Quasiparticle dynamics in overdoped Bi$_{1.4}$Pb$_{0.7}$Sr$_{1.9}$CaCu$_2$O$_{8+δ}$: Coexistence of superconducting gap and pseudogap below $T_c$

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Abstract
Photoexcited quasiparticle relaxation dynamics in overdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8+δ}$ ($T_c=65$ K, hole doping $p=0.22$) single crystal is investigated as a function of temperature. We provide evidence of a $\sim 22$ meV pseudogap ($T^*\approx 100$ K) at this doping level. Our data support the scenario where both the superconducting gap and pseudogap coexist in the superconducting state. Our results also suggest an increased scattering rate between electrons and spin fluctuations as the sample enters the pseudogap phase.
All hole-doped cuprate high-temperature superconductors (HTSCs) exhibit an unusual normal state that is characterized by the opening of a gap in the electronic spectrum, at a temperature \( T^* \) above the superconducting (SC) transition temperature \( T_c \). Much theoretical and experimental effort has been spent in ascertaining the origin of this gap, called the pseudogap (PG) [1], for the answer may prove crucial in the understanding of high-\( T_c \) superconductivity. A fundamental issue regarding the PG phase is [2]: does it compete with, is unrelated to, or is a precursor of, superconductivity? Related to this is the number of energy gaps below \( T_c \): a single energy gap would imply that the PG is a precursor state, while two gaps would suggest that the PG is a competing or coexisting phase [3]. Another issue is the understanding of the HTSC phase diagram: does the \( T^* \) line merge with \( T_c \) on the overdoped side, or does it cross the SC dome and falls to zero at a quantum critical point? In the former case, does the PG phase coexist with the SC phase below \( T_c \), or is it a precursor to superconductivity by smoothly evolving into the SC phase below \( T_c \)?

A variety of experimental techniques have sought to answer some of these questions. The existence of the PG in underdoped hole-doped HTSCs is now not in doubt, but the issue is not so clear in the overdoped regime. Angle-resolved photoemission (ARPES) data in Bi-2212 [4] revealed a “peak-dip-hump” feature in the SC phase which persists above \( T_c \) in the PG phase. Two energy scales were associated with the PG — a low-energy one given by the location of the leading-edge midpoint (before the “peak”), and a high-energy one given by the position of a broad peak (“hump”) near the (\( \pi,0 \)) point. The low-energy PG smoothly evolves into the SC gap upon going from the underdoped to the overdoped regime and disappears in overdoped samples, while the high-energy PG persists in overdoped samples up to a hole doping \( p \approx 0.22 \). On the other hand, tunneling spectroscopy data [5], whose PG energies correspond to the high-energy scale in ARPES, showed that the PG exists in a highly overdoped sample (\( T_c = 56 \) K). Other measurements like \( c \)-axis transport [6] also observed a PG for an overdoped sample with \( p = 0.22 \), though it is not clear whether it is measuring the low-energy or high-energy PG.

Ultrafast time-domain pump-probe spectroscopy has shown to be a useful tool in studying the nonequilibrium carrier dynamics in HTSCs. This technique can differentiate between different quasiparticle (QP) excitations by their different relaxation timescales and thus distinguish the different phases, for example, the PG phase in cuprate and pnictide HTSCs.
In these experiments, a pump pulse first breaks Cooper pairs into QPs which rapidly relax to states close to the Fermi energy ($E_F$) by electron-electron and electron-phonon scattering. The presence of a gap near $E_F$ causes a relaxation bottleneck, so that carriers accumulate in states near the gap edge, and subsequent relaxation and recombination dynamics give rise to a transient change in optical transmission or reflection of a time-delayed probe pulse which can be measured. These studies helped in gaining information on the nature of low-energy electronic structure of correlated electron systems like HTSCs where the dynamics is sensitive to presence of a gap.

Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi-2212) is one of the most studied HTSCs because of its extremely large anisotropy and cleavability, containing only CuO$_2$ planes and not chains, and the possibility of growing samples with a larger range of $T_c$’s. The interpretation of the femtosecond QP dynamics in Bi-2212 with regards to the PG phase have only been reported for underdoped [7] and optimally doped [8] samples. Liu et al. [7] reported the coexistence of the PG and SC QPs in the SC state of an underdoped sample by simultaneously detecting two distinct components via tuning the probe beam polarization and energy. Cao Ning et al. [8] interpreted the dynamics of an optimally doped sample using the Rothwarf-Taylor (RT) model and showed the coexistence of a BCS-like temperature ($T$)-dependent SC gap and a $T$-independent PG. In this paper, we analyze pump-probe data of an overdoped (OD) Bi-2212 single crystal sample. In fitting the $T$-dependence of the relaxation amplitude in the SC state, we require the presence of two $T$-dependent gaps to fit the data. Our data are consistent with the scenario where the SC gap and the PG coexist below $T_c$, i.e. the “two-gap” scenario.

The experiments were performed on single crystals of Bi-2212 grown using the traveling-solvent-floating-zone method [11]. The OD sample has been doped with Pb to obtain a $T_c$ of 65 K ($p=0.22$). The value of $T_c$ obtained from magnetization data collected using Magnetic Property Measurement System (MPMS) correspond to the midpoint of the SC transition. The onset of superconductivity occurs at 68 K. The hole-doping value ($p$) were obtained from the $T_c$ values using the parabolic law [12]: $T_c = T_c^{max}[1 - 82.6(p - 0.16)^2]$, where $T_c^{max}=95$ K. The sample was cleaved before data were taken.

In our experiment, an 80-MHz Ti:Sapphire laser produces sub-50 fs pulses at $\approx$ 800 nm (1.55 eV) as a source of both pump and probe pulses. The pump and probe pulses were cross-polarized. The pump spot diameter was 60 $\mu$m and that of probe was 30 $\mu$m. The
FIG. 1: (color online) Waterfall plots of transient reflection $\Delta R/R$ versus pump-probe time delay at different temperatures. Solid lines at 32 K and 100 K are two-exponential fits.

The reflected probe beam was focused onto an avalanche photodiode detector. The photoinduced change in reflectivity ($\Delta R/R$) was measured using lock-in detection. In order to minimize noise, the pump beam was modulated at 100 kHz with an acousto-optical modulator. The experiments were performed with an average pump power of 500 $\mu$W, giving a pump fluence of $\sim0.3$ $\mu$J/cm$^2$ and a photoexcited QP density of $\sim1 \times 10^{-3}$/unit cell, showing that the system is in the weak perturbation limit. The probe intensity was approximately 10 times lower. The $T$ rise of the illuminated spot has been accounted for in all the data.

In Figure 1, we show the time dependence of $\Delta R/R$ at various temperatures above and below $T_c$. At low temperatures, a fast positive $\sim100$ femtosecond (fs) decay ($A_{fast}$) and
a slow picosecond (ps) negative decay ($A_{\text{slow}}$) were observed, with the negative signal disappearing above $T_c$. We therefore attribute $A_{\text{slow}}$ to the reformation of SC order following photoexcitation. Above $T_c$, a two-exponential positive decay was seen up to 300 K.

Using a two-exponential decay function, we extract the temperature dependence of the relaxation amplitudes and relaxation times, as shown in Fig. 2(a) and Fig. 3. Notice that $A_{\text{slow}}$ (1) crosses zero at $\sim 65$ K, and (2) exhibits a dip at $\sim 100$ K. The slow relaxation time $\tau_{\text{slow}}$, on the other hand, exhibits an upturn near two temperatures: (1) $65$ K ($T_c$) and (2) $100$ K, which we denote as $T^*$, the PG temperature. Both the dip in $A_{\text{slow}}$ at $100$ K, and the upturn in $\tau_{\text{slow}}$ at $65$ K and $100$ K, were reproducible upon re-cleaving the same sample.

In order to analyze the $T$-dependence of $A_{\text{slow}}$ quantitatively, we use the model proposed by Kabanov et al. [13]. The $T$-dependence of the relaxation amplitude in the SC state for an isotropic $T$-dependent gap $\Delta_c(T)$ is given by

$$G(T) \propto \frac{\epsilon_I/(\Delta_c(T) + k_B T/2)}{1 + \zeta \sqrt{\frac{2k_BT}{\pi \Delta_c(T)}} \exp[-\Delta_c(T)/k_B T]},$$

where $\epsilon_I$ the pump laser intensity per unit cell, and $\zeta$ is a constant. The above expression for $G(T)$ describes a reduction in the photoexcited QP density with increase in temperature, due to the decrease in gap energy and corresponding enhanced phonon emission during the initial relaxation. On the other hand, the $T$-dependence of the relaxation amplitude for a $T$-independent gap $\Delta_p$ is given by

