In-Plane Thermal Conductivity of Nd_2CuO_4: Evidence for Magnon Heat Transport

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We report the temperature and magnetic field dependence of the in-plane thermal conductivity (κ_{ab}) of high-quality monocrystalline Nd_2CuO_4. Isothermal measurements of the field dependence of κ_{ab} at low temperatures (2 K $\leq T \leq$ 5 K) show no change in κ_{ab} below a critical magnetic field H_c ($H_c \approx$ 4.5 T for $H \parallel [100]$) and $H_c \approx 2.5$ T for $H \approx\parallel [110]$). Above H_c, κ_{ab} more than doubles as H is increased to 9 T. At H_c, there is a transition from a noncollinear to a collinear arrangement of the Nd and Cu spins and a collapse of the gap, Δ, in an acoustic magnon branch at $\mathbf{k} = 0$. Closure of this gap appears to allow the conduction of substantial amounts of heat by acoustic magnons.

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Thermal conductivity measurements can be used to probe magnetic excitations in low dimensional magnetic insulators [1–5]. In the quasi-two-dimensional (2D) Heisenberg antiferromagnet $K_2V_3O_8$, we recently reported a remarkable increase in the in-plane thermal conductivity, κ_{ab}, as a function of magnetic field [1]. $K_2V_3O_8$ is tetragonal at all temperatures and the magnetic structure consists of antiferromagnetic planes of V^{4+} (spin = 1/2) arranged in a body-centered square lattice. In this system, the additional heat conduction is found to coincide with the closure of the anisotropy gap, Δ_a, in the acoustic magnon spectrum at zero wave vector, i.e., $\mathbf{k} = 0$ [1,6,7]. Additional heat conduction occurs only for magnetic fields larger than $\Delta_a/g\mu_B$. It was not clear to us if this additional channel for heat conduction is a feature of most quasi-2D antiferromagnets or only peculiar to $K_2V_3O_8$.

In this Letter, we report in-plane thermal conductivity measurements of Nd_2CuO_4 as a function of temperature and magnetic field. Nd_2CuO_4 is a quasi-2D antiferromagnet, where the dominant magnetic interaction is the antiferromagnetic exchange ($J_{\text{Cu-Cu}}/k_B \sim 1000$ K) between Cu spins in the same layer [see Fig. 1(a)] [8,9]. The magnetism of all of the rare-earth (R) cuprate R_2CuO_4 systems, including Nd_2CuO_4, has been extensively studied because of the entanglement of magnetism and superconductivity in doped members of these compounds. Although the magnetism and spin-wave spectrum of Nd_2CuO_4 is complicated because of the presence of two magnetic ions (Nd and Cu) that are coupled to each other [8,9], the low-temperature behavior of the in-plane thermal conductivity in a magnetic field is remarkably similar to that of $K_2V_3O_8$. The present results thus suggest that the excess heat conduction observed in a magnetic field large enough to close a gap in an acoustic magnon branch may be a general feature of quasi-2D antiferromagnets. Although not discussed in the present Letter, similar features in the magnetic field dependence of κ_{ab} have also been observed in crystals of Pr_2CuO_4.

The Nd_2CuO_4 single crystals used for this study were grown using both the traveling-solvent-floating-zone (TSFZ) technique and the flux method. Four-point thermal conductivity measurements were performed in a physical property measurement system from quantum design. The Cernox thermometers we used have a weak field dependence of $[T(H = 9\,\text{T})\text{-}T(H = 0)]/T(H = 0) =$ 3.1% at 2 K and less at higher temperatures [10]. According to our previous measurements on nonmagnetic materials, the apparent field-induced thermal conductivity change is less than 4% at 2 K and smaller at higher temperatures. The results obtained from the TSFZ crystal

