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Enhanced coherent oscillations in the superconducting state of underdoped $YBa_2Cu_3O_{6+x}$ induced via ultrafast terahertz excitation

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We utilize intense, single-cycle terahertz pulses to induce collective excitations in the charge-density-wave-ordered underdoped cuprate $YBa_2Cu_3O_{6+x}$. These excitations manifest themselves as pronounced coherent oscillations of the optical reflectivity in the transient state, accompanied by minimal incoherent quasiparticle relaxation dynamics. The oscillations occur at frequencies consistent with soft phonon energies associated with the charge-density-wave, but vanish above the superconducting transition temperature rather than that at the charge-density-wave transition. These results indicate an intimate relationship of the terahertz excitation with the underlying charge-density-wave and the superconducting condensate itself.

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I. INTRODUCTION

An intriguing possibility in photonics is the use of light to control matter [1], especially in complex materials where multiple phases can possess similar energy scales and are coupled. The hallmark of these phases can be imprinted in single-particle excitations, such as quasiparticles, or in collective modes. A step toward the goal of full functional control is to induce coherent and collective transient states that could cooperatively connect these intertwined phases [2]. One area important for demonstrating the induction of cooperative dynamics is in complex oxides such as cuprates [3], where precise tuning of material functionality, e.g., in metamaterials [4,5] for instance, can be exploited.

A recent highlight in quantum materials research is the discovery of an incommensurate charge-density-wave (CDW) state in bulk underdoped cuprate $YBa_2Cu_3O_{6+x}$ (YBCO) [3,6,7], found to be enhanced when superconductivity (SC) is suppressed by a strong magnetic field [7], indicating that the CDW and SC are competing. In addition, anomalous temperature-dependent softening of a set of phonons, precisely at the CDW wave vector, has also been reported [8], highlighting the role of the lattice to both CDW and SC phases. Moreover, the recent observation of a CDW in the Bi-based family of superconductors [9] clearly indicates that the coexistence of density waves (and in some cases stripe and checkerboard charge modulations) and SC is a ubiquitous phenomenon in the underdoped cuprates. While the precise relationship of the CDW state to high-temperature SC is still under investigation, the cooperative interplay between CDW, SC, and the lattice makes the cuprates a renewed arena for manipulating and controlling complex quantum phases, testing the control of function with light.

Optical pump experiments on CDW-ordered YBCO [10] have demonstrated the excitation of coherent dynamics with a frequency of ∼1.8 THz, a lower frequency than previously observed [11]. However, the coherent excitations are obscured by a large incoherent background that decays on a time scale of a few picoseconds [Fig. 1(c)]. This incoherent background arises from the fact that the photon energy of the optical pulse is of the order of 1.5 eV, creating "hot" electrons well above the Fermi level with energies significantly higher than the natural energy scale of elementary excitations in solids (e.g., superconducting gap, phonons, and magnons, ranging from 10 to 300 meV). As a consequence, a significant amount of incoherent excitations are also generated due to the "hot" electron scattering with many underlying degrees of freedom. This scattering often dominates the dynamical landscape of the relaxation process and obscures coherent collective behavior. In this regard, mid-IR [12] or terahertz [13] radiation can be an ideal pump for time-resolved experiments on these types of materials to minimize these unwanted excitations in the relaxation process and access electron states near the Fermi energy at the nodal point for a d-wave superconducting system [Fig. 1(b)]. Today, the advances in THz technology [14] allow for the generation of intense, single-cycle terahertz pulses, demonstrating the capability of manipulating coherent, elementary excitations in solids, such as magnons [15,16], electromagnons [17], or in the triggering of an insulator-to-metal transition [18]. Here, we use intense THz pulses to excite an underdoped superconducting cuprate [Fig. 1(a)] exhibiting competing SC and CDW ground

