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LETTER TO THE EDITOR

Observation of a transverse magnetization in the ordered phases of the pyrochlore magnet $Gd_2Ti_2O_7$

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Abstract

We have performed a detailed transverse magnetization study of the pyrochlore antiferromagnet $Gd_2Ti_2O_7$. A transverse magnetization of about $10^{-3}M_{sat}$ is observed in the low-temperature ordered phases. These measurements result in the refinement of the $Gd_2Ti_2O_7$ phase diagrams. Observation of a transverse magnetization indicates loss of the cubic symmetry in some of the magnetic phases and provides new information for a better understanding of the complicated magnetic ordering of $Gd_2Ti_2O_7$.

Gadolinium compounds $Gd_2Ti_2O_7$ and $Gd_2Sn_2O_7$ provide good examples of the Heisenberg antiferromagnets on the pyrochlore lattice. The magnetic Gd^{3+} ions are located at the vertices of the network of corner sharing tetrahedra. The nearest-neighbour Heisenberg exchange interaction is strongly frustrated in this geometry—nearest-neighbour exchange coupling alone is not sufficient to select a unique ground state of the pyrochlore antiferromagnet. However, both of these compounds orders near 1 K [1]. This ordering is caused by the subtle interplay of weaker interactions (e.g. dipolar coupling, further-neighbour exchange couplings and single-ion anisotropy).

The magnetic phase diagrams of $Gd_2Ti_2O_7$ [2], as determined from the specific heat measurements, reveal a variety of ordered magnetic phases. Two phase transitions are observed in zero external field at $T_{N1} = 1.05$ K and $T_{N2} = 0.75$ K. At low temperatures a transition to the saturated phase is observed at a field near 6 T, while an additional phase transition is found near $\mu_0 H \sim 3$ T for the $\mathbf{H} \parallel [110], [111]$. The latter phase transition is reported to be absent for the $\mathbf{H} \parallel [211]$. The two-step phase transition in zero field and the field-induced transitions in this compound are not understood yet. Moreover, the experimentally determined phase diagram for the $\mathbf{H} \parallel [211]$ [2] is not consistent with the general thermodynamic restrictions [3] forbidding existence of a point where three second-order transition lines meet. Identification of the zero-field magnetic structure by the means of neutron scattering [4] yields a $4\mathbf{k}$-structure with $\mathbf{k} = (1/2, 1/2, 1/2)$ where 1/4 of the Gd ions remains disordered at $T_{N2} < T < T_{N1}$.

These ions order at the second phase transition at T_{N2}. The high-temperature ordered phase ($T_{N2} < T < T_{N1}$) preserves cubic symmetry[3], while the symmetry of the low-temperature phase is not clearly understood, being hidden in the complications of the 4**k**-structure.

The present work was stimulated by the prediction of a possible appearance of a transverse magnetization in the classical Heisenberg pyrochlores above some critical fields [5]. A transverse magnetization change is a sensitive indicator for the magnetic phase transition [6]. The transverse magnetization could appear in the crystal if the magnetic field direction deviates from the main axes of the susceptibility tensor. In the case of the cubic symmetry the susceptibility tensor is isotropic. Thus, no transverse magnetization could be observed in the magnetic phases with cubic symmetry.

To measure the transverse magnetization we have used a capacitance torquemeter. The sensitive element of the torquemeter was a plane capacitor with the parallel plates formed by the rigid base and the flexible bronze cantilever. The sample was glued to the flexible cantilever and the experimental cell was mounted on the dilution refrigerator equipped with a 6 T cryomagnet. The cantilever was thermalized with the mixing chamber. The magnetic field was applied perpendicular to the capacitor plates. The experimental cell was connected to the General Radio capacitance bridge, which was balanced before the measurements. The imbalance of the bridge was measured as a function of the applied magnetic field. The maximal capacitance change during the measurements was about 20%. The capacitance of the experimental cell at $H = 0$ was about 1.4 pF. Cantilever deformations were found to be always reversible. A $Gd_2Ti_2O_7$ single crystal was prepared following the same technique as described in [7]. Samples were oriented and cut into rectangular shape with the dimensions of about $0.6 \times 0.9 \times 1.3$ mm^3.

