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Two-Dimensional Localization in Thin Copper Films

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Resistance measurements have been made on low-resistivity thin Cu films (50–500 Å) between 1 and 20 K. A logarithmic temperature dependence of the resistance is observed. For a resistance per square of $R < 20\ \Omega/\square$, good agreement is obtained with the localization theory. The amplitude of the resistance variation and its large magnetic-field dependence exclude possible Coulomb-interaction effects predicted by Altshuler *et al.* as the main source of the observed behavior.

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Based on the scaling ideas of Thouless,[1] Abrahams *et al.*[2] recently predicted that in a thin metallic film, electronic states should become localized at low temperatures, resulting in a logarithmic increase of the resistance as the temperature is lowered.[3] Experimental studies of this effect have been reported in ultrathin (and presumably granular) Au-Pd films[4] and in inversion layers.[5] The resistance per square $R_\square$ of these samples was typically 1000 $\Omega/\square$ or more. Although the results did show the predicted $\ln T$ dependence, the magnitude of the effect was only a half to a quarter of that predicted by the theory. Similar problems have arisen in the study of one-dimensional (1D) localization in highly disordered[6] and amorphous[7] systems and have been attributed by Thouless[8] to a linear temperature dependence of the electron-phonon scattering time characteristic of highly disordered two-level systems.[9] An alternative interpretation of these results in terms of the Coulomb interaction has been proposed by Altshuler, Aronov, and Lee.[10]

We report for the first time on measurements of 2D localization in clean metallic (copper) films, with a mean free path of the order of the film thickness and a resistance per square of the order of 10 $\Omega/\square$. The resistance of these films shows the expected logarithmic increase at low temperatures. The strength of the effect is in agreement with the localization theory but disagrees strongly with the theory of Altshuler *et al.*, in view of the Hartree correction which becomes very important for clean films. Slightly thinner films, approaching the discontinuity limit, show a weaker effect, similar to what was observed by Dolan and Osheroff.[4] However, in this limit it is quite difficult to distinguish between the predictions of the localization and the Coulomb theory.

Before discussing the implication of these results for further studies of localization in one and two dimensions, we first describe the experimental conditions.

The Cu samples were prepared in a commercial stainless-steel, diffusion-pumped chamber, at a pressure of 10^{-6} Torr. The clean metal films were made by evaporating MRC 99.999% pure copper from a resistively heated molybdenum boat onto a Corning 7059 glass substrate held at room temperature. The evaporation rate was about 10 Å/s. By means of a sliding mask system a wide range of Cu thicknesses could be evaporated on one substrate and within a single evaporation run. The film thickness was measured with a quartz-crystal thickness monitor which has been calibrated with an optical interference technique.

Using photolithographic techniques we obtained on each substrate a set of Cu strips with different thicknesses, each one allowing four-terminal measurements of the film resistance. The conducting channel was 0.235 mm wide and the potential probes were separated by 4 mm. The applied photoresist layer also served as a protection for the thin Cu films.

The resistance versus temperature measurements were performed between 20 and 1 K in a temperature stabilized ^{4}He cryostat. Because of the very small resistivity changes which are of the order of 10^{-9} Ω cm, precautions had to be taken to minimize external noise inputs and to increase the sensitivity of the measuring system.

With use of a battery-powered current supply, two Solartron 7075 digital voltmeters (sensitivity 10^{-7} V) and a Minc computer (Digital) for averaging, resistance changes of $\Delta R/R \sim 10^{-6}$ could be detected at low currents. Within this sensitivity the measured values at a fixed temperature were stable over several hours. This high sensitivity was necessary for an accurate measurement of resistance changes in the thicker films (see Fig. 1). In the following we discuss only data taken at low electric fields, where the resistance is field independent. At higher electric fields, the resistance was found to decrease in a way similar to that reported by other authors (using the notation of Ref. 5, we found $p' = 2.4$ at $R_\square \sim 100$ Ω/□).

The localization theory of Abrahams *et al.* predicts that

$$\frac{\Delta R_\square}{R_\square^2} = \frac{\alpha P}{2} \frac{e^2}{\pi^2 \hbar} \ln 10. \qquad (1)$$

Here $\Delta R_\square$ is the increase of $R_\square$ over one decade

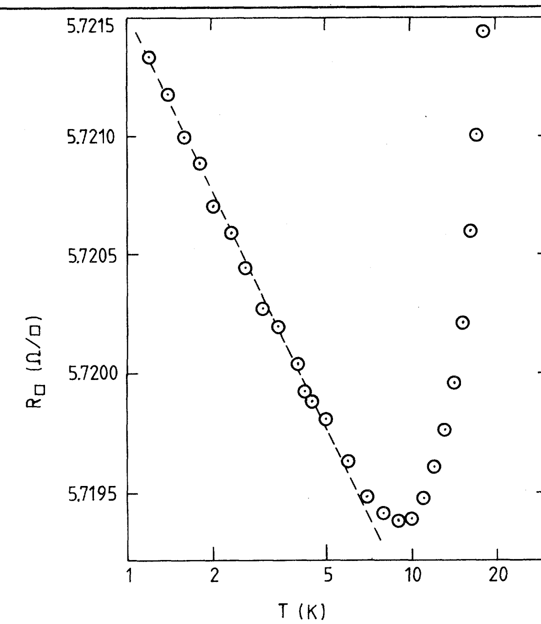

FIG. 1. Resistance vs temperature for a Cu film with thickness $d = 119$ Å and resistivity $\rho_{4.2\,K} = 6.8 \times 10^{-6}$ Ω cm.

