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Dislocation Nucleation and Propagation During Thin Film Deposition Under Tension

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Abstract: Using molecular dynamics method, we study the nucleation of dislocations and their subsequent propagation during the deposition of tungsten thin films under tension. Aiming to reveal the generic mechanisms of dislocation nucleation during the deposition of polycrystalline thin films, the case of tungsten on a substrate of the same material is considered. The substrate is under uniaxial tension along the $[111]$ direction, with the thermodynamically favored $(0\bar{1}1)$ surface being horizontal. The simulation results indicate that the nucleation starts with a surface step, where a surface atom is pressed into the film along the $[11\bar{1}]$ direction. This process leads to the generation of a half dislocation loop of Burgers vector $\frac{1}{2}[11\bar{1}]$ along the (112) plane, which is about 73^o from the horizontal plane. The dislocation propagates along the $[\bar{3}11]$ direction. As a result of the dislocation nucleation, a sharp surface step is eliminated.

keyword: dislocation nucleation, dislocation propagation, thin films, and molecular dynamics.

1 Introduction

Thin films, in particular polycrystalline thin films, are almost ubiquitous to modern engineering, because of their low defect concentration ease of processing. Together with the scientific curiosity, the applications have led to intensive investigation of polycrystalline thin film processing. Polycrystalline aluminum or copper thin films in the form of metal lines, for example, form good interconnections of transistors in integrated circuits. It is well known that grain boundaries and dislocations are open paths of fast diffusion, which is responsible for the electromigration failure [Mahajan and Sree Harsha(1998)]. The fast diffusion can be reduced or eliminated if the microstructure, in particular the texture, in the aluminum thin films is well controlled. Multiscale materials modeling has served an effective tool in understanding the evolution of grains and the dynamics of dislocations. Based on this approach, an atomistic simulator ADEPT [Huang, Gilmer, and Diaz de la Rubia (1998); Huang and Gilmer(1999); Gilmer, Huang, Diaz de la Rubia, Torre and Baumann(1999); Huang and Gilmer(2001a); Huang and Gilmer(2001b); Baumann, Chopp, Diaz de la Rubia, Gilmer, Greene, Huang, Kodambak, O'Sullivan, and Petrov(2001)] has been developed. However, the effects of dislocation in polycrystalline thin films have not been considered in ADEPT, particularly at the atomistic level.

The nucleation of dislocations at surfaces and their subsequent propagation into the polycrystalline thin films bears some similarities to the epitaxial thin films. For the polycrystalline thin films, dislocations nucleate during film growth, as if the films are deposited on a stressed substrate that is made of the same materials. During epitaxy, on the other hand, dislocations nucleate when films are deposited on a substrate of either the same or different materials; generally stress exists in hetero-epitaxy. In view of the common features existing between the dislocation dynamics in the two types of thin films, it is worthwhile mentioning studies in the case of epitaxial thin films.

The theoretical investigation of dislocation dynamics in epitaxial thin films has gone through two stages: analytical formulation and atomistic simulation. The analytical formulation of dislocations in thin films started with Frank and van der Merwe(1949). In this theory, a simple model of one-dimensional springs on a periodically modulated substrate was used. The two competing factors are the strain energy of thin films due to the film-substrate mismatching, and the extra energy associated with the strain field of dislocations. A dislocation is nucleated when its presence leads to the reduction of the total energy. For a given film-substrate system, this critical condition translates into a critical thickness of the

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thin film, at which the strain energy due to film-substrate mismatching is so large that the total energy decreases with the generation of a dislocation. The formulation is based on energy minimization among possible configurations of thin films. However, it is not clear how the dislocations nucleate and how they propagate. In an attempt to address the issue of dislocation nucleation and propagation, Matthews and Blakeslee(1974) assumed a semi-circular loop on the film surface as the nucleus of dislocation. They derived a critical radius of the loop, beyond which the dislocation will propagate into the film and below which the dislocation will be absorbed by the surface. Based on this nucleation mechanism and extension of threading dislocations, more factors are considered in his later calculation [Matthews(1975)].

