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Ferrimagnetism in the rare-earth iron garnets: a Monte Carlo study

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Abstract
We investigate classical vector spin models of the rare-earth iron garnet ferrimagnets yttrium iron garnet (YIG) and gadolinium iron garnet (GdIG) using Monte Carlo simulations. Critical temperatures agree well with experiment. A compensation point is observed in GdIG, again in good agreement with experiment.

1. Introduction

Ferrimagnets are materials in which different sublattices have opposing magnetic moments of unequal magnitude. Thus, unlike antiferromagnets, they have a nett moment at low temperatures, which vanishes at a critical temperature T_c. Furthermore, since the sublattice magnetizations will not, in general, have the same temperature dependence, there is the possibility of their cancellation at some lower temperature T_{comp}, known as a compensation point.

Ferrimagnetic materials have been known and studied for a long time [1, 2]. The 'classic' materials are the spinels and garnets [2, 3]. In the present work we focus on the rare-earth iron garnets, yttrium iron garnet (YIG) and gadolinium iron garnet (GdIG). Both have critical temperatures near 550 K (560 K and 565 K, respectively) and GdIG has a compensation point at 290 K. There has been a great deal of work, both experimental and theoretical, on both of these materials over the past 50 years, and one can claim that they are well understood. Theoretical treatments are largely based on a Heisenberg model of localized spins at the magnetic sites, interacting via short range exchange interactions. However, the model has only been analysed within the molecular-field approximation (MFA) [4] or, at low temperatures via spin wave (SW) theory [5]. It is well known that MFA is quantitatively unreliable. In particular it overestimates the critical temperature, in this case by some 20%, and therefore leads to a corresponding underestimate of exchange parameters, obtained by fitting the MFA results to experiment [5]. The motivation for the present work has been to go beyond MFA, by using classical Monte Carlo simulations, which allows a proper treatment of thermal fluctuations. Our only approximation is to represent the quantum spins by classical vectors, which can point in any direction in space. For large quantum number S (here $S = 5/2, 7/2$) this should be a good approximation, except at very low temperatures.

2. Models and methods

The structure of the rare-earth iron garnets, with chemical formula written usually as $R_3Fe_5O_{12}$, is complex. Each cubic unit cell contains 8 formula units, i.e. 160 ions. The Fe^{3+} cations lie on two inequivalent sublattices (conventionally labelled 'a' and 'd'), and the rare-earth ions lie on a third sublattice (labelled 'c'). Each unit cell has, respectively, 16, 24, 24 a, d, and c sites. Figure 1 shows the positions of the cations in one unit cell.

Each a site has 8, 6, 6 nearest neighbour a, d, c sites respectively, while the corresponding numbers for d and c sites are 4, 4, 2 and 4, 2, 4.

The spin Hamiltonian is written in the form

$$H = 2\sum_{\langle i,j\rangle} J_{ij}\mathbf{S_i}\cdot\mathbf{S_j} \qquad (1)$$

with interactions restricted to nearest neighbours between each species. The magnetic ions Fe^{3+}, Gd^{3+} are S-state ions ($L = 0$) and therefore crystal-field effects are expected to be unimportant. In principle, one should also include dipolar interactions in the Hamiltonian. However, these are believed to be small in these materials [5], and are usually neglected.

In principle the values of the exchange parameters could be estimated by *a priori* electronic structure calculations but, to our knowledge, this has not been attempted. In practice they have been estimated by fitting to experimental results. The more reliable approach has been to fit measured

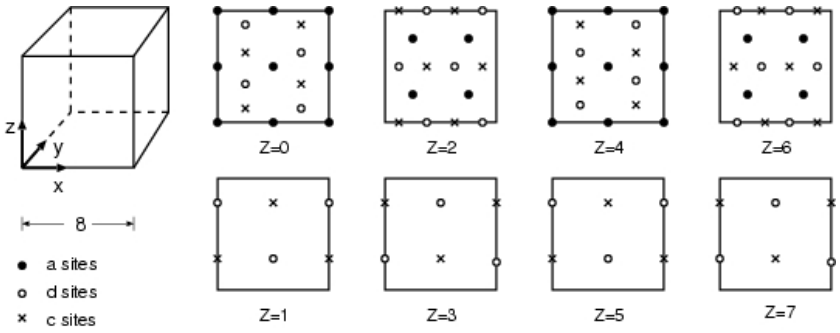

Figure 1. Positions of a, d, c sites in a cubic unit cell of the garnet structure. The panels show slices parallel to the x–y plane, for $z = 0$–7.

Table 1. Exchange parameters for YIG and GdIG (values in K).

