Thermal Conductivity in the Triangular-Lattice Antiferromagnet $Ba_3CoSb_2O_9$

K Naruse[1], T Kawamata[1], M Ohno[1], Y Matsuoka[1], H Sudo[1], H Nagasawa[1], Y Hagiya[1], T Sasaki[2] and Y Koike[1]

[1] Department of Applied Physics, Tohoku University, Sendai 980-8579, Japan
[2] Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

E-mail: `tkawamata@teion.apph.tohoku.ac.jp`

Abstract. We have measured the thermal conductivity in the *ab*-plane, $\kappa_{\rm ab}$, and along the *c*-axis, $\kappa_{\rm c}$, of single crystals of the $S = 1/2$ triangular-lattice antiferromagnet $Ba_3CoSb_2O_9$ in zero field and magnetic fields. In zero field, it has been found that both $\kappa_{\rm ab}$ and $\kappa_{\rm c}$ show a similar broad peak around 40 K, suggesting that the thermal conductivity due to phonons is dominant in this compound and that the mean free path of phonons is suppressed so much by strong magnetic-fluctuations due to the magnetic frustration. It has also been found that both $\kappa_{\rm ab}$ and $\kappa_{\rm c}$ show a kink at the antiferromagnetic transition temperature $T_{\rm N}$. In magnetic fields parallel to the *ab*-plane up to 14 T, magnetic-field dependences of both $\kappa_{\rm ab}$ and $\kappa_{\rm c}$ at temperatures below and above $T_{\rm N}$ have been found to be understood taking into account the phonon-magnon scattering in the so-called 120° structure phase and the up-up-down (UUD) phase, while the minima of $\kappa_{\rm ab}$ and $\kappa_{\rm c}$ observed in the middle of the UUD phase have not been understood. This suggests the occurrence of some change of the magnetic state in the middle of the UUD phase.

1. Introduction

Thermal conductivity is a useful probe to investigate the change of a magnetic state and phase transitions, because the change and phase transitions affect the thermal conductivity due to phonons, $\kappa_{\rm phonon}$, via the phonon-magnon scattering. For example, the thermal conductivity shows various changes by the application of magnetic field in several spin-gap systems such as $SrCu_2(BO_3)_2$ [1, 2], $TlCuCl_3$ [3, 4] and $Ba_3Mn_2O_8$ [5], because the number of magnetic excitations scattering phonons is determined by the magnitude of the spin gap depending on magnetic field, leading to the change of $\kappa_{\rm phonon}$. Recently, furthermore, interesting behaviors of thermal conductivity have been reported in various geometrically frustrated pyrochlore antiferromagnets $R_2Ti_2O_7$ (R : rare-earth elements) [6, 7]. In $Tb_2Ti_2O_7$, extremely low values of $\kappa_{\rm phonon}$ have been observed in the spin-liquid state due to the strong magnetic-fluctuations [6]. In $R_2Ti_2O_7$ (R : Tb, Gd and Er), a complicated magnetic-field dependence of the thermal conductivity has been observed in correspondence to the change of the magnetic state [6, 7].

The compound $Ba_3CoSb_2O_9$ is a spin system where Co^{2+} spins with the spin quantum number $S = 1/2$ form an uniform triangular-lattice in the *ab*-plane and the layer of the triangular-lattice is repeatedly stacked along the *c*-axis. From ESR and magnetization measurements, the exchange interaction between the nearest neighboring spins in the *ab*-plane, J, and that between the triangular-lattice layers, J', have been estimated to be 18.5 K and 0.48 K, respectively [8]. An

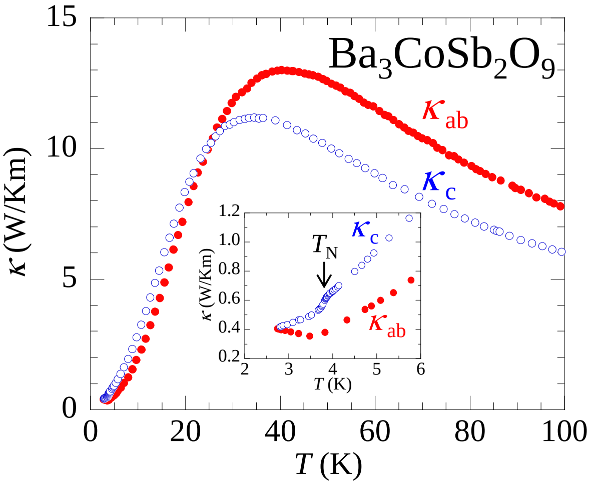

Figure 1. Temperature dependence of the thermal conductivity in the *ab*-plane, κ_{ab}, and along the *c*-axis, κ_c, of $Ba_3CoSb_2O_9$ single crystals in zero field. The inset shows the magnified plot around the antiferromagnetic transition temperature T_N = 3.8 K indicated by the arrow.

antiferromagnetic (AF) ordered state with the so-called 120° structure in the *ab*-plane appears at low temperatures below the AF transition temperature, T_N, ∼ 3.8 K due to the weak J' in zero field [9]. In high magnetic fields above 9 T parallel to the *ab*-plane, the so-called up-up-down (UUD) structure, where two thirds of spins are parallel and the others are antiparallel to the magnetic-field direction, appears at low temperatures below ∼5 K [8]. In the UUD state, the magnetization shows a 1/3 magnetization plateau in a field-range of 9–15 T at 1.3 K.