Although simplistic, the analytical theory [Frank and van der Merwe(1949)] qualitatively predicted the critical thickness when dislocations start to be present in thin films. Efforts were made to achieve more quantitative predictions by considering some characteristics of the dislocations - such as their screw or edge nature [People and Bean(1985)]. Still, these theories suffer from two intrinsic drawbacks. First, the thermodynamic equilibrium assumption ignores kinetic effects during thin film deposition. However, surfaces of thin films are seldom in equilibrium during deposition and they are generally rough, and the surface roughness will affect the nucleation of dislocations. Second, the elastic theory ignores the effects of the crystal structure, the importance of which is well known today [Hirth and Lothe(1982); Campbell, Foiles, Huang, Hughes, King, Lassila, Nikkel, Diaz de la Rubia, Shu and Smshlyaev(1998)].

Atomistic simulations are immune from these two drawbacks of the continuum theories. However, most of the atomistic simulations have been two-dimensional (2D) or quasi two-dimensional, because of the limited computational capacity available. In a 2D molecular dynamics study, [Dong, Schnitker, Smith, and Srolovitz(1998)] found that the critical thickness of dislocation nucleation depends not only on the magnitude, but also the sign of mismatching. Under compression, a dislocation was found to nucleate by squeezing out one atom near a depression. Under tension, on the other hand, the first dislocation was found to nucleate by relative shift of two neighboring columns of atoms.

Recently, we studied dislocation nucleation and propagation of tungsten thin films deposited on a compressed substrate [Liu, Shi, Woo and Huang(2001)], using three-dimensional molecular dynamics. The results confirmed that the dislocation nucleus is indeed a half-loop. Further, it was demonstrated that both glissile and sessile dislocations could be nucleated, although one atom was always ejected at the nucleation point. Similar process under tension has yet to be studied, and is the subject of this paper. The simulation method is described in Section 2, and simulation results are presented with discussions in Section 3. Finally, we summarize the results in Section 4.

2 Simulation Method

We use the classical molecular dynamics (MD) method to investigate the nucleation and propagation of dislocations during thin film deposition. Tungsten, a body-center-cubic metal, is chosen as a representative material, because (1) it is almost elastically isotropic, and (2) it does not have Shockley partial dislocations. These two features enable a simpler analysis at the start. Ackland's potential in the Finnis-Sinclair form [Finnis and Sinclair(1984), Ackland and Thetford(1987)] is used in our calculations. The tungsten atoms are deposited on a stressed tungsten substrate, with the $(0\bar{1}1)$ surface horizontal. This surface orientation is thermodynamically the most preferred. The two horizontal directions are $[111]$ and $[\bar{2}11]$; the first direction is along one of the Burgers vectors.

To minimize the computational effort, dimensions of the simulation cell are set to be 47 Å, 125 Å and 14 Å along the $[111]$, $[\bar{2}11]$ and $[0\bar{1}1]$ directions, respectively, in our simulations. According to our previous studies [Liu, Shi, Woo and Huang(2001)], size effects are unimportant when a simulation cell of this dimension is used. The periodic boundary condition is applied along the two horizontal directions, leaving two free surfaces along the $[0\bar{1}1]$ direction. Atoms within the bottom 7 Å (that is 1.5 times the cutoff distance of the interatomic potential) of the substrate are fixed to their BCC lattice positions, to mimic the bulk of one grain in polycrystalline thin films. Atoms within the top 7 Å of the substrate are subjected to random frictional forces, and effectively serve as a thermal bath [Nyberg and Schlick(1991)]. A uniaxial tensile stress of 13 GPa is applied to the substrate along the $[111]$ direction. This stress is represented by the corresponding strain, and the two are related by using the Parrinelo-Rahman algorithm [Parrinello and

