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Ultrafast quenching of electron–boson interaction and superconducting gap in a cuprate superconductor

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Ultrafast spectroscopy is an emerging technique with great promise in the study of quantum materials, as it makes it possible to track similarities and correlations that are not evident near equilibrium. Thus far, however, the way in which these processes modify the electron self-energy—a fundamental quantity describing many-body interactions in a material—has been little discussed. Here we use time- and angle-resolved photoemission to directly measure the ultrafast response of self-energy to near-infrared photoexcitation in high-temperature cuprate superconductor. Below the critical temperature of the superconductor, ultrafast excitations trigger a synchronous decrease of electron self-energy and superconducting gap, culminating in a saturation in the weakening of electron–boson coupling when the superconducting gap is fully quenched. In contrast, electron–boson coupling is unresponsive to ultrafast excitations above the critical temperature of the superconductor and in the metallic state of a related material. These findings open a new pathway for studying transient self-energy and correlation effects in solids.

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The dynamics of electrons and atoms interacting with intense and ultrashort optical pulses is one of the emerging fields in physics. Strong optical pulses have been used as powerful tools to measure the electron–phonon interaction in solids^{1,2}, to investigate fundamental dynamical processes in semiconductors^{3,4} and to modulate the lattice structure of solids by creating dynamical states with new properties^{5–8}. These methods are particularly exciting in the context of correlated materials in which intense optical fields can drive a transition from an insulating to a metastable metallic phase⁹, can induce transient signatures of superconductivity¹⁰, can lead to anisotropic modulation of the electron–phonon coupling¹¹ and can disentangle the different dynamics in governing the superconducting and pseudogap phase of cuprates^{12–15}.

Despite the large amount of new physics revealed, most studies, thus far, use all-optical techniques that do not directly probe quasiparticles or carry any momentum information. As a consequence, the way fundamental quantities such as the electron self-energy and many-body interactions evolve outside equilibrium is often inferred indirectly. When probed in a time-resolved manner, these quantities have the potential to reveal insights on the microscopic properties of solids^{1,16}. Recent developments in high-resolution time- and angle-resolved photoemission spectroscopy (trARPES) now make these studies possible. So far, however, most of the trARPES studies have focused on recombination dynamics of photo-induced quasiparticle population^{17–21} and gap dynamics^{5,7,22,23}.

Here we present a study on the high-temperature cuprate superconductor $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi2212), and compare it with metallic $\text{Bi}_{1.76}\text{Pb}_{0.35}\text{Sr}_{1.89}\text{CuO}_{6+\delta}$ (Bi2201). In cuprate materials such as these, there is known to be a universal electron self-energy renormalization effect (a kink in the dispersion) that signifies the coupling of the electrons to bosons^{24–28}. However, whether this kink is related in any way to superconductivity is highly debated. Using trARPES, we tracked the temporal evolution of the electron self-energy renormalization and the superconducting gap after the system is perturbed by a femtosecond pump pulse. We found that, in superconducting Bi2212, the real part of the self-energy and the superconducting gap are markedly suppressed. Both effects also saturate at the same pump fluence. In contrast, in the normal state of Bi2212 and metallic heavily overdoped Bi2201, the suppression of the electron self-energy is far weaker. The results open a new avenue of investigation into self-energy and electron–boson interactions in solids.

Data were taken on an ultra-high resolution setup as previously described²⁹. In order to avoid the effects of photon-induced electric fields¹⁶, we limit our study to relatively low fluence (between 4 and $24 \mu\text{J cm}^{-2}$ or between 0.004 and 0.009 V/ a_0) and long time delay (≥ 300 fs). However, the fluence used in this study is still high enough to drive the full closure of the superconducting gap on the Fermi arc²³.

