จุฬาลงกรณ์มหาวิทยาลัย

ทุนวิจัย

กองทุนรัชดาภิเษกสมโภช

รายงานวิจัย

การจำลองสร้างฟิล์มบางบนแผ่นรองรับที่มีลวดลาย

โดย

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Simulations of thin film growth on patterned substrates

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<u>บทคัดย่อ</u>

การสร้างฟิล์มบางบนแผ่นรองรับที่มีลวดลายเป็นเทคนิคที่ชั้นของฟิล์มบางถูกสร้างบนแผ่นรองรับที่มีการกำหนดลวดลายไว้ก่อนแล้ว แบบจำลองอย่างง่ายได้ถูกใช้เพื่อศึกษากระบวนการสร้างลวดลายที่มีลวดลายแตกต่างกันสองแบบ ได้แก่ ลวดลายที่เรียบและลวดลายที่เป็นคาบ โดยมีจุดมุ่งหมายเพื่อกำหนดเงื่อนไขในการสร้างที่ทำให้ฟิล์มที่สร้างขึ้นสามารถถอดแบบลวดลายดั้งเดิมได้และเพื่อกำหนดว่าลวดลายดั้งเดิมมากเพียงใดที่จะคงอยู่ได้ไปจนถึงเวลาที่สนใจ ความน่าจะเป็นของการคงอยู่ได้ถูกใช้เพื่อกำหนดสัดส่วนของลวดลายที่ยังคงอยู่ เราพบว่าในการสร้างลวดลายที่เรียบอุณหภูมิของแผ่นรองรับที่มีค่าสูงจะส่งผลให้มีการเพิ่มระยะการแพร่บนพื้นผิวของอะตอมที่กำลังเคลื่อนที่และสามารถช่วยเพิ่มค่าสภาพการคงอยู่ของลวดลายได้ ถ้าอุณหภูมิของแผ่นรองรับสูงพอฟิล์มจะถูกสร้างในรูปแบบชั้นต่อชั้นและลวดลายที่เรียบจะคงอยู่ได้เป็นเวลานาน ส่วนในการสร้างลวดลายที่เป็นคาบการเพิ่มระยะการแพร่บนพื้นผิวช่วยให้เกิดความราบในส่วนที่เรียบของลวดลายแต่ทำลายรูปร่างเค้าโครงของลวดลาย เราพบว่าอุณหภูมิของแผ่นรองรับจะต้องเป็นค่าที่ไม่น้อยจนเกินไปหรือมากจนเกินไป ค่าที่เหมาะสมที่สุดจะขึ้นอยู่กับขนาดของลวดลายโดยลวดลายที่มีขนาดของลักษณะเด่นใหญ่กว่าจะสามารถคงอยู่ได้เป็นระยะเวลาที่นานกว่า สุดท้ายเราได้ปรับปรุงนิยามของความน่าจะเป็นของการคงอยู่เพื่อที่จะให้ได้ค่าความน่าจะเป็นที่สอดคล้องยิ่งขึ้นกับลักษณะพื้นผิวที่จำลองขึ้น

Project Title Simulations of thin film growth on patterned substrates

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Abstract

Patterned substrate growth is a technique that layers of thin film are grown on a substrate with a predetermined pattern. A simple model is used to study patterned growth process with two different types of pattern: flat pattern and periodic pattern. The goals are to determine growth conditions that enable the grown film to reproduce the original pattern, and to determine how much of the original patterns survive up to a specific time. The persistence probability is used to determine fractions of survived pattern. We found that in flat patterned growth, a high substrate temperature which results in a long surface diffusion length of moving atoms can help increase the persistence probability of the pattern. If the substrate temperature is high enough, the film is grown in layer-by-layer mode and the flat pattern persists for a long time. In periodic pattern growth, long surface diffusion length helps with the smoothness of the flat parts of the pattern but destroys the outline shape of the pattern. We found that the substrate temperature has to be a moderate value, not too low and not too high. The optimal value depends on the size of the pattern. A pattern with a bigger feature size can persist for a longer period of time. Finally, we suggest a modified definition of the persistence probability in order to have a probability that agrees better with the simulated morphology.

Acknowledgment

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Table of Contents

List of Figures

List of Symbols

ℓ	diffusion length
W	interface width
h	film height
P	persistence probability
t	growth time
ML	monolayer
L	substrate size
r	pattern size
h_0	pattern height
P_n	modified persistence probability
ε	percentage of accepted error
Δh	height of accepted error

Chapter 1 Introduction

Physics of surfaces and interfaces has been an interesting and active research topic in the field of condensed matter for a long time. There have been a lot of research work, experimentally and theoretically, on the study of thin film growth on flat substrates and scientists have gained profound understandings in the subject. On the theoretical side, many growth models have been introduced [1-7] in order to use computer simulation technique as a tool to study kinetic properties of the evolving surface.

These days, growing interests in nanotechnology make the surface and interface physics even more important. Modern techniques can imprint nano and sub-nano scale structure on a substrate and then grow thin film on top of the substrate in order to fabricate nano scale device such as quantum dot and quantum wire. The process of growing thin film on a substrate with a specific structure is known as patterned substrate growth process. Obviously, the goal in this process is that after we grow thin film on top of the substrate, the pattern should still exist on the film surface. In order to be able to control the growth process and have a high quality film, i.e. a film that the original pattern on the substrate survives the growth process, we also need to determine parameters that give information about the pattern survival rate. There is no guarantee that the same knowledge scientists have acquired regarding growth process on a flat substrate will still be valid with patterned growth. Hence, there is an increasing interest in the study of patterned substrate growth process [8-10].

In our study here, we focus on the theoretical aspects of molecular beam epitaxy growth on patterned substrate. To be specific, our goal is to use a simulation model to investigate a growth process with a predetermined pattern embedded into the substrate before the film is grown. We want to determine an appropriated parameter or parameters which can tell us about the survival rate of the pattern. We also want to determine growth conditions that help keep the original pattern after the growth process is completed. We note that this study is not aimed only to acquire knowledge to control patterned substrate growth process in experiments, but it also has an interesting aspect from a fundamental point of view. The flat substrate growth and patterned substrate growth are, in fact, two physical processes that are very similar to each other. The only difference is their initial conditions. If we can have insights into a patterned substrate growth, we can compare this with the flat substrate growth knowledge in literature [1-5], and see whether these two seemingly similar processes share the same physical properties. This will let us know how much the initial conditions can impact a physical process.

Chapter 2 Models and Methods

In this work, our goal is to study molecular beam epitaxy (MBE) growth of thin film on patterned substrates. MBE is a technique that can grow very high quality thin films. It is used in the fabrication of such things as optoelectronic devices, quantum structures, and other devices that require very high accuracy. In the MBE technique, neutral atoms or molecules of the film material are thermally evaporated from a source or sources. These atoms form a beam that is directed toward a heated substrate. The atoms are deposited on the substrate. Some of the deposited atoms have enough energy to leave the surface and desorp back. Most of the deposited atoms, however, stay on the substrate and diffuse around on the surface before eventually be incorporated onto the film. The growth process is generally performed with a slow growth rate which is determined by the number of averaged layers grown per unit time. There are three main mechanisms that affect the grown surface. These are the deposition, diffusion and desorption of the atoms. In most MBE processes, very few atoms desorp from the surface so in the study of MBE growth the desorption process is usually neglected. That leaves two major competing processes: deposition of atoms that induces roughness on the film versus surface diffusion of atoms that smoothens the film. In experiments, the controllable parameters associated with these two processes are the deposition rate and the substrate temperature. The temperature of the substrate is the factor that determines energy, and hence mobility, of the surface atoms. MBE growth can also be studied theoretically as well, especially via computer modeling. In our study, we use results from simulation results of a computer model to try to understand patterned MBE growth.

2.1 Discrete Growth Models

Since the study of MBE growth by the use of computer simulations has attracted a great deal of attention from researchers in the field, many discrete growth models are proposed [1-7] to describe the kinetic phenomena of surface growth. These models are also used as tools to determine essential properties of the problems.

The model we use in this study is the Das Sarma-Tamborenea (DT) model [4] introduced in 1991 as a simple model to study MBE growth on a flat substrate. For comparison, however, we discuss two other well known growth models in this section too. These are the Random Deposition (RD) model [1] and the Molecular Beam Epitaxy (MBE) model [5].

