Abrupt transition in quasiparticle dynamics at optimal doping in a cuprate superconductor system

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Abstract

We report time-resolved measurements of the photoinduced change in reflectivity, $\Delta R$, in the Bi$_2$Sr$_2$Ca$_{1-y}$Dy$_y$Cu$_2$O$_{8+\delta}$ (BSCCO) system of cuprate superconductors as a function of hole concentration. We find that the kinetics of quasiparticle decay and the sign of $\Delta R$ both change abruptly where the superconducting transition temperature $T_c$ is maximal. These coincident changes suggest that a sharp transition in quasiparticle dynamics takes place precisely at optimal doping in the BSCCO system.
Pump and probe methods in optical spectroscopy have opened a new window on the properties of quasiparticles in cuprate superconductors and other highly correlated electron systems [1]. In experiments based on these methods, ultra-short pump pulses inject quasiparticles at densities that are continuously variable from well above to well below the thermal equilibrium level. Time-delayed probe pulses measure changes in the reflectivity or transmissivity that result from the presence of nonequilibrium quasiparticles, providing information about their recombination rates, transport, and optical properties. These studies have been carried out extensively in the cuprate superconductors, yielding a rich, complex, yet poorly understood array of experimental observations. One of the central observations, and possibly the most puzzling, has been the behavior of the quasiparticle recombination rate, \( \gamma \), as a function of temperature, \( T \), and photoinjected density, \( \Delta n_{ph} \). Two classes of behavior are found: in class (1) \( \gamma \) appears to vanish as \( T \rightarrow 0 \) and \( \Delta n_{ph} \rightarrow 0 \) [2, 3, 4, 5, 6], while in class (2) \( \gamma \) remains essentially constant with decreasing \( T \) [9, 10, 11] and \( \Delta n_{ph} \) [12]. Another, seemingly distinct, puzzle concerns the sign of the photoinduced change in sample reflectivity, \( \Delta R \), which can be either positive or negative [2, 9, 13, 14, 15].

Here we report measurements of \( \Delta R \) and \( \gamma \) in the Bi\(_2\)Sr\(_2\)Ca\(_{1-y}\)Dy\(_y\)Cu\(_2\)O\(_{8+d}\) (BSCCO) system of cuprate superconductors as a function of hole concentration, \( x \), that considerably clarify the conditions under which these behaviors appear. As discussed below, the key to successfully exploring the BSCCO system was to eliminate the effects of laser-induced heating. Once this is accomplished, we find that the dynamics change from class (1) to (2) at exactly \( x_m \), the value for which the superconducting transition temperature \( T_c \) is maximal. Moreover, we find that the sign of \( \Delta R \) reverses at \( x_m \) as well. These coincident changes suggest that an abrupt transition in quasiparticle dynamics takes place precisely at optimal doping in the BSCCO system.

Time-resolved optical spectroscopy was performed using pump and probe pulses of photon energy 1.5 eV and duration 80 fs from a mode-locked Ti:Sapphire oscillator. Because the BSCCO crystals are optically thick at the laser wavelength of 820 nm, the changes in optical response were probed by measuring the reflected probe power. Fig. 1 is a plot of the initial reflectivity change, \( \Delta R \), normalized to the reflectivity \( R \), as a function of the energy per area, \( \Phi_L \), deposited by each pump pulse. The underdoped (\( T_c=71 \) K) sample was thermally anchored to a Cu plate maintained at 5 K. The \( \Delta R \) values plotted as open symbols were measured using the full repetition rate of the oscillator, which is 90 MHz. At this repetition
rate, the slope of $\Delta R/R$ vs. laser intensity changes abruptly when $\Phi_L$ reachess 0.8 $\mu$J/cm$^2$. This effect was not observed in studies of underdoped YBa$_2$Cu$_3$O$_{6.5}$ crystals with similar values of $T_c$ and the same conditions of photoexcitation [5, 8].

