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Femtosecond Optical Detection of Quasiparticle Dynamics in Single-Crystal $Bi_2Sr_2CaCu_2O_{8+\delta}$ *

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Quasiparticle dynamics of an optimally doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal is investigated by the femtosecond pump–probe technique. Temperature dependences of amplitude of the photoinduced differential reflectivity and the relaxation time show the evidence of strong phonon bottleneck. The experimental results are analysed by the Rothwarf–Taylor model.

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In recent years, the femtosecond pump–probe technique has been widely utilized to study nonequilibrium carrier dynamics in cuprate superconductors.[1−6] In experiments, a pump pulse creates nonequilibrium quasiparticles (QP) through photoexcitation. These nonequilibrium QPs rapidly thermalize through electron–electron ($e-e$) and electron–phonon ($e-ph$) interactions and then reach states near the gap edge. The subsequent relaxation and recombination dynamics can be observed by measuring changes in reflectivity or transmission of a time-delayed probe pulse. Since the dynamics of nonequilibrium QP is extremely sensitive to the presence of a gap, it is capable of giving direct information about the low-lying electronic structure and temperature dependence of the QP lifetime. Compared to the frequency-domain optical spectroscopy, such as infrared reflectivity and Raman spectroscopy, the pump–probe technique enables the separation of different spectral components by their lifetime, therefore, can resolve long-standing controversial issues regarding the origin of the different components in the low-energy spectra of the cuprates.

Recently, Gay *et al.*[1] have reported the measurement of femtosecond dynamics of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO). They found that it is impossible to fit the carrier dynamics of BSCCO by a simple model although the bi-particle recombination can explain some of the results. To clarify the nonequilibrium QP dynamics in BSCCO, in this Letter we report femtosecond time-resolved experiments in an optimally doped BSCCO single crystal. It is found that temperature dependences of amplitude of the photoinduced differential reflectivity and the relaxation time show the evidence of strong phonon bottleneck.

Our optimally doped BSCCO single crystals were grown from a mixture of Bi_2O_3, CuO, $SrCO_3$, and $CaCO_3$ by the travelling solvent floating zone method with a transition temperature of 89 K.[7] Small pieces of these crystals with a typical size of $0.6 \times 0.6 \times 0.05\,\mathrm{mm}^3$ were glued onto Si substrates, which were mounted on a cold finger of a liquid-helium continuous-flow optical cryostat.

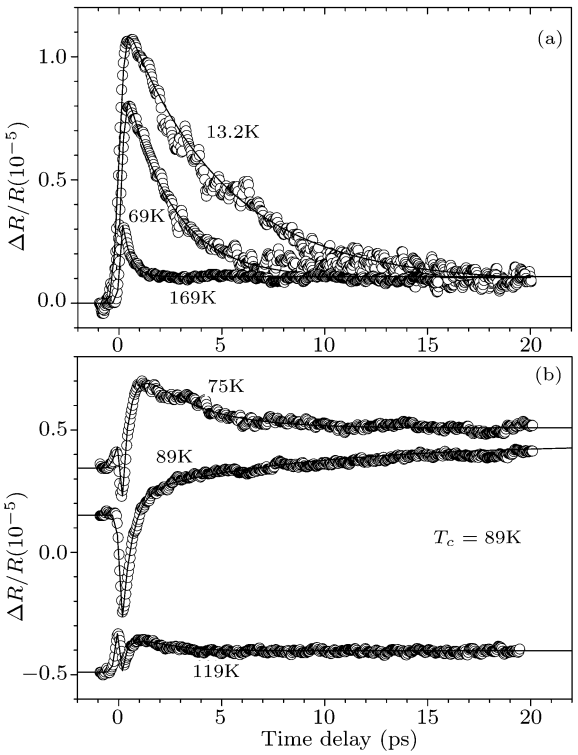

Fig. 1. Photoinduced change in reflectivity $\Delta R/R$ as a function of time delay at different temperatures. The solid curves are theoretical fits to the data.

The time-resolved measurements were performed using the standard pump–probe technique, with a commercial Ti:sapphire mode-locked laser, which pro-

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duced 100-fs-wide pulses at the wavelength 790 nm at repetition rate 82 MHz. The average energy of the pump and probe pulses are 0.1 and 0.02 nJ, respectively. The diameter of both the beams was 100 μm and the surface was parallel to the *ab* crystal plane. Preliminary measurements on these samples revealed that the pump pulses increase the temperature of probed spot by about 9 K which is attributed to the much lower thermal conductivity of BSCCO, particularly in the *c*-axis direction.[8] In order to extract $\Delta R/R$, the train of the pump pulses was modulated at 100 kHz by an acousto-optic modulator, and the output of the photodiode was sent to an digital lock-in amplifier for phase sensitive detection of the 100 kHz component. To avoid coherent artifacts and also to reduce the scattering of pump beam into the detector, the pump and probe beams were cross polarized. Our instrumental limit for the smallest observable change in optical reflectivity is about 10^{-7}.

