SYSTEMATIC STUDY OF INFLUENCE OF GROWTH PARAMETERS ON ISLAND MORPHOLOGY DURING MOLECULAR BEAM EPITAXY GROWTH: A MONTE CARLO STUDY

Shankar Prasad Shrestha
Department of Physics, Tribhuvan University, Patan Campus, Patan Dhoka, Nepal
and
The Abdus Salam International Centre for Theoretical Physics, Trieste, Italy
and
Chong-Yun Park
Department of Physics Sung Kyun Kwan University, Suwon 440-746, South Korea.

Abstract

We have made a systematic study of influence of diffusion flux ratio (D/F), diffusional anisotropy (DA) and sticking anisotropy (SA) on island morphology to show the influence of each growth parameter on island morphology in presence of the other growth parameters. Our results show that the influence of D/F ratio and DA on island morphology depends on the sticking anisotropy of the adatoms. At the intermediate anisotropic case, increase in D/F ratio results in transition of the island morphology from 1d nature to 2d nature. In anisotropic diffusion case, D/F ratio can change the growth direction of the island morphology. We also find that only sticking anisotropy is not sufficient to produce elongated islands, low D/F ratio is also essential.

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1Regular Associate of ICTP. Corresponding author. shankarpds@yahoo.com, shankarpds@hotmail.com
Introduction

In the recent years, with the development of the analytical tool such as scanning tunneling microscope the study of submonolayer epitaxial growth of metal and semiconductor has received considerable attention [1-7]. The experimental investigation of the island morphologies formed during submonolayer deposition on various system shows the formation of various types of island morphologies ranging from 1d chain like [8-10], compact 2d [11-15], to fractal like [16-17] structure. Understanding the factors governing the shape of clusters in diffusion controlled aggregation is very essential in the MBE film growth due to much interest of nanostructure from fundamental as well as technical importance. Several groups have studied the evolution of the growth of islands morphologies as well as the scaling of islands during submonolayer epitaxy using Monte Carlo simulation [18-24]. Using various models, effect of anisotropy of diffusion [15, 25-29] and binding strength [26-27, 30] between adatoms and adatom and island on island shape were studied as well. For example, in the simulation study by Ferrando et al.[26] and Mottet et al.[27] the growth of island morphologies were studied using a model with deposition, diffusion and fully reversible aggregation on a rectangular substrate and also were applied to the case of low coverage homoepitaxial growth of Ag and Cu(110). They have shown that depending on flux and temperature, the island morphology varies from small isotropic cluster to 1d strips. The formation of strips was attributed to sticking anisotropy. Similarly, in Mo et al.’s [2] investigation on submonolayer growth of Si on Si(100), the observed shape anisotropy were also attributed to the anisotropy in sticking probability and not due to diffusional anisotropy. Using different model, the elongation of islands were also studied [29]. The anisotropic corner rounding processes were identified to be responsible for the island elongation. The elongated islands were obtained in isotropic diffusion case also when the attachments at both island sides are different. The influence of diffusional anisotropy and binding strength between adatoms and the adatom and island border on the island aspect ratio were studied using temperature, surface diffusion barrier and adatom binding energy as parameters [30]. Using solid on solid model of the epitaxial films growth based on random deposition the influence of the critical number of adatoms lateral bonds and the deposition rate on possible surface morphology anisotropy were also studied [31]. In spite of great deal of work, systematic study of influence of each parameter in presence of the other parameter on island morphology is still lacking. In this paper, we report how each parameter (diffusion to flux ratio (D/F), diffusional anisotropy (DA) and sticking anisotropy (SA)) show its influence on island morphology in different combination of the other two parameters.
Model and simulation

Our simulation and model used closely resembles the Monte Carlo solid-on-solid simulation model used by Labella [13]. Using this model, they have successfully generated the similar island morphologies obtained by STM at different temperature. By combined study of simulated morphologies and temperature dependent STM morphologies, they have successfully determined the diffusion parameter for Ga on GaAs(001)-2x4 surfaces. In our simulation, the monomer encounters the following process before the final island morphology is formed.

