INVESTIGATION OF ENERGETIC DEPOSITION OF Au/Au (001) THIN FILMS BY COMPUTER SIMULATION

Q.Y.Zhang
State Key Laboratory for Materials Modification by Laser, Ion and Electron Beams, Dalian University of Technology, Dalian 116024, China

Z.Y.Pan, G.Q.Zhao, and J.Y.Tang
Accelerator-based Atomic and Nuclear Physics Laboratory, Institute of Modern Physics, Fudan University, Shanghai 200433, China

ABSTRACT

A new computer simulation method for film growth, the kinetic Monte Carlo simulation in combination with the results obtained from molecular dynamics simulation for the transient process induced by deposited atoms, was developed. The behavior of energetic atom in Au/Au(100) thin film deposition was investigated by the method. The atomistic mechanism of energetic atom deposition that led to the smoothness enhancement and the relationship between the role of transient process and film growth mechanism were discussed. We found that energetic atoms cannot affect the film growth mode in layer-by-layer at high temperature. However, at temperature of film growth in 3-dimensional mode and in quasi-two-dimensional mode, energetic atoms can enhance the smoothness of film surface. The enhancement of smoothness is caused by the transient mobility of energetic atoms and the suppression for the formation of 3-dimensional islands.

1. INTRODUCTION

Well-controlled interface and surface morphologies in film growth are required for many applications, such as in microelectronics, optoelectronics, X-ray optics and magnetic materials. To controll the film growth, several methods based on energetic atom deposition, *e.g.* ion beam assisted deposition (IBAD), sputter deposition, accelerated molecular beam epitaxy(AMBE), and energy-filtered ion beam deposition (IBD),*etc.*, have been developed in the past few years. In the experiments it has been observed that the use of low energetic atoms can result in enhancing the smoothness of surface and in suppressing the formation of 3-dimensional islands during the film epitaxial growth [1-5]. These encouraging results promote the present study to understand the atomistic mechanisms in the energetic atom deposition.

Many studies of thin film growth in different energetic atom deposition technologies by computer simulations with molecular dynamics (MD) method and Monte Carlo (MC) method have revealed some atomistic mechanisms of thin film growth[6-12]. Chason and co-workers suggested a transient ion-induced defects smoothing mechanism and found the phenomenon of ion-enhanced island coarsening in IBAD growth by kinetic MC simulation[6-8]. Gilmore and Sprague simulated the energetic atom growth of Ag/Ag(001) film by MD method[9-10]. Villarba et al. investigated the atomic exchange processes in sputter deposition of Pt/Pt(111) system using MD method and found that by raising the

incident kinetic energy of deposited Pt atom from 0.25 eV to 10 and 20 eV, an increasing area of impact atop a heptamer and straight type A and B steps on Pt(111) results in push-out events where incident atoms get incorporated into the growthing layer[12].

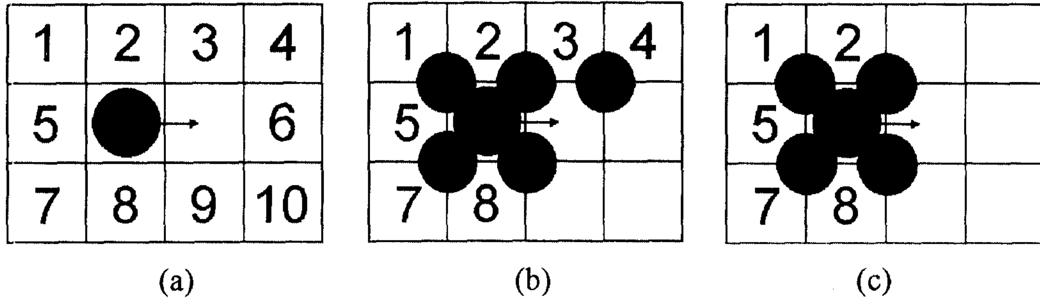
To understand the film growth mechanism of energetic atom deposition, two different processes must be distinguished, transient process and diffusion process. In the energetic atom deposition, the transient process caused by energetic atoms is as important as diffusion process. The transient process includes the transient mobility of energetic atoms and adatoms, and of the defects produced by energetic atoms. In the transient process, all the motion of atoms and the production of defects occur in a few pico-seconds, which cannot be counted in the simulation of film growth by kinetic MC method. However, the diffusion process, involving in-plane diffusion and interlayer diffusion, adatom cooperation with defects, and nucleation, generally occurs in nanoseconds to seconds depending on the temperature and deposition rate. It is impossible to consider the diffusion process with enough simulation time in the MD method.

