Polarized Raman Response of Two-Dimensional Quasiperiodic Antiferromagnets:
Configuration-Interaction versus Green’s Function Approaches
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We study Raman response of Heisenberg antiferromagnets on the $C_{5v}$, Penrose and $C_{mm}$ Ammann-Beenker lattices within and beyond the Loudon-Fleury second-order perturbation scheme intending to explore optical features peculiar to quasiperiodic magnets. Within the Loudon-Fleury mechanism, we find one and only Raman-active mode of $E_2$ symmetry without any dependence on linear incident and scattered polarizations. Beyond the Loudon-Fleury mechanism, two more symmetry species $A_1$ and $A_2$ are activated via dynamic ring exchange and chiral spin fluctuations, respectively, which can be extracted by the use of circular as well as linear polarizations. We employ Green’s functions on one hand and configuration-interaction wavefunctions on the other hand to calculate the multimagnon contributions to inelastic light scatterings. Demonstrating the great advantage of the configuration-interaction scheme, we reveal that a major portion of the Shastry-Shraiman fourth-order Raman intensity is mediated by multimagnon fluctuations.

A variety of quasiperiodic magnetic crystals\(^1\)–\(^4\) have renewed the theoretical exploration of novel magnetism of geometric origin. The theoretical and Ammann-Beenker lattices in two dimensions attract much interest in this context. Their Ising\(^5\)\(^,\)\(^6\) and Heisenberg\(^7\)\(^–\)\(^10\) models were calculated by the linear spin-wave (SW) theory and Monte Carlo (MC) methods, whereas their Hubbard models\(^11\)\(^,\)\(^12\) were investigated within a mean-field approximation. In the context of inelastic-neutron-scattering experiments, SW findings for the dynamic structure factor of the Ammann-Beenker Heisenberg antiferromagnet reveal intriguing excitation features possibly due to the quasiperiodicity,\(^13\) the coexistence of linear soft modes near the magnetic Bragg peaks at low frequencies, self-similar structures at intermediate frequencies, and flat bands at high frequencies. We may take further interest in inelastic light scatterings in quasiperiodic magnets, which strongly reflect their lattice symmetry and potentially bring brandnew information by virtue of the light polarization degrees of freedom.

Recent technical progress of manipulating optical laticess\(^13\)\(^–\)\(^15\) also deserves special mention. Two-dimensional (2D) quasiperiodic potentials with five-\(^16\) or eight-fold\(^17\)\(^,\)\(^18\) rotational symmetry were theoretically designed in terms of standing-wave lasers and indeed observed via Bragg diffraction.\(^14\)\(^,\)\(^16\) By trapping ultracold bosonic or fermionic atoms in optically tunable potentials, we can change the on-site interaction in two dimensions attract much interest in this context. Their Ising\(^5\)\(^,\)\(^6\) and Heisenberg\(^7\)\(^–\)\(^10\) models were calculated by the linear spin-wave (SW) theory and Monte Carlo (MC) methods, whereas their Hubbard models\(^11\)\(^,\)\(^12\) were investigated within a mean-field approximation. In the context of inelastic-neutron-scattering experiments, SW findings for the dynamic structure factor of the Ammann-Beenker Heisenberg antiferromagnet reveal intriguing excitation features possibly due to the quasiperiodicity,\(^13\) the coexistence of linear soft modes near the magnetic Bragg peaks at low frequencies, self-similar structures at intermediate frequencies, and flat bands at high frequencies. We may take further interest in inelastic light scatterings in quasiperiodic magnets, which strongly reflect their lattice symmetry and potentially bring brandnew information by virtue of the light polarization degrees of freedom.

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Thus motivated, we study Raman responses of 2D quasiperiodic spin-\(^\frac{1}{2}\) Heisenberg antiferromagnets on the $C_{5v}$, Penrose\(^21\)\(^,\)\(^22\) and $C_{mm}$, Ammann-Beenker\(^23\)\(^,\)\(^24\) lattices, described by the Hamiltonian $H = J \sum_{\langle i,j \rangle} S_i \cdot S_j$ ($J > 0$), where $\sum_{\langle i,j \rangle}$ runs over all pairs of connected vertices. With their bipartite structures in mind, we introduce Holstein-Primakoff bosons\(^25\) and expand the bosonic Hamiltonian in powers of the inverse spin magnitude $1/S$, denoting the terms of order $S^m$ by $H^{(m)}$. When we decompose the quartic boson operators $H^{(0)}$ into quadratic terms $H_{BL}^{(0)}$ and residual normal-ordered quartic interactions : $H^{(0)}$ : using Wick’s theorem,\(^26\)\(^,\)\(^27\) the up-to-$O(S^2)$ bosonic Hamiltonian reads

$$ H = H^{(2)} + H^{(1)} + H_{BL}^{(0)} + : H^{(0)} : \equiv H_{BL}^+ + : H^{(0)} :. $$ (1)

First we diagonalize the bilinear Hamiltonian $H_{BL}$ via the generalized Bogoliubov transformation,\(^8\)\(^,\)\(^28\)\(^,\)\(^29\)

$$ H_{BL} = \sum_{m=0}^2 \sum_{\sigma=\pm} \sum_{l=1}^m c_{l \sigma}^\dagger c_{l \sigma}^\dagger, $$ (2)

where $E^{(2)}(m)$ is the classical ground-state energy and $E^{(m)}(m = 1, 0)$ are the $O(S^m)$ quantum corrections to it, while $a_{m \sigma}^\dagger$ creates an antiferromagnetic ($\sigma = +$) or ferromagnetic ($\sigma = -$) SW\(^30\)\(^,\)\(^31\) of energy $E^{(m)}_m$ for the vacuum state $|0\rangle_{BL}$, enhancing or reducing the ground-state magnetization, respectively.\(^32\)\(^,\)\(^33\) Then we take account of the two-body interactions : $H^{(0)}$ : in two ways, i.e., Green’s function (GF) perturbative and configuration-interaction (CI) variational approaches. Shastry and Shraiman\(^34\)\(^,\)\(^35\) formulated magnetic Raman scatterings in an antiferromagnetic insulator by perturbatively treating the single-band Hubbard model in the limit of sufficiently strong correlation $U$ compared to hopping $t$.\(^36\) With the electron band being half filled, the second-order vertex\(^37\) $R$ results in the well-known Loudon-Fleury operator\(^37\) consisting of pair exchange interactions $S_i \cdot S_j$, whereas the fourth-order vertices\(^4\) $R$ contain three-spin scalar-chirality terms $\propto S_i \cdot (S_j \times S_k)$ and four-spin ring-exchange terms $\propto (S_i \cdot S_j)(S_k \cdot S_l)$.\(^38\)\(^–\)\(^40\) With varying incident photon frequency $\omega_m$,\(^2\) $R$ predominates in the far-resonant regime, $t \ll |U - \hbar \omega_m|$, while\(^4\) $R$’s are also of major importance in the near-resonant regime, $t \approx |U - \hbar \omega_m|$. The Raman operators are classified according to the number of their constituent spin operators,\(^2\) $R = \sum_{m=2}^{2n} \sum_{\tau=2}^{2n} \sum_{j=1}^{2n} R(n = 1, 2)$, and each component can be expanded in $1/S$ terms in the bosonic language,

$$ R = \sum_{m=0}^\infty \sum_{\langle i,j \rangle} \sum_{\tau=2}^{2n} \sum_{j=1}^{2n} |2n\rangle R^{(m)}(\tau) \left| \frac{m-n}{2} \right| \sum_{l=1}^{m-n} \sum_{\tau=2}^{2n} \sum_{j=1}^{2n} |2n\rangle R^{(m)}(\tau) \left| \frac{m-n}{2} \right|, $$ (3)

where $|2n\rangle R^{(m)}(\tau)$, consisting of $2 \langle 0 \leq l \leq m \rangle$-magnon (2M) vertices, is of order $S^{-m}$. Via the Bogoliubov transformation, (3) truncated at $m = 2$, i.e., the up-to-$O(S^0)$ vertices become

$$ R = \sum_{n=1}^{1} \sum_{\langle i,j \rangle} \sum_{\tau=2}^{2n} \sum_{j=1}^{2n} |2n\rangle R^{(m)}(\tau) \left| \frac{m-n}{2} \right|, $$ (4)

where $|2n\rangle R^{(m)}(\tau)$ are normal-ordered with respect to the quasi-particle magnon operators,\(^2\) $R$ merely contribute to elastic (Rayleigh) scatterings and are thus omitted hereafter.
Putting \( \mathcal{R}(t) \equiv e^{iH_0 t}e^{-iH_0 t}/\hbar \) for any operator \( \mathcal{R} \), we define the 2M-mediated Raman intensities at absolute zero as:  
\[
\int_{-\infty}^{\infty} dt \frac{d^2 \mathcal{R}_m(t)}{2\pi} \sum_{n,n'=1} \langle 0|\{\mathcal{R}_m(t),\mathcal{R}_m(0)\}|n\rangle \equiv \langle p|I_{2M}(\omega)\rangle. \tag{5}
\]

Once we find the exact ground state of the Heisenberg Hamiltonian, \( H(0) = E_0(0) \), the Loudon-Fleury \( (p = 2) \) and Shastryl-Shraiman \( (p = 4) \) intensities can be exactly calculated as:  
\[
\langle p|I(\omega)\rangle = \int_{-\infty}^{\infty} dt \frac{d^2 \mathcal{R}_m(t)}{2\pi} \sum_{n,n'=1} \langle 0|\{\mathcal{R}_m(t),\mathcal{R}_m(0)\}|n\rangle = -\frac{i}{\pi} L \lim_{\omega \to \omega_0} \Im \langle 0|\mathcal{R}_m(t)|n\rangle \equiv \langle p|I_{2M}(\omega)\rangle. \tag{6}
\]

Magnet-magnon interactions significantly modify the Raman spectra, because pair-exchange and multiple-spin cyclic-exchange Raman vertices, emergent with and beyond the Loudon-Fleury scheme, respectively, play qualitatively different roles in inelastic photon scatterings. First we demonstrate this by a renormalized perturbation theory. The magnon GFs (S46b) and (S46d), the 2M-mediated Raman scattering intensities are calculated as:  
\[
\langle p|I_{2M}(\omega)\rangle = -\text{Im} \int_{-\infty}^{\infty} dt \frac{d^2 \mathcal{R}_m(t)}{2\pi} \sum_{n,n'=1} \langle 0|\{\mathcal{R}_m(t),\mathcal{R}_m(0)\}|n\rangle \equiv \text{Im} \int_{-\infty}^{\infty} dt \frac{d^2 \mathcal{R}_m(t)}{2\pi} \mathcal{G}_m(t); \tag{7}
\]

where we numerically obtain the coefficients \( \langle p|\mathcal{R}_m(t)\rangle \) and \( \langle p|\mathcal{G}_m(t)\rangle \).

