Absence of the superconducting collective excitations in the photoexcited nonequilibrium state of underdoped YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{y}.

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Abstract

Recent observation of the light-induced superconducting (SC)-like transient response in the c-axis optical conductivity far above the SC transition temperature $T_c$ in underdoped YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{y} (YBCO) has attracted great attention in the field of high-$T_c$ superconductors. Since then, various theoretical and experimental studies have been devoted to elucidating its microscopic origin. One prominent fingerprint of the light-induced superconductivity is the emergence of $1/\omega$-like spectral behavior in the imaginary part of the optical conductivity in the terahertz (THz) frequency range. However, the spectral profile can also be described by the Drude response of the quasiparticles (QPs) with a substantially low scattering rate. To circumvent this critical ambiguity, we investigated the photoinduced nonequilibrium state in underdoped YBCO samples using the nonlinear THz optical responses originating from the SC collective excitations: the THz Kerr effect that reflects the Higgs mode and SC QPs responses, and the ac-driven Josephson current. Upon the near-infrared (NIR) photoexcitation above $T_c$, the $1/\omega$-like spectral behavior in the imaginary part of the optical conductivity emerges, whereas the THz nonlinear responses associated with those collective modes were not observed. These results indicate that the NIR-pump induced state exhibiting the $1/\omega$-like response above $T_c$ is distinct from the long-range ordered SC state in equilibrium. According to these observations, the possible origin of the irregularly coherent charge carrier response along the $c$-axis is discussed.
I. INTRODUCTION

Recent advancements in ultrafast light sources and spectroscopic techniques have accelerated the study of nonequilibrium dynamics in versatile quantum materials. One of the most intriguing examples is the photoinduced superconductivity in high-temperature cuprate superconductors. Upon photoexcitation, a plasma edge-like structure was observed in transient c-axis terahertz (THz) reflectivity spectra in stripe-ordered La-based cuprate superconductors La$_{1.673}$Eu$_{0.37}$Sr$_{0.125}$CuO$_4$ [1] and La$_{2-x}$Ba$_x$CuO$_4$ [2] and interpreted as the Josephson plasma edge of the photoinduced superconducting (SC) state. Remarkably, the photoinduced SC-like behavior was also identified in underdoped YBa$_2$Cu$_3$O$_y$ (YBCO) samples that do not exhibit the stripe order, even at room temperature depending on the doping [3,4]. Since then, various theoretical [5-18] and experimental studies [19-34] have been devoted to elucidating the microscopic origin of the observed photoinduced SC-like behavior. In these experiments, the transient emergence of the 1/ω-like spectral profile in the imaginary part of the optical conductivity $\sigma_2(\omega)$, which is identical to the response of the SC condensate in equilibrium, is interpreted as a fingerprint of the photoinduced superconductivity[1-4]. However, ambiguity remains that one cannot distinguish the 1/ω-like response in $\sigma_2(\omega)$ from a Drude response of the quasiparticles (QPs) with a substantially low scattering rate in the limited spectral window of the THz spectroscopy [33]. Accordingly, another ultrafast probe of the SC order parameter is desired to have a deeper insight into the observed SC-like behavior.

For this purpose, we shed light on the THz nonlinear optical responses originating from the collective excitations of the SC order parameter: (i) a collective amplitude mode of the order parameter, i.e., the Higgs mode, and (ii) the ac-driven Josephson current. While the Higgs mode has been elusive over decades because it lacks the linear coupling with the electromagnetic field, recent developments of intense THz light sources [35,36] have made it possible to observe the Higgs mode in conventional superconductors through the nonlinear light-Higgs coupling [37-41]. More recently, the observation of the Higgs mode has been extended to MgB$_2$ [42], cuprates [43-45], and iron-based superconductors [46-49]. In the case of cuprate superconductors, it has also been revealed that an intense THz pulse polarized along the c-axis induces ac-Josephson current and gives rise to the THz nonlinear responses [50-54]. In this study, we employed the THz nonlinear optical responses of those collective modes to probe the SC order parameter in the photoexcited nonequilibrium state of underdoped YBCO on the ultrafast time scale.

II. EXPERIMENTS

First, we performed optical pump and THz probe (OPTP) measurements using an underdoped YBCO single crystal ($T_c = 61$ K, UD61) grown by the pulling method [55]. The SC transition
temperature was determined by the magnetic susceptibility measurement using a SC quantum interference device (SQUID) as described in Appendix A. The crystal was cut and polished to give an ac surface whose size is \( a \times b \times c = 3 \times 1 \times 2 \text{ mm}^3 \). Figure 2(a) illustrates a schematic of the OPTP spectroscopy. The output of a regenerative amplified Ti:sapphire laser system with the central wavelength of 800 nm, pulse duration of 100 fs, pulse energy of 4 mJ, and repetition rate of 1 kHz, was divided into three beams: one for the generation of the THz probe pulse, one for the optical pump pulse, and the other for the gate pulse for the electro-optic (EO) sampling. The THz probe pulse was generated by the optical rectification in a ZnTe (110) crystal. The reflected THz pulse from the sample was detected by the EO sampling in a 2-mm-thick ZnTe (110) crystal. The OPTP experiment was also performed with the 1.5-\(\mu\)m pump pulse generated from an optical parametric amplifier (OPA). We present the OPTP results in the Results section A.

Next, optical pump-third-harmonic generation (THG) probe (OP-THG) measurements were performed in reflection geometry for the same UD61 sample. We generated an intense narrowband THz pulse employing the tilted-pulse-front technique with a LiNbO\(_3\) crystal [35,36]. To obtain the narrowband THz pulse with the center frequency of 0.5 THz, we inserted bandpass filters (BPFs) in the incident THz beam pass. The peak electric field (\(E\)-field) of the narrowband THz pulse was estimated as 25 kV/cm by EO sampling in the 380-\(\mu\)m-thick GaP (110) crystal placed inside the cryostat. To extract the third harmonic (TH) components of the reflected THz pulse, 1.5 THz BPFs were inserted in the reflected THz beam pass so that the fundamental harmonic (FH) component at 0.5 THz was eliminated. The reflected THz pulse from the sample was detected by EO sampling in the 2-mm-thick ZnTe (110) crystal. The OP-THG results are displayed in the Results section B.

Finally, to investigate the in-plane response of the photoinduced nonequilibrium state, we utilized the THz pump-optical probe (TPOP) spectroscopy in reflection geometry using a detwinned \(ab\)-plane YBCO single crystal (\(T_c = 78\) K, UD78), whose size was \(a \times b \times c = 2.5 \times 2 \times 1\) mm\(^3\) (see Appendix A for the determination of \(T_c\)). The intense single THz pulse was generated and detected in the same manner as the THG measurements. The THz peak \(E\)-field was estimated as 350 kV/cm at the sample position. In the Result section C, we first show the TPOP signal in YBCO that reflects the SC order parameter. Next, we discuss the photoinduced dynamics of the SC order parameter based on the optical pump-TPOP (OP-TPOP) measurements.

III. RESULTS

A. Photoexcited nonequilibrium state studied by OPTP spectroscopy

1. Equilibrium THz responses

In Figs. 1(a)-(d), we present the equilibrium optical constants of UD61 along the \(c\)-axis measured by THz time-domain spectroscopy (THz-TDS) in reflection geometry. As shown in Fig. 1(a), the
reflectivity spectrum displays the plasma edge associated with the Josephson plasma resonance (JPR) below $T_c = 61$ K. As the temperature is lowered, the Josephson plasma edge shows a blueshift and approaches 1.2 THz at 20 K, manifesting the growth of the superconductivity. Concomitantly, a peak appears at the JPR frequency in the loss function spectrum $-\text{Im}(1/\varepsilon(\omega))$ plotted in Fig. 1(b). Figures 1(c) and (d) show that below $T_c$, the spectral weight of $\sigma_1(\omega)$ is suppressed, and $\sigma_2(\omega)$ exhibits an increasing tendency toward lower frequency described by the $1/\omega$-profile, reflecting the development of the SC phase stiffness. These spectral features agree with the previous studies on the c-axis optical response of YBCO [56,57]. Since the frequency range of the measured optical constants is limited to above 0.7 THz because of the sample size, the optical constants below 0.7 THz are estimated by fitting the complex optical conductivity by the two-fluid model described in Appendix B.

2. THz transient responses in the photoexcited state below $T_c$

Figure 2(b) shows the transient THz reflectivity along the c-axis of UD61 at 20 K measured at the pump-probe delay time $t_{pp} = 3.2$ ps with the pump fluence of 3.4 mJ/cm$^2$ and the center wavelength of 800 nm. A redshift of the Josephson plasma edge is observed after the photoexcitation, indicating that the superconductivity is destroyed by the photoexcitation. The transient reflectivity change at the selected pump-probe delay times $t_{pp}$ is displayed in Fig. 2(c). For all the positive delay times, the negative reflectivity change is discerned below the equilibrium JPR frequency of 1.2 THz, which indicates the redshift of the Josephson plasma edge. It is counter-intuitive that the JPR remains, although redshifted, even after the intense photoexcitation with the fluence of 3.4 mJ/cm$^2$, which is much larger than the threshold fluence of $F_{\text{th}} = 10$-200 $\mu$J/cm$^2$ for the total destruction of the superconductivity in cuprate superconductors as reported in the time-resolved optical spectroscopy [58-62] and angle-resolved photoemission spectroscopy (ARPES) measurements [63-65]. This non-vanishing JPR even under the intense photoexcitation far above the threshold fluence $F_{\text{th}}$, is because the penetration depth of the 800-nm pump pulse $d_{\text{pump}} = 0.22 \mu$m [32] is much shorter than that of the THz probe pulse $d_{\text{THz}} = 8 \mu$m at 1 THz estimated from the refractive index at 20 K obtained by the THz-TDS measurements.

