Transition temperature of proximity-effect thin sandwiches*

Guy Deutscher† and Ora Entin-Wohlman‡
Department of Physics, University of California, Los Angeles, California 90024

Zvi Ovadyahu
Department of Physics and Astronomy, Tel Aviv University, Tel Aviv, Israel

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We study the effect of an interface barrier on the T_c of a superconducting-normal (S-N) metal sandwich in the Cooper limit. The calculation is based on the diffusion approximation of de Gennes and the barrier is accounted for by appropriate boundary conditions at the interface. In contrast with the original de Gennes result, it is shown that for finite thicknesses T_c does not depend only on the N to S thickness ratio but also on the absolute value of the thickness. An analysis of experimental data obtained on Pb-Cu sandwiches shows that this finite-thickness correction is important even at very small thicknesses and for rather transparent barriers. Assuming that the penetration probability σ shows only random variations in a series of samples prepared under the same nominal conditions, the best fit between theory and experiment is obtained for a BCS coupling parameter in Cu equal to 0.015.

I. INTRODUCTION

The Cooper limit of a superconducting-normal metal (S-N) sandwich is attained when both slabs are thinner than their respective coherence lengths. In this limit the superconducting properties do not vary essentially across the metals. This eliminates the problems associated with space dependence of the pair potential and thus this limit is amenable to a rather complete solution, which may be used to study the BCS interaction parameter of the normal side.

Neglecting the effect of the barrier between S and N, de Gennes[1] obtained

$$\ln(1.14\hbar\omega_0/k_BT_{DG}) = (1+R)/(\lambda_s + R\lambda_N). \quad (1)$$

Here T_{DG} is the transition temperature when the barrier effect is completely neglected, and

$$R = N_N d_N / N_S d_S, \quad (2)$$

where d_N, d_S are the thicknesses of N, S, respectively, N_N, N_S are the respective densities of states at the Fermi level, and

$$\lambda_{S,N} = N_{S,N} V_{S,N}, \quad (3)$$

where V_S, V_N are the respective BCS interaction potentials. The result (1) was derived in the diffusion approximation in which it is assumed[2]: (i) both metals are "dirty," i.e., their mean free paths are much smaller than the respective coherence lengths, (ii) the superconducting properties vary slowly over distances of the order of the mean free path. The second condition should be modified when the effect of the barrier is studied. An expression similar to (1) was obtained by Silvert and Cooper[2] as an upper bound for $\ln(1.14\hbar\omega_0/k_BT_c)$.

The result (1) of de Gennes does not depend on the absolute values of the thicknesses but only on R. Thus T_c measurements of Cooper-limit sandwiches seem to be a simple tool to investigate λ_N. However, in T_c measurements done on thin Pb-Cu samples we found that T_c (in the Cooper limit) decreases when d_S, d_N are reduced at constant R. The arguments in the articles by Silvert and Cooper[2] and McMillan,[3] suggest that the disagreement with the de Gennes formula is due to the neglect of the barrier effect. As was pointed out by de Gennes,[1] the barrier can be accounted for by generalizing the boundary conditions at the interface. In this article we derive an expression for T_c using the generalized boundary conditions. They introduce an effective length L, roughly of the order of d_N/σ,[4] where σ is the penetration probability of the barrier. We show that T_c is always higher than T_{DG} and find a condition, involving σ, under which T_{DG} is approached.

In our thin Pb-Cu samples this condition does not hold even assuming $\sigma = 1$, although the films are in the Cooper limit. We use our T_c data to calculate σ for different values of λ_N. Assuming that all samples have the same σ, or at least that σ varies randomly among them, the correct λ_N is the one for which σ does not have any systematic variation as a function of thickness. In this way we find that λ_N of Cooper is 0.015.

