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Effect of controlled disorder on quasiparticle thermal transport in $Bi_2Sr_2CaCu_2O_8$

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Low-temperature thermal conductivity κ of optimally doped $Bi_2Sr_2CaCu_2O_8$ was studied before and after the introduction of point defects by electron irradiation. The amplitude of the linear component of κ remains unchanged, confirming the universal nature of heat transport by zero-energy quasiparticles. The induced decrease in the absolute value of κ at finite temperatures allows us to resolve a nonuniversal term in κ due to conduction by finite-energy quasiparticles. The magnitude of this term provides an estimate of the quasiparticle lifetime at subkelvin temperatures.

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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 (angle-resolved photoemssion spectrocscopy) 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]

Low-temperature thermal conductivity κ has proven to be an instructive probe of such excitations. A nonvanishing linear term in thermal conductivity of optimally doped $YBa_2Cu_3O_7$ (Y123) for $T \rightarrow 0$ was the first solid evidence for a finite density states of nodal quasiparticles at zero energy.[5] Moreover, as expected for the case of a *d*-wave gap,[6,7] the amplitude of this term was found to be universal; i.e., independent of impurity concentration.[5] Recently, Durst and Lee[3] showed that, regardless of Fermi-liquid corrections, this amplitude is intimately related to the fine structure of the superconducting gap in the vicinity of the nodes. Subsequently, Chiao *et al.*[4] found a quantitative agreement between this gap structure as deduced from thermal conductivity and the one directly observed by ARPES studies[8] in optimally doped $Bi_2Sr_2CaCu_2O_8$ (Bi2212). On the other hand, data on electronic specific heat[9–11] and penetration depth[12,13] are limited to $T > 2$ K and probe excitations within a higher energy interval where the density of states is a linear function of energy. The temperature dependence of electronic specific heat in Y123 is in agreement with theoretical expectations for the gap structure near the nodes.[4,11] The same holds for the thermal variation of superfluid density obtained by penetration-depth studies assuming a finite Fermi-liquid correction to charge transport.[8] Observing these remarkable quantitative agreements, Chiao *et al.*[4] argued that this is a strong indication for the validity of Fermi-liquid treatment of the nodal quasiparticles. However, one important parameter of this picture, the impurity bandwidth γ, the energy scale below which the density of states becomes constant, is yet to be measured experimentally.

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 $YBa_2Cu_4O_8$.[14] 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 (Ref. 15 and 16) and Bi2212 (Ref. 13) have documented a steep increase in the scattering time of quasiparticles below T_c. According to recent ARPES measurements by Valla *et al.*[17] 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 $Bi_2Sr_2CaCu_2O_8$ 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 κ_{qp} at finite temperatures. This allows us to resolve the component of heat transport arising from finite-energy quasiparticles.

Two single crystals of $Bi_2Sr_2CaCu_2O_8$ were used in this study with typical dimensions of $1.0 \times 0.3 \times 0.02$ mm^3.

