Pair-field susceptibility and superconducting tunneling: A macroscopic approach

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The dc I-V characteristics of tunnel junctions between weak and strong superconductors characterized by transition temperatures T_c and T_c', respectively ($T \approx T_c << T_c'$), exhibit an excess pair current. This current is proportional to the imaginary part of the generalized pair-field susceptibility $\chi(k,\omega)$, which characterizes the dynamics of the weak superconductor, where ω and k are proportional, respectively, to the voltage across and the magnetic field in the junction. We show here that this result follows in a simple way from the interaction that gives rise to the usual Josephson current below T_c, and can in fact be viewed as a second-order Josephson effect. Above T_c the order parameter on the strong side acts as an effective conjugate field to induce an order parameter on the weak side, yielding a proportional Josephson current between the two electrodes. We also interpret this result to yield a charge-imbalance relaxation time for the effective gapless superconductor on the weak side. Below T_c the induced part of the order parameter interacts with the usual Josephson current to produce an excess current in a manner analogous to the way an external oscillator coupled to a junction results in a constant-voltage step in the I-V characteristic. In both temperature regimes the magnitude of this excess current is related to an appropriate time-dependent Ginzburg-Landau equation. Finally, the effect of thermal voltage noise on the excess current is considered. Throughout, a heuristic approach is used to bring out new aspects of the problem and to make the physical content of the theory accessible.

I. INTRODUCTION

The study of the dynamics of the superconducting order parameter is of great interest, in part because it provides a test of the application of the ideas of nonequilibrium statistical mechanics to superconductors where theory has worked so well for equilibrium properties, and in part because dynamical theory may have some real application to some of the many superconducting devices that have been under development. Therefore, one would like to infer experimentally, as directly as possible, the form of the equations that govern the time evolution of the order parameter. The experimental determination of the frequency and wave-vector-dependent pair-field susceptibility is in many instances the most direct probe of order-parameter dynamics. Although it has been studied theoretically[1-4] and experimentally[5-7] for more than a decade, the physical basis of the measurement of the pair-field susceptibility and its connection with more familiar aspects of the Josephson effect and the macroscopic equations for the superconducting order parameter have not been described completely.

In magnetic systems, the most direct methods for the determination of the dynamics of the magnetization M involve the measurement of the wave-vector- and frequency-dependent magnetic susceptibility $\chi_M(k,\omega)$, and the structure function $S(k,\omega)$ related via the fluctuation-dissipation theorem. A superconducting system does *not* have a thermodynamic conjugate field which will induce superconductivity in a normal metal in the same way that a laboratory magnetic field H produces a finite magnetization in a paramagnet.[1] However, in both the superconducting and magnetic cases, within the spirit of mean-field theory, one can treat the internal interaction within the material as an effective field acting on a given region of the material. In the superconductor, this internal interaction is long range (on the order of the coherence length ξ) and gives rise to the well-known proximity effect in a clean contact between a superconductor and a normal metal. It has perhaps not been generally appreciated, however, that the same type of physical effect (although weaker) occurs across a tunneling barrier, and it is precisely this type of coupling that gives rise to the usual Josephson effects between two superconducting electrodes. Thus, even in a tunnel junction between a superconductor and a normal metal, there is a small induced order parameter on the normal side. Furthermore, there is an "excess" supercurrent across the junction, which is a measure of the generalized superconducting pair-field susceptibility $\chi(k,\omega)$. Similar considerations are relevant when both electrodes are superconducting, and the excess pair current for $V\neq 0$ is also related to the susceptibility.[6]

In this paper we will consider in detail a geometry consisting of a tunnel junction between two films at a temperature T such that one (the "primed film")is far below its critical temperature T_c', and the other (the "unprimed film") is above, near, or slightly below its transition at T_c. This corresponds to the usual experimental situation, and means that the background quasiparticle current across the junction will be small, and that the effect on the order parameter of the primed film is likely to be weak. In order to understand the physical basis for the experimental determination of the pair-field susceptibility we will examine the form of the excess pair current and its relation to the susceptibility within the context of time-dependent Ginzburg-Landau theory, in contrast to earlier detailed derivations which relied on Green's-function formalism[3,4] or linear response theory.[2] This approach represents a generalization of the initial treatment of the problem of the pair-field susceptibility by Ferrell.[1] Part of the procedure is also related to the work of Kulik[8] on the fluctuation resistance of a tunnel junction above its critical temperature. Throughout, connections will be made to other types of measurements and other theoretical viewpoints, in order to clarify the underlying physics. We will first examine the case of $T>T_c$, followed by the more complex regime $T<T_c$. Then we will consider the effect of thermal noise and conclude with a general discussion.

