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2007 J. Phys.: Condens. Matter 19 145271

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Single-ion anisotropy and transverse magnetization in the frustrated gadolinium pyrochlores

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Received 8 August 2006

Published 23 March 2007

Online at stacks.iop.org/JPhysCM/19/145271

Abstract

A single-ion anisotropy of the planar type is found in the pyrochlore antiferromagnet oxides $\text{Gd}_2\text{M}_2\text{O}_7$ ($M = \text{Ti}, \text{Sn}$); its strength is comparable with the strength of the exchange coupling. Models considering the effects of the planar anisotropy predict the appearance of a transverse magnetization in an applied magnetic field. A detailed experimental study of $\text{Gd}_2\text{Ti}_2\text{O}_7$ single crystals reveals that a transverse magnetization is really present at low temperatures. The magnetic phase diagram of $\text{Gd}_2\text{Ti}_2\text{O}_7$ is refined.

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

Pyrochlore antiferromagnets, where magnetic ions form a lattice of corner-sharing tetrahedra, demonstrate a high degree of geometric frustration. A nearest-neighbour exchange interaction alone cannot select a unique ground state, thus a classical Heisenberg pyrochlore should remain in the macroscopically degenerated disordered spin-liquid state down to $T = 0$ [1]. Rare-earth oxides $\text{Gd}_2\text{M}_2\text{O}_7$ ($M = \text{Ti}, \text{Sn}$) are usually considered as examples of Heisenberg pyrochlores, since the spin-orbit coupling should be small for the S-state Gd^{3+} ion [2]. However, both titanate and stannate order at temperatures near 1 K. Their ordered states are quite different: while $\text{Gd}_2\text{Sn}_2\text{O}_7$ orders via a first-order phase transition into a $\mathbf{k} = 0$ state [3], $\text{Gd}_2\text{Ti}_2\text{O}_7$ undergoes a two-step phase transition ($T_{\text{N1}} = 1.05$ K and $T_{\text{N2}} = 0.75$ K) into a complicated partially disordered $\mathbf{k} \neq 0$ structure [4]. The H - T phase diagrams of $\text{Gd}_2\text{Ti}_2\text{O}_7$ [5] evidence two phase transitions induced by an external field at $\mu_0 H_{c1} \sim 3$ T and $\mu_0 H_{c2} \sim 6$ T. While the selection of the $\mathbf{k} \neq 0$ state in $\text{Gd}_2\text{Ti}_2\text{O}_7$ could be explained by the interplay of further neighbour exchange interactions in the model of dipolar pyrochlore [3, 6], the nature of the phase transitions in the gadolinium titanate still remains an enigma.

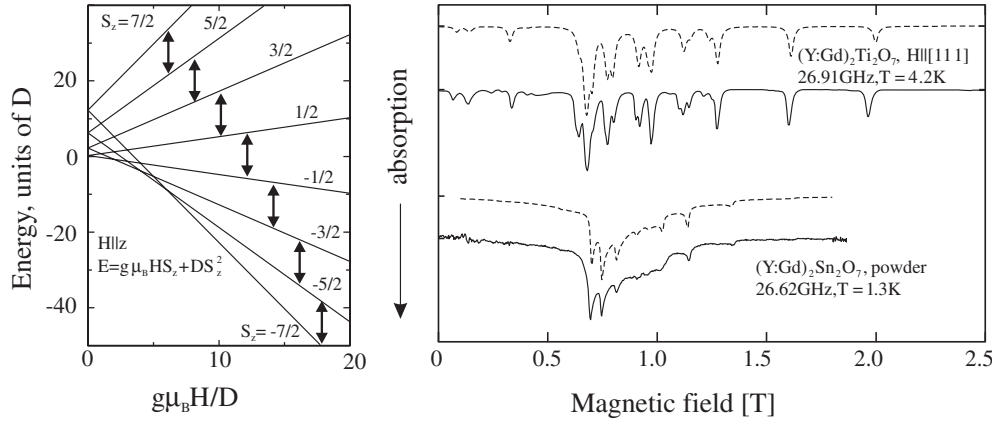


Figure 1. Left panel: scheme of the energy levels of the crystal field split $S = 7/2$ multiplet. Vertical arrows mark positions of the dipolar transitions ($\Delta S_z = \pm 1$) with the same energy. Right panel: examples of the observed ESR absorption spectra in $(Y_{0.995}Gd_{0.005})_2Ti_2O_7$ and $(Y_{0.995}Gd_{0.005})_2Sn_2O_7$ (solid) and modelled ESR absorption lines (dashed).

