Effect of controlled disorder on quasiparticle thermal transport in Bi$_2$Sr$_2$CaCu$_2$O$_{8}$

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Low temperature thermal conductivity, $\kappa$, of optimally-doped Bi2212 was studied before and after the introduction of point defects by electron irradiation. The amplitude of the linear component of $\kappa$ remains unchanged, confirming the universal nature of heat transport by zero-energy quasiparticles. The induced decrease in the absolute value of $\kappa$ at finite temperatures allows us to resolve a nonuniversal term in $\kappa$ due to conduction by finite-energy quasiparticles. The magnitude of this term provides an estimate of the quasiparticle lifetime at subkelvin temperatures.

Fifteen years after their discovery, high-$T_c$ superconductors continue to attract significant attention and provoke intense debate [1]. One central issue is the extent of validity of the Fermi-liquid picture for describing the electronic excitations in these systems. The properties of the metallic state (even at optimal doping) appear to remain beyond such a picture. But, are there well-defined quasiparticles ($qp$) deep in the superconducting state as suggested by ARPES measurements [2]? And if yes, at which energy scale do they break down? To answer these questions, low-energy excitations in the superconducting state are under intense scrutiny [3,4].

Despite such marked accomplishment by the quasiparticle picture to treat the low-energy nodal excitations, numerous challenges remain. One is the absence of a linear term in low-temperature thermal conductivity of underdoped stoichiometric YBa$_2$Cu$_4$O$_8$ [5]. As there is no obvious reason to suppose that the structure of the superconducting gap in this system is radically different from that of the Y123 parent compound, this result suggests that somewhere in the underdoped regime, the Fermi-liquid picture might break down. Another issue is the effect of superconductivity on $qp$ lifetime. Transport studies in both Y123 [6,7] and Bi2212 [3] have documented a steep increase in the scattering time of quasiparticles below $T_c$. According to recent ARPES measurements by Valla et al. [8] on Bi2212, however, the $qp$ lifetime is not affected by the onset of superconductivity. This surprising discrepancy remains to be explained.

Here, we present a study of subkelvin thermal conductivity in optimally-doped Bi$_2$Sr$_2$CaCu$_2$O$_{8}$ before and after the introduction of vacant and interstitial sites by electron irradiation. We found that the zero-energy $qp$ conductivity remains constant despite the introduction of pair-breaking defects, while there is a substantial decrease in $qp$ thermal conductivity, $\kappa_{qp}$, at finite temperatures. This allows us to resolve the component of heat transport arising from finite-energy quasiparticles.
Two single crystals of Bi$_2$Sr$_2$CaCu$_2$O$_8$ were used in this study with typical dimensions of 1.0 x 0.3 x 0.02 mm$^3$. Thermal conductivity was measured with a conventional two-thermometer-one-heater set-up. Point defects were introduced by exposing the sample to a 2.5 MeV electron beam created by Van de Graaf accelerator at the Laboratoire des Solides Irradiés in Palaiseau, France. Fig. 1 show the effect of electron irradiation on electrical resistivity and the corresponding low temperature thermal conductivity of the two samples. As seen in the two insets, irradiation leads to a decrease in the critical temperature and an increase in normal-state resistivity, $\rho$, as reported in previous electron-irradiation studies [16]. Both pristine samples have a transition temperature ($T_c$ = 92K) and $d\rho/dT$ ($\rho = \rho_0 + bT\mu\Omega$ cm, with $b = 1.5\mu\Omega$ cmK$^{-1}$) characteristic of optimally doped Bi2212 crystals [15]. In the case of sample $a$ ($\rho_0 \sim 60\mu\Omega$), an irradiation flux of 6.0 x 10$^{19}$ e$^-$/cm$^2$ leads to a fourfold increase in residual resistivity ($\rho_0 \sim 240\mu\Omega$ cm) and a 20 K decrease in $T_c$ (73K). Moreover, a curvature appears in the temperature-dependence of resistivity which may suggest a doping change. In order to explore such possibility, we have measured the room-temperature thermopower, $S$(300K), which is a well known indicator of doping levels in high-$T_c$ superconductors [24]. The determined values of post-irradiation samples $a$ and $b$ (after the third irradiation) are $-0.1 \pm 0.1$ and $5.0 \pm 0.1\mu V/K$. The values of corresponding un-irradiated crystals are $-0.8 \pm 0.4$ and $2.4 \pm 0.1\mu V/K$, thus in both samples irradiation appear to have caused a slight “under-doping”. According to the study on $S$(300K) of Bi2212 [25], $\Delta T_c$ due to a possible variation in the doping level here are 1.8 K and 4.5 K for samples $a$ and $b$, respectively. These changes are considerably smaller than $\Delta T_c$ due to the pair-breaking effect from electron irradiation, 20 and 50 K, and thus we will assume that the effect on $qp$ transport arising from a small change in the doping level is negligible in comparison. It must be noted here that the thermal conductivity of sample $b$ was not measured before irradiation. Thus in the following discussions, we will concentrate on sample $a$ and will use the data on the sample $b$ only as supplementary evidence.

