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Quasiparticle relaxation dynamics in n-type superconductor $La_{2-x}Ce_xCuO_4$

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Abstract

We have studied the photoexcited carrier dynamics in the electron-doped $La_{2-x}Ce_xCuO_4$ (LCCO). In general, there is a slowing down of the relaxation as the temperature decreases. However, the quasiparticle lifetime is found to exhibit a quasi-divergence as the temperature approaches T_c from below. We have also observed picosecond rise time in the electron-doped LCCO, which is attributed to the Cooper pair breaking dynamics. The experimental results are analyzed by Rothwarf–Taylor model.

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Keywords: Quasiparticle relaxation; n-Type superconductor

1. Introduction

In recent years, femtosecond pump-probe technique has been widely utilized to study nonequilibrium carrier dynamics in strongly correlated electron systems such as cuprate superconductors [1–11], heavy fermions [12,13] and charge density waves [14,15]. This technique involves the measurement of small changes in reflectivity or transmission caused by photoexcitation of quasiparticles (QP). Since the dynamics of nonequilibrium QP is extremely sensitive to the presence of a gap, it is capable of giving direct information about the temperature dependence of the QP lifetime and the gap magnitude. Most experimental results were interpreted by a phenomenological Rothwarf–Taylor (RT) model [16] which considers the phonon bottleneck in the QP recombination. On the other hand, Serge et al. [5] argued that the relaxation of photoinduced reflectivity is due to the dynamics of QP thermalization rather than their recombination into condensates.

Up to now, most femtosecond time-resolved experiments in high T_c superconductors were performed in hole-doped cuprates. On the other hand, the electron-doped cuprates show a number of distinct features which are different from the hole-doped cuprates [17–25]. For hole doped cuprates, it is commonly accepted that the pairing order parameter has d-wave symmetry [23]. However, for electron-doped cuprates, no consensus has been reached on the pairing symmetry. In addition, the transition temperature is lower and the doping range for superconductivity phase is narrower than those of the hole-doped cuprates. It is also interesting that there are two kinds of charge carriers in the electron-doped cuprates [19]. So far there has been only one report on the femtosecond relaxation dynamics in the electron-doped $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ [26]. Recently, we have reported some preliminary results of time-resolved experiments in an electron-doped superconductor $La_{2-x}Ce_xCuO_4$ (LCCO) thin film [9]. In this paper, we shall make a more detailed study on the photoexcited carrier dynamics in this electron-doped sample. In general, there is a slowing down of the relaxation of the photoexcited QP as the temperature decreases. However,

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the QP lifetime is found to exhibit a quasi-divergence as the temperature approaches T_c from below. We have also observed picosecond rise time in the electron-doped LCCO. The experimental results are analyzed by RT model.

2. Experimental details

The electron-doped LCCO thin film deposited on a (1 0 0) SrTiO$_3$ substrate was prepared by on-axis dc magnetron sputtering using a single source ceramic La–Ce–Cu–O with the composition ratio of cation La:Ce:Cu = 1.89: 0.11:1.0. The optimal-doped film had a single T′-type structure. The X-ray diffraction measurements showed that the film was highly *c*-axis oriented. The thickness was about 1500–2000 Å. The sample was near optimally doped with the highest T_c of 26 K. A more detailed description of the film preparation is given in Ref. [20].

The photoinduced differential reflectivity measurements were performed using a standard pump-probe technique. The light source was a commercial Ti:sapphire mode-locked laser, which operated at 82 MHz repetition rate with pulse duration 100 fs and wavelength centered at approximately 790 nm. The laser output was split into a pump and a probe beam with average power of 10 mW and 2 mW, respectively. The diameter of both beams on the sample surface was 100 μm and the surface was parallel to the *a–b* plane of sample. With these light intensities, the temperature of the sample increased by about 3 K. The train of the pump pulses was modulated at 1 KHz by a mechanical chopper and the probe beam reflected from the sample was collected by a photodetector and measured by an EG&G7265 lock-in amplifier. To avoid coherent artifacts and also reduce the scattering of pump beam into the detector, the pump and probe beams were cross polarized. The samples were mounted on a cold finger in a liquid-helium continuous-flow optical cryostat.

3. Experimental results and data analysis

Fig. 1 presents the temporal evolution of the photoinduced differential reflectivity $\Delta R/R$ at a few temperatures. From the logarithmic plots (see the inset), it is evident that the data can be fit by a single exponential decay when $T > 40$ K. On the other hand, below 40 K there exists two distinct relaxation processes, one with a lifetime of a few picoseconds and the other with sub-nanosecond lifetime. We plot the picosecond component of the relaxation time τ as a function of temperature in Fig. 2. It shows that, as T increases, τ first decreases to reach a minimum. Then it is followed by an anomalous increase before T_c is reached. We also found that, above T_c, the relaxation time which is much longer than what is expected for relaxations in a metal, decreases again with the increase of T.

