Dynamical Axions in $U(1)$ Quantum Spin Liquids

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Since their proposal nearly half a century ago, physicists have sought axions in both high energy and condensed matter settings. Despite intense and growing efforts, to date, experimental success has been limited, with the most prominent results arising in the context of topological insulators. Here, we propose a novel mechanism whereby axions can be realized in quantum spin liquids. We discuss the necessary symmetry requirements and identify possible experimental realizations in candidate pyrochlore materials. In this context, the axions couple to both the external and the emergent electromagnetic fields. We show that the interaction between the axion and the emergent photon leads to a characteristic dynamical response, which can be measured experimentally in inelastic neutron scattering. This Letter sets the stage for studying axion electrodynamics in the highly tunable setting of frustrated magnets.

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Quantum spin liquids (QSLs) are long-range entangled phases of matter with fractionalized spins and emergent gauge fields [1–3]. One of the most fascinating examples is a $U(1)$ QSL, where the gauge structure resembles quantum electrodynamics [4,5]. Vastly different from our universe, this emergent quantum electrodynamics violates Lorentz symmetry, is strongly coupled, and contains magnetic monopoles. Such $U(1)$ QSLs have attracted substantial interest due to proposed experimental realizations [6–8] in a class of frustrated quantum rare-earth pyrochlore magnets called quantum spin ice (QSI) [9–15].

The long-wavelength effective description of a $U(1)$ QSL in $3+1$D with only gapped matter is in terms of an emergent $U(1)$ gauge field whose quanta are photons, electric gauge charges corresponding to fractionalized spin excitations (spinons), and dual magnetic monopoles (visons) [16]. At energies below the matter gap, the universal properties of $U(1)$ QSL phases stem from their emergent photons governed by the Maxwell Lagrangian $\mathcal{L}_\gamma = (|\boldsymbol{e}|^2 - |\boldsymbol{b}|^2)/2$, where $\boldsymbol{e}$ and $\boldsymbol{b}$ are emergent electric and magnetic fields [see Supplemental Material (SM) [17] for our choice of electromagnetic units].

A tantalizing modification to the Maxwell Lagrangian that has been entertained both in high energy and condensed matter physics is the axion term $\mathcal{L}_{\varphi\gamma} = \alpha\varphi(\boldsymbol{e}\cdot\boldsymbol{b})/\pi$ [18], where $\alpha = e^2/(4\pi c)$ is the fine structure constant (Here, c is the "speed of light" and e is the elementary "electric charge," both associated with the emergent gauge structure.) [19] and φ is a real pseudoscalar called the axion field. In high-energy physics, axions are considered one of the best motivated particles beyond the standard model, acting as a remedy to the strong CP problem [20], naturally arising in string theory [21], and playing a role as a possible dark-matter candidate [22]. Despite decades of intensive experimental efforts, such axions have not been observed [23].

Axions can also emerge as collective excitations in condensed matter systems [18,24]. Indeed, the fluctuations of any (anti)ferromagnetic ordering with a pseudoscalar order parameter couple as a dynamical axion field at long wavelengths. In the context of topological insulators, a spacetime constant φ (in which case, the axion term is called a θ term) plays an important role in the electromagnetic response [25–27], and the influence of dynamical axionic fluctuations on the external electromagnetic fields has been studied in considerable detail [28–31]. Additionally, they have been discussed in the context of topological superconductors [32] as well as Weyl semimetals [33,34].

