JOURNAL OF MATERIALS SCIENCE 31 (1996) 685–693

A three-dimensional non-epitaxial atomistic growth model for thin-film deposition: effect of surface mobility

SHI XU, G. Q. LU*
*School of Electrical and Electronic Engineering, and *School of Mechanical and Production Engineering, Nanyang Technological University, Nanyang Avenue, Singapore 2263*

The effect of the atomic mobility on a film surface has been studied by using a three-dimensional atomistic thin-film deposition model which simulates three-dimensional thin-film images, surface profiles and cross-sectional area pictures. In addition, quantitative results of surface RMS roughness, average film thickness, atomic coordination number and its distribution, and solid fraction of the deposited thin films, were obtained from the simulations. When the film surface mobility increased from 0.3 to 3.0, RMS roughness decreased from 6.5 to 1.1, solid fraction increased from 0.27 to 0.56 and average film thickness decreased from 40 to 28, due to the reduction of the voids within the film. The full-width half magnitude of the atomic coordination distribution became narrower indicating the increased degree of crystallization. With increase in surface mobility crossing the boundary to 1.5, the film evolved from a porous or loose columnar structure with voids, to a densely packed fibrous grain structure which can be categorized by the zone structure models.

1. Introduction

Thin films have been widely used for electronic, magnetic, optical, protective and tribological applications. The deposition processes are usually far from thermodynamic equilibrium and the structure and properties of thin films largely depend on the deposition techniques and conditions. Movchan and Demchishin [1] originated the structure zone model (SZM) to categorize films produced by physical vapour deposition (PVD) processes. Their observations were based on the electron-beam evaporated films under optical microscopy of cross-sections. When the substrate temperature is low ($T/T_m < 0.3$, where T_m is the melting temperature of the film material), little surface mobility exists, resulting in tapered columnar structure with domed tops on columns. Voids are present between columns and the surface is rough. Films in this form are classified as Zone 1. Zone 2 is categorized by the columnar structure without voids, and a smoother film surface morphology than in Zone 1, due to increased surface mobility when T/T_m is between 0.3 and 0.5. When T/T_m is between 0.5 and 1.0, bulk self-diffusion occurs, resulting in equiaxed grains and an even smoother surface than in Zone 2.

Thornton extended the model into sputtering processes [2] by observing fractured films in the scanning electron microscope. In Thornton's SZM, Zone T is defined as a transition between Zone 1 and Zone 2. Zone T is a region consisting of "a dense array of poorly defined fibrous grains without voided boundaries".

Computer simulation of thin-film growth is of great significance and has attracted the attention of many researchers in recent years. It can be used to replace the analytical solution of theoretical equations to provide a better understanding of and new insights into the growth mechanisms. Many important parameters and effects can be easily investigated by simulation, while they may prove to be experimentally difficult or expensive.

Most of the simulation models can be categorized into either analytical or atomistic models [3–10]. The atomistic models normally result in simulated two-dimensional film microstructure and morphology. However, the two-dimensional atomistic simulations [5–9] over-simplified the real atomistic film deposition. Some three-dimensional atomistic deposition models have been reported [10], generating various film solid fractions ranging from 0.128–0.582. However, data are rare and some structural readjustments in the models are artificial. A good review of the recent literature on computer models for thin-film deposition is available [11].

In our recent letter [12], we reported a three-dimensional atomistic model to simulate the effect of the atomic substrate surface mobility on the nucleation and island growth of thin films. In this paper the effect of the film surface atomic mobility on film morphology and structure is presented.

0022–2461 © *1996 Chapman & Hall*

2. Model development and simulation technique

2.1. Model development

The physical concept of the model is described as follows.

1. The substrate is flat, defect-free with dimensions 70 × 70 atom diameters.

2. A single atom impinges from a random position far above the substrate and down to the substrate at a normal incidence angle at constant time intervals.

3. A single atom migrates on the substrate surface (substrate surface mobility, γ_1) to a random direction at a constant jumping step, γ_1, before coalescence with other atoms or settling down on the substrate.

4. Atoms deposited on adatoms are subjected to a variable film surface mobility, γ_2, i.e. an atom moves randomly within a circular area with radius of γ_2 until a settling position with smallest height is found.

In the present model, the re-evaporation of atoms on the substrate and the effect of different substrates are not considered. The basic settling criterion for an atom on the substrate is one atom being in contact with three other closest adatoms [13]. The length unit used in the model and in this paper is the atom diameter length.

2.2. Simulation techniques

The model is programmed in FORTRAN and is summarized in a flowchart as shown in Fig. 1. The program begins by generating two random x and y coordinates for an atom. The z coordinate is initially fixed at a position (1000) far above the substrate. The atom then moves straight down to the substrate. CHECK routine then checks whether there is any adatom in the way. If there is one adatom in the way, the incoming atom will stick to it by routine STICK1. If there are two adatoms in the way, routine STICK2 is called and the coming atom will settle down by contacting both adatoms. If there are more than two adatoms, the incoming atom will settle down by making three contacts with three adatoms using routines CNTAC1, CNTAC2 and CNTAC3.

If there are no other adatoms in the way, the incoming atom will be placed on the substrate and migrate to a random direction at a given step. During the migration, if there is an adatom in the way, the incoming atom will stick to it and stop there by routine STICK1. If there are two adatoms, the incoming atom will contact both adatoms and stop by routine STICK2. If there are more than two adatoms, the coming atom will then make three contacts by routines CNTAC1, CNTAC2 and CNTAC3 and settle down. If this atom sits on other adatoms, it will move around within a given film surface mobility range to finalize its position in a lowest place to the substrate. Finally the atom's three coordinates and relations between this atom and other adatoms are recorded. The whole process is repeated until the total number of atoms deposited reaches a preset number (120 000) atoms in this work). The detailed descriptions of CNTAC1, CNTAC2 and CNTAC3 can be found elsewhere [13].

