Dynamics of quasiparticles in superconductors

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The dynamics of quasiparticles in superconductors are studied using Sn-oxide-Sn tunnel junctions excited by 30-ns argon-laser pulses. The recombination lifetime and the closure of the superconducting energy gap as a function of laser power are measured. The relationship of these measurements to current theories of quasiparticle dynamics is discussed.

Recently there has been considerable interest in photoexcitation of quasiparticles in superconductors.¹⁻³ In an experiment by Testardi, ¹ pulses of laser light were used to drive thin superconducting lead films normal in times of the order of 10^{-6} sec or less. Stimulated by these experimental results, Owen and Scalapino² proposed a model of a nonequilibrium superconductor where they treated the unpaired electrons as a Fermi gas still characterized by a lattice temperature T but with a different chemical potential. From this model, they predicted a first-order phase transition to the normal state at some critical number of excited quasiparticles. Using a cw He-Ne laser, Parker and Williams³ reported measurements on Sn and Pb tunnel junctions at low excitation levels and their results were in general consistent with the theoretical model.

We report here an experimental study of the dynamics of quasiparticles in superconducting Sn under pulsed conditions. We do this by studying the I-V characteristics of Sn-oxide-Sn tunnel junctions excited by 30-ns argon-laser pulses. Measurements of the quasiparticle recombination time as a function of temperature and light intensity are presented and detailed I-V curves are studied. We find that the energy gap decreases continuously as a function of incident laser power but that the transition at the energy gap is appreciably broadened. We will compare our results and their implication with the theory proposed by Owen and Scalapino.

A standard-cavity-dumped argon laser with peak power of about 50 W was used. To minimize the stray leakage light from the laser cavity, we have used another acoustooptic coupler externally. Most measurements of the recombination lifetime and gap dependence on laser power were performed on a Sn junction of dimensions 0.2×0.2 mm and of a total thickness of about 3200 Å. The junction was deposited on a glass substrate and immersed in liquid helium (with the bath temperature varied from 1.2 to 4.2 K). The junction was biased at voltages less than the energy gap and hence the quasiparticle density was measured directly. The pulse current induced by the laser light was first amplified and then fed into a PAR boxcar integrator. The signal as a function of time was then displayed on a X-Y recorder. We have been careful in analyzing our circuit to be sure that our measurement of the recombination time was not limited by the RC time of our junction.

The measured recombination time as a function of temperature is shown in Fig. 1 and fitted with a function $y = y_0 (\Delta/kT)^{1/2} \exp(\Delta/kT)$, where 2Δ is the energy gap. Only one parameter y_0 is used in fitting the data. The general agreement between the measured temperature dependence and the function implies that the recombination time is proportional to the number of thermally excited quasiparticles, as expected.⁴ The deviation from the theoretical plot at the lowest temperature T = 1.2 K can be explained by the results shown in the inset of Fig. 1. Here we show our signal as a function of time for different laser intensities. The "narrowing" of our signal pulses at higher laser intensity is clear. The relative laser intensities for the A and B curves shown were 1 and 3, respectively. The dependence of the decay time on laserlight intensity can be easily understood. The relaxation of two quasiparticles to form a Cooper pair depends on the number of excited quasiparticles. At the lowest temperature there are so few thermally excited quasiparticles that in order to obtain a reasonable signal, the number of laserinduced quasiparticles is always comparable to or greater than the number of thermally excited quasiparticles and therefore the recombination time is effectively shortened. This "over-injection" is probably also present in all the previous measurements of recombination time^{5,6} at the lowest temperatures. Our pulse method has the advantage of being able to show directly the dependence of the relaxation time on laser intensity.

Two clarifications are certainly needed concerning the recombination time measured by our pulselight method. First, since the photon energy is a few orders of magnitude larger than the energy gap, the photon energy presumably will first be shared by many quasiparticles. There are both

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FIG. 1. Measured recombination lifetime as a function of temperature. The solid line is the theoretical curve with $y_0 = 3.5 \times 10^{-10}$ sec. The insert is the PAR boxcar output of the signal as a function of time. The relative light intensities are 1 and 3.

theoretical and experimental studies^{7,8} which suggest that the excited quasiparticles decay via a two-step process. The first consists of a relaxation to energy close to Δ , either by electronelectron interaction or by the emission of a phonon. The second step, recombination of two quasiparticles with energy close to Δ to form a Cooper pair, is accompanied by the emission of a phonon of energy 2Δ . It is known that the first step is much faster and is therefore not observed in our experiment. Our measurements will then be those of the relaxation times of the second process. The second question is the effect of reabsorption^{9,10} of the 2Δ phonon emitted during the recombination processes on our measurement of recombination time. This is known to have the effect of a simple modification of the factor y_0 . Because of many uncertainties in calculating this factor (film thickness, substrate, coupling to He, etc.) we present only the experimentally measured lifetime.