In this letter, we propose a new computer simulation method, the kinetic Monte Carlo simulation in combination with molecular dynamics simulation results about the transient process caused by energetic atoms, and apply the method to the simulation of Au/Au (001) homoepitaxial growth based on energetic atom deposition. In our computer simulation program, the transient process considered was based on the MD simulation results, which supplied the detail description of local environment change for energetic atom deposition. The diffusion process was simulated by kinetic MC method. We explore the atomistic mechanism of energetic atom deposition to lead the smoothness enhancement and reveal the relationship between the role of transient process and film growth mechanism at different temperature.

2. SIMULATION METHODS

In our MD simulation, a NVT canonical ensemble system contained 576 substrate atoms was used. The transient process in our simulations was defined as the change of local environment of surface induced by energetic atoms in the time of 3 ps after the atom deposited. To investigate the transient processes of energetic atom, 6 kinds of adlayer configurations contained 0, 1, 4, 9, and 16 atoms were considered in the simulation. The atoms with the energy of 0.1 eV and 10.0 eV were deposited at the different positions of adlayers and substrate surface from a distance of 0.6 nm to the topmost atom on the surface. The adlayer configurations were relaxed at 0 K for 30 ps prior to the deposition. The simulations of transient process were carried out using the velocity-Verlet method with variable steps, where the maximum integration step was 5 fs. The Au-Au interaction potential with embedded-atom method (EAM)[13] was used in the MD simulation and in the calculation of diffusion barriers. The diffusion barriers were calculated by the method similar to the one adopted by Breeman *et al.*[14]. The kinetic MC method we used is similar to the one proposed by Voter[15]. In the kinetic MC simulation program, we considered 1024 local configurations of adatom for in-plane diffusion and near to 1024 configurations for interlayer diffusion process involving two kinds of local environments(to see fig.1). The initial surface is a 160×160×4 flat array contained 51200 atoms. The deposition rate of energetic atoms is 0.025 monolayers/s (ML/s). The hopping rate of an adatom for a specific move is given by $h = \nu \exp(-E_B / kT)$, where ν

$=1.0\times10^{12}$ Hz is the attempt frequency, E_B is the height of the energy barrier that the adatom needs to cross for that specific move, k is Boltzmann's constant, and T is the temperature. Periodic boundary condition was applied to both of MD and kinetic MC simulations.



(a) (b) (c)

Fig. 1 The local enviroments of adatom diffusion (a) in-plane diffusion, (b) inter-layer diffusion I, (c) inter-layer diffusion II.

We investigated the behaviors of an energetic atom deposited onto the different positions of adlayer and substrate surface by the MD simulation and found the statistical law of transient process caused by energetic atoms. Each deposition was simulated more than 100 times by choosing the incident position in the area of 0.5 $a_0\times0.5$ a_0 around the center of deposition randomly. We found that the push-out probabilities of atoms in adlayer and substrate surface are relative with the energy of incident atom and the nearest neighbor coordination number in plane. The atom with the energy of 0.1 eV can not push out any atom even from a 4-atom cluster on surface. Therefore the downward funneling model, which was proposed by Evans *et al.*[16] and has been applied in many kinetic MC simulations of film growth[14,17], is a good approach for the investigation of film growth with very low energetic atom deposition, such as evapor phase deposition. By raising the incident atom energy to 10 eV, however, the atom near the deposition center can be pushed out from the adlayer of 16-atom cluster with the probability of 0.15. When the atom deposited at a asymmetric position, such as the position near to the corner or the edge of adlayer, the probability to find an atom remaining at its position is much lower than that at the center. The probability to find an atom at the corner is about 0.24 to 0.36, and the probability at the edge is about 0.56 to 0.64. When the incident atom deposited at the center of small adlayer, such as 4-atom cluster, the probability to find an atom at the positions the atom had occupied is 0.4.

