Absence of Residual Quasiparticle Conductivity in the Underdoped Cuprate $YBa_2Cu_4O_8$

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We report measurements of the in-plane thermal conductivity κ of the stoichiometric underdoped cuprate $YBa_2Cu_4O_8$ (Y124) below 1 K. $\kappa(T)$ is shown to follow a simple phononic T^3 dependence at the lowest temperature T for both current directions, with a negligible linear quasiparticle contribution. This observation is in marked contrast with behavior reported in optimally doped cuprates, and implies that extended zero-energy (or low-energy) quasiparticles are absent in Y124.

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Experimental evidence for d-wave superconductivity in high-T_c cuprates (HTC) is now well established. The presence of nodes in the gap is expected to produce a finite density of well-defined quasiparticle (QP) excitations at low energies that dominate the low-T physics. For a *pure* d-wave superconductor with line nodes on the Fermi surface, for example, the density of states (DOS) is linear in the excitation energy, giving rise to a T^2 dependence of the low-T specific heat and thermal conductivity (assuming a constant scattering rate). This excitation spectrum, however, is altered significantly in the presence of impurities. For an anisotropic two-dimensional superconductor with scattering in the unitary limit, a band of impurity states is expected to develop whose width γ grows with increasing impurity concentration n_{imp}, leading to a finite zero-energy DOS [1–4]. Lee showed that the residual conductivity is independent of n_{imp}, the result of compensation between the increased DOS and a reduction in the associated transport lifetime [2]. This residual, or "universal" conductivity develops at low T in the so-called "dirty" limit, $k_B T \le \gamma$. Similarly, the low-T $\kappa(T)$ will be dominated by QP states in the vicinity of the line nodes, and is given by the expression [4]

$$\kappa_{\mathrm{res}}/T \approx (n/d)\,(k_B^2/3)\,(v_F/v_2)\,, \qquad (1)$$

where n/d is the stacking density of CuO_2 planes and v_F and v_2 are the energy dispersions (QP velocities) perpendicular and tangential to the Fermi surface, respectively.

The issue of localization of electronic states in d-wave superconductors, however, is a complex problem and many competing viewpoints prevail [2–8]. Balatsky and Salkola [5], for example, argue that the long-range nature of hopping between impurity states along the nodal directions leads to strong overlap of the impurity wave functions along the diagonals of the square lattice, and ultimately to "extended" impurity states. Senthil *et al.* [6,7], however, argue that quantum interference effects destabilize the extended QP states and lead to a vanishing DOS at zero energy (see also [8] for a similar conclusion). They coined the phrase, "superconducting thermal insulator," [7] to describe such a superconductor with localized states.

Experimentally, the observation of a finite linear term in $\kappa(T)$ in pure and Zn-doped $YBa_2Cu_3O_{7-\delta}$ (Y123) by Taillefer *et al.* [9] appeared to confirm the existence of zero-energy quasiparticles. Moreover, the size of this term was indeed found to be universal, i.e., independent of Zn concentration, in agreement with Lee's prediction. Similar behavior was later reported for $Bi_2Sr_2CaCu_2O_8$ (Bi2212) by Behnia *et al.,* using irradiated crystals [10]. More recently, the magnitude of κ_{res}/T in Bi2212 ($\approx$0.15 $\mathrm{mW/cm\,K^2}$) was shown to be consistent with absolute values of v_F/v_2 estimated from angle-resolved photoemission spectroscopy (ARPES) [11,12].

Despite this apparent consistency between theory and experiment, κ_{res}/T has only been reported for two compounds, both at their optimum doping level, and it is not immediately obvious how κ_{res}/T will vary across the phase diagram. ARPES [11] and penetration depth measurements [13] indicate that v_F/v_2, and thereby κ_{res}/T, increase in the underdoped regime. On the other hand, in certain underdoped cuprates, where T_c has been suppressed in high magnetic fields [14,15], there is a marked tendency towards localization below T_c, suggesting a *vanishing* QP contribution at low T. Clearly, low-T $\kappa(T)$

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measurements on underdoped cuprates are important to help clarify this seemingly contradictory behavior.

