



Two distinct kinetic regimes for the relaxation of light-induced superconductivity in $\text{La}_{1.675}\text{Eu}_{0.2}\text{Sr}_{0.125}\text{CuO}_4$

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We address the kinetic competition between charge striped order and superconductivity in $\text{La}_{1.675}\text{Eu}_{0.2}\text{Sr}_{0.125}\text{CuO}_4$. Ultrafast optical excitation is tuned to a midinfrared vibrational resonance that destroys charge order and promptly establishes transient coherent interlayer coupling in this material. This effect is evidenced by the appearance of a longitudinal plasma mode reminiscent of a Josephson plasma resonance. We find that coherent interlayer coupling can be generated up to the charge-order transition $T_{\text{CO}} \approx 80$ K, far above the equilibrium superconducting transition temperature of any single layer cuprate. Two key observations are extracted from the relaxation kinetics of the interlayer coupling. First, the plasma mode relaxes through a collapse of its coherence length and not its density. Second, two distinct kinetic regimes are observed for this relaxation, above and below spin-order transition $T_{\text{SO}} \approx 25$ K. In particular, the temperature-independent relaxation rate observed below T_{SO} is anomalous and suggests coexistence of superconductivity and stripes rather than competition. Both observations support arguments that a low temperature coherent stripe (or pair density wave) phase suppresses c -axis tunneling by disruptive interference rather than by depleting the condensate.

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Stripe order in cuprates is closely associated with the suppression of superconductivity, although the interplay between these orders remains a subject of much debate. In these materials, superconductivity is thought to be supported in two-dimensional CuO_2 planes and made three dimensional by interlayer Josephson tunneling. However, both in-plane and out-of-plane coherence can be strongly affected by ordering of charges and spins, or by lattice deformations. Small perturbations in doping, applied field, or pressure can tune the energy landscape between orders, suppressing or supporting the superconducting state.

This phase competition is especially dramatic in the lanthanum copper oxides, which exhibit “striped” spin and charge-ordered states, typically stabilized by an underlying lattice distortion [1,2]. The charge and spin orders also organize within the CuO_2 planes. Static stripe orders, first discovered in $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$ (LNSCO $_x$) [3], are also found in $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ (LBCO $_x$) [4,5] and $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$ (LESCO $_x$) [6,7] [see phase diagram in Fig. 1(a)]. Charge stripes are associated with the suppression of superconductivity in this family of compounds, with bulk superconductivity completely destroyed at $x = 1/8$ doping [8,9] where the lattice spacing synchronizes with the stripe periodicity.

Recent theoretical [10–13] and experimental [9,14,15] work suggests that this suppression is not a simple competition between charge density wave and superconducting instabilities. Rather, superconductivity may coexist with charge ordering, which imposes a space dependence on the superconducting order parameter phase [11]. This spatial modulation—often referred to as a “pair density wave” state—suppresses the total Josephson tunneling by disruptive interference [12]. According to this view, three-dimensional superconductivity

would only be achieved below the temperature at which the superconducting c -axis coherence length exceeds the quadrupled unit cell spacing [16] [see inset in Fig. 1(a)]. This assessment is supported by susceptibility [9] and resistivity and thermopower [15] measurements that suggest a fluctuating two-dimensional (2D) superconductivity regime survives in the spin-order state, up to $T_{\text{SO}} = 40$ K in LBCO 1/8, and one-dimensional correlations persist up to the charge-order transition, $T_{\text{CO}} = 54$ K.

In this work, we used femtosecond laser excitation to reintroduce c -axis coherent coupling in stripe-ordered $\text{La}_{1.675}\text{Eu}_{0.2}\text{Sr}_{0.125}\text{CuO}_4$ (LESCO 1/8) and examined the relaxation kinetics. LESCO 1/8 single crystals were grown using the traveling solvent floating zone technique. They were characterized by resistivity and magnetization measurements and were found to be nonsuperconducting down to 5 K. [17] The charge- and spin-ordered regimes for this doping are indicated by the dashed vertical line on the phase diagram in Fig. 1(a).

