Journal of Physics: Conference Series **150** (2009) 042188 doi:10.1088/1742-6596/150/4/042188

Low-frequency spin dynamics of the frustrated pyrochlore magnet $Gd_2Ti_2O_7$

A I Smirnov[1], S S Sosin[1], V N Glazkov[1], O A Petrenko[2], G Balakrishnan[2] and M E Zhitomirsky[3]

[1] P. L. Kapitza Institute for Physical Problems RAS, 119334 Moscow, Russia
[2] Department of Physics, University of Warwick, Coventry CV4 7 AL, UK
[3] Commissariat á l'Energie Atomique, DSM/DRFMC/SPSMS, 38054 Grenoble, France

E-mail: smirnov@kapitza.ras.ru

Abstract. The adiabatic $T(H)$ curves measured in a Heisenberg pyrochlore $Gd_2Ti_2O_7$ prove the existence of a macroscopic number of local soft modes in this compound. A spin gap of 25 GHz, developing in $Gd_2Ti_2O_7$ on cooling down to 1.3 K (in the collective paramagnetic phase), was observed by ESR spectroscopy. ESR of diamagnetically diluted crystals revealed the single-ion anisotropy energy to be about a quarter of Curie-Weiss temperature. This might be responsible for the observed energy gap. Below 1 K, in the magnetically ordered phase, the spin excitations have a three-branch spectrum with two energy gaps.

Pyrochlore antiferromagnets have attracted much attention as exchange systems on a crystal lattice with a macroscopically degenerate ground state. This degeneracy arises due to the frustration of exchange bonds and prevents conventional antiferromagnetic ordering down to very low temperatures (see, e.g. Ref. [1]). $Gd_2Ti_2O_7$ with Gd^{3+} ($S = 7/2$, $L = 0$) ions was considered as a Heisenberg pyrochlore system. It remains paramagnetic when cooling through the Curie-Weiss temperature $\theta = 10$ K, and demonstrates a two-step magnetic ordering at low temperatures with $T_{N1} = 1.0$ K and $T_{N2} = 0.7$ K [2]. The classical spins on the pyrochlore lattice have specific excitations, which may be ascribed to rotations of antiferromagnetically correlated spins on hexagon loops. These are local soft modes, which are naturally combined with the macroscopic degeneracy of the classical ground state. These modes were detected in neutron scattering experiments on another related compound, the cubic spinel $ZnCr_2O_4$ [3], by measurements of a structure factor, corresponding to spin hexagons. The aim of the present paper is to collect and review our recent experimental results in order to establish a full description of the low-energy spin dynamics of $Gd_2Ti_2O_7$.

The macroscopic number of degenerate soft modes is predicted to give rise to a residual entropy, which may be affected by an external magnetic field. This entropy should result in an enhanced cooling of the spin system during the adiabatic demagnetization. In contrast to conventional paramagnets, where the cooling is limited to temperatures of about θ, this cooling would be effective even in the temperature range $T \ll \theta$. We performed an adiabatic demagnetization experiment (see Ref. [4] for details). The observed adiabatic $T(H)$ curves are shown in Fig. 1 by the symbols, while the solid lines present the results of classical Monte Carlo simulations of the process. The only parameter used for these calculations is the exchange integral $J = 0.3$ K, taken from the value of θ. The dotted lines represent the experimental

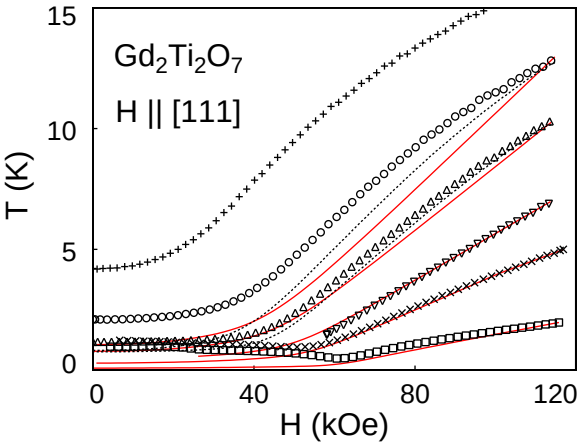

Figure 1. Adiabatic $T(H)$ curves[4]. *Reprinted figure with permission from S. S. Sosin, L. A. Prozorova, A. I. Smirnov, A. I.Golov, I. B. Berkutov, O. A. Petrenko, G. Balakrishnan, and M. E. Zhitomirsky, Phys. Rev. B* ***71****, 094413 (2005).* ©*2005 by the American Physical Society.*