The ideal torquemeter response is only due to the transverse magnetization component $\mathbf{M}_\perp$ aligned along the flexible cantilever. As the magnetic field **H** is applied perpendicular to the cantilever plate, the torque $\mathbf{M}_\perp \times \mathbf{H}$ acts on the sample. This torque is compensated by the bending of the cantilever resulting in the change of the experimental cell capacitance ΔC detected as the bridge imbalance. Thus, the torquemeter output could be expressed as $U(T, H) \propto \Delta C \propto M_\perp H$.

A real torquemeter is also slightly sensitive to the longitudinal magnetization component $\mathbf{M}_\parallel$ via the magnetic-balance response due to the small uncontrolled field gradient at the sample position. Another uncontrolled effect is a torque due to the demagnetization tensor anisotropy for the non-spherical samples—an elongated sample in a magnetic field tends to rotate its longest axis parallel to the field direction. These two parasitic effects should be present even for the cubic crystals. They are proportional to H^2 at low fields and are expected to change their field dependence at the saturation field. They are expected to be almost temperature independent since the magnetic susceptibility of $Gd_2Ti_2O_7$ does not change strongly below 1 K [1].

Field dependences of the torquemeter response taken at different temperatures are shown in figure 1. Above 1 K the occurrence of a transverse magnetization is forbidden by the cubic symmetry of the paramagnetic phase. The observed response is due to the parasitic effects of longitudinal magnetization and demagnetization-induced torque. The maximal amplitude of the response due to these parasitic effects can be estimated from the amplitude of the torquemeter response at 6 T in the paramagnetic phase. The shape of the torquemeter response changes as the temperature decreases below 1 K, i.e. when the sample enters the ordered state. For $\mathbf{H} \parallel [110], [111]$ a strong hump-shaped deviation of the torquemeter output from the high-temperature behaviour appears near 3 T. The amplitude of this hump at low temperatures

[3] As one can see from reference [4], at $T_{N1} > T > T_{N2}$ all $\langle 111 \rangle$ directions are the third-order symmetry axes. Thus, the symmetry of this phase is at least tetrahedral and belongs to the cubic symmetry class.

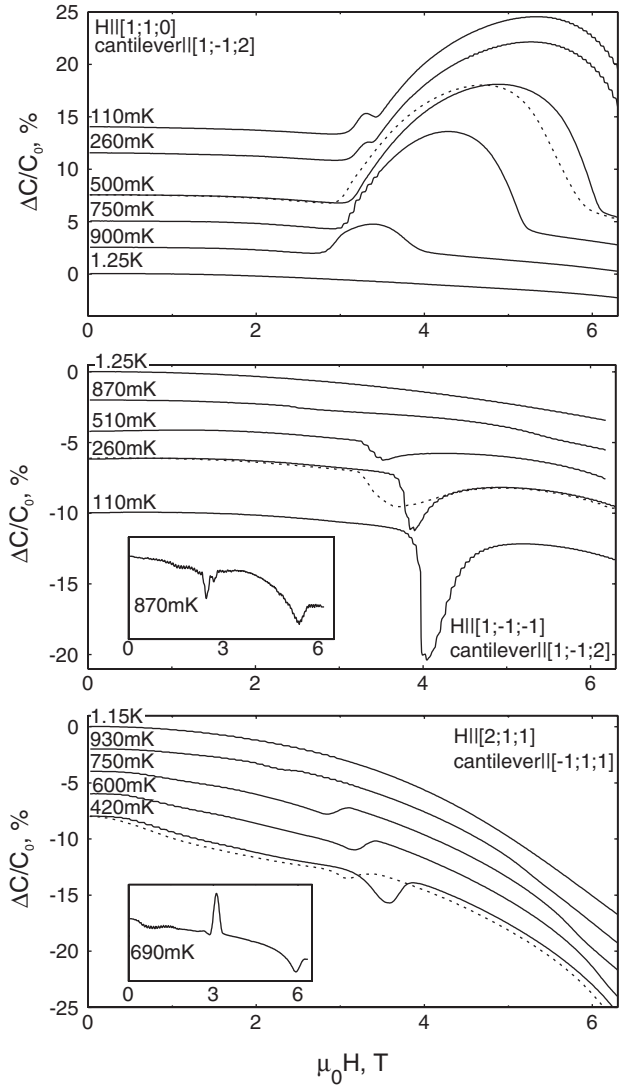

Figure 1. Field dependences of the torquemeter response for different field orientations. Insets: field dependences of the field derivative of the torquemeter response for the corresponding orientations. Solid curves (main panels and insets): data measured on increasing magnetic field. Dashed curves: examples of the data measured on decreasing field.

