Electron localization in two-dimensional percolating gold films

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We have investigated the temperature dependence and magnetoresistance of thin semicontinuous gold films. We have observed a decrease of the quantum corrections to the conductivity due to two-dimensional localization effects for samples close to the percolation threshold. For these samples, quasi-one-dimensional effects appear to become important, presumably developing around the singly connected links of the percolation network. The experimental data are shown to be consistent with a model based on scaling laws of percolation and localization.

I. INTRODUCTION

It has been known for some time that quantum corrections to the conductivity can be expected to occur near the percolation threshold of metal-insulator mixtures.[1] More recently, the interplay between percolation and Anderson localization has been examined theoretically within the framework of the scaling theory of localization[2] and in particular the occurrence of an Anderson transition above the percolation threshold p_c has been shown to be relevant for the understanding of the transport and superconducting properties of a number of metal-insulator mixtures.[3] Furthermore, the relevance of the fractal geometry of the percolation network has been well established for the upper superconducting critical field[4,5] and superconducting fluctuation effects above T_c.[6]

This paper is devoted to a study of weak localization effects in percolating semicontinuous gold films. We have observed that the anomalous magnetoresistance (MR) characteristic of weak two-dimensional (2D) localization saturates near p_c. An interpretation of this result is proposed in terms of a model that describes weak localization on fractals. Very near p_c additional effects such as a strong dependence of the quantum correction to the conductivity on the measuring current and the appearance of an additional MR at high fields are observed. They are shown to be consistent with the occurrence of quasi-1D effects probably at or near the singly connected links or bottle necks of the infinite cluster.

In Sec. II we review very briefly the theoretical background of percolation and weak localization in two dimensions and describe our model. Experimental results are given in Sec. III, data analysis is discussed in Sec. IV, and finally, Sec. V is devoted to conclusions.

II. THEORETICAL BACKGROUND

According to percolation theory, the conductivity σ of a metal-insulator mixture goes to zero at p_c as $\sigma \propto |p-p_c|^{\mu}$. The percolation correlation length diverges near p_c as $\xi_p \propto |p-p_c|^{-\nu}$, and one can define an average conductivity on scales $L \ll \xi_p$. Using a scaling approach it was shown[7] that $\langle \sigma(L) \rangle \propto L^{-\mu/\nu}$ for $L \ll \xi_p$. This result was tested and confirmed experimentally for real systems.[8] Gefen *et al.*[2] considered the scaling theory of localization in the context of percolation. For a homogeneous 2D system the well-known result is[9]

$$\sigma(L) \propto \sigma_0 + \ln L \ .$$

For a d-dimensional fractal they obtained a new functional dependence of the quantum correction to the conductivity:

$$\sigma(L) \propto \sigma_0 - AL^{d-2+\mu/\nu} \ .$$

The temperature dependence of the quantum correction in two dimensions in the absence of a magnetic field, and on spin-orbit and magnetic impurity scattering is given by

$$\Delta\sigma(T) = -\frac{e^2\alpha}{2\pi^2\hbar} \ln\left[\frac{\tau_\epsilon}{\tau_0}\right] \propto \alpha_T \ln T \ , \tag{1}$$

where α is of order 1, $\alpha_T = \alpha P$, and $\tau_\epsilon \propto T^{-P}$, where τ_ϵ is the inelastic relaxation time (τ_0 is the elastic relaxation time). Since there is only one time scale, τ_ϵ, involved, the argument used by Gefen *et al.* may be applied straightforwardly to a percolation network on scales $L \ll \xi_p$, giving a power-law temperature dependence.

In the presence of spin-orbit interaction in the homogeneous case the quantum correction to the conductivity involves more than one characteristic time, and it is not clear how the temperature dependence in this case should be modified for a fractal geometry.

