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Thermodynamic Properties of Geometrically Frustrated $S = 1/2$ XY Antiferromagnet $Er_2Sn_2O_7$

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The magnetic susceptibility, specific heat and entropy of geometrically frustrated $S = 1/2$ XY antiferromagnet $Er_2Sn_2O_7$ are reported. It is shown, that temperature dependence of magnetic susceptibility above nominally 10 K is governed by crystal field effects. Systematic study of specific heat in magnetic fields up to 7 T enabled the estimation of exchange coupling, $J/k << 4.6$ K. Investigation of isothermal change of magnetic entropy upon magnetization from 1 T to 7 T revealed the absence of enhanced magnetocaloric effect. The obtained results suggest that frustration parameter $f \approx 140$ proposed for $Er_2Sn_2O_7$ earlier may be significantly overestimated.

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1. Introduction

Geometrically frustrated magnetic systems are of interest due to novel ground states arising from inability of the system to minimize the energy of all pairwise interactions simultaneously. Rare-earth pyrochlores with general formula $RE_2A_2O_7$ (RE = Dy, Ho, Er, Gd, Yb, A = Ti, Sn, Mo) are compounds with high level of frustration. Unlike its counterpart $Er_2Ti_2O_7$, which orders magnetically at 1.2 K [1], no indication of long-range ordering has been reported in $Er_2Sn_2O_7$ down to 100 mK [2]. The comparable values of Curie-Weiss temperatures for both compounds suggest that the frustration index $f = \Theta_{cw}/T_N \approx 14/0.1 = 140$ of the $Er_2Sn_2O_7$ is significantly higher than that found in $Er_2Ti_2O_7$. The obtained value of f locates $Er_2Sn_2O_7$ to highly frustrated regime.

The purpose of the present work is to verify the proposed value of f by more detailed study of thermodynamic properties.

2. Experimental details

The compound $Er_2Sn_2O_7$ was synthesized following established procedure [2]. Subsequent X-ray analysis did not reveal impurities present in the sample. The structure of $Er_2Sn_2O_7$ consists of corner sharing tetrahedra, magnetic Er(III) ions are located in the corners. The local easy plane is oriented perpendicularly to the line connecting the centre of the tetrahedron and the corner. The magnetic susceptibility of polycrystalline sample was studied in commercial Quantum Design SQUID magnetometer from 2 K to 300 K in magnetic field of 0.1 T. Specific heat was investigated in Quantum Design PPMS system with ^{3}He insert from 0.4 K to 25 K in magnetic fields up to 7 T. Entropy was calculated numerically using standard extrapolations at low temperatures.

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3. Results and discussion

Temperature dependence of the inverse susceptibility is linear above 10 K, the deviation appears at lower temperatures, see Fig. 1, inset. The analysis of the susceptibility above 30 K using Curie-Weiss law yielded $\Theta_{cw} = -14$ K [2]. However, Θ_{cw} may not be the result of exchange coupling only, since at higher temperatures excited doublets may become populated. Therefore, temperature dependence of the population ratio between the ground state and the first and the second excited doublet was constructed, adopting the values of the energies from neutron scattering data [2]. As can be seen, the change of slope in inverse susceptibility at 10 K may be associated just with the occupation of higher crystal field (CEF) levels. Thus, the contribution of exchange coupling to Θ_{cw} may be significantly smaller.

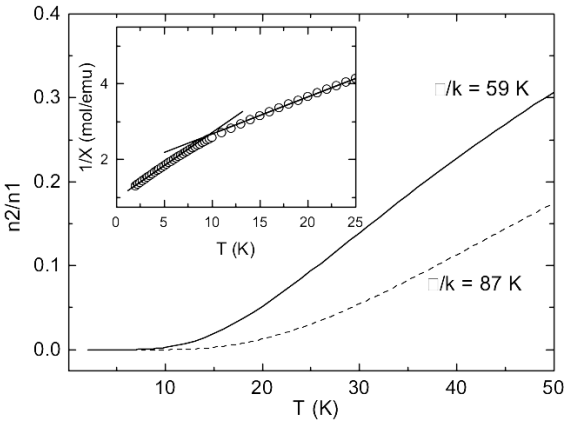

Fig. 1. Temperature dependence of occupation ratio between the ground state and the first excited state with $\Delta/k = 59$ K and second excited state with $\Delta/k = 87$ K. Inset: Detail of temperature dependence of the inverse susceptibility. Solid lines are guides for eyes.

