ELECTRON INTERFERENCE IN A NORMAL METAL INDUCED BY SUPERCONDUCTING CONTACTS

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In the course of a tunneling study of the superconducting proximity effect between lead and silver films we have observed an oscillation in the tunneling characteristics of junctions where tunneling is into the silver film (<2000 Å) backed by lead. In thin silver films (<500 Å) we observe an image of the lead phonon spectrum in the first derivative of the tunneling characteristic and in thicker films (500-2000 Å) this structure oscillates with both voltage and film thickness. We believe that this is a result of electron interference in the normal metal. A similar mechanism, that of quasiparticle interference in a superconductor, was invoked by Anderson and one of the authors[1] to explain the effects observed by Tomasch[2] in tunneling into thick superconducting films.

In a bulk superconductor the E_k-vs-k diagram can be represented[3] as in Fig. 1(a). Electrons injected by tunneling at energy ω enter the superconductor as a linear superposition of quasiparticle excited states at momenta k_1 and k_2 where

$$k_{1,2} = k_F \pm (\omega^2 - \Delta^2)^{1/2}/v_F \qquad (1)$$

For a uniform superconductor the probability of electron injection is proportional to the incoherent sum of the injection probabilities into the two channels k_1 and k_2. As pointed out by Cohen, Falicov, and Phillips,[4] the coherence factors drop out after summing over the two states and the tunneling current is proportional to the final density of states. For the case of a superconducting Pb film of thickness d backed by Ag the part of the electron wave injected into the state k_1 propagates across the Pb film and is scattered into the state k_2 by the gap perturbation in the Ag. This wave propagates back across the Pb film and interferes with the k_2 part of the initial electron wave. This interference between the two channels is observed in the tunneling characteristic as an oscillation of period

$$\frac{|k_1 - k_2|d}{2\pi} = \frac{2d(\omega^2 - \Delta^2)^{1/2}}{2\pi\hbar v_F} \qquad (2)$$

as found by Tomasch in both Pb and In.

Now consider the case reported here of an Ag film of thickness d backed by superconducting Pb. The injected electron propagates across the Ag film to the interface as an electron in state k_1' [Fig. 1(b)] and is scattered into a hole state k_2' by the energy-gap function in the Pb film. This energy-gap function can pair an excited electron with an electron inside the Fermi sea leaving a hole excitation. The hole propagates back across the Ag film in state k_2' but, since Ag is a normal metal, it cannot interfere with the original electron wave. In order for interference to occur the hole must be reflected at the Ag surface, propagate to the interface, be scattered into the electron state k_1'

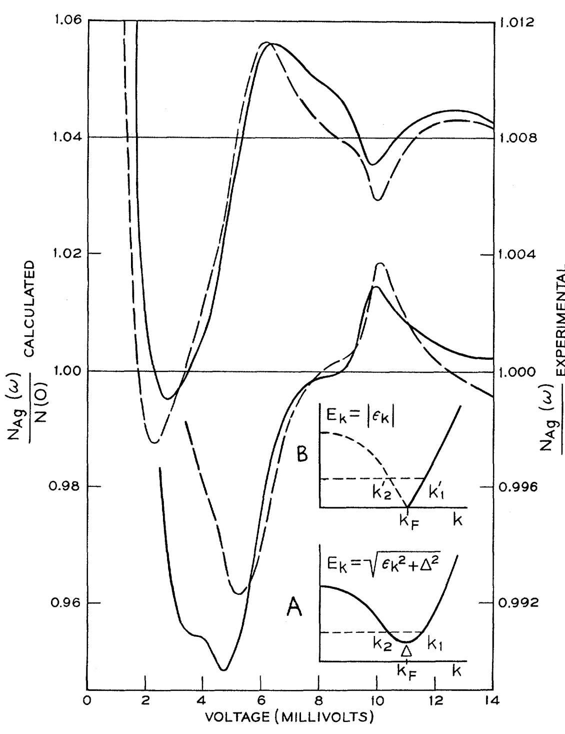

FIG. 1. Solid lines show measured densities of states for two junctions with silver film thicknesses of 1000 Å (upper) and 470 Å (lower) and lead thickness of 850 Å. Dashed lines show the calculated variation of density of states. The value $N_{Ag}(\omega)/N(0) = 1$ has been shifted vertically by 0.04 (left-hand scale) for the upper curves. Inserts (a) and (b) show the E_k-vs-k diagrams for a superconducting and normal metal, respectively.

