Theory of Quasiparticle Charge Imbalance Induced in a Superconductor by a Supercurrent in the Presence of a Thermal Gradient

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(Received 30 July 1979)

The charge-imbalance voltage induced in a clean superconductor by a supercurrent density $\vec{J}_s$ in the presence of a temperature gradient ∇T is shown to be $V = 2\pi v_F l \lambda^2 \Delta (\vec{J}_s \cdot \nabla T)/3c^2 k_B T^2 \cosh^2(\Delta/2k_B T) g_{NS}(1-Z)$, where v_F, l, and $\lambda(T)$ are the Fermi velocity, mean free path, and penetration depth, g_{NS} is the normalized tunnel conductance of the probe junction, and $Z(\Delta/T)$ is a known function. This result agrees well with the experiments of Clarke, Fjordbøge, and Lindelof.

Pethick and Smith[1] have predicted that a quasiparticle charge imbalance Q^* and potential V will be established when the superfluid moves at a velocity $\vec{v}_s$ in a superconductor along which there exists a temperature gradient ∇T. In the clean limit and for $\Delta \ll k_B T$ (Δ is the energy gap) they find

$$\frac{eV}{E_F} = \frac{\pi}{6} \frac{\Delta}{k_B T} \frac{\tau}{g_{NS}} \vec{v}_S \cdot \frac{\nabla T}{T}, \tag{1}$$

where τ is "a characteristic time for charge relaxation", E_F is the Fermi energy, and g_{NS} is the normalized tunneling conductance to the normal-metal probe used to detect V.[2,3] Near T_c, v_s is proprotional to $j_s(1-t)^{-1}$, where j_s is the supercurrent density, and $t = T/T_c$. Clarke, Fjordbøge, and Lindelof[4] observed this potential in Sn films, and found that V was proportional to $j_s \nabla T$ at a given temperature, and that $V g_{NS} T/ j_s \nabla T$ diverged as $(1-t)^{-1}$. Thus to bring the temperature dependence predicted by (1) into agreement with the experimental observations, it is necessary for τ to be proportional to Δ^{-1}. For a superconducting film in which the inelastic mean free path is much less than the elastic mean free path (not a physically realizable situation), Pethick and Smith interpret τ as the inelastic charge relaxation time, $\tau_{Q^*} = (4k_B T/\pi\Delta)\tau_E$ (τ_E is the inelastic scattering time at E_F at T_c), obtaining the correct temperature dependence but a voltage that exceeds the experimental values by two to three orders of magnitude. If one replaces τ by the temperature-independent impurity-scattering time, one obtains approximately the right magnitude for the voltage at temperatures near $0.9T_c$, but an incorrect temperature dependence near T_c, namely $(1-t)^{-1/2}$. In this Letter we give an alternative theory for this effect, applicable to clean films where the quasiparticle momentum $\vec{p}_k$ is a good quantum number. Our result fits the observed temperature dependence over the entire experimental range ($t = 0.5-0.99$) and it is in reasonable agreement with the experimentally determined magnitude for clean films.

We consider first the situation in which $\vec{v}_s = 0$, so that the quasiparticle excitation spectrum is

