Raman Scattering Polarization and Single Spinon Identification in Two-Dimensional Kitaev Quantum Spin Liquids

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Perfect depolarization of the Loudon-Fleury inelastic visible-light scattering in the Kitaev honeycomb model is well known. Though it happens in Heisenberg Kagome and triangular antiferromagnets as well, yet we prove it to be of geometric origin rather than peculiar to quantum spin liquids. A Kitaev spin liquid in the square planar geometry indeed exhibits polarized Raman spectra containing different symmetry species, each brought by symmetry-compatible spinon geminate excitations, i.e., arising from symmetry-compatible direct product representations made of double-valued irreducible representations of mediating-spinon-belonging gauged k-point symmetry groups. We combine a standard point-symmetry-group analysis of the Raman vertex in the real space and an elaborate projective-symmetry-group analysis of Raman-scattering-mediating Majorana spinons in the reciprocal space to identify emergent spinons singly.

The Kitaev honeycomb model has made a major breakthrough in the study of quantum spin liquids (QSLs) explicitly visualizing partons as elementary excitations. Majorana spinons accompanied by emergent \( Z_2 \) gauge fields are characteristic of the Kitaev QSL and Raman spectroscopy is particularly useful in diagnosing them. Within the Loudon-Fleury (LF) mechanism valid for strongly correlated electrons, the Raman vertex commutes with background gauge fields and can selectively excite spinons.

The LF vertex for the gauge-ground Kitaev honeycomb QSL yields a completely depolarized Raman response, but the depolarization is no longer perfect beyond the LF theory in an external field and with integrability-breaking perturbations such as Heisenberg and o-flattening, exchanges, whether intralayer or interlayer. On the other hand, the depolarization of the LF Raman response occurs in Heisenberg frustrated antiferromagnets as well, including a U(1) Dirac spin-liquid state on the regular Kagome lattice and a \( Z_2 \) dimer-liquid phase on the equilateral triangular lattice. There is an argument that depolarization of Raman response may be characteristic of QSLs.

We are thus motivated to discuss Raman responses of various two-dimensional Kitaev models (Fig. 1). We make symmetry arguments in two ways. Point-symmetry analysis of the LF vertex [cf. Eq. (16)] in the real space reveals what is the decisive factor for Raman scattering polarization, and then, projective-symmetry analysis of the gauge-ground Majorana Hamiltonian [cf. Eq. (4)] in the reciprocal space shows which mode of polarized Raman responses, if any, is attributable to which combination of spinon eigenmodes.

The Kitaev Hamiltonian (cf. Fig. 1) reads

\[
\mathcal{H} = - \sum_{\langle r, x, r', x' \rangle} J_{\alpha(r,x,r',x')}(\sigma^\alpha_{r,x})(\sigma^\alpha_{r',x'})
\]

where \( (\sigma^\alpha_{r,x}, \sigma^\beta_{r,x}, \sigma^\gamma_{r,x}) \) (l = 1, \ldots, \( N^2 \equiv L/g \); \( \alpha = 1, \ldots, q \)) are the Pauli matrices attached to the \( l \)-th site in the \( l \)-th unit at \( r \) and obey the usual commutation relations \( [\sigma^\alpha_{r,x}, \sigma^\beta_{r',x'}] = 2i\delta^\alpha_\beta \delta_{r,x} \sum_{\gamma=xyzt} e_{\gamma \delta} \sigma^\gamma_{r',x'} \) while \( \langle r, x, r', x' \rangle \) runs over 3\( L/2 \) nearest-neighbor bonds with \( \alpha(r_1, x_1, r_2, x_2, r_3) = x \) as a function of \( r_1 \) and \( r_2 \) : \( x \) taking \( x, y, \) and \( z \) once every \( r_1 \) \( : \). The coupling constants \( J_{\alpha(r,x,r',x')} \) are all set to \( J > 0 \) in the following. We start with the pure honeycomb lattice containing \( L/2 \) hexagons [Fig. 1(a)], next decorate this putting triangles and \( L/3 \) diamonds [Fig. 1(b)] or \( L/4 \) hexagons and squares [Fig. 1(c)] and \( L/12 \) dodecahedrons [Fig. 1(d)], and then proceed to a square analog decorated with diamonds containing \( L/4 \) diamonds and \( L/4 \) octagons [Fig. 1(d)]. Denoting the primitive translation vectors in each lattice by \( a \) and \( b \), we adopt a periodic boundary condition, \( r_i + Na = r_i + Nb = r_i \). The number of sites \( L \) reads \( qN^2 \) with \( q = 2 \) [Fig. 1(a)], \( q = 6 \) [Fig. 1(b)], \( q = 12 \) [Fig. 1(c)], and \( q = 4 \) [Fig. 1(d)].

We introduce four Majorana fermions at each site as \( \sigma^\alpha_{r,A} = i\eta^\alpha_{r,A} \) with \( \eta^\alpha_{r,A}(\eta^\alpha_{r,A}^\dagger) = 2\delta^\alpha_\beta \delta_{r,A} \delta_{r,A} \), \( \sigma^\alpha_{r,r'} = 2i\delta_{r,r'} \), and \( \eta^\alpha_{r,A} = 0 \) to have

\[
\mathcal{H} = iJ \sum_{\langle r, x, r', x' \rangle} \hat{u}_{r,x}^{r',x'} \sigma^\alpha_{r,x} \sigma^\alpha_{r',x'}, \quad (2)
\]

where the nearest-neighbor bond operators \( \hat{u}_{r,x}^{r',x'} \) commute with each other as well as the Hamiltonian (2) and therefore behave as \( Z_2 \) classical variables, \( u_{r,x}^{r',x'} = \pm 1 \). For an \( N_g \)-sided polygon, we multiply its constituent spin operators in the anticlockwise manner to define the flux operator

\[
\hat{W}_p \equiv \prod_{\langle r, x, r', x' \rangle \in \partial \hat{p}} \sigma^\alpha_{r,x}(\alpha(r,x,r',x') \sigma^\alpha_{r',x'})
\]

\[
= (-i)^{N_g} \prod_{\langle r, x, r', x' \rangle \in \partial \hat{p}} \hat{u}_{r,x}^{r',x'} \quad (3)
\]
also commutes with the Hamiltonian, whether (1) or (2), and thus behaves as a classical variable, $W_p = \pm 1$ or $\pm i$ according as $N_p$ is even or odd. We have a U(1) gauge flux, $W_p = e^{i\phi_p} (\pi < \phi_p \leq \pi)$, in general. Each Kitaev lattice consists of $\frac{q}{2}$ gauged polynomials with their flux variables satisfying $\prod_{p=1}^N W_p = 1$. Under the periodic boundary condition, there are two more nontrivial flux operators $W_c \equiv \{a \equiv \{a, b\}\}^{10,32}$ wrapping around the torus in the directions $a$ and $b$, each with eigenvalues $\pm 1$ in Figs. 1(a)–1(c) and $\pm 1$ or $\pm i$ according as $N$ is even or odd in Fig. 1(d). We set $N$ equal to a sufficiently large even number, 8192 or more. We have $2^q$ + 1 flux configurations $\{W_p, W_c\}$, each available from a set of $2^q/2^q + 1 = 2^q - 1$ different bond configurations $\{u^{(\alpha)}_{\langle c, c'\rangle}\}$. The eigenspectrum of (2) depends on $\{u^{(\alpha)}_{\langle c, c'\rangle}\}$ only through $\{W_p, W_c\}$.

