Energy Gap Reduction in Superconducting Tin Films by Quasiparticle Injection

J. Fuchs, P. W. Epperlein, M. Welte, and W. Eisenmenger
Physikalisches Institut der Universität Stuttgart, D 7000 Stuttgart 80, Germany
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In Sn-*I*-Sn-*I*-Pb tunneling structures the energy gap Δ_{Sn} of Sn is reduced by quasiparticle injection via single-particle tunneling between the Sn films. Δ_{Sn} as function of the quasiparticle density is probed by the Pb contact and found in agreement with the theory of Owen and Scalapino. An instability of the energy gap of Sn is observed at the critical gap reduction ratio predicted by this theory for a first-order phase transition.

Nonequilibrium quasiparticle distributions in superconductors can be produced by photon[1-3] and phonon[4] irradiation or by quasiparticle[5] injection via tunneling. Under constant injection conditions the stationary quasiparticle energy distribution is determined by the energy distribution of the primary quasiparticle injection or excitation rates, by the energy dependence of relaxation and recombination probabilities, and by secondary quasiparticle excitation and pair-breaking rates via phonon absorption. Since phonons are emitted in quasiparticle decay, the phonon escape probability from the superconducting film into the substrate and the intrinsic phonon decay also have a strong influence on the stationary quasiparticle energy distribution. Whereas the general problem of the quasiparticle distribution can be solved numerically,[6] two important simple models have been discussed in the past: For the limit of recombination lifetimes long compared to relaxation times, Owen and Scalapino[7] proposed a nonequilibrium quasiparticle distribution in which the excess number of quasiparticles is characterized by a chemical potential $\mu^* > 0$ and their energy distribution by the unperturbed lattice temperature T. Since most superconducting films show high phonon trapping[8] by pairbreaking, Parker[9] proposed a model in which an elevated temperature $T^* > T$ describes the number of quasiparticles *and* their energy distribution. A significant difference between the two models is that the "μ^* model" predicts a first-order phase transition as the number of excess quasiparticles is increased, whereas the "T^* model" does not. Different experiments with optical excitation of quasiparticles[2,3,10] did not give clear evidence in favor of one of the two models.

In this communication we report on experiments with quasiparticle injection via tunneling between two Sn films and probing the energy gap and the quasiparticle population with a Pb contact. In accord with the μ^* model we find that the gap reduction as function of the quasiparticle density is stronger than in the thermal case and we observe an instability of the energy gap at the predicted critical gap reduction.

The sample consists of two overlapping Sn films and one Pb film, width and thickness of each film being 1.4 mm and 1000 Å, respectively (Fig. 1). Silicon single crystals are used as substrates which are cooled by direct contact to the liquid-He bath on the backside. The front surface with the Sn-*I*-Sn-*I*-Pb structure can be kept under vacuum or also exposed to liquid He. By 15-min glow-discharge oxidation in O_2 at 100 mTorr the tunneling resistance in the Sn-*I*-Sn junctions resulted with the higher voltage asymptotic value of $R_\infty \geq 25$ mΩ. For the Sn-*I*-Pb junctions typical values are $R_\infty \approx 1$ mΩ. The increased tunneling resistance in the Sn-*I*-Sn contact was necessary for obtaining a high stationary quasiparticle population at injection currents below the critical currents of the film structure. This allows high battery voltages and primary quasiparticle injection energies at multiples of the energy gap with successive relaxation-phonon emission and reabsorption by pairbreaking increasing the effective rate of quasiparticle excitations.

Using conventional electronic measuring tech-

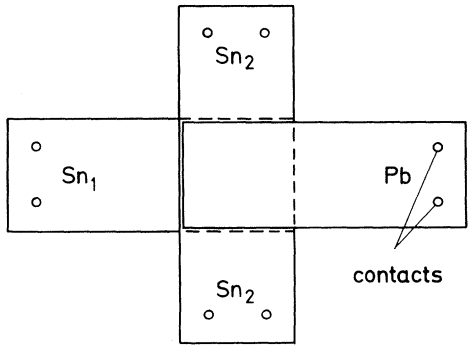

FIG. 1. Sample configuration. The center film Sn_2 is used for the critical current measurement.

