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Carrier relaxation time divergence in single and double layer cuprates

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Abstract. We report the transient optical pump-probe reflectivity measurements on single and double layer cuprate single crystals and thin films of ten different stoichiometries. We find that with sufficiently low fluence the relaxation time (τ_R) of all samples exhibits a power law divergence with temperature (T) : $\tau_R \propto T^{-3\pm0.5}$. Further, the divergence has an onset temperature above the superconducting transition temperature for all superconducting samples. Possible causes of this divergence are discussed.

PACS. 74.78.Bz High-T_c films – 74.72.-h Cuprate superconductors (high-T_c and insulating parent compounds) – 74.25.Gz Optical properties

1 Introduction

To place our experimental results in context, we provide a synopsis of what is known about the behavior of photoexcited carriers in simple metals [1–7], BCS superconductors [8–11] and cuprates [12–18]. The photoelectrons thermalize with other electrons, this process is characterized by a thermalization time (τ_{TH}). It was originally though that this process would be very fast (< 10 fs) [19] in metals since the Coulomb interaction occurs as the speed of light and, from two-photon photoemission measurements, the lifetime of electron excited $\geq$ 1 eV above the chemical potential is < 10 fs [19,20]. The initial distribution of electrons is not a Fermi-Dirac distribution, and changes quickly [21], but as the absorbed energy is redistributed among electrons a Fermi-Dirac distribution develops [21]. It was a surprise when the measured τ_{TH} for simple metals (Cu, Ag, Au) was 0.5 ps –2 ps [5,6,21]. This long τ_{TH} was attributed primarily to phase space limitations due to Fermi-Dirac statistics. The measured thermalization time in metals is almost temperature independent, but reports in the literature were not conducted at sufficiently low actual temperatures to observe any T^2 behavior characteristic of a Fermi liquid metal [1–7,22]. In superconductors, the thermalization time increases across the superconducting phase transition, a result generally attributed to phase space limitations due to a gap in the carrier excitation spectrum.

Relaxation of excited carriers, characterized by a relaxation time (τ_R) was first considered in the two-temperature model [23]. For an uncorrelated (simple) metal, the predictions were (i) $\tau_R \propto T$ for temperatures down to $T \sim \theta_{\mathrm{DEBYE}}/5$, where θ_{DEBYE} is the Debye temperature, and (ii) $\tau_R \propto T^{-3}$ for $T \ll \theta_{\mathrm{DEBYE}}/5$ [7,22]. However, experiments on simple metals [1–7], while confirming prediction (i), establish that the divergence at low

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Table 1. Experimental procedures.

Sample	$La_{2-x}Sr_xCuO_4$	La-doped $Bi_2Sr_2CuO_{6+z}$	$Bi_2Sr_2CaCu_2O_{8+\delta}$
Type	Thin film (100 nm)	Single crystal	Single crystal
Underdoped	$X = 0.06$, 0.08, 0.10		$T_C = 85$ K
Near optimally doped	$X = 0.15$		$T_C = 92$ K
Overdoped	$X = 0.25$	$T_C = 18$ K, 25 K	
Non-superconducting	$X = 0.30$		

temperatures is not observed. References [1–7] argue that the divergence is not observed because of phase space limitations on possible scattering events at low temperatures.

Both theory [8,24,25] and experiment [9–11,13,15] in BCS and cuprate superconductors report that τ_R and quasiparticle lifetime increase across the superconducting phase transition, results attributed to the same phase space limitations caused by a gap in the carrier excitation spectrum.

In BCS superconductors, the carrier relaxation time diverges as the superconducting phase transition is approached from the superconducting state [11,26–29]. The experimental data shows a change of $\sim 5x$ between $T = 0.98T_C$ and $0.995T_C$. At low temperatures, the relaxation time is, in the limit of zero fluence, expected to change as $\tau_R \propto \exp(\Delta/k_BT)$, where (Δ) is the superconducting gap and k_B is the Boltzmann constant [11,29]. In the cuprates, there have been reports arguing for a divergence in the relaxation time across T_C, although the data show a change of only $\sim 2x$ and the measurements were performed at considerably higher fluence than the lowest fluencies we report [30]. There were early hints that the relaxation time increased at low temperatures [16,18]. More recently, experimentalists have reduced the minimum fluence at which such experiments can be performed. Segre et al. [17] and Schneider et al. [31] have reported a surprising result: $\tau_R \propto T^{-N}(N \sim 3)$ at low temperatures. Reference [17] argues that the divergence is caused by the superconducting phase transition. Reference [31] provides no single explanation for the observation, which was limited to single-layer cuprate samples.

