Further calculations allowing for other relaxation mechanisms are needed to understand this quantitatively.

In the absence of an external current applied to the superconductor, the experiment is similar to that performed by Falco[4] and we now calculate the voltages expected then. The total current is the sum of a normal component, $\vec{j}_n = -L\nabla T$, analogous to a thermoelectric current in a normal metal, and a supercurrent $e\rho_s\vec{v}_s$. L is the equivalent of a normal-state thermoelectric coefficient, and ρ_s is the superfluid density. In the absence of a net current,

$$\vec{j} = \vec{j}_n + \vec{j}_s = -L\nabla T + e\rho_s\vec{v}_s = 0 \tag{16}$$

or

$$\vec{v}_s = L\nabla T/\rho_s e, \tag{17}$$

and the charge-imbalance voltage near T_c is given by

$$eV/E_F = \tfrac{2}{3}\alpha(L\tau/\rho_s g_{NS})(\nabla T)^2/eT. \tag{18}$$

For Al the superfluid velocity becomes $\sim 10^{-5}$ cm/s, for $|\nabla T| = 0.1$ K/cm, if the mean free path for elastic scattering is comparable with the zero-temperature coherence length. In making this estimate we have used a typical value ($\sim 10^{-8}$ V/K) of the normal-state thermopower at the transition temperature. The charge-imbalance voltage is some seven orders of magnitude less than that which can be achieved by application of an external current.[9]

In summary, we have demonstrated that the combined effects of an externally applied supercurrent and a temperature gradient can lead to appreciable charge-imbalance voltages. The observed effect follows the prediction (15) well as regards the explicit dependence on $\vec{v}_s$ and ∇T. The measured time is considerably less than our estimates for a pure metal, thus emphasizing the need to consider additional mechanisms for charge relaxation.

We are grateful to our experimental colleagues, John Clarke, Birgit R. Fjordbøge, and Poul Erik Lindelof, for many stimulating discussions of the work presented here. We also thank Mr. J. Beyer Nielsen and Dr. Y. A. Ono for theoretical discussions. This work was supported in part by the National Science Foundation under Grant No. DMR78-21068.

[1]See, e. g., Yu. M. Galperin, V. L. Gurevich, and V. I. Kozub, Zh. Eksp. Teor. Fiz. 66, 1387 (1974) [Sov. Phys. JETP 39, 680 (1974)]; A. G. Aronov, Zh. Eksp. Teor. Fiz. 67, 178 (1974) [Sov. Phys. JETP 40, 90 (1975)].

[2]D. J. VanHarlingen and J. C. Garland, Solid State Commun. 25, 419 (1978).

[3]A. M. Guénault and K. A. Webster, J. Phys. (Paris), Suppl. 39, C6-539 (1978).

[4]C. M. Falco, Phys. Rev. Lett. 39, 660 (1977).

[5]J. Clarke, B. Fjordbøge, and P. E. Lindelof, following Letter [Phys. Rev. Lett. 43, 642 (1979)].

[6]C. J. Pethick and H. Smith, J. Phys. (Paris), Suppl. 39, C6-488 (1978).

[7]C. J. Pethick and H. Smith, Ann. Phys. (N.Y.) 119, 133 (1979).

[8]The role of impurity scattering for the present problem is discussed by P. E. Lindelof, B. R. Fjordbøge, and J. Bindslev Hansen, to be published; and by A. Schmid and G. Schön, to be published; and by J. Clarke and M. Tinkham, to be published.

Supercurrent-Induced Charge Imbalance Measured in a Superconductor in the Presence of a Thermal Gradient

John Clarke,[a] B. R. Fjordbøge, and P. E. Lindelof
Physics Laboratory I, H. C. Ørsted Institute, University of Copenhagen, DK-2100 Copenhagen Ø, Denmark
(Received 21 May 1979)

A pair-quasiparticle potential difference arising from a quasiparticle charge imbalance has been observed in superconducting tin films along which there exist both a supercurrent, I, and a temperature gradient, ∇T. The voltage is proportional to $I\nabla T$ at a given temperature, in agreement with the prediction of Pethick and Smith, and diverges as $(1-T/T_c)^{-1}$ for given values of I and ∇T.