The main panels of Fig. 1 present the low temperature thermal conductivity data. The persistence of a sizeable phonon contribution, $\kappa_{ph}$, to the total heat transport complicates the determination of $qp$ conductivity. As $T \rightarrow 0$, $\kappa_{ph}$ is expected to enter the boundary scattering (or ballistic) regime, where phonon mean-free-path is limited by the dimensions of the crystal and $\kappa_{ph}$ becomes proportional to $T^3$. Above this regime, $\kappa_{ph}$ should increase more slowly due to the decrease in phonon mean-free-path. Therefore, by plotting $\kappa/T$ against $T^2$, one can resolve the zero-intercept, $\kappa_{00}/T$, corresponding to the residual linear component of $\kappa$ that is a signature of residual $qp$ conductivity. As seen in fig. 1b, there is a finite $\kappa_{00}/T$ term in the thermal conductivity of sample $a$ both before and after irradiation. The magnitude of this term shows little or no change in spite of the drastic increase in the number of defects. Note that the original contacts were kept throughout the successive measurements for each crystal, the sizeable uncertainty in $\kappa$ geometric factor does not hamper the effect of disorder. If we assume a linear extrapolation of the normal-state resistivity, the scattering rate in sample $a$ becomes four times larger after irradiation. If we further take the enhancement in the $qp$ lifetime below $T_c$ into account, the induced increase in impurity scattering rate becomes even larger. The amplitude of $\kappa_{00}/T$ found in sample $a$ (0.16 ± 0.03 mW/K$^2$cm) is similar to that of sample $b$ (0.13 mW/K$^2$cm) as well as to those reported in previous studies (0.14-0.15 mW/K$^2$cm) [21,4]. Theory predicts an increase in $\kappa_{00}/T$ at sufficiently high levels of impurity concentration due to the impurity-induced change in $T_c$ [22]. Therefore, considering that the sample $b$ after the third irradiation is nearly twenty times dirtier than the pristine sample $a$ (judging from their respective $\rho_0$ values), the “universal” character found in the values of $\kappa_{00}/T$ among all samples here is quite surprising. A quantitative comparison to the fine structure of the superconducting energy gap around the nodes can then be performed by inserting the value of $\kappa_{00}/T$ into the following equation [4]:

$$\kappa_{00}/T = \frac{k_B^2 v_F n}{3h v_2 d},$$

where $v_F$ and $v_2$ are $qp$ Fermi velocity and gap velocity at the nodes in the gap. One then obtains $\frac{\kappa_{00}}{T} = 21\pm3$ which is close to the ratio obtained by ARPES measurements [3]. The same conclusion has been previously drawn by Chiao et al. [3].

Before discussing the finite temperature $qp$ conductivity further, let us focus on phonon conductivity. For the sample $a$, if one can assume that the system enters the ballistic regime for $T < 0.25$ K, the extracted cubic term for phonon conductivity is $\kappa_{ph}/T^3 \sim 2.6$mW/K$^4$cm. This can be compared to the theoretically expected value using $\kappa_{ph} = 1/3\beta v_{ph}^3 T^3$, where $\beta$ is the phonon specific heat coefficient and $v_{ph}$ is the average sound velocity. Taking the transverse dimensions of our crystal along with the available values for $\beta = 0.0095$mJ/K$^4$cm$^{-3}$ [23] and $v_{ph} = 3600$mms$^{-1}$ [24], $\kappa_{ph}/T^3$ should be 5.9 mW/K$^4$cm, approximately twice the value determined from our data. One possible source for this discrepancy is that our measurement stops above the onset temperature of phonon ballistic conductivity. In the study reported by Chiao et al., the ballistic regime appears only below 130 mK. In this case, a downward curvature of the $\kappa/T$ curve would lead to a slight decrease in the finite intercept and the estimated $\kappa_{00}$ would become 0.14 mW/K$^2$cm (see Fig. 2a) but remains within our experimental uncertainty.

