Quasiparticle relaxation dynamics in underdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ by two-color pump-probe spectroscopy

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We investigate the relaxation dynamics of photoexcited quasiparticles (QPs) in underdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ ($T_c = 78$ K). By changing the excitation energy and polarization of the probe beam, two different types of relaxation dynamics, associated with superconducting (SC) and pseudogap (PG) QPs, are quantitatively analyzed independently. From the temperature dependencies, we obtained the SC gap, $\Delta_{SC}(0) = 24$ meV, using BCS-type temperature-dependent gap and the pseudogap, $\Delta_{PG} = 41$ meV. The pump fluence ($F$) dependence of the SC-dominated transients shows a contribution of the PG component above the saturation condition of the SC component ($F_{th} = 16 \mu J/cm^2$), where Cooper pairs with long-range order are fully destroyed within the photoexcited volume. Assuming a temperature-independent PG decay time, we successfully isolate the native SC transient even above $F_{th}$ by subtracting the PG response from the original data. In the saturation regime, the exponential decay (recovery of SC) is fast ($\tau_{SC} \sim 2-3$ ps), suggesting an efficient nonequilibrium phonon relaxation in this compound. We also find a flat-top response preceding the exponential decay at $F > F_{th}$, which appears as a delay of SC recovery in the original data. This response is visible over the whole temperature range below $T_c$ and its duration increases with increasing $F$. The response is attributable to a photoinduced SC to non-SC phase transition arising from excitation by the nonthermal QPs and/or high-frequency phonons. The consistently near-constant magnitude of the PG response at the start of the SC state recovery from the non-SC phase suggests a correlation between the SC and PG QPs.

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I. INTRODUCTION

The coexistence of superconducting (SC) gap and pseudogap (PG) is a fascinating feature in high-$T_c$ superconductors, and underdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (UD-Bi2212) is one of the most extensively studied cuprates from this point of view. Scanning tunneling microscopy/spectroscopy (STM/STS) and angle-resolved photoemission spectroscopy (ARPES) are the two major techniques of investigating the connection between SC and PG, where the former characterizes the spatial variations of the gaps, while the latter characterizes their momentum distributions. The recent STS and ARPES studies on UD-Bi2212 and Bi2201 (La) reveal that the SC gap located in the nodal region starts to open below $T_c$, while the PG shows no clear change across $T_c$.

Time-resolved optical spectroscopy provides another way to resolve the quasiparticles (QPs) associated with SC and PG in the time domain. Although momentum integrated energy spectra around the Fermi energy are often broad and featureless in superconductors, QP dynamics with femtosecond resolution allows us to separate the overlapping contributions more easily. Furthermore, nonequilibrium condition achieved by ultrashort pulse laser has a potential to directly reveal the pairing dynamics in time. The photoinduced vaporization of the SC condensate has been realized recently. For Bi2212, an ultrafast depletion of the SC condensate on a picosecond (ps) time scale has been demonstrated.

In this kind of experiments, the optical pump-probe method is typically employed, where the nonequilibrium dynamics of QPs induced by an intense pump pulse is traced by a probe pulse with a variable delay time between the two pulses. The pump pulse with an energy higher than the energy gap of SC/PG can be utilized to break Cooper pairs into QPs excited into nonequilibrium high-energy states. The nonequilibrium carriers immediately relax down to states near the gap via electron-electron and electron-phonon scattering, resulting in quasiequilibrium populations of QPs and high-frequency phonons. They then reach equilibrium by high-frequency phonon decay. A theoretical framework for analyzing the transient data has being established on the basis of comprehensive and systematic experiments on various superconductors involving BCS-type cuprates, Fe-based superconductors, and other related compounds. In the cuprates, the distinction of the QP dynamics between SC and PG has been realized by the characteristics of each dynamics, such as relaxation time, temperature dependence, or dependence on the external fields. The differences in transition probability and matrix elements for optical probe transitions also provide a possibility to selectively enhance the QP dynamics detection of SC/PG, which allows us to analyze each dynamics individually.

