A systematic study of femtosecond quasiparticle relaxation processes in $La_{2-x}Sr_xCuO_4$

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A systematic study of quasiparticle recombination and relaxation dynamics in $La_{2-x}Sr_xCuO_4$ for $x=0.06$, 0.1, 0.115, 0.15, and 0.2 unambiguously shows the presence of two distinct components in the relaxation, one being present only below T_c while the other vanishes at much higher temperatures. The two components are attributed to the intrinsically inhomogeneous ground state in cuprates. Since there are no charge reservoirs in the 2-1-4 system, the presence of two components can only be attributed to inhomogeneity of the CuO_2 planes.

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I. INTRODUCTION

Time-resolved (TR) experiments in cuprate superconductors have shown that valuable information can be obtained about the lifetimes of low-energy electronic states as well as the energy level structure around the Fermi level using femtosecond laser spectroscopy. A combination of modeling and detailed experimental studies on cuprates have consistently revealed the simultaneous presence of multiple relaxation processes, implying the possibility of at least two different species of quasiparticle (QP) states at low temperatures, which could be understood as a signature of an electronically inhomogeneous state.[1–3] Two of these QP relaxation processes appear on the picosecond time scale, while one is slower, with relaxation on the microsecond time scale attributed to the relaxation of intragap localized states.[4] A number of experiments probing the local structure of copper oxide superconductors on short time scales, such as x-ray absorption fine structure (XAFS) and neutron pair-distribution function (PDF) measurements, have suggested that these materials are structurally inhomogeneous.[5–8] These inhomogeneities—particularly if it can be shown that they exist in the CuO_2 planes—may have profound and fundamental consequences for pairing and superconductivity. We should note that there is still substantial controversy regarding the details, such as magnitude and time scale—and indeed relevance—of such inhomogeneities.[9,10] Associated with structural inhomogeneities, one would also expect to observe inhomogeneities in the electronic structure, which may be significantly harder to determine, as it amounts to deconvolving multicomponent inhomogeneous electronic spectra, which cannot be done convincingly without direct measurement of the lifetimes of the states involved, such as can be done with TR femtosecond spectroscopy.

Most TR experiments showing two-component relaxation so far were performed on cuprates with a multiple-layer structure, consisting of CuO_2 planes and transition-metal-oxide charge-reservoir layers or Cu-O chains. Thus, the two-component relaxation observed in time-resolved spectroscopy could not unambiguously be attributed to CuO_2 planes, and naturally it was suggested that the two relaxation components might be attributed to CuO_2 planes and charge-reservoir layers, respectively. From analysis of the systematic QP relaxation measurements in $La_{2-x}Sr_xCuO_4$ (LSCO) as a function of doping x and temperature, we show that in spite of the fact that there are no distinct out-of-plane charge reservoirs, two different QP relaxation processes are observed. One exhibits "pseudogap" behavior, which can be modeled with QP recombination across a T-independent gap Δ_p, while the other is associated with a T-dependent superconducting gap $\Delta_c(T)$. We also show that both Δ_p and $\Delta_c(T)$ vary systematically with doping, in agreement with other measurements in $La_{2-x}Sr_xCuO_4$. We discuss the observed QP relaxation in terms of our current understanding on the low-energy electronic structure with preformed pairs above T_c, and a phase-coherent state with a collective mean-field-like gap below T_c. In both temperature ranges, the QP relaxation proceeds across a large gap, indicating that the nodal QPs are either inaccessible or missing.

II. EXPERIMENTAL DETAILS

The experiments were performed on a series of high-quality $La_{2-x}Sr_xCuO_4$ single crystals grown in an optical furnace[11] with $x=0.06$ ($T_c=6$ K), 0.10 ($T_c=30$ K), 0.115 ($T_c=31$ K), 0.15 ($T_c=39$ K), and 0.2 ($T_c=30$ K). The TR experiments were performed using the standard pump-probe technique, the details of which have been reported elsewhere.[12] The light was incident along the c axis of the crystals, with 50-fs pulses at 800 nm from a Ti:sapphire laser with a repetition rate of 86 MHz as a source for both excitation (pump) and probe pulses. The pump beam was focused on a 100-μm full width half maximum spot on the sample while the probe diameter was <80 μm. The measurements have been performed at a wide range of excitation intensities ranging from 4×10^{-8} J/cm^2 to 2.8×10^{-6} J/cm^2 (corresponding to energies of 1.3–84 pJ per pulse).