II. EXCESS PAIR TUNNELING CURRENT FOR $T>T_c$

In order to calculate the excess supercurrent above T_c, we start with the usual Ginzburg-Landau free-energy density in the unprimed film,

$$f_0=\alpha\,|\psi|^2+\beta/2\,|\psi|^4 -(\hbar^2/2m^*)\,|\nabla\psi-(2ie/\hbar)A\,|^2 , \quad (1)$$

where $\psi=|\psi|e^{i\theta}$ is the complex order parameter in the film. Above T_c the quartic term can be ignored. In the present case in which the film is one electrode of a tunnel junction, an additional term in the free energy is necessary to account for the superconducting interaction between the two films of the junction. We present this interaction by the Josephson free energy $F_J=-(\hbar I_0/2e)\cos\gamma$, which gives rise in the usual way to the supercurrent in the junction $I_s=I_0\sin\gamma$. Here γ is the gauge-invariant phase difference across the barrier, and I_0 is the critical current, which is inversely proportional to the normal-state junction resistance R_N and goes as $|\psi|$ (provided this quantity is small). We emphasize that because of this interaction, there will be an induced order parameter which permits the definition of a nonzero supercurrent even above T_c. The only difference between this case and that of the usual Josephson current, as will be shown below, is that since both the magnitude and phase of this induced order parameter are fixed by the interaction, there is not the internal degree of freedom associated with the phase which characterizes the usual supercurrent-phase relation below T_c.

If we assume that the thickness $d<<\xi\equiv(\hbar^2/2m^*|\alpha|)^{1/2}$, the effective coherence length in the film, the interaction is uniform and we can express it in terms of a free-energy density

$$f_J=(\hbar j_0/2ed)\cos\gamma =(\hbar\bar{C}/2ed)\,|\psi|\cos\gamma , \quad (2)$$

where $j_0=\bar{C}\,|\psi|$ is a critical current density such that $j_J=j_0\sin\gamma$ is the supercurrent density in the junction. In the regime of interest, where $|\psi|$ is small and $T\approx T_c<<T_c'$, j_0 is given by[1]

$$j_0\approx(G_N\Delta_{\rm in}/4e)\ln(T_c'/T) , \quad (3)$$

where G_N is the normal conductance per unit area of the junction, and $\Delta_{\rm in}$ is the induced BCS gap parameter related to $|\psi|$ (for a dirty superconductor) by

$$\Delta_{\rm in}=|\psi|(4k_BT\hbar/\pi n\tau)^{1/2} . \quad (4)$$

Here n and τ are the electron density and impurity scattering time in the normal state.

Consider the case where there is a constant voltage V_0 across the junction and an external magnetic field H_0 in the plane of the junction (see Fig. 1).

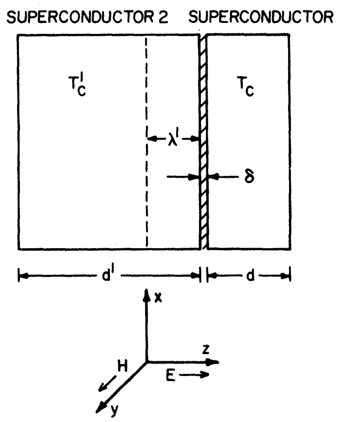

FIG. 1. Schematic of tunnel junction for measurement of pair-field susceptibility, for $T \approx T_c << T_c'$. The magnetic field is in the y direction, the pair field is modulated in the x direction, and the electric field and current flows are in the z direction.

Then γ can be written

$$\gamma=\theta-2eV_0t/\hbar-(2e/\hbar)\int A\,dz$$
$$=\theta-\omega_0t-q_0x\ , \tag{5}$$

where

$$q_0=(2e/\hbar)(\lambda'+d/2+\delta)H_0\ , \tag{6}$$

and it is assumed that the penetration depth λ' of the primed film is much less than its thickness, while $d<<\lambda$ for the unprimed film (δ is the barrier thickness, usually negligible). Then

$$f_J=-\frac{\hbar\overline{C}}{4ed}\{\psi\exp[-i(q_0x+\omega_0t)]+\psi^*\exp[+i(q_0x+\omega_0t)]\}\ . \tag{7}$$

If we define $f=f_0+f_J$, one can perform the usual variational calculation[9] to obtain the time-dependent Ginzburg-Landau (TDGL) equation

$$\tau_{\rm GL}\,|\alpha|\left[\frac{\partial}{\partial t}-\frac{2ie\phi}{\hbar}\right]\psi=\frac{\delta f}{\delta\phi^*}\ , \tag{8}$$

where $\tau_{\rm GL}=\pi\hbar/8k_B\,|T-T_c|$ is the Ginzburg-Landau relaxation time and ϕ is the local electric potential. This leads to

$$\tau_{\rm GL}\frac{\partial\psi}{\partial t}+\psi-\xi^2\nabla^2\psi=C_1\exp[i(q_0x+\omega_0t)]\ , \tag{9}$$

where $C_1=\hbar\overline{C}/(4ed\,|\alpha|)$ and we take $\phi=0$ (the other film is at voltage V_0), and also neglect A in $\nabla-2ieA/\hbar$, since for small fields its effect can be largely accounted for as a shift in T_c.[10] The quantity on the left of Eq. (9) is the usual TDGL equation for $T>T_c$. If the right-hand side were equal to zero, then in steady state one would have $\psi=0$. If the quantity on the right included thermal fluctuations, we would still have the thermal average of the order parameter $\langle\psi\rangle=0$, although $\langle|\psi|^2\rangle\neq0$, giving rise to excess electrical conductance of the film in the normal state above T_c.[11] In the present case, the quantity on the right can be viewed as an effective "pair field" $\zeta(x,t)$ which is conjugate to the order parameter and excites a particular mode of oscillation with wave vector q_0 and frequency ω_0:

$$\zeta(x,t)=\alpha C_1\exp[i(q_0x+\omega_0t)]\ . \tag{10}$$

(The normalization has been chosen so that $\zeta=-\partial f_J/\partial\psi^*$.) It is a remarkable fact, attributable to the gauge invariance of the Josephson coupling, that the application of time-and-space—independent electromagnetic fields in the junction leads to a temporally and spatially modulated effective pair field.

