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Low-energy spin dynamics of Heisenberg pyrochlore magnets

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Abstract

The low-temperature magnetic resonance in two related Heisenberg pyrochlores $Gd_2Ti_2O_7$ and $Gd_2Sn_2O_7$ is studied. The gapped acoustic branches are found in both compounds. Weakly dispersive soft modes are observed in the saturated phase of $Gd_2Ti_2O_7$, in agreement with the predictions of the spin-wave theory.

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1. Introduction

Magnetic frustration arises in the case that all paired interactions in the system cannot be simultaneously minimized. The distinct feature of strongly frustrated magnets (e.g. on kagome, garnet or pyrochlore lattices) is the macroscopic degeneracy of their magnetic ground states giving rise to delay in a long-range ordering down to temperatures much lower than the characteristic exchange energy. This degeneracy is lifted due to weaker forces, with the resulting ordering capable of being different even for systems with similar crystal structures. Two Heisenberg pyrochlore magnets $Gd_2Ti_2O_7$ and $Gd_2Sn_2O_7$ is a nice illustration of the above principles. Both compounds have the Curie–Weiss constant of about 10 K while their ordering appears at temperatures below 1 K. The phase diagrams and the properties of ordered phases are quite different: two Néel temperatures at $T_{N1,2} \simeq 1.0$ and 0.75 K and two field transitions at $H_{c1,2} \simeq 30$ and 60 kOe (H_{c2} is the saturation field) are observed in $Gd_2Ti_2O_7$ [1], while the only ordered phase is found in $Gd_2Sn_2O_7$ [2]. According to neutron scattering results [3], the magnetic structure of $Gd_2Ti_2O_7$ is described by four wave vectors of $\mathbf{k} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ type with partial disorder of one fourth of magnetic ions. Recent study of $Gd_2Sn_2O_7$ [4] suggests its magnetic ions to be fully ordered in the plane "cross-type" structure with $\mathbf{k} = 0$.

Another important consequence of a macroscopic degeneracy is that the excitation spectrum in this case contains soft, almost dispersionless modes in the whole Brillouin zone which were indirectly shown to exist by enhanced magnetocaloric effect in $Gd_2Ti_2O_7$ [5]. The purpose of the present work is the ESR study of exotic ordering in both compounds as well as an attempt to detect directly the soft modes in the low-energy part of their resonance spectra.

2. Experiment and discussion

The magnetic resonance of a single crystal $Gd_2Ti_2O_7$ and a powder sample of $Gd_2Sn_2O_7$ were studied at temperatures between 0.4 and 30 K over a wide frequency and field range. In the high temperature range $T \gg T_N$ both systems demonstrate a paramagnetic resonance with an isotropic g-factor of about 2.0 gradually converting into a wide nonresonant band of absorption in the vicinity of 1 K (much stronger in amplitude for $Gd_2Ti_2O_7$). At $T = 0.4$ K

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several lines of an antiferromagnetic resonance were observed in the absorption spectrum of $Gd_2Ti_2O_7$ at magnetic fields applied parallel and perpendicular to the [1 1 1]-axis of the sample. Two of them have gaps $\Delta_{1,2}(H = 0) = 32$ and 39 GHz (A and B on Fig. 1, respectively), the first one decreasing and the second increasing in field slightly dependent on its orientation. At fields above H_{c1} one more resonance appeared at both field directions (line C), monotonously falling in the intermediate field range $H_{c1} < H < H_{c2}$ and softening at $H = H_{c2}$. The saturated phase is characterized by additional resonance modes: lines D and E for $H \| [1\,1\,1]$ and lines D and E1–E2 for $H \perp [1\,1\,1]$. The modes E and E1–E2 were found to have exact linear frequency-field dependence with the slope corresponding to g-factor 1.95. The resonance spectrum of $Gd_2Sn_2O_7$ sample consists of only two branches with equal gap values of 33.5 GHz and a field evolution quite similar to those observed for lines A and B in the spectrum of $Gd_2Ti_2O_7$. No other absorption was detected at the noise level of about 0.5% of the output signal.

The detailed interpretation of the data on $Gd_2Ti_2O_7$ is labored by the absence of the microscopic model which would initially give the correct ground state of the system. In the low field range one can apply the "spin-hydrodynamic" approach for the magnetic structure with cubic or tetrahedral symmetry and isotropic susceptibility, the properties being consistent with the 4k-structure proposed in Ref. [3]. It yields the following cubic equation for eigen frequencies of three acoustic modes of the spectrum (see Ref. [6] for details):

$$(\nu^2 - \Delta_1^2)(\nu^2 - \Delta_2^2)^2 + \gamma^2\nu^2(\Delta_1^2 H_\parallel^2 + \Delta_2^2 H_\perp^2 - \nu^2 H^2) = 0,$$

where $\Delta_{1,2}$ are the gap values, $H_\parallel$ and $H_\perp$ are field components with respect to [1 1 1] axis, $\gamma = g\mu_B/h$ is a gyromagnetic ratio. These formulae (shown by solid lines on Fig. 1) reflect the qualitative features of lines A and B in both principal orientations of the magnetic field.

