Perspective

Quantum Science and the Search for Axion Dark Matter

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The dark-matter puzzle is one of the most important open problems in modern physics. The ultralight axion is a well-motivated dark-matter candidate, conceived to resolve the strong-*CP* problem of quantum chromodynamics. Numerous precision experiments are searching for the three nongravitational interactions of axionlike dark matter. Some of the searches are approaching fundamental quantum limits on their sensitivity. This Perspective describes several approaches that use quantum engineering to circumvent these limits. Squeezing and single-photon counting can enhance searches for the axion-photon interaction. Optimization of quantum spin-ensemble properties is needed to realize the full potential of spin-based searches for the electric dipole moment and the gradient interactions of axion dark matter. Several metrological and sensing techniques, developed in the field of quantum information science, are finding natural applications in this area of experimental fundamental physics.

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I. DARK MATTER—THE SIGNATURE OF PHYSICS BEYOND THE STANDARD MODEL

Only six parameters are needed to fit all experimental data gathered on the behavior of our Universe at cosmological scales [1]. The resulting model is rich, yet strange. It assumes that general relativity and the standard model of particle physics describe the basic physics of the Universe throughout its history. Yet, at present time, only approximately 5% of the energy density of the Universe is in the form of nucleons, electrons, photons, or neutrinos—the particles that are described by the standard model [2]. Most of the energy in the Universe appears to be in the form of *dark energy*, which drives the accelerating expansion of the Universe. The best-studied possibility is that this acceleration is due to the Einstein cosmological constant Λ, equivalent to the vacuum energy of empty space [3,4]. Most of the matter in the Universe is in the form of *dark matter*, which is nonbaryonic and has feeble interactions (or none at all) with the standard-model particles, aside from gravity. These observations are the basis of the lambda cold dark matter (ΛCDM) model.

The early evidence for dark matter long predates the ΛCDM model. A detailed account of the long history of the dark-matter concept can be found in Ref. [5]. Today, evidence for dark matter is based on numerous observations, including galaxy-rotation curves, baryonic acoustic oscillations, and temperature anisotropies of the cosmic microwave background [6,7]. The fact that none of the standard-model particles can explain these observations is an indication that dark matter represents evidence for physics beyond the standard model. Since it has only been observed through its gravitational effects, there is a wide range of dark-matter candidates, spanning 90 orders of magnitude in mass [8,9].

The weakly interacting massive particle (WIMP) is a well-known candidate that has inspired a large number of ultrasensitive experiments of increasing complexity and scale [10,11]. To date, there has been no unambiguous detection and fundamental backgrounds (the neutrino floor) will soon start to limit the sensitivity of direct WIMP searches [2,12–14]. Additionally, recent experiments at the LHC, as well as laboratory-scale experiments sensitive to permanent electric dipole moments (EDMs) of atoms and molecules, have placed stringent constraints on theoretical frameworks, such as supersymmetry, that support WIMP dark matter [15–18]. Nevertheless, the WIMP concept can be expanded and broadly defined WIMPs remain viable dark-matter candidates [8].

Axions and axionlike particles are in the class of ultralight dark-matter candidates that has attracted increasing attention in the past decade [19]. Quantum

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chromodynamics (QCD) axions are considered to have the best theoretical motivation, because they solve the strong-*CP* problem [20]. This problem is briefly formulated as follows. Strong interactions, described by QCD, are allowed to violate the combined charge-conjugation C and spatial inversion (parity) P discrete symmetries. Such violation is quantified by the θ parameter that enters the QCD Lagrangian and would give rise to, e.g., a permanent EDM of the neutron [21]. Yet, stringent experimental limits on the neutron EDM and other *CP*-violating strong interactions indicate that this violation is a factor of approximately 10^{10} smaller than expected [22,23]. This constitutes the strong-*CP* problem. The Peccei-Quinn mechanism solves this problem, by introducing a new scalar field that changes the QCD Lagrangian so that instead of θ being a constant, it takes on a dynamical effective value that is driven to zero due to spontaneous symmetry breaking [24,25]. The elementary excitation of this field corresponds to the QCD axion, the pseudo-Goldstone boson of the Peccei-Quinn symmetry [26,27].

Since its inception, the axion concept has had a deep impact on theoretical physics, emerging naturally from grand unified theories, models with extra dimensions, and string theory [28]. A plenitude of light pseudoscalar bosons (*axionlike particles, ALPs*) arise readily in string-theory compactifications, as a result of symmetries broken at large energy scales f_a [28–30]. Generic expectations from string theory predict large values for f_a, up to the grand unified (approximately 10^{16} GeV) and Planck (approximately 10^{19} GeV) scales [29,31–34].

There are many other dark-matter candidates, including relaxions and other scalar fields [35], hidden photons [36], superfluid dark matter [37], Planck-scale dark matter [38], sterile neutrinos [39], and primordial black holes [40,41]. Arguments can be made about the degree of theoretical motivation of each specific candidate but the lack of unambiguous experimental evidence of nongravitational dark-matter interactions signifies the importance of keeping the dark-matter search as broad as possible [8,42,43].

Dark matter is not the only way to resolve puzzles in astrophysical observations, such as galactic rotation curves. Other potential solutions modify the theory of gravitational interactions and dynamics [44,45]. However, these modified gravity theories suffer from several problems and most scientists in this field pursue the cold-dark-matter paradigm [46].