The magnetoconductance in the presence of spin-orbit interaction has been calculated in the weakly localized regime.[10] Neglecting Zeeman splitting and the electron-electron interaction one obtains the following result for the magnetoconductance ($d=2$):

$$\Delta\sigma = -\frac{e^2}{2\pi^2\hbar}\left[\Psi\left[\frac{1}{2}+\frac{1}{a\tau_0}\right] - \frac{3}{2}\Psi\left[\frac{1}{2}+\frac{1}{a\tau_1}\right] + \frac{1}{2}\Psi\left[\frac{1}{2}+\frac{1}{a\tau_2}\right]\right] \equiv -\frac{e^2}{2\pi^2\hbar} f_2 \ , \tag{2}$$

where Ψ is the digamma function, $a = 4DeH/\hbar c$, $\tau_1^{-1} = 4\tau_{so}^{-1} + 2\tau_s^{-1} + \tau_\epsilon^{-1}$, $\tau_2^{-1} = 6\tau_s^{-1} + \tau_\epsilon^{-1}$, D is the electronic diffusion constant, and τ_0, τ_{so}, τ_s, and τ_ϵ are the re-

laxation times due to the elastic, spin-orbit, magnetic impurity, and inelastic scattering, respectively. Notice, that $1/a$ is also the characteristic time for diffusion over the Landau radius. There is no known way to revise this magnetoconductance formula for a fractal geometry.

In this paper we propose an alternative model for localization phenomena in 2D percolation networks. Within the framework of this model, the percolation network—at a given (nonzero) temperature T—can be divided into two basic parts, if $\xi_p \gg (\langle L_\epsilon \rangle, w)$ (w indicates the width of the channel). One part, on the scale of $\langle L_\epsilon \rangle \simeq [D(\tau)\tau_\epsilon]^{1/2}$ consists of many loops of different radii smaller than or of the order of $\langle L_\epsilon \rangle$. The second part is the rest of the network which on this scale does not contain loops smaller than L_ϵ.

The first part can be regarded as a 2D system with an average sheet resistance

$$R(\langle L_\epsilon \rangle) = r_\square \left[\frac{L_\epsilon}{w} \right]^{\mu/\nu}$$

and the second is nearly one dimensional and on the scale L_ϵ has a resistance $R_{L_\epsilon} = r_\square L_\epsilon / w$. Here $r_\square$ is the sheet resistance of the conducting channels. The magnetoresistance, MR, in one dimension in the presence of spin-orbit interaction was derived and studied by Santhanan *et al.*:[11]

$$\frac{\Delta R}{R} = \frac{e^2}{\pi\hbar} \frac{r_\square L_\epsilon}{w} f_1 , \tag{3}$$

where

$$f_1 = \frac{3}{2} \left[1 + \frac{H^2}{48 H_1 H_w} \right]^{-1/2} - \frac{1}{2} \left[1 + \frac{H^2}{48 H_2 H_w} \right]^{-1/2} - 1, \quad H_1 = \frac{\hbar c}{4eD\tau_1}, \quad H_2 = \frac{\hbar c}{4eD\tau_2}, \quad H_w = \frac{\hbar c}{4ew} .$$

To summarize, we propose that the MR of the percolation network will have two contributions [if $\xi_P \gg (\langle L_\epsilon \rangle, w)$]:

$$\frac{\Delta R_\square}{R_\square} = -\frac{e^2 a_2}{2\pi^2\hbar} R(\langle L_\epsilon \rangle) f_2 + \frac{e^2 a_1}{\pi\hbar} \frac{r_\square L_\epsilon}{w} f_1 . \tag{4a}$$

Here, $a_1 + a_2 = 1$ (a_1 and a_2 are relative contributions of 2D and 1D parts to the total relative change in the resistance). Equation (4a) is in agreement with a recent theoretical analysis.[12]

For a homogeneous 2D film, $a_1 = 0$, $a_2 = 1$, and $R(L) = R_\square$, and for a one-dimensional wire, $a_2 = 0$, $a_1 = 1$, and Eq. (4a) is reduced to Eqs. (2) and (3), respectively. Note that, once $L_\epsilon \gg w$, the contribution of the second term in Eq. (4a) becomes negligible for small magnetic fields, since the characteristic fields in the second term are then much larger than in the first one, and prefactors are roughly the same. Thus, for $H^2 < H_1 H_w, H_2 H_w$, we get

$$\Delta\sigma = \frac{\Delta R_\square}{R_\square^2} = -\frac{e^2 a_2}{2\pi^2\hbar} \alpha_H f_2 . \tag{4b}$$

Here $\alpha_H \equiv R(\langle L_\epsilon \rangle)/R_0$.

