Erosion of nodal Fermi spheres in nonequilibrium *d*-wave superconductors

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The relaxation of a *d*-wave superconductor that has been driven out of equilibrium by an optical pulse is investigated. We consider a simple model for the low-energy nonequilibrium state, one in which the unpaired quasiparticles form Fermi spheres near the gap nodes, and calculate the decay rate of the quasiparticle population due to phonon-assisted recombination. In the high density limit for the photoinjected quasiparticles, the decay rate dn/dt is found to vary as $n^{5/2}$, which differs from the n^2 form commonly adopted in phenomenological models of the relaxation dynamics. In the low density limit, the decay is exponential. From numerical estimates, we determine that phonon-assisted recombination could play an important role over the picosecond time scales of current interest. We compare our results to pump-probe optical experiments on high T_C cuprates and find reasonable agreement for the decay rate in underdoped YBCO for low laser intensity.

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Over the last several years, there have been numerous reports of femtosecond-resolved optical pump-probe measurements on high T_C cuprates in the superconducting state.[1–9] The data have revealed intriguing properties of the picosecond dynamics, including unusual dependence on laser intensity, temperature and doping. For example, recent measurements on BSCCO (Ref. 8) uncovered a change in the qualitative behavior, of both the initial optical response and its subsequent decay, that occurs suddenly as a function of doping at a value close to that giving optimal T_C. Such data could have far-reaching implications for the study of the high T_C phase diagram. In order to interpret them, some understanding of the nonequilibrium state of the *d*-wave system that occurs shortly after the arrival of the pump pulse is needed.

There is recent theoretical work aimed at understanding the dynamics of high T_C superconductors in the nonequilibrium state induced by a laser pulse.[10–13] The initial response of the system to a visible photon involves interband transitions and high-energy electron-electron intraband scattering in the strongly correlated system. In order to make some progress, people usually assume that after a sequence of fast high-energy relaxation processes, the system is left with a nonequilibrium distribution of low-energy (i.e., nodal) quasiparticles and phonons, the decay of which is governed by slower equilibration mechanisms. Of these slow relaxation rates, the conduction of heat out of the system by phonons that escape into the substrate and the recombination of quasiparticles into Cooper pairs are expected to be important. If the former rate is slowest, then heating is the only long-term effect of the pump laser (this is referred to as the T^* model and the strong bottleneck regime).[13,14] If the latter process is slowest, then the phonons and quasiparticles can achieve a common temperature while there remains an excess number of unpaired electrons. In a *d*-wave superconductor at a sufficiently low temperature, this corresponds to the presence of Fermi spheres of quasiparticles surrounding each node, described by Fermi distributions with a nonzero chemical potential $\mu(t)$ (this is called the μ^* model[11,15] and the weak bottleneck regime). The final relaxation is the erosion of the Fermi spheres because of quasiparticle recombination and the return to the true equilibrium $\mu(\infty)=0$.

The two slow relaxation rates described above may be competitive in high T_C superconductors over time and temperature scales of interest. Nevertheless, it is important to characterize the dynamics expected when one or the other of these relaxation processes is dominant.

In this paper, we consider the μ^* model and study the decay of the nonequilibrium quasiparticle population due to phonon-assisted recombination. We obtain a rate equation that governs the population decrease (or, equivalently, the collapse of the nodal Fermi spheres), and estimate the time scale over which the decay occurs. The quasiparticle number is found to obey $dn/dt \propto -n^{5/2}$ in the limit that the Fermi gas at the nodes is degenerate (i.e., $\mu(t) \gg k_B T$). In the nondegenerate limit, which is expected for vanishing pump intensity, the decay is exponential with a time constant that is proportional to T^{-3}. The latter behavior has been observed in underdoped YBCO samples for very low laser intensity,[6] and the time constant that we calculate is in reasonable agreement with the measured value. Our results suggest that phonon-assisted recombination can play an important role in the dynamic optical response over picosecond time scales for $T \ll T_C$, which contradicts earlier work[1] claiming that the recombination lifetime is of the order μs at low measurable temperatures.

We begin by obtaining an equation that governs the time evolution of the nodal Fermi sphere. The equation depends on the recombination lifetime for a single quasiparticle, which we calculate next. Finally, we determine the photoexcited quasiparticle density and compare our results to recent data before concluding.

