Magnon Heat Transport in doped La$_2$CuO$_4$

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(Dated: January 14, 2002)

We present results of the thermal conductivity of La$_2$CuO$_4$ and La$_{1.8}$Eu$_{0.2}$CuO$_4$ single-crystals which represent model systems for the two-dimensional spin-1/2 Heisenberg antiferromagnet on a square lattice. We find large anisotropies of the thermal conductivity which are explained in terms of two-dimensional spin-1/2 Heisenberg antiferromagnet on a square lattice. This is consistent with previous results obtained in Ref. [9]. Several conventional mechanisms which may be intrinsically quasi-ballistic, i.e., dissipationless, thereby leading to high magnetic thermal conductivities $\kappa_{\text{mag}}$. Of particular interest is the mean free path $l_{\text{mag}}$ of the heat-current carrying magnetic excitations. Based on a kinetic approach it has been shown recently [1], that $l_{\text{mag}}$ for the spin ladder compound is of the order of 3000 Å. Direct microscopic evaluation of the mean free path is an open issue yet, with conflicting conclusions published [2].

In this letter we turn to two-dimensional (2D) quantum magnets by investigating the thermal conductivity of La$_2$CuO$_4$ which realizes the spin-1/2 Heisenberg antiferromagnet on the square lattice. The magnetic structure consists of Cu$^{2+}$-ions in CuO$_2$-planes extending along the crystallographic $a$- and $b$-directions. A strong antiferromagnetic intraplanar exchange coupling $J/k_B \approx 1550$ K [3] is present whereas the interplanar exchange $J_\perp$ is negligible ($J_\perp/J \approx 10^{-5}$) [2].

Our primary focus will be on a broad peak at high temperatures ($T$) in $\kappa(T)$ which we observe if measured by a thermal current parallel to the CuO$_2$-planes, but which is absent for the perpendicular direction. This is consistent with previous results obtained in Ref. [2]. Several attempts to explain this peak have been made, e.g. scattering processes of acoustic phonons with soft optical phonons [4] or magnons [5]. It has been speculated also, that the high-$T$-maximum could be related to heat carried by magnetic excitations [4]. However, its origin has never been elucidated unambiguously. Here, and employing the effects of doping by various types of impurities we provide clear evidence in favor of the anisotropic heat conduction to be due to 2D magnons indeed. We extract the mean free path from our data using an approach similar to that for the case of (Sr, Ca)$_{1.4}$Cu$_{24}$O$_{41}$ [1]. It will be shown that $l_{\text{mag}}$ does not only scale linearly with the reciprocal concentration of magnetic impurities which are generated by Zn-ions, but moreover, that it is roughly equal to the mean unidirectional distance between these ions.

We have measured $\kappa$ of La$_2$CuO$_4$ and La$_{1.8}$Eu$_{0.2}$CuO$_4$ single crystals as well as La$_2$Cu$_{1-z}$Zn$_z$O$_4$ ($z = 0, 0.005, 0.008, 0.01, 0.02, 0.05$) polycrystals as a function of $T$. Stoichiometric oxygen contents were achieved by annealing in high vacuum ($p < 10^{-4}$ mbar) for 1-3 hours at 800° C. The preparation of the samples [5] and the experimental method [5] have been described elsewhere.