In our previous paper, we selectively isolated the transient dynamics associated with SC and PG QPs in UD-Bi2212 by changing the probe beam energy and polarization. We also showed that two distinct components can be detected simultaneously below $T_c$. However, the paper lacked quantitative details. In this paper, we report experimental studies on UD-Bi2212 by using a similar selective method as before. The photoexcitation with a low-repetition-rate laser allows quantitative analysis of the dynamics with high precision. In addition to the temperature dependence, we carry out the pump fluence dependence of the dynamics. At fluences where the
SC component is saturated, the PG component rises above the SC signal. At the same time, a delay of SC recovery appears and increases linearly with increasing pump fluence. We also extract the saturated SC dynamics by subtracting the PG component obtained above $T_c$ from the original data. The response is clearly visible even in the vicinity of $T_c$, indicating a SC to non-SC phase transition with an efficient nonthermal phonon distribution.

**II. EXPERIMENTAL**

The sample used in this work was underdoped Bi2212 single crystal with $T_c \approx 78$ K grown by the traveling solvent floating zone method. The time-resolved reflectivity change $\Delta R$ was measured using a two-color pump-probe setup and was carried out with a freshly cleaved sample mounted in an optical liquid-He flow cryostat. The two pulses were derived from a 250-kHz Ti:sapphire regenerative amplifier (RegA) and a signal of an optical parametric amplifier (OPA) pumped by RegA. Due to the low repetition rate, we can neglect the laser heating, as justified in Sec. III A from the data obtained at various fluences. We employed a combination of the excitation energies (wavelengths) of 1.55 eV (800 nm) and 0.95 eV (1300 nm), where the latter energy is slightly lower than that used in the previous report ($\sim 1.07$ eV).

Depending on the measurement, either 1.55/0.95 eV or vice versa were used for the pump/probe energy ($E_{pr}/E_{pu}$). The pump and probe beams were coaxially overlapped and focused onto the $ab$ plane of the crystal by a lens ($f = 50$ mm) located on-axis, allowing for the precise spatial overlap between the beams. The beam diameters from RegA and OPA were measured to be 30 and 40 μm at the sample position, respectively. In most cases, the collinearly polarized pump and probe beams were set to be parallel to $a$ axis of the crystal ($E_{pu} \parallel E_{pr} \parallel a$). The fluence of the pump beam ($F$) ranged from 9 to $\sim 900$ μJ/cm$^2$. We set the probe fluence about three orders of magnitude smaller than the pump. Owing to the energy difference between the pump and probe beams, we can spectroscopically distinguish the reflection beams for detection. The time resolution was estimated to be around 75 fs from cross correlation measurements.

**III. RESULTS AND DISCUSSIONS**

A. Temperature dependence

In Fig. 1, we plot the temperature dependence of $\Delta R/R$ transients at various excitation conditions at low excitation fluences. The upper panels show the density plots of the data, where the red and blue correspond to positive and negative signs of $\Delta R$, respectively. With excitation/probe energies of $E_{pu}/E_{pr} = 0.95/1.55$ eV [see Figs. 1(a) and 1(d)], only a negative $\Delta R/R$ transient is present below $T_c$, characteristic of the QP relaxation associated with the SC state. No remarkable polarization dependence was observed with this pump-probe energy combination. In contrast, a positive component is dominant in the reverse combination; $E_{pu}/E_{pr} = 1.55/0.95$ eV [see Figs. 1(b) and 1(c)]. This component remains constant across $T_c$; then decreases monotonically with further increasing temperature, and finally disappears around $T^* \approx 180 \sim 210$ K. Thus we assign the positive $\Delta R/R$ transient to QP relaxation associated with the PG. In the case of $E_{pr} \parallel a$ probe polarization [see Figs. 1(c) and 1(f)], an additional small negative component appears in the temperature region below $T_c$, which can be identified as a small contribution of the SC component observed in Figs. 1(a) and 1(d). On the other hand, the negative component is nearly absent and only the positive component is apparent when $E_{pr} \parallel b$ [see Figs. 1(b) and 1(e)].

