Magnetic-Field Tuning of Light-Induced Superconductivity in Striped $La_{2-x}Ba_xCuO_4$

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Optical excitation of stripe-ordered $La_{2-x}Ba_xCuO_4$ has been shown to transiently enhance superconducting tunneling between the CuO_2 planes. This effect was revealed by a blueshift, or by the appearance of a Josephson plasma resonance in the terahertz-frequency optical properties. Here, we show that this photoinduced state can be strengthened by the application of high external magnetic fields oriented along the c axis. For a 7 T field, we observe up to a tenfold enhancement in the transient interlayer phase correlation length, accompanied by a twofold increase in the relaxation time of the photoinduced state. These observations are highly surprising, since static magnetic fields suppress interlayer Josephson tunneling and stabilize stripe order at equilibrium. We interpret our data as an indication that optically enhanced interlayer coupling in $La_{2-x}Ba_xCuO_4$ does not originate from a simple optical melting of stripes, as previously hypothesized. Rather, we speculate that the photoinduced state may emerge from activated tunneling between optically excited stripes in adjacent planes.

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Charge and spin ordered phases are found throughout wide regions of the phase diagrams of high-T_C cuprates [1–7]. These orders tend to compete with the superconducting state, reducing its coherence. A well-studied case is that of single-layer compounds, in which the doped holes arrange themselves in one-dimensional charge stripes separated by regions of oppositely phased antiferromagnetic order in the CuO_2 planes. In $La_{2-x}Ba_xCuO_4$ (LBCO), for example, stripes completely suppress superconductivity at $x = 1/8$ doping [8,9], and coexist with it at lower and higher doping values [see phase diagram in Fig. 1(a)].

Recent experimental evidence [20,21] suggests that the individual striped planes may be made up of a spatially modulated superfluid, a so-called "pair-density-wave" (PDW), in which the interlayer Josephson tunneling is frustrated by symmetry [see 90° stacking in inset of Fig. 1(a)] [22–24].

Pressure and magnetic fields have been used to tune the striped state in LBCO and in related compounds. In particular, hydrostatic pressures of few GPa were shown to increase T_C [25] and to partially suppress charge order [26]. On the other hand, magnetic fields $H \lesssim 10$ T were shown to amplify the effect of dynamical layer decoupling, leading to a reduction of interlayer tunneling [27,28] and the stabilization of charge and spin order [29,30].

More recently, optical excitation with femtosecond laser pulses has emerged as a means to drive the interplay between stripes and superconductivity [31], enhancing one or the other transiently. Excitation of either the in-plane Cu-O stretching mode in nonsuperconducting $La_{1.675}Eu_{0.2}Sr_{0.125}CuO_4$ ($LESCO_{1/8}$) [32,33], or of $La_{2-x}Ba_xCuO_4$ with high-energy (1.5 eV) optical pulses [34,35], were both shown to enhance interlayer tunneling [36].

In these photoinduced superconductivity experiments, the enhancement was achieved at or near 1/8 doping levels, and it was generally interpreted as a consequence of the removal of frustration by ultrafast melting of stripes [37,38].

Here, we study this effect by tuning the relative strength of equilibrium interlayer tunneling and stripes before photoexcitation. This is investigated for different doping levels, as a function of temperature, and in the presence of external magnetic fields up to 7 T. The experiments yield highly surprising results and identify a correlation between the strength of the equilibrium stripe phase (before it is optically melted) and the photoinduced superconducting state.

The $La_{2-x}Ba_xCuO_4$ single crystals used in our experiments were grown at two nominal Ba concentrations $x = 9.5\%$ and 11.5% [9]. Both of these compounds are superconducting, with transitions at $T_C \simeq 32$ and 13 K, respectively. At 9.5% doping, superconductivity, charge and spin order all appear at the same temperature $T_{CO} \simeq T_{SO} \simeq T_C \simeq 32$ K, while these transitions become decoupled at higher hole concentrations [$T_{CO} \simeq 53$ and $T_{SO} \simeq 40$ K for $x = 11.5\%$, see phase diagram in Fig. 1(a)].

