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# Spin dynamics in the hyperkagome compound $Gd_3Ga_5O_{12}$

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We present neutron inelastic-scattering results on the low-temperature magnetic state of the frustrated-hyperkagome compound $Gd_3Ga_5O_{12}$ gadolinium gallium garnet (GGG). Our neutron-scattering studies reveal a remarkable range of time scales. Short-range spatial correlations appear static within the instrumental resolution (50 $\mu$eV). Three distinct inelastic modes are found at 0.04(1), 0.12(2), and 0.58(3) meV at 0.06 K. The lowest and highest energy excitations show spatial dependencies indicative of dimerized short-range antiferromagnetic correlations that survive to high temperatures, comparable to the nearest-neighbor exchange interactions. Our results suggest that the ground state of a three-dimensional hyperkagome compound differs distinctly from its frustrated counterparts on a pyrochlore lattice and reveal a juxtaposition of co-operative paramagnetism and strong-dimerized coupling. These results are surprising since GGG is often classified as a strongly frustrated system with a manifold of connected states for which one would expect a continuum of gapless excitations.

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## I. INTRODUCTION

Accumulated evidence suggests that magnetic frustration provides an excellent path to exotic magnetic order.[1–5] Evocative names such as spin liquids, spin glasses, and spin ice are associated with the frustration of magnetic spins. In a true spin liquid, due to a manifold of degenerate states, the spins remain fluctuating at temperatures much lower than the interaction energies.[3] An illustrative example of a spin liquid with a large spin value $S \gg \frac{1}{2}$, a co-operative paramagnet, is $Tb_2Ti_2O_7$. $Tb_2Ti_2O_7$ remains disordered down to the lowest temperatures[6] with an excitation spectrum that reveals, in addition to crystal-field excitations,[7] a quasielastic width of fluctuating spins that slow down with decreasing temperature but remain fluctuating down to 0.05 K.[8] A second example of a co-operative paramagnet is the kagome antiferromagnet deuteronium jarosite which shows gapless magnetic excitations extending out to at least 20 meV with a linear temperature dependence of the spin-fluctuation rate.[9] These two examples highlight the continuum of liquidlike quasielastic scattering typically observed in a co-operative paramagnet.

Structurally, rare-earth garnets such as gadolinium gallium garnet (GGG) are one of very few realizations of a hyperkagome structure, a three-dimensional lattice of corner sharing triangles. It can be considered as a depleted pyrochlore lattice in which only 3/4 of the vertices on each tetrahedron are occupied by a magnetic ion. Another recently discovered hyperkagome compound is $Na_4Ir_3O_8$.[10] In fact, GGG consists of two interpenetrating lattices of corner-sharing triangles with magnetic exchange mediated via $J_1$, $J_2$, and $J_3$, the near neighbor, adjacent triangle, and adjacent sublattice exchange interactions, respectively,[11] see Fig. 1. In GGG the magnetic $Gd^{3+}$ spins ($S=7/2$) are considered as Heisenberg spins due to single-ion anisotropy of less than 0.04 K (Ref. 12) yet the non-negligible dipole exchange, $D=0.7$ K, could lead to anisotropy.[11] GGG shows a Curie-Weiss temperature of −2.3 K indicative of antiferromagnetic (AF) interactions with no order down to 0.025 K.[11,13] Indications of short-range order were hinted at by bulk measurements[13,14] and proof was obtained by neutron diffraction that revealed a spin liquidlike ground state down to $T=0.14$ K. Additional development of sharper but not resolution limited magnetic diffraction peaks is observed below 0.14 K.[15] True long-range magnetic order is achieved at sufficiently low temperatures via the application of 1 T.[16–18]

The dynamic nature of GGG had been studied via the indirect measurements of $\mu$SR and the more direct technique of Mössbauer spectroscopy. Two $\mu$SR studies confirmed the absence of long-range order down to 0.025 K, however, these studies disagree on the nature of the slowing down of the spin fluctuations. Dunsiger *et al.*[19] found a linear decrease in Gd spin fluctuations below 1 K which extrapolated to 8.2 $\mu$eV at 0 K while Marshall *et al.*[20] observed a temperature-independent relaxation below 0.2 K. Theoretically, a linear temperature dependence of the spin-fluctuation rate has been predicted for classical spins on some frustrated

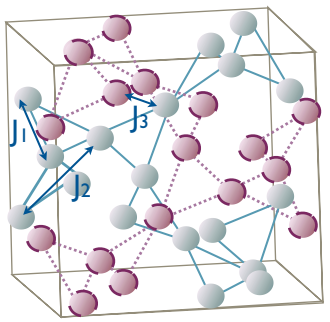


FIG. 1. (Color online) The garnet structure of GGG with two interpenetrating hyperkagome lattices showing the interatomic, intertriangular, and sublattice exchange interactions, $J_1$, $J_2$, and $J_3$, respectively. For clarity only the $Gd^{3+}$ are shown.