Here, we investigate the effects of an emergent dynamical axion in $U(1)$ QSLs. This scenario has received limited attention in the literature and remains poorly understood beyond some work on the effect of θ terms [35–38]. As in topological insulators, the dynamical axion we study couples to external electromagnetic fields. However, it also couples to the emergent electrodynamics, giving rise to a vacuum with elementary photons and axions that has been hitherto inaccessible in other contexts. We study this internal axion electrodynamics by investigating the dynamical structure factor and show how it leads to prominent signatures accessible through inelastic neutron scattering. Namely, we find a characteristic two-particle continuum associated with the threshold production of axion-photon pairs. We further discuss possible experimental realizations in QSI on the breathing pyrochlore lattice, where an appropriate magnetic order interacts with the emergent gauge field as an axion field, see Fig. 1. This identifies breathing pyrochlore QSL candidates, like

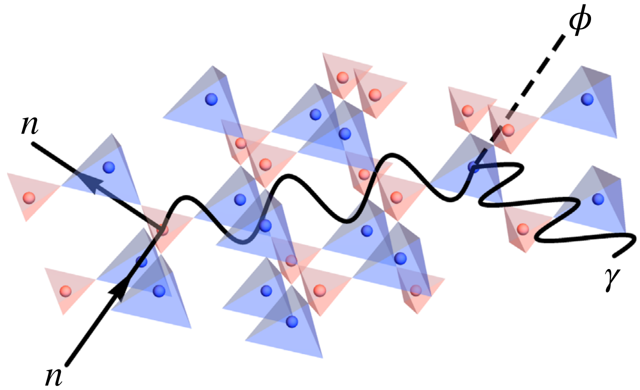

FIG. 1. In the breathing pyrochlore lattice, the A (red) and B (blue) tetrahedra have different volumes, breaking inversion symmetry. A staggered spinon charge density, with positive (red sphere) and negative (blue sphere) spinons on A and B tetrahedra, respectively, breaks time reversal [40]. The combined order couples as an axion to the internal electrodynamics. The embedded Feynman diagram illustrates one of the neutron scattering processes brought about by photon-axion pair production.

$Ba_3Yb_2Zn_5O_{11}$ [39], as a possible condensed-matter realization of emergent axion electrodynamics.

Low-energy effective action.—$U(1)$ QSLs are described by the deconfined phase of $U(1)$ lattice gauge theory [41]. This is well established in microscopic spin models [6,42–44]. Regularizing the axion term on a lattice is a long-standing open problem in lattice gauge theory, and current proposals involve complicated, beyond nearest-neighbor interactions [45–48].

We hereby leave these technicalities, which do not affect the universal physics, for future work, and we focus, instead, on coarse-graining the microscopic degrees of freedom and writing down the leading symmetry-allowed terms in a long-wavelength effective action [17]. We separate the axion field into a vacuum expectation value and fluctuations: $\varphi(\boldsymbol{x},t) \equiv \theta + \phi(\boldsymbol{x},t)$. This leads to the low-energy effective action $S_{\mathrm{eff}} = \int dt d^3\boldsymbol{x}(\mathcal{L}_\gamma + \mathcal{L}_\phi + \mathcal{L}_{\varphi\gamma})$, where

$$\mathcal{L}_\gamma = \frac{1}{2}(|\boldsymbol{e}|^2 - |\boldsymbol{b}|^2), \tag{1a}$$

$$\mathcal{L}_\phi = \frac{J}{2}[(\partial_t\phi)^2 - v^2|\nabla\phi|^2 - \Delta^2\phi^2], \tag{1b}$$

$$\mathcal{L}_{\varphi\gamma} = \frac{\alpha}{\pi}(\theta + \phi)\boldsymbol{e}\cdot\boldsymbol{b}, \tag{1c}$$

(See SM [17] for additional discussion of S_{eff}'s construction). $\mathcal{L}_\gamma$ and $\mathcal{L}_{\varphi\gamma}$ are the Maxwell and axion terms, governing the emergent electric and magnetic fields $\boldsymbol{e}(\boldsymbol{x},t)$ and $\boldsymbol{b}(\boldsymbol{x},t)$ with $\alpha \sim 1/10$ the emergent fine structure constant [19]. $\mathcal{L}_\phi$ is the free theory of φ, where the phenomenological parameters J, v, and Δ correspond to the axion's stiffness, asymptotic speed, and gap, respectively. Except when $v = c$, the emergent speed of light, the axion sector of the theory violates the emergent Lorentz invariance.

