Entangled tetrahedron ground state and excitations of the magneto-electric skyrmion material Cu$_2$OSeO$_3$

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The strongly correlated cuprate Cu$_2$OSeO$_3$ has recently been identified as the first insulating system exhibiting a skyrmion lattice phase. Using a microscopic multi-boson theory for its magnetic ground state and excitations, we establish the presence of two distinct types of modes: a low energy manifold that includes a gapless Goldstone mode and a set of weakly dispersive high-energy magnons. These spectral features are the most direct signatures of the fact that the essential magnetic building blocks of Cu$_2$OSeO$_3$ are not individual Cu spins, but rather weakly-coupled Cu$_4$ tetrahedra. Several of the calculated excitation energies are in nearly perfect agreement with reported Raman and far-infrared absorption data, while the magneto-electric effect determined within the present quantum-mechanical framework is also fully consistent with experiments, giving strong evidence in the entangled Cu$_4$ tetrahedra picture of Cu$_2$OSeO$_3$. The predicted dispersions along with the dynamical dipole and quadrupole spin structure factors call for further experimental tests of this picture.

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Introduction — The experimental discovery of skyrmions in the chiral metallic helimagnets MnSi [1, 2], Fe$_1$–xCu$_2$Si [3, 4] and FeGe [5], almost 20 years after their theoretical prediction [6, 7], has prompted an enormous interest in the community, both on the theory and the experiment side. Skyrmions are localized magnetization textures with non-trivial topology which, under certain conditions [6–8], may condense into a lattice, in analogy to the Abrikosov vortices in type-II superconductors [9], and the so-called “blue phases” in chiral liquid crystals [10]. The recent discovery of skyrmions [11] in Cu$_2$OSeO$_3$ has opened an alternative route for realizing this kind of physics, since this material is an insulating oxide with localized spins-1/2 [12]. Moreover Cu$_2$OSeO$_3$ shows a linear magneto-electric (ME) effect [11, 13, 14] which in principle allows the skyrmions to be manipulated by an external electric field without losses due to joule heating [15–17].

A microscopic understanding of this rich set of physical phenomena in Mott insulating Cu$_2$OSeO$_3$ starts from a description of its magnetic interactions. Close inspection of the superexchange interactions between localized Cu$^{2+}$ spins, as calculated from atomistic ab initio methods [18, 19], reveals the presence of weaker and stronger magnetic bonds. Due to this separation of energy scales, the degrees of freedom that order below T$_C$ ≃ 60 K and twist in the helical or the skyrmion lattice phase are not the bare Cu$^{2+}$ spins but instead tetrahedral entities (see Fig. 1(b)), which are, nevertheless, also deeply quantum-mechanical (QM) in nature [19].

Here we show that the notion of weakly-interacting Cu$_4$ tetrahedra gloriously survives a full quantitative QM treatment of the problem and has profound implications for the magnetic excitation spectrum as well as for the ME properties of Cu$_2$OSeO$_3$. Using a multi-boson approach that incorporates the strong tetrahedral entities from the outset—in this way capturing the dominant portion of the QM correlations—we establish the presence of two distinct types of excitations: (i) a 4-branch low-energy manifold that includes a gapless Goldstone mode corresponding to long-wavelength modulations of the local order parameter, and (ii) a higher-energy set of more weakly dispersive intra-tetrahedron excitations. As we show below, a number of excitation energies (at the $\Gamma$ point) are in nearly perfect agreement with reported Raman [20] and far-infrared [21] absorption data, lending strong support to the entangled tetrahedra picture of this compound. Given that this picture can be also tested by other spectroscopic techniques, such as inelastic neutron scattering, we explicitly calculate the dipolar and quadrupolar dynamical structure factors for the entire spectrum. We also explore the ramifications of this framework for the ME coupling by calculating the induced electric polarization $\mathbf{P}$ in different crystallographic directions as a function of the orientation of an applied magnetic field $\mathbf{H}$. Recent experimental observations [11, 13, 14] are fully consistent with the calculated direction of $\mathbf{P}$, again supporting the fundamental notion that the essential magnetic building blocks of Cu$_2$OSeO$_3$ are not individual Cu spins, but entangled Cu$_4$ tetrahedra.

Magnetic ground state — Cu$_2$OSeO$_3$ has the non-centrosymmetric P2$_1$3 space group, similar to the metallic B2O helimagnets. There are two symmetry inequivalent Cu$^{2+}$ sites, Cu$_1$ and Cu$_2$, residing at the Wyckoff positions 4a and 12b, respectively [12, 20, 23]. These sites form a distorted, 3D pyrochlore lattice of corner sharing tetrahedra, consisting of one Cu$_1$ and three Cu$_2$ ions...
FIG. 1. (color online) (a) Distorted pyrochlore structure of Cu$_2$OSeO$_3$. Solid (dashed) lines indicate the strong (weak) exchange couplings. For clarity, the longer-range $J'_{O,O}$ coupling (see text) is not shown here. (b) The energy spectrum of an isolated strong tetrahedron [22].

