Anomalous Behavior of the Electron Density of States in Strongly Disordered Amorphous Composite Indium Plus Indium-Oxide Films

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Tunneling measurements on 3D amorphous composite indium plus indium-oxide films reveal anomalous features in the normal-state electron density of states $N_n(E)$. Above 2 meV, $N_n(E)$ increases linearly with $\ln(E)$ as expected for 2D films, rather than $\sqrt{E}$ as expected for 3D films. However, the magnitude of the $\ln(E)$ term scales with resistivity $\rho_{4.2}$, not sheet resistance $R_{\square}$.

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For a decade there have been numerous studies of localization and electron-electron interaction effects in disordered conductors. One issue is how disorder, combined with the Coulomb interaction between electrons, affects the normal-state density of states $N_n(E)$ near the Fermi level, $E=0$. Theory explains that for weak disorder ($k_F l \gg 1$), $N_n(E)$ has a minimum at the Fermi level $E=0$ and increases as E moves away from $E=0$. A universal result of the theory is that the correction $\delta N_n(E)$ is proportional to $\sqrt{E}$ [$\ln(E)$] for three-dimensional [2D] materials.[1,2] While results on several materials support this theory,[3-7] the tunneling data presented here disagrees systematically with theory and cast doubt on its universality.

The conductor here is amorphous composite indium plus indium oxide (a-InO_x) fabricated by reactive ion-beam sputtering in the presence of a partial oxygen pressure, following Ref. 8. TEM measurements[9] indicate that a-InO_x consists of amorphous indium oxide, in atomic ratio 1 to 1, mixed with occasional crystalline grains of semiconducting In_2O_3. As film resistivity increases, the size of the grains decreases and they become rarer. The films are very reproducible across a single substrate and from day to day. The resistivity $\rho(T)$ and mean-field transition temperature $T_{c0}(\rho)$ are consistent with published results for similar deposition parameters.[10,11]

Many physical properties of a-InO_x have been studied.[9-16] Electric-field-effect measurements[12] on similar films indicate that the $k_F l$ (l is electron mean free path) of our films ranges from 2 to 5, which is close to the strong-scattering regime. The conduction-electron density from Hall effect measurements is about 4×10^{20} cm^{-3} for the range of resistivities studied here. As discussed in Ref. 16, TEM measurements,[9] a study of the metal-insulator transition,[12] and the effect of a transport supercurrent on the superconducting density of states[16] show that a-InO_x behaves like a microscopically homogeneous material rather than a filamentary or granular material, e.g., granular Al.

We obtain $N_n(E)$ from the differential conductance

$G_j(V,T)\equiv \partial I/\partial V|_T$

of $Al/AlO_x/a\text{-}InO_x$ and $Al_{0.99}Mn_{0.01}/AlO_x/a\text{-}InO_x$ normal/insulator/superconductor junctions measured at various temperatures. Incorporating superconducting effects on the density of states into the function $N_1(E)$, we have[16]

$$G_j(T,V)=C\int_{-\infty}^{\infty}dE\,N_1(E)N_n(E)\times[-\partial f(E-eV)/\partial(eV)|_T]\,, \quad (1)$$

$$\approx CN_n(eV) \text{ at } eV\gg\Delta\,, \quad (2)$$

where $N_1(E\gg\Delta)\approx1$, f is the Fermi function, Δ is the order parameter which is less than 1 meV, and C is a constant proportional to the intrinsic conductance of the tunnel barrier. We neglect the thermal smearing represented by $\partial f/\partial(eV)|_T$ in the integral since observed features in $G_j(V)$ are much broader than a thermal energy k_BT (≈0.3 meV at 4 K).

Junctions are fabricated by evaporating an Al or Al(1% Mn) strip on a glass substrate, depositing SiO to define the junction area, oxidizing in air, and finally depositing a cross strip of InO_x. The thickness and $R_{\square}$ of the Al strip is about 2000 Å and less than 1 Ω. Mn is used to suppress the T_{c0} of Al, which is needed to study superconducting properties of InO_x. Data are taken with a conventional ac technique. The typical junction area is about 0.1–0.01 mm^2 and the normal junction resistance ranges from 500 Ω to 200 kΩ. We fabricate several junctions on an InO_x strip at the same time to check the quality of the junctions and homogeneity of the strip.[15] The normalized conductances $G_j(V,T)/G_j(0,T)$ are the same within 1% for all junctions made at the same time.

A small corrections to G_j is needed, in principle, because of the asymmetry of the tunnel barrier.[7,17] However, we observe that G_j for both polarities of bias voltage differ by only 1%–3% up to 100 mV, which is small enough to neglect in the interpretation of data.