Results

Electronic dispersion. Figure 1a shows the equilibrium ($t = -1$ ps) and non-equilibrium ($t = 1$ and 10 ps) ARPES intensity as a function of energy and momentum for a nearly optimally doped sample measured with pump fluence $24 \mu\text{J cm}^{-2}$ at 17 K, far below T_c , along the nodal direction. The equilibrium spectrum shows the widely studied renormalization kink at ~ 70 meV (refs 24–27). Upon pumping, at a delay time of 1 ps, a clear loss of spectral weight can be observed, which is mainly confined between the Fermi energy and the kink energy¹⁸. It takes ~ 10 ps for the transient spectra to recover back to the equilibrium state. The effect of laser pumping on the dispersion is shown in Fig. 1b, where the equilibrium (black solid line) and transient (red solid

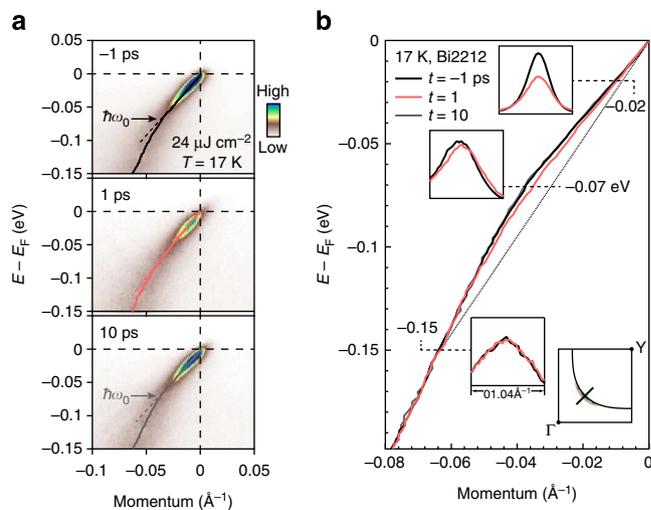


Figure 1 | Time-resolved spectra on a nearly optimally doped Bi2212.

(a) Equilibrium (before pumping, $t = -1$ ps) and transient (after pumping, $t = 1$ and 10 ps) photoelectron intensity (represented by false colour) as a function of energy and momentum measured along a nodal cut for a pump fluence of $24 \mu\text{J cm}^{-2}$. The bold solid black lines are the momentum distribution curve (MDC) dispersions at the corresponding delay time. The arrows mark the position of the kink at $\hbar\omega_0 \sim 70$ meV. (b) MDC dispersions for different delay times (-1 , 1 and 10 ps). Insets show comparisons of MDCs before pumping (-1 ps) and after pumping (1 ps) for a series of binding energies (-0.15 , -0.07 and -0.02 eV).

line) dispersions are compared. The dispersions are extracted in the standard way by fitting momentum distribution curves (MDCs) to a Lorentzian functional form³⁰. The comparison between the dispersion curve after a long delay time ($t = 10$ ps; grey line) with the equilibrium curve provides an estimate of our error bars. The most obvious pump-induced change occurs near the kink energy, as shown by the shift of MDC peak position in this energy range, in contrast to the MDCs near the Fermi level or at much higher binding energies, where the shift is negligible (see inset). Specifically, for delay time $t = 1$ ps, the Fermi velocity increases by 0.13 eV \AA (equilibrium 1.87 eV \AA) at binding energy below the kink energy and remains approximately unchanged above, resulting in an apparent softening of the kink strength (such as coupling strength) as pumping is turned on. The Fermi velocity is extracted from the slope in the dispersion between ~ 70 meV and E_F , as $v = dE/dk$ ³¹.

Self-energy changes below T_c . The differences between equilibrium and transient spectra can be analysed by extracting the electron self-energy $\Sigma = \Sigma' + i\Sigma''$, shown in Fig. 2. To extract the effective real part of the self-energy at different delay time, we took measured dispersions and subtracted featureless linear bare bands with the same velocity at each delay time (see, for example, the dotted line in Fig. 1b). Such techniques are commonly used to analyse the electron–boson interaction at equilibrium^{24,30}. At equilibrium (black dots) Σ' is reminiscent of a spectrum of modes, as previously reported^{26,27,32,33}, and its maximum is at the 70 meV kink position. The most significant pump-induced effect is the suppression of Σ' in the proximity of the kink energy (between 40 and 90 meV; Fig. 2a, red dots), in line with a softening of the coupling strength, as shown in Fig. 1. This suppression intensifies as fluence increases, and eventually saturates when superconductivity is completely suppressed (Fig. 3), as we will discuss later. Within our resolution, we observe no pump-induced energy shift in the peak position of Σ' (ref. 34). In the same