The ground-flux-configuration sector of (2) reads

$$\mathcal{H} = \frac{i J}{2} \sum_{\langle \alpha, \beta \rangle} \sum_{\langle \alpha', \beta' \rangle} \sum_{\langle \alpha'', \beta'' \rangle} u^{(\alpha)}_{\langle c, c'\rangle} c_{\alpha', \beta'} c_{\alpha'', \beta''} c_{\beta', \alpha'},$$

(4)

where $\tilde{a}_{\langle c, c'\rangle} \equiv u^{(\alpha)}_{\langle c, c'\rangle} \delta$ no longer depend on the position $r$. Carrying out the Fourier transformation

$$\gamma_{k, \alpha} = \frac{1}{\sqrt{N}} \sum_{\alpha = 1}^N e^{i k r \lambda} c_{\alpha', \beta'} c_{\alpha', \beta'},$$

(5)

with $\gamma_{-k, \alpha} = \gamma^\dagger_{k, \alpha}$ in mind yields

$$\mathcal{H} = i J \sum_{\alpha = 1}^N \frac{1}{\sqrt{N}} \sum_{\alpha' = 1}^N u^{(\alpha)}_{\langle c, c'\rangle} \gamma^\dagger_{k', \alpha'} \gamma_{k, \alpha},$$

(6)

We define $q/2$ (complex) bond fermions

$$f_{k, \alpha} = \gamma_{k, 2\alpha-1} + i \gamma_{k, 2\alpha}, \; f^\dagger_{k, \alpha} = \gamma_{k, 2\alpha-1} - i \gamma_{k, 2\alpha},$$

(7)

at each momentum to process (6) into

$$\mathcal{H} = \sum_{\alpha = 1}^N f^\dagger_{k, \alpha} \mathcal{H}_k f_{k, \alpha} = \sum_{\alpha = 1}^N f^\dagger_{k, \alpha} \mathcal{H}^{(\alpha)}_{k} f_{k, \alpha},$$

(8)

with vectors of dimension $q$ and matrices of dimension $\frac{q}{2} \times \frac{q}{2}$

$$\mathcal{H}_k \equiv \left[ f^\dagger_{k, 1} \cdots f^\dagger_{k, \frac{q}{2}} \right] \left[ f_{k, 1} \cdots f_{k, \frac{q}{2}} \right]^\dagger,$$

(9)

$$\mathcal{H}^{(\alpha)}_{k} \equiv \left[ f^\dagger_{k, 1} \cdots f^\dagger_{k, \frac{q}{2}} \right] \mathcal{H}^{(\alpha)}_{k} f_{k, 1} \cdots f_{k, \frac{q}{2}},$$

(10)

Having in mind that the $\pm k$ blocks $\mathcal{H}_{+k}$, of the Hamiltonian (8) are related through two different unitary transformations, $\tilde{a}_{\langle c, c'\rangle} \equiv M^{\dagger} \mathcal{H}_{-k} M = C^\dagger \mathcal{H}_{k} C$, $M$ reads $[M]_{\langle \alpha, \beta \rangle \langle \alpha', \beta' \rangle} = [M]_{\langle \alpha', \beta' \rangle \langle \alpha, \beta \rangle} = 1$ ($\lambda = 1, \cdots, \frac{q}{2}$) and otherwise consists of 0, while $C^\dagger$ is a gauged twofold rotation,30 we find that $\mathcal{H}_{\pm k}$ have the same set of eigenvalues and every such set consists of $q/2$ pairs of eigenvalues $\pm \varepsilon_{\pm k, \lambda}$ ($\lambda = 1, \cdots, \frac{q}{2}$).33 Arranging creation operators for spinon particles and holes into a column vector,

$$\mathcal{A}_{\pm k} = \left[ a^\dagger_{\pm k, 1} \cdots a^\dagger_{\pm k, \frac{q}{2}} \alpha^\dagger_{\pm k, 1} \cdots \alpha^\dagger_{\pm k, \frac{q}{2}} \right]^\dagger,$$

(12)

and defining their energies as

$$\mathcal{E}_{\pm k} = \text{diag}\{\mathcal{E}_{k, 1}, \cdots, \mathcal{E}_{k, \frac{q}{2}}, -\mathcal{E}_{k, 1}, \cdots, -\mathcal{E}_{k, \frac{q}{2}}\},$$

(13)

the gauge-ground Hamiltonian (4) reads

$$\mathcal{H}_g = \sum_{\alpha = 1}^N \mathcal{A}^\dagger_{\pm k} \mathcal{E}_{\pm k} \mathcal{A}_{\pm k} = \sum_{\alpha = 1}^N \sum_{\alpha' = 1}^N \mathcal{A}^\dagger_{\pm k, \alpha} \mathcal{A}_{\pm k, \alpha'} \left( \alpha^\dagger_{\pm k, \alpha} \alpha^\dagger_{\pm k, \alpha'} - \frac{1}{2} \right),$$

(14)

where the eigenvalues $\varepsilon_{\pm k, \lambda}$ lie on $\{\pm \varepsilon_{\pm k, \lambda}\}$ are nonnegative.

Given a lattice of point symmetry $P_{\text{sym}}$, a wavevector $k$ in the first Brillouin zone of its reciprocal lattice belongs to the $k$-point symmetry group (isotropy group of $k$) $P_k \subseteq P_{\text{sym}}$, which consists of point symmetry operations $P_k$ such that $P_k k = k \equiv P k$ with $P$ being $0$ or a reciprocal lattice vector. Each $\mathcal{H}_{\pm k}$ of the gauge-ground Majorana Hamiltonian (8) may belong to a projective $k$-point symmetry group $P_{\text{sym}}^{(k)}$, which is the $2^{\tilde{v}}$-gauge extension of a point symmetry group $P_{\text{sym}}^{(k)}(\text{sym}) = \sum_{\alpha = 1}^n \mathcal{A}^\dagger_{\pm k, \alpha} \mathcal{A}_{\pm k, \alpha}$ of point symmetry operators $\alpha^\dagger_{\pm k, \alpha} \alpha_{\pm k, \alpha}$ and quasiparticle occupation operators $\alpha^\dagger_{\pm k, \alpha} \alpha_{\pm k, \alpha}$ and therefore straightforwardly apply it to quasiparticle states with given background gauge fields $\{u^{(\alpha)}_{\langle c, c'\rangle}\}$. Physical and unphysical states in each gauge-fixed block of the Hilbert space can be distinguished according as the number of emergent (complex) fermions in them is even or odd. Physical states against the ground gauge fields consist of even numbers of quasiparticles $\alpha^\dagger_{\pm k, \alpha} \alpha_{\pm k, \alpha}$, and each $W_p$’s of the constituent polynomials in the ground state are all $-1$, or either of $+i$ and $-i$ according as their $N_p$’s are $4l, 4l + 2$, or $2l + 1$ with $l \in \mathbb{N}$ (Fig. 1).31,41 Apart from the twofold degeneracy due to the constituent triangles, $W_k = \cdots = W_k = \pm i$ in Fig. 1(b), the ground states of the gauged torus Figs. 1(a)–1(d) are all quadruply degenerate due to the topological eigenvalues, $W_k = \pm 1$ and $W_k = \pm 1$.30

We calculate the intensity of Raman scattering through the LF vertex $\mathcal{S}^{(k)}$ in the ground state \(|0\rangle\),

$$I(\omega) = \int_{-\infty}^{\infty} \frac{dt}{2\pi} e^{-i\omega t} \langle 0| e^{i\alpha \mathcal{S}^{(k)} \mathcal{R} e^{-i\alpha \mathcal{S}^{(k)}}} \mathcal{R}|0\rangle; \mathcal{R} \equiv \mathcal{R}^\dagger,$$

(15)

$$\equiv -J \sum_{\langle r, r' \rangle \langle x, x' \rangle} (e^{i\mathcal{S}^{(k)}}_{\langle r, r' \rangle} e^{i\mathcal{S}^{(k)}}_{\langle x, x' \rangle} \delta^{(r, r')}_{\langle x, x' \rangle}) \delta^{(r, r')}_{\langle x, x' \rangle}$$