of temperature. The parameter αP, with P the power of T for the appropriate inelastic scattering mechanism and $\alpha = 1$, is expected to be equal to 3 for 3D electron-phonon scattering and to 2 in the 2D case in the clean limit.[1] For electron-electron scattering a value of 2 is predicted. In the investigated temperature range, our Cu films with $R < 20$ Ω/□ are in the clean limit. Therefore, one should expect $\Delta R/R^2$ values between 5.73×10^{-5} Ω^{-1} ($\alpha P = 2$) and 8.59×10^{-5} Ω^{-1} ($\alpha P = 3$).

The theory of Altshuler, Aronov, and Lee[10] makes a prediction similar to Eq. (1), with $\alpha P = 1$. There is, however, a Hartree correction to this exchange term. It has the same form as the exchange term [Eq. (1)] but is reduced by a factor $(1 - F)$. The screening length k^{-1} in Cu is much shorter than the film's thickness. Hence a calculation of F based on 3D screening applies,[11] $F = (1/x) \ln(1 + x)$ with $x = [2(k_F/k)]^2$, from which we estimate, for Cu, $F = 0.6$. The effective value of αP from this theory is then $\alpha P = 0.4$, quite different from the values compatible with the localization theory $\alpha P \gtrsim 2$.

For all films in this resistance range, we find a logarithmic increase of the resistance at low temperature that can be described by Eq. (1) with $\alpha P \gtrsim 2$ (Fig. 2). The $R_\square$ value of these films varies as $1/d^2$ for $R_\square \lesssim 15$ Ω/□ (larger thicknesses), indicating that the electron mean free path

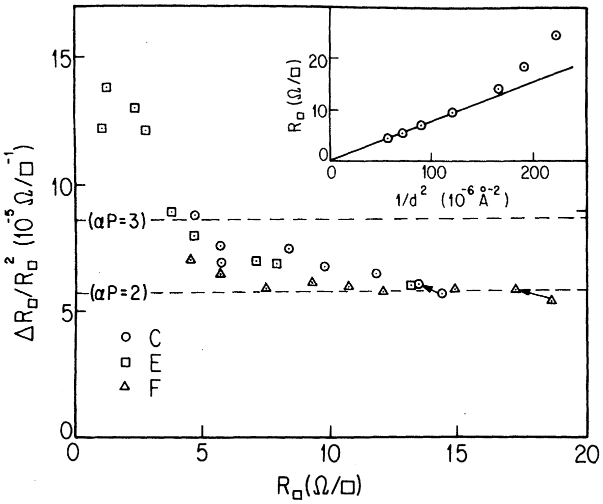

FIG. 2. Variation of $\Delta R_\square/R_\square^2$ as a function of $R_\square$ for three different series of films.

TABLE I. Sample characteristics for one of the series shown in Fig. 1.

d (Å)	$R_\square$ ($\Omega/\square$)	$10^5\ \Delta R_\square/R_\square^2$ [$(\Omega/\square)^{-1}$]
130	4.53	7.10
110	5.7	6.54
94	7.52	5.95
86	9.36	6.19
78	10.68	5.97
72	12.1	5.80
67	14.84	5.89
62	18.73	5.46
	17.27[a]	5.92[a]

[a]Corrected (see Ref. 12).

is thickness limited[12] (see inset of Fig. 2). Table I gives the samples characteristics for one of the series shown in Fig. 2.

The data for $5\ \Omega/\square \lesssim R_\square \lesssim 20\ \Omega/\square$ is fully consistent with $\alpha P=2$. For $R_\square \lesssim 5\ \Omega/\square$, $\Delta R_\square/R_\square^2$ shows a systematic increase above the value corresponding to $\alpha P=2$. In this connection, we note that the characteristic phonon wavelength in Cu at $T\simeq 4$ K is of the order of 100 Å. This is the thickness at which $R_\square \sim 5\ \Omega/\square$ (see Table I). Hence, the observed increase could be due to a crossover between a 2D regime at $R_\square > 5\ \Omega/\square$ ($\alpha P=2$) and a 3D regime at $R_\square < 5\ \Omega/\square$ ($\alpha P=3$) (if we assume that the electron phonon scattering dominates over the electron-electron scattering).

