Zn-Substitution Effect on the Thermal Conductivity of the Two-Dimensioinal Spin-Gap System $SrCu_2(BO_3)_2$ and the Two-Dimensional Antiferromagnetic System $Cu_3B_2O_6$ Single-Crystals

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Abstract

We have measured the thermal conductivity κ in the two-dimensional plane of $SrCu_{2-x}Zn_x(BO_3)_2$ (x = 0, 0.02) and $Cu_{3-x}Zn_xB_2O_6$ (x = 0, 0.03) single-crystals grown by the floating-zone method. It has been found that the temperature dependence of κ exhibits an anomalous peak in the spin-gap state of $SrCu_2(BO_3)_2$ and also in the antiferromagnetic ordered state of $Cu_3B_2O_6$. Through the partial substitution of Zn for Cu, the peak does not change in the former, while it is strongly reduced in the later. It is concluded that the peaks observed in $SrCu_2(BO_3)_2$ and $Cu_3B_2O_6$ are caused by the enhancement of κ due to phonons and magnons, respectively.

Key words:
$SrCu_2(BO_3)_2$, $Cu_3B_2O_6$, thermal conductivity, spin gap, antiferromagnetic order, Zn-substitution effect, single crystal

Recently, the thermal conductivity κ in low-dimensional quantum spin systems has attracted great interest, because large thermal conductivity due to magnons, κ_{magnon}, was observed in such several cuprates as regarded as one-dimensional spin systems, namely, $Sr_{14}Cu_{24}O_{41}$[1–3], $CuGeO_3$[4,5], Sr_2CuO_3 and $SrCuO_2$[6]. As for two-dimensional spin systems, an anomalous peak was observed in the temperature dependence of κ in the spin-gap state of $SrCu_2(BO_3)_2$[7,8] with the two-dimensional $CuBO_3$ plane. The peak was understood to be caused by large contribution of the thermal conductivity due to phonons, κ_{phonon}. A similar anomalous peak was also observed in the antiferromagnetic ordered state of $Cu_3B_2O_6$ with the two-dimensional $Cu_3B_2O_6$ plane[9]. In this case, on the other hand, the peak was understood to be caused by large contribution of κ_{magnon}[10]. In general, however, it is difficult to separate the contributions of κ_{magnon} and κ_{phonon} from each other within the theoretical fitting analysis, because it usually contains some ambiguity. Therefore, the discusstion on the origin of the anomalous peaks of κ observed in $SrCu_2(BO_3)_2$ and $Cu_3B_2O_6$ does not seem to be conclusive. In order to distinguish the contributions of κ_{magnon} and κ_{phonon} from each other, it is effective to study effects of the partial substitution of Zn for Cu on κ, because Zn^{2+}(the spin quantum number $S = 0$) is expected to shorten the mean free path of magnons running in the Cu^{2+}($S = 1/2$) network. Actually, it has been pointed out in $Sr_{14}Cu_{24}O_{41}$ that magnons have a larger tendency to be scatterd by Zn than phonons[3]. In this paper, therefore, we investigate Zn-substitution effects on κ in the partially Zn-substituted $SrCu_2(BO_3)_2$ and $Cu_3B_2O_6$.

Single crystals of $SrCu_{2-x}Zn_x(BO_3)_2$ (x = 0, 0.02) and $Cu_{3-x}Zn_xB_2O_6$ (x = 0, 0.03) were grown by the floating-zone method[7,9]. Thermal conductivity measurements were carried out by the conventional steady

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state method at the High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University.

