Diffusive Quasiparticle Instability toward Multiple-Gap States in a Tunnel-Injected Nonequilibrium Superconductor

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Experimental observations of multiple-gap states in a strongly tunnel-injected nonequilibrium superconductor are reported. A phenomenological model based on quasiparticle diffusion successfully accounts for the qualitative features of these observations.

It has been believed for some time that a homogeneous nonequilibrium superconductor can become unstable with respect to formation of a spatially inhomogeneous nonequilibrium state.[1-3] Recently, Dynes, Naranamurti, and Garno (DNG)[4] and Gray and Willemsen (GW)[5] reported observations of an instability in a nonequilibrium state created by tunnel injection of quasiparticles and probed via a second tunnel junction. DNG interpreted their results as indicating an intrinsic instability toward a new type of spatially inhomogeneous superconducting state, while GW showed their results were also consistent with a less fundamentally interesting load-line-switching effect involving nonuniform quasiparticle injection.[6] In this Letter, we report experiments which differ from those of DNG and GW in the crucial respect that our injection voltages were extended well above the gap voltage, so that quasiparticle injection was uniform over the injected superconductor film. Under these conditions we show that an intrinsic instability does occur.[7]

In our experiments we used three-film, two-tunnel-junction structures on glass substrates. With the notation S_1-I-S_2-I-S_3, S_1 was a Pb-Bi film with a gap parameter $\Delta_1 \cong 1.7$ meV, and S_2 and S_3 were Al films. $\Delta_3 \cong 0.3$ meV, and Δ_2 was made appreciably smaller than Δ_3 by controlling evaporation conditions; $\Delta_3 - \Delta_2$ ranged from 10 to 100 μeV among our samples. Film 3 was in contact with the substrate and had a thickness ~1000 Å; film 2 had a thickness ~500 Å. The differences in material, gap parameters, and thicknesses among the three films were designed to minimize nonequilibrium perturbations in films 1 and 2 due to pair breaking by recombination phonons diffusing throughout the sample structure.[8] The junction areas were 380×380 μm^2. S_1-I-S_2 was used as an injector junction to create a nonequilibrium quasiparticle population in film 2, which was then probed using S_2-I-S_3 as a detector junction. The specific resistances of the injector junctions ranged from 10^{-5} to 10^{-2} Ω cm^2 and the specific resistances of the detector junctions were typically 10^{-3} to 10^{-2} Ω cm^2. The junction edges were not covered, but the junction films were carefully aligned to prevent direct tunneling between films 1 and 3. In most samples the detector-junction area common to the injection junction was 90% or more of the total area. All junctions studied were of high quality and free from excess current, shorts, etc.

For relatively low-resistance injector junctions biased at the gap-sum voltage, we observed behavior generally similar to that reported by DNG[4] and by GW.[5] We believe this situation represents a special case. Here we wish to focus on the more general situation exemplified in Fig. 1.

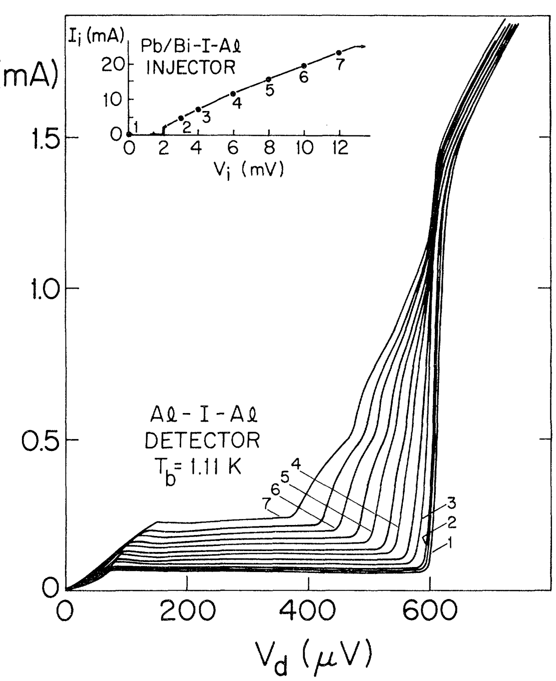

FIG. 1. Detector and injector (inset) current-voltage characteristics. Detector-curve numbers correspond to indicated points on injector characteristic.

