Phonon-Assisted Tunneling in Superconductors*

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In an earlier publication, two of us[1] reported the existence of polarity-independent excess currents in electron tunneling between superconductors at voltages $eV < \Delta_a(T) + \Delta_b(T)$ [eV is the electronic charge times the applied voltage and $\Delta(T)$ is one half the energy gap of the superconductor at the operating temperature T]. One type of excess current, characterized by a sharp temperature-independent jump at $eV = \Delta(T)$, has been attributed to a mechanism suggested by Schrieffer[2] involving double-particle tunneling (DPT) via pair dissociation or recombination. After subtracting the normal single-particle tunneling (SPT) current and where applicable, the DPT current, an additional excess current was found for Pb–Pb junctions which decreased rapidly with decreasing temperature and which increased approximately exponentially with voltage. It was suggested that this type of excess current is associated with thermally assisted single-particle tunneling processes involving phonons, photons, or electrons. In this paper we present further data on the strongly temperature-dependent excess currents in Pb–Pb junctions and a theoretical calculation of thermal phonon-assisted tunneling processes.

Approximately 20-Å-thick barrier layers for the Pb–Pb junctions were prepared by either completely oxidizing an evaporated Al film deposited over the first Pb layer or by oxidizing the surface of the Pb layer itself. Both methods yield samples with similar current–voltage (I–V) characteristics. Such characteristics for a typical Pb–Pb junction are shown in Fig. 1 for two different temperatures; 4.2° and 2.2°K. At 2.2°K the DPT current is clearly discernable, whereas at the higher temperature the DPT current is completely masked by the normal SPT current. The additional excess current is obtained by subtracting the theoretical SPT and DPT currents from the experimental curves. A plot of the normalized excess current $J/J_0(0)$ [the ratio of the actual excess current to the current jump at $eV = 2\Delta(0)$] vs reciprocal temperature for several values of $eV/2\Delta(T)$ is shown in Fig. 2. It should be pointed out that this type of excess current was not readily apparent in the I–V curves of any other junctions studied—i.e., Sn–Sn, Al–Al, Sn–Pb, and Sn–Tl. This is attributed in part to the strong coupling[3] nature of Pb, the presence of large temperature-independent excess currents in some of the other junctions, and the difficulty of accurately subtracting theoretical curves from experimental curves, especially in a region of large negative resistance.

Because the thermal phonon-assisted SPT process is the one most likely to make the major contribution, it is this mechanism which we have calculated and compared to experiment. In phonon-assisted SPT, a phonon is absorbed by a ground-state pair on one side of the oxide barrier creating two quasiparticles, one on each side of the barrier; this results in the transfer of a single electron from one superconductor to the other.

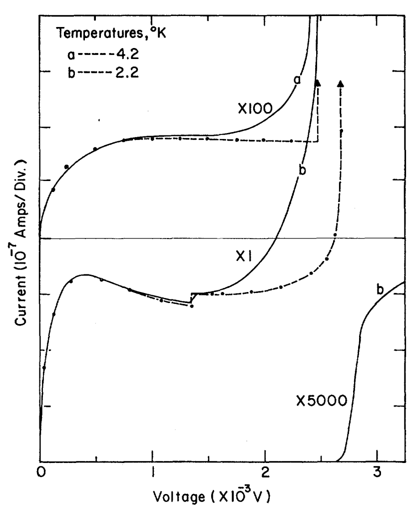

FIG. 1. Typical experimental I–V curves (solid lines) for Pb–Pb junctions. The dashed lines are theoretical SPT plus DPT I–V curves.

A many-body Hamiltonian for the superconducting junction may be derived[4] using a basis set consisting of the one-electron eigenfunctions ψ_l and ψ_r, left- and right-hand functions with a long tail of

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[1] B. N. Taylor and E. Burstein, Phys. Rev. Letters **10**, 14 (1963).

[2] J. R. Schrieffer and J. W. Wilkins, Phys. Rev. Letters **10**, 17 (1963).

[3] The calculated phonon-assisted tunneling current in Al–Al junctions is about 1/20 that in Pb–Pb junctions.

[4] L. Kleinman (to be published).

magnitude T on the "wrong" side of the oxide barrier[5]:

$$H = H_l + H_r + H_{ll}^{\rm elph} + H_{rr}^{\rm elph} + H_{lr}^{\rm elph} + H_{rl}^{\rm elph} + H_{\rm int}\,. \qquad (1)$$

The term of interest that destroys an l electron, creates an r electron and destroys a phonon of wave vector $\mathbf{k}$, and polarization s is

$$H_{lr}^{\rm elph} = \sum_{\mathbf{pqkG}s\sigma} A_{\mathbf{kG}s}[T_{\mathbf{q}} b^l_{\mathbf{k}s} + T_{\mathbf{p}} b^r_{\mathbf{k}s}]\delta(\mathbf{q}-\mathbf{p}-\mathbf{k}-\mathbf{G})\, c^{r\dagger}_{\mathbf{q}\sigma} c^{l}_{\mathbf{p}\sigma}\,, \qquad (2)$$
$$+ \text{ herm. conj.}$$

where $A_{\mathbf{kG}s}$ represents the usual factors[6] in the electron–phonon Hamiltonian and $\mathbf{G}$ is a reciprocal lattice vector. We take account in Eq. (2) only of phonons in the two superconductors; the fact that the experimental data is independent of which oxide is used indicates that the oxide phonons play a negligible role.

