



HAL
open science

COMPUTER SIMULATIONS OF THIN FILM GROWTH FOR HOMO AND HETERO EPITAXY

T. Kaneko, R. Yamamoto

► **To cite this version:**

T. Kaneko, R. Yamamoto. COMPUTER SIMULATIONS OF THIN FILM GROWTH FOR HOMO AND HETERO EPITAXY. Journal de Physique Colloques, 1990, 51 (C1), pp.C1-843-C1-848. 10.1051/jphyscol:19901132 . jpa-00230042

HAL Id: jpa-00230042

<https://hal.science/jpa-00230042>

Submitted on 4 Feb 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

COMPUTER SIMULATIONS OF THIN FILM GROWTH FOR HOMO AND HETERO EPITAXY

T. KANEKO and R. YAMAMOTO

Department of Materials Science, Faculty of Engineering, University of Tokyo, Tokyo 113, Japan

/Abstract/ We simulated a growth of metal films with two models of surface diffusion and investigated the relationships between the evolution of surface step densities and growth condition. In one case, we modeled thermally activated surface migration by the Arrhenius relation. When the substrate temperature was above 430K, clear periodic oscillations of the number of steps on the surface were observed. In the other case, epitaxial growth at 0K was simulated in which we modeled that deposited atoms can move by the hopping which use the kinetic energy of the atom. The step density can oscillate with one monolayer growth period when an impinged atom could hop more than 10 times in one direction even at 0K. In the random-hopping-migration model, when deposited atom can hop more than 28 times, the step density oscillation can be obtained.

1-INTRODUCTION

Since the discovery of intensity oscillations in reflection high energy electron diffraction (RHEED) during epitaxial growth a few years ago, they have been the subject of intensive investigation both experimentally and theoretically. The observation of temporal oscillations of RHEED beam intensity with a period equal to the one monolayer growth time offers a reliable measure of the growth rate and has provided the most direct evidence for the layer-by-layer growth mode. In the field of the crystal growth of metal films, RHEED beam intensity oscillations were observed for the first time during the growth of Cu in a Cu/Mo multilayered film at 1986¹⁾. Later, similar oscillations were observed at a number of materials and the same interpretation as in semiconductor epitaxy in terms of a periodically varying surface step density in unit area has been accepted.

To date, a variety of computer simulations have been made to understand and control the crystal growth of films, in particular for semiconductor films. In Section 2, we modeled surface migration by the Arrhenius relation and we have shown that the substrate temperature above 430K is needed for observation of the oscillation of surface step density. This substrate temperature can afford such a sufficient surface diffusion in which that an adatom is able to jump to one of the nearest-neighbor sites 400 times during the growth of one monolayer. However, in the RHEED experiments of epitaxial metal growth, RHEED intensity oscillations occur at very low temperature in the cases of Ni and Mo growing on W²⁾, Au and Ag on Si³⁾, Cu and Fe on Ag⁴⁾, whereas the surface diffusion length in these cases is almost zero. The cause of the difference between the experimental and simulated of temperature at which RHEED intensity oscillation can be observed is not well understood. In the present paper, we have investigated

the possibility that deposited atoms can move across a surface not only by thermally activated surface diffusion process but also by the hopping due to the kinetic energy of a deposited atom and RHEED intensity oscillation at low temperatures.

2-GROWTH BY THERMAL ACTIVATED SURFACE DIFFUSION

The basic assumptions used in our model are as follows. Only single atoms are mobile and impinge on the substrate surface. Mobile atoms migrate on the surface through jumping to one of the nearest-neighbor sites. Only the nearest-neighbor interactions are taken into account in computing the potential energy. The surface of the substrate is free from the sites of preferred nucleation, e.g., defects or impurities. The periodic boundary conditions are employed. An atom that has jumped up from the upside border of the lattice appears again at the bottom of the lattice. No decomposition of clusters occur. Reevaporations of atoms from the surface are not considered in the simulations. We assumed the temperature of the surface to be low enough for neglecting reevaporation.