In this Letter, we do not consider effects from the dynamical gapped electric gauge charges (spinons) and magnetic monopoles (visons) in S_{eff}. Futhermore, S_{eff} could include terms with external electric and magnetic fields $\boldsymbol{E}(\boldsymbol{x},t)$ and $\boldsymbol{B}(\boldsymbol{x},t)$ interacting with φ (i.e., $\mathcal{L}_{\mathrm{ext}} \sim \varphi\boldsymbol{E}\cdot\boldsymbol{B}$). Here, we focus on how the axion interacts with the emergent electromagnetic fields, Eq. (1c), as this physics is new to the QSL context. We briefly return to the coupling to external electromagnetic fields at the end of this Letter.

It is well known that the θ term is a total derivative and does not affect the dynamical equations of motion in the bulk. However, the presence of the axion term modifies the emergent Gauss's law as a Gauss-Witten law [49]

$$\nabla\cdot\boldsymbol{e} = -\frac{\alpha}{\pi}\nabla\cdot(\varphi\boldsymbol{b}). \tag{2}$$

This reveals the Witten effect, where the axion field causes magnetic monopoles to accumulate electric charge [50]. For our purposes, it will also be important in identifying the degrees of freedom to which external probe-fields couple.

Axions in quantum spin ice.—Before turning to our results on the dynamics of axion-photon production, we contextualize them to the case of QSI. This is both because QSI has become a familiar model system for $U(1)$ QSLs [6–8], and also, because of its direct experimental relevance [9–15].

We need to identify an experimentally motivated order parameter, φ, that interacts with the emergent photon as an axion. To be consistent with the field-theoretic modeling above, upon coarse graining, φ must be a local time-reversal odd pseudoscalar to linearly couple with $\boldsymbol{e}\cdot\boldsymbol{b}$.

It is tempting to consider a common magnetic order parameter in pyrochlore systems [51], namely,

$$Q_t = (-1)^t \mathrm{div}_t S^{(z)}. \tag{3}$$

Here, t labels tetrahedra on the pyrochlore lattice and $(-1)^t = \pm 1$ depending on the sublattice (A or B). $\mathrm{div}_t S^{(z)}$ is the lattice divergence of $S^{(z)}$—the component of the spin along the local easy axis—which yields the spinon charge at t. Evidently, Q_t is an order parameter for a staggered background of spinons, which corresponds to the spins developing all-in-all-out order and spontaneously breaking time-reversal symmetry [40]. However, Q_t is even under inversion and does not have the correct symmetry properties.

Therefore, we are brought to consider the breathing pyrochlore lattice, where A and B tetrahedra are of unequal size and, thus, no longer relate under inversion (see Fig. 1). For small enough breathing anisotropy, the $U(1)$ QSL phase of QSI remains stable [52]. The breathing anisotropy only modifies the microscopic ring-exchange scale, changing the value of the internal speed of light but not the stability of the effective field theory.

