# Temperature- and magnetic-field-dependent thermal conductivity of pure and Zn-doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals

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The thermal conductivity $\kappa$ of $Bi_2Sr_2CaCu_2O_{8+\delta}$ is measured in pure and Zn-doped crystals as a function of temperature and magnetic field. The in-plane resistivity is also measured on the identical samples. Using these data, we make a crude estimate of the impurity-scattering rate $\Gamma$ of the pure and the Zn-doped crystals. Our measurement show that the ''plateau'' in the $\kappa(H)$ profile is not observed in the majority of our Bi-2212 crystals, including one of the cleanest crystals available to date. The estimated values of $\Gamma$ for the pure and Zn-doped samples allow us to compare the $\kappa(H)$ data with the existing theories of the quasiparticle heat transport in *d*-wave superconductors under magnetic field. Our analysis indicates that a proper inclusion of the quasiparticle-vortex scattering, which is expected to play the key role in the peculiar behavior of the $\kappa(H)$, is important for a quantitative understanding of the quasiparticle heat transport in the presence of the vortices.

Thermal conductivity $\kappa$ of a superconductor is one of the few probes which allow us to investigate the quasiparticle (QP) density and its scattering rate in the superconducting (SC) state. It is now believed that the SC state of the high-$T_c$ cuprates is primarily $d_{x^2-y^2}$,[1] where it has been found that the magnetic field induces extended QP's whose population increases as $\sqrt{H}$.[2,3] Also, the QP scattering rate in the cuprates has been found to drop very rapidly below $T_c$,[4] which causes a pronounced peak in the temperature dependence of $\kappa$.[5] In 1997, Krishana *et al.* reported an intriguing result from their measurement on the magnetic-field dependence of $\kappa$ in $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) at temperatures below 20 K.[6] They observed, in the profile of $\kappa(H)$, a sharp break at a '' transition field'' $H_k$ and a subsequent plateau region where $\kappa$ does not change with magnetic field. Krishana *et al.* proposed an interpretation that $H_k$ marks a phase transition from the $d_{x^2-y^2}$ state to a fully gapped $d_{x^2-y^2}+id_{xy}$ (or $d_{x^2-y^2}+is$) state and thus in the plateau region there are little thermally excited QPs which contribute to the heat transport. This interpretation appears to be fundamentally related to the high-$T_c$ mechanism and therefore attracted much attention both from theorists[7–14] and from experimentalists.[15–18]

An independent test of this unusual behavior of $\kappa$ has been reported by Aubin *et al.*;[15] although the plateaulike feature was essentially reproduced in their measurement, when the data were taken with field sweep up and down, Aubin *et al.* observed a rather large jump in $\kappa$ upon field reversal and consequently the $\kappa(H)$ profile had a pronounced hysteresis. The fact that the $\kappa$ value in the ''plateau'' depends on the history of the applied magnetic field casts a serious doubt on the Krishana *et al.*'s interpretation. Moreover, Aubin *et al.* reported that an *increase* in $\kappa$ with magnetic field was observed at subkelvin temperatures, which strongly suggests the presence of a finite density of QP's at low temperatures and thus is incompatible with the fully gapped state.[16] Although these newer results suggest that a novel phase transition in the gap symmetry is not likely to be taking place, the plateau in the $\kappa(H)$ profile and the sensitiveness of $\kappa$ on the magnetic-field history are still to be understood. One interesting information one can draw from these experiments is that phonons are *not* scattered by vortices in cuprate superconductors.[6,15,19]

Motivated by these experiments, there appeared several theories that try to capture the essential physics of the QP heat transport in the *d*-wave superconducting state. It has become rather clear that a proper inclusion of the QP-vortex scattering rate is necessary for explaining the observed magnetic-field dependence; however, there has been no consensus yet as to *how* the QP-vortex scattering is taken into account. To improve our understanding of the QP heat transport under magnetic field, quantitative examinations of various theories in the light of actual data are indispensable. For Bi-2212, however, previously published data from various groups do not provide enough information; for example, the impurity scattering rate, which is the most important parameter that controls the thermal conductivity behavior, are not known for clean Bi-2212 crystals. [Rather surprisingly, no electrical resistivity data have been supplied for samples which were used in recent studies of $\kappa(H)$ profile of Bi-2212.[6,15,16,18]]

