Journal of the Korean Vacuum Science & Technology; JKVST
Vol.6, No.1, 2002, pp.22～29

Multi-scale Simulation of Energetic Deposition: Form MD Simulation to KMC Simulation

Qing-Yu Zhang†

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

(Received)

Abstract

The process of energetic deposition has been simulated by using molecular dynamics (MD) simulation and kinetic Monte Carlo (KMC) simulation. The atomistic mechanisms of film growth in energetic deposition are discussed.

1. Introduction

It is very important to increase the deposition energy of atoms and/or ions in the thin film synthesis. The increase of deposition energy can considerably improve the properties of thin films synthesized. In the past years, many methods based on energetic deposition, such as ion beam assisted deposition (IBAD), accelerated molecular beam epitaxy, pulsed laser deposition (PLD), etc. have been developed and have become important techniques in film preparation. [1,2] Though it was observed that the use of energetic atoms in film deposition can result in the growth of thin film at lower temperature with better quality than that formed by vapor deposition [3-5], little has been know on the film growth mechanism of energetic deposition at an atomic level.

2. Modeling of Energetic Deposition: Md Simulation

In MD simulation, however, the process of film growth cannot be simulated in the huge number of atoms contained and in the time scale involved in experiment, so that a reasonable model should be established. In the present work, the simulation method is same as reference [6].

2.1. Evolution of kinetic energy of incident atom

Figure 1 shows the evolution of kinetic energy of incident atoms with time. The results are those averaged over 50 deposition events and the initial substrate temperature is 300 K. In the figure, we can see the kinetic energy of the incident atom increases first, the increment is about 2-3 eV, which originates from the latent heat of binding energy. The kinetic energy of incident atoms increases from the beginning to its maximum value in about 0.2-0.3 ps drops very sharply. After that, the kinetic energy dissipates in the form of damping oscillation in 1-2 ps. It is very interesting that the time spent in the process of kinetic energy dissipation of incident atoms is not obviously dependent on the incident energy of deposited atoms. This result implies that the incident atom loses its kinetic energy in a very short time (about 0.2-0.4 ps) after impinging on the surface. The excess energy

† E-mail : qyzhang@dlut.edu.cn

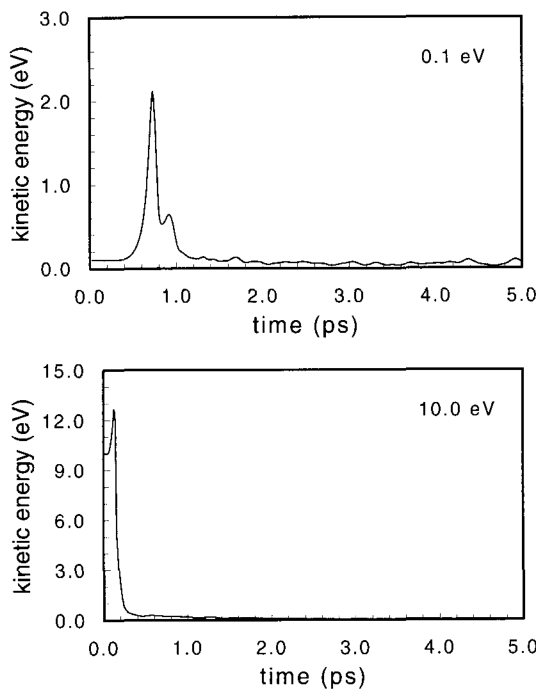

Fig. 1. Evolution of kinetic energy of incident atoms.

