Torque equilibrium spin wave theory of Raman scattering in an anisotropic triangular lattice antiferromagnet with Dzyaloshinskii-Moriya interaction

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We apply torque equilibrium spin wave theory (TESWT) to investigate an anisotropic XXZ antiferromagnetic model with Dzyaloshinskii-Moriya (DM) interaction in a triangular lattice. Considering the quasiparticle vacuum as our reference, we provide an accurate analysis of the non-collinear ground state of a frustrated triangular lattice magnet using the TESWT formalism. We elucidate the effects of quantum fluctuations on the ordering wave vector based on model system parameters. We study the single magnon dispersion, the two-magnon continuum using the spectral function, the Raman spectrum of bimagnon and trimagnon excitations. We present our results for the $HH$, $VV$ and the $HV$ polarization Raman geometry dependence of the bimagnon and trimagnon excitation spectrum where $H(V)$ represents horizontal (vertical) polarization. Our calculations show that both the $HH$ and the $HV$ polarization spectrum can be used to determine the degree of anisotropy of our system. We calculate the Raman spectra of $\text{Ba}_2\text{CoSb}_2\text{O}_9$ and $\text{Cs}_3\text{CuCl}_4$.

I. INTRODUCTION

The effect of quantum fluctuations on the ground state and the phase diagram of frustrated magnets has been a topic of interest in recent years [1–8]. The two-dimensional triangular lattice antiferromagnet (TLAF) is a canonical example of a frustrated magnetic system. There are several examples of TLAF, see Table I. Due to the melting of long-range magnetic order by frustration [9, 10], the two-dimensional triangular lattice is considered as a natural spin liquid candidate [11, 12]. Even when magnetic long-range-order exists, the competition between various interactions have consequences on the ground state and the phase diagram, especially for low-dimensional spin systems [13, 14]. Quantum fluctuations can be non-negligible even for ordered magnets with 120◦ spiral order [15–17].

The non-collinear spin structure of the triangular lattice leads to interesting phenomena such as the presence of a roton minimum [18, 19] and a continuum of high-energy magnons [20–22]. Similar to superfluid $^4\text{He}$ [23] and fractional quantum Hall systems [24], Zheng et. al. [20–22] defined the $M$ and $M'$ points of the Brillouin zone (BZ) of a triangular lattice as rotonlike points. The formation of the local minimum is caused by quantum fluctuations [25, 26]. The roton signal has been observed in inelastic neutron scattering experiments [16, 18, 27, 28]. However, the nature of the high-energy continuum in the triangular lattice is still controversial. The continuous excitation at high energy [20–22] may come from fractional excitation of a proximate spin-liquid phase [29–33] or from strong magnon-magnon interactions [34–36]. In this context, Raman spectroscopy serves as a powerful tool to probe lattice distortions and the effect of ground state quantum fluctuations. It has already been used to detect magnon excitations in TLAF [37–43]. Raman’s advantage is the sensitivity to polarization geometry [44] and magnon-magnon interactions [45, 46], which is helpful for studying the high energy continuum.

Raman bimagnon calculation for anisotropic TLAF has been calculated within the framework of interacting spin wave theory with the scattering operator defined similar to that of a square lattice [46]. However, the presence of divergence in the ordering wave vector and singularity of the spin wave spectrum calls for renewed attention to accurately describe the non-collinear frustrated triangular lattice magnet [47] beyond the $1/S$-spin wave theory analysis. The Raman spectrum should be carefully reconsidered with appropriate quantum fluctuation effects and with proper underlying lattice symmetry. The recently established torque equilibrium spin wave theory (TESWT) considers the spin Casimir effect of a non-collinear system caused by the zero-point quantum fluctuations [13]. This formalism cures the ordering wave vector of any divergence. The computed phase diagram of the anisotropic TLAF is consistent with series expansion (SE) and modified spin wave theory (MSWT) [3, 48]. Since quantum fluctuations cause modification of the ordering wave vector, its influence on the magnon and multi-magnon excitations (bi- and tri-magnon) is an important question to investigate.

In this study, we extend the analysis of the $J-J'$ triangular lattice Heisenberg magnet to the case of a XXZ model with DM interaction. First, we apply TESWT to obtain the ordering wave vector. We find that the DM interaction is more favorable to stabilizing the helix state compared to XXZ anisotropy. Similar to the quasi-one-dimensional helimagnets [14], the mere presence of XXZ anisotropy can lead to a shift in the phase boundary. Second, we calculate the spectral function within the TESWT framework. We find that the magnon excitations are more stable with DM interaction and XXZ anisotropy. Our calculations, show the presence of quasiparticle excitation and continuum in the spectral function. Third, we calculate the polarization-dependent bi- and trimagnon Raman spectrum under TESWT. Distinct from the non-interacting calculation, the bimagnon spectrum in the

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TABLE I. Antiferromagnetic triangular lattice materials. We report Raman scattering spectrum for Ba$_2$CoSb$_2$O$_9$ and Cs$_2$CuCl$_4$. The torque equilibrium spin wave theory approach used to compute the Raman spectrum can be applied to any of the ordered triangular lattice materials listed below. The last column states the ordering wave vector.

| Material          | $T_N (K)$ | Space group | $Q$          |
|-------------------|-----------|-------------|--------------|
| Ba$_2$CoSb$_2$O$_9$ [21, 49, 50] | 3.8       | P6$_3$/mmc  | (2/3,0,1)    |
| CuCrO$_2$ [51, 52] | 24.2      | R3m         | (0.658,0,0)  |
| a-SrCr$_2$O$_4$ [17, 53] | 43        | Pmmm        | (0.6609,0,1) |
| a-GaCr$_2$O$_4$ [16, 54] | 43        | Pmmm        | (0.6659,0,1) |
| LuMnO$_3$ [22]    | 90        | P6$_3$cm    | (2/3,0,0)    |
| Cs$_2$CuCl$_4$ [20, 47, 55] | 0.62      | Pnma        | (0.530,0,0) [20] |
| Cs$_2$CuBr$_4$ [55–57] | 1.4       | Pnma        | (0.575,0,0)  |

