Momentum-Resolved Ultrafast Electron Dynamics in Superconducting Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$

R. Cortés,$^{1,2}$ L. Rettig,$^{1,3}$ Y. Yoshida,$^4$ H. Eisaki,$^3$ M. Wolf,$^{1,2}$ and U. Bovensiepen$^3$

$^1$Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany
$^2$Abt. Physikalische Chemie, Fritz-Haber-Institut d. MPG, Faradayweg 4-6, D-14195 Berlin, Germany
$^3$Fakultät für Physik, Universität Duisburg-Essen, Lotharstr. 1, D-47048 Duisburg, Germany
$^4$National Institute of Advanced Industrial Science and Technology, Tsukuba, Ibaraki 305-8568, Japan

(Dated: November 5, 2010)

The non-equilibrium state of the high-$T_c$ superconductor Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ and its ultrafast dynamical properties have been investigated by femtosecond time- and angle-resolved photoemission spectroscopy (trARPES) well below the critical temperature. We probe optically excited quasiparticles at different electron momenta along the Fermi surface and detect metastable quasiparticles near the antinode. Their decay through e-e scattering is blocked by a phase space restriction to the nodal region. The lack of momentum dependence in the decay rates is in agreement with relaxation dominated by Cooper pair recombination in a boson bottleneck limit.

PACS numbers: 74.47.J-, 74.25.Jb, 74.72.-h

The pairing mechanism responsible for the high-$T_c$ superconductivity in the cuprates remains one of the most challenging problems of current Solid State Physics, after more than two decades of research. In this context, angle-resolved photoemission spectroscopy (ARPES) has proven to be a very powerful experimental technique, providing information on the single-particle spectral function of these materials with a very high energy and momentum resolution [11]. However, it gives limited information on the coupling of single particle states with collective excitations, which seems to be essential to understand the ground state of the high-$T_c$ superconductors (HTSC) [12]. Additional information can be obtained from femtosecond (fs) time-resolved optical and THz techniques [3–10], which allow to study the quasiparticle (QP) interactions responsible for the relaxation of a photoexcited non-equilibrium state. This, in the case of the HTSC, is considered to provide a tool to study the interactions responsible for the pairing of QPs forming Cooper pairs. The analysis of QP decay dynamics has led to a controversy whether the decay follows a bimolecular recombination or proceeds in a boson bottleneck regime [12] [11]. However, these experiments inherently lack momentum resolution and thus can only be related to the electronic band structure in an indirect way. Finally, theoretical works related with these optical studies [11] [12] have provided further insight into the QP dynamics, but they have arrived to conclusions about the metastability of the nonequilibrium QPs which were up to now difficult to prove by experimental means.

Complementary to ARPES and all optical time-resolved techniques, femtosecond time- and angle-resolved photoemission spectroscopy (trARPES) provides momentum and energy resolved information on the single particle spectral function and its temporal evolution, allowing a direct investigation of the QP relaxation along the Fermi surface (FS). However, first investigations on cuprate superconductors using trARPES [13] did not develop a fully momentum-resolved study of their ultrafast dynamics nor used low enough excitation densities to avoid the instant vaporization of the superconducting condensate.

In this letter, we report on the ultrafast electron dynamics in superconducting Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi-2212) investigated at different points of its normal state FS by trARPES. Our data show that the density of nonequilibrium QPs created by photoinduced breaking of Cooper pairs is momentum dependent and related to the size of the superconducting gap. In contrast, the recombination rate of these QPs shows no sign of momentum or excitation density dependence. Our results provide experimental evidence of the transient stabilization of QPs off the node, due to scattering phase space restrictions caused by energy and momentum conservation in a d-wave superconductor. They also demonstrate that the net QP recombination rate in Bi-2212 is determined by the decay rate of the bosons emitted in this process (boson bottleneck).