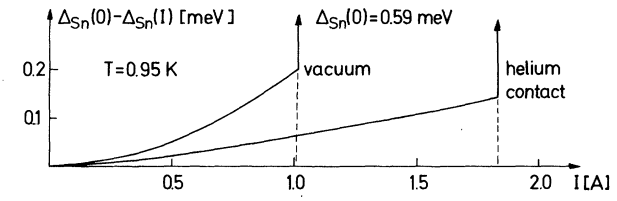

FIG. 2. Energy gap reduction $\Delta_{Sn}(0)-\Delta_{Sn}(I)$ measured with the Pb-I-Sn contact of the sample as function of the injection current I in the Sn-I-Sn junction.

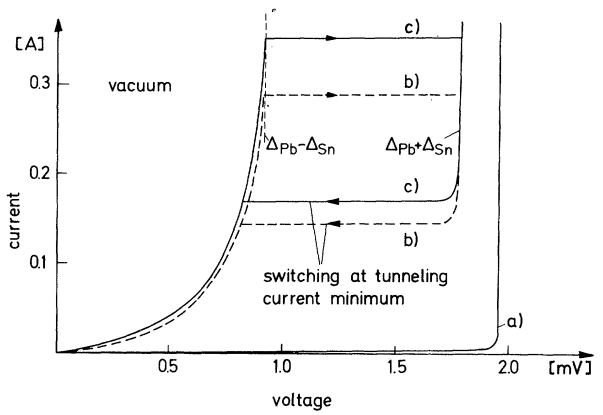

FIG. 3. I-V characteristic of the Pb-I-Sn junction: a, $T=0.96$ K without injection in the Sn-I-Sn contact, b, $T=0.96$ K with current injection of 0.9 A in the Sn-I-Sn contact. c, $T=2.92$ K—no current injection.

niques no differences between I-V curves under dc or pulsed conditions (repetition frequency 500 Hz, pulse duration 3 μs, tunnel junction under vacuum or He contact) could be detected, and we conclude that simple substrate heating and phonon backscattering from the substrate as possible for glass substrates is avoided. The quasiparticle concentration in Sn under either injection or thermal conditions is measured by the tunneling current of the Sn-I-Pb junction in the voltage-bias regime $\Delta_{Pb}-\Delta_{Sn}<eV<\Delta_{Pb}+\Delta_{Sn}$, whereas the energy gap reduction of Sn is directly obtained from the steep current increase at $eV=\Delta_{Pb}+\Delta_{Sn}$. In the entire regime of temperature and quasiparticle concentrations of our measurements we found no changes of the energy gap of Pb.

Figure 2 shows the energy gap reduction $\Delta_{Sn}(0)-\Delta_{Sn}(I)$ with increasing injection current I for a Sn-I-Sn junction with $R_\infty=32.2$ mΩ measured at $eV=\Delta_{Sn}+\Delta_{Pb}$ by the Sn-I-Pb contact [$\Delta_{Sn}(0)$= energy gap of Sn without quasiparticle injection; $\Delta_{Sn}(I)$= energy gap of Sn as function of quasiparticle injection current]. The energy gap becomes unstable[11] at injection currents of 1.03 or 1.7 A for vacuum or He contact conditions, respectively. The critical current in the Sn films was determined to be 2.7 A at the same temperature of 0.95 K. The maximum gap reductions at the onset of the instability are $\Delta_{Sn}(0)-\Delta_{Sn}(I)=0.20$ meV under vacuum and $\Delta_{Sn}(0)-\Delta_{Sn}(I)=0.137$ meV liquid He contact. There numbers correspond to $\Delta_{Sn}(I)/\Delta_{Sn}(0)=0.65$ for vacuum and $\Delta_{Sn}(I)/\Delta_{Sn}(0)=0.76$ for He contact. Different samples show scattering of these values by $\pm 10\%$. The μ^* model predicts the first-order phase transition at $\Delta_{Sn}(I)/\Delta_{Sn}(0)=0.60$ for Sn at 0.95 K in satisfactory agreement with the observed instability of the energy gap. At higher temperatures the experimental critical gap reduction ratio decreases as predicted by the μ^* model. The differences in quasiparticle concentration and gap reduction between vacuum conditions and liquid-He contact for equal injection current result from the strongly reduced 2Δ-phonon trapping[12] with He contact.