An fcc lattice in which the [111] direction is perpendicular to the film surface was used in this study. The sample lattice with 400 (20x20) sites was employed. We modeled surface migration by the Arrhenius relation. Surface diffusion coefficient D_s is expressed as

$$D_s = D_0 \exp(-E_s/kT) , \quad (1)$$

where k is Boltzmann's constant, T the temperature of the substrate, and E_s the energy barrier to diffusion of an atom, respectively. According to Gjostein⁵⁾, D_s of fcc metals is given by the following expression in the substrate temperature range T , $T/T_m < 0.75$ where T_m is the melting point,

$$D_0 = 0.014 \text{ cm}^2/\text{s} \quad (2)$$

and

$$E_s = 13T_m \text{ cal/mol.} \quad (3)$$

The prefactor D_0 for Ni was set as $0.014 \text{ cm}^2/\text{s}$ and E_d as 0.975 eV , respectively. A free atom can jump in one of six possible directions on a trigonal lattice. In the present calculations, the actual direction of each jump was selected by means of a random number generator.

Five simulation runs were carried out for each substrate temperature. We monitored the growth by summing the number of uncombined bonds parallel to the surface. This provides a measure of the number of steps. The averaged values of the number of steps on the surface were calculated for five runs.

Time dependences of the number of atomic steps on the surface of 20x20 sites are shown in Fig.1 for the range of substrate temperature from 160K to 470K. We used 'layers' here as a unit of time in the horizontal axis in Fig.1. One layer denotes the required time for the impingement of enough atoms for one monolayer growth. Since the deposition rate in the present simulations was set at 0.5 monolayer/s , one layer is equivalent to two seconds. Below 400K, periodic oscillations of the number of steps are not observed, and the number of steps increases rapidly until 0.5 monolayer growth and then continues increasing slowly. At 400K, the number of steps increases until 0.5 layer but then decreases a little until one monolayer growth. Above 430K, the oscillations continue over 10 times. The maximum number of steps of each oscillation shows no change at 470K, but the minimum number of steps increased gradually. In Fig.2, we have shown the growth configurations of the surface after the deposition of enough atoms for five complete monolayers at substrate temperatures of 160K (Fig.2(a)) and 470K (Fig.2(b)). The growth mode is clearly three dimensional at 160K, and there are four incomplete layers from the third to the sixth layer. At 470K the third layer has been completed and the number of incomplete layers is three. Even at 470K, the growth was not completely layer-by-layer mode, whereas the number of steps showed the oscillatory behavior. A large difference in the configurations of deposited atoms between the growth at 160K and that at 470K was the number of clusters. The effect of enhanced surface diffusion is apparent by the smaller number of growing cluster at higher temperatures.

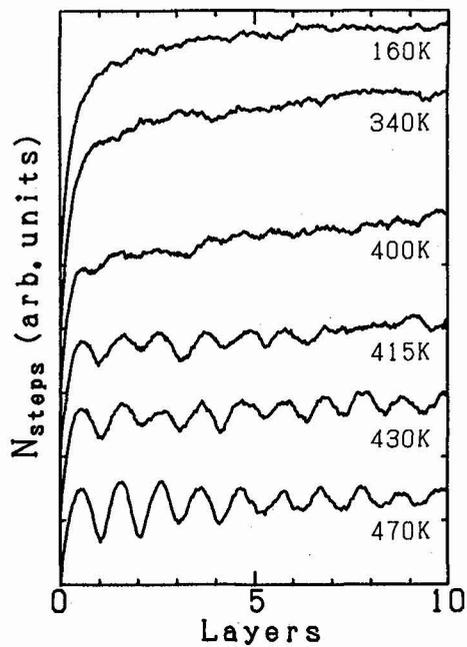


Fig.1 The time dependences of the number of steps on growing surface in thermal activated surface diffusion model.

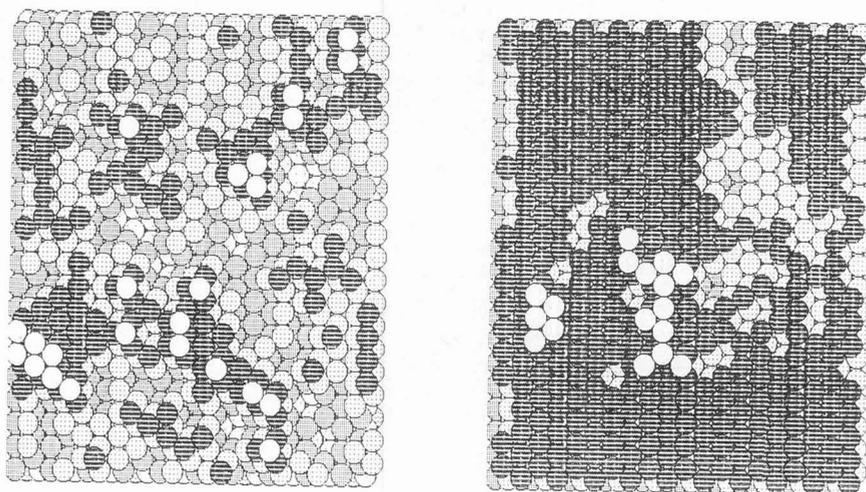


Fig.2 The growth configurations of surface after the deposition of enough atoms for five complete monolayers at substrate temperatures of 160K(a) and 470K(b).

