The superconductor insulator transition in systems of ultrasmall grains

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We present transport measurements on quench condensed granular Pb films in which the grains are 40 – 80Å in diameter. These films show a cross over from an insulator to a superconductor behavior as the nominal thickness of the layer is increased. This transition is different in nature than those seen in quench condensed systems reported in the past where the films were either uniform or granular with grain sizes on the order of 200Å. We discuss possible physical mechanisms for these transitions.

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The superconductor insulator transition (SIT) in thin layers has attracted a lot of interest for close to three decades and has been lately revived due to the possibility that it is a quantum phase transition at T=0 [1–6]. A number of theoretical scenarios have been suggested to try to explain why superconductivity is destroyed as the thickness of the layer is decreased. These can be classified into two major classes. The first invokes a ”Bosonic” picture in which isolated superconducting islands exist in a non superconducting matrix [4]. Here, Cooper pairs are localized on the islands resulting in an insulating behavior of the film. The other scenario [5] proposes localization of single electrons as a result of disorder and Coulomb interaction effects.

The technique of quench condensation, i.e. sequential evaporation on a cryogenically cold substrate under UHV conditions, has proven to be a very useful tool for studying the SIT [7–10]. Depending on the choice of substrate, this technique enables one to probe both mechanisms of the SIT: If the samples are quench condensed on a passivated substrate (such as SiO), they grow in a granular manner so that the film brakes up into separated islands. The average distance between the islands decreases upon adding material. In these samples there is a critical thickness $d_c$, below which no conductivity can be measured. For granular Pb, $d_c \approx 100Å$. Once the thickness, $d$, of the sample is larger than $d_c$, the sheet resistance, $R$, drops exponentially with thickness until, for $R \leq 6k\Omega$, it switches to a normal Ohmic behavior ($R \propto 1/d$). STM measurements on quench condensed Pb granular films close to $d_c$ [11–13] show that the grains are about 200Å in diameter and 50-80Å in height. On the other hand when the substrate is pre-coated by a thin layer of amorphous Ge (which is insulating at 4K), the sample grows uniformly and is continuous at a thickness of 1-2 monolayers of material [7] after which the sample obeys the usual $R \propto 1/d$ dependence. It is believed that the large number of dangling bonds in the Ge layer act as nucleation centers for the evaporated metals, thus the deposited adatoms are less mobile and the sample grows homogeneously.

These two geometries lead to two different types of SIT. Figure 1 compares the behavior of uniform and granular quench condensed films of Pb on a SiO substrate.

![Figure 1](image_url)

**FIG. 1.** Resistance versus temperature for sequential layers of quench-condensed granular Pb (left) and uniform Pb evaporated on a thin Ge layer (right). Different curves correspond to different nominal thickness.

In the uniform film it is seen that for $R > 6k\Omega$ the sample is insulating. As the film thickness is increased and the resistance is decreased, a superconducting transition evolves with increasing critical temperature, $T_c$, which approaches the bulk value (7.2K for Pb) for thick enough samples. The superconducting transitions themselves are relatively sharp and well defined. In the granular film a crossover from an insulating to superconducting behavior is also observed. However, here $T_c$ is not very well defined and the superconducting transitions have broad tails which become sharper as material is added to the film. If $T_c$ is defined as the temperature at which the resistance starts dropping exponentially, it has bulk value and barely changes throughout the entire SIT. Moreover, even on the insulating side of the SIT the curve changes its slope at $T = T_c$, reflecting the influence of superconductivity even in the thinnest measurable samples. A similar effect is seen in the properties of the energy gap, $\Delta$. Tunneling measurements show [14,15] that while in the uniform film $\Delta$ grows with increasing thickness so
that the ratio \( \frac{\Delta}{T_c} \) is roughly constant, in the granular case the bulk energy gap is measured even on the insulating side of the transition and its does not change with additional material.

These differences are attributed to a different nature of the SIT [14]. In the granular case the grains are large enough to sustain superconductivity with bulk properties. However, for the high resistance samples, there are phase fluctuations between the grains leading to an insulating R-T curve. As more material is added the Josephson coupling increases and eventually superconducting percolation is achieved. The situation is different for the uniform geometry where the film is believed to be homogeneous on an atomic level. In this case the SIT is due to suppression of the order parameter amplitude at ultra-low thickness due to an interplay between coulomb interactions and disorder.

So far, all studies on quench condensed based SIT have been performed on either extreme of very homogeneous layers or granular films with grains large enough to have bulk superconductor characteristics. In this paper we describe results obtained on films that bridge these two extremes. We grow granular Pb films in which the grains are too small to sustain bulk superconducting properties and investigate the nature of the SIT in these samples. This geometry is intriguing. Though much attention has been directed towards understanding the nature of superconductivity in grains in which the discrete electronic level spacing is larger than \( \Delta \), [16, 17], not much is known about an array of coupled ultrasmall grains.

