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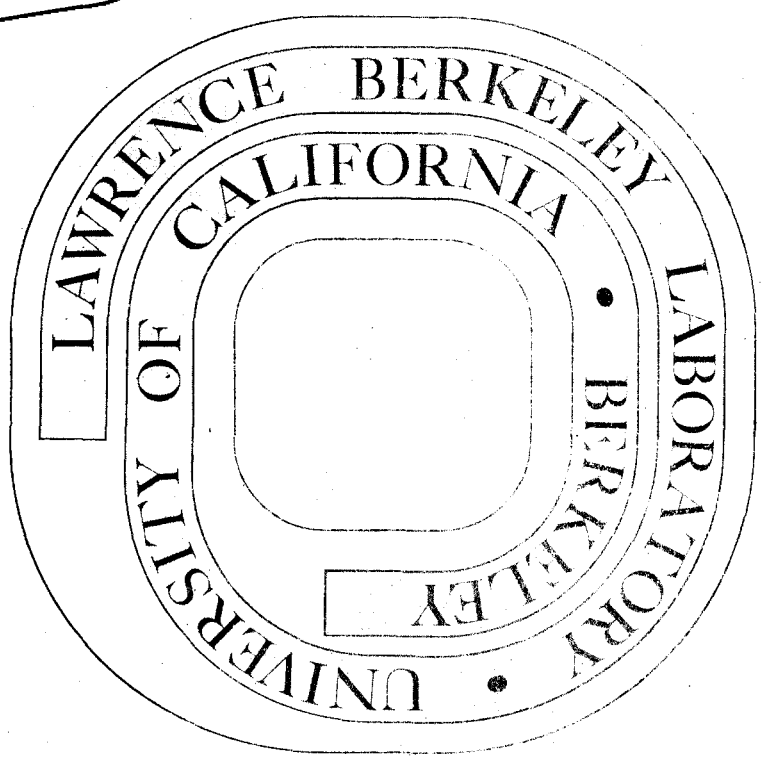

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Theory of Pair-Quasiparticle Potential Difference in Non-Equilibrium Superconductors*

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ABSTRACT

A theory is given of the observable potential difference between pairs and quasiparticles due to the imbalance in the populations of the electron-like and hole-like branches of the excitation spectrum of a superconductor, caused by injection of a quasiparticle current.

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In the preceding Letter,[1] it was shown experimentally that when a quasiparticle current is converted into a pair current in a superconductor, there is a quasiparticle potential in the non-equilibrium region that differs from the chemical potential of the pairs. In this Letter, we calculate the form and magnitude of this potential difference.

The non-equilibrium processes are assumed to occur uniformly in a superconductor S of volume Ω (Fig.1). An electron current I injects electrons via the quasiparticle junction N'S and extracts pairs via the

3f

Josephson[2] junction SS'. A superconducting probe S_p, weakly coupled to S through a second Josephson junction SS_p, measures the pair chemical potential μ_p in S, while a normal probe N_p, in weak contact with S via the quasiparticle junction SN_p, measures the quasiparticle potential. Any emf V between the two probes is measured by a null method that draws no current. The four tunnel junctions ensure that the non-equilibrium processes do not spread significantly into the other conductors, and in addition that only electrons, and not pairs, may be exchanged between S and N_p.

