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Three Dimensional Simulation of Thin Films Growth

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Abstract

A simple three dimensional model are employed to investigate the relation between film microstructure and deposition condition (substrate temperature, deposition rate, deposition angle and substrate roughness). Increasing substrate temperature and deposition rate leads to fewer and smaller voids, smoother surface and higher film density. As the deposition angle increases, the film microstructure changes from a dense film with few voids to highly porous structure of well-formed columns. In this model, hard sphere travels in a straight line and relaxes on lattice point with longer catchment length. It is found that surface diffusion plays an important role in thin film growth and relaxation depends on the surrounding geometry of the previously deposited atoms. Using this model as a base for our calculations, the relation between deposition parameters and film structure is discussed in details and found to be consistent with measurements published in the literatures.

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Introduction

In the physical vapor deposition (PVD), atoms are deposited from a gas phase onto a substrate without the decomposition of one chemical species into other species, as in chemical vapor deposition. The parameters that describe the PVD deposition process include deposition rate, deposition angle (i.e., the angle the velocity vector of a depositing atom makes with the substrate normal), the substrate temperature, etc [1-7]. These deposition parameters all play an important role in determining the microstructure and physical property of PVD-grown films. There have been considerable attention to effects of these parameters in simulation of thin film growth.

Due to the non-equilibrium nature of all vapor deposition methods, the resulting film properties are generally highly dependent on preparation parameters. In most cases it is

the physical structure of thin film which is either directly responsible to the film property or related to them : for example, textured surfaces of Si and Ge for solar absorption [8] and optical data storage [9], magnetic anisotropy [10], and a-Si:H for photovoltaics [11]. The most important and commonly reported microstructure consists of columns of different density material.It is now well known that this density and the grain size directly depend on the substrate temperature [2-5,12,13]. The columnar structure was first reported by Movchan and Demcheshin [12] who examined thick films of metal oxides and proposed three zone model depending on the ratio of substrate temperature (T_s) to the melting point of the evaporation (T_m).

The microstructure of PVD grown films has been divided into three distinct zones based upon the substrate temperature during deposition. At substrate temperatures below approximately $0.3T_m$ the films tend to exhibit a very porous columnar microstructure, this porous structure is increasingly obvious as the deposition angle become larger(i.e., increasing oblique). Experimental observation suggests that the angle of orientation of these columns (relative to the substrate normal) β, in these so-called zone I microstructure,is generally smaller than the deposition angle(relative to the substrate normal) α: i.e., the columnar structure is oriented more nearly perpendicular to the substrate than the deposition flux. At substrate temperature $0.3T_m < T_s < 0.45T_m$ microstructure is columnar with smooth surface and at $T_s > 0.45T_m$ there are bright surface with recrystallization grain structure.

Recently, scanning tunneling microscopy (STM) has enabled us to view atoms and their arrangement during the evolutionary growth of thin films. New results, related to the nucleation/initialgrowth stage of thin films have obtained, consisting of pyramid like structure(mound morphology), the size and separation of which increase with film thickness [14]. The mound separation shows coarsening versus growth time with $L(t) \sim t^n (n = 1/4)$, consistent with the theory based on capillary between mounds.A monte carlo simulation on a simple square lattice system [15] showed that the value of n is sensitive to the growth temperature, T_s.

Many authors have investigated the zone model structure of different materials [2-7,12,13] and found a porous columnar network in zone I, it's size increase with temperature. From the theoretical or simulation point of view, this structure may be related to the shadowing effect which is based on the principle that peaks receive a greater deposition flux than the valleys. Thus instead of the surface growing at a uniform rate, the peaks grow at a faster rate than the valleys leading to the development of the structure seen in the zone I region [16]. Dirks and Leamy [1] in their hard disc atomistic simulation of thin film growth, and Messier and Yehoda [17] who describe the shadowing effect as "natural clustering due to ballistic aggregation" concluded that the low adatom mobility in zone I can lead to self-shadowing which results in the microstructure of columnar morphology with micro-voids. Thus the shadowing effect plays an important role in the growth of columnar crystal in thin films. As substrate temperature increases a second mechanism known as surface diffusion is activated. The growth of grains in the microstructure is attributed to the surface diffusion of atoms. In the tow-dimensional surface of the film, atoms are able to move around and can diffuse across grain boundaries from one grain to another. Increasing the substrate temperature increases the activation energy

