# Ultrafast nonequilibrium carrier relaxation in single-crystal $Nd_{1.85}Ce_{0.15}CuO_{4-y}$

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We present a temperature-dependent investigation of the femtosecond relaxation dynamics of the *n*-type superconductor $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ using ultrashort pulses from a Ti-sapphire laser in a transient absorption, pump-probe configuration. The energy relaxation time of the single crystals in the normal state was observed to increase by over an order of magnitude as the critical temperature was approached. This behavior has been modeled by the scattering of electrons with two-dimensional phonons. In the superconducting state, an increase and subsequent saturation of the relaxation time has been discerned. A possible reason for this is a complex interplay among the characteristic relaxation times, including scattering and recombination of quasiparticles optically excited by ultrashort laser pulses and order parameter relaxation close to $T_c$.

Among the myriad of experimental techniques employed to shed light on the microscopic origin of superconductivity, photoexcitation studies have often played an important role.[1] Continuing advances in the development of ultrafast-pulse lasers have presented opportunities to study the carrier and quasiparticle dynamics of various materials directly in the time domain. These sources now provide optical pulses with duration typically between 50 and 100 fs, which is less than or comparable to electron-phonon energy relaxation times in the relevant materials. The first pump-probe transient absorption studies to use these ultrashort pulses concentrated on the inelastic relaxation of nonequilibrium electrons in metals.[2] They were soon followed by a number of attempts to investigate the nonthermal carrier distributions in high-temperature superconductors.[3–5] By providing information on the dynamics of quasiparticles, such studies have contributed to our understanding of the nonequilibrium superconducting state and also helped to ascertain technologically relevant questions concerning the relative importance of bolometric and nonbolometric response to optical stimulation.

Transient absorption, when observed in *d*-band metals and high-$T_c$ superconductors at temperatures above $T_c$, is frequently interpreted as thermomodulation of optical reflectivity and is believed to arise from excitations by photons to empty states near the Fermi level.[6,7] Since the "pump" pulse employed is of very short duration, this external thermal stimulation is only present for a very short time interval. A subsequent rapid internal thermalization among the carriers then leads to a smearing of the Fermi energy, i.e., emptying states below the Fermi level and increasing their occupancy above the Fermi level. The overall effect is a very fast change in the reflectivity (or transmissivity) of the sample detected by a second, perfectly synchronized and variably delayed, "probe" pulse. The reflectivity subsequently relaxes on a longer time scale as the carriers undergo inelastic scattering with phonons. This so-called "energy smearing" model has proved useful in explaining the behavior of both *d*-band metals and a number of hole-dominated high-$T_c$ perovskites.[4,6,7]

The nonequilibrium distribution caused by pulsed laser stimulation shows a dramatic change in character for temperatures near and below the superconducting transition point. Formation of the superconducting condensate (Cooper pairs) at an energy $\Delta$ below the Fermi level has a decisive influence on the absorption of the pump and probe electromagnetic radiation. For instance, a 2-eV pump photon, along with the resulting phonons from the multiple cascading process as the pairs recondense, can break up to 100 or so Cooper pairs, leading to a dramatic change in the absorption—and thus also in the reflectivity and transmissivity—of the probe.

Since the pump-induced change in the imaginary part of the dielectric function, $\Delta\epsilon_2$, is the actual variable which corresponds to the state occupancy in the sample, and thus also the quantity proportional to the pump-induced change in absorption, it would be best to have its value determined. However, $\Delta\epsilon_2$ is strictly dependent on both the fractional reflectivity and transmissivity changes ($\Delta T/T$) in a high-$T_c$ superconductor, and the single crystals used in this study were prohibitively thick to allow any measurement of transmissivity. Regardless, it is assumed here that enough similarities exist between the high-$T_c$ crystals under study and other electron-transport materials such as gold that results obtained from $\Delta R$ measurements are induced by the thermomodulation (i.e., $\Delta R/R \propto \Delta\epsilon_2$) and are relevant to relaxation-time studies in superconductors.

Previous studies in our group and elsewhere have demonstrated the power of femtosecond pump-probe techniques in providing insight into the relaxation time and the carrier-phonon interaction in several hole-type high-$T_c$ perovskites. In this work, we address the topic of quasiparticle dynamics in an electron-type high-$T_c$ material, namely $Nd_{1.85}Ce_{0.15}CuO_4$(NCCO). The existence of electron-doped high-$T_c$ perovskites has already demonstrated that superconductivity is not exclusively tied to

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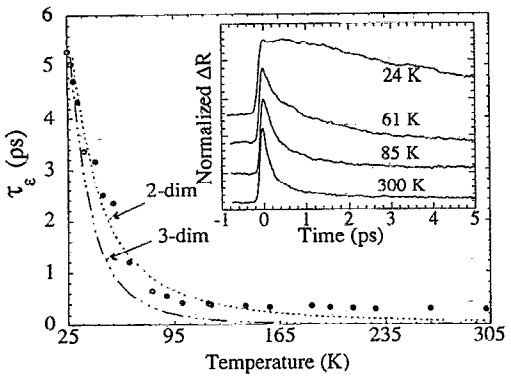


FIG. 1. Temperature dependence of the instantaneous energy relaxation time and the two-temperature-model fit using 2D and 3D phonon spectra. Inset: Normalized reflectivity changes ($\Delta R/R \approx 10^{-3}$) vs pump-probe delay time with the sample in the normal state.

hole-type conduction.[8] Here we present a comparison of relaxation-time behavior between the two types of materials.