(16)

$$\equiv i J \sum_{\langle r, r' \rangle \langle x, x' \rangle} (e^{i\mathcal{S}^{(k)}}_{\langle r, r' \rangle} e^{i\mathcal{S}^{(k)}}_{\langle x, x' \rangle} \delta^{(r, r')}_{\langle x, x' \rangle} \delta^{(r, r')}_{\langle x, x' \rangle}) \delta^{(r, r')}_{\langle x, x' \rangle},$$

where $\mathcal{S}^{(k)}_{\langle r, r' \rangle} = \cos \varphi_{\langle r, r' \rangle}$ and $\mathcal{S}^{(k)}_{\langle x, x' \rangle} = \sin \varphi_{\langle x, x' \rangle}$ are the incident and scat-
Fig. 2. (Color online) Spinon excitation energies $\varepsilon_{k,\lambda}$ ($\lambda = 1, \cdots , 5$) and Raman intensities $I(\omega)$ of the gauge-ground Kitaev pure (a), triangle (b), and square-hexagon (c)–honeycomb models. All the three bands in (c) are doubly degenerate due to the primitive-translation-invariant gauged twofold rotational symmetry. The Raman responses do not depend on $(\varphi_\mu, \varphi_\kappa)$ at all.

How many Raman-active modes are possible in the lattice geometry is most decisive of whether and how the intensity depends on the light polarization. Since the pure and decorated honeycomb Kitaev QSLs of triangular geometry, Figs. 1(a)–1(c), have one and only Raman-active mode $E_2$ of dimensionality two with in-plane basis functions such that

$$
E_{E_{2}1}^2 = \frac{\cos^2(\varphi_\mu + \varphi_\kappa)}{2},
$$

$$
E_{E_{2}2}^2 = \frac{\sin^2(\varphi_\mu + \varphi_\kappa)}{2},
$$

their Raman responses $I(\omega)$ are completely depolarized (Fig. 2), regardless of further details such as whether or not the ground state spontaneously breaks time reversal symmetry$^{25}$ and whether the spinon excitation spectrum is gapped or gapless$^{27}$.

The identity representations (18a) and (19a) commute with their Hamiltonians (1), resulting in Rayleigh scattering. Having in mind that $(0|\mathcal{R}_E|0, \mu, \mu) = \delta_{\mu\mu}$, $(0|\mathcal{R}_B|0, \mu, \mu)$ no longer depends on $\mu$ with $|0\rangle$ being invariant under every symmetry operation of $P$. We find that the symmetry species $\mathcal{E}_j$ of $I(\omega)$ should be mediated by spinon geminate excitations containing the same representation $\mathcal{E}_j$.

$$
I(\omega) = \sum_j \mu \sum_{\mu, \mu} \mathcal{R}_E^\mu(\omega)(E_{E_{j}\mu}^\mu)^2 = \sum_j \mu \sum_{\mu, \mu} \mathcal{R}_B^\mu(\omega)(E_{B_{j}\mu}^\mu)^2.
$$

$$
\mathcal{R}_E^\mu(\omega) \equiv \int_{-\infty}^{\infty} \frac{dt e^{i\omega t}}{2\pi \hbar L} (0|e^{i\mu t} \mathcal{R}_E^\mu|0) = \frac{1}{L} \sum_{\lambda=1}^{N} \sum_{\mu=1}^{g/2} \frac{\mu}{L} \sum_{\lambda=1}^{N} \sum_{\mu=1}^{g/2} \left(0|e^{i\mu t} \mathcal{R}_E^\mu|0^\mu\lambda\right)^2 \delta(\hbar\omega - \varepsilon_{\mu,\lambda} - \varepsilon_{-\mu,\lambda}).
$$

The diamond-square Kitaev QSL of $C_{6}$ symmetry, Fig. 1(d), has two one-dimensional Raman-active modes $B_1$ and $B_3$. The basis functions remain the same as (21), but $R_{B_1}^\mu(\omega) \neq R_{B_3}^\mu(\omega)$, yielding the Raman response $I(\omega) = (E_{B_{1}\mu}^\mu)^2 + (E_{B_{2}\mu}^\mu)^2$.
Heisenberg antiferromagnets on the Kagome$^{18,20}$ and triangular$^{19,23}$ lattices as well as the honeycomb Kitaev QSL.$^5$ However, it is the case with an ordered Heisenberg antiferromagnet on the $C_{5v}$, Penrose lattice as well.$^{19}$ while the present $C_{4v}$-diamond-square, $D_{3h}$-harmonic-honeycomb,$^5,13)$ and $T$- and $O_h$-polyhedral$^{10}$ Kitaev QSLs exhibit strong polarization in their Raman responses. One and only multidimensional Raman-active mode, depending on the background lattice geometry rather than whether liquid or solid, is the key ingredient in depolarization of Raman response.

Now we consider which symmetry species of the $C_{4v}$-gauged-lattice$^{22}$ Raman spectrum originates in which combination—in the sense of energy, momentum, and symmetry—of spinon eigenmodes. We show in Fig. 3(a) the spinon dispersion relations of the gauge-ground Kitaev diamond-square model. Each spinon eigenmode $\epsilon_k, \lambda$ belongs to a double-valued irreducible representation of the double group $\tilde{P}_k$.$^{36}$ The gauge-ground Majorana Hamiltonian (4) in the real space for Fig. 1(d) is invariant to $\tilde{C}_{4v}$.$^{22}$ while each block $\tilde{H}_k$ of its Fourier transform (6) in the reciprocal space belongs to any of subsets $\tilde{P}_k \subseteq C_{4v}$. We demand that $\tilde{P}_k$ should keep $\tilde{H}_k$ invariant. Under the present Fourier transformation (5), $\tilde{P}_k$ should consist of primitive-translation-invariant gauged point symmetry operations, i.e., $\tilde{P}_k$ may be written as $\tilde{P}_0^k$ with $\tilde{P}_0^k \subseteq \tilde{P}_0 = C_{4v}$. $\tilde{P}_0^k$ amounts to $\tilde{C}_{2v}$ even at the highest symmetry points $\Gamma$ and $M$ to have one and only double-valued irreducible representation $E_2$ and thus bring doubly degenerate spinon excitations, as is shown in Fig. 3(a). As we move away from them, $\tilde{P}_0^k$ further reduces from $\tilde{C}_{2v}$ to $C_2$ or lower and the two-dimensional real irreducible representation $E_2$ splits into two one-dimensional complex ones $E^{(1)}_2$ and $E^{(2)}_2$ to lift the band degeneracy. Irreducible representations of double groups for the gauged diamond-square (reciprocal) lattice are detailed in Ref. 35.