HV polarization displays a single peak feature with magnon-magnon interactions considered. We find that the bimagnon intensity is polarization-independent for the isotropic TLAF. However, the bimagnon excitation occurs only in HV polarization for the anisotropic TLAF. In the HV polarization, spatial anisotropy reduces the bimagnon intensity and peak energy. DM interaction also reduces its intensity, especially in a system with increasing spatial anisotropy. In spite of a spin gap, DM interaction induces an upshift of the bimagnon peak a system with increasing spatial anisotropy. In spite of a spin energy. DM interaction also reduces its intensity, especially in a system with increasing spatial anisotropy. However, the bimagnon excitation occurs only in HV polarization for the TLAF. We also compute the Raman spectra of Ba$_2$CoSb$_2$O$_9$ and Cs$_2$CuCl$_4$. For Ba$_2$CoSb$_2$O$_9$, its Raman spectrum has a sharp peak and a broad shoulder in both the HH and the HV polarization. For Cs$_2$CuCl$_4$, its Raman spectrum has a sharp bimagnon peak in HV polarization. We find that the primary contribution to the Raman intensity in the HH polarization comes from the trimagnon excitation. However, in the HV polarization, both the bimagnon and the trimagnon excitation mix and give a broad spectrum.

This article is organized as follows. In Sec. II we introduce the XXZ model with spatial anisotropy and DM interaction. In Sec. III we compute the spin wave spectrum and spectral function by applying TESWT. In Sec. IV we utilize TESWT to calculate the bimagnon and trimagnon Raman spectrum. In Sec. IV A we derive the expressions for the Raman operator, their polarization dependence, and the magnon-magnon interaction effects. In Sec. IV B we present and discuss our results on spatial anisotropy, spin anisotropy, magnon-magnon interaction, and polarization dependence. Then we present the Raman spectra of Ba$_2$CoSb$_2$O$_9$ and Cs$_2$CuCl$_4$. Finally, in Sec. V we provide our conclusions.

II. MODEL

Triangular lattice materials can contain spatial or spin anisotropies. In the case of Cs$_2$CuCl$_4$ and Cs$_2$CuBr$_4$, DM interaction is present and generates a spin gap. However, the large gap of Cs$_2$CuBr$_4$ in the energy dispersion cannot be generated exclusively by DM interaction [57, 58]. Additionally, spin-orbit coupling may lead to XXZ anisotropy which has been used to explain the presence of a gapped spectrum in some TLAF materials [49, 59]. Amongst them is Ba$_2$CoSb$_2$O$_9$. Even though it is spatially isotropic in its exchange interaction, its magnetization is well explained by a spin-1/2 XXZ (spin-anisotropic) model on a triangular lattice [60–62]. Thus, to conduct a thorough study of the frustrated TLAF systems, we consider XXZ anisotropy in addition to anisotropic exchange interaction and DM interaction. Our model is written as

$$\mathcal{H} = J \sum_{\langle ij \rangle} \left[ S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z \right] + J' \sum_{\langle ij \rangle} \left[ S_i^x S_j^z + S_i^z S_j^z + \Delta S_i^x S_j^x \right] - \sum_{\langle ij \rangle} D \cdot \left( S_i \times S_j \right),$$

where $\langle ij \rangle$ refers to nearest-neighbor bonds on the triangular lattice and $\delta_{1,2}$ are the nearest-neighbor (nn) vectors along the diagonal bonds, see Fig. 1(a). The four parameters $(J, J', D, \Delta)$ in the model correspond to the exchange constants along the horizontal bonds, the exchange constants along the diagonal bonds, DM interaction along the $l_0$ direction ($D>0$) [20], and XXZ spin anisotropy, respectively.

The spin spiral ground state can be described by an ordering wave vector $Q$. To analyze the spin wave spectrum of this magnetic model, we first transform from the lab to the rotated local coordinate frame [47]. Then, the successive applications of the Holstein-Primakoff (HP), Fourier, and Bogoliubov transformations give us the effective first-order 1/S
expansion Hamiltonian as
\[
\mathcal{H}_{\text{eff}} = \sum_k \left[ (S_\varepsilon k + \delta \varepsilon k) c_k^\dagger c_k + \frac{O_k}{2} (c_k^\dagger c_k - c_k c_- k) \right]
\]
\[
+i \sqrt{\frac{S}{2N}} \sum_{k} \Phi_\sigma (1, 2; 3) c_k^\dagger c_2^\dagger c_3
\]
\[
+ \frac{1}{3!} \sum_{k} \Phi_\sigma (1, 2, 3) c_k^\dagger c_2^\dagger c_3^\dagger + \text{H.c.}
\]
\[
+ \frac{1}{8N} \sum_{k} \Phi_\sigma (1, 2; 3, 4) c_k^\dagger c_2^\dagger c_3^\dagger c_4,
\]

where \(c_k^\dagger (c_k)\) is the quasiparticle creation (annihilation) operator in momentum space. \(1, 2, \ldots\) denote \(k_1, k_2, \ldots\). The formulae of the vertex coefficients \(\Phi_\sigma\), \(\Phi_\sigma\), and \(\Phi_\sigma\) are given in Appendix A. We set \(J = 1\) meV in all our subsequent calculations. Thus, our model has three parameters \((J', D, \Delta)\), where \(J'\) and \(D\) represent the relative interaction strengths.

In the next section, we will analyze the above bosonized Hamiltonian for its spin wave spectrum. As mentioned before, the \(1/S\)-spin wave theory cannot treat the quantum fluctuations appropriately, which in turn leads to divergences and singularities in the calculation of the ground state [14]. Thus, we will analyze this model using TESWT.

