

# Topographic study of sputter-deposited film with different process parameters

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In this study, molecular dynamics simulation is employed to investigate the surface topography of thin films produced by the sputtering process for different parameters such as substrate temperature, incident energy, and incident angle. Interface width is used to quantify the quality of the deposited film. The Morse potential is used to model the atomic interaction between atoms. From the results of this study, it is found that for lower substrate temperature, lower incident energy, and larger incident angle, the growing film structure tends toward a three-dimensional columnar structure, and a rougher film is produced. Conversely, for higher substrate temperature, higher incident energy, and smaller incident angle, the growing film structure tends toward a two-dimensional (Frank–van der Merwe) quasi-layer-by-layer structure, and a smoother film is produced. Finally, average surface kinetic energy is found to be an important factor in determining the surface properties produced in the process. Generally, the produced film is smoother when the average surface kinetic energy is larger. © 2001 American Institute of Physics. [DOI: 10.1063/1.1371007]

## I. INTRODUCTION

Thin films with high surface quality are applied widely in advanced technologies such as very large scale integrated circuits,<sup>1</sup> optical films,<sup>1,2</sup> and superconductors.<sup>3,4</sup> In the fabrication of integrated circuits, the wafer surface must be smooth enough to allow high-density microcircuits.<sup>1</sup> In optical components, smooth topography is necessary to reduce scattered light. The surface properties of the multilayer junction of a superconductor significantly affect the superconducting properties. Furthermore, the surface topography even influences the magnetic properties and Curie temperature of a superconducting film.<sup>5,6</sup> Thus, film smoothness is a serious topic of study and a variety of experimental methods have been used to investigate surface topography. Previously utilized methods include atomic force microscopy, scanning tunneling microscopy, scanning electron microscopy, stylus measurements, x-ray scattering, and optical scattering.<sup>1–4,7–9</sup> However, these experimental methods cannot observe the transient morphologies of growing films. As a consequence, understanding the formation of the different transient morphologies must be investigated with the aid of simulation.

To date, Monte–Carlo<sup>10–13</sup> and molecular dynamics (MD) analytic methodologies have been used to simulate the deposition process. Evans *et al.*<sup>14</sup> performed MD simulations in which interface width, defined as the root mean square roughness, was used to evaluate film surface quality. Their

study reported the simulated topographies for different deposition models which included random immobile deposition, simplified continuous downward funneling, discretized downward funneling, and one-hop transient mobility. A hard-sphere potential model was used in their study. Interaction forces between atoms were assumed to begin only when atoms collided, and forces were assumed discontinuous. The repulsive and attractive forces of metallic bonds and also the effects of van der Waals forces were ignored. Further, the substrate temperature was assumed to remain constant at 0 K. Thus, the impact energy of an incident atom was completely absorbed by the substrate atoms. As a consequence, the movement of an incident atom on the film surface could not be simulated accurately in their study. Srolovitz and co-workers used two-dimensional MD simulation to investigate the effect of working parameters including incident energy,<sup>15,16</sup> substrate temperature,<sup>15,16</sup> and incident angle.<sup>16</sup> Their simulation results showed the void volume fraction, and surface roughness of the film structure increased as the substrate temperature and incident energy decreased. Furthermore, their study included incident angle self-shadowing and showed that film density decreased very slowly when the incident angle was lower than 45°, but the film density decreased very rapidly when the incident angle exceeded 45°. Similarly, Zhou and co-workers also used two-dimensional MD simulation to investigate vacancy concentration with three working parameters, substrate temperature, incident energy, and incident angle.<sup>17</sup> However, the two-dimensional simulation can not adequately reflect the physical model in real deposition process. Kelchner and DePristo also studied the surface topographies for low energy cluster deposition at a substrate temperature of 0 K.<sup>18</sup> The discussed issue was

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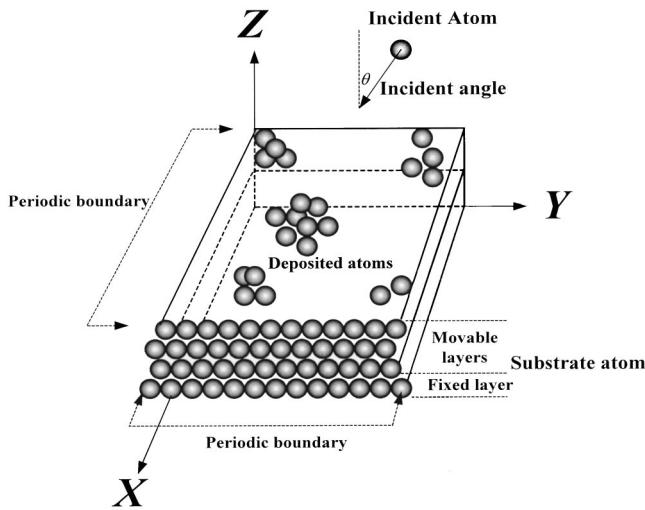


