Disorder-induced spin-liquid-like behavior in kagome-lattice compounds

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Quantum spin liquids (QSLs) are an exotic state of matter that is subject to extensive research. However, the relationship between the ubiquitous disorder and the QSL behaviors is still unclear. Here, by performing comparative experimental studies on two kagomé-lattice QSL candidates, TmSb$_3$Zn$_2$O$_{14}$ and TmSb$_3$Mg$_2$O$_{14}$, which are isostructural to each other but with strong and weak structural disorder, respectively, we show unambiguously that the disorder can induce spin-liquid-like features. In particular, both compounds show dominant antiferromagnetic interactions with a Curie-Weiss temperature of -17.4 and -28.7 K for TmSb$_3$Zn$_2$O$_{14}$ and TmSb$_3$Mg$_2$O$_{14}$, respectively, but remain disordered down to about 0.05 K. Specific heat results suggest the presence of gapless magnetic excitations characterized by a residual linear term. Magnetic excitation spectra obtained by inelastic neutron scattering (INS) at low temperatures display broad continua. All these observations are consistent with those of a QSL. However, we find in TmSb$_3$Zn$_2$O$_{14}$ which has strong disorder resulting from the random mixing of the magnetic Tm$^{3+}$ and nonmagnetic Zn$^{2+}$, that the low-energy magnetic excitations observed in the specific heat and INS measurements are substantially enhanced, compared to those of TmSb$_3$Mg$_2$O$_{14}$ which has much less disorder. We believe that the effective spins of the Tm$^{3+}$ ions in the Zn$^{2+}$/Mg$^{2+}$ sites give rise to the low-energy magnetic excitations, and the amount of the random occupancy determines the excitation strength. These results provide direct evidence of the mimicry of a QSL caused by disorder.

I. INTRODUCTION

Quantum spin liquids (QSLs) represent a novel state of matter in which spins are highly entangled, but do not order nor freeze even in the zero-temperature limit.5–7 Such a state does not involve any spontaneous symmetry breaking, which is beyond Landau’s paradigm for a phase and the associated transition.8 They are proposed to host fractional excitations and emergent gauge structures, and thus are promising candidates for quantum computation.9 Furthermore, high-temperature superconductivity may emerge from carrier doping a QSL.10,11 Thus, it has been a long-sought goal to achieve the QSL state. However, spins often tend to order at low temperatures.12 One approach to the goal is to introduce geometrical frustration into a low-spin system to enhance quantum fluctuations, so magnetic exchange interactions cannot be satisfied simultaneously among different lattice sites and the static magnetic order is prohibited.13–15 By now, a number of QSL candidates resulting from geometrical frustrations have been proposed and explored experimentally,16,17 and some typical examples include organic triangular-lattice systems κ-(ET)$_2$Cu$_2$(CN)$_3$18–20 and EtMg$_2$C$_6$[Pd(dmit)$_2$]$_2$.21,22 Inorganic triangular-lattice compound ZnCu$_3$(OH)$_6$Cl$_2$.23–25 Inorganic triangular-lattice compound YbMgGaO$_4$.26–28 and its sister compound YbZnGaO$_4$.29–31, more recently found triangular-lattice system delafossites,32–34. The disorder-free delafossites with effective spin-1/2 moments provide an excellent platform to unveil the QSL nature in a clean system. However, for most of these QSL candidates, the magnetic or nonmagnetic disorder can be significant, and complicates the interpretation of the intrinsic physics of the investigated systems.35–37 For this reason, how disorder affects the QSL state is still a controversial issue.

Recently, $REE_3$Sb$_3$M$_2$O$_{14}$ as a new family of two-dimensional kagomé-lattice compounds were synthesized,38–40 where $REE^{3+}$ represents rare-earth ions and $M^{2+}$ denotes nonmagnetic Zn$^{2+}$ or Mg$^{2+}$ ions. Among these compounds, Tm$_3$Sb$_3$Zn$_2$O$_{14}$ was proposed to be a QSL.41 The Tm$^{3+}$ ion has an electron configuration of $4f^{12}$ and, according to the Hund’s rule, it has a total angular momentum of $J = 6$ with a 13-fold degener-
acy. Considering the crystal-electric-field (CEF) effect, the degeneracy of \( J \) will be lifted, which was suggested to give rise to a non-Kramers doublet ground state\(^{57}\). In III B we will discuss the ground state and CEF excitations in more details. There is no signature of magnetic phase transition by magnetic susceptibility, heat capacity, and muon-spin relaxation measurements, suggesting the possible realization of a gapless QSL ground state\(^{57}\). However, it was shown that there was a significant site mixing between the magnetic Tm\(^{3+}\) and nonmagnetic Zn\(^{2+}\) sites\(^{57}\), causing a strong disorder effect that could impact the proposed QSL state.