All measurements were carried out in a superconducting magnet with optical access, with magnetic fields up to 7 T applied along the c direction, and for temperatures down to ~5 K [see Fig. 1(b) for experimental geometry].

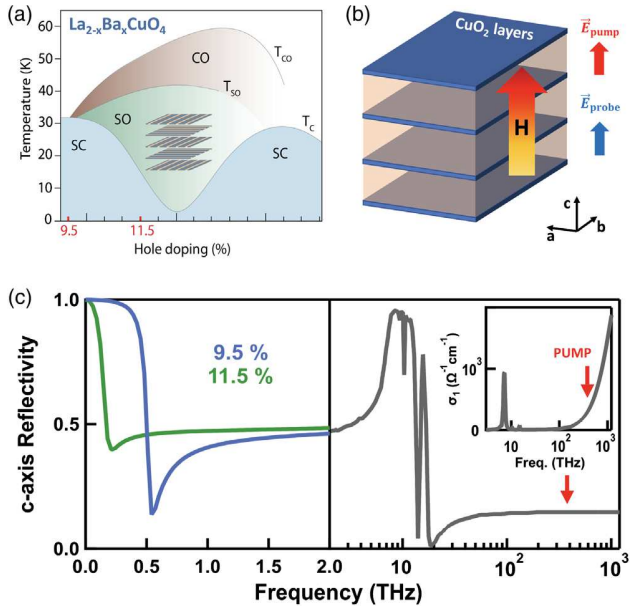

FIG. 1. (a) Temperature-doping phase diagram of $La_{2-x}Ba_xCuO_4$, as determined in Ref. [9]. T_C, T_{SO}, and T_{CO} indicate the superconducting, spin-order, and charge-order transition temperatures, respectively. The inset shows the 90° periodic stacking of CuO_2 planes in the stripe phase. (b) Cartoon depicting the experimental geometry. We used optical pump and THz probe pulses polarized both along the c axis. The experiment was performed in presence of magnetic fields up to 7 T, oriented along the c direction. (c) Equilibrium c-axis optical properties of LBCO. Left panel: THz reflectivity of both samples at $T = 5$ K (no magnetic field), resulting from fits to the experimental data [10]. Right panel and inset: broadband c-axis reflectivity and optical conductivity of LBCO ($x = 9.5\%$) from Ref. [19]. Red arrows indicate the pump photon energy.

