Evidence for a large magnetic heat current in insulating layered cuprates

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The in-plane thermal conductivity k of the two-dimensional antiferromagnetic monolayer cuprate $Sr_2CuO_2Cl_2$ is studied. Analysis of the unusual temperature dependence of k reveals that at low temperatures the heat is carried by phonons, whereas at high temperatures magnetic excitations contribute significantly. A comparison with other insulating layered cuprates suggests that a large magnetic contribution to the thermal conductivity is an intrinsic property of these materials.

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There is growing experimental evidence that spin excitations may contribute significantly to the heat current in low-dimensional spin systems. This seems to be well established for one-dimensional (1D) systems.[1–5] For example, in the insulating spin-ladder material $Sr_{14-x}Ca_xCu_{24}O_{41}$, a large magnetic contribution k_m to the thermal conductivity k can be derived from a pronounced double-peak structure of k along the ladder direction.[2,4] The situation is less clear in two-dimensional spin systems. These are, however, of particular importance due to their relevance for high-temperature superconductivity.[6,7] A double-peak structure comparable to that in 1D systems is found in the in-plane thermal conductivity of insulating 2D cuprates such as La_2CuO_4 (LCO) and $YBa_2Cu_3O_6$ (YBCO) (Refs. 8–10). This may indicate a sizable magnetic contribution to the heat current at high temperatures.[8] However, the phononic thermal conductivity k_{ph} may show a double-peak structure also, as a result of pronounced (resonant) scattering in a narrow temperature range. Such scattering may arise from the presence of local magnetic excitations, as was recently shown for the 2D spin-dimer system $SrCu_2(BO_3)_2$ (Ref. 11), or it may arise from the presence of soft phonon modes.[9] The latter was suggested for LCO and YBCO, in which soft modes, e.g., associated with tilt distortions of the CuO polyhedra are known to be present.[9,12] An additional complication arises from a strong sensitivity of the double-peak structure to oxygen doping.[9]

A material of particular interest in this context is $Sr_2CuO_2Cl_2$ (SCOC). It is structurally very similar to LCO. It contains CuO_2 layers as in LCO, but the out-of-plane oxygen ions at the apices of the CuO_6 octahedra are replaced by Cl and La by Sr. The material has following advantages compared to LCO and YBCO (see, e.g., Ref. 6).

(1) SCOC does not exhibit any distortion from tetragonal symmetry down to at least 10 K so that there is no structural instability associated with soft tilting modes.

(2) Because of the absence of tilt distortions, the magnetic properties are simpler than those of LCO. For example, there is no Dzyaloshinski-Moriya exchange interaction. Thus, SCOC is believed to represent the best realization of a two-dimensional $S=1/2$ square-lattice Heisenberg antiferromagnet.

(3) In contrast to LCO and YBCO, SCOC cannot be doped easily with charge carriers.

In this paper, we present measurements of the in-plane thermal conductivity k of SCOC. We identify a double-peak structure from a pronounced high-temperature shoulder around 230 K. Analysis of these data, and a comparison to LCO and YBCO shows that it is very unlikely that the double-peak structure arises from anomalous phonon damping due to scattering on soft lattice modes or magnetic excitations. The data indicate instead a large magnetic thermal conductivity at high temperatures as an intrinsic feature of the insulating 2D cuprates.

We studied a single crystal of $Sr_2CuO_2Cl_2$ of rectangular form ($1\times3\times4$ mm^3) with the short direction along the crystallographic c axis. It was grown by the traveling-solvent floating zone method. The thermal conductivity was measured with the heat current within the CuO_2 planes by a conventional steady-state method using a differential Chromel-Au+0.07%Fe-thermocouple. Typical temperature gradients were of the order of 0.2 K. The absolute accuracy of our data is restricted by uncertainties in the sample geometry, whereas the relative accuracy is of the order of a few percent.[13]

We show in Fig. 1 the in-plane thermal conductivity of SCOC as a function of temperature. We identify a maximum at $\approx$30 K and a shoulder at high temperatures around 230 K. The pronounced low-temperature maximum of k indicates the high crystal quality. We note that k is independent of a magnetic field ($\leqslant$8 T) applied within the CuO_2 planes perpendicular to the heat current. For comparison, we show in Fig. 1 the in-plane thermal conductivity of a single crystal of LCO, measured by Nakamura *et al.* (Ref. 8). These data also reveal a double-peak structure. The absolute value of k at the low-temperature maximum is smaller than in SCOC. One reason may be that LCO is more sensitive to defects, resulting, e.g., from excess oxygen, which introduces lattice defects and hole doping and thus reduces the mean free path of the heat-carrying excitations.