each, see Fig. 1(a). For present purposes, we consider only the Heisenberg-type exchange interactions described by the Hamiltonian $H_{heis} = \sum_{(ij)} J_{ij} S_i \cdot S_j$ (where $J_{ij}$ are the exchange couplings between sites $i$ and $j$). The much weaker Dzyaloshinskii-Moriya (DM) interactions [18, 19] affect only the very low frequency portion of the excitation spectrum close to the $\Gamma$ point. Four of the five most relevant exchange paths [19] are indicated in Fig. 1: $J^AF_s = 170$K, $J^AF_m = -128$K (within strong tetrahedra, solid lines) and $J^AF_w = 27$K and $J^FM_w = -50$K (connecting strong tetrahedra, dashed lines). The fifth coupling is a longer-range exchange, $J^FM_{O,O} = 45$K, connecting Cu$_1$ with Cu$_2$ sites across the diagonals of alternating Cu$_1$–Cu$_2$ hexagon loops [19].

The presence of stronger ($J^AF_s$ and $J^AF_m$) and weaker couplings ($J^AF_w$, $J^FM_w$, and $J^FM_{O,O}$) suggests that we take as a starting point a tetrahedron–factorized wave function $|\Psi\rangle = \prod_n |\psi\rangle_t$, where $|\psi\rangle_t$ is a QM state living in the 16-dimensional Hilbert space of the strong tetrahedron $t$. Doing so is equivalent to solving the single tetrahedron mean field (TMF) Hamiltonian [19] $H^{(t)}_{TMF} = H^{(t)}_0 + V^{(t)}_{MF}$, where $H^{(t)}_0$ contains the intra-tetrahedra couplings, and $V^{(t)}_{MF}$ the exchange fields exerted from $t' \neq t$ at a mean field level. The ensuing energy eigenstates of $H_0$ are shown in Fig. 1(b) above, and can be labeled by the total spin $S$, its projection $S_z$, and the irreducible representations $\lambda$ of the point group $C_{3v}$. The ground state is an $A_1$–triplet with a large excitation gap of $\sim 275$ K. A finite $V_{MF}$ mixes states with different $S$, so that it is no longer a good quantum number. The point group symmetry remains $C_{3v}$, however, thus the $A_1$–triplet can only admix with the $A_1$–quintet. So for an infinitesimal symmetry breaking magnetic field along $z$, the ground state of $H^{(t)}_{TMF}$ reads

$$|\psi\rangle_t = \cos \frac{\alpha}{2} |1, 1, A_1\rangle_t + \sin \frac{\alpha}{2} |2, 1, A_1\rangle_t,$$

where the variational parameter $\alpha$ controls the degree of spin mixing and the local moments, since

$$\langle S^z_1 \rangle = -\frac{1}{4} (\cos \alpha + \sqrt{3} \sin \alpha), \quad \langle S^z_2 \rangle = \frac{1}{3} (1 - \langle S^z_1 \rangle).$$

Incidentally, the total moment per tetrahedron is $\langle S^z \rangle = 1$ regardless of $\alpha$, which corresponds to a 1/2 magnetization plateau. For $\alpha = 0$, $\langle S^z \rangle = -\frac{1}{4}$ and $\langle S^z \rangle = \frac{5}{12}$, while in the coupled limit the minimization of $\langle \psi | H^{(t)}_{TMF} | \psi \rangle_t$ yields $\alpha = 0.337205$, $\langle S^z_1 \rangle = -0.37(9)$ and $\langle S^z_2 \rangle = 0.45(9)$. The reduced values of the spin lengths compared to the classical $S^z_1 = \frac{5}{2}$ reflect the fact that $|\psi\rangle$ is highly entangled and cannot be decomposed into a site-factorized form. Such a local moment reduction has indeed been observed experimentally [12].

**Magnetic fluctuations and excitations**—Owing to the QM nature of $|\Psi\rangle$, including the fluctuating part $H = \sum_t H^{(t)}_{TMF}$ requires a multi-boson generalization [24–27] of the standard spin-wave expansion [28–30]. Such multi-boson theories have previously been employed successfully in quantum spin models with dimerized [31], quadrupolar [32, 33], or nematic phases [34, 35].

We first introduce bosonic operators $a_{n,\nu}(R)$, with $n = 1-16$ and $\nu = 1-4$, such that $a_{n,\nu}(R)|0\rangle$, where $|0\rangle$ is the vacuum, gives the $n$-th eigenstate of $H^{(t)}_{TMF}$ at the $\nu$-th tetrahedron inside the unit cell $R$. The wavefunction $|\Psi\rangle$ can be thought of as a coherent state $\prod_n R(a_{n,\nu}(R)) M |0\rangle$ as $M \to \infty$. The remaining 15 bosons per tetrahedron play the role of generalized “tetrahedral spin waves”. Now, given that each tetrahedron can only be in one of the 16 states $|n\rangle$, the bosons must satisfy the hard-core constraint $\sum_n a_{n,\nu}^\dagger(R)a_{n,\nu}(R) = 1$. Following the standard approach [28], to treat this constraint we allow for $M$ bosons per tetrahedron instead of 1, and replace the condensed bosons with

$$a_{1,\nu} = a_{1,\nu}^\dagger = \sqrt{M - \sum_{n=1}^M a_{n,\nu}a_{n,\nu}},$$

and then perform a $1/M$ expansion up to quadratic terms. In the bosonic Hamiltonian, the zeroth-order scalar corresponds to the mean-field energy, the first-order terms vanish in the ground state, while the second-order terms provide the quadratic spectrum by a standard Bogoliubov diagonalization [22].