Since any perturbative renormalization is hard to tractable for more-than-3M GFs, we decompose 4M GFs into 2M GFs as (S61) on one hand and into 3M and 1M GFs as (S61) on the other hand. The renormalized 1M GFs reduce to the Hartree-Fock solutions (S48), whereas the 2M and 3M ones are calculated through ladder-approximation Bethe-Salpeter equations (S49) and (S55), respectively.

The Raman operator is written as a rank-2 tensor doped with the polarization vectors of the incident (\( \varepsilon_{m} \)) and scattered (\( \varepsilon_{sc} \)) photons,  
\[
\langle p|R|\rangle = \sum_{\mu,\nu=1} \langle p|\mathcal{R}_{\mu,\nu}|\varepsilon_{m}^{\mu}\varepsilon_{sc}^{\nu}\rangle, \tag{8}
\]

where we set \( \varepsilon_{m} \) and \( \varepsilon_{sc} \) parallel to the lattice plane (\( \varepsilon_{m}^{\perp} = \varepsilon_{sc}^{\perp} = 0 \)) to reduce \( \langle p|R|\rangle \) to a \( 2 \times 2 \) matrix. In terms of the point symmetry group \( \mathcal{P} \) of the lattice, this is rewritten as  
\[
\langle p|R|\rangle = \sum_{\mu,\nu=1} \langle p|E_{P,\mu,\nu}|\varepsilon_{P,\mu,\nu}\rangle, \tag{9}
\]

where \( \langle p|E_{P,\mu,\nu}|\varepsilon_{P,\mu,\nu}\rangle \) runs over the Raman-active irreducible representations \( \Sigma_{P} \) of \( \mathcal{P} \), each with dimensionality \( d_{P,\mu,\nu} \), are the \( \mu \)th polarization-vector basis function and Raman vertex for \( \Sigma_{P} \), respectively. Since the ground state \( |0\rangle \) is invariant under every symmetry operation of \( \mathcal{P} \), any expectation value between Raman vertices of different symmetry species for it goes to zero. We can therefore classify the Raman intensities as to symmetry species,  
\[
\int_{-\infty}^{\infty} dt \frac{d^2 \mathcal{R}_m(t)}{2\pi} \sum_{n,n'=1} \langle 0|\{\mathcal{R}_m(t),\mathcal{R}_m(0)\}|n\rangle \equiv \langle p|I_{2M}(\omega)\rangle. \tag{10}
\]

Considering that \( \langle p|E_{P,\mu,\nu}|\varepsilon_{P,\mu,\nu}\rangle \) runs over the Raman-active irreducible representations \( \Sigma_{P} \), we find  
\[
\langle |I(\omega)|^2 \rangle = \sum_{\mu=1}^{d_{P,\mu,\nu}} \sum_{\nu=1}^{d_{P,\mu,\nu}} \sum_{\mu'=1}^{d_{P,\mu',\nu'}} \sum_{\nu'=1}^{d_{P,\mu',\nu'}} \langle p|E_{P,\mu,\nu}|\varepsilon_{P,\mu,\nu}\rangle \langle p|E_{P,\mu',\nu'}|\varepsilon_{P,\mu',\nu'}\rangle \tag{11}
\]

When \( \mathcal{P} = \mathcal{C}_{5v} \), (9) contains two 1D and one 2D symmetry species, whose basis functions and vertices are given by  
\[
E_{C_{5v}:1} \equiv e_{\mu}^{\mu_{m},\mu_{sc}} + e_{\nu}^{\nu_{m},\nu_{sc}}, \quad E_{C_{5v}:2} \equiv e_{\mu}^{\mu_{m},\mu_{sc}} - e_{\nu}^{\nu_{m},\nu_{sc}} \tag{12}
\]

e_{\mu}^{\mu_{m},\mu_{sc}} = \langle p|E_{P,\mu,\nu}|\varepsilon_{P,\mu,\nu}\rangle \tag{12}
\]

For the linear polarizations \( \varepsilon_{m(\mu,m_{sc})} = (\cos \phi_{m(\mu,m_{sc})}, \sin \phi_{m(\mu,m_{sc})}, 0) \), (12) reads \( E_{C_{5v}:1} = \cos \phi \cdot e_{\mu} + \sin \phi \cdot e_{\nu} \). Since \( \langle p|E_{P,\mu,\nu}|\varepsilon_{P,\mu,\nu}\rangle \) commutes with the Heisenberg Hamiltonian, the \( \Lambda_{1} \) species is Raman inactive within the Loudon-Fleury scheme. The \( \Lambda_{2} \) species is also Loudon-Fleury-Raman inactive. Since the Raman operator is time-reversal-invariant, the \( \langle p|E_{P,\mu,\nu}|\varepsilon_{P,\mu,\nu}\rangle \) are also time-reversal-antisymmetric. The second-order pair-exchange Raman vertices are all time-reversal-invariant.

For the 2D Penrose lattice this consists of one and only Raman-active species \( E_{2} \) to yield depolarized spectra,  
\[
\langle |I(\omega)|^2 \rangle = \langle p|E_{E_{2}:1}^2 |\varepsilon_{E_{2}:1}^2 \rangle \tag{12}
\]
the Heisenberg Hamiltonian, whereas $^{[4]}R_{\mathbf{S}1}$ comprises chiral spin fluctuations $S_y \times S_x$ breaking the time-reversal symmetry, both of which drive the fourth-order Raman response to depend on the light polarization,

$$^{[4]}I(\omega) = |^{[4]}C_{\mathbf{E}1}^{\mathbf{A}2}(\omega) + |^{[4]}C_{\mathbf{A}1}^{\mathbf{A}2}(\omega) \cos \phi_c + |^{[4]}C_{\mathbf{A}2}^{\mathbf{A}2}(\omega) \sin \phi_c|$$

as is demonstrated in Figs. 1(b) and 1(b')

Figures 1(a) and 1(a') present almost the same observations, while Figs. 1(b) and 1(b') show artificial differences at $\hbar \omega \approx 5J$. Since we cannot directly evaluate more-than-3M GFs in practice, $^{[4]}I(\omega)$ and more generally Raman responses beyond the Loudon-Fleury scheme, containing significant multimagnon contributions, are much harder to reliably calculate in terms of GFs than $^{[3]}I(\omega)$ obtainable from the established Bethe-Salpeter equation. Even though we can calculate 3M GFs, for instance, there may be some different Bethe-Salpeter-like manners of renormalization to complicate matters further. In order to reliably evaluate the role of multimagnon scatterings in novel Raman responses beyond the Loudon-Fleury scheme, we propose an alternative approach utilizing CI variational wavefunctions. Once we proceed to the fourth-order perturbation scheme, the polarized Raman spectra (15) for linearly polarized components of the proceed to the fourth-order perturbation scheme, the polarized ized significant multimagnon contributions, are much harder to reli-

$^{[4]}I(\omega) \equiv \sum_{n=1}^{4} |^{[4]}C_{[\mathbf{A}1]^{\mathbf{A}2}}^{\mathbf{E}1}(\omega)|^2$, (15)

We verify the CI evaluation (17) in Fig. 2. The CI findings for the $E_2$ and $A_2$ symmetry species are in very good agreement with the exact solutions obtained by a recursion method based on the Lanczos algorithm. While the $E_2$ scattering is Loudon-Fleury-Raman active and arises chiefly from pair-exchange spin fluctuations, a nonnegligible portion of this scattering intensity is mediated by 4M fluctuations, as revealed by our CI scheme. While the $A_2$ scattering intensity is small compared to the other symmetries, it is so interesting as to allow for directly observing the spin chirality $\mathbf{A}_2$ Raman response is emergent in the honeycomb and kagome lattices consisting of nonparallellograms but impossible in the square and triangular lattices comprising rhombuses. The present scattering of $A_2$ symmetry owes to the quasiperiodic geometry whose rank $D$, i.e., the smallest number of wavevectors that can span the whole diffraction pattern of the crystal by their integral linear combinations, is larger than the actual physical dimension $d$. The $d = 2$ Penrose and Ammann-Beenker lattices have the same indexing dimension $D = 4$. Our CI scheme well reproduces the $A_1$ exact solution as well but somewhat overestimates its 4M spectral weight. This is because the $A_1$ symmetry species becomes Raman active due to ring exchange interactions such as $S_x S_y S_x S_y$, $S_x S_y S_x S_y$, and $S_x S_y S_x S_y$, the latter two of which are essentially described by six or more Holstein-Primakoff bosons and therefore sensitive to the Hamiltonian of $O(S^{-1})$.

What will happen to clusters without $C_{5v}$ symmetry? The $A_1$ and $A_2$ symmetry species remain Loudon-Fleury-
Figure 3 shows comparative CI and GF calculations of the 2D Ammann-Beenker lattice of C₈, point symmetry, the above six of which are compared with the exact solutions. All other details are the same as Fig. 2.

Figure 3 reveals our CI scheme to be much superior to the conventional GF approach. Indirect evaluation of the 4M GFs cannot reproduce the significant 4M-mediated scattering intensity in general. The spin-chirality-driven A₂ scattering intensity is especially misunderstood by the GF approach. Indeed low-energy peaks are almost of 2M character, but high-energy ones clearly owe to both 2M and 4M scatterings, as is revealed by the CI calculations. The inaccurate 4M GFs are totally ignorant of the mixed character of these A₂ scatterings and such is the case with every symmetry species. The essential features of all the calculational schemes developed are further demonstrated in Supplemental Material.⁵⁵

We have demonstrated the advantages of CI over GF in analyzing Raman scattering intensities according to mediating magnons. Once we go beyond the Loudon-Fleury second-order perturbation scheme, or sometimes even within it, we will find out any spectral weight of multimagnon character correctly only by evaluating the Raman correlation functions G₂M(t) directly. Real-frequency dynamic quantities cannot be obtained directly from path-integral calculations such as quantum MC (QMC) findings. Relevant imaginary-time correlation functions have to be first calculated and then continued to real frequency. Laplace transforms are difficult to invert numerically and maximum-entropy analytic continuation, for instance, of QMC data is not necessarily successful even for small periodic clusters.⁵⁵ Under such circumstances,
our elaborate CI approach can open up a new path of calculating dynamic properties. In terms of its ability to clarify what kind of intermediate states are essential in which scattering channel, we note that a newly developed representation theory of the quantum affine Lie algebra for the spin-1/2 XXZ infinite chain\(^{70,71}\) opened the door to a full understanding of its dynamics. We are aware of how much percentage of its total structure factor intensity two- and four-spinon intermediate states contribute.\(^{72,73}\) Dynamic spin structure factors of quasiperiodic magnets\(^{3}\) are also analyzable in full detail with the present CI scheme.