FIG. 1. Simulation model of the deposition process.

mainly the influence of different clusters, consisting of different numbers of atoms, on the formation of different surface topographies. The effect of substrate temperature as well as the other working parameters was not analyzed in their studies.

Zhou and Wadley<sup>19</sup> used three-dimensional MD simulation to investigate the effects of incident angles, with special focus on atomic reflection and resputtering. Surface damage was analyzed, but film structures were not considered. In a related study, Robbemond and Thijsse<sup>20</sup> used MD to simulate the topography of film produced by the evaporation process with a low incident energy of 0.17 eV. Only two specific incident angles were considered in their study. Was and co-workers<sup>21</sup> adopted a two-dimensional MD model to investigate the surface roughness induced by hillocks. Iizuka and Hoshida<sup>22</sup> utilized Morse potential to simulate growing film morphologies at two specific substrate temperatures. In their simulation, incident atoms arrived randomly at angles ranging from 0° to 60°. However, their simulated substrate contained only 168 atoms per plane. With a substrate this small, the incident angle can be considered the same for all incident atoms.

To allow a deeper understanding of the growth mechanism of thin films, we employ three-dimensional MD simulation to create a more accurate model of deposition conditions, thereby allowing three-dimensional atomic movement to be taken into account. Both topography and surface roughness of the growing film are investigated for different working parameters, including substrate temperature, incident energy, and incident angle.

## II. SIMULATION MODEL

The simulation model consists of three parts: the sputtering model, the model of atomic interaction, and the roughness calculation.

The formation of a three-dimensional thin film produced in a deposition process under sputtering process conditions is modeled as shown in Fig. 1. In this study, both substrate and incident atoms are Cu, with the substrate being assumed a

TABLE I. Morse potential parameters used in the simulation.

| Interaction pair | $D$ (eV) | $\alpha$ (Å <sup>-1</sup> ) | $r_o$ (Å) |
|------------------|----------|-----------------------------|-----------|
| Cu–Cu            | 0.3429   | 1.3588                      | 2.6260    |

perfect crystal. The velocity of an incident atom is calculated from the incident energy assumed in the simulation and the following:

$$V_{\text{atom}} = \sqrt{\frac{2 \times E_{\text{atom}}}{m}}, \quad (1)$$

where  $E_{\text{atom}}$  represents the incident energy and  $m$  is the atomic mass. In order to more closely simulate conditions of the real deposition process, incident atoms are periodically and individually generated at random locations in the  $x$ – $y$  plane. Furthermore, all incident atoms are sputtered at a distance which is much larger than the truncated distance of the atom from the substrate. This prevents initial interaction of the incident atoms with the substrate atoms. In our simulation, each incident atom is located at a vertical distance six times the truncated distance from the substrate, whereas the truncated distance is 2.5 (6.565 Å) in the dimensionless Morse potential unit adopted herein.

The substrate is modeled by a four-layered structure with a total of 288 atoms in each layer. To prevent the substrate atoms from shifting, the bottom layer is fixed, so the energy transfer from the incident atoms to the substrate is assumed to occur only in the upper three layers. Moreover, the substrate is assumed to be an ideal heat sink, resulting in a constant substrate temperature. The velocities of the atoms in the upper three layers follow the Maxwell–Boltzmann distribution, which is rescaled at each time step according to the substrate temperature in the simulation.<sup>23</sup> Periodic boundary conditions are applied in the  $x$  and  $y$  directions, with an open boundary in the  $z$  direction. A total of 1440 atoms, being equal to 5 ML, is deposited on the substrate for each simulation.

