Photoinduced Nonequilibrium Response in Underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ Probed by Time-Resolved Terahertz Spectroscopy

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Intense laser pulses have recently emerged as a tool to tune between different orders in complex quantum materials. Among different light-induced phenomena, transient superconductivity far above the equilibrium transition temperature in cuprates is particularly attractive. Key to those experiments was the resonant pumping of specific phonon modes, which was believed to induce superconducting phase coherence by suppressing the competing orders or modifying the structure slightly. Here, we present a comprehensive study of photoinduced nonequilibrium response in underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$. We find that upon photoexcitations, the Josephson plasma edge in the superconducting state is initially removed accompanied by quasiparticle excitations, and subsequently reappears at a frequency lower than the static plasma edge within a short time. In the normal state, an enhancement or weaker edgelike shape is indeed induced by pump pulses in the reflectance spectrum accompanied by simultaneous rises in both real and imaginary parts of conductivity. We compare the pump-induced effects between near- and midinfrared excitations and exclude phonon pumping as a scenario for the photoinduced effects above. We further elaborate that the transient responses in the normal state are unlikely to be explained by photoinduced superconductivity.

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High- T_c superconducting cuprates are highly anisotropic materials. The conducting CuO_2 layers are separated by different block layers, leading to less-conducting or insulatorlike c -axis dc and optical responses in the normal state. The low-frequency c -axis optical spectra of high- T_c superconducting cuprates are dominated by infrared active phonons with a very small contribution from free carriers. However, when the cuprate system goes into the superconducting state, a very sharp plasma edge suddenly develops in the c -axis reflectivity spectrum. The plasma edge is caused by the condensed superfluid carriers via the Josephson tunneling effect, which is referred to as the Josephson plasmon edge (JPE) [1–3]. The manifestation of such a c -axis JPE is taken as an optical evidence for the occurrence of superconductivity.

Ultrashort pulses now provide unique opportunities to manipulate different orders and physical properties in complex electronic materials. An unexpected finding is that intense ultrafast excitation can induce a Josephson

plasmalike edge in the reflectance spectra and $1/\omega$ -like dependence in the imaginary part of conductivity along the c axis of cuprates in the normal state, which was taken as an indication of light-induced transient superconductivity. The effect was first observed notably 5 ps after excitation in a stripe-ordered cuprate $\text{La}_{1.675}\text{Eu}_{0.2}\text{Sr}_{0.125}\text{CuO}_4$ at 10 K whose T_c is less than 2 K [4], then in underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ (YBCO) at a temperature even above room temperature [5,6]. In those measurements, the pump excitation wavelength is tuned to about $15\text{ }\mu\text{m}$ ($\sim 20\text{ THz}$) in the midinfrared (MIR) region which is assumed to be resonant with specific phonon modes of oxygen (e.g., an in-plane Cu-O stretching mode or apical oxygen mode relative to CuO_2 planes). The resonant pumping of the specific phonon mode is believed to be the key to light-induced superconductivity. The observations also motivated theoretical studies on transient superconductivity along the direction of the resonant phonon pumping [7–13].

A near-infrared (NIR) pump is also used to investigate the photoexcitation-induced effect in cuprates. Nicoletti *et al.* reported a surprisingly large effect on $\text{La}_{1.885}\text{Ba}_{0.115}\text{CuO}_4$: a blueshift of JPE with an even enhanced edge amplitude when $T < T_c$, and a light-induced edge when $T > T_c$ in the stripe ordered state whose amplitude is comparable with that seen in the equilibrium superconducting state [14,15]. A significant NIR pump-induced effect was recently also observed in

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time-resolved terahertz measurements on other compositions of $\text{La}_{2-x}\text{A}_x\text{CuO}_4$ ($\text{A} = \text{Ba}, \text{Sr}$) and electron-doped $\text{Pr}_{0.88}\text{LaCe}_{0.12}\text{CuO}_4$ [16–20]. Those results are notably different from the above reports on $\text{La}_{1.675}\text{Eu}_{0.2}\text{Sr}_{0.125}\text{CuO}_4$ [4] and $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ [5,6], where the relative change of reflectivity is less than 20% even after taking the penetration depth mismatch into account. Nonetheless, up to now there is no report about c -axis terahertz measurement on YBCO driven by NIR pulse excitations.

Aiming at addressing two critical issues on the photo-induced effect in YBCO, that is, whether phonon pumping is essential in inducing the effect, and whether the induced edgelike shape in the reflectance spectrum and $1/\omega$ -like dependence in the imaginary part of conductivity could be unambiguously attributed to transient superconductivity, here we present a comprehensive NIR and MIR pump, c -axis terahertz probe measurement on underdoped YBCO superconductors. First, we investigate, the non-equilibrium states after NIR excitations below and above T_c , which suggest a strong depletion of superconducting condensate and a significant increase of quasiparticles, respectively. Second, we show that similar transient responses are detected after tuning the pump to be resonant with the phonon near 650 cm^{-1} . We elaborate that the present study does not favor positive answers to those questions above.

Details of sample growth and spectral measurements are presented in the Supplemental Material [21]. Figure 1(a) shows the temperature-dependent magnetization data for the two crystals measured under the magnetic field of 50 Oe in a Quantum Design Physical Property Measurement System. They show a sharp superconducting transition temperature at $T_c = 55\text{ K}$ ($x = 0.55$) and 35 K ($x = 0.45$), respectively. Figure 1(b) presents the broadband reflectivity spectra $R(\omega)$ of the two samples along the c axis at three different temperatures: 5 K, just above T_c and 300 K, which are measured by Fourier transform infrared spectrometers in combination with a time-domain terahertz spectroscopy system (details are presented in the Supplemental Material [21]).