II. THE ROLE OF QUANTUM SCIENCE IN FUNDAMENTAL DISCOVERY

The idea of trying to detect nongravitational interactions of dark matter in a laboratory is many decades old [5]. Yet, despite numerous sensitive experimental searches of increasing complexity and scale, there has been no unambiguous detection. One way to guide our efforts toward a potential discovery is to consider recent technological advances that may allow experiments to probe previously unexplored territory. Quantum information science (QIS) is at the forefront of technological innovation [47]. Originating at the foundations of quantum mechanics, the field of quantum science was stimulated by its convergence with information science, when it was realized that quantum mechanical machines can perform certain information-processing tasks faster than any classical computer. It is still unknown if and how truly large-scale quantum machines can be built, beyond the noisy intermediate-scale quantum (NISQ) devices [48]. But the concepts and tools of QIS have already inspired a broad spectrum of novel applications, in fields ranging from the life sciences to materials science [49,50]. Quantum metrology and quantum sensing are already making a significant impact on fundamental physics [51].

A. What is “quantum”?

Do we need to define what is meant by “quantum,” in order to consider how quantum science can be used to search for dark matter? After all, it is the scientific reach of a particular technology that ultimately determines its promise, rather than an arguably arbitrary attribute of “quantumness” [52]. Is a photomultiplier a quantum sensor? It counts photons, quanta of electromagnetic field, with sensitivity beyond the standard quantum limit of a linear amplifier measuring the electromagnetic field. Is a superconducting quantum interference device (SQUID) a quantum sensor? It makes use of the Josephson effect—the quantum interference of the superconducting wave function, split into two paths, interrupted by Josephson junctions. And it has the word “quantum” in the acronym. Answers to such questions are matters of perspective but, inevitably, they do guide the scope of this Perspective. However, it is important to keep in mind that the ultimate focus is on the potential sensitivity improvements, the feasibility, and the scientific merit of an approach.

Let us loosely define the “first quantum revolution,” which gave birth to “quantum 1.0” technologies, based on lasers, semiconductor, and superconductor devices. The promise of the “second quantum revolution” is the development of “quantum 2.0” technologies, with the potential to result in impressive performance leaps. For example, a number of approaches aim to evade the standard quantum limit (SQL) of measurement, by making use of entanglement, squeezing, back-action evasion, or strong correlations. More broadly, QIS has developed many ideas for how to optimize preparation, transmission, control, and measurement of correlated quantum states in systems such as spin ensembles, atomic interferometers and clocks, color centers, and superconducting devices.

B. The pioneering efforts

The search for gravitational waves by the Laser Interferometer Gravitational-Wave Observatory (LIGO) is the pioneering example of how "quantum 2.0" technologies can make a direct impact on fundamental physics discoveries. Injection of squeezed light into the interferometer improves the photon-shot-noise limit to the sensitivity of the Advanced LIGO detectors [53]. At frequencies above 50 Hz, the sensitivity improvement is up to 3 dB, which corresponds to a 40–50% increase in the expected gravitational-wave-event detection rate [54]. At lower frequencies, the sensitivity is degraded by quantum back action, as radiation pressure induces motion of interferometer mirrors. However, frequency-dependent squeezing can achieve a broadband reduction of quantum noise [55,56]. These quantum technologies are crucial to the scientific reach of the gravitational-wave-observing programs [57]. Indeed, there is an intriguing possibility that merger events observed by terrestrial gravitational-wave detectors are due to primordial black holes, which could contribute a significant fraction, if not the entirety, of the dark-matter abundance [40,41,58,59].

The pioneering efforts of incorporating quantum technologies into gravitational-wave detectors have been a key inspiration for the rapid growth of research activity in the field of quantum sensing and metrology [51]. There are also numerous potential applications for quantum sensors, in fields ranging from chemistry and biology to medicine and geology [60]. Even within fundamental physics, there are many diverse ideas for how quantum technology can be applied. One example is the development of high-efficiency pattern recognition algorithms, based on quantum annealing, in order to speed up track-reconstruction analysis and jet identification in subatomic collider experiments [61–63]. Another area of opportunity is the application of quantum computational resources to simulate high-energy quantum field theories [64], perform nuclear structure calculations [65], and model neutrino-nucleus scattering [66].

This Perspective focuses on a specific area of intense search for new fundamental physics: the use of quantum technologies to accelerate direct searches for ultralight axionlike dark matter. Many of the ongoing and proposed experiments in this field are approaching, or have already reached, sensitivity levels where quantum resources are needed to achieve their scientific goals. This is also where several of the quantum approaches, having matured in QIS, can find a natural application. Thus there is both a need and an opportunity to use quantum science and engineering. Notably, there are other avenues for sensitivity improvements that are also being pursued, such as development of high-magnetic-field technologies [67] and high-quality-factor resonators [68–71].

III. NONGRAVITATIONAL INTERACTIONS OF AXIONLIKE DARK MATTER

The Big Bang cosmology of the axion depends on whether the spontaneous breaking of the Peccei-Quinn symmetry occurs before, during, or after inflation [72–75]. Axions can be produced in the early Universe nonthermally via the misalignment mechanism [76–78] and via thermal production through axion couplings to the standard-model plasma [79,80]. The misalignment mechanism dominates for large values of the Peccei-Quinn symmetry-breaking energy scale, generating a coherent oscillating axion field $a \approx a_0 \cos(2\pi \nu_a t)$. The oscillation frequency corresponds to the axion Compton frequency $\nu_a = m_a c^2/h$, where m_a is the axion mass and h is Planck's constant. The field amplitude determines the stored energy density. If we assume that this axion field is the primary component of dark matter, then its amplitude on Earth can be calculated from the local galactic dark-matter energy density: $m_a^2 a_0^2/2 = \rho_{\rm DM} \approx 4 \times 10^{-42}$ GeV^4 [9,81]. The kinetic energy of the axionlike-dark-matter field introduces small corrections to its frequency spectrum. The standard halo model predicts the spectral shape with line width $(v_0^2/c^2)\nu_a \approx 10^{-6}\nu_a$, where $v_0 \approx 220$ km/s is the circular rotation speed of the Milky Way galaxy at the location of the Sun [82–84].