In order to derive the rate equation for the nodal quasiparticle population, one may consider the lowest-order contribution of phonon-assisted recombination to the inverse lifetime of a quasiparticle with momentum $\mathbf{k}$ and a given spin, which is given by the Golden Rule as

$$\tau_{\mathbf{k}}^{-1} = 2\pi \sum_{\mathbf{k}'} g_{\mathbf{q}}^2(\mathbf{k}) L_{\mathbf{k},\mathbf{k}'}^2 f_{\mathbf{k}'}(1+n_{\mathbf{q}})\delta(\omega_{\mathbf{q}} - E_{\mathbf{k}} - E_{\mathbf{k}'}), \quad (1)$$

where $\mathbf{q}=\mathbf{k}+\mathbf{k}'$, and $\omega_{\mathbf{q}}$, $E_{\mathbf{k}}$ is the phonon, quasiparticle energy, and $g_{\mathbf{q}}(\mathbf{k})$ is the electron-phonon matrix element. The Fermi and Bose functions are written as $f_{\mathbf{k}}$ and $n_{\mathbf{k}}$, respectively (note that $f_{\mathbf{k}} \equiv f(E_{\mathbf{k}})$ always refers to the Fermi function with μ included) and $L_{\mathbf{k},\mathbf{k}'}^2 = (u_{\mathbf{k}} v_{\mathbf{k}'} - v_{\mathbf{k}} u_{\mathbf{k}'})^2$ is a BCS coherence factor. Equation (1) is valid only in the clean limit, that is when $k_B T \gg \gamma$, where γ is the constant impurity scattering rate of nodal quasiparticles. The total reduction in the population of quasiparticles due to such recombination is $\Sigma_{\mathbf{k},\sigma} f_{\mathbf{k}} \tau_{\mathbf{k}}^{-1}$.

An expression similar to Eq. (1), but with different occupation factors, holds for the rate of quasiparticle creation (phonon-induced pair breaking). By taking the difference of pair recombination and creation, one obtains the net recombination rate of quasiparticles as

$$\frac{dn}{dt} = -[1 - e^{-2\beta\mu}] \sum_{\mathbf{k}\sigma} f_{\mathbf{k}} \tau_{\mathbf{k}}^{-1}. \quad (2)$$

The explicit μ-dependence of Eq. (2) accounts for the presence of photoinjected quasiparticles: When $\mu=0$, the quasiparticles are in chemical equilibrium with the condensate and there is no net recombination.

If both T and μ are significantly smaller than the gap maximum Δ_0, then Eq. (2) can be written as

$$\frac{dn}{dt} = -\frac{1}{\Delta_0^2}(1 - e^{-2\beta\mu}) \int d\epsilon \epsilon f(\epsilon) \tau^{-1}(\epsilon), \quad (3)$$

where the recombination lifetime $\tau^{-1}(\epsilon)$ is the average of $\tau_{\mathbf{k}}$ over the energy contour $E_{\mathbf{k}}=\epsilon$. Starting from Eq. (3), the number of quasiparticles n will be written in the conventional units of $4N_0\Delta_0$, where N_0 in the normal-state density of states. Also, we use $2\Delta_0 = v_2 k_f$, where k_f is the length of the wave vector from the Brillouin zone center to the node and v_2, which is the slope of the gap along the Fermi surface at the node, will always be the quantity for which experimental estimates are obtained.

The total number of quasiparticles in a CuO_2 plane n is related to the chemical potential μ by

$$n = \frac{1}{\Delta_0^2} \int_0^\infty d\epsilon \epsilon \frac{1}{e^{\beta(\epsilon-\mu)} + 1}. \quad (4)$$

Taking the time derivative of this equation and comparing it to Eq. (3), we find that

$$\frac{d\mu}{dt} = -\frac{1}{k_B T} \frac{1 - e^{-2\beta\mu}}{\ln(1 + e^{\beta\mu})} \int d\epsilon \epsilon f(\epsilon) \tau^{-1}(\epsilon), \quad (5)$$

which is the desired equation of motion for the collapse of the Fermi sphere. The degenerate (nondegenerate) limit is the lowest-order term in $\mu/k_B T$ ($k_B T/\mu$).

