Observation of the Thermal Conductivity due to Spins in the One-Dimensional Antiferromagnetic Ising-Like Spin System \( A\text{Co}X_3 \) (\( A = \text{Rb, Cs}; \ X = \text{Cl, Br} \))

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Thermal conductivity measurements have been carried out for single crystals of the one-dimensional (1D) antiferromagnetic (AF) Ising-like spin system \( A\text{Co}X_3 \) (\( A = \text{Rb, Cs}; \ X = \text{Cl, Br} \)). A shoulder originating from the thermal conductivity due to spin excitations, \( \kappa_{\text{spin}} \), has been observed around 60 K in the temperature dependence of the thermal conductivity along the \( c \)-axis parallel to spin chains for every \( A\text{Co}X_3 \). To our knowledge, this is the first observation of \( \kappa_{\text{spin}} \) in 1D AF Ising-like spin systems. It has been found that the magnitude of \( \kappa_{\text{spin}} \) in \( A\text{Co}X_3 \) is a little smaller than those in 1D AF Heisenberg spin systems. The reason is discussed in terms of the Heisenberg-like character in \( A\text{Co}X_3 \).

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1. Introduction

The thermal conductivity in low-dimensional quantum spin systems has attracted great interest, because various remarkable anomalies associated with the low dimensionality of the spin correlation have been observed in these spin systems. In particular, the thermal conductivity due to spin excitations, $\kappa_{\text{spin}}$, has been observed markedly in one-dimensional (1D) antiferromagnetic (AF) Heisenberg spin systems,\textsuperscript{1–17} two-dimensional AF Heisenberg spin systems\textsuperscript{18–26} and a 1D ferromagnetic Heisenberg spin system also.\textsuperscript{27,28} It has been found that the magnitude of $\kappa_{\text{spin}}$ tends to increase with increasing exchange interaction between the nearest neighboring spins in 1D AF Heisenberg spin systems.

The Hamiltonian of a 1D AF system is described using the XXZ Heisenberg model as follows:

$$ H = J \sum_i \left( s_i^z s_{i+1}^z + \varepsilon \left( s_i^x s_{i+1}^x + s_i^y s_{i+1}^y \right) \right), $$

where $s = (s^x, s^y, s^z)$ is the spin, $J$ the intrachain exchange interaction between the nearest neighboring spins and $\varepsilon$ the anisotropy parameter of the spin. When spins are isotropic, namely, $\varepsilon = 1$, the spin system is called a Heisenberg one. When $\varepsilon = 0$, on the other hand, the spin system is called an Ising one. Using the spin raising and lowering operators, $s^+ = s^x + is^y$ and $s^- = s^x - is^y$, respectively, Eq. (1) is rewritten as follows:

$$ H = J \sum_i \left( s_i^z s_{i+1}^z + \frac{\varepsilon}{2} \left( s_i^x s_{i+1}^x + s_i^y s_{i+1}^y \right) \right). $$

The second and third terms are so related to the propagation of spin excitations as to be likely important for the appearance of $\kappa_{\text{spin}}$. Therefore, $\kappa_{\text{spin}}$ is supposed to be very small in Ising spin systems. Theoretically, in fact, this is supported.\textsuperscript{29} Experimentally, no $\kappa_{\text{spin}}$ has been observed in the quasi-1D AF spin system BaCo$_2$V$_2$O$_8$ where $\varepsilon \sim 0.5$.\textsuperscript{30,31}