The contribution of the single-ion anisotropy has so far been neglected for the gadolinium compounds, since Gd^{3+} is an S-state ion. However, a small single-ion anisotropy is usually present on the Gd^{3+} ions [7]. It is due to the strong spin-orbit coupling of the 4f-electrons, which mixes $^8S_{7/2}$ and $^6P_{7/2}$ states. Thus, determination of the single-ion anisotropy parameters in the gadolinium pyrochlores and analysis of their effects on the magnetic properties of these compounds are important issues.

Study of the single-ion effects in the concentrated magnet $Gd_2M_2O_7$ is hampered by the presence of the inter-spin couplings. To avoid spin-spin interactions we have grown samples of the isostructural non-magnetic compounds: $Y_2Ti_2O_7$ single crystals and $Y_2Sn_2O_7$ polycrystals, with a small fraction of the Y ions (nominally 0.5%) substituted by the Gd ions [8, 9]. The crystal lattice parameters of the corresponding yttrium and gadolinium compounds are very close, thus we expect that the crystal field on the rare-earth site is the same.

Electron spin resonance (ESR) is a very sensitive method to look for small crystal field splittings. ESR absorption is due to the dipolar transitions with $\Delta S_z = \pm 1$. Should the ground-state multiplet be split, the resonance conditions $\Delta E = \hbar\omega$ for the different dipolar transitions would be fulfilled at the different fields (see scheme in figure 1). This would result in the multi-component ESR absorption spectrum. The ESR study was performed at frequencies of 18–100 GHz using a set of home-made transmission-type ESR spectrometers equipped with an He-cooled cryomagnet. The ESR absorption spectra were recorded as the field dependences of the transmitted microwave signal. Observation of several absorption components on the measured ESR absorption spectra (figure 1) indicates the presence of a single-ion anisotropy on the Gd ions.

Note that the analysis of the observed spectra is complicated by the presence of the four inequivalent positions with respect to the magnetic field (since the main anisotropy axis for the Gd ion is aligned along the corresponding local trigonal axis) and, additionally, by the averaging over powder particle orientations in the case of $(Y_{0.995}Gd_{0.005})_2Sn_2O_7$. Absorption components corresponding to certain transitions can be identified; their frequency-field dependences are characteristic for the case of a crystal-field split $S = 7/2$ multiplet [8, 9]. Additionally, we have performed a numerical modelling of the ESR absorption spectra (see dashed lines in figure 1). This modelling automatically accounts for the above-mentioned complications. The calculated