Since it is impossible and unreasonable to use the limit configurations of adlayers in the kinetic MC simulation, so a general description about the change induced by energetic atom should be given. By comparing the events of energetic atoms deposited at different positions of different adlayers, we found the push-out probability of atoms in adlayers can be written as

$$P_{\text{push}} = 1 - P_{\text{D}} n_{\text{b}} \rho / a_u , \quad (1)$$

where P_D is the probability to find an atom with one nearest neighbor atom in plane, P_D = 0.2 for 10.0 eV atom, n_b is the coordination number of an atom in the adlayer or surface layer, ρ is the distance of the atom to the center of deposition, a_u =0.5 a_0, a_0 is the lattice constant. The push-out probability calculated by (1) is very close to the results of MD

simulation with the error less 15.0 % for different adlayer configuration and different deposition position. The distribution probabilities of the push-out atom can be written as

$$P_{\text{find}}^{\text{ad}} = \frac{1}{\sqrt{2\pi a_u}} \exp[-(\frac{R_{\text{P}} - R_{\text{D}}}{a_u})^2] \cos\theta\Delta\theta, \quad (2)$$

where R_{P} is the distance from the position of the atom to be found to the center of deposition, R_{D} is the effective radius of energetic atom, R_{D} =2.83 a_u for 10.0 eV atom, θ is the angle between $\mathbf{R}_{\text{P}}$ and $\mathbf{R}_{\text{sum}}$, where $\mathbf{R}_{\text{sum}} = \sum_{i=1}^{n_b} \mathbf{R}_i + R_{\text{D}}\mathbf{u}$, $\mathbf{R}_i$ is the vector of the bind of the atom to its nearest neighbor. The distribution probability of deposited atom can be written as

$$P_{\text{find}}^{\text{dp}} = \frac{1}{\sqrt{2\pi a_u}} \exp[-(\frac{R_{\text{P}} - R_{\text{D}}}{a_u})^2] \cos\varphi\Delta\varphi, \quad (3)$$

where φ is the angle between $\mathbf{R}_{\text{P}}$ and $\mathbf{R}_{\text{sum}}^{\text{ad}} = -\sum_{i=1}^{n} \mathbf{R}_i^{\text{ad}}$, n is the number of atoms pushed out, $\mathbf{R}_i^{\text{ad}}$ is the vector of displacement of the atom pushed out.

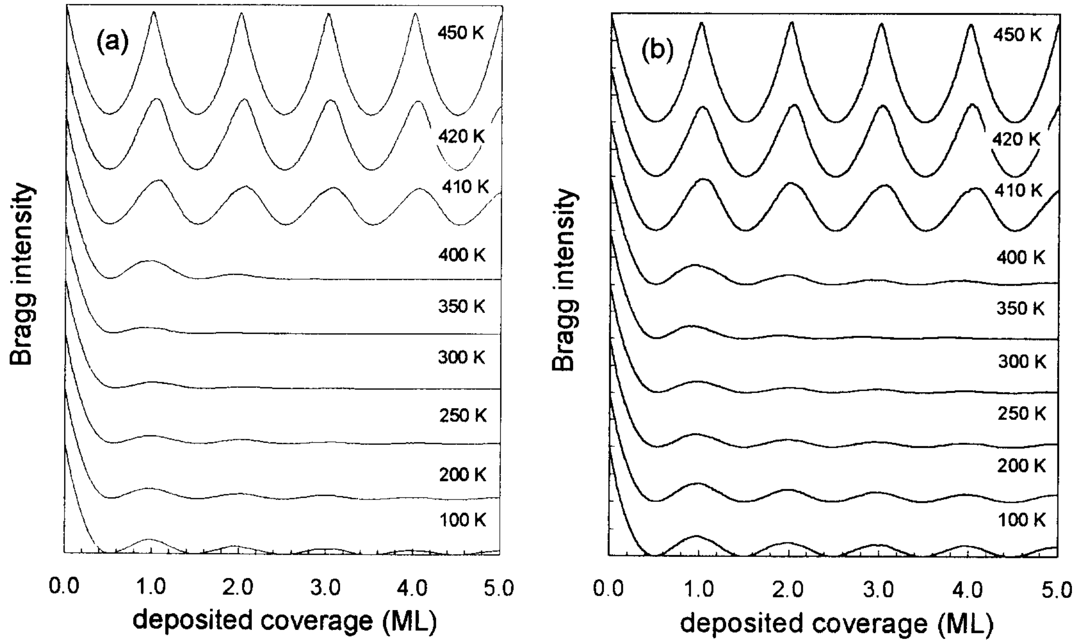


Fig.2 The Bragg intensities in kinematic approach for film growth at different temperature (a) the downward funneling model, (b) the energetic atom deposition