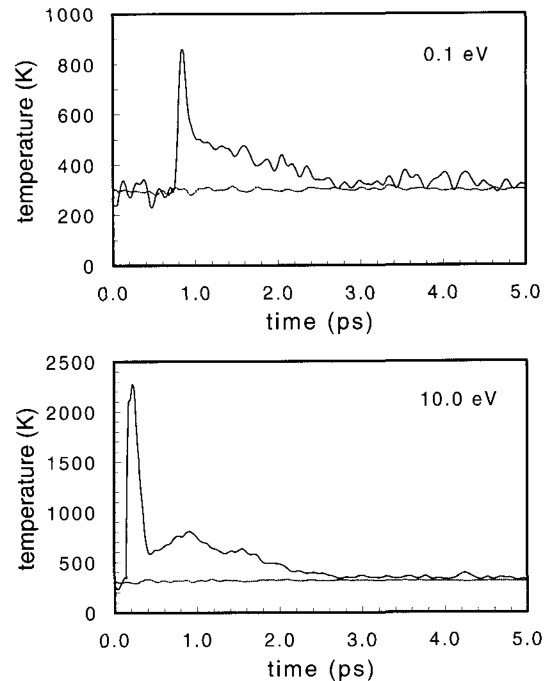

Fig. 2. Evolution of substrate temperature in region A (—) and region B (-----) .

dissipation of deposited atoms in the form of damping oscillation resulted from the diffusion of thermal energy in local environment.

2.2. Evolution of substrate temperature

To investigate the evolution of substrate temperature, the substrate is divided into two regions. Region A includes the atoms in the range of $R_A \leq r_{cut}$ around the incident point on the surface, where r_{cut} is the cut-off radius of Au-Au potential in EAM. Region B is in the range of $R_B \geq r_{cut}$. Figure 2 is the evolution of substrate temperature in regions A and B. In the figures, we can see that the temperature in region B is almost independent of time. The temperature in region A, however, changes sharply, and decreasing in the form of damping oscillation. There is a phenomenon that should be noted: the average temperature in region A after arriving its maximum value is decreased sharply, then declines in the form of damping oscillation. In 2.6-3.0 ps, the average temperature declines to the initial temperature in all cases of incident energies. This result shows that to study the film growth by using MD simulation method, the interval of atom deposition should not be less than 2.5-3.0 ps in the simulation model containing about 100 atoms per monolayer. In other words, the deposition rate should be less than 0.4 atoms/ps. Otherwise, the substrate will be heated continuously by the dissipation of kinetic energy of incident atoms and latent heats, and the substrate temperature will be increased. In our simulation, 3 ps are chosen as the interval of atom deposition.

2.3 Growth mode and morphology of thin films

Figure 3 shows that Bragg diffraction intensity of film growth with 0.1 eV incident atom declines in the

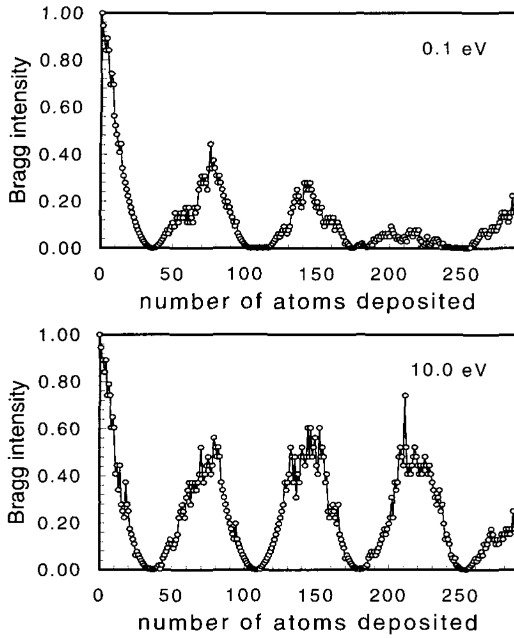

Fig. 3. Bragg diffraction intensities of films deposited with 0.1 eV and 10.0 eV incident atoms at 300 K.

