Office (Durham) under Grant No. DA-ARO-D-31-124-G844.

[1]Y. B. Kim, C. F. Hempstead, and A. R. Strnad, Phys. Rev. 139, A1163 (1965).
[2]R. G. Jones, E. H. Rhoderick, and A. C. Rose-Innes, Phys. Letters 24A, 318 (1967).
[3]D. E. Farrell, I. Dinewitz, and B. S. Chandrasekhar, Phys. Rev. Letters 16, 91 (1966).
[4]I. Giaever, Phys. Rev. Letters 15, 825 (1965); P. Solomon, Phys. Rev. Letters 16, 50 (1966).
[5]F. Seitz, The Modern Theory of Solids (McGraw-Hill Book Company, Inc., New York, 1940), p. 162.
[6]P. W. Anderson, N. R. Werthamer, and J. M. Luttinger, Phys. Rev. 138, A1157 (1965).
[7]B. B. Schwartz, Bull. Am. Phys. Soc. 11, 106 (1966).
[8]J. Bardeen and M. J. Stephen, Phys. Rev. 140, A1197 (1965).

SUPERCONDUCTIVE PAIRING ACROSS ELECTRON BARRIERS*

Morrel H. Cohen and D. H. Douglass, Jr.†
Department of Physics and Institute for the Study of Metals, The University of Chicago, Chicago, Illinois
(Received 29 May 1967)

We consider the possibility of superconductive pairing of two electrons separated by a barrier. We argue that such pairing is possible in principle for all metals and may lead to high transition temperatures.

Systems comprised of two superposed metal films S_1 (thickness t_1) and S_2 (thickness t_2) separated by an insulating barrier B (thickness t_B), as in Fig. 1(a), have been intensively studied in connection with superconductive tunneling.[1-3] It has been assumed that both electrons of the Cooper pairs in such systems lie either in S_1 or S_2. In this Letter we consider the possibility of superconductive pairing across B, i.e., Cooper pairs with one electron in S_1 and one in S_2. We find that such pairing is possible in principle and would lead to novel effects including the possibilities of higher transition temperatures for known superconductors and of superconductivity in previously nonsuperconducting materials, e.g., the magnetic metals.

Apart from partial reflection at the S_1B and BS_2 interfaces, B does not present a barrier to phonons. An electron in S_1 can emit a phonon which subsequently travels across B and is absorbed by an electron in S_2, Fig. 1(b), resulting in an attractive, phonon-induced interaction V_{ph}^{12} comparable in magnitude with the interaction within a single film V_{ph}^{11}. The Coulomb interaction across the barrier V_C^{12}, on the other hand, differs greatly from that within a single film V_C^{11}. Because the screening cloud around an electron is of radius λ_{TF}, the static screening length, electrons on opposite sides of the barrier interact via a dynamically screened Coulomb interaction similar to that in the bulk. The closest distance of approach, however, is only t_B. Provided t_B, t_1, and t_2 exceed λ_{TF}, V_C^{12} is reduced essentially to zero for low-energy transfers, leaving a residuum at higher energies which acts as an attractive interaction.[4] Dynamical effects such as the exchange of low-frequency surface plasmons[5,6] could provide an additional attractive interaction. These crude arguments suggest that the net interaction across the barrier, $V_T^{12} = V_{\text{ph}}^{12} + V_C^{12}$, could be attractive for all choices of metals S_1 and S_2. Such an attractive interaction would give rise to pairing across the barrier as a possible new channel whereby superconductivity could be

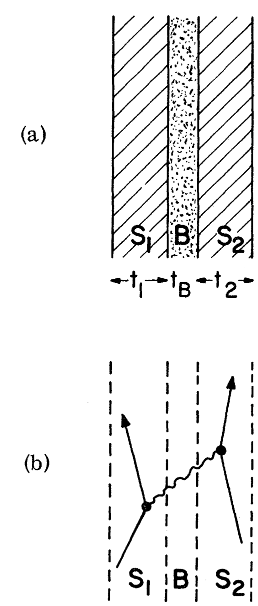

FIG. 1. (a) Metal (S_1)-insulator (B)-metal (S_2) sandwich. (b) Feynman diagram for phonon-induced electron-electron interaction across the barrier V_{ph}^{12}.

established.[7]

The pair amplitude $F(x,y)=\langle\psi_\uparrow(x)\psi_\downarrow(y)\rangle$ can be decomposed into a set of four pair amplitudes $F_{ij}(x,y)$, where $i=1$ refers to x in S_1, $i=2$ refers to x in S_2, and similarly for j and y. The case $i\neq j$ refers to pairing across the barrier. At the transition temperature the integral equation satisfied by the F_{ij} is linear, and, for small tunneling amplitudes, the different F_{ij} are decoupled. Each Cooper pair amplitude F_{ij} has an associated transition temperature $T_{ij}=T_{ij}$ related to the corresponding total interaction V_T^{ij}. The superconducting transition temperature of the sandwich T_c is $\max(T_{ij})$. Energy gap parameters Δ_{ij} corresponding to the F_{ij} may be defined.