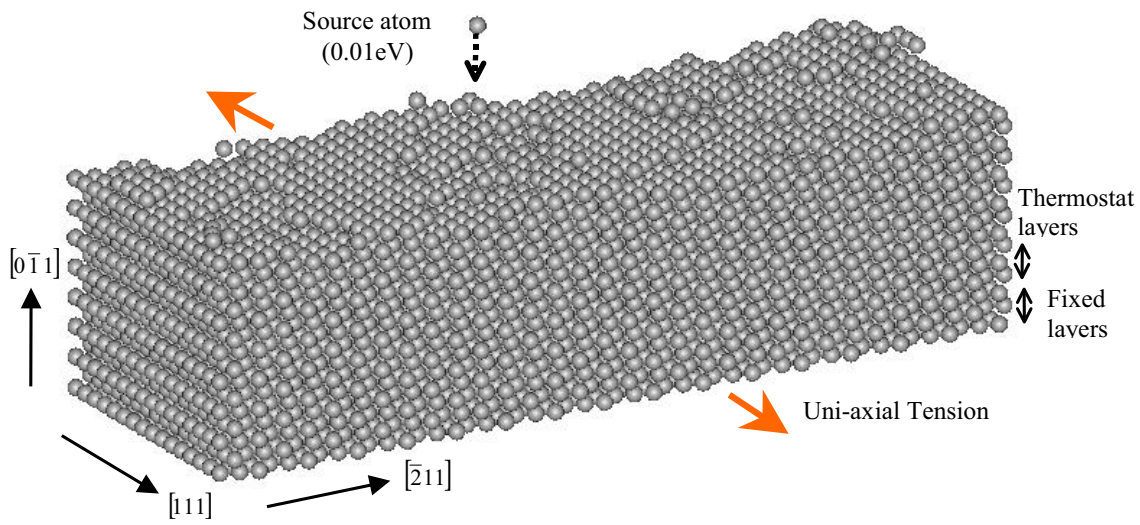

Figure 1 : A typical simulation cell, with directions of trajectory of deposited atoms and applied stress indicated.

Rahman(1981)] and their relation is given in [Liu, Shi, Woo and Huang(2001)]; details of the correlation are presented in the following Section.

The stressed substrate is bombarded by tungsten atoms, which carry 0.01 eV of kinetic energy and head toward the substrate vertically. This particle source essentially simulates a 100% collimated physical vapor deposition process. The small kinetic energy is chosen to minimize the surface over-heating. The simulation cell, after deposition of 14 atomic layers, is shown in Figure 1.

It is worth mentioning that the film in Figure 1 is deposited at a deposition rate of 1 meter per second, in contrast to the typical value of 1 micron per minute. This high deposition rate is a result of the short time that can be reached in molecular dynamics simulations. The most serious consequence of this high deposition rate is the lack of diffusion during film deposition. As compensation, we keep the substrate at a very high temperature, 2500K - the melting temperature of the tungsten is about 3680K. The temperature is first brought to 2500K using the Langevin force [Nyberg and Schlick(1991)], which is then removed to keep the simulation cell a micro-canonical system [Liu, Woo and Huang(1999)]. As the results in the following Section indicate, the amount of atomic diffusion in the simulations is equivalent to that during an experiment at 340K under 1 micron per minute [Liu, Woo, and Huang(in preparation)].

During each simulation, positions of deposited atoms are recorded periodically for analysis. In order to facilitate the visualization, the thermal fluctuation of the positions is eliminated by quenching the temperature of the atoms to 0.1 K. An “atomic movie” is prepared using a sequence of the atomic positions to identify dislocations. Both potential energy and the von Mises atomic shear stress are used to differentiate atoms near a dislocation from the rest. Because high potential energy and shear stress might exist on atoms that are far away from any dislocations, we take this “atomic movie” as guidance only. The dislocation position and configuration are uniquely defined using atomic positions. The dislocation Burgers vector is defined by SF/RH Burgers circuits [Bilby, Bullough and Smith(1955)].

3 Simulation Results

To estimate whether atoms are allowed to have sufficient diffusion during our deposition simulation, we have studied the diffusion of an atom on the $(0\bar{1}1)$ flat surface. Following Soneda and Diaz de la Rubia(1998), we divided the simulated diffusion process into N sub-processes and the diffusion coefficients are found to converge when each sub-process on average contains at least one or two fundamental jumps [Liu, Woo, and Huang(1999)]. Under a deposition rate of 1 meter per second at 2500K as used in our simulation, the distance that a deposited atom diffuses before burial is about 3 nm. Such a diffusion distance is realized in deposition at 340K under realistic deposition rate - 1 micron per minute, according to the calculated adatom diffusion co-