Similar to the previous results on underdoped YBCO [22,23], the out-of-plane reflectivities are dominated by phonons in the far-infrared region. Among all those phonons, the one located near 650 cm^{-1} , assigned as an A_{2u} mode involving the bonds between apical oxygens and copper atoms [24], has been drawing much attention for it may be related to the enhancement of superconductivity in YBCO [25]. Below 85 cm^{-1} , the reflectivity becomes smooth. An upward curvature suggests the presence of free-carrier contribution in the normal state. The $x = 0.55$ sample apparently has a higher reflectivity and shows a stronger upward curvature at a very low frequency. With increasing temperature, one can also observe an increase of low-energy reflectivity. On the other hand, in the superconducting state, sharp JPEs develop with reflectivity minimums locating at 40 cm^{-1} for $x = 0.55$ and 22.4 cm^{-1} for $x = 0.45$, respectively.

FIG. 1. Sample characterization and broadband reflectivity spectra along the c axis in the equilibrium state. (a) Temperature-dependent magnetization measurements. (b) Out-of-plane reflectance spectra of YBCO are governed by phonons in the far-infrared region. In superconducting states, sharp Josephson plasmon edges show up in the terahertz regime. Dashed lines are the low-frequency extrapolations used for Kramers-Kronig transformation. The gray shaded areas show the energy scale and spectral range of pump pulses. (c),(d) The colored thick lines show the real and imaginary parts of conductivity in $x = 0.55$ at 60 and 300 K. The black and gray thin lines show the fitting results of the Drude-Lorentz model. (e),(f) Reflected terahertz electric field in time and frequency domain measured by a time-domain terahertz spectroscopy system, whose measurement range is plotted as a shaded area in (b)–(d).

Figures 1(c) and 1(d) show the real and imaginary parts of conductivity below 550 cm^{-1} , derived from the Kramers-Kronig transformation of broadband reflectivity, for the $x = 0.55$ sample at 60 and 300 K in the normal state, respectively. An increase of low-frequency spectral weight in $\sigma_1(\omega)$ from 60 to 300 K is clearly observed, suggesting an increased contribution from free carriers. The result is consistent with the change of reflectivity mentioned above. Concurrently, $\sigma_2(\omega)$ in the low frequency shows an enhancement, as displayed in the inset of Fig. 1(d). The thin black and gray lines are fitting curves which we shall explain in the discussion section.

Figure 1(e) shows the waveforms from time-domain terahertz measurements $E_0(t)$ on the $x = 0.55$ sample at two different temperatures. Figure 1(f) shows the amplitude of the Fourier transformation of the terahertz electric field, i.e., $|E_0(\omega)|$. The oscillations observed in the time domain at 5 K lead to a strong dip feature at 40 cm^{-1} in the frequency domain, which is in good agreement with the JPE dip observed in Fourier transform infrared reflectance measurements.

In our pump-probe experiments, two selective pump wavelengths, as indicated by gray shaded areas in Fig. 1(b), are used to interrogate the pump-induced change in the terahertz regime. MIR pump pulses are tuned to $15\ \mu\text{m}$ ($667\ \text{cm}^{-1}$), being resonant with the phonon near $650\ \text{cm}^{-1}$, and NIR pump pulses are set to $1.28\ \mu\text{m}$ ($7810\ \text{cm}^{-1}$), whose energy scale is notably higher than all the phonons and collective modes in YBCO.

We now present an NIR pump-induced change in the terahertz regime in the superconducting state first. Because the two samples exhibit qualitatively the same behaviors, we show only the data collected on the $x = 0.55$ sample in the main text, while the whole set of data on the $x = 0.45$ sample are available in the Supplemental Material [21]. Figure 2(a) displays the decay procedure of the maximum absolute value of pump-induced change, $|\Delta E_{\text{max}}|/E_{\text{peak}}$, after excitation at $1.28\ \mu\text{m}$ by a fluence of $1\ \text{mJ}/\text{cm}^2$ (peak electric field of $\sim 3.9\ \text{MV}/\text{cm}$). We define the time zero at the position where $|\Delta E_{\text{max}}|/E_{\text{peak}}$ starts to change. Roughly a 7% maximum relative change is seen at 3 ps after excitation. The signal decays to $\sim 37\%$ of the maximum signal within 40 ps before reaching a metastable state [16,19]. Then, the pump-induced relative change $\Delta E(t, \tau)/E_{\text{peak}}$ at time delay τ in the superconducting state could be measured by fixing the terahertz gate line at the fixed delay time τ . The methods for determining the phase of $\Delta E(t, \tau)$ relative to static $E(t)$ are presented in the Supplemental Material [21]. Figure 2(b) shows $\Delta E(t, \tau)/E_{\text{peak}}$ at two representative delay times $\tau = 3\ \text{ps}$ and $40\ \text{ps}$.

The amplitudes of Fourier transformation of $\Delta E(t, \tau)$, i.e., $|\Delta E(\omega, \tau)|$, are displayed in Fig. 2(c). $|\Delta E(\omega, \tau)|$ behaves quite differently compared with the static reflected electric field of terahertz $|E_0(\omega)|$ (plotted in gray lines). The oscillations of the pump-induced terahertz signal $\Delta E(t, \tau)/E_{\text{peak}}$ in the time domain with an oscillation period of $\sim 0.85\ \text{ps}$ give a pronounced peak near $40\ \text{cm}^{-1}$ in the frequency domain $|\Delta E(\omega, \tau)|$, suggesting that the pump-induced change occurs predominantly near the static JPE position.

