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Ill-behaved convergence of a model of the $Gd_3Ga_5O_{12}$ garnet antiferromagnet with truncated magnetic dipole–dipole interactions

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Abstract

Previous studies have found that calculations which consider long-range magnetic dipolar interactions truncated at a finite cut-off distance R_c predict spurious (unphysical) long-range ordered phases for Ising and Heisenberg systems on the pyrochlore lattice. In this paper we show that, similar to these two cases, calculations that use truncated dipolar interactions to model the $Gd_3Ga_5O_{12}$ garnet antiferromagnet also predict unphysical phases with incommensurate ordering wavevector $\mathbf{q}_{ord}$ that is very sensitive to the dipolar cut-off distance R_c.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

There are currently many highly frustrated magnetic materials being experimentally studied where the magnetic species consist of a rare-earth 4f ion, such as Ho^{3+}, Dy^{3+}, Gd^{3+} or Tb^{3+}, which can have a large magnetic dipole moment. Because of the large moment, the long-range dipole–dipole interactions in these systems are an important part of the full spin Hamiltonian. The role of dipolar interactions in highly frustrated magnetic Ising systems has been systematically investigated for the three-dimensional pyrochlore lattice of corner-sharing tetrahedra [1–3].

In highly frustrated Heisenberg antiferromagnets of corner-sharing triangles or tetrahedra, any state with zero total magnetic moment on each elementary triangle or tetrahedral unit is a classical ground state [4]. There are an infinite number of such spin configurations and this is why these systems fail to develop conventional magnetic order at nonzero temperature [4]. In cases where dipolar interactions are somewhat weaker than nearest-neighbour exchange interactions, one might have naively assumed that the long-range dipolar interactions can be

truncated at a finite cut-off distance, R_c, since the nearest-neighbour exchange energetically controls and enforces the nearest-neighbour correlations. Previous studies on the Ising spin ice pyrochlore systems [1–3] and the Heisenberg pyrochlore antiferromagnet [5, 6] have found that this naive expectation is erroneous and that truncating the dipolar interactions at a finite cut-off distance R_c leads to spurious (unphysical) long-range ordered phases, and that it is crucial to consider dipolar interactions to infinite distance ($R_c = \infty$). In this paper we report a third example of a highly frustrated spin system which is very sensitive to the dipolar cut-off. Specifically, we consider a Heisenberg model on the three-dimensional garnet lattice structure of corner-shared triangles. Our work extends the study of a dipolar Ising version of the model on a garnet lattice [7] and is relevant to the ultimate understanding of the nature of the incommensurate spin–spin correlations that develop in the $Gd_3Ga_5O_{12}$ garnet (GGG) antiferromagnet below a temperature of 500 mK [8, 9].

2. Model and method

In order to investigate the problem of an adequate treatment of the dipolar interactions in GGG we consider below a minimal model Hamiltonian H for it. To best expose the physics of a truncated dipolar lattice sum, we ignore the effects of the quantum nature of the Gd^{3+} spins, lattice disorder [8], exchange interactions beyond nearest neighbours [8, 10] and possible single ion anisotropy [12], all of which are potentially important for a thorough quantitative understanding of GGG. We describe the magnetic Gd^{3+} spins $\mathbf{S}$ as classical and isotropic $n = 3$ component (Heisenberg) vectors of length $\sqrt{(S(S+1))}$ ($S = 7/2$) coupled by frustrated antiferromagnetic nearest-neighbour exchange and long-range dipolar interactions, of strength $J = 0.107$ K and $D = 0.0457$ K, respectively [10, 11]:

$$H = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + D \sum_{i>j} \frac{1}{r_{ij}^3} \left[\mathbf{S}_i \cdot \mathbf{S}_j - 3(\mathbf{S}_i \cdot \hat{r}_{ij})(\mathbf{S}_j \cdot \hat{r}_{ij}) \right]. \tag{1}$$

In equation (1), i, j span the sites of the GGG lattice (see figure 2 in [10] for the GGG lattice structure) which are separated by vectors $\mathbf{r}_{ij} \equiv r_{ij}\hat{r}_{ij}$ of directions $\hat{r}_{ij}$; $\langle i, j \rangle$ denotes pairs of nearest neighbours. The size of the GGG conventional cubic cell is $a = 12.349$ Å [8].

