Electron tunneling into strongly disordered films: The influence of structure on electron-electron interactions

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We present measurements of the electronic tunneling density of states of ultrathin films with sheet resistances at 8 K in the range $100\ \Omega < R < 100\ \mathrm{k}\Omega$. We find that in the strongly disordered regime the Coulomb anomaly in the tunneling density of states grows in strength with R in a manner which depends strongly on film structure. These data demonstrate that the effective electron-electron interactions in disordered films depend on film structure in this regime. We discuss the implications of this behavior for the properties of superconductors near the two-dimensional superconductor-to-insulator transition.

The effects of static disorder on the transport properties of two-dimensional (2D) electronic systems have been studied intensively for the past 20 years. These efforts have led to a detailed understanding of the low-temperature transport properties of weakly disordered systems, i.e., systems for which $k_F\ell > 1$ where k_F is the Fermi wave number and ℓ is the elastic mean free path. This understanding is embodied in the theories of weak localization[1] and disorder-enhanced electron-electron (e^--e^-) interaction effects.[2] In particular, these perturbation theories can account for the anomalous logarithmic increase in resistance with decreasing temperature, the negative magnetoresistance observed in many systems, and the depression in the density of states at the Fermi energy. A very remarkable aspect of these phenomena is that the size of these effects scales with sheet resistance R, and is quite insensitive to the details of the materials making up the 2D system.[1–3] For example, the small corrections to the sheet resistance are nearly the same in 2D electron gases in metal-oxide-semiconductor field-effect transistors,[3] ultrathin metal films,[1] and some of the nonsuperconducting cuprate materials[4] that have equal sheet resistances.

By comparison, the present understanding of strongly disordered systems ($k_F\ell \leq 1$ and $R \simeq h/e^2$), where the perturbation theories of weak localization and disorder-enhanced e^--e^- interactions break down, is significantly less complete.[5] In this regime, interesting phenomena such as the superconductor-to-insulator (SI) transition[6–9] and the crossover from extended state to hopping-dominated transport occur.[3,5] These phenomena are believed to result from the growth and interplay of localization and e^--e^- interaction effects[9,10] and to explain them requires insight into the relative importance of these two influences. Consequently, experiments such as electron tunneling[2,11–13] that only probe e^--e^- interaction effects, or vice versa, are of great value.

In this paper, we present tunneling measurements of the density of states in high-R ultrathin films. These were performed in order to investigate the disorder-enhanced e^--e^- interactions in the strongly disordered regime. These interactions shift the energies of the "free electron" states away from the Fermi energy E_F, depressing the density of states near E_F. Electron tunneling measures this density of states and thus, the size of the energy shifts caused by disorder-enhanced e^--e^- interactions. The actual structure in the voltage dependence of the conductance of a tunnel junction has been deemed the Coulomb anomaly.[2] We have investigated films which have structures ranging from islanded to uniformly disordered. In the strongly disordered regime, the Coulomb anomaly depends on R and strongly and systematically on film structure; unlike the weakly disordered regime. Specifically, the anomaly is stronger, grows faster, and extends to higher voltages in the more uniform films. These results imply that the disorder-enhanced e^--e^- interactions depend on film structure, as well as R, in the strongly disordered regime. They are of direct relevance to the 2D SI transition since disorder-enhanced e^--e^- interactions are believed to drive it.[9]

The films were deposited onto fire-polished glass substrates which were held at 8 K and had been previously prepared with Au contact pads and an $\mathrm{Al/AlO}_x$ strip. The Al and AlO_x served as the counterelectrode and barrier, respectively, for the tunnel junction.[12,14] Electron tunneling and transport measurements were performed *in situ* using standard techniques. In a given experimental run, we investigated a series of films with different R by sequential evaporations onto a single substrate without breaking vacuum or warming the cryostat. Throughout this paper we assume $R \propto (n\ell t)^{-1}$, the Drude form, where n is the carrier density in a film and t, the film thickness, is the mass per unit area of the film divided by its bulk density. By choosing different materials and deposition procedures we have been able to produce films with comparable values of n which had different values of ℓ for the same R. Here we present data on Ag, Sn, and Pb films that were deposited directly onto glass substrates and Pb films that were deposited onto $\approx$2 monolayers of quench-condensed Ge. The results of previous transport and superconducting tunneling experiments on these and other similarly deposited films indicate that the carrier densities of these films are comparable to their bulk values at the thicknesses considered in these experiments.[6,7,14,15]