Deposition and diffusion: Monomers are deposited randomly at constant flux (F) of 1/600 ML per sec, on a square lattice of size 600x600. The deposited monomers are allowed to diffuse to the nearest neighbour (NN) sites by random jump. Each jumping adatom is selected at random among the existing monomers, which are not in contact to the other adatom or island in the substrate. Similarly, the NN site for jump of the adatom is also selected randomly. The jump at the selected site is allowed only if it is not occupied by any other atom. In order to take account of diffusional anisotropy, any attempt to jump to the NN site in x direction or y direction is made successful according to the preset value of the diffusional anisotropy parameter (DA). The parameter DA is a dimensionless parameter which is simply a diffusional probability ratio DX/DY, where DX (DY) is the diffusion probability of monomer along X and Y direction in unit of jump per second. When the depositing monomer encounters the top of existing monomer or island, it is allowed to jump in one of the NN site chosen at random. If the NN site is also the top of an island, then for simplicity another site of deposition is chosen. For low coverage, this will not affect the resulting morphology because probability of such event is quite low. The deposition is stopped when final coverage of 0.07 is reached. In order to avoid the edge effect, periodic boundary condition were applied in both X and Y direction.

Nucleation and aggregation: When the deposited monomer or diffusing monomer encounters the another monomer in nearest neighbour site, a stable two adatoms island is allowed to nucleate with certain probability depending on the initial preset sticking anisotropy parameter (SA). As in the diffusional anisotropy case, the parameter SA is simply a sticking probability ratio (SY/SX), where SY (SX) is the sticking probability along Y and X direction of the substrate. Thus, in isotropic sticking case, the bonding between adatoms along normal and parallel to [001] direction are equally probable, where as in anisotropic sticking case, bonding along the normal direction is highly improbable as compared to the bonding in parallel direction. Since we do not allow island dissociation, our simulation is basically for low temperature MBE island growth. When the deposited monomer or diffusing monomer encounters an
existing island, it is allowed to stick to the island. When it sticks to its nearest neighbour site, the number of atoms in the island is increased by one. If the adatom does not stick to the NN site then it is treated as a monomer and allowed for further diffusion. In order to study the influence of each parameter DA, SA and D/F on the island morphology, we have monitored the island morphology as well as island anisotropy parameter $\sigma$ for various values D/F ratios 600 to $6\times10^5$. These all simulation steps was repeated for the different fixed values of (DA) parameter and these again are repeated for the different values of SA.

**Results**

Our simulation of deposition of submonolayer amount (0.07ML) with the simple model produced various types of island morphologies depending on directional hopping ratio, directional sticking probability ratio and D/F ratio. Influence of each term on the island morphology is discussed below. Influences of DA and D/F ratio on the island morphology were studied by performing each simulation at different fixed values of SA.

Fig. 1(a-k) depicts the morphologies of the clusters at various D/F ratio and diffusional anisotropy’s for isotropic sticking case. The horizontal panel from left to right depicts the variation in island morphologies with respect to D/F ratio for three different diffusional anisotropy parameter (DA = 1(upper), $10^3$(middle), $\propto$(lower panel)). Similarly, the vertical panel from top to bottom depicts the variation of island morphologies with respect to DA for three different D/F ratios (J=10$^1$(left), $10^3$(middle), $10^5$(right panel)). At low D/F ratio (Fig. 1(a)), the island is small with many isolated atoms. At high D/F ratio, the islands show fractal like structure. Also as expected, as D/F ratio increases the number density of island decreases whereas island size increases. This behavior is found to be true for all studies at three different fixed DA values. Similar growth patterns of the islands are observed in other studies at the other different DA values (Fig. 1(d-k)). A closer look on the islands for anisotropic diffusion case (Fig. 1(i, j and k)) reveals that when D/F is low a large number of islands are elongated along direction of fast diffusion (Y-direction) where as when D/F ratio is high the islands seem to be more elongated along the direction perpendicular to the direction of fast diffusion (X-direction). Comparing Fig. 1 (a, b and c) with Fig. 1(i, j and k) we find that when diffusion is isotropic, the influence of D/F is observed to only increase the island size and it does not lead to any shape anisotropy. But, when diffusion is anisotropic, D/F ratio results slight shape anisotropy.