of the surface atoms, and then the rate of surface diffusion increases [5]. The effect of atom mobility caused by thermally activated hopping processes is modeled by Muller [18,19]. It was found that from a certain substrate temperature onwards the migration rate of adatoms to shadowed region is large enough and causes void incorporation during growth. therefore structure changes at the transition temperature from a porous columnar microstructure to a film with no micro-voids.

Computer simulation of thin film growth 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 the growth mechanism. Many important parameters and effects can be easily studied from simulations while they are difficult to study experimentally. Most of the simulation models can be categorized into either analytic or atomistic [18-26]. The atomistic models usually are 2D simulation of film microstructure and morphology [27]. Some 3D atomistic deposition models have been reported [26] generating various film solid fractions ranging from 0.128 to 0.582.

In this paper, we report the growth of thin films in all three zone of SZM, by proposing a simple simulation model in which the relaxation depends on the surrounding geometry of the previously deposited atoms, while the influence of different parameters, namely, substrate temperature, deposition rate,surface roughness, angle of incidence and surface diffusion are also discussed.

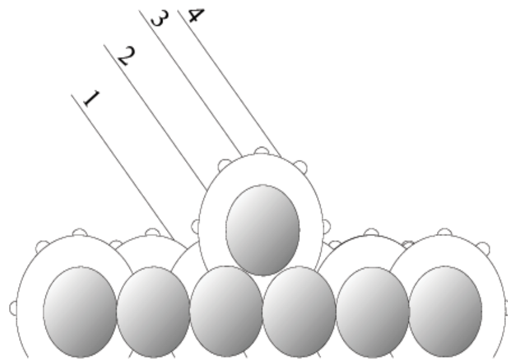
The model

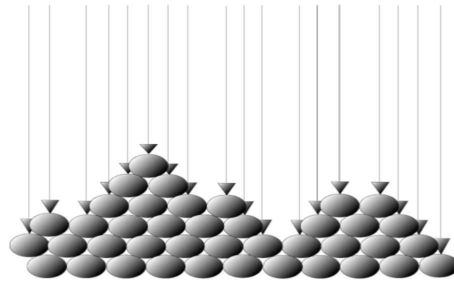
Muller [18,19] and Dirks and Leamy [1] used the idea of Henderson et al [20] ideas in order to simulate the growth of thin films with low adatom mobility in two dimensions. Savaloni and Gholipoor [27] have used two-dimensional simulation to study the dependence of microstructure on the deposition rate and deposition angle and substrate temperature. Atoms (hard disc)travel along straight liens, which make an angle α with the normal to substrate. When atoms impinge on the surface they migrate if they have enough energy, else they relax in the position that has longer catchment length.

In this model we study three dimensional simulation of thin film growth. In order to account relaxation process of the arriving atoms, we consider lines that are parallel with trajectory of arriving atoms (figure 1(a)). If an atom is located between two lines, the relaxation will be into the site positioned between two lines. The distance between these parallel lines is called catchment length and provides a basis for determining where the atom hits on surface and relaxes.Then we can determine the nearest 'cradle'. Every atom can have 12 neighbors.

On this basis, one can define a "catchment length" which corresponds to the relaxation probability of deposited atom into each unoccupied lattice point. Figure 1(b), (c)represents a surface with peaks and valleys and shows the respective catchment length.

For inclusion of surface diffusion process in this model, the stochastic gas model [36] was employed. If the adatom is not buried under the following atoms, and if it has enough thermal energy to overcome the local potential (activation energy)barrier then it will perform random hops.





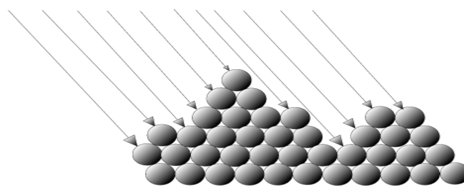


Figure 1: (a)Catchment length, when an atom can occupy the lattice point between two atoms.Distance between lines 1,2 is more than 3, 4 then probability of it's site is more than the other.(b)Catchment length for different unoccupied possible lattice points in normal evaporation.(c)Catchment length for different unoccupied possible lattice points in oblique evaporation.

