14th Czech and Slovak Conference on Magnetism, Košice, Slovakia, July 6–9, 2010

Scattering of Phonons in $CsMnCl_3 \cdot 2H_2O$

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The thermal conductivity of the quasi-one-dimensional $S = 5/2$ Heisenberg antiferromagnet $CsMnCl_3 \cdot 2H_2O$ with the intrachain interaction $J/k_B = 3$ K was experimentally studied at temperatures from 2 to 25 K. The data analysis performed within the Debye model with the relaxation-time approximation unambiguously indicates the presence of the scattering of phonons on magnetic subsystem.

PACS numbers: 75.30.Et, 75.40.Cx, 66.70.Lm, 75.30.Ds

1. Introduction

Recent theoretical predictions of the ballistic heat transport in a $S = 1/2$ Heisenberg chain model renewed the interest in experimental studies of the thermal conductivity of low-dimensional magnets [1]. However, in real systems magnons interact with the phonon bath, which leads to the mutual scattering of heat carriers.

This work deals with experimental studies of the heat transport in $CsMnCl_3 \cdot 2H_2O$ (CMC), a quasi-one-dimensional $S = 5/2$ Heisenberg antiferromagnet with the intrachain interaction $J/k_B = 3$ K [2]. The magnetic system undergoes a phase transition to the ordered state at $T_N = 4.89$ K. The analysis of the thermal conductivity data suggests that the phonons represent the main heat carriers in CMC.

2. Experimental details

CMC crystallizes in the orthorhombic system ($Pcca$ space group), the unit cell parameters $a = 9.060$ Å, $b = 7.285$ Å, $c = 11.455$ Å. The thermal conductivity was measured with the standard steady-state heat-flow method with one heater in the temperature range from 2 to 25 K. The heat power was applied along the magnetic chains running along the a axis. Two samples were cut from a piece of a single crystal to the shape of a rectangular parallelepiped with the following dimensions: $13 \times 1.6 \times 0.75$ mm^3 (I) and $6 \times 1 \times 0.3$ mm^3 (II), respectively. The surface of the latter was further mechanically treated. The thermal conductivity of the sample (I) was studied in a commercial PPMS device with a TTO option, while a special sample holder [3] was used for the sample (II).

3. Results and discussion

The temperature dependence of the thermal conductivity of both samples is characterized by appearance of a round maximum at about 5 K (Fig. 1a). While the maximum of sample (I) possesses a double structure, the feature measured for the sample (II) is wide and flat. The thermal conductivity is sample dependent at least at the temperatures up to 15 K. CMC represents a magnetic insulator, thus only phonons and magnons can be considered as potential heat carriers. The velocity of acoustic phonons was approximated by a sound velocity experimentally studied at room temperature [4] providing the longitudinal velocity of sound $v_l = 4000$ m s^{-1} and the transversal components $v_{t1} = v_{t2} = 1500$ m s^{-1}.

Following the approach described in Ref. [5], we calculated the average value of the velocity, $v = 1562$ m s^{-1} using the equation $v = v_l \frac{2s^2+1}{2s^3+1}$, where $s = \frac{v_l}{v_t}$. The magnon group velocity was estimated by straightforward differentiating the experimental spin excitation spectra, $v = \frac{d\omega}{dq}$, obtained by inelastic neutron scattering experiments at low temperatures [6]. Then the intrachain magnon group velocity near the Brillouin zone centre ($q_x \approx 0$), was calculated, $v_M = 2070$ m s^{-1}. The comparison of the phonon and intrachain magnon velocities suggests that

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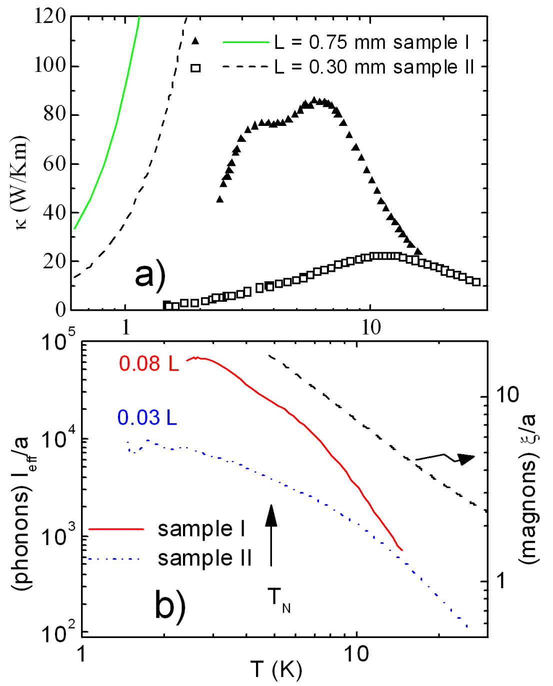

Fig. 1. (a) Temperature dependence of thermal conductivity of CMC. The lines represent the relation $k = \frac{1}{3}CvL$. (b) Temperature dependence of phonon mean free path and intrachain magnetic correlation length.

both types of carriers should mediate the heat transport, while the interchain magnons will act as scattering centers.

