

Superconductivity in ultrathin quench-condensed Pb/Sb and Pb/Ge multilayers

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(Received 6 February 1995)

We present the results of *in situ* transport and tunneling measurements on quench-condensed Pb/Sb and Pb/Ge multilayers. The superconducting transition temperature (T_c) and the density of states increase concurrently with the number of Pb layers (n) and eventually saturate. Both the variation of T_c with n and the saturated value of T_c can be accounted for by electron transfer between Pb layers through localized states in the Sb(Ge). We find that the basic mechanism for superconductivity in these multilayers is not fundamentally altered from that in homogeneous amorphous superconducting films. Our results clarify the role the Ge or Sb underlayer and overlayer plays in the transport and superconductivity of uniform quench-condensed films.

Superconductor-normal-metal (SN) and superconductor-insulator (SI) multilayers have been under intensive study for more than a decade.¹ These systems exhibit many interesting dimensional effects in their superconducting transition temperature (T_c),^{2,3} critical field (H_c),⁴ and vortex properties,⁵ which may have their parallels in the high- T_c cuprate superconductors. These multilayered structures offer potential insight in the study of the mechanism of high- T_c superconductivity because of the similarity with the layered nature of the cuprates. The multilayer systems are particularly adaptable in that one can artificially control some important physical parameters. The nature of the superconducting layer can be controlled by adjusting its T_c and sheet resistance (R_\square) through the variation of its thickness. The interlayer coupling strength can in turn be controlled by varying the insulating layer thicknesses and by doping it with magnetic or nonmagnetic impurities.

It has been suggested that the high T_c 's in the cuprate superconductors are a result of the Josephson coupling between the CuO_2 layers.^{6,7} Theoretical studies^{7,8} have shown that single-particle hopping between CuO_2 layers is rendered incoherent, thus strongly suppressed, due to the non-Fermi-liquid nature of the in-plane ground state. In other words, it is energetically unfavorable to have a single-particle hopping from one CuO_2 plane to the other because of the strong electronic correlation effects. On the other hand, the Josephson pair tunneling does not experience this so-called orthogonality catastrophe and remains coherent.⁷ It is suggested that the interlayer pair tunneling amplifies the pairing mechanism in each individual layer regardless of its origin, resulting in a much higher T_c .

Strongly interacting electronic behavior is not restricted to the high- T_c compounds. Many low- T_c superconductors such as the heavy fermion materials also exhibit such behavior. An even simpler case which has been extensively studied is the ultrathin homogeneous amorphous films of conventional superconductors.⁹⁻¹² It has been demonstrated that, as the film is made thinner, T_c decreases as a linear function of $1/d$ where d is the

thickness. Associated with this, R_\square increases, and in the region of superconductivity the films are in the weakly localized regime. The superconductivity is suppressed in such a way that T_c and the energy gap Δ_0 decrease concurrently while both remain sharp. Furthermore, the depression of T_c is accompanied by a reduction in the normal state electronic density of states (DOS).¹² These changes are presumably due to the enhancement of the Coulomb interaction.¹³⁻¹⁵ Disorder hinders the screening of the electron-electron interaction, making the dynamical Coulomb interaction long-ranged at finite frequencies. The effect becomes particularly strong in thin films because the screened Coulomb potential diverges with decreasing film thickness. A sufficiently thin disordered film is in fact a strongly correlated electron system, as indicated by a depressed T_c and a strong logarithmic correction to the DOS.¹² Therefore, a multilayered structure consisting of alternate layers of ultrathin disordered superconductor and insulator could be a good model system for the cuprate superconductors. In a cuprate superconductor both the number of CuO_2 planes in a unit cell and the interlayer coupling strength are fixed, making it difficult to study the correlation between these parameters and superconductivity. On the other hand, in SI multilayers T_c can be measured as a function of the number of layers, and the coupling strength between the superconducting layers can be continuously adjusted by changing the thickness of the insulating layer.