The raw data on the time dependence of the photoinduced change in reflectivity $\Delta R/R$ is shown in Fig. 1 for $x=0.1$, 0.115, 0.15, and 0.2. The data are consistent with previous measurements on this compound,[13,14] but were measured over a wider temperature and doping range.

The dynamics are strongly temperature dependent, which is emphasized by the semilogarithmic scale plot in Fig. 1. Above T_c, a fast response is observed, with a rise time that is limited by the experimental resolution of $\approx$70 fs (labeled B in Fig. 1). This signal has a weakly T-dependent amplitude

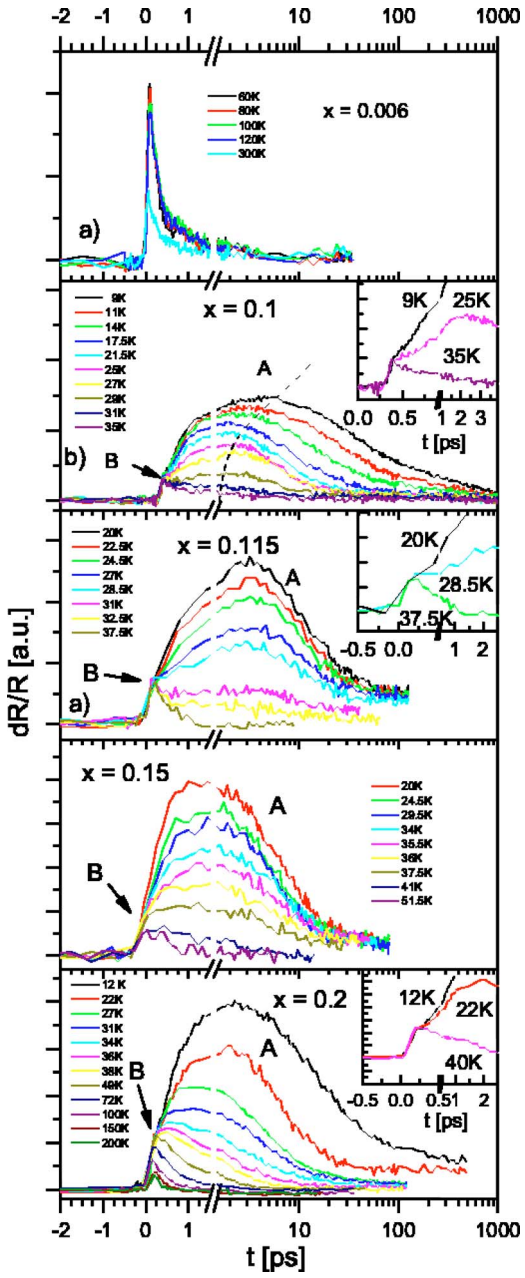

FIG. 1. (Color online) The photoinduced reflection dynamics $\Delta R/R$ in $La_{2-x}Sr_xCuO_4$ single crystals taken with photoexcitation intensities $1.7\times10^{-7}-2\times10^{-6}$ J/cm^2 for different doping x as a function of temperature. Two distinct components (A and B) are clearly resolved (see also insets).

and decay time. Below T_c, a second component becomes evident (A in Fig.1) whose rise time is about 1 ps (which is somewhat temperature dependent, being slightly shorter at higher temperatures). Both the amplitude $\Delta R/R$ and the relaxation time τ_R of component A are strongly T dependent. The difference in rise times of the two components is an important feature of the data, which enables us to distinguish between the two components below T_c. In particular, the insets to Fig. 1 clearly demonstrate that the two components are coexisting down to the lowest temperatures measured. Moreover, it is also clear that the amplitude of component B is roughly constant below T_c, consistent with previous reports on other cuprates.