The induced order parameter which satisfies Eq. (9) takes the form

$$\psi_{\rm in}=\frac{C_1\exp[i(q_0x+\omega_0t)]}{i\omega_0\tau_{\rm GL}+(1+\xi^2q_0^2)}\ . \tag{11}$$

This can also be seen within the context of linear response theory, where one can define the order-parameter response function $\chi(x-x',\,t-t')$ such that

$$\psi_{\rm in}(x,t)=\int\int\zeta(x',t')\chi(x-x',t-t')dx'dt'\ . \tag{12}$$

In terms of the Fourier transforms of these quantities, this becomes

$$\psi_{\rm in}(k,\omega)=\zeta(k,\omega)\chi(k,\omega)\ , \tag{13}$$

where $\chi(k,\omega)$ is the wave-vector- and frequency-dependent order-parameter susceptibility. In the present case

$$\zeta(k,\omega)=\alpha C_1\delta(k-q_0)\delta(\omega-\omega_0) \tag{14}$$

and

$$\chi(k,\omega)=\{\alpha[i\omega\tau_{\rm GL}+(1+k^2\xi^2)]\}^{-1}\ , \tag{15}$$

so that

$$\psi_{\rm in}(x,t)=\zeta(x,t)\chi(q_0,\omega_0)\ . \tag{16}$$

In other works,[2] χ is usually defined to correspond to $\Delta_{\rm in}$ rather than $\psi_{\rm in}$ as is done in Eq. (12). In that case, $\chi(k,\omega)$ would be normalized so that α in Eq. (15) is replaced by $N(0)(T-T_c)/T_c$, where $2N(0)$ is the normal electronic density of states at the Fermi level.

One can now obtain the time-average excess supercurrent $\bar{I}_{\rm ex}$ from

$$\begin{aligned}\bar{I}_{\rm ex}&=\int_0^w\int_0^L dx\,dy\bar{j}_J\\&=\bar{C}w\int_0^L dx\,\langle {\rm Im}\{\psi_{\rm in}(x,t)\exp[-i(q_0x+\omega_0t)]\}\rangle_t\\&=\frac{\hbar C^2wL}{4ed}{\rm Im}\chi(q_0,\omega_0)=\frac{\hbar}{4ed}\bar{C}^2\frac{wL}{\alpha}\frac{\omega_0\tau_{\rm GL}}{[(\omega_0\tau_{\rm GL})^2+(1+q_0^2\xi^2)^2]}\ .\end{aligned}\tag{17}$$

This general relationship between $\bar{I}_{\rm ex}$ and χ is also valid below T_c, as will be shown in the next section.

Equation (17) indicates that the excess current $I_{\rm ex}=I_0\sin\gamma$ is time independent (as is γ), even when $\omega_0=2eV_0/\hbar\neq0$. This appears to violate the ac Josephson relation,[12] but the resolution of this paradox can be achieved, together with significant physical insight, if we introduce the concept of charge imbalance.[13] The charge imbalance (or branch imbalance) Q^* is the net charge density of the quasiparticles which constitute the "normal fluid" of a two-fluid picture of a superconductor. In equilibrium the electronlike and holelike excitations taken together have zero net charge. In the nonequilibrium situation, the charge imbalance Q^* in the quasiparticles is balanced by an equal and opposite charge in the superfluid, which corresponds to a proportional shift in the chemical potential of the condensate by $\Phi=Q^*/2N(0)e$.

In the classic experiment of Clarke,[14] a charge imbalance is achieved by the tunneling of a normal current I_n across an oxide barrier from a normal metal electrode into a superconductor. Since this current will leave the superconducting film as supercurrent, there must be an accumulation of "normal charge" Q^* in the film, proportional in steady state to the time τ_{Q^*} in which this nonequilibrium distribution decays:

$$Q^*=I_n\tau_{Q^*}/\Omega\ ,\tag{18}$$

where Ω is the nonequilibrium volume. In the case of the determination of the pair-field susceptibility of a normal metal we are focusing on the normal film. In the absence of superconducting order, there can be no charge imbalance since relaxation cannot occur. However, because of the interaction with the strongly superconducting electrode, the normal film becomes effectively a "gapless superconductor" with a small order parameter. In the present case, the excess pair current is a supercurrent injected into the normal film, and this current presumably leaves the film at the boundaries as normal current. In the same way as before, this will produce a charge imbalance

$$Q^*=-I_{\rm ex}\tau_{Q^*}/\Omega\ ,\tag{19}$$

for some effective relaxation time τ_{Q^*}. The negative sign is taken because the process is essentially the opposite of that described by Eq. (18).

