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Photoinduced Changes of Reflectivity in Single Crystals of $YBa_2Cu_3O_{6.5}$ (Ortho II)

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We report measurements of the photoinduced change in reflectivity of an untwinned single crystal of $YBa_2Cu_3O_{6.5}$ in the ortho II structure. The decay rate of the transient change in reflectivity is found to decrease rapidly with decreasing temperature and, below T_c, with decreasing laser intensity. We interpret the decay as a process of thermalization of antinodal quasiparticles, with a rate determined by inelastic scattering of quasiparticle pairs.

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The desire to understand the pseudogap phenomenon continues to dominate research in the high-T_c cuprates. At issue is the nature of the partial gap that onsets well above T_c in underdoped materials. The gap is likely to be a manifestation of either fluctuating or long-range order. A key question is whether this order competes with or enhances superconductivity, or is, in fact, a fluctuating form of the superconducting phase itself.

One approach to answering this question is to study the dynamics of nonequilibrium quasiparticles, which can be extremely sensitive to a gap [1,2]. A promising technique to study such dynamics is optical pump and probe spectroscopy. In this type of experiment a pulse of light stimulates a nonequilibrium state with excess energy and quasiparticle density. A weaker pulse, which is delayed relative to the pump, probes the nonequilibrium state by detecting changes in the absorptance or reflectance. Dynamics can be measured with subpicosecond time resolution while anisotropy can be explored by varying the polarization of the pump and probe beams.

In this paper we report pump and probe measurements performed on an untwinned single crystal of $YBa_2Cu_3O_{6.5}$ (Ortho II). Together with $YBa_2Cu_4O_8$ it is one of two underdoped materials in which doping does not introduce disorder. We find, in contrast with an earlier report [3] on underdoped films, that the decay rate of the photoinduced change in reflectivity, $\Delta R/R$, depends strongly on the excitation density and temperature. Because it vanishes in the limit that both tend to zero, we interpret the decay rate as an inelastic scattering rate of quasiparticle pairs. We present evidence that the decay corresponds to a process of thermalization, rather than recombination, of the nonequilibrium quasiparticle population. The dynamics of thermalization change suddenly upon crossing from the pseudogap regime to the superconducting state.

We measured the decay of the $\Delta R/R$ at a photon energy of 1.5 eV due to excitation with photons of the same energy. The optical pulses are produced by a mode-locked Ti:sapphire laser, and have duration 100 fs, pulse to pulse separation 12 ns, and center wavelength 800 nm. Both the pump and probe beams were focused onto the sample with a 20 cm focal length lens, yielding a spot size with 90% of its intensity within a 100 μm diameter. A photoelastic modulator varied the pump intensity at 100 kHz and a vibrating mirror oscillating at 40 Hz varied the time delay between pump and probe. This double modulation provides sensitivity to $\Delta R/R$ of 10^{-7}.

Figure 1a shows $\Delta R/R$ due to absorption of a pump pulse of energy 0.07 nJ vs time (t), at several temperatures. These reflectivity transients were measured with pump and probe polarization aligned perpendicular to the chain axis. The amplitude of $\Delta R/R$ increases and the decay rate decreases as the temperature is lowered. Figure 1b shows the temperature dependence of the maximum amplitude $(\Delta R/R)_0$. The reflectivity change is smaller when measured with pump and probe parallel to the chains, particularly above T_c.

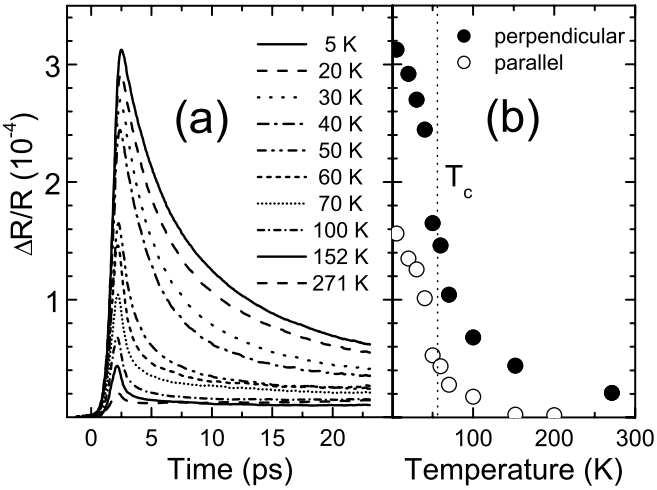

FIG. 1. (a) $\Delta R/R$ vs t at constant pump intensity for different temperatures (pump and probe both polarized perpendicular to chains). (b) Maximum amplitude of $\Delta R/R$ vs T for polarization parallel and perpendicular to the chains.

