Optical observation of fermionic partons in Kitaev spin balls

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Abstract. We demonstrate a Kitaev spin liquid in a polyhedral cluster and propose detecting its fractional excitations—Majorana spinons—by magnetic Raman scattering. While little polarization dependence of the Raman spectra at sufficiently low temperatures is usual with quantum spin liquids, the present observations are primarily of geometric origin. The Raman scattering intensity peaks melt with increasing temperature, to be more precise, with increasing number of background gauge-flux excitations.

1. Introduction
Quantum spin liquids (QSLs) [1] have no conventional magnetic order but feature fractional excitations—partons—and emergent gauge degrees of freedom [2]. The Kitaev model [3] is exactly solvable to have a QSL ground state and its elementary excitations consist of itinerant Majorana fermions and local gauge vortices, called spinons and visons, respectively. While Kitaev-type interactions are found in some honeycomb iridates [4], they can be defined on any lattice with coordination number three. We consider a Kitaev spin liquid in “zero dimension” in an attempt to identify and characterize Majorana spinons on a rigorous basis. Magnetic Raman scattering is an excellent probe to detect Majorana spinons separately from visons.

2. Kitaev Hamiltonian
We consider the Kitaev Hamiltonian on a dodecahedral lattice of \( L \equiv 20 \) sites

\[
H = - \sum_{\lambda=x,y,z} \sum_{\langle i,j \rangle_\lambda} J_\lambda \sigma^\lambda_i \sigma^\lambda_j; \quad [\sigma^\lambda_i, \sigma^\mu_j] = 2i \delta_{ij} \epsilon_{\lambda\mu\nu} \sigma^\nu_i,
\]

where \( \langle i,j \rangle_\lambda \) runs over nearest neighbors with \( \lambda \) depending on the \((i,j)\) pair [Fig. 1(a)]. We set \( J_\lambda \)'s all equal to \( J \) in what follows. Let

\[
\sigma^\lambda_i = i \eta^\lambda_i c_i; \quad c_i^\dagger = c_i, \quad (\eta^\lambda_i)^\dagger = \eta^\lambda_i; \quad \{\eta^\lambda_i, \eta^\mu_j\} = 2 \delta_{ij} \delta_{\lambda\mu}, \quad \{c_i, c_j\} = 2 \delta_{ij}, \quad \{\eta^\lambda_i, c_j\} = 0.
\]

Then the Hamiltonian (1) reads

\[
H = i \sum_{\lambda=x,y,z} \sum_{\langle i,j \rangle_\lambda} J_\lambda \hat{u}_{\langle i,j \rangle_\lambda} c_i c_j; \quad \hat{u}_{\langle i,j \rangle_\lambda} \equiv i \eta^\lambda_i \eta^\lambda_j.
\]

Since the bond operators \( \hat{u}_{\langle i,j \rangle_\lambda} \) commute with the Hamiltonian as well as each other, they are treatable as classical \( \mathbb{Z}_2 \) variables, \( u_{\langle i,j \rangle_\lambda} = \pm 1 \). Every anticlockwise multiplication—viewed from the outside of the dodecahedron—of the spin operators around pentagonal plaquette \( p \),

\[
\hat{W}_p = \prod_{\langle i,j \rangle_\lambda \in \partial p} \sigma^\lambda_i \sigma^\lambda_j = (-i)^{\delta} \prod_{\langle i,j \rangle_\lambda \in \partial p} \hat{u}_{\langle i,j \rangle_\lambda} (p = 1, 2, \cdots, 3L/5),
\]
The symmetry group of the Kitaev dodecahedron is thus given by a gauge extension of the pure spin model (3) is not invariant under \( c \) gauge transformations. The gauge transformation \( R \equiv \exp(i \pi \tau^3) \) reads operating on all the other sites. Both indeed recover the initial bond configuration. Every \( u_{<i,j>} \) configuration, we can diagonalize the Majorana Hamiltonian into