Very recently, Demsar and colleagues [22] have performed similar pump probe measurements on heavy fermion metals $YbXCu_4$ and their non-magnetic, non-heavy fermion counterparts $LuXCu_4$ (X= Ag, Cd, In). Reference [22] reports an increase of more than two orders of magnitude in τ_R below the Kondo temperature for the heavy fermion compounds, but no increase for the $LuXCu_4$ materials.

In this report, we concentrate on the relaxation time behavior of both single and double layer cuprates, including $La_{2-x}Sr_xCuO_4$, $Bi_2Sr_2CuO_{6+z}$ (La-doped) and $Bi_2Sr_2CaCu_2O_{8+\delta}$. As we establish below, we obtain three experimental results: (i) at sufficiently low fluence, both single and double layer cuprates exhibit a power law divergence in the relaxation time: $\tau_R \propto T^{-3\pm0.5}$. The divergence is observed in both superconducting and metallic, non-superconducting samples. (ii) The onset temperature of the divergence is in the normal state for all samples, leading us to argue that the divergence is a normal-state property. (iii) As we increase the fluence, we find that the relaxation time is more affected by the superconducting phase transition.

2 Experimental procedures

We reproduced the results on multiple samples of the same stoichiometry, and for multiple times on different parts of the same sample (Tab. 1). Sample preparation for $La_{2-x}Sr_xCuO_4$ thin films and La-doped $Bi_2Sr_2CuO_{6+z}$ has been described elsewhere [32–34]. Single crystals of $Bi_2Sr_2CaCu_2O_{8+\delta}$ were synthesized 'at-stoichiometry' from a modified floating-zone process at a growth rate of ~ 0.1 mm/h in a vertical infra-red image furnace, cleaved from an as-grown crystal and individually post-annealed to different levels of oxygen content (doping) [35]. We confirmed the results in four different laboratories: Vanderbilt, Rochester, William and Mary and Los Alamos, using different femtosecond Ti:sapphire laser systems [36–39]. Pump pulse energy and energy densities as low as 3×10^{-12} joule and 5×10^{-8} joule/cm^2 were used. The photodetectors (home-built) and phase sensitive detection scheme, have also been described elsewhere [39]. The noise voltage level (including electronics and scattered light) allowed us to detect voltages as small as 3×10^{-7} volts. The pulse width varied between 30 fs [38] and 260 fs [36].

To estimate the actual sample temperature, we considered the heating that a continuous wave laser with an equivalent power would produce. Some of the energy absorbed from the pump pulse does not completely leave the illuminated region in 12 ns, the time between pulses. This results in a temperature increase ($\Delta T_{\text{heating}}$) determined by the temperature of the lattice before illumination and the fluence used. We obtained spectra at several pump power levels. We also used the value of the sample superconducting transition temperature T_C, measured by susceptibility, as a benchmark temperature and confirmed our estimate of sample heating. With these checks, we were able to obtain temperature estimates to an accuracy of ±2 K (the heating effects in all the data presented here have already been accounted for).

Figure 1 shows a portion of a data set taken on La doped $Bi_2Sr_2CuO_{6+z}$ at 12 K. Note that only about 5% of the total reflectivity change occurs while the pump pulse illuminates the sample, so virtually all the reflectivity

