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Critical behavior of the electrical resistance of very thin Cr films

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Measurements of the thickness dependence of the electrical resistance of two very thin Cr films deposited at substrate temperatures of 385 and 360°C and pressures of 10^{-7} and 10^{-6} Torr are reported. The various conduction mechanisms have been identified and a metal-insulator region spanning about 5.5 nm of thickness has been found. In this region the resistance of both films follows the scaling law $R \propto (d - d_c)^{-t}$ with $t = 1.34 \pm 0.11$ and d_c equal to 1.64 ± 0.20 and 1.03 ± 0.20 nm.

The earlier Monte Carlo calculations of the critical exponent associated with conduction in two-dimensional systems yielded a value of $t = 1.10 \pm 0.05$.¹ More recently Zabolitzky² calculated that $t/\nu = 0.973 \pm 0.005$ which, using the generally accepted value of the correlation length exponent $\nu = \frac{4}{3}$, gives $t = 1.297 \pm 0.005$. This value agreed remarkably well with values obtained by other researchers (see references in Ref. 2) using different techniques. Experimental confirmation of the value of t has so far involved model percolating systems only. Smith and Lobb³ found t to be approximately 1.3. By punching holes of various geometries in a slightly anisotropic two-dimensional conducting medium Mendelson and Karioris⁴ found t in the range 1.1 to 1.6. Recently Dubson and Garland⁵ determined t to be 1.29 ± 0.03 for site percolation on a square lattice and 1.34 ± 0.07 for random-void continuum percolation, proving that t depends only on the dimensionality of the system. Evidently a value of 1.3 is firmly established for ideal two-dimensional systems, i.e., the resistance of such a system is proportional to $(p - p_c)^{-1.3}$, where p is the probability of occupation of a conducting square on the insulating substrate.

Various conduction mechanisms exist in real two-dimensional systems. Near the onset of conduction at very low average film thicknesses, highly activated conduction mechanisms between isolated islands^{6,7} occur. As the islands start coalescing with increasing thickness, continuous metallic paths are formed and the system undergoes a metal-insulator transition. As the thickness increases further, the conductivity is influenced by grain boundary, surface, and roughness scattering. The purpose of the present report was to see if a critical exponent can be determined in the presence of other conducting mechanisms in very thin films. Measurements of the resistance of two Cr films, one in the thickness region $0 < d < 26$ nm and the other in $0 < d < 10$ nm, have been carried out under slightly different deposition conditions. For very thin films it is reasonable to assume that $p \propto d$

and, hence, the resistance should scale as $(d - d_c)^{-t}$. A direct experimental determination of the critical thickness d_c which marks the onset of the metal-insulator region is precluded by the highly activated conduction mechanisms at very low film thicknesses. It has nevertheless been found that d_c can be indirectly inferred from the measurements and that t can be determined with an accuracy of $\pm 8\%$ in Cr films.

A Varian vacuum system consisting of two vacsorb pumps, a titanium pump and a vacion pump was used to produce a pressure of $< 4 \times 10^{-7}$ Torr at all stages of the first set of measurements during which Cr was deposited on a Corning glass (No. 7059) substrate at a temperature of 385 K and a deposition rate of 0.1 nm s^{-1} . Prior to deposition, the substrate was chemically cleaned and subsequently maintained at this temperature in the bell jar until the pressure reached the above value. Simultaneously the Cr source of purity 99.99% was baked for a few hours at a temperature corresponding to a reddish color to degas it. The thickness of the film during the deposition was monitored by an Inficon crystal oscillator which registered the mass of Cr deposited on a target close to the Cr substrate and equidistant (~ 30 cm) from the Cr source. Since the substrate's resistance is very large ($> 10^{10} \Omega$), the constant current generator had to be activated by connecting an external resistor R_e across its output. This reduces the fractional output voltage across the film by a factor $(1 + R/R_e)^{-1}$ but allows the determination of the film resistance R as a function of the film thickness in our four-probe system, starting at zero thickness. The film dimensions were $l \times w \cong 3.9 \times 1.6 \text{ cm}^2$. The largest error in the measurements arose from the determination of the film thickness which was estimated to be ± 0.2 nm at all thicknesses. In what follows the film deposited at a substrate temperature of 385°C and a pressure of 10^{-7} Torr will be referred to as film 1. Film 2 was deposited at 360°C and 10^{-6} Torr.

