Spinon Magnetic Resonance of Quantum Spin Liquids

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We describe electron spin resonance in a quantum spin liquid with significant spin-orbit coupling. We find that the resonance directly probes spinon continuum, which makes it an efficient and informative probe of exotic excitations of the spin liquid. Specifically, we consider spinon resonance of three different spinon mean-field Hamiltonians, obtained with the help of projective symmetry group analysis, which model a putative quantum spin liquid state of the triangular rare-earth antiferromagnet $YbMgGaO_4$. The band of absorption is found to be very broad and exhibit strong van Hove singularities of single spinon spectrum as well as pronounced polarization dependence.

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Introduction.—Electron spin resonance (ESR) and its variants in magnetically ordered systems—ferromagnetic and antiferromagnetic resonances—represent one of the most precise and frequently used spectroscopic probes of excitations of magnetic media. The essence of the magnetic resonance technique consists of measuring absorption of electromagnetic radiation (usually in the microwave range of frequencies) by a sample material that is (typically) subjected to an external static magnetic field. The absorption is caused by coupling of magnetic degrees of freedom to the magnetic field of the electromagnetic wave. Given the very large wavelength of the microwave, the ESR absorption is driven by zero wave vector ($\mathbf{q} = 0$ or vertical) transitions between states with different S^z projections of magnetic dipole moment on the direction perpendicular to the magnetic field of the electromagnetic (EM) radiation.

In a spin system with isotropic exchange, the absorption spectrum of an ac magnetic field is a δ-function peak at the frequency equal to that of the Zeeman energy, independent of the exchange interaction strength. This is a consequence of the fact that, at $\mathbf{q} = 0$, EM radiation couples to the total magnetic moment, which for an SU(2) invariant system commutes with the Hamiltonian [1]. Therefore, any deviation of the absorption spectrum from the δ-function shape implies violation of the spin-rotation symmetry, caused either by anisotropic terms in the Hamiltonian (explicit symmetry breaking) or by the development of long-range magnetic order below the critical temperature (spontaneous symmetry breaking). This is the key reason for ESR's utility.

The goal of our work is to explore applications of ESR to a highly entangled phase of magnetic matter—the quantum spin liquid (QSL) [2]. This intriguing novel quantum state manifests itself via nonlocal elementary excitations—spinons—which behave as fractions of ordinary spin waves. The local spin operator becomes a composite of two or more spinons, which immediately implies that dynamic spin susceptibility measures the multispinon continuum. In principle, the best probe of the spinon continuum is provided by inelastic neutron scattering, which probes spinons at finite wave vector $\mathbf{q}$ and frequency ω. By now, several textbook-quality experiments have provided us with unambiguous signatures of multiparticle continua [3–5]. In practice, however, such state of the art measurements require large high-quality single crystals, which quite frequently are not available.

We posit here that ESR, with its exceptionally high-energy resolution, represents an appealing complimentary spectroscopic probe of spinons—spinon magnetic resonance (SMR). The key requirement for turning it into a full-fledged probe of spinon dynamics consists of the absence of spin-rotational invariance. This requirement stems from the aforementioned "insensitivity" of ESR to the details of excitations spectra in SU(2) invariant magnetic materials. Note that the SU(2) invariance is, at best, a theoretical approximation to the real world materials which always suffer from some kind of magnetic anisotropy.

Moreover, over the past 15 years, the field of QSL has evolved dramatically away from the spin-rotational invariance requirement explicit in many foundational papers [6–8]. The absence of spin-rotational invariance has evolved from the "real world" annoyance to the virtue [2,9,10]. Indeed, the first and still the most direct and unambiguous demonstration of the gapless QSL phase came from Kitaev's exact solution of the fully anisotropic honeycomb lattice model [11], which does not conserve total spin.

