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Two-Dimensional Electrical Conductivity in Quench-Condensed Metal Films

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The resistance of ultrathin films condensed on low-temperature substrates has been measured. All of the materials studied (Au, Cu, Al, Sn, and Pb) show a transition from activated to metallic behavior as the resistance is reduced below 30 000 Ω/□. This result gives strong evidence in support of the concept of a minimum two-dimensional conductivity. In Pb films with resistance > 30 000 Ω/□, the onset of superconductivity is marked by an increase in activation energy.

There is growing evidence[1,2] that as the resistance of a two-dimensional electron system is increased, for example by reducing the electron density in inversion layers[3] or by changing topology in thin metal films,[4] metallic conduction cannot persist above a resistance of $\sim 0.12 e^2/\hbar = 30\,000\ \Omega/\square$. This limit occurs at the Ioffe-Regel[5] condition, when the electronic mean free path is equal to the Fermi wavelength. Above this limit conduction becomes activated. It is clear that such a high resistance cannot be reached in a uniform metal film, where the Fermi wavelength is comparable to the lattice spacing, as even a poor conductivity of approximately $10^4\ (\Omega\ \mathrm{cm})^{-1}$ would require a film thickness of less than an interatomic spacing. Hence when films reach the resistance of 30 000 Ω/□, they must be nonuniform, consisting of islands connected by narrow constrictions or by tunnel barriers. There has been considerable discussion concerning the energy barriers determining this activated transport. The splitting of energy levels within each island or constriction due to the size effect may give rise to the activation energy,[1,2] or the charging energy of such small particles may be dominant.[6]

In this Letter we present a study of films made by evaporation of Pb, Sn, Au, Al, and Cu onto a substrate held near 4.2 K. Such quench-condensed films show conductivity at a lower average thickness than films of the same materials made at higher temperatures. Further, *in situ* measurement of the resistance versus temperature has allowed us to increase the thickness of the films progressively until activated conductivity is no longer observed. This transition from activated to metallic behavior occurs near a resistance of 30 000 Ω/□. We further observe that in Sn films the resistance varies as $\exp(T^{-1})$, whereas in Au and Cu it varies as $\exp(T^{-1/2})$. In the latter cases the extrapolated high-temperature resistance is close to 30 000 Ω/□. In Pb films we observe tunneling between superconducting islands in the activated regime and supercurrents due to Joseph-

son coupling between islands once the film is metallic.

The films are prepared and measured in a small evaporator which is completely immersed in liquid helium.[7] This consists of a 11.5-cm-long stainless steel nipple of 3.8 cm i.d. with the ends closed by Varian Conflat flanges. The lower flange provides the filaments for evaporation; the upper flange supports a sample block with substrate, thermometer, and heater and a quartz crystal for film-thickness measurement. A radiation shield, soldered to the copper gasket of the upper flange, holds a mask in front of the substrate. This substrate, of fire-polished glass, has two thin (a few hundred angstroms) gold contacts evaporated across its width. The chamber is evacuated at room temperature and then cooled. Although we have no measure of the pressure in this system when it is immersed in helium, it should be extremely low because of cryopumping on all surfaces. Initially a film is evaporated until some conduction is measured between the gold contacts. (During evaporation the substrate warms to between 5 and 10 K.) The resistance of this film is then measured from 4.2 to 10 K using a Keithley 616 digital electrometer. The field along the film is <1 V/cm and the current is proportional to voltage within the range of fields used. A large temperature range cannot be covered since above about 15 K annealing occurs in most of the films [the $R(T)$ curves are not reversible]. Next, more metal is evaporated and the sequence of evaporation and measurement is followed until the film resistance is about 1 kΩ. The average film thickness at each step is measured with the quartz crystal which has been previously calibrated with a much thicker film and an optical interference device.

The resistance of our films at 4.2 K versus thickness is shown in Fig. 1. Once the films begin to conduct (defined as the limit of our measurements, $\approx 10^{11}$ Ω/□) the decrease of resistance with thickness is very rapid and the dependence appears to be exponential. In Fig. 1, the resistance changes by 10^6 for an increase in average thickness of only ≈5 Å (Cu) or ≈6 Å (Au). This is in rough agreement with a model in which we assume the film to be made up of islands separated by a distance, s, which is reduced during each evaporation of new film. If the tunneling barrier between islands is vacuum with height ≈4 V, then the tunneling probability[8] increases by 10^6 if s decreases by 7 Å, which is comparable to the increase in film thickness. If the tunneling occurs through the glass instead, a barrier height of 2 V gives a change of $\sim 10^5$ for a decrease of 7 Å.