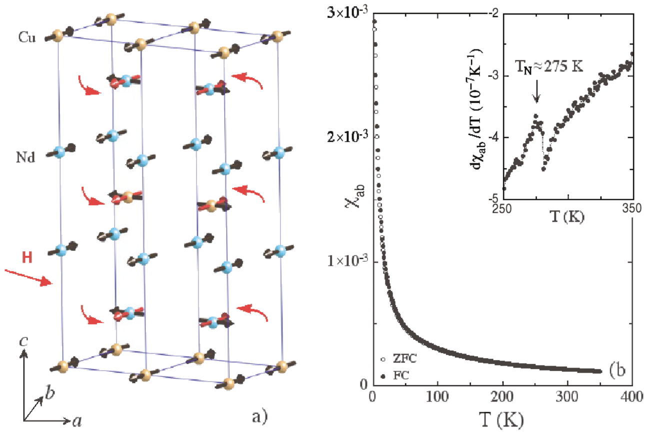

FIG. 1 (color). (a) Magnetic structure of Nd_2CuO_4 below 30 K. The brown dots denote the Cu ions and the blue ones Nd ions. Red arrows point the rotation direction of spins when H is applied along the [100] direction. (b) Temperature dependence of the in-plane magnetic susceptibility (χ_{ab}) of Nd_2CuO_4 measured by applying $H = 1000$ Oe along the ab plane. The inset is the plot of $d\chi_{ab}/dT$ vs T between 250 and 350 K, where the Néel temperature is indicated.

(a rod with dimensions of ϕ 4 mm × 10 mm) and the flux-grown plate [$(3 \times 2 \times 0.5)$ mm^3] are identical within experimental error.

In Nd_2CuO_4, the Cu sublattice orders antiferromagnetically at $T_N \approx 275$ K [Fig. 1(b)]. On further lowering temperature, the large exchange coupling between the Cu and Nd polarizes the Nd spins and induces an ordered moment. Below about 30 K, the ordered arrangement of the Cu and Nd spins is noncollinear as shown in Fig. 1(a) [8,9,11–14]. The temperature dependence of the ordered Nd moment is qualitatively similar to the in-plane susceptibility data shown in Fig. 1(b), around $1.5\mu_B$ at 2 K and $0.05\mu_B$ at 50 K [8]. At 5 K, the application of a magnetic field of about 4.5 T along the [100] direction results in a spin-flop transition, in which the Cu and Nd spins oriented along the [100] direction rotate in-plane by 90° [8,12,14]. This results in a collinear spin structure in which all of the ordered moments are approximately perpendicular to H. If a smaller field ($\sim$ 0.7–2 T) is applied along the [110] direction, there is a spin-flop transition in which all of the spins rotate in-plane by about 45°, giving rise to the same collinear spin structure. The field, at which each of these transitions occurs, has been studied using several techniques [8,9,11–15].

In an insulating magnetic crystal, heat can be carried by phonons and/or magnons. As the first approximation, the total thermal conductivity can be written as $\kappa \approx \kappa_{\mathrm{ph}} + \kappa_{\mathrm{mag}}$. Here, κ_{ph} and κ_{mag} are the thermal conductivities due to acoustic phonons and acoustic magnons, respectively. In addition, magnons can scatter phonons and vice versa. For both κ_{ph} and κ_{mag}, the simplest expression is $\kappa_i = (1/\mathrm{D})C_i v_i d_i$ ($i =$ ph, mag), where D is the dimension, C_i is the specific heat due to phonons or magnons, v_i is the sound velocity or the magnon group velocity, and d_i is the mean-free path.

The main panel of Fig. 2 presents the temperature dependence of κ_{ab} of Nd_2CuO_4 for $H = 0$ (open circles) and 9 T (solid circles) with $H \parallel ab$. In both cases, κ_{ab} varies nonmonotonically with temperature and exhibits a broad maximum near 250 K, a shallow minimum near 100 K, followed by a sharp maximum at 20 K. The broad maxima near 250 K does not normally arise from phonon heat transport in an insulating crystal. Similar high-temperature maxima in κ_{ab} have been seen in high-quality La_2CuO_4 crystals and other lightly hole-doped cuprates [16–20]. While it was attributed to the phonon damping for $YBa_2Cu_3O_{6+x}$ [16], very recent work suggests that magnetic excitations associated with the Cu sublattice carry heat [2,5,21]. Below $\sim$100 K, κ_{ab} increases with decreasing temperature and rapidly decreases after reaching a maximum at $\sim$20 K. This behavior is typical of phonon heat conduction in insulating crystals.