In this paper, we present results of our measurements of $\kappa(T)$ and $\kappa(H)$ of well-characterized Bi-2212 crystals, together with their in-plane resistivity ($\rho_{ab}$) data. To look for the effect of changing impurity-scattering rate, we measured both pure and 0.6%-Zn-doped crystals. The crystals used here are single domained (without any mosaic structure nor grain boundaries) and have very good morphology, which were confirmed by x-ray analyses and the polarized-light optical microscope analysis. In the data presented here, neither the pure sample nor the Zn-doped sample show any plateaus in the $\kappa(H)$ profile in the temperature and the magnetic-field regime where the plateaus have been reported. In fact, we have found that the plateau in the $\kappa(H)$ profile is not a very reproducible feature (we have thus far observed the plateau-like feature in only two samples out of more than 30 samples measured) and we have not yet conclusively sorted out what determines the occurrence of the plateau;[20] we therefore decided to show only the data that are representative of the majority of the samples. Using all the data of $\rho_{ab}(T)$, $\kappa(T)$, and $\kappa(H)$, we try to estimate the electronic thermal conductivity $\kappa_e(T)$ and make a rough estimate of the impurity scattering rate $\Gamma$ for the pure and Zn-doped samples. Our data offer a starting point for the quantitative understanding of the QP heat transport in the *d*-wave superconductors in the magnetic fields, where an interplay between the QP-vortex scattering and the QP-impurity scattering apparently plays an important role.

The single crystals of Bi-2212 are grown with a floating-zone method and are carefully annealed and quenched to obtain a uniform oxygen content.[21] Both the pure and the Zn-doped crystals $[Bi_2Sr_2Ca(Cu_{1-z}Zn_z)_2O_{8+\delta}]$ are tuned to the optimum doping by annealing at 750 °C for 48 h, after which the transition width of about 1.5 K (measured by the dc magnetic susceptibility measurements) is achieved; this indicates a high homogeneity of these crystals. The Zn-doped crystal contains $(0.6\pm0.1)\%$ of Zn (namely $z=0.006\pm0.001$), which was determined by the inductively coupled plasma spectrometry. The zero-resistance $T_c$'s are 92.4 K for the pure crystal and 84.5 K for the Zn-doped crystal. We first measure the temperature dependence of $\rho_{ab}$ using a standard four-probe technique, and then measure the thermal conductivity $\kappa$ of the same sample. The temperature dependence of $\kappa$ from 1.6 to 160 K are measured in zero field using calibrated AuFe-Chromel thermocouples. The precise magnetic-field dependence of $\kappa$ is measured with a steady-state technique using a small home-made thin-film heater and microchip Cernox thermometers. The bottom end of the sample is anchored to a copper block whose temperature is carefully controlled within 0.01% stability and accuracy. Since we need to measure the change in $\kappa$ with a very high accuracy, the small magnetic-field dependence of the thermometers were carefully calibrated beforehand using a $SrTiO_3$ capacitance sensor and a high-resolution capacitance bridge. As a check for the measurement system and the calibrations, we first measured the thermal conductivity of nylon and confirmed that the reading is indeed magnetic-field independent within 0.1% accuracy.

Figure 1(a) shows the $\rho_{ab}(T)$ data of the pure and Zn-doped samples. If we define the residual resistivity $\rho_0$ by the extrapolation of the *T*-linear resistivity to 0 K, we obtain slightly negative $\rho_0$ for the pure sample; this is always the case for the cleanest Bi-2212 crystals grown in our laboratory. As is expected, the Zn-doped sample gives larger $\rho_0$, which is about 10 $\mu\Omega$ cm. We note that the uncertainty in the absolute magnitude of $\rho_{ab}$ and $\kappa$ in our measurements are less than $\pm5\%$ in this work. This is achieved by determining the sample thickness, which is usually the main source of the uncertainty, by measuring the weight of the sample with 0.1 $\mu$g resolution. We used relatively long samples here (the length of the pure and the Zn-doped samples were 4.5 and 6.5 mm, respectively), with which the errors in estimating the voltage contact separation and the thermocouples separation are less than $\pm5\%$. It should also be noted that a sophisticated technique to make the current contacts uniformly on the side faces of the crystals are crucial for reliably measuring $\rho_{ab}$ of Bi-based cuprates.