by the Pb energy-gap function, and propagate as an electron to the Ag film surface. Here it can interfere with the original electron wave. This interference produces an oscillation in the tunneling characteristic, of period

$$\frac{|k_1'-k_2'|2d}{2\pi}=\frac{4d\omega}{2\pi\hbar v_F}. \quad (3)$$

The calculation of the tunneling density of states for this case of Ag backed by Pb is similar to that presented in Ref. 1. We assume that the energy-gap function is Δ_ω in the Pb film and vanishes in the Ag film, and calculate the Green's function to second order in Δ_ω. If $\tilde{G}_\omega{}^0(rr')$ is the Green's function of the normal metal at energy ω in the Nambu representation, the Green's function in the superconducting state is

$$\tilde{G}_\omega(rr')=\tilde{G}_\omega{}^0(rr')+\int d\bar{r}\tilde{G}_\omega{}^0(r\bar{r})\Delta_\omega(\bar{r})\tau_1\tilde{G}_\omega{}^0(\bar{r}r') + \int d\bar{r}d\bar{r}'\,\tilde{G}_\omega{}^0(r\bar{r})\Delta_\omega(\bar{r})\tau_1\tilde{G}_\omega{}^0(\bar{r}\bar{r}')\Delta_\omega(\bar{r}')\tau_1\tilde{G}_\omega{}^0(\bar{r}'r'), \quad (4)$$

where

$$\Delta_\omega(r)=0,\ 0<x<d,$$
$$=\Delta_\omega,\ x>d. \quad (5)$$

We assume specular scattering at the free silver surface so that $\Delta_\omega(-x)=\Delta_\omega(x)$. The tunneling density of states is[5]

$$N(\omega)=\mathrm{Im}\,\mathrm{Tr}\tilde{G}_\omega(rr)|_{x=0}. \quad (6)$$

The Green's function for the pure normal metal is

$$\tilde{G}_\omega{}^0(R)=-(m/2\pi\hbar R)\exp[i\omega R/\hbar v_F] \times[i\sin k_FR+\tau_3\cos k_FR]. \quad (7)$$

After performing the integrations of Eq. (4) we find for the density of states

$$N_{Ag}(\omega)=N(0)\,\mathrm{Re}\left[1+\frac{\Delta_\omega{}^2}{2\omega^2}F\left(\frac{4d\omega}{\hbar v_F}\right)\right], \quad (8)$$

where

$$F(y)=\int_1^\infty \exp[ixy](dx/x^2). \quad (9)$$

$F(y)$ is an oscillating function of $y=4d\omega/\hbar v_F$ with period ~2π. We expect impurity scattering or diffuse scattering to reduce the magnitude of the interference term [and possibly to change the detailed shape of $F(y)$]. Thus we expect to observe the oscillations only for relatively clean films with mean free paths ~d, a condition satisfied in the experiments described below.

In the derivation of Eq. (8) we have made a weak-coupling approximation by neglecting the renormalization function Z_ω. However, Eq. (8) is valid for a strong-coupled superconductor, such as lead, if one uses for Δ_ω the renormalized energy-gap function.[3,5]

Note that, except for the interference factor F, Eq. (8) is just the approximate expression (for $\omega \gg \Delta$) for the tunneling density of states of a single Pb film.[5] For thin Ag films we expect to see the "Pb phonon structure"[6] in $N_{Ag}(\omega)$ and for thicker Ag films we expect to see the magnitude of this structure oscillate with period $4d\omega/2\pi\hbar v_F$.