The compound ACoX$_3$ ($A =$ Rb, Cs; $X =$ Cl, Br) is regarded as a quasi-1D AF Ising-like spin system where the space group of the crystal structure is $P6_3/mmc$ at room temperature. The AF spin chains consist of face-shared CoX$_6$ octahedra stacked along the $c$-axis, and the spin chains compose a triangular lattice in the $c$-plane. The Ising-like anisotropy of Co$^{2+}$ spins along the $c$-axis is due to the spin-orbit interaction. The Hamiltonian of ACoX$_3$ is given by Eq. (1), where $\varepsilon$ is estimated from Raman scattering,\textsuperscript{32–34} neutron scattering,\textsuperscript{35–37} magnetization\textsuperscript{38} and ESR measurements\textsuperscript{39} to be below 0.24 and smaller than that of BaCo$_2$V$_2$O$_8$, as listed in Table I. It is known that ACoX$_3$ exhibits successive phase transitions at low temperatures $T_{N1}$ and $T_{N2}$, owing to the interchain interaction.\textsuperscript{33,40–42} The magnetic state is param-
agnetic at high temperatures above $T_{N1}$. At intermediate temperatures between $T_{N1}$ and $T_{N2}$, a partially ordered phase is formed, where two thirds of spin chains are ordered like a honeycomb lattice in the $c$-plane owing to the AF interchain interaction. At low temperatures below $T_{N2}$, all Co$^{2+}$ spins are ordered, so that a ferrimagnetic order is formed. Moreover, it is known that RbCoBr$_3$ undergoes two structural transitions at $T_{s1}$ and $T_{s2}$. Values of $T_{N1}$, $T_{N2}$, $T_{s1}$ and $T_{s2}$ are listed in Table I together with those of $J$ and the ratio of the interchain interaction, $J'$, to $J$.

In this paper, in order to investigate the contribution of spin excitations to the thermal conductivity in 1D AF Ising-like spin systems, we have measured the thermal conductivity of ACoX$_3$ ($A = \text{Rb, Cs}; \ X = \text{Cl, Br}$).

2. Experimental

Single crystals of ACoX$_3$ ($A = \text{Rb, Cs}; \ X = \text{Cl, Br}$) were grown by the Bridgman technique using equimolar mixture of AX and CoX$_2$ sealed in an evacuated quartz tube. Both of the thermal conductivity along the $c$-axis parallel to spin chains, $\kappa_{\parallel c}$, and that in the $c$-plane perpendicular to spin chains, $\kappa_{\perp c}$, were measured by the conventional steady-state method. The typical dimensions of a single crystal used for the measurements were 4 mm along the direction of heat current and $2 \times 2$ mm$^2$ perpendicular to the direction. One side of a single crystal was anchored on the copper heat sink with indium solder. A chip-resistance of 1 kΩ (Alpha Electronics MP1K000) was attached as a heater to the opposite side of the single crystal with Araldite epoxy adhesive. The temperature gradient across the crystal was measured with two Cernox thermometers (Lake Shore Cryotronics, CX-1050-SD). The error of the absolute value of the thermal conductivity obtained was estimated to be about 10% on account of errors of the crystal geometry. Thermal conductivity measurements in magnetic fields up to 14 T were also carried out for RbCoCl$_3$ using a superconducting magnet.

3. Results and Discussion

Figure 1 shows the temperature dependence of $\kappa_{\parallel c}$ and $\kappa_{\perp c}$ of single crystals of ACoX$_3$ in zero field. It is found that a peak appears at a low temperature below 10 K in both $\kappa_{\parallel c}$ of every ACoX$_3$ and $\kappa_{\perp c}$ of RbCoCl$_3$, while a shoulder is observed around 60 K in only $\kappa_{\parallel c}$ of every ACoX$_3$. Since ACoX$_3$ is an insulator, the thermal conductivity is given by the sum of the contribution of phonons and spin excitations. The thermal conductivity due to phonons, $\kappa_{\text{phonon}}$, is usually more isotropic than $\kappa_{\text{spin}}$, because the anisotropy of the correlation between atoms is generally much smaller than that of the spin correlation in a low-dimensional spin
system. Moreover, the temperature dependence of $\kappa_{\perp c}$ is typical of $\kappa_{\text{phonon}}$. Therefore, it is concluded that the peak at a low temperature below 10 K is due to $\kappa_{\text{phonon}}$ and that the shoulder around 60 K is due to $\kappa_{\text{spin}}$. Similar temperature dependence of the thermal conductivity along spin chains has been observed in several 1D AF Heisenberg spin systems such as Sr$_2$CuO$_3$ and SrCuO$_2$.

