Photoexcitation of Quasiparticles in Nonequilibrium Superconductors*

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We report measurements on superconducting tunnel junctions illuminated with optical radiation, which confirm the model of nonequilibrium superconductors proposed by Owen and Scalapino. Measurements of the chemical potential μ^* for quasiparticle excitations and of the temperature dependence of the quasiparticle recombination time are presented.

Recent experiments by Testardi[1] have shown that thin superconducting films can be driven normal by laser light and that this change of state is not simply due to lattice heating. Testardi suggested that this effect may be due to an excess number of quasiparticles induced by the flux of optical photons. Owen and Scalapino[2] have developed a model of superconductors in which the number of quasiparticles is larger than the usual thermal-equilibrium number as a result of an external pair-breaking mechanism such as optical photons. They assume that the quasiparticles come to thermal equilibrium with the lattice in a time short compared to the recombination time for quasiparticles but do not reach thermal equilibrium with the pair state. The energy distribution of the nonequilibrium quasiparticles is characterized by the lattice temperature T and an additional chemical potential μ^*. In this Letter we report experimental results obtained from superconducting tunnel junctions illuminated with laser light, which support the model proposed by Owen and Scalapino and which determine an effective chemical potential for quasiparticles.

Their result for the energy gap of a nonequilibrium superconductor, valid for low reduced temperature, is

$$(\Delta/\Delta_0)^3 = \{[(\Delta/\Delta_0)^2 + n^2]^{1/2} - n\}^2, \tag{1}$$

where n is the excess quasiparticle number density in units of $4N(0)\Delta_0$. $N(0)$ is the single-spin density of states and Δ_0 the unperturbed energy gap at $T=0$. This reduces for small n to

$$\Delta/\Delta_0 \cong 1 - 2n. \tag{2}$$

If a thin-film superconductor is uniformly illuminated with optical radiation, quasiparticles will be produced at a rate proportional to the absorbed optical flux P, and will recombine at a rate proportional to n/τ, where τ is an effective recombination time.[3] In steady state

$$n = \frac{rP\tau}{4N(0)\Delta_0\mathcal{V}}, \tag{3}$$

where $\mathcal{V}$ is the volume of the illuminated superconductor and r the number of quasiparticles produced per photon if P is expressed in photons per second. We assume that the excess number of quasiparticles is small compared to the number in the absence of the laser light. Equations (2) and (3) predict that (1) the gap will decrease in proportion to the optical intensity, and (2) the gap will decrease exponentially with decreasing temperature since τ is approximately proportional to $e^{\Delta/kT}$ at low reduced temperatures.

The experiment is straightforward. Superconducting tunnel junctions, either Sn or Pb, with dimensions 0.8×0.3 mm^2 are prepared in the conventional manner. The film thicknesses are typically 1000 to 2000 Å. While the quasiparticles are produced in an optical penetration depth of a few hundred angstroms, the excess quasiparticle density should be approximately uniform throughout the thickness of the illuminated film since the diffusion length for quasiparticles is typically many micrometers.[4] The junctions are biased by a constant current source on the rapidly rising portion of the I-V curve at a voltage of 2Δ. The light from a He-Ne laser illuminating the junction through the walls of the glass Dewar is mechanically chopped and the resulting modulation of the energy gap is measured with a lock-in amplifier. The laser intensity is varied using a set of calibrated neutral density filters.

In these experiments it is not difficult to distinguish between lattice heating effects and quasiparticle photoexcitation. Heating of the lattice will produce neither of the effects mentioned above but will result in a signal that decreases with decreasing temperature because the BCS gap becomes less temperature dependent at low temperatures. To separate these effects, measurements were made on junctions prepared on glass and sapphire substrates and at temperatures above and below the λ transition of liquid helium. Lattice heating was observed with both substrates above T_λ. The heating effects ob-

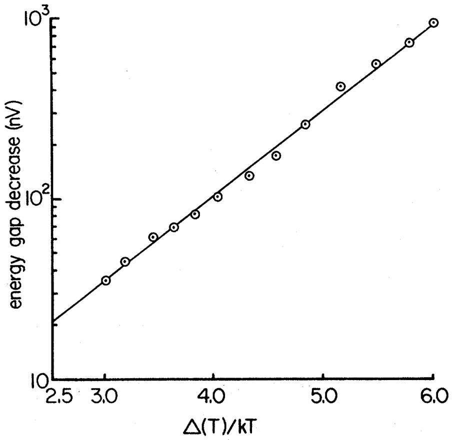

FIG. 1. The decrease in the energy gap of Sn versus $\Delta(T)/kT$ at constant laser intensity. The solid line indicates the theoretical temperature dependence of the quasiparticle recombination time in Sn.