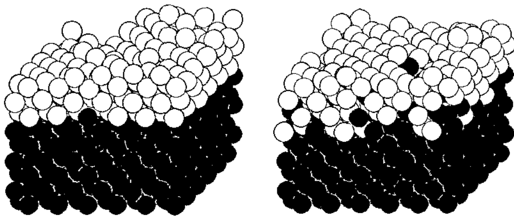

Fig. 4. Morphologies of films deposited with (left) 0.1 eV, (right) 10.0 eV incident-atom energies at 300 K.

form of damping oscillation; those obtained at incident atom energies of 10.0 eV, however, do not decline obviously. This result implies that Au-film growth with 10.0 eV incident atoms is very similar to layer-by-layer growth mode, *i.e.,* a Frank-van der Merwe growth (F-M growth). The film growth with 0.1 eV incident atoms, however, is more like Stranski-Krastanov growth (S-K growth), *i.e.* layer-by-layer growth followed by 3-dimensional island formation. In other words, the film growth mode changes from S-K growth to F-M growth with the increase of incident atom energy from 0.1 eV to 10.0 eV. This interpretation can also be confirmed by observing the morphology of thin film as shown in Fig.4, where the film (white atoms) is shown on the substrate (black atoms) after the deposition of 220 atoms (about 3 monolayers).

2.4 Effects of energetic atom on film growth

To reveal the transient motion of energetic atoms and their effect on the mechanism of film growth, the transport properties of deposited atoms and adatoms have been investigated. We define two kinds of lateral redistribution of atomic transport as shown in Figs. 5 and 6, where the statistical results are calculated with 288 deposition events. Figure 5 is the lateral redistribution of deposited atoms moved from the initial positions on the surface to the first surface sites

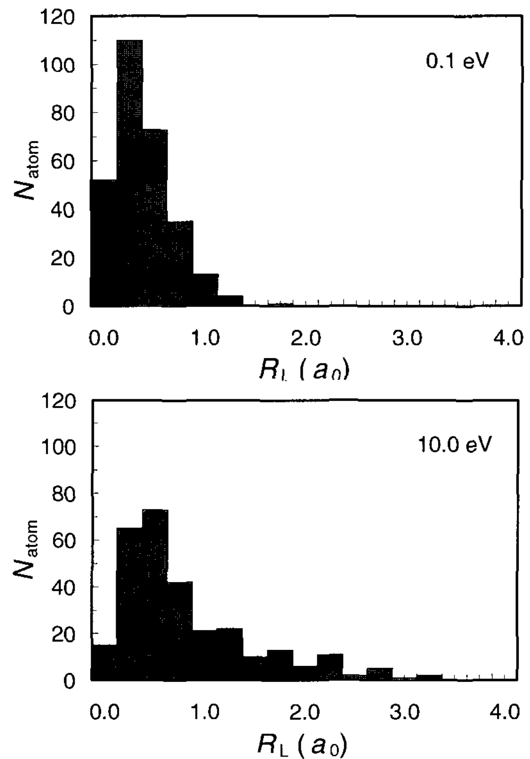

Fig. 5. Lateral redistribution of deposited atoms.

occupied, *i.e.* the occupied sites before the next incident atom is deposited. This redistribution can describe the transient motion of deposited atoms at surface in relaxation process and is related to the incident atom energy and surface morphology, hence we name this redistribution as the lateral redistribution of deposited atoms. Figure 6 is the lateral redistribution of deposited atoms displaced from the first occupied sites on the surface to the final occupied sites, *i.e.* the occupied sites after the end of the simulation. This redistribution can reflect the effect of incident atoms on adatoms and atoms in the film and the effect of diffusion caused by substrate temperature, and we call this redistribution as adatomic lateral redistribution.