Another important aspect of Fig. 1b is a sizable irradiation-induced decrease in $\kappa(T)$ for the entire temperature range (0.13 K < $T$ < 0.9K). We begin by noting that the typical wavelength of acoustic phonons may
be estimated to vary as \( \lambda_{ph} = h\nu_{ph}/k_B T = 173 \text{nm/K}. \) Thus, at subkelvin temperatures, the spatial extension of lattice vibrations is more than two orders of magnitude larger than the size of introduced point defects and thus \( \kappa_{ph} \) should not be affected by irradiation. For this reason, the observed decrease must be exclusively due to an increase in the \( qp \)-defect scattering rate. However, the \( qp \) conductivity is expected to become independent of impurity scattering rate for \( T_c < \gamma \), the impurity bandwidth. Such condition is finally met in sample \( b \) after the second and the third irradiation where \( \kappa \) is virtually unchanged (see the lower panel of Fig.1). In relatively cleaner samples, the marked decrease in \( \kappa \) infers that \( \gamma \) must lie beyond our range of measurements.

Fig. 2 depicts the change in \( \kappa(T)/T \) of sample \( a \) before and after irradiation. \( \Delta \kappa/T \) is linear in \( T \), revealing a quadratic temperature dependence. The most plausible explanation for this result is to concede that \( qp \) conductivity of the pristine sample includes a quadratic term which is heavily diminished by irradiation. A fit to the data of Fig. 2 for the whole temperature range yields \( \Delta \kappa = aT + bT^2 \) with \( a = 0.005 \text{mW/K}^2 \text{cm} \) and \( b = 0.19 \text{mW/K}^2 \text{cm} \). The small size of the linear intercept indicates again that irradiation has left the linear term of \( qp \) conductivity intact. We now compare the amplitude of the term \( b \) with what is expected from the finite-energy \( qp \) contribution. For energies exceeding \( \gamma \), the density of states varies linearly with energy \( 2 \gamma N(E) = \frac{2}{\pi\hbar^2} \frac{1}{v_F v_2} E \).

This leads to a quadratic temperature dependence of specific heat \( C_e \)

\[
C_e = \frac{18\zeta(3)}{\pi} \frac{k_B^3 n}{h^2} \frac{1}{v_F v_2} T^2
\]

where \( \zeta(3) \approx 1.20 \) is a numerical factor. Thermal conductivity of these excitations can be estimated via kinetic theory; \( \kappa_e = \frac{4\zeta(3)k_B^2 n}{h^2 \frac{1}{v_F v_2}} \tau_e \). Here, \( \tau_e \) is the electronic scattering time. The temperature dependence of \( \tau_e \) governs the temperature dependence of electronic thermal conductivity. The relative weight of this term to the universal linear term can be estimated to be:

\[
\frac{\kappa_e}{\kappa_{00}} = \frac{18\zeta(3)k_B \tau_e T}{\pi \hbar}
\]

Note that while \( \kappa_{00}/T \) is universal, \( \kappa_e \) is not: its magnitude decreases with increasing disorder. The size of the quadratic term coefficient appearing in \( \Delta \kappa(T) \) (\( b = 0.19 \text{mW/K}^2 \text{cm} \)) corresponds to an electronic scattering time of 1.3 ps. On the other hand, \( qp \) lifetime can be estimated from \( \rho = \frac{\pi n e^2}{m^* \tau_e} \) and \( \omega_p = (4\pi ne^2/m^*)^{1/2} \), where \( \omega_p \) is the Drude plasma frequency. Using \( \omega_p = 1.1 \text{eV} \) \( \frac{22}{22} \), \( \rho(100K) = 200 \mu\Omega \text{cm} \) and \( \rho_0 = 60 \mu\Omega \text{cm} \), one finds \( \tau_e(100K) \approx 0.05 \text{ ps} \) and \( \tau_e(0K) \approx 0.2 \text{ ps} \), provided that the scattering rate is also linear in temperature for \( T < T_c \). Our results suggest a significant increase in \( qp \) lifetime, and thus, a substantial reduction in scattering rate below \( T_c \), in agreement with microwave conductivity data \( \frac{13}{13} \). It is instructive to compare the magnitude of \( \tau_e \) in Bi2212 and Y123. In the latter system, \( \tau_e \) of high-quality crystals is of the order of 7 ps \( \frac{13}{13} \) and is reported to approach 20 ps \( \frac{24}{24} \) in BaZrO\(_4\) grown crystals. For Bi2212, recent study on the complex conductivity by Corson et al. showed the \( qp \) lifetime to approach \( \sim 1 \text{ ps} \) \( \frac{27}{27} \) at low temperatures, similar to the value calculated independently from our measurements. Further, assuming the scattering rate to remain constant \( \frac{24}{24} \), one can estimate the size of \( \kappa_e \) upward in temperature. The deduced value at 5K (\( \sim 4.8 \text{ mW/Kcm} \)) can be compared via the Wiedemann-Franz law to the available data on charge conductivity which is limited to \( T > 5K \) \( \frac{13}{13} \). This yields \( \frac{\kappa_{00}(5K)}{\sigma} = 21.3 \text{ nW/Kcm} \), very close to the Sommerfeld value (\( \lambda_0 = 24.5 \text{ nW/Kcm} \)). Thus, our interpretation appears to be consistent with what is known from charge conductivity.

