University of Nebraska - Lincoln
DigitalCommons@University of Nebraska - Lincoln

Si-Hwang Liou Publications | Research Papers in Physics and Astronomy

December 1990

Relaxation time of the order parameter in a high-temperature superconductor

G.L. Eesley
Department of Physics, General Motors Research Laboratories, Warren, Michigan

J. Heremans
Department of Physics, General Motors Research Laboratories, Warren, Michigan

M.S. Meyer
Department of Physics, General Motors Research Laboratories, Warren, Michigan

G.L. Doll
Department of Physics, General Motors Research Laboratories, Warren, Michigan

Sy_Hwang Liou
University of Nebraska-Lincoln, sliou@unl.edu

Follow this and additional works at: https://digitalcommons.unl.edu/physicsliou

 Part of the Physics Commons

Eesley, G.L.; Heremans, J.; Meyer, M.S.; Doll, G.L.; and Liou, Sy_Hwang, "Relaxation time of the order parameter in a high-temperature superconductor" (1990). *Si-Hwang Liou Publications*. 39.
https://digitalcommons.unl.edu/physicsliou/39

This Article is brought to you for free and open access by the Research Papers in Physics and Astronomy at DigitalCommons@University of Nebraska - Lincoln. It has been accepted for inclusion in Si-Hwang Liou Publications by an authorized administrator of DigitalCommons@University of Nebraska - Lincoln.

Relaxation Time of the Order Parameter in a High-Temperature Superconductor

G. L. Eesley, J. Heremans, M. S. Meyer, and G. L. Doll
Physics Department, General Motors Research Laboratories, Warren, Michigan 48090-9055

S. H. Liou
Behlen Laboratory of Physics and Center for Materials Research and Analysis, University of Nebraska-Lincoln, Lincoln, Nebraska 68588-0111
(Received 7 September 1990)

We present femtosecond time-resolved measurements on the high-T_c superconductor $Tl_2Ba_2Ca_2Cu_3O_{10}$. At temperatures below T_c, we observe a relaxation process which is distinct from the equilibrium of hot carriers in the normal state. Our results demonstrate an increasing relaxation rate as the superconducting gap opens. This is consistent with the behavior of conventional metallic superconductors.

PACS numbers: 74.40.+k, 73.50.Gr, 78.47.+p

Investigations of nonequilibrium superconductivity have demonstrated the important role of the order parameter in describing a superconductor perturbed by photons or injected carriers.[1] On sufficiently short time scales, these perturbations can throw the order parameter (i.e., the energy gap Δ) out of equilibrium with both the phonon and the superconducting system.[2] The return to equilibrium can be characterized by a limiting relaxation time τ_s, which is proportional to T/Δ and diverges in the vicinity of the critical temperature T_c.[3,4]

In the case of high-temperature copper-oxide superconductors, tunneling experiments have measured the presence of an energy gap $\Delta(T)$ which closes at T_c. Although the behavior is suggestive of the Bardeen-Cooper-Schrieffer (BCS) theory, the values of $2\Delta(0)/kT_c$ are several times larger than those observed in conventional metallic superconductors.[5] This and other characteristics have yet to be reconciled with an acceptable theoretical account.

Recent time-resolved optical investigations of $YBa_2Cu_3O_7$ (Refs. 6 and 7) and $Bi_2Sr_2Ca_2Cu_3O_{10}$ (Ref. 6) have revealed an abrupt change in carrier relaxation time when the samples become superconducting. Han *et al.*[7] have analyzed their results in terms of a two-fluid model which predicts a quasiparticle-density-dependent relaxation time. In this Letter, we report detailed temperature-dependent measurements of the carrier relaxation time in superconducting films of $Tl_2Ba_2Ca_2Cu_3O_{10}$. We show for the first time that in the vicinity of T_c the distinct change in relaxation rate is proportional to $\Delta(T)$ and related to the normal-carrier inelastic relaxation rate. Our results are consistent with the behavior of conventional superconductors, and indicate that phonons play a significant role.

