Conduction in granular aluminum near the metal-insulator transition

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The electrical resistance has been measured on granular aluminum specimens with room-temperature resistivities between 1.5×10^{-3} and 1.3×10^{-1} Ω cm, from 0.3 K to room temperature, in magnetic fields up to 9 T. The results show the importance of electron correlation effects on both sides of the metal-insulator transition. The Mott hopping law is not observed in any specimen over the whole temperature range. The results suggest the possibility of superconductivity in the insulating phase.

Granular metals show a metal-insulator transition with features which make these materials particularly useful for the study of questions of localization and electron-electron interaction which are of current interest. Granular aluminum has the added advantage that its superconducting properties provide a sensitive probe of electron behavior.

We have measured the electrical resistance of a series of granular aluminum specimens on both sides of the metal-insulator transition over a wide span of temperatures (from 0.3 to 300 K) and magnetic fields (from 0 to 9 T). The results allow us to distinguish between four regimes as a function of the room-temperature resistivity, ρ_{RT}:

(i) The conduction properties of specimens with ρ_{RT} up to about 10^{-3} Ω cm are similar to those of homogeneous impure metals. The normal-state resistance R_N changes very little with temperature.

(ii) For somewhat higher values of ρ_{RT} (up to about 5×10^{-3} Ω cm) the temperature coefficient of resistance is negative. Below 5 K the resistance approaches a finite value at $T=0$, with $R_N(T) = [R_N(0)](1-A\sqrt{T})$. (See Fig. 1.) The specimens are superconducting with a transition temperature T_c which is approximately independent of ρ_{RT}.[1]

(iii) For high-resistivity specimens ($\rho_{RT} \gtrsim 5 \times 10^{-2}$ Ω cm) the resistance varies as $R = R_0 \exp(T_0/T)^{1/2}$ over a wide temperature range. (See Fig. 2.) These specimens show no signs of superconductivity.

(iv) Between regimes (ii) and (iii) is an intermediate regime in which the change of R_N with T is slower than in (iii), but at least linear in $\log T$. (See Fig. 3.) The specimens are superconducting, but with significantly reduced values of T_c.[2]

The large temperature span of our present measurements leads to two further conclusions which we would like to emphasize:

(v) The high-temperature behavior of the specimens in the metallic regime (ii) and in the intermediate regime (iv) is similar. It is only at low temperatures that the variation of the resistance makes it possible to differentiate between them. (See Fig. 3.)

(vi) The Mott variable-range hopping law $R = R_0 \times \exp(T_0/T)^{1/4}$ is not observed in any specimen over the whole temperature range, and, in particular, never at the lowest temperatures. (See Fig. 4.)

Paragraphs (ii), (iii), and (vi) lend support to theories which emphasize the importance of correlation effects in disordered materials.[3,13] The distinctive feature of granular metals (as compared to doped semiconductors) which allows the correlation effects to be observable over a broad temperature range is that they can have a low conductivity as well as a

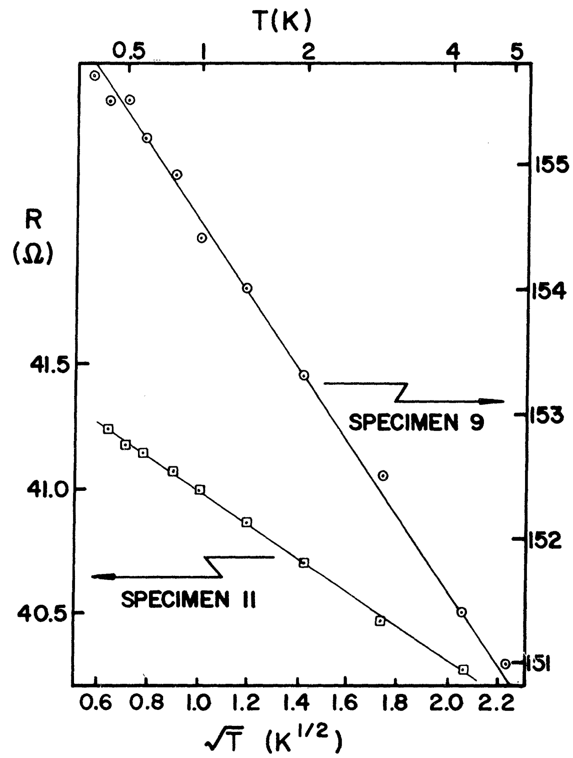

FIG. 1. Normal-state resistance as a function of $\sqrt{T}$ for the metallic specimens, 9 and 11.

