Superconductivity and the electronic density of states in disordered two-dimensional metals

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We have measured the superconducting and normal-state properties of ultrathin quench-condensed Pb and Sn films by electron tunneling and transport. The normal-state measurements illustrate a substantial reduction in the electronic density of states about the Fermi surface. Good agreement with theory is obtained for this reduction. The collapse of T_c can be accounted for by the reduced density of states but some problems stand in the way of a full description of the superconducting behavior.

For a highly disordered metal, it is known that superconductivity disappears in the vicinity of the metal-insulator transition. For a pair wave function $\psi=\psi_0 e^{-i\phi}$, either amplitude reduction (ψ_0) or phase breaking (ϕ) will result in loss of superconductivity, and these two modes manifest themselves differently. In the case of amplitude reduction, T_c and the energy gap Δ_0 both remain well defined and decrease.[1,2] In the case of phase breaking, T_c (onset of superconductivity) remains unchanged,[3,4] but the current carrying capacity disappears, the transition width increases until the material has no region of zero resistance, and the energy gap Δ_0 broadens substantially.[3] These two limiting cases can be accessed in real materials by controlling the material morphology (either homogeneous or inhomogeneous on a length scale less than the coherence length). In two dimensions both limits have been identified, and it has been determined that in the homogeneous case, a simple Eliashberg description of superconductivity is inadequate.[1]

Why Δ_0 and T_c decrease continuously in the homogeneous case in ultrathin films remains an issue, and it is this problem we address. Previous work[5] has suggested Couloumb interactions as a possible cause of these effects. By tunneling and transport measurements we *directly* measure the electronic density of states for ultrathin films and show that it is profoundly altered, presumably by Coulomb interactions. This change in the density of states is compared with recent theories and found to be in good agreement. Using this density of states, we find that the superconducting transition temperature should indeed be reduced an amount which is consistent with that observed experimentally. Some problems remain, however, and these will be discussed.

The measurements described here were performed on films evaporated at low temperature (4–7 K) in a cryogenic evaporator that has been described previously.[6] The tunnel junctions used were of the configuration Al/oxide/disordered superconductor. Prior to cooling down the apparatus, gold contact pads and a narrow Al strip, which was allowed to oxidize for a short time in air, were evaporated onto a fire-polished glass substrate. At low temperatures, a 1–2 monolayer Ge film was evaporated onto the substrate followed by a series of Sn or Pb film evaporations. The Sn and Pb films became electrically continuous at a thickness t_0, ~ 6 and ~ 2 atomic layers, respectively. The conductivity grew nearly linearly with film thickness beyond t_0. The deviations from linearity in the regime where these studies were performed were only a few percent and this convinces us that these films are homogeneous. In calculations of mean free paths of the electrons in these films, we use an effective thickness that is equal to the total equivalent mass thickness of the film minus t_0. The Al oxide and Ge acted as the barrier for the tunnel junction. With this arrangement we evaporated successive films of the metal, Pb or Sn, and measured the sheet resistance as a function of temperature and the conductance of the tunnel junction for each. Changes in the tunnel conductance from film to film can be entirely attributed to changes in the characteristics of the film and not to changes in the Al oxide and Ge tunnel barrier, as the barrier is *identical* in all measurements for a given series of films. The tunnel junctions were of very high quality, showing almost no leakage current due to nontunneling conduction.[1]

The conductance of a tunnel junction can be written

$$G=N_2\int_{-\infty}^{\infty} N_1(E)\frac{\partial f(E+V)}{\partial V}P(E)dE\ , \tag{1}$$

where E is the energy relative to the Fermi energy, f is a Fermi function, V is the voltage across the junction, and N_2 and $N_1(E)$ are the densities of states of the bulk metal electrode, which we take as a constant, and the quench condensed film, respectively. $P(E)$ is the tunneling probability which on the voltage scales here we take to be $\sim$const$=P_0$. At low temperatures the Fermi functions are sharp and Eq. (1) reduces to

$$G=N_2N_1(V)P_0\ .$$

Thus, a measurement of the voltage dependence of the conductance of a bulk metal-insulator quenched-condensed film tunnel junction at low temperatures gives a measure of the density of states of the film as a function

of energy. We will present tunneling conductance data in the normalized form,

$$\tilde{G}=G(R_\square,V)/G(R_\square\approx 0,V)\ .$$

This normalization eliminates the barrier effects and N_2.