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For relatively high-resistance injectors, we observed no instability for injection currents on the gap-sum step in the injector characteristic. Rather, for injection currents well above the gap-sum step we observed the appearance of structures in the detector characteristic which we believe correspond to the nucleation and growth in film 2 of regions β with a reduced gap $\Delta_{2\beta}$, and of still further regions γ and δ with gaps $\Delta_{2\gamma}$ and $\Delta_{2\delta}$ intermediate in value between the "main" gap $\Delta_{2\alpha}$ and $\Delta_{2\beta}$. These new gaps are reflected in both the gap-sum and the gap-difference structures in the detector characteristic. About two-thirds of our samples showed as many as three distinct reduced gaps.

The reduced gaps were more easily detected and followed in the first derivative of the detector characteristic. By measuring the corresponding peak heights in a dV_d/dI_d vs I_d plot as a function of injector current I_i and extrapolating to zero peak height, we found for the sample of Fig. 1 the following threshold currents for the successive appearances of the reduced-gap regions: $I_{i\beta}=6.4$ mA, $I_{i\gamma}=8.1$ mA, and $I_{i\delta}=10.7$ mA. These threshold currents are indicated in Fig. 2, which shows the dependence of the gaps on injection current for this sample. The regions appeared continuously in second-order fashion and the corresponding gap-structure signals grew continuously in size with increasing injection current. All the threshold currents increased monotonically with increasing bath temperature. The threshold current for the appearance of the second-gap region remained within the range 5 ± 1 mA for injector specific resistances ranging over two orders of magnitude, so long as the sample (i.e., film 1) was in direct contact with superfluid helium. This was so whether the threshold occurred on the gap-sum step in the injector characteristic or above it. $I_{i\gamma}$ behaved similarly. Coating samples with a thin (a few microns) coating of photoresist reduced the threshold currents by a factor of 2 or 3 and increased the gap difference $\Delta_{2\alpha}-\Delta_{2\beta}$.[9] For samples in direct contact with helium we observed no hysteretic switching in the injector characteristic,[4,5] but did occasionally observe some indication of such switching in photoresist-coated samples.

This behavior is qualitatively different from that reported by DNG[4] and by GW[5] and cannot be attributed to inhomogeneous quasiparticle injection.[5,6] We now outline a model based on previous theoretical work on diffusive quasiparticle instabilities in nonequilibrium superconductors[10,11] which can account for our observations.

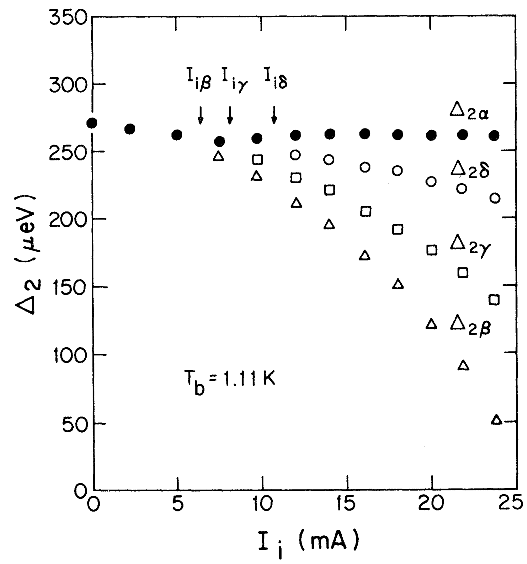

FIG. 2. Dependence of gaps on injection current.

