Electronic Inhomogeneity and Breakdown of the Universal Thermal Conductivity in Cuprate Superconductors

X. F. Sun, S. Ono, Yasushi Abe, Seiki Komiya, Kouji Segawa, and Yoichi Ando

Central Research Institute of Electric Power Industry, Komae, Tokyo 201-8511, Japan

(Dated: March 23, 2022)

We report systematic, high-precision measurements of the low-T (down to 70 mK) thermal conductivity \( \kappa \) of YBa\(_2\)Cu\(_3\)O\(_x\), Lax\(_{2-\delta}\)Sr\(_x\)CuO\(_4\) and Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{8+\delta}\). No careful examinations of the Zn- and hole-doping dependences of the residual thermal conductivity \( \kappa_0/T \), as well as the in-plane anisotropy of \( \kappa_0/T \) in Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{8+\delta}\), indicate a breakdown of the universal thermal conductivity, a notable theoretical prediction for \( d \)-wave superconductors. Our results point to an important role of electronic inhomogeneities, that are not considered in the standard perturbation theory for thermal conductivity, in the under- to optimally-doped regime.

PACS numbers: 74.25.Fy, 74.25.Dw, 74.72.Bk, 74.72.bk, 74.72.Dn

The “gapless” nature of the \( d \)-wave superconductivity in high-\( T_c \) cuprates opened a new possibility to study the electronic state at low temperature through quasiparticle (QP) heat transport [1, 2, 3, 4]. In the presence of disorder, QPs are created near the gap nodes with an “impurity bandwidth” \( \gamma \) [14] and carry heat; intriguingly, those QPs are also scattered by the same disorder, and in some cases the effects of creation and scattering balance, causing the thermal conductivity \( \kappa \) due to QPs to become independent of the disorder concentration at low \( T \). This phenomenon is called the “universal thermal conductivity”, which is best understood in the framework of the self-consistent \( T \)-matrix approximation (SCTMA) theory [14] for \( BCS d \)-wave superconductors. In the SCTMA theory, when \( \gamma \) is in the universal limit \( k_BT \ll \gamma \ll \Delta_0 \) (\( \Delta_0 \) is the maximum gap), the residual component \( \kappa_0/T \) (\( T \to 0 \) limit of \( \kappa/T \)) becomes independent of the QP scattering rate \( \Gamma \) and is expressed as

\[
\frac{\kappa_0}{T} = \frac{k_B^2 n}{3h c} \left( \frac{v_F}{v_2} \right) \left( \frac{v_2}{v_F} \right) \geq \frac{k_B^2 n v_F}{3h c v_2}, \tag{1}
\]

with \( n \) the number of CuO\(_2\) planes per unit cell, \( c \) the c-axis lattice constant, and \( v_F \) (\( v_2 \)) the QP velocity normal (tangential) to the Fermi surface at the node. This is useful because, if valid, one can obtain \( \Delta_0 \) by the bulk measurement of \( \kappa \) [5, 6].

Experimentally, \( \kappa_0/T \) was reported to be approximately independent of the impurity scattering in nearly optimally-doped YBa\(_2\)Cu\(_3\)O\(_y\) (YBCO) [7, 8] and Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{8+\delta}\) (Bi2212) [9], and its magnitude for Bi2212 was consistent with Eq. (1) using the angle-resolved photoemission spectroscopy (ARPES) results [17]. Hence, it has been considered that the universal thermal conductivity is essentially realized in cuprates. However, the experimental results so far [7, 8, 9, 10, 11, 12, 13] carry rather large error bars and thus it has not been clear to what extent \( \kappa_0/T \) is “universal”. Moreover, Hussey et al. [10] found an absence of \( \kappa_0/T \) in an underdoped YBa\(_2\)Cu\(_4\)O\(_8\), which questions the universal scenario and suggests a QP localization phenomenon that does not show up in the SCTMA theory. In this regard, an unusual QP localization in magnetic field was recently observed in underdoped La\(_{2-\delta}\)Sr\(_x\)CuO\(_4\) (LSCO) [12, 13], and, while it is obviously related to the magnetic-field-induced spin/charge order [12, 18], the mechanism of this localization is not yet understood.