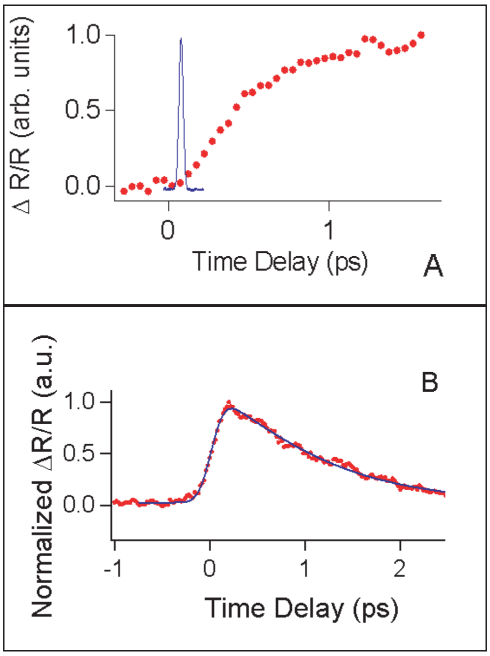

Fig. 1. A: Expanded view of transient reflectivity versus time delay, for La-doped $Bi_2Sr_2CuO_{6+z}$ single crystal sample at 12 K (closed circles). The thin line is a temporally accurate representation of the pump-probe cross correlation and represents the time that the laser pulse is incident on the sample. B: Raw data (closed circles) taken on $La_{1.7}Sr_{0.30}CuO_4$ at 60 K, thin line is our fit to the data.

change occurs after all photoelectrons are absorbed. The long rise time compared with the pump probe cross correlation allows us to accurately fit the electron thermalization time, the process of electron-electron scattering which leads to the creation of many more electron-hole pairs than are initially created by photoexcitation. Sun et al. [21] and Fann et al. [5] considered the situation where electron thermalization and relaxation via phonon emission take place at the same time. This leads to transient reflectivity response modeled by:

$$\Delta R \propto A(T)\{H(t)[1-\exp(-t/\tau_{TH})][\exp(-t/\tau_R)]\}. \quad (1)$$

Where $A(T)$ is an amplitude of the transient, $H(T)$ is the Heaviside function that accounts for the experimental resolution, τ_{TH} is the electron thermalization time constant and τ_R is the relaxation time constant.

3 Experimental results

We analyzed how τ_R depends on temperature for three different materials. In Figure 2 we show a log-log plot of τ_R versus T which displays a power law divergence: $\tau_R = CT^{-N}$ for all of the samples. Within our experimental error, $N = 3.0 \pm 0.5$ is the same for all samples, from the lowest temperature to well above T_C for the superconducting samples. The data do not lie on top of each other, indicating differences in the coefficient C. In earlier reports [30] τ_R is peaked at T_C and almost constant below T_C. Our data is not in contradiction to these earlier measurements but rather the experimental conditions are different. In particular the fluence of our current measurements is more then an order of magnitude lower. We found that the lower fluence was needed to observe a divergent relaxation time. The onset temperatures for all samples are well into normal state. return to base line even after 120 ps. Figure 3B is a plot of log of relaxation time τ_R versus the log of temperature T. The straight line is there to guide the eye and represents a power law of ∼2.6. Figure 3C shows the same data on a semilog plot comparing τ_R to $(1/T)$. The data in Figure 3C clearly do not fall on a straight line indicating that the change in relaxation time as a function of temperature is not exponential as follows from Rothwarf-Taylor equations in the low excitation limit when bi-particle recombination governs the SC state recovery [8].

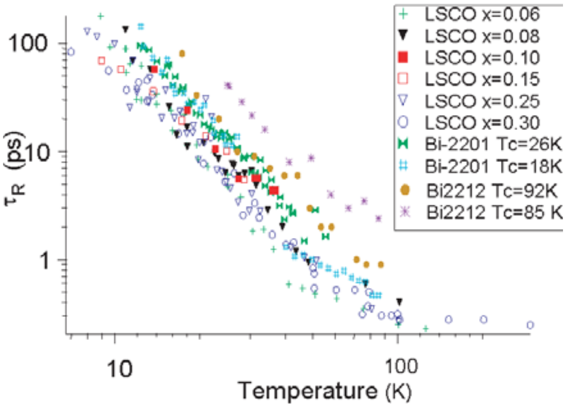

Fig. 2. Plot of ln τ_R versus ln$T$ for all ten samples.

Figure 3A shows the transient reflectivity data taken on a near optimally doped ($T_C = 92$ K) $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal. The raw data is shown without smoothing. However, some of the data are the average of up to 10 scans. The relaxation time decreases from 80 ps at 17 K to 0.8 ps at 87 K. The data are offset from one another for ease of reading. Note that the data taken with a lattice temperature of 17 K do not.

