MONTE CARLO SIMULATION OF HYPERTHERMAL PHYSICAL VAPOR DEPOSITION

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Abstract—Low-pressure sputtering and ionized vapor deposition processes create atomic fluxes with kinetic energies in the 1.0–20 eV (and above) range. The impact energy of these hyperthermal atoms significantly effects the surface morphology and structure of vapor deposited films. Recent molecular dynamics simulations of metal atom interactions with a metal surface have established the energy and angular dependence of many of the impact energy induced mechanisms of atomic assembly including biased diffusion, atomic reflection, resputtering, and thermal transient induced “athermal” diffusion. These four effects have been incorporated into an earlier two-dimensional kinetic Monte Carlo model that analyzes the thermally driven multipath diffusional processes active during vapor deposition (Y. G. Yang et al., Acta Mater., 45 (1997) 1445). The contributions of the energy-dependent mechanisms to surface morphology were found to grow in importance as the substrate temperature was reduced and/or as the rate of deposition increased. The simulation methodology was used to establish functional dependence of surface roughness upon the atom’s kinetic energy and its direction of incidence during the hyperthermal deposition of nickel vapor. The simulations reveal the existence of a minimum surface roughness at an incident angle which increased with impact atom kinetic energy. Modification of the impact energy is shown to be a viable means for controlling surface morphology during physical vapor deposition under high deposition rate, low deposition temperature growth conditions.

Keywords: Physical vapor deposition (PVD); Monte Carlo simulation; Thin films

1. INTRODUCTION

The growth of materials from the vapor phase is widely used to create thin films and coatings [1–3]. Increasingly stringent levels of film perfection are often needed. For example, thin film devices based upon the giant magnetoresistance (GMR) effect require the growth of Ni$_{88}$Fe$_{17}$/Cu/Ni$_{83}$Fe$_{17}$ planar multilayers in which the interfacial roughness is less than a few angstroms and intermixing of the dissimilar metal layers is negligible [1–3]. Vapor deposition strategies that achieve this microstructure exploit kinetically limited growth mechanisms. However, the design of deposition processes that achieve atomic scale perfection by controlling the kinetics of film growth is complicated because of the many mechanisms that contribute to the atomic assembly process [4, 5]. Interest has therefore developed in the use of atomistic computer simulation techniques to better understand and model the atom-by-atom assembly of thin films [4–15].

The atomic assembly events that occur in vapor deposition processes are “enabled” by both thermally activated atomic diffusion and hyperthermal atom impact induced mechanisms. To realistically simulate the growth of films under energetic conditions, a model must address both conventional thermally activated processes and the energetic impact effects responsible for additional atomic rearrangement mechanisms.

The impingement of energetic atoms with a growth surface can induce numerous physical and chemical phenomena [4, 5, 16]. Depending upon the energy and the surface condition, an incident atom can be trapped on the surface, implanted beneath it, reflected, and/or undergo long range biased diffusion in the impact direction on the surface. It can also cause localized heating which may induce atomic rearrangements that would normally not occur (athermal diffusion). For high-energy impacts, they may even cause resputtering of the already deposited atoms [4, 5]. Chemical reactions can occur with other chemisorbed species or a gas-phase atom or a physisorbed
molecule. Electronic scale processes resulting from inelastic scattering are also possible, but are less likely to influence the final structure of the film [16].

The complex effects of incident atom energy during vapor deposition are pervasive for many low pressure sputter deposition processes. This is because sputtered atoms are emitted from targets with a broad energy spectrum extending from a few tenths of an electron volt up to 20 eV or more [17]. This spans the energy barriers of many atomic assembly processes [4, 5, 18]. While experiments indicated that many impact phenomena can cause a change of the local atomic configurations [17, 18], it is difficult to quantify their individual contributions. It is also difficult to know how these mechanisms are dependent upon the incident energy and angle, deposition rate and substrate temperature.

