Correlation of pseudogap, superconducting ordering and superconducting fluctuations with electron–phonon interactions in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8+\delta$

Yong Li$^{1,2,6}$, Weizheng Liang$^{3,4,6}$, Ning Li$^{3}$, Yongliang Chen$^{2}$, SN Luo$^{7,*}$ and Yong Zhao$^{1,2,5,*}$

1 School of Physical Science and Technology, Southwest Jiaotong University, Chengdu, Sichuan, 610031, People’s Republic of China
2 Superconductivity and New Energy R & D Center, Key Laboratory of Advanced Technologies of Materials (Ministry of Education), Southwest Jiaotong University, Chengdu, Sichuan, 610031, People’s Republic of China
3 Key Laboratory of Advanced Technologies of Materials (Ministry of Education), and Institute of Material Dynamics, Southwest Jiaotong University, Chengdu, Sichuan 610031, People’s Republic of China
4 The Peac Institute of Multiscale Sciences, Chengdu, Sichuan 610031, People’s Republic of China
5 College of Physics and Energy, Fujian Normal University, Fuzhou, Fujian, 350117, People’s Republic of China
6 Authors to whom any correspondence should be addressed.
7 Equal contribution.

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Abstract

The relationship between anomalous pseudogap and superconducting states is a fascinating but controversial subject in high temperature superconductors. Here, we investigate the different quasiparticle dynamics with femtosecond transient optical spectroscopy in underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8+\delta$ at 3–250 K. Below $T_c$, these results reveal the coexistence of the anomalous pseudogap and superconducting states. The Mattis–Bardeen formula for the Cooper-pairing superconducting gap and the Kabanov model for the pseudogap state, the measurements yield a temperature-dependent superconducting gaps and two temperature-independent pseudogaps below and above the superconducting transition $T_c$. Above the superconducting transition temperature, the pseudogap state can be explained as an incipient condensation of Cooper pairs with short coherence length. The quasiparticle dynamics in the superconducting and pseudogap states are correlated well with electron–phonon interaction characteristics.

1. Introduction

The relationship between anomalous pseudogap (PG) and superconducting (SC) states is always a hotly debated topic in high temperature superconductors [1–18]. Two main scenarios can be distilled from relevant experimental and theoretical studies [3, 7–10, 12, 13]. In one scenario, PG is dominated by a hidden order which competes with superconductivity below SC transition temperature $T_c$ [8, 12]. In the other scenario, PG is a precursor to superconductivity, an incipient condensation of Cooper pairs with short coherence length [13]. Both scenarios are closely related with basic interactions that may dominate the superconductivity of cuprates. Therefore, revealing the exact relation between SC and PG states and underlying mechanisms is important for understanding the Cooper pairing mechanism, the core issue of superconductivity. By means of such advanced techniques as electron tunneling spectroscopy, angle-resolved photoemission spectroscopy (ARPES), and electron Raman scattering, some characteristics of PG, including the coexistence of and the relationship between PG and SC, are revealed [5–10]. However, these methods are subject to their own limitations, and it is difficult to detect simultaneously the microscopic dynamic processes of the quasiparticles underlying the PG and SC states.
Recently, ultrafast optical pump–probe spectroscopy has ultrahigh temporal resolutions, thus advantageous for elucidating the nature of electron relaxation by studying non-equilibrium dynamics after a sudden nonthermal photoexcitation in superconductors [14, 19–25]. The measured signal is a relative change in reflectivity \( R(t) = \Delta R(t)/R_0 \), where \( R_0 \) refers to the reflectivity at zero pump fluence. Ultrafast pump–probe spectroscopy can identify the dynamics of quasiparticles in both the PG and SC states [19, 22, 25], and in the meantime detect the microscopic dynamical processes related to electron phonon interactions (EPI) [14, 26–28]. Beyond that, time-resolved spectroscopy shows certain advantages in measuring the EPI strength than Raman or neutron scattering [28]. Photo-excitation of charge carriers, and then relaxation kinetics by electron–electron scattering and electron–phonon collisions to quasi-particle states near the Fermi energy within \( \sim 100 \) fs where PG–SC phase changes occur, can be captured by measuring transient reflectivity \( \Delta R(t)/R_0 \) [14]. In the limit of the local non-equilibrium regime, different components can be directly distinguished in real time by virtue of the reflectivity \( \Delta R(t)/R_0 \) variation. Ultrafast time-resolved spectroscopy has been a powerful tool to study different interacting dynamics of charge and lattice degrees of freedom in high temperature superconductors.

In this work, we study the correlation between Cooper pairs and the PG phase in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212) single crystals by femtosecond transient optical spectroscopy. We obtain the dynamics of quasiparticles of PG and SC states, identify EPI, and establish the correlation of order parameters of SC and PG with EPI.