For the saturated phase $H > H_{c2}$, the ground state in the nearest exchange approximation simply corresponds to ferromagnetic alignment of all spins, with four spin–wave branches existing in the excitation spectrum. Two of them are former dispersionless soft modes acquiring the linear in field gap $\nu_{1,2} = \gamma(H - H_{c2})$. In the reality, they should be splitted by weak interactions resulting in finite frequencies also at $H = H_{c2}$. The lines E and E1–E2 of the spectrum are suggested to correspond to this type of excitations. Other two modes are dispersive magnon branches. One of them can be related to the line D while the last one has a gap of about $\gamma H_{c2} \sim 200$ GHz out of our experimental conditions.

Theoretical analysis of the low field resonance spectrum in $Gd_2Sn_2O_7$ on the assumption of the plane magnetic structure proposed in Ref. [4] is forthcoming. The microscopic model explaining the absence of other absorption lines in its resonance spectrum should be also constructed (Fig. 2).

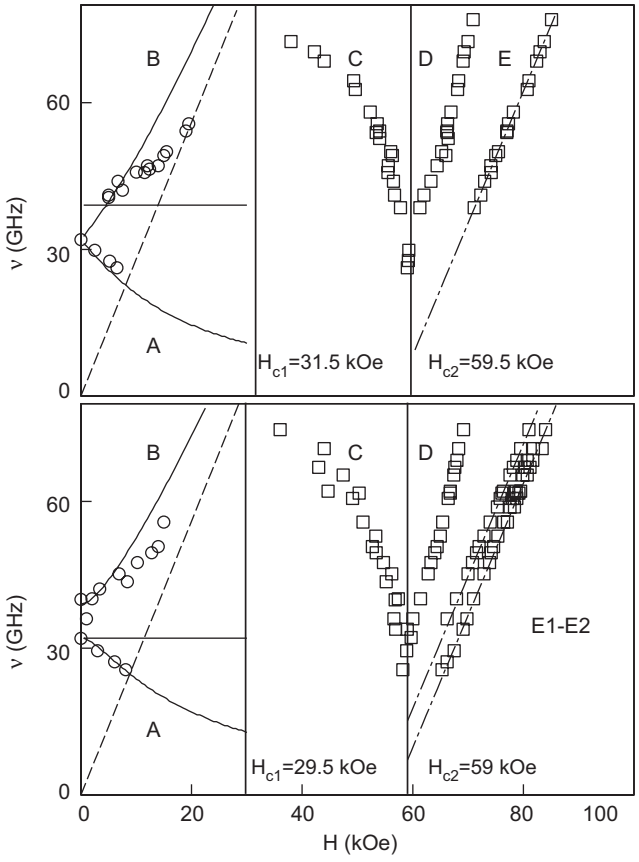

Fig. 1. Frequency field diagram of the resonance spectrum in $Gd_2Ti_2O_7$ at $T = 0.42$ K for $H \| [1\,1\,1]$ (upper panel) and $H \perp [1\,1\,1]$ (lower panel); solid lines are low field theoretical calculations, dashed-dotted lines are linear fits as described in the text, dashed line is a paramagnet with $g = 2.0$.

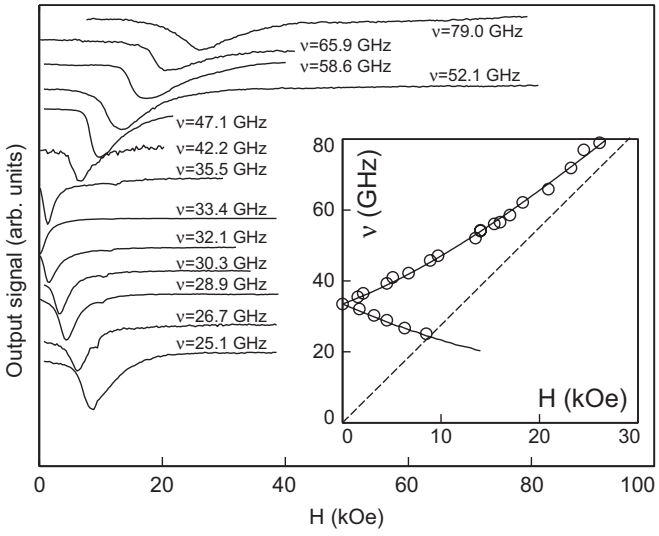

Fig. 2. Resonance absorption lines in $Gd_2Sn_2O_7$ at various frequencies for $T = 0.45$ K; the corresponding $\nu(H)$ diagram is shown on the inset; solid lines are guide-to-the-eye for the two branches, dashed line is a paramagnet with $g = 2.0$.

3. Conclusions

In summary, the experimental study of the magnetic resonance in two related strongly frustrated compounds $Gd_2Ti_2O_7$ and $Gd_2Sn_2O_7$ is carried out. In the disordered phase they demonstrate identical paramagnetic properties. The acoustic parts of their low-temperature spectra are also found similar, those in $Gd_2Ti_2O_7$ being consistent with the recently proposed 4*k*-structure. In the spin saturated phase of $Gd_2Ti_2O_7$, our experiment provides the first direct observation of the quasi-local soft modes in a pyrochlore magnet. Their behaviour under magnetic field agrees well with the spin–wave theory and confirms the previous suggestions on the origin of the enhanced magnetocaloric effect.

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