A multilayer model is used to obtain the authentic pump-induced change of optical properties, which is disguised by the non-negligible mismatch of the penetration depth of pump and probe pulses. The detailed calculation method and rationality of the model are presented in the Supplemental Material [21]. All the transient optical constants shown below are calculated with the multilayer model. It deserves to be remarked that, although the static electric field $|E_0(\omega)|$ has sufficiently high signal down to less than $10\ \text{cm}^{-1}$, the pump-induced signal of $|\Delta E(\omega, \tau)|$ in the superconducting state changes predominantly near JPE and has vanishing spectral weight only below $18\ \text{cm}^{-1}$ depending on time delays [see Fig. 2(c)], which makes the calculated optical constants dubious below $18\ \text{cm}^{-1}$. We distinguish those data from others with color-fading dots in Fig. 2.

Figure 2(d) shows the transient reflectivity at selective time delays. The NIR excitations dramatically change the low-frequency c -axis response. Even before reaching

FIG. 2. Pump-induced changes after excitation of $1.28\ \mu\text{m}$ at $5\ \text{K}$. (a) The decay procedure of $|\Delta E_{\text{max}}|/E_{\text{peak}}$ after being excited by a fluence of $1\ \text{mJ}/\text{cm}^2$ (peak electric field of $\sim 3.9\ \text{MV}/\text{cm}$). E_{peak} is the maximum value of $E_0(t)$ at a static position. (b) The pump-induced relative change $\Delta E(t, \tau)/E_{\text{peak}}$ in the time domain at 3 and 40 ps. (c) Fourier transformed spectrum of $\Delta E(t, \tau)$. Gray lines represent the Fourier transformation of the static reflected electric field divided by a coefficient. (d) Transient reflectivity $R(\omega, \tau)$. (e) Real part of conductivity $\sigma_1(\omega, \tau)$. (f) Imaginary part of conductivity $\sigma_2(\omega, \tau)$. Some portions are plotted in color-fading dots; for the light-induced measurement signal there has almost vanishing spectral weight, which makes those calculated data points unreliable. Optical constants in the static state are plotted in gray lines.

the maximum pump-probe signal, e.g., at the time delay $\tau = 1.1$ and 1.5 ps, reflectivity near JPE is strongly modified: the reflectivity values below static JPE are suppressed and those above JPE enhanced. The observations indicate a breakdown of Josephson tunneling along the c axis and a development of quasiparticle excitations. At the maximum position of the pump-induced signal, i.e., at 3 ps, a small edge reappears at the lower energy scale. Then, the edge gets sharper at subsequent time delays and shifts towards static JPE at a higher energy scale. This procedure reflects the recovery of Josephson tunneling after excitation.

The depression and recovery procedure of the Josephson plasmon mode and quasiparticle excitations can be seen more clearly in real and imaginary parts of conductivity, i.e., $\sigma_1(\omega)$ and $\sigma_2(\omega)$, respectively. In the static state, the values of $\sigma_1(\omega)$ along the c axis are rather low, which is shown as the gray lines in Fig. 2(e). Upon NIR excitations, the low-frequency spectral weight develops in $\sigma_1(\omega)$, which reflects pump-induced quasiparticle excitations. The quasiparticle spectral weight increases sharply upon initial pumping, reaches a maximum at the time delay close

to the maximum signal, and then decreases at subsequent time delays. Meanwhile, $\sigma_2(\omega)$ deviates quickly from that in the static state which roughly follows $1/\omega$ dependence arising from the superconducting condensate, as shown in Fig. 2(f). The deviation becomes most prominent at the time delay near the maximum pumping signal, then gets smaller at further time delays. Those results strongly suggest that the superconducting condensate is heavily disturbed or destroyed upon NIR pumping and gradually recovered with time delays.

Figure 3 summarizes the NIR pump-induced change after the excitation at $1.28 \mu\text{m}$ by a fluence of $2 \text{ mJ}/\text{cm}^2$ (peak electric field of $\sim 5.5 \text{ MV}/\text{cm}$) at the temperature of 60 K. Three representative time delays shown in Fig. 3(a) will be discussed. The middle panel of Fig. 3(b) shows the measured pump-induced change $\Delta E(t, \tau)$ at 1.5 ps, which represents the maximum photoinduced response in the normal state. The upper and lower panels show $\Delta E(t, \tau)$ at 0.65 and 3 ps, which locate at half of the rise and decay procedures, respectively. According to the raw experimental data in Figs. 3(a) and 3(b), there are essentially three differences in the normal state compared

FIG. 3. Pump-induced changes after excitation at $1.28 \mu\text{m}$ by a fluence of $2 \text{ mJ}/\text{cm}^2$ (peak electric field of $\sim 5.5 \text{ MV}/\text{cm}$) in the normal state. (a) The decay procedure of $|\Delta E_{\text{max}}|/E_{\text{peak}}$ at 60 K is plotted in a solid line. Three representative time delays are labeled in colored triangles. The decay procedure profile at 5 K is plotted in a dashed line for comparison. (b) The pump-induced relative change $\Delta E(t, \tau)/E_{\text{peak}}$ in the time domain at three different time delays. (c) Fourier transformed spectrum of $\Delta E(t, \tau)$. Gray lines indicate the Fourier transformation of the static reflected electric field divided by a coefficient. (d),(f) Transient reflectivity $R(\omega, \tau)$, real part of conductivity $\sigma_1(\omega, \tau)$, imaginary part of conductivity $\sigma_2(\omega, \tau)$, respectively. The colored dots are experimental data and the black lines are fitting curves. Optical constants in the static state are plotted in gray lines.

with that in the superconducting state. First, the rise and decay times of $|\Delta E_{\max}|/E_{\text{peak}}$ are relatively short in the normal phase as shown in Fig. 3(a). Second, the pump-induced change of the reflected electric field is significantly smaller in the normal phase compared with that in the superconducting state. Third, the damped oscillations with an oscillation period of ~ 0.85 ps observed in the superconducting state at 5 K disappears completely in the pump-induced change $\Delta E(t, \tau)$, and the time duration of $\Delta E(t, \tau)$ is within 3 ps in the normal phase, as shown in Fig. 3(b). Figure 3(c) shows the amplitude of the Fourier transformation of $\Delta E(t, \tau)$, which covers nearly all the terahertz regime generated in our experiments. $\Delta E(t, \tau)$ and $E_0(t)$ in the normal state is multiplied by a Blackman window function before doing a Fourier transformation to reduce the noise effect, which will not affect the calculated transient optical constants.