One approach to the modeling of hyperthermal deposition utilizes molecular dynamics (MD) techniques to simulate energetic impact processes. The MD method solves for the trajectories of each atom in a crystal by integrating Newton’s equations of motion using an interatomic force law [4–7, 9–11]. Since the lattice atom vibration is explicitly traced in this approach, the forces on atoms must be calculated several times per lattice vibration period. This leads to intensive computational demands. As the shortest lattice vibration periods are about $10^{-13}$ s, only 1–10 ns of real time can be simulated. Many thermally activated atomic relaxations take microseconds or longer to complete and are not represented in an MD simulation of vapor deposition. However, since the MD method evolves atomic configurations by explicitly analyzing interatomic forces, it rigorously incorporates the atomic assembly mechanisms initiated by the high kinetic energy atom impacts that are frequently completed in less than 5 ps [4–7].

Kinetic Monte Carlo (KMC) methods are being developed to analyze multipath thermally activated diffusion [8, 14, 15]. These techniques evolve atomic configurations by identifying their thermally activated atomic jump paths and implementing a probabilistic scheme for their subsequent execution. In KMC analyses of film growth, the likelihood that an atom jumps from one lattice site to another depends upon its local atomic configuration which can be characterized by an activation energy and a jump attempt frequency [8]. The simulation first deduces the set of jump probabilities for every allowed jump path using precalculated activation energies and then executes jumps according to their relative probabilities. After a jump is executed, time is advanced by a computational time step determined by the residence time of the system, i.e. the reciprocal of the sum of the jump rates for all the allowed jump paths of the system. This process is then repeated until the time between atom arrivals is exhausted. A new atom is added and the algorithm iterated.

Two- (2D) and three-dimensional (3D) forms of the KMC method have been developed to treat diffusion on defective growth surfaces. The 2D methods enable the surface morphology evolution to be simulated, but the out-of-plane constraint upon diffusion restricts the predictive accuracy. Three-dimensional approaches are computationally expensive to implement, especially if an off-lattice approach is used. Nonetheless, both approaches are beginning to enable an investigation of the effects of substrate temperature, deposition rate and incidence angle of the flux upon the surface morphology and void structure of physical vapor deposited thin metal films grown either on flat or featured surfaces (like those used for dual damascene grown integrated circuit interconnects) [19–21]. While the KMC methods address the numerous thermally activated routes of atomic assembly, they have not yet incorporated the effects of adatom energy or possibly important contribution from the adatom’s potential energy (i.e. the latent heat of condensation) which is released during each atom’s condensation.

Here, the computationally efficient 2D KMC method for the analysis of multipath thermal diffusion during low energy vapor deposition has been modified to incorporate the effects of atom potential and kinetic energy. The resulting energy-dependent KMC model is then used to systematically investigate trends between the surface morphology of vapor deposited nickel films and the adatom kinetic energy, substrate temperature and deposition rate.

2. SIMULATION METHODOLOGY

A variety of phenomena can occur following the energetic impingement of an atom with a surface. They can be broadly classified as either thermally activated or energy impact induced. Examples of the latter are adatom reflection and resputtering of previously deposited material. The phenomena initiated by energetic interactions are generally completed within a few picoseconds [4]. This is usually much smaller than the atom residence times at the substrate temperatures of typical interest [22]. As a result, the energetic reassembly events and thermal diffusion can be treated sequentially. In other words, following each adatom arrival, the impact energy effects can be treated first using approximations described in the following sections, followed by regular KMC simulated thermal diffusion. In order to incorporate the energy effects, we utilize results from molecular dynamics simulations. These results have been fitted to functional relationship linking atom assembly processes to the energy and angle of adatom impacts.

2.1. MD analysis of atom–surface interactions

To calculate the nickel adatom–surface interaction an embedded atom method (EAM) determined by Foiles et al. [23] was used as the interatomic potential. The free parameters of the model have been fitted with experimental material properties such as the lattice constant and the sublimation energy. Because the
sublimation energy is exactly reproduced [23], the EAM correctly accounts for the energy transfer during atom collisions with a surface. On the other hand, high-energy impacts cause atoms to approach each other closely and short-range interaction dominates. If the impact energy is not too high (say, less than 100 eV), the EAM potential should reasonably capture the impact phenomenon and accurately reveal the subsequent atomic relaxation processes because its environment-dependent potential form realistically describes the energetics of local non-equilibrium atomic configurations.