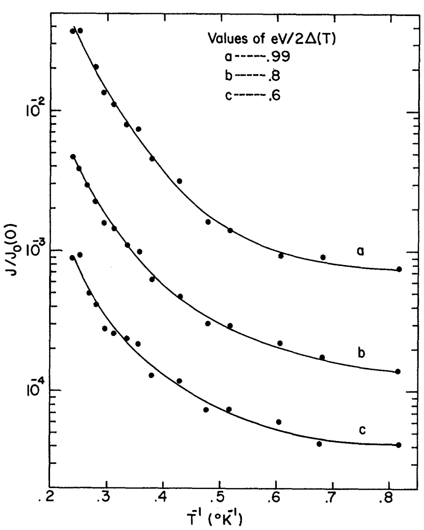

FIG. 2. Normalized excess current $J/J_0(0)$ vs reciprocal temperature for several values of $eV/2\Delta(T)$.

In order to eliminate $H_{\rm int}$ from Eq. (1), the electron creation and destruction operators are written in terms of the Bogoliubov quasiparticle operators[7] resulting in

$$\sum\nolimits_\sigma c^{r\dagger}_{\mathbf{q}\sigma} c^{l}_{\mathbf{p}\sigma} = (u^r_{\mathbf{q}} v^l_{\mathbf{p}} + u^l_{\mathbf{p}} v^r_{\mathbf{q}})\gamma^{r\dagger}_{\mathbf{q}\uparrow}\gamma^{l\dagger}_{-\mathbf{p}\downarrow} \qquad (3)$$

plus terms that destroy quasiparticles. (We assume the temperature low enough for the thermally excited quasiparticles to be negligible.) The determination of the current using the golden rule and Eqs. (2) and (3) is straightforward but the integrations over d^3p, d^3q, d^3k are rather tedious and will be presented elsewhere.[4] We have performed the calculation for normal ($\mathbf{G} = 0$) and umklapp ($\mathbf{G} \neq 0$) processes and in addition have calculated the current due to two-phonon processes. Only the one-phonon (umklapp processes) is appreciable and the result is

$$J = J_0(T)\,\frac{Lm^2I^2(\mathbf{G})[\mathbf{G}\cdot\hat{e}_s]^2}{\Pi\hbar^3 MsNGK_F^2}\left(\frac{kT}{\hbar s}\right)^2 \times D_1\left(\frac{2\Delta(T)-eV}{kT}\right), \qquad (4)$$

where a sum over the 14 reciprocal lattice vectors $G < 2K_F = 3.14 \times 10^8$ cm^{-1} and over the polarization vectors $\hat{e}_s$ is implied. $J_0(T)$ is the discontinuity in the current at $eV = 2\Delta(T)$ and

$$D_1(x) = \int_x^\infty (e^x - 1)^{-1} x\,dx\,.$$

In deriving Eq. (4) we have used the polycrystalline nature of the superconducting films to justify an average over crystal orientation.[8]

The transverse and longitudinal sound velocities in lead are $S_L = 2.35 \times 10^5$ cm/sec and $S_T = 1.27 \times 10^5$ cm/sec. Since $J \sim s^{-3}$ we neglect the longitudinal modes and take $\Sigma_{\mathbf{G},s}(\mathbf{G}\cdot\hat{e}_{\mathbf{k}s})^2 = \frac{2}{3} \times 14\overline{G}^2$. The electron–phonon matrix element $I(\mathbf{G})$ can be calculated from the Pines[9] formula or alternatively, the average value estimated by Rothwarf and Cohen[10] from high-temperature restivity data can be used. They obtained $I^2\overline{G}^2/N^2 = 6.1 \times 10^{-52}$ ergs^{2}cm^4 while an average of the Pines formula over the fourteen G's yields 8.7×10^{-52} ergs^{2}cm^4. N is the number density of lead atoms, 3.32×10^{22} cm^{-3}, and M their mass, $3.83 \times 10^5 m_e$. L is the thickness of the superconductors (assumed to be identical); however, it may not exceed the distance over which a basis function may be approximated by a plane wave. Thus L probably represents the mean free path for normal electrons. On taking $L = 170$ Å, $\overline{G} = 2.35 \times 10^8$cm^{-1}, and $I = 7 \times 10^{-52} N^2\overline{G}^2$ ergs^{2}cm^4, we obtain

$$J = 3.44 \times 10^{-4} J_0(T) T^2 D_1\{[2\Delta(T) - eV]/kT\}\,. \; (5)$$

For lead $2\Delta(0) = 4.2kT_c$ and $T_c = 7.2$°K so $J = 3.4 \times 10^{-4} J_0(T) T^2\ D_1\{[30.24\Delta(T)T\Delta(0)]$

[5] It is interesting to note that H_T, the term responsible for the usual single particle tunneling, does not appear in H. This is because our basis functions with their long tails on the wrong side of the barrier have already accounted for this.