The computing time of the program is increased exponentially with the number of atoms because all the adatoms will be manoeuvered together (coordinate transformation and overlap checking) when an atom is being deposited. In order to reduce the computing time, only those adatoms which might be used in the process are made active. The computing time is approximately linear with the number of atoms. When the program runs on PC-486DX2 50 MHz it takes between 20 and 40 h to deposit 120 000 atoms, depending upon the mobility settings. The results presented in this paper are from the simulations run on Sun SparcII workstation and the computing time is typically within 24 h for 120 000 atoms.

The surface RMS roughness, average film thickness, coordination number and solid fraction of the film are obtained by running an analysis program called ANALYSE. Three-dimensional and cross-sectional area pictures are generated by a plotting program also written in FORTRAN. Three-dimensional surface profiles are plotted by graphics package AXUM.

3. Results and discussion

Parameters of the eleven simulations conducted are given in Table I. The atomic mobility on the film surface is considered as a nearest-neighbour hopping process [14]. The mobility is proportional to the hopping rate of an Arrhenius form and can be expressed as $\gamma \propto \exp[-E_B/(kT + E_{in})]$, where γ is the mobility, k is Boltzmann's constant, E_B is the atomic hopping barrier, T is the substrate temperature, and E_{in} is the atom incoming energy. The quantitative relation of the surface mobility to those physical parameters may be found by validating the results of simulation with experimental data. In this work, however, the mobility is regarded as a lumped simulation parameter taking into account the effect of the substrate temperature, film atomic hopping barrier, and energy of the incoming atoms. Our previous work revealed that the atomic mobility on a substrate only affects the initial nucleation and coalescence stage and has minimum effect on film morphology and structure when the film becomes continuous [14]. In this work, the atomic mobility on a substrate is fixed at 5.0, which is a reasonable value for metal on glass at room temperature where E_B is relatively small.

It is well known that, for the evaporation process, the energy of the incoming atoms is quite low (about 0.1 eV). In order to obtain a film with a smoother surface and a higher density, an elevated substrate temperature is necessary to result in higher atom mobilities on the substrate and the film surface. At the same temperature, films made by the sputtering process normally have smoother surface profile and higher density compared with those made by the evaporation process. This is because the energy of the incoming atoms in the sputtering process is higher than that of those in the evaporation process to generate higher mobilities, and hence better film quality.