As shown in Figs. 1(d) and 1(e), $\Delta R/R$ transients are well reproduced by a single exponential relaxation (thin solid lines), ensuring that we can uniquely evaluate the relaxation dynamics.

**FIG. 1.** (Color online) (Top) Density plots of $\Delta R/R$ transients as a function of temperature, obtained at (a) $E_{pu}/E_{pr} = 1.55/0.95$ eV (800/1300 nm) with $F = 9 \mu$J/cm$^2$ and (b) $E_{pu}/E_{pr} = 0.95/1.55$ eV (1300/800 nm), $F = 75 \mu$J/cm$^2$ and $E_{pr} \parallel a$ in (b) and $F = 25 \mu$J/cm$^2$ and $E_{pr} \parallel b$ in (c). (Bottom) Corresponding $\Delta R/R$ transients at typical temperatures.
of the SC and PG QPs at each excitation condition. Let us briefly comment on the selectivity of SC/PG component in the optical pump-probe measurement. In cuprates superconductors, a two-component signal consisting of SC and PG components has been widely observed, where the magnitude of the signal reflects the change of the interband transition probabilities induced by the photoexcited QPs. Since the transition probability depends on the probe energy and polarization, we can enhance/suppress one of the SC/PG components by changing the excitation condition. For the lowest temperature, which is comparable to the typical value obtained in other cuprates, it is possible to consider that the photoinduced change of the transition probabilities for PG (SC) component is strongly reduced in (d) ([e]), resulting in a single exponential relaxation with SC (PG). We emphasize that there should exist a small contribution from the minority [PG in (d) and SC in (e)] in each excitation condition, but its magnitude is negligibly small as compared with the dominant signal. This is consistent with the fact that we see a small contribution of SC component below \( T_c \) in Figs. 1(e) and 1(f), where the change of probe polarization breaks the balance to cancel out the SC component owing to the dipole matrix element for the transition. The two-component signal is also seen in the SC-dominated transients when increasing the pump fluence above the saturation of SC component, which will be discussed later and in Sec. III B in more detail.

To quantitatively confirm the assignment of SC and PG components, we plot in Fig. 2 the \( \Delta R/R \) dependencies of SC amplitudes together with the fits (thin solid lines) characterized by the SC gap \( \Delta_{SC} \) and pseudogap \( \Delta_{PG} \). Assuming sufficiently weak photoexcitation (so that the photoinduced QPs and phonon occupations are small compared to their equilibrium concentrations) and a momentum-independent gap, \( \Delta R/R \) amplitude is given by

\[
\frac{\Delta R}{R} \propto \left[ \Delta_{SC}(T) + \frac{k_BT}{2} \right]^{-1} \times \left\{ 1 + \frac{k_BT}{\Delta_{SC}(T)} \exp \left[ -\frac{\Delta_{SC}(T)}{k_BT} \right] \right\}^{-1}
\]

where \( g \) represents the ratio of bosonic and electronic density of states that contribute the photoinduced QP density. We used BCS-type \( \Delta_{SC}(T) \) and obtained \( \Delta_{SC}(0) = 24 \) meV, which is slightly larger than the values obtained from other experiments, but still consistent by taking into account the variation in the momentum space. The agreement with the data is also good enough to ensure the identification of the SC component. For the PG component, assuming the strong bottleneck condition (QP relaxation is governed by the hot phonon relaxation) and weak photoexcitation, \( \Delta R/R \) amplitude is given by

\[
\frac{\Delta R}{R} \propto \left[ 1 + \frac{2\nu k_BT}{N(0)\hbar \Omega_c} \exp \left( -\frac{\Delta_{PG}}{k_BT} \right) \right]^{-1}
\]

where \( N(0) \approx 2 \) eV\(^{-1}\)cm\(^{-1}\)spin\(^{-1}\) is the electronic density of states per unit cell and \( \nu \) and \( \Omega_c \) are, respectively, the number of bosons in the PG relaxation and their cutoff frequency. The best fit to the data at \( E_{pr} \parallel a \) (open circles) with Eq. (2) using \( T \)-independent \( \Delta_{PG} \) shows \( \Delta_{PG} = 41 \) meV, which is also consistent with the value obtained from the STS. In the same figure, we plot the amplitude of \( \Delta R/R \) at \( E_{pr} \parallel b \) (triangles) whose \( T \) dependence is consistent with the data at \( E_{pr} \parallel a \) and its fitting curve.

