Femtosecond optical studies of cuprates


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ABSTRACT

Femtosecond optical reflectivity measurements of La$_{2-x}$Sr$_x$CuO$_4$, La$_2$CuO$_{4+y}$, Bi$_2$Sr$_2$CuO$_{6+z}$ and Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ thin films and single crystal samples indicate qualitative changes with fluence. At the lowest fluencies, there is a power law divergence in the relaxation time. The divergence has an onset temperature of 55±15K, independent of whether the sample is in the superconducting or normal states. At slightly higher fluencies, still perturbative, the additional response does not exhibit this power law divergence. At quite high fluencies- no longer perturbative- the metallic samples exhibit oscillations in the reflectivity amplitude. The period of these oscillations varies with the probe wavelength but not with the pump wavelength. The oscillations exhibit a decay time as long as 10 nsec.

1. INTRODUCTION

There have been several reports of studying cuprates and simple metals using the technique of femtosecond optical reflectivity.[1-22] The technique provides direct information on carrier dynamics in these systems.[2] In metals, the classic model is called the ‘two-temperature’ model.[1] Developed in the U.S.S.R. in 1956, this model assumes that after light is absorbed by electrons, these photoelectrons (PE’s) thermalize immediately with the other electrons (TE’s) and establish an effective electron temperature. The TE’s then relax by dissipating energy to the lattice on a relaxation time (τ) that is typically the time scale of lattice vibrations (~ 1ps). The system then, much more slowly, removes the energy from the illuminated region by thermal conduction (~ 1-10 ns). Several reports on the cuprates have shown that the relaxation time changes across the superconducting phase transition temperature (Tc), and is (~ 0.3 ps) well above Tc and almost constant at (1-10 ps) below Tc. In simple metals, an extension of the two-temperature model has been developed [4-9] to account for the measured finite time in which PE’s reach thermal equilibrium with the other electrons (TE’s).

We report two qualitatively new results. At sufficiently low fluencies, which we term “lowest fluence,” we measure a divergence in (τ), the relaxation time, with temperature (T). We find that for several different cuprate samples, (τ) ∝ T^{-N} (N = 2.5 ± 0.5). This divergence depends on fluence. As the fluence increases, the additional reflectivity signal shows
the behavior described above, not the divergence measured at sufficiently low fluencies. The other new result is at very high fluencies, for which we observe oscillations in the reflectivity response. These oscillations are very low frequency, as low as 20 GHz, and long-lived, with damping times as long as 10 ns. In the remainder of this paper, we present experimental information, the results, and a brief discussion of the results.

2. EXPERIMENTAL

Figure One illustrates schematically the laser system used. We reproduced the data shown in at least two laboratories, and for much of the data in four laboratories. The pump fluence (energy/area) for the “lowest fluence” measurements ranged from 3 x 10^{-12} joule/pulse (4 x 10^{-8} joule/cm^2/pulse) to 2.5 x 10^{-11} joule/pulse (3.2 x 10^{-7} joule/cm^2/pulse); for “perturbative fluence” from 1.2 x 10^{-10} joule/pulse (1.6 x 10^{-6} joule/cm^2/pulse) to 6 x 10^{-10} joule/pulse (8 x 10^{-6} joule/cm^2/pulse); for “high fluence” from 5 x 10^{-8} joule/pulse (7 x 10^{-4} joule/cm^2/pulse) to 5 x 10^{-7} joule/pulse (7 x 10^{-3} joule/cm^2/pulse). The inset of Fig. 1 shows a typical reflectivity versus time delay spectrum.

We fabricated samples of La_{2-x}SrxCuO_4 (0.04 \leq x \leq 0.30) thin films on SrTiO_3 and buffered LaSrAlO_3 on SrTiO_3, La_2CuO_4+x thin films on SrTiO_3 and LaSrAlO_3, Bi_2Sr_2CuO_{6+x} single crystals (doped with La) and thin films (not doped with La) on LaSrAlO_3. Thin films were fabricated in three different laboratories and yielded virtually identical results. Figure Two shows the in-plane resistivity of the La_{1.70}Sr_{0.30}CuO_4 samples, which are metallic but non-superconducting. The residual resistivity is \sim 40 \mu\Omega \cdot cm, which corresponds to a mean free path of approximately 24 nm, much longer than the superconducting coherence length of cuprates (1-2 nm). The sample exhibits T^2 behavior at low temperatures (T) and \sim T behavior at higher temperatures. Resistivity data is not sensitive to small inhomogeneities. X-ray diffraction data taken on these samples (not shown) indicate uniform films (within the limitations of such data). Single crystals of Bi_{2-x}Sr_xCaCu_2O_y were grown by the Floating Zone method using an infrared image furnace at low final pass growth rates of 0.05 - 0.1 mm/hr. At optimal oxygen doping, Tc = 95K. Oxygen content was varied by annealing as-grown crystals in different partial pressures of oxygen.

