Monte Carlo modeling of thin film deposition: Factors that influence 3D islands.

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ABSTRACT

In this paper we discuss the use of atomisitc Monte Carlo simulations to predict film microstructure evolution. We discuss physical vapor deposition, and are primarily concerned with films that are formed by the nucleation and coalescence of 3D islands. Multi-scale modeling is used in the sense that information obtained from molecular dynamics [1] and first principles calculations [2] provide atomic interaction energies, surface and grain boundary properties and diffusion rates for use in the Monte Carlo model. In this paper, we discuss some fundamental issues associated with thin film formation, together with an assessment of the sensitivity of the film morphology to the deposition conditions and materials properties.

Keywords: ionized physical vapor deposition, island coalescence, nanocluster arrays.

1   INTRODUCTION

Modeling based on Monte Carlo (MC) simulations is employed to predict film microstructure evolution, e.g., texture development, intergranular porosity, and grain size distributions. The model can encompass the wide range of materials properties and morphologies that are exhibited by these films, and facilitate the selection of materials and processes that will optimize the film properties [3-5].

An advantage of atomistic simulations is the crystal lattice, which is implicit in the models, along with the associated anisotropies that control crystal morphology. For this reason, many important phenomena are present, without having use artifices to introduce anisotropic properties. These models exhibit faceting, variations in surface diffusion rates with orientation, and grain boundary grooving.   These are direct consequences the inter-atomic potential and the resulting interactions between atoms on the crystal lattice sites.

2   MODEL

We have developed a model that provides efficient simulations of materials having several different crystal lattices structures [3]. Monte Carlo (MC) models of crystal growth are usually based on the repetition of several basic events. This type of model has the advantage of fast computation, because of the simplicity of MC events, while including diffusion rates and other mechanisms based on more detailed modeling; i.e. molecular dynamics or first principles, calculations. Although the energetics and mechanisms employed in the MC model are not exact replicas of the more detailed models, the MC approach fills an important gap in the modeling hierarchy. It provides sufficient structural complexity to treat the surface geometry essential to a realistic description of the crystallization process; i.e., it includes the steps and islands (2D and 3D) necessary to model the essential features of surface roughening transitions, kinetic roughening, and nucleation. At the same time, it can treat processes that occur over the relatively large length and time scales of real deposition systems.

The simulation of sputter deposition proceeds by selecting one of two events, injecting a sputtered atom or selecting a film surface atom for a diffusion jump. The relative probabilities for selecting these events depend on the conditions being simulated. In particular, they depend on the ratio of the impingement frequency of sputtered atoms to the hop frequency of atoms at the surface. Sputtered atoms are initially placed at random positions in a plane above the substrate and are assigned velocities corresponding to ejection angles selected randomly using a cosine distribution. These "sputtered" atoms are moved along the chosen trajectories in small increments until they reach a lattice site having at least one occupied neighboring site. The atom is placed at this position, and a new event is selected. Surface diffusion hops are performed by selecting a surface atom from a book-keeping list by using a random selection scheme, and moving it to a neighboring empty site up to the third neighbor distance.   Atoms in different coordination sites on the surface are selected with different probabilities, based on the diffusion hop rate, as discussed above.

Our model also accounts for the atomic rearrangements in the film resulting from collisions with energetic particles, such as those produced by sputtering. These collisions can increase the density of refractory films that often grow with voids or columnar crystallites. They can also affect the shapes of 3D islands that are usually formed at low coverage when more labile materials are deposited onto foreign substrates.

3   SUBSTRATE DIFFUSION

The sputter deposition of the metals Al and Cu usually results in 3D island nucleation and, eventually, coalescence. The relatively high diffusivity of these materials permits the transport of deposited material within islands so as to reduce the total of the surface, interfacial, and bulk free energies. Voids and pinholes are formed as

the islands coalesce. Monte Carlo simulations combined with analytical modeling of island growth and coalescence allows us to predict the thicknesses at which island coalescence occurs, and thus the minimum thickness required to produce a continuous film.

