Proximity effect in ultrathin Pb/Ag multilayers within the Cooper limit

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We report on transport and tunneling measurements performed on ultrathin Pb/Ag (strong-coupled superconductor/normal metal) multilayers evaporated by quench condensation. The critical temperature and energy gap of the heterostructures oscillate with addition of each layer, demonstrating the validity of the Cooper limit model in the case of multilayers. We observe excellent agreement with a simple theory for samples with layer thickness larger than 30 Å. Samples with single layers thinner than 30 Å deviate from the Cooper limit theory. We suggest that this is due to the ''inverse proximity effect'' where the normal-metal electrons improve screening in the superconducting ultrathin layer and thus enhance the critical temperature.

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The superconducting proximity effect is a well-known phenomenon and has drawn a lot of interest both from the fundamental and the practical points of view. In a high transmission contact between a superconductor and a normal metal the superconducting wave function varies smoothly across the interface causing a suppression of the pair amplitude in the superconductor and an enhancement of pairing on the normal side. The characteristic distance in which superconductivity ''leaks'' into the normal region is the normal-state coherence length $\xi_N=\sqrt{\hbar D/2\pi kT}$, where D is the diffusivity, and the length scale at which superconductivity is suppressed in the superconducting side is the superconducting coherence length ξ_S.

In the Cooper limit[1] (in which both the superconductor and the normal-metal layers are smaller than the characteristic coherence lengths ξ_S and ξ_N, respectively) the behavior of an *S*-*N* bilayer is well described within the framework of the de Gennes model.[2,3] The electrons experience an average pairing interaction (average between the two materials) and T_C can be described within the BCS weak-coupling form adapted to the Cooper limit proximity effect in the following way:

$$kT_C=1.13\hbar\omega_D e^{-1/N(0)V}, \tag{1}$$

$$[N(0)V]_{S+N}=\frac{d_S[N(0)V]_S+d_N[N(0)V]_N}{d_S+d_N},$$

where ω_D is the Debye frequency, $N(0)V$ is the pairing interaction [$N(0)$ being the density of states at the Fermi level and V the pairing interaction], and $d_{S,N}$ is the thickness of the superconducting and normal-metal films, respectively. This model assumes that the Debye frequencies in the two metals are similar and it is based on the understanding that the electrons spend $N_Nd_N/(N_Nd_N+N_Sd_S)$ of their time in the normal metal and $N_Sd_S/(N_Nd_N+N_Sd_S)$ in the superconductor (N_N and N_S are the density of states in the normal metal and in the superconductor, respectively).

The above picture is modified in the case of strong-coupled superconductors where T_C is given by the McMillan expression:[4]

$$T_C=\frac{\omega_D}{1.45}\exp-\left[\frac{1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right], \tag{2}$$

where λ is the dimensionless electron-phonon coupling coefficient and μ^* is the effective Coulomb repulsion. In a normal-metal/strong-coupled superconductor within the Cooper limit, one can expect λ in Eq. (2) to be replaced by an average electron-phonon coupling:[5]

$$\lambda_{S+N}=\frac{\lambda_Sd_S+\lambda_Nd_N}{d_S+d_N}. \tag{3}$$

The proximity effect in the Cooper limit has been observed experimentally in numerous systems of superconductor/normal-metal bilayers.

In this paper, we study systems of multilayers of superconductor/normal metal (Pb/Ag) in which the total thicknesses of the metals are comparable to the relevant coherence lengths. The transport and tunneling experimental results confirm that the basic idea of the Cooper limit notion, i.e., that the electrons experience an average pairing, is valid also in a multilayer sample. We observe good *quantitative* agreement with theory as long as the thickness of the layers is larger than ~30 Å while thinner layers deviate from the theoretical predictions. We suggest that the simple picture is modified in ultrathin layers due to effects of screening of *e*-*e* interactions (the inverse proximity effect as previously reported[6]).

The Pb and Ag layers were prepared by quench condensation, i.e., evaporation on a cryogenically cooled substrate within the measurement apparatus.[7] This allows *in situ* sequential depositions under UHV conditions and simultaneous transport and tunneling measurements on a single sample. This method has several essential advantages for the study of the superconducting proximity effect.[8] Since Pb and Ag are immiscible, the alloying of these two materials is very improbable even at room temperatures. Alloying becomes practicably negligible when the samples are quench condensed and are kept below $T=10$ K throughout the entire experiment (sample growth and measurement). In addition, the extremely clean environment of the experiment leads to very