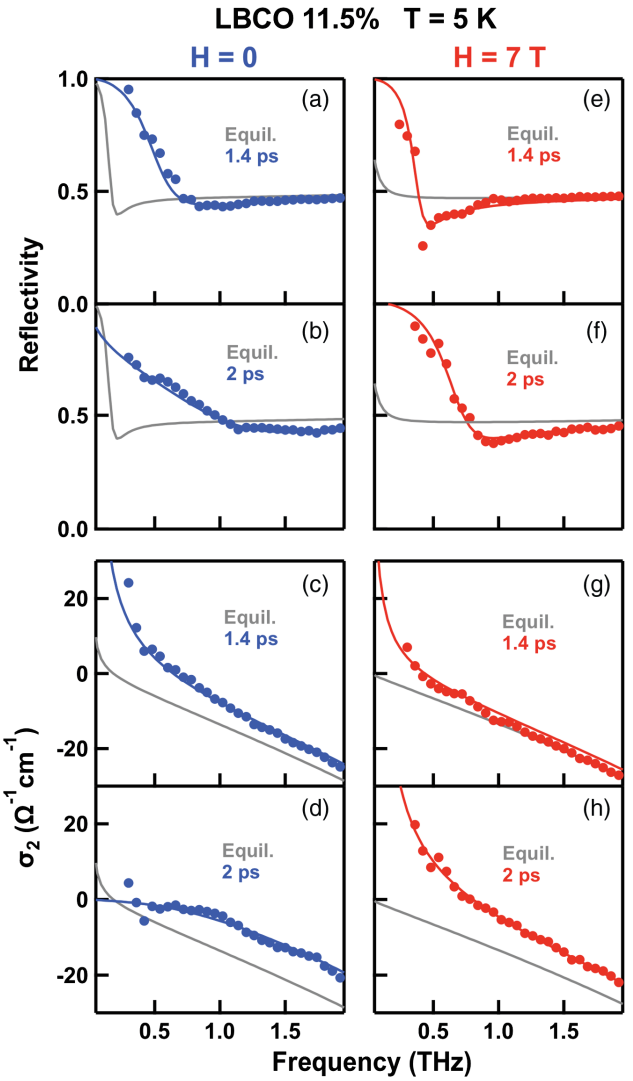

FIG. 2. (a),(b) c-axis THz reflectivity of $La_{1.885}Ba_{0.115}CuO_4$ measured at $T = 5$ K (no magnetic field), at equilibrium (gray lines) and at two pump-probe time delays ($\tau = 1.4$ and $\tau = 2$ ps) after excitation (blue circles). Fits to the transient spectra are shown as blue lines. (e),(f) Same quantities as in (a),(b) measured in presence of a 7 T magnetic field (transient data are shown here in red). (c),(d) Zero-field imaginary conductivity spectra corresponding to the reflectivities shown in (a),(b). (g),(h) In-field imaginary conductivities corresponding to the data of (e),(f).

The equilibrium optical properties [19] at both doping levels were determined using single-cycle THz pulses polarized along the c axis [see Fig. 1(b)], whose electric field profile was measured after reflection from the sample surface at different temperatures, both below and above T_C [10]. The c-axis equilibrium reflectivities extracted with this procedure are shown in the left-hand panel of Fig. 1(c) for $H = 0$ and $T = 5$ K. Both spectra are characterized by a Josephson plasma resonance (JPR), which appears at ~0.5 THz for $x = 9.5\%$ and at ~0.2 THz for $x = 11.5\%$.

In the time-resolved experiments, the LBCO crystals were photoexcited with ~100 fs, 800 nm wavelength laser pulses, also polarized along the c axis [see Fig. 1(b)], at a fluence of ~2 mJ/cm^2. The pump photon energy is indicated by arrows in the spectra of Fig. 1(c). Changes in the real and imaginary c-axis optical properties were retrieved for different time delays after photoexcitation with a temporal resolution of ~350 fs [39], having accounted for the pump-probe penetration depth mismatch [10].

The optical response of the 11.5%-doped compound ($T < T_C$), measured in absence of magnetic field, is displayed in Figs. 2(a)–2(d), both at equilibrium (gray) and at two different pump-probe time delays after photoexcitation (blue circles). As already reported in Refs. [34,35], this material was found to be the most photosusceptible within the LBCO family, displaying optically enhanced (induced) superconductivity all the way up to $T_{SO} \simeq 40$ K.

The $\tau = 1.4$ ps data show a response compatible with a strong optically induced increase in interlayer tunneling strength, evidenced by a blueshift of the reflectivity edge from ~0.2 to ~0.6 THz [see Fig. 2(a)], and, correspondingly,

by an enhancement in the low-frequency imaginary conductivity $\sigma_2(\omega)$ [Fig. 2(c)] [10]. The data shown in Figs. 2(a) and 2(c) could be fitted using a model that describes the optical response of a Josephson plasma, for which the dielectric function is expressed as $\tilde{\varepsilon}(\omega) = \varepsilon_\infty(1 - \omega_J^2/\omega^2)$. Fits to the transient spectra are displayed as blue lines [10].

At longer time delay ($\tau = 2$ ps), the response evolved into that of a conductor with finite momentum relaxation rate, as evidenced by the broadening of the reflectivity edge [Fig. 2(b)] and by a downturn of $\sigma_2(\omega)$ at low frequency [Fig. 2(d)] [10]. Notably, the response functions at this longer time delay were fitted using a conventional Drude model for metals [blue lines in Figs. 2(b) and 2(d)], for which the complex dielectric function is expressed as $\tilde{\varepsilon}(\omega) = \varepsilon_\infty[1 - \omega_P^2/(\omega^2 + i\Gamma\omega)]$. Here, ω_P and Γ are the carrier plasma frequency and momentum relaxation rate, respectively [10].