The temperature dependence of the quantum correction to the macroscopic dc conductivity will also contain two contributions. In the weak localization regime [$L_\epsilon \ll \xi_l$, where ξ_l is the localization length] the one-dimensional part varies with temperature as

$$\Delta\sigma(T) = -\frac{e^2}{2\pi^2\hbar} \left[\frac{T_0}{T} \right]^{P/2}$$

when $T \ll T_0$. Here T_0 is the temperature at which $\tau_\epsilon = \tau_0$. In the regime where $L_\epsilon \simeq \xi_l$, $\sigma(T) \propto T^{-2}$.[13]

In the case when $\xi_p \ll L_\epsilon$ we expect the homogeneous formula [Eq. (2)] to be valid. The same model could be used to describe interaction effects. The distinction between 1D and 2D parts of the network in this case should be made by introducing the temperature length $L_T = (\hbar D/k_B T)^{1/2}$ instead of L_ϵ. The temperature dependence of the resistance due to interactions is similar to that of the localization in 2D and 1D systems, and usually cannot be distinguished experimentally. The MR in two dimensions and one dimension due to interaction effects can be distinguished from MR caused by localization, since the former usually has a much higher characteristic field.

III. EXPERIMENT AND RESULTS

The samples were obtained by vacuum evaporation of pure Au (99.999 and 99.99 [samples marked by asterisk (*)]) from Al_2O_3 coated Mo boats onto glass substrates. As was shown and discussed in a previous paper,[14] there is a critical average thickness t_c below which the conductivity of Au samples goes to zero. The variation of the conductivity with the average thickness t in the vicinity of the critical thickness [$\sigma \propto (t - t_c)^\mu$, with $\mu = 1.25 \pm 0.05$] was shown to be due to a variation of the surface coverage, i.e., to reflect the percolation law $\sigma \propto (p - p_c)^\mu$.

Microscopic observations as well as this behavior of the conductivity indicated that the films had a percolation structure. This was alter confirmed by the direct observation of the self-similar behavior $R_\square \propto L^{\mu/\nu}$.[8] Measured values of $R_\square$, being substituted into $R_\square = r_\square (\xi_p/w)^{\mu/\nu}$ allow us to estimate the approximate values of ξ_p for different samples (we estimate that $r_\square \simeq 20\Omega$ for samples not marked by an asterisk and twice as large for marked samples). The characteristic parameters of the sample are summarized in Table I.

All the samples have a metallic behavior, i.e., the resistivity goes down with temperature at high temperatures and shows only a weak increase below 5 K (see Fig. 1). It is remarkable that all samples, with $R_\square$ values ranging from about $10\Omega/\square$ up to $30\,000\Omega/\square$, show similar resis-

TABLE I. Summary of the characteristic parameters.

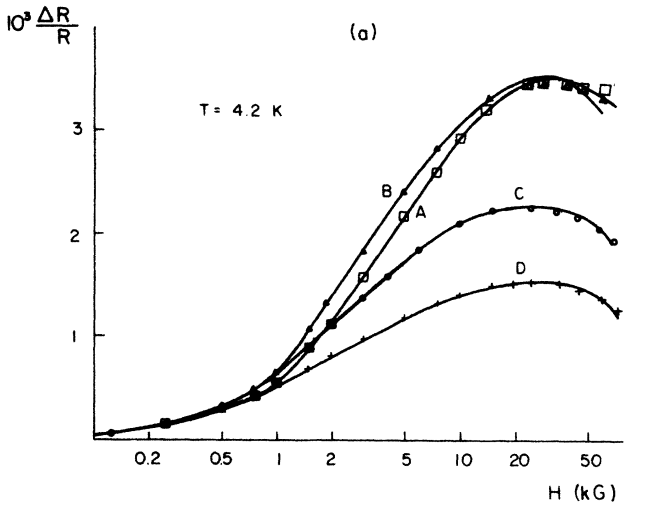
Sample	$\bar{l}$ (Å)	$R_{\square,4.2}$ (Ω)	$\xi_p = \frac{R_\square(150\text{ Å})^a}{r_\square}$	$L_{\epsilon,4.2}$ (Å)	$R_R = \frac{R_{300}}{R_{4.2}}$
A		≃30000	22.5 μm	950	1.08
B		678	5100 Å	950	1.09
C		272	2000 Å	1250	1.09
D		100	750 Å	1550	1.1
*A**	36	600	2250 Å	550	1.3
*B**	39	280	1000 Å	700	1.2
*C**	41	140	500 Å	900	1.1
*D**	67	25	(homogeneous)	2000	1.16
*E**	81	8.5	(homogeneous)	2800	1.3