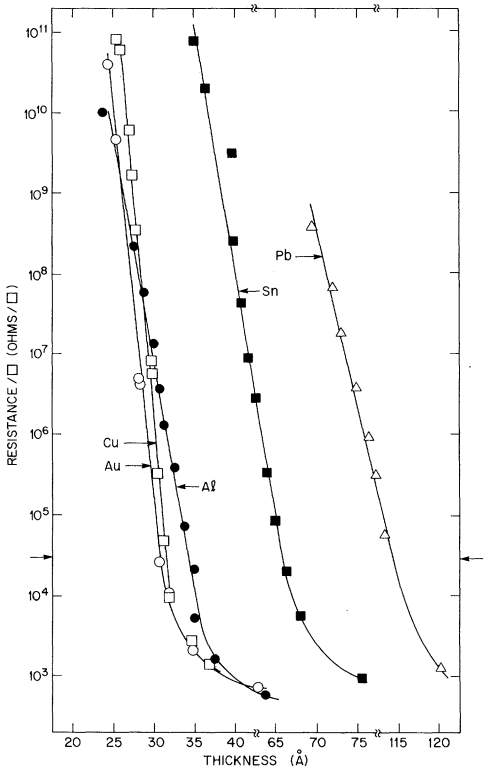

FIG. 1. Resistance vs average film thickness for the materials studied. All resistance measurements were taken at 4.2 K except for the case of Pb where the measurements were taken at 7.5 K.

The temperature dependence of resistance of the Cu and Au films is shown in Fig. 2. The fit in this case is noticeably better to $\exp(T^{-1/2})$ than to $\exp(T^{-1})$, but other fractional powers of T cannot be ruled out. Near 30 000 Ω/□ the activated behavior becomes weak, but persists somewhat below this value. These very disordered quenched films do not develop a resistance that increases with *increasing* temperature even when very thick; hence "metallic" conduction is taken as a resistance independent of temperature. Below 30 000 Ω/□, the conduction apparently becomes "metallic."

A most interesting feature of Fig. 2, which is not understood, is that extrapolations of the plots to high temperatures appear to converge within a factor 10 of the value of 30 000 Ω/□, but fall below this value. Thus the pre-exponential term in the activated behavior appears to be close to this limiting metallic resistance. The $1/T^{1/2}$ de-

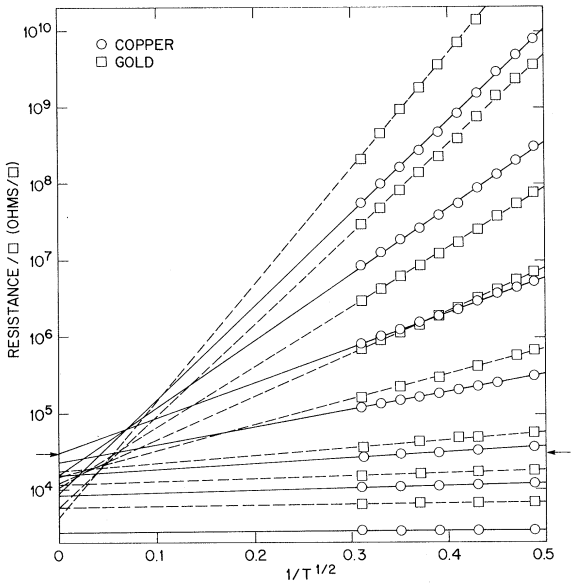

FIG. 2. Resistance vs $1/T^{1/2}$ for copper and gold. High-temperature extrapolations for the high-resistance curves all tend to converge to ~ 30 000 Ω/□.

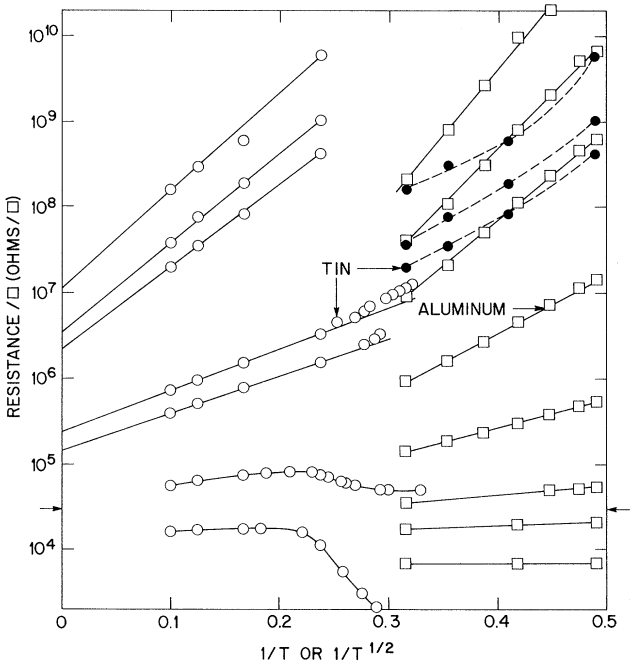

FIG. 3. Resistance vs $1/T$ for tin (open circles) and $1/T^{1/2}$ for aluminum (squares) and tin (closed circles). Notice the effect of superconductivity of Sn at $1/T = 0.22\ K^{-1}$.

pendence itself is also of some interest, in that it has been explained[6] by assuming a relationship between the diameter (d) of the islands (or particles in the case of cermets) and their separations. It follows that if $s/d = \text{const}$, a $1/T^{1/2}$ dependence of log$R$ results. While this relationship follows from the argument that the average density of the film or cermet must be constant, it is surprising that this dependence is widely observed, in that one might expect the current to flow in regions that do not exhibit average behavior—for example, between large particles which, by chance, occur closer together than expected.

