First publ. in: Journal of superconductivity and novel magnetism ; 24 (2011), 1-2. - S. 421-425

Dynamical Structural Instabilities in $La_{1.9}Sr_{0.1}CuO_4$ Under Intense Laser Photoexcitation

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Abstract The femtosecond timescale photoinduced reflec tivity response in $La_{1.9}Sr_{0.1}CuO_4$ under intense laser photoexcitation is reported as a function of intensity and temperature. We observe saturation of the response, which corresponds to the destruction of both superconducting gap and the pseudogap. We find that the saturation thresholds scale approximately as Δ^2, where $\Delta = \Delta_S$ for the superconducting gap and $\Delta = \Delta_P$ for the pseudogap where $\Delta_P/\Delta_S \approx 10$. The pseudogap destruction threshold correlates with the onset of a structural phase transition measured with time-resolved electron diffraction.

Keywords Superconductivity · Time-resolved optical spectroscopy · Pseudogap · Cuprate superconductors · Quasiparticle relaxation

1 Introduction

The interplay between single-particle excitations and collective-ordering dynamics in cuprates has been a subject of great interest, particularly the connection between structural and electronic ordering on different length and time scales [1]. Recently, femtosecond electron diffraction was used to investigate the structural changes in $La_2CuO_{4+\delta}$ occurring after photoexcitation by intense laser pulses [2]. At a threshold of around 5 mJ/cm^2 the material has been shown to exhibit bimodal behavior, indicating the existence of an optically induced phase transition to a state which does not exist under equilibrium conditions. The transition is characterized by a large c axis lattice expansion, which shows two metastable positions typically described by a double-well potential in the expansion of the free energy.

In a study conducted at relatively low photoexcitation densities in the range up to 100 μJ/cm^2 it was shown that the superconducting state can be controllably destroyed with femtosecond laser pulses while monitoring the quasiparticle density in real time [3], revealing details of the energy relaxation pathways in the superconducting state of the cuprate superconductors. A good phenomenological understanding of the non-equilibrium quasiparticle (QP) dynamics was obtained within the framework of Rothwarf and Taylor [4] model, which describes the QP and pairing boson population dynamics. The model [4–9] applies to phonons and spin excitations as pairing bosons equally well, and is independent of the origin of the gap (or pseudogap) giving it general applicability [5]. Indeed, apart from classical [10–12] and cuprate superconductors [13–16], the model has been used to describe electronic relaxation across the gap in numerous charge-density wave systems [17, 18], the relaxation of electrons between low-energy electronic states in Jahn–Teller ordered systems [19] and in heavy-electron systems [20].

Unique to the femtosecond spectroscopy studies of cuprate superconductors is the ubiquitous observation of two distinct components in the ultrafast relaxation dynamics [14–16]. One component is present only below T_C and can be attributed to relaxation across the superconducting gap $\Delta_s(T)$. The other component is present up to

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much higher temperature T^* (usually called the pseudogap temperature). It has been associated—based on its systematic evolution of T^* with doping [13–16] and excellent quantitative agreement with other frequency domain spectroscopies—with relaxation processes across the pseudogap Δ_p. The simultaneous presence of the pseudogap and SC gap responses at low temperatures was explained in terms of intrinsic phase separation [14]. The origin of the pseudogap remains unresolved at present, with numerous theoretical proposals still actively pursued, ranging from spin fluctuation [21], order-parameter fluctuation [22], pre-formed pairs, to Coulomb-induced mesoscale phase separation [1]. With so many different proposals, it is of great importance for further progress in the field to try and deduce the origin of the pseudogap using new experimental approaches.

In the present experiments we vary the photoexcitation laser fluence over nearly four orders of magnitude, exceeding the excitation density required to suppress superconductivity [3] by more than two orders of magnitude, aiming at determining the optical threshold for the destruction of the pseudogap in $La_{1.9}Sr_{0.1}CuO_4$. This gives us some valuable new data on the robustness of the pseudogap and as a consequence also about its origin.