[a] $r_\square = 20\ \Omega$ (for samples without the asterisk). $r_\square = 40\ \Omega$ (for samples marked by the asterisk). The width of the channel $w = 150$ Å.

tivity ratios R_R between ambient and liquid He temperatures. This proves that on the local (channel) scale *all* samples are indeed similar and that the large values of $R_\square$ are due to the macroscopic (percolative) structure. When the low-temperature dependence is compared to formula (1) predicted by the scaling theory of localization and also by an alternative theory based on electron-electron interaction effects, we find that the prefactor $\alpha_T = \alpha_P$ is reduced from $\alpha_T = 2$ for homogeneous samples down to $\alpha_T = 0.2$ for the sample closest to p_c (Table II).

In the presence of a magnetic field in both the perpendicular and parallel orientations, Au films exhibit a positive MR (Fig. 2). The data was taken for fields up to 7 T at different temperatures. The MR decreases as the temperature increases, and at 77 K it is completely unobservable. Qualitatively, the dependence of MR on temperature and the sign of MR are as expected for samples with strong spin-orbit interaction. However, a quantitative analysis shows that the prefactor of the logarithmic term α_H which in this regime should be equal to 1 (for homogeneous 2D samples) decreases considerably for samples near the continuity threshold (Table II).

A similar observation was reported by Kawaguti and Fujimori,[15] who explained it in terms of the homogeneous formula (2) by introducing an unreasonably high value of τ_0 in the first term of (2). They tested this formula for their dirtiest sample at different temperatures. However,



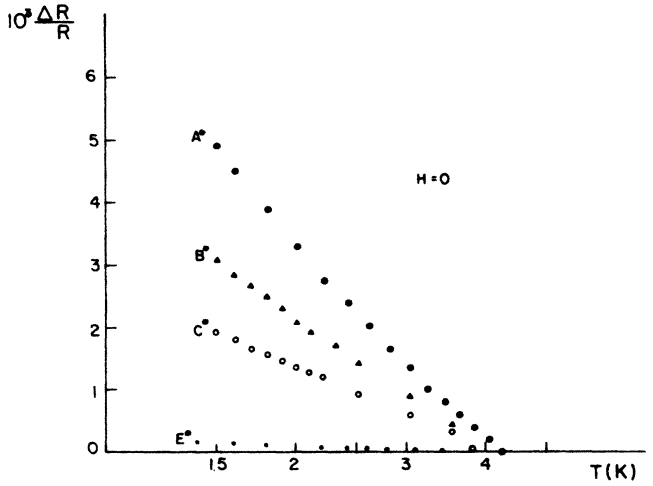


FIG. 1. Variation of the resistance with temperature for samples marked by asterisk.

FIG. 2. Magnetoresistance data points and fitted curves. In (a) note the similarity between samples *A* and *B*. In (b) the higher values of H_ϵ is presumably due to a higher scattering rate (see text).

TABLE II. The parameters extracted from the data analysis.

