Spin Hamiltonian, Competing Small Energy Scales and Incommensurate Long Range Order in the Highly Frustrated Gd$_3$Ga$_5$O$_{12}$ Garnet Antiferromagnet

Taras Yavors’kii, Matthew Enjalran, and Michel J.P. Gingras

$^1$ Department of Physics and Astronomy, University of Canterbury, Private Bag 4800, Christchurch, New Zealand
$^2$ Department of Physics, Southern Connecticut State University, New Haven, CT 06515, USA
$^3$ Department of Physics and Astronomy, University of Canterbury, Private Bag 4800, Christchurch, New Zealand

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Despite the availability of a spin Hamiltonian for the Gd$_3$Ga$_5$O$_{12}$ garnet (GGG) for over twenty-five years, there has so far been little theoretical insight regarding the many unusual low temperature properties of GGG. Here we investigate GGG in zero magnetic field using mean-field theory. We reproduce the spin liquid-like correlations and, most importantly, explain the positions of the sharp peaks seen in powder neutron diffraction experiments. We show that it is crucial to treat accurately the long-range nature of the magnetic dipolar interactions to allow for a determination of the small exchange energy scales involved in the selection of the experimental ordering wave vector. Our results show that the incommensurate order in GGG is classical in nature, intrinsic to the microscopic spin Hamiltonian and not caused by weak disorder.

The diversity of empirical data collected over the past fifteen years has demonstrated that geometrically frustrated triangular and tetrahedral arrangements of antiferromagnetically coupled spins are highly partial towards the realization of exotic correlated phases in magnetic materials $^1$ $^2$ $^3$. The reason for the rich and typically material specific properties of frustrated magnets is understood. It stems from their sensitivity to perturbations beyond the frustrating nearest-neighbor antiferromagnetic (AFM) exchange which, on its own, leads to a macroscopic number of exactly degenerate and competing, hence fragile, classical ground states. In this paper we show, through a careful theoretical analysis of neutron scattering experiments, that the extensively studied Gd$_3$Ga$_5$O$_{12}$ garnet (GGG) is precisely such a system, though evidence for this fact emerges from a perspective on the problem that has heretofore escaped scrutiny.

GGG displays a gamut of complex and interesting low temperature magnetic phenomena. In zero magnetic field, the behavior of GGG is uniquely rich. The nonlinear magnetic susceptibility $\chi_3$ peaks at $T_g \sim 180 \text{ mK}$ $^3$, indicating a spin glass transition $^5$. However, muon spin relaxation $^6$ $^7$ and Mössbauer spectroscopy $^8$ find persistent spin dynamics down to $T \ll T_g$. Meanwhile, powder neutron scattering data $^9$ $^{10}$ indicate that GGG is on the verge of developing true incommensurate long-range magnetic order with a correlation length ($\xi \approx 100 \text{ Å}$) extending over 8 cubic unit cells below 140 mK.

A Hamiltonian $\mathcal{H}$ describing GGG, that we shall explicitly define below, has long been available $^{11}$. It assumes classical Gd$^{3+}$ spins, is parameterized as a sum of empirical exchange contributions up to third nearest-neighbors as well as a magnetic dipolar contribution, and ignores potentially important quantum fluctuations or disorder inherent to GGG $^4$. Previous numerical studies based on $\mathcal{H}$ have been unable to provide a quantitative explanation for the bulk $^4$ and dynamical $^6$ $^7$ $^8$ properties of GGG or the incommensurate spin-spin correlations that develop below 200 mK $^3$ $^{10}$. This could be interpreted as evidence that exotic mechanisms involving either quantum fluctuations or disorder effects are at play in GGG. For example, it has been suggested that disorder, such as Gd$^{3+}$ on Ga$^{3+}$ sites, is responsible for nucleating regions with the spin order observed in neutron scattering in zero magnetic field $^3$ $^{10}$.