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The T dependence shown in Fig. 1 is consistent with reports of $\Delta R/R$ in underdoped (twinned) thin films excited at 1.5 eV and probed at either near-infrared [1] or far-infrared [4] frequencies. As emphasized in Ref. [4], the increase of $\Delta R/R$ with decreasing T tracks the growth of a spin resonance peak seen by neutron scattering [5,6] and a gaplike feature seen in optical conductivity [7,8]. Furthermore, $\Delta R/R$ shows the same behavior with varying carrier concentration as do these spectroscopic features, namely abrupt onset at T_c for near optimal doping [9] and a slow onset above T_c for underdoped materials [1,4]. Such considerations suggest that 1.5 eV photons ultimately generate low-energy excitations, despite the fact that they initially create an electron-hole pair whose energy is much larger than the maximum gap Δ_0.

It is generally agreed that the generation of low-energy quasiparticles occurs via a rapid cascade process, in which the parent electron-hole pair creates a large number of excitations at much lower energy. What has remained less clear is the mechanism by which this nonequilibrium population of quasiparticles causes a change in reflectivity in the visible region of the spectrum. However, recent measurements [4,10] point to a mechanism for changes in reflectivity at 1.5 eV.

Measurements of the change in THz conductivity of an underdoped YBCO ($YBa_2Cu_3O_{6+x}$) film show that photoexcitation at 1.5 eV removes spectral weight from the condensate δ function [10]. Far-IR measurements [4] reveal that this spectral weight reappears at frequencies above the maximum superconducting gap, or $\sim$100 meV. The Kramers-Kronig relations dictate that this redistribution of spectral weight changes the real part of the dielectric function at much higher frequencies (ω) according to $\Delta\epsilon_1(\omega) = -(8/\omega^4)\Sigma$, where $\Sigma = \int d\omega' \, \Delta\sigma_1(\omega')\omega'^2$ and $\Delta\sigma_1$ is the change in the real part of the optical conductivity. The change in reflectivity at 1.5 eV, $\Delta R/R = -0.225\Delta\epsilon_1$, can be obtained from literature [11] values for the dielectric function, $\epsilon_1 = 1.1$ and $\epsilon_2 = 2.7$.

The rate of increase of the photoinduced reflectivity with increasing pump intensity provides a quantitative test of the link between $\Delta R/R$ and Σ. According to the mechanism described above, ΔR reaches an upper limit, $\Delta R_{\rm sat}$, when photoexcitation shifts the entire condensate spectral weight to nonzero frequency. Figure 2 shows $(\Delta R/R)_0$ at 15 K vs $u \equiv \lambda E/4\pi r_0^2$, the average energy density deposited by the pump in the volume that is probed ($\lambda = 1.3 \times 10^{-5}$ Å is the penetration depth of the pump beam, E is the energy of the pump pulse, and $r_0 = 42$ μm is the $1/e$ radius of the laser intensity). There is a clear tendency toward saturation evident in the data, which allows us to estimate $\Delta R_{\rm sat}$. The dashed line shows the best fit to a simple saturation model, $\Delta R_0/\Delta R_{\rm sat} = [\lambda u(0)]^{-1} \int dz(1 - e^{-u(z)/u_{\rm sat}})u(z)$, where $u(z)$ is the average energy density at a depth z below the sample surface. The best fit parameters are $(\Delta R/R)_{\rm sat} = 1.1 \times 10^{-3}$ and $u_{\rm sat} = 0.43$ J/cm^3.