The Bi-2212 samples studied in this work were nearly optimally doped single crystals with a transition temperature $T_c = 88$ K. They were cleaved in situ in ultrahigh vacuum ($\sim 8 \cdot 10^{-11}$ mbar) at 30 K, where the experiments were carried out. In the trARPES measurements the samples were excited by 55 fs laser pulses with a photon energy of 1.5 eV (pump beam), at 300 kHz repetition rate. Absorbed fluences, $F$, between 6 and 139 $\mu$J/cm$^2$ were used. The transient electron distribution was probed by time-delayed 80 fs, 6 eV laser pulses (probe beam) giving rise to photoelectrons, which were detected by a time-of-flight spectrometer. The energy resolution was typically 50 meV, the momentum resolution was $0.05 \text{ Å}^{-1}$ and the time resolution $< 100$ fs, see [14] for details. By means of a slanted sample holder [14] it was possible to reach points along the FS correspond-
ing to FS angles, $\phi$, between 45° (nodal point) and 18° (Fig.1(a)), in spite of the low photon energy of the probe beam.

The equilibrium electronic band structure around the Fermi level was studied by laser-based ARPES, using only the 6 eV beam. ARPES spectra (Fig.1(b)) were taken along arcs in the reciprocal space cutting the normal state FS (Fig.1(a)). In the spectra measured at the FS ($\phi = 27°$ and 63°) are highlighted with red thick lines in the lower panel. (c) ARPES spectra measured at $\phi = 18°$ as a function of the temperature.

FIG. 1. (Color online) (a) Sketch of the normal state FS of Bi-2212. Circles mark the FS angles, $\phi = 18°$, $27°$, $37°$ and $45°$, considered in this work. Some of the arcs cutting the FS along which the ARPES spectra were taken are also shown. (b) ARPES spectra and their representation as a false color plot, measured along the red arc in (a). The spectra measured at the FS ($\phi = 27°$ and 63°) are highlighted with red thick lines in the lower panel. (c) ARPES spectra measured at $\phi = 18°$ as a function of the temperature.

FIG. 2. (Color online) (a) trARPES spectra measured at several pump-probe delays. The depletion of the superconducting peak and the increase of spectral weight above the Fermi level, in relation to the spectrum measured before optical excitation, are shadowed in yellow (bright) and gray (dark), respectively; (b) Depletion of the superconducting peak (yellow (bright) area between the vertical gray lines in (a)) and increase of the intensity above the Fermi level (gray (dark) area in (a)) as a function of the pump-probe delay.

Dependent analysis of the evolution of that spectral weight at $E > E_F$.

trARPES spectra were measured at four points of the FS, with $\phi = 18°$, $27°$, $37°$ and $45°$ (Fig.1(a)), for different pump fluences, $F = 6, 14, 33, 139 \mu J/cm^2$. Next, the normalized trARPES intensity change with respect to the intensity before the arrival of the pump pulse, $\Delta I(t)/I$, was determined for $E > E_F$ (Fig.2). The decay of $\Delta I(t)/I$ in the measurements with $F \leq 33 \mu J/cm^2$ was fitted to a single-component exponential decay, $\Delta I(t)/I = A \exp(-t/\tau) + B$, convoluted with a 100 fs width Gaussian accounting for the time resolution. $A$ is the excitation amplitude, $\tau$ is the relaxation time of the nonequilibrium QPs and $B$ accounts for heat diffusion effects [19]. For larger fluences, an additional decay component with smaller $\tau$ is observed in $\Delta I(t)/I$, see the inset of Fig.3(b). We fit these data by a biexponential decay, accounting for the slow and fast component, see Fig.3.

First we focus on the slower contribution and its momentum dependence. Fig. 4 shows the momentum-dependent amplitudes $A$ and decay times $\tau$ obtained by fitting $\Delta I(t)/I$. All the fluences considered here show -- within error bars -- a constant $\tau \sim 2.5$ ps, see panel (a), and a decrease in $A$ with increasing $\phi$, panel (b). Albeit the error bars of $\tau$ increase for larger $\phi$ due to the simultaneous reduction in $A$ we can exclude that a similarly strong variation as in $A$, which is up to 8 times, occurs for $\tau$. We rather find that $\tau$ depends only very weakly or is even independent on the FS angle. In particular the
data for $F = 32 \mu J/cm^2$ support this conclusion.