This calculation provides direct access to the residual spin length corrections, which are an important diagnostic for the reliability of the multi-boson approach. It quantifies the strength of residual quantum fluctuations, which should be small for our theoretical approach to be consistent. We find (see SM for details) $\delta S_1 = 0.011129$ and $\delta S_2 = -0.003709$, which are very small indeed. This
implies that most of the quantum correlations have already been captured by $|\Psi\rangle$ and that tetrahedral spin-wave fluctuations have a negligible influence on the spin lengths: our multi-boson expansion is thus based on very firm ground.

The resulting magnetic excitation spectrum is shown in Figs. 2(c-d). Most of the features can be understood by comparing with the energy levels of $H_{\text{TMF}}$ for $\alpha=0.337205$, show in Fig. 2(b). The lowest excitations of $H_{\text{TMF}}$ from $n=1\rightarrow n'=2$ give rise to four modes (counting the number of tetrahedra per unit cell) with $\delta M=1$. One of them gives the quadratic Goldstone mode (associated with the breaking of SU(2) symmetry down to U(1)), and the remaining three modes are at $\sim 130$ K at the $\Gamma$ point. The excitation to $n'=3$ gives another four modes with $\delta M=2$, which sit at $\sim 225$ K and are dispersionless. Excitations to $n'>4$ give rise to higher energy modes above $\sim 320$ K which are all weakly dispersive. On the basis of this excitation spectrum one can directly conclude that weakly-coupled magnetic tetrahedra are the essential magnetic building blocks of Cu$_2$OSeO$_3$.

Comparison to Raman and far-infrared experiments — Gnezdilov et al.[20] have reported a very large number of Raman absorption lines in the energy scale of interest, at least five of which are most likely of magnetic origin, given that they appear below $T_C$ and the fact that the magnetic transition is not accompanied by any structural distortion. For comparison, these lines are shown by square symbols on top of the calculated spectrum in Fig. 2(c) and are labeled by A (86 cm$^{-1}$), B (203 cm$^{-1}$), C (261 cm$^{-1}$), D (270 cm$^{-1}$), and E (420 cm$^{-1}$). The lines A, B and D have also been observed in far-infrared absorption spectra reported by Miller et al.[21]. Except for line B, all other lines are very close to one of the calculated absorptions. The agreement, especially for line A is striking, showing that this line corresponds to the three low-lying modes at $\sim 130$ K discussed above. Furthermore, the fact that the energy of these modes depends essentially only on the weak inter-tetrahedra exchange couplings gives strong confidence in the accuracy of $J_{w}^{AF}$, $J_{w}^{FM}$ and $J_{O}^{AF}$. Turning to line B, if this mode is magnetic then its deviation from the three modes predicted around 340 K would suggest that the strong intratetrahedra couplings $J_{x}^{FM}$ and $J_{x}^{AF}$ are overestimated in [19], which is plausible given that these energy scales do not affect much the T-dependence of the susceptibility and the value of $T_C$ [19]. While further experiments are needed to clarify this point, the nearly perfect agreement of the remaining modes (especially A) lends strong support to the entangled tetrahedra picture of Cu$_2$OSeO$_3$.

Dynamical structure factors — To give predictions for further experiments, we calculate the dipole and quadrupole matrix elements between the magnetic ground state $|i\rangle$ and excited states $|f\rangle$. In terms of the
magnetic dipole operators $S_i^\alpha$, the quadrupolar operators, defined on bonds (ij), are $Q_{ij}^{\alpha\beta} = S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha$. For simplicity we restrict ourselves here to the ones on the strong bonds:

$$Q_{s,AF}^{\alpha\beta}(\mathbf{R}) = S_{1,1}^\alpha S_{1,1}^\beta + S_{1,1}^\beta S_{1,1}^\alpha + S_{1,3}^\alpha S_{1,3}^\beta + S_{1,3}^\beta S_{1,3}^\alpha,$$

$$Q_{s,FM}^{\alpha\beta}(\mathbf{R}) = S_{2,2}^\alpha S_{2,2}^\beta + S_{2,2}^\beta S_{2,2}^\alpha + S_{2,4}^\alpha S_{2,4}^\beta + S_{2,4}^\beta S_{2,4}^\alpha,$$

where $t$ is any of the four tetrahedra inside the unit cell $\mathbf{R}$.