 

lattices.[21,22] A Mössbauer spectroscopy study,[23] observed fluctuating Gd spins down to 0.027 K with a decrease in spin fluctuating rate from 11.9 $\mu$eV at 0.4 K to 0.12 $\mu$eV at 0.09 K. Most recently, Ghosh *et al.*[24] pointed to a different dynamical phenomena, in the low-temperature phase below 0.14 K, in which fluctuating uncompensated moments coexist with unsaturated AF order and defect centered clusters.

Theoretically Yavors'kii *et al.*[25] were able to reproduce the spatial correlations of the low-temperature ($T<0.14$ K) phase by taking into account the nearest neighbor, $J_1=0.107$ K,[11] and dipole exchange interactions.[11,13,26] The ordering phenomena are perturbed by much smaller exchange interactions $J_2$ and $J_3$ (Refs. 25 and 26) that dictate the incommensurate ordering wave vector of the low-temperature phase $T<0.14$ K.

This work presents inelastic neutron-scattering study on a hyperkagome structure, GGG, in which we show a spin-liquid state that is, contrary to expectation, concomitant with distinct-gapped modes pointing toward singlet-triplet excitations arising from short-range AF correlations. These results thus shed light on the spin dynamics of frustrated hyperkagome structures.

## II. EXPERIMENTAL DETAILS

Neutron time-of-flight measurements were performed at the spectrometer IN5 of the Institut Laue-Langevin.[27] IN5 was set up to an incident energy of $E_i=1.94$ and 3.27 meV with average elastic linewidths of 50 and 80 $\mu$eV, respectively, full width at half maximum (FWHM). The resolution was determined using a standard incoherent scatterer. The temperature dependence of the scattering function $S(Q,\omega)$ was measured between 0.06 and 9 K with an identical empty cell at 2 K measured as background. The sample used in this work is that used in the previous work of Petrenko *et al.*[15] containing 99.98% of the nonabsorbing isotope $^{160}$Gd. High-resolution neutron diffraction using D1A of the Institut Laue Langevin, $\lambda=1.9$ Å, was used to determine the level of possible disorder on the Ga/Gd sites. The refinement revealed a fully stochiometric sample, the error of the site occupations indicated the upper limit of disorder to be less than 2%.

Figure 2 shows the powder averaged scattering function $S(Q,\omega)$ at 0.06 K. We observe a well-developed elastic line and three inelastic contributions labeled INS1, INS2, and INS3. The wave-vector transfer and energy-dependent powder averaged scattering cross section can be characterized by a $\delta(\omega)$ function with $Q$-dependent intensity for the elastic line and each of the inelastic contributions with a lorentzian form with $Q$-dependent characteristic energy ($\Omega_Q$), linewidth ($\Gamma_Q$), and intensity [$I_i(Q)$]

$$S_{\mathrm{INSi}}(Q,\omega)=I_i(Q)\frac{1}{\pi}\{n(\omega)+1\}F^2(Q)\times\frac{4\omega\omega_Q\Gamma_Q}{(\omega^2-\Omega_Q^2)^2+4\omega_Q^2\Gamma_Q^2}, \quad (1)$$

where $F(Q)$ is the ionic form factor[28] and $\{n(\omega)+1\}$ is the thermal population factor. The dynamic response described by a Lorentzian form corresponds to an exponential decay of

