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Short Communication

## Experimental observation of a doubly percolating system

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**Résumé.** — Nous montrons qu'au cours de la formation des couches minces d'antimoine apparaissent deux seuils de percolation. Le premier correspond à la transition isolant-conducteur déjà observée sur d'autres films métalliques. Le deuxième reflète la cristallisation de la couche et peut être compris grâce à la percolation par invasion.

**Abstract.** — We show that antimony thin films present two percolation transitions. The first corresponds to the well-known insulator-conductor transition already observed in other metallic films. The second corresponds to the crystallization of the film and may be compared to invasion percolation.

The percolation model has become a powerful tool in the modeling of many natural systems where connectivity plays the leading part (for a review, see [1-3]). The important point in percolation is the existence of a threshold above which long-range interactions can be found. Usually, percolation is introduced by the following example: the sites of a primitively insulating network are progressively filled at *random* with conducting material and, above a critical conductor concentration (the percolation threshold) a current can flow between the edges of the network. Experimentally, many groups [4-7] have argued that the insulator-conductor transition observed during the growth of metallic thin films on insulator substrates could be understood in the framework of the percolation model. In this paper, we want to show an experiment where, for the first time to our knowledge, a *double* percolation takes place. We first observe an insulator-poor conductor transition and then a poor conductor good conductor one.

We have measured the electrical conductance of ultra thin-films obtained by molecular beam deposition of antimony on an insulator substrate as a function of the deposit thickness. The antimony metal heated at about 853 K is vaporized and deposited at room temperature on a corning glass substrate with two predeposited chromium electrodes (residual pressure  $10^{-4}$  Pa).

The thickness is simultaneously controlled by a crystal quartz rate monitor. To measure the desorption rate on the substrate, the thickness of the deposit was checked by Rutherford Backscattering Spectrometry (R.B.S.). For electrical measurements, the applied potential is 1 V, the minimum measurable conductance with our device being about  $2 \times 10^{-14}$  S. The sample characteristics are the following: length of the film (between the two electrodes): 3 mm, width of the electrode: 1.5 mm. The growth of thin films prepared by antimony deposition has already been extensively studied by several workers. It has been established that:

- i) the incident  $\text{Sb}_4$  molecules nucleate on preferential sites, giving raise to islands which diameters increase along with matter deposition (three dimensional layer growth) [8, 9];
- ii) below a critical thickness almost all supported antimony particles are in the amorphous state (see for example [9-11] and references therein). A few particles are crystallized [11];
- iii) above the critical thickness, crystallization occurs [9-11]. The few crystallized particles can be some embryos of the amorphous film crystallization [11];
- iv) the critical thickness corresponds to the thickness at which the film becomes almost continuous [12, 13].

The experimental current versus thickness curve obtained for the antimony deposition at room temperature is shown in figure 1. A more detailed account of the experiments, including transmission electron microscopy observations will be published separately [14]. Below 18.5 nm, the supported particles are well separated, allowing no detectable current to cross the sample. Then a first current jump occurs, indicating an insulator-conductor transition. This jump has been observed by many groups [4-7] working on evaporation of thin metallic films and is interpreted as the percolation of the metallic crystalline phase. Then, the first current jump of figure 1 can be attributed to the percolation of the amorphous phase (recall that the particles are essentially amorphous, point (ii)) This is confirmed by the absolute magnitude of the conductance around 21 nm, characteristic of amorphous antimony [15, 16]. At this stage, many amorphous islands begin to be connected with their neighbors. From (iii) and (iv), it can be deduced that, at that stage, the film begins to crystallize. The few crystalline islands propagate their structure to the amorphous particles they are connected to and the proportion of the crystalline phase increases with increasing thickness (which is proportional to the time). Eventually, a continuous *crystalline* path crosses the sample, joining the two electrodes. Then it seems reasonable to interpret the second jump at 30 nm as the percolation of the crystalline phase among the amorphous one. This has been confirmed by electron microscopy observations [14].

The previous analysis can be qualitatively improved by using the effective medium approximation [17, 18] to calculate the conductance of the film. The different phase proportions are defined as follows: a given island can be isolated and amorphous (with probability  $p_1$ ), isolated and crystalline ( $p_4$ ), connected to another island and amorphous ( $p_2$ ) or connected and crystalline ( $p_3$ ). In the following we should neglect  $p_4$  (the number of isolated crystalline islands corresponds to the few crystallized particles of point (iv) and remains always weak [11]). Then  $p_1 + p_2 + p_3 = 1$ . In the effective medium approximation, the distribution of conductances  $\rho(\sigma)$  is replaced by a homogeneous lattice, where all bonds have the same conductance  $\Sigma$ .  $\Sigma$  is determined by the condition (we assume a coordination number  $z = 4$  for our two-dimensional lattice)

$$\int \rho(\sigma) \frac{\Sigma - \sigma}{\sigma + \Sigma} d\sigma = 0 \quad (1)$$

Using the distribution  $\rho(\sigma) = p_1 \delta(\sigma - \sigma_1) + p_2 \delta(\sigma - \sigma_2) + p_3 \delta(\sigma - \sigma_3)$ , where  $\delta$  is the Dirac distribution, one gets easily the condition

$$p_1 \frac{\Sigma - \sigma_1}{\Sigma + \sigma_1} + p_2 \frac{\Sigma - \sigma_2}{\Sigma + \sigma_2} + p_3 \frac{\Sigma - \sigma_3}{\Sigma + \sigma_3} = 0 \quad (2)$$