In this work, in order to identify the role of disorder in the QSL candidates, we choose Tm\(_3\)Sb\(_2\)Zn\(_2\)O\(_{14}\) and its sister compound Tm\(_3\)Sb\(_3\)Mg\(_2\)O\(_{14}\) with quantifiable disorder, that is, strong and weak disorder in the former and latter, respectively, and investigate how disorder affects the material’s magnetic properties. On one hand, the absence of magnetic order and spin freezing down to \( \sim 50 \) mK far below the Curie-Weiss temperature (\( \Theta_{CW} \)), the presence of residual linear term in the specific heat, and the observation of broad gapless magnetic excitations resemble those of gapless QSLs. On the other hand, we find that the disorder resulting from the random mixing of magnetic Tm\(^{3+}\) and nonmagnetic Zn\(^{2+}\) or Mg\(^{2+}\) in the Tm layers is intimately correlated with the strength of the low-energy magnetic excitations. In particular, in Tm\(_3\)Sb\(_2\)Zn\(_2\)O\(_{14}\) which has strong disorder, as identified from the structural refinement and CEF excitations, the value of the residual linear term in the specific heat is about 4 times larger, and the intensity of the gapless spin excitations in the INS spectra is greatly enhanced, compared to those of Tm\(_3\)Sb\(_3\)Mg\(_2\)O\(_{14}\) with much less disorder. These results demonstrate conclusively that disorder in a geometrically frustrated compound can make it mimic a QSL.

II. EXPERIMENTAL DETAILS

Polycrystalline samples of Tm\(_3\)Sb\(_2\)Zn\(_2\)O\(_{14}\) (La\(_3\)Sb\(_2\)Zn\(_2\)O\(_{14}\)) and Tm\(_3\)Sb\(_3\)Mg\(_2\)O\(_{14}\) (La\(_3\)Sb\(_3\)Mg\(_2\)O\(_{14}\)) were synthesized by conventional solid-state reactions with stoichiometric amounts of Tm\(_2\)O\(_3\) (99.99%), Lu\(_2\)O\(_3\) (99.99%), Sb\(_2\)O\(_3\) (99.99%), ZnO (99.99%), and MgO (99.99%) powders. The mixtures of the precursor compounds of Tm\(_3\)Sb\(_2\)Zn\(_2\)O\(_{14}\) (La\(_3\)Sb\(_3\)Zn\(_2\)O\(_{14}\)) were carefully ground and sintered at 1200 \( ^\circ \)C for 3 days. For Tm\(_3\)Sb\(_3\)Mg\(_2\)O\(_{14}\) (La\(_3\)Sb\(_3\)Mg\(_2\)O\(_{14}\)) samples, a higher reaction temperature of 1350 \( ^\circ \)C and longer reaction time of 5 days were required to obtain the pure phase. X-ray diffraction (XRD) data were collected in an x-ray diffractometer (X’TRA, ARL) using the Cu-K\(\alpha\) edge with a wavelength of 1.54 \( ^\circ \)A. Rietveld refinements on the data were run by the EXPGUI and GSAS programs\(^{55,59}\). The dc magnetic susceptibility was measured in a Quantum Design physical property measurement system (PPMS, EverCool). Specific heat above 2 K was measured on 5.6-mg Tm\(_3\)Sb\(_2\)Zn\(_2\)O\(_{14}\) and 4.7-mg Tm\(_3\)Sb\(_3\)Mg\(_2\)O\(_{14}\) samples, respectively, in a PPMS DynaCool equipped with a dilution refrigerator.

INS experiments on 5.3-g Tm\(_3\)Sb\(_2\)Zn\(_2\)O\(_{14}\) and 4.5-g Tm\(_3\)Sb\(_3\)Mg\(_2\)O\(_{14}\) polycrystalline samples were carried out on PELICAN equipped with a dilution refrigerator, a cold neutron time-of-flight (TOF) spectrometer located at ANSTO at Lucas Heights, Australia. The powders were loaded into a pure copper can in the dilution refrigerator, which was able to cool down to around 50 mK. The incident neutron wavelength was selected as \( \lambda \sim 4.69 \) \( ^\circ \)A, corresponding to an incident energy of 3.69 meV and an energy resolution of \( \Delta E = 0.067 \) meV (half width at half maximum, HWHM). For each temperature, we collected data for about 12 hours. The CEF experiments were performed on 8-g powders on a thermal triple-axis spectrometer TAIPAN at ANSTO. The powders were loaded into an aluminum can and then mounted onto a closed-cycle refrigerator which could reach 1.6 K. A pyrolytic graphite (PG) filter was placed after the sample to reduce contaminations from higher-order neutrons. The beam collimations were 4°-40°-sample-40°-4°. A fixed-final-energy (E\(_f\)) mode with E\(_f\) = 14.87 meV was used in the measurements. The resulting energy resolution was about 0.41 meV (HWHM). Measurements were performed under vertical-focusing conditions for both the monochromator and analyzer on TAIPAN.