Laser excitation has proven a powerful tool to control lattice [18–21] and electronic [22–25] properties and drive phase transitions. The $15\ \mu\text{m}$ midinfrared (MIR) pump used here, tuned to an in-plane Cu-O mode, has been shown to promptly reduce charge stripe order [26] and reintroduce c -axis coherent coupling [17]. The c -axis optical response below 2.6 THz was interrogated as a function of time delay after photoexcitation. Single-cycle THz pulses were generated via 800 nm excitation of a photoconductive antenna. The time resolution of the experiment was limited by the THz bandwidth to about 300 fs. The experiment was performed in reflection geometry, with the MIR pump at normal incidence and the THz s -polarized along the c axis at a 30° angle of incidence. The reflected equilibrium

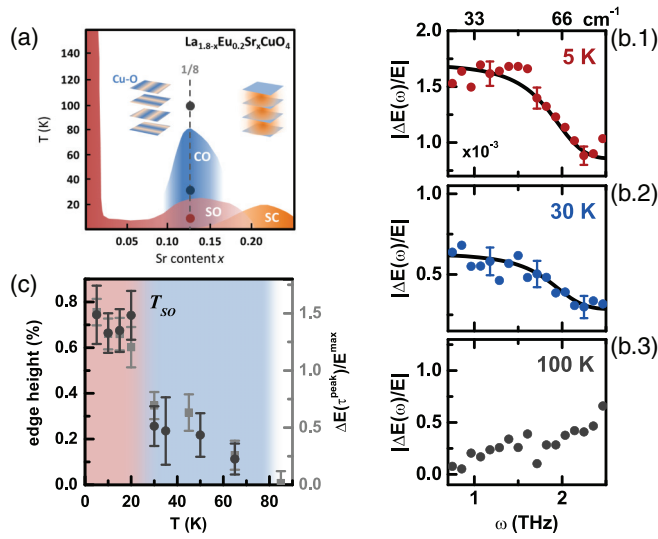


FIG. 1. (Color online) Phase diagram of LESCO and transient response at 5, 30, and 100 K. (a) Phase diagram of LESCO, based on Ref. [28] indicating regions of bulk superconductivity (SC) and static spin (SO) and charge (CO) order. The static stripes suppress c -axis coupling of the CuO_2 planes (inset cartoon, left), with bulk superconductivity restored at dopings in which the stripe order is reduced (inset cartoon, right). (b) The raw transient reflectivity changes measured 1.8 ps after MIR excitation. At 5 K (b.1) and 30 K (b.2), an edge is apparent at 1.5–2 THz. The black lines indicate the reflectivity spectrum (rescaled) due to a longitudinal plasma mode, shown as a guide to the eye. Above the charge-ordered transition temperature T_{CO} no edge is observed [shown at 100 K; (b.3)]. (c) The size of the reflectivity edge remains approximately constant below the spin-order transition temperature T_{SO} , but drops rapidly between T_{SO} and T_{CO} (dark gray circles). This trend is also reflected in the time domain, by the change in the THz amplitude $\Delta E(\tau)/E^{\text{max}}$ measured at the peak of the response (light gray squares). The full delay traces $\Delta E(\tau)/E^{\text{max}}$ are shown in Fig. 4.

THz field, $E(t)$, and the pump-induced changes to the field at time delay τ , $\Delta E(t, \tau)$, were measured via electro-optic sampling to capture the full amplitude and phase response.

The raw change in reflectivity, given by the Fourier transformed quantity $|\Delta \tilde{E}(\omega, \tau)/\tilde{E}(\omega)| \equiv \Delta R/R$, is shown in Fig. 1(b) at $\tau = 1.8$ ps after excitation, which corresponds to the peak amplitude of the transient response. Below the charge-order transition temperature, the transient spectra show the appearance of a reflectivity edge between 1.5 and 2 THz. Figure 1(b.1) shows 5 K base temperature and Fig. 1(b.2) shows 30 K, above T_{SO} . Note that the appearance of this effect has been reported in Ref. [17] up to 10–20 K. Improvements in the experimental apparatus and a fourfold increase in the excitation fluence (to ~ 4 mJ/cm²) make it possible here to detect a qualitatively similar but smaller edge far above T_{SO} , up to between 70 and 80 K [dark gray dots, Fig. 1(c)]. Above T_{CO} , the THz response shows no reflectivity edge [Fig. 2(b.3) at 100 K].