The same experiment was repeated in a 7 T magnetic field, oriented along the c axis. At equilibrium, the application of the magnetic field induces vortices within the CuO_2 layers, stabilizes charge and spin order [30], and suppresses interlayer Josephson coupling [27] without appreciably affecting superconductivity in the planes (as $H_{c2} \gg 7$ T). The magnetic-field-induced suppression of interlayer superfluid stiffness is evidenced by the redshifted equilibrium JPR, which for zero field is found at ~0.2 THz [gray line in Fig. 2(a)], while in a 7 T magnetic field it is quenched to below the probed frequency range [<0.15 THz, gray line in Fig. 2(e)].

Similar to the data of Fig. 2(a), also in this case a clear edge appeared in the transient reflectivity measured 1.4 ps after photoexcitation [Fig. 2(e), red circles]. However, here the transient plasma resonance remained sharp for longer time delays [Fig. 2(f)], revealing superconductinglike optical properties even at $\tau = 2$ ps. This is also evidenced by the diverging $\sigma_2(\omega \to 0)$ in Fig. 2(h) [10]. Remarkably, unlike the zero-field data in Figs. 2(a)–2(d), the transient optical properties measured at 7 T could be fitted with the Josephson plasma formula at both time delays shown in Figs. 2(e)–2(h).

The coherent character of this newly discovered optically enhanced superconducting state in the presence of high magnetic fields is even more evident in Fig. 3. Here, we show, for different data sets, the dynamical evolution of the interlayer phase correlation length, extracted from the Drude fits of Fig. 2 and defined as $\xi_c = 2\omega_P L/\Gamma$ (here L is the CuO_2 layer separation) [33]. Note that at the earliest time delays, for which the optical properties could also be fitted with a JPR model, ξ_c is at least ~20 unit cells. The upper bound is here determined by the frequency resolution of our measurement and it corresponds to carrier mobilities larger than $\sim 10^4$ cm^2/(V s). Importantly, these are extremely high values which would be unprecedented for out-of-plane transport in a highly resistive normal state oxide, and are rather strongly suggestive of a transient

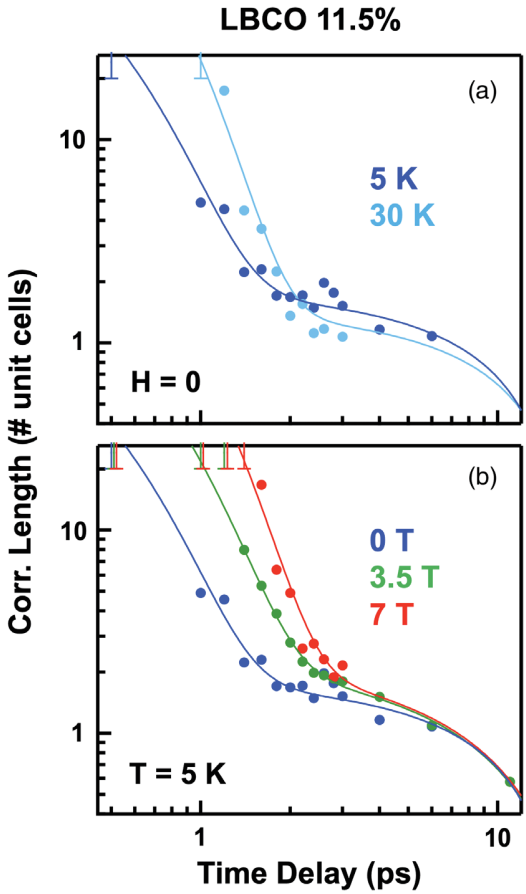

FIG. 3. Dynamical evolution of the interlayer phase correlation length measured in $La_{1.885}Ba_{0.115}CuO_4$ for different temperatures and applied magnetic fields. The values were extracted from the fits of Fig. 2 [10].

superconducting state (see Sec. S4 in the Supplemental Material [10] for an extended discussion on this topic).

