

Quasiparticle dynamics across the full Brillouin zone of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ traced with ultrafast time and angle-resolved photoemission spectroscopy

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A hallmark in the cuprate family of high-temperature superconductors is the nodal-antinodal dichotomy. In this regard, angle-resolved photoemission spectroscopy (ARPES) has proven especially powerful, providing band structure information directly in energy-momentum space. Time-resolved ARPES (trARPES) holds great promise of adding ultrafast temporal information, in an attempt to identify different interaction channels in the time domain. Previous studies of the cuprates using trARPES were handicapped by the low probing energy, which significantly limits the accessible momentum space. Using 20.15 eV, 12 fs pulses, we show for the first time the evolution of quasiparticles in the antinodal region of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ and demonstrate that non-monotonic relaxation dynamics dominates above a certain fluence threshold. The dynamics is heavily influenced by transient modification of the electron-phonon interaction and phase space restrictions, in stark contrast to the monotonic relaxation in the nodal and off-nodal regions. © 2015 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [<http://dx.doi.org/10.1063/1.4933133>]

After more than 25 years of intense investigation, there is still no universal agreement on the coupling mechanism in the high-temperature cuprate superconductors. Key information comes from the coexistence, and possibly competition, between the superconducting (SC) phase and the pseudogap,¹⁻³ as well as other phases with translationally broken symmetry, in different regions of the Brillouin zone (BZ) and across the cuprates' phase diagram.^{4,5} To complicate matters, both phonon- and spin-mediated interactions,^{6,7} believed to be responsible for the emergent superconductivity, are predicted to vary substantially across the BZ, and especially in the nodal and antinodal regions. Angle-resolved photoemission spectroscopy (ARPES) has provided a great deal of experimental phenomenology, e.g., kinks in the band dispersion,^{8,9} viewed as fingerprints of the relevant many-body interactions, but so far has failed to reveal the nature of the electron-boson coupling in a uniform and compelling way.

A complementary view is offered by time-resolved techniques where different interactions can potentially be disentangled in the temporal domain. Ultrafast optical and terahertz

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techniques have provided a wealth of information,^{10–12} while being troubled by the inherent problem of relating the sample's reflectivity to the electron dynamics, and lacking momentum resolution. Moreover, the question of how exactly quasiparticle (QP) dynamics is related to the relevant order parameter is difficult to answer with optical methods. Expanding ARPES into the time domain offers the possibility to directly monitor QP and SC gap relaxation in energy-momentum space.¹³ Recent pioneering studies on the cuprates using 6 eV photons have been carried out in the nodal direction,¹⁴ challenging the nodal-antinodal dichotomy,¹⁵ and in the low perturbation limit, where surprisingly lacking,¹⁶ or very weakly-dependent momentum-resolved dynamics was observed.¹⁷ In order to properly map the QP and gap dynamics in cuprates, one needs to access information from the full BZ. Unfortunately, using only 6 eV photons does not allow one to probe the BZ boundaries of the cuprates, and key information from the true antinodal region remains elusive. A solution to this problem is offered by high-harmonic-generation (HHG) based time-resolved ARPES (trARPES),¹⁸ expanding the range of accessible energy and momentum, and offering exceptionally high temporal resolution. HHG trARPES has been demonstrated in studying ultrafast melting of charge-density waves,^{19,20} QP relaxation in heavy-fermion compounds,^{21,22} and graphene.^{23,24}

A promising route of investigating QP and order parameter response in the cuprates is offered by applying strong pump pulse at low temperature, sufficient to suppress SC non-thermally, while tracing its dynamical recovery on ultrashort time scales. Since the pair-breaking process is related to QP recombination by time-reversal symmetry, and therefore, both processes must involve the same mediating boson, strong perturbation of the cuprates may offer important new insights. This aspect has been exploited in all-optical experiments,²⁵ and recently using 6 eV trARPES.^{26,27} The body of work accumulated using 6 eV trARPES in the nodal and off-nodal regions indicates that above a critical fluence, sufficient to close the SC gap, a two-component QP dynamics emerges: a fast femtosecond dynamics appears, present for various doping levels, and similar to the relaxation above T_c , suggesting that the system is in a transient metallic state. The following picosecond dynamics, on the other hand, only present at low temperatures correlates very well with the re-entrance of the system in the SC state. The recovery rate of the SC gap appears to be momentum-independent, while as a function of increasing fluence, it increases as well. The QP recovery rate, on the other hand, shows clear momentum dependence, with relaxation away from the node occurring at a faster rate. The question of how QPs behave in the antinode where the SC is the largest has remained unanswered. Here, we employ a HHG-based trARPES to study the ultrafast electron dynamics in photoexcited $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi-2212), with the goal of answering this question and comparing the relaxation of QP from the nodal and antinodal regions in the BZ of the cuprates.