Recent experiments and theories have demonstrated that the corrections to the density of states at the Fermi energy of a two-dimensional disordered metal are logarithmic.[7–9] These corrections are due to the enhancement of the Coulomb interactions between electrons due to the reduced diffusivity induced by disorder. More specifically, Altshuler, Aronov, and Zuzin,[10] and Lopes dos Santos and Abrahams[11] have shown for a film of thickness d that these corrections have the form

$$\frac{\delta N}{N_0}=\frac{-e^2}{8\pi^2\hbar}R_\square \ln\left[\frac{E}{\hbar D(2\pi/d)^2}\right]\ln\left[\frac{(2\pi)^2E}{\hbar D\kappa^4 d^2}\right]\ , \qquad (2)$$

where

$$\kappa=\frac{2me^2k_Fd}{\pi\hbar^2}\ ,$$

m is the electron mass, e is the electron charge, D is the electronic diffusivity, and N_0 is the density of states of the bulk disordered metal. For high sheet resistance films, Eq. (2) predicts that the correction to the density of states can be quite substantial and can extend to energies beyond the average phonon energies in the material.

In Fig. 1(a), we have plotted $\tilde{G}$ measured at 4.2 K versus voltage for a series of Pb films of thicknesses from $\sim$1 to 20 atomic layers for 2000 $\Omega/\square > R_\square > 100\ \Omega/\square$. We used the tunnel conductance of a 40 $\Omega/\square$ film for the normalization. The superconducting energy gap observed at lower voltages does not affect this normalization. These data show a striking decrease in the density of states at the Fermi surface as the sheet resistance of these films increases. For example, N_1 (V) $< 0.5N_0$, for a film of 2000 $\Omega/\square$. At low voltages, these data are consistent with a logarithmic rise of N (V). Other data on uniform Sn films show a crossover to $\sqrt{V}$ dependence at higher voltages (>10 mV). We presume this to be a dimensional crossover from two to three dimensions at higher energies.[7] At higher $R_\square$ we observe the density of states to go to zero, signaling an insulator. We emphasize that this is the first measurement which has allowed a direct comparison of the density of states for various $R_\square$. This is possible because the same oxide barrier is used for all films. Previous studies[7,8] have involved multiple junctions.

To compare these data to Eq. (2), in Fig. 2 we have plotted the normalized conductance at 5 mV for a series of Pb films and at 10 mV for a series of Sn films versus sheet resistance. Comparisons at any energy are equally valid and are equally good. We specifically chose these voltages (energies), as they are close to the average phonon energies in Pb and Sn, respectively. It is the density of states at the average phonon energy which is the relevant quantity for superconductivity. The dashed line in the plot gives the predicted behavior using Eq. (2), assuming n denotes the bulk number density of electrons, k_f denotes the free-electron Fermi wave vector, and relating the sheet resistance to the diffusion constant through

$$D=2E_F/ne^2dR_\square\ .$$

These parameters yield $D\simeq 4$ cm^2/s, which implies that $k_Fl\simeq 10$, where l is the elastic mean free path. Since the theory is perturbative, we expect the theory to be valid only out to $R_\square\simeq 0.5\ k\Omega/\square$. In the valid region, the theory agrees quite well with the data for both Pb and Sn.

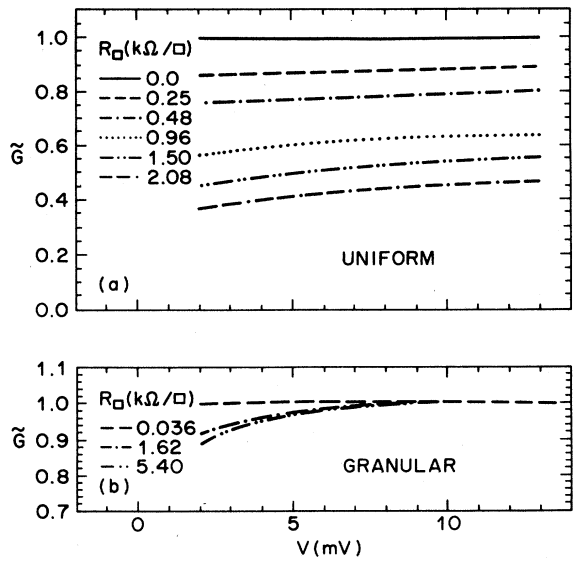

FIG. 1. (a) Normalized tunneling conductance of normal bulk Al—Al_2O_3—Pb film as a function of voltage for a series of uniform Pb films at 4.2 K. (b) Same as (a) for a series of *granular* Pb films at 7.6 K.