The Morse potential is adopted herein and used to determine the interactive force among atoms. This model is a representative potential of the pair type, with a simple functional form shown below:<sup>24</sup>

$$\phi(r_{ij}) = D \{ \exp\{-2\alpha(r_{ij} - r_o)\} - 2 \exp\{-\alpha(r_{ij} - r_o)\} \}, \quad (2)$$

where  $r_{ij}$ ,  $r_o$ ,  $D$ , and  $\alpha$  correspond to the distance between atoms  $i$  and  $j$ , the nearest atomic distance at equilibrium, the cohesion energy and the value fitted to the bulk elastic module,<sup>25</sup> respectively. The Morse potential parameters related to Cu used in this study are listed in Table I.<sup>24</sup>

The interface width  $W$ ,<sup>14–16</sup> defined as the root mean square roughness of the surface atoms on the exposed layers, is used to quantify the variation of surface roughness and is calculated by the following equation:

$$W^2 = \sum_{j=0}^{j=\infty} (Z_j - \bar{Z})^2 N_j, \quad (3)$$

TABLE II. Morse potential dimensionless units used in the simulation.

| Unit            |                       | Dimensionless unit                               |
|-----------------|-----------------------|--------------------------------------------------|
| Length          | $r$ (Å)               | $r^* = r/r_o$                                    |
| Time step       | $dt$ (s)              | $dt^* = dt \cdot \sqrt{\frac{D}{m \cdot r_o^2}}$ |
| Temperature     | $T$ (K)               | $T^* = T \cdot \frac{k}{D}$                      |
| Energy          | $E$ (eV)              | $E^* = E/D$                                      |
| Interface width | $W$ (Å <sup>2</sup> ) | $W^* = \frac{W}{r_o^2}$                          |

where  $N_j$  is the net number of exposed atoms which are stopped or migrating in layer  $j$  at a certain time step,  $j=0$  corresponds to the top substrate layer,  $\bar{Z}$  represents the mean height of the film surface, and  $Z_j$  the height of the exposed atoms. Gear’s predictor–corrector algorithm<sup>26</sup> is used to calculate the trajectories of atoms in the simulation. All units used in the simulation are dimensionless and listed in Table II, where  $m$  and  $k$  correspond to the mass of the atom and Boltzmann’s constant, respectively.

III. RESULTS AND DISCUSSION

Discussions are presented in the following three subsections, each subsection treating the effects of one of the three working parameters, substrate temperature, incident energy, and incident angle.

A. Substrate temperature

The substrate temperatures simulated in Figs. 2 and 3 are 300, 600 and 1000 K, and are applied to the upper three layers of the substrate. The other conditions remain the same, these conditions being zero incident angle, 0.5 eV incident energy, and 0.1 atom/ps deposition rate. In Fig. 2, the hori-

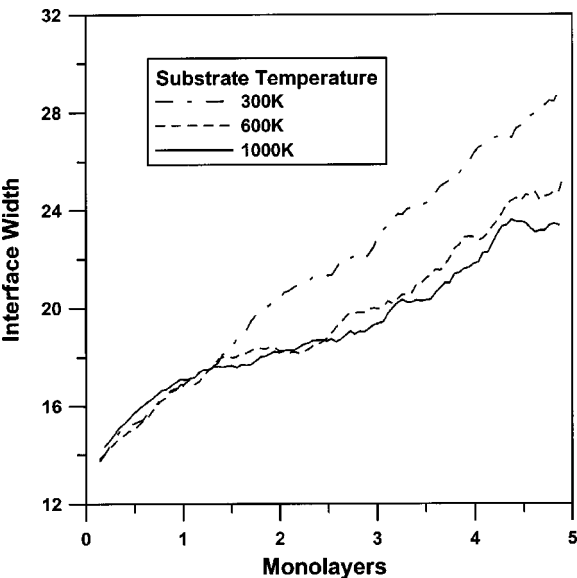


FIG. 2. Interface width variation at substrate temperatures of 300, 600, and 1000 K, and the other deposition conditions remain the same: zero incident angle, 0.5 eV incident energy, and 0.1 atom/ps deposition rate.