To extract the transient c-axis THz response at the photoexcited surface region of YBCO samples, various works have utilized models which suppose that the photoinduced refractive index change relaxes along the sample depth direction within the pump penetration depth [3,4,24,32,33]. However, it was pointed out that such models can give rise to artifacts when one extracts the optical constants of the photoexcited surface region, particularly in the SC state [30]. In fact, it has been demonstrated that, below $T_c$, the transient optical constants in La$_{2-x}$Sr$_x$CuO$_4$ are well described by the pump-induced sample heating caused by the injected pump energy in particular for pump-probe
delay times when the system reaches a quasi-thermal equilibrium state [30]. Since the typical electron-phonon scattering time in cuprate is ~ 6 ps [66], it is reasonable to apply this heating model to the present case of YBCO after \( t_{pp} = 6 \) ps. Accordingly, we analyze the transient optical constants at \( t_{pp} = 12.8 \) ps using this heating model, whose details are described in Appendix C. Figure 2(d) shows the spatial distribution of the pump fluence \( F(z) \) (magenta, left axis) and the sample temperature in a quasi-equilibrium \( T_f(z) \) (light blue, right axis) obtained by the heating model simulation for the incident pump fluence of 3.4 mJ/cm\(^2\). One can see that the temperature rise penetrates deeper into the sample than the pump penetration depth.

In Fig. 3, we compare the optical constants obtained by the heating model simulation to the experimental data measured at \( t_{pp} = 12.8 \) ps with the fluence of 3.4 mJ/cm\(^2\) (red) and 0.4 mJ/cm\(^2\) (blue). Here, we define the pump-induced phase shift of the reflected THz E-field as \( \theta(\omega) = \arg(r_{\text{off}}(\omega)/r_{\text{on}}(\omega)) \), where \( r_{\text{on}}(\omega) \) and \( r_{\text{off}}(\omega) \) are the complex reflectivity when the pump pulse is on and off, respectively. By using the literature values of the pump penetration depth [32] and specific heat [67], the heating model well reproduces the pivotal experimental observations without any adjustable parameters: the redshift of the JPR in the reflectivity, which is more clearly identified in the negative reflectivity change \( \Delta R/R \) around the equilibrium JPR frequency of 1.2 THz, and the decrease in the phase \( \theta(\omega) \) around the JPR frequency in equilibrium, both of which are attributed to the destruction of the superconductivity. The \( \Delta R/R \) spectrum obtained by the experiment in Fig. 3(b) is slightly different from that obtained by the simulation in Fig. 3(e) above 1.4 THz, probably due to the QP excitation effect, which is not considered in the heating model simulation. We note that the heating model simulation is not applicable at earlier pump-probe delay times where the electron-lattice system does not reach the quasi-equilibrium state. Nevertheless, in Appendix D we demonstrate that the pump-induced transient THz responses at \( t_{pp} = 3.2 \) ps are reasonably well reproduced by assuming that the superconductivity in the photoexcited surface region is partially suppressed and its optical constants are described by those at 60 K in equilibrium (destroyed layer model). This result clarifies that the superconductivity is also suppressed by the photoexcitation even for earlier delay times, which is consistent with the negative reflectivity change below 1.2 THz for all the positive \( t_{pp} \) shown in Fig.2 (c) caused by the redshift of the Josephson plasma edge.

3. THz transient responses in the photoexcited state above \( T_c \)

Now we examine the photoexcited nonequilibrium state of UD61 above \( T_c \). Figure 4(a) shows the 800-nm pump-induced THz reflectivity change \( \Delta R/R \) at 100 K measured at \( t_{pp} = 0.8 \) ps (red) and 9.6 ps (green) with the pump fluence of 3.4 mJ/cm\(^2\). The \( \Delta R/R \) spectrum at \( t_{pp} = 0.8 \) ps exhibits a plasma edge-like behavior similar to the equilibrium JPR below \( T_c \) in accordance with the previous studies [3,4,24,32]. We plot the reflectivity change computed by the heating model
simulation at 100 K in Fig. 4(a) by the blue dashed curve. Notably, the reflectivity change expected from the heating model simulation is much smaller than the experimental data, showing that the non-thermal effect plays a crucial role above $T_c$.

Figure 4(b) presents the spatial distribution of the pump fluence (magenta, left axis) and the sample temperature (light blue, right axis) obtained by the heating model simulation. The light-induced temperature rises at 100 K with the pump fluence of 3.4 mJ/cm² is limited within a length scale of the penetration depth $d_{pump}$ because the specific heat is much larger than that below $T_c$. Therefore, even though the surface layer models extracting the optical constants in the photoexcited surface region might cause artifacts below $T_c$ as mentioned above, they serve as reasonable models above $T_c$ where the refractive index exhibits a monotonic temperature dependence, as shown in Figs. 15(a) and (b) in Appendix C. Similar models have been utilized to investigate the transient THz optical constants in photoexcited semiconductors [68]. In the following, we present the optical constants at the photoexcited surface region extracted by the so-called single layer (SL) model: we suppose that the refractive index depends on the sample depth $z$ as

$$n^{SL}(\omega, z) = \begin{cases} n_{surf}(\omega), & (0 \leq z \leq d_{pump}), \\ n_{eq}(\omega), & (z > d_{pump}). \end{cases}$$  

(1)

Here, $n_{surf}(\omega)$ and $n_{eq}(\omega)$ are the complex refractive index at the photoexcited surface region and that in equilibrium. By considering the multiple reflections inside the photoexcited surface region, we compute the total optical response of the sample observed by the THz probe pulse $n_{eff}(\omega)$. Finally, we obtain the refractive index at the surface $n_{surf}(\omega)$ such that $n_{eq}(\omega)$ reproduces the experimentally observed complex refractive index.

In Fig. 5, we plot the real and imaginary parts of the optical conductivity at the photoexcited surface region at 100 K obtained by the SL model. As shown in Fig. 5(b), the imaginary part of the optical conductivity $\sigma_2(\omega)$ measured at $t_{pp} = 0.8$ ps (red circles) exhibits the $1/\omega$-like behavior, which coincides well with that at 40 K in equilibrium without the photoexcitation (blue dashed curve). Concomitantly, the real part of the optical conductivity $\sigma_1(\omega)$ exhibits a slight increase as plotted in Fig. 5(a). Although the spectral feature of $\sigma_2(\omega)$ at $t_{pp} = 0.8$ ps is almost identical to that in SC state, the pump-induced increase in $\sigma_1(\omega)$ and $\sigma_2(\omega)$ at $t_{pp} = 0.8$ ps can be simultaneously fitted by the Drude model:

$$\Delta \sigma(\omega) = \frac{i\varepsilon_0\omega_p^2}{\omega + i\gamma}$$

(2)

Here, $\varepsilon_0$ is the vacuum permittivity, and $\omega_p$ and $\gamma$ are the plasma angular frequency and scattering rate, respectively. Figures 5(a) and (b) show the fitting curves to the optical conductivity at $t_{pp} = 0.8$ ps with $\omega_p = 29$ THz and $\gamma/2 \pi = 0.14$ THz. Similarly, the pump-induced change in $\sigma_1(\omega)$ and
\( \sigma_2(\omega) \) measured at \( t_{pp} = 1.3 \) ps can be simultaneously fitted by the Drude model with \( \omega_p = 44 \) THz and \( \gamma/2\pi = 1.0 \) THz, as shown by the green circles in Figs. 5(c) and (d). Figure 5(e) summarizes the time evolution of the pump-induced change in the imaginary part of the optical conductivity \( \Delta \sigma_2(\omega) \). The vertical arrows denote the peaks in \( \Delta \sigma_2(\omega) \) spectra. The peak frequency increases with the delay time \( t_{pp} \), which is also identified in the previous study [24]. However, it is challenging to distinguish whether the observed transient increase in \( \sigma_2(\omega) \) reflects the SC or Drude response due to the limited frequency range of the THz spectroscopy. To clarify this problem, we examine the photoexcited nonequilibrium state in YBCO using the THz nonlinear optical responses arising from the Higgs mode (section B) and ac-Josephson current (section C).

Before moving to the THz nonlinear responses, we further investigate the temperature dependence of the photoinduced transient THz response. To study the temperature dependence of the photoinduced plasma-edge-like response above \( T_c \) with a better signal-to-noise ratio, we focus on the pump-induced change in the THz probe \( E \)-field instead of the transient optical conductivity spectrum as follows. Figure 6(a) shows the 800-nm pump-induced change in the THz \( E \)-field \( \Delta E(t_{gate}) \) above \( T_c \) as a function of the probe-gate delay time \( (t_{gate}) \) with the pump-probe delay time fixed at \( t_{pp} = 0.8 \) ps. The corresponding power spectra of \( \Delta E(t_{gate}) \), denoted by \( \Delta I \), are plotted in Fig. 6(b). The spectral weight of \( \Delta I \) integrated from 0.4 THz to 1.0 THz divided by the reflected THz power without photoexcitation, \( \int d\omega \frac{\Delta I}{I} \), is displayed in Fig. 6(c) as a function of the pump-probe delay time \( t_{pp} \) (magenta, left axis). The dynamics of frequency-integrated \( \Delta I/I \) follows the time evolution of \( [\omega \sigma_2(\omega)]^2 \) at \( \omega/2\pi = 0.4 \) THz obtained from the SL model analysis (light blue, right axis). Thus, the frequency-integrated intensity \( \Delta I/I \) can be used as an indicator of the pump-induced increase in \( \sigma_2(\omega) \). We present the temperature dependence of \( \Delta I/I \) in Fig. 6(d). Here, the black horizontal line denotes the noise floor of \( \Delta I/I \) at 300 K estimated from the data at the negative delay, \( t_{pp} = -9.6 \) ps. As the temperature increases, the integrated intensity \( \Delta I/I \) decreases and approaches the noise floor around 210 K which is consistent with the previous study of OPTP measurements for similarly doped YBCO [24]. Notably, this temperature is close to the pseudogap opening temperature \( T^* \) of similarly doped YBCO, reported as 235±10 K in neutron scattering [69], while lower than 260±40 K in Nernst measurement [70] and 270±20 K in optical spectroscopy measurement [71]. We will discuss this point in the discussion section.