Sample	α_T	$a_2\alpha_{H,2.1}$	$a_2\alpha_{H,4.2}$	$a_2\alpha_{H,9.1}$	$R(L_{\epsilon 4.2})$	$H_{\epsilon,2.1}$	$H_{\epsilon,4.2}$	$H_{\epsilon,9.1}$	H_{so}
A	0.2	0.008	0.007		220 Ω	100 G	175 G		6 kG
B	0.35	0.4	0.35	0.28	240 Ω	89 G	170 G	460 G	5.8 kG
C		0.58	0.52	0.48	140 Ω	58 G	100 G	450 G	5.6 kG
D			0.8		80 Ω		67 G		5.0 kG
*A**	0.8	0.51	0.5		300 Ω	380 G	470 G		5.5 kG
*B**	1	0.74	0.72		200 Ω	250 G	350 G		6.0 kG
*C**	1.4	0.8	0.8		110 Ω	150 G	190 G		5.0 kG
*D**			1		25 Ω		42 G		2.5 kG
*E**	2		1		8.5 Ω		20 G		1.0 kG

since the clean samples exhibit the correct slope α_H up to relatively high fields, the value of τ_0 should be very small even for clean samples. Thus, for dirtier samples τ_0 should be negligible. In the next section, where we present our interpretation of the data, we give some estimates about the value of τ_0 which is roughly 1 order of magnitude smaller than the value introduced in Ref. 15.

The MR and temperature dependence of the sample closest to the percolation threshold were measured using different intensities of the current I (Fig. 3). We found that the temperature dependence of the sample resistance strongly depends on the value of the current intensity in the range 10 nA $< I <$ 10 μA. The value of α_T given in Table II is reduced by a factor $\sim$25 as the current is increased from 10 nA up to 10 μA. However, in this range of measuring currents the MR at low magnetic fields *remains unaffected* by the current increase. We also observe the appearance of an additional positive MR at high magnetic fields for small currents (Fig. 4), which becomes more pronounced at lower temperatures. This behavior is suggestive of nonhomogeneous heating, which does not affect the parts responsible for the low-field (2D) MR, but rather the more weakly connected (quasi-1D) parts of the infinite cluster. For a current $I = 50$ μA we observed a macroscopic heating of the sample which leads to a strong decrease of the MR at all magnetic fields and to a vanishing temperature dependence. A similar, but much smaller, effect (α_T reduces by a factor $\sim$2) was also observed in sample *B* (not shown).

IV. DATA ANALYSIS AND DISCUSSION

The magnetoresistance data was fitted to formula (4b), where we dropped the first term in f_2 [Eq. (2)] neglecting the effect of elastic collisions on the MR. The reason for this is as follows: our estimation of τ_0 gives a very high value of the characteristic field $H_e = \hbar c/4De\tau_0$. Since the physical thickness of the gold channels is of the order of 50 Å and the scattering of electrons at the surface boundaries is known to be diffuse, we expect the value of

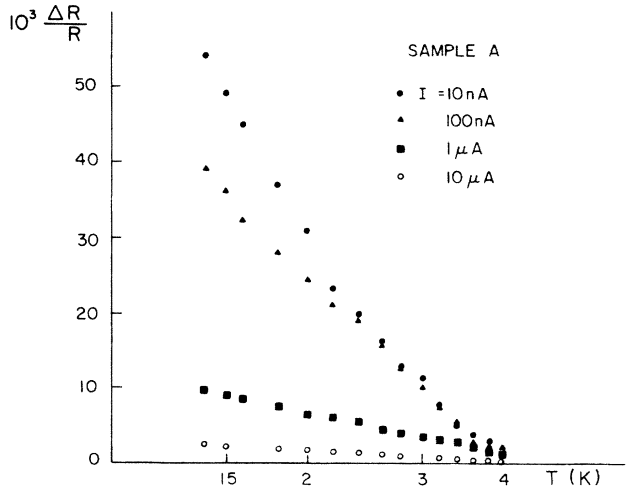


FIG. 3. Variation of the resistance of sample *A* with temperature for different measuring currents.