Every LF scattering is mediated by a momentum-locked spinon geminate excitations $\tilde{\alpha}_{\lambda}^k, \tilde{\alpha}_{\lambda}^{k', j'}$ and characterized by its direct-product representation made of double-valued irreducible representations $\tilde{E}_2$ and $\tilde{E}_{2'}$ of the double group $\tilde{P}_0^k.$$^{22}$ Direct-product representations of a nonabelian group are not necessarily irreducible, even though the constituent representations are irreducible. Those relevant to spinons in pair with wavevectors $\pm \mathbf{k}_n$ decompose into single-valued irreducible representations of $\tilde{P}_0^k$, as is shown in Table I and illustrated in more detail in Ref. 36. Every single-valued irreducible representation of $\tilde{P}_0^k$ remains the same as that of the corresponding point symmetry group $\tilde{P}_0$, $\tilde{E}_2 \otimes \tilde{E}_{2'} = \bigoplus_j \tilde{E}_j \otimes \tilde{E}_{j'} = \bigoplus_j \tilde{E}_j$. Every direct product of the two same representations reads a sum of symmetric and/or antisymmetric representations, $\tilde{E}_2 \otimes \tilde{E}_{2'} = \bigoplus_j \tilde{E}_j \otimes \tilde{E}_{j'} = \bigoplus_j \tilde{E}_j$. The fermionic geminate excitations $\tilde{\alpha}_{\lambda}^k, \tilde{\alpha}_{\lambda}^{k', j'}$ and $\tilde{\alpha}_{\lambda}^k, \tilde{\alpha}_{\lambda}^{k', j'}$ should have antisymmetric representations. Thus and thus, along high symmetry points in the Brillouin zone, we can reveal the symmetry species of spinon geminate excitations $\tilde{\alpha}_{\lambda}^k, \tilde{\alpha}_{\lambda}^{k', j'}$, each compatible with one or more of irreducible representations of the full point symmetry group $C_{4v}$ (Table II) and bringing spectral weights $I(k, -k; \omega)$ [Fig. 3(c)] when their compatible symmetry mode(s) of $C_{4v}$ are Raman active. The Raman-active modes of the $C_{4v}$ gauged lattice are $\mathbf{B}_1$ and $\mathbf{B}_2$, selectively available from $\langle \varphi_{\mathbf{B}_1} \varphi_{\mathbf{B}_2} \rangle = (0, 0)$ and $(0, \pi)$, respectively. Suppose we consider two spinons

**Table I.** Direct-product representations made of double-valued irreducible representations $\tilde{E}_2 \otimes \tilde{E}_{2'}$ and their decompositions into single-valued irreducible representations $\tilde{E}_j$, which are underlined when they are relevant to Raman scattering, for gauged $k$-point symmetry groups $\tilde{P}_k$.

| $\tilde{P}_k$ | $\tilde{E}_2 \otimes \tilde{E}_{2'}$ | $\tilde{E}_j$ |
|--------------|-----------------------------|----------|
| $C_{2v}(1)$  | $E^{(1)}_2 \otimes E^{(2)}_2$ | $E_2$    |
| $C_{2v}(2)$  | $E^{(1)}_2 \otimes E^{(2)}_2$ | $E_2$    |
| $C_{4v}$     | $E^{(1)}_2 \otimes E^{(2)}_2$ | $E_2$    |

**Table II.** Compatibility relations between irreducible representations of $C_{4v}$ and those of its subgroups $C_{2v}$ and $C_2$. The gauged diamond-square lattice Fig. 1(d) is invariant to $\tilde{C}_{4v}$ while each block $\tilde{H}_k$ of the gauge-ground Majorana Hamiltonian (4) belongs to any of subgroups $\tilde{P}_k \subseteq C_{4v}$.

| $C_{4v}$ | $A_1$ | $A_2$ | $B_1$ | $B_2$ | $E$ |
|----------|-------|-------|-------|-------|-----|
| $C_{2v}$ | $A_1$ | $A_2$ | $B_1$ | $B_2$ | 2B  |
| $C_{2v}(1)$ | A | A | A | B | 2B  |


α†(k,k)/√2:λ and α†−(k,k)/√2:λ′ (λ,λ′= 1, 2) on the ways from Γ to M of C2v symmetry. The E₁(2)⊗E1(2) pairs belong to the symmetry species B of C2v, compatible with LF-Raman-inactive A₂ and Raman-active B₁ of C2v, and therefore bring spectral weights \[ \sum_{k_1=1}^{2} \mathcal{I}[k, -k; (ξ_{k_1} + e_{-k_1})/\hbar] \equiv 2\mathcal{I}[k; (ξ_{k_1} + e_{k_2})/\hbar] \] at \((φ_{sc}, φ_{nc}) = (0, 0)\). The E1(3)⊗E1(3) pairs belong to the symmetry species A of C2v, compatible with LF-Raman-inactive (LF-Rayleigh) A₁ and Raman-active B₂ of C2v, and therefore bring spectral weights \[ \sum_{k_1=1}^{2} \mathcal{I}[k_1, 2k_2; (ξ_{k_1} + e_{k_2})/\hbar] \equiv \mathcal{I}[k; 2e_{k_1}/\hbar] + \mathcal{I}[k; 2e_{k_2}/\hbar] \] at \((φ_{sc}, φ_{nc}) = (0, ½)\), where single spinon excitation modes \(e_{k_1}\) and \(e_{k_2}\) each separately appear.

The key ingredient of depolarized Raman response is one and only multidimensional Raman-active mode of geometric origin. It occurs in honeycomb Heisenberg antiferromagnets without any frustration, while it breaks down in Kitaev QSLs in nontriangular geometry. Polarized Raman spectra of Kitaev QSLs serve to identify their single spinon excitation modes as functions of their momenta, even though the total momentum of each pair of mediating spinons is locked to zero. Polarized photons can distinguish between spinon geminate excitations of different symmetries and therefore reveal the projective symmetries of their constituent spinons. While the depolarization of Raman response in Kitaev QSLs trivially breaks down with higher-order vertices under increasing itinerancy and decreasing correlation, the breakdown is possible within the solvable Hamiltonian and LF scheme. On the one hand nontriangular geometries are realizable on honeycomb lattices with site dilution and/or bond disorder, but on the other hand Kitaev QSLs in the cylinder and only multidimensional Raman-active mode of geometric origin are ideal targets of the present approach.