In the following, to gain a deeper insight into the origin of the NIR pump-induced \( 1/\omega \)-like behavior in \( \sigma_2(\omega) \), we investigate the THz nonlinear optical responses arising from the SC collective excitations in the photoexcited state.
B. Photoexcited nonequilibrium state studied by THz THG

1. THz THG measurements

In Fig. 7(a), we present the power spectra of the reflected narrowband THz E-field polarized along the c-axis of UD61 at the selected temperatures. The TH component is clearly identified at 1.5 THz below $T_c$. Figures 7(b) and (c) show the integrated FH and TH intensities as a function of the incident THz peak E-field ($E_{in}$) at 20 K and 60 K, respectively. Here, we integrate the FH intensity from 0.3 to 0.7 THz and the TH intensity from 1.2 to 1.8 THz. At 20 K, the TH intensity follows $E_{in}^6$ and the FH intensity follows $E_{in}^2$, indicating that the observed TH component originates from the THG process in the SC state. In contrast, at 60 K, the integrated intensity around 1.5 THz (denoted as TH in Fig. 7(c) for convenience) follows $E_{in}^2$. This 1.5 THz component at 60 K is attributed to the finite leakage of the FH component, which transmits the 1.5-THz BPF placed after the sample. We note that the integrated TH intensity for the lowest $E$-field deviates from the expected power law dependence displayed by solid lines in both Figs. 7(b) and (c), because the signal level is close to the noise floor, which is estimated by integrating the power spectra from 2.5 to 3.1 THz, i.e., outside the TH frequency range [72,73].

To discuss the temperature dependence of the TH intensity, we evaluate the third-order nonlinear susceptibility normalized by the FH penetration depth $\chi^{(3)}_{dep}$ [74,75]. Here, we subtract the leakage of the FH intensity from the TH intensity and consider the Fresnel reflection loss of the incident FH E-field at the sample surface. The details of the analysis for $\chi^{(3)}_{dep}$ are described in Appendix G. The temperature dependence of $\chi^{(3)}_{dep}$ is plotted by the red circles in Fig. 7(d), showing a good agreement with that of the superfluid density $n_s$ obtained by the THz-TDS measurements represented by the gray circles. This agreement is consistent with the temperature dependence expected for the THG mediated by the THz-driven ac-Josephson current, which is proportional to the superfluid density [53]. Therefore, the observed THG is attributed to the ac-Josephson current driven by the intense THz pulse polarized along the c-axis and serves as the ultrafast probe of the SC order parameter under the photoexcitation. In the following, we utilize this THz-THG signal to investigate the photoinduced nonequilibrium state.

2. OP-THz THG measurements

Figure 8(a) illustrates the schematic of the OP-THG measurements for UD61 in reflection geometry. In Fig. 8(b), we show the power spectra of the reflected narrowband THz pulse at 20 K when the 800-nm photoexcitation is “On” (blue) and “Off” (gray) with the pump-probe delay time of $t_{pp} = 3.2$ ps. The frequency-integrated TH intensity $I_{TH}$ (integrated from 1.2 to 1.8 THz) decreases by 5.9% when the photoexcitation is “On” compared to that when the photoexcitation is “Off.” Following the destroyed layer model described in Appendix D, the superconductivity is destroyed
within the photoexcited surface depth of 0.94 µm with the pump fluence of 3.4 mJ/cm². In this situation, the TH intensity is estimated to decrease by 5.3% compared to that without photoexcitation, consistent with the OP-THG results. Figure 8(c) plots the pump-induced change in the TH intensity \( \Delta I_{TH} \) at 20 K at the selected delay times. For all the delay times, the TH intensity is reduced, and no enhancement is identified within the error bars. These results reinforce that the NIR photoexcitation destroys the superconductivity below \( T_c \).

Next, we examine the photoinduced nonequilibrium state above \( T_c \) by the OP-THG spectroscopy. As described in the Results section A, in the OPTP measurements at 100 K with the pump fluence of 3.4 mJ/cm², we observed the \( 1/\omega \)-like spectral profile in \( \sigma_2(\omega) \) at \( t_{pp} = 0.8 \) ps, which is considerably identical to that in the equilibrium SC state at 40 K. If this transient state can be recognized as the 40 K-superconductivity, one should expect a finite THG at 100 K after photoexcitation.

First, to estimate the expected TH intensity in the photoexcited nonequilibrium state at 100 K, we consider the penetration depth mismatch between the 800-nm pump and 0.5-THz pulses, schematically drawn in Fig. 9(a). Assuming that the photoexcited surface region of the sample turns to the SC state, whose superfluid density \( n_s \) is the same as that at 40 K, that surface region should have the same third-order nonlinear susceptibility \( \chi^{(3)}_{dep} \) as that at 40 K. Since the 800-nm pump beam diameter is 2 mm and sufficiently larger than that of the THz pulse (0.55 mm), the volume of the SC region should be simply proportional to the pump penetration depth \( d_{pump} \). Here, the penetration depth at 800 nm is set to \( d_{pump} = 0.22 \) µm from the literature [32]. We calculate the penetration depth for 0.5 THz at 40 K as \( d_{THz} = 5.7 \) µm using the refractive index obtained in the THz-TDS measurements. Hence, the ratio of the SC volume at 100 K in the photoexcited state measured at \( t_{pp} = 0.8 \) ps to that at 40 K in equilibrium is estimated as \( d_{pump}/d_{THz} = 3.9\% \).

Second, we evaluate the FH \( E \)-field penetrating inside the sample, as discussed in Appendix G. For the FH \( E \)-field inside the sample at 100 K after photoexcitation, we consider the effect of the multiple reflections inside the photoexcited surface region. Using the pump-induced surface refractive index at 100 K measured at \( t_{pp} = 0.8 \) ps and refractive index at 40 K in equilibrium, the ratio of \( B_{THG}(\omega) \) (defined by Eq. (G5) in Appendix G) at 0.5 THz is computed as 44. Combining this result with the SC volume ratio, we estimate that \( (d_{pump}/d_{THz})^2 B_{THG}(\omega) = (0.039)^2 \times 44 = 6.7\% \) of the TH intensity at 40 K without the photoexcitation should be observed at 100 K in the photoexcited state at \( t_{pp} = 0.8 \) ps.

In the upper panel of Fig. 9(b), we present the 800-nm pump-induced change in the TH power spectrum \( \Delta I_{TH} \) at 100 K measured at \( t_{pp} = 0.8 \) ps and the TH power spectrum \( I_{TH} \) at 40 K without the photoexcitation multiplied by 6.7%. Importantly, the pump-induced change in the TH signal, \( \Delta I_{FH} \), was not identified at 100 K within the error bars (see the lower panel of Fig. 9(b) for the
enlarged view) despite the emergence of $1/\omega$-like spectral profile in $\sigma_2(\omega)$ upon the photoexcitation. Figure 9(c) displays the pump-induced change in the FH power spectrum ($\Delta I_{FH}$, upper panel) and the TH power spectrum ($\Delta I_{TH}$, lower panel) as a function of the delay time $t_{pp}$ measured at 100 K. Although $\Delta I_{FH}$ increases after the photoexcitation, $\Delta I_{TH}$ does not exhibit a measurable change at any delay time. Furthermore, we simulate the time evolution of the THz TH intensity after the photoexcitation by assuming that the SC order is transiently induced by the photoexcitation, as shown in Fig. 9(d) (see Appendix H for the simulation details). The simulated THz TH intensity follows the dynamics of the pump-induced SC order represented in the inset of Fig. 9(d). These results apparently contradict the interpretation that the $1/\omega$-like spectral profile in the transient $\sigma_2(\omega)$ spectrum is attributed to the superconductivity.

In the next section, we examine the Higgs-mode response with THz $E$-field polarized along the $ab$-plane of the UD78 YBCO sample, which directly manifests the SC order parameter.

**C. Photoexcited nonequilibrium state studied by TPOP spectroscopy**

1. **TPOP measurements**

Figure 10(a) presents the THz pump-induced reflectivity change $\Delta R_{THz}/R$ for UD78 when the THz pump polarization is along the $a$-axis, and the 800-nm optical probe polarization is along the $b$-axis. At 20 K, we identify a negative signal in $\Delta R_{THz}/R$ with a rise time of $\sim 1$ ps and decay time of $\sim 5$ ps. As the temperature increases, this decaying component decreases and vanishes around $T_c$. This result supports the interpretation that the decaying component is ascribed to the THz pump-induced QPs excitation in the SC state, which was also observed in another cuprate superconductor Bi$_2$Sr$_2$CaCu$_2$O$_8+x$ (Bi2212) [43]. In addition to the decaying component, at 20 K, we identify a small shoulder at the delay time of $t_1 = 0.48$ ps and a peak at $t_2 = 1$ ps, which correspond to the peak positions of the squared THz $E$-field shown in the upper panel of Fig. 10(a). This signal is identified up to $T_c$. Accordingly, these shoulder and peak structure in the time-domain signal is attributed to the instantaneous THz nonlinear response which follows the squared THz $E$-field, i.e., the THz Kerr signal [76]. Correspondingly, the amplitude of $\Delta R_{THz}/R$ at $t_2$ is proportional to $E_{THz}^2$ as shown in Fig. 10(b). The THz Kerr effect has also been observed in underdoped Bi2212 single crystals and attributed to the Higgs mode response from the polarization and doping dependences [43]. Likewise, in the present case of YBCO the THz Kerr signal is considered to reflect a contribution from the Higgs mode. Recent theoretical studies have shown that there indeed exists the contribution of Higgs mode to the THz Kerr signal in cuprates [77] while its magnitude depends on the extent of disorder [78]. At present, further theoretical clarification on the relative magnitude of the Higgs-mode contribution and the other contributions, including QPs effect in the THz Kerr
signal, remains to be resolved. Here, we can use this THz Kerr signal as an indicator of the SC order parameter.

