Triplons, magnons, and spinons in a single quantum spin system: SeCuO₃

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Quantum magnets display a wide variety of collective excitations, including spin waves (magnons), coherent singlet-triplet excitations (triplons), and pairs of fractional spins (spinons). These modes differ radically in nature and properties, and in all conventional analyses any given material is interpreted in terms of only one type. We report inelastic neutron scattering measurements on the spin-1/2 antiferromagnet SeCuO₃, which demonstrate that this compound exhibits all three primary types of spin excitation. Cu₁ sites form strongly bound dimers while Cu₂ sites form a network of spin chains, whose weak three-dimensional (3D) coupling induces antiferromagnetic order. We perform quantitative modeling to extract all of the relevant magnetic interactions and show that magnons of the Cu₂ system give a lower bound to the spinon continua, while the Cu₁ system hosts a band of high-energy triplons at the same time as frustrating the 3D network.

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The exotic collective excitations observed in magnetic materials emerge from the rich spectrum of possible effects when quantum spin fluctuations act in different geometries, dimensionalities, and with different degrees of frustration. When fluctuations push a system beyond robust magnetic order and textbook spin waves, common types of excitation include triplons arising from structural dimerization [1–3] and order and textbook spin waves, common types of excitation when fluctuations push a system beyond robust magnetic order and textbook spin waves, common types of excitation when fluctuations push a system beyond robust magnetic order and textbook spin waves, common types of excitation. Here we investigate the coexistence of multiple excitation types by an inelastic neutron scattering (INS) study of SeCuO₃. This compound displays both qusilocalized high-energy states and weak magnetic order at low temperatures. We demonstrate that the excitation spectrum has one triplon branch, dispersing weakly around 27 meV, and a magnonlike branch below 4 meV, whose associated scattering intensity shows the clear fingerprints of spinon continua. By model calculations using linear spin-wave theory and perturbative methods, we deduce the interaction parameters of a minimal magnetic Hamiltonian, allowing us to describe SeCuO₃ in terms of two interacting spin subsystems, namely, dimers and chains, each of which shapes the magnetic excitations of the material. The S = 1/2 quantum magnet SeCuO₃ [30] has a monoclinic unit cell with space group P2₁/n and lattice parameters a = 7.71 Å, b = 8.24 Å, c = 8.50 Å, and β = 99.12°. Two
FIG. 1. Atomic structure and magnetic interactions of SeCuO$_3$. (a) Schematic representation of the atomic structure showing Cu$_1$ (orange), Cu$_2$ (blue), Se (green), and O (pink) atoms. (b) Projection on the ac and (c) on the ab plane, indicating the magnetic interactions of Table I. (d) Perspective view highlighting the Cu$_2$ chains and the direct interactions connecting them into coupled, buckled planes. (e) Geometry of the effective interactions mediated between Cu$_2$ atoms by the Cu$_1$ dimer units: $J_D$ and $J_D'$ are given in terms of the two different Cu$_1$-Cu$_2$ interactions, $J_{12}'$ (green) and $J_{12}$ (purple), and the dimer interaction, $J_D$ (black), by Eq. (2).

crystallographically inequivalent Cu sites, Cu$_1$ and Cu$_2$, are each surrounded by six O atoms, forming Cu$_2$O$_4$ plaquettes, with the remaining two O atoms forming the elongated octahedra represented for the Cu$_2$ atoms in Figs. 1(a) and 1(e). This elongation favors the $d_{z^2}$-y$^2$ orbitals, ensuring strong Cu$_1$ dimer units (orange shading in Fig. 1) of edge-sharing plaquettes whose superexchange paths have a Cu$_1$-O-Cu$_1$ angle of 101.9º [31]. Including the Cu$_2$ $d_{z^2}$-y$^2$ orbitals led to the proposal of a weakly coupled network of linear (Cu$_2$-Cu$_1$-Cu$_1$-Cu$_2$) tetramers [31], although this scenario cannot explain the magnetic susceptibility below 90 K. Recent nuclear quadrupole resonance measurements confirmed the formation of singlet states at $T \lesssim 200$ K [32], and together with nuclear magnetic resonance, electron spin resonance, and torque magnetometry experiments [33,34] were interpreted as reflecting two essentially decoupled subsystems, the strong, local Cu$_1$ dimers and weakly coupled Cu$_2$ spins hosting magnetic order below $T_N = 8$ K.