Importantly, a large number of very interesting and not yet understood materials, such as α-$RuCl_3$ [12], $YbMgGaO_4$ [13,14], $Yb_2Ti_2O_7$ [15,16], and many other pyrochlores [17], and even organic BEDT-TTF and BEDT-TSF salts [18], showing promising QSL-like features are known to possess significant spin-orbit interaction and are described by spin Hamiltonians with significant asymmetric exchange and pseudodipolar terms. It is precisely this class of low-symmetry spin models we focus on in the present study.

We illustrate our idea by considering a spin-liquid state proposed to describe a spin-orbit-coupled triangular lattice Mott insulator $YbMgGaO_4$. The appropriate spin Hamiltonian has been argued to be that of an XXZ model with interactions between nearest (with $J \sim 1$ K) and next-nearest neighbors on the triangular lattice, together with a pseudodipolar term [19–21] of $J_{\pm\pm}$ kind in notations of [15], between nearest neighbors ($J_{\pm\pm} \sim 0.2$ K). Most recently, polarized neutron scattering data were interpreted in favor of significant $J_{z\pm}$ interaction [22]. This Hamiltonian does not conserve total spin $\mathbf{S}_{\rm tot}$.

Inelastic neutron scattering experiments reveal broad spin excitations continuum [23,24], consistent with fractionalized QSL with spinon Fermi surface. At the same time, experimental evidence of significant disorder effects [21,24–26], capable of masking "pristine" physics of the material, is mounting.

Our goal here is to add to the ongoing discussion on the nature of the ground state of $YbMgGaO_4$ by pointing out that ESR can serve as a very useful probe of QSL with significant built-in spin-orbit interactions. We therefore accept spin-liquid hypothesis and focus on fermionic U(1) symmetric spin-liquid ground states, proposed for this material previously [23,27]. We rely on the well-established projective symmetry group (PSG) analysis of possible U(1) spin liquids [27–31]. The spin-orbital nature of the effective spin-$1/2$ local moment of the Yb^{3+} ion implies that, under the space group symmetry operations, both the direction and the position of the local spin are transformed. The symmetry operations include translations $T_{1,2}$ along the major axis $\mathbf{a}_{1,2}$ of the crystal lattice, a rotation C_2 by π around the in-plane vector $\mathbf{a}_1 + \mathbf{a}_2$, a counterclockwise rotation C_3 by $2\pi/3$ around the lattice site, and the (three-dimensional) inversion I about the lattice site. Following [27], it is convenient to combine C_3 and I operations into a composite one $\bar{C}_6 \equiv C_3^{-1} I$. (Note that the original C_6 lattice rotation by $2\pi/6$ around the lattice site is *not* the symmetry of $YbMgGaO_4$ due to alternating—above and below the plane—location of oxygens at the centers of consecutive elementary triangles [13].)

These symmetries strongly constrain possible U(1) mean-field spinon Hamiltonians and result in eight different PSG states, of U1A and U1B kind. U1A states maintain periodicity of the original lattice and their band structure consists of just two spinon bands. U1B states are π-flux states with doubled unit cell. Equivalently, their band structure contains four spinon bands. For the sake of simplicity, we focus on the U1A family in the following (description of U1B increases algebraic complexity without adding any new essential physics). The U(1) mean-field spinon Hamiltonian is parameterized by several hopping amplitudes: $t_{1,2}$ describes spin-conserving hopping between the nearest and the next-nearest neighbors and $t'_{1,2}$ describes analogous non-spin-conserving hops. PSG analysis fixes relative phases between hopping amplitudes on the bonds related by the space group operations (see Supplemental Material [32] for details of the derivation). The magnitudes of these hoppings are not determined by PSG. This requires a separate variational calculation of the ground state energy, which is not attempted here. We do expect, on physical grounds, that, for the spin model with predominant isotropic nearest-neighbor spin exchange and subleading asymmetric $J_{\pm\pm}$ terms, the following estimate should hold $t_1 > t'_1 > t_2 > t'_2$.