Interestingly enough, our equations for the pair potentials in N, S are of the same functional form as the ones which can be derived from McMillan's[3] tunneling model. In McMillan's model the barrier effect is described in terms of σ and the average path length L_N between collisions with the barrier. Comparing our equations with those of McMillan, we relate the effective length L to

L_N. McMillan treats the tunneling through the barrier in second-order perturbation theory. Thus his model describes a barrier with low penetration probability. On the other hand, de Gennes[1] approximation is applicable to a "transparent" barrier. Our theory links the two cases and is suitable for comparison with experimental data, where σ may have intermediate values.

In Sec. II we derive the expression for T_c in the Cooper limit. In Sec. III we apply the theory to our experimental data, and Sec. IV includes the conclusions.

II. TRANSITION TEMPERATURE

To find the transition temperature, we first determine the pair potentials Δ_N, Δ_S in the two films. At T_c, $\Delta(x)$ is given by a linear integral equation[1]

$$\Delta(x) = V(x) \int dx' \, \Delta(x') k_B T \sum_{\omega} H(xx', \omega) . \tag{4}$$

The interface between the two slabs is at $x = 0$. Here $\hbar\omega = \pi k_B T(2n+1)$ and the sum runs over all integers n. The kernel $H(xx', \omega)$ satisfies the sum rule[1]

$$\int dx' \, H(xx', \omega) = \pi N(x)/\hbar |\omega| , \tag{5}$$

where $N(x)$ is the local density of states.

In the dirty limit $H(xx', \omega)$ satisfies a diffusion equation, with two boundary conditions at $x = 0$.[1,2] One of them is obtained from the sum rule[5]

$$D(x') \frac{d}{dx'} H(xx', \omega) \Big|_{x'=0^-}^{x'=0^+} = 0 , \tag{6}$$

where D_N, D_S are the diffusion coefficients. For the other boundary condition, de Gennes[1] writes down the general form

$$H(xx', \omega)|_{x'=0^+} = \frac{N_S}{N_N} \left(H(xx', \omega) + L \frac{d}{dx'} H(xx', \omega) \right) \Big|_{x'=0^-} . \tag{7}$$

The result (1) for T_{DG} is obtained when the second term on the right-hand side is neglected. The effective length L involves σ and is discussed as follows. De Gennes argues that when $\sigma \gg l/\xi$ (l is the mean free path and ξ the coherence length), then $L \sim l$. When the film is much thicker than the coherence length $\{[dH(xx', \omega)/dx']/H(xx', \omega)\}|_{x'=0^-} \simeq \xi^{-1}$, and the second term on the right-hand side of (7) is of the order l/ξ compared to the first term. Thus it is negligible in the dirty limit. When the film is very thin

$$\frac{1}{H(xx', \omega)} \frac{d}{dx'} H(xx', \omega) \Big|_{\substack{x=0^+ \\ x'=0^-}} = \frac{1}{\xi_N(\omega)} \tanh \frac{d_N}{\xi_N(\omega)} \simeq \frac{d_N}{\xi_N^2(\omega)} , \tag{8}$$

where

$$\xi_N^2(\omega) = D_N/2\hbar|\omega| \tag{9}$$

and $d_N \ll \xi_N(\omega)$ in the Cooper limit. Thus for $L \sim l$ ($\sigma \gg l/\xi$), the second term on the right-hand side of (7) is of the order $l d_N/\xi_N^2$ compared to the first term, and therefore negligible.

To account for a barrier with low and intermediate values of σ, we retain the L term in (7). To solve for $H(xx', \omega)$, we assume that in the Cooper limit $H(xx', \omega)$ is essentially constant when x, x' are varied in one of the slabs. Thus we have to find three values: $H(NN, \omega)$ (x, x' in the N side), $H(SS, \omega)$, and $H(SN, \omega) = H(NS, \omega)$. From (5) we get

$$\begin{aligned} d_N H(NN, \omega) + d_S H(NS, \omega) &= \pi N_N/\hbar|\omega| , \\ d_N H(SN, \omega) + d_S H(SS, \omega) &= \pi N_S/\hbar|\omega| , \end{aligned} \tag{10}$$

