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Nonequilibrium Superconducting States with Two Coexisting Energy Gaps

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(Received 21 November 1978)

The effect of tunnel currents in superconducting junctions on the energy gap is calculated. For certain parameters, two different gaps can exist. The stability of these solutions is investigated and at a certain voltage a "first-order transition" is found. This result explains the experimentally observed inhomogeneous states in superconducting tunnel junctions.

Recent experiments on superconducting tunnel junctions[1,2] suggest that for certain injection currents and voltages a superconductor sustains simultaneously two different values of the energy gap. Existing phenomenological models[3] predict that above a critical density of the excitations n_c, the superconductor has an intrinsic instability with respect to the formation of a spatially inhomogeneous state. This is not in agreement with the experimental results of Gray and Willemsen.[2] These authors interpreted the effect by existing inhomogeneities in the probes, with a lower gap value, which grow with increasing total current.

In this Letter we describe microscopically a superconducting tunnel junction consisting of an *injector* and a *probe* and find that two stable values of the energy gap can exist in the probe. In accord with the experiments, at a certain voltage a first-order phase transition takes place where the part of the probe with the lower gap and larger injection current density grows while the part with the larger gap and lower injection current density decreases. The relative size of the two regions is controlled by the total injection current. This is analogous to a liquid-gas transition at a certain pressure where the relative volumes are controlled by the total volume. Existing spatial inhomogenities will serve as nucleation centers for the low-gap region. To this extent, our result is similar to the model of Ref. 2. However, we find the existence of the two gaps to be an intrinsic property of the superconductor and the size of the low-gap regions grows continuously from zero to the size of the probe. Furthermore, our approach describes the gap enhancement by quasiparticle extraction, investigated in the experiments by Chi and Clarke.[4] We will present a detailed analysis in two limits, near T_c and near $T=0$. Qualitatively, the same results can be obtained for any intermediate temperature.

We assume that the injector is thick and, in contrast to the probe, is not appreciably perturbed by the tunneling processes, and also that the phonons remain in equilibrium. The modification of the density of excitations in the probe affects Δ, the magnitude of the gap in the probe.

(Notice that the current of excitations differs from the electric current since electronlike and holelike excitation contribute with different relative signs.) Following Schmid and Schön,[5] we find δf_E, the deviation of the quasiparticle distribution function in the probe from a thermal distribution $f^0(E)$, from the Boltzmann equation:

$$\mathfrak{N}_1(E)\delta\dot{f}_E - K(\delta f) = P_E - \mathfrak{N}_1(E)[\partial f^0(E)/\partial E]\Delta\dot{\Delta}/E. \quad (1)$$

$\mathfrak{N}_1(E)$ is the normalized BCS density of states and $K(\delta f)$ describes the inelastic electron-phonon scattering. $K(\delta f)$ can be split into a "scattering-out" term $-\tau_E^{-1}\mathfrak{N}_1(E)\delta f_E$, and a "scattering-in" term, an integral operator. The perturbation describing the current of excitations is

$$P_E = 2B\mathfrak{N}_1(E)\{\mathfrak{N}_1^{\,i}(E - eV)[f^0(E-eV) - f^0(E)] + (eV \leftrightarrow -eV)\},$$

where $B^{-1} = R\Omega 8e^2N_0$, R is the resistance of the junction, Ω the volume of the probe, N_0 the normal density of states, and $\mathfrak{N}_1^{\,i}$ refers to the injector. The thickness of the probe is small, and no spatial variations in this direction occur. For the moment we also neglect spatial variations in the junction plane. The effect of δf_E on Δ follows for temperatures near the critical temperature of the probe from the Ginzburg-Landau (GL) equation[5]