\[
\mathcal{H} = \sum_{k=1}^{L/2} \frac{\varepsilon_k}{2} \left( \alpha_k^+ \alpha_k - \alpha_k \alpha_k^+ \right) = \sum_{k=1}^{L/2} \varepsilon_k \left( n_k - \frac{1}{2} \right); \quad n_k = \alpha_k^+ \alpha_k, \quad c_i = \sum_{k=1}^{L/2} (\psi_{ik} \alpha_k + \psi_{ik}^* \alpha_k^+),
\]

where \( c_i \)'s are recomplexified into \( \alpha_k \)'s to obtain the eigenvalues \( \varepsilon_k \) which are positive definite.

3. Projective symmetry group

Before proceeding to optical observation, we make a symmetry argument of this Kitaev spin ball [Fig. 1(b)]. While a dodecahedron belongs to the point group \( \text{Ih} \), the dodecahedral Kitaev spin model (3) is not invariant under \( \text{Ih} \) group operations. Even if we choose bond variables so as to give the uniform \( W_p \) configuration, neither simple rotation nor inversion keeps these local gauge fields invariant. Then we extend a simple point group by gauge transformations [5].

Suppose we rotate the gauge-ground Kitaev Hamiltonian anticlockwise about a three-fold axis, say \( n \), of the dodecahedron by an angle of \( \frac{2\pi}{3} \), which is denoted by \( R(\frac{2\pi}{3}, n) \). Then, bond variables may be changed but the ground \( W_p \) configuration remains unchanged. Any two sets of bond variables giving the same \( W_p \) configuration can be converted to each other by local gauge transformations. The gauge transformation \( c_i \rightarrow -c_i \) reads \( u_{<i,j>} \rightarrow -u_{<i,j>} \) with \( j \) running over the nearest neighbor sites of site \( i \). \( A(\frac{2\pi}{3}, n) \) operates on 18 sites, while \( -A(\frac{2\pi}{3}, n) \) reads operating on all the other sites. Both indeed recover the initial bond configuration. Every rotation \( R \subset \text{I} \) can be followed by two sets of local gauge transformations to keep any ground-state bond-variable configuration invariant. Since the inversion reverses all the pentagonal plaquette fluxes \( W_p \), any gauged inversion cannot restore the initial bond-variable configuration. The symmetry group of the Kitaev dodecahedron is thus given by a gauge extension of the pure rotation subgroup of the icosahedral group \( \text{Ih} \), which is denoted by \( \tilde{\text{I}} \). This extended group is double valued, hence called a projective symmetry group (PSG) [6]. If we apply the gauge \( \frac{2\pi}{3} \) rotation three times to any bond-variable configuration, it is multiplied by \(-1\), which is reminiscent of the case with a half-odd integer spin. Note that \( \tilde{\text{I}} \subset \text{SU}(2) \) and \( \text{I} \subset \text{SO}(3) \).
We can characterize every eigenstate of the Hamiltonian (5) in terms of this symmetry group unless the uniform $W_p$ configuration is broken. Every eigenstate of the gauge-ground Kitaev dodecahedron belongs to any of the irreducible representations of $\mathbf{I}$ and the degeneracy of each eigenvalue is given by the dimension of its belonging representation [Fig. 1(a)].

4. Magnetic Raman scattering

Within the Loudon-Fleury perturbation theory [7, 8], magnetic Raman intensity reads

$$I(\omega) = \frac{1}{2\pi\hbar L} \int_{-\infty}^{\infty} dt \, e^{i\omega t} \langle e^{i\frac{2\pi}{L} \mathbf{R}^{\text{sc}}} \mathbf{R} \rangle_T; \quad \mathbf{R} = -\sum_{\lambda=x,y,z} \sum_{i<j,\lambda} J_{\lambda}(\mathbf{e}_{in} \cdot \mathbf{d}_{ij})(\mathbf{e}_{sc} \cdot \mathbf{d}_{ij})\sigma_{\lambda}^i \sigma_{\lambda}^j,$$