We report the observation of a pair-quasiparticle potential difference,[1,2] arising from a quasiparticle charge imbalance, in a superconducting Sn film along which there exists both a supercurrent, I, and a temperature gradient, ∇T. Such an effect has been predicted by Pethick and

Smith.[3]

Our experimental configuration is shown in the inset of Fig. 3. First, a Sn film typically 300 nm thick and 0.1 mm wide in the middle region was evaporated onto a $32\times 7\times 1\text{-mm}^3$ soda glass or silicon substrate maintained at either liquid nitrogen or room temperature. The Sn was oxidized in air for 5 to 15 min, and three Cu(+3% Al) disks 0.8 to 1.3 μm thick and 2 mm in diameter were deposited. Finally three Pb strips 1 mm wide and about 200 nm thick were evaporated. The thickness and mean free path, l, of the Sn strips and the junction resistance at T_c, $R_{\text{jn}}(T_c)$, are listed in Table I for five samples. In a given experimental run one of the three Sn-SnO_x-Cu tunnel junctions was used to detect the quasiparticle potential in the superconducting Sn film relative to the pair potential.[1] The Pb strips eliminated nearly all the resistance of the Cu that would otherwise generate both an excessive Johnson noise and spurious thermoelectric effects. The Cu was sufficiently thick and dirty to eliminate pair tunneling between the two superconductors in the temperature range where we measured. Thin PbSn solder leads were attached to the films with In pellets, and connected to Nb wires to make superconducting current (I) and voltage (V) leads. The use of superconducting current leads enabled us to apply a current without heating the substrate [except above the In transition (~ 3.4 K) where a negligible heating occurred], while the use of superconducting voltage leads eliminated spurious thermoelectric voltages. The superconducting voltage lead was attached to a region of the Sn where $I=0$. If $I\neq 0$ and $\nabla T\neq 0$ at the point of attachment, this lead would still measure the pair potential at temperatures below the In transition, but not above it.

The sample was mounted in a vacuum can. Each end of the substrate was clamped to a Cu block, on which was wound a heater, connected to the top of the can via a suitable thermal conductance. Two Allen-Bradley carbon thermometers were attached to the rear side of the substrate with G. E. varnish. None of the leads connected to the substrate perturbed its temperature distribution significantly. Outside the can the voltage leads were connected in series with a resistor of $\sim 3\times 10^{-5}\ \Omega$ and the superconducting input coil of a S.H.E. SQUID operated as a null-balancing voltmeter. Thus, the quasiparticle potential was measured at (nearly) zero current[1] with a resolution limited by the Johnson noise in the resistor and the junction. The can was immersed in superfluid helium, and the cryostat was surrounded by a double Mumetal shield.

To make a measurement, we applied current to one or both heaters until the substrate attained the desired temperature gradient. The presence of a gradient always generated a voltage, presumably of the same origin as the voltages observed by Falco[4] in a similar configuration. This voltage, at most 1 pV, was small compared with the voltages generated by the applied supercurrent. When a steady gradient had been established, *we defined the voltage to be zero at $I=0$.* We increased the current I in steps, and measured the voltage V for each step. We took great care to ensure that the Sn was not driven normal. For example, after taking data at a given gradient, we could raise the temperature of the colder end of the sample until $\nabla T=0$, and check that $V=0$ at the highest current used. In Figs. 1 and 2 we plot V vs I for five values of ∇T, and V vs ∇T for ten values of I, for a representative sample. The quasiparticle potential is positive relative to the pair potential if the (conventional) current and ∇T are in the same direction. V is proportional to I over the accessible current range (up to three decades) and very nearly proportional to

TABLE I. Properties of five samples.

Sample No.	Sn thickness (nm)	l^{a} (nm)	$R_{\text{jn}}(T_c)$ (Ω)	$Vg_{\text{NS}}T(1-t)A/I\nabla T$ ($10^{-16}\ \Omega\ \text{cm}^3$)	Comments
1	400	57	1.1×10^{-4}	2.5	Glass; different junctions on same substrate
2	400	57	1.35×10^{-5}	3.8	
3	250	57	1.2×10^{-5}	0.8	Glass; ground plane added
4	320	428	2.0×10^{-5}	1.2	Glass
5	310	57	2.0×10^{-5}	2.1	Si

[a] Calculated from the resistance ratio without correction for size effects.

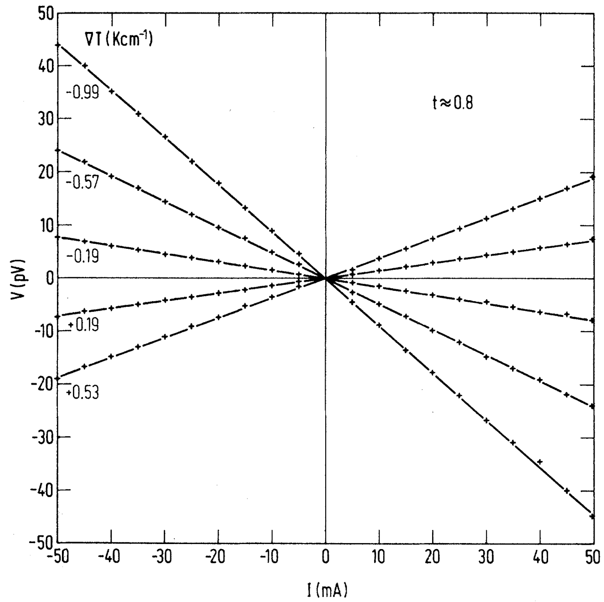

FIG. 1. V vs I for five values of ∇T for sample 4.

