Nonequilibrium Superconductivity and Quasiparticle Dynamics Studied by Photoinduced Activation of mm-Wave Absorption

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We present a study of nonequilibrium superconductivity in $DyBa_2Cu_3O_{7-\delta}$ using photoinduced activation of mm-wave absorption. We monitor the time evolution of the thin film transmissivity at 5 cm^{-1} subject to pulsed infrared radiation. In addition to a positive bolometric signal we observe a second, faster, decay with a sign opposite to the bolometric signal for $T > 40$ K. We attribute this to the unusual properties of quasiparticles residing near the nodes of an unconventional superconductor, resulting in a strong enhancement of the recombination time. [S0031-9007(97)04710-8]

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The occurrence of zeros in the superconducting gap for certain values of the momentum $\hbar\vec{k}$ at the Fermi surface of high T_c superconductors has a number of intriguing consequences for the dynamical behavior and lifetime of the quasiparticles at low temperatures, which has only recently begun to attract the attention of researchers in the field. Because of the presence of these zeros (or nodes) the reduction in the superfluid fraction (ρ_s) [1,2] and specific heat [3] is proportional to $H^{1/2}$, where H is the magnetic field. Also, a strong reduction of the quasiparticle scattering rate ($1/\tau$) below T_c [4–6] provides evidence that the dominant scattering mechanism has an electronic signature.

In this Letter we present a study of the quasiparticle dynamics using photoinduced activation of mm-wave absorption (PIAMA). In this pump/probe experiment we use a free electron laser [7] (FELIX) which is continuously tunable from 100 to 2000 cm^{-1} as a pump to create a temporary excited state of a superconductor. FELIX produces macropulses with a stepwise "off-on-off" intensity profile ("on" for $3 < t < 7$ μs in Fig. 1), consisting of 5000 micropulses (1–5 ps). The step response of the complex dielectric constant is monitored at 5 cm^{-1} using the combination of a backward wave oscillator (BWO) and a fast waveguide diode detector as a probe to measure the transmission through a superconducting film as a function of time. The mm-wave detector circuit was selected as a compromise between sensitivity and speed of detection, resulting in an overall time resolution of 1 μs. This choice of experimental parameters is optimal for the detection of small changes induced in the dielectric function by the infrared (IR) pulse at, as we will see, the scale of the lifetime of nonequilibrium superconductivity in high T_c's.

We used films of $DyBa_2Cu_3O_{7-\delta}$ which were prepared by rf sputtering on $LaAlO_3$ substrates. The film thickness was 20 nm and T_c was 88 K. Optimal surface quality was obtained by using Dy instead of Y. This substitution does not affect the superconducting properties. A detailed description of the preparation, characterization and the mm-wave dielectric properties of these films has been given elsewhere [8,9].

The $LaAlO_3$ substrate supporting the film is plan parallel, with a thickness $D = 0.054$ cm and a refractive index $n = 4.70$. At $k/2\pi \approx 5\,cm^{-1}$, which is our probe frequency, the dielectric function of the film $|\epsilon|$ ranges from 10^4 to 10^6 depending on temperature, while $(kd)^{-2} \approx 3 \times 10^8$. Hence the films are optically thin and $(kd)^{-2} \gg |\epsilon| \gg 1$. In that limit the Fresnel expression for transmission through the film/substrate system is

