Electron Tunneling Determination of the Order-Parameter Amplitude at the Superconductor-Insulator Transition in 2D

J. M. Valles, Jr.,⁽¹⁾ R. C. Dynes,⁽²⁾ and J. P. Garno⁽³⁾

⁽¹⁾Materials Science Institute, University of Oregon, Eugene, Oregon 97403 ⁽²⁾Department of Physics, University of California at San Diego, La Jolla, California 92093

⁽³⁾AT&T Bell Laboratories, 600 Mountain Avenue, Murray Hill, New Jersey 07974

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We have investigated the behavior of the superconducting energy gap Δ in ultrathin films of quench condensed Bi near the superconductor-insulator (SI) transition. From electron tunneling measurements on these films, we conclude that Δ becomes very small and approaches zero at the SI transition. We studied high-sheet-resistance films with T_{c0} 's as low as 0.19 K. This is a factor of 40 lower than the low-sheet-resistance film T_{c0} of 6.4 K.

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As the amount of disorder is increased in a metallic system the conduction-electron states begin to localize, and the strength of the effective electron-electron interactions increases. These two effects hinder the formation of the normally superconducting state in a disordered material. The localization of the electrons opposes the formation of a coherent state over distances longer than the localization length. The enhanced repulsive electron-electron interactions will reduce the net attractive interaction that is required for Cooper pairing. In two-dimensional systems the effects of increasing disorder on the superconducting state are dramatic and eventually drive a transition from a superconducting to an insulating state [1-6].

To qualitatively describe this transition it is helpful to consider the superconducting order parameter $\Psi \propto \Delta_0^{1/2} e^{i\phi}$ where the amplitude of the order parameter, Δ_0 , is the zero-temperature energy gap and ϕ is its phase. If a film is composed of islands that are large enough to have independent superconducting properties then the superconducting state can be weakened by reducing the (Josephson) coupling between the islands. This is accomplished by reducing the tunneling probability between the islands which results in an increase in the normal-state sheet resistance, R_{\Box} , of the film. Fluctuations in the phase of the order parameter ensue, which when strong enough destroy the long-range phase coherence across the film [2,6,7]. The situation has been shown to be different for the case where the morphology of the film is more homogeneous, i.e., for disorder on shorter length scales. In systems that are not too close to the superconducting to insulating transition, increases in R_{\Box} have been shown to lead to well-defined decreases in the amplitude of the order parameter [7,8] and the mean-field transition temperature, T_{c0} [3-11], in direct contrast with the islanded film case. One might expect that very near the superconducting to insulating transition, fluctuations in the amplitude of the order parameter would play a dominant role in driving the transition. Recently, however, it has been argued that even in these systems, phase fluctuations dominate the physics of the superconducting to insulating transition [6,12]. In this paper we present tunneling and transport results on uniformly disordered Bi films that are very close to the superconducting to insulating transition. We find that the energy gap in these films is reduced significantly from its bulk value, showing that the amplitude of the order parameter continues to decrease at the same rate as T_{c0} even near the transition. That is, the amplitude of the order parameter is extremely small or zero at the superconductor to insulator transition in the homogeneous case. Fluctuations in the amplitude must therefore be at least as important as fluctuations in the phase of the order parameter.

The electron tunneling and transport measurements described here were performed on films evaporated onto substrates that were held at low temperatures (4-7 K) in a cryogenic evaporator. The substrates were thermally connected to the mixing chamber of a dilution refrigerator giving a temperature range for our measurements of 0.13 < T < 7.0 K. Contact pads and an Al strip of width 0.01 in. were deposited on the substrate prior to cooling down the apparatus. The Al strip had a small amount of Mn impurities in it to prevent it from superconducting. It was oxidized in air and served as the counterelectrode in the tunnel junctions, Al/oxide/Bi film. At low temperatures, a 1-2-monolayer film of Sb was evaporated onto the substrate and Al oxide surfaces followed by a series of Bi evaporations. The width of the Bi film was 0.1 in, so that the tunnel junction area was 0.1 in. ×0.01 in. The Sb film is an insulator and helps the Bi form a very thin uniform, continuous film [13]. For example, the mass of Bi per unit area for a film with $R_{\Box}(7 \text{ K}) = 8 \text{ k}\Omega/\Box$ was equivalent to only four atomic layers of crystalline Bi. After each Bi evaporation the resistance of the film as a function of temperature, and the voltage dependence of the tunnel junction conductance, $G_i(V)$, were measured. The Sb film and the Al oxide serve as the tunnel barrier. The completed tunnel junction had the standard cross stripe geometry. Care was taken to assure that the measurements of $G_i(V)$ with the Bi film in the normal state