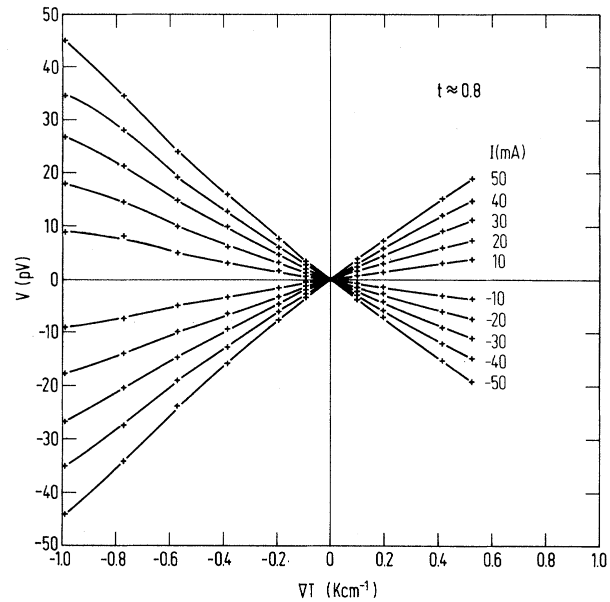

FIG. 2. V vs ∇T for ten values of I for sample 4. At each value of ∇T, the voltage is defined to be zero at $I=0$.

∇T. The small deviations from linearity in Fig. 2 are caused by errors in estimating the junction temperature from the two thermometer readings, and the fact that we did not correct the gradients estimated from the two thermometers for the temperature-dependent thermal conductance of the substrate.

The measured voltage is inversely proportional to the measured normalized junction conductance, g_{NS},[1,2] which we determined separately by applying a current to the lead i and one of the leads I. To eliminate the temperature dependence of g_{NS}, which was somewhat sample dependent, we have plotted $Vg_{NS}/I\nabla T$ versus reduced temperature, t, in Fig. 3. $Vg_{NS}/I\nabla T$ diverges as $t\to 1$, and falls off steadily with decreasing temperature at low temperatures.

At this point we discuss briefly several possible experimental problems: (1) A simple calculation indicates that the thin films should not significantly perturb the temperature distribution of a glass substrate, and that the gradient in the Sn film should be the same as that in the substrate, even in the vicinity of the overlying films. As a check, we prepared a sample (No. 5) on a Si substrate with a thermal conductance three orders of magnitude greater than glass. The signal generated was not significantly different (Table I). (2) The temperature gradient along the copper film together with the magnetic field in its plane generated by I give rise to transverse thermoelectric effects, but voltages generated this way are estimated to be at least two orders of magnitude below the observed values. Besides, we would not expect such effects to have the temperature dependence shown in Fig. 3.[3] The supercurrent tends to concentrate at the edges of the Sn film except under the Pb films, which act as ground planes. To investigate possible effects due to current redistribution near the edges of the Pb film, after studying sample No. 3 we coated the films with a thin ($\lesssim 1\ \mu$m) layer of Duco cement, and deposited a large Pb ground plane. The measured voltages without and with the ground plane agreed to within the scatter in the data.[4] Over most of the temperature range the penetration depth is less than the film thickness and the supercurrent is excluded from the interior of the film. However, the charge imbalance should be uniform across the thickness of the film which is much less than the charge-imbalance diffusion length.

Finally, we compare our results with the theory of Pethick and Smith,[3] who predict that for a superconductor near T_c the quasiparticle potential is given by [Eq. (15) of Ref. 3]

$$eV=\frac{\pi\Delta(T)}{4k_BT}\,\frac{\tau E_F\vec{j}\cdot\nabla T}{e\rho_s(T)g_{NS}T}\,. \tag{1}$$