In order to consider the deposition rate in our simulation model, we use the following procedure:the mean number of hops N, which an adatom performs on an ideal surface at the substrate temperature T_s in the time interval Δt between two newly arriving vapor atoms, is determined by the equation

$$N = \frac{\tau_m}{\tau_h} \tag{1}$$

where τ_m is the deposition time for the growth of one atomic layer and is defined by:

$$\tau_m = \frac{a_d}{r} \tag{2}$$

in which a_d is the average distance between atoms in the direction perpendicular to the substrate surface(which for a film with bulk density will be close to the lattice spacing), r is the deposition rate, and τ_h is the mean time between hops which is determined by the substrate temperature(T_s), the energy needed for one hope (Q) and the lattice vibration frequency (υ) [28]:

$$\tau_h = \frac{1}{\upsilon}\exp(\frac{Q}{KT_s}) \tag{3}$$

Here Q is the surface activation energy, υ is of the order of $10^{14}s^{-1}$, and K is the Boltzmann constant.

By using Boltzman statistics, the probability of the energy of an atom being more than E is [29]

$$P(\varepsilon > E) = 1 - \exp(\frac{-E}{KT_s}) \tag{4}$$

Hence

$$E = -KT_s \ln(1 - P^N) \qquad 0 < P < 1 \tag{5}$$

Where P is a uniformly distributed random number in the interval(0,1), and can be sampled using the Monte carlo rejection technique [30,31].

Muller [18] proposed the following equation for obtaining the local activation barrier:

$$\begin{aligned} B_{ij} &= Q & N_i < N_j \\ B_{ij} &= Q + (N_j - N_i)\phi & otherwise \end{aligned} \tag{6}$$

where ϕ is the energy of a single bond, Q is the activation energy for surface diffusion, N_i is the number of nearest neighbors. Q is parameterized in terms of the temperature and the metal melting temperature according to the experimental results of Neumann and Hirschwald (valid for $T_s < 0.5T_m$) [18,32]:

$$Q = (5 + \frac{20}{3}\frac{T_s}{T_m})KT_m. \tag{7}$$

By comparing the thermal energy with local activation energy barrier, the possibility of adatom migration is investigated. If this adatom has enough energy, it will migrate to site with lowest energy. The diffusion length of an adatom is implemented using

$$\bar{\chi} = \frac{1}{2}a_d(\frac{\tau_m}{\tau_h})^{\frac{1}{2}}. \tag{8}$$

Simulation of oblique evaporation is considered by tilting and aligning the perpendicular lines of figure 1(a),(b)with the direction of atoms. As shown in figure 1(c), this causes a change in the catchment length of particular site and increases the shadowing effect. Therefore film should grow obliquely.

The model is programmed using Java and is summarized in flowchart given in figure 2. The atomic arrangement of the substrate is simulated in step 1, and in step 2 an atom is generated. In step 3 atoms trajectory and in step 4 hit position on substrate are evaluated. Then position to cradle is found based on catchment length in step 5. Migration possibility is investigated based on equation(5)-(7)in step 6. After the first evaporated atom is relaxed to a position on the substrate surface, the determination of the capture length and probable relaxation sites for the following atom is repeated.

Result and discussion

In our simulation, we consider the film to have five mono-layers and the number of atoms, n, are 2000. The number of deposited atom is 1000. In order to simulate on infinite system, periodic boundary conditions are applied.