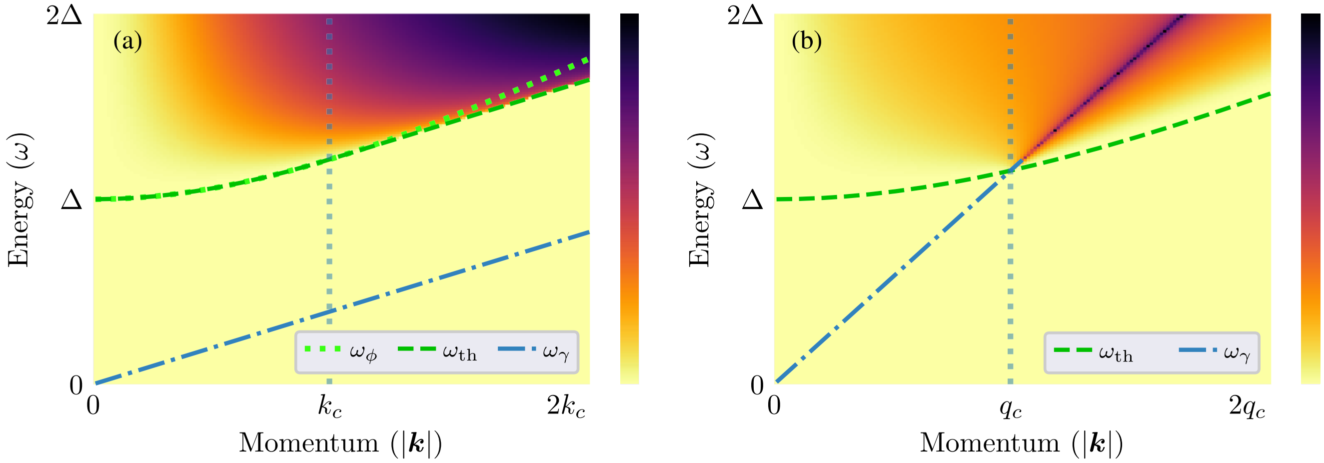

FIG. 2. The dynamical structure factor $\mathcal{F}^{aa}(\boldsymbol{k},\omega)$ exhibits a two-particle continuum from axion-photon pair production above a threshold $\omega_{\rm th}$ [green dashed line, see Eq. (7)]. Below threshold, the delta function response due to single photon states is indicated by ω_γ (blue dash-dotted line). Depending on the kinematics of the axion dispersion, either the axion can travel faster than the speed of light (a) or not (b). In the former case, the critical momentum beyond which the axion would travel faster than the speed of light is k_c. In the latter case, the single-photon dispersion enters the photon-axion continuum above momentum q_c, leading to a resonance centered at ω_γ (vertical dotted lines in the respective panels).

Breathing anisotropy is characterized by the lattice order parameter

$$\Delta V_l = a^{-3}(V_l^{(A)} - V_l^{(B)}), \quad (4)$$

where, $V_l^{(A)}$ ($V_l^{(B)}$) is the volume of the A (B) tetrahedron with corner at site l, and a is the lattice constant. Evidently, ΔV is a time-reversal even real pseudoscalar, and the product $Q\Delta V$ finally has the correct symmetry. Coarse-graining and decomposing the order parameters into their vacuum expectation values and amplitude fluctuations, we obtain an axion field

$$\varphi(\boldsymbol{x},t) \equiv \theta + \phi(\boldsymbol{x},t) \sim \langle\Delta V\rangle\langle Q\rangle + \langle\Delta V\rangle\delta Q(\boldsymbol{x},t). \quad (5)$$

Here, we have neglected fluctuations of ΔV since lattice distortions generally are higher energy than the magnetic fluctuations.

The dynamics of QSI at energies below the spinon gap is governed by a six-spin ring-exchange term [6]. Since ϕ and e arise from single-body operators while b arises from a six-body operator, the lattice axion term is an eight-body subleading ring-exchange term. Given that a six-body term is important to the dynamics of the QSL phase, an eight-body term may be subleading, but not negligible.

Axion-photon production.—To probe the coupled dynamics of the axion-photon system, we study the dynamical structure factor (DSF) of the coarse-grained spin magnetic moment $\boldsymbol{S}(\boldsymbol{x},t)$. This can be measured by neutron scattering and is often used to probe candidate spin liquids [9,12,14,15,53]. The DSF $\mathcal{F}^{ij}(\boldsymbol{k},\omega)$ is given by the imaginary part of the spin susceptibility associated with the correlation function $\langle S^i(\boldsymbol{k},\omega)S^j(-\boldsymbol{k},-\omega)\rangle$, where $i,j=x,y,z$.

The most interesting contribution to the DSF within the low-energy effective theory, Eq. (1), arises from the threshold production of axion-photon pairs. While we do not model spinon-axion interactions, we note that they could modify the DSF in the spin-flip channel and cause an additional multiparticle continuum [54,55].