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Supplemental Material for
Polarized Raman Response of Two-Dimensional Quasiperiodic Antiferromagnets: Configuration-Interaction versus Green’s Function Approaches

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S1. Spin-Wave Hamiltonian

We discuss antiferromagnetic Heisenberg models on the two-dimensional (2D) Penrose and Ammann-Beenaker lattices of point symmetry C_{5v} and C_{10h}, respectively, both of which are described by the Hamiltonian

\[ \mathcal{H} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \quad (J > 0), \]  

(S1)

where \( \mathbf{S}_i \) is the spin-1/2 operator at site \( i \) and \( \sum_{\langle i,j \rangle} \) runs over all pairs of connected vertices. Since the Penrose and Ammann-Beenaker lattices are both bipartite, we divide them each into two sublattices, A with \( L_A \) sites and B with \( L_B \equiv L_\text{L} - L_A \) sites, respectively. We introduce the Holstein-Primakoff (HP) bosonic spin deviation operators\(^1\)

\[ S_i^+ = (2S - a_i^+ a_i) \frac{1}{2} a_i \quad \text{and} \quad S_i^- = (2S - a_i^+ a_i) \frac{1}{2}, \]

(S2)

where the site indices are understood as \( i \in \text{A} \) and \( j \in \text{B} \). We expand the Hamiltonian (S1) in powers of the inverse spin magnitude \( 1/S \),

\[ \mathcal{H} = \mathcal{H}^{(2)} + \mathcal{H}^{(1)} + \mathcal{H}^{(0)} + O(S^{-1}), \]  

(S3)

where \( \mathcal{H}^{(m)} \), on the order of \( S^m \), reads

\[ \mathcal{H}^{(2)} = -JS^2 \sum_{i \in \text{A}} \sum_{j \in \text{B}} l_{i,j} \quad \mathcal{H}^{(1)} = JS \sum_{i \in \text{A}} \sum_{j \in \text{B}} l_{i,j} \]

\[ \times \left\{ (a_i^+ a_i + b_i^+ b_i) \right\} \quad \mathcal{H}^{(0)} = -J \sum_{i \in \text{A}} \sum_{j \in \text{B}} l_{i,j} \]

\[ \times \left\{ a_i^+ b_i^+ b_j + \frac{1}{4} \left( a_i^+ a_i a_j b_j + a_i^+ b_i^+ b_j^+ b_j + \text{H.c.} \right) \right\}, \]  

(S4)

with \( l_{i,j} \) being 1 for connected vertices \( i \) and \( j \), otherwise 0. We decompose the \( O(S^0) \) quartic Hamiltonian \( \mathcal{H}^{(0)} \) into quadratic terms \( \mathcal{H}^{(0)}_{\text{BL}} \) and normal-ordered quartic terms \( \mathcal{H}^{(0)}_{\text{H.c.}} \); through Wick’s theorem,\(^2\)

\[ a_i^+ a_j b_j = : a_i^+ a_j b_j : \]

\[ + 2 \left( \mathcal{B}_{\text{BL}}(a_i^+ a_j b_j) + \mathcal{B}_{\text{BL}}(a_j^+ b_j a_i) \right); \]

\[ - 2 \left( \mathcal{B}_{\text{BL}}(a_i^+ b_j a_i) + \mathcal{B}_{\text{BL}}(a_j^+ a_j b_j) \right); \]

\[ a_j^+ b_j^+ b_j = : a_j^+ b_j^+ b_j :, \]

\[ + 2 \left( \mathcal{B}_{\text{BL}}(a_j^+ b_j a_i) + \mathcal{B}_{\text{BL}}(a_j^+ b_j a_j) \right); \]

\[ - 2 \left( \mathcal{B}_{\text{BL}}(a_j^+ a_i) + \mathcal{B}_{\text{BL}}(a_j^+ b_j a_j) \right); \]

\[ \mathcal{B}_{\text{BL}} = \mathcal{B}_{\text{BL}}(0)^{\text{BL}} = \mathcal{B}_{\text{BL}}(0)^{\text{BL}} + \mathcal{B}_{\text{BL}}(0)^{\text{BL}} \quad \mathcal{B}_{\text{BL}}(0)^{\text{BL}} = \mathcal{B}_{\text{BL}}(0)^{\text{BL}} + \mathcal{B}_{\text{BL}}(0)^{\text{BL}} \quad \mathcal{B}_{\text{BL}}(0)^{\text{BL}} = \mathcal{B}_{\text{BL}}(0)^{\text{BL}} + \mathcal{B}_{\text{BL}}(0)^{\text{BL}}, \]  

(S5)

where \( |0\rangle_{\text{BL}} \) denotes the quasiparticle magnon vacuum. The up-to-\( O(S^0) \) bosonic Hamiltonian reads

\[ \mathcal{H}^{(0)} = \mathcal{H}^{(2)} + \mathcal{H}^{(1)} + \mathcal{H}^{(0)}_{\text{BL}} : \mathcal{H}^{(0)}_{\text{H.c.}} :. \]  

(1)

Let us express the bilinear Hamiltonian \( \mathcal{H}_{\text{BL}} \) as

\[ \mathcal{H}_{\text{BL}} = e^1 \mathcal{M} e + \sum_{m=0}^2 \mathcal{E}^{(m)}, \quad \mathcal{M} = \left[ \begin{array}{cc} A & C^T \\ C & B \end{array} \right], \]  

(S6)

where we define the row vectors \( a^T \) and \( b^T \) of dimension \( L_A \) and \( L_B \), respectively,

\[ e^1 = a^T a + b^T b \quad \text{and} \quad C = a^T b + b^T a \equiv \left[ a^T, b^T \right], \]  

(S7)

the matrices \( A, B, \) and \( C \) of dimension \( L_A \times L_A, L_B \times L_B, \) and \( L_A \times L_B, \) respectively,

\[ [A]_{i,j} = \delta_{i,j} \sum_{j \in \text{B}} l_{i,j} \quad [B]_{i,j} = \delta_{i,j} \sum_{j \in \text{B}} l_{i,j} \quad [C]_{i,j} = \delta_{i,j}, \]

(S8)

and the constants

\[ E^{(2)} = \mathcal{E}^{(2)} \equiv \mathcal{E}^{(2)}(1), \quad E^{(1)} = -JS \sum_{i \in \text{A}} \sum_{j \in \text{B}} l_{i,j}, \]  

(1)
\[ \hat{E}^{(0)} = J \sum_{i \in X} \sum_{\mu \in B} l_{i,\mu} \left( BL(0)|a_i^\dagger a_\mu|0\rangle_{BL} \right. \\
+ \frac{1}{2} \left( BL(0)|a_i a_\mu|0\rangle_{BL} + BL(0)|a_i^\dagger b_\mu^\dagger|0\rangle_{BL} \right) \\
+ BL(0)|a_i^\dagger a_\mu|0\rangle_{BL} BL(0)|b_\mu^\dagger b_\mu^\dagger|0\rangle_{BL} \\
+ BL(0)|a_i a_\mu|0\rangle_{BL} BL(0)|a_\mu b_\mu|0\rangle_{BL} \left. \right) \times \left( BL(0)|a_i^\dagger b_\mu^\dagger|0\rangle_{BL} + BL(0)|a_\mu b_\mu|0\rangle_{BL} \right) \right]. \]

We carry out the Bogoliubov transformation
\[ c = X \alpha; \quad X \equiv \begin{bmatrix} S & U \\ V & T \end{bmatrix}, \]
where we define the matrices \( S, T, U \), and \( V \) of dimension \( L_x \times L_x, L_y \times L_y, L_x \times L_y, \) and \( L_y \times L_y \), respectively, to obtain the ferromagnetic and antiferromagnetic magnon operators \[ \left[ a_{i \uparrow}^+, \ldots, a_{L_x}^+ a_{i \downarrow}^+, \ldots, a_{L_x}^+ \right] \equiv a^+ \].

By virtue of the bosonic commutation relations, the Bogoliubov transformation matrix \( X \) satisfies\(^{4-6}\)
\[ X \Gamma X^\dagger = \Gamma; \quad \Gamma \equiv \begin{bmatrix} -I(L_x) & 0 \\ 0 & I(L_y) \end{bmatrix}, \]
\[ X \Gamma X^\dagger = \Gamma; \quad \Gamma^\prime \equiv \begin{bmatrix} -I(L_y) & 0 \\ 0 & I(L_x) \end{bmatrix}, \]

where \( I(L) \) denotes the \( L \times L \) identity matrix. Demanding that \( X \) should diagonalize \( M \), we obtain
\[ X \Gamma X^\dagger = \Gamma \Gamma^\prime \equiv \begin{bmatrix} \varepsilon_i^\uparrow, \ldots, \varepsilon_{L_x}^\uparrow, \varepsilon_i^\downarrow, \ldots, \varepsilon_{L_x}^\downarrow \end{bmatrix} \equiv E, \]

where the eigenvalues \( \varepsilon_i^\pm \) are non-negative. Multiplying (13) by \( X \Gamma^\prime \) from the left yields
\[ \Gamma M X = X \Gamma^\prime E. \]

The column vectors of \( X \) and the diagonal elements of \( \Gamma^\prime E \) are the right eigenvectors and their eigenvalues for \( \Gamma M \), respectively.