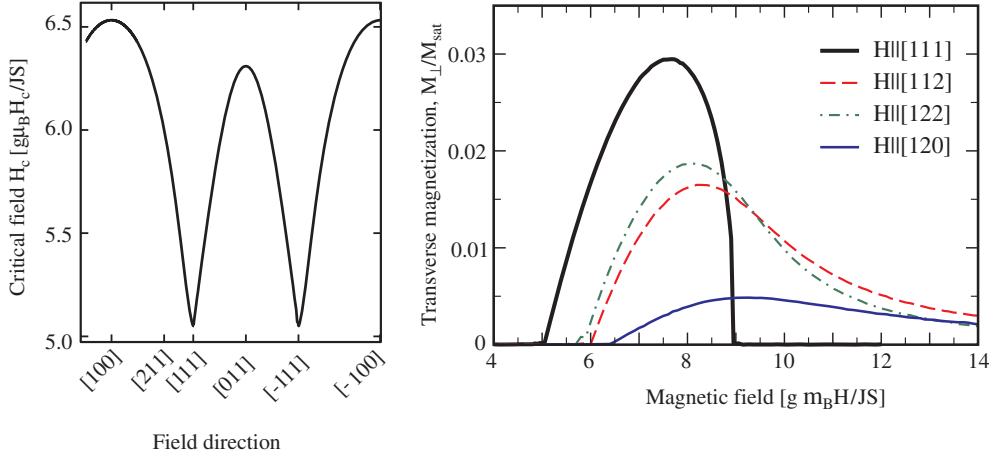


Figure 2. Left panel: orientation dependence of the critical field for the model (1). Right panel: modelled field dependences of the transverse magnetization at several orientations for $D = 0.75J$.

curves reproduce the experimental curves well. The fitting parameters in our model are the parameters of the single-ion anisotropy. We found that, both for the titanate and the stannate, the second-order axial term $D\hat{S}_z^2$ dominates; the higher-order terms provide only minor corrections to the splitting of the energy levels [8, 9]. The single-ion anisotropy is of the easy plane type ($D > 0$) for both compounds. The main anisotropy constant D is equal to 0.223 K for the $\text{Gd}_2\text{Ti}_2\text{O}_7$ and 0.140 K for the $\text{Gd}_2\text{Sn}_2\text{O}_7$. This equals $0.75J$ and $0.5J$ for the titanate and the stannate, respectively (here J is the exchange integral value for the corresponding compound).

Effects of the staggered planar anisotropy on the ordering of a pyrochlore antiferromagnet were considered by other authors [3, 11]. Here we focus on his effects on the magnetization process. We consider a simplified model of the classical pyrochlore which takes into account a nearest-neighbour exchange and a staggered planar anisotropy [8]:

$$\hat{\mathcal{H}} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + D \sum_i (\mathbf{n}_i \cdot \mathbf{S}_i)^2 - \mathbf{H} \cdot \sum_i \mathbf{S}_i. \quad (1)$$

Here \mathbf{n}_i is the unit vector in the direction of the local trigonal axis, $D > 0$. Note that this model is only qualitative in application to the *real* gadolinium pyrochlores.

The model (1) could be reduced to the consideration of a single tetrahedron. At zero field, the constraint on the total spin per tetrahedron $\mathbf{S}_{\text{tetr}} = 0$ could be fulfilled by keeping all spins in their respective easy planes. Moreover, this classical ground state remains infinitely degenerate; this degeneracy can be parametrized by a single continuous parameter per tetrahedron. This degeneracy remains if a small magnetic field is applied. As the magnetic field increases, the parametrization parameter space shrinks, but up to a certain critical field both exchange and anisotropy energies could be minimized simultaneously. Below the critical field, the magnetization process of the anisotropic pyrochlore (1) is exactly the same as that of the classical Heisenberg pyrochlore with the susceptibility $\chi = 1/(2J)$. The critical field value H_c does not depend on the value of the anisotropy constant. It is determined by the orientation only (see figure 2).

Above H_c the magnetization process of the anisotropic pyrochlore deviates from the pure Heisenberg case. Exchange and anisotropy energies cannot be minimized simultaneously any more and the spins begin to deviate from their easy planes. Since the anisotropy axes directions