To access the full spin dynamics of SeCuO$_3$, we grew a 1 g single-crystal sample by chemical vapor transport. Thermal neutron INS measurements were performed on the IN8 spectrometer (ILL [35]) to probe the (hkh) scattering plane. The low-energy dynamics were studied on the cold-neutron spectrometers ThALES (ILL [36]) and 4F1 (LLB), the latter experiment probing the (hkh) scattering plane. Full details of the instrumental setups employed are provided in Sec. S1 of the Supplemental Material (SM) [37]. The measured intensities, $I(q, \omega)$, are directly proportional to the dynamical structure factor, $S(q, \omega)$, for scattering processes at momentum transfer $q$ and energy transfer $\omega$.

In Fig. 2 we present the high-energy dynamics of SeCuO$_3$ as measured on IN8. We obtained $I(q, \omega)$ for $q$ points along two orthogonal high-symmetry directions. At 2 K, each energy scan [Fig. 2(a)] has a resolution-limited peak that we fit with a Gaussian at all $q$ points to extract a weak dispersion around 26.5 meV [Fig. 2(b)], with smooth changes in intensity [Fig. 2(c)]. At 15 K, i.e., above $T_N$, the peak shows only a minimal downward shift and increased broadening [Fig. 2(a)]. Figures 2(d)–2(f) confirm that this mode persists at least until 120 K, i.e., far beyond $T_N$, and that its width is captured by the Lorentzian component of a Voigt line shape.

The weak $q$ dependence of this excitation indicates its nature as a near-localized triplon of the Cu$_1$ dimers, whose energy is given by $J_D$ in Fig. 1. Its Lorentzian width increases linearly with temperature until a value of 4 meV [Figs. 2(d) and 2(e)], which we will show reflects the coupling to the excitations of the Cu$_2$ subsystem. However, the primary thermal effect is intrinsic, as shown in Fig. 2(f) by comparing the mode amplitude with the probability, $[1 + 3 \exp(-J_D/k_B T)]^{-1}$, of finding a $J_D$ dimer in its singlet state at temperature $T$, which further confirms the triplon nature of this mode.

Turning to the low-energy dynamics measured at 2 K on ThALES and 4F1, representative background-subtracted constant-$q \omega$ scans are shown in Fig. 3. A strong low-energy mode is present at all $q$, but a continuum of scattering states persists above this feature, at least up to the highest measured energy of 4.5 meV. To visualize this continuum scattering, we present our intensity data as color maps in Fig. 4, and note that it appears in all three dimensions of reciprocal space. We return below to a detailed discussion of this continuum.

For a systematic analysis we perform a Gaussian fit to the peak at the lower edge of the continuum (Fig. 3) and collect two separate intensity contributions, $I_p$ from the Gaussian and $I_c$ from the excess scattering at all higher energies. The upper panels of Fig. 4 show the values of $I_p(q)$ and $I_c(q)$ extracted from 74 energy scans. The lower panels show a
FIG. 2. Triplon excitation. (a) Intensity, $I(\omega)$, at $q = (0.3 \, 0.3 \, 0.5)$, measured at low (blue) and intermediate (red) temperatures with the difference shown in black. (b) Dispersion, $\omega(q)$, of the triplon in two orthogonal $q$ directions; shading represents the width (FWHM) of the mode at each $q$ point. (c) Integrated intensity, $I(q)$, for the same directions. (d)–(f) Thermal evolution at $q = (0.3 \, 0.3 \, 0.0)$. (d) Lorentzian width, $\Gamma_C$ (red), compared to $k_B T$ (blue). (e) $\omega(q, T)$; shading indicates the instrumental resolution of 1.8(2) meV (red) and the Lorentzian profile (blue). (f) Normalized $I(q)$ in red compared with the thermal singlet population (blue).

well-defined band with a maximum of 4 meV and a small gap, $\Delta = 0.42(3)$ meV, where $I_p(q)$ becomes large due to the magnetic order.

We expect that, with the exception of a term opening a sum over bonds in the set $\{J_m\}$ coupling two quasiindependent magnetic sublattices, the low intensity indicates that they are very weak.

The high-energy response as a triplon of the Cu 1 subsystem with $J_D = 26.5$ meV, we build up our knowledge of the terms in Eq. (1) by next describing the low-energy response as a consequence of the decoupled Cu 2 subsystem, i.e., by neglecting the second term. For this we seek a set of interaction parameters that, used in an effective Hamiltonian of the same form as the last term of Eq. (1), reproduces the magnon dispersions and intensities in Fig. 4. As shown in Fig. 1, we allow both near-neighbor couplings, $\{J_m\}$, and long-distance “effective” couplings, $\{J_p\}$, over paths that include the polyhedra of other Cu 1 atoms and whose microscopic origin therefore lies in the second term of Eq. (1), as we discuss further below.