3. RESULTS

3.1. Lowest fluence

Figure Three shows reflectivity data versus time delay for La_{1.70}Sr_{0.30}CuO_4, a metallic, nonsuperconducting thin film sample. The data were taken at actual (during illumination by the pump) sample temperatures from 7K- 32K. The initial slope (change versus time) of the response is approximately temperature independent over this temperature range. The amplitude of the signal decreases by a factor of 3x over this temperature range. The relaxation after reaching maximum amplitude is markedly longer at lower temperatures.
Figure 3 Femtosecond optical reflectivity versus time delay for La$_{1.70}$Sr$_{0.30}$CuO$_4$ thin film sample. The data were taken at temperatures of 7K, 13K, 17K, 21K, 24K and 32K. The temperature at which different spectra were taken can be determined by noting that the amplitude of the signal decreases monotonically with temperature.

Figure 4 Femtosecond optical reflectivity versus time delay for La$_{1.94}$Sr$_{0.06}$CuO$_4$ thin film sample. The data were taken at temperatures of 11K, 20K, 36K and 51K.

Figure 5 Femtosecond optical reflectivity versus time delay for Bi$_2$Sr$_2$CuO$_{6+x}$ single crystal sample (doped with La). The data were taken at temperatures of 12K, 14K, 18K, 23K, 33K and 43K. The spectra exhibit a monotonic decrease in the relaxation time with increasing temperature.

Figure Four shows reflectivity data versus time delay for La$_{1.94}$Sr$_{0.06}$CuO$_4$, an underdoped superconducting thin film sample. The initial slope of the response is approximately temperature independent over this temperature range. The amplitudes have been normalized, but in fact the amplitude decreases monotonically with temperature. The relaxation after reaching maximum amplitude is markedly longer at lower temperatures.

Figure Five shows reflectivity data versus time delay for Bi$_2$Sr$_2$CuO$_{6+x}$ superconducting single crystal sample. The initial slope of the response is approximately temperature independent over this temperature range. The amplitudes have been normalized, but in fact the amplitude decreases monotonically with temperature. The relaxation after reaching maximum amplitude is markedly longer at lower temperatures and decreases monotonically with increasing temperature.

Figure Six shows data taken on near-optimally doped Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ single crystal samples (T$_C$ = 92K, 10-90% transition width < 2K). At a sample temperature during pump pulse illumination of 17K, the data show a long relaxation response, with a relaxation time of ~ 70 ps. By contrast, at 77K, still in the superconducting state, the relaxation time is much faster at ~ 1 ps. These data, and data taken on slightly underdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ (T$_C$ = 87K, 10-90% transition width ~ 5K) and overdoped (T$_C$ = 75K, 10-90% transition width ~ 3K) single crystal samples, establish that there is a quasi-divergence in the relaxation time versus temperature for sufficiently low fluence for these two-layer cuprate samples.

Figure Seven shows the relaxation time versus actual temperature during pump illumination for nine different samples, including LSCO thin films (x= 0.06, 0.08, 0.10, 0.15, 0.25, and 0.30), BSCCO-2201 single crystals (two different stoichiometries of La) and BSCCO-2212 single crystals. The values of the superconducting transition temperature range from 0K (LSCO, x=...
0.30) to 92K (optimally doped BSCCO-2212). The data all show a power law divergence in the relaxation time ($\tau$), with $\tau \propto T^{-N}$, $N = 2.5 \pm 0.5$, depending on sample. There are small changes in the relaxation time across the superconducting phase transition temperatures, but the dominant response is that of a power law divergence. The divergence occurs at an onset temperature of $55 \pm 15K$ for single layer cuprate samples. For the double layer, BSCCO-2212 samples, we could not take data above 80K at the lowest fluence. The extrapolated onset temperature is $125 \pm 15K$ for the double layer, BSCCO-2212 samples, as shown in the inset of Fig. 7. The onset temperature is in the normal state for single layer cuprates and the extrapolated onset temperature for double layer cuprates is also in the normal state.