We aim to identify the critical (or, soft) modes of model (1) for which a magnetic instability first develops as the temperature T is reduced. We consider the soft mode spectrum in the Gaussian (mean-field theory, or MFT) approximation [13]. Following reference [13], we apply MFT to calculate the neutron scattering intensity $I(\mathbf{q})$. This allows for a convenient way of analysing the physical influence of finite R_c on the magnetic correlations as well as a direct comparison with experimental data [8]:

$$I(\mathbf{q}) = [f(|\mathbf{q}|)]^2 \times \lim_{N\to\infty} 1/N \sum_{ij} \langle \mathbf{S}_i^{\perp} \cdot \mathbf{S}_j^{\perp} \rangle e^{i\mathbf{q}\cdot\mathbf{r}_{ij}}. \tag{2}$$

Here, angular brackets denote a thermal average, N is the number of spins, $\mathbf{S}_i^{\perp}$ represents the components of spin $\mathbf{S}_i$ at site i perpendicular to the scattering vector $\mathbf{q}$ and $f(|\mathbf{q}|)$ is the magnetic form-factor of Gd^{3+} [14]. The MFT expression for $I(\mathbf{q})$ [13] is obtained from the eigenvalues $\lambda^{\alpha}(\mathbf{q})$ and eigenvectors of the basis-diagonalization of the Fourier transform of exchange and dipolar interactions in equation (1) [13]:

$$I(\mathbf{q}) = [f(|\mathbf{q}|)]^2 \sum_{\alpha} \frac{|\mathbf{F}_{\perp}^{\alpha}(\mathbf{q})|^2}{(n - \lambda^{\alpha}(\mathbf{q})/T)}. \tag{3}$$

Here $\alpha = \{1, \ldots, n \cdot N_b = 36\}$ enumerates the eigenvalues and the vector $\mathbf{F}_{\perp}^{\alpha}(\mathbf{q})$ incorporates information on the eigenvectors and represents the role of the paramagnetic form factor of

the GGG primitive unit cell, which contains $N_b = 12$ ions. The ordering wavevector $\mathbf{q}_{\rm ord}$ is given by locating in the first Brillouin zone the global maximum, $\lambda_{\rm max}$, of the maximum value (upper branch), $\lambda^{\rm up}(\mathbf{q})$, among the 36 $\lambda^{\alpha}(\mathbf{q})$ eigenvalues. The mean field critical temperature, $T_{\rm c}^{\rm MFT} = \lambda_{\rm max}/n$, is used to define a (positive) dimensionless temperature $\tau = T/T_{\rm c}^{\rm MFT} - 1$ to serve as a natural temperature scale.

3. Results

With the aim of studying the effect of the dipolar cut-off on the magnetic correlations of model (1), we first compute, for various $R_{\rm c}$, $\lambda^{\rm up}(\mathbf{q})$ for arbitrary $\mathbf{q}$ in the first Brillouin zone. Technically, we calculate the $\lambda^{\alpha}(\mathbf{q})$ modes on a finite 32^3 $\mathbf{q}$-space grid in the zone, and obtain their values at any $\mathbf{q}$ using a three-dimensional cubic interpolation procedure. We verify the grid independence of the results by considering denser grids and by cross-checking the interpolated results with exact calculations at judiciously chosen $\mathbf{q}$ values.