All models discussed here are under solid-on-solid constraints. This means there is no desorption, no overhanging, and no bulk vacancy formation on the growth front. All deposited atoms must become part of the growing film. Periodic boundary condition is also used in all simulations to prevent edge effects. Each atom is simplified to a simple unit square block of equal size. Details of the deposition and diffusion processes depend on rules of each model.

2.1.1 Random Deposition Model

The RD model is the simplest discrete growth model that includes only the deposition process while desorption and diffusion of atoms are not allowed [1]. An atom is dropped on a randomly chosen site. The atom falls vertically until it reaches the top of that random site and then it is incorporated there permanently. This completes a deposition of one atom and the process is repeated for the next atoms until the film reaches desired thickness.

The characteristic of this model is that each lattice site is grown independently and the surface of the growing film is uncorrelated. The interface of the film is extremely rough because there is no diffusion at all. The RD model is generally studied statistically from the point of view of statistical physicist. However, for the study of MBE growth, the model is too unrealistic and not a suitable model.

2.1.2 Molecular Beam Epitaxy Model

To create a more realistic model, a so-called MBE model is introduced [1,5]. In this model, deposition and diffusion processes are included. All atoms on the surface can diffuse. The hopping rate of each surface atom is calculated from the Arrhenius hopping rate which depends on the substrate temperature and the initial bonding configuration of that atom. The atoms continue to hop around until they are buried by other atoms. This model is sometimes called the full diffusion MBE model to emphasize that all surface atoms can diffuse.

The MBE model is one of the most realistic discrete models because of its extensive details in the diffusion process. However, everytime that a surface atom hop to a different site, bonding configurations of other surface atoms in the vicinity are changed. So after one hop made by one atom, hopping rates of all surface atoms in the area must be re-calculated. The simulation of this model requires a lot of computational time and so the model cannot be used for a system with a large substrate.

2.1.3 Das Sarma-Tamborenea Model

The DT model was introduced in 1991 by S.Das Sarma and P.Tamborenea [4,11] as a simple model for MBE growth. It was created in such a way that its diffusion rules are much simpler than the MBE model but still captures the essence of real MBE growth process. The model includes the deposition and the diffusion on the interface but does not allow any atom on the surface to desorp. In this model, the diffusion process is instantaneous, i.e. each atom is deposited on to the substrate at a randomly chosen site, diffuse to a “better” site and then the atom is incorporated at that latter site. Once the atom is incorporated, it becomes a part of the growing film and can no longer move. This rule makes the DT model much easier to simulate compared to the full diffusion MBE model.

In the DT model, each atom is assumed to be a simple cubic. Atoms are dropped onto the substrate at random sites. The important thing is, in this model, only one atom is dropped at a time. After the atom reaches the bottom, it can diffuse within a finite diffusion length ℓ to search for its final site according to the diffusion rule of the model. Once the atom finds its final site, it stays there permanently and the next atom is dropped.

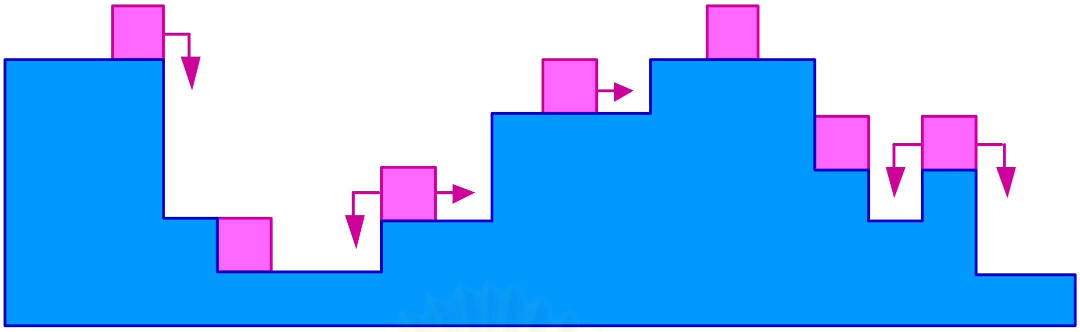

Figure 2.1 A schematic diagram showing the diffusion rule for the DT model in one dimensional substrate. The diffusion length here is $\ell = 1$.

The diffusion rule for this model [4,11], shown schematically in Fig. 2.1 for $\ell = 1$ case, is quite simple. At first the atom checks to see the number of nearest neighbor bonding it can have at the random site it is dropped upon. If there are at least two bonds, the atom will choose that random site as its incorporation site and it will stay there without diffusing any further. However, if the atom can have only one bonding (with a neighbor directly beneath it), it will search its nearest neighbors and see how many bonding it can form if it goes there. If one of the nearest neighbors offers more bonding than the original site, the atom diffuses there. If there are many (i.e. more than one) nearest neighbors that offer more than one bonding, then the atom chooses its final site by a random pick among those nearest neighbors. However, if all the nearest neighbors offer only one bonding each, the atom will search further away from its original site. If the atom reaches its limit, i.e. the pre-determined diffusion length ℓ, and still has not found a better site than the original site, then it stays at its original site. We should emphasize here that the possibility that two atoms will "fight" for the same final site does not exist. This is because in the DT model, only one atom is allowed to move at a time. Once the moving atom chooses the final site and be incorporated at that site, only then the next atom is dropped and allowed to move on the surface.

2.2 Surface Roughness

In the study of thin film growth, the first thing we can investigate is the surface morphology of the grown film. In simulation it is easy to plot the height of the surface as a function of the position on the substrate. From the plot, we can usually judge how “rough” the film is just by taking a look at the morphology. But our judgment is not universal and what considered rough by one researcher may be considered smooth by another researcher. So we need a quantity that can judge “roughness” of a thin film surface universally. That quantity is the interface width W defined as [1]

$$W(t) = \left\langle \left(h(x,t) - \bar{h}(t)\right)^2 \right\rangle_x^{1/2}$$

where $h(x,t)$ is the height of the surface at site x at time t, $\bar{h}(t)$ is the average height of the film at that time which can be thought of as the average thickness of the film, and the brackets $\langle ... \rangle_x$ means the quantity in the bracket is averaged over all value of x on the substrate.

From the definition of the interface width, it can be interpreted that W tells us how much the actual height of the film at various position on the substrate differ from its average value. This is, by definition, the standard deviation of $h(x,t)$. In general, the interface width increases as a function of growth time. It means, in general, the longer you grow the film (or the thicker the film), the rougher the film surface becomes.

2.3 Persistence Probability

When studying growth on a flat substrate, the most important thing is to determine how smooth, or how rough, the film is as deposition process continues through time. A quantity used to determine the roughness of a film is the root mean square value of the height fluctuation, also known as the interface width, of the film. However, when the film is grown on a patterned substrate, the most important thing is not the roughness of the film anymore. The question has now become, how much of the original pattern survive the growth process. To answer this question, the persistence probability is used in the study of thin film growth on patterned substrates. The persistence probability is conventionally defined as [8]

$$P(t) = \left\langle \prod_{s=1}^{t} \delta_{h(x,s),h(x,0)+s} \right\rangle.$$

Here $h(x,s)$ is height of the simulated surface at the lattice site x at time s and $h(x,0)+s$ is the height of the substrate at site x (i.e. before the growth process begins because time is set at zero) plus the average thickness of the grown film. Basically, $h(x,0)+s$ is the desired height of the film at the site x. The notation δ is the Kronecker delta function which takes on the value of unity when its two indices are equal and becomes zero otherwise ($\delta_{i,j} = 1$ when $i = j$), $\prod$ denotes the product of the delta function from time $s = 1$ ML through $s = t$ ML, and the angular brackets $\langle ... \rangle$ represents the average of the quantity over all lattice sites on the substrate.

By definition, $P(t)$ means that survival of the pattern at time t is counted only when the initial pattern is reproduced exactly every time after each monolayer deposition, from the very beginning of the growth process until the film is grown to t ML.

2.4 Noise Reduction Technique

In all atomistic models, there are unavoidable stochastic noises during the growth process. The most dominant noise is the noise associated with the deposition process. When using a discrete model such as the DT model, the simulation starts from the deposition of thin film process which is a process where atoms are deposited on the substrate at randomly chosen sites. This randomness produces noise which causes roughness on the surface. To reduce the noise effects, a noise reduction technique is utilized in this work.