Figure 4 shows raw data taken on a superconducting, La-doped $Bi_2Sr_2CuO_{6+z}$ single crystal sample (T_C= 25 K). The relaxation time decreases from 88 ps at 12 K to 0.8 ps at 43 K. Figure 4B shows a plot of log τ_R versus log T. The straight line is there to guide the eye and represents a power law ∼3.5. Figure 4C shows a semilog plot of the same data. Note that log-log plot (power law divergence) yields a better fit than the semilog (exponential divergence) plot.

Figure 5A shows some of the raw data taken on a non-superconducting $La_{1.70}Sr_{0.30}CuO_4$ thin film sample. The relaxation time decreases from 85 ps at 7 K to 2 ps at 32 K. The inset of Figure 5A shows a log-log plot of the ab-plane conductivity σ_{ab} versus temperature. The inset of Figure 5A shows data with the residual resistivity (49 $\mu\Omega$ cm subtracted). There is no indication of percolative superconducting paths down to $T = 7$ K. The

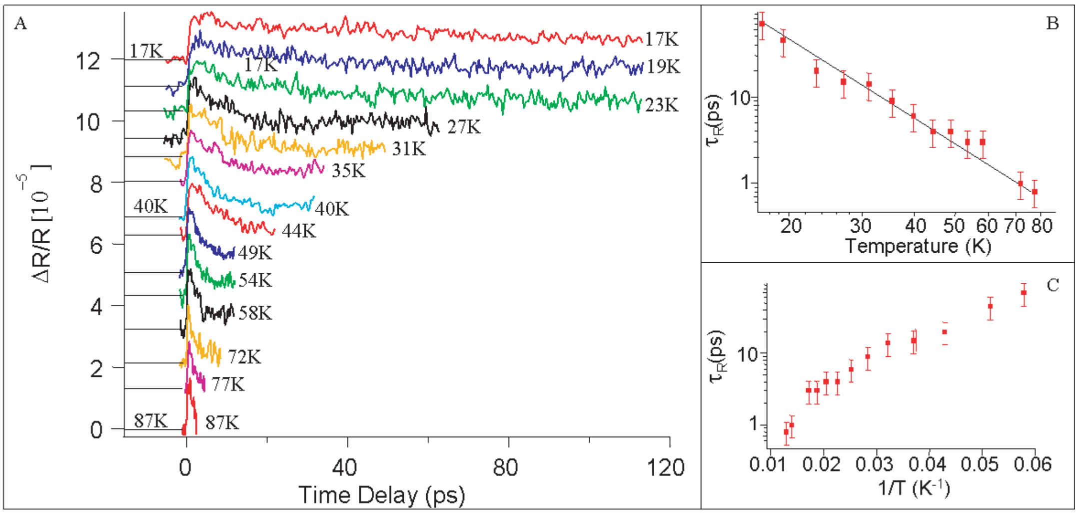

Fig. 3. A: Transient reflectivity versus time delay for $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal sample with T_C= 92 K. Lowest temperature starting at the top with temperatures to the right of each data set. B: Log-log plot of relaxation time versus temperature for $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal sample with T_C= 92 K. C: Semi-log plot of relaxation time versus $(1/T)$ for $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal sample with T_C= 92 K.

