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Evidence for Ballistic Thermal Conduction in the One-Dimensional Spin System Sr_2CuO_3

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Abstract. We have measured the thermal conductivity along the b axis, κ_b, parallel to the one-dimensional (1D) spin chain of $Sr_2Cu_{1-x}Pd_xO_3$ single crystals including nonmagnetic impurities of Pd^{2+}, in order to investigate possible ballistic thermal conduction. It has been found that the mean free path of spinons estimated from the thermal conductivity due to spinons, which is obtained by subtracting the thermal conductivity due to phonons from κ_b using the Debye model, is comparable with the average length of finite spin chains between spin defects estimated from the magnetic susceptibility measurements. This proves that the thermal conduction due to spinons at low temperatures in the 1D spin system Sr_2CuO_3 is ballistic.

Keywords: Thermal conductivity, Low-dimensional quantum spin system, Impurity effects
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Recently, thermal conductivity in low-dimensional quantum spin systems has attracted interest, because a large thermal conductivity due to spin excitations has been found in various materials [1–4]. In particular, in one-dimensional (1D) integrable Hamiltonian spin systems with the spin quantum number $S = 1/2$, it has theoretically been predicted that the thermal conduction due to spinons is ballistic [5, 6]. In fact, it has been reported that the thermal conductivity due to spinons, κ_{sp}, is large in Sr_2CuO_3 with 1D $S = 1/2$ chains [3, 4] and this compound can be regarded as an ideal 1D antiferromagnetic Heisenberg chain system. If the thermal conduction is ballistic, it is expected that the mean free path of spinons, l_{sp}, changes according to the length of finite spin chains.

In this paper, in order to confirm the ballistic nature of κ_{sp} in Sr_2CuO_3, we have measured the thermal conductivity along the b axis parallel to the 1D spin chain, κ_b, for $Sr_2Cu_{1-x}Pd_xO_3$ (x = 0, 0.004, 0.010) single crystals including nonmagnetic impurities of Pd^{2+}. We then compared the value of l_{sp} estimated from κ_{sp} with the average length of finite spin chains, between spin defects, estimated from the magnetic susceptibility measurements.

Single crystals of $Sr_2Cu_{1-x}Pd_xO_3$ (x = 0, 0.004, 0.010) were grown by the traveling-solvent floating-zone method and then were annealed at 870 °C for 72 h in Ar [7]. Thermal-conductivity measurements were carried out by the conventional steady-state method. The magnetic susceptibility was measured using a SQUID magnetometer (Quantum Design, MPMS-XL5).

Figure 1 shows the temperature dependence of κ_b in $Sr_2Cu_{1-x}Pd_xO_3$ (x = 0, 0.004, 0.010). The temperature dependence of the thermal conductivity along the a axis perpendicular to the spin chain, κ_a, of Sr_2CuO_3 is also shown in the inset of Fig. 1. For $x = 0$, κ_a increases with decreasing temperature and exhibits a peak at ∼ 25 K. This is a typical behavior of the thermal conductivity due to phonons, κ_{ph}. On the other hand, κ_b increases with decreasing temperature and exhibits a small shoulder due to κ_{sp} at around 70 K in addition to the peak due κ_{ph} at ∼ 25 K. These behaviors are similar to previous results [3, 4]. For x = 0.004 and 0.010, it is found that both peaks at ∼ 25 K and the small shoulder around 70 K are suppressed by Pd^{2+} doping. These results suggest that both phonons and spinons are scattered by Pd^{2+} so that their mean free paths become shorter.

First, κ_{ph} is estimated from the fit of the data of κ_a using the following equation based on the Debye model [8], as shown by the solid line in the inset of Fig. 1.

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

where $x = \hbar\omega/k_B T$, ω is the phonon angular frequency, v_{ph} is the phonon velocity and Θ_D is the Debye temperature. The phonon scattering rate τ_{ph}^{-1} is assumed to be given by the summation of scattering rates due to various scattering processes as follows,

$$\tau_{ph}^{-1} = \frac{v_{ph}}{L_b} + A\omega^4 + B\omega^2 T \exp(-\frac{\Theta_D}{bT}), \qquad (2)$$

where L_b, A, B and b are constants. The first term represents the phonon scattering by boundaries; the second, the phonon scattering by point defects; the third, the phonon-phonon scattering in the umklapp process. Here, Θ_D is set to 470.8 K from our specific heat measurements