Here, we choose to use the long surface diffusion length noise reduction technique [12]. In this technique, the surface diffusion length (ℓ) of each atom is increased from the default value of unity. This means we increase the maximum lateral length that an atom can move. It is obvious that an atom with a longer diffusion length ($\ell > 1$) will have more chance to find a more appropriated incorporation site. In the DT model, the atom with a long surface diffusion length will be able to search further away from its random deposition site for the incorporation site with a large coordination number. This situation will result in a smoother interface.

Although the long surface diffusion length here is just a computer simulation technique, it actually relates to a controllable parameter in experiments: the temperature of the substrate T. When T is increased, the surface atoms have more thermal energy which raise their mobility because they can break bonds at their original deposition sites and travel for a relatively large distance. So large T corresponds to large ℓ. To be explicit, the diffusion length varies with the substrate temperature as [13]

$$\ell \propto \exp\left(- \frac{1}{k_B T} \right)$$

when k_B is the Boltzmann constant. It should be noted here, however, that the substrate temperature cannot be set too high because it is assumed in this study that the desorption rate is too small to have any effect on the surface.

Chapter 3 Results and Discussions

In this chapter, we present our simulation results and offer discussions. The results are separated into 2 groups. The first group is results of growth on a "flat pattern" substrate. The second group is from growth of thin films on substrates with periodic patterns. All simulation results presented here are from our studies of the DT model on one dimensional substrates with the periodic boundary condition to prevent edge effects.

3.1 Flat Patterned Growth

The initial configuration for flat patterned growth is that height at the starting time ($t = 0$) is set to zero at all lattice sites, i.e. $h(x, t = 0) = 0$ for $x = 1,2,3,...,L$ when L is the size of the substrate. Three quantities are studied in this case: the surface morphology of the simulated film, the interface width (W), and the persistence probability (P).

Let us first investigate the film morphology. Fig. 3.1 shows the time evolution of a typical surface of a film simulated from the DT model with only nearest neighbor diffusion, i.e. $\ell = 1$. It is clear from Fig. 3.1 that once the deposition process begins, the film roughness increases. This is expected as the diffusion rules allow atoms to move only to their nearest neighbors at the most, so it is difficult for atoms to find the most appropriated sites that will help maintain the smoothness of the surface. In order to improve the film quality, the long surface diffusion length noise reduction technique was applied. The results are shown in Fig. 3.2 where surface morphologies after 100 ML deposition are illustrated. The diffusion length is set to be $\ell = 0, 1, 10, 50$. The $\ell = 0$ simulation is obviously just the RD model and the surface is extremely rough. The $\ell = 1$ system is the original nearest neighbor diffusion DT model as seen in Fig. 3.1 (the middle line at 100 ML). The top two lines here show the DT morphologies with longer surface diffusion length. It is striking how smooth the surfaces become when the moving atoms are allowed to search further away from the randomly chosen deposition sites. The longer an atom can move, the smoother the

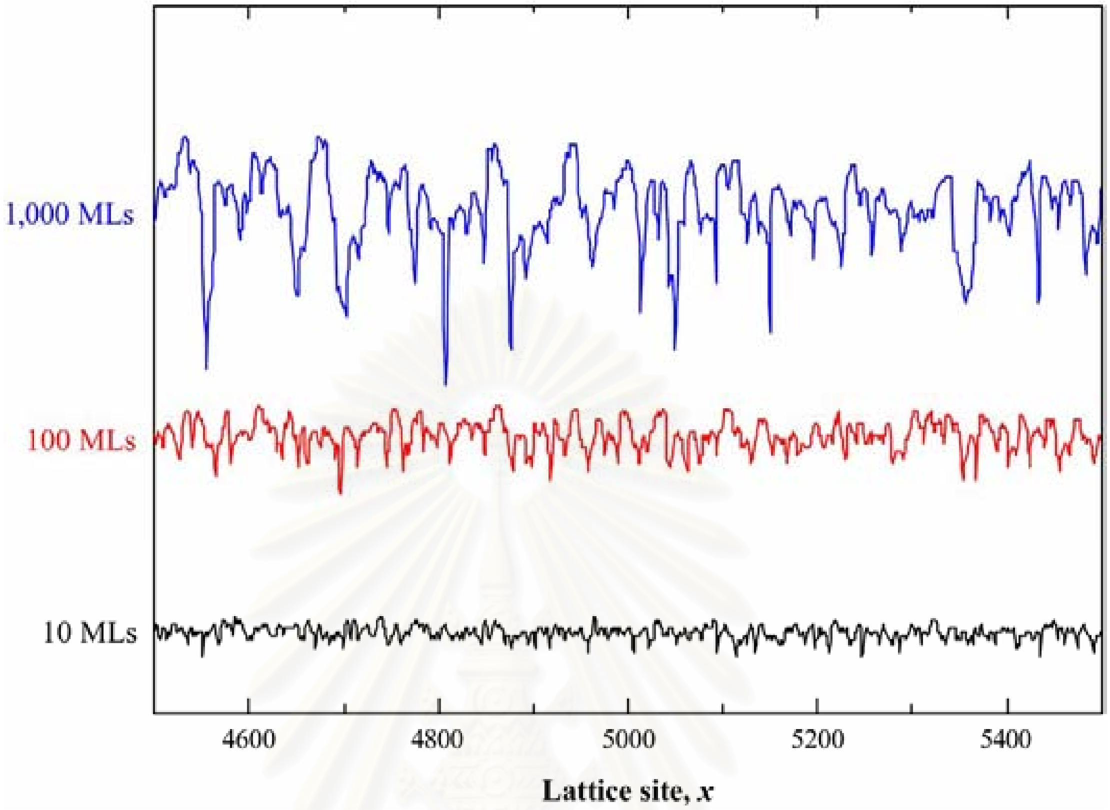

Figure 3.1 Dynamical morphology shows the time evolution of the kinetically rough thin film grown by using the DT model. The diffusion length is fixed at $\ell = 1$.

surface is because the moving atom has more chance to diffuse to a site with larger numbers of bonding, i.e. a site that is a groove/pit in the surface.

The interface width results for the DT simulations on flat substrate growth are shown in Fig. 3.3 for various values of ℓ. As discussed earlier in section 2.2, W increases with time in general. Results in Fig. 3.3 confirm that. It is also found here that when the diffusion length of moving atoms increases, which means the atoms have higher mobility, the surface roughness decreases. This result agrees with the morphologies in Fig. 3.2 which show that when ℓ is larger, the surface is smoother. There is a striking characteristic in Fig. 3.3 ; the oscillation of W in $\ell > 1$ systems which is especially obvious when $\ell = 50$ and $\ell = 100$. The oscillation indicates that the thin film is being grown in layer-by-layer mode [12] which means one layer is grown completely before the next layer is formed. Starting from a flat layer, atoms are incorporated into the film as the next layer is grown and W increases during this