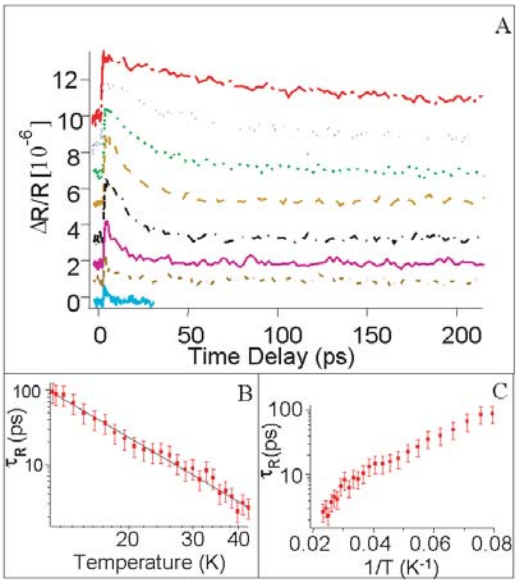

Fig. 4. A: Transient reflectivity for La-doped $Bi_2Sr_2CuO_{6+z}$ with T_C = 25 K. Temperatures starting with the top data set and going down are: 12 K, 15 K, 18 K, 22 K, 25 K, 29 K, 36 K, 43 K. B: Log-log plot of relaxation time versus temperature for La-doped $Bi_2Sr_2CuO_{6+z}$ with T_C = 25 K. C: Semi-log plot of relaxation time versus temperature for La-doped $Bi_2Sr_2CuO_{6+z}$ with T_C = 25 K.

resistivity changes only slightly, from about 53 $\mu\Omega$ cm to 64 $\mu\Omega$ cm, over the temperature range in which the relaxation time changes by a factor of 40, indicating that disorder does not play a major role on the relaxation time. It is worth noting that we have confirmed this result with multiple $La_{1.70}Sr_{0.30}CuO_4$ thin film samples.

Figure 5B is a log-log plot of τ_R versus T. The straight line is the power law $N = 2.5$ and is there to guide

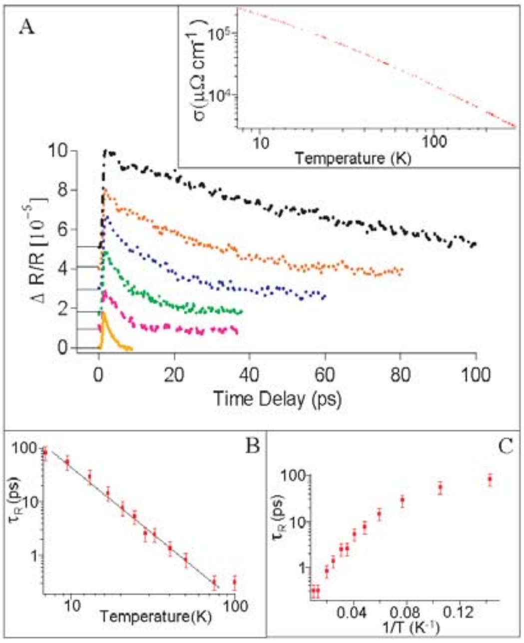

Fig. 5. A: Transient reflectivity versus time delay, for overdoped, metallic, non-superconducting $La_{1.70}Sr_{0.30}CuO_4$ thin film sample taken at temperatures noted. Inset: log-log plot of d.c. conductivity versus temperature; the residual resistivity of 49 $\mu\Omega$ cm was subtracted from the measured resistivity. Temperatures starting at the top data set and working down are; 7 K, 13 K, 17 K, 21 K, 24 K, 32 K. B: Log-log plot of relaxation time versus temperature for non-superconducting $La_{1.70}Sr_{0.30}CuO_4$.

the eye. Note that the data above ~80 K on this sample do not follow the straight line indicating an onset of ~80 K. Figure 5C is a semilog plot again indicating the non-exponential behavior of the relaxation time.

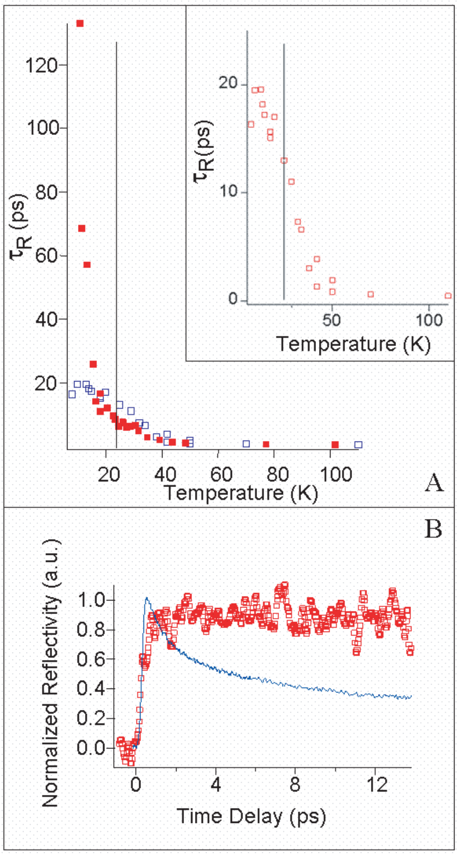

Fig. 6. A: Relaxation time versus temperature for $La_{1.92}Sr_{0.08}CuO_4$ low fluence (closed squares) and high fluence (open squares) data are shown. B: Raw data files of $La_{1.92}Sr_{0.08}CuO_4$ for the low fluence (open square) and the high fluence (solid line) both at their lowest temperatures.

