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Field-induced transitions in the highly frustrated magnet gadolinium gallium garnet – long- or short-range order?

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Abstract. In gadolinium gallium garnet (GGG) the antiferromagnetically interacting Gd ions are located on two corner-sharing triangular sublattices. The magnetic system is therefore highly frustrated and does not order in zero field. Antiferromagnetic (AF) order can only be induced by the application of an external magnetic field. We have measured the magnetic diffraction pattern of a single crystal of GGG at temperatures between 50 mK and 0.9 K in fields of up to 4 T. These experiments reveal that the H–T phase diagram has a much more complicated nature than previously assumed. Three groups of magnetic Bragg peaks were found: strong ferromagnetic peaks, saturating in fields above 2.5 T, strong AF peaks with a maximum intensity at the lower-field phase boundary around 0.6 T and weak AF peaks which have a maximum intensity at the upper phase boundary around 1.5 T. The widths of the strong AF peaks are limited by the instrument resolution only in the vicinity of the lower phase boundary.

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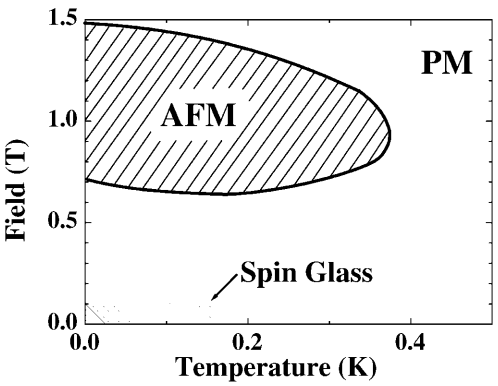

Fig. 1. Low-temperature magnetic phase diagram of GGG, Ref. [1, 2] showing the paramagnetic (PM), antiferromagnetically ordered (AFM) and spin glass phases

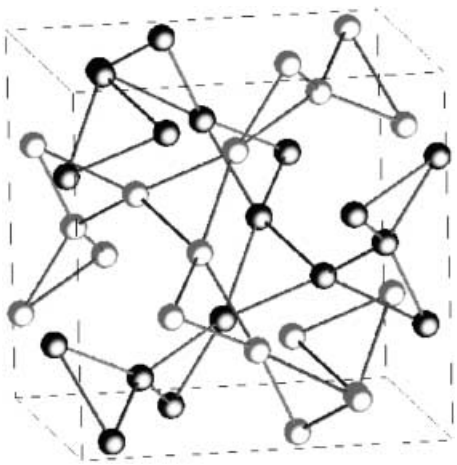

Fig. 2. Positions of the magnetic Gd ions in a garnet structure. There are 24 magnetic ions per unit cell, they are divided into two interpenetrating sublattices

It has been known for more than twenty years that Gadolinium Gallium Garnet, $Gd_3Ga_5O_{12}$, has an extraordinary low-temperature phase diagram [1, 2], as shown on Fig. 1. While having a relatively high Curie-Weiss temperature ($\Theta_{CW} \sim -2$ K) GGG possesses no long-range magnetic order down to 25 mK. In the currently measured temperature range ordering can be induced only by an applied magnetic field of around 1 T. Such a peculiar behaviour has been attributed to the combination of the two factors. Firstly, in GGG, the magnetic Gd ions are located on two interpenetrating corner-sharing triangular sublattices, as shown on Fig. 2, and there is strong geometrical frustration of the magnetic interactions. Secondly, GGG is magnetically very isotropic – in sharp contrast with the rest of rare-earth garnets single-ion anisotropy in GGG is almost zero and, apart from dipole-dipole anisotropy, it is a perfect Heisenberg antiferromagnet. Despite all experimental efforts, the situation regarding the origin of field induced order in GGG remains far from clear. The H–T