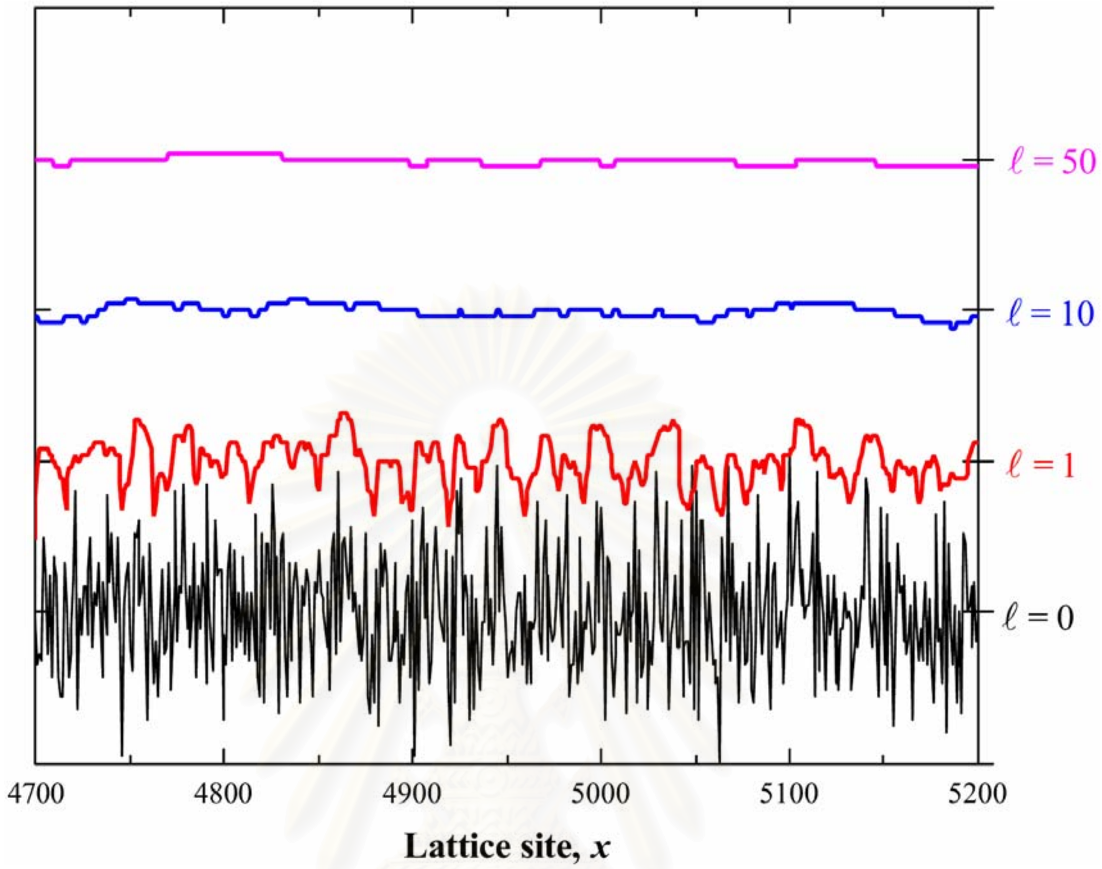

Figure 3.2 Morphologies at $t = 100$ MLs for the DT model with various diffusion length.

process until the interface width reaches its peak when the layer is half filled. As new atoms are incorporated into this half filled layer, the standard deviation of $h(x,t)$ decreases. W reaches its minimum value when the layer is completely filled, forming a flat surface again. The whole process is then repeated for the next layer. Eventually, the film cannot be grown in this layer-by-layer mode due to the stochastic noise associated with the growth process, so the amplitude of the oscillation decreased continuously in time until the oscillation eventually damp out completely. Note that this layer-by-layer oscillatory surface roughness is seen experimentally in RHEED results as well when the substrate temperature is sufficiently high.

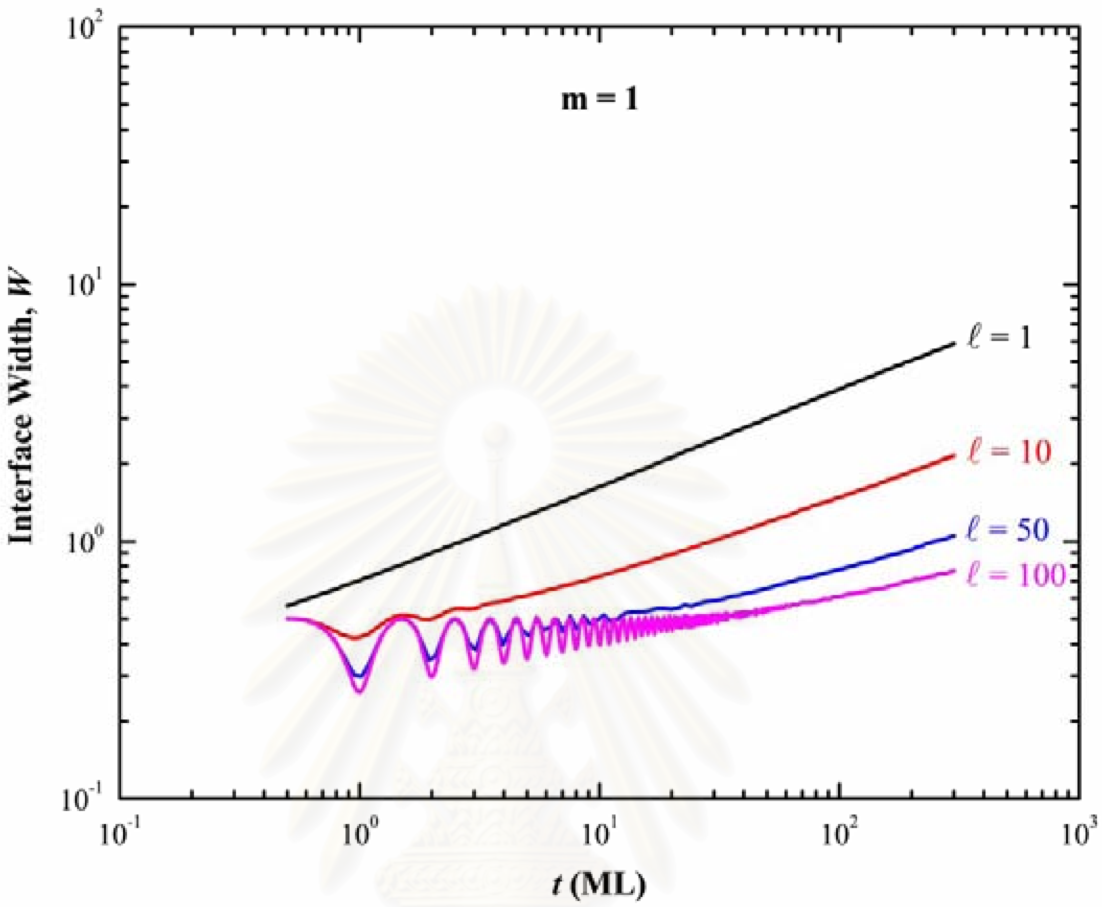

Figure 3.3 The interface width vs. time plots for various diffusion length.

In the flat patterned substrate growth, the pattern here is just a completely flat substrate. So the goal is to keep the film flat after growth process is finished. From the interface width plot in Fig. 3.3, we can conclude that increasing the diffusion length to a very large value can help reduce the surface roughness. However, when ℓ is small and the film is rough, can we decide how much of the "pattern" is still maintained? To answer this question, we need to study the persistence probability introduced earlier in the literature [8]. Our $P(t)$ results are plotted as a function of time in Fig. 3.4. Let us concentrate on the original DT model with $\ell = 1$ first. In this system, the morphology is very rough and the interface width increases rather rapidly in time. It can then be assumed that the persistence probability of the pattern in this system should decrease quickly in time as well. This is exactly what is seen in Fig. 3.4, that $P(t)$ starts from its maximum possible value ($P = 1$) and then decreases to $P \approx 10^{-5}$ before the film thickness reaches 20 ML. Since the substrate here is $L = 10{,}000$ lattice sites, the probability $P(t) = 10^{-5}$ means only an average of one site in the whole substrate can maintain the "theoretical height" throughout the growth process up to the time t. When the diffusion length is increased, both the morphologies and the interface width results suggest that more of the original pattern can survive growth process for longer period of time. The results in Fig. 3.4 agree well with that assumption, with the decay rate of $P(t)$ smaller when ℓ is larger. From this plot, we can determine how thick we can grow the film at a fixed ℓ when the acceptable level of persistence probability is specified. For example, when we need at least 1% of the pattern in the grown film, i.e. $P(t) \geq 10^{-2}$, we can grow the film to approximately 40 ML with $\ell = 100$. However, if the diffusion length in the system is $\ell = 1{,}000$, then the film can be growth to approximately 80 ML. It should be emphasized that ℓ is not just a computational parameter. The diffusion length relates directly with the temperature of the substrate in experiment. To have larger ℓ, one needs to increase the substrate temperature when growing a real film experimentally.

There is also an interesting point in Fig. 3.4, that for $\ell = 5{,}000$, $P(t) = 1$ throughout the whole process, which is up to 200 ML in this study. This data means after 200 ML deposition, the surface of this film is still totally flat like the substrate. It can be explained that, in the system with $L = 10{,}000$ that we have here, $\ell = 5{,}000$ indicates that all atoms can search the whole substrate for the most appropriated sites. There will not be any groove left after each layer is grown. The layer-by-layer growth mode can be maintained throughout the entire growth time up to infinite film thickness. This is a little interesting bit from the computational point of view but has no real impact on experiments because it is not possible to have a system with atom mobility being so large that the diffusion length reaches $5{,}000$ lattice sites. In order to do so, the substrate temperature must be extremely high that the substrate, and the chamber for that matter, will melt away long before we can reach the desired diffusion length.