### III. SPIN WAVE SPECTRUM BY TESWT

Torque equilibrium spin wave theory formalism gives the correct ground state and phase diagram for spin spiral magnets [13, 14, 47]. The phase diagram that results from TESWT formalism is consistent with previous numerical calculations [3, 48]. The essential conceptual difference between spin wave theory and TESWT is the reference ground state. The former considers the classical vacuum as the ground state, while the later considers the quasiparticle vacuum state as the correct starting point. Though linear spin wave spectrum \(\varepsilon_k\) is physically well-behaved at the classical ordering wave vector \(Q_\sigma = (Q_{cl}, 0, 0)\), it yields an incorrect ground state wave vector, see Fig. 2. Within the TESWT approach the goal is to find a redefined Hamiltonian whose classical ordering wave vector \(\overline{Q}_\sigma\) is equal to the final ordering wave vector \(Q\) of the original state. Henceforth, the tilde variable will signify parameters of the torque equilibrium shifted Hamiltonian.

To implement TESWT we rewrite the quadratic term of our model as \(H_2(J', D, \Delta, Q) = \overline{H}_2(J', \tilde{D}, \tilde{\Delta}, \tilde{Q}) + H_2\), where the superscript \(c\) represents the counterterm which will regularize the original singular Hamiltonian. Due to the small values of \(D\) and \((1 - \Delta)\) in real materials, we take \(\tilde{D} = D\) and \(\tilde{\Delta} = \Delta\). Next, the spin Casimir torque is defined as

\[
T_{\text{sc}}(Q) = \sum_k \left| \langle \Psi_{\text{vac}} | \partial H_{\text{vac}} / \partial Q \right| \Psi_{\text{vac}} \rangle,
\]

where \(\Psi_{\text{vac}}\) represents the expectation value of the quasiparticle vacuum state. Next, we utilize the torque equilibrium condition, within the approximation of \(T_{\text{sc}}(Q) = \tilde{T}_{\text{sc}}(Q)\), to obtain the final ordering wave vector as

\[
\frac{\partial E_{\text{eff}}(Q)}{\partial Q} + \sum_k \frac{\partial E_k}{\partial Q} = 0,
\]

where \(F = F(J', \tilde{D}, \tilde{\Delta}, Q)\) (where \(F\) is an arbitrary operator). The corresponding functions are shown in Appendix A.

Figure 2(a) shows the ordering vector \(Q\) of the spin-1/2 system obtained using TESWT. Without DM interaction and XXZ anisotropy, the TLAF orders in an antiferromagnet phase for \(J' \geq 1.2\). DM interaction influences the ordering vector more than XXZ anisotropy. It enlarges the region of spiral phase. Fig. 2(b) shows the difference of ordering vector between TESWT and linear spin wave theory (LSWT). For \(J' \leq 1\), TESWT gives a smaller \(Q\) (compared to LSWT) and the ground state becomes closer to the ferromagnet. Whereas, with \(J' \geq 1\) TESWT gives a larger \(Q\) and the ground state will be nearly antiferromagnetic in arrangement. Thus we conclude that the spin Casimir effect induces collinear arrangement of spins. Note, since we are analyzing a coplanar non-collinear spin configuration we will restrict our XXZ anisotropy values. It is evident from Fig. 2(b) that with DM interaction, the difference between TESWT and LSWT becomes smaller is minimized, indicating that it weakens quantum fluctuations. The Hamiltonian shift results in the one-loop torque equilibrium effective Hamiltonian given by

\[
\tilde{\mathcal{H}}_{\text{eff}} = \sum_k \left[ (S \tilde{\varepsilon}_k + \tilde{\delta} \varepsilon k) c_k^\dagger c_k + \frac{\tilde{O}_k}{2} (c_k^\dagger c_k - c_k c_- k) \right]
\]

\[
+ \frac{S}{2N} \left[ \sum_{k} \Phi_\sigma (1, 2; 3) c_k^\dagger c_2^\dagger c_3 \right]
\]

\[
+ \frac{1}{3!} \sum_{k} \Phi_\sigma (1, 2, 3) c_k^\dagger c_2^\dagger c_3^\dagger + \text{H.c.}
\]

\[
+ \frac{1}{8N} \sum_{k} \Phi_\sigma (1, 2; 3, 4) c_k^\dagger c_2^\dagger c_3^\dagger c_4,
\]
with

$$
\epsilon_k^a = \frac{1}{\epsilon_k} (\Delta \vec{A}_k \vec{B}_k - \vec{B}_k \vec{A}_k) - \vec{\tilde{\varepsilon}}_k,
$$

$$
\hat{\Omega}_k^a = \frac{1}{\epsilon_k} (\vec{A}_k \vec{B}_k - \vec{B}_k \vec{A}_k).
$$

In such a non-colinear spin system, we consider the renormalization of magnon dispersion up to 1/3 order. Thus, the counterterm contributions from $H_3$ and $H_4$ are neglected [13, 14]. Within this scheme the first-order renormalized Green’s function in the one-loop approximation is given by

$$
G^{-1}(k, \omega) = \omega - \Delta \vec{\tilde{\varepsilon}}_k - \sum S_{\epsilon\theta}^a(k - \delta_k) + \sum \delta_{\epsilon\theta}^a(k, \omega) + \sum \delta_{\epsilon\theta}^b(k, \omega),
$$

where $S_{\epsilon\theta}^a$ is the counterterm from $H_2$, $\delta_{\epsilon\theta}^a$ describes the quartic correction following mean-field averages. $\delta_{\epsilon\theta}^b(k, \omega)$ are the self-energy contributions from the cubic interaction and are given by

$$
\delta_{\epsilon\theta}^b(k, \omega) = \frac{S}{4N} \sum_k \frac{|\vec{D}_a(k_1 - k_1, \cdots, k_1)|^2}{\omega - S_{\epsilon\theta}^a(k) - S_{\epsilon\theta}^b(k) + \imath \omega^*},
$$

$$
\delta_{\epsilon\theta}^b(k, \omega) = -\frac{S}{4N} \sum k \frac{|\hat{D}(k_1 - k_1, \cdots, k_1)|^2}{\omega + S_{\epsilon\theta}^a(k) + S_{\epsilon\theta}^b(k) - \imath \omega^*}.
$$

