

## Theory of Phonon-Assisted Tunneling in Superconductors\*

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Taylor and Burstein have found a temperature-dependent current in Pb-Pb superconductor-oxide-superconductor tunnel junctions in excess of the usual thermally excited quasiparticle current. We here calculate the current arising from a process in which a phonon is absorbed by a ground-state pair on one side of the oxide barrier resulting in a pair of quasiparticles, one on each side of the barrier, and the transfer of one electron across the barrier. One-phonon normal and umklapp as well as two-phonon umklapp processes are examined. The one-phonon umklapp process dominates and yields a current in excellent agreement with the experimental excess current both as a function of temperature and of voltage.

### I. INTRODUCTION

TAYLOR and Burstein<sup>1</sup> have made a careful study of current versus voltage in several superconductor-metal oxide-superconductor tunnel junctions for voltages less than  $2\Delta$ , the energy to excite a pair of quasiparticles. After subtracting off the two-particle tunneling<sup>2</sup> which sets in at  $V=\Delta$  and the tunneling due to thermally excited quasiparticles,<sup>3</sup> they found an excess current which was strongly temperature-dependent for Pb-Pb junctions and temperature-independent for all other junctions they examined. They suggested and we here show that this temperature-dependent current is due to a phonon-assisted tunneling process. The same process must also be present in the weaker coupling superconductors but is masked by the temperature-independent "leakage" current (which is as yet unexplained) as well as by the thermally excited quasiparticle current.

*Note added in proof.* [We now believe the temperature-independent "leakage" current to be due to the tunneling of ground-state pairs which emit phonons thus conserving energy on tunneling (to be published).]

Bardeen<sup>4</sup> has shown that a many-particle tunneling matrix element may be defined in terms of basis functions which are eigenfunctions of the Hamiltonian on one side of and in the oxide barrier but drop rapidly to zero on the other side. Cohen, Falicov, and Phillips<sup>5</sup> have used this idea to write the total Hamiltonian  $H=H_a+H_b+H_T$ , where  $H_a$  and  $H_b$  are the exact Hamiltonians for the superconductors on either side of the barrier

$$H_T = \sum_{pq\sigma} T_{pq} C_{p\sigma} a^\dagger C_{q\sigma} b + T_{qp} C_{q\sigma} b^\dagger C_{p\sigma} a \quad (1)$$

transfers electrons from superconductor  $a$  to superconductor  $b$  and vice versa and  $T_{pq}$  are matrix elements

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<sup>1</sup> B. N. Taylor and E. Burstein, Phys. Rev. Letters **10**, 14 (1963).

<sup>2</sup> J. R. Schrieffer and J. W. Wilkins, Phys. Rev. Letters **10**, 17 (1963).

<sup>3</sup> I. Giaever and K. Megerle, Phys. Rev. **122**, 1101 (1961).

<sup>4</sup> J. Bardeen, Phys. Rev. Letters **6**, 57 (1961).

<sup>5</sup> M. H. Cohen, L. M. Falicov, and J. C. Phillips, Phys. Rev. Letters **8**, 316 (1962).

for tunneling of *normal electrons* across the oxide layer. Phonon-assisted tunneling may be obtained by including the screened electron-phonon terms,  $H_a^{e1ph}$  and  $H_b^{e1ph}$ , in the Hamiltonian,

$$H_a^{e1ph} = \sum_{\mathbf{G}, \mathbf{p}', \mathbf{p}, \mathbf{k}, \sigma} (\hbar/2M\omega_{\mathbf{k}s}NV)^{1/2} \hat{e}_{\mathbf{k}s} \cdot (\mathbf{k}+\mathbf{G}) I(\mathbf{k}+\mathbf{G}) \\ \times C_{p\sigma} a^\dagger C_{p'\sigma} b_{\mathbf{k}s}^a \delta(\mathbf{p}-\mathbf{p}'-\mathbf{k}+\mathbf{G}) + \text{H.c.}, \quad (2)$$

where  $\mathbf{G}$  is a reciprocal lattice vector,  $V$  the volume of the superconductor,  $N$  the number density of ions of mass  $M$ ,  $\omega_{\mathbf{k}s}$  the frequency of a phonon of wave number  $\mathbf{k}$  in the  $s$ th mode whose polarization vector is  $\hat{e}_{\mathbf{k}s}$ , and  $b_{\mathbf{k}s}^a$  is the destruction operator for a phonon on side  $a$  of the barrier.  $I(\mathbf{p}-\mathbf{p}')$  is a matrix element which may be obtained either theoretically<sup>6</sup> or from comparison with experimentally measured high-temperature resistivity.<sup>7</sup> We neglect scattering by phonons in the thin oxide layer, which seems to be negligible compared to scattering by phonons in the superconductors.

The electron creation and destruction operators are written in terms of the quasiparticle operators by the Bogoliubov<sup>8</sup> transformation

$$C_{p\uparrow}^\dagger = u_p \gamma_{p\uparrow}^\dagger + v_p \gamma_{-p\downarrow} \\ C_{-p\downarrow}^\dagger = u_p \gamma_{-p\downarrow}^\dagger - v_p \gamma_{p\uparrow}, \quad (3)$$

and second-order perturbation theory (i.e., one order in  $H_T$  and one order either in  $H_a^{e1ph}$  or  $H_b^{e1ph}$ ) leads to the contributions diagrammed in Fig. 1. In 1(a) a ground-state pair on one side of the oxide layer is scattered by  $H_T$  into a pair of quasiparticles—one on each side of the layer—then a phonon is absorbed by the quasiparticle on side  $a$ . Figure 1(b) represents the same process except that the phonon is absorbed by the quasiparticle on side  $b$ . In Fig. 1(c) and 1(d) a ground-state pair on one side of the oxide layer is scattered by a phonon into a pair of quasiparticles on the same side, then one of the quasiparticles is scattered to the other side by  $H_T$ . The quasiparticles may have either electron

<sup>6</sup> D. Pines, Phys. Rev. **109**, 280 (1958).