III. RESULTS

A. Structure and magnetic susceptibility

Figures 1(a) and 1(b) show the schematics of the crystal structure and the two-dimensional kagome layer of Tm\(_3\)Sb\(_2\)Zn\(_2\)O\(_{14}\), respectively. Magnetic Tm\(^{3+}\) ions forming corner-shared kagome-lattice layers are well separated by nonmagnetic layers and have an ABC stacking arrangement along the c axis\(^{56,57}\). We have performed Rietveld refinements on the XRD data of Tm\(_3\)Sb\(_2\)Zn\(_2\)O\(_{14}\) and the results are shown in Fig. 1(c). For the \( R3m \) space group with a perfect kagome lattice, there should be some Bragg reflections below 30 degrees, which are absent in our data. In order to capture the absence of low-angle reflections, we have to allow some mixings between the Tm\(^{3+}\) and Zn\(^{2+}\)(1) sites. Such a site-mixing model was used in Ref. 57 to analyze the XRD data in Tm\(_3\)Sb\(_2\)Zn\(_2\)O\(_{14}\) as well. In order to keep the stoichiometry, we need to free the site mixing between the Tm\(^{3+}\) and Zn\(^{2+}\)(2) sites as well, which was not considered in Ref. 57. The detailed refinement parameters are listed in Table I. Our refinement results show that there are around 35% Zn\(^{2+}\) ions occupying Tm\(^{3+}\) positions, and 55% and 56% Tm\(^{3+}\) ions occupying Zn\(^{2+}\)(1) and Zn\(^{2+}\)(2) positions, respectively. The strong site mixing of Tm\(^{3+}\) and Zn\(^{2+}\) reduces the distinctness of their original positions and
then increases the crystallographic symmetry, i.e., from kagomé to triangular within the $a$-$b$ planes. This naturally explains the absence of the low-angle Bragg reflections in Fig. (c).

The random site mixing between the magnetic and nonmagnetic sites is expected to have a strong impact on the magnetic properties, which is indeed the case as will be discussed later. To address this issue, it is better to have a compound that is isostructural to Tm$_3$Sb$_3$Zn$_2$O$_{14}$ but has less disorder resulting from the random mixing of the magnetic and nonmagnetic ions. For this purpose, we have replaced the nonmagnetic Zn in Tm$_3$Sb$_3$Zn$_2$O$_{14}$ with Mg and synthesized Tm$_3$Sb$_3$Mg$_2$O$_{14}$, which has less disorder as we show below. We have performed similar refinements and the results are shown in Fig. (d) and Table I. The XRD pattern for Tm$_3$Sb$_3$Mg$_2$O$_{14}$ shown in Fig. (d) is almost the same as that for Tm$_3$Sb$_3$Zn$_2$O$_{14}$ shown in Fig. (c), except for the additional reflections below 30 degree, which is expected for the $R3m$ space group with perfect kagomé layers. As shown in Table I, the crystal structure for both compounds is the same, but the amount of Tm$^{3+}$ in the Mg$^{2+}$ sites is significantly reduced. We believe that the different amounts of disorder in Tm$_3$Sb$_3$Zn$_2$O$_{14}$ and Tm$_3$Sb$_3$Mg$_2$O$_{14}$ are due to the different radii of the Zn$^{2+}$ and Mg$^{2+}$ ions. Compared to the smaller Mg$^{2+}$, the larger Zn$^{2+}$ ions are closer to Tm$^{3+}$ ions in size, and thus it is easier to occupy each other randomly. In support of this point, it has been reported that when the radius of the rare-earth ion becomes large enough to have an obvious difference from that of Zn$^{2+}$ ion, such as Dy$^{3+}$ or larger ones in the 4$f$ row, the disorder effect will be weakened significantly.

We further characterize both compounds by measuring the magnetic susceptibility ($\chi$) with a magnetic field of 0.1 T, and the results are shown in Figs. (e) and (f). There is no indication of long-range magnetic order down to 2 K. In addition, the absence of the bifurcation of zero-field-cooling (ZFC) and field-cooling (FC) data also indicates that there is no spin freezing at the lowest temperature measured. The inverse susceptibility is linear for most of the temperature range, except for the slight deviation at low temperatures, as shown in the inset of Figs. (e) and (f). Such a deviation may be associated with the development of short-range magnetic correlations, and is commonly observed in QSL candidates. It can also be explained with the thermal population of CEF levels and we will discuss it later. From the Curie-Weiss fits, we obtain the Curie-Weiss temperature $\Theta_{CW}$ of -17.4 and -28.7 K for Tm$_3$Sb$_3$Zn$_2$O$_{14}$ and Tm$_3$Sb$_3$Mg$_2$O$_{14}$, respectively, implying dominating antiferromagnetic spin correlations in these materials. These results are consistent with the QSL state expected for a kagomé-lattice system with antiferromagnetic interactions and strong geometrical frustration. On the other hand, the disorder resulting from the random mixing of the magnetic and nonmagnetic sites discussed earlier can be also responsible for

![Figure 1](https://example.com/figure1.png)
TABLE I. Room-temperature XRD pattern refinements for Tm₃Sb₂Zn₂O₁₄ and Tm₃Sb₃Mg₂O₁₄.