Both compounds, SCOC and LCO, are antiferromagnetic insulators. In an insulator, the heat is usually carried by phonons. The typical behavior of k_{ph} of a crystalline insulator is shown by the solid lines in Fig. 1. These curves represent fits to the low-temperature maximum of k (fitted below

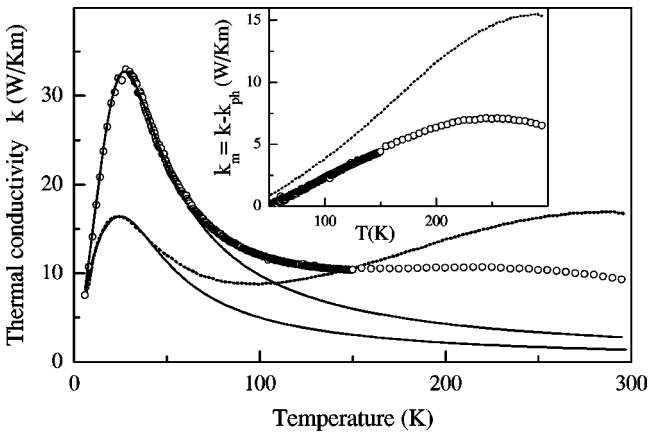

FIG. 1. In-plane thermal conductivity $k(T)$ of $Sr_2CuO_2Cl_2$ (circles) and La_2CuO_4 [dotted line; data from Nakamura *et al.* (Ref. 8)]. Solid lines: fits to k_{ph} using the Debye model (Refs. 11,14). The fitting parameters for $Sr_2CuO_2Cl_2$ (La_2CuO_4) are $D/10^{-17}$ s $=3.4(2.6)$; $P/10^{-43}$ s$^3=0.15(21)$; $U/10^{-30}$ s^2/K$=1.0(2.2)$; and $u=4.9(4.4)$. The point defect scattering (P) is smaller in SCOC. Inset: The magnetic contribution $k_m=k-k_{ph}$ for $Sr_2CuO_2Cl_2$ (circles) and La_2CuO_4 (dotted line) (see text).

about 50 K) of SCOC and LCO, using the standard Debye model for the thermal conductivity of acoustic phonons:[11,14]

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

Here, Θ_D is the Debye temperature and v_{ph} is the sound velocity. Due to the lack of experimental data for SCOC we use for both compounds the values reported for LCO [$\Theta_D \approx 385$ K;[15] $v_{ph}\approx 5.2\times 10^3$ m/s (Ref. 16)]. ω is the phonon frequency, $x=\hbar\omega/k_BT$, and $\tau(x,T)$ is the phonon relaxation time given by

$$\tau^{-1}=\frac{v_{ph}}{L}+D\omega^2+P\omega^4+UT\omega^3\exp\left(\frac{\Theta_D}{uT}\right). \qquad (2)$$

The four terms refer to the scattering rates for boundary scattering, scattering on planar defects, on point defects, and phonon-phonon Umklapp scattering, respectively. $L\approx 1$ mm is the sample length, and D, P, U, and u are fitting constants. The low-temperature data are described very well by these fits. The fit parameters are given in the caption of Fig. 1. The decrease of k_{ph} at high temperatures is due to phonon-phonon Umklapp scattering.

For following reasons, it is very unlikely that the high-temperature increase of k is due to conventional heat transport by phonons.

(1) The contribution to k from acoustic phonons, as described above, decreases at high temperatures.

(2) The contribution of optical phonons to the heat current is usually much smaller than that of acoustic phonons, even in compounds with a very large number of atoms in the unit cell,[5,17] so that heat transport by optical phonons is very unlikely to cause the high-temperature maximum.