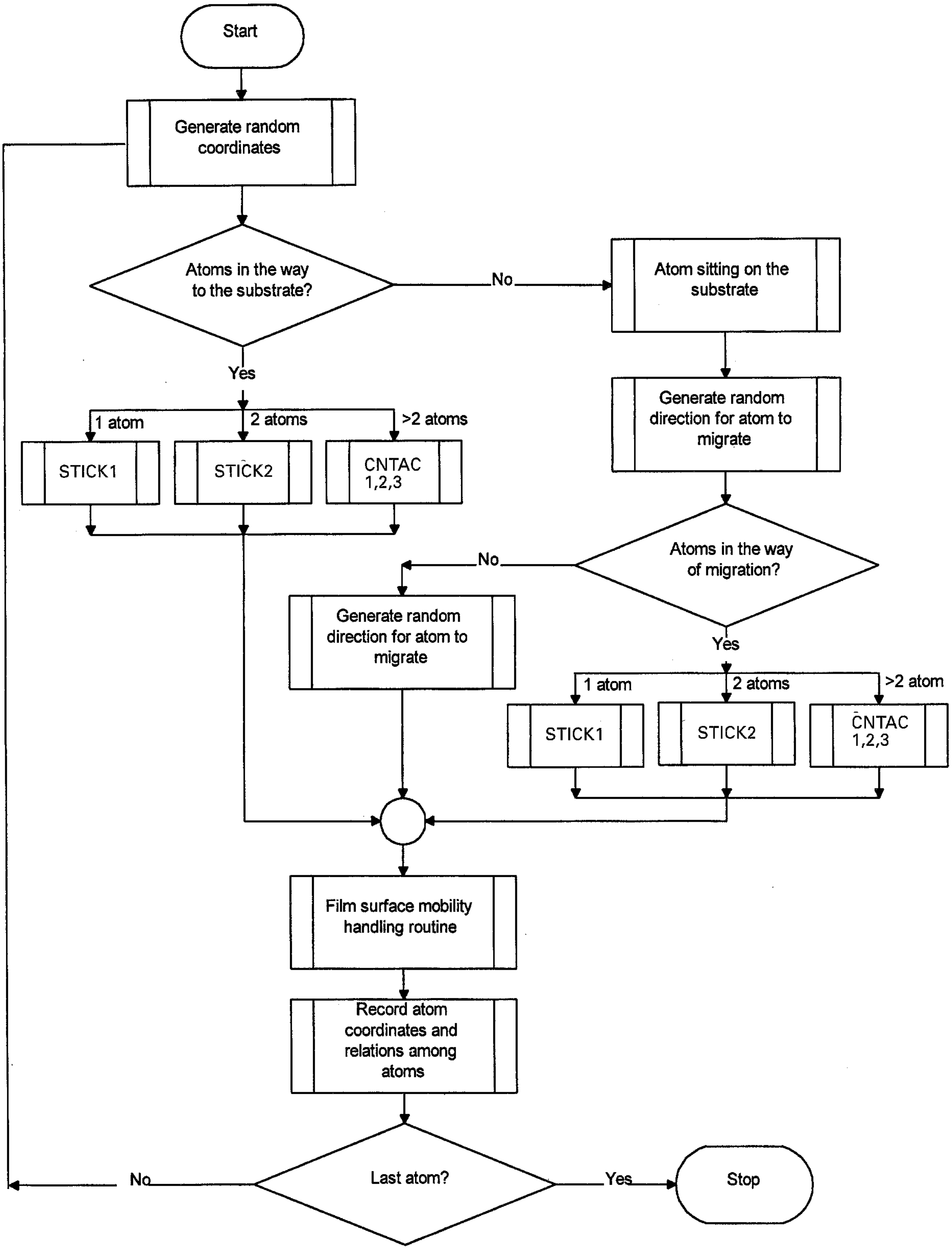


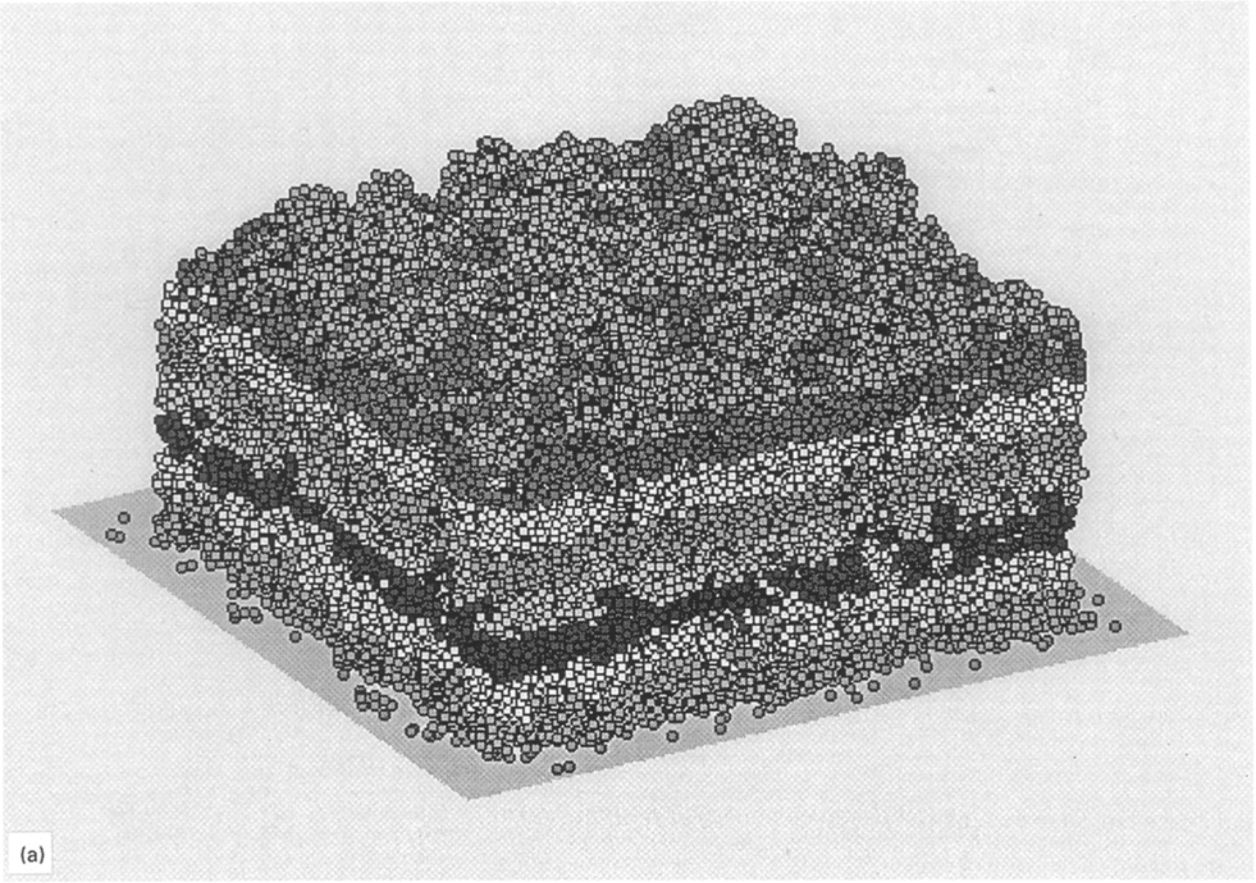
Figure 1 The flowchart of the simulation program.

TABLE I Simulation parameters

S/N	Deposition area (atom diameters)	Number of atoms deposited	Film surface mobility (atom diameters)
1	70 × 70	120 000	0.3
2	70 × 70	120 000	0.5
3	70 × 70	120 000	0.7
4	70 × 70	120 000	1.0
5	70 × 70	120 000	1.5
6	70 × 70	120 000	2.0
7	70 × 70	120 000	2.5
8	70 × 70	120 000	3.0

3.1. Effect of film surface mobility on film morphology

The film surface morphology is profiled by dividing the film surface into 1 × 1 grids to obtain the height of the film column within every other grid. In the same process, the values of average film thickness and the average RMS roughness of the film surface are obtained. Fig. 2 shows three-dimensional pictures of the simulated thin film nos. 2 and 6 with surface mobility 0.5 and 2.0, respectively. Fig. 3 shows two series of snap shots of these two films during their growing process. When there are only 3000 atoms, the island



