Direct observation of the coexistence of pseudogap and superconducting quasi-particles in Bi2212 by time-resolved optical spectroscopy

Y. H. Liu1, Y. Toda2, K. Shimatake2, N. Momono4, M. Oda1, and M. Ido1
1Department of Physics, Hokkaido University, Sapporo 060-0810, Japan
2Department of Applied Physics, Hokkaido University, Sapporo 060-0810, Japan
3Division of Innovative Research CRIS, Hokkaido University, Sapporo 001-0021, Japan
4Department of Materials Science and Engineering, Muroran Institute of Technology, Muroran 050-8585, Japan

(Dated: February 21, 2008)

We report the ultra-fast optical response of quasi-particles (QPs) in both the pseudogap (PG) and superconducting (SC) states of underdoped (UD) Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ (Bi2212) single crystal measured with the time-resolved pump-probe technique. At a probe energy $\hbar \omega_{pr}=1.55$ eV, it is found that the reflectivity change $\Delta R/R$ changes its sign at exactly $T_c$, which allows the direct separation of the charge dynamics of PG and SC QPs. Further systematic investigations indicate that the transient signals associated with PG and SC QPs depend on the probe beam energy and polarization. By tuning them below $T_c$ two distinct components can be detected simultaneously, providing evidence for the coexistence of PG and SC QPs.

PACS numbers: 68.37Ef, 74.72.Hs, 74.25.-q, 74.50.+r

The relationship between the anomalous PG state and high-$T_c$ superconductivity is still an important open issue in condensed matter physics. Two main scenarios have been put forward to help us to understand this relationship well within a theoretical framework. One proposes that the PG state is a precursor of high-$T_c$ superconductivity. The other emphasizes that the PG state is a precursor of high-$T_c$ superconductivity. In this picture, high-$T_c$ superconductivity originates from the PG state where Cooper pairs are formed but lack long-range phase coherence, which will be established below $T_c$. In early research, angle-resolved photoemission spectroscopy (ARPES) and electron tunneling spectroscopy revealed that the PG could smoothly evolve into the SC gap as the temperature fell below $T_c$, while electronic Raman scattering (ERS) data showed that nodal and antinodal gaps coexisted below $T_c$. In fact, this discrepancy can be understood well in terms of the scenario of the “Fermi-arc” superconductivity, which is supported by additional high-resolution data recently obtained again by ERS and by ARPES, revealing that for UD cuprate superconductors below $T_c$ there are two energy scales in the nodal and antinodal regions on the Fermi surface. Such a high consistency of different experimental data validates the contention that the SC gap located in the nodal region, called “Fermi-arc” superconductivity, is distinct from the PG located in the antinodal region for UD cuprate superconductors.

Distinguishing the charge dynamics of QPs in the antinodal and nodal regions will be helpful as regards revealing the mechanism responsible for them. Time-resolved pump-probe optical spectroscopy with femtosecond resolution is a powerful tool for determining the charge dynamics of carriers in superconductors. In such measurements, a pump pulse with a higher energy than the SC energy gap breaks Cooper pairs into two electrons (or photoexcited carriers) and excites them to a non-equilibrium high-energy state. Subsequently the probe pulse detects, within the delay time between the pump and probe pulses, the process by which photoexcited carriers recombine into a superconducting condensate. Below $T_c$, the PG does not change into the SC gap for UD cuprate superconductors, meaning that the relaxation process of SC QPs (Cooper pairs) should differ from that of PG QPs. Therefore, in principle this technique can be used to distinguish different relaxation processes involving PG and SC QPs in UD Bi2212 by measuring the sample reflectivity change $\Delta R/R$. Previously pump-probe experiments revealed that $\Delta R/R$ could be either positive or negative, indicating that $\Delta R/R$ of the UD Bi2212, using the same technique as described in Ref. [19], revealed that the two transitions can be selectively detected at temperatures far below the CDW transition temperatures. In this letter, we report the direct identification of the charge dynamics of PG and SC QPs by measuring $\Delta R/R$ of the UD Bi2212, using the same technique as that described in Ref. [19].

