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Phys. Rev. A **108**, 010101 — Published 17 July 2023

DOI: 10.1103/PhysRevA.108.010101

Probing fundamental physics with spin-based quantum sensors

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(Dated: May 23, 2023)

The applications of spin-based quantum sensors to measurements probing fundamental physics are surveyed. Experimental methods and technologies developed for spin-based quantum information science have rapidly advanced in recent years, and these tools enable increasingly precise control and measurement of spin dynamics. Theories of beyond-the-Standard-Model physics predict, for example, discrete-symmetry-violating electromagnetic moments correlated with particle spins, exotic spin-dependent forces, and coupling of spins to ultralight bosonic dark matter fields. Spin-based quantum sensors can be used to search for these myriad phenomena, and offer a methodology for tests of fundamental physics that is complementary to particle colliders and large-scale particle detectors. Areas of technological development that can significantly enhance the sensitivity of spin-based quantum sensors to new physics are highlighted.

I. INTRODUCTION

There are multiple profound mysteries in fundamental physics, ranging from the nature of dark matter and dark energy to the origin of the matter-antimatter asymmetry of the universe. In turn, there are a plethora of theoretical proposals to explain these mysteries. However, despite intense scientific activity, there are currently few if any clear experimental signatures indicating how best to unravel these mysteries. Consequently, in this era it is advantageous to cast a wide net in the search for new physics. A powerful, versatile, and relatively low-cost approach is to use the techniques, systems, and devices developed in the rapidly-growing field of quantum information science (QIS). Quantum systems can be made extremely sensitive to external perturbations. Indeed, much of the work in quantum science is focused on how to minimize this sensitivity, in order to prevent decoherence. Here we outline a complementary approach, which seeks to maximize the sensitivity of quantum systems to new fundamental physics.

There are a growing number of experiments that make use of quantum resources and systems to search for spin-dependent interactions of novel origin, which are predicted by a wide variety of beyond-the-Standard-Model physics theories [1, 2]. Experimental techniques for precision measurement of such spin-dependent interactions have substantially advanced over recent decades, in no small part because they share a common foundation with the robust program of research on spin-based quantum sensors for measurement of magnetic fields, magnetic resonance, and related phenomena. Furthermore, control and measurement of spins, spin ensembles, and quantum materials is at the heart of many QIS and quantum computing schemes [3–5]. Thus the development of spin-based quantum sensors offers significant opportunities for cross-fertilization between fundamental and applied research.

In the context of searches for beyond-the-Standard-Model physics, precision measurements using the tools of QIS, magnetic resonance, and atomic, molecular, and optical (AMO) physics are complementary to collider-based high-energy-physics research [1, 2]. Precision experiments searching for discrete-symmetry-violating permanent electric dipole moments (EDMs), exotic spin-dependent interactions mediated by new light bosons, and spin-dependent couplings to ultralight bosonic dark matter fields [e.g., axions, axion-like particles (ALPs), and dark/hidden photons] can probe new physics associated with energy scales far beyond the reach of modern particle colliders [1, 2]. This is because precision-measurement experiments[1] are designed to detect extremely subtle energy shifts.[2] Because of their energy resolution, such experiments can be sensitive to physics gen-

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[1] Note that while the experiments described herein are commonly referred to as "precision-measurement" experiments, since the overarching goal of these experiments is to reveal new physics, the more relevant metric is "sensitivity." Sensitivity refers to discovery potential of an experiment, while precision refers to how finely an experiment can measure a given quantity.

[2] Experiments have reached astonishing sensitivity to frequency

erated by new high-mass particles. For example, EDM searches are now sensitive to CP-violation due to virtual particles with masses $M \gtrsim 10\,\mathrm{TeV}/c^2$ [9–12]. Precision magnetic resonance-based searches for axion-like dark matter [13] are sensitive to ALPs arising from spontaneous symmetry breaking at scales f_a reaching up to the grand unified theory (GUT) scale ($\sim 10^{16}$ GeV) and Planck scale ($\sim 10^{19}$ GeV) [14].

Improving the sensitivity of spin-based sensors will extend the reach of such experiments to higher energies as coupling constants typically scale proportionally to $1/M$ or $1/f_a$. Spin-based sensors can also be used as particle detectors by precisely measuring and characterizing changes to the environment caused by new particle interactions. Because precision experiments are often carried out at the "table-top" scale involving relatively small teams of researchers and relatively fast timelines from conception to data, they offer affordable opportunities to explore many creative theoretical scenarios of beyond-the-Standard-Model physics.

In terms of technological development of instrumentation essential for expanding the reach of precision spin-based sensors for fundamental physics research, there are a number of high priority areas:

- Find ways to enhance the number of polarized spins N via optical pumping and other hyperpolarization methods [15] and quantum control techniques, as the shot-noise-limited sensitivities of spin-based sensors generally scale proportionally to $1/\sqrt{N}$ [16];
- Develop methods and find systems to achieve the longest possible spin coherence times τ, since measurement sensitivity generally scales as $1/\sqrt{\tau}$ [16];
- Improve fundamental sensitivity of spin-based sensors via new measurement schemes involving, for example, quantum back-action evasion [17] and rapid averaging of quantum uncertainty in highly correlated spin systems (e.g., ferromagnets [18, 19]);
- Study new atomic, molecular, and condensed-matter systems that feature enhanced sensitivity to beyond-the-Standard-Model physics, such as ferroelectric crystals [20, 21], polyatomic molecules [22], and deformed nuclei [23];
- Advance tools, such as comagnetometers [6, 24] and quantum sensor networks [25], to control and eliminate systematic errors and spurious technical noise;
- Find techniques to increase the bandwidth of spin-based sensors to explore higher frequencies [26] and therefore higher boson masses in dark matter haloscope searches;
- Develop methods to speed up the scanning rate of magnetic-resonance-based dark matter haloscope searches [27] in order to explore larger ranges of boson masses over a given measurement time;
- Design and implement new strategies for spin-based sensors at smaller length scales to probe higher mass exotic bosons that mediate forces at smaller length scales [28];
- Enhance the accuracy of spectroscopic measurements and theoretical calculations of atomic, molecular, and nuclear systems to enable new tests of fundamental interactions [29].
- In cases where the spin precession time is not limited by the spin coherence time, enhance energy resolution beyond the apparent shot-noise-limit using entangled spin states.

II. SEARCHES FOR NEW PHYSICS

Measurements of spins can probe new physics in three primary ways:

- First, new physics may break symmetries of the Standard Model, giving rise to novel responses of Standard Model spins to other Standard Model fields (Sec. III).
- Second, the new physics may directly affect the spin, for example, via an interaction between a new field and the spin (Secs. IV – VI).
- Third, the environment of the spin may be affected by the new physics and the spin can discover the new physics by sensing changes to its environment (Sec. VII).

The canonical science target for the first kind of effect, namely, the breaking of Standard Model symmetries by new physics, is the search for the permanent electric dipole moment (EDM) of fundamental particles. If a fundamental particle possesses an EDM, an applied electric field will cause the spin of the particle to precess. Such a dipole moment violates CP symmetry (the combined symmetry of charge conjugation, C, and parity, P) and it is a natural facet of many theories of physics beyond the Standard Model [30]. Indeed, the existence of such CP violation is indicated by the existence of the matter-antimatter asymmetry in the universe [31].

Key science targets that cause the second kind of effect, namely, direct effects on the spin itself, include particles such as axions, ALPs, massive vector bosons and other ultra-light bosons [14]. Particles of this kind emerge in several theoretical frameworks that are aimed at solving outstanding problems of the Standard Model such as the strong-CP [32, 33] and hierarchy [34–37] problems. They are also predicted to emerge as a generic consequence of

shifts approaching the pHz (10^{-12} Hz) scale, corresponding to energy scales $\lesssim 10^{-26}$ eV [6–8].

string theory [38, 39]. The key reason for the ubiquity of such particles in these extensions of the Standard Model is due to effective field theory [14]. Given a light field, interactions with the spin of Standard Model fermions is one of the dominant channels that would allow this light field to interact with Standard Model particles and fields in a technically natural [3] way. These fields are thus natural portals into the "ultra-violet" or high energy/mass regime. Such bosonic fields can be detected by sourcing them in the laboratory with spin-polarized (or unpolarized) test masses or by looking for a cosmological abundance of such bosons. The latter possibility is well motivated since many cosmological scenarios (such as inflation) can naturally produce a relatively large cosmic abundance of these particles [40, 41]. If discovered, these particles thus have the potential of solving both the problem of dark matter as well as unveiling other mysteries of the early universe. In addition, it is also possible that complex dark sectors could directly source these long-range fields giving rise to new long-range interactions between the dark matter and Standard Model spins [42]. In light of poor observational constraints on such particles, it is vital to develop technological probes that are able to cover wide swaths of parameter space. The developments in QIS technologies over the past decade now makes a broad probe of parameter space experimentally feasible [2, 43].

