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KEr(MoO₄)₂ AND CsDy(MoO₄)₂ — LOW-DIMENSIONAL ISING MAGNETS

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In the present work the behaviour of cluster excitations in magnetic field, B , parallel to the easy axis was experimentally studied by the measurements of KEr(MoO₄)₂ and CsDy(MoO₄)₂ specific heat, C , vs. B dependence studied at a constant temperature. Corresponding Monte Carlo simulations of C revealed that despite of the fact that the Ising exchange-coupling models proved useful for a sufficient description of the specific heat of these dipolar systems in $B = 0$, the analysis of current experimental data requires a partial renormalization of exchange-coupling constants previously obtained from $B = 0$ analysis. The influence of crystal field effects and long-range character of spin correlations is discussed.

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Spin dynamics of one-dimensional (1D) magnetic systems has been a subject of intensive theoretical and experimental investigations for a few decades. Several classes of 1D magnetic materials have been reported, enabling the study of the influence of the anisotropy and spin value on the character of the ground state and magnetic excitations [1]. One of the extensively studied classes of low-dimensional systems is represented by a series with a general formula AR(MO₄)₂, where A = Cs, K, Rb... , R = La, Ce, Pr,...,Lu and M = Mo, W. In these ionic compounds with a layered crystal structure the R³⁺ ions occupy sites which are sufficiently far apart from one another so that a dipolar coupling represents the dominating interaction among magnetic moments [2, 3].

KEr(MoO₄)₂ and CsDy(MoO₄)₂ represent the best-studied compounds from the group of rare-earth dimolybdates. The ground state of a free Er³⁺ ion is ⁴I_{15/2}. In the KEr(MoO₄)₂ lattice the degenerated electronic state is split into eight Kramers doublets with the lowest energies $E_0 = 0$ and $E_1 = 15 \pm 2$ cm⁻¹. The Ising character of this compound is quite well established by a strong g -factor anisotropy of the lowest doublet with $g_a = 1.8$, $g_b < 0.9$ and $g_c = 14.7$. The g -factors of the higher doublet are $g'_a = 2.3$ and $g'_b, g'_c < 1$ [2]. Theoretical calculations considering

only dipolar interactions and crystal-field effects have shown that the magnetic structure of the ground state consists of ferromagnetic chains parallel to the c -axis with magnetic moments oriented along the chains [2]. The moments in neighbouring chains are coupled antiferromagnetically. The authors suggest that in the pure dipolar approach the compound can be treated as a quasi-1D magnetic system. The suggestions were later confirmed by $\text{KEr}(\text{MoO}_4)_2$ specific heat studies [4]. The analysis of the data in zero magnetic field performed within nearest-neighbour exchange-coupling model has revealed that in the temperature range from 0.5 K to 5 K the magnetic system can be approximated by $S = 3/2$ Blume–Capel model on rectangular lattice with intrachain interaction $J'_1/k_B = 0.38$ K, interchain interaction $J'_2/J'_1 \approx -0.2$ and single-ion anisotropy $D/k_B = 10$ K. It was found that for the given set of parameters at temperatures lower than 2 K this model converges to $S = 1/2$ Ising model on the rectangular lattice with effective intrachain interaction $J_1/k_B = 0.85$ K and effective interchain interaction $J_2/J_1 \approx -0.2$. (The J' and J are related by the transformation $J' = (4/9)J$ due to different spin magnitudes of the used models.) Preliminary studies of the contribution of spin cluster excitations to the specific heat in non-zero magnetic field applied along the c -axis was reported in [4]. The analysis of the data performed within the same models revealed that the data in $B \neq 0$ require renormalization of the J'_1 , J'_2 parameters. Two possible mechanisms were suggested to explain the origin of the renormalization; existence of two non-equivalent magnetic centres in the $\text{KEr}(\text{MoO}_4)_2$ unit cell (in other words a non-zero tilting angle between local anisotropy axes of both centres) and the dipolar nature of magnetic correlations.

This work is devoted to the detailed experimental study of magnetic field dependence of $\text{KEr}(\text{MoO}_4)_2$ specific heat at a constant temperature in magnetic fields applied parallel to the c -axis which was declared by the previous studies to be the easy axis of the magnetic system. The role of a tilting angle in the aforementioned renormalization is experimentally investigated in a similar dipolar system $\text{CsDy}(\text{MoO}_4)_2$.

The magnetic field dependence of $\text{KEr}(\text{MoO}_4)_2$ specific heat was experimentally studied by using a standard quasi-adiabatic calorimetry [4] at the constant temperature $T = 4.27$ K in magnetic fields up to 1 T. Since the studied compound can be treated as a magnetic insulator, the lattice contribution was separated using the method described elsewhere [4]. The resultant magnetic specific heat is shown in Fig. 1. It can be seen that these new more detailed measurements re-confirmed the fact that the renormalization of J'_1 , J'_2 parameters of $S = 3/2$ Blume–Capel model is required to achieve a better agreement between the theory and experiment. A previous study of phase transitions in magnetic field [5] revealed the influence of exchange interaction. It was found that in the a -direction it becomes comparable with the strength of dipolar interactions so that J'_1 , J'_2 parameters of $S = 3/2$ Blume–Capel model represent some effective parameters $J'_{\text{eff}} = J'_{\text{dip}} + J'_{\text{exch}}$ where the dipolar part will be effectively changed in the magnetic field.

