Thermal Conductivity of the Quasi One-Dimensional Spin System $\text{Sr}_2\text{V}_3\text{O}_9$

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Abstract. We have measured the thermal conductivity along the [101] direction, $\kappa_{[101]}$, along the [10¯1] direction, $\kappa_{[10\bar{1}]}$, and along the $b$-axis, $\kappa_b$, of the quasi one-dimensional $S=1/2$ spin system $\text{Sr}_2\text{V}_3\text{O}_9$ in magnetic fields up to 14 T, in order to find the thermal conductivity due to spinons and to clarify whether the spin-chains run along the [101] or [10¯1] direction. It has been found that both $\kappa_{[101]}$, $\kappa_{[10\bar{1}]}$ and $\kappa_b$ show one peak around 10 K in zero field and that the magnitude of $\kappa_{[10\bar{1}]}$ is larger than those of $\kappa_{[101]}$ and $\kappa_b$. By the application of magnetic field along the heat current, the peak of $\kappa_{[10\bar{1}]}$ is markedly suppressed, while the peaks of $\kappa_{[101]}$ and $\kappa_b$ little change. These results indicate that there is a large contribution of spinons to $\kappa_{[10\bar{1}]}$ and suggest that the spin-chains run along the [101] direction.

1. Introduction

Recently, the thermal conductivity in low-dimensional quantum spin systems has attracted interest, because a large contribution of magnetic excitations to the thermal conductivity has been observed in some compounds regarded as low-dimensional quantum spin systems. In one-dimensional (1D) antiferromagnetic (AF) Heisenberg spin systems with the spin quantum number $S = 1/2$, especially, it has theoretically been proposed that the thermal conduction due to magnetic excitations, namely, spinons is ballistic [1, 2, 3]. In fact, a large contribution of spinons to the thermal conductivity has been observed in the $S=1/2$ 1D AF Heisenberg spin system $\text{Sr}_2\text{CuO}_3$ [4]. Moreover, the ballistic nature of the thermal conduction due to spinons has experimentally been confirmed in $\text{Sr}_2\text{CuO}_3$ [5].

The compound $\text{Sr}_2\text{V}_3\text{O}_9$ contains three kinds of vanadium ions in the unit cell. Two of them are nonmagnetic $V^{5+}$ ions located in $\text{VO}_4$ tetrahedra, and the rest is $V^{4+}$ ions with $S = 1/2$ located in $\text{VO}_6$ octahedra. The $\text{VO}_6$ octahedra are connected with each other by sharing an oxygen at the corner along the [101] direction, as shown Fig. 1. The $ac$-plane with the magnetic network is weakly stacked along the $b$-axis. The magnetic properties are understood in terms of the $S=1/2$ 1D AF chain model with the exchange interaction between the nearest spins, $J = 82$ K, estimated from the susceptibility measurements [6]. However, it has been suggested from the ESR measurements [7] and a theory by Koo and Whangbo [8] that the spin-chain direction is
not the [101] direction but the [10\overline{1}] direction, along which VO$_6$ octahedra are connected via a VO$_4$ tetrahedron as shown Fig. 1.

Therefore, in order to find the contribution of spinons to the thermal conductivity and also to clarify whether the spin-chain direction is the [101] or [10\overline{1}] direction, we have measured the thermal conductivity of Sr$_2$V$_3$O$_9$ along the [101], [10\overline{1}] directions and the $b$-axis. The precise measurement of thermal conductivity needs a large-size single crystal. Therefore, we have attempted to grow large-size single crystals of Sr$_2$V$_3$O$_9$ by the floating-zone (FZ) method.

2. Experimental

First, polycrystalline powder of Sr$_2$V$_2$O$_7$ was prepared by the solid-state reaction method, in order to prepare a feed rod for the FZ growth. The prescribed amount of SrCO$_3$ and V$_2$O$_5$ powder was mixed in the molar ratio of SrCO$_3$ : V$_2$O$_5$ = 2 : 1 and prefired in air at 700 °C for 72 h. After pulverization, the prefired powder of Sr$_2$V$_2$O$_7$ was mixed with VO$_2$ powder in the molar ratio of Sr$_2$V$_2$O$_7$ : VO$_2$ = 1 : 1 and isostatically cold-pressed at 600 bar into a rod of 7 mm in diameter and ~100 mm in length. Then, the rod was sintered at 540 °C in Ar for 24 h. As a result, a sintered feed rod was prepared. The FZ growth was carried out by the twice-scanning technique in an infrared heating furnace equipped with a double ellipsoidal mirror (NEC Machinery Corp, Model SC-K15HD-H). A high-density premelted feed rod was prepared through the first scan using the sintered feed rod. In the first scan, the molten zone was scanned at a speed of ~20 mm/h under flowing Ar of 1.5 atm. Next, the second scan, namely, a usual growing procedure was carried out using the premelted feed rod at the growth rate of 1.0 mm/h in the same atmosphere as in the first scan. Thermal-conductivity measurements were...
3. Results and Discussion

We have succeeded in growing a single-crystal rod, owing to the stable upkeep of the molten zone during the FZ growth. Figure 2(a) shows an as-grown single-crystal rod with ~ 6 mm in diameter and ~ 100 mm in length. The grown crystals were characterized by the x-ray back-Laue photography, as shown in Fig. 2(b). Although the grown crystals were composed of several domains, the diffraction spots were very sharp. The dimensions of the single-domain region were typically ~ 6 mm in diameter and ~ 25 mm in length. The single crystals were also confirmed by the powder x-ray diffraction to be of the single phase without any impurity phases. Accordingly, it is concluded that we have succeeded in the growth of high-quality single-crystals. The high quality was supported by the magnetic-susceptibility result that no Curie term due to impurities and/or lattice defects was observed at very low temperatures.