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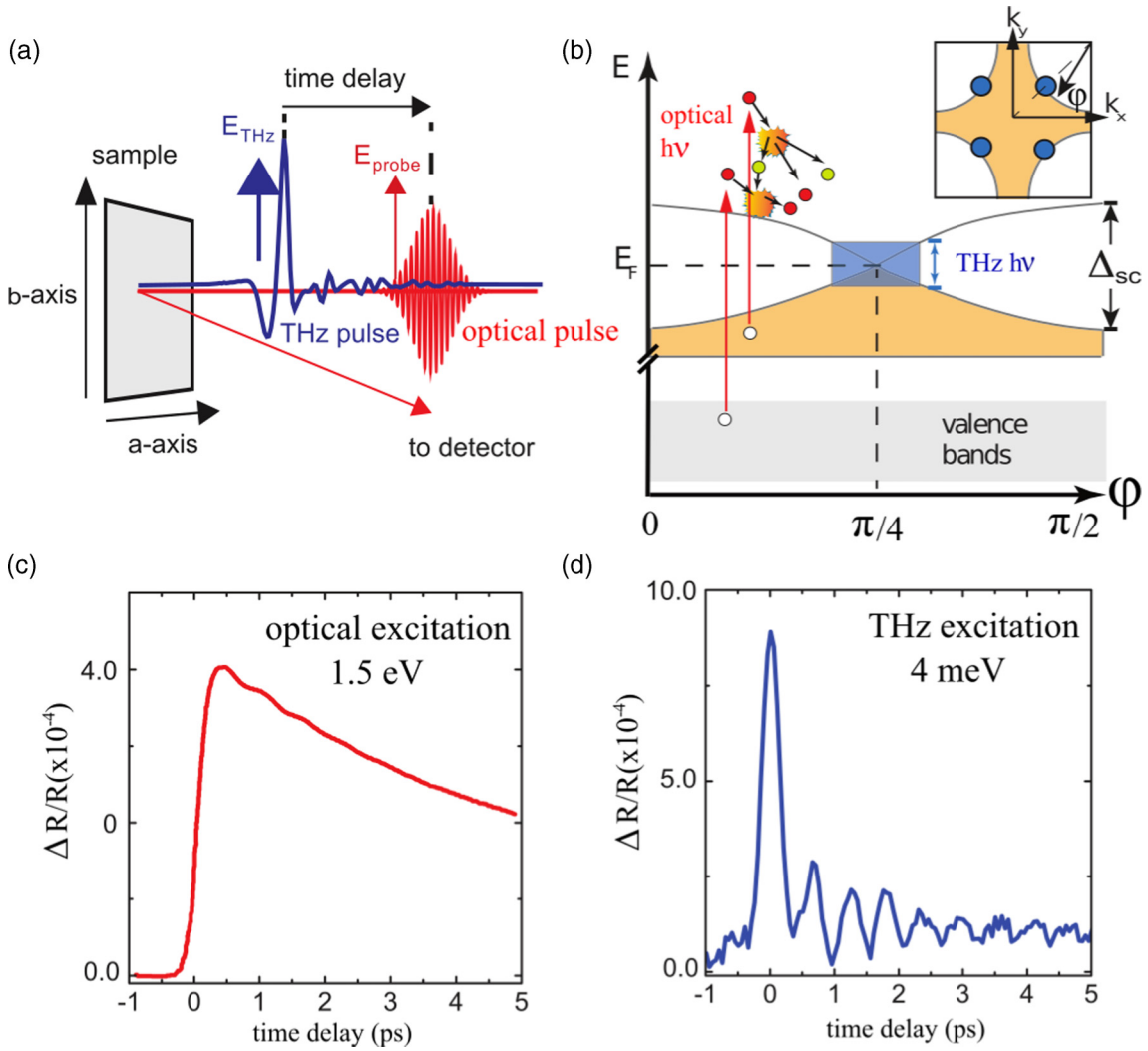

FIG. 1. (Color online) (a) An illustration of the experimental setup (not drawn to scale). (b) A cartoon of the band structure of a d-wave superconductor exhibited in YBCO. An 800 nm pump pulse, with a photon energy of ~1.5 eV, excites a large number of "hot" electrons well above the Fermi energy and can in principle excite quasiparticles over the entire Brillouin zone. A THz frequency pump pulse, which has a much lower energy (~4 meV) and which is on the same scale as the electron states near the nodal point ($\varphi = \pi/4$), can couple to these near-node states directly. (c) The time evolution of the 800 nm reflectivity change after an optical excitation of 800 nm at 15 K (adapted from Hinton *et al.* [10] with permission). (d) The time trace of the 800 nm reflectivity change due to a THz excitation at 10 K. The optical excitation [(c), red curve] used a fluence of ~1.5 μJ/cm^2 and results in a large number of photoinduced quasiparticles as seen as a large background in the figure. The THz data [(d), blue curve] uses a fluence of ~44 μJ/cm^2 and shows the large oscillation with minimal quasiparticle generation.

states and demonstrate that the low photon energy fields are able to induce pronounced coherent, collective excitations in the superconducting state [Fig. 1(d)].