were not influenced by the sheet resistance of the Bi film. With this arrangement, we can correlate all changes in the tunnel junction conductance with changes in R_{\Box} and so with changes in the film properties and not to changes in the tunnel junction barrier. We present tunneling and transport data on three Bi films, two with high R_{\Box} (near the superconducting to insulating transition), and one with a very low R_{\Box} . All three were evaporated in sequence onto the same film and tunnel junction. We have obtained qualitatively similar results on Pb films in other experimental runs. In all cases, the tunnel junctions were of high quality, showing essentially no leakage current due to nontunneling conduction.

Recent measurements on ultrathin Bi films have shown that the superconducting to insulating transition occurs near $R_{\Box}(T > T_c) \simeq 6.45 \text{ k}\Omega/\Box$ [4]. We have measured the temperature dependence of the sheet resistance and the tunneling density of states of similar uniform Bi films in order to evaluate the behavior of the amplitude of the order parameter near this superconductor to insulator transition. In Fig. 1 we plot the $R_{\Box}(T)$ for two Bi films with $R_{\Box}(7 \text{ K}) \cong 8.0$ and 5.8 k Ω/\Box . Both films show behavior suggesting that at T=0 they would be insulators (dR/dT < 0), but then have reasonably sharp superconducting transitions. In the case of the 8.0-k Ω/\Box film the minimum resistance that we measured was limited by the base temperature of our refrigerator. Clearly both films show mean-field transition temperatures that are greatly depressed from the thick Bi film value, $T_{c0} \simeq 6.4$ K, which was achieved upon further evaporation. If we take the midpoint of $R_{\Box}(T)$ to give T_{c0} then we find that $T_{c0}=0.7$ and 0.19 K for the 5.8- and 8.0-k Ω/\Box films, respectively. These films are very close to the superconducting to insulating transition. In fact we find that the 8.0-k Ω/\Box film superconducts, in contrast with earlier measurements on Bi films evaporated onto Ge [4], for which a case was made that the SI transition occurred at $h/4e^2 \approx 6.5 \text{ k}\Omega$.



FIG. 1. Resistance as a function of temperature for two sequentially evaporated films that had sheet resistances at 7 K, $R_{\Box}(7 \text{ K})$, of 5.8 and 8.0 k Ω/\Box .

A slightly thinner film with higher R_{\Box} is insulating. We speculate that the discrepancy between these experiments results from the different preplating elements, Sb and Ge, that were used. Together the results support the idea that SI transition does not occur at a universal value of R_{\Box} . The purpose of this paper is to study the amplitude of the superconducting order parameter on the superconducting side of the SI transition.

We can learn about the amplitude of the order parameter or the energy gap near the superconductor to insulator transition by measuring the tunneling density of states of these films. The conductance of a normal-metal/oxide/ superconducting film tunnel junction can be written [2]

$$G_j(V,T) \propto N_2 \int_{-\infty}^{+\infty} N_S(E) N_N(E) \frac{df(E+eV)}{dE} P(E) dE ,$$
(1)

where N_2 is the density of states of the Al electrode which is assumed constant, $N_S(E) = |E|/(E^2 - \Delta^2)^{1/2}$ is the BCS density of states, $N_N(E)$ is the normal-state density of states, E is the electron energy measured from the Fermi energy E_F , f is the Fermi function, and P(E)is the tunneling probability which on the voltage scales considered here is approximately constant. At low temperatures ($T \ll T_{c0}$) the Fermi functions are sharp and Eq. (1) reduces to

$$G_i(V) \propto N_2 N_S(eV) N_N(eV) . \tag{2}$$

Thus, a measurement of the voltage dependence of the conductance at low temperatures gives a measure of the density of states of the film as a function of energy.