2 Experimental

The time-resolved photoinduced reflectivity experiments were performed on freshly cleaved surfaces of a high-quality $La_{1.9}Sr_{0.1}CuO_4$ single crystal ($T_C = 30$ K) grown in an optical furnace [23, 24]. We used the pump and probe pulses at the wavelength of $\lambda = 805$ nm (~ 1.5 eV) from an amplified 250-kHz Ti:Sapphire laser system covering the excitation fluence from $F = 4 \times 10^{-5}$ mJ/cm^2 to 10 mJ/cm^2. The pump beam was incident along the c axis of the crystal and focused onto a spot with diameter $\rho_{pump} = 100$ μm, while the probe diameter was ~80 μm (both were measured accurately with a pinhole) to ensure homogeneous excitation density.

Linearly polarized 50 fs pulses first excite electrons from the occupied to the unoccupied hybridized (predominantly Cu–O) states within 1.5 eV of the Fermi level. The photoexcited carriers rapidly relax via e–e and e–ph relaxation processes to the states near the Fermi energy [13]. The density of the photoexcited QPs at the gap edge is probed in real time with an excited state absorption process using a second suitably delayed (probe) laser pulse. The transient change in reflectivity $\Delta R(t)/R$ (which for small $\Delta R/R$ is linearly proportional to the transient photoinduced absorption[1]) is thus directly proportional to n_{qp} at time t [13].

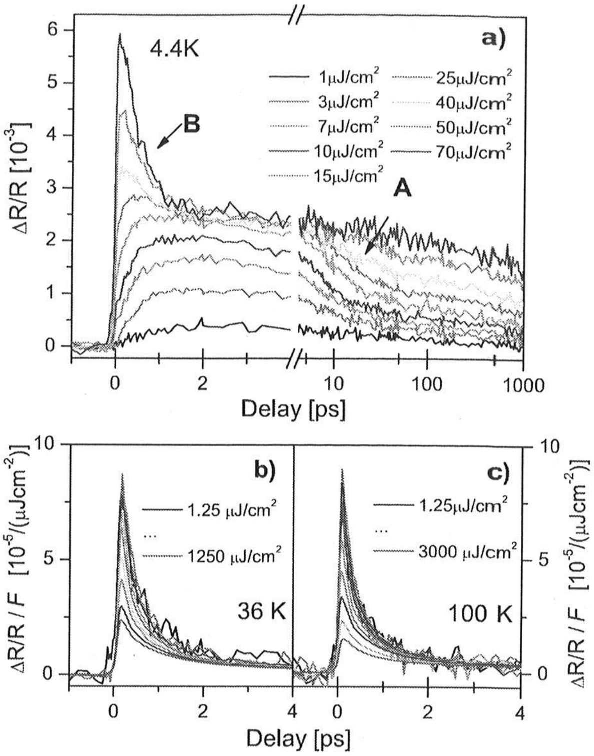

Fig. 1 The photoinduced reflectivity $\Delta R/R$ in $La_{1.9}Sr_{0.1}CuO_4$ taken at various photoexcitation fluences ($T_C = 30$ K). (**a**) Data taken in the superconducting state at 5 K. (**b**) an (**c**) Data taken above T_C at 36 K and 100 K. Data in (**b**) and (**c**) are normalized with respect to the excitation fluence. The two distinct relaxation components are marked as A and B in (**a**). Only relaxation component B remains above T_C

Taking into account the absorption depth $\lambda_{op} = 150$ nm at 810 nm [25], the photoexcited QP density at the gap Δ can be estimated [13] as $n_{qp} = F/\lambda_{op}\Delta$. Under the assumption that the gap Δ remains unchanged after photoexcitation these fluence densities correspond to $n^s_{qp} \approx 10^{18}$ to 10^{22} cm^{-3} for the superconducting gap ($\Delta_s \approx 8$ meV) and $n^p_{qp} \approx 10^{17}$ to 10^{21} cm^{-3} for the pseudogap $\Delta_p \approx 60$ meV.