Figures 3(d)–3(f) present the transient optical constants calculated with the multilayer model at the three time delays. For all three of those time delays after excitation, a slight enhancement of reflectivity can be observed. At 0.65 ps, the enhancement at lower frequency dominates more than that at higher. At the maximum pump-induced response position, 1.5 ps, transient reflectivity ascends collectively. Then, the reflectivity turns to decay, as seen at 3 ps. Enhancement of low-frequency reflectivity usually reflects an enhanced contribution from free carriers, which is also supported by the simultaneous rise of $\sigma_1(\omega)$ and $\sigma_2(\omega)$. It indicates that the NIR pump pulses turn to result in quasiparticle excitations. We shall address this issue further in the discussion part. The fitting curves depicted in Figs. 3(d)–3(f) will also be explained there.

We also perform MIR pump-terahertz probe experiments on YBCO in superconducting and normal states as shown in Fig. 4, in which the MIR pump pulses are tuned to $15 \mu\text{m}$ and resonant with the apical-oxygen-related phonon mode. The MIR pump is at a fluence of $1 \text{ mJ}/\text{cm}^2$ and the peak electric field is $\sim 1.5 \text{ MV}/\text{cm}$. In both the superconducting and normal state, the sample exhibits similar rise and decay procedures of $|\Delta E_{\max}|/E_{\text{peak}}$ after the excitation by MIR [Fig. 4(a)] and NIR [Fig. 3(a)] pump pulses, though the decay of $|\Delta E_{\max}|/E_{\text{peak}}$ after MIR excitation appears more significant. Figure 4(b) shows the waveform of a pump-induced change of the reflected terahertz electric field at a maximum response position, $\tau = 3$ ps, in the superconducting state ($T = 5 \text{ K}$). Figures 4(c)–4(e) show the calculated transient optical constants, $R(\omega, \tau)$, $\sigma_1(\omega, \tau)$, $\sigma_2(\omega, \tau)$, using the above-mentioned multilayer model. The transient responses by NIR excitations at the same fluence and same time delay are also shown in those panels as dashed lines for comparison. Similar to NIR pump pulses, in the superconducting state, MIR pulses also turn to remove JPE upon exciting and then drive YBCO into a state with JPE at a lower energy scale together with some spectral weight increase arising from excited quasiparticles. Figures 4(f)–4(i) show the

FIG. 4. Comparison between the pump-induced effects after the excitation of $15 \mu\text{m}$ (solid lines) and $1.28 \mu\text{m}$ (dashed lines) pulses at $1 \text{ mJ}/\text{cm}^2$. (a) The decay procedure of $|\Delta E_{\max}|/E_{\text{peak}}$ at 5 and 60 K. (b) The pump-induced change of the terahertz electric field at 5 K in the time domain. (c)–(e) Transient optical constants, $R(\omega, \tau)$, $\sigma_1(\omega, \tau)$, $\sigma_2(\omega, \tau)$ at 5 K. (f) The pump-induced change of the terahertz electric field at 60 K in the time domain. (g)–(i) The transient optical constants at 60 K in the normal state. Gray lines are the optical constants in the equilibrium state.

transient responses at the time delay $\tau = 1.5$ ps by MIR excitations in the normal state (60 K), together with those by NIR excitations. We find that MIR excitations also lead to an increase of reflectivity and enhancement in both real and imaginary parts of conductivity, implying quasiparticle excitations. Although the pump-induced changes after NIR excitations [dashed lines in Figs. 4(d) and 4(e)] are more significant than those after the MIR pump, there is no essential difference between those two transient responses, as seen in Fig. 4.

Below we will make a brief comparison between the present experimental results with others reported in Refs. [5,6,26]. In the superconducting state, the transient reflectivities at 1.1 and 1.5 ps are close to the reported results after being excited by $15 \mu\text{m}$ (MIR) at the fluence of $4 \text{ mJ}/\text{cm}^2$ and the peak electric field of $\sim 3 \text{ MV}/\text{cm}$ [see Fig. 5(j) in Ref. [6]]. Both of those measurements show that the transient reflectivity is a gentle slope with a dip near the static JPE in the superconducting state, which denotes a depletion of superconducting condensate and excitation of quasiparticles here. However, the observed feature was interpreted as the static JPE being fixed accompanied by

