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Observation of Thermally Induced Potential in a Superconductor

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Potential differences have been measured between a normal probe and a superconducting probe attached to a superconductor held in a temperature gradient. These observations suggest that a thermoelectrically generated quasiparticle current is flowing in the superconductor and allow evaluation of the thermal transport coefficient for the quasiparticles.

The first measurement of a thermoelectric transport coefficient in a superconductor was reported by Meissner in 1927.[1] It was found that a circuit consisting of two metals gave rise to no thermoelectromotive force when both metals were superconducting. This, and many subsequent experiments,[2] have shown that with the exception of flux flow[3] and certain electrostatic phenomena[4] all conventional thermoelectric effects vanish in the superconducting state. However, as Ginzburg[5] first noted, there exists in a superconductor the possibility of a simultaneous flow of a normal current of density $\vec{j}_n = L_T(-\nabla T)$ and a supercurrent $\vec{j}_s = -\vec{j}_n$. This prediction of a "superconducting fountain effect" was qualitatively verified by Clarke and Freake[6]; however, they were unable to obtain quantitative information on the transport coefficient L_T. More recently[7] calculations based on the two-fluid model have predicted that this flow of normal current in a superconductor gives rise to a nonquantized contribution to the magnetic flux in a loop made up of two different superconductors. Experimental data[8-10] indicate the existence of such a magnetic flux with a value of as much as five orders of magnitude[10] larger than predicted by theory. This discrepancy, coupled with the opportunity to study quasiparticle transport and relaxation processes in a nonequilibrium superconductor, prompted us to make measurements of the pair and quasiparticle electrochemical potentials in a superconductor held in a temperature gradient. It has been predicted[11] that in nonequilibrium situations in which the electron and hole branches of the quasiparticle excitation spectrum are unequally populated, the quasiparticles in a superconductor may be described by a different electrochemical potential from that which describes the pairs. This Letter reports our initial results showing the first experimental evidence for a pair-quasiparticle electrochemical potential difference in a superconductor in a temperature gradient.

A normal-metal tunnel junction and superconducting metal probe were placed in a nonequilibrium region of a thin-film superconductor. It has been shown[12] that this allows a direct measurement of the quasiparticle and pair electrochemical potentials to be made. A strip of 99.999%-purity Sn in the form of a "T" (Fig. 1) of width $d \sim 2$ mm, length ~2 cm, and thickness ~2000–4000 Å was evaporated onto a 1-mm-thick sapphire substrate and oxidized. A small rectangle (~2.5

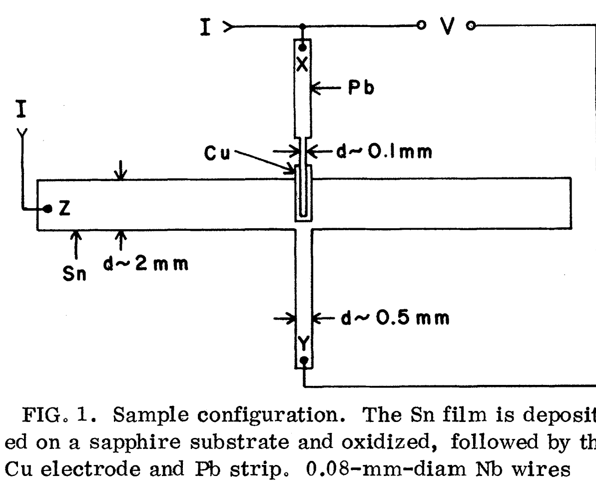

FIG. 1. Sample configuration. The Sn film is deposited on a sapphire substrate and oxidized, followed by the Cu electrode and Pb strip. 0.08-mm-diam Nb wires connected to a superconducting quantum interference device voltmeter allow the potential difference between points X and Y to be measured.

mm ×0.6 mm ×1.5 μm thick) of Cu alloy containing 5% Al was then evaporated in the center of the strip to form the normal probe. Finally, a 3000-Å Pb film was deposited over the Cu to reduce the normal-probe resistance. This was necessary in order to reduce the Johnson noise in the sample and thus enable very low-voltage measurements to be made. The thermal conductivity of the sapphire substrate ensured that the temperature gradient in the vicinity of the junctions was changed by less than 0.1% by the presence of the metal films.

Additional leads attached to the sample enabled the resistance of the normal probe to be measured by applying a current between X and Z (Fig. 1) and measuring the voltage developed between X and Y. Typical junctions had a resistance of $(1-4)\times10^{-5}$ Ω immediately below the T_c of Sn which, as shown in Fig. 2, increased approximately at the rate expected for a normal-superconductor (NS) tunnel junction. The voltage between X and Y was measured with a superconducting quantum interference device voltmeter in a feedback configuration. The resolution of this system was approximately 2×10^{-13} V rms with a 1 sec averaging time. It was also possible to inject a current through the normal probe to verify that Josephson tunneling was not taking place through the Cu. Such tunneling would couple the pair electrochemical potentials in the Sn and Pb, making the measurements described here impossible.