Theoretically, whereas the SCTMA theory is a perturbation theory, there are other non-perturbative approaches [14, 19, 20] that tend to predict QP localizations. Actually, when the superconducting (SC) state is inhomogeneous, the SCTMA theory is clearly inappropriate and nonperturbative theories becomes necessary. Since the cuprate superconductors have an inherent tendency to develop electronic inhomogeneities in the SC state because of the short coherence length, and indeed various sorts of electronic inhomogeneities are being discovered in cuprates [21, 22, 23, 24], it is important to sort out the consequence of the inhomogeneity in the low-energy physics probed by the QP heat transport.

In this Letter, we critically examine the universal thermal conductivity by studying YBCO, LSCO, and Bi2212 for various doping levels; what is crucial to this work is that our samples are among the best single crystals available for these three systems, well characterized by various transport properties [25, 26, 27, 28, 29, 30], and the \( \kappa \) measurements are done with a small absolute uncertainty of less than 10%. In our experiments, we find three features that all indicate a breakdown of the universal thermal conductivity upon a closer look: i) a slight Zn substitution for Cu induces notable suppression of \( \kappa_0/T \) in underdoped and optimally-doped samples of all the three systems; ii) in Bi2212, both the magnitude and the doping dependence of the gap parameter \( v_F/v_2 \) obtained from \( \kappa_0/T \) via Eq. (1) differ significantly from the ARPES results [17, 31]; iii) a large in-plane anisotropy of \( \sim 2 \) is observed for \( \kappa_0/T \) in Bi2212. We discuss that the electronic inhomogeneity plays a major role in the departure from the universality by causing a partial QP localization as suggested by the nonperturbative theories.
High-quality single crystals of YBa$_2$(Cu$_{1-z}$Zn$_z$)$_2$O$_y$, La$_{2-x}$Sr$_x$Cu$_{1-z}$Zn$_z$O$_4$ and Bi$_{2}$Sr$_2$Ca(Cu$_{1-z}$Zn$_z$)$_2$O$_8$+$\delta$ are grown by a flux method [25], a traveling-solvent floating-zone method [26] and a floating-zone method [28, 29], respectively. The actual Zn concentration $z$ is determined by the inductively-coupled plasma atomic-emission spectroscopy (ICP-AES). All the YBCO samples are perfectly detwinned and $\kappa$ is measured along the $a$ axis to avoid the additional electronic heat transport coming from the Cu-O chains. For a doping-dependence study of Bi2212, Bi$_2$Sr$_2$CaCu$_2$O$_8$+$\delta$ crystals as well as Bi$_2$Sr$_2$Ca$_{1-x}$Y$_x$Cu$_2$O$_8$+$\delta$ (Dy-Bi2212) crystals (also produced by the floating-zone method) are carefully annealed at 400–800°C in suitable atmospheres to tune the oxygen content. We label the Bi2212 samples as UD70, OP94, OD70, etc., by their doping regimes and $T_c$ values, and the Dy-Bi2212 samples as Dy81, Dy45, etc., by their $T_c$ values (all in underdoped regime) [28]. The thermal conductivity measurement in millikelvin region is done by a conventional steady-state “one heater, two thermometer” technique in a dilution refrigerator [11, 12].

We emphasize that our $\kappa$ data are reproducible with typical variations of less than 10%, thanks to the consistently high quality of the crystals and a good control of the doping level [25, 26, 27, 28, 29].