The molecular dynamics results were based on simulations conducted on (111) nickel surface. A cell of 24(224)×9(111)×14(220) (x, y, z) was used with periodic boundary conditions applied along x and z axes and the free surface of (111) normal to y. A fixed substrate temperature was applied to a region of two atomic layers at the bottom while the atoms above were left free. To simulate the impact, a nickel atom with desired incident energy and incident angle was introduced at a random location above the surface. The positions and velocities of all the atoms were then calculated throughout the impact process (about 2–5 ps) and were recorded as a function of time. To reduce statistical variations, the results were presented as an average of five separate runs using different random number seeds.

The calculations identified four major assembly mechanisms associated with atom impact: adatom reflection, resputtering of the previously deposited atoms, biased diffusion and thermal spike induced atom rearrangement (Fig. 1). Briefly, atom reflection occurs most frequently for oblique angles of incidence when the atom’s energy exceeds a threshold value (Fig. 1a). The threshold for nickel is about 15 eV and the reflection is most likely for incident angles between 65 and 80°. Resputtering is observed most significantly for angles in the range of 30–45° and at higher energy (Fig. 1b). When an impact does not initiate the processes above, the incoming atom can be trapped by attractive forces near the surface while its lateral momentum enables the adatom to skip along the surface. This is referred to as biased diffusion (Fig. 1c). Finally, every atom impact results in a temporary increase in local surface temperature (thermal spike). It results from the combined contributions of the latent heat of condensation and dissipation of the incident kinetic energy. This transient thermal spike can induce local atomic rearrangement (Fig. 1d).

The four mechanisms above have been systematically studied as a function of the atom energy and incidence angle. Details can be found in Refs. [4, 5]. Here, relevant results from these analyses have been fitted to functional relationships summarized in Tables 1–3 to facilitate subsequent KMC execution. The function forms were fitted in a way that they look physically meaningful. In equation (1), for example, θc is the minimum and θm the optimal angle to create an atom reflection. Equation (2) shows that the threshold energy in creating a reflection is 15 eV. In quantifying the thermal spike, a continuum medium approximation was made for the crystal. The space- and time-dependent surface temperature T(s,t) (s position relative to impact site and t time after impact), equation (12) in Table 3, was a solution of the standard Fourier equation for conduction heat transfer [24]. The initial effective temperature at the impact site came from applying equipartition theorem in which the average kinetic energy was obtained from MD calculations.

2.2. The energy-dependent KMC model

During an evaporation process at low deposition rate and/or high substrate temperature, thermally activated atomic jumping allows a thin film to reduce its overall energy and defect content. KMC methods can simulate the overall effect of multipath atomic jumps during deposition [8], and hence the structure of vapor deposited films. Incident atoms are introduced into the system at random positions above the substrate and are assigned an incident angle by random sampling of a pre-defined distribution. In the interval between atom impacts with the surface, the key steps of the diffusion simulation include: (1) calculating the
### Table 1. Fitted equations for reflection probability, angular and energy distribution of reflected atoms