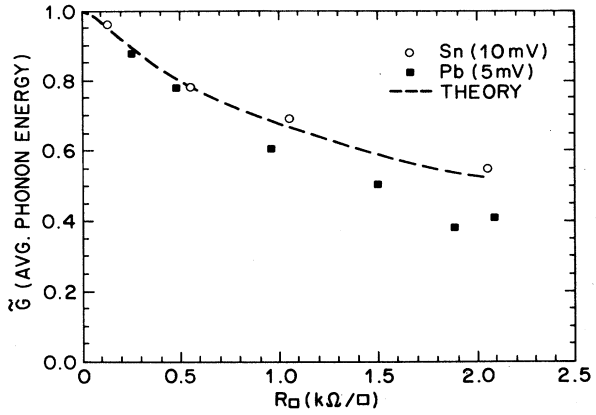

FIG. 2. Normalized tunneling conductance of series of Sn films at 10 mV and series of Pb films at 5 mV as a function of $R_\square$. The dashed line is Eq. (2) using bulk Pb parameters. The curve predicted by Eq. (2) for Sn is nearly identical.

As a contrast to the uniform film behavior, we show $\tilde{G}$ for a series of quench-condensed granular Pb films of thickness $\simeq 120$ Å in Fig. 1(b). These films were prepared in a manner similar to that described in Ref. 3. This is the case of phase destruction of superconductivity where T_c does not change. The corrections to the density of states for these films are qualitatively different from those seen in Fig. 1(a). While $\tilde{G}$ in the disordered granular films does decrease logarithmically near the Fermi energy ($E < 5$ meV) in the same manner as equivalent $R_\square$ uniform films, its overall value is independent of $R_\square$. More importantly, $N_1(E)$ at the average phonon energy depends very little on $R_\square$.

We want to emphasize that both the uniform and granular film data quantitatively agree with Eq. (2) at low voltages.[3] This is true even though one varies $R_\square$ in the two cases in completely different ways. For uniform films one changes $R_\square$ by changing the film thickness at constant D, while for granular films one changes $R_\square$ by changing D at approximately constant thickness. The different overall behaviors can be attributed to the fact that the uniform film is much thinner than the granular film (30 versus 120 Å). Thus, the behavior of the density of states at low energies is determined by the sheet resistance, while the overall behavior is determined by the film thickness.

We now relate these altered properties of the normal state to the superconducting properties of the uniform films. The density of electronic states at the Fermi energy is one of the parameters that determines the superconducting transition temperature in a conventional metal; typically, the higher $N(E_f)$, the higher T_c. We have made measurements of the resistive transitions on uniform Sn and Pb films and have plotted the T_c data in Fig. 3(a). We note two aspects of these data. First, there is a

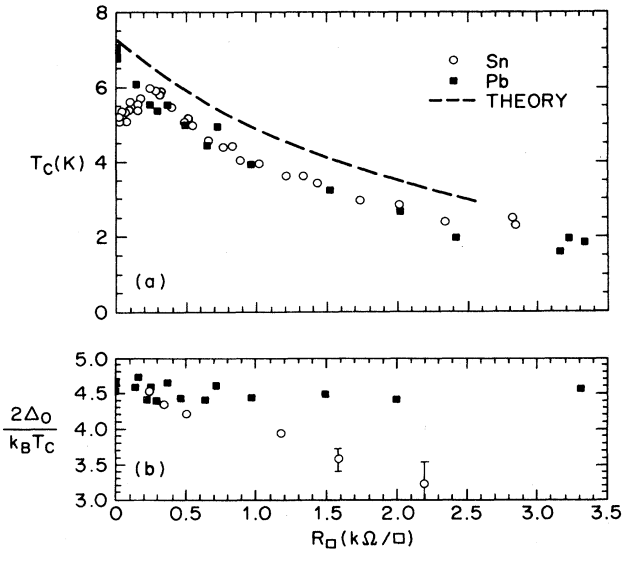

FIG. 3. (a) Superconducting transition temperature vs sheet resistance for Sn (○) and Pb (■) films. (b) $2\Delta_0/k_BT_c$ as a function of sheet resistance for Sn and Pb films. In BCS theory $2\Delta_0/k_BT_c = 3.53$. The Pb data are from Ref. 1.

peak in the Sn data at $R_\square = 250\ \Omega/\square$. We believe that the peak in T_c in the Sn data is due to a structural transition from an amorphous film at high $R_\square$ to a mixed amorphous-granular film at low $R_\square$. In the following, we will only consider the data from Sn films for which $R_\square > 250\ \Omega/\square$. Second, for $R_\square > 250\ \Omega/\square$, the data for the Pb and Sn films are identical to within the scatter. This is an unexpected result.