An electron of energy E_k injected from N' into S has a probability $u_{k>}^2$ of entering the electron-like branch ($k>k_F$) and a probability $u_{k<}^2 = v_{k>}^2$ of entering the hole-like branch ($k<k_F$) of the excitation spectrum. (See Fig.2) Here[3] $u_k^2(\xi_k) = \frac{1}{2}(1+\xi_k/E_k) = v_k^2(-\xi_k)$, and $E_k = (\Delta^2 + \xi_k^2)^{\frac{1}{2}}$; $k_>$ and $k_<$ refer to the two states with $\xi_k = \pm(E_k^2 - \Delta^2)^{\frac{1}{2}}$. We define $n_>$ and $n_<$ as the quasiparticle populations per unit volume of the respective branches. The quantity $Q = n_> - n_<$ then represents the excess population of the electron-like branch over the hole-like branch, and, as we shall show, is closely related to V. If Q is disturbed from its equilibrium value (zero), it relaxes with a characteristic time τ_Q, the <u>branch mixing time</u>. Branch mixing occurs through scattering processes governed by the coherence factor[3] $(u_{k<}\, u_{k>} - v_{k<}\, v_{k>})^2$. This coherence factor vanishes for elastic scattering in an isotropic uniform superconductor, since $u_{k<} = v_{k>}$ and $v_{k<} = u_{k>}$, for any $k_<$ and $k_>$ having the same E_k, and hence connected by elastic scattering. Thus, in this simple case, branch mixing is forbidden. Branch mixing <u>can</u> occur through inelastic scattering processes, in which $E_{k>} = E_{k<} \pm E_q$, where E_q is the energy of the phonon emitted or absorbed in the scattering process.

Branch mixing can also occur by elastic scattering processes if the gap is anisotropic or if the gap is spatially inhomogeneous (as usually happens near the surface[4]), since in these cases the symmetry relations between $u_{k<}$ and $v_{k>}$ no longer hold. The relative importance of the elastic and inelastic branch mixing processes depends on the temperature and on the properties of S[5].

Once the two branches have come into equilibrium with each other, their total population ($n_> + n_<$) may still be out of thermal equilibrium with the condensate. The excess recombine to form pairs with a recombination time[6] τ_R that is greater than τ_Q. Both τ_Q and τ_R are usually much longer than the relaxation time of the superfluid, τ_{GL}. Contrary to the theory of Rieger *et al*[7], the bottleneck in the equilibration process of our theory is τ_Q, rather than τ_{GL}.

We calculate the potential V developed between N_p and S_p by a current I that tends to increase the electron-like quasiparticle population and decrease the pair population[8]. The reduction in the number of pairs has the effect of decreasing k_F. However, a space charge is created so that the chemical potential of the pairs (including the electrostatic potential) is restored everywhere to its equilibrium value, μ_p; we can refer all voltages to this value. The electron injection generates perturbations $\delta f_{k>}$ and $\delta f_{k<}$ on the electron- and hole-like branches, where the δf_k do not necessarily refer to thermal equilibrium. The current through the junction SN_p when N_p is maintained at a potential μ_p/e is[9]

$$\delta I = \frac{G_{NN}}{e}\left\{\int_\Delta^\infty \frac{E_{k>}}{(E_{k>}^2-\Delta^2)^{\frac{1}{2}}}\,(v_{k>}^2 - u_{k>}^2)\,\delta f_{k>}\,dE_{k>} + \int_\Delta^\infty \frac{E_{k<}}{(E_{k<}^2-\Delta^2)^{\frac{1}{2}}}\,(v_{k<}^2 - u_{k<}^2)\,\delta f_{k<}\,dE_{k<}\right\}. \qquad (1)$$

G_{NN} is the tunneling conductance[10] for the junctions SN_p when S is normal. Since $E(E^2 - \Delta^2)^{-\frac{1}{2}} = E/|\epsilon|$, and $(v_{k>}^2 - u_{k>}^2) = -(v_{k<}^2 - u_{k<}^2) = |\epsilon|/E$, (1) reduces to

$$\delta I = \frac{G_{NN}}{e} \int_{\Delta}^{\infty} (\delta f_{k>} - \delta f_{k<}) dE_k = \frac{G_{NN} Q^*}{2N(0)e} , \tag{2}$$

where $$Q^* = 2N(0) \int_{\Delta}^{\infty} (\delta f_{k>} - \delta f_{k<}) dE_k . \tag{3}$$

$N(0)$ is the density of states at the Fermi level for electrons of one spin. The voltage required between the two probes to null the current is then $V = \delta I/G_{NS}$, where G_{NS} is the tunneling conductance of the junction SN_p. The measured voltage is therefore

$$V = \frac{Q^*}{2N(0)e g_{NS}} , \tag{4}$$

where $g_{NS} = G_{NS}/G_{NN}$ is just the normalized tunneling conductance[10] for an SN junction in the low voltage limit. We see immediately that V is proportional to Q^*; an excess quasiparticle population with $Q^* = 0$ does <u>not</u> give rise to a quasiparticle potential different from μ_p.

