{Hz}}$, generated by the inner layer of the magnetic shield (63) and sensed by the rubidium (light blue region) and xenon (blue region) spins. We find that only at a few frequencies (and, in particular, at harmonics of the mains hum) the magnetic noise becomes dominant over the probe noise.

To characterize the response transverse fields, we inject white magnetic noise along the $\hat{x}$ direction with a spectral density of $5.6 \text{pT} / \sqrt{\text{Hz}}$ as presented in Fig. 5B. To highlight the response of the xenon nuclei, we tune the Floquet parameters of the rubidium vector magnetometer to be most sensitive to fields along the $\hat{y}$ direction, suppressing the response to $\hat{x}$ fields. Thus, we gain sensitivity to the response of the xenon spins to the injected noise while suppressing its direct effect on the rubidium magnetometer. The Lorentzian response of the xenon near its NMR frequency validates Eq. 7 and enables also to estimate its parameters, yielding $\xi = 2.2$. The injection of magnetic noise to characterize the detector was used only for the particular demonstration in Fig. 5B to highlight the coupling of magnetic noise and its decomposition to contributions of Rb and Xe. The routine calibration protocols that calibrate $\xi$ use coherent sinusoidal signals instead as described in (41).
The detailed and strict search procedure and the methodology of the data analysis are described in (41), taking into account the exact measurements of the signal and the statistical properties of the ALPs. Nonetheless, we find it insightful to exemplify the spectral sensitivity to anomalous fields of the detector based on a short measurement of the sensor in the limit of $v_{\text{rel}} \to 0$ as shown in Fig. 5C. It is apparent that, when white noise dominates the spectrum, the sensitivity follows the Lorentzian NMR shape of the nuclear spins, and a high sensitivity is limited to a narrow frequency band, whose width is determined by the xenon's spectral width $\Gamma_{\text{Xe}} = 0.3$ Hz.

It is also possible to give an approximate estimate of the excluded $b_{\text{DM}}$ and the typical $\theta_X$ from the measurement in Fig. 5A at $f_{\text{DM}} \approx 116$ Hz in resonance with the NMR frequency. For the characteristic noise spectral density $\sqrt{\text{PSD}(N)} \approx 100 \text{fT}/\sqrt{\text{Hz}}$, a finite measurement interval $T \approx 3000$ s, which is shorter than the ALP coherence time, and $\xi = 2.2$, we can exclude coherent anomalous fields that are larger than $b_{\text{DM}} \gtrsim \sqrt{\text{PSD}(N)/(\xi T)} \approx 1$ fT. This field also corresponds to a minimal tilt of the Xe spins by about $\theta_X \gtrsim 2 \times 10^{-8}$, obtained by Eq. 4 for $\Gamma_{\text{Xe}} = 0.3$ Hz. Note that these approximate estimates compared the power of the signal to the noise variance to determine exclusion [signal-to-noise ratio (SNR) = 1], whereas the actual constraints presented in Fig. 2 effectively set higher SNR to obtain the 95% C.L. in the likelihood test.

**Data processing**

We automatically excluded data from our analysis if it matched at least one of the following criteria (quality cuts): (i) substantial variation in the sensor parameters between two subsequent calibration measurements, (ii) saturation of the detector that is identified via substantial decrease in the noise variance, and (iii) a substantial increase in the noise, which is identified in spectral regions of the signal being far away from the NMR resonance. For (i), we vetoed the entire measurement, whereas for cuts related to both (ii) and (iii) data vetoing is limited to a finite measurement window.

We analyze the data using the log-likelihood ratio test to constrain the presence of ALPs at frequency $f_{\text{DM}}$ with a width determined by the signal coherence time and the effects of Earth's rotation on the signal. Each measurement was used to constrain a frequency range of width 2 to 6 Hz around the resonance. Bounds were set on ALP-neutron interactions, as well as quadratic interactions with neutrons. To accurately account for the velocity distribution of the DM (see Eq. 1), we followed the suggested analysis in (19). We used the asymptotic formulas in (64) for the distribution of the log likelihood. For each measurement, the noise was assumed to be white and was estimated using sideband analysis in the frequency domain away from the NMR resonance.

All analysis procedures and cuts were designed in a blinded fashion, and decided in advance before looking at the data, to eliminate bias. However, after unblinding, we found that less than 0.1% of the spectral domain of the search range was statistically inconsistent with the white noise model. For this part, we have refined the statistical tests to treat transient and coherent magnetic noise. All data are found consistent with the refined model. The ALP stochastic properties, the statistical analyses, and post-unblinding changes are detailed in (41).
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