In this paper, we have measured the thermal conductivity in the *ab*-plane, κ_{ab}, and along the *c*-axis, κ_c, of $Ba_3CoSb_2O_9$ single crystals in zero field and magnetic fields parallel to the *ab*-plane, in order to investigate the magnetic state and the change of the magnetic state by the application of magnetic field.

2. Experimental

Single crystals of $Ba_3CoSb_2O_9$ were grown by the floating-zone method. The quality of the single crystals was checked by the x-ray back-Laue photography to be good. Thermal conductivity measurements were carried out by the conventional steady-state method. Magnetic fields up to 14 T were applied.

3. Results and discussion

Figure 1 shows the temperature dependence of κ_{ab} and κ_c of $Ba_3CoSb_2O_9$ single crystals in zero field. It is found that both κ_{ab} and κ_c show a broad peak around 40 K and then decrease with decreasing temperature. Since the magnitude of the peak and the temperature dependence of κ_{ab} are almost the same as those of κ_c, respectively, it is suggested that the contribution of phonons is dominant in the thermal conductivity. It is well known that the typical temperature dependence of κ_{phonon} in nonmagnetic insulators shows a peak around 10 K. Therefore, the appearance of the broad peak around 40 K in $Ba_3CoSb_2O_9$ may be due to strong magnetic-fluctuations, which arise from the frustration of Co^{2+} spins forming triangular-lattices and decrease the mean free path of phonons, l_{phonon}.

As shown in the inset of Fig. 1, κ_{ab} shows a kink and κ_c drops at T_N. At low temperatures below T_N, κ_{ab} increases and κ_c slowly decreases with decreasing temperature. Since κ_{phonon} is expressed as the product of the specific heat of phonons, C_{phonon}, the velocity of phonons, v_{phonon}, and l_{phonon} and v_{phonon} is little dependent on temperature, the temperature dependence of κ_{phonon} is determined by the temperature dependence of C_{phonon} and l_{phonon}. Therefore, the changes of κ_{ab} and κ_c below T_N are understood to be due to the increase of l_{phonon} owing to

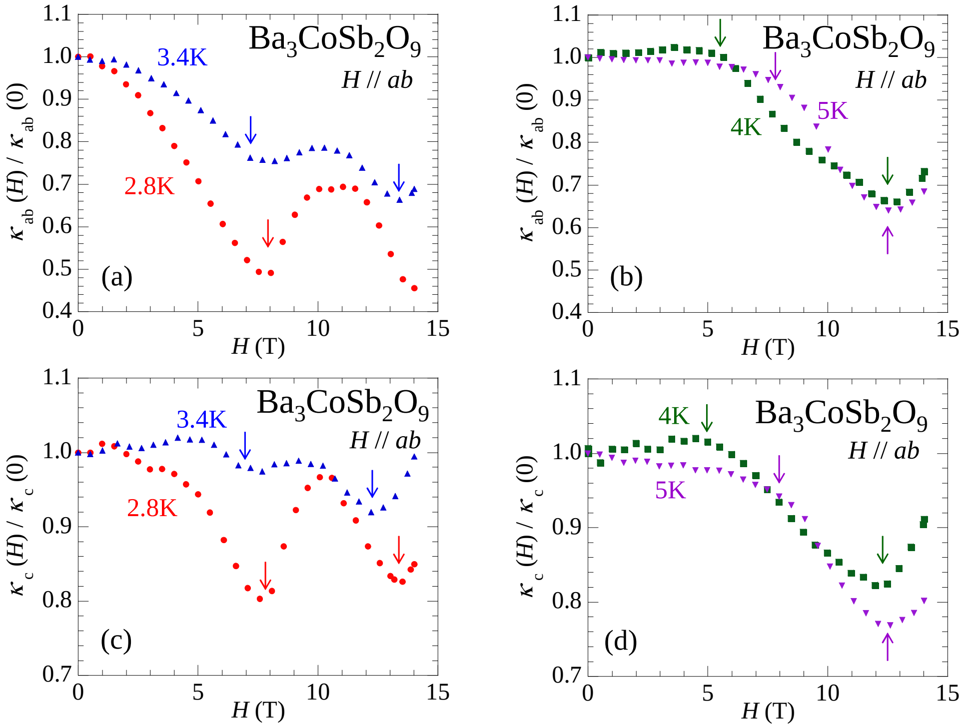

Figure 2. Magnetic-field dependence of the thermal conductivity normalized by the values in zero field for $Ba_3CoSb_2O_9$ single crystals. (a) and (b) show $\kappa_{ab}(H)/\kappa_{ab}(0)$ at low and high temperatures, respectively. (c) and (d) show $\kappa_c(H)/\kappa_c(0)$ at low and high temperatures, respectively. Magnetic fields are applied parallel to the *ab*-plane. Arrows indicate fields where the thermal conductivity drastically changes.