The dependence of R on t is shown in Fig. 1 for all

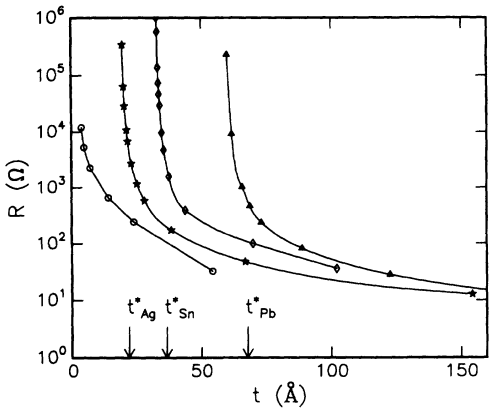

FIG. 1. Sheet resistances measured at 8 K versus thickness for Pb deposited on Ge on glass (circles), and Ag (stars), Sn (diamonds), and Pb (triangles) deposited on glass. The arrows indicate the thickness t^* and sheet resistance R^* at which $k_F\ell \simeq 1$ for each of the films. They are Ag, $t^*_{Ag} = 22$ Å, $R^* = 5$ kΩ, Sn, $t^*_{Sn} = 37$ Å, $R^* = 1.5$ kΩ, and Pb, $t^*_{Pb} = 66$ Å, $R^* = 1.1$ kΩ.

the films considered here. The arrows indicate the thicknesses t^*, or sheet resistances R^*, at which $k_F\ell \sim 1$. Roughly, $k_F\ell \sim 1$ separates the films into two groups with qualitatively different physical processes dominating their electronic transport. The nearly exponential drop of R with t for $k_F\ell \ll 1$ is characteristic of films composed of islands between which electrons must tunnel.[6,16] Moreover, for the Pb films, H_{c2}, the perpendicular upper critical field, has a temperature dependence at low temperatures that indicates that the majority of these islands are weakly coupled to one another and have dimensions that are comparable to t.[14] In the opposite regime, R approaches a $(t)^{-1}$ dependence, indicating that ℓ does not depend on t and the films have a more uniform structure. Again, $H_{c2}(T)$ measurements on these films corroborate this picture. Thus, we can create films that have the same R but must have different structures. For the sake of convenience we will refer to the films with $k_F\ell \ll 1$ as islanded and those in the opposite limit as uniformly disordered. The remainder of this paper addresses the question of how the Coulomb anomaly and, correspondingly, the disorder-enhanced e^--e^- interactions in these films differ in these two extremes for films of equal R.

The Coulomb anomaly in the tunneling density of states in weakly disordered electronic systems was first described by Altshuler and Aronov.[2] These workers demonstrated that the enhanced effective e^--e^- interactions that result from static disorder in a metallic system reduce the tunneling density of states $N(E)$ in the vicinity of E_F. In an ultrathin film, the reduction has the form[2,17]

$$\frac{\delta N}{N_0} = -\lambda_2 \frac{e^2}{8\pi^2\hbar} R \ln\left[\frac{E}{\hbar D(2\pi/t)^2}\right], \quad (1)$$

where N_0 is the density of states for the bulk disordered metal, D is the electronic diffusivity, and E is the energy of an electron measured relative to E_F. λ_2 depends on the form of the effective e^--e^- interactions and for long range Coulomb interactions[2]

$$\lambda_2 = \ln\left[\frac{(2\pi)^2 E}{\hbar D \kappa^4 t^2}\right], \quad (2)$$

where $\kappa = \frac{2me^2 k_F t}{\pi\hbar^2}$. The other factors in Eq. (1) arise from the growth of electron correlations with disorder. Experimental studies[11–13] of the Coulomb anomaly in ultrathin films revealed the predicted[2] logarithmic energy dependence of the density of states. The strength and disorder dependence of the anomaly, however, varied from system to system.[11–13] These discrepancies have been serious enough that the form of the effective e^--e^- interactions (e.g., long or short range) has not been established.[17] The experiments described here addressed this question, and extended measurements of the tunneling density of states into the regime where R approaches and exceeds R_Q, i.e., the regime of the SI transition.