A detailed calculation of $N_{Ag}(\omega)$ for $d=1460$ Å and $v_F=1.0\times10^8$ cm/sec is shown in Fig. 2. The lower curves show the real and imaginary parts of the interference factor $F(y)$ plotted versus both energy and y. The middle curves show the real and imaginary parts of Δ_ω obtained from an analysis of tunneling data on a single lead film.[6] The resulting $N_{Ag}(\omega)$ obtained from (8) is shown as the dashed line of the upper two curves. The solid line is an experimentally measured $N_{Ag}(\omega)$ for a junction Al-I-Ag/Pb 15 where tunneling was into a 1460-Å Ag film backed by 1010 Å of Pb. The Fermi velocity $v_F=1.0\times10^8$ cm/sec was chosen to give the best fit to the experimental data. Note that the magnitude of the calculated structure is about five times that found experimentally. From our study[7] of the proximity effect in this Ag-Pb system we have concluded that it is essential to take into account the properties of the Ag-Pb interface. To obtain reasonable agreement between theory and experiment for the transition temperature and energy gap of the system we have represented the interface by a potential barrier with a transmission

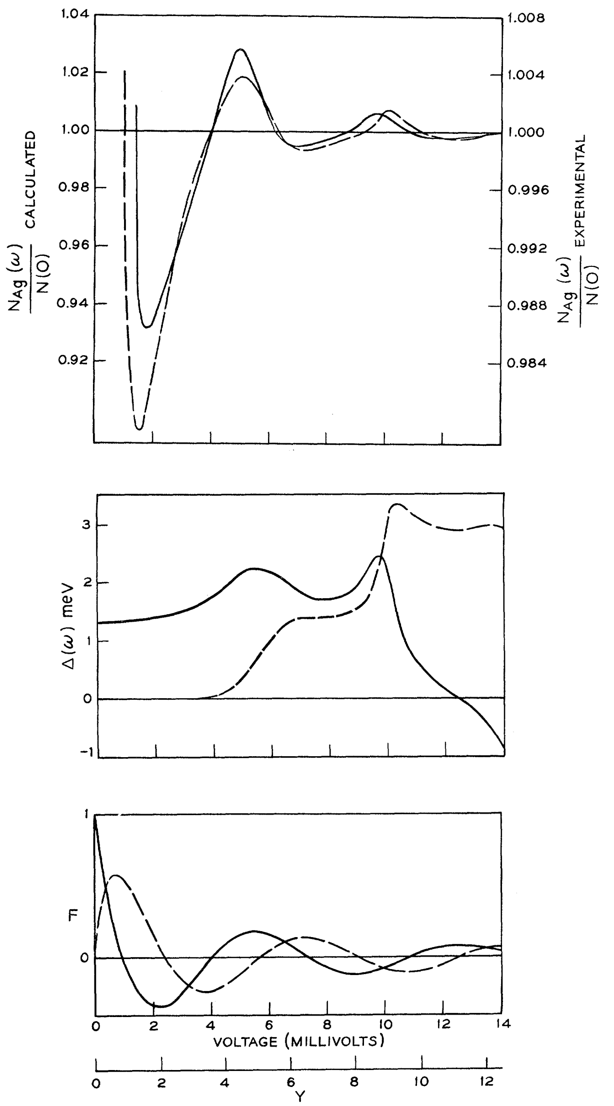

FIG. 2. The lower curves show the real (solid line) and imaginary (dashed line) parts of the interference term $F(y)$ plotted against y and against voltage for $d=1460$ Å. The middle curves are the real (solid line) and imaginary (dashed line) parts of the gap parameter Δ_ω for lead. The upper solid curve is a measured tunneling density of states for a 1460-Å Ag film backed by 1010 Å of lead. The upper dashed curve is a calculated density of states obtained from Eq. (8) using $F(y)$ and Δ_ω shown below.

coefficient $t \approx 0.4$. This presumably takes into account the mismatch of the electronic wave functions and the presence of real contamination at the interface. The inclusion of the surface barrier in this way reduces the interference term of Eq. (8) by the factor $t^2 \approx 0.16$ which accounts for the magnitude discrepancy.

A similar comparison between calculated and measured values of $N_{\rm Ag}(\omega)$ is shown in Fig. 1 for two more junctions where the thickness of the silver film has been reduced to 1000 and 470 Å, respectively. In Ag/Pb 13 where $d=470$ Å, the function $F(\omega)$ has a period of ~22 meV and the familiar structures in the density of states corresponding to transverse and longitudinal phonon peaks simply appear "upside down" compared to tunneling characteristics obtained for Pb films.[6] (But note that the magnitude of the effects is only ~6% of that in Pb films.) When d is 1000 Å in Ag/Pb 14 the period of $F(\omega)$ is ~10 meV and its effect in the longitudinal region (~10 meV) is particularly striking. The calculated plots (dashed lines) for both junctions of Fig. 1 were also obtained using a value of $v_{\rm F}=1.0\times10^8$ cm/sec.