With this in mind, we have carried out low-T $\kappa(T)$ measurements on the underdoped cuprate Y124 ($T_c = 80$ K), which is a self-doped, stoichiometric cuprate and therefore relatively free of disorder. Below 0.25 K, $\kappa(T) \propto T^3$ for both a- and b-axis currents, consistent with a phonon heat conduction in the ballistic regime. The residual linear QP term, however, is either absent or is negligibly small, with an upper bound of 0.02 mW/cm K^2. Clearly, the universal conductivity scenario breaks down dramatically in underdoped Y124. One compelling possibility is that the QP states in Y124 are localized at low T, due to the proximity to the superconductor/insulator (S/I) boundary, and therefore do not contribute to the low-T heat transport.

The Y124 crystals were grown by a flux method described elsewhere [16]. For this particular study, three platelike crystals were selected, two with their longest dimension along the b axis, and the other along the a axis. Approximate dimensions were $0.25 \times 0.16 \times 0.015$ mm^3 for the κ_a crystal (labeled hereafter as $a\sharp 1$) and $1 \times 0.09 \times 0.05$ mm^3 and $0.8 \times 0.25 \times 0.06$ mm^3 for the two κ_b crystals, $b\sharp 1$ and $b\sharp 2$. $T_c = 80$ K for all crystals, with a transition width, measured resistively, of less than 1 K.

$\kappa(T)$ for each crystal was measured between 0.14 and 1 K using a conventional steady-state four-probe technique that allowed the electrical resistivity $\rho_{a,b}(T)$ of each sample to be measured *in situ* without changing the contact configuration. Gold wires were attached as electrical contacts using Dupont 6838 silver paint. The $\rho_{a,b}(T)$ behavior was found to be in excellent agreement with previous measurements [17], with room temperature values, $\rho_a = 350\ \mu\Omega$ cm and $\rho_b = 90\ \mu\Omega$ cm (for both b-axis crystals). This large in-plane anisotropy arises from the high conductivity of carriers on the quasi-1D CuO chains that run parallel to the b axis (see schematic inset of Fig. 1) and confirms not only the high quality of the crystals used in this study but also that the current flow in each case is uniaxial. The temperature gradient was measured by two RuO_2 thermometers connected to the "voltage" contacts through the gold wires, and the thermometers were supported by long, thin superconducting Nb-Ti wires to minimize heat losses. Uncertainties in the absolute magnitudes of $\kappa(T)$ [$\rho(T)$], mainly due to the finite contact dimensions on these small crystals, are estimated to be about 15% for κ_b (ρ_b) and about 25% for κ_a (ρ_a).

The $\kappa(T)$ data for all crystals are shown on double-logarithmic axes in Fig. 1. The variation of $\kappa_b(T)$ for the two b-axis crystals is almost identical over the whole temperature range studied, giving us confidence in the reproducibility of our data. Below 0.25 K, κ_a and κ_b both vary approximately as T^3, consistent with phonon heat transport in the boundary-scattering limit. Above 0.25 K, $\kappa_a(T)$ deviates more strongly from a T^3 dependence. The origin of the enhancement of κ_b over κ_a above $T = 0.25$ K is not understood at present, though we assume it reflects an additional contribution to κ from the CuO chains. This could reflect a substantial anisotropy in the lattice heat conduction identified in measurements at higher T [18]. Alternatively, the QP conductivity on the chains might develop swiftly above 0.25 K (note that the CuO chains, being quasi-1D, may be susceptible to charge ordering at very low T). Further measurements in a magnetic field are envisaged to help clarify the origin of this anisotropy.

