Spin dynamics in the skyrmion-host lacunar spinel GaV₄S₈

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In the lacunar spinel GaV₄S₈, the interplay of spin, charge, and orbital degrees of freedom produces a rich phase diagram that includes an unusual Néel-type skyrmion phase composed of molecular spins. To provide insight into the interactions underlying this complex phase diagram, we study the spin excitations in GaV₄S₈ through inelastic neutron scattering measurements on polycrystalline and single crystal samples. Using linear spin wave theory, we describe the spin wave excitations using a model where V₄ clusters decorate an FCC lattice. The effective cluster model includes a ferromagnetic interaction and a weaker antisymmetric Dzyaloshinskii-Moriya (DM) interaction between the neighboring molecular spins. Our work clarifies the spin interactions in GaV₄S₈ and supports the picture of interacting molecular clusters.

I. INTRODUCTION

Magnetic skyrmions are topological spin textures characterized by an integer topological number and have potential for applications in spintronics devices [1–5]. Since the first discovery of the Bloch-type skyrmions in MnSi [6], different types of magnetic skyrmions have been identified [7–9]. Among them, the Néel-type skyrmion first realized in the polar system GaV₄S₈ has drawn great attention [10–15]. Distinct from the spin whorls in the Bloch-type skyrmions, Néel-type skyrmions are composed of spins rotating in the radial planes, which provides new opportunities for skyrmion manipulations [16–18].

GaV₄S₈ belongs to the lacunar spinel family of compounds with a chemical formula AM₄X₈ (A = Ga, Ge; M = V, Mo, Nb, and Ta; X = S and Se) [20–31], where the magnetic M ions form a breathing pyrochlore lattice as shown in Fig. 1(a). GaV₄S₈ is a magnetic semiconductor where the magnetism is associated with the unpaired valence electrons localized in the V₄ tetrahedra [32]. Assuming Ga and S take their most common valence configuration, charge balance yields Ga₃⁺V₄⁺S₈⁻ with 7 valence electrons per V₄ cluster. Due to the short V-V metal bonds in the smaller V₄ tetrahedra, the V 3dℓ-bands hybridize with each other, leading to a molecular spin-1/2 that is almost evenly distributed across the V₄ tetrahedra [33]. At temperatures below Tₘ ~ 44 K, GaV₄S₈ undergoes a cubic (Fm3m)-rhombohedral (R3m) transition due to a Jahn-Teller (JT) distortion, which elongates the V₄ tetrahedra along the [111] directions and results in an easy axis anisotropy for the V₄ spins as shown in Fig. 1. This anisotropy stabilizes a ferromagnetic (FM) ground state below Tₘ ~ 6 K [10, 11]. For temperatures between Tₘ and the long range order transition of Tₕ ~ 12.8 K, GaV₄S₈ has a thermal-fluctuation-stabilized cycloidal phase with a periodic-

FIG. 1. (a) Crystal structure of GaV₄S₈ highlighting the breathing pyrochlore V-sublattice composed of a periodic arrangement of large and small V₄ tetrahedra. (b) Molecular orbital diagrams of the V₄ tetrahedra in GaV₄S₈ with (below Tₘ) and without (above Tₘ) the Jahn-Teller distortion [19].

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The powder averaged INS spectra reveal spin wave excitations along the high symmetry directions, and the spectra are reproduced using a classical Heisenberg model that incorporates the ferromagnetic exchange interactions and the DM interactions.

**III. RESULTS AND DISCUSSION**

Figure 2 presents the powder-averaged INS spectra of GaV₄S₈ measured using SEQUOIA with \( E_i = 18 \) meV at \( T = 5 \) and 20 K. In the FM phase at 5 K, the spectra exhibit a single excitation emerging from \( Q = 0 \) with a bandwidth of \( \sim 6 \) meV. At larger wave-vector transfers the scattering decreases in intensity consistent with the cross-section being magnetic in origin. With a higher incident neutron energy of \( E_i = 40 \) meV (not shown), we confirm that no additional magnetic scattering is observed beyond that evident in Fig. 2(a). At 20 K (Fig. 2(b)), weak diffuse inelastic scattering is observed. This indicates that the spin-spin correlations survive above \( T_N = 12.8 \) K as expected for a material with geometrical frustration such as GaV₄S₈.
The INS data collected on the single crystal array at $T = 1.7$ K in the FM phase are summarized in Fig. 3. Reciprocal lattice unit (r.l.u.) indices are denoted in the cubic space group for convenience. Spin waves emanating from the ferromagnetic wave vectors are observed along the (001) and (110) directions as shown in Figs. 3(a) and (b), respectively. The periodicity of the FM spectrum can clearly be seen in the constant-energy slice in the $(HHL)$ plane. Calculated spin wave excitation spectra along the (d) $(−1−1L)$ (e) $(H−1)$ directions. (f) Calculated constant-energy slice at $E = 3$ meV in the $(HHL)$ plane. Measurements and calculations are further described in the text.