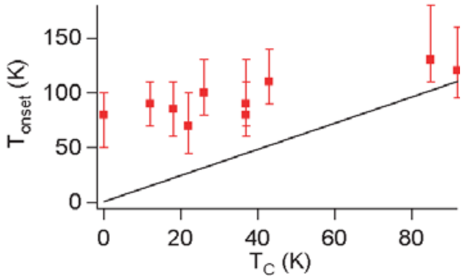

Fig. 7. Onset temperature versus superconducting transition temperature. The straight line is $T_{\rm ONSET} = T_C$.

Figure 6A shows a comparison of the relaxation time versus temperature for an $La_{1.92}Sr_{0.08}CuO_4$ sample with data taken at perturbative fluence (5 μJ/cm^2) and at low fluence (50 nJ/cm^2). At a perturbative fluence, the effect of the superconducting phase transition at 24 K (solid vertical line) shows up clearly, as emphasized in the inset. Figure 6B shows the raw data taken at the lowest temperatures accessible for the two fluencies. There is a clear difference in the relaxation time between the lowest fluence (133 ps at 10 K) and perturbative fluence (19 ps at 10 K). Both data sets show the same relaxation times at high temperatures, well into the normal state. However, note that at low temperatures, the relaxation time becomes very long when the fluence is reduced. For such low fluencies, the change across T_C is a small effect compared to the seeming divergence at low temperatures.

To understand the physics of this relaxation time divergence, it is important to compare the onset temperature of the divergence to the value of T_C. Figure 6 establishes that at high temperatures, the relaxation time is independent of fluence. For samples such as shown in Figure 6, we obtained the onset temperature directly from the data. However, there are samples for which we could not get a signal (using the lowest fluence) up to high temperatures. For such samples, we measured the relaxation time at high temperatures using a perturbative fluence, measured the divergence at low temperatures using the lowest fluencies, and extrapolated the relaxation time from low to high temperatures. We define the temperature at which the relaxation time power law divergence intersects the high temperature values as the onset temperature ($T_{\rm ONSET}$). Figure 7 shows ($T_{\rm ONSET}$) versus T_C. The straight line is the $T_{\rm ONSET} = T_C$ line. The error bars reflect the uncertainty in the extrapolation and in the exact power law of the divergence. Figure 7 establishes several significant points: i) $T_{\rm ONSET} > T_C$ for all samples; ii) the difference between $T_{\rm ONSET}$ and T_C becomes less as T_C increases. The difference ranges from 80 K ($T_C = 0$ K) to 28 K ($T_C = 92$ K); iii) including the error bars, $T_{\rm ONSET}$ and T_C are indistinguishable for the BSCCO-2212 optimally doped sample ($T_C = 92$ K).

4 Discussion

We considered several explanations of the measured relaxation time divergence.

4.1 Heavy fermion analog;
4.2 The opening of the superconducting gap;
4.3 Traditional two temperature model;
4.4 Changes in electron-phonon scattering processes with fluence;
4.5 Spatially inhomogeneous samples.

4.1 Heavy fermion analog

As noted in the introduction, heavy fermion systems exhibiting a Kondo temperature also exhibit a marked increase in τ_R at low temperatures, while the nonmagnetic analogous materials exhibit no such increase. Reference [22] provides both data and analysis that the increase in τ_R arises from the increase in the electronic heat capacity below the Kondo temperature and the strongly suppressed electron-phonon scattering. For cuprate samples, however, the electronic heat capacity versus temperature is known [40] and does not display a marked increase below the $T_{\rm ONSET}$ (Fig. 5). So this basis for expecting τ_R to increase at low temperatures does not exist for the cuprates. What the heavy fermion report [22] does demonstrate is that the relaxation time divergence is observed in correlated electron materials other than the cuprates.