The eigenvalues of \( \Gamma M \) comprise \( L_- \) negative and \( L_+ \) positive eigenvalues,\(^{5,6}\)
\[ \Gamma E = E^\prime \equiv \begin{bmatrix} \varepsilon_1^\pm, \ldots, \varepsilon_{L_x}^\pm, \varepsilon_1^\pm, \ldots, \varepsilon_{L_x}^\pm \end{bmatrix}. \]

Having in mind that \( (\Gamma^\prime)^2 = I(L) \), we find \( E = \Gamma E^\prime \). Then the non-negative eigenvalues \( \varepsilon_i^\pm \) read \( \varepsilon_i^\pm = \pm \varepsilon_i^\pm \), \( \varepsilon_i^\pm = \varepsilon_i^\pm \) to yield the diagonal one-body Hamiltonian
\[ \mathcal{H}_{BL} = \sum_{m=0}^2 \varepsilon_i^m + \frac{1}{2} \sum_{i=1}^{L_x} \varepsilon_i^\uparrow a_i^\dagger a_i^\dagger + \sum_{i=1}^{L_y} \varepsilon_i^\dagger a_i a_i^\dagger. \]

Denoting the \( O(S^m) \) component of \( \varepsilon_i^\uparrow \) by \( \varepsilon_i^{(m)} \), we express the \( O(S^0) \) quantum corrections to the classical ground-state energy \( E^{(0)} \) as
\[ E^{(m)} = \hat{E}^{(m)} + \sum_{i=1}^{L_x} \varepsilon_i^{(m)} (m = 1, 0). \]
The quartic interaction : $\mathcal{H}^{(0)}$ : is given by

$$
\mathcal{H}^{(0)} := -J \sum_{i \in A} \sum_{j \in B} \sum_{\ell, \ell', \ell''} V^{(i)}_{\ell, \ell', \ell''} \alpha^+_{\ell} \alpha^+_{\ell'} \alpha^-_{\ell''} \alpha^-_{\ell'''}
$$

with $V^{(2)}_{\ell, \ell', \ell''} = V^{(3)*}_{\ell, \ell', \ell''}$, $V^{(5)}_{\ell, \ell', \ell''} = V^{(6)*}_{\ell, \ell', \ell''}$, and $V^{(7)}_{\ell, \ell', \ell''} = V^{(8)*}_{\ell, \ell', \ell''}$. Figure S2 shows the magnon-magnon interactions $V^{(i)}_{\ell, \ell', \ell''}$, diagrammatically. We give the magnon-number-conserving interactions explicitly in particular,

$$
V^{(1)}_{\ell, \ell', \ell''} = \frac{1}{4} \left( s_{\ell} s_{\ell'} v_{\ell''} v_{\ell''} + s_{\ell} s_{\ell''} v_{\ell'} v_{\ell'} + s_{\ell'} s_{\ell''} v_{\ell'} v_{\ell'} + s_{\ell'} s_{\ell''} v_{\ell'} v_{\ell'} \right)
$$

and

$$
V^{(4)}_{\ell, \ell', \ell''} = u_{\ell} u_{\ell'} u_{\ell'} v_{\ell''} v_{\ell''} + u_{\ell} u_{\ell'} u_{\ell'} v_{\ell''} v_{\ell''} + u_{\ell} u_{\ell'} u_{\ell'} v_{\ell''} v_{\ell''}
$$

in terms of the matrix elements $s_{l_{ij}} \equiv [S]_{l_{ij}}, u_{l_{ij}} \equiv [U]_{l_{ij}}$, and $v_{l_{ij}} \equiv [V]_{l_{ij}}$ defined in (S10).

**S2. Magnetic Raman Scattering**

Following the Shastry-Shraiman perturbation theory, we derive spin-$\frac{1}{2}$ magnetic Raman operators from the half-filled single-band nearest-neighbor Hubbard model

$$
\mathcal{H} = U \sum_i c^+_i c^+_i c_i c_i - t \sum_{\langle i,j \rangle} \sum_{\sigma=\uparrow,\downarrow} (c^+_i c_{j\sigma} + c^+_j c_{i\sigma}) + \text{H.c.},
$$

$$
\equiv \mathcal{H}_U + \mathcal{H}_F
$$

where $c^+_i$ creates an electron with spin $\sigma$ at site $i$. We assume that $0 < t \ll U$. We replace all the hopping terms $c^+_i c_{j\sigma}$ in $\mathcal{H}_F$ with $c^+_i c_{j\sigma} \exp \left[ \mathbf{F} \cdot \mathbf{r} \right] \mathcal{A}(\mathbf{r}) \cdot d\mathbf{r}$, where $c$ and $e$ are the elementary electric charge and the light velocity, respectively, and then the applied electric field $\mathbf{E}(t)$ reads as $-\partial \mathbf{A}(t)/\partial t$. Suppose $\gamma_{\mathbf{q}, \omega}$ creates a photon of momentum $\mathbf{q}$, energy $\hbar \omega$, and polarization $p$, then the second-quantized vector potential is written as

$$
A(\mathbf{r}) = \sum_{\mathbf{q}, \omega} \frac{\hbar^2}{V \omega q} \left( e_{\mathbf{q}, \mathbf{p}} \gamma_{\mathbf{q}, \omega} e^{\mathbf{q} \cdot \mathbf{r}} + e_{\mathbf{q}, \mathbf{p}}^* \gamma_{\mathbf{q}, \omega} e^{-\mathbf{q} \cdot \mathbf{r}} \right)
$$

with $V$ being the appropriate volume of the sample. For visible light, we may put $e^{i\mathbf{q} \cdot \mathbf{r}} \approx 1$ and therefore denote $A(\mathbf{r})$ sim-
ply by a hereafter. Then the electron-photon-coupled Hamiltonian reads

$$\mathcal{H}_{\text{el-ph}} = \mathcal{H} + \Omega + \sum_{m=1}^{\infty} \frac{\langle m | \mathcal{J} | \Omega \rangle}{\hbar c} \sum_{q, \phi} \hbar \omega_q \gamma_q^\phi \gamma_q^\phi,$$

for all \(m \neq 0\), \(\Omega \equiv \sum_{q, \phi} \hbar \omega_q \gamma_q^\phi \gamma_q^\phi\).

$$[m | \mathcal{J} | \Omega] \equiv -i \sum_{i} \sum_{j} \frac{1}{m!} \left[ \gamma_i \gamma_j \gamma_i \gamma_j \right] \left[ \left( \frac{-ie}{\hbar c} A \cdot d_{ij} \right)^m + \text{H.c.} \right],$$

$$= -i \sum_{i,j} \sum_{\sigma, \tau} \frac{1}{m!} \left[ \gamma_i \gamma_j \gamma_i \gamma_j \right] \left[ \left( \frac{-ie}{\hbar c} A \cdot d_{ij} \right)^m \right]$$

(S23)

with \(d_{ij} \equiv r_j - r_i\).

Let the photoinduced current operators \( [m | \mathcal{J} | \Omega] \) be perturbations to \( \mathcal{H} + \Omega \). The transition between arbitrary states, \( | i \rangle \) and \( | f \rangle \) of energy \( e_i \) and \( e_f \), each being a product of electronic and phononic states, are rated as

$$W_{if} = \frac{2 \pi}{\hbar} \langle f | \mathcal{J} | i \rangle^2 \delta(e_f - e_i).$$

Any Raman scattering contains two photons, starting with an incident photon and ending in a scattered photon, where (S22) is explicitly written as

$$A \equiv \sqrt{\frac{\hbar c^2}{V \omega_n} e_m \gamma_{q, \sigma_n}} + \sqrt{\frac{\hbar c^2}{V \omega_n} e_m \gamma_{q, \sigma_n}},$$

with \(\omega_n(q, \sigma_n), q_m(q, \sigma_n)\), and \(e_m(q, \sigma_n)\) being the frequency, momentum, and polarization of the incident (scattered) photon, respectively. The Raman transition matrix \( \mathcal{J} \) in proportion to \( A^2 \) reads

$$\mathcal{J} = [2 | \mathcal{J} | [1 | \mathcal{J} | \Omega - \mathcal{H}_U - \mathcal{H}_T [1 | \mathcal{J} | \Omega],$$

(S26)

Every magnetic Raman scattering demands that the electronic state should belong in the singly-occupied ground-state manifold at the beginning and end, where \([m | \mathcal{J} | \Omega] \), inducing a single electron transfer, singly has no contribution to the transition rate, \( \langle f | [m | \mathcal{J} | i \rangle = 0 \). Relevant intermediate states obtained by operating \([1 | \mathcal{J} | \Omega \) on the initial state each have one doublelon-holon pair together with no photon or two photons. The photonic state is also singly occupied at the beginning and end. Considering that \( t \ll U \), we regard both \([m | \mathcal{J} | \Omega \) and \( \mathcal{H}_U \), as perturbations to \( \mathcal{H}_U \) and therefore express the effective Raman operator as

$$\mathcal{R} = [1 | \mathcal{J} | \Omega - \mathcal{H}_U - \mathcal{H}_T [1 | \mathcal{J} | \Omega - \mathcal{H}_U - \mathcal{H}_T [1 | \mathcal{J} | \Omega,$$

(S27)

where \( \mathcal{P} \) is the projection operator to the singly-occupied ground-state manifold.

No-photon and two-photon intermediate states are higher in energy than the ground state by \(U - \hbar \omega_n\) and \(U + \hbar \omega_n\), respectively. Assuming that the incident photon energy is comparable to the electronic correlation energy, \( t \ll |U - \hbar \omega_n| \ll U \), we may replace \( (\mathcal{E}_i - \mathcal{H}_U)^{-1} \) by \( (\hbar \omega_n - U)^{-1} \) with the single occupancy at every site in mind, we express the electron operators in terms of the spin operators, 

$$\mathcal{P}^+_{s_i, s_j} \mathcal{P}^+ = \frac{1}{2} \delta_{s_i, s_j} + \sum_{\mu, \nu} S^\mu_i \left[ s^\mu_j \right] \left[ s^\nu_j \right] \left[ s^\gamma_i \right],$$

where \(s^\alpha\)’s are the Pauli matrices.

Fig. S3. Fourth-order electron hopping paths. (a)–(d) and (a’–d’) are cyclic paths, while (a’)–(d’) are round paths. Solid arrows create or annihilate a doublelon-holon pair arising from \(1 | \mathcal{J} | \Omega\), whereas broken arrows correspond to electron transfer arising from \( \mathcal{H}_U \).

The lowest-order \( n = 0 \) term in (S27), which is of second order in \( t \), reads

$$\mathcal{R} = - \frac{1}{U - \hbar \omega_n} [1 | \mathcal{J} | \Omega - \mathcal{H}_U - \mathcal{H}_T [1 | \mathcal{J} | \Omega - \mathcal{H}_U - \mathcal{H}_T [1 | \mathcal{J} | \Omega,$$

where \( \mathcal{P} \) is the projection operator to the singly-occupied ground-state manifold.