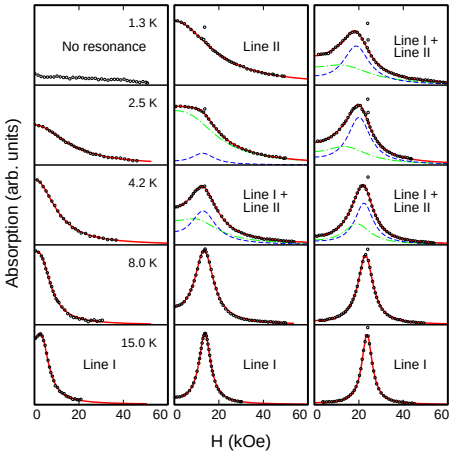

Figure 2. ESR absorption at 9 GHz (left), 36 GHz (middle), 70 GHZ (right).

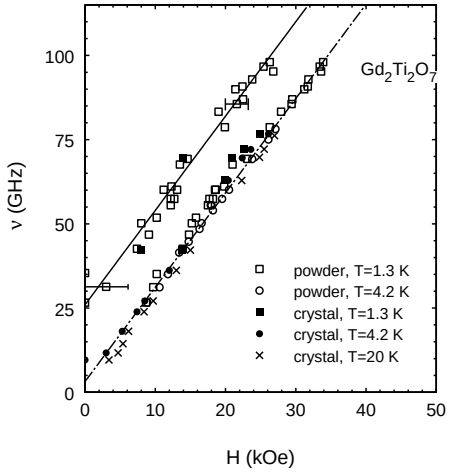

Figure 3. ESR frequency-field dependences at T=1.3 K and T >4 K, **H** ∥ [111]. Lines are linear fits [5].

Fig.2 is reprinted with permission from [5] S. S. Sosin, L. A. Prozorova, A. I. Smirnov, P. Bonville, G. Jasmin - Le Bras, O. A. Petrenko, Phys. Rev. B ***77****, 104424 (2008).* ©*2008 by the American Physical Society.*

temperature corrected for the influence of the lattice, i.e., the extrapolation to a purely spin system. A very good correspondence between the observed or corrected $T(H)$ curves and the calculations confirms the concept of degenerate soft modes.

As a further step, we looked for spin excitations by means of electron spin resonance (ESR) spectroscopy in the frequency range 9 to 100 GHz. At $T \gg \theta$ there is a single ESR line with g-factor of 2.0. The resonances become wider on cooling below θ and at $T = 1.3$ K their behaviour is frequency-dependent: 9 GHz ESR disappears below 2 K, the 36 GHz resonance transforms into a wide band with the absorption maximum at zero field and the 70 GHz absorption maximum is shifted from the high-temperature position towards zero field. The recorded absorption lines were fitted by two Lorentzians, as presented in Fig. 2. Frequency-field dependences of spectral components, determined by fitting are given in Fig. 3. The upper branch has an energy gap of 25 GHz and a linear frequency-field dependence. This gap is of an unknown nature and is unexpected in the exchange approximation. The single ion magnetic anisotropy was suggested to be the origin of the gap.

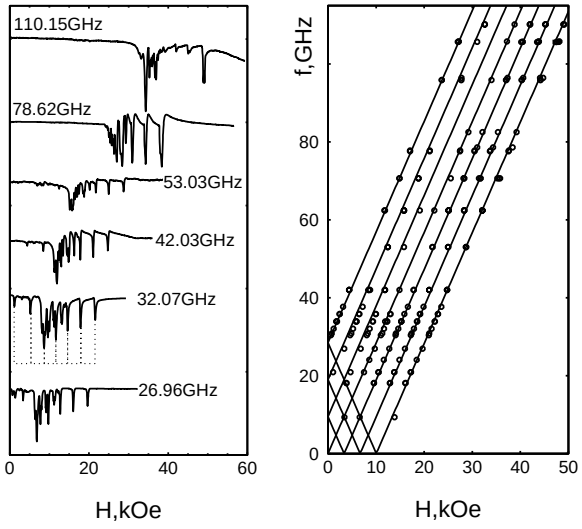

Figure 4. ESR lines and spectrum for magnetically diluted samples at $T = 4.2$ K [6]. Lines: theory. *Reprinted figure with permission from V. N. Glazkov, M. E. Zhitomirsky, A. I. Smirnov, H.-A. Krug von Nidda, A. Loidl, C. Marin and J.-P. Sanchez, Phys. Rev. B* ***72****, 020409(R) (2005). ©2005 by the American Physical Society.*