The larger value of $\Delta R_\square/R_\square^2$ at low $R_\square$ (thick films) could also be attributed to contamination of the films by a small amount of magnetic impurities (< 2 ppm). If we assume that their concentration is the same in all films prepared simultaneously, the Kondo contribution to $\Delta R_\square/R_\square^2$ should vary as $R_\square^{-3/2}$ [since $R_\square \sim 1/d^2$ and $\Delta\rho^{\text{Kondo}} = (\Delta R_\square)\,d = \text{const}$]. Even if we attribute the totality of the resistance increase at $R_\square \sim 1\ \Omega/\square$ to a Kondo effect, the resulting $\Delta R_\square/R_\square^2$ at $R_\square \sim 10\ \Omega/\square$ [$\sim 0.5\times10^{-5}\ (\Omega/\square)^{-1}$] would still be a small correction. Also it would seem to be an unlikely coincidence if an (uncontrolled) amount of magnetic impurities would precisely give $\Delta R_\square/R_\square^2 \sim 6\times10^{-5}\ (\Omega/\square)^{-1}$ in so many samples.

Our results at $R_\square \lesssim 20\ \Omega/\square$ fully meet two important requirements for a meaningful comparison with the localization theory. First, their low resistance per square ensures that the condition necessary to ensure the validity of the logarithmic correction to the conductivity [Eq. (15) of Ref. 2] is fully met. Second, since the mean free path is much larger than the interatomic distance, disorder effects should be unimportant and the electron-phonon scattering time should have its bulk value. We therefore consider that the agreement between our data and the prediction of Abrahams *et al.*[2] constitutes an experimental verification of their theory. At the same time it appears that the theory of Altshuler *et al.* is not capable of explaining the behavior of our clean copper films.

Moreover, preliminary measurements of $R_\square$ as a function of a magnetic field applied perpendicular to the films shows an important negative magnetoresistance even at magnetic fields as low as 100 G. Following recent calculations by Altshuler *et al.*,[11] this effect is consistent with localization and should be absent if the logarithmic increase of R with T was due to electron correlation effects. A detailed report on magnetic field effects will be published elsewhere.[13]

For Cu films close to the continuity limit ($R_\square \gtrsim 100\ \Omega/\square$), we obtain significantly smaller values of $\Delta R_\square/R_\square^2$, corresponding to an effective $\alpha P \lesssim 1$, as first reported by Dolan and Osheroff[4] for ultrathin AuPd films. This lower value αP may be due to a number of reasons: appearance on the substrate of macroscopic uncovered regions on a scale larger than $L_T(T) = (v_F \tau_{\text{ind}} l)^{1/2}$ (which would enhance the measured value of $R_\square$); disorder effects on $\tau_{e\text{ph}}$[8]; and predominance of the Coulomb interaction at large $R_\square$.

In conclusion we wish to suggest that further studies of localization can and should be conducted on clean metal films. This is particularly true for the study of localization in one dimension, for which the T^{-2} dependence of the resistance predicted by Thouless remains to be observed. Copper wires, with thicknesses of 100 Å, widths of 1 μm, and lengths of 1 cm would seem to be well suited for that purpose.

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[12]Deviation from $R_{\square} \propto 1/d^2$ at small thickness indicates that films tend to become discontinuous with the apparition of macroscopic cracks and holes. The measured value of $R_{\square}$ is then enhanced above that of the material itself, which we expect should still vary as $1/d^2$. For deviations from linearity less than 15%, we have corrected the measured value of $R_{\square}$ as indicated by the arrows in Fig. 2.
[13]L. Van den Dries, C. Van Haesendonck, Y. Bruynseraede, and G. Deutscher, to be published.

Conductivity Cusp in a Disordered Metal

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A tendency toward a cusp at zero temperature in the electrical conductivity of Si crystals doped with P is observed. It is found that, within the metallic state, decreasing P concentration enhances the cusp and then rapidly changes its sign as a pseudogap opens. Such a cusp has been predicted for a disordered metal in which Coulomb interactions dominate the scattering.

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The physics of disordered Fermi systems involves both the effects of localization and of Coulomb interactions, but their relative importance and interplay remain unresolved. Fundamentally different theories of localization[1] and of electronic interactions[2,3] both agree with recent observations of a logarithmic dependence of the resistivity upon temperature and electric field in two-dimensional (2D) metals[4] and with the behavior of thin wires.[5] Hall-effect experiments[6] on 2D electron inversion layers support the Coulomb-interaction approach. In contrast, neither theory is supported by whisker and colloidal particle studies.[7] Our results on dc conductivity and far-infrared transmission of 3D metallic samples of Si:P appear to be explicable within the Coulomb-interaction picture.

We have made four-probe resistance measurements of metallic, uncompensated samples of Si:P as a function of T at frequencies ~10 Hz. Wires of Au:Sn were spot welded to a freshly etched sample surface to make low-resistance,