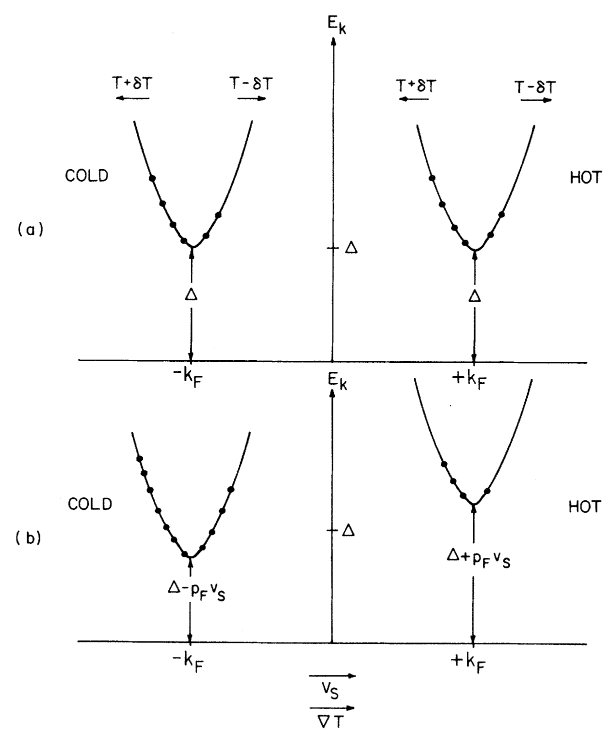

FIG. 1. (a) Schematic representation of quasiparticle excitations in presence of thermal gradient. (b) Same as (a), but with $\vec{v}_s$ also present.

symmetric around the Fermi surface, but $\nabla T \neq 0$.[5] Quasiparticles moving from the left in Fig. 1(a) are at an effective temperature $T-\delta T$, while those from the right are at $T+\delta T$, where T is the local temperature. Thus, there is an imbalance in the populations of the $k>k_F$ and $k<k_F$ branches on the right-hand side of the Fermi surface, but an equal and opposite imbalance on the left-hand side; as a result, the value of Q^* is zero. If we impose a superfluid velocity $\vec{v}_s$, the excitation energies take the form[6,7]

$$E_k = (\epsilon_k{}^2 + \Delta^2)^{1/2} + \vec{p}_k \cdot \vec{v}_s, \tag{2}$$

where ϵ_k is the one-electron energy relative to the chemical potential, and $\vec{p}_k$ is the electron momentum. There is now an asymmetry in the excitation spectrum about the Fermi surface, as indicated in Fig. 1(b), so that the population imbalances on the two sides no longer cancel exactly. This net charge imbalance is the origin of the observed potential, and is the quantity that we now calculate.

Consider a superconductor with transverse dimensions small compared with the penetration depth, so that any supercurrent flows uniformly. We assume that $p_F v_s \ll \Delta$ and $l\,|\nabla T/T| \ll 1$, where p_F is the Fermi momentum and l is the electronic mean free path. Our derivation is based on the general Boltzmann-equation solution quoted by Ziman[8] for the case of a uniform temperature gradient, in which the population of the state $\vec{k}$ at point $\vec{r}$ is the equilibrium population for the temperature at the point one mean free path back along the trajectory. This holds for isotropic scattering, whether elastic or inelastic, since that scattering erases all memory of the direction of the previous random-walk trajectory. Thus,

$$\delta f_{\vec{k}} = f_0\left(\frac{E_{\vec{k}}{}^0 + \vec{v}_s \cdot \vec{p}_{\vec{k}}}{T - \vec{l}_{\vec{k}} \cdot \nabla T}\right) - f_0\,\frac{E_{\vec{k}}{}^0}{T} \quad . \tag{3}$$

Here f_0 is the Fermi function, and $\delta f_{\vec{k}}$ is the departure from the occupation number when both $\vec{v}_s$ and ∇T are zero. As argued above, one gets a net Q^* only if both $\vec{v}_s$ and ∇T are nonzero. The relevant noncanceling term is

$$\delta f_{\vec{k}} = -\frac{\partial^2 f_0}{\partial E\,\partial T}\,(\vec{v}_s \cdot \vec{p}_{\vec{k}})(\vec{l}_{\vec{k}} \cdot \nabla T). \tag{4}$$

