Observation of Thermoelectrically Induced Charge Imbalance in Superconducting Aluminum

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The quasiparticle transport current induced in a superconducting aluminum film by a temperature gradient has been detected by measurements of the spatially decaying charge imbalance generated near the end of the sample where the current is divergent. The results are in good agreement with theory when the nonuniformity of the temperature gradient is taken into account. In particular, the inferred value of the thermopower agrees well with the value obtained when the aluminum is in the normal state.

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Ginzburg[1] pointed out in 1944 that a temperature gradient, $\vec{\nabla} T$, in a superconductor should produce a quasiparticle current, $\vec{j}_N = L_T(-\vec{\nabla} T)$, that is everywhere cancelled by a counterflowing supercurrent, $\vec{j}_S$; here, $L_T(T)$ is the thermoelectric transport coefficient. There have been several experiments to investigate this effect, but the results have often been in conflict with each other and in disagreement with theoretical predictions. For example, attempts to observe an asymmetry in the critical current of superconducting weak links in the presence of a temperature gradient have produced conflicting results. Experiments to measure the magnetic flux in a superconducting bimetallic ring generally produced results that were not only several orders of magnitude larger than the theoretical prediction, but that also exhibited a different temperature dependence. Details of these and related experiments and references thereto can be found in the recent reviews by Schön[2] and Van Harlingen.[3]

Near the boundary of a superconductor across which there is heat flow, Ginzburg's original prediction is modified by the presence of charge imbalance[4] arising from the interconversion of normal current and supercurrent.[5] Recent measurements[6,7] of the excess thermoelectric voltage produced by the charge-imbalance region in superconductor–normal-metal–superconductor sandwiches have been in good agreement with theory. In this Letter, we report an experiment in which we measured directly the spatial variation of the charge imbalance produced by the divergence in the thermoelectrically induced quasiparticle current near the end of a superconducting film.

It can be shown[2,3,5,6] that $\vec{j}_N$ and the charge imbalance[4] per unit volume, Q^*, in the superconductor are related by the equations

$$\vec{\nabla} \cdot \vec{j}_N = -Q^*/\tau_{Q^*}, \tag{1}$$

and

$$\vec{j}_N = [\sigma/2N(0)e^2](-\vec{\nabla} Q^*) + L_T(-\vec{\nabla} T). \tag{2}$$

Here, $\tau_{Q^*}^{-1}$ is the charge-imbalance relaxation rate, $\sigma(T)$ is the electrical conductivity, and $N(0)$ is the single-spin electron density of states. We now apply these equations to a one-dimensional superconductor ($0 \leq x < \infty$) in the presence of a temperature gradient dT/dx. The constraint $j_S(x) + j_N(x) = 0$ holds throughout the superconductor, but, in addition, the requirement that each current must be continuous across the boundary imposes the condition $j_S(0) = j_N(0) = 0$. As a consequence, there must be divergence in j_N near the end of the superconductor, and a corresponding production of charge imbalance. For a uniform temperature gradient ($d^2T/dx^2 = 0$), one can solve Eqs. (1) and (2) subject to the boundary condition to find

$$Q^*(x) = 2N(0)e^2 \lambda_{Q^*} S^S (dT/dx) \exp(-x/\lambda_{Q^*}), \tag{3}$$

where $\lambda_{Q^*} \equiv [\sigma \tau_{Q^*}/2N(0)e^2]^{1/2}$ is the charge relaxation length, and $S^S \equiv L_T/\sigma$ is the thermopower in the superconducting state. One can detect the charge imbalance by means of a normal electrode making a tunneling contact to the superconductor. For zero tunneling current, the voltage across the junction is[4] $V(x) = Q^*(x)/2N(0)e^2 g_{NS}(0)$, where $g_{NS}(0)$ is the zero-bias tunneling conductance normalized to its value at the transition temperature of the superconductor. In the present experiment, we measure $V(x)$ with a series of probes near the end of a superconducting film in the presence of a known temperature gradient, and hence deduce both S^S and the decay length of Q^*.