In order to check the amount of sample heating from the laser, we compare the \( T \) dependencies of SC amplitudes measured at various pump fluences [inset of Fig. 2(a)]. Here, the fluences of the two data sets correspond to \( F \) above saturation condition for SC component (where the SC condensate is temporarily destructed, see Sec. III B). A similar \( T_c \) was found at \( F = 70 \) \( \mu J/cm^2 \) (closed triangles) while a small (~5 K) apparent shift of \( T_c \) due to the laser heating is observed at \( F = 300 \) \( \mu J/cm^2 \) (open triangles). This implies that the laser heating is negligibly small for \( F \) up to a few hundred \( \mu J/cm^2 \).

Under the saturation condition, the assumption of weak photoexcitation in Eq. (1) is no longer valid. Instead, we employ the Mattis-Bardeen (MB) formula to characterize the data, which are given by

\[
\frac{\Delta R}{R} \propto \Delta_{SC}(T)^2 \ln \left( \frac{\hbar \omega}{\Delta_{SC}(T)} \right)
\]

where \( \hbar \omega \) is the photon energy. This formula represents a \( T \)-dependent difference in reflectivity between the normal (non-SC) and SC states and an assumption that the reflectivity in
to the amplitude, the decay time in each component shows a noticeable difference between low- and high-fluence cases. This featureless dependence is also consistent with the data very well with Eq. (4) shown by the thin line in Fig. 3(a). Here, we used $n_{ph}$ extracted from the measured $T$ dependence of amplitude [see Fig. 2(a)] and $n_T = N(0) \sqrt{2 \pi \Delta_{SC}(T) k_B T} \exp \left[-\Delta_{SC}(T)/k_B T\right]$ using $\Delta_{SC}(T)$ given by the BCS functional form. From the fit, we obtained $\Delta_{SC}(0) = 4.3$ meV, which is significantly smaller as compared with $\Delta_{SC}(0) = 24$ meV estimated from the fit to the $T$ dependence of the amplitude. Similar underestimation was observed by Kabanov et al.,$^8$ where they suggested that both the assumption of temperature-independent $\gamma$ and the underestimate of the laser heating lead to a reduction of $\Delta_{SC}(0)$. However, we can exclude the latter contribution, especially in the measurement with low pump fluence, because of both the low repetition rate of the laser and identical processes of phonon escape, particularly those associated with nanoscale textures (stripes).$^3$

For the comprehensive assignment of coexisting SC and PG components, we show the decay times of both the low repetition rate of the laser and identical processes of phonon escape, particularly those associated with nanoscale textures (stripes).$^3$ For the comprehensive assignment of coexisting SC and PG components, we show the decay times of both the low repetition rate of the laser and identical processes of phonon escape, particularly those associated with nanoscale textures (stripes).$^3$

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We removed this component in (b) and (d) by subtracting $\Delta R/R$ transient at $T = 260$ K from the original ones at all temperatures. The additional component is nearly identical to the PG responses observed at the reverse pump/probe energies ($E_{pu}/E_{pr} = 1.55/0.95$ eV), confirming the assignment. With increasing temperature above $T_c$, this PG component reaches its maximum, then gradually decreases, and finally disappears around $T^*$. On the other hand, below $T_c$, this component coexists with the native SC component, as has been demonstrated in the previous papers.7,13–22,27