If our simple arguments about V_T^{12} are correct, T_{12} always exists. Supposing that $T_1(=T_{11})>T_2(=T_{22})$ for convenience, we have two possibilities: case I, $T_c=T_{12}>T_1$, and case II, $T_c=T_1>T_{12}$. In case I the superconductivity just below $T_c=T_{12}$ is associated entirely with pairing across the barrier. In thick films this pairing is confined to a layer of depth R in the vicinity of the barrier; there is no associated energy gap. In films thinner than R the pairing is uniform, and the energy gap is Δ_{12}. Here R is the range of $V_{\rm ph}^{12}$ and is given by $v_{\rm F}/\omega_{\rm D}$ or the phonon mean free path, whichever is the smaller. In case II, the transition temperature is T_1, unaffected by interaction across the barrier.

Case I offers an intriguing possibility for increased transition temperatures. We have carried out a highly oversimplified model calculation of T_{12} for S_1 equal to S_2, for t_1, t_B, and t_2 less than R, and for t_B greater than λ_{TF}. We have assumed the pair amplitude F_{12} to be independent of position, and the kernels in the integral equation for Δ_{12} to be replaced by square wells, i.e., a Tolmachev model.[8] In particular, the Coulomb kernel is assumed to vanish for either quasiparticle energy below ω_l, a lower cutoff expressing the effect of screening, and to be unaffected above ω_l. We find

$$T_{12}=1.14\theta_{\rm D}\exp(-1/\rho_{12}), \tag{1}$$

where

$$\rho_{12}=\rho_{\rm ph}^{12}+\rho_c^{12}. \tag{2}$$

We estimate that

$$\rho_{\rm ph}^{12}=a\xi\rho_{\rm ph}^{11}, \tag{3}$$

where ξ, smaller than one, results from acoustic mismatch and a from the phase change of the phonon in crossing the barrier. For a perfectly flat barrier, a becomes

$$a=\tfrac{1}{2}(1+|\sin 2k_{\rm F}t_B|/2k_{\rm F}t_B). \tag{4}$$

Our assumptions concerning the Coulomb kernel lead to

$$a\rho_c/(3-\rho_c\ln\omega_u/\omega_{\rm D})>\rho_c^{12}>0 \tag{5}$$

for ρ_c^{12}, provided $\omega_l>\omega_{\rm D}$. Here ω_u is the upper cutoff for the Coulomb interaction. Equation (5) for ρ_c^{12} should be compared with the corresponding result for ρ_c^{11},

$$\rho_c^{11}=-\frac{\rho_c}{1+\rho_c\ln\omega_u/\omega_{\rm D}}, \tag{6}$$

where ρ_c is the magnitude of the Coulomb kernel for S_1. The upper limit for ρ_c^{12} in (5) corresponds to the most favorable choice of ω_l,

$$\rho_c\ln\omega_u/\omega_l=1. \tag{7}$$

The possibility that T_{12} greater than

$$T_{11}=1.14\theta_{\rm D}\exp(-1/\rho_{11}), \tag{8}$$

$$\rho_{11}=\rho_{\rm ph}^{11}+\rho_c^{11}, \tag{9}$$

arises from the reversal of the sign of the Coulomb contribution in (5) relative to (6). For T_{12} to exceed T_{11} this sign reversal must compensate for the diminution of $\rho_{\rm ph}^{12}$ by the factors a and ξ. Deviations from planar geometry can increase a towards unity.

For this simple model, high values of T_{12} are favored by large $\rho_{\rm ph}^{11}$ and by large ρ_c, i.e., by large density of states, large $|V_{\rm ph}|$, and large V_c. Among superconductors, the best candidates for having T_{12} significantly larger than T_1 are those metals in which a large $\rho_{\rm ph}^{11}$ is nearly canceled by a large $|\rho_c^{11}|$. Quite large values of T_{12} may occur for metals which are nonsuperconducting in the bulk because of a large $|\rho_c^{11}|$, i.e., a large ρ_c can be expected to occur for magnetic metals.

