Localization and negative magnetoresistance in thin copper films

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The electrical properties of thin Cu films with resistance per square in the range 1 to 100 Ω/□ have been studied. For temperatures below 10 K, the resistance of the films increases logarithmically with decreasing temperature and the magnitude of the rise becomes larger as the resistance per square is increased. The results are in good agreement with the predictions of Abrahams *et al.* for localization in two dimensions. The effect of a magnetic field has also been investigated. The negative magnetoresistance observed in a perpendicular field is in good agreement with the recent theory of Altshuler *et al.*: At low fields ($H < 10^{-2}$ T), no magnetoresistance is observed; for 10^{-2} T $< H < 1$ T the magnetoresistance changes logarithmically with the magnetic field. For very high fields ($H \gtrsim 5$ T), localization is destroyed, as predicted by theory. In a field parallel to the film plane no negative magnetoresistance is observed. A detailed analysis of the results indicates that spin-orbit effects (of moderate strength) must be taken into account.

I. INTRODUCTION

Recent theories[1,2] of electron localization in quasi-two-dimensional (2D) systems have predicted an anomalous logarithmic increase of the metallic resistance at low temperature. An alternate theory[3] based on electron-electron Coulomb interactions predicts, in two dimensions, a similar logarithmic dependence.

Several experiments indeed indicated anomalous temperature-dependent rises in the resistance for two-dimensional systems at low temperatures.[4–9] We[6] reported resistance measurements on continuous thin Cu films of various thicknesses. The logarithmic increase of the resistance at low temperatures ($T \lesssim 10$ K) was in agreement with the scaling theory of localization[2] for two-dimensional pure metal films with a resistance per square 5 Ω/□ $\lesssim R \lesssim$ 20 Ω/□. Similar measurements in AuPd films[4] ($R > 100$ Ω/□), silicon inversion layers[5] ($R \gtrsim 1$ kΩ/□), ultrathin Pt films[7] ($R > 1$ kΩ/□), oxidized Cu films[8] ($R > 100$ Ω/□), and In oxide films[9] ($R > 100$ Ω/□) also showed a logarithmic decrease of the conductance although smaller than predicted by theory. Moreover, the presence of Coulomb interactions (correlation effects) in these experiments could not be excluded. In good metallic thin Cu films the screening length for the Coulomb interaction is very short and its effect should therefore be small.[6,10] Nevertheless, for a complete theoretical analysis, interaction and localization effects must be taken into account.

More recent theoretical[11,12] and numerical[13] calculations suggest that the most appropriate experiment to distinguish between electron localization and correlation effects is the application of a magnetic field perpendicular to the film plane. In the case of localization one expects a negative magnetoresistance, even at low fields ($H \sim 10^{-2}$ T), and the Hall coefficient should be independent of temperature. Correlation effects, on the other hand, produce no negative magnetoresistance and give a temperature-dependent Hall coefficient.

For Si inversion layers a negative magnetoresistance has been observed at low magnetic fields[14,15] ($H \lesssim 10^{-1}$ T) indicating that localization is present. At higher fields ($H > 1$ T) the negative magnetoresistance disappeared and the change of the Hall coefficient with temperature is in quantitative agreement with the correlation theory.[15,16] Those results indicate that localization as well as correlation effects are present in Si inversion layers. Evidence for magnetic-field-dependent localization was also reported for ultrathin Pt films,[7] oxidized Cu films,[8] and In oxide films.[9]

We present here the results of our study of the localization and magnetoresistance in continuous thin Cu films. These results are important for three reasons:

(1) The data were obtained in "good 2D metals" where $k_F l_{el} >> 1$ (k_F is the Fermi wave vector and l_{el} is the elastic impurity scattering length; typically $k_F l_{el} \simeq 100$). Indeed, since the Cu films have a resistivity of the order of 1 $\mu\Omega$ cm at 4.2 K the conduction electrons can be described as plane waves. This criterion must be fulfilled in order to apply the first-order perturbation theory which gives rise to the logarithmic correction in the metallic conductivity. An indication that higher-order correction terms may be neglected is also provided by the measured relative resistance variation which ranges between 10^{-4} for a Cu-film thickness $d_{Cu} \simeq 200$ Å and 10^{-2} for $d_{Cu} \simeq 50$ Å. It is our belief that a good metallic behavior of the 2D electron gas is the key condition for a meaningful check on weak localization effects in two dimensions.