Before presenting our main results, we wish to demonstrate that the results are not affected by superconductivity in the a-InO_x strip, or by high current densities in the a-InO_x strip at high junction bias voltages, or by the presence of Mn in the Al counterelectrode. Supercon-

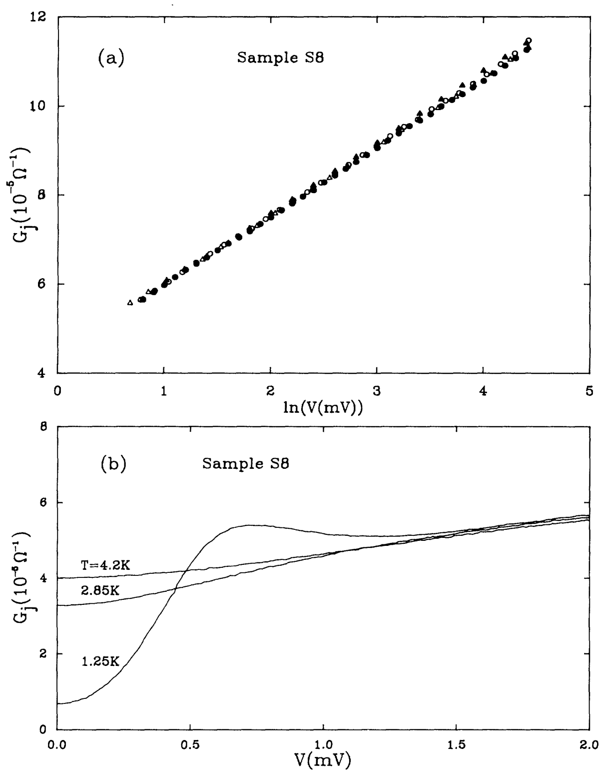

FIG. 1. (a) Measured G_j vs $\ln(V)$ of sample $S8$ above 2 mV at different T and voltage polarity. Open circles, $T=4.2$ K; solid circles, $T=2.85$ K (T_{c0}); open triangles, $T=1.25$ K; solid triangles, opposite polarity at $T=4.2$ K. T_{c0} is the mean-field superconducting transition temperature of a-InO_x film. (b) G_j vs V of sample $S8$ at $V<2$ mV.

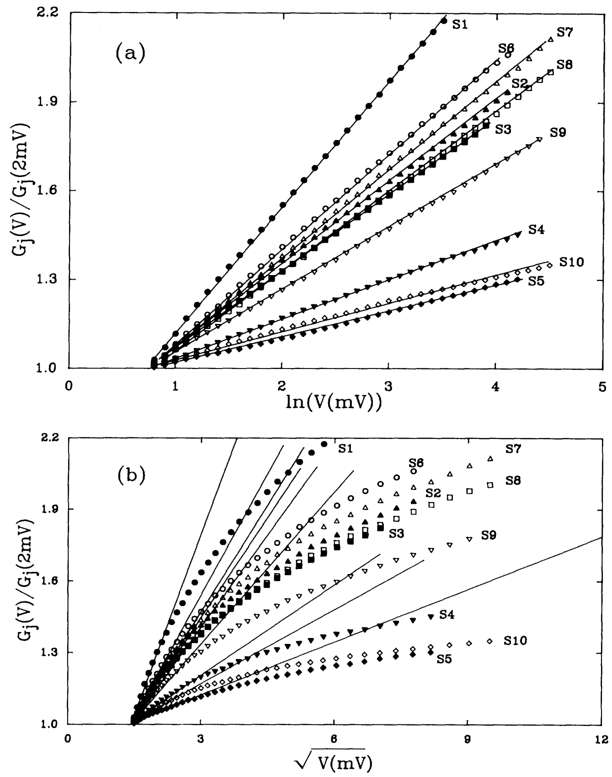

FIG. 2. (a) Measured G_j vs $\ln[V(\text{mV})]$. (b) G_j [shown in (a)] vs $[V(\text{mV})]^{1/2}$. Deviation from $\sqrt{V}$ is clearly shown for all samples.

ductivity serves only the technical purpose of making the a-InO_x strip an equipotential, thereby keeping the current through the junction as uniform as possible.

Figures 1(a) and 1(b) show $G_j(V)$ of a junction of $T>T_{c0}$, $T=T_{c0}$, and $T<T_{c0}$. $G_j(V\le 2\text{ mV})$ at $T<T_{c0}$ clearly shows a superconducting effect, which proves the quality of the junction. At high bias from 2 to 100 mV, the data at the three different T merge into virtually a single curve. This result is strong support that the measured G_j is indeed a measure of the normal-state density of states N_n, and not a stray effect due to distortion of current distribution through the junction (the critical current density of the InO_x strip far below T_{c0} is on the order of 10^9 A/m^2, which is 3–5 orders of magnitude greater than the current density in the junction area up to 100 mV).