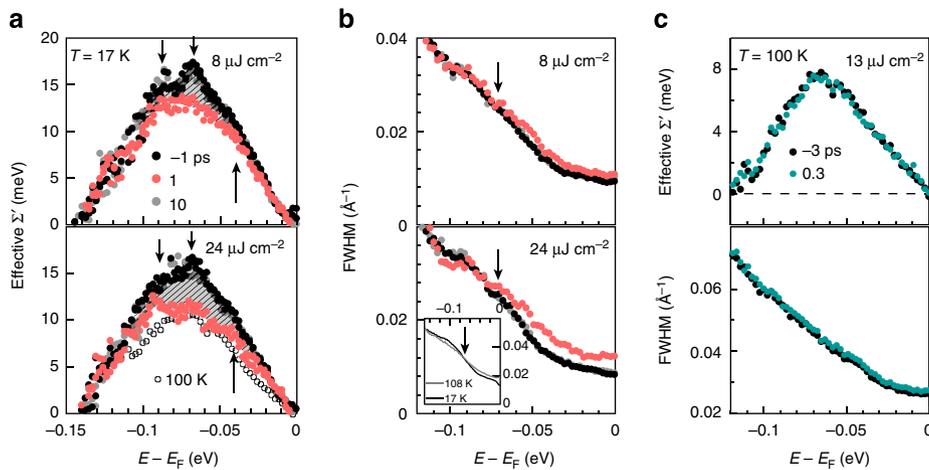


Figure 2 | Equilibrium and transient nodal electron self-energies. (a) Real part of electron self-energy (Σ') at different delay times ($t = -1, 1$ and 10 ps) for pump fluences 8 and $24 \mu\text{J cm}^{-2}$ measured at 17 K (below T_c). Σ' measured at equilibrium temperature 100 K (above T_c) is plotted in the lowest panel for comparison. (b) The corresponding MDC width as a function of energies. The black arrows mark the energy $\sim \hbar\omega_0$ where non-equilibrium and equilibrium MDC width separate with each other. Inset in the lowest panel shows non-equilibrium MDC width at 17 and 108 K (ref. 32). (c) Σ' and MDC width, similar to that shown in a,b but measured in the normal state with pump fluence $13 \mu\text{J cm}^{-2}$, at an equilibrium temperature of 100 K.