In spite of this apparent consistency, this analysis cannot accommodate the broader theoretical picture of \( qp \) transport in \( d \)-wave superconductors elaborated during the recent years. First, a linear \( N(E) \) implies an energy-dependent \( \tau_e \). In the unitary limit, for example, this leads to a cubic (instead of a quadratic) behavior for finite-energy \( qp \) transport for \( T > \gamma \). Second, the size of \( \gamma \) strongly depends on the impurity density \( (n_{imp}) \) as well as the scattering phase shift \( \delta \). In the unitary \( (\delta = \pi/2) \) limit, \( \gamma \) increases substantially with increasing scattering rate, \( \Gamma \); \( \gamma \sim (\Gamma \Delta_0)^{1/2} \). In the Born \( (\delta = 0) \) limit, its enhancement with \( \Gamma \) is largely attenuated for \( \Gamma \ll \tau_e \); \( \gamma \sim \Delta_0 \exp(-\frac{\Delta_0}{\Delta_0}) \). Here \( \Gamma = \frac{1}{\tau_e} \) and \( \Delta_0 \) is the magnitude of the superconducting gap \( \frac{4}{4} \). Now, with \( \tau_e \sim 1 \text{ ps} \) (which would yield \( \Gamma \sim 3K \)) and \( \Delta_0 \sim 40 \text{ meV} \) \( \frac{28}{28} \), \( \gamma \) may be estimated to be 35K in the unitary limit and virtually zero in the Born limit. Thus, the size of \( \gamma \) found in this study is in sharp contrast with what is expected in the unitary limit. A slight deviation from the unitary limit can produce a linear \( \kappa(T)/T \) above its universal value in a limited temperature range \( \frac{23}{23} \), however, preliminary investigations reveal that a reasonable phase shift only cannot account for the magnitude of the excess conductivity observed here \( \frac{23}{23} \). Clearly, our findings constitute a challenge to existing theory.

In summary, we have studied the effect of electron irradiation on the low temperature thermal conductivity of optimally doped \( Bi_2Sr_2CaCu_2O_8 \). The quasiparticle contribution to heat transport was found to contain two distinct components, a linear term associated with zero-energy quasiparticles and a quadratic term originating from finite-energy quasiparticles. The linear term remains insensitive to the number of defects. The magnitude of the quadratic term allowed us to make a new independent estimation of quasiparticle scattering time at low temperatures, implying a significant increase in the quasiparticle transport lifetime below \( T_c \) as indicated by other probes.
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FIG. 1. Upper panel: Low-temperature thermal conductivity, $\kappa/T$, of sample $a$ is plotted as a function of $T^2$. The thin lines are guides to the eye. The thick line represents the expected asymptotic lattice conductivity at the ballistic regime when the phonon mean-free-path attains the average sample size (see text). The inset shows the resistivity data on the same sample. Lower panel: Same for sample $b$. Note that the thermal conductivity of this sample in the pristine state was not measured.

FIG. 2. Upper panel: The change in the thermal conductivity of the sample before and after electron irradiation divided by temperature and plotted as a function of temperature. The straight line is a fit to the data revealing a negligible intercept and a quadratic variation of $\Delta \kappa(T)$. Lower panel: Thermal conductivity of the sample before and after irradiation together with a sketch of electronic and lattice components of thermal conductivity in the pristine sample.

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Bi-2212 sample a

\[
\frac{\kappa}{T} \text{ (mW/K}^2\text{cm)}
\]

\[\rho \text{ (} \mu\Omega \text{ cm)}\]

before

after

Bi-2212 sample b

\[
\frac{\kappa}{T} \text{ (mW/K}^2\text{cm)}
\]

\[\rho \text{ (} \mu\Omega \text{ cm)}\]

\[T^2(K^2)
\]