To decompose the THz pump-induced reflectivity change $\Delta R_{\text{THz}}/R$ for the UD78 sample into the THz Kerr and decaying components, we fit $\Delta R_{\text{THz}}/R$ with the following model [43]:

$$\frac{\Delta R_{\text{THz}}}{R}(t) = A \int_0^\infty dt' E_{\text{THz}}(t-t'-t_0)^2 \exp\left(-\frac{t'}{\tau_a}\right)$$

$$+ B \exp\left(-\frac{t-t_0}{\tau_b}\right) \left[1 - \text{erf}\left(-\frac{4(t-t_0)\tau_d+\tau_d^2}{2\sqrt{2}\tau_d\tau_b}\right)\right].$$

(3)

The first term denotes the THz Kerr component, which follows the squared THz $E$-field $E_{\text{THz}}(t)^2$ convoluted with an exponential function with a decay constant of $\tau_a = 300$ fs. The second term describes the decaying component, whose rise and decay times are described by $\tau_b$ and $\tau_d$, respectively. The parameters of $t_0$ and $\tau_b$ are determined at 20 K and fixed for all the temperatures. The amplitude of the THz Kerr component ($A$), that of the decaying component ($B$), and the relaxation time of the decaying component ($\tau_d$) are the fitting parameters. We note that the amplitude of the THz Kerr component $A$ is fixed above $T_c$. The gray curve in Fig. 10(a) is the fitting curve for each temperature.

In Fig. 10(c), we summarize the temperature dependence of the THz Kerr and the decaying components. The absolute amplitude of the decaying component $B$ increases below $T_c$, which reinforces the interpretation that the decaying component is attributed to the QPs in the SC state. Furthermore, the absolute amplitude of the THz Kerr component $A$ displays a sharp increase below $T_c$, indicating its relevance to the superconductivity.

Having established that the TPOP spectroscopy can probe the SC order parameter, we apply this ultrafast spectroscopic technique to examine the photoinduced nonequilibrium state in the $ab$-plane of the sample through the OP-TPOP measurements.

2. OP-TPOP measurements

The configuration of the OP-TPOP measurements is schematically depicted in Fig. 11(a). First, the 1.4-$\mu$m pump pulse drives the system out of equilibrium. Next, the pump-induced change in the system is detected through the THz pump-induced reflectivity change $\Delta R_{\text{THz}}/R$ probing at 800 nm. If the 1.4-$\mu$m pump-800-nm probe delay time ($t_{\text{NIR}-\text{Pr}}$) is fixed, one can measure the THz pump-induced reflectivity change $\Delta R_{\text{THz}}/R$ under the 1.4-$\mu$m pump-induced nonequilibrium state by sweeping the delay time of the THz pump pulse with respect to the 800-nm probe pulse ($t_{\text{THz}-\text{Pr}}$).

Here, by sweeping the delay stage of the THz pulse while fixing that of the 800-nm probe pulse, $\Delta R_{\text{THz}}/R$ at all $t_{\text{THz}-\text{Pr}}$ records the same effect induced by the 1.4-$\mu$m pump illuminated at $t_{\text{NIR}-\text{Pr}}$, as investigated in the previous OPTP measurements [4,79-84]. Since we identify the NIR pump-
induced increase in the imaginary part of the transient optical conductivity of the UD61 sample when the NIR pump polarization is along the $a$-axis (Appendix E) or the NIR pump wavelength is tuned to 1.5 µm (Appendix F), here we employ the 1.4-µm pump pulse polarized along the $a$-axis, which can be easily separated from the reflected 800-nm probe pulse from the sample.

Figure 11(b) plots the THz pump-induced reflectivity change $\Delta R_{\text{THz}}/R$ at 20 K as a function of $t_{\text{THz-Pr}}$ with (red curve, denoted by “On”) and without (gray curve, denoted by “Off”) the 1.4-µm NIR excitation when the 1.4-µm pump-800-nm probe delay time is fixed at $t_{\text{NIR-Pr}} = 0.5$ ps. While the extraction of the instantaneous THz Kerr response from $\Delta R_{\text{THz}}/R$ signal is difficult due to the limited signal-to-noise ratio in the OP-TPOP measurements, the overall amplitude of $\Delta R_{\text{THz}}/R$ decreases with the 1.4-µm optical excitation, in good agreement with the photoinduced destruction of the superconductivity below $T_c$ observed in the OPTP and OP-THG experiments.

In Fig. 12(a), we present the THz pump-induced reflectivity change $\Delta R_{\text{THz}}/R$ at 100 K, i.e., above $T_c$ as a function of $t_{\text{THz-Pr}}$ with (red curve, denoted by “On”) and without (gray curve, denoted by “Off”) the 1.4-µm photoexcitation under the 1.4-µm pump fluence of 560 µJ/cm², for the delay time between the 1.4-µm pump and 800-nm probe pulses fixed at $t_{\text{NIR-Pr}} = 0.4$ ps. For comparison, we show $\Delta R_{\text{THz}}/R$ at 20 K without the 1.4-µm photoexcitation by the blue dotted curve in Fig. 12(a). Qualitatively, neither the THz Kerr component nor the decaying component emerges at any $t_{\text{THz-Pr}}$’s investigated here. To quantitatively discuss the amplitude of $\Delta R_{\text{THz}}/R$ at 100 K with the NIR photoexcitation, we integrate $\Delta R_{\text{THz}}/R$ from $t_{\text{THz-Pr}} = 0$ to 2.4 ps (the red shaded area in Fig. 12(a)) and compare it to that without the NIR photoexcitation. Figure 12(b) plots the integrated $\Delta R_{\text{THz}}/R$ without the NIR photoexcitation as a function of temperature. Apparently, the integrated $\Delta R_{\text{THz}}/R$ at 100 K shown by the red diamond remains intact upon the NIR photoexcitation within the error bar and does not show an enhancement that is expected if one interprets the $1/\omega$-like response in $\sigma_2(\omega)$ as a signature of the photoinduced superconductivity. These results indicate that the NIR photoexcited state in the UD78 sample is unlikely attributed to the SC state equivalent to that below $T_c$ in equilibrium.

IV. DISCUSSION

In our study, neither the $ab$-plane nonlinear THz signal, including the THz Kerr effect and the incoherent QPs response, nor the nonlinear Josephson current along $c$-axis is identified in the NIR photoexcited nonequilibrium state above $T_c$, while all of them are observed in the equilibrium SC phase below $T_c$. Hence, it is hard to attribute the NIR pump-induced $1/\omega$-like spectral response observed in the imaginary part of the optical conductivity $\sigma_2(\omega)$ to the SC phase equivalent to that in equilibrium. Here, we should note that the similar $1/\omega$-like response in $\sigma_2(\omega)$ previously reported under the MIR pump excitation in YBCO may have a different origin, which shows a clear
resonance when the pump photon energy is tuned to the apical oxygen phonon energy [3,4,32]. Recently, the parametric amplification of the Josephson plasmons mediated by the resonantly driven phonon at 80 meV using the MIR pump pulse has been proposed as a mechanism for the photoinduced superconductivity [15,18,34]. This parametric process might be one possible scenario to explain the $1/\omega$-like transient response in $\sigma_2(\omega)$ in the present study with 800 nm (1.55 eV) or 1.4 μm (0.89 eV) NIR optical pump if the NIR pump pulses can drive the phonon at 80 meV off-resonantly. As presented in Appendix E, however, the $1/\omega$-like response in $\sigma_2(\omega)$ is also observed when the 800-nm pump is polarized along the $a$-axis, making a stark contrast to the previous work with the MIR pump where the $1/\omega$-like response was observed only for the $c$-axis pump [24]. This polarization dependence indicates that the apical oxygen phonon is irrelevant in the case of the NIR photoexcitation. Furthermore, it should be noted that in the proposed framework of this parametric process, the superconductivity should be transiently enhanced even below $T_c$ [15,18,34]. On the contrary, our experimental results demonstrate that the 800-nm photoexcitation destroys the superconductivity both in the $ab$-plane and along the $c$-axis. Therefore, it is hard to attribute the present result of NIR photoexcitation to the parametrically amplified plasmon process.

It is highly nontrivial that the photoexcited state in underdoped YBCO far above $T_c$ exhibits such a coherent carrier transport along the $c$-axis with a substantially low scattering rate, less than 0.15 THz, albeit with the incoherent $c$-axis transport in equilibrium [85]. Even though the photoinduced state lacks the SC collective modes, it is still tempting to infer that the observed coherent charge carrier response is related to superconductivity. Following this scenario, the phase-fluctuating SC state without the long-range order may be a candidate to explain the lack of the SC collective modes. In this view, it is indicative that the coherent Drude responses have been observed previously in equilibrium along the $c$-axis far above $T_c$ [86], and for the in-plane response even below $T_c$ [87], both of which were interpreted as a signature of the phase-fluctuating superconductivity. It is also notable that a recent theoretical study has shown that photoexcitation enhances the $d$-wave SC correlation but with a substantially short pairing correlation length [88]. Though it assumes the in-plane photoexcitation of the stripe-ordered cuprates, the enhanced superconductivity with substantially short-range correlation may be relevant to our scenario of the photoinduced phase-fluctuating superconductivity.