In Fig. 2 the conductance of the tunnel junction for the 5.8- and 8.0-k Ω/\Box films at T = 0.13 K and a very low R_{\Box}



FIG. 2. Tunnel junction conductances as a function of voltage for the $8.0 \cdot k \Omega/\Box$ (lowest curve) and $5.8 \cdot k \Omega/\Box$ (middle curve) films at T = 0.13 K and for a $R_{\Box}(7 \text{ K}) = 75 \Omega/\Box$ film (upper curve) at $T \approx 0.3$ K. The junction conductances are normalized by that of the $75 \cdot \Omega/\Box$ film measured at a temperature above its T_c . Inset: The data for the 5.8- and $8.0 \cdot k \Omega/\Box$ films on an expanded scale.

film, 75 Ω/\Box , at T < 1.0 K, are plotted as a function of voltage. The tunnel junction conductances of the two high R_{\Box} films have been normalized to the normal-state conductance for the very low R_{\Box} film which is essentially voltage independent over this range and equal to 0.063 $k \Omega^{-1}$. This procedure eliminates P and is legitimate because the 75- Ω/\Box film is the result of subsequent evaporations on the same junction. There are three important features of these data. First, the overall tunnel junction conductance of the high R_{\Box} for these films is smaller over this voltage range than that of the low R_{\Box} film, e.g., $G_j(5)$ mV, 5.8 k Ω) \simeq 0.4G_i (5 mV, 75 Ω/\Box). Second, there is a "cusp" in G_i at V=0. These features are characteristic of tunneling into disordered thin films and have been studied extensively [14,15]. Finally, there is a bump in $G_i(V)$ at a low voltage in the 5.8-k Ω/\Box film which appears below the transition temperature of this film. This bump is clearer in the expanded data in the inset of Fig. 2.

The first two features are qualitatively what one would expect to see based on observations made in tunneling measurements on lower R_{\Box} films. It has been shown that in low R_{\Box} films, $R_{\Box} \ll h/4e^2$, disorder-enhanced Coulomb interactions lead to logarithmic corrections to the density of states of the form

$$\frac{\delta N_n(E)}{N_0} = -\frac{1}{16\pi} \frac{R_{\Box}}{(6.5 \text{ k}\Omega)} \times \ln\left(\frac{|E|}{\hbar D(2\pi/d)^2}\right) \ln\left(\frac{(2\pi)^2 |E|}{\hbar D\kappa^4 d^2}\right) \quad (3)$$

in the weak disorder limit [15]. D is the electronic diffusivity, κ is the 2D screening wave number, and d is the film thickness. These corrections lead to a cusp and an overall reduction in $G_j(V)$ similar to what we observe in this strongly disordered film. In earlier work, we showed that the overall reduction in $G_j(V)$ in junctions on films with R_{\Box} up to 500 Ω/\Box , for which the density of states correction at 5 mV (the average phonon energy relevant for superconductivity in Pb) is about 10%, agrees quantitatively with this model [9]. Thus, we believe that the data on these high R_{\Box} films show that disorder-enhanced Coulomb interactions suppress the tunneling density of states very strongly, by more than 50% at 5 meV.

Near V = 0 a "bump" can be observed in $G_j(V)$ for the 5.8-k Ω/\Box film at T = 0.18 K $< T_{c0} = 0.7$ K. This is more clear on the smaller voltage scale of the inset of Fig. 2. We associate this bump with the peak in the density of states at $V = \Delta/e$ in this superconducting film. In Fig. 3 we plot $G_j(V)$ at T = 0.13 K normalized to $G_j(V)$ at 0.61 K over a smaller voltage scale. This normalization removes most of the energy dependence of the normal-state density of states and brings out the structure in the superconducting density of states. While this normalization procedure is only approximate, we believe that it is