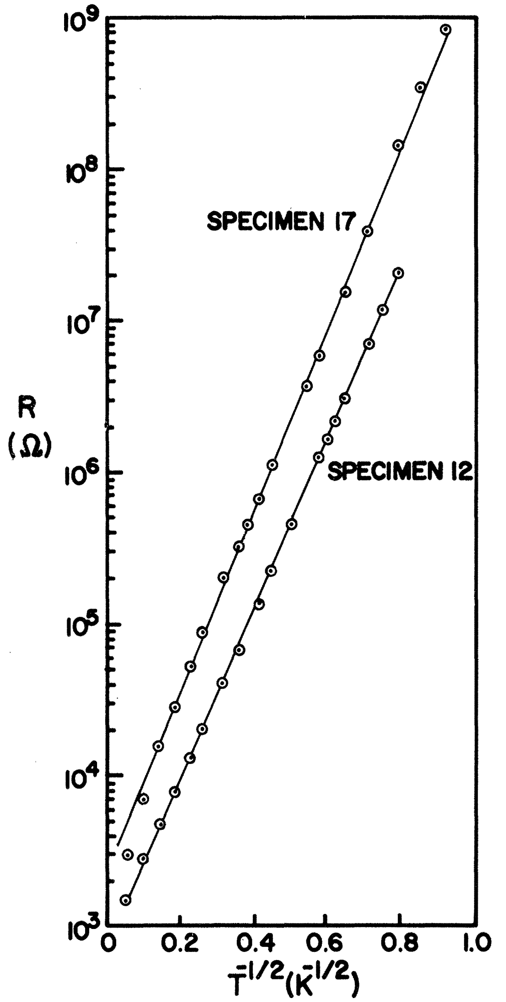

FIG. 2. Log$R$ as a function of $T^{-1/2}$ for the insulating, nonsuperconducting specimens, 12 and 17. (The measured resistance of specimen 17 has been multiplied by a factor of 9.3. This normalizes the graphs to the same geometrical factor; i.e., it makes the ratio of the plotted resistances for the two specimens equal to the ratio of their resistivities.)

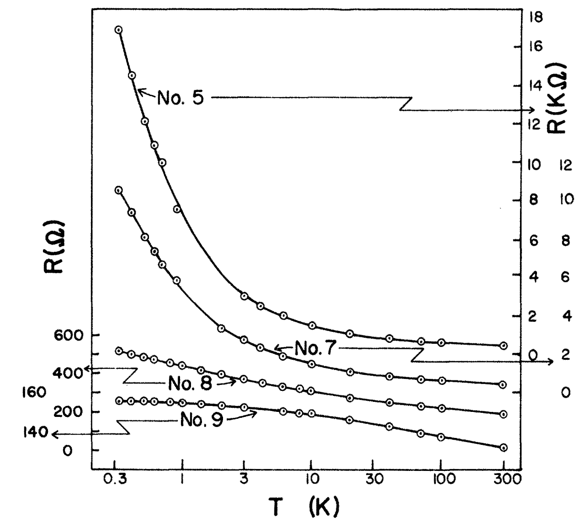

FIG. 3. Normal-state resistance as a function of log$T$ for the metallic specimen, 9, and specimens 8, 7, and 5, which are in the intermediate regime.

