Momentum-Resolved Ultrafast Electron Dynamics in Superconducting $Bi_2Sr_2CaCu_2O_{8+\delta}$

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The nonequilibrium state of the high-T_c superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$ and its ultrafast dynamics have been investigated by femtosecond time- and angle-resolved photoemission spectroscopy 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, since their decay toward the nodal region through *e-e* scattering is blocked by phase space restrictions. The observed lack of momentum dependence in the decay rates is in agreement with relaxation dynamics dominated by Cooper pair recombination in a boson bottleneck limit.

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The complete understanding of 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 [1]. 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) [1,2]. Additional information can be obtained from femtosecond (fs) time-resolved optical and THz techniques [3–10], which allow studying the quasiparticle (QP) interactions responsible for the relaxation of a photoexcited nonequilibrium state. Applied to the HTSC, such time-resolved techniques have investigated the microscopic processes that take part in photoexcited QP recombination into Cooper pairs (CPs). In fact earlier optical works attributed the observed time scales to such QP recombination [3–6,8–10]. But they have also lead to several controversies [4,6–8,11,12] due to the inherent lack of momentum resolution of these experiments, which only allows relating them to the electronic band structure in an indirect way.

Complementary to ARPES and all optical time-resolved techniques, femtosecond time- and angle-resolved photoemission spectroscopy (trARPES) allows overcoming these limitations as it provides both momentum and energy resolved information on the single-particle spectral function in combination with its temporal evolution. This facilitates a direct investigation of the QP relaxation along the Fermi surface (FS) just above the superconducting gap. However, first investigations on cuprate superconductors by trARPES only investigated the dynamics along the nodal direction [13,14].

In this Letter, we report on the ultrafast electron dynamics in superconducting $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) investigated at different points of its FS, from the nodal towards the antinodal point, by trARPES. Our data show that the density of nonequilibrium QPs created by photoinduced breaking of CPs is momentum dependent and related to the size of the superconducting gap. In contrast, the observed recombination rate of these QPs exhibits no sign of momentum 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.

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 ($\sim 8 \times 10^{-11}$ 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), from a Ti:sapphire amplifier operating 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 $Å^{-1}$ and the time resolution <100 fs, see [15] for details. By means of a slanted sample holder [15] it was possible to reach points along the FS corresponding to FS angles, ϕ, between 45° (nodal point) and 18° [Fig. 1(a)], despite the low photon energy of the probe pulses.

The equilibrium electronic band structure around the Fermi level was studied by laser-based ARPES, using

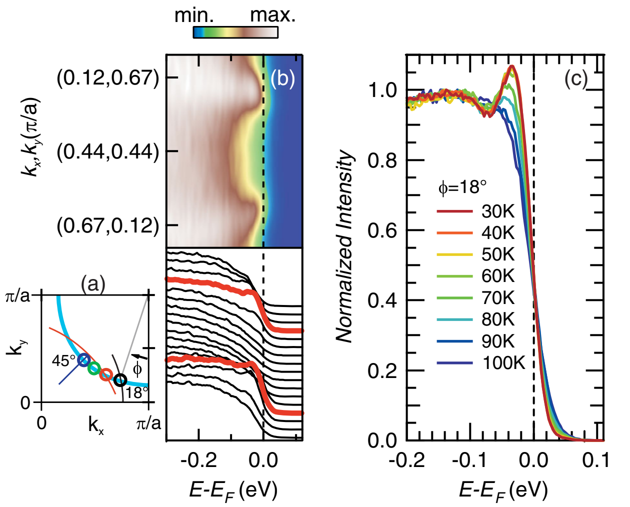

FIG. 1 (color online). (a) Sketch of the normal state FS of Bi-2212. Circles mark the FS angles, $\phi = 18°$, $27°$, $37°$ and $45°$, considered in this work. Some of the arcs cutting the FS along which the ARPES spectra were taken are also shown. (b) ARPES spectra and their representation as a false 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. The intensity of every spectrum has been normalized to its value at -0.15 eV.

