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INTERCONNECT FILMS II: ATOMISTIC SIMULATIONS OF
FILM DEPOSITION ONTO INCLINED SURFACES**

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To be published in

**Proceedings of the MRS Spring Meeting, 1999,
Session L, Polycrystalline Metal and Magnetic Thin Films**

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ABSTRACT

We present three dimensional lattice Monte Carlo simulations of sputter deposition onto inclined surfaces. For this purpose, we use a model of an fcc material and we vary the substrate inclination, the texture, and the atomic mobility. In this way we can examine different conditions and mechanisms involved in depositing barrier layer and interconnect films. We obtain results on the density and the roughness of the films. We find that the film texture has dramatic effects on the film density and smoothness. Surprisingly, we find that the surface diffusion coefficients may not be critical parameters; changes in the coefficients by several orders of magnitude may cause only a small change in the film morphology when depositing onto non-wetting substrates.

I. INTRODUCTION

Barrier layer films are used in the microelectronics industry to prevent the intermixing of successive layers of different materials. For example, in the case of Cu interconnects, thin layers of Ta or its alloys are often used to prevent the Cu from diffusion into the SiO₂ layer, and from there into the active device regions. Barrier layer materials are chosen for properties such as low diffusivity in order to retard interdiffusion. However, precisely because of their small diffusion coefficients, they are susceptible to kinetic roughening of the surface during growth, and to the incorporation of voids and pinholes during the deposition of the film. Furthermore, the sidewalls of vias and trenches present additional difficulties, since the particles arriving on these surfaces arrive at oblique angles of incidence. It is known that a flux of atoms deposited at oblique angles can lead to a columnar film morphology [1,2], i.e. a film consisting of irregular columns often separated from one another by void regions. Voids between the columns are clearly detrimental to the function of the film, since they provide paths for interdiffusion between the layers on either side. A large number of experiments and much theoretical work has been devoted to characterizing the film properties and to explaining the self-shadowing process that leads to columnar growth [1]. The film densities and column angles have been studied in some detail [1,2,3]. Experiments show that the column inclination is tilted away from the substrate normal in the direction of the incident particle flux [1].

In this paper, we present simulations using a lattice Monte Carlo (MC) model, with ballistic deposition, realistic binding energies, surface diffusion, and grain boundaries. We study columnar growth on inclined substrates by modeling a target corresponding to a real experimental situation [4]. We present the influence of the surface diffusion coefficient and the presence of grain boundaries on the resulting morphology, surface roughness, and film density.

II. MODEL

ADEPT is a 3D MC model that has been developed to simulate metal films deposited by physical vapor deposition. The basic features of ADEPT have been described in a previous publication (see Ref.[5]) and are summarized in this section. Only the new characteristics are described in detail here.

In order to simulate the magnetron sputtering apparatus presented in an accompanying paper [4], we approximated the target by a square, with the geometry shown in Figure 1. The normal to the target is directed toward the center of the substrate. We define Ψ_m to be the angle between the normal to the substrate and the line joining the centers of the target and the substrate. Thus, Ψ_m denotes the substrate tilt with respect to the mean direction of the sputtered atom flux. Atoms arriving at the substrate have trajectories with angles in the range $\Psi \in [\Psi_m - 15^\circ, \Psi_m + 15^\circ]$, where Ψ is measured from the substrate normal and in the plane containing the substrate normal and the line of centers. Similarly, the angles measured in the perpendicular plane have $\phi \in [-15^\circ, +15^\circ]$. The interval bounds are chosen to correspond roughly to the experimental geometrical configuration; i.e., target/substrate distance and target size. We also assumed that the angular probability distribution is uniform for Ψ and ϕ . By this approximation, we neglect any intrinsic variation in the angular distribution of the target, and we assume that the substrate is small enough to neglect variations in the flux across its surface.

Atomic surface diffusion is described by diffusion coefficients of the form $D = D_0 \exp(-E_m/kT)$, where the prefactor D_0 and the activation energy for diffusion E_m are coordination dependent. A diffusion hop rate v is determined for each atom in the film based on its coordination number; it is related to D through the relation, $v = 4D/\lambda^2$, where λ is the hop distance. Atoms are selected for diffusion events with a probability proportional to v . Potential energies for the initial and final states are calculated using an embedded atom potential for the lattice MC model, but chosen to mimic the potential energies obtained using molecular dynamics. If the potential energy decreases from the initial to the final state, the event is executed with the rate v ; otherwise it is executed with a rate $v \exp(-\Delta U/kT)$ where ΔU is the potential energy increase.

