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Simulation of structural transitions in thin films

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Thin-film growth was simulated by the deposition of two-dimensional discs on to a smooth substrate. The films were characterized for structure, orientation and density. The incorporation of increased adatom mobility during film growth resulted in a transition from a highly defected and voided columnar deposit to a dense, more crystalline columnar deposit. The simulation results duplicate the general features of the Zone 1 to Zone 2 structural transition in physical vapour-deposited thin films, and confirm the importance of adatom surface mobility in producing this transition.

1. Introduction

The microstructure of physical vapour-deposited thin films is strongly dependent on deposition conditions, particularly substrate temperature. Movchan and Demchishin [1] proposed a structure zone model (SZM) or classification of films into three zones according to the ratio of substrate temperature during deposition to melting temperature of the film, T/T_M. The low temperature, low adatom mobility Zone 1 structure at $T/T_M < 0.3$ consists of highly defected columns separated by voided boundaries. The Zone 2 structure, $0.3 < T/T_M < 0.5$, consists of dense columnar grains with dislocations and defects primarily in boundary regions. In the high-temperature Zone 3 structure, $T/T_M > 0.5$, where bulk diffusion processes are active, the structure consists of equiaxed grains with a smooth surface. Subsequently, Thornton [2] identified a Zone T structure, intermediate to Zones 1 and 2, consisting of a dense array of poorly defined fibrous grains. The Zone 1 to Zone T transition results from energetic particle bombardment of the growing film surface. Of interest in this paper is the Zone 1 to Zone 2 transition, believed to be due to an increase in adatom surface diffusion with increasing temperature. Evidence for the importance of adatom diffusion is given by the grain-width dependence on T/T_M for Zone 2 structures, which yields an activation energy approximately equal to that for surface diffusion [3].

Computer simulations of thin-film deposition have helped to clarify the processes involved in microstructure formation. In 1974, Henderson *et al.* [4] reported the random deposition of three-dimensional hard spheres on to a surface, with subsequent relaxation to the nearest pocket formed by three spheres. Due to the effects of clustering and shadowing, these simple growths reproduced the general features of low-mobility Zone 1 films: columnar structures with an orientation in agreement with the experimentally observed tangent rule [5]. Subsequent simulation work has been reviewed by Leamy *et al.* [6] and the fractal properties of these ballistic deposits have been studied by Meakin [7]. Although only small numbers (relative to a real film) of particles are deposited in such simulations, statistical analyses of these deposits show many properties, such as surface roughness and density variation with deposition angle, that agree with the characteristics of real films.

The effects of increased adatom mobility have been studied by Kim *et al.* [8] who allowed the incident particles to bounce several times before they were incorporated into the deposit. This artificial motion created deposits of higher density that still retained a columnar structure. Transitions to denser, more crystalline deposits with increased temperature have also been seen in the more complex molecular dynamics simulations by Leamy *et al.* [6] using a Lennard–Jones potential to model atomic interactions, and by Müller [9], who introduced thermally activated hopping within the deposit. The purpose of this work is to utilize computer simulation to provide a clear confirmation of the importance of adatom mobility in the Zone 1 to Zone 2 transition, by explicitly incorporating increased adatom surface mobility with a simple ballistic deposition model for two-dimensional hard discs.

2. Computer simulation

Thin-film growth was simulated by the random deposition of two-dimensional hard discs (atoms) on to a smooth substrate at an angle of incidence α with the substrate normal, as shown in Fig. 1. To determine an atom trajectory from the top of the simulation boundary, an x coordinate was generated using a pseudo-random number generator on the interval $(0, L)$. The first point of contact (impact position) with previously deposited atoms was calculated using the linear (ballistic) atom trajectory. If an atom hit the substrate, it remained at that point. An atom incident on the film was allowed to relax to a "cradle" position between other atoms on the film surface according to the surface mobility scheme used.

To simulate "low" adatom mobility, the incident atom was relaxed to the nearest cradle point at which it touched at least two other atoms. Deposits using

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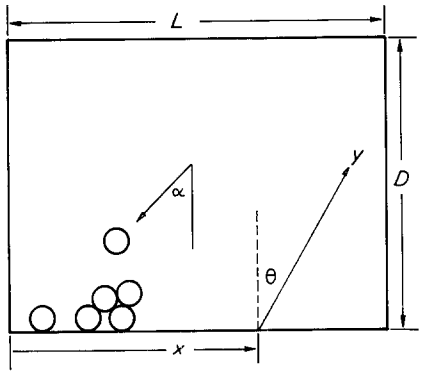

Figure 1 An illustration of deposition at angle of incidence α and also of the (x, y) coordinate system used for the analysis of column orientation.

this low-mobility relaxation scheme have been studied in detail by other authors [10]. To introduce increased or "medium" adatom surface mobility, the incident atom was relaxed to the surface cradle with the highest coordination number (greatest number of bonds to other atoms) within a radius of three atomic diameters of the initial impact position. In the case of cradle positions with equal coordination number, the final site chosen was closest to the impact position. This simple relaxation process was chosen as it is analogous to the surface diffusion of a real adatom to a site of lowest potential energy. For further increased adatom mobility, "high" mobility simulations followed an algorithm identical to the medium mobility simulations, but with the final cradle site a maximum of four atomic diameters distant from the impact position. A sticking coefficient of unity was used for all simulations.