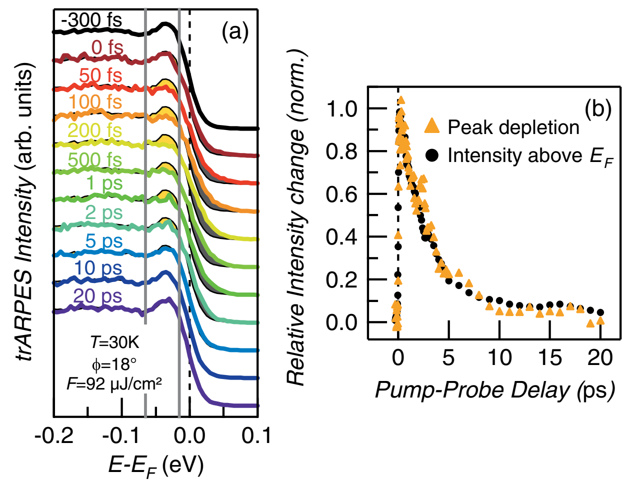

FIG. 2 (color online). (a) trARPES spectra measured at several pump-probe delays. The depletion of the superconducting peak and the increase of spectral weight above the Fermi level, in relation to the spectrum measured before optical excitation, are shadowed in yellow (bright) and gray (dark), respectively. (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.

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, a sharp peak is observed, which is known to be a direct consequence of the superconducting state [1]. The temperature dependence of this superconducting peak is shown for $\phi = 18°$ in Fig. 1(c). Its disappearance above T_c corroborates its relation with superconductivity.

The excitation of the sample by the 1.5 eV pump pulse produces a depletion of the superconducting peak, as well as an increase of the spectral weight above the Fermi level, E_F, [Fig. 2(a)] which are different than the ones produced by a mere increase of the temperature, see Fig. 1(c). We find that both quantities have the same evolution with the pump-probe delay [Fig. 2(b)]. This shows that the increase of the spectral weight at $E > E_F$ corresponds to the creation of a nonequilibrium density of QPs by breaking CPs and the decrease of that spectral weight can be attributed to the recombination of these QPs. Thus, our observations demonstrate that the time dependent spectral weight above E_F directly reflects the dynamics of the recovery of the superconducting condensate after photoexcitation. This result provides experimental justification for the description of the dynamics of the superconducting state in terms of the photoinduced QP density assumed in fs time-resolved optical and THz works [4,6,8,10,11]. We proceed now by a momentum-dependent analysis of the evolution of that spectral weight at $E > E_F$.

trARPES spectra were measured at four points of the FS [Fig. 1(a)], for four different pump fluences [see Fig. 3(b)]. 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. 3). Here, the intensity above E_F before the arrival of the pump pulse is mainly due to the finite energy resolution, which is larger than the thermal broadening at 30 K and the size of the superconducting gap at any ϕ. The decay of $\Delta I(t)/I$ in the measurements with $F \leq 32$ μJ/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, τ is the relaxation time of the nonequilibrium QPs and B accounts for heat diffusion effects [16]. For larger F, an additional decay component with smaller τ 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 (Fig. 3).

First we focus on the slower contribution and its momentum dependence. Figure 4 shows the momentum-dependent amplitudes A and decay times τ 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 pronounced decrease in A with increasing ϕ, panel (b). Albeit the error bars of τ increase for larger ϕ due to the simultaneous reduction in A, we can exclude that a similarly strong variation as in A occurs for τ. We rather find that τ depends only very weakly or is even independent on the FS angle. In particular the data for $F = 32$ μJ/cm^2 support this conclusion.