Science targets for the third possibility, namely, the use of spins to detect the effects of new physics on the environment of the spin, includes the detection of crystal damage caused by dark matter interactions and the ability to use spins to detect changes caused to surfaces at the single atom level, with the changes being produced as a result of dark matter interactions [44]. The former phenomenon could conceivably be used to identify the direction of dark matter induced nuclear recoil while the latter could potentially be used to detect light dark matter.

III. SEARCHES FOR PARITY- AND TIME-REVERSAL-VIOLATING ELECTRIC DIPOLE MOMENTS (EDMS)

The first way that sensitive measurements of spin dynamics can probe new physics identified in Sec. II is via searches for discrete symmetry violations. The primary focus of recent research has been measurement of *permanent* electric dipole moments (EDMs) in atomic, molecular, and nuclear systems. There have been a number of reviews on the topic of EDMs, see for example Refs. [1, 2, 30, 45–53]. A nonzero EDM $\boldsymbol{d}$ of an elementary or composite particle must be proportional to the total angular momentum $\boldsymbol{F}$ of the system (a fact that follows from the Wigner-Eckart theorem and the fact that no additional quantum numbers are required to describe the system, see, for example, Refs. [45, 54]). Since $\boldsymbol{d}$ is odd with respect to mirror-symmetry (parity, P) and even under time-reversal (T) while $\boldsymbol{F}$ is even under P and odd under T, the existence of an EDM violates P and T symmetries. Thus an EDM is a result of what are classified as P- and T-violating fundamental interactions, and, assuming CPT invariance, CP-violating interactions. Such symmetry-violating interactions can endow elementary particles such as electrons and quarks with EDMs, which can in turn create EDMs of atoms, molecules, and nuclei. Symmetry-violating interactions between constituent particles of composite systems can also induce electrical polarization along $\boldsymbol{F}$ and generate EDMs.

The predominance of matter over antimatter is incompatible with Standard Model mechanisms of baryogenesis [55], and it is widely believed that the missing ingredient is a new, larger source of CP violation that would also generate EDMs. A wide variety of beyond-the-Standard-Model theories predict EDMs near present experimental sensitivities. For instance, existing experimental limits on EDMs have established some of the most stringent constraints on supersymmetric theories, in many scenarios beyond constraints from collider experiments [56].

Depending on whether the atomic or molecular system studied is *paramagnetic* (with unpaired electron spins) or *diamagnetic* (with closed electron shells but nonzero nuclear spin), different types of physics can be probed: EDM experiments with paramagnetic systems can target electron EDMs and CP-violating electron-nucleon interactions; diamagnetic systems can target nuclear EDMs and CP-violating hadronic and other semileptonic interactions. Thus it is valuable to develop techniques and experiments to study both paramagnetic and diamagnetic systems.

The general approach of EDM experiments is to search for the combined effect of a P- and T-odd Hamiltonian and an applied electric field $\boldsymbol{E}$, which results in an energy shift $\pm\Delta E_{\rm edm}$ for a given quantum state of the atom or molecule, where the sign of the effect depends on the projection of the spin along the quantization axis. A preliminary consideration is that in the nonrelativistic limit there is no energy shift when $\boldsymbol{E}$ is applied to a neutral system, even if it is composed of particles possessing nonzero EDMs. This is because particles will rearrange upon application of the applied field $\boldsymbol{E}$ so that the internal field $\boldsymbol{E}_{\rm int}$ cancels $\boldsymbol{E}$ at the positions of the constituent particles, a result known as Schiff's theorem [57]. However, relativistic effects not only evade Schiff's theorem

[3] "Technical naturalness" refers to the scenario where a dimensionless coupling constant g describing an interaction in a theory is $\ll 1$ because of symmetry breaking. There exists a symmetry which if respected implies $g = 0$, but if the symmetry is broken as some high energy scale, at low energies it can be nonzero but quite small compared to unity. The property of technical naturalness protects the coupling constant g from large perturbative corrections that would tend to increase its value closer to unity at low energy scales where the measurements are performed.

but can even lead to enhancement of EDM observables [48, 58]. Because relativistic effects are more prominent in heavy atoms, ΔE_{edm} can be significantly enhanced in systems with large atomic number Z [48, 58], and thus EDM experiments employ heavy atoms such as Tl, Th, Cs, Hg, and Xe. Typically the system is spin polarized via optical pumping or some other hyperpolarization technique such that it is in a superposition of quantum states with opposite EDM-induced energy shifts. A nonzero EDM will cause the polarized spins to precess in the presence of $\boldsymbol{E}$ by an angle $\phi = 2\Delta E_{\text{edm}}\tau/\hbar$, where for maximum precession the time τ is given by the spin-coherence time. The best achievable energy resolution for a single-particle measurement is $\hbar/(4\tau)$ (a consequence of the energy-time uncertainty relation); measuring with N uncorrelated systems for a total time t gives an energy resolution of δE

$$\delta E \approx \frac{\hbar}{4}\frac{1}{\sqrt{\tau t N}}\,. \tag{1}$$

Considering this approach, there are several general areas of technological development that can advance the fundamental sensitivity of EDM measurements:

- increase ΔE_{edm} by finding atomic and molecular systems with maximal enhancement factors;
- improving hyperpolarization and quantum control techniques so that the total number N of polarized atoms/molecules can be increased;
- achieve longer spin-coherence times τ.

At least equally important is improving control of systematic errors that could mimic EDM signals. Among the most pernicious systematic effects that have plagued generations of EDM experiments are those due to uncontrolled magnetic fields $\boldsymbol{B}$ that couple to the magnetic dipole moments of the atoms or molecules, causing Larmor precession of spins. While many magnetic field effects can be distinguished from effects due to EDMs by reversal of the direction of $\boldsymbol{E}$, there can be $\boldsymbol{B}$-fields correlated with the direction of $\boldsymbol{E}$ due to leakage currents as well as motional magnetic fields $\propto \boldsymbol{E}\times\boldsymbol{v}/c$, where $\boldsymbol{v}$ is the particle velocity in the lab frame. Magnetic-field-related systematic errors are generally reduced using the technique of *comagnetometry* [6], where simultaneous measurements in the same volume are carried out on either different species [59] or different quantum states of the same species [60].

In addition to comagnetometry, controlling and monitoring the magnetic environment of an EDM experiment should also make use of ultra-stable and sensitive magnetometers surrounding the experiment. Optically pumped atomic magnetometers [61, 62] and in general magnetometers probed by light are subject to shifts that challenge their stability (although, it should be noted, that there are techniques to ameliorate or cancel such systematic effects, see, e.g., Refs. [63–65]). Nuclear spin magnetometers, in particular those based on ^{3}He [66, 67], have the potential as quantum sensors to provide the unprecedented stability required for future EDM experiments.

Earlier generations of electron-EDM experiments generally employed paramagnetic atomic systems like Cs [68] and Tl [59, 69], and there are ongoing atomic EDM experiments employing advances in laser-cooled and trapped atoms and other state-of-the-art QIS methods [70–72]. However, in recent years the focus has shifted to molecular systems such as YbF [73], ThO [9, 10], and HfF^+ [11, 12]. The molecular systems have enabled orders-of-magnitude improvements in sensitivities to electron EDMs through their larger enhancement factors which increase ΔE_{edm} as compared to atomic systems as well as opening a variety of techniques to control and reduce systematic errors. Efficient systematic error control in molecular EDM experiments is accomplished by experimenting on particular molecular states that have reduced sensitivities to magnetic perturbations while retaining sensitivity to EDM-induced effects, and by using optical and radio-frequency fields for quantum control to switch between different quantum states that allow rapid measurement and cancellation of many systematic errors. Further improvements in cooling [74] and control of molecules [4, 75], extending spin-coherence times [76, 77], increasing the number of polarized molecules [78–80], and advances in comagnetometry [22] and other methods to control systematic effects are among the paths toward further advances. In addition to ongoing experiments [9–12, 73], a number of new experiments are under development [81].

The leading diamagnetic (nuclear) EDM experiment has employed Hg atoms [8], complementary to direct measurements of the neutron EDM [82]. The sensitivity of the Hg EDM experiment results from a relatively high density of optically polarized atoms ($N \sim 10^{14}$) and long coherence times (hundreds of seconds), as well as a variety of auxiliary measurements and techniques developed over the years to reduce systematic errors [83]. Searches for EDMs of diamagnetic atoms in other systems have been carried out [84–89]; many of these are ongoing efforts with the prospect of improving measurement accuracy by orders of magnitude, such as the radium EDM search in which several upgrades are in the process of being implemented [90, 91]. There are also a number of new experiments that have the potential to explore unconstrained parameter space for symmetry violating effects in the nuclear sector [22, 92–98], such as the CENTREX experiment that employs a cold beam of TlF molecules [99], a search particularly sensitive to the proton EDM [100, 101].