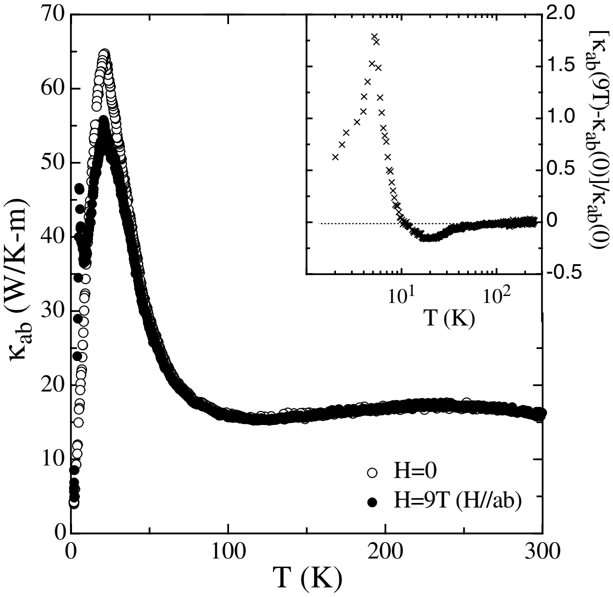

FIG. 2. Linear plot of temperature dependence of κ_{ab} of Nd_2CuO_4 at $H = 0$ (open circles) and $H = 9$ T (solid circles) with $H \parallel ab$. The inset shows the temperature dependence of magnetothermal conductivity $\kappa_{ab}(H)$-$\kappa_{ab}(0)/\kappa_{ab}(0)$ at $H = 9$ T. Note that the application of magnetic field induces a peak near 5 K and a sign change of $\kappa_{ab}(H)$-$\kappa_{ab}(0)/\kappa_{ab}(0)$ at 10 K.

The overall effect of a field of 9 T on κ_{ab} is shown in the inset of Fig. 2 plotted as $[\kappa_{ab}(9\ \mathrm{T})$-$\kappa_{ab}(0)]/\kappa_{ab}(0)$ versus T. Above $\sim$80 K, a field of 9 T does not have any detectable effect on κ_{ab}. However, there is a large change in $[\kappa_{ab}(9\ \mathrm{T})$-$\kappa_{ab}(0)]/\kappa_{ab}(0)$ below $\sim$80 K and it is this change that is the main focus of this Letter. With decreasing T, $[\kappa_{ab}(9\ \mathrm{T})$-$\kappa_{ab}(0)]/\kappa_{ab}(0)$ initially decreases. After decreasing about 20%, $[\kappa_{ab}(9\ \mathrm{T})$-$\kappa_{ab}(0)]/\kappa_{ab}(0)$ reaches a minimum at 20 K, below which it increases with decreasing T and changes the sign at 10 K. Of prominence is the H-induced peak at 5 K, where $\kappa_{ab}(9\ \mathrm{T})$ increases by about 180% compared to $\kappa_{ab}(0)$.

If the heat is carried only by phonons at low temperatures, $\kappa_{ab}(\mathrm{T})$ should be proportional to C_{ph}, since v_{ph} and d_{ph} are approximately constants. The low-temperature specific heat of Nd_2CuO_4 is complicated by the exchange splitting of the Nd ground-state doublets [22], which produce a Schottky peak near 1.5 K for $H = 0$ and 3 K for $H = 9$ T [23]. Could the maxima in $[\kappa_{ab}(9\ \mathrm{T})$-$\kappa_{ab}(0)]/\kappa_{ab}(0)$ near 5 K result from the Schottky contribution? If it were the dominant factor in the low-temperature behavior of κ_{ab}, one would expect κ_{ab} to increase with decreasing temperature for $1.5\ \mathrm{K} < T < 10$ K. As can be seen from Fig. 2, $\kappa_{ab}(0)$ varies smoothly with T, showing no sign of upturn as seen in specific heat [22]. Thus, we believe that the peak in $[\kappa_{ab}(9\ \mathrm{T})$-$\kappa_{ab}(0)]/\kappa_{ab}(0)$ cannot be simply attributed to the Schottky effect. Furthermore, careful modeling of the temperature and field dependence of the Scottky anomaly has shown that, within experimental resolution, it is totally due to nondispersive magnon modes (no group velocity) associated with the Nd spins [24]. These magnons serve only as scatterers in heat transport.