Through the chemical potential shift Φ, $Q^*\neq0$ affects the ac Josephson relation, which can be more correctly expressed in the form

$$\frac{d\gamma}{dt}=2\Delta\mu_s/\hbar\ ,\tag{20}$$

where $\Delta\mu_s$ is the difference across the junction of the condensate electrochemical potential $\mu_s=e\phi-\Phi$, and as before ϕ is the electric potential. The Josephson relation is obeyed only if $\Delta\mu_s=0$, which can occur only if $\Delta\Phi=eV_0$. If we neglect the small chemical potential shift on the strongly superconducting side, this implies that the relaxation time must satisfy the relation

$$\tau_{Q^*}=2N(0)e^2V_0Lwd/I_{\rm ex}\ .\tag{21}$$

Substituting the expression for $I_{\rm ex}$ from Eq. (17) into Eq. (21), and taking the low voltage $q_0=0$ limit, we obtain

$$\begin{aligned}\tau_{Q^*}&=\hbar^2N(0)/(4\alpha\tau_{\rm GL}C_1^2)\\&=mN(0)D/|\psi_{\rm in}|^2=2k_BT\hbar/\pi\Delta_{\rm in}^2\ ,\end{aligned}\tag{22}$$

where we have used the theory of dirty superconductors, including Eq. (4) and the relation $\xi^2=D\tau_{\rm GL}$ ($D=v_F^2\tau/3$ is the normal-state diffusivity), together with the fact that $2N(0)=3n/mv_F^2$.

The relaxation time inferred in Eq. (22) is identical to that derived by more standard methods[15] for charge imbalance relaxation in a gapless superconductor below T_c.

III. EXCESS PAIR CURRENT FOR $T<T_c$

A similar approach can be used to compute the excess current below T_c, although the details of the procedure are more complicated for several reasons. First, the simple time-dependent Ginzburg-Landau equation is no longer valid except in special cases, and a properly generalized TDGL equation is likely to be much more complicated, if indeed one exists at all. Second, there is an equilibrium nonzero value of the order parameter, and as shown below, the dynamical equations for small induced variations of the order parameter are different for variations in the magnitude ("longitudinal" mode) and variations in the phase ("transverse mode). Finally, there is, of course, the usual dc Josephson current associated with the equilibrium value of the order parameter, which is much larger than the excess pair current, and must be depressed using a voltage and/or magnetic field in order to see the desired effect.

We will begin by using the simple TDGL equation, which is expected to be valid for $T<T_c$ only for gapless superconductors. In the same way as we obtained Eq. (9), we have

$$\tau_{\rm GL}\frac{\partial\psi}{\partial t}-(1-|\psi|^2/\psi_{\rm eq}^2)\psi-\xi^2\nabla^2\psi =C_1\exp[i(q_0x+\omega_0t)] \;, \quad (23)$$

where the differences of sign relative to Eq. (9) are due to the fact that α has changed sign. Since we are looking for small variations in ψ about its equilibrium value $\psi_{\rm eq}\,e^{i\theta_{\rm eq}}$, we define

$$\psi\equiv\psi_{\rm eq}fe^{i\theta}\approx\psi_{\rm eq}(1+g+i\theta) \;, \quad (24)$$

where $g=1-f$ and θ are real functions small compared to unity, and $\theta_{\rm eq}=0$. Substituting this in, and separating the real and imaginary parts, we obtain

$$\left[\tau_{\rm GL}\frac{\partial}{\partial t}+2-\xi^2\nabla^2\right]g=C_1\cos(q_0x+\omega_0t)/\psi_{\rm eq} \;, \quad (25)$$

$$\left[\tau_{\rm GL}\frac{\partial}{\partial t}-\xi^2\nabla^2\right]\theta=C_1\sin(q_0x+\omega_0t)/\psi_{\rm eq} \;. \quad (26)$$

Clearly, the longitudinal variation of the order parameter $\delta\psi_L=\psi_{\rm eq}g$ exhibits different behavior than the transverse variation $\delta\psi_T=\psi_{\rm eq}i\theta$. Equations (25) and (26) can be solved to yield

$$\delta\psi^L=\zeta(x,t)\chi^L(q_0,\omega_0)+\text{c.c.} \;, \quad (27)$$

$$\delta\psi^T=\zeta(x,t)\chi^T(q_0,\omega_0)-\text{c.c.} \;, \quad (28)$$

where

$$\chi^L(k,\omega)=[2\,|\,\alpha\,|\,(i\omega\tau_{\rm GL}+2+\xi^2k^2)]^{-1} \quad (29)$$

and

$$\chi^T(k,\omega)=[2\,|\,\alpha\,|\,(i\omega\tau_{\rm GL}+k^2\xi^2)]^{-1} \;, \quad (30)$$

and the effective pair field $\zeta(x,t)$ is the same as in Eq. (10) and c.c. represents the complex conjugate. Note that this problem can no longer be described simply by linear response theory, since there is also a "conjugate-linear" response proportional to the complex conjugate of $\zeta(x,t)$. However, as the conjugate term gives a contribution at frequency $2\omega_0$, it is only the linear response term that contributes to the dc excess current:

$$\bar{I}_{\rm ex}=\frac{\hbar\bar{C}^2}{4ed}\,{\rm Im}[\chi^L(q_0,\omega_0)+\chi^T(q_0,\omega_0)] \;. \quad (31)$$

If we define the total susceptibility $\chi=\chi^L+\chi^T$, then this takes exactly the same form as Eq. (17) for $T>T_c$. This current is, of course, in addition to the usual Josephson supercurrent for $V=0$, $H\approx0$, $I_s=\bar{C}wL\psi_{\rm eq}\sin\gamma$. It is important to note that the excess supercurrent is of higher order in the small coupling constant $\bar{C}$, and can only be seen when the usual supercurrent is not present or is greatly reduced in magnitude.