In this paper, we present the results of *in situ* transport and tunneling measurements on a series of quench-condensed SI multilayers. The samples fail to show any dramatic increase in T_c with the number of bilayers (n). Generally, T_c increases smoothly with n but eventually saturates. The saturated values of T_c , T_{cs} , are lower than the T_c of the bulk superconductor and are dependent on the insulator thickness. The increase of T_c with n is accompanied by a concurrent increase in the measured DOS. We show that the results can be accounted for exclusively by electron transfer through the insulating layers. We present evidence that it is the high density of localized states in the amorphous insulator that is re-

sponsible for the strong electron diffusion.

The film growth and measurements were performed in a cryogenic evaporator similar to that described earlier.¹⁶ The whole assembly was immersed in liquid He so that all the deposition and measurements were done under UHV. Oxidized Si with preevaporated thin Au/Ge contact pads were used as the substrates. Prior to cooling down the apparatus, a thin Mn-doped Al stripe was evaporated and allowed to oxidized in air for ~ 15 min. The junctions produced this way were usually very high quality SIN tunnel junctions. Pb was used as the superconductor, and both Sb and Ge were used as the insulator, yielding similar results. The evaporation sources were arranged in such a way that they were on a line parallel to the the film stripe to avoid superconducting shorts at the edges. A quartz-crystal oscillator was used to monitor the film thickness.

We began by depositing an 8-Å-thick layer of Sb. It has been shown that extremely thin and homogeneous Pb films can be grown on such a buffer layer.^{9–12} We then deposited a Pb layer followed by another Sb layer (a Pb/Sb bilayer). The film resistance versus temperature was measured as well as the differential conductance of the tunnel junction at various temperatures. The same measurements were repeated each time we put down more Pb/Sb bilayers, until T_c and the tunneling spectrum stopped changing.

In Fig. 1 we present a typical set of transport and tunneling data on a Pb(10 Å)/Sb(10 Å) multilayer. Figure

1(a) shows the evolution of R_{\square} versus T with the number of Pb/Sb bilayers. R_{\square} decreases and T_c increases with n , while the superconducting transition becomes somewhat sharper. As shown in Fig. 1(b), T_c (defined as the temperature at $R_N/2$) increases smoothly with n and saturates at ~ 5.3 K, a value much lower than the bulk Pb T_c of 7.2 K. On the other hand, the normal state conductance of the multilayer increases linearly with n . Namely, the multilayers have the same R_{\square} but lower T_c than a homogeneous Pb film with the same total Pb thickness. A plot of T_c versus R_{\square} lies significantly below that for a pure Pb film. The sharpness of the superconducting transitions correlates with T_c in the same way as in homogeneous Pb films. This fact, coupled with the linearity between the conductance and n , leads us to believe that the multilayers have a well-defined and uniform layer growth. There is little intermixing between Pb and Sb. This conclusion is further supported by the observation of tunneling behavior in *in situ* fabricated Pb/Sb/Pb junctions, which we will discuss later.

Another notable feature in the data is the close correlation between T_c and the normal state DOS. Figure 1(c) shows the normal state tunneling conductance curves taken at around 6.5 K for the same series of multilayers. Because the junction barrier is identical for the same series of multilayers, the changes in the tunnel conductance can be attributed entirely to the variation of the DOS in the multilayers. In Fig. 1(d) we plot the tunnel conductance as a function of $\log_{10} V$. All the curves