$$\pi\dot{\Delta}/8T - \chi\Delta = (\alpha - \beta\Delta^2)\Delta, \quad (2)$$

where $\chi = -\int_{-\infty}^{\infty} dE\, E^{-1}\mathfrak{N}_1(E)\delta f_E$ and $\alpha = (T_c - T)/T_c$, $\beta = 7\zeta(3)/8\pi^2T^2$. We obtain analytic results if we assume that the energy gap of the injector and the voltage are small Δ_i/T, $|eV|/T \ll 1$. The linear term of an expansion of P_E in powers of eV/T is localized in a narrow energy region $|E| \lesssim \Delta_i + |eV| \ll T$. Consequently, the corresponding contribution $\delta f_E^{(1)} = \tau_0 P_E/\mathfrak{N}_1(E)$ to the stationary nonequilibrium distribution, is obtained neglecting the "scattering-in" term and taking the scattering time τ_E at $E = 0$. Depending on the relative values of Δ, Δ_i, and V, $\delta f_E^{(1)}$ describes an increase or decrease of the density of excitations near the gap edge. The resulting $\chi^{(1)}(\Delta, V)$ also changes sign, and both gap reduction or gap enhancement are possible. By contrast, the quadratic term of the expansion of P_E always results in a gap reduction. Since $\delta f_E^{(2)}$ is not localized near the gap edge, the "scattering-in" term cannot be neglected. A variational calculation[6] yields $\chi^{(2)} = -1.4B\tau_0(eV/2T)^2$. This contribution can be interpreted as an effective increase in temperature. For the following discussion we shall neglect it, although it may be important for a quantitative analysis.

In the considered limit $(\Delta, \Delta_i, |eV| \ll T)$, we find

$$\chi^{(1)}(\Delta, V) = -2\theta(\Delta + \Delta_i - |eV|)B\tau_0|eV/2T|g\{(d - |eV|)K(k) + (c-d)\pi(\alpha^2, k)\}, \quad (3)$$

where K and π are complete elliptic integrals of the first and third kind, $g = 2/[(a-c)(b-d)]^{1/2}$, $\alpha^2 = (b-c)/(b-d)$, $k^2 = \alpha^2(a-d)/(a-c)$, and a, b, c, d are the parameters $|eV| \pm \Delta_i$ and $\pm\Delta$ assigned such that $a > b > c > d$. In order to obtain the stationary solution of the GL equation including the nonequilibrium term, we employ graphical constructions. The intersections of $-\chi^{(1)}(\Delta, V)$ and the curve $\alpha - \beta\Delta^2$ yield possible solutions of Δ. A large positive value of $\chi^{(1)}(\Delta, V)$ at $|eV| = \Delta_i - \Delta$ (for $\Delta_i > \Delta$), corresponding to a net extraction of excitations, results in a large gap enhancement at this voltage. This is in qualitative agreement with the experimental results of Chi and Clarke.[4]

For our present problem, larger voltages are of interest. As shown in Fig. 1, $\chi^{(1)}(\Delta, V)$ has a step structure at $|eV| = \Delta_i + \Delta$. This step corresponds to the step in the $I(V)$ characteristic of an ideal semiconductor-insulator-semiconductor tunnel junction at the same voltage, however, $\chi^{(1)}$ is finite at low voltages $|eV| \lesssim \Delta + \Delta_i$ and zero above. In the vicinity of the step, we can approximate Eq. (3) by

$$\chi^{(1)}(\Delta, V) = B\tau_0|eV/2T|\pi(\Delta_i/\Delta)^{1/2}\theta(\Delta + \Delta_i - |eV|). \quad (4)$$

For suitably chosen parameters (e.g., $eV = 2.2\Delta_i$ in Fig. 1) we find three solutions of the GL equation denoted by Δ_1, Δ_2, and Δ_3. In addition $\Delta = 0$ is a solution. From an analysis of the time-dependent equations, we find that $\Delta = 0$ and Δ_2 are unstable, whereas Δ_1 and Δ_3 are locally stable. At low voltages we find only one enhanced superconducting solution, whereas at high voltages the perturbance $\chi^{(1)}$ vanishes. In the inset of Fig. 1, the solutions Δ as a function of the voltage are shown. Obviously, in the range where $\Delta(V)$ has

