Generation of Charge Imbalance in a Superconductor by a Temperature Gradient

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We predict that a superconductor carrying a supercurrent in a temperature gradient ∇T will develop a charge-imbalance voltage proportional to $\vec{v}_s \cdot \nabla T$, where $\vec{v}_s$ is the superfluid velocity. The magnitude of the voltage is estimated.

Attempts to measure thermoelectric effects in superconductors have led to results which are difficult to understand in terms of current physical ideas. On the theoretical side, most attention has been devoted to the normal currents generated by a temperature gradient,[1] but the results of experiments designed to measure these effects are too large to be accounted for by theory,[2] and also depend on surface properties of the sample.[3] Falco[4] has attempted to measure the chemical-potential difference between the normal and superfluid components generated by a temperature gradient, but it is not yet clear how to interpret his results. One of the difficulties in both of the above experiments is that the effects to be measured are very small. In this Letter we show that much larger chemical-potential differences between the normal and superfluid components can be generated by passing a current through a superconductor in a temperature gradient. In the following Letter, Clarke, Fjordbøge, and Lindelof[5] describe the first measurements of this effect.

Let us calculate the charge imbalance generated in a current-carrying superconductor in a temperature gradient. We consider the case of a clean superconductor, and the excitations may therefore be characterized by their momentum $\vec{p}$. As we have described in detail elsewhere,[6,7] the voltage developed between a superconductor and a normal probe is proportional to the deviation, $\delta Q_n^{\text{l.e.}}$, of the quasiparticle charge from its local equilibrium value. This is given by

$$\delta Q_n^{\text{l.e.}} = \sum_{\vec{p}\sigma} q_{\vec{p}}\, \delta f_{\vec{p}}^{\text{l.e.}}, \tag{1}$$

where $\delta f_p^{\text{l.e.}}$ is the deviation of the quasiparticle distribution function from its local equilibrium value, and $q_p = u_p^2 - v_p^2 = \xi_p / E_p$ is the effective charge of a quasiparticle. Here u_p and v_p are the usual coherence factors, ξ_p is the energy of a normal-state quasiparticle measured from the chemical potential, and the quasiparticle energy E_p is given by

$$E_p = (\xi^2 + \Delta^2)^{1/2}, \tag{2}$$

where Δ is the gap. We shall work with positive-energy quasiparticles throughout.

The quasiparticle distribution function $f_{\vec{p}}$ is found by solving the Boltzmann equation, which has the usual form

$$\frac{\partial E_{\vec{p}}'}{\partial \vec{p}} \cdot \frac{\partial f_{\vec{p}}}{\partial \vec{r}} - \frac{\partial E_{\vec{p}}'}{\partial \vec{r}} \cdot \frac{\partial f_{\vec{p}}}{\partial \vec{p}} = \left(\frac{d f_{\vec{p}}}{dt}\right)_{\text{coll}}. \tag{3}$$

Here $(df_{\vec{p}}/dt)_{\text{coll}}$ denotes the collision term. In the streaming term the quasiparticle energy $E_{\vec{p}}'$ is that in the laboratory frame, in which the superfluid moves with velocity $\vec{v}_s$, and is therefore given by

$$E_{\vec{p}}' = E_p + \vec{p} \cdot \vec{v}_s. \tag{4}$$

The quasiparticle velocity is

$$\partial E_{\vec{p}}' / \partial \vec{p} = \vec{v}_p + \vec{v}_s, \tag{5}$$

where $\vec{v}_{\vec{p}} = (\xi_p / E_p)\vec{p}/m^*$ and m^* is the effective mass. We are interested in situations where spatial variations are slow, and therefore in the streaming term in (3) we may replace $f_{\vec{p}}$ by a local equilibrium function $f_{\vec{p}}^{\text{l.e.}} = \{\exp[E_{\vec{p}}'/k_B T(\vec{r})] + 1\}^{-1}$. To first order in $\vec{v}_s$ and first order in ∇T the left-hand side of the Boltzmann equation (3) is

$$-(\vec{p} \cdot \vec{v}_s)\left(\vec{v}_p \cdot \frac{\nabla T}{T}\right) \frac{\partial}{\partial E_{\vec{p}}}\left(E_p \frac{\partial f_p^0}{\partial E_p}\right) - \vec{v}_s \cdot \frac{\nabla T}{T} E_{\vec{p}} \frac{\partial f_{\vec{p}}^0}{\partial E_{\vec{p}}}, \tag{6}$$

where f^0 denotes the equilibrium Fermi function $f(E)$. The first term in (6) contains a term which is independent of the direction in momentum space, and which is odd in ξ, and this can lead to a charge imbalance. The part of the first term which contains quadrupolar terms, and the second term, which is even in ξ, will not contribute to the charge imbalance. The latter term will, however, give rise to a change in the gap. The rate at which quasiparticle charge is generated is found by multiplying the driving term (6) by $\xi_{\vec{p}}/E_{\vec{p}}$ and summing over momentum space. We find

$$(dQ_n/dt)_{\text{gen}} = -n\alpha\vec{v}_s\cdot(\nabla T)/T, \tag{7}$$

where the dimensionless quantity α is given by

$$\alpha = 2\int_0^\infty d\xi\, \frac{\xi^2}{E^2}\,\frac{\partial}{\partial E}\left(E\,\frac{\partial f^0}{\partial E}\right), \tag{8}$$

and n is the electron density. For $T \to T_c$, α tends to $\frac{1}{4}\pi\Delta/k_B T_c$. This shows that, as one might expect, for a normal metal no charge imbalance is generated, since all the charge is then in the normal component.