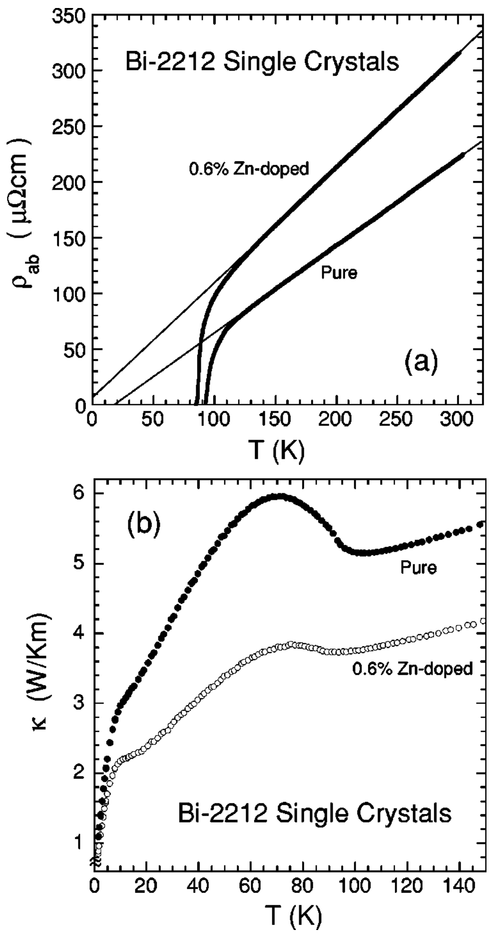


FIG. 1. $T$ dependence of (a) $\rho_{ab}$ and (b) $\kappa$ of pure and 0.6%-Zn-doped Bi-2212 crystals in zero field. The thin solid lines are *T*-linear fits to the $\rho_{ab}(T)$ data.

Figure 1(b) shows the temperature dependence of $\kappa$ of the two samples in zero field. The size of the peak in $\kappa(T)$ of the pure crystal is very large for Bi-2212 (the enhancement from the minimum near $T_c$ to the peak is 16%). This implies that the pure crystal reported here is one of the cleanest Bi-2212 crystals available to date. The Zn-doped sample shows not only a smaller magnitude of $\kappa$ but also a significantly suppressed peak in $\kappa(T)$ below $T_c$; this is caused by a larger impurity scattering rate (which limits the enhancement of the QP mean free length below $T_c$) and is consistent with the Zn-doping effect in $YBa_2Cu_3O_{7-\delta}$ (YBCO).[22,23]

Figure 2 shows $\kappa(H)$ profiles of the two samples at 7.5 and 12.5 K. The data are normalized by the zero-field value of $\kappa$. Neither of these samples show any plateaulike feature below 6 T. [The $\kappa(H)$ data of the Zn-doped sample is almost flat within our sensitivity, but we do not call it a plateau.] Note that Ref. 6 reported the plateaus to be observed above 0.9 T at 7.5 K and above 2.6 T at 12.5 K, which are well within the range of our experiment. The data in Fig. 2 were taken in the field-cooled (FC) procedure, in which the sample is cooled in the presence of a magnetic field, and therefore the magnetic induction in the sample remains homogeneous. The FC data are expected to give information