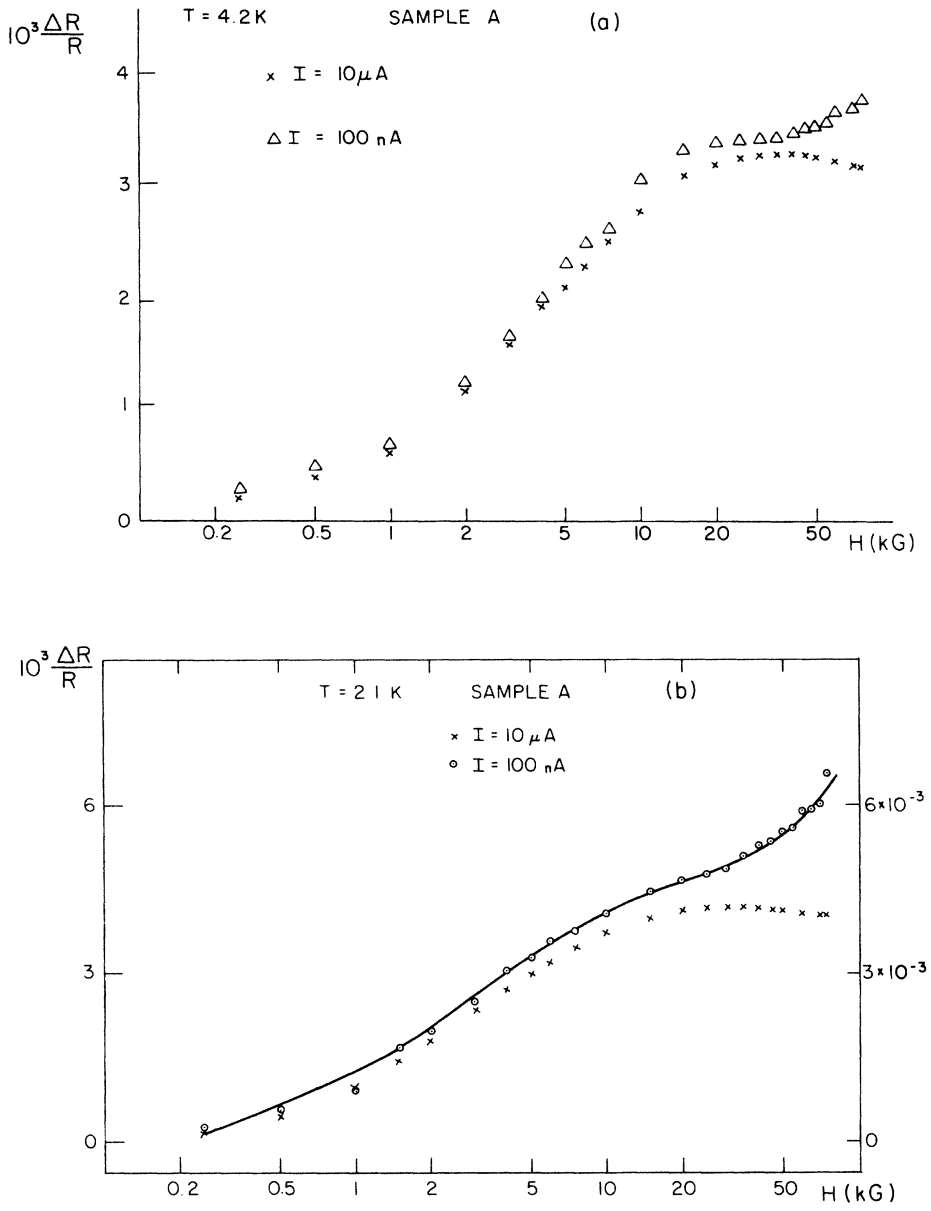


FIG. 4. MR curves of sample A for different curves (a) 4.2 K and (b) 2.1 K.

l, the elastic mean free path, to be close to 50 Å. Moreover, using Mathiessen's rule we can extract a value for l from the temperature resistivity ratio, which is $R_R \simeq 1.1$, which also gives $l \simeq 50$ Å. Substituting this value of l in $l = V_f \tau_0$ and using $V_f = 1.4 \times 10^8$ cm/s, we obtain for the characteristic field $H_e \simeq 6$ T.