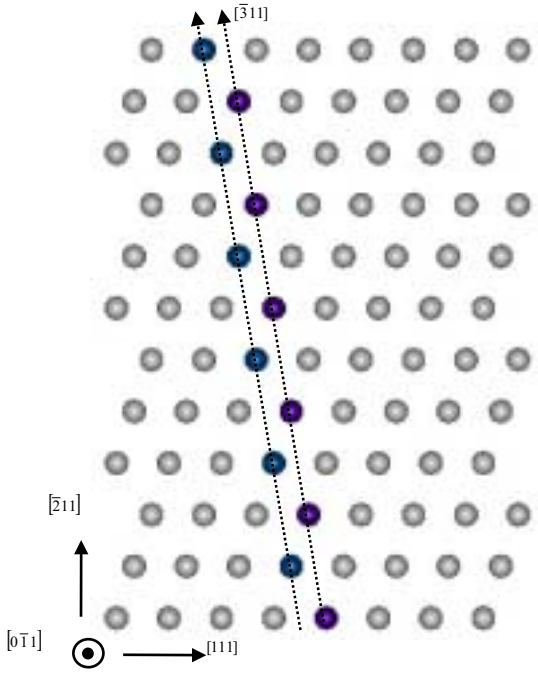

Figure 2 : Atoms on the $(0\bar{1}1)$ horizontal surface - only part of the simulation cell is shown for clarity. The dark color indicates atoms squeezed into the film along the $[11\bar{1}]$ direction. The two dotted lines indicate directions of dislocation extension, and that the extension follows a zig-zap path between the two parallel lines. The circle with a solid dot indicates the direction point out of the paper.

efficient; the burial time is taken to be that of growing one monolayer [Liu, Shi, Woo and Huang(2001); Liu, Woo, Huang].

Using the setup of the aforementioned substrate and temperature, and under the uniaxial tension of 13GPa, a dislocation nucleates when 11-13 atomic layers of film are deposited. Along the direction of tension, the magnitude of strain is 0.055. The nucleation of dislocation is first identified using the "atomic movie", and then confirmed by configuration analyses. A dislocation nucleates near a step, and propagates into the film as a half-loop, on the two neighboring (112) planes as shown in Figure 2. One of the $(1\bar{1}0)$ layers is shown in Figure 3a-c; the nucleation starts at the encircled atom. First a step is formed on the surface, as is shown in Figure 3a; the step would also show up in parallel $(1\bar{1}0)$ layers. The atom near the step, as indicated by the circle, moves into the film along the $[11\bar{1}]$ direction, as shown in Figure 3b. Afterwards, the atom at the upper edge of the step relaxes down to eliminate the sharp step, as shown in Figure 3c; the energy is likely minimized by the elimination of the step. This nucleus of dislocation propagates along the $[\bar{3}11]$ line. This process generates a dislocation, and the corresponding Burgers vector has a component of $[\bar{1}\bar{1}1]$ in the

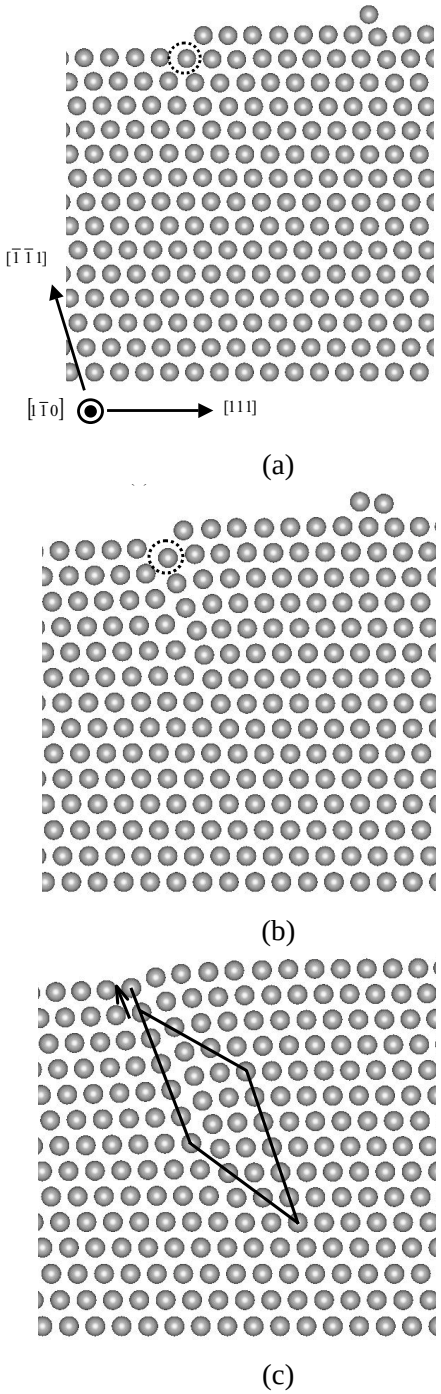

Figure 3 : Atomic positions in one of the $(1\bar{1}0)$ atomic layers containing the dislocation nucleus at, (a) 1873 ps, (b) 1883 ps, and (c) 1893 ps after the deposition starts. The circle with a solid dot indicates the direction point out of the paper.

