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The effect of crystallinity on the morphology of evaporated Al-Ge thin films

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Résumé. — Des mélanges Al-Ge ont été préparés par évaporation simultanée des constituants sur des supports de verre chauffés à environ 180 °C. A cette température les deux constituants sont cristallisés. Les microographies prises au microscope électronique montrent une structure aléatoire. Les mesures de la résistivité donnent un seuil de percolation indiquant également une structure aléatoire. Ces résultats sont fort différents de ceux obtenus lorsque les mélanges sont évaporés à température ambiante, dans lesquels le Ge est amorphe et l'aluminium granulaire. Ils confirment la corrélation entre cristallinité et morphologie suggérée par Deutscher et al.

Abstract. — Al-Ge mixtures have been co-evaporated onto glass substrates heated to ~180 °C. At this temperature both the Al and the Ge were crystalline. Electron micrographs show a random structure. Resistivity measurements give a percolation threshold also indicating a random structure. These results are in contrast to the granular aluminium in an amorphous Ge matrix which is obtained on room temperature substrates, and support the correlation between crystallinity and morphology in evaporated films, as proposed by Deutscher et al.

It has been suggested by Deutscher et al. [1] that there exists a correlation between the crystallinity of an evaporated binary mixture of metal and insulator and its morphology. In reference [1] it was predicted that whenever both the metal and insulator are crystalline, the structure will be random. Consequently, the percolation threshold $X_c$ in three dimensions (3-D) will be about 15 vol. % [1]. However, when the metal is crystalline and the insulator is amorphous the structure will consist of metallic grains covered by insulator mantles. This is the usual granular structure which has a 3-D conduction threshold near 50 % [2]. Thus it was expected that the percolation threshold would be sensitive to the crystallinity of the mixture. In this letter we show results for Al-Ge which support this prediction.

Al-Ge co-evaporated onto room temperature substrates has a granular structure, with crystalline Al grains in an amorphous Ge matrix [1, 3]. Its percolation threshold is about 55 %.

In this work the Ge was induced to be crystalline by evaporating onto heated substrates. Two kinds of substrates were used. Both were microscope slides held at a temperature of ~180 °C. However, one was first covered by an evaporated NaCl film of about 2 000 Å thickness which was later dissolved in water to obtain films for electron microscopy.

The Al and the Ge were co-evaporated from two electron beam guns through a mask with slits to a combined thickness of about 1 200 Å. Each substrate contained nine samples with different metal concentrations due to different distances of the slits from the sources. The combined evaporation rate onto both substrates was ~40 Å/s and the pressure during evaporation was ~$5 \times 10^{-6}$ torr.

The microscopic structure of the films was investigated using a TEM. Typical structure is shown in figure 1. It is seen to be random. The diffraction pattern (Fig. 1) shows that the Ge is crystalline. From dark field micrographs the characteristic size of the crystallites (of both the Al and the Ge) was found to be about 250 Å.

Electrical measurements were made using the four terminal method. The results for the resistivity vs. metal concentration for the two types of substrate are shown in figure 2. As can be seen, the percolation threshold was the same for both substrates within the experimental error (25 % ± 4 %). In accordance with percolation theory it has been observed (see insert in Fig. 2) that near the percolation threshold the resistivity $\rho$ obeys the power law $\rho \sim (X - X_c)^{-t}$.

We have found $t = 0.9 \pm 0.2$ on NaCl and $t = 1.05 \pm 0.15$ on glass. Both values are consistent with the 2-D exponent predicted by theory [4], $t_{2D} = 1.1$, but certainly not with the 3-D value $t_{3D} = 1.7$.

Now, what seems like a contradiction arises; viz., the critical exponent $t$ has the 2-D value, while
the measured percolation threshold (25 %) is intermediate between the 2-D value (50 %) and the 3-D one (15 %). This is easily seen to be a dimensionality effect [5, 6] due to the fact that the ratio of the thickness \( d \) of the film to the grain size \( a \) is not a large number. Assuming that the critical concentration for a film of thickness \( d \) is given by [5-7]

\[
X_c(d) = X_c(\infty) + (A/n)^{1/v_3}
\]

where \( v_3 \) is the correlation length exponent in 3-D, \( A \) is a constant and \( n \sim d/a \) is the number of 2-D layers, and using \( X_c(\infty) = 0.15 \) and the values of Hoshen et al. [7] \( v_3 = 0.98 \) and \( A = 0.33 \) [5], we find for our films with \( d = 1 \, 200 \, \text{Å} \) that \( X_c(\infty) = 21 \% \). This is in reasonable agreement with the measured \( X_c(25 \% \pm 4 \%) \). We also note that in the concentration range 25 % \(< X < 35 \% \) the correlation lengths

\[
\xi_{2D} \approx 250 \, \text{Å} \, (X - 0.25)^{-4/3}
\]

and

\[
\xi_{3D} \approx 250 \, \text{Å} \, (X - 0.15)^{-0.85}
\]

are of the order of or larger than the films’ thickness.

The film is therefore, effectively 2-D in that range (see insert of Fig. 2).

In summary, we conclude that our Al-Ge films were indeed random, and that this was achieved because both the metal and the insulator were crystalline. We have shown, in addition, that the percolation threshold \( X(d) \) is larger than its 3-D value when the film thickness is only several times the grain size, and that 2-D critical behaviour is observed near \( X(d) \). This effect could be observed in the crystalline Al-Ge films precisely because they were random, in contrast to the much studied granular films.

References