We find that the dipolar term of equation (1) selects a unique ordering wavevector $\mathbf{q}_{\rm ord}$ with corresponding mode $\lambda^{\rm up}(\mathbf{q}_{\rm ord})$ out of the massively degenerate spectrum of soft modes of the nearest-neighbour model at any cut-off distance $R_{\rm c} > 1$. The ordering wavevector $\mathbf{q}_{\rm ord}$ was found to belong to the (hhl) planes of the first Brillouin zone for all $R_{\rm c} > 1$. However, its location in those planes is very sensitive to $R_{\rm c}$. We display this in figure 1 by showing the dependence of the $\lambda^{\rm up}(\mathbf{q})$ modes on $\mathbf{q}$ in the (hhl) plane for $R_{\rm c} = 3, 4, 5, 100, 1000$. We note their three important properties as reflected in figure 1. First, for each value of $R_{\rm c}$, $\lambda^{\rm up}(\mathbf{q})$ is characterized by a relatively small overall dispersion $\lambda_{\rm max}/\lambda_{\rm min} - 1 \approx 10\%$ throughout the zone, where $\lambda_{\rm min}$ is the global minimum of the branch. Even within the interval 1% below $\lambda_{\rm max}$ (or at 13% of the overall dispersion, as delineated by the outermost isolines), $\lambda^{\rm up}(\mathbf{q})$ covers the major part of the (hhl) plane. Second, though the general topology of $\lambda^{\rm up}(\mathbf{q})$ within the Brillouin zone is preserved with varying dipolar cut-off distances $R_{\rm c}$, its fine structure is manifestly sensitive to $R_{\rm c}$. The ordering wavevector $\mathbf{q}_{\rm ord}$ displays a non-monotonic dependence upon $R_{\rm c}$, as shown in figure 1 by its movement throughout the zone. Third, $\mathbf{q}_{\rm ord}$ converges to a well-defined value, and so does the dispersion of $\lambda^{\rm up}(\mathbf{q})$ away from $\mathbf{q}_{\rm ord}$ at large values of $R_{\rm c}$, as is evident by comparing the $R_{\rm c} = 1000$ panel with the limiting case of $R_{\rm c} = \infty$, recast by the Ewald method [13]. These are the central results of the paper.

We now proceed to calculate the neutron scattering intensity $I(\mathbf{q})$ of model (1) within the MFT scheme, equation (3). Unlike the spectra $\lambda^{\alpha}(\mathbf{q})$, $I(\mathbf{q})$ can be directly compared to experiments, such as the one on a powder sample of GGG in zero external magnetic field [8]. We determine the powder intensity $I(q)$ by numerically calculating the spherical average of $I(\mathbf{q})$, which entails the separate application of a cubic interpolation procedure to the numerator and denominator of equation (3). To illustrate the influence of $R_{\rm c}$ on the spin–spin correlations of model (1), we show in figure 2 the MFT powder scattering profiles $I(q)$ of model (1) at several $R_{\rm c} = 3, 10, 100, \infty$.

The $I(q)$ profiles show different degrees of dependence on the dipolar cut-off distance $R_{\rm c}$ at different dimensionless MFT temperatures τ (figure 2). At temperatures sufficiently far from the mean-field critical regime (figure 2, $\tau \geqslant 0.1$), the magnetic correlations are not very sensitive to $R_{\rm c}$. In fact, it has been found in Monte Carlo simulations [8] that paramagnetic liquid-like correlations of GGG can be well described by completely ignoring the dipolar term. At $\tau \approx 0.01$ the profiles start to capture the effect of the dipolar interactions on the magnetic correlations. The effect turns out to depend on $R_{\rm c}$. Indeed, at $\tau \approx 0.001$[4] the profiles clearly display a specific q-dependence sensitive to the chosen $R_{\rm c}$.

[4] The powder MFT Bragg intensities grow as $\log|\tau|$ as opposed to the $1/\tau$ growth of the $\mathbf{q}$-dependent intensities. This explains the need to consider rather small τ in order to theoretically approach the critical regime.