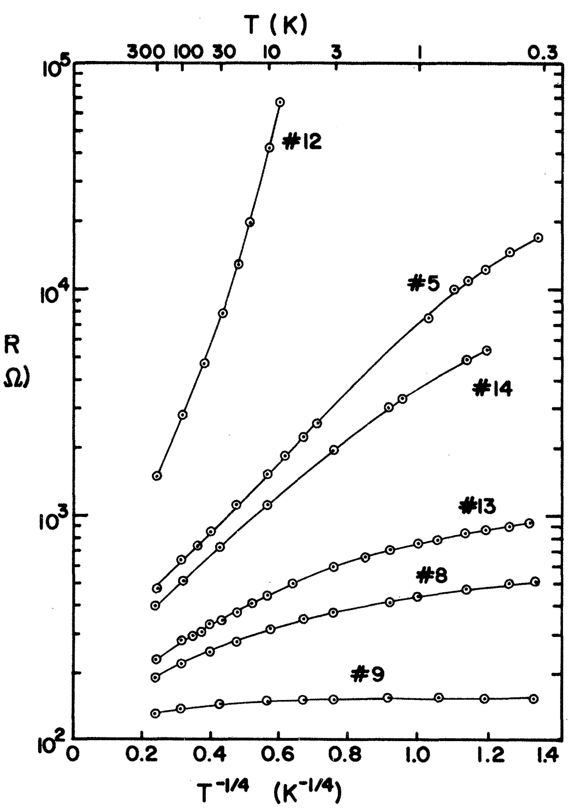

FIG. 4. Log$R$ as a function of $T^{-1/4}$ for six specimens. Specimen 9 is in the metallic regime, specimen 12 is in the insulating, strong-localization regime, and the others are in the intermediate, weak-localization regime.

high density of states. The implication is, however, that at sufficiently low temperatures all disordered semiconductors will exhibit correlation effects, so that the limiting behavior as T goes to zero will never be that of the Mott hopping law.[4]

The characteristics of our specimens are given in Table I. They were made by evaporating pure aluminum in the presence of a small amount of oxygen. The aluminum then forms metallic grains surrounded by amorphous aluminum oxide. By varying the oxygen pressure the resistivity can be changed over a wide range, from the metallic to the insulating regimes. We report here on specimens with the values of ρ_{RT} from 1.5×10^{-3} to 1.3×10^{-1} Ω cm. In this region the grain size is known to be rather uniform,

TABLE I. Characteristics of the specimens. ρ_{RT} is the resistivity at room temperature, $\rho_{4.2}$ the resistivity at 4.2 K, and T_c is the temperature at which the resistance has dropped to half of its maximum value.

Specimen number	ρ_{RT} (10^{-3} Ω cm)	$\rho_{4.2}$ (10^{-3} Ω cm)	T_c (K)
11	1.5	1.7	2.34
9	5.5	6.3	2.31
8	8.3	15	2.03
13	9.6	22	1.81
14	16	69	1.47
7	20	93	1.41
5	20	98	1.17
12	63	15 000	0
17	130	96 000	0

with grain diameters near 30 Å.[1] Since the thickness of the films (about 1 μm) is larger than any length relevant to the conduction process, we expect the specimens to behave like three-dimensional systems.

All but two of the specimens become superconducting and exhibit superconducting fluctuations well above the transition temperature. The low-temperature measurements were therefore made in magnetic fields up to 9 T and the normal-state resistance was determined by extrapolation to zero field.

We begin with the results for our most resistive specimens, 12 and 17. Figure 2 shows that over a resistance variation of four decades the resistance follows the relation $R = R_0 \exp(T_0/T)^{1/2}$. This form of variation was also found earlier, between 1.2 and 4.2 K, in two insulating, nonsuperconducting specimens.[2] Sheng *et al.*[5] have derived such a formula for granular metals by considering the effect of the charging energy $E_c = E_c(d,s)$ on the conductance of paths linking grains of separation s and diameter d. They make the two special assumptions that s is proportional to d, and that there is a broad grain-size distribution, so that conduction is dominated by paths with an optimal value of s and therefore also d. Our results, together with the measured grain-size distribution of similar specimens, are incompatible with the second of these assumptions.[6]