In obtaining Eq. (5), we assumed that the recombination rate $d\mu/dt$ is either much faster or much slower than the time variation of the temperature dT/dt. In either case, recombination tends to relax the quasiparticle distribution toward a Fermi function with $\mu=0$ and temperature T. However, only in the latter case will the value of T correspond to the measured temperature of the substrate T_s. The time-dependent part of the quasiparticle population is given by

$$\delta n = \frac{1}{4N_0\Delta_0} \sum_{\mathbf{k}\sigma} [f_{\mathbf{k}}(\mu, T) - f_{\mathbf{k}}(0, T)]. \quad (6)$$

In this paper, we refer to δn as the number of photoexcited quasiparticles even though this terminology is not accurate when $T \neq T_s$.

Before proceeding, we address two possible concerns with the model. First, since phonons emitted by recombination can break pairs, one might expect that a nonequilibrium phonon distribution needs to be considered for consistency. For small $\mu/k_B T$, the entire system is close to equilibrium so this effect is higher order in $\mu/k_B T$. For large $\mu/k_B T$, it can be neglected, since it is far more probable that a phonon emitted from recombination will be absorbed in quasiparticle scattering than in pair breaking. To see this, one may consider that for large $\mu/k_B T$ quasiparticles fill the states with energy less than μ and an emitted phonon has energy between 0 and 2μ. Scattering can occur for arbitrary phonon energy since quasiparticles are available at the Fermi surface. Pair breaking requires a minimum energy of 2μ in order for there to be unfilled states to accept the created quasiparticles, which leaves no phase space for single-phonon recombination creation at $T=0$. The rate for pair breaking is clearly higher order in $k_B T/\mu$ in the degenerate limit, so it is not unreasonable to assume that the phonons are in equilibrium (at the instantaneous temperature T). Second, we are considering phonon-assisted recombination but not quasiparticle-quasiparticle recombination processes (i.e., Auger-type processes that, at low energy, are equivalent to recombination with emission of a spin fluctuation).[16,17] Several authors, e.g., Refs. 6 and 11, have pointed out that quasiparticle-quasiparticle recombination cannot dissipate energy from the system and thus cannot relax it to true equilibrium. However, it could relax a μ^*-distribution toward a T^* distribution, and give a transient response due to a difference in the optical properties of these models.[11] Although both processes may contribute, we consider only the phonon-assisted process for simplicity. Some aspects of quasiparticle-quasiparticle recombination have been treated previously.[12]

We now use Eq. (1) to calculate $\tau(\epsilon)$ in the clean limit, and its dependence on μ, and insert it into Eq. (5). (We also study the dirty limit by calculating the electron self-energy evaluated to lowest order in the electron-phonon interaction with a constant scattering rate γ included for electrons, which is appropriate when $\mu \ll k_B T \ll \gamma$, see Ref. 18.)

The momentum integral in Eq. (1) can be simplified by considering the energy-conserving δ-function. For $\mu + k_B T \ll \omega_D$, where ω_D is the Debye frequency, only processes involving acoustic phonons and quasiparticles at opposite nodes are possible (all others are suppressed by a factor $e^{-\beta(\omega_D-\mu)}$). The condition for energy conservation in the opposite node-acoustic phonon case is illustrated in Fig. 1. The fact that $v_2, v_f \gg c_s$, where c_s is the speed of sound for any acoustic mode, implies that energy conservation can only be satisfied if the phonon wave vector has a large component in the direction normal to the CuO_2 plane. To a good approxi-

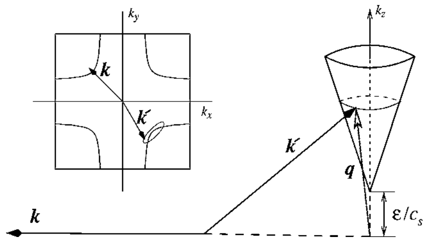

FIG. 1. Low-energy phonon-assisted recombination in d-wave superconductors viewed along the (001) and (110) directions. A quasiparticle, with wave vector $\mathbf{k}$ and energy ϵ, recombines with a quasiparticle $\mathbf{k}'$ and emits a phonon $\mathbf{q}$. To conserve energy and momentum, $\mathbf{q}$ and $\mathbf{k}'$ must lie on the surface of the cone. The cone is narrow (it has slopes v_f/c_s and v_2/c_s along and into the page), so the phonon wave vector is nearly parallel to the k_z axis. The figure is approximate.

mation, the phonon energy can be taken to be $\omega_{\mathbf{q}}=c_s|q_z|$. (The argument implies that for large $\beta\mu$ the nonequilibrium distribution of phonons emitted from recombination is unusual since phonons propagate along nearly the same line.) This simplification makes trivial the integrals along the in-plane energy contours.

