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The superconducting proximity effect as a tool to investigate metal films and interfaces

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Abstract. The superconducting proximity effect is measured in sandwiches of thin Pb films and the alkali metals Cs, Rb, K and Na. The T_c-dependence provides information about the interface barriers between Pb and the alkalis. Such a barrier is particularly large in Pb/Cs sandwiches. It is not due to impurities or oxydation. In the presence of a sufficiently strong barrier a special form of the Cooper limit can be applied to calculate the transition temperature of the sandwich.

PACS. 74.50.+r Tunneling phenomena; point contacts, weak links, Josephson effects – 73.20.-r Electron states at surfaces and interfaces – 71.20.Dg Alkali and alkaline earth metals

1 Introduction

When a thin superconducting film is covered with a normal conductor its transition temperature is lowered. This phenomenum is known as the "Superconducting Proximity Effect" (SPE), and it was in tensively studied in the 1960s and 70s [1–7] (see for example [8]). (In those days one had the hope that an extrapolation of T_c would yield a finite transition temperature for normal conductors such as the noble metals.) However, there has been a continuous interest in this effect over the years [9–13] which extended recently into superconductor-ferromagnetic metal sandwiches (see for example [14]). In the present paper we want to revive the SPE as a tool to investigate interfaces between metal films.

In recent years we have investigated the properties of thin alkali metal films [15–23]. We observed a number of properties which were quite unexpected:

- The Hall effect and resistance of thin Cs increased dramatically when the film was covered in situ with small concentrations of (s, p) impurities.
- Sandwiches of quench-condensed Fe covered with a film of Cs or K showed a mean free path in the alkali film which can be up to five times the thickness of the alkali film. This means that the electrons in the Cs or K are almost perfectly specularly reflected at the free surface and the interface with the Fe. The latter is quite surprizing since the Fe is very disordered and the electrons should be diffusively scattered at the interface.
- Recently we used the SPE in a Pb/K/Pb sandwich to study the local electronic properties of the K while it was coveraged with sub-mono-layers Pb. On the one hand the coverage of the K with Pb appeared to localize the electrons in the K while on the other hand the T_c-reduction of the first Pb film by the K remained unaltered.

The properties of interfaces and the ability of electrons to cross them are important in many physical phenomena and applications. To name one example for the latter, the giant magneto-resistance in magnetic multi-layers depends critically on the ability of the conduction electrons to cross "freely" from one film to the next. The goal of this paper is to investigate the SPE in Pb/Ak films (in the following we use the symbol "Ak" as synonym for any of the four alkali metals Cs, Rb, K and Na) and to show that the SPE is a suitable tool to obtain information about interfaces.

2 Experiment

In the present investigation we prepare sandwiches of Pb/Ak at liquid helium temperatures. In most experiments we quench condense a Pb film of 13 to 14 nm thickness and a resistance of about 100 Ω. The Pb is then covered in several steps with an alkali metal. After each evaporation the film or sandwich is annealed, the original Pb film up to 40 K and the sandwiches up to 35 K. Then the superconducting transition curve is recorded and the magneto-resistance and Hall resistance are measured at 9.5 K in the field range between –7 T and +7 T. In these experiments one has to be careful that the edges of the

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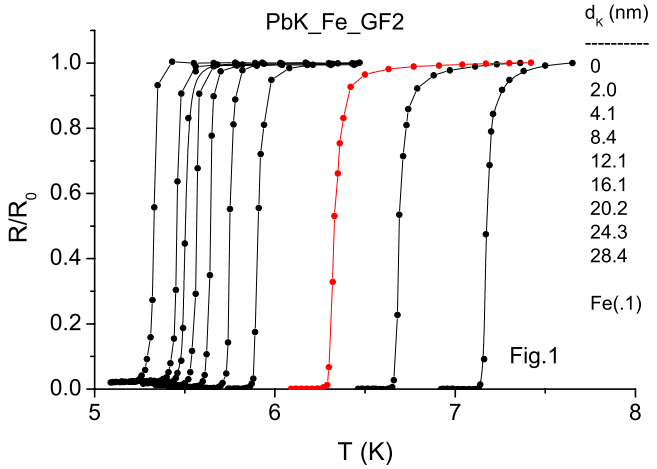

Fig. 1. The superconducting transition curves for Pb with different coverages of K.

Pb film are covered by the normal metal. Otherwise one obtains double transition curves. Figure 1 shows a set of transition curves for Pb/K sandwiches.

In Figures 2a–d the dependence of T_c on the coverage with Cs, Rb, K and Na is plotted. The full circles represent the experimentally measured T_c. The curves are discussed below.