The samples used in our measurements were shiny, platelike single crystals ($2\times2\times0.2$ mm$^3$) grown by the self-flux method with the $c$ axis perpendicular to the plate.[9] A dc magnetic susceptibility measurement revealed the onset of superconductivity to be at 24 K, with a transition width of about 3 K. To measure the photoinduced change of reflectivity, we used a pump-probe arrangement similar to ones reported previously (e.g., Ref. 3). Our short-pulse light source was a passively mode-locked Ti-sapphire laser with a center wavelength of 780 nm (1.59 eV), a pulse width of 75 fs, and a pulse repetition rate of 76 MHz. The change in refractive index induced by carriers heated by the pump pulse resulted in the change of reflectivity of the probe pulse, which was sensed by a silicon photodiode and plotted versus the pump-probe delay time. The pump pulse was perpendicularly polarized to the probe pulse to avoid interference. The measurements were typically carried out with the pump beam—4 mW average power—focused into a spot of 50 $\mu$m diameter and the probe beam, with an average power of 1 mW, focused into a 30-$\mu$m diameter. Decreasing the power of the probe beam resulted in noisier traces, but also yielded essentially the same reflectivity curves, indicating that there are no probe-induced effects on the data in this fluence regime. Heating due to the average power of the laser pulse trains was estimated to be less than 2 K.

In the inset of Fig. 1, the change of reflectivity versus pump-probe delay time at a series of sample temperatures above $T_c$ (corrected for laser average power heating) is shown. The essential features consist of, first, a rapid transient rising to a peak at a reference of 0 ps, followed by a temperature-dependent exponential-like decay. The detected probe beam signal, which has its photon energy close to the interband transition across the charge transfer gap [1.5 eV for $Nd_2CuO_4$ (Ref. 10)], is a direct measure of the relaxation of the nonequilibrium electrons near $E_f$. If we assume that the measured reflectivity is proportional to $\Delta\epsilon_2$ for this NCCO sample, the positive change of reflectivity we observed indicates probing above the Fermi level.

The temporal evolution of the $\Delta R/R$ signal may be explained as follows. The excited hot electrons first thermalize via *e-e* collisions within a very short time, $\sim$200 fs. This is characterized by the rise time and is in good agreement with other experiments.[11] The initial, picosecond-regime decay results from cooling of the hot electrons via inelastic scattering with phonons, while the subsequent slow, nearly flat decay arises from the phonon-phonon relaxation, which is typically in the nanosecond range. The instantaneous energy relaxation time, $\tau_\epsilon$, may be used to describe the rapid inelastic scattering of electrons with phonons.[12] In order to extract $\tau_\epsilon$ from the experimental data, we perform a least-square fit to the initial decaying part of the reflected time-domain signal using $c_1+c_2\exp(-t/\tau_\epsilon)$. The time constant of the exponential term gives $\tau_\epsilon$, while the $c_1$ models the heat diffusion out of the illuminated volume.

In Fig. 1, we plot the temperature-dependent $\tau_\epsilon$ for $T>T_c$. A rapid increase in $\tau_\epsilon$ is noted as the temperature is lowered, with the onset of the rise particularly noticeable below 100 K. This contradicts the behavior of the $p$-doped high-$T_c$ materials such as $YBa_2Cu_3O_7$, which exhibit a sharp rise in $\tau_\epsilon$ only in the vicinity of their critical temperatures.

In a nonequilibrium, two-temperature system where electrons are at a temperature $T_e$ and lattice at $T_L$, an initial decay is given by the instantaneous energy relaxation time: $\tau_\epsilon=\Delta E_e/\dot{E}_e$, where $\Delta E_e=\gamma(T_e^2-T_L^2)/2$ and $\dot{E}_e$ is the rate of energy transfer from the electrons to the lattice. At temperatures much less than the Debye temperature, $\theta_D$($\approx$320 K for NCCO), it can be shown that[12,13]

$$\frac{\tau_\epsilon(T_L)}{\tau_\epsilon(T_0)_{3D}}=C_3\frac{\theta_D}{T_0}\left(\frac{\theta_D}{T_e}\right)^3\frac{1-(T_L/T_e)^2}{1-(T_L/T_e)^5},$$
$$\frac{\tau_\epsilon(T_L)}{\tau_\epsilon(T_0)_{2D}}=C_2\frac{\theta_D}{T_0}\left(\frac{\theta_D}{T_e}\right)^2\frac{1-(T_L/T_e)^2}{1-(T_L/T_e)^4}. \quad (1)$$

