Localization and Electron-Interaction Effects in the Magnetoresistance of Granular Aluminum

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Measurements of the magnetoresistance are reported for a series of granular aluminum specimens on the metal side of the metal-insulator transition. A clear separation can be made between the effects of localization and those of electron-electron interactions. Sufficiently far on the metal side localization effects dominate. At high fields, interactions account for the observed dependences on resistivity and temperature, both of which are absent in the localization theory.

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The phenomena which occur on the metallic side of the metal-insulator transition have recently been investigated intensively.[1] Theories of electron localization[2] as well as those related to electron interactions[3] have been invoked to describe the results. Both approaches often lead to similar descriptions, and it is then difficult to distinguish the effects of the two mechanisms and to assess their relative importance.

The magnetoresistance is among the few properties with decisively different behavior depending on whether it is primarily the result of localization or of electron interactions. In both cases the change in conductivity with magnetic field, $\Delta\sigma = \sigma(H,T) - \sigma(0,T)$, is, for three-dimensional systems, expected to be linearly related to H^2 for low fields and to $\sqrt{H}$ for high fields, but localization leads to a negative magnetoresistance[4] (positive $\Delta\sigma$) while electron interactions lead to a positive magnetoresistance.[5,6]

A recent experiment on phosphorus-doped silicon shows a predominantly positive magnetoresistance, with properties which are to a large extent consonant with those expected from electron interactions, and with the localization-related behavior only indirectly inferred.[5] We report here measurements on granular aluminum which allow both effects to be clearly identified and distinguished. The magnetoresistance is negative at all temperatures and in all specimens which have been measured, so that we conclude that localization effects dominate. An analysis in terms of the localization theory of Kawabata[4] and the electron-interaction theory of Lee and Ramakrishnan and co-workers[5,6] shows that the interaction effects become increasingly important as the metal-insulator transition is approached,[7] although their positive contribution to the magnetoresistance never becomes sufficiently large to overcome the negative localization component.

The specimens are made by electron-beam evaporation of pure aluminum in the presence of a small amount of oxygen. They are similar to those which we have previously described,[8,9] and consist of grains of aluminum, about 30 Å in size, surrounded by varying amounts of Al_2O_3. The film thickness is about 1 μm so that the specimens are three dimensional. Heat-capacity experiments[8] have shown that the specimens have properties to a large extent like those of homogeneous metals, so that, although there may be disorder-related percolation effects, gross inhomogeneities do not play an important role.

With increasing amounts of oxide between the grains the room-temperature resistivity ρ_{RT} increases. For values of ρ_{RT} below 10^{-2} Ω cm the specimens are metallic, although they may show signs of the approach to the metal-insulator transition. Between about 10^{-2} and 2×10^{-2} Ω cm there is an intermediate regime in which the resistance varies approximately logarithmically at low temperatures. For higher ρ_{RT} the resistance varies exponentially and the specimens become insulators as T goes to zero.[9]

Most of our measurements were made in a magnetic field perpendicular to the plane of the specimens. Two samples were also measured in a parallel field. No difference was observed.

In the metallic and intermediate regimes the specimens are superconducting, with values of the upper critical field at $T=0$ constant at about 3.6 T.[10] From the constancy of the critical field at the value of the paramagnetic limit we infer that at this field the destruction of superconductivity is the result of its disappearance in the grains themselves, and not just the result of the quenching of the coupling between the grains.[11] The magnetoresistance which we observe is still increasing at our highest field (9.2 T), and the fractional change is greatest in specimens with high resistivity which show no evidence of superconductivity. We therefore rule out the possibility that the magnetoresistance effects which we observe are related to either pair formation or quasiparticle tunnelling associated with superconductivity.

At high fields and low temperatures the localization theory[4] predicts a unique linear relationship between $\Delta\sigma$ and $\sqrt{H}$, with a slope $A \equiv d\sigma/d\sqrt{H}$ independent of temperature and specimen resistivity, in the region of the theory's applicability, i.e., for $k_F l \gg 1$. (k_F is the Fermi wave vector and l the electronic mean free path.)

The electron-interaction theory[5,6] also predicts a linear relationship between $\Delta\sigma$ and $\sqrt{H}$, but with a slope of opposite sign, which depends on $k_F l$ and on the Hartree factor F which is between 0 and 1.[12] For pure bulk aluminum $F = 0.54$. Lee and Ramakrishnan[6] have shown that the two contributions to A simply add and that for the combination of both effects A should be proportional to $1 - 1.0F(m^*/m)^{1/2}(k_F l)^{-1/2}$, where m^*/m is the effective mass ratio.

We illustrate the relation between $\Delta\sigma$ and $\sqrt{H}$ in Fig. 1 for a specimen whose room-temperature resistivity is 8×10^{-3} Ω cm. In Fig. 2 we show the values of A which we observe at the lowest temperature of our measurements, i.e., near 0.3 K.[13] It is apparent from the figure that A is indeed roughly independent of ρ_{RT} in the metallic regime, although at a value which is about three times that of Kawabata. For higher values of ρ_{RT} the value of A drops, and we ascribe the change to the increasing admixture of the component of opposite sign which results from electron interactions.