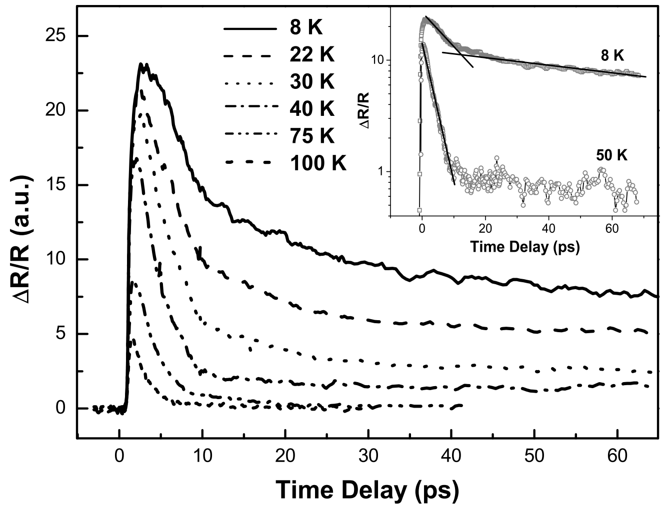

Fig. 1. Photoinduced differential reflectivity $\Delta R/R$ as a function of time delay at different temperatures. The inset shows the data at 8 K and 50 K on the logarithmic scale. The solid lines in the inset are the theoretical fits.

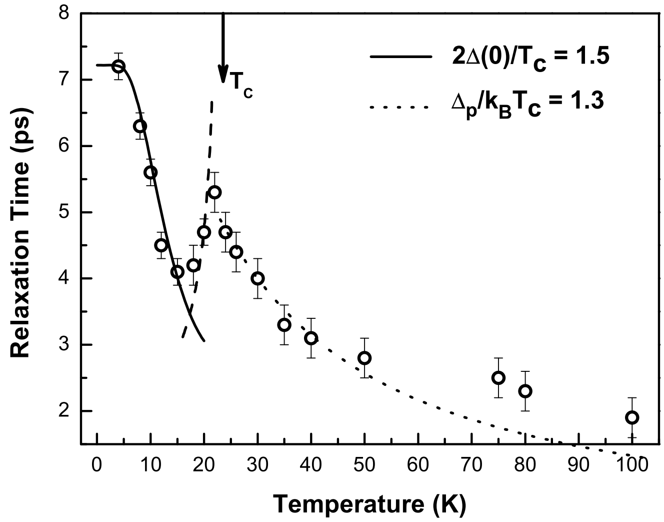

Fig. 2. The temperature dependence of the relaxation time. The solid, dashed and dotted curves are theoretical fits to the data in the low-temperature, near-T_c and above-T_c regions, respectively.

Now, we analyze the experimental results. It is generally believed that for most conventional superconductors as well as cuprates, the nonequilibrium QP dynamics is governed by phonon bottleneck mechanism, where the superconducting state recovery dynamics are governed by the lifetime of high frequency phonon (HFP) with frequency $\hbar\omega \geqslant 2\Delta$, which are in thermal equilibrium with QPs. Here, Δ is the gap magnitude. Theoretically, this dynamics can be interpreted by a phenomenological RT model [16] which involves the evolution of QP and HFP populations. In Fig. 2, the most noticeable feature is the appearance of a quasi-divergence of the relaxation time as T approaches T_c from below. This anomaly is attributed to the closing of a superconducting gap. Specifically, near T_c the relaxation is governed by the anharmonic decay time of HFPs, which is $\tau_{ph} \propto 1/\Delta(T)$ [3] as plotted in Fig. 2 (dashed curve).

Let us turn to the QP relaxation at low temperatures. Fig. 2 shows the increase of the relaxation time as

$T \to 0$, which was previously attributed to the bi-particle recombination process [4]. However, Kabanov et al. [16] have demonstrated that this behavior can be explained by the RT model in the strong bottleneck regime. According to their theory, the T-dependence of τ^{-1} is given by

$$\tau^{-1}(T) = \Gamma[\delta(\zeta n_T + 1)^{-1} + 2n_T], \tag{1}$$

where Γ, δ, and ζ are T-independent fit parameters. The density of thermally excited QPs n_T at temperature T is $n_T(T) \propto \sqrt{\Delta(T)T}\exp(-\Delta(T)/T)$, where $\Delta(T) = 1.74\Delta(0)\sqrt{1 - T/T_c}$ according to the BCS theory. The solid curve in Fig. 2 is the fit to our data, from which we obtain $2\Delta(0)/T_c = 1.5$. We then consider the relaxation process above T_c. It is found that the relaxation time is much longer than what is expected for metallic relaxation, indicating the presence of a T-independent pseudogap Δ_P below T^*. The effects of the pseudogap on relaxation process can be described by RT model. Thus, Eq. (1) is applicable if we replace $n_T(T)$ by $n_T \propto T\exp(-\Delta_P/k_B T)$. The dotted line in Fig. 2 is the theoretical fit with $\Delta_P/k_B T_c = 1.3$.