An $\exp(T_0/T)^{1/2}$ dependence arising from the Coulomb energy has also been derived for hopping conduction by Efros and Shklovskii.[3] Efros[7] showed that their expression should be valid for $kT \ll e^6 N(0)/kT_0$. For the values of T_0 from Fig. 2 of 170 K for specimen 12 and 200 K for 17, with the density of states, $N(0)$, for bulk aluminum this criterion leads to $T \ll 10^4$ K. This result suggests that the $\exp(T^{-1/2})$ temperature dependence is not observed in impurity semiconductors because $N(0)$ is usually lower by several orders of magnitude. Support for this view comes from the experiments of Redfield[8] on very heavily doped gallium arsenide, with $N(0) \sim 1.5 \times 10^{31}$ $(\mathrm{erg\,cm^3})^{-1}$. The application of Efros's formula shows that the $\exp(T^{-1/2})$ dependence is expected in Redfield's specimens below about 20 K, and that is indeed what is observed.

The validity of the theory of Efros and Shklovskii[3] must, however, remain in doubt until questions about their derivation are resolved.[9] In order to apply it to our system it will also be necessary to show that it is applicable in granular materials.

We now turn to the other specimens. Figure 3 shows that at high temperatures R varies slowly, with conductivities σ given by $d\sigma/d\log_{10}T = 20 \pm 7$ $(\Omega\,\mathrm{cm})^{-1}$. At low temperatures the specimens behave very differently. The resistance of specimen 9 tends to a finite value (as is true also for specimen 11) while the resistances of the others continue to increase down to the lowest temperatures of the measurments although less rapidly than the $\exp(T_0/T)^{1/2}$ behavior of specimens 12 and 17.

The similar behavior at high temperatures at the same time as the very different behavior at low temperatures is consistent with the implications of the scaling theory of localization[10] for three-dimensional systems discussed by Imry.[11] According to his analysis the temperature dependence of the conductivity depends on the relative importance of two lengths. One is a temperature-independent length ξ which shows how close a system is to the metal-insulator transition, and which diverges at the transition. The other is the mean electron-diffusion distance between inelastic scatterings, l_{in}, which depends on the temperature and on the disorder. At high temperatures, when $l_{in} \ll \xi$, it is not possible to tell whether a specimen is on the metallic or the insulating side of the transition.

Specimens 9 and 11 are in the extended-state regime, but in both of them the temperature dependence of the resistance exhibits the incipient effects of localization. Figure 1 shows that for these two specimens, between 0.3 and 5 K, $\sigma = \sigma_0(1 + A\sqrt{T})$, with $A = 0.018 \pm 0.001$ $\mathrm{K}^{-1/2}$. We can compare the $\sqrt{T}$ variation with the interpolation formula $\sigma \propto (1/\xi) + (1/l_{in})$ which has been suggested[12] for the region where both lengths influence the conductivity. The two are consistent if l_{in} is proportional to $T^{-1/2}$.

The electron-electron interaction theory of Altshuler and Aronov[13,14] also leads to a temperature dependence of σ linear in $\sqrt{T}$ in a three-dimensional disordered metal. However, in both theories the values of A are predicted to be in the ratio of $\sigma_0^{-3/2}$, i.e., about 7 for specimens 9 and 11, and not unity as we find.

Altshuler and Aronov[14] have discussed another way in which the $\sqrt{T}$ dependence could arise in a granular metal, namely, through the effect of electron-electron interactions on the resistance of the tunnel junctions between individual grains. The coefficient A for a specimen comprising an array of junctions then depends only on the properties of the metal of the grains and could therefore be the same for our specimens 9 and 11.

We return to the question of whether the specimens in the intermediate regime (iv) are metallic. If they are, the metal-insulator transition and the superconductor-normal transition coincide. The question is equivalent to asking whether the resistances of specimens 5, 7, 14, 13, and 8 approach a finite value as T goes to zero. For these specimens R varies at least linearly with $\log T$ at the lowest temperatures of our measurements. Metallic behavior would require a change to a slower than $\log T$ variation of the resistance at still lower temperatures. Our data show no sign of such a change down to 0.3 K, and to this extent suggest the possibility of superconductivity in the insulating phase.[15] Under these conditions superconductivity could have interesting and unusual characteristics, such as, for example, the reentrant behavior discussed by Simanek[16] and Efetov.[17]

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