<table>
<thead>
<tr>
<th>Equation no.</th>
<th>Equations</th>
<th>Parameters</th>
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<tbody>
<tr>
<td>(1)</td>
<td>Reflection probability as a function of $\theta$ at $E_i = 70.0$ eV</td>
<td>$\theta_i = 22^\circ$, $\theta_m = 72^\circ$, $p = 0.55$, $\lambda = 1.43$</td>
</tr>
<tr>
<td></td>
<td>$Y_r(\theta) = \min \left{ 1, p + p \sin \left[ 90.0 + 180.0 \left( \frac{\theta - \theta_0}{\theta_m - \theta_0} \right)^{1.43} \right] \right}$</td>
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<tr>
<td>(2)</td>
<td>Reflection probability as a function of $E_i$ at $\theta = 80^\circ$:</td>
<td>$E_i = 15$ eV, $p = 31.5$, $\lambda = 2.03$</td>
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<tr>
<td></td>
<td>$Y_r(E_i) = 1.0 - \exp \left[ - \left( \frac{E_i - E_i c}{p} \right)^{1.38} \right]$</td>
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</tr>
<tr>
<td>(3)</td>
<td>Reflection probability as a function of incident angle and incident energy:</td>
<td>$E_i = E_i c$</td>
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<td></td>
<td>$Y_r(E_i, \theta) = \frac{Y_r(E_i)}{Y_r(E_i - 70.0$ eV)}</td>
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<tr>
<td>(4)</td>
<td>Angular distribution of reflected atoms as a function of incident angle and energy:</td>
<td>$\beta = 1.38$, $G_i = -39.1 + [E_i - 39.1]$, $p_n$=normalization factor (integral of $\rho$ equals 1)</td>
</tr>
<tr>
<td></td>
<td>$\rho(\dot{\zeta}) = p_n (\dot{\zeta} - \theta)^{\alpha} (90.0 - \dot{\zeta})^{\beta}$</td>
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<td></td>
<td>where $\alpha = \frac{\beta (\theta_m - \theta_0)}{90.0 - \theta_0}$</td>
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</tr>
<tr>
<td></td>
<td>$\beta = 2.1$, $c = 1.07$, $E_0 = -13.39$, $\theta_0 = 50.8^\circ$</td>
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<tr>
<td>(5)</td>
<td>Energy distribution of reflected atoms as a function of incident angle and energy:</td>
<td>$\beta = 2.1$, $c = 1.07$, $E_0 = -13.39$, $\theta_0 = 50.8^\circ$</td>
</tr>
<tr>
<td></td>
<td>$\rho(E) = p_n E^{\alpha} (E_i - E)^{\beta}$</td>
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<tr>
<td></td>
<td>where $\alpha = \frac{\beta E_i}{E_i - E}$</td>
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<tr>
<td></td>
<td>$E_f = (c E_i + E_0) \sin \left[ \frac{90.0 (\theta - \theta_0)}{90.0 - \theta_0} \right]$</td>
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jump rates ($p_i$) for all possible jump paths; (2) selecting and executing a jump according to its relative jump rates; (3) summing up the rates of all possible jumps ($P = \sum_i p_i$) and marching the system clock forward by a time interval $t_n$ ($t_n = 1/P$). The new atomic configuration is then used to compute new jump rates and the algorithm is repeated until the time between atom arrivals is exhausted. This algorithm is able to account for the effects of deposition rate, temperature and incident flux distribution upon thin film growth. By using the relationships in Tables 1–3, the KMC model can be extended to include the effects of incident energy.

#### 2.2.1. Incorporation of reflection and resputtering

Three steps were used to incorporate reflection and resputtering events. The first step identified the type of event. Suppose that an atom with a kinetic energy $E$ approaches a surface at an incident angle (defined as the angle between the local surface normal and the incident direction), its probability for reflection or
Table 3. Fitted equations for local effective temperature and biased diffusion distance

<table>
<thead>
<tr>
<th>Equation no.</th>
<th>Equations</th>
<th>Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>(11) Biased diffusion distance:</td>
<td>$d(\theta, E_i) = \frac{z E_i \theta</td>
<td>\theta - \theta_a</td>
</tr>
<tr>
<td>(12) Local effective temperature due to latent heat release and incident kinetic energy:</td>
<td>$T(\alpha) = T_{\text{sub}} + (66.0 + 3.6E_i) \times \exp \left( \frac{0.09\alpha}{\nu + 0.05} \right)$</td>
<td>$T_{\text{sub}} = \text{substrate temperature}$, $E_i = \text{incident energy}$, $\alpha = \text{elapsed time}$, $x = \text{distance from adatom}$</td>
</tr>
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</table>

resputtering can be calculated using equation (3) in Table 1 and equation (8) in Table 2, respectively. A random number (between 0 and 1) is then generated and compared with the probability. If the random number is less than the probability, the corresponding reflection or resputtering event is executed. Otherwise the program proceeds to simulate biased diffusion and effects of the thermal spike as described below. Cases exist where both reflection and resputtering events can occur. In that case, the kinetic energy and emitting direction are saved for the resputtered atoms and the reflection is first executed. Once reflection is completed, the data for the resputtered atom is then retrieved and the atom is treated as new incident atom. In step 2, the reflected or the resputtered atom is ballistically moved until it impinges upon new surface, or removed from the system when no new surface will be encountered. A third step is used to determine the possibility of further reflection and/or resputtering events if a reflected or resputtered atom still retains sufficient kinetic energy. The same procedure is repeated until no further reflection and resputtering events are possible.
The local incident angle $\theta$ needed for the calculations described above depends on the local surface normal at the impact site. This can be defined for a small surface area near the impact. In the case of a flat plane, its value remains the same regardless the size of the sample area, but for a rough surface, the sample area needs to be large enough to fully cover the local information, yet small enough to avoid inaccuracy caused by features such as sharp corners and other rough sites. Various test calculations were performed to determine an appropriate size for defining the local normal. It was found that five surface atoms generally give rise to reasonable surface normal and were hence used for all the simulations.

