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Metal-Insulator Transition in Granular Aluminum

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The metal-insulator transition in granular aluminum has been observed. The resistivity has been varied over six orders of magnitude from metallic to a material where the conduction is via variable-range hopping. As the transition is approached from the metallic side, tunneling measurements have shown a $V^{1/2}$ dependence of the density of states, in agreement with recent theories.

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The metal-insulator transition has been a subject of study for decades.[1] Recently, the nature of this transition and the concept of minimum metallic conductivity has come under scrutiny as the concepts of localization[2] and electron correlation[3] evolve. Recent results[4] on two-dimensional (2D) systems illustrate the importance of these effects. We report here tunneling measurements into granular 3D Al films in the vicinity of this transition. We find that Coulomb effects[3] are dominant approaching the transition and that large square-root singularities in the electronic density of states correlate with electron diffusivity. The details of the density of states are found to be in agreement with the theory of Altshuler and Aronov[5] and with the recent theory of the metal-insulator transition of McMillan.[6]

The measurements reported here are for samples with resistivities at 4.2 K ranging from $\sim 10^{-7}$ to 1 Ω cm. The samples consist of "granular aluminum" films, i.e., Al films prepared in an oxygen atmosphere. There is substantial literature on the preparation and properties of these films.[7,8] Our films were prepared by evaporation from resistively heated filaments in an oxygen partial pressure ranging from $\sim 5\times 10^{-7}$ to 2×10^{-4} Torr. In addition to maintaining constant pressure during evaporation it was also imperative that the evaporation rate be kept constant, as a small variation in rate here resulted in inhomogeneities in the film. The samples reported here were evaporated at a rate of ~20 Å/sec and had thicknesses ranging from 100 to 5000 Å. All samples consisted of a bottom film of clean Al, subsequently oxidized to form the tunnel barrier, and a cross stripe of the granular Al. With suitable lead configurations the film resistance and the junction resistance could both be measured using four terminal probes.

The dependence of the superconducting transition temperature T_c with resistivity $\rho(4.2\text{ K})$ is shown in Fig. 1. These results are in agreement with previous investigations[7-10] of the superconductivity of granular Al. With increasing resistivity, the T_c is observed to rise to 2.2 K, at which point it saturates. With increasing ρ, T_c remains relatively constant and then begins to broaden substantially and decrease. The tunneling measurements reported here indicate that correlated with this decrease in T_c is a decrease in the density of states $N(E)$ at E_F. The metal-insulator transition occurs when $N(E_F)$ goes to zero (and superconductivity disappears). From Fig. 1 this apparently occurs at a resistivity $\rho \gtrsim 10^{-1}$ Ω cm

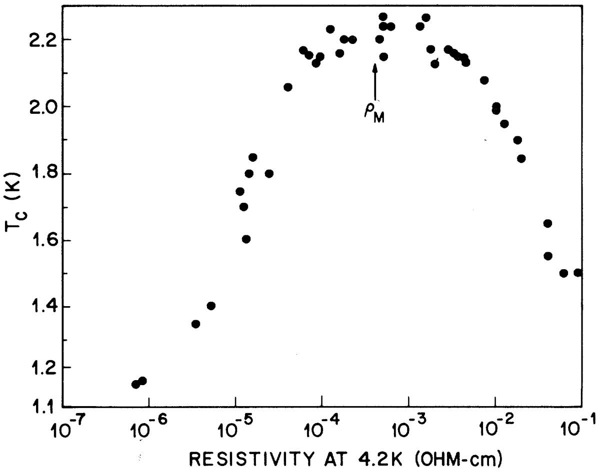

FIG. 1. T_c vs resistivity at 4.2 K. ρ_M is the value of the resistivity where $k_F\lambda = 1$. λ is the mean free path.

and this conclusion is corroborated by the tunneling results. The point labeled ρ_M is where $k_F\lambda = 1$, with λ being the mean free path determined from the resistivity. It is in this region that Mott[1] anticipates an abrupt metal-insulator transition with the conductivity dropping to zero. Although it is clear that nothing precipitous happens here, it is at this point that there are changes in the material which ultimately result in the metal-insulator transition at a resistivity three to four orders of magnitude higher.[11]

At even higher resistivities (1 Ω cm and above) the $\rho(T)$ demonstrates the exponential behavior expected for random systems with localized electrons, i.e., $\log \rho \propto T^{-1/4}$, indicating variable-range hopping from localized sites. We believe these samples are on the insulating side of the metal-insulator transition. This result is intuitively sensible in view of Fig. 1, as it is difficult to imagine superconductivity well into the insulating region. The nature of the gradual destruction of superconductivity with increasing resistivity (or decreasing diffusivity) is not clear at the moment. It is clear, however, that such a correlation should exist.