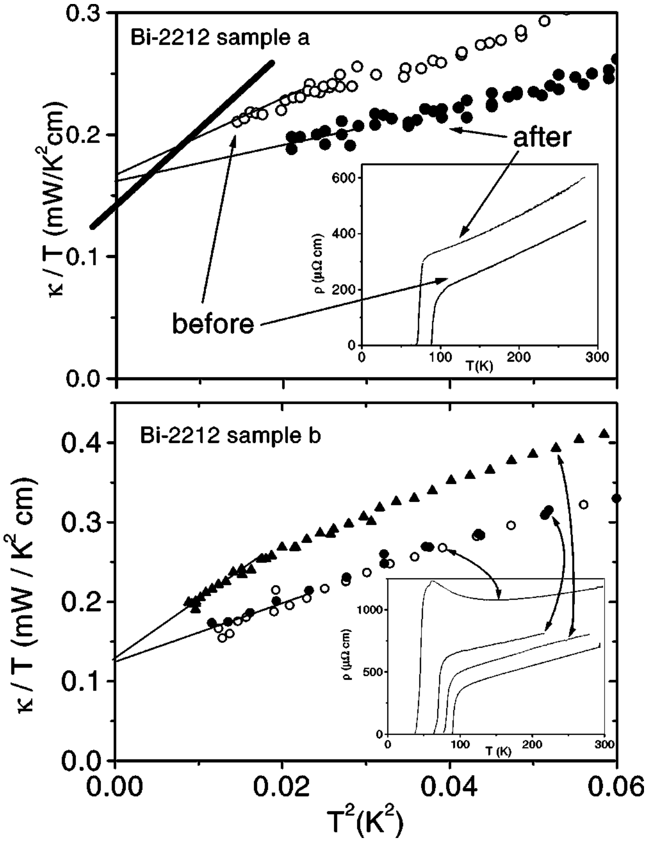

FIG. 1. Upper panel: Low-temperature thermal conductivity κ/T of sample *a* is plotted as a function of T^2. The thin lines are guides to the eye. The thick line represents the expected asymptotic lattice conductivity at the ballistic regime when the phonon mean-free-path attains the average sample size (see text). The inset shows the resistivity data on the same sample. Lower panel: Same for sample *b*. Note that the theral conductivity of this sample in the pristine state was not measured.

Thermal conductivity was measured with a conventional two-thermometer(RuO2)-one-heater setup. The thermometers were connected to the sample via gold wires and were supported by long, thin superconducting wires of Nb-Ti to minimize heat losses. 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. Figure 1 shows 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 ρ as reported in previous electron-irradiation studies.[18] Both pristine samples have a transition temperature (T_c $=92$ K) and $d\rho/dT$ ($\rho=\rho_o+bT$ $\mu\Omega$ cm, with $b=1.5$ $\mu\Omega$ cm K^{-1}) characteristic of optimally doped Bi2212 crystals.[19] In the case of sample *a* ($\rho_o\sim60\mu\Omega$), an irradiation flux of $6.0\times10^{19}e^-/\mathrm{cm}^2$ leads to a fourfold increase in residual resistivity ($\rho_0\sim$ $240\mu\Omega$ cm) and a 20 K decrease in T_c (73 K). Moreover, a curvature appears in the temperature dependence of resistivity that may suggest a doping change. In order to explore such possibility, we have measured the room-temperature thermopower, $S(300\ \mathrm{K})$, which is a well-known indicator of doping levels in high-T_c superconductors.[20] The determined values of postirradiation samples *a* and *b* (after the third irradiation) are -0.1 ± 0.1 and $5.0\pm0.1\mu$V/K. The values of corresponding unirradiated crystals are -0.8 ± 0.4 and $2.4\pm0.1\mu$V/K, thus in both samples irradiations appear to have caused a slight "under-doping." According to the study on $S(300\ \mathrm{K})$ of Bi2212,[20] ΔT_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 ΔT_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 κ_{ph} to the total heat transport complicates the determination of qp conductivity. As $T\rightarrow0$, κ_{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 κ_{ph} becomes proportional to T^3. Above this regime, κ_{ph} should increase more slowly due to the decrease in phonon mean-free-path. Therefore, by plotting κ/T against T^2, one can resolve the zero-intercept κ_{00}/T corresponding to the residual linear component of κ that is a signature of residual qp conductivity. As seen in Fig. 1(b), there is a finite κ_{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, thus the sizeable uncertainty in κ due to their 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* has become four times larger after irradiation. In fact, the qp lifetime is believed to grow at a rate much faster than T linear below T_c.[21] Thus, the true increase in impurity-scattering rate due to irradiation must be even larger. The amplitude of κ_{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).[22,4] Theory predicts an increase in κ_{00}/T at sufficiently high levels of impurity concentration due to the impurity-induced change in T_c.[23] Therefore, considering that the sample *b* after the third irradiation is nearly twenty times dirtier than the pristine sample *a* (judging from their respective ρ_o values), the "universal" character found in the values of κ_{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 κ_{00}/T into the following equation:[3]

$$\kappa_{00}/T=\frac{k_B^2}{3\hbar}\frac{v_F}{v_2}\frac{n}{d}, \tag{1}$$

where v_F and v_2 are qp Fermi velocity and gap velocity at the nodes in the gap. One then obtains $v_F/v_2=21\pm 3$ that is close to the ratio obtained by ARPES measurements.[8] The same conclusion has been previously drawn by Chiao *et al.*[4]