(2) All our Cu films show electronic localization, and the universality of the effect is not influenced by the preparation conditions or metallic sheet resistance. The effect is already present in films with $R = 1\ \Omega/\Box$ ($d_{Cu} \simeq 200$ Å) and is still effective in films with $R = 1\ k\Omega/\Box$ ($d_{Cu} \simeq 40$ Å), which is the limit of electrical continuity for the Cu films. Moreover, the localization is probably independent of the microscopic origin of the impurity scattering since resistance changes of more than 20% due to aging effects, thermal cycling, or evaporation conditions do not influence the agreement with theory.

(3) Spin-orbit interactions are weaker than in heavier metals,[4,7] and also weaker than in oxidized Cu films.[8] As predicted[12,17] this allows one of the most fundamental signatures of the localization phenomenon to be observed unequivocally: a negative magnetoresistance in perpendicular fields, no magnetoresistance in a parallel field.

The paper is organized as follows. In Sec. II we analyze the resistance-versus-temperature measurements in view of the scaling localization theory. In Sec. III we compare the magnetoresistance measurements with the theory of Altshuler *et al.*[11] (AKLL) and Hikami *et al.*[12] (HLN) for a 2D electron gas. Finally, in Sec. IV we discuss corrections due to electron-electron and spin-orbit interactions.

II. LOCALIZATION IN THIN Cu FILMS

The Cu films are prepared by evaporating MRC 99.999% pure Cu from a resistively heated molybdenum boat onto a Corning 7059 glass substrate held at room temperature (pressure during deposition $\sim 10^{-6}$ torr). Several Cu films with thicknesses varying between 50 and 200 Å could be deposited in one evaporation cycle. The average sample thickness is measured with a quartz-crystal thickness monitor mounted inside the sample holder and calibrated with an optical interference technique. The four-terminal resistance measurements were carried out on Cu strips whose size (4.00×0.235 mm^2) was defined by photolithographic techniques. The applied photoresist layer also served as a protection for the thin Cu films. Using a battery powered current supply and HP 3456 A digital voltmeters, resistance changes $\Delta R/R < 10^{-5}$ could still be detected at low currents. Precaution was taken to keep the applied electric field low enough so that R was field independent (no electron heating). The resistance-versus-temperature and magnetic field measurements were performed in a temperature-stabilized ^{4}He cryostat. A magnetic field up to 4.5 T, supplied by a superconducting magnet, could be applied either perpendicular or parallel to the film plane.

In the scaling theory of localization in zero field at finite temperature T, the resistance per square R depends logarithmically on the inelastic scattering length $l_{in} = v_F \tau_{in}$ where v_F is the Fermi velocity and τ_{in} is the inelastic scattering time (which is proportional to T^{-P}). The increase ΔR of R when the temperature is decreased over one decade is given by[2]

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

where $\alpha = 1$ for the electron gas.