Here $A$ represents the density of photoexcited quasiparticles, which shows a momentum dependence strikingly similar to the gap function of a $d$-wave superconductor. This can be understood by an indirect excitation process during which the order parameter is projected into the unoccupied electronic band structure. We now aim at an explanation of the processes active in the decay of that photoexcited state. Recalling that the pump-induced changes at $\phi = 27^\circ$ with $F = 139 \mu J/cm^2$ and $F = 33 \mu J/cm^2$, using a logarithmic vertical scale, is shown as an inset in (b). The fitting to exponential decays (see text) is shown as thin gray lines. In the inset, only the fitting of the spectrum measured at $33 \mu J/cm^2$ to a single-component exponential decay function for $t > 100$ fs is shown. To fit the spectrum measured at $139 \mu J/cm^2$ an additional component is needed.

FIG. 3. (Color online) Relative trARPES intensity change above the Fermi level, $\Delta I/I$, measured at different FS angles, $\phi$, using a pump fluence $F = 32 \mu J/cm^2$ (a) and measured at $\phi = 27^\circ$, using different pump fluences (b). A zoom of the spectra measured at $\phi = 27^\circ$ with $F = 139 \mu J/cm^2$ and $F = 33 \mu J/cm^2$, using a logarithmic vertical scale, is shown as an inset in (b). The fitting to exponential decays (see text) is shown as thin gray lines. In the inset, only the fitting of the spectrum measured at $33 \mu J/cm^2$ to a single-component exponential decay function for $t > 100$ fs is shown. To fit the spectrum measured at $139 \mu J/cm^2$ an additional component is needed.

Here $A$ represents the density of photoexcited quasiparticles, which shows a momentum dependence strikingly similar to the gap function of a $d$-wave superconductor. This can be understood by an indirect excitation process during which the order parameter is projected into the unoccupied electronic band structure. We now aim at an explanation of the processes active in the decay of that photoexcited state. Recalling that the pump-induced changes observed in Fig. 2(a) occur within $50$ meV around $E_F$, i.e. close to the superconducting gap, we take QPs with energies on the order of the gap or smaller into account. We first consider QPs at $\phi = 45^\circ$, i.e. at the node, where no gap is found, and illustrate QP relaxation in Fig. 3(c). At $T = 0$ K, QPs with infinitesimally small energy can only scatter with other QP exactly at $\phi = 45^\circ$, because at smaller $\phi$ the scattering partner cannot overcome the gap due to energy conservation. At higher $T$ QPs have larger energies and the scattering phase space is increased because the secondary QPs at other $k$ points can now overcome the gap near the nodal line. Far off the nodal line towards smaller $\phi$ this scattering channel is blocked for QP with energies about the gap size, as sketched in Fig. 3(d), process (1). Looking at our data we find that the amplitudes are actually larger far off the node and the decay is simply described for all momenta by a single exponential decay exhibiting constant $\tau$. Therefore, we find no indication of scattering towards the node. We conclude that albeit QPs just above the gap could gain energy through momentum redistribution (process (1), Fig. 3(d)) this channel is prohibited because the scattering partners required by momentum and energy conservation are not available. As a consequence QPs off the node become transiently stabilized, in agreement with the more sophisticated theoretical analysis of Refs. 4, 12.

Having excluded intraband e-e scattering as a relaxation channel for the QP population above the gap, we
face the question how to explain the observed 2.5 ps decay time. This time compares well to the one observed in earlier optical investigations [3, 6, 9, 10]. We recall that on this very same time scale the recovery of the superconductor in the strong bottleneck regime, as pointed out by an analytic solution [12].

However, such a dynamics can also be found in a strong bottleneck regime, as pointed out by an analytic solution [12].

Finally, we consider the second and faster component observed in the decay of $\Delta I(t)/I$ for $F = 139 \mu$J/cm². Although our work aims particularly on the slower component, we note that the fast decay contribution is connected to scattering with QPs excited near the node at these higher pump fluences [12] and/or to a partial evaporation of the SC condensate [8, 10]. However, further details are out of the scope of the current letter and will require additional studies as a function of the pump fluence and temperature.

