

Evidence for two coupled subsystems in the superconducting state of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

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We used a pump-probe technique to measure the transient change of optical reflectivity of both $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, of various dopings, and slightly underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$ thin films. For the $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ films, our data demonstrate the coexistence, in the superconducting state, of two coupled subsystems with different relaxation times and different contributions to the optical reflectivity. One subsystem is associated with the superconducting phase. By contrast, the data from $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$ shows that the coupling between the two subsystems is weak or absent.

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For cuprate superconductors, the nature of both superconducting and normal states continues as a major challenge to condensed-matter physics.¹ Several studies demonstrated that ultrafast optical spectroscopy provides important, though sometime controversial, insights into the properties of two-layer cuprates.^{2–10} In this paper we present femtosecond pump-probe measurements on single-layer $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and double-layer $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$ cuprate films grown on $\text{SrTiO}_3(100)$. Our main results are the following: (1) In the double-layer cuprates, the reflectivity increases during the pump pulse (~ 100 fs), and reaches a maximum in less than 300 fs; relaxation begins *immediately* afterwards.¹¹ In contrast, the decay of the reflectivity signal in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ is delayed by as long as 1.4 ps. We present a model showing that such an unusually delayed relaxation originates from the coexistence of two coupled subsystems with different optical properties and dynamics. (2) The relaxation time for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in the superconducting state is nearly an order of magnitude longer than for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ or $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$. In contrast, well into the normal state $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (all doping levels), $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$ exhibit virtually identical behaviors. It should be noted that previous reports⁶ indicated that the single layer material $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$ —at least for near optimal doping—also exhibits a distinct plateau and a long relaxation response in the superconducting state, both of which are absent well into the normal state.

The samples that we measured were 100 nm thick and were grown with pulsed laser deposition. The details of sample preparation are provided in Ref. 12. We measured five sample types: underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x=0.08$, $T_c=24$ K), optimally doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x=0.15$, $T_c=36$ K), slightly overdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x=0.20$, $T_c=22$ K), underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ ($T_c=80$ K), and underdoped $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$ ($T_c=82$ K). The T_c values for the samples were determined by susceptibility measurements. The T_c values have a 10–90% width of 2 K or less for all samples in this paper. The experimental setup for time-resolved measurements is similar to Fig. 2.4 of Ref. 13. Our

light source was a Ti:sapphire laser at a wavelength of 800 nm and an 80-MHz repetition rate. We determined the pump pulse width to be 100 fs, and the probe pulse width to be 56 fs giving an overall temporal resolution of 120 fs. We determined the pulse widths using second harmonic generation from a b - BaB_2O_4 crystal. For each type of sample, we measured the change in optical reflectivity versus delay time between pump and probe pulses. Our instrumental limit for the smallest observable change in optical reflectivity is 2×10^{-7} , as determined by systematically reducing the laser power on a photodetector by using neutral density filters of known attenuation. The maximum fractional change in the transient optical reflectivity varied from $\sim 3 \times 10^{-5}$ to $\sim 1.2 \times 10^{-4}$, depending on sample and temperature. All data have been repeatedly reproduced. We took great care to insure that the laser power was kept quite low so that we only weakly perturbed the system. We used average laser power of 10 mW, and the illuminated spot on the sample was approximately 60 μm in diameter. This corresponds to an energy during one pulse of 1.2×10^{-10} J and an energy per area during one pulse of 4×10^{-6} J/cm². This is approximately 1.5×10^{13} photons/cm² per pulse. Because we were unable, for technical reasons, to measure the absolute transmissivity, we could not obtain the optical density of the films. Using the approximation that one photon excites ~ 30 carriers in the superconducting state,^{7,8} this would lead to the excitation of $\sim 4.5 \times 10^{14}$ carriers/cm². These numbers are comparable to those in earlier reports on weakly perturbing cuprates.^{7,8}

Figure 1 illustrates the (raw data) changes in reflectivity versus time. The rise (R), plateau (P), and decay (D) times are illustrated by inset I of Fig. 1. We measured data for time delay periods as long as 60 ps as shown by inset II of Fig. 1. Figure 1(a) defines the zero time delay. The rise time is defined as the time during which the differential reflectivity increases from 10% to 90% of the total change. The rise time is followed by a plateau, defined as the period of time during which the differential reflectivity changes by less than 2%. By comparing Fig. 1(a) with the data, [Figs. 1(b)–1(e)], note that the rise time for all data is longer than the instrumental time. Figures 1(b) and 1(c) establish that there is a marked change between the superconducting state and the normal