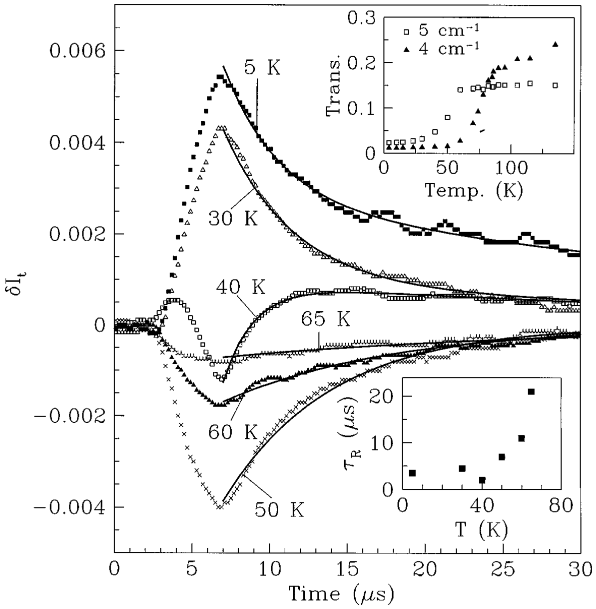

FIG. 1. Change in transmission, δI_t for $DyBa_2Cu_3O_{7-\delta}$ on $LaAlO_3$, shown for several temperatures. The far-infrared pulse enhances transmission at low temperatures, while it reduces it at temperatures higher than 40 K. The exponential fits are shown as the solid lines. Inset, upper right corner: temperature dependence of the unperturbed transmission at 4 and 5 cm^{-1}. Inset, lower right corner: Temperature dependence of the faster relaxation time, τ_R.

$$I_t = \left|(2 - ikd\epsilon)\frac{\cos\psi}{2} - (i + in^2 + kd\epsilon)\frac{\sin\psi}{2n}\right|^{-2}, \tag{1}$$

where $\psi = nkD$. The effect of increasing the temperature is to transfer spectral weight from the condensate to the quasiparticles, while at the same time reducing the quasiparticle lifetime. The net result is that both $|\epsilon'|$ and ϵ'' are reduced as the temperature increases, and the thin film transmission increases. In the inset in Fig. 1 the mm-wave transmission through a $\mathrm{DyBa_2Cu_3O_{7-\delta}}$ film of 20 nm thickness on a $\mathrm{LaAlO_3}$ substrate is displayed for $k/2\pi = 5\ \mathrm{cm}^{-1}$ and for $k/2\pi = 4\ \mathrm{cm}^{-1}$. The former corresponds to a larger sensitivity to the quasiparticles (represented by ϵ'') as compared to the latter frequency. A detailed analysis has been given elsewhere [9]. Most significant for the identification of a possible bolometric response is the *monotonic* temperature dependence of the transmission over the entire temperature interval.

In Fig. 1 the photoinduced change in transmission (δI_t) of the same film is shown between 5 and 65 K. The probe frequency is $5\ \mathrm{cm}^{-1}$. The pump frequency is $k/2\pi = 800\ \mathrm{cm}^{-1}$, with a power of $\simeq$10 mJ/pulse. Here and in Fig. 2 the curves have been calibrated against variations in the incident power of FELIX. We see that for temperatures lower than 40 K, the IR pulse enhances the transmissivity of the thin film. However, around 40 K the situation changes and the transmission after the IR pulse is reduced instead. δI_t is smaller at higher temperatures and becomes undetectable above 75 K. The ordinary *monotonic* behavior seen in the temperature dependence of the unperturbed mm-wave transmission indicates that a simple heating of the sample cannot account for the fact that $\delta I_t < 0$ above 40 K. The fits shown in Fig. 1 correspond to a linear combination of a slow (τ_B) and a fast (τ_R) decay: $\delta I_t = i_B e^{-t/\tau_B} + i_R e^{-t/\tau_R}$. The slow component i_B has a weak time dependence ($\tau_B \gg 45\ \mu\mathrm{s}$) on the interval displayed in Fig. 1 and is reduced from $i_R/3$ at 5 K to zero for $T > 40$ K. The prefactor of the fast component ($4 < \tau_R < 25\ \mu\mathrm{s}$) changes sign at 40 K. The changes in transmission as a function of pump frequency show a rather nonmonotonic behavior and have been summarized in Fig. 2 for several temperatures ranging from 5 to 60 K. Plotted are the maxima (minima) of the positive (negative) peak intensities obtained after calibrating against the changes in incident power. For comparison we display in the same figure the absorption coefficient in the superconducting film $A_f = 1 - R_f - T_f$ of the IR light, where R_f is the reflectivity of the substrate-supported film, and T_f is the transmission through the film into the substrate. We calculated A_f without adjustable parameters from the experimentally determined a- and c-axis dielectric function of YBaCuO [10,11] and $\mathrm{LaAlO_3}$ [12] using the Fresnel equations for light of mixed polarization incident at an angle of 45° on a 20 nm thick, c-axis oriented YBaCuO film on a $\mathrm{LaAlO_3}$ substrate, identical to the experimental situation. Optical absorption in the substrate occurs at 185, 427, and 651 cm^{-1}. For the film A_f has minima at these frequencies and maxima at 290, 600, and 760 cm^{-1}, which is due to resonant reflection at the substrate/film interface for frequencies matched to the longitudinal phonons of the substrate. The main conclusion from Fig. 2 is that δI_t tracks the laser power deposited in the film, not in the substrate. This demonstrates that PIAMA probes changes in the physical state of the superconductor, while secondary effects due to substrate heating can be excluded.