There are four mean-field Hamiltonians in the U1A family, labeled by $U1A_{n_{C_2} n_{\bar{C}_6}}$ ($n_{C_2}, n_{\bar{C}_6} \in \{0, 1\}$).They have the simple form

$$H = \sum_{\mathbf{k}} (f^\dagger_{\mathbf{k}\uparrow}, f^\dagger_{\mathbf{k}\downarrow}) \begin{pmatrix} \omega_{\mathbf{k}} + \epsilon_{\mathbf{k}} & \eta_{\mathbf{k}} \\ \eta^*_{\mathbf{k}} & \omega_{\mathbf{k}} - \epsilon_{\mathbf{k}} \end{pmatrix} \begin{pmatrix} f_{\mathbf{k}\uparrow} \\ f_{\mathbf{k}\downarrow} \end{pmatrix}, \qquad (1)$$

where $\mathbf{k}$-dependent $\omega_{\mathbf{k}}, \epsilon_{\mathbf{k}}, \eta_{\mathbf{k}}$ are listed in [32]. Spin-orbit interaction appears via spin-nonconserving hopping η_k in (1). The U1A00 state is characterized by finite $\omega_{\mathbf{k}}$ and zero $\epsilon_{\mathbf{k}}$ and $\eta_{\mathbf{k}}$, while U1A01 and U1A11 have $\omega_{\mathbf{k}} = 0$ and finite $\epsilon_{\mathbf{k}}$ and $\eta_{\mathbf{k}}$. In the calculations below, we set $t_1 = 1$ and $t'_1 = 0.8$, $t'_2 = 0.3$ for U1A11, while $t_1 = 0$ for U1A01 and we choose $t'_1 = 1$, $t_2 = 0.8 t'_1$, $t'_2 = 0.4 t'_1$ for it. U1A10 turns out to be nonphysical since its Hamiltonian matrix is zero, $t_{1,2} = t'_{1,2} = 0$. The "accidental" nature of the U1A00 state is manifested by the absence of any spin-dependent hopping in its Hamiltonian—this state happens to be more symmetric than the spin Hamiltonian it describes and is characterized by the large Fermi surface [23].

We focus on most physically relevant U1A01 and U1A11 states, for which $\omega_{\mathbf{k}} = 0$. The resulting fermion bands are easy to find, $E_{\nu=1,2}(\mathbf{k}) = (-1)^\nu E(\mathbf{k}) = (-1)^\nu \sqrt{\epsilon_{\mathbf{k}}^2 + |\eta_{\mathbf{k}}|^2}$. The U1A11 state possesses symmetry-protected Dirac nodes at Γ and M points of the hexagonal Brillouin zone, while U1A01 has additional Dirac nodes at K points as well.

Interaction with monochromatic radiation linearly polarized along direction $\hat{\mathbf{n}} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ is described by $V(t) = -\mathbf{h}(t) \cdot \mathbf{S}_{\rm tot}$, i.e.,

$$V(t) = h e^{-i\omega t} \mathbf{n} \cdot \frac{1}{2} \sum_{\mathbf{r}} (f^\dagger_{\mathbf{r}\uparrow}, f^\dagger_{\mathbf{r}\downarrow}) \boldsymbol{\sigma} \begin{pmatrix} f_{\mathbf{r}\uparrow} \\ f_{\mathbf{r}\downarrow} \end{pmatrix}. \qquad (2)$$

Within linear response theory, the rate of energy absorption $I(\omega) = -\omega \chi''_{\hat{n}\hat{n}}(\omega) |h|^2/2$ is determined by the imaginary part of $\mathbf{q} = 0$ Fourier transform of the dynamic susceptibility [1] $\chi_{\hat{n}\hat{n}}(t, \mathbf{r}) = -i\Theta(t) \langle [\mathbf{S}_{\mathbf{r}}(t) \cdot \hat{\mathbf{n}}, \mathbf{S}_{\mathbf{0}}(0) \cdot \hat{\mathbf{n}}] \rangle$, with Θ being the Heaviside function. Straightforward calculation gives

$$\chi_{\hat{n}\hat{n}}(\omega) = \frac{1}{4N} \sum_{\mathbf{k}} \frac{n_{\mathbf{k}\alpha} - n_{\mathbf{k}\beta}}{\omega + E_\alpha(\mathbf{k}) - E_\beta(\mathbf{k}) + i0} \times (U^+_{\mathbf{k}} \sigma^a U_{\mathbf{k}})_{\alpha\beta} (U^+_{\mathbf{k}} \sigma^b U_{\mathbf{k}})_{\beta\alpha} \hat{n}^a \hat{n}^b. \qquad (3)$$