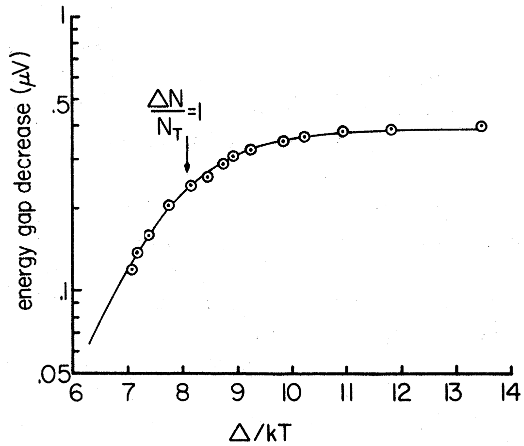

FIG. 2. The decrease in the energy gap of Pb versus $\Delta(T)/kT$. The solid line indicates the predicted temperature dependence of the decrease in the energy gap.

served with glass substrates were several times larger than those observed with sapphire. Below T_λ where the thermal conductivity of liquid helium increases substantially, no lattice heating was observed with either substrate. All the results reported in this Letter were obtained from junctions on sapphire and at temperatures less than T_λ.

The predicted behavior was observed with Sn-Sn-oxide–Sn tunnel junctions. It was verified that the energy gap decreased in proportion to increasing laser intensity over 2 orders of magnitude of optical power. The decrease in the energy gap at constant laser intensity was measured as a function of temperature from 2.17 to 1.14 K. The results obtained from a 400-Ω junction biased at 1 μA are shown in Fig. 1 where the change in the gap in nanovolts is plotted against $\Delta(T)/kT$, where $\Delta(T)$ is the measured gap. The temperature dependence of the recombination time for low reduced temperatures,[5] indicated by the solid line in Fig. 1, is $[\Delta(T)T]^{-1/2}e^{\Delta(T)/kT}$. The outstanding agreement is obtained using only a scaling factor as an adjustable parameter.

A similar measurement of the decrease of the energy gap, obtained from a 6.9-Ω Pb-Pb-oxide-Pb junction biased at 10 μA as a function of $\Delta(T)/kT$, is shown in Fig. 2. For small values of $\Delta(T)/kT$, the signal increases at approximately an exponential rate consistent with the temperature dependence of the recombination time of Pb. For temperatures such that $\Delta(T)/kT \gtrsim 10$, the signal is almost independent of temperature. Such behavior occurs when the excess number density of quasiparticles $\Delta N = 4N(0)\Delta_0 n$ becomes larger than the thermal-equilibrium number density N_T.[6] Rothwarf and Taylor calculate for all values of ΔN

$$\Delta N/N_T = (1 + 2\tau r P/N_T \mathcal{V})^{1/2} - 1. \tag{4}$$

The solid curve in Fig. 2 indicates the behavior predicted by Eq. (4) using both n and τ as adjustable parameters to fit the data. The arrow in Fig. 2 indicates the position on the curve corresponding to $\Delta N/N_T = 1$. For low reduced temperatures

$$N_T = 4N(0)[\tfrac{1}{2}\pi\Delta(T)kT]^{1/2}e^{-\Delta(T)/kT}, \tag{5}$$

$$\Delta N/N_T = 0.80\, n[\Delta(T)/kT]^{1/2}e^{\Delta(T)/kT}. \tag{6}$$

Using the experimental value of n at the position indicated by the arrow, Eq. (6) predicts $\Delta(T)/kT = 8.5$ when $\Delta N/N_T = 1$. This value is in good agreement with the experimental value of 8.1.

The calculation of Rothwarf and Taylor reduces to $n = (2N_T \tau r P/\mathcal{V})^{1/2}/4N(0)\Delta_0$ for $\Delta N/N_T \gg 1$. The change in the energy gap now varies as the square root of the laser intensity rather than as the intensity. This behavior is observed in Pb for $\Delta(T)/kT \gtrsim 10$.