The sample configuration is shown in Fig. 1. An Al film, typically 300 nm thick, was evaporated onto a 1-mm-thick glass substrate, and a strip 500 μm wide was prepared by etching. A series of probes was fabricated on top of the Al film with a photolithographic procedure. Prior to the depostion of the probes, the Al surface was ion milled and thermally oxidized. The probes consisted of 800 nm of Cu (3 wt.% Al) followed by about 5 nm of Fe and 200 nm of Pb (5 wt.% In). The PbIn eliminated the series resistance of the probes that would have degraded the voltage sensitivity, while the Fe layer prevented any Josephson tunneling between the PbIn and the Al. Near the end of the film the probes were 2 μm wide with a 6 μm separation between centers; the sensitivity of the measurement was limited to about 0.2 pV $Hz^{-1/2}$ by the Johnson noise in the tunneling resistance, typically 0.5 mΩ. At greater distances from the end, the probes were made wider to produce a lower tunneling resistance and hence an enhanced sensitivity. The voltage developed across each junction was measured with a null-balancing dc SQUID voltmeter[8] relative to a wide probe far from the end of the film. The voltmeter was switched to each probe in turn by means of a mechanical superconducting switch operated from outside the cryostat. In another region of the Al film, a sample was etched to enable us to measure the normal state thermopower, again with the aid of the null-balancing voltmeter. One end of the substrate was clamped to a copper block in a vacuum can, while a noninductive heater was glued to the other end to enable us to establish a temperature gradient. Three carbon-resistance thermometers were glued to the reverse side of the substrate; two determined the temperature gradient and the third measured the temperature in the vicinity of the probes. Since the temperature drop along the sample in the region of interest was typically 2 mK or less, we define the average temperature to be the sample temperature T_s. The vacuum can was immersed in liquid helium, the temperature of which was lowered to about 1.0 K.

Of the many samples studied, we have investigated three in detail. The transition temperature, T_c, of sample 1 was 1.231 K, and the normal state thermopower, S^N, just above T_c was $(-1.05 \pm 0.06)\times 10^{-8}$ V K^{-1}, a value in good agreement with values in the literature.[9] Figure 2 shows the measured value of $V(x)$ vs x at $T_s = 0.978T_c$ for $dT/dx = 28.1$ K m^{-1}. The voltage drops from 5.1 pV at the probe 6 μm from the end of the sample to 0.52 pV at the probe 68 μm from the end. The signal was independent of the polarity of the heater current. We observed zero voltage at probes far from the end of the sample, and at all probes when the temperature of the superconducting strip was raised uniformly, thereby eliminating the possibility that the voltage arose from a thermoelectric effect in the junctions or in the measuring circuit. If one attempts to fit a straight line to the data, the inferred decay length is 29 ± 4 μm. This length was

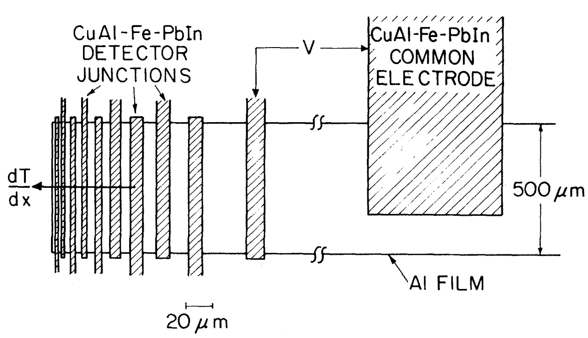

FIG. 1. Sample configuration. Note the two different length scales.

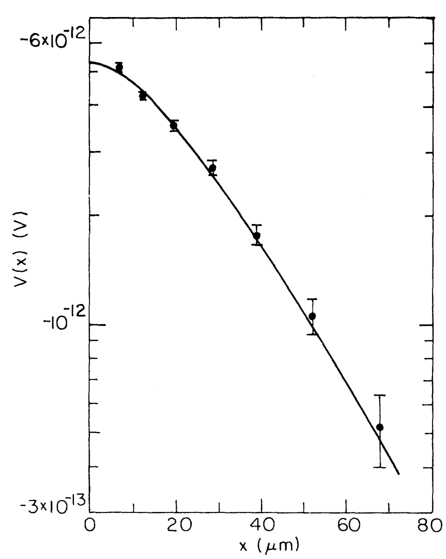

FIG. 2. Measured values of $V(x)$ for sample 1 ($T_s = 0.978T_c$, $dT/dx = 28.1$ K m^{-1}) in presence of temperature gradient. Solid line is from Eq. (4) with $\lambda_{Q^*} = 14.5$ μm, $\lambda_T = 19$ μm, and $S^S = -3.0\times 10^{-8}$ V K^{-1}.