At longer time delays, for which the response is well described by the Drude model, ξ_c becomes finite and exhibits a collapse, within a few picoseconds, down to values close to 1–2 unit cells. This collapse became slower and not faster when the temperature was raised to 30 K [Fig. 3(a)], and it was observed to be even slower in presence of a 7 T magnetic field [Fig. 3(b)]. Notably, the application of magnetic field resulted, for certain time delays ($\tau \simeq 1$–2 ps), in a tenfold increase in ξ_c.

A relevant piece of information is added here by the measurements performed at $T_C < T < T_{SO}$ [Fig. 3(a)] [10]. In this case, transient superconductivity emerged from the equilibrium normal state, displaying a larger ξ_c than that found at lower temperatures. This observation suggests that, for $H = 0$, the transient superconductor induced from the normal state is "more coherent" than that induced by exciting the superconducting state at $T < T_C$.

Importantly, all data reported in Fig. 3 display relaxation dynamics to a state with only partially reduced coherence.

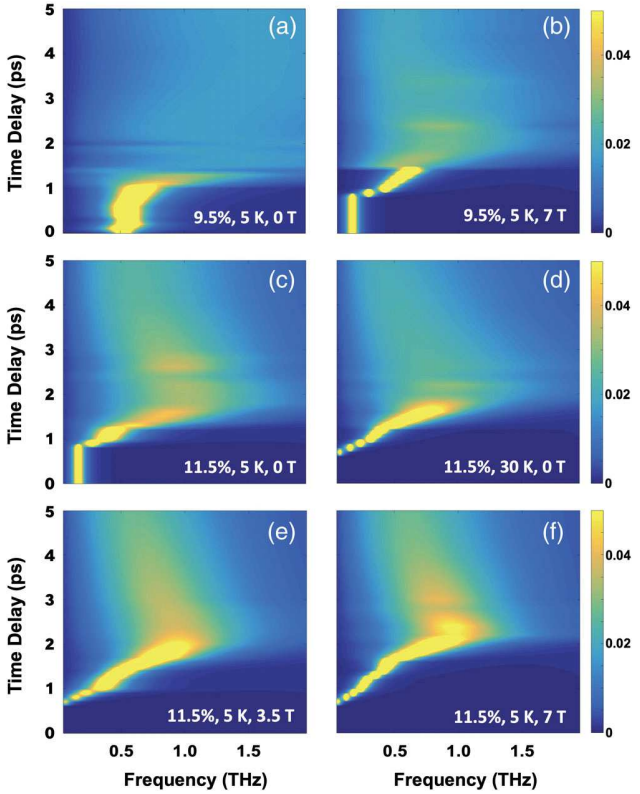

FIG. 4. Dynamical evolution of the energy loss function of photostimulated LBCO under various starting conditions (different doping levels, sample temperatures, and applied magnetic field values), as extracted from fits to the data. Panels (a),(b) refer to $La_{1.905}Ba_{0.095}CuO_4$ at $T = 5$ K in absence (a) and in presence (b) of a 7 T magnetic field. Panels (c)–(f) show instead the response of $La_{1.885}Ba_{0.115}CuO_4$, measured at $T = 5$ K and $H = 0$ (c), $T = 30$ K and $H = 0$ (d), $T = 5$ K and $H = 3.5$ T (e), $T = 5$ K and $H = 7$ T (f) [10].

At $\tau \gtrsim 2$ ps, the data could still be fitted with momentum relaxation rates of $\Gamma \simeq 1$ THz (corresponding to $\xi_c \sim 1$–2 unit cells, see also Ref. [34]), values that are anomalously low for conventional incoherent charge transport, and are instead compatible with a strongly fluctuating superconducting state [40].

The full dynamical response, determined for a wide variety of initial conditions (different doping values, temperatures, external magnetic fields) is shown in Fig. 4. Here, we plot the frequency- and time-delay-dependent energy loss function, $\mathrm{Im}[-1/\tilde{\varepsilon}(\omega,\tau)]$, as extracted from fits to the transient optical response functions. This quantity exhibits a sharp peak at the JPR frequency, which acquires a finite width Γ as soon as momentum relaxation processes set in.