3 Results and Discussion

The photoinduced reflectivity change $\Delta R/R$ as a function of time delay for different excitation intensities F is shown for $T = 4.4$ K, 36 K and 100 K in Fig. 1. Below T_C (Fig. 1a)) we identify two relaxation processes with very different dynamics, which we label as A and B. Signal A is visible only below T_C and in accordance with previous work is assigned to the QP recombination across the SC gap $\Delta_s(T)$ [14]. In contrast to signal B, whose rise time is resolution limited, the rise time of signal A is about 1 ps at low F. This long rise time has been interpreted as the time required for the QP population to build up [5, 10]. Signal B is present

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[1] For larger fluences, we can expect deviations from linearity due to photoinduced changes of the electronic band structure.

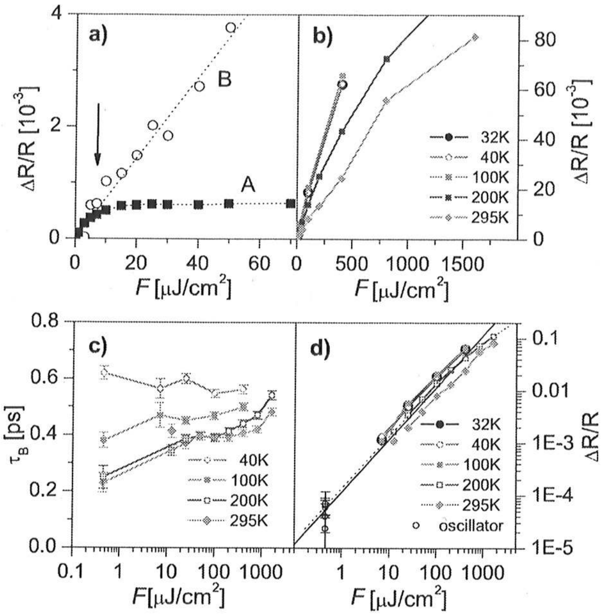

Fig. 2 Behavior of the amplitude of the photoinduced response as a function of fluence at different temperatures. (**a**) The amplitude of component A and B in superconducting state at 5 K. The threshold fluence for the destruction of superconductivity is marked with an *arrow*. (**b**) The amplitude of component B in normal state as a function of the excitation fluence. (**c**) The relaxation time of component B as a function of the fluence (**d**) Same as (**b**) but in log–log graph

from the lowest T to temperatures far above T_C. It gradually decreases as the temperature is increased above the pseudogap temperature T^* and—in agreement with previous low-fluence experiments—is assigned to the recombination across the "pseudogap" Δ_p [13, 14].

The data for all F can be fit using a single exponential decay for each of the two components A and B [14, 26, 27]. From the fit we extract the QP lifetimes and signal amplitudes of the superconducting and pseudogap responses τ_A and τ_B as functions of T and F. The results of the fitting procedure are shown on Fig. 2. Below T_C (Fig. 2a)) we see that at low F component A is dominant. As F is increased, the amplitude of signal A first increases linearly with F and saturates above ~12 μJ/cm^2. The saturation of $\Delta R/R$ signifies that all of the pairs in the condensate are photoexcited into QP states, i.e. the condensate is completely destroyed, as previously described [3]. At the same time, as the QP density (signal A) starts to saturate, signal B starts to become more visible. In this fluence range the amplitude of component B remains linear in the fluence. By further increasing the fluence the signal B becomes sublinear and above ~500 μJ/cm^2 starts to show saturation (Fig. 2b), c)). The relaxation time of component B is a few 100 fs and drops with an increasing temperature and only weakly changes with fluence.