Eq.(4) is quite general, and does not require the two branches to be separately in thermal equilibrium. To see under what circumstances separate thermal equilibrium does occur, we must consider the tunneling and relaxation processes in more detail. A simple case to consider is electron injection at high bias voltages ($\gg \Delta/e$), when the majority of the excitations will be electron-like. High energy excitations decay rapidly by phonon emission[11] into lower energy states. A study of the coherence factors indicates that the ratio of the probability of a quasiparticle changing branches to the probability of its staying on its own branch is roughly Δ/E_i, where E_i is the initial energy. Thus the high energy excitations mostly remain on their own branch during the first inelastic process. Subsequent scattering processes

tend to bring each branch separately into thermal equilibrium, and also to equalise the populations of the two branches. Near T_c, Δ approaches zero, and the branch mixing process becomes very slow. (It cannot occur at all in the normal state.) We may then assume that the mixing occurs between two populations which are separately near equilibrium.

Hence, near T_c the definition of a chemical potential for each branch becomes meaningful. For each branch we may then write $\delta f_k = -(\partial f_k / \partial E_k)\delta\mu$, where $\delta\mu$ is the displacement of the corresponding chemical potential from μ_p, and f_k is the Fermi function. For such δf_k, (3) and (4) lead to

$$V = \frac{\mu_> - \mu_<}{2e} \frac{Q^*}{Q} . \tag{5}$$

But when chemical potentials are defined, Q and Q* are related by

$$\frac{Q^*}{Q} = \int_\Delta^\infty -\left(\frac{\partial f}{\partial E}\right) dE \Bigg/ \int_\Delta^\infty \frac{E}{(E^2 - \Delta^2)^{\frac{1}{2}}} \left(-\frac{\partial f}{\partial E}\right) dE = \frac{2f(\Delta)}{g_{NS}} \tag{6}$$

which approaches unity as T approaches T_c, and (5) simplifies to

$$V = (\mu_> - \mu_<)/2e . \tag{7}$$

Note that V is zero[12] if $\mu_> = \mu_<$, even if both differ from μ_p. Near T_c, $\dot{Q}$ due to injection equals $I/e\Omega$ for all bias voltages, $Q^* \approx Q \approx I\tau_Q/e\Omega$, and from (4) we obtain the final result

$$V = \frac{I\tau_Q}{2e^2\Omega N(0) g_{NS}} . \tag{8}$$

Let[9] us now estimate τ_Q. We assume $eV_{inj} \gg kT_c$, so high-energy electron-like quasiparticles dominate the injected population, and $\dot{Q} = I/e\Omega$. First we find how the electrons cool, then how the branch imbalance Q relaxes.

Initially consider the cooling of electrons when the sample temperature T is zero. Then only spontaneous phonon emission occurs,

and the probability per unit time of energy loss between ϵ and $\epsilon + d\epsilon$ is $2\epsilon^2 d\epsilon/\tau_\theta(k\theta)^3$. The quadratic dependence on ϵ results from combining the appropriate density of states with the square of the electron-phonon matrix element, both proportional to ϵ. The maximum energy loss is the Debye energy $k\theta$. τ_θ is the scattering time at $T = \theta$, as inferred from electrical or thermal conductivity. If we characterize the injected quasiparticle distribution by its mean energy kT^*, we can compute the rate of decrease of T^* due to phonon emission. The result is

$$T^* \approx \theta(3\tau_\theta/16t)^{1/3} \qquad (9)$$

after the electrons have cooled enough that $T^{*3}(t) \ll T^{*3}(0)$. Inserting numerical values for tin ($\tau_\theta = 2 \times 10^{-14}$ sec, $\theta = 200^\circ$K, and $T_c = 3.8^\circ$K), we find that the time required to cool down to T_c is 5×10^{-10} sec. If T is finite, the instantaneous cooling rate is reduced, and the final approach of T^* to T is exponential.