3.2. Perturbative fluence

Figure Eight shows the reflectivity versus time delay spectra for a La$_2$CuO$_4+\delta$ superconducting thin film sample; the superconducting transition temperature of this sample is 25K. The initial slopes of the spectra are approximately the same for all temperatures. The relaxation time does not show a power law divergence, and does show a marked change across the superconducting phase transition temperature, as shown in the inset of Fig. 8. The dominant relaxation time response is almost constant, and fast, in the normal state, with the relaxation time as fast as 0.26 ps. There is a large increase in the relaxation time when entering the superconducting state, with values for this type of sample ~ 6 ps, similar to several earlier reports. As reported earlier by some of us and by other investigators, the relaxation time at temperatures below approximately 0.8 $T_C$ is almost constant.[14-21]
Figure 10  Reflectivity versus time delay spectra for La$_{1.70}$Sr$_{0.30}$CuO$_4$ thin film grown on SrTiO$_3$. The top spectrum was taken for pump/probe wavelengths of 800/800 nm, the middle spectrum for 400/800 nm, and the bottom spectrum for 800/400 nm. The inset shows the oscillation frequency versus the probe wavevector.

Figure 11  Reflectivity versus time delay for: top: 39 nm La$_2$CuO$_4$ on SrTiO$_3$; second: 52 nm La$_2$CuO$_4$ on SrTiO$_3$; third: 76 nm La$_{1.70}$Sr$_{0.30}$CuO$_4$ on SrTiO$_3$; bottom: 100 nm La$_{1.85}$Sr$_{0.15}$CuO$_4$ on LaSrAlO$_3$ buffer on SrTiO$_3$.

3.3. High fluence
Figure Nine shows reflectivity versus time delay data taken at high fluence for a La$_2$CuO$_4$ thin film grown on SrTiO$_3$. Data taken at nominal sample temperatures of 15K and 285K are shown; we also measured the sample at four other temperatures between 15K and 285K (data not shown). There is an oscillation in the reflectivity amplitude. The oscillation period, and the damping of the oscillations, are independent of temperature. We have data (not shown) indicating that the damping time (\(\eta\)), assuming that the amplitude decreases as (e^{-t/\eta}), can be as long as 10 nsec.

Figure Ten shows reflectivity versus time delay taken on a La$_{1.70}$Sr$_{0.30}$CuO$_4$ thin film grown on SrTiO$_3$. The top spectrum was taken for pump/probe wavelengths of 800/800 nm, the middle spectrum for 400/800 nm, and the bottom spectrum for 800/400 nm. These and other data not shown indicate that the period of oscillation depends on the probe wavelength but does not depend on the pump wavelength. The inset of Fig. 10 shows the oscillation frequency versus the probe wavevector. The two quantities are related in approximately a linear fashion.

Figure Eleven shows reflectivity versus time delay data for four spectra, including: top: 39 nm LCO on STO; second: 52 nm LCO on STO; third: 76 nm LSCO on STO; bottom: 100 nm LSCO on buffer LSAO/STO. The top and second spectra were taken on the same material, grown on the same substrate, but the film thickness varies by a factor of x 1.3. The top, second, and third spectra span a factor of x2.0 in film thickness, with no difference in oscillation period. The oscillation period does not depend on film thickness and is the same for LSCO and LCO films grown on SrTiO$_3$. The third and bottom spectra were taken on the same material with slightly different film thicknesses, but the substrate was different. The third spectrum was taken from a film grown directly on SrTiO$_3$, while the bottom spectrum was taken from a film grown on a 100 nm LaSrAlO$_3$ buffer layer on top of SrTiO$_3$. The bottom spectrum shows very strong damping and a different oscillation period compared to all the films grown directly on SrTiO$_3$. Both the period of oscillation and the damping depends markedly on whether the film is grown on SrTiO$_3$ or on LaSrAlO$_3$.

4. DISCUSSION AND CONCLUSIONS

4.1. Lowest fluence
The main result from studies of cuprates using the lowest feasible fluence is the power law divergence in the relaxation time as shown in Fig. 7. The divergence has been measured for a total of eight different types of samples, including three different single-layer cuprates, so the behavior seems a general property of cuprates. This divergence begins at an onset temperature that is independent of the superconducting state temperature (Fig. 7 inset). The data indicate that for these samples, the power law divergence of the relaxation time is a property of the normal state, in contrast to the conclusion drawn by Ref. 22. One of us (JD) and colleagues has analyzed the relaxation time versus temperature expected assuming a fully gapped electronic density of states.[22] The analysis predicts an exponential divergence in the relaxation time. However, such analysis does not account for our data on the metallic, nonsuperconducting samples (which have no gap). We have no microscopic model for this behavior.
Figure 12 illustrates schematically the extension of the two-temperature model developed in Refs. 4-9. The extension involves a finite thermalization between the photoelectrons (PE’s) and the other electrons (TE’s), rather than the immediate thermalization assumed in Ref. 1. Following Refs. 4-9, we can model the reflectivity response by:

\[
\Delta R \propto A(T) [H(t) \left[ 1 - \exp(-t/\tau_{TH}) \right] \left[ \exp(-t/\tau_P) \right]]
\]