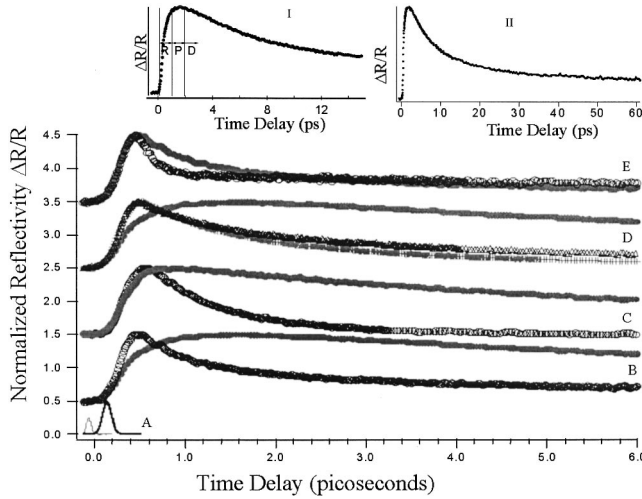


FIG. 1. Change of the reflectivity (normalized to maximum change) vs the time delay. Inset I illustrates the $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ data from (b) out to a delay period of 15 ps defining rise (R), plateau (P) and decay (D) parts of the response. Inset II: same data as inset I for the full delay period of 60-ps delay time. (a) Overlap of probe pulse (smaller, narrower Gaussian) and pump pulse (larger, wider Gaussian), that defines the zero time delay. (b) Underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in the superconducting state (14 K, ●), and normal state (58 K, ○). (c) Slightly overdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in the superconducting state (12 K, ●) and normal state (53 K, ○). (d) Superconducting state spectra of underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (14 K, ●), underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (46 K, Δ), and underdoped $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$ (47 K, +). (e) Normal-state spectra of underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (58 K, ●) and underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (117 K, ○).

state of underdoped and of overdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. There is also a marked change for optimally doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. Specifically, the rise time is much longer, the plateau time is visible, and the relaxation time is much longer in the superconducting state. Figure 1(d) is one of the essential results: it shows that there is a marked difference between $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and either $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ or $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$ in the superconducting state. Note the longer rise time for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. Note as well that $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ exhibits a marked plateau period, while neither two layer cuprate material exhibits a plateau period. Figure 1(e) shows that this difference between one layer and two-layer cuprates virtually disappears well into the normal state.

The quantitative differences between, for instance, underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($T_c/T_{\text{CMAX}} \sim 0.6$) and underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ ($T_c/T_{\text{CMAX}} \sim 0.85$) in the superconducting state is significant. The rise time can be as long as ~ 800 fs for the underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. The plateau period in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ lasts as long as 1.4 ps. In contrast, the plateau time in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$ is absent or too short to observe (less the 50 fs).¹⁴ Furthermore, in the superconducting state, the decay time in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ measured after the end of the plateau is quite long. However, underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$ are quite similar in the normal state ($T/T_c > 2$), as illustrated in Fig. 1(e).

Note that a short rise time is expected in metals due to the

delayed thermalization via the $e^- - e^-$ interaction.¹⁵⁻¹⁷ We also measure a rise time of ~ 300 fs in the normal state for all our samples. However, as can be seen in the inset of Fig. 2(a), underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in the superconducting state actually displays two different regimes of reflectivity rise. The earlier rise regime lasts for ~ 400 fs, followed by a longer rise time of ~ 400 fs. This “kink” indicates that even the initial thermalization is delayed for underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in the superconducting state. The data in Fig. 1 directly establish the two main points of this report: (1) For $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in the superconducting state, the relaxation is delayed by as much as 1.4 ps. (2) The relaxation time for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in the superconducting state is an order of magnitude longer than for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ or $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$.

We analyzed the time dependence of our data using two methods: (a) Fitting one decaying exponential after the end of the plateau period. (b) A two-exponential fit described by Eq. (3), including part of the rise time and all of the plateau period. We used method (a) to analyze our $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ data. Our results are consistent with those reported in Ref. 13. It is important to note that our two exponential method (b) is not the same as the two exponential method used in Ref. 8. We found that method (a) leads to an excellent fit—but only in the normal state—while method (B) was needed to fit the rise and plateau regimes in the superconducting state.