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1) A. Kitaev, Ann. Phys. (N.Y.) 321, 2 (2006).
2) J. Knolle and R. Moessner, Annu. Rev. Condens. Matter Phys. 10, 451 (2019).
3) Y. Motome and J. Nasu, J. Phys. Soc. Jpn. 89, 032002 (2020).
4) L. Savary and L. Balents, Rep. Prog. Phys. 80, 016502 (2017).
5) J. Knolle, G.-W. Chern, D. L. Kovrizhin, R. Moessner, and N. B. Perkins, Phys. Rev. Lett. 113, 187201 (2014).
6) P. A. Fleury and R. Louden, Phys. Rev. 166, 514 (1968).
7) B. S. Shastry and B. I. Shraiman, Phys. Rev. Lett. 65, 1068 (1990).
8) B. S. Shastry and B. I. Shraiman, Int. J. Mod. Phys. B 5, 365 (1991).
9) B. Perreault, J. Knolle, N. B. Perkins, and F. J. Burnell, Phys. Rev. B 92, 094439 (2015).
10) S. Yamamoto and T. Kimura, J. Phys.: Conf. Ser. 1220, 012003 (2019).
11) B. Perreault, J. Knolle, N. B. Perkins, and F. J. Burnell, Phys. Rev. B 94, 060408(R) (2016).
12) B. Perreault, J. Knolle, N. B. Perkins, and F. J. Burnell, Phys. Rev. B 95, 104427 (2016).
13) B. Perreault, S. Rachel, F. J. Burnell, and J. Knolle, Phys. Rev. B 95, 184429 (2017).
14) I. Rousochatzakis, S. Kourtis, J. Knolle, R. Moessner, and N. B. Perkins, Phys. Rev. B 100, 045117 (2019).
15) J. G. Rau, E. K.-H. Lee, and H.-Y. Kee, Phys. Rev. Lett. 112, 077204 (2014).
16) H. Tomishige, J. Nasu, and A. Koga, Phys. Rev. B 97, 094403 (2018).
17) K. Slagle, W. Choi, L. E. Chern, and Y. B. Kim, Phys. Rev. B 97, 115159 (2018).
18) O. Cépas, J. O. Haerter, and C. Lhuillier, Phys. Rev. B 77, 172406 (2008).
19) N. Perkins and W. Brenig, Phys. Rev. B 77, 174412 (2008).
20) W.-H. Ko, Z.-X. Liu, T.-K. Ng, and P. A. Lee, Phys. Rev. B 81, 024414 (2010).
21) G. Misguich and F. Mila, Phys. Rev. B 77, 134421 (2008).
22) See Supplemental Material Sect. S1 where gauged point symmetry operations on the gauge-group Majorana fermionic Hamiltonian (4) are illustrated in detail.
23) X.-G. Wen, Phys. Rev. B 65, 165113 (2002).
24) F. Wang and A. Vishwanath, Phys. Rev. B 74, 174423 (2006).
25) H. Yao and S. A. Kivelson, Phys. Rev. Lett. 99, 247203 (2007).
26) J. Nasu and Y. Motome, Phys. Rev. Lett. 115, 087203 (2015).
27) S. Yang, D. L. Zhou, and C. P. Sun, Phys. Rev. B 76, 180404(R) (2007).
28) L. N. Karnaíkho, Europhys. Lett. 102, 57007 (2013).
29) A. Bao, H.-S. Tao, H.-D. Liu, X.Z. Zhang, and W.-M. Liu, Sci. Rep. 4, 6918 (2014).
30) G. B. Halász, J. T. Chalker, and R. Moessner, Phys. Rev. B 90, 035145 (2014).
31) O. Petrova, P. Mellado, and O. Tchernyshyov, Phys. Rev. B 90, 134404 (2014).
32) F. Zachoche and M. Vojta, Phys. Rev. B 92, 014403 (2015).
33) K. O’Brien, M. Hermanns, and S. Trebst, Phys. Rev. B 93, 085101 (2016).
34) S. Yamamoto, J. Ohara, and M. Ozaki, J. Phys. Soc. Jpn. 79, 044709 (2010).
35) See Supplemental Material Sect. S2 where irreducible representations of the double groups \(C_{4v}, C_{2v}, C_{2}\) are listed with their characters.
36) See Supplemental Material Sect. S3 where the point symmetry group \(P, k\)-point symmetry group \(P_{k}\), and gauged \(k\)-point symmetry group \(P_{k}^G\) for the diamond-square (reciprocal) lattice Fig. 1(d) are detailed, and then, spinon-geminate-excitation-relevant direct-product representations of the double groups \(C_{2v}^{1(2)}(τ = z, x, y, a, b)\) and \(C_{2v}^{1(2)}^{(a) ab}\) are listed with their characters and decomposed into single-valued irreducible representations of the corresponding point symmetry groups \(C_{2v}^{1(2)}\) which are compatible with one or more of irreducible representations of the full point symmetry group \(C_{2v}\).
37) M. S. Dresselhaus, G. Dresselhaus, and A. Jorio, Group Theory: Application to the Physics of Condensed Matter (Springer, Berlin, 2008).
38) H. Yao, S.-C. Zhang, and S. A. Kivelson, Phys. Rev. Lett. 102, 217202 (2009).
39) F. L. Pedrocchi, S. Chesi, and D. Loss, Phys. Rev. B 84, 165414 (2011).
40) M. Udagawa, Phys. Rev. B 98, 220404(R) (2018).
41) P. Mellado, O. Petrova, and O. Tchernyshyov, Phys. Rev. B 91, 041103(R) (2015).
42) T. P. Devereaux and R. Hackl, Rev. Mod. Phys. 79, 175 (2007).
43) F. Vernay, T. P. Devereaux, and M. J. P. Gingras, J. Phys. Condens. Matter 19, 145243 (2007).
44) T. Inoue and S. Yamamoto, arXiv: 2004.09850.
45) A. J. Willans, J. T. Chalker, and R. Moessner, Phys. Rev. B 84, 115146 (2011).
46) M. Gohlke, R. Verresen, R. Moessner, and F. Pollmann, Phys. Rev. Lett. 119, 157203 (2017).
47) K. Suzuki and S. Yamamoto, J. Phys.: Conf. Ser. 1220, 012046 (2019).
Supplemental Material for
Raman Scattering Polarization and Single Spinon Identification in
Two-Dimensional Kitaev Quantum Spin Liquids

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We detail the projective-symmetry-group technique employed in the text with many schematic demonstrations and apply it to the Z₂-gauged diamond-square lattice in the context of its Raman response mediated by spinon geminate excitations.

S1. Projective Symmetry Operations on Gauge-Ground Kitaev Spin Planes

We discuss the Kitaev Hamiltonian

\[ \mathcal{H} = -J \sum_{\langle r_{i}, r_{j}, r_{k}, r_{l} \rangle} \sigma_{r_{i}, \lambda}^{i} \sigma_{r_{j}, \lambda}^{j} \sigma_{r_{k}, \lambda}^{k} \sigma_{r_{l}, \lambda}^{l}, \]  

(S1)
on various two-dimensional lattices, where \((\sigma_{r_{i}, \lambda}^{i}, \sigma_{r_{j}, \lambda}^{j}, \sigma_{r_{k}, \lambda}^{k}, \sigma_{r_{l}, \lambda}^{l})\) \((i = 1, \ldots, N^{2} \equiv L/g, \lambda = 1, \ldots, g)\) are the Pauli matrices attached to the \(i\)th site in the \(i\)th unit at \(r_{i}\) and \(\langle r_{i}, r_{j}, r_{k}, r_{l} \rangle \lambda \) runs over 3L/2 nearest-neighbor bonds. Denoting the primitive translation vectors in each lattice by \(a\) and \(b\), we adopt a periodic boundary condition, \(r_{i} + N a = r_{i} + Nb = r_{i}\). The number of sites \(L\) reads \(qN^{2} = 2\) [Figs. S1(a)], \(q = 6\) [Figs. S1(b)], \(q = 12\) [Figs. S1(c)], and \(q = 4\) [Figs. S1(d)].

When we introduce four Majorana fermions at each site \(\gamma_{\langle r_{i}, r_{j}, r_{k}, r_{l} \rangle} \gamma\), we find that the gauged honeycomb belongs to the double point symmetry \([S1(d)]\).

The flux operator defined for an \(N_{g}\)-sided polygon \(p\)

\[ \hat{W}_{p} = \prod_{\langle r_{i}, r_{j}, r_{k}, r_{l} \rangle} \sigma_{r_{i}, \lambda}^{i} \sigma_{r_{j}, \lambda}^{j} \sigma_{r_{k}, \lambda}^{k} \sigma_{r_{l}, \lambda}^{l}, \]  

(S6)
also behaves as a classical variable, \(W_{p} = \pm 1\) or \(\pm i\) according as \(N_{g}\) is even or odd. The eigenspectrum of (S3) depends on the bond configuration \(\{\hat{u}_{\langle r_{i}, r_{j}, r_{k}, r_{l} \rangle}^{\alpha} \}\) through the flux configuration \(\{\hat{W}_{p} \}\). The ground-state flux configuration is such that all \(W_{p}\)'s are \(-1, +1\), or either of \(+i\) and \(-i\) according as their \(N_{g}\)'s are \(4, 4, 2, \) or \(2, 1\) with \(l \in \mathbb{N}\) [cf. Figs. S1(a)–S1(d)].

The pure-, triangle-, and square-hexagon-honeycomb lattices are of \(C_{6v}\) point symmetry \([S1(a)–S1(c)]\), while the diamond-square lattice is of \(C_{6v}\) point symmetry \([S1(d)]\). Once these lattices are gauged in the context of the Kitaev quantum spin liquid, they no longer belong to their original point symmetry groups \(P_{\text{org}}\) but may become invariant under gauged point symmetry operations \(\hat{P} \in \hat{P}_{\text{org}} \) with \(\hat{P} \notin \hat{P}_{\text{org}}\). Note that \(\hat{P}\) is not necessarily equal to \(P_{\text{org}}\).