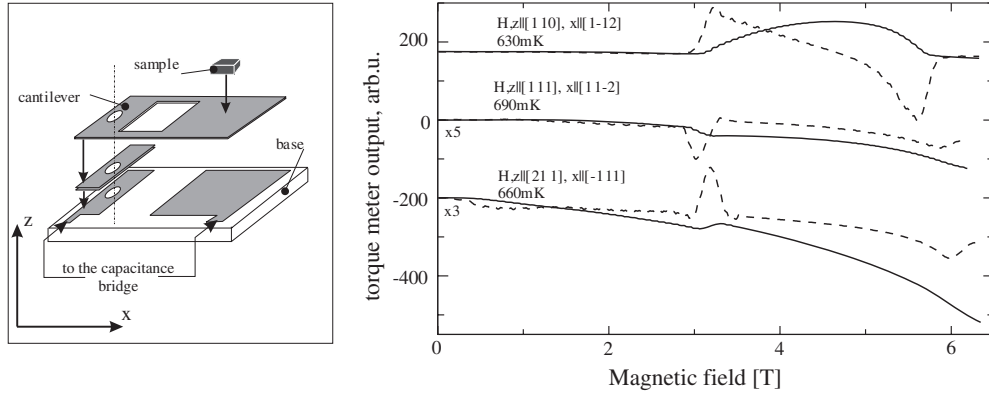


Figure 3. Left panel: scheme of the experimental cell for the magnetic torque measurements. Right panel: solid curves—torquemeter output field dependences at different orientations. Dashed curves—field derivatives of the corresponding torquemeter output curves.

are staggered, these deviations are different for the different vertices of the tetrahedron. As a result, a non-compensated transverse magnetization appears at $H > H_c$. The numerically calculated field dependence of the transverse magnetization is shown in figure 2. The magnitude of the transverse magnetization, calculated for $D = 0.75J$ (the value determined for $\text{Gd}_2\text{Ti}_2\text{O}_7$) is several per cent of the saturation magnetization.

To check the prediction of the possible appearance of a transverse magnetization in the pyrochlore antiferromagnet submitted to a magnetic field, we have performed a detailed transverse magnetization study of $\text{Gd}_2\text{Ti}_2\text{O}_7$ single crystals [10]. The measurements were made using a home-made capacitance torquemeter (figure 3). The sample is mounted on a flexible cantilever. A torque $\mathbf{M} \times \mathbf{H}$ acting on the sample in the magnetic field is compensated by the elastic bending of the cantilever. The latter is detected as a change of the cell capacitance. The torquemeter response is $\Delta C \propto M_x H_z + \alpha M_z H_z$; here α should be zero for the perfect experimental conditions. The parasitic contribution $M_z H_z$ is due to the magnetic-balance effect in the presence of a field gradient in addition to the demagnetization-induced torque.

The parasitic effects could be estimated in the paramagnetic phase, since its cubic symmetry forbids transverse magnetization. In the ordered state we observe a change in the torquemeter response field dependence, with two anomalies located at low temperatures near 3 and 6 T (figure 3). Subtracting the parasitic contribution determined in the paramagnetic phase from the low-temperature curves, we recover the field dependences of the transverse magnetization (figure 4). We have found that the transverse magnetization is present in the low-temperature ordered phases ($T < T_{N2}$) both in low and in high fields. No definite conclusions could be drawn from the present data about the high-temperature ordered phase ($T_{N1} > T > T_{N2}$), which has a cubic symmetry according to the neutron scattering experiments [4]. The maximal amplitude of the observed transverse magnetization estimated from the cantilever elastic constant corresponds to 10^{-3} of the saturation magnetization. By plotting the observed anomalies of the torquemeter output on the $(H-T)$ plane, we obtain the magnetic phase diagrams of $\text{Gd}_2\text{Ti}_2\text{O}_7$ (figure 5). Our results correspond well to the published diagrams [5], except for the $\mathbf{H} \parallel [211]$ orientation. Here we observe a phase transition line which was not reported before.

Observation of the transverse magnetization in the low-temperature ordered phases of $\text{Gd}_2\text{Ti}_2\text{O}_7$ indicates that the cubic symmetry is lost there. This finding could be important