In Figs. 4(c) and 4(d), we summarize the fluence dependencies of SC ($E_{pu}/E_{pr} = 0.95/1.55$ eV) and PG ($E_{pu}/E_{pr} = 1.55/0.95$ eV) components, respectively. The pump fluence $F$ varies up to 230 $\mu$J/cm$^2$ in (c) and 900 $\mu$J/cm$^2$ in (d). The amplitude of each component shows a saturation with increasing $F$. According to the inhomogeneous-excitation saturation model established by Kusar et al.,8 we determine the saturation threshold fluence $F_{th}$ by the value at which $\Delta R/R$ reaches $2\Delta/(1 + 2\lambda)/(1 + \rho^2)$ of the maximum of $\Delta R/R$, where $\lambda = \lambda_{pu}/\lambda_{pr}$, with optical penetration depths for pump ($\lambda_{pu}$) and probe ($\lambda_{pr}$), and $\rho = \rho_{pu}/\rho_{pr}$, with beam radii for pump ($\rho_{pu}$) and probe ($\rho_{pr}$). Using $\lambda = 124/174$ nm for 0.95/1.55 eV, extracted from the reflectance of nearly-optimally-doped Bi2212,32 and the measured values $\rho = 15/20$ $\mu$m for 0.95/1.55 eV, we obtained $F_{th} = 16$ $\mu$J/cm$^2$ for the SC component and 80 $\mu$J/cm$^2$ for the PG component. Note that, for the PG component, the saturation value of $\Delta R/R$ was not clearly determined due to the experimental limit of $F$, and therefore the exact $F_{th}$ should be larger than 80 $\mu$J/cm$^2$.

From $F_{th}$ for the SC component, we calculate the deposited optical SC-state destruction energy density $U_r = F_{th}/k_B = F_{th}(1 - \rho^2)/\lambda_{pu}$ with beam radii for pump ($\rho_{pu}$) and probe ($\rho_{pr}$), and $\lambda_{pu}$, which is much larger than the thermodynamic SC condensation energy of $U_c = 0.5$ K/Cu for UD-Bi2212 ($p \approx 0.12$).33 As previously suggested by Kusar et al.,6 the discrepancy between $U_r$ and $U_c$ indicates an efficient fast dissipation of the photoexcitation energy by phonons.6 Let us now consider the $F$ dependencies of the decay times [insets of Figs. 4(c) and 4(d)]. In contrast to the qualitatively similar $F$ dependence of the SC and PG amplitudes, the decay times in each component reveal unique $F$ dependencies, which are similar to those observed in the $T$ dependencies. Indeed, as shown by the thin line in the inset of Fig. 4(c), the reduction of $\tau_{SC}$ with increasing $F < F_{th}$ is well characterized by Eq. (4) using parameters derived from the $T$ dependence [see Fig. 3(a)]. With further increase of $F$, the decay time deviates from the model curve, and then increases upward. It is important to note that $\tau_{SC}$ in the upward region is slightly overestimated since $\Delta R/R$ transient itself deviates from the single exponential decay. This deviation behavior will be discussed in detail in the next section (Sec. III C). On the other hand, $\tau_{PG}$ shows a modest increase with increasing $F$ from 0.7 ps at the lowest $F$ to $\sim 1.4$ ps at the highest $F$.

![Figure 4](https://example.com/fig4)

**FIG. 4.** (Color online) $\Delta R/R$ transients at 10 K excited by various pump fluences with (a) $E_{pu}/E_{pr} = 0.95/1.55$ eV and (b) $E_{pu}/E_{pr} = 1.55/0.95$ eV and $E_{pu} \parallel a$. (c) and (d) Fluence dependencies of amplitudes and decay times (insets) for the corresponding excitation conditions. The saturation threshold fluence $F_{th}$ for destructing the superconducting condensate in the excited volume was estimated to be $16 \mu$J/cm$^2$ from (c). In the saturation regime, the decay times of SC ($\tau_{SC}$) obtained from the standard exponential fitting are overestimated. The correction of $\tau_{SC}$ will be presented in Fig. 6(c).