The measured RHEED intensity oscillations always show a damping of their amplitude of oscillations. Van Hove and Cohen⁶⁾ have found that the damping envelope of measured intensity oscillations is qualitatively different for low-index and vicinal surfaces. They found that on a low-index surface, there was an initial transient, after which the intensity oscillation maxima damped slowly to a nearly constant, nonzero value. In contrast, on vicinal surfaces the intensity oscillations damped rapidly, and the maximum and minima decayed to an intermediate steady state value.

In Fig.3, the measured RHEED intensity oscillation during the growth of Ni on a W substrate²⁾ and the calculated step density oscillation at 470K are shown. Since a higher step density implies a lower RHEED intensity, the ordinate of the graph in Fig.3 (b) is inverted. Their oscillatory behaviors resemble to a typical damped oscillation on a low-index surface. The maximum number of steps of each oscillation show no change but the minimum number of steps increases gradually.

In computer simulations we needed a substrate temperature above 430K for observation of the oscillation of the number of steps. At this substrate temperature a sufficient surface diffusion length is giving so that an adatom is able to jump to one of the nearest-neighbor site 400 times during the growth of one monolayer.

3-DIFFUSION BY KINETIC ENERGY OF EVAPORATED ATOM

A growing surface was simulated by 64x64 point square grid resembling a (001) substrate of a simple cubic lattice for a convenience of calculations. Some test runs were also made on the 32x32 point and 16x16 point lattices.

We have set the substrate temperature to be 0K. At 0K re-evaporation and thermally activated surface diffusion can be neglected. The migration of an atom was assumed to take place by using its kinetic energy in the vapor phase.

The arrival of an atom to the growing surface has been simulated by generating atoms at random points on the grid. It was assumed that only single atoms impinge upon the surface. Any atom can only migrate once just after its arrival on the surface. The migrated atom can hop H times in one migration process and H is only one adjustable parameter which we used in the present study. Since we assumed that only single atoms are mobile on the surface, the migrations were terminated when other atoms existed at the nearest-neighbor sites of the migrating atom. Two different type of migration were considered. One type is the one-directional-hopping-migration, in which the hopping direction of an impinged atom is randomly selected from among four possible directions once at its arrival. Then the atom hops to the nearest-neighboring site in this direction. The other type is the random-hopping-migration. In this model of migration, the direction

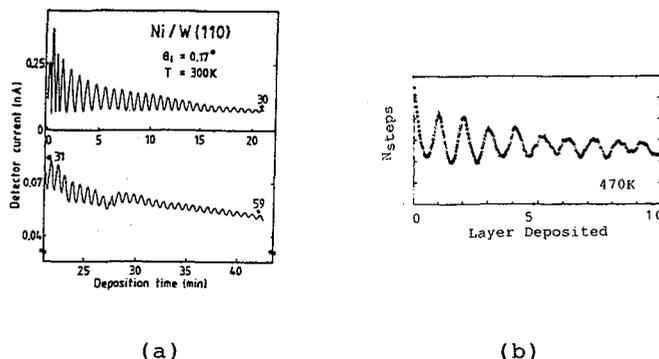


Fig.3 (a) Measured RHEED intensity oscillation in the case of Ni on W(110). (b) The evolution of the number of steps during the growth at 470K. Note that the ordinate has been inverted to correspond with (a).

of hopping is selected at random each time before its hopping. So, atoms migrate by the random walk.

Very slow deposition rate was assumed. After the preceding atom impinged and already migrated the next atom was allowed to impinge. The crystal growth was simulated in the presented model by the repetition of the impingement of an atom and its successive migration.

In the one-directional-hopping-migration model the time dependences of the step density on the surface of 64x64 sites are shown in Fig.4 for the range of the number of hopping H from 0 to 64. When no migration of deposited atoms take place ($H=0$), periodic oscillation of the step density was not observed and the step density increased rapidly. For the case in which the number of hopping is 8 times the oscillation with small amplitude appeared shown until the growth of five layers and then the step density slowly increased. When we set H at above 16, the oscillation with a regular amplitude continue until 10 layers. In Fig.5, the evolution of the step density with time is shown for the range of the number of hopping from 5 to 11 in detail. At $H=5$ there occurs no oscillation. For the range of H for 6 to 9 small irregular oscillations can be observed shown but it does not continue to ten layer growth. When H is larger than 10, the oscillation became settled and had regular period exactly equal to the one monolayer growth time.