Technological improvements that can enhance the sensitivity of EDM experiments include any methods that result in longer spin-coherence times, such as longer beam lines, slower/colder beams, and trapping of molecules which can lengthen spin-coherence times by orders of magnitude. Sensitivity can also be improved by increasing count rates via beam cooling and focusing, more efficient probing/detection methods, improved trapping techniques, and brighter molecular sources. It is important to note that all three of the leading electron EDM searches with molecules [10–12, 73] are presently statistics limited, meaning that technological advances in the aforementioned areas can lead directly to improved sensitivity.

An important area of technological development is toward the use of deformed nuclei for EDM searches [23]. Because the motion of a nucleus within an atom or molecule is deeply nonrelativistic, Schiff's theorem [57] implies that any nuclear EDM is mostly screened from external fields. Nonetheless, symmetry violating nuclear interactions can change the nuclear charge and current distributions, and lead to nonzero energy shifts due to finite-nuclear-size effects described by the Schiff moment [102]. Deformed nuclei that possess a reflection antisymmetric shape in the nuclear frame, such as Fr, Ra, Th, and Pa that may have static octupole deformations, have enhanced nuclear Schiff moments (by orders of magnitude) and therefore lead to comparably larger atomic and molecular EDMs [23, 103–106].

Another rapidly developing technology, useful not only for nuclear EDM experiments but also for a wide range of searches for beyond-the-Standard-Model physics, are new methods for nuclear spin comagnetometry [87, 107–109]. These techniques can improve control of systematic errors, often the limiting factor in EDM experiments.

A new direction of particular interest is the use of polyatomic molecules for EDM searches, which can enable application of laser cooling techniques [110] in conjunction with internal comagetometry and full polarization [22, 111]. Polyatomic molecules show considerable promise for both electron and nuclear EDM experiments.

A different approach is to develop solid-state systems for EDM experiments [112, 113]. Such solid-state EDM experiments sacrifice the long spin-coherence times possible in gas-phase atomic and molecular experiments for a significantly larger signal due to the higher density of spins in a solid. As first suggested in Refs. [114, 115], an electron EDM search can be carried out using unpaired election spins bound to a crystal lattice: when an electric field $\boldsymbol{E}$ is applied, if the electrons possess a non-zero EDM the spins will become oriented parallel to $\boldsymbol{E}$ and generate a nonzero magnetization [20, 116, 117]. The inverse experiment can also be performed, where a material is magnetized (spin-polarized) and one searches for electric polarization due to a nonzero electron EDM [118]. Technological improvements are needed to reduce systematic errors in such solid-state EDM experiments, for example due to heating and dielectric relaxation.

In the longer term, it is likely that advances along multiple fronts will allow the frontiers of EDM searches to be pushed even further. For example, using heavy polar molecules with deformed nuclei in an EDM experiment taking full advantage of state-of-the-art cooling, trapping, and molecular production could allow sensitivity to symmetry-violating interactions many orders of magnitude beyond what is possible today [98]. Combining the Schiff-moment enhancement of an octupole-deformed nucleus with the relativistic enhancement, there are molecular species such as ^{229}ThO, ^{229}ThOH, ^{229}ThF$^+$, and ^{225,223}RaOH$^+$, ^{225,223}RaOCH$_3^+$, ^{225,223}RaF, ^{225,223}RaAg, and ^{223}FrAg that are up to 10^6 times more sensitive per particle to CP-violating physics than ^{199}Hg [119–121]. Note that dedicated institutes for low-energy nuclear science research, such as the Facility for Rare Isotope Beams (FRIB), TRI-University Meson Facility (TRIUMF), and Isotope mass Separator On-Line (ISOLDE), have the capability to produce these isotopes for use in practical quantities and enable precursor spectrocopic studies [122]. Excellent candidates are Ra-containing molecules [123], since Ra has a well-studied nuclear deformation [105, 124], and many Ra-containing molecules can be laser cooled. For example, RaOCH$_3^+$ was recently synthesized, captured in an ion trap and cooled [97], opening the potential for an experiment that takes advantage of the advanced quantum control techniques possible with cold ions [11, 12, 98, 125]. A novel related concept is to use the radioactive species ^{229}Pa, which may be a highly deformed nucleus, embedded in an optical crystal to search for its strongly enhanced symmetry-violating magnetic quadrupole moment or nuclear Schiff moment [92].

Another route is to combine the advantages of the long coherence times and quantum control possible in gas-phase atomic and molecular experiments with the high spin densities possible in solid-state systems [126–132]. The idea is to trap atoms and molecules with high intrinsic sensitivity to symmetry-violating interactions within inert cryogenic crystal matrices. In order for an EDM experiment based on this approach to surpass the sensitivities of gas-phase experiments, it is essential that both high density of the target species is achieved and that the target species retains all the key properties that enable quantum control and sensing in the inert crystal environment (long coherence times and efficient polarization and read-out of spin states). While experiments with alkali atoms in solid hydrogen and solid helium have demonstrated long coherence times and efficient optical pumping and probing [133–135], the alkali atom densities so far have been low. On the other hand, both high alkali atom density and relatively long spin-coherence times ($\tau \equiv T_2 \sim 0.1\,$s) have been demonstrated in solid parahydrogen [136–138]. While there are experimental hurdles yet to be overcome, such as relatively short spin-ensemble dephasing times (T_2^*) due to the polycrystalline nature of the parahydrogen samples used so far [136, 137], there are viable paths forward to taking full advantage of the possibilities of this system by, for example, creating single-crystal cryogenic samples [139] and higher purity parahydrogen matrices [140]. New experiments using rubidium atoms trapped in solid neon matrices show promising results in terms of spin coherence and the ability to optically control and readout the rubidium spin properties [141].

Many of the EDM experiments described here rely on quantum sensing and control of spin ensembles, analogous to those used in QIS, and can therefore borrow new tools from this rapidly-advancing field [3–5]. As highlighted above, QIS methods have already been implemented in EDM searches. A variety of techniques for quantum control and readout have been used to take advantage of the rich internal structure of molecules [142, 143] both for QIS applications [75, 125, 144, 145] and for EDM searches [11, 12]. There are a number of new approaches that may offer synergistic opportunities, such as cavity-enhanced readout of solid-state spin sensors [146], employing quantum entanglement between atoms and molecules to transduce quantum information across widely varying frequencies [147], and using the coupling between phonons and polar molecules trapped in Coulomb crystals for non-optical quantum logic operations [148]. Quantum entanglement and spin squeezing have been shown to improve signal-to-noise [149–152] over measurement time scales shorter than the relevant coherence time [16, 26, 153, 154], which could be useful for enhancing measurement bandwidth and improving single-shot measurement precision.

It is widely believed that new sources of CP-violation are required to explain the cosmological matter-antimatter asymmetry [155]. Consequently, there are a wide range of beyond-the-Standard-Model theories predicting observable EDMs "just around the corner" of present experimental sensitivities [56]. Discovery of a nonzero EDM would herald the existence of new particles, and can explore new physics from particles with masses beyond the direct reach of any conceived accelerator [1, 2].

IV. SEARCHES FOR EXOTIC SPIN-DEPENDENT INTERACTIONS USING MAGNETOMETRY AND COMAGNETOMETRY

The second class of precision experiments highlighted in Sec. II are direct searches for exotic spin-dependent interactions originating from beyond-the-Standard-Model physics. Many theories predict the existence of new force-mediating bosons that couple to the spins of Standard Model particles [2]. Regardless of the specifics of the fundamental theory, if the new interaction respects rotational invariance, there are only a relatively small number of long-range interaction potentials that can exist as described in detail in Refs. [156–159]. The range of such a fundamental interaction is parameterized by the Compton wavelength of the force-mediating boson: $\lambda_c = \hbar/(mc)$, where m is the boson mass. For example, exchange of an exotic spin-0 boson (such as an axion [156]) with pseudoscalar coupling to fermion 1 and scalar coupling to fermion 2 leads to a monopole-dipole potential of the form:

$$\mathcal{V}_{ps}(r) = \frac{g_p^{(1)} g_s^{(2)} \hbar}{8\pi m_1 c} \boldsymbol{S}_1 \cdot \hat{\boldsymbol{r}} \left(\frac{1}{r\lambda_c} + \frac{1}{r^2} \right) e^{-r/\lambda_c} , \quad (2)$$

where $g_p^{(1)}$ and $g_s^{(2)}$ parameterize the vertex-level pseudoscalar and scalar couplings, respectively, $\boldsymbol{S}_1$ is the spin of fermion 1, m_1 is mass of fermion 1, and $\boldsymbol{r} = r\hat{\boldsymbol{r}}$ is the displacement vector between the fermions. The potential $\mathcal{V}_{ps}(r)$ causes an associated spin-dependent energy shift. The basic experimental program is thus to hunt for all possible types of interactions at various length scales between Standard Model fermions (typically electrons, protons, and neutrons in the case of AMO experiments). Through the framework of Refs. [156–159], the results of experiments can be interpreted in terms of fundamental physics theories [2].

