Proceedings of the 15th Czech and Slovak Conference on Magnetism, Košice, Slovakia, June 17–21 2013

The Rare-earth Based Single-ion Magnet $CsNd(MoO_4)_2$

V. Tkáč[a,*], K. Tibenská[b], A. Orendáčová[a], M. Orendáč[a], A. G. Anders[c], V. Pavlík[d], A. Feher[a]

[a]Institute of Physics, Faculty of Sciences, P. J. Šafárik University, Park Angelinum 9, 041 54 Košice, Slovakia
[b]Faculty of Aeronautics, Technical University, Rampová 7, 041 21 Košice, Slovakia
[c]Institute of Low Temperature Physics and Engineering, Lenin Av. 47, 3 101 64 Kharkov, Ukraine
[d]Institute of Experimental Physics SAS, Watsonova 47, 040 01 Košice, Slovakia

Specific heat, magnetization and DC susceptibility of the single crystal $CsNd(MoO_4)_2$,a layered rare-earth dimolybdate, have been investigated nominally, in the temperature range from 100 mK to 300 K in the magnetic field up to 5 T, applied along the a axis. The analysis of the experimental data revealed the absence of a phase transition to the magnetic ordered state down to 100 mK. The application of a standard two-level model yielded an excellent agreement with the specific heat data above 2 K in nonzero magnetic field indicating a weakness of magnetic correlations and a predominant occupation of the ground-energy doublet. The latter indicates a large energy separation between the ground and first excited doublet. These measurements suggest that $CsNd(MoO_4)_2$ can represent a good realization of a single-ion magnet.

DOI: 10.12693/APhysPolA.126.244

PACS: 75.40.-s, 75.60.-d

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 alkali 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^3_{2h}) 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^3 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^{4}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 $\sim$ 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

*corresponding author; e-mail: vladimir.tkac@student.upjs.sk

ordered state 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 nonzero 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.

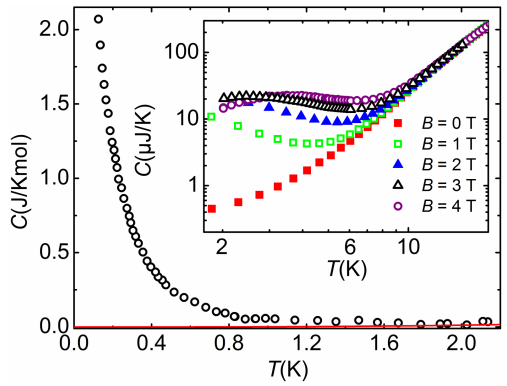

Fig. 1. The temperature dependence of the total specific heat of $CsNd(MoO_4)_2$ in zero magnetic field (circle). The solid line represents a phonon contribution $C_{ph} = bT^3$. Inset: The temperature dependence of the heat capacity of $CsNd(MoO_4)_2$ in nonzero magnetic fields.

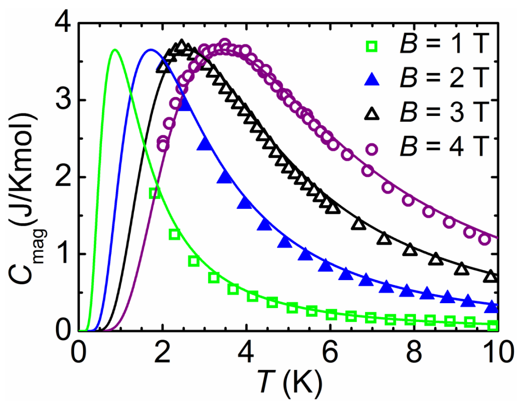

Fig. 2. The temperature dependence of the magnetic specific heat C_{mag} of $CsNd(MoO_4)_2$ obtained from the total specific heat by subtracting of a phonon contribution bT^3. The solid lines correspond to the specific heat of a two-level model with $g = 3.08$.

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)

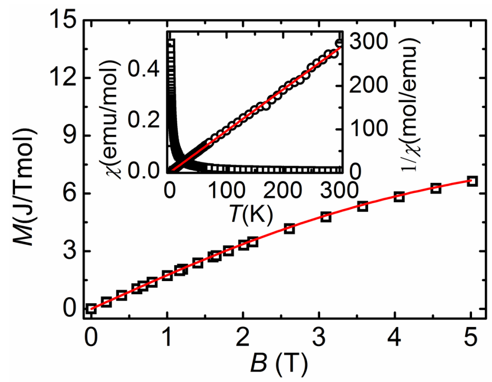

Fig. 3. The field dependence of the magnetization at the constant temperature $T = 5$ K. The solid line represents Brillouin function with $g = 3.08$. Inset: Temperature dependence of the DC susceptibility (square) and the inverse susceptibility (circle). The solid line represents fit to Curie-Weiss law.

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

This work was supported by the projects APVV 0132-11, ITMS 26220120005 and VEGA 1/0143/13. Financial support from US Steel DZ Energetika is greatly acknowledged.

References

[1] A. Orendáčová, M. Orendáč, A. Feher, *Czech. J. Phys.* **52**, 307 (2002).

[2] A.A. Pavlyuk, L.P. Kozeeva, K.G. Folin, V.G. Gladyshev, V.S. Gulyaev, V.S. Pivtsov, A. A Kaminskii, *Inorg. Mater* **19**, 767 (1983).

[3] A. Feher, P. Štefányi, A. Orendáčová, E.E. Anders, A.G. Anders, S.V. Volotskii, A.I. Zvyagin.S.V. Startsev, E.N. Khatsko, A.S. Chernyi, *Fiz. Niz. Temp.* **14**, 1304 (1988).

[4] P. Štefányi, A. Feher, A. Orendáčová, A.S. Chernyi, *Fiz. Niz. Temp.* **16**, 224 (1990).