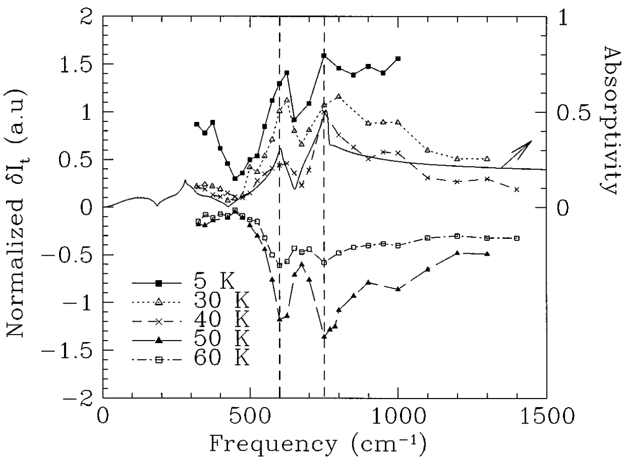

FIG. 2. Normalized δI_t as a function of frequency, shown for temperatures ranging from 5 to 60 K. Also shown is the absorptivity within the film (solid line).

The amplitude of the slow component, τ_B, in Fig. 1 corresponds to an increase in temperature of 13 and 0.2 K for the 5 and 40 K curves, respectively. A crude estimate of the increase in temperature based on the input laser power and the specific heat of the film/substrate system gives $\Delta T = 9$ and 0.2 K, respectively. We therefore attribute the slow response to bolometric heating of the film/substrate system. At higher temperatures the specific heat is too large, and ΔT is insignificant. We observed a similar bolometric response for MgO supported NbN thin superconducting films, in which case the faster decay was absent within the limitations of the time resolution of our detector. A more extensive discussion of this work is presented elsewhere [13].

Let us now consider the faster decay, τ_R. For sufficiently low frequencies ($\omega\tau \ll 1$) the inductive response is proportional to the condensate amplitude ($\epsilon' \propto \rho_s$), and will be reduced during and following the IR pulse, so that $\delta\rho_s < 0$. Here we are interested in the behavior of the quasiparticle response, which is represented by the finite value of ϵ''. The latter is proportional to the density of quasiparticles and their lifetime ($\epsilon'' \propto \rho_{\mathrm{qp}}\tau$). Because of transfer of spectral weight from the condensate to the quasiparticle peak we expect that $\delta\rho_{\mathrm{qp}} > 0$ in the nonequilibrium state following the IR pulse. With PIAMA we attempt to probe the time evolution of variations in the volume density of quasiparticles ($\delta\rho_{\mathrm{qp}}$). The highest sensitivity to the latter variations relative to those of the condensate is obtained for $\cos\psi = 0$, which is also the experimental situation in Figs. 1 and 2. In that case the transmission coefficient varies as

$$\frac{2n^2}{I_t^2}\,\delta I_t = -[kd\epsilon']^2\frac{\delta\rho_s}{\rho_s} - kd\epsilon''[1 + n^2 + kd\epsilon''] \times \frac{\delta\rho_{\mathrm{qp}}\tau}{\rho_{\mathrm{qp}}\tau}\,. \qquad (2)$$

Immediately following the laser excitation the excess quasiparticles have an enhanced nonequilibrium temperature T^*. During a short time interval of order 1 ns the quasiparticles thermalize to the equilibrium temperature. *If* the quasiparticle recombination time (τ_R) is long compared to the thermalization time, this leads to a transient state with *cold* excess quasiparticles, where ρ_{qp} is larger than the equilibrium value, but where τ is at its equilibrium value, resulting in a δI_t which is negative. Note that this situation is different from ordinary heating of the sample, where the quasiparticle peak also broadens ($\delta\tau < 0$). The net result in the latter case is a reduction of ϵ''. The possibility of cold excess quasiparticles was the subject of extensive investigations in conventional superconductors [14–16].