Let us start with a simple example. Figure S1(a) illustrates gauged point symmetry operations on the gauge-ground Kitaev honeycomb Hamiltonian. Suppose we rotate it by \(\pi/3\) about the normal vector, denoted by \(C_{6}\), and then gauge some Majorana fermions as \(c_{r_{i}, \lambda} \rightarrow -c_{r_{i}, \lambda}\), or equivalently, change the signs of their relevant bonds as \(u_{\langle r_{i}, r_{j}, r_{k}, r_{l} \rangle}^{\alpha} \rightarrow -u_{\langle r_{i}, r_{j}, r_{k}, r_{l} \rangle}^{\alpha}\), so as to recover the initial bond configuration. Any local gauge transformation \(c_{r_{i}, \lambda} \rightarrow -c_{r_{i}, \lambda}\) reverses its three surrounding bond variables. Given a point symmetry operation \(P \in C_{6v}\), there are two such local gauge transformations, denoted by \(\pm \Lambda(P)\). Let us abbreviate a couple of these serial transformations as \(\Lambda(P)P \equiv \tilde{P}\) and \(-\Lambda(P)P \equiv \tilde{P}\) and denote them unifiedly as \(\tilde{P}\). We find that the gauged honeycomb belongs to the double group \(C_{6h}\), and therefore \(\hat{P} = \hat{P}_{\text{org}}\) in this case.

Figure S1(b) illustrates mirror operations as well as gauged rotations on the gauge-ground Kitaev triangle-honeycomb Hamiltonian. Every mirror operation reverses all the flux variables \(W_{p}\) of the constituent triangles, while any local gauge transformation \(c_{r_{i}, \lambda} \rightarrow -c_{r_{i}, \lambda}\) results in reversing the signs of bonds in pair in the three surrounding polygons to keep their flux variables \(W_{p}\) unchanged. We find that the symmetry group of the gauged triangle honeycomb is not \(C_{6v}\) but \(C_{6h}\). Note that \(\hat{P} \neq \hat{P}_{\text{org}}\) in this case. In the same manner, we find that \(\hat{P} = \hat{P}_{\text{org}} \equiv \hat{C}_{6v}\) for the gauged square-hexagon honeycomb and \(\hat{P} = \hat{P}_{\text{org}} \equiv \hat{C}_{6h}\) for the gauged diamond square.
\( W_p = +1, -1, -1 \)

\( (b) \bar{C}_6 \)
\[
\begin{align*}
C_{2} \rightarrow -\Lambda(C_{2}) \\
C_{6} \rightarrow -\Lambda(C_{6}) \\
\sigma_{x} \rightarrow \Lambda(\sigma_{x}) \\
\sigma_{y} \rightarrow \Lambda(\sigma_{y}) \\
\end{align*}
\]

\( W_{p} = +1 - 1 - 1 \)

\( \overline{C_{6v}} = C_{6v} \)
Fig. S1. Gauged rotations and mirror operations of gauge-ground Kitaev spin planes consisting of the pure (a)-, triangle (b)-, square-hexagon (c)-honeycomb and diamond-square (d) lattices. Mirror operations \( \sigma \in C_{6v} \) of (b) can be followed by no such gauge transformation as to recover the initial bond configuration. Rotations and mirror operations \( C_4, C_4^{-1}, \sigma_x, \sigma_y \in C_{4v} \) of (d) can be followed by no such gauge transformation as to recover the initial bond configuration with the primitive translation vectors remaining unchanged.
S2. Irreducible Representations of Double Groups for the Gauged Diamond-Square Lattice

We denote the orders of a point symmetry group $P$ and its double covering group $\overline{P}$ by $g^P$ and $g^\overline{P}$, respectively. Suppose the double cover $\overline{P}$ to be the $Z_2$-gauge extension of $P \subset O(3)$. Two group elements $P_1 \in P$ and $P_2 \in \overline{P}$ are conjugate when we find such an element $\overline{P} \in \overline{P}$ as to satisfy

$$\overline{P}_1 \equiv \overline{P} \overline{P} \overline{P}^{-1}.$$  \hfill (S7)

Every set of conjugate elements forms a class. The classes of the double group of $C_{4v}$ and its subgroups read

$$\overline{C}_{4v} : \{E, [E], [2\overline{C}_4], [2\overline{C}_2], [\overline{C}_2, \overline{C}_2], \}
\overline{C}_{2v} : \{E, [E], [\overline{C}_2], [\overline{C}_2, \overline{C}_2], [\overline{C}_2, \overline{C}_2], \}
\overline{C}_2 : \{E, [E], [\overline{C}_2], \}$$

Supposing the $q$th class $C_q$ ($q = 1, \cdots, n_P^P$) of $P$ to consist of $h_q$ elements, it reads $\{h_q \overline{P}_1, h_q \overline{P}_2, \cdots, h_q \overline{P}_{h_q}\}$.

The number of (complex) irreducible representations equals how many classes are in the group. Since all the single-valued (complex) irreducible representations of $P$, amounting to $n_P^P$, remain unchanged in $\overline{P}$, we find $n_P^P = n_P^P$ double-valued (complex) irreducible representations in $\overline{P}$. When we denote the $i$th (complex) irreducible representation of $P (\overline{P})$ by $\Xi_i (\overline{\Xi}_i)$ and its dimensionality by $d_P^{\overline{P}} (d_{\overline{P}}^{\overline{P}})$, we have

$$\sum_{i=1}^{n_{C_{4v}}^{C_{4v}}} \left( d_{\overline{P}}^{\overline{P}} \right)^2 = g^{C_{4v}} = 8,$$

$$\sum_{i=1}^{n_{C_{2v}}^{C_{2v}}} \left( d_{\overline{P}}^{\overline{P}} \right)^2 = g^{C_{2v}} = 4,$$

$$\sum_{i=1}^{n_{C_2}^{C_2}} \left( d_{\overline{P}}^{\overline{P}} \right)^2 = g^{C_2} = 2,$$

in an attempt to determine the dimensionality of the double-valued (complex) irreducible representations $d_P^{\overline{P}} (i = n_C^P + 1, \cdots, n_P^P)$. The characters of $\overline{C}_2$ are such that

$$\chi_{\overline{C}_2} (\overline{P}) = \chi_{\overline{P}} (P) (i = 1, \cdots, n_P^P),$$

$$\chi_{\overline{C}_2} (\overline{P}) = -\chi_{\overline{P}} (P) (i = n_C^P + 1, \cdots, n_P^P).$$

When $\overline{P}$ and $P$ belong to the same class, i.e., $\chi_{\overline{C}_2} (\overline{P}) = \chi_{\overline{P}} (P)$, we immediately find

$$\chi_{\overline{C}_2} (\overline{P}) = \chi_{\overline{P}} (P) = 0 (i = n_C^P + 1, \cdots, n_P^P).$$  \hfill (S13)

The character orthogonality theorems of the first and second kinds read\(^4\)

$$\sum_{\overline{P} \in \overline{C}_q} \frac{d_P^{\overline{P}}}{2} \chi_{\overline{P}} (\overline{P}) = g^{\overline{P}} \delta_{qr},$$

$$\sum_{\overline{P} \in \overline{C}_q} \frac{d_P^{\overline{P}}}{2} \chi_{\overline{P}} (\overline{P}) = g^{\overline{P}} \delta_{q'r',}.$$  \hfill (S14)

When we denote the $h_q$ elements of $C_q$ distinguishably as $\{\overline{P}_j^q, \cdots, \overline{P}_{h_q}^q\}$, we can define structure constants as

$$\sum_{\overline{P} \in \overline{C}_q} \overline{P}_q^j \overline{P}_q^r \overline{P}_q^{(f)} = h_q \sum_{i=1}^{h_q} \sum_{j=1}^{h_q} \sum_{r=1}^{h_q} \overline{P}_q^i \overline{P}_q^j \overline{P}_q^{(f)}.$$  \hfill (S16)

to have another relation,

$$h_q h_q \overline{P}_{\overline{P}} (C_q) \overline{P}_q^j (C_q) = d_P^{\overline{P}} \sum_{i=1}^{h_q} h_q \sum_{j=1}^{h_q} \sum_{r=1}^{h_q} \overline{P}_q^i \overline{P}_q^j \overline{P}_q^{(f)}.$$  \hfill (S17)

With Eqs. (S13), (S14), (S15), and (S17) in mind, we can obtain characters of both single- and double-valued (complex) irreducible representations of any double group $P$. We list in Tables S1–S3 characters of irreducible representations of the double group $\overline{C}_{4v}$ and those of its subgroups $\overline{C}_{2v}$ and $\overline{C}_2$ with particular emphasis on the relation between $P$ and $\overline{P}$.

