like a solid, and the values of R_{xx} and R_{yy} depend on the thickness of the sample.

To demonstrate the effect of the rotation of the polarization of the shear wave, it is possible to perform measurements of the nondiagonal elements of Z by using crossed transducers. Such results will be soon published.

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[1]F. Brochard, J. Phys. (Paris) 32, 685 (1971).
[2]J. L. Ericksen, Arch. Ration. Mech. Anal. 4, 231 (1960), and 9, 37 (1962).
[3]F. M. Leslie, Quart. J. Mech. Appl. Math. 19, 357 (1966), and Arch. Ration. Mech. Anal. 28, 265 (1968).
[4]P. Martinoty and S. Candau, Mol. Cryst. Liquid Cryst. 14, 243 (1971).
[5]P. E. Cladis and M. Kleman, to be published.

Experimental Observation of Pair-Quasiparticle Potential Difference in Nonequilibrium Superconductors*

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It is shown experimentally that when a quasiparticle current is converted into a pair current in a superconductor, the quasiparticle potential in the nonequilibrium region differs from the pair chemical potential.

In a recent Letter, Rieger, Scalapino, and Mercereau[1] developed a theory of nonequilibrium superconductivity. They considered a current I flowing through a superconductor S of volume Ω so that quasiparticles were injected and pairs extracted, and found that the pair and quasiparticle chemical potentials (μ_p and μ_{qp}) differed by

$$(\mu_{qp}-\mu_p)/e = I\tau_{\rm GL}/24e^2\Omega N(0). \quad (1)$$

In (1), $\tau_{\rm GL}$ is the Ginzburg-Landau relaxation time, and $N(0)$ the density of states at the Fermi level for electrons of one spin.

In this Letter, we show experimentally that the quasiparticle potential in a nonequilibrium superconductor differs from μ_p/e; however, the data do not support (1) in detail. In the following Letter,[2] a new theory is presented which is in good agreement with the experimental results. In general, the quasiparticle chemical potential is not a well-defined quantity, and is replaced by a "quasiparticle potential" which arises from the imbalance of electronlike and holelike excitations. Throughout S, μ_p is constant (as in Ref. 1), and the difference between the quasiparticle potential and μ_p/e is shown to be[2]

$$V = I\tau_{\rm Q}/2e^2\Omega N(0)g_{NS}. \quad (2)$$

In (2), $\tau_{\rm Q}$ is the relaxation time for the electronlike and holelike imbalance,[2] and g_{NS} is the normalized conductance[3] of an NS tunnel junction in the low-voltage limit. This result should be a good approximation over all temperatures.

To observe these nonequilibrium effects, we require a superconductor S of small volume into which quasiparticles are injected from a normal electrode and from which pairs are extracted into a superconducting electrode. The quasiparticle potential is measured by a normal probe which exchanges single electrons with S, while μ_p/e is measured by pair exchange with a superconducting probe. If the normal injection electrode were in good metallic contact with S, electrons incident on the interface from the normal metal with energies less than the energy gap Δ would undergo Andreev[4] reflection as holes, and pairs would be transmitted into the superconductor. Thus significant nonequilibrium effects would be observed only when $\Delta \lesssim kT$. Furthermore, good metallic contact of either the normal probe or normal injection electrode with S could substantially depress the condensation amplitude in S by means of the proximity effect.[5] These difficulties may be circumvented by coupling both normal metals to S through tunnel junctions. Ideally,[2] one would also like to couple the superconducting probe and electrode to S via Josephson[6] junctions, so that the nonequilibrium processes would be confined to S. In practice, this configuration would be difficult to fabricate, and in the experiment only the normal electrode and probe were coupled via junctions. However, the geometry was such that the quasiparticle-pair conversion did take place in a well-defined volume.