One of the primary experimental strategies is to employ a sensitive detector of torques on spins and then bring that spin-based torque sensor within $\sim \lambda_c$ of an object that acts as a local source of an exotic field (e.g., a large mass or highly polarized spin sample). Such experiments are closely analogous to spin-based magnetometry [61, 62], where the effect of an ambient magnetic field $\boldsymbol{B}$ is measured by sensing the $\boldsymbol{\mu} \times \boldsymbol{B}$ torque on spins with magnetic moment $\boldsymbol{\mu}$. This is equivalent to measuring the magnetic-field-induced energy shift between Zeeman sublevels via observation of the time-evolution of a coherent superposition of spin states in the probed system. Exotic spin-dependent interactions act as "pseudo-magnetic fields" and generate analogous effects, albeit with couplings to Standard Model particles that can be completely different from those due to a real magnetic field [160, 161].

The central technology in these experiments is the spin-based sensor employed. The accessible parameter space depends on the overall sensitivity, which determines how small a coupling can be observed, as well as the size and geometry of the sensor, which determines what interaction range λ_c (boson mass m) can be probed. Since the observable in these experiments is a spin-dependent energy shift, just as in the case of the EDM experiments discussed in Sec. III, a sensor employing N independent spins with coherence time τ has a shot-noise-limited sensitivity described by Eq. (1). However, as noted in Ref. [162], a practical benchmark for comparison of different magnetometer technologies is the *energy resolution limit* (ERL). A heuristic argument supporting the ERL comes from considering measurement of a magnetic field B using a sensor whose active element fills a volume V. Suppose that the measurement is carried out over a time t and results in a determination of the measured magnetic field to be $B_0 + \Delta B$, where B_0 is the true mean (expectation) value of the field and ΔB characterizes the measurement error. The associated measurement bias in the average magnetostatic energy, ΔE_B, is

$$\Delta E_B \approx \frac{V}{2\mu_0} \langle \Delta B^2 \rangle , \quad (3)$$

where μ_0 is the magnetic permeability of free space and $\langle \cdots \rangle$ indicates the average. Multiplying ΔE_B by the measurement time t yields a quantity with units of ac-

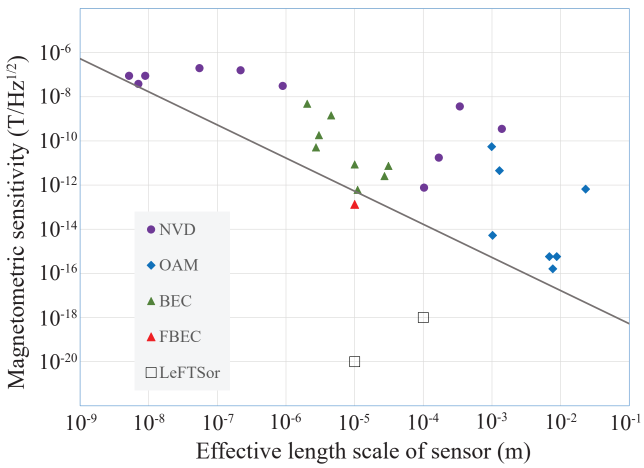

FIG. 1. Summary of the size and sensitivity of spin-based magnetometers. Experimentally demonstrated magnetometers are represented by filled markers, projected sensitivity of proposed magnetometers are represented by unfilled markers. The gray line indicates the energy resolution limit (ERL) described by Eq. (4). The purple circles correspond to nitrogen-vacancy diamond (NVD) magnetometers [164–171], the green triangles correspond to atomic Bose-Einstein condensate (BEC) magnetometers [150, 172–179], and the blue diamonds correspond to optical atomic magnetometers (OAM) [149, 180–186]. The red triangle represents the sensitivity of the recently demonstrated single-domain ferromagnetic BEC magnetometer (FBEC) that surpasses the ERL [187]. Levitated ferromagnetic torque sensors (LeFTSors), represented by the unfilled black squares, are predicted to surpass the ERL by many orders of magnitude [18, 19]. Figure adapted with permission from Ref. [162]; does not include non-spin-based magnetic sensors based on, for example, superconducting quantum interference devices (SQUIDs).

tion. If one assumes that quantum mechanics imposes a lower limit, equal to Planck's constant $\hbar$, on the contribution of the magnetic field measurement uncertainty to the action, one arrives at the ERL:

$$\langle \Delta B^2 \rangle \gtrsim \frac{2\mu_0 \hbar}{V t} \, . \tag{4}$$

For a detailed discussion of the ERL and the origin of magnetometric sensitivity limits specifically for spin-precession-based sensors, see Ref. [163].

Therefore, a major technological leap in the search for exotic spin-dependent interactions at various length scales would be to find methods to surpass the ERL. One promising technology along these lines is the development of levitated ferromagnetic torque sensors (LeFTSors) [18, 19, 188–192]. The active sensing element consists of a hard ferromagnet, well isolated from the environment by, for example, levitation over a superconductor via the Meissner effect. The mechanical response of the levitated ferromagnet to an exotic spin-dependent interaction can be precisely measured using a superconducting quantum interference device (SQUID). For sufficiently slow rotational motion of the ferromagnet, its angular momentum is dominated by its intrinsic spin, and it behaves as a gyroscope [18]. For faster motion, the levitated-ferromagnet dynamics are dominated by pendulum-like librational motion [19]. In either regime, LeFTSors are predicted to be able to surpass both the ERL and even the standard quantum limit (SQL) for uncorrelated spins described by Eq. (1).

The ability of LeFTSors to achieve this sensitivity is a result of the high correlation of the electron spins in a ferromagnet, which are locked together along a well-defined local direction by magnetic anisotropy, ultimately converting the field measurement into a mechanical measurement [19]. The quantum uncertainty in the spin orientation is rapidly averaged by the strong internal interactions in the ferromagnet [18]. In the case of a LeFTSor, the ultimate quantum- and thermal-noise-limited uncertainty in the measurement of a magnetic field is derived from the fluctuation-dissipation theorem [18]:

$$(\Delta B)^2 \gtrsim \frac{2\alpha k_B T}{\hbar \omega_0^2 \gamma^2} \frac{1}{N t^3} \, , \tag{5}$$

where α is the Gilbert constant, k_B is Boltzmann's constant, T is the temperature, ω_0 is the ferromagnetic resonance frequency, γ is the gyromagnetic ratio, and N is the number of polarized spins. For a micron-scale ferromagnet levitated above a perfect superconductor at cryogenic temperatures, the magnetometric sensitivity (5) can surpass the ERL (4) and SQL for uncorrelated spins (1) by many orders of magnitude. Practical limits on the sensitivity, well above the ultimate limit (5), are predicted to arise due to magnetic coupling of the spin-fluctuations to the non-zero external magnetic field [193] and, for example, perturbation due to collisions with residual gas molecules [18, 191].

Recently, a magnetic field sensor surpassing the ERL was demonstrated: a single-domain spinor Bose-Einstein condensate (BEC) [187]. Similar to the LeFTSor concept, ultracold two-body interactions in the BEC create a fully coherent, single-domain state of the atomic spins that enables the system to evade the ERL that limits traditional spin-based sensors. The experiment described in Ref. [187] confirms the principles underlying the promise of next-generation torque sensors such as LeFTSors, and also emphasizes the connection to highly correlated spin systems of particular interest for QIS applications [194].

A variety of other directions to improve fundamental and practical sensitivity of spin-based magnetometers are being explored, including bandwidth enhancement via spin squeezing [26, 195, 196], spin-polarized matter-wave interferometry [197–199], and methods to utilize many-body collective correlation among spins [200]. A new high-frequency magnetometer based on electron spin resonance, operating in the MHz – GHz region, has demonstrated sensitivity at the pT level and has the potential to reach sub-fT sensitivity [201]. Another example that underscores the usefulness of QIS methods for probing exotic spin-dependent interactions is an experiment measuring the interaction between the ground-state spin-1/2 valence electrons of two entangled $^{88}Sr^+$ ions [202]. The coherent cooperative spin dynamics of the pair of $^{88}Sr^+$ ions was restricted to a decoherence-free subspace that was immune to collective magnetic field noise. This allowed the experiment to probe exotic spin-spin interactions between electrons with orders-of-magnitude greater accuracy than achievable in prior measurements [203].

Beyond the intrinsic sensitivity, the principal challenge in experiments searching for exotic spin-dependent interactions is understanding and eliminating systematic errors: clearly distinguishing exotic spin-dependent interactions from mundane effects due to, for example, magnetic interactions. This is a theme in common with the EDM searches discussed in Sec. III, and many similar technical approaches avail themselves. Ideally, the local source of the exotic field can be manipulated in such a way as to modulate its effects, thereby providing a signal with a well-characterized time-dependence that can be distinguished from background. In addition, a variety of independent measurements can be used to monitor, control, and identify systematic errors. Importantly, in searches for exotic spin-dependent potentials, the sought-after effect is not due to a real magnetic field, but rather a *pseudo-magnetic field*. Therefore, by comparing the response of two different systems, effects from magnetic fields can be distinguished from effects due to exotic spin-dependent interactions. This is the essence of *comagnetometry* [204], where the same field, magnetic or otherwise, is simultaneously measured using two different ensembles of atomic or nuclear spins, reviewed in Ref. [6].