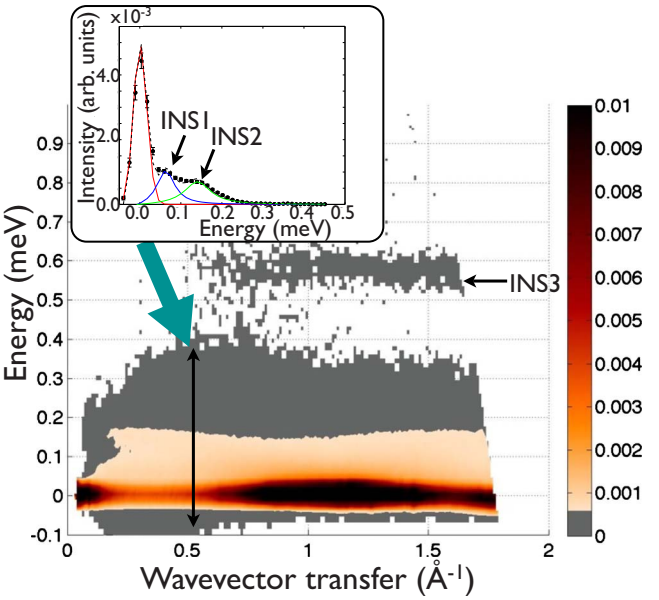


FIG. 2. (Color online) Powder-averaged scattering function $S(Q,\omega)$ of GGG at 0.06 K with incident $E_i=1.94$ meV. Insert is a cut at wave-vector transfer of $Q=0.5$ Å$^{-1}$ showing the elastic lineshape and two low-lying excitations with the corresponding fits as described in the text.

excitations in time, given in terms of a damped harmonic oscillator: $\Omega_Q^2=\omega_Q^2+\Gamma_Q^2$. Equation (1) and the elastic $\delta(\omega)$ are convolved with the instrumental resolution[27] to enable a fit to the data at each position of wave-vector transfer. The data are well described by a Gaussian elastic line, the FWHM of which is fixed by the vanadium standard, and three further inelastic contributions at 0.04(1), 0.14(2), and 0.58(3) meV, see Fig. 2.

The elastic scattering is shown in Fig. 3(a) for 0.06 and 0.25 K. At 0.06 K the elastic part of Eq. (1) represents 82% of the total scattering. This scattering is reminiscent of a spin liquidlike structure factor. In agreement with earlier work,[15] incommensurate Bragg peaks corresponding to longer range correlations develop below 0.14 K [see Fig. 3(b)]. Within the present resolution of the elastic line the spin dynamics observed by $\mu$SR (Refs. 19 and 20) and Mössbauer[23] would appear static. In spite of the onset of longer range correlations we have not observed any sign of associated spin waves at the lowest temperatures. It is possible that these are too weak to be observed since the correlations remain finite on the scale of 100 Å.[15]

The inelastic-scattering reveals the most unusual features. The three-gapped inelastic peaks, INS1, INS2, and INS3 are all dispersionless within the resolution probed. These excitations do not originate from either local vibrational excitations or crystal-field excitations since their dependence on wave-vector transfer neither increases with $|Q|$ nor follows the $Gd^{3+}$ form factor.[28,29] The possibility that the higher energy peak, INS3, is a crystal-field excitation affected by an internal molecular field can be excluded as this is not compatible with the specific-heat data.[11]

The $Q$-dependent powder averaged scattering cross section at 0.06 K of the three inelastic peaks are shown in Fig.

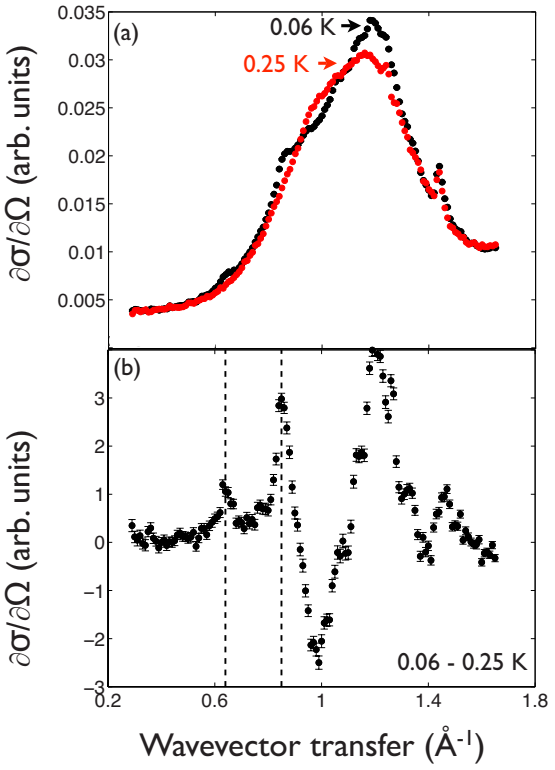


FIG. 3. (Color online) (a) Elastic scattering cross section at 0.06 and 0.025 K. (b) Difference in elastic scattering cross section between 0.06 and 0.25 K. The dashed line corresponds to longer ranged ordered peaks observed by Petrenko *et al.*(Ref. 15).