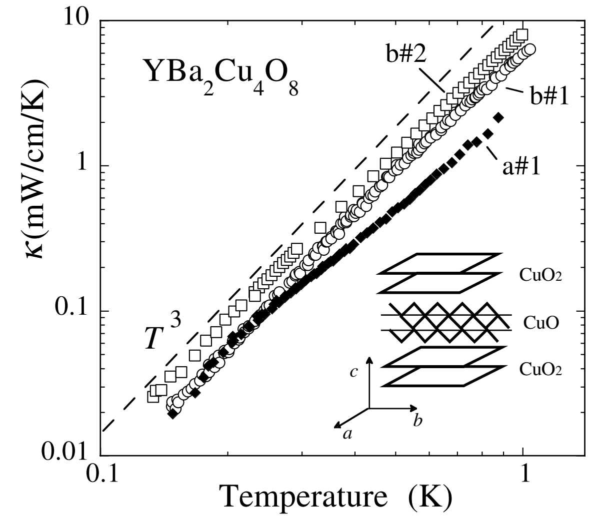

FIG. 1. $\kappa_a(T)$ and $\kappa_b(T)$ of Y124, plotted on double-logarithmic axes. The dashed line represents the T^3 dependence expected for phonon heat transport in the ballistic regime. The inset shows a simple schematic of the crystal structure of Y124.

In order to look for evidence of a linear κ_{res}, we have replotted the low-T data in Fig. 2 as $\kappa_{a,b}/T$ versus T^2 and fitted each data set below 0.25 K to the expression $\kappa_{a,b} = AT + BT^3$ [19]. The coefficients for each fit are $A = 0.011$, -0.007, and 0.006 (± 0.02) mW/cm K^2 and $B = 6.50$, 7.00, and 10.67 (± 0.50) mW/cm K^4 for $a\sharp 1$, $b\sharp 1$, and $b\sharp 2$, respectively.

In the boundary-scattering limit, κ_{ph} is given by

$$\kappa_{\text{ph}} = 1/3\beta\langle v_{\text{ph}}\rangle l_0 T^3, \tag{2}$$

where β is the phonon specific heat coefficient, $\langle v_{\text{ph}}\rangle$ is the average acoustic sound velocity, and $l_0 = 2w/\sqrt{\pi}$ is the maximum phonon mean-free path. Here, w represents a mean width of the rectangular-shaped crystal. Taking the dimensions of our crystals and suitable values for β ($= 0.5 \pm 0.1$ mJ/mol K^4) [20] and $\langle v_{\text{ph}}\rangle$ ($= 5 \pm 1 \times 10^5$ cm/s) [21], we obtain estimates for $\kappa_{\text{ph}}/T^3 = 4.1 \pm 1.2$, 5.25 ± 1.5, and 9.55 ± 2.0 mW/cm K^4 for $a\sharp 1$, $b\sharp 1$, and $b\sharp 2$, respectively [22]. Given the uncertainties in measuring dimensions and contact distances, we believe these values compare favorably with the experimental values. More importantly, the size of the T^3 term for the two b-axis crystals scales well with w, and we assign the T^3 contribution simply to the phonon contribution in the ballistic regime.

The most striking result here is the complete absence (to within experimental error) of the residual linear term in the low-T $\kappa(T)$ for *both* chain and plane current directions. It