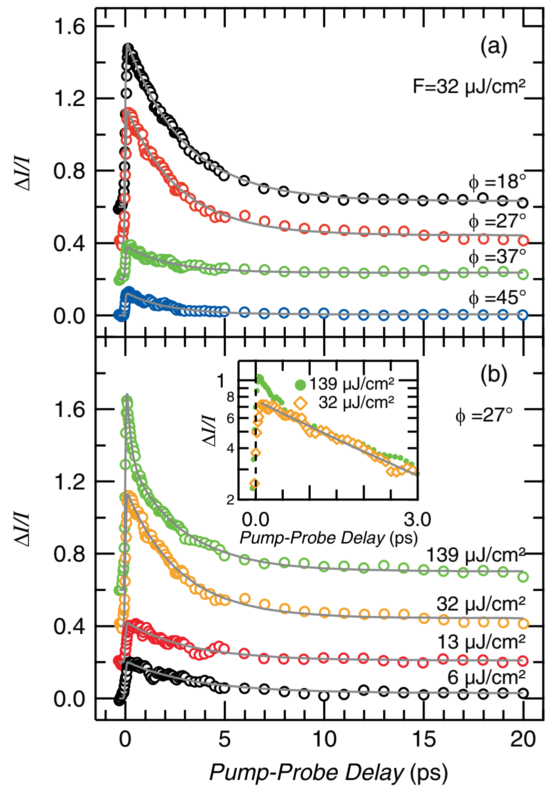

FIG. 3 (color online). Relative trARPES intensity change above E_F, $\Delta I/I$, measured at different FS angles, ϕ, using a pump fluence $F = 32\ \mu\mathrm{J/cm}^2$ (a) and measured at $\phi = 27°$, using different pump fluences (b). A zoom of the spectra measured at $\phi = 27°$ with $F = 139\ \mu\mathrm{J/cm}^2$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

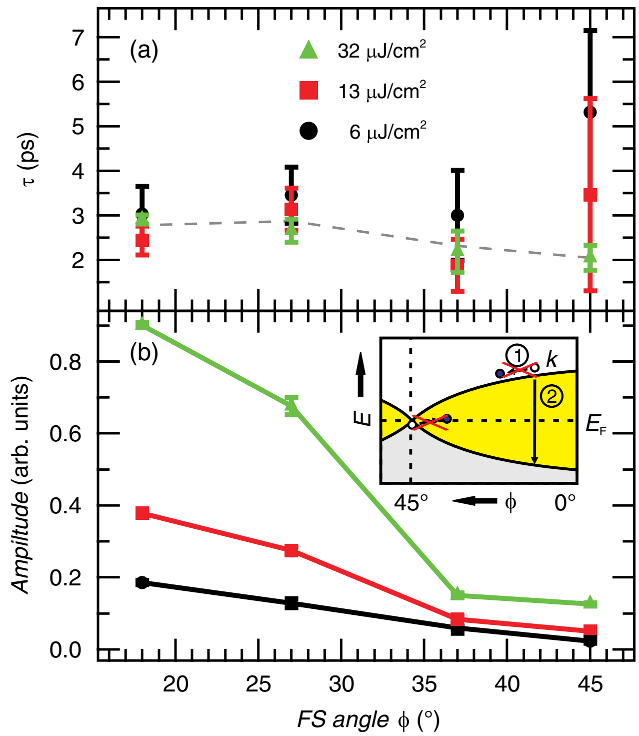

FIG. 4 (color online). Relaxation time (a) and excitation amplitude (b), obtained from the fitting to exponential decays (see text), as a function of the FS angle, ϕ. Lines are guides to the eye. The inset in (b) illustrates potential decay processes of photoexcited quasiparticles discussed in the text. The energy per pump pulse was kept constant, which lead in angle-dependent studies to a variation in F due to changes in reflectivity and illuminated area. The data were corrected accordingly.

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 suggests that the photoexcitation process promotes a fraction of CPs (their density decreases for larger ϕ towards the node) by *CP* breaking to unoccupied QP states just above the gap maintaining the initial momentum. In more detail, QPs are initially excited to energies up to the order of the pump photon energy and then relax within the first few hundred femtoseconds [17] by inelastic scattering processes, exciting new QPs to states just above the gap. In the following we discuss the decay processes of this low energy photoexcited state. Looking at our data we find that the amplitudes A are larger far off the node, while the decay is simply described for all momenta by a single exponential decay exhibiting constant τ. Therefore, we find no indication of preferential scattering between different points of the FS. This implies that albeit QPs just above the gap could gain energy through e-e scattering toward the node [process (1), inset of Fig. 4(b)] this channel is blocked. The reason might be the lack of the scattering partners required by momentum and energy conservation, due to the presence of the d-wave superconducting gap. As a consequence QPs off the node become transiently stabilized, in agreement with the conclusions of the theoretical analysis of Refs. [4,12], which were up to now difficult to prove by experimental means.