4(a) and inset. The INS3 reveals spatial correlations corresponding to that expected from a singlet-triplet excitation of a dimerized state[30] and can be modeled using the following powder averaged formula:

$$I(Q) \propto A(T)\left[1 - \frac{\sin(Qd)}{Qd}\right], \tag{2}$$

where $d$ is the separation between spins and $A(T)$ is a temperature-scaling factor linked to the canonical partition function proportional to the thermal distribution of the singlet ground state and the triplet excited state, $A(T)=1/[1+3\exp(-J_{NN}/k_BT)]$. Here $J_{NN}$ is the nearest-neighbor exchange energy $J_{NN}=J_1S(S+1)=1.68$ K.[14,30] In this case, a dimer can be understood as short-range order of AF coupled Gd spins within a cluster effectively shielded from its neighboring cluster. Magnetic interactions between clusters therefore, can be neglected.

The dashed lines in Fig. 4(a) correspond to the neutron scattering of Gd dimers with near-neighbor distance $d=3.7915$ Å, as described in Eq. (2). Clearly the INS3 data are well described by such a near neighbor model. Interestingly the energy FWHM of INS3, $\Gamma$, is not resolution limited with $\Delta E=0.05(1)$ meV at 0.06 K, corrected for instrumental resolution. The excitation lifetime is inversely proportional to $\Gamma$, $\tau=\Gamma^{-1}$, thus indicating that the Gd dimers are short lived.

Figure 4(a) inset shows the integrated intensity of the wave-vector transfer for INS1 and INS2. The $Q$ dependence

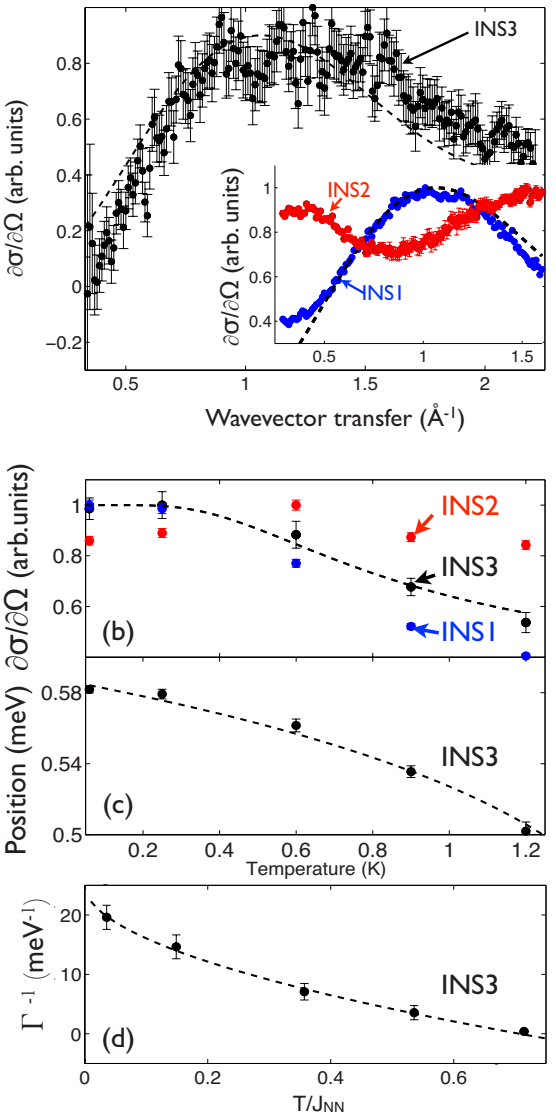


FIG. 4. (Color online) (a) $Q$-dependent integrated intensity of INS3. The dashed lines correspond to a model of short-range AF correlation with only near neighbor interactions. The inset shows the integrated intensities of INS1 and INS2. (b) Temperature dependence of the integrated intensity for INS1, INS2, and INS3. The dashed line is the temperature dependence expected for a singlet to triplet excitation with an exchange interaction $J_{NN}=J_1S(S+1)=1.68$ K and follows closely the integrated intensity of INS3. (c) Peak position in energy of INS3 [dashed line is a fit to the data with a power law with the peak position falling into the elastic line at $T=1.67(2)$ K]. (d) $T/J_{NN}$ dependence of the excitation lifetime $\Gamma^{-1}$ of INS3. Dashed line is the algebraic function $\tau=\mathcal{A}T^{\xi}$ with an exponent $\xi=0.53\pm0.13$.