<sup>7</sup> A. Rothwarf and M. Cohen, Phys. Rev. **130**, 1401 (1963).

<sup>8</sup> See, e.g., S. T. Beliaev, *The Many-Body Problem* (John Wiley & Sons, Inc., New York, 1959), p. 360 ff.

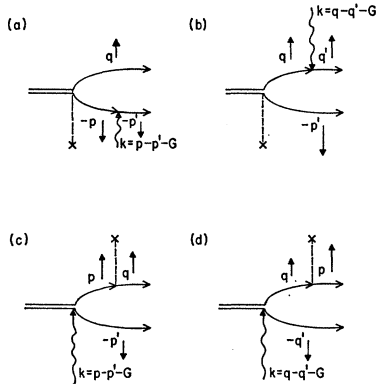


FIG. 1. Diagrams of phonon-assisted tunneling processes. (a) A ground-state pair on one side of the oxide layer is scattered by  $H_T$  into a pair of quasiparticles, one on each side of the oxide layer, then a phonon is absorbed by one of the quasiparticles; (b) same as (a) but the phonon is absorbed by the other quasiparticle; (c) a ground-state pair on one side of the oxide layer is scattered by a phonon into a pair of quasiparticles then one of the quasiparticles is scattered by  $H_T$ ; (d) same as (c) but the ground-state pair is on the other side of the barrier.

or hole-like character, so that current is always flowing from  $a$  to  $b$  in the diagrams just discussed even though the quasiparticles are going from  $b$  to  $a$  in half the processes.

## II. TUNNELING HAMILTONIAN INCLUDING PHONON SCATTERING

Although the procedure just discussed is straightforward and the diagrams give a good picture of the physics, the integrals over intermediate states required for the second-order perturbation theory are difficult if not impossible to perform. Instead of using the Bardeen procedure with its wave functions which are not good eigenfunctions everywhere, we deal with a basis set consisting of exact one-electron eigenfunctions.<sup>9</sup> Because of the inversion symmetry in our model (before the voltage drop is applied across the oxide barrier) the eigenfunctions may be written as  $\psi_p^e$  and  $\psi_p^o$ , even and odd under inversion. (Prange<sup>10</sup> has discussed tunneling from this point of view also.) These are degenerate to within  $1/V$ , where  $V$  is the volume of the superconductor. Since the barrier is not infinitely high  $\psi_p^o$  and  $\psi_p^e$  will differ by some small fraction of a wavelength giving a kinetic energy difference proportional to  $1/V$ . The contribution of the oxide layer region to the kinetic and potential energy expectation values of  $\psi_p^o$  and  $\psi_p^e$  will differ; but due to the normalization of the wave functions, this too is down by a factor  $1/V$ . Thus, we treat  $\psi_p^o$  and  $\psi_p^e$  as exactly degenerate.

We may now construct "left"- and "right"-side wave functions which are orthogonal to one another,

$$\psi_{ap} = (\psi_p^o + \psi_p^e)/\sqrt{2} \quad \psi_{bp} = (\psi_p^o - \psi_p^e)/\sqrt{2}. \quad (4)$$

<sup>9</sup> We will not need the wave function in the oxide layer since any effects due to the wave function in the oxide are down by a factor  $V_{ox}/V_{PB}$ .

<sup>10</sup> R. E. Prange, Phys. Rev. **131**, 1083 (1963).

If the barrier height were infinite,  $\psi_p^o$  and  $\psi_p^e$  would be exactly in phase and would result in  $\psi_{ap}$  and  $\psi_{bp}$  being truly right- and left-side wave functions. In general,  $\psi_p^o$  and  $\psi_p^e$  will be slightly out of phase resulting in a long tail on the "wrong" side of  $\psi_{ap}$  and  $\psi_{bp}$ . This tail is proportional to  $T_p$  where

$$T_p^2 = 16E_F V_0^{-2} \cos^2\theta (V_0 - E_F \cos^2\theta) e^{-2\beta d} \quad (5)$$

$$\beta = [2m\hbar^{-2}(V_0 - E_F \cos^2\theta)]^{1/2}.$$

Because only electrons at the Fermi surface will tunnel,  $E_F$  represents the energy of the electron,  $V_0$  the height of the barrier,  $d$  its thickness, and  $\theta$  the angle  $\mathbf{p}$ , the wave vector of the electron, makes with the barrier normal. (See Fig. 2.)

We now write the annihilation operator

$$\Psi(x) = \sum_{p\sigma} C_{p\sigma}^a \psi_{ap} + \sum_{q\sigma} C_{q\sigma}^b \psi_{bq}, \quad (6)$$

and the second quantized Hamiltonian

$$H = \int dx \Psi^\dagger(x) [\hat{p}^2/2m + V(x) + \sum_{\mathbf{kG}s} A_{\mathbf{kG}s} e^{i(\mathbf{k}+\mathbf{G}) \cdot \mathbf{x}} (\eta_{\mathbf{k}s}^a + \eta_{\mathbf{k}s}^b)] \Psi(x) + \frac{1}{2} \int \int \Psi^\dagger \Psi^\dagger Q \Psi \Psi, \quad (7)$$

where

$$A_{\mathbf{kG}s} = (\hbar/2M_{\mathbf{k}s}NV)^{1/2} \hat{e}_{\mathbf{k}s} \cdot (\mathbf{k} + \mathbf{G}) I(\mathbf{k} + \mathbf{G})$$

and

$$\eta_{\mathbf{k}s}^a = \hat{b}_{\mathbf{k}s}^{a\dagger} + \hat{b}_{-\mathbf{k}s}^a.$$