In the case of Al films (Fig. 3) it is difficult to decide on the correct temperature dependence although log$R$ vs $1/T^{1/2}$ seems to be a better fit than $1/T$. Extrapolations of the $1/T^{1/2}$ dependences are again fairly close to 30 000 Ω/□. However, Sn films above 4.2 K definitely fit $1/T$ much more closely than $1/T^{1/2}$, and in this case there is no common extrapolation of the plots for each thickness. From Fig. 1 it can be seen that the Sn film is more than twice as thick (at a given resistance) as the Au, Cu, or Al films. Again, in both the Al and Sn, below ~30 000 Ω/□ the films apparently become "metallic" in that the temperature dependence of the resistance disappears.

Below 4.2 K, superconducting effects are observed in the Sn film and these are much more striking in the case of Pb films, as shown in Fig. 4. For the thickest Pb film the transition to the superconducting state is complete and the midpoint of the transition is at 6.6 K. However for the film with resistance close to 30 000 Ω/□ the transition is broadened, especially below its midpoint, while in the films with resistance near 10^5 Ω/□ the resistance first decreases at T_c and then increases at lower temperatures. For higher-resistance films the activation energy increases below T_c, i.e., the films become *more* resistive when superconducting. This behavior can be understood in a qualitative way by analogy with well-known superconducting tunneling experiments. The thinnest films are over 100 Å thick (see Fig. 1) and hence, under the assumption that the film is comprised of individual islands, these are still superconducting with a T_c of 6.6 K since size effects are not serious within grains of this size. At T_c, opening of the energy gap makes it more difficult to tunnel between islands, as in a superconductor-insulator-superconductor tunnel junction at voltages below the gap. Alternatively, one thinks of the energy gap as an additional mismatch between energy levels of adjacent islands, applying whether this mismatch is due to a charging energy or to size quantization. In the lower-

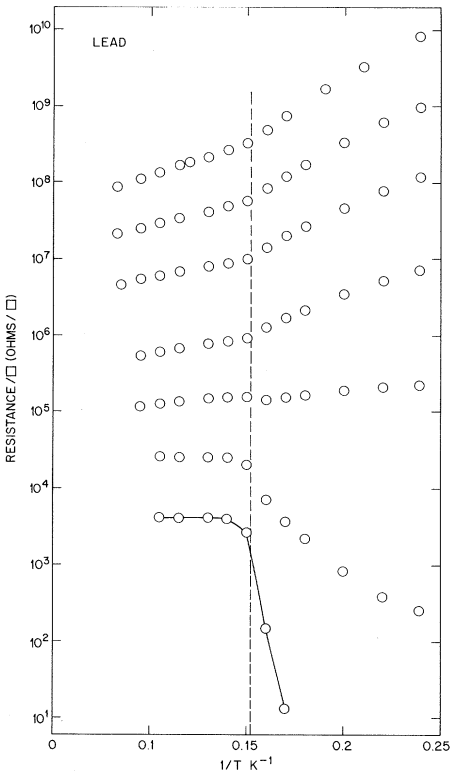

FIG. 4. Resistance vs $1/T$ for lead. For resistances greater than 30 000 Ω/□, the temperature dependence is activated with an increase at T_c. Below 30 000 Ω/□ "metallic" behavior is observed with superconductivity below T_c.

resistance films Josephson coupling between islands becomes strong enough for supercurrents to flow through the film as a whole. For granular superconductors, Abeles[9] has pointed out that this will not happen unless the grains are in intimate electrical contact, which suggests that the Pb film becomes "metallic" because the islands begin to touch. Certainly from Fig. 4 it is clear that in the range of values where the resistance ceases to be activated (10^4–10^5 Ω/□), the grains couple together sufficiently strongly to result in a superconducting film. This observation further supports the concept of a minimum metallic conductivity.

In passing, we comment that various models have been proposed for conduction in a localized or activated regime[10] and recently much thought has been directed toward understanding metallic conductivity[11] in the strong-scattering regime (electronic mean free path $\approx a_0$, the lattice spacing). From Figs. 2–4 it is clear that experimentally the transition from this metallic behavior to activated is a continuous one and models describing conduction in either limit should, conceptually at least, be equipped to describe this transition.

In summary, we have performed resistance measurements on ultrathin films grown on low-temperature substrates. The resistance versus film thickness suggests that these films consist of metallic islands and that, for thin films, the conduction is via tunneling. This property makes this an ideal system to study conduction in two dimensions. It is observed that the temperature dependence of the resistance follows an $\exp(1/T^{1/2})$ dependence for Au and Cu, and tends toward $\exp(1/T)$ in the case of Sn. Al lies somewhere between these two dependences. Although this temperature behavior is not understood, it is characteristic of all the materials studied that for a resistance in the vicinity of 30 000 Ω/□, the conduction changes from activated to metallic. In the case of the superconducting materials, the film displays superconductivity at and below this critical value of resistance. These observations provide strong support for the concept of minimum metallic conductivity in a two-dimensional system.

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