Thermal Conductivity of the $S = 1/2$
Quasi-One-Dimensional Ferromagnetic Spin System
CsCuCl$_3$

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Abstract. We have measured the thermal conductivity along the $c$-axis, $\kappa_{||c}$, parallel to ferromagnetic spin-chains of single crystals of the $S = 1/2$ quasi-one-dimensional spin system CsCuCl$_3$ in magnetic fields up to 14 T, in order to investigate the thermal conductivity due to spins, $\kappa_{\text{spin}}$, and the change of thermal conductivity corresponding to the change of the spin state. In the temperature dependence of $\kappa_{||c}$, no contribution of $\kappa_{\text{spin}}$ has been observed, while a dip has been observed at the antiferromagnetic phase transition temperature, $T_N$. Furthermore, it has been found that $\kappa_{||c}$ at a low temperature of 5.1 K below $T_N$ changes with increasing field perpendicular to the $c$-axis in good correspondence to the field-induced change of the spin state.

1. Introduction
Thermal conductivity in low-dimensional quantum spin systems has attracted interest, owing to the large contribution of the thermal conductivity due to spins, $\kappa_{\text{spin}}$ [1, 2]. Especially, a number of studies on $\kappa_{\text{spin}}$ have been carried out in the spin quantum number $S = 1/2$ one-dimensional (1D) antiferromagnetic (AFM) systems [3–11]. The large contribution of $\kappa_{\text{spin}}$ has been observed as a peak or a shoulder in the temperature dependence of the thermal conductivity along spin chains together with the contribution of the thermal conductivity due to phonons, $\kappa_{\text{phonon}}$. In the 1D ferromagnetic (FM) systems, on the other hand, the contribution of $\kappa_{\text{spin}}$ has not been observed clearly, though several studies on $\kappa_{\text{spin}}$ have been performed [12–16].

The compound CsCuCl$_3$ belongs to the so-called $ABX_3$ family including well-known quasi-1D spin systems with frustration. The compound contains linear spin-chains of face-sharing CuCl$_6$ octahedra running along the $c$-axis. The spin chains separated by Cs$^+$ ions form an equilateral triangular lattice in the $c$-plane. The intrachain exchange interaction $J$ is FM and estimated as $\sim 28$ K, while the interchain exchange interaction $J'$ is AFM and estimated as $\sim 5$ K [17]. Therefore, frustration is expected to exist more or less between the spin chains. The CuCl$_6$ octahedra are distorted due to the Jahn-Teller effect, so that the Dzyaloshinsky-Moriya interaction is induced between spins along the $c$-axis. An AF long-range order appears at the phase transition temperature, $T_N$, $\sim 10.7$ K, owing to $J'$ [18]. The spin structure at low temperatures below $T_N$ is helical with the pitch of $\sim 5.1^\circ$ along the $c$-axis owing to the combination of $J$ and the Dzyaloshinsky-Moriya interaction. In the $c$-plane, the so-called $120^\circ$
Figure 1. Temperature dependence of the thermal conductivity along the c-axis, $\kappa_{\parallel c}$, parallel to spin chains in zero field and magnetic fields perpendicular to the c-axis.

spin structure is formed owing to $J'$ and the spin directions are confined in the c-plane [18]. Moreover, it has been known that the pitch decreases by the application of magnetic field perpendicular to spin chains [19–21].

In this paper, we have measured the thermal conductivity along the c-axis, $\kappa_{\parallel c}$, parallel to spin chains of CsCuCl$_3$ in order to investigate the contribution of $\kappa_{\text{spin}}$. Furthermore, we have also investigated the relation between the spin state and the behavior of the thermal conductivity, because thermal conductivity is a good probe detecting a change of the spin state in a spin system via the change of the scattering rate of heat carries [22–26].

2. Experimental
Single crystals of CsCuCl$_3$ were grown from solution. Thermal conductivity measurements were carried out by the conventional steady-state method in magnetic fields up to 14 T.