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**Table S1.** Irreducible representations of the double group $\overline{C}_{4v}$ and their characters.

| $C_{4v}$ | $|E|$ | $|2C_4|$ | $|2C_2|$ | $|\overline{C}_2|$ | $|\overline{C}_2|$ |
|----------|-------|-------|-------|-------|-------|
| $\overline{C}_{4v}$ | $\overline{C}_{4v}$ | $\overline{C}_{4v}$ | $\overline{C}_{4v}$ | $\overline{C}_{4v}$ | $\overline{C}_{4v}$ |
| $A_1$ | 1 | 1 | 1 | 1 | 1 |
| $A_2$ | 1 | 1 | -1 | -1 | -1 |
| $B_1$ | 1 | -1 | 1 | -1 | -1 |
| $B_2$ | 1 | -1 | -1 | 1 | -1 |
| $E$ | 2 | 0 | 0 | 0 | 0 |

**Table S2.** Irreducible representations of the double group $\overline{C}_{2v}$ and their characters.

| $\overline{C}_{2v}$ | $|E|$ | $|2C_2|$ | $|\overline{C}_2|$ | $|\overline{C}_2|$ |
|----------|-------|-------|-------|-------|
| $\overline{C}_{2v}$ | $\overline{C}_{2v}$ | $\overline{C}_{2v}$ | $\overline{C}_{2v}$ | $\overline{C}_{2v}$ |
| $A_1$ | 1 | 1 | 1 | 1 |
| $A_2$ | 1 | 1 | -1 | -1 |
| $B_1$ | 1 | -1 | 1 | -1 |
| $B_2$ | 1 | -1 | -1 | 1 |
| $E_1$ | 2 | 0 | 0 | 0 |

**Table S3.** Irreducible representations of the double group $\overline{C}_2$ and their characters.

| $\overline{C}_2$ | $|E|$ | $|C_2|$ | $|\overline{C}_2|$ |
|----------|-------|-------|-------|
| $\overline{C}_2$ | $\overline{C}_2$ | $\overline{C}_2$ | $\overline{C}_2$ |
| $A_1$ | 1 | 1 | 1 |
| $B_1$ | 1 | 1 | -1 |
| $\bar{E}_1$ | 2 | 0 | 0 |

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\(^4\) character orthogonality theorem of the fourth and second kinds
S3. Direct-Product Representations of Double Groups for the Gauged Diamond-Square Lattice

Inelastic visible-light scatterings within the Loudon-Fleury scheme5,8 are mediated by “momentum-locked” spinon geminate excitations $\alpha_{k}^{i} \alpha_{k}^{j}$. In order to analyze the polarized Raman spectra of the gauged diamond-square lattice Fig. S1(d), we formulate direct-product representations of gauged k-point symmetry groups $P_{k}' \subseteq P \equiv \mathbb{C}_{4v}$ at high symmetry points of the diamond-square reciprocal lattice. When we need to specify the principal axis $\tau$ for n-fold rotations and/or the normal vector $\sigma$ for mirror operations, we replace the usual Schönflies notation $\mathbb{C}_{n}(r\sigma)$ by $\mathbb{C}_{n}(r\sigma')$ for the sake of saving space.

The isotropy group of $k$ in the first Brillouin zone $P_{k}$ consists of symmetry operations $P_{k}$ such that

$$P_{k}k = k + K \equiv k$$  \hspace{1cm} (S18)

with $K$ being $0$ or a reciprocal lattice vector. The $\mathbb{Z}_{2}$-gauged $k$-point symmetry group $P_{k}'$ keeps the $k_{0}$ block $\mathcal{H}^{k}_{0}$ of the gauge-ground Majorana Hamiltonian (S3) invariant. Under the present Fourier transformation (S4), $P_{k}'$ should consist of primitive-translation-invariant gauged point symmetry operations, i.e., $P_{k}'$ may be written as $P_{0}'$ with $P_{0}' \subseteq P_{0}$. $P_{0}'$ equals the full point symmetry group of the background lattice $P_{0}\equiv \mathbb{C}_{4v}$ and reads $\mathbb{C}_{4v}$ for the diamond-square lattice. Figure S1(d) shows that the gauge transformations $\mathcal{A}(C_{4}), \mathcal{A}(C_{4}^{-1}), \mathcal{A}(\sigma_{x})$, and $\mathcal{A}(\sigma_{y})$ recover the initial ground gauge configuration but break the primitive translation symmetry. Such gauged point symmetry operations keep none of the original $k_{0}$ blocks $\mathcal{H}_{k}^{k_{0}}$ ($k = 1, \cdots, N^{2}$) invariant and demand that the Fourier transformation (S4) be modified. Thus, every $P_{0}'$ is limited to a proper subset of $P_{0}$. $P_{0}' \subseteq P_{0}$ for the diamond-square lattice. We show in Fig. S2 how $P_{k}$ and $P_{0}'$ read at high symmetry points of the diamond-square reciprocal lattice. $P_{k}'$ becomes $\mathbb{C}_{2v}$ even at the highest symmetry points $\Gamma$ and $M$. The thus-defined projective symmetry group of the $\mathbb{Z}_{2}$-gauged diamond-square lattice reads $L \wedge \mathbb{C}_{2v}$ with $L = \{mn | m \in \mathbb{Z} \} \times \{nb | n \in \mathbb{Z} \}$.

Each momentum-locked spinon geminate excitation $\alpha_{k}^{i} \alpha_{k}^{j}$ consists of double-valued irreducible representations of the gauged $k$-point symmetry group $P_{0}'$ at points $\pm k_{s}$ and have a direct-product representation, $\mathcal{E}_{i} \otimes \mathcal{E}_{j}$ $(i, i' = n_{0} + 1, \cdots, n_{c})$. Since its characters read

$$\chi_{\mathcal{E}_{i} \otimes \mathcal{E}_{j}'}(P_{0}') = \chi_{\mathcal{E}_{i}'}(P_{0}') \chi_{\mathcal{E}_{j}'}(P_{0}') = \chi_{\mathcal{E}_{i} \otimes \mathcal{E}_{j}}(P_{0})$$  \hspace{1cm} (S19)

it results in single-valued irreducible representations of $P_{0}'$, i.e. those of the corresponding point symmetry group $P_{0}'$. 