Applying (S28) to (S29) and discarding Rayleigh (elastic scattering) terms, we obtain what they call the Loudon-Fleury Raman vertex \(^{(10)} \)

$$\mathcal{R} = \frac{2 \pi e^2}{\hbar \omega_n} \frac{2 \pi e^2}{\sqrt{V \omega_n} \omega_n} U - \hbar \omega_n \sum_{(i,j)} \left( e_m \cdot d_{i,j} \right) s_i \cdot s_j.$$
With this in mind, the $n = 1$ Raman vertex reads

$$d_{i\sigma} = (-1)^{\delta_{i\sigma}} c_{i\sigma}, \quad c_{i\sigma}^{\dagger} = (-1)^{\delta_{i\sigma}} d_{i\sigma}. \tag{S31}$$

Substituting (S31) into (S28) with the single-occupancy constraint $\sum_{\sigma} d_{i\sigma} d_{i\sigma} = 1$ in mind yields

$$\mathcal{P}^{\dagger}_{i\sigma} c_{i\sigma} \mathcal{P} = \mathcal{P}(-1)^{\delta_{i\sigma} + \delta_{i\sigma'}} \left( \delta_{i\sigma, i\sigma} - d_{i\sigma} d_{i\sigma'} \right) \mathcal{P} = \mathcal{P} d_{i\sigma}^{\dagger} d_{i\sigma'} \mathcal{P}, \tag{S33}$$

showing that the electron and hole at each site have the same spin projection, and therefore, (S32) vanishes. Likewise, all the odd-integral-$n$ vertices contribute nothing to the Raman intensity of our system (S23).

The next leading term is $n = 2$ in (S27), which is of fourth order in $r$, reads

$$\mathcal{R} = \frac{-1}{(U - i\hbar\omega_m)^4} \mathcal{P}^{\dagger} \mathcal{H}_r \mathcal{H}_r^{\dagger} \mathcal{P} = \frac{2\pi e^2}{\hbar V \sqrt{\omega_m \omega_c}} \sum_{(i_1, i_2, j_1, j_2)} \left\{ -4Q \left[ (S_{i_1} \cdot S_{i_2}) (S_{i_1} \cdot S_{j_1}) + (S_{i_1} \cdot S_{j_1}) (S_{i_2} \cdot S_{j_2}) - (S_{i_1} \cdot S_{i_2}) (S_{j_1} \cdot S_{j_2}) \right] + 2i \sum_{n=1}^{4} \mathcal{D}_n S_{i_1} \cdot S_{i_2} \cdot S_{j_1} \cdot S_{j_2} + \frac{2}{n} \sum_{n=1}^{4} \mathcal{D}_n S_{i_1} \cdot S_{i_2} \cdot S_{j_1} \cdot S_{j_2} \right\}; \tag{S35}$$

Figure S3 shows in what order many electrons move in a variety of fourth-order hopping paths. After some algebra, we find the fourth-order Raman vertex as

$$\mathcal{R} = \frac{2\pi e^2}{\hbar V \sqrt{\omega_m \omega_c}} \sum_{(i_1, i_2, j_1, j_2)} \left\{ \begin{array}{l} -4Q \left[ (S_{i_1} \cdot S_{i_2}) (S_{i_1} \cdot S_{j_1}) + (S_{i_1} \cdot S_{j_1}) (S_{i_2} \cdot S_{j_2}) - (S_{i_1} \cdot S_{i_2}) (S_{j_1} \cdot S_{j_2}) \right] + 2i \sum_{n=1}^{4} \mathcal{D}_n S_{i_1} \cdot S_{i_2} \cdot S_{j_1} \cdot S_{j_2} + \frac{2}{n} \sum_{n=1}^{4} \mathcal{D}_n S_{i_1} \cdot S_{i_2} \cdot S_{j_1} \cdot S_{j_2} \end{array} \right\}; \tag{S35}$$

where $\sum_{(i_1, i_2, j_1, j_2)}$ run over four-site-cyclic and three-site-round paths, respectively, and we abbreviate $d_{n,n'} \equiv r_{n'} - r_n$ as $d_{n,n'}$ with $1 \leq n, n' \leq 4$. The Shastry-Shraiman fourth-order mechanism is of major importance in the Raman response when the incident photon energy $i\hbar\omega_m$ is in the near-resonant regime, $t \approx |U - i\hbar\omega_m|$.
Intending to further express the Raman operators (S30) and (S35) in terms of the bosonic language, we classify them according to the number of their constituent spin operators, which we shall denote by \( \tau \),

\[
\mathcal{R} = \sum_{n=1}^{\infty} \sum_{r=2}^{2n} [2n]_R \cdot \sum_{m=0}^{2n} \sum_{l=0}^{l} [2n]_R^{(r-m)} \cdot \mathcal{R}_{2M}^{(r-m)}.
\]

where \([2n]_R^{(r-m)}\) and \([2n]_R^{2M}\) consist of the Heisenberg pair exchange, three-spin scalar-chirality, and four-spin ring-exchange terms, respectively. They also read descending powers of the spin magnitude and each further break into components of different numbers of mediating magnons,

\[
\sum_{n=0}^{\infty} \sum_{m=0}^{n} \sum_{l=0}^{m} [2n]_R^{(r-m)} \cdot \mathcal{R}_{2M}^{(r-m)} = \sum_{n=0}^{\infty} \sum_{m=0}^{n} [2n]_R^{(r-m)} \cdot \mathcal{R}_{2M}^{(r-m)}.
\]

is a linear combination of terms containing 2l magnon operators. We truncate the inverse-spin-magnitude series (3) at \( m = 2 \) to have the up-to-\( O(S^0) \) Raman vertices

\[
\mathcal{R} \approx \sum_{n=1}^{\infty} \sum_{r=0}^{2n} [2n]_R^{(r-m)} \cdot \mathcal{R}_{2M}^{(r-m)} = \sum_{n=1}^{\infty} \sum_{m=0}^{n} [2n]_R^{(r-m)} \cdot \mathcal{R}_{2M}^{(r-m)},
\]

where setting \( p \) equal to 2 and 4 corresponds to the Loudon-Fleury second-order and Shastri-Shraiman fourth-order perturbation schemes, respectively. We employ Wick’s theorem so as for all the vertices \([2n]_R^{(r-m)} \cdot \mathcal{R}_{2M}^{(r-m)}\) to be normal-ordered with respect to the quasiparticle magnon operators. \([2n]_R^{2M}\) merely contribute to elastic (Rayleigh) scatterings and are henceforth omitted.

With a tacit understanding of site indices being used as \( i, k \in A \) and \( j, l \in B \), various spin interactions are written in terms of HP bosons as

\[
S_i \cdot S_j = -S^2 + S \left( a^\dagger_i a^i k^j a^j b^j b^j - \frac{1}{4} \left( a^\dagger_i a^i a^i a^i b^j b^j b^j b^j + H.c. \right) \right) + O(S^{-1}).
\]

\[
S_i \cdot S_k = S^2 + S \left( a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i b^k b^k - \frac{1}{4} \left( a^\dagger_i a^\dagger_i a^\dagger_i a^\dagger_i a^i a^i a^i a^i a^i a^i a^i a^i b^k b^k b^k b^k + H.c. \right) \right) + O(S^{-1}),
\]

\[
S_j \cdot S_l = S^2 + S \left( b^j b^j b^j b^j b^j b^j b^j b^j b^j b^j + H.c. \right) + O(S^{-1}),
\]

\[
i S_i \cdot (S_j x S_k) = S^2 \left( a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i b^j b^j b^j b^j b^j b^j b^j b^j b^j b^j + H.c. \right) + O(S^{-1}),
\]

\[
i S_j \cdot (S_k x S_l) = S^2 \left( a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i b^j b^j b^j b^j b^j b^j b^j b^j b^j b^j + H.c. \right) + O(S^{-1}),
\]

\[
S_i \cdot S_j (S_k x S_l) = S^4 - S^3 \left( a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i b^j b^j b^j b^j b^j b^j b^j b^j b^j b^j + H.c. \right)
\]

\[
+ S^2 \left( a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i b^j b^j b^j b^j b^j b^j b^j b^j b^j b^j + H.c. \right) + O(S^{-1}),
\]

\[
S_i \cdot S_k (S_j x S_l) = S^4 - S^3 \left( a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i b^j b^j b^j b^j b^j b^j b^j b^j b^j b^j + H.c. \right)
\]

\[
+ S^2 \left( a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i a^i b^j b^j b^j b^j b^j b^j b^j b^j b^j b^j + H.c. \right) + O(S^{-1}).
\]
Via the Bogoliubov transformation (S10), the two-magnon (2M)- and four-magnon (4M)-mediated Raman scattering operators read

\[ \sum_{n=1}^{p/2} \sum_{l,l' } R_{2M} = \sum_{l,l' } |p W_{1,l,l'}^{(1)} e^{i \alpha_{l}^{+} a_{l}^{+} a_{l}^{+} a_{l}^{+} } + \sum_{l,l' } |p W_{1,l,l'}^{(2)} e^{i \alpha_{l}^{+} a_{l}^{+} a_{l}^{+} a_{l}^{+} } + \sum_{l,l' } |p W_{1,l,l'}^{(3)} e^{i \alpha_{l}^{+} a_{l}^{+} a_{l}^{+} a_{l}^{+} } + \sum_{l,l' } |p W_{1,l,l'}^{(4)} e^{i \alpha_{l}^{+} a_{l}^{+} a_{l}^{+} a_{l}^{+} } | (S44) \]

with \( p/2 \) and \( p W_{1,l,l'} \) and

\[ \sum_{n=1}^{p/2} \sum_{l,l' } R_{4M} = \sum_{l,l' } |p X_{1,l,l’,l’}^{(1)} e^{i \alpha_{l}^{+} a_{l}^{+} a_{l}^{+} a_{l}^{+} } + \sum_{l,l' } |p X_{1,l,l’,l’}^{(2)} e^{i \alpha_{l}^{+} a_{l}^{+} a_{l}^{+} a_{l}^{+} } + \sum_{l,l' } |p X_{1,l,l’,l’}^{(3)} e^{i \alpha_{l}^{+} a_{l}^{+} a_{l}^{+} a_{l}^{+} } + \sum_{l,l' } |p X_{1,l,l’,l’}^{(4)} e^{i \alpha_{l}^{+} a_{l}^{+} a_{l}^{+} a_{l}^{+} } | (S45) \]

with \( p X_{1,l,l’,l’} \) and \( p X_{1,l,l’,l’}^{(4)} \) and

\[ \sum_{n=1}^{p/2} \sum_{l,l' } R_{4M} = \sum_{l,l' } |p X_{1,l,l’,l’}^{(2)} e^{i \alpha_{l}^{+} a_{l}^{+} a_{l}^{+} a_{l}^{+} } + \sum_{l,l' } |p X_{1,l,l’,l’}^{(3)} e^{i \alpha_{l}^{+} a_{l}^{+} a_{l}^{+} a_{l}^{+} } + \sum_{l,l' } |p X_{1,l,l’,l’}^{(4)} e^{i \alpha_{l}^{+} a_{l}^{+} a_{l}^{+} a_{l}^{+} } | (S44) \]