To further explore the origin of the dramatic effect of a magnetic field on κ_{ab}, we measured the field dependence of κ_{ab} between 0 and 9 T at fixed temperatures. Figure 3 shows κ_{ab} versus H at $T = 2$, 5, 10, and 18 K for (a) $H \parallel [100]$ and (b) $H \approx\parallel [110]$, respectively. For $T = 2$ and 5 K, κ_{ab} has little change until H reaches a critical value H_c, that depends on the direction of the magnetic field. When H is applied along the [100] direction, $H_c \approx 4.5$ T, a field coinciding with that for the noncollinear to collinear spin-flop transition [8,12,14]. When H is applied nearly along the [110] direction, we obtain $H_c \approx 2.5$ and 0.65 T (not shown) for TSFZ- and flux-grown crystals, respectively. Compared to the value of 0.7 T determined from magnetization [12] and neutron scattering measurements [14] on flux-grown crystals for the spin-flop transition, H_c for our TSFZ-grown crystal is somewhat large. Apart from the possibility that H_c may be crystal-growth-technique dependent, it should be mentioned that the applied field in our experiments may has not been exactly parallel to the [110] direction of the TSFZ-grown crystal due to the unusual growth habit. Nevertheless, as noted by Petitgrand and co-workers [9], these spin-flop transitions coincide with a collapse of a gap Δ in one branch of the magnon spectrum of Nd_2CuO_4 at $\mathbf{k} = 0$.

The isothermal measurements of the field dependence of κ_{ab} at 2 and 5 K [see Fig. 3(a)] are remarkably similar to the behavior we observed in $K_2V_3O_8$ [1]. In $K_2V_3O_8$, the magnon spectrum has been measured [7] and is relatively simple, consisting of a doubly degenerate acoustic branch with a small anisotropy gap Δ_a at $\mathbf{k} = 0$. Closure of this gap with a magnetic field of magnitude $H_c = \Delta_a/g\mu_B$ results in a rapid increase of κ_{ab} for $H > H_c$. The magnon spectrum of Nd_2CuO_4 is considerably more complicated [8,9,25], consisting of several low-energy branches (< 1 meV) with excitations associated primarily with Nd spins and high-energy excitations (> 1 meV) involving mainly Cu spins. Most of the low-energy and high-energy modes are essentially optical magnon modes that cannot carry heat, as their group velocity is near 0. These optical modes, however, can scatter acoustic phonons and magnons. Sachidanandam and co-workers predict an acoustic magnon mode with large dispersion (high velocity) and a very small gap at $\mathbf{k} = 0$ [8]. (To the best of our knowledge, this mode has not yet been experimentally observed.) We believe it is primarily magnon heat conduction associated with this mode that is responsible for the dramatic increase in κ_{ab} in Nd_2CuO_4 when $H > H_c$. For $H \parallel [100]$, $H_c = 4.5$ T implies a gap of about 0.3 meV, which is about 60 times larger than that estimated by theory [8]. This discrepancy between experimental and theoretical values of the in-plane gap may be due to the simplified model calculation, which did not consider additional exchange-interaction complications, as pointed out by Petitgrand and co-workers [9].