3 Theory and discussion

Werthamer [3] derived an implicit set of equations for the transition temperature of a sandwich of two superconductors. The superconductor with the lower transition temperature is generally called the normal conductor because at the T_c of the sandwich it would be in the normal conducting state. In this theory the gap function $\Delta(\mathbf{r})$ in the superconductor is proportional to $\cos[k_s(z+d_s)]$ and in the normal conductor proportional to $\cosh[k_n(z-d_n)]$, with $z=0$ at the interface. The $k_{s,n}$ are the inverse superconducting coherence lengths. For disordered (dirty) metal films they are given by

$$\ln\left(\frac{T_s}{T_c}\right) = \chi\left(\xi_s^2 k_s^2\right),\ \ln\left(\frac{T_c}{T_n}\right) = -\chi\left(-\xi_n^2 k_n^2\right)$$
$$\chi(z) = \Psi\left(\frac{1}{2}+\frac{1}{2}z\right) - \Psi\left(\frac{1}{2}\right)$$
$$\xi^2 = \frac{\hbar v_F l}{6\pi k_B T_c} = \frac{\pi\hbar k_B}{6e^2 T_c}\frac{\sigma}{\gamma} = D\tau_T$$
$$\tau_T = \frac{\hbar}{2\pi k_B T_c}. \qquad (3.1)$$

Here T_s, T_n and T_c are the transition temperatures of the two superconductors ($T_s > T_n$) and the sandwich, $\xi_{s,n}$ are the thermal coherence lengths, σ the conductivity, γ the Sommerfeld constant (which stands for the density of states), D the diffusion constant, v_F the Fermi velocity, τ_T the thermal time, and $\psi(x)$ the digamma function.

At the interface Werthamer originally used the boundary condition that $\frac{1}{\Delta}\frac{d\Delta}{dz}$ is continous. De Gennes [24] derived instead that $\Delta/(NV)$ and $(D/V)(d\Delta/dz)$ are continous at the interface, the latter only in the "dirty" limit. The combined function $(D/V)(d\Delta/dz)/\Delta/(NV) = DN(d\Delta/dz)/\Delta$ is then continous as well. This yields boundary condition

$$N_s\xi_s^2 k_s \tan(k_s d_s) = N_n\xi_n^2 k_n \tanh(k_n d_n) \qquad (3.2)$$

which determines T_c. The equations (3.1, 3.2) together are often called the Wertheimer-deGennes theory. We will abriviate the theory as the WG-theory.

The WG-theory is restricted to the dirty case, where the mean free path of the conduction electrons is much smaller than the superconducting coherence length. It is interesting to note that Werthamer's equations require only three parameter of each superconductor besides the thickness: the transition temperature, the resistivity and the density of states (Sommerfeld constant).

Deutscher and DeGennes [8] point out the calculation for $\Delta(x)$ within each superconductor uses only one root for the decay constant (the smallest Matsubara frequency). Nevertheless Werthamer's equations were successfully applied to a number of experimental results (see for example [8,6]). Therefore it is interesting to compare the experimental results with the WG-theory.

In Figures 2a–d we calculate the transition temperature with the WG-theory for the different Pb/Ak sandwiches. As far as we know the alkali metals are not superconducting. On the other hand the WG-theory requires a finite transition temperature T_n for both metal films. Therefore we treat the alkali metals as hypothetical superconductors with a transition temperature of $T_n = 10^{-5}$ K. (It turns out that the choice of $T_n = 10^{-2}$ K does not make any difference in the theoretical transition temperatures at the experimental thicknesses of the alkali metals. Only in an extremely small thickness range of the alkali metals $d_n \to 0$ does one find a small difference (which, however, influences the initial slope dT_c/dd_n at zero thickness of the alkali metal.) We recognize from Figure 2 that the heavy alkali metals such as Cs show a large deviation from the WG-theory while the sandwich with the light Na is relatively close to the theoretical curve. For Pb/Rb and Pb/K the region of small alkali thickness appears to be reasonably reproduced by the WT.

The initial slope dT_c/dd_n of T_c versus the thickness of the normal metal at $d_n = 0$ can be derived from Werthamer's theory and is given by

$$\frac{d_s}{T_s}\frac{dT_c}{dd_n} = -\frac{\pi^2}{4}\frac{N_n}{N_s}\chi^{-1}\left(-\ln\left(\frac{T_s}{T_n}\right)\right).$$

The expression $\chi^{-1}\left(-\ln\left(\frac{T_s}{T_n}\right)\right)$ approaches the value one for a large (T_s/T_n) ratio, but for $T_n = 10^{-2}$ K (with $T_s = 7.2$ K) one has $\chi^{-1}\left(-\ln\left(7.2/10^{-2}\right)\right) = -0.75$. Since the Werthamer value of the initial slope depends strongly on the choice of T_n while at finite thickness there is no such dependence we prefer to compare the value $\Delta T_c/\Delta d_n$ between experiment and theory. Here Δd_n is the smallest