In the on-shell approximation, the first-order renormalized magnon energy can be calculated as

$$
\omega_k = S\vec{\tilde{\varepsilon}}_k + S\epsilon_k^a + \sum_{\epsilon\theta}^a(k, \vec{\tilde{\varepsilon}}_k) + \sum \delta_{\epsilon\theta}^a(k, \vec{\tilde{\varepsilon}}_k) + \sum \delta_{\epsilon\theta}^b(k, \vec{\tilde{\varepsilon}}_k).
$$

The real and imaginary part of $\omega_k$ are magnon dispersion and magnon decay, respectively. To obtain an intuitive understanding of the single magnon excitation, we calculate the spectral function, which is defined as

$$
A(k, \omega) = -\frac{1}{\pi} \text{Im} G(k, \omega).
$$

In Fig. 3 we report our spectral function calculation for various model parameters. The intensity plots show broadening of the quasi-particle excitation and presence of two-magnon continuum in all the panels, which is consistent with a previous study on isotropic TLAF [36]. Thus, magnon-magnon interactions are important in the triangular lattice. Compared to Ref. 36, spatial anisotropy causes a downshift of the continuum energy. In Fig. 3(a) the dispersion shows no gap at the ordering wave vector. This is consistent because the DM interaction is set to zero and the Hamiltonian is at the spin-isotropic point $\Delta = 1$. From Fig. 3(b) and 3(c) we observe that DM interaction and XXZ anisotropy can suppress damping and stabilize magnon excitations. Both XXZ anisotropy and DM interaction can generate gaps at the ordering wave vector, thereby reducing magnon decay. However, DM interaction has a greater effect of suppression on magnon decay than XXZ anisotropy. In Fig. 3(b), the consistency between the spectral function and dispersion indicates that the on-shell calculation is more reasonable with DM interaction. However, the spectrum is inconsistent with the on-shell dispersion when $J>1$, see Fig. 3(d). Thus, we restrict our Raman calculations to parameters where the spatially anisotropic exchange interaction does not exceed one. In fact, this is a valid parameter regime for real materials [47, 57, 59, 60].

### IV. TORQUE EQUILIBRIUM SPIN WAVE THEORY

**Raman Spectrum**

Next we study the polarized Raman scattering of TLAF to investigate the bi- and trimagnon excitation behavior. We consider the magnon-magnon interactions to study the continuum shown in Fig. 3. Raman scattering is a valid tool to detect magnon excitations due to its sensitivity to magnon-magnon interactions and polarization [37–41, 44–46]. A previous RIXS calculation [47] on TLAF has investigated the polarization-independent bi- and trimagnon spectrum. An unpolarized Raman scattering has been reported in $\alpha$-SrCr$_2$O$_4$ [41], which is a distorted TLAF. As the highest energy of one magnon is about 20 meV [17], the high-energy peak above 40 meV in Raman experiment [41] of $\alpha$-SrCr$_2$O$_4$ may be a trimagnon excitation. In addition to the unpolarized Raman detection, the polarization-dependent Raman spectrum may provide another view to study the features of bi- and trimagnon excitation. In comparison to RIXS, Raman scattering is a more mature technique which can detect the $\mathbf{q} \approx 0$ magnon excitation. Till date, from a theoretical perspective, a substantial number of studies have been pursued within LSWT and an interacting framework to investigate the Raman spectrum of TLAF Heisenberg model [37, 38, 45, 46]. However, since LSWT leads to a divergent ordering wave vector and fails to describe the ground state, it is not suitable to calculate the magnon excitation. Thus, we apply the TESWT to study the Raman bi- and trimagnon excitation of TLAF. One of the key developments reported in this paper is on trimagnon calculation and our discussion of the polarization dependence of the bi- and trimagnon excitation. Neglecting polarization, the bimagnon intensity is zero at the $\Gamma$ point. This implies being in the $A_1$ mode of the multi-magnon excitation. However, the real spectrum of the anisotropic TLAF is polarization dependent. Next, we discuss the polarization dependence and how it helps to analyze the composition of Raman spectrum.

#### A. Raman scattering operator and interactions

Standard perturbation theory formalism applied to electron-radiation interaction can be used to compute the Raman scattering cross-section [63–65]. The expression for the polarization-dependent second-order Raman scattering operator of our model is

$$
\hat{\mathcal{O}} = \sum_{i \neq \delta j} \mathcal{P}_j(\theta, \phi) \times \left[ J_{\delta j}(S_i^+ S_i^{- \delta j} + S_i^- S_i^{+ \delta j} + \Delta S_i^+ S_i^{\delta j}) - D_j \cdot (S_i \times S_i^{\delta j}) \right],
$$

where $\delta_j$ denote the lattice vectors: $\delta_1 = \left( \frac{1}{2}, 0, \frac{\sqrt{3}}{2} \right)$, $\delta_2 = \left( \frac{1}{2}, 0, -\frac{\sqrt{3}}{2} \right)$ and $\delta_3 = (1, 0, 0)$ [66]. The polarization geometry and the symmetry of the experimental setup are captured in the $\mathcal{P}_j(\theta, \phi)$ operator coefficient. We consider the polarization of the incoming and outgoing light as $\hat{\mathbf{e}}_{in} = (\cos \theta, 0, \sin \theta)$
and $\tilde{E}_{\text{out}} = (\cos \phi, 0, \sin \phi)$, respectively, where $\theta$ and $\phi$ are defined with respect to the $x_0$ axis. The sketch of an experimental geometry is shown in Fig. 1(b). Since we are considering a quasi-2D TLAF, for generality, we calculated the Raman spectrum of both the isotropic ($C_{3v}$) and the anisotropic case ($C_{2v}$), respectively. The Raman-active modes of the $C_{3v}$ and $C_{2v}$ systems are given by the irreducible representations $A_1 + E$ and $A_1 + A_2$, respectively.

