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TIME DEPENDENCE OF AREA COVERAGE OF THE N-TH LAYER DURING THIN FILM GROWTH AT LOW TEMPERATURES

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ABSTRACT

This paper describes a model representing growth of thin solid films at low substrate temperatures. We derive an expression for the Area coverage of the n^{th} layer, $A_n(R,t)$. For small time scales, i.e. small compared to the time to grow a monolayer, $A_n(R,t) \sim t^n$. Results of the numerical simulation are in excellent agreement with the above predictions.

Key words: Thin solid films, model, simulation, and area coverage.

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1. INTRODUCTION

Thin solid films are used in a variety of important applications including integrated circuits, information storage devices, and coatings for wear protection and corrosion resistance [1]. A feature crucial to the appropriateness of a film to a given application is it *physical structure*. This is because most of the technologically important properties of a film, i.e. electrical, mechanical, optical, acoustical and magnetic, are strongly dependent on the structure of the deposited film. Hence a clear understanding of the nature and the cause of film structures are technologically important and remain a topic of theoretical challenge.

Existing models for thin film growth can be divided in to two categories as macroscopic and microscopic growth models [2-3]. Although detailed information down to very small length scales could in principle be obtained, microscopic model are difficult to solve analytically and computer intensive. Inability to provide detailed information about the physical structure is one of the most crucial disadvantages of the macroscopic models

In this article we describe a microscopic model that describes film growth at low temperatures. It is suitable for explaining any of the atomistic deposition techniques [4] in the limit, where surface and bulk rearrangement processes of the deposited atoms are inactive, i.e. at low substrate temperatures. The model can be solved either numerically or analytically. It can be used, in addition to explaining the relevant experiments, to check the accuracy of simulations by comparing their predictions with that of the model in the appropriate temperature limit. This article also presents such comparison.

2. THE GROWTH MODEL

In this section we will introduce the model [5]. Films are grown over carefully prepared cleaned substrates. Surfaces of the substrates are, in general, rough in the atomic scale and are known to be the cause of columnar growth observed at low growth temperatures [6]. We assume in our model that the surface of the substrate to be flat. Analytical studies become simpler with this assumption and can be readily relaxed during the corresponding simulations.

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Surface of the substrate is considered as a two-dimensional (2D) lattice with one atom at the center of each of the N_0 unit cells. During a typical growth experiment the over-layer material and hence its crystal structure and the lattice parameters, in general, differ from that of the substrate. This "misfit" gives rise to a build up of strain energy. It is known that up to a certain height the over-layers atom follows the crystal structure corresponds to that of the underlining substrate. Beyond this critical height, which depends on the two materials and the experimental conditions, over-layer material collapses to energetically more favorable crystal structure such as that of the over-layer material [7]. Our model considers only those films with thickness less than this critical height. We assume that the depositing atoms can only take the position right above the center of the unit cell, i.e. above an already deposited atom or on an atom of the substrate yet unoccupied. We assume further that any given incoming atom is equally like to select any of the unit cells. For temperatures above the activation energies of various surface and bulk transport mechanisms, migration of adatoms is the rule and the model neglects this possibility [8]. For this reason, the model is not appropriate for explaining films grown at elevated temperatures.

Once the growth is allowed to proceed, according the assumptions specified above, it is possible to *analytically* determine certain measurable quantities. On the other hand any quantity of interest should be able to obtain by means of the corresponding simulation.

We now present the derivation of an expression for the *Area coverage* of the n^{th} layer above the surface of the substrate at any time *t*. Let us denote this quantity by $A_n(t)$.

In terms of the number of occupied sites in the n^{th} layer at time t, $N_n(t)$, we define $A_n(t)$ as

$$A_n(t) \equiv \frac{N_n(\mathrm{t})}{N_0}$$

Let us assume that the growth proceeded at the rate R layers per second for a period of t seconds. Hence the total number of deposited atoms and the average height of the film is given by RtN_0 and Rta respectively. Here a represents the distance between adjacent layers. It should be obvious that at the end of time *t*, the probability of having *m* adatoms, on top of each other, above *any* unit cell of the substrate, $P_m(R,t)$, is [9]

$$P_m(R,t) = \frac{(Rt)^m}{m!} e^{-Rt}.$$

In terms of $P_m(R,T)$ one can immediately write the following expression for the Area coverage,

$$A_n(R,t) = \sum_{m \geq n} P_m(R,t).$$

Last two equations imply

$$A_n(R,t) = 1 - \left(\sum_{\ell=0}^{n-1} \frac{(Rt)^\ell}{\ell!} \right) e^{-Rt} \qquad (1).$$

Last equation represents an exact result. It implies, for time scales smaller compared to the time required to grow a monolayer, i.e. $t << 1/R$, that (see appendix)

$$A_n(R,t) = \frac{(Rt)^n}{n!} \qquad (2).$$

Deviation from this simple power law behavior is to be expected even at small time scales. The reason for that is the effect of *geometrical shadowing* [10] of the less grown or "valley" regions of the growing film by the "hills".