A strip of Al (XX') (Fig. 1) of width $d \sim 3$ mm and thickness ~1500 Å was evaporated onto a glass slide and oxidized. Next, a strip (YY') of Sn (thickness t 2000 to 5000 Å) was evaporated across the Al strip to form a tunnel junction. The volume of Sn overlaying the Al was the region where the quasiparticle-pair conversion occurred. The sample was removed from the evaporator, so that the Sn oxidized somewhat, and a layer of varnish applied, leaving a window about 1 mm square in the middle of the junction. A diagonal strip (ZZ') of Cu was then evaporated to form the normal probe. To reduce the resistance of this probe and so make possible very low-voltage measurements, a 5000-Å film of Pb was deposited over the Cu. The Cu was sufficiently thick (>1 μm), and sufficiently dirty, that no Josephson tunneling[6,7] between the Sn and Pb was possible. The resistance of the Al-Sn injection junction, typically 10 Ω, was much higher than the resistance of the Al strip forming the junction, so that quasiparticles were uniformly injected into the Sn. The barrier between the Sn and Cu was of much lower resistance, but was sufficiently thick to effectively quench the proximity effect; this result was verified in a separate experiment.

The voltage V between Y and Z was measured with a superconducting galvanometer[8] in series with a resistor by means of a null-balancing technique, the whole circuit being immersed in liquid helium. The resistance of the Sn-oxide-Cu-Pb junction was determined by applying a current between Y' and Z' and measuring the voltage between Y and Z. This resistance was dominated by the oxide layer, and was typically 10^{-5} Ω at the transition temperature T_c of the Sn. As the temperature was lowered, the resistance of the junction increased at approximately the rate predicted for an SN tunnel junction.[3] At the lower temperatures, the resolution of the voltmeter was seriously reduced by the high resistance thus introduced into the circuit, and relatively high injection currents (I) were required, typically 5 to 20 mA. Near T_c, the currents used were 0.1–1 mA. Thus at all temperatures the voltage across the Al-Sn junction was much greater than Δ and proportional to I.

Data were rejected from samples in which the current-voltage characteristics of the Al-Sn junction indicated that metallic shorts might be present, and the quasiparticle injection therefore highly nonuniform. Average values of V from acceptable samples[9] for each thickness of Sn are shown in Fig. 2. For electron injection into the Sn (Al negative relative to Sn), the Cu probe was negative with respect to μ_p. Near T_c, V reversed exactly when I was reversed, and was linear in I. Below about $0.8T_c$, V did not

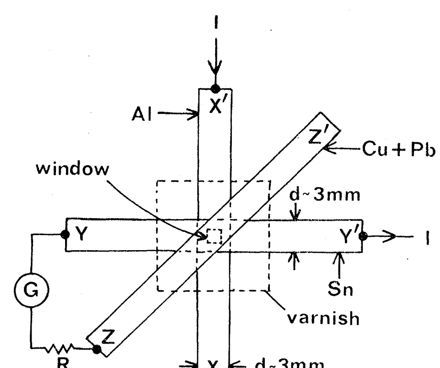

FIG. 1. Sample configuration. In order of deposition, the films are Al (XX'), Sn (YY'), varnish, Cu (ZZ'), and Pb (ZZ'). Galvanometer G and resistor R measure the potential difference V between Y and Z.

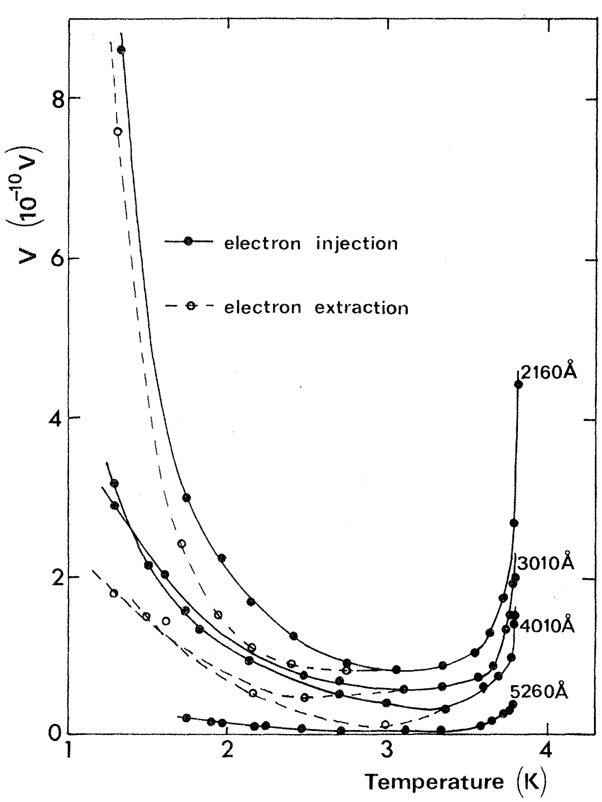

FIG. 2. Potential difference V between Y and Z for four thicknesses of Sn versus temperature. V is normalized to the injection current (I) of 1 mA. Sample area, 0.1 cm^2.

reverse exactly, $|V|$ being larger for electron injection than for electron extraction. This asymmetry is thought to be due to the energy dependence of $N(0)$ which has been neglected in both theories. The rapid rise in V as the temperature is lowered below $0.8T_c$ is explained by the presence of g_{NS} in the denominator of (2). This feature is absent from (1), which is intended to be valid only near T_c, where $g_{NS} \simeq 1$.