To summarized this part, we have found that the flat pattern on the substrate can last for a longer period of time when the diffusion length in the model is increased. However, when the model followed nearest neighbor diffusion rule, the flat

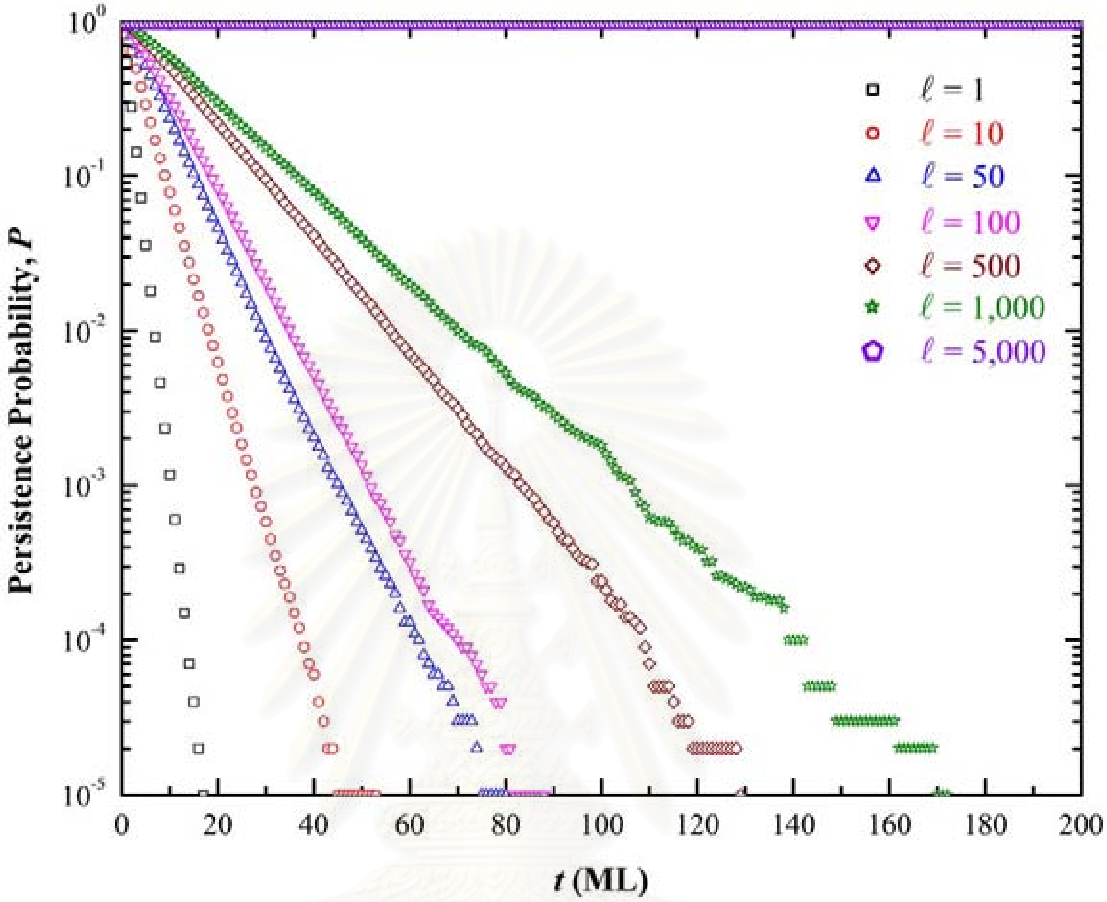

Figure 3.4 The persistence probability versus time plots for the DT model with various diffusion length.

pattern of the substrate was destroyed very quickly after deposition process begins. This is confirmed by the study of the surface morphologies, the interface width versus time plots, and the plots of the persistence probability as a function of growth time. There is no limit to how large the diffusion length can be in our simulations as it was found that the survival rate of the original pattern continue to improve as the diffusion length becomes larger.

3.2 Periodic Patterned Growth

The flat patterned growth presented in the previous section seems to be trivial because it is actually just surface growth problems which have been done on flat substrates. It was studied here from the patterned growth point of view for the sake of completeness and also for comparison with results from growth on other types of pattern. This was also a good chance for us to have a better understanding of the nature of the persistence probability. In this section, we continue our study by imposing a periodic pattern on our substrates. This type of growth process is a very interesting case because it offers a new technique to fabricate highly ordered nanostructure devices. In our study here, the initial pattern is designed to be a series of blocks placed at an equal interval throughout the substrate. All the blocks have the same shape and size. The width of each block is denoted by r and the original height of each block is h_0. This is illustrated in Fig. 3.5 for a system with a substrate of size $L = 10{,}000$, $r = 1{,}000$ and $h_0 = 100$ lattice sites. To understand this problem, we utilized the same tool to study the survival rate of the pattern as in the previous section, i.e. the persistence probability.

From our results of flat patterned growth in the previous section, it is clear that the long surface diffusion length can indeed reduce noise and help the original pattern to persist through a long period of growth time when compared with the $\ell = 1$ model. With this knowledge in mind, we expect similar results for the periodic patterned

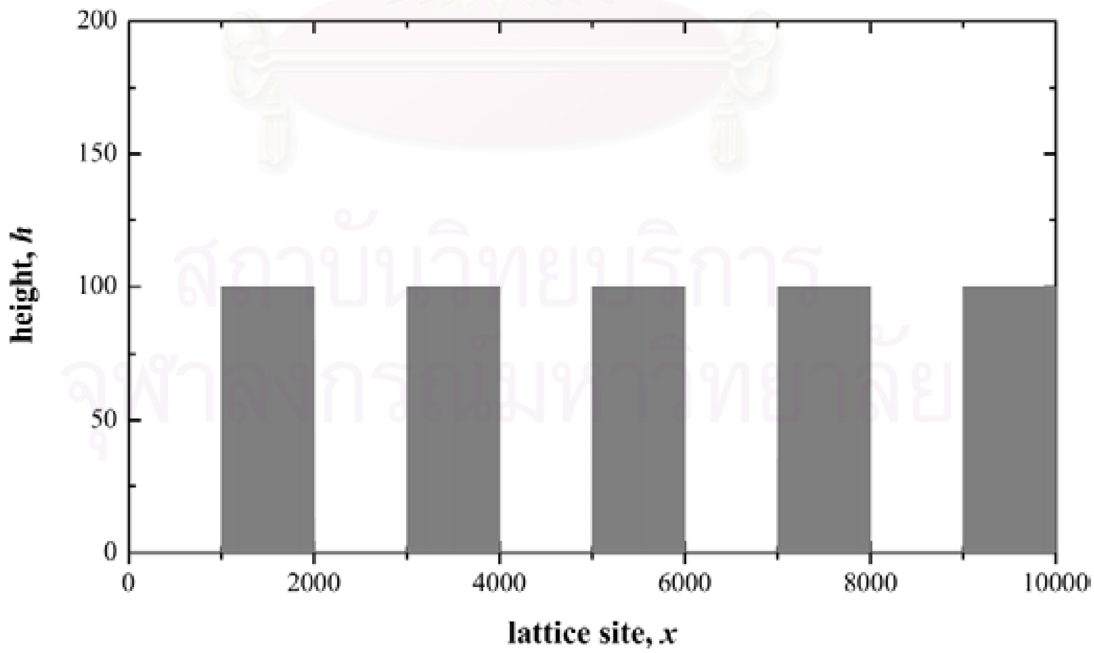

Figure 3.5 The periodic patterned substrate.

growth in this section. However, our results in Fig. 3.6 indicate that our expectation is not correct as longer diffusion length does not always provide better survival rate for the periodic pattern growth. The details are seen in Fig. 3.6. When ℓ is increased from 1 to $10, 50,$ and then 100, the decay rates of the persistence probability continue to decrease as we expect. When $\ell = 500$, however, $P(t)$ decreases rapidly after 100 ML. In fact, after approximately 135 ML, the persistence probability of the system with $\ell = 100$ is better than that of the system with larger diffusion length, i.e. $\ell = 500$. For systems with an even longer diffusion length ($\ell = 1000$ and $\ell = 5000$ in Fig. 3.6), the persistence probabilities reduce with a dramatic decay rate and it looks like the original pattern is destroyed after only a few mono-layers deposition.

