Kinetic Monte Carlo simulations of three-dimensional self-assembled quantum dot islands*

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By three-dimensional kinetic Monte Carlo simulations, the effects of the temperature, the flux rate, the total coverage and the interruption time on the distribution and the number of self-assembled InAs/GaAs (001) quantum dot (QD) islands are studied, which shows that a higher temperature, a lower flux rate and a longer growth time correspond to a better island distribution. The relations between the number of islands and the temperature and the flux rate are also successfully simulated. It is observed that for the total coverage lower than 0.5 ML, the number of islands decreases with the temperature increasing and other growth parameters fixed and the number of islands increases with the flux rate increasing when the deposition is lower than 0.6 ML and the other parameters are fixed.

Keywords: Monte Carlo simulations, self-assembled quantum dot islands, growth parameters

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1. Introduction

Self-assembled quantum dots (QDs) are intensively studied due to their unique optical and electronic properties, with potential applications in optoelectronics and semiconductor devices.[1,2] The InAs/GaAs hetero-structure is a typical example; it is characterized by a large lattice mismatch and undergoes a growth transition from a two-dimensional (2D) layer to three-dimensional (3D) islands.[3,4] The optical properties of the 2D layer and 3D islands have also been studied.[5]

Experimental data indicate that the synthesis of an InAs/GaAs island array is sensitive to ambient growth parameters, exhibiting both thermodynamic and kinetic features.[6,7] The mechanisms behind the transition from flat pyramids to sharp domes have been studied intensively.[8–13] Using atomic force microscopy, Arciprete *et al.* showed clearly that different coverage usually corresponds to different QD size. The transition from thermodynamically to kinetically controlled QD self-assembly was also studied by Musikhin *et al.*[14] both experimentally and theoretically.

It is argued that the surface diffusivity and mobility could be affected by the strain field on the surface and even by the quantum size effect.[15–19] Recent first-principles calculations of Si and Ge adatoms on Si (001) and Ge (001) surfaces clearly show the correlation between the surface diffusion and mobility and the applied strain field.[15] The diffusion anisotropy of adatoms due to the applied strain field was also observed,[15] and enhanced evaporation caused by the local strain was reported.[20] An epitaxial system can maintain a lower free energy by transferring atoms from the island edge to the upper layer, because the transition leads to a decrease in the contact area between the substrate and a new layer.[13] Thus, atomic transitions to the upper layers lead to a relaxation in the local strain field. The tradeoff between the cost of the additional surface energy and the gain of energy due to elastic relaxation is the very driving force for the transition from flat pyramids to sharp domes.[11,12,18,19,21,22] These arguments provide a reason to consider situations in which the atom hopping probability to an upper layer can be higher than that to a lower layer. The kinetics of such a process could be described by adjusting the probability for the atoms to hop up or hop down, which could subsequently lead to a flat cluster or sharp 3D island self-assembly, and ultimate control over the synthesis mechanisms.

While some simple computational approaches for QD self-assembly have been discussed before, e.g., Ref. [23], no complete 3D QD self-assembly model has been developed so far in which the growth from a flat cluster to 3D islands and the corresponding island size equalization can be clearly illustrated during the growth process. Such a model would be vital to understanding the parametric variations of the surface patterns to the conventional growth parameters, such as flux rate, interruption time and temperature. We therefore propose a fast multiscale 3D kinetic Monte Carlo (KMC) QD growth model. Furthermore, the depositions with different growth parameters (temperature, flux rate and total coverage) are studied to show how they affect the equalization and the number of is-

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lands during the deposition process in the InAs/GaAs (001) system. Our model has also shown clearly the importance of the interruption time during the growth of QD islands. We believe that this model will provide an attractive means for simulating the strain-controlled ordering of QDs on surfaces. We note that, while the material parameters in our simulation are for InAs/GaAs (001), the two species In and As in the InAs QD islands are assumed to be equal. In other words, they are indistinguishable in the KMC simulation.[24,25]

This paper is organized as follows. In Section 2, we present the structure models and kinetic Monte Carlo simulations used in this work. The influence of the growth parameters (temperature, flux rate, total coverage and interruption time) on self-assemble InAs/GaAs QD islands are shown in Section 3. Finally, the conclusion is drawn.