Our measurement scheme is similar to that used for generating nonequilibrium electron-lattice temperatures in metals.[8] An ultrashort-duration light pulse is absorbed by the carriers which rapidly thermalize to a temperature in excess of the phonon system. The subsequent energy relaxation between electrons and phonons is monitored by optically probing the temperature-dependent reflectivity change as a function of time delay after the excitation pulse. In the case of a superconductor, we expect that a sufficiently fast thermo-optical perturbation will decrease the order parameter Δ by destroying a fraction of the superconducting carriers.[9]

The present measurements were performed using a tunable laser system producing repetitive 80-fs-duration light pulses at an average power of 70 mW ($\sim$0.85 nJ/pulse). The laser was tuned to a wavelength of 612 nm (2.03 eV) and the power was split to provide synchronous excitation and probe pulses in a power ratio of $\sim$10 to 1. The measurements were made with $\leq$ 1.8 mW incident power focused onto the sample surface within an $\sim$30-μm diameter. The pulses were orthogonally polarized, with the probe polarized in the plane of incidence and both pulses incident on the surface at an angle of roughly 30°. The effective time resolution and zero delay were determined by measuring the temporal cross-correlation function of the probe and excitation pulses at the sample position, yielding a full width at half maximum (FWHM) of 160 ± 20 fs.

Superconducting films were produced by magnetron sputtering from a single composite oxide target, and deposited to a thickness of $\sim$1 μm on $LaAlO_3$ substrates.[10] Subsequent annealing produced a $Tl_2Ba_2Ca_2Cu_3O_{10}$ (2:2:2:3:10) superconducting phase with an onset temperature of 120 K and zero resistance at 106 K. The crystal structure was examined by high-resolution x-ray-diffraction measurements which determined a c-axis periodicity of 3.5647 nm in a direction perpendicular to the plane of the film. The misalignment of ordered grains was examined by a rocking curve through the (0014) diffraction peak, which exhibited a FWHM $< 0.22°$.

Transient reflectivity measurements were performed at cryostat temperatures ranging from room temperature down to 80 K. Figure 1 shows a series of our measurements in the vicinity of the critical temperature. The vertical axis is the fractional reflectivity change $\Delta R/R$

© 1990 The American Physical Society

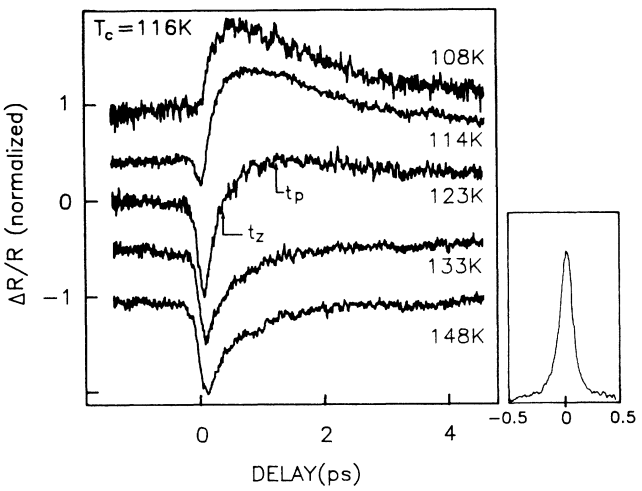

FIG. 1. Transient fractional reflectivity change vs time delay after excitation. Each transient has been normalized to its peak value ($\Delta R/R \sim 1.5\times10^{-4}$) and offset for clarity. The temperatures correspond to the illuminated sample temperature; the cryostat temperatures are, from top to bottom, 80, 86, 95, 105, and 120 K (see Ref. 15). For $T=123$ K, zero-crossing and peak-signal times are indicated by t_z and t_p, respectively. Inset: The measurement system cross correlation.