II. EXPERIMENT

The sample [19] was single-crystal $YBa_2Cu_3O_{6+x}$ with ortho-III oxygen ordering [20], with $x = 0.75$. The critical temperature (T_c) for the superconducting transition is 75.2 K and the doping level is $p = 0.133$. The high-quality detwinned single crystals of YBCO are platelets with the broad face perpendicular to the c axis, and were measured with the polarization along either the a or b crystallographic directions. The CDW transition temperature T_{CDW} was found to occur at ~150 K using resonant x-ray scattering [20]. Strong, single-cycle THz pulses were generated from a $LiNbO_3$ crystal via optical rectification of 800 nm femtosecond laser pulses using the tilted-phase front technique [21]. The resulting THz pulses had energies up to 3 μJ with a central wavelength of about 0.6 THz, but carried significant spectral intensity up to ~3 THz [Figs. 2(e) and 2(f)]. The full width at half maximum (FWHM) pulse duration is about 500 fs. Peak THz electric fields at the sample plane were measured to be ~400 kV/cm using electro-optical sampling in a 100 μm thick (110) GaP crystal at the sample location. As sketched in Fig. 1(a), the pump pulse was followed by a weak, 800 nm wavelength probe beam of 100 fs pulse duration to detect the change of the reflectivity, known to be sensitive to transient variations of the dielectric function. Both pulses were incident ~20° from normal to the surface of the sample, which was mounted inside a He cryostat, allowing the ability to vary the temperature in the range the 7–300 K. The reflected optical beam was detected

