MULTIPHONON EFFECTS IN TUNNELLING BETWEEN METALS AND SUPERCONDUCTORS

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From electron tunnelling experiments Giaever, Hart, and Megerle¹ have shown that the dI/dV - Vcharacteristic for superconducting lead has two small steps at biases of approximately $2E_{\varphi}$ and $4E_g$, E_g being the full energy gap. It is our purpose to report further structure in the tunnelling characteristic; specifically, the positions of at least seven steps have been determined, indicating corresponding structure in the density-ofstates distribution. Some limited structure has been predicted recently from solutions of the integral gap equation by Swihart² and by Culler, Fried, Huff, and Schrieffer.³ While the latter solution is exact, it rests on the assumption of a simple Debye phonon spectrum. By invoking an Einstein distribution, Morel and Anderson⁴ have commented that harmonic structure should be expected but we will show that the observed structure is too extensive to be explained by this approach alone.

The extensive structure in the tunnelling characteristics was revealed with the aid of equipment previously used by Chynoweth, Logan, and Thomas.⁵ It displays automatically on an X - Y re-



FIG. 1. First and second differentiation of the current through an Al-Al₂O₃-superconducting Pb sandwich as a function of bias. (Because of the instrumentation involved, the second derivative trace appears upside down.)

corder both dI/dV and d^2I/dV^2 as a function of the dc voltage across a superconducting lead-insulatornormal metal sandwich. Figure 1 shows such traces, considerably reduced in size, for the case of an evaporated Al-Al₂O₃-Pb sandwich at 1.6°K. The dI/dV plot exhibits several humps and up to four of these can be seen on the original traces. The positions of these humps are more easily taken as peaks from the d^2I/dV^2 plot, which also resolves several more at higher biases. The peaks can also be seen near 4.2°K, and similar structure is observed when bulk polycrystalline lead is used.

We have plotted in Fig. 2 the locations of successive peaks, the scatter being due to measurements both at different times and on a number of different samples. It can be seen that the peak positions are given empirically by

$$E_n = \Delta' + n\theta, \tag{1}$$

with the intercept $\Delta' = (1.4 \pm 0.3) \times 10^{-3}$ eV which is



FIG. 2. Locations of peaks in the tunnelling characteristic for $Al-Al_2O_3$ -Pb sandwiches.

in satisfactory agreement with the established value for the half-gap, Δ , 1.34×10^{-3} eV. The slope gave $\theta = 3.7 \times 10^{-3}$ eV and it was found that θ did not change with temperature from 1.6° K to above 4.2° K. According to Brockhouse et al.⁶ the bulk of the transverse acoustic phonon frequencies fall in the range $(4.1 \pm 0.8) \times 10^{-3}$ eV. It therefore seems reasonable to identify θ with the transverse acoustic phonon of lead. The longitudinal acoustic energies are of order 8×10^{-3} eV but there is apparently no repeated structure in the derivative plots which can be attributed to these phonons. This implies that the electron-phonon deformation coupling constants, α^2 , are larger for TA than LA phonons.

The experimental values for the relative magnitudes of the peaks are given in the first row of Table I. These values are fitted rather well by $S_1 = S_2 = 1$, $S_n (n \ge 2) = C^{n-2}$ where C = 0.38 (second row Table I). This value of C is suggestively close to the coupling constant Δ/θ , though it must be emphasized that the observed magnitudes have not been adjusted to include the effects of changes in the peak shapes.

A possible dynamical explanation for the structure is that during tunnelling the carriers emit phonons. Kane⁷ has pointed out, however, that in this case the phonons should be characteristic of the insulator rather than the lead. As the structure is unchanged with different insulators, we conclude that the peaks must be associated with intrinsic properties of the quasi-particles in the superconductor and must result from a proper solution of the gap equation.

Giaever, Hart, and Megerle explain the steps they observed in terms of the energy dependence of the gap parameter $\Delta(E_k)$, where the thermodynamic quasi-particle energy E_k is given in terms of the normal particle energy ϵ_k measured from the Færmi energy by

$$E_k = \left[\epsilon_k^2 + \Delta_k^2\right]^{1/2}.$$
 (2)

Swihart has discussed the determination of Δ_k in an isotropic superconductor from the gap equation (at T = 0)

$$\Delta_{k} = \sum_{q} \alpha^{2} \frac{\Delta_{k+q}}{E_{k+q}} V_{k,k+q}.$$
 (3)

He has used the Frohlich-Bardeen expression for the kernel,

$$V_{k,k+q} = \frac{\hbar \omega_q}{-(\hbar \omega_q)^2 + (\epsilon_k - \epsilon_{k+q})^2}, \qquad (4)$$

which describes electron-phonon scattering in the normal metal.