Here, $n_{\mathbf{k}\alpha}$ is the occupation number of the band α, $U_{\mathbf{k}}$ is the unitary diagonalizing matrix connecting the spinor of original fermions to that of the band ones, $f_{\mathbf{k},\alpha} = (U_{\mathbf{k}})_{\alpha\beta} b_{\mathbf{k},\beta}$, and summation over repeated indices is implied. Equation (3) shows that, in the spin-degenerate U1A00 state, for which $n_{\mathbf{k}\alpha} = n_{\mathbf{k}\beta}$, the susceptibility is strictly zero. Therefore, in agreement with general discussion above, no energy absorption occurs in the absence of an external magnetic field for this state. The condition $n_{\mathbf{k}\alpha} \approx n_{\mathbf{k}\beta}$ is also satisfied at high temperature of the order of spinon bandwidth (which is of the order of exchange J) when spinon resonance disappears. We therefore expect the width of the resonance to increase when the temperature is lowered. It is worth noting that the lowest temperature of the ESR study [14] is 1.8 K, which makes it a high-temperature measurement.

At zero temperature absorption at frequency ω is possible only via vertical transitions from the filled lower band ($\alpha = 1, n_{\mathbf{k}1} = 1$) to the empty upper one ($\beta = 2$, $n_{\mathbf{k}2} = 0$), and therefore

$$\chi''_{\hat{n}\hat{n}}(\omega) = -\frac{\pi}{4N}\sum_{\mathbf{k}}\delta[\omega - 2E(\mathbf{k})](U^{+}_{\mathbf{k}}\sigma^a U_{\mathbf{k}})_{12} \times (U^{+}_{\mathbf{k}}\sigma^b U_{\mathbf{k}})_{21}\hat{n}^a\hat{n}^b. \quad (4)$$

After some algebra, the product of matrix elements 12 and 21 of the rotated Pauli matrices in the equation above simplifies to

$$\chi''_{\hat{n}\hat{n}}(\omega) = -\frac{\pi}{4N}\sum_{\mathbf{k}}\frac{\delta[\omega - 2E(\mathbf{k})]}{E(\mathbf{k})^2}[(\epsilon^2_{\mathbf{k}} + \eta''^2_{\mathbf{k}})\sin^2\theta\cos^2\phi + (\epsilon^2_{\mathbf{k}} + \eta'^2_{\mathbf{k}})\sin^2\theta\sin^2\phi + |\eta_{\mathbf{k}}|^2\cos^2\theta]. \quad (5)$$

It can be shown [32] that the omitted off-diagonal terms, containing products $\hat{n}^x\hat{n}^y, \hat{n}^x\hat{n}^z$, and $\hat{n}^y\hat{n}^z$, are all zero. Moreover, terms proportional to $\cos^2\phi$ and $\sin^2\phi$ are actually equal, so that the absorption only depends on the azimuthal angle θ with respect to the normal to the magnetic layer.

Two features of this result are worth noting. First, the absorption takes place over the wide band of frequencies, $\min(E) = 0 < \omega/2 < \max(E)$, which covers the full bandwidth of two-spinon continuum. Second, Eq. (5) describes "zero-field absorption," which does not require any external static magnetic field **B**. Both of these are a direct consequence of the absence of spin conservation in Eq. (1).