The resistances of films 1 and 2 as a function of average film thickness in the region $0 < d < 10$ nm are shown in

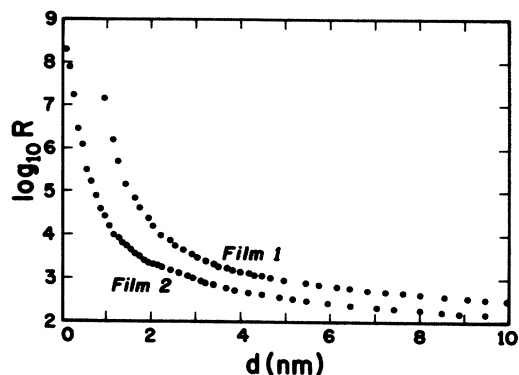


FIG. 1. The \log_{10} of the resistance R vs film thickness d of Cr films 1 and 2 deposited under different conditions. R is in Ω . Not all the experimental points are shown.

Fig. 1. Conduction in film 1 starts at a thickness of about 1 nm. Hereafter R decreases rapidly by a factor of 10^3 as the thickness changes from 1 to 3 nm. Conduction in film 2 appears at ~ 0.2 nm which points to the effect of the deposition parameters in determining the size of the islands in the two films. The initial rapid decrease in resistance below ~ 2 nm is believed to arise from activated conduction mechanisms which exist at thicknesses close to the onset of conduction and the formation of continuous metallic paths at the slightly higher critical thicknesses of 1.64 and 1.03 nm as found below. This is indicated in region A of Fig. 2 which shows the conductivity of the two films as a function of film thickness. The conductivity increases rapidly in region B and then much more slowly in region C.

In analyzing the results we first fitted the relation⁸ $Rd = K_0 + K_1/d + K_3/d^3$ to the measurements on film 1 in region C of Fig. 2 (32 experimental points) where the conductivity is apparently influenced by grain boundary and roughness scattering as described by the second and third terms of this equation. A least-squares fit in the

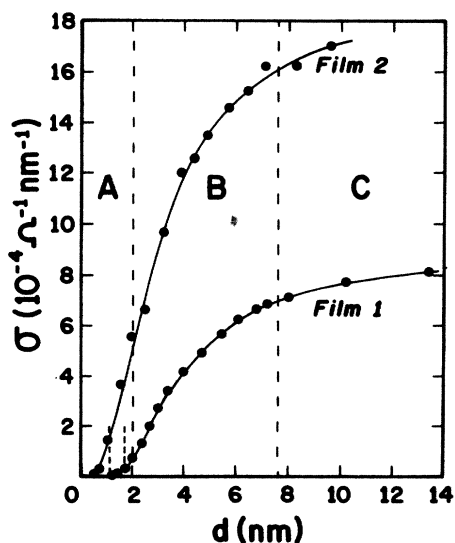


FIG. 2. The conductivity σ of Cr films 1 and 2 vs film thickness d . Not all the experimental points are shown. Regions A, B, and C are referred to in the text. The vertical dashed lines below 2 nm indicate the critical thickness of each film.

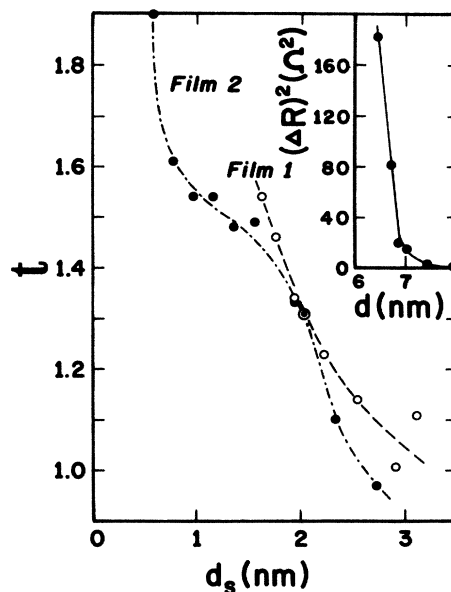


FIG. 3. The critical exponent t vs the starting thickness d_s . The inset shows $(\Delta R)^2 = (R - R_{\text{fit}})^2$ vs thickness d of film 1.

thickness region $8 \leq d \leq 13.5$ nm gave $K_0 = 2.85 \times 10^3 \Omega \text{ nm}$, $K_1 = 7.156 \times 10^2 \Omega \text{ nm}^2$, and $K_3 = 2.359 \times 10^5 \Omega \text{ nm}^4$. A plot of $\Delta R^2 = (R - R_{\text{fit}})^2$ as a function of thickness from 6 to 8 nm is shown in the inset of Fig. 3. (R changes by only 20% in this region.) From ~ 7.6 to 13.5 nm $\Delta R^2 < 2.5 \Omega^2$ which indicates that the above relation gives a very good description of the data in the fitted region. Although the fitted points were confined to the upper limit of 13.5 nm ΔR^2 seldom exceeded $2.5 \Omega^2$ for thicknesses up to 26 nm. As can be seen from the inset in Fig. 3, different conduction mechanisms start affecting the conductivity below 7.4 ± 0.2 nm which we will use in the subsequent paragraph to set a lower limit on the value of the critical exponent. A fit of the above relation in the region $2 < d < 8$ nm produced $K_1 < 0$ which is physically unacceptable. If only the first two terms are fitted, i.e., with $K_3 = 0$, a very bad fit is obtained.