Note that $\vec{l}_{\vec{k}} \equiv l\hat{v}_{\vec{k}} = l\,\mathrm{sgn}(\epsilon)\vec{p}_{\vec{k}}/p_F$, since the group velocity along $\vec{p}_{\vec{k}}$ is $dE/d\epsilon = (\epsilon/E)v_F$, and the factor ϵ/E is canceled by a factor $|E/\epsilon|$ in the scattering time.[9] Although the coherence factors involved in this cancellation are modified by v_s, the mean free path remains isotropic at least to lowest order[10] in v_s as is implicitly assumed here. Then an elementary geometrical argument shows that, averaged over a sphere, the factors in parentheses in (4) reduce to $\frac{1}{3}p_F\,l\,\mathrm{sgn}(\epsilon)\vec{v}_s \cdot \nabla T$. Thus, the normal charge[11] δQ_n is given by

$$\begin{aligned}\delta Q_n &= 2N(0)\int_{-\infty}^{\infty} \frac{\epsilon}{E}\langle \delta f\rangle\, d\epsilon \\ &= \tfrac{4}{3}N(0)p_F\,l\,\vec{v}_s \cdot \nabla T \int_{\Delta}^{\infty}\left(-\frac{\partial^2 f_0}{\partial E\,\partial T}\right)dE \\ &= \tfrac{4}{3}N(0)p_F\,l\,\vec{v}_s \cdot \nabla T\,\frac{\partial f_0(\Delta)}{\partial T},\end{aligned} \tag{5}$$

where $\langle \delta f\rangle$ refers to the angular average of $\delta f_{\vec{k}}$ at fixed ϵ, and we have used the fact that $\epsilon\, d\epsilon = E\,dE$.

Now, this δQ_n is *not* the same as Q^*, as was first pointed out by Pethick and Smith[11] and further elaborated by Kadin, Smith, and Skocpol.[12] The argument can be summarized as follows: Maintenance of electrical neutrality requires that the total electronic charge $\sum v_k{}^2 + \sum(1-2v_k{}^2)f_k$ remain equal to the total charge of the positive ion cores. Thus, first-order changes in v_k and f_k about their equilibrium values $v_k{}^0$ and $f_k{}^0$ are constrained by $\sum(1-2f_k{}^0)\,\delta v_k{}^2 + \sum(1-2v_k{}^{02})\,\delta f_k = 0$. But all the $\delta v_k{}^2$ are determined from the shift δE_F in Fermi energy of the condensate by the relation $\delta v_k{}^2 = (\Delta^2/2E_k{}^3)\,\delta E_F$. The quasiparticle charge Q^* is also directly proportional to this shift, since it is equal and opposite to the change in condensate charge. Thus we have

$$\begin{aligned} Q^* &= -2N(0)\,\delta E_F = 2N(0)\frac{\sum(1-2v_k{}^{02})\,\delta f_k}{\sum(1-2f_k{}^0)(\Delta^2/2E_k{}^3)} \\ &= \frac{\sum q_k{}^0\,\delta f_k}{1-Z} \equiv \frac{\delta Q_n}{1-Z},\end{aligned} \tag{6}$$

where $Z = 2\int_{\Delta}^{\infty} f_0(E)(\Delta^2/E^3)(E/\epsilon)\,dE = 2\int_{\Delta}^{\infty}(-\partial f_0/\partial E)(\epsilon/E)\,dE$ is the function discussed by Clarke *et al.*[13] Near T_c, $Z \to 1 - \pi\Delta/4k_BT$, so that Q^* is "amplified"[14] by a factor $4k_BT/\pi\Delta$ relative to δQ_n. As $T\to 0$, however, Z becomes exponentially small, and Q^* is nearly equal to δQ_n. Equating the expression for Q^* obtained from (5) and (6) with[3] $2N(0)g_{NS}eV$, we obtain

$$eV = \tfrac{2}{3}p_F\,l\,\frac{\partial f_0(\Delta)/\partial T}{g_{NS}(1-Z)}\,\vec{v}_s \cdot \nabla T. \tag{7}$$

Writing $\vec{v}_s = \vec{J}_s/n_s e$, where n_s is determined from[7] $\lambda^2(T) = mc^2/4\pi n_s e^2 \approx \lambda^2(0)(1-t^4)^{-1}$, and evaluating $\partial f_0(\Delta)/\partial T$, we find from (7)

$$V = \frac{2\pi}{3}\frac{v_F}{c}\, l\, \frac{\lambda^2(0)}{1-t^4} \times \frac{\Delta}{k_B T \cosh^2(\Delta/2k_B T) g_{NS}(1-Z)} \frac{\vec{J}_s \cdot \nabla T}{cT}. \quad (8)$$