In Fig. 2 the magnetic dipole transition matrix elements (Fig. 2(c)) and the quadrupole ones $Q_{s,AF}^{\alpha\beta} = Q_{s,AF}^{\alpha\beta} + Q_{s,FM}^{\alpha\beta}$ (Fig. 2(d)) are indicated by the width of the lines in the spectrum. Dashed lines denote excitations where the matrix element vanishes exactly due to selection rules, as e.g. in the dipole case for transitions with $\delta M=2$ (such transitions are instead allowed in $Q^{xx-uv}$ and $Q^{xy}$). The dipole transitions are particularly strong for the lowest four branches, whereas the quadrupolar ones have appreciable weights at both low and high energy modes.

The dipole matrix elements are directly relevant for inelastic neutron scattering (INS) experiments, since the INS intensity is proportional to the dynamical spin structure factor $S^{\mu\nu}(\mathbf{k},\omega) = \sum f |i(S_{\mathbf{k}}^{\mu}|f)(S_{\mathbf{k}}^{\nu}|i)|^2 \delta(\omega - \omega_{f})$. In addition, these results are relevant in light scattering experiments, since the response to the electric and magnetic components of the incoming light is given by the magnetic and dielectric susceptibilities, $\chi_{\alpha\beta}(\omega) = \sum f |i(S_{\mathbf{k}}^{\alpha}|f)(S_{\mathbf{k}}^{i\beta}|i)|^2 \delta(\omega - \omega_{f})$ and $\chi_{\alpha\beta}(\omega) = \sum f |i(P_{\mathbf{k}}^{\alpha}|f)(P_{\mathbf{k}}^{i\beta}|i)|^2 \delta(\omega - \omega_{f})$, where $\mathbf{P}$ is the electric polarization. The latter is related to quadrupolar spin transition operators through the ME coupling [13] (see also below).

**Magneto-electric effect** — As the quadratic corrections to the state $|\Psi\rangle$ are extremely small, $|\Psi\rangle$ provides accurate information about other GS expectation values, such as for instance those of quadrupole spin operators. The coarse-grained electric polarization $\mathbf{P}$ [13], in particular, can be expressed as a sum over quadrupolar spin contributions from symmetry inequivalent bonds [36]:

$$P^{\alpha}(\mathbf{R}) = \xi_{s,AF}(Q^{\beta\gamma}_{s,AF}(\mathbf{R})) + \xi_{s,FM}(Q^{\beta\gamma}_{s,FM}(\mathbf{R})) + \ldots$$

where $\chi$ is the dielectric susceptibility, $\xi_{s,AF}$, $\xi_{s,FM}$ ME coupling constants, and $Q_{s,AF}$, $Q_{s,FM}$ are defined in Eq. (4) above. (The explicit form of these operators is given in the SM). For a magnetic field pointing along $\Omega = (\sin \vartheta \cos \varphi, \sin \vartheta \sin \varphi, \cos \vartheta)$, we find

$$\mathbf{P}/\chi = P(\sin \varphi \sin 2\vartheta, \cos \varphi \sin 2\vartheta, \sin 2\varphi \sin 2\vartheta)$$

where $P = \xi_{s,AF}Q_{s,AF} + \xi_{s,FM}Q_{s,FM} + \ldots$, and

$$Q_{s,AF} = \frac{1}{8} (-2 + \cos \alpha - 3\sqrt{3} \sin \alpha) = -0.34,$$

$$Q_{s,FM} = \frac{3}{8} (\cos \alpha + \sqrt{3} \sin \alpha) = 0.56,$$

$$Q_{w,AF} = Q_{w,FM} = (S_{1}^z)^2 = 0.17,$$

$$Q_{w,FM} = (S_{2}^z)^2 = 0.21.$$
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[36] Note that the phase space of isolated spins-1/2 leaves no room for quadrupolar degrees of freedom, which is why the minimal ME coupling discussed here involves more than one Cu spins.
SUPPLEMENTARY MATERIAL

In this supplement we introduce the local tetrahedron–basis used in our calculation and give details on the multi-boson spin-wave theory.

Tetrahedron basis

Table I provides the symmetry-classified eigenbasis of a tetrahedron in the decoupled limit.