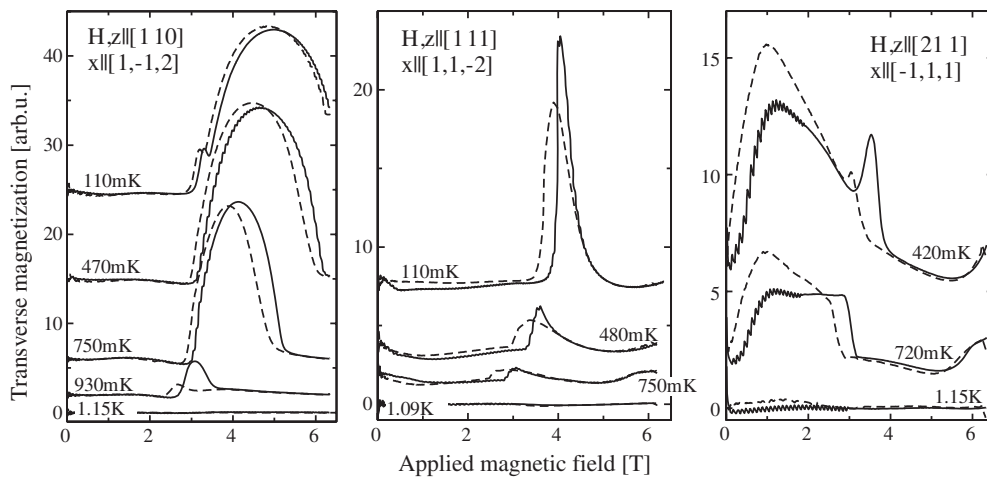


Figure 4. Field dependences of the transverse magnetization in $Gd_2Ti_2O_7$ at different orientations. Solid curves—data measured on increasing magnetic field; dashed curves—on decreasing field.

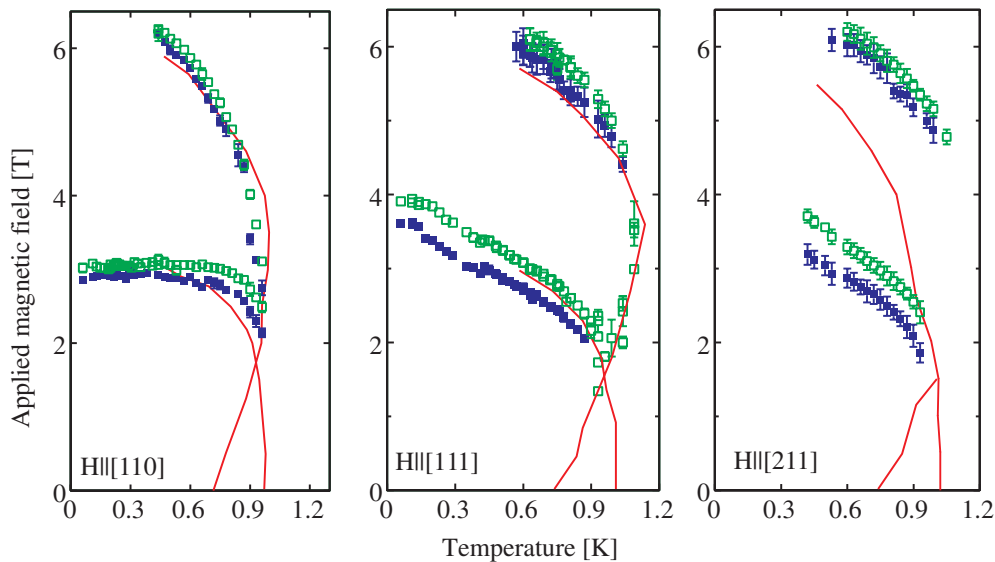


Figure 5. $(H-T)$ phase diagrams of $Gd_2Ti_2O_7$. Open symbols (green)—data taken on increasing field; closed symbols (blue)—data taken on decreasing field; solid lines (red)—data from [5].

for the deciphering of the low-temperature ordered structures of gadolinium titanate and for a better understanding of its phase diagrams.

Acknowledgments

The authors thank S Sosin (Kapitza Institute) and I Sheikin (CNRS-Grenoble/HMFL) for discussions concerning the results and experimental techniques. The Kapitza Institute team (VG and AS) thanks the Russian Foundation for Basic Research for support of their work

(grant no. 04-02-17294-a). One of the authors (VG) thanks the National Science Support Foundation for support of his studies.

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