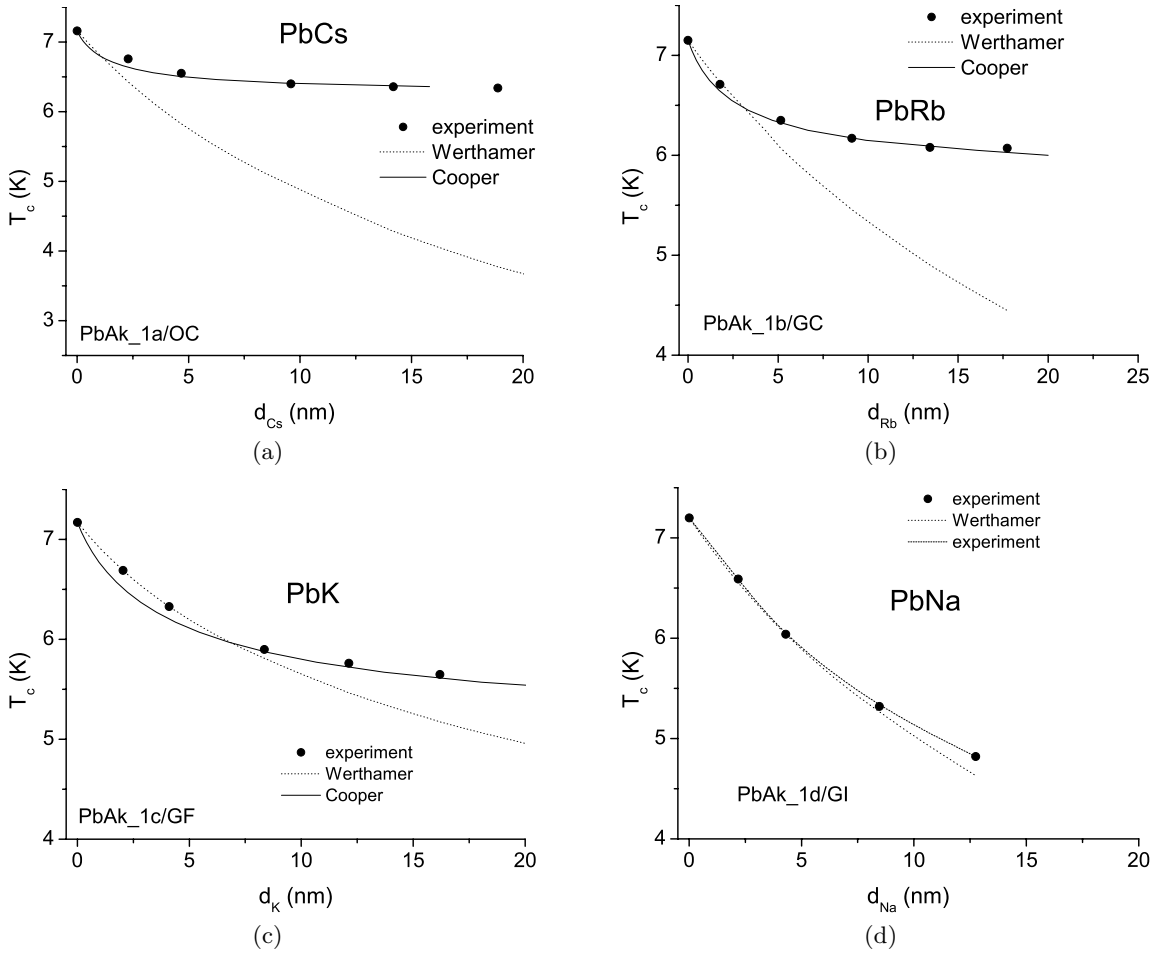

Fig. 2. The transition temperatures of Pb/Ak multilayers as a function of the alkali thickness. The dotted curves are calculated with the theory by Werthamer. For Pb/Cs, Pb/Rb and Pb/K the full curves are calculated within the Cooper model with a barrier between the Pb and the alkali film. For Pb/Na the full curve is a guide to the eye.

evaporated film thickness of the alkali metal and ΔT_c is the T_c-reduction due to this alkali coverage. (We will still call the ratio $\Delta T_c/\Delta d_n$ the initial slope.) These values are collected in Table 1 for the four alkali sandwiches studied. For the alkali metals we used the density of states as given in Ashcroft and Merman [25]. In collumn three the factor N^*/N_0, the ratio between the (experimental) density of states N^* and the free electron value N_0, is given. For the very disordered Pb we used a density ratio of 2.5 because disordered Pb has an even higher gap ratio $2\Delta_0/k_BT_c = 4.6$ than that of pure Pb and therefore a larger value of $(1+\lambda)$ than pure (annealed) Pb, where $(1+\lambda)$ is the electron-phonon enhancement factor.

The initial slope does not depend on the mean free path in either metal. The same is true for the small values of Δd_n. In Werthamer's theory it only depends on the ratio of the density of states. For the Pb sandwiched with Na, K and Rb the experimental initial slope agrees within 15% with the prediction of the WG-theory. For Cs the experimental initial slope is much smaller than Werthamer's prediction. We believe that this is caused by some kind of barrier at the Pb-Cs interface.