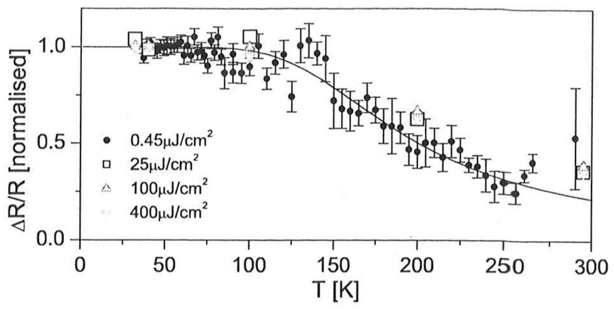

Fig. 3 Temperature behavior of the amplitude of component B at different excitation fluences Data at the fluence 0.45 μJ/cm^{2}was taken with a laser with 88 MHz repetition rate. The *line* represents a fit of (1) to the data [13]

From Fig. 3 we see that temperature behavior of the amplitude normalized with its low temperature value does not change significantly with the fluence up to 400 μJ/cm^2. The model from reference [13] describes the temperature behavior of the amplitude of the signal due to relaxation over a temperature-independent gap for the case of low excitation:

$$\frac{\Delta R}{R}(T) \propto n_{qp}(T) = \frac{\varepsilon_I/\Delta_p}{1 + C\exp(-\Delta_p/k_B T)}. \qquad (1)$$

Here $C = 2\nu/N(0)\Omega_C$ where ν is the number of phonon modes per unit cell participating in the relaxation, Ω_C is phonon frequency cut off, and $N(0)$ is the density of states at Fermi energy ($C \approx 30$). ε_I is energy density deposited by laser pulse. The magnitude of the temperature-independent pseudogap Δ_p obtained from the fit is $\Delta_p \approx 700$ K ± 100 K (see Fig. 3).

In contrast to the relatively low excitation energy density of $\varepsilon_s \approx 2$ K/Cu needed to destroy the superconducting condensate [3] the pseudogap response remains linear in the excitation fluence up to 200 K/Cu ($F_0 = 500$ μJ/cm^2). We used a model from reference [3] ((6) & (7) from Appendix to [3]) to obtain the excitation fluence required to saturate the signal and therefore destroy the pseudogap state. The value obtained from the fit (Fig. 4b)) is $F = 0.75$ mJ/cm^2 corresponding to the absorbed energy density $\varepsilon_p \approx 340$ K/Cu. If we compare this value to the energy density needed to destroy the superconducting state we find $\varepsilon_p/\varepsilon_s \approx 170$. This is close to the ratio of the BCS condensation energies [28] $N(0)\Delta_p^2/N(0)\Delta_s^2 \approx 80$, where $N(0)$ is the density of states at Fermi energy and $\Delta_S/k_B \approx 80$ K for the superconducting gap and $\Delta_p/k_B \approx 700$ K [14] for the pseudogap.

When we increase the excitation fluence over 8 mJ/cm^2 the behavior of the signal changes qualitatively with the fast response changing the sign (Fig. 4a)). For accurate determination of the threshold fluence at which this happens the experimental geometry needs to be taken into account. Because of the finite absorption depth and the Gaussian profile