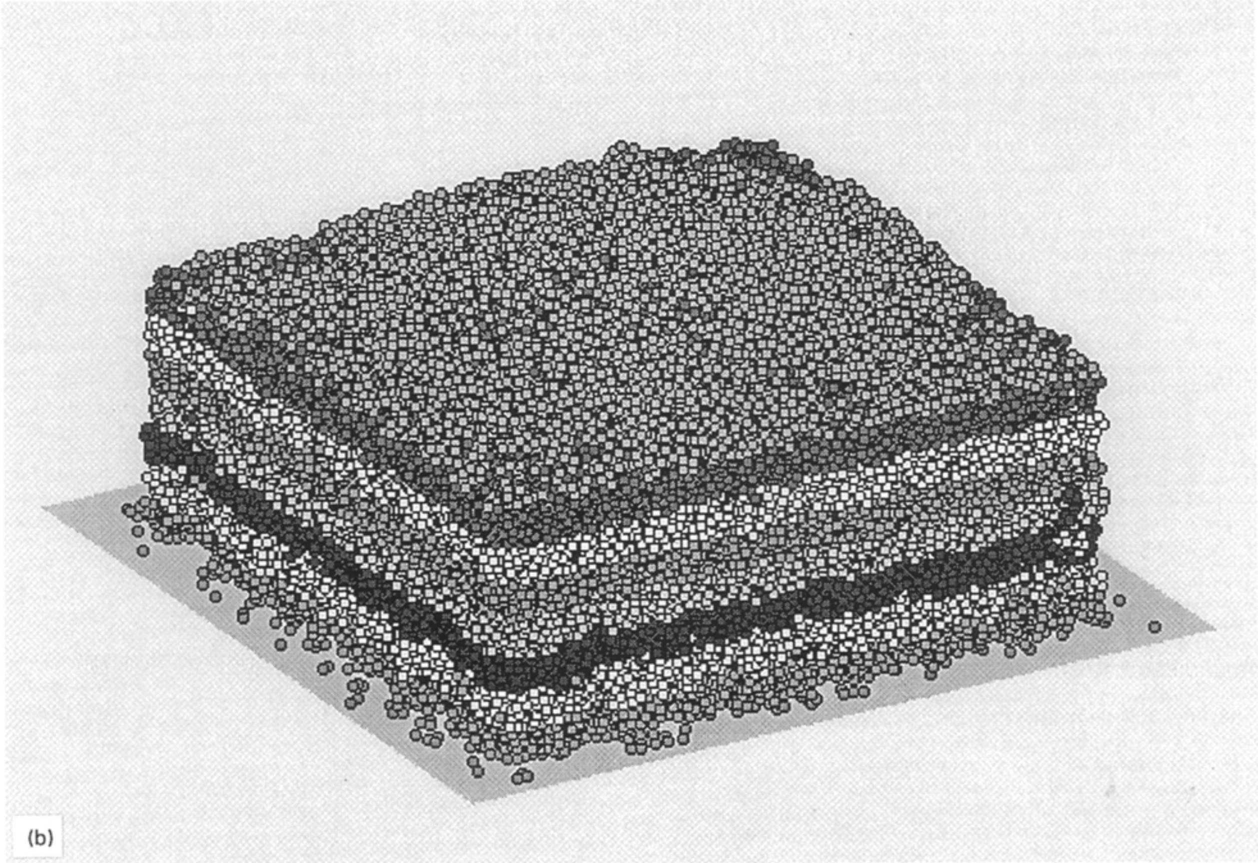


Figure 2 Three-dimensional pictures of the simulated thin films with film surface mobilities of (a) 0.5, (b) 2.0. Deposition area 70 × 70 and 120 000 atoms. Substrate mobility is fixed at 5.0.

density and size are almost the same for these two films. However, the two films grow into different morphology when the films become continuous. The film of mobility 0.5 has an obviously rougher surface (RMS roughness ~ 3.7), while the surface of the other film of mobility 2.0 is much smoother (RMS roughness ~ 1.2). The similar starting morphology results from the same atomic mobility on the substrate surface. At