and from (7)–(9)

$$H(SS, \omega) = (N_S/N_N) H(SN, \omega)(1 + L d_N/\xi_N^2) . \tag{11}$$

Solving (10) and (11) and inserting into (4) we obtain

$$\Delta_N = \lambda_N \left(\frac{R}{1+R} A + \frac{1}{1+R} F \right) \Delta_N + \lambda_N \frac{1}{1+R} (A - F) \Delta_S , \tag{12}$$

$$\Delta_S = \lambda_S \left(\frac{1}{1+R} A + \frac{R}{1+R} F \right) \Delta_S + \lambda_S \frac{R}{1+R} (A - F) \Delta_N ,$$

where R, λ_N, λ_S are given by (2), (3), and

$$A = \pi k_B T_c \sum_{\omega} \frac{1}{\hbar|\omega|} , \tag{13}$$

$$F = \pi k_B T_c \sum_{\omega} [\hbar|\omega| + (1+R) D_N/2L d_N]^{-1} . \tag{14}$$

Using the BCS frequency cutoff ω_0 we write (13), (14) as follows:

$$A = \int_0^{\omega_0} d\omega \frac{1}{\omega} \tanh(\hbar\omega/2k_B T_c) , \tag{15}$$

$$F = \int_0^{\omega_0} d\omega \frac{\omega^2}{\omega^2 + [(1+R) D_N/2L d_N]^2} \frac{1}{\omega} \tanh(\hbar\omega/2k_B T_c) . \tag{16}$$

Equations (12), (15), and (16) are of the same functional form as those which can be derived from McMillan's[3] equations (11) and (16). [Note that Eqs. (37)–(39) in Ref. 3 are derived for $\lambda_N = 0$.] They will be identical if we make the identification

$$D_N/2L d_N = \Gamma_N = \hbar/2\tau_N , \tag{17}$$

where τ_N is the average time that an electron spends in N before penetrating the barrier and escaping into S.[6] τ_N is given in terms of σ and L_N (the average electron path length between collisions with the barrier),

$$\tau_N = L_N/v_F\sigma . \tag{18}$$

Using $D_N \simeq \hbar v_F l$ we find

$$L = (l/d_N)(L_N/\sigma) . \tag{19}$$

L_N is given in Ref. 3 by

$$L_N = 2d_N B(l/d_N) , \tag{20}$$

where B is a function of l/d_N. In very thin films with diffuse reflections at the boundaries the mean free path is thickness limited

$$1/l = 1/l_0 + p(1/d_N) , \tag{21}$$

where l_0 is the mean free path as $d_N \to \infty$, and p is a parameter that characterizes the scattering at the boundaries (i.e., $p=1$ for a diffuse scattering and $p=0$ for a specular reflection). For perfectly clean film $l_0 \gg d_N$, so that l is dominated by boundary scattering and B will be a constant, $B \sim 2$.[5] In this case $L_N \simeq 4d_N$. On the other hand, for $l_0 \ll d_N$, $L_N \simeq 4l_0$. Thus in general

$$L_N \simeq 4l. \tag{22}$$

The transition temperature is determined from the requirement that Eqs. (12) have a nontrivial solution

$$A \equiv \ln \frac{1.14\hbar\omega_0}{k_B T_c} = \frac{1 + R - F(\lambda_S R + \lambda_N)}{\lambda_S + R\lambda_N - F(1+R)\lambda_S\lambda_N} . \tag{23}$$