In Fig. 1 we show the measured variation $R - R_{10}$ (R_{10} is the sheet resistance at 10 K) as a function of temperature for seven typical Cu films prepared in different vacuum runs. One clearly sees that the amplitude of the logarithmic variation increases rapidly with increasing R, i.e., decreasing l_{el}. The normalized resistance increase changes from $\Delta R/R^2 = 8.8 \times 10^{-5}\ (\Omega/\Box)^{-1}$ for the thickest film towards $\Delta R/R^2 = 3.8 \times 10^{-5}\ (\Omega/\Box)^{-1}$ for the 45-Å thick film. A compilation of the measured values of $\Delta R/R^2$ is shown in Fig. 2(a). The divergence of $\Delta R/R^2$ for $R \rightarrow 0$ is probably due to a very small amount of magnetic impurities in our Cu films. The Kondo effect caused by these impurities should give rise to $\Delta R/R^2 \propto R^{-3/2}$ when we make the reasonable assumption that the contamination of the Cu films by magnetic impurities

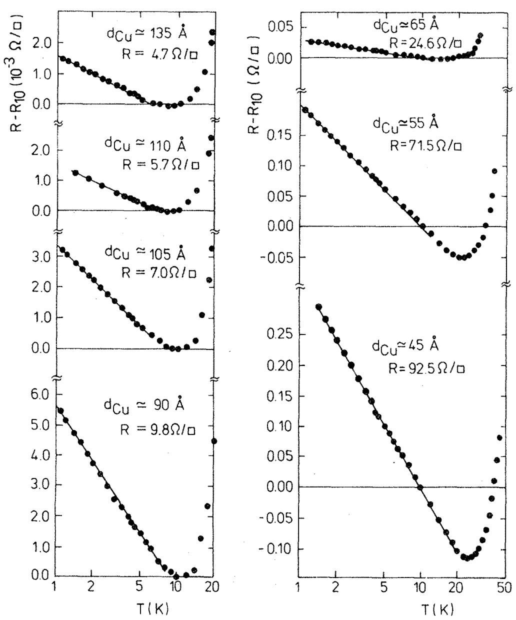

FIG. 1. Resistance R minus the resistance R_{10} at 10 K, as a function of temperature T for different Cu film thicknesses.

is independent of the thickness d_{Cu} (Ref. 6). We have calculated this contribution by assuming that the Kondo effect dominates at $R \lesssim 1\ \Omega/\square$ [full line, Fig. 2(a)]. The corrected data, shown in Fig. 2(b), correspond to $\alpha P \simeq 2$ for $R \lesssim 20\ \Omega/\square$; there seems to be a systematic decrease of $\Delta R/R^2$ at higher resistances. In this connection we note that the mean free path is thickness limited ($R \propto 1/d_{\mathrm{Cu}}^2$) only for $R \lesssim 20\ \Omega/\square$ [see inset, Fig. 2(b)].

The logarithmic increase of the resistance given by Eq. (1) is only valid when $l_{\mathrm{in}}(T) >> l_{\mathrm{el}}$. For an inelastic scattering length $l_{\mathrm{in}}(T) < l_{\mathrm{el}}$ the electron states become completely delocalized. In that case, we recover the usual fast increase of R due to the electron-phonon scattering which rapidly changes above 10 K in Cu. Since in our Cu films we observe $R \propto 1/d_{\mathrm{Cu}}^2$, and assuming that $d_{\mathrm{Cu}} \sim l_{\mathrm{el}}$, we conclude that $R \propto 1/l_{\mathrm{el}}^2$. The temperature T_{min} at which the minimum in the R-vs-T curve occurs is then approximately given by $T_{\mathrm{min}} \simeq bR^{1/2P}$ where b is a constant. Using the results of Fig. 1 we estimate $P \simeq 1.5$, a value which is not unreasonable (either for electron-phonon or electron-electron interaction).

III. MAGNETIC FIELD EFFECTS

The magnetoresistance theory[11,12] predicts that a magnetic field H will decrease the localization ef-