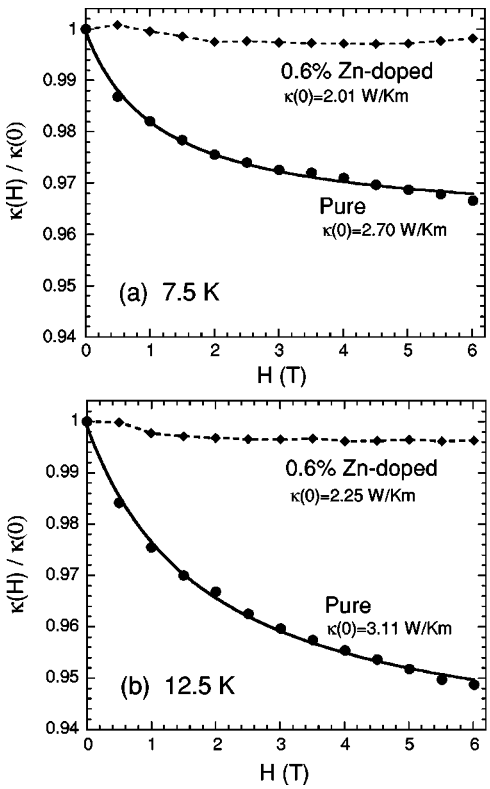


FIG. 2. Normalized $\kappa(H)$ profiles of the pure and Zn-doped samples at (a) 7.5 K and (b) 12.5 K. The solid lines are the fits to Eq. (1) with: (a) $\kappa_{ph}=2.60$ W/K m, $\kappa_e=0.10$ W/K m, and $p=0.88$ for the 7.5 K data, and (b) $\kappa_{ph}=2.91$ W/K m, $\kappa_e=0.20$ W/K m, and $p=0.53$ for the 12.5 K data.

which is free from complications due to vortex pinning, while the zero-field-cooled (ZFC) data (for which the sample is cooled in zero field and the magnetic field is swept at a constant temperature) are subject to such complications. We emphasize that the FC data should be examined to look for the intrinsic effect of vortices on the QP transport. All the previously published data on the plateau[6,15,16,18] are, however, ZFC data.[24]

It has been proposed that the magnetic-field dependence of $\kappa$ of the high-$T_c$ cuprates are described by

$$\kappa(H,T)=\frac{\kappa_e(T)}{1+p(T)|H|}+\kappa_{ph}(T), \quad (1)$$

where $\kappa_e$ is the electronic part of $\kappa$ in zero field and $\kappa_{ph}$ is the phonon part.[26] Equation (1) was proposed first by Yu *et al.*[27] and later by Ong and co-workers.[5,19,25] This expression utilizes the finding that the phonon thermal conductivity of the cuprates is independent of the magnetic field. The parameter $p(T)$ is proportional to the zero-field value of the QP mean free path. We found that the $\kappa(H)$ data of our clean samples are reasonably well described by Eq. (1). The solid lines in Figs. 2(a) and 2(b) are fits of the data to Eq. (1). The fitting parameters suggest that the phononic contribution to $\kappa$ is as large as 96% and 93% at 7.5 and 12.5 K, respectively; this is essentially a reflection of the fact that the changes in $\kappa$ with the magnetic field are very small at these temperatures. Ong *et al.* reported[19] that $p(T)$ is about 2.3 $T^{-1}$ for underdoped YBCO at 7.5 K, while it is 0.88 $T^{-1}$ for our pure Bi-2212 at 7.5 K. This is an indication that the QP transport is dirtier in Bi-2212 compared to YBCO; we will elaborate on these $p(T)$ values later. The magnetic field dependence of $\kappa$ of the Zn-doped sample is too small to make a reliable fit to Eq. (1), but one can infer that the ratio of the phononic contribution in the Zn-doped sample is even larger than in the pure sample.

In the normal state, the electronic thermal conductivity $\kappa_e$ and the electrical conductivity $\sigma$ are related by $\kappa_e/T=L\sigma$, where $L$ is called the Lorenz number. In simple metals, $L$ is usually constant at high temperatures (Wiedemann-Franz law) and the free-electron model gives $L_0=2.44\times10^{-8}$ W Ω/K$^2$. When the electron-electron correlation becomes strong, $L$ becomes smaller than the free electron value $L_0$; for YBCO, $L$ has been estimated to be around $1.0\times10^{-8}$ W Ω/K$^2$ near $T_c$.[22,28] Using this value of $L$ and the $\rho_{ab}$ data of our crystals, we can roughly estimate $\kappa_e$ and $\kappa_{ph}$ above $T_c$. Such estimation for the pure sample gives $\kappa_e$ and $\kappa_{ph}$ values of about 1.6 and 3.6 W/K m, respectively, at 120 K. (120 K is the lower bound for the temperature range where the effect of the superconducting fluctuations on $\rho_{ab}$ is negligible.) It has already been established that $\kappa_{ph}$ does not change much just below $T_c$ and that it is mostly $\kappa_e$ that causes the peak.[5,29] We can therefore infer (from the $\kappa_e$ value at 120 K and the total $\kappa$ at the peak) that $\kappa_e$ at the peak is about 1.7 times larger than in the normal state. In the same manner, we can estimate for the Zn-doped sample $\kappa_e$ and $\kappa_{ph}$ to be about 1.0 and 2.9 W/K m at 120 K, respectively, and the enhancement of $\kappa_e$ at the peak is inferred to be a factor of 1.2. This analysis indicates that the effect of 0.6%-Zn doping is weaker for $\kappa_{ph}$ (which is decreased by 19% at 120 K upon the 0.6%-Zn doping) than for $\kappa_e$ (which is decreased by 38% at 120 K).