In Fig. 5, we can see that the maximum values in distributions are about 0.5 a_0 for both high and low incident atom energies. This result means that most of incident atoms go to the nearest four-fold site around the incident point. The difference induced by the

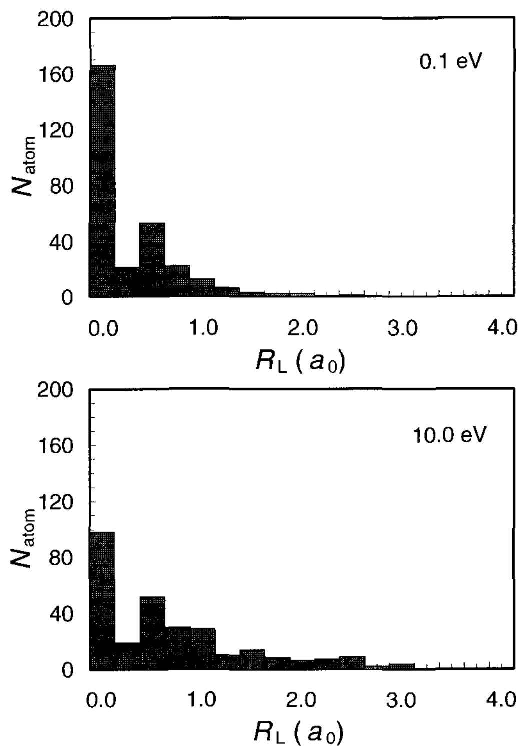

Fig. 6. Adatomic lateral redistribution.

incident atom energy is the change of lateral redistribution function. The number of atoms in the distribution at long distances is increased with increasing the incident energy. In other words, the increase of incident atom energy increases the "transient" mobility of deposited atoms. The increase of "transient" mobility of deposited atoms is one of the reasons, to prompt the change of the film growth mode from S-K growth to F-M growth.

In Fig. 6, adatomic lateral redistribution of 10.0 eV incident atom is different from that of 0.1 eV incident atoms. This result means that the deposited atom with low incident atom energy cannot displace adatom to a long distance. With the increase of incident atom energy from 0.1 eV to 10.0 eV, both "transient" mobility of deposited atoms and adatomic mobility are increased. The increase of "transient" mobility of deposited atoms and adatomic mobility prompts the film growth in layer-by-layer mode.

It should be pointed out that the conclusions are obtained without consideration of adatom diffusion on film surface. In other words, the results only or mainly reflect the effects of "transient" motion of deposited atoms. On the other hand, the results obtained by molecular dynamics simulation only reflect the change of local environment about the deposited atom in film growth. By considering the adatom diffusion, the film growth mode may be different from the results obtained above.

3. Modeling of Energetic Deposition: Kmc Simulation

In general, the deposition process is a transient dynamic process and can be studied by MD simulation. The diffusion process, however, generally occurs in nanoseconds to seconds depending on the temperature and deposition rate. In the diffusion process, some events are very important to the film growth, such as the motion of adatoms in large scale, the nucleation of adatoms, the growth of islands, and

the recombination of adatoms with vacancies. It is impossible to consider the diffusion process with enough simulation time in the MD simulation. In this study, based on the MD simulation results of deposition process of energetic atoms, a method has been devised to describe the change of local morphology in deposition process and then applied to the investigation of the film growth with energetic deposition by kinetic MC simulation.

3.1 Nucleation and morphology of film growth in early stages

Figure 7 contains the perspective-view images of simulated surfaces at various temperatures. The images represent the surfaces of film growth after 0.2 ML energetic atoms have been deposited on substrates. We can see that the morphology of film growth is dominated by the substrate temperature. At the temperature of 100 K, the main morphological entities on the surface are monomers, dimers, trimers, small clusters and monomer strings. The morphology of film growth at low temperatures results from the low mobility of adatoms. The mobility of an adatom can be quantitatively described by the hopping rate of the adatom in a certain time. We calculated the hopping rate of adatoms with some typical local environments as a function of temperature in the time of deposition interval. Calculation results indicate that the hopping rate of adatom with any local environment is much less than 1.0 when the temperatures below 200 K. This means the nucleation and island growth at low temperatures mainly depend on the increase of the adatom number density and the transient move of energetic atom on the substrate. With the increase of substrate temperature, monomers and dimers considerably decrease and islands obviously grow up. At the temperature of 300 K, monomer strings are the major morphological entities on the surface. At the temperature above 400 K, however, the morphological entities are mainly islands. The shape of islands on the surface at