It is possible to extract an order-of-magnitude estimate of the numerical value of the recombination time τ from estimates of the optical reflectivity and the parameter r. The maximum value of r results if all the photon energy produces quasiparticles of energy Δ. A minimum value of r results if the photon produces quasiparticles of average energy equal to the Debye energy and further interactions of the quasiparticles create phonons rather than more quasiparticles. In the case of Pb, this reasoning leads to $220<r<1500$. We assume $r=1000$, the optical reflectivity is 70%, and $N(0)=1\times10^{22}$ states $\mathrm{eV}^{-1}\,\mathrm{cm}^{-3}$.[7] The effective recombination time for Pb at low reduced temperatures, obtained from Eq. (4) and the data in Fig. 2, is $\tau=0.4\times10^{-11}T^{-1/2}e^{15.5/T}$ which is approximately an order of magnitude larger than the theoretical estimate[5] of $0.3\times10^{-12}\times T^{-1/2}e^{15.7/T}$. However, Rothwarf and Taylor have pointed out that the experimental lifetime may appear longer than the actual recombination time because the phonons produced by the recombination will themselves create quasiparticles. According to their analysis, this will result in an experimental lifetime approximately a factor of 5 larger than the true lifetime for our geometry.

In addition to observing the behavior of the energy gap, we have used the I-V curve of tunnel junctions to determine the chemical potential μ^* introduced by Owen and Scalapino to characterize the energy distribution of quasiparticles in nonequilibrium superconductors. This parameter is determined by the usual condition

$$N=4N(0)\int_0^\infty \{1+\exp[\beta(E-\mu^*)]\}^{-1}\,d\epsilon, \qquad (7)$$

where $E=(\epsilon^2+\Delta^2)^{1/2}$, N the total number of excitations (electrons and holes), and ϵ an energy measured relative to the conventional Fermi level. The parameter μ^* appears only in the Fermi function. Since the I-V curve of a superconducting tunnel junction is an integral over the density of states of the superconductors and the appropriate Fermi functions, the effective chemical potential μ^* can be determined from the I-V curve. We take as a model of an illuminated tunnel junction one superconductor unperturbed and the second specially homogeneous and characterized by $\Delta(n)$ and $\mu^*(n)$. This assumption is reasonable if the junction is uniformly illuminated by the laser light. The large diffusion length insures that the excess quasiparticles are uniformly distributed over the thickness of the illuminated film and the small tunneling probability ensures that the second superconductor is unperturbed. Following the standard procedure of calculating I-V curves of superconducting tunnel junctions,[8] we calculate

$$\frac{I(n)}{I(0)}\simeq\tfrac{1}{2}\left(1+\exp\{\beta[\mu^*(n)+\Delta(0)-\Delta(n)]\}\right), \qquad (8)$$

subject to the condition $2\beta\lesssim eV\lesssim\Delta(0)+\Delta(n)$; V is the voltage across the junction. $I(n)$ is the tunneling current of the illuminated junction and $I(0)$ the current of the unilluminated junction. This

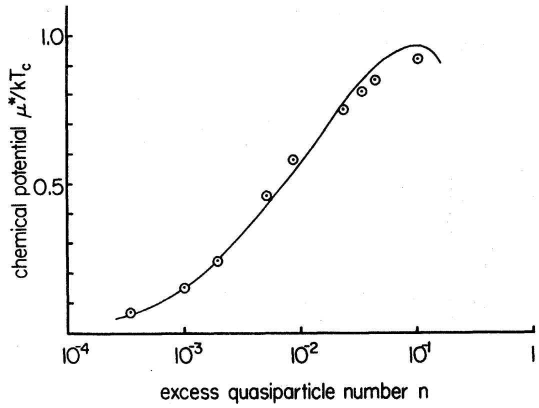

FIG. 3. Measured values of the effective chemical potential in units of kT_c versus measured values of n at $T/T_c=0.30$. The solid line is the theoretical prediction.

result is accurate to approximately 10% for $n<0.1$.

The observed I-V curves of illuminated tunnel junctions are essentially identical in shape to those of unilluminated junctions in the restricted voltage range but scaled by a multiplicative factor dependent on the optical power. The ratio $I(n)/I(0)$ and the energy gap $\Delta(n)$ are determined from the I-V curve and the parameter μ^* calculated from Eq. (8). The corresponding value of n is determined from Eq. (2). In Fig. 3 are shown the observed values of μ^* divided by kT_c obtained from a 2.5-Ω Sn–Sn-oxide–Sn tunnel junction at temperature $T=1.15$ K (reduced temperature 0.30) plotted against the measured value of n. The solid line in Fig. 3 is the predicted behavior of $\mu^*(n)$ at a reduced temperature of 0.30.[9] The agreement is excellent except at large values of n where the experimental values fall below the theoretical values. This small difference would result if the temperature of the superconductor was 0.1 K above the temperature of the helium bath. This is not an unreasonable temperature difference, considering that the junction is illuminated by 0.5 W of optical power at the largest value of n.