To estimate the rate of Q-relaxation, we take the coherence factor for branch mixing to be zero except for transitions involving a state within $\sim\Delta$ of the bottom of the distribution. Near T_c, where $\Delta \ll kT$, mixing is slow, and we may assume that T^* has reached T_c before Q relaxes. In that case, approximately $\Delta(T)/kT_c$ of all transitions involve branch crossing, and we find[13]

$$\tau_Q = \frac{0.068\tau_\theta(\theta/T_c)^3}{\Delta(T)/\Delta(0)} = \frac{2 \times 10^{-10}\text{ sec}}{\Delta(T)/\Delta(0)} . \qquad (10)$$

The temperature dependence is as found experimentally[1]. Considering the crudeness of the model, the numerical agreement of the coefficient ($\frac{1}{2}$ the measured value) is quite reasonable. The fact that (10) fits the data even for $T \ll T_c$, where the assumption of equilibration of T^*

at T_c is inappropriate, may be explained as follows: For $T \ll T_c$, $\Delta \approx \Delta(0) = 1.76kT_c$; thus, even somewhat before T^* has reached T_c, all phonon emission processes have roughly 50% probability of branch crossing. Hence, Q relaxes while the injected electrons are cooling through the vicinity of T_c, no matter how low T is. In this dynamic situation, the chemical potentials $\mu_>$ and $\mu_<$ are not really well-defined or useful concepts.

The detailed computation[9] of this simultaneous cooling and Q-relaxation process turns out to be rather delicate and model-dependent. Moreover, any residual gap anisotropy not destroyed by the short mean free path[14] provides an additional Q-relaxation mechanism. For a typical mean free path $\ell_o = 1000$Å, we estimate that the r.m.s. residual gap anisotropy is roughly 1%. Near T_c, where Δ is small, its contribution to $1/\tau_Q$ is negligible compared to that of the phonons, but for $\Delta = \Delta(0)$ and $T^* \approx T_c$, its contribution is estimated to be of the same order of magnitude as that of the phonon mechanism. Moreover, its contribution increases as T^* decreases, while the phonon mechanism decreases as T^{*2}. Thus even a tiny residual gap anisotropy will assure that Q relaxes before T^* falls much below T_c.

We have not attempted corrections for the difference between Q^* and Q. Very near T_c, $Q^* \approx Q$. In the low temperature regime, Q relaxes largely while T^* is still above T_c, where Q^*/Q typically lies in the range 0.7-1.0. Thus the error due to this source is probably smaller than the uncertainty in the calculation of τ_Q in the low temperature regime, and we use (8) at all temperatures.

We are pleased to acknowledge the hospitality of the Cavendish Laboratory during the course of this work, and to thank Professor A.B. Pippard, Dr. J.R. Waldram, Dr. C.J. Adkins, and Dr. B.D. Josephson for numerous helpful conversations.