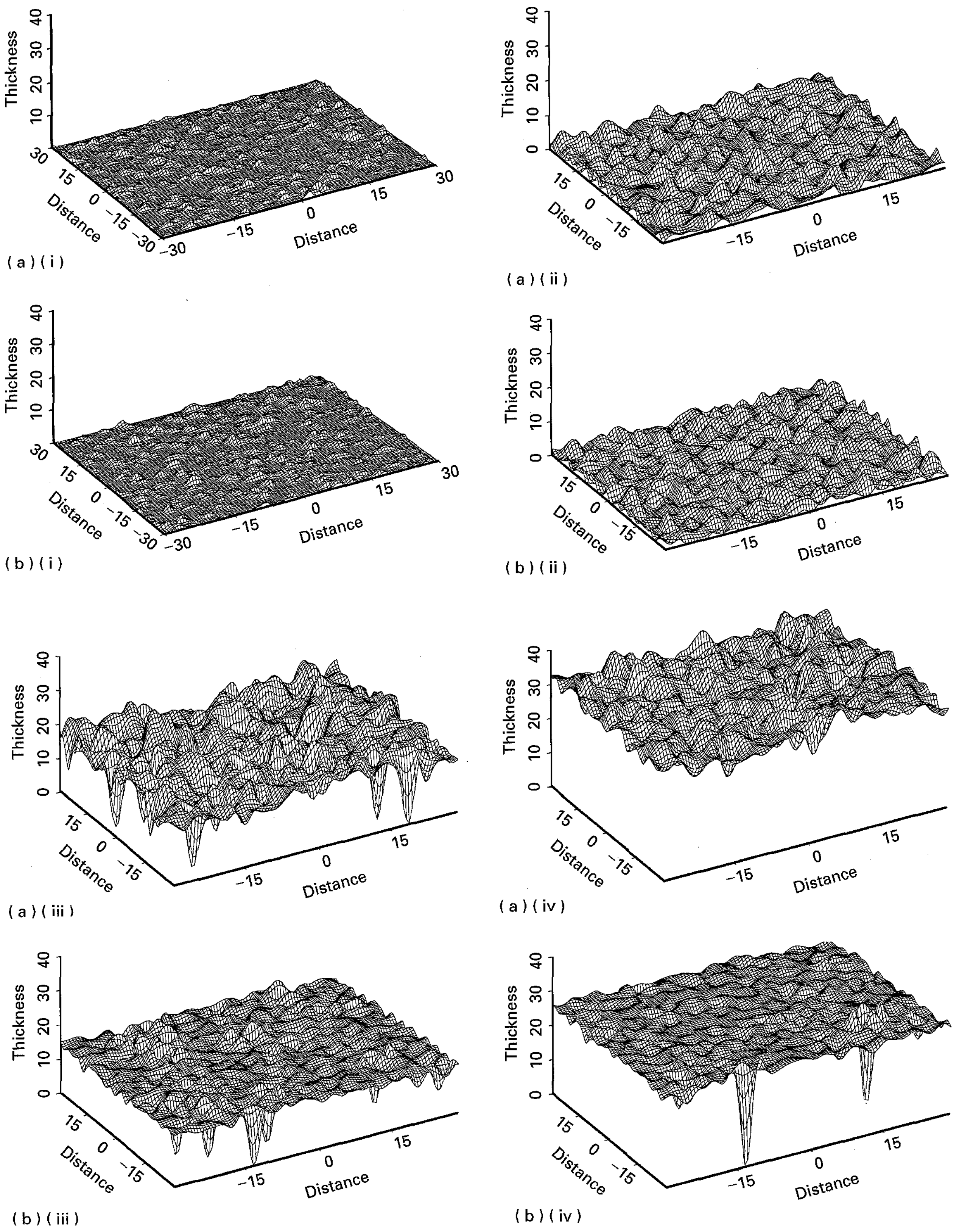


Figure 3 Snap-shots of the films with film surface mobilities of (a) 0.5, (b) 2.0. (i) 3000 atoms; (ii) 10 000 atoms; (iii) 60 000 atoms; and (iv) 110 000 atoms.

this stage the substrate surface mobility is a predominant parameter. After the film becomes continuous the film surface mobility becomes the dominant parameter. Films grow into different surface morphology because of their different film surface mobilities.

The quantitative plot of RMS roughness of the film surface and film average thickness versus the film surface mobility is given in Fig. 4. It is seen that the film surface roughness reduces rapidly from 6.5 to 1.2 and film thickness from 40 to 28 when film surface mobility increases from 0.3 to 1.5. Afterwards, they appear to reduce at a much slower pace. Roughness changes from 1.2 to 1.1 and thickness from 28.2 to 27.8 when mobility increases from 1.5 to 3.0.

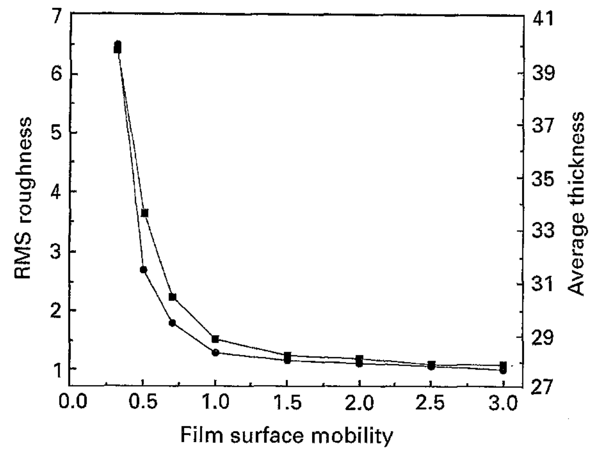


Figure 4 The film surface RMS roughness (●) and average thickness (■) versus the film surface mobility.

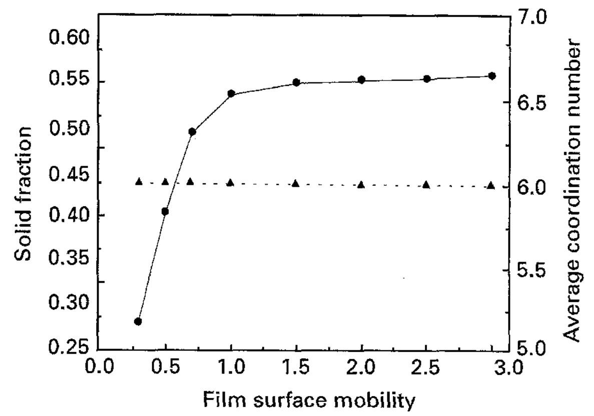


Figure 5 The (●) solid fraction and (▲) average atomic coordination number versus the film surface mobility.

3.2. Effect of film surface mobility on film structure

Fig. 5 shows the change of the solid fraction and the average atomic coordination number of the film with the film surface mobility. The solid fraction shows a rapid increase from 0.26 to 0.54 when mobility increases from 0.3 to 1.5, thereby resulting in a denser film. Considering that the solid fractions for f'c'c and b'c'c structures are 0.74 and 0.68, respectively, and distance between nearest atoms in a solid is about 20% less than the sum of two atomic radii, 0.26 solid fraction means about 42% and 45% of the bulk material density for f'c'c and b'c'c structures, respectively, and 0.45 solid fraction about 88% and 95% of the bulk density. Fig. 6 shows the cross-sectional pictures of the films with film surface mobility 0.3, 0.5, 1.0, and 2.0. The changes of film density and void distribution are clearly shown in these pictures.