2. Experiment

2.1. Femtosecond pump–probe time-resolved optical reflectivity

The Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ single crystals piece laid on a copper bracket in optical cryostat system. The beam is allowed to enter the cryogenic system and fall on the sample. The cryogenic system can be cooled down to liquid helium temperature. And, the surrounding magnetic field is less than 0.3 Oe in the cryogenic box. The phase locked amplifier with digital signal processors has been used in the ultrafast time-resolved optical reflectivity. The split pump–probe scheme is a synchronously pumped tunable mode-locked laser. The pulsed Ti:sapphire laser is used in the time-resolved measurements of the reflectivity experiment. The more details can see the previous work [29].

We use a Ti:sapphire laser system delivering \( \sim 35 \) fs pulses at 80 MHz, with a center wavelength of \( \sim 800 \) nm (1.55 eV). The laser pulse at 800 nm is used for both pump and probe beams. The pump fluence is about \( \sim 9 \) \( \mu \)J cm$^{-2}$. The probe intensity is 10 times weaker than the pump intensity. Both the probe and pump beams are incident on a freshly cleaved sample surface along the \( c \)–axis, overlapping with each other. The Bi2212 crystals are flux grown via the traveling solvent floating zone method in an infrared image furnace. The SC phase transition, which takes place at \( T_c = 75 \) K in our sample. The PG opening temperature \( T^\ast \) is determined from the resistivity measurement and the phase diagram [30].

2.2. Magnetization measurements

The magnetic property was performed by physical property measurement system (MPMS, Quantum Design). The inset of the figure has shown the result of magnetization measurement in figure 1, and the superconductivity transition temperature is in 75 K. As shown in the \( M\text{--}T \) curves, the low-field
temperature dependence of the magnetization transition (FC or ZFC) is clean and neat indicate that the samples hardly contain any other intermediate phase in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ system.

3. Results and discussion

Figure 1 shows the $\Delta R(t)/R_0$ time series at 3–240 K. Here $t$ denotes time delay of the probe beam with respect to the pump beam. The sign of $\Delta R(t)/R_0$ reverses across the SC transition temperature $T_c = 75$ K [figures 2 and 3(a) and (b)], similar to references [12, 15]. Meanwhile, the relaxation process is significantly slowed down around $T_c$. However, different from a previous study, no sign change in $\Delta R(t)/R_0$ occurs at the PG opening temperature $T^* = 216$ K, but the relaxation process slows down around $T^*$. Compared with the SC component, we think the PG state is very weak in this work. This is the primary reason we are unable to gain some more obvious changes in figure 4(b) and 3(b). There is another fact that the various pump power can directly affect the shape of transient optical spectroscopy $\Delta R(t)/R_0$. And, the signs depend on the relative contributions of multiple microscopic relaxation processes; in addition to relaxation of the quasiparticles excited in the SC and PG states, there also exists scattering of carriers via EPI.

An experimental $\Delta R(t)/R_0$ time series can be well described with a two-exponential decay function,

$$\mathcal{R}(t) \equiv \frac{\Delta R(t)}{R_0} = A_s \exp \left( -\frac{t}{\tau_s} \right) - A_f \exp \left( -\frac{t}{\tau_f} \right),$$

(1)

where $A$ and $\tau$ are the amplitude and lifetime of a relaxation process, and subscripts $s$ and $f$ denote slow and fast processes, respectively. Figure 2(d) presents a typical fitting to the $\Delta R(t)/R_0$ time series with
Figure 4. (a) Amplitude $A_f$ and (b) decay time $\tau_f$ of transient reflectivity for the fast decay process (EPI) at different temperatures.

equation (1) at $T = 10$ K. All the time series at different temperatures can be well fitted with equation (1). Below $T_c$, the slow process has a typical relaxation time ($\tau_s$) of several $\sim$ps, which is related to pairing electrons in the SC or PG states [14, 22, 27]. On the other hand, $\tau_f$ is about 100–500 fs for the fast process, which is related to EPI. As described the two exponential decay function, the two processes can be fitted by the negative and positive exponential function in the different transient reflectivity $\Delta R(t)/R_0$.