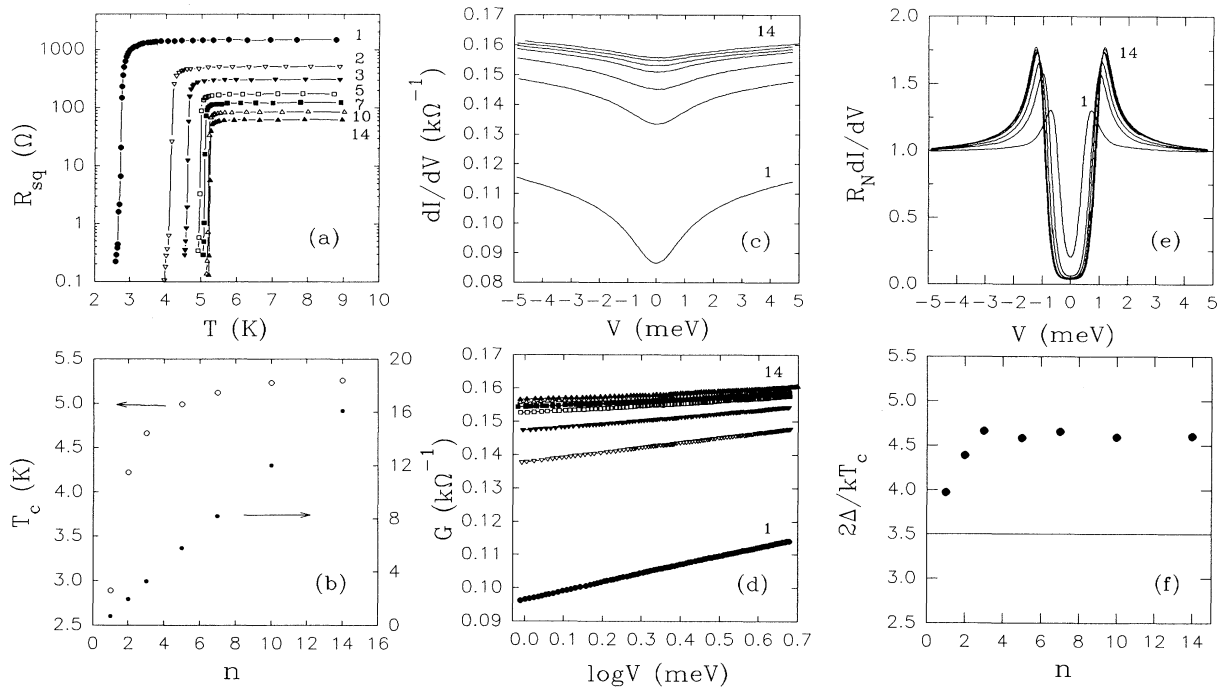


FIG. 1. (a) Sheet resistance as a function of temperature for a series of Pb(10 Å)/Sb(10 Å) multilayers. The numbers are the number of Pb/Sb bilayers. (b) The variation of T_c and conductance with the number of bilayers for the same series. (c) The normal state tunnel conductance for the same series. Note that the junction barrier was identical for the entire series. (d) The normal state tunnel conductance vs $\log_{10} V$. (e) The normalized superconducting tunnel conductance curves for the same series. (f) The BCS constant $2\Delta/kT_c$ for the series.

become linear. The absolute value of the DOS increases with n while the slope decreases. Namely, the corrections to the DOS are logarithmic and the variation in the extent of the corrections coincides with the change in T_c , consistent with the pure Pb case.¹² Since the T_c saturates much below the bulk Pb T_c , we cannot normalize the conductance curves to obtain precisely the extent of the depression in DOS relative to bulk Pb. Thus, a direct comparison is not possible between the DOS of a multilayer and a homogeneous Pb film with the same T_c . However, the correlation between T_c and the DOS closely resembles the pattern for pure Pb films. It indicates that the microscopic mechanisms responsible for the T_c variation in the multilayers and homogeneous Pb films are the same.

Figure 1(e) shows the low temperature ($T < T_c$) tunneling conductance curves for this series normalized by their respective normal state tunneling curves. The normalization eliminates the logarithmic correction and the barrier effects, the result is simply the thermally smeared superconducting DOS,¹⁷

$$N'_s(eV) = \int_{-\infty}^{\infty} N_s(E)[f(E) - f(E + eV)]dE, \quad (1)$$

where $f(E)$ is the Fermi function and $N_s(E)$ is the superconducting DOS at $T = 0$. Since there is little inelastic gap broadening in these uniform films,^{10,12} we can use the standard BCS superconducting DOS, $E/\sqrt{E^2 - \Delta^2}$, as $N_s(E)$. Using Eq. (1) and adjusting Δ we obtain excellent fits to the data. Therefore, the superconducting DOS is still of the BCS form; there is no modification of the DOS by the multilayered structure. Furthermore, the parameter $2\Delta/kT_c$, which is a measure of the electron-phonon coupling strength, remains constant in all samples, just like in homogeneous Pb films.¹⁰