the suppression of the phonon-magnon scattering caused by the suppression of the magnetic fluctuations in the AF ordered state.

Figure 2 shows the magnetic-field dependence of $\kappa_{ab}(H)$ and $\kappa_c(H)$, normalized by the values in zero field, $\kappa_{ab}(0)$ and $\kappa_c(0)$, respectively, of $Ba_3CoSb_2O_9$ single crystals in magnetic fields parallel to the *ab*-plane at various temperatures. At low temperatures below T_N, it is found that both κ_{ab} and κ_c decrease with increasing field in low fields, where the 120° structure is formed, as shown in Figs. 2 (a) and (c). The decrease is interpreted as being due to the enhancement of magnetic fluctuations scattering phonons with increasing field, because magnetic fields applied parallel to the *ab*-plane disturb the ordered state with the 120° structure. With further increasing field, it is found that both κ_{ab} and κ_c increase above ∼8 T indicated by arrows in Figs. 2 (a) and (c). The fields ∼8 T indicated by the arrows are in good agreement with those where the transition from the 120° structure phase to the UUD phase occurs, as shown in the H–T magnetic phase diagram of Fig. 3 obtained from the magnetic susceptibility and specific heat measurements [10, 11]. The increase above ∼8 T is interpreted as being due to the decrease of the number of magnetic excitations, because the UUD phase has a spin gap [12, 13]. In high magnetic fields above 8 T, it is found that both κ_{ab} and κ_c show a peak around 11 T and a dip around 13 T. The dip fields ∼13 T indicated by arrows in Figs. 2 (a) and (c) are plotted by stars in Fig. 3. It is found that the dip fields are located in the middle of the UUD phase. However, the magnitude of the spin gap in the UUD phase is expected to disappear at the boundary of the UUD phase and show the maximum in the middle of the UUD phase. Therefore, the peaks around 11 T and the dips around 13 T cannot be interpreted in terms of the change of

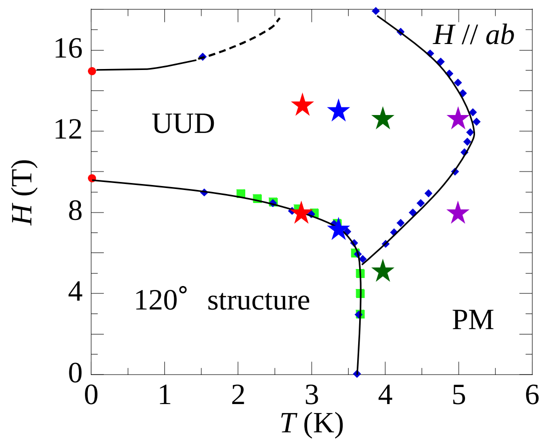

Figure 3. H–T magnetic phase diagram of $Ba_3CoSb_2O_9$ in magnetic fields parallel to the ab-plane. Data are from refs. [10, 11]. Stars correspond to arrows in Fig. 2.

the magnitude of the spin gap with changing field. These results suggest that some magnetic change takes place in the middle of the UUD phase showing the 1/3 magnetization plateau.

At high temperatures above T_N, on the other hand, it is found that both κ_{ab} and κ_c are almost constant in low magnetic fields below ∼5 T, as shown in Fig. 2 (b) and (d). This means that the number of magnetic excitations does not change with increasing field in the paramagnetic (PM) state. It is found that both κ_{ab} and κ_c start to decease at ∼8 T with increasing field. The fields ∼8 T indicated by arrows in Figs. 2 (b) and (d) are also plotted by stars in Fig. 3. It is found that the fields are a little smaller than the boundary between the PM and UUD phases. The decrease above ∼8 T implies that the increase of the number of magnetic excitations, but this cannot be explained by the increase of the spin gap in the UUD phase. The decrease above ∼8 T may be due to the enhancement of magnetic fluctuations, which has been observed around the phase boundary in the specific heat measurements [10]. That is, the strong magnetic-fluctuations around the phase boundary may scatter phonons markedly, leading to the decrease in thermal conductivity. This may be the reason why the fields ∼8 T indicated by arrows are a little smaller than the phase boundary. In high magnetic fields, it is found that both κ_{ab} and κ_c show a dip around 13 T. The dip fields ∼13 T indicated by arrows in Figs. 2 (b) and (d) are also plotted by stars in Fig. 3. It is found that the dip fields are located in the middle of the UUD phase as well as those at low temperatures. As mentioned above, therefore, it is very likely that the magnetic state changes in the middle of the UUD phase. However, no anomaly has been observed in the field dependence of the specific heat around 13 T [10]. Accordingly, it is necessary to investigate the magnetic state using another probe such as NMR and neutron scattering measurements.