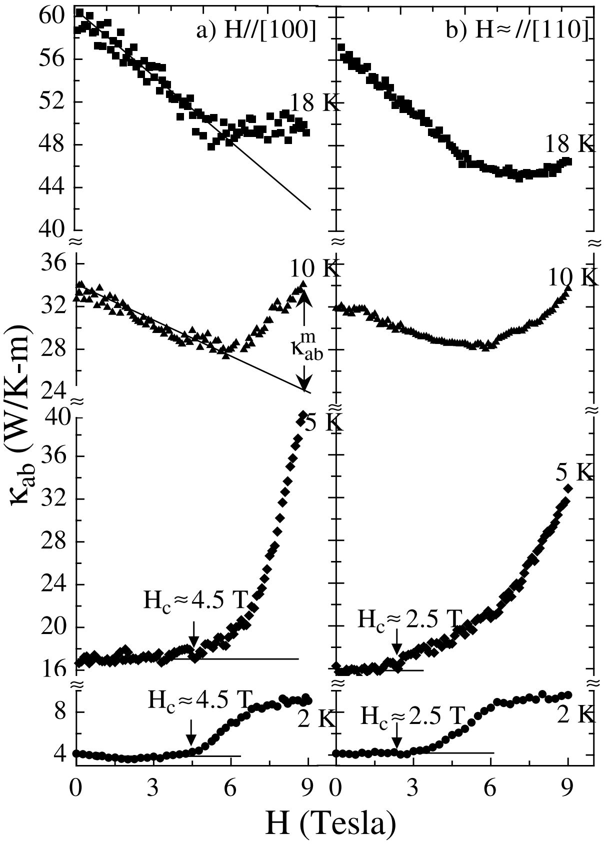

FIG. 3. Magnetic field dependence of κ_{ab} of Nd_2CuO_4 at $T = 2$ (solid circles), 5 (diamonds), 10 (triangles), and 18 K (squares) for (a) $H \parallel [100]$ and (b) $H \approx\parallel [110]$, respectively. The dashed lines are an estimate of thermal conductivity change due to phonon-magnon scattering.

When $H = H_c$, the lowest acoustic magnon branch becomes gapless at $\mathbf{k} = 0$ and a new magnetic ground state is formed. A plausible interpretation of the experimental results is that only magnons near $\mathbf{k} = 0$, which collapse into the gapless lower branch, carry a significant amount of heat. In the case of $K_2V_3O_8$, the experimental data strongly suggest that the temperature and field dependence of the excess thermal conductivity for $H > H_c$ is proportional to the population of these magnons P_m [1]. Phenomenologically, P_m can be expressed as

$$P_m(T, H > H_c) = \frac{e^{-[(\Delta - g\mu_B H)/(k_B T)]}}{e^{-[(\Delta - g\mu_B H)/(k_B T)]} + e^{-[(\Delta + g\mu_B H)/(k_B T)]}} - \frac{1}{1 + e^{-[(2\Delta)/(k_B T)]}} \qquad (1)$$

for $H > H_c$ and $P_m = 0$ for $H \le H_c$. Though this is a purely phenomenological expression, it seems to provide a reasonable description of our experimental results from Nd_2CuO_4 as is shown in Figs. 4(a) and 4(b). The solid line

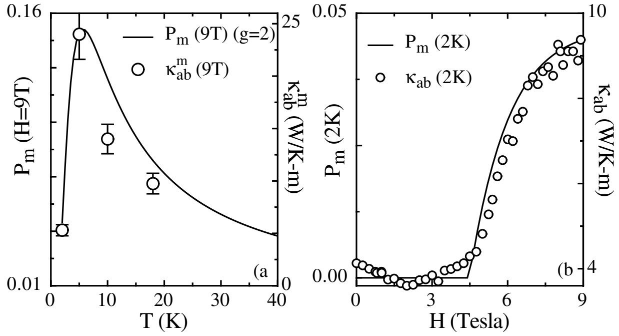

FIG. 4. (a) Comparison of magnon-related thermal conductivity κ_{ab}^m (open circles) with the magnon population (P_m) residing in the lower branch of the magnon spectrum at $H = 9$ T (solid line); (b) magnetic field dependence of P_m (solid line) and κ_{ab} (open circles) at 2 K. As the experimental data are for $H \parallel [100]$, $H_c = 4.5$ T are used for calculating P_m.

in Fig. 4(a) is the plot of P_m versus T, corresponding to $H_c = 4.5$ T and $H = 9$ T. Note that P_m (T) peaks at 5 K, consistent with the experimental observation (open circles). For the unique field dependence of κ_{ab} at 2 and 5 K, Eq. (1) also works very well, as demonstrated in Fig. 4(b).

For temperatures slightly larger than 5 K, κ_{ab} is no longer constant with H for $H < H_c$, but more or less linearly decreases with increasing H (see dashed lines in Fig. 3). At higher temperatures, there are more acoustic phonons and thermally excited optical magnons. It is likely that the initial decrease of κ_{ab} is due to the scattering of acoustic phonons by a growing number of optical magnons [2]. Simple models suggest that, in antiferromagnetic systems, the phonon-magnon interaction reduces the thermal conductivity as H is increased [1,2]. The successive increase of κ_{ab} in the high-field regime is, again, because of acoustic magnons which carry considerable amounts of heat when the gap is closed by the magnetic field.