Since the factor $\Delta/(1-Z)$ varies only from $4k_B T_c/\pi$ at T_c to $\Delta(0) = 1.76 k_B T_c$ at $T=0$, the temperature dependence is dominated by the divergence of $\lambda^2 \sim n_s^{-1} \sim (1-t)^{-1}$ near T_c and the exponential temperature dependences at low temperatures which reflect the freezing out of the quasiparticle population. Sufficiently near T_c, (8) simplifies to $V = 2v_F l \lambda^2(0) \vec{J}_s \cdot \nabla T / 3c^2 g_{NS}(1-t)T$. To estimate the voltage expected near T_c for the only clean Sn sample (No. 4) of the experimental work[4] we set $v_F = 0.65\times10^8$ cm/sec, $l = 4.3\times10^{-5}$ cm, and $\lambda(0) = 5.0\times10^{-6}$ cm to find $V g_{NS} T(1-t)/J_s|\nabla T| = 0.5\times10^{-16}\ \Omega\ \text{cm}^3$, in reasonable agreement with the measured value of $1.2\times10^{-16}\ \Omega\ \text{cm}^3$.

In order to test the predicted temperature dependence more fully, we return to (8). Figure 2 shows the remarkably good fit of this function to the experimental data of Ref. 4, fitted at a single point. Note that this formula reproduces even such a subtle feature as the rise of the data above the simple $1/(1-t)$ behavior before the fall at low temperatures dictated by the $\cosh^2(\Delta/2k_B T)$ factor.

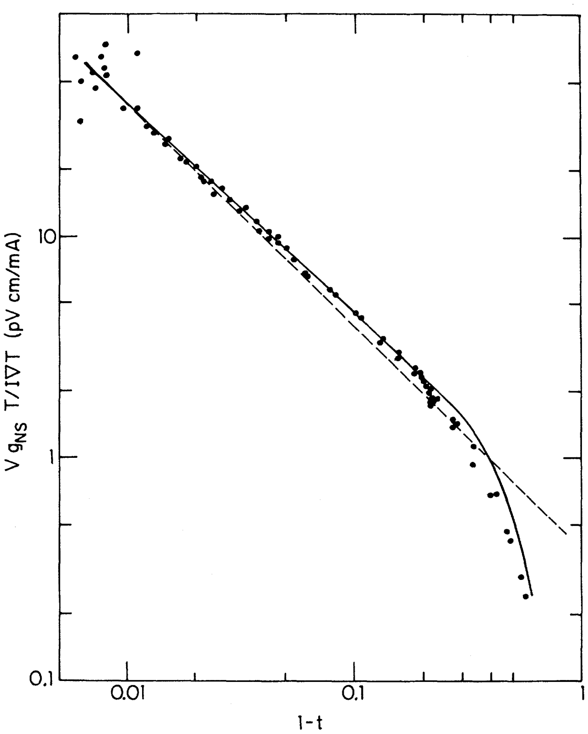

FIG. 2. $V g_{NS} T/I|\nabla T|$ vs $(1-t)$. Solid circles represent experimental data; dashed line has slope of −1; solid line is (8), fitted at one point.

It is of interest to compare the processes causing charge imbalance in this experiment with those in situations where the generation occurs at a well-defined spatial discontinuity, as in tunnel injection,[2] or current through an N-S interface[15] or phase-slip center.[16] In these latter experiments, charge generation and relaxation processes are quite distinct, being even spatially separated. Thus, for example, if one enhances the Q^* relaxation rate by the application of a magnetic field[17] or addition of magnetic impurities[18] while retaining the same generation rate, the steady-state value of Q^* will decrease. By contrast, in the present experiment the charge imbalance generation is a *volume* process, not a surface one, and generation and relaxation processes cannot be independently varied. As a result, the net branch imbalance charge is imposed by the presence of the temperature gradient, the magnitude of the effect depending only on the transport mean free path which limits the distance over which the gradient is effective in producing a nonequilibrium population. If this view is correct, *times* such as τ_E and τ_{Q^*} play no direct role in determining the magnitude of Q^*, and hence introduction of a small concentration of magnetic impurities (for example) should not change the size of the effect, in contrast to the situation in the other experiments cited above. Our viewpoint also provides a natural explanation for the fact that Schmid and Schön[19] find results differing from ours only by logarithmic factors in a diverse variety of limiting cases (clean, dirty, strong pair breaking, weak pair breaking) in which they make approximate solutions of the Boltzmann equation.