$$\mathcal{E}_{i} \otimes \mathcal{E}_{j} = \{ \mathcal{E}_{i} \otimes \mathcal{E}_{j} \} \otimes \{ \mathcal{E}_{i} \otimes \mathcal{E}_{j} \}$$  \hspace{1cm} (S21)

$$[\mathcal{E}_{i} \otimes \mathcal{E}_{j}] = \{ \mathcal{E}_{i} \} \times \{ \mathcal{E}_{i} \}$$  \hspace{1cm} (S22)

$$[\mathcal{E}_{i} \otimes \mathcal{E}_{j}] = \{ \mathcal{E}_{i} \} \times \{ \mathcal{E}_{j} \}$$  \hspace{1cm} (S23)

$$[\mathcal{E}_{i} \otimes \mathcal{E}_{j}] = \{ \mathcal{E}_{i} \} \times \{ \mathcal{E}_{j} \}$$  \hspace{1cm} (S24)

$$[\mathcal{E}_{i} \otimes \mathcal{E}_{j}] = \{ \mathcal{E}_{i} \} \times \{ \mathcal{E}_{j} \}$$  \hspace{1cm} (S25)

![Fig. S2. The diamond-square lattice of $\mathbb{C}_{4v}$ point symmetry with primitive cells encircled by dotted lines. $k$-point symmetry groups (isotropy groups of $k$) $P_{k}$ at high symmetry points $k$ of the diamond-square reciprocal lattice, each consisting of symmetry operations $P_{k}$ such that $P_{k}k = k + K \equiv k$ with $K$ being $0$ or a reciprocal lattice vector. $\mathbb{Z}_{2}$-gauged $k$-point symmetry groups $P_{k}'$ under the Fourier transformation (S4), i.e., $P_{k}'$, at high symmetry points $k_{0}$ of the diamond-square reciprocal lattice, each keeping the $k_{0}$ block $\mathcal{H}_{k_{0}}$ of the Fourier-transformed gauge-ground Majorana Hamiltonian (S5) invariant. Note that $P_{k}' \subseteq P_{k} \subseteq P_{0}\equiv \mathbb{C}_{4v}$.](image_url)
We can obtain characters of any direct-product representation using Eqs. (S24) and (S25) as well as (S19), which of our interest are listed in Tables S4 and S5. Direct-product representations for germinate excitations of different Majorana spinon eigenmodes are not necessarily made of different irreducible representations but may be made of the same ones. Those made of different irreducible representations can be decomposed into irreducible representations by Eq. (S20), while those made of the same ones by Eqs. (S22) and (S23). Direct-product representations for germinate excitations of degenerate Majorana spinon eigenmodes are also the latter case. The thus-obtained decompositions into irreducible representations are all listed in Table S6.

Any symmetry species \( \Xi_j \) of the \( \overline{C_{4v}} \)-gauged-lattice Raman spectrum belongs to \( C_{4v} \), while spinon germinate excitations yield one or more irreducible representations \( \bigoplus_j \Xi_j \) of its subgroup \( P'_0 \subset P_0 = C_{4v} \). When \( P'_0 \subset C_{4v} \), every irreducible representation \( \Xi_j \) of \( P'_0 \) is compatible with one or more irreducible representations \( \Xi_j \) of \( C_{4v} \), in other words, every irreducible representation \( \Xi_j \) of \( C_{4v} \) reads a direct sum of one or more irreducible representations \( \Xi_j \in P'_0 \). When we denote the representation \( \Xi_j \) of \( C_{4v} \) within its subgroup \( P'_0 \) by \( \Xi_j \downarrow P'_0 \), the compatibility relation reads

\[
\Xi_j \downarrow P'_0 = \bigoplus_{k \in \Xi_j} \sum_{q=1}^{n_k} \frac{h_q}{P'_0} \chi_{P'_0}(C_q) \chi_{\Xi_j}(C_q)
\]

We summarize in Table S7 the thus-obtained compatibility relations between \( C_{4v} \) and its subgroups \( C_{2v} \) and \( C_2 \).

| Table S4. Direct-product representations made of double-valued irreducible representations of the double group \( C_{2v} \) and their characters. |
|---------------------------------------------------------------|
| \( \Xi_j \downarrow \Xi_{j'} \) | \( [E] \) | \( [E] \) | \( [C_2] \) |
| \( [E] \oplus [E] \) | 3 | -1 | -1 | -1 |
| \( [E] \oplus \left[ E_{1/2} \right] \) | 1 | 1 | 1 | 1 |

| Table S5. Direct-product representations made of double-valued irreducible representations of the double group \( C_2 \) and their characters. |
|---------------------------------------------------------------|
| \( \Xi_j \downarrow \Xi_{j'} \) | \( [E] \) | \( [E] \) | \( [C_2] \) |
| \( [E] \oplus [E] \) | 1 | -1 | -1 |

| Table S6. Direct-product representations made of double-valued irreducible representations \( \Xi_j \downarrow \Xi_{j'} \) and their decompositions into single-valued irreducible representations \( \Xi_j \), which are underlined when they are relevant to Raman scattering, for gauged \( k \)-point symmetry groups \( P'_0 \). |
|---------------------------------------------------------------|
| \( P'_0 \) | \( \Xi_j \downarrow \Xi_{j'} \) | \( \Xi_{j'} \downarrow \Xi_{j''} \) |
| \( \Xi_2(2v) \) | \( \Xi_2(2v) \downarrow \Xi_2(2v) \) | \( \Xi_2(2v) \downarrow \Xi_2(2v) \) |
| \( \Xi_2(2) \) | \( \Xi_2(2) \downarrow \Xi_2(2) \) | \( \Xi_2(2) \downarrow \Xi_2(2) \) |

| Table S7. Compatibility relations between irreducible representations of \( C_{4v} \) and those of its subgroups \( C_{2v} \) and \( C_2 \). The \( Z_2 \)-gauged diamond-square lattice is invariant to \( C_{4v} \) \( \equiv \bigoplus \) \( P_0 \) [Fig. S1(d)], while each block \( \Xi_{j'} \) of the gauge-group Majorana Hamiltonian (S3) belongs to its subgroup \( P'_0 \), where \( P'_0 \subset P_0 \subset \bigoplus \). When we adopt the Fourier transformation (S4), \( P'_0 \) reads \( \Xi'_0 \) with \( \Xi'_0 = \Xi_0 \). The Raman-active modes of \( C_{4v} \) are underlined and their compatible symmetry species of \( P'_0 \) determine which type of spinon germinate excitations is relevant to which symmetry species of the Raman scattering intensities. |
|---------------------------------------------------------------|
| \( C_{4v} \) | \( A_1 \) | \( A_2 \) | \( B_1 \) | \( B_2 \) | \( E \) |
| \( C_{2v} \) | \( A_1 \) | \( A_2 \) | \( A_1 \) | \( B_1 \) | \( B_2 \) |
| \( C_2 \) | \( A \) | \( A \) | \( A \) | \( 2B \) |
| \( C_{2v}, C_2 \) | \( A \) | \( A \) | \( A \) | \( A \) |

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1) P. Malliando, O. Petrova, and O. Tchernyshev, Phys. Rev. B 91, 041103(R) (2015).
2) X.-G. Wen, Phys. Rev. B 65, 165113 (2002).
3) O. Petrova, P. Malliando, and O. Tchernyshev, Phys. Rev. B 90, 134404 (2014).
4) M. S. Dresselhaus, G. Dresselhaus, and A. Jorio, Group Theory: Application to the Physics of Condensed Matter (Springer, Berlin, 2008).
5) P. A. Fleury and R. Loudon, Phys. Rev. 166, 514 (1968).
6) B. S. Shastry and B. I. Shraiman, Phys. Rev. Lett. 65, 1068 (1990).
7) B. S. Shastry and B. I. Shraiman, Int. J. Mod. Phys. B 5, 365 (1991).
8) J. Knolle, G.-W. Chern, D. L. Kovrizhin, R. Moessner, and N. B. Perkins, Phys. Rev. Lett. 113, 187201 (2014).