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Hall coefficient and conduction in a 2D percolation system

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Résumé. — Nous présentons des mesures complémentaires de la conductivité et du coefficient de Hall dans des films d'or semicontinus bidimensionnels. On montre que la conductance de couche près du seuil de percolation suit une loi de puissance avec l'exposant $\mu = 1,25 \pm 0,05$. Le coefficient de Hall ne montre pas de comportement critique près du seuil.

Abstract. — Complementary measurements of the conductivity and the Hall coefficient in 2D semicontinuous thin films of Au have been performed. It is shown that the sheet conductance near the percolation threshold obeys a power law with the exponent $\mu = 1.25 \pm 0.05$. The Hall coefficient does not exhibit critical behaviour in the vicinity of the threshold.

Dilute metals have been widely studied in recent years, both theoretically and experimentally [1, 2]. In particular metal-insulator mixture films have been shown to be well described by percolation theory near their conduction threshold [2, 3]. For example the conductivity, σ , near that threshold obeys a power law of the type

$$\sigma \propto (p - p_c)^\mu, \quad p \geq p_c \quad (1)$$

with p being the metal fraction [3-6]. Measurements of the 2D to 3D crossover in the resistivity [6] have also confirmed that description.

Most measurements have been performed [3-6] in thin mixture films. Close enough to the percolation threshold a 2D behaviour is always observed since the percolation correlation length becomes large compared to the film thickness. Nevertheless, 2D percolation behaviour *has never been studied in a pure 2D system with respect to transport properties*. In recent papers, the existence of geometric percolation characteristics in semicontinuous thin films of Pb [7] and Au [8] films have been reported. We report here the first quantitative study of transport properties (conductivity and Hall effect) showing that the underlying geometry indeed affects the macroscopic transport properties of such films.

The Hall coefficient is complementary to conductivity, but depends on independent microscopic properties of the metal. It has been predicted by several authors that the macroscopic Hall coefficient will have a power law behaviour in the vicinity of the percolation threshold, in 3D the exponent being between 0.3-0.5 [9-10]. Such behaviour was observed in granular films of Al-Al₂O₃ [11]. In 2D, however, it was shown already several years ago [12] as well as more recently [9, 10] that no divergence should be observed, the Hall coefficient remaining that of the pure metal in the whole metallic range.

In this letter we show that equation (1) for 2D holds with $\mu = 1.25 \pm 0.05$ and that the Hall coefficient remains constant near p_c and is close to that of the pure metal.

Micrographs of our Au films in the vicinity of the percolation threshold (not shown) indicate the same morphology as that studied in references [7] and [8], i.e. that of a random percolation network. The measurements of the conductivity were performed *in situ* during the evaporation using a 4-wire voltage source of 10 mV [14]. The average thickness $\bar{t}$ was read from the analogue output of the quartz crystal thickness monitor. The values of σ and $\bar{t}$ were recorded by an on-line computer.

Figure 1 shows the variation of the sheet conductance $G_{\square}$ with $\bar{t}$. $G_{\square}$ was obtained after correcting for contact series resistance ($\sim 1.5 \Omega$). For thicknesses smaller than $\bar{t}_c = 22 \text{ \AA}$ the film is discontinuous. Above $\bar{t}_c$ the log-log plot in figure 2 shows a power law variation of G as a function of $(\bar{t} - \bar{t}_c)$, with an exponent $\mu = 1.25 \pm 0.05$.

It is known [15, 16] that the conductivity of polycrystalline gold films of uniform thickness obeys perfectly the size effect law of Fuchs [17] in both limits of thicknesses, i.e. $K = \frac{\bar{t}}{l_e} \gg 1$ and $K \ll 1$, where $l_e = 400 \text{ \AA}$ is the mean free path for gold at room temperature, if diffuse reflection of the boundaries are assumed. The theoretical curve of the conductivity variation with thickness is shown in figure 1 by a solid line. Our results fit this curve for thicknesses greater than $50 \text{ \AA}$. Hence beyond $50 \text{ \AA}$ the film is essentially *uniform*. This value of the thickness is roughly twice the value of $\bar{t}_c$ and since the value of the critical concentration in 2D is $p_c \simeq 50 \%$, we conclude that in the range $22\text{--}50 \text{ \AA}$ the fraction of the covered area varies essentially linearly with $\bar{t}$. In other words the proportionality $(p - p_c) \propto (\bar{t} - \bar{t}_c)$ is not just valid for small values of $(p - p_c)$ but holds over an extended range of concentration. This claim is admittedly based upon indirect evidence. It could in principle be checked on films removed from the vacuum system, but the instability problems of

Fig. 1. — Thickness dependence of the sheet conductance. Fuchs curve has been calculated with $l_e = 400 \text{ \AA}$.

