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Thermal Conductivity of a Layered $CsGd(MoO_4)_2$ Crystal

K. Tibenská[a,*], V. Tkáč[b], A. Orendáčová[b], M. Orendáč[b], A. Feher[b], J. Šebek[c] and V. Sechovský[d]

[a]Faculty of Aeronautics, Technical University, Rampová 7, 041 21 Košice, Slovak Republic

[b]Institute of Physics, P.J. Šafárik University, Park Angelinum 9, 040 00 Košice, Slovak Republic

[c]Institute of Physics AS CR, Na Slovance 2, 18221 Prague, Czech Republic

[d]Charles University in Prague, Faculty of Mathematics and Physics, Ke Karlovu 5, 12116 Prague, Czech Republic

The thermal conductivity of $CsGd(MoO_4)_2$ has been studied in the temperature range from 2 to 50 K in zero magnetic field. The analysis of the data performed within the Debye model with the relaxation-time approximation revealed the presence of the scattering of phonons by critical fluctuations. The behaviour of phonon mean free path at the lowest temperatures is discussed.

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1. Introduction

Rare-earth dimolybdates besides their practical use in laser technology, represent a good realization of low-dimensional Ising and XY models with spin 1/2 [1, 2]. In these layered crystal structures, lattice instability resulting from the electron–phonon coupling and orbital degeneracy has been detected producing the variety of structural phase transitions. The study of the thermal conductivity of $CsDy(MoO_4)_2$ [2] confirmed the relation of the observed Jahn–Teller transition with the phonon subsystem while the field induced Jahn–Teller transition in $KEr(MoO_4)_2$ at low temperatures is not fully understood [3]. The single crystal of $CsGd(MoO_4)_2$ has been prepared by the flux method. The title compound $CsGd(MoO_4)_2$ crystallizes in the orthorhombic system (space group D_{2h}^3) with the unit cell parameters $9.52 \times 5.07 \times 8.05$ Å^3. In the cleavage plane (100), the crystal layers contain chains of Gd^{3+} ions running along the c axis. The ground state of the Gd^{3+} ions is $^8S_{7/2}$. Previous studies [4] determined the intrachain coupling $J_1/k_B \approx 0.6$ K, interchain coupling $J_2 \approx 0.03J_1$ and the Debye temperature, $\Theta_D = 174$ K. The onset of the magnetic long range order has been observed at 0.45 K.

The present work deals with the analysis of the thermal conductivity of $CsGd(MoO_4)_2$ to search for the presence of spin-phonon coupling expected in the system.

2. Experimental details

The thermal conductivity was measured using the steady-state one-heater method in the temperature range from 2 to 50 K in zero magnetic field. The sample was cut in the shape of a rectangular parallelepiped with the shortest dimension $L = 0.53$ mm. The measurement was performed in a Physical Property Measurement System (PPMS) from Quantum Design. A commercially available sample holder for electrical resistance measurement was modified for the measurement of thermal conductivity. Two Lake Shore CX-1050-BC thermometers and a VITROHM heater were attached directly to a sample of dimensions $3.7 \times 0.53 \times 8$ mm^3. Heat power was applied along the c axis. The accuracy of the measurement was 5%.

3. Results and discussion

$CsGd(MoO_4)_2$ belongs to magnetic isolators, thus phonons and magnetic excitations can be considered as heat carriers. The average velocity of the phonons $v_{fon} = 2130$ m/s was estimated from the Debye temperature. The real velocity of phonons can be lower than the calculated value, as the calculation was based on the average Debye temperature. Given very low values of magnetic interactions (≈ 0.5 K), it can be assumed that the velocity of the magnons $v_{mag} \sim JS^2$ will be at least one order of magnitude lower than the acoustic phonon velocity. The comparison of v_{fon} and v_{mag} suggests that in $CsGd(MoO_4)_2$ the phonons will be dominant heat carriers, and the magnetic excitations will scatter the phonons. In the thermal conductivity analysis, the mean free phonon path, l, is an important parameter which should achieve a saturated value at lowest temperatures close to L, the shortest crystal dimension.