We begin with the steady-state solution of the Rothwarf-Taylor equations,[12] modified to include quasiparticle diffusion,

$$N^2=N_T{}^2+(I_0-\nabla\cdot\vec{J})(2R)^{-1}[1+(\tau_{es}/\tau_B)],$$

$$I_0\equiv I_{qp}+\left(\frac{2\tau_{es}}{\tau_B+\tau_{es}}\right)I_{ph} . \qquad (1)$$

Here N is the quasiparticle concentration and N_T is the thermal-equilibrium quasiparticle concentration. I_{qp} and I_{ph} are, respectively, the quasiparticle and phonon injection rates, assumed uniform across the film interfaces. R is the quasiparticle recombination coefficient, τ_B is the phonon pair-breaking time, and τ_{es} is the phonon escape time. We assume that the quasiparticle diffusion current density $\vec{J}$ is given by the Scalapino-Huberman expression,[11]

$$\vec{J}=-[2D/N(0)\Delta_0](N_c-N)\nabla N+\tfrac{1}{2}D\xi^2\nabla(\nabla^2 N), \qquad (2)$$

where D is the quasiparticle diffusion constant, $N(0)$ is the Bloch single-spin density of states at the Fermi surface, Δ_0 is the zero-temperature equilibrium gap parameter, and ξ is the zero-temperature coherence length. N_c is a critical quasiparticle concentration above which the effective quasiparticle chemical potential decreases with increasing quasiparticle concentration. This effect is the key to the occurrence of a diffusive quasiparticle instability because it leads to quasiparticle diffusion from regions of low concentration to regions of high concentration when the critical concentration is exceeded.

Given our experimental evidence for the existence of separate regions, within each of which the gap is fairly constant, together with indications from theoretical discussions of similar systems,[13] it seems reasonable to consider a model in which it is assumed that in the fully developed multigap state the quasiparticle concentration is constant within each region and has nonzero gradients only at the boundaries of the regions. We further assume that the normal component of the quasiparticle diffusion current density at the boundary s of such a region can be approximated by the first term of Eq. (2):

$$J_n = -[2D/N(0)\Delta_0][N_c - N(s)](\partial N/\partial n)_s, \qquad (3)$$

where $(\partial N/\partial n)_s$ is the spatial gradient of N along the direction n in the plane of the film and normal to the boundary s. The stabilizing effect of the second term in Eq. (2) can be simulated by imposing some finite value on $(\partial N/\partial n)_s$. $(\partial N/\partial n)_s$ is weakly dependent on I_0 compared with the factor $[N_c - N(s)]$, so we can take it to be a constant without losing any essential features of the model.

With these assumptions, we now consider two distinct regions α and β, at an injection rate above the threshold for the two-gap state. Integration of Eq. (1) over each region, with use of Eq. (3), yields

$$\begin{aligned} &2R(N_\alpha^2 - N_T^2) = \hat{I}_0 - D_\alpha[N(s) - N_c], \\ &2R(N_\beta^2 - N_T^2) = \hat{I}_0 + D_\beta[N(s) - N_c], \end{aligned} \qquad (4)$$

where $\hat{I}_0 = I_0[1 + (\tau_{es}/\tau_B)]$,

$$D_\alpha = \frac{2D}{N(0)\Delta_0}\left(\frac{L_s}{A_\alpha}\right)\left(1 + \frac{\tau_{es}}{\tau_B}\right)\left(\frac{\partial N}{\partial n}\right)_s,$$

and

$$D_\beta = (A_\alpha/A_\beta)D_\alpha.$$

A_α and A_β are the areas of regions α and β, L_s is the perimeter of the boundary between the two regions, and the normal in $(\partial N/\partial n)_s$ is the outward normal from region α into region β. At the boundary we also have

$$2R[N^2(s) - N_T^2] = \hat{I}_0. \qquad (5)$$

Elimination of $N(s)$ among Eqs. (4) and (5) gives equations for N_α and N_β,

$$\begin{aligned} &2R(N_\alpha^2 - N_c^2) \\ &\quad = \hat{I}_0 - I_\beta + D_\alpha N_c - D_\alpha[N_c^2 + (2R)^{-1}(\hat{I}_0 - I_\beta)]^{1/2}, \\ &2R(N_\beta^2 - N_c^2) \\ &\quad = \hat{I}_0 - I_\beta - D_\beta N_c + D_\beta[N_c^2 + (2R)^{-1}(\hat{I}_0 - I_\beta)]^{1/2}, \end{aligned} \qquad (6)$$

where we have used the definition $I_\beta \equiv 2R(N_c^2 - N_T^2)$.