In this letter we address the adequacy of the Hamiltonian $\mathcal{H}$ of Ref. $^{11}$ to explain the low temperature properties of GGG. It has been demonstrated $^9$ $^{12}$ that $\mathcal{H}$ with only a nearest-neighbor exchange interaction can describe liquid-like spin correlations $^9$ $^{10}$. We show below that $\mathcal{H}$ can also describe well the incommensurate spin-spin correlations and that there is no obvious reason to extend $\mathcal{H}$. Rather, it is the correct procedure to treat its many competing small energy scales, in particular the long range dipolar interactions, and the unusually precise specification of weak second and third nearest-neighbor exchange couplings, that are foremost needed, and that have been missed so far.

In order to correctly parameterize $\mathcal{H}$ and the resulting spin correlations we rely on neutron scattering experiments $^9$ $^{10}$, which are the most direct and unambiguous probe of magnetic correlations. Our basic hypothesis is that conventional criticality controls the development of the sharp peaks observed in neutron experiments $^9$ $^{10}$, and that, as in conventional magnets, a mean-field theory (MFT) treatment of the pertinent Hamiltonian should be able to capture the Gaussian regime in the approach to criticality. Our finding of a unique ordering wave vector $q_{ord}$ shows that GGG, unlike the AFM nearest-neighbor Heisenberg pyrochlore, is not pathological and is amenable to such a soft-mode determination and calculation of the neutron scattering intensity profile. We show that GGG is very similar to the now well understood dipolar spin ice materials $^{13}$ $^{14}$, where a MFT that takes into account the long-range nature of the dipolar interaction $^{13}$ (i) identifies the correct ordering wave...
vector \[14\], and (ii) correctly predicts the symmetry of the scattering close to the critical temperature.

We examine the spin correlations in GGG by calculating the powder neutron scattering intensity \(I(Q)\) and comparing it with experimental data \[4\]. The scattering \(q\)-vector-dependent intensity is given by

\[ I(q) = \frac{F(q)^2}{N} \sum_{ij} \langle \hat{S}_i \cdot \hat{S}_j \rangle e^{iq \cdot \mathbf{r}_{ij}}, \]

obtained for the model Hamiltonian \[11\]

\[ \mathcal{H} = \mathcal{H}_c + \mathcal{H}_{\text{dip}} \]

\[ \mathcal{H}_c = \sum_{i>j} \{ \sum_{k=1}^{4} J_3 \delta_{\mathbf{r}_{ij}, \mathbf{r}_k} \mathbf{S}_i \cdot \mathbf{S}_j \} \quad \text{and} \quad \mathcal{H}_{\text{dip}} = \sum_{i>j} \{ D(\mathbf{r}_1/\mathbf{r}_{ij})^3 \} \mathbf{S}_i \cdot \mathbf{S}_j - 3 \langle \mathbf{S}_i \cdot \mathbf{r}_{ij} \rangle \langle \mathbf{S}_j \cdot \mathbf{r}_{ij} \rangle \].

The scattering \(q\)-space grid with 32\(^3\) points in the first Brillouin zone, and construct a three dimensional cubic interpolation scheme to separately evaluate the numerator and the denominator of the MFT \(I(q)\) (cf. Eq. A37 in Ref. \[16\]). For \(q \approx q_{\text{ord}} + \mathbf{G}\), where Bragg peaks occur (\(G\) is a FCC reciprocal lattice vector), we back up the interpolation by calculating \(I(q)\) exactly \[16\]. This ensures \(q\)-grid size independent results \[16\]. \(I(Q)\) is computed by spherically averaging \(I(q)\) numerically.