2. Kinetic Monte Carlo simulations

In order to check the reliability of the proposed technique, we perform the KMC simulations. In our imitation process, it includes the deposition of the third material atoms and the diffusion of the atoms on the substrate. Starting with a perfect, defect free wetting layer, which is not simulated explicitly, we deposit adatoms randomly with a given flux rate. The probability distribution of random numbers is uniform. The models are based on a 2D solid-on-solid model[26,27] of epitaxial growth presented by Pan *et al.*[28] and Lam *et al*,[29] which assumes a simple cubic lattice structure with neither vacancies nor overhangs. Our models are developed from the former work.[30] In order to account for the adsorption and desorption motion of atoms, an up-down ratio is also introduced. We all know that the direction in which the atom in the same plane moves to is random and the probability of the movement is equivalent. The atoms on the substrate can still diffuse after the nucleation unless their neighboring positions are all occupied by other atoms. The hopping rate of the top most atom follows the Arrhenius law[31]

$$p = \upsilon_0 \exp\left(\frac{E_S + E_N + E_{SW} - \Delta E}{\kappa_B T}\right), \tag{1}$$

where ΔE is the strain energy, $\kappa_B T$ is the thermal energy, κ_B represents the Boltzmann constant, υ_0 is the attempt frequency and E_S and E_N are the binding energies to the surface and the neighboring atoms, respectively. In order to explain the motion of the atoms, an additional parameter, Schwöbel step-edge barrier term E_{SW}, is introduced. Most former works just took the neighboring atoms as being in the same plane, however, the neighboring atoms in the up and the down layers should be considered in a 3D simulation. So, in our work, we take into account the interaction between the neighboring atoms in different planes.

We know that there are three kinds of bondings between two different atoms, as shown in Fig. 1. As a result of the disparate bonding mode, the bonding energies are diverse between the two atoms, which can be expressed as E_{AA}, E_{AB} and E_{BB}. Figure 2 illustrates all the atom diffusion processes considered in our simulations, which include an adatom moving on a flat terrace, stepping down at a step edge and detaching from a step edge. The activation barriers of the diffusion processes are determined by the local bonding of the initial configuration and the interatomic bond energies. For the adatom terrace diffusion, the activation barrier is given by the bond energy of the adatom to its neighbor underneath. The activation barrier for the adatom stepping down is the terrace diffusion barrier plus the additional Schwöbel step-edge barrier. Similarly, the activation barrier for the adatom detaching from a step edge is the terrace diffusion barrier plus the bond energy of the adatom to the neighboring atom at the step edge. In the simulations, we take into account the three two-component situations mentioned above. It should be noted that dealing with effective atom hopping on a square lattice and the usage of effective energies have to be understood as a mean-field approach. Implicitly, properties which lead to local fluctuations of the binding energies are taken into account. Such properties include, for example, the surface reconstruction and changes in the ratio, which might occur in both time and space. As long as these fluctuations are small compared to the time scale of the fastest process modeled explicitly, there is no expected influence to the validity of our model. However, for convenience, we simplify the simulations by using equation $E_{AA} = E_{AB} = E_{BB}$. We firstly set the strength of all single nearest bonds E_b to be 0.2 eV and reduce it by a factor δ for the next nearest neighbors. To calculate the diffusion barrier, the binding energy $E_{n,initial}$ at the initial site and the binding energy at the hop site $E_{n,hop}$ are respectively calculated by the following equations:

$$E_N = E_{n,initial} - gE_{n,hop}, \tag{2}$$

$$E_{n,initial} = mE_b + \delta nE_b, \tag{3}$$

$$E_{n,hop} = m'E_b + \delta n'E_b, \tag{4}$$

where m (m') and n (n') are the numbers of the nearest and the next nearest atoms, respectively. A coupling constant $g = 0.3$ is used in the equation, which describes a weak coupling between the two adjacent lattice sites. We note that, as the interaction between the substrate and depositing atoms will decrease as the atom height increases, the substrate atom binding energy to the surface E_S is an exponential decay with the increasing distance

$$E_S = mE_{S0}\exp(h_{atom}), \tag{5}$$

where E_{S0} is set as 1.35 eV and h_{atom} is the height of the formed island. The material parameters used in our simulations are those of the three species (Si, Ge, Sn). Thus, we take the Ge/Si (001) system with embedded Sn inclusions as an example to investigate the Ge QR self-assembly. The lattice constants of Si, Sn and Ge are chosen to be 5.43 Å, 6.489 Å and 5.658 Å, respectively. Finally, to account for the up and down motion of the atoms, an additional factor μ is introduced,[31] which is defined as the ratio of the probability for edge atoms to hop up to hop down. The factor μ should be larger than 1. Otherwise, all the atoms will tend to move to lower layers.