exceeds significantly the response that could be ascribed to the parasitic effects; i.e., observed hump-shaped signals are due to the appearance of the transverse magnetization. At high fields the hump disappears and the torquemeter response is close to that observed in the paramagnetic state. For the **H** ∥ [110] this hump has well defined low-field and high-field edges, which allow us to estimate the critical fields of appearance and disappearance of the transverse magnetization. For the **H** ∥ [111] the transverse magnetization signal has a sharp low-field edge only.

The evolution of the torquemeter response for the **H** ∥ [211] geometry is different. As the sample is cooled down below 1 K the *low-field* part of the torquemeter response deviates from

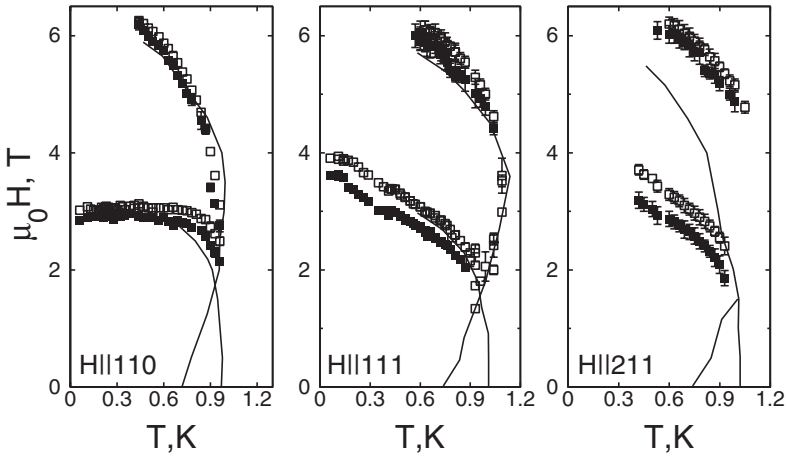

Figure 2. H–T phase diagrams. Open symbols: data taken on increasing field. Closed symbols: data taken on decreasing field. Solid curves: data from [2].

the paramagnetic H^2 behaviour. Developing hump demonstrates a sharp high-field edge. For this orientation we were unable to perform measurements below 400 mK for technical reasons. Amplitudes of the torquemeter response deviations from the high-temperature behaviour is not high enough to ascribe unambiguously these changes to the transverse magnetization effects. However, to attribute this signal to the parasitic effects, one has to assume that the sample magnetization at low fields (below 3 T) almost reaches the saturation and then decreases. Neglecting this unlikely scenario, we conclude that this response is also connected with the appearance of the transverse magnetization.

A transition to the saturated phase can be found for the $\mathbf{H} \parallel [111], [211]$ orientations by taking the field derivatives of the torquemeter output (see insets in figure 1). They change sharply near 6 T from the field-dependent behaviour to a constant owing to the change of the field dependences of the parasitic effects at the saturation field.

A hysteresis both in the position and in the form of the transverse magnetization related hump is observed for all orientations (see figure 1). At increasing fields appearance and disappearance of the transverse magnetization are always observed at higher field values than for decreasing field. The hump is also more pronounced on the experimental curves taken on increasing field. This hysteresis does not disappear as the magnetic field sweep rate is tenfold decreased.

Identified features of the torquemeter response (i.e. fields of appearance and disappearance of the transverse magnetization and fields of the transition to the saturated phase) were used to draw the (H–T) diagrams shown in figure 2. These features correlate very well with the known phase diagrams [2] for the $\mathbf{H} \parallel [110], [111]$ orientations of the applied magnetic field. However, our results for the $\mathbf{H} \parallel [211]$ suggest that there is a phase transition line near $\mu_0 H = 3$ T that was not detected in the known specific heat measurements [2]. This suggested phase transition line is in agreement with general thermodynamic restrictions [3]. We have measured the field dependence of the specific heat in the $\mathbf{H} \parallel [211]$ geometry at $T = 620$ mK using a Quantum Design PPMS calorimeter (figure 3). Our specific heat data demonstrate clear transitions near 3.1 and 6.1 T in perfect agreement with the transverse magnetization measurements.