In Eqs. (25) and (26), both g and θ obey diffusive equations, i.e., with only a first derivative in time. For BCS superconductors with a gap in the excitation spectrum, both the experimental observations[6] and the theory[16–18] suggest that the transverse mode should be propagating, corresponding to a second time derivative and damped waves in the spectrum of excitations. As an example of how the present approach would deal with a generalized TDGL equation which contains this propagating mode, consider the equation of Kramer and Watts-Tobin,[19] as modified by Skocpol and Octavio[20]:

$$\frac{\tau_{\rm GL}}{(1+\Gamma^2 f^2)^{1/2}}\left[\frac{\partial}{\partial t}\frac{2ie\phi}{\hbar}+\frac{2i\tau_E}{\hbar}\dot{\Phi}+\frac{\Gamma^2}{2}\frac{\partial f^2}{\partial t}\right]\psi=(1-f^2)\psi+\xi^2\left[\nabla-\frac{2ieA}{\hbar}\right]^2\psi\,, \tag{32}$$

where τ_E is the inelastic scattering time, and $\Gamma=2\tau_E\Delta(T)/\hbar$. We will work in the experimentally accessible limit $\Gamma>>1$. Note that as $\Gamma\rightarrow 0$, Eq. (32) reduces to the simple TDGL equations, except for the term $\dot{\Phi}$. This term was added to account for charge imbalance waves,[21] a counterflow of normal and supercurrent which constitutes the progagating transverse mode discussed above.

Adding in the pair field ζ and proceeding as before, we obtain the pair of equations

$$\tau_{\rm GL}\Gamma\dot{g}+2g-\xi^2\nabla^2 g =[\hbar\bar{C}/4ed\,|\,\alpha\,|\,\psi_{\rm eq}(t)]\cos(q_0x+\omega_0 t)\,, \tag{33}$$

$$(2\tau_{\rm GL}/\hbar\Gamma)(\Phi+\tau_E\dot{\Phi})-\xi^2\vec{\nabla}\cdot\vec{q} =[\hbar\bar{C}/4ed\,|\,\alpha\,|\,\psi_{\rm eq}(T)]\sin(q_0x+\omega_0 t)\,, \tag{34}$$

where $\vec{q}=\vec{\nabla}\theta-2ie\vec{A}/\hbar$ is the gauge-invariant phase gradient. From the first equation we obtain

$$\chi^{(L)}(k,\omega)=[2\,|\,\alpha\,|\,(i\omega\tau^{(L)}+2+\xi^2k^2)]^{-1}\,, \tag{35}$$

where $\tau^{(L)}=\tau_{\rm GL}\Gamma=\pi^3k_BT\tau_E/7\zeta(3)\Delta(T)$ is the longitudinal relaxation time for a superconductor with a gap.[15] $[\zeta(3)\equiv 1+2^{-3}+3^{-3}+\cdots=1.202\ldots]$

We can solve the transverse equation by applying the condition of electroneutrality within a two-fluid picture, that $\vec{\nabla}\cdot\vec{J}=0$, where $J=J_n+J_s$ is the total current density, $J_n=\sigma E$ the normal current, and $J_s=|\,\psi\,|^2e\hbar q/m$ the supercurrent. Using this to eliminate q, we obtain the following wave equation in Φ:

$$\left[\tau_J\tau_E\frac{\partial^2}{\partial t^2}+(\tau_J+\tau_E)\frac{\partial}{\partial t}+1-\Lambda^2\nabla^2\right]\Phi=\frac{\hbar\Gamma}{2\tau_{\rm GL}}\frac{\hbar C}{4ed\,|\,\alpha\,|\,\psi_{\rm eq}(T)}[\omega_0\tau_J\cos(q_0x+\omega_0t)+\sin(q_0x+\omega_0t)]\,, \tag{36}$$

where

$$\tau_J=\tau_{\rm GL}/u=2k_BT\hbar/\pi\Delta^2(T)\qquad [u=\pi^4/14\zeta(3)=5.79] \tag{37}$$

is the supercurrent relaxation time, and

$$\Lambda=\xi(\Gamma/u)^{1/2}=(D\tau_{Q^*})^{1/2} \tag{38}$$

is the quasiparticle diffusion length corresponding to the charge imbalance relaxation time $\tau_{Q^*}=4k_BT\tau_E/\pi\Delta(T)$. From this, we obtain the transverse susceptibility

$$\chi^{(T)}(k,\omega)=\Gamma(1+i\omega\tau_J)/\{2(i\omega\tau_{\rm GL})[(1+i\omega\tau_J)(1+i\omega\tau_E)+\Lambda^2k^2]\}\,. \tag{39}$$

For high frequencies $\omega>>\tau_J^{-1}$, τ_E^{-1}, this clearly exhibits a resonance at a frequency

$$\omega=k\Lambda/(\tau_J\tau_E)^{1/2}\,, \tag{40}$$

corresponding to a wave with velocity

$$v=\Lambda/(\tau_J\tau_E)^{1/2}=[2D\Delta(T)/\hbar]^{1/2}\,. \tag{41}$$

The same will also be true, of course, in the expression for the excess current given by Eq. (31).