The $\tau_s(T)$ and $A_s(T)$ curves for the slow process both show anomalous changes at 75 K and 216 K, corresponding to the SC transition temperature $T_c$ and the PG opening temperature $T^*$ [figure 3(a)], respectively, suggesting that the slow relaxation process is related to the SC and PG states. The anomalous changes are due to the bottleneck effect: upon photo-excitation, the electrons are excited to an energy level far above the Fermi surface, then relax to right above the SC or PG gap, and simultaneously release their excess energy by emitting phonons. Part of the emitted phonons, with energy $\hbar \omega_{ph} > 2\Delta_{SC}$ (or $\hbar \omega_{ph} > 2\Delta_{PG}$), re-excite electron–hole pairs between SC gap (or PG gap) to form quasi-equilibrium quasiparticles, and break the Cooper pairs. Here, $\hbar = h/2\pi$, $h$ is Planck’s constant, $\omega_{ph}$ is phonon frequency, and $\Delta$ is band gap. On the other hand, the recombination of electron–hole pairs will directly affect the Cooper pairs. The competition between the formation and disintegration of Cooper pairs evolves in a way dictated by phonon population, i.e., the number of quasiparticles, and their life time shows anomalous changes [14, 28, 31].

Fitting to the $\tau_s(T)$ and $A_s(T)$ curves yield band gaps of SC and PG states. Quantitatively, the $A_s(T)$ data in figure 3(a) can be analyzed with the Mattis–Bardeen theory [32] at the high frequency limit for a Cooper-pairing superconductor gap, and the Kabanov et al model [33, 34] for a temperature-independent PG. From the Mattis–Bardeen theory ($\hbar \omega \gg 0$), the reflectivity changes as a function of temperature for SC is [32, 34]

$$R_{SC}(T) \propto \frac{2\Delta(T)}{\hbar \omega_{op}} \ln \left[ \frac{1.47\hbar \omega_{op}}{\Delta(T)} \right],$$

where $\omega_{op}$ is the photon energy, $\Delta(T) = \Delta_0[1 - (T/T_c)^2]$. Under the phonon bottleneck condition, the reflectivity change for PG can be written as [33]:

$$R_{PG}(T) \propto \left[ 1 + B \exp \left( \frac{\Delta_{PG}}{k_B T} \right) \right],$$

where $k_B$ is Boltzmann constant, $B = 2v/[N(0)\hbar \Omega_C]$, $N(0)$ is the density of electronic states at the Fermi level, and $v$ is the number of bosons involved in relaxation process across the PG, which is high frequency phonons here. $\Omega_C$ is the cutoff frequency of the bosonic spectrum, and $\Omega_C = 0.1$ eV from reference [33]. For $v = 10–20$, $N(E) \approx 2.5–5$ eV$^{-1}$ cell$^{-1}$ spin$^{-1}$.
The $A_c(T)$ data can be well fitted with equations (2) and (3) as shown in figure 3(a). The fitting yields $\Delta_0 = 43$ meV for the SC states, and $\Delta_{\text{PG1}} = 37$ meV below $T_c = 75$ K, and $\Delta_{\text{PG2}} = 24$ meV between 75 K and 216 K for the PG states. The experimental data below $T_c$ can be well fitted by considering the contributions both from SC and PG [the red line in the inset of figure 3(a)], but not from SC only [the green line in the inset of figure 3(a)]. This demonstrates the coexistence of the SC and PG states below $T_c$.

As regards the second feature, i.e., a significant fluctuation of $\tau_\gamma$ near $T_c$ (referred as the SC fluctuation), it is originated from the opening of SC gap and resulting bottleneck effect: the high frequency phonons re-excite the Cooper pairs hence induce fluctuation of the superfluid density, $n_s$ [35, 36], manifesting as the fluctuation in $\tau_\gamma$. Therefore, it is reasonable to use the phonon-induced pair breaking model to quantitatively fit the experimental data of $\tau_\gamma(T)$. For the phonon-induced pair breaking process, the gap-dependent lifetime of the quasiparticles can be described as [33]

$$\tau_\gamma \propto \frac{h\omega_{ph}^2}{12\Gamma_{\omega}^2\Delta(T)^2} \ln \left[ \frac{\epsilon_f}{2N(0)\Delta(0)^2} + \exp \left( -\frac{\Delta(T)}{k_0 T} \right) \right].$$

(4)

Here $\omega_{ph}$ is the frequency of high frequency phonons, $\epsilon_f$ is the absorbed incident laser pulse energy density per unit cell, and $\Gamma_{\omega}$ is the phonon line-width and treated as a constant. As shown in figure 3(b), the fitting with equation (4) to the $\tau_\gamma(T)$ data below $T_c$ yields $\Delta_0 = 43$ meV, identical to the value obtained from the $A_c(T)$ data. In addition, a smaller fluctuation in $\tau_\gamma(T)$ also occurs at $T = T^*$ [figure 3(b)], consistent with the anomalous peak $A_c(T^*)$ in figure 3(a).