Fig. 1(a, d, i), 1(b, e, j), 1(c, f, k)(along the vertical panel) depicts the variation of the island morphologies with respect to DA for 3 different D/F ratios (DA = 1, $10^3$ and $10^5$). It is seen from the figure that at low D/F ratio the diffusional anisotropy does not
show any marked variation in island size, where as at high D/F ratio (Fig. 1c, f, k) the island size for isotropic diffusion is observed to be larger than the island size for low DA. As reported earlier [2-3], the number density of island is also found to increase with diffusional anisotropy. The reason for this effect is explained in the discussion section. As mentioned above, the number density of island increases with diffusional anisotropy. When D/F is low (D/F=1), the island size is small and the change of DA from DA = 1 to DA = \( \propto \) leads to slight change in the island elongation along the direction of fast diffusion. But when D/F ratio is high (D/F = 10^5), the island size is big and islands are slightly elongated along the direction perpendicular to the fast diffusion direction [3, 30]. This implies that at low D/F ratio the growth is along the direction of fast diffusion where as at high D/F ratio it is along the direction perpendicular to it. This result contradicts the results of Mo. et al. [2] in which slightly different model was used. In their study no shape change were observed.

Fig. 2(a-k) depicts the morphologies of the clusters at various D/F ratio and DA values for anisotropic sticking case. At low D/F ratio, small preferential direction in their orientation of the island is observed. For intermediate D/F ratio, the islands are one dimensionally elongated. At high value of D/F ratio, more elongated 1d islands are observed and only few islands is found to be slightly 2d nature. Similar influence of D/F ratio is observed in all DA cases. Its influence for island elongation is much more effective in isotropic diffusion case as compared to anisotropic diffusion case. Similar results were observed by Ferrendo et al.[26] with fully reversible aggregation model on rectangular lattice using kinetic Monte Carlo simulation. Using temperature as a parameter and for the case of isotropic surface and anisotropic bonding energies, similar elongated islands were also reported [30].

Fig. 2(a, d, i), (b, e, j) and (c, f, k) depict the variation of the island morphologies with respect to the diffusional anisotropy for three different D/F ratio for anisotropic sticking case. From the figure it is seen that the island shapes (Fig. 2(b, c)) is much more elongated then the island shape for DA=0 (Fig. 2(j, k)). This result shows that the diffusional anisotropy does not lead island elongation but instead it shortens the island length. Thus, higher the diffusional anisotropy lesser will be the island elongation. The same effect is observed for all D/F ratio studied (see Fig. 2 vertical panels).

In order to study the effect of directional sticking probability on the island morphology, we have fixed the DA value and variation in morphology with D/F ratio were monitored for different SA values. The same procedure was repeated for different fixed DA values. Here we present the results for DA = 1 (isotropic diffusion case) and DA = \( \propto \) (perfect anisotropic diffusion case).

The horizontal panel in Fig. 1(a, b, c), Fig. 3(a, b, c) and Fig. 2(a, b, c) depict
the variation in island morphology with respect to D/F ratio for the three different fixed SA values (SA = 1, 10^{-3}, 0) for isotropic diffusion case. Comparing these figures, we find that when SA = 1, the variation of D/F ratio will not lead to any shape anisotropy. But, when SA value is decreased to SA = 10^{-3}, the variation of D/F is observed to change the shape of the island from 1d like nature at low D/F ratio to 2d like nature at high D/F ratio (see Fig. 3(a, b, c)). When SA value is further decreased to zero, the islands are observed to be almost 1d type for all D/F ratios. (See Fig. 2 (a),(b) and (c)). The horizontal panel in Fig. 1(i, j, k), Fig. 2(i, j, k), Fig. 4(i, j, k) depict the variation of morphology with D/F ratio at 3 different fixed values of SA (SA=1, 0 and 10^{-3}) for DA = \infty case. As discussed earlier, when SA = 1 or SA = 0 the variation of D/F ratio results only slight change in the island morphology. But, when SA = 10^{-3}, the variation of D/F leads to the morphology transition from 1d nature at low D/F value to 2d nature at high D/F value. At high D/F ratio, the islands are observed to be more compact and elongated structure (Fig. 4c). Thus, the island morphology transition with D/F ratio takes place at intermediate sticking condition.