Here $\tau_\epsilon(T_0)$ is the electron relaxation time for a temperature, $T_0$, close to the Debye temperature, and $C_2$ and $C_3$ are constants which depend on the energy position of the averaged electrons.[13] The top equation holds for three-dimensional (3D) phonons, the lower for two-dimensional (2D) phonons. In the case of equilibrium transport, we can define $\tau_\epsilon$ as $\tau_E$, and the top equation reduces to $\tau_E(T_e)/\tau_E(T_0)_{3D}=C(\theta_D/T_0)(\theta_D/T_e)^3$ with $C=2C_3/5$.The factor $C$ is estimated to be 1/93 for an averaged thermal quasiparticle and 1/8.4 for a quasiparticle right at the Fermi surface. During the initial cooling, the lattice temperature can be approximated by the sample temperature, while the electron temperature depends on the heating pulse: $T_e=\sqrt{T_L^2+2\Delta E/\gamma}$. In our case, $\Delta E\approx0.11$ J/cm$^3$ and $\gamma\approx0.3$ mJ/cm$^3$ K$^2$.[14] We obtain the constants from $\tau_\epsilon(T_c)$ and $\tau_\epsilon$(300 K) to be $C_3\approx1/21.5$ and $C_2\approx1/2.6$, suggesting that we are indeed probing near the Fermi level. Shown in Fig. 1 are both the experimental $\tau_\epsilon$ and the relaxation time obtained from the two-temperature model for both 2D and 3D phonons. The 2D phonon calculation

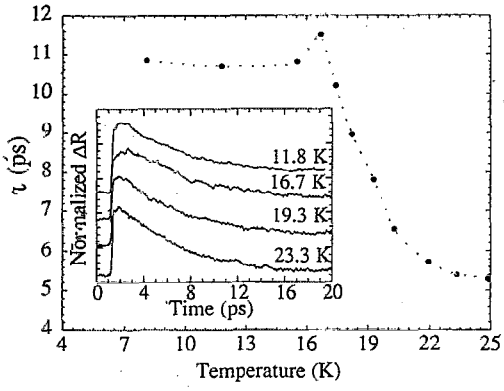


FIG. 2. Temperature-dependent relaxation time below $T_c$. The dashed line is a guide to the eye. Inset: Normalized reflectivity changes vs pump-probe delay time for $T < T_c$.

gives a better fit to $\tau_\epsilon(T)$ than that for 3D phonons, as the latter shows a much sharper temperature dependence. This may be related to the intrinsic 2D Cu-O plane in the $Nd_2CuO_4$ structure.

The inset of Fig. 2 shows the reflected signals versus pump-probe delay time in a reduced temperature range extending into the superconducting domain, while the actual change in relaxation time versus temperature (derived in the same way as before with $c_i$ fixed) is presented in the main part of Fig. 2. Several relaxation processes are possible here. It is most likely that the nonequilibrium electrons will first relax via inelastic scattering with phonons and then recombine to form paired electrons within the recombination time $\tau_r$. Close to $T_c$, the picture could be further complicated by the fact that the order parameter is very sensitive to any external perturbation. Theoretical calculations for conventional superconductors by Kaplan *et al.*[15] show that as temperature decreases, the quasiparticle recombination time increases because of the decrease in the population of quasiparticles, while the scattering time has a rather weak temperature dependence for excitation energies exceeding the gap. We believe that a complex interplay among these characteristic times is the cause of the saturation in the relaxation time observed in Fig. 2. In the absence of a detailed theoretical description, however, it is not possible to specify the role of each of the relaxation processes.

In comparison with the known results on $YBa_2Cu_3O_7$ (YBCO) and $Bi_2Sr_2Ca_2Cu_3O_{10}$ (BSCCO), the temperature dependence of the relaxation time for the NCCO well above $T_c$ is qualitatively similar, i.e., subpicosecond in duration and essentially constant. However, while $\tau_\epsilon$ in the former compounds only rises appreciably as the temperature approaches $T_c$ from above, for NCCO it begins to rise substantially above $T_c$, as indicated by the temperature dependence described in Eq. (1) and seen in Fig. 1. Furthermore, below $T_c$ the plateau in $\tau_\epsilon$ for the NCCO is roughly three to five times larger than reported results for YBCO and BSCCO. Also, while the peak in $\tau_\epsilon$ appears somewhat suppressed for the NCCO compared to other $YBa_2Cu_3O_7$ measurements,[16] it is true our NCCO sample had a rather broad transition of $\sim 3$ K, which might have led to a smearing of the divergence of $\tau_\epsilon$.[17,18]

In conclusion, femtosecond, time-resolved measurements of the normal-state, temperature-dependent electron-phonon energy relaxation times for single crystals of the electron-type copper oxide superconductor $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ have been demonstrated. A model which assumes 2D phonons has been found to provide a marginally better fit than a system of 3D phonons. Below $T_c$, the behavior of the relaxation time is likely to be the result of an interplay of a combination process with scattering and recombination of quasiparticles photogenerated by an ultrashort-pulse laser and the order parameter relaxation close to $T_c$. Further studies are planned, particularly on thin films of $Nd_{1.85}Ce_{0.15}CuO_{4-y}$, so that time-resolved transmissivity changes can be measured along with the reflectivity signals, thus conclusively determining $\Delta\epsilon_2$.

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