In the lower panel of Fig. 1 we present the thermal conductivity of single crystalline La$_2$CuO$_4$ if measured parallel ($\kappa_{ab}$) and perpendicular ($\kappa_{c}$) to the CuO$_2$-planes as a function of $T$. The pronounced anisotropy of $\kappa$ strongly resembles that of spin ladder compounds [1]: $\kappa_{ab}$ exhibits a $T$-dependence which is typical for insulating crystalline materials with pure phononic thermal conductivity $\kappa_{\text{ph}}$. In contrast to this, the low-$T$ peak of $\kappa_{ab}$ is followed by a minimum around 80 K and a strong increase of $\kappa_{ab}$ which eventually develops into a broad peak around 300 K that clearly exceeds the low-$T$ peak. Several conventional mechanisms which may be invoked to explain this unusual $T$-dependence and the resulting anisotropy can be dismissed. First, we can exclude effects of radiative heat transport since the optical properties of La$_2$CuO$_4$ are almost isotropic in the relevant energy-range below $h\nu \lesssim 0.1$ eV [6]. Second, electronic contributions to the thermal current can be excluded since the material is insulating. Therefore, phononic heat transport is expected to dominate. However, and third, a scenario solely based on thermal conduction by acoustic phonons is very unlikely since an-
crease of scattering usually causes a negative slope of $\kappa(T)$ for intermediate and higher $T$, in contrast to our observation. Hence, either an unusual scattering process acts on the acoustic phonons or an additional contribution apart from the conventional phononic background must be present for $\kappa_{ab}$.

We now turn to the upper panel of Fig. 1 where $\kappa_{ab}(T)$ and $\kappa_c(T)$ of La$_{1.8}$Eu$_{0.2}$CuO$_4$ are shown. Both curves strongly resemble our findings for La$_2$CuO$_4$, yet exhibiting obvious differences: the low-$T$-peaks of $\kappa_{ab}$ and $\kappa_c$ are slightly larger and more sharply shaped than in the case of La$_2$CuO$_4$. Furthermore, a step-like anomaly is present at $T_{LT} \approx 135$ K. Above $T_{LT}$ $\kappa_c$ remains almost constant and stays below the value for the undoped case, while $\kappa_{ab}$ also exhibits a high-$T$ maximum at $T \approx 270$ K which is even larger than in La$_2$CuO$_4$.

The difference between $\kappa_c$ of Eu-doped and of pure La$_2$CuO$_4$ is attributed to a difference in phononic heat conduction. Upon doping La$_2$CuO$_4$ with Eu, enhanced scattering of phonons reduces $\kappa_c$ for $T > T_{LT}$, where both compounds have the same structure. The anomaly at $T_{LT}$ (La$_{1.8}$Eu$_{0.2}$CuO$_4$) signals the transition to a new structural phase for $T < T_{LT}$ where $\kappa_{ph}$ is enhanced. The structural peculiarities of rare earth doped cuprates are well known [14]. Their influence on $\kappa_{ph}$ will be discussed in detail in a forthcoming paper [14].

Despite the increase in scattering of phonons due to Eu-impurities in La$_{1.8}$Eu$_{0.2}$CuO$_4$ the high-temperature peak of $\kappa_{ab}$ is of larger magnitude than in La$_2$CuO$_4$. This excludes the peak to stem from the previously mentioned heat transport by acoustic phonons. The high-$T$ peaks in $\kappa_{ab}$ of both compounds therefore must originate from a different heat transport channel. In principle, two different kinds of excitation could generate such an additional heat current. One possibility is heat transport by dispersive optical phonons, the other one is thermal current carried by magnetic excitations. Magnetic heat transport is the most likely explanation, since the magnetism is truly two-dimensional and the refoc could explain the observed anisotropy. This is not true for dispersive optical phonons, which certainly are present in this material, but are found along all crystal axes [13].

This result is corroborated by the strong suppression of the high-$T$ peak in hole-doped La$_{2-x}$Sr$_x$CuO$_4$ [9], even at a very low Sr-content of $x \approx 0.01$ [10]. On the one hand such an effect of doping is not to be expected in the case of a high-$T$ peak originating from optical phonons, since the lattice impurities induced by the Sr-ions are similar to the Eu-impurities discussed above. On the other hand the strong frustration of antiferromagnetism upon doping of mobile holes (cf. Ref. [12] and references therein) would provide for a satisfying explanation for the suppression of a peak of magnetic origin. Note in this context that Eu-doping leaves the CuO$_2$-planes and therefore the magnetism almost unaffected. Hence we conclude, that the high-$T$ peak of $\kappa_{ab}$ originates from magnetic excitations which propagate only within the CuO$_2$-planes. $\kappa_{ab}$ therefore consists of a usual phonon background and a magnon contribution $\kappa_{mag}$ while $\kappa_c$ is purely phononic.