However, in the paramagnetic phase above $T_{\rm N}$, all magnons are strongly damped due to a small ordered region, characterized by the value of the correlation length ξ calculated for a classical Heisenberg chain with the spin $S = 5/2$ (Fig. 1b). The calculations provide the estimation of ξ at $T_{\rm N}$, being equal $\approx$ 20 lattice parameters, which indicates that in CMC phonons are the main heat carriers.

Assuming the dominance of the phonon heat transport, we calculated the phonon free path l in the wide temperature range using the relation from kinetic theory of gases, $k = \frac{1}{3}Cvl$, where k is the experimental thermal conductivity, C is the volume lattice heat capacity of CMC derived in Ref. [2] and v is the average phonon velocity (Fig. 1b).

In the absence of a magnetic subsystem and structural defects, the saturated l value should roughly correspond to the shortest dimension L of a crystal [5]. It is evident that at lowest temperatures the mean free path is nearly two orders of magnitude lower than the shortest dimension of the crystal. Since the low temperature l value achieved nearly saturation, an additional scattering mechanism of phonons can be expected.

In CMC, we can expect the additional phonon scattering on magnetic excitations as well as the scattering on external surfaces arising from a layered crystal structure. The presence of the additional scattering effects at low temperatures is evident from the comparison of low-temperature experimental data and results of theoretical calculation of the thermal conductivity based on the kinetic theory of gases; individual parameters have already been described (Fig. 1a).

Besides the phonon boundary scattering, other scattering mechanisms should be involved, namely the scattering on point defects and Umklapp processes. For this purpose, we used Debye model with the relaxation time approximation [5]. In this approximation, the thermal conductivity can be calculated

$$k = \frac{k_{\rm B}}{2\pi^2 v}\left(\frac{k_{\rm B}}{\hbar}\right)^3 T^3 \int_0^{\frac{\Theta_{\rm D}}{T}} \tau(x)\frac{x^4\,{\rm e}^x}{({\rm e}^x-1)^2}\,{\rm d}x\,.$$

We considered the boundary phonon scattering with $\tau_{\rm bd}^{-1} = \frac{v}{L}$, the phonon Umklapp processes characterized by $\tau_{\rm um}^{-1} = UT\omega^3\,{\rm e}^{-\Theta_{\rm D}/uT}$ and the scattering by point defects with $\tau_{\rm pt}^{-1} = P\omega^4$. Then considering

$$\frac{1}{\tau} = \frac{1}{\tau_{\rm bd}} + \frac{1}{\tau_{\rm um}} + \frac{1}{\tau_{\rm pt}}\,,$$

a fitting procedure was performed.

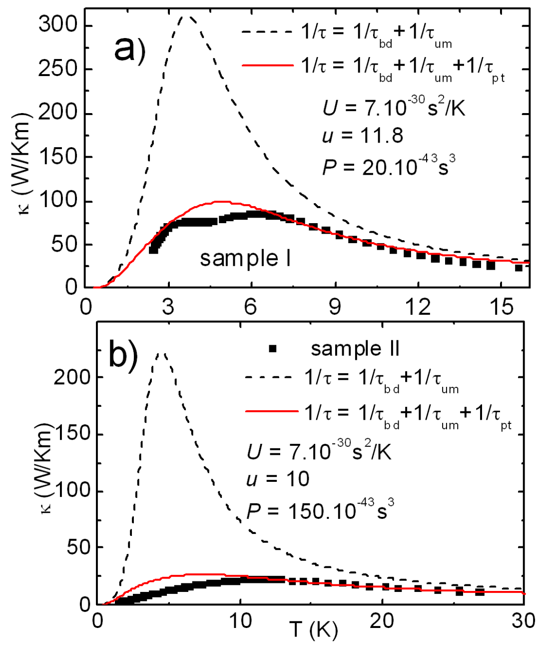

Fig. 2. Temperature dependence of thermal conductivity of CMC. Lines represent theoretical predictions (see text).

Assuming $\Theta_{\rm D} = 205$ K, $v = 1562$ m s^{-1}, $L = 0.75$ mm and 0.3 mm, the best agreement has been achieved for the parameters u, U, P listed in Fig. 2. Despite the introduction of other scattering processes, a more sophisticated analysis is necessary to explain such subtle features as the double maximum or behavior at lowest temperatures.

4. Conclusion

In conclusion, magnon and phonon velocities have been estimated. The results together with the calculated phonon mean free path in CMC suggest that phonons are the main heat carriers and magnons should act as scattering centers. The analysis of the thermal conductivity within a Debye model with the relaxation-time approximation indicates the need to incorporate other scattering processes. The scattering of phonons by a magnetic subsystem will be a subject of future studies.

Acknowledgments

This work has been supported by VEGA grant 1/0078/09, project APVT-0006-07 and ERDF EU project No. ITMS26220120005. Financial support from US Steel DZ Energetika is greatly acknowledged. The work of V.S. was supported by the grant LA308 of the Ministry of Education of Czech Republic.

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