$$P(T) \propto \frac{\epsilon_I/\Delta_p}{1 + \zeta \exp(-\Delta_p/k_B T)}.$$  

We first fit $A_{\text{slow}}(T > T_c)$ with $P(T)$, shown by the dashed line in Fig. 2(a) — the fit obviously does not reproduce the dip in $A_{\text{slow}}$ at $100$ K. Next, we proceeded to fit $A_{\text{slow}}(T)$ with the difference $G'(T) - G(T)$, where $G(T)$ is a function of the $T$-dependent SC gap $\Delta_{\text{SC}}(T)$ which closes at $T_c$, and $G'(T)$ is a function of the $T$-dependent PG $\Delta_{\text{PG}}(T)$ which closes at $T^*$. Both $\Delta_{\text{SC}}(T)$ and $\Delta_{\text{PG}}(T)$ are assumed to obey the BCS $T$-dependence in this overdoped regime. The results are shown as solid lines in Fig. 2(a). The dip in $A_{\text{slow}}$ at $100$ K is reproduced. The fitted values of $T_c$ and $T^*$ are $71$ K and $104$ K, respectively. We attribute the discrepancy between the data and fitted lines near $T_c$ to fluctuation effects. Nevertheless the quality of the fits to $\tau_{\text{slow}}(T)$ later are not affected. The fitted values of the zero-temperature gaps are $\Delta_{\text{SC}}(0) = (3.0 \pm 0.2)k_B T_c$ and $\Delta_{\text{PG}}(0) = (4.1 \pm 1.5)k_B T_c$. The fitted value of the zero-temperature SC gap $\Delta_{\text{SC}}(0)$ agrees well with tunneling data $[2\Delta_{\text{SC}}(0)/k_B T_c = 5.3]$. 

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FIG. 2: (a) $T$ dependence of $A_{\text{slow}}$ (solid circles), with fit of data above $T_c$ with $P(T)$ (dashed line), and fit of data from 28 K to $T^*$ with $G''(T) - G(T)$ (solid line). The fitted values of $T_c$ and $T^*$ are 71 K and 104 K, respectively. (b) Doping dependence of the PG as determined by the position of leading-edge midpoint (○, left axis) and high-energy feature ( Kı, right axis) in the ($\pi,0$) ARPES spectra from Bi-2212. The solid circle corresponds to the PG energy scale (22 meV) deduced from our data. The dome represents the $d$-wave mean-field approximation $\Delta(x) = 4.3 k_B T_c(x)/2$. Dashed line is a guide to the eye. Adapted with permission from Fig. 62 of Ref. 4. Copyright 2003 by the American Physical Society.
Note that our treatment of the PG to be $T$-dependent was motivated by the dip of $A_{\text{slow}}$, and the concurrent upturn of $\tau_{\text{slow}}$, at 100 $K$ ($\approx T^*$). Compare this with pump-probe data of other cuprates, such as $Y_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ [9], $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$ [15], and optimally doped Bi-2212 [8], where no dip in the relaxation amplitude, nor upturn in the relaxation time, was seen at $T^*$, and so the PG’s there were treated to be $T$-independent. We attribute this difference to the large amount of overdoping of our sample. In tunneling data on an overdoped ($T_c = 74.3 \text{ K}$) Bi-2212 sample, the PG has a smaller magnitude than the UD PG, and has already almost vanished at 89 $K$ [2, 5]. The attribution of a temperature dependence to our OD PG is also consistent with the behavior of $\tau_{\text{slow}}$ near $T^*$ — the rapid appearance of a (pseudo)gap at $T^*$ presents a relaxation bottleneck, which causes an upturn in $\tau_{\text{slow}}$ at $T^*$. If the PG were to be $T$-independent, there would not be a relaxation bottleneck, and we would not have seen the upturn in $\tau_{\text{slow}}$ at $T^*$. Our analysis suggests that, even in the OD regime, there is a coexistence of the SC and PG phase below $T_c$. Our results are thus consistent with the “two-gap” scenario. Our conclusions are also consistent with recent tunneling data on an overdoped Bi-2201 sample [16], where two gaps were observed below $T_c$. Note that our value of $\Delta_p(0) \approx 22 \text{ meV}$, is consistent with the trend in the positions of the leading-edge mid-point of ARPES data at lower dopings (see Fig. 2(b)). It is interesting to see that our pump-probe technique yields values of the PG energy scale that coincide with the low-energy PG scale from ARPES.