$(1\bar{1}0)$ plane; as will be seen later, this component is also the whole Burgers vector.

To identify the geometry of this dislocation, atoms in seven (112) layers neighboring the dislocations are projected along the $[111]$ direction, as shown in Figure 4. A small half-loop is nucleated in Figure 4a, and it extends by propagation to a near circular half-loop in Figure 4b.

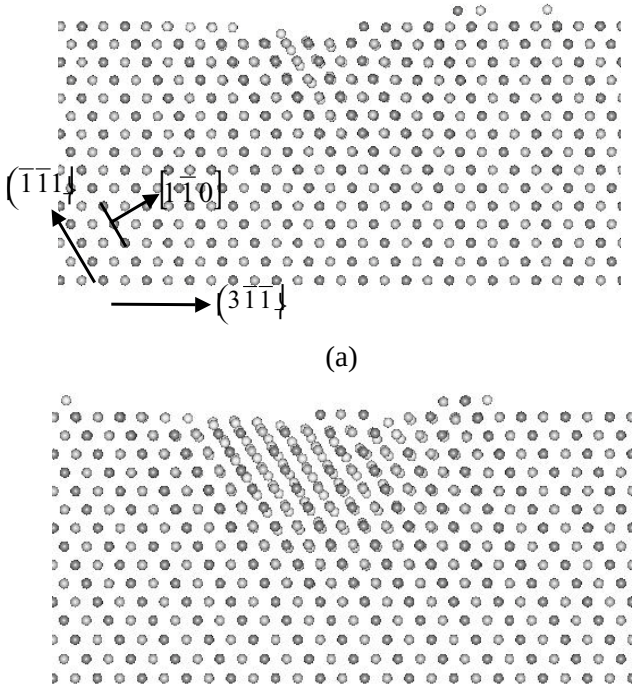

Figure 4 : Projection of atomic positions in seven (112) layers neighboring the dislocation, along the $[111]$ direction - only part of the simulation cell is shown for clarity. The snapshot is taken at (a) 1883 ps, and (b) 1889 ps after the deposition starts. The grey scale of atoms indicates the depth of various (112) layers. The normal vector of the (112) plane is also indicated by an arrow in (a).

In another simulation, similar phenomena are observed. In this case, the step is reversed, favoring nucleation of a dislocation on the (121) plane. The nucleation starts by squeezing one atom along the $[\bar{1}1\bar{1}]$ direction. Both the (121) plane and the $[\bar{1}1\bar{1}]$ direction are symmetry images of the (112) plane and the $[11\bar{1}]$ direction when the step is reversed. It is worth mentioning, the two glide planes, (112) and (121), also have the largest Schmid factor in the simulated stress condition. The (211) plane is the other one that has the largest Schmid factor. However, the corresponding $\langle 111 \rangle$ direction is the $[\bar{1}11]$, which is in the horizontal $(0\bar{1}1)$. If an atom was to squeeze in along the $[\bar{1}11]$ direction, the tensile stress under the surface is not released. As a result, nucleation of the corresponding dislocation does not minimize the total energy. This scenario is consistent with the fact that no dislocation on the (211) plane has been observed; validation of the hypothesis using energy calculation is under way.

4 Conclusions

Using the molecular dynamics method, we have studied the dislocation nucleation and propagation during deposition of tungsten thin films under a uniaxial tension. Our results show that a dislocation nucleates near a surface step and propagates in four steps:

1. First, one atom near a surface step is squeezed into the film along a $\langle 111 \rangle$ direction.
2. The atom at the nearby upper corner of the surface step follows the first atom to move down, and resulting in the elimination of the surface at one atomic layer.
3. The insertion of atoms downward extends along a $\langle 113 \rangle$ line on the horizontal surface.
4. The dislocation nucleus also expands by propagating towards the film-substrate interface.

The dislocation nucleation is found to occur on two $\{112\}$ planes with the largest Schmid factor. Dislocation on one of these planes is not observed; it is probably due to the fact that nucleation of a dislocation on this plane does not minimize the total energy.

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