Symmetry restricts the interactions that axions and ALPs can have with the particles of the standard model (such as electrons, photons, and nuclei) [19,81]. There are three possible interactions. The defining axion interaction that solves the strong-*CP* problem is the coupling to the gluon field [85]. At low energies, this leads to the nucleon electric dipole moment (EDM) interaction

$$\mathcal{H}_{\rm EDM} = g_d a \mathbf{E}^* \cdot \boldsymbol{\sigma}/\sigma, \tag{1}$$

where g_d is the coupling strength, σ is nucleon spin, and $\mathbf{E}^*$ is an effective electric field that depends on the nuclear, atomic, and condensed-matter environment of the nucleon [86]. This corresponds to a parity- and time-reversal-violating nucleon EDM, given by $d = g_d a$. Since the axion field oscillates at the Compton frequency ν_a, this is not a constant but an oscillating EDM. Recalling the strong-*CP* problem, we note that this corresponds to an oscillating QCD θ parameter: $\theta(t) = a(t)/f_a$, with g_d inversely proportional to f_a [81,87]. The EDM coupling generates axion mass and for the QCD axion $m_a \approx \Lambda_{\rm QCD}^2/f_a$, where $\Lambda_{\rm QCD} \approx 200$ MeV is the QCD confinement scale [81,88].

Axions and ALPs can couple to standard-model fermions, such as nucleons or electrons, via the gradient interaction with Hamiltonian

$$\mathcal{H}_{\rm gr} = g_{\rm gr} \nabla a \cdot \boldsymbol{\sigma}, \tag{2}$$

where $g_{\rm gr}$ is the coupling strength and σ is the fermion spin.

The final interaction is with electromagnetic fields, commonly written in terms of the Lagrangian

$$\mathcal{L}_{a\gamma\gamma} = g_{a\gamma\gamma} a\mathbf{E} \cdot \mathbf{B}, \tag{3}$$

where $g_{a\gamma\gamma}$ is the coupling strength and $\mathbf{E}$ and $\mathbf{B}$ are the electric and magnetic fields.

Generically, axions and ALPs can possess all these couplings. Direct experimental searches for dark matter can be thought of as transducers, the goal of which is to detect the effects of an interaction of a specific dark-matter candidate with the experimental apparatus by converting them into an electromagnetic signal that is calibrated to give some information about this interaction. We consider separately the experiments searching for the electromagnetic interaction of axionlike dark matter with photons [see Eq. (3)] and interaction of axionlike dark matter with spins [see Eqs. (1) and (2)].

The possible mass range of axionlike dark matter is extremely broad. An approximate estimate of the mass range where the existence of axionlike particles is theoretically and experimentally allowed is 10^{-19} eV to 10^{-1} eV [2]. Focusing on the QCD axion, this window narrows somewhat, to 10^{-12} eV–10^{-1} eV [19]. Only a tiny fraction of this window has been explored in laboratory experiments. The ADMX experiment has searched for the electromagnetic interaction of axion dark matter in the mass range 2.66–3.31 μeV [89,90] and there is a program for extending the upper range to 16.5 μeV. There are many other ideas for how to cover a much broader swath of axion masses [91]. Approaches based on quantum engineering are an important part of these efforts (see Fig. 1).

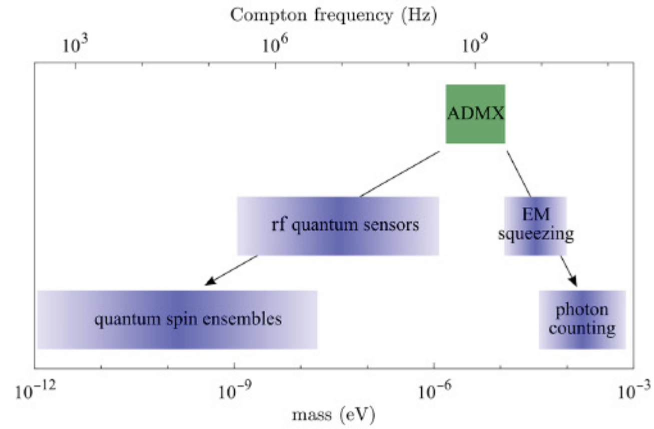

FIG. 1. The potential QCD axion dark-matter mass range (bottom scale) and the corresponding Compton frequencies (top scale). The ADMX experiment aims to cover the mass range between 2.5 and 8.3 μeV and future modifications will seek to double the upper end to 16.5 μeV [89,90,92]. This is only a small fraction of the possible mass range. Approaches based on quantum engineering are an important part of the efforts to broaden the search. Squeezing of quantum fluctuations of the electromagnetic field and photon counting will extend the search at higher masses and radio-frequency quantum sensors will enable lumped-element-based experiments at lower masses. Still lower masses will be explored by experiments that make use of quantum spin ensembles.