IV. EFFECT OF THERMAL VOLTAGE NOISE ON EXCESS PAIR CURRENT

The previous derivations assumed that the voltage across the tunnel junction is a constant. In real junctions, there will always be some voltage noise, which will act to broaden the peaks in the excess pair current

$I_{ex}(V)$. The calculation of the effect of voltage noise, which is rather similar in detail to the calculation of the thermally induced radiation linewidth in Josephson junctions,[22] is summarized below.

Consider a tunnel junction with effective dynamic resistance R and capacitance C. In addition to the dc voltage V_0 across the junction, there is also a thermally generated noise voltage V_n which assumed to be described by Gaussian statistics. The spectral density $J(\omega)$ of the noise is given by

$$J(\omega)=\langle\,|V_n(\omega)|^2\rangle=\frac{1}{2\pi}\int d\tau\langle V_n(t)V_n(t+\tau)\rangle_t e^{i\omega t}=2k_BTR/\pi[1+\omega^2(RC)^2]\ , \tag{42}$$

where $\langle\ \rangle_t$ represents a time average.

The expression for both ψ_{in} and I_{ex} must be modified to include V_n. The spatial dependence carries through as before, so we will consider the $q=0$ case. The effect of the noise is to introduce an additional time-dependent phase factor which results in

$$\psi_{in}(t)=(\hbar\bar{C}/4ed)\int dt'\exp\left[i\omega_0 t'+\left[\frac{2ie}{\hbar}\right]\int^{t'}V_n(t'')dt''\right]\chi(t-t')\ . \tag{43}$$

The excess current is then of the form

$$\begin{aligned}I_{ex}(V_0)&=(\bar{C}wL)\mathrm{Im}\left\{\left\langle\psi_{in}^*(t)\exp\left[-i\omega_0 t-\left[\frac{2ie}{\hbar}\right]\int^{t}V_n(t'')dt''\right]\right\rangle_t\right\}\\&=(\hbar\bar{C}^2wL/4ed)\mathrm{Im}\int dt'\left\langle e^{-i\omega_0(t-t')}\chi(t-t')\exp\left[\frac{2ie}{\hbar}\right]\int_{t'}^{t}V_n(t'')dt''\right\rangle_t\ .\end{aligned} \tag{44}$$

Changing variables, the time average can be brought into a single exponential factor:

$$I_{ex}=(\hbar C^2wL/4ed)\mathrm{Im}\int d\tau e^{-i\omega_0\tau}\chi(\tau)F(\tau)\ , \tag{45}$$

where

$$F(\tau)=\left\langle\exp\left[\left[\frac{-2ie}{\hbar}\right]\int_0^{\tau}V(t-t_1)dt_1\right]\right\rangle_t=\exp\left[\left[\frac{-2e^2}{\hbar^2}\right]\int_0^{\tau}\int_0^{\tau}dt_1dt_2\langle V(t-t_1)V(t-t_2)\rangle_t\right]\ . \tag{46}$$

This last reduction is a standard result from the statistics of Gaussian random noise.[23] Following Ref. 22,

$$\begin{aligned}F(\tau)&=\exp\left[\left[\frac{-8e^2}{\hbar}\right]\int d\omega\,J(\omega)\sin^2(\omega\tau/2)/\omega^2\right]\\&=\exp(-4e^2Rk_BT/\hbar^2)\{\,|\tau|+RC[\exp(-|\tau|/RC)-1]\}\ .\end{aligned} \tag{47}$$

From this we take the Fourier transform to obtain the real distribution $\tilde{F}(\omega)$, which can be expressed in terms of a confluent hypergeometric function,[22] but in the limits RC small (compared to the relevant times in χ) and RC large, it is given simply by

$$\tilde{F}(\omega)=\begin{cases}\Gamma_1/[\pi(\omega^2+\Gamma_1^2)] & (RC\text{ small})\\ \exp(-\omega^2/\Gamma_2^2)/(\pi^{1/2}\Gamma_2) & (RC\text{ large})\ ,\end{cases} \tag{48}$$

where $\Gamma_1=4e^2Rk_BT/\hbar^2$ and $\Gamma_2=(2e/\hbar)(2k_BT/C)^{1/2}$. In terms of $\tilde{F}(\omega)$, Eq. (44) can be written

$$I_{ex}(V_0)=(\hbar\bar{C}^2wL/4ed)\mathrm{Im}\int d\omega\,\chi(\omega_0+\delta\omega)\tilde{F}(\delta\omega)=\int d(2e\delta V/\hbar)I_{ex}^0(V_0+\delta V)\tilde{F}(2e\delta V/\hbar)\ , \tag{49}$$

where $I_{ex}^0(V)$ is the excess current in the absence of noise.