(3) The out-of-plane thermal conductivity k_c of LCO behaves as the in-plane thermal conductivity k at low temperatures, but k_c shows no indication of a high-temperature maximum.[8] Such a strongly temperature-dependent anisotropy is not expected for purely phononic heat conduction.

Additional phonon scattering, active in a narrow temperature range close to the minimum of k, may, in principle, cause a double-peak structure. However, resonant scattering on local magnetic excitations as in $SrCu_2(BO_3)_2$ (Ref. 11) cannot be the correct explanation in the present case. In the 2D square-lattice cuprates, the dispersion of magnetic excitations ranges from ≈ 0 to $2J/k_B \gtrsim 2000$ K (J is the in-plane exchange constant) so that there is no reason that scattering on magnetic excitations should be most pronounced in a narrow temperature interval around 100 K.[11,19]

Additional phonon damping from scattering on soft lattice modes, as suggested in Refs. 9 and 18, is unlikely as a cause of the double peak for the following reasons.

(1) There are no lattice instabilities in SCOC, rendering this mechanism unimportant for this material.

(2) The double-peak structure is also present in the tetragonal low-temperature phase of Eu-doped LCO, in which no soft tilting modes should be present either.[10]

(3) The absence of the double-peak structure of k_c in LCO (Ref. 8) implies that anomalous phonon scattering would have to be active only for the in-plane thermal conductivity k. Such a strong anisotropy of the phonon-phonon scattering is not expected. Finally, note that the finding $k_c < k$ in LCO (Ref. 8) provides evidence against any scattering scenario as a cause of the double-peak structure. Such a scattering, if active only for k, but absent for k_c, implies $k<k_c$, in contradiction to the experimental results.

The data of Fig. 1 (in particular $k>k_c$) are most naturally explained, if an additional channel of heat transport for the in-plane thermal conductivity is present. In an undoped insulating 2D Heisenberg antiferromagnet with an electronic gap $\gtrsim 1.5$ eV, the only candidate for heat transport next to phonons are magnetic excitations. Their thermal conductivity k_m adds to that of the phonons, i.e., $k=k_{ph}+k_m$. For a quantitative estimate of k_m from the data, we subtract k_{ph} as obtained from the fit of the low-temperature maximum from k. Note that k_m cannot be obtained at $T\lesssim 100$ K in this way, because Eq. (1) was fitted to the *total* k below 50 K. Remarkably, k_m is of comparable magnitude (roughly of the order 10 W/Km) in both compounds (see inset of Fig. 1). The maximum of k_m is at ≈ 245 K in SCOC and at ≈ 285 K in LCO.

Is a magnetic contribution of this size reasonable? We estimate k_m using the kinetic equation in 2D:[14]

$$k_m=\frac{1}{2}c_m v_m \ell_m. \qquad (3)$$

Here c_m is the magnetic specific heat and ℓ_m is the mean free path of the magnetic excitations. The velocity v_m of long-wavelength spin waves is $v_m^{SCOC}\approx 1.06\times 10^5$ m/s and $v_m^{LCO}\approx 1.16\times 10^5$ m/s, which is obtained from $v_m=\sqrt{8}SZ_cJa/\hbar$. Here Z_c is the Oguchi correction and a denotes the lattice constant.[20] Values of J for various 2D cuprates are given in Table I. Note that v_m is much larger than v_{ph}, as a result of $J \gg k_B\Theta_D$. For the specific heat c_m, we use the theoretical result shown in the inset of Fig. 2, which comes from the

TABLE I. Position T_H of the high-temperature maximum of k_m, Neel-temperature T_N, in-plane magnetic exchange coupling constant J, and the ratio J/k_BT_H for three insulating 2D cuprates. Note that J/k_BT_H is very similar in all three compounds. T_N is from Refs. 6,7,32. The values of J are derived from the two-magnon Raman scattering and infrared bimagnon-plus-phonon absorption data (Refs. 25,33), where the Oguchi correction has been taken into account.[20] The data for k in YBCO and LCO are taken from Refs. 9 and 8, respectively.