First, we show the effect of Zn-substitution on the low-$T$ QP heat transport for different doping regimes. Figure 1 shows the $\kappa/T$ vs $T$ plots for YBCO, LSCO, and Bi2212 systems in the underdoped and optimally-doped regimes. The $T=0$ intercepts of the linear fits to the lowest-$T$ data give the residual QP component $\kappa_0/T$, which is decreased upon slight Zn substitution in all these systems [33], whereas within the SCTMA theory $\kappa_0/T$ is predicted to increase with $\Gamma$ when the system is not in the universal limit [13, 24]. We note that in YBCO at $y=7.00$, $\kappa_0/T$ decreases by $\sim$20% with 0.6% of Zn [Fig. 1(b)]; while this behavior suggests a departure from the universal behavior given our small error bar ($\sim$10%) [33], it is not inconsistent with the previous results by Taillefer et al. for $y=6.9$ [1, 2], where the error bar was as large as $\sim$40%. A more pronounced suppression in $\kappa_0/T$ of $\sim$40% is observed upon 0.6% of Zn-substitution in underdoped YBCO at $y=6.50$ [Fig. 1(a)], for which there has been no previous report. Apparently, the present higher-precision measurements indicate that all three systems do not strictly display the universal behavior in the underdoped and optimally-doped regimes; furthermore, the observed suppression of $\kappa_0/T$ by Zn substitution is opposite to what is expected from the SCTMA theory. If we remember that a non-perturbative theory that takes into account the gap inhomogeneities induced by impurities predicts a QP localization [12], it seems most likely that the observed behavior is essentially due to the Zn-induced electronic inhomogeneity in the form of nonsuperconducting droplets [22].

Figure 2 shows the Zn-substitution effect on $\kappa$ in overdoped LSCO and Bi2212. In overdoped cuprates, serious electron-phonon decoupling occurs at very low $T$ [36, 37], causing the QP thermal conductivity to be undetectable.
by the conventional steady-state technique. As a result, a strong downturn shows up in the lowest-\(T\) data of \(\kappa/\tau\) for overdoped samples \[36, 37\], which is also the case with our data in Fig. 2. [Our optimally-doped Bi2212 also shows this problem below 100 mK, see Fig. 1(f)]. It is therefore impossible to precisely determine \(\kappa_0/\tau\) of these samples from the present data. Nevertheless, for a qualitative evaluation of the Zn-substitution effect, one can crudely estimate \(\kappa_0/\tau\) by a linear fitting to a higher-temperature range, as shown in Fig. 2, and it would be safe to conclude that upon Zn substitution \(\kappa_0/\tau\) is enhanced in these overdoped samples. This trend is opposite to that observed in underdoped and optimally-doped samples in Fig 1, and is probably understandable within the SCTMA theory \[15, 34\]. Hence, there is a crossover near optimum doping beyond which the nodal QPs behave more ordinarily.

We next show the doping dependence of \(\kappa_0/\tau\) for Bi2212, where the gap parameters have been reliably obtained by ARPES studies \[17, 31\]. Figures 3(a) and 3(b) show the low-\(T\) thermal conductivity (along the \(a\) axis) of Bi2212 and Dy-Bi2212 single crystals, and Fig. 3(c) summarizes the doping dependence of \(\kappa_0/\tau\). Here, the hole concentration per Cu, \(p\), is determined by the method employing the Hall coefficient proposed in Ref. \[38\]. Clearly, \(\kappa_0/\tau\) decreases with decreasing doping and approaches zero upon entering the nonsuperconducting region — this behavior is essentially the same as that observed in LSCO \[2, 3\] and YBCO \[11\]. The gap parameter \(v_F/v_2\) calculated using Eq. \(1\) is shown in Fig. 3(d); also included in this panel are the \(v_F/v_2\) values directly obtained from ARPES for several underdoped Bi2212 crystals \[31\]. Not only the opposite trend in the doping dependence, but also the difference in the \(v_F/v_2\) values from \(\kappa_0/\tau\) and from ARPES, demonstrate an inadequacy of Eq. \(1\). To be more precise, \(\kappa_0/\tau\) of 0.051 W/Km for the optimally-doped Bi2212 yields \(v_F/v_2 = 65\) and \(\Delta_0 = 10\) meV (assuming a simple \(d_{x^2−y^2}\) gap), in contrast to \(v_F/v_2 = 20\) and \(\Delta_0 = 35\) meV obtained from ARPES \[17, 31\]. The reason why \(\kappa_0/\tau\) is much larger than what the ARPES-derived \(v_F/v_2\) would predict via Eq. \(1\) is not clear, but this should be understood in relation with the electronic inhomogeneity, given that there is likely a large variation in the SC gap magnitude in this material \[21, 24\]. It might be that, depending on the nature of the inhomogeneity, sometimes the QP creation effect overwhelms the QP scattering effect (which may be the case near optimum doping) and sometimes the opposite happens (possibly in the underdoped regime). Clearly, more work is called for on the theoretical part.