Figure 2 illustrates the results of our data fitting for all samples. Figure 2(a) shows the rise time versus normalized temperature for all doping levels of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, Fig. 2(b) illustrates the plateau time versus normalized temperature, and Fig. 2(c) illustrates the exponential fit versus normalized temperature. We found it necessary to use method (B)—with two exponentials—in the superconducting state, while method (A)—one exponential—provide a satisfactory fit to the normal-state data. The rise time for our $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$ data, consistent with Ref. 9, is 260 ± 20 fs for all temperatures. In our experiments, we excite carriers to the upper Hubbard band. Reference 18 measured BSCCO-2212 materials, and found an electron lifetime in the upper Hubbard band of ~ 50 fs. This indicates that the much longer signal rise time observed here is associated with the thermalization within the lower Hubbard band. We note the recent photoemission report on $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (Ref. 19), which argues that the electronic band structure is quasi-one-dimensional for underdoped samples. We speculate that the long rise time for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in the superconducting state is due to the effects of superconductivity on the lower Hubbard band states. In a sense, this must be so: while $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ exhibit the same $\sim 260 \pm 20$ -fs signal rise time for temperatures well above T_c , they differ substantially below T_c even though the upper Hubbard band is unaffected by whether the material is in the superconducting or normal state.

To interpret the delayed thermalization in the $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ superconducting state, we modeled the relaxation as comprising two interacting subsystems A and B that are out of equilibrium with each other. As the photoexcited carriers thermalize during the first ~ 300 fs (Refs. 15–17)