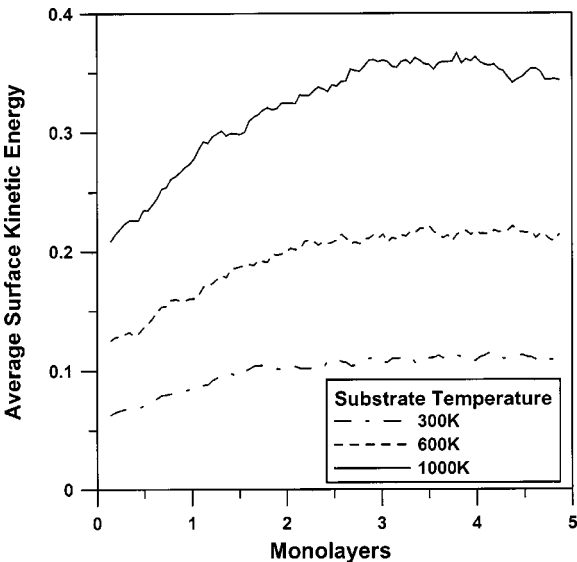


FIG. 3. Average surface kinetic energy at substrate temperatures of 300, 600, and 1000 K, and the other deposition conditions remain the same: zero incident angle, 0.5 eV incident energy, and 0.1 atom/ps deposition rate.

zontal axis represents the number of deposited layers as calculated from the number of the deposited atoms. It is observed that the topography of the substrate at 1000 K is smoother than at 300 and 600 K.

Figure 3 shows that the average surface kinetic energy is higher when the substrate temperature is higher. From the microscale viewpoint, temperature represents the average kinetic energy of atoms. At higher temperature, the atoms of the substrate act as less of a temperature sink, so the film surface remains at higher temperature. In other words, the surface atoms possess greater kinetic energy, resulting in the atoms moving on the surface with correspondingly greater mobility and increased migration distance. Isolated clumping of incident atoms is discouraged because of the increased mobility and greater energy exchange between atoms moving on the surface. The elevation of mobility is helpful for producing smoother surfaces.

The morphological formations during deposition at 1000 and 300 K are shown in Figs. 4 and 5, respectively. In Fig. 4, two-dimensional (Frank–van der Merwe) quasi-layer-by-layer growth is obtained at 1000 K. However, in Fig. 5, three-dimensional columnar growth is found at the lower temperature of 300 K. These two results are consistent with Movchan’s<sup>27</sup> and Thornton’s<sup>28</sup> studies, and indicate that higher substrate temperature promotes the growth of a quasi-layer-by-layer film structure.

In actual sputtering experiments, the energy transferred from incident atoms to the substrate is absorbed by the large number of substrate atoms. In addition, the substrate temperature is adjusted and maintained by a power supply controller in the substrate. Thus, the temperature of the substrate can be considered to be constant in actual sputtering deposition experiments.<sup>29,30</sup> In our MD simulations, therefore, the velocities of the upper three atomic layers of the substrate are rescaled so as to maintain a constant temperature at each time step in order to better model the energy transfer from