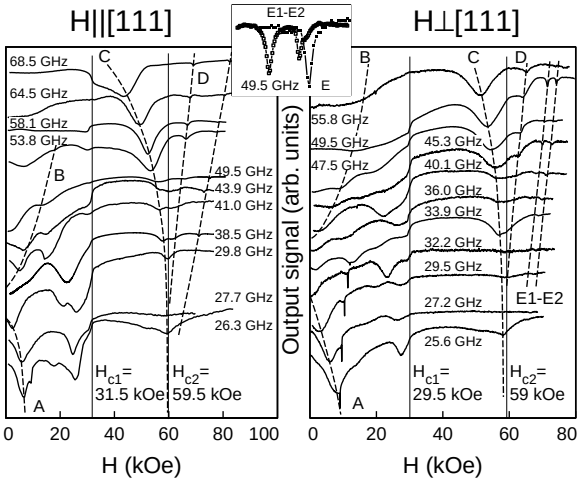

Figure 5. ESR lines in the ordered phases of $Gd_2Ti_2O_7$ at $T = 0.42$ K. [7].

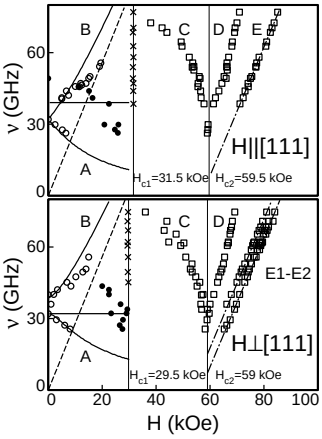

Figure 6. Spectrum of ordered phases [7]. Solid lines: theory, dash-dotted lines: linear fits.

Figures 5, 6 are reprinted with permission from S. S. Sosin, A. I. Smirnov, L. A. Prozorova, G. Balakrishnan, and M. E. Zhitomirsky Phys. Rev. B ***73****, 212402 (2006). ©2006 by the American Physical Society.*

A measurement of the anisotropy constant was performed using diamagnetically diluted crystals $(Y_{0.995}Gd_{0.005})_2Ti_2O_7$ [6], where the Gd^{3+}-ions are in the same crystal field as in $Gd_2Ti_2O_7$, but do not interact because of a nonmagnetic dilution by yttrium. The ESR spectra in Fig. 4 demonstrate a strong effect of the single ion anisotropy, which splits the ESR line into a multicomponent spectrum. For $\mathbf{H} \parallel [111]$ one can choose seven equidistant resonances, one of which has the largest field. A standard analysis [8] of these lines gives the main term in the single-ion easy-plane anisotropy $D \approx 0.25$ K. Other lines were attributed to Gd-ions, with the local crystal field axes aligned along the other {111} directions. Details of the analysis are presented in Ref. [6]. The single-ion anisotropy energy $DS_z^2 = 2.73$ K is not negligible compared to the exchange energy presented by the value of $\theta = 10$ K. Note that the observed energy gap of $Gd_2Ti_2O_7$ exactly corresponds to the transition $|5/2\rangle \rightarrow |7/2\rangle$ of an isolated Gd^{3+} ion at the field parallel to the local anisotropy axis. Other transitions have smaller frequencies. However,

at $T \ll \theta$, the spectrum of $Gd_2Ti_2O_7$ should be related to the excitations above the collective ground state, which should be triplets. The influence of short-range correlations on excitations in quantum-disordered gapless spin systems was first reported for 1D $S = 5/2$ spin chains [9]. The observed ESR shift was attributed to the combined effect of the dipolar interaction and a single-ion anisotropy. A similar effect in pyrochlores requires theoretical studies. The observed temperature and the field evolution of ESR signals resembles the case of triplet excitations split by crystal and magnetic fields. Once the nature of the collective excitations is known precisely, a detailed analysis of this problem can be performed.