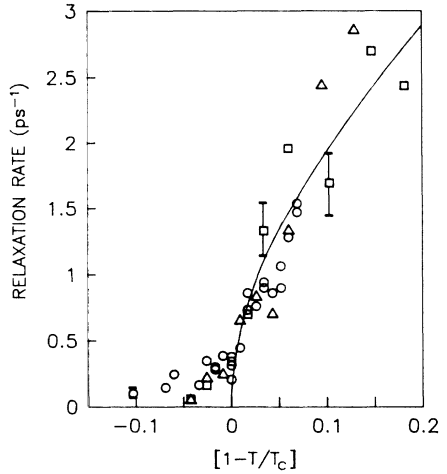

FIG. 2. Order-parameter relaxation rate τ_s^{-1} vs reduced temperature ($T_c=116$ K). Data sets marked by ○, △, and □ were obtained at power levels of P_0, $0.8P_0$, and $0.5P_0$. Typical error bars are shown, which only account for the τ_n measurement uncertainty applied to Eq. (1). The solid curve is the fit: $[\tau_s=2.2\{k_BT/\Delta(T)\}\tau_n]^{-1}$.

versus time delay of the probing pulse relative to the heating pulse. The signal levels have been normalized to their respective peak values and offset from zero for clarity.

At high temperatures the signal is characterized by a negative ΔR transient which decays within $\sim$0.5 ps. The decay time τ_n increases as temperature decreases from 300 K, although the difference between the 148- and 133-K data is less than our measurement accuracy of $\pm$40 fs. This response is typical of metallic systems in which nonequilibrium carriers cool via inelastic collisions with phonons.[11] The correspondence between the measured τ_n and the electron-phonon coupling has been developed by Allen[12] and demonstrated in a number of conventional superconductors.[13]

As the temperature is lowered into the transition region ($T\simeq123$ K), we observe the onset of a positive ΔR which appears to decay over several picoseconds. This signal exhibits increasing amplitude and *decreasing decay time* (τ_s) *as the temperature is lowered* further, consistent with the observations of Han *et al.*[7] in $YBa_2Cu_3O_7$. We believe this behavior is suggestive of the destruction of superconducting pairs during the excitation pulse, and the subsequent relaxation of nonequilibrium carriers back into the superconducting state. As the temperature is lowered further below T_c, the restoring force between carriers becomes stronger and the relaxation into the superconducting state occurs faster.[14] This trend is opposite to that of normal carriers, and it is not observed in nonsuperconducting copper oxides.[6,7]

In the vicinity of T_c, the determination of the $+\Delta R$ decay time is obscured by the presence of the normal-carrier signal ($-\Delta R$). This is not unexpected, since superconducting carriers can be destroyed by the optical "injection" of hot normal carriers which rapidly thermalize into states above the energy gap. During the rapid carrier-carrier thermalization, excess energy can excite more carriers from below the gap and lead to a nonequilibrium carrier distribution.[9] We expect that this situation is generated during the $\sim10^{-13}$-s heating pulse, and the subsequent temperature relaxation occurs via normal-carrier inelastic scattering ($-\Delta R$) and the dynamic return of the energy gap to equilibrium ($+\Delta R$).

We can model the transient reflectivity signal by the sum of two exponentials of opposite polarity, with decay times τ_s and τ_n. It is simple to show that the time delay between the signal zero crossing at t_z and the maximum at t_p (see Fig. 1) satisfies the relation

$$\ln(\tau_s/\tau_n)-(t_p-t_z)[1/\tau_n-1/\tau_s]=0. \quad (1)$$

This provides a means of determining τ_s from a measurement of τ_n and the time difference (t_p-t_z). This method is independent of the relative signal amplitudes, and does not require an absolute determination of the zero time delay between excitation and probe pulses. However, we do assume that the *normal*-carrier cooling time τ_n is approximately temperature independent in the range $0.2\geq1-T/T_c\geq0$. This is supported by the fact that we are unable to resolve any change in τ_n for temperatures 20 K in excess of the onset region.

The measurement system response (cross correlation) is deconvolved from the reflectivity transients, and by fitting the signal at 133 K, we obtain $\tau_n=0.35\pm0.04$ ps for the inelastic cooling time. It is clear, however, that

the signal contains a small contribution (~10%) with much longer decay time, possibly due to thermal diffusion out of the optical skin depth.[8]

The results of several temperature- and power-dependent[15] measurements are shown in Fig. 2, where we plot the *relaxation rate* τ_s^{-1} versus reduced temperature. A prominent feature of these data is the sudden increase in the rate at a particular temperature which falls in the superconducting onset region. We believe the discontinuity in slope is an indication of the normal-to-superconducting transition and have defined this point as occurring at T_c.