	T_H (K)	T_N (K)	J/k_B (K)	J/k_BT_H
$YBa_2Cu_3O_6$	200	>400	1125	5.6
$Sr_2CuO_2Cl_2$	245	260	1220	5.0
La_2CuO_4	285	320	1390	4.9

extrapolation of the high-temperature series for the partition sum (see the Appendix). The maximum is $c_{\max}=0.4612(5)Nk_B$ at $k_BT_{\max}=0.5956(1)J$. Given v_m, c_m, and using k_m as shown in Fig. 1, we obtain an estimate of $\ell_m(T)$, using Eq. (3). ℓ_m decreases strongly with increasing temperature (Fig. 2). At room temperature, $\ell_m/a\approx 30$ for SCOC and ≈ 75 for LCO. These values are not excessively large—much larger values of ℓ_m have been found in one-dimensional spin systems[2–4]—rendering a magnetic contribution to the heat current in SCOC and LCO very plausible.

For a better understanding of k_m, it is instructive to discuss the magnetic correlations in the quasi-2D cuprates, in particular, the in-plane magnetic correlation length $\xi_m(T)$. In a 2D antiferromagnet, long-range order with $\xi_m=\infty$ is restricted to $T=0$. With increasing temperature, spin-flips (or magnons) are excited, which reduce ξ_m by breaking the long-range correlation. In the quasi-2D materials considered here, the finite magnetic ordering temperature T_N is determined by the *inter*plane interaction,[6,7] which is much weaker than the in-plane exchange interaction J, so that $k_BT_N \ll J$ (see Table I). In the ordered state at $T<T_N$, $\xi_m=\infty$. For $T>T_N$, ξ_m is still large because of the large J. We show $\xi_m(T)$ of SCOC and LCO as inferred from magnetic neutron scattering[7] in Fig. 2. Above T_N, ξ_m is indeed much larger than the lattice constant. However, ξ_m decreases strongly with increasing temperature, approximately according to Ref. 7 $\xi_m(T)\simeq\exp(2\pi J/k_BT)$.

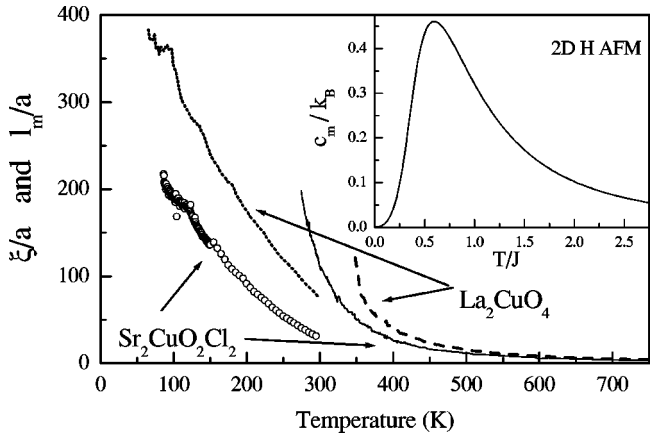

FIG. 2. Magnetic mean free path $\ell_m(T)/a$ (circles: $Sr_2CuO_2Cl_2$; dotted line: La_2CuO_4) as extracted from the data shown in Fig. 1 (see text), and in-plane magnetic correlation length $\xi_m(T)/a$ (solid line: $Sr_2CuO_2Cl_2$; dashed line: La_2CuO_4) as obtained from neutron scattering (Ref. 7). Here, $a\simeq 3.9$ Å is the lattice constant. Inset: Specific heat c_m/k_B of a $S=1/2$ square-lattice Heisenberg antiferromagnet (see the Appendix).

From these considerations we may draw following conclusions for the magnetic heat current.

(1) We expect that k_m is determined by the large in-plane exchange interaction J and not by the interplane interactions. Therefore, no significant anomaly of k_m is expected at T_N. We note that, for the same reason, the anomaly of the magnetic specific heat at T_N is very small and could not be detected experimentally.[6,24]

(2) At least above T_N, the heat-carrying magnetic excitations are not the familiar collective excitations of a magnetically ordered state (i.e., conventional magnons), but rather magnetic excitations (of triplet character) in a spin-liquid state. Note, however, that also at $T<T_N$, the nature of the magnetic excitations of the 2D cuprates is under intensive debate.[25–29]