Fig. 2. — Power law dependence of the sheet conductance near $\bar{t}_c$. The slope $\mu = 1.25 \pm 0.05$.

thin gold films discussed below would probably render such a check very problematic. We note that our conductivity measurements also imply that films with $\bar{t} \lesssim 50$ Å are in fact composed of grains or clusters of grains with a fairly uniform physical thickness $t \simeq 50$ Å.

This description is consistent with resistivity ratio measurements on Hall effect samples, which give a value independent of $\bar{t}$, even near $\bar{t}_c$ (Table I). However, the value of $\bar{t}_c$ is about 50 % larger than for *in situ* measured films. A still higher value of $\bar{t}_c$ ($\gtrsim 60$ Å) is reported in reference [8]. $\bar{t}_c$ values are known to vary significantly depending on details of samples preparation. We note that *in situ* samples were deposited on glass, and Hall effect samples on glass covered with SiO.

Table I.

$\bar{t}$ (Å)	$R_{\square}$ (Ω) (at 300 K)	$R_{\square}$ (Ω) (at 4.2 K)	$\frac{V(H)}{I.H} \times 10^{-2}$ (C/m ⁴)	$R_{H2D} \times 10^{11}$ (C/m ³)	$R_{H3D} \times 10^{11}$ (C/m ³)
36	775	600	2	10	7.2
41	155	140	1.9	9.5	7.8
67	29	25	1.4	9.6	9.6
81	11	8.5	1.1	8.9	8.9
100	9	7	0.65	6.5	6.5

Furthermore, measurements on our Hall effect samples and those of reference [8] were performed *after* removal from the vacuum system. This is probably the main reason for the higher values of $\bar{t}_c$ and p_c ($p_c \simeq 74\%$ from Ref. [8]) of these films, compared to those measured *in situ*: gold films are known to be fairly unstable, room temperature annealing leading to partial agglomeration [18]. The dispersion in the conductivity values of our Hall effect samples (Fig. 1) is probably also related to the films instability. It appears that only *in situ* conductivity measurements are appropriate for a precise determination of critical indices in gold films.

The Hall effect samples were prepared with a special arrangement of contacts [13] by evaporation of Au from a boat unto a microscopic slide at room temperature. Before deposition the slide was covered with a positive photoresist to allow lift-off photolithography. A 400 Å SiO film provided the support for the Au film after dissolving the photoresist in acetone for TEM observation. Films with $t \gtrsim 70$ Å were found to be essentially uniform. In a pure 2D system the Hall coefficient should not depend on $\bar{t}$ (the average thickness) if, as we have argued, the thickness t of covered areas (physical thickness) remains constant for all concentrations $p < 1$. The quantity that should remain constant near p_c is therefore $V(H)/IH$. This is actually what is observed (Table I) for the 2 thinner samples. We define :

$$R_{H2D} = \frac{V(H) t}{I \cdot H}. \quad (2)$$

For thicker samples for 3D formula $R_H = V(H) \bar{t}/IH$ should be applied.

In order to compare Hall coefficients for the whole range of thicknesses, we used equation (2) with $t = 50$ Å for the first two samples and 3D formula for the three thicker ones. The Hall coefficient shows a very moderate increase near p_c . The two thicker samples which are considered uniform should have the bulk value of the Hall coefficient which is $R_{Au} = 7.2 \times 10^{-11}$ [C/m³] in reasonable agreement with the data. The Hall coefficient was found to be independent of the applied magnetic field up to 1.3 T and of the current density up to 1×10^3 A/cm². The measurements were performed at 300 K, 77 K and 4.2 K. The Hall coefficient in this range of temperature remained unchanged, while as noted above the resistance decreased at low temperature.

In conclusion, the conductivity of semi-continuous Au films near the percolation threshold shows a power law behaviour with a conductivity index $\mu = 1.25 \pm 0.05$ in excellent agreement with the latest transfer matrix computation ($\mu = 1.28 \pm 0.03$) for 2D systems [19]. The Hall constant remains near the threshold very close to the bulk value, also in agreement with theoretical predictions.

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