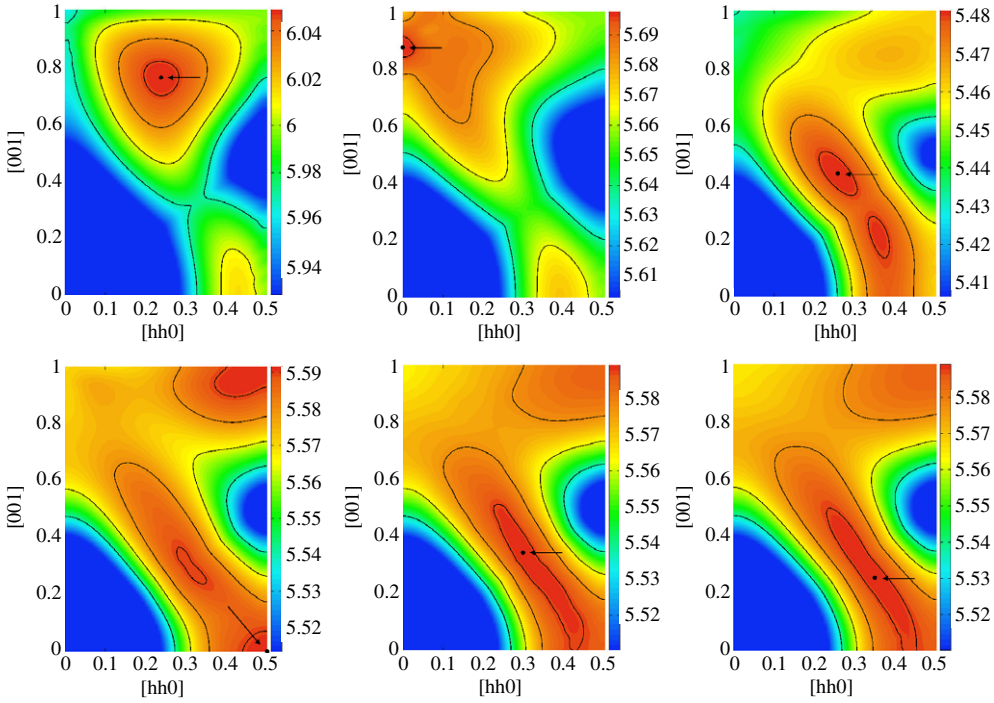

Figure 1. Each panel shows the upper branch $\lambda^{\mathrm{up}}(\mathbf{q})$ of eigenvalues of model (1) as a function of wavevector $\mathbf{q}$ in the (*hhl*) plane at various cut-off distances R_c: $R_c = 3, 4, 5$ (upper panels, left to right) and $R_c = 100, 1000, \infty$ (lower panels). The global maximum $\lambda_{\max}$ defines the ordering wavevector $\mathbf{q}_{\mathrm{ord}}$ (denoted by an arrow). Its location is a complex function of R_c. The isolines are drawn at 0.5%, 3%, 7%, 13%, of the overall dispersion downhill from the global maximum; the actual values of the dispersions as well as of $\lambda_{\max}$ depend on R_c ($\mathbf{q}$ is measured in units of $2\pi/a$).

Starting from this regime, the influence of small R_c becomes uncontrollable, as seen by the formation of Bragg peaks at spurious ordering wavevectors (cf figure 1). Figure 2 shows that even the consideration of a rather large dipolar cut-off of $R_c = 100$ does not allow one to reproduce magnetic correlations consistent with the physical $R_c = \infty$ limit. This, together with the incommensurability of the fundamental ordering wavevector of the physical $R_c = \infty$ case, $\mathbf{q}_{\mathrm{ord}} = 2\pi/a$ (0.348 0.348 0.253), strongly warns against using a standard Monte Carlo method with periodic boundary conditions to tackle this problem. Moreover, we anticipate that a $1/L^3$ finite-size correction of the real space representation of the Ewald interactions used to recast the dipolar lattice sums would be sufficiently large for numerically accessible system sizes so as to prohibit a quantitative disentanglement of the role of perturbative terms [8, 11, 12] to model (1), presumably a necessary condition for obtaining a quantitative description of the experimental incommensurate magnetic correlations [8] in GGG.