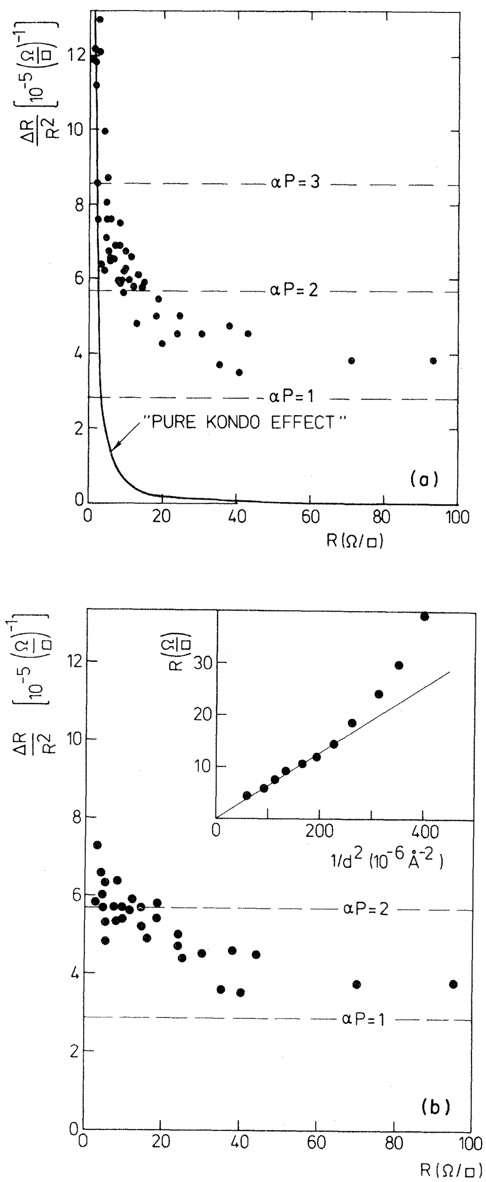

FIG. 2. (a) Normalized increase $\Delta R/R^2$ of the resistance between 10 and 1 K vs resistance per square R. The full line is the theoretical calculation of $\Delta R/R^2$ for the Kondo effect caused by 1 at. ppm Cr in the Cu films. (b) Corrected $\Delta R/R^2$ vs R data taking into account a dominating Kondo effect for $R \lesssim 1\ \Omega/\square$. Deviations from $R \propto 1/d_{\mathrm{Cu}}^2$ are important for $R \gtrsim 20\ \Omega/\square$ (see insert).

fect as soon as the size of the first Landau orbital $l_L = (\hbar/4eH)^{1/2}$ becomes of the same order as the inelastic diffusion length in two dimensions $(\frac{1}{2} l_{\mathrm{el}} l_{\mathrm{in}})^{1/2}$. Above this threshold field the resistance decreases logarithmically with the magnetic field strength. The exact variation

$\delta R_\perp$ [$=R(H=0)-R(H)$] of the resistance in a perpendicular field H is given by

$$\frac{\delta R_\perp}{R^2}=\frac{-e^2}{2\pi^2\hbar}\left[\ln\left[\frac{\hbar/2e}{l_{el}l_{in}H}\right]-\psi\left[\frac{1}{2}+\frac{\hbar/2e}{l_{el}l_{in}H}\right]\right], \tag{2}$$

where ψ is the digamma function. At low fields Eq. (2) reproduces the field-independent logarithmic divergence of R with $l_{in}(T)$ given by Eq. (1). When

$$l_L^2 << \tfrac{1}{2}l_{in}l_{el} \quad (H>>H_c=\hbar/2el_{in}l_{el}),$$

the digamma function reduces to a constant and a $\ln H$ variation is obtained. In a parallel field, the magnetoresistance is predicted to be zero. Equation (2) is no longer valid at very high fields when $l_L<l_{el}$: in that case the electron states become completely delocalized.

We will now show that the observed negative magnetoresistance in our thin Cu films reproduces the main features of the magnetoresistance theory and indicates the presence of 2D localization effects. Figure 3 shows typical magnetoresistance curves in a perpendicular and in a parallel field. In the perpendicular orientation a measurable negative magnetoresistance appears at a field of about 10^{-2} T. At a field of about 10^{-1} T, the variation of $\delta R/R^2$ is of the order of 1×10^{-5} $(\Omega/\Box)^{-1}$. In a parallel field no magnetoresistance is observed, which rules out spin effects as the source of the negative magnetoresistance in the perpendicular orientation. This behavior reproduces the main qualitative features of the localization theory.