To summarize, the transport and tunneling data on these quench-condensed Pb/Sb(Ge) multilayers are remarkably similar to those on homogeneous Pb. We conclude that the underlying mechanism for superconductivity (or for the suppression of superconductivity) is not fundamentally altered by the multilayered structure. No dramatic enhancement of T_c , as a result of interlayer Josephson coupling, is observed. Yet in the multilayers, the T_c and the DOS increase with the *total* Pb thickness at a much reduced rate compared with homogeneous Pb, and the saturated T_c is lower than the bulk Pb T_c . This dependence on the total Pb thickness implies that the Pb layers are coupled. All these observations can be quantitatively understood by introducing an electron transmission coefficient, t , through the Sb layer. Namely, during the superconducting coherence time, τ_ξ , a fraction (t) of the electrons in each Pb layer will transfer across the Sb barrier. This fraction of electrons sees an effective thickness of $2d_0$, with d_0 being the single Pb layer thickness. Therefore, in terms of superconductivity, a Pb/Sb multilayer with n bilayers is equivalent to a single layer of Pb with a thickness of

$$d_{\text{eff}} = d_0 + td_0 + t^2d_0 + \dots + t^{n-1}d_0. \quad (2)$$

With an appropriate choice of t , the T_c versus d_{eff} curve

should overlap with the T_c versus d curve for homogeneous Pb film. On the other hand, as $n \rightarrow \infty$ in Eq. (2),

$$d_{\text{eff}(s)} = \frac{d_0}{1-t}. \quad (3)$$

Hence t can also be obtained by matching T_{cs} with the T_c of homogeneous Pb with a thickness of $d_0/(1-t)$. We have analyzed the data for a number of Pb/Sb(Ge) multilayers with this approach. Figure 2(a) shows T_c versus n for four Pb/Ge and five Pb/Sb multilayers. The graph looks very busy because of the different Pb and Sb(Ge) thicknesses. The picture is significantly simplified when we plot T_c against $1/d_{\text{eff}}$ in Fig. 2(b). Here d_{eff} is calculated from d_0 and t , and t is first obtained from T_{cs} . Quite remarkably, every series now overlaps with the T_c versus $1/d$ curve for homogeneous Pb (solid line). The variation in the Pb layer thickness does not affect our analysis because t only depends on the Sb(Ge) thickness. This is compelling evidence that this analysis is self-consistent and the increase of T_c with n in the multilayers results from electron transfer through the Sb(Ge) layers.

In Fig. 3 we plot the transmission coefficient versus the Sb or Ge thickness (d_I). t depends sensitively on d_I . There is some scatter in the data because of small uncertainties in the determination of the thicknesses from run to run. Nevertheless, it is clear that t does not depend linearly on d_I . In fact, it is much closer to an exponential dependence, as one would expect from tunneling processes. The solid line in Fig. 3 is an exponential fit with a thickness offset of 7.5 Å.

The large transmission coefficient could be caused by pinholes, direct tunneling, or multistep tunneling via the localized states. Pinholes in the Sb layers could cause charge transfer between the Pb layers. The linear scaling between the conductance and n and the sharpness of the superconducting transitions do not rule out the existence of pinholes in Sb. In order to better understand the nature of the Sb barriers, we have fabricated Pb/Sb/Pb cross-stripe junctions by quench-condensing, with the Sb barrier thickness ranging from 15 to 40 Å. The I - V characteristics were then measured *in situ*. Two types of I - V characteristics were observed on different

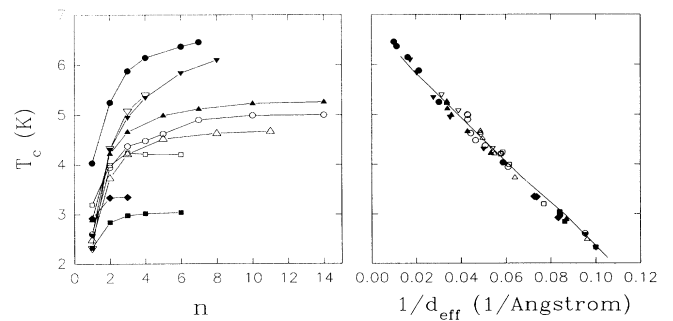


FIG. 2. (a) T_c vs the number of bilayers for nine different series of Pb/Ge and Pb/Sb multilayers. (b) T_c vs the effective Pb thickness (as defined in text) for the nine series of multilayers. The solid line is T_c vs $1/d$ for a homogeneous Pb film.