Comagnetometers are in fact the most sensitive devices for measuring energy differences between quantum states, in some cases achieving precision at the $\sim 10^{-26}$ eV level [7, 205, 206]. Presently the most sensitive alkali-atom/noble-gas comagnetometers are based on *spin-exchange-relaxation-free* (SERF) atomic magnetometry combined with a scheme where the magnetization of a noble gas species self-compensates the magnetic field, and enabling nearly background-free searches for exotic spin-dependent interactions [24, 207]. Other methods have reached similar sensitivity using a variety of atomic systems via simultaneous measurement of spin-precession in different samples [8, 206].

Presently, comagnetometer technology is limited by effects due to the combination of magnetic field gradients and imperfect sample overlap, atomic collisions, surface interactions that differentially affect the atomic species, and quantum back-action. A number of techniques to circumvent these limitations are being explored. For example, in Ref. [109], quantum control methods are used to average away deleterious effects and precession is measured "in the dark" without external fields applied in order to reduce background effects. In the case of nuclear spin measurements in liquid samples, the problem of magnetic field gradients is overcome in an ensemble of identical molecules by carrying out comagnetometry with different nuclear spins in each identical molecule, suppressing effects of gradients by over an order-of-magnitude as compared to overlapping samples of different atoms/molecules [107, 108].

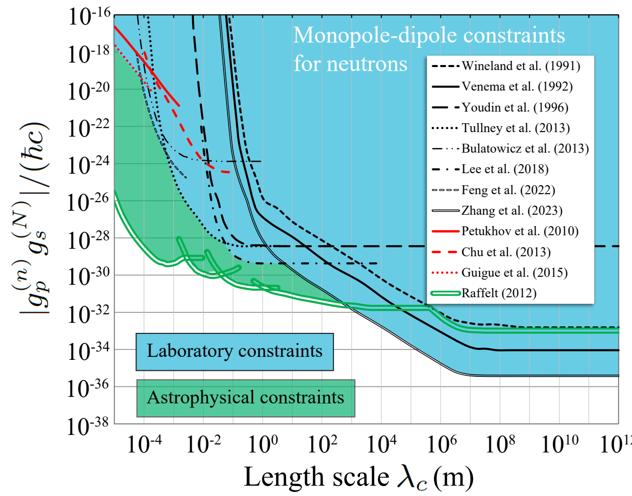

FIG. 2. Constraints on the ALP-mediated monopole-dipole interaction between nucleons and neutrons, $|g_p^{(n)} g_s^{(N)}|/(\hbar c)$, as a function of ALP Compton wavelength (λ_c) as described by Eq. (2), adapted and updated from Refs. [2, 14].[a] Experiments using comagnetometry [208–215] are indicated by black lines, experiments using magnetometry are indicated by red lines [216–218], and astrophysical constraints are indicated by the green double line [219]. Experiments at different length scales measure interaction ranges corresponding to different ALP Compton wavelengths λ_c, and thus different ALP masses m.

[a] Importantly, for the experiments of Venema et al. (1992) [208] and Wineland et al. (1991) [209], a factor of 4π error in the results is corrected in the above plot as compared to the corresponding plots in Refs. [2, 14]. This has the important qualitative consequence that the laboratory experiment of Venema et al. (1992) [208] surpasses astrophysical constraints in the long ALP Compton wavelength (λ_c) limit.

Magnetometer and comagnetometer technology has been applied to a wide variety of experiments searching for new spin-dependent interactions. Experiments using spin-based sensors and spectroscopy have been able to search for interactions with ranges from the nanometer-scale [220–225] to the Earth-scale [208, 209, 215, 226–229], and have probed interactions of protons [220, 224, 229–232], neutrons [7, 208, 209, 214, 215, 231–235], electrons [203, 231, 235–249], and even antimatter [250, 251]. To get an overall idea of the state-of-the-art in experimental methods, a representative survey of the use of spin-based sensor technology in searches for the monopole-dipole interaction described by Eq. (2) for neutron spins [208–218] is shown in Fig. 2. In Fig. 2, constraints on the dimensionless coupling constant $|g_p^{(n)} g_s^{(N)}|/(\hbar c)$ using comagnetometers are indicated by the black lines, and constraints obtained using ^3He magnetometry are indicated by red lines. Parameter space excluded by laboratory experiments is indicated by the light blue shaded region and astrophysical constraints

[219] are shown by the double green lines and green shaded region. It is evident that the best laboratory constraints are obtained using comagnetometry techniques, and these techniques are at the level of precision where for many boson masses they explore parameter space beyond the astrophysical constraints, highlighting the importance of further technological improvements along this research direction. Here again, QIS techniques offer intriguing possibilities: for example, a BEC-based ^{87}Rb comagnetometer, employing the $F = 1$ and $F = 2$ hyperfine manifolds as colocated magnetic sensors, has recently demonstrated significant suppression of magnetic noise and the potential to search for exotic spin-dependent interactions at sub-mm distance scales [252].

If λ_c is at or below the atomic or molecular scale, experimental searches often rely on comparing high-precision measurements to high-accuracy atomic and molecular calculations based on Standard Model physics, as described, for example, in Refs. [159, 221–223, 225, 251]. The idea in these studies is that disagreement between theory and experiment can be interpreted as a possible hint of new physics, while good agreement between theory and experiment can be interpreted as a constraint on new physics scenarios. In these cases, improvements in spectroscopic measurement techniques must be accompanied by similar improvements in calculations: these are examples of measurements where the sensitivity of the method depends on the precision and accuracy of both experiment and theory. Thus there are usually advantages to studying simpler atomic and molecular systems that can be well understood. This is a situation similar in many respects to the long-running program of atomic parity violation measurements and calculations used to test electroweak unification [2], which, of course, can also be used to place bounds on exotic parity-violating interactions [253]. Note also that EDM measurements (Sec. III) can be used to constrain atomic- and molecular-scale symmetry violating interactions [254], and experiments with antimatter open up the possibility of testing if exotic interactions are symmetric with respect to charge-conjugation symmetry [251].

V. SPIN-BASED SENSOR NETWORKS

The searches for exotic spin-dependent interactions mediated by "new" bosons described in Sec. IV employ a local source for the new potential and a spin-based sensor to detect the effects of that potential. Another possibility is that the new bosons can be abundantly generated by astrophysical processes: for example, as dark matter produced in the early universe [255], or through some cataclysmic astrophysical process such as those occurring near black holes [256–258]. In these scenarios, the existence of the new bosons could be directly detected through their interactions with electronic or nuclear spins as reviewed in Ref. [259].

If exotic ultralight bosons ($m \lesssim 1\,\mathrm{eV}/c^2$) such as axions, ALPs, or dark/hidden photons make up the majority of dark matter and have negligible self-interactions, their phenomenology is well-described by a classical field oscillating at the Compton frequency $\omega_c = mc^2/\hbar$. However, due to topology or self-interactions, such ultralight bosonic fields can form stable, macroscopic field configurations in the form of boson stars [260–262] or topological defects (e.g., domain walls, strings, or monopoles [263]). Even in the absence of topological defects or self-interactions, bosonic dark matter fields exhibit stochastic fluctuations [264]. Additionally, as noted above, it is possible that high-energy astrophysical events could produce intense bursts of exotic ultralight bosonic fields [265]. In any of these scenarios, instead of being bathed in a uniform flux, terrestrial detectors witness transient events when ultralight bosonic fields pass through Earth [266].

Such transient phenomena could easily be missed by experimenters when data are averaged over long times to increase the signal-to-noise ratio as is done in the searches described in Secs. III and IV. Detecting such unconventional events presents several challenges. If a transient signal heralding new physics is observed with a single detector, it would be exceedingly difficult to confidently distinguish the exotic-physics signal from the many sources of noise that generally plague precision spin-based sensor measurements. However, if transient interactions occur over a global scale, a network of spin-based sensors geographically distributed around the Earth could search for specific patterns in the timing, amplitude, phase, and polarization of such signals that would be unlikely to occur randomly, as illustrated in Fig. 3. By correlating the readouts of many sensors, local effects can be filtered away and exotic physics could be distinguished from prosaic Standard-Model physics [267–269].