Normal incidence on flat surface at different substrate temperatures and deposition rates

Figure 3(a) shows the result of a typical simulation of Ni ($T_m = 1726, \phi = -0.74$ev) film at 400K substrate temperature with deposition rate of $R = 0.1 MLs^{-1}$. In all figures, yellow spheres are buried atoms and red spheres are diffused atoms and black spheres, belong to the substrate. Increasing the substrate temperature (figure 3(b), $T_s = 750K$, $R = 0.1 MLs^{-1}$), increases diffused atoms, while at $T_s = 400K$ only a few atoms are diffused. The result for higher deposition rates are similar to that of figure 3(a). This effect is the result of increasing the number of buried atoms at high deposition rates as well as low mobility at lower temperatures. By increasing the substrate temperature to 850K, the diffusion processes dominate other processes(figures 4(a)-(d)), as expected. From figures 4(a)-(d), it is observed that by increasing the deposition rate, the number of buried atoms increases, the yellow and red spheres represent adatoms, buried atoms and diffused atoms respectively. The number of diffused atoms decreases by increasing the deposition rate. This is because of the bombardment of deposited atoms by the next atoms. At higher rates, the transition from zone I structure to higher zones happens at higher temperatures. Initially, instead of columnar structure, structure similar to zone I is produced.

Figures 3,4 also show that with increasing the substrate temperature, as is experimentally observed, the microstructure of the film changes from porous or columnar to densely packed or re-crystallite films without micro-voids(particulary, more pronounced for lower deposition rates). This behavior is also seen in other models(e.g.see [18]).

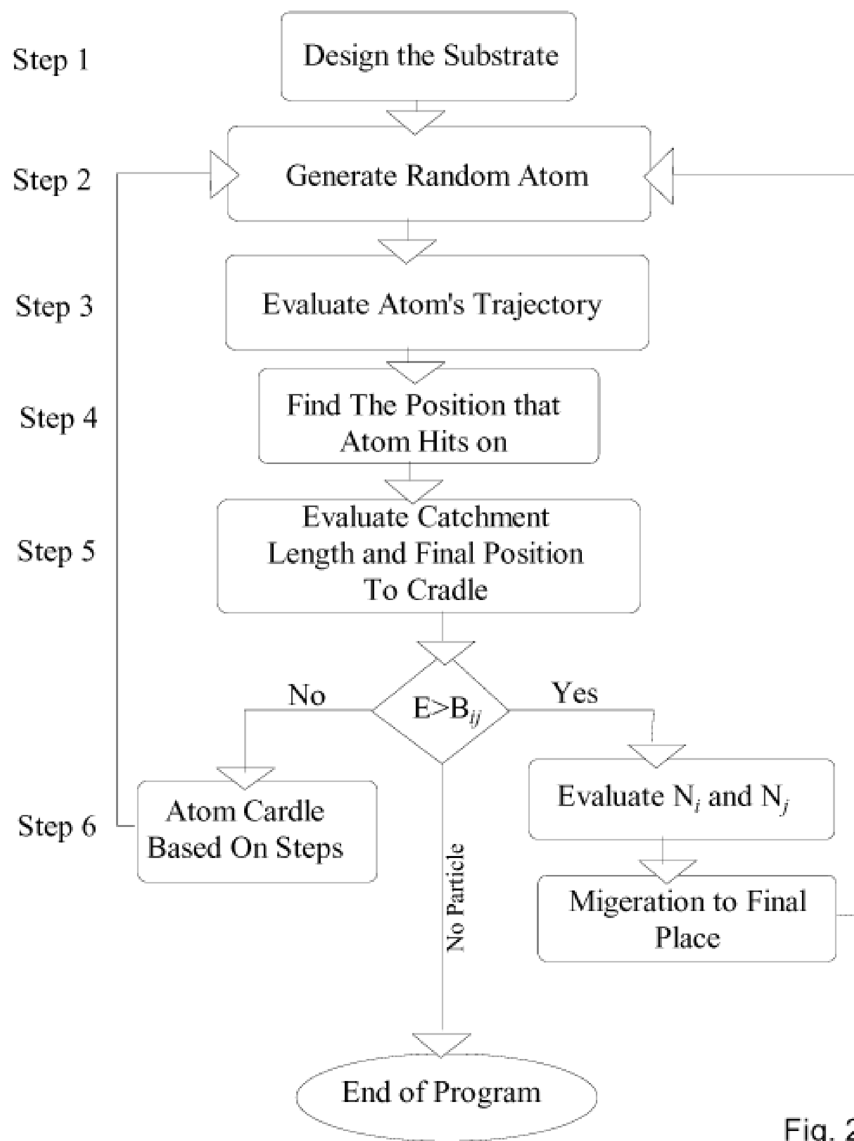


Figure 2: Flowchart of the algorithm for the deposition of a layer.