A least-squares fit of $R \propto (d - d_c)^{-t}$ to the data on film 1 in region B of Fig. 2 using d_c as a variable revealed that $1.3 < d_c < 2.6$ nm, the experimental points displaying a distinct concave upward and downward feature near the beginning of the fits for d_c equal to 1.3 and 2.6 nm, respectively. A similar analysis pointed to the existence of a critical thickness in film 2. To set an upper limit on t which is complicated by activated conduction mechanisms, the following procedure was carried out. Sliding least-square fits were performed on sets of data each spanning 5.5 nm of thickness for each film, starting at thicknesses in the range $0.5 < d_s < 3.5$ nm (e.g., with $d_s = 2$ nm, one fit would cover the range $2 < d \leq 7.5$ nm, large enough to cover the metal-insulator region and to justify a meaningful statistical analysis). The results of these calculations are shown in Fig. 3. For each film t decreases with increasing d_s . From the variation of χ^2 with t , as shown in Fig. 4, the upper limit of the critical exponent is taken to occur at $t = 1.45$ as indicated by the vertical dashed line in this figure. The rapid increase in

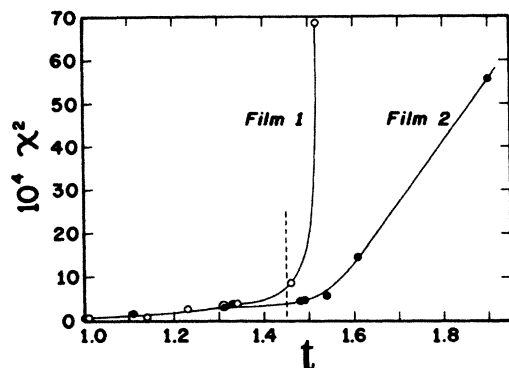


FIG. 4. χ^2 vs the critical exponent t for Cr films 1 and 2. The vertical dashed line marks the upper limit of t .

χ^2 above this value is attributed to the effects of the activated conduction mechanisms. As for the determination of the lower limit, application of the scaling law becomes physically meaningless for values of d exceeding 7.6 nm as found in the previous paragraph. This implies $d_s = 2.1$ nm and $t = 1.23$ with the aid of Fig. 3. Hence, $t = 1.34 \pm 0.11$. The critical thicknesses corresponding to this value of t are 1.64 ± 0.20 and 1.03 ± 0.20 nm for films 1 and 2, respectively. Figure 5 shows the resistance versus $d - d_c$ for the two films in the region where the conductivity is dominated by the formation of continuous metallic paths. The uncertainty in t amounts to only ± 0.005 (two standard deviations) for these fits, the much larger error of ± 0.11 reflecting the overlapping effects of the other conduction mechanisms into the metal-insulator region.

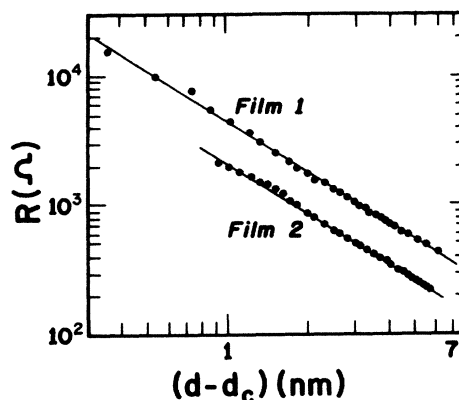


FIG. 5. The resistance R of Cr films 1 and 2 vs $d - d_c$ in the metal-insulator region. The straight lines have a slope of -1.31 and $d_c = 1.68$ and 1.11 nm for the two films, respectively.

We, therefore, conclude that the percolating region in Cr films can be reasonably well identified and described by the scaling law $R \propto (d - d_c)^{-t}$ with $t = 1.34 \pm 0.11$. The conductivity of film 1 in the thickness region $8 < d < 26$ nm can be very well accounted for using the relation $Rd = K_0 + K_1/d + K_3/d^3$. Further work on this aspect of Cr films is being carried out.

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