Single-crystal optimally doped Bi-2212 samples ($T_c = 91$ K) were cleaved *in situ* at a pressure of 7×10^{-11} Torr. The high-energy photons were produced via the process of high-harmonic generation by focusing 600 μJ pulses in a quasi-static gas cell backed by 30 Torr of argon. The 13th harmonic of an amplified Ti:Sapphire system (Red Dragon, KMLabs) at 20.15 eV was selected by a time-delay compensated double-grating monochromator, resulting in a flux of $\sim 10^{10}$ photons/harmonic/s.¹⁸ Both pump and probe beams were s-polarized. The laser system was operated at 10 kHz repetition rate precluding significant heating effects. Data were taken with a hemispherical analyzer (SPECS Phoibos 150); measurements from any given sample and orientation were limited to ~ 3 h, guaranteeing that sample deterioration was not an issue. We photoexcited the sample with 1.55 eV pulses at an absorbed fluence of 600 $\mu\text{J}/\text{cm}^2$ and probed the transient electronic states (Fig. 1(a)) with 12 fs extreme ultraviolet (XUV) pulses. To capture the evolution of the system from the most relevant portions of the cuprates' BZ, we collected data in the nodal, off-nodal (at $k_{\parallel} = 0.58 \text{ \AA}^{-1}$, perpendicular to the Γ -M direction, see inset in Fig. 3(b)) and antinodal regions. The time resolution of 35 fs was established by a cross-correlation measurement (between the near-infrared and XUV pulses) of the hot-electron response from a clean metal surface. This key feature, allowing for tracing ultrafast dynamics, is made possible by the broad bandwidth of the XUV pulse, 250 meV, which in turn determines the energy resolution of the experiment. This indicates that currently trARPES based on HHG sources is better suited to study QP dynamics rather than directly monitoring the

evolution of the SC gap; however, recent improvements indicate that improved energy resolution is possible in the near future. The spot size of the pump beam was set to $400\ \mu\text{m}$ and that of the probe beam to $100\ \mu\text{m}$. Equilibrium spectrum from the antinodal region at 13 K taken with the XUV beam only is shown in Fig. 1(b). The overall large-scale features are clearly reproduced;²⁸ the pronounced SC peak in the vicinity of the Fermi energy (FE) is resolved by using conventional Helium lamp source at very similar photon energy, 21.2 eV (He I).

In order to demonstrate the procedure for extracting information from the data, we show in Fig. 2(a), a representative contour plot in energy-time space from the off-nodal region after photoexcitation, restricted to $\text{FE} \pm 500\ \text{meV}$, and to time delays up to 1 ps. It clearly reveals the depletion of spectral weight below the FE, accompanied by a corresponding increase above the FE at time zero, which is understood in terms of creation of electron and hole distributions by the pump pulse. Taking two cuts in energy space, Fig. 2(b), the populations equally separated from the FE are seen to relax on identical time scales, in agreement with the symmetry of Bogolyubov states above and below the FE. Alternatively, cuts in the time domain, Fig. 2(c), reveal the redistribution of spectral weight following photoexcitation. In the following, we will concentrate exclusively on the electronic part.