IV. QUANTUM-ENHANCED SENSORS OF ELECTROMAGNETIC FIELDS

Searches for the electromagnetic interaction of axionlike dark matter are based on the idea that the dark-matter field can excite an electromagnetic field sensor, such as a microwave cavity, an optical resonator, or a lumped-element circuit coupled to a sensitive detector [93]. This excitation takes place via the interaction in Eq. (3). Microwave-cavity-based haloscopes search for conversion of the axionlike-dark-matter field into photons within a high-quality-factor resonant cavity, placed inside a strong magnetic field (see Fig. 2). The ADMX experiment is the most mature search of this type. It has achieved a level of sensitivity sufficient to search for dark-matter axions with masses between 2.66 and 3.31 μeV and, under the assumption that all of dark matter is in the QCD axion field, it has excluded the QCD axion-photon couplings predicted by plausible models for this mass range [89,90]. A number of cavity-based axion-dark-matter searches are in development or already exploring ALP masses up to approximately 50 μeV [23,94–102]. In order to search for lower-mass axions and ALPs coupled to photons, it is possible to use lumped-element circuits [103–110]. This concept is based on a modification of Maxwell's equations by the $g_{a\gamma\gamma} a\mathbf{E} \cdot \mathbf{B}$ interaction: in the presence of a large static magnetic field B_0, axionlike dark matter acts as a source of an oscillating magnetic field, the amplitude of which is proportional to B_0 [93,111].

A. Electromagnetic squeezing

For a microwave-cavity haloscope, the estimate for the axion-induced power that has to be detected at the Compton frequency is approximately 10^{-24} W [112]. Experiments have to be designed to operate at ultralow temperatures, to minimize the impact of thermal noise. Zero-point quantum fluctuations of the electromagnetic field are a fundamental source of noise at the level of $h\nu/2$, where h is Planck's constant and ν is the frequency of the electromagnetic field mode. This noise limits the reach of microwave-cavity haloscopes at frequencies above a few gigahertz. Vacuum squeezing can circumvent this limit and speed up the frequency sweep rate of a resonant search [113]. This has been implemented by the HAYSTAC collaboration, which has demonstrated a doubling of their search rate [94,100]. The HAYSTAC approach can be understood using a noise model with two main contributions: N_1 and N_2. N_1 is the noise sourced by the internal cavity loss, as a consequence of the fluctuation-dissipation

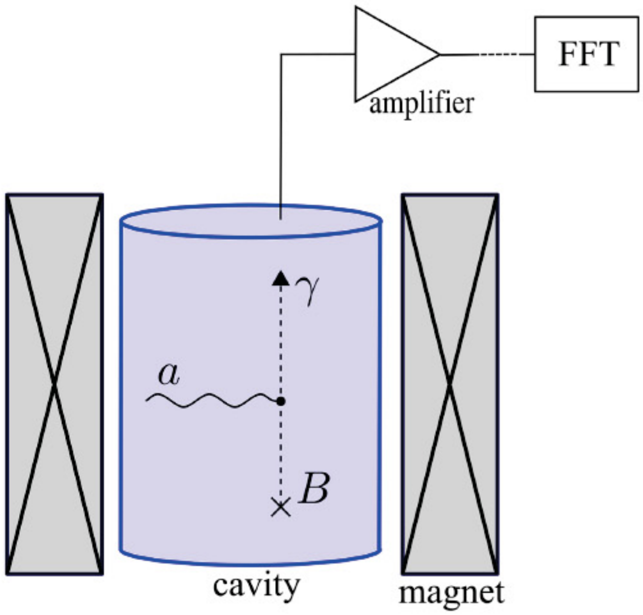

FIG. 2. A conceptual schematic of the ADMX haloscope. A resonant cavity is placed in a strong magnetic field created by a superconducting magnet inside a dilution refrigerator. Dark-matter axions are converted to photons via the electromagnetic interaction in Eq. (3). The resulting electromagnetic signal occupies a narrow band near the axion Compton frequency ν_a. It is coupled out of the cavity and into a sensitive amplifier and detection chain. The search for the unknown axion mass is performed by tuning the cavity resonance.

theorem. Its spectral shape is Lorentzian, with a spectral width determined by the cavity quality factor. N_2 is the noise incident on, and reflected from, the cavity. This is Johnson-Nyquist noise, originating from a 50-Ω termination, held at a cryostat base temperature [see Fig. 3(a)] [114]. In the limit where these two contributions dominate over the noise of the subsequent amplifier chain, the width of the band of potential axion masses that are simultaneously probed for a given cavity-tuning step is determined by the range of frequencies where N_1 dominates over N_2 [see Fig. 3(b)]. In HAYSTAC, the cavity output port is coupled to two Josephson parametric amplifiers (JPAs) via a nonreciprocal element (circulator). The noise N_2 is coupled into the cavity via one of the circulator ports. The first JPA (labeled "squeezer") squeezes one of the quadratures of this vacuum state before it enters the cavity, where it can be displaced by the interaction with the axion-dark-matter field. The second JPA (labeled "amplifier") unsqueezes the state after it exits the cavity, amplifying the previously squeezed quadrature. Squeezing does not affect the ratio of the axion-induced signal S to the intrinsic cavity noise N_1, which dominates near the cavity resonance. But it does reduce the contribution from the noise N_2, which makes it beneficial to overcouple the cavity and broaden the frequency band over which N_1 dominates over N_2 and therefore the band of potential axion masses that are simultaneously probed for a given cavity-tuning step [see Fig. 3(c)]. This allows larger tuning steps and speeds up the search at a given axion coupling sensitivity [113]. The HAYSTAC collaboration search for the electromagnetic interaction of axion dark matter has placed stringent limits on axions with mass near 17 μeV [100].