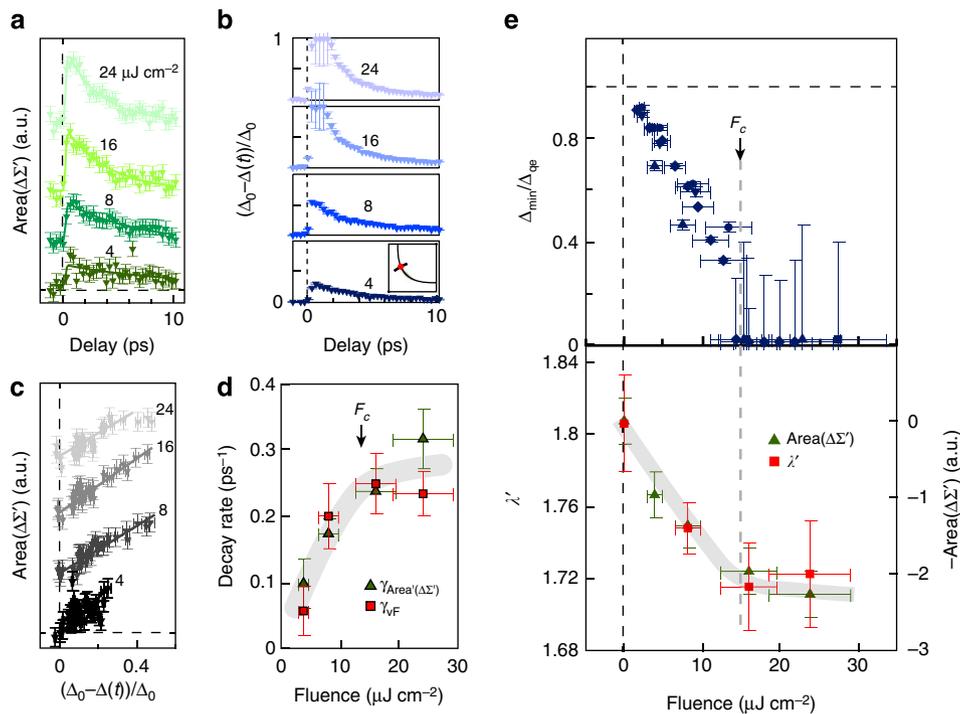


Figure 3 | Dynamics of pump-induced change. (a) Pump-induced change of nodal Σ' (vertically offset for clarity) as a function of delay time for different fluences. The error bars of $\text{Area}(\Delta\Sigma')$ are estimated by the maximum difference of $\text{Area}(\Delta\Sigma')$ among the measurements in equilibrium state. (b) Gap versus pump-probe delay time for an off-nodal cut (inset) at different fluences. The changes of energy gap are normalized to the maximum gap. (c) Pump-induced change of the Σ' (offset for clarity) as a function of the energy gap at each delay time as extracted from a,b for four different fluences. Because of the limited energy resolution (23 meV, comparable with the equilibrium gap size), only energy gap changes $< 50\%$ are reliable and shown. (d) The recovery rate of nodal $\Delta\Sigma'$ and the nodal coupling strength λ' as a function of fluence. (e) The pump-induced non-equilibrium minimum energy gap, maximum changes of $\text{Area}(\Delta\Sigma')$ and electron-boson coupling strength at 70 meV as a function of pump fluence. The coupling strength is given by $\lambda' = v_0/v_F$ as functions of pump fluence (v_0 (v_F), bare (Fermi) velocity below (above) the kink energy). The error bars of the change of Σ' and λ' are absolute maximum variations before $t = 0$, and the others are s.d. from fitting.

figure (bottom panel of Fig. 2a), we directly compare the equilibrium Σ' at 100 K with the pump-induced Σ' at a similar electronic temperature as measured from the width of the Fermi edge^{17,18,23}. While the effect of temperature on the equilibrium

Σ' extends over the entire energy range, the effect of pumping is smaller overall and is mainly confined within the kink energy (see also Supplementary Fig. 1). This suggests that optical pumping induces an effect beyond increasing the temperature.

In Fig. 2b, we show the imaginary part of the self-energy, Σ'' , that is proportional to the full-width at half maximum of the MDCs. In agreement with Σ' , the main pump-induced change is confined between the kink energy and the Fermi level (vertical black arrow) and increases as fluence increases. This is in contrast to thermal effects, where the temperature causes a change of Σ'' over the entire energy window (see inset in Fig. 2b and Supplementary Fig. 1). In Fig. 2c, we show the comparison of equilibrium and transient Σ' and Σ'' at equilibrium temperature 100 K, above T_c . In sharp contrast with the low-temperature behaviour, the pump-induced changes of the self-energy are negligible in the normal state up to the highest applied fluence $24 \mu\text{J cm}^{-2}$ (we note, however, that this might no longer be valid at very high fluences, which would substantially affect the entire band structure^{16,35}). These results might suggest that the pump-induced changes of self-energy are sensitive to the presence of superconductivity and are not induced by trivial thermal broadening effects.

We note that the absence of a shift in the kink energy, when the superconducting gap closes, suggests that the electron–boson interaction falls beyond the standard Migdal–Eliashberg theory for superconductivity as also suggested by equilibrium ARPES experiments^{36,37}. Indeed, within this standard theory, the energy dispersion has a square-root divergence at energy $\Delta_0 + \Omega$ in $\Sigma'(\omega)$ (Ω is the boson energy and Δ_0 is the superconducting gap at zero temperature)³⁸. Therefore, when the pump drives the superconducting gap to zero, one expects a shift of the kink energy and of Σ' towards lower binding energy by the magnitude of the superconducting gap, even for the nodal cut³⁹.