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phase diagram of GGG has been thoroughly studied through susceptibility, heat capacity, thermal conductivity, and magnetocaloric measurements [2, 3]. Peculiarly, the lower field boundary of the AF phase is shown to have distinct minimum at $T \sim 0.2$ K (see Fig. 1), in analogy with the minimum in the melting curve of ^{4}He. Our earlier powder neutron diffraction studies showed clearly the development of short-range magnetic order without an applied field [4] and the appearance of long range order in an applied magnetic field [5]. The existence of the ordered phase was also confirmed by Monte Carlo simulations [2, 6]. Muon spin relaxation studies [7], however, have found that the spin correlations in GGG appear to remain essentially dynamic at low temperatures even in applied magnetic fields of up to 1.8 T. One possible scenario of magnetic ordering in GGG has been suggested recently by Zhitomirsky et al. [8]. Namely, for a classical Heisenberg antiferromagnet on a garnet lattice, a stabilisation of a collinear state by thermal fluctuations (through an order by disorder effect) has been found at one-third of the saturation field. For such a model a field induced collinear state has a spin gap and produces a magnetisation plateau.

In this paper we report the results of neutron diffraction measurements on a single crystal of GGG. The experimental results give general support to the proposed ordering mechanism, although further investigations may be required to draw a firm conclusion.

1 Experimental details

Polycrystalline GGG was synthesised starting from stoichiometric quantities of Gd_2O_3 and Ga_2O_3 by a solid diffusion reaction at $T = 1400\,^\circ$C for 12 hours. In order to minimise neutron absorption we have prepared a sample with 99.98% of the non-absorbing isotope, ^{160}Gd. This isotope was supplied by Oak Ridge National Laboratory. The single crystal sample of GGG was then grown by the floating zone method using a two mirror infra-red image furnace.

The neutron diffraction measurements were carried out using the E1 instrument at Berlin Neutron Scattering Center, Hahn-Meitner-Institut. The E1 triple axis spectrometer was used in a double-axis mode, with a pyrolytic graphite (002) monochromator providing a neutron wavelength of 2.41 Å. A pyrolytic graphite filter was installed in the incident beam to remove higher order contamination. The horizontal collimation was $40' - 80' - 40'$. Typical intensities of the main nuclear Bragg peaks were several hundred counts per second.

The measurements were performed in the temperature range between 50 mK and 900 mK using an Oxford Instruments dilution refrigerator insert. The horizontal scattering plane contained the (hk0) reflections so that an external magnetic field provided by a vertical split-pair cryomagnet was applied along the [001] direction.

2 Results and discussion

Before applying a magnetic field, we have attempted to find the magnetic Bragg peaks associated with the "spin freezing" in zero field. These peaks have been found in a powder diffraction pattern at temperatures below 140 mK [4] and incommensurate positions, such as $|Q_1| = 0.64$ Å^{-1} and $|Q_2| = 0.85$ Å^{-1} and several others. Our attempts to find single crystal peaks of the (hk0) type with a scattering vector length equal to Q_1 or Q_2 were unsuccessful.

On application of an external field two sets of magnetic Bragg peaks appeared, ferromagnetic and antiferromagnetic. The former have intensity increasing with the field and saturate at higher fields, the latter have intensity growing in low fields, reaching a maximum, and then decreasing and almost disappearing at a field above 2.5 T. Figure 3 shows the evolution of the integrated intensity of the three different magnetic peaks at $T = 60$ mK with applied magnetic field. For the purposes of clarity, the intensity of the ferromagnetic peak, (220), has been divided by 10, while the intensity of relatively weak antiferromagnetic peak, (330), has been multiplied by the same amount.

The stronger AF peaks, such as (200), have maximum intensity in a field of about $H_{\rm low} \approx 0.6$ T, which corresponds to the lower-field phase boundary on the $H - T$ phase diagram. The same field is marked by the start of intensity rising for the weaker AF peaks, such as (330), and for all the FM peaks. The weaker AF peaks have maximum intensity in a field of about $H_{\rm high} \approx 1.5$ T, which corresponds to the saturation field – an upper-field phase boundary. Note the nonzero intensity of the AF peaks in fields well above $H_{\rm high}$. The FM peaks demonstrate a significant variation of intensity only in the region between $H_{\rm low}$ and $H_{\rm high}$.

Interestingly, the width of the (200) peak is limited by the instrumental resolution only in a small region of fields around $H_{\rm low}$. The width of the weaker AF peaks always seem to remain above the resolution limit. This observation suggests that the true long range magnetic order in GGG is induced only in the vicinity of the lower-field phase boundary in agreement with theoretical predictions [8], while the region of field between $H_{\rm low}$ and $H_{\rm high}$ corresponds to an enhancement of the short range AF order. There are, however, at least two possible objections to this statement.