The corrections to the density of states can be obtained from measurements of the conductance of an Al/AlO_x/ultrathin film tunnel junction for which[12]

$$G_j(V) = \int_{-\infty}^{\infty} N_{Al} N_{film} \frac{\partial f(E+eV)}{\partial(eV)} P(E) dE, \quad (3)$$

where E is the electron energy relative to E_F, f is the Fermi distribution, V is the voltage across the junction, and N_{Al} and N_{film} are the densities of states of the Al strip and the ultrathin film, respectively. $P(E)$ is the tunneling probability. We will present the tunneling data in the form

$$g(V) = \frac{G_j(V,R)}{G_j(V,0)} \frac{G_j(2mV,0)}{G_j(2mV,R)}, \quad (4)$$

where $G_j(V,R)$ is the conductance for a film of sheet resistance R and $G_j(V,0)$ is the conductance of the film with the lowest sheet resistance in a given experimental run. This "normalization" process eliminates the effects of $P(E)$ and N_{Al} on the data and thus, for $eV \gg k_BT$, $g(V) \propto N(eV)$.[12] $g(V)$ was symmetric in voltage to $\leq 2\%$, and as a result we will only present data for a single voltage polarity.

The measurements of $G_j(V)$ were performed using standard low frequency ac techniques. The junctions had resistances that ranged from 6 to 100 kΩ. The measured voltage dependences of the $G_j(V)$ were independent of junction resistance. High junction resistances like these were essential for accurate measurements of the tunneling density of states in these films. Careful checks were made to ensure that the measured conductances corresponded to the actual junction conductance and were not influenced by the voltage drops that occur in finite-resistance films. This was simple for the superconducting samples where a direct comparison could be made of the high voltage junction conductance (beyond the superconducting energy gap) with the film in the superconducting state, where the film resistance is zero, and normal state, where the film resistance is finite. For the data presented here, these measured conductances were identical to within our

resolution. In the very worst case, the nonsuperconducting Pb/Ge film (R = 12 kΩ), the film resistance influenced the measurement of the junction conductance at the 10% level. Also, measurements on the samples that were superconductors were either performed at temperatures above the superconducting transition temperature or in high magnetic fields in order to avoid the complicating influence of the superconducting density of states on the data. We note that all of our results are magnetic field independent up to 8 T.

We compare the tunneling conductances for a range of R of two sets of films with very different values of t^* in Fig. 2. The difference between the Pb/Ge films and the Sn films is striking for R >1 kΩ. The Coulomb anomaly is larger and grows significantly faster with R in the Pb/Ge films than in the Sn films. In fact, the growth of the Coulomb anomaly for the Sn films saturates at the highest R. The films are similar in the respect that the Coulomb anomaly depends logarithmically on voltage over some range of voltage. For the Pb/Ge films, this logarithmic dependence extends to approximately 40 meV for films with R < 6 kΩ. For the Sn films, the size of the logarithmic region shrinks with increasing R and the voltage dependence of g beyond this region is weaker, in agreement with previous work.[12] These data unambiguously show that at fixed R the depression in the density of states due to disorder is stronger and extends to higher energies in the Pb/Ge films than in the Sn films.