and \( \lambda_{j}^{i} \) is understood to be infinitesimal. Next we consider the ladder-approximation Bethe-Salpeter equations for the 2M GFs

\[ G_{l,l'}^{k,k'}(\omega) = G_{l,l'}^{k,k'}(\omega) = \frac{1}{\hbar} \sum_{p,p'} G_{l,l'}^{k,k'}(\omega) \times \sum_{i \in \Lambda} \sum_{j \in \Lambda} l_{i,j} V_{i,j}^{(4)} (i,j,p,p',q,q') G_{l,l'}^{k,k'}(\omega), \]  

\[ G_{l,l'}^{k,k'}(\omega) = G_{l,l'}^{k,k'}(\omega) = \frac{1}{\hbar} \sum_{p,p'} G_{l,l'}^{k,k'}(\omega) \times \sum_{i \in \Lambda} \sum_{j \in \Lambda} l_{i,j} V_{i,j}^{(9)} (i,j,p,p',q,q') G_{l,l'}^{k,k'}(\omega), \] 

\[ G_{l,l'}^{k,k'}(\omega) = G_{l,l'}^{k,k'}(\omega) = \frac{1}{\hbar} \sum_{p,p'} G_{l,l'}^{k,k'}(\omega) \times \sum_{i \in \Lambda} \sum_{j \in \Lambda} l_{i,j} V_{i,j}^{(10)} (i,j,p,p',q,q') G_{l,l'}^{k,k'}(\omega), \] 

denoting the unperturbed 2M GFs by

\[ G_{l,l'}^{k,k'}(\omega) = \frac{1}{\hbar} \sum_{p,p'} G_{l,l'}^{k,k'}(\omega) \times \sum_{i \in \Lambda} \sum_{j \in \Lambda} l_{i,j} V_{i,j}^{(4)} (i,j,p,p',q,q') G_{l,l'}^{k,k'}(\omega), \] 

\[ G_{l,l'}^{k,k'}(\omega) = \frac{1}{\hbar} \sum_{p,p'} G_{l,l'}^{k,k'}(\omega) \times \sum_{i \in \Lambda} \sum_{j \in \Lambda} l_{i,j} V_{i,j}^{(9)} (i,j,p,p',q,q') G_{l,l'}^{k,k'}(\omega), \] 

\[ G_{l,l'}^{k,k'}(\omega) = \frac{1}{\hbar} \sum_{p,p'} G_{l,l'}^{k,k'}(\omega) \times \sum_{i \in \Lambda} \sum_{j \in \Lambda} l_{i,j} V_{i,j}^{(10)} (i,j,p,p',q,q') G_{l,l'}^{k,k'}(\omega), \] 

We solve the eigenvalue equations obtained from (S49a)–(S49c),

\[ |\omega^{|V|} \rangle = \hbar (i \sigma_{\tau}^{\prime} \sigma_{\tau}^{\prime} l_{l} e^{i \alpha_{l}^{+} a_{l}^{+} a_{l}^{+} a_{l}^{+} } |0\rangle, \] 

where \( \omega^{|V|} \) are the column vectors of dimension \( L_{l}L_{r} \) whose \((l_{l}-1)_{l_{l}}^{L_{l}}+k_{l} \sigma_{l}^{\prime} \sigma_{l}^{\prime} \)-elements are given by

\[ \left[ g_{l_{l}^{k_{l}^{l}}}^{\sigma_{l}^{\prime} \sigma_{l}^{\prime}} \right]_{(l_{l}-1)_{l^{L_{l}}}+k_{l} \sigma_{l}^{\prime} \sigma_{l}^{\prime}}^{k_{l} \sigma_{l}^{\prime} \sigma_{l}^{\prime}} = 0 \left[ \sigma_{l}^{\prime} \sigma_{l}^{\prime} \right]_{(l_{l}-1)_{l^{L_{l}}}+k_{l} \sigma_{l}^{\prime} \sigma_{l}^{\prime}}^{k_{l} \sigma_{l}^{\prime} \sigma_{l}^{\prime}}, \] 

whereas \( \omega^{|V|} \) are the matrices of dimension \( L_{l}L_{r} \times L_{l}L_{r} \) whose \((l_{l}-1)_{l_{l}}^{L_{l}}+k_{l} \sigma_{l}^{\prime} \sigma_{l}^{\prime} \)-elements are given by

\[ \left[ V^{+} \right]^{(l_{l}-1)_{l^{L_{l}}}+k_{l} \sigma_{l}^{\prime} \sigma_{l}^{\prime} + l_{r} \sigma_{l}^{\prime} \sigma_{l}^{\prime}} = \delta_{k_{l}^{l},k_{l}^{l}} \left[ \epsilon_{l_{l}^{l}}^{\prime} + \epsilon_{l_{l}^{l}}^{\prime} \right] - \sum_{i \in \Lambda} \sum_{j \in \Lambda} l_{i,j} V_{i,j}^{(3)} (i,j,k_{l}^{l},k_{l}^{l}), \] 

\[ \left[ V^{+} \right]^{(l_{l}-1)_{l^{L_{l}}}+k_{l} \sigma_{l}^{\prime} \sigma_{l}^{\prime} + l_{r} \sigma_{l}^{\prime} \sigma_{l}^{\prime}} = \delta_{k_{l}^{l},k_{l}^{l}} \left[ \epsilon_{l_{l}^{l}}^{\prime} + \epsilon_{l_{l}^{l}}^{\prime} \right] - 2 \sum_{i \in \Lambda} \sum_{j \in \Lambda} l_{i,j} V_{i,j}^{(9)} (i,j,k_{l}^{l},k_{l}^{l}), \]
\[
\mathcal{V}^\downarrow \mid (k\downarrow,l\downarrow,k\downarrow,l\downarrow,\ldots) = \delta_{k\downarrow,l\downarrow} \delta_{k\downarrow,l\downarrow} \left( \bar{e}_{k\downarrow} + \bar{e}_{l\downarrow} \right) - 2J \sum_{i,A} \sum_{j,B} l_{ij} V^{(1)}_{ijklk,l,l,l} \quad \text{(S53c)}
\]

The Lehmann representation (11) of the 2M GFs reads
\[
G_{k\downarrow,l\downarrow}^{k\downarrow,l\downarrow} (\omega) = \sum_{j=0}^{L_{\downarrow}-1} \frac{\hbar g_{j\downarrow}^{k\downarrow,l\downarrow} \left( \bar{g}_{j\downarrow}^{k\downarrow,l\downarrow} \right)^*}{\hbar \omega - \hbar \Omega_{j\downarrow}^{k\downarrow,l\downarrow} + \eta} \quad \text{(S54)}
\]

Likewise, we consider the three-leg-ladder analogs of the Bethe-Salpeter equations for the 3M GFs
\[
G_{k\downarrow,l\downarrow}^{k\downarrow,l\downarrow} (\omega) = \frac{J}{\hbar} \sum_{p,-p} \sum_{q,-q} \sum_{q',-q'} G_{p\downarrow,p\downarrow}^{k\downarrow,l\downarrow} (\omega)_{BL} \times \sum_{i,A} \sum_{j,B} l_{ij} \left( \frac{1}{2} \delta_{p\downarrow,q'} V_{ij,p\downarrow,p\downarrow,q'}^{(9)} + \frac{1}{2} \delta_{q\downarrow,q'} V_{ij,p\downarrow,p\downarrow,q'}^{(4)} + \frac{1}{2} \delta_{p\downarrow,q'} V_{ij,p\downarrow,p\downarrow,q'}^{(4)} \right) G_{l\downarrow,l\downarrow}^{q\downarrow,q\downarrow} (\omega)_{BL}
\]
\[
G_{k\downarrow,l\downarrow}^{k\downarrow,l\downarrow} (\omega) = \frac{J}{\hbar} \sum_{p,-p} \sum_{q,-q} \sum_{q',-q'} G_{p\downarrow,p\downarrow}^{k\downarrow,l\downarrow} (\omega)_{BL} \times \sum_{i,A} \sum_{j,B} l_{ij} \left( \frac{1}{2} \delta_{p\downarrow,q'} V_{ij,p\downarrow,p\downarrow,q'}^{(9)} + \frac{1}{2} \delta_{q\downarrow,q'} V_{ij,p\downarrow,p\downarrow,q'}^{(4)} + \frac{1}{2} \delta_{p\downarrow,q'} V_{ij,p\downarrow,p\downarrow,q'}^{(4)} \right) G_{l\downarrow,l\downarrow}^{q\downarrow,q\downarrow} (\omega)_{BL}
\]

denoting the unperturbed 3M GFs by
\[
G_{k\downarrow,l\downarrow}^{k\downarrow,l\downarrow} (\omega)_{BL} = \int_{-\infty}^{\infty} dt e^{i\omega t} \left[ G_{k\downarrow,l\downarrow}^{k\downarrow,l\downarrow} (\omega)_{BL} G_{l\downarrow,l\downarrow}^{k\downarrow,l\downarrow} (\omega)_{BL} G_{k\downarrow,l\downarrow}^{k\downarrow,l\downarrow} (\omega)_{BL} \right]
\]
\[
\frac{\hbar \delta_{k\downarrow,l\downarrow} + \delta_{k\downarrow,l\downarrow}}{\hbar \omega - \delta_{k\downarrow,l\downarrow}^\sigma - \delta_{k\downarrow,l\downarrow}^\pi + \eta}
\]
\[
G^{\sigma\pi\sigma}_{k\downarrow,l\downarrow} = \hbar \Omega^{\sigma\pi\sigma}_{k\downarrow,l\downarrow} G^{\sigma\pi\sigma}_{k\downarrow,l\downarrow}
\]
\[
\text{S57}
\]

where \(g^{\sigma\pi\sigma}_{k\downarrow,l\downarrow}\) are the column vectors of dimension \(L_{\downarrow}^2 L_{\sigma}\) whose \([(k_{\sigma\downarrow} - 1)L_{\downarrow}^2 + (k_{\pi\downarrow} - 1)L_{\sigma} + k_{\varsigma\downarrow}]\)-elements are given by
\[
\left[ g^{\sigma\pi\sigma}_{k\downarrow,l\downarrow} \right]_{(k\downarrow_{\sigma\downarrow}-1)L_{\downarrow}^2 + (k\downarrow_{\pi\downarrow}-1)L_{\sigma} + k\downarrow_{\varsigma\downarrow}} \equiv g^{\sigma\pi\sigma}_{k\downarrow,l\downarrow} = \left( \delta_{k\downarrow\sigma\downarrow} \delta_{l\downarrow\pi\downarrow} \delta_{k\downarrow\varsigma\downarrow} \right)_{l\downarrow\sigma\downarrow}
\]
\[
\text{S58}
\]

