Time-Resolved, Far-Infrared Studies of Excess Quasiparticles in Superconductors

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Superconductivity is a phenomenon where electrons form pairs and an energy gap Δ develops in the electronic density of states both above and below the Fermi energy. The typical energy scale is several meV. This energy gap, a key parameter indicating the overall strength of the superconducting state, can be sensed by far-infrared spectroscopy. Electronic transitions across the full gap (energy 2Δ) correspond to breaking pairs, producing excitations called quasiparticles.

Breaking a significant number of pairs leads to a non-equilibrium condition where the superconducting state is weakened (indicated by a smaller energy gap). The return to equilibrium involves the recombination of the excess quasiparticles into pairs, releasing energy of at least 2Δ (per recombination event), usually as a phonon. The rate for this process involves the interaction between quasiparticles, which is of fundamental interest for any theory of superconductivity. Kaplan *et al*[1] have calculated the recombination time for a number of elemental BCS superconductors, obtaining characteristic values in the 10 to 100 picosecond range, depending on material and temperature. But the system does not truly relax on this time scale, since the resulting excess 2Δ phonons usually break other pairs at a similar rate[1]. So the excess energy is temporarily trapped in a coupled system of quasiparticles and phonons. The coupled system eventually relaxes as the phonons escape to other parts of the specimen, but this process can be slow compared to the recombination rate or pair-breaking rate, providing sufficient time for the excess quasiparticles and phonons to reach equilibrium with each other.

In an experiment where a fixed energy is deposited into the superconductor, this internal equilibrium condition allows one to determine how the excess energy is distributed between the excess quasiparticles and excess phonons. For example, consider the case where the quasiparticle lifetime is extremely long compared to the phonon lifetime against pair-breaking. Pairs of quasiparticles are rarely removed by recombination events, and the few that do occur are quickly followed by pair-breaking events that restore the quasiparticle pairs. So one would find the excess energy in the form of a large number of quasiparticles and very few phonons. This can be quantified as[2]

$$n_{qp}(T)\Big/n_0 = \frac{\Delta_0}{\Delta(T)}\frac{1}{1+2\tau_B(T)/\tau_R(T)} \qquad (1)$$

where $\tau_R(T)$ is the quasiparticle recombination time, $\tau_B(T)$ is the phonon pair-breaking time, n_0 is the quasiparticle density if all the excess energy is in the form of quasiparticles, and $\Delta(T)$ is the energy gap. The factor of 2 is a consequence of the fact that pair breaking creates two quasiparticles while removing only one phonon, and a subscript of "0" indicates a T=0K value. Kaplan et al demonstrated that the recombination and pair-breaking times for a BCS weak coupled superconductor follow universal temperature dependences, shown in Figure 1. Simple scale factors handle variations between materials, and the figure shows results using scale factors appropriate for Pb, *i.e.* $\tau(0)_R$ = 70 ps and $\tau(0)_B$= 34 ps. Substituting into the expression for $n_{qp}(T)/n_0$ yields curves of various shapes depending on

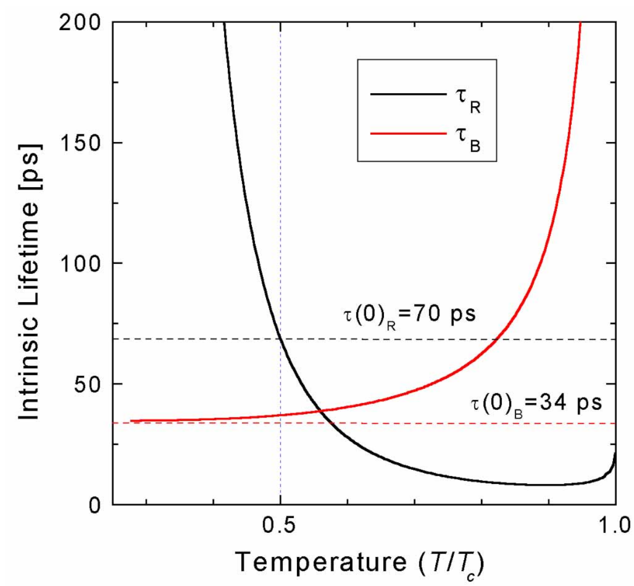

Figure 1. *Calculated intrinsic recombination time (black curve) and phonon pair breaking time (red curve) as a function of temperature for Pb. These universal curves are applicable to other BCS superconductors by suitable choice for the parameters* $\tau(0)_R$ *and* $\tau(0)_B$.

the ratio $\tau(0)_R/\tau(0)_B$. Thus, a measurement of $n_{qp}(T)/n_0$ can be used to determine this ratio and compare it with theory.