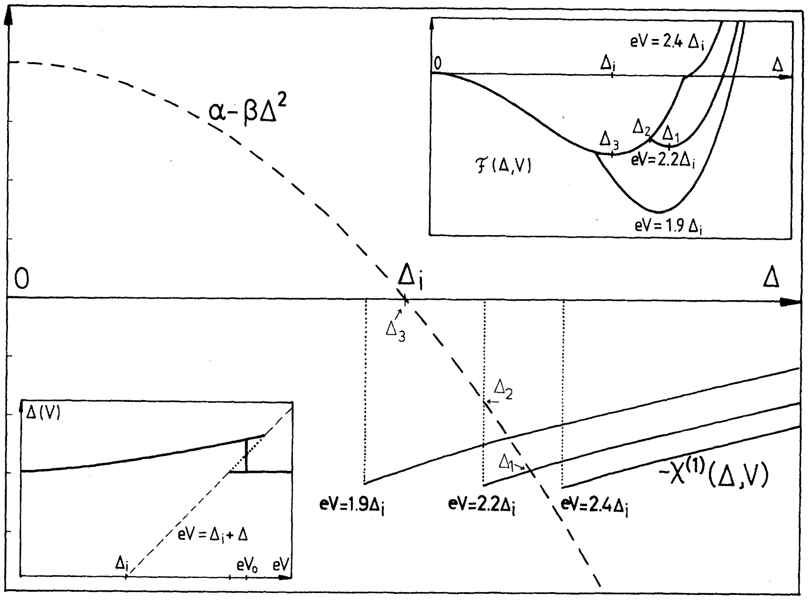

FIG. 1. Graphical solution of the GL equation for voltages $|eV| > \Delta_i$. The constants α, β, and B in $\chi^{(1)}$ are chosen arbitrarily, the higher-order term $\chi^{(2)}$ is neglected. The unperturbed solution is chosen to be $\Delta_0 = \Delta_i$. $\chi^{(1)}(\Delta,V)$ has a step at $\Delta = |eV| - \Delta_i$. For the same parameters, the generalized free energy $\mathfrak{F}(\Delta,V)$ is shown in the upper inset, while the solutions $\Delta(V)$ are shown in the lower inset.

two values with $\Delta_i + \Delta_1 > |eV| > \Delta_i + \Delta_3$, two significantly differing values of the current density are obtained. The enhancement of the gap, as long as $|eV| < \Delta_i + \Delta$, was also found by Peskovatskii and Seminozhenko[7] and by Chang.[8] However, in both papers the possibility of two different gap solutions was not noticed.

In order to find which of the two locally stable solutions is globally stable, we follow the analysis performed by Schmid.[9] The probability of a solution Δ is given by $W(\Delta) \propto \exp(-\mathfrak{F}(\Delta)/T)$ where the generalized free energy is $\mathfrak{F}(\Delta) = -2N_0\Omega \times \int_0^\Delta d\Delta'[\alpha - \beta\Delta'^2 + \chi(\Delta')]\Delta'$. A plot of $\mathfrak{F}$ is given in the inset of Fig. 1. The situation is clearly analogous to a first-order phase transition. At low voltages, Δ_1 is the globally stable "phase." At a certain voltage V_0, which for small perturbations is $|eV_0| = \Delta_i + (\Delta_1 + \Delta_3)/2$, we have $\mathfrak{F}(\Delta_1) = \mathfrak{F}(\Delta_3)$, and a transition between Δ_1 and Δ_3 can take place. With increasing total current, the size of the region in the Δ_1 "phase" with small injection-current density decreases in favor of the region in the Δ_3 "phase" with large injection-current density. At high enough voltages, the Δ_3 "phase" is globally stable. In addition, the metastable states will result in a hysteresis under suitable conditions. The effect of $\chi^{(2)}$ is to reduce Δ_1 and Δ_3 below the values found here, but the difference $\Delta_1 - \Delta_3$ is preserved.

Even in the state where the two gaps coexist, the distribution function δf_E (in the limit considered) is spatially homogeneous. Therefore, spatially inhomogeneous problems are described simply by adding the space derivatives to the GL equation. We find[10,11] that at $V = V_0$ a stationary wall separating the two phases can exist. Apart from the fact that the location of the wall can be shifted arbitrarily, this solution is locally stable. This confirms our result of the stable coexistence of the two phases at the voltage $V = V_0$. On the other hand, droplets or periodic structures correspond to saddle-point solutions and are therefore unstable.