In terms of the Bogoliubov magnons the polarized Raman scattering operator takes the following form

$$
\hat{\mathcal{O}} = \sum_{\mathbf{k}} \tilde{B}_{\mathbf{k}} (c_{\mathbf{k}} c_{-\mathbf{k}} + c_{\mathbf{k}}^{\dagger} c_{-\mathbf{k}}^{\dagger}) + \sum_{\mathbf{k}, \mathbf{p}} \tilde{\mathcal{F}} (\mathbf{p}, -\mathbf{k} - \mathbf{p}, \mathbf{k}) (c_{\mathbf{p}} c_{-\mathbf{k} - \mathbf{p}} c_{\mathbf{k}} + c_{\mathbf{p}}^{\dagger} c_{-\mathbf{k} - \mathbf{p}}^{\dagger} c_{\mathbf{k}}^{\dagger}),
$$

(12)

where the scattering matrix element $\tilde{B}_{\mathbf{k}}$ and $\tilde{\mathcal{F}} (\mathbf{k}, -\mathbf{k} - \mathbf{p}, \mathbf{p})$ are given by

$$
\tilde{B}_{\mathbf{k}} = S \sum_{j=1}^{3} \mathcal{P}_j (\theta, \phi) [\bar{\mu}_j \bar{\nu}_j - (\bar{\mu}_j^2 + \bar{\nu}_j^2)] \lambda_j,
$$

(13)

and

$$
\tilde{\mathcal{F}} (\mathbf{k}, -\mathbf{k} - \mathbf{p}, \mathbf{p}) = \sqrt{\Sigma} \sum_{j=1}^{3} \mathcal{P}_j (\theta, \phi) \times \left[ \xi_j (\bar{\mu}_p \bar{\nu}_p + \bar{\nu}_p \bar{\mu}_p) + \zeta_j (\bar{\mu}_k \bar{\nu}_k + \bar{\nu}_k \bar{\mu}_k) \right].
$$

(14)

In the above equations we have introduced the following functions

$$
\xi_{jk} = 2[\Delta J_j + J_j \cos (\mathbf{Q} \cdot \delta_j) - D_j \sin (\mathbf{Q} \cdot \delta_j)] \cos (\mathbf{k} \cdot \delta_j) - 4[J_j \cos (\mathbf{Q} \cdot \delta_j) - D_j \sin (\mathbf{Q} \cdot \delta_j)],
$$

$$
\lambda_{jk} = [\Delta J_j - J_j \cos (\mathbf{Q} \cdot \delta_j) + D_j \sin (\mathbf{Q} \cdot \delta_j)] \cos (\mathbf{k} \cdot \delta_j),
$$

$$
\zeta_{jk} = -[J_j \sin (\mathbf{Q} \cdot \delta_j) + D_j \cos (\mathbf{Q} \cdot \delta_j)] \sin (\mathbf{k} \cdot \delta_j).
$$

(15)

For the $C_{3v}$ symmetry the $\mathcal{P}_j (\theta, \phi)$ coefficient is given by the following function

$$
\mathcal{P}_j (\theta, \phi) = \epsilon_{\text{out}} (\theta) \left( \begin{array}{c} p_1 \\ p_2 \\ p_3 \end{array} \right) \epsilon_{\text{out}}^T (\phi) \alpha_j,
$$

$$
\mathcal{P}_j (\theta, \phi) = \epsilon_{\text{out}} (\theta) \left( \begin{array}{c} p_1 \\ p_2 \\ p_3 \end{array} \right) \epsilon_{\text{out}}^T (\phi) \alpha_j
$$

(16)

with $\alpha_j^{A_1} = 1, \alpha_j^{E_1} = -2a_j^{E_1} = -2a_j^{E_2} = 1/2, \alpha_j^{E_2} = 0$ and $a_j^{E_1} = -a_j^{E_2} = \sqrt{3}/4$ [37, 38]. $\epsilon_{\text{out}}^T$ is the transpose of $\epsilon_{\text{out}}$. Within the polarization defined above, the scattering spectrum is only dependent on the $(p_1, p_2)$ coefficients. To simplify, we chose $p_1 = p_2 = 1$ in all subsequent calculations. For the $C_{2v}$
symmetry the $P_j(\theta, \phi)$ coefficient is given by

$$P_j(\theta, \phi) = e^{i\eta_j^3} \begin{pmatrix} p_5 \\ p_6 \\ p_7 \end{pmatrix} e^{i\eta_j^3} \begin{pmatrix} 0 \\ p_8 \\ p_9 \end{pmatrix} e^{i\eta_j^3},$$

with $\eta_j^3 = 1$, $\eta_j^3 = 0$, and $\eta_j^3 = -\frac{\sqrt{3}}{4}$. Within the defined polarization, the spectrum is independent of the $p_6$ coefficient. To simplify, we set $p_5 = p_7 = p_8 = 1$ in our computations.