A closely related method that has been proposed to speed up an axion search scan is to couple the axion-sensitive cavity to an auxiliary resonant circuit via two-mode squeezing and state-swapping interactions [115]. We should note that both this approach and single-mode squeezing can enhance the scan rate at lower frequencies, where the axion-sensitive cavity has an appreciable thermal photon population [113,116]. Thus they are promising experimental directions in this regime. At higher frequencies, however, the technique of photon counting can completely evade the standard quantum limit applicable to linear measurements of electromagnetic fields.

B. Photon counting

Photon counting surpasses the standard quantum limit by measuring only the amplitude and not the phase of the electromagnetic field. For cavity-based experiments searching for the electromagnetic interaction of axion dark matter, this means replacing the linear amplifier and detection chain in Fig. 2 with a photon detector. The detector dark-count rate becomes the key figure of merit, as the Poisson fluctuations in the background count rate limit the experimental sensitivity. At optical and infrared frequencies, photomultipliers, avalanche photodiodes, and superconducting nanowire single-photon detectors (SNSPDs) are well-established technologies [117,118]. For example, SNSPDs have recently been used to place limits on dark photon dark matter in the electronvolt mass range [119]. However, such sensors are not suited to detecting lower-energy microwave quanta.

One possibility for detecting microwave photons is to use highly excited Rydberg states in alkali atoms [120,121]. This approach has recently seen renewed interest [122]. Another approach, spurred by advances in superconducting device technology, has led to the development of single-microwave-photon detectors in which input photons are coupled to a superconducting qubit, the state of which is measured to detect, and even count, the incoming photons [123–125]. Such devices have already found applications for magnetic resonance spectroscopy of small ensembles of electron spins [126]. The use of microwave-photon counting for an ultralight-dark-matter search has been explored in recent work that has implemented quantum nondemolition measurements of cavity photons [127]. A cavity photon shifts the transition frequency of the transmon qubit due to the qubit-cavity interaction. The

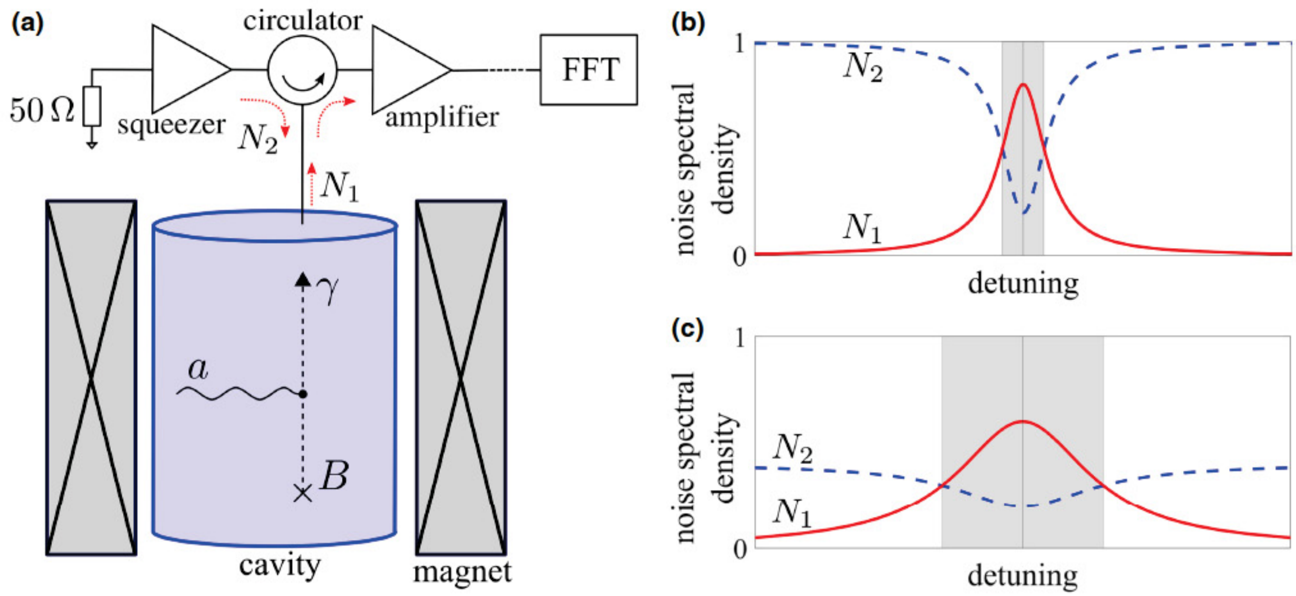

FIG. 3. (a) A conceptual schematic of the HAYSTAC haloscope. The detection scheme is similar to that of the ADMX, consisting of a resonant cavity in a strong magnetic field. HAYSTAC operates at microwave frequencies, where zero-point quantum fluctuations of the electromagnetic field are the dominant noise source. (b) The frequency dependence of the Johnson-Nyquist noise originating in the cavity (N_1) and reflected from the cavity (N_2), plotted as a function of the detuning from cavity resonance. The noise spectral density is in units of single-quadrature vacuum noise $h\nu_c/4$, where ν_c is the cavity frequency. The gray band indicates the range of frequencies where internal cavity noise dominates. (c) By squeezing one of the quadratures of the vacuum state incident on the cavity, HAYSTAC is able to reduce the reflected noise and overcouple the cavity, which broadens the frequency band over which internal cavity noise dominates [the gray region; note that the x axis spans the same scale as in (b)]. This increases the sensitive bandwidth at each cavity tuning step and therefore speeds up the axion search.