We calculate $\tau^{-1}(\epsilon)$ using the procedure described above and the electron-phonon matrix element from Ref. 17 and obtain

$$\tau^{-1}(\epsilon) = \frac{N_0\alpha_c}{\Delta_0 c_s}F^2(k_f)\int_0^\infty dxx(x+\epsilon)f(x)[1+n(\epsilon+x)], \quad (7)$$

where α_c is the c-axis lattice constant and $F^2(k_f)$ is a constant with the dimensions of energy:

$$F^2(k_f) = \frac{g^2\eta^2}{2MNc_s^2}. \quad (8)$$

M is the mass of the unit cell, N is the total number of unit cells, and g is the electron-phonon coupling energy, (which is equal to the derivative of a hopping matrix element t, in the effective single-band Hamiltonian, with respect to bond length multiplied by the lattice constant). The dimensionless factor η^2 comes from the electron-phonon matrix element, we estimate that η^2 is of order 10^{-2} (this is discussed in a footnote in Refs. 19 and 20).

In the degenerate limit $\mu \gg k_BT$, the integral in Eq. (7) is equal to $\mu^2(\mu/3+\epsilon/2)$. After substituting this value into Eq. (5), evaluating the integral for $\mu \gg k_BT$, and expressing the result in terms of quasiparticle number, one obtains

$$-\frac{dn}{dt} = \frac{2\Lambda}{3}n^{5/2}, \quad \mu \gg k_BT, \quad (9)$$

which has the solution

$$\delta n(t) = n(t) = \frac{n(0)}{[1+\Lambda n^{3/2}(0)t]^{2/3}}, \quad \mu \gg k_BT. \quad (10)$$

The relaxation is similar to, but distinguishable from, second-order kinetics for which $dn/dt \propto -n^2$.

The time constant Λ^{-1} is given by

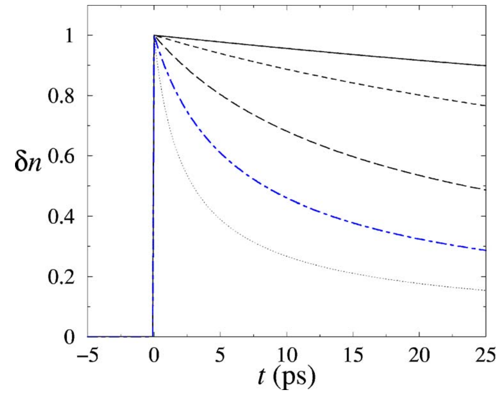

FIG. 2. (Color online) The population of photoexcited quasiparticles δn in the degenerate limit, given by Eq. (10). The normalized curves correspond to different values of the initial population $\delta n(0)$. In the top curve $\delta n(0)=n_0\equiv 0.004$; and the rest, in descending order, are for $\delta n(0)=2n_0$, $5n_0$, $10n_0$, and $20n_0$. Superconductivity breaks down at roughly $40n_0$ (according to Ref. 10).

$$\Lambda = \frac{\sqrt{8}\Delta_0^2\alpha_c}{c_s}\left(\frac{N_0g^2\eta^2}{2NMc_s^2}\right). \quad (11)$$

Using experimental values for cuprates (the main variation between different cuprate materials comes from the gap magnitude), we obtain $\Lambda^{-1}=1-10$ fs.

For sufficiently long times, the condition $\mu > k_BT$ must be violated. After this, the spread of the quasiparticles in $\mathbf{k}$ space is no longer affected by μ. During this final stage of relaxation, recombination occurs between one of the few remaining photoexcited quasiparticles and one of the much larger number of thermal quasiparticles [if $\mu(t=0) \ll k_BT$, then this is the only stage of relaxation in our model]. Repeating the calculations above for $\mu \ll k_BT$, we find that the nonequilibrium quasiparticle population follows:

$$\delta n(t) = \delta n(0)e^{-\Lambda(T)t}, \quad \mu \ll k_BT, \quad (12)$$

where the time constant is given as

$$\Lambda(T) = \frac{3}{\ln(2)\sqrt{8}}\left(\frac{k_BT}{\Delta_0}\right)^3\Lambda. \quad (13)$$

All of the preceding results are for the clean limit $\gamma \ll k_BT,\mu$. In the dirty nondegenerate limit, the result is Eqs. (12) and (13) with one of the powers of (k_BT/Δ_0) replaced by $(\gamma/2\Delta_0)\ln(\Delta_0/\gamma)$.