The Bardeen-Pines<sup>11</sup> canonical transformation is as-

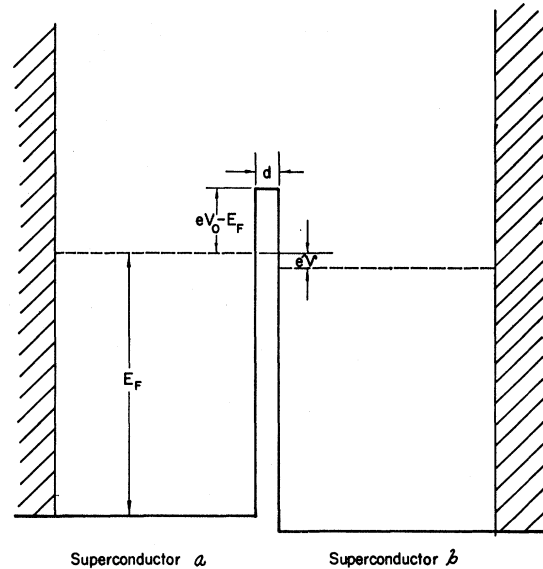


FIG. 2. Potentials in superconductor-metal oxide-superconductor junction. The voltage drop  $\mathcal{U}$  applied across the oxide layer of thickness  $d$  is 4 to 5 orders of magnitude smaller than the potential barrier height  $V_0$  and is drawn to an exaggerated scale here.

<sup>11</sup> J. Bardeen and D. Pines, Phys. Rev. **99**, 1140 (1955).

summed to have been made so that  $I(\mathbf{k}+\mathbf{G})$  is the matrix element for the *screened* electron-phonon interaction and  $Q$  contains the phonon-mediated attractive interaction between electrons. The Bogoliubov transformation [Eq.(3)] can now be used to diagonalize the Hamiltonian approximately.<sup>8</sup> Thus, the first term of  $H$  may be written

$$H = H_a + H_b + H_a^{\text{el ph}} + H_b^{\text{el ph}} + H_{ab}^{\text{el ph}} + H_{ba}^{\text{el ph}}, \quad (8)$$

where  $H_a^{\text{el ph}}$  is given by Eq. (2),

$$H_a = \sum_{p\sigma} \epsilon_p C_{p\sigma} a^\dagger C_{p\sigma}^a \quad (9)$$

and

$$H_{ab}^{\text{el ph}} = \sum_{\mathbf{G}\mathbf{p}\mathbf{q}\mathbf{k}\mathbf{s}\sigma} A_{\mathbf{k}\mathbf{G}\mathbf{s}} [T_{\mathbf{q}} b_{\mathbf{k}\mathbf{s}}^a + T_{\mathbf{p}} b_{\mathbf{k}\mathbf{s}}^b] \times \delta(\mathbf{q}-\mathbf{p}-\mathbf{k}-\mathbf{G}) C_{\mathbf{q}\sigma} b^\dagger C_{\mathbf{p}\sigma}^a + \text{H.c.} \quad (10)$$

$$H_{ba}^{\text{el ph}} = \sum_{\mathbf{G}\mathbf{p}\mathbf{q}\mathbf{k}\mathbf{s}\sigma} A_{\mathbf{k}\mathbf{G}\mathbf{s}} [T_{\mathbf{q}} b_{\mathbf{k}\mathbf{s}}^a + T_{\mathbf{p}} b_{\mathbf{k}\mathbf{s}}^b] \times \delta(\mathbf{p}-\mathbf{q}-\mathbf{k}-\mathbf{G}) C_{\mathbf{p}\sigma} a^\dagger C_{\mathbf{q}\sigma}^b + \text{H.c.} \quad (11)$$

Writing the electron operators in terms of the quasiparticle operators we may now obtain the phonon-assisted tunneling current taking  $H_{ab}^{\text{el ph}}$  and  $H_{ba}^{\text{el ph}}$  to only first order in perturbation theory. Two-phonon processes which previously were third order are now second order in perturbation theory (one order in  $H_{ab}^{\text{el ph}}$  or  $H_{ba}^{\text{el ph}}$  and one order in  $H_a^{\text{el ph}}$  or  $H_b^{\text{el ph}}$ ).

Because  $\psi_{a\mathbf{p}}$  and  $\psi_{b\mathbf{p}}$  are good orthogonal eigenfunctions on both sides of the barrier, the usual Bardeen tunneling term  $H_T$  does not appear in Eq. (8). This was to be expected; for the long tails on  $\psi_{a\mathbf{p}}$  and  $\psi_{b\mathbf{p}}$  have already accounted for this tunneling.

The second (interaction) term of the Hamiltonian [Eq.(7)] essentially vanishes under the Bogoliubov transformation except for a negligible remainder  $H_{\text{int}}$ . Included in this negligible term will be terms still smaller by a factor  $T$  which tunnel electrons across the barrier. Since the interaction term contains no phonon operators, it does not contribute to the phonon-assisted tunneling in any order.

### III. PHONON-ASSISTED TUNNELING CURRENT

We consider a voltage drop  $\mathcal{U}$  applied across the oxide barrier such that  $e\mathcal{U}$ , the energy gained by an electron traversing the barrier from  $a$  to  $b$  is less than  $2\Delta$ , the energy needed to create a quasiparticle pair. Thus, the only energy conserving processes induced by  $H_{ab}^{\text{el ph}}$  are those in which a phonon is destroyed. We further limit ourselves to voltages large enough and temperatures low enough that the reverse current induced by  $H_{ba}^{\text{el ph}}$  is negligible. We also require the temperature to be low enough for the thermally excited quasiparticles to be negligible. Then from Eq. (3),

$$\sum_{\sigma} C_{\mathbf{q}\sigma} b^\dagger C_{\mathbf{p}\sigma}^a = (u_{\mathbf{q}} b_{\mathbf{p}}^a + u_{\mathbf{p}} a_{\mathbf{q}}^b) \gamma_{\mathbf{q}\uparrow} b^\dagger \gamma_{\mathbf{p}\downarrow} a^\dagger \quad (12)$$