Comparison of the inferred behaviors of $\kappa_e$ below $T_c$ with the theoretical calculations of Ref. 22 gives us the idea of the magnitude of the impurity-scattering rate $\Gamma$ in our samples. As is discussed above, $\kappa_e$ can be inferred to be about 1.7 times enhanced at the peak; comparison of this enhancement factor with the calculations for various $\Gamma/T_c$ values[22] suggests that $\Gamma/T_c$ of our pure sample is about 0.05. The position of the peak (at $T/T_c\sim0.7$) is also consistent with the theoretical calculation for $\Gamma/T_c\sim0.05$. Similarly, the inferred enhancement factor of $\kappa_e(T)$ of the Zn-doped sample suggests $\Gamma/T_c$ to be about 0.2. Although these estimates are very crude, the estimated values of $\Gamma/T_c$ of our Bi-2212 samples imply that the scattering rate increases by $\Gamma/T_c\sim0.25$ per 1% of Zn, which is of the same order of magnitude as that for YBCO.[22,30] The estimated $\Gamma/T_c$ of our pure Bi-2212 is still notably larger than that for pure YBCO, for which $\Gamma/T_c$ has been estimated to be $\sim0.01$.[22,30] This is essentially a reflection of the fact that the peak in $\kappa(T)$ of Bi-2212 is much smaller compared to that of YBCO, and is probably caused by the intrinsic disorder of the crystalline lattice of Bi-2212 (the modulation structure along the $b$ axis).

The above-mentioned difference in $\Gamma/T_c$ between our pure Bi-2212 ($\Gamma/T_c\sim0.05$) and pure YBCO ($\Gamma/T_c\sim0.01$) indicates that the QP mean free path in zero field, $l_0$, is roughly five times longer in pure YBCO compared to that in pure Bi-2212. This observation yields useful information on the QP scattering cross section of the vortices, $\sigma_{tr}$, in Bi-

2212. It was discussed in Refs. 19 and 25 that parameter $p(T)$ appearing in Eq. (1) can be expressed as $p(T) = l_0\sigma_{tr}/\phi_0$. (Note that $\sigma_{tr}$ is the scattering cross section in two dimensions.) As is already discussed, the $p(T)$ value at 7.5 K is 0.88 $T^{-1}$ for our pure Bi-2212, while it is 2.3 $T^{-1}$ for YBCO; given the indication that $l_0$ is roughly five times longer in YBCO, the ratio of the $p(T)$ values of the two systems suggests that $\sigma_{tr}$ should be approximately a factor of 2 larger in Bi-2212. Since $\sigma_{tr}$ has been estimated to be 9 nm for YBCO,[19] we obtain $\sigma_{tr}\sim 18$ nm for Bi-2212. The origin of this difference in $\sigma_{tr}$ between the two systems might be related to the difference in the structure of the vortex lines and should be a subject of future studies. In any case, the estimate of $\sigma_{tr}\sim 18$ nm gives $l_0\sim 0.1$ $\mu$m for pure Bi-2212 at 7.5 K.