To compute the DSF, we must connect $\boldsymbol{S}(\boldsymbol{x},t)$ to the degrees of freedom present in the effective field theory. Below the spinon gap, physical states satisfy the divergence-free condition $\boldsymbol{\nabla}\cdot\boldsymbol{S}=0$. Comparing this to the Gauss-Witten law, Eq. (2), up to a nonuniversal constant, $\boldsymbol{S}(\boldsymbol{x},t)$ is, therefore, given by

$$\boldsymbol{S}(\boldsymbol{x},t) = \boldsymbol{e}(\boldsymbol{x},t) + \frac{\alpha}{\pi}\varphi(\boldsymbol{x},t)\boldsymbol{b}(\boldsymbol{x},t). \quad (6)$$

Equation (6) can be further confirmed by noting that the emergent vector potential and spin magnetic moment are canonically conjugate [6,56,57]. Identifying the coarse-grained $\boldsymbol{S}(\boldsymbol{x},t)$ as the canonical momentum of the effective field theory, the variation $\delta S_{\rm eff}/\delta\boldsymbol{e}$ again yields Eq. (6).

With this identification, the DSF $\mathcal{F}^{ij}(\boldsymbol{k},\omega)$ must be transverse. Solving the Schwinger-Dyson equation to leading loop order, we find (see SM [17]),

$$\mathcal{F}^{ij} = \left(\delta^{ij} - \frac{k^i k^j}{k^2}\right)\begin{cases}\mathcal{F}_\gamma(\boldsymbol{k},\omega)\delta(\omega - c|\boldsymbol{k}|) & \omega < \omega_{\rm th}(\boldsymbol{k}) \\ \mathcal{F}_{\phi\gamma}(\boldsymbol{k},\omega) & \omega > \omega_{\rm th}(\boldsymbol{k})\end{cases}. \quad (7)$$

Here, $\omega_{\rm th}$ indicates the threshold energy above which axion-photon pairs can be produced at total momentum $\boldsymbol{k}$. The associated two-particle continuum is represented by

$\mathcal{F}_{\phi\gamma}$, which in the absence of the emergent axion vanishes. Below the threshold, only single photon states are available and the DSF contains a delta function reflecting the photon dispersion, weighted by the function $\mathcal{F}_{\gamma} = (\pi + \alpha^2\theta^2/\pi)\omega$. This reduces to the well known result for standard $U(1)$ QSLs when $\theta = 0$ [56,58].

In general, the effective theory is not Lorentz invariant unless the asymptotic velocity of the axion field, v, equals the speed of light, c. Thus, there are two distinct kinematic regimes for axion-photon production corresponding to $v \geq c$ and $v < c$, as illustrated in Figs. 2(a) and 2(b), respectively. In the case $v \geq c$, there is a critical momentum $k_c = \Delta c/(v\sqrt{v^2 - c^2})$ beyond which the axion travels faster than c. This leads to the threshold

$$\omega_{\text{th}} = \begin{cases} \omega_{\phi}(\boldsymbol{k}) & |\boldsymbol{k}| < k_c \\ \omega_{\gamma}(|\boldsymbol{k}| - k_c) + \omega_{\phi}(k_c) & |\boldsymbol{k}| \geq k_c \end{cases}, \tag{8}$$

where $\omega_{\phi}(|\boldsymbol{k}|) = \sqrt{\Delta^2 + v^2|\boldsymbol{k}|^2}$ and $\omega_{\gamma}(|\boldsymbol{k}|) = c|\boldsymbol{k}|$ are the axion and photon dispersions, respectively. Physically, for external momenta less than k_c, all of the momentum is carried by the axion at threshold. For greater momenta, the axion travels at c and the rest of the momentum is carried by the photon. While the full functional form of $\mathcal{F}_{\phi\gamma}$ is cumbersome (see SM [17]), the behavior just above threshold is governed by characteristic exponents,

$$\mathcal{F}_{\phi\gamma}(\boldsymbol{k}, \omega) \sim \begin{cases} (\omega - \omega_{\text{th}})^3 & |\boldsymbol{k}| \leq k_c \\ \sqrt{\omega - \omega_{\text{th}}} & |\boldsymbol{k}| > k_c \end{cases}. \tag{9}$$