2.2.2. Biased diffusion. To execute biased diffusion, the incident atom’s skipping distance $d(\theta,E)$ is first calculated using equation (11) in Table 3. The execution of biased diffusion on a smooth surface simply moves the incident atom in the direction of its momentum by the skipping distance. To move an atom on a rough surface, the skipping distance was first compared with the unit move distance (taking to be the nearest neighbor distance). If the skipping distance is greater, the atom is moved by the unit distance. Then the remaining skipping distance is reduced by a predetermined value to account for friction of a rough surface. The procedure continues until the skipping distance is completely consumed. Mathematically, this can be described by:

\[ S_{i+1} = S_i - Ca \]  

(1)

where $S_i$ is the remaining skipping distance at the $i$th jump, $a$ the unit distance, $C$ is a coefficient and $S_{i+1}$ the remaining skipping distance at $(i+1)$th jump. As long as $S_{i+1}$ is positive and greater than $a$, we continue to move the atom.

The parameter $C$ was obtained based on the statistic analysis of MD results for skipping on rough surfaces. The values of $C$ are usually different at each jump site $i$. For example, $C_i = 1$ for a perfect smooth surface. $C_i$ is larger when the atom moves down or against a ledge, and $C_i = S_i/a$ (i.e. stop after the jump) when the atom encounters a sharp valley or a higher ledge and so on.

2.2.3. Transient thermal diffusion. When an energetic atom impinges upon a growing film, rearrangement among atoms in the thermal spike zone near the impact site takes place. The extent of this local rearrangement depends upon both the energy (or equivalently the temporary rise of temperature) acquired by the atoms near the impact site and their neighborhood restraint. The atom with highest energy yet least neighborhood restraint has the highest probability to jump to a lower energy site. The criterion for a jump to occur then can be expressed by

\[ P_i > P_r \Rightarrow P_i = \left( \frac{E}{Q_i} \right)^n \]  

(2)

where $E$ is the energy of an atom in the thermal spike zone (obtained using equation (12) in Table 3), $Q_i$ the neighborhood restraint taking to be the activation energy for $i$th jump pathway, and $n$ and $P$ are adjustment parameters. Suppose that an atom has a pathway meeting the condition of equation (2), it is then said to have a probability to jump to a new place through that path. When many atoms have the probability to jump in different paths, the jump simulation is conducted in such a way so that probability for the jump in the $i$th pathway scales with $P_i$. Once the jump is made, $P_i$ is updated for all the paths. This procedure was repeated until no atoms have probability to move.

The parameters $n$ (energy sensitivity index) and $P$ (a cut-off number) were determined by matching the configurations obtained using MD simulations at short time/length scale to those obtained from KMC calculations at the same conditions. Using the values of $P$ and $n$ determined this way, MD and KMC simulations give similar configurations at various atom kinetic energies ranging from 0.1 to 2.0 eV under normal incidence condition.

3. RESULTS AND DISCUSSION

The KMC model described above was used to explore the importance of various kinetic energy mechanisms. Based on a 2D approximation of nickel deposition [8], the substrates were constructed to contain eight rows of 200 close-packed nickel atoms. Periodic boundary conditions were employed laterally to minimize the effect of small system size. Eight thousand atoms were deposited for each condition. To quantify the surface morphology, a surface roughness parameter was calculated as the ratio of the surface area of a simulated configuration to that of the smooth substrate [22, 25] (for a flat surface, the roughness ratio=1.0; while for a rough surface, the roughness ratio >1.0). To minimize statistic variation of the results, 10 separate simulations (each with a different random number seed) were conducted and the average used for a roughness data point. Since this is a 2D model, the actual temperatures used for deposition are not directly related to those of a 3D system. To compensate, we note that the 2D vacancy formation energy is about 2/3 of the corresponding 3D value [26]. Thus, a homologous temperature is also given, based on a melting temperature, $T_m$, of 1150 K, which is 2/3 of the 3D nickel melting temperature of 1726 K.