Upon applying a magnetic field of 8 T no significant changes of $\kappa$ were detected. This is, however, consistent with a magnetic origin of the high-$T$ peak since the corresponding Zeeman-energy is orders of magnitude smaller than the magnetic exchange coupling $J$.

In order to extract $\kappa_{mag}$ from $\kappa_{ab}$ we make use of the anisotropy of $\kappa$ assuming that the phononic part $\kappa_{ab,ph}$ of $\kappa_{ab}$ is roughly proportional to $\kappa_c$ which is justified by only weakly anisotropic elastic constants [13]. Since the magnetic contributions roughly follow a $T^2$-law (see below) they are expected to be negligible in the range of the low-$T$ peak. Therefore, for La$_2$CuO$_4$ as well as for La$_{1.8}$Eu$_{0.2}$CuO$_4$ a reasonable estimate of $\kappa_{ab,ph}$ is achieved by scaling the corresponding data for $\kappa_c$, such as to match its low-$T$ peaks with that of $\kappa_{ab}$. In Fig. 1 the data thus obtained for $\kappa_{ab,ph}$ of both compounds are represented by dashed lines. $\kappa_{mag}$ was extracted by subtracting $\kappa_{ab,ph}$ from $\kappa_{ab}$ (open squares in Fig. 1). This procedure involves significant uncertainties only at low $T$ where $\kappa_{mag} \ll \kappa_{ab,ph}$. At intermediate and higher $T$ the relative uncertainties are smaller and rather concern the magnitude of $\kappa_{ab,ph}$ than its slope. Therefore we assume that in this $T$-range the difference between the extracted and the true $\kappa_{mag}$ is roughly constant.

Following standard linearized Boltzmann-theory we have \( \kappa = \frac{1}{2} \frac{1}{(2\pi)^3} \int v_k l_k c_k \frac{d}{dE} n_k dk \) for the 2D thermal conductivity of a single magnon dispersion branch (la-
belled by $i$, where $v_k, l_k, \epsilon_k$ and $n_k$ denote velocity, mean free path, energy and Bose-function of a magnon. Note that $\kappa_{mag}^i$ of a three-dimensional ensemble of planes, as realized in La$_2$CuO$_4$, is given by $\kappa_{mag}^i = \frac{2v_k^2}{\kappa_{ph}^i}$, where $c = 13.2$ Å is the lattice constant of La$_2$CuO$_4$ perpendicular to the planes. Then the total $\kappa_{mag}$ results from summing up $\kappa_{mag}^i$ of each magnon branch.

In order to calculate $\kappa_{mag}^i$ we approximate the magnon dispersion relation $\epsilon_k$ of the two branches $i = 1, 2$ with the 2D-isotropic expression $\epsilon_k = \epsilon_k = \sqrt{\Delta_i^2 + (\hbar v_0 k)^2}$, which describes the dispersion observed experimentally [17, 18] for small values of $k$. Here, $v_0$ is the spin wave velocity while $\Delta_1$ and $\Delta_2$ denote the spin gaps of each magnon branch. For clarity, we define the characteristic temperature $\Theta_M = (\hbar v_0 \sqrt{\pi})/(a k_B)$ where $a = 3.8$ Å is the lattice constant of the CuO$_2$-planes [19]. Assuming a momentum independent mean free path, i.e. $l_k \equiv l_{mag}$, we obtain for each magnon branch:

$$\kappa_{mag}^i = \frac{v_0 k_B l_{mag} T^2}{2a^2 c} \Theta_M^{-1} \int_{x_{0,i}}^{x_{max}} x^2 \sqrt{x^2 - x_{0,i}^2} \frac{dn(x)}{dx} dx \cdot (1)$$

