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

M Uesaka$^1$, T Kawamata$^2$, N Kaneko$^1$, M Sato$^1$, K Kudo$^3$, N Kobayashi$^3$, Y Koike$^1$

$^1$ Department of Applied Physics, Tohoku University, 6-6-05 Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan
$^2$ Nishina Center for Accelerator-Based Science, RIKEN, 2-1 Hirosawa, Wako 351-0198, Japan
$^3$ Institute for Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan
E-mail: uesaka@apph.tohoku.ac.jp

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 Sr$_2$V$_3$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 Sr$_2$CuO$_3$ [4]. Moreover, the ballistic nature of the thermal conduction due to spinons has experimentally been confirmed in Sr$_2$CuO$_3$ [5].

The compound Sr$_2$V$_3$O$_9$ contains three kinds of vanadium ions in the unit cell. Two of them are nonmagnetic V$^{5+}$ ions located in VO$_4$ tetrahedra, and the rest is V$^{4+}$ ions with $S=1/2$ located in VO$_6$ octahedra. The 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
Figure 1. Crystal structure of \( \text{Sr}_2\text{V}_3\text{O}_9 \). \( \text{V}^{5+} \) ions and \( \text{V}^{4+} \) ions are located in \( \text{VO}_4 \) tetrahedra and \( \text{VO}_6 \) octahedra, respectively. \( \text{VO}_6 \) octahedra are connected with each other along the [101] direction by sharing an oxygen at the corner and are also connected via a \( \text{VO}_4 \) tetrahedron along the [10\overline{1}] direction.

Figure 2. (a) Picture of an as-grown single-crystal rod of \( \text{Sr}_2\text{V}_3\text{O}_9 \). (b) X-ray back-Laue photography of an as-grown single-crystal in the x-ray parallel to the \( b \)-axis.

not the [101] direction but the [10\overline{1}] direction, along which \( \text{VO}_6 \) octahedra are connected via a \( \text{VO}_4 \) 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 \( \text{Sr}_2\text{V}_3\text{O}_9 \) 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 \( \text{Sr}_2\text{V}_3\text{O}_9 \) by the floating-zone (FZ) method.

2. Experimental

First, polycrystalline powder of \( \text{Sr}_2\text{V}_2\text{O}_7 \) was prepared by the solid-state reaction method, in order to prepare a feed rod for the FZ growth. The prescribed amount of \( \text{SrCO}_3 \) and \( \text{V}_2\text{O}_5 \) powder was mixed in the molar ratio of \( \text{SrCO}_3 : \text{V}_2\text{O}_5 = 2 : 1 \) and prefired in air at 700 °C for 72 h. After pulverization, the prefired powder of \( \text{Sr}_2\text{V}_2\text{O}_7 \) was mixed with \( \text{VO}_2 \) powder in the molar ratio of \( \text{Sr}_2\text{V}_2\text{O}_7 : \text{VO}_2 = 1 : 1 \) and isostatically cold-pressed at 600 bar into a rod of 7 mm in diameter and \( \sim100 \) 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 \( \sim20 \) 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
Figure 3. Temperature dependence of the thermal conductivity of Sr$_2$V$_3$O$_9$ along the [101] direction, $\kappa_{[101]}$, along the [101] direction, $\kappa_{[10\overline{1}]}$, and along the $b$-axis, $\kappa_b$, in zero field and a magnetic field of 14 T parallel to the respective heat current.

Figure 4. Temperature dependence of the difference between $\kappa_{[10\overline{1}]}$ and $\kappa_b$ in zero field and 14 T.

carried out by the conventional steady-state method.

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 $\sim 6$ mm in diameter and $\sim 100$ 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 $\sim 6$ mm in diameter and $\sim 25$ 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 [101] direction, $\kappa_{[10\overline{1}]}$, 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 $\kappa_{[101]}$, $\kappa_{[10\overline{1}]}$ and $\kappa_b$ show a peak around 10 K. The magnitude of $\kappa_{[10\overline{1}]}$ at the peak is larger than those of $\kappa_{[101]}$ and $\kappa_b$. Since Sr$_2$V$_3$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 $\kappa_{[10\overline{1}]}$. By the application of magnetic field parallel to the heat current, the peak of $\kappa_{[10\overline{1}]}$ 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 $\kappa_{[10\overline{1}]}$, 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 [101] direction but 84.48˚ tilted from the [101] 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 [101] direction is very roughly estimated to be $\kappa_{[101]} - \kappa_b$, as shown in Fig. 4. However, this is not simply accepted as the temperature dependence of $\kappa_{\text{spinon}}$ along the [101] 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[101] 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 [101] 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 $\kappa_{[101]} - \kappa_b$ 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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