According to the fluctuation-dissipation theorem the Raman scattering intensity can be related to the multi-magnon momentum irreducible vertex. For the Raman process, the scattering momentum follows the Feynman diagrams of the interaction, which are given by

$$\chi_2(\omega) = \int_0^\infty dt e^{i\omega t} \sum_{kk'} \bar{G}_{kk'}(T_c c_k(\tau) c_{-k}^\dagger(\tau) c_{-k'} c_{k'}),$$

$$\chi_3(\omega) = \int_0^\infty dt e^{i\omega t} \sum_{kk'} \bar{T}_{kk'} \bar{T}_{k'k'}$$

where $T_c$ is the time-ordering operator. ($\bar{\cdot}$) is the average of the ground state. Here, we study the case of zero temperature $\beta = 1/k_B T$. According to Fermi’s golden rule, the non-interacting scattering intensity is related to the bare Green’s function $G_0(k, \omega) = 1/(\omega - \omega_{k}^{(0)} + i\nu^\text{F})$ with $\omega_{k}^{(0)} = S \delta_{kk}$ in quasi-particle representation. Applying Wick’s theorem, the non-interacting spectrum can be calculated as

$$I_2(\omega) = 2 \sum_k \bar{G}_k^2 \delta(\omega - \omega_{k}^{(0)} - \omega_{-k}^{(0)}),$$

$$I_3(\omega) = 6 \sum_{kk'} \bar{T}_{kk'} \bar{T}_{k'k'} \delta(\omega - \omega_{k}^{(0)} - \omega_{-k}^{(0)} - \omega_{-k'}) = 0.$$  

The non-interacting result is shown in Fig. 4 and will be discussed in Sec. IV B along with the interacting case.

Next, we consider the $1/S$ correction to the bimagnon excitation. The two-particle propagator $\Gamma_{kk}(\omega)$ is corrected by vertex function $\Gamma_{kk}(\omega, \omega')$ following the Bethe-Salpeter equation, which is given by

$$\Pi_{kk}(\omega, \omega') = 2i \sum_{kk'} \bar{G}_{kk'}(\omega + \omega') G_{-k}(-\omega') \Gamma_{kk'}(\omega, \omega').$$

The vertex corrections follow the Feynman diagrams of the RIXS calculation [67]. They are expressed as

$$\Gamma_{kk}(\omega, \omega') = \delta_{kk} + \sum_{kk'} 2i \int \frac{d\omega_1}{2\pi} G_{kk'}(\omega + \omega_1) G_{k-k'}(-\omega_1)$$

where $V_{kk'}^{\text{IR}}(\omega', \omega_1) = V_{kk'}^3(\omega', \omega_1) + V_{kk'}^4(\omega', \omega_1)$ is the two-particle irreducible vertex. For the Raman process, the scattering momentum $q \approx 0$, thus leading to the disappearance of vertices

| $\omega/3JS$ | HH | HV |
|-------------|-----|-----|
| 0           | 0.5 | 0.5 |
| 1           | 0.5 | 0.5 |
| 2           | 0.5 | 0.5 |

FIG. 4. Non-interacting bimagnon (solid lines) and trimagnon (dashed lines) Raman spectrum of spin-1/2 system in (a) HH and (b) HV polarization. DM interaction and XXZ anisotropy are absent in the system. The red and black lines are calculated with $J' = 1$ under $C_2$ symmetry. The blue and green lines are calculated with $J' = 0.5$ under $C_2$ symmetry.

The four-point vertex $V_{kk'}^{ab}$ originating from the quartic Hamiltonian is given for our Raman case as

$$V_{kk'}^{ab} = \frac{1}{4N} \left[ 4(G_{0(k_2+q), k}^1 + iG_{0(k_2-q), k}^1) \delta_{kk'} + \sum_{kk'} G_{0(k_2+q), k}^1 \delta_{kk'} \right]$$

Next, considering the appropriate time domain of $\tau \in (-\infty, \infty)$, the expression for the interacting bimagnon Raman intensity is given by

$$I_2(\omega) = -\text{Im} \sum_{m,n} \left[ \hat{\chi}_{mn}(\omega) - \hat{\chi}_{nm}(\omega) \right].$$

In the above calculation we will assume that the two on-shell magnons are created and annihilated in the intermediate propagators with $\omega' \approx -\omega_{k}^{(0)} = -S \delta_{kk} + S \epsilon_{k}$ and $\omega_1 \approx -\omega_{k}^{(0)} = -S \delta_{kk} - S \epsilon_{k}$ [47, 67]. We treat the bimagnon susceptibility $\hat{\chi}$ through employing the discrete variables (m, n, l) to replace the continuous momenta $(k, k', k_{1})$ in Brillouin zone. Thus, we can write Eq. (21) in matrix form $\hat{\chi} = D(\hat{1} - \hat{\Gamma}^{-1}) \hat{D}$.  

### B. Bimagnon and trimagnon Raman spectrum

We calculate the spectrum under HH, VV, and HV polarization (as realized in experimental set-up), where $H$ and $V$
represent the horizontal and vertical polarization of the incoming and outgoing light, as shown in Fig. 1(b). For example, in our notation $HV$ polarization implies $\delta_{in} = H$ and $\delta_{out} = V$. The Raman signal from the isotropic TLAF can be explored by one of these three polarization choices originating from the irreducible representations $\sigma \AA + \sigma \FF \left( HH, VV \right)$ and $\sigma \FF \left( HV \right)$ modes. Note, $HH = VV$ only for the anisotropic TLAF.

The non-interacting Raman spectra for the isotropic cases are shown in Fig. 4. The bimagnon spectrum is polarization-independent since the red solid lines of Fig. 4(a) and Fig. 4(b) are the same. However, the trimagnon is polarization-dependent since the black dashed lines of Fig. 4(a) and Fig. 4(b) behave differently. The trimagnon intensity in the $HH$ polarization is much greater than that in the $HV$ polarization. For the anisotropic case, the Raman signal under $HH$ ($VV$) polarization and $HV$ polarization stems from $\eta \AA$ and $\eta \FF$ mode, respectively. As the Raman scattering operator of the bimagnon in $A_1$ mode is commutable with $H_2$, the bimagnon intensity in this channel is zero, see the blue solid line in Fig. 4(a). Different from the two-peak feature observed in the Raman response of the isotropic TLAF in Fig. 4, the bimagnon spectrum of the anisotropic case presents a single peak structure with a downshift in peak energy.