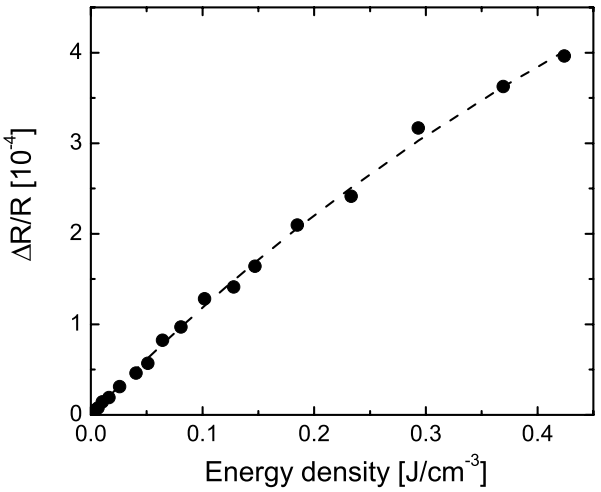

FIG. 2. $\Delta R/R$ vs excitation density at 15 K. The dashed line is the best fit to the saturation model described in the text.

We now compare the above value of $\Delta R_{\rm sat}$ with that expected if all the condensate spectral weight is shifted away from $\omega = 0$. The value of $\Delta\epsilon_1$ at saturation is $\omega_p^2\langle\omega^2\rangle/\omega^4$, where ω_p is the plasma frequency corresponding to the condensate spectral weight and $\langle\omega^2\rangle$ is the mean square frequency shift. Using $\omega_p = 1.5$ eV [12] and $\Delta\epsilon_{1\rm sat} = 4.9 \times 10^{-3}$, we obtain $\langle\omega^2\rangle = (130 \text{ meV})^2$. That this frequency shift is comparable to that directly observed by Kaindl *et al.* [4] strongly supports our interpretation of the photoinduced change in reflectivity.

We now turn to the rate of decay of $\Delta R/R$ measured as a function of T and pump intensity I. We have seen in Fig. 1 that the decay of $\Delta R/R$ becomes slower as T decreases. Figure 3a, a plot of the reflectivity change at 15 K created by pulses of varying intensity, shows that the decay rate also slows as the excitation density decreases.

The decays shown in Fig. 3a are nonexponential in that the instantaneous rate of decay, $\gamma(t) \equiv -d[\ln(\Delta R)]/dt$, decreases as t increases. To characterize the intensity dependence of the decay rate in a relatively simple way, we focus on the initial decay rate, or $\gamma(0)$. We estimate $\gamma(0)$ from the slope of $\Delta R(t)/R$ between 100% and 75% of $(\Delta R/R)_0$, normalized by the average value of $\Delta R(t)/R$ in that interval. Given our time resolution, this procedure provides a good estimate of $\gamma(0)$ for rates slower than $\sim$1 ps^{-1}. Figure 3b shows $\gamma(0)$ as a function of $(\Delta R/R)_0$, obtained by analysis of the curves similar to the ones in Fig. 3a. $\gamma(0)$ varies linearly with $(\Delta R/R)_0$ and extrapolates to a nonzero intercept as the excitation density approaches zero.

The dependence of $\gamma(0)$ on excitation density is consistent with two-particle kinetics, for example, a process in which a pair of quasiparticles scatter off of each other. For high excitation densities the rate of such scattering is proportional to the nonequilibrium density. As the excitation density decreases, becoming smaller than the thermal population, $\gamma(0)$ approaches the equilibrium scattering rate at that temperature [13,14].