| irrep | state | notation | $a_i$ boson | excitation energy |
|-------|-------|----------|-------------|------------------|
| $A_1 \otimes D^{(1)}$ | $\frac{1}{\sqrt{6}}((\downarrow\downarrow\uparrow) + |\downarrow\downarrow\downarrow) + |\downarrow\downarrow\downarrow - 3|\uparrow\uparrow\downarrow)$ | $|1, \Omega, A_1)$ | $a_{1,1}^{\Omega, A_1}$ | $\omega_{S=1}^{A_1} = 0$ |
| $E \otimes D^{(0)}$ | $\frac{1}{2\sqrt{3}(2|\downarrow\downarrow\uparrow) - |\downarrow\downarrow\downarrow - |\uparrow\uparrow\uparrow - 2|\uparrow\uparrow\downarrow) + (\uparrow\uparrow\uparrow) + (\uparrow\uparrow\downarrow) + (\uparrow\uparrow\downarrow)$ | $|0, 0, E_0)$ | $a_{1,0,0, E_0}$ | $\omega_{S=0}^{E_0} = \frac{J_{AF} - J_{FM}}{2}$ |
| $A_1 \otimes D^{(2)}$ | $\frac{1}{\sqrt{6}}((\downarrow\downarrow\uparrow) + |\downarrow\downarrow\downarrow) + |\downarrow\downarrow\downarrow - 3|\uparrow\uparrow\downarrow)$ | $|1, \Omega, E_0)$ | $a_{1,1}^{\Omega, E_0}$ | $\omega_{S=2}^{E_0} = 2J_{AF}$ |
| $E \otimes D^{(1)}$ | $\frac{1}{2\sqrt{6}}((-2|\downarrow\downarrow\uparrow) + |\downarrow\downarrow\downarrow) + |\downarrow\downarrow\downarrow - 2|\uparrow\uparrow\downarrow) + (\uparrow\uparrow\uparrow) + (\uparrow\uparrow\downarrow) + (\uparrow\uparrow\downarrow)$ | $|0, 0, E_1)$ | $a_{1,0,0, E_1}$ | $\omega_{S=1}^{E_1} = \frac{3(J_{AF} - J_{FM})}{2}$ |
| $A_1 \otimes D^{(1)}$ | $\frac{1}{\sqrt{6}}((\downarrow\downarrow\uparrow) + |\downarrow\downarrow\downarrow) + |\downarrow\downarrow\downarrow - 3|\downarrow\downarrow\downarrow)$ | $|1, \Omega, E_1)$ | $a_{1,1}^{\Omega, E_1}$ | $\omega_{S=2}^{E_1} = 2J_{AF}$ |

Spin polarized ground state

Here we introduce the general form of the SU(2) symmetry breaking ground state using spin polarized states. The ground state of a single tetrahedron is the 3-fold degenerate $A_1$–triplet $|1, S^z \rangle_{A_1}$, where $S^z = -1, 0, 1$. The corresponding wavefunction for this triplet pointing in the direction $\Omega = (\cos \varphi \sin \theta, \sin \varphi \sin \theta, \cos \theta)$ reads

$$|1, \Omega \rangle_{A_1} = \cos \frac{\theta}{2} |1, 1 \rangle_{A_1} + e^{i\varphi} \sin \theta \frac{1}{\sqrt{2}} |1, 0 \rangle_{A_1} + e^{2i\varphi} \sin^2 \frac{\theta}{2} |1, \Omega \rangle_{A_1}.$$  

Similarly, for the $S = 2$ excitation of the decoupled limit:

$$|2, \Omega \rangle_{A_1} = -e^{-i\varphi} \cos \frac{\theta}{2} \sin \theta |2, 2 \rangle_{A_1} + (\cos^2 \theta - \sin^2 \frac{\theta}{2}) |2, 1 \rangle_{A_1} + \frac{1}{2} \sqrt{3} e^{i\varphi} \sin 2\theta |2, 0 \rangle_{A_1}$$

$$- e^{2i\varphi} (\cos^2 \theta - \cos^2 \frac{\theta}{2}) |2, J \rangle_{A_1} + e^{3i\varphi} \sin \frac{\theta}{2} \sin \theta |2, \Omega \rangle_{A_1}.$$  

The inter-tetrahedron interactions admix $|1, \Omega \rangle_{A_1}$ with $|2, \Omega \rangle_{A_1}$, and so the mean-field ground state reads

$$|\Omega \rangle_{A_1} = \cos \frac{\alpha}{2} |1, \Omega \rangle_{A_1} + \sin \frac{\alpha}{2} |2, \Omega \rangle_{A_1}.$$  


The spin expectation value of the Cu1 site for Eq. 13 is \( \langle \Omega | Q \Omega^\dagger | \Omega \rangle = S_1 \Omega \), with \( S_1 = -\frac{1}{\sqrt{3}}(\cos \alpha + \sqrt{3} \sin \alpha) \). The dipole expectation value of the Cu2 sites in the ground state has the same form, with \( S_{2,3,4} = \frac{1}{\sqrt{3}}(4 \cos \alpha + \sqrt{3} \sin \alpha) \). In the decoupled limit \( \alpha \rightarrow 0 \), \( S_1 \rightarrow -\frac{1}{\sqrt{3}} \) and \( S_{2,3,4} \rightarrow \frac{5}{\sqrt{3}} \). Note, that the ground state of Eq. 13 corresponds to a 1/2 magnetization plateau, regardless of \( \Omega \) and \( \alpha \), since the total magnetization of a tetrahedron is \( \sum_{i=1-4} S_i = 1 \), yielding \( M/M_{\text{sat}} = 1/2 \).

The ground state expectation value of the quadrupole operators defined for a bond of two copper ions can be expressed as

\[
\langle \Omega | Q^m | \Omega \rangle = Q \sin \varphi \sin 2\vartheta \\
\langle \Omega | Q^{z^2} | \Omega \rangle = Q \cos \varphi \sin 2\vartheta \\
\langle \Omega | Q^{z^2-y^2} | \Omega \rangle = Q \cos 2\varphi \sin^2 \vartheta \\
\langle \Omega | Q^{z^2-y^2} | \Omega \rangle = Q(\cos 2\varphi \sin^2 \vartheta + \cos 2\vartheta) ,
\]

where \( Q = \frac{1}{8}(-2 + \cos \alpha - 3\sqrt{3} \sin \alpha) \) on the antiferromagnetic, and \( Q = \frac{3}{8}(\cos \alpha + \sqrt{3} \sin \alpha) \) on the ferromagnetic bonds.