Since a given quasiparticle density in the μ^* model has a smaller energy distribution width than in thermal equilibrium excitation or in the T^* model, the same quasiparticle density results in a stronger gap reduction for the μ^* model.[13] This is demonstrated by the Sn-I-Pb I-V curves of Fig. 3 with and without injection by the Sn-I-Sn junctions in vacuum. Curve a is the Sn-I-Pb characteristic without injection at 0.96 K. Curve b shows the reduction of the Sn energy gap by an injection current of 0.9 A also at this temperature. In curve c the same energy-gap reduction was obtained by increasing the bath temperature to 2.92 K without current injection. The different quasiparticle densities in curves b and c are clearly demonstrated by the difference in the tunneling current minima[14] in the voltage range $\Delta_{Pb}-\Delta_{Sn}<eV<\Delta_{Pb}+\Delta_{Sn}$ leading to voltage backswitching along the load line. The thermal current minimum in curve c and the corresponding quasiparticle density is 17% higher as compared to the injection case in curve b for the same gap reduction, in satisfactory agreement with the μ^* model. By calibration using the thermal tunneling current together with the calculated thermal quasiparticle density N_T we obtained from the tunneling current minimum[14] the quasiparticle densities for different injection currents and gap reductions. The result in Fig. 4 shows the gap reduction ratio $\Delta_{Sn}(n)/\Delta_{Sn}(0)$ at $T=0.95$ K as function of the quasiparticle density parameter[7] $n=\Delta N/4N(0)\Delta_{Sn}(0)$ with $\Delta N=N_{inj}-N_T$; N_{inj} is the quasiparticle density by current injection, $N(0)$ is the

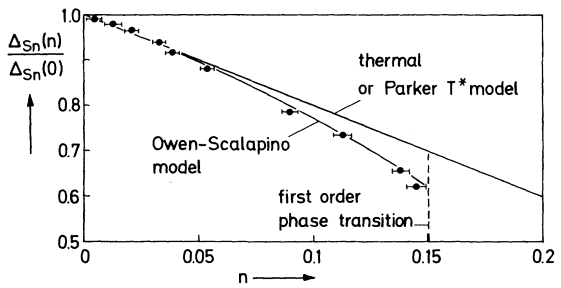

FIG. 4. Energy-gap reduction ratio $\Delta_{Sn}(n)/\Delta_{Sn}(0)$ as function of the quasiparticle concentration parameter n. The theoretical prediction of Owen-Scalapino and Parker are introduced as full lines.

Bloch density of states at the Fermi level, and $\Delta_{Sn}(0)$ is the energy gap of Sn at $T=0$ without injection. For comparison the calculated $\Delta_{Sn}(n)/\Delta_{Sn}(0)$ values for the μ^* model and for the T^* model are introduced together with the first-order phase-transition limit of Owen-Scalapino. The measurement shows good agreement with the μ^* model.

The general agreement of the experimental results with the Owen-Scalapino prediction of a first-order phase transition and a gap reduction by injected quasiparticles significantly different from thermal conditions exclude heating. This is strongly supported by the observations that at lower injection currents in the Sn junction the dependence of the energy gap reduction from injection current increases sharply at the battery voltage of $4\Delta_{Sn}$ as well as at $eV=6\Delta_{Sn}$. These structures cannot be explained by heating and are also known from phonon generation and detection[15] experiments indicating the onset of the successive emission and reabsorption of 2Δ-relaxation phonons.