| Compound          | Atom           | Wyckoff position | x       | y       | z       | Occ. | Uₘ₀ (Å²) | a (Å)  | c (Å)  | χ²  |
|-------------------|----------------|------------------|---------|---------|---------|------|----------|--------|--------|-----|
| Tm₃Sb₂Zn₂O₁₄     | Tm(1)         | 3a               | 0       | 0       | 0       | 0.55 | 0.45(3)  | 7.354(2) | 16.9956(5) | 3.84 |
|                   | Zn(1)         | 3a               | 0       | 0       | 0       | 0.55 | 0.45(3)  | 7.354(2) | 16.9956(5) | 3.84 |
|                   | Zn(2)         | 3b               | 0       | 0       | 0.5     | 0.44 | 0.56(4)  | 0.025 | 7.354(2) | 16.9956(5) | 3.84 |
|                   | Sb            | 9d               | 0.5     | 0       | 0       | 1   | 1        | 1      | 1      |     |
|                   | O(1)          | 6c               | 0       | 0       | 0.393  | 1   | 1        | 1      | 1      |     |
|                   | O(2)          | 18h              | 0.504(3)| -0.504(3)| 0.117(2)| 1   | 1        | 1      | 1      |     |
|                   | O(3)          | 18h              | 0.143(3)| -0.143(3)| -0.026(2)| 1   | 1        | 1      | 1      |     |
| Tm₃Sb₃Mg₂O₁₄     | Mg(1)         | 3a               | 0       | 0       | 0       | 0.55 | 0.18(1)  | 0.025 | 7.2772(5) | 17.2278(1) | 3.67 |
|                   | Mg(2)         | 3b               | 0       | 0       | 0.5     | 0.93 | 0.07(1)  | 1      | 1      |     |
|                   | Tm(1)         | 3a               | 0       | 0       | 0       | 1   | 1        | 1      | 1      |     |
|                   | Tm(2)         | 3b               | 0       | 0       | 0.5     | 0.18 | 0.05(1)  | 1      | 1      |     |
|                   | Tm(3)         | 18h              | 0.522(2)| -0.522(2)| 0.139(1)| 1   | 1        | 1      | 1      |     |
|                   | Sb            | 9d               | 0.5     | 0       | 0       | 1   | 1        | 1      | 1      |     |
|                   | O(1)          | 6c               | 0       | 0       | 0.615(2)| 1   | 1        | 1      | 1      |     |
|                   | O(2)          | 18h              | 0.132(2)| -0.132(2)| -0.058(1)| 1   | 1        | 1      | 1      |     |
|                   | O(3)          | 18h              | 0.132(2)| -0.132(2)| -0.058(1)| 1   | 1        | 1      | 1      |     |

T = 300 K, wavelength of the x-ray λ = 1.54 Å, and space group: R3m.
Uₘ₀ denotes the isotropic atom displacement from the equilibrium positions, and χ² represents the goodness of fitting.

B. CEF excitations

To further investigate the disorder effect in these two compounds, we measured the CEF excitations, and the experimental results are shown in Fig. 2. The experimental CEF spectra of Tm₃Sb₃Mg₂O₁₄ are shown in Fig. 2(b). There are three well-isolated CEF transitions centered at 4, 23.5, and 33.5 meV, respectively, with no asymmetry nor significant broadening observed. For Tm₃Sb₂Zn₂O₁₄, there are two CEF levels observed at 1.6 and 4.5 meV. Both peaks are slightly broader than the instrument resolution of 0.41 meV but still relatively sharp. However, another one centered at 19 meV is asymmetric and much broader. To analyze the experimental results in Tm₃Sb₂Zn₂O₁₄, we need to consider the random site mixing between the Tm³⁺ and Zn²⁺ ions, which is strong as shown in Table I. A Zn²⁺(1) cation with surrounding six O²⁻ anions produces a ZnO₆ polyhedra, while the Zn²⁺(2) and Tm³⁺ cations are both surrounded by eight O²⁻ anions. As a result, the Tm³⁺ ions at the original and Zn(1) sites have different ligand environments and should give rise to two different sets of CEF excitations associated with these two sites. This indeed makes the CEF pattern of Tm₃Sb₂Zn₂O₁₄ more complicated than that of Tm₃Sb₃Mg₂O₁₄. By comparing their experimental results in Figs. 2(a) and 2(b), we believe the CEF transitions observed at 4.5, 24.8, and 33.5 meV of Tm₃Sb₂Zn₂O₁₄ can be attributed to excitations of the Tm³⁺ ions at the original sites, since these energies are almost the same as those of Tm₃Sb₃Mg₂O₁₄, although the one at 33.5 meV is weaker than that in the latter. In Tm₃Sb₂Zn₂O₁₄, the remaining two CEF levels at 1.6 and 18.5 meV which are completely absent in Tm₃Sb₃Mg₂O₁₄ should result from the CEF excitations of Tm³⁺ cations at the Zn(1) sites. In this case, the asymmetric and very broad peak around 19 meV for Tm₃Sb₂Zn₂O₁₄ shown in Fig. 2(a) is actually composed of two CEF levels centered at 18.5 and 24.8 meV resulting from two different sites. These results show clearly that the site mixing between the magnetic and nonmagnetic ions will have a strong impact on the CEF excitations.