The complex optical properties of the photoinduced state were calculated from the THz response and equilibrium properties measured by Fourier transform infrared (FTIR) spectroscopy. The THz probe samples a crystal volume on order 100 times greater than the 15 μm pump. To isolate the optical response of the photoexcited volume alone we model the system as a single excited layer on a bulk volume that remains in equilibrium [27] (see Supplemental Material [28]).

Figure 2 shows the optical response of LESCO 1/8 at two temperatures, below and above the spin-order transition $T_{SO} \approx 25$ K. [6,29] In both regimes, the transient response (dots) is characterized by the appearance of a longitudinal plasma mode at ~ 1 THz. This mode is most clearly discerned in the real dielectric response $\epsilon_1(\omega)$, shown in Fig. 2(a), which exhibits a zero crossing near the mode resonance. The Ohmic conductivity $\sigma_1(\omega)$ is only weakly affected by the pump, maintaining an insulatorlike response [Fig. 2(b)], suggesting the pump is not producing significant quasiparticle excitations. The inductive conductivity, $\sigma_2(\omega)$, which is negative in equilibrium (gray line) and approaches zero as

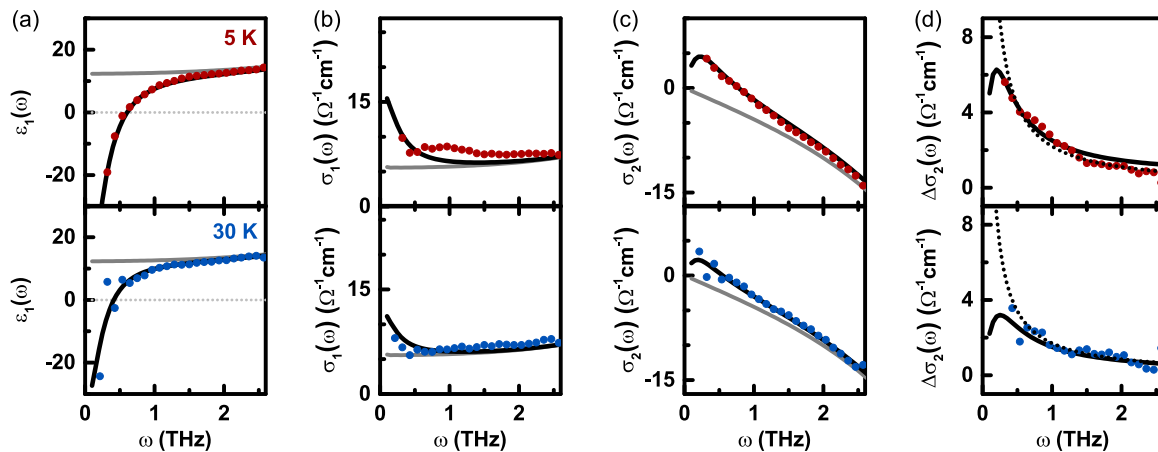


FIG. 2. (Color online) Transient optical properties at 5 and 30 K. Upon MIR excitation, LESCO 1/8 develops a high-mobility state, shown 2.8 ps after excitation at 5 K (red, first row) and at the peak of the response at 30 K (blue, second row). The equilibrium response is shown in gray. The transient state can be fit (black lines) by a longitudinal plasma mode, as described in the main text. (a) The mode is characterized by a zero crossing in the real dielectric function, $\epsilon_1(\omega)$. (b) The Ohmic conductivity $\sigma_1(\omega)$ remains insulating, showing only a small enhancement at lowest frequencies. (c) The inductive conductivity $\sigma_2(\omega)$ becomes positive and diverging towards low frequency. (d) The pump-induced changes $\Delta\sigma_2(\omega) = \sigma_2(\omega) - \sigma_2^{\text{eq}}(\omega)$ diverge as $1/\omega$ (dotted line).