3. NUMERICAL WORK

We carried out a simulation using MATLAB, in which a film was grown under the conditions specified in the previous section. A 2D null matrix represented the substrate.

In order to represent the deposition, a unit matrix with appropriate number of its elements set to zero, was added to the substrate during each time step. The locations of the zero matrix elements were selected at random. As the growth proceeds, instructions were written in the program to keep a track of the area coverage of each layer as a function of time.

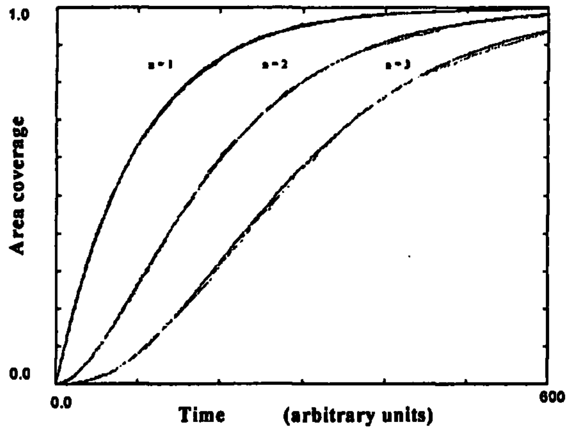

Fig. 1 Time evolution of Area coverage, $A_n(t)$, for the first three over-layers. Smooth lines represent theory and the others the simulation.

In Figure 1 we compare, the model predictions with the results of the simulation. In order to avoid overcrowding, the time evolution of the *area coverage* of only the first three over-layers are presented in the figure. Simulation corresponds to Figure 1 was carried out for long enough duration so that even the third layer of the film is almost filled. On a Pentium II 400 MHz machine, it took only *few minute* of CPU time to execute this program. The size of the 2D matrix, which represents the size of the substrate, is 50 X 50. As the plots indicate, there is an excellent agreement between the theory and the simulation.

4. DISCUSSION

We presented a growth model, valid at low temperatures, to represent atomistic growth techniques such as Sputtering, CVD, and MBE. The model was solved analytically to obtain an expression for the time evolution of area coverage for each over-layers. Numerical results are found to be in excellent agreement with the theory. The model and in particular the above solution that it renders can be use to verify the accuracy of more sophisticated simulations developed to represent growth of thin films at elevated temperatures.

We are aware of the existence of an experimental study carried out to measure the time evolution of area coverage of each over layers. Based on the plots given in that work it was found that a near power low behavior of the Area coverage. Present work resulted as an attempt to explain that experiment and, we are unable to trace that work during the literature search.

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APPENDIX

Derivation of the power low behavior at small time scales. Two methods are presented here.

(i) If one starts from $A_n(R,t) = \sum_{m\geq n}^{\infty} P_m(R,t)$

which can be written

$$A_n(R,t) = \sum_{\ell\geq n}^{\infty} \frac{(Rt)^\ell}{\ell!} \approx \frac{(Rt)^n}{n!}.$$

Hence the claimed $(Rt)^n$ behavior emerges naturally.

(ii) If one starts from

$$, A_n(R,t) = 1 - \left(\sum_{\ell=0}^{n-1} \frac{(Rt)^\ell}{\ell!} \right) e^{-Rt}$$

he must be careful to retain enough terms in the expansion of e^{-Rt}.

For example, if needed to evaluate $A_4(R,t)$ starting from the above equation, we must proceeds as follows

$$\begin{aligned} A_4(R,t) &= 1 - \left(\sum_{\ell=0}^{4-1} \frac{(Rt)^\ell}{\ell!} \right) e^{-Rt} \\ &\approx 1 - \underset{[\text{exact}]}{\left[1 + (Rt) + \frac{(Rt)^2}{2!} + \frac{(Rt)^3}{3!} \right]} \underset{[\text{approximat e}]}{\left[1 - (Rt) + \frac{(Rt)^2}{2!} - \frac{(Rt)^3}{3!} + \frac{(Rt)^4}{4!} \right]} \\ &= \frac{(Rt)^4}{4!}. \end{aligned}$$