Finally, we can recover the field dependences of the transverse magnetization. To account for the parasitic effect contribution, we subtracted the torquemeter response measured in the

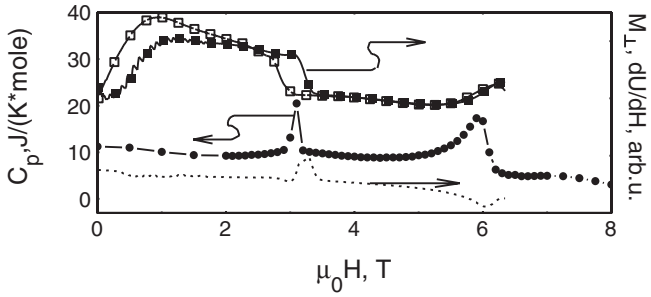

Figure 3. Field dependence of the specific heat (circles) and its correspondence to the results of transverse magnetization measurements. Dashed line—field derivative of the torquemeter output; squares—determined field dependences of the transverse magnetization measured at increasing (closed) and decreasing (open) field. For all curves $T = 620$ mK, $\mathbf{H} \parallel [211]$.

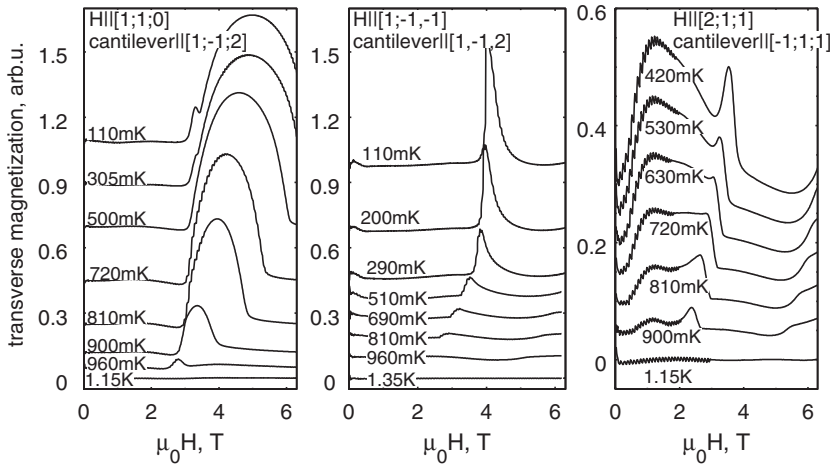

Figure 4. Field dependences of the normalized transverse magnetization (see text). All curves correspond to the measurements made on increasing magnetic field.

paramagnetic phase (at $T_0 = 1.15 \ldots 1.35$ K) from the low temperature response. This approach yields the following expression for the transverse magnetization per unit mass:

$$|M_\perp(T, H)| \propto \frac{|U(T, H) - U(T_0, H)|}{Hm} \tag{1}$$

where m is the sample mass. The field dependences of the so-extracted transverse magnetization are presented in figure 4. Note that for the $\mathbf{H} \parallel [211]$ orientation at $T_{N1} > T > T_{N2}$ (810 and 900 mK curves) the transverse magnetization appears only above a given magnetic field. This observation is in agreement with the high symmetry of the high-temperature ordered phase, which forbids the transverse magnetization. The critical field for the appearance of the transverse magnetization corresponds well to the known phase boundary between ordered phases.

The magnitude of the transverse magnetization can be roughly estimated from the elastic constant of the cantilever. This yields a value of $3 \times 10^{-3} M_{sat}$ for the maximal amplitude of the observed transverse magnetization.

Summarizing our results, we have observed a transverse magnetization in the low-temperature magnetically ordered phases of the pyrochlore antiferromagnet $Gd_2Ti_2O_7$. This means unambiguously that the cubic symmetry is lost in the low-temperature ordered phases of $Gd_2Ti_2O_7$. We have also refined the magnetic phase diagrams and have detected a new phase transition line for $\mathbf{H} \parallel [211]$.

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