The solid curve in Fig. 2 is the predicted temperature dependence of the relaxation rate. This dependence, $\tau_s^{-1} \propto \Delta(T)/T$, was reported several years ago by Tinkham,[4] later refined by Schmid and Schon,[3] and demonstrated by Schuller and Gray in aluminum.[14,16] We have used the BCS weak-coupling $\Delta(T)/\Delta(0)$ from Ref. 5, where it was shown to agree with tunneling measurements of the gap in high-T_c materials. We believe Fig. 2 demonstrates a correlation between the carrier relaxation rate and $\Delta(T)/T$ in the vicinity of T_c. Although our data do not provide a precise confirmation of the detailed temperature dependence, they clearly demonstrate an *increasing relaxation rate as the temperature is lowered and the gap opens.* This correlation supports our contention that the measured relaxation rate of $+\Delta R/R$ corresponds to a process in which the order parameter is directly involved. A clean divergence of τ_s is not observed at T_c, and this is most likely due to sample inhomogeneities (and/or fluctuations) which smear the critical point and lead to the rather slow decrease in relaxation rate in the onset region ($T > T_c$).

In addition to predicting a $\Delta(T)$ dependence for the relaxation rate, it has been shown that the "longitudinal" relaxation time can be related to the normal inelastic relaxation time by[3]

$$\tau_s = [\pi^3/7\zeta(3)]\{k_B T/\Delta(T)\}\tau_n = 3.69\{k_B T/\Delta(T)\}\tau_n, \text{ if } h/\Delta(T)\tau_n \ll 1. \quad (2)$$

In Eq. (2), $\zeta(3)$ is the Rieman zeta function, and k_B and h are the Boltzmann and Planck constants, respectively. The solid curve in Fig. 2 results from a fit of $\tau_s = C \times \{k_B T/\Delta(T)\}\tau_n$ to our data. Using the value $\Delta(0) = 25$ meV,[17] the fit yields $C = 2.2$ as compared to 3.69 in Eq. (2). We note, however, that our measurements do not fall in the limit $h/\Delta(T)\tau \ll 1$, where relaxation broadening of energy levels is negligible.

Because the measured decay times appear to be independent of excitation intensity,[15] we are confident that the perturbation is small enough to permit comparison with the linearized treatment[3] which leads to Eq. (2). However, the absolute values for τ_s are indeed sensitive to the value used for the normal-carrier relaxation time τ_n in Eq. (1). Thus, any temperature dependence in τ_n over the range of our data (~20 K) will influence τ_s as the temperature is decreased.

An important connection exists between our measurement of the order-parameter relaxation time and the recent work of Friedl, Thomsen, and Cardona.[17] They observe anomalous linewidth broadening for specific phonon modes in $RBa_2Cu_3O_{7-\delta}$ (R = Er,Y,Dy,Tm) for $T < T_c$. The excess broadening is attributed to coupling with carriers which are intimately involved with the superconducting transition. Near $1 - T/T_c \simeq 0.04$, the observed broadening of $\Delta\nu \simeq 2$ cm^{-1} corresponds to an additional relaxation rate of $1/\tau = \pi c \Delta\nu \simeq 0.2$ ps^{-1}, which is comparable to our measured rate of ~1 ps^{-1}. It is very likely that both experiments are probing relaxation mechanisms involved with the condensation of carriers into the superconducting state. The work of Friedl, Thomsen, and Cardona[17] examines the carrier-phonon scattering by measuring the rate of change in phonon-mode occupation. On the other hand, we examine the order-parameter relaxation by measuring the rate of carrier pairing which may be mediated by phonons.

In conclusion, femtosecond time-resolved measurements on high-T_c materials permit the observation of order-parameter relaxation processes distinctly from the thermal equilibration of hot carriers in the normal state. We have shown that the temperature dependence of the order-parameter decay rate is in qualitative agreement with theory and experiments on conventional metallic superconductors. Comparison of our order-parameter relaxation rate with the anomalous phonon relaxation rate in similar materials indicates that phonons play a significant role in high-temperature superconductivity.

We thank G. P. Meisner, J. R. Smith, D. Morelli, and T. W. Capehart for helpful discussions.