As a controversial topic, PG state arises from the incipient formation of Cooper pairs with short coherence length [13], and the SC state is a result of condensation of the short-coherence-length Cooper pairs. Others think that the PG states competes with SC states [9, 12]; consequently, the PG state should be suppressed by the SC state below $T_c$. While, the above quantitative analysis reveals the coexistence of the SC and PG states below $T_c$. PG shows a slightly larger gap values below $T_c$ ($\Delta_{\text{PG1}} = 37$ meV) than above $T_c$ ($\Delta_{\text{PG2}} = 24$ meV). One reason may be that the PG state is an intermediate state between Cooper pairs with the phase fluctuation and the global phase coherence. After the transient collisional excitation by laser, the breaking of Cooper pairs will interact with each other.

As mentioned above, the fast process with a time scale of 100–500 fs, was assigned to the EPI mechanism [37–39]. The fitting results in figure 4 displays anomalies at $T_c$ and $T^*$ in the both the $A_c(T)$ and $\tau_\gamma(T)$ curves, corresponding to the anomalies at the same temperatures in the slow relaxation process. This reveals a strong correlation of SC and PG states with the EPI in Bi2212. The reason for this is that the phonon can be looked as one kind of medium, which is related to the phase fluctuation and the global phase coherence.

In EPI, the coupling constant $\lambda$ of Bi2212 can be characterized with the second moment

$$\lambda \langle \omega_{ph}^2 \rangle = 2 \int \alpha^2 F(\omega_{ph}) \omega_{ph} \omega_{ph} d\omega_{ph}$$

of the Eliashberg spectral function $\alpha^2 F(\omega_{ph})$ [28, 40]. A theoretical framework expressing the electron–phonon scattering time $\tau_{e-ph}$ in terms of $\lambda \langle \omega_{ph}^2 \rangle$ is often provided by the two-temperature model (TTM) [41] and nonequilibrium model (NEM) [42]. (Here subscripts e and ph refer to electron and phonon, respectively.) Generally, the choice of a model (TTM or NEM) for determining $\tau_{e-ph}$ is based on the relatively stringent requirement regarding the fluence dependence of $\tau_{e-ph}$. For TTM, it requires that $\tau_{e-ph} \ll \tau_{e-ph}$. Here $\tau_{e-e}$ is the electron–electron scattering time and $\tau_{e-ph}$ is electron–phonon scattering time. By assuming that the Fermi–Dirac distribution is created instantly following photoexcitation, the TTM often neglects e–e thermalization process, which lead to a discrepancy between the experimental results and the TTM model [43]. And Groeneveld et al ascribed this discrepancy mainly to the fact that the e–e and e–ph thermalization times are comparable at low temperature [44].

While, the deviation of the electron distribution from quasi-equilibrium population might not much influence on the energy relaxation under the high temperature condition. So that TTM is a reliable approximation at high temperatures. In addition, a fluence-dependent phonon mode has not been observed, and to the best of our knowledge has never been observed in cuprates. Here, we choose NEM to describe the fluence dependence of $\tau_{e-ph}$ in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$. For NEM, relaxation can be described with the kinetic Boltzmann equation using electron–electron and electron–phonon collision integrals [45], where the electrons and phonons are both out of equilibrium. The kinetic Boltzmann equation can be solved numerically [44] or analytically [42, 46]. The analytical treatment yields

$$\lambda \langle \omega_{ph}^2 \rangle = \frac{2\pi}{3} \frac{k_0 T_1}{\hbar \tau_{e-ph}}.$$  

(5)

Here $T_1$ is lattice temperature. To obtain an estimate of $\lambda$ from the experiments, we express the second moment of the Eliashberg function as the product of a dimensionless electron–phonon coupling constant.
and the square of a characteristic phonon frequency $\omega_0$: $\lambda \langle \omega_{ph}^2 \rangle = \lambda \omega_0^2$. The estimate of $\omega_0$ and then $\lambda$ requires detailed knowledge of the Eliashberg spectral function. This can be extracted from other experiments such as optical absorption [47], neutron scattering [48], ARPES [49–51], and tunneling [52–54]. For estimate, we use $\omega_0 \sim 40$ meV for Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$, and it follows that $\lambda = 0.25$ for the $\tau_f(T)$ at $T = 3$ K in figure 4(b).

4. Conclusion

In summary, the quasiparticle dynamics of cuprate Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ is investigated with femtosecond transient optical spectroscopy. The coexistence of the SC and PG states in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ is observed below $T_c$, and PG in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ can be explained as an incipient condensation of Cooper pairs with short coherence length. The fast decay time $\tau_f$, related to the EPI, shows anomaly near $T_c$ and $T^*$, suggesting the correlation of EPI and SC/PG states. The strength of electron–phonon coupling is calculated as $\lambda = 0.25$ at 3 K.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

ORCID iDs

Yong Li https://orcid.org/0000-0002-2059-5537
Weizheng Liang https://orcid.org/0000-0001-9244-6875
S N Luo https://orcid.org/0000-0002-7538-0541
Yong Zhao https://orcid.org/0000-0001-6375-0982

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