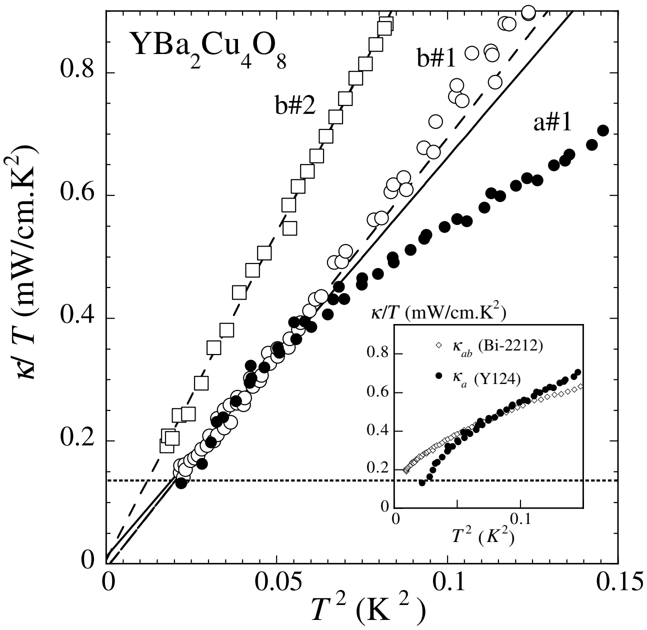

FIG. 2. κ/T versus T^2 below 0.4 K for $a \sharp 1$ (closed circles), $b \sharp 1$ (open circles), and $b \sharp 2$ (open squares). Fits to the expression $\kappa = AT + BT^3$ below 0.25 K are indicated by dashed lines for $b \sharp 1$ and $b \sharp 2$ and by a solid line for $a \sharp 1$. The dotted line represents the universal conductivity limit for Bi2212. Inset: comparison of κ_a/T for Y124 (closed circles) and κ_{ab}/T of Bi2212 [10] (open diamonds), measured in the same apparatus.

should be emphasized, of course, that a zero linear term in κ_b naturally implies a negligible $\kappa_{\rm res}$ for the planes along the b direction also. Thus, we have effectively confirmed the absence of the universal QP term in Y124 in three different crystals. Moreover, for there to be any finite zero-T intercept in κ/T, it would require $\kappa_{\rm ph}(T)$ below 0.15 K to vary as T^{3+n} with $n > 0$, which is simply not physical, given that the lattice heat capacity is strictly cubic below 1 K. Thus, we are confident that the main result of this Letter, namely, the absence of $\kappa_{\rm res}$ in Y124, is robust. For comparison, we also show, in the inset of Fig. 2, κ_{ab}/T data for optimally doped Bi2212 [10] measured with the same experimental setup [23]. In Bi2212, we can clearly distinguish a finite $\kappa_{\rm res}/T \approx 0.15$ mW/cm K^2 (shown by a dotted line), that is *an order of magnitude larger* than the upper limits for $\kappa_{\rm res}/T$ in Y124.

Despite the overwhelming support for d-wave pairing in HTC, there is still limited, direct evidence for a $d_{x^2-y^2}$ order parameter in Y124. Recently, however, the penetration depth along both the a and b axes in Y124 has been found to follow power-law T dependences down to 2 K [24,25], behavior consistent with a linear gap structure at the planar nodes and proximity effect coupling between the planes and chains [26]. Of course, at this stage, we cannot rule out completely the possibility of a finite gap in Y124, but, if it does exist, its magnitude would be vanishingly small. The orthorhombic distortion in Y124, induced by the chains, introduces some $d + s$ admixture in the gap function. As the s component is increased from zero, the position of the nodal lines is first shifted away from (π, π), but as the s component becomes comparable with the d component, a nodeless gap may form. Second, magnetic impurities are thought to induce a local, imaginary component that could also give rise to a fully gapped state and a suppression of low-T thermal transport [27]. This latter possibility, however, is not supported by specific heat data taken on crystals similar to those studied here which show no sign of a low-T Schottky anomaly arising from such magnetic impurities [20]. In what follows, therefore, we assume that nodal lines are present in Y124 and turn to consider the nature of the low-energy QP states in their vicinity.

From (1), we see that $\kappa_{\rm res}/T$ is directly proportional to the ratio v_F/v_2 at the nodal positions, so a negligible $\kappa_{\rm res}/T$ may indicate a sharp gap feature at the nodes at very low energies, induced either by doping, impurities, or structural modifications. However, as noted above, v_F/v_2 is expected to *increase* as we move into the underdoped regime, and indeed, independent estimates of v_F/v_2 from the slope of the low-T penetration depth [25] in Y124 yield an estimate for $\kappa_{\rm res}/T$ that is *larger* than those for both optimally doped Bi2212 and Y123. Moreover, given that band structure estimates for v_F are similar for Y123 and Y124 [28], even with our upper bound estimate for $\kappa_{\rm res}/T$ ($= 0.02$ mW/cm K^2), we obtain a physically unrealistic value ($v_F/v_2 \approx 2$) for the gap slope within the nodes in Y124. It appears unlikely, therefore, that the gap structure can itself explain a value of $\kappa_{\rm res}/T$, 1 order of magnitude lower than in Y123.