The atomic average coordination number, however, does not change with surface mobility and stays at about 6.0. Similar results have been observed by several researchers and are due to the three-contact stability criterion [15]. Fig. 7 shows the distribution of

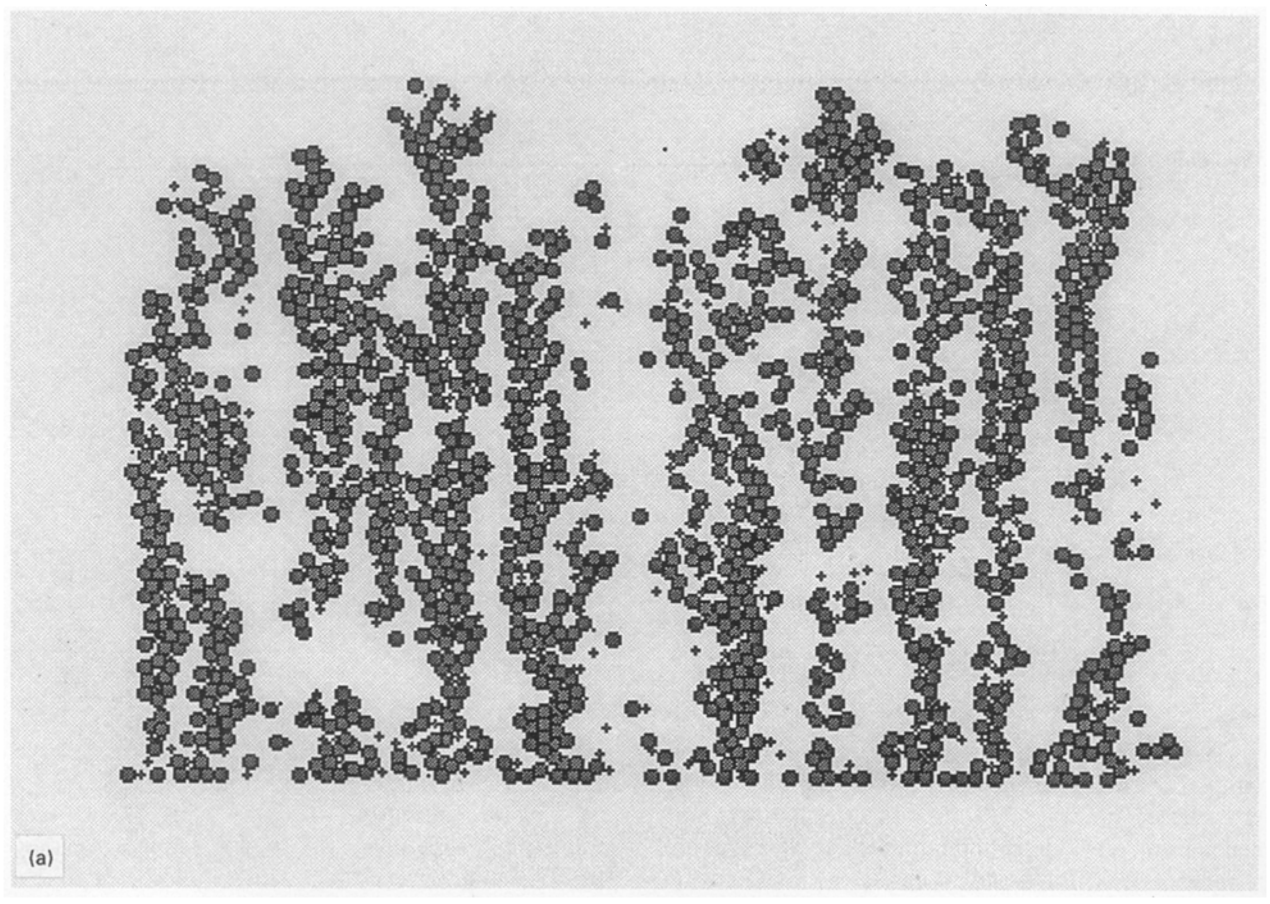
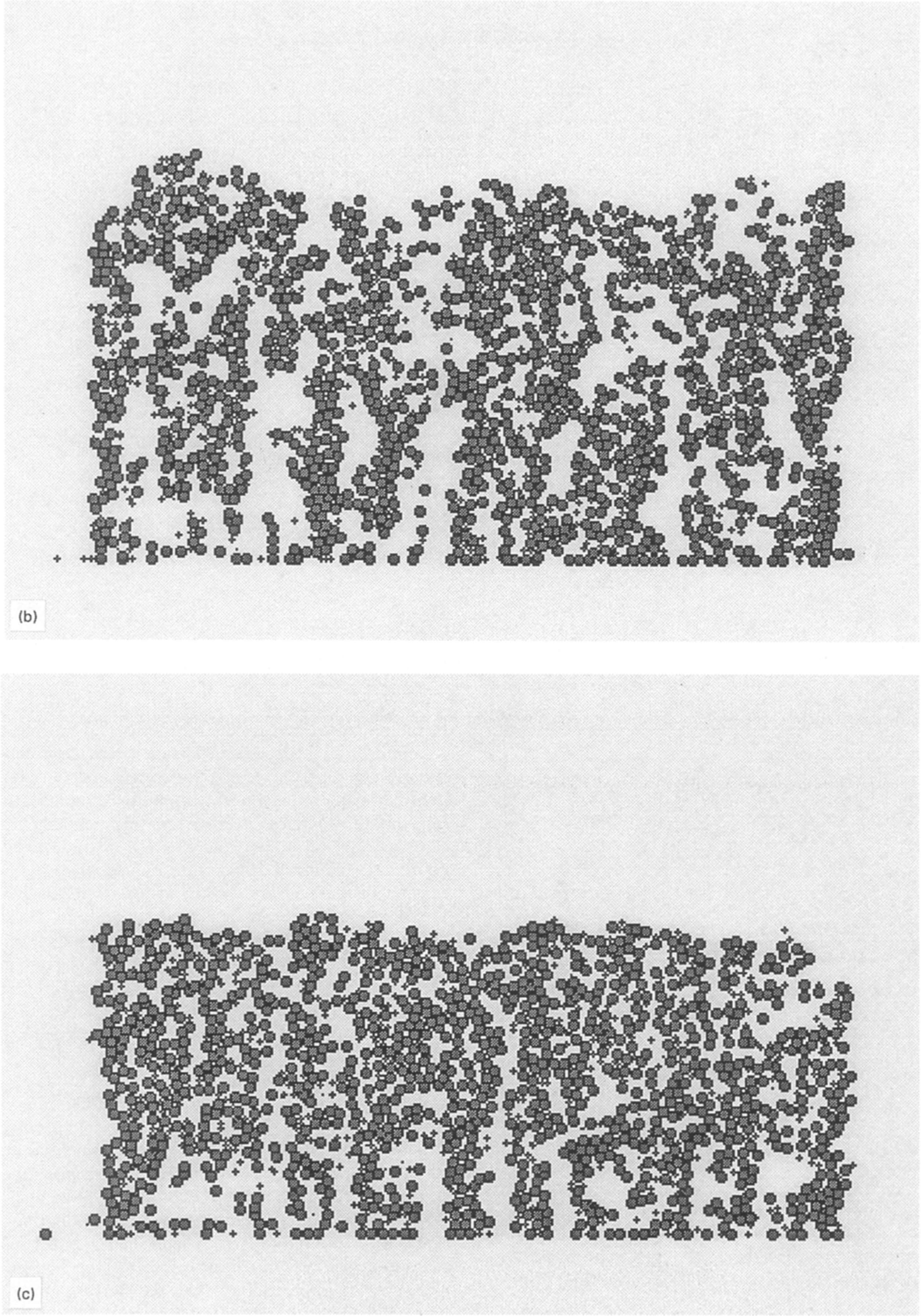