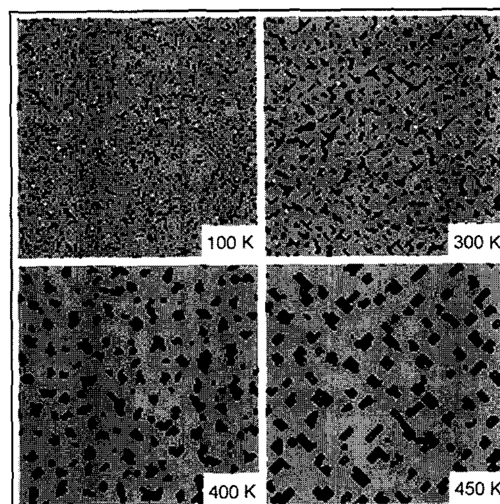

Fig. 7. Simulated film surfaces taken at various temperatures after deposition of 0.2 ML energetic atoms.

450 K is more regular than that at 400 K. This phenomenon can be attributed to the increase of diffusion capability of the adatom. Adatom diffusion dominates the nucleation, island growth, and the change of island shape. The calculation results of hopping rates show that dimer and monomer strings are no longer stable and the interlayer diffusion of adatoms can occur with high probability at the temperatures higher than 400 K. Those are the main reasons causing the morphology change with the increase of temperature.

3.2. Growth mode and atomistic mechanism

The Bragg intensity calculated with a kinematic approach and roughness of film surface versus deposited coverage at various temperatures are shown in Fig. 8. In the figures we can see that the Bragg intensity oscillates in the form of similar amplitude at the temperatures higher than 410 K. This implies the film growth proceeds in the form of an almost perfect layer-by-layer mode at these temperatures. For temperatures of 400 K to 300 K, Bragg intensity

considerably decreases in the form of damping oscillations with the increase of coverage. In these cases, film growth is a typical 3-dimensional island growth mode. At the temperature below 250 K, the damping of the oscillations decreases and the amplitudes of Bragg intensity oscillations increase. When the temperature decreases down to 100 K, the Bragg intensity oscillates in the form of almost the same amplitude again though the amplitude of Bragg intensity oscillations is much lower than that at high temperatures. A similar phenomenon of film growth was first discovered by Kunkel et al. in the experiment of homoepitaxial Pt(111)-films growth and was named as "re-entrant layer-by-layer growth" or quasi-two-dimensional layer-by-layer growth [7]. The behavior of film growth with energetic deposition is similar to the simulation result with conventional vapor phase deposition. In other words, the film growth mode is mainly dominated by the temperature of film growth.

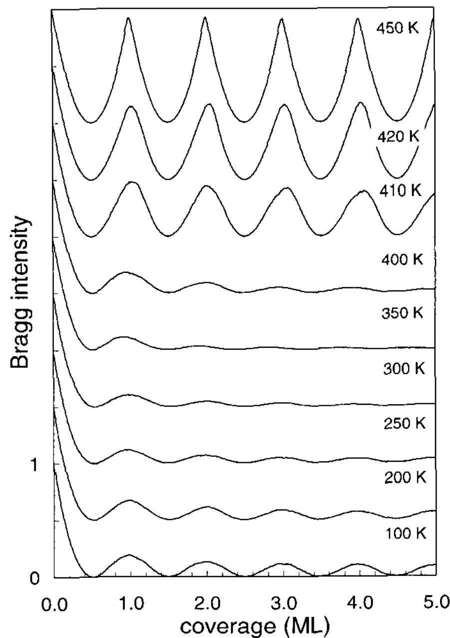

Fig. 8. Bragg intensity of film surface versus coverage taken at various temperatures after deposition of 5.0 MLs energetic atoms.