From the experimental results the deviation between experiment and WG-theory for the Pb/Cs sandwich is particularly striking. One might object that the small reduction in T_c is caused by an oxide layer between the Pb and the Cs. We exclude this interpretation for several reasons: (i) The results have been reproduced in several experiments, (ii) the other alkali metal such as Na and K would show an even stronger tendency to oxidation.

To investigate this question we prepare a sandwich in which the Pb is first covered with 2 nm of Na and afterwards with Cs of increasing thickness. In Figure 3 the transition temperature of the Pb/Na/Cs sandwich is plotted versus the alkali thickness $d_{\mathrm{Na}} + d_{\mathrm{Cs}}$. For comparison the T_c-dependence of the Pb/Cs and the Pb/Na sandwiches are shown in the same figure. One realizes that the transition temperature of the Pb/Na/Cs sandwich is much closer to the Pb/Na sandwich than to the Pb/Cs sandwich. This demonstrates that, after bridging the contact between the Pb and the Cs, one finds a similar T_c-reduction as in the Pb/Na sandwich. It is not the electronic properties of the Cs which yield the small initial slope and the weak reduction of T_c in the Pb/Cs sandwich.

Table 1. The experimental value of $\frac{\Delta T_c}{\Delta d_n}$ and the prediction according to Werthamer's theory for the different alkali sandwiches.

Alkali metal	exper. code	$\frac{N^*}{N_0}$	d_n (nm)	$\left(\frac{\Delta T_c}{\Delta d_n}\right)_{\rm exp}$	$\left(\frac{\Delta T_c}{\Delta d_n}\right)_{\rm Wh}$	$\frac{\tau_{T_c}}{\tau_f}$
Na	GI	1.3	2.18	0.280	0.32	—
K	GF	1.2	2.04	0.235	0.27	0.08
Rb	GC	1.3	1.77	0.249	0.25	0.052
Cs	OC	1.5	2.29	0.175	0.31	0.035

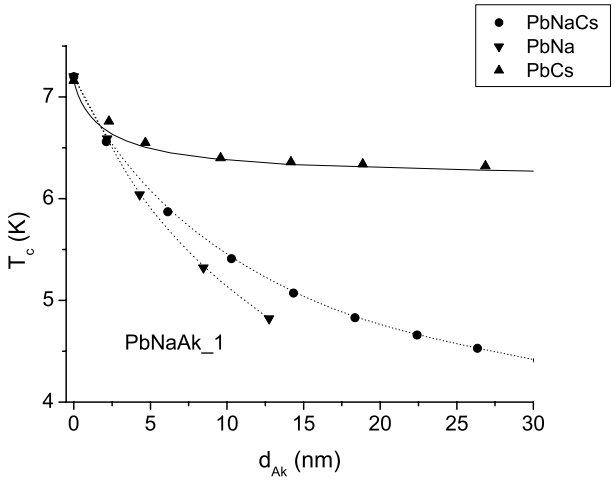

Fig. 3. The transition temperatures of Pb/Na/Cs multilayers as a function of the alkali thickness (full circles). The transition temperatures of the Pb/Cs sandwiches (up triangles) and the Pb/Na sandwiches (down triangles) are shown for comparison. The full curve for the PbCs is calculated in the Cooper model with a barrier between the Pb and Cs films.

Instead the interface between Pb and Cs must create some kind of obstacle which makes it harder for the electrons to cross the interface. About the nature of this obstacles one can only speculate at the present time. However, we are quite sure that it is not due to dirt or oxygen. And neither the Pb nor the Cs are individually responsible for the obstacle because in the Pb/Na/Cs sandwich the more aggressive Na faces each of the two and the obstacle (at each interface) is either absent or much smaller than in the Pb/Cs interface.

If one compares the experimental T_s curves for Pb with Cs, Rb, K and Na with the theory of Wertheimer one recognizes right away that the deviation between experiment and the Wertheimer theory decreases in going from Cs to Na. It is not obvious that the Wertheimer theory has to be correct for our Pb/Ak sandwiches; in fact this theory may be not appropriate at all for the alkali metals. But the Pb/Na/Cs sandwich demonstrates for the Pb/Cs case that the deviation is not due to the Cs but the interface. Therefore it is suggestive that all the interfaces Pb/Ak present some kind of obstacles for the transmission of the electrons through the interface, where the strength of the obstacle decreases in going from Pb/Cs to Pb/Na. This does not contradict the fact the experimental initial slopes for the three alkali metals agree quite well with the Wertheimer theory. This we demonstrate for the Cooper limit of the SPE.