This idea forms the basis for the Global Network of Optical Magnetometers for Exotic physics searches (GNOME), an international collaboration operating spin-based sensors all over the world, specifically targeting beyond-the-Standard-Model physics [25, 270]. The magnetometric sensitivity of each GNOME sensor is $\approx 100\,\mathrm{fT}/\sqrt{\mathrm{Hz}}$ over a bandwidth of $\approx 100\,\mathrm{Hz}$ [270]. Each magnetometer is located within a multi-layer magnetic shield to reduce the influence of magnetic noise and perturbations while still retaining sensitivity to many exotic fields [161]. Even with the magnetic shielding, there are inevitably some transient noise spikes associated with the local environment (and possibly with global effects like the solar wind, changes to the Earth magnetic field, etc.). Therefore, each GNOME sensor uses auxiliary unshielded magnetometers and other sensors (such as accelerometers and gyroscopes) to measure relevant environmental conditions, enabling exclusion/vetoing of data with known systematic issues [270]. The signals from GNOME sensors are recorded with accurate timing provided by the global positioning system (GPS) using a custom GPS-disciplined data acquisition system [271] with temporal resolution $\lesssim 10\,\mathrm{ms}$ (determined by the magnetometer bandwidth), enabling reconstruction of events

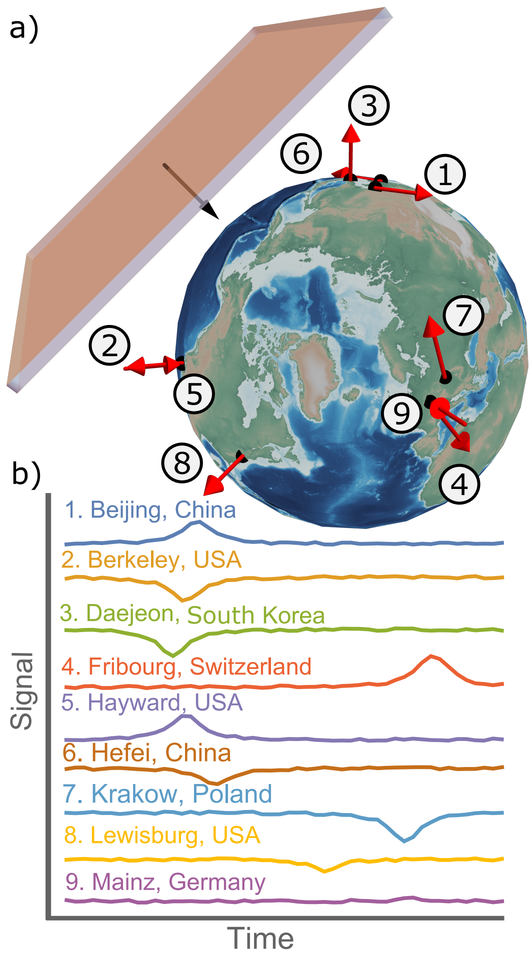

FIG. 3. (a) Schematic representation of a ALP field topological defect (domain wall) passing through the Earth, with the location and sensitive direction of GNOME sensors marked by arrows. (b) As the topological defect passes through various GNOME stations, signals appear in the magnetometer data at particular times. The sign and amplitude of the signals depend on the orientation of the sensor with respect to the domain wall and the atomic species used. Figure from Ref. [268].

that propagate at $\lesssim c$ across the Earth ($R_E/c \approx 40\,\mathrm{ms}$). The broad geographical distribution of sensors enables GNOME to achieve good spatial resolution and act as an "exotic physics telescope" with a baseline comparable to the diameter of the Earth [265].

GNOME searches for a class of signals different from those probed by most other experiments, namely transient and stochastic effects that could arise from ALP fields of astrophysical origin passing through the Earth during a finite time. Depending on the particular hypothesis tested, GNOME is sensitive to ALPs with masses between $\approx 10^{-17}\,\mathrm{eV}$ and $\approx 10^{-9}\,\mathrm{eV}$, and can probe parameter space unconstrained by existing laboratory experiments and astrophysical observations discussed in Sec. IV. A search for ALP domain walls has already been carried out [268, 272], and there are ongoing efforts to search for boson stars [273], carry out intensity interferometry using GNOME to detect stochastic fluctuations of dark matter fields [274], perform multimessenger "exotic physics" astronomy [265], and probe other scenarios [42]. New data analysis efforts and upgrades of GNOME magnetometers to noble gas comagnetometers [275–277] are underway. Most importantly, correlated searches with spin-based sensors offer the possibility to hunt for the unexpected.

Another interesting scenario is the case of kinetically-mixed[4] hidden-photon dark matter. Earth itself may act as a transducer to convert hidden-photon dark matter into a monochromatic oscillating magnetic field[5] at the surface of the Earth [282]. The induced magnetic field from the hidden photons would then have a characteristic global vectorial pattern that can be searched for with unshielded magnetometers dispersed over the surface of the Earth. GNOME is insensitive to such kinetically-mixed hidden-photon dark matter because of the magnetic shields enclosing the magnetometers [161, 281]. Instead, a network of unshielded magnetometers is required. Searches for dark/hidden photons and ALPs using a publicly available dataset from the SuperMAG Collaboration [283, 284] established experimental constraints on such scenarios that are competitive with astrophysical limits [285–287] and the CAST experiment [288] in the probed mass ranges (from around $10^{-18}\,\mathrm{eV}$ to $10^{-16}\,\mathrm{eV}$). A dedicated unshielded magnetometer network targeting hidden photon dark matter may be able to extend the probed parameter space.

There may also be opportunities for QIS techniques to play a key role in next-generation dark-matter searches with quantum sensor networks. As already noted, entanglement and spin-squeezing can increase sensor bandwidth [26, 195, 196], which could expand the range of accessible parameter space. Another intriguing possibility is the use of a network of entangled quantum sensors [289–294].

[4] In models with more than one $U(1)$ gauge symmetry, it is always mathematically possible to make a transformation to new definitions of the associated gauge fields that mix the associated kinetic terms in the Lagrangian [278]. This is the case for models with hidden photons [279], as they result from another $U(1)$ gauge symmetry in addition to the usual one that gives rise to electromagnetism. The practical consequence is that there can be different bases for the eigenstates of interactions as compared to the eigenstates of mass, etc. The situation is analogous to that realized in nature for neutrinos [280]. Neutrinos exhibit mixing because there are different eigenstates for neutrino masses and neutrino flavors (interactions).

[5] The concept of this effect is analogous to that in hidden-photon dark matter experiments carried out using laboratory-scale conducting shields [14, 281]. In this case, the lower atmosphere of the Earth is an insulating gap sandwiched between the conductive interior of the Earth below and ionosphere above. Hidden-photon dark matter drives oscillating currents at the interfaces of the Earth and ionosphere with insulating lower atmosphere (via the kinetic mixing effect), and these surface currents generate a detectable magnetic field.

VI. MAGNETIC RESONANCE SEARCHES FOR ULTRALIGHT BOSONIC DARK MATTER FIELDS

In contrast to some of the scenarios discussed in Sec. V, the simplest assumption for the nature of ultralight ($m \lesssim 1\,\mathrm{eV}/c^2$) bosonic dark matter postulates that the bosons are virialized in the gravitational potential of galaxies such as the Milky Way and manifest as classical fields oscillating at the Compton frequency ω_c. The bosonic dark matter field can cause spin precession via couplings to nuclear and electron spins, and since the field oscillates at a particular frequency the broad and versatile tools of *magnetic resonance* can be used to detect the spin interaction.

An axion (or ALP) field $a(\boldsymbol{r},t)$, which to be dark matter must be nonrelativistic, can be described approximately by

$$a(\boldsymbol{r},t) = a_0 \cos\left(\boldsymbol{k}\cdot\boldsymbol{r} - \omega_c t + \phi_0\right) , \tag{6}$$

where $\boldsymbol{k} \approx m\boldsymbol{v}/\hbar$ is the axion wave vector ($\boldsymbol{v}$ is the relative velocity between the sensor and the field), ϕ_0 is a random phase offset, and a_0 is the average field amplitude, which can be estimated by assuming the average energy of the axion field comprises the totality of the local dark matter energy density $\rho_{dm} \approx 0.4\,\mathrm{GeV/cm^3}$

$$\langle a_0^2 \rangle \approx \frac{2\hbar^2}{c^2}\frac{\rho_{dm}}{m^2} . \tag{7}$$

The axion field has a finite coherence time due to the random kinetic energy of the constituent axions, leading to a broadening of the line shape to a part in $\sim 10^6 \sim c^2/v^2$ as discussed in Refs. [295, 296], as well as stochastic fluctuations of amplitude, phase, and $\boldsymbol{k}$ [264].