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 $\kappa_{ph}=1/3\beta\langle v_{ph}\rangle l_{ph}T^3$, where β 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} (Ref. 24) and $v_{ph}=3600$ ms^{-1},[25] κ_{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.*,[4] the ballistic regime appears only below 130 mK. In this case, a downward curvature of the κ/T curve would lead to a slight decrease in the finite intercept and the estimated κ_{00} would become 0.14 mW/K^2 cm [see Fig. 1(a)] but remains within our experimental uncertainty.

Another important aspect of Fig. 1(b) is a sizable irradiation-induced decrease in $\kappa(T)$ for the entire temperature range (0.13 K$<T<0.9$ K). We begin by noting that the typical wavelength of acoustic phonons may be estimated to vary as $\lambda_{ph}=hv_{ph}/k_BT=173$ 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 κ_{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 $T<\gamma$, the impurity bandwidth. Such condition is finally met in sample *b* after the second and the third irradiation where κ is virtually unchanged (see the lower panel of Fig. 1). In relatively cleaner samples, the marked decrease in κ infers that γ must lie below our range of measurements.

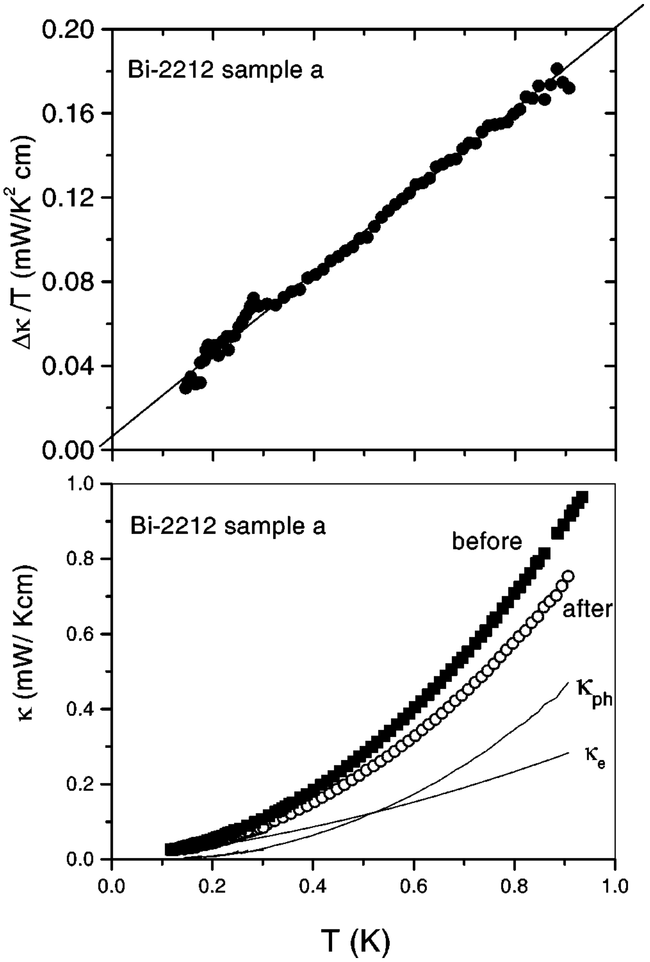

FIG. 2. Upper panel: The change in the thermal conductivity of the sample before and after electron irradiation divided by temperature and plotted as a function of temperature. The straight line is a fit to the data revealing a negligible intercept and a quadratic variation of $\Delta\kappa(T)$. Lower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