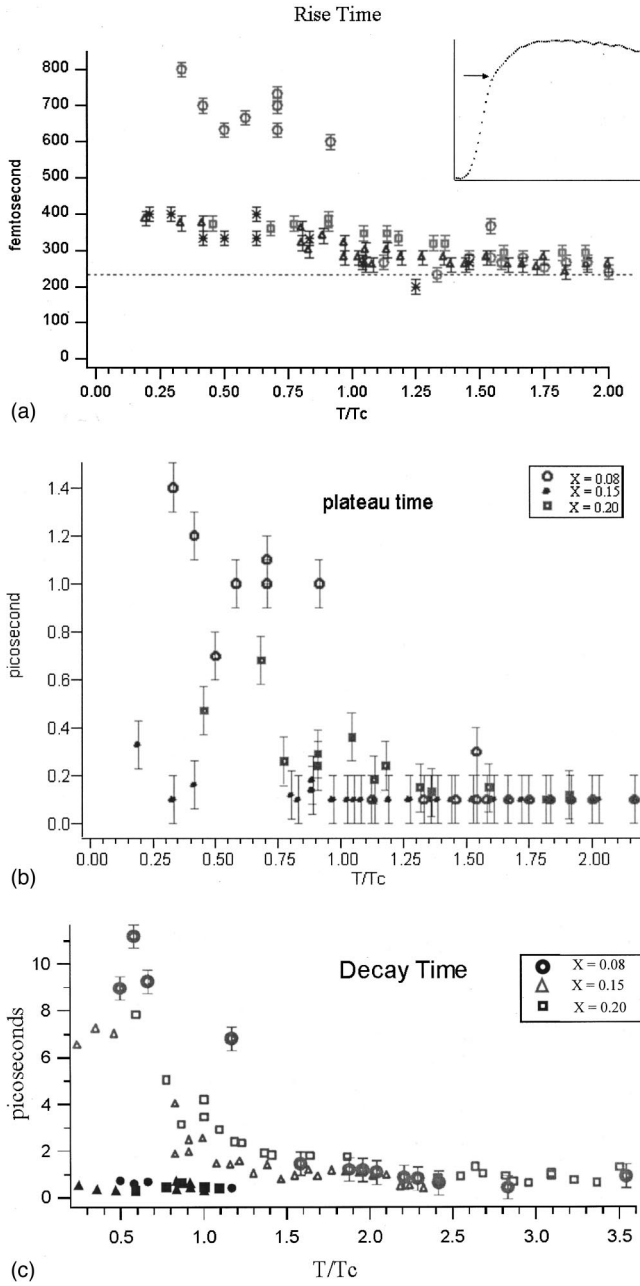


FIG. 2. Rise, plateau, and relaxation times vs normalized temperature for underdoped (○), optimally doped (△), and slightly overdoped (□) $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. (a) rise times. The dashed line represents the rise time of the double layer cuprates. The starred (*) rise time represent an extrapolation of the first slope of the underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ data. Inset: Raw data from underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in the superconducting state. The arrow highlights the change of slope. The first slope is used to determine starred rise times. (b) Plateau time vs temperature for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. There is no plateau for the double-layer cuprates. (c) Time constants obtained using method B in the superconducting state, and method A in the normal state. Open markers: the longer time constant ($1/\Gamma_-$). Closed markers: short time constant ($1/\Gamma_+$). The error bars are the same for all sets of data.

they transfer energy to subsystems A and B . To a first approximation, the reflectivity $R(t) = R[E_A(t), E_B(t)]$ can then be characterized by the time-dependent excitation energies $E_i(t) = \sum \varepsilon n_\varepsilon$, $i=A$ and B , where ε are the energies of the elementary excitations in the system and n_ε the corresponding distribution functions.²⁰ For weak photoexcitation conditions, the reflectivity $R(t) = R[E_A(t), E_B(t)]$ can be expanded in terms of E_i .

By retaining the first term in such a Taylor expansion¹⁷ for the differential reflectivity $\Delta R(t) = R(t) - R(0)$ we obtain

$$\Delta R(t) = \Delta R_A(0) \frac{E_A(t)}{E_A(0)} + \Delta R_B(0) \frac{E_B(t)}{E_B(0)}, \quad (1)$$

where $\Delta R_i(0)$ is the contribution to the differential reflectivity at $t=0$, coming from system (i). The time evolution is

$$\frac{dE_i}{dt} = -\Gamma_i E_i + G_{ij} E_j, \quad (2)$$

where G_{ij} describes the rate at which energy is transferred between systems (i) and (j) due to the interactions between them. In the weak coupling limit $G_{AB}G_{BA} < |\Gamma_A - \Gamma_B|^2$, the solution of Eqs. (1) and (2) has the form

$$\Delta R(t) = \Delta R_0(t) + \left[G_{BA} \Delta R_B(0) \frac{E_A(0)}{E_B(0)} + G_{AB} \Delta R_A(0) \frac{E_B(0)}{E_A(0)} \right] \frac{e^{-\Gamma_- t} - e^{-\Gamma_+ t}}{|\Gamma_A - \Gamma_B|}, \quad (3)$$

where

$$\Gamma_\pm = \frac{1}{2} [\Gamma_A + \Gamma_B \pm \sqrt{(\Gamma_A - \Gamma_B)^2 + 4G_{AB}G_{BA}}]. \quad (4)$$

In the above equations, the contribution $\Delta R_0(t)$, coming from two noninteracting subsystems, decays with two relaxation times $\sim 1/\Gamma_-$ and $1/\Gamma_+$. The second term, on the other hand, which is proportional to the interaction strength, grows in magnitude during a time interval $\sim (1/\Gamma_+)$. It is important to note that even though the magnitude of the second term is suppressed for weak interactions as compared to $\Delta R_0(t)$, it dominates the time dependence during the time scales $\sim (1/\Gamma_+)$, where $(1/\Gamma_+) \sim (1/\Gamma_A) \ll (1/\Gamma_-) \sim (1/\Gamma_B)$, provided that

$$\Delta R_A(0) \ll \frac{G_{BA}}{|\Gamma_A - \Gamma_B|} \frac{E_A(0)}{E_B(0)} \Delta R_B(0). \quad (5)$$

Under the above condition the differential reflectivity will display a rise and plateau regime, determined by the second term in Eq. (3), even for weak interaction between the two subsystems. The above two exponential relaxations describes the experimental data very well with the chi square fit to the data < 0.04 .

In the superconducting state, the decay time for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ is much longer than for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ or $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$ [Fig. 2(c)], which implies that (Γ_-) is small compared to $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ or $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$. This is consistent with the condition that $(1/\Gamma_+) \sim (1/\Gamma_A) \ll (1/\Gamma_-) \sim (1/\Gamma_B)$. In the normal state, the relaxation follows a single

exponential decay, with similar magnitudes for all three cuprate systems since the decay time becomes comparable [Fig. 2(c)].