Another possible scenario is that the observed SC-like behavior in $\sigma_2(\omega)$ is associated with the pseudogap because the 800-nm pump-induced THz reflectivity change persists up to the pseudogap opening temperature $T^*$, as mentioned in the Results section A-3. While we cannot exclude the possibility of a simple coincidence, there are two reasons why we presume that this onset temperature is possibly related to the pseudogap. First, the photoinduced transient increase in $\sigma_2(\omega)$ has also been reported in variously underdoped YBCO samples up to $T^*$ with the MIR [3,4,24] and
NIR pulse excitation [33]. Second, previous optical spectroscopy [56,71,89-91] and theoretical studies [92-94] have revealed that the equilibrium c-axis optical response of cuprates is sensitive to the electronic density of states in the antinodal region where the pseudogap opens: the interlayer hopping integral for cuprates strongly depends on the \( ab \)-plane momentum \( \mathbf{k} \) of carriers as \( t_\perp(\mathbf{k}) \propto [\cos(k_a) - \cos(k_b)]^2 \) (\( a \) and \( b \) are the lattice constants for \( a \) and \( b \) axis, respectively) [92,93], which is maximum at the antinode. Accordingly, the photoinduced SC-like behavior in \( \sigma_2(\omega) \) might be related to the pseudogap in the present case of YBCO.

Though the origin of the pseudogap is still under intensive debate [91,95-98], some theoretical studies have recently suggested the presence of a pair-density wave (PDW) state at high temperature above \( T_c \) and its relation with the pseudogap has been discussed [99-101]. The existence of the PDW has been reported by the scanning tunneling microscopy (STM) measurements in Bi2212 far below \( T_c \) [102,103] and even in the high-temperature regime up to \( 1.5T_c \) [104]. Interestingly, it has been theoretically proposed that the photoexcited state of the PDW can exhibit the \( 1/\omega \)-like response in \( \sigma_2(\omega) \) without showing the Meissner effect [16]. This scenario does not seem to contradict our experimental results that the collective excitations of the SC order parameter are absent in the photoexcited state out of equilibrium.

V. CONCLUSION

We studied the NIR pump-induced nonequilibrium state in underdoped YBCO samples. First, we performed the OPTP spectroscopy for the UD61 sample along the c-axis. We observed the redshift of the Josephson plasma edge in the transient THz reflectivity spectrum after the 800-nm photoexcitation below \( T_c \). On the other hand, we identified the \( 1/\omega \)-like response in the imaginary part of the optical conductivity \( \sigma_2(\omega) \) immediately after the photoexcitation above \( T_c \) up to the pseudogap temperature, consistent with the previous reports [3,4,24,33].

Next, we investigated the NIR photoexcited nonequilibrium state using the THz nonlinear optical responses in the SC state: THG signal arising from ac-driven Josephson current (c-axis) and THz-Kerr effect (\( ab \)-plane), both of which act as an indicator of the SC order parameter. Below \( T_c \), these THz nonlinear responses displayed a reduction in intensity upon the photoexcitation, confirming that the photoexcitation destroys the superconductivity. On the other hand, neither the THz nonlinear response from the ac-Josephson current nor the \( ab \)-plane THz Kerr effect was identified in the NIR photoexcited nonequilibrium state above \( T_c \), despite the appearance of the \( 1/\omega \)-like transient response in the imaginary part of the c-axis optical conductivity. These results indicate that the photoinduced state that exhibits the \( 1/\omega \)-like response along the c-axis imaginary part of the optical conductivity is distinct from the equilibrium SC state below \( T_c \).

At present, it is hard to specify the microscopic origin of the unusually coherent charge carrier response along the c-axis.
that appeared in the photoexcited state of underdoped YBCO above $T_c$, but it deserves further experimental and theoretical investigation and will provide important insights for the understanding of electronic states above $T_c$ in underdoped cuprate superconductors.

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APPENDIX A: DETERMINATION OF THE SUPERCONDUCTING TRANSITION TEMPERATURE $T_c$

Figures 13(a) and (b) show the magnetic moments for UD61 and UD78 YBCO single crystals measured by SQUID under zero-field cooling. The SC transition temperature $T_c$ is determined by the onset of the drop in the magnetic moment.

APPENDIX B: EQUILIBRIUM OPTICAL CONSTANTS

We measured the optical constants along the $c$-axis of UD61 in equilibrium using the THz-TDS measurements in reflection geometry. Since the frequency range of the obtained optical constants is limited above 0.7 THz due to the sample size, the optical constants below 0.7 THz are estimated by fitting the complex optical conductivity $\sigma(\omega) = -i\varepsilon_0\omega(\varepsilon(\omega) - \varepsilon_b)$ with the following two-fluid model [32,33,105]:

$$\varepsilon(\omega) = \varepsilon_b - \frac{n_s}{\omega^2} - \frac{\omega_d^2}{\omega^2 + i\gamma_d\omega} - \sum_j \frac{S_j^2}{\omega^2 - \omega_j^2 + i\gamma_j\omega}. \quad (B1)$$

Here, $n_s$ is superfluid density and set to zero above $T_c$, $\omega_d$ and $\gamma_d$ are the plasma angular frequency and the scattering rate of the Drude component, respectively. The scattering rate $\gamma_d$ is fixed to 10 THz, which is a typical value for the similarly doped YBCO [33,105]. The parameters of $\omega_j$, $\gamma_j$, and $S_j$ are the central angular frequency, scattering rate, and oscillator strength of the $j$-th oscillator representing the infrared active phonons, respectively. We adopt the parameters of these oscillators taken from Ref. [105] and fixed at all temperatures. $\varepsilon_b$ is the background dielectric constant of YBCO and set to 4.5 from the literature value [106].

The fitting results for the real and imaginary parts of the optical conductivity are shown in Figs. 14(a) and (b), respectively. Whereas the fitting reasonably reproduces the imaginary part of the
optical conductivity $\sigma_2(\omega)$, the real part $\sigma_1(\omega)$ slightly deviates from the fitting, possibly because the phonon parameters are fixed. Figures 14(c) and (d) plots the temperature dependence of the fitting parameters $n_s$ and $\omega_d^2$, respectively. The superfluid density $n_s$ displays an increase below $T_c$, manifesting the growth of the SC phase stiffness.

**APPENDIX C: THE DETAILS OF THE HEATING MODEL SIMULATION**

We perform the heating model simulation reported in the previous work of Ref. [30] as follows. Because the typical electron-phonon scattering time in cuprate is $\sim 6$ ps [66], the pump pulse energy is absorbed by the sample after this time scale. Thus, it is reasonable to consider that the sample temperature reaches a quasi-equilibrium value after $t_{pp} = 6$ ps. Assuming that the optical pump fluence $F_0$ decays exponentially inside the sample with the pump penetration depth $d_{\text{pump}}$ as $F(z) = F_0 \exp(-z/d_{\text{pump}})$, one can compute the pump energy density absorbed at the depth $z$ of the sample $I(z)$ as

$$I(z) = (1 - R) \left( -\frac{dF(z)}{dz} \right) = (1 - R) \frac{F_0}{d_{\text{pump}}} \exp \left( -\frac{z}{d_{\text{pump}}} \right), \quad (\text{C1})$$

where $F_0$ is the pump fluence, and $R$ is the reflectivity at the pump wavelength. In the case of 800-nm pump pulse, $R = 0.1$ and $d_{\text{pump}} = 0.22$ µm from the literature [32]. If the energy of $I(z)$ is absorbed by the sample, the temperature at the depth $z$ of the sample $T_f(z)$ can be connected to $I(z)$ by the following integral equation:

$$I(z) = \int_{T_i}^{T_f(z)} dT N C_s(T). \quad (\text{C2})$$

Here, $T_i$ is the initial temperature of the sample, $C_s(T)$ is the specific heat of the sample, and $N$ is the number of molecules in the excited volume. In the case of YBCO, it is known that the specific heat $C_s(T)$ can be described by the following equation over a wide temperature range [32,67,107]:

$$C_s(T) = \gamma_s T + \beta_s T^3, \quad (\text{C3})$$

where $\gamma_s = 2.3$ mJ mol$^{-1}$ K$^{-2}$ and $\beta_s = 0.394$ mJ mol$^{-1}$ K$^{-4}$ are the electronic and lattice coefficients to the specific heat taken from the literature values [67]. We compute $T_f(z)$ for a given $F_0$ by numerically solving Eq. (C2) with Eqs. (C1) and (C3). Using the temperature dependence of the complex refractive index [we present the real part $n_1(\omega)$ in Fig. 15(a) and the imaginary part $n_2(\omega)$ in Fig. 15(b) for selected frequencies], the computed $T_f(z)$ is converted to the spatial distribution of the complex refractive index as shown in Figs. 15(c) and (d).

Lastly, using the obtained spatial distribution of the complex refractive index, we calculate the effective optical response by dividing the sample surface region into layers [3,4,30]. In this study,
we divided the sample surface of thickness 30 μm into 3000 layers and calculated the effective refractive index as a result of the multiple reflections. We summarize the obtained transient reflectivity $R$, reflectivity change $\Delta R/R$, and phase shift $\theta$ in Figs. 3(d)-(f) for the pump fluence of $F_0 = 3.4$ mJ/cm$^2$ (red) and 0.4 mJ/cm$^2$ (blue).