The coefficients in Eq. (2) are such [13] that below 40 K δI_t is dominated by $\delta\rho_s$ (causing $\delta I_t > 0$), whereas above 40 K $\delta\rho_{\mathrm{qp}}$ dominates (resulting in $\delta I_t < 0$). Interestingly 40 K presents a borderline case, where δI_t switches sign from positive to negative when the pump intensity exceeds a threshold value. This observation is consistent with a series of experiments as a function of incident laser power, pulse duration, and pump frequency [13].

A lifetime of several microseconds for the nonequilibrium state is surprisingly long compared to typical values reported for conventional superconductors, ranging from 1 ns to 1 μs. Before discussing those considerations which are specific to the high T_c superconductors we recall that the most important processes responsible for inelastic scattering are electron-electron interactions and inelastic scattering by spin and charge fluctuations and phonons. At this point we want to stress that a net decrease of the *number* of quasiparticles only results from phonon-assisted quasi-particle-pair recombination, i.e., events where two quasiparticles with momentum $\hbar k$ and $\hbar k'$ and energy E_k and $E_{k'}$ are converted to a Cooper pair and a phonon with momentum $\hbar(k + k')$ and energy $\Omega_{k+k'}$. A similar conversion of quasiparticle pairs into electronic collective modes may exist. However, as such modes can not escape into the substrate, they will be converted back and forth into quasiparticles without reducing the lifetime of the excited electron plasma.

In the cuprates several factors conspire to suppress the phonon-assisted quasiparticle recombination processes. The vertex for phonon-assisted quasiparticle recombination is the bare electron-phonon coupling constant (g) multiplied by the coherence factor $M_{kk'} = u_{k'}v_k + v_{k'}u_k$. In isotropic s-wave superconductors the lowest energy levels accessible to a quasiparticle have an energy Δ, so that the quasiparticles are equally distributed along the Fermi surface. In a d-wave superconductor one expects quite different behavior: While cooling down to an energy of order k_BT, the excess quasiparticles relax toward the nodes. After this relaxation is completed, recombination processes will only generate phonons with an energy of order k_BT and momentum of order $\hbar q_{ph} \approx k_BT/v_s$, where v_s is the sound velocity. As $T \ll k_Fv_s$, it follows that $q_{ph} \ll k_F$. Hence most recombination processes will involve two quasiparticles in nodes at opposite sides of the Fermi surface. The coherence factor $M_{kk'}$ is proportional to Δ_k/k_BT, which becomes zero at the nodes. Hence our first observation is that the quasiparticles relax to those regions where the gap has its zeros, thus separating them from the region in k space with the largest pairing amplitude. This in turn leads to a strong suppression of quasiparticle recombination processes. This first observation is robust, and applies to general k values of the nodes, but also if zeros of the gap occur in coordinate space, e.g., in the chain bands, at defects, vortices, etc.

For an isotropic s-wave superconductor energy conservation requires that the recombination process is fully suppressed if 2Δ exceeds the Debye frequency. For a d-wave superconductor the situation is perhaps even more intriguing. In thermal equilibrium the quasiparticles are concentrated near the nodes. Near the nodes the two-dimensional energy-momentum relation has the functional form $E_k^2 = (\partial_k\Delta)^2k_t^2 + \hbar^2v_F^2k_l^2$, where k_t and k_l are the momenta parallel and perpendicular to the Fermi surface measured relative to the node, and $\partial_k\Delta$ is the transverse momentum derivative of the superconducting gap at the node. If $\partial_k\Delta$ (70 meV Å at 4 K [17]) exceeds the sound velocity ($v_s = 30$ meV Å [18]), the constraints on momentum and energy conservation cannot be satisfied, leading to a suppression of this process. Hence our second observation is that the phonon-assisted quasiparticle recombination is suppressed due to kinematical constraints when a large gap opens. An analogy exists to the A phase in superfluid ^{3}He, where the relaxation rate of quasiparticles near the nodes has an algebraic (T^4) temperature dependence due to the reduction of the available phase space in the superfluid phase [19]. Together these arguments imply that there is a strong suppression of quasiparticle phonon scattering near the nodes, in particular of quasiparticle recombination processes. We used Fermi's golden rule [20]

$$\sum_k \frac{f_k}{\tau_R} = \sum_{k,k'} g^2|M_{kk'}|^2\,\mathrm{Im}\frac{f_kf_{k'}(1 + n_q)\delta_{k'}^{q-k}}{E_k + E_{k'} - \Omega_q - i0^+}\,,$$