To investigate the dependence of T_c on σ and d_N we have to consider the function F. We write (16) in the form

$$F = \int_0^{\Theta_D/T_c} dx \frac{x^2}{x^2 + (a/k_B T_c)^2} \frac{1}{x} \tanh\frac{x}{2} , \tag{24}$$

where

$$a = (1+R) D_N/2Ld_N \simeq (1+R)\hbar v_F\sigma/8l \simeq (1+R)\hbar v_F\sigma/8d_N \tag{25}$$

and we used $\hbar\omega_0 = k_B\Theta_D$. Now since $\Theta_D/T_c \gg 1$, the $\tanh\frac{1}{2}x$ in the integrand may be approximated by 1 and consequently

$$F \simeq \tfrac{1}{2} \ln[1 + (k_B\Theta_D/a)^2] . \tag{26}$$

From (1) and (23) it is seen that T_{DG} is approached when F becomes very small. This occurs for $k_B\Theta_D/a \ll 1$, which can be written as follows:

$$\frac{T_c}{\Theta_D} \gg \frac{d_N l}{\xi_N^2(T_c)} \frac{4}{(1+R)\pi\sigma} , \tag{27}$$

where $\xi_N^2(T) = D_N/2\pi k_B T$. Thus it is seen that when σ is small, it becomes difficult to fulfill this condition. In Sec. III it will be shown that (27) does not hold, even for $\sigma \sim 1$, for our Cooper-limit samples. In the case where (27) holds and F is small we find [from (1), (23), (25), (26)]

$$\ln\frac{T_c}{T_{DG}} \simeq \left(\frac{\lambda_S - \lambda_N}{\lambda_S + R\lambda_N}\right)^2 \frac{32R}{(1+R)^2} \left(\frac{k_B\Theta_D}{\hbar v_F\sigma}\right)^2 l^2 . \tag{28}$$

Since in very thin films $l \sim d_N$, T_{DG} is approached from above as a linear function of d_N^2.

III. COMPARISON WITH EXPERIMENTAL DATA

Samples were prepared by thermal evaporation from tungsten boats onto glass substrates at room temperature. High vacuum (better than 10^{-6} Torr) was employed and the lead film was applied on top of the Cooper film in all cases (Pb and Cu 99.999% purity). Thickness measurements were performed *in situ* by a quartz crystal installed as close as physically possible to the substrate. Care was taken to keep a fixed geometry for all evaporations. The thickness ratio of Cu to Pb was kept equal to 1.50 ± 0.02 (i.e., $R = 0.85$) for the entire series. Within 5 min from preparation, samples were mounted in a ^{4}He cryostat where all subsequent measurements were taken. These included resistivity and transition-temperature measurements in a four-terminal con-

TABLE I. Experimental results for T_c, d_N, and the ratio of the resistance at room temperature (R_{RT}) to that at liquid-helium temperature (R_{He}). The quantity $\sigma k_B\Theta_D/a$ was calculated from the last equality in (25) for $R=0.85$, $\Theta_D = 100$ K. The mean free path l was computed from $\rho l \sim 6\times10^{-1}\,\Omega$ m, using resistance measurements at 4.2 K.

T_c (K)	$d_N(10^{-6}$ cm)	$\sigma k_B\Theta_D/a$	$A=\ln(114/T_c)$	R_{RT}/R_{He}	$l(10^{-6}$ cm)
2.4	2.09	0.757	3.86	1.67	2.1
2.8	2.52	0.912	3.71	1.62	1.9
3.05	2.74	0.992	3.62	1.61	2.5
3.3	3.48	1.260	3.54	1.72	2.5
3.88	3.93	1.423	3.38	1.75	3

TABLE II. F calculated from (23) and σ calculated from (25) and (26).

$\lambda_N=0.05$		$\lambda_N=0.02$		$\lambda_N=0.01$		$\lambda_N=0$		$\lambda_N=-0.01$		$\lambda_N=-0.02$		$\lambda_N=-0.05$	
F	σ	F	σ	F	σ	F	σ	F	σ	F	σ	F	σ
0.733	0.413	0.935	0.322	0.983	0.305	1.037	0.286	1.075	0.274	1.120	0.261	1.203	0.238
0.985	0.366	1.137	0.308	1.174	0.296	1.218	0.282	1.247	0.273	1.284	0.262	1.348	0.245
1.121	0.341	1.249	0.296	1.280	0.286	1.319	0.275	1.344	0.267	1.377	0.258	1.431	0.244
1.243	0.379	1.351	0.337	1.377	0.328	1.412	0.316	1.433	0.309	1.463	0.299	1.508	0.285
1.485	0.330	1.557	0.306	1.575	0.300	1.602	0.292	1.617	0.287	1.640	0.281	1.669	0.272