Next, we turn our attention to the study of the energy gap as a function of laser intensity. We note that in all of the following discussions the number of laser-excited quasiparticles is always much greater than the number of thermally excited quasiparticles at T = 1.2 K.

Before presenting the experimental results, we will briefly review the theoretical aspects of this problem. In the BCS theory of superconductivity the gap equation is given by

$$\frac{1}{V} = \sum_{k} \frac{1}{2E_{k}} (1 - 2f_{k}) ,$$

where $E_{k} = (\Delta^{2} + \epsilon_{k}^{2})^{1/2}$, the summation is a sum over all momentum k, V is the effective matrix element for the pair interaction, f_k is the distribution function, and ϵ_k is the one-electron Bloch energy measured from the Fermi energy. The interesting question arises as to what will be the quasiparticle distribution for a superconductor under an external dynamic pair breaking influence such as a flux of photons. Since it is known that the bottleneck on the relaxation of quasiparticles is that of the recombination process, Owen and Scalapino² suggested that this would give rise to an energy distribution still characterized by the lattice temperature T but now with a different chemical potential, namely $f_k = (e^{(E-\mu)/kT} + 1)^{-1}$, with the chemical potential μ determined by the excess number of quasiparticles. The gap dependence on n as derived by Owen and Scalapino at T = 0 K is illustrated in Fig. 2, where *n* is the number of excess quasiparticles in units of $4N(0)\Delta_0$, N(0) is the density of states at the Fermi level per spin, and $2\Delta_0$ is the energy gap at T = 0 K. The dashed part of the curve is energetically unfavorable, having an energy greater than that of the normal state and the first-order phase transition at n = 0.15, $\Delta = 0.62 \Delta_0$ is indicated. Also shown in Fig. 2 is the gap dependence on n, with f_{k} now given by the thermal distribution. Although the two distributions are quite different for the two cases, the gap dependences are similar at low excitation levels. Hence it appears that the most unequivocal test of the model is a determination of the nature of the transition of Δ . Presumably, the phase transition will manifest itself as a sudden discontinuity of the energy gap as a function of laser light. Therefore, we bias our junction at $eV = 2\Delta$ and study the signal as a function of laser intensity. In the insert of Fig. 3, we have shown the signal



FIG. 2. Gap dependence on *n*, the number of excess quasiparticles in units of $4N(0)\Delta_0$, for the Owen-Scalapino model and thermal distribution, respectively. The dashed part of the curve is energetically unfavorable and the first-order phase transition is indicated.



FIG. 3. The two dashed curves are the junction at thermal equilibrium with no light on. The solid lines are I-V characteristics under pulse-light conditions. The number in each curve is the relative light intensity, but the scale is different from the scale in Fig. 1. The insert is the signal as a function of laser intensity (arbitrary unit) for three different bias points A, B, and C.

in mV as a function of laser intensity at three different bias points and no discontinuity has been observed. In Fig. 3, we have also displayed the I-Vcharacteristics of the junction under pulse-light conditions with different laser-light intensities. Compared with the thermal I-V curves of our junction, we find that these two sets are similar at low biases, but the curves under laser light have much broader gap characteristics at $eV \sim 2\Delta$. One possible explanation for both the absence of any discontinuity in the closure of the energy gap and the gap broadening has been suggested by Rice.¹¹ Namely, although the film is indeed undergoing a first-order phase transition to the normal state, it tends to happen first in small pockets, and these normal regions grow as we keep on pumping with laser light. The observed I-V curves will therefore always be an average over the normal and superconducting regions. The result is then an apparent broadening at the gap. Another possibility is that the distribution of quasiparticles is actually very close to that of a thermal distribution at a higher temperature; therefore, no discontinuity is expected. In order that this explanation be consistent, the broadening of the gap would then suggest that there must be a temperature variation of the order of 1 K over the junction. A simple thermalconductivity calculation indicates that this is indeed possible with only a very small variation either in the reflectivity of the film or the energy density of the laser beam. The question of the relevant time for achieving the thermal distribution is a subtle one. Our measurement of the recombination time does indicate that the recombination time is indeed faster than our light pulses at high excitation levels; therefore, it is conceviable that it might require much faster light pulses (maybe of the order of y_0) for achieving the bottleneck and a nonequilibrium distribution.

In conclusion, we have been able to study the decay of quasiparticles as a function of temperature using pulsed laser light. We have also demonstrated the shortening of the lifetime of these quasiparticles at higher levels of laser excitation and have obtained I-V curves of Sn junctions under pulsed light conditions. Although we cannot determine exactly the energy distribution of these photoexcited quasiparticles in the superconductor, we do find that the gap can be changed continuously. Since superconducting thin films were used as a 2Δ phonon generator, ⁸ the tunability of 2Δ (by varying the laser intensity) might be useful in tuning the phonons so generated.

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