while \(\mathcal{V}^{\sigma\pi\sigma}\) are the matrices of dimension \(L_{\downarrow}^2 L_{\sigma} \times L_{\downarrow}^2 L_{\sigma}\) whose \([(k_{\sigma\downarrow} - 1)L_{\downarrow}^2 + (k_{\pi\downarrow} - 1)L_{\sigma} + k_{\varsigma\downarrow}]\) elements are given by
\[
\left[ \mathcal{V}^{\sigma\pi\sigma} \right]_{(k\downarrow_{\sigma\downarrow}-1)L_{\downarrow}^2 + (k\downarrow_{\pi\downarrow}-1)L_{\sigma} + k\downarrow_{\varsigma\downarrow}} = \delta_{k\downarrow\sigma\downarrow} \delta_{l\downarrow\lll \pi\downarrow} \delta_{k\downarrow\varsigma\downarrow} \left( \bar{e}_{k\downarrow} + \bar{e}_{l\downarrow} + \bar{e}_{l\downarrow} \right) - J \sum_{i,A} \sum_{j,B} l_{ij} \left( 2\delta_{k\downarrow\llll \pi\downarrow} V_{ij,k\downarrow,k\downarrow,l,l,l}^{(9)} + \delta_{k\downarrow\llll \llll} V_{ij,k\downarrow,k\downarrow,l,l,l}^{(4)} + \delta_{k\downarrow\llll \llll} V_{ij,k\downarrow,k\downarrow,l,l,l}^{(4)} \right)
\]
\[
\text{S59a}
\]
\[
\left[ \mathcal{V}^{\sigma\pi\sigma} \right]_{(k\downarrow_{\sigma\downarrow}-1)L_{\downarrow}^2 + (k\downarrow_{\pi\downarrow}-1)L_{\sigma} + k\downarrow_{\varsigma\downarrow}} = \delta_{k\downarrow\sigma\downarrow} \delta_{l\downarrow\llll \pi\downarrow} \delta_{k\downarrow\varsigma\downarrow} \left( \bar{e}_{k\downarrow} + \bar{e}_{l\downarrow} + \bar{e}_{l\downarrow} \right) - J \sum_{i,A} \sum_{j,B} l_{ij} \left( 2\delta_{k\downarrow\llll \pi\downarrow} V_{ij,k\downarrow,k\downarrow,l,l,l}^{(9)} + \delta_{k\downarrow\llll \llll} V_{ij,k\downarrow,k\downarrow,l,l,l}^{(4)} + \delta_{k\downarrow\llll \llll} V_{ij,k\downarrow,k\downarrow,l,l,l}^{(4)} \right)
\]
\[
\text{S59b}
\]

The Lehmann representation of the 3M GFs reads
\[
G_{k\downarrow,l\downarrow}^{k\downarrow,l\downarrow} (\omega) = \sum_{j=0}^{L_{\downarrow}-1} \frac{\hbar g_{j\downarrow}^{k\downarrow,l\downarrow} \left( \bar{g}_{j\downarrow}^{k\downarrow,l\downarrow} \right)^*}{\hbar \omega - \hbar \Omega_{j\downarrow}^{k\downarrow,l\downarrow} + \eta}
\]
\[
\text{S60}
\]

Since any perturbative renormalization is hardly tractable for more-than-3M GFs, we decompose them into less-than-4M GFs. The 4M GFs can be approximated by the 2M GFs on one hand,
\[
2G_{k\downarrow,l\downarrow}^{k\downarrow,l\downarrow} (\tau) = 2(0)T \left[ \alpha_{k\downarrow}^{\dagger}(\tau) \alpha_{k\downarrow}(\tau) \alpha_{l\downarrow}^{\dagger}(\tau) \alpha_{l\downarrow}(\tau) \alpha_{k\downarrow}^{\dagger}(\tau) \alpha_{l\downarrow}^{\dagger}(\tau) \right]
\]
\[
\text{S61}
\]

and by the 3M and 1M GFs on the other hand,
\[
4G_{k\downarrow,l\downarrow}^{k\downarrow,l\downarrow} (\tau) = 4(0)T \left[ \alpha_{k\downarrow}^{\dagger}(\tau) \alpha_{k\downarrow}(\tau) \alpha_{l\downarrow}^{\dagger}(\tau) \alpha_{l\downarrow}(\tau) \alpha_{k\downarrow}^{\dagger}(\tau) \alpha_{l\downarrow}^{\dagger}(\tau) \right]
\]
\[
\text{S62}
\]
Since the coefficient of the Raman correlation function \( ^{1}X_{l,t',t''} \) is symmetric with respect to the replacements \( l_{+} \leftrightarrow l'_{+} \) and \( l_{-} \leftrightarrow l''_{+} \), the 4M GFs can be expressed as

\[
G_{l_{+},l',l'',t'}^{k,k',k'',t''}(t) \simeq 2G_{l_{+},l',l'',t'}^{k,k',k'',t''}(t) + \frac{i}{2}G_{l_{+},l',l'',t'}^{k,k',k'',t''}(t),
\]

(S62)

and their Fourier transforms are given by

\[
G_{l_{+},l',l'',t'}^{k,k',k'',t''}(\omega) \simeq \int_{-\infty}^{\infty} dt \, e^{i\omega t} \left[ 2G_{l_{+},l',l'',t'}^{k,k',k'',t''}(t) + \frac{1}{2}G_{l_{+},l',l'',t'}^{k,k',k'',t''}(t) \right]
\]

\[
= \int_{-\infty}^{\infty} \frac{ds}{2\pi} \left( 2G_{l_{+},l',l'',t'}^{k,k',k'',t''}(\omega - s) + \frac{1}{2}G_{l_{+},l',l'',t'}^{k,k',k'',t''}(\omega - s) \right)
\]

\[
= 2 \sum_{\alpha l_{+}} \frac{1}{\hbar \omega - \hbar \Omega_{0}^{\alpha} + \hbar \omega_{l'_{+}} + i\eta} \frac{1}{2} \sum_{\alpha l_{+}} \frac{1}{\hbar \omega - \hbar \Omega_{0}^{\alpha} + \hbar \omega_{l''_{+}} + i\eta}.
\]

(S63)

S4. Irreducible Decomposition of Raman Operators

The 2D Raman operator reads

\[
[^{\mu}]R = \sum_{\mu_{m},\nu_{m},y} e_{m}^{\mu} [^{\mu}]R^{\mu \nu} e_{m}^{\nu},
\]

(8)

where \( e_{m}^{\mu} \equiv (e_{m}^{x}, e_{m}^{z}) \) and \( e_{c}^{\mu} \equiv (e_{c}^{x}, e_{c}^{z}) \) are the unit vectors indicating the polarizations of incident and scattered photons, respectively, while \( [^{\mu}]R^{\mu \nu} \) is the \((\mu, \nu)\)-element of \([^{\mu}]R\) in Cartesian coordinates. We introduce four matrices

\[
\Xi_{1} = \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}, \quad \Xi_{2} = \begin{bmatrix} 0 & 1 \\ -1 & 0 \end{bmatrix}, \\
\Xi_{3} = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}, \quad \Xi_{0} = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}
\]

(S64)

with their Hilbert-Schmidt inner products satisfying

\[
\text{Tr} \left[ \Xi_{i} \Xi_{j} \right] = \sum_{\mu_{m},\nu_{m},\nu_{c}} \Xi_{i}^{\mu \mu_{m}} \Xi_{j}^{\nu_{c} \nu_{m}} = 2 \delta_{i,j}
\]

(S65)

to rewrite the Raman operator (8) into

\[
[^{\mu}]R = \sum_{i=0}^{\infty} E_{\Xi,i} \left([^{\mu}]R^{\mu \nu} e_{m}^{\nu} \right)
\]

(S66)

where \( E_{\Xi,i} \equiv e_{m}^{x} e_{c}^{x} + e_{m}^{z} e_{c}^{z} \), \( E_{\Xi,1} \equiv e_{m}^{x} e_{c}^{x} - e_{m}^{z} e_{c}^{z} \), \( E_{\Xi,2} \equiv e_{m}^{x} e_{c}^{x} - e_{m}^{z} e_{c}^{z} \), \( E_{\Xi,3} \equiv e_{m}^{x} e_{c}^{x} + e_{m}^{z} e_{c}^{z} \).

We recall the irreducible decomposition of the Raman operator for an arbitrary point symmetry group \( P \).

\[
[^{\mu}]R = \sum_{\mu_{1},\nu_{1}} d_{\Xi_{1}}^{\mu_{1} \nu_{1}}[^{\mu_{1}}R^{\mu_{1} \nu_{1}}],
\]

(9)

to consider 2D lattices of \( C_{nv} \) point symmetry in general. The polarization-vector basis functions \( E^{\alpha}_{\Xi,i} \) relevant to Raman scattering have their equivalent in \( E_{\Xi,i} \)'s [cf. (12) and (S66a)]. We list their correspondence relations in Table I.