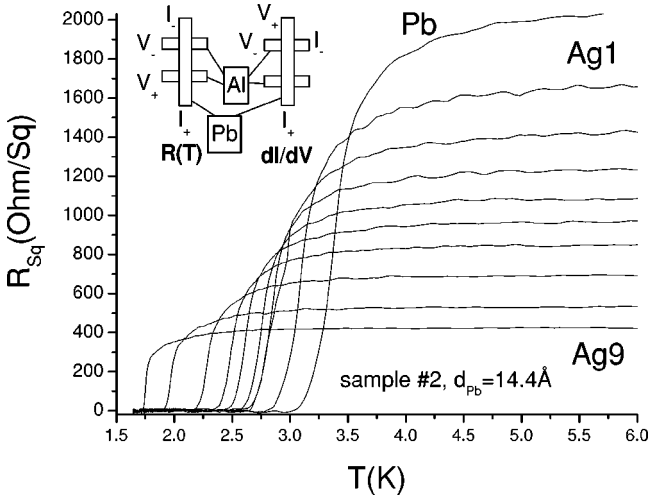

FIG. 1. Resistance per square versus temperature of a Pb/Ag bilayer (sample 2). The thickness of Ag varies from 0 Å (Pb) to 14.4 Å (Ag9). Each evaporation corresponds to $\approx$1.6 Å. The T_C decreases monotonically as normal-state resistance is decreased. Inset: Illustration of the sample geometry.

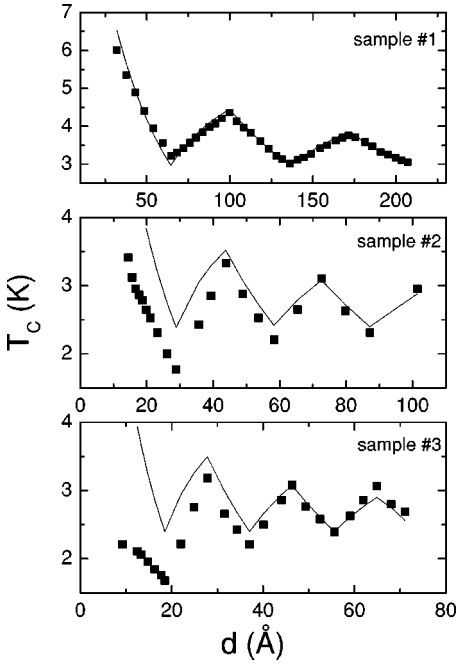

FIG. 2. Superconducting critical temperatures extracted from the $R(T)$ measurements in the case of three different thickness of the Pb/Ag layer. The thicknesses of the Pb and Ag layers in the bilayer are 32.5 Å (top graph), 14.4 Å (middle graph), and 9.3 Å (bottom graph). The solid lines are the theoretical fits using Eqs. (2) and (3).

high quality interfaces between the superconducting layers and the normal-metal layers. Such clean interfaces provide ideal conditions for studying proximity effects.

The geometry of the samples used in this study is shown in the inset of Fig. 1. We begin by evaporating two strips of Al onto a room-temperature polished glass substrate. These strips are used both as voltage leads and as the base electrode for tunneling measurements. Then, we allow a native oxide barrier to grow on the Al for 20 min. We connect metallic leads to the substrate and place it in an evaporation chamber which is then pumped out and immersed in liquid helium. A 10-Å Ge adhesion layer is then quench condensed across the Al strips, permitting the evaporation of a continuous layer of Pb as thin as 5 Å.[7] The thicknesses are monitored by a calibrated quartz crystal situated in the chamber. Multilayers, made of base units of Pb/Ag bilayers having different thicknesses, are quench condensed across the Al strips. This configuration allows the simultaneous measurements of transport properties of the multilayers (thus determining the critical temperature), and tunneling into the multilayers (for determination of the energy gap). We performed these measurements at incremental stages of the evaporation of each of the Pb or Ag layers. All measurement were performed in a screened room using standard ac methods.

In this paper, we present data on three Pb/Ag systems having different base unit thickness: Sample 1 where $d_{\mathrm{Pb}}=d_{\mathrm{Ag}}\approx 32$ Å, sample 2 where $d_{\mathrm{Pb}}=d_{\mathrm{Ag}}\approx 15$ Å, and sample 3 where $d_{\mathrm{Pb}}=d_{\mathrm{Ag}}\approx 9$ Å. Each of these unit bilayers is within the cooper limit; ξ_N of the Ag at low temperatures is of the order of 1 μm and ξ_S of the amorphous Pb (which is in the dirty limit) at low temperature is given by $\xi'=\sqrt{\xi_0 l}$, where ξ_0 is the clean limit coherence length (for Pb this is 800 Å) and l is the mean free path which is approximately an interatomic distance, about 2.7 Å in our samples.[9] These values yield a zero-temperature coherence length, $\xi'\approx 46$ Å, which is larger than each one of our Pb layers. Furthermore, as the temperature increases, the coherence length increases until it diverges at $T=T_C$.