We conclude from all these measurements that the simple model of nonequilibrium superconductors proposed by Owen and Scalapino is quantitatively correct. The excess quasiparticles are in thermal equilibrium with the lattice at temperature T but the energy distribution is characterized by an effective chemical potential with the predicted behavior. Consequently, the "bottleneck" in the relaxation process of quasiparticles is the recombination time, and other interaction times in a superconductor such as electron-electron, electron-phonon, and branch mixing[10] must be considerably shorter. In addition, the behavior of the energy gap of an illuminated superconductor can be used to measure the quasiparticle recombination time in superconductors.

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Some Results Concerning the Crossover Behavior of Quasi–Two-Dimensional and Quasi–One-Dimensional Systems*

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A magnetic system with intraplanar and interplanar interaction strengths J and RJ is is treated. Rigorous relations are established concerning the first few derivatives with respect to R of the susceptibility $\chi(R)$. Considering $\chi(R) = b_0 + b_1 R + b_2 R^2 + \cdots$, we find b_1 and the order of magnitude of b_2. Hence we can predict when the system "crosses over" from d-dimensional to $\bar d$-dimensional behavior (e.g., for quasi–two-dimensional systems, $d=2$, $\bar d=3$, while for quasi–one-dimensional systems, $d=1$, $\bar d=3$). These results also support scaling with respect to the anisotropy parameter R.

There has recently been considerable interest[1-3] in systems with "lattice anisotropy" (different coupling strengths in different lattice directions). Consider, e.g., the d-dimensional nearest-neighbor (nn) Ising system with Hamiltonian

$$\mathcal{H} = -J \sum_{\vec v_i = \vec v_j}^{\text{nn}} s_i s_j - RJ \sum_{\vec u_i = \vec u_j}^{\text{nn}} s_i s_j \equiv \mathcal{H}_0 + R\mathcal{H}_1, \qquad (1)$$

where $\vec r_i \equiv (x_1, x_2, \cdots, x_{\bar d}) \equiv (\vec u_i, \vec v_i)$ with $u_i = (x_1, \cdots, x_d)$, and $\vec v_i \equiv (x_{d+1}, \cdots, x_{\bar d})$. For example, very recently there have been extensive calculations[1] concerning the case $d=2$, $\bar d = 3$, corresponding to a "square to simple-cubic crossover." Henceforth we shall consider this system for the purpose of specificity and clarity; thus $\vec r_i \equiv (x_i, y_i, z_i) \equiv (\vec u_i, z_i)$, $J = J_{xy}$, $RJ = J_z$. In the last paragraph we treat briefly the case $d=1$, $\bar d = 3$.

The system described by (1) is interesting because critical-point exponents, according to the universality hypothesis,[2] should depend only upon lattice dimensionality; and hence when $R \to 0$ (and the lattice "crosses over" from $\bar d$ dimensions), we expect anomalous behavior. This crossover behavior would be observable *if* we could vary R continuously to zero.

Another interesting property of the weakly coupled layers is that even for $R \neq 0$ the system is essentially two-dimensional at high temperature. Yet when it is sufficiently close to the critical temperature $T_c(R)$, it is *three*-dimensional. Hence there is a crossover region $T_A(R) \geq T \geq T_B(R)$ where the system transits from $d=2$ to $\bar d = 3$ (cf. Fig. 1).

The crossover region is only a loosely defined concept. To be quantitatively precise, we shall define $T_p(R)$ as the solution of $\chi(T_p, R)/\chi(T_p, 0) - 1 = p$ and we arbitrarily choose $T_A(R) = T_{0.01}(R)$, since this is roughly the temperature for which the deviation (1%) of the reduced susceptibility $\bar\chi(R) \equiv \chi/\chi_{\text{Curie}}$ from the two-dimensional value $\bar\chi(0)$ becomes experimentally appreciable.[4]

This crossover behavior is most easily explained in the context of the scaling hypothesis,[2] where we assume that there exist three numbers