The Cooper limit applies when the gap function $\Delta(\mathbf{r})$ can be treated as constant in each metal of the film. This is fulfilled when the thickness of each metal is much smaller than its coherence length. If the exchange of electrons between the superconductor and the normal conductor is strongly reduced then the Cooper limit is valid in a considerably larger thickness range. As discussed in Appendix A one can formulate the following condition: Mark all electrons in a small energy range about the Fermi surface in the superconductors. Follow their density distribution as a function of time while they can propagate into the normal conductor. If their density is at all times reasonably constant within the superconductor, then the gap function $\Delta(\mathbf{r})$ will be a constant Δ_s (at the transition temperature) and the Cooper case applies. This can, for example, happen for relatively thick films if the escape time from the superconductor into the normal conductor is long.

In Appendix A we derive the Cooper limit with a barrier between the two metals from a special version of the linear gap equation. The theory requires one fitting parameter, the transmission rate $1/\tau_{sn}$ from the Pb into the alkali film. The resulting theoretical curves are plotted in Figures 2a, b, c as full curves and the fitted rates τ_{T_c}/τ_t are collected in Table 1. They describe the behavior of T_c at finite thickness d_n of the alkali metal films quite well, in particular the saturation in the Pb/Cs sandwich. Surprisingly the WG-theory gives a better fit for the sandwiches with Rb, K and Na at very small alkali thicknesses.

For low transmission rate $1/\tau_{sn}$ through the interface and large normal conductor thickness the electrons of the superconductor escape with a rate of $1/\tau_{sn}$ from the superconductor. This acts as a pair-weakening rate of $1/\tau_{sn}$ and reduces the transition temperature as

$$\frac{\Delta T_c}{T_s} = \frac{\pi^2}{2}\frac{\hbar}{2\pi k_B T_c}\frac{1}{\tau_{ns}}.$$

Ashida et al. [26] calculated the transition of superconductor – normal conductor sandwiches with barriers in between. They described the strength of the barrier by the coefficient of reflectivity R at the interface. (In the absence of a barrier the value of R is not zero but given by density of states ratio). They find in the limit $(R-1) \ll 1$ for the transition temperature

$$\frac{\Delta T_c}{T_s} = \frac{\pi^2}{16}\frac{\hbar}{2\pi k_B T_c}\frac{(1-R)\,v_s}{d_s}.$$

The two result are compared in Appendix A.

A full numerical solution of the linear gap equation (A.4) with an adjustable transmission rate between the Pb and the alkali film would be desirable. Presently we are developing the software for such a solution.

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Appendix A

In close vicinity of the transition temperature the superconducting gap function $\Delta(\mathbf{r})$ is very small and the "gap equation" can be linearized [27]

$$\Delta(\mathbf{r}) = V(\mathbf{r}) \int d^3\mathbf{r}' \frac{1}{\tau_T} \sum_\omega H_\omega(\mathbf{r},\mathbf{r}') \Delta(\mathbf{r}') \quad \text{(A.1)}$$

$$H_\omega(\mathbf{r},\mathbf{r}') = G^*_\omega(\mathbf{r},\mathbf{r}') G_\omega(\mathbf{r},\mathbf{r}') \quad \text{(A.2)}$$

$$\frac{1}{\tau_T} = \frac{2\pi k_B T}{\hbar}. \quad \text{(A.3)}$$

Here $\Delta(\mathbf{r})$ is the gap function at the position $\mathbf{r}$, $\omega_n = (n+1/2)/\tau_T$ are the Matsubara frequencies, $N(\mathbf{r}')$ is the (BCS)-density of states for one spin direction, $V(\mathbf{r})$ is the effective electron-electron interaction at position $\mathbf{r}$. The function $H_\omega(\mathbf{r},\mathbf{r}')$ is the product of the two single electron Green functions $G_\omega(\mathbf{r},\mathbf{r}')$ of a Cooperon. Following de Gennes [8,24], Lueders [28] and one of the authors [29] we use a different approach to solve the gap equation. Since the G_w represent the amplitude of an electron traveling (at finite temperature) from $\mathbf{r}'$ to $\mathbf{r}$ the product $G^*_\omega(\mathbf{r},\mathbf{r}') G_\omega(\mathbf{r},\mathbf{r}')$ describes the pair amplitude of the Cooperon to travel from $\mathbf{r}'$ to $\mathbf{r}$. Since the two G_w are conjugate complex to each other and are independent of the spin direction, the pair amplitude is identical to the probability of a single electron to travel from $\mathbf{r}'$ to $\mathbf{r}$. The function $H_\omega(\mathbf{r},\mathbf{r}')$ can be expressed by the function $F(\mathbf{r},0;\mathbf{r}',t')$ which gives the probability of an electron to travel from $\mathbf{r}'$ to $\mathbf{r}$ during the time interval $|t'|$ (departing at $\mathbf{r}'$ at the negative time t' and arriving at $\mathbf{r}$ at the time $t=0$) while it experiences an exponential damping of $\exp(2|\omega|t')$

$$H_\omega(\mathbf{r},\mathbf{r}') = \int_{-\infty}^{0} dt' e^{2|\omega|t'} F(\mathbf{r},0;\mathbf{r}',t') N(\mathbf{r}').$$

This yields the gap equation

$$\Delta(\mathbf{r}) = V(\mathbf{r}) \int d^3\mathbf{r}' \int_{-\infty}^{0} \frac{dt'}{\tau_T} \sum_\omega e^{2|\omega|t'} F(\mathbf{r},0;\mathbf{r}',t') N(\mathbf{r}') \Delta(\mathbf{r}'). \quad \text{(A.4)}$$

The function $\sum_\omega e^{-2|\omega|t}$ describes the exponentially decaying coherence of the Cooperons.