Previously we discussed the importance of magnon-magnon interaction within the context of the spectral function of a TLAF. Thus, we calculated the interacting Raman multimagnon spectrum using Eq. (25). Fig. 5 shows the interacting bimagnon intensity with different parameters under $HV$ polarization. We note that the bimagnon intensity with $HH$ ($VV$) polarization is almost zero. We study the effect of magnon-magnon interactions, spatial anisotropy, DM interaction, XXZ anisotropy, and spin value on Raman bimagnon spectrum. Considering interactions, the spectrum in the $HV$ polarization displays a single peak structure in the isotropic model compared to the two-peak structure of the non-interacting calculation. Spatial anisotropy decreases the intensity and the peak energy. DM interaction shifts the peak towards higher energy. Although XXZ anisotropy introduces a gap, the bimagnon peak has a slight downshift in energy.

As anisotropy increases, the system tends to behave like a quasi-1D spin chain. Thus, it reduces the bimagnon intensity and leads to the downshift of the peak similar to what is predicted to occur in the RIXS spectrum [47] at the roton scattering momentum $q = M$ and $q = M'$. However, unlike the RIXS spectrum, the DM interaction causes a decrease in the Raman intensity, compare Fig. 5 to Fig. 5(b). The reduction effect is also seen in the XXZ model if we compare Fig. 5(a) to Fig. 5(c). This reduction can be attributed to the influence of DM interaction and XXZ anisotropy on the bimagnon scattering matrix element. With increasing spatial anisotropy, DM interaction plays a more important role. Thus, the Raman intensity reduction is more prominent in the systems with greater spatial anisotropy. The renormalized dispersion is shown in Fig. 3. DM interaction introduces a spin gap, resulting in the higher peak energy in Fig. 5(b) compared to Fig. 5(a). Although a tiny gap is generated by XXZ anisotropy, the bimagnon peak shifts to lower energy slightly in Fig. 5(c) compared to Fig. 5(a). In addition, the large spin value weakens the quantum fluctuations and magnon-magnon interactions. Thus, compared to spin-1/2 systems, a spin-3/2 material will cause an energy upshift with vanishing shoulder, similar to the non-interacting case.

We study the Raman spectrum of two real materials Ba$_3$CoSb$_2$O$_9$ (isotropic TLAF) and Cs$_3$CuCl$_4$ (anisotropic TLAF). The Raman spectrum of Ba$_3$CoSb$_2$O$_9$ in $C_{3v}$ symmetry is shown in Fig. 6(a) and Fig. 6(b). For Ba$_3$CoSb$_2$O$_9$, with DM interaction set to zero and $J = J'$ the ground state is close to a 120$^\circ$ non-collinear magnetic order. We choose our calculation parameters based on the experiment of Susuki et al. Ref. 60. The Raman spectrum of Cs$_3$CuCl$_4$ in $C_2$, symmetry is shown in Fig. 6(c) and 6(d). The fit parameters for Cs$_3$CuCl$_4$ were chosen from our earlier TESWT INS fitting result [47].

The Ba$_3$CoSb$_2$O$_9$ Raman spectrum shows two prominent features for both the $HH$ and $HV$ polarization. In the $HH$ polarization there is a clear separation of energy excitation. The bimagnon peaks around $\approx 1.1J$, whereas the trimagnon peaks around $\approx 3.3J$. However, the bimagnon intensity is much greater than the trimagnon response in the $HH$ polarization geometry. For the $HV$ case, the Raman intensity is dominated by the bimagnon signal. For Cs$_3$CuCl$_4$ the bimagnon signal is almost zero in the $HH$ channel. Thus, the trimagnon is the main contribution. This response is opposite to that observed in Ba$_3$CoSb$_2$O$_9$. Hence, we can use the $HH$ signal as a signature to identify anisotropic behavior in a TLAF. The trimagnon signal is broad and spreads over an energy range of $\approx J - 3J$. In contrast, the $HV$ polarization for Cs$_3$CuCl$_4$ supports a non-zero signal for both the bi- and the trimagnon intensity. The bimagnon peaks at $\approx 0.6J$ and the trimagnon is maximum around $\approx 1.2J$. This behaviour is qualitatively similar to what we observe for Ba$_3$CoSb$_2$O$_9$ Raman signal in the $HH$ geometry. The bimagnon intensity for Ba$_3$CoSb$_2$O$_9$ is greater than Cs$_3$CuCl$_4$. This can be explained by the presence of stronger spin coupling along the
diagonal bonds for the isotropic TLAF. Furthermore, comparing Fig. 6(b) to Fig. 6(d) we find that the trimagnon response survives only in the anisotropic case, consistent with Fig. 4(b). Thus, we can also judge the degree of anisotropy of the system from the HV polarized Raman spectrum.

V. CONCLUSION

We applied TESWT to calculate the bi- and the trimagnon Raman spectrum of an isotropic and an anisotropic TLAF. We extended TESWT to the XXZ model considering both spatial anisotropy and DM interaction. Our calculation is a demonstration of the validity to apply the TESWT formalism to Raman spectroscopy analysis. We computed the TESWT corrected ordering wave vector, the on-shell dispersion, the spectral function, and the Raman spectrum in both $C_{3v}$ and $C_{2v}$ symmetry for HH and HV polarization.