A more recent exact determination of Δ_k has been made by Culler <u>et al.</u> using the Eliashberg⁸ kernel

$$V_{k,k+q} = \frac{(E_{k+q} + \hbar\omega_q)}{(E_{k+q} + \hbar\omega_q)^2 - E_k^2}.$$
 (5)

Their results are similar to those of Swihart in that the main structure in the resulting density of states consists of two humps at $k\theta_D$ and $2k\theta_D$ (θ_D being the Debye temperature). Their use of a Debye phonon spectrum might be expected to smear out the more closely spaced harmonic structure we observe.

A further approximate solution using the Eliashberg kernel and assuming an Einstein phonon spec trum has been made by Morel and Anderson. They note that harmonic structure should occur at 2θ , 3θ , 4θ , etc., θ being the phonon energy chosen, but this does not appear in their solution as it is regarded as a small correction in the weak-coupling limit and is neglected. If the harmonic structure is retained, we can obtain an upper bound on the magnitudes of successive peaks using the following arguments. The gap equation may be solved qualitatively by taking advantage of isotropy to

Table I. Observed and predicted peak strengths in the tunnelling characteristics.

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Peak number	S ₀	S ₁	S ₂	S ₃	<i>S</i> ₄	S ₅	Se	s ₇
Observed peak strength	>25	1	0.93	0.37	0.17	0.07	0.02	0.01
Power law dependence			1	0.38	0.14	0.05	0.02	0.008
Calculated one-phonon peak strength	1	1	0.38	0.072	0.009	0.0009	0.00007	
$C = E_g/\theta = 0.38$								

transform to integration over $E_{k+q} = E'$. In the density of states given by Bardeen, Cooper, and Schrieffer⁹ there is an $x^{-\nu_2}$ singularity for $E' = \Delta(0) = E_g/2$. Invoking the usual extremal arguments which apply when integrals contain such singularities,¹⁰ we replace E_{k+q} in the neighborhood of the singularity by $E_g/2$. Then for $E_k = E_g/2 + \theta$ the kernel goes through a resonance and $\Delta(E_k)$ changes sign from $\Delta(0)$. This approximation actually overestimates the magnitude of the resonance and hence the peak strengths.

The effect of this resonance is to flatten E_k vs ϵ_k as Δ goes through 0 in the neighborhood of $E_g/2 + \theta$, which means a corresponding peak in the density of states at this point. We can now repeat the argument at $E_g/2 + 2\theta$; we find the peak in the density of states at $E_g/2 + \theta$ again switches the sign of $\Delta(E_k)$. By continuing in this manner, switching of $\Delta(E_k)$ accompanied by peaks in the density of states at $E_g/2 + n\theta$ is found to occur.

Now extend the qualitative solution to give an upper bound on peak intensities. According to (3) the pulse from each peak *n* on the peak *n* +1 is proportional to the strength of peak *n* and to Δ/E_n . This yields a series of linear equations for peak strengths; here $C = E_g/2\theta$ is a coupling parameter, and $E_{\rho}/2$ is neglected compared with θ :

$$S_1 = 1, S_2 = CS_1, S_3 = CS_2/2 \cdots,$$
 (6)

which can be solved to give $S_{n+1} = C^n / n!$.

In the third row of Table I we have tabulated these theoretical values. The theoretical upper limits are much too small to explain the experimental peak intensities. A result closer to the experimental values might be obtained from a more complicated multiphonon Eliashberg kernel of the form

$$V_{k+q,k} = \sum_{n=1}^{\beta} \beta_n \frac{E_{k+q} + n\theta}{(E_{k+q} + n\theta)^2 - E_k^{-2}},$$
 (7)

where β_n are multiphonon coupling constants. Peak n + 1 could then result from the pulse of the very strong peak at the gap and from the pulses of peaks $1 \cdots n$ instead of from the pulse of peak n alone, as for the one-phonon kernel.

Multiphonon interactions can be obtained from higher order single phonon terms or from anharmonic terms in the lattice Hamiltonian; both of these are usually small at He temperatures. For example, phonon spontaneous decay broadening gives $h/\tau_p \sim 0.04 \theta$ for transverse phonons in Pb at helium temperatures.⁶

It appears that by the use of the Eliashberg kernel, assuming an Einstein phonon distribution, harmonic structure similar to that observed should result from an exact solution of the gap equation. But further understanding of the strong electron-phonon interactions involved will be necessary to explain the relatively slow decay of the strengths of the harmonics.

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¹⁰Integrals of this kind frequently occur in Fermi surface problems, where they can also be evaluated by the method of steepest descents.

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