In order to compare our experimental results with Eq. (2) we have corrected the data taking into account the normal positive magnetoresistance which occurs at high fields. At $T=77$ K we observe only a positive magnetoresistance (linearly increasing with H). The data shown in Fig. 4 have been corrected by subtracting this positive magnetoresistance from the field dependence of R at low temperatures, using Kohler's rule (actually in our Cu samples the resistance varied only by 5% between 77 and 4.2 K). For magnetic fields $H<2$ T this correction is small ($<10\%$) and a comparison with theory is authorized. Figure 4 shows the negative magnetoresistance in a perpendicular field at 4.2 K for three typical samples with different sheet resistance R. In accordance with Eq. (2), the high-field variation of $\delta R_\perp/R^2$ is logarithmic. The slope of $\delta R_\perp/R^2$ vs $\ln H$ is the same for the three samples. Its value is about 20% smaller than that predicted by Eq. (2); this agreement, although not perfect, is remarkable since it does not involve any adjustable parameter. The value of H_c is an increasing function of R, in qualitative agreement with the expression $H_c=\hbar/2el_{in}l_{el}$. A fit to Eq. (2) over a range of magnetic fields up to a few

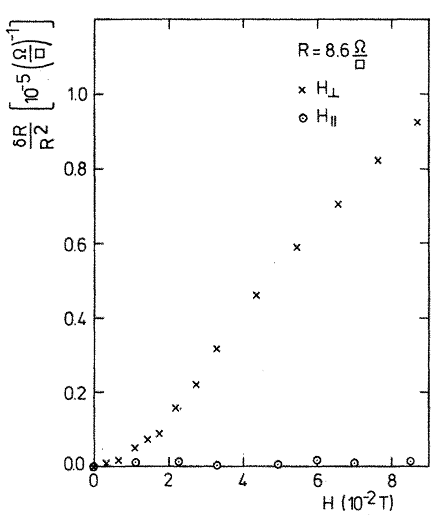

FIG. 3. Low field transverse (×) and parallel (o) magnetoresistance for a Cu film with $R=8.6$ $\Omega/\Box$.

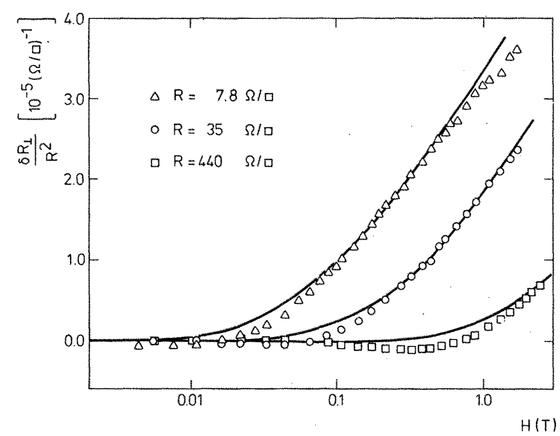

FIG. 4. Normalized negative magnetoresistance $\delta R_\perp/R^2$ as a function of a perpendicular magnetic field H (logarithmic scale) for three Cu films with different values of the resistance per square R. The full lines show the theoretical curves calculated from Eq. (2): $l_{el}l_{in}=3.48\times10^6$ Å^2 for $R=7.8$ $\Omega/\Box$, $l_{el}l_{in}=0.92\times10^6$ Å^2 for $R=35$ $\Omega/\Box$, and $l_{el}l_{in}=0.98\times10^5$ Å^2 for $R=440$ $\Omega/\Box$.

la shows reasonable agreement, although there are some systematic deviations at low fields (in particular in the thinner film for which the magnetoresistance is actually positive at low fields), and at high fields, where, as already noted, the experimental slope is smaller than the theoretical one. Finally, the magnetoresistance shown in Fig. 4 supports qualitatively the notion that localization is completely destroyed for $H > (\hbar/2e)l_{\rm el}^{-2}$. For the Cu film with $R = 7.8\ \Omega/\Box$ we observe already a tendency towards saturation above a field of 1 T. On the other hand, the film with $R = 35\ \Omega/\Box$ still shows the $\ln H$ behavior at 2 T, and the film with $R = 440\ \Omega/\Box$ has not yet reached the $\ln H$ regime.