We have performed such measurements by time-resolved pump-probe spectroscopy. The technique[3] exploits the electron bunching in the VUV ring, producing ~ 350 ps duration pulses of far-infrared radiation. A mode-locked Ti:sapphire laser, synchronized to the VUV far-infrared pulses, serves as an excitation (pump) source. The photons from the laser break a small fraction of the superconducting pairs, leading to excess quasiparticles and a non-equilibrium state that evolves with time. This non-equilibrium state can be sensed as a small weakening (downward shift) of the superconducting energy gap, and increased far-infrared absorption. Using the pump-probe method, we follow this absorption (and therefore the excess quasiparticles) as a function of time. Results for a Pb film on sapphire, shown in Figure 2, yield an excess quasiparticle decay time of a few nanoseconds. This is the relaxation time for the coupled system of excess quasiparticles and phonons. The magnitude of this signal provides a measure of the excess quasiparticle density, and varies with temperature as expected from equation 1. We have measured this temperature dependent quasiparticle signal for Pb[2] and a number of other materials, with the results shown in Figure 3. Also shown are theoretical calculations for $n_{qp}(T)/n_0$ assuming values of $\tau(0)_R/\tau(0)_B$ ranging from 2 to 20. The behavior is generally consistent with eq. 1, supporting the analysis of Kaplan

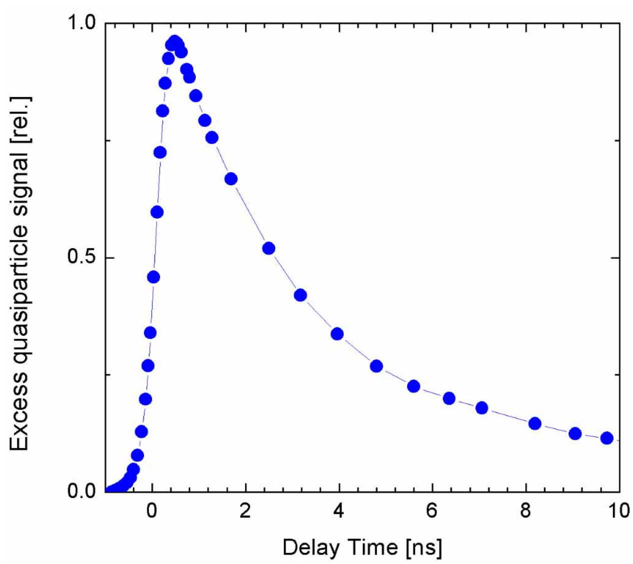

Figure 2. *Time-dependent relaxation of excess quasiparticles in a thin superconducting film following pair-breaking by a short laser pulse, determined by pump-probe far-infrared spectroscopy.*

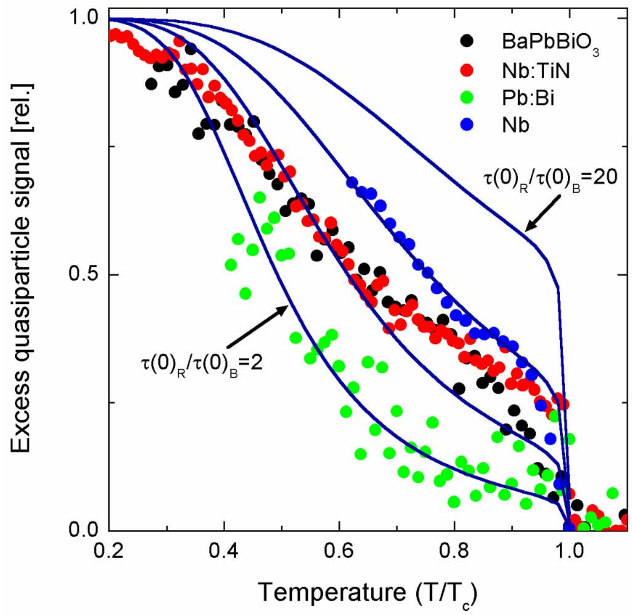

Figure 3. *Measured excess quasiparticle fraction for 4 different superconductors along with calculations assuming values for $\tau(0)_R/\tau(0)_B$ ranging from 2 to 20.*

et al. Materials with strong-coupling and a significant density of low energy phonons tend to have small values for this ratio. But there are some discrepancies, some of which may be due to possibly invalid assumptions of the theory (i.e. weak-coupling). For example, the energy gap does not grow as rapidly for T just below T_c in a strong-coupled superconductor. A smaller gap for T near T_c will cause $n_{qp}(T)/n_0$ to reach a higher value, consistent with the measured results for both Nb:TiN and Ba(Pb)BiO_3.We are presently investigating these details.

Acknowledgements

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