of the integrated intensity of INS1 follows closely the behavior of INS3, the dashed line represents the line shape of Eq. (2). However, at low and high wave-vector transfer the model of short-range correlations fails indicating that extra terms remain important for a full description of the ground state. The integrated intensity of INS2 shows a minimum close to the position in reciprocal space that corresponds to nearest-neighbor interactions indicating that the origin of INS2 is very different to that of INS1 and INS3.

The temperature dependence of the normalized-integrated intensities of INS1, INS2, and INS3 are shown in Fig. 4(b). The dashed line represents the thermal behavior expected from a singlet-triplet excitation, $A(T)$ from Eq. (2) with an exchange interaction $J_{NN}=1.68$ K. The dashed line follows closely the integrated intensity of INS3 thus further validating the notion of a dimerized short-range AF ordered state. However, neither INS1 nor INS2 follow the temperature dependence of INS3. The integrated intensity of INS1 follows a trend similar to INS3, albeit with a reduced exchange interaction $J=1.3$ K, up to 0.6 K but at higher temperatures does not follow this trend. The integrated intensity of INS2 has a maximum at 0.6 K.

Further information concerning the INS3 excitation is revealed in Fig. 4(c). Unlike INS1 and INS2, INS3 shows a strong temperature dependence in its energy position and the excitation lifetime. The peak energy position can be followed by a power-law function with parameters $\beta=0.12(1)$ and falls to zero at $T=1.67(2)$ K. The excitation lifetime is not linear as observed in certain frustrated compounds[9] but can instead be described by the algebraic form $\tau=\mathcal{A}T^{\xi}$ with $\xi=0.53\pm 0.13$, Fig. 4(d). The relevance of these parameters becomes clear when reviewing recent theoretical work by Robert *et al.*[31] showing that, in contrast to the pyrochlore lattice,[21,22,32] sufficient temporal and spatial stiffness in a classical kagome antiferromagnet can give rise to magnetic excitations corresponding to acoustic and optical modes in addition to a soft mode. The temperature dependence of the lifetime of the excitations, $\tau$, would follow an algebraic dependence $\tau=\mathcal{A}T^{\xi}$ with $\xi=0.18$ for inelastic scattering in the regime of co-operative paramagnetism, $T/J_{NN}\leq 0.1$. In addition, Zhitomirsky indicates that such temporal and spatial stiffness is required to develop multipolar ordered states at low temperatures in both kagome and hyperkagome structures.[33] The excitations observed in this work cannot be assigned to acoustic excitations as these would remain dispersive and originate from Bragg peaks, even with powder averaging. It is possible to assign the nondispersive-gapped excitations to optical or soft modes with a high-energy mode at $\omega=0.58$ meV $\sim 5.4J_1$ and not $2J_1$ as predicted.[31] Robert *et al.* calculated the excitations for a classical Heisenberg kagome AF. It is well-known that substantial long-range dipole exchange interactions play an important role, such a mode would therefore, be lifted upwards and may display an alternative temperature dependence of the fluctuation rate. Analogous to phonons, a magnetic optical mode can arise from a localized perturbation of interactions as found in the short-range dimerized interactions displayed by INS1 and INS3.

The data presented in this work sheds light on the unusual magnetic ground state of the hyperkagome structure GGG. These results reveal that magnetic order in GGG corresponds to remarkably different time scales leading to the simultaneous development of short-range ordered antiferromagnetic dimerized order with milli-electron-volt ($10^{-3}$) dynamics (this work), co-operative paramagnetism with $\mu$eV ($10^{-6}$) dynamics,[23] protected spin clusters with pico-electron volt ($10^{-12}$) dynamics[24] and static order. This is highly unusual in a compound in which a continuum of excitations is expected to be characteristic of the dynamic nature of the magnetic ground state.

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