3. RESULTS AND DISCUSSION

With the above formula, we performed the kinetic MC simulation of Au/Au(001) thin film growth with energetic atom deposition and compared the results with the simulations based on downward funneling model. In Fig. 2 the Bragg intensity calculated with kinematic approach is plotted as a function of deposited coverage for downward funneling model and for energetic atom deposition at different growth temperature. In

our model, the reentrant growth can be observed from the change of Bragg intensity with the growth temperature. By compared the Bragg intensity of energetic atom deposition with that of downward funneling model, we can find the role of energetic atom in the film growth is related to the growth temperature. At the temperature below 410 K, The effect of energetic atoms is to increase the Bragg intensity of film growth and the lower the temperature the more obvious the effect. This result indicates that the role of energetic atom is to enhance the smoothness of surface and to promote the film growth in 2-dimensional growth mode. At the temperature above 420 K, however, the difference of Bragg intensity between in downward funneling model and in energetic atom deposition is not obvious any more. In fact we found that the Bragg intensity of energetic atom deposition at 450 K is slightly lower than that of downward funneling model by comparing the numerical results of simulations or by enlarging the Bragg intensity scale. These results imply that effect of energetic atoms is to roughen the surface of growing film.

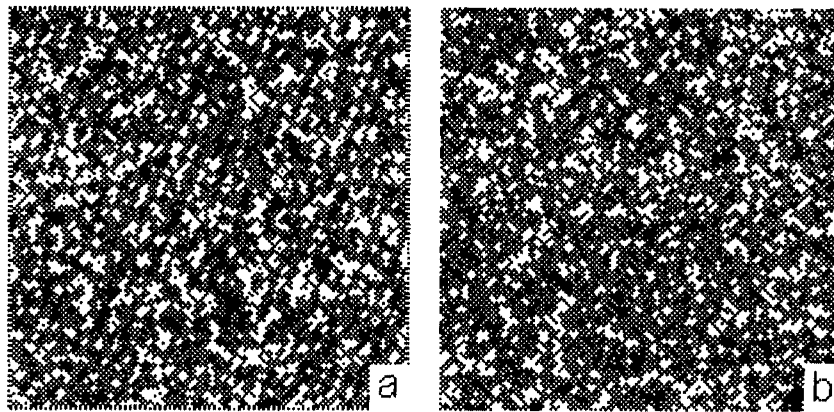


Fig.3 The snapshots of films after deposition of two monolayers at 200 K (a) the downward funneling model, (b) the energetic atom deposition.

By analyzing the mechanism of film growth, we can make clear and understand the role of energetic atoms deposition in film growth. At high temperature the film growth is in the mode of layer-by-layer due to the high mobility of adatoms. Because the mobility of adatom is much higher than the transient mobility induced by energetic atoms, the local environment change of surface caused by energetic atoms can be eliminated by the diffusion of adatom, so that the diffusion process still dominates the film growth in the mode of layer-by-layer at the case of energetic atom deposition. The effect of energetic atoms is to decrease the perfection of film, such as the increase of defects and adatoms. At low temperature, however, the adatom mobility is very low and islands with large scale cannot be formed by the diffusion process, so the film growth is in the mode of quasi-two-dimensional growth. In this case the transient mobility of energetic atoms can make the deposited atom to go longer distance than the atom in downward funneling model can. The energetic atom can also destroy the small islands formed on the surface. In fact, The role of energetic atoms is to enlarge the island size and lower the height of islands. In other words, the effect of energetic atoms in the deposition is to enhance the smoothness of film. The conclusion can be shown by the comparison of film

morphologies simulated at 200 K with downward funneling model and with energetic atom model as shown in fig. 3, where two monolayers were deposited. One can see that the morphology of film in energetic atom deposition is flatter than that in downward funneling model. The snapshots of films simulated are very similar to the images of scanning tunneling microscopy (STM) obtained by Esch et al. in IBAD experiments[18].