Alternative rough estimation of the exchange interaction may be obtained from specific heat data below 10 K,

where higher CEF states are not thermally populated. For this purpose $Er_2Sn_2O_7$ specific heat data were compared with the prediction for $S = 1/2$ paramagnet with $g = 5.15$, obtained from ESR study [3]. Whereas for low magnetic fields the comparison fails completely, in magnetic field of 4 T, the major features of the data below 10 K are reproduced reasonably, see Fig. 2. This result suggests that at magnetic field of 4 T the energy scale is determined predominantly by Zeeman splitting of the doublet and not by the energy of exchange coupling. Consequently, adopting non-equation $g\mu_B BS >> zJS^2$, a rough estimation $J/k << 4.6$ K is obtained. This estimation shifts the originally proposed value of frustration parameter f to significantly lower values.

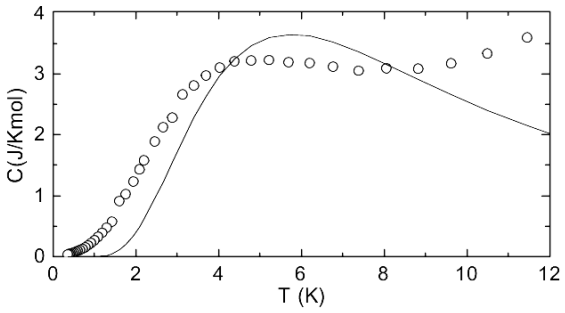

Fig. 2. Temperature dependence of specific heat of $Er_2Sn_2O_7$ (empty circles). Schottky contribution for two-level system is denoted by the solid line.

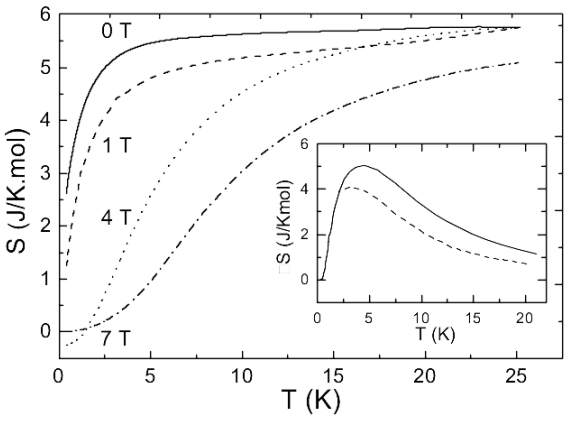

Fig. 3. Temperature dependence of entropy of $Er_2Sn_2O_7$ in various magnetic fields. Inset: Comparison of isothermal change of entropy of $Er_2Sn_2O_7$(dashed line) and $S = 1/2$ paramagnet (solid line) for magnetization from 1 T to 7 T.

It is known, that in geometrically frustrated systems the enhanced magnetocaloric effect may be anticipated, due to the condensation of localized magnon modes at saturation magnetic field [4]. This phenomenon was observed, for example in $Gd_2Ti_2O_7$ [5]. In order to clarify the potential existence of enhanced magnetocaloric effect in $Er_2Sn_2O_7$, temperature dependence of entropy in various magnetic fields was calculated from specific heat data. For magnetic fields in 0 T, 1 T and 4 T the saturation value of the entropy was adjusted at high temperatures to be equal to $S = 5.76$ J/K/mol, as anticipated for a system with spin 1/2, see Fig. 3. The small negative value of the entropy at low temperatures in magnetic field of 4 T appears due to the used approximation. On the other hand, no indication of the saturation is observed for magnetic field of 7 T, since this magnetic field splits the energy levels far from each other and in the used temperature range the excited level is not sufficiently populated. In order to clarify the magnetocaloric behavior of $Er_2Sn_2O_7$ the temperature dependence of isothermal change of the entropy was constructed for isothermal magnetization from 1 T to 7 T.

Such a field range may cover the saturation field for $Er_2Sn_2O_7$. Indeed, the saturation field for $Gd_2Ti_2O_7$ is about 6 T, whereas 1.7 T was proposed for $Er_2Ti_2O_7$ [1]. If the enhanced magnetocaloric effect in $Er_2Sn_2O_7$ is present, then its isothermal change of the entropy should be more pronounced than that for an $S = 1/2$ paramagnet under the same experimental conditions. The comparison suggests that no enhanced magnetocaloric effect is present in $Er_2Sn_2O_7$. The origin of this behavior may be attributed to strong planar anisotropy in the studied material.

4. Conclusions

The investigation of magnetic susceptibility and specific heat of $Er_2Sn_2O_7$ suggests significantly lower value of frustration parameter f, than previously reported. Magnetocaloric behaviour seems to be governed by single - ion anisotropy rather than frustration.

Acknowledgments

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