In Figure 3, we present the evolution of QP in photoexcited Bi-2212 on a longer, picoseconds, scale from the nodal, off-nodal, and antinodal regions of the BZ, integrated in the range from FE to $\text{FE} + 500\ \text{meV}$. The QP relaxation in the nodal region is monotonic, as shown in Fig. 3(a), and it fits well to an exponential recovery with a time constant of $\tau = 640\ \text{fs}$ (see captions), followed by slower dynamics on the few picoseconds time scale, as expected from previous studies.^{14,15} A qualitatively similar but significantly faster loss of spectral weight ($\tau = 150\ \text{fs}$) is observed in the off-nodal region (Fig. 3(b)), in accord with recently observed dynamics.¹⁷ We cannot rule out the presence of a relaxation component on a longer time scale, which is commonly observed in other experiments; however, this is below the sensitivity of our apparatus. In stark contrast, the behavior of QP in the antinodal region, presented in Fig. 3(c), is substantially different: a slow build-up of the initial peak in the signal, occurring for $\sim 300\ \text{fs}$, is followed by ultrafast relaxation for $\sim 200\ \text{fs}$, resulting in a dip of the spectral weight, indicated by an arrow above the letter D. This trend abruptly changes and is followed by an increase of the QP density, peaking at 700 fs (indicated by an arrow above the letter P), followed by an exponential recovery with a time constant of 1.85 ps.

Qualitatively, the dynamics in the three regions can be understood by first noting that the pump pulse evaporates the SC condensate and induces a transition to a metallic-like phase.^{29–31}

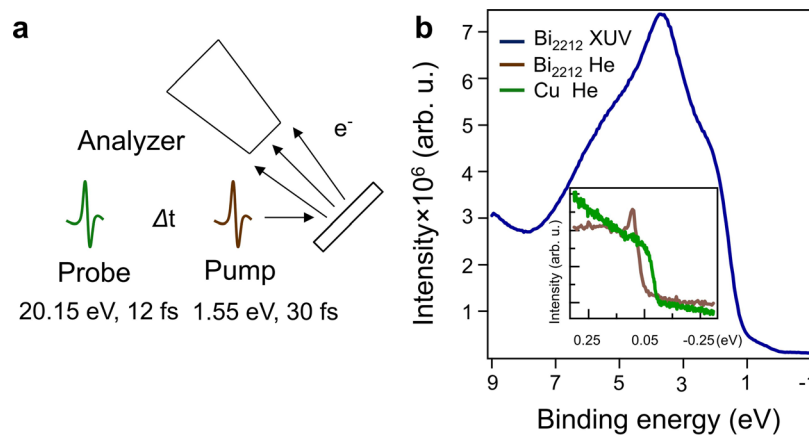


FIG. 1. Experimental geometry and static photoemission from Bi-2212. (a) Near-infrared pulses at 1.55 eV are used to photoexcite the sample. Following a controlled delay Δt , synchronized ultrafast extreme ultraviolet pulses at 20.15 eV are used to probe the transient electronic states by recording two-dimensional energy-momentum maps with an electron analyzer. (b) Angle-integrated photoemission spectrum from the antinodal region obtained using the laser-based XUV source only (blue). Inset shows the appearance of the SC peak (brown), downshifted from the Fermi energy, as measured from copper (green) with high-resolution (30 meV) He lamp ($\hbar\omega = 21.2\ \text{eV}$).