Figure 4(a) displays the response of the 9.5%-doped compound [10] below T_C at zero field, which was already reported in Ref. [34]. This material, for which the equilibrium superconducting phase is robust (see equilibrium JPR at ~0.5 THz) and stripe order is weak [9], only shows marginal superconductivity enhancement for $\tau \lesssim 1$ ps, and then, at later delays, it evolves abruptly into an incoherent state, characterized by an overdamped loss function peak.

When a 7 T magnetic field is applied to the 9.5% compound [Fig. 4(b)], the equilibrium interlayer superfluid stiffness is reduced (the JPR at $\tau \leq 0$ ps is now at ~0.2 THz) and, concomitantly, stripe order is enhanced by ~30% [30]. Here, the optically induced effect was different from that measured in the same material at $H = 0$ and resembled that found at 11.5% doping. Within ~1.5 ps after photoexcitation, a notable blueshift of the equilibrium JPR developed, reaching values close to 0.5 THz [10]. This observation suggests that interlayer Josephson tunneling, which was almost completely quenched by the magnetic field, can be transiently revived also at 9.5% doping by photoexcitation. However, this revival occurs only over a short time interval, and for $\tau \gtrsim 1.5$ ps the system evolves abruptly into an incoherent state.

The middle and lower panels of Fig. 4 [panels (c)–(f)] show the $\mathrm{Im}[-1/\tilde{\varepsilon}(\omega,\tau)]$ function of the 11.5% compound, for which selected optical spectra are displayed in Fig. 2 and the extracted phase correlation lengths are reported in Fig. 3 [10]. Note that this compound is characterized by weaker equilibrium interlayer superfluid stiffness and by a far stronger stripe order than that found at 9.5% doping (by a factor of ~5) [9].

The dynamical evolution of the loss function in this compound displays, for all temperatures and applied magnetic field values, a more coherent character than that found in $La_{1.905}Ba_{0.095}CuO_4$ [Figs. 4(a)–(b)]. In particular, the data taken at T = 5 K and $H = 7$ T [Fig. 4(f)] are those showing the sharpest resonance, with the longest lifetime.

All results above clearly indicate that optically enhanced (induced) superconductivity in $La_{2-x}Ba_xCuO_4$ correlates with the strength of the equilibrium stripe order. Indeed, the stronger the stripes, the longer the photoinduced coherence, as shown by systematically changing the doping level, sample temperature, and external magnetic field. An exception to this trend is found in the temperature dependent response of LBCO 11.5% [Fig. 3(a) and Figs. 4(c)–(d)]. Here, the transient lifetime is clearly enhanced in the 30 K data, although the equilibrium charge order is reduced by ~15% with respect to lower temperatures [9].

The observations above can be interpreted by considering the role of the equilibrium, preexisting c-axis superfluid stiffness in determining the properties of the transient state in the LBCO compounds investigated in our study. Our data suggest that a more robust superconductor at equilibrium adversely affects the strength and lifetime of the photoinduced interlayer coupling [see Fig. 4(a)], while a weak or completely absent pre-existing condensate [Fig. 4(d)–(f)] promotes the transient superconducting state.

We speculate that the equilibrium and optically induced interlayer tunneling might indeed originate from two

separate entities. The former is a spatially homogeneous in-plane condensate, which contributes to the interlayer superfluid stiffness at equilibrium, and whose density is higher for dopings away from 1/8 (as in LBCO 9.5%). The latter, within a PDW picture [23], may be understood as originating from superconducting stripes, whose Josephson tunneling is activated via photoexcitation and whose strength, as for the stripes, is maximum close to 1/8 doping. The equilibrium superconductor and the photoexcited PDW may well compete rather than cooperate with each other, similar to how static charge- and spin order compete with 3D superconductivity at equilibrium.

In the aggregate, the results reported above set new constraints on the overall origin of light-induced superconductivity in single-layer cuprates, which appears to result from the photoexcited striped phase and perhaps is not easily explained as simple optical melting of stripes.

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