We begin by presenting the results for the initial bilayer. Figure 1 shows the resistance versus temperature of sample 2 as a layer of 14.5-Å-thick Ag is grown in steps of $\approx$1.55 Å on top of a 14.4-Å-thick initial Pb layer. Such measurements are used to extract T_C by taking the midpoint of the transition ($R_N/2$).[10] It is seen that the initial Pb layer has a T_C which is smaller than the bulk value of 7.2 K. This is typical for very thin superconducting layers which are known to exhibit a superconductor–insulator transition as a function of film thickness (and R_{sq}).[11–13] It has been argued[14,20] that as R_{sq} (the disorder) increases, the electronic screening is reduced. Coulomb interactions are then enhanced leading to a weakening of the superconducting coupling. This explains the decrease of T_C in the Pb films when the thickness is reduced from 30 Å to 9 Å as is seen in Fig. 2. The enhancement of Coulomb interactions for high R_{sq} is also evident in Fig. 3 which depicts the (dI/dV)-V measurements (proportional to the density of states) of a 9-Å-thick-Pb film with increasing layers of Ag, taken at temperatures above T_C. It is seen that in the ultrathin Pb layer, the density of states at the Fermi level is relatively low. The tunneling conductance exhibits a strong voltage (energy) dependence demonstrating that this is a strongly Coulomb correlated system. The density of states at the junction interface dramatically increases when layers of Ag are evaporated *on top of the Pb*, presumably due to enhancement of screening and, hence, suppression of the Coulomb interactions.[14]

As subsequent layers of Ag are added (and R_{sq} is reduced

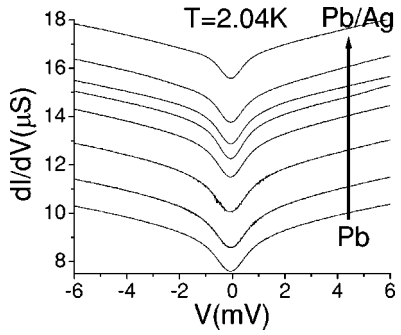

FIG. 3. dI/dV versus V (proportional to the density of states as a function of energy) at temperatures above T_C (2.04 K) of an ultrathin Pb layer (10 Å) with an increasing Ag (up to 10 Å) layer on top.

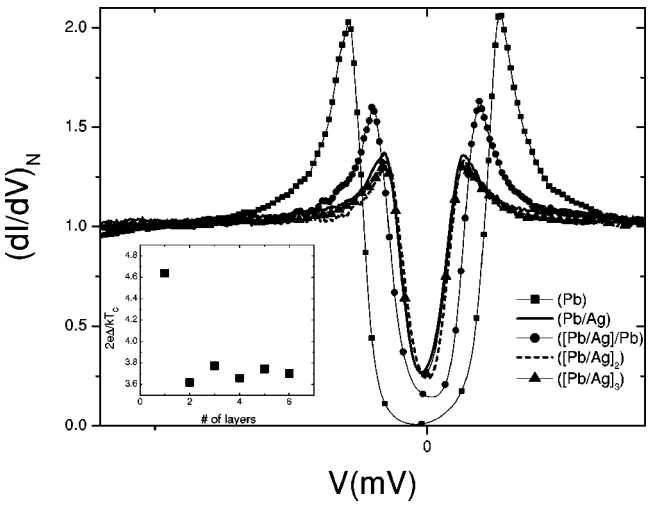

FIG. 4. dI/dV versus V, normalized to the normal-state conductance, for sample 1 taken at $T=1.65$ K for each Pb and Ag sublayer. The oscillations of the gap with thickness are easily observed. The inset shows $2e\Delta/kT_C$ as a function of the number of layers. Note that this ratio is nearly constant for the last few layers.

as $1/d_{\mathrm{Ag}}$) T_C is decreased even further due to the proximity effect as evident in Fig. 1. Since the bilayer is in the Cooper limit, T_C is indeed expected to decrease as a function of the normal-metal thickness. We note, however, that this is not always the case. We have shown[6] that when the initial Pb layer is extremely thin (thinner than that of sample 3) the first added Ag sublayers cause T_C to *increase* in what seems to be an inverse proximity effect. This effect was first observed experimentally by Shapovalov *et al.*[15,16] and was treated theoretically by Finkelstein.[17] Based on tunneling measurements,[6] we interpreted the observations as being due to the fact that the Ag layers enhance screening in the system thus suppressing the *e-e* interactions which are responsible for the T_C suppression.[18] This trend opposes the usual proximity effect and becomes more important in thinner superconducting films.