One of us (M.T.) would like to acknowledge the hospitality of the Institut für Theorie der Kondensierten Materie, Universität Karlsruhe, and is a Senior U. S. Scientist Awardee of the Alexander von Humboldt Foundation and the other (J. C.) acknowledges the hospitality of the same institution and the Cavendish Laboratory, Cambridge, and is in receipt of a Guggenheim Foundation Fellowship. We are grateful to B. R. Fjordbøge, P. E. Lindelof, C. J. Pethick, G. Schön, A. Schmid, and H. Smith for many helpful discussions. The support of this work by the U. S. National Science Foundation, the U. S. Office of Naval Research, and the U. S. Department of Energy is gratefully acknowledged.

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

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

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

[4]J. Clarke, B. R. Fjordboge, and P. E. Lindelof, Phys. Rev. Lett. 43, 642 (1979).

[5]If $\nabla T \neq 0$, there is a small superfluid velocity even in the absence of an applied current. The resulting potential $\sim |\nabla T|^2$ is much smaller than the effect treated here. It was discussed by D. F. Heidel and J. C. Garland [J. Phys. (Paris), Colloq. 39, C6-492 (1978)] using an approach similar to that of Ref. 1.

[6]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)].

[7]See, for example, M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975).

[8]J. M. Ziman, *Principles of the Theory of Solids* (Cambridge Univ. Press, 1964), Chap. 7.

[9]J. Bardeen, G. Richayzen, and L. Tewordt, Phys. Rev. 113, 982 (1959); P. Wyder, Rev. Mod. Phys. 36, 116 (1964).

[10]R. Orbach, private communication. Moreover, Yu. M. Galperin, V. L. Gurevitch, and V. I. Kozub, Zh. Eksp. Teor. Fiz. 65, 1045 (1973) [Sov. Phys. JETP 38, 517 (1974)], have shown explicitly that the usual Boltzmann equation [e. g., Eq. (7.14) in Ref. 8] can be used to describe quasiparticles in a superconductor in the presence of a superfluid velocity, provided one uses (2) for E_k. The solution to this Boltzmann equation [e. g., Eq. (7.92) of Ref. 8] is then (3).

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

[12]A. M. Kadin, L. N. Smith, and W. J. Skocpol, to be published.

[13]J. Clarke, U. Eckern, A. Schmid, G. Schön, and M. Tinkham, Phys. Rev. B 20, 3933 (1979).

[14]This "amplification" does not shift the steady-state value of Q^* in an "injection" experiment, since the rates of generation and relaxation are "amplified" equally; it simply speeds up the approach to equilibrium. (See Ref. 12.) Here, However, δf_k itself is specified, and the factor $(1-Z)^{-1}$ enters naturally.

[15]A. B. Pippard, J. G. Shepherd, and D. A. Tindall, Proc. Roy. Soc. London, Ser. A 324, 17 (1971).

[16]W. J. Skocpol, M. R. Beasley, and M. Tinkham, J. Low Temp. Phys. 16, 145 (1974).

[17]A. M. Kadin, W. J. Skocpol, and M. Tinkham, J. Low Temp. Phys. 33, 481 (1978).

[18]T. R. Lemberger and J. Clarke, Bull. Am. Phys. Soc. 24, 328 (1979).

[19]A. Schmid and G. Schön, Phys. Rev. Lett. 43, 793 (1979).