As an example, consider the case with $T>T_c$ and RC small. Then

$$I_{ex}=\frac{\hbar\bar{C}^2wL}{4ed\alpha}\frac{\omega_0/\Gamma}{(\omega_0/\Gamma)^2+(1+q_0^2\xi^2)^2}\ , \tag{50}$$

where $\Gamma=1/\tau_{GL}+\Gamma$. In general, thermal voltage noise will act to broaden peaks in the excess current, but it will not shift their centers. Reasonable estimates of the parameters R and C suggest that thermal voltage noise may be an important consideration in attaining a quantitative fit between theory and experiment.

Any other type of voltage noise present in an actual experiment can be treated in much the same way, and will have a similar effect. Fluctuation in the magnitude of the magnetic field in the junction will similarly broaden the peaks in the dependence $I_{\text{ex}}(H)$. Finally, an external rf signal could also be treated within this picture in a similar manner.

V. DISCUSSION AND CONCLUSIONS

In this concluding section we will review the major results established earlier, and discuss their relationship to other points of view in order to illustrate the physical significance of these results. The fundamental origin of the excess pair current is the Josephson coupling between the strong superconductor and the weakly superconducting or normal film. This can be expressed in terms of a free-energy density $f_J = -(\hbar\bar{C}/2ed)\,|\,\psi\,|\cos\gamma$ where $\bar{C}\sim 1/R_N$ (R_N is the normal-state resistance of the tunnel junction). The coupling acts as a weak generalized proximity effect, and can be characterized in terms of an effective pair field

$$\zeta(x,t) = (\hbar\bar{C}/4ed)\exp[i(q_0x+\omega_0 t)]\ , \tag{51}$$

acting across the junction, where $q_0 \sim H_0$ and $\omega_0 \sim V_0$. It is a remarkable fact, attributable to the gauge invariance of the Josephson coupling, that dc electromagnetic fields in the junction give rise to this manifestly ac effective pair field. For $T > T_c$, $\zeta(x,t)$ gives rise to an induced order parameter given by

$$\psi_{\text{in}}(x,t) = \zeta(x,t)\chi(q_0,\omega_0)\ , \tag{52}$$

which when substituted into the usual expression for the dc Josephson current density $j_J = \bar{C}\,|\,\psi\,|\sin\gamma$, results in the excess pair current of the form

$$\begin{aligned} I_{\text{ex}} &= (4edA/\hbar)\text{Im}(\psi_{\text{in}}\zeta^*) \\ &= (4edA/\hbar)\,|\,\zeta\,|^2\text{Im}[\chi(q_0,\omega_0)]\ . \end{aligned} \tag{53}$$

(A is the area of the tunnel junction.) Evidently, $I_{\text{ex}} \sim 1/R_N^2$, as the square of the coupling between the two films. This is in contrast to the usual dc Josephson effect found when both films are below their transition temperatures where $I_c \sim 1/R_N$. Thus the excess pair current can be seen as a higher-order response due to the same Josephson coupling.

The pair-field susceptibility $\chi(k,\omega)$ can be determined from the definition of linear response and TDGL theory. Equation (53) is true in general, and does not depend on a particular TDGL equation, since any linearized dynamical equation would serve.[4] Even in the absence of a dynamical equation, more general theoretical techniques not discussed in the present paper would enable the calculation of χ from microscopic theory:

$$\chi(\vec{r}-\vec{r}\,',t-t') = \begin{cases} (-i/\hbar)\langle[\hat{\psi}(\vec{r},t),\hat{\psi}^\dagger(\vec{r}\,',t')]\rangle\ , & t>t' \\ 0, & t<t' \end{cases} \tag{54}$$

where $\hat{\psi}$ is the quantum-mechanical order parameter operator, [ ] represents the commutator of the operators, and $\langle\ \rangle$ the expectation value at finite temperature. This definition is equivalent to a standard susceptibility using quantum-mechanical linear response theory.[2]

We emphasize that although the results of measurements of the pair-field susceptibility are related to the spectrum of spontaneous thermal fluctuations within the weak superconductor, they are in no sense due to these fluctuations. The excess current being a Josephson current, it depends on the existence of a phase correlation across the barrier. This does not exist with the thermal fluctuations, which therefore do not contribute to the excess current. In a measurement of excess conductivity *within* a film above T_c, on the other hand, only correlations *within* the film are important, and to these thermal fluctuations *do* contribute. In fact, thermal fluctuations will act against well-defined features in the excess current. We have worked out explicitly the case of thermal fluctuations in the voltage across the junction, which can significantly broaden the peaks in $I_{\text{ex}}(V)$, but at least to first order should not affect its center or its overall intensity.

Another point which has not previously been emphasized is that the measurement of the excess supercurrent for $T > T_c$ can be viewed as the reverse of the usual charge-imbalance experiment. Here supercurrent is injected into a normal metal, and the effective relaxation time τ_{Q^*} goes as $1/I_{ex}$. For the simple TDGL equation, for $q_0 = 0$ and small V_0, $\tau_{Q^*} = 2k_B T\hbar/\pi\Delta_{in}^2$.