4. Summary

We have measured κ_{ab} and κ_c of $Ba_3CoSb_2O_9$ single crystals in zero field and magnetic fields. In zero field, it has been found that both κ_{ab} and κ_c show a similar broad peak around 40 K, suggesting that κ_{phonon} is dominant and that l_{phonon} is suppressed so much by strong magnetic-fluctuations due to the frustration of Co^{2+} spins. It has also been found that both κ_{ab} and κ_c show a kink at T_N, owing to the increase of l_{phonon} in the AF ordered state. By the application of magnetic field parallel to the ab-plane in the 120° structure phase at low temperatures, it has been found that both κ_{ab} and κ_c decrease with increasing field due to the enhancement of magnetic fluctuations arising from the instability of the 120° structure. With further increasing field, it has been found that both κ_{ab} and κ_c increase above ∼8 T, where the transition from the 120° structure phase to the UUD phase occurs, owing to the decrease of the number of magnetic excitations in the UUD phase with a spin gap. In high magnetic fields above 8 T in the UUD

phase, it has been found that both $\kappa_{\rm ab}$ and $\kappa_{\rm c}$ show a peak around 11 T and a dip around 13 T, which cannot be interpreted in terms of the change of the magnitude of the spin gap. Even at high temperatures above $T_{\rm N}$, it has also been found that both $\kappa_{\rm ab}$ and $\kappa_{\rm c}$ show a dip around 13 T in the UUD phase. These results suggest that some magnetic change takes place in the middle of the UUD phase showing the 1/3 magnetization plateau.

Acknowledgments

The thermal conductivity measurements were performed at High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University. This work was supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

References

[1] Kudo K, Noji T, Koike Y, Nishizaki T and Kobayashi N 2001 *J. Phys. Soc. Jpn.* **70** 1448
[2] Hofmann M, Lorenz T, Uhrig G S, Kierspel H, Zabara O, Freimuth A, Kageyama H and Ueda Y 2001 *Phys. Rev. Lett.* **87** 047202
[3] Kudo K, Yamazaki M, Kawamata T, Noji T, Koike Y, Nishizaki T, Kobayashi N and Tanaka H 2004 *J. Phys. Soc. Jpn.* **73** 2358
[4] Kudo K, Yamazaki M, Kawamata T, Noji T, Koike Y, Nishizaki T, Kobayashi N and Tanaka H 2004 *J. Magn. Magn. Mater.* **272–276** 214
[5] Ke W P, Wang X M, Fan C, Zhao Z Y, Liu X G, Chen L M, Li Q J, Zhao X and Sun X F 2011 *Phys. Rev.* B **84** 094440
[6] Li Q J, Zhao Z Y, Fan C, Zhang F B, Zhou H D, Zhao X and Sun X F 2013 *Phys. Rev.* B **87** 214408
[7] Zhang F B, Li Q J, Zhao Z Y, Fan C, Li S J, Liu X G, Zhao X and Sun X F 2014 *Phys. Rev.* B **89** 094403
[8] Susuki T, Kurita N, Tanaka T, Nojiri H, Matsuo A, Kindo K and Tanaka H 2013 *Phys. Rev. Lett.* **110** 267201
[9] Shirata Y, Tanaka H, Matsuo A and Kindo K 2012 *Phys. Rev. Lett.* **108** 057205
[10] Zhou H D, Xu C, Hallas A M, Silverstein H J, Wiebe C R, Umegaki I, Yan J Q, Murphy T P, Park J-H, Qiu Y, Copley J R D, Gardner J S and Takano Y 2012 *Phys. Rev. Lett.* **109** 267206
[11] Koutroulakis G, Zhou T, Batista C D, Kamiya Y, Thompson J D, Brown S E and Zhou H D 2013 *Preprint* cond-mat/1308.6331
[12] Tsujii H, Rotundu C R, Ono T, Tanaka H, Andraka B, Ingersent K and Takano Y 2007 *Phys. Rev.* B **76** 060406R
[13] Fujii Y, Hashimoto H, Yasuda Y, Kikuchi H, Chiba M, Matsubara S and Takigawa M 2007 *J. Phys.: Condens. Matter* **19** 145237