The microscopic explanation usually given for the absence of superconductivity in ferromagnetic metals involves a Coulomb repulsion strong enough for the exchange interaction to cause ferromagnetism and Fermi surfaces of different size for opposite spins so that perfect pairing is impossible. The strong Coulomb repulsion enhances the tendency towards pairing across the barrier in a sandwich structure; perfect opposite-spin pairing could occur if the magnetization pointed oppositely in S_1 and S_2. One could thus expect superconductivity in S_1-B-S_2 sandwiches, although it may be difficult to freeze in antiparallel magnetizations. It might be easier to study Pd, which is nearly ferromagnetic, and then to follow the superconductivity into the ferromagnetic state in dilute PdNi alloys. Similarly, one could expect superconductivity in sandwiches made of antiferromagnetic metals. States in S_1 would be paired with their time-reversed conjugates in S_2, requiring spatial coherence of the domain structure in S_1 with that in S_2. Again, it might be easier to study nonmagnetic CrV alloys ($\lesssim 70\%$ V) and then to follow superconductivity into the antiferromagnetic state in alloys containing more Cr.

The above predictions are moot, based as they are on indirect physical argument and highly oversimplified models. Even finding superconductivity in sandwiches of normal or magnetic metals would not constitute firm evidence for the existence of pairing across barriers. An experiment more specifically indicative than one which merely establishes superconductivity is necessary. Suppose a supercurrent to flow in S_1 parallel to the barrier. If pairing across the barrier exists, a corresponding supercurrent must flow in S_2 if S_2 is an element of a closed superconducting circuit.[9] On the other hand, if the supercurrent in S_2 is constrained to flow at a velocity different from that in S_1 (an extreme case would be S_2 an element of an open circuit), a voltage must develop which is proportional to the velocity difference for low velcoities but no larger than the energy gap. Observation of these and related phenomena would provide strong support for our ideas; such experiments are in progress here.

We have examined the existing experimental literature for evidence bearing on our ideas. We have found reports of relevant experiments on W,[10] Al,[11,12] Tl,[13] and Fe[14] films, respectively.

W films deposited in a relatively poor vacuum were found to have transition temperatures as high as 4°K[10]; the bulk transition temperature of W is 0.01°K. X-ray patterns showed only broadened αW lines in unannealed films, from which lower limits to the W "particle" sizes are inferred. The transition temperature increased with decreasing particle size. Decrease in particle size by itself may increase T_c[15]; however, the observed increases appear too large to have occurred at particle sizes of ~100 Å. It is conceivable that gaseous impurities were concentrated between the "particles" leading locally to an -S-B-S- structure. Assuming such a structure, pairing across the barriers could lead to high transition temperatures T_{12}. If we choose the conservative value of $\frac{1}{2}$ for both a and ξ, use Garland's estimates[16] for ρ_{ph}[11], ρ_c, and ρ_c[11], and insert these into Eqs. (1)-(6), we find that a T_{12} of 4°K would require a ρ_c[12] of only 30% of the maximum given by Eqs. (5) and (7). We conclude that pairing across the barrier provides a possible explanation for the high T_c's observed in W films. Similar analyses of the T_c enhancements observed for Al films[12,13] lead us to conclude that pairing across the barrier is a possible explanation there, as well.

Rühl[13] has reported T_c observations for Tl-(Tl oxide)-Tl sandwiches with $t_1 \simeq 70$ Å, $t_B \simeq 20$ Å, and t_2 variable. Once sufficient Tl has been deposited to convert all of the oxygen layer chemisorbed on the oxide to oxide, S_2 begins to build up and T_c to increase rapidly. The increase in T_c is 0.3°K for a t_2 of only ~3 Å, which is three times the enhancement caused by the charge depletion of S in the S-B structure. The enhancement is probably not associated with a size-affected superconductivity of S_2 because t_2 is too small for a continuous electrical path through S_2. We infer the transition occurs in S_1; S_2 thus influences the superconductivity of S_1 through ~20 Å of coherent oxide layer. Again pairing across the barrier provides a possible interpretation. Observations of T_c enhancements in a variety of S_1-B-S_2 structures described in the next Letter[17] can also be interpreted in terms of pairing across the barrier.

Mikhailov et al.[14] have reported the observations of superconductivity with $T_c > 4$°K in Fe films deposited at He temperatures. They evaporated Fe through a rotating pin hole onto a cold cylindrical substrate; the films disintegrated at hydrogen temperatures. Subsequent investigations in which great care was taken with regard to vacuum and to cleanliness failed to reproduce their results.[18,19] These facts suggest to us that the Fe films of Mikhailov et al. had a jelly-roll-like structure of layers of condensed gas. Their remarkable observation of superconductivity in Fe could therefore be explained by pairing across a barrier.

We are indebted to K. L. Ngai for detailed assistance with the formal theory and to E. Economou for discussion of surface plasmons.