FOOTNOTES

1. J. Clarke, preceding Letter.
2. B.D. Josephson, Phys. Letters 1, 251 (1962).
3. J. Bardeen, L.N. Cooper, and J.R. Schrieffer, Phys. Rev. 108, 1175 (1957).
4. W.L. McMillan and P.W. Anderson, Phys. Rev. Letters 16, 85 (1966).
5. For a discussion, see W.A. Phillips, Proc. Roy. Soc. A309, 259 (1969).
6. The total excess population, and hence τ_R, may be measured by a tunnel junction biased at a non-zero voltage: e.g., K.E. Gray, A.R. Long, and C.J. Adkins, Phil. Mag. 20, 273 (1969).
7. T.J. Rieger, D.J. Scalapino, and J.E. Mercereau, Phys. Rev. Letters 27, 1787 (1971).
8. We assume the dimensions of S (Fig.1) are small compared with the characteristic length over which branch mixing occurs, $\lambda = (\ell_o v_F \tau_Q)^{\frac{1}{2}}$, introduced by A.B. Pippard, J.G. Shepherd, and D.A. Tindall [Proc. Roy. Soc. A324, 17 (1971)]. ℓ_o is the mean free path.
9. M. Tinkham, to be published.
10. D.H. Douglass,Jr., and L.M. Falicov, Prog. Low Temp. Phys. IV (Ed. C.J. Gorter), North-Holland, Amsterdam (1964).
11. Phonons emitted with energies $> 2\Delta$ have a high probability of exciting a pair into two quasiparticles. We assume that this process populates the two branches equally, so that Q is unaffected.
12. A small contribution to V from the increase of $n_> + n_<$ might be expected because of the energy dependence of N(0).

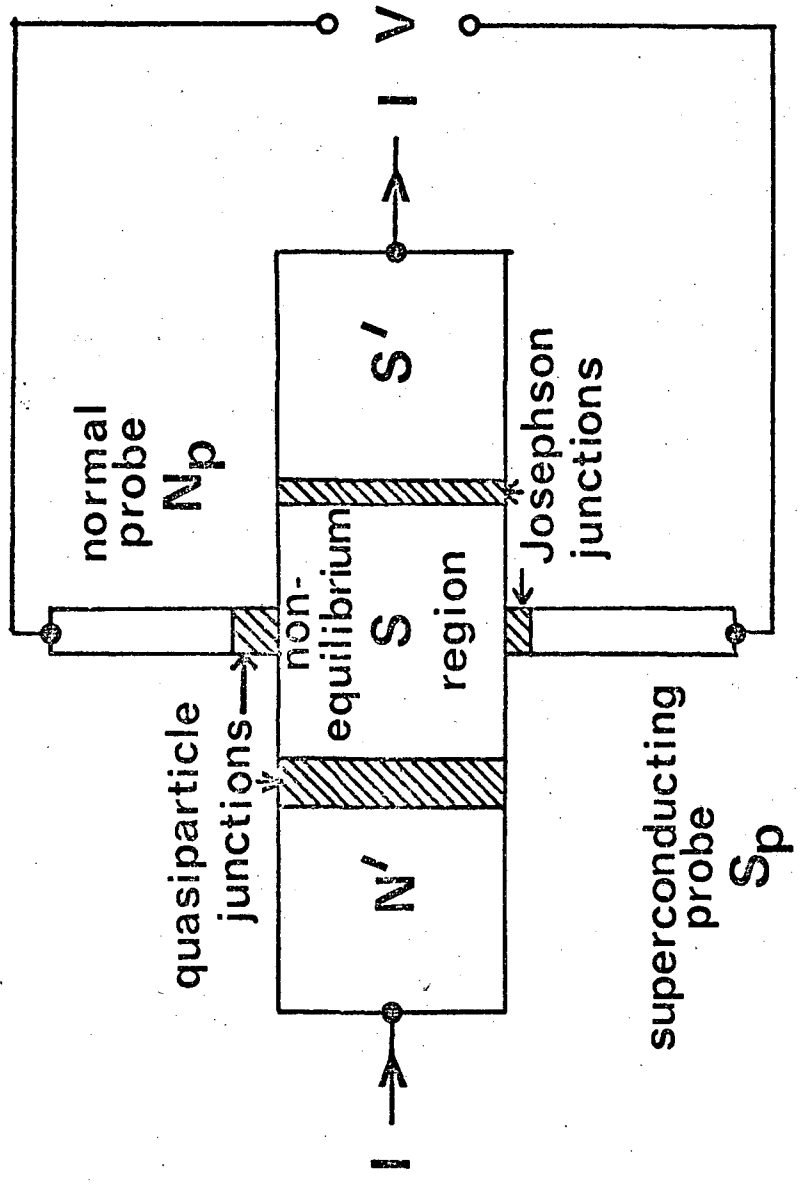
13. Very close to T_c, (10) will become longer than τ_R, which therefore may represent an upper limit to τ_Q.

