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LETTER TO THE EDITOR

Percolation threshold and mean grain size in Al_xSi_{1-x} thin films

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Abstract. We determined the critical composition x_c of the percolation threshold in polycrystalline Al_xSi_{1-x} thin films with a new method, using structural arguments only. The result $x_c = (0.55 \pm 0.05)$ agrees with the results of the commonly used methods. Moreover, our model explains the wide spread of experimental values of x_c as reported in the literature.

The critical composition x_c of metal insulator films at the percolation threshold marks the cross-over from the metallic to the granular behaviour [1]. Usually, such films are not amorphous but polycrystalline. In the case of polycrystalline samples, we assume that the percolation threshold will be approached, if each metallic grain is covered by a complete layer of the insulator. Knowing the solubility of the insulator in the metallic grains, the number of complete layers of insulator between the metallic grains (which is called the covering coefficient F) can be easily calculated; in the case of FCC structure, F is given by

$$F = \frac{(1 - x - S)}{x} \left(\frac{4\pi}{3}\right) D \left(\frac{r_R^2}{g_M^3}\right)$$

where the grains are assumed to be cubic.

Here, x is the concentration of the metal, S is the fraction of the insulator material which is dissolved in the metallic grains, r_I is the atomic radius of the insulator, g_M is the length of one elementary cell of the metal and D is the mean size of the crystals. Following the discussion given above, the critical compositions x_c will be reached when $F = 1$. Consequently, the critical composition depends on the solubility S and on the grain size D.

The grain size, however, depends on the insulator concentration, the material of the substrate and the conditions of the preparation of the sample. Thus, the critical concentration cannot be a well defined function of the fraction of the insulator. This result easily explains the wide spread of critical compositions ($0.12 < x_c < 0.6$, [1]–[5]) found for different materials or even for different preparation routines at the same material.

It is the purpose of this Letter to test the model given above, i.e. to compare the results of two standard methods with the predictions of our model, using Al_xSi_{1-x} films

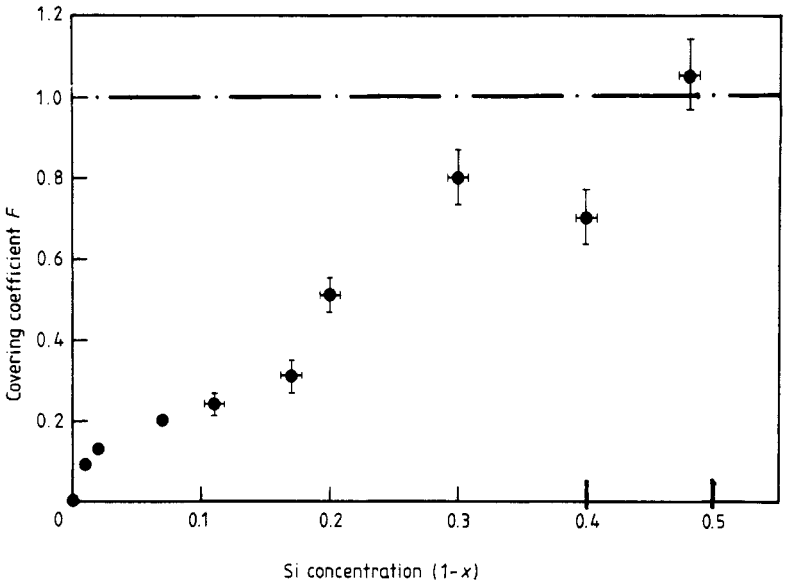

Figure 1. Covering coefficient F of the metallic grains versus insulator concentration. The critical region is the crossover from $F < 1$ to $F > 1$.

as an example. As evaluated from electron diffraction patterns, the solubility of Si in Al is known to be $S = 0.01$ [6]; the rest of the Si segregates into the grain boundaries, so that a homogeneous coverage of the grain boundaries can be expected. Moreover, the mean grain size decreases with increasing Si concentration [7], so that such films are very proper for this experiment.