See Fig. 3(a) for frequency dependent line cuts of the full DSF illustrating these two behaviors. Notice that the naive expectation, based on the density of axion-photon states, goes as $(\omega - \omega_{\text{th}})^2$ for $|\boldsymbol{k}| < k_c$ and $\sqrt{\omega - \omega_{\text{th}}}$ for $|\boldsymbol{k}| > k_c$. The difference can be traced to the momentum dependence of the interaction.

In the case that $v < c$, the axion never reaches the speed of light so $\omega_{\text{th}} = \omega_{\phi}$. However, the single-photon dispersion crosses above ω_{th} for momenta greater than $q_c = \Delta/\sqrt{c^2 - v^2}$. In this regime, a single photon can decay into an axion-photon pair and the DSF develops a resonant peak above threshold, see Fig. 3(b). The Fano-like asymmetry [59] of the resonant line shape can be traced to the interference between the $\boldsymbol{e}$ operator in Eq. (6) creating a single-photon state which decays into an axion-photon pair and the $\varphi\boldsymbol{b}$ operator directly producing such a pair.

The experimental signatures of the axion discussed, thus far, are from interactions with photons. It is interesting to consider whether a dynamical axion field can be detected through inelastic neutron scattering in the absence of an emergent electrodynamics. In this case, there are no long-wavelength pseudovectors at the Γ point to couple to the neutron spin. Returning to the example of breathing pyrochlore, this indicates that fluctuations of Q remain hidden to neutrons unless there is coexisting $U(1)$ fractionalization.

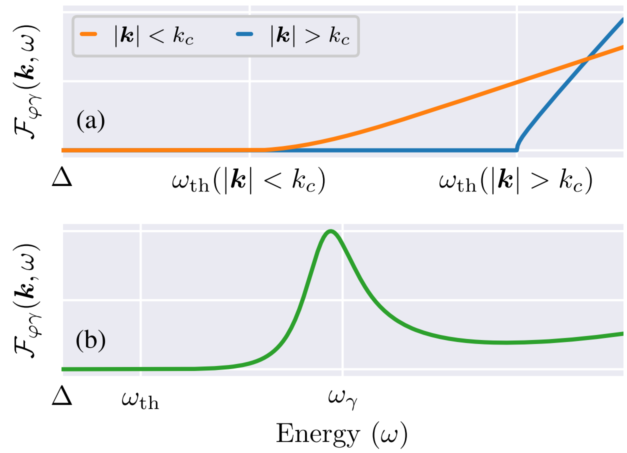

FIG. 3. Line cuts of the dynamical structure factor $\mathcal{F}_{\phi\gamma}(\boldsymbol{k}, \omega)$ at fixed momentum $|\boldsymbol{k}|$ illustrating, for different kinematic regimes, (a) the turn-on of the axion-photon continuum above threshold and (b) the single photon resonance on top of the continuum. For convenience, we have chosen parameters that produce a broad resonance with visible Fano asymmetry on the scale of the background.

Coupling to external electromagnetic fields.—On symmetry grounds, the order parameter $\phi \sim Q\Delta V$ couples to the external electromagnetic field, leading to a magnetoelectric polarizability in linear response. Since the charge gap is large in these materials, the response of the conduction and valence bands to the magnetic ordering should be weak [25], producing a small electronic contribution to the response. Rather, we expect a significant contribution would rely on the Khomskii polarization mechanism [60], wherein an electric dipole moment arises in tetrahedra that host spinon excitations pointing in the direction of the minority spin (For example, the minority spin of a three-in-one-out tetrahedron is the single outward-pointing spin.). In nonbreathing pyrochlores, this cancels for adjacent spinon-antispinon pairs; with breathing, it need not. In the absence of an applied magnetic field, the staggered Q order parameter corresponds to a spinon-antispinon zinc-blende crystal with no preferred orientation for the minority spins, and the associated Khomskii polarization cancels [61]. An applied magnetic field would make certain spins preferentially "minority" for the existing zinc-blende crystal, which, in turn, leads to a linear electric polarization. A quantitative study of this microscopic mechanism is beyond the scope of the present Letter.