3.1. Relative contributions of mechanisms

Simulations of vapor deposition at an incident energy of 40 eV, a deposition rate of 0.1 µm/min, and a normal cosine angular distribution for the incident flux and two different temperatures were carried out to examine the contribution of each atomic assembly mechanism (Fig. 2). The use of a high energy of 40 eV can more clearly reveal the energy effects (this would mimic the conditions when a metal flux was
Contributions of various growth mechanisms in hyperthermal physical vapor deposition. The substrate temperatures \( \frac{T}{T_m} \) were 0.17 and 0.43, respectively. The deposition rate was 0.1 \( \mu \)m/min, and the kinetic energy was 40 eV. The incident flux had a cosine distribution. The deposition of 8000 nickel atoms was simulated. The configurations in (a) result from ballistic deposition with only nearest neighbor relaxation after atoms’ impingement. The configurations from (b) to (g) result from combined mechanisms sequentially added one at a time to show the difference that each additional one makes.

First, a ballistic deposition model was used to show the configuration of deposited film with very limited atomic relaxation. This was done by simply placing the depositing atom at the lattice site nearest to the location of impingement and then freezing it. The simulated configurations are shown in Fig. 2a. Obviously, these configurations are independent of temperature. The very rough and highly porous structure results from self shadowing. This is likely to represent the porous configuration under extremely low temperature conditions. The traditional KMC model was then used to simulate the effects of thermal diffusion and the results are shown in Fig. 2b. It represents thermal deposition excluding only latent heat. By successively turning on the mechanisms of athermal diffusion due to latent heat release (i.e. thermal spike at zero incident energy), atom reflection, resputtering, biased diffusion, and athermal diffusion due to impact energy (i.e. thermal spike at non-zero incident energy), the KMC simulations described earlier were carried out to reveal the effects of individual mechanisms. The results are shown as Fig. 2c–g.

Low-temperature \( \frac{T}{T_m} = 0.17 \) configurations shown in the left column of Fig. 2 were first examined. Comparison between Fig. 2a and b shows that thermal diffusion significantly increases the column size. Fig. 2c indicates that the latent heat release can cause considerable atomic rearrangements that lead to even larger column sizes and less inter-column voids. High-energy reflection and resputtering are not seen...
to have noticeable effects on the film structures (Fig. 2d and e). This is because relatively fewer reflection and resputtering events occur under the simulated conditions. More importantly a reflection or a resputtering event only results in redeposition of atoms at different locations and does not cause a local atomic rearrangement. Fig. 2f shows that the inclusion of biased diffusion resulted in fully densified structure and much smaller surface roughness. This occurs because the biased diffusion enables atoms to move by either a short or a long distance in the direction of incidence, promoting their attachment at lattice ledges. However, the biased diffusion only affects the adatoms, not the other atoms adjacent to the impinging site. By incorporating thermal spike (including latent heat and kinetic energy contributions) induced rearrangement from neighboring atoms, a fully dense and rather flat configuration was obtained (Fig. 2g).

At higher temperature of $T/T_m = 0.43$, effects associated with various energy mechanisms became smaller. The effects of intensive thermal diffusion overwhelm the energy induced atomic rearrangement.

3.2. Effect of kinetic energy on surface morphology

It has been shown that the contributions of energy mechanisms to the atomic assembly process depend on temperature and is especially significant when thermal diffusion is low. To systematically explore the energy effects, the full hyperthermal KMC model was used to simulate vapor deposition at various incident energies from 0 to 20 eV at a fixed low substrate temperature of $T/T_m = 0.17$, a deposition rate of 0.1 $\mu$m/min, and a standard cosine flux angular distribution. Four representative configurations are shown in Fig. 3.

It is clear that increasing incident energy helps decrease surface roughness and promote a denser film. At zero incident energy (thermal energy only), Fig. 3a, the configuration is a porous columnar structure with a rough surface (the roughness ratio was found to be 3.22). It results from limited atomic mobilities and self-shadowing. Because there are no biased diffusion, reflection and resputtering at low incident energies, only the latent heat release enables the local reconstruction of atoms. However, the effect is limited. The microstructure is typical of zone I structure observed in experiments [18]. At 5 eV, Fig. 3b, the porosity significantly decreases and the roughness ratio was found to reduce to 1.67. Since reflection and resputtering were not activated at this energy, the observed change was a result of a stronger thermal spike induced athermal diffusion and biased diffusion. This trend continued as the incident energy was increased to 10 eV, Fig. 3c, where the roughness ratio was reduced to 1.55 and voids were completely eliminated. Finally, at 20 eV, Fig. 3d, the surface became much smoother and the roughness ratio was further reduced to 1.29. When the kinetic energy is at 20 eV or higher, several new mechanisms were activated. First, reflection became possible. A reflected atom generally tends to move in the forward direction and is less likely to attach to an existing mound. When it jumps onto the edge of the mound, it enables the mound to grow in the horizontal rather than in the thickness direction. Secondly, resputtering can occur at 20 eV. Resputtering is mostly significant at surface asperities as these places are exposed to more incoming flux. The preferential etching of these sites also promoted smoothness.