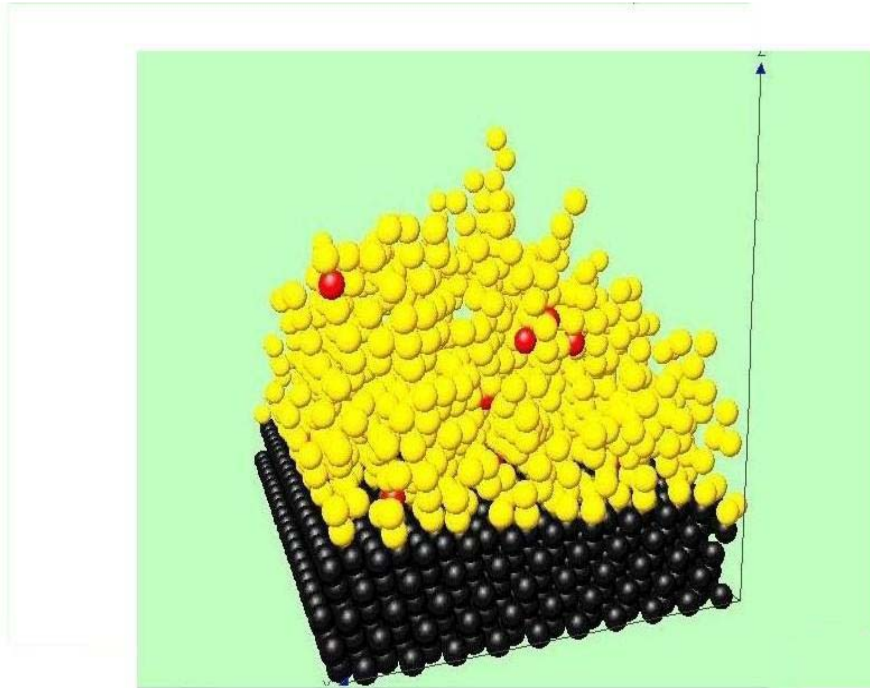


Fig.3a

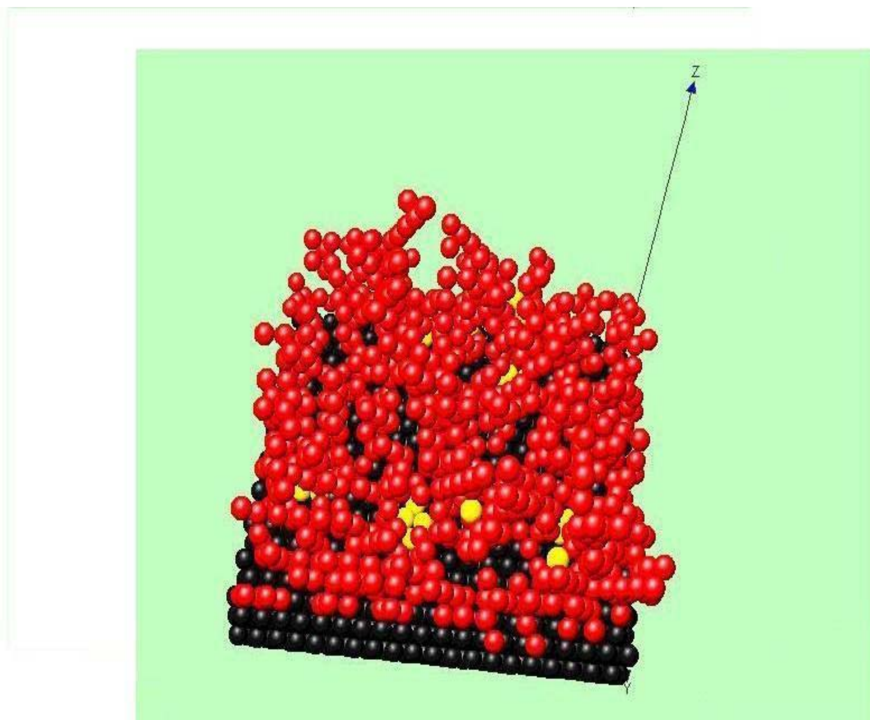


Fig3b

Figure 3: Typical thin film growths on smooth surfaces at normal incidence and for deposition rate of$R = 0.1MLs^{-1}$. (a) $T_s = 400K$, (b)$T_s = 750K$.Red spheres:diffused atoms,yellow spheres:buried atoms.