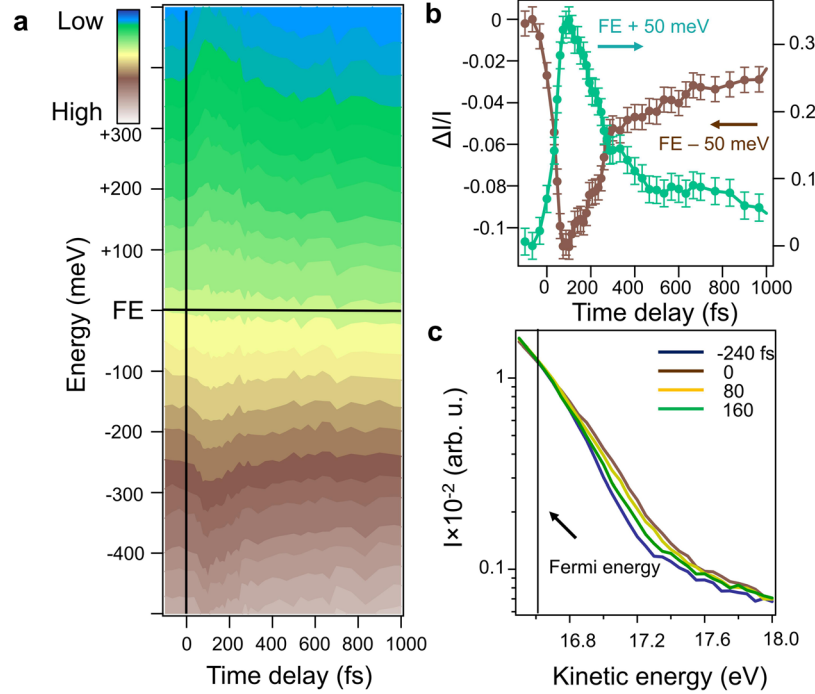


FIG. 2. Time-resolved photoemission spectra from the off-nodal region. (a) Two-dimensional contour map (raw data) of the optically induced modification of the photoemission spectra as a function of electron binding energy and time taken at 13 K and at a fluence of $600 \mu\text{J}/\text{cm}^2$. Horizontal and vertical black lines indicate the positions of the FE and time zero, respectively. The position of the FE was determined by noting the transition from hole-like to electron-like relaxation of the transient electronic structure. (b) Cuts in the horizontal direction representing the evolution of electrons (green) and holes (brown) separated by $\pm 50 \text{ meV}$ (this number represents the energy steps we used to collect data) from the FE. The vertical axis is the change in trARPES intensity normalized to the signal before the pump pulse. (c) Cuts in the vertical direction showing the change in photoemission spectra at various pump-probe delays.

The states in the nodal region are gapless both below and above T_c , allowing for efficient cooling of the electrons via interaction with strongly coupled phonon modes. This relaxation is followed by slower picoseconds dynamics dominated primarily by anharmonic phonon decay.¹⁴ The more rapid loss of QP spectral weight from the off-nodal region is tentatively assigned to an enhanced scattering at this wave vector, with the most likely candidate being a charge- or spin-density wave fluctuation.^{17,32} In contrast, the QP dynamics in the antinode differs

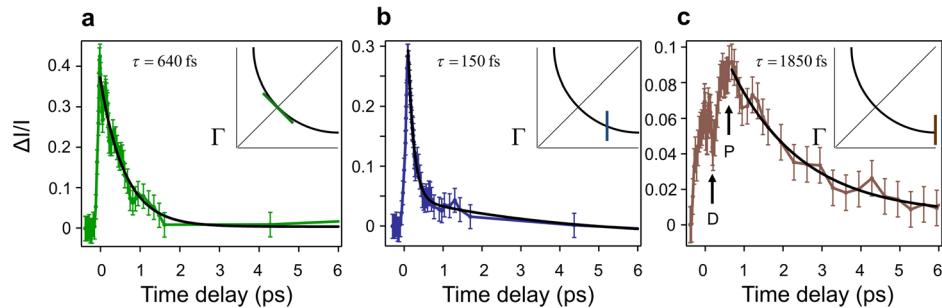


FIG. 3. Evolution of QPs from different regions of the Bi-2212 Brillouin zone. (a)–(c) Dynamics of electronic states, integrated up to 500 meV above the FE, from the nodal (a), off-nodal (b), and antinodal (c) regions taken at 13 K and at a fluence of $600 \mu\text{J}/\text{cm}^2$. Solid black curves are fits to the data at positive time delays using a double exponential decay function (except (c)), with the dominating time constant at short delays depicted. The insets show schematically the locations in k -space where data were taken. The error bars represent the standard deviation of the fits to the data. Letters D and P and the corresponding arrows in (c) indicate the positions where the dip (at 200 fs) and peak (at 700 fs) of the signal occur, as described in the text.