*Research has been supported in part by the National Aeronautics and Space Administration, and by the Army Research Office, Durham. In addition, it has benefited from support of Materials Science at the University of Chicago by the Advanced Research Projects Agency.

†Alfred P. Sloan Fellow.

[1]I. Giaever and K. Megerle, Phys. Rev. 122, 172 (1961).

[2]B. D. Josephson, Advan. Phys. 14, 419 (1965).

[3]We suppose B to be sufficiently thick that electron tunneling across it influences the gross thermodynamic properties of the sandwich negligibility.

[4]That a repulsive interaction which vanishes for low-energy transfer acts as though it were an additional attractive pairing interaction in the presence of $V_{\rm ph}$ may readily be seen from the Tolmachev two-square-well model. R. A. Ferrell, Phys. Rev. 111, 1214 (1955); J. W. Garland, Phys. Rev. 153, 460 (1967).

[5]Ferrell, Ref. 4.

[6]E. Economou, to be published.

[7]Although we discuss our planar geometry here for convenience, specimens of more complex geometry containing continuous metallic regions separated by barriers are probably still more favorable for pairing across barriers.

[8]N. N. Bogoliubov, V. V. Tolmachev, and D. V. Shirkov, New Method in the Theory of Superconductivity (Academy of Sciences of USSR, 1958).

[9]We note that this is a dc current transformer. Suppose S_1 and S_2 are the same material. Pairing requires that the velocities are the same in S_1 and S_2. If t_1 and t_2 are both less than R, the secondary to primary current ratio is just t_2/t_1.

[10]O. F. Kammerer and M. Strongin, Phys. Letters 17, 224 (1965); Basic Problems in Thin Film Physics, edited by R. Niedermayer and H. Mayer (Vandenhoek and Ruprecht, Göttingen, 1966), p. 511.

[11]M. Strongin, O. F. Kammerer, and A. Paskin, Phys. Rev. Letters 14, 949 (1965).

[12]Abeles et al., Phys. Rev. Letters 17, 632 (1966).

[13]W. Rühl, in Proceedings of the Ninth International Conference on Low Temperature Physics, edited by J. G. Daunt, D. O. Edwards, F. J. Milford, and M. Yaqub (Plenum Press, New York, 1965), p. 475.

[14]Yu. G. Milhailov et al., Zh. Tekh. Fiz. 29, 931 (1958) [translation: Soviet Phys.-Tech. Phys. 4, 844 (1959)].

[15]C. J. Thompson and J. M. Blatt, Phys. Letters 5, 6 (1963); A. Paskin and A. D. Singh, Phys. Rev. 140A, 1965 (1965); V. Z. Kresin and B. A. Tavger, Zh. Eksperim. i Teor. Fiz. 50, 1689 (1966) [translation: Soviet Phys.—JETP 23, 1124 (1966)]; R. Parmenter, to be published.

[16]J. W. Garland, to be published.

[17]M. Strongin, O. F. Kammerer, D. H. Douglass, Jr., and Morrel H. Cohen, following Letter [Phys. Rev. 19, 121 (1967)].

[18]B. G. Lazarev et al., Zh. Eksperim. i Teor. Fiz. 40, 105 (1961) [translation: Soviet Phys.—JETP 13, 75 (1961)].

[19]J. C. Suits, Phys. Rev. 131, 588 (1967).

EFFECT OF DIELECTRIC AND HIGH-RESISTIVITY BARRIERS ON THE SUPERCONDUCTING TRANSITION TEMPERATURE OF THIN FILMS*

M. Strongin and O. F. Kammerer
Brookhaven National Laboratory, Upton, New York

and

D. H. Douglass, Jr.,† and Morrel H. Cohen
Department of Physics and Institute for the Study of Metals, The University of Chicago, Chicago, Illinois
(Received 29 May 1967)

Increases in the transition temperature of a structure consisting of alternate 60-Å layers of superconductor S and barrier material B are observed whenever the sequence SBS is formed. The effect was observed for Al, Zn, In, Sn, and Pb and for a variety of barrier materials.

In this paper we present the results of a series of experiments on films deposited onto substrates cooled to low temperatures. These experiments were initiated to investigate the effects predicted by Cohen and Douglass.[1] They have predicted that a thin sandwich of superconductor-dielectric-superconductor could have a T_c quite different from the superconductor alone. To investigate this idea a structure was made consisting of alternate layers (~60 Å) of pure superconductor (S) and electron barrier material (B) which was either a dielectric or high-resistivity metal. The main result of these experiments is that whenever S was evaporated so as to form the combination -SBS, an increase in transition temperature