Figure 6 Cross-sectional area pictures of the films with film surface mobilities of (a) 0.3, (b) 0.5, (c) 1.0, and (d) 2.0. Deposition area 70 × 70 and 120000 atoms.



atomic coordination number within the film. It is observed that the full-width half magnitude (FWHM) decreases rapidly from 4.9 to 2.9 with mobility increasing from 0.3 to 1.5. Afterwards, the FWHM (~2.8) decreases slowly. The curves with mobility 2.0 and 3.0 are not plotted in the figure because they virtually coincide with the curve of 2.5. Less divergence in the coordination number represents a film with better

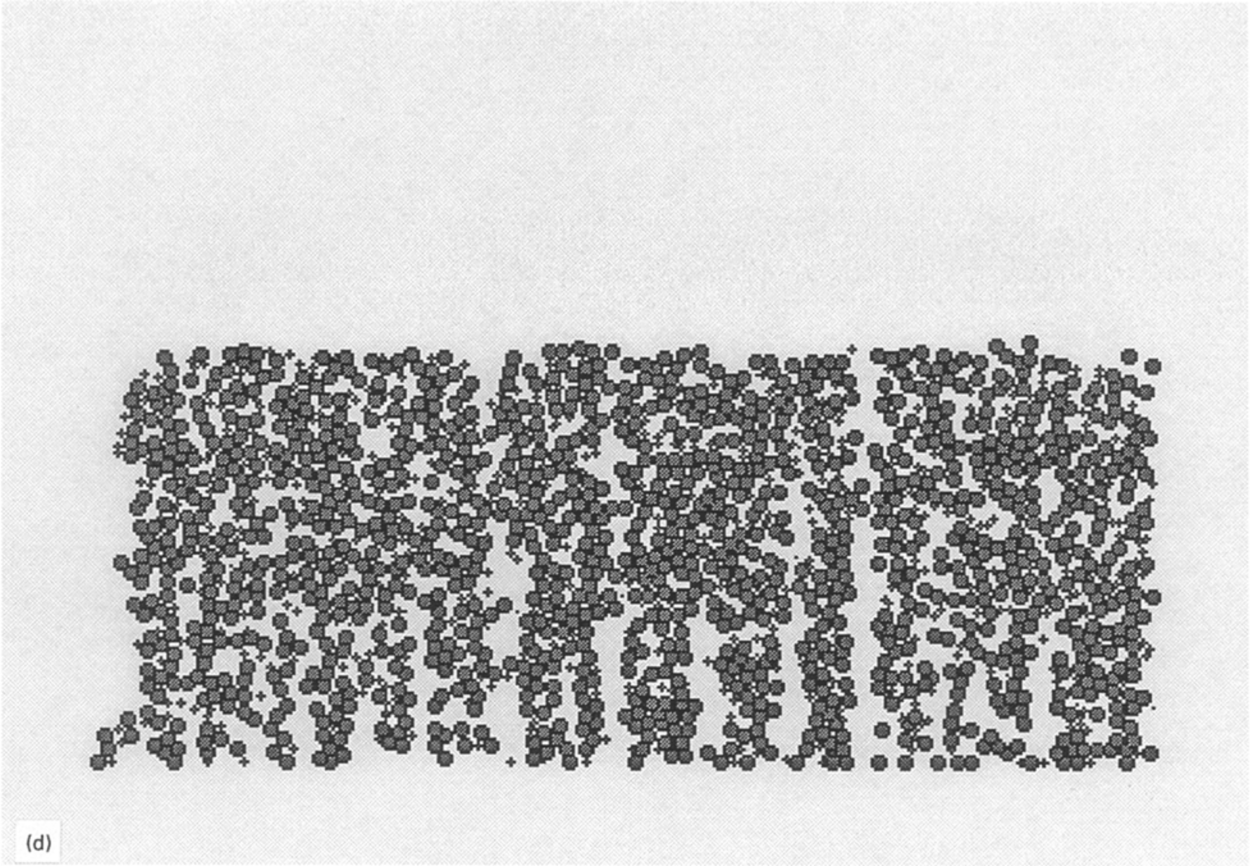


Figure 6 Continued

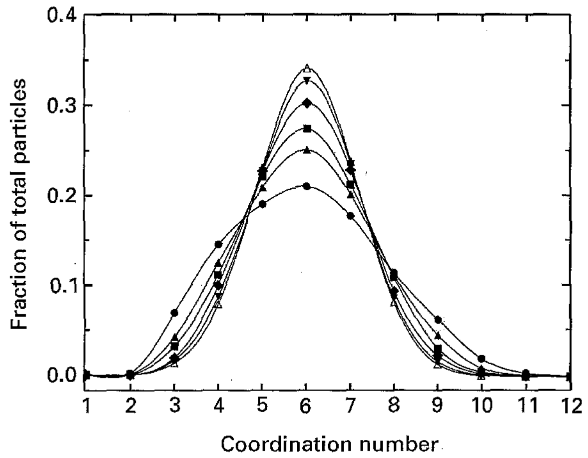


Figure 7 The distribution of the atomic coordination number versus the film surface mobility: (●) 0.3, (▲) 0.5, (■) 0.7, (◆) 1.0, (▼) 1.5, (△) 2.5.

ordered structure, or the creation of larger microcrystal grains. Therefore, the divergence of the coordination number can be regarded as a measure of the degree of crystallization in the film. With higher film surface mobility, films are more crystallized.

3.3. Discussion

From the results presented above, the simulated film grows in a clear island growth mode. The film surface mobility, as a lumped parameter with the effect of substrate temperature, atomic bonding energy and incoming atom energy, determines the film morphology and structure. With the increase in surface mobility, the film surface becomes smoother and the film density is higher. For a surface mobility below 1.5, the film surface roughness, average film thickness (for the same amount of atoms), and FWHM of the atomic coordination number distribution, reduce rapidly with a rapid increase in film solid fraction. For films of mobility above 1.5, further changes in solid fraction, roughness, thickness and FWHM of the coordination number distribution are much slower. It is suggested that a mobility of 1.5 is a rough boundary to differentiate between two different film structures and morphologies.

With the clear visualisation of the film morphology and structure as shown in Figs. 2, 3 and 6, it is seen that the films of mobility below 1.5 exhibit loose columnar growth which belongs to Zone I (porous structure with voids) in Movchan and Demchishin's structure zone model [1] and in Thornton's structure zone model [2]. As a result, the solid fraction is small, FWHM of the atomic coordination number distribution is wide, and the surface is rough. The fact that the average atomic coordination number is unchanged with mobility indicates that, with the increase of mobility, the voids within the film are reduced and the film becomes less porous. However, the narrowing of FWHM of the coordination number distribution may indicate