The decrease in $\Delta R/R$ with increasing $\Phi_L$ occurs when photoexcitation begins to drive the surface of the sample into the normal state. The origin of this effect is the steady state increase in the surface temperature of the sample due to laser-induced heating. The effects of surface heating are more severe than in the YBCO system because c-axis thermal conductivity of BSCCO is much smaller [16]. To overcome the laser-heating problem, we inserted an acousto-optic pulse-picker at the laser output. The solid symbols in Fig. 1 indicate values of $\Delta R/R$ obtained when the pulse-picker reduces the 90 MHz pulse repetition rate of the laser to 5 MHz (at the same time the diameter of the illuminated area on the sample was reduced from 75 to 30 $\mu$). When the average power is thus reduced (by a factor of $\sim 100$) the discontinuity in the slope of $\Delta R$ vs. $\Phi_L$ disappears and the growth of $\Delta R/R$ with $\Phi_L$ is essentially linear. Further reduction of the repetition rate to 2.5 MHz produced no further changes in the either the amplitude or the subsequent decay of $\Delta R/R$, indicating that effects of laser heating are negligible at this power level.

Eliminating problems associated with laser heating makes it possible to investigate the
nonequilibrium state of the BSCCO family of superconductors at low temperature. In this work we studied eight BSCCO crystals whose \( T_c \)'s range from 42 K (underdoped) to 77 K (overdoped). To control hole concentration over a wide range, we combined both oxygen tuning and Dy-doping. We achieved the target \( T_c \)'s by floating-zone growth of three different kinds of single crystals with the following Bi:Sr:Ca:Dy ratio: (a) 2.10 : 1.91 : 1.03 : 0, (b) 2.11 : 1.88 : 0.79 : 0.23, and (c) 2.13 : 1.82 : 0.70 : 0.36. The crystals were subsequently annealed for 2 to 14 days, depending on the annealing temperature, in air or an argon+oxygen environment.

We now describe the evolution of the photoinduced \( \Delta R \) with hole concentration. Fig. 2 presents an overview of the changes, showing \( \Delta R \) (at \( \Phi_L = 0.9 \mu J/cm^2 \)) as a function of time for three representative samples: underdoped (\( T_c = 71 \) K), optimally-doped (\( T_c = 94.5 \) K), and overdoped (\( T_c = 77 \) K). \( \Delta R \) changes from positive for the underdoped sample to negative for the overdoped sample. In the sample with \( T_c = 94.5 \) K, \( \Delta R \) is a superposition of signals of both sign, indicating that the crossover takes place precisely at optimal doping.

Figs. 3(a) and 3(b) illustrate the crossover in kinetics that takes place at the same hole concentration at which \( \Delta R \) changes sign. The top panels show \( \Delta R(t)/R \) at different values of \( \Phi_L \), for the same under and overdoped samples as in Fig. 2. In order to compare the decay rates at different \( \Phi_L \), we scale the amplitude to the same value near time zero. For each
FIG. 3: Top panels: Fractional change in reflectivity as a function of time for an (a) underdoped ($T_c=71$ K) and (b) overdoped sample ($T_c=77$ K), for several values of the pump fluence in the range from 0.1 to 1.0 µJ/cm$^2$. The plots are scaled to have the same value at $t = 0$, illustrating that the decay rate depends on fluence in underdoped samples but not in overdoped samples. Bottom panels (c) and (d): Initial decay rate as a function of initial $\Delta R/R$ for the curves directly above.

set of curves, a plot of the initial decay rate, $\gamma(0)$, vs. $\Phi_L$ appears below. In underdoped samples $\gamma(0) \propto \Phi_L$, with essentially zero intercept (we estimate an experimental uncertainty in $\gamma(0)$ of 0.005 ps$^{-1}$, approximately $10^{-2}$ of the maximum $\gamma(0)$ of 0.6 ps$^{-1}$). The adjacent panels illustrate that the kinetics of photoexcitations in overdoped samples are substantially different - the decay rate of the excited state remains large as $\Phi_L$ is lowered.

The abruptness of the change in sign and decay rate of $\Delta R$ with hole concentration, $x$, is shown Figs. 4(a) and 4(b). Fig. 4(a) is a plot of $\Delta R$ vs. $x$ measured at 0.2 ps after arrival of the pump pulse. The values of $x$ were obtained from the $T_c$’s using the empirical formula given in Ref. [17]. Fig. 4(b) shows the initial decay rate of $\Delta R$ for the same set of samples, measured at $\Phi_L = 0.3$ µJ/cm$^2$. At this relatively low $\Phi_L$, the lifetime changes by a factor $\sim 50$ as $x$ varies from just below to just above $x_m$. In the following, we discuss the origin of
FIG. 4: Fractional change in reflectivity (top panel) and initial lifetime (bottom panel) as a function of hole concentration. All data measured at pump laser fluence 0.3 $\mu J/cm^2$. The collapse of the quasiparticle lifetime and sign change of $\Delta R$ both occur at optimal doping.

the two transitions, starting with the sign of $\Delta R$.