Finally, we consider the rise-time dynamics in the electron-doped LCCO. As shown in Fig. 3a, the electron-doped LCCO is characterized by a long rise time of picosecond time scale; while, in most cuprates, the rise times are shorter than 100 fs. Similar behavior has been observed in MgB_2 [6] and $La_{2-x}Sr_xCuO_4$ [7]. Fig. 3b presents the temperature dependence of the time when $\Delta R/R$ reaches maximum. Physically, when there is strong electron–phonon coupling and the superconducting gap is small compared to the phonon cutoff frequency, then during the initial avalanche process most of the energy will go into HFPs. The anomalous rise-time dynamics is attributed to Cooper pair breaking by these phonons. The pair-breaking dynamics can be analyzed by RT model. In the initial pair-breaking stage, the relaxation of HFP can be neglected, then the time evolution of the QP density $n(t)$ is given by [6]

$$n(t) = \frac{\beta}{R}\left[-\frac{1}{4} - \frac{1}{2\tau_1} + \frac{1}{\tau_1}\frac{1}{1 - K\exp(-t\beta/\tau_1)}\right]. \tag{2}$$

Here, R is the bare QP recombination rate; β is the probability for pair breaking by HFPs; K and τ_1 are dimensionless parameters determined by the initial conditions. The rise time is related to τ_1, which is given by

$$\frac{1}{\tau_1} = \sqrt{\frac{1}{4} + \frac{2R}{\beta}(n_0 + 2N_0)}, \tag{3}$$

where n_0 and N_0 are the initial QP and HFP densities after photoexcitation, respectively. The solid line in the inset in Fig. 3a is the theoretical fit to the experimental data at 8 K using Eq. (2). Now, let us consider the temperature dependence of the rise time. In the weak perturbation limit, where the photoinduced concentrations of PQs and HFPs are much less than the corresponding thermally excited concentrations, we have $n_0 \simeq n_T$ and $N_0 \simeq N_T$ with N_T the concentration of the thermally excited HFP. Since n_T and N_T increase with the temperature, according to Eq. (3), the rise time decreases as the temperature increases. It is worth mentioning that in the electron-doped LCCO the condition of small superconducting gap for Cooper pair breaking dynamics is consistent with the fact that it has low T_c of 26 K.

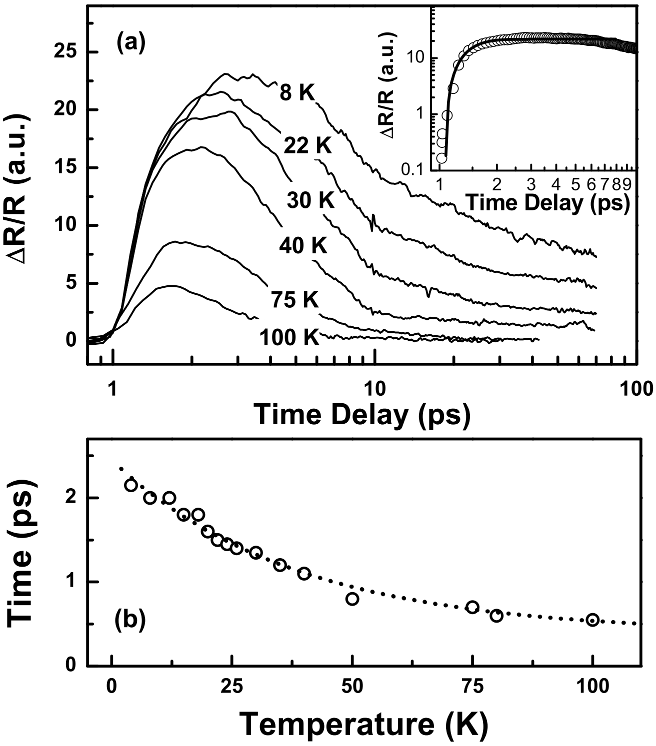

Fig. 3. (a) Photoinduced differential reflectivity $\Delta R/R$ as a function of time delay at different temperatures. Note that the x-axis is plotted on the logarithmic scale and all traces have been shifted horizontally by 1 ps. Inset is the theoretical fit (solid line) to the experimental data (open circles) at 8 K using Eq. (2). (b) Temperature dependence of the time when $\Delta R/R$ reaches maximum. The dotted curve is a guide to eye.

4. Conclusion

Up to now, most femtosecond time-resolved experiments have been performed in hole-doped cuprates, which have shown the importance of the phonon bottleneck in the nonequilibrium QP dynamics. In this paper, we study the photoexcited carrier dynamics in the electron-doped LCCO and find that the QP relaxation is again governed by the phonon-bottleneck mechanism. We have also observed picosecond rise time in the electron-doped LCCO, suggesting that after photoexcitation energy is initially relaxed to HFPs, instead of electron–electron thermalization.

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