We find that \(\mathcal{H}\) with dipolar interactions and exchange interactions beyond nearest-neighbor possesses a unique \(q_{\text{ord}}\). Our numerical study suggests that the soft mode selection in the full \(\mathcal{H}\) is highly sensitive to the values of \(J_2\) and \(J_3\). This finding is in general agreement with results from Monte Carlo simulations of GGG \[9,10\] (see also Ref. \[11\] for a comment on the sensitivity of the GGG specific heat on \(J_3\)). However, our crucial new observation is that \(q_{\text{ord}}\) is also very sensitive to an ad-hoc choice of the cut-off distance \(R_c\) in \(\mathcal{H}_{\text{dip}}\). We demonstrate this by plotting in Fig. 1 \(I(Q)\) for various \(R_c\) for \(J_2 = -0.003\) K and \(J_3 = 0.010\) K (values from \[11\]). For \(R_c \lesssim 10^5\), not only is \(q_{\text{ord}}\) a non-monotonous function of \(R_c\), as the positions of the peaks indicate, but so is the unit cell magnetic form factor, as is evident from the significant changes in the overall shape of the diffuse scattering patterns near criticality \((\tau \approx 1)\). Fig. 1 demonstrates that \(\mathcal{H}\) with different low \(R_c\) gives completely different spin-spin correlations in the critical regime. Only for rather large cut-offs does \(q_{\text{ord}}\) become insensitive to \(R_c\), approaching the \(R_c = \infty\) limit only when \(R_c \gtrsim 10^3\). Figure 1 suggests that it is essential to set \(R_c\) to its true \(R_c = \infty\) limit to avoid inducing spurious ordered phases by treating the lattice sum in \(\mathcal{H}_{\text{dip}}\) by a short cut-off method, as was done previously \[9,10,11,12,17\]. We henceforth acknowledge this and implement the Ewald method \[16\] to process \(\mathcal{H}_{\text{dip}}\) with \(R_c = \infty\).

The scattering profiles in Fig. 1 and the \(q\) positions of the sharp peaks, even in the \(R_c = \infty\) limit, are incompatible with the experimental data \[9\] (cf. Fig. 3, upper panel). However, the uncertainty in previously estimated values of \(J_2\) and \(J_3\) \[11\] allows fine-tuning them in order to obtain a better match with the low temperature data \[9\]. To proceed, we are guided by the general understanding of disordered systems near the critical boundary between long-range ordered and spin glass phases \[3\]. Specifically, we make the reasonable assumption that,
given the sizeable and saturated $\xi \sim 100$ Å correlated regions below 140 mK \cite{9}, the weak disorder in GGG freezes-in the $q$-dependence of quasi-critical incommensurate correlated regions as $T_J$ is crossed upon cooling. We therefore use a two-step fitting procedure to determine $J_2$ and $J_3$. First, we establish whether the model $\mathcal{H}$ allows for a $q_{\text{ord}}$ compatible with the experiment. Second, we examine whether the model with a given $\{J_2, J_3\}$ set is able to reproduce the overall structure of correlations both in the paramagnetic and “frozen-in critical” regimes \cite{9}.

We find that $q_{\text{ord}}$ for $T_c = \infty$ belongs to the (hhl) reciprocal plane, with its location within the plane highly dependent on $J_2$ and $J_3$. To determine the optimum $J_2$ and $J_3$, we generate a sequence of data points $\{\tilde{Q}\} \equiv \{q_{\text{ord}}(J_2, J_3) + \mathbf{G}\}$, and then match these points to the experimental sequence of sharp peaks in powder GGG data \cite{9}. To do so, we introduce a penalty function, $\mathcal{P}(J_2, J_3)$, that provides a measure of mismatch between the first three clearly discernible and strongest experimental peaks at $Q_1^* = 0.64$ Å$^{-1}$, $Q_2^* = 0.85$ Å$^{-1}$ and $Q_3^* = 1.97$ Å$^{-1}$ of Ref. \cite{9} (see 43mK experimental data in top panel of Fig. \ref{fig:3}) and the closest theoretically determined peaks: $\mathcal{P} = 100 \times \max_{i=1,2,3} \Delta Q_i$, where $\Delta Q_i = \min_{\tilde{Q} \in \{\tilde{Q}\}} |\tilde{Q} - Q_i^*|$ with the numerical factor 100 chosen to set the scale, $\mathcal{P} = 1$, for a maximum mismatch of 0.01Å$^{-1}$. A map of $\mathcal{P}$ in the $J_2 - J_3$ plane is shown in Fig. \ref{fig:2}. A good match between theory and experiment is denoted in black. The discrepancy between experiment and theory increases as the gradation of gray varies from black to white.