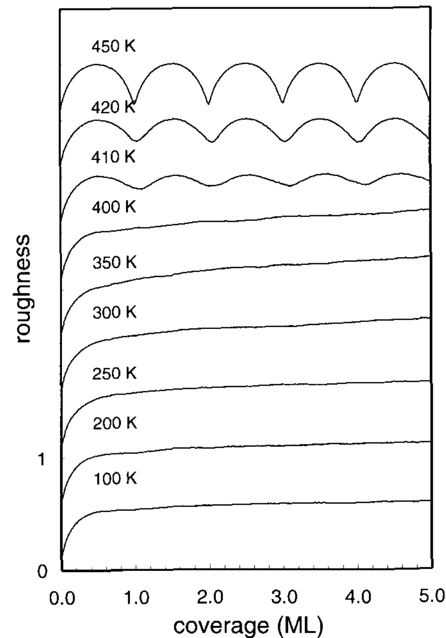

Fig. 9. Roughness of film surface versus coverage taken at various temperatures after deposition of 5.0 MLs energetic atoms.

Although the behavior of Bragg intensity oscillation at low temperature is similar to that at high temperatures, the nature of film growth for the two cases is very different. From the change of roughness of film surface at various temperature as shown in Fig. 8, we can see that the roughness of film growth changes in the form of oscillation with the increase of coverage at high temperatures. This oscillation of roughness is a feature of real layer-by-layer growth. At low temperatures, however, the change of roughness is the linear increase with the increase of coverage after 0.5 MLs atoms are deposited. In the view point of roughness the film growth at low temperatures is the same as 3-dimensional island growth. The enhancement of Bragg intensity oscillation is the downward funneling effect caused by numerous small islands. At low temperatures, islands grow up very slowly because of the low mobility of adatom. The morphological entities of film growth are monomers and small islands. These small islands induce the efficient interlayer mass transport. This also

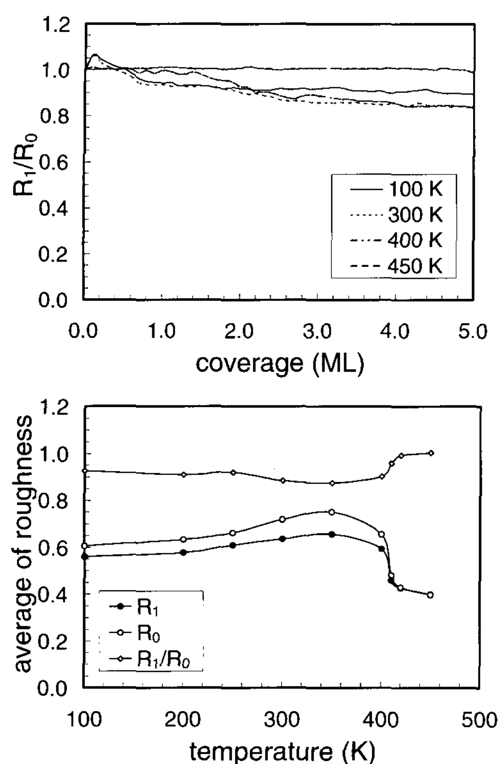

Fig. 10. (a) Evolution of roughness ratio of energetic deposition to conventional vapor phase deposition with the increase of coverage at various temperatures, (b) Average roughness of films at different temperature, where R_0 is roughness of conventional vapor phase deposition and R_1 is roughness of energetic deposition.

means that the morphology of film growth is rough on an atomic scale at low temperatures.