4.2 Opening of the superconducting gap

We considered the possibility that the relaxation time divergences arises due to the presence of the superconducting gap. As we have mentioned, there is an increase in quasiparticle relaxation reported in different cuprates across T_C [41,42]. The maximum lifetime reported at low temperatures for $YBa_2Cu_3O_x$, ~140 ps [41], is comparable to the maximum lifetime we have measured (180 ps; Fig. 4). Further, Quinlan et al. [42] predict a $\tau_R \propto T^{-3}$ behavior, due to scattering off spin fluctuations, at $T \leq 0.25 T_C$, consistent with our data. However, we have seen a power law divergence in non-superconducting samples, at temperatures well above T_C (Fig 7) as well as in both highly underdoped and highly overdoped samples. These results are difficult to reconcile with scattering off spin fluctuations or with the superconducting phase transition as the main effect causing the observed divergence. As Figures 3C, 4C and 5C establish, the relaxation time divergence is not accounted for by a uniform BCS gap.

4.3 Traditional two temperature model

The two temperature model [7,23] predicts $\tau_R \propto T^{-3}$, at low temperatures in simple metals. However, we also considered that no study of simple metals in the literature reported such a divergence. To investigate the issue experimentally, we measured the relaxation time using a ~7 nm thick gold (Au) film and the same fluence used for cuprate samples. Our work confirms earlier work reported in reference [7] taken at higher fluence. Figure 8 shows a series of spectra taken at sample temperatures between 10 K –280 K. We used probe pulses with a width of 30 fs, so the relaxation time in the raw data is accurate.

Unlike the cuprate data, there is no indication of a divergence in the relaxation time down to temperatures ~10 K. The experimental comparison suggests that the two temperature model, which was developed for uncorrelated metals, is not applicable even at the low fluencies we used. Earlier studies of simple metals [5,6] argued that the relaxation time vs. temperature was affected by the incomplete thermalization of the photoelectrons. However, this argument does not work for cuprates at low temperatures, where the thermalization time (< 50 fs for metallic samples, 1–5 ps for superconducting samples) are much less than the relaxation times (as long as 180 ps). It is also quite difficult to explain, within the context of the two temperature model, why we measure the same relaxation dynamics for samples in the superconducting state and for metallic, non-superconducting samples. Finally, it is difficult to reconcile the two temperature model to the behavior of the relaxation time with fluence (Fig. 6) since for perturbative fluence the relaxation time depends only on the electron-phonon coupling constant, which does *not* depend on photon fluence.

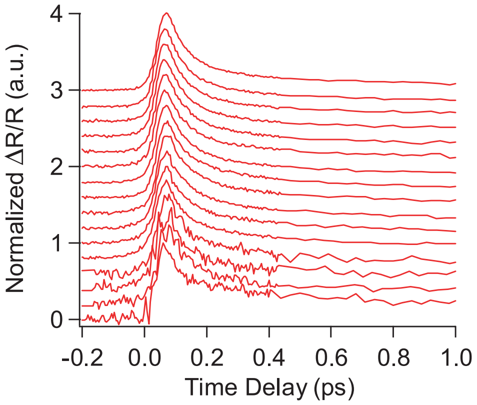

Fig. 8. Series of reflectivity spectra taken on 7 nm thick gold film at temperatures (from bottom to top) of 4, 20, 30, 50, 60, 80, 100, 120, 140, 160, 180, 200, 220, 240, 260, and 280 K. The 4–50 K data were taken at 5 times lower fluence.

4.4 Changes in electron-phonon scattering processes with fluence

In work beginning in the late 1960's, scientists studied electrical resistivity measurements of very pure metallic samples [43,44]. These early studies relied on low-energy methods such as the radio-frequency size effect (RFSE). They developed a model for electron-phonon scattering that included both emission and absorption of phonons [43,44]. In the Debye approximation, this model predicted an electron relaxation time proportional to T^{-3} [45]. Studies of the relaxation time in metals at low temperature ($\leq$ 10 K) date to pioneering work by Haberland and Shiffman [43] and Gantmakher and Leonov [44], who reported finding an electron scattering time $\propto T^{-2}$ to T^{-3}. This is remarkably similar to our results. Later Chambers outlined the theory for the radio-frequency size effect [46] and concluded that a T^{-3} dependence was expected because Gantmakher's measurements involved all scattering angles. Chambers contrasted this T^{-3} behavior with electrical resistivity, which is dominated by large angle scattering due to the $(1 - \cos\theta)$ weighting factor, which leads to a different temperature dependence. We have compared the transport relaxation time (Fig. 5A inset) with our data and do find that they exhibit different temperature dependences.