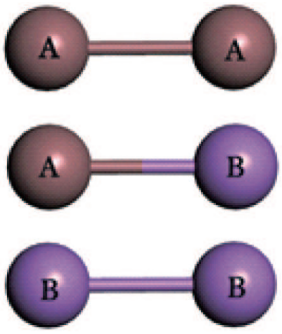


Fig. 1. (color online) Schematic diagram of three kinds of bonding modes between two atoms.

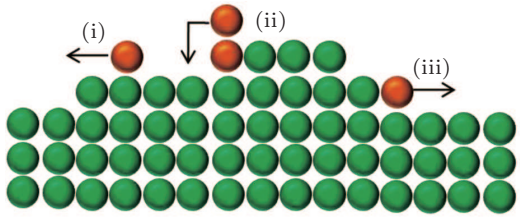


Fig. 2. (color online) Illustration of the diffusion processes considered in the simulation. (i) Adatoms move on the flat terrace; (ii) step down at a step edge; (iii) detach from a step edge.

3. Results and discussion

Using the 3D QD self-assembly approach described above, we can now simulate the epitaxial growth process. The growth model is on a 200×200 grid with the periodic boundary condition and the model material is InAs/GaAs (001). While there are two different species In and As in a compound InAs QD, we do not distinguish them in our KMC diffusion simulation;[32,33] the chemical species information is implied in the mismatch strain and elastic stiffnesses used in the strain energy calculation. The effects of growth parameters (temperature, flux rate, total coverage and interruption time) on the quality of QDs growing are studied as follows.

3.1. The temperature influencing on the growth of QD islands

The island size equalization relies on the movement of atoms on the surface during the epitaxial growth. It could start at the very beginning of the deposition process if a low flux rate is given.[32,33] Figure 3 shows the effect of different temperatures on island size distributions. It is clearly observed from Fig. 3 that for a lower temperature, there is no uniform shape and there are a lot of thin film shape distributions on the substrate. The phenomenon can be explained as follows. When the atoms deposit on the surface of the GsAs substrate, due to a low temperature, there is not enough thermal activation energy to overcome the diffusion barrier, then the atoms only diffuse in a narrow range or even in the home position; the atoms do not overcome the band energies and the substrate bondage to jump to the upper layers. For a high temperature, the atomic thermal activation energy increases to overcome some diffusion barriers and some atoms can overcome the substrate bondage, which leads to the formation of small islands. Meanwhile, the atoms begin to diffuse to the upper layers after overcoming the substrate bondage and the islands become higher. As the temperature increases, small isolated islands tend to join onto large ones in their proximity and the island distribution becomes more and more ordered. For example, when T = 800–900 K, the islands have a fixed shape and the distributions are more uniform than that when T is lower than 800 K.

Figure 4 demonstrates further the relation between the total coverage and the number of islands at different temperatures. For a low temperature, there are more single islands with a smaller total coverage. At the beginning of the deposition process, the number of islands is larger. With longer growth time, the single atoms tend to join into isolated islands in their proximity, which then join onto larger ones and the number of islands decreases. Figure 4 illustrates that the number of islands is sensitive to the total coverage when it is less than 0.5 ML. For a small total coverage ($C < 0.5$ ML), as the temperature increases, the atomic thermal activation energy increases and the probability of atoms joining into islands becomes larger. So the number of islands decreases and the atomic average size becomes bigger.

With the total coverage increasing, the temperature of the substrate rises, which results in more deposited atoms diffusing to the upper layers, higher atomic islands and a smaller coverage of atoms. However, when the substrate temperature is lower, it is hard for the atoms to diffuse to the upper layers and the coverage of atomic islands becomes bigger, which leads to islands growing quickly along the horizontal direction. It is concluded that with more total coverage and a higher temperature, the height of atomic islands tends to be higher so that the number of islands decreases more slowly.