The canonical axion of quantum chromodynamics (QCD), a consequence of the Peccei-Quinn mechanism introduced to solve the strong-CP problem [32, 33], naturally couples to the gluon field and generates an oscillating EDM $\boldsymbol{d}_n(t)$ along the nuclear spin orientation $\hat{\boldsymbol{\sigma}}_n$ [43],

$$\boldsymbol{d}_n(t) = g_d a(\boldsymbol{r},t)\hat{\boldsymbol{\sigma}}_n , \tag{8}$$

where g_d is the coupling parameter (inversely proportional to the associated symmetry-breaking scale f_a). Axions can also couple directly to Standard Model spins $\hat{\boldsymbol{\sigma}}$ through the gradient interaction [43], described for nuclear spins by the Hamiltonian

$$\mathcal{H}_g = g_{aNN}\boldsymbol{\nabla} a(\boldsymbol{r},t)\cdot\hat{\boldsymbol{\sigma}}_n , \tag{9}$$

which, in analogy with the Zeeman effect, shows that $\boldsymbol{\nabla} a(\boldsymbol{r},t)$ acts as a pseudo-magnetic field with amplitude B_a:

$$B_a \approx \frac{g_{aNN}}{\hbar\gamma_n}\sqrt{2\hbar^3 v^2 c\rho_{dm}} , \tag{10}$$

where γ_n is the nuclear gyromagnetic ratio. (An analogous situation occurs for other fermions, but characterized by different coupling constants.)

In either case, there appears an oscillating torque on spins due to the axion field. For the axion-gluon (EDM) interaction of Eq. (8) this torque is given by

$$\boldsymbol{\tau}_{\mathrm{EDM}} = \boldsymbol{d}_n(t)\times\boldsymbol{E}^*, \tag{11}$$

where $\boldsymbol{E}^*$ is an effective electric field, which depends on the atomic and nuclear structure of the spin system under study [21]. For the axion-fermion interaction of Eq. (9) this torque is

$$\boldsymbol{\tau}_{\mathrm{grad}} = \boldsymbol{\mu}_n\times\boldsymbol{B}_a(t), \tag{12}$$

where $\boldsymbol{\mu}_n \propto \hat{\boldsymbol{\sigma}}_n$ being the nuclear magnetic moment. Therefore the interaction between an axion dark matter field and nuclear spins is equivalent to that of an oscillating magnetic field as illustrated in Fig. 4, and consequently the tools of magnetic resonance can be used to search for axion dark matter. This is the central concept of the Cosmic Axion Spin Precession Experiment (CASPEr) [13, 21, 298–300].

Nuclear magnetic resonance (NMR) experiments involve measuring nuclear spin dynamics in an applied bias field $\boldsymbol{B}_0$ that determines the Larmor frequency $\Omega_L = \gamma_n B_0$, although $\boldsymbol{B}_0$ can be near zero in zero-to-ultralow field (ZULF) NMR experiments [301] – a technique used in Refs. [299, 300]. In CASPEr, like other dark matter haloscope experiments, the oscillating field is assumed to always be present, corresponding to case of continuous-wave (cw) NMR [302]. The magnetic field is scanned, and if $\Omega_L \approx \omega_c$, a resonance occurs and the spins are tilted away from the direction of $\boldsymbol{B}_0$ and precess at Ω_L, generating a time-dependent magnetization that can be measured, for example, by induction through a pick-up loop or with a SQUID.

The CASPEr experimental program is divided into two branches: CASPEr Electric, which searches for an oscillating EDM $\boldsymbol{d}_n(t)$, and CASPEr Gradient, which searches for an oscillating pseudo-magnetic field $\boldsymbol{B}_a(t)$ [297]. A key to CASPEr's sensitivity is the coherent "amplification" of the effects of the axion dark matter field through a large number of polarized nuclear spins. Therefore an important technological development is the ability to carry out NMR on the largest possible number of spins: this requires large nuclear spin ensembles with high polarization, a focus of CASPEr research efforts, which include thermal polarization, optical polarization, and dynamic nuclear polarization [302]. Another area of focus is optimization of spin ensemble coherence time, making use of quantum control and decoupling schemes [302]. Identifying the optimal spin species and materials with large effective electric fields is especially important for CASPEr Electric, where the detectable signal is proportional to E^*. Optimal atomic systems are heavy (large atomic number Z) and optimal materials have broken inversion symmetry, such as ferroelectric

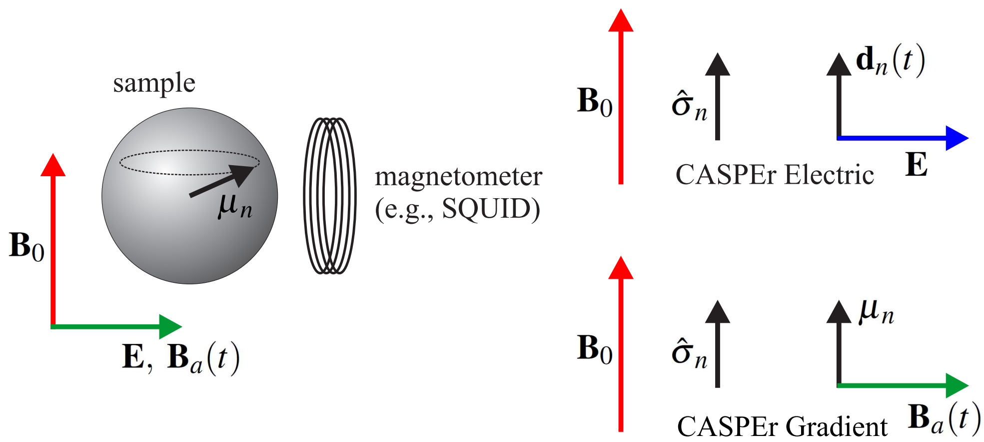

FIG. 4. *Left-hand side*: Schematic diagram of the CASPEr experiment. When the Larmor frequency matches the axion Compton frequency, $\Omega_L \approx \omega_c$, the nuclear spins in the sample are tipped away from their initial orientation along $\boldsymbol{B}_0$ due to the axion-induced torque. The precessing magnetization at Ω_L can be detected with a magnetometer (such as a SQUID) placed near the sample. *Right-hand side*: Experimental geometries for CASPEr Electric (top) and CASPEr Gradient (bottom). In both cases, the nuclear spins $\hat{\boldsymbol{\sigma}}_n$ are oriented along a leading magnetic field $\boldsymbol{B}_0$. An oscillating torque, $\boldsymbol{\tau}_{\rm EDM} = \boldsymbol{d}_n(t) \times \boldsymbol{E}$ in the case of CASPEr Electric and $\boldsymbol{\tau}_{\rm grad} = \boldsymbol{\mu}_n \times \boldsymbol{B}_a(t)$ in the case of CASPEr Gradient, tips the nuclear spins away from $\boldsymbol{B}_0$ if the Larmor frequency Ω_L matches ω_c. Figure adapted from Ref. [297].

solids [21]. Optimizing the coupling of the spin ensemble to the readout sensor that measures its dynamics is yet another area of focus. Quantum back-action effects will eventually limit the sensitivity of NMR experiments to axion dark matter, and therefore back-action evasion techniques will need to be developed for CASPEr experiments approaching fundamental spin projection noise sensitivity limits [302].

The QUAX (QUaerere AXion) experiment [303–305] searches for axion dark matter in a manner similar to CASPEr but by exploiting the interaction of axions with electron spins. The QUAX experiment searches for a coupling of the form (9) but with the nuclear coupling g_{aNN} replaced by the electron coupling g_{aee}, and the electron spin $\hat{\boldsymbol{\sigma}}_e$ playing the role of the nuclear spin $\hat{\boldsymbol{\sigma}}_n$. Ten spherical yttrium iron garnet (YIG) samples are coupled to a cylindrical copper cavity by means of an applied static magnetic field, and the resulting photon-magnon hybrid system acts as an axion-to-electromagnetic field transducer. This transducer is then coupled to a sensitive radiofrequency (RF) detector (a quantum-limited Josephson parametric amplifier). The QUAX experiment is one of the most sensitive RF spin magnetometers ever realized, able to measure fields as small as 5.5×10^{-19} T with nine hours of integration time [305].

Clearly, there is significant overlap between CASPEr and QUAX techniques and those used to search for static EDMs (Sec. III) and exotic spin-dependent interactions (Sec. IV). Indeed, in Refs. [306–311], noble gas comagnetometers, a spin-polarized torsion pendulum, and apparatuses used for EDM experiments were used as spin-based haloscopes to place limits on axion-like dark matter in the low mass range, corresponding to low Compton frequencies. Of note are the development of Floquet masers [312] and spin-amplifiers [313] that may expand the nominal bandwidth of noble gas comagnetometers and enable parallel dark matter searches in different frequency ranges.

The Axion Resonant InterAction Detection Experiment (ARIADNE) experiment [28, 314] is another example of how spin-based sensors can be employed to search for new physics. ARIADNE, like CASPEr and QUAX, aims to use magnetic resonance techniques to search for axions and ALPs, and specifically targets the QCD axion. ARIADNE employs an unpolarized source mass and a spin-polarized ^{3}He low-temperature gas to search for a QCD-axion-mediated spin-dependent interaction: the monopole-dipole coupling described by Eq. (2) and discussed in Sec. IV. In contrast to dark matter haloscopes like CASPEr and QUAX, whose signals depend on the local dark matter density at the Earth, the signal in the ARIADNE experiment does not require axions to constitute dark matter and can be modulated in a controlled way. ARIADNE probes QCD axion masses in the higher end of the traditionally allowed axion window, up to 6 meV, a mass range inaccessible to any other existing experiment. Thus ARIADNE fills an important gap in the search for the QCD axion in this important region of parameter space.