Now let us discuss the observed magnetic-field dependence of $\kappa$ in conjunction with the estimated values of $\Gamma/T_c$ for each sample. Kübert and Hirschfeld (KH) calculated the magnetic-field dependence of $\kappa_e$ that comes from the QP's Doppler shift around the vortex.[31] Good agreements between the KH theory and experiments have been reported for very low temperatures, where an increase of $\kappa$ with magnetic field has been observed.[16,23,32] The numerical calculation by KH show that $\kappa(H)$ is already an increasing function of $H$ at $T = 0.2T_c$ for a dirty case, $\Gamma/T_c = 0.1$; this is clearly in disagreement with our data and indicates the necessity of an inclusion of the QP-vortex scattering into the calculation.

The theory proposed by Franz[13] tries to incorporate the effect of QP-vortex scattering; heuristically, Franz supposed that the QP-impurity scattering and the QP-vortex scattering are separable and additive. In this theory, the zero-field scattering rate $\sigma_0$ (which is a sum of the impurity scattering rate and the inelastic scattering rate) is directly related to the total change in $\kappa$ with the magnetic field, $\Delta\kappa$, which is expressed as $\Delta\kappa = (2.58T/\sigma_0 - 1)\kappa_{00}$ for $\sigma_0 < T$, and as $\Delta\kappa = (2.15T/\sigma_0)^2\kappa_{00}$ for $\sigma_0 > T$. In these expressions, $\kappa_{00}$ is the universal thermal conductivity.[30,33] Assuming that the inelastic scattering is negligible ($\sigma_0 \approx \Gamma$) at 7.5 K, we can estimate $\Delta\kappa$ for the pure and Zn-doped samples using these equations. The results are $\Delta\kappa \approx 0.4$ W/K m for the pure sample and $\Delta\kappa \approx 0.1$ W/K m for the 0.6%-Zn-doped sample. (In the calculation, we used $\kappa_{00}/T = 0.015$ W/K$^2$ m which is reported for Bi-2212.[32].) When we turn to our data, the values of $\Delta\kappa$ actually measured in our experiments are much smaller; at 7.5 K, $\Delta\kappa$ up to 6 T is 0.086 and 0.008 W/K m for the pure and the Zn-doped samples, respectively. Clearly, the theory overestimates $\Delta\kappa$ and the overestimation is particularly large for the Zn-doped sample. This comparison implies that the rather simple treatment of the separable QP-impurity scattering and the QP-vortex scattering is not accurate enough, particularly when the impurity scattering rate becomes comparable to the vortex scattering rate.

Vekhter and Houghton (VH) (Ref. 14) have recently proposed a theory which explicitly considers the interplay between the vortex-lattice scattering and the impurity scattering. Unfortunately, numerical calculations for the magnetic-field dependence of $\kappa$ at intermediate temperatures are available only for a very clean case ($\Gamma/T_c = 0.006$),[14] and we cannot make a direct comparison to our data. Qualitatively, however, the theory predicts that a steep drop in $\kappa(H)$ at low fields is expected only when $T$ is larger than $\Gamma$. The estimated $\Gamma/T_c$ values of our samples suggest that the condition $T > \Gamma$ is satisfied in the pure sample, but is not satisfied in the 0.6%-Zn-doped sample in the temperature range of Fig. 2.

In summary, the temperature and magnetic-field dependences of the thermal conductivity $\kappa$ are measured in pure and Zn-doped Bi-2212 crystals that are well characterized. The $\rho_{ab}(T)$ data taken on the identical samples are used for extracting the electronic thermal conductivity $\kappa_e$ above $T_c$. The temperature and magnetic-field dependences of $\kappa$ clearly reflect the difference in the impurity-scattering rate $\Gamma$ in the crystals. It is found that in the majority of our Bi-2212 crystals (including one of the cleanest crystals available to date) the ''plateau'' in the $\kappa(H)$ profile is not observed and the $\kappa(H)$ profile around 10 K are reasonably well described by Eq. (1), which was originally proposed for YBCO. We estimate $\Gamma$ of our crystals by comparing the behaviors of $\kappa_e$ below $T_c$ with the calculation by Hirschfeld and Putikka.[22] The estimated values of $\Gamma$ for the pure and Zn-doped samples allow us to compare the $\kappa(H)$ data with the existing theories of the QP heat transport in *d*-wave superconductors under magnetic field.

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