$\omega \rightarrow 0$, instead increases divergently towards low frequency in the photoexcited state [Fig. 2(c)]. The response turns positive below 0.75 THz at 5 K and 0.5 THz at 30 K. The conductivity change, $\Delta\sigma_2(\omega) = \sigma_2(\omega) - \sigma_{2,\text{eq}}(\omega)$, scales as $1/\omega$, as shown by the dotted line in Fig. 2(d).

In equilibrium, LESCO 1/8 has no plasma mode in this frequency range at any temperature. However, the transient mode coincides with the frequency of the Josephson plasma resonance (JPR) of related underdoped superconducting lanthanides [14,30]. A Josephson plasma mode is a generic feature of superconductivity in cuprates which arises due to the tunneling of pairs between CuO_2 planes. Its frequency ω_p is related to the condensate density, compressibility, and the geometric spacing of the planes [31]. Within one family of compounds, the JPR frequency scales with the superfluid density as $\omega_{\text{JPR}}^2 \propto n_{\text{SF}}$, implying that the transient state reported here has a condensate density of roughly half that of near-optimal doped $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ in equilibrium.

Hence, although the appearance of a longitudinal plasma mode by itself does not uniquely prove superconductivity, the quantitative agreement with the optical response of related superconducting compounds makes the assignment of a superconducting state by far the most likely explanation.

The solid black lines in Fig. 2 show a fit to a single longitudinal plasma mode utilizing only two free parameters [32] with the Drude form,

$$\tilde{\epsilon} = \tilde{\epsilon}_{\text{eq}} - \frac{\omega_p^2}{\omega^2 - i\omega\Gamma},$$

where $\tilde{\epsilon}_{\text{eq}}$ is the equilibrium dielectric function. The transient response is best fit by $\omega_p = 2.45$ THz (1.65 THz) at 5 K (30 K) with a scattering rate of $\Gamma \approx 0.25$ THz.

Within this model, the scattering rate term Γ encompasses all transient processes that impact the mobility along the c axis. The scattering rate is extremely low, and cannot be reconciled with the properties of an incoherent plasma, but is consistent with a superconducting state with finite coherence length such as that seen in the equilibrium superconductor $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ near T_c [33]. At resonance, transport occurs across the CuO_2 planes with a velocity $2\omega_p L$, where L is the CuO_2 plane separation. The rate Γ can be related to the coherence length d of the c -axis plasma by $d = 2\omega_p L / \Gamma$.

Note that the reflectivity edge and the zero crossing of $\epsilon_1(\omega)$ do not appear exactly at the plasma resonance, ω_p , but are shifted due to decoherence as well as other intraband contributions to $\epsilon_{1,\text{eq}}$, which can be captured in the THz regime by a single parameter ϵ_{FIR} . For long coherence lengths, $d \rightarrow \infty$, the zero crossing occurs near the screened frequency $\tilde{\omega}_p = \omega_p / \sqrt{\epsilon_{\text{FIR}}}$ and shifts to the red as d decreases. If $\Gamma > \tilde{\omega}_p$, the zero crossing is entirely lifted. For the plasma mode $\tilde{\omega}_p$ reported here, we take $\epsilon_{\text{FIR}} = 30$, a standard value for cuprates [34,35].

Figure 3 shows the evolution of $\epsilon_1(\omega)$ as a function of delay time after photoexcitation at 5 K [Fig. 3(a.1)], 35 K [Fig. 3(a.2)], and 65 K [Fig. 3(a.3)]. From fits to the optical response, we extract the time evolution of the screened plasma mode $\tilde{\omega}_p$ [Fig. 3(b), bottom] and the coherence length d [Fig. 3(b), top]. After the transient state is formed, both quantities initially decay following a 2 ps (1 ps) time scale at 5 K (35 K). At longer time delays, the plasma frequency