The behavior predicted by Eqs. (6) is shown schematically in Fig. 3. As the injection current I_i is increased from zero, N increases from N_T, linearly at first and then tending toward a $I_i^{1/2}$ dependence, as described by Eq. (1) with the diffusion term omitted because N is constant throughout the film. When N reaches the critical concentration N_c, the quasiparticle concentration curve bifurcates; a high-concentration (and hence low-gap) region β appears, within which the concentration increases continuously as the injection current is further increased. The concentration in the α region first decreases slightly, then increases again. When it reaches N_c, a second bifurcation should occur, spawning a second high-concentration region. Within the model, such bifurcations may occur repeatedly.

Crude as it is, this model successfully reproduces all of the qualitative features of our experimental observations. At a series of threshold injection currents, $I_{i\delta} > I_{i\gamma} > I_{i\beta}$ we observe the appearance of superconducting regions with quite

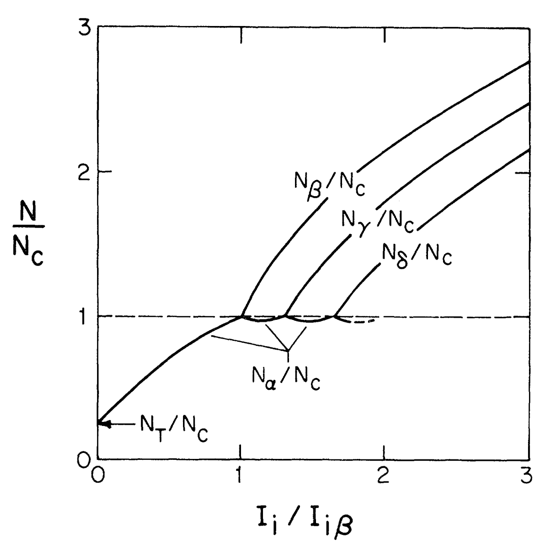

FIG. 3. Schematic dependence of quasiparticle concentration on injection current according to the model described in text.

well-defined gaps $\Delta_{2\alpha} > \Delta_{2\delta} > \Delta_{2\gamma} > \Delta_{2\beta}$. In the model N_α exhibits small decreases between thresholds; we have experimentally observed a slight increase (~ 5 μeV) in $\Delta_{2\alpha}$ above the first threshold. $\hat{I}_0$ differs from I_0 by a phonon-trapping factor; this is consistent with the observed differences in threshold currents between samples with and without photoresist coatings. The absence of third- and fourth-gap states for injection at the gap edge in our experiments (as well as in Refs. 4 and 5) is also consistent with the model; the larger A_β which occurs in this case increases $I_{i\gamma}$, making observation of the third-gap state more difficult. Using the model and experimental data, we estimate the effective width of the boundary between regions to be ~ 1 μm. The theory of Scalapino and Huberman[11] indicates a length scale for the spatial inhomogeneity in Al of about 30 μm, much larger than the boundary width, consistent with our assumption of uniform quasiparticle concentrations within regions. The boundary width is proportional to the phonon-trapping factor, consistent with the observed greater smearing of the gap structure in photoresist-coated samples.

The general agreement between the predictions of this model and our experimental observations supports our conclusion that we have observed an intrinsic quasiparticle diffusion instability toward a spatially inhomogeneous nonequilibriun superconducting state.

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Electron-Phonon Enhancement of Electron-Electron Scattering in Al

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The influence of the electron-phonon interaction on electron-electron scattering in simple metals has been described within the framework of Landau Fermi-liquid theory. The predicted electron-electron scattering contribution to the low-temperature resistivity of Al is enhanced by a factor of ~20 by the electron-phonon interaction and is in excellent agreement with recent experiments.

The behavior of the ideal resistivity of Al at low temperatures has been the object of controversy for nearly a decade. Recently van Kempen and co-workers[1-3] have found that below 2 K $\rho_i(T)$ has a T^2 component which they interpreted as being due to electron-electron scattering, i.e., $\rho_i(T) \simeq \rho_{ee}(T) = AT^2$, where $A = 2.8 \times 10^{-15}$ Ω mK^{-2}. This result is qualitatively in accord with earlier