plus terms which destroy one or two nonexistent quasi-

particles. The coherence factor<sup>12</sup>

$$L_{\mathbf{p}\mathbf{q}}^2 = (u_{\mathbf{q}} b_{\mathbf{p}}^a + u_{\mathbf{p}} a_{\mathbf{q}}^b)^2 = \frac{1}{2} [1 - (\epsilon_{\mathbf{p}} \epsilon_{\mathbf{q}} - \Delta^2) / E_{\mathbf{p}} E_{\mathbf{q}}], \quad (13)$$

where  $E_{\mathbf{q}} = (\Delta^2 + \epsilon_{\mathbf{q}}^2)^{1/2}$  and  $\epsilon_{\mathbf{q}} = (\hbar^2/2m)(\mathbf{q}^2 - K_F^2)$  may be approximated by  $L_{\mathbf{p}\mathbf{q}}^2 = 2[1 + \Delta^2/E_{\mathbf{q}}E_{\mathbf{p}}]$  with  $\mathbf{p}$  and  $\mathbf{q}$  restricted to magnitudes greater than  $K_F$ .

The phonon-assisted current is then given by applying the golden rule

$$J = (4\pi e/\hbar) \left| \sum_{\text{out}} \sum_{\mathbf{G}\mathbf{p}\mathbf{q}\mathbf{k}\mathbf{s}} A_{\mathbf{k}\mathbf{G}\mathbf{s}} T_{\mathbf{p}} L_{\mathbf{p}\mathbf{q}} \right. \\ \left. \times \langle \Psi_{\text{out}} | \gamma_{\mathbf{q}\uparrow} b^\dagger \gamma_{\mathbf{p}\downarrow} a^\dagger b_{\mathbf{k}\mathbf{s}}^a | \Psi_{\text{in}} \rangle \right|^2 \delta(E_{\mathbf{q}} + E_{\mathbf{p}} - \hbar\omega - e\mathcal{U}) \quad (14)$$

with the condition that  $\mathbf{q}-\mathbf{p}=\mathbf{k}+\mathbf{G}$ . The wave functions in (14) are defined by

$$\Psi_{\text{in}} = |O_N^a, O_N^b, n_{\mathbf{k}\mathbf{s}}^a\rangle, \\ \Psi_{\text{out}} = \gamma_{\mathbf{K}\downarrow} a^\dagger \gamma_{\mathbf{K}'\uparrow} b^\dagger |O_{N-1}^a, O_{N'+1}^b, n_{\mathbf{k}\mathbf{s}}^a - 1\rangle, \quad (15)$$

where  $O_N^a$  represents the ground state of superconductor  $a$  with  $N$  electrons and  $|n_{\mathbf{k}\mathbf{s}}^a\rangle$  is the phonon state vector with  $n_{\mathbf{k}\mathbf{s}}$  phonons in mode  $(\mathbf{k}, s)$ . The phonons on side  $b$  have contributed a factor of 2 to Eq. (14). Using  $\langle n_{\mathbf{k}\mathbf{s}} - 1 | b_{\mathbf{k}\mathbf{s}} | n_{\mathbf{k}\mathbf{s}} \rangle = N_{\mathbf{k}\mathbf{s}} = (e^{\hbar\omega/kT} - 1)^{-1}$  and changing the sum over  $\mathbf{p}$  and  $\mathbf{q}$  to an integral with  $K^2 dK = (Km/\hbar^2) d\epsilon_K \approx (K_F m/\hbar^2) (E_K^2 - \Delta^2)^{-1/2} E_K dE_K$ , we obtain (dropping the explicit indication of the sum over phonon modes  $s$ )

$$J = \frac{8\pi e V^2}{\hbar (2\pi)^6} \left( \frac{K_F m}{\hbar^2} \right)^2 \int d\Omega_{\mathbf{p}} d\Omega_{\mathbf{q}} |A_{\mathbf{k}\mathbf{G}\mathbf{s}} T_{\mathbf{p}}|^2 N_{\mathbf{k}\mathbf{s}} \\ \times \int \frac{E_{\mathbf{p}} E_{\mathbf{q}} + \Delta^2}{E_{\mathbf{p}} E_{\mathbf{q}}} \frac{E_{\mathbf{p}} dE_{\mathbf{q}}}{(E_{\mathbf{p}}^2 - \Delta^2)^{1/2}} \frac{E_{\mathbf{q}} dE_{\mathbf{q}}}{(E_{\mathbf{q}}^2 - \Delta^2)^{1/2}}. \quad (16)$$

We have made the usual assumption that  $A_{\mathbf{k}\mathbf{G}\mathbf{s}}$  and  $T_{\mathbf{p},\mathbf{q}}$  vary only slightly over the range of values of  $E_{\mathbf{p}}$  and  $E_{\mathbf{q}}$  which contributes to  $\mathcal{J}$ , the second double integral in Eq. (16). Integrating over  $E_{\mathbf{q}}$  we obtain

$$\mathcal{J} = \int_{\Delta}^{\hbar\omega + e\mathcal{U} - \Delta} E_{\mathbf{p}} (\hbar\omega + e\mathcal{U} - E_{\mathbf{p}}) (E_{\mathbf{p}}^2 - \Delta^2)^{-1/2} \\ \times [(\hbar\omega + e\mathcal{U} - E_{\mathbf{p}})^2 - \Delta^2]^{-1/2} dE_{\mathbf{p}}. \quad (17)$$

Now make the substitution  $x = E_{\mathbf{p}} - \Delta$  and  $\hbar\omega + e\mathcal{U} - 2\Delta = \delta$  and

$$\mathcal{J} = \int_0^{\delta} [(x+\Delta)(\Delta+\delta-x) + \Delta^2] [x^2 + 2\Delta x]^{-1/2} \\ \times [(\delta-x)^2 + 2\Delta(\delta-x)]^{-1/2} dx. \quad (18)$$

We have assumed that we are at a temperature well below  $T_c$ . Since for lead  $2\Delta = 4.2kT_c$ , we have  $\delta \ll \Delta$ , i.e., larger  $\delta$ 's require phonons of higher frequencies which are not present at these low temperatures. Thus,

<sup>12</sup> Equation (13) follows directly from the definitions of  $u$  and  $v$ :  $u = 2^{-1/2} (1 + \epsilon/E)^{1/2}$ ,  $v = 2^{-1/2} (1 - \epsilon/E)^{1/2}$ .