**APPENDIX D: DESTROYED LAYER MODEL**

To elucidate the optical properties within ~ 6 ps after photoexcitation when the hating model is not applicable, we develop the following “destroyed layer model” as schematically illustrated in Fig. 16(a). Here, we assume that the superconductivity in the photoexcited surface region with a depth of $d_0$ is destroyed and the optical constants are described by that at 60 K. Firstly, by considering the multiple reflections of the THz probe pulse inside the photoexcited surface region, we compute the effective reflectivity $R_{\text{dest}}$. Figure 16(b) shows that the THz reflectivity change spectra $\Delta R/R$ at 20 K measured at $t_{pp} = 3.2$ ps are reasonably fitted with $(R_{\text{dest}} - R_{\text{eq}})/R_{\text{eq}}$, where $d_0$ is the only fitting parameter and $R_{\text{eq}}$ denotes the equilibrium reflectivity.

Secondly, if the superconductivity is destroyed within the thickness of $d_0$ at the sample surface, the attenuated pump intensity at the sample depth of $z = d_0$ should reach the SC threshold energy $I_{\text{th}},$

$$F_0 \exp \left(-\frac{d_0}{d_{\text{pump}}} \right) = F_{\text{th}}.$$  \hspace{1cm} (D1)

Thus, the destroyed layer length $d_0$ can be written as

$$d_0 (I_0) = \begin{cases} d_{\text{pump}} \ln \left(\frac{F_0}{F_{\text{th}}} \right) + d_{\text{offset}} \left( F_0 \geq F_{\text{th}} \right) \\ d_{\text{offset}} \left( 0 \leq F_0 < F_{\text{th}} \right). \end{cases}$$  \hspace{1cm} (D2)

Here, we add an offset $d_{\text{offset}}$ to prevent $d_0$ going to zero for $F_0 < F_{\text{th}}$. The destroyed layer length $d_0$ obtained by the fitting in Fig. 16(b) is plotted as a function of the pump fluence $F_0$ in Fig. 16(c). We fit the obtained $d_0$ using Eq. (D2) with the adjustable fitting parameters of $F_{\text{th}}$ and $d_{\text{offset}}$ while fixing the 800-nm pump penetration depth $d_{\text{pump}} = 0.22$ μm [32]. One can see that the obtained $d_0$ follows the fitting curve by Eq. (D2) and the SC threshold energy is estimated as $F_{\text{th}} = 70$ μJ/cm$^2$, whose energy scale is in good agreement with the results of the previous time-resolved pump-probe measurements [58-62] and ARPES measurements in cuprate superconductors [63,64]. Hence, the destroyed layer model reveals that the photoexcitation destroys the superconductivity even before the system reaches the thermal quasi-equilibrium state.
APPENDIX E: PUMP POLARIZATION DEPENDENCE IN THE C-AXIS TRANSIENT OPTICAL RESPONSES

We performed the OPTP experiments by setting the 800-µm pump polarization along the a-axis while keeping the THz probe polarization along the c-axis. Figure 17 plots the 800-nm pump-induced transient reflectivity change $\Delta R/R$ along the c-axis of UD61 at 20 K. $\Delta R/R$ can be fitted by the destroyed layer model described in Appendix D using the parameter of $d_0 = 0.43$ µm. This value of $d_0$ is longer than the 800-nm pump penetration depth of 0.11 µm along the a-axis, which is computed from the optical constants in the literature [108], as expected in the destroyed later model.

In Fig. 18, we present the transient increase in the c-axis optical conductivity at the photoexcited surface at 100 K when the pump polarization is parallel to the c-axis (red) and a-axis (green) using the single layer model in Eq. (1). Remarkably, both the real and imaginary parts of the optical conductivity for the a-axis pump exhibit increase, similar to those for the c-axis pump. These results reveal that the NIR pump-induced transient increase in $\sigma_1(\omega)$ and $\sigma_2(\omega)$ does not depend on the pump polarization, contrary to the previous work with the MIR photoexcitation where the transient increase in $\sigma_2(\omega)$ was discerned only for the c-axis pump above $T_c$ [24].

APPENDIX F: PUMP WAVELENGTH DEPENDENCE IN THE C-AXIS TRANSIENT OPTICAL RESPONSES

In this Appendix F, we present the OPTP results using the 1.5-µm pump pulse. Figure 19(a) shows the 1.5-µm pump-induced transient reflectivity along the c-axis of UD61 at 20 K for the pump fluence of 0.75 mJ/cm². We identify a redshift of the JPR, indicating that the superconductivity is destroyed by the 1.5-µm photoexcitation. The corresponding pump-induced transient reflectivity change $\Delta R/R$ is presented in Fig. 19(b), which can be fitted by the destroyed layer model described in Appendix D using the parameter of $d_0 = 4.0$ µm. In Fig. 19(c), we plot the results of the destroyed layer model for the 1.5-µm photoexcitation. The pump fluence dependence of the destroyed layer length $d_0$ follows Eq. (D2), and the threshold energy to destroy the superconductivity is extracted as $F_{th} = 60$ µJ/cm², in good agreement with the case of the 800-nm photoexcitation.

Figure 20 displays the 1.5-µm pump-induced transient optical conductivity at the photoexcited surface region at 100 K above $T_c$. Here, we adopt the 1.5-nm pump penetration depth of 0.78 µm for the c-axis from the literature [32]. The imaginary part of the optical conductivity $\sigma_2(\omega)$ exhibits an increasing trend toward lower frequency, and a concomitant increase in the real part $\sigma_1(\omega)$ is discerned. The overall behaviors both below and above $T_c$ are very similar to those for the 800-nm photoexcitation. It should be noted that these results are consistent with the previous OPTP
measurements for underdoped YBCO samples using similar NIR pump wavelengths of 1.4-µm [32] and 1.28-µm [33].

APPENDIX G: EVALUATION OF THE TEMPERATURE DEPENDENCE OF THE THz THG

In this appendix, we describe the procedure to evaluate the temperature dependence of the THz THG intensity arising from the ac-driven Josephson current. First, we remove the leakage of the FH component in the following manner. Since the FH leakage intensity at TH frequency \( I_{\text{leak}} \) follows the squared incident FH E-field \( E_{\text{in}}^2 \) and the third-order nonlinear TH intensity \( I_{\text{THG}} \) follows \( E_{\text{in}}^6 \), as shown in Figs. 7(b) and (c), the measured TH intensity \( I_{\text{meas}} \) is expressed as a function of the incident FH E-field \( E_{\text{in}} \) by

\[
I_{\text{meas}}(E_{\text{in}}) = I_{\text{THG}}(E_{\text{in}}) + I_{\text{leak}}(E_{\text{in}}) = \left( \chi^{(3)} E_{\text{in}}^3 \right)^2 + \alpha_{\text{leak}} E_{\text{in}}^2. \tag{G1}
\]

Here, \( \chi^{(3)} \) is the third-order nonlinear susceptibility, and \( \alpha_{\text{leak}} \) is a coefficient for the leakage. The leakage component can be subtracted by measuring the TH intensity for two different incident FH E-fields of \( E_{\text{in}} \) and \( \beta E_{\text{in}} \), where the coefficient \( \beta \) is controlled by wire grid polarizers in the path of the incident THz beam. In this case, the nonlinear TH intensity \( I_{\text{THG}}(E_{\text{in}}) \) can be written as

\[
I_{\text{THG}}(E_{\text{in}}) = \frac{I_{\text{meas}}(E_{\text{in}}) - I_{\text{meas}}(\beta E_{\text{in}})/\beta^2}{1 - \beta^4}. \tag{G2}
\]

During the measurements, \( \beta \) is set to 0.75. Figure 21(b) shows the obtained TH intensity \( I_{\text{THG}} \) as a function of temperature (magenta curve). We note that \( I_{\text{THG}} \) is the TH intensity integrated from 1.2 to 1.8 THz after subtracting the leakage. The TH intensity \( I_{\text{THG}} \) displays a sharp increase below \( T_c \) and a maximum at 52 K. This peak at 52 K is due to the screening effect of the THz E-field when decreasing the temperature, as discussed below.

The screening effect of the incident FH E-field into the sample and outgoing TH E-field from the sample is evaluated using the complex refractive index \( n(\omega) \) obtained by the THz-TDS measurements. The situation is depicted in Fig. 21(a). The screened FH E-field inside the sample \( E_{\text{scr}}(\omega_0) \) is related to the incident FH E-field \( E_{\text{in}}(\omega_0) \) as

\[
E_{\text{scr}}(\omega_0) = \frac{2}{1 + n(\omega_0)} E_{\text{in}}(\omega_0) = t_{12}(\omega_0) E_{\text{in}}(\omega_0). \tag{G3}
\]

Here, \( t_{12}(\omega_0) \) is the Fresnel transmission coefficient for the FH angular frequency \( \omega_0 \) from the air to the sample. The FH E-field inside the sample \( E_{\text{scr}}(\omega_0) \) generates the TH E-field \( E_{\text{sample}}(3\omega_0) = \chi^{(3)} E_{\text{scr}}(\omega_0)^3 \). Thus, the observed TH E-field \( E_{\text{THG}}(3\omega_0) \) is written by
\[ E_{\text{THG}}(3\omega_0) = \frac{2n(3\omega_0)}{1 + n(3\omega_0)} E_{\text{sample}}(3\omega_0) = t_{21}(3\omega_0)\chi^{(3)}E_{\text{scr}}(\omega_0)^3, \tag{G4} \]

where \( t_{21}(3\omega_0) \) is the Fresnel transmission coefficient for the angular frequency \( 3\omega_0 \) from the sample to the air. Using Eqs. (G3) and (G4), the third-order nonlinear susceptibility \( \chi^{(3)} \) is expressed as

\[ \chi^{(3)} = \frac{E_{\text{THG}}(3\omega_0)}{t_{12}(\omega_0)^3 E_{\text{in}}(\omega_0)^3 t_{21}(3\omega_0)} = \frac{\sqrt{B_{\text{THG}}(\omega_0) I_{\text{THG}}(3\omega_0)}}{I_{\text{in}}(\omega_0)^3}, \tag{G5} \]

Here, \( I_{\text{in}}(\omega_0) \) and \( I_{\text{THG}}(3\omega_0) \) are the intensity of the incident FH E-field and observed TH E-field, respectively. The coefficient \( B_{\text{THG}} \) at 0.5 THz is computed using the refractive index \( n(\omega) \) and plotted as the light blue dashed curve in Fig. 21(b).