To understand this difference between the flat pattern and periodic pattern growth, we investigate the time evolution of the growing morphology for the system with $\ell = 1000$. The results are shown in Fig. 3.7. The grey area indicates the original patterned substrate before the growth process starts. This is a periodic pattern system with $L = 10{,}000$, $r = 1{,}000$ and $h_0 = 100$ as already shown in Fig. 3.5. The red area shows the addition to the film after 30 ML deposition while the blue and pink area indicate the film after 50 and 100 ML deposition respectively. It is obvious from these morphologies that during the first 30 ML, the newly deposited atoms do not nucleate on top of any of the original blocks at all. All newly deposited atoms hop down to fill the empty space between the blocks. Although we did not expect this result, it actually makes sense. This is because the diffusion rule of the DT model requires that a moving atom search for a final incorporation site where it can increase number of bondings. So when a new atom is dropped on top of a completely flat block, it has only one bonding with its neighbor underneath it and no lateral bonding at all. According to the diffusion rule of the model, the atom searches, within $\ell = 1000$ sites, for a site that can offer larger number of bond. The atom finds that final site at the bottom edge of the block where it can form one bond with the neighbor below and one lateral bond with the side of the block. This leaves the top of the block flat as before, and any new atom deposited on the top of the block faces the same situation, and eventually ends up at the bottom in the similar process. After 30 ML deposition,

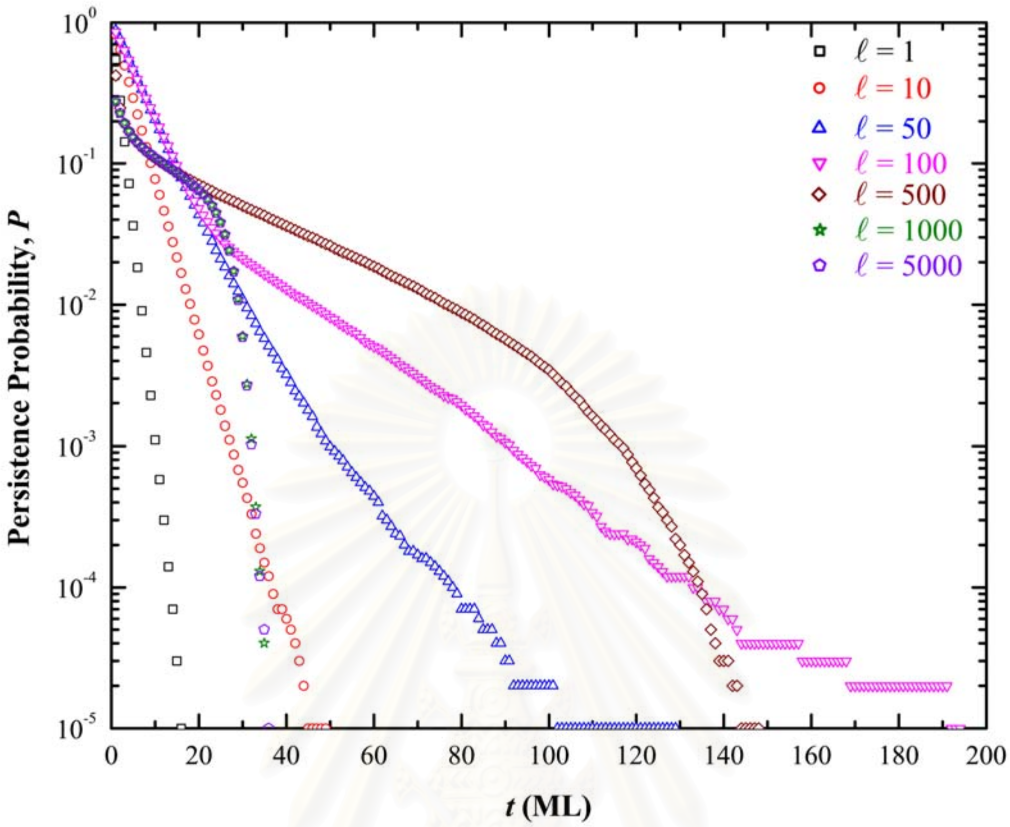

Figure 3.6 The persistence probability versus time plots for the periodic patterned growth with various diffusion length.

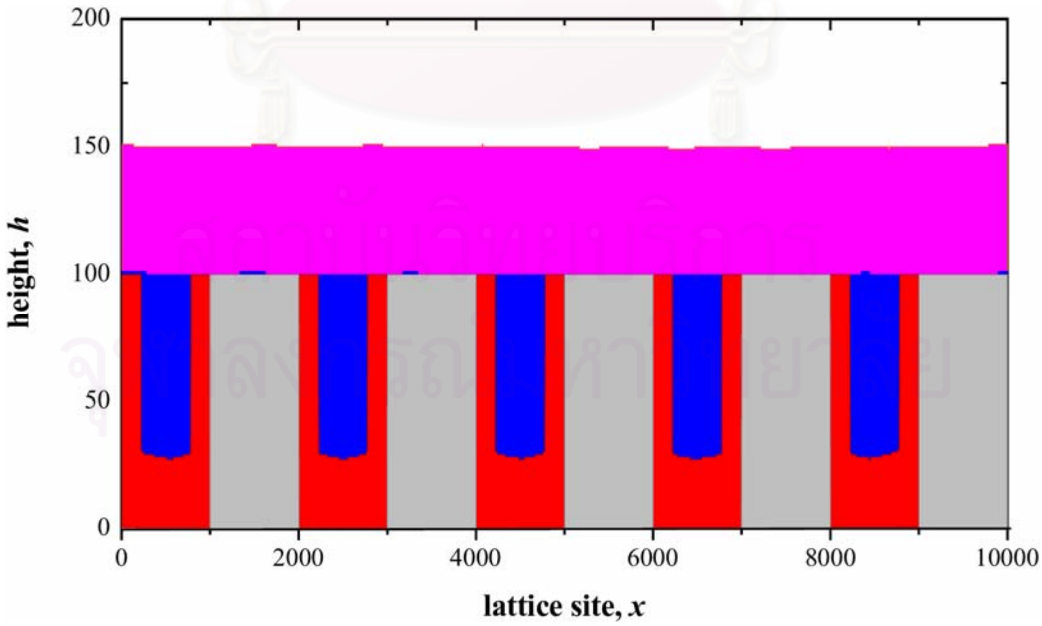

Figure 3.7 The periodic patterned film with a fixed $\ell = 1000$ at $t = 0$ (grey), $t = 30$ (grey and red), $t = 50$ (grey, red and blue), and $t = 100$ (grey, red, blue and pink) MLs.

the pattern of the film is changed to a series of blocks with $r > 1000$ and $h_0 < 100$. After a total of 50 ML deposition, a total of 500000 atoms are deposited, and that is the number of atoms required to completely fill all the empty spaces between the original blocks. The film is turned into a flat surface without any information of the original pattern left. Any additional deposited atoms do not “know” that there is a pattern hidden in the substrate, so the growth process after 50 ML is a flat substrate growth. Since the diffusion length in this case is comparatively large, the surface remains relatively smooth throughout the growth process.

These results shown in Figs. 3.6 and 3.7 and discussed extensively above point out that a longer surface diffusion length is not always good for patterned growth. However, nearest neighbor diffusion only ($\ell = 1$) is not good either as the surface of the growing film is too rough. So how big the diffusion length (in other words, how high the substrate temperature) should be in a patterned substrate growth? The answer to this question depends on the size of the original pattern, i.e. r in our simulations. Once the diffusion length is equal to $r/2$, all atoms can hop down from the top of the original blocks. A close examination of Fig. 3.6 leads to our conclusion as follows.

3.2.1 Effects of diffusion length and size of the pattern

When ℓ is very small ($\ell = 1$ and $\ell = 10$ in Fig. 3.6), the persistence probability decays in almost the same way as $P(t)$ in the flat substrate cases (Fig. 3.4), which is an exponential decay. This is because ℓ is so small when compared with r so the deposited atoms can search for final sites in the vicinity of the deposition sites only. In this situation, most of the deposited atoms do not come across the edge of the blocks, so they do not “see” the pattern. The diffusion situation for these atoms, which are the majority, are exactly the same as in flat substrate growth. So it is understandable that the persistence probabilities in the systems with very small diffusion length are similar to those from the flat pattern studies. This means the pattern is destroyed quickly because with small ℓ, the surface (which is originally smooth both on top of the blocks and between the blocks) becomes rough in a short time from the noise.