Eq. (18) simplifies to

$$g = \Delta \int_0^\delta dx x^{-1/2} (\delta - x)^{-1/2} = \pi \Delta. \quad (19)$$

Before doing the integration over angles we simplify  $T^2$  [Eq. (5)]

$$\beta = [2m\hbar^{-2}(V_0 - E_F - E_F \sin^2\theta_q)]^{1/2} \\ \approx [2m\hbar^{-2}(V_0 - E_F)]^{1/2} \\ \times [1 + E_F \sin^2\theta_q / 2(V_0 - E_F)]. \quad (20)$$

Since  $\beta$  with its  $\sin^2\theta_q$  appears exponentially, we may set  $\cos^2\theta_q = 1$  wherever it appears in  $T$ . Thus,

$$T_p^2 \approx 16E_F V_0^{-2} (V_0 - E_F) \exp[-2d\hbar^{-1}(2m(V_0 - E_F))^{1/2}] \\ \times \exp[-E_F d\hbar^{-1} \sin^2\theta_q (2m/(V_0 - E_F))^{1/2}], \quad (21)$$

and

$$J = C \int \int \sin\theta_p \sin\theta_q \\ \times \exp[-E_F d\hbar^{-1} \sin\theta_q (2m/(V_0 - E_F))^{1/2}] \\ \times I^2(\mathbf{k} + \mathbf{G}) [(\mathbf{k} + \mathbf{G}) \cdot \hat{e}_{\mathbf{k}s}]^2 k^{-1} \\ \times [e^{\hbar s k / kT} - 1]^{-1} d\theta_p d\theta_q d\varphi_p d\varphi_q \quad (22)$$

where

$$C = \frac{32eV}{(2\pi)^4} \left( \frac{K_F m}{\hbar^2} \right)^2 \frac{E_F (V_0 - E_F)}{V_0^2} \left( \frac{\Delta}{2M_s N} \right) \\ \times \exp[-2d\hbar^{-1}(2m(V_0 - E_F))] \quad (23)$$

and  $s$ , the speed of sound, appears because we have replaced  $\omega_{\mathbf{k}s}$  appearing in  $N_{\mathbf{k}s}$  and in  $A_{\mathbf{k}G_s}$  by  $ks$ .

#### IV. NORMAL PROCESSES

We now look at normal processes ( $\mathbf{G} = 0$ ). The electron-phonon matrix element  $I(\mathbf{k})$  is a slowly varying function of  $\mathbf{k}$  and for low temperatures may be replaced by  $I(0) = \frac{2}{3}E_F$  according to the Bardeen self-consistent calculation.<sup>13</sup> Now  $\mathbf{k}$  depends on the angle between  $\mathbf{p}$  and  $\mathbf{q}$  so we change our variables of integration from  $\Omega_p$  and  $\Omega_q$  to  $\Omega_{p-q}$  and  $\Omega_q$ . The integrations over  $\phi_q$  and  $\phi_{q-p}$  yield a factor  $(2\pi)^2$ . Substituting  $\mu = \sin\theta_q$  and  $d\mu = d\theta_q$  (remember  $\cos\theta_q \approx 1$ ) the integral over  $\theta_q$  becomes

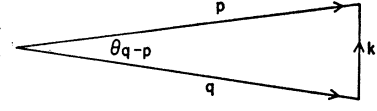
$$\int_0^\infty \exp[-\mu E_F d\hbar^{-1} (2m/(V_0 - E_F))^{1/2}] \mu d\mu \\ = \frac{1}{2} \hbar ((V_0 - E_F)/2m)^{1/2} (E_F d)^{-1}.$$

To do the integral over  $\theta_{q-p}$  we assume pure longitudinal ( $\mathbf{k} \cdot \hat{e}_{\mathbf{k}s} = k$ ) and transverse ( $\mathbf{k} \cdot \hat{e}_{\mathbf{k}s} = 0$ ) modes and use  $\frac{1}{2}k = K_F \sin\frac{1}{2}\theta_{q-p}$  (see Fig. 3) which yields

$$\sin\theta_{q-p} d\theta_{q-p} = k dk / K_F^2. \quad (24)$$

<sup>13</sup> J. Bardeen, Phys. Rev. **52**, 688 (1937).

FIG. 3. Incoming ( $\mathbf{q}$ ) and outgoing ( $\mathbf{p}$ ) electron wave vectors and phonon wave vector ( $\mathbf{k}$ ) for normal process.



Thus,

$$J = (2/9)E_F C (2\pi)^2 \hbar ((V_0 - E_F)/2m)^{1/2} d^{-1} K_F^{-2} \\ \times \int_{(2\Delta - e\mathcal{U})/\hbar s}^\infty (e^{\hbar k s / kT} - 1)^{-1} k^2 dk, \quad (25)$$

where the lower limit of integration arises from energy conservation,  $\hbar s k \geq 2\Delta - e\mathcal{U}$ . So that we finally obtain

$$J = J_0 \frac{L_3 E_F m}{9\pi \hbar M_s N} \left( \frac{kT}{\hbar s} \right)^3 D_2 \left( \frac{2\Delta - e\mathcal{U}}{kT} \right), \quad (26)$$

where

$$D_n(x) = \int_x^\infty (e^x - 1)^{-1} x^n dx$$

is the tabulated Debye integral,

$$J_0 = 2e\Delta L_1^2 E_F (V_0 - E_F)^{3/2} (2m)^{1/2} (\pi d \hbar^2 V_0^2)^{-1} \\ \times \exp[-2d\hbar^{-1}(2m(V_0 - E_F))^{1/2}]$$

is the discontinuity in the current<sup>2</sup> at  $e\mathcal{U} = 2\Delta$ , and the dimensions of the superconductors are  $L_1 \times L_1 \times L_3$ . The current depends linearly on the thickness of the superconductor. This is because we have assumed the electron wave functions are plane waves extending over the entire crystal. In actual fact the wave functions are plane wave like only over small distances  $\lambda$ , where  $\lambda$  may actually be  $L_3$  or  $\lambda$  may represent the mean free path for a normal electron—whichever is shortest. For  $\lambda = 170 \text{ \AA}$ , about  $\frac{1}{3}$  of the thickness of the superconducting films,<sup>14</sup> we find  $J = 1.7 \times 10^{-7} J_0 T^3 D_2((2\Delta - e\mathcal{U})/kT)$ , about a factor of  $10^3$  too small to account for the observed current.