Here, the integral is dimensionless but temperature dependent via $x_{0,i} = \Delta_i/(k_B T)$ and $x_{max}$. The upper boundary $x_{max}$, however, may be set to infinity without affecting the fit at temperatures $T \lesssim 300$ K. Since from neutron scattering experiments $v_0 \approx 1.287 \cdot 10^5$ m/s [5] as well as $\Delta_1/k_B \approx 26$ K and $\Delta_2/k_B \approx 58$ K [17] are well known quantities, $l_{mag}$ is the only unknown parameter in Eq. 1. Assuming $l_{mag}$ to be $T$-independent at low $T$, we can use Eq. 1 in order to check the $T$-dependence of our experimental $\kappa_{mag}$, which for $T \gtrsim \Delta_{1,2}$ should roughly be $\kappa_{mag} \propto T^2$, as the integral is only weakly $T$-dependent in this $T$-range. Furthermore, $l_{mag}$ can be extracted from the data. For $T \gtrsim 250$ K interactions between the magnons become important [20] which requires a renormalization of the spin wave parameters and therefore we restrict the application of Eq. 1 with constant $l_{mag}$ to $T \lesssim 250$ K.

### Table I: Fit parameter of $\kappa_{mag}$ for single (S) and poly-crystals (P). The relative errors for $l_{mag}$ of the poly-crystals amount to about 10%. In all cases the errors have been obtained using different estimates for $\kappa_{ph}$.

| Sample                        | $l_{mag}$ (Å) | Fit interval (K) |
|-------------------------------|---------------|------------------|
| S La$_2$CuO$_4$              | 558 ± 140     | 70–158           |
| S La$_{1-x}$Eu$_x$CuO$_4$    | 1157 ± 60     | 54–131           |
| P La$_2$CuO$_4$              | 815           | 70–140           |
| P La$_2$CuO$_{0.995}$Zn$_{0.005}$O$_4$ | 515     | 70–158           |
| P La$_2$CuO$_{0.992}$Zn$_{0.008}$O$_4$ | 435     | 82–164           |
| P La$_2$CuO$_{0.997}$Zn$_{0.003}$O$_4$ | 266     | 100–173           |
| P La$_2$CuO$_{0.995}$Zn$_{0.005}$O$_4$ | 153     | 118–200           |
| P La$_2$CuO$_{0.992}$Zn$_{0.007}$O$_4$ | 49      | 141–223           |

FIG. 2: Open circles: Thermal conductivity $\kappa$ of La$_2$Cu$_{1-x}$Zn$_x$O$_4$ poly-crystals ($z = 0, 0.005, 0.01, 0.05$) as a function of $T$. Solid lines: extrapolated $\kappa_{ph}$.

Fitting Eq. 1 to the experimental data we allow for an additive shift of the $\kappa_{mag}$-curve which yields a further free parameter apart from $l_{mag}$ and accounts for the aforementioned uncertainties in the magnitude of $\kappa_{mag}$ [21]. For both compounds satisfactory fits were obtained at intermediate $T$-ranges. These are listed in Table I together with the resulting values for $l_{mag}$. The fitting curves are represented by solid lines in Fig. 1. While the slight deviations between the fitted and experimental data towards low $T$ are due to the uncertainties in $\kappa_{ab,ph}$ in this range, the deviations at high $T$ can be understood in terms of the $T$-dependence of $l_{mag}$ due to enhanced magnon-magnon scattering.

The data are consistent with a constant $l_{mag}$ for $T$ both, within the fit interval and below, indicating that in this range $T$-dependent scattering processes like magnon-magnon scattering or even magnon-phonon scattering may be discarded. Therefore, relevant processes seem to be sample-boundary scattering or scattering at static magnetic defects. For La$_{1-x}$Eu$_x$CuO$_4$ and La$_2$CuO$_4$ we find $l_{mag} \approx 1160$ Å and $l_{mag} \approx 560$ Å, respectively. Since these values are far too small to correspond to the crystal dimensions, magnon-defect scattering is the most likely candidate. This conclusion is consistent with the fact that $\kappa_{mag}$ and $l_{mag}$ are quantitatively different for La$_{1-x}$Eu$_x$CuO$_4$ and La$_2$CuO$_4$: since the magnetic properties of both compounds are expected to be identical in essence, unequal $\kappa_{mag}$ can arise only due to a difference in densities of the magnetic defects that restrict $l_{mag}$.