profoundly. An initial slow build-up of spectral weight around zero delay reflects the time necessary to destroy the SC order at the location where the gap is the largest. As the system undergoes a photoinduced transition to the normal phase, rapid QP cooling lasting about ~ 200 fs (Refs. 29 and 30) occurs, characteristic for metallic systems. An abrupt change of this trend, manifested by an increase of the QP signal, can be attributed to the “build-up” of QPs in the antinode. This situation resembles a bottleneck effect, where QPs cannot relax to lower energy states fast enough, and their density increases. This interplay leads to a maximum accumulation of QP weight at ~ 700 fs, after which monotonic relaxation is observed. We posit that this overall non-monotonic behavior is entirely due to non-thermal effects occurring only in the first picosecond after photoexcitation. We cannot assert that the dynamics on the long picosecond time scale can be attributed to the re-entrance of the system in the SC state since we drive the system far from equilibrium and we expect that following the ultrafast melting of the SC order the lattice will be found at an elevated temperature as well. However, this electron-lattice thermalization is expected to occur on the several picosecond scale; therefore, the non-monotonic behavior we observe in the antinode is purely a non-thermal effect. The significantly different relaxation of QPs from the most important regions of the BZ of the cuprates constitutes a novel observation of clear momentum-dependent dynamics, and adds a new dimension to the nodal-antinodal dichotomy.

Additional information about the QP dynamics can be inferred by tracing the QP population of different states above the FE on an ultrashort 1 ps scale, as shown in Figure 4. QP photoexcited well above the FE (FE + 500 meV) display dynamics that is quite similar in all investigated k -regions (bottom plots in Fig. 4). The behavior of QP at lower binding energy, albeit at energies significantly higher than the SC gap, varies significantly in different portions of the BZ. The speed of recovery of nodal QP is progressively reduced from $\tau = 169$ fs down to $\tau = 1033$ fs (Fig. 4(a)), while that of off-nodal QP is reduced only to about $\tau = 212$ fs. In addition, the peak of the signal in the off-nodal region appears ~ 100 fs after zero delay (Fig. 4(b)), indicative of a time delay necessary to populate these states with QPs. In general, a reduction of QP relaxation rate is expected for states closer to the FE due to phase-space restrictions;³³ however, the significant difference between the nodal and off-nodal decrease in spectral weight provides further support that the QP dynamics in the off-nodal region is due to a scattering mechanism, rather than direct QP recombination. Finally, the dynamics in the antinodal region evolve from monotonic for QP well above the FE energy to the complex double-peak structure for states closer to FE.

The observed momentum-dependent QP relaxation is expected to vary with the amount of energy deposited in the electronic subsystem by the pump pulse. To study this in more detail, we have performed a fluence-dependent study in the antinodal region where the effect of the SC gap dynamics is most obvious. At an absorbed fluence of $100 \mu\text{J}/\text{cm}^2$, the signal (Fig. 5(a), bottom) is seen to decrease monotonically, qualitatively similar to the dynamics in the nodal and off nodal regions, and in agreement with previous studies.⁶ At the higher fluence of $400 \mu\text{J}/\text{cm}^2$, a dip in the decay at 100 fs is observed followed by an increase of the signal, peaking at ~ 500 fs. These features are also observed at higher fluence ($600 \mu\text{J}/\text{cm}^2$) with the dip and the delayed peak appearing at progressively later times, 200 fs and 700 fs, respectively. This trend indicates that as the number of QP created by the pump beam increases, the system is driven further away from equilibrium, and more time is needed to relax from the metallic state; consequently, the bottleneck effect occurs at later times.

To study the influence of temperature on the observed electron dynamics, we collected data at 140 K when the system is in the normal state, and at an absorbed pump fluence of $600 \mu\text{J}/\text{cm}^2$. As evidenced in Fig. 5(b), the QP relaxation in the nodal and antinodal regions (integrated in the range from FE to FE + 500 meV) is very similar to each other in the normal state. This result is expected in the absence of the SC order, and it confirms that the presence of the d -wave SC gap is the dominant reason behind the momentum-dependent relaxation dynamics. This also suggests that the dynamics we observe are not related to the pseudogap phase.