	Reference	Year	J_{ad}	J_{aa}	J_{dd}	J_{ac}	J_{dc}	
YIG	Harris	1963	45.7	9.1	9.1			SW magnetization
YIG	Plant	1977	39.8	8.0	8.0			Magnon spectra
YIG	Srivastava	1982	30.04	6.45	12.05			MFA
YIG	Cherepanov	1993	39.8	3.8	13.4			Magnon spectra
GdIG	Harris	1963	45.7	9.1	9.1	2.52	10.1	SW
GdIG	Dionne	1970	36.4	−12.2	−17.1	−1.29	6.76	MFA
GdIG	Srivastava	1982	30.04	6.45	12.05	0.60	1.80	MFA

magnon spectra obtained by inelastic neutron scattering to theoretical spin wave results [6, 7], or to fit low temperature magnetization data to SW results [5]. Other workers have fitted thermodynamic data over an extended temperature range to the MFA results [8, 9], but, as already remarked above, this leads to a significant underestimate of the parameters. Table 1 lists some of the parameter sets that have been used by previous workers.

In YIG the only magnetic ions are the Fe^{3+}, with $S = 5/2$. The dominant exchange interaction is J_{ad} between nearest neighbour sites. This is antiferromagnetic and results in antiparallel alignment of the a and d moments. Ferrimagnetism arises simply from the unequal numbers of a and d sites. Nearest neighbour interactions J_{aa} and J_{dd} within each sublattice are also important. There is some apparent disagreement in the literature about the sign of these. Dionne [8] gives these as ferromagnetic, (actually for GdIG, but by implication also for YIG), but the most recent and most comprehensive study [7] clearly states that all interactions are antiferromagnetic. Thus there is a degree of frustration in each sublattice, overcome by the dominant coupling J_{ad}. In our Monte Carlo work we have used the most recent set [7] $J_{ad} = 39.8$ K, $J_{dd} = 13.4$ K, $J_{aa} = 3.8$ K.

In GdIG the c sites are occupied by Gd^{3+} ions, with $S = 7/2$. The most important additional exchange couplings are believed to be J_{ac} and J_{dc}. The values of the exchange parameters are less well known for GdIG. We have used the parameter set of Harris [5] (table 1).

We use a standard Monte Carlo procedure with single spin updates [10], with classical vector spins of length $\sqrt{(}S(S+1))$. As the Hamiltonian is rotationally invariant in spin space, the total moment can rotate freely without energy cost. To avoid difficulties associated with this we include a small uniaxial magnetic anisotropy in the Hamiltonian, to yield a preferred ordering axis. Our results are not affected significantly by this. A number of runs are taken at each temperature, to ensure reasonable statistics. We have used systems of L^3 unit cells, with $L = 4, 5, 6$; i.e. 4096, 8000, 138 24 sites. These are found to be large enough to avoid significant finite-size effects. Various quantities are 'measured', as averages over the corresponding time series. In our plots we show the specific heat and sublattice magnetizations as functions of temperature. The specific heat peak is used to give the critical temperature, and the sublattice magnetizations yield the total moment.

3. Results

3.1. YIG

Figure 2 shows some of our results for YIG, in the form of specific heat and (normalized) sublattice magnetizations versus temperature. The joined point are for $L = 6$ (138 24 sites). We also show, as unconnected points, specific heat results for $L = 4$ (4096 sites). There is no systematic difference between the two cases, confirming that our system is large enough. The specific heat peak, at 520 K, gives an estimate of the critical temperature. This should be compared with the experimental value of 560 K. The magnetization data can be used to compute the total moment—there is no compensation point, in agreement with experiment. Of course, since we use a classical model, our results will not yield the correct low temperature behaviour. In particular, the specific heat will not fall to zero, as it should.

3.2. GdIG

Similar results for GdIG are shown in figure 3. The specific heat peak occurs at 550 K. This is to be compared to the

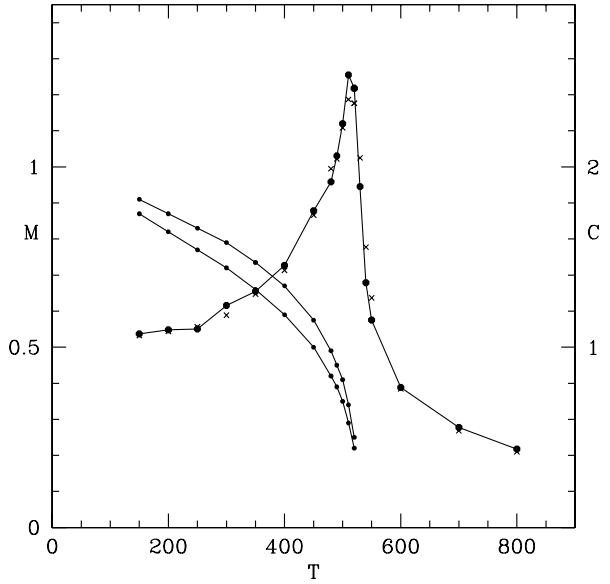

Figure 2. Monte Carlo results for YIG. The filled circles are MC results for a system of 138 24 sites. The crosses are specific heat results for a smaller system of 4096 sites. For convenience both magnetizations (upper and lower curves are, respectively, for a and d sublattices) are shown as positive. The lines are simply guides for the eye.