When ℓ is not too small but still less than $r/2$ ($\ell = 50$ and 100 in Fig. 3.6), the behavior of $P(t)$ in Fig. 3.6 can be separated into two time regimes. In the early time, $P(t)$ decay exponentially and the plots are straight lines. After 30 - 40 ML, $P(t)$ decay at smaller rates which means the persistence of the pattern is better. This is because the diffusion length is long enough to maintain some "smoothness" of the original pattern and at the same time the diffusion length is not too long (compared to the pattern size) so many atoms can be incorporated on top of the blocks and help keep the information of the pattern.

When $\ell = r/2$ ($\ell = 500$ in Fig. 3.6), the behavior of $P(t)$ is very interesting. In the very early time, the persistence probability decays very quickly (very sharp slope). After approximately 10 – 20 ML, however, the decay rate becomes smaller until after 100 ML that the decay rate becomes very large again. The reason for these changes is that at very early time, the diffusion length is exactly equal to half of the block size. This means all atoms deposited on the top of a block have to be incorporated at the bottom edge of the block and the persistence decays quickly. Soon afterward, the blocks in the film become slightly bigger. The increase of the pattern size means at this time the diffusion length (which is fixed at ℓ) is less than half the size of a block and some atoms can stay on the top surface. This helps with the survival rate of the pattern and the persistence probability decay rate slows down which means the film can keep the pattern for some time.

And finally, when ℓ is very large, i.e. $\ell \geq r$ ($\ell = 1{,}000$ and 5,000 in Fig. 3.6), the persistence probabilities in these cases decays at a dramatic rate, i.e. the pattern is destroyed within an extremely short deposition time. This is because none of the newly deposited atom is allowed to stay on top of the blocks, as described in detail earlier.

From the above results, a diffusion length that is too small or too large compared to the pattern size (r) is not a good choice for periodic patterned growth because the "pattern" can be thought of a combination of two things: a series of flat surfaces on the top and bottom of the blocks, and the shape of the blocks. Both must be maintained throughout the growth but they require different technique. Long diffusion length is needed to maintain the smooth flat surfaces but it destroys the structure of the periodic block series. Small diffusion length will help keep the original block width for a long time but the flat parts become rough after a short growth time. The appropriated value for ℓ is in between a very small and very large value. From our results, the best value for ℓ seems to be a large value that is still less than $r/2$. To confirm that the optimal value of surface diffusion length depends critically on the pattern size, we study a system with a different r. In Fig. 3.8, the persistence probabilities of a system with $r = 200$ and varying ℓ are shown. The substrate size and the block original height are the same as before : $L = 10{,}000$ and $h_0 = 100$. It is obvious that the statistical behavior of $P(t)$ is the same as described before, with an abrupt drop of $P(t)$ when $\ell = 150$ which is greater than $r/2$ in this system. This leads to our conclusion that large diffusion length can still help with the

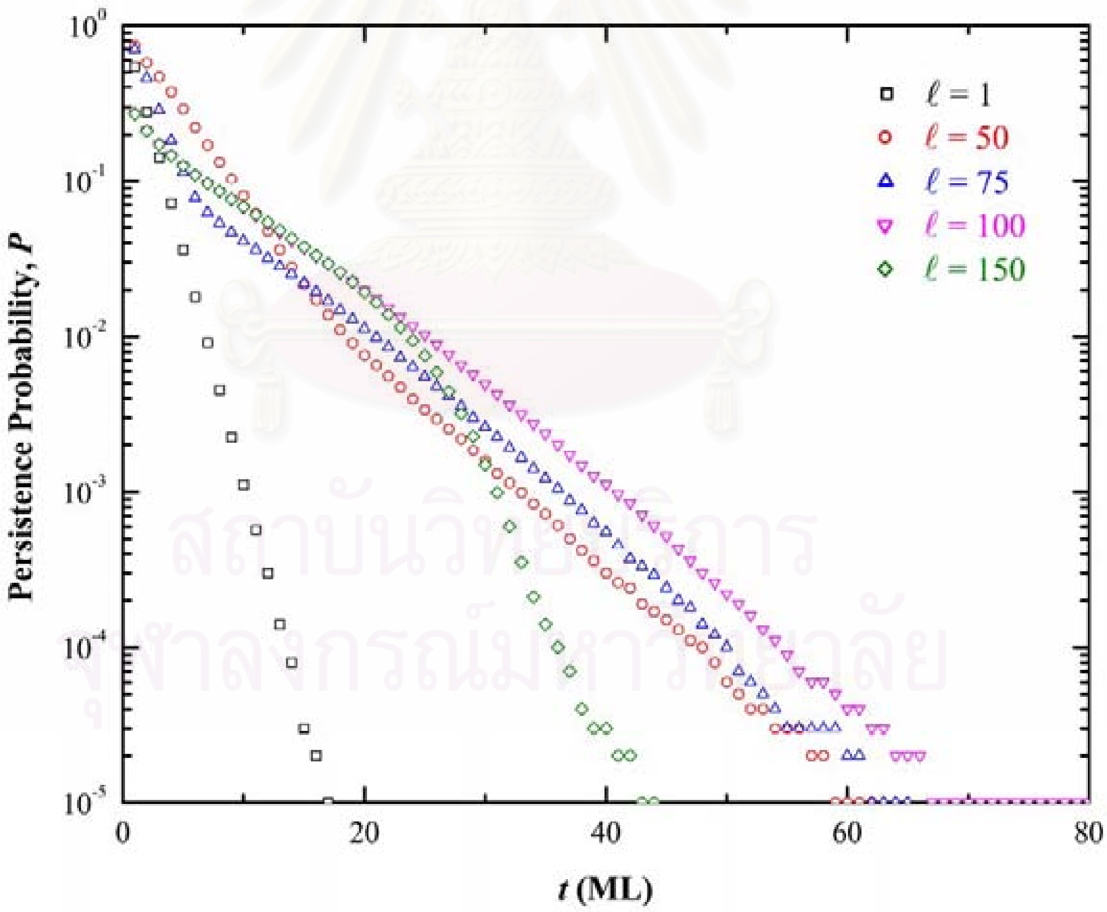

Figure 3.8 The persistence probability versus time plots for periodic patterned substrate with a smaller pattern size, $r = 200$. The diffusion length is varied.

survival rate of the periodic pattern similar to what we find for flat pattern growth. However, the diffusion length must be less than half of the pattern size. The size of the desired pattern is a very crucial factor to determine the most appropriated value of ℓ.

3.2.2 Effects of height of the pattern

The pattern size as discussed above is only the lateral size, i.e. the width of the block. Another factor we should consider is the vertical size of the pattern. So far the height of the block, h_0, is fixed at a constant value of 100. To study effects of h_0, we change the pattern height from 100 to 50 and 200. The results are shown in Figs. 3.9 and 3.10 for $h_0 = 50$ and $h_0 = 200$ respectively. In both systems, $L = 10{,}000$ and $r = 1{,}000$. When the diffusion length is relatively small ($\ell = 1, 10, 50$ and 100), the persistence probability of the two systems with different h_0 are statistically the same. In the case when $\ell = r/2 = 500$, however, it can be seen that the pattern persists better in the system with a larger h_0. This is because when ℓ is large enough, the empty space between blocks can be completely filled after a deposition of $h_0/2$ ML. So for the system with $h_0 = 50$, the empty space between blocks are completely filled after only 25 ML deposition while it take a much longer time, 100 ML deposition, to completely filled the space in the system with $h_0 = 200$. In other words, it takes more time to completely destroy the original pattern when the pattern height is greater, so the persistence probability in such systems decay slower. When $\ell > r/2$ the difference in the decay rates of $P(t)$ for various h_0 is even more obvious. Looking back at our results for $h_0 = 100$ in Fig. 3.6, it agrees with the above explanation as well.

It should be noted here that although the height of the pattern effects the calculated persistence probability when $\ell \geq r/2$, the value of h_0 has very little impact on $P(t)$ when the diffusion length is smaller. The vertical size of the pattern has no part in the determination of the optimum ℓ either.