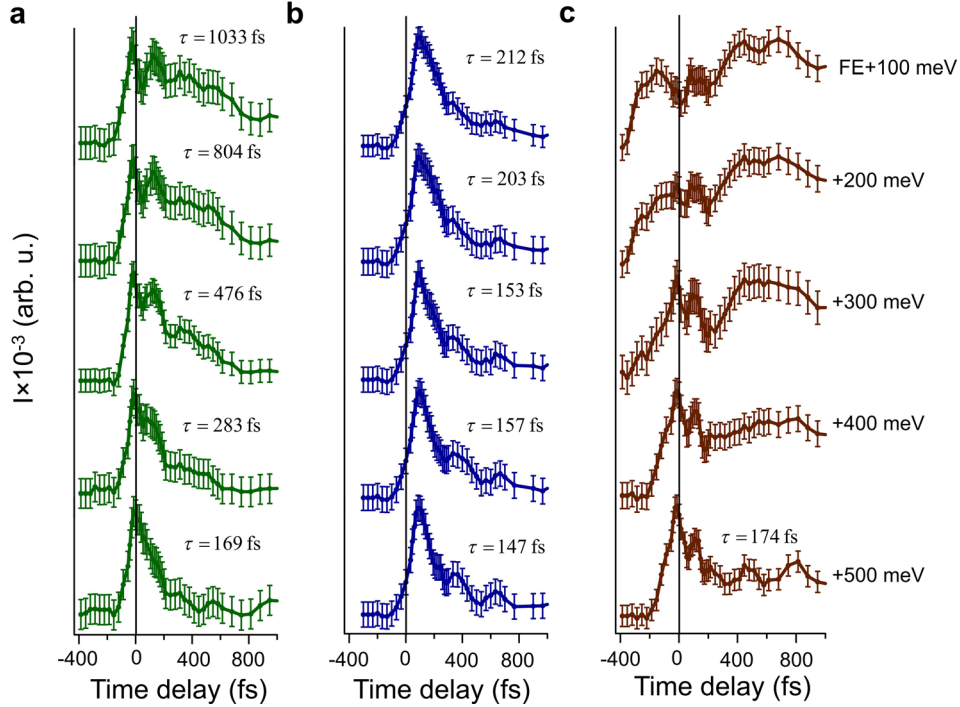


FIG. 4. Energy-resolved QP dynamics from different regions of the Bi-2212 Brillouin zone. (a)–(c) Relaxation of electronic states at different binding energy above the FE from the nodal (a), off-nodal (b), and antinodal (c) regions taken at 13 K and at a fluence of $600 \mu\text{J}/\text{cm}^2$. The scale of the vertical axes is the same as in Figure 3. Vertical lines indicate the position of zero delay. The time constants shown are extracted from fitting single exponential decay curves to the corresponding plots at positive time delays. The error bars represent the standard deviation of the fits to the data.

In general, the non-monotonic QP relaxation in the antinode above certain fluence level, and only below T_c , is the most interesting aspect of our work. In order to discuss this in more detail, we note that the dynamics of QP are governed, in our case, by transient modification of the band structure, coupled with time-dependent changes in all electron-boson coupling channels. These pump-induced processes act together to influence the recovery of the system back towards equilibrium. It is tempting to associate the picosecond dynamics that evolves after ~ 1 ps with the re-entrance of the system in the superconducting state, as the re-opening of the SC gap would act to “lift” QPs,²⁷ as observed at around 700 fs. However, due to the strong pump fluence used in our experiment, well-above the SC vaporization threshold,²⁹ the system is quite likely driven far out of equilibrium and SC probably is not re-established during the first few picoseconds that we are interested in. An alternative scenario involves the intrinsic inhomogeneity, well-known to exist in the cuprates.

Here, following the ultrafast evaporation of the SC state, the system is found as a mixture of two phases: a SC volume where slow, picosecond dynamics governs the QP relaxation, and normal, metallic phase where typical relaxation on the femtosecond scale occurs.³⁰ In this case the increase of spectral weight at ~ 700 fs is associated with the growth of fracture of the SC volume. However, it is difficult to corroborate this explanation experimentally, and hence, we propose that the re-opening of the SC gap occurs on a much later time scale; therefore, transient band structure modification is not the primary reason for the peculiar behavior of antinodal QPs. While at low fluence (Fig. 5(a)) and above T_c (Fig. 5(b)), QPs at all energy states manage to rapidly relax, as evidenced by the universal monotonic dynamics, in the high fluence case the QPs closer to the FE accumulate after several hundred femtoseconds. This also indicates that scattering of antinodal QPs to other regions in the BZ is not an efficient relaxation channel. Rather, the modification of the electron-boson coupling above a certain threshold of photogenerated QPs “prevents” efficient relaxation. We conclude that above this threshold QPs