#### V. UMKLAPP PROCESSES

The small value for  $J$  obtained from normal electron-phonon scattering is due to the factor  $(\mathbf{k} + \mathbf{G})^2$  appearing in Eq. (22). For  $\mathbf{G} \neq 0$  the scattering will be much stronger. We therefore examine the umklapp processes. Since the vectors  $\mathbf{G}$  are fixed in the reciprocal lattice we must also measure  $\theta_p$ ,  $\theta_q$ ,  $\phi_p$ , and  $\phi_q$  with respect to the reciprocal lattice and not with respect to the barrier normal as before. Since  $\mathbf{k}$ , the phonon wave vector, is very small, the electron wave vectors  $\mathbf{p}$  and  $\mathbf{q}$  are constrained to lie in directions such that  $\mathbf{q} - \mathbf{p} \approx \mathbf{G}$ . (See Fig. 4.) The tunneling matrix element depends exponentially on the angle the barrier normal makes with  $\mathbf{q}$  or equivalently with the reciprocal lattice vector  $\mathbf{G}$ . It is believed that the lead film consists of tiny crystallites randomly oriented.<sup>14</sup> Therefore, in addition to the integrations in Eq. (22)

<sup>14</sup> B. N. Taylor and E. Burstein (private communication).

we average over the angle  $\mathbf{N}$  makes with  $\mathbf{q}$ . The integration over  $\theta_{\mathbf{N}\mathbf{q}}$  proceeds exactly as the integration over  $\theta_{\mathbf{q}}$  did for the normal case and we obtain<sup>15</sup>

$$J = \frac{1}{4} C \hbar ((V_0 - E_F)/2m)^{1/2} d^{-1} E_F^{-1} \int \int d\Omega_p d\Omega_q I^2(\mathbf{k} + \mathbf{G}) \times [(\mathbf{k} + \mathbf{G}) \cdot \hat{\mathbf{e}}_{\mathbf{k}s}]^2 k^{-1} (e^{\hbar s k/kT} - 1)^{-1}, \quad (27)$$

where a sum over the 14 reciprocal lattice vectors smaller than  $2K_F$  is to be understood. (Since  $\mathbf{k}$  is very small, momentum cannot be conserved for  $G > 2K_F$ ). We change our variables of integration from  $\Omega_q$  and  $\Omega_p$  to  $\Omega_{(\mathbf{q}+\mathbf{p})/2}$  and  $\Omega_{\mathbf{q}-\mathbf{p}}$ , take  $\mathbf{k} + \mathbf{G} \approx \mathbf{G}$ , set  $k = [(\mathbf{G} - \mathbf{K})^2 + G^2 \psi^2]^{1/2}$  (see Fig. 4, Ref. 16) and integrate over  $\phi_{(\mathbf{q}+\mathbf{p})/2}$  and  $\phi_{\mathbf{q}-\mathbf{p}}$  obtaining  $(2\pi)^2$  to get

$$J = C \pi^2 \hbar I^2(\mathbf{G}) (\mathbf{G} \cdot \hat{\mathbf{e}}_{\mathbf{k}s})^2 ((V_0 - E_F)/2m)^{1/2} d^{-1} E_F^{-1} \times \int \int \sin \theta_{(\mathbf{p}+\mathbf{q})/2} \sin \theta_{\mathbf{q}-\mathbf{p}} [(\mathbf{K} - \mathbf{G})^2 + G^2 \psi^2]^{-1/2} \times \{ \exp[(\hbar s/kT)((\mathbf{K} - \mathbf{G})^2 + G^2 \psi^2)^{1/2}] - 1 \}^{-1} \times d\theta_{(\mathbf{p}+\mathbf{q})/2} d\theta_{\mathbf{q}-\mathbf{p}}. \quad (28)$$

If we take  $\mathbf{G}$  to be the polar axis of our spherical coordinate system,  $\theta_{(\mathbf{q}+\mathbf{p})/2} = \rho$ . We may equally well integrate over  $\psi$ , the complement of  $\rho$  (see Fig. 4). Corresponding to Eq. (24), we have

$$(G/K_F^2) dK \approx (K/K_F^2) dK = \sin \theta_{\mathbf{q}-\mathbf{p}} d\theta_{\mathbf{q}-\mathbf{p}}, \quad (29)$$

also since we will get contributions to the integral only for small  $\psi$ ,  $\sin \psi \approx \psi$  and therefore

$$\sin \theta_{(\mathbf{q}+\mathbf{p})/2} \sin \theta_{\mathbf{q}-\mathbf{p}} d\theta_{(\mathbf{q}+\mathbf{p})/2} d\theta_{\mathbf{q}-\mathbf{p}} \rightarrow (G/K_F^2) \psi dK d\psi.$$