All measurements were conducted in a screened room with Mumetal and superconducting shields reducing the field in the sample vacuum chamber to less than 10^{-6} T. The temperatures of both ends of the sample were measured to within an uncertainty of less than 1 mK with use of calibrated Ge resistors. Any leaks in the vacuum chamber (which would reduce the temperature gradient along the sample) could be readily detected as a reduction of the thermal time constant of the entire system (approximately 15 min).

The voltage developed per unit temperature difference for one sample is shown in Fig. 3. All samples which showed no evidence of a superconducting short through the normal probe exhibited the same behavior shown in this figure, except that the absolute magnitudes varied by as much as a factor of 8. This is not too surprising since we are measuring a quantity proportional to a thermal transport coefficient. This coefficient is extremely sensitive to impurities and can be ex-

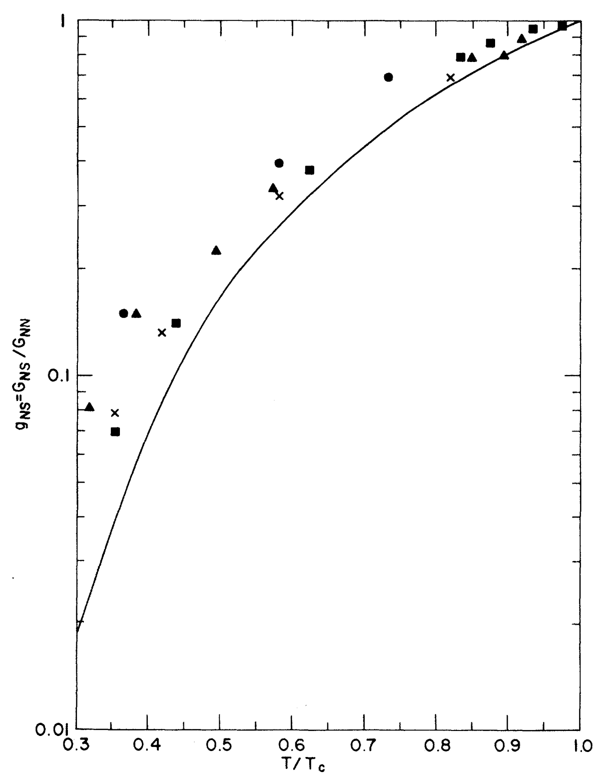

FIG. 2. Normalized low-voltage conductance g_{NS} for four samples as a function of temperature compared with theory (solid line).

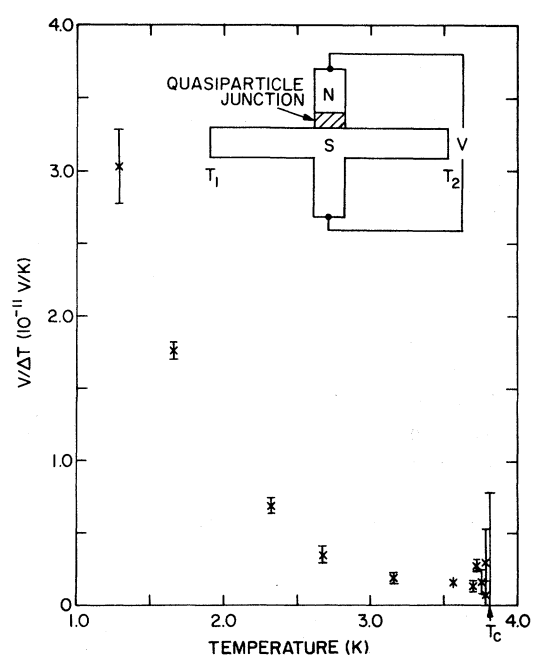

FIG. 3. Potential difference between X and Y per unit temperature difference across the sample vs temperature for a 2510-Å Sn film.

pected to vary widely for thin-film specimens. The data shown in Fig. 3 have the lowest magnitude of those measured. This thermally induced potential is linear in ∇T over at least a factor-of-10 change in the temperature gradient. Reversing the sign of ∇T does not reverse the sign of the observed voltage and leaves the magnitude unchanged to within 15–30%. This failure to reproduce the magnitude of V for opposite sign of ∇T lies outside the experimental uncertainty of the measurements and perhaps is due to a small normal-thermoelectric-voltage contribution from the Cu. Such a conventional themoelectric potential would change sign with ∇T and could cause the asymmetry that we observed.

This potential is reminiscent of that observed by Clarke[12] in his branch-mixing experiment. In that experiment, an imbalance in the population of the electron and hole branches of the excitation spectrum created by tunnel injection led to a potential difference between normal and superconducting probes. Similar effects have also been observed near superconductor/normal boundaries carrying a transport current[13] and within Josephson weak links excited by rf radiation.[14] In all of these experiments, a flow of normal excitations is accompanied by a difference between the quasiparticle and pair electrochemical potentials. Tinkham has shown[11] that such quasiparticle-pair electrochemical-potential differences can only arise from an electron-hole branch imbalance. These results, together with the data presented here, are evidence that such a branch imbalance induced by a thermally generated quasiparticle current is the cause of the potential measured in the present experiment.