For $T < T_c$, the situation is somewhat more complicated due to the fact that there is an equilibrium order parameter ψ_{eq}. However, the same pair field $\zeta(x,t)$ as before induces a change $\delta\psi_{in} = \delta\psi^L + \delta\psi^T$ in order parameter, where $\delta\psi^L$ is along the direction of ψ_{eq} (in the complex plane) and $\delta\psi^T$ is perpendicular to it. These two components satisfy different equations with corresponding susceptibilities χ^L and χ^T, and

$$\delta\psi_{in} = \zeta(x,t)[\chi^L(q_0,\omega_0) + \chi^T(q_0,\omega_0)] + \zeta^*(x,t)(\chi^{L*} - \chi^{T*}) . \quad (55)$$

This is not strictly a linear response, but since $I_{ex} \sim \mathrm{Im}(\delta\psi_{in}\zeta^*)$ only the term linear in ζ contributes to the time-averaged $\bar{I}_{ex}$; the "conjugate linear" term yields a current $\sim\cos(2\omega_0 t)$. In this context, the ac pair field $\zeta(x,t)$ acts analogously to an external ac voltage of frequency $\omega = 2eV/\hbar$ across the Josephson junction. In the latter situation the external signal at ω mixes with the internal Josephson oscillator at ω to give a dc constant voltage step, together with an oscillatory current at 2ω. The position along the constant voltage step is related to the phase difference between the external and internal oscillators. In the present case, this phase difference is fixed by the interaction that gives rise to $\delta\psi_{in}$, so that the additional external degree of freedom is absent.

The form of $\delta\psi_{in}$ is also more complicated below T_c than above. There is no single simple TDGL equation which can describe a superconductor for $T < T_c$ under all conditions. The longitudinal mode is expected to obey a diffusive equation in general, except for a resonance at $\hbar\omega = \Delta_{eq}$.[24] The transverse mode is expected to be propagating for a BCS supercondutor, corresponding at high frequencies to charge imbalance waves with velocity $v = (2D\Delta/\hbar)^{1/2}$. We have used a generalized TDGL equation which satisfies these criteria (except for the gap resonance) to derive expressions for χ^L and χ^T and the resulting $\bar{I}_{ex}$. Whichever equation one uses, $\bar{I}_{ex} \sim 1/R_N^2$, just as for $T > T_c$.

It has been the aim of the present paper to generate more interest in, and understanding of, the concept of pair-field susceptibility measurements. In particular, although the excess pair current is small and often difficult to observe experimentally, these experiments provide what is in many cases the only direct check of the dynamical equations governing the order parameter.

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[1] R. A. Ferrell, J. Low Temp. Phys. 1, 423 (1969).
[2] D. J. Scalapino, Phys. Rev. Lett. 24, 1052 (1970).
[3] H. Takayama, Prog. Theor. Phys. (Kyoto) 46, 71 (1971).
[4] M. Dinter, J. Low Temp. Phys. 32, 529 (1978).
[5] J. T. Anderson, R. V. Carlson, and A. M. Goldman, J. Low Temp. Phys. 8, 29 (1972).
[6] R. V. Carlson and A. M. Goldman, Phys. Rev. Lett. 31, 880 (1973); 34, 11 (1975); J. Low Temp. Phys. 25, 67 (1976).
[7] F. E. Aspen and A. M. Goldman, Phys. Rev. Lett. 43, 307 (1979); J. Low Temp. Phys. 43, 559 (1981).
[8] I. O. Kulik, Zh. Eksp. Teor. Fiz. 59, 937 (1970) [Sov. Phys.—JETP 32, 510 (1971)].
[9] M. Cyrot, Rep. Prog. Phys. 36, 103 (1973).
[10] S. R. Shenoy and P. A. Lee, Phys. Rev. B 10, 2744 (1974).
[11] W. J. Skocpol and M. Tinkham, Rep. Prog. Phys. 38, 1049 (1975).
[12] R. A. Ferrell and D. J. Scalapino, private communication.
[13] C. J. Pethick and H. Smith, Ann. Phys. (New York) 119, 133 (1979).
[14] J. Clarke, Phys. Rev. Lett. 28, 1363 (1972).
[15] A. Schmid and G. Schön, J. Low Temp. Phys. 20, 207 (1975).
[16] A. Schmid and G. Schön, Phys. Rev. Lett. 34, 941 (1975).
[17] S. N. Artemenko and A. F. Volkov, Zh. Eksp. Teor. Fiz. 69, 1764 (1975) [Sov. Phys.—JETP 4 , 896 (1976)].
[18] O. Entin-Wohlman and R. Orbach, Ann. Phys. (New York) 116, 35 (1978); I. O. Kulik, O. Entin-Wohlman, and R. Orbach, J. Low Temp. Phys. 43, 591 (1981).
[19] L. Kramer and R. J. Watts-Tobin, Phys. Rev. Lett. 40, 1041 (1978).

[20]M. Octavio and W. J. Skocpol, Physica 107B, 173 (1981).
[21]A. M. Kadin, L. N. Smith, and W. J. Skocpol, J. Low Temp. Phys. 38, 487 (1980).
[22]A. I. Larkin and Yu N. Ovchinnikov, Zh. Eksp. Teor. Fiz. 53, 2159 (1967) [Sov. Phys.—JETP 26, 1219 (1968)].
[23]See, e.g., C. W. Helstrom, *Statistical Theory of Signal Detection,* 2nd ed. (Pergamon, New York, 1968), p. 53.
[24]G. Schön and V. Ambegaokar, Phys. Rev. B 19, 3515 (1979).