We now make the following substitutions:  $(\hbar s/kT)(\mathbf{K} - \mathbf{G}) = r \sin \omega$ ,  $(\hbar s/kT)G\psi = r \cos \omega$ , and hence,  $(\hbar s/kT)^2 G dK d\psi = r dr d\omega$  yielding

$$J = C \pi^2 \hbar I^2(\mathbf{G}) (\mathbf{G} \cdot \hat{\mathbf{e}}_{\mathbf{k}s})^2 (kT/\hbar s)^2 \times ((V_0 - E_F)/2m)^{1/2} (GK_F^2 E_F d)^{-1} \times \int_{-\pi/2}^{\pi/2} \int_{(2\Delta - e\mathcal{U})/kT}^{\infty} r \cos \omega [e^r - 1]^{-1} dr d\omega \quad (30)$$

or

$$J = J_0 \frac{L m^2 I^2(\mathbf{G}) (\mathbf{G} \cdot \hat{\mathbf{e}}_{\mathbf{k}s})^2 (kT)^2}{\pi \hbar^3 M s N G K_F^2} D_1 \left( \frac{2\Delta - e\mathcal{U}}{kT} \right). \quad (31)$$

We must now sum Eq. (31) over the fourteen  $\mathbf{G}$ 's smaller than  $2K_F$ . There are eight (1,1,1)  $\mathbf{G}$  vectors of length  $2.20 \times 10^8 \text{ cm}^{-1}$  and six (2,0,0) vectors of length  $2.54 \times 10^8 \text{ cm}^{-1}$ ;  $K_F = 1.57 \times 10^8 \text{ cm}^{-1}$ . We may either calculate  $I(\mathbf{G})$  from the Pines<sup>6</sup> formula or use the average value estimated by Rothwarf and Cohen<sup>7</sup> from high-temperature resistivity measurements. They obtain  $I^2 \bar{G}^2/N^2 = 6.1 \times 10^{-52} \text{ erg}^2 \text{ cm}^4$  while an average of the

<sup>15</sup> The extra factor of  $\frac{1}{2}$  comes from  $\int_0^\pi \sin \theta_{\mathbf{N}\mathbf{q}} d\theta_{\mathbf{N}\mathbf{q}} = 2$ .

<sup>16</sup>  $\mathbf{K} = \mathbf{q} - \mathbf{p}$  so that  $\mathbf{K} = \mathbf{k} + \mathbf{G}$ . Therefore,  $k^2 = K^2 + G^2 - 2KG \cos \psi$ , where  $\psi$  is the angle between  $\mathbf{k}$  and  $\mathbf{G}$ . Since  $\mathbf{k}$  is very small so is  $\psi$  and expanding the cosine we obtain  $k^2 = (K - G)^2 + KG\psi^2 \approx (K - G)^2 + G^2 \psi^2$ .

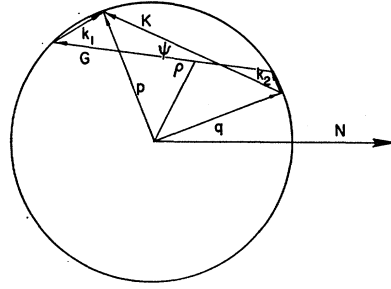


FIG. 4. Vectors and angles involved in umklapp scattering.  $\mathbf{G}$ , a reciprocal lattice vector is taken as the polar axis of the coordinate system;  $\mathbf{N}$  is the normal vector to the oxide barrier;  $\mathbf{K}$  is the difference between the incoming electron wave vector  $\mathbf{q}$  and the outgoing electron wave vector  $\mathbf{p}$ ;  $\rho$  is the polar angle  $\mathbf{p} + \mathbf{q}$  makes and  $\psi$  is its complement; the phonon wave vector  $\mathbf{k} = \mathbf{K} - \mathbf{G}$  is the sum of the two pieces labeled  $\mathbf{k}_1$  and  $\mathbf{k}_2$ .

Pines formula over the fourteen  $\mathbf{G}$ 's yields  $8.7 \times 10^{-52} \text{ erg}^2 \text{ cm}^4$ . The transverse and longitudinal sound velocities in lead are  $s_L = 2.35 \times 10^5 \text{ cm/sec}$  and  $s_T = 1.27 \times 10^5 \text{ cm/sec}$ ; since  $J \sim s^{-3}$  we will neglect the longitudinal modes and take  $\sum_{\mathbf{G},s} (\mathbf{G} \cdot \hat{\mathbf{e}}_{\mathbf{k}s})^2 = \frac{2}{3} \times 14 \bar{G}^2$ . Using  $N = 3.32 \times 10^{22} \text{ cm}^{-3}$ ,  $M = 3.83 \times 10^5 m_e$ ,  $L = 170 \text{ \AA}$ ,  $\bar{G} = 2.35 \times 10^8 \text{ cm}^{-1}$  and  $I^2 = 7 \times 10^{-52} N^2 \bar{G}^{-2} \text{ erg}^2 \text{ cm}^4$  we obtain

$$J = 3.44 \times 10^{-4} J_0 T^2 D_1((2\Delta - e\mathcal{U})/kT). \quad (32)$$

## VI. COMPARISON WITH EXPERIMENT

For lead  $2\Delta(0) = 4.2kT_c$  and  $T_c = 7.2^\circ \text{K}$ , so  $J = 3.4 \times 10^{-4} J_0(T) T^2$

$$D_1 \left( \frac{30.24\Delta(T)}{T\Delta(0)} \left( 1 - \frac{e\mathcal{U}}{2\Delta(T)} \right) \right);$$

we use the BCS<sup>17</sup>  $\Delta(T)/\Delta(0)$ . In Fig. 5 we plot  $J$  against  $T$  for  $e\mathcal{U}/2\Delta(T) = 0.8$ . For intermediate temperatures the theoretical and experimental curves agree very well. At higher temperatures the theoretical current is too small. This is to be expected because of the simplifications, valid only at low temperatures, which we made in going from Eq. (18) to Eq. (19). At low temperatures the experimental current is seen to become temperature-independent. This must be due either to a small "leakage" current like the one observed for other superconductors<sup>1</sup> or to the error in subtracting the other tunneling processes from the total observed current. In any event the true temperature-dependent part of the current must continue to decrease exponentially with temperature. In Fig. 6 we plot  $J$  against  $e\mathcal{U}/2\Delta$  for  $T = 2.86^\circ$ . Theory and experiment are in excellent agreement for  $e\mathcal{U}/2\Delta \leq 0.95$ . It is believed<sup>14</sup> that due to the difference in thermal expansion between the lead film and its glass substrate, the lead is subjected to large stresses. Even if the stress were uniform, this would result in a different  $\Delta$  for each crystalite depending on

<sup>17</sup> J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

its orientation. Thus, when  $eU/2\Delta > 0.95$ , there may actually be some crystalites for which  $eU/2\Delta \sim 1$ . The normal single-particle current tunneling through these crystalites would easily account for the excess experimental current. Hence, we conclude that over those ranges of  $U$  and  $T$  for which both theory and experiment can be expected to be valid, their agreement is excellent.