III. DATA ANALYSIS

In this paper we concentrate on the analysis of the temperature dependence of the amplitude $\Delta R/R$, which gives systematic information on the gap structure as a function of x and temperature.[15] As noted above, above T_c the photoinduced reflectivity $\Delta R/R$ (component B) can be well described by a single relaxation process, while below T_c the photoinduced reflectivity trace $\Delta R/R$ consists of two distinct contributions (A and B) with different rise times [which is clear from the kink in the raw data at $t<0.3$ ps (see insets to Fig. 1)]. The determination of amplitudes of the two components as a function of temperature is straightforward over the entire temperature range, i.e., by simple reading out from the raw data. However, the large difference in magnitudes of the two components (below T_c A$\gg$B) prevents us from extracting the relaxation time τ_B at temperatures below T_c, as was done in the case of $YBa_2Cu_3O_{7-x}$ (YBCO) (Ref. 1) or $YBa_2Cu_4O_8$ (Y124).[3] On the other hand, both components are well fitted by single exponential decay, i.e., both at $T\gg T_c$, where only component B is present, as well as at $T\ll T_c$, where A$\gg$B; the relaxation is well fitted by $\exp(-t/\tau)$. In addition to picosecond relaxation dynamics, some long time scale dynamics ($\tau\gg10$ ns) is also found with similar temperature dependence that has been reported for other cuprates,[4] which will not be discussed in detail here.

Due to poor low-temperature thermal conductivity in cuprates,[16] the heating of the laser illuminated spot in these experiments with high repetition laser systems can be substantial. However, proper accounting of heating effects using different pump intensities shows that the relaxation dynamics in $La_{2-x}Sr_xCuO_4$ are independent of the excitation intensity in the range of intensities used here (spanning nearly two orders in magnitude). In Fig. 2(a) we show the temperature dependence of the relaxation time τ taken at several laser fluences for $x=0.1$. Heating of the probed spot due to laser excitation has been determined using the method given in Ref. 12, showing that the relaxation time is in this range of excitations independent of pump intensity over a wide temperature range. Similarly, the amplitudes of the photoinduced reflectivity signals are linearly proportional to the pump intensity, as expected.[15] Figure 2(b) presents the temperature dependence of amplitudes of components A and B over the temperature range $20<T<300$ K.

A. Temperature dependence of the photoinduced quasiparticle density

The very different rise times (see Fig. 1) of the two relaxation processes allow us to analyze the T dependence of the two components in more detail. The temperature dependences of the two amplitudes (A and B) are shown in Fig. 3 for different doping levels x. Clearly the temperature dependences of the two components are very different. As already mentioned above, component B is evident throughout the entire temperature range, while component A is present only

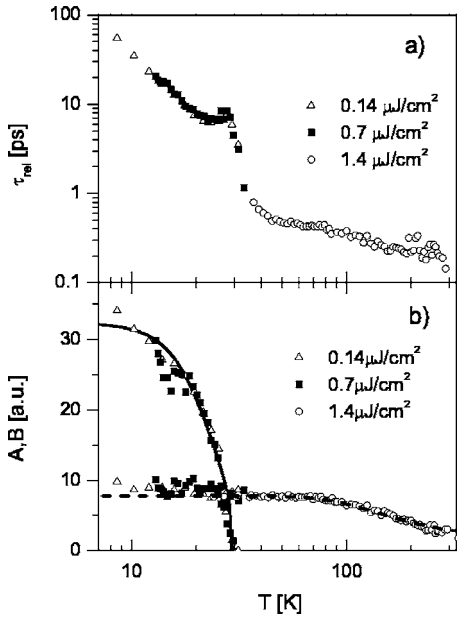

FIG. 2. (a) The temperature dependence of the relaxation time obtained using a single exponential decay fit to the data. (b) The temperature dependence of the amplitude of components A and B normalized to the excitation fluence for $La_{1.9}Sr_{0.1}CuO_4$ taken at several excitation intensities (the fluence of the probe beam was 0.18 $\mu J/cm^2$ in all data sets). The amplitudes of the two transients were directly read out from the raw data. Continuous heating due to laser excitation is accounted for as described in Ref. 12.