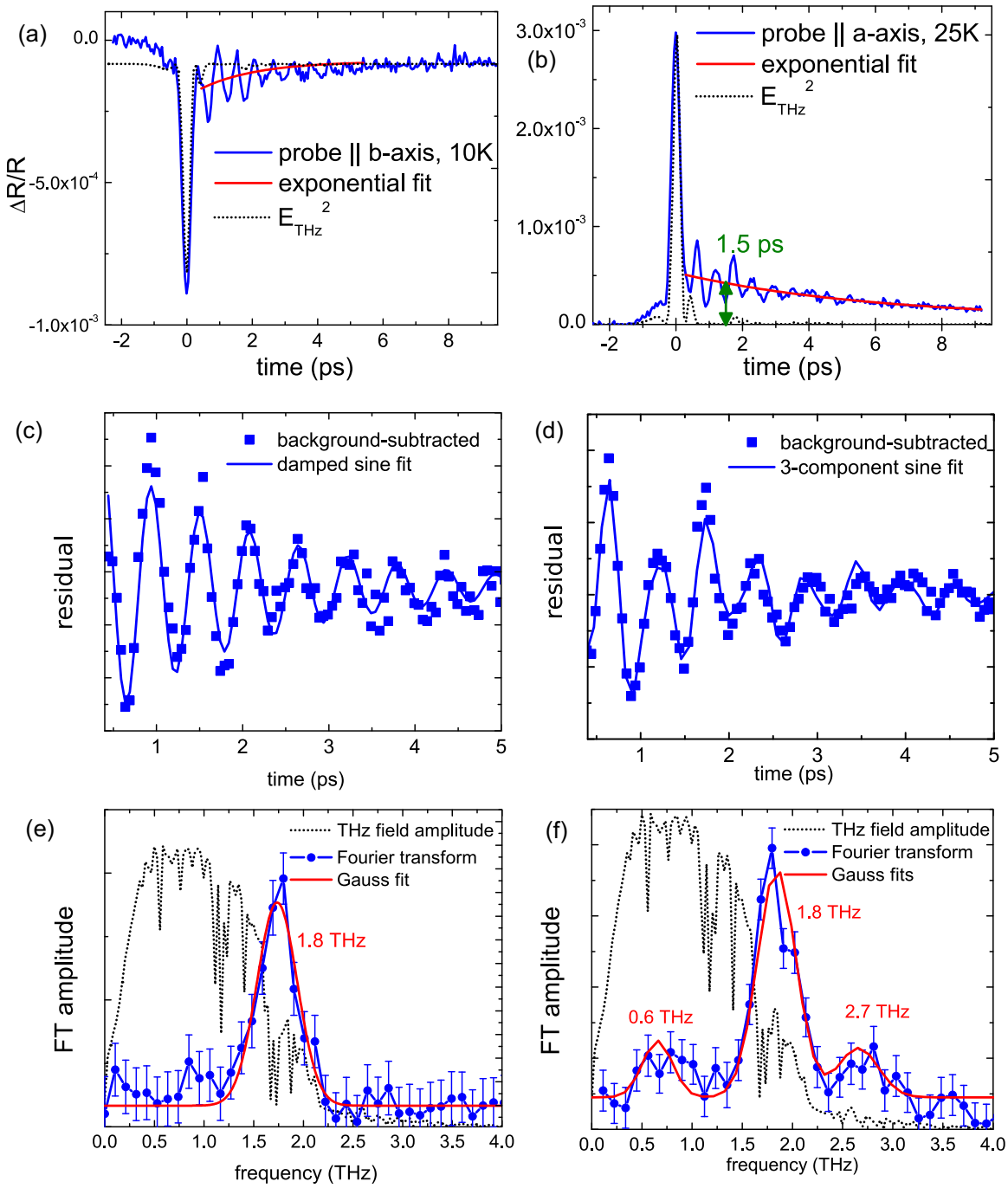

FIG. 2. (Color online) THz-induced reflectivity change with the probe polarization (a) parallel to the YBCO *b* axis at 10 K and (b) parallel to the *a* axis at 25 K. The black dotted lines show the square of the THz field for comparison. The THz *E* field is parallel to the *b* axis in both cases. (c), (d) Oscillatory contribution to the signal after subtracting an exponential fit to the data, shown as a red curve in (a) and (b), respectively. The solid lines are damped sine fits with (c) one frequency and (d) three frequency components, for the two different polarizations. (e) and (f) show the Fourier transforms of the background subtracted oscillations in (c) and (d), respectively. The error bars in the Fourier transform were obtained by error propagation of the time domain uncertainty into the frequency domain. The dotted line shows the Fourier amplitude spectrum of the THz pump pulse in both geometries. The red curve in (f) is the result of a fit to three separate Gaussian functions to each of the three modes, displaying the central frequency of each.

on a silicon photodiode and the measurement was performed using a standard lock-in technique. Both the THz and the probe pulse were linearly polarized. Measurements were carried out for the four possible combinations of both polarizations along either the *a* or *b* directions.

III. RESULTS AND DISCUSSION

As indicated in Figs. 2(a) and 2(b), the THz-induced transient reflectivity at low temperatures closely tracks the THz pulse intensity profile initially [dotted line in Figs. 2(a)