In Fig. 1, it is noticed that neither sharp dip is observed at $T_{\text{N1}}$ nor at $T_{\text{N2}}$ for every ACoX$_3$, while a sharp dip has been observed at the magnetic transition temperature in every direction of thermal conductivity for BaCo$_2$V$_2$O$_8$ and several three-dimensional magnets. The reason for neither sharp dip at $T_{\text{N1}}$ nor at $T_{\text{N2}}$ for ACoX$_3$ is not clear at present, though several low-dimensional AF spin systems such as Sr$_2$CuO$_3$ and La$_2$CuO$_4$ have shown no sharp dip at their magnetic transition temperatures.

In Fig. 1, it is also noticed that neither clear change is observed at $T_{s1}$ nor at $T_{s2}$ for RbCoBr$_3$. Since the structural phase transitions are caused by sliding motions of spin chains along the $c$-axis, it may be reasonable that $\kappa_{\parallel c}$ is not affected by the transitions.

Here, we estimate the magnitude of $\kappa_{\text{spin}}$. Since $\kappa_{\text{spin}}$ is given by the subtraction of $\kappa_{\text{phonon}}$ from $\kappa_{\parallel c}$ as follows:

$$\kappa_{\text{spin}}(T) = \kappa_{\parallel c}(T) - \kappa_{\text{phonon}}(T), \quad (3)$$

the estimation of $\kappa_{\text{phonon}}$ is necessary at first. Based on the Debye model with the relaxation time approximation, $\kappa_{\text{phonon}}$ is given by

$$\kappa_{\text{phonon}}(T) = \frac{k_B}{2\pi^2 v_{\text{phonon}}} \left(\frac{k_BT}{\hbar}\right)^3 \int_0^{\Theta_D/T} dx \frac{x^4 e^x}{(e^x - 1)^2} \tau_{\text{phonon}}(x, T), \quad (4)$$

where $k_B$ is the Boltzmann constant, $\hbar$ the Planck constant, $\Theta_D$ the Debye temperature, $v_{\text{phonon}}$ the velocity of phonons, $\tau_{\text{phonon}}$ the relaxation time of the phonon with the angular frequency $\omega$, and $x = \hbar\omega/k_BT$. The $v_{\text{phonon}}$ is calculated as

$$v_{\text{phonon}} = \frac{k_B \Theta_D}{\hbar} \left(6\pi^2 n\right)^{-1/3}, \quad (5)$$

where $n$ is the number density of atoms. The phonon scattering rate, $\tau_{\text{phonon}}^{-1}$, is given by the sum of scattering rates in various scattering processes as follows:

$$\tau_{\text{phonon}}^{-1}(\omega, T) = \frac{v_{\text{phonon}}}{L_b} + P\omega^4 + D\omega + U\omega^2 T \exp(-\Theta_D/\mu T), \quad (6)$$