We fit $\omega(q)$ using linear spin-wave (LSW) theory, as implemented in the spinw package [38], obtaining an excellent account of the INS peak positions when the interaction parameters of Fig. 1 have the values reported in Table I. This fit contains two magnon branches, one of which has over 90% of the intensity we measure. The LSW theory delivers an accurate account of $I_p(q)$ for the strong branch with no further fitting, as Fig. 4 makes clear, underlining its success in capturing all the leading physics of the magnon spectrum.

Turning to percent-level discrepancies, in the LSW treatment the weak branch has a very low [$\mathcal{O}(1\%)$] intensity, whereas the intensities measured in Fig. 4 are in general 1%–5% of the strong branch. While this discrepancy suggests that the magnetic Hamiltonian of SeCuO 3 contains further terms (beyond those in Table I) coupling two quasiindependent magnetic sublattices, the low intensity indicates that they are very weak.

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The interactions of Table I define a Cu 1 magnetic lattice composed of chains aligned in the $a - c$ direction, whose energy scale, $J_1$, exceeds by a factor of 10 all the interchain couplings. From Fig. 1(a), $J_1$ connects Cu 2 spins through the SeO 3 tetrahedra, a superexchange path not so far considered [31–34]. This coupled-chain character provides an immediate indication for the origin of the continuum scattering in Figs. 3 and 4 as the break-up of $\Delta S = 1$ magnons into spinons at energies beyond their confinement scale. The four additional Cu 2 interactions ensure both strong interchain frustration in all three directions and the weak magnetic order at $T < T_N$.

TABLE I. Superexchange interaction parameters within the Cu 1 and Cu 2 subsystems, in meV, obtained by fitting the dispersion data of Figs. 2(b) and 4. The geometry of the interactions is shown in Fig. 1.

| Cu 1-Cu 1 | Cu 2-Cu 2 |
|-----------|------------|
| $J_D$ | $J_1$ | $J_\perp$ | $J_0$ | $J_a$ | $J_\beta$ |
| 26.5(6) | 3.39(13) | 0.39(3) | −0.19(2) | 0.34(2) | 0.35(2) |
The small magnon gap can be reproduced with a tiny Ising anisotropy of $J_{\perp}$, $\delta J_1 = 0.018$ meV, which has negligible influence on the dynamics away from the zone center.

To go beyond the independent-subsystem interpretation of the excitation spectrum, we restore the coupling between Cu$_1$ and Cu$_2$ atoms in the second term of Eq. (1), without which the triplon measured in Fig. 2 would be nondispersive. All the interactions between the Cu$_2$ atoms given in Table I are required to fit the dispersion data of Fig. 4 in multiple reciprocal-space directions. However, they include not only the near-neighbor couplings $J_{1|1}$, $J_{1|2}$, and $J_6$, but also the couplings $J_{10}$ and $J_{12}$ whose long superexchange paths proceed directly across the Cu$_1$-Cu$_1$ dimer (Fig. 1); as a result, in a self-consistent model, these should be effective couplings arising as a consequence of $J_D$ and of two Cu$_1$-Cu$_2$ coupling parameters, $J_{12}^\gamma$ [Fig. 1(e)].

To estimate $J_{12}^\gamma$, we perform a perturbative analysis of the two four-site units shown in Fig. 1(e), as detailed in Sec. S2 of the SM [37]. The ground state in the limit $J_D \gg J_{12}^{\gamma}$, $|\Phi_0\rangle = |s_1\rangle \otimes |s_2\rangle$, is the product of two singlets on each pair of Cu$_1$ and Cu$_2$ sites. In the three lowest excited states, the Cu$_1$ dimer remains in a singlet while the Cu$_2$ spins form a triplet, $|l_{12}\rangle$, with $l = +, 0, -$. The energy difference gives the effective coupling between the two Cu$_2$ atoms,

$$J_\gamma = \frac{J_{12}^{\gamma}}{2} + \frac{1}{4} \left[ \frac{3(J_{12}^{\gamma})^2 - 2J_DB_{12}^{\gamma}}{\sqrt{J_D^2 + (J_{12}^{\gamma})^2}} \right] \rightarrow \frac{3}{4} \frac{(J_{12}^{\gamma})^2}{J_D}.$$  \hspace{1cm} (2)

From the fitted values of the effective couplings $J_{10}$ and $J_{12}$ (Table I), we deduce the microscopic coupling parameters to be $J_{12}^{\alpha} = 3.47$ meV and $J_{12}^{\beta} = 3.52$ meV. Our LSW fits verify that any effective coupling between atoms connected by a path involving both $J_{12}^{\alpha}$ and $J_{12}^{\beta}$ must be vanishingly small.