U1A11 state.—Figure 1 shows scaled absorption intensity, $2I(\omega)/|h|^2 = -\omega\chi''_{\rm nn}(\omega)$, for different polarizations. Polarization dependence is strong. The plot is obtained by numerical integration of (5), with frequency steps of $\Delta\omega = 0.05$, over the primitive cell of the reciprocal lattice [$\mathbf{k} = (k_1, k_2)$, where $k_{1,2} \in (0, 2\pi)$; see [32] for details]. We approximate the delta function by the Lorenzian $\delta(x) \approx \pi^{-1}d/(d^2 + x^2)$ with $d = 0.01$. We checked that $d = 0.05$ results in the same outcome. As expected, and also easy to check analytically, $\chi''_{\hat{n}\hat{n}}(\omega) \sim \omega$ at small frequencies. This is the consequence of Dirac nodes at Γ and M points.

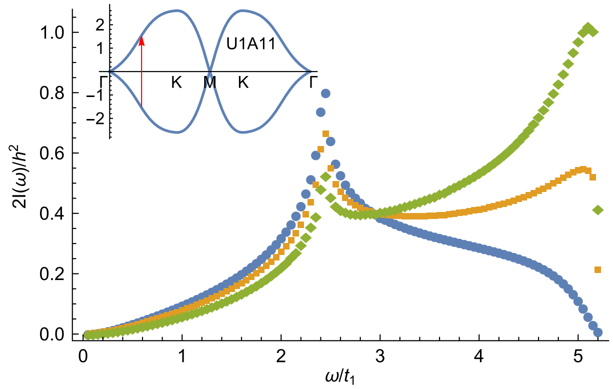

FIG. 1. Plot of $2I(\omega)/|h|^2$ vs ω/t_1 for different polarizations $\theta = 0$ (blue dots), $\pi/4$ (orange squares), and $\pi/2$ (green rhombi) for U1A11 state. The inset shows spinon band structure along the high-symmetry path Γ-K-M-K-Γ in the Brillouin zone. The vertical red line illustrates optical transitions between spinon bands.

Behavior near the upper boundary, $\omega \approx 3\sqrt{3}$, is determined by the vicinity of the K point where $\epsilon(\mathbf{K}) = \text{const}$, while $\eta(\mathbf{K}) = 0$. As a result, one obtains $\chi''_{zz} \sim 3\sqrt{3} - \omega$, while at $\theta = \pi/2$ susceptibility terminates discontinuously in a steplike fashion, $\chi''_{\hat{n}\hat{n}} = \chi''_{xx} \sim \Theta(3\sqrt{3} - \omega)$. The rounding of the step-function behavior in Fig. 1, for $\theta \neq 0$, is caused by the finite width of the numerical delta function used in the integration over the Brillouin zone. The peak in the middle of the absorption band, at $\omega \approx 2.43$, is caused by the van Hove singularity, of the saddle point kind, of $E(\mathbf{k})$ at $\mathbf{k}_0 = (k_0, 2\pi - k_0)$ and symmetry-related points. Here, $k_0 \approx 1.97$ and $E(\mathbf{k} \approx \mathbf{k}_0) \approx 1.215 + 1.31(k_1 + k_2)^2 - 0.23(k_1 - k_2)^2$. The saddle point produces logarithmically divergent contribution, $\chi''_{\hat{n}\hat{n}} \sim \ln|\omega - 2.43|$, which matches numerical data in Fig. 1 perfectly.

U1A01 state.—SMR of this phase is shown in Fig. 2. It is seen to host two van Hove singularities, which can be

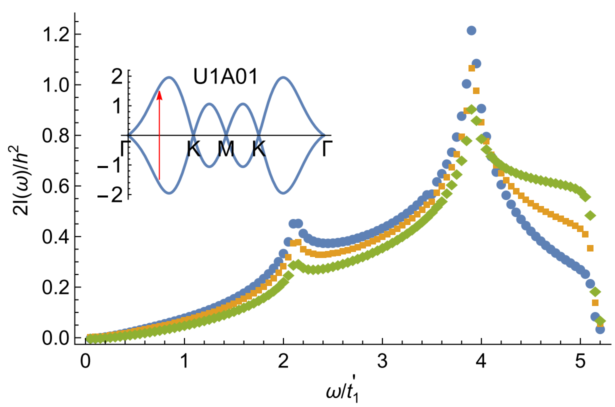

FIG. 2. Plot of $2I(\omega)/|h|^2$ vs ω/t'_1 for different polarizations $\theta = 0$ (blue dots), $\pi/4$ (orange squares), and $\pi/2$ (green rhombi) for U1A01 state. The inset shows spinon dispersion along the path in Fig. 1.