In Fig. 3(b) we also plot $2\Delta_0/k_BT_c$ versus $R_\square$ for a series of uniform Sn and Pb films. Δ_0 is determined from fits of the tunneling characteristics using a Bardeen-Cooper-Shreiffer (BCS) form for the density of states.[1] Both Pb and Sn behave like strongly coupled superconductors at low $R_\square$ ($2\Delta_0/kT_c \cong 4.5$); a remarkable result for the Sn, which is weakly coupled in bulk crystalline form but perhaps not surprising in the amorphous state. This similarity between Pb and Sn strikes us as interesting, and will be a subject of further investigation. As $R_\square$ increases, $2\Delta_0/k_BT_c$ in Sn films decreases toward the BCS value, while in Pb it remains unchanged. In fact, we cannot rule out the possibility that $2\Delta_0/k_BT_c$ in Sn may be less than the BCS value of 3.53 at high $R_\square$.

If we naively accept that the electronic density of states is reduced as illustrated in Fig. 1(a) and that this is the relevant density of states for the pairing interaction, we would expect that T_c should be reduced. Indeed, a simple calculation taking the density of states $N(E)$ at a value of E corresponding to the average phonon energy $\langle \omega_{ph} \rangle$ does suggest that this reduction is responsible for the T_c depression. As was pointed out earlier, however, the problem with this simple approach is that an Eliashberg description ignoring the altered Coulomb interactions does not work,[1] and the problem needs to be addressed at a more fundamental level. We emphasize that the physics here is very different from that investigated several years ago where disorder produced a smearing in the electronic density of states (in the A15 superconductors, for example). In that case, peaks in the density of states were reduced to a broadened final value and T_c saturated at a new value. In this case, we are not increasing the disorder (reducing the electronic mean free path) and affecting the single-particle density of states. These are many-body effects.

Belitz[13] has considered the effects of disorder-induced reductions in $N(E)$ on superconductivity within a strong-coupling theory for dirty superconductors. In an approximation in which he keeps only diffusive contributions to the dependence of T_c on disorder, he derives an expression for T_c as a function of $\tilde{G}$ near the Fermi energy. We have plotted his result in Fig. 3(a) using the appropriate bulk Pb parameters and relating $\tilde{G}$ to $R_\square$ with Fig. 1(a). The theory predicts a smaller depression of T_c than that which is observed, but it does follow the general trend of the data. Indeed, if in the limit of $R_\square \to 0$ the T_c were ≈ 6.4 K and not 7.2 K, the agreement would be very good. It is possible that the Pb also experiences a transition below 250 $\Omega/\square$ as in the case of Sn and the appropriate T_c for comparison is this lower value of 6.4 K. In addition, by assuming a Debye model for the phonon spectrum, Belitz predicts that for Pb and Sn $2\Delta_0/k_BT_c$

should decrease slightly with $R_\square$; a reduction due to weaker electron-phonon coupling. For Pb, the predicted decrease is greater than that observed, while for Sn the predicted decrease is less than that observed.

Two other theories attribute the reduction of T_c with $R_\square$ to the reduction of the density of states of these films, due to enhanced Coulomb interactions. Fu and Yu[14] derive a set of generalized Eliashberg equations for two-dimensional disordered superconductors that allow for an energy-dependent normal state density of states and a frequency-dependent Coulomb interaction. They solve the equations numerically using Eq. (2) for the normal state density of states of the film and obtain results in qualitative agreement with the $T_c(R_\square)$ data for uniform Pb films. This approach, however, should be limited to low $R_\square$ films for which $\delta N/N_0$ is not too large. It is also difficult to see how the theory of Fu and Yu could describe the ratio $2\Delta_0/kT$ we obtain.

Eckern and Pelzer[15] use a path integral formulation of the problem of dirty superconductors in two dimensions that does not include strong-coupling corrections. They also find T_c to decrease with $R_\square$ by roughly the same amount as we observe in Pb and Sn. $2\Delta_0/k_BT_c$ should decrease with $R_\square$ for Pb films, unlike that observed. They suggest that the source of this discrepancy may be the absence of strong-coupling corrections in their theory. Again, we do not have an explanation for the differences between Pb and Sn.

It is clear, however, that these various theories, as well as simple intuition, suggest that the reduction in T_c for ultrathin films comes as a result of a substantial reduction in the density of states in the energy range relevant to the pairing interaction. The observed reduction in $N(E)$ is in good agreement with theories of the correction due to 2D Coulomb interaction, and this reduction is adequate to account for the reduced superconducting transition temperature T_c. Problems remain, however. It is still not clear why the Eliashberg description of strong coupling breaks down in this highly disordered limit, and why there are marked differences in the ratio $2\Delta_0/k_BT_c$ in the case of Pb and Sn illustrated in Fig. 3(b).

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