Next we analyze the $T$-dependence of the slow relaxation time $\tau_{\text{slow}}(T)$ by using the RT model [17]. This is a phenomenological model that describes the dynamics of photoexcited QPs and high-frequency phonons (HFPs), where the presence of a gap in the electronic density of states (DOS) gives rise to a bottleneck for carrier relaxation. When two QPs with energies $\geq \Delta$ (\(\Delta\) is the gap magnitude) recombine, a HFP is created with $\omega > 2\Delta$. These HFPs trapped within the excited volume can further rebreak Cooper pairs and act as a bottleneck for QP recombination. Hence the SC recovery is governed by the decay of the HFP population. In the SC state ($T < T_c$), the $T$-dependence of $\tau_{\text{slow}}^{-1}$ is determined by the $T$-dependence of the amplitude $A_{\text{slow}}(T)$ and is given by [8, 18, 19]:

$$\tau_{\text{slow}}^{-1}(T) = \Gamma \left\{ \delta G(T) + \eta \sqrt{\Delta_{SC}(T)/T} \exp\left[-\Delta_{SC}(T)/T\right] \right\} \times \left[ \Delta_{SC}(T) + \alpha T \Delta_{SC}(T)^4 \right], \quad (3)$$

while in the PG phase ($T_c < T < T^*$), $G(T)$ is replaced by $G'(T)$, and $\Delta_{SC}(T)$ replaced by $\Delta_{PG}(T)$, with $\Gamma$, $\delta$, $\eta$ and $\alpha$ as fitting parameters. Figure 3(a) shows the $T$-dependence of
FIG. 3: (a) Solid circles: $\tau_{\text{slow}}(T)$ data. Solid line: Fit to Eq. (3) for $T < T_c$. Dotted line: Fit to Eq. (3) for $T_c < T < T^*$. Dashed line: Fit to Eq. (4) for $T < T_c$. (b) $\tau_{\text{fast}}(T)$ data (solid circles), with fit to $1/T^n$ (solid line) where $n=1.3$. 
The good fits show that the relaxation dynamics in both the SC phase and the PG phase can be explained by the presence of a relaxation bottleneck due to a gap in the DOS. The term $[\Delta_{SC}(T) + \alpha T \Delta_{SC}(T)^4]$ in Eq. (3) accounts for the $T$-dependence of phonon decay rate and ensures that the values of $\Delta_{SC}(0)$ and $\Delta_{PG}(0)$ obtained from fits to $A_{slow}(T)$ and $\tau_{slow}(T)$ are the same [19]. Also note that $\Delta_{PG}(0)/k_B T^* = 2.6$ — this ratio is consistent with the value (2.4) obtained from tunneling data of an OD sample with $T_c = 82$ K [2, 20], providing additional justification that the crossover to the PG phase really does take place at 100 K.

One might question the wisdom of using just $G(T)$ and $\Delta_{SC}(T)$ component in Eq. (3). After all, in OD Bi-2212, the gap distribution on the sample surface is more homogeneous than in underdoped or optimally-doped samples [21, 22]. One may suspect therefore that, in the SC state, the SC gap and PG add in quadrature to yield an effective gap $\Delta_{eff}(T) = \sqrt{\Delta_{SC}(T)^2 + \Delta_{PG}(T)^2}$ [23]. We attempt to fit the data of $\tau_{slow}$ below $T_c$ using

$$\tau_{slow}^{-1}(T) = \Gamma \left\{ \delta[G(T) + G'(T)] + \eta \sqrt{\Delta_{eff}(T)} T \exp[-\Delta_{eff}(T)/T] \right\} \times [\Delta_{eff}(T) + \alpha T \Delta_{eff}(T)^4].$$

(4)

The poor fit of Eq. (4) (dashed line) to data, shown in Fig. 3(a), shows that this “effective-gap” picture does not work. ARPES data, near the antinodes of an OD Bi-2212 sample ($T_c = 86$ K) [24], are also inconsistent with the SC gap and PG adding in quadrature.