To analyze the sign change of $\Delta R$ we consider the change in the real part of the dielectric function, $\Delta\varepsilon_1$, caused by the presence of nonequilibrium excitations. In general, a change in the occupation of states shifts optical spectral weight from one frequency range to another, conserving the total weight. The sign of $\Delta\varepsilon_1$ caused by such shifts depends on the relative ordering of the measurement frequency, $\omega_0$, and frequencies where spectral weight is lost and gained. For a superconductor described by BCS theory, an increase in the number density of quasiparticles shifts spectral weight from the condensate $\delta$-function at $\omega = 0$ to frequencies near the quasiparticle scattering rate, $1/\tau$, or the gap $\Delta$, in the clean, or dirty limit, respectively. Assuming that $\omega_0 \gg \max\{\Delta, 1/\tau\}$, the Kramers-Kronig relations imply that $\Delta\varepsilon_1 \approx -(8/\omega_0^4) \int d\omega \Delta\sigma_1(\omega)\omega^2$, where $\Delta\sigma_1$ is spectrum of the conductivity that was removed from the $\delta$-function. As $\Delta\sigma_1$ is positive, the resulting $\Delta\varepsilon_1$ must be negative.
From the Fresnel equation and literature values \[\epsilon(\omega)\] for \(\epsilon(\omega)\) we obtain \(R^{-1}\partial R/\partial \epsilon_1 = 0.02\) at the measurement energy of 1.5 eV. Thus the spectral weight shifts expected for a BCS superconductor correspond to \(\Delta R < 0\) and can account for the photoinduced response in overdoped, but not underdoped BSCCO \cite{19, 20}.

To obtain \(\Delta \epsilon_1\) of the opposite sign requires that a fraction of the spectral weight removed from the \(\delta\)-function either shifts to \(\omega \gg \omega_0\), or becomes broad on the scale of \(\omega_0\). In other words, some spectral weight must be distributed on the scale of the electronic bandwidth, rather than the frequency scale of \(1/\tau\) or \(\Delta\). In this case \(\Delta \epsilon_1\) acquires a positive contribution, given approximately by \(8A/\omega_0^2\), where \(A\) is the portion of the spectral weight removed from the condensate that is shifted to high frequencies \cite{21}. Using this relation, we can infer the value of \(A\) that generates a given positive value of \(\Delta R\). To express \(A\) in a fashion that can be compared with other experiments, we convert conductivity spectral weight to the electron kinetic energy \cite{22}, \(i.e.\) \(A \equiv \int \sigma d\omega = (\pi e^2/2\hbar^2 d)\langle -T \rangle\). The largest \(\Delta R\) that we obtain, \(\sim 4 \times 10^{-4}\), corresponds to \(\langle -\Delta T \rangle = 3\) meV. We note a possible connection between the photoinduced \(\Delta \epsilon_1\) and the thermally induced \(\Delta \epsilon_1\) recently reported in BSCCO \cite{21}. When spectral weight is removed from the \(\delta\)-function by raising the temperature, \(\Delta \epsilon_1\) in the near-infrared is positive and similar in magnitude to the photoinduced changes reported here.

We next discuss the discontinuous change in decay rate that takes place at the same hole concentration at which \(\Delta R\) changes sign. The sudden change in rate at \(x_m\) marks the transition from \(\gamma(0) \propto \Phi_L\) on the underdoped side of the phase diagram and to \(\gamma(0)\) independent of \(\Phi_L\) on the overdoped side. For underdoped samples, the decay rate is a linear function of the density of photoinduced excitations. Such "second-order kinetics" occurs when individual, isolated excitations are stable (or metastable) and the rate of decay is limited by the frequency of two-quasiparticle encounters. The transition to a \(\Phi_L\)-independent decay rate at optimal doping indicates that excitations that were metastable on the underdoped side of the phase diagram become unstable on the overdoped side. Below, we discuss the nature of these excitations and potential explanations for the metastable-unstable transition.