In the case of the random-hopping-migration the time dependences of the step density on the surface of 64x64 sites are shown in Fig.6 for the range of the number of hopping H from 4 to 64. When H is below 12, no oscillation has occurred. Stable oscillation with a period equal to the one monolayer

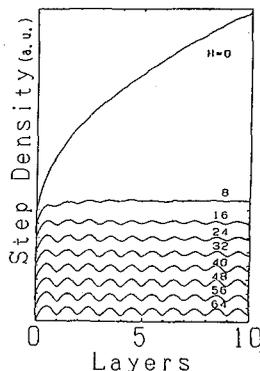


Fig.4 The time dependences of the step density in the one directional migration model.

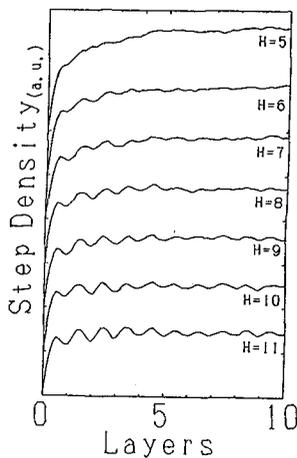


Fig.5 The step density evolution in one directional migration model.

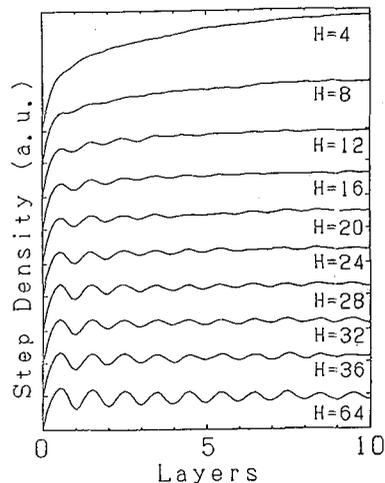


Fig.6 The time dependences of the step density in the random-hopping migration model.

growth time could be shown, when the number of hopping H is set at greater than 28. In the random walk theory effective migration length is given by $(Ha)^{-1/2}$, where a is the distance between the nearest-neighboring sites. When H is 28, effective migration length is about $5a$. It is astonishing that the step density of growing surface could oscillate even when the deposited atoms are able to migrate such a short effective migration distance.

Compared the one-directional-migration model with the random-hopping-migration one at $H=64$, the damping envelope of oscillations was qualitatively different. It was found that in the one-direction model the amplitude of step density oscillation is nearly constant and the minima and the maxima did not change. In contrast, in the random-hopping model the oscillations of the step density damped gradually and the maxima and minima of the step density decay to an intermediate steady state value.

4-CONCLUSION

We simulated a growth of metal films with two models of surface diffusion. In one case, we modeled thermally activated surface migration by the Arrhenius relation. When the substrate temperature was above 430K, clear periodic oscillations of the number of steps on the surface were observed. In the other case, epitaxial growth at 0K was simulated and we modeled that deposited atoms can move by the hopping which use the kinetic energy of the atom. The step density can oscillate with one monolayer growth period when an impinged atom could hop more than 10 times in one direction even at 0K. In the random-hopping-migration model, when deposited atom can hop more than 28 times, the step density oscillation can be shown.

REFERENCES

- /1/ Kaneko, T., Imafuku, M., Kokubu, C., Yamamoto, R. and Doyama, M.: J.Phys.Soc.Jpn. 55 (1986) 2903.
- /2/ Lilienkamp, G. Kozioł, C. and Bauer, E.: 'Reflection High-Energy Electron Diffraction and Reflection Electron Imaging of Surfaces' eds. P.K.Larsen and P.J.Dobson (Plenum Press, New York, 1988) p.489.
- /3/ Jałochowski, M. and Bauer, E.: Phys.Rev.B 38 (1988) 8622.
- /4/ Egelhoff, Jr., W.F. and Jacob, I.: Phys.Rev.Lett. 62 (1989) 921.
- /5/ Gjostein, N.A.: 'Surface and Interfaces' (Syracuse Univ.Press, 1967).
- /6/ Van Hove, J.M. and Cohen, P.I.: J.Cryst.Growth 81 (1987) 13.