This equation has a very transparent interpretation. From a given position $\mathbf{r}'$ and at a given time $t'<0$ there are $N(\mathbf{r}')\Delta(\mathbf{r}')$ electrons (representing the pair amplitude) propagating into all directions of the metal and experiencing scattering. Along the way their number decays exponentially as $\sum_\omega e^{-2|\omega|t}$. At a given time, for example at $t=0$ and at each position $\mathbf{r}$ one sums the contribution of all surviving electrons from all $(\mathbf{r}',t')$, forming the integral $\int d\mathbf{r}' \int_{-\infty}^{0} dt'$ and multiplies the result with $V(\mathbf{r})$. The result has to reproduce self-consistently everywhere the gap function $\Delta(\mathbf{r})$.

If one is dealing with a time dependent gap function the derivation of a time-dependent Ginzburg-Landau equation from $(A.4)$ is straight forward by replacing the time 0 by t and $\Delta(\mathbf{r}), \Delta(\mathbf{r}')$ by $\Delta(\mathbf{r},t), \Delta(\mathbf{r}',t')$ [29].

Since we are here not interested in time-dependent gap functions we may shift the time scale by starting the propagation at $\mathbf{r}'$ at the time $t=0$ and arriving at $\mathbf{r}$ at the time $t>0$. Then we have

$$\Delta(\mathbf{r}) = V(\mathbf{r}) \int d^3\mathbf{r}' \int_0^{\infty} \frac{dt'}{\tau_T} \sum_\omega e^{-2|\omega|t} F(\mathbf{r},t;\mathbf{r}',0) N(\mathbf{r}') \Delta(\mathbf{r}'). \quad \text{(A.5)}$$

The great advantage of this description is the fact that a major part of solving the gap equation involves only the dynamics of the conduction electrons. We demonstrate this first for the Cooper limit of an SN-sandwich.

Cooper case without barrier: We assume that the electron-electron interaction in the normal metal is zero $V_n = 0$. In this case the gap function is only non-zero in the superconductor and both $\mathbf{r}'$ and $\mathbf{r}$ lie in the superconducting film. Since in the Cooper limit $\Delta(\mathbf{r}')$ is constant and has the value Δ_s in the superconducting film one obtains for the gap equation

$$\Delta_s = V_s \int_S d^3\mathbf{r}' \int_0^{\infty} \frac{dt}{\tau_T} \sum_\omega e^{-2|\omega|t} F(\mathbf{r},t;\mathbf{r}',0) N_s \Delta_s.$$

The integration $d\mathbf{r}'$ extends only over the superconducting film. The electron propagation function $F(\mathbf{r},t;\mathbf{r}',0)$ describes the probability of $N(\mathbf{r}')\Delta_s$ electrons which start at $(\mathbf{r}',0)$ (in the superconductor) and arrive at t at the position $\mathbf{r}$ (in S). (Although the start and end points of the electron path lie in the superconductor the path can extend into the normal conductor as well.) Since the electrons propagate roughly with the Fermi velocity perpendicular to the film plane the time to move the distance d_s or d_n is very short compared with the thermal coherence time $\tau_T = \hbar/(2\pi k_B T)$. Therefore any electron – independently of where it started – will be found, after a very short time, in the superconductor with the probability $p_s = \frac{d_s N_s}{d_s N_s + d_n N_n}$. Therefore one obtains $\int_S d^3\mathbf{r}' F(\mathbf{r},t;\mathbf{r}',0) = \frac{d_s N_s}{d_s N_s + d_n N_n}$ for (almost) any time t.

This yields the equation

$$\Delta_s = V_s N_s \frac{d_s N_s}{d_s N_s + d_n N_n} \int_0^\infty \frac{dt'}{\tau_T} \sum_\omega e^{2|\omega|t'} \Delta_s$$

$$1 = V_s N_s \frac{d_s N_s}{d_s N_s + d_n N_n} \frac{1}{\tau_T} \sum \frac{1}{2\,|\omega|} \tag{A.6}$$

$$\text{with } \frac{1}{\tau_T} \sum \frac{1}{2\,|\omega|} = \sum_{n=0}^{n_c} \frac{1}{n+\frac{1}{2}}.$$

$$n_c = \frac{\Theta_D}{2\pi T_c},\ n_{c0} = \frac{\Theta_D}{2\pi T_s}$$

The equation (A.6) is the well known Cooper condition for the transition temperature of the sandwich where the attractive interaction is given by $V_{eff} = V_s \frac{d_s N_s}{d_s N_s + d_n N_n}$, resulting in the transition temperature $T_c = 1.14\Theta_D \exp\left[-1/N_s V_{eff}\right]$. For the initial slope in the Cooper limit one obtains then

$$\frac{d_s}{T_s}\frac{dT_c}{dd_n} = -\frac{1}{V_s N_s}\frac{N_n}{N_s}.$$

It should be pointed out that the initial slope in the Cooper limit does not exactly agree with the result by Werthamer, emphasizing that Werthamer's solution contains approximations.