where $L_b$, $P$, $D$, $U$ and $\mu$ are fitting parameters. The first term represents the phonon scattering by boundaries. The second term represents the phonon scattering by point defects. The third term represents the phonon scattering by lattice distortions. The fourth term represents the phonon-phonon scattering in the umklapp process.
Using Eqs. (4)–(6) and putting $\Theta_D$ at 310 K estimated from the Mössbauer measurements, the data of $\kappa_{\perp c}(T)$ in RbCoCl$_3$ are well fitted in a wide temperature-range as shown by the black line in Fig. 1, indicating that $\kappa_{\perp c}(T)$ is due to only $\kappa_{\text{phonon}}(T)$. The obtained parameters are listed in Table II. The estimation of $\kappa_{\text{phonon}}(T)$ in $\kappa_{\parallel c}(T)$ of ACoX$_3$ is performed by the fit of the data of $\kappa_{\parallel c}(T)$ at low temperatures below 25 K with Eqs. (4)–(6), because $\kappa_{\text{spin}}(T)$ decreases markedly with decreasing temperature at low temperatures of $k_B T \ll J$ in usual low-dimensional spin systems. In the fitting, values of $\Theta_D$ of RbCoCl$_3$ and CsCoBr$_3$ are put at those estimated from the Mössbauer measurements, and those of RbCoBr$_3$ and CsCoCl$_3$ are put at the average value of those of RbCoCl$_3$ and CsCoBr$_3$, considering the weight of atoms. The value of $L_b$ is given by the distance between two terminals of temperature on a single crystal. The best-fit results of $\kappa_{\text{phonon}}(T)$ are shown by blue solid lines in Fig. 2. Shaded areas in Fig. 2 indicate errors of $\kappa_{\text{phonon}}(T)$. Values of parameters used for the best fit are listed in Table II. It is found that values of $P$ obtained in $\kappa_{\parallel c}$ and $\kappa_{\perp c}$ of RbCoCl$_3$ are very different from each other, which is unreasonable. The difference of the $P$ value is caused by the analysis using the Debye model where the anisotropy of $v_{\text{phonon}}$ is not taken into account. That is, the anisotropy of $v_{\text{phonon}}$ appears to bring about the difference of the $P$ value between $\kappa_{\parallel c}$ and $\kappa_{\perp c}$. Using Eq. (3), $\kappa_{\text{spin}}(T)$ is obtained as shown by red circles in Fig. 2. It is found that $\kappa_{\text{spin}}(T)$ exhibits the maximum at a high temperature above 50 K and that the maximum value of $\kappa_{\text{spin}}(T)$ is about 3 W/Km in RbCoCl$_3$, 4 W/Km in CsCoCl$_3$, CsCoBr$_3$, and 0.9 W/Km in RbCoBr$_3$. Here, it is noted that $\kappa_{\text{spin}}(T)$ is a little underestimated, because $\kappa_{\text{spin}}(T)$ is neglected at low temperatures below 25 K. Values of $\kappa_{\text{spin}}(T)$ in RbCoBr$_3$ are smaller than those of $\kappa_{\text{spin}}(T)$ in the other ACoX$_3$. Since structural phase transitions occur at $T_{s1}$ and $T_{s2}$ only in RbCoBr$_3$, the lattice mismatch may be marked in RbCoBr$_3$ on account of the difference of the ionic radius between Rb$^+$ and Br$^-$. Therefore, the lattice mismatch may induce disorder of the lattice and suppress $\kappa_{\text{phonon}}(T)$ and $\kappa_{\text{spin}}(T)$.

Figure 3 displays the maximum values of $\kappa_{\text{spin}}$ in ACoX$_3$ and several 1D AF Heisenberg spin systems plotted as a function of $J$. It is found that the maximum values of $\kappa_{\text{spin}}$ in 1D AF Heisenberg spin systems tend to be proportional to $J$. This is simply understood to be due to the velocity of spin excitations, $v_{\text{spin}}$, being proportional to $J$ in spite of the specific heat, $C_{\text{spin}}$, being inversely proportional to $J$ at low temperatures in 1D AF Heisenberg spin systems, because $\kappa_{\text{spin}}$ is proportional to $C_{\text{spin}} v_{\text{spin}}^2$. It is found that the maximum values of $\kappa_{\text{spin}}$ in ACoX$_3$ are located a little below the shaded zone indicating the rough proportionality between the maximum value of $\kappa_{\text{spin}}$ and $J$. The bandwidth of the magnetic dispersion in an 1D Heisenberg spin system is proportional to $J$, so that $v_{\text{spin}}$ is proportional to $J$ at low
temperatures. In an 1D AF quasi-Ising spin system, on the other hand, the bandwidth, namely, $\nu_{\text{spin}}$ is expected to be proportional to $\varepsilon J$, according to Eq. (2). Using values of $\varepsilon J$ instead of those of $J$ in Fig. 3, therefore, the maximum values of $\kappa_{\text{spin}}$ in $A\text{Co}X_3$ are found to shift just into the shaded zone. Accordingly, this result strongly suggests that a little Heisenberg-like character with $\varepsilon \neq 0$ brings about a significant bandwidth, leading to the appearance of $\kappa_{\text{spin}}$ even in an 1D AF Ising-like spin system. Here, it is noted that $\kappa_{\text{ijc}}$ of $\text{RbCoCl}_3$ little changes by the application of magnetic field of 14 T along the $c$-axis in a wide temperature-range of 6–140 K, as shown in Fig. 1. That is, $\kappa_{\text{spin}}$ appears not to be affected by the magnetic field of 14 T. The magnetic field of 14 T may be too small to affect $\kappa_{\text{spin}}$ of $\text{RbCoCl}_3$ being markedly observed at high temperatures around 70 K. Furthermore, it is not clear why no $\kappa_{\text{spin}}$ has been observed in the quasi-1D AF spin system $\text{BaCo}_2\text{V}_2\text{O}_8$ in spite of the sizable value of $\varepsilon \sim 0.5$. To be more conclusive, therefore, thermal conductivity measurements in the other 1D AF Ising-like spin systems are necessary.