$$\sum_k \frac{f_k}{\tau_i} = \sum_{k,k'} g^2|L_{kk'}|^2\delta_{k'}^{q+k} \times \mathrm{Im}\frac{f_k(1 - f_{k'})n_q + f_{k'}(1 - f_k)(1 + n_q)}{E_{k'} - E_k - \Omega_q - i0^+} \qquad (3)$$

to compute the thermal averages of the recombination

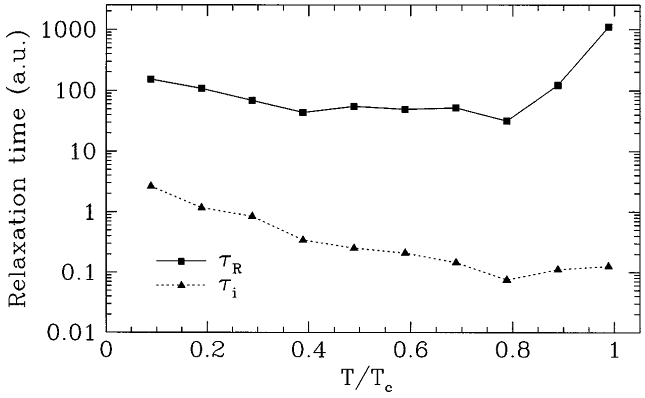

FIG. 3. Calculated temperature dependence of the quasiparticle phonon scattering time (τ_i, triangles) and the quasiparticle recombination time (τ_R, squares).

lifetime τ_R and the inelastic scattering time τ_i numerically. Here f_k and n_q are the Fermi-Dirac and Bose-Einstein distribution functions of quasiparticles and phonons, respectively, and $L_{kk'} = u_{k'}v_k - v_{k'}u_k$. We adopted a $d_{x^2-y^2}$ order parameter with $\Delta_{\max} = 25\,\mathrm{meV}$. The resulting temperature dependence of τ_R and τ_i is displayed in Fig. 3. Most importantly we notice that $\tau_R \gg \tau_i$ at all temperatures. The same calculation assuming an isotropic s-wave gap confirms the earlier result that τ_R and τ_i are equal for $T \rightarrow T_c$ in s-wave superconductors [21]. A small admixture of s-wave symmetry of the type "$d + s$" merely breaks the fourfold rotation symmetry of the quasiparticle dispersion, without affecting the physical picture. With an admixture of the type "$d + is$" the energy of the quasiparticles near the nodes is increased to $E_k^2 = (\partial_k\Delta)^2 k_t^2 + \hbar^2 v_F^2 k_l^2 + \Delta_s^2$, causing a further suppression of the available phase space for recombination processes, while $|M_{kk'}|$ increases near the nodes. The net effect on τ_R depends on $\partial_k\Delta$, v_F, and Δ_s.

Finally we discuss our observations in relation to time scales obtained with other experimental techniques. Using microwave experiments the scattering time is found to change from 100 fs at 90 K to less than 10 ps at 40 K. With pump/probe experiments using visible light a decay of 0.2 ps has been observed, which was associated with the lifetime of quasiparticles near the Fermi energy, along with a second slow decay of at least 20 ns [22]. A relaxation of the resistivity within a few nanoseconds [23] has been attributed to nonequilibrium quasiparticle generation by hot phonons. Based on an analysis of the critical flux-flow velocity Doettinger *et al.* [24] determined an inelastic scattering time ranging from 10 ps at 80 K to 0.1 μs at 40 K. The time scale of several microseconds reported in this Letter is much longer. We attribute this to the fact that the quasiparticle recombination time is always longer than the inelastic scattering time, which is the sum of all electron-electron and electron-phonon scattering processes, as is demonstrated by the numerical calculation of the two electron-phonon time constants τ_R and τ_i presented above.

In conclusion, we have observed a nonequilibrium state with a lifetime of several microseconds in $DyBa_2Cu_3O_{7-\delta}$ using photoinduced activation of mm-wave absorption. The nonequilibrium state is clearly distinct from bolometric heating of the superconductor. The long time constant seems to reveal an unusually long quasiparticle recombination time, which can be understood as a consequence of the highly peculiar nature of quasiparticles near the nodes in these materials. Along with other factors, such as the amplitude of the gap, the presence of nodes distinguishes these materials from conventional superconductors.

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