The MR data was fitted using a least-squares-fitting procedure and for each sample we have extracted the parameters

$$a_2 \alpha_H = \frac{a_2 R(\langle L_\epsilon \rangle)}{R_\square} ,$$

H_ϵ, and H_{so}. They are summarized in Table II. Figure 2 shows the experimental points and the fitted curve. The fit is excellent for the samples with low and medium $R_\square$ and not as good for samples close to the percolation threshold at high fields. The value of $a_2 \alpha_H$ in (4a) which should be equal to 1 for homogeneous films varies from 1 for low-$R_\square$ films to $\frac{1}{100}$ for samples close to the percolation threshold.

Our samples can be classified in the following way.

(1) Homogeneous 2D samples (E^*,D^*) for which we obtain $\alpha_T = 2$, $\alpha_H = 1$ ($a_2 = 1$), and the variation of H_ϵ and H_{so} with $R_\square$ is consistent with the 2D theory of localization.

(2) Nonhomogeneous samples that can be divided into 2 categories. In the first category (samples C^*,B^*,A^*,D,C) the fit to the 2D localization theory is still good if $a_2 \alpha_H$ is allowed to take a value smaller than 1 and α_T is smaller than 2. Both parameters decrease as $R_\square$ increases. For these samples the inhomogeneous heating effects are not observed; therefore, it is plausible to assume that $a_2 = 1$ ($a_1 = 0$). $R(\langle L_\epsilon \rangle)$ and H_ϵ *increase* with $R_\square$, and *both* $R(\langle L_\epsilon \rangle)$ and H_ϵ are temperature dependent (Table II).

We call these samples "inhomogeneous 2D."

The second category of inhomogeneous samples is represented by samples A and B. Here we observe a strong dependence of α_T on the measuring current and an additional positive MR at high magnetic fields for small measuring currents (sample A), while, as described before, $a_2\alpha_H$ remains unchanged. This suggests that a_1 is now finite, as discussed below. The prefactors α_T and $a_2\alpha_H$ are reduced as $R_\square$ is increased. The value of H_ϵ is saturated, and the value of $a_2R_\square(L_\epsilon)$ for sample A is smaller than for sample B. As we now show, this classification is consistent with an interpretation based on the scaling laws of localization and percolation.

As was mentioned in Sec. II, we have two basic length scales involved in the problem, L_ϵ and ξ_p. For the inhomogeneous 2D samples L_ϵ is just of the order of $\xi_p(L_\epsilon/\xi_p$ varies roughly from $\frac{1}{2}$ to 2 for all these samples, except for A^* where the ratio is about 4). Therefore, any heating effects must affect similarly all parts of the sample, since electrons are thermalized over the length L_ϵ. The localization behavior is two dimensional, but the values of H_ϵ and $R(\langle L_\epsilon\rangle)$ are ξ_p dependent and the prefactors α_T and $R(\langle L_\epsilon\rangle)/R_\square$ are reduced as ξ_p/L_ϵ is increased. The variation of $R(\langle L_\epsilon\rangle)/R_\square$ with temperature is due to the variation of the ratio ξ_p/L_ϵ. The temperature dependence is essentially logarithmic except for the dirtier sample A^* for which it is slightly stronger. This is, probably, also due to the variation of the ratio ξ_p/L_ϵ with temperature. The higher values of H_ϵ and $R(\langle L_\epsilon\rangle)$ for the samples marked by an asterisk and the weaker dependence of H_ϵ on temperature are, probably, due to a high concentration of magnetic impurities and a higher value of the microscopic resistivity of the channels $r_\square$.