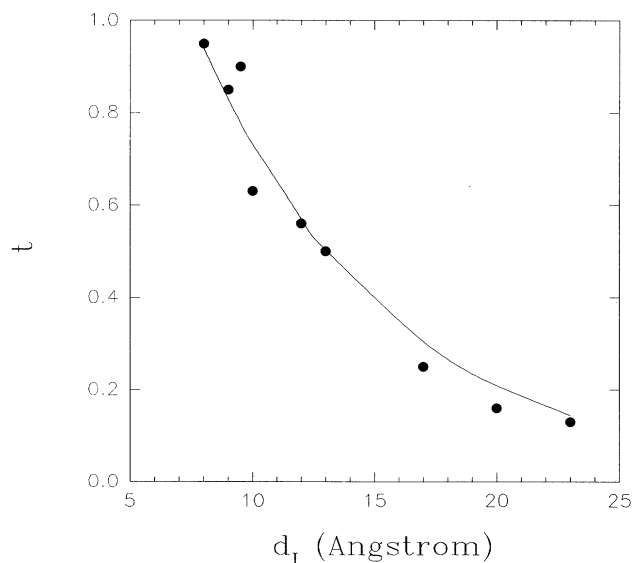


FIG. 3. The transmission coefficient t vs the insulating layer thickness for the nine series shown in Fig. 2. The solid line is an exponential fit with a thickness offset of 7.5 Å.

junctions, as represented in Fig. 4. The first type (curve *a*) results from electrical shorts between the Pb layers, which is indicative of the presence of pinholes in the Sb barrier; the second type (curve *b*) is typical of multistep tunneling,¹⁸ which indicates that the barrier is free of pinholes. More importantly, the appearance of pinholes does not necessarily correlate with the barrier thickness. We have observed the second type of I - V curve in a junction with a thinner barrier than those of junctions with shorts. Therefore, although pinholes may be present in the Sb layers, its density is so low that the average distance between the pinholes is much larger than the superconducting coherence length. Hence the pinholes are ir-

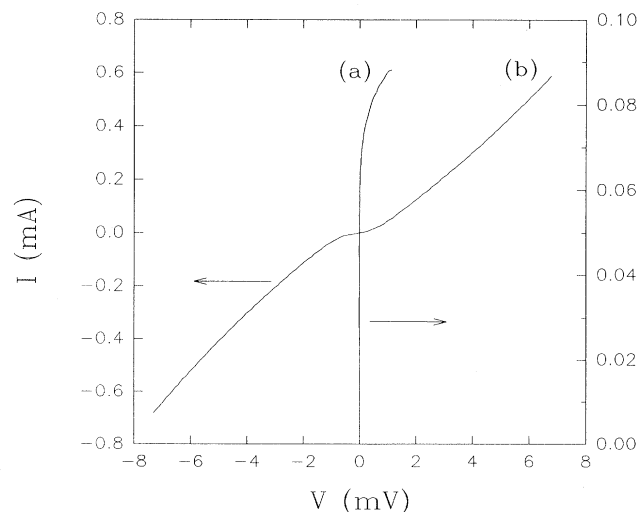


FIG. 4. Two types of I - V characteristics obtained on Pb/Sb/Pb junctions. (a) I - V curve due to electrical shorts in the Sb barrier. (b) I - V curve due to multistep tunneling via localized states in Sb.

relevant as far as superconductivity is concerned in these multilayers. The behavior displayed in Fig. 2 cannot be caused by such a low density of pinholes.