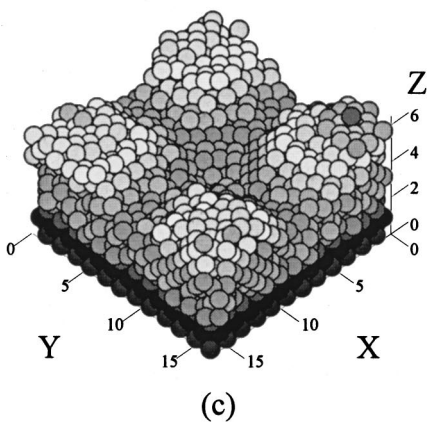
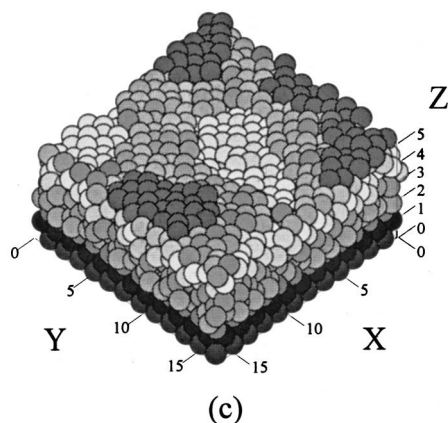
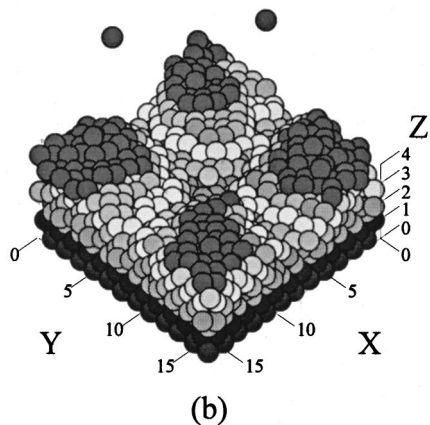
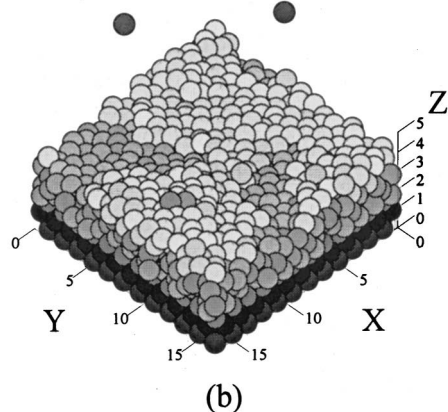
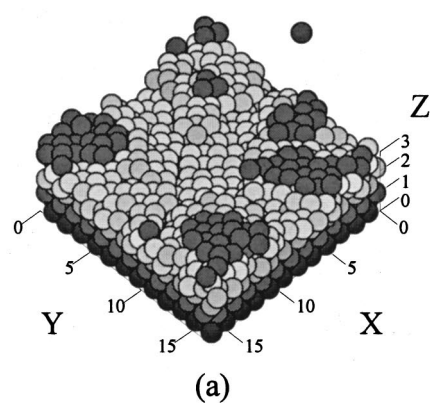
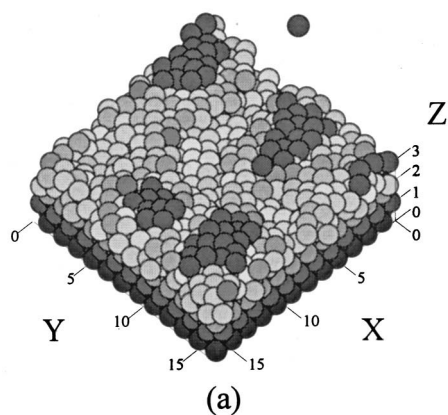


FIG. 4. Morphology at the substrate temperature of 1000 K with zero incident angle, 0.5 eV incident energy, and 0.1 atom/ps deposition rate (a) 1 ML, (b) 3 ML, and (c) 5 ML.

incident atoms to substrate.<sup>23</sup> In keeping with experimental observations, setting a higher rescaling temperature lowers the amount of energy transferred from film to substrate. The surface atoms thereby possess higher average surface kinetic energy, which improves the diffusion ability and final surface smoothness.

### B. Incident energy

Figure 6 shows the interface width variation for incident energies of 0.5, 2, and 10 eV. The other sputtering conditions remain the same: incident angle of zero, substrate tempera-

FIG. 5. Morphology at substrate temperature of 300 K with zero incident angle, 0.5 eV incident energy, and 0.1 atom/ps deposition rate (a) 1 ML, (b) 3 ML, and (c) 5 ML.

ture of 300 K, and deposition rate of 0.1 atom/ps. The results indicate that the higher incident energy is, the smoother film surface will be. Similarly, the atoms moving on the surface can migrate further, which is helpful for producing a smoother film surface. The energy transferred to the surface atoms increases as the incident energy increases. This can be seen in Fig. 7, where the average surface kinetic energy is higher when the incident energy is higher.

The simulated morphological formation during deposition for an incident energy of 10 eV is shown in Fig. 8. The result shows a two-dimensional (Frank-van der Merwe) quasi-layer-by-layer growth. A three-dimensional columnar growth structure is found for an incident energy of 0.5 eV, as

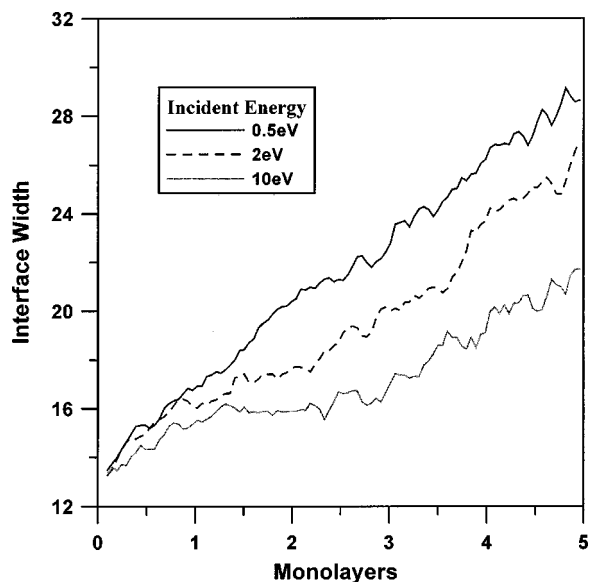


FIG. 6. Interface width variation at incident energies of 0.5, 2, and 10 eV, and the other conditions remain the same: zero incident angle, 300 K substrate temperature, and 0.1 atom/ps deposition rate.