Self-energy versus superconducting gap. To further investigate this matter, we utilize the unique advantage of trARPES by simultaneously monitoring the pump-induced changes in both the electron self-energy and the superconducting gap. Figure 3 compares the pump-induced change in Σ' and the non-equilibrium superconducting gap. The latter is measured for a cut on the Fermi arc (see inset of Fig. 3b). Energy gaps at each delay time are obtained by fitting symmetrized energy distribution curves to an energy-resolution-convolved phenomenological BCS model⁴⁰, which is widely used in characterizing the energy gap in

cuprates^{23,41} (see also Supplementary Fig. 2). The temporal evolution of the area of $\Delta\Sigma'$ (hatched area in Fig. 2a) and the superconducting gap for different fluences are shown in panels a and b of Fig. 2, respectively. In agreement with Fig. 2, Σ' is weakly affected at low fluences and shows substantially slower initial recovery rate (0.1 ps^{-1}) than at higher fluences (0.32 ps^{-1} ; Fig. 3d). Similarly, pumping only weakly affects the superconducting gap at low fluence (bottom measurements in Fig. 3b) and eventually drives it to a full closure at high fluences (top measurements)^{20,23}. A similar fluence dependence is also observed for the Fermi velocity (Supplementary Fig. 3). The pump-induced change in the self-energy and the non-equilibrium superconducting gap at each delay time are plotted in Fig. 3c for several fluences, showing an unexpected linear relation for all fluences. At the highest fluence, a small deviation from linearity is observed, possibly because of additional broadening of the spectra, causing an underestimate of the superconducting gap.

In Fig. 3e, we show the fluence dependence of the maximal near-nodal gap shift (Fig. 3b) and compare it with the area of the maximal self-energy change (from Fig. 3a) and the coupling constant. In a simple electron–boson coupling model⁴², the coupling strength λ is directly related to the bare and dressed Fermi velocities according to $\lambda' \equiv \lambda + 1 = v_F/v_0$. Alternatively, the change in λ can be approximated by the integral of the self-energy change between equilibrium and pumped values (see hatched areas in Fig. 2) because near the Fermi energy $\lambda = \lim_{\omega \rightarrow 0} \partial\Sigma'/\partial\omega$, namely, $\Sigma'(\omega) \approx \lambda\omega$, thus $\int_0^{\omega_c} \Delta\Sigma'(\omega)d\omega \approx \Delta\lambda\omega_c^2/2$. Both characterization methods are shown (for more details, see also Supplementary Fig. 3). Interestingly, as we drive the superconducting gap to a gradual melting by increasing the excitation density, we find that the electron–boson interaction decreases in a similar fashion and eventually saturates (see also Supplementary Fig. 4) when the superconducting gap is fully quenched. The saturation effect is in contrast with thermal broadening, where continuous smearing of the kink occurs as the temperature gradually increases³².

Figure 3d shows the recovery rate of the integrated self-energy change (from Fig. 3a) and λ' (Supplementary Fig. 3). In the low-fluence regime, we find that both of these rates increase linearly with fluence in a similar fashion to the bimolecular recombination of non-equilibrium quasiparticles^{20,43}. As the fluence