The temperature dependence of the mean free phonon path (Fig. 1) has been determined from the kinetic equation $\kappa = Cvl/3$. The high temperature phonon specific heat C of $CsGd(MoO_4)_2$ was approximated by the specific heat of the isostructural diamagnetic $KLu(MoO_4)_2$ [5]; the good coincidence of both the lattice specific heats was tested at low temperatures. The large difference between the L value and the calculated maximum mean free path L_0 is evident (Fig. 1) and can be caused by following reasons: (i) $CsGd(MoO_4)_2$ has a layered structure and therefore the phonon scattering occurs at the inner surfaces; (ii) the estimated phonon velocity can be smaller than the calculated value, therefore the mean free phonon path will be larger (iii); the

* corresponding author; e-mail: katarina.tibenska@tuke.sk

mean free phonon path depends also on the phonon scattering on structural defects and on the magnetic subsystem. Correspondingly, the thermal conductivity was analyzed using the Debye model with the approximation of the relaxation time including phonon scattering at sample boundaries, point defects and Umklapp scattering represented by the total relaxation time $\tau^{-1} = v/L + P\omega^4 + UT\omega^3 \exp(-\Theta_D/uT)$. U, u and P are free parameters and ω is the phonon frequency.

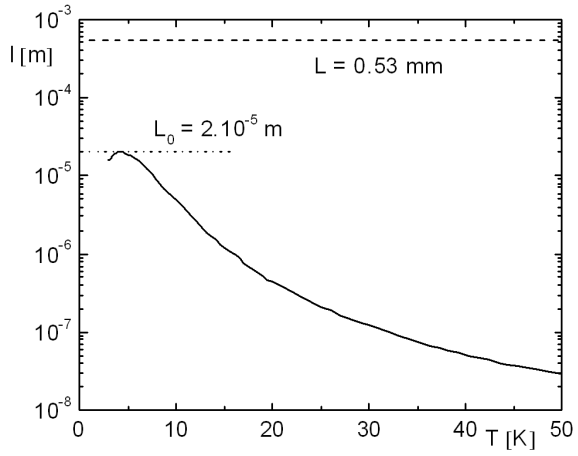

Fig. 1. The temperature dependence of the mean free phonon path in $CsGd(MoO_4)_2$ (solid line). The dashed line represents the lowest crystal dimension.

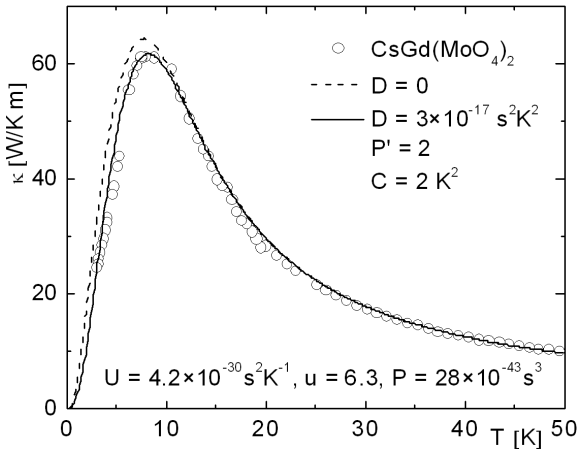

Fig. 2. The temperature dependence of thermal conductivity of $CsGd(MoO_4)_2$. The dashed line represents the theoretical prediction of the thermal conductivity, including the boundary scattering, Umklapp process and scattering on point defects. Solid line represents the same theoretical prediction when including the effect of the volume magnetostriction.

Considering only the mentioned scattering processes, a difference between the theoretical prediction and experimental data was observed at temperatures below 10 K (Fig. 2). In $CsGd(MoO_4)_2$ the formation of magnetoelastic modes resulting from the single-ion magnetostriction is expected due to the crossing of acoustic phonon branches and spin energy levels. The effect manifests as a resonance scattering of phonons on magnons which can be observed at the temperature $T_0 \approx E/4k_B$. When considering crystal field splitting [4] and magnetic interactions, the highest spin energy level is roughly $E/k_B \approx 2$ K, correspondingly, the resonance scattering should occur at about ≈ 0.5 K. The estimated temperature well coincides with the temperature of the phase transition to the ordered state T_N, which implies the combination of the single-ion and volume magnetostriction. Including the relaxation time corresponding to the single-ion magnetostriction did not improve the agreement between the data and theory. On the other hand, including the phonon scattering on the critical fluctuations resulting from the presence of the volume magnetostriction, $\tau^{-1} = D\omega^2/[(T - T_N)^{P'} + C]$, where D, P' and C are free parameters, significantly improved the agreement between the data and theory (Fig. 2).

4. Conclusion

In conclusion, the analysis of the thermal conductivity of $CsGd(MoO_4)_2$ revealed that the dominant heat carriers are phonons, which are scattered by critical fluctuations of the magnetic subsystem. In future, further studies at lower temperatures and nonzero magnetic field will be performed.

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