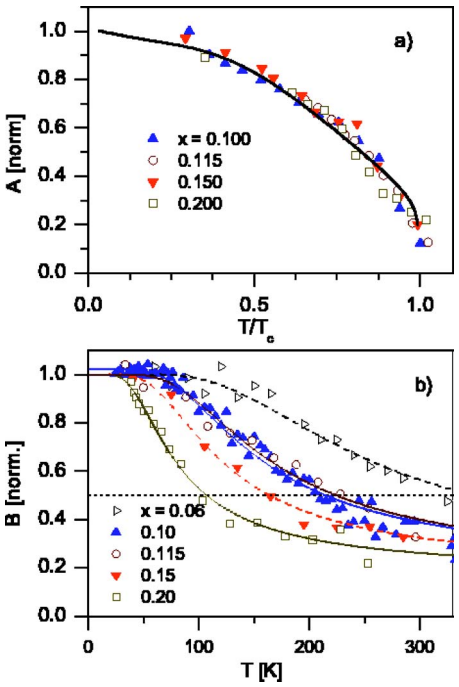

FIG. 3. (Color online) The temperature dependence of the magnitudes of the two photoinduced reflectance components (A and B) in $La_{2-x}Sr_xCuO_4$ for different doping levels—symbols. The lines present the best fit to the data using Eqs. (1) and (2) with $\Delta_c(0)$ and Δ_p as fitting parameters (see text). All the data have been normalized to their low-temperature values, and therefore the cross section of the fit with horizontal line in panel (b) corresponds to the temperature T^* (see text).

below T_c. Above T_c, we observe that the amplitude of signal B systematically diminishes with increasing temperature, similar to previous observations on YBCO,[15] $Hg_1Ba_2Ca_2Cu_3O_{8+\delta}$ (Hg1223),[2] and Y124.[3] To quantify the systematics in temperature dependence of signal B as a function of doping, we plot the temperature where the signal drops to half its maximum-low-temperature value (T^*) as a function of doping x in Fig. 4(a), together with T_c. The systematic variation of T^* with x is very similar to the one obtained in YBCO (Refs. 1 and 15) and qualitatively follows the doping dependence of the pseudogap temperature extracted by other experimental techniques. We should emphasize the qualitative doping dependence of T^*, since different criteria for its determination (e.g., T^* is the temperature where the signal B drops to 1/3 of its low temperature value) would give different absolute values of T^*, while the qualitative doping dependence would remain the same.

As discussed already in several publications[1–3] the presence of two distinct components in photoinduced carrier relaxation dynamics, one of which (A) is sensitive to the superconducting phase transition, whereas the other (B) becomes evident below the characteristic temperature T^*—which coincides with the pseudogap temperature observed by other spectroscopic techniques—implies that photoexcited carrier relaxation dynamics is sensitive to the opening of the superconducting gap and pseudogap, respectively. The coexistence of two components at temperatures below T_c, on the other hand, suggests that intrinsic phase separation persists all the way to very low temperatures, i.e., the superconducting ground state is phase separated. In order to quantify the doping dependence of the superconducting gap Δ_c and pseudogap Δ_p we fit the temperature dependence of amplitudes A and B using the model of Kabanov *et al.*,[15] assuming QP recombination across a T-dependent BCS-like gap Δ_c (below T_c) and a T-independent pseudogap Δ_p under bottleneck conditions.[15] The model has been shown to give an accurate value of the gap not only in cuprates but also in low dimensional charge-density wave systems.[17,18] In this model, the amplitude of the photoinduced reflectivity transient $|\Delta R/R|$ is linearly proportional to the photoexcited quasiparticle density $|\Delta R/R| \propto n_{qp}$, which is in turn given by

$$n_{qp} = \frac{\mathcal{E}_I/\Delta_p}{1 + B\exp(-\Delta_p/k_BT)}, \quad \Delta_p = \text{const}, \tag{1}$$

$$n_{qp} = \frac{\mathcal{E}_I/(\Delta_c(T) + k_BT/2)}{1 + B\sqrt{\frac{2k_BT}{\pi\Delta_c(T)}}\exp[-\Delta_c(T)/k_BT]}; \quad \Delta_c(T) = \Delta_{BCS}(T). \tag{2}$$

Here $B = 2\nu/N(0)\hbar\Omega$, in which $N(0)$ is the density of states at E_F, Ω is the characteristic phonon cutoff frequency, ν is the effective number of phonon modes per unit cell participating in the recombination process, Δ_p is the magnitude of the temperature-independent pseudogap, and $\Delta_c(T)$ the magnitude of the temperature-dependent gap. A BCS