a weak new edge appearing at a higher energy scale [5,6]. In order to explain the edge appearing at a higher energy, they assumed nonhomogeneous excitations in the pumped regime even after considering the penetration depth mismatch. They claimed the superconductivity in a great portion (80%) of the pumped volume stays unaffected with that in a small portion (20%) being enhanced (see Fig. FS3.1 in the Supplemental Material of Ref. [6]). That explanation cannot be supported by our experiments. In fact, the spectral shape of transient reflectivity at any time delay presented here could not be reproduced by a combination of two different Josephson plasma edges with different volume fractions. Furthermore, the transient reflectivity at a subsequent time depicts a re-emergence and recovery procedure of the static JPE, which indicates that suppression of the static JPE upon excitations is a more plausible explanation. Our result reveals that the Bruggeman's effective medium model is not applicable in the superconducting state. A recent report posted by Liu *et al.* [26] confirms the spectral shape we observed in the superconducting state when they use similar MIR pump fluence at 1.5 mJ/cm² (peak electric field at ~ 1.4 MV/cm), indicating a depletion of the superconducting condensate. However, when they increase the MIR pump to a higher fluence of 8 mJ/cm² and the peak electric field of ~ 3 MV/cm (see Fig. S8 in the Supplemental Material of Ref. [26]), they observe a transient increase in superfluid density in the superconducting state. We remark that we could not make a comparison on the MIR regime due to the limitation on the MIR pump fluence in our experiments. Nevertheless, we performed the measurement with NIR (1.28 μ m) excitation at a higher fluence of 6 mJ/cm² (peak electric field of ~ 9.5 MV/cm) in both the superconducting and the normal state, in which the peak electric field is much higher than other reports on YBCO. Rather than an enhancement or emergence of superconductivity, the results still indicate superconductivity depletion and quasiparticle excitations after a strong NIR pump. Details can be found in the Supplemental Material [21].

The present work enables us to address two crucial issues with respect to the photoinduced transient superconductivity. The first one is whether or not the phonon resonant pump is essential for the photoinduced effect. In the last few years, mode-selective optical control became particularly attractive because it is argued based on the MIR pump that the coherent excitation of a certain phonon mode can suppress the charge order and simultaneously enhance superconductivity in cuprates. It has motivated many theoretical studies in this direction [7–13]. Since we find qualitatively the same effect by the NIR and MIR pump, the phonon resonant pump could be essentially ruled out. Following our work, the recent results by Liu *et al.* also indicate that NIR pump pulses behave in a similar fashion and have a significant photoexcitation effect on YBCO [26]. Moreover, we shall emphasize that the calculated

transient optical constants using the multilayer model are quite sensitive to the estimated penetration depth of pump pulses. The pump-induced change turns to get more significant when the penetration depth of pump pulses get more inconspicuous compared with that of the probe one (find in Supplemental Material [21]). At the phonon mode position, the penetration depth gets to a minimum and changes quickly when the tuning frequency deviates from the phonon, which could lead to maximum photoinduced effects even when raw experimental data $\Delta E(t, \tau)/E_{\text{peak}}$ are nearly the same.

The second important issue is whether or not the observed phenomenon in the normal state could be attributed to photoinduced transient superconductivity. We shall address this issue from the pump-induced change in both transient $R(\omega)$ and transient $\sigma_2(\omega)$. Our measurement indeed confirms the presence of photoinduced enhancement in transient $R(\omega, \tau)$ [or an edgelike upturn in $\Delta R/R$ (see Supplemental Material [21] for data on the $x = 0.45$ sample)] by both NIR and MIR excitations. However, the enhancement or edgelike shape is rather weak in the calculated reflectance [Fig. 3(d)]. The results are similar to the data reported previously [5,6,27]. However, the interpretation of the transient enhancement as the emergence of a new Josephson plasma edge relies on the analysis based on nonhomogeneous excitations, even after taking the penetration depth mismatch of the pump and probe pulses into account. A quantitative fit yields roughly 20% of the volume fraction in the pumped regime being transformed into the transient superconducting component and contributes to the edgelike upturn [6]. We elaborated above that our measurement results are not compatible with the nonhomogeneous excitations in the superconducting state, then there is no reason that the nonhomogeneous excitations could be present in the normal state. On this basis, we cannot attribute the observed enhancement or edgelike shape to the transient superconductivity which gives rise to a JPE in the normal state.

We now discuss the transient enhancement in the imaginary part of conductivity. The superconductivity-related response manifests naturally in the complex conductivity. At zero temperature, the real part of conductivity of a superconductor is condensed into a delta function at zero frequency, and by Kramers-Kronig transformation, the imaginary part of conductivity goes as $1/\omega$ dependence:

$$\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega) = \frac{\omega_{ps}^2}{8}\delta(0) + i\frac{\omega_{ps}^2}{4\pi\omega}, \quad (1)$$

where ω_{ps}^2 is the condensed plasma frequency and related to the London penetration depth by $\omega_{ps}^2 = 1/\lambda_L^2$. According to this equation, the $1/\omega$ dependence in $\sigma_2(\omega)$ could be taken as an indication of superconductivity, and the $4\pi\omega\sigma_2(\omega)$ at the zero frequency limit could be used to estimate the superconducting condensate or London

penetration depth. Recently, $1/\omega$ dependence observed in transient $\sigma_2(\omega)$ was widely taken to be the other evidence for the photoinduced transient superconductivity in the literature [5,6,14,18,26–28]. We would like to remark that the observation of $1/\omega$ dependence in $\sigma_2(\omega)$ within a very limited range of frequency is not a sufficient condition for the identification of superconductivity. When a compound changes from the normal state to a superconducting state, the low-frequency spectral weight in $\sigma_1(\omega)$ must be reduced due to the condensate to the delta function at zero frequency. The missing spectral weight in $\sigma_1(\omega)$ can also be used to derive the superconducting condensate:

$$\omega_{ps}^2 = 8 \int_0^\infty [\sigma_1(\omega, T > T_c) - \sigma_1(\omega, T \ll T_c)] d\omega. \quad (2)$$

Indeed, a loss of low-frequency spectral weight in $\sigma_1(\omega)$ has always been observed in cuprate superconductors across the superconducting transitions in the equilibrium state, including measurements polarized along the c axis [29]. However, this was not the case for the above-mentioned works on laser-induced response in the normal state [5,6,14,18,26–28]. In those works, when a $1/\omega$ dependence or an upward increase with decreasing frequency was identified in $\sigma_2(\omega)$, the low-frequency $\sigma_1(\omega)$ does not drop compared with the values in the static state. Instead, the low-frequency spectral weight of $\sigma_1(\omega)$ still shows an increase. The simultaneous enhancement in both transient $\sigma_2(\omega)$ and $\sigma_1(\omega)$ is in contradiction to the superconducting condensate.