shown in Fig. 5. These two results indicate that higher incident energy helps promote the growth of a quasi-layer-by-layer film structure. Confirming evidence is supplied by Qiang's study,<sup>31</sup> which likewise found that larger incident energy produces smoother film surface.

### C. Incident angle

The interface width variation at incident angles of  $0^\circ$ ,  $25^\circ$ ,  $45^\circ$ , and  $75^\circ$  is shown in Fig. 9. The other sputtering conditions are the same: deposition rate of 0.1 atom/ps, substrate temperature of 300 K, and incident energy of 2 eV. The results indicate that a larger incident angle makes a rougher surface. The velocity of the incident atoms consists

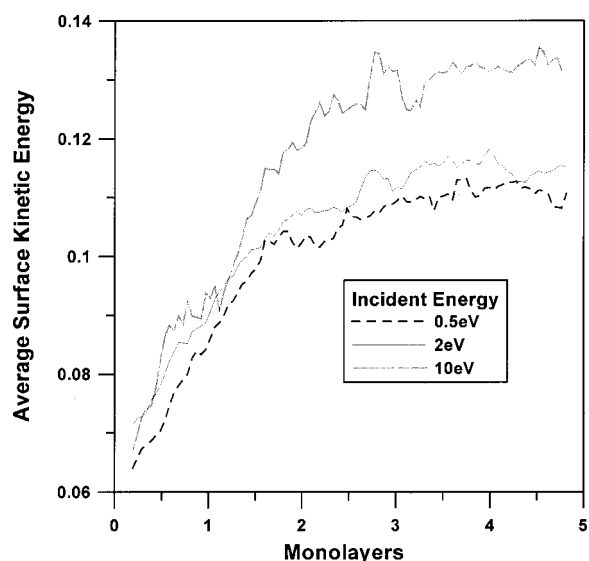


FIG. 7. Average surface kinetic energy at incident energies of 0.5, 2, and 10 eV, and the other conditions remain the same: zero incident angle, 300 K substrate temperature, and 0.1 atom/ps deposition rate.

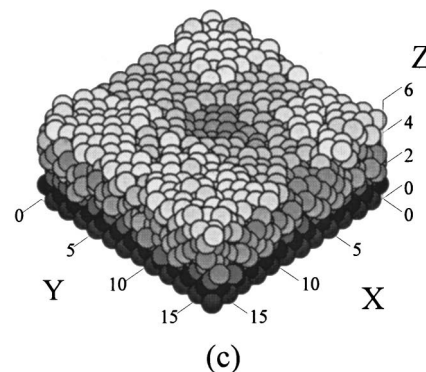
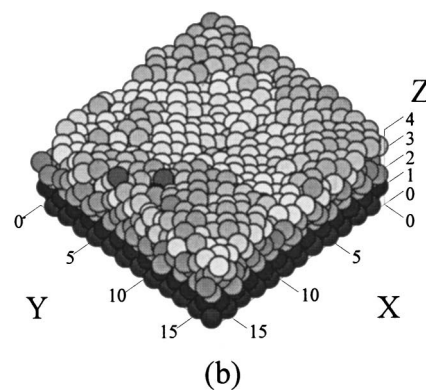
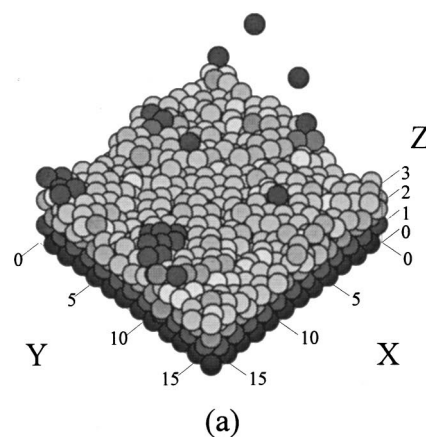