We argue that the charge transfer is largely due to tunneling processes, including both direct tunneling and multistep tunneling. Direct tunneling is elastic; the electrons in one metal layer tunnel into another's available states without changing their energy. Since the tunneling length in these amorphous materials is ~ 10 Å,¹⁸ we expect the direct tunneling to be the dominant process when the insulating layer is thin. When the Sb(Ge) layer becomes thicker than 10 Å the multistep tunneling gradually takes over.¹⁸ The multistep tunneling is an inelastic process; the electrons tunnel through the barrier via several localized states by emitting or absorbing phonons. Its tunneling spectrum differs from that for direct tunneling in that the I - V curve shows significant nonlinearity at high biases. It also allows for subgap tunneling even when the metallic layers are superconducting, and the gap edge is severely smeared, just as shown in Fig. 4 (curve *b*). However, the observation of tunneling through localized states in such thin Sb layers is somewhat surprising. The density of localized states at the Fermi level in *a*-Si or *a*-Ge grown at room temperature is typically $\sim 10^{18}$ states/cm³ eV implying that the distance between the localized states is ~ 100 Å,¹⁹ much larger than the Sb thicknesses in these multilayers. In order for the multistep tunneling to be the dominant process in the multilayers, the density of the localized states has to be $\sim 10^{21}$ states/cm³ eV. This is not impossible in these highly disordered quench-condensed films, and this number is consistent with an estimate using the proximity effect data discussed later.

The high density of localized states in Sb(Ge) could be a reason why we failed to observe any enhancement in superconductivity due to the mechanism proposed by Chakravarty *et al.*⁷ Single electron tunneling between the superconducting layers, which is fatal to this mechanism, is greatly enhanced by the localized states. In order to do this experiment correctly, one would have to use insulating barriers free of localized states, which is apparently difficult to accomplish *in situ* at low temperatures.

In the study of ultrathin homogeneous amorphous superconducting films by quench-condensing, a buffer layer of 1–2 monolayers of Sb or Ge is necessary to support the uniform growth of the superconductor. The role of the buffer layer on the transport and superconductivity of these films has long been an unsettled issue. Our results on these multilayers suggest that this buffer layer plays an important role in both, because of the high density of localized states. First, the localized states provide alternative diffusion paths for the transport of electrons in Pb. This effectively increases the thickness of Pb slightly, thereby reducing the sheet resistance and increasing the T_c . On the other hand, the high density of localized states reduces the average BCS coupling constant, $N(0)V$, because it enables the superconducting electrons to extend into the buffer layer. In other words, the buffer layer also tends to decrease the T_c as a result of the proximity effect with the localized states. The net effect of the buffer layer is a compromise of these two

effects. Because of the linear dependence of the T_c on $1/d_s$, we have $\Delta T_c \propto \delta d_s/d_s^2$, i.e., T_c is much more sensitive to small thickness changes when the film is thin. We therefore expect the first effect to be strong when d_s is small. As d_s increases and T_c approaches the bulk T_c , T_c becomes much less sensitive to d_s . In this limit the proximity effect becomes dominant. We have directly confirmed these conjectures by comparing the T_c of uniform Pb films of different thicknesses with and without an Sb top layer. Similar results were reported by Strongin *et al.*,⁹ who studied the effect of a Ge overlayer on uniform Pb films. The presence of localized states in the Ge layer gives an appropriate microscopic origin for the explanation offered by the authors.

Figure 5 shows the results of two of such measurements. In both runs the thicknesses of the Sb underlayer were the same, while the Pb films were 9 Å and 60 Å thick with a T_c of 2.2 K and 6.0 K, respectively. An Sb top layer of ~ 20 Å was then deposited on the Pb film. For the thin Pb film, the Sb top layer reduces the R_{\square} by 1/4 and *increases* its T_c by as much as 0.2 K [Fig. 5(a)]. In contrast, the Sb top layer *decreases* the T_c by 0.15 K on the thick Pb film [Fig. 5(b)]. The increase in T_c for the thin Pb film is due to the increased effective thickness and the associated decrease in R_{\square} . In the latter case, the T_c and the DOS of the Pb film are very close to the bulk values. We can safely assume that the T_c change is overwhelmingly due to the proximity effect. We can then estimate the density of the localized states from ΔT_c for the thick film. In the proximity effect, the T_c of a superconductor is affected by a different material in its proximity through the averaging of the BCS interaction constant, $N(0)V$, over a length of the coherence length. In the Cooper limit, the effective BCS coupling constant for a Pb/Sb bilayer is

$$N(0)V = \frac{N_{\text{Pb}}(0)d_{\text{Pb}}}{N_{\text{Pb}}(0)d_{\text{Pb}} + N_{\text{Sb}}(0)d_{\text{Sb}}} N_{\text{Pb}}(0)V_{\text{Pb}}. \quad (4)$$