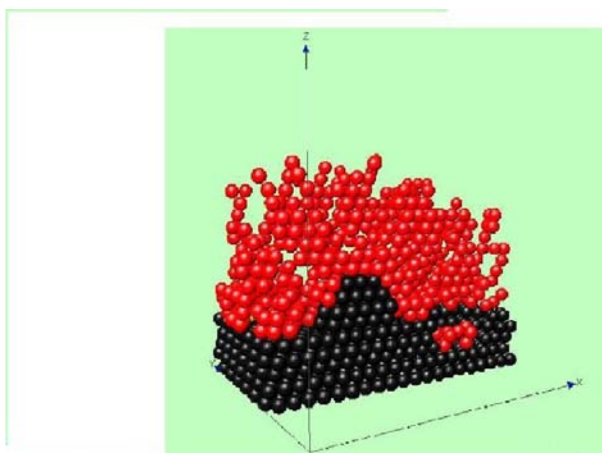


Fig5a

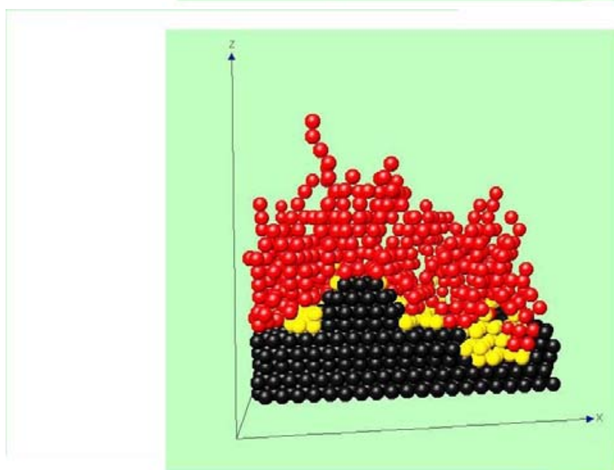


Fig5b

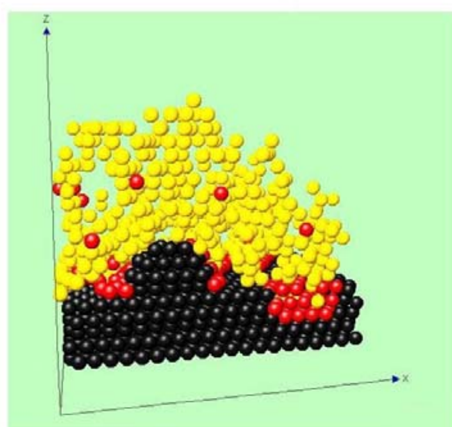
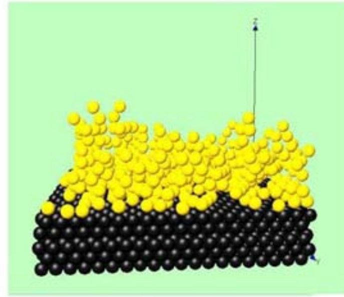
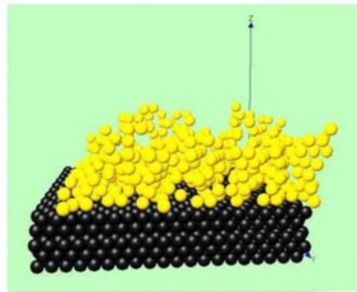
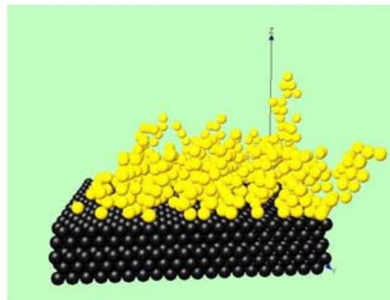


Fig5c

Figure 4: Typical thin film growths on rough surfaces at $T_s = 850K$, normal incidence and for different deposition rates of: (a) $R = 0.001MLs^{-1}$ (b)$R = 0.1MLs^{-1}$ (c)$R = 10MLs^{-1}$. Atoms color is as defined in figure 3.