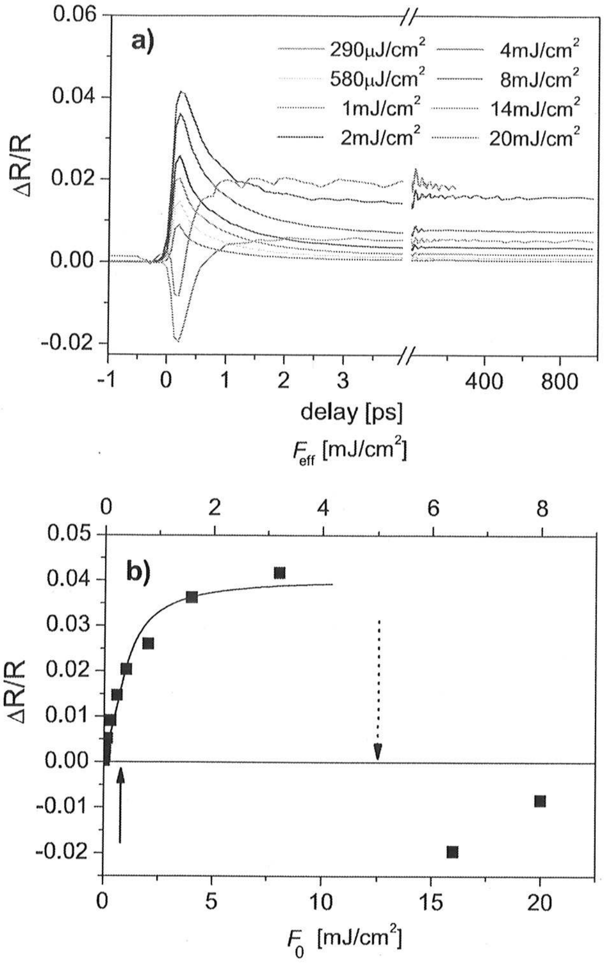

Fig. 4 Measurements taken at 36 K with the fluence up to 20 mJ/cm^2. (**a**) The photoinduced reflectivity change. (**b**) The amplitude of the fast component as a function of the fluence. *Line* is a fit of the model from reference [3] (see text) to the data. The saturation value of $F = 0.75$ mJ/cm^2 obtained from fit is marked by a *full arrow*. The fluence on x-scale in (**b**) is given in measured values (*bottom*) and effective values top (see text). The threshold value for photoinduced structural phase transition measured by Gedik et al. [2] is marked by *dotted arrow*. In (**a**) oscillations can be resolved with the period of ~35 ps. These are a result of the coherent superposition of the double reflection of the probe pulse from the first surface and from the moving strain caused by an inwardly moving sound wave [29]

of the laser beam the response has to be integrated over the inhomogeneously excited sample [3]:

$$F_{eff} = \frac{\oint_{sample} F(R)\,dR}{\oint_{sample} dR} = \frac{2F_0\rho_{pump}^2}{3(\rho_{pump}^2 + \rho_{probe}^2)}. \qquad (2)$$

Here ρ_{pump}^2 and ρ_{probe}^2 are the diameters of the pump and probe beams, F is the fluence and F_0 the fluence at the center of the laser beam. For our experiments this gives $F_{eff} \approx 0.4F_0$. If we take this into account the fluence needed to change the sign of the photoinduced signal is around 5 mJ/cm^2 (2200 K/Cu). This is similar to the fluence reported by Gedik et al. [2] where a non-equilibrium structural phase transition was observed. Since in their experiment the probe penetration depth (electrons) ~10 nm is much smaller than the pump penetration depth their measured fluence is directly comparable to our effective fluence. Although the excitation fluence is similar in both experiments it is worth noting that in the photoinduced reflectivity change we do not observe any of the characteristic time scales reported by Gedik et al. [2].

In Fig. 4a) oscillations can be resolved with the period of ~35 ps. This oscillation is a result of coherent superposition of the double reflection of the probe pulse from the first surface and from the moving strain of the sound wave [29].

4 Conclusions

We measured the photoinduced reflectivity in La$_{1.9}$Sr$_{0.1}$CuO$_4$ within a wide range of excitation fluence. In the normal state, where the signal is associated with the relaxation across the pseudogap, the signal is linear in the excitation fluence up to $F_0 = 500$ μJ/cm^2. The scaling of the saturation threshold of Δ_s^2 and Δ_p^2 with F_s and F_p is particularly intriguing. The response changes qualitatively (change of sign) at a fluence of ~5 mJ/cm^2, which was reported [2] to be the threshold fluence for a photoinduced non-equilibrium structural phase transition. This indicates that the pseudogap state is correlated with structural instabilities in La$_{2-x}$Sr$_x$CuO$_4$.

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