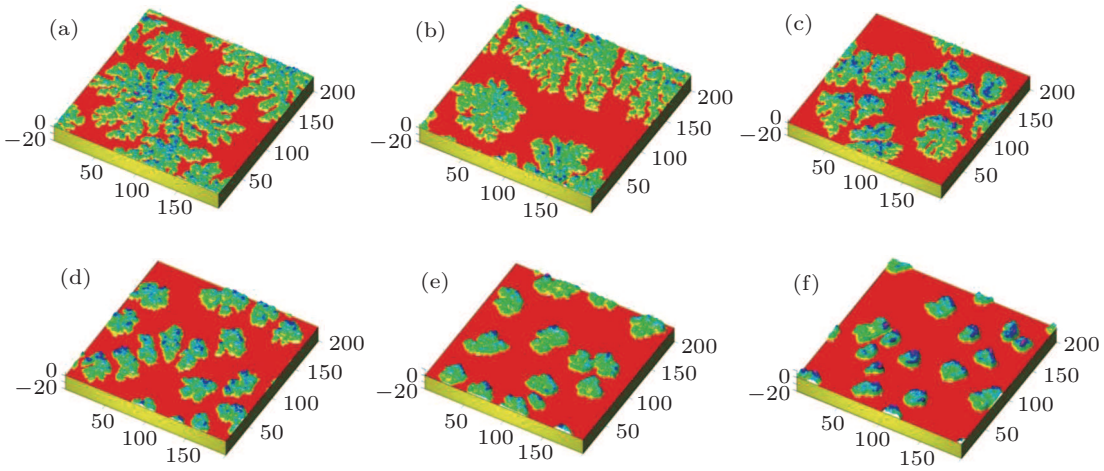


Fig. 3. (color online) The 3D island distributions for different temperatures ((a) 400 K, (b) 500 K, (c) 600 K, (d) 700 K, (e) 800 K, (f) 900 K) with up-down ratio ρ=20. Fix growth parameters are flux rate $F = 1$ ML/s, total coverage $C = 1$ ML and $E_b = 0.2$ eV.

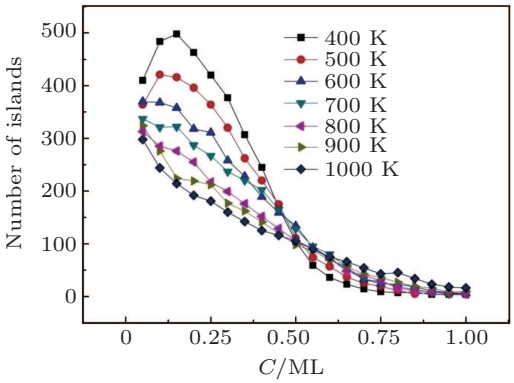


Fig. 4. (color online) The relationship between the atomic total coverage and the number of islands at different temperatures.

3.2. The flux rate influencing on the growth of QD islands

The flux rate represents the rate of the material depositing on the substrate surface that does not affect any growth process of self-assemble QDs, e.g., strain energy and diffusion barrier. However, the flux rate influences the atomic density and the nucleation rate of islands per unit of time, so it is still a key factor affecting the growth of semiconductor QDs.

Figure 5 shows the effect of the flux rate on the distributions of islands. For a low flux rate, e.g., $F = 0.01$ ML/s, the deposited atoms have more time to move to the equilibrium position and to assemble together during the deposition process. On the contrary, for a high flux rate, there is not enough time for the deposited atoms to diffuse and then there is a cluster of islands. Therefore, for the same growth parameters, with lower flux rates, the size equalization of islands will be better; it is the opposite case with high flux rates. Therefore, besides the up-down ratio, the flux rate is also important for obtaining an ordered and uniform-size QD island distribution. This is particularly true as a low flux rate is required for size equalization starting from the beginning of the deposition process.