Using Eq. (1) we obtain quantitative agreement with our data using three parameters: the (A), amplitude of the reflectivity signal, \(\tau_{TH}\) the thermalization between PE’s and TE’s, and \(\tau_P\), the relaxation time to the lattice. Our data show that the model of Refs. 4-9, which leads to Eq. (1), has one clear limitation. We find that we must make the amplitude (A) temperature-dependent, while the model would assume that the amplitude of the reflectivity signal is temperature independent. This is particularly clear at lower temperatures (below 100K), where we must allow (A) to vary with temperature to obtain quantitative fits to the data. Thus, while Eq. (1) and the model behind it give excellent fits to individual spectra, obtaining quantitative agreement with sets of data involves changing the overall amplitude factor with no physical justification or explanation.

4.2. Perturbative fluence

The main result of using somewhat higher fluence is that instead of a power law divergence, we measure a very fast relaxation time in the normal state and a much slower- but almost temperature independent- relaxation time in the superconducting state. This is the result in several earlier reports, which all used fluencies at or higher than those we used in these measurements. We have also reported elsewhere [21] that at intermediate fluencies between “lowest” and “perturbative” we measure both behaviors: there is a part of the reflectivity change that exhibits a divergence and another part of the change that exhibits the behavior shown in Fig. 8. The data indicate that at the lowest fluencies, the reflectivity signal exhibits a divergence. When the fluence is increased beyond some value of approximately 5 x 10^{-11} joule/pulse (8 x 10^{-7} joule/cm^2/pulse), the additional reflectivity signal does not exhibit a divergence. Thus, at fluencies we term “perturbative,” the dominant reflectivity signal does not show a divergence.

Comparing the “lowest fluence” and the “perturbative fluence” data lead both to new insight and puzzlement. For a BCS superconductor, both \(\tau_{TH}\) and \(\tau_P\) would be affected by the opening of the superconducting gap, and neither would be affected if the sample is metallic but nonsuperconducting. Consider the data of Fig. 13. This shows the electron-electron thermalization time versus temperature for nonsuperconducting samples (La_{0.3}Sr_{0.7}CuO_4) and superconducting samples (La_{0.3}Sr_{0.7}CuO_4 thin films and Bi_{2}Sr$_2$CuO$_6$ (doped with La) single crystals). \(\tau_{TH}\) is fast (~ 0.1 ps) and temperature independent for the non-superconducting samples; it is the same for superconducting samples well into the normal state. However, \(\tau_{TH}\) shows a marked increase at or near T_c for all superconducting samples at both lowest and perturbative fluencies. So at perturbative fluences both \(\tau_{TH}\) and \(\tau_P\) are affected by the phase transition, while at the lowest fluencies only \(\tau_{TH}\) is affected by the phase transition.
3.3. High fluence
The main result of the high fluence data is the presence of reflectivity oscillations. The period of oscillation depends on the probe wavelength, but neither on the pump wavelength, the film thickness, nor the nominal sample temperature. We have varied the pump fluence over a factor of 10x, and the oscillation period and damping are independent of pump fluence. The oscillation frequency and the probe wavevector are approximately linearly related. There are reports in the literature of reflectivity oscillations,[23-27] including reports that the oscillation period depends linearly on the probe wavelength.[25,27] However, the systems discussed in Refs. 25 and 27 are insulators or semiconductors. The systems for which the oscillation period depends on the probe wavelength are also systems for which the oscillation period is linearly proportional to the film thickness.[25,27] For our data, the results are ambiguous. On the one hand, the oscillation period is independent of film thickness. On the other hand, both the damping and the oscillation period is affected by the choice of substrate. Also, all the systems we have measured are metals. For metals, the dominant reflectivity response is due to the conduction band electrons, in marked contrast to the behavior of insulators and semiconductors. We have no microscopic model of why metallic systems act like insulators in exhibiting reflectivity oscillations at high fluencies.

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REFERENCES