Further potential mechanism responsible for the observed deviations in Fig. 1 can be the influence of the tilting angle between local anisotropy axes of Er^{3+} ions in the unit cell. The tilting angle does not exceed $\approx 20^\circ$ [2]. Since the experiments

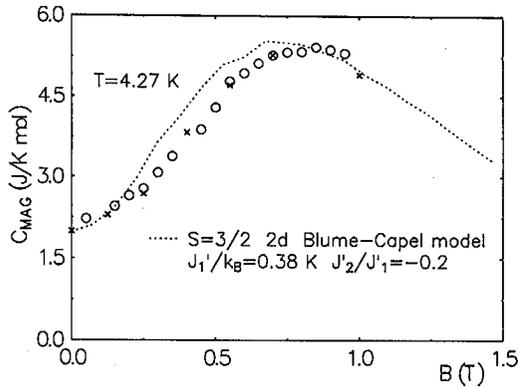


Fig. 1. Magnetic field dependence of $KEr(MoO_4)_2$ specific heat at $T = 4.27$ K; o — present paper, x — paper [4].

were performed in the orientation $B \parallel c$, the tilting angle between the field and local anisotropy axes of both sets of Er^{3+} ions was the same, being equal to $\pm 10^\circ$. To estimate experimentally the role of the tilting angle, the $CsDy(MoO_4)_2$ specific heat vs. magnetic field dependence is to be measured in $B \parallel c$. In this compound there are two non-equivalent magnetic ions with the mutual tilting angle 10° [6], thus in the orientation $B \parallel c$ the tilting angle between the field and local anisotropy axes of both sets of Dy^{3+} ions will be equal to $\pm 5^\circ$. This low value of the angle should not lead to the deviations observed in Fig. 1.

Recent theoretical studies of the system considering only dipolar nature of magnetic correlations and crystal field effects have revealed that the ground-state configuration has the same chain character as that predicted for $KEr(MoO_4)_2$ [6]. However, in comparison with $KEr(MoO_4)_2$, the strength of interchain interactions is much larger. This fact corresponds with previous specific heat studies in $B = 0$ indicating that the system can be described by $S = 1/2$ Ising model on the square lattice with $J/k_B = 0.56$ K [7]. To estimate magnetic field dependence of $CsDy(MoO_4)_2$ specific heat, the results of [8] were used where the temperature dependence of specific heat in a constant magnetic field $B \parallel c$ was studied.

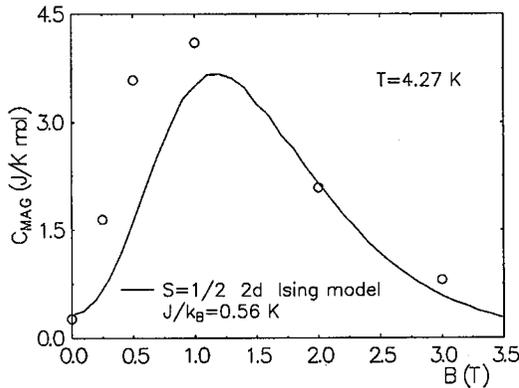


Fig. 2. Magnetic field dependence of $CsDy(MoO_4)_2$ specific heat at $T = 4.27$ K (o).

The obtained "sketch" of the dependence is in Fig. 2. The data are compared with the behaviour of $S = 1/2$ Ising model on the square lattice with intralayer exchange interaction $J/k_B = 0.56$ K in $B \parallel z$. The theoretical prediction was calculated by the Monte Carlo method with parameters described in detail elsewhere [4]. Despite of the preliminary character of the data, it is clear that also in this case a different value of J should be used to improve the agreement with the experiment. Since the tilting angle in $\text{CsDy}(\text{MoO}_4)_2$ is much lower than that in $\text{KEr}(\text{MoO}_4)_2$, it is obvious that mainly a dipolar nature of magnetic correlations will be responsible for changing of effective parameters of the used exchange-coupling models. This observation revealed that at least in the case of specific heat the role of magnetic non-equivalence is not so important as it was assumed earlier. In future, a more detailed experimental study of $\text{CsDy}(\text{MoO}_4)_2$ specific heat vs. B dependence at a constant temperatures is to be performed which together with presented $\text{KEr}(\text{MoO}_4)_2$ data will be used to verify the aforementioned assumption on the dipolar interactions. The theoretical study of the specific heat could give a clue to the insight into the microscopic processes e.g. the character of spin cluster excitations formed in magnetic field in low-dimensional Ising systems with long-range interactions [4].

Acknowledgments

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