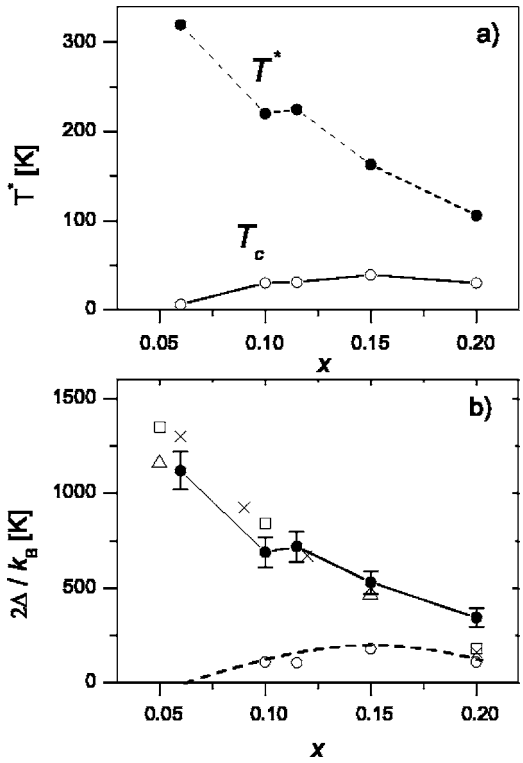

FIG. 4. (a) The effective pseudogap temperature T^*, defined as the temperature at which B drops to half its maximum value and T_c as a function of x. (b) The pseudogap Δ_p and collective gap Δ_c, obtained by fitting the data using Eqs. (1) and (2), as a function of x (solid and open circles, respectively). Δ_p is compared to E_Δ from angle-resolved photoemission spectroscopy (Ref. 20) (open triangles) and magnetic susceptibility (Ref. 21) (crosses) (Ref. 22) (open squares).

functional form $\Delta_{BCS}(T)$ was used to describe the T-dependent gap $\Delta_c(T)$ below T_c. The values of constants used are: $\nu=10$, $N(0)=2$ eV^{-1} spin^{-1} cell^{-1} (Ref. 19), and $\Omega_c=0.1$ eV. In fact Eqs. (1) and (2) are rather weakly dependent on B, so the temperature dependences of A and B are determined essentially by $\Delta_c(0)$ and Δ_p, respectively.

As seen in Fig. 3(a), the temperature dependence of the amplitude of component A shows universal behavior. In particular the data scales with T_c, and the ratio $2\Delta_c/k_BT_c \approx 4.7$. This ratio is considerably smaller than the ratio of 8-9 observed in YBCO,[1] which can be seen from the raw data on the temperature dependence of A, which is, in YBCO, much steeper near T_c than in LSCO. In fact, similar scaling can be performed also for the component B, where the data scale with T^*, as was done for the case of YBCO.[15]

The doping dependence of the pseudogap Δ_p and superconducting gap $\Delta_c(0)$ obtained from the fits are shown in Fig. 4(b). The extracted values of pseudogap Δ_p are compared with values obtained from photoemission[20] and magnetic susceptibility,[21,22] and we find quite a good agreement between the three data sets.

Finally, for completeness we plot in Fig. 5 the temperature dependence of relaxation time τ for various doping levels. As evident from the data, the temperature dependence (temperature scale normalized to T_c) is very similar for all doping

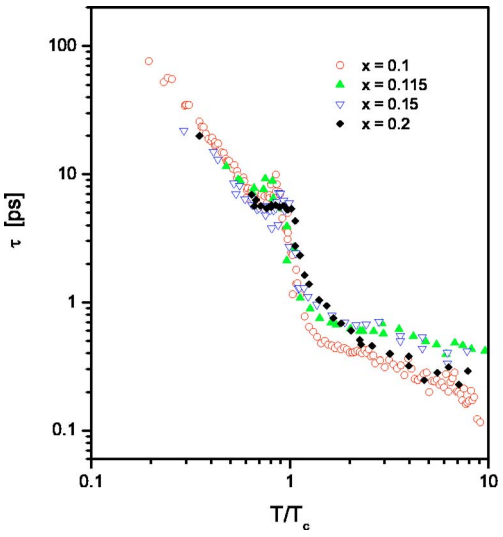

FIG. 5. (Color online) The temperature dependence of the relaxation time τ in LSCO for several doping levels x.