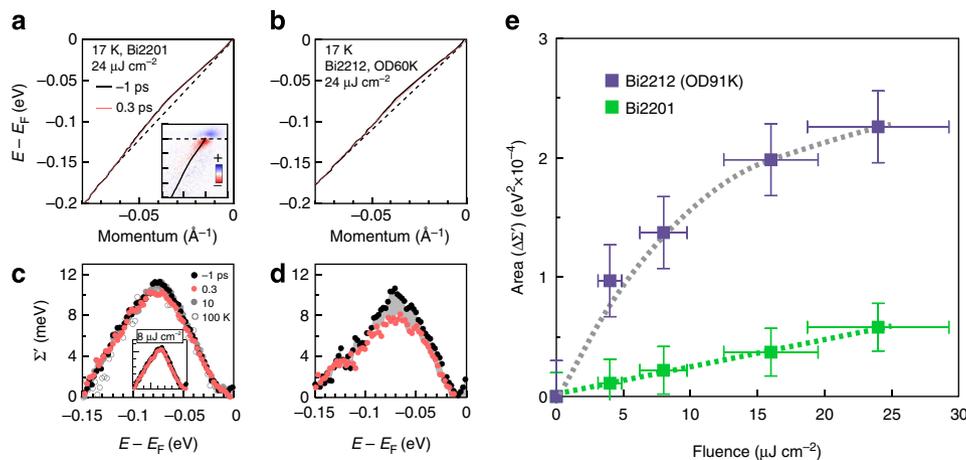


Figure 4 | Results on a heavily overdoped Bi2201 and an overdoped Bi2212. (a,b) MDC dispersions of equilibrium ($t = -1 \text{ ps}$) and non-equilibrium ($t = 0.3 \text{ ps}$) states, for a pump fluence of $24 \mu\text{J cm}^{-2}$ for the overdoped Bi2201 and the overdoped Bi2212, respectively. False-coloured inset in **a** shows the difference between equilibrium ($t = -1 \text{ ps}$) and non-equilibrium ($t = 0.3 \text{ ps}$) raw spectral image. (c,d) Real part of the electron self-energy (Σ') at different delay time ($t = -1, 0.3$ and 10 ps) for pump fluence $24 \mu\text{J cm}^{-2}$ for the overdoped Bi2201 and the overdoped Bi2212, respectively. The equilibrium Σ' measured at 100 K on the overdoped Bi2201 is represented by empty circles for comparison in **c**. The inset in **c** shows Σ' spectra for a pump fluence of $8 \mu\text{J cm}^{-2}$. (e) Fluence dependence of the Area($\Delta\Sigma'$) for the optimally doped Bi2212 and heavily overdoped Bi2201. The bold lines are guides to the eye. The error bars in **d** are absolute maximum variations before $t = 0$.

approaches the critical fluence, both rates saturate, marking the onset of different recombination processes, a behaviour that appears to be dictated by the closing of the superconducting gap at F_c . The behaviour is also consistent with previous reports of non-equilibrium quasiparticle recombination in an optimally doped and underdoped Bi2212 (refs 20,21), and with a time-resolved optical reflectivity study⁴⁴.

Self-energy changes above. To better understand the nature of the reported pump-induced change in the self-energy of Bi2212 superconductor, in Fig. 4, we study the effect of pumping on the electron–boson coupling in a metal in the same low-fluence regime and at the same low temperature. To make a meaningful comparison with Bi2212, we report data for an heavily overdoped Bi2201 ($T_c < 2$ K). Bi2201 has very similar crystallographic and electronic structure to Bi2212, and an only slightly weaker 70 meV dispersion kink²⁵. Moreover, Bi2201 can be grown in the heavily overdoped regime, with T_c lower than a few Kelvin, making it possible for us to study its normal state at 17 K, the same temperature as the study of optimally doped Bi2212.

Figure 4a shows the pump-induced changes in the electronic dispersion of heavily overdoped metallic Bi2201 along the nodal direction. Although qualitatively the pump-induced changes of spectral intensity are similar to those in superconducting optimally doped Bi2212 (ref. 18), there are significant differences between the two. Despite the apparent kink feature, the pump-induced changes in Σ' are negligible at lower fluences (inset of Fig. 4c), and show a very small increase with fluence over the entire range. Note that, consistent with the notion that the critical fluence marks the threshold above which all Cooper pairs are broken²¹, no critical fluence is observed in the Σ' in the normal state of this heavily overdoped sample (Fig. 4e). This difference is not likely owing to the weaker kink in Bi2201. Indeed, at the highest fluence (Fig. 4c), the change of Σ' is still at least a factor of 2 smaller than the corresponding change observed in overdoped Bi2212—displayed in Fig. 4d—despite the comparable kink strength. The change of Σ' in Bi2201 is a factor of 4 smaller than optimally doped Bi2212, in which the kink strength is larger only by a factor of 40% (see Fig. 4e and the coupling strength in Supplementary Fig. 5). In contrast to Bi2212, the pump-induced changes in Bi2201 can be accounted for by thermal smearing of the dispersion with temperature^{33,45}, as shown by the comparison of equilibrium and transient self-energy at an equilibrium temperature of 100 K. We caution that these considerations apply to the low pump fluence regime. This behaviour is also consistent with the expected behaviour for a metal in which temperature smears the kink feature and reduces the magnitude of the maximum of Σ' without any change in the electron–boson coupling³⁸.