Ramsey protocol is repeatedly applied to the qubit, performing a quantum nondemolition cavity-photon-number parity measurement [51,124]. The resulting detection efficiency is approximately 41% and the false positive probability, proportional to the dark-count rate, is approximately 4×10^{-4} [127]. This detector has been used in a proof-of-principle search, which has set limits on the kinetic mixing parameter of hidden photon dark matter with mass near 25 μeV [127]. However, no axion limits have been set, because searching for axion dark matter requires the presence of a strong magnetic field, which would interfere with the detector operation. A possible way forward is to separate the axion-sensitive cavity, placed in a strong magnetic field, from the detector readout cavity. Achieving this while maintaining low dark-current rate and high detection efficiency is the formidable technical challenge that remains to be overcome [128].

The squeezing and the microwave-photon counting are complementary approaches. Squeezing should be deployed at low frequencies (axion masses), where it can accelerate an axion search even in the presence of thermal photons, which would be a deleterious background for a photon-counting device. At high frequencies, photon counting is more effective. It eliminates noise due to vacuum fluctuations, since no real photons are generated by vacuum. Photon counting is also more robust than squeezing against loss, because vacuum fluctuations in lossy components add electromagnetic field noise but do not generate real photons. The crossover frequency depends on the technical aspects of each approach but can be estimated to be approximately 10 GHz [112]. Both the squeezing the photon-counting approaches need to solve the technical challenges that arise from the fundamental incompatibility of superconducting quantum devices with high magnetic fields.

C. Radio-frequency quantum sensors and atomic systems

In the lower mass range, at radio frequencies, thermal noise dominates over zero-point quantum fluctuations. The crossover is set by the typical dilution-refrigerator experiment temperature of approximately 100 mK, which corresponds to 2 GHz. Nevertheless, the experimental sensitivity of lumped-element resonant axion-dark-matter searches benefits from low readout-amplifier noise, all the way down to the standard quantum limit [129]. The reason for this can be understood by means of an argument similar to the description of the HAYSTAC squeezing approach. Lower readout-amplifier noise does not change the on-resonance ratio of a potential axion signal to the thermal noise but it does increase the sensitivity bandwidth over which thermal noise dominates over readout noise and it therefore speeds up the axion search frequency scan [116]. SQUIDs have been used as readout amplifiers in several lumped-element axionlike-dark-matter searches [107,109, 110] and they can be optimized to achieve near-quantum-limited performance [130]. The DM Radio collaboration is developing a radio-frequency quantum upconverter (RQU) device, with tunable noise impedance, and compatibility with high-quality-factor operation of the LC resonator that couples to axion dark matter [104,116]. The ultimate goal is not only to achieve the standard quantum limit on amplification but to develop an approach that goes beyond this limit, making use of back-action evasion to increase the sensitivity bandwidth.

Atomic systems provide an alternative approach to quantum sensing of electromagnetic fields. As mentioned previously, Rydberg atoms are sensitive detectors of microwave electric fields [131,132]. A number of experiments have explored entanglement and squeezing in order to make optimal use of quantum resources [133]. One of the applications most relevant for dark-matter searches is sensing of radio-frequency electric fields, which can be sourced by hidden photons or the axion-photon interaction. A recent experiment has demonstrated 240 nV $m^{-1}/\sqrt{Hz}$ sensitivity, using a two-dimensional trapped-ion crystal with approximately 150 ions [134]. The device measures the center-of-mass motion of the trapped-ion crystal, probed via the collective electronic spin of the $^9Be^+$ ions. Entanglement between these two degrees of freedom allows measurements that evade quantum back action and thermal noise, achieving an electric field sensitivity of approximately 4 dB below the standard quantum limit. The demonstrated sensitivity compares favorably to that of Rydberg atom electrometers. Another favorable feature of the trapped-ion technology for an axion-dark-matter search is the inherent presence of the strong magnetic field, which is necessary to convert axion dark matter into an electromagnetic field. This is in contrast to superconducting detector technologies, which must be operated near zero magnetic field. The most significant limitation of the trapped-ion approach would be the disadvantage of detecting an oscillating electric field, compared to a magnetic field, by the suppression factor, which scales as the ratio of the experimental size and the axion Compton wavelength. Nevertheless, if the trapped-ion platform can be scaled up, it has the potential to be competitive with existing limits on the electromagnetic interaction of axionlike dark matter in the nanoelectronvolt range.

V. SPIN SENSORS

Historically, experiments sensitive to axion-photon interaction have dominated searches for ultralight axionlike dark matter. However, as noted in Sec. III, there are two other nongravitational interactions that can be used to detect axion dark matter. These interactions couple axions to instrinsic spin angular momentum of electrons, nucleons, or nuclei. Quantum engineering of spin systems has been an active research direction in the field of quantum science, so it is natural to ask how these tools can aid the search for these interactions.