FIG. 3. Tunnel junction conductance of the 5.8-k Ω/\Box film at T=0.13 K normalized to its conductance at T=0.61 K.

sufficiently good to give us a superconducting density of states that is very near the true answer. The shape of the normalized conductance qualitatively resembles that expected for tunneling into a BCS superconductor. There is a peak in the normalized conductance and a reduction of the number of states near the Fermi energy. The deviations from perfect BCS behavior may be partially due to the normalization procedure that we have had to use and may also reflect the fact that the density of states in a strongly disordered superconducting film may be different from the BCS form because of quasiparticle lifetime or fluctuation effects [2,16,17]. For such films where T_c is so severely reduced, it is not unexpected that fluctuation effects are eventually observed. Nevertheless, the signature of the superconducting energy gap is clear. We can estimate the size of the energy gap from these data by measuring the voltage at which the normalized conductance crosses 1 [18]. We get $\Delta \approx 0.082$ meV. Taken with the resistive midpoint we get $2\Delta_0/k_BT_c \approx 2.7$. In view of the substantial normalization effects this is reasonably close to the BCS value. This implies that this high- R_{\Box} Bi film has nearly BCS-like characteristics.

On the highest R_{\Box} film (8.0 k Ω/\Box), we indeed observe the beginning of the opening of a gap as shown in the inset of Fig. 2. Because we are not at sufficiently reduced temperature, T/T_c 0, and the potential normalization errors are more severe, however, more quantitative analysis seems inappropriate at the moment.

In earlier work, on uniform ultrathin films of Pb and Sn, the reduction in T_c with R_{\Box} was shown to be accompanied by a reduction in Δ in such a way that $2\Delta_0/k_BT_{c0} \approx \text{const}$ for $R_{\Box} \leq 3 \text{ k} \Omega/\Box$ [8,9]. The films showed BCS behavior with reduced transition temperatures. The reduction in Δ and T_{c0} correlated with a reduction in the tunneling density of states due to disorder-enhanced electron-electron interactions. The size of the reduction in $N_n(E)$ has been shown to be sufficient to account for the depression of T_c and Δ in these films [9,19]. Our results here suggest that this trend continues up to the superconducting to insulating transition. That is, T_{c0} and Δ_0 decrease at the same rate even in films near the superconducting to insulating transition and at the transition, T_{c0} and Δ go to zero. The concomitant reduction in the tunneling density of states at the average electron-phonon frequency implies that the reduction in T_{c0} and Δ_0 are due to disorder-enhanced electron-electron interactions [9,19]. We conclude that the amplitude of the order parameter is extremely small or zero at the superconducting to insulating transition in these homogeneous films. We add, however, that in the granular case we reach a very different conclusion. The amplitude Δ_0 is finite at the transition and T_{c0} is not well defined [2,7].

Recently, theories of the SI transition have been proposed that argue that the transition is driven by increased phase fluctuations of the order parameter [12]. In this picture, below a critical resistance but in the critical regime the Cooper pairs form a Bose superfluid. Above the critical resistance these bosons localize and a superfluid of vortices exists. Since the vortices move freely through the system there are large phase fluctuations, dissipation, and, consequently, insulating behavior. What is essential to this picture is that the amplitude of the order parameter must be finite and robust in the vicinity of this transition. The formation of vortices requires a well-developed order-parameter amplitude. Our work here shows that in uniform Bi films, the amplitude of the order parameter (energy gap) goes to zero or becomes extremely small near the transition, i.e., it is not well developed. It is small enough that fluctuations in *its* amplitude should become significant. While we cannot rule out from the data of Fig. 3 that there may be some fluctuation effects observed, it is clear that $\Delta \rightarrow 0$ as $T_c \rightarrow 0$ and there are fermions in the system close to the transition. Thus, we argue that a theoretical description of the superconducting to insulating transition in these uniform Bi films must take fluctuations in the amplitude as well as the phase of the order parameter into account. We emphasize that these tunneling measurements show very clearly that in this uniform case $\Delta \rightarrow 0$ at the transition, unlike in the granular case where it has been shown that Δ remains finite through the transition [2].

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