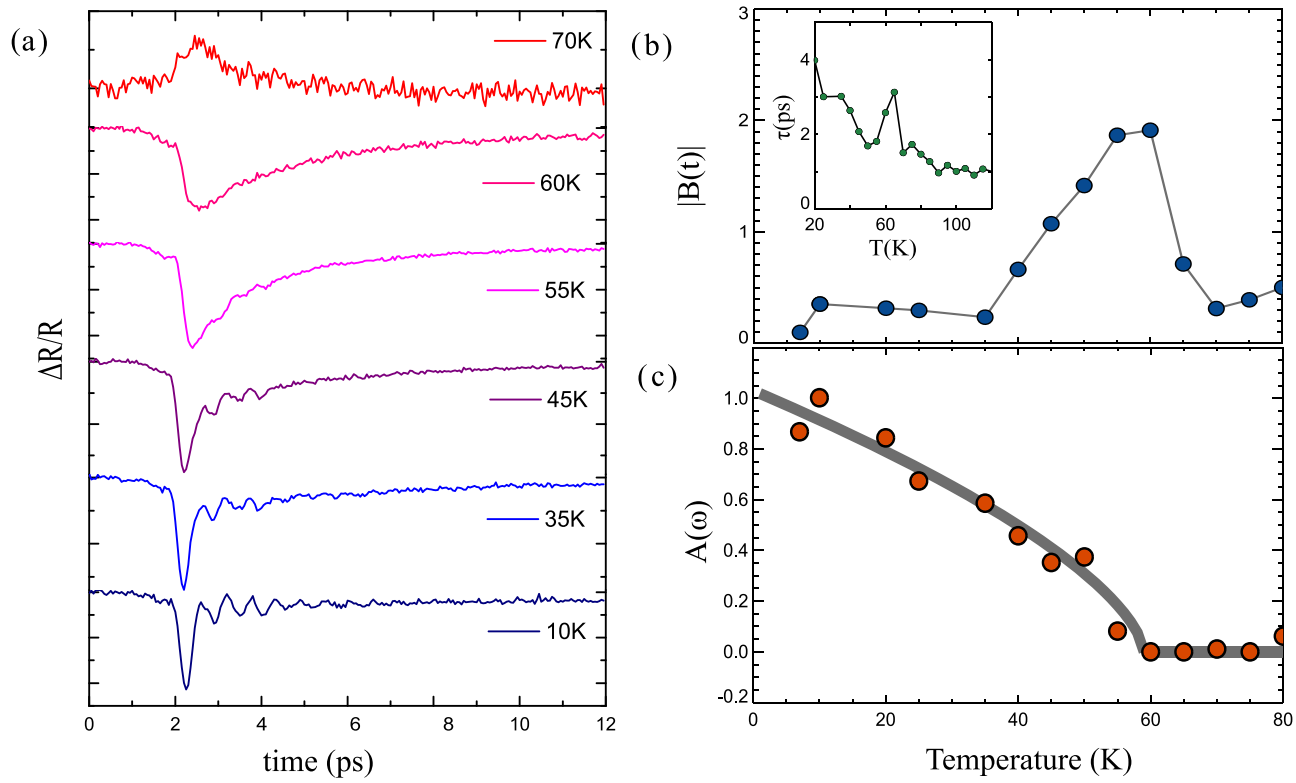

FIG. 3. (Color online) (a) Temperature-dependent traces of the THz-induced reflectivity change $\Delta R/R$ with probe polarization parallel to the b axis. The evidence of the increase of the relaxation time near T_c, as well as the oscillations below T_c, are both clearly evident in the raw data. (b) Incoherent background $|B(t)|$ plotted with temperature for $t = 1.5$ ps, representing the photoinduced quasiparticle number. Inset: The single-exponential relaxation time $\tau(T)$. A similar peak observed in the plot of the relaxation time on approaching T_c was previously observed in YBCO [27] and has been attributed to the "phonon bottleneck" effect. (c) The amplitude of the Fourier mode at 1.8 THz as a function of temperature. The amplitude was calculated by fitting a damped harmonic oscillator response function to the Fourier transform of the oscillatory signal after normalizing by the reflectivity.

and 2(b)], and is then followed by a pronounced, coherent oscillation with almost negligible incoherent background [Fig. 1(d)], in stark contrast to the optically induced dynamics [Fig. 1(c)]. In general, the sign of the reflectivity signal changes if the E field of the 800 nm laser is parallel to the a or b axis of the YBCO crystal. The polarization of the THz pulse did not affect the sign of the pump-probe signal.

The frequency of these oscillations can be extracted by performing Fourier analysis on the time trace of the reflectivity change after subtracting a single-exponential fit to the data, the residual of which is shown in Figs. 2(c) and 2(d). When the polarization of the probe beam is along the b axis, one single mode of 1.8 THz is seen [Fig. 2(e)]. This frequency is consistent with that of the coherent oscillations induced by an 800 nm optical pump pulse, suggesting a similar origin. When tuning the polarization of the probe beam parallel with the a axis, three modes can be resolved: a dominant 1.8 THz mode and two additional modes at 0.6 and 2.7 THz [Fig. 2(f)]. Notably, the 1.8 and 2.7 THz modes match the energies of the strongly renormalized phonon modes exactly at the wave vector corresponding to the CDW modulation [8], measured by inelastic x-ray scattering. This indicates that the coherent oscillations measured via transient reflectivity are due to the presence of the CDW state. While these softened modes are located at a finite momentum, the expected Brillouin zone folding due to the CDW breaks translational symmetry, making these modes optically accessible at the zone center. The 0.6 THz mode and its origin require further investigations. Because it appears while pumping in the a-axis direction, it is plausible that it is associated with the chain ordering or with some underlying electronic C_2 symmetry or nematicity of the CuO_2 planes [22–24]. Only one mode has been reported in time-resolved optical pump experiments in YBCO [10] and $La_{(2-x)}Sr_xCuO_4$ [25].