3. Results and Discussion
Figure 1 shows the temperature dependence of $\kappa_{\parallel c}$ parallel to spin chains in zero field and magnetic fields perpendicular to the c-axis at low temperatures below ~ 100 K. It has been found that $\kappa_{\parallel c}$ show a peak around 10 K with a dip at $T_N$ in zero field. This kind of dip at $T_N$ has been observed in several antiferromagnets [5, 27, 28], and is interpreted as being due to the strong scattering of heat carries caused by the critical fluctuations around $T_N$ or due to the increase of the mean free path of heat carries just below $T_N$ on account of the marked reduction of the scattering caused by the development of the long-range order. Therefore, this behavior of $\kappa_{\parallel c}$ in zero field is likely to be mainly due to the contribution of $\kappa_{\text{phonon}}$ affected by the scattering of phonons by magnetic excitations. The contribution of $\kappa_{\text{spin}}$ cannot be observed clearly in CsCuCl$_3$. Recently, single crystals of CsCuCl$_3$ of a single domain with homochirality of spins have been grown by Kousaka et al. [29]. On the other hand, our single crystals of the zigzag shape may be of multi-domains. Therefore, $\kappa_{\text{spin}}$ might strongly be suppressed due to the strong scattering of magnons at the domain boundaries.

By the application of magnetic field perpendicular to the c-axis, it has been found that $\kappa_{\parallel c}$ at low temperatures below $T_N$ is suppressed, as shown Fig. 1. To see the suppression in detail, the
Figure 2. Magnetic-field dependence of the thermal conductivity along the $c$-axis normalized by the value in zero field, $\kappa_{||c}(H)/\kappa_{||c}(0)$, of CsCuCl$_3$ in magnetic fields perpendicular to the $c$-axis. Three arrows on the left and right sides mean $120^\circ$ and coplanar 2-1-type spin structures, respectively.

The magnetic-field dependence of $\kappa_{||c}$ normalized by the value in zero field, $\kappa_{||c}(H)/\kappa_{||c}(0)$, at 5.1 K is shown in Fig. 2. It has been found that $\kappa_{||c}(H)/\kappa_{||c}(0)$ monotonically decreases with increasing field up to $\sim 9$ T, becomes constant between $\sim 9$ T and $\sim 12$ T, and then decreases again above $\sim 12$ T. This behavior is very similar to the magnetic-field dependence of the incommensurability, $\delta$, which is proportional to the pitch of the helical spin structure along the $c$-axis [19–21]. It is known that the helical and $120^\circ$ spin structures are maintained in low magnetic fields below $\sim 9$ T, though the pitch of the helical structure along the $c$-axis decreases with increasing field [21]. The $120^\circ$ spin structure in the $c$-axis is expected to become unstable by the application of magnetic field in the $c$-plane. Therefore, it is likely that $\kappa_{||c}$ is suppressed with increasing field due to the enhancement of the magnetic fluctuations scattering phonons.

In a region between $9$ T and $16$ T, it is known that the helical and the so-called coplanar 2-1-type spin structures, where two thirds of spins are roughly parallel and the others are roughly antiparallel to the magnetic-field direction as shown in Fig. 2, are formed. The constant value of $\kappa_{||c}(H)/\kappa_{||c}(0)$ in this region implies that the number of magnetic excitations scattering phonons does not change so much. This is consistent with the locking phenomenon of the spin structure in this region observed in the neutron scattering experiment [21] and with the plateau-like behavior in this region observed in the magnetization curve also [30].

In high magnetic fields above $\sim 12$ T, on the other hand, the suppression of $\kappa_{||c}$ can be explained by the enhancement of magnetic fluctuations scattering phonons, because the field-induced transition to the non-helical commensurate phase with $\delta = 0$ takes place at $\sim 16$ T [20, 21]. The behavior of $\kappa_{||c}$ is also roughly consistent with the neutron scattering result that the value of $\delta$ starts to decrease at $\sim 14$ T with increasing field [20, 21].

4. Conclusions
We have measured $\kappa_{||c}$ parallel to spin chains of CsCuCl$_3$, in order to investigate the presence or absence of $\kappa_{\text{spin}}$ and the change of the spin state. Neither peak nor shoulder due to the contribution of $\kappa_{\text{spin}}$ has been observed in the temperature dependence of $\kappa_{||c}$ in zero field. This may be due to the strong scattering of magnons at boundaries of domains with homochirality of spins in the helical spin structure along the $c$-axis. By the application of magnetic field perpendicular to the $c$-axis up to $14$ T, it has been found that $\kappa_{||c}$ at 5.1 K below $T_N$ decreases...
with increasing field up to $\sim 9$ T, is constant between $\sim 9$ T and $\sim 12$ T, and decreases again above $\sim 12$ T. These behaviors have been explained as being due to the change of $\hat{\kappa}_{\text{phonon}}$ caused by the change of the scattering of phonons by magnetic excitations due to the field-induced change of the spin state.

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