We will briefly compare our results to recent experiments by making the tentative assumption that the differential reflectance $\Delta R/R$, measured at optical frequency, decays with time in proportion to the number of remaining quasiparticles.[11,13] For high pump intensity Φ and low temperature, the degenerate limit, Eq. (10), may be achieved. In Fig. 2, we plot n in the degenerate limit, as given by Eq. (10). We have chosen values for $n(0)$ (see caption of Fig. 2) such that the evolution occurs over the time scale probed by recent experiments, which is 10 ps, and have used $\eta=0.1$. The behavior is qualitatively similar to that seen in the optical response of underdoped cuprates,[8] but the observed decay rate is proportional to $n(0)$ rather than $n^{3/2}(0)$. This discrepancy might point to the importance of a phonon-heat bottleneck, i.e., of the dT/dt term discussed above, for high-intensity measurements.[13]

For low intensity, we expect that the nondegenerate limit, Eqs. (12) and (13), is valid. In measurements made on ortho-II YBCO, Segre *et al.*[6] observed that the low-intensity decay rate is proportional to T^3, in agreement with Eq. (13). If we use $\Delta_0=66$ meV for Ortho-II (Ref. 21) and $\eta^2=0.1$, then our result for the decay rate matches that extracted from the data. This value of η^2 is larger by a factor of 10 than the rough estimate given above, but is not an implausible value.[19] This is suggestive that, for low enough pump intensity, phonon-assisted recombination may determine the time evolution of the optical response on the picosecond time scale.

The fact that our low-T recombination lifetime is orders of magnitude smaller than that calculated by Feenstra[1] should be discussed. Feenstra claimed that, because the quasiparticle velocity is larger than the sound velocity, energy momentum cannot be conserved in recombination processes involving acoustic phonons and quasiparticles at opposite nodes (which results in exponentially slow recombination at low T). This conclusion is only true if the phonons are constrained to propagate along the CuO_2 plane. For *in-plane* phonons, energy momentum is conserved when the phonon-wave vector is zero, i.e., when both quasiparticles lie exactly at the nodes, but cannot be conserved if quasiparticles are displaced from the nodes in any direction, since the quasiparticle energy increases more quickly than the phonon energy. However, if the emitted phonon propagates along the c axis, then it can carry away energy without any change in the in-plane momentum. (The quasiparticles can have any momentum along the c axis since they are assumed to be confined in real space.) By bending the phonon wave vector away from the c axis, energy conservation involving phonons and opposite-node quasiparticles is satisfied, as in Fig. 1. Thus, in a correct treatment in which phonons propagating out of the CuO_2 plane are considered, the lifetime for low-temperature quasiparticle recombination in equilibrium is found to be of the order 10–100 ps at 10 K, as above.

In summary, we have studied the effect of phonon-assisted recombination on the relaxation of a d-wave superconductor in a low-energy nonequilibrium state. The time evolution of the nodal Fermi sphere, which contains the photoexcited quasiparticles prior to recombination, has been calculated in both the degenerate and nondegenerate limits. There is good agreement between the result for the nondegenerate limit and observations made on underdoped YBCO with low laser intensity. The degenerate limit results may be useful toward understanding high-intensity data, although it is likely that the effects of laser heating and the associated phonon heat bottleneck are important in this case. Here, our work may complement phenomenological rate-equation approaches in which terms associated with both phonon heat conduction and recombination are included.[13]

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[19] An estimate of the coupling constant g for modes that stretch nearest-neighbor bonds has been obtained, from thermal conductivity data, but those associated with next-nearest neighbors, g' and out-of-plane neighbors $g_\perp$ are unknown. By considering all modes, we determine that $\eta \approx \max(g_\perp/g, [c_s/v_2]\mathbf{g}'/\mathbf{g}, [c_s/v_2]^2)$. If we roughly approximate $g'/g \approx t'/t$, then we get $\eta^2 \approx 10^{-2}$ for the second estimate. The third estimate gives $\eta^2 \approx 10^{-3}$, which sets a lower bound.

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