The data on the time dependence of the photoinduced change in reflectivity $\Delta R/R$ at different temperatures is shown in Fig. 1. The dynamics shows three distinct temperature regions. The sign of $\Delta R/R$ is positive and the signal shows a single exponential decay with decay time τ_1 when the temperature T is below 72 K or above 129 K [see Fig. 1(a)]. On the other hand, the temporal behaviour of $\Delta R/R$ becomes complicated when 72 K$< T <$ 129 K [Fig. 1(b)]. To analyse these data, we decomposed the total signals, using the nonlinear least squares fitting method, into separate single-exponential processes (Fig. 2). For the single exponential decay, $\Delta R/R$ is corresponding to the solution of the equation

$$\frac{d(\Delta R/R)}{dt} = g(t) - \frac{\Delta R/R}{\tau}, \qquad (1)$$

where $g(t) = g_0 \exp(-2t^2/\tau_p^2)$ represents the photoexcitation with Gaussian pulse of width τ_p, and τ refers to the relaxation time of QP. The solid lines in Fig. 2 are superposition of these single-exponential relaxation processes. It is found that in addition to a positive relaxation process of decay time τ_1 (dashed curve), there exists a negative sub-picosecond relaxation process of decay time $\tau_2 \approx 500$ fs (dotted curve), which is nearly independent of temperature. Moreover, near T_c another negative relaxation process with a lifetime $\tau_3 \approx 9$ ps (dash-dotted curve) appears and disappears quickly as $T > T_c$. We note that in fitting the data we have to take into account the long-lived background especially when $T > 129$ K, which has a lifetime longer than 12 ns.

Figure 3 presents the temperature-dependent amplitude $\Delta R/R$ and relaxation time τ_1 of the positive picosecond component, while those of the sub-picosecond negative component are presented in the insets. The amplitude of the positive picosecond component [Fig. 3(a)] is almost constant at low temperatures followed by a rapid decrease as T_c is approached, while it diminishes systematically with increasing temperature above T_c. On the other hand, the amplitude of the sub-picosecond negative component increases as the temperature increases, reaching the maximum at T_c, and then decreases and disappears with further increase of temperature.

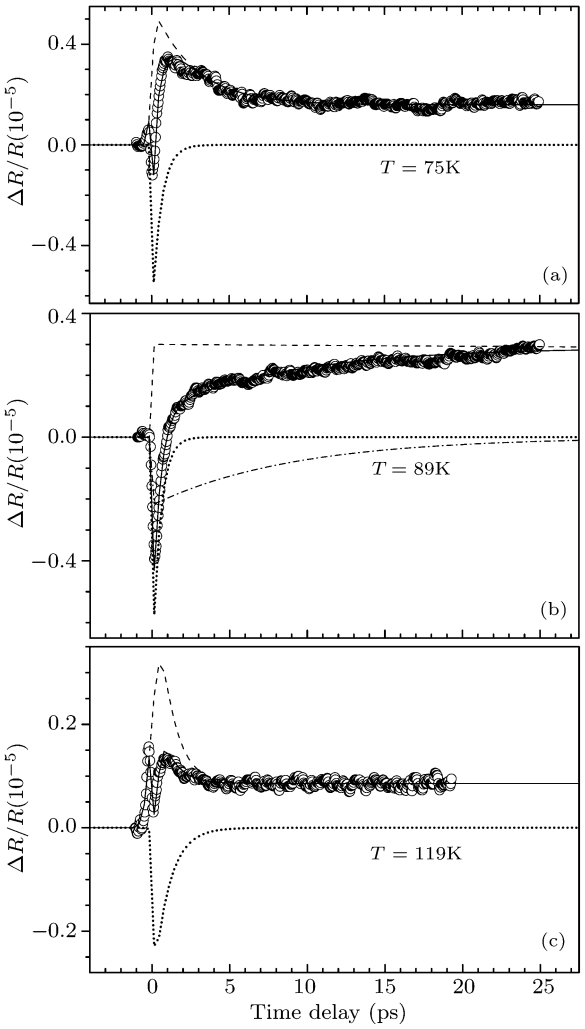

Fig. 2. Photoinduced change in reflectivity $\Delta R/R$ at (a) 75 K, (b) 89 K and (c) 119 K decomposed into constituting single exponential relaxation processes. The dashed, dotted and dash-dotted curves represent the positive, sub-picosecond negative and 9 ps negative components, respectively. The solid lines are superpositions of these single-exponential processes.