Figure \ref{fig:2} shows that even small (at the $\approx 1\%$ level of $J_1$) changes in $J_2$ and $J_3$ show up as large changes in the ordering wave vector and, correspondingly, in $\mathcal{P}$ too. There exist only relatively small domains (shown in black) that minimize $\mathcal{P}$. It turns out, however, that for each of these domains, the overall shape of the MFT powder scattering profile changes noticeably even though $q_{\text{ord}}$ remains almost constant at the value of $2\pi/a \approx 0.29, 0.29, 0$. Similarly to the standard experimental procedure to analyze powder neutron scattering data, once $q_{\text{ord}}$ is determined, one needs to solve for the magnetic structure. The sensitivity of the unit cell magnetic form factor to $\{J_2, J_3\}$ leads to a significant qualitative change in the diffuse scattering pattern near criticality, hence offering a second optimization channel for the determination of $\{J_2, J_3\}$.

Figure \ref{fig:3} shows the dependence of the theoretical $I(Q)$ on $\{J_2, J_3\}$ within the black domains of Fig. \ref{fig:2}. A comparison of such theoretical profiles with the experimental one reveals that only in the upper left black domain are the theoretical and experimental profiles similar; a strong discrepancy in other domains allows us to exclude them from further consideration. Within the upper left domain, the match is visually better for rightmost values of $J_2$ with the restriction that its value cannot cross the domain boundary at $-0.004$K.

We support and further refine the above analysis by fitting the MFT scattering profiles to the experimental $I(Q)$ in the paramagnetic regime. For each value of $J_2$ and $J_3$ we consider a $\chi^2$-type penalty function for the overall shape of $I(Q)$, which is adjusted by parameters responsible for a uniform and linear in $|Q|$ background terms, as well as by the overall scale factor. With $\tau$ as a free fitting parameter, we do the fits in the interval
To conclude, we have found that, as in spin ices \[1,13\] [14], an ad-hoc $R_c$ cut-off of the dipolar interactions at less than a few hundred nearest-neighbor distances leads to spurious long-range ordered phases. Only a proper treatment of an infinite $R_c$ allows one to get a handle on the very small exchange interactions beyond nearest neighbors which dictate the incommensurate ordering wave vector $q_{\text{ord}}$. With a reasonably well parameterized Hamiltonian now in hand, further theoretical studies to explore the origins of the complex phenomena displayed by GGG in zero and nonzero magnetic field may now be possible. Perhaps most importantly, we have identified the likely reciprocal plane that contains $q_{\text{ord}}$, with $q_{\text{ord}} \approx 2\pi/a [0.29, 0.29, 0]$. With this prediction available, it may now be possible to perform single crystal neutron scattering measurements in a reflection geometry to beat the $^{157}$Gd absorption problem in high quality single crystals with natural Gd abundance \[18\], and allow for a quantitative investigation of the development of correlations in GGG at low temperatures. This may prove a fruitful endeavor to help shed light on the mysteries of GGG.

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\[Q = 0.26 - 1.57 \text{Å}^{-1}\] which allows for the complete shape determination of the first experimental broad diffusive scattering peak at $1.05 \text{Å}^{-1}$ without a $|Q|^2$ background correction. Most importantly, an analysis of the (paramagnetic) data at 175 mK \[6\] allows us to confirm the outcome of the analysis of the critical data at 43 mK and reject all but the aforementioned upper left domain of Fig. 2. Within that domain the data impose conservative error bars on $J_2$: $-0.012 \leq J_2 \leq -0.004$ K, with limits on $J_3$ set by that same domain, i.e. $-0.003 \leq J_3 \leq 0.012$ K, see Fig. 2.

The MFT $I(Q)$ with the optimized $J_n$ values captures the features of the experimental $I(Q)$ well (see bottom panel of Fig. 2). The optimization procedure to match the three strongest experimental peaks leads to a theoretical sequence of peaks that can be identified on the experimental profile as rather well-distinguished peaks and cusps. The match (Fig. 3) between MFT and experimental $I(Q)$ suggests post-factum that the $q_{\text{ord}}$ dependence of the correlations can be described by MFT.

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