Both of the interactions in Eqs. (1) and (2) can be written in the following way:

$$\mathcal{H}_\sigma = \hbar\gamma \mathbf{B}^* \cdot \boldsymbol{\sigma}, \tag{4}$$

where γ is the gyromagnetic ratio and $\mathbf{B}^*$ is an oscillating pseudomagnetic field that exerts a torque on spin $\boldsymbol{\sigma}$. Note that this pseudomagnetic field does not obey Maxwell's equations; it is sourced by the axion-dark-matter field. In the case of the gradient interaction, $\mathbf{B}^*_{\rm gr} = g_{\rm gr}\nabla a/(\hbar\gamma)$ and in the case of the EDM interaction, $\mathbf{B}^*_{\rm EDM} = g_d a\mathbf{E}^*/(\hbar\gamma\sigma)$ [135]. In both cases, $\mathbf{B}^*$ oscillates at the Compton frequency ν_a of the axion field. The objective of experiments that search for these interactions is to detect the effect of this oscillating pseudomagnetic field on the evolution of the spin ensemble under study.

There are two approaches: detecting the energy deposited by the dark-matter field via spin flips and detecting coherent evolution or energy shift of spins as a result of the effective interaction in Eq. (4). The QUAX experiment takes the former approach, measuring the energy deposited by electron spin flips in the permeable material yittrium iron garnet (YIG) inside a microwave cavity [136–138]. There are related experiments and proposals, making use of interaction with magnons and with atomic electrons in rare earths [139–141]. The coherent approach is typically used to search for lower axion masses, in experiments that use nuclear spins. The Cosmic Axion Spin Precession Experiments (CASPEr) use nuclear magnetic resonance to search for the EDM and the gradient interactions of axionlike dark matter with nuclear spin ensembles [86,142–147]. The closely related comagnetometer experiments search for the gradient interaction with nuclear spins [148–150]. Stringent limits on the interaction in Eq. (4) at low axion masses can also be extracted from analysis of static EDM experimental data [151,152].

Most of the existing spin-based experimental approaches can be thought of as extensions of an experimental scheme that searches for the axion electromagnetic coupling but with a spin ensemble acting as the transducer between the axions and the electromagnetic detector. For instance, the QUAX experimental scheme is similar to a microwave-cavity axion search shown in Fig. 2, with a YIG spin sample placed inside the cavity [138]. The CASPEr experimental scheme is similar to a lumped-element circuit search at radio frequencies, with a nuclear spin sample acting as the transducer. In comagnetometer experiments, the role of the electromagnetic field detector is often played by a laser polarimeter. On the one hand, the use of spins

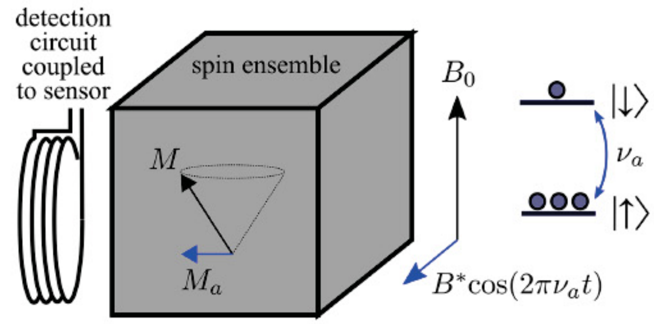

FIG. 4. A conceptual schematic of the CASPEr search for the EDM and gradient interactions of axionlike dark matter. The axionlike-dark-matter field interacts with the spin ensemble via the oscillating effective magnetic field B^*. Experiments search for the resulting tilt and precession of the spin-ensemble magnetization M, when the spin Larmor frequency is on resonance with the axion Compton frequency ν_a. The search for the unknown axion mass is performed by varying the leading field B_0, which tunes the spin Larmor frequency.

as transducers increases experimental complexity, since in addition to all the technical parameters in an electromagnetic search (such as sensor noise and coupling), one needs to calibrate and optimize spin-ensemble characteristics. On the other hand, this creates more freedom and opportunity to utilize materials and spin-ensemble control techniques that can dramatically improve sensitivity.

The interaction Hamiltonian in Eq. (4) gives rise to a torque on each spin, the magnitude of which is quantified by the Rabi frequency $\Omega_a = \gamma B^*/2$. In a resonant coherent detection experiment, the spin ensemble is placed in an external bias magnetic field B_0. Resonance occurs if the spin Larmor frequency $\gamma B_0/(2\pi)$ matches the oscillation frequency of the pseudomagnetic field B^*, which is the axion Compton frequency ν_a. If the initial spin orientation is along the bias magnetic field B_0, then the torque tilts them away from this direction. The experimental observable is the steady-state oscillating transverse magnetization

$$M_a = uM_0\Omega_a T_2 \cos(2\pi\nu_a t), \tag{5}$$

where $M_0 = p\hbar\gamma n$ is the equilibrium magnetization of the spin ensemble with polarization fraction p and number density n, T_2 is its spin coherence time, and u is a dimensionless spectral factor that takes into account the inhomogeneous broadening of the spin ensemble and the detuning between the axion Compton frequency and the spin Larmor frequency [147]. One of the ways to measure the transverse magnetization M_a is with a pickup coil that inductively couples the spin ensemble to an electromagnetic sensor (see Fig. 4). There are other more complex detection methods that make use of atomic magnetometers or comagnetometers [145,150,153].