In order to confirm the signals we have observed here indeed originate from CEF excitations, some Q scans located at representative energy levels were performed. In Figs. 2(c) and 2(d), the behavior of monotonic decrease of intensities with increasing Q follows the magnetic form factor of a Tm³⁺ ion, distinct from phonon scatterings. Moreover, INS results of a nonmagnetic reference compound La₃Sb₂Mg₂O₁₄ show phonon scattering is only pronounced at a larger range of Q ≥ 10 Å⁻¹ (Ref. 52), which is far away from our currently investigated area.

The rare-earth ions in the materials are strongly influenced by the electrostatic environment they occupy. Therefore, in order to quantitatively identify how the (2J + 1)-fold (J = 6) spin-orbital degeneracy is lifted by the CEF effect, a CEF analysis was performed, and the effective Hamiltonian was obtained by the point-charge model according to the point-group symmetry 23:

\[
H_{CEF} = \sum_{n,m} B_{nm}^{m} O_{n}^{m},
\]

where \(O_{n}^{m}\) and \(B_{nm}^{m}\) are the Steven operators 24 and CEF parameters, respectively. In the following, \(B_{nm}^{m}\) are calcu-
of $J_z = \pm 6$ states:

\[ |+\rangle \approx \frac{\sqrt{2}}{2} (|6\rangle + |-6\rangle), |\rangle \approx \frac{\sqrt{2}}{2} (|6\rangle - |\rangle). \tag{2} \]

Due to the low symmetry, the CEF peaks observed experimentally are not sufficient to simulate all the CEF parameters. In addition, the disorder would further influence the simulation. In this case, we simply do the symmetry analysis and obtain the effective CEF Hamiltonian by point-charge approximation. Since the lifting of the degeneracy is mainly determined by the point-group symmetry, we think the analysis is sufficient to identify the low-lying states, which are very far away from other higher energy levels. Further fitting would revise the higher energy levels, but the components of the non-Kramers doublet would not change significantly.

A similar non-Kramers doublet was also reported for Ho$_3$Sb$_3$Mg$_6$O$_{14}$ (Ref. 65) and Tb$_3$Sb$_3$Mg$_6$O$_{14}$ (Ref. 66), which share the same structure. The fitting analysis of the same structure of other crystals also supported that the symmetry analysis is qualitatively efficient for the low-lying states.

For the Tm$^{3+}$ cations at the Zn(1) sites in Tm$_2$Zn$_2$Sb$_3$O$_{14}$, the ligand environment is a squashed oxygen octahedron, whose symmetry is higher than the original sites. The CEF with $D_{3d}$ point group symmetry will split the 13-degenerate states into five singlets and four doublets. It has been revealed that two low-lying singlets would compose the nearly degenerate non-Kramers doublet. The components of the nearly degenerate non-Kramers doublet ground state are primarily $J_z = \pm 6$ and $\pm 3$ states. Therefore, an effective spin-1/2 can be defined in this doublet. Due to the occupations of the Tm$^{3+}$ cations at the original and Zn(1) sites, the two different ligand environments lead to different CEF excitations, which results in a very different CEF pattern shown in Fig. 2(a).

**TABLE II.** The CEF parameters obtained from the point-charge approximation.

| $B_{i}^{\text{meV}}$ | meV       |
|-------------------|-----------|
| $B_{1}^{1}$       | -1.40866  |
| $B_{2}^{1}$       | 2.02733   |
| $B_{3}^{1}$       | 0.79967   |
| $B_{4}^{1}$       | -0.00252  |
| $B_{5}^{1}$       | -0.01587  |
| $B_{6}^{1}$       | 0.00610   |
| $B_{7}^{1}$       | 0.03075   |
| $B_{8}^{1}$       | 0.02983   |
| $B_{9}^{1}$       | -0.00002  |
| $B_{10}^{1}$      | -0.00007  |
| $B_{11}^{1}$      | 0.00004   |
| $B_{12}^{1}$      | 0.00016   |
| $B_{13}^{1}$      | -0.00017  |
| $B_{14}^{1}$      | -0.00054  |
| $B_{15}^{1}$      | -0.00005  |

**lated based on the 15 nontrivial CEF parameters shown in Table II with the point-charge approximation.**

Since the CEF of Tm$^{3+}$ at the original sites has a very low symmetry of the $C_{2h}$ point group with eight oxygen ligands, the 13-fold degeneracy of the ground-state manifold $3H_8$ is expected to be completely lifted. Fortunately, the diagonalization of Eq. (1) predicts that the splitting of two low-lying singlets is small enough to be regarded as a nearly degenerate non-Kramers doublet. The effective spin in the doublet is an easy-axis moment, and the components are mostly $J_z = \pm 6$ along the easy axis. If we choose our local axes such that the two-fold rotation symmetry is about the $y$ axis and the easy axis is the $z$ axis, the doublet shown in Table II can be well described by symmetric and antisymmetric combinations...
TABLE III. Eigenvalues and eigenvectors for the non-Kramers doublet ground state of Tm$^{3+}$ at the original sites. The first column indicates the energies and the rest indicate the antisymmetric and symmetric states of the doublet.