To understand the spin excitations in GaV$_4$S$_8$, we develop an effective Hamiltonian based on molecular $V_4$ spins. Recently Dally et al. have argued that the spin distribution is uniform over the $V_4$ tetrahedra[33]. Therefore, for the description of the spin wave dispersion in GaV$_4$S$_8$, the breathing pyrochlore lattice formed by the V ions can be effectively treated as a face-centered cubic (FCC) lattice of the $V_4$ molecular spins. A similar approach has been used to understand the spin-spin interactions in the Cr-breathing pyrochlore materials[36,38]. Unlike the aforementioned Cr-based materials where intra-cluster interactions are evident, the uniform spin distribution over the $V_4$ tetrahedra only modulates the intensities through a magnetic form factor $\int dr \exp(−i\mathbf{kr})\rho(r)$, where $\rho(r)$ describes a ferromagnetic spin density that is equally distributed over the 4 V sites, $\mathbf{k}$ is the wavevector transfer. It should be noted that a recent neutron diffraction experiment on GaV$_4$S$_8$ proposed the spins within the $V_4$ tetrahedra may be tilted by an angle of 39(8)$^\circ$[39]. Such an uncertainty in spin alignment may modify the exact form of the $V_4$ form factor but will not qualitatively change our effective model as long as the major spin components are ferromagnetic. The coupling strengths between the $V_4$ spins can then be parameterized by comparing the experimental data to the dynamical spin structure factor on a FCC lattice. Using the linear spin wave theory implemented in the SpinW software package[40], we consider an effective model with NN exchange and DM interactions:

$$H = \sum_{\langle ij \rangle} J \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\langle ij \rangle} D \cdot (\mathbf{S}_i \times \mathbf{S}_j),$$

where $\langle ij \rangle$ denotes the NN bonds with spins $\mathbf{S}_i$ and $\mathbf{S}_j$ located at the centers of the $V_4$ tetrahedra. The spin magnitude is assumed to be $S = 1/2$ per $V_4$ tetrahedra, which is consistent with the ordered moment of $\sim 0.25 \mu_B$ per V ion[33]. The direction of the DM vector is fixed based on the FCC lattice symmetry. According to the cycloidal pitch and the mean-field analysis of magnetization measurements reported in Ref.

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**FIG. 3.** Inelastic neutron scattering intensity as a function of energy and wave-vector transfer for GaV$_4$S$_8$ at T=1.7 K. Spin wave excitations along the (a) $(−1−1L)$ and (b) $(H−1)$ directions at $T = 1.7$ K measured using the MACS instrument. (c) Constant-energy slice of the INS spectrum at $E = 3$ meV in the $(HHL)$ plane. Calculated spin wave excitation spectra along the (d) $(−1−1L)$ (e) $(H−1)$ directions. (f) Calculated constant-energy slice at $E = 3$ meV in the $(HHL)$ plane.
we fixed the DM interaction to the reported value of $D = 0.13$ meV. To fit the data, we extracted 26 different energy and momentum values from the experimentally observed spin waves along the high symmetry directions $(110)$ and $(001)$. A particle swarm oscillation (pso) fitting method implemented in the SpinW software package is used to fit the data. The spin wave modes within the energy bin size are binned together and considered as one mode in the fit. The fit to the 2D dispersion data is carried out multiple times with more than 50 fitting cycles at each time. The goodness of fit (or R-value) of all those fits is around 2.7. Based on the value of the fitting parameter after all those runs; the value of exchange interaction $J$ and its uncertainty is optimized as $-0.72(3)$ meV. As already noted, the DM interaction is fixed based on the cycloidal pitch and the mean field analysis of magnetization measurements reported in Ref. [10]. The ratio of $D$ and $J$ in GaV$_4$S$_8$ is around 0.18 which is comparable to many other skyrmion host materials. For example, $D/J \sim 0.18$ stabilizes the skyrmion state in MnSi [41,42]. Whereas the nearest neighbor $D/J$ value of 0.39 in the skyrmion host bulk material Cu$_2$OSeO$_4$ [43] is larger than its value for GaV$_4$S$_8$. In the skyrmion ground state of a series of Mn$_{1-x}$Fe$_x$Ge $(0 < x < 1)$ compounds [44], the value of $D/J$ varies in the range of (0.02 to 0.6) depending upon the value of $x$.