levels. The relaxation time increases by more than an order of magnitude upon decreasing the temperature through T_c. Upon further cooling, the relaxation time further increases, reaching almost 100 ps at the lowest temperatures that could be accessed (CW laser heating has already been accounted for). Indeed, similar temperature dependence has been observed on other cuprate materials,[2,13,24] suggesting that the behavior is similar in all cuprates. In fact, recently the same temperature dependence of the superconducting state recovery dynamics has been observed on MgB_2,[25] suggesting that the behavior may be a quite general property of superconductors. Extending the temperature range—compared to recent reports—to room temperature, there is another feature of the data that is worth mentioning. Namely, we observe that the relaxation time in the normal state also shows some temperature dependence; τ decreases upon increasing the temperature all the way to the highest temperatures measured. This is an important fact, since it presents further evidence that the normal state response is indeed governed by the presence of the pseudogap in the density of states and is not related to a simple electron-phonon thermalization that governs the relaxation dynamics in normal metals.[32–34] Namely, in conventional metals the recovery of the photoinduced reflectivity is governed by electron-phonon thermalization, where electron-phonon thermalization time increases upon increasing the temperature at temperatures above $\approx \Theta_{Debye}/5$.[32–34] This is clearly not observed in LSCO. Together with the observation that relaxation time increases also in the superconducting state, this suggests that the slowing down of the relaxation upon decreasing the temperature is a general phenomenon of relaxation across the narrow gap in the density of states.

IV. DISCUSSION

The time-resolved spectroscopic data on LSCO as a function of doping shows very similar behavior to that previously reported on other cuprates.[1–3] In particular, the observation

of two distinct components in relaxation at temperatures below T_c is consistent with the data on YBCO,[1] Y124,[3] Hg1223,[2] $Tl_2Ba_2Ca_2Cu_3O_{10}$ (Tl2223),[23] and $Tl_2Ba_2CuO_{6+\delta}$ (Tl2201).[24] In fact, it is safe to say that this observation seems to be a general feature in cuprates. Secondly, the doping dependence of the two components suggests that one is associated with quasiparticle recombination dynamics across the temperature-dependent superconducting gap, while the other describes the recombination dynamics across a temperature-independent pseudogap, whose magnitude is strongly doping dependent. Interestingly, the pseudogap is present over the entire doping range (also in the strongly overdoped samples), i.e., there is no sign of quantum criticality near optimum doping as inferred from some studies of the doping dependence of the pseudogap. Since most of the time-resolved studies performed thus far were performed on cuprates with a multiple-layer structure, consisting of CuO_2 planes and transition-metal-oxide charge-reservoir layers or Cu-O chains, the two-component relaxation observed in time-resolved spectroscopy could not unambiguously be attributed to CuO_2 planes. Naturally it was suggested that the two relaxation components might be attributed to CuO_2 planes and CR layers, respectively. Therefore the observation of two-component relaxation dynamics in LSCO is particularly important, since it implies that it is an intrinsic property of cuprates and presents a strong argument for the phase separation in CuO_2 planes of the cuprates.