To reveal the role of energetic atom in the film growth, we compared the simulated results with those obtained in the simulation of conventional vapor phase deposition with the downward funneling model [8]. At the temperatures of 100 K to 200 K, the Bragg intensity of film growth with energetic deposition increases by about 40%. For temperatures of 250 K to 400 K, the increase of Bragg intensity is about 20%. At the temperatures higher than 410 K, the increase of Bragg intensity sharply drops down to 0 when increasing the temperature up to 450 K. In comparison to conventional vapor phase deposition, the roughness of film growth with energetic deposition decreases about 15% in the temperatures of 100 K to 400 K. For the temperatures above 410 K, the roughness of energetic deposition is almost the same as that of conventional vapor phase deposition. These results imply that the role of energetic atoms in film growth is to enhance the smoothness of morphology at the temperatures below 400 K. At the temperatures higher than 410 K, the influence of energetic atom can not dominate the behavior of film growth due to the increase of diffusion capability of adatoms.

The evolution of roughness ratio of energetic deposition to conventional vapor phase deposition with the increase of coverage at various temperatures and the average roughness of film growth are shown in Fig.9. We can clearly see that the roughness of film growth with energetic deposition is increased in the early stages of film growth (the coverage below 0.5 MLs) at low temperatures. The increase of roughness can be interpreted by the sputtering effect of energetic atoms on substrates. The impact of energetic atoms with substrate increases the defects on substrate. On the another hand, the increase of adatom number density induced by sputtering effect promotes the nucleation of film growth in the early stages. After the energetic atoms are deposited to more than 1 ML, the roughness of film growth is much lower than that in conventional vapor phase deposition at the temperatures of 100 K to 400 K. The decrease of roughness can be attributed to the contribution of sputtering effect of energetic atoms and the results of transient mobility of deposited atoms. On the one hand, the energetic atoms suppresse growth of 3-dimensional islands by a sputtering effect, on the another hand, the transient mobility of energetic atom induces the efficient interlayer mass transport.

4. Conclusions

(1) At the substrate temperature of 300 K, film

growth mode changes from S-K to F-M growth with the increase of incident atom energy from 0.1 eV to 10.0 eV. The reason of film growth mode change can be attributed to the increase of "transient" mobility of deposited atoms and adatoms.

(2) In the early stages of film growth, the morphology of film growth with energetic deposition is mainly dominated by the substrate temperature. The role of the energetic atom is to influence the local morphology of film growth. The influence of energetic atoms on the morphology of film growth is greater at low temperature than that at high temperature.

(3) Energetic atoms can promote the nucleation and island growth in the early stages at low temperatures. The promotion of the nucleation and island growth can be attributed to the increase of adatom number density and the low diffusion capability of adatom.

(4) The mode of film growth with energetic deposition is mainly dominated by the temperature. The morphology of film growth with energetic deposition is smoother than that with conventional vapor phase deposition at the temperatures of 100 K to 400 K. The role of energetic atom in the film growth is to suppress the growth of 3-dimensional islands and to increase the interlayer mass transport.

References

[1] A. Rockett, S. A. Barnett, and J. E. Greene, J. Vac. Sci. Technol. B **2**, 306 (1984).

[2] D. Marton, in J. W. Rablais (ed.), "*Low Energy Ion-Surface Interactions*", (Wiley, New York, 1994) p482; K. Miyake and K. Ohashi, Jpn. J. Appl. Phys. **32**, L120 (1993).

[3] C. Schwebel, F. Meyer, G. Gautherin, and C. Pellet, J. Vac. Sci. Technol. B **4**, 1153 (1986).

[4] Y. Namba and T. Mori, J. Vac. Sci. Technol. **13**, 693 (1976).

[5] M. Kawasaki and H. Uchiki, Surf. Sci. **388**, L1121 (1997).

[6] Q.Y. Zhang, Z.Y. Pan, and J.Y. Tang, Acta Phys. Sinica(Oversea Edition), **8**, 296 (1999).

[7] R. Kunkel, B. Poelsema, L. K. Verheij, and F. Comsa, Phys. Rev. Lett. **65**, 733 (1990).

[8] Q.Y. Zhang, T.C. Ma, Z.Y. Pan, and J.Y. Tang, Acta Phys. Sinica **49**, 297 (2000), in Chinese.