As a good approximation, we use the BCS formula

$$T_c = \frac{\Theta_D}{1.45} \exp\left(-\frac{1}{N(0)V}\right), \quad (5)$$

where Θ_D is the Debye temperature for Pb, which we assume is not changed by a small addition of Sb. Sub-

stituting Eq. (4) into Eq. (5) and noting that $N_{\text{Pb}}(0) \gg N_{\text{Sb}}(0)$, we obtain

$$\frac{\Delta T_c}{T_c} = \frac{N_{\text{Sb}}(0)d_{\text{Sb}}}{N_{\text{Pb}}(0)d_{\text{Pb}}} \frac{1}{N_{\text{Pb}}(0)V_{\text{Pb}}}. \quad (6)$$

Using $[N(0)V]_{\text{Pb}} \sim 0.3$, $N_{\text{Pb}}(0) \sim 10^{23}$, and $d_{\text{Pb}}/d_{\text{Sb}} = 3$, we get $N_{\text{Sb}}(0) \sim 10^{21}$ states/cm³ eV. This is consistent with the estimate we obtained earlier, which gives us some confidence in this interpretation.

The high density of localized states in quench-condensed Sb and Ge also bears consequences on some experiments designed to directly probe the long-range Coulomb interaction in ultrathin disordered metal films through screening. Because of the long-range character of the Coulomb interaction in these ultrathin films, a ground plane that is in close proximity with but electrically well isolated from a thin film can screen the Coulomb interaction in the thin film. If the film is a

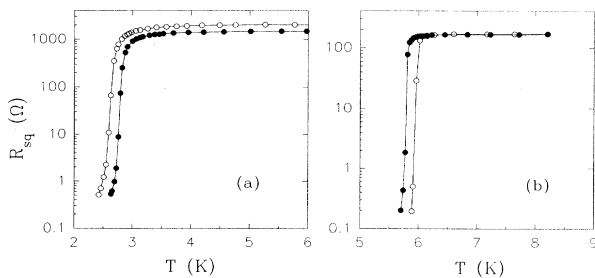


FIG. 5. R_{\square} vs T for two homogeneous Pb films (60 Å and 9 Å) without (open circles) and with (closed circles) an Sb top layer. Note T_c goes down in the 60 Å film and goes up in the 9 Å film with the addition of the Sb top layer.

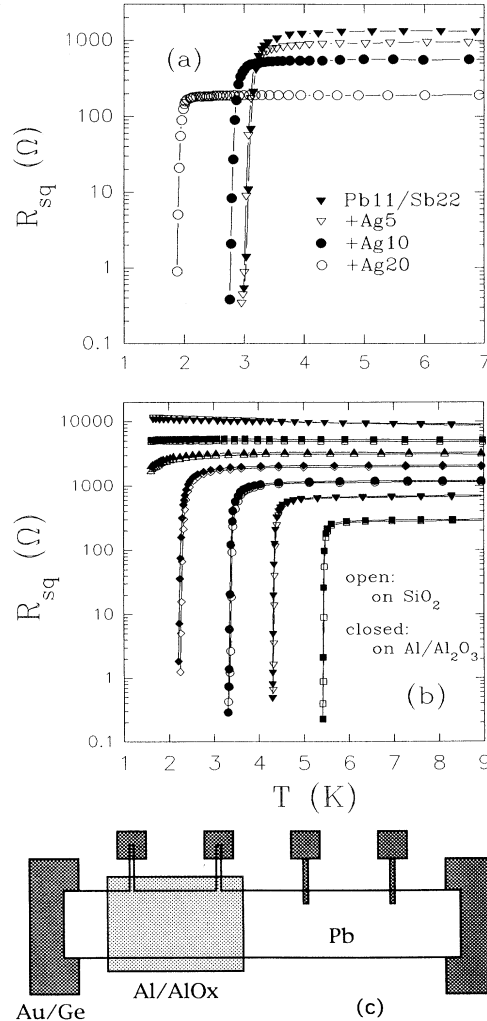


FIG. 6. (a) R_{\square} vs T for a Pb/Sb/Ag sandwich at different Ag thicknesses. (b) R_{\square} vs T for two sections of a Pb film on bare SiO₂ substrate (open symbols) and on Al/Al₂O₃ (closed symbols). (c) The sample geometry for (b).