As a matter of fact, even for a simple Drude response, the imaginary part of conductivity $\sigma_2(\omega)$ could show approximately a $1/\omega$ dependence if $\omega \gg 1/\tau$ ($1/\tau$ can be considered as a scattering rate). $\sigma_2(\omega)$ starts to decrease only when ω becomes smaller than $1/\tau$. When the scattering rate is very small, it becomes hard to distinguish between a Drude response and a superconducting response solely from the frequency dependence of $\sigma_2(\omega)$ in the very limited terahertz measurement frequency range. As shown in Fig. 1(b), the reflectivities of the two samples in the normal state are not completely flat, but show a clear increase feature at the low-frequency limit, implying the presence of a free-carrier contribution to the c -axis optical response. The free-carrier contribution increases with increasing doping levels as reflected in the two samples. Actually, in an earlier study on the c -axis charge dynamics on $\text{La}_{1-x}\text{Sr}_x\text{CuO}_4$, the c -axis reflectance shows clearly the upturn feature in the terahertz regime upon increasing doping and temperature [2]. Similar changes were also observed for YBCO [22,23]. It is well known that, for cuprate superconductors, the charge conduction along the c axis is rather complex, which involves oxygen bonding with virtual Cu 4s orbitals [30–32]. The c -axis hopping integral is strongly in-plane momentum dependent, being zero at the nodal direction and maximum at the antinodal

direction for the simple tetragonal cuprate system. Furthermore, the scattering rates of charge carriers contributing to the c -axis conductivity could be different at a different region of the Fermi surface or Fermi arc. The presence of Cu-O chains in YBCO further complicates the conduction. Therefore, it is not expected that the quasiparticle contribution to the c -axis conductivity could be well explained by a simple Drude response. Nonetheless, the Drude response picture can still be used to explain the trend of low-frequency optical data contributed from the quasiparticles.

To analyze the low-frequency Drude-like increase feature in $R(\omega)$ and compare all optical constants in both the equilibrium and nonequilibrium states more quantitatively, we try to fit $R(\omega)$, $\sigma_1(\omega)$, and $\sigma_2(\omega)$ simultaneously by using a Drude-Lorentz model

$$\epsilon(\omega) = \epsilon_\infty - \sum_{i=1} \frac{\omega_{pi}^2}{\omega^2 + i\omega/\tau_i} + \sum_{j=1} \frac{S_j^2 e^{i\theta_j}}{\omega_j^2 - \omega^2 - i\omega/\tau_j}. \quad (3)$$

We find that the low-frequency optical constants could be well reproduced simply by using two Drude components and a number of Lorentz components for phonon peaks. As an example, Figs. 1(c) and 1(d) show the Drude-Lorentz fit to the frequency dependence of $\sigma_1(\omega)$ and $\sigma_2(\omega)$ in the equilibrium state for the $x = 0.55$ crystal at 60 and 300 K in the far-infrared region, respectively. In the inset of Fig. 1(d), we show the enlarged part of $\sigma_2(\omega)$ at low frequency; an enhancement of $\sigma_2(\omega)$ is seen simply at an elevated temperature. We also find that the transient optical constants after excitations at 60 K in Figs. 3(d)–3(f), and even increasing the pump fluence to 6 mJ/cm² with the peak electric field being ~ 9.5 MV/cm (shown in the Supplemental Material [21]), could be approximately reproduced simultaneously by the model with enhanced plasma frequencies and slightly reduced scattering rates of Drude components. Although the fitting parameters (shown in the Supplemental Material [21]) may only reflect the trend of free-carrier evolution for the reasons mentioned above, the fitting clearly illustrates that the upward increase or a $1/\omega$ -like behavior in $\sigma_2(\omega)$ in the very narrow measured frequency range could not be uniquely explained by the transient superconductivity. Quasiparticle excitations offer an alternative way to explain the observed phenomenon, which could be similar to the results of some single-color pump-probe experiments [33–35].

We emphasize that the optical constants after NIR or MIR excitations—i.e., the simultaneous enhancements in $R(\omega, \tau)$, $\sigma_1(\omega, \tau)$, and $\sigma_2(\omega, \tau)$ —evolve in a way very similar to the increase of the doping level or even increase of temperature. It is thus reasonable to attribute the effect to the contribution of photoexcited quasiparticles. We would like to point out that, in early studies, the quasiparticle scattering rate along the c axis was believed to be very large, for the conductivity is very small. Since we observe a

clear Drude-like increase of upturn of $R(\omega)$ at a very low frequency in the equilibrium state, it turns out that the normally assumed very large scattering rate may not be true. This issue should be further studied.

In summary, we observe a strong pump-induced spectral change in terahertz frequency, predominantly near the energy scale of the Josephson plasmon edge below T_c . The edge is almost removed upon initial photoexcitations, indicating a collapse of superconducting condensate. After a short time delay we observe the reappearance of a JPE at a frequency lower than the static JPE, whose feature becomes more pronounced and shifts slightly to a higher energy scale with delay time evolving. Meanwhile, quasiparticle excitations develop and contribute to the spectral weight in the real part of conductivity. Above T_c , a much smaller pump-induced effect is detected. An enhancement or weak edgelike shape develops in the reflectance spectrum, which also results in an increase of both real and imaginary parts of conductivity. In addition, we find very different time-scales for achieving the maximum pump-probe signal between $T < T_c$ and $T > T_c$. We elaborate that the pump-induced effect above T_c is unlikely to be explained by photoinduced transient superconductivity. In addition, there is no substantial difference between the near- and midinfrared pump cases both in the superconducting and normal state, which indicates that a phonon resonant pump as a scenario for the photoexcitation effect can be excluded.