Since the Ge under-layer appears to be the cause for the uniformity of the films, the natural way to try and reduce the size of the grains would be to grow a very thin layer of Ge prior to the Pb evaporation. This turns out to be very difficult because 1-2 monolayers of Ge are enough to cause atomically uniform films. Alternatively, one can achieve a suitable substrate for ultrasmall grains by applying different evaporation conditions than those used in previous works. Conventional quench condensed films, such as those described in figure 1, were deposited in a vacuum can set-up immersed in a liquid He\( ^4 \) bath, resulting in UHV conditions (\( P < 10^{-11} \text{mbar} \)) due to cryopumping of the can. The samples described in this work were grown in a normal high-vacuum evaporation chamber equipped with a cold finger capable of reaching temperatures as low as 4K. The base pressure in the chamber was \( 10^{-8} \) mbar. This pressure is low enough to allow relatively pure evaporations but it is still a few orders of magnitude higher than that in the previous set-ups. In this system one can expect particles from the residual pressure in the chamber to cryopump on the substrate, which is the coldest element in the set-up. As time goes by nucleation centers form on the substrate, causing the granularity of the evaporated material to decrease. Indeed, depending on the waiting time before evaporation, we achieve samples with \( d_c \) between 20 and 50Å. This should be compared to \( d_c \approx 100 \text{Å} \) for granular Pb and \( d_c \approx 5 \text{Å} \) for uniform Pb.

The resistance versus temperature curves for several samples having \( d_c \) between 45 and 22Å are shown in figure 2. These curves were measured using standard 4 probe AC techniques and making sure, for each point on the curve, that the I-V characteristics are in the linear regime. An SIT is observed for all these samples. However, the details of the transition are different than both those of uniform and those of granular systems. In fact the samples have characteristics of both types of geometries. On one hand \( T_c \) changes considerably as more material is added and on the other hand the transitions exhibit broad tails.

![Figure 2](image-url)  

**FIG. 2.** Resistance versus temperature of Pb quench condensed samples having \( d_c = 45, 33 \) and 22Å from top to bottom respectively. The dashed lines are the calculated curves that are expected according to equation 2 for large grains.

The fact that these samples are granular in nature can be seen from a number of experimental findings. First, \( d_c \) is much larger then a monolayer indicating that the films are not atomically uniform. Secondly, the resistance as a function of thickness (for \( d > d_c \)), as shown in
figure 3, begins with a sharp exponential dependence before turning Ohmic, reflecting the tunneling nature of the transport. In addition the I-V curves shown in the inset of figure 3, exhibit a series of hysteretic jumps, typical to a granular system which contains an array of Josephson junctions with different critical currents. These cause avalanche bursts of the I-V curve at discrete voltages [18].

Hence, the film is composed of grains. Nevertheless, the critical temperatures we measure for all the layers are smaller than that of the bulk. This together with the relatively small \( d_c \) of the films lead to the conclusion that the grains are considerably smaller then those in the previously studied granular structures. Using a simple model based on the geometries deferred from the STM measurement in the large grains we estimate the average grain size in our samples to be around 40-80Å in diameter and about 50Å in height. The electronic level spacing, \( d^* \) in a single grain can be calculated using the free electron expression for the density of states, \( N(E_F) \):

\[
d^* = \frac{1}{N(E_F)} = \frac{2\pi^2\hbar^2}{mk_F\Omega}
\]

where \( \Omega \) is the volume of the grain. For our Pb grains this yields a level spacing of \( \sim 0.75 \) meV which is smaller than the superconducting energy gap, \( \Delta = 1.4meV \). Thus, superconductivity can be expected to be suppressed and the critical temperature of the grains may indeed be smaller than that of the bulk.

Because the geometry of these systems lies between that of the uniform films and the large-grain films, one could expect that the physical properties would also show intermediate behavior. This is not the case. Since \( d_c \) is geometry and material dependent, the relevant parameter for comparing different systems is the normal-state sheet resistance, \( R_N \), which reflects the degree of disorder, rather than the film thickness. In the large grain systems, the resistance versus temperature for \( R_N < 80\k \) and \( T < T_c \) follows an exponential behavior (see figure 1) and can be expressed in the following way [19]:

\[
R = R_0 e^{\frac{T}{T_c}}
\]

Though the cause for this dependence is not yet understood, it turns out to be universal for all quench condensed granular superconductors. Moreover, the slope of the curves, \( \frac{T}{T_c} \), depends only on \( R_N \) and not on the material or the \( T_c \) [20]. Our samples do not follow this dependence. Instead, \( R \) versus \( T \) is always slower than an exponential law and the slope of the curves everywhere is less steep then what it would be for a similar \( R_N \) in the usual granular system. This can be seen in figure 2 where we have sketched curves as they would have been measured for large-grain systems having a similar \( T_c \) and \( R_N \). Clearly the small-grain samples exhibit a much slower dependence on temperature. This trend becomes weaker as \( d_c \) decreases, as expected when the sample approaches uniformity.