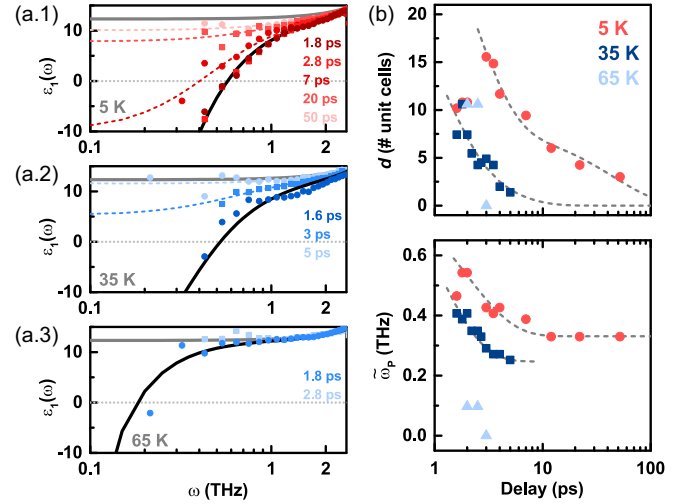


FIG. 3. (Color online) Time dependence of the transient response. (a) The real dielectric function $\epsilon_1(\omega)$ (alternating circle and square dots) as a function of pump-probe delay at 5 K (a.1), 35 K (a.2), and 65 K (a.3). The response is fit with a single longitudinal mode, as described in the main text (solid black and dashed lines). The drop in coherence length causes a flattening in the low frequency $\epsilon_1(\omega)$. (b, top) The c -axis coherence length of the transient plasma as a function of delay for three temperatures: 5 K (circles), 35 K (squares), and 65 K (triangles). The length is expressed in units of the CuO_2 plane spacing. The decay can be fit with a double exponential (dashed lines) with time constants 2 ps (1 ps) and 45 ps (4 ps) at 5 K (35 K). (b, bottom) The plasma frequency ω_p redshifts at early times, decaying to a constant value following single exponential (dashed lines) of 2 ps (1 ps) at 5 K (35 K).

stabilizes to a finite value, indicating that the carrier density does not reduce significantly throughout the relaxation. This observation offers further support that the transient state is not consistent with an anomalously high-mobility quasiparticle excitation, which would relax via a depletion of the carrier density. Rather, the decay of the plasma mode is characterized by a dramatic decrease in the correlation length from ~ 15 unit cells (10 nm) to zero.

The highest temperature at which the longitudinal mode could be seen in $\epsilon_1(\omega)$ in our frequency window was 65 K. The low-frequency cutoff is limited by the sample size and by day-to-day alignment to between 0.2 and 0.4 GHz. From the optical response, and the amplitude of the reflectivity changes $\Delta E(\tau)/E^{\text{max}}$, we extrapolate that the maximum base temperature at which the transient state can be induced is between 70 and 80 K, which is approximately the charge-ordering temperature T_{CO} .

The relaxation time scales are also captured by the changes in reflectivity measured at the peak of the THz field, $\Delta E(\tau)/E^{\text{max}}$, which could be measured with finer delay steps. Figure 4(a) shows $\Delta E(\tau)/E^{\text{max}}$ as a function of pump-probe delay τ . Two lifetimes could be extracted, shown in blue in Fig. 4(b). The lifetimes measured from the coherence length decay are shown in red for comparison. In both the spin-ordered and charge-ordered regimes, the time scales of the double exponential decay are commensurate with the decay of the coherence length. Both lifetimes exhibit two distinct