Here, ρ_s is the superfluid density, which is pro-

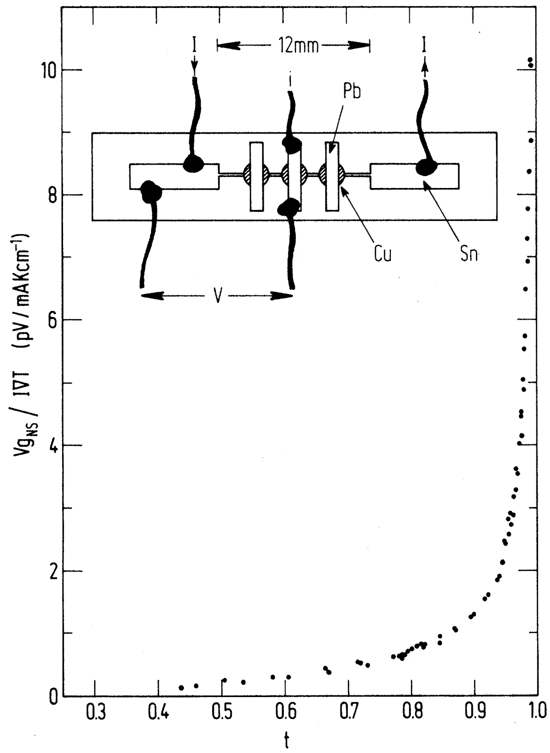

FIG. 3. $Vg_{NS}/I\nabla T$ vs reduced temperature, t, for sample 4. Inset shows sample configuration.

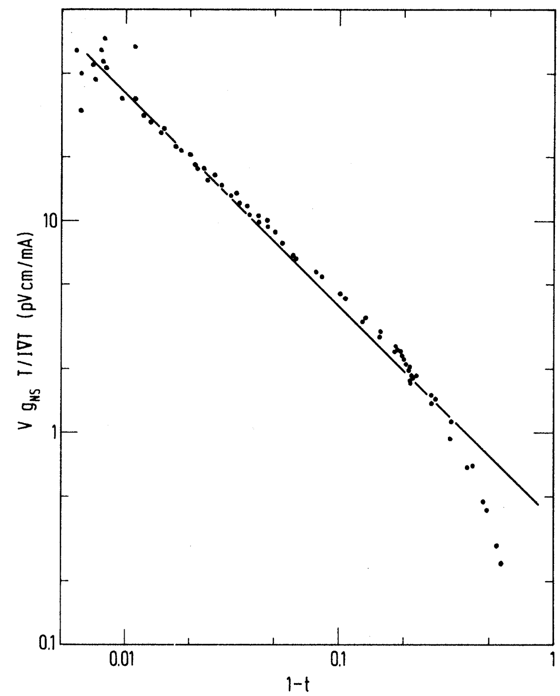

FIG. 4. $Vg_{NS}T/I\nabla T$ vs $(1-t)$ for sample 4. Line is drawn with slope -1.

portional to $1-t$, E_F is the Fermi energy, $\Delta(T)$ is the energy gap, and τ is a characteristic time for quasiparticle-charge relaxation. The sign of V and its dependence on $\vec{j}\cdot\nabla T$ are consistent with our experimental results (see Fig. 4). In the limit where the inelastic-scattering rate is much greater than the elastic-scattering rate, which is definitely not the case for our samples, Pethick and Smith set $\tau = 4k_BT\tau_{inel}(0)/\pi\Delta(T)$, where $\tau_{inel}(0)$ is the electron-phonon scattering time at T_c and at the Fermi energy. With this value of τ, Eq. (1) yields the observed temperature dependence near T_c, but a value of $Vg_{NS}T(1-t)/j\nabla T$, $5\times10^{14}\ \Omega\ \mathrm{cm}^3$, that is two to three orders of magnitude greater than the values listed in Table I (A is the cross section of the Sn films). Thus, it appears that additional scattering mechanisms that will produce a smaller characteristic time τ must be taken into account. It should be borne in mind that, at least in the context of the Pethick-Smith[3] theory, the time inserted in Eq. (1) must be proportional to Δ^{-1}.

In summary, the sign of the observed quasiparticle potential and its dependence on I and ∇T are correctly predicted by the theory of Pethick and Smith.[3] However, the theory makes a definite prediction for the temperature dependence and magnitude of the effect only in the limit where inelastic scattering dominates elastic scattering, and further theoretical work is required for the experimentally accessible limit in which elastic scattering dominates.[5]

We are indebted to Ole Eg for sample preparation. We are grateful to Chris Pethick and Henrik Smith for many stimulating conversations. This work was supported by the Philips Foundation of 1958 and the Danish Natural Science Research Council. One of us (J.C.) acknowledges receipt of a Guggenheim Fellowship.

[a]On sabbatical leave from the Department of Physics, University of California, and Materials and Molecular Research Division of the Lawrence Berkeley Laboratory, Berkeley, Cal. 94720.

[1]J. Clarke, Phys. Rev. Lett. 28, 1363 (1972).

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

[3]C. J. Pethick and H. Smith, preceding Letter [Phys. Rev. Lett. 43, 640 (1979).

[4]C. M. Falco, Phys. Rev. Lett. 39, 660 (1977).

[5]P. E. Lindelof, B. R. Fjordbøge, and J. Bindslev Hansen, to be published; A. Schmid and G. Schön, to be published; J. Clarke and M. Tinkham, to be published.