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[1] K. Tamasaku, Y. Nakamura, and S. Uchida, *Charge Dynamics across the CuO₂ Planes in La_{2-x}Sr_xCuO₄*, *Phys. Rev. Lett.* **69**, 1455 (1992).
 [2] S. Uchida, K. Tamasaku, and S. Tajima, *c-Axis Optical Spectra and Charge Dynamics in La_{2-x}Sr_xCuO₄*, *Phys. Rev. B* **53**, 14558 (1996).
 [3] C. C. Homes, T. Timusk, R. Liang, D. A. Bonn, and W. N. Hardy, *Optical Conductivity of c Axis Oriented YBa₂Cu₃O_{6.7}: Evidence for a Pseudogap*, *Phys. Rev. Lett.* **71**, 1645 (1993).
 [4] D. Fausti, R. I. Tobey, N. Dean, S. Kaiser, A. Dienst, M. C. Hoffmann, S. Pyon, T. Takayama, H. Takagi, and A. Cavalleri, *Light-Induced Superconductivity in a Stripe-Ordered Cuprate*, *Science* **331**, 189 (2011).
 [5] S. Kaiser, C. R. Hunt, D. Nicoletti, W. Hu, I. Gierz, H. Y. Liu, M. Le Tacon, T. Loew, D. Haug, B. Keimer, and A. Cavalleri, *Optically Induced Coherent Transport far above T_c in Underdoped YBa₂Cu₃O_{6+δ}*, *Phys. Rev. B* **89**, 184516 (2014).
 [6] W. Hu, S. Kaiser, D. Nicoletti, C. R. Hunt, I. Gierz, M. C. Hoffmann, M. Le Tacon, T. Loew, B. Keimer, and A. Cavalleri, *Optically Enhanced Coherent Transport in*

YBa₂Cu₃O_{6.5} by Ultrafast Redistribution of Interlayer Coupling, *Nat. Mater.* **13**, 705 (2014).
 [7] A. Subedi, A. Cavalleri, and A. Georges, *Theory of Non-linear Phononics for Coherent Light Control of Solids*, *Phys. Rev. B* **89**, 220301(R) (2014).
 [8] S. J. Denny, S. R. Clark, Y. Laplace, A. Cavalleri, and D. Jaksch, *Proposed Parametric Cooling of Bilayer Cuprate Superconductors by Terahertz Excitation*, *Phys. Rev. Lett.* **114**, 137001 (2015).
 [9] Z. M. Raines, V. Stanev, and V. M. Galitski, *Enhancement of Superconductivity via Periodic Modulation in a Three-Dimensional Model of Cuprates*, *Phys. Rev. B* **91**, 184506 (2015).
 [10] M. A. Sentef, A. Tokuno, A. Georges, and C. Kollath, *Theory of Laser-Controlled Competing Superconducting and Charge Orders*, *Phys. Rev. Lett.* **118**, 087002 (2017).
 [11] J. I. Okamoto, W. Hu, A. Cavalleri, and L. Mathey, *Transiently Enhanced Interlayer Tunneling in Optically Driven High-T_c Superconductors*, *Phys. Rev. B* **96**, 144505 (2017).
 [12] N. Bittner, T. Tohyama, S. Kaiser, and D. Manske, *Possible Light-Induced Superconductivity in a Strongly Correlated Electron System*, *J. Phys. Soc. Jpn.* **88**, 044704 (2019).
 [13] A. Klein, M. H. Christensen, and R. M. Fernandes, *Laser-Induced Coherent Control of an Electronic Nematic Quantum Phase Transition*, *arXiv:1809.05600v1*.
 [14] D. Nicoletti, E. Casandruc, Y. Laplace, V. Khanna, C. R. Hunt, S. Kaiser, S. S. Dhesi, G. D. Gu, J. P. Hill, and A. Cavalleri, *Optically Induced Superconductivity in Striped La_{2-x}Ba_xCuO₄ by Polarization-Selective Excitation in the Near Infrared*, *Phys. Rev. B* **90**, 100503(R) (2014).
 [15] E. Casandruc, D. Nicoletti, S. Rajasekaran, Y. Laplace, V. Khanna, G. D. Gu, J. P. Hill, and A. Cavalleri, *Wavelength-Dependent Optical Enhancement of Superconducting Interlayer Coupling in La_{1.885}Ba_{0.115}CuO₄*, *Phys. Rev. B* **91**, 174502 (2015).
 [16] S. J. Zhang, Z. X. Wang, L. Y. Shi, T. Lin, M. Y. Zhang, G. D. Gu, T. Dong, and N. L. Wang, *Light-Induced New Collective Modes in the Superconductor La_{1.905}Ba_{0.095}CuO₄*, *Phys. Rev. B* **98**, 020506(R) (2018).
 [17] S. J. Zhang, Z. X. Wang, D. Wu, Q. M. Liu, L. Y. Shi, T. Lin, S. L. Li, P. C. Dai, T. Dong, and N. L. Wang, *Photoinduced Metastable State with Modulated Josephson Coupling Strengths in Pr_{0.88}LaCe_{0.12}CuO₄*, *Phys. Rev. B* **98**, 224507 (2018).
 [18] D. Nicoletti, D. Fu, O. Mehio, S. Moore, A. S. Disa, G. D. Gu, and A. Cavalleri, *Magnetic-Field Tuning of Light-Induced Superconductivity in Striped La_{2-x}Ba_xCuO₄*, *Phys. Rev. Lett.* **121**, 267003 (2018).
 [19] K. A. Cremin, J. Zhang, C. C. Homes, G. D. Gu, Z. Sun, M. M. Fogler, A. J. Millis, D. N. Basov, and R. D. Averitt, *Photoenhanced Metastable c-Axis Electrodynamics in Stripe-Ordered Cuprate La_{1.885}Ba_{0.115}CuO₄*, *Proc. Nat'l. Acad. Sci. U.S.A.* **116**, 19875 (2019).
 [20] H. Niwa, N. Yoshikawa, K. Tomari, R. Matsunaga, D. Song, H. Eisaki, and R. Shimano, *Light-Induced Nonequilibrium Response of the Superconducting Cuprate La_{2-x}Sr_xCuO₄*, *Phys. Rev. B* **100**, 104507 (2019).