Grain boundaries are modeled using a single crystal lattice, but assigning a grain orientation parameter Ω around the surface normal to each atom in the film. A deposited atom is assigned the same grain orientation as one of its nearest neighbors. An atom is considered to belong to a grain boundary when at least one of its nearest neighbors is in another grain than the one to which it belongs. Two kinds of events are possible for grain boundary atoms: a reorientation event to match that of a neighboring atom, or a migration event to a neighboring empty site, as described above. The reorientation event is executed with a rate determined by the product of a Boltzmann factor for any potential energy increase, and a prefactor calculated from molecular dynamics [6]. A reduced binding energy is assigned to a bond between atoms from two different crystallites. This value is a function of the mismatch angle between the two grains,

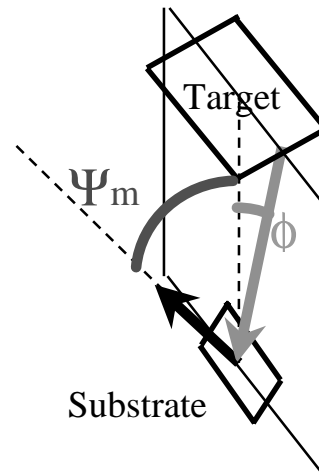


Figure 1: Substrate/target geometry, and angles used to describe the range of trajectories of atoms in the flux arriving at the substrate.

and is determined from molecular dynamics and experimental results (see Ref.[5]). This model describes very closely a polycrystalline film where the grains have a common out-of-plane orientation (texture), but different orientations in the plane. The model is appropriate for determining the influence of the grain boundary energy on surface morphology including grain boundary grooving, and possibly for assessing effects of grain boundaries on overall surface mass transport.

The principal difference between this model and earlier MC calculations such as those of Family [7] or Meakin and Jullien [8], is the fact that the present model is consistent with full three dimensional crystal surfaces. There is no approximation such as the no-overhang condition for solid-on-solid models. For this reason, this model can treat columnar growth, with effects of faceting on closed-packed surfaces such (100) and (111). In addition, the diffusion rates and atom energies are based on LDA and MD calculations of real materials, such as Al.

III. RESULTS

1. Simulation setup

Our model is based on the Al fcc crystal, because of the availability of surface atom potential energies and surface diffusion coefficients from first principles and molecular dynamics simulations [6,9]. In order to simulate results appropriate for refractory materials such as Ta, Ti, or TiN, we scale down the diffusion coefficients by a variable factor $s=D/D_{Al}$ with $10^{-6} \leq s \leq 10^{-1}$. The deposition rate is $1\mu\text{m}/\text{min}$, and we are working at room temperature. The substrate consists of an atomic layer containing 2500 to 10000 atoms depending on the substrate orientation. Periodic boundary conditions are applied in the lateral directions, and in all the simulations 50 monolayers (ML) are deposited onto this substrate. The imperfect wetting between the film and the substrate is included in the model by weakening all the film-substrate bonds by 0.15eV .

2. Morphology variation with substrate inclination and texture

In Figures 2(a), (b), we present simulations of polycrystalline {100} textured films for two different diffusivities, $s=10^{-1}$ and 10^{-4} . The substrate is inclined by angles of $\Psi_m=0^\circ$, 30° , 60° (See Fig.1). In these pictures, each shade of gray represents a different grain orientation. For $\Psi_m=0^\circ$, the polycrystalline films are nearly 100% dense, and consist of grains whose long dimensions are vertical. In the case where $\Psi_m=30^\circ$, voids are present in the grain boundaries and grains are inclined toward the direction of the impinging atom flux. Finally, a regime with completely isolated columns is reached for $\Psi_m=60^\circ$, where the large angle of incidence of the beam enhances self-shadowing. Note that high mobility produces stronger faceting (Fig.2(a)) and grain boundary grooving (0° of Fig.2(a)). At this point, columns are clearly inclined toward the impinging atom direction while the film is rough and underdense. We expect faceting (e.g. fig.2(a)) to produce columns with an inclination significantly different from the empirical tangent rule [10]. A quantitative study of the column angle with substrate inclination and atomic diffusivity is under investigation. Recent ADEPT [5] and molecular dynamics simulations [2] suggest a column formation mechanism initiated by kinetic roughening. Once hills and valleys have formed by kinetic roughening, the hills intercept more material than the valleys, and this results in an instability and has its ultimate outcome in a columnar structure. This process is dependent (i) on a large incident angle of impinging atoms, preventing the flux from reaching the bottom of any surface depressions [2], and (ii) on a low mobility of surface atoms which prevents the stabilizing influence of mass transport processes driven by surface free energy to