Having excluded scattering toward the node as an active relaxation channel for the QP population above the gap, we focus now on the observed momentum-independent decay time of 2.5 ps [Fig. 4(a)]. This value compares well to reports of earlier optical investigations [3,6,9,10]. We recall that on this very same time scale the recovery of the superconducting peak was observed (Fig. 2) and thus we can safely identify the decay of the QPs with recombination into CPs [process (2), inset of Fig. 4(b)]. This requires coupling of a QP with momentum $\hbar k$ with one at $-\hbar k$ and a boson with twice the gap energy. Still, the momentum independence of the decay times is intriguing. The increase in the gap function for smaller ϕ should favor higher energy gain near the antinode increasing the decay rate. Also the momentum dependence in the spin-fluctuation-mediated pairing interaction [18] and in e-ph coupling [19] suggests a variation of τ with ϕ. However, in a d-wave superconductor, the interaction of k-dependent QP recombination, CP breaking by the bosons emitted during the recombination, and the relaxation rate of those bosons might result in a compensating effect, leading to the observed momentum independence of τ. This scenario might be the one of the so called boson bottleneck regime, which appears in the context of the phenomenological Rothwarf-Taylor equations (RTE) [20].

The RTE are widely used to analyze CP recombination in fs time-resolved optical and THz studies [4,6,8,10,11]. At low T where the photoexcited QP density, n^*, is considerably larger than the thermal one, n_T, i.e. $n^* \gg n_T$, the relaxation of the low energy photoexcited QPs is governed by the probability to find a recombination partner leading to a recombination rate with a quadratic dependence on the QP density (second-order kinetics) [6,11]. At higher $T < T_c$, in the boson bottleneck regime, an exponential decay is predicted and experimentally observed [3,5,9]. It arises from the competition between CP recombination emitting a boson with twice the gap energy and CP breaking by that boson. In this regime the relaxation is determined by the decay rate of these bosons and thus it is independent of the excitation density. Alternatively, the exponential decay observed at higher T, where $n_T > n^*$, has been also explained by means of the RTE not as due to a boson bottleneck but due to the recombination of the photoexcited QPs with the thermal ones into CPs [6]. The decay that we find at 30 K is well described by a single exponential function exhibiting momentum and pump fluence independent decay times. However, it cannot be explained considering only recombination with thermal QPs. CP pair formation requires the two QPs involved in the process to have opposite momenta and, as most of the thermally excited QPs are in the nodal region, this would imply that the photoexcited QPs scatter toward the node before they recombine with the thermal ones, contrary to our observation. Therefore only the existence of a boson bottleneck can explain all our results and we can conclude that Bi-2212 is in the strong bottleneck regime. This challenges the conclusion of previous optical studies [6,7] about the absence of a bottleneck, based on the observation of a second-order kinetics. However, such a dynamics can also be found in a strong bottleneck regime, as pointed out by an analytic solution of the RTE [11].

Although a compensating effect in the boson bottleneck regime might explain the observed momentum-independent decay times, this regime is a specific case of the RTE. The RTE are the most suitable theoretical framework currently available to discuss these trARPES results, but they do not take into account momentum degrees of freedom, which are very relevant in this case. Therefore, the development of a more sophisticated theoretical description beyond the RTE, taking the symmetry of the order parameter into account, would be necessary.

Finally, we consider the second and faster component observed in the decay of $\Delta I(t)/I$ for $F = 139\ \mu\mathrm{J/cm}^2$. Although our work aims particularly on the slower component observed at low excitation densities, 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 transition to the normal state [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, which is explained by blocking of e-e scattering away from the node. The decay of these QPs is dominated by recombination into CPs and, in the low fluence limit, is well described by a single exponential with momentum and pump fluence independent decay time. This demonstrates that Bi-2212 is in the boson bottleneck regime.

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