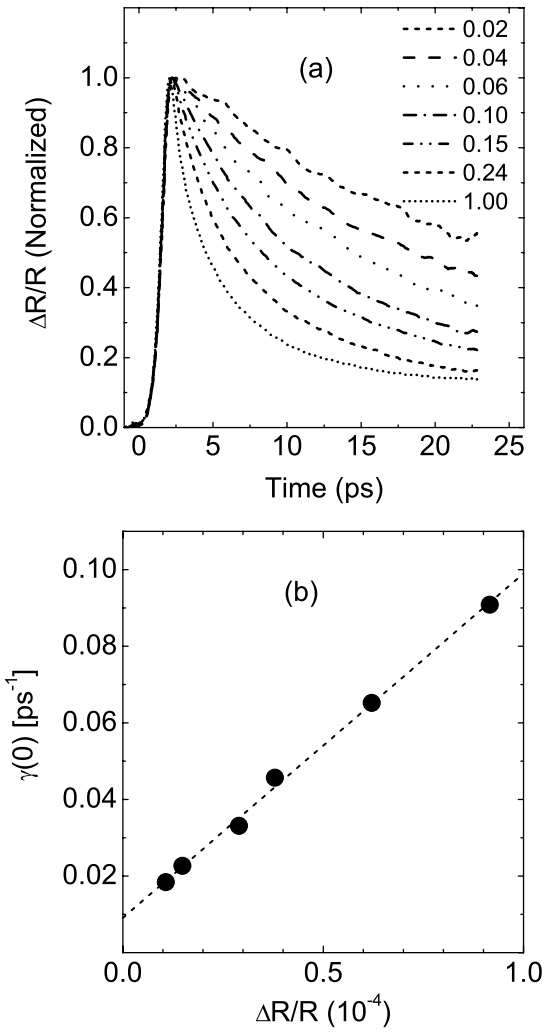

FIG. 3. (a) Decay of $\Delta R/R$ for several pump intensities at 15 K. (b) Initial decay rate $\gamma(0)$ vs $(\Delta R/R)_0$.

Figure 4 presents the temperature dependence of the decay rate for a range of intensities, in the form of a logarithmic plot of $\gamma(0)$ vs T. Points with the same (open) symbol show $\gamma(0)$ measured at the same excitation density. The data sets coincide at high temperatures but diverge at $T_c = 57$ K. This indicates that the dependence of $\gamma(0)$ on I starts abruptly upon crossing from the pseudogap to the superconducting state. The solid symbols show the decay rate in the limit of zero excitation density, γ_{th}, as determined from the intercept of plots such as in Fig. 3b. The same quantity is plotted on linear axes in the inset. For $10 < T < 40$ K, γ_{th} appears to vary as T^3, although given the experimental uncertainty an activated form cannot be excluded.

The decay rates shown in Fig. 4 were measured with the probe beam polarized perpendicular to the chains. We find that the rates measured with the probe polarized parallel to the chains are indistinguishable, within the sensitivity of our measurement, for all T and I. The absence of polarization dependence suggests that the carrier relaxation processes probed in this experiment take place in the CuO plane and not in the chain layers. Given this observation,

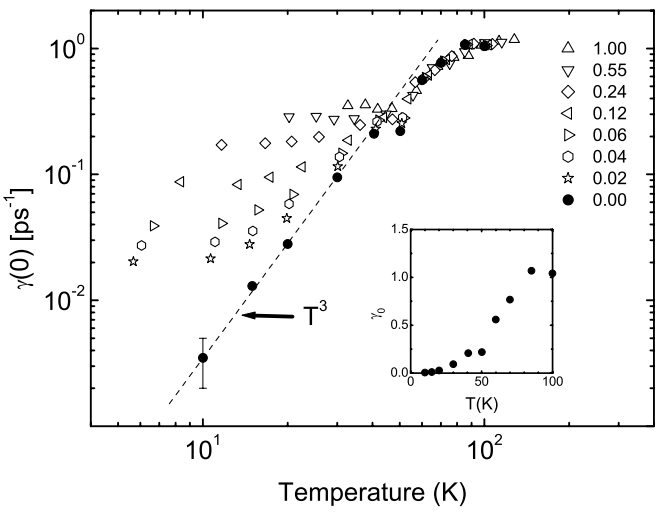

FIG. 4. Initial decay rate $\gamma(0)$ vs T at fixed I (open symbols). Decay rate in low I limit, γ_{th}, vs T (solid symbols in main panel and inset).

we speculate that the difference in $\Delta R/R$ for the orthogonal polarizations (Fig. 1b) is due to anisotropy in the spectral weight shifted by the nonequilibrium quasiparticles.

Several aspects of the decay rate of $\Delta R/R$ suggest a strong connection to quasiparticle dynamics and the density of states. For example, the decay rate appears sensitive to the opening of the pseudogap, decreasing near T^* in a manner which is remarkably similar to the nuclear spin relaxation rate in underdoped YBCO [15]. The onset of superconductivity also affects the decay dynamics—the decay rate depends on excitation density only below T_c. Finally the T^3 dependence of γ_{th} is consistent with a prediction for the recombination and scattering rate of nodal quasiparticles [16].