Simulation results for films deposited on substrates with widely varying diffusivities are illustrated in Fig. 1. It is apparent from these results that low substrate diffusivities are desirable for the formation of a dense, singly connected film. The surface specific density of nucleation events is expected to vary as $(D_S)^{-1/3}$ when two Al atoms form a super-critical nucleus [6]. The grain densities at the substrate in these simulations follow this relationship closely, although the density deviates below this trend at very low diffusivities, because of the coalescence of islands in this regime.

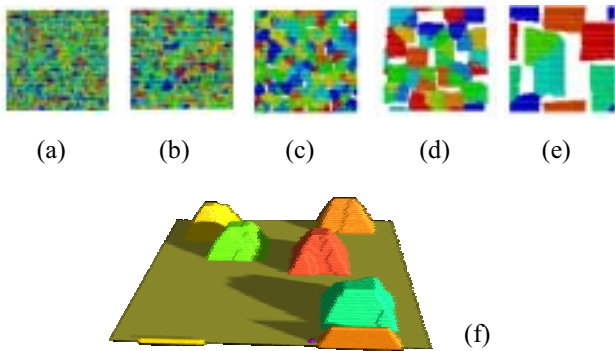(a)   (b)   (c)   (d)   (e)

(f)

Figure 1. Cross-sections through the base of films simulated by the MC program. 3D islands are formed because the film-substrate bonds are set to 70% of that for Al-Al. The different shades of gray indicate different grains, derived from different nucleation events, and the white areas are bare substrate regions. In this case Al is deposited onto substrates at T=200K, and with adatom diffusion coefficients D_S: (a) $D_S/D_{Al}=10^{-4}$, (b) $D_S/D_{Al}=10^{-2}$, (c) $D_S/D_{Al}=1$, (d) $D_S/D_{Al}=10^2$, (e) $D_S/D_{Al}=10^4$. The films illustrated correspond to (001) texture, the cross-section is 35nm by 35nm, and ten (001) layers were deposited. The island morphologies at $D_S/D_{Al}=10^2$ are shown in (f). Both (111) and (001) facets are present, although (111) predominate.

The coalescence of the islands can lead to a dense, singly connected film, but it is apparent from the results shown in Fig. 1 that the film thickness at which this occurs depends on the substrate diffusivity. Figure 2 shows the thickness at which 95% of the substrate sites are covered by the film. The dependence of the critical film thickness on D_S can be understood based on the $(D_S)^{-1/3}$ relationship for the density of nuclei in the layer, and the shapes of islands with different bond energies with the substrate. From Fig. 2 it is apparent that the strength of film-substrate

bond has a strong effect on the ability of the film to cover the substrate.
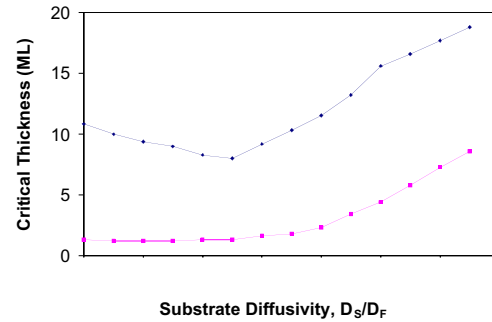

Figure 2. Thickness at which the film covers 95% of the substrate. The lower curve corresponds to a film-substrate bond energy which is 90% of the film-film bond, and for the upper curve it was reduced to 50%. Otherwise, the parameters are identical to those in Fig. 1.

Unfortunately, substrate diffusivities much below that of the film material are not achievable for most real materials. Weak bonding, which leads to de-wetting and 3D islands is usually accompanied by low diffusion barriers on the substrate. 3D islands are also frequently observed in the case of strong film-substrate bonding, where lattice mismatch between the substrate and the film materials causes strain in the film. Monolayer amounts of the film material will adhere to the substrate in a uniform film, because of the strong bonds. Subsequent layers do not have the advantage of these strong bonds, attach weakly to the underlying film, and often exhibit 3D islands. Furthermore, the effective substrate diffusivity is essentially that of the film material, because the atoms of the subsequent deposit are moving on the layer of film material.