The THz-induced dynamics [26] are strongly temperature dependent, as shown in Fig. 3(a). As the temperature is raised, the coherent oscillations weaken and finally become undetectable near the superconductivity transition temperature, rather the CDW transition. The incoherent dynamics, which can be described by a single-exponential recovery time, become dominant. Above T_c, and for probe polarization parallel to the b axis, we observe a sign reversal of the transient reflectivity and a weak, featureless signal persists to about 200 K. These temperature-dependent dynamics are described quantitatively by parametrizing the transient reflectivity by

$$B(t,T) = \Delta R(t,T)/R(T) = C_0 + C(T)e^{-(t/\tau + \varsigma)}, \quad (1)$$

where the amplitude of the incoherent response is the value of this underlying exponential function [shown in Fig. 3(b)],

τ is the decay time of the incoherent component [inset of Fig. 3(b)], C_0 is a constant background, and ς is the phase. In order to highlight the oscillatory component, we take the difference between the data and this exponential fit [Figs. 2(c) and 2(d)], and calculate the Fourier transform $A(\omega)$ shown in Figs. 2(e) and 2(f).

The temperature-dependent dynamics are found to be very different than those induced by 800 nm optical pump pulses [10,25]. First, while the collective oscillations induced by optical excitation persist to temperatures well above T_c, namely, up to T_{CDW}, the THz-induced collective dynamics disappear slightly below T_c [Fig. 3(c)]. Second, unlike optical excitations, neither the frequency nor the phase of the oscillations shifts as a function of temperature. Lastly, the temperature dependence of the incoherent dynamics is also very different. On the one hand, the temperature evolution of the recovery time τ [inset of Fig. 3(b)] displays an increase just below T_c, a behavior ascribed to the presence of a "phonon bottleneck" near the point the SC gap closes and the photoexcited quasiparticles and low-energy phonons are in a state of quasiequilibrium [27]. On the other hand, $|B(t,T)|$ also shows a peak just below T_c [Fig. 3(b)], but *vanishes* at low temperatures, in contrast with the temperature-independent behavior observed in all-optical experiments [25,27]. We note that the presence of the incoherent dynamics must be due to Cooper pair breaking [28]. However, we estimate that pair breaking due to Cooper pair acceleration by the THz electric field is not large enough to completely destroy the condensate. For instance, using a rough approximation [29], the maximum supercurrent density can be calculated as $j_{\max} = (ne^2 E_{\max})/m\omega$. We estimate this supercurrent density to be $\sim 10^{11}$ A/m^2, which is smaller compared to the depairing [30] current density of 3×10^{12} A/m^2 in YBCO. The THz-induced coherent dynamics are unlikely to originate purely from this pair breaking process; rather, they are more likely owing to the field-induced modification of the balance between SC, the CDW, and the lattice [28].

IV. CONCLUSION

We have demonstrated that intense THz pulses can induce strong, coherent oscillations indicative of collective behavior below T_c in underdoped YBCO. The observed response reproduces soft phonon modes reported in YBCO and is markedly different from that induced by above band-gap optical excitation. THz excitation provides a unique mechanism for driving this system out of equilibrium through interacting with the CDW state, but only in the presence of the SC state. The coherent dynamics may be ubiquitous among the underdoped cuprates and reveals a perspective for investigating the interplay between SC and CDW. This work paves the way for future investigations with advanced probe methods, for example, the use of ultrafast resonant x-ray scattering may provide information by directly monitoring the CDW dynamics. These future directions could reveal the interplay between SC and CDW physics, thought to be of importance in all families of cuprates, and can be used to further explore the properties of high-temperature superconductors.

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