Although these values are large compared to the couplings within the Cu$_1$ subsystem in Table I, their real effect on the spin dynamics is suppressed strongly by $J_D$, as the structure of Eq. (2) makes clear. Values of 3–4 meV are consistent with the width, $\Gamma_{\perp}$, of the triplon at high temperatures [Fig. 2(d)], which indicates its coupling to incoherent excitations. The perturbative treatment of Eq. (2) provides upper bounds for the $J_{12}^{\gamma}$ values and thus is the opposite limit to the LSW approach, which cannot describe the full system of Cu$_1$ and Cu$_2$ atoms. The two approaches indicate the range of possible renormalization effects due to quantum fluctuations, which is widest at intermediate energies (corresponding to the spinon continua). The extent of renormalization to LSW theory in SeCuO$_3$ can be gauged from the magnetic order on the Cu$_2$ sublattice, which despite its three-dimensional (3D) nature is $\mu_2 < 0.8\mu_B$ [32]. The interactions $J_{12}^{\gamma}$ induce order on the Cu$_1$ sublattice, although $\mu_1 \approx 0.35\mu_B$ is very weak even at the lowest temperatures, and hence a full description lies well beyond the LSW approximation. The relative canting of the $\mu_1$ and $\mu_2$ moment directions [31] and the intensity transfer to the weak magnon branch (Fig. 4) gauge the physical effects of terms omitted in the minimal model of Eq. (1).
We return now to the most unexpected feature of our INS data, the strong continuum scattering observed directly above the magnon peaks in Figs. 3 and 4. Interpreting this as deconfined spinons requires the definitive exclusion of alternative origins. Continuum scattering above a one-magnon band arises naturally due to multimagnon processes and has long been known in both 3D [39] and 2D antiferromagnets [40]. In this situation, the ratio of the integrated intensities, $I_p$ in the one-magnon sector and $I_c$ in the putative multimagnon sector (Fig. 4), cannot exceed a well-defined limit. In Table II we average both quantities along four $\mathbf{q}$ directions, and note further that our measured energy range may not capture the upper edge of the continuum (Fig. 3), whence the ratio $\kappa_{\text{min}} = I_c/I_p$ constitutes a lower bound.

We find that $I_c$ is of the same order as $I_p$, making their ratio far greater than those found in multimagnon scattering studies [39,40]. The LSW prediction for this ratio can be deduced from the spin reduction (quantum fluctuation above the magnon peaks in Figs. 3 and 4). Interpreting this INS data, the strong continuum scattering observed directly above the one-magnon band. However, the frustrating inter-chain interactions, which allow this spinonic character in a system with magnetic order, mean that the resulting continua (Fig. 4) are far from the familiar single-chain form, which was found in Ref. [29] for magnons and spinons coexisting in the quasi-1D limit. The problem of partially confined spinons has recently received considerable attention in some of the paradigm Heisenberg models within frustrated quantum magnetism [41–45], and SeCuO3 presents a materials example of this complex situation. While a detailed analysis lies beyond the scope of the present study, the locations of continuum scattering in Fig. 4 and the intensity ratios for magnon and spinon contributions will provide essential input for a complete theoretical description.

To conclude, we have investigated a member of the class of coupled-cluster, multisubsystem quantum magnetic materials in which magnon, triplon, and spinon excitations are present simultaneously. In SeCuO3, the clusters are strong Cu1 dimers and the second sublattice, Cu2, is a network of spin chains on which weak magnetic order appears below $T_N = 8$ K. By neutron spectroscopy we have determined not only the intrasublattice interactions but also the Cu1-Cu2 interactions that make the Cu1 triplon mode weakly dispersive, induce small Cu1 moments, and create frustrating interactions in the Cu2 sublattice, which contribute to the emergence of spinon continua above the magnons. From our results, SeCuO3 encapsulates the challenge of describing the coherent quantum correlations, or quantum entanglement, that in many systems lie beneath the (unentangled) veneer of magnetic order, and mandates an integrated theoretical treatment of how these correlations lead to all three coexisting excitation types.

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Table II. Integrated peak intensity, $I_p$, and continuum intensity, $I_c$, averaged along four high-symmetry directions. The lower row presents the LSW theoretical result for the spin reduction, $\Delta S_t = 0.13$, measured [32] on the Cu$_2$ sublattice.

| Direction | $I_p$ | $I_c$ | $\kappa_{\text{min}}$ |
|-----------|-------|-------|----------------------|
| $[h, 2, k]$ | 90(4) | 61 | 0.69(4) |
| $[0, k, 0]$ | 133(5) | 99 | 0.85(4) |
| $[h, 3, k]$ | 98(4) | 125 | 1.28(4) |
| $\{\frac{1}{2}, k, \frac{1}{2}\}$ | 85(4) | 56 | 0.65(4) |
| $\Delta S_t = 0.13$ | 0.47 | 0.15 | 0.32 |

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Correction: Support information was missing in the Acknowledgment section and has been inserted.