nearly independent of temperature. We measured λ_{Q^*} independently by passing a current through one of the probe junctions, and measuring the decay length of the induced charge imbalance from the voltages produced at nearby probes. The observed voltages decayed exponentially with distance from the junction through which current was injected, and yielded $\lambda_{Q^*} = 14.5 \pm 1.0$ μm at $T_s = 0.978T_c$.[10] The length λ_{Q^*} varied rather accurately as $(1 - T_s/T_c)^{1/4}$ as expected.[4] In view of the different magnitudes and temperature dependences of λ_{Q^*} and the length measured in the thermoelectric experiment, we are forced to conclude that the latter length is not λ_{Q^*}. We must therefore reexamine the assumptions involved in the derivation of Eq. (3).

In our theoretical discussion, we assumed that $d^2T/dx^2 = 0$; if this term is in fact nonzero, there will be an additional thermoelectric contribution to the charge imbalance.[11,12] Since the heat current must enter the film from the substrate via a Kapitza boundary resistance, d^2T/dx^2 will be nonzero near the end of the sample over a distance λ_T that depends on the Kapitza resistance and the thermal conductivities of the substrate and film. Since the thermal conductance of the Al film is roughly 20 times less than that of an equal width of the glass substrate, to a first approximation we can assume that the presence of the film does not perturb the temperature dependence of the substrate significantly. In this approximation, one finds $\lambda_T = (gd/y_K)^{1/2}$, where g and d are the thermal conductivity and thickness of the film, and y_K is the Kapitza conductance of a unit area between the film and the substrate. From the measured electrical resistivity (5.4 nΩ m) and the Wiedemann-Franz law we estimate $g \approx 6$ W m^{-1} K^{-1}, while we assume[13] $y_K \approx 5\times10^3$ W m^{-2} K^{-1}. Setting $d \approx 300$ nm, we estimate $\lambda_T \approx 20$ μm. In a quasi-one-dimensional model dT/dx is zero at $x = 0$, and it is easy to show that $dT(x)/dx = (dT/dx)|_\infty[1 - \exp(-x/\lambda_T)]$. Substituting this expression in Eqs. (1) and (2), we find

$$Q^*(x) = \frac{2N(0)e^2\lambda_{Q^*}{}^2}{\lambda_{Q^*}{}^2 - \lambda_T{}^2} \times (\lambda_{Q^*}e^{-x/\lambda_{Q^*}} - \lambda_T e^{-x/\lambda_T}) S^S \left.\frac{dT}{dx}\right|_\infty . \quad (4)$$

We have fitted Eq. (4) to the experimental data in Fig. 2 using the measured value of λ_{Q^*}. The fit yields $\lambda_T = 19 \pm 4$ μm, in good agreement with our estimate. The values obtained for the thermopower, $S^S(T_s/T_c)$, at the highest temperature

TABLE I. Values of thermopowers in the superconducting (S^S) and normal (S^N) states for three samples.

Sample	$S^N(T_c)$ (10^{-8} V K^{-1})	T_s/T_c	$S^S(T_s/T_c)$ (10^{-8} V K^{-1})
1	-1.05 ± 0.06	0.991	-2.7 ± 0.6
2	-0.94 ± 0.06	0.992	-1.4 ± 0.3
3	-1.12 ± 0.06	0.997	-1.0 ± 0.3

T_s/T_c at which we obtained data are listed in Table I for all three samples. The corresponding values of S^N, obtained just above T_c, are also listed. The value of S^S for sample 1 exceeds S^N by about a factor of 2, while the values of S^S and S^N for samples 2 and 2 are in quite good agreement. Given the limitations of our model, we feel that the fit of the theory to the data and the agreement between the values of S^S and S^N are very satisfactory.

In summary, we have measured the spatial variation of the charge imbalance induced near the end of a superconducting aluminum film by a temperature gradient. A simple model that takes into account the fact that d^2T/dx^2 is also nonzero agrees well with the data, and yields values of the thermopower in the superconducting state that are in reasonable agreement with the measured values in the normal state. A more detailed account of this work will be submitted for publication elsewhere.

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