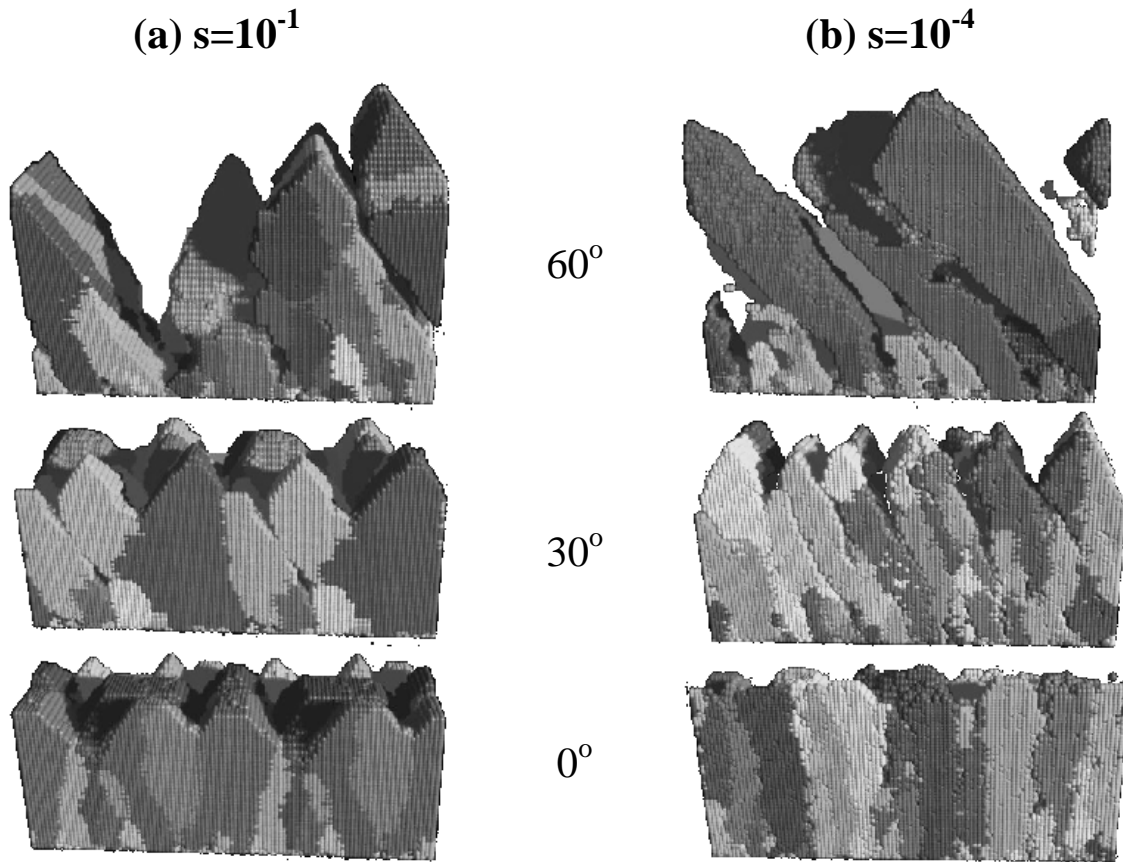


Figure 2: Morphology variation as a function of substrate inclination $\Psi_m = 0^\circ, 30^\circ, 60^\circ$, in the case of two different diffusivities $s=10^{-1}$ (a) and 10^{-4} (b). The different shade of grey represent different grain orientations

smooth the surface [5]. These two points lead to the shadowing of certain areas of the film for geometrical reasons.