The question at issue is whether these earlier reports relate to the energy relaxation that we measure. The photon energy used is very low ($\leq 4 \times 10^{-3}$ meV) compared to the change in the Fermi-Dirac distribution due to the light absorbed in our experiment ($\leq$ 1 meV). This strongly suggests that the RFSE measurements is more a measure of momentum relaxation than of energy relaxation.

Another argument against this explanation is based on estimating the fraction of carriers in the thermalized electron group that relaxes through dissipation. We compare the fraction at the lowest fluence and temperatures used (where a divergence is observed) to higher but still perturbative fluence (where no fluence is observed). Assuming that 10–30% of the light is absorbed, the density of electronic states is ~1 electron/eV, and that the increase in electron temperature is ∼3 K, the fraction of carriers in the 300 meV wide conduction band in the thermally excited electron subsystem ranges from 8×10^{-5} (divergence observed) to 1.6×10^{-2} (no divergence measured).

We conclude, tentatively, that the scattering processes are similar over this fluence range, and that there should be no significant change in scattering with fluence. A realistic theoretical calculation is needed to definitively rule out this possibility.

4.5 Spatially inhomogeneous samples

We also considered the possibility that the samples are intrinsically inhomogeneous. This is by itself plausible, given the recent reports by scanning tunneling microscopy (STM) [47] and other methods [47,48]. There are no calculations on this possibility, so we can provide only qualitative arguments.

STM data indicate that at least some cuprate samples have two types of regions that exhibit a difference in the joint density of electronic states. One exhibits a superconducting state gap at low temperatures and the other (termed the β-region in the literature) displays STS spectra similar to the normal-state vortex region when a magnetic field above H_{c1} is applied [49]. Carriers excited in the two regions should then exhibit considerably different relaxation times, faster in the metallic regions. This is qualitatively consistent with the data of Figure 6, showing two different relaxation time responses. It should be noted that the metallic regions in which we expect a faster response are also the regions where a superconducting gap is seen to have opened. In the β regions we do not expect to see the opening of a gap. We therefore speculate that these regions may exhibit the long relaxation time. However, it requires detailed and careful calculations to establish whether the dominant response at the lowest fluence should occur in the β-regions. If this premise is confirmed by detailed calculations, the idea would be qualitatively consistent with the argument that as the fluence increases, the additional signal will come predominantly from the superconducting regions, so at somewhat higher fluence there will be a marked change in the relaxation time across the superconducting phase transition while at the lowest fluence the signal is only slightly affected by the phase transition.

It is also important to consider this idea and whether it can be consistent with a power law temperature divergence. If the β-regions are semiconducting (unlikely but still not established), the divergence should exhibit an exponential temperature dependence, in contrast to the data. If the β-regions are a simple metal, then the results on metallic, overdoped, non-superconducting samples (Fig. 5) become inexplicable. The STM results indicate that the β-regions exhibit a non-zero density of states at the chemical potential, so determining relaxation behavior in such regions requires detailed calculations not currently available. If the reflectivity signal comes predominantly from the β-regions at lowest fluencies, this would account for the insensitivity of such data to the superconducting phase transition. At higher fluence, the predominant signal arises from the regions that exhibit a superconducting gap, and this is when we clearly see the effect of the superconducting phase transition. These data also indicate that even the overdoped, metallic, non-superconducting samples exhibit this relaxation time divergence, and thus are spatially inhomogeneous. This idea also accounts for how widespread the divergence is in the cuprates. Including other reports [16–18,31], there are indications or definitive measurements of a relaxation time divergence in YBCO, BSCCO-2201, BSCCO-2212, LSCO, Tl-2201 and Hg-1223.

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