Figure 5 shows that the temperature dependence of the resistance is quenched at high fields, another major prediction of localization theory.

However, some rather serious difficulties with the interpretation of the data appear when we consider the temperature dependence of the magnetoresistance. As seen in Fig. 6 it is only very weakly temperature dependent, while we would expect the field $H_c \propto \tau_{\rm in}^{-1}(T)$ to vary like T^P. A (probably related) difficulty is the small value of $l_{\rm in}$ required to fit the experimental curves: about 10^5 Å instead of 10^7 Å that we would expect from electron-phonon scattering at this temperature. A further problem is that the field H_c varies as R, i.e., as $l_{\rm el}^{-2}$, rather than $l_{\rm el}^{-1}$ as expected (Fig. 7). A possible explanation for the lowering of $l_{\rm in}$ in our Cu films is given by Schmid.[18] Owing to the relative large impurity scattering ($l_{\rm el} \lesssim 100$ Å) the inelastic scattering length for electron-phonon and electron-electron interaction must be renormalized and depends upon $l_{\rm el}$. In this way the reduction of $l_{\rm in}$ with decreasing $l_{\rm el}$ observed in our Cu films can be understood.

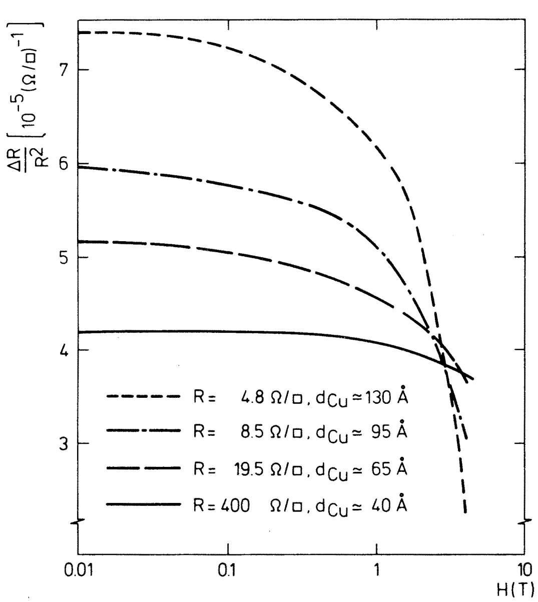

FIG. 5. Normalized increase $\Delta R/R^2$ of the resistance between $T = 10$ and 1 K vs magnetic field H (logarithmic scale) for four Cu films with varying thickness.

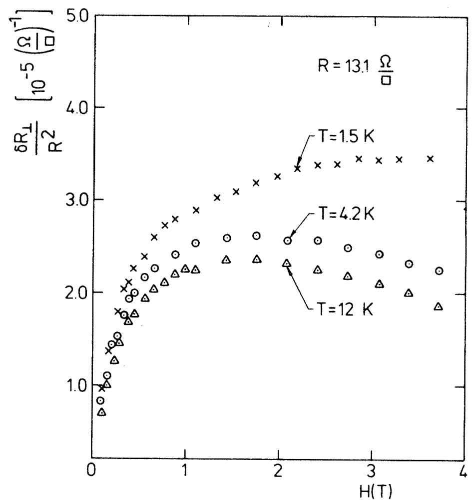

FIG. 6. Normalized transverse magnetoresistance $\delta R_\perp/R^2$ as a function of the magnetic field H (linear scale) for a film with $R = 13.1\ \Omega/\Box$ at three different temperatures.