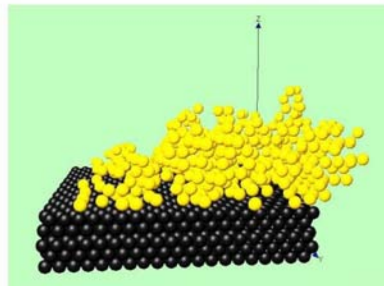


Figure 5: Typical thin film growths on smooth surfaces at different incidence angles ($T_s = 400K$, $R = 0.1$): (a)$\alpha = 15$ (b) $\alpha = 30$ (c) $\alpha = 45$ (d) $\alpha = 60$. Atoms color is as defined in figure 3.

Normal incidence on rough surfaces as a function of deposition rate and substrate temperature

The effect of substrate roughness is examined by exposing a rough surface to the incoming atoms. The rough surface is simulated by production of peaks and valleys of different heights and depths on the substrate surface. At $T_s = 850K$ similar results with no considerable change are obtained for all deposition rates R=0.001,0.1,10. As expected the self-shadowing effect as well as the roughness of the surface, on smooth surfaces, causes the largest column-like growths with void in between, is also observed. At $T_s = 850K$(figures 5(a)-(c))the number of buried adatoms are increased, at high deposition rates $10MLs^{-1}$, as discussed in section III.A. At lower deposition rates, due to increase diffusion processes [3] and for reduced or non-existence of buried adatoms, the expected zone III structure is obtained(figures(a)-(c)). However at lower deposition rates, on top of the large irregularities, the film surface stands higher than the smooth section of the substrate surface, consistent with the shadowing theory proposed by Srolovitz et al [33].

Oblique incidence on flat surfaces

Since its discovery in 1959 [33], people have tried to use the oblique incidence effect to explain the observed phenomena. The origin of the special oblique structure of films has been related to the shadowing phenomenon during film growth. Nieuwenhuizen and Haanstra[34] described the relation between the column and the deposition angle in thier experimental results using the following relation $\tan\alpha = 2\tan\beta$. This equation which is called "tangent law", valid for deposition angle $\alpha \leq 60$. Because of shadowing, the film should consist of inclined columns with the bundles in which α and β are the vapor incidence direction and columnar inclination, respectively. Mazor et al [35] claim that in zone I the tangent rule is approximately valid, while the tangent of tilt of columns in zone II will be smaller than zone I due to enhanced surface diffusion.

Figures 6(a)-(d) show that results of 400K substrate in which the angle of the growth of bundles is clearly inclined towards the incidence atoms and agreement with the tangent rule is obtained.

Conclusion

A three-dimensional hard sphere ballistic aggregation model for thin film growth is reported. The influence of substrate temperature, deposition rate, angle of incidence, self-shadowing effect and substrate roughness are investigated, and qualitative agreements with the prediction of structure zone model(SZM)and theoretical results of Srolovitz [16] are obtained. Although the results at low substrate temperature (i.e. zone I) are in dendritic form and reflect very low diffusion processes at higher substrate temperature, the model produces consistent result with SZM. If these are compared with the result of Muller [18] who used Henderson's [1,20] prescription for adatom relaxation, one can observe that Muller's results are associated with high rate of diffusion at low substrate

temperature (i.e. zone I) and transition to structure of zone III takes place at zone I temperature. Therefore, it may be proposed that by some modifications to the simple model proposed here, one may obtain agreement with SZM in all three zones.

In general, this simple model provides almost all predicted results and agrees well with observations. The advantage of this model to those previously reported is closer to the real film growth due to the simplicity of the assumptions made.

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