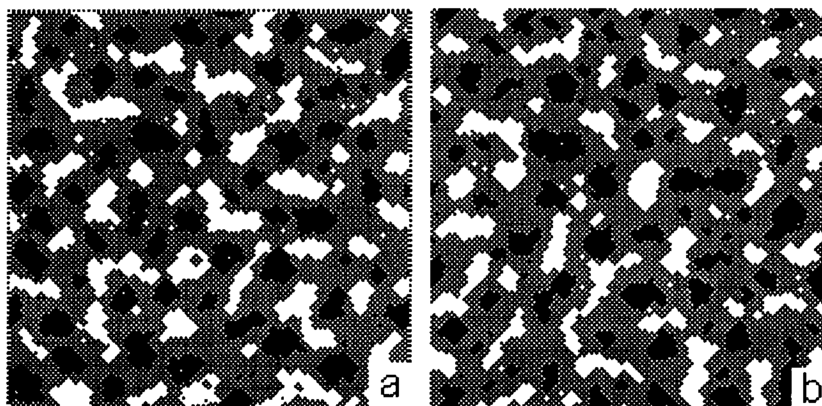


Fig.4 The snapshots of films after deposition of two monolayers at 400 K (a) the downward funneling model, (b) the energetic atom deposition.

At temperature of film growth between in layer-by-layer mode and in quasi-two-dimensional mode, namely the temperature range of 250 K to 410 K, the limit mobility of adatoms cannot drive the adatom to diffuse in long distance and there are not enough adatoms to hope down from upper layer, so that the film growth is in 3-dimensional mode. The role of energetic atoms is to suppress the 3-dimensional island growing large. In this case, the transient mobility of the deposited atom can cause the deposited atoms on top layer of small islands to hop down. At the same time, the adatoms with limit mobility can eliminate the defects produced by energetic atoms. The snapshots of films simulated with downward funneling model and with energetic atom deposition at 400 K are compared in fig.4, where the films were deposited for two monolayers. One can see that the number of atoms in the topmost layer in downward funneling model is more than that in energetic atom deposited model.

4. CONCLUSION

In conclusion, we have developed a simulation method to study the film growth of energetic atom deposition by using kinetic Monte Carlo simulation in combination with the results obtained from molecular dynamics simulation for the transient process. We found the role of energetic atoms is different in different film growth mode and at different temperature. At high temperature, energetic atom cannot affect the film growth mode when the film grows in layer-by-layer. At temperature of film growth in 3-dimensional mode and in quasi-two-dimensional mode, energetic atom can enhance the smoothness of film surface, which is similar to the results observed in experiments. The enhancement is caused by the transient mobility of energetic atoms and the suppression for the formation of 3-dimensional islands.

REFERENCES

1. C.Schwebel, F.Meyer, G.Gautherin, and C.Pellet, J. Vac. Sci. Technol. **B4** (1986) 1153
2. Y.Namba and T.Mori, J. Vac. Sci. Technol. **13** (1976) 693
3. E.Chason, P.Bedrossian, K.M.Horn, J.Y.Tsao, and S.T.Picraux, Appl. Phys. Lett., **57** (1990) 1793
4. J.M.Millunchik and S.A.Barnett, Appl. Phys. Lett., **65** (1994) 1136
5. C-H.Choi, R.Ai, and S.A.Barnett, Phys. Rev. Lett., **67** (1991) 2826
6. B.K.Kellerman, E.Chason, J.A.Floro, S.T.Picraux, and J.M.White, Appl. Phys. Lett., **67** (1995) 1703
7. E.Chason, P.Bedrossian, J.E.Houston, J.Y.Tsao, B.W.Doson, and S.T.Picraux, Appl. Phys. Lett., **59** (1991) 3533
8. E.Chason and B.K.Kellerman, Nucl. Instru. Methods, **127/128** (1997) 225
9. C.M.Gilmore and J.A.Sprague, Phys. Rev., **B44** (1991) 8950
10. J.A.Sprague and C.M.Gilmore, Thin Solid Films, **272** (1996) 244
11. A.Robbemond and B.J.Thijsse, Nucl. Instr.&Methods, **127/128** (1997) 273
12. M.Villarba and H.JOnsson, Surf. Sci., **324** (1995) 35
13. S.M.Foiles, M.I.Baskes, and M.S.Daw, Phys. Rev., **B33** (1986) 7983
14. M.Breeman, G.T.Barkema, M.H.Langelaar, and D.O.Berma, Thin Solid Films, **272** (1996) 195
15. A.F.Voter, Phys. Rev., **B34**(1986) 6819
16. J.W.Evans, D.E.Sanders, P.A.Thiel, and A.E.Depristo, Phys. Rev., **B41**(1990) 5410
17. J.Jacobsen, K.W.Jacobsen, P.Stoltze, and J.K.Nørskov, Phys. Rev. Lett., **74**(1995) 2295
18. S.Esch, M.Breeman, M.Morgenstern, and T.Michely, Surf. Sci., **365** (1995) 187