The simplest interpretation of the connection between $\gamma(0)$ and quasiparticle dynamics is that $\Delta R(t)/R$ reflects the decay of the nonequilibrium quasiparticle density. According to this view, $\gamma(0)$ is the rate at which quasiparticle pairs scatter into the condensate and γ_{th} is its value in thermal equilibrium. However, this description of the decay of $\Delta R/R$ cannot be complete because for quasiparticles to scatter into the condensate they must shed their excess energy. In the model of Ref. [16] the excess energy appears as a spin fluctuation, which is built from particle-hole excitations of the same band of electrons that form the condensate. Therefore the "recombination" process essentially scatters one pair into another, with no reduction in the number or energy density. This is analogous to a problem in the interpretation of transport experiments, where quasiparticle scattering alone cannot reduce the total momentum [17].

The actual recombination rate, in the sense of recovery of the equilibrium quasiparticle density, would likely be much slower than the quasiparticle scattering described above. Decay of the quasiparticle density requires that excess energy flow irreversibly away from the system of superconducting electrons to the lattice vibrations [18]. We

expect such a process to be extremely slow on the time scale of quasiparticle scattering and to exhibit single rather than two-particle kinetics. This expectation is based on the physics of the "phonon bottleneck" in s-wave superconductors [13,14], an effect which may be even more restrictive in d-wave superconductors.

The bottleneck arises because the process in which a quasiparticle pair emits a phonon and scatters into the condensate is reversible—the emitted phonon can immediately regenerate the quasiparticle pair. The quasiparticle density relaxes at the rate at which the typical phonon energy decreases below the threshold for pair creation. Not only is this rate slower than the quasiparticle scattering rate, it is also approximately independent of the excess quasiparticle density. Experiments with pulsed photoexcitation have provided convincing evidence that the relaxation of the quasiparticle density in Pb films, for example, is limited by the phonon bottleneck [19,20].

The relaxation of the quasiparticle density may be even slower in the cuprates than in conventional superconductors. In s-wave superconductors the phonons decouple from the electrons when their energy becomes less than 2Δ. In the d-wave superconductors, where there is no threshold for pair creation, the typical phonon energy must degrade to $\sim k_B T$ in order for the electronic system to reach equilibrium. The extremely weak coupling between nodal quasiparticles and phonons compounds this effect. The decay of a quasiparticle pair into an acoustic phonon is kinematically forbidden because the quasiparticle velocity is larger than the sound velocity [18].

If relaxation of the nonequilibrium density is extremely slow, why does $\Delta R/R$ decay on a picosecond time scale? The remaining possibility is that the decay corresponds to thermalization, rather than recombination. If $\Delta R/R$ is not a direct measure of the quasiparticle density, but proportional instead to Σ as we have argued previously, it can decrease during thermalization, despite a constant, or even increasing, density of quasiparticles. Microwave measurements show that Σ for nodal particles is small, because their spectral weight is confined to a very narrow (~ 10 μeV) Drude peak [21]. The ~ 100 meV spectral weight shifts [4], and consequently large values of Σ, are likely due to the antinodal quasiparticles. These considerations suggest that the decay of $\Delta R/R$ measured at 1.5 eV tracks the conversion of quasiparticles from a nonthermal distribution, which has substantial Σ, to a quasithermal distribution in which Σ is a factor 10^8 smaller. Measurements in which the nonequilibrium state is probed in the frequency range of the Drude conductivity [18] show much longer decay times (typically milliseconds), consistent with an experiment which selectively probes the nodal quasiparticle density.

In summary, we present measurements of the photoinduced change in reflectivity in an underdoped crystal of YBCO, and discuss a new perspective on this effect. We interpret the decay rate of $\Delta R/R$ as the inelastic scattering rate of quasiparticle pairs, which sets the time scale for thermalization of the nonequilibrium population. The thermalization rate decreases with the development of the pseudogap, becomes excitation density dependent in the superconducting state, and decreases rapidly with decreasing T in the low excitation limit. Coordinated measurements of photoinduced changes in optical response over a broad spectral range, from terahertz to visible frequencies, are needed to test this perspective.

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