In the low-temperature limit ($\Delta, \Delta_i \gg T$) qualitatively similar results can be found. Particularly interesting are injection voltages close to the sum of the two gaps: $[(\Delta_i + \Delta) - |eV|]/\Delta \ll 1$. In this case P_E and consequently δf_E are localized near the gap edge. Integrating the Boltzmann equation with respect to the energy, we find an equation for the total number of excitations per unit volume $n = 4N_0 \int_0^\infty dE\, \mathfrak{N}_1(E)\delta f_E$ in the form of a linear Rothwarf-Taylor equation.[12] In the stationary case,

$$2n/\tau_R(E = \Delta) = 4N_0 \int_0^\infty dE\, P_E = |I(V)/e\Omega|. \qquad (5)$$

In deriving Eq. (5), the localization of δf_E allowed us to neglect the energy dependence of the recombination rate and take its value at the gap edge[13] $\tau_R^{-1}(E=\Delta)=1.19\tau_0^{-1}(\Delta/T_c)^3(T/\Delta)^{1/2}\exp(-\Delta/T)$. Also, within corrections of order $\exp(-\Delta/T)$, $\int_0^\infty dE\,P_E$ is proportional to the injection current. The effect of nonequilibrium excitations on the order parameter follows from the self-consistency relation $\Delta=\lambda\Delta\int_0^{\omega_D}dE\,E^{-1}\mathfrak{N}_1(E)[\tanh(E/2T)-2\delta f_E]$. Again the localization of δf_E allows us to simplify this equation by setting $(\Delta/E)\delta f_E\approx\delta f_E$. Thus, to lowest order in $\epsilon=(\Delta-\Delta_0)/\Delta_0$, where Δ_0 is the unperturbed $T\approx 0$ gap, we obtain

$$\epsilon=-(n/2N_0\Delta_0)\theta((|eV|-\Delta_0-\Delta_i)/\Delta_0-\epsilon). \qquad (6)$$

The step in the injection current at the voltage $|eV|=\Delta+\Delta_i$ leads to a corresponding step in n, which is made explicit in Eq. (6). For suitably chosen parameters, we find again two locally stable solutions $\epsilon_0=0$ and $\epsilon_1=-n/2N_0\Delta_0$. The physical interpretation of the two solutions is straightforward. First we notice that near $T=0$ there are no excitations which could be extracted from the system, and an injected current increases their number. At a voltage $|eV|$ slightly below $\Delta_i+\Delta_0$, the probe can be either in the unperturbed gap state Δ_0 with no current or in a low-gap state Δ_1 with finite injection current and consequently increases excitation number stabilizing the low-gap value. Again, in a current biased experiment, the relative size of the regions with the different gaps is controlled by the total injection current. The value of the voltage V_0 at which this transition occurs is fixed by the condition that a stable wall exists separating the two regions. If we assume that $\Delta(x)$ is determined by the local value of $n(x)$ and include in Eq. (5) the spatial derivatives, we find[10,14] $eV_0=\Delta_i+(\Delta_0+\Delta_1)/2$.

Our results are in good agreement with the experiments[1,2]: (1) While the tunnel junction is biased at a point on the vertical part of the current voltage characteristic $I(V)$, the probe splits into regions with two different gaps; (2) the difference between the two gaps is proportional to the junction conductivity; (3) the hysteresis along the lower branch of $I(V)$ has been detected. Near T_c, as long as the condition $|eV|\approx\Delta_i+\Delta\ll T$ is satisfied, we find the two coexisting gaps to have enhanced and unperturbed values respectively. The correction $\chi^{(2)}$ in an expansion in $|eV|/T$ results in a reduction of both gap values. This is important for a quantitative comparison with experiments. At low temperatures, we even find that the two gaps have unperturbed and reduced values respectively. At any intermediate temperature the strong voltage dependence of the tunneling processes at $|eV|\approx\Delta_i+\Delta$ results in a steplike modification of the quasiparticle distribution. Thus, we expect that the coexistence of two gaps can be found at all temperatures below T_c and the values of the gaps will be intermediate between the limiting results. For a quantitative comparison with experiment one should also take into account that nonthermal phonons are created[8] which break pairs and further reduce the gap.

In the case of stimulation of superconductivity by microwaves or by phonons at a certain temperature, two stable solutions are also found[7] (normal and superconducting or both superconducting with different gaps, respectively). However, the transition between these solutions occurs abruptly, since there is no external variable, such as the total injection current in the present problem, to control the transition.

It is a pleasure to acknowledge stimulating discussions with V. Ambegaokar, U. Eckern, A. Schmid, M. Schmutz, and E. Siggia. This work was supported in part by the U. S. Office of Naval Research under Contract No. N00014-78-C-0666, Technical Report No. 2.

This work was started while one of us (G.S.) was a visitor at Cornell University.

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