![Figure 5](https://example.com/fig5)

**FIG. 5.** (Color online) (a) Density plot of $\Delta R/R$ transients as a function of temperature at $E_{pu}/E_{pr} = 0.95/1.55$ eV with $F = 70 \mu$J/cm$^2$. (b) Density plot after subtraction of $T$-independent component from (a) by $\Delta R/R(T) - \Delta R/R(T = 260)$ K. (c) and (d) Corresponding $\Delta R/R$ transients at typical temperatures.

**C. Dynamics of superconducting condensate at $F \gg F_{th}$**

In this section, we concentrate our attention to the SC dynamics above the saturation condition at $E_{pu}/E_{pr} = 0.95/1.55$ eV. Figure 6(a) shows $\Delta R/R$ transients excited with various $F > F_{th}$ at 10 K, where, due to the saturation of the SC component, the contribution of the PG component becomes apparent as a positive peak together with an instantaneous negative peak that exists throughout the whole temperature range as mentioned in Fig. 5(a). The most striking feature of the saturated dynamics in the SC component is the appearance of a delay preceding the onset of the SC component recovery marked by arrows in Fig. 6(a).

In order to minimize the PG component contribution to $\Delta R/R$ in the delay region at high $F$ to obtain better insight...
Here, we assumed that the PG amplitude dependence of the PG component up to Δ1R/R does not significantly change with F/Δ1R.

Amplitude of the PG component at T = 90 K (>Tc). Each plot is shifted vertically. For comparison, ΔR/R obtained with the lowest F are shown in the top. (c) Plot of the exponential decay time (τSC, open circles) and the delay of the exponential decay (t∞, closed circles and triangles) as a function of F. The positions of t∞ are indicated by arrows in (a) and (b). (d) ΔR/R(T = 10 K) = CΔR/R(90 K) fitted with a delayed exponential decay (dashed line), where C is ΔRPG(T)/ΔRPG(90 K)≈ 1.3. (e) ΔR(t∞)/R = ΔRreq/R at 90K, where ΔRreq/R is defined in (b). The amplitude of the PG component at F∞ is also shown (triangle). The error bars represent the standard deviation of ΔR/R.

FIG. 6. (Color online) Typical ΔR/R transients at Eµ/Epr = 0.95/1.55 eV with various F > Fth (a) at 10 K (<Tc) and (b) at 90 K (>Tc). Each plot is shifted vertically. For comparison, ΔR/R obtained with the lowest F are shown in the top. (c) Plot of the exponential decay time (τSC, open circles) and the delay of the exponential decay (t∞, closed circles and triangles) as a function of F. The positions of t∞ are indicated by arrows in (a) and (b). (d) ΔR/R(T = 10 K) = CΔR/R(90 K) fitted with a delayed exponential decay (dashed line), where C is ΔRPG(T)/ΔRPG(90 K)≈ 1.3. (e) ΔR(t∞)/R = ΔRreq/R at 90K, where ΔRreq/R is defined in (b). The amplitude of the PG component at F∞ is also shown (triangle). The error bars represent the standard deviation of ΔR/R.

The subtracted transients [shown in Fig. 6(d)] show at the highest F a flat-top response, which follows the initial negative ~100 fs peak. While it is not clear whether the initial peak is an artifact of the subtraction procedure, the flat-top response seems a quite robust feature, since it is consistently present also in T-dependent subtracted intermediate-F transients shown in Fig. 7.

The F dependence of the flat-top duration, t∞, obtained from the delayed-exponential-function fits, is shown by the solid circles in Fig. 6(c) and appears consistent with the shifts of the negative peak position obtained from the raw data (triangles), ensuring the validity of the subtraction procedure. A linear fit to the data shows that the delay of the SC recovery intercepts zero at F = 20 μJ/cm², which is approximately equal to Fpr ≈ 16 μJ/cm², suggesting that the delayed recovery starts with the saturation of the SC response.