Figure 4 shows another novel property observed in Bi2212; namely, the QPs show very different abilities to conduct heat along the \(a\) and \(b\) axes that correspond to the two orthogonal nodal directions. Indeed, the data in Fig. 4 demonstrate that in the \(T → 0\) limit the anisotropy in \(\kappa_0/\tau\) is more than a factor of 2. If the universal scenario holds here and Eq. \(1\) is valid, the in-plane anisotropy would mean that the SC gap is not the standard \(d_{x^2−y^2}\), a possibility discussed in Ref. \[29\]; however, given the large anisotropy for \(T → 0\) and the problems in the universal scenario, it seems more natural to consider that the source of the anisotropy is not an asymmetry in the SC gap but is the breakdown of the
universal scenario. To corroborate this interpretation, the $\kappa_0/T$ values for both $a$ and $b$ axes measured here in our optimally-doped Bi2212 are much larger than the values reported previously \cite{3,4}; such a dependence of $\kappa_0/T$ on the sample source suggests that the details of the electronic inhomogeneities matter in Bi2212.

As for the possible source of the in-plane anisotropy in Bi2212, it is useful to notice that the latest “gap map” data obtained by the scanning tunneling microscope experiments by McElroy et al. \cite{21} present clear anisotropy in the electronic inhomogeneity; in Fig. 2B of Ref. \cite{21}, the variation of the gap magnitude is rather smooth along the $a$ axis, while it is much steeper along the $b$ axis. If a stronger electronic inhomogeneity causes a larger tendency for QP localization, as the calculations based on the Bogoliubov-de Gennes equation suggest \cite{14,19}, one would expect $\kappa_b$ to become smaller than $\kappa_a$ in the situation as indicated in Ref. \cite{21}. Hence, the unusual in-plane anisotropy of $\kappa_0/T$ in Bi2212 is likely to be related to the anisotropic electronic inhomogeneity, that is indirectly caused by the supermodulation structure peculiar to this system.

A clear message one can take is that the QP thermal conductivity is unexpectedly sensitive to the electronic inhomogeneity, whatever the nature of the inhomogeneity is; it can be the local suppression of the SC gap by Zn impurities \cite{22}, or it can be the gap inhomogeneity produced by dopant atoms \cite{21}. The qualitative change in the Zn-substitution dependence near optimum doping suggests that a strong tendency to form an electronic inhomogeneity is present only in the under- to optimally-doped regime; in this regard, it is useful to note that the dynamics of photo-induced QPs in our Bi2212 crystals shows a sharp change at optimum doping, and a more BCS-like behavior is found in the overdoped regime \cite{39}. In any case, for a better understanding of the QP properties in cuprates, one should definitely take into account the QP localization due to electronic inhomogeneities, and the present work demonstrates a crucial role of the nanoscale inhomogeneity in the low-energy physics of high-$T_c$ cuprates.

We greatly thank K. Behnia, P. J. Hirschfeld, N. E. Hussey and J. Takeya for helpful discussions. This work was supported by the Grant-in-Aid for Science provided by the Japan Society for the Promotion of Science.

