The Rare-earth Based Single-ion Magnet CsNd(MoO$_4$)$_2$

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Specific heat, magnetization and DC susceptibility of the single crystal CsNd(MoO$_4$)$_2$ have been investigated nominally, in the temperature range from 100 mK to 300 K and magnetic field up to 5 T. The systems offer a possibility to study low-dimensional phenomena with the strong magnetic anisotropy, changing from the easy-plane to the easy-axis type [1]. The measurements of the specific heat, DC susceptibility and magnetization of CsNd(MoO$_4$)$_2$ have been performed to identify the magnetic subsystem of the compound. The results of the data analysis will serve as a starting point to the investigation of the spin dynamics determining the response of the studied SIM to the application of a high-frequency magnetic field.

1. Introduction

The present work is devoted to the study of CsNd(MoO$_4$)$_2$, the candidate for novel mononuclear lanthanide-based single-ion magnets (SIMs). The compound belongs to the group of the rare-earth dimolybdates ARE(MoO$_4$)$_2$, where A represents alkaline metal. The systems offer a possibility to study low-dimensional phenomena with the strong magnetic anisotropy, changing from the easy-plane to the easy-axis type [1]. The measurements of the specific heat, DC susceptibility and magnetization of CsNd(MoO$_4$)$_2$ have been performed to identify the magnetic subsystem of the compound. The results of the data analysis will serve as a starting point to the investigation of the spin dynamics determining the response of the studied SIM to the application of a high-frequency magnetic field.

2. Experimental details

CsNd(MoO$_4$)$_2$ crystallizes in the orthorhombic system (space group $D_{2h}^2$) with the cell parameters $a = 9.55$ Å, $b = 8.23$ Å, and $c = 5.13$ Å [2]. The ground multiplet of Nd$^{3+}$ is $^4I_9/2$ and is split in the crystal field of low-symmetry into 5 doublets. Specific heat has been experimentally studied in the temperature range from 120 mK to 2 K in zero magnetic field using a dual-slope method. The sample with mass 24.9 mg was attached to the heat reservoir in the dilution $^3$He-$^4$He refrigerator. Specific heat measurement in the temperature range from 2 to 20 K and magnetic field up to 4 T has been performed in the commercial Quantum Design PPMS device on the sample with $m = 3.7$ mg. The sample was glued on "L" shaped silver holder to achieve the orientation of the magnetic field parallel to the $c$ axis. DC susceptibility and magnetization studies of the single crystal with $m = 13.7$ mg have been performed in the same magnetic-field orientation in the commercial Quantum Design SQUID magnetometer in the temperature range from 2 to 300 K and magnetic fields up to 5 T.

3. Results and discussion

Temperature dependence of the specific heat in zero magnetic field is characterized by a slight decrease down to 1 K and a significant upturn appearing below 0.4 K (Fig. 1). CsNd(MoO$_4$)$_2$ is a magnetic insulator, thus the total specific heat consists of the phonon and magnetic contribution, the former manifesting above 1 K as $bT^2$ dependence and the latter as $a/T^2$. The fitting procedure in the temperature interval between 1 and 2 K yielded estimations of the separate contributions with $a = 0.058$ J/Kmol and $b = 0.0015$ J/K$^2$mol. Rather steep upturn at low temperatures reflects the formation of magnetic correlations. Previous specific heat studies of CsGd(MoO$_4$)$_2$ [3] and CsDy(MoO$_4$)$_2$ [4] observed a phase transition to the ordered magnetic state at 0.45 K and 1.3 K, respectively. In both cases, the specific heat near the phase transition reached the values 25–30 J/Kmol while the specific heat of CsNd(MoO$_4$)$_2$ is about 2 J/Kmol at the lowest temperature, $T = 120$ mK. The magnetic entropy, $S = 1.62$ J/Kmol, calculated in the whole experimental temperature interval represents only 30% of the maximal entropy of the system with an effective spin $1/2$. The low values of the entropy and specific heat indicate that the transition to the magnetic
The rare-earth based single-ion magnet CsNd(MoO$_4$)$_2$ will be located at temperatures well below 120 mK. Applied magnetic field induces a round anomaly shifting to higher temperatures with increasing magnetic field [Fig. 1 (inset)].

The behavior of the magnetic specific heat in non-zero magnetic field (Fig. 2) can be well described with a standard two-level model. The application of the model yielded the best agreement with the $g$-factor value 3.08.

![Fig. 1. The temperature dependence of the total specific heat of CsNd(MoO$_4$)$_2$ in zero magnetic field (circle). The solid line represents a phonon contribution $C_{ph} = bT^3$. Inset: The temperature dependence of the heat capacity of CsNd(MoO$_4$)$_2$ in nonzero magnetic fields.](image1)

![Fig. 2. The temperature dependence of the magnetic specific heat $C_{mag}$ of CsNd(MoO$_4$)$_2$ obtained from the total specific heat by subtracting a phonon contribution $bT^3$. The solid lines correspond to the specific heat of a two-level model with $g = 3.08$.](image2)

The analysis suggests that higher energy doublets do not contribute to the specific heat at low temperatures and CsNd(MoO$_4$)$_2$ can be approximated by the model of an ideal paramagnet with the effective spin $S' = \frac{1}{2}$ at temperatures above 2 K. Accordingly, the magnetic field dependence of the magnetization at the constant temperature 5 K can be well described using Brillouin function with $g = 3.08$ (Fig. 3). Similarly, DC susceptibility data can be fitted to Curie-Weiss law (Fig. 3 insert) with Curie paramagnetic temperature $\theta = -1.05$ K and $g = 3.3$ when assuming $S' = \frac{1}{2}$.

4. Conclusions

Analysis of the specific heat, magnetization and DC susceptibility indicates very weak magnetic correlations in CsNd(MoO$_4$)$_2$ and dominant influence of the crystal field produced by the first coordination sphere of Nd$^{3+}$ ion. The crystal field is responsible for rather large energy separation between the ground and first excited doublet. These features suggest CsNd(MoO$_4$)$_2$ can be considered as a representative of a single-ion magnet in which magnetic relaxation can be expected. In future, the study of the dynamic properties will be performed.

Acknowledgments

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References