When these data are combined with the results from the Ag and Pb films, we find that these differences in the Coulomb anomaly are strongly correlated with t^*. By taking the slopes $g' \equiv \frac{\partial g}{\partial \ln V}$ of linear fits of the type shown in Fig. 2, we can measure the strength of the Coulomb anomaly and illustrate this correlation. The results, shown in Fig. 3, demonstrate that the strength of the Coulomb anomaly is greatest in the films with the smallest t^* or, equivalently, largest R^*. At R = 12 kΩ the strength of the Coulomb anomaly in the Pb/Ge films

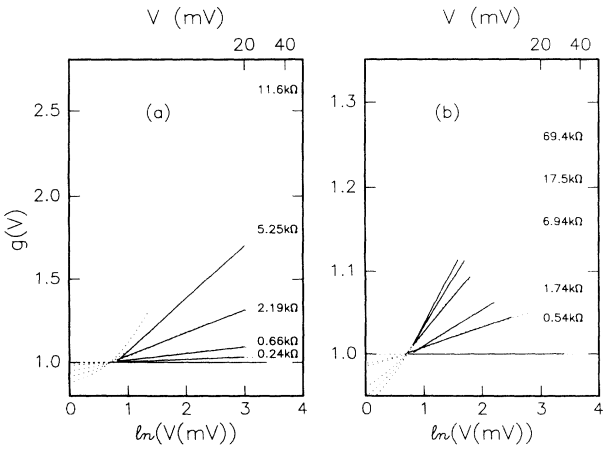

FIG. 2. Tunnel junction conductance normalized to the conductance at 2 mV as a function of voltage on a logarithmic scale for the (a) Pb/Ge and (b) Sn films. Note the different y-axis scales in the two figures. The solid lines are linear fits and the dotted lines are the data.

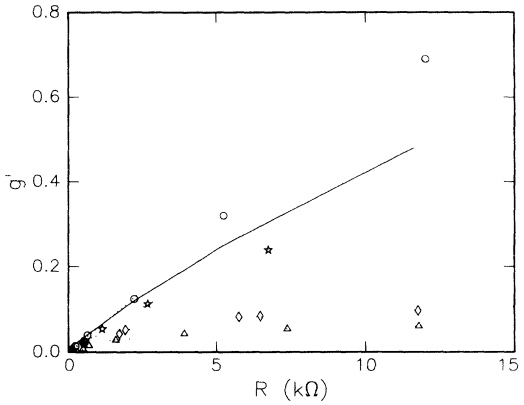

FIG. 3. The slopes of the linear fits shown in Fig. 2 and from other experiments as a function of R for the Pb/Ge (circles), Ag (stars), Sn (diamonds), and Pb (triangles) films. The solid line is the prediction of the theory of Ref. 10 using free electron parameters for Pb and the Drude model.

is more than a factor of 10 larger than that in the Pb films deposited directly on glass. As R approaches and exceeds R^*, the growth in the slope slows significantly, as is evident in the data for the Pb and Sn films in the inset of Fig. 4. More quantitatively, g' grows with the logarithm of R and appears to scale as t^{*-1} as we show in Fig. 4 for the Sn and Pb films. It is important to note that this behavior extends past the resistance quantum, h/e^2 = 25 kΩ. We do not observe a divergence in the slope at ≃ 10 kΩ as has been reported previously.[12] The voltage range over which g is logarithmic shrinks with increasing R as we noted in reference to Fig. 2. Qualitatively similar crossovers to a weaker voltage dependence were observed by other workers[11,12] and were attributed to a crossover from two- to three-dimensional e^--e^- interaction effects.[2] The voltage dependence of our data

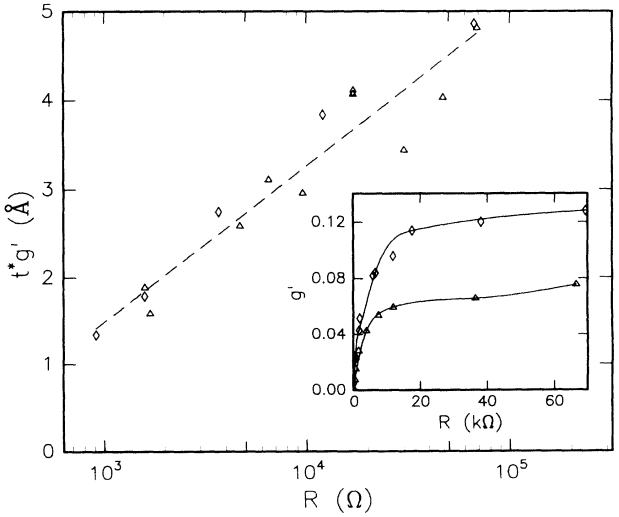

FIG. 4. t^*g' for Sn (diamonds) and Pb (triangles) plotted versus the logarithm of R for the films with $k_F\ell \leq 1$. The dashed line is a guide to the eye. Inset: g' for Sn and Pb plotted on a linear scale.