We stress the significance of Eq. (5). In normal metals, there are two subsystems—the phonon bath and the electron bath. Again, in BCS superconductors, the two-fluid model leads to two subsystems. Neither of these exhibit a delayed rise or plateau period, which is observed if Eq. (5) is satisfied. Physically, Eq. (5) means that the subsystem with the shorter (faster) relaxation time contributes less to the total reflectivity change than the other subsystem. Equation (5) is consistent with Fig. 2(c), in which we find two different relaxation times—differing by approximately one order of magnitude—in the superconducting state. In principle, an alternative explanation for the rise time and plateau time data would be that the electron-electron thermalization time and the electron-phonon relaxation time are close to the same. In a good metal, this is not the case. We note that in the superconducting state, the rise time (<1 psec) and the slower relaxation time (5–10 psec) are significantly different, while the faster relaxation time (~ 1 psec) is only slightly slower than the rise time. There is the possibility, then, that the faster relaxation time might be a combination of thermalization and relaxation, rather than simply relaxation. This possibility warrants further theoretical modeling and experimental investigation.

Let us now briefly speculate as to the origin of the two coexisting subsystems *A* and *B*. First, since we only observe the two exponential relaxations below T_c , one of the two subsystems is in the superconducting phase. Second, our data indicate that the above coexistence occurs the most strongly in the underdoped regime, where the anomalous normal state properties occur. Third, according to Eq. (5), the dielectric function and optical spectrum must depend weakly on the photoinduced distribution function of the elementary excitation of the subsystem with the shorter relaxation time. In fact, for finite temperatures, the coexistence of two phases is expected in the proximity of a quantum critical point that may govern the anomalous normal state properties of the cuprates.²¹ Note that in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, as the doping decreases below a critical value $x_C \sim 0.20$, a rapid collapse of the superfluid density was observed.^{22,23} The optically excited carriers can flip their spin as they scatter and will transfer some of their energy to such low-spin excitations, leading to $E_A(t)$. Since spin excitations do not contribute directly to the optical spectrum ΔR_A should be smaller than ΔR_B , and the condition of Eq. (5) is satisfied. $E_B(t)$ would then be the energy due to the electronic excitations, with a rather long relaxation time $\sim (1/\Gamma_B)$ of the order of 10 ps due to the formation of the superconducting gap and the resulting

bottleneck effect on relaxation. Any reduction of phase space for scattering could lead to a long relaxation time. In this regard, we note the recent photoemission study of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ indicating a quasi-one-dimensional electronic band structure for underdoped samples, just the reduction of phase space that would lead to a long relaxation time.¹⁹ While the reduction of phase space should result in a longer decay time, it seems unlikely that this is the sole cause for the difference in the response of the single compared to the double layer cuprates. Further theoretical studies are required to determine the effects of a quantum phase transition on the relaxation.

A second possibility is to identify subsystem *B* with a pseudogap normal phase that coexists with the superconducting phase *A*, similar to the analysis of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ in Ref. 8. We could then have $\Delta R_A \ll \Delta R_B$ since for underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, the superfluid density is suppressed and the sample is inhomogeneous due to phase separation. The condition of Eq. (5) would then be satisfied if, in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, the relaxation time in the pseudogap phase is much longer than the relaxation time in the superconducting phase. In $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, according to Ref. 8, the opposite is true.

In summary, we reported on the superconducting and normal-state transient optical reflectivity in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, and $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$. We find qualitatively different behavior for the single-layer cuprate compared to the double-layer cuprates in the superconducting state; these differences disappear at temperatures well into the normal state. The data directly establish that $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in the superconducting state consists of two interacting subsystems. A theoretical analysis provides details of the conditions on the interaction between the subsystems consistent with the data. We speculate briefly on two possible explanations as to the physical origins of subsystems *A* and *B*. Relaxation can have a strong effect on the nature of a quantum phase transition, and vice versa, while experiments such as the above are well suited to study such effects. Independent of our speculations, it is noteworthy that—directly from the data—one of the two subsystems is associated with the superconducting state, since the rise and plateau intervals decrease sharply above T_c and are essentially identical to $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ well above T_c .

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