Detailed simulations on the effect of incident energy have also been conducted at various substrate temperatures. The roughness obtained from these simulations is plotted in Fig. 4 as a function of incident energy at different fixed substrate temperatures. It can be seen that at a low temperature of $T/T_m = 0.17$, the surface roughness initially decreases very rapidly as the incident kinetic energy increases up to 15 eV. Further increases of energy then cause only a small change in the roughness. Increasing the substrate temperature causes a roughness decrease over the whole energy range. In addition, the effects of incident energy on surface roughness became less
Fig. 4. Effect of incident energy on surface roughness of nickel films under different substrate temperatures \((T/T_m)\), (a) 0.17; (b) 0.22; (c) 0.26; and (d) 0.30. The deposition rate was 0.1 \(\mu\)m/min and the incident angle had a cosine distribution.

significant as temperature was increased. For temperature above \(T/T_m = 0.26\), the effect of energy becomes negligible. As a result, energy modification is more effective at low temperatures.

The functional dependence between surface roughness and incident energy at different deposition rates was also calculated at a fixed \(T/T_m = 0.22\) and a cosine angular deposition flux. The results of this calculation are shown in Fig. 5. It shows that at the highest simulated deposition rate of 10 \(\mu\)m/min, increasing the incident energy from 0 to 15 eV significantly reduced the surface roughness. The roughness appeared to be saturated when the energy was increased above 15 eV. Similar to increasing temperature, it can be seen that decreasing the deposition rate reduced both the roughness and its energy dependence over the whole energy range simulated. Especially, the roughness becomes almost independent of incident energy when the deposition rate was decreased below 0.01 \(\mu\)m/min at \(T/T_m = 0.22\).

The simulations demonstrate the interplay between thermal diffusion and various energy related mechanisms on surface roughness. At either low substrate temperatures or high deposition rates, thermal diffusion is not significant and the flattening of the surface is then mainly achieved by energy related mechanisms such as athermal diffusion, biased diffusion, reflection and resputtering. At high temperatures or low deposition rates, thermally activated diffusion is often sufficient to enable the formation of a flat dense structure and the effects of energy mechanisms are overwhelmed.

Extensive experimental results have shown that the use of energetic atoms during thin films deposition can promote layer-by-layer growth and that energetic atoms can activate various surface assembly processes [27–33]. Almost all experimental analyses attributed the formation of dense and flat structures during hyperthermal energy deposition to the dissociation of surface asperities by energetic bombardment induced atomic reconstruction. The dissociated atoms preferentially reach step edges, resulting in a layer-by-layer epitaxial growth. However, these desirable results are often compromised by deleterious effects of energetic impacts. For instance, when energy increases above a threshold, permanent damage to the lattice increases more rapidly than the annealing processes are capable of healing, resulting in a sharp increase on the number of defects [30, 33]. Thus there is an optimum energy for achieving a layer-by-layer epitaxial growth while maintaining high crystalline quality films which are relatively defect free [30].

3.3. Effect of incidence angle on surface morphology

Thin films grown under low substrate temperature/low impact energy conditions are generally known for their characteristic (and often detrimental) columnar structure caused by self-shadowing. Self-shadowing is particularly strong when incident atoms are away from the substrate normal. To more clearly show the interplay of incident energy and incident angle, the energy-dependent KMC model has been used to simulate depositions where all atoms have the same incident energy and incident direction.