does not, however, approach the square root dependence at higher voltages that would be expected for a three-dimensional system.[2] We have not yet been able to fit these data to any simple power law form.

Taken as a whole, these data show that the anomaly in the tunneling density of states depends on t^* and, therefore, film structure in the strongly disordered limit. Since disorder-enhanced e^--e^- interactions are responsible for it, we suggest that these interactions must also depend on film structure.

At $R < R^*$, the junction conductances of all of the Pb/Ge, Ag, and Sn films roughly satisfy the predictions of Eq. (1). That is, g' depends nearly linearly on R with a slope that depends weakly on material.[2,17] We can make the most thorough quantitative comparison with the theoretical predictions of Eqs. (1) and (2) in the case of the Pb/Ge film for which the range of R below R^* is largest. The solid curve in Fig. 3 was calculated using the thickness of the Pb deposited on the Ge and the free electron carrier density for Pb. The theoretical curve agrees with the data out to 2 kΩ, at which point the data deviate above the prediction. In this and earlier work that showed that the magnitude of the tunneling density of states followed theory, it has been implicitly assumed that the underlying Ge does not affect the carrier density in the Pb. This agreement supports the validity of this assumption. Deviation from theory at 2 kΩ is reasonable, since the correction to the density of states predicted by this perturbation theory[17] is 45% of the density of states. Earlier work[12] on Sn films quench condensed on glass agreed with theory up to about 1 kΩ. Thus, in the weakly disordered limit, the data agree with the interpretation that this zero bias anomaly stems from a many-body e^--e^- interaction effect in disordered systems.[2,12,15]

It should be noted that this agreement implies that the influence of the counterelectrode on the effective e^--e^- interactions in the film is minimal. This is in accord with some earlier tunneling measurements on Sn films,[12] but disagrees with some made on InO films,[11] for which the screening of the counterelectrode had to be considered in order to explain the magnitude of the observed Coulomb anomaly.[2] We believe that the salient difference between our systems and the InO system is the much larger carrier density in ours than in theirs. This implies that the screening length is much longer in the InO films and, therefore, they are more susceptible to electromagnetic interactions with the counterelectrode. Recent transport experiments on metal/insulator/metal sandwiches that were designed to measure the e^--e^- interactions between metal layers support this explanation.[18,19]

The fact that $g'(R)$ deviates from Eq. (1), for $R > R^*$, is not surprising since the theory[2] only applies to homogeneously disordered systems for which $k_F\ell > 1$. Nevertheless, given the excellent agreement between data and theory at low R and the gradual evolution of the data with increasing R, we feel comfortable in assuming that this Coulomb anomaly still stems from corrections to $N_{\text{film}}(E)$ that result from disorder-enhanced e^--e^- interactions. The logarithmic R and inverse thickness dependences of g' suggest a possible picture for the manner in which e^--e^- interaction effects grow with increasing R in these islanded films. According to Eq. (1), the Coulomb anomaly depends on the form of the interaction potential, through λ_2, and the strength of the e^--e^- correlations, through the other factors in $\delta N/N_0$. Different length scales influence these two factors.[2] For two electrons that interact and exchange an energy E, the length scale associated with the correlations is $L_{\text{corr}} = \sqrt{\hbar D/E}$, i.e., the distance that two electrons diffuse during their interaction time. Typically, L_{corr} is on the order of a few hundred Å at $E \sim 1$ meV, for strongly disordered systems. The longest length scale associated with the Coulomb interaction is determined by the frequency-dependent screening length and it is given by $L_{\text{scr}} = \hbar D\kappa/E$.[2,18] For disordered systems, $L_{\text{scr}} \sim 1\ \mu\text{m} \gg L_{\text{corr}}$.[18] It seems natural, therefore, that the screening properties of the electron gas, which determine λ_2, are primarily governed by the ability of the conduction electrons to diffuse over distances encompassing many islands, and the correlation effects are primarily governed by the diffusion of the electrons on length scales comparable to or less than the size of the individual islands in the film. Since changes in R largely reflect changes in the long length scale properties of the film, we suggest that the growth in g' with R in more islanded films is dominated by the degradation of the ability of the electron gas to screen on length scales greater than the average island size and not by changes in the strength of the e^--e^- correlations. Indeed, if we explicitly assume that the electron correlations do not change in this regime, Eqs. (1) and (2) predict, in agreement with the data, that g' grows with $\ln(R)$ and $(t^*)^{-1}$. The former dependence comes through the λ_2 term [Eq. (2)] and the latter through the linear dependence on R.