Figure 1 shows the temperature dependence of κ along the a-axis (i.e. pallalel to the $CuBO_3$ plane), κ_a, of $SrCu_{2-x}Zn_x(BO_3)_2$. The κ_a exhibits an anomalous enhancement at low temperatures below 11 K and shows a peak at 3 K in both x = 0 and 0.02 . The peak does not change through the Zn substitution, suggesting that the peak observed in the spin-gap state of $SrCu_2(BO_3)_2$ is due to the enhancement of κ_{phonon}. That is, κ_{phonon} is interpreted to be enhanced in the spin-gap state, because the mean free path of phonons l_{phonon} increases on account of the reduction of the phonon-magnon scattering. This result is consistent with the previous reports[7,8]. On the other hand, the deviation at high temperatures above 20 K may be caused by the change of κ_{magnon} rather than κ_{phonon}, because κ_{phonon} is not expected to be reduced through the Zn substitution at high temperatures above 20 K. This is becauce κ_{phonon} is not reduced through the Zn substitution even at low temperatures below 20 K where l_{phonon} is much longer than that at high temperatures above 20 K.

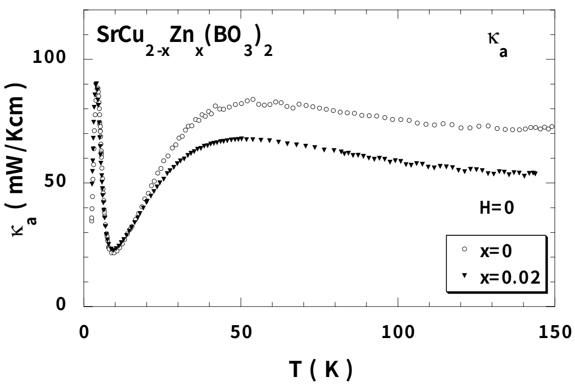

Fig. 1. Temperature dependence of the thermal conductivity along the a-axis in the two-dimensional $CuBO_3$ plane of $SrCu_{2-x}Zn_x(BO_3)_2$. Open circles and closed triangles denote the data of x = 0 and 0.02, respectively.

Figure 2 shows the temperature dependence of κ along the direction $bc\#1$, $\kappa_{bc\#1}$, of $Cu_{3-x}Zn_xB_2O_6$. Here, $bc\#1$ is the direction where the magnetization exhibits the minimum value in the two-dimensional bc plane[10]. The $\kappa_{bc\#1}$ exhibits an anomalous increase at low temperatures below the Néel temperature T_N = 10 K and shows a peak at 5 K in x = 0. The peak is strongly reduced through the Zn substitution. This result suggests that the peak observed in the antiferromagnetic ordered state of $Cu_3B_2O_6$ is due to κ_{magnon}. The reduction of the peak through the Zn substitution corresponds to the decrease of the mean free path of magnons. That is, it is concluded that κ_{magnon} is

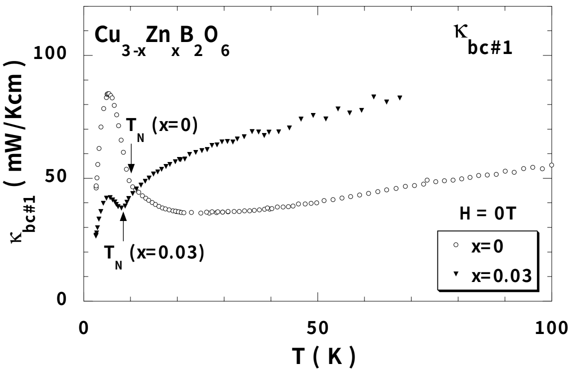

Fig. 2. Temperature dependence of the thermal conductivity along the direction $bc\#1$ in the two-dimensional $Cu_3B_2O_6$ plane of $Cu_{3-x}Zn_xB_2O_6$. Open circles and closed triangles denote the data of x = 0 and 0.03, respectively.

enhanced in the antiferromagnetic ordered state of $Cu_3B_2O_6$ on account of the increase of the mean free path of magnons. At high temperatures above T_N, on the other hand, $\kappa_{bc\#1}$ in x = 0.03 is larger than that in x = 0, which may be attributed to the increase of the sound velocity through the Zn substitution.

In conclusion, the anomalous peaks of κ observed in $SrCu_2(BO_3)_2$ and $Cu_3B_2O_6$ are regarded as being due to the enhancement of κ_{phonon} and κ_{magnon} respectively, because the peak does not change in the former while it is strongly reduced in the later through the partial substitution of Zn for Cu.

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