Now, we analyse the experimental results. It is generally believed that for most conventional superconductors as well as cuprates, the nonequilibrium QP dynamics is governed by phonon bottleneck mechanism.[9] In the strong bottleneck regime, two QPs with energies larger than Δ recombine to create a high frequency phonon (HFP) with energy larger than 2Δ. Here, Δ is the gap magnitude. If the relaxation of

HFP is slow, an HFP can subsequently break a Cooper pair and create two QPs, causing bottleneck for the QP relaxation. As a result, the recovery dynamics is governed by the lifetime of the 2Δ phonon.

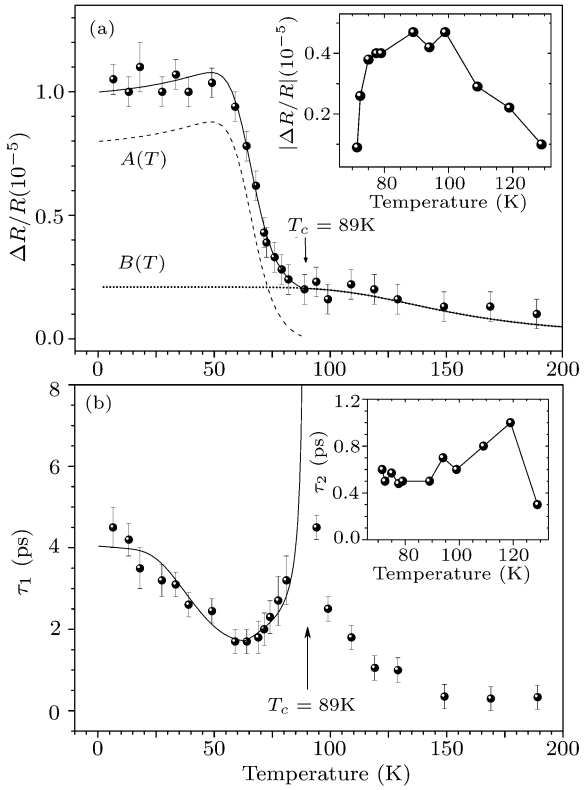

Fig. 3. (a) Temperature dependence of the amplitude $\Delta R/R$ of the positive and sub-picosecond negative (inset) components. The fit (solid curve) is made using the sum of $A(T)$ (dashed curve) and $B(T)$ (dotted curve). (b) Temperature dependence of relaxation time of the positive and sub-picosecond negative (inset) components. The solid curve is the theoretical fit.

We first discuss the temperature dependence of the amplitude of the positive picosecond component. Let us assume that the positive component of $\Delta R/R$ is caused by the excited-state absorption of the probe pulse from photoexcited QP states and therefore, is proportional to the photoexcited carrier density n_{pi}. In the limit of small n_{pi}, the temperature dependence of the amplitude $\Delta R/R$ for an isotropic BCS-like T-dependent gap $\Delta_c(T)$ and a T-independent gap Δ_p are given by[2]

$$A(T) \propto \frac{\varepsilon/[\Delta_c(T) + k_B T/2]}{1 + \zeta\sqrt{2k_B T/\pi\Delta_c(T)}\exp[-\Delta_c(T)/k_B T]}, \tag{2}$$

$$B(T) \propto \frac{\varepsilon/\Delta_p}{1 + \zeta\exp(-\Delta_p/k_B T)}, \tag{3}$$

where ζ is a constant and ε is the pump laser intensity per unit cell. As shown in Fig. 3(a), we can obtain a good fit to the temperature dependence of $\Delta R/R$ by assuming the coexistence of these two gaps. The values of $\Delta_c(0)$ and Δ_p obtained from the best fit are $4k_B T_c$ and $9k_B T_c$, respectively. The similar two-gap behaviour is observed on $Tl_2Ba_2Ca_2Cu_3O_{10}$,[10] $YBa_2Cu_3O_{7-\delta}$[2] and $HgBa_2Ca_2Cu_3O_{8+\delta}$.[3]