The above picture relies on the assumption that these films are uniformly disordered on length scales comparable to L_{scr}. For this to be the case, they must be far from the percolation threshold where the film structure would become inhomogeneous on all length scales.[20] We can test the validity of this assumption by considering how the absolute value of the conductance of the tunnel junctions varies with film sheet resistance. Within a percolation picture, the paths through which electron transport takes place become more and more widely separated as the sheet resistance of a film is increased. Very close to the percolation threshold, only a small fraction of the film participates in transport. Electrons can only tunnel into areas through which electron transport can take place. Since the conductance of a tunnel junction is directly proportional to the area into which the electrons can tunnel, one expects this conductance to change dramatically as the percolation threshold is approached. We see no evidence of this behavior. For example, the junction conductance at 30 mV for the Sn films varies by less than 5% as their sheet resistances change from $\simeq 300$ to $\simeq 70$ kΩ. We take this as strong evidence that these films are not near a classical percolation threshold.

Our results are of direct relevance to systems near the SI transition. A large body of work has shown that the underlying structure of a superconducting film strongly influences the qualitative manner by which it proceeds through the SI transition.[6–8] In islanded films, the transition occurs (with increasing R) through the growth

of fluctuations in the phase of the superconducting order parameter and the eventual loss of long range phase coherence.[6] In more uniform films (e.g., Pb/Ge)[7] the amplitude of the superconducting order parameter decreases with increasing R and in some instances may disappear at the transition.[8] For both cases, theories[9] intimate that the growth of e^--e^- interactions with disorder is primarily responsible for the transition. These models[9] *assume* that the interactions depend on whether a film has a uniform or islanded structure. Our results support this picture and give insight into which interaction forms are appropriate for different experimental systems. In order to reduce the amplitude of the superconducting order parameter, it seems reasonable that the e^--e^- interaction effects must modify the electronic states participating in the formation of Cooper pairs. In Pb and Sn, for example, the majority of these states are 5–10 meV from E_F and thus we expect the amplitude of their order parameters to be degraded,[9] since the Coulomb anomaly extends to those energies. The Pb/Ge films follow this expectation (see also Ref. 15). The Coulomb anomaly in the islanded Pb and Sn films, in contrast, does not affect the "pairing electron" states nearly as strongly and as a result the amplitude of the superconducting order parameter changes very little through their SI transitions.[12] Rather, the degradation of screening on long length scales, as discussed above, leads to the enhancement of intergrain repulsive Coulomb interactions that compete with intergrain Josephson coupling.

In summary, we have measured the Coulomb anomaly in ultrathin high sheet resistance films that have structures ranging from islanded to nearly uniform. In the strongly disordered limit, the strength of this anomaly depends on film structure. This implies that the e^--e^- interactions in these films depend on their structure. The energy scale over which this dependence is apparent is on the order of meV. These results are, therefore, of direct relevance to studies of the metal-to-insulator transition in disordered systems and the 2D SI transition for which the pertinent energy scales are of this order or less.

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