The evolution of T_C as more Pb and Ag layers are added is shown in Fig. 2 for the three samples. It is seen that T_C oscillates when we change between Pb and Ag depositions reaching a local minimum or maximum at the completion of each layer. The oscillation amplitude decreases with increasing number of bilayers and T_C appears to approach a value of ≈ 3 K in all three samples. The oscillation behavior is also observed in the energy-gap measurements which are illustrated in Fig. 4 for sample 1. The dI/dV curves were measured at $T=1.65$ K, well below the T_C of the multilayer. Note that despite that fact that tunneling is performed into the bottom Pb layer, the results are still affected by adding the sixth layer on top. This clearly demonstrates that within the Cooper limit, the tunneling electrons probe the *entire* sample and the superconducting parameters are determined by the mean properties of *all* the layers in the multilayer. Figure 4 also shows the ratio $2e\Delta/kT_C$ as a function of the number of layers. The superconducting gap was evaluated from a classical temperature-dependent BCS fit. It is seen that this ratio crosses over from a value of 4.7 (which is close to the strong-coupling limit of ~ 4.8) to 3.6 (which is close to the week-coupling limit of ~ 3.5). We would like to point out that the tunneling measurements were performed at relatively low temperature. The fact that T_c and Δ track on each other is further experimental evidence that the Cooper limit is valid here. Hence, the relatively constant value of $2e\Delta/kT_C$ as we add the last few layers gives us further confidence in the validity of our model.

We have attempted to fit the curves in Fig. 2 to the proximity effect modified McMillan expression of Eq. (2) using an average λ for the Cooper limit [Eq. (3)] in the spirit of the de Gennes model for Pb and Ag. These are shown in Fig. 2 where we used the known values for Pb: $\lambda_S=1.55, \omega_D=105$ K, $\mu^*=0.11$,[19] and taking a low electron-phonon coupling for Ag, $\lambda_N=0.2$, without any fitting parameters. It is seen that the experimental results are in excellent agreement with this model for sample 1 (where $d_{\mathrm{Pb}}=d_{\mathrm{Ag}}\approx 32$ Å). We note that the values for λ, ω_D, and μ^* are not adjusted, but accepted values and so the agreement is quite impressive. However, when we used the same parameters for the other samples with thinner base layers, we get substantial deviations from the model for the first few layers (Fig. 2). In fact, in sample 1 (and in the theory) the oscillations are superimposed on a global decrease of T_C, in sample 2 the background is roughly constant and in sample 3 T_C globally *increases* with the number of bilayers. We argue that this behavior is an extension of the inverse proximity effect observed in the ultrathin bilayers.[6,15,16] The large Coulomb interactions characteristics of these thin samples are continuously weakened with increasing numbers of Pb/Ag bilayers, thus, as more layers are added, we approach the Cooper limit proximity effect theory.

In summary, we have shown that the de Gennes considerations regarding the Cooper limit are valid for the case of multilayers until the layers become very thin. Using a simple model, we were able to describe very accurately the variation of the critical temperature and the energy gap as a function of the number of layers in the thick limit. We have shown that for very thin layers the model breaks down indicating that other physics is involved. We suggest that this breakdown is due to the effect of strong Coulomb interactions in

the superconducting ultrathin films. These interactions are screened out as normal metal is added to the superconductor resulting in an inverse proximity effect which must also be taken into account in the thinnest layers.

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[9] This value was calculated from the density of Pb and is also consistent with the conductivity measurements for 1–2 monoatomic thickness of our films.
[10] The relatively broad transition is a consequence of superconductor fluctuations which introduce some uncertainty when evaluating T_C. This can be overcome by fitting the curves using the Aslamazov-Larkin fluctuation theory [L.G. Aslamasov and A.I. Larkin, Phys. Lett. **26A**, 238 (1968)]. Such fits yield T_C's which are very close to the midpoint of the transition as shown in Ref. 6.
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[18] Figure 3 illustrates very clearly that Coulomb interactions play an important role even in the normal state. The addition of Ag on the back of the film in which tunneling is performed affects the background density of states as the Coulomb effects are reduced with additional Ag.
[19] It is well known from a variety of tunneling measurements that the value of μ^* is very similar for all superconductors and ranges from 0.11 to 0.13. See, for example, Ref. 4 and P.B. Allen and R.C. Dynes, Phys. Rev. B **12**, 905 (1975). Note that the McMillan expression is rather insensitive to these slight variations.
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