From the delayed-exponential fits [dashed lines in Fig. 6(d)], we obtain more accurate values of τSC in the saturation regime [shown as open circles in Fig. 6(c)], that show a much weaker dependence on F than τSC obtained by the nondelayed exponential fit [see inset of Fig. 4(c)].

In Fig. 7 we summarize the T dependence of the saturated SC component dynamics obtained by the subtraction procedure performed on the data of Fig. 5. The flat-top response is clearly visible throughout the whole temperature range below Tc. Unlike the divergent-like increase of the superconducting response duration with increasing T near Tc, the flat-top shows a finite t∞ (~3 ps at F = 70 μJ/cm²) even in the vicinity of Tc. Note that the delay of SC recovery is confirmed also by the shift of the negative peak in the raw data [see Figs. 5(a) and 5(c)].
The flat-top response indicates, due to the saturation properties of the SC component, that in the excited volume the SC gap is temporarily completely closed and the SC condensate remains completely destroyed during \( t_{\text{th}} \). Despite the unreliability of the subtraction during the initial few hundred femtoseconds, the raw data around the threshold indicate [see Fig. 4(a)] that the SC gap closing happens rather fast on a few hundred fs timescale, which is slightly faster than in \( \text{La}_{1-x}\text{Sr}_{x}\text{CuO}_{4} \).\(^{6}\) During the flat-top duration, QPs and hot phonons relax but do not contribute to the SC condensation. The subsequent fast (\( \sim 2 \) ps) partial recovery of the SC condensate indicates that the relaxation, during and immediately after the flat-top response, can not be associated with the heat diffusing thermally out of the experimental volume, but it must be associated with relaxation of a transient nonthermal state of the system characterized by an excess of the high-energy excitations.\(^{23}\) The existence of the transient nonthermal state is supported also by the absence of an increase of the relaxation time near the saturation threshold, which is observed in the case of the thermal transition near \( T_c \) [see Fig. 3(a)].

The two most obvious relaxation pathways in such nonthermal system are (i) the diffusion of hot nonthermal QPs and/or high-frequency phonons out of the experimental volume and (ii) their energy relaxation to the thermal (low-frequency) phonon bath. The lower bound for the timescale of (i) can be estimated on the basis of the measured in-plane low-\( F \) hot-QP diffusion constant in \( \text{YBa}_2\text{Cu}_3\text{O}_6 \).\(^{35,36}\) \( \kappa_{\text{sh}} \simeq 20–24 \text{ cm}^2/\text{s} \). In the present case, due to the pancake-like geometry of the excitation volume, only the out-of-plane diffusion can contribute. Taking into account the high anisotropy of UD-\( \text{Bi}_{2212} \), the timescale of (i), \( \tau_c \gg \tau_{\text{sh}} = \lambda_{\text{pu}}/\kappa_{\text{sh}} \simeq 6 \text{ ps} \), clearly indicates the dominance of process (ii).

Our observations therefore suggest that, after the initial fast closure of the SC gap, the system remains in a transient nonthermal normal state for up to \( t_{\text{th}} \sim 2 \) ps at the highest excitation density. During this time, the hot nonthermal QPs and high-frequency phonons are cooled by the low-frequency phonon bath. After \( t_{\text{th}} \), the population of the hot QPs and high-frequency phonons drops to such level that the SC condensate (and gap) can start to recover. Due to the nonthermal character of the transient normal state a fundamental question appears: what is the exact pathway of the SC state destruction and recovery?

Recently, it was proposed that the photoinduced transition to the normal state (and back) might be of first order.\(^{24,25}\) Our data indicate that there is no abrupt changes in the photoinduced reflectivity during the recovery, which would be an indication of the first-order transition. However, the excitation density is, due to the finite penetration depth, highly inhomogeneous and any abrupt changes could be easily smeared out. Moreover, the SC rise-time dynamics is obscured by other components preventing us to reliably observe any fast SC component switching during the SC state destruction. On the basis of the present data and in the absence of a second-order transition model valid at high \( F \), which would fit the data, it is therefore not possible to rule out a first-order transition.