4. Conclusion

To conclude, we have identified another example of a highly frustrated Heisenberg antiferromagnetic system, namely that on the garnet lattice, where the selection of the soft mode is sensitive to an *ad hoc* cut-off distance R_c of the dipolar interactions. This adds to the

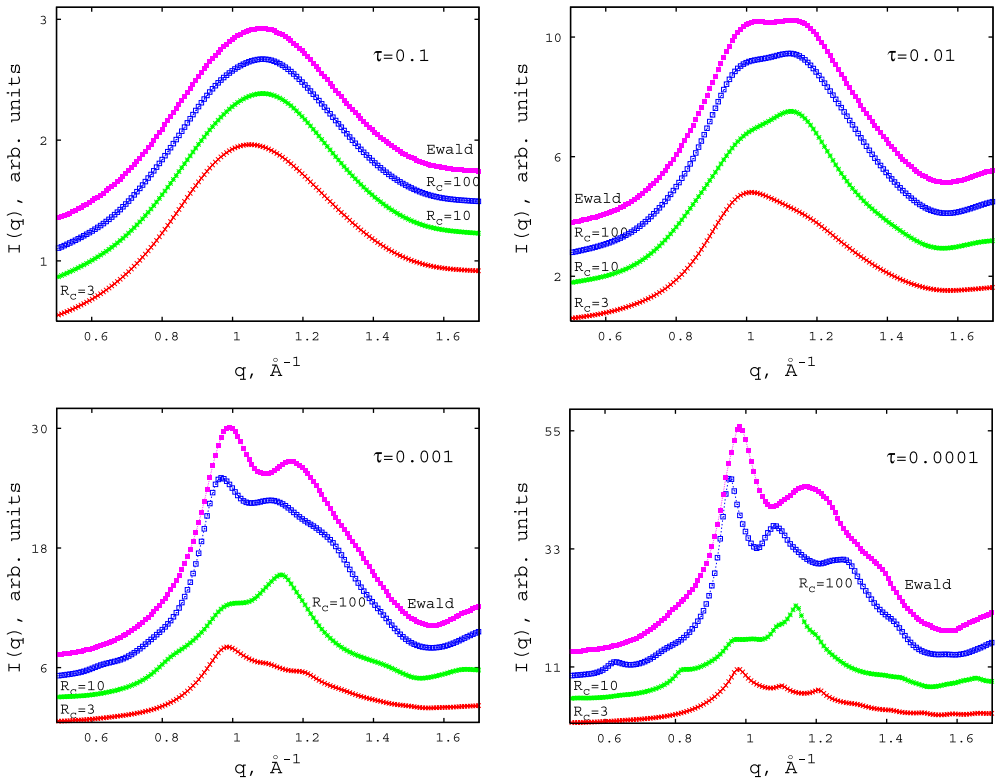

Figure 2. Theoretical powder neutron scattering profiles of model (1) at the dipolar cut-off distances $R_c = 3, 10, 100, \infty$ and the dimensionless temperatures $\tau = 0.1, 0.01, 0.001, 0.0001$. At $\tau = 0.1$ the profiles are not sensitive to the cut-off distance and in fact reflect the properties of the nearest-neighbour Heisenberg model without dipolar interactions. With the approach to the critical temperature the profiles acquire dispersions that are unique reflections of different ordering tendencies of model (1) at different R_c. The profiles are uniformly off-set for clarity.

cases of the Ising (spin ice) [1–3] and the dipolar Heisenberg antiferromagnets [5, 6], both on the pyrochlore lattice. Continued progress in understanding GGG may be possible provided the dipolar interactions are treated at their physical infinite cut-off limit $R_c = \infty$. Finally, we note that the positions and relative intensity of the $I(q)$ maxima differ dramatically from those found experimentally (figure 2(a) of [8]); an adjustment of τ does not solve the discrepancy. This may indicate that a quantitative description of the low-temperature spin–spin correlations in GGG requires inclusion of exchange interactions beyond nearest neighbours [10, 11] and/or single ion anisotropy.

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