Next we describe the magnetic resonance in the ordered phase [7]. The observed ESR lines and spectra at $T = 0.42$ K are presented in Figs. 5, 6. On cooling the sample through T_{N1} and T_{N2} the widened ESR line transforms into a complex multicomponent spectrum. As shown in Fig. 6, there are two energy gaps and three branches in the low-field range. The changes of the spectrum at the magnetic field H_{c1} and H_{c2} correspond to a phase transition detected earlier [2] and to the saturation field, correspondingly. The analysis [7] based on the macroscopic theory results in the conclusion that the two-gap and three branch spectrum corresponds to a noncollinear nonplanar spin structure. At $H > H_{c2}$ the ground state corresponds to a ferromagnetic alignment of spins. Here the four spin-wave modes (according to a number of ions in a unit cell) are given as [10]

$$\nu_{1,2} = \tilde{\gamma}H - 8JS \ , \ \nu_{3,4} = \tilde{\gamma}H - 2JS(2 \mp \sqrt{1 + 3\eta_{\mathbf{k}}}) \qquad (1)$$

where $\eta_{\mathbf{k}}$ is a certain combination of lattice harmonics ($\eta_{\mathbf{k}=0} = 1$). $\nu_{1,2}$ are soft modes with no dispersion acquiring a finite gap above the saturation field $\tilde{\gamma}H_{c2} = 8JS$. Additional interactions should produce the weak dispersion resulting in finite gaps at $\mathbf{k} = 0$. We suggest that the lines E,E1,E2 of the spectrum demonstrating exact linear field dependencies with small gaps correspond to this type of mode. The other two modes are dispersive branches with gaps $\nu_{3,\mathbf{k}=0} = 0$ and $\nu_{4,\mathbf{k}=0} = \tilde{\gamma}H_{c2}$. The line D should be the mode ν_3 while the mode ν_4 lies above the spectrometer range.

In summary, we have tested the low-frequency spin dynamics of the pyrochlore antiferromagnet $Gd_2Ti_2O_7$ by means of thermodynamic and electron spin resonance methods. Low-temperature cooling via adiabatic demagnetization confirms the presence of local quasi-soft modes. Microwave spectroscopy indicates an energy gap in the correlated low temperature state at temperatures above the magnetic ordering. The spectrum of the antiferromagnetic resonances of the low temperature ordered phase consists of three branches with two energy gaps and indicates a complicated nonplanar spin ordering.

Acknowledgments

The work is supported by the Russian Foundation for Basic Research.

References

[1] Canals B and Lacroix C 2000 *Phys. Rev. B* **61** 1149
[2] Petrenko O A, Lees M R, Balakrishnan G and, Paul D McK 2004 *Phys. Rev. B* **70** 012402
[3] Lee S-H, Broholm C, Ratcliff W, Gasparovic G, Huang Q, Kim T H and Cheong S-W 2002 *Nature* **418** 856
[4] Sosin S S, Prozorova L A, Smirnov A I, Golov A I, Berkutov I B, Petrenko O A, Balakrishnan G and Zhitomirsky M E 2005 *Phys. Rev. B* **71** 094413
[5] Sosin S S, Prozorova L A, Smirnov A I, Bonville P, Jasmin-Le Bras G and Petrenko O A 2008 *Phys. Rev. B* **77** 104424
[6] Glazkov V N, Zhitomirsky M E, Smirnov A I, Krug von Nidda H-A, Loidl A, Marin C and Sanchez J-P 2005 *Phys. Rev. B* **72** 020409(R)
[7] Sosin S S, Smirnov A I, Prozorova L A, Balakrishnan G and Zhitomirsky M E 2006 *Phys. Rev. B* **73** 212402
[8] Abragam A and Bleaney B 1970 *Electron Paramagnetic Resonance of Transition Ions* (Oxford: Clarendon Press)
[9] Nagata K and Tazuke Y 1972 *J. Phys. Soc. Japan* **32** 337
[10] Zhitomirsky M E 2003 *Phys. Rev. B* **67** 104421