Fig. 1(a), Fig. 2(a), Fig. 3(a), Fig. 1(b), Fig. 2(b), Fig. 3(b) and Fig. 1(c), Fig. 2(c), Fig.3(c) depict the variation of the island morphology with SA for three different D/F ratios. When SA=1 and D/F ratio is high, the ramified island morphology is observed (Fig. 1c). When SA value is decreased further to 10^{-3} and 0, the island morphology is observed to get more elongated and finally show almost 1d natured. Similar morphology transitions are observed for anisotropic diffusion case also. As seen by comparing Fig. 1(i), Fig. 2(i), Fig. 4(i), Fig. 1(j), Fig. 2(j), Fig. 4(j) and Fig. 1(k), Fig. 2(k), Fig. 4(k). Thus, when diffusion is isotropic or anisotropic the island shape change drastically with the change in SA value. This shows that SA plays major role in the island morphology change as reported by earlier workers [2, 26, 29].

In order to characterize the degree of anisotropy and the compactness of the islands, we have calculated the anisotropy parameter \( \sigma \) [26], using the relation given below.

\[
\sigma = \frac{(<np>-<nn>)}{(<np>+<nn>)}
\]

where \(<np>\) and \(<nn>\) are the average number of parallel and normal nearest neighbor bonds for a given set of D/F, DA and SA value. When all islands are 1 dimensional, the value of \( \sigma \) becomes 1. As the island shows 2d character, the value of \( \sigma \) decreases and it becomes to zero when average number of parallel and normal bonds are equal.

Fig. 5 depicts the variation of the island anisotropy parameter \( \sigma \) with D/F for different values of DA for isotropic sticking case (SA=1) and anisotropic sticking (SA=0) case. We see that when sticking between the adatoms is isotropic the value of the anisotropy parameter \( \sigma \) is always low (<0.1) for all D/F ratio and DA values. When
diffusion is isotropic (DA=1) the $\sigma$ value remain always 0 for all D/F ratios. For anisotropic diffusion case (DA>1), we see that the value of $\sigma$ increases slowly from a negative value at low D/F region to a positive value at high D/F region. This variation is more clearly shown in the inset to Fig. 5 in which the vertical axis is expanded. The variation of $\sigma$ with D/F for all diffusional anisotropy values (DA=1 to $\infty$) is found to follow similar trend.

It is interesting to note that (Fig. 5 inset) as D/F ratio increases the anisotropy parameter increases and beyond certain value of D/F depending on diffusional anisotropy, the anisotropy parameter $\sigma$ changes its sign. This behaviour is observed for all DA ratios except for DA=1 case where it is always zero. This implies that when the value of D/F ratio is low the $\sigma$ value is negative and the islands are slightly more elongated along the direction of fast diffusion direction. At high D/F ratios the $\sigma$ becomes positive and the islands get more elongated along the direction perpendicular to the direction of fast diffusion. Further, we have also observed that when D/F ratio is low, $\sigma$ value decreases to negative value with increase in DA, and at high DA values, it remains approximately constant. In high D/F ratio case, increase in DA values results in increase of $\sigma$ in positive direction. This is more clearly shown in Fig. 6. This implies that in isotropic sticking and anisotropic diffusion case, when D/F ratio is low, the island is more elongated along the Y-direction implying that island growth take place along the direction of fast diffusion. At sufficiently high D/F ratio $\sigma$ becomes positive, which implies that island growth take place along the direction perpendicular to the fast diffusion direction, which is observed in island morphology study also.