A series of calculations were carried out at different incident angles, but at a fixed temperature of \(T/T_m = 0.26\), a fixed deposition rate of 0.1 \(\mu\)m/min, and a fixed incident energy of 20 eV. Representative configurations are shown in Fig. 6. At normal inci-
Fig. 6. Effect of incident angle on surface morphology of nickel films at a substrate temperature of 300 K (\(T/T_m\) = 0.26). The deposition rate was 0.1 \(\mu\)m/min and the kinetic energy was 20 eV. The deposition of 8000 nickel atoms was simulated.

didence of \(\theta = 0^\circ\), mounds on a rough surface can be clearly identified. The mounds grow broader and the number of the mounds decreased as the incident angle was increased to 20°. The surface became considerably smooth at \(\theta = 40^\circ\) and the mounds can barely be identified. Further increases in the incidence angle to 60° reversed the trend and caused the surface to become rougher.

More simulations on the effects of incident angle were carried out and the data for surface roughness as a function of incident angle are summarized in Figs 7 and 8. Fig. 7 compares the results for four incident energies at fixed \(T/T_m\) = 0.22 and deposition rate of 0.1 \(\mu\)m/min. Fig. 8 compares the results for four substrate temperatures at the same deposition rate and a 10 eV energy. Fig. 7 shows that at zero incident energy, the surface roughness initially increases slowly with increasing incident angle up to 40°, then rapidly increases when the incident angle is beyond about 40°. This implies that a strong self-shadowing induced defective structure can not be healed by latent heat induced atomic relaxation alone. The rapid increase of roughness at 40° results from the formation of voided columnar structure. With the incident energy increased to 5 eV, a dramatic change is observed on the surface roughness versus the incident angle relationship. The surface roughness initially decreases as incident angle is increased to about 30°, then starts to increase as the incident angle is further increased. A roughness minimum is seen around 30–40°. The surface roughness also becomes very significant at incident angle up to 60°. Compared with the low energy curve, the overall roughness has been greatly reduced. With an increase in energy to 10 eV and then to 20 eV, roughness is continuously reduced. This reduction is more significant at high incident angles. Nevertheless, the roughness versus incident
angle trend remains similar. Interestingly, the incident angle at which the minimum roughness is obtained increases with increasing energy. This occurs because high energy activates more surface processes which causes surface asperity flattening and a consequent reduction of self-shadowing. Fig. 7 also shows that an increase of energy from 0 to 5 eV causes more flattening than an increase of energy from 10 to 20 eV. This indicates that the effects of athermal and short-range biased diffusion on smoothing the surface are significant.

The effect of substrate temperature on the roughness–incident angle curves is similar to that of incident energy as seen in Fig. 8. Increasing the temperature generally reduces the roughness, especially at high incident angles. For an incident energy of 10 eV, the angle for the minimum roughness increases from about 35 to about 42° as the temperature increases from \( T/T_\text{eq} = 0.17 \) to 0.30. This arises because thermal diffusion promotes surface reconstruction that flattens a rough surface and eliminates voided columnar structure.

At the energy levels treated above, reflection and resputtering were not significant. The observed microstructures were mainly caused by the thermal spike induced athermal diffusion and biased diffusion. Biased diffusion had a significant effect during oblique angle deposition. When all atoms had the same incident angle, the unidirectional biased diffusion on the surface promote local layer-by-layer growth and resulted in a smooth surface. This effect increases with increasing the incident angle. However, large incident angles can cause self-shadowing, resulting in rough surfaces. There is a compromise between these two mechanisms. The optimal angle for minimum roughness depends on many process conditions including the incident energy.

4. CONCLUSIONS

An energy-dependent kinetic Monte Carlo method has been developed to simulate sputter deposition on a flat substrate. The method allows a realistic study of incident energy effects during deposition and their interplay with other conditions, such as substrate temperature, deposition rate, and incident angle. Application of the method to the deposition of nickel films reveals that:

1. In addition to thermal diffusion, the primary energy induced contributions to the thin film’s densification and smoothing come from short-range atomic rearrangement mechanisms including biased and thermal spike induced athermal diffusion, while those from reflection and resputtering are secondary and cause only redeposition.

2. Surface roughness decreases with increasing kinetic energy. The effect becomes stronger as the substrate temperature is lowered and/or the rate of deposition is increased.

3. For directional hyperthermal deposition, a minimum surface roughness at a nonzero angle of incidence is observed. The angle corresponding to the minimum roughness increases with increasing kinetic energy, substrate temperature and with decreasing deposition rate.

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