**Bosonic representation**

As described in the main text, our bosonic operators create the \( C_{3v} \otimes SU(2) \) eigenbasis of Table 1 so that \( |S,m\rangle_R = a_{S,m,R}^\dagger |0\rangle \) and \( |0\rangle \) is the vacuum. The form of the spin operators \( S^a \) are too excessive to show here, nonetheless the intra-tetrahedron Hamiltonian and the total magnetization \( S^z = \sum_{i=1-4} S_i^z \) are diagonal in this basis:

\[
\mathcal{H}_0 = -\frac{5 J_s^{AF} + 3 J_s^{FM}}{4} \sum_{m=\{1,0,1\}} a_{1,m,Ai}^\dagger a_{1,m,Ai} - \frac{3(J_s^{AF} + J_s^{FM})}{4} \sum_{E=\{E_1,E_2\}} a_{0,0,E}^\dagger a_{0,0,E} + \frac{3(J_s^{AF} + J_s^{FM})}{4} \sum_{m=\{1,0,1\}} a_{1,m,E}^\dagger a_{1,m,E} + \frac{3(J_s^{AF} - 3 J_s^{FM})}{4} \sum_{m=\{2,...,2\}} a_{1,m,E}^\dagger a_{1,m,E} \ ,
\]

\[
S^z = \sum_m a_{S,m,R}^\dagger a_{S,m,R} , \quad (19)
\]

The spin raising operator of a tetrahedron can be written as

\[
S^+ = \sum \sqrt{S(S+1) - m(m+1)} a_{S,m+1,R}^\dagger a_{S,m,R} , \quad (20)
\]

where the sum is over the 16 dimensional tetrahedron basis.

**Multi-boson theory and matrix elements**

In the multi-boson theory, solving the constraint \( \sum_{\{S,m,R\}} a_{S,m,R}^\dagger a_{S,m,R} = M \) for the \( a_{1,1,Ai}^\dagger \) and \( a_{1,1,Ai} \), we perform the replacement of Eq. (3) of main text. The intra-tetrahedron Hamiltonian then becomes

\[
\mathcal{H}_0 = -\frac{5 J_s^{AF} + 3 J_s^{FM}}{4} M + \frac{J_s^{AF} - 3 J_s^{FM}}{2} \sum_{E=\{E_1,E_2\}} a_{0,0,E}^\dagger a_{0,0,E} + 2 J_s^{AF} \sum_{m=\{2,...,2\}} a_{1,m,E}^\dagger a_{1,m,E} + \frac{3(J_s^{AF} - J_s^{FM})}{2} \sum_{m=\{1,0,1\}} a_{1,m,E}^\dagger a_{1,m,E} , \quad (21)
\]

Resetting \( M \) to 1, we obtain the dispersionless uncoupled spectrum, where the ground state is \( -5 J_s^{AF} - 3 J_s^{FM} \) (note that there are four tetrahedra in the unit cell) and the bosons in Eq. (21) play the role of the excitations. For each tetrahedron there are two modes at \( \frac{J_s^{AF} - 3 J_s^{FM}}{2} \), five at \( 2 J_s^{AF} \) and six at the energy \( \frac{3(J_s^{AF} - J_s^{FM})}{2} \). Furthermore, there are two zero-energy bosons \( a_{1,0,Ai}^\dagger \) and \( a_{1,1,Ai}^\dagger \) per tetrahedron.
The matrix elements $\langle 0 | S^2 | f \rangle$, related to the intensity of the inelastic neutron spectrum, are easily obtained. First we replace Eq. (3) (of main text) in the spin operators then perform a $1/M$-expansion up to second order in bosons:

$$S^2 = 1 + \sum (m - 1) a_S^\dagger S,m,R a_S^S,m,R ,$$

with summation over the tetrahedron basis, except for the ground state. Similarly,

$$S^+ = \sum \sqrt{S(S + 1) - m(m + 1)} a_S^\dagger S,m+1,R a_S^S,m,R + \sqrt{2} a_{1,0,A_1}^\dagger$$

where the sum is over the 15 bosons defined for a tetrahedron (the boson representing the ground state is excluded).

In $\langle 0 | S^2 | f \rangle$ the final state $| f \rangle$ represents one of the 15 bosons $a_S^S,m,R$, and $| 0 \rangle$ is the boson vacuum, thus for the matrix elements it is enough to consider $S^\alpha$ up to the first order in bosons, the two-boson operator terms don’t have a contribution. Then $S^+ \approx \sqrt{2} a_{1,0,A_1}^\dagger$, $S^- \approx 2a_{1,0,A_1}$, and $S^z \approx 1$. As a consequence, $S^x$ and $S^y$ give finite matrix elements only with the mode $a_{1,0,A_1}^\dagger$.