Depolarization of the Loudon-Fleury second-order Raman response, such as \( (14) \), is the consequence of \( [^{1}R] \) containing one and only multidimensional irreducible representation.\(^{15}\) In Table I, neither \( C_{2n} \) nor \( C_{4v} \) meets this criterion, while all the rest do. Let us investigate \( C_{nv} \) symmetry operations on \( \Xi_{i} \), intending to reveal the possible dimensionality \( d_{\Xi_{i}}^{\mu_{1} \nu_{1}} \). We denote the matrix representation for a point symmetry operation \( P \in P \) by \( \mathcal{P} \). Setting \( P \) to the rotation \( C_{nv}^{n} \) in \( C_{nv} \),

\[
\mathcal{P} = \begin{bmatrix} \cos \frac{2\pi}{n} & -\sin \frac{2\pi}{n} \\ \sin \frac{2\pi}{n} & \cos \frac{2\pi}{n} \end{bmatrix}
\]

(S67)

we obtain

\[
\mathcal{P}^{-1} \Xi_{0} \mathcal{P} = \Xi_{0},
\]

(S68a)

\[
\mathcal{P}^{-1} \Xi_{1} \mathcal{P} = \Xi_{1} \cos \frac{4\pi}{n} = \Xi_{3} \sin \frac{4\pi}{n},
\]

(S68b)
and \( (S55a) \) and \( (S55b) \) for 3M GFs) in mind, we retain only the magnon-number-conserving interactions and properties of the A\(_3\) symmetry species, where we specify a particular lattice to the point symmetry groups each, because it depends on the lattice shape which symmetry species is Raman active. In the case of \( P = C\(_{2v}\) \), not only \( \Xi_0 \) but also \( \Xi_3 \) may belong to the A\(_3\) symmetry species and the coefficients of their linear combination depend on further details of the lattice.

| \( P \)                         | \( \Xi_0 \) | \( \Xi_1 \) | \( \Xi_2 \) | \( \Xi_3 \) |
|-------------------------------|------------|------------|------------|------------|
| \( C\(_{2v}\) \) (ladder)     | \( A_1 : 1 \) | \( A_2 : 1 \) | \( A_1 : 1 \) | \( A_1 : 1 \) |
| \( C\(_{3v}\) \) (kagome)     | \( A_1 : 1 \) | \( E : 1 \) | \( A_2 : 1 \) | \( E : 2 \) |
| \( C\(_{4v}\) \) (square)     | \( A_1 : 1 \) | \( B_2 : 1 \) | \( A_2 : 1 \) | \( B_1 : 1 \) |
| \( C\(_{6v}\) \) (triangular) | \( A_1 : 1 \) | \( E_2 : 1 \) | \( A_2 : 1 \) | \( E_2 : 2 \) |
| \( C\(_{6v}\) \) (honeycomb)  | \( A_1 : 1 \) | \( E_2 : 1 \) | \( A_2 : 1 \) | \( E_2 : 2 \) |
| \( C\(_{5v}\) \) (Penrose)    | \( A_1 : 1 \) | \( E_2 : 1 \) | \( A_2 : 1 \) | \( E_2 : 2 \) |
| \( C\(_{5v}\) \) (Ammann-Beenker)| \( A_1 : 1 \) | \( E_2 : 1 \) | \( A_2 : 1 \) | \( E_2 : 2 \) |

we have

\[
P^{-1} \Xi_0 P = \Xi_0, \quad \Xi_1 P = -\Xi_1 \cos \frac{4\pi l}{n} + \Xi_3 \sin \frac{4\pi l}{n}, \quad P^{-1} \Xi_2 P = -\Xi_2, \quad \Xi_3 \cos \frac{4\pi l}{n} + \Xi_1 \sin \frac{4\pi l}{n}. \]

(S70a)

(S70b)

(S70c)

(S70d)

\( \Xi_0 \) and \( \Xi_2 \) each correspond to a 1D irreducible representation for any point symmetry group \( C\(_{nv}\) \), whereas \( \Xi_1 \) and \( \Xi_3 \) span a 2D irreducible representation unless \( n = 2 \) or \( n = 4 \). No quasiperiodic lattice in two dimensions belongs to either of \( C\(_{2v}\) \) and \( C\(_{4v}\) \), and therefore, depolarization of \( [2]l(\omega) \), which we observe in Figs. 1(a) and 1(a') is common to all 2D quasiperiodic lattices.

S5. Configuration-Interaction Formalism

In order to give a precise description of multimagnon-mediated inelastic light scatters, we consider interactions between the up-to-4M basis states

\[
[0M] \equiv [0]_{BL}, \quad [2M]^{l}_{L} \equiv \alpha_{l}^{+} \alpha_{l}^{-} [0]_{BL}, \quad [4M]^{l,r}_{L} \equiv \sqrt{1 + \delta_{l,r}} \alpha_{l}^{+} \alpha_{l}^{-} [0]_{BL} (1 \leq \ell, \ell' \leq L_{\sigma}).
\]

(S71a)

(S71b)

(S71c)

Considering that the Hamiltonian as well as the Raman operator commutes with the total magnetization, any other 2M and 4M states, changing the total magnetization, are ineffective in the ground-state Raman response. The up-to-\( O(5) \) 2M-4M-configuration-interaction (CI) Hamiltonian is formally written as

\[
\mathcal{H} = \mathcal{H}_{\text{bl}} + \mathcal{H}^{(0)}.
\]

With the ladder-approximation Bethe-Salpeter equation formalism \cite{[5.49a)–(5.49c) for 2M GFs and (5.55a) and (5.55b) for 3M GFs]} in mind, we retain only the magnon-number-conserving interactions \( V^{(1)}_{ij,j,j',j''} \), \( V^{(4)}_{ij,j,j',j''} \) and \( V^{(9)}_{ij,j,j',j''} \) in our CI scheme. Then (S72) reduces to the block-diagonal Hamiltonian

\[
\mathcal{H} = \sum_{m=0}^{3} E^{(m)} = \begin{bmatrix} 0 & \langle 2M|H|2M \rangle_{L}^{(1)} & \cdots & \langle 2M|H|2M \rangle_{L}^{(3)} \\ \langle 2M|H|2M \rangle_{L}^{(2)} & \cdots & \langle 2M|H|2M \rangle_{L}^{(3)} \\ \vdots & \cdots & \vdots \\ \langle 2M|H|2M \rangle_{L}^{(3)} & \cdots & \langle 2M|H|2M \rangle_{L}^{(3)} \\ \end{bmatrix}
\]

(S73)
If we discard the 4M basis states (S71c) in (S73), the resultant 2M-CI findings for $|\psi_I|_{2M}\langle\omega|$ are exactly the same as the ladder-approximation Bethe-Salpeter calculations. There is a complete correspondence of the 2M sector (S73b) of any CI magnon-number-conserving block-diagonal Hamiltonian with the 2M Bethe-Salpeter interaction matrix $V^{(4)}$ (S53a). (S49b) and (S49c) are irrelevant to any calculation of $|\psi_I|_{2M}\langle\omega|$ but necessary for calculating $|\psi_I|_{2M}\langle\omega|$, or more precisely, for decomposing the 4M GFs as (S62) and (S62'). When we go beyond the Loudon-Fleury second-order perturbation theory and take an interest in multimagnon-mediated Raman intensities, the 2M-4M-CI scheme is much superior to any tractable self-consistent GF formalism.

S6. Configuration-Interaction versus Green’s Function Calculations of Raman Spectra

We show the cluster-size and calculational-scheme dependence of Raman spectra in full detail for both the Penrose (Figs. S4 to S6) and Ammann-Beenker (Figs. S7 and S8) lattices, intending to demonstrate the superiority of the 2M-4M-CI scheme over the others especially in evaluating 4M-mediated scattering intensities.

For the $L = 16$ Penrose (Fig. S4) and $L = 25$ Ammann-Beenker (Fig. S7) clusters, all the perturbative and variational calculations are compared with the exact solutions. The Hartree-Fock approximation cannot reproduce any of the major peak positions. The 2M-CI formulation, which is equivalent to the 2M Bethe-Salpeter equation, is good at reproducing the low-energy peaks essentially of 2M character but poor in describing higher-energy spectral weight. The 2M-4M-CI formulation overcomes this drawback owing to its precise evaluation of 4M-mediated scattering intensities. This is not the case with any of the GF findings. Indeed both (3 + 1)M and (2 + 2)M approximate evaluations of the 4M Raman correlation function $|\psi_I|_{2M}\langle\omega|$ lead to a reasonable reduction of the high-energy excess spectral weight, but neither of them gives such a satisfactory description of the intermediate-energy scattering bands of 2M-4M-mixed character as to be obtained through the 2M-4M-CI scheme. Such observations are common to both the Penrose and Ammann-Beenker lattices and hold good for all the symmetry species but $A_1$. Any excess 4M scattering intensity for the $A_1$ symmetry species present in the SW calculations should rather be ascribed to the up-to-$O(5)$ expansion of $R$ than otherwise. Note that the exact diagonalization calculation of the Raman intensity (6) is performed with the spin operator expressions (S30) and (S35), whereas any SW calculation, whether in the GF description (7) or through the CI scheme (17), is performed with the up-to-$O(5)$ approximate vertices (4) written in terms of the magnon operators (S44) and (S45), with the aim of revealing the scenario of inelastic light scattering.

With increasing system size, the spectral shape and/or density change in a complicated manner, but the balance in intensity sharing between 2M and 4M scatterings remains qualitatively the same. The energy ranges in which 2M scattering intensities dominate for the three symmetry species each remain almost unchanged from those of the smallest cluster calculated and this is essentially the case with 4M scattering intensities as well. Indeed, neither of the range of the eigenvalue distribution nor the specific heat curve is sensitive to the system size for the $L \geq 31$ Penrose and $L \geq 33$ Ammann-Beenker lattices, as was already shown in Fig. S1. Eigenvalues corresponding to the one or two highest coordination numbers have no serious effect on bulk properties.
Fig. S4. CI and GF calculations of the Shastry-Shraiman fourth-order Raman intensities $I_{2\mu}(\omega) \equiv \sum_{\nu=1}^{2L} |I_{2\mu\nu}(\omega)|$ for the $L = 16$ 2D Penrose lattice of $C_{5v}$ point symmetry in comparison with the exact solutions, where the perturbation parameter $t/(U - \hbar \omega)$ is set to $9/10$ and every spectral line is Lorentzian-broadened by a width of $0.1J$. From the top to the bottom, the calculational schemes employed are the Hartree-Fock approximation [(a1) to (c1)], which retains only the magnon vacuum in (S71a) and substitutes the eigenstates and eigenvalues of $H_{\text{HF}}(2)$ for the basis states $|\nu\rangle$ and their energies $\epsilon_{\nu}$, with $N_{\text{CI}}$ reducing to $L$ in (17), 2M-CI [(a2) to (c2)], which is equivalent to solving the 2M Bethe-Salpeter equation (S49a), 2M-4M-CI [(a3) to (c3)], $2M + 4M$ [approximated by (S62′)]-GF [(a4) to (c4)], and $2M + 4M$ [approximated by (S62)]-GF [(a5) to (c5)]. The pure symmetry components are extracted from three polarization combinations, (15) with $\phi = 0, \pi/2$ and (16) with $\sigma_{\text{out}} = -1$. All the SW calculations of $I_{2\mu}(\omega)$ each are distinguishably colored.
Fig. S5. The same as Fig. S4 for the $L = 41$ 2D Penrose lattice of $C_{5v}$ point symmetry without any exact solution available.
Fig. S6. The same as Fig. S4 for the $L = 56$ 2D Penrose lattice of $C_{5v}$ point symmetry without any exact solution available.
Fig. S7. The same as Fig. S4 for the $L = 25$ 2D Ammann-Beenker lattice of $C_{8v}$ point symmetry.
Fig. S8. The same as Fig. S7 for the $L = 57$ 2D Ammann-Beenker lattice of $C_{8v}$ point symmetry without any exact solution available.
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