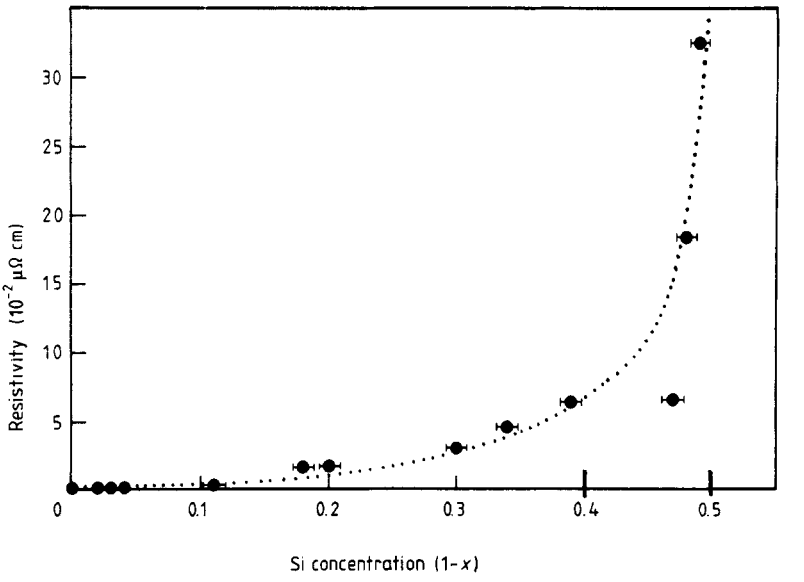

Figure 2. Resistivity of the samples of 30 nm thickness versus insulator concentration. A sharp change in the slope of the curve is observed between $0.4 < (1 - x) < 0.5$.

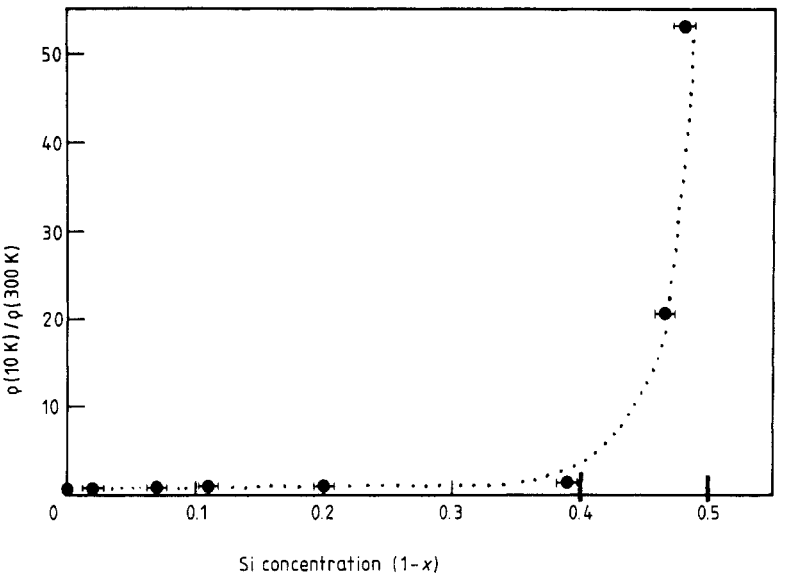

Figure 3. The value of $\rho(10\,\mathrm{K})/\rho(300\,\mathrm{K})$ versus insulator concentration. The percolation threshold is marked by a change in the slope in the range $0.4 < (1 - x) < 0.5$.

Usually, the percolation threshold is determined by considering the dependence of the resistivity ρ of the samples on the insulator concentration $(1 - x)$. A sudden change in the slope of this curve should mark the critical composition. A further method is the measurement of the temperature dependence of ρ. The ratio of $r = \rho(10\,\mathrm{K})/\rho(300\,\mathrm{K})$ should increase from the metallic and weak localisation regime $(0 < r < 2)$ to values above 10 when the percolation threshold is reached.

The results for the covering coefficient as defined above are shown in figure 1. The mean crystallite size was determined from TEM pictures with magnifications up to $(1:2) \times 10^5$. As can be seen from figure 1, the critical value ($F = 1$) is reached for

$$x_c = (0.55 \pm 0.05).$$

The results of the standard methods described in the text are presented in figures 2 and 3. Evaluating these plots, the critical value again is

$$x_c = (0.55 \pm 0.05).$$

Comparing the results, very good agreement is found between our model and the commonly used methods. Our model, however, explains why no universal critical value can be found in the case of polycrystalline films. Only in the case of purely 'amorphous' samples (if there really are such films: compare reference [8]) can a sharp and universal value be expected.

Using the example of Al_xSi_{1-x} thin films, we have shown that the percolation threshold in polycrystalline metal–insulator samples depends on both the solubility of the insulator in the metallic grains and the grain size. The critical value of the composition determined with the present model agrees with the result of the standard methods. Consequently, no universal value of the critical composition can be expected in the case of polycrystalline samples, so that the percolation threshold must be determined for each material and preparation routine.

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