To facilitate comparison of the two theories, it is convenient to multiply the right-hand side of (1) by g_{NS}^{-1}, and to compute the quantity $\zeta = V\Omega g_{NS} I^{-1}$ from the experimental data. We then compare ζ in turn with the two expressions $\tau_{GL}/24e^2N(0)$ from (1), which varies as Δ^{-2} near T_c, and $\tau_Q/2e^2N(0)$ from (2), which varies[2] as Δ^{-1} near T_c. In Fig. 3 we have plotted ζ for data taken from the three thinnest samples averaged over the asymmetry at low temperatures. V appears to be proportional to Ω^{-1}, as predicted by both theories. The temperature dependence of the data reflects the behavior of the characteristic time. The solid curve represents Δ^{-1}, fitted to the measured T_c (3.81 K), and the average value of ζ at low temperatures. The fit is surprisingly good over the whole temperature range. For comparison, the crosses indicate a Δ^{-2} curve, fitted in the same way: Acceptable agreement with experiment could be obtained only by choosing $T_c \simeq 4.0$ K, a value much higher than that observed experimentally. It appears that a characteristic time porportional to Δ^{-1}, rather than Δ^{-2}, fits the data more adequately, a conclusion that supports the validity of (2) over that of (1).

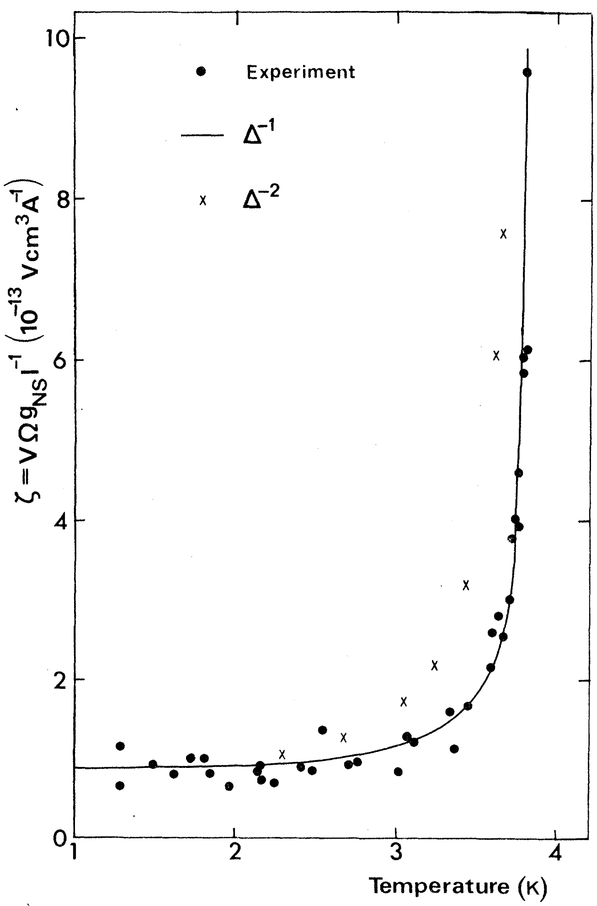

FIG. 3. Plot of $\zeta = V\Omega g_{NS} I^{-1}$ versus temperature for the three thinnest samples.

From (1) and the low-temperature data of Fig. 3, we find that the Ginzburg-Landau time required to fit the data would be approximately 5×10^{-9} sec, about 2 orders of magnitude higher than any acceptable value. From (2) and the data of Fig. 3, we find that in dirty Sn, $\tau_Q = 4 \times 10^{-10}\, \Delta(0)/\Delta(T)$ sec, where $\Delta(T)/\Delta(0)$ is the normalized gap. This result is in satisfactory agreement with the theoretical estimate.[2] The characteristic length[10] over which quasiparticle-pair conversion occurs, $\lambda = (l_0 v_F \tau_Q)^{1/2}$, is roughly 5 μm at low temperatures, where we have taken the mean free path l_0 as 1000 Å.