From figure 5, we also see that when sticking is anisotropic (SA=0), the value of $\sigma$ is always high (>0.8) for all D/F ratios except at low value D/F ratio. This shows that the resulting island morphology always show approximately 1d nature. Thus, when bond strength of adatoms along one direction is very high as compared to bond strength along the other, the resulting growth structure is always approximately 1d natured irrespective of D/F ratio and diffusional anisotropy of the surface. However, when D/F ratio is low the $\sigma$ value is quite low. Comparing the $\sigma$ versus D/F curve for isotropic and anisotropic sticking case we find that in isotropic sticking case (SA=1), the value of $\sigma$ is always low (<0.1) for all D/F ratios and DA values and for anisotropic sticking case (SA=0), the value of $\sigma$ always high. This shows that sticking anisotropy drastically influence the island morphology. When SA=0 the islands always show 1d natured provided D/F ratio is high. This is clearly visible in island morphology, which is discussed earlier.

Fig. 7 depicts the variation of $\sigma$ with D/F ratio at different values of stick probability ratios (SA=1 - 0) for isotropic diffusion case (DA=1). The curve for SA=0 and SA=1 is repeated here for comparison purpose. It is seen in the figure that when SA
value is either very high (SA=1) or very low (SA=0), the $\sigma$ value always tends to remain constant with respect to variation of D/F ratio. But for the intermediate value of SA, the $\sigma$ value changes drastically with D/F ratio. This shows that, with respect to D/F ratio the morphology transition from 2d to 1d occurs only if bonding between adatoms neither highly isotropic nor highly anisotropic. From Fig. 7, we also see that, higher the sticking anisotropy (lesser the SA value), the more steep variation of $\sigma$ with respect to D/F ratio is observed.

From Fig. 7 it is also clearly seen that sticking probability ratio drastically influences the anisotropy parameter $\sigma$ (along the vertical line for fixed D/F ratio). At low D/F values sticking ratio (SA>=10^{-2}) is enough to produce almost 1d islands ($\sigma$~1) but as D/F ratio increases, the $\sigma$ value decreases and the island starts to show 2d nature. Therefore, only sticking anisotropy is not sufficient to produce elongated island but low D/F ratio is also essential. For all D/F ratios the island morphology is greatly affected by sticking probability ratio.

Fig. 8 depicts the variation of $\sigma$ with D/F at different SA values for perfect anisotropic diffusion case (DA = $\propto$). The variation of $\sigma$ with respect D/F ratio is very much similar to the isotropic diffusion case, but in this case the $\sigma$ value starts decreasing at lower value of D/F ratio as compared to the anisotropic case. Therefore, as in the case of isotropic diffusion, the influence of D/F on $\sigma$ depends on sticking anisotropy parameter. In anisotropic diffusion case also, the sticking probability drastically change the morphology of the island which is true for all D/F ratio (Fig. 8)

**Discussion**

The reason for the effect observed for diffusional anisotropy can be qualitatively explained as follows. When the diffusion is highly anisotropic, the adatoms jumps only along the line. Therefore, there are only two probable sites for jump (see Fig. 9a). Among these two sites, only one site is new with respect to previous jump, because before jumping to the present site, the adatom was in one of the present available site. Thus, for each jump there is effectively only one possibility of finding new site, which is not visited in earlier jump. For the isotropic diffusion case, the possibility of finding new site is three (see Fig. 9b). Which means that for isotropic case, for the same D/F ratio the probability nucleation or growth event is three times larger than for anisotropic case. For high D/F ratio, most of monomers are already attached to the island and the island growth dominates the island nucleation, which leads to larger island and smaller number density. In anisotropic diffusion case, this probability of finding new site is low as compared to the isotropic diffusion case. Due to this, the adatoms tends to remain isolated and this leads to higher nucleation probability then growth. Thus, the number
density for anisotropic case will be higher and island size will be smaller which is observed in the simulation.

**Summary and conclusion**
We have presented Monte Carlo results for a systematic study of the influence of diffusional anisotropy, sticking anisotropy and D/F ratio on island morphology during submonolayer deposition using irreversible nucleation and growth model. The main findings are summarized below.