4. Summary

Thermal conductivity measurements have been carried out for single crystals of the 1D AF Ising-like spin system $A\text{Co}X_3$ ($A = \text{Rb, Cs}; X = \text{Cl, Br}$). It has been found that $\kappa_{\perp c}$ perpendicular to spin chains shows an only peak at a low temperature below 10 K and that $\kappa_{\parallel c}$ parallel to spin chains shows a shoulder around 60 K besides a peak at a low temperature below 10 K. It has been concluded that the peak is due to $\kappa_{\text{phonon}}$ and that the shoulder around 60 K is due to $\kappa_{\text{spin}}$. To our knowledge, this is the first observation of $\kappa_{\text{spin}}$ in 1D AF Ising-like spin systems. It has been found that the magnitude of $\kappa_{\text{spin}}$ in $A\text{Co}X_3$ estimated by the subtraction of $\kappa_{\text{phonon}}$ from $\kappa_{\parallel c}$ is a little smaller than those in 1D AF Heisenberg spin systems and that the magnitude of $\kappa_{\text{spin}}$ tends to be proportional to $\varepsilon J$ universally in both $A\text{Co}X_3$ and 1D AF Heisenberg spin systems. Accordingly, it has been concluded that a little Heisenberg-like character with $\varepsilon \neq 0$ brings about a significant bandwidth of spin excitations, leading to the appearance of $\kappa_{\text{spin}}$ even in an 1D AF Ising-like spin system. To be more conclusive, thermal conductivity measurements in the other 1D AF Ising-like spin systems are necessary.

Acknowledgment

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Figure captions

Fig. 1. (Color online) Temperature dependence of the thermal conductivity along the \(c\)-axis, \(\kappa_{\parallel c}\), in \(A\text{CoX}_3\) \((A = \text{Rb, Cs}; X = \text{Cl, Br})\) and the thermal conductivity in the \(c\)-plane, \(\kappa_{\perp c}\), in \(\text{RbCoCl}_3\) in zero field. The black line is the best-fit result of \(\kappa_{\perp c}\) in \(\text{RbCoCl}_3\) using the thermal conductivity due to phonons given by Eqs. (4)–(6) with parameters listed in Table II. Red circles show \(\kappa_{\parallel c}\) of \(\text{RbCoCl}_3\) in a magnetic field of 14 T applied along the \(c\)-axis.

Fig. 2. (Color online) Temperature dependence of the thermal conductivity along the \(c\)-axis, \(\kappa_{\parallel c}\), in (a) \(\text{RbCoCl}_3\), (b) \(\text{RbCoBr}_3\), (c) \(\text{CsCoCl}_3\), (d) \(\text{CsCoBr}_3\). Blue solid lines show \(\kappa_{\text{phonon}}\) estimated using Eqs. (4)–(6). Shaded areas demonstrate errors of \(\kappa_{\text{phonon}}\). Red circles show \(\kappa_{\text{spin}}\) obtained by subtracting \(\kappa_{\text{phonon}}\) from \(\kappa_{\parallel c}\).