where $\mathbf{e}_{in}$ ($\mathbf{e}_{sc}$) is the polarization vector of incident (scattered) light, while $\mathbf{d}_{ij}$ is the vector connecting the $i$th and $j$th sites. Having in mind that $[\mathbf{R}, \hat{u}_{<i,j,\lambda}] = 0$, the magnetic Raman operator is expressed in terms of the fermionic quasiparticles as

$$\mathbf{R} = \frac{1}{2} \sum_{k,l=1}^{L/2} \left[ X_{kl}(2\alpha_k \alpha_l - \delta_{kl}) + Y_{kl} \delta_{kl} + Y_{kl} \alpha_k \alpha_l \right];$$

$$X_{kl} = \sum_{\xi=x,y,z} \sum_{i<j,\lambda} iJ_{\lambda}(\mathbf{e}_{in} \cdot \mathbf{d}_{ij})(\mathbf{e}_{sc} \cdot \mathbf{d}_{ij})u_{<i,j,\lambda} [\psi^*_i \psi_{jl} - \psi^*_j \psi_{il}],$$

$$Y_{kl} = \sum_{\xi=x,y,z} \sum_{i<j,\lambda} iJ_{\lambda}(\mathbf{e}_{in} \cdot \mathbf{d}_{ij})(\mathbf{e}_{sc} \cdot \mathbf{d}_{ij})u_{<i,j,\lambda} [\psi^*_i \psi_{jl} - \psi^*_j \psi_{il}].$$

Note that $[\mathbf{R}, \mathcal{H}] \neq 0$ in general. Neglecting the Rayleigh and anti-Stokes terms, the scattering intensity mediated by matter fermions with a particular $W_p$ configuration is calculated as

$$I_{\{W_p\}}(\omega) = \frac{1}{2L} \sum_{k,l=1}^{L/2} \left[ 2|X_{kl}|^2 n_k (1-n_l) \delta(h\omega + \epsilon_k - \epsilon_l) + |Y_{kl}|^2 (1-n_k)(1-n_l) \delta(h\omega - \epsilon_k - \epsilon_l) \right].$$

Then we take a thermal average to obtain the spectrum,

$$I(\omega) = \frac{1}{Z} \sum_{\{W_p\}} \sum_{\{n_k\}} \langle \{n_k\} | e^{-\beta \mathcal{H}} I_{\{W_p\}}(\omega) \mathcal{P} | \{n_k\} \rangle_{\{W_p\}} \equiv \left< I_{\{W_p\}}(\omega) \right>_{\mathcal{T}},$$

where $Z = \sum_{\{W_p\}} \sum_{\{n_k\}} \langle \{n_k\} | e^{-\beta \mathcal{H}} \mathcal{P} | \{n_k\} \rangle_{\{W_p\}}$, $W_p$’s each run over $\pm i$, $n_k$’s each run over 0 and 1, and $\mathcal{P}$ projects out all the unphysical combinations of $\{W_p\}$ and $\{n_k\}$ [9].

Figure 2(a) shows $I(\omega)$ with various polarization vectors in the $xy$-plane at absolute zero, where Eq. (8) consists only of the second term and therefore every Raman-active mode is characterized by a product representation of two quasiparticle eigenstates belonging to either $I_2$ or $G_2$ representation. On the other hand, the Raman spectra can be characterized by irreducible representations of the lattice point group $\mathbf{I}_h$. The Kitaev dodecahedron has only one Raman-active mode belonging to the five-dimensional $H_5$ representation of $\mathbf{I}_h$. When we decompose the Raman operator as $\mathbf{R} = \sum_{i=1}^{5} E_{H_5(i)} \mathbf{R}^{H_5(i)}$ neglecting the irrelevant Rayleigh component of $\mathbf{A}_{g}$ symmetry, the contributions $\langle e^{i\frac{2\pi}{L} \mathbf{R}^{H_5(i)}} e^{-i\frac{2\pi}{L} \mathbf{R}^{H_5(i)}(T=0)} \rangle_{\mathcal{T}}$ are all degenerate with each other as long as the ground state is a QSL [10]. Then $I(\omega)$ reads