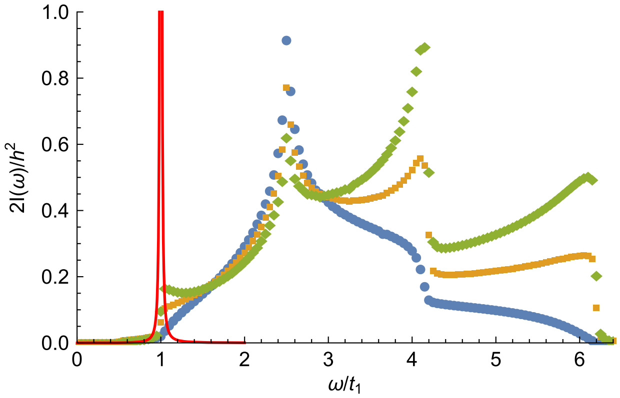

FIG. 3. Plot of $2I(\omega)/|h|^2$ vs $\omega/(2t_1)$ for polarizations $\theta = 0$ (blue dots), $\pi/4$ (orange squares), and $\pi/2$ (green rhombi) for U1A11 state in the presence of magnetic field $B_z = 1$. Note the appearance of strong van Hove singularity at $\omega \approx 4.2t_1$. The thin red line shows Zeeman response of U1A00 state.

qualitatively understood as a direct consequence of the additional, in comparison with the U1A11 state, Dirac cone in the spinon dispersion at the K point. The presence of the symmetry-protected node at the K point results in a stronger variation of spinon dispersion in the Brillouin zone and causes the appearance of additional saddle points.

U1A11 state in magnetic field.—An external magnetic field adds further variations to the spinon absorption intensity. We illustrate this with the case of U1A11 state subject to magnetic field $\mathbf{B} = B_z\hat{z}$ along the normal to the magnetic layer. It should be noted that PSG analysis underlaying our consideration assumes time-reversal (TR) symmetry. Therefore, we treat the magnetic field perturbatively, by coupling it to the local TR-odd combination of spinons, which is just $B_z S^z_{\mathbf{r}} \sim B_z f^\dagger_{\mathbf{r}\alpha}\sigma^z_{\alpha\beta} f_{\mathbf{r}\beta}$. Thus, magnetic field enters (1) via $\epsilon_{\mathbf{k}} \to \epsilon_{\mathbf{k}} - B_z/2$ and gaps out Dirac nodes. The minimal excitation energy becomes $\min(E) = B_z/2$ and absorption intensity acquires threshold behavior $I(\omega) \sim \Theta(\omega - B_z)$. This behavior is illustrated in Fig. 3, which also shows development of additional spectral features at $\omega \approx 4.2$; see [32]. The in-plane magnetic field lowers symmetry of the spin Hamiltonian further and its consideration is left for future studies.

This unusual response should be contrasted with that of the large-Fermi-surface state U1A00. Here, $\mathbf{B} = B_z\hat{z}$ leads to the Zeeman splitting of spinon up- and down-spin bands $E_\nu = \omega_{\mathbf{k}} \mp B_z/2$, and therefore, according to (3), one finds the standard result for magnetically isotropic media $\chi''_{\hat{n}\hat{n}}(\omega) \sim \sin^2\theta\delta(\omega - B_z)$. This is consistent with the earlier analysis of [33], where a weak magnetic field $\mathbf{B} = B_z\hat{z}$ was added to the mean-field Hamiltonian similarly. Off the Γ point, i.e., for $\mathbf{q} \neq 0$, one finds broad continuum corresponding to the spinon particle-hole excitations [33].