An alternative method to improve substrate coverage is to reduce the self-diffusion rate on the surface of the film. Our simulations show that reductions in the temperature are highly effective in improving the substrate coverage. The increased island density facilitates coalescence. This effect is also confirmed by experiments, as illustrated for the case of Al deposited onto a Ti substrate in Fig. 3. At the highest temperature of 400°C, large 3D islands form on a bare substrate, whereas at 200°C the film seems to cover the substrate except for a few pinholes. It is sometimes useful to program the temperature during deposition to optimize the properties of the film. Initial deposition onto a cold substrate will provide percolation of the islands at an early stage, and then increasing the temperature can facilitate mass transport to fill any pinholes or defects in the film.

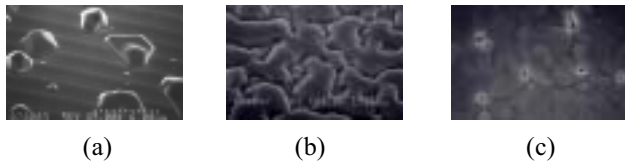<div style="text-align:center">(a)   (b)   (c)</div>

Figure 3. Scanning electron micrographs of Al sputtered onto Ti substrates: nucleation and coalescence of islands. (a) Average film thickness of 140nm, T=400°C, (b) film thickness of 80nm, T=300°C, (c) film thickness of 140nm, T=200°C. In (a) the width of the picture is 15μm, and in (b) and (c) it is 4 μm. .

4 ENERGETIC PARTICLE BOMBARDMENT

The control of energetic ions and neutrals has long been recognized as an important method to influence the film structure. Although these particles are in essential part of sputtering, the kinetic energies of the particles as they impinge on the substrate can be modified, most conveniently by controlling the Ar pressure in the sputtering chamber. The effect of energetic particles on the shapes of the 3D islands is one mechanism for possible improvement of film coverage. Re-sputtering of atoms from the 3D islands causes some reduction of the height to width ratio, and this effect will favor coalescence.

Our simulations show that the sputtered particles from an Al target, with a 400eV potential on the target, have rather low energies and only a small influence on the aspect ratios of the 3D islands. This is true even at low Ar pressures, where the mean free path of the sputtered particles is longer than the target-substrate distance. Higher energy Ar ions in the range of 50eV, for example, cause significant flattening of the islands due to the downward momentum of the ions, and increase the coalescence of islands. Thus, a facility to accelerate Ar ions toward the substrate should improve coverage. The energies of reflected Ar atoms depends on the mass of the target atoms, and since Al has a smaller mass than Ar, the energies are low.

An example of sputtering of a target with large atomic mass is shown in the MC simulations of Ta illustrated in Fig. 4. A different growth mechanism from island-merger operates in this system, since the extremely low mobility of Ta surface atoms does not allow the material to rearrange in such a way as to form 3D islands. In this case columnar growth is observed, with low density films and void regions traversing the film [5].

The influence of the particles coming from the target is considerable in this case. The density of the film is increased by the re-sputtering events, and the number of pores is dramatically reduced.
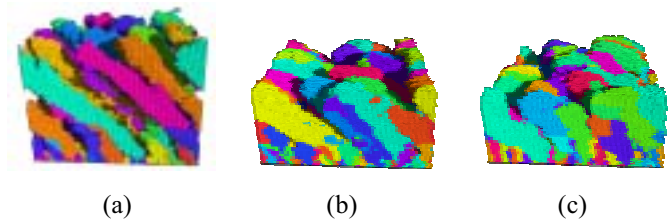

<div style="text-align:center">(a)   (b)   (c)</div>

Figure 4. MC simulations of sputtered Ta films at T=30°C (a) Film deposited without energetic particles. There is some energy imparted to the impinging Ta atoms due to attractive interatomic forces, and this amounts to about 6eV/atom. (b) film formed with Ta atoms with an initial kinetic energy that averages 10eV. (c) film produced with both 10eV Ta atoms and 50eV reflected Ar atoms. The energies for the Ta and Ar atoms were obtained using TRIM simulations of the 400eV Ar ion impinging on the Ta target. .

5 CONCLUSION

The MC simulations provide a method to assess different techniques to improve film quality, and to select deposition conditions and properties that are likely to have a significant impact on the films. Our understanding of the film deposition process is advanced by the ability to mdify individual parameters or external conditions.

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