UD Bi2212 single crystals with $T_c=76$ K (hole concentration 0.11) are grown by the traveling solvent floating
zone method using an infrared image furnace. The time-resolved reflectivity change \( \Delta R/R \) is measured by using the pump-probe technique on the sample mounted in a cryostat whose temperature ranges from 4 to 300 K. Both the probe and pump beams are crossed and incident in the c-axis direction on freshly cleaved surfaces. The laser power induced heating effect has been accounted for by measuring the temperature dependence of the \( \Delta R/R \) amplitude at different given powers. The pump-probe experimental configurations have been described in detail elsewhere [19].

Figure 1(a) shows the temperature evolution of \( \Delta R/R \) as a function of delay time measured at a probe energy \( \hbar \omega_{pr} = 1.55 \text{ eV} \) and a pump energy \( \hbar \omega_{pu} = 1.07 \text{ eV} \) over a wide temperature range from 15 to 280 K. This waterfall plot shows that the sign changes of \( \Delta R/R \) occur just at the SC transition temperature \( T_c \) and the PG-opening temperature \( T^* \), respectively. It should be pointed out that the \( T^* \) determined here is consistent with the result of the tunneling spectroscopy [20]. The sign of \( \Delta R/R \) below \( T_c \) and above \( T^* \) is positive, whereas it is negative between \( T_c \) and \( T^* \). Hereafter, we define the positive (negative) signal as one with a positive (negative) sign. Using a single-component exponential decay function \( \Delta R/R(T, t) = A(T) \exp(-t/\tau) \), where \( A(T) \) is the amplitude of \( \Delta R/R \) as a function of temperature \( T \) and \( \tau \) is the relaxation time of QPs, we can fit the data well [solid lines in Figs. 1(b), (c) and (d)] and achieve the relaxation times for different QPs, whose temperature dependence is shown in Fig. 2(a). Considering the noticeable sign changes of \( \Delta R/R \) present at \( T_c \) and \( T^* \), we naturally assign the positive component appearing below \( T_c \) with a slow decay of \( \sim 2.5 \text{ ps} \) to SC QPs (Cooper pairs), which corresponds to the recombination time of Cooper pairs, consistent with those obtained on Y123 [18] and UD Bi2212 film [15] using two-component analysis. Another component that appears above \( T_c \) and fades out at \( T^* \) is ascribed to PG QPs with a relaxation time of \( \sim 0.5 \text{ ps} \). The third component appearing above \( T^* \) is a step-function response with a relaxation time of \( \sim 0.8 \text{ ps} \) [Fig. 1(d)], which is a typically bolometric effect in the metal [21] and similar to that observed in YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) (Y123) at 300 K [22]. This observation is consistent with ARPES results showing that UD Bi2212 is a metal above \( T^* \) [23]. In this letter, we focus our attention on the two components present below \( T^* \). The temperature dependence of the amplitude of \( \Delta R/R \) is shown in Fig. 2(b), which shows two features: (1) the amplitude of the positive SC component decreases with increasing temperature and becomes zero at \( T_c \); (2) the negative PG component starts to appear at \( T_c \) and fades out around \( T^* \).

Figures 3(a) and (b) show the effect of the probe beam polarization on \( \Delta R/R \) measured at \( \hbar \omega_{pr} = 1.55 \text{ eV} \) below and above \( T_c \). It is worthwhile noting that \( \Delta R/R \) shows no dependence on the pump beam polarization, which is consistent with previous reports on Y123 [18], but is closely related to that of the probe beam. Below \( T_c \) the transient \( \Delta R/R \) is independent of the probe polarization and two identical signals are obtained for \( E_{pr\parallel} \) and \( E_{pr\perp} \) (\( E_{pr\parallel} \) and \( E_{pr\perp} \) denoted as the probe polarizations parallel and orthogonal to the b axis of the crystal, respectively.). Above \( T_c \), however, the \( \Delta R/R \) measured with \( E_{pr\perp} \) is negative while that measured with \( E_{pr\parallel} \) is negligible, which may be owing to the anisotropy of the
to fit the SC and PG components, respectively, by fixing that used in Refs. [14, 15] and illustrated in the inset of spectively. In this case, the analysis method is similar to of two components that have slow and fast decays, re-
change of two fittings. It can be seen that this method works component to PG QPs. Below

equal to that measured at \( \hbar \omega_{pr} = 1.55 \) eV in the PG state shown in Fig. 1(c). Since each kind of QP corre-
sponds to one relaxation time, it is natural to assign this state shown in Fig. 1(c). Since each kind of QP corre-
proving this change in the PG component with probe energy.