a transformation from a random amorphous structure to a polycrystalline grain structure. Films with mobility above 1.5 can be categorized into Zone II in Movchan and Demchishin's structure zone model, and Zone T or Zone II in Thornton's model, which is characterized by more densely packed fibrous or columnar grains. It is obvious that the films with mobility above 1.5 are of higher density with a smoother surface and almost without voids compared to the films with mobility below 1.5.

4. Conclusion

A three-dimensional atomistic thin-film deposition model is successfully developed. Compared to the previous two-dimensional models, our model can simulate realistic thin film images together with the surface profiles and cross-sectional area pictures. In addition, quantitative results of the surface RMS roughness, average film thickness, atomic coordination number and its distribution, and solid fraction of the deposited thin films, are obtained from the simulation. It is shown that with the increase in surface mobility from 0.3 to 3.0 (crossing the boundary 1.5) the film evolves from a porous or loose columnar structure with voids to a densely packed fibrous grain structure which can be categorized by the zone structure models. With higher surface mobility the film becomes smoother with a higher density and more crystallized. The model is being developed further to take into account the re-evaporation of single atoms, critical cluster size, migration of single atoms on substrate surface, etc.

References

1. B. A. MOVCHAN and S. V. DEMCHISHIN, *Fiz. Met. Metalloved* **28** (1969) 653.
2. J. A. THORNTON, *Ann. Rev. Mater. Sci.* **7** (1977) 239.
3. S. DAS SARMA, I. K. MARMORKOS and S. M. PAIK, *Surf. Sci.* **228** (1990) 28.
4. R. HRACH and M. SOBOTKA, *Int. J. Electron.* **69** (1990) 49.
5. KARL-HEINZ MÜLLER, *J. Appl. Phys.* **58** (1985) 2573.
6. S. MÜLLER-PFEIFFER, H. VAN KRANENBURG and J. C. LODDER, *Thin Solid Films* **213** (1992) 143.
7. H. J. LEAMY, G. H. GILMER and A. G. DIRKS, *Curr. Top. Mater. Sci.* **6** (1980) 309.
8. D. J. HENDERSON, M. H. BRODSKY and P. CHAUDHARY, *Appl. Phys. Lett.* **25** (1974) 641.
9. PAUL MEAKIN, P. RAMANLAL, L. M. SANDER and R. C. BALL, *Phys. Rev. A* **34** (1986) 5091.
10. R. JULLIEN and P. MEAKIN, *Europhys. Lett.* **4** (1987) 1385, *and references therein.*
11. RUDOLF HRACH, *Int. J. Electron.* **69** (1990) 55.
12. SHI XU and G. Q. LU, *J. Mater. Sci. Lett.*, **13** (1994) 1629.
13. G. Q. LU, L. B. TI and K. ISHIZAKI, *Mater. Manuf. Proc.* in press.
14. PAVEL SMILAUER, M. R. WILBY and D. D. VVEDENSKY, *Phys. Rev. B* **47** (1993) 4119.
15. J. RODRIGUEZ, C. H. ALLIBERT and J. M. CHAIX, *Powder Technol.* **47** (1986) 25.

Received 23 August 1994
and accepted 17 May 1995