In the junction shown in Fig. 1, the a-InO_x strip effectively is an equipotential even in the normal state since $R_{\text{jct}}\approx 30$ kΩ is much larger than the sheet resistance of the a-InO_x strip, $R_\square\approx 0.25$ kΩ. In lower-resistance junctions where the a-InO_x is not an equipotential, we observe that G_j is different above and below T_{c0}, as expected. We also observe that $G_j(V)$ is independent of T below T_{c0}. Hence we obtain $N_n(E)$ from $G_j(V)$ measured at $T\ll T_{c0}$.

Plots of $G_j(V)$ vs $\ln(V)$ are shown in Figs. 2(a) and 2(b), respectively, for many samples with different film thicknesses and resistivities, as given in Table I. Al(1%

TABLE I. Sample parameters: sheet resistance $R_\square(4.2\text{ K})$, film thickness d, residual resistivity $\rho_{4.2}$, dimensional crossover energy $E_{\text{cross}}=4\pi^2\hbar D/d^2$. D is estimated from the free-electron relation, and the electron density $n\approx 4\times 10^{20}$ cm^{-3} is taken from Ref. 12.

Sample	$R_\square$ (kΩ)	d (Å)	$\rho_{4.2}$ (mΩ cm)	E_{cross} (meV)
$S1$	2.80	350	9.8	0.45
$S2$	1.74	350	6.0	0.55
$S3$	1.60	350	5.6	0.79
$S4$	1.16	350	4.0	1.15
$S5$	0.68	350	2.4	1.84
$S6$	4.26	180	7.7	2.17
$S7$	0.28	2200	6.2	0.018
$S8$	0.25	2100	5.3	0.023
$S9$	2.80	180	5.0	3.34
$S10$	1.64	180	2.95	5.67

Mn) was used as a normal electrode for samples $S1$ to $S5$ and pure Al for $S6$ to $S10$ to check the effect of Mn on G_j. No quantitative difference in $G_j(V)$ is observed, as seen in Fig. 2.

The linear increase of $N_n(eV)$ with $\ln(V)$ rather than $\sqrt{V}$ above 2 mV is evident from comparison of Figs. 2(a) and 2(b). A small power of the voltage such as $V^{0.1}$ perhaps fits up to about 30 mV, but $\ln(V)$ is still better. Note that the disorder-induced correction to N_n is not small; it ranges from 30% to 100% between 2 and 100 mV.

Although the $\ln(E)$ dependence suggests that the films are 2D, the magnitude of the $\ln(V)$ correction to N_n scales with $\rho_{4.2}$, not sheet resistance $R_\square$, which suggests 3D behavior. This is illustrated in Fig. 3, which depicts the dependence of the $\ln(V)$ correction on $\rho_{4.2}$ and $R_\square$. Figure 3(b) shows that A, a measure of the correction defined by $d[G(V)/G(2\text{ mV})]/d[\ln(V)]$, remains unchanged while $R_\square$ increases by 20 times. On the other hand, A is linear with $\rho_{4.2}$ within 10% uncertainty as seen in Fig. 3(a). Specifically, $N_n(E) \approx N_n(2\text{ meV}) \times \{1+A\ln[E/(2\text{ meV})]\}$, where $A \approx 0.042\rho_{4.2}(\text{m}\Omega\,\text{cm})$.

A crossover from 2D ($\ln V$) to 3D ($\sqrt{V}$) behavior in the weak-scattering limit is expected when eV exceeds $E_{\text{cross}} \approx \hbar D(2\pi/d)^2$ such that the electron diffusion distance in a characteristic quantum time $\hbar/E_{\text{cross}}$ is roughly the film thickness. (Although this result is often quoted without the 2π, the observed crossover in polycrystalline indium oxide suggests 2π should be included.[5]) Values of E_{cross} for our films range from 0.6 to 2 meV, except $S9$ and $S10$ as given in Table I, showing that $N_n(E)$ should show 3D behavior above 2 mV. Neglecting the 2π lowers E_{cross} and makes our results even more surprising. No crossover is observed, even including $S9$ and $S10$.

For a quantitative comparison with results on other materials which show a $\sqrt{E}$ dependence, we draw a line to fit the initial rise of data (roughly between 2 and 7 mV) by $(eV)^{1/2}$ as shown in Fig. 2(b), and determine the correlation gap E_c from[18]

$$N_n(eV) = N_n(0)[1+(eV/E_c)^{1/2}]\,. \qquad (3)$$

The magnitude of E_c estimated in this crude fashion is comparable with other materials,[4-6] although the energy dependence of $N_n(E)$ is distinctly different.