Now, we turn to the Rothwarf–Taylor (RT) model,[11] which describes the evolution of QP and HFP populations during the superconductor recovery. Recently, Kabanov *et al.*[9] have obtained analytical solutions of the model in the limit of a strong and a weak bottleneck. According to the RT model, the evolution of QP and HFP populations is determined by

$$\frac{dn}{dt} = \eta N - Rn^2, \tag{4}$$

$$\frac{dN}{dt} = -\frac{\eta N}{2} + \frac{Rn^2}{2} - \gamma(N - N_T), \tag{5}$$

where n and N are the concentrations of QP and HFP, respectively; η is the probability for pair breaking by HFP; R is the QP recombination rate; N_T is the concentration of HFP in thermal equilibrium; and γ is the decay rate of HFP. From Eqs. (4) and (5), we obtain

$$d(2N + n)/dt = -2\gamma(N - N_T). \tag{6}$$

Under the condition of strong bottleneck (i.e. $\gamma \ll \eta$), we can assume that during the recovery process, QP and HFP are in quasi-equilibrium, therefore we have $\eta N \approx Rn^2$. Also, the thermal equilibrium concentrations of HFPs and QPs (n_T) satisfy the detailed balance $\eta N_T \approx Rn_T^2$. Then, Eq. (6) becomes

$$\left(4n + \frac{\eta}{R}\right)\frac{dn}{dt} = -2\gamma(n^2 - n_T^2). \tag{7}$$

We define the initial relaxation rate as $\tau^{-1} = |dn(t)/dt|/(n_s - n_T)$ at $t \to 0$.[12] Considering the weak photoexcitation case ($n \approx n_T$), in the low temperature limit ($4N_T \ll \eta/R$), we have[9]

$$\tau^{-1} = \frac{2R\gamma}{\eta}(n_s + n_T), \tag{8}$$

where n_s is the quasistationary concentration of QPs preceding the relaxation. Since the signal amplitude $A \propto n_s - n_T$ and $n_T(T) \propto \sqrt{\Delta_c(T)T}\exp[-\Delta_c(T)/T]$,[2] assuming R, γ and η to be temperature independent, then Eq. (8) can be rewritten as

$$\tau_1^{-1}(T) = C\{DA(T) + E\sqrt{\Delta_c(T)T} \cdot \exp[-\Delta_c(T)/T]\}, \tag{9}$$

where C, D and E are the fitting parameters. The above expression can fit the data only up to about $0.7T_c$ and fail to reproduce the upturn of τ_1 near T_c. This is because γ is strongly dependent on the temperature near T_c. Specifically, near T_c the relaxation is governed by the anharmonic decay time of HFPs,

where the relaxation rate is proportional to $\Delta_c(T)$,[2,3] therefore, due to the close superconducting gaps the relaxation time τ_1 becomes divergent. In our experiment we obtain $\tau_1 \approx 150$ ps and 1000 ps at $T = 84$ K and 89 K, respectively [not shown in Fig. 3(b)]. We include this T-dependence of γ into $C(T)$ so that $C(T) \propto \Delta_c(T)$ when $0.7T_c < T < T_c$. The solid curve in Fig. 3(b) is the theoretical fit, from which we obtain $2\Delta_c(0) = 1.7k_BT_c$. Finally, we note that the relaxation time above T_c is much longer than expected for metallic relaxation, indicating the presence of a T-independent pseudogap Δ_p. The value of $\Delta_c(0)$ extracted from fitting the T dependence of the relaxation time is somewhat lower than the value extracted from the fit to the amplitude of the positive component. Similar things occur in fitting the data of MgB_2 and $Tl_2Ba_2CuO_{6+\delta}$. Reasons for this inconsistency have been discussed by Kabanov *et al.*[9]

The present measurements show that there exist negative signals around the transition temperature (72 K$< T <$ 129 K). The sign of $\Delta R/R$ is related to the mechanism of the optical response of the probe beam and depends on the details of the electronic band structure. The photoinduced probe signal can also originate from photoexcitation transfer of spectral weight from the condensate δ function at $\omega = 0$ to higher frequency.[4,13] In our sample the coexistence of the positive and negative signals with different decay times leads us to suggest that independent mechanisms are operating. From the time scale and the near-independence temperature, we suggest that the negative signal with sub-ps decay time is due to the thermalization of the hot electrons before bottleneck, which can be probed through the photoinduced redistribution of spectral weight.[4] This is consistent with the experimental results shown in Fig. 1(b), where the negative signal appears at the initial rising edge of the positive signal. Now, we discuss the 9 ps negative component. According to Fig. 2(b), the positive component at $T = 89$ K is almost a constant, which may indicate the divergence of the decay time as $T \to T_c$ due to the closing of the superconducting gap. On the other hand, we may also consider the positive component as a long-lived background and explain the 9 ps negative component as the relaxation process which is directly related to the decay of HFP. However, since the energy band structure and its temperature dependence have not been well defined the sign change of the signal just near T_c is difficult to explain.

In conclusion, we have performed femtosecond optical detection of QP dynamics in an optimally doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal. Our results show clear evidence of strong phonon bottleneck. We also find the coexistence of a BCS-like T-dependent gap and a T-independent pseudogap.

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