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Non-equilibrium Superconductivity and Quasiparticle Dynamics studied by Photo Induced Activation of Mm-Wave Absorption (PIAMA).

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We present a study of non-equilibrium superconductivity in $DyBa_2Cu_3O_{7-\delta}$ using photo induced activation of mm-wave absorption (PIAMA). 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 quasi-particles residing near the nodes of an unconventional superconductor, resulting in a strong enhancement of the recombination time.

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The occurrence of zero's 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 quasi-particles at low temperatures, which has only recently begun to attract the attention of researchers in the field. Due to the presence of these zero's (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 quasi-particle 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 quasi-particle dynamics using Photo Induced 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 macro-pulses with a stepwise off-on-off intensity profile ('on' for $3<t<7\mu$s 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 $(kd)^{-2} \approx 3 \cdot 10^8$. 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

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

where $\psi = nkD$. The effect of increasing the temperature is to transfer spectral weight from the condensate to the quasi-particles, while at the same time reducing the quasi-particle 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 of Fig. 1 the mm-wave transmission through a $DyBa_2Cu_3O_{7-\delta}$ film of 20 nm thickness on a $LaAlO_3$ substrate is displayed for $k/2\pi = 5\,cm^{-1}$ and for $k/2\pi = 4\,cm^{-1}$. The former corresponds to a larger sensitivity to the quasi-particles (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 photo induced change in transmission (δI_t) of the same film is shown between 5 and 65 K. The probe frequency is $5\,cm^{-1}$. The pump frequency is $k/2\pi = 800\,cm^{-1}$, 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 can not 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 ($\tau_B \gg 45 \mu s$) on the interval displayed in Fig. 1 and is reduced from $i_R/3$ at $5K$ to zero for $T > 40K$. The prefactor of the fast component ($4 \mu s < \tau_R < 25 \mu s$) changes sign at 40 K.

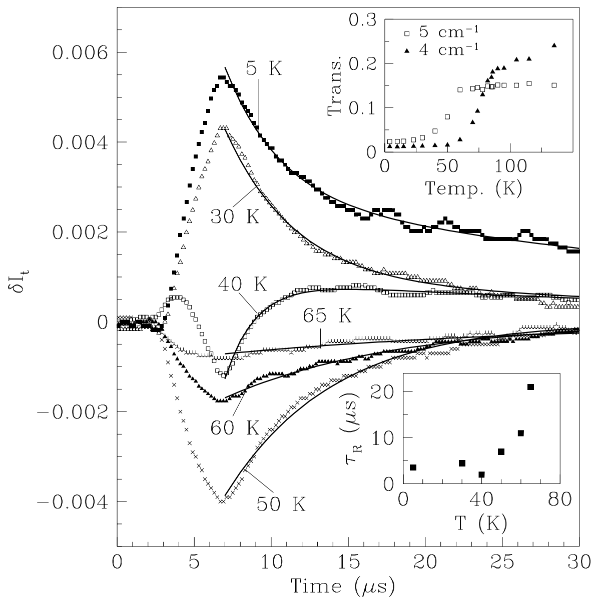

FIG. 1. Change in transmission, δI_t for $DyBa_2Cu_3O_{7-\delta}$ on $LaAlO_3$, shown for several temperatures. The FIR-pulse enhances transmission at low temperatures, while it reduces it at temperatures higher than 40 K. The exponential fits are shown as the solid lines. Inset, upper right corner: 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.

The changes in transmission as a function of pump frequency show a rather non-monotonic 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 $LaAlO_3$ [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 $LaAlO_3$ 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.

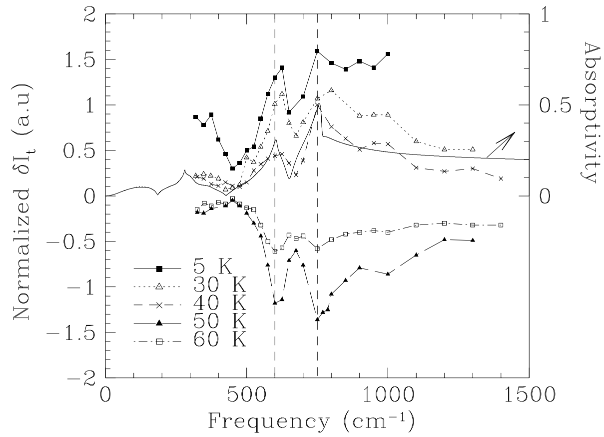

FIG. 2. Normalized δI_t as a function of frequency, shown for temperatures ranging from 5 to 60 K. Also shown is the absorptivity within the film (solid line).

The 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.