The linear dependence of \(\Delta R\) on \(\Phi_L\) suggests that the excitations that give rise to \(\Delta R\) are not nodal quasiparticles. More precisely, it suggests that photoexcited quasiparticles do not immediately thermalize towards the nodes, establishing a degenerate Fermi-Dirac distribution with an effective chemical potential or temperature. If this were the case, the
spectral weight removed from the condensate would be proportional to $\Delta n_{ph}^{1/2}$ and $\Delta R$ would be proportional to $\Phi_L^{1/3}$ \cite{23}. The experimental observation that instead $\Delta R \propto \Phi_L$ suggests that photoexcited quasiparticles form a nondegenerate gas, as do quasiparticles injected in an s-wave superconductor. The scenario that emerges is that injected quasiparticles cascade to the antinodal regions of the Brillouin zone where the density of states is very large. In underdoped samples they remain there, prevented from thermalizing further (for reasons we speculate on below) until an encounter with another quasiparticle. The rapid, $\Delta n_{ph}$-independent decay of $\Delta R$ that appears at optimal doping may indicate that the bottleneck for thermalization of antinodal particles disappears above $x_m$.

The scenario described above bears a certain relationship to ARPES investigations \cite{24} of the quasiparticle self-energy as a function of $x$, particularly to the recent emphasis on a "dichotomy" between nodal and antinodal excitations \cite{25,26,27}. The dichotomy exists on the underdoped side, where excitations near the node are coherent quasiparticles and antinodal excitations are incoherent. With increasing $x$ the quasiparticle coherence extends further from the node and appears to encompass the entire Fermi surface in overdoped samples. We speculate that the bottleneck in underdoped materials exists because the incoherent nodal excitations cannot readily convert to coherent nodal excitations. When the entire FS becomes coherent the bottleneck is removed.

Further evidence that the excitations in underdoped samples are incoherent comes from the second-order recombination coefficient, $\beta$, defined such that $\gamma(0) = \beta \Delta n_{ph}$ \cite{28}. To estimate $\beta$ we assume that each photon creates $\hbar \omega_0 / \Delta$ particles, where $\Delta$ is the antinodal creation energy of $\sim 35$ meV. Converting photon to quasiparticle number as above, we obtain $\beta \approx 0.1 \text{ cm}^2/\text{s}$ for all the underdoped samples measured in this study. If a particle is incoherent, its bandlike motion is frustrated and the rate of recombination can become limited by the time needed to diffuse the average distance between them. For a 2D "diffusion-limited reaction" process $\beta \approx D$. It is therefore interesting to compare $\beta$ with the quantum diffusion of a fermion in 2D. At the localization limit $k_F l = 1$, $D = 1/h N_F$ or $h/2m^*$, where $N_F$ and $m^*$ are the Fermi level density of states and effective mass, respectively. If we take $m^* = 3m$ as suggested from optical measurements \cite{29}, then $D = 0.15 \text{ cm}^2/\text{s}$, which is remarkably close the experimental estimate for $\beta$.

In conclusion, we have observed an abrupt change in quasiparticle decay rate and the sign of the photoinduced $\Delta R$ in BSCCO superconductors precisely at optimal doping. On
the underdoped side, the photoinjected quasiparticles appear to propagate incoherently and cause some condensate spectral weight to shift to very high frequencies. The sign of spectral weight shift is consistent with recent suggestions of a kinetic-energy driven transition to the superconducting state \[21\]. The sudden change in the quasiparticle dynamics at \(x_m\) may signal the onset of antinodal quasiparticle coherence in optimal and overdoped samples. The change in the sign of \(\Delta R\) at \(x_m\) suggests that reduction in kinetic energy with the onset of superconductivity occurs primarily in the underdoped regime.