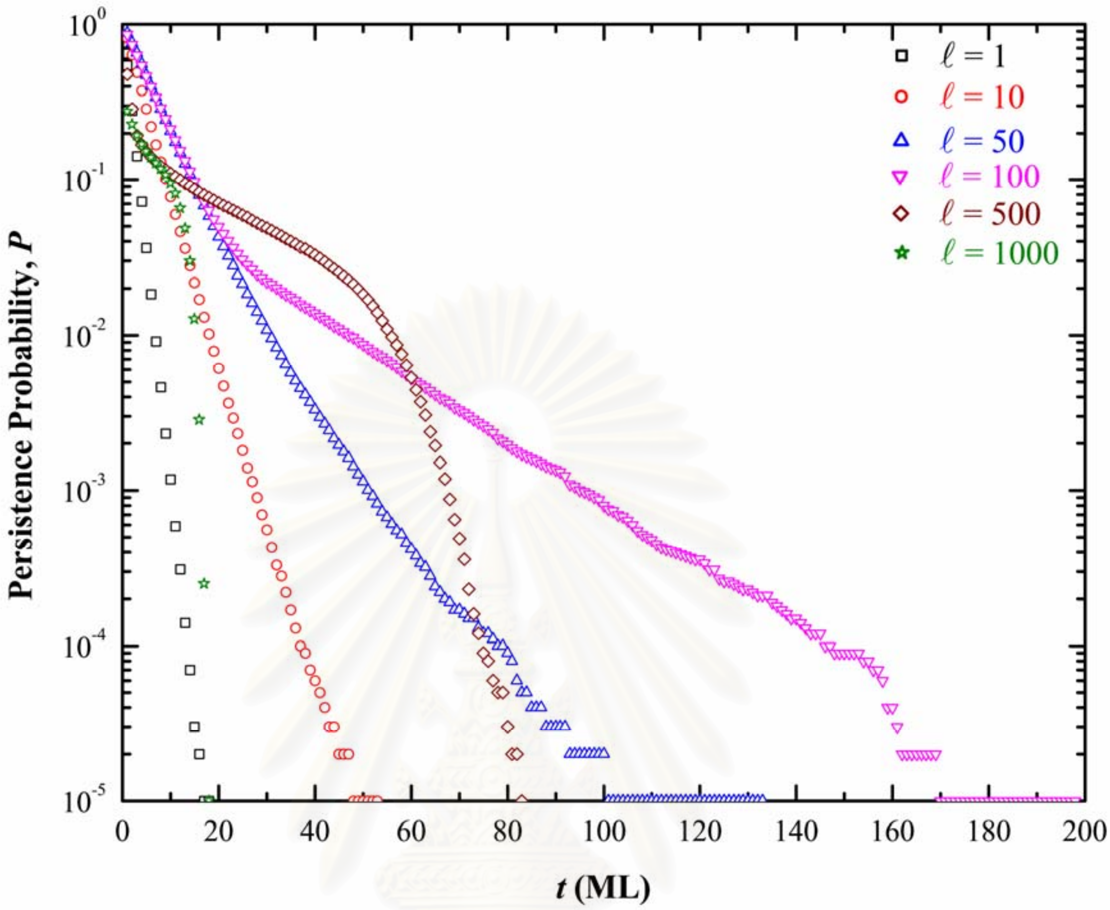

Figure 3.9 The persistence probability versus time plots for periodic patterned substrate with $r = 1000$ and $h_0 = 50$. The diffusion length is varied.

3.3 Modified Persistence Probability

All the persistence probabilities presented here in our work are calculated from the conventional definition of $P(t)$ [8] as defined in section 2.3 which is a very strict definition. The pattern is counted as "survive" only when the exact pattern is reproduced. A closer inspection of $P(t)$ shown in Fig. 3.6 raise an interesting question when compared with the morphologies results. Let us concentrate on the system with a moderate diffusion length at $\ell = 50$. Snapshots of surface morphology of the system at $t = 10$, 50, 100 and 200 ML are shown in Fig. 3.11. Although the simulated film we obtained after 50 layers deposition is relatively rough compares to the original substrate, the intended pattern – a series of blocks – can still be seen very clearly. The

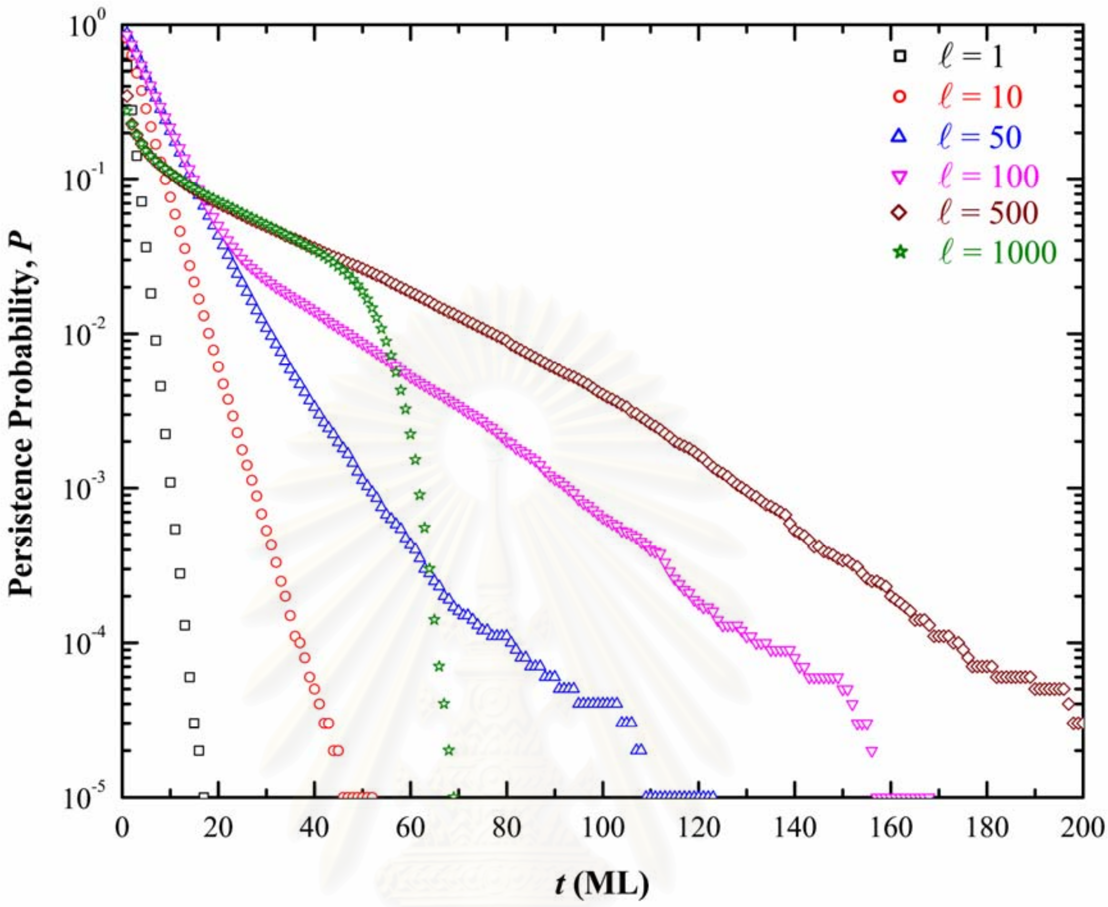

Figure 3.10 The persistence probability versus time plots for periodic patterned substrate with $r = 1000$ and $h_0 = 200$. The diffusion length is varied.

calculated persistence probability in Fig. 3.6, on the other hand, is only approximately 0.001 from the $\ell = 50$ data in Fig. 3.6, which is an extremely small value. From Fig. 3.6, $P(t)$ is practically zero at $t = 100$ ML while the obtained morphology in Fig. 3.11 still shows a good reproduction of the original pattern. Even at 200 ML where the calculated $P(t)$ is too small to be seen on Fig. 3.6 plot, the grown film still exhibits a clear series of block with approximately the same r and h_0 as in the substrate. From experimental point of view, this creates a question of how reliable the calculated persistence probability is.

Before addressing this question, we note that the decay of $P(t)$ is very rapid because of the way $P(t)$ is