Discussion.—We studied the dynamical interplay of axion and gauge fields in the context of quantum spin liquids, where they both appear as emergent phenomena. We identified a microscopic order parameter with the right symmetry to give rise to an emergent axion field. This order is natural in quantum spin ice systems on breathing pyrochlore lattices [52], such as the QSL candidate $Ba_3Yb_2Zn_5O_{11}$ [39,62–64]. The two main ingredients are the breaking of lattice inversion symmetry coexisting with low energy fluctuations of a scalar antiferromagnetic order parameter Q. From Eq. (5), a finite θ parameter requires $\langle Q \rangle \neq 0$. Classically, such an order can coexist with Coulomb liquid behavior, for example, in the fragmented phase of spin ice [65,66], which has been recently observed in iridate materials [67–69]. Our long-wavelength symmetry analysis suggests that the quantum limit of a fragmented spin liquid on breathing pyrochlore would be a $U(1)$ QSL phase with both a θ term and a dynamical axion. Notice, however, that the dynamical part of the axion, ϕ, comes from the amplitude fluctuations of Q, and, therefore, is present regardless of $\langle Q \rangle$.

Unlike the axion electrodynamics studied in topological insulators and superconductors, and in Weyl semimetals [28–30,32–34], the quasiparticle content of $U(1)$ QSLs includes magnetic monopoles. In the presence of a finite θ term, the celebrated Witten effect induces the monopoles to acquire an electric charge, becoming dyons, which, in turn, leads to possible changes in the thermodynamic behavior of the system. For instance, condensation of dyons can lead to distinct symmetry patterns in the neighboring ordered phases [36,45,48].

Here, we focused, instead, on a dynamical axion, which modifies the inelastic response and may be observed directly in neutron scattering. Using a long-wavelength description, we investigated the behavior of the dynamical structure factor and demonstrated how it is qualitatively modified by the axion-photon continuum (see Fig. 2). Our results provide an avenue for observing signatures of emergent axion electrodynamics in a class of frustrated magnetic systems. These signatures crucially depend on the presence of the underlying $U(1)$ gauge structure and, therefore, also provide direct evidence of QSL behavior.

Dynamical axions in our universe were first hypothesized over 40 years ago as a remedy for the strong *CP* problem in the standard model [70]. Today, they are the subject of intense experimental searches as candidates for dark matter [22], yet to be observed [23]. In condensed matter systems, while pseudoscalar collective modes are not so elusive, their coupling to a fully internal electrodynamics is not readily available. In this Letter, we demonstrated how one can realize and access an emergent axion electrodynamics in the context of quantum spin liquids. Speculatively, this provides a test bed for high-energy physics that cannot currently be probed directly due to limitations of experiments or, more dramatically, limitations in the content of the universe itself.

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[16] The terminology for $U(1)$ QSL excitations differs among communities. We denote the spinon as the electric gauge charge, sourcing emergent electric fields. Spinons are referred to as magnetic monopoles in the spin ice literature, as they source real magnetization fields. The excitations we call magnetic monopoles source emergent magnetic fields and undergo the Witten effect. In spin ice, they are often referred to as visons.
[17] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.130.076701 for a discussion of (i) the electromagnetic units used throughout the manuscript, (ii) the local easy axes of the (breathing) pyrochlore lattice in quantum spin ice, (iii) the symmetry

properties of the emergent gauge field and axion, and (iv) the calculation of the dynamical structure factor.
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