The simulated powder average signal is presented in Figs 3(c) and 2(d). A model obtained by accounting the form factor of V$_4$ tetrahedra, Fig. 2(d), reproduces the measured powder spin spectra with better reliability than a model with the form factor of single V$^{3+}$ ion, Fig. 2(c), providing further support for the molecular picture for the V$_4$ tetrahedra. The exchange parameters reproduce spin wave like powder average excitation with zone center $|Q|=0$. The calculated energy at each $|Q|$ overlaps with the experimentally measured excitation energy. As the form factor of V$_4$ tetrahedra changes significantly, our model indicates that the intensity of excitation spectrum decreases rapidly with the increase in the value of $|Q|$, consistent with the experimental observation. The simulated spin wave spectra, including all domain contributions, for single crystal measurements are shown in the lower panels of Fig. 3. The energy scale and the features of the calculated excitation spectrum are in accord with the experimental data.

Including further neighbor interactions does not result in a discernible increase in the quality of the fit with the present data set. Additionally, we note that the observed ferromagnetic ground state originates in part due to anisotropy which stems from the rhombohedral structural distortion at 44 K. Such anisotropic terms are to too small to be extracted from the data and analysis presented here.

Figures 4(a) and (b) present the scattering intensity at fixed energy transfers as a function of L along the (-1 -1 L) and H along the $(H H -1)$ directions. Near the zone boundary, the line-cuts shows broader peak-width along the $(H H -1)$ direction compared to the (-1 -1 L) direction. The calculated constant energy line-cuts at the corresponding energies are over-plotted to the data. The calculated line-cuts successfully captures the peak position and broadness present in the data. The deviation between the model and the data near 5.5 meV is likely due to larger background scattering in this region of the spectrum.

We now compare the value of exchange coupling determined here with the value determined through other approaches [10,45]. Assuming the rhombohedral structure, Zhang et al. [45] calculated the NN (interplane) exchange coupling, using first-principles calculations and symmetry analysis. They found values in the range of $[-0.16, -1.68]$ meV depending on the value of the effective Coulomb potential. They reported a ratio $D/J$ of $\sim 0.16$ stabilizes the noncollinear spin structures in GaV$_4$S$_8$, which is consistent with our model. The scenario of different coupling strengths due to the rhombohedral distortion is also discussed in Ref. [45]. Mean-field analysis of magnetization measurements in the Ref. [10] showed the NN exchange interaction along the easy axis $\sim 0.38$ meV and an extremely weak mag-
netic anisotropy with a ratio of exchange couplings parallel and perpendicular to the easy axis $\sim 1.08$. The single magnon mode observed in our MACS experiment suggests the couplings strengths among the $V_4$ tetrahedra to be close to uniform even in the rhombohedral phase. As noted previously, the experimental resolution of our measurements is not sufficient to distinguish small anisotropy terms, our value of exchange coupling $J$ is fairly consistent with the value reported through other approaches.

IV. CONCLUSIONS

We have successfully grown single crystals of GaV$_4$S$_8$ of sufficient size to study the spin dynamics using INS. Our INS experiments reveal well-defined spin wave excitations with a bandwidth of $\sim 6$ meV, and an effective FCC lattice model of molecular $V_4$ spins reproduces the INS spectra. In addition to the ferromagnetic couplings of $J = -0.72(3)$ meV between neighboring $V_4$ spins, strong DM interactions with a magnitude of $D \sim 0.13$ meV are also confirmed. Our work clarifies the spin couplings in GaV$_4$S$_8$ and will facilitate the further understanding of its rich phase diagram.

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