$$I(\omega) = \frac{1}{2\pi\hbar L} \int_{-\infty}^{\infty} dt \, e^{i\omega t} \langle e^{i\frac{2\pi}{L} \mathbf{R}^{H_5(1)}} e^{-i\frac{2\pi}{L} \mathbf{R}^{H_5(1)}(T=0)} \rangle_{\mathcal{T}} \sum_{i=1}^{5} E^2_{H_5(i)};$$

hence comes very little polarization dependence originating merely in the coefficients $E^2_{H_5(i)}$. There is no polarization dependence at all in a planar honeycomb Kitaev spin liquid [8], where the Raman operator consists only of the two-dimensional $E_2$ representation of the point group $C_{6v}$ as $\mathbf{R} = \sum_{i=1}^{2} E_{E_2(i)} \mathbf{R}^{E_2(i)}$ with $E_{E_2(1)} = (e_{in}^x e_{sc} - e_{in}^y e_{sc})/\sqrt{2}$ and $E_{E_2(2)} = (e_{in}^y e_{sc} + e_{in}^x e_{sc})/\sqrt{2}$, and satisfy $\sum_{i=1}^{2} E^2_{E_2(i)} = 1/2$ as well as $\langle e^{i\frac{2\pi}{L} \mathbf{R}^{E_2(1)}} e^{-i\frac{2\pi}{L} \mathbf{R}^{E_2(1)}(T=0)} \rangle_{\mathcal{T}} = \langle e^{i\frac{2\pi}{L} \mathbf{R}^{E_2(2)}} e^{-i\frac{2\pi}{L} \mathbf{R}^{E_2(2)}(T=0)} \rangle_{\mathcal{T}}$. However, no or little polarization dependence of the magnetic Raman spectra is not necessarily
the case with QSLs. It is primarily of geometric origin. We will demonstrate elsewhere strongly polarization-dependent magnetic Raman spectra in Kitaev spin clusters of octahedral symmetry. A strong polarization dependence is observed in three-dimensional Kitaev QSLs [10] as well.

Figure 2(b) shows $I(\omega)$ as a function of temperature. As soon as visons are thermally excited, the Hamiltonian (3) no longer belongs to $\hat{I}$. Vison excitations can be measured by the parameter

$$w_P = \frac{1}{Z} \sum_{\{W_p\}} \sum_{\{n_k\}} \langle\{n_k\}\rangle e^{-\beta\hat{H}\hat{w}_P}\langle\{n_k\}\rangle_{\{W_p\}}; \quad \hat{w}_P = \frac{5}{3L} \sum_{p=1}^{3L/5} \hat{W}_p$$

and observed through specific heat. Figure 2(c) shows their temperature dependences. $w_P = 1$ without any vison at $T = 0$, while $w_P = 5040/2048/12 = 0.205078125$ with visons emerging at random in the $T \to \infty$ limit. At $k_BT/J = 0.2$, visons are almost fully excited and therefore the symmetry-definite Raman scattering intensity peaks melt away. Matter fermions are excited at much higher temperatures. Specific heat is thus doubly peaked with increasing temperature.

5. Concluding remarks

Gapless algebraic quantum spin liquids [11] such as critical Heisenberg antiferromagnets in one dimension are also attracting much interest in the context of fractional spinon excitations, where Jordan-Wigner fermions behave as elementary spin-$\frac{1}{2}$ entities and they are indeed detected in pairs by inelastic neutron scattering [12]. Our proposal consists of observing fractional magnetic excitations—Majorana spinons—by inelastic light scattering rather than usual magnetic probes. PSG characterization of magnetic Raman spectra mediated by fractional quasiparticles may be available to other QSL nanostructures such as hexagonal fragments. We hope this study will stimulate further chemical explorations of Kitaev spin systems, say, nanoribbons and nanotubes.

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