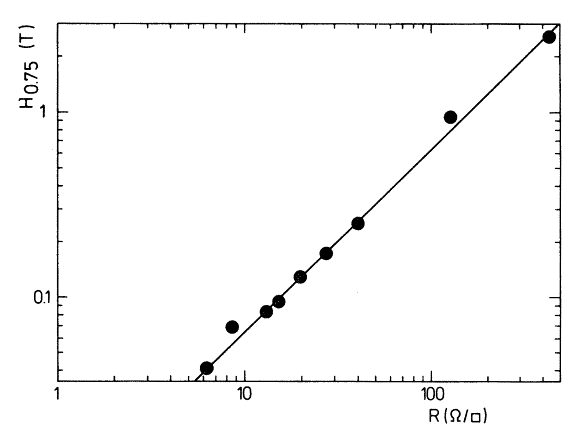

FIG. 7. The field, $H_{0.75}$ at which the transverse negative magnetoresistance $\delta R_\perp/R^2$ reaches $0.75 \times 10^{-5}\ (\Omega/\Box)^{-1}$, as a function of the resistance per square R. The full curve corresponds to $H_{0.75} = aR$, where the constant a is depending upon the structure of the Cu films.

IV. CORRECTIONS TO THE LOCALIZATION THEORY: ELECTRON-ELECTRON AND SPIN-ORBIT INTERACTION

The main predictions of the localization theory—a logarithmic increase of the resistance at low temperatures with a prefactor corresponding to a value of $\alpha P \simeq 2$, a negative magnetoresistance at low perpendicular fields, and no magnetoresistance in a parallel field—are all clearly observed in Cu films. However, the theory in its simplest form [Eqs. (1) and (2)] is unable to explain a number of important experimental results:

(1) The decrease of αP at large values of R (even after correcting for a probable Kondo contribution) [Fig. 2(b)].

(2) The weak temperature dependence of the field H_c, which should, in principle, vary as T^P (Fig. 6).

(3) The linear variation of H_c with R rather than with the resistivity (Fig. 7).

(4) The weak positive magnetoresistance at low fields in the thinner Cu films (Fig. 4).

(5) The fact that experimentally the prefactor of $\ln H$ is somewhat smaller (by about 20%) than $e^2/2\pi^2\hbar$.

It appears that points (1)–(4) can be interpreted as the manifestation of a spin-orbit interaction of moderate strength in Cu films, while point (5) can be equally well explained by the presence of electron-electron interaction. The theory[12,17] predicts that in the presence of a spin-orbit interaction characterized by a scattering time $\tau_{so} \lesssim \tau_{in}$, the perpendicular field at which a negative magnetoresistance appears is determined by the product $l_{so} l_{el}$ rather than by $l_{in} l_{el}$. This explains point (2) since l_{so} is temperature independent, and point (3) since l_{so} is proportional to l_{el}, which is itself proportional to $R^{1/2}$ in the case of continuous films [$l_{el} \propto d$, see insert, Fig. 2(b)]. The theory also predicts that for sufficient strong spin-orbit scattering the magnetoresistance is positive at weak fields which nicely fits with point (4). This interpretation is confirmed by Fig. 8, which shows that the positive magnetoresistance becomes more pronounced at low temperatures (due to a larger l_{in}), although at high fields the behavior remains essentially unchanged.