In conclusion, we studied the momentum dependence of the transient population and decay times of photoexcited QPs in the high-$T_c$ superconductor Bi-2212 by means of femtosecond trARPES. We observe a transient stabilization of the photoexcited QPs created by Cooper pair breaking, which is explained by blocking of e-e scattering away from the node. The decay of these QPs is well described by a single exponential with momentum and pump fluence independent decay time, which demonstrates that Bi-2212 is in the boson bottleneck regime.

R.C. acknowledges the Alexander von Humboldt Foundation. This work has been funded by the DFG through BO 1823/2-2.

---

[1] A. Damascelli, Z. Hussain, and Z.-X. Shen, Rev. Mod. Phys., 75, 473 (2003)
[2] A. A. Kordyuk, S. V. Borisenko, V. B. Zabolotnyy, J. Heck, M. Knupfer, J. Fink, B. Büchner, C. T. Lin, B. Keimer, H. Berger, A. V. Pan, S. Komiya, and Y. Ando, Phys. Rev. Lett., 97, 017002 (2006)
[3] J. Demsar, B. Podobnik, V. V. Kabanov, T. Wolf, and D. Mihailovic, Phys. Rev. Lett., 82, 4918 (1999).
[4] N. Gedik, P. Blake, R. C. Spitzer, J. Orenstein, R. X. Liang, D. A. Bonn, and W. N. Hardy, Phys. Rev. B, 70, 014504 (2004)
[5] P. Kusar, J. Demsar, D. Mihailovic, and S. Sugai, Phys. Rev. B, 72, 014544 (2005)
[6] R. A. Kaindl, M. A. Carnahan, D. S. Chemla, S. Oh, and J. N. Eckstein, Phys. Rev. B, 72, 060510(R) (2005)
[7] N. Gedik, M. Langner, J. Orenstein, S. Ono, Y. Abe, and Y. Ando, Phys. Rev. Lett., 95, 117005 (2005)
[8] P. Kusar, V. V. Kabanov, S. Sugai, J. Demsar, T. Mentel, and D. Mihailovic, Phys. Rev. Lett., 101, 227001 (2008)
[9] Y. H. Liu, Y. Toda, K. Shimatake, N. Momono, M. Oda, and M. Ido, Phys. Rev. Lett., 101, 137003 (2008)
[10] C. Giannetti, G. Coslovich, F. Ciell, G. Ferrini, H. Eisaki, N. Kaneko, M. Greven, and F. Parmigiani, Phys. Rev. B, 79, 242502 (2009)
[11] V. V. Kabanov, J. Demsar, and D. Mihailovic, Phys. Rev. Lett., 95, 147002 (2005)
[12] P. C. Howell, A. Rosch, and P. J. Hirschfeld, Phys. Rev. Lett., 92, 037003 (2004)
[13] L. Perfetti, P. A. Loukakos, M. Lisowski, U. Bovensiepen, H. Eisaki, and M. Wolf, Phys. Rev. Lett., 99, 197001 (2007).

[14] F. Schmitt, P. S. Kirchmann, U. Bovensiepen, R. G. Moore, L. Rettig, M. Krenz, J. H. Chu, N. Ru, L. Perfetti, D. H. Lu, M. Wolf, I. R. Fisher, and Z. X. Shen, Science, 321, 1649 (2008).

[15] R. W. Schoenlein, W. Z. Lin, J. G. Fujimoto, and G. L. Eesley, Phys. Rev. Lett., 58, 1680 (1987).

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

[17] T. Dahm, V. Hinkov, S. V. Borisenko, A. A. Kordyuk, V. B. Zabolotnyy, J. Fink, B. Buechner, D. J. Scalapino, W. Hanke, and B. Keimer, Nature Physics, 5, 217 (2009).

[18] K.-P. Bohnen, R. Heid, and M. Krauss, Europhys. Lett., 64, 104 (2003).