To first order, a branch imbalance should not arise in a superconductor in a thermal gradient since the increase in electronlike excitations and decrease in holelike excitations on one side of the Fermi surface is precisely balanced by the opposite effect on the other side of the Fermi surface. However, there is not exact electron-hole symmetry since the two branches of the excitation spectrum have opposite curvature. This lack of symmetry, coupled with an energy-dependent relaxation time,[15] will give rise to a branch imbalance.

If we assume that such a branch imbalance exists in our experiment, it is possible to extract information on the thermal transport coefficient for the quasiparticles from the data shown in Fig. 3. The quasiparticle current produced by the temperature gradients involved in this experiment should lead to a small asymmetry near the bottom of the electron and hole branches of the excitation spectrum. Another way in which a similar asymmetry could be produced is by injecting quasiparticles into a superconductor at very low voltages via a tunnel junction. Although to obtain detailed information will require a theory directly applicable to this experiment, the data shown on Fig. 3 can be interpreted using the theory of Tinkham[11] for tunneling generation of an electron-hole imbalance in the appropriate limit of small injection voltages. In this limit, the voltage is given by

$$V \leqslant \frac{I f(\Delta)}{2e^2 N(0)\Omega g_{\mathrm{NS}}^2}\tau_Q \quad [eV_{\mathrm{inj}} \ll \Delta(T)], \qquad (1)$$

where I is the injected quasiparticle current, $f(\Delta)$ is a Fermi factor, $N(0)$ is the one-spin density of states at the Fermi surface, Ω is the volume sampled by the voltage probes, g_{NS} the normalized low-voltage conductance of the NS junction (Fig. 2), and τ_Q is the branch-imbalance relaxation time.[12] The equality sign in Eq. (1) is valid near T_c. Since there exist both theory[15] and experiments[16] for τ_Q in Sn, and since all of the other quantities in Eq. (1) are known or can be measured, it is possible to use Eq. (1) to extract a value for the transport coefficient L_T by assuming $I = L_T(-\nabla T)$. The results of such an analysis for three different samples are shown in Fig. 4.

Because of the small temperature gradients

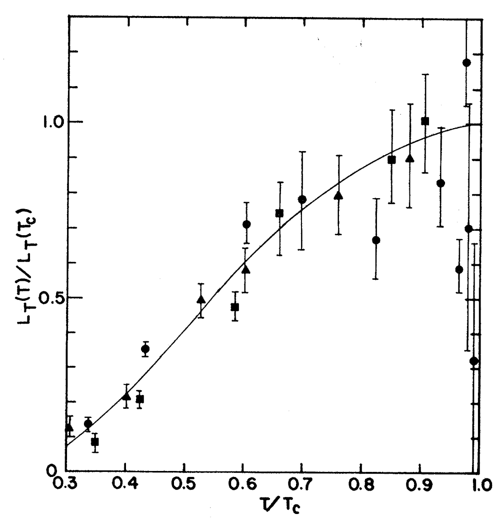

FIG. 4. Plot of the transport coefficient L_T normalized to its value at T_c vs temperature for three samples. The solid line is from a theory by Gal'perin, Gurevich, and Kozub (Ref. 18) in which $L_T(T_c)$ is taken as the value in the normal state just above T_c.

necessary and the concomitant small voltages developed, it is not possible at present to obtain information very near T_c; at lower temperatures we find the data to fall off smoothly, apparently to zero. In absolute magnitude, $|L_T| = 2-16$ A/cm K near T_c. Given that in the normal state $L_T = \sigma S$, where σ is the electrical conductivity and S is the thermoelectric power, this transport coefficient corresponds to normal-state thermoelectric powers between 3 and 24 μV/K. This is about what would be expected from measurements on the normal-state thermoelectric power in Sn just above T_c.[17]

A theory for L_T has been developed by Gal'perin, Gurevich, and Kozub.[18] This theory, which does not explicitly take into account the character of the electron and hole branches of the excitation spectrum, describes quasiparticle relaxation using the same mean free path appropriate to the normal state. The result of this theory with a one-parameter fit to the data at T_c is shown as a solid line in Fig. 4. As can be seen, the fit to the experiment data is good. However, without a reliable theory for interpreting the experimentally measured voltages in terms of a transport coefficient, one cannot comment further on this agreement.

In summary, we have observed a difference between the quasiparticle and pair electrochemical potentials in thin-film superconductors held in a temperature gradient. This potential diverges at low temperature and, within the resolution of our data, approaches a constant at T_c. It is possible to use our data to extract a value for the thermal transport coefficient L_T appropriate for the normal excitations in the superconductor. Unlike the experiments on bimetallic superconducting loops,[8-10] this analysis leads to values for L_T near T_c which are in rough agreement with those in the normal state. We also find L_T to fall off to zero at low temperatures. However, in the absence of a satisfactory theory to interpret these observations, this agreement with normal-state values must be regarded as very approximate. In particular, a satisfactory theory for thermoelectric effects in nonequilibrium superconductors must take into account the directional dependence of the distortion of the Fermi surface by a thermal gradient as well as the various relaxation mechanisms appropriate in the nonequilibrium state.

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