FIG. 8. Morphology at incident energy of 10 eV with zero incident angle, 300 K substrate temperature and 0.1 atom/ps deposition rate (a) 1 ML, (b) 3 ML, and (c) 5 ML.

of  $x$ ,  $y$ , and  $z$  components. The  $z$  velocity component causes the incident atom to collide (interact) with the surface atoms directly. It is proposed that when incident atoms possess the same velocity, the amount of the energy transferred from incident atoms to surface atoms depends on the  $z$  velocity component of the incident atoms. At different incident angles, however, the  $z$  component differs, changing the amount of transferred energy and thus the average surface kinetic energy. The influence of the incident angle is on the amount of the energy transferred from the incident atoms to the surface atoms. A larger incident angle leads to less energy transfer to the surface atoms because the normal impact



FIG. 9. Interface width variation at incident angles of 0°, 25°, 45°, and 75°, and the other conditions remain the same: 2 eV incident energy, 300 K substrate temperature, and 0.1 atom/ps deposition rate.

energy is proportional to the square of cosine value of the incident angle. This result can also be seen from the average surface kinetic energy, shown in Fig. 10.

Film morphology for an incident angle of 25°, shown Fig. 11, is smoother than that for the of 75° incident angle, shown in Fig. 12. Inspection of Fig 12 (c) shows a deep groove formed between two columnar structures. Together, these results indicate that a larger incident angle encourages three-dimensional columnar film structure.

The trend of larger average surface kinetic energy resulting in smoother surface at different incident angles is shown in Figs. 9 and 10. Inspection of Fig. 9 shows the interface widths with incident angles of 25° and 45° are very similar, while the interface width at an incident angle of 75° is much

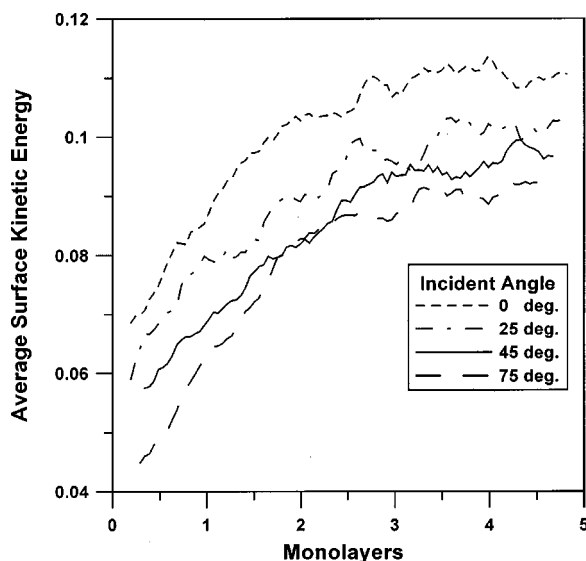


FIG. 10. Average surface kinetic energy at incident angles of 0°, 25°, 45°, and 75°, and the other conditions remain the same: 2 eV incident energy, 300 K substrate temperature, and 0.1 atom/ps deposition rate.

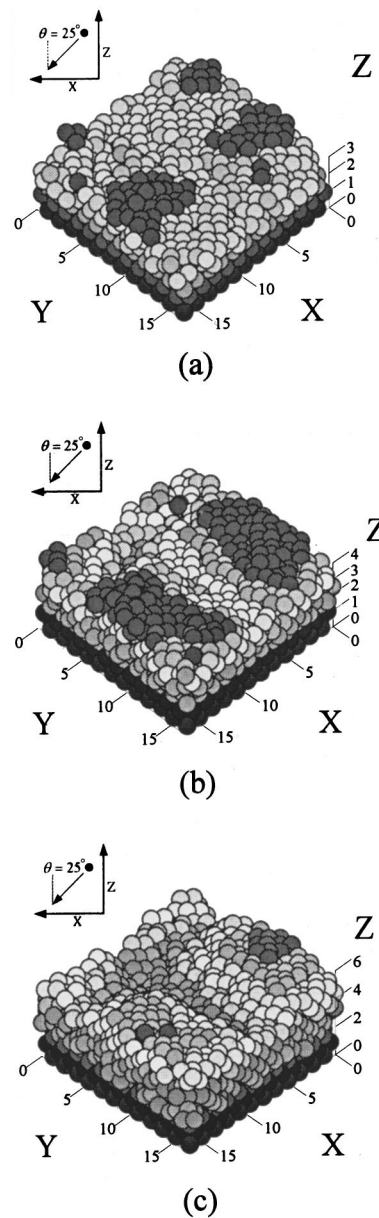


FIG. 11. Morphology at incident angle of 25° with 2 eV incident energy, 300 K substrate temperature, and 0.1 atom/ps deposition rate (a) 1 ML, (b) 3 ML, and (c) 5 ML.