While noting that there is a strong similarity in the temperature dependence of amplitudes[1–3] and relaxation times[13] among LSCO and other cuprates, we should also mention several differences. One of the striking differences is the rise-time dynamics of the component A, as already noted in Ref. 13. While in YBCO (Ref. 1) and Hg1223 (Ref. 2), the rise-time dynamics was found to be resolution limited at all temperatures, in LSCO the rise time of the component A, which describes the relaxation across the superconducting gap, was found to be ≈ 1 ps. In fact, similarly long rise-time dynamics was observed also on single layer BiSCO at low temperatures.[13] The rise time also shows some temperature dependence, as evident from the raw data in Fig. 1, where the position of the maximum in induced change in reflectivity shifts to shorter times upon increasing temperature [see the dashed line in panel (b) of Fig. 1]. Similar temperature dependence of the rise time has been recently observed in MgB_2, where it was attributed to Cooper-pair breaking by high-frequency phonons created in the initial avalanche relaxation of photoexcited quasiparticles.[25] It was argued that the fact that the anomalous rise-time dynamics was observed in MgB_2, while in most of the cuprates the rise time is shorter than 100 fs, is a result of a small superconducting gap $\Delta_c(0)$ compared to the phonon cutoff frequency Ω together with strong electron-phonon coupling. In this scenario, during the initial avalanche process, where photoexcited carriers inelastically scatter with other electrons (break Cooper pairs) and phonons, most of the energy goes into the phonon subsystem. Suppression of superconductivity is a result of these high-frequency phonons breaking Cooper pairs until quasiequilibrium between high-frequency phonons and quasiparticles is reached. When comparing LSCO to other cuprates studied thus far, the energy of the superconducting gap is comparatively smaller in LSCO. From our data it follows that in LSCO $\Delta_c(0) \lesssim 15$ meV, while in YBCO or Hg1223 $\Delta_c(0) \approx 50$ meV, while the phonon cutoff frequency $\Omega \approx 0.1$ eV for both materials. This suggests that the smaller the ratio $\Delta_c(0)/\Omega$, the more energy goes initially in the phonon subsystem, and the longer is the process of Cooper-pair breaking. The observation that in LSCO the rise time of the component B, associated with recombination across the pseudogap, is much shorter than that of the component A seems to support the above statement since $2\Delta_p \approx \Omega \gg 2\Delta_c(0)$. Interestingly, in LSCO the extracted value of Δ_p over the entire range of doping is much larger than $\Delta_c(0)$ [see Fig. 4(b)]. This is in contrast to YBCO, where near optimal doping the two are quite comparable.[1] It is interesting to note that the doping dependence, and more importantly the magnitudes of Δ_p in YBCO (Ref. 1) and LSCO are quite comparable. This observation may be relevant for deducing the nature of the temperature-independent gap in cuprates, i.e., whether Δ_p is the pairing energy in the preformed pairing model, or Δ_p is the local gap in the particle-hole channel, and T^* is the temperature where inhomogeneities appear.[26]

From time-resolved spectroscopic data we cannot infer the lateral size of the inhomogeneities. While the neutron,[7] extended XAFS (EXAFS) (Ref. 8), or tunneling data[27] suggest that the typical size is of the order of several naometers,[28] several transport measurements on ultraclean samples infer that the low-temperature mean free path is in the 0.1–1 μm range,[29,30] which is in apparent contradiction with these observations. Based on this, one could argue that the observed two-component response is due to disorder in the samples studied. However, the fact that the experimental results on photoinduced dynamics are very similar in a variety of cuprate compounds with different purity levels, thin films, and single crystals, seems to rule out this possibility, and suggests that phase separation is intrinsic.

V. CONCLUSIONS

Probably the most fundamentally important result of these experiments is the unambiguous observation of two simultaneously present relaxation components associated with Δ_p and $\Delta_c(T)$ below T_c for all x in $La_{2-x}Sr_xCuO_4$. Since $La_{2-x}Sr_xCuO_4$ has no "charge-reservoir" structure, the two components cannot be attributed to structurally separated electronic subsystems, and are observed over the entire phase diagram. Although such relaxation has been observed at selected doping levels in BiSCO (Ref. 31) and Tl1223,[23] as well as Y124,[3] systematic measurements on YBCO show a two-component relaxation only in the optimally doped and overdoped region.[1] The reason for this is probably related to a symmetric resonance of the probe optical process with interband transitions involving transitions to, and from, the states near E_F, as discussed in detail in Ref. 3. Thus, from the nearly ubiquitous presence of such multiple relaxation processes in the hole–doped cuprates, we seemingly cannot avoid the conclusion that the two processes are associated with an inhomogeneous state in the CuO_2 planes. The length scale of the inhomogeneity is certainly well below the wave-

length of light, since no inhomogeneity could be detected with optical experiments. In other words, should the length scale be much larger than the wavelength of light, the optical response would be strongly dependent on the position on the sample, which is not observed in any optical measurements. Therefore we conclude that the length scale is probably on the nanometer scale, as EXAFS and neutron PDF measurements suggest. Thus certain regions of the nanoscopically inhomogeneous state, e.g., local pairs are gapped by Δ_p, while other more extended regions have a collective, T-dependent gap $\Delta_c(T)$. As more detailed data on the electronic structure become available at different T and higher resolution, the kinematics for LSCO can be examined in further detail.

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