The density and roughness of the polycrystalline films are plotted in Fig.3(a),(b). In Figure 3(a), we have plotted the film density as a function of substrate orientation. For this purpose we averaged the density of the film over 10 layers by calculating the ratio of the number of occupied sites to the total number of sites in the layer. We averaged these densities at a thickness where the film density is well established (between the 40th and 50th layers for {100} polycrystalline films and between the 20th and 30th layers for {111}). The roughness of the film as a function of substrate inclination is shown in Fig.3(b). The RMS roughness σ is calculated by measuring the surface height above each site in the substrate. In the case of overhangs, where the surface is multi-valued, the highest occupied lattice position is selected as the surface height. Figures 3(a),(b) show a dramatic difference in the behavior of the {100} and {111} textured films. The {111} textured film is dense and smooth for all values of the substrate inclination. On the contrary, for {100} texture, the film shows an abrupt decrease in the density and an increase in the roughness at large angles. This difference between the two surfaces is not determined by surface energy effects, since the specific surface energy is approximately the same in this case [9]. Instead, it is a result of differences in the kinetics of layer nucleation on the two surfaces. The potential energies of adatoms on {100} and {111} surfaces differ by about 0.3eV, with the {100} adatom having the lower energy. As a consequence, the concentration of adatoms on {100} is higher than on {111}, where adatoms tend to diffuse to neighboring surface orientations or to step edges [5]. This leads to faster nucleation on {100} and facilitates the growth in the vertical direction instead of the lateral growth needed to densify the film. This result can be

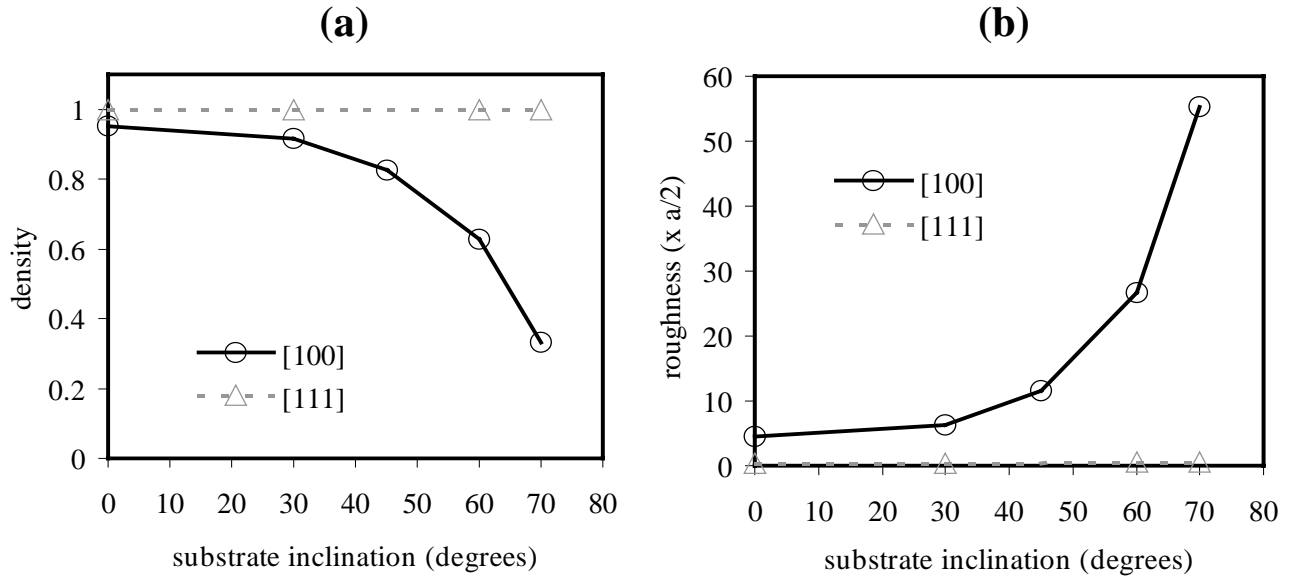


Figure 3: (a) Film density averaged over 10 layers as a function of the substrate inclination. (b) roughness of the film as a function of the substrate inclination. The roughness is in reduced units ($a/2$) where a is the lattice parameter in the fcc structure. The solid and dashed lines correspond to $\{100\}$ and $\{111\}$ textures, respectively.

generalized to other crystal lattice structures, where the surface orientation with the densest atomic packing has a high adatom potential energy, and the texture with this orientation produces the densest films.