Simulations were performed on a film segment 85 atoms wide, and were terminated when the thickness reached 70 atomic diameters. Dependent on the resultant film density, a typical simulation involved the deposition of 3200 to 4600 atoms. Periodic boundary conditions were imposed along the x coordinate in order to reduce finite size effects in the simulations. The computer code was written in Fortran 77 and ran on a Zenith 158 microcomputer with execution times of approximately 4, 30 and 160 min for simulations with low, medium and high mobility, respectively.

3. Results and discussion

Simulations were performed at various angles of incidence between 0 and 65° and for low, medium and high adatom mobility. The results discussed in detail here are for $\alpha = 50°$, which show structural trends with mobility that are typical for simulations at any angle of incidence studied. Films L, M, and H, deposited at $\alpha = 50°$ and at low, medium, and high mobility, respectively, are shown in Fig. 2. The average film density for these simulations is shown in Table I, and was calculated relative to a hexagonal closest-packed structure and measured between the 20% and 80% points of the film thickness so as to avoid substrate and surface effects. Also shown in Table I are the average coordination numbers for the first and second nearest neighbours of the film particles in neighbour positions identical to those for a hexagonal closest-packed arrangement of discs.

The trend in Films L, M, and H with increasing mobility shows three features similar to those of

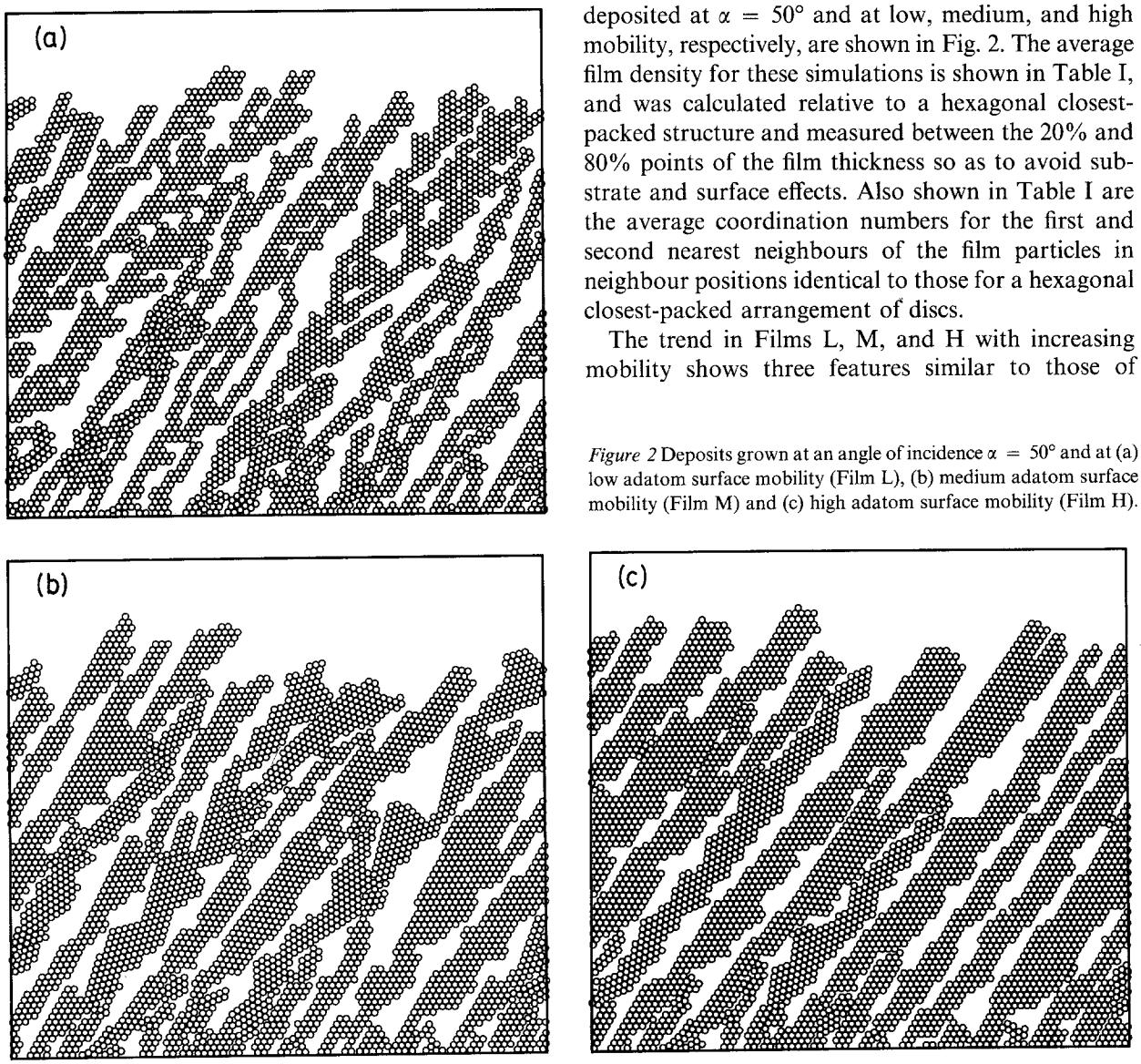

Figure 2 Deposits grown at an angle of incidence $\alpha = 50°$ and at (a) low adatom surface mobility (Film L), (b) medium adatom surface mobility (Film M) and (c) high adatom surface mobility (Film H).

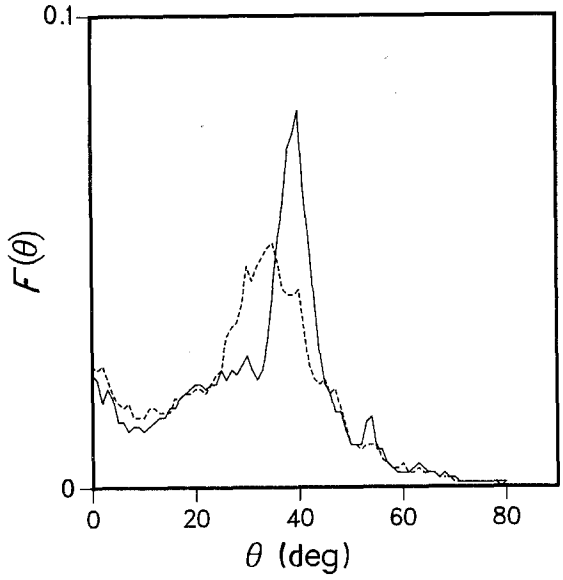

Figure 3 The orientation function $F(\theta)$ for (– – –) Film L (low adatom mobility) and (——) Film H (High adatom mobility).

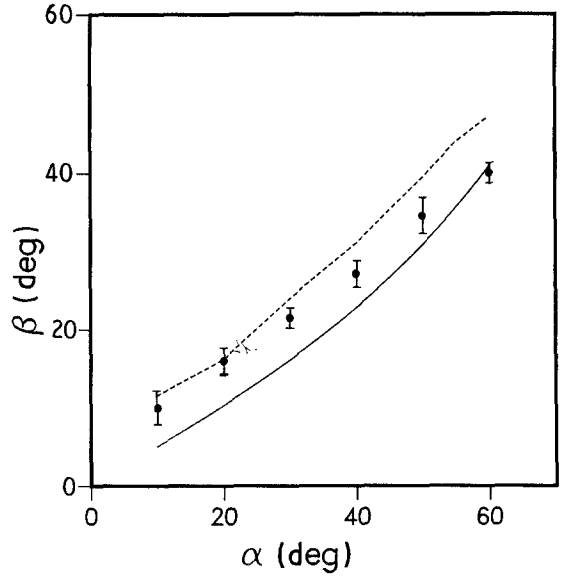

Figure 4 Column orientation β as a function of deposition angle of incidence α: (– – –) Meakin *et al.* [12], (——) tangent rule, (●) data from high-mobility simulations. Error bars indicate the standard deviation from the mean of up to five simulations.

a Zone 1 to Zone 2 transition in real films. First, a substantial density increase is observed, from $\varrho(\mathrm{L}) = 0.61$ to $\varrho(\mathrm{H}) = 0.75$. Second, a decrease in dislocations and voids inside the columns and an increase in average coordination number is seen, similar to the improved crystallinity of columnar grains in a real Zone 2 film. Finally, the simulated columns of Film H maintain a dense particle packing orientation throughout the column, analogous to the growth of column crystallites in a localized epitaxial process for Zone 2 coatings [11]. Film H retains the shadowing-induced columnar structure in spite of high surface adatom mobility. In real coatings, a columnar morphology is eradicated only at deposition temperatures high enough to create atomic mobility in the bulk of the film, and leading to a structural transition to Zone 3.