Finally, the theory predicts a sign reversal of the temperature dependence of the resistance, with α changing from $1(\tau_{so}/\tau_{in} \rightarrow \infty)$ to $-1/2(\tau_{so}/\tau_{in} \rightarrow 0)$. At least qualitatively, this explains the decrease of αP at large values of R. Experimentally, however, there is no indication of a sign reversal. In this connection we note that according to the simple argument of Anderson[19] (loss of one degree of freedom of the electron gas in the strong spin-orbit coupling limit) this sign reversal should not actually take place. Our experiments would be consistent with α changing from 1 to 1/2 rather than from 1 to $-1/2$. A related question is the role played by electron-electron interactions in the temperature and field dependence of the resistance. If spin-orbit interactions are ignored, electron-electron interactions result in the following corrections:

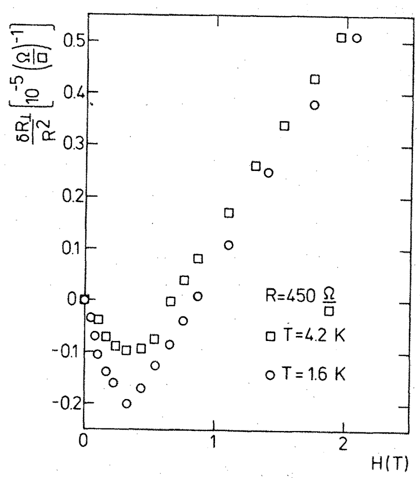

FIG. 8. Positive and negative magnetoresistance at low fields for a 45-Å-thick Cu film.

(i) An additional contribution to the temperature dependence of the resistance given by[3]

$$\frac{\Delta R}{R^2}=(1-F)\frac{e^2}{2\pi^2\hbar}\ln 10 \, . \tag{3}$$

(ii) A positive contribution to the magnetoresistance given by[20]

$$\frac{\delta R_\perp}{R^2}=\frac{F}{2}\frac{e^2}{2\pi^2\hbar}\ln\left[\frac{\hbar/2e}{l_{el}^2 H}\right] . \tag{4}$$

In Eqs. (3) and (4), F is the three-dimensional (3D) screening integral. For Cu we calculated[6] $F \simeq 0.6$. With this value, the correction [Eq. (4)] gives an excellent agreement with the measured magnetoresistance (to better than 5%) with no adjustable parameter. This argument, however, may be partly fortuitous in view of the fact that Eq. (4) must be modified when spin-orbit interactions are not negligible,[21] as is surely the case in the Cu films. The effect of electron-electron interactions is more difficult to identify than that of localization (including its spin-orbit correction). It could very well be that the value $\alpha P \simeq 2$ observed in the thicker Cu films is the sum of a localization term ($\alpha P \simeq 1.5$ instead of 2 due to the spin-orbit correction) and of an interaction term corresponding to $(1-F) \simeq 0.5$, but we have no direct proof that this is actually the case. A further question to which we have no answer is that of the actual temperature dependence of τ_{in}. Because of the spin-orbit correction, it can not be obtained simply from the temperature dependence of H_c. In this connection magnetoresistance measurements at much lower temperatures, in the regime $\tau_{\text{in}} >> \tau_{\text{so}}$, where a more pronounced positive magnetoresistance at low fields is expected, would probably be helpful also.

V. CONCLUSION

The essential signature of quantum localization in 2D— a negative magnetoresistance in small perpendicular fields, no magnetoresistance in parallel fields—has been clearly observed in Cu films with conductances 3 orders of magnitude larger than the characteristic value $e^2/\hbar$. When a spin-orbit interaction of moderate strength[22] is assumed, we find that the overall agreement with theory is truly impressive. Some points—the temperature dependence of the resistance in the strong spin-orbit coupling limit, the exact role played by electron-electron interaction, and the temperature dependence of the inelastic scattering time at low temperature—deserve further study.

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[22]P. Yee and W. D. Knight, Phys. Rev. B 11, (1975). For Cu these authors found $l_{\text{so}} \simeq 150\ l_{\text{el}}$ which is in very good agreement with the values obtained from the corrected $\delta R_{\perp}/R^2$ vs H data. For a Cu film with $R = 7.8\ \Omega/\Box$ we find $l_{\text{el}} \simeq 95$ Å and $l_{\text{so}} \simeq 1.5 \times 10^4$ Å.