The preferential orientation of the silver films was determined to be 110 perpendicular to the film surface. This was found by an electron-diffraction measurement on a 2500-Å Ag film which was evaporated onto an oxidized aluminum film on a sapphire substrate. This reproduces the condition of the Ag films used in the junctions. The experiment measures a Fermi velocity in the 110 direction which is on the belly of the Fermi surface.[8] (The Fermi surface contacts the zone boundary in the 111 direction.) We would, therefore, expect a Fermi velocity comparable to the free electron value of 1.4×10^8 cm/sec. To determine an accurate value of $v_{\rm F}$ we need to observe many oscillations in a thick clean silver film. From our measurements on these thin films we suggest $v_{\rm F}=(1.1\pm0.2)\times10^8$ cm/sec in the 110 direction.

Junctions of the type Al-I-Ag/Pb were prepared in the order shown. An aluminum film was evaporated, allowed to oxidize, crossing strips of Ag were evaporated and covered with lead films at room temperature. This procedure can be questioned because of the possibility of diffusion and alloying effects. We believe that plots of the kind shown in Figs. 1 and 2 indicate that these problems are not present in the Ag/Pb system.

We have found a striking verification of the space dependence of the theory of superconductivity as originally proposed by Gor'kov.[9] We have also observed the interplay between the space dependence (given by the interference factor F) and the time dependence (given by the ω dependence of Δ_ω) of the strong-coupled theory due to Eliashberg.[10] The time dependence of the theory for a uniform superconductor has already been verified in some detail.[5,6] In addition we have a verification of (6), namely that tunneling measures the Green's function at the tunneling surface.

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Note added in proof.—It has been brought to our attention that P. D. de Gennes and D. Saint-James [Phys. Letters 4, 151 (1963)] have solved a theoretical model similar to that considered here (but for energies less than Δ) and, indeed, oscillations are quite apparent in their results. We have not succeeded in making a sufficiently thick, clean Ag film to observe the oscillations at these low energies.

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EFFECT OF ELECTRON-SPIN PARAMAGNETISM ON THE CRITICAL FIELD OF THIN Al FILMS*

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We report measurements of the critical field parallel to the plane of ~100-Å Al films, through the field region where spin paramagnetism limits the critical field. The question of electron-spin polarization and superconductivity is of great importance because of the absence of a general explanation of the measurements of the Knight shift (the shift in the nuclear resonance frequency due to the additional field produced by the net electron polarization). The Knight-shift measurements[1] in superconducting tin, mercury, and aluminum yield values at 0°K that are about $\frac{2}{3}$ of the normal-state values. On the basis of the BCS theory where there is complete spin pairing at 0°K, one would expect zero Knight shift at 0°K if only spin polarization effects were important. Various explanations advanced to explain the Knight-shift measurements, including some which do not rely only on spin polarization, have been discussed by Schrieffer.[1] The present measurements yield information about the spin polarization, through its effect on the critical field, and thus complement the Knight-shift measurements.

The effects of electron paramagnetism on the upper field of superconductors were treated by Chandrasekhar[2] and Clogston[3] for the limiting case of a superconductor showing complete field penetration, no Meissner effect, at 0°K. The Clogston[3] calculation, which compares the paramagnetic energy of the normal metal in a field to the gap energy of a superconductor with no Meissner effect, gives the upper limit of the critical field. We have made a simple extension of the Clogston argument to a film at 0°K that does show diamagnetism. From this calculation and our data we can estimate a value for the upper paramagnetic limited critical field for the "no Meissner effect" case.

In the discussions to follow we discuss our data in terms of the simple two-fluid temperature dependence[4]; this temperature dependence is used for convenience and because better theoretical estimates such as those of de Gennes and Tinkham[5] and Maki[6] do not, for our purposes, differ too significantly from this depen-