A statistical analysis of the films was performed using the method of Sikkens *et al.* [10] to determine the average angle β of the columns with the substrate normal. A column orientation function $F(\theta)$ was defined as

$$F(\theta) = \frac{\cos^2\theta}{\varrho(1-\varrho)\,D^2}\int_0^L \left\{\int_0^{D/\cos\theta} [\varrho(x, y) - \varrho]\,\mathrm{d}y\right\}^2 \mathrm{d}x \quad (1)$$

using the coordinate system shown in Fig. 1. ϱ is the average density of the film and $\varrho(x, y)$ is the point density of the film, either 0 or 1 dependent on the presence of an atom at that coordinate. Thus a film with columns strongly oriented at an angle β will show a maximum in the orientation function $F(\theta)$ at $\theta = \beta$. Plots of $F(\theta)$ in Fig. 3 for Films L and H show preferred orientations of $\beta(\mathrm{L}) = 36°$ and $\beta(\mathrm{H}) = 40°$, respectively, with the increased orientation strength of Film H due to the improved crystallinity of the deposit.

TABLE I Densities and average coordination numbers for a hexagonal closest-packed (h c p) arrangement of discs and for Films L, M, and H

	h c p	Film L	Film M	Film H
Density	1.0	0.61	0.70	0.75
Coordination number				
1st nearest neighbour (1.00 diameters)	6.0	4.51	4.89	5.14
2nd nearest neighbour (1.725 diameters)	6.0	3.48	3.83	4.47

The column orientation in real coatings follows an empirical relationship called the tangent rule [5]:

$$2\tan\beta = \tan\alpha \quad (2)$$

where α is the deposition angle and β is the angle of orientation of the columns, measured from the substrate normal. This relationship has been verified for deposition angles up to 80°. Extensive ballistic deposition simulations by Meakin *et al.* [12], similar to the low-mobility simulations reported here, showed a deviation from the tangent rule. To determine the behaviour of the column orientation β for deposition at high adatom mobility, a series of simulations were performed at angles of incidence from 10 to 60°. These simulation results are shown in Fig. 4, along with the tangent-rule prediction and the results of Meakin *et al.* [12]. The orientation for high adatom mobility (Zone 2) growths is approximately intermediate to the other results. At the present time, the reason for this small deviation in column orientation between computer-simulated growths and real growths is not understood.

For real thin films, it has been observed that film density decreases almost linearly with $\tan\alpha$ [6, 13]. This relationship is due to the longer shadows cast at larger angles of incidence α, leading to wider voids between columns. Fig. 5 shows the density variation for simulated high-mobility films as a function of $\tan\alpha$ for $\alpha = 10$ to 65°, and a linear fit to the data. The good fit demonstrates that the simulated film densities have a behaviour, with $\tan\alpha$, similar to real films.

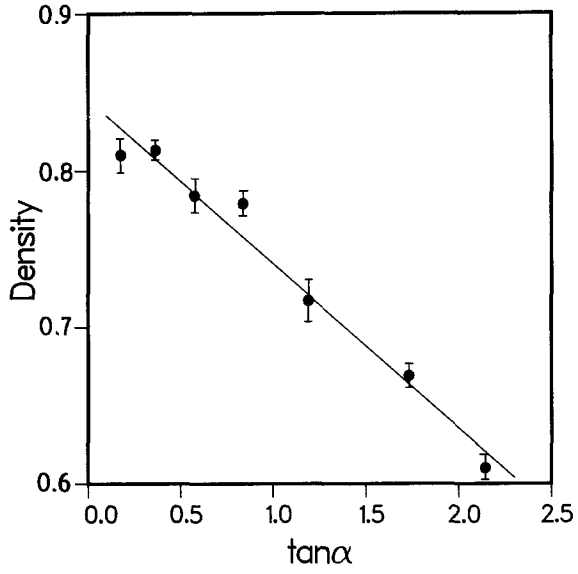

Figure 5 Deposit density of high-mobility simulations as a function of the tangent of the deposition angle of incidence. The straight line is a least-squares fit to the data. Error bars indicate the standard deviation from the mean of up to five simulations.

4. Conclusions

Computer simulations of thin-film deposition were performed by irreversible ballistic deposition of two-dimensional discs. Adatom surface mobility was included by a simple process in which adatoms relaxed to the surface bond site of highest coordination number within a radius of up to four disc diameters from the impact position. Deposits without adatom mobility had a highly dislocated and voided columnar structure typical of Zone 1 coatings. Deposits including adatom mobility had structural features similar to those of Zone 2 coatings; increased density, fewer dislocations, and a consistent particle packing arrangement throughout each column. Deposit density and orientation varied with deposition angle in a manner similar to real films. These simulation experiments have provided a confirmation of the importance of adatom surface mobility in creating a Zone 1 to Zone 2 thin-film structural transition.

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