* Present address: CERC,AIST, Tsukuba 205-8562, Japan

[1] L. Taillefer et al., Phys. Rev. Lett. 79, 483 (1997).
[2] M. Chiao et al., Phys. Rev. Lett. 82, 2943 (1999).
[3] M. Chiao et al., Phys. Rev. B 62, 3554 (2000).
[4] S. Nakamae et al., Phys. Rev. B 63, 184509 (2001).
[5] N. E. Hussey et al., Phys. Rev. Lett. 85, 4140 (2000).
[6] J. Takeya, Y. Ando, S. Komiya, and X. F. Sun, Phys. Rev. Lett. 88, 077001 (2002).
[7] M. Sutherland et al., Phys. Rev. B 67, 174520 (2003).
[8] D. G. Hawthorn et al., cond-mat/0502273.
[9] R. W. Hill et al., Phys. Rev. Lett. 92, 027001 (2004).
[10] Y. Ando et al., Phys. Rev. Lett. 92, 247004 (2004).
[11] X. F. Sun, K. Segawa, and Y. Ando, Phys. Rev. Lett. 93, 107001 (2004); Phys. Rev. B 72, 100502(R) (2005).
[12] X. F. Sun, S. Komiya, J. Takeya, and Y. Ando, Phys. Rev. Lett. 90, 117004 (2003).
[13] D. G. Hawthorn et al., Phys. Rev. Lett. 90, 197004 (2003).
[14] For a review, see N. E. Hussey, Adv. Phys. 51, 1685 (2002).
[15] M. J. Graf, S-K. Yip, J. A. Sauls, and D. Rainer, Phys. Rev. B 53, 15147 (1996).
[16] A. C. Durst and P. A. Lee, Phys. Rev. B 62, 1270 (2000).
[17] A. Damascelli, Z.-X. Zhen, and Z. Hussain, Rev. Mod. Phys. 75, 473 (2003).
[18] V. P. Gusynin and V. A. Miransky, Eur. Phys. J. B 37, 363 (2004); M. Takigawa, M. Ichioka, and K. Machida, J. Phys. Soc. Jpn. 73, 2049 (2004).
[19] W. A. Atkinson and P. J. Hirschfeld, Phys. Rev. Lett. 88, 187003 (2002).
[20] S. Vishveshwara, T. Senthil, and M. P. A. Fisher, Phys. Rev. B 61, 6966 (2000).
[21] K. McElroy et al., Science 309, 1048 (2005).
[22] S. H. Pan et al., Nature (London) 403, 746 (2000).
[23] M. Vershinin et al., Science 303, 1995 (2004).
[24] C. Howald, P. Fournier, and A. Kapitulnik, Phys. Rev. B 64, 100504(R) (2001).
[25] K. Segawa and Y. Ando, Phys. Rev. Lett. 86, 4907 (2001); Y. Ando and K. Segawa, Phys. Rev. Lett. 88, 167005 (2002).
[26] S. Komiya, Y. Ando, X. F. Sun, and A. N. Lavrov, Phys. Rev. B 65, 214553 (2002); S. Komiya, H.-D. Chen, S.-C. Zhang, and Y. Ando, Phys. Rev. Lett. 94, 207004 (2005).
[27] S. Komiya and Y. Ando, Phys. Rev. B 70, 060503(R) (2004).
[28] Y. Ando et al., Phys. Rev. B 62, 626 (2000).
[29] Y. Ando et al., Phys. Rev. Lett. 88, 147004 (2002).
[30] Y. S. Lee et al., Phys. Rev. B 72, 054529 (2005); W. J. Padilla et al., Phys. Rev. B 72, 060511(R) (2005).
[31] J. Mesot et al., Phys. Rev. Lett. 83, 840 (1999).
[32] Actual Dy contents of the Dy-Bi2212 samples are determined by ICP-AES: 0.20 (Dy80); 0.27 (Dy52); 0.34 (Dy42); 0.36 (Dy36); 0.51 (Dy0). Dy0 is a nonsuperconducting sample close to the SC boundary.
[33] While the slope of the linear fit is usually determined by the cross section of the sample due to the boundary scattering of phonons, the large slopes in our Zn-free YBCO are indicative of the $T^3$ contribution of the thermally-excited QPs in very clean samples \cite{3}, which testifies the “ultraclean” nature of our YBCO crystals. This additional $T^3$ term does not affect the determination of $\kappa_0/T$.
[34] Y. Sun and K. Maki, Europhys. Lett. 32, 355 (1995).
[35] For those YBCO data, the uncertainty in $\kappa_0/T$ comes mostly from geometrical-factor errors (5–10%) and much less from the linear fitting.
[36] M. F. Smith, J. Paglione, M. B. Walker, and L. Taillefer, Phys. Rev. B 71, 014506 (2005).
[37] S. Nakamae et al., Phys. Rev. B 68, 100502(R) (2003).
[38] Y. Ando et al., Phys. Rev. B 61, R14956 (2000); 63, 069902(E) (2001).
[39] N. Gedik et al., Phys. Rev. Lett. 95, 117005 (2005).