Based on our calculations we find that even for our system the spin Casimir effect can induce an arrangement of collinear spins [13]. Although DM interaction and XXZ anisotropy stabilize the spiral order, spin wave theory is unable to predict an accurate ordering wave vector for the system. Thus, one needs to account for the presence of spin Casimir torque introduced by zero-point quantum fluctuations. This reduces the range of the spiral phase. Both XXZ anisotropy and DM interaction introduces a gap at the ordering wave vector, leading to suppression of damping and magnon excitation stabilization. We note that DM interaction brings about a stronger suppression effect on magnon decay than XXZ anisotropy. We also discuss the sensitivity of Raman spectrum to polarization, system parameters ($J'$, $D$, $\Delta$, $S$), and magnon-magnon interactions. We find that large spin values cause an energy upshift with vanishing shoulder due to the weakened quantum fluctuations and magnon-magnon interactions. We also compute the bi- and the trimagnon Raman spectrum for $\text{Ba}_3\text{CoSb}_2\text{O}_8$ and $\text{Cs}_2\text{CuCl}_4$. In isotropic TLAF, the Raman spectrum is sensitive to polarization for trimagnon excitation and independent from polarization for bimagnon excitation. However, the converse holds true for the anisotropic lattice. The bimagnon excitation is polarization dependent. We propose that the degree of anisotropy of the system can be judged using either the HH or the HV polarization. Thus, we show that TESWT is a reliable method to calculate the Raman spectrum of frustrated magnetic materials and to analyze the underlying spectrum.

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Appendix A: $1/S$ spin wave THEORY

In this Appendix we state the introduced functions under $1/S$-LSWT, which can be easily applied under the TESWT. The classical energy $E_0(\mathbf{Q})$ is given by

$$E_0(\mathbf{Q}) = NS^2(J + \eta) = NS^2\gamma$$  \hspace{2cm} (A1)

with

$$J_k = J_0 \cos k_x + 2J' \cos k_x \cos k_y \cos \frac{\sqrt{3}}{2},$$

$$\eta = 2D \sin \frac{k_x}{2} \cos \frac{\sqrt{3}}{2} k_y. \hspace{2cm} (A2)$$

The classical ordering vector $Q_{cl}$ is obtained by solving the following self-consistent equation

$$\nabla \mathbf{Q} E_0(\mathbf{Q}) = 0. \hspace{2cm} (A3)$$

The bare magnon dispersion is $S \epsilon_k$ with

$$\epsilon_k = \sqrt{A_k^2 - B_k^2}, \hspace{2cm} (A4)$$

where

$$A_k = \frac{1}{2}(2\Delta J_k + \gamma \eta_k + \gamma \eta_k - 4\gamma),$$

$$B_k = \frac{1}{2}(\gamma \eta_k + \gamma \eta_k - 2\Delta J_k). \hspace{2cm} (A5)$$

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig6.png}
\caption{Interacting bimagnon and non-interacting trimagnon Raman spectra of $\text{Ba}_3\text{CoSb}_2\text{O}_8$ and $\text{Cs}_2\text{CuCl}_4$. The left (right) column is under HH (HV) polarization. The first line is for $\text{Ba}_3\text{CoSb}_2\text{O}_8$ with $(J', D, \Delta) = (1, 0, 0.954)$. The second line is for $\text{Cs}_2\text{CuCl}_4$ with $(J', D, \Delta) = (0.316, 0.025, 1)$.}
\end{figure}
The rest of the quadratic terms in $\mathcal{H}_{\text{eff}}$ is obtained from a mean-field decoupling of the quartic Hamiltonian with

$$
\delta \epsilon_k = (u_k^2 + v_k^2) \delta A_k + 2u_k v_k \delta B_k,
$$

$$
O_k = (u_k^2 + v_k^2) \delta B_k + 2u_k v_k \delta A_k,
$$

where $u_k$ and $v_k$ are the Bogoliubov transformation coefficients given by

$$
u_k = -\text{sgn}(B_k) \sqrt{\frac{A_k}{2} - \frac{1}{2}},$$

and

$$
\delta A_k = \frac{A_k}{2} + \frac{1}{2N} \sum_p \frac{1}{e_p A_p} \left[ A_p (A_{k-p} + B_{k-p} - A_k - A_p) + B_p \left( \frac{B_k}{2} + B_p \right) \right],
$$

$$
\delta B_k = \frac{B_k}{2} - \frac{1}{2N} \sum_p \frac{1}{e_p B_p} \left[ B_p (A_{k-p} + B_{k-p} - A_k - A_p) + A_p \left( \frac{A_k}{2} + \frac{B_k}{2} \right) \right].
$$

The cubic interaction terms are defined as

$$
\Phi_{(1, 2; 3, 4)} = \left[ \tilde{\gamma}_1 (u_1 + v_1) (u_2 u_3 + v_2 v_3) + \tilde{\gamma}_2 (u_2 + v_2) (u_1 u_3 + v_1 v_3) + \tilde{\gamma}_3 (u_1 + v_1) (u_2 v_3 + v_2 u_3) + \tilde{\gamma}_4 (u_1 v_2 + v_1 u_2) (u_3 + v_3) \right].
$$

The quartic interaction term is given by

$$
(1, 2, 3) = \left[ \tilde{\gamma}_1 (u_1 + v_1) (u_2 u_3 + v_2 v_3) + \tilde{\gamma}_2 (u_2 + v_2) (u_1 u_3 + v_1 v_3) + \tilde{\gamma}_3 (u_1 + v_1) (u_2 v_3 + v_2 u_3) + \tilde{\gamma}_4 (u_1 v_2 + v_1 u_2) (u_3 + v_3) \right].
$$

where $C_k$ is

$$
C_k = A_k + B_k.
$$

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[66] Note, in our earlier publication Jin et al. [47], there was a typographical error in the reported RIXS scattering operator expression. The expression was missing the DM interaction term which we considered in our study. The correct reported form of the RIXS operator expression should have been \( \mathcal{R}_\delta = \sum_{i,a} e^{i\delta} [J_{a} S_i \cdot S_{i+a} - D_{a} (S_i \times S_{i+a})] \).

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