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[1] R. D. Averitt and A. J. Taylor, J. Phys.: Condens. Matter 14, R1357 (2002).
[2] P. Gay, D. C. Smith, C. J. Stevens, C. Chen, G. Yang, S. J. Abell, D. Z. Wang, J. H. Wang, Z. F. Ren, and J. F. Ryan, J. Low Temp. Phys. 117, 1025 (1999).
[3] D. C. Smith, P. Gay, C. J. Stevens, D. Z. Wang, J. H. Wang, Z. F. Ren, and J. F. Ryan, J. Low Temp. Phys. 117, 1059 (1999).
[4] J. Demsar, R. Hudej, J. Karpinski, V. V. Kabanov, and D. Mihailovic, Phys. Rev. B 63, 054519 (2001).
[5] G. P. Segre, N. Gedik, J. Orenstein, D. A. Bonn, R. X. Liang, and W. N. Hardy, Phys. Rev. Lett. 88, 137001 (2002).
[6] M. L. Schneider, J. Demsar, Y. Glinka, A. Klimov, A. Krapf, S. Rast, Y. H. Ren, W. D. Si, Y. Xu, X. H. Zeng, et al., Europhys. Lett. 60, 460 (2002).
[7] N. Gedik, J. Orenstein, R. X. Liang, D. A. Bonn, and W. N. Hardy, Science 300, 1410 (2003).
[8] N. Gedik, P. Blake, R. C. Spitzer, J. Orenstein, R. X. Liang, D. A. Bonn, and W. N. Hardy, Phys. Rev. B 70, 014504 (2004).
[9] S. G. Han, Z. V. Vardeny, K. S. Wong, O. G. Symko, and G. Koren, Phys. Rev. Lett. 65, 2708 (1990).
[10] R. D. Averitt, G. Rodriguez, A. I. Lobad, J. L. W. Siders, S. A. Trugman, and A. J. Taylor, Phys. Rev. B 63, 140502 (2001).
[11] J. Demsar, B. Podobnik, V. V. Kabanov, T. Wolf, and D. Mihailovic, Phys. Rev. Lett. 82, 4918 (1999).
[12] W. Albrecht, T. Kruse, K. Leo, and H. Kurz, Appl. Phys. A 57, 203 (1993).
[13] G. L. Eesley, J. Heremans, M. S. Meyer, G. L. Doll, and S. H. Liou, Phys. Rev. Lett. 65, 3445 (1990).
[14] D. H. Reitze, A. M. Weiner, A. Inam, and S. Etemad, Phys. Rev. B 46, 14309 (1992).
[15] D. Dvorsek, V. V. Kabanov, J. Demsar, S. M. Kazakov, J. Karpinski, and D. Mihailovic, Phys. Rev. B 66, 020510 (2002).
[16] M. F. Crommie and A. Zettl, Phys. Rev. B 43, 408 (1991), part A.
[17] M. R. Presland, J. L. Tallon, R. G. Buckley, R. S. Liu, and N. E. Flower, Physica C 176, 95 (1991).
[18] J. Hwang, T. Timusk, and G. D. Gu, Nature 427, 714 (2004).
[19] J. Demsar, R. D. Averitt, V. V. Kabanov, and D. Mihailovic, Phys. Rev. Lett. 91, 169701
[20] N. Gedik, J. Orenstein, R. Liang, D. A. Bonn, and W. N. Hardy, Phys. Rev. Lett. 91, 169702 (2003).

[21] H. J. A. Molegraaf, C. Presura, D. van der Marel, P. H. Kes, and M. Li, Science 295, 2239 (2002).

[22] R. Kubo, J. Phys. Soc. Japan 12, 570 (1957).

[23] J. P. Carbotte and E. Schachinger, Phys. Rev. B 70, 014517 (2004).

[24] A. Damascelli, Z. Hussain, and Z. X. Shen, Rev. Mod. Phys. 75, 473 (2003).

[25] T. Yoshida, X. J. Zhou, T. Sasagawa, W. L. Yang, P. V. Bogdanov, A. Lanzara, Z. Hussain, T. Mizokawa, A. Fujimori, H. Eisaki, et al., Phys. Rev. Lett. 91, 027001 (2003).

[26] X. J. Zhou, T. Yoshida, D. H. Lee, W. L. Yang, V. Brouet, F. Zhou, W. X. Ti, J. W. Xiong, Z. X. Zhao, T. Sasagawa, et al., Phys. Rev. Lett. 92, 187001 (2004).

[27] K. M. Shen, F. Ronning, D. H. Lu, F. Baumberger, N. J. C. Ingle, W. S. Lee, W. Meevasana, Y. Kohsaka, M. Azuma, M. Takano, et al., Science 307, 901 (2005).

[28] A. Rothwarf and B. N. Taylor, Phys. Rev. Lett. 19, 27 (1967).

[29] Y.-S. Lee, K. Segawa, Y. Ando, and D. N. Basov, Phys. Rev. B 70, 014518 (2004).