The critical temperatures measured in these samples also do not fall between those of the two extreme geometries. Figure 4 depicts \( T_c \) as a function of \( R_N \) for a number of our samples compared to that of uniform films [14]. It is seen that all samples with \( R_N < 1\k \) have critical temperatures smaller than that of the uniform film. The smaller the \( d_c \) the smaller are the critical temperatures (though this trend seems to stop as the sample approaches uniformity).
Hence, regarding the tail broadness, the small grain films are more extreme than the large grain films, while, concerning the critical temperatures they fall shorter than those of uniform films.

The results presented above imply that the mechanism of the SIT in the ultrasmall grain system is different than both the transition in a uniform film, which is dominated by order parameter amplitude suppression, and that of the large-grain systems which is governed by phase fluctuations. Though the mechanism is not yet understood it is clear that fluctuations in either the phase or the amplitude of the order parameter alone is not enough to account for the findings. Rather, it seems reasonable that a combination of both parameters are involved in the behavior of these systems. In order to achieve superconductivity, two conditions have to be fulfilled. One is that all the grains in the conduction percolation network would be close enough to allow Josephson coupling and the second is that they all be superconducting. Unlike the other two geometries where all the film turns superconducting at once, the ultrasmall grain systems may involve a distribution in $T_c$ due to a distribution in grain size. Hence, at temperatures smaller than the bulk $T_c$ many of the grains are non-superconducting while others may have turned superconducting. This leads to a situation (which does not occur in the large-grain films) in which grains can be close enough to allow strong Josephson coupling, and yet the sample will not have long-range superconductivity because many of the grains are not superconducting. Adding small amounts of material to the layer causes the average grain size to grow (either by coalescence or by increasing the tunneling between grains) thus increasing the average critical temperature. The samples described in this work demonstrate the nature of the transition in systems in which very small regions of superconductivity are embedded in a non-superconducting matrix. This is believed to be the case in many composite samples studied recently (see for example [3]). The R-T dependences of any dirty superconductor can be used as a fingerprint to determine the geometry of the superconducting material and the governing SIT mechanism.

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[1] A.F. Hebard and M.A. Paalanen, Phys. Rev. Lett. 65, 927 (1990).
[2] A. Yazdani and A. Kapitulnik, Phys. Rev. Lett. 74, 3037 (1995); N. Mason and A. Kapitulnik, Phys. Rev. Lett. 82, 5341 (1999).
[3] V.F. Gantmakher et al., JETP Lett. 68, 345 (1998); V. Gantmakher et al, Physica B 284-288, 649 (2000); V. Gantmakher et al, to be published.
[4] M.P.A. Fisher, G. Grinstein and S. Grivin, Phys. Rev. Lett. 64, 587 (1990); M.P.A. Fisher, Phys. Rev. Lett. 65, 923 (1990); A. Gold, Z. Phys. B87, 169 (1992).
[5] [10] R.P. Barber and R.E. Glover III, Phys. Rev. B40, 6754 (1990).
[6] A.T. Truscott, Ph.D. Thesis, University of California, San Diego (1999)
[7] A. Frydman and R.C. Dynes, To be published in Phil. Mag.
[8] A. Frydman, E.P. Price and R.C. Dynes, Phys. Rev. B49, 3409 (1994).
[9] J.M. Valles, Jr, S. Hsu, R.C. Dynes and J.P. Garno, Physica B 197, 522 (1994) and references within.
[10] R.P. Barber, L.M. Merchant, A. La Porta and R.C. Dynes, Phys. Rev. B40, 3409 (1994).
[11] P.W. Anderson, J. Phys. Chem. Solids. 11, 28 (1959); D.C. Ralph, C.T. Black and M. Tinkham, Phys. Rev. Lett. 74, 3241 (1995); Phisica 218B, 258 (1996); Phys. Rev. Lett. 78, 4087 (1997).
[12] For a comprehensive review see J. van Delft, Ann. Phys. (Leipzig) 10, 1 (2001)
[13] A. Frydman, E.P. Price and R.C. Dynes, Solid State Commun. 106, 715 (1998)
[14] L. Marchent, J. Ostrick, R.P. Barber Jr. and R.C. Dynes, Phys. Rev. B
[15] O. Naaman, A. Frydman and R.C. Dynes, in preparation.