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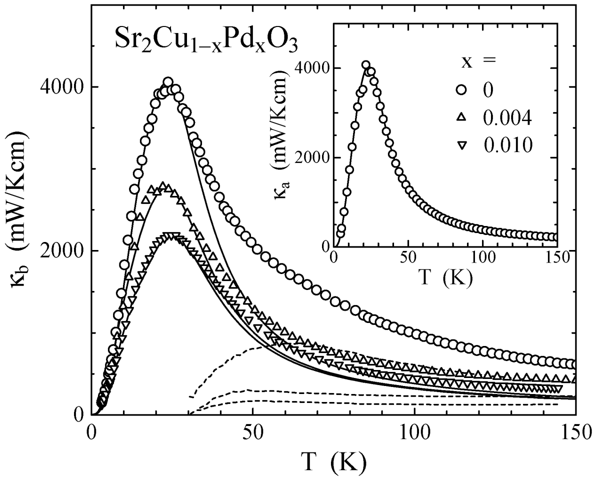

FIGURE 1. Temperature dependence of κ_b parallel to the spin chains of $Sr_2Cu_{1-x}Pd_xO_3$ with $x = 0$, 0.004, 0.010. The inset shows the temperature dependence of κ_a perpendicular to the spin chains of Sr_2CuO_3. The solid lines are κ_{ph} estimated using the Debye model. The dashed lines are κ_{sp} obtained by subtracting κ_{ph} from κ_b for $x = 0$ (upper), 0.004 (middle) and 0.010 (lower).

and v_{ph} is calculated as $v_{ph} = \Theta_D(k_B/\hbar)(6\pi^2 n)^{-1/3}$, where n is the number density of atoms. On the other hand, κ_{ph} of the Pd-doped Sr_2CuO_3 is estimated by the fit to the data of κ_b at low temperatures below ~ 25 K, adjusting L_b and A, which depend on the phonon scattering by boundaries and by point defects, respectively, under the assumption that the phonon-phonon scattering in the umklapp process is not affected by the small doping of Pd^{2+}. Then, κ_{sp} is estimated by subtracting the fitting curve of κ_{ph} from the data of κ_b, as shown by dashed lines in Fig. 1.

Next, l_{sp} is calculated from κ_{sp}, using the following equation, namely, the product of the specific heat, the velocity and the mean free path of spinons based on the $S = 1/2$ 1D Heisenberg model [9, 10].

$$\kappa_{sp} = \frac{\pi N a k_B^2 T}{3\hbar} l_{sp}, \tag{3}$$

where N is the number of spins per unit volume and a is the distance between spins in the chain. The calculated values of l_{sp}^{-1} are fit to the following equation [3].

$$l_{sp}^{-1} = AT\exp(-T^*/T) + L^{-1}, \tag{4}$$

where A, T^* and L are constants. The first term represents the scattering in the umklapp process and T^* is the characteristic temperature. The second term represents the temperature-independent spinon scattering by spin defects, etc. L is regarded as the upper-limit value of l_{sp} at low temperatures. The obtained values of L are listed in Table 1.

Finally, in order to estimate the average length of finite spin chains between spin defects, L_{imp}, the number of free spins per Cu, x_{Curie}, is obtained from the Curie constant. This is estimated from the fit of the magnetic susceptibility data using the function given by the sum a Curie-Weiss term, a Cu^{2+} spin term suggested by Eggert *et al.*[11], a Van Vleck paramagnetic term and a core diamagnetic term. The obtained values of x_{Curie} are roughly a half of x values for $x = 0.004$ and 0.010, respectively, as listed in Table 1. This is reasonable, because one antiferromagnetic spin chain with an odd number of spins has a free spin of $S = 1/2$ and one chain with an even number of spins exhibits $S = 0$. Consequently, one free spin is induced, on average, by two spin defects [12, 13]. Values of L_{imp} calculated from x_{Curie} are also listed in Table 1. Surprisingly, it is found that the values of L_{imp} are very close to those of L. In this analysis, since κ_{ph} is estimated using the data at low temperatures (below ~ 25 K), κ_{sp} is underestimated to be zero in this temperature range. That is, the L value is underestimated. In any case, this means that l_{sp} at low temperatures is approximately limited by L_{imp}. That is, the thermal conduction due to spinons is limited at low temperatures only by scattering from spin defects. Accordingly, it is concluded that the thermal conduction due to spinons at low temperatures is ballistic.

TABLE 1. Parameters estimated from the thermal conductivity and the magnetic susceptibility measurements in $Sr_2Cu_{1-x}Pd_xO_3$.

x	x_{Curie}	L (Å)	L_{imp} (Å)
0	0.0010	2107.8	1956.5
0.004	0.0026	821.4	752.6
0.010	0.0042	498.2	465.8

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