For the inhomogeneous mixed 2D-1D samples (A,B), where $\xi_p \gg L_\epsilon$ the quantum diffusion of electrons *during* the time τ_ϵ in some parts of the network resembles a 1D motion. We propose that these parts are responsible for the high-field MR as well as for the major part of the temperature dependence (for small measuring currents). In this regime we anticipate the saturation of $R(\langle L_\epsilon\rangle)$ and H_ϵ since 2D quantum interference effects occur on scales for which the network is self-similar. Since the value of a_2 becomes smaller as ξ_p/L_ϵ increases, the value of $a_2R(\langle L_\epsilon\rangle)$ for sample B is somewhat higher than for sample A. For the same reason as for the inhomogeneous 2D samples, the MR curves for A and B exhibit different values of H_ϵ and $R(\langle L_\epsilon\rangle)$ at different temperatures.

We now turn to the anomalous heating effect observed in these samples. We first observe that at large electric fields (and over a wide range of electric fields) the temperature dependence is not completely suppressed, as would be the case in a homogeneous sample.[16] It is weakened, but there is no sign of saturation at low temperatures. This is indicative of nonhomogeneous heating.

Indeed, close to p_c, the number of independent current carrying paths, which varies as $(\xi_p)^{-1}$, is strongly reduced. The current is then strongly concentrated in the regions at the singly connected links of the infinite cluster and the neighboring regions which constitute the dilute parts of the network (and are along the quasi-1D parts). Heating effects will preferentially occur at and near the singly connected links, *and can remain local* since $L_\epsilon \ll \xi_p$. This nonhomogeneous heating therefore suppresses the high-field (1D) MR without affecting the low-field (2D) MR up to $I=10\ \mu$A (Fig. 4).

We cannot conclude whether there is a quantitative agreement between the high-field MR and the predicted 1D MR in Eq. (4a), since the range of magnetic fields is insufficient for that purpose. The characteristic field at which 1D MR effects become important is $(48H_2H_w)^{1/2}$. Taking at 2.1 K $H_2=100$ G, and with $w\simeq 100$ Å, this field is of the order of 15 kG. This is consistent with the data shown in Fig. 4(b). Interaction effects would also occur in the same range of fields. More data at lower temperatures and higher fields is required to distinguish between 1D localization and 1D interaction effects.

Since the 2D contribution to the MR is negative at high magnetic fields (finite spin-orbit) and 1D MR makes a positive contribution, the total MR at high magnetic fields can be confused with a saturation behavior, if one performs the measurement in certain range of currents. We conjecture that this is the reason for the saturation of the MR in gold films at high magnetic fields reported recently.[17]

By comparing the values of the prefactors $a_2R(\langle L_\epsilon\rangle)$ for samples A and B and assuming the saturation of $R(\langle L_\epsilon\rangle)$ we estimate the value of a_1 to be at least 0.1 and not much larger than that (the observed nonlinearity in sample B being 1 order of magnitude smaller than in sample A). Nevertheless, the value of α_T is reduced much less than $a_2\alpha_H$. This is especially pronounced for sample A, for which the value of α_T is about 25 times larger than α_H. This is an indication that *the one dimensional parts* of the cluster make a *dominant contribution* to the temperature dependence. Moreover, the dependence is not logarithmic, since the value of α_T constantly increases as the temperature decreases. A log-log plot of this temperature dependence gives a power close to -1. In short, the 1D parts constitute a small fraction of the infinite cluster ($a_1\simeq 0.1$), but make a dominant contribution to the temperature dependence.

This surprising result suggests that many of the 1D channels have a localization length ξ_l not much larger than L_ϵ. In this regime it was shown[13] that the conductance of a wire should decrease with temperature as T^{-2}. A decrease of the temperature by a factor of 2 (from 4 K down to 2 K) should give a 40% change of the total resistance since $a_1=0.1$. The observed 5% change suggests that the wires are close but not yet in the Thouless regime ($L_\epsilon \leq \xi_l$), which, as far as we know, has not yet been observed for 1D wires.

V. CONCLUSIONS

The conventional MR formula (2) is irrelevant for sufficiently inhomogeneous samples ($\xi_p \gg L_\epsilon$). We propose that it should then be modified in the way presented in Eq. (4b). The variation of the parameters α_H and α_T with $R_\square$ confirms the validity of the scaling laws of percolation and localization. The high-field MR and the temperature dependence for small measuring currents is attributed to quasi-1D localization and interaction effects.

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