Fig. 3. (Color online) Dependence on the intrachain interaction, \(J\), of the maximum value of \(\kappa_{\text{spin}}\) in the 1D AF Ising-like spin system \(A\text{CoX}_3\) \((A = \text{Rb, Cs}; X = \text{Cl, Br})\) (closed circles) and in 1D AF Heisenberg spin systems (open circles).\(^{1,10,17,50,51}\) Diamonds show dependence on \(\varepsilon J\) of the maximum value of \(\kappa_{\text{spin}}\) in \(A\text{CoX}_3\). The shaded zone indicates the rough proportionality between the maximum value of \(\kappa_{\text{spin}}\) and \(J\).
Table I. Magnetic parameters (the intrachain exchange interaction, $J$, the anisotropy parameter of spin, $\varepsilon$, the ratio of the interchain exchange interaction, $J'$, to $J$, the magnetic phase transition temperatures, $T_{N1}$ and $T_{N2}$) and structural parameters (the structural phase transition temperatures, $T_{s1}$ and $T_{s2}$) of ACoX$_3$ ($A = \text{Rb, Cs}$; $X = \text{Cl, Br}$).

|       | $J$ (K) | $\varepsilon$ | $J'/J$ | $T_{N1}$ (K) | $T_{N2}$ (K) | $T_{s1}$ (K) | $T_{s2}$ (K) |
|-------|---------|---------------|---------|--------------|--------------|--------------|--------------|
| RbCoCl$_3^a$ | 140–152 | 0.091–0.15 | 0.02–0.11 | 28 | 11 | - | - |
| RbCoBr$_3^b$ | 194 | - | 0.025 | 37 | 31 | 90 | 37 |
| CsCoCl$_3^c$ | 126–150 | 0.097–0.16 | 0.013–0.12 | 21 | 9 | - | - |
| CsCoBr$_3^d$ | 124–182 | 0.11–0.24 | 0.019–0.13 | 28 | 10 | - | - |

$^a$ Ref.33, 34, 38. $^b$ Ref.42, 43. $^c$ Ref.32, 34–40. $^d$ Ref.32, 34, 36, 38, 41.

Table II. Parameters used for the fit of the temperature dependences of the thermal conductivity, $\kappa_{||c}$ and $\kappa_{\perp c}$, in ACoX$_3$ with Eqs. (4)–(6).

|       | $\kappa$ | $\Theta_D$ (K) | $L_\eta$ (mm) | $P$ ($10^{-43}$ J s$^{-1}$) | $D$ ($10^{-6}$) | $U$ ($10^{-18}$ sK$^{-1}$) | $\mu$ |
|-------|---------|----------------|---------------|----------------------------|-----------------|----------------------------|------|
| RbCoCl$_3$ | $\kappa_{||c}$ | 310 | 1.48 | 52.5 ± 1.9 | 36.1 ± 1.0 | 39.8 ± 0.4 | 7.66 ± 0.05 |
|       | $\kappa_{\perp c}$ | 310 | 2.03 | 0.82 ± 0.35 | 43.3 ± 1.3 | 19.0 ± 0.4 | 9.8 ± 0.2 |
| RbCoBr$_3$ | $\kappa_{||c}$ | 300 | 1.26 | 16.9 ± 3.8 | 86.0 ± 5.7 | 35.0 ± 1.0 | 10.0 ± 0.4 |
|       | $\kappa_{\perp c}$ | 300 | 1.65 | 3.6 ± 5.6 | 0 ± 2.5 | 15.5 ± 1.7 | 8.3 ± 0.8 |
| CsCoCl$_3$ | $\kappa_{||c}$ | 300 | 1.54 | 0.24 ± 1.40 | 0.08 ± 2.69 | 9.23 ± 0.69 | 8.82 ± 0.50 |
Fig. 1.
Fig. 2.
Fig. 3.

Intrachain exchange interaction $J$ (K)

Maximum value of $\kappa_{\text{spin}}$ (W/Km)

- Sr$_2$CuO$_3$
- BaCu$_2$Si$_2$O$_7$
- Sr$_2$V$_3$O$_9$
- KCuF$_3$
- LiCuVO$_4$
- CsCoBr$_3$
- CsCoCl$_3$
- RbCoCl$_3$
- RbCoBr$_3$