Finally, we consider that the Josephson current induced by the FH incident field is proportional to the area perpendicular to the \( c \)-axis where the FH field penetrates inside the sample, because the nonlinear polarization is induced within the volume of the penetration depth from the surface by the THz pulse in reflection geometry [74,75]. If the FH beam area on the sample is fixed for all the temperatures, this volume is proportional to the FH penetration depth, which can be calculated as \( d_{\text{THz}}(\omega_0) = c/[2n(\omega_0)\omega_0] \). It is convenient to define the third-order nonlinear susceptibility per depth \( \chi_{\text{dep}}^{(3)} = \chi^{(3)}/d_{\text{THz}} \) when we consider the expected TH intensity at 100 K after photoexcitation. We present the obtained \( \chi_{\text{dep}}^{(3)} \) as a function of temperature in Fig. 7(d).

**APPENDIX H: SIMULATION OF THE THz THG INTENSITY AFTER PHOTOEXCITATION**

We simulate the time evolution of the THz THG intensity driven by the Josephson current under the 800-nm photoexcitation applying the procedure in Ref. [47], which is in the case of the Higgs mode. When a THz pulse polarized along the \( c \)-axis of YBCO \( (E(t) = E_0 \sin(\omega_0 t)) \) is irradiated, the interlayer phase difference of the SC order parameter \( \theta(t) \) advances in time according to the Josephson relation as

\[ \frac{\partial \theta(t)}{\partial t} = \frac{2e d}{\hbar} E(t), \tag{H1} \]

Here, \( 2e \) is the Cooper pair charge, \( d \) is the interlayer spacing, and \( \hbar \) is the Planck’s constant \( h \) divided by \( 2\pi \). Following the time evolution of \( t = \theta_0 \cos(\omega_0 t) \quad (\theta_0 = 2e d E_0/\hbar) \), the Josephson current \( I(t) \) is induced and given by
\[ I(t) = I_c \sin(\theta(t)) \approx I_c \left[ \theta_0 \cos(\omega_0 t) - \frac{\theta_0^3}{6} \cos^3(\omega_0 t) \right], \]  \quad (H2)

where \( I_c \) is the critical current and proportional to the superfluid density \( n_s \) \[109\]. For an intense THz \( E \)-field, the second term in the right side of Eq. (H2) gives rise to the THG.

Now we consider the time evolution of the THz THG intensity if the 800-nm photoexcitation induces the superconductivity. In the OP-THG experiment, the THz \( E \)-field was measured by sweeping the THz pulse arrival time at the sample \( t_{\text{probe}} \) while fixing the timing between the pump and the sampling pulse for the EO-sampling \( t_{pp} = t_{\text{samp}} - t_{\text{pump}} \), where \( t_{\text{samp}} \) is the timing of the sampling pulse for the EO-sampling and \( t_{\text{pump}} \) is the time when the 800-nm pump pulse arrives at the sample. This configuration ensures that the THz \( E \)-field waveform is composed of the data points measured at the same delay time \( t_{pp} \) \[4,79-84\]. Here, we assume that the superfluid density \( n_s \) is induced after 800-nm photoexcitation and advances in time \( t_{pp} \) as shown in the inset of Fig. 9(d). In the simulation, we employ the THz \( E \)-field used in the experiment \( E(t_{\text{samp}} - t_{\text{probe}}) \). Since the critical current \( I_c \) is proportional to \( n_s(t_{\text{samp}} - t_{\text{pump}}) \), Eq. (H2) can be rewritten as

\[ I^{wp}(t_{\text{samp}} - t_{\text{pump}}, t_{\text{samp}} - t_{\text{probe}}) \propto n_s(t_{\text{samp}} - t_{\text{pump}}) \sin(\theta(t_{\text{samp}} - t_{\text{probe}})). \]  \quad (H3)

Here, \( t_{\text{probe}} \) is the time when the THz pulse arrives at the sample. The emitted \( E \)-field from this Josephson current is expressed as

\[ E^{wp}(t_{\text{samp}} - t_{\text{pump}}, t_{\text{samp}} - t_{\text{probe}}) = -\frac{\partial}{\partial t_{\text{samp}}} I^{wp}(t_{\text{samp}} - t_{\text{pump}}, t_{\text{samp}} - t_{\text{probe}}). \]  \quad (H4)