| $E$ (meV) | $|−6⟩$ | $|−5⟩$ | $|−4⟩$ | $|−3⟩$ | $|−2⟩$ | $|−1⟩$ | $|0⟩$ | $|1⟩$ | $|2⟩$ | $|3⟩$ | $|4⟩$ | $|5⟩$ |
|----------|-------|-------|-------|-------|-------|-------|-----|-----|-----|-----|-----|-----|
| 0.000    | (0.701 0.010 -0.079 0.042 0.008 -0.024 0.000 -0.024 -0.008 0.042 0.079 0.010 -0.701) |
| 0.018    | (0.701 0.008 -0.086 0.031 0.001 -0.017 0.024 0.017 0.001 -0.031 -0.086 -0.008 0.701) |

C. Specific heat results

![Graphs showing specific heat results](image)

FIG. 3. (a) and (b) Specific heat results of Tm$_3$Sb$_3$Zn$_2$O$_{14}$ and Tm$_3$Sb$_3$Mg$_2$O$_{14}$ at ultralow temperatures. The specific heat of nonmagnetic references La$_3$Sb$_3$Zn$_2$O$_{14}$ and La$_3$Sb$_3$Mg$_2$O$_{14}$ are also shown. Solid lines are the fits with Debye model as $C_p \sim T^3$ for the nonmagnetic compounds. Insets show the low-temperature $C_p/T$ vs. $T^2$. Dashed lines are the linear fits. (c) and (d) Magnetic specific heat ($C_m$) results of Tm$_3$Sb$_3$Zn$_2$O$_{14}$ and Tm$_3$Sb$_3$Mg$_2$O$_{14}$ after subtracting the contribution from the lattice using nonmagnetic reference compounds La$_3$Sb$_3$Zn$_2$O$_{14}$ and La$_3$Sb$_3$Mg$_2$O$_{14}$, respectively.

We performed ultralow-temperature specific heat ($C_p$) measurements of these two compounds and the results are shown in Fig. 3. Figure 3(a) shows the specific heat results of Tm$_3$Sb$_3$Zn$_2$O$_{14}$ down to 70 mK. There is no sharp A-type peak expected for a well-defined phase transition. Instead, there is a kink around 7 K. This kink temperature almost coincides with the temperature when the susceptibility deviates from the Curie-Weiss behavior shown in Fig. 1(c). In some other QSL candidates, there is a more obvious hump which probably corresponds to the establishment of short-range spin correlations. We conjecture that the underlying physics for the kink observed here is similar. Another possible explanation is that it results from the thermal population of the low-lying CEF level located at 1.6 meV as shown in Fig. 3(a). As for a two-level system with the energy splitting of 1.6 meV, it gives rise to a maximum at 7.7 K which is close to the kink temperature observed in both $\chi(T)$ and $C(T)$. Generally speaking, the specific heat at low temperatures can be fitted as $C_p = \gamma T + \beta T^3$ for a system with gapless fermionic excitations (Refs. 17 and 22), where the linear $T$ term and $T^3$ term denote electronic and phononic contributions, respectively. Compared with the nonmagnetic reference compound La$_3$Sb$_3$Zn$_2$O$_{14}$ behaving well as $C_p/T \sim T^2$, which is reasonable, since it is an insulator. Tm$_3$Sb$_3$Zn$_2$O$_{14}$ has a large residual linear term. In the inset of Fig. 3(a), we plot $C_p/T$ as a function of $T^2$ to focus on the low-temperature part, and the linear extrapolation to absolute zero temperature yields a finite linear term coefficient $\gamma \sim 31.5(6) \text{ mJ mol}^{-1} \text{ K}^{-2}$. Such an observation is quite unusual for an insulator, and is often interpreted to be due to the fermionic fractional excitations such as spinons of a QSL. The specific heat results of Tm$_3$Sb$_3$Mg$_2$O$_{14}$ depicted in Fig. 3(b) exhibit similar behaviors. The kink shifts to a higher temperature of around 17 K, and it may reflect the fact that the exchange interaction is stronger than that in Tm$_3$Sb$_3$Zn$_2$O$_{14}$, which is consistent with the higher $\Theta_{CW}$ in Tm$_3$Sb$_3$Mg$_2$O$_{14}$. We believe the kink should have the same origin as discussed earlier for Tm$_3$Sb$_3$Zn$_2$O$_{14}$. The specific heat also exhibits a linear term, as shown in the inset of Fig. 3(b). However, the $\gamma$ value of $8.9(7) \text{ mJ mol}^{-1} \text{ K}^{-2}$ is more than three times smaller than that of $31.5(6) \text{ mJ mol}^{-1} \text{ K}^{-2}$ of Tm$_3$Sb$_3$Zn$_2$O$_{14}$, indicating much less density of states at low energies in Tm$_3$Sb$_3$Mg$_2$O$_{14}$. Considering the substantial amount of disorder in Tm$_3$Sb$_3$Zn$_2$O$_{14}$, we believe that the enhancement of the $\gamma$ is due to the disorder resulting from the strong site mixing of Tm$^{3+}$ and Zn$^{2+}$ ions. We also present the magnetic specific heat ($C_m$) results of these two compounds in Figs. 3(c) and 3(d). The kinks around 7 K in Tm$_3$Sb$_3$Zn$_2$O$_{14}$ and 17 K in Tm$_3$Sb$_3$Mg$_2$O$_{14}$ are more clearly shown after subtracting the phonon contributions.