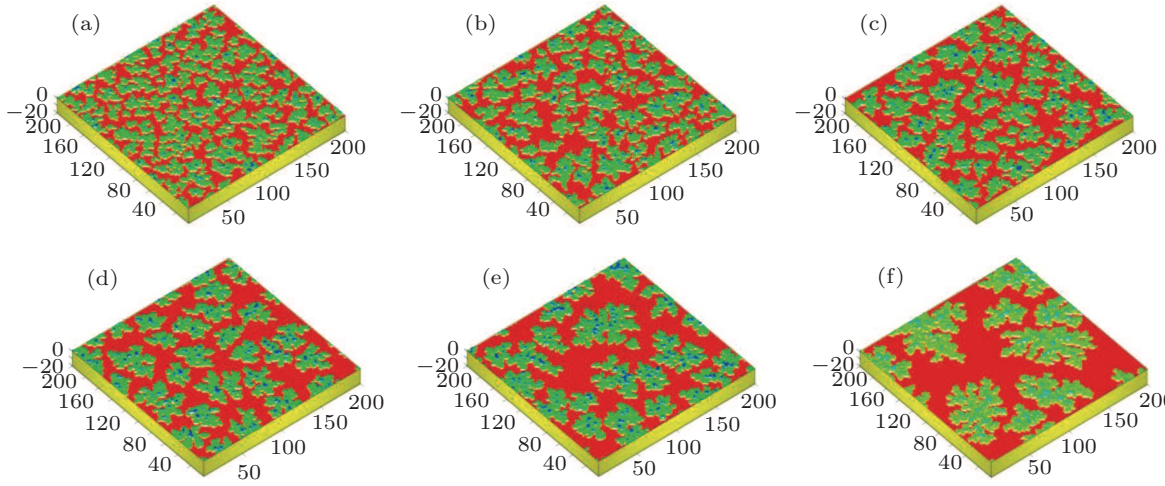


Fig. 5. (color online) The 3D island distributions for different flux rates ((a) 2 ML, (b) 1 ML, (c) 0.5 ML, (d) 0.1 ML, (e) 0.05 ML, (f) 0.01 ML) with up-down ratio $\rho = 20$. Fix growth parameters are temperature $T = 800$ K, total coverage $C = 1$ ML and $E_b = 0.8$ eV.

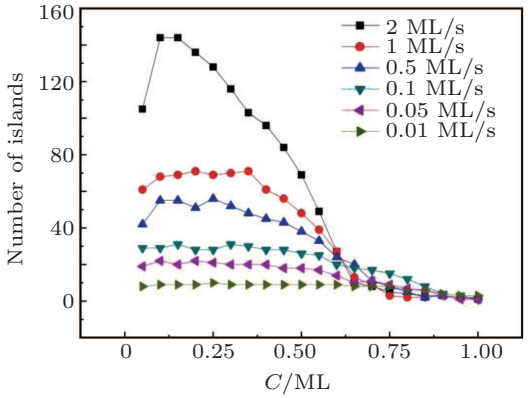


Fig. 6. (color online) The relationship between the number of islands and the total coverage at different flux rates.

Figure 6 shows the relationship between the total coverage and the corresponding number of islands for different flux rates, which again displays clearly the island size and number variation during growth. It clearly demonstrates the following. (i) With a high flux rate ($F = 2$ ML/s, 1 ML/s, 0.5 ML/s), the number of islands increases firstly and then decreases gradually during the deposition process. The reason is that for a high flux rate, there is not enough time for the deposited atoms to diffuse and the range of diffusion is small. At the beginning of the growth process, there are many isolated islands and the number of islands is directly proportional to the total coverage. With a longer growth time, the isolated islands tend to join onto the large ones in their proximity so the number of islands decreases. (ii) For a low flux rate ($F < 0.5$ ML/s), there is a longer diffusion time for the deposited atoms, so the number of islands is small. With more deposited atoms, the isolated islands join onto larger ones, so the number of islands decreases as the deposited time becomes longer. (iii) Figure 6 displays clearly that the number of islands is sensitive to the total coverage with high flux rates ($F = 2$ ML/s, 1 ML/s, 0.05 ML/s) while that changes very slowly for low flux rates ($F < 0.5$ ML/s).

3.3. The interruption time influencing on the growth of QD islands

Finally, the effect of the growth interruption on QDs growing is studied. The growth interruption, a stage after atoms deposition before capper material depositing, is another important factor influencing the growth quality. It is observed from experiments that it plays a very important role in the surface appearance of crystal.[1] During the growth interruption, the deposited atoms do not stop diffusing. On the contrary, it is easier for the deposited atoms to move to the equalization positions and for the system to be in the thermal equilibrium state. It is essentially different from growth by dynamics and that by thermo kinetics, so the growth interruption is very important for QDs' appearance.