Figure 3 shows the temperature dependence of the thermal conductivity along the [101] direction, $\kappa_{[101]}$, along the [10\bar{1}] direction, $\kappa_{[10\bar{1}]}$, and along the b-axis, $\kappa_b$, in zero field and a magnetic field of 14 T parallel to the respective heat current.

In zero field, both $\kappa_{[101]}$, $\kappa_{[10\bar{1}]}$ and $\kappa_b$ show a peak around 10 K. The magnitude of $\kappa_{[10\bar{1}]}$ at the peak is larger than those of $\kappa_{[101]}$ and $\kappa_b$. Since $\text{Sr}_2\text{V}_3\text{O}_9$ is an insulator, the thermal conductivity is described as the sum of the thermal conductivity due to phonons, $\kappa_{\text{phonon}}$, and due to spinons, $\kappa_{\text{spinon}}$. It is known that the anisotropy of $\kappa_{\text{phonon}}$ is usually not so large and that the contribution of $\kappa_{\text{spinon}}$ markedly appears along the direction where the magnetic interaction is strong. Therefore, the large anisotropy of the thermal conductivity is guessed to be due to a large contribution of $\kappa_{\text{spinon}}$ to $\kappa_{[10\bar{1}]}$. By the application of magnetic field parallel to the heat current, the peak of $\kappa_{[10\bar{1}]}$ around 10K is suppressed with increasing field, while there is little change in $\kappa_{[101]}$ and $\kappa_b$, as shown in Fig. 3. This result also supports the guess that there is a large contribution of $\kappa_{\text{spinon}}$ to $\kappa_{[10\bar{1}]}$, because $\kappa_{\text{spinon}}$ is expected to be affected by the application of a magnetic field.
comparable with $J/(g\mu_B)$ ($g$: the $g$-factor, $\mu_B$: the Bohr magneton). Furthermore, the little change in $\kappa_{[101]}$ and $\kappa_b$ by the application of magnetic field indicates that the contribution of $\kappa_{\text{spinon}}$ is very small along the $[101]$ direction and $b$-axis. Accordingly, it is concluded that the spin-chain direction is the $[101]$ direction, as suggested from the ESR measurements [7] and the theory by Koo and Whangbo [8].

Here, we estimate $\kappa_{\text{spinon}}$ in $\kappa_{[101]}$, where both $\kappa_{\text{spinon}}$ and $\kappa_{\text{phonon}}$ are included. In the temperature dependence of $\kappa_{[101]}$, only one peak is observed around 10 K, indicating that both peaks of $\kappa_{\text{spinon}}$ and $\kappa_{\text{phonon}}$ are overlapped. Therefore, it is very hard to estimate $\kappa_{\text{spinon}}$ and $\kappa_{\text{phonon}}$ separately. As for $\kappa_{[101]}$, a small contribution of $\kappa_{\text{spinon}}$ to $\kappa_{[101]}$ is guessed to exist, because the $[101]$ direction is not exactly perpendicular to the $[10\overline{1}]$ direction but 84.48˚ tilted from the $[10\overline{1}]$ direction. As for $\kappa_b$, it is due to only $\kappa_{\text{phonon}}$. Therefore, neglecting the anisotropy of $\kappa_{\text{phonon}}$, $\kappa_{\text{spinon}}$ along the $[10\overline{1}]$ direction is very roughly estimated to be $\kappa_{[10\overline{1}]} - \kappa_b$, as shown in Fig. 4. However, this is not simply accepted as the temperature dependence of $\kappa_{\text{spinon}}$ along the $[10\overline{1}]$ direction, because unusual two peaks appear around 4 K and 28 K. What is remarkable at least is that the peak around 4 K is strongly suppressed by the application of magnetic field while the other peak around 28 K little changes. Therefore, it is expected that the peak around 4 K is attributed to the contribution of $\kappa_{\text{spinon}}$. On the other hand, it is likely that the peak around 28 K appears because of the difference of $\kappa_{\text{phonon}}$ between the $[10\overline{1}]$ direction and the $b$-axis. Accordingly, at least these results indicate that the temperature dependence of $\kappa_{\text{spinon}}$ exhibits a peak around 4 K in $\kappa_{[101]}$. In order to estimate the value of $\kappa_{\text{spinon}}$ in Sr$_2$V$_3$O$_9$ exactly, the estimate of the anisotropy of $\kappa_{\text{phonon}}$ between the $[10\overline{1}]$ direction and $b$-axis is necessary.

4. Summary

Large-size single-crystals of Sr$_2$V$_3$O$_9$ have successfully been grown by the FZ method and the thermal conductivity have been measured in magnetic fields up to 14 T. The magnitude of $\kappa_{[101]}$ in zero field is larger than those of $\kappa_{[101]}$ and $\kappa_b$. By the application of magnetic field, only $\kappa_{[101]}$ is suppressed. These anisotropic behaviors suggest that the spin-chains run along the $[101]$ direction. Moreover, it is concluded from the field effect of $\kappa_{[10\overline{1}]} - \kappa_b$ related to the behavior of $\kappa_{\text{spinon}}$ that the temperature dependence of $\kappa_{\text{spinon}}$ exhibits a peak around 4 K.

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