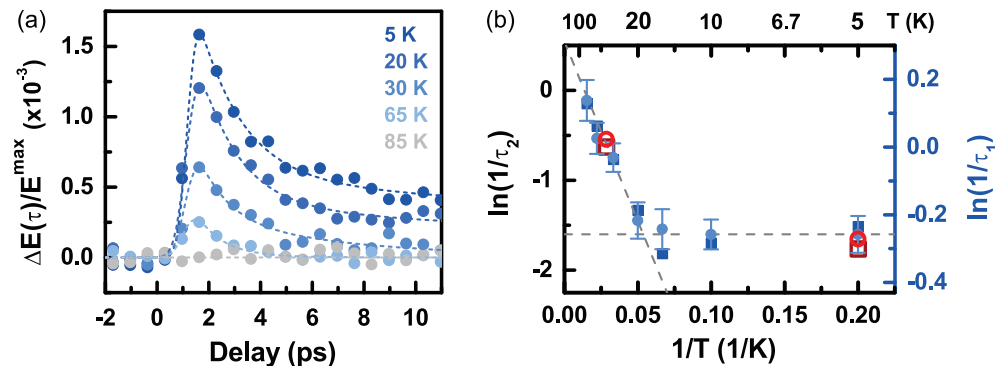


FIG. 4. (Color online) Temperature dependence of the transient lifetime. (a) The time profile of the transient state as a function of pump-probe delay. The vertical axis indicates the transient change of the peak of the THz probe field. For clarity, each point shown represents an average of ten delay measurements. The lifetime of the transient state was extracted using a double exponential fit (dashed lines), with each exponential given roughly equal weight, $50\% \pm 10\%$. (b) An Arrhenius plot of the relaxation rate as a function of temperature. The short lifetime τ_1 (dark blue squares) and the long lifetime τ_2 (light blue circles) track the decay of the coherence length of the transient plasma. [Lifetimes used in the fits shown in Fig. 3(b) are plotted in red.] The lifetime of the transient state remains temperature independent (horizontal gray dashed line) below the spin-order transition temperature, $T_{SO} \approx 25$ K. Above this transition, the lifetime exhibits an exponential temperature dependence, with an energy scale of 4 meV for τ_2 and 0.8 meV for τ_1 (gray dashed line).

regimes. Below T_{SO} , the lifetimes remain temperature independent. Above T_{SO} , where only static charge order remains, the lifetime drops exponentially with base temperature.

The exponential dependence of the relaxation between $T_{SO} < T < T_{CO}$ can be reconciled with the expected kinetic behavior for a transition between two distinct thermodynamic phases separated by a free energy barrier. This is quantitatively captured by the slope of the logarithmic plot in Fig. 4(b), which reflects an activated relaxation of the type $\exp(-E_{\text{barrier}}/k_B T)$. From a double exponential relaxation with lifetimes τ_1 and τ_2 , we extract an energy scale $E_{\text{barrier}} \sim 40$ K (4 meV) and 9 K (0.8 meV), respectively. Interestingly, this energetic regime corresponds to the energy scale of spin fluctuations measured in LSCO 1/8 [36–38], suggesting that the transition between the two phases may be regulated by spin rearrangements. This is consistent with the observation that strong spin fluctuations develop in equilibrium above T_{SO} and survive up to T_{CO} [39,40].

The departure from activated behavior for $T < T_{SO}$ may therefore be related to the freezing out of spin fluctuations. A temperature-independent relaxation rate would be compatible with quantum coherent tunneling between two states, for instance between superconducting and pair density wave phases at constant carrier density. In this picture, Cooper pairing would be superimposed or intertwined [41–43] with the stripe phase, where the dynamical destruction of stripes allows a finite Josephson current rather than driving pairing directly. Indeed, as discussed in Ref. [17], the prompt time scale of the appearance of the longitudinal mode renders it unlikely that the optical excitation is causing pair formation

[44] but rather suggests that pairing in the planes persists in equilibrium.

We have reported the generation of a transient high-mobility state in LESCO 1/8 that survives up to the charge-order transition. This transient state is characterized by the appearance of a longitudinal plasma mode in the low-frequency THz response, which we attribute to *c*-axis superconducting coupling. A first striking observation is that the transient plasma mode can be induced all the way up to the charge-order transition temperature $T_{CO} \sim 80$ K, a large temperature for this family of cuprates. This observation further substantiates theoretical ideas that charge-ordered cuprates remain frustrated superconductors above their equilibrium transition temperature, and that pairing persists even above the spin-ordering temperature T_{SO} . We find that two distinct regimes of phase competition exist between stripe order and transient superconductivity. The lifetime of the photoinduced state remains essentially independent of base temperature below T_{SO} and follows an Arrhenius-like behavior in the charge-ordered regime, up to T_{CO} . Our measurements support the growing body of evidence for 2D superconducting pairing in the charge-ordered state of cuprates.

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