[6] J. M. Ziman, *Electrons and Phonons* (Oxford University Press, London, 1962).

[7] S. T. Beliaev, *The Many Body Problem* (John Wiley & Sons, Inc., New York, 1959), p. 360 ff.

[8] At low temperatures umklapp tunneling processes can occur only when $\mathbf{N}K_F - \mathbf{G}$ lies very near the Fermi surface ($\mathbf{N}$ is a unit vector normal to the oxide barrier).

[9] D. Pines, Phys. Rev. **109**, 280 (1958).

[10] A. Rothwarf and M. Cohen, Phys. Rev. **130**, 1401 (1963).

$[1 - eV/2\Delta(T)]\}$. In Fig. 3 we plot J against $eV/2\Delta(T)$ for $T = 2.86°$. It can be seen that theory and experiment are in excellent agreement for $eV/2\Delta(T) < 0.95$. We believe that due to the difference in thermal expansion between the lead film and

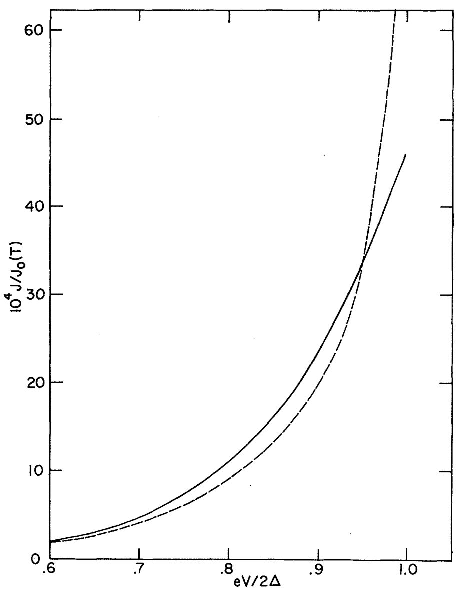

FIG. 3. Theoretical (solid line) and experimental (dashed line) plots of normalized current, $J/J_0(T)$ vs voltage for $T = 2.86°K$.

its glass substrate, the lead is subjected to large stresses. Even if the stress were uniform, this would result in a different Δ for each crystalite, depending on its orientation. Thus, when $eV/2\Delta(T) > 0.95$ there may be some crystalites for which $eV/2\Delta \sim 1$. Normal SPT currents arising from such crystalites could easily account for the excess experimental current.

In Fig. 4 we plot $J/J_0(T)$ against T for $eV/2\Delta(T) = 0.8$. For intermediate temperatures the theoretical and experimental curves agree well. At higher temperatures the theoretical current is too small. This we believe is due to approximations, valid only at low temperatures, which we were required to make in our derivation of Eq. (4). At low temperatures the experimental current is seen to become temperature-independent. This may be due to the error in subtracting the other tunneling processes from the total observed current when the temperature-dependent part is so small, or it may be a real temperature-independent current similar to, though smaller than, that observed for other junctions.[1] The true tem-

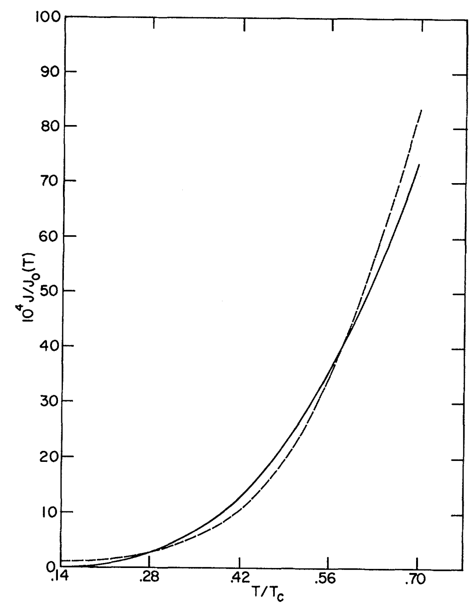

FIG. 4. Theoretical (solid line) and experimental (dashed line) plots of normalized current $J/J_0(T)$ vs reduced temperature for $eV/2\Delta(T) = 0.8$.

perature-dependent current must fall exponentially as the theoretical curve does at low temperatures. Hence, we conclude that over those ranges of V and T for which both theory and experiment are expected to be valid, their agreement is excellent.