(3) Our data analysis reveals that ℓ_m decreases strongly with temperature above 100 K. This decrease overcompensates the increase of c_m, which explains, why the maximum of k_m occurs at a temperature much lower than that of c_m. One might argue that the strong decrease of ℓ_m is related to that of ξ_m, since the mean free path of a conventional magnon should not be larger than $\xi_m(T)$. In fact, our data yield $\ell_m(T)<\xi_m(T)$, confirming such a view. We note, however, that for magnetic excitations in a spin liquid, this argument does not hold: the results on the spin-ladder compound $Sr_{14-x}Ca_xCu_{24}O_{41}$ obtained in the spin liquid state without long-range order reveal a mean free path of the magnetic excitations much larger than ξ_m.[2,4]

A double-peak structure of k has also been found in other insulating monolayer cuprates[30,34] and in the insulating bilayer compound $YBa_2Cu_3O_6$.[9] In YBCO, as in LCO, one observes pronounced anisotropy, i.e., k_c does not show a high-temperature maximum. Given the existence of a large k_m in the monolayer cuprates, the high-temperature maximum of YBCO is also likely to be of magnetic origin. We find a systematic variation of the temperature T_H of the high-temperature maximum of k_m with J for the three different insulating cuprates, as shown in Table I.

The data in Fig. 1 show that the high-temperature maximum is more pronounced in LCO than in SCOC. A related observation is the rather weak high-temperature anomaly of k in Pr_2CuO_4.[30,34] In view of the rather similar magnetic properties of the insulating cuprates, one would expect, however, a similar magnitude of k_m.[31] One may suspect that the different magnitude of k_m is related to a slightly different charge carrier doping in the samples, arising, e.g., from a variation of the oxygen content. It is well known that doping with mobile charge carriers suppresses magnetic correlations in the CuO_2 planes very effectively. Accordingly, one expects that k_m is also suppressed strongly, which is indeed found experimentally.[35] For example, in LCO and YBCO,

doped with a few percent of mobile holes per Cu, the high-temperature maximum of k completely disappears.[8–10] However, this cannot explain the larger k_m in LCO, since SCOC as a clean, undoped material should then have the largest k_m, which is not found experimentally. Note that the absolute value of k_m is not understood generally. It is, e.g., unclear why k_m is so extraordinarily large in $Sr_{14-x}Ca_xCu_{24}O_{41}$ and rather small in the Bechgaard salts.[2,3,5] It has been pointed out that the magnitude of k_m may be determined in part by the coupling between magnetic excitations and the lattice,[3] which is essential to establish a temperature gradient for the magnetic excitations.

In summary, the in-plane thermal conductivity of $Sr_2CuO_2Cl_2$ shows an unusual temperature dependence with a pronounced shoulder at high temperatures. It is unlikely that this behavior can be explained in terms of a purely phononic heat current. In particular, an unusual damping of the phononic heat current due to resonant scattering on soft lattice modes can be excluded, since there is no structural instability in $Sr_2CuO_2Cl_2$. Our data indicate a large magnetic contribution to the in-plane heat current. A comparison to other insulating layered cuprates suggests that a magnetic contribution to the heat current is an intrinsic property of these materials. The absolute magnitude of the magnetic contribution differs strongly for the various cuprates for reasons yet to be clarified.

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APPENDIX

To deduce $c(T)$ from the high-temperature series,[21] this series is converted into series for the entropy s in the energy per site e (Ref. 22). The extrapolations are stabilized by information on the ground-state energy $e_0=-0.669437(5)$ (Ref. 23), the maximum entropy $s=\ln 2$, and the expected low-energy power law $s(e)\propto(e-e_0)^{2/3}$. For the latter, Padé approximants are applied to $s'(e)/s(e)-2/(3(e-e_0))$ (Dlog Padé approximation). Very good results are obtained (reliable error estimate 10^{-2} from comparing diagonal to nondiagonal Padé approximants). The result shown in the inset of Fig. 2 (error 10^{-3}) is obtained by approximating

$$\left[\frac{(e-e_0)s'(e)}{s(e)}-\frac{2}{3}\right]\ln\left(\frac{e-e_0}{1-e_0}\right), \tag{A1}$$

which allows for multiplicative logarithmic corrections, yielding $c(T)\propto T^2[A+\ln^{-\gamma}(1/T)]$ with $A=0$ and $\gamma=1.05(5)$. We do not, however, exclude a small finite value of A as found in spin-wave theory.

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