14. P.W. Anderson, J. Phys. Chem. Solids 11, 26 (1959); D. Markowitz and L.P. Kadanoff, Phys. Rev. 131, 563 (1963).

FIGURE CAPTIONS

Fig. 1. Schematic diagram of non-equilibrium experiment. Quasiparticles are injected into S from N', and pairs extracted into S'. S_p measures the pair chemical potential in S, while N_p measures the quasiparticle potential.

Fig. 2. Excitation spectrum of superconductor with energies referred to μ_p. There are $n_>$ excitations on the electron-like branch ($k > k_F$), and $n_<$ on the hole-like branch ($k < k_F$). The imbalance $Q = n_> - n_<$.


V
I
S'
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probe
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Josephson
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FIG. 1

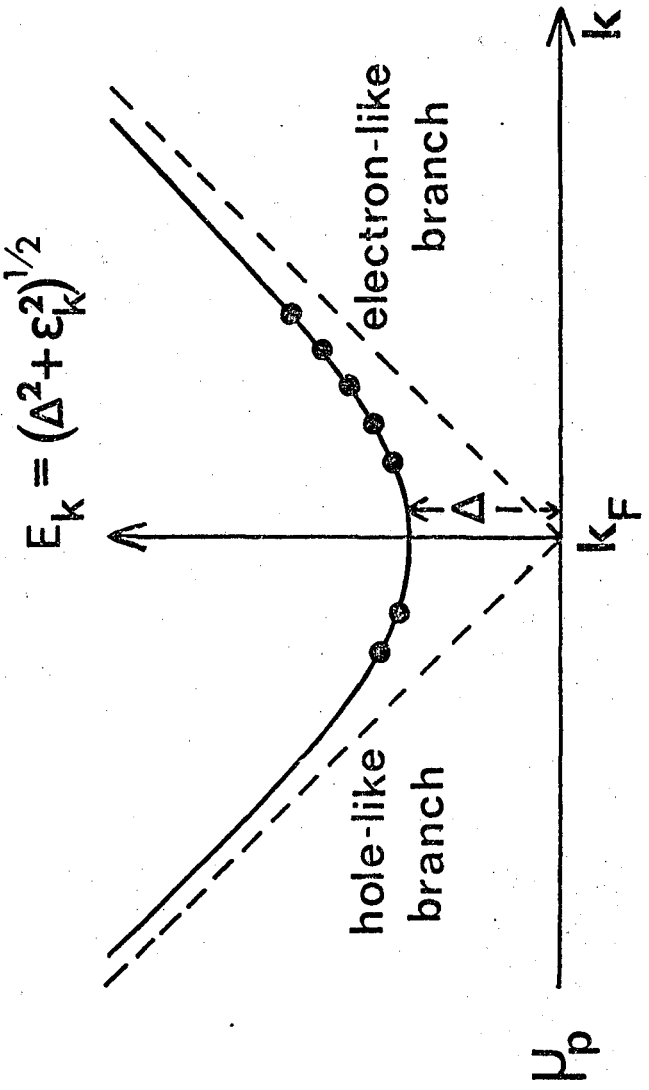
$E_k = (\Delta^2 + \varepsilon_k^2)^{1/2}$
electron-like branch
hole-like branch
Δ
k_F
k
μ_p

FIG. 2

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