superconductor, the screening should decrease the sheet resistance and increase the T_c . Many attempts have been made^{20,21} to observe the screening effect. However, the results are conflicting. In a recent experiment,²¹ a small increase in T_c (0.02 K) was observed when a thick Au film was deposited on top of an anodized Ta film with a depressed T_c . In contrast, in another set of experiments, Yap and Bergmann,²⁰ measured the change in T_c of a Bi/Sb/Bi sandwich while varying the thickness of the top Bi layer *in situ*. No increase in T_c was observed even when the top Bi layer was as thick as the bottom one. This was cited as evidence against the presence of long-range Coulomb interaction in two-dimensional disordered films. However, several aspects of the experiment make the interpretation of the data somewhat ambiguous. First, from our current results we know that the two Bi layers were not electrically well isolated from each other because of the high density of localized states in the Sb layer. Second, the T_c of the bottom Bi layer was about 2/3 of the T_c for bulk amorphous Bi. The screening length in this Bi film is not expected to be much longer than that in bulk Bi. Finally, the top Bi layer had to be kept no thicker than the bottom one. It did not make a good ground plane since its screening length was not any shorter than that in the bottom Bi layer. Therefore, the screening effect by the second Bi layer, if there is any, is expected to be small. It is better to use a normal metal as the second metal layer so that one can make it thick enough to be a good ground plane. We have measured the T_c of a quench-condensed Pb/Sb/Ag sandwich with different Ag thickness. The results are shown in Fig. 6(a). No T_c increase was observed with the addition of the Ag top layer. On the contrary, the T_c decreases steadily with increasing Ag thickness, indicating a strong proximity effect between Pb and Ag through the Sb barrier. Again, this result can be attributed to the electron diffusion through the localized states in Sb. In order to get around this problem, we have measured simultaneously R_{\square} versus T for two adjacent sections of a Pb film on a bare SiO₂ substrate and on Al/Al₂O₃ respectively. The sample geometry is shown in Fig. 6(c). The Au/Ge contacts and the high purity Al film were deposited before the substrate was loaded into the appa-

ratus. The Al film was $\sim 200 \text{ \AA}$ thick and was oxidized in air for several hours. An Al/Al₂O₃/Pb junction made this way would have a resistance of $> 10^8 \Omega$; we are confident that the Al was electrically well isolated from the Pb film. As shown in Fig. 6(b), both the R_{\square} and the T_c are almost identical for the two sections. We have repeated the same experiment several times, changing the substrate orientation relative to the sources to eliminate the effect from any potential thickness gradient on the Pb film. The results were consistent. Our results put an upper limit of $\sim 20 \text{ \AA}$ on the screening length in these ultrathin homogeneous disordered films. Although the same conclusion was reached from other studies,^{2,20} the complications from the proximity effect and electron transfer through the insulating layer were eliminated in our experiments. In addition, the ability to vary the film thickness *in situ* enabled us to establish unambiguously the absence of the screening effect throughout the insulator-superconductor transition.

In summary, we have carried out extensive *in situ* transport and tunneling measurements on ultrathin quench-condensed Pb/Sb and Pb/Ge multilayers. Despite the strongly correlated nature of the ultrathin Pb layers, we did not observe any change in the basic mechanism of superconductivity in the multilayers. The electrical properties of the insulating layers were characterized directly through *in situ* fabricated Pb/Sb/Pb junctions. We find that the density of localized states in these quench-condensed Sb and Ge layers is unusually high. The localized states play an important role in the transport and superconducting properties of the Pb/Sb(Ge) multilayers, as well as homogeneous superconducting films grown on a Ge or Sb buffer layer. We have also demonstrated more convincing and direct evidence for the absence of the screening effect in homogeneous two-dimensional superconducting films.

It is a pleasure to acknowledge helpful discussions with S. Kivelson, S. Chakravarty, R. Barber, and F. Sharifi. We also thank S. I. Applebaum for discussions and a critical reading of the manuscript. This work was supported by ONR Grant No. N0001491J1320.

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