Cooper case with barrier: In the next step we consider the Cooper case with a barrier between the superconducting and the normal metal film. An electron in S has a finite transmission rate through the interface which is proportional to the density of states in the normal conductor N_n and inversely proportional to the thickness of the superconductor d_s.

$$\frac{1}{\tau_{sn}} = \alpha \frac{1}{d_s} N_n.$$

We follow the fate of such an electron, denoting the probability to be in S or N as n_s and n_n, where $(n_s + n_n) = 1$. Then we have

$$\frac{dn_s}{dt} = -\alpha n_s \frac{1}{d_s} N_n + \alpha n_n \frac{1}{d_n} N_s$$

$$\frac{dn_n}{dt} = \alpha n_s \frac{1}{d_s} N_n - \alpha n_n \frac{1}{d_n} N_s.$$

The solution of this equation is

$$n_s = n_\infty + n_\Delta \exp\left(-\frac{t}{\tau_r}\right) \tag{A.7}$$

$$\text{with } n_\infty = \frac{N_s d_s}{(N_n d_n + N_s d_s)},\ n_\Delta = \frac{d_n N_n}{(N_n d_n + N_s d_s)}$$

$$\frac{1}{\tau_r} = \alpha \left(\frac{N_n}{d_s} + \frac{N_s}{d_n}\right) = \frac{1}{\tau_{sn}} + \frac{1}{\tau_{ns}}$$

where τ_r^{-1} is the Cooperon relaxation rate. It is the sum of the transmission rates through the interface in both directions. This yields the following gap equation

$$\Delta_s = V_s N_s \int_0^\infty \frac{dt}{\tau_T} \sum_\omega \left(n_\infty e^{-2|\omega|t} + n_\Delta e^{-\left(2|\omega| + \frac{1}{\tau_r}\right)t}\right) \Delta_s$$

or

$$1 = \frac{V_s N_s}{d_s N_s + d_n N_n} \times \left(d_s N_s \sum_{n=0}^{n_c} \frac{1}{\left(n+\frac{1}{2}\right)} + d_n N_n \sum_{n=0}^{n_c} \frac{1}{\left(n + \frac{1}{2} + \frac{1}{2}\frac{\tau_T}{\tau_r}\right)}\right) \tag{A.8}$$

This equation shows already qualitatively the effect of the barrier for the two limiting cases:

- weak transmission, $\tau_T/\tau_r < 1$: In this case the sum in the second term on the right side of equation (A.8) is almost the sum as the first sum. One obtains almost the transition temperature of the pure superconductor. T_c is given by the implicit equation

$$1 = V_s N_s \sum_{n=0}^{n_c} \frac{1}{n+\frac{1}{2}} - V_s N_s \frac{d_n N_n}{(N_n d_n + N_s d_s)} \left[\psi\left(\frac{1}{2} + \frac{1}{2}\frac{\tau_T}{\tau_r}\right) - \psi\left(\frac{1}{2}\right)\right]$$

- strong transmission, $\tau_T/\tau_r > 1$: In this case one obtains

$$1 = V_s N_s \frac{d_s N_s}{(N_n d_n + N_s d_s)} \sum_{n=0}^{n_c} \frac{1}{n+\frac{1}{2}} + \frac{d_n N_n}{(N_n d_n + N_s d_s)} \sum_{n=0}^{n_c} \frac{1}{n + \frac{1}{2} + \frac{1}{2}\frac{\tau_T}{\tau_r}}$$

  where the second term on the right side can be neglected for large τ_T/τ_r and the remaining part yields just the Cooper transition temperature without a barrier.