Since the experimental critical film current of 2.7 A in our Sn films is higher than the critical injection current of 1 A in vacuum and 1.7 A in He, we expect magnetic influences to be small. This is consistent with the essential agreement of the critical gap reduction ratios found for both phonon-escape conditions and for different samples. A small magnetic field contribution may possibly explain the slightly higher critical $\Delta_{Sn}(I)/\Delta_{Sn}(0)$ value for the measurements with liquid-He contact. With respect to the vacuum case the parallel field component by a film current of 1 A in a stripe of 1.4 mm width amounts to 45 Oe with only minute influence on the quasiparticle properties[16] and the energy gap.

In order to check the μ^* model condition that the relaxation times τ_{rel} must be short compared to the recombination time τ_{eff}, we used our sample structure for a stationary measurement of these time constants in the same way as earlier performed with Al-I-Al-In structures by Miller and Dayem.[17] We find $\tau_{rel}=5.2\times10^{-10}$ sec for a quasiparticle energy $E=2\Delta$ above the Fermi level and $\tau_{eff}=5\times10^{-9}$ sec ($\pm3\times10^{-9}$ sec) for a gap reduction ratio of $\Delta(n)/\Delta(0)=0.65$. *Note.*—Close to the gap edge τ_{rel} varies as $(E-\Delta)^{-3.5}$. We also note that recombination phonon trapping factors[18] in our samples are of the order of 100 under vacuum conditions. The excess energy of phonons and quasiparticles exceeding 2Δ or Δ, respectively, is degraded in successive phonon reabsorption and re-emission steps even if relaxation rates are slower than recombination. The low-energy phonons with $|E_p|<2\Delta$ emitted in relaxation are no reabsorbed and have a comparatively high escape rate into the substrate crystal. Under these conditions 2Δ phonon trapping results in additional "cooling" of excited quasiparticles to the bath temperature thereby producing a quasiparticle distribution which can be well approximated by the "μ^* model." Fast low-energy phonon escape and quasiparticle injection at moderate energies therefore appear favorable for produc this distribution. In contrast, quasiparticle distributions generated by optical[3,10] or high-energy heat-pulse excitation[4] may be better approximated by the "T^* model."

From the present experiment the existence of a first-order phase transition can be only concluded in the sense of the observation of an instability[19] of the energy gap; see Fig. 2. An intermediate state, as discussed in prior experimental[3,10] and theoretical[20] work, is not ruled out since we observed steplike further switching processes[11] until the normal conducting state is reached for the entire film. But this behavior can also be explained by assuming spatial inhomogenities of the energy gap or of the tunneling probability. A recent comment of Elesin[21] on the possible breakdown of the μ^* model used the argument that the quasiparticle occupation numbers cannot exceed the value 0.5, or, in other terms, μ^* cannot exceed Δ unless stimulated recombination reduces abruptly the recombination time. For our experimental conditions we checked that up to the critical quasiparticle population the occupation numbers for a distribution according to 0.95 K do not exceed 0.5.

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Stability of Radiation-Stimulated Superconductivity

Albert Schmid*
Institut für Physik, Universität Dortmund, West Germany
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Recent observations of radiation-stimulated superconductivity above the thermodynamic transition temperature reveal the existence of a first-order phase transition between a normal and a superconducting state. In this Letter, a Langevin equation for the order parameter is derived, which is appropriate for the nonequilibrium steady-state process under consideration, and which explains the observed discontinuous phase transition.

The prediction of Eliashberg[1] on radiation-stimulated superconductivity seems now to have found acceptance from the experimental point[2,3] of view, particularly since superconductivity above the transition temperature has been observed[3] in a homogeneous material. It is well known that thermally excited electrons and holes oppose superconductivity; this explains the phase transition at finite temperatures. Less known, however, is the fact that excitations near the gap edge are more detrimental to superconductivity than those which are off the edge. Quite generally, a classical radiation field tends to spread the energy distribution of the excitations and thus remove them from the gap edge. In essence, this is the explanation of radiation-stimulated superconductivity.

In a certain range of temperatures, Eliashberg's theory allows three solutions of the gap equation, a situation which reminds us of van der Waal's theory of real gases. Furthermore, Klapwijk, van der Berg, and Mooij[3] have observed a discontinuous transition between the normal and superconducting states, including hysteresis, which is similar in appearance to a first-order phase transition. However, standard thermody-