Discussion.—Physical arguments leading to Eq. (5) are very general and rely on the absence of long-range magnetic order, existence of fractionalized elementary excitations, which ensure a continuumlike response to external probes, and significant built-in spin-orbit interaction, which leads to nonconservation of spin and makes zero-field absorption possible in a wide range of frequencies. All of these are very generic conditions, which are satisfied by essentially every model of spin liquids of U(1) and Z_2 type [but not by spin-conserving SU(2) ones]. The restriction to low-symmetry spin liquids is not really a handicap, as it turned out that the number of possible spin liquids with reduced U(1) and Z_2 vastly outnumbers that of SU(2) symmetric ones [30,34,35]. In particular, the SMR should be present in the celebrated Kitaev's honeycomb model [11], as was emphasized in dynamic structure calculations of [36–39]. There too one can see anisotropic spin structure factor $\mathcal{S}^{aa}(\mathbf{q} = 0, \omega)$, with $\mathcal{S}^{zz} \neq \mathcal{S}^{xx/yy}$, and sharp van Hove singularities in the Majorana fermion density of states. The similarity is not accidental—it follows from the linear mapping between Majorana and projective spinon representations [40,41]. Unlike the situation described here, in the exactly solvable gapless Abelian region, dynamic response appears above a finite threshold energy (which is the energy cost of creating Z_2 fluxes). However, generic spin exchange perturbations turn the response gapless [42], so that $\mathcal{S}^{aa}(\mathbf{q} = 0, \omega) \sim \omega$ at low energy. Resonant inelastic x ray, Raman scattering, and parametric pumping of the Z_2 Kitaev spin liquid results in a gapless and extended energy continuum too [43–46].

Our theory can be broadly thought of as an extension of one-dimensional theories of ESR in spin chains with Dzyaloshinskii-Moriya interactions [1,47–49]. In one dimension, the fractionalized nature of spinons is very well established and theories based on them describe ESR experiments exceedingly well, both in gapless [50–52] and gapped [53,54] settings.

Another important connection is provided by electric dipole spin resonance, which describes absorption of EM radiation in conductors with pronounced spin-orbit interaction, which mediates coupling of the ac electric field to the electron spin [55]. Here, spin-rotational asymmetry causes strong absorption, which is controlled by the real part of optical conductivity [56–62].

Somewhat surprisingly, energy absorption due to coupling of spins to the ac electric field is also possible in strong Mott insulators, provided they are built of frustrated triangular units, in which virtual charge fluctuations produce spin-dependent electric polarization [63,64]. Hints of this physics were recently observed in herbertsmithite and α-$RuCl_3$ antiferromagnets [65–67].

Simple calculations of SMR presented here are based on mean-field spinon Hamiltonians derived with the help of PSG formalism. They do not include gauge fluctuations, which undoubtedly are present in the theory. These fluctuations are certain to affect exponents characterizing sharp features of $\chi''_{\hat{n}\hat{n}}(\omega)$, such as, for example, behavior near the van Hove singularity and/or near the lower or upper edge of the two-spinon continuum. (Disorder, in the form of Mg/Ga mixing, leads to distribution of g factors [25], which also broadens magnetic response.) In addition, by analogy with critical Heisenberg chain [68], we expect four-spinon contributions to the susceptibility to affect the high-frequency behavior. However, these important effects cannot reduce spinon absorption bandwidth and eliminate other outstanding features of the SMR found here. It should also be noted that SMR is not specific to fermionic spinons and indeed extension of the theory to bosonic PSG is possible as well [69,70]. We therefore conclude that spinon magnetic resonance represents an efficient and informative probe of exotic excitations of spin-orbit-coupled quantum spin liquids.

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Note added.—Recent manuscript [71] contains a detailed comparison of ground state energies of various U(1) PSG states.

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