Finally, we comment on the contribution of the PG QPs to the photoinduced SC to non-SC phase transition. Assuming that in the PG state \( \Delta R(R)/R \) directly corresponds to the population of the nonequilibrium PG QPs, we use the data above \( T_c \) in Fig. 6(b) to estimate the population of the nonequilibrium PG QPs at the moment when the SC condensate starts to recover. In Fig. 6(e), we therefore plot \( \Delta R(t_{\text{th}})/R - \Delta R_{\text{eq}}/R \) at 90K, where \( \Delta R_{\text{eq}}/R \) is the \( \Delta R/R \) averaged between 9 and 10 ps [see Fig. 6(b)]. For comparison, the amplitude of the PG component at \( F_{\text{th}} \) is also shown [triangle in Fig. 6(e)]. The result shows a virtually constant QP density at \( t_{\text{th}} \), suggesting the existence of a critical PG QPs density level correlated with the start of the SC condensation. In other words, the PG QPs also need to cool below a critical level before the SC condensate can recover, suggesting the correlation between the SC and PG QPs.\(^{36}\)

It is surprising that the gap recovery starts so fast after the complete gap destruction. However, we note that the recovery time of the SC state is also very fast, a few picoseconds, which is much faster than that of \( \text{MgB}_2 \) (\( \sim 100 \) ps)\(^{12}\) and even faster than in \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) (\( \sim 10 \) ps). Also note that the fast recovery time in Bi2212 was widely observed not only for UD samples\(^{22,25}\) but also for the optimally-doped (\( \sim 2.5 \) ps from time-resolved ARPES\(^{2,37}\)) as well as overdoped samples (\( \sim 3.3 \) ps).\(^{24}\)

**IV. CONCLUSIONS**

We investigated the relaxation dynamics of photoexcited quasiparticles (QPs) in UD Bi2212 (\( T_c = 78 \) K). The transient \( \Delta R/R \) dynamics associated with SC and PG QPs were selectively isolated from each other by changing the probe beam energy and polarization, enabling us to evaluate the individual dynamics quantitatively. Below the saturation condition, both the temperature and pump-fluence dependencies of SC transients agree well with those predicted from Rothwarf-Taylor model. From the temperature dependencies, we obtained \( \Delta_{\text{SC}}(0) = 24 \text{ meV} \) using BCS-type temperature-dependent gap and \( \Delta_{\text{PG}} = 41 \text{ meV} \), both of which are consistent with the values obtained from other measurements.

The pump fluence dependence of the SC-component-dominated transients shows a contribution of the PG component above the saturation of the SC component (\( F_{\text{th}} = 16 \mu \text{J/cm}^2 \)), where the SC condensate is fully destroyed within the photoexcited volume. We also found a delay of the SC state recovery whose duration time increases linearly with increasing \( F > F_{\text{th}} \). The delay of the SC state recovery is associated with a picosecond-timescale transient normal state. The subsequent fast (picosecond) partial SC state recovery suggests that the phonon and QP population remain highly nonthermal upon photoexcitation, on a time scale of a few picoseconds. During the nonthermal relaxation phase, the energy relaxation proceeds from the high-energy QPs and phonons, which are strongly coupled to phonons that do not break SC pairs. The latter can be the low-frequency phonons or maybe the phonons in the layer isolated from the Cu-O plane.

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et al.}

PHYSICAL REVIEW B 84, 174516 (2011)

8 V. V. Kabanov, J. Demsar, and D. Mihailovic, Phys. Rev. Lett. 95, 147002 (2005).
34 The $T$-independent negative peak, which is observed at high $T$ [see Figs. 5(a) and 5(c)] might be overestimated by the scaling by $C$.
36 On the basis of this scenario, one can expect the $T$-independent $I_{vM}$, which is confirmed in the $T$ range below $\sim 50$ K of Fig. 7. The increase of $I_{vM}$ (decrease of PG QPs at $I_{vM}$) above $T \sim 50$ K could be explained by the increasing number of the thermal PG QPs.