Collisions of quasiparticles convert quasiparticle charge into charge associated with the pairs. We describe the relaxation phenomenologically by the equation

$$(dQ_n/dt)_{\text{coll}} = -\delta Q_n^{\text{l.e.}}/\tau, \tag{9}$$

where τ is a characteristic time for charge relaxation. By equating (7) and (9) one finds

$$\delta Q_n^{\text{l.e.}} = n\alpha[(\vec{v}_s\cdot\nabla T)/T]\,\tau. \tag{10}$$

The voltage difference measured between the superfluid and a normal probe is given by[7]

$$eV = \delta Q_n^{\text{l.e.}}/2N(0)g_{\text{NS}}, \tag{11}$$

where $N(0)$ is the density of states at the Fermi surface, for one-spin population, and g_{NS} is the tunnel conductance divided by its normal-state value. The electronic charge is $e = -|e|$. Thus we find

$$eV/E_F = \tfrac{2}{3}(\alpha\tau/g_{\text{NS}})(\vec{v}_s\cdot\nabla T)/T, \tag{12}$$

where $E_F = p_F^2/2m^*$ denotes the Fermi energy.

The characteristic time τ depends on the particular relaxation mechanism under consideration. Let us now first consider the effect of electron-phonon scattering. For many charge-imbalance situations one can show that for $T \to T_c$, $\tau = (4/\pi)(k_B T_c/\Delta)\tau_{\text{inel}}(0)$, where $\tau_{\text{inel}}(0)$ is the normal-state electron-phonon scattering time at the Fermi energy. One method of doing this is by expanding the solution of the Boltzmann equation for the superconductor in terms of the eigenfunctions of the collision operator for the normal state.[7] The present situation is somewhat different, but one may expect to obtain a reasonable quantitative estimate of the effect near T_c using this value for τ. For $T \to T_c$, $\alpha \simeq \frac{1}{4}\pi\Delta/k_B T$, and $g_{\text{NS}} \simeq 1$, and therefore, taking $\tau \sim (4/\pi)(k_B T/\Delta) \times\tau_{\text{inel}}(0)$, we then find

$$eV/E_F = \tfrac{2}{3}\tau_{\text{inel}}(0)(\vec{v}_s\cdot\nabla T)/T. \tag{13}$$

To estimate V for a pure superconductor, we observe that the maximum velocity one can use is of order the critical velocity $\sim\Delta/p_F$, where p_F is the Fermi momentum. The maximum voltages that can be developed are therefore of order

$$|eV|/\Delta \sim v_F\tau_{\text{inel}}(0)|(\nabla T)/T|. \tag{14}$$

For Al, $\tau_{\text{inel}}(0)$ is $\sim 10^{-8}$ s and $v_F \sim 10^8$ cm s^{-1}. Thus since Δ typically corresponds to a temperature of order 1 K, for a temperature gradient of order 0.1 K/cm, one expects a maximum voltage of $\sim 10^{-6}$ V. The electron-phonon scattering rates for other metals are generally much larger than for Al, and consequently the voltages developed will be correspondingly smaller.

Since impurity scattering dominates electron-phonon scattering in the metallic films used for charge-relaxation experiments, we shall briefly discuss the rate of charge relaxation due to the scattering by impurities, whose importance has been stressed by a number of other workers.[8] In the presence of superflow such processes contribute to the relaxation of charge, essentially because of the anisotropy of the quasiparticle energy (4). Since the scattering takes place on a surface of constant energy, but with varying effective charge, such processes contribute to the overall rate of charge relaxation.

To facilitate comparison with experiment we now write the expected voltage in terms of the supercurrent $\vec{j}_s = e\rho_s\vec{v}_s$, with ρ_s being the superfluid density:

$$eV/E_F = \tau\,\alpha(\vec{j}_s/e)\cdot\nabla T/\rho_s g_{\text{NS}}T. \tag{15}$$

A companion Letter gives details of the experimental arrangement for measuring the charge-imbalance voltage produced by a temperature gradient and a current flowing in the sample.[5] The measured voltages confirm the validity of (15), both with regard to the functional dependence on $\vec{v}_s$ and ∇T as well as the sign of the voltage. The characteristic relaxation time τ deduced from the measurements is considerably less than that we estimate for phonon scattering alone.

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.

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

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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