Let us consider the quantum limits on the sensitivity of the scheme described above. There is still the standard quantum limit on the sensitivity of the electromagnetic field sensor that measures the spin magnetization M_a. This SQL, and the approaches that can circumvent it, have been discussed in Sec. IV in the context of searches for the axion-photon coupling. In addition, there is now another standard quantum limit—in the context of spins, this is called *spin-projection noise* (SPN) [154]. A simple way to understand SPN is to consider the following thought experiment. Suppose that a single spin-1/2 is prepared in the "spin-up" state, namely the quantum state with the $s_z = 1/2$ spin projection along the z axis. Then a measurement of the s_y spin component is performed. There are two possible outcomes, $s_y = +1/2, -1/2$, and they are equally likely. If this sequence is repeated N times or the experiment is performed on N uncorrelated spins, then the (random) mean value of the s_y spin component is normally distributed, with mean 0 and standard deviation $\sqrt{N}/2$. The uncertainty in the transverse spin projection corresponds to a $\delta\theta \approx 1/\sqrt{N}$ uncertainty in the polar angle of the spin or the spin ensemble. In a spin ensemble with number density n, there are $N = nV$ spins in volume V and the SPN magnetization is given by $M_{\mathrm{SPN}} \approx (\hbar\gamma/V)\sqrt{N} = \hbar\gamma\sqrt{n/V}$. As a benchmark, for proton nuclear spins in $V = 1$ cm^3 of water, $\mu_0 M_{\mathrm{SPN}} \approx 10$ fT, where μ_0 is the permeability of free space.

The existence of SPN was noted by Bloch in 1946 and it has been measured in macroscopic spin ensembles in a number of nuclear magnetic resonance (NMR) experiments [155–157]. For spin-based axion-dark-matter searches, conversion of SPN limits to the relevant coupling strength, g_{gr} or g_d, is analyzed in Ref. [158]. Some of the existing experimental efforts are now approaching SPN-limited sensitivity [147,149]. Technical challenges that have to be overcome include the necessity of a low-magnetic-field noise environment, the requirement of probing a broad range of Larmor frequencies, and, when searching for the EDM interaction, the need to work with static solid crystals [147]. These are unusual regimes from the point of view of NMR spectroscopy, where experiments are usually done at a fixed magnetic field, and solid-state NMR is usually done with magic-angle spinning [159,160]. Nevertheless, spin-based axion-dark-matter searches are likely to reach SPN-limited sensitivity in the next few years [158]. Accomplishing this will require quantum engineering of the spin-ensemble properties, including polarization and coherence time, as well as optimization of the electromagnetic sensors that are used for ensemble evolution readout. For example, the problem of readout back action on the spin ensemble (somewhat misleadingly called "radiation damping" in NMR spectroscopy) will have to be addressed, especially for gradient-interaction searches [158]. Spin-based experiments are likely to benefit from some of the accomplishments described in Sec. IV. For instance, superconducting microwave-photon detectors have already been used for magnetic resonance spectroscopy of small ensembles of electron spins [126]. In the near future, the RQU devices, with their tunable noise impedance and potential SQL-limited sensitivity, will play an important role in optimized spin-ensemble readout at radio frequencies.

Is it possible to use quantum engineering to go beyond the SPN? This question has inspired intense research, including ideas such as spin squeezing, entangled states, and quantum error correction [133,161,162]. There are also long-standing arguments that assert that many of these approaches cannot make significant improvements to the signal-to-noise ratio of an *optimized* experiment with SPN-limited sensitivity [163,164]. However, even if this is true, there may be other ways to engineer spin ensembles in order to improve such experiments. For example, in Sec. IV, we describe how squeezing can achieve an improvement in the useful bandwidth of a search for axionlike dark matter and therefore speed up the search over the axion Compton frequencies. Specific spin-ensemble quantum engineering schemes will be explored in the next 3–5 years. The maximization of sensitivity to small background fields necessitates the use of macroscopic spin ensembles, on the order of one mole, which leads to unprecedented technical challenges (listed in the previous paragraph), compared to much-smaller-scale demonstration experiments. Nevertheless, there are grounds for significant optimism, given the flexibility that is created by choosing the spin ensemble with optimal properties, such as coherence, and combining it with state-of-the-art precision sensors and metrology techniques. If fundamental sensitivity limits, imposed by thermal and quantum noise, can be reached, we are likely to see important scientific breakthroughs in spin-based axion-dark-matter searches within the next 5–10 years.

VI. OUTLOOK

The dark-matter puzzle is one of the most compelling leads in our search for physics beyond the standard model. After decades of searching for the WIMP, there is a rapidly growing recognition that experimental efforts should be broadened and the QCD axion is a very well-motivated target. The nature of experimental searches for axionlike dark matter offers clear opportunities for the powerful toolbox of quantum science to expand the scientific reach of these searches. Some of these opportunities are sketched in Fig. 1. The ADMX haloscope is searching for the QCD axion and is projected to cover the 2.5–8.3 μeV mass range by 2025. Quantum technologies create opportunities to expand the search range by orders of magnitude, potentially 1 peV to 1 meV. The approaches covered in this Perspective include quantum spin ensembles, the optimal sensitivity of which is at lower axion masses, radio-frequency quantum sensors, electromagnetic cavity squeezing and entanglement, and microwave-photon counting using qubits. Whether or not one of these approaches helps discover the axion depends, of course, on its unknown mass and interactions. Perhaps unexpected discoveries will be made. Quantum science is letting us look in places where no one has ever looked before. Whatever the outcome of the search for axionlike dark matter, there is no doubt that the development of quantum precision measurement methods will lead to new devices, sensors, and technologies, which may find diverse applications in fundamental and applied science. After all, quantum engineering is already a key driving force behind innovation and scientific progress in disciplines ranging from computer science to systems engineering and the life sciences.

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