[1] *Nonequilibrium Superconductivity,* edited by D. N. Langenberg and A. I. Larkin (North-Holland, New York, 1986).

[2] C. S. Owen and D. J. Scalapino, Phys. Rev. Lett. **28**, 1559 (1972).

[3] A. Schmid and G. Schon, J. Low Temp. Phys. **20**, 207 (1975).

[4] M. Tinkham, Phys. Rev. B **6**, 1747 (1972).

[5] M. Lee, D. B. Mitzi, A. Kapitulnik, and M. R. Beasley, Phys. Rev. B **39**, 801 (1989), and references therein.

[6] J. Chwalek, C. Uher, J. F. Whitaker, G. A. Mourou, J. Agostinelli, and M. Lelental, Appl. Phys. Lett. **57**, 1696 (1990).

[7] S. G. Han, K. S. Wong, Z. V. Vardeny, O. G. Symko, and G. Koren, Bull. Am. Phys. Soc. **35**, 678 (1990).

[8] G. L. Eesley, Phys. Rev. Lett. **51**, 2140 (1983); R. Schoenlein, W. Z. Lin, J. G. Fujimoto, and G. L. Eesley, Phys. Rev. Lett. **58**, 1680 (1987).

[9] W. H. Parker and W. D. Williams, Phys. Rev. Lett. **29**, 924 (1972).

[10] M. Hong, S. H. Liou, D. D. Bacon, G. S. Grader, J. Kwo, A. R. Kortan, and B. A. Davidson, Appl. Phys. Lett. **53**, 2102

(1988).

[11]We associate ΔR with the inelastic scattering of carriers near the Fermi level because we find that $\tau_n(T)$ scales approximately as the inverse resistivity (ρ) of our sample; i.e., $\tau_n(300\ \mathrm{K})/\tau_n(133\ \mathrm{K}) \simeq 0.45(\pm 0.15)$ and $[\rho(300\ \mathrm{K})/\rho(130\ \mathrm{K})]^{-1} \simeq 0.5$. This is expected in the linear regime where the Drude model of resistivity yields $\rho \sim 1/\tau \sim T$. See, for example, L. Forro, G. L. Carr, G. P. Williams, D. Mandrus, and L. Mihaly, Phys. Rev. Lett. **65**, 1941 (1990).

[12]P. B. Allen, Phys. Rev. Lett. **59**, 1460 (1987).

[13]S. D. Brorson, A. Kazeroonian, J. S. Moodera, D. W. Face, T. K. Cheng, E. P. Ippen, M. S. Dresselhaus, and G. Dresselhaus, Phys. Rev. Lett. **64**, 2172 (1990).

[14]I. Schuller and K. E. Gray, Phys. Rev. Lett. **36**, 429 (1976).

[15]Because average heating of the illuminated volume will downshift the measured T_c relative to the unperturbed sample, power-dependent measurements are essential. Of the five data sets shown in Fig. 2, three were obtained with $P_0 \simeq 1.8$ mW, one at $0.8P_0$, and one at $0.5P_0$. From the temperature shifts required to overlap the apparent T_c at different powers, we determine an average heating of 28, 23, and 13 K, respectively. The $P_0 \rightarrow 0$ extrapolated $T_c = 116$ K falls in the electrical-resistivity onset region for superconductivity. A temperature rise of $\Delta T_{av} = (1-R)P_0/Kd\sqrt{\pi} \simeq 15$ K is expected for $R = 0.1$, $d = 30$ μm, and $K \simeq 2$ W/mK. For ΔT_{av}, see J. F. Ready, *Effects of High-Power Laser Radiation* (Academic, New York, 1971), Chap. 3; for K, we use a value for crystalline $La_{1.96}Sr_{0.04}CuO_4$: D. T. Morelli, G. L. Doll, J. Heremans, M. Dresselhaus, A. Cassanho, D. Gabbe, and H. Jenssen, Phys. Rev. B **41**, 2520 (1990).

[16]I. Schuller and K. E. Gray, Solid State Commun. **23**, 337 (1977).

[17]B. Friedl, C. Thomsen, and M. Cardona, Phys. Rev. Lett. **65**, 915 (1990).