For $K_2V_3O_8$, the specific heat shows an anomaly at the spin-flop transition [26]. Such an anomaly has not been observed in Nd_2CuO_4 down to 2 K. This implies that, for Nd_2CuO_4, acoustic magnons associated with heat transport cannot be easily extracted from specific heat measurements that integrate over all available excitations. On the other hand, the spin-flop transition for $H \parallel [100]$ is visible only in magnetization data for temperatures below 2 K [12,26]. By contrast, this transition is still clearly observable at 18 K in the thermal conductivity data shown in Fig. 3, illustrating the sensitivity of this technique in studying certain types of magnetic excitations.

In all magnetically ordered materials, there is an anisotropy gap in the acoustic spin-wave branch that simply reflects a preferred direction for the ordered magnetic moments. This gap also reflects the *anisotropy* in the dominant exchange-interaction J. Hence, the magnitude of the gap can be significantly smaller than J. In the quasi-2D antiferromagnetic Nd_2CuO_4, we have demonstrated that the collapse of this gap with a magnetic field results in a remarkable increase in heat conduction. Although we have provided a simple phenomenological explanation of this effect, there is clearly a need for a good theoretical understanding of this phenomenon.

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[1] B.C. Sales, M. Lumsden, S. Nagler, D. Mandrus, and R. Jin, Phys. Rev. Lett. **88**, 95901 (2002).
[2] M. Hofmann *et al.*, Phys. Rev. Lett. **87**, 47202 (2001); Phys. Rev. B **67**, 184502 (2003).
[3] A.V. Sologubenko, K. Gianno, H.R. Ott, U. Ammerahl, and A. Revcolevschi, Phys. Rev. Lett. **84**, 2714 (2000).
[4] A. Vasil'ev *et al.*, Phys. Rev. Lett. **81**, 1949 (1998).
[5] C. Hess *et al.*, Phys. Rev. B **64**, 184305 (2001); Phys. Rev. Lett. **90**, 197002 (2003).
[6] M.D. Lumsden, B.C. Sales, D. Mandrus, S.E. Nagler, and J.R Thompson, Phys. Rev. Lett. **86**, 159 (2001).
[7] M. Lumsden and S. Nagler (unpublished).
[8] R. Sachidanandam *et al.*, Phys. Rev. B **56**, 260 (1997).
[9] D. Petitgrand, S.V. Maleyev, Ph. Bourges, and A.S. Ivanov, Phys. Rev. B **59**, 1079 (1999).
[10] B. Brandt, D. Liu, and L. Rubin, Rev. Sci. Instrum. **70**, 104 (1999).
[11] M. Matsuda *et al.*, Phys. Rev. B **42**, 10 098 (1990).
[12] A. Cherny *et al.*, Phys. Rev. B **45**, 12 600 (1992).
[13] J. Lynn *et al.*, Phys. Rev. B **41**, 2569 (1990).
[14] S. Skanthakumar, J.W. Lynn, J.L. Peng, and Z.Y. Li, J. Appl. Phys. **73**, 6326 (1993).
[15] S. Zherlitsyn *et al.*, Physica (Amsterdam) **211B**, 168 (1995).
[16] J.L. Cohn, C.K. Lowe-Ma, and T.A. Vanderah, Phys. Rev. B **52**, 13 134 (1995).
[17] Y. Nakamura *et al.*, Physica (Amsterdam) **185C–189C**, 1409 (1991).
[18] A. Inyushkin *et al.*, Physica (Amsterdam) **194B–196B**, 479 (1994).
[19] A. Inyushkin *et al.*, Physica (Amsterdam) **235C–240C**, 1487 (1994).
[20] P. Henning *et al.*, J. Supercond. **8**, 453 (1995).
[21] X. Sun, J. Takeya, S. Komiya, and Y. Ando, Phys. Rev. B **67**, 104503 (2003).
[22] T. Brugger *et al.*, Phys. Rev. Lett. **71**, 2481 (1993).
[23] L. Shan *et al.*, cond-mat/0209380.
[24] N.T. Hien *et al.*, Phys. Rev. B **57**, 5906 (1998).
[25] P. Thalmeier, Physica (Amsterdam) **266C**, 89 (1996).
[26] R. Jin *et al.* (unpublished).