larger. The self-shadowing mechanism is cited herein to explain this phenomenon. In a related study,<sup>16</sup> simulation results revealed film density decreased very slowly with incident angles lower than 45°, but film density decreased very rapidly when the incident angle exceeded 45°. Conversely, surface roughness increased very slowly with incident angles lower than 45°, but surface roughness increased very rapidly when the incident angle exceeded 45°. This is consistent with our simulation results which found very similar interface widths at 25° and 45°, with much larger interface width at 75°.

#### IV. CONCLUSION

In this article, MD simulation is utilized to study the topography of thin films grown by the sputtering deposition

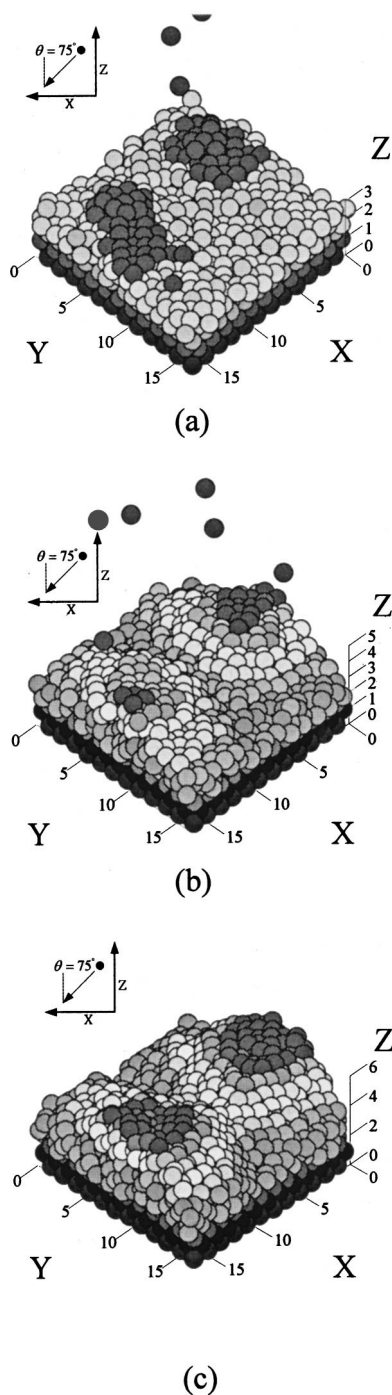


FIG. 12. Morphology at incident angle of  $75^\circ$  with 2 eV incident energy, 300 K substrate temperature, and 0.1 atom/ps deposition rate (a) 1 ML, (b) 3 ML, and (c) 5 ML.

process for different working parameters such as substrate temperature, incident energy, and incident angle. Interface width is used to quantify surface roughness. Average surface kinetic energy is proposed as a measure of thermal diffusivity of the surface atoms. Indeed, our simulation results found that surface roughness is strongly dependent on the surface kinetic energy of the deposited film. From this study, several important conclusions are drawn.

(1) There is less energy transfer into sublayer atoms as

the substrate temperature increases. The increase of surface temperature (thermal diffusivity) leads to production of a smoother film surface.

(2) Higher incident energy leads to larger amounts of incident atom energy transferred to the surface atoms. Smoother films are thereby produced.

(3) There is less energy transferred to surface atoms as the incident angle increases. Rougher surfaces are thus produced.

(4) With decreasing substrate temperature, decreasing incident energy, and increasing incident angle, the growing film tends toward a three-dimensional columnar structure. Conversely, with increasing substrate temperature, increasing incident energy, and decreasing incident angle, the growing film tends toward a two-dimensional (Frank–van der Merwe) quasi-layer-by-layer structure.

(5) The average surface kinetic energy is an important factor in determining the surface roughness produced. Generally, the produced film is smoother when the average kinetic surface energy is larger.

Trends for three different sputtering deposition processing parameters are investigated in this study. The final results can be used as reference in the theoretical and practical fabrication of thin films for improved control of surface roughness.

## ACKNOWLEDGMENTS

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