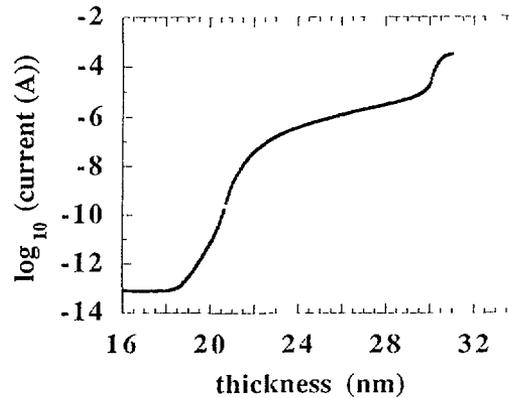


Fig. 1. — Experimental current versus thickness curve obtained at room temperature. Deposition rate:  $0.05 \text{ nm.s}^{-1}$

Taking  $\sigma_1 = 0$ ,  $\sigma_2 = 1$  and  $\sigma_3 = a$ , one gets

$$\Sigma(p_1, p_2, p_3) = 1/2 \left\{ -a(p_1 + p_2 - p_3) - (p_1 - p_2 + p_3) + \Delta^{1/2} \right\} \quad (3)$$

with

$$\Delta = \{a(p_1 + p_2 - p_3) + (p_1 - p_2 + p_3)\}^2 - 4a(p_1 - p_2 - p_3). \quad (4)$$

It is of course possible to study  $\Sigma$  as a function of two of the  $p_i$ . We prefer here to connect the different phase proportions to the thickness of the film, in order to reproduce the experimental curve of figure 1. The experimental observations suggest that the  $p_i$  evolutions could be reproduced by the following functions:

$$p_1 = \frac{1}{1 + \exp \frac{e - e_1}{w_1}}, \quad p_3 = \frac{1}{1 + \exp - \frac{e - e_3}{w_3}} \quad \text{and} \quad p_2 = 1 - p_1 - p_3. \quad (5)$$

$e_1$  (resp.  $e_3$ ) represents the thickness at which  $p_1$  (resp.  $p_3$ ) begins to decrease (resp. increase). The  $w_i$  characterize their evolution rate. We see in figure 2 that we are now able to reproduce the main features of the experimental curve. Evidently, we are not trying to fit the experiments, but rather to show that the essential phenomena have been caught.

We have presented an experimental system where a double percolation is found. Clearly, our theoretical analysis should be improved. For example, the amorphous-crystalline transition, which starts from some isolated embryos could possibly be compared to some kind of *invasion* percolation [19]. A detailed study will be published separately [20]. The evolution of the phase proportions has to be improved to render it more physical and much has to be said on the interpretation of the first jump in the framework of pure, geometrical percolation [21]. From an experimental point of view, it will be interesting to follow the conductance evolution as a function of time (which should be proportional to  $p_3$ ) at constant thickness (then at constant  $p_1$ ) after the first jump.

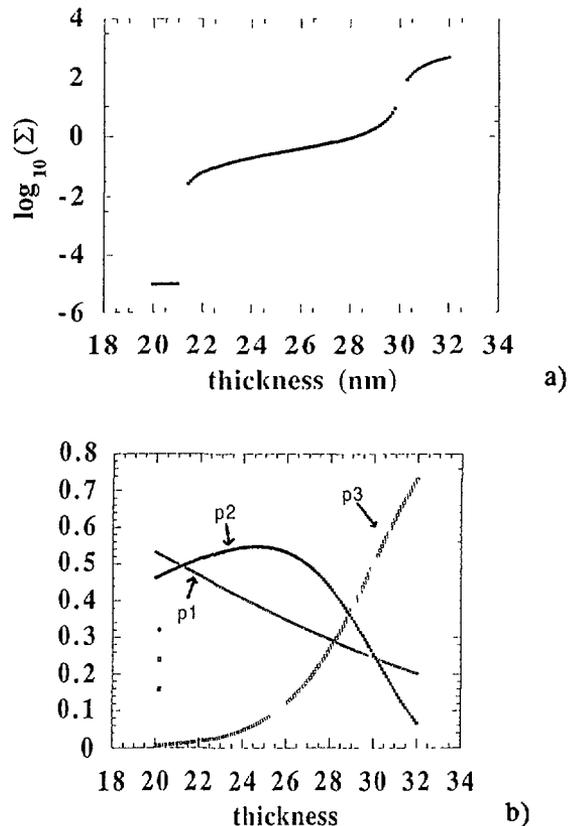


Fig. 2. — a) Conductance versus thickness curve calculated using the EMA approximation. The conductivity before 21 nm has arbitrarily been fixed at  $10^{-5}$  S.cm $^{-1}$ . The figure 2b shows the evolution of  $p_1$ ,  $p_2$  and  $p_3$ . The following values have been assumed in equation (5):  $\epsilon_1 = 21$ ,  $\epsilon_3 = 30$ ,  $w_1 = 8$  and  $w_3 = 2$ .

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