It might be remarked that an experiment performed by Ginsberg[11] in an attempt to measure the *recombination* time of injected quasiparticles, in fact would have demonstrated the effects described here if the voltage resolution had been high enough.

Notice also that the configuration of Fig. 1 represents a "superconducting transistor": A current between X' and Y' develops a voltage across Y (or Y') and Z. The device is of course passive and achieves no power gain, but could possibly be used as an impedance transformer.

Finally, we consider the implications of these experiments for the determination of e/h using the Josephson effect.[12] Electromagnetic radiation of frequency ω induces constant-voltage current steps on the characteristic of a Josephson junction whenever $n\hbar\omega = 2\Delta\mu_p$, where $\Delta\mu_p$ is the difference in *pair* chemical potential across the junction, and n is an integer. If the current and voltage leads, which are of course normal,[13] on one side of the junction (or on both sides) are within a distance λ (say), the quasiparticle potential difference measured by the voltage leads will differ from $2\Delta\mu_p/e$, and an error in e/h will result. However, in all published determinations of e/h, the current and voltage leads were well separated, and the errors due to nonequilibrium effects utterly negligible.

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[1]T. J. Rieger, D. J. Scalapino, and J. E. Mercereau, Phys. Rev. Lett. 27, 1787 (1971).

[2]M. Tinkham and J. Clarke, following Letter [Phys. Rev. Lett. 28, 1366 (1972)].

[3]D. H. Douglass, Jr., and L. M. Falicov, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland, Amsterdam, 1964), Vol. IV.

[4]A. F. Andreev, Zh. Eksp. Teor. Fiz. 46, 1823 (1964) [Sov. Phys. JETP 19, 1228 (1964)].

[5]P. G. de Gennes, Rev. Mod. Phys. 36, 225 (1964).

[6]B. D. Josephson, Phys. Lett. 1, 251 (1962).

[7]J. Clarke, Proc. Roy. Soc., Ser. A 308, 447 (1969).

[8]J. Clarke, Phil. Mag. 13, 115 (1966).

[9]Usually, two out of four samples of a given thickness of Sn were acceptable and were reproducible to perhaps 20%. Only one 5260-Å sample exhibited a good Al-Sn junction characteristic, which was of such low resistance that the injection was undoubtedly very nonuniform; these data were not used in Fig. 3.

[10]Since $t \ll \lambda \ll d$, the quasiparticle-pair conversion was uniform throughout the region where the Sn overlapped the Al.

[11]D. M. Ginsberg, Phys. Rev. Lett. 8, 204 (1962).

[12]W. H. Parker, D. N. Langenberg, A. Denenstein, and B. N. Taylor, Phys. Rev. 177, 639 (1969).

[13]Depending on the nature of the junction, the leads may form tunnel junctions or metallic contacts with the superconductors. In the latter case, the nonequilibrium effects will be greatly reduced, except near T_c, as pointed out in the text.

Theory of Pair-Quasiparticle Potential Difference in Nonequilibrium Superconductors*

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

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 nonequilibrium region that differs from the chemical potential of the pairs. In this Letter, we calculate the form and magnitude of this potential difference.

The nonequilibrium 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 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 nonequilibrium 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.

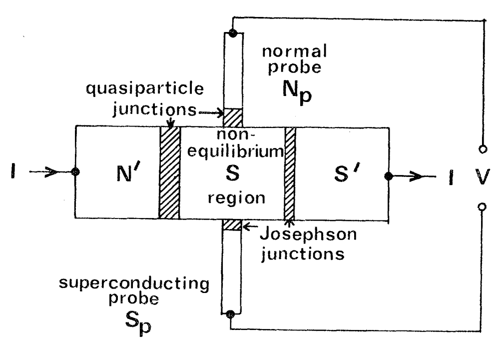

FIG. 1. Schematic diagram of nonequilibrium experiment. Quasiparticles are injected into S from N', and pairs extracted into S'. S_p measures the pair chemical potential in S, while N_p measures the quasiparticle potential.