- [21] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevX.10.011056> for the detailed information of the experiments, calculations, and fitting procedure. Experimental data with higher pump fluence and with a lower doping level ($x = 0.45$ sample) are also available.
- [22] C. C. Homes, T. Timusk, D. A. Bonn, R. Liang, and W. N. Hardy, *Optical Properties along the c -axis of $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$, for $x = 0.50 \rightarrow 0.95$ Evolution of the Pseudogap*, *Can. J. Phys.* **73**, 663 (1995).
- [23] S. Tajima, J. Schützmann, S. Miyamoto, I. Terasaki, Y. Sato, and R. Hauff, *Optical Study of c -Axis Charge Dynamics in $\text{YBa}_2\text{Cu}_3\text{O}_y$: Carrier Self-Confinement in the Normal and the Superconducting States*, *Phys. Rev. B* **55**, 6051 (1997).
- [24] C. C. Homes, T. Timusk, D. A. Bonn, R. Liang, and W. N. Hardy, *Optical Phonons Polarized along the c Axis of $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$, for $x = 0.50.95$* , *Can. J. Phys.* **73**, 663 (1995).
- [25] R. Mankowsky, A. Subedi, M. Först, S. O. Mariager, M. Chollet, H. T. Lemke, J. S. Robinson, J. M. Glowina, M. P. Minitti, A. Frano, M. Fechner, N. A. Spaldin, T. Loew, B. Keimer, A. Georges, and A. Cavalleri, *Nonlinear Lattice Dynamics as a Basis for Enhanced Superconductivity in $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$* , *Nature (London)* **516**, 71 (2014).
- [26] B. Liu, M. Forst, M. Fechner, D. Nicoletti, J. Porras, B. Keimer, and A. Cavalleri, *Two Pump Frequency Resonances for Light-Induced Superconductivity in $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$* , [arXiv:1905.08356](https://arxiv.org/abs/1905.08356).
- [27] C. R. Hunt, D. Nicoletti, S. Kaiser, D. Präpper, T. Loew, J. Porras, B. Keimer, and A. Cavalleri, *Dynamical Decoherence of the Light Induced Interlayer Coupling in $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$* , *Phys. Rev. B* **94**, 224303 (2016).
- [28] C. R. Hunt, D. Nicoletti, S. Kaiser, T. Takayama, H. Takagi, and A. Cavalleri, *Two Distinct Kinetic Regimes for the Relaxation of Light-Induced Superconductivity in $\text{La}_{1.675}\text{Eu}_{0.2}\text{Sr}_{0.125}\text{CuO}_4$* , *Phys. Rev. B* **91**, 020505(R) (2015).
- [29] D. N. Basov, S. I. Woods, A. S. Katz, E. J. Singley, R. C. Dynes, M. Xu, D. G. Hinks, C. C. Homes, and M. Strongin, *Sum Rules and Interlayer Conductivity of High- T_c Cuprates*, *Science* **283**, 49 (1999).
- [30] L. B. Ioffe and A. J. Millis, *Zone-Diagonal-Dominated Transport in High- T_c Cuprates*, *Phys. Rev. B* **58**, 11631 (1998).
- [31] T. Xiang and J. M. Wheatley, *c Axis Superfluid Response of Copper Oxide Superconductors*, *Phys. Rev. Lett.* **77**, 4632 (1996).
- [32] N. E. Hussey, M. Abdel-Jawad, A. Carrington, A. P. Mackenzie, and L. Balicas, *A Coherent Three-Dimensional Fermi Surface in a High-Transition-Temperature Superconductor*, *Nature (London)* **425**, 814 (2003).
- [33] J. Demsar, B. Podobnik, V. V. Kabanov, T. Wolf, and D. Mihailovic, *Superconducting Gap Δ_c , the Pseudogap Δ_p , and Pair Fluctuations above T_c in Overdoped $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ from Femtosecond Time-Domain Spectroscopy*, *Phys. Rev. Lett.* **82**, 4918 (1999).
- [34] P. Kusar, V. V. Kabanov, J. Demsar, T. Mertelj, S. Sugai, and D. Mihailovic, *Controlled Vaporization of the Superconducting Condensate in Cuprate Superconductors by Femtosecond Photoexcitation*, *Phys. Rev. Lett.* **101**, 227001 (2008).
- [35] L. Stojchevska, P. Kusar, T. Mertelj, V. V. Kabanov, Y. Toda, X. Yao, and D. Mihailovic, *Mechanisms of Nonthermal Destruction of the Superconducting State and Melting of the Charge-Density-Wave State by Femtosecond Laser Pulses*, *Phys. Rev. B* **84**, 180507(R) (2011).