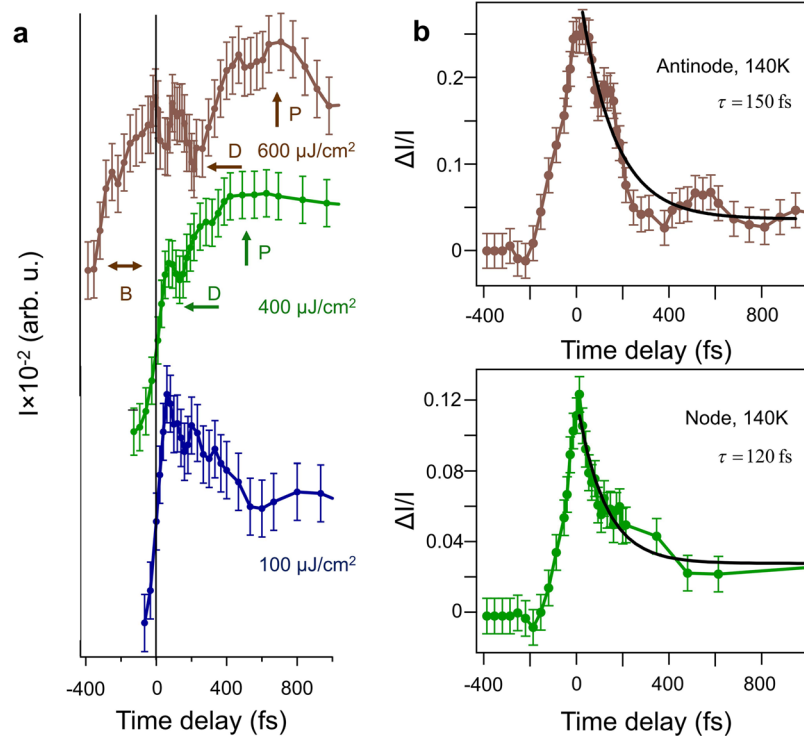


FIG. 5. Evolution of QP dynamics as a function of fluence and temperature. (a) Relaxation in the antinodal region at varying fluence. Letters D and P and the corresponding arrows indicate the positions where the dip and peak of the signal occur at different fluences. Letter B and the double-pointing arrow indicate the time interval (~ 300 fs) over which the SC signal grows. (b) Comparison of QP relaxation from the antinodal (brown) and nodal (green) regions at 140 K. The error bars represent the standard deviation of the fits to the data.

at lower energy states cannot relax by, e.g., emission of hot phonons until the density of these phonons decreases, quite likely due to anharmonic effects. The competition between QP relaxation from high energy states and restricted relaxation of QP from low energy states results in a bottleneck effect and the observed non-monotonic relaxation.

Finally, our results demonstrate an obvious failure of the widely used assumption of uniformly established transient electronic temperature following ultrafast optical excitation. The ultrafast modification of the electronic band structure quite possibly in tandem with the change in electron-phonon coupling results in QP recombination with very different rates across the Brillouin zone, as demonstrated in our experiment, leading to a variety of k -dependent effective transient temperatures. This indicates the inadequacy of the generally accepted trend of using an “elevated” temperature, based on the estimation of a “hot” Fermi function, extracted from fits in the nodal region, as our results show that this assumption is not applicable to the QP dynamics in the antinodal portion of the Brillouin zone. Therefore, any realistic attempt to simulate the dynamics of photoexcited QP should take into account the momentum dependence of the transient electronic temperature.

Our observation of distinct QP dynamics in the nodal, off-nodal, and antinodal regions on ultrafast timescales represents a novel piece of information about the femtosecond relaxation in cuprates driven far out of equilibrium. Through the combination of energy, momentum, and time sensitivity, we are able to show the significant differences in the relaxation dynamics between the nodal, off-nodal, and antinodal regions. We propose that the observed dichotomy in dynamics corresponds to the strongly momentum-dependent transiently modified electron-boson coupling, governing the evolution of the QP system on very short, ~ 1 ps time scales. This observation provides opportunity for future studies and applications of the dynamic properties of wide class of high-temperature superconductor materials.

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