For the QCD axion, the scalar and dipole coupling constants $g_s^{(N)}$ and $g_p^{(N)}$ appearing in Eq. (2) are correlated with the axion mass m. As discussed earlier, the axion-mediated spin-dependent interaction manifests as a pseudo-magnetic field $\boldsymbol{B}_a$. In the ARIADNE experiment, this $\boldsymbol{B}_a$ (if it exists) can be used to resonantly drive spin precession in the laser-polarized cold ^{3}He gas.

This is accomplished by spinning an unpolarized tungsten mass sprocket near the ^{3}He vessel. As the teeth of the sprocket pass by the sample at Ω_L, the magnetization in the longitudinally polarized He gas begins to precess about the axis of an applied field. This precessing transverse magnetization is detected with a SQUID. The ^{3}He sample acts as an amplifier to transduce the small fictitious magnetic field B_a into a larger real magnetic field detectable by the SQUID, similar to the approach of the CASPEr Gradient experiment [297]. Superconducting shielding is needed around the sample to screen it from ordinary magnetic field noise which would otherwise limit the sensitivity of the measurement [315, 316]. The ARIADNE experiment sources the axion field in the lab (like the experiments discussed in Sec. IV, and can explore all mass ranges in the sensitivity band simultaneously, unlike other haloscope experiments which must scan over the possible axion oscillation frequencies ω_c by tuning a magnetic field [13, 21] or cavity [317, 318].

Future prospects for improvements in the search for novel spin dependent interactions could include investigations with a spin polarized source mass, or improved sensitivity with new cryogenic or quantum technologies. Spin squeezing or coherent collective modes in ^{3}He could offer prospects for improved sensitivity beyond the Standard quantum limit of spin projection noise [319], potentially allowing sensitivity all the way down to the SQUID-limited sensitivity. This would allow one to rule out the axion over a wide range of masses, and when combined with other promising techniques [13, 21, 320–322], and existing experiments [317, 318] already at QCD axion sensitivity, could enable a search for the QCD axion over its entire allowed mass range.

VII. SPIN-BASED SENSORS AS DARK MATTER PARTICLE DETECTORS

While Sections IV-VI focus on the use of spin-based sensors to search for axions, bosons, and other new fundamental physics that behaves as a field, spin-based sensors can also be used to search for exotic massive particles. The scattering of dark matter in crystals is a well-developed approach to search for canonical weakly interacting massive particle (or WIMP) dark matter. Searches for WIMP dark matter are soon expected to hit an irreducible background, namely, the coherent scattering of neutrinos from the Sun. This problem is particularly acute for low-mass (a few GeV) WIMPs. There are important scientific reasons to probe WIMP cross-sections below the neutrino floor since such cross-sections are natural in models where the WIMP interacts with Standard Model particles via the Higgs boson. One way to probe the dark matter parameter space below the neutrino floor is to develop detectors that are able to identify the direction of the nuclear recoil caused by the scattering of dark matter. Since the location of the Sun is known, one may veto all scattering events that point away from the Sun, rejecting all events due to solar neutrinos. The dark matter, being relatively isotropic,[6] will induce scattering events in all directions, permitting an unambiguous detection. The key challenge that needs to be overcome to implement this concept is that directional detection needs to be accomplished in a sample with a large enough ($\gtrsim$ ton scale) target mass since the WIMP cross-sections of interest are so small that existing state-of-the-art, ton-scale detectors have so far found nothing. For a practical detector, this requires the ability to perform directional detection in the solid/liquid state so that the detector is sufficiently compact.

This challenge could conceivably be met in a solid state detector via the concept explored in Ref. [44]. The scattering of the dark matter displaces an atom off its lattice location and the displaced atom kicks many other atoms off their locations. This causes a tell-tale damage track, $\sim 10-100\,$nm, in the crystal that points to the direction of the incoming dark matter. The created damage can be measured using techniques established in the fields of solid-state quantum sensing and quantum information processing. The detection concept would utilize conventional localization techniques to identify the location of an event of interest to within $\sim$mm precision. Diffraction limited optics can then be used to achieve micron-scale localization. Optical superresolution or high resolution X-ray-nanoscopy techniques can then be used to measure the damage track at the nanometer scale. One way to accomplish this superresolution imaging is to use NV-center spin spectroscopy in polycrystalline diamond. This technique can also be implemented in a variety of other wide bandgap semiconductors such as divacancies in silicon carbide.

In the near term, work towards such a solid-state, WIMP detector with directional sensitivity is centered around demonstrating the capability to locate and determine the direction of nuclear recoil damage tracks in diamond or other crystals. This requires adaptation and development of existing techniques, but the current state-of-the-art is not far from the requisite sensitivity and resolution [44]. In the medium term, such a detector will require position-sensitive instrumentation with spatial resolution at the millimeter scale, as well as development of crystal-growth techniques to create large volumes of radiopure, structurally homogeneous crystals. With appropriate development, this approach offers a viable path towards directional WIMP detection with sensitivity below the neutrino limit.

Spin-based sensors may also be useful as low-mass dark matter particle detectors. For low-mass dark matter par-

[6] Although it should be noted that because of the relative motion of the Earth with respect to the galactic rest frame, there is expected to be a preferential flux of dark matter from the direction of Cygnus [14]. This is not to mention the possibility of nonvirialized streams of dark matter that could also produce preferential directions of dark matter flux [323].

ticles, not only are interactions rare because of the exceedingly small cross-sections but also the deposited energy in the detector is extremely small, so both high sensitivity and low background are required. In Ref. [324], a new method for detecting low-mass dark matter particles is proposed. The idea is that if a dark matter particle deposits a small amount of energy ($\gtrsim 1\,\mathrm{meV}$) into a high-quality crystalline solid, that energy will eventually be converted into ballistic phonons travelling to the crystal surface. If the crystal surface is covered by a van der Waals liquid helium film, the phonons can cause quantum evaporation of He atoms. At low temperature (below $\sim 100\,\mathrm{mK}$) ^{3}He atoms in liquid helium reside at the surface in Andreev bound states [325]. After being evaporated, the ^{3}He atoms can be collected on another surface covered with a van der Waals film of isotopically enriched ^{4}He. The ^{3}He atoms can be localized at mK temperatures to bound electron states on this second helium film [326], and subsequently detected by sensing their magnetic moments, by measuring, for example, decoherence of electron spin qubits [327]. This methodology opens the possibility of single ^{3}He atom detection and dark matter particle detection at the $\sim 1\,\mathrm{meV}$ scale [324].

VIII. CONCLUSION

Much of what is now known about the structure and composition of molecules and materials was originally revealed through spin-based measurements such as nuclear magnetic resonance and electron spin resonance. As QIS continues to advance the level of control over spin systems, new opportunities are emerging to use the same techniques to search for new fundamental physics in a parallel and complementary manner to large-scale particle accelerators and direct particle detectors. There are a range of spin-based experiments that can be employed to search for a variety of effects. Searches for permanent electric dipole moments with atoms, molecules, and spins in solids can probe for symmetry violations and thereby test possible explanations for the matter-antimatter imbalance in the universe. Spin-based magnetometers and global networks of such detectors can search for and discover or constrain the parameter space for new particles and fields. Spins in solids can also serve as novel particle detectors by using them as *in-situ* probes for the signatures left behind from particle impacts, and ^{3}He spins evaporated from liquid helium films on crystal surfaces could be used as low-mass dark matter particle detectors. While many such efforts are already underway, there remain tremendous opportunities for innovations in spin-based quantum sensors that will enhance their sensitivity, accuracy, and range of potential fundamental physics targets.

ACKNOWLEDGMENTS

This work is adapted from a white paper entitled "Quantum Sensors for High Precision Measurements of Spin-dependent Interactions" for the U.S. particle physics community's "Snowmass" planning process [328]. Important contributions to the writing of the original white paper were made by Surjeet Rajendran and Thomas Cecil, to whom we are deeply indebted. The authors appreciate the insightful comments and suggestions on the manuscript from Nicolò Crescini, Reuben Shuker, Andrew Jayich, and Morgan Mitchell. The authors are also sincerely grateful to Dong Sheng, Shaobo Zhang, and Zheng-Tian Lu for bringing the error in previously published versions of Fig. 2 to our attention. The work of DFJK was supported by the U.S. National Science Foundation under grant PHYS-2110388. The work of DB was supported in part by the Cluster of Excellence "Precision Physics, Fundamental Interactions, and Structure of Matter" (PRISMA+ EXC 2118/1) funded by the German Research Foundation (DFG) within the German Excellence Strategy (Project ID 39083149) and also in part upon work from COST Action COSMIC WISPers CA21106, supported by COST (European Cooperation in Science and Technology).

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