Figure 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 that 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 $a=0.005$ mW/K^2 cm and $b=0.19$ mW/K^3 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 γ, the density of states varies linearly with energy[3,4]

$$N(E)=\frac{2}{\pi\hbar^2}\frac{1}{v_Fv_2}E. \tag{2}$$

This leads to a quadratic temperature dependence of specific heat[4]

$$C_e=\frac{18\zeta(3)}{\pi}\frac{k_B^3}{\hbar^2}\frac{n}{d}\frac{1}{v_Fv_2}T^2 \tag{3}$$

where $\zeta(3)\simeq 1.20$ is a numerical factor. Thermal conductivity of these excitations can be estimated via kinetic theory; $\kappa_e=\frac{1}{3}C_ev_F^2\tau_e$. Here, τ_e is the electronic scattering time. The temperature dependence of τ_e dictates the temperature dependence of electronic thermal conductivity, $\kappa_e\propto T^2\tau_e(T)$. The relative weight of this term to the universal linear term can be estimated to be

$$\frac{\kappa_e}{\kappa_{00}}=\frac{18\zeta(3)k_B\tau_eT}{\pi\hbar}. \tag{4}$$

Note that while κ_{00}/T is universal, κ_e is not: its magnitude decreases with increasing disorder. The quadratic dependence of κ_e on temperature suggests that τ_e in this temperature range is nearly T independent. The size of the quadratic term coefficient appearing in $\Delta\kappa(T)(b=0.19\ \mathrm{mW/K^3\,cm})$ corresponds to an electronic scattering time of 1.3 ps. On the other hand, qp lifetime can be estimated from $\rho=m^*/ne^2\tau_e$ and $\omega_p=(4\pi ne^2/m^*)^{1/2}$, where ω_p is the Drude plasma frequency. Using $\omega_p=1.1$ eV,[22] $\rho(100\ \mathrm{K})=200\ \mu\Omega\,\mathrm{cm}$ and $\rho_0=60\ \mu\Omega\,\mathrm{cm}$, one finds $\tau_e(100\ \mathrm{K})\simeq 0.05$ ps and $\tau_e(0\ \mathrm{K})\simeq 0.2$ 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.[13] It is instructive to compare the magnitude of τ_e in Bi2212 and Y123. In the latter system, τ_e of high-quality crystals is of the order of 7 ps (Ref. 15) and is reported to approach 20 ps (Ref. 26) in BaZrO$_3$-grown crystals. For Bi2212, recent study on the complex conductivity by Corson *et al.* showed the qp lifetime to approach ~ 1 ps (Ref. 27) at low temperatures, similar to the value calculated independently from our measurements. Further, assuming the scattering rate to remain constant for $T>1$ K,[26] one can estimate the size of κ_e upward in temperature. The deduced value at 5 K (~ 4.8 mW/K cm) can be compared via the Wiedemann-Franz law to the available data on charge conductivity that is limited to $T>5$ K.[13] This yields $(\kappa_e/\sigma_1 T)(5\ \mathrm{K})\sim 21.3\ \mathrm{nW}/\Omega\,\mathrm{K}$, very close to the Sommerfeld value ($L_0=24.5\ \mathrm{nW}/\Omega\,\mathrm{K}$). 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 τ_e. In the unitary limit, for example, this leads to a cubic (instead of a quadratic) behavior for finite-energy qp transport for $T>\gamma$.[7] Second, the size of γ strongly depends on the impurity density (n_{imp}) as well as the scattering phase shift δ. In the unitary ($\delta=\pi/2$) limit, γ increases substantially with increasing scattering rate, Γ; $\gamma\sim(\Gamma\Delta_0)^{1/2}$. In the Born ($\delta=0$) limit, its enhancement with Γ is largely attenuated for $\Gamma\ll T_c$; $\gamma\sim\Delta_0\exp(-\Delta_0/\Gamma)$. Here $\Gamma=1/2\tau_e$ and Δ_0 is the magnitude of the superconducting gap.[6] Now, with $\tau_e\sim 1$ ps (which would yield $\Gamma\sim 3$ K) and $\Delta_0\sim 40$ meV,[28] γ may be estimated to be 35 K in the unitary limit and virtually zero in the Born limit. Thus, the size of γ 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,[29] however, preliminary investigations reveal that a reasonable phase shift only cannot account for the magnitude of the excess conductivity observed here.[30] 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$_2$Sr$_2$CaCu$_2$O$_8$. 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 an 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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