The term which describes the decrease of A should be proportional to $F/\sqrt{l}$ and hence to $F\sqrt{\rho}$. The solid line in Fig. 2 is drawn in accord with this relation, for a constant value of F. For $m^*/m = 1.4$, $k_F = 1.75$ Å^{-1}, and $\rho = \rho_{RT}$ it corresponds to a value of $F^2/\rho l$ of 7.0×10^9 Ω^{-1} cm^{-2}. For $\rho l = 9\times10^{-12}$ Ω cm^2,[14] the value of F is 0.25.

It is a consequence of the localization theory[4] that at a fixed high field (in the region where $\Delta\sigma$ varies as $\sqrt{H}$) the value of $\sigma(H, T)$ should be independent of temperature. The interaction theory, on the other hand, predicts that at a fixed and sufficiently high field $\sigma(H,T)$ should be proportional to $\sqrt{T}$. The theory of Ref. 6 shows that the two effects are additive. A graph of $1/R$ as a function of $\sqrt{T}$, taken from Fig. 1, is shown in Fig. 3. It exhibits the linear relationship expected from the theory. This temperature dependence is to be contrasted with the faster, approximately logarithmic dependence in zero field[9] which includes both the contribution from localization and that from interaction effects.[12,15]

According to the interaction theory,[5,6] at a fixed high field, $(1/\sigma)d\sigma/d\sqrt{T} = 0.12T_F^{-1/2}(k_F l)^{-3/2} \times (\frac{4}{3} - F)$. This relationship allows a calculation of the slope of the line on Fig. 3. For $\rho l = 9\times10^{-12}$ Ω cm^2, $F = 0.25$, and $\rho = \rho_{RT} = 8\times10^{-3}$ Ω cm, we find $(1/\sigma)d\sigma/d\sqrt{T} = 0.005$, while the experimental value is 0.2. We see that although the form of temperature dependence is correctly predicted, there is poor agreement with the prefactor. We do not know whether the disagreement is the re-

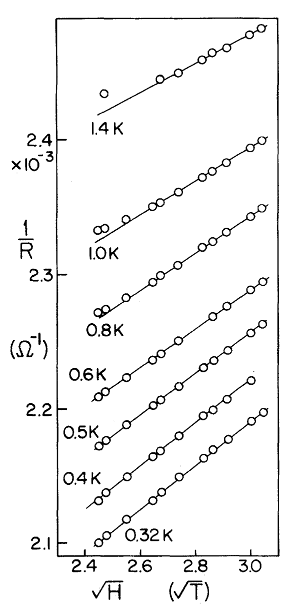

FIG. 1. $1/R$ vs $\sqrt{H}$ for specimen 8. $\rho_{RT} = 8\times10^{-3}$ Ω cm.

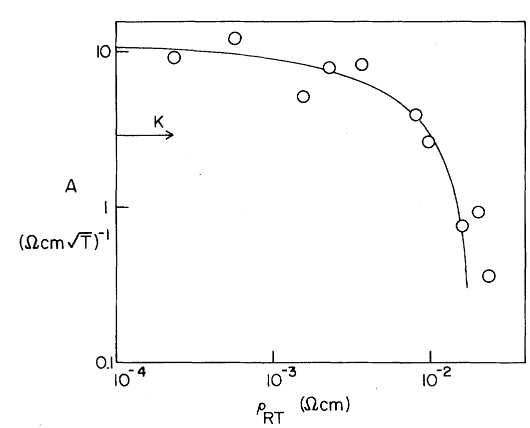

FIG. 2. Log-log plot of $A \equiv d\sigma/d\sqrt{H}$ as a function of the room-temperature resistivity ρ_{RT}.

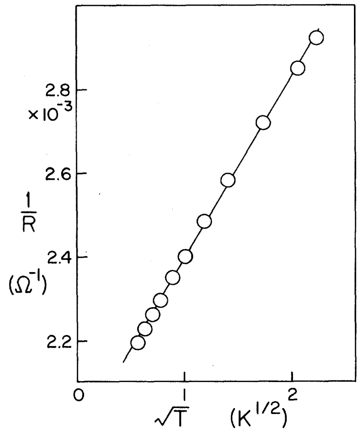

FIG. 3. $1/R$ vs $\sqrt{T}$ at a field of 9.2 T for specimen 8.

sult of a deficiency in the theory of the interactions or of the localization.

We now turn to the consideration of the low-field region where $\Delta\sigma$ is proportional to H^2. Figure 4 shows the quantity $\Delta R/R_0\Delta H^2$ vs T, where $R_0 \equiv R(0,T)$. According to Kawabata[4] $\Delta\sigma/\sigma_0$ is proportional to $(\tau_e/\tau)^{3/2}(\omega_c\tau)^2$, where τ_e is the inelastic and τ the elastic scattering time, $\omega_c = eH/mc$, and $\sigma_0 = ne^2\tau/m$. If τ_e is proportional[16] to $1/T$ this leads to values of the quantity plotted in Fig. 4 proportional to $T^{-3/2}\rho^{-1/2}$, which is clearly inconsistent with our results.