Further investigations indicate that the transient \( \Delta R/R \) also strongly depends on probe energy \( \hbar \omega_{pr} \). In Figs. 3(c) and (d) are shown the data measured after exchanging the pump and probe energies used above, i.e. measured at \( \hbar \omega_{ps} = 1.55 \) eV and \( \hbar \omega_{pr} = 1.07 \) eV. For \( E_{pr \perp} \) the sign of \( \Delta R/R \) measured at \( \hbar \omega_{pr} = 1.07 \) eV is positive throughout the whole temperature range [Fig. 3(c)]. Above \( T_c \), the one-component exponential decay function \( \Delta R/R (T, t) = \Lambda(T) \exp(-t/\tau_{PG}) \) is used to fit the data well and we therefore assign the fast-decay component to PG QPs and the slow-decay component to SC QPs. For \( E_{pr \parallel} \) the detected signal consists of two distinct compo-
nents with opposite signs below \( T_c \), as seen in Fig. 3(d); the fast-decay component is positive and the slow-decay component is negative. A two-component decay function, \( \Delta R/R (T, t) = \Lambda(T) \exp(-t/\tau_{PG}) - \beta(T) \exp(-t/\tau_{SC}) \), is used to obtain a good fit with the data measured below \( T_c \), using parameters of \( \tau_{PG} \approx 0.5 \) ps and \( \tau_{SC} \approx 2.5 \) ps. One fitting curve is shown as an example in the inset of Fig. 3(d). With increasing the temperature, the amplitude of the negative component decreases and becomes zero at \( T_c \), suggesting that it originates from SC QPs. Above \( T_c \) only the positive component remains whose relaxation time is about 0.5 ps, suggesting that the positive component observed below \( T_c \) is due to PG QPs. Therefore, we directly observe the coexistence of PG and SC QPs in UD Bi2212, which have different relaxation dynamics. This time-domain observation of the coexistence of PG and SC QPs below \( T_c \) is in good agreement with real-space measurements such as those obtained with scanning tunneling microscopy/spectroscopy (STM/STS) with atomic resolution, which reveal that SC and PG gaps coexist in real space in UD Bi2212 [9, 25]. Here, we would like to emphasize that the coexistence of PG and SC QPs does not mean the “inhomogeneous phase separation” of the PG and SC states in real space. Recent STM/STS results suggest that both the PG and SC QPs should be distributed uniformly for a disorder-free UD Bi2212 sample below \( T_c \).
In order to obtain detailed information on the energy dispersion of the charge dynamics of PG and SC QPs below \( T_c \), we further investigate the effect of probe energy on the transient signal at 20 K, indicating that either the PG or SC component can be selectively detected at a specified probe energy. Figure 4(a) shows the signals measured at probe energies of 1.03 to 1.15 eV with \( E_{pr} \parallel \), which is normalized to the amplitude of the SC component to minimize the effect of the pump power on the signal. As is clearly seen, two components originating with SC and PG QPs can be simultaneously detected, as discussed above. More interestingly, with increasing signal. As is clearly seen, two components originating which is normalized to the amplitude of the SC component to minimize the effect of the pump power on the signal. As is clearly seen, two components originating with SC and PG QPs can be simultaneously detected, as discussed above. More interestingly, with increasing signal.

| \( \Delta R \) and \( \Delta R \) are obtained by fitting the data using the method \( \omega \perp \omega \parallel \) for \( \omega \| \omega \perp \parallel \), as shown in Fig. 3(c). Careful fitting analysis indicates that at \( h\omega_{pr} = 0.98 \) eV the PG component dominates the signal. Increasing the probe energy results in a decrease in the PG component fraction and an increase in the SC component fraction; in the probe energy range from 1.12 to 1.77 eV, only the SC signal is detected. This trend is illustrated in Fig. 4(d) by solid circles, where \( \Delta R\parallel \) and \( \Delta R\perp \) are obtained by fitting the data using the method described in the inset of Fig. 3(c). Note that the signals measured at \( h\omega_{pr} = 1.03 \) and 1.10 eV are similar to those measured previously for La214 and Y123 samples \[14, 27\] that consist of the mixed SC and PG components.