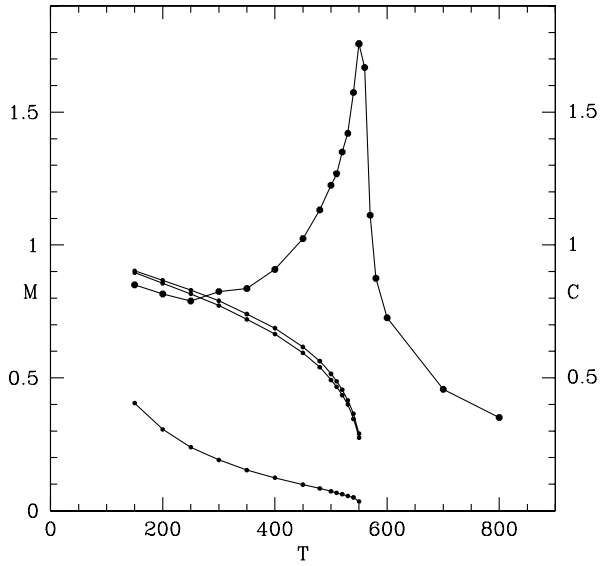

Figure 3. Monte Carlo results for GdIG for a system of 13824 sites. The specific heat and the three sublattice magnetizations (a, d, c from top to bottom) are shown versus temperature, for the parameter set of Harris [5].

experimental value of 564 K. The sublattice magnetizations for the a and d sublattices (Fe^{3+} spins) vary with temperature in a way very like that in YIG. However the sublattice c magnetization (Gd^{3+} spins) shows a rapid decrease at lower temperatures. This reflects the relatively weak coupling between the Gd and Fe spins. This rapid change, in the region 100–200 K, should be reflected in the specific heat, and indeed there appears to be a broad peak at 150 K in our results. We have not explored this in detail.

The total magnetization of GdIG, to within a normalization factor, is

$$M_{\text{ferri}} = -15\sigma_{\text{d}} + 10\sigma_{\text{a}} + 21\sigma_{\text{c}}. \tag{2}$$

This is shown, versus temperature, in figure 4. We note the compensation point at 330 K, where M_{ferri} changes sign. This is in reasonable agreement with the experimental value of 290 K.

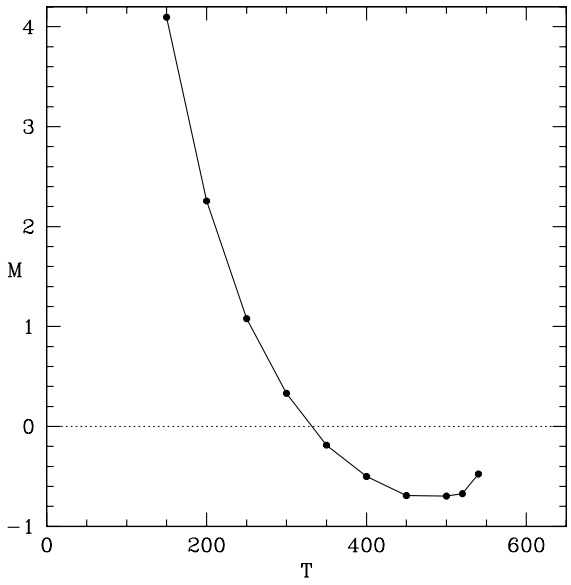

Figure 4. Total moment for GdIG, showing the compensation point.

4. Summary and conclusions

We have used classical Monte Carlo simulations to compute the thermodynamic properties of a vector spin model for the rare-earth iron garnet ferrimagnets YIG and GdIG. Thus we are able to go beyond the usual molecular-field approach (MFA). Using exchange parameters, obtained, by previous workers, by fitting magnon spectra from inelastic neutron scattering measurements, we find critical temperatures in good agreement with the experimental values. For GdIG we find a compensation point at 330 K, in good agreement with experiment. Thus we have demonstrated that the same set of exchange parameters suffices, as it should, to describe both the low temperature properties, in the spin wave regime, and the thermodynamic properties at higher temperatures. More generally, we have demonstrated that a classical Monte Carlo approach is useful to study complex magnetic materials. It is possible therefore to study other ferrimagnets, such as other rare-earth garnets and spinels. It seems surprising that this approach has not been commonly used before. The only related work, of which we are aware, is a study of gadolinium gallium garnet [11], which is an interesting frustrated antiferromagnet. Stanica *et al* [12] have reported results of a Monte Carlo study of the rare-earth iron garnets, but based on an Ising description, which neglects the transverse fluctuations of the spins. A preliminary report of this work has been given previously [13].

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