Powers other than $E^{0.5}$ are observed in some materials. $E^{0.6}$ is seen in 3D Au-Ge mixtures,[3] and a crossover from $E^{0.5}$ to $E^{0.3}$ at $E \approx E_c$ is reported for amorphous 3D Nb-Si alloys.[4] A crossover from $\ln(E)$ to $\sqrt{E}$ is observed in polycrystalline indium oxide at E_{cross}. However, there are no other reports of $\ln(E)$ behavior in 3D materials besides the present work.

Because a-InO_x is a multiphase material, the issue of homogeneity is especially important. There is considerable circumstantial evidence that the films are homogeneous. Tunneling data below T_{c0} show strong super-

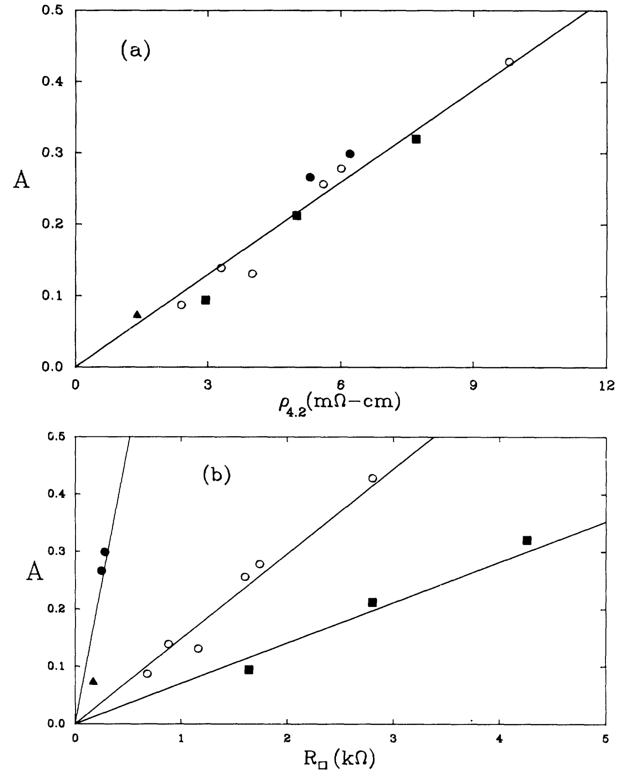

FIG. 3. (a) A, slope estimated from Fig. 2(a), vs $\rho_{4.2}$. Solid squares, 180 Å; open circles, 350 Å; solid triangle, 840 Å; solid circles, 2200 and 2100 Å. (b) A [shown in (a)] vs $R_\square$. This clearly shows that the magnitude of the $\ln(V)$ correction correlates with $\rho_{4.2}$, not $R_\square$.

conducting structure indicating that the tunneling current is going into the superconducting amorphous-indium-oxide component rather than the occasional crystalline In_2O_3 crystallites. Studies of the resistive transition[11] show that its width can be understood entirely in terms of the Kosterlitz-Thouless transition, showing that effects of inhomogeneity on the transition width are negligible. Furthermore, the effect of a supercurrent in the a-InO_x strip on the conductance of the tunnel junction is in quantitative agreement with the dirty-limit theory based on the measured film resistivity.[15,16] This would not be true if the film were very granular, like granular Al, or filamentary.

One might argue that the film is somehow layered with thin conducting regions between insulating layers, so that the film is always 2D regardless of its thickness. This is an unlikely explanation of the data. In this view, the conducting layers must be thinner than 20 Å to make E_{cross} larger than 100 mV. (If the 2π should not be included in the definition of E_{cross}, then the thickness must be less than 4 Å.) It is hard to understand how such thin continuous layers could be formed in such a regular way from sample to sample independent of deposition conditions.

While our samples are outside of the critical region near the metal-insulator transition, it is interesting to note theoretical progress inside the critical region where both localization and Coulomb interactions between electrons are important. McMillan[18] proposed a scaling theory that incorporates both localization and interaction effects by introducing two scaling parameters, a Coulomb coupling constant κ and conductance g. He predicts the singularity given in Eq. (3) by assuming that κ and g are related to $N_n(0)$, in which the singularity in N_n plays an important role in his scaling argument, although the validity of this assumption has been questioned.[19,20] Also Gefen and Imry[21] provided a similar scaling argument taking into account both localization and Coulomb interactions between electrons, and they predict a deviation from $\sqrt{E}$ dependence at $E \gg E_c$. Finkelstein[20] performed a field-theoretical renormalization-group calculation, taking full account of the interaction with the localization effect suppressed. Later, Castellani *et al.*[22] rederived Finkelstein's results by perturbative treatment, and found $N_n(E) \approx E^{1/4}$ in 2D, not logarithmic, when the interaction is long ranged. A corresponding result in 3D is not available.

At this moment, no theory provides a proper interpretation of the data. It is fair to say that the various theoretical descriptions in the strongly scattered limit have not reached a general consensus yet and more data are needed.

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