Another important consideration is the size of the impurity band, γ. We recall that, in the unitary limit, the universal conductivity regime develops below $k_BT \leq \gamma$. Thus, the observation of $\kappa_{\rm res}$ depends not only on the temperature range of the experiment but also on the energy scale of the impurity band, imposed by the scattering phase shift. However, by taking values for the b-axis residual resistivity $\rho_0 = 0.5\ \mu\Omega$ cm [17] and plasma frequency $\omega_p = 2.5$ eV [29], we obtain a *lower bound* estimate of γ in the unitary limit of 14 K, some 2 orders higher than the base temperature of our measurements. [Unfortunately, similar analysis for $J \parallel a$ cannot be performed due to the difficulties in estimating ρ_0 from $\rho_a(T)$.] Even in the opposite (Born) limit, where the impurity band becomes exponentially small, one expects a QP contribution from the continuum states that is even higher than the predicted $\kappa_{\rm res}/T$, and so the same arguments should still apply.

Given these simple yet rather compelling arguments, we are left to consider how QP localization might account for the absence of $\kappa_{\rm res}/T$ in underdoped Y124. As mentioned above, high magnetic field measurements on the underdoped cuprates $La_{2-x}Sr_xCuO_4$ [14] and $Pr_{2-x}Ce_xCuO_4$ [15] revealed that $\rho_{ab}(T)$, though "metallic" at high T, tends towards localization as $T \rightarrow 0$ K. The origin of this localization phenomenon is unknown at present. However, if the field-induced destruction of superconductivity leads directly to an insulating phase, then it is not unreasonable to assume that the superconducting state itself is characterized by *localized* QP excitations [7]. The crossover from metallic to insulating behavior in the normal state (i.e., above H_{c2}), as we approach the Mott insulator, suggests an increasing role of long-range interactions on the mobility of low-energy quasiparticles. The superfluid condensate, suppressed in the vicinity of an impurity, becomes ineffective in screening the Coulomb repulsion between quasiparticles in the bound state [30]. As we approach the parent insulator, we expect such interactions to grow, eventually leading to insulating behavior of the quasiparticles above the superconducting condensate and a negligible QP contribution to the low-T heat transport [31]. Such localization in a nominally clean superconductor is an exciting prospect, and measurements on Zn-doped or irradiated Y124 are envisaged to investigate this possibility further. We note here that most impurity models for d-wave (cuprate) superconductivity fail to take into account the developing role of long-range interactions between quasiparticles as the S/I boundary is approached. We hope, therefore, that this Letter stimulates renewed theoretical efforts toward understanding the nature of QP excitations in the CuO_2 planes, deep inside the superconducting state, and in particular in stoichiometric crystals on the underdoped side of the phase diagram.

In conclusion, we have measured the low-T $\kappa(T)$ of stoichiometric, underdoped Y124 and have found that, in marked contrast to optimally doped Y123 and Bi2212, the universal QP conductivity term is absent. We have considered several interpretations of this intriguing result, including localization of the quasiparticles themselves, due to enhanced long-range interactions as we approach the S/I boundary. Prior to this work, the observation of universal QP conductivity in Y123 and Bi2212 had been widely regarded as solid support for the picture of long-lived quasiparticles above the superconducting ground state of HTC. Our surprising result offers important and timely counterevidence that the residual conductivity term is nonuniversal, and we hope it encourages further debate and investigation into this critical and still controversial issue.

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