As shown in Fig. 7, the shape of islands is irregular and the distribution of islands is asymmetrical just at the end of growth. However, with a longer interruption time, the edge of islands becomes smoother and the islands become more isolated. At $t = 40$–50 s, the shape of the islands is basically stable, the islands are more ordered and the system approaches the thermal equilibrium state gradually.

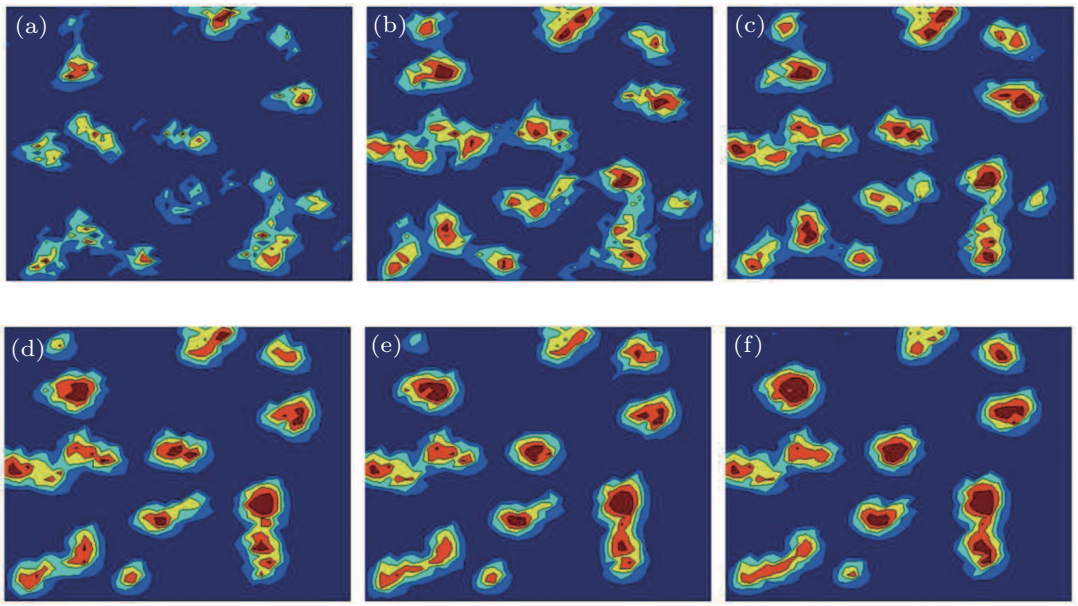


Fig. 7. (color online) The influence of different interruption times ((a) 0 s, (b) 10 s, (c) 20 s, (d) 30 s, (e) 40 s, (f) 50 s ML) on 3D island shape and distributions with up-down ratio ρ=20. The fixed growth parameters are temperature T=600 K, flux rate F=0.1 ML/S and total coverage C=0.15 ML.

4. Conclusion

In this paper, we propose a 3D QD epitaxial growth model by enhancing our former x, y-plane growth model. The effects of temperature, flux rate, total coverage and interruption time on the island equalization and distribution are studied; furthermore, combining the flux rate and the total coverage, the temperature and the total coverage, the number of islands that change is illustrated in the simulations during and after the deposition is demonstrated. The main conclusions from our 3D KMC model are as follows.

(i) The island size equalization could start at the very beginning of the deposition process if the flux rate is low. For other fixed growth parameters, as the temperature increases, the islands become more and more ordered and the island shape becomes more homogeneous. However, the number of islands decreases with the temperature decreasing when the total coverage $C < 0.5$ ML.

(ii) For other fixed growth parameters, with a low flux rate, there is enough time for the deposited atoms to diffuse and for the islands to join onto ordered and homogeneous islands. So the shape of the islands is more ordered and the size of the islands is bigger as the flux rate decreases. For a low flux rate, when the total coverage $C < 0.5$ ML, the number of islands decreases with a longer growth time.

(iii) When the atoms stop growing, the shape of the islands is irregular and the distribution is nonuniform. During the growth interruption, the deposited atoms keep diffusing. With the interruption time increasing, an ordered island distribution with uniform size can be obtained.

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