We must finally look into the possibility of there being additional processes which could contribute to the temperature-dependent current and spoil the agreement between the theory and experiment. One such process could be tunneling assisted by phonons in the oxide layer. If we assume the same electron-phonon coupling in the oxide as in the superconductor this is down by a factor  $L_{ox}/2\lambda \approx 20/340$  from the phonon processes in the superconductors. There are obviously other factors involved as well which are difficult to estimate theoretically because of a lack of knowledge about the phonon modes in the oxide layer. However, additional experiments<sup>14</sup> on samples with Pb-oxide layers (the original samples had Al-oxide layers), yielded identical currents which seem to confirm the negligibility of the oxide phonons.

Another group of processes is the many phonon assisted tunneling. These require integrations over intermediate states which in general we were unable to perform. However, we were able to make the calculation for second-order umklapp processes which should be the most important of the many phonon processes. This is because the energy of the intermediate states is proportional to  $(\mathbf{G} + \mathbf{k}_1 + \mathbf{p}_1)^2 - K_F^2 \approx G^2$  when averaged

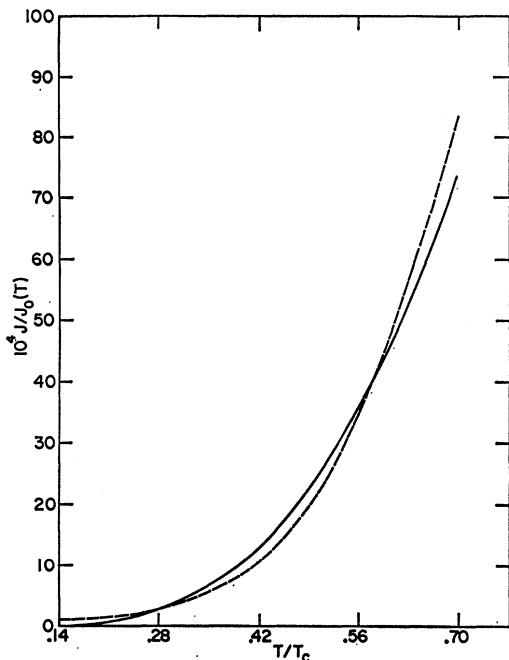


FIG. 5. Theoretical (solid line) and experimental (dashed line) plots of current in units of  $J_0$ , the discontinuity in current at  $eU = 2\Delta$ , versus temperature. The voltage is fixed at  $eU/2\Delta = 0.8$ .

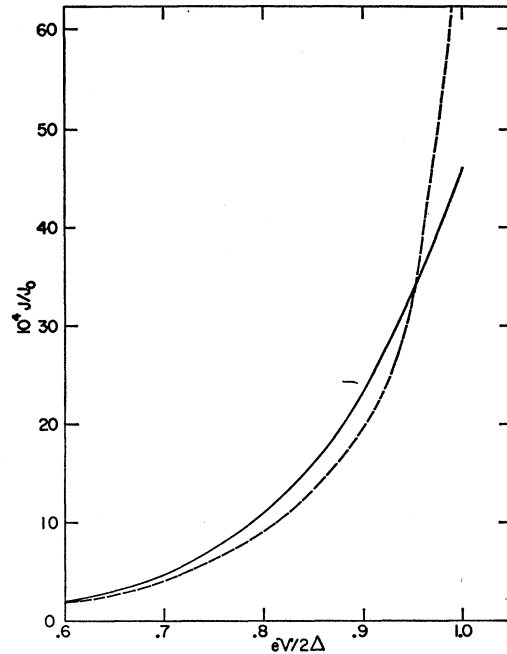


FIG. 6. Theoretical (solid line) and experimental (dashed line) plots of current versus voltage for  $T = 2.86^\circ\text{K}$ .

over the angle  $\mathbf{G}$  makes with  $\mathbf{N}$  (and hence  $\mathbf{p}_1$ ) where  $\mathbf{p}_1$  represents the wave vector of the incoming electron and  $\mathbf{k}_1$  the wave vector of the first phonon. With this simplification the energy denominator causes no trouble when  $\mathbf{k}_1$  or  $\mathbf{p}_1$  are integrated over. Because energy is not conserved in the intermediate state, all  $\mathbf{G}$ 's (not only those for which  $G \leq 2K_F$ ) must be summed over until  $G^2 I^2(\mathbf{G}) \sim [G/(G^2 + K_s^2)]^2$  is negligible<sup>6</sup> where  $K_s^2 = 3.73 \times 10^{16} \text{ cm}^{-2}$  is a screening factor. After performing integrals only slightly more tedious than those involved in the one-phonon calculations we obtain

$$J = 7.9 \times 10^{-8} J_0 T^3 F(2\Delta - eU/2kT), \quad (33)$$

where

$$F(x) = \int_x^\infty \frac{t[t - \ln(e^t - 1)]}{e^t - 1} dt + \int_0^x \frac{t[2x - t - \ln(e^{2x-t} - 1)]}{e^t - 1} dt. \quad (34)$$

Thus the two-phonon processes are down by more than three orders of magnitude from the one-phonon umklapp processes.

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