(a) The influence of D/F ratio and diffusional anisotropy on island morphology is observed to depend on the sticking probability ratio. In the isotropic sticking case, when diffusion is isotropic, D/F ratio is observed to increase the island size only and it does not lead to any shape anisotropy whereas, when diffusion is anisotropic D/F ratio is observed to change the direction of island elongation direction slightly. Also in the isotropic sticking case, when D/F ratio is low, increase in DA leads to island elongation along the direction of fast diffusion whereas when D/F ratio is high, it leads to elongation along the perpendicular direction.

(b) In the anisotropic sticking case, when diffusion is isotropic, increase in D/F leads to highly elongated islands whereas when diffusion is anisotropic, only slight elongation of islands are observed. In this case, for all values of D/F ratios, increase in DA is observed to cause decrease in island elongation. This shows that DA does not lead to island elongation but instead is shortens.

(c) In isotropic as well as anisotropic diffusion case, when sticking is isotropic, change of D/F ratio does not lead to shape anisotropy. When sticking is isotropic islands always show 2d nature and when it is highly anisotropic the islands show 1d nature. At intermediate sticking ratio, the D/F ratio results in transition of island morphology from 1d nature at low D/F ratio to 2d nature at high D/F ratio.

(d) When diffusion is either isotropic or anisotropic the island shape is observed to change drastically with SA. The sticking probability ratio is observed to show its influence in all cases of diffusional anisotropy and D/F ratios. Further, only sticking anisotropy is not observed to be sufficient to produce the elongated island. Because, if D/F ratio is also high it results 2d natured island. Therefore, low D/F ratio is also essential.

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References

**Figure Captions**

Fig. 1. Section (100*100) of typical island morphologies obtained from the simulation for isotropic sticking case \((SA=1)\). The horizontal panel are morphologies at \(D/F = 1\) (upper), \(10^3\) (middle) and \(10^5\) (lower) and the vertical panel are morphologies at \(DA = 1\) (left), \(10^3\) (middle) and \(\propto\) (right), respectively.

Fig. 2. Section (100*100) of typical island morphologies obtained from the simulation for anisotropic sticking case \((SA=0)\). The horizontal panel are morphologies at \(D/F = 1\) (upper), \(10^3\) (middle) and \(10^5\) (lower) and the vertical panel are morphologies at \(DA = 1\) (left), \(10^3\) (middle) and \(\propto\) (right), respectively.

Fig. 3. Section (100*100) of typical island morphologies obtained from the simulation for isotropic diffusion case \((DA = 1)\). The horizontal panel are morphologies at \(D/F = 1\) (left) \(10^3\) (middle) and \(10^5\) (right) for \(SA = 10^{-3}\).

Fig. 4. Section (100*100) of typical island morphologies obtained from the simulation for anisotropic diffusion case \((DA = \propto)\). The horizontal panel are morphologies at \(D/F = 1\) (left) \(10^3\) (middle) and \(10^5\) (right) for \(SA = 10^{-3}\).

Fig. 5. Variation of \(\sigma\) with \(D/F\) ratio for 7 different \(DA\) values for isotropic sticking case \((SA=1, \text{ lower})\) curve and anisotropic sticking case \((SA=0 \text{ upper})\) curve. Inset to the figure shows the same curve for \(SA = 1\) with vertical scale expanded.

Fig. 6. Variation of \(\sigma\) with \(DA\) for 7 different \(D/F\) ratio for isotropic sticking case \((SA=1, \text{ lower})\) curve and anisotropic sticking case \((SA=0 \text{ upper})\) curve.

Fig. 7. Variation of \(\sigma\) with \(D/F\) ratio for 7 different sticking parameter \(SA\) for isotropic diffusion \((DA = 1)\), lower)

Fig. 8. Variation of \(\sigma\) with \(D/F\) ratio for 7 different sticking parameter \(SA\) for anisotropic diffusion \((DA = \propto)\)

Fig. 9. Schematic top view of the adatoms (filled circle) showing possible jump sites (a) two jump sites (1, 2) for anisotropic diffusion case and (b) four jump sites (1, 2, 3 and 3) for isotropic diffusion case. Gray filled circle represent the position of the adatoms before jumping to the current site.
Fig. 5

Fig. 6
Fig. 7

Fig. 8

Fig. 9