figuration. The Cu thicknesses and the transition temperatures of the samples are listed in Table I. It is immediately apparent from Table I that T_c varies significantly as a function of d_N. In order to see whether T_c approaches the de Gennes limit T_{DG}, we calculated $\sigma k_B \Theta_D/a$ from (25) for $\Theta_D=100$ K. Results are listed in Table I. It is seen that even for $\sigma \sim 1$ condition (27) is not satisfied and F is not smaller than 1. Thus expression (1) for T_{DG} cannot be used to calculate λ_N, although our samples are in the Cooper limit.

The value of F can be calculated from (23) assuming a given value for λ_N. Then σ can be found from (25) and (26). We have followed this procedure for seven values of $\lambda_N = \pm 0.05$,[7,8] ± 0.02, ± 0.01, and 0. In these calculations $\lambda_S = 0.39$, $R=0.85$, and $l=d_N$. Results are listed in Table II. Using the values of the measured l and equation (21) one observes that l is thickness limited. This is further supported by R_{RT}/R_{He} values which range from 1.58 to 1.75 (Table I) while a Cu sample of 6000 Å evaporated under the same conditions has a R_{RT}/R_{He} of ~10. Thus we conclude that $l \sim d_N$ is the relevant value to use in the present calculations. With a relation of the form $\sigma=\sigma_0+\alpha F$ we used F, σ, to calculate α by a least-mean-squares fit. Values of α for the different λ_N's are listed in Table III. Assuming that σ is independent on the normal-metal thickness, i.e., can have only random variations among the samples, we conclude from Table III that the best value for λ_N is $\lambda_N=0.015$. There is strong experimental evidence to support this assumption. Adkins and Kington[5] have shown that their energy-gap measurements in Cu-Pb sandwiches were consistent with a constant $\sigma \sim 0.25$,[6] for 200 Å $< d_{Cu} <$ 2000 Å. Freake[9] gives $\sigma=0.29$ both from T_c and gap measurements, again independent of thickness for 200 Å $< d_{Cu} <$ 10 000 Å. Therefore, it seems to us that assuming σ independent of thickness is the best way to analyze our results. It turns out (Table II) that one then obtains an average value for $\sigma=0.3$, in very good agreement with the above estimates.

TABLE III. Slope of σ as a function of F for different values of λ_N (from a least-mean-squares fit of the values in Table II).

λ_N	α
0.05	−0.093
0.02	−0.012
0.01	0.0096
0	0.029
−0.01	0.039
−0.02	0.056
−0.05	0.087

IV. CONCLUSIONS

An expression for T_c of a proximity-effect sandwich in the Cooper limit was derived. The penetration probability of the barrier between N and S was accounted for by generalized boundary conditions at the interface. These introduce an effective length $L \sim l/\sigma$ which is related to L_N of McMillan's tunneling model.[3]

We have derived a condition, Eq. (27), under which T_c approaches the usual result T_{DG} which does not depend explicitly on the normal-metal thickness. It was found that thin Pb-Cu samples do not satisfy this condition, and the transition temperature is quite higher than the one predicted by de Gennes.

If one accepts that σ varies only randomly among the samples, a best fit to the data is obtained for $\lambda_N=0.015$.

It appears that a fairly exact knowledge of σ is needed to calculate λ_N. This can be extracted from tunneling density-of-states measurements.[3,5] However, our procedure may yield information about the sign of λ_N in other cases, e.g., when an assumed value of positive or zero λ_N results with a σ larger than 1. We hope to follow this point for Pb-Ag sandwiches.

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‡Permanent address: Department of Physics and Astronomy, Tel-Aviv University, Tel Aviv, Israel.

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