Calculating the quadrupole matrix elements can reveal which modes are (electrically) active in optical measurements. The quadrupoles up to linear order in boson operators have the following form

$$Q^{xy} \approx -\frac{1}{2}, \quad Q^{x^2-y^2} \approx \frac{1}{2} a_{1,1,A_1}^\dagger + h.c., \quad Q^{xy} \approx \frac{i}{2} a_{1,1,A_1}^\dagger + h.c., \quad Q^{yz} \approx \frac{i}{2} a_{1,0,A_1}^\dagger + h.c., \quad Q^{zx} \approx \frac{1}{2\sqrt{2}} a_{1,0,A_1}^\dagger + h.c.$$ (24)

It follows from Eq. (24) that the quadrupoles $Q^{yz}$ and $Q^{zx}$ or in other words the $x$ and $y$ components of the electric polarization give finite matrix element for the mode $a_{1,0,A_1}^\dagger$, $Q^{xy}$ and $Q^{x^2-y^2}$ couple to $a_{1,1,A_1}^\dagger$, while $Q^{zx}$ has no matrix element in the uncoupled case.

The coupled limit is more complicated. First we need to rotate the basis so that the new boson $\tilde{a}_{1,1,A_1}^\dagger$ creates the ground state

$$\tilde{a}_{1,1,A_1}^\dagger = \cos \frac{\alpha}{2} a_{1,1,A_1}^\dagger + \sin \frac{\alpha}{2} a_{2,1,A_1}^\dagger ,$$

$$\tilde{a}_{2,1,A_1}^\dagger = -\sin \frac{\alpha}{2} a_{1,1,A_1}^\dagger + \cos \frac{\alpha}{2} a_{2,1,A_1}^\dagger .$$ (26)

The variational parameter $\alpha$ is calculated from minimizing the energy. $S$ is not a good quantum number anymore, the multipoles and the bosons of the four different tetrahedra are mixed by the inter-tetrahedron couplings and the modes acquire dispersion. After replacing $a_{1,1,A_1}^\dagger$ and $\tilde{a}_{1,1,A_1}$ with $\sqrt{M - \tilde{a}_{1,1,A_1}^\dagger \tilde{a}_{2,1,A_1}^\dagger - \sum a_S^\dagger S,m,R a_S^S,m,R}$ where the summation is over the 14 unchanged bosons, we perform the $1/M$ expansion and keep the terms in the Hamiltonian up to quadratic order in bosons

$$H = M^2 H(0) + M^{3/2} H(1) + M H(2).$$ (27)

The constant term $H(0)$ is the mean-field energy, the term $H(1)$ is linear in boson operators and is identically zero for the variational solution that minimizes the energy, and $H(2)$ represents the quadratic part that can be diagonalized using Bogoliubov transformation. In particular, $H(2)$ takes the following form:

$$H(2) = \frac{1}{2} \begin{pmatrix} A_k & A_{-k} \end{pmatrix}^T \begin{pmatrix} C & D \end{pmatrix} \begin{pmatrix} A_k & A_{-k} \end{pmatrix}$$

where $C = C^\dagger$ and $D = D^T$ are 60 $\times$ 60 matrices and the vector $A_k^\dagger$ contains the 60 excitations of the four tetrahedra in the unit cell (15 per tetrahedron):

$$A_k^\dagger = \begin{pmatrix} a_{1,1,A_1}^\dagger, \ldots, a_{1,4}^\dagger \end{pmatrix} ,$$

where the bosons of one tetrahedron are

$$a_{1,n}^\dagger = \begin{pmatrix} a_{1,1,A_1}^\dagger(k), \ldots, a_{1,4,E_2}^\dagger(k) \end{pmatrix} ,$$

The equation of motion reads

$$i\dot{a}_{S,m,R}(k) = [a_{S,m,R}(k), H(2)] = \omega_{S,m,R}(k) a_{S,m,R}(k)$$ (31)
So, to find the linear combinations of bosons that diagonalize $\mathcal{H}^{(2)}$ we need to solve the eigenvalue problem

$$
\begin{pmatrix}
C & D \\
-D^\dagger & -C
\end{pmatrix}
\begin{pmatrix}
u_n \\
v_n
\end{pmatrix}
= \omega_n
\begin{pmatrix}
u_n \\
v_n
\end{pmatrix}
$$

(32)

The physical eigenvectors (with positive eigenvalues) are associated with the states that diagonalize the Hamiltonian $\mathcal{H}^{(2)}$.

$$
\alpha_{\mu,k}^\dagger = u_\mu \cdot A_k^\dagger + v_\mu \cdot A_{-k}
$$

(33)

Then $[\mathcal{H}^{(2)}, \alpha_n^\dagger] = \omega_n \alpha_n^\dagger$ and the bosons $\alpha_n^\dagger$ satisfy the commutation relation $[\alpha_n, \alpha_m^\dagger] = \delta_{nm}$. The Hamiltonian $\mathcal{H}^{(2)}$ becomes diagonal:

$$
\mathcal{H}^{(2)} = \sum_{\mu=1}^{60} \omega_\mu \left( \alpha_\mu^\dagger \alpha_\mu + \frac{1}{2} \right).
$$