Discussion

The effects observed in this paper cannot be simply explained by transient heating. Indeed, a comparison at similar temperatures between the temperature dependence of the transient self-energy and the equilibrium self-energy shows quite different trends. At equilibrium the temperature-dependent self-energy changes over a broad range of energies³², in contrast to the transient self-energy, where the changes are confined to the vicinity of the kink energy. In addition, the equilibrium self-energy gradually changes as the equilibrium temperature increases, and the effect persists even above T_c . In contrast, the pump-induced change in the self-energy saturates at the critical fluence and does not show any change above T_c . Moreover, the temperature dependence of the density of the initial non-equilibrium quasiparticles population is not proportional to the inverse of the specific heat²¹, as expected

in the case of thermal dynamics. Finally, the observation that bimolecular recombination dominates quasiparticle scattering processes in the low-fluence regime further supports the non-thermal nature of the reported effects^{20,43}.

These observations, together with the negligible pump-induced effect in the normal state and in a metallic compound (Fig. 4) validate the intrinsic nature of the pump-induced phenomena reported here. Whether these effects are due to a change of the electron–boson coupling constant as in an equilibrium picture or are a consequence of photo-induced delocalization of charge that leads to a reduction of the Jahn–Teller effect^{46,47} and to an apparent softening of the electron–boson interaction, they suggest the exciting possibility that the effects responsible for the self-energy renormalization are an important booster of a large pairing gap^{26–30}. trARPES experiments in which one can coherently pump a specific phonon mode may provide fundamental new insights into this piece of the puzzle.

Methods

Time-resolved ARPES. In our trARPES experiments^{18,20,29}, an infrared pump laser pulse ($h\nu = 1.48$ eV) drives the sample into a non-equilibrium state. Subsequently, an ultraviolet probe laser pulse (5.93 eV) photoemits electrons that are captured by a hemispherical analyser in an ARPES setup. The system is equipped with a Phoibos 150-mm hemispherical electron energy analyser (SPECS). The spot size (full-width at half maximum) of pump and probe beams are ~ 100 and ~ 50 μm , respectively, and the pump fluence is calculated using the same method as previously reported²⁰. The repetition rate of the pump and probe beams used in the measurements is 543 kHz. Time resolution is achieved by varying the delay time (t) between the probe and pump pulses. For $t < 0$, the probe pulse arrives before the pump pulse, and hence corresponds to an equilibrium measurement. For $t > 0$, the probe pulse arrives after the pump pulse, and hence corresponds to a non-equilibrium measurement. The zero delay and time resolution are determined by a cross-correlation of pump and probe pulses measured on hot electrons of polycrystalline gold integrated in a 0.4-eV kinetic energy window centred at 1.0 eV above the Fermi energy. The total energy resolution is ~ 22 meV, time resolution is ~ 310 fs (Supplementary Fig. 6) and the momentum resolution is ~ 0.003 \AA^{-1} at nodal point at the Fermi energy. In the electron self-energy analysis, the Lucy–Richardson iterative deconvolution method is applied to mitigate the effect of the energy resolution⁴⁸.

Samples. Nearly optimally doped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ ($T_c = 91$ K) and heavily overdoped $\text{Bi}_{1.76}\text{Pb}_{0.35}\text{Sr}_{1.89}\text{CuO}_{6+\delta}$ single crystals were grown by the travelling solvent floating zone method. The overdoped Bi2212 sample ($T_c = 60$ K) and heavily overdoped Bi2201 sample ($T_c < 2$ K) were obtained by annealing the as-grown optimally doped Bi2212 and Bi2201 in high pressure oxygen. The T_c values of the samples were characterized by superconducting quantum interference measurements. Samples were cleaved *in situ* in ultra-high vacuum with a base pressure $< 5 \times 10^{-11}$ Torr.

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Author contributions

W.Z., C.L.S. and T.L.M. carried out the measurements on Bi2212. C.H., C.L.S. and W.Z. took the measurement on Bi2201. W.Z., C.H., C.L.S. and T.L.M. were responsible for data analysis. Bi2212 samples were prepared by H.E. Bi2201 sample was prepared by K.K., T.A. and Y.K. A.L. was responsible for the experimental planning and infrastructure. All authors contributed to the interpretation and writing of the manuscript.

Additional information

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