The most important finding of our present investigation is the direct observation of coexisting PG and SC QPs below \( T_c \) [Fig. 3(d) and Fig. 4(b)], which agrees well with findings obtained using electronic Raman scattering \[1, 2, 3\], ARPES \[4, 5, 6\] and STM/STS \[8, 9, 25\] that two energy scales coexist below \( T_c \). For UD Bi2212, the SC gap located in nodal regions is distinct from the PG in antinodal regions on the Fermi surface, which suggests that below \( T_c \) the PG does not evolve smoothly into the SC gap. As for the time-resolved pump-probe measurement, to our knowledge, it is the first time there has been an unambiguous and direct assignment of the relaxation processes associated with PG and SC QPs based on the sign changes of \( \Delta R/R \) at \( T_c \) and \( T^* \) [Fig. 1(a)], whose relaxation times are \( \sim 0.5 \) ps and \( \sim 2.5 \) ps, respectively. At present, however, it is still difficult to infer whether the PG state is a friend or foe of high \( T_c \) superconductivity \[28\].

However, the present investigations clarify an important long-standing puzzle with respect to the sign of the transient \( \Delta R/R \) measured by time-resolved optical spectroscopy. We verify that the sign of \( \Delta R/R \) depends both on \( h\omega_{pr} \) and on the probe beam polarization, which may be due to the anisotropy of the probe transition matrix elements and the inter-band transition probability \[24\]. Both strongly depend on the probe energy \( h\omega_{pr} \) and the band structure of the material \[24\]. Therefore, a single SC or PG component can be detected separately by tuning the probe polarization and energy. This method for separating the contributions of PG and SC QPs to \( \Delta R/R \) is useful for investigating the charge dynamics of QPs in other high- \( T_c \) superconductors in order to clarify the universal relationship between the SC and PG states.

This work was supported by the 21st century COE program “Topological Science and Technology” and Grants-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

---

[1] Electronic address: yhaoliu@hotmail.com

[2] M. Oda, N. Momono and M. Ido, Supercond. Sci. Technol. 13, R139 (2000); and references therein.

[3] M. Le Tacon et al., Nature Physics 2, 537 (2006).

[4] K. Tanaka et al., Science 314, 1910 (2006).

[5] T. Kondo et al., Phys. Rev. Lett. 98, 27004 (2007).

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

[7] M. Oda et al., J. Phys. Soc. Jpn. 69, 983 (2000).

[8] K. McElroy et al., Phys. Rev. Lett. 94, 197005 (2005).

[9] A. Hashimoto et al., Phys. Rev. B 74, 064508 (2006).

[10] S. D. Branson et al., Phys. Rev. Lett. 64, 2172 (1989).

[11] S. G. Han et al., Phys. Rev. Lett. 65, 2708 (1990).

[12] G. L. Eesley et al., Phys. Rev. Lett. 65, 3445 (1990).

[13] V. V. Kabanov et al., Phys. Rev. B 59, 1497 (1998).

[14] J. Demus et al., Phys. Rev. Lett. 82, 4918 (1999).

[15] H. Murakami et al., Europhys. Lett. 60, 288 (2002).

[16] N. Gedik et al., Phys. Rev. Lett. 95, 117005 (2005).

[17] T. N. Thomas et al., Phys. Rev. B 53, 12436 (1996).

[18] C. J. Stevens et al., Phys. Rev. Lett. 78, 2212 (1997).

[19] K. Shimatake, Y. Toda, and S. Tanda, Phys. Rev. B 75, 115120 (2007).

[20] R. M. Dipasupil et al., J. Phys. Soc. Jpn. 71, 1535 (2002).

[21] G. L. Easley, Phys. Rev. Lett. 51, 2140 (1983).

[22] A. Hashimoto et al., J. Phys. Soc. Jpn. 71, 1535 (2002).

[23] M. R. Norman et al., Nature 392, 157 (1998).

[24] D. Dvorscek et al., Phys. Rev. B 66, 020510(R) (2002).

[25] Y. H. Liu et al., Phys. Rev. B 75, 212507 (2007).

[26] Y. H. Liu et al., J. Phys. Chem. Solids, in press.

[27] S. Rast et al., Phys. Rev. B 64, 214505 (2001).

[28] M. R. Norman, D. Pines, and C. Kallin, Adv. Phys. 54, 715 (2005).