(34)

To check the reliability of the multiboson spin wave approach, we calculated the quantum fluctuations of the two copper sub-lattices. For Cu, the $z$ component of the spin operator in the multiboson formalism can be written as

$$
S^z_1 = -\frac{1}{4} \cos \alpha + \frac{\sqrt{3}}{4} \sin \alpha + \left( \frac{1}{4} \cos \alpha - \frac{\sqrt{3}}{4} \sin \alpha \right) a_1^\dagger a_{1,0,A_1} + a_{1,0,A_1}^\dagger a_1
$$

$$
+ a_{0,0,E_1,0}^\dagger a_{0,0,E_1,0} + a_{0,0,E_2}^\dagger a_{0,0,E_2} + \frac{1}{2} \left( \frac{1}{4} \cos \alpha - \frac{\sqrt{3}}{4} \sin \alpha \right) a_{1,1,E_1,0}^\dagger a_{1,1,E_1,0} + a_{1,1,E_1,0} a_{1,1,E_1,0}^\dagger
$$

$$
+ a_{1,2,0}^\dagger a_{1,2,0} a_{1,2,0}^\dagger a_{1,2,0} + \frac{1}{2} \left( \frac{1}{4} \cos \alpha - \frac{\sqrt{3}}{4} \sin \alpha \right) a_{1,1,E_1,0}^\dagger a_{2,0}^\dagger a_{2,0} a_{1,1,E_1,0} + a_{1,1,E_1,0}^\dagger a_{2,0} a_{2,0}^\dagger a_{1,1,E_1,0} + h.c.
$$

(35)

When calculating the expectation value, the linear operator terms do not give a contribution and we can use the spin length obtained from the variational solution: $S^z_1 = -\frac{1}{4} \cos \alpha + \frac{\sqrt{3}}{4} \sin \alpha$. Then the spin expectation value on Cu sites becomes

$$
\langle S^z_1 \rangle = S^z_1 - S^z_1 \sum_k \left( \langle a_{1,0,A_1,k}^\dagger a_{1,0,A_1,k} \rangle + \langle a_{1,0,E_1,k}^\dagger a_{1,0,E_1,k} \rangle + \langle a_{1,0,E_2,k}^\dagger a_{1,0,E_2,k} \rangle + \langle a_{2,0,A_1,k}^\dagger a_{2,0,A_1,k} \rangle 
$$

$$
+ \langle a_{0,0,E_1,0}^\dagger a_{0,0,E_1,0} \rangle + \langle a_{0,0,E_2}^\dagger a_{0,0,E_2} \rangle \right) 
$$

$$
- \left( S^z_1 + \frac{1}{2} \right) \sum_k \left( \langle a_{1,1,E_1,k}^\dagger a_{1,1,E_1,k} \rangle + \langle a_{1,1,E_2,k}^\dagger a_{1,1,E_2,k} \rangle + \langle a_{2,2}^\dagger a_{2,2} \rangle \right) 
$$

$$
- \left( S^z_1 - \frac{1}{2} \right) \sum_k \left( \langle a_{1,1,E_1,k}^\dagger a_{1,1,E_1,k} \rangle + \langle a_{1,1,E_2,k}^\dagger a_{1,1,E_2,k} \rangle + \langle a_{2,2}^\dagger a_{2,2} \rangle \right) 
$$

$$
- \left( S^z_1 - \frac{1}{4} \right) \sum_k \left( \langle a_{1,2,0}^\dagger a_{1,2,0} a_{1,2,0}^\dagger a_{1,2,0} \rangle - \left( S^z + \frac{1}{4} \right) \langle a_{1,2}^\dagger a_{1,2} \rangle \right) 
$$

$$
- \frac{1}{2} \sum_k \left( \langle a_{2,0,A_1,k}^\dagger a_{2,0,A_1,k} \rangle + \langle a_{0,0,E_2,k}^\dagger a_{0,0,E_2,k} \rangle - \langle a_{1,0,E_1,k}^\dagger a_{0,0,E_1,k} \rangle + h.c. \right) 
$$

\]
\[-\frac{\sqrt{3}}{4} \sum_k \left( \langle a_{1,1,A_1,k}^\dagger a_{2,1,A_1,k} \rangle + h.c. \right) - 2S_z^\tau \sum_k \langle \tilde{a}_{1,1,A_1,k}^\dagger \tilde{a}_{2,1,A_1,k} \rangle \right) \]

(36)

To determine $\delta S_1 = \langle S_z^\tau \rangle - S_z^\tau$, we use the inverse transformation of (33) and exploit the fact that in the boson vacuum, i.e. the ground state $\langle \alpha_{\mu,k}^\dagger \alpha_{\mu,k} \rangle = 0$ and $\langle \alpha_{\mu,-k}^\dagger \alpha_{\mu,-k} \rangle = 1$. The spin fluctuation on the Cu$_2$ site can be determined in a similar way.