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 K 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 quasi-particle response, which is represented by the finite value of ϵ''. The latter is proportional to the density of quasi-particles and their lifetime ($\epsilon'' \propto \rho_{qp}\tau$). Due to transfer of spectral weight from the condensate to the quasi-particle peak we expect that $\delta\rho_{qp} > 0$ in the non-equilibrium state following the IR-pulse. With PIAMA we attempt to probe the time-evolution of variations in the volume density of quasi-particles ($\delta\rho_{qp}$). 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:

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

Immediately following the laser excitation the excess quasi-particles have an enhanced non-equilibrium temperature T^*. During a short time-interval of order 1 ns the quasi-particles thermalize to the equilibrium temperature. *If* the quasi-particle recombination time (τ_R) is long compared to the thermalization time, this leads to a transient state with *cold* excess quasi-particles, 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 quasi-particle peak also broadens ($\delta\tau < 0$). The net-result in the latter case is a reduction of ϵ''. The possibility of cold excess quasi-particles was the subject of extensive investigations in conventional superconductors [14–16].

The coefficients in Eq. (2) are such [13], that below 40K δI_t is dominated by $\delta\rho_s$ (causing $\delta I_t > 0$), whereas above 40 K $\delta\rho_{qp}$ dominates (resulting in $\delta I_t < 0$). Interestingly 40 K presents a bordercase, 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 μs 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 quasi-particles only results from phonon assisted quasi-particle-pair recombination, *i.e.* events where *i.e.* two quasi-particles 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 quasi-particle 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 quasi-particles without reducing the lifetime of the excited electron plasma.

In the cuprates several factors conspire to suppress the phonon-assisted quasi-particle recombination processes. The vertex for phonon-assisted quasi-particle 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 quasi-particle have an energy Δ, so that the quasi-particles 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 quasi-particles 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 $T \ll k_F v_s$, it follows that $q_{ph} \ll k_F$. Hence most recombination processes will involve two quasi-particles 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 quasi-particles relax to those regions where the gap has its zero's, thus separating them from the region in k-space with the largest pairing amplitude. This in turn leads to a strong suppression of quasi-particle recombination processes. This first observation is robust, and applies to general k-values of the nodes, but also if zero's 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 quasi-particles are concentrated near the nodes. Near the nodes the two-dimensional energy-momentum relation has the functional form $E_k^2 = (\partial_k\Delta)^2 k_t^2 + \hbar^2 v_F^2 k_l^2$ 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$\mathring{A}$ at 4 K [17]) exceeds the sound-velocity ($v_s = 30$ meV$\mathring{A}$ [18]), the constraints on momentum and energy conservation can not be satisfied, leading to a suppression of this process. Hence our second observation is, that the phonon assisted quasi-particle recombination is suppressed due to kinematical constraints when a large gap opens. An analogy exists to the A-phase in superfluid ^3He, where the relaxation rate of quasi-particles 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 quasi-particle phonon scattering near the nodes, in particular of quasi-particle recombination processes. We used Fermi's Golden Rule [20]

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

$$\sum_k \frac{f_k}{\tau_i} = \sum_{k,k'} g^2|L_{kk'}|^2\delta_{k'}^{q+k}$$

$$\mathrm{Im}\frac{f_k(1-f_{k'})n_q + f_{k'}(1-f_k)(1+n_q)}{E_{k'} - E_k - \Omega_q - i0^+} \quad (3)$$

to compute the thermal averages of the recombination 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 quasi-particles 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.

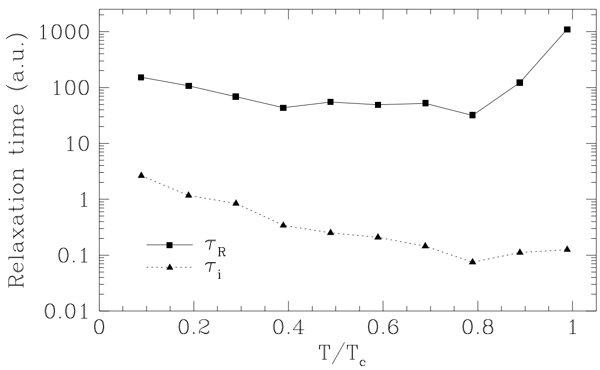

FIG. 3. Calculated temperature dependence of the quasi-particle-phonon scattering time (τ_{in}, triangles) and the quasi-particle recombination time (τ_R, squares).

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 4-fold rotation symmetry of the quasi-particle dispersion, without affecting the physical picture. With an admixture of the type 'd+is' the energy of the quasi particles 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 micro-wave 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 life-time 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 ns [23] has been attributed to non-equilibrium 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 timescale of several μs reported in this Letter is much longer. We attribute this to the fact that the quasi-particle 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 non-equilibrium state with a life-time of several μs in $DyBa_2Cu_3O_{7-\delta}$ using photo induced activation of mm-wave absorption (PIAMA). The non-equilibrium state is clearly distinct from bolometric heating of the superconductor. The long time-constant seems to reveal an unusually long quasi-particle recombination time, which can be understood as a consequence of the highly peculiar nature of quasi-particles 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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