3. Effect of reduced diffusivity

In this section we analyze the effect of diffusivity on the film morphology. For this purpose, we computed the film density in the case of a 60° substrate inclination, for $\{100\}$ and $\{111\}$ textured films. The results of these calculations are plotted in Figure 4. First, the $\{111\}$ textured films are denser than $\{100\}$ over the entire range of the diffusion coefficient ratio. But the films corresponding to the two textures approach approximately the same density for very low values of s , where the diffusion coefficients are so small that the two orientations have similar kinetics; for low s atoms stick close to the point where they land on the surface. Next, the $\{111\}$ textured films remain close to 100% density for a wide range of diffusivities, and the density decreases only for a diffusivity reduction greater than 10^4 . On the contrary, the $\{100\}$ textured films are seriously underdense for all values of s . The $\{100\}$ densities may be increased somewhat by including in the model the high energy Ar atoms reflected from the target onto the

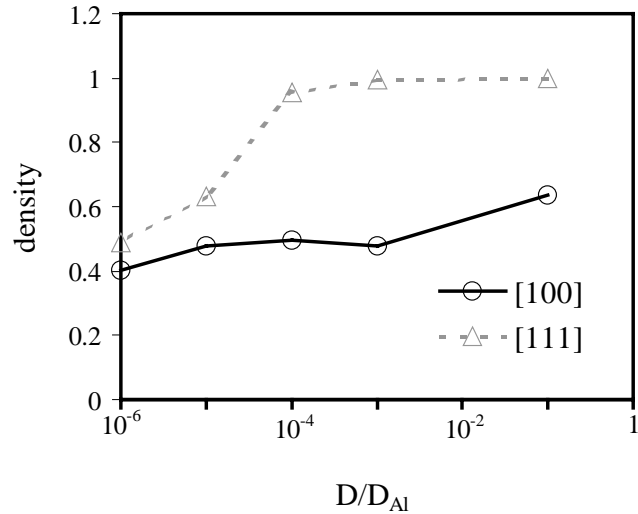


Figure 4: Density of the film averaged over 10 layers as a function of the substrate inclination in the 60° case. The solid and dashed lines correspond respectively to $\{100\}$ and $\{111\}$ textures.

film, since their momenta are directed toward the substrate, and would tend to knock out atoms from the higher sections of the film and to fill in the depressions. The inclusion of these energetic collisions with the film would also increase the surface diffusion rate, although the results of figure 4 show that the density is not very sensitive to the amount of surface diffusion. The insensitivity of the density and film morphology to the surface diffusion kinetics is of importance to the modeling effort, since it shows that just approximate values of the diffusion coefficient may already be sufficient to provide accurate film properties.

IV. CONCLUSIONS

We have presented Monte Carlo simulations of deposition onto inclined surfaces using conditions in accordance with magnetron sputtering experiments [4]. We have examined the effects of texture surface orientation, and of the surface diffusion coefficients on the density and roughness of the film morphology in a model material implemented with scaled Al parameters.

The most intriguing result of our studies is the dramatic effect of texture on the density and porosity of films. Films with texture corresponding to the low-index surface with the highest adatom potential energy were found to produce superior films for barrier layers. This is a result of the anisotropy of the growth kinetics, due to slow nucleation rates of new layers on these orientations. Grains with these orientations grow preferentially in the lateral directions, and this reduces shadowing effects, inhibits columnar growth, and produces dense, low porosity films.

Since the fact that we used Al with scaled parameters enables us to only do a qualitative comparison to experimental data of real barrier films (see Ref.[4]). As soon as the microscopic parameters to refractory materials (like Ti, TiN, etc) are available our model should allow a direct comparison between simulations and experiments. A deeper understanding of the above described phenomena, both through experiments and simulations, may lead to better understanding and control of texture formation, and thus ultimately to advances in thin film technology.

ACKNOWLEDGMENTS

This work was supported by a NSF/DARPA VIP contract through the University of Illinois and by U.S. Department of Energy, Office of Science, Laboratory Technology Division under contract DE-AC05-96OR22464 with Lockheed Martin Energy Research Corp. and contract DE-AC05-76OR00033 with Oak Ridge Associated Universities.

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