D. INS spectra

We now turn to the INS measurements at low energies to gain further insights into the magnetic state. Figures 4(a) and 4(c) show the magnetic excitation spectra (raw data) collected for the polycrystalline sample of Tm$_3$Sb$_3$Zn$_2$O$_{14}$ at three characteristic temperatures. The INS results contain two pronounced features. First is...
the flat excitation band between \( E = 1.1 \) and \( 2.1 \) meV. These excitations are the low-lying CEF excitations of the \( \text{Tm}^{3+} \) ions as also shown in Fig. 2(a). Second is another broad and nearly flat excitation band below the CEF excitations, approximately in the energy range of 0.3 to 0.6 meV. The intensity weakens at 5 K [Fig. 4(b)] and disappears at 30 K [Fig. 4(c)]. To investigate the low-energy excitations in detail, we integrate the intensities with energy ranging from 0.3 to 0.6 meV to avoid contaminations from the CEF excitations and elastic scattering, and plot the integrated intensity as a function of \( Q \) in Fig. 4(d). It is clear that there is a broad peak centered at \( Q \sim 1 \) Å\(^{-1}\) which corresponds to the \( \Gamma \) point in the second Brillouin zone of a kagomé lattice. The \( Q \) and temperature dependence of these excitations indicate that they are of magnetic origin\(^{31,69}\). Figures 4(e)-4(g) show the INS spectra of \( \text{Tm}_3\text{Sb}_2\text{Mg}_2\text{O}_{14} \). As discussed earlier and shown in Fig. 2(b), the first CEF level of \( \text{Tm}_3\text{Sb}_2\text{Mg}_2\text{O}_{14} \) is 4 meV, which is beyond the energy range in Fig. 4. As a result, we do not observe the CEF excitations in Figs. 4(e)-4(g) for \( \text{Tm}_3\text{Sb}_2\text{Mg}_2\text{O}_{14} \). The INS measurements were performed on the samples with similar weights and thus similar amount of magnetic \( \text{Tm}^{3+} \) ions. Furthermore, we used the same experimental setup and equal counting time. Therefore, Figs. 4(a)-4(c) and 4(e)-4(g) can be compared directly. Different from that in \( \text{Tm}_3\text{Sb}_2\text{Zn}_2\text{O}_{14} \), the low-energy excitations are barely visible in Fig. 4(e) for \( \text{Tm}_3\text{Sb}_2\text{Mg}_2\text{O}_{14} \). We integrate the intensity between 0.3 and 0.9 meV, but since the intensity is too weak, we need to subtract the background data at 60 K to make it visible. The so-obtained intensity as a function of \( Q \) is shown in Fig. 4(d). There also appears to be a broad peak centered at \( Q \sim 1 \) Å\(^{-1}\), and the intensity increases as the temperature decreases. However, the overall intensities are significantly weaker than those in \( \text{Tm}_3\text{Sb}_2\text{Zn}_2\text{O}_{14} \).

To further investigate the broad excitations centered at \( Q \sim 1 \) Å\(^{-1}\), we integrated the intensities with \( Q \) ranging from 0.6 and 1.4 Å\(^{-1}\) at various temperatures, and the integrated intensities are plotted as a function of energy in Fig. 5. The intensities follow similar temperature dependence for both compounds: on the energy-loss side \( (E > 0) \), intensities increase with decreasing temperature; on the energy-gain \( (E < 0) \) side, the intensities are suppressed at low temperatures due to the detail balance. These results indicate that the intensities are resulting from intrinsic magnetic excitations. Moreover, the magnetic signals are dominated by the inelastic scattering, while the quasi-elastic scattering has no temperature dependence.

It is reasonable that the spectral weight is mainly distributed in the inelastic channels for a system with moderate spin interactions in which \( \Theta_{\text{CW}} \) is -17.4 and -28.7 K for \( \text{Tm}_3\text{Sb}_2\text{Zn}_2\text{O}_{14} \) and \( \text{Tm}_3\text{Sb}_2\text{Mg}_2\text{O}_{14} \), respectively. We integrate the elastic channels and plot the data collected at \( \sim 50 \) mK, and there is no magnetic Bragg peak observed in Figs. 5(c) and 5(d), which also proves the absence of magnetic order for both compounds down to \( \sim 50 \) mK.