We perform the numeric solution of equation (A.8) for the Pb/Cs sandwich. For this purpose one has to solve the equation

$$d_s N_s \left(\sum_{n=0}^{n_c} \frac{1}{\left(n+\frac{1}{2}\right)} - \frac{1}{V_s N_s}\right) + d_n N_n \left(\sum_{n=0}^{n_c} \frac{1}{\left(n + \frac{1}{2} + \frac{1}{2}\frac{\tau_T}{\tau_r}\right)} - \frac{1}{V_s N_s}\right) = 0 \tag{A.9}$$

with

$$n_c = \frac{\Theta_D}{2\pi T_c},\ n_{c0} = \frac{\Theta_D}{2\pi T_s} \tag{A.10}$$

$$\rho = \frac{\tau_T}{\tau_r} = \frac{\tau_T}{\tau_{sn}}\left(1 + \frac{d_s N_s}{d_n N_n}\right)$$

$$\frac{1}{V_s N_s} = \sum_{n=0}^{n_{c0}} \frac{1}{\left(n+\frac{1}{2}\right)}.$$

The resulting T_c-values are plotted in Figures 2a and 3 as full curves. For the transmission rate we used the value $1/\tau_{sn} = 0.035/\tau_{T_s} = 0.035 \times 2\pi k_B T_s/\hbar = 2.0 \times 10^{11}$ s^{-1} or $\tau_{sn} = 5 \times 10^{-12}$ s. ($1/\tau_{sn}$ is transmission rate for an electron in the Pb film to escape into the Cs film). It is much longer than the superconducting time constant $\tau_{T_s} = \hbar/(2\pi k_B T_s) = 1.7 \times 10^{-13}$ s and the ballistic flight time of the Pb electrons with (normalized) Fermi velocity (in z-direction): $\tau_b = 2d/v_F^* = 4 \times 10^{-14}$ s.

Similar evaluations are performed for the Pb/Rb and Pb/K sandwiches. The transmission rate ratios τ_{T_s}/τ_{sn} are included in Table 1. For the Pb/Na sandwich the agreement was not sufficient to include it in the Figure 2d. All the theoretical curves for the Cooper case with barrier yield a too large initial slope. For small thicknesses of the normal conductor the relaxation rate becomes arbitrarily large according to relation (A.7) This is unphysical, however, because the electron needs at least the ballistic flight time τ_b to cross over from the superconductor into the normal conductor. Therefore this range has to be excluded from the evaluation. Going from Pb/Cs to Pb/Na the alkali thickness range in which the barrier approach is not appropriate increases.

Ashida et al. [26] calculated the transition temperature of SN-sandwiches with barriers in-between. They characterize the strength of the barrier by the coefficient of reflectivity R at the interface. (In the absence of a barrier the value of R is not zero but given by density of states ratio.) They find in the limit $(R-1) \ll 1$ for the transition temperature (in our notation)

$$\frac{\Delta T_c}{T_s} = \frac{\pi^2}{16} \frac{\hbar}{2\pi k_B T_c} \frac{(1-R)\, v_s}{d_s}.$$

This limit $(R-1) \ll 1$ corresponds to in our notation to the limit $1/\tau_{sn} \ll 1/\tau_T$. If we follow Ashida et al. and expand A.9 in terms of the pair breaking parameter ρ we obtain

$$\sum_{n=0}^{n_c} \frac{1}{\left(n+\frac{1}{2}+\frac{1}{2}\rho\right)} \approx \sum_{n=0}^{n_c} \frac{1}{n+\frac{1}{2}} - \frac{\rho}{2} * \frac{1}{2}\pi^2$$

$$\left(\frac{d_n N_n}{d_s N_s}\right)^{-1} = -1 + \frac{\frac{\rho}{2} * \frac{1}{2}\pi^2}{\left(\sum_{n=0}^{n_c} \frac{1}{\left(n+\frac{1}{2}\right)} - \frac{1}{V_s N_s}\right)} \tag{A.11}$$

which yields

$$\ln\left(\frac{T_s}{T_n}\right) = \frac{\pi^2}{4} \frac{\tau_T}{\tau_{sn}}$$

or

$$\frac{\Delta T_c}{T_s} = \frac{\pi^2}{4} \frac{\hbar}{2\pi k_B T_c} \frac{1}{\tau_{sn}}.$$

The calculation of $\overline{1/\tau_{sn}}$ in terms of R is straight forward

$$\overline{\frac{1}{\tau_{sn}}} = \frac{\int_0^1 \frac{v_F \cos\theta}{2d_s} (1-R)\, d(\cos\theta)}{\int_0^1 d(\cos\theta)} = \frac{v_F\,(1-R)}{4d_s}.$$

This expansion yields a perfect agreement between the results of Ashida et al. and ours. However, the result is somewhat surprising since the T_c does not depend on the thickness of the normal conductor. Our numerical evaluation shows that this is not quite correct. The reason is the following.

The value of $\rho = \frac{\tau_T}{\tau_r} = \frac{\tau_T}{\tau_{sn}} \left(1 + \frac{d_s N_s}{d_n N_n}\right)$ diverges for small thickness of the normal conductor, always reaching the limit of strong transmission for $d_n \to 0$. That means that at small thickness of the normal conductor one expects the full Cooper reduction of the transition temperature. This means in other words that even for $(R-1) \ll 1$ the expansion by Ashida et al. has to exclude the range of very small normal conductor thickness. When d_n increases the transmission into the normal conductor becomes less effective, the superconducting film behaves more isolated and the T_c-reduction is much less effective. This is the experimental observation, in particular for the Pb/Cs sandwich.

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