By sweeping \( t_{\text{probe}} \) while keeping \( t_{pp} = t_{\text{samp}} - t_{\text{pump}} \), we can calculate \( E^{wp} \) for each \( t_{pp} \). The simulated TH intensity of \( E^{wp} \) is shown as a function of \( t_{pp} \) in Fig. 9(d).
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Figure 1 The $c$-axis equilibrium optical constants of UD61 obtained by THz time-domain spectroscopy (THz-TDS): (a) the reflectivity, (b) loss function, (c) real and (d) imaginary part of the optical conductivity.
Figure 2 (a) Schematic illustration of the optical pump-THz probe (OPTP) spectroscopy. (b) The 800-nm pump-induced transient reflectivity along the $c$-axis of UD61 at 20 K with the pump fluence of 3.4 mJ/cm$^2$ (blue solid curve). The pump polarization is set parallel to the $c$-axis. Eq in the legend denotes equilibrium. (c) The 800-nm pump-induced transient reflectivity change at 20 K with the pump fluence of 3.4 mJ/cm$^2$ at selected pump-probe delay times $t_{pp}$. The delay time $t_{pp}$ for each data is shown in the right. (d) The pump fluence $F(z)$ for $F_0 = 3.4$ mJ/cm$^2$ (magenta curve, left axis) and the corresponding sample temperature $T_f(z)$ (light blue curve, right axis) obtained by the heating model simulation for the initial sample temperature of $T_i = 20$ K.
Figure 3 Comparison of the optical constants in the photoexcited nonequilibrium state obtained in (a-c) the experiment and those computed by (d-f) the heating model simulation. (a) The pump-induced transient reflectivity, (b) reflectivity change $\Delta R/R$, and (c) phase shift $\theta$ at 20 K measured at $t_{pp} = 12.8$ ps. Here, both the pump and probe polarizations are parallel to the c-axis of UD61. (d), (e) and (f) are the corresponding results computed by the heating model simulation. The inset in (d) shows the imaginary part of the optical conductivity at the surface (magenta) and the bulk (light blue) used in the heating model simulation for the pump fluence of 3.4 mJ/cm$^2$. 
Figure 4 (a) The 800-nm pump-induced reflectivity change along the $c$-axis of UD61 at 100 K with the pump fluence of 3.4 mJ/cm². The pump polarization is set parallel to the $c$-axis. The error bars are displayed as colored bands. The blue dashed curve shows the result of the heating model simulation at 100 K. (b) The spatial distribution of the pump fluence $F(z)$ (magenta, left axis) and the sample temperature $T_f(z)$ (light blue, right axis) computed by the heating model simulation for the initial sample temperature of $T_i = 100$ K.
Figure 5 (a), (b) The real and imaginary parts of the transient optical conductivity along the c-axis of UD61 at 100 K with the 800-nm pump fluence of 3.4 mJ/cm² measured at $t_{pp} = 0.8$ ps (red circles). Here, the pump polarization is along the c-axis. Eq in the legend denotes equilibrium. The solid orange curves are the fits to the data using the Drude model. (c), (d) The real and imaginary parts of the transient optical conductivity along the c-axis at 100 K with the pump fluence of 3.4 mJ/cm² measured at $t_{pp} = 1.3$ ps (green circles). The solid orange curves are the fits to the data using the Drude model (e) The time evolution of the pump-induced change in the imaginary part of the optical conductivity at 100 K with the 800-nm pump fluence of 3.4 mJ/cm². The delay time $t_{pp}$ for each data is shown in the right. The vertical arrows denote the peak positions in $\Delta \sigma_2(\omega)$.
Figure 6 (a) The 800-nm pump-induced change in the reflected THz $E$-field waveform along the $c$-axis of UD61 (denoted by $\Delta E(t_{gate})$) with the pump fluence of 3.4 mJ/cm$^2$ measured at the indicated temperatures above $T_c$. The pump polarization is parallel to the $c$-axis. (b) The FFT intensity of $\Delta E(t_{gate})$ (denoted by $\Delta I(\omega)$) corresponding to (a). The standard errors of the measurements are displayed as colored bands. (c) The integrated intensity of $\Delta I(\omega)$ normalized by the reflected THz power as a function of the pump-probe delay time $t_{pp}$ measured at 100 K (magenta, left axis). The integral region is shown by the shaded area (magenta) in (b). The horizontal black dashed line denotes the noise floor estimated from the integrated FFT intensity at the negative delay time. The squared pump-induced change in the imaginary part of the optical conductivity multiplied by the angular frequency, $(\omega \Delta \sigma_2)^2$, at $\omega/2\pi = 0.4$ THz is also shown in the right axis (light blue). (d) The integrated FFT intensity of $\Delta I(\omega)$ normalized by the reflected THz power without photoexcitation as a function of temperature. The horizontal dashed black line indicates the noise floor estimated from the integrated FFT intensity at 300 K measured at $t_{pp} = -9.6$ ps.
Figure 7 (a) The FFT power spectra of the reflected narrowband THz $E$-field from UD61 at selected temperatures using the THz peak $E$-field of 25 kV/cm. (b) The frequency-integrated FH (magenta squares) and TH (light blue squares) intensity as a function of the incident THz peak $E$-field ($E_{in}$) at 20 K. The FH intensity is integrated from 0.3 to 0.7 THz, and the TH intensity is integrated from 1.2 to 1.8 THz. The solid lines are the guides to the eye with a slope of 2 (magenta line) and 6 (light blue line). The gray squares show the intensities on the noise floor, which are estimated by integrating the FFT power spectra as in (a) from 2.5 to 3.1 THz. The gray dashed line denotes the fitting curve to the noise-floor intensities with a constant. (c) The same plot as (b), but at 60 K. The solid lines are the guides to the eye with a slope of 2. In the legend, NF denotes the noise floor. (d) Temperature dependence of the third-order nonlinear susceptibility per depth $\chi^{(3)}_{dep}$ (red) and the superfluid density $n_s$ (gray). The error bars for $n_s$ are the fitting errors in the two-fluid model. The black vertical dashed line denotes $T_c$. 
Figure 8 (a) Schematic of the optical pump-THG probe (OP-THG) spectroscopy in reflection geometry. (b) The FFT power spectra of the reflected narrowband THz E-field at 20 K measured at $t_{pp} = 3.2$ ps when the 800-nm pump pulse is “On” (blue curve) and “Off” (gray curve). Here, both the 800-nm pump and 0.5-THz polarizations are parallel to the c-axis of UD61. (c) The 800-nm pump-induced change in the TH intensity at 20 K with the pump fluence of 3.4 mJ/cm$^2$ at selected pump-probe delay times $t_{pp}$. The delay time $t_{pp}$ for each data is shown in the right.
Figure 9 (a) Schematic illustration of the pump-induced SC volume at 100 K compared to that below $T_c$. (b) The upper panel shows the pump-induced change in the FFT power spectrum around TH frequency for UD61 at 100 K (red curve). The green dashed curve is 6.7% of the FFT power spectrum at 40 K without the photoexcitation. Both the 800-nm pump and 0.5-THz polarizations are along the c-axis of UD61. The lower panel shows the expanded plot of the upper panel for the pump-induced change in FFT power spectrum at 100 K. The standard errors of the measurements are displayed by the colored bands. (c) The pump-induced change in the FH intensity ($\Delta I_{FH}$, upper panel) and TH intensity ($\Delta I_{TH}$, lower panel) at 100 K as a function of the pump-probe delay time $t_{pp}$. The FH intensity changes are integrated from 0.3 to 0.7 THz, and the TH intensity change is integrated from 1.2 to 1.8 THz. In the lower panel, $\Delta I_{TH}$ at 100 K is normalized by the integrated TH intensity without photoexcitation at 40 K from 1.2 to 1.8 THz, and 6.7% of the integrated TH intensity at 40 K is shown by the green dashed line. (d) Simulated time evolution of the TH intensity. The inset plots the time evolution of the superfluid density $n_s$ after photoexcitation normalized by its maximum value, which is used in the simulation.
Figure 10 (a) The THz pump-induced reflectivity change $\Delta R_{THz}/R$ for UD78 at selected temperatures. Here, the polarization of the THz pump pulse is along the $a$-axis, and that of the 800-nm probe pulse is along the $b$-axis. The gray curves are the fits to the data using Eq. (3). The upper panel shows the waveform of the squared THz $E$-field $E_{THz}(t)^2$. The two peaks in $E_{THz}(t)^2$ in the time domain are denoted as $t_1$ (= 0.48 ps) and $t_2$ (= 1 ps) (horizontal black dashed lines). (b) The THz peak $E$-field-dependence of $\Delta R_{THz}/R$ at 20 K measured at the delay time of $t_{THz-Pr} = 1$ ps. The dashed line is the guide to the eye with a slope of 2. (c) Temperature dependence of the fitting parameters. The error bars are the fitting errors. The black vertical dashed line denotes $T_c$. 
Figure 11 (a) Relation between the 1.4-μm pump, THz pump, and 800-nm probe pulses in the time domain for the OP-TPOP measurements. Here, we sweep the delay stage of the THz pump pulse while fixing the delay stages of the 1.4-μm pump and 800-nm probe pulse. (b) The THz pump-induced reflectivity change $\Delta R_{THz}/R$ for UD 78 at 20 K as a function of the THz pump-800-nm probe delay time $t_{THz-Pr}$ with the 1.4-μm photoexcitation (red curve denoted by “On”) and without the 1.4-μm photoexcitation (gray curve denoted by “Off”). Here, the 1.4-μm pump fluence is 220 μJ/cm², and the 1.4-μm pump-800-nm probe delay time is fixed at $t_{NIR-Pr} = 0.5$ ps.
Figure 12 (a) The THz pump-induced reflectivity change $\Delta R_{THz}/R$ for UD78 at 100 K as a function of the THz pump-800-nm probe delay time $t_{THz-Pr}$ with 1.4-µm photoexcitation (red curve denoted by “On”) and without 1.4-µm photoexcitation (gray curve denoted by “Off”). Here, the 1.4-µm pump fluence is 560 µJ/cm², and the 1.4-µm pump-800-nm probe delay time is fixed at $t_{NIR-Pr} = 0.4$ ps. (b) Comparison of the integrated THz pump-induced reflectivity change $\Delta R_{THz}/R$ from $t_{THz-Pr} = 0$ to 2.4 ps with the 1.4-µm photoexcitation at 100 K (red diamond) and that without the 1.4-µm photoexcitation as a function of temperature (light blue circles). The gray diamond is the integrated THz pump-induced reflectivity change at 100 K without the 1.4-µm photoexcitation. The black vertical dashed line denotes $T_c$. The error bars are the standard error of the $\Delta R_{THz}/R$ amplitude for $t_{THz-Pr} < 0$.

Figure 13 (a), (b) The magnetic moment of the UD61 (ac-plane) and UD78 (ab-plane) YBCO single crystals, respectively. The black vertical dashed lines denote the determined $T_c$. 
Figure 14 (a), (b) Fitting results of the real and imaginary parts of the optical conductivity along the c-axis of UD61 using the two-fluid model described in Appendix B, respectively. The open circles are the data, and the dashed curves are the fitting curves using Eq. (B1). (c), (d) The fitting parameters of $n_s$ and $\omega_d^2$ as a function of temperature. The error bars are the fitting errors. The black vertical dashed lines denote $T_c$. 
Figure 15 (a), (b) Temperature dependence of the real and imaginary parts of the c-axis refractive index of UD61 in equilibrium. The black vertical dashed lines denote $T_c$. (c), (d) Spatial distribution of the real and imaginary parts of the complex refractive index $n(\omega, z)$ constructed in the heating model simulation.
Figure 16 (a) Schematic illustration of the destroyed layer model presented in Appendix D. (b) The 800-nm pump-induced THz reflectivity change $\Delta R/R$ along the $c$-axis of UD61 at 20 K. The black curves are the fits to $\Delta R/R$ using the destroyed layer model. (c) The destroyed layer length $d_0$ obtained by the destroyed model as a function of the 800-nm pump fluence at 20 K (red circles). The solid red curve is the fit to the data with Eq. (D2). The fluence lower than the determined threshold energy $F_{th}$ is shaded by gray.
Figure 17 The 800-nm pump-induced THz reflectivity change $\Delta R/R$ along the $c$-axis of UD61 at 20 K when the pump polarization is along the $a$-axis. The black curve is the fitting to $\Delta R/R$ using the destroyed layer model.

Figure 18 (a), (b) The real and imaginary parts of the transient optical conductivity along the $c$-axis of UD61 at 100 K with the pump fluence of 3.4 mJ/cm$^2$ measured at $t_{pp} = 0.8$ ps when the 800-nm pump polarization is parallel to the $c$-axis (red) and $a$-axis (green). The standard errors of the measurements are displayed as colored bands.
Figure 19 (a) The pump-induced transient reflectivity along the $c$-axis of UD61 at 20 K with the pump wavelength of 1.5 µm (orange solid curves). Eq in the legend denotes equilibrium. The pump polarization is set parallel to the $c$-axis. (b) The pump-induced transient reflectivity change corresponding to (a). The black curve is the fitting to $\Delta R/R$ using the destroyed layer model. (c) The destroyed layer length $d_0$ obtained by the destroyed model as a function of the 1.5-µm pump fluence at 20 K (orange circles). The solid orange curve is the fit to the data with Eq. (D2). The fluence lower than the determined threshold energy $F_{\text{th}}$ is shaded by gray.
Figure 20 (a), (b) The real and imaginary parts of the transient optical conductivity along the c-axis of UD61 at 100 K with the 1.5-μm pump fluence of 0.75 mJ/cm² measured at $t_{pp} = 0.4$ ps. The standard errors of the measurements are displayed as colored bands. Eq in the legend denotes equilibrium.

Figure 21 (a) Schematic illustration of the THG in reflection geometry to consider the effect of the THz $E$-field screening inside the sample (Appendix G). (b) Temperature dependence of the TH intensity without the leakage for UD61 ($I_{THG}$, magenta curve). The light blue dashed curve is the screening coefficient $B_{THG}$ defined by Eq. (G5) at 0.5 THz as a function of temperature. The black vertical dashed line denotes $T_c$. 