IV. DISCUSSIONS AND CONCLUSIONS

How do we understand the INS results given the strong site mixing demonstrated from the XRD, CEF,
FIG. 5. Energy dependence of the integrated neutron scattering intensity in the $Q$ range between 0.6 to 1.4 Å$^{-1}$ for Tm$_3$Sb$_3$Zn$_2$O$_{14}$ (a) and Tm$_3$Sb$_3$Mg$_2$O$_{14}$ (b). Solid lines are guides to the eye. Dashed lines represent the instrumental resolutions. Elastic neutron scattering data for (c) Tm$_3$Sb$_3$Zn$_2$O$_{14}$ and (d) Tm$_3$Sb$_3$Mg$_2$O$_{14}$, obtained by integrating the intensity in an energy window of [-0.1,0.1] meV. The data were collected on a time-of-flight spectrometer PELICAN at various temperatures.

and specific heat results? At first glance, the low-energy magnetic excitations in Tm$_3$Sb$_3$Zn$_2$O$_{14}$ are distinct from conventional spin-wave excitations, but similar to the spectrum expected from the deconfined spinons in QSLs. However, we believe the disorder-induced low-energy excitations will be a more natural explanation. Since the low-lying state of Tm$^{3+}$ at the original sites is a nearly degenerate non-Kramers doublet and dominated by $J_z = \pm 6$ components by our CEF analysis, the effective spin residing in the doublet is more likely to behave as a multipole, which is not directly accessible for neutron scattering that has a selection rule of $\Delta S = \pm 1$ (Ref. [24]). This explains nicely why the low-energy excitations are so weak in Tm$_3$Sb$_3$Mg$_2$O$_{14}$, which has much less disorder. We need to point out that a well-isolated singlet ground state of Tm$^{3+}$ at the original sites in Tm$_3$Sb$_3$Mg$_2$O$_{14}$ was also proposed. Although we cannot rule out this possibility, it will not affect the conclusion for the absence of low-energy magnetic excitations resulting from the original sites, as there will be no extra magnetic excitations below the first CEF level from the isolated singlet ground state. We believe the spin excitations resulting from the effective spin of Tm$^{3+}$ in the Zn$^{2+}$ (2) sites will not give rise to the intensities observed in the INS experiment either as these two sites have similar CEF environments with eight O$^{2-}$ anions surrounding them. For Tm$_3$Sb$_3$Zn$_2$O$_{14}$ with a strong disorder effect, a large amount of Tm$^{3+}$ ions leave their original sites and occupy Zn$^{2+}$ (1) position which is surrounded by six O$^{2-}$ anions, different from the situation in their original sites. The CEF environment with a higher symmetry may preserve the effective spin-1/2 resulting from the dominating $J_z = \pm 6$ and $\pm 3$ components in the non-Kramers doublet, and in this case the dipole moments of the non-Kramers doublet are effective and can give rise to the low-energy magnetic excitations observed in the INS experiment. Furthermore, the Zn$^{2+}$ (1) sites are at the center of the hexagon. This will tune the kagomé lattice to triangular lattice effectively. In this case, the peak center of the excitation intensity $Q \sim 1$ Å$^{-1}$ corresponds to the M point of the twice-enlarged Brillouin zone in the triangular lattice due to the reduction of the $a$ axis by half. This is fully consistent with the observation of negative Curie-Weiss temperature for the dominant antiferromagnetic interactions. Of course, INS measurements on single crystals are desirable in order to better reveal the momentum distribution of the spectral weight. Nevertheless, our observation of the site-mixing-induced broad gapless excitations is fully in line with previous reports in YbZnGaO$_4$ (Ref. [51]), Yb$_2$TiO$_5$ (Ref. [52]), and κ-(ET)$_2$Cu[N(CN)$_2$]Cl (Ref. [53]).

At this point, we conclude that the disorder resulting from the site mixing of the magnetic Tm$^{3+}$ ions and the nonmagnetic ones is responsible for the absence of magnetic order, CEF excitations, specific heat, and the INS spectra. Intriguingly, the amount of disorder is manifested in the value of the residual linear term in the specific heat and the strength of the low-energy magnetic excitations. Then a natural question is what is the ground state with no disorder? One possibility is the valence-bond-solid state, in which two nearest-neighbor antiparallel spins form a singlet—such a state will not exhibit a magnetic order either. Another possibility is the gapped QSL state. In both cases, disorder will induce the low-energy magnetic excitations that contribute to the specific heat and magnetic neutron scattering. Although to distinguish these two states requires further experimental efforts, our results here demonstrate directly that by bringing disorder into such a geometrically frustrated system, a spin-liquid-like state can be induced. We believe this conclusion holds for a broad class of frustrated magnets including both the organic and inorganic compounds in the presence of either magnetic or nonmagnetic disorder. With many efforts, both experimental and theoretical, the role of disorder in the realization towards the spin-liquid-like behaviors has been made prominent. This also poses a great challenge for experimentalists in identifying a QSL.
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