A further difficulty is that in Kawabata's theory the crossover between the H^2 and the $\sqrt{H}$ regions occurs at a field which is proportional to the specimen resistivity. This should lead to an increasing $\sqrt{H}$ region as the specimen resistivity decreases, which is opposite to the dependence we observe. These discrepancies indicate that the temperature dependence of τ_e is more complicated than the simple $1/T$ form often assumed[16] and that in addition τ_e may also depend on resistivity.

We note in this connection that according to Refs. 4 and 5 the contribution to $\Delta\sigma$ at low fields from interaction effects is smaller than that from localization by a factor $(k_F l)^2$. In our lowest-resistivity specimens interaction effects on $\Delta\sigma$ should therefore be negligible.

We conclude that granular aluminum behaves differently from phosphorus-doped silicon in that the localization effects dominate the magnetoresistance, although the interaction effects are crucial for understanding the variation with temperature and resistivity. The fact that localization-related effects play a more important role has made it possible for us to identify a region ($\rho_{RT} \lesssim 2\times 10^{-3}$ Ω cm) where there is no observable

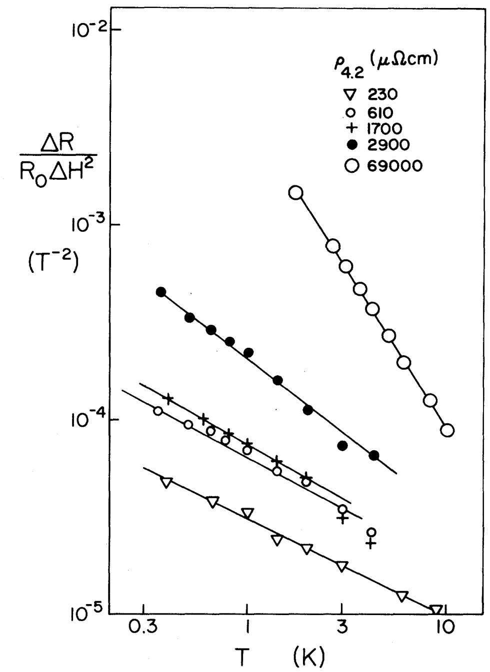

FIG. 4. $\Delta R/R_0\Delta H^2$ vs T for several specimens.

contribution from interactions to the coefficient A. The increasing role of interactions for higher values of ρ_{RT} is then easily distinguished and continues to the vicinity of the metal-insulator transition.

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Minimum Metallic Conductivity and Thermopower in Thin Palladium Films

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The effects of two-dimensional electron localization have been studied by measuring thermopower and resistivity of thin (18–25 Å) films of palladium. "Metallic" samples have a small thermopower which tends to zero as $T \to 0$. Insulating samples have large thermopower which increases as $T \to 0$. The metal-insulator transition in Pd films thus involves the opening of a gap at the Fermi energy. With use of the thermopower to define the metal-insulator transition a critical resistivity of ~ 30 kΩ/□ is found.

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There has been renewed interest in the problem of electron localization in two dimensions due to a number of recent theoretical advances[1-4] and considerable experimental work.[5-8] Rather than a minimum metallic conductivity of $R_{2D}^{-1} \approx (30\,000\ \Omega/\Box)^{-1}$, single-parameter scaling theories predict that all states are localized in two dimensions.[1] A logarithmic temperature increase in the resistivity for low-$R_\Box$ samples crosses over smoothly to an exponential increase (with decreasing temperature) for $R_\Box > R_{2D}$.[2] Another picture suggests that the logarithmic behavior for $R_\Box < R_{2D}$ may be understood in terms of electron-electron interactions.[3] Experiments on thin metal films have been interpreted as evidence for either localization or interaction effects[6,8] while recent experiments on electron inversion layers tend to support the interaction picture.[7]

In this Letter we present thermopower measurements which clearly indicate the opening of an energy gap as $R_\Box(T)$ increases above R_{2D}. In terms of the thermopower there is a sharp distinction between the metallic and insulating states. We find that this metal-insulator transition occurs quite rapidly in the resistance region of R_{2D}. Since the temperature dependence of the resistance of these films is virtually indistinguishable from that in previous studies of thin films we believe our results are quite general. While resistance (and magnetoresistance) measurements probe the density and mobility of the carriers, the thermoelectric power (S) probes their energy distribution. If the energy distribution falls to zero width about the Fermi energy as $T \to 0$ we will have $S \to 0$. Such is the usual case for metals as well as for variable-range hopping in three and two dimensions.[9] If S increases as $T \to 0$ there is evidence for the existence of an energy gap.

We find a striking qualitative change in the thermopower as the thickness of our films is lowered so that the resistivity crosses R_{2D}. For films with $R_\Box < R_{2D}$ the thermopower is approximately independent of $R_\Box$ and decreases with decreasing temperature. For films with $R_\Box > R_{2D}$ the low-temperature thermopower increases with decreasing temperature, with higher-resistivity films showing a larger thermopower. The thermopower measurements therefore indicate that the density of states at the Fermi level vanishes