In order to check our analysis quantitatively it would be desirable to measure the density of static magnetic defects of our crystals. In lack of such methods we have performed measurements of $\kappa$ on samples with a well defined density of magnetic defects. Such defects can be induced in La$_2$CuO$_4$ by substituting a small amount of...
Cu$^{2+}$-ions by non-magnetic Zn$^{2+}$-ions. Representative results on our polycrystalline samples of La$_2$Cu$_{1-z}$Zn$_{z}$O$_4$ are presented in Fig. 4. As is expected for doping of static structural and magnetic defects, the Zn-impurities lead to a gradual suppression of both, the phonon as well as the magnon contribution to $\kappa$.[23] Due to the polycrystalline nature of our samples anisotropic information on $\kappa$ is averaged over. We therefore estimate the phonon contributions $\kappa_{\text{ph}}$ by fitting a factor of its maximum by $\kappa_{\text{ph}} = \alpha / T + \beta$ and by extrapolating this fit towards high $T$ (solid lines in Fig. 4). In turn, $\kappa_{\text{mag}}$ on the polycrystals is obtained by $\kappa_{\text{mag}} = \kappa - \kappa_{\text{ph}}$.

Note, that the measured $\kappa_{\text{mag}}$ is smaller than the intrinsic $\kappa_{\text{mag}}$ of these compounds by the factor of 2/3 due to averaging over all three components of the $\kappa$ tensor.[24] As shown in Fig. 4 $\kappa_{\text{mag}} = (3/2)\kappa_{\text{mag}}$ systematically decreases with increasing Zn-content as it is expected for static defects. Analyzing these data we proceed analogous to the undoped case by using Eq. 1. However, we neglect a slight reduction of the wave velocity and changes of the spin gaps induced by the Zn-ions. These effects lead to corrections smaller than the experimental error. The fits are represented by the solid lines in Fig. 4 the resulting $l_{\text{mag}}$ and the fit intervals are reproduced in Table 1. For typical transport experiments it is expected that the mean free path of the quasi-particles is proportional to the reciprocal defect concentration, i.e., to $1 / z$. We therefore plot $l_{\text{mag}}$ as a function of $1 / z$ in the inset of Fig. 4. The linear scaling between $l_{\text{mag}}$ and $1 / z$ is clearly confirmed. From a linear fit including the origin (solid line) we find that $l_{\text{mag}} \approx 0.74 a / z$. Since $1 / z$ is equal to the mean unidirectional distance of Zn-ions, i.e. static magnetic defects, $l_{\text{mag}}$ gives a direct measure of these distances.[24]. Therefore, this result confirms the above quantitative analysis of $\kappa_{\text{mag}}$ of the single crystals based on Eq. 4.

In conclusion, both, qualitatively as well as quantitatively our results strongly suggest, that the high-$T$ peak observed in $\kappa_{\text{ab}}$ of doped and undoped La$_2$CuO$_4$ arises due to magnon heat transport which is confined to the CuO$_2$-planes. We note, that in general a large $\kappa_{\text{mag}}$ is rarely observed at high $T$ and requires a unique synergy of magnon-phonon coupling[27], large $l_{\text{mag}}$ and high spin wave velocity $v_0$. Its realization in La$_2$CuO$_4$ may lead to future use of magnetic heat transport as a tool to study the interactions of magnetic excitations with other quasiparticles like holes or phonons.

We acknowledge support by the DFG through SP1073.

![FIG. 3: Main panel: Magnon thermal conductivity $\kappa_{\text{mag}}$ of La$_2$Cu$_{1-z}$Zn$_{z}$O$_4$ ($z = 0, 0.005, 0.008, 0.01, 0.02, 0.05$) as a function of $T$ (open circles). Solid lines: fits according to Eq. 4. Inset: $l_{\text{mag}}$ as a function of $1 / z$ in units of lattice constants $a$. Solid line: fit line through origin.](image)

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