We now turn to the fast component, which is positive at all temperatures. A two-exponential positive decay was also seen in optimally-doped Bi-2212 [25] — there the authors attributed the fast decay to coupling between electrons and “hot” phonons (i.e. phonons that are strongly coupled to the electrons), while the slow decay was due to anharmonic coupling between the hot phonons and the cold lattice bath. Figure 3(b) shows the temperature dependence of $\tau_{fast}$ — notice its rise with decreasing temperature, before peaking at $T^*$ and decreasing to $\sim$100 fs at 30 K. Notice also the slight change in slope of $\tau_{fast}$ at $T_c$. The change in behavior of $\tau_{fast}$ at $T^*$, and to a lesser extent at $T_c$, is intriguing — they suggest that the fast relaxation may result from an admixture of electron-phonon and electron-spin fluctuation coupling. The peak at $T^*$, and its subsequent decrease below $T^*$, may be due to an increased scattering rate between electrons and spin fluctuations as the sample enters the PG phase. This scenario is further confirmed by the $T$-dependence of $\tau_{fast}$ above $T^*$ — a fit to $1/T^n$ yields $n = 1.3$, which disagrees with the behavior predicted by Kabanov and
Alexandrov [26] for the electron-phonon relaxation time for good \((n=2)\) and poor \((n=3)\) metals.

We have performed ultrafast time-resolved photoinduced reflectivity measurements on overdoped \(\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}\) single crystals. Our data are consistent with the formation of a pseudogap phase at \(T^*=100\) K, which coexists with the superconducting phase below \(T_c\). We also see an increased scattering rate between electrons and spin fluctuations as the sample enters the pseudogap phase. Experimental studies on other moderate-to-extreme overdoped cuprates are clearly needed to confirm whether the pseudogap exists, is also temperature-dependent, and whether \(\tau_{fast}\) also peaks at \(T^*\), in these materials.

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[1] T. Timusk and B. Statt, Rep. Prog. Phys. 62, 61 (1999).
[2] Ø. Fischer et al., Rev. Mod. Phys. 79, 353 (2007).
[3] C. Panagopoulos and T. Xiang, Phys. Rev. Lett. 81, 2336 (1998).
[4] A. Damascelli, Z. Hussain, and Z.-X. Shen, Rev. Mod. Phys. 75, 473 (2003).
[5] C. Renner et al., Phys. Rev. Lett. 80, 149 (1998).
[6] T. Shibauchi et al., Phys. Rev. Lett. 86, 5763 (2001).
[7] Y. H. Liu et al., Phys. Rev. Lett. 101, 137003 (2008).
[8] N. Cao et al., Chin. Phys. Lett. 25, 2257 (2008).
[9] J. Demsar et al., Phys. Rev. Lett. 82, 4918 (1999).
[10] E. E. M. Chia et al., Phys. Rev. Lett. 104, 027003 (2010).
[11] N. Ichikawa, Ph.D. thesis, University of Tokyo, 1999.
[12] M. R. Presland et al., Physica C 176, 95 (1991).
[13] V. V. Kabanov, J. Demsar, B. Podobnik, and D. Mihailovic, Phys. Rev. B 59, 1497 (1999).
[14] S. Hufner, M. A. Hossain, A. Damascelli, and G. A. Sawatzky, Rep. Prog. Phys. 71, 062501
(2008).

[15] J. Demsar et al., Phys. Rev. B 63, 054519 (2001).

[16] M. C. Boyer et al., Nat. Phys. 3, 802 (2007).

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

[18] V. V. Kabanov, J. Demsar, and D. Mihailovic, Phys. Rev. Lett. 95, 147002 (2005).

[19] E. E. M. Chia et al., Phys. Rev. B 74, 140409(R) (2006).

[20] R. Dipasupil, M. Oda, N. Momono, and M. Ido, J. Phys. Soc. Jpn. 71, 1535 (2002).

[21] K. McElroy et al., Science 309, 1048 (2005).

[22] J. W. Alldredge et al., Nat. Phys. 4, 319 (2008).

[23] C.-C. Chien, Y. He, Q. Chen, and K. Levin, Phys. Rev. B 79, 214527 (2009).

[24] W. S. Lee et al., Nature 450, 81 (2007).

[25] L. Perfetti et al., Phys Rev Lett 99, 197001 (2007).

[26] V. V. Kabanov and A. S. Alexandrov, Phys. Rev. B 78, 174514 (2008).