The first objection comes from the fact that there is no requirement that the AF peaks corresponding to the appearance of the long-range order in GGG should necessarily lie in the [hk0] scattering plane when the field is applied along the [001] direction. It is possible that besides (200) and (330)

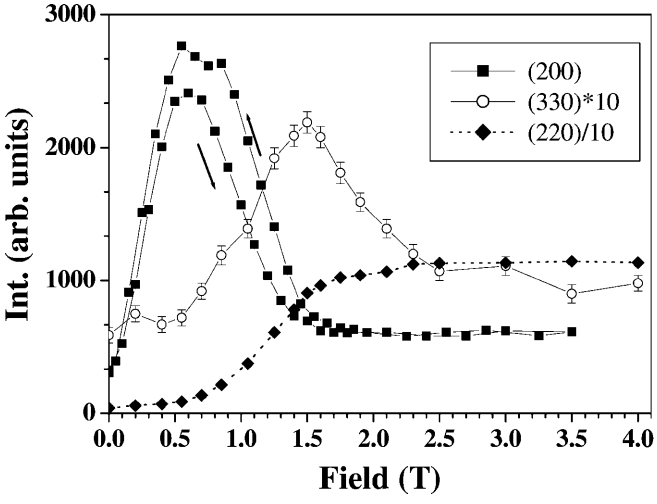

Fig. 3. Field dependence of the integrated intensity of the three magnetic peaks in GGG measured at $T = 60$ mK, $H \parallel [001]$

types AF peaks found in this study there are some other out of plane Bragg peaks associated with the ordering. In neutron diffraction data on a powder sample [5] in addition to the (200) peak, the (210) peak as well as an incommensurate magnetic peak located between them have been found. These extra peaks are clearly seen in a powder diffraction pattern for $0.4 < H < 1.6$ T, but they have not been observed in the present single crystal experiment.

The second objection is suggested by the unusual temperature dependence of the AF Bragg peaks. Figure 4 shows the integrated intensity of the (200) peak measured at $H = 0.6$ T. The intensity remains nearly constant in the temperature range $50 < T < 500$ mK and decreases significantly only when the temperature is increased above 0.7 K. Experimental restrictions have not allowed us to warm the sample above 0.84 K, however, a rough estimate of $T \sim 0.9$ K can be given for the transition temperature to a disordered phase. This result is in sharp contrast with the previous measurements of the specific heat and magnetic susceptibility of GGG [2], which have indicated the existence of the ordered phase in the temperature range up to 0.4 K, as shown on Fig. 1.

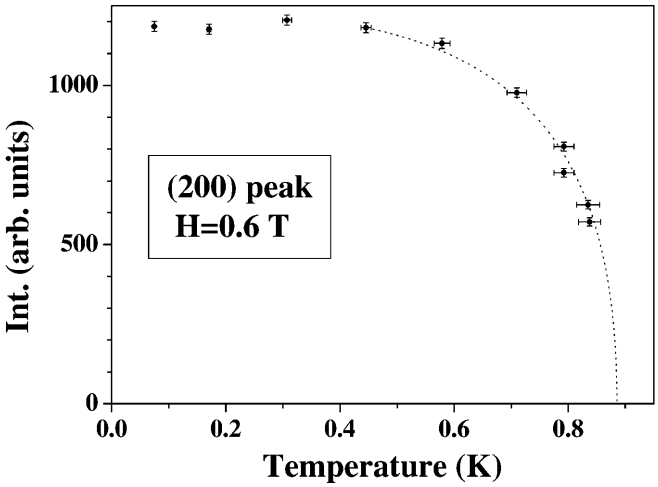

Fig. 4. Temperature dependence of the integrated intensity of the (200) peak measured at $H = 0.6$ T, $H \parallel [001]$. *Dotted line* is the guide for the eye

To summarise, GGG is an intriguing magnetically frustrated system where it is difficult to project out a single ground state in an applied magnetic field. The current state of the experimental studies clearly demonstrates the interest but also suggests that considerably more effort is required to extract the underlying features and to identify the dominant interactions.

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