The central result of this paper is the measurement of the tunneling density of states $N(E)$. It is predicted[5,6] that the Coulomb corrections will be evident in this quantity as square-root singularities, symmetric about E_F. Within smearing factors of approximately a few kT, a measurement of the conductance dI/dV of a metal-insulator-metal tunnel junction is a determination of this density of excitations in the metal. We have chosen a superconductor as the counter electrode in these measurements and have performed the measurements at 0.95 K where both films are in the superconducting state. This was chosen because the observation of the superconducting energy gap is strong evidence for the quality of the tunnel junction and for the claim that tunnel injection is indeed the form of charge transfer. On the other hand, the superconducting corrections to $N(E)$ from dI/dV measurements are rather minor on the voltage scales of these measurements and so the data will be presented as a measure of dI/dV and interpreted as the density of excitations $N(E)$.

For the films with resistivities $< 10^{-4}$ Ω cm, the junctions displayed the usual tunnel characteristics. The superconducting energy gap of both the Al counter electrode and the granular Al were extremely sharp and the tunnel conductance dI/dV showed a slowly varying parabolic shape representative of barrier-shape[12] effects. With increasing resistivity "zero-bias anomaly" increased in strength and dominated the conductance curves above the superconducting energy gap. We note that qualitatively similar behavior was seen in the pioneering work of Abeles.[8] Anticipating the theories of Altshuler and Aronov[5] and McMillan,[6] we plot the normalized conductance for selected junctions as a function of $V^{1/2}$ in Fig. 2. It is seen that beyond the voltage region, where the conductance is dominated by the superconducting energy-gap structure, dI/dV varies as $V^{1/2}$, a result similar to that observed by McMillan and Mochel.[13] Altshuler and Aronov,[5] considering electron-electron interactions in the weakly scattered limit (where $k_F\lambda > 1$) showed that there was a repulsion in the density of states symmetrically about E_F with a change in the density of states $\delta N(E) \propto E^{1/2}$, a result consistent with the observations here. These measurements are on films, however, in the *strongly* scattered limit. McMillan,[6] considering the electron-localization problem and the metal-insulator transition, concluded that localization, correlation, and screening all had to be included on an equal footing. In the limit of low diffusivity (strong scattering or high resistivity), correlation effects become very important and the screening length diverges at the metal-insulator transition. On the metallic side of the transition, he predicted a density of states variation,

$$N(E) = N(0)[1 + (E/\Delta)^{1/2}], \qquad (1)$$

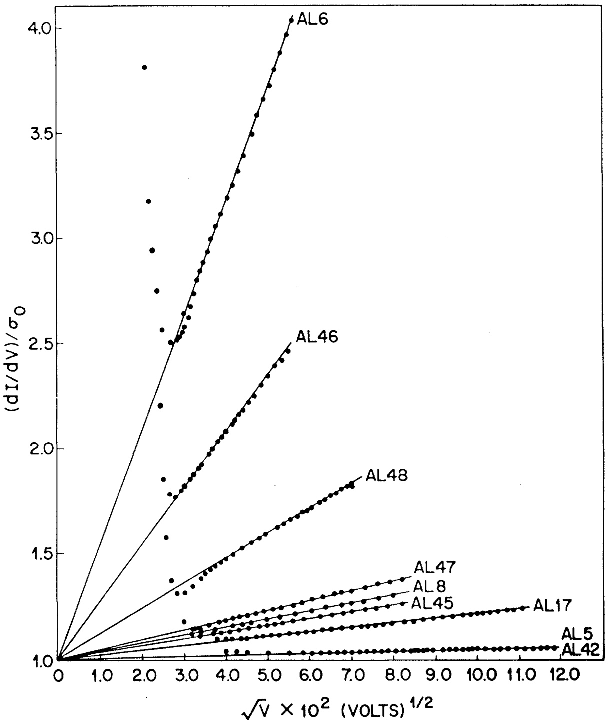

FIG. 2. The measured normalized conductance of selected samples as a function of $V^{1/2}$. At lower bias, the superconducting energy-gap structure dominates and has been omitted. The sample configurations are Al–insulator–granular-aluminum and the resistivities of the various samples are AL6, 4.26×10^{-2} Ω cm; AL46, 5.95×10^{-2} Ω cm; AL48, 1.89×10^{-2} Ω cm; AL47, 4.43×10^{-3} Ω cm; AL8, 9.84×10^{-3} Ω cm; AL45, 2.05×10^{-2} Ω cm; AL17, 3.20×10^{-3} Ω cm; AL5, 6.15×10^{-4} Ω cm; and AL42, 3.28×10^{-4} Ω cm.

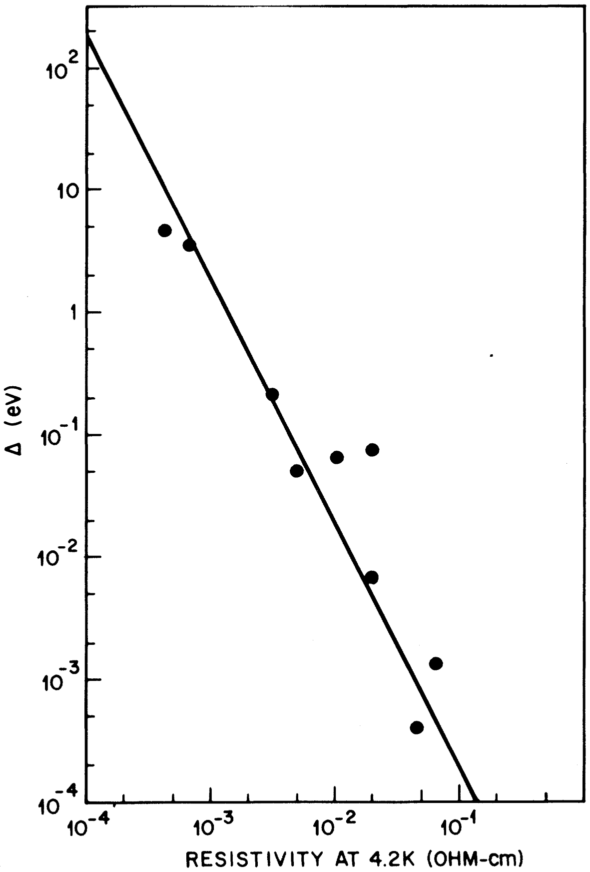

FIG. 3. Correlation gap Δ extracted from the data of Fig. 2 with use of Eq. (1). The straight line is a line of slope − 2 suggesting a quadratic power-law dependence of Δ on resistivity.

a result similar to that of Altshuler and Aronov. Here Δ is the "correlation gap," an energy corresponding to the screening length. With increasing resistivity, the screening length increases, and diverges at the metal-insulator transition, while Δ collapses to zero. On the insulating side of the transition, Δ again increases. This collapse of Δ with diffusivity is clearly seen in Fig. 2, as the slope of these curves is a measure of 1/Δ. An analysis of these conductance curves with use of Eq. (1) yields numerical values for Δ ranging from several electron volts at ρ_M (the screening length is $\sim a_0$) to less than a millielectronvolt just before the transition. This dependence is shown in Fig. 3 for the conductance curves of Fig. 2. We believe that the scatter in the data is due to inhomogeneities in the film, because tunneling is a surface probe while a measurement of resistivity probes the bulk. The scatter in the data represents our ability to control the oxide concentration throughout the thickness of the film. There is nevertheless a distinct inverse dependence of Δ on ρ and the straight line drawn in the figure has a slope of 2. The data is not of sufficient quality for one to assert that the power-law dependence of Δ with ρ has been determined, but it certainly is consistent with a $1/\rho^2$ dependence. Altshuler and Aronov determined that Δ should vary as $1/\rho^3$ in the weak scattering limit. In the strongly scattered limit, McMillan's model anticipates an exponent of 2 (Ref. 14) and the experimental results are certainly consistent with this power. At any rate, the qualitative observation of the collapse of this energy Δ with increasing resistivity is in agreement with the theory of McMillan. Although it would be more correct to plot Δ versus resistivity at $T=0$ K, it is difficult to measure ρ at lower T because of the superconductivity. From $\rho(T)$ measurements we anticipate the correction due to this approximation to be small even near the metal-insulator transition. On the logarithmic scale of Fig. 3, it would be of no consequence.

In summary, we have observed the metal-insulator transition in granular Al. Coming from the metallic side it is heralded by an increasing zero-bias anomaly which has a $V^{1/2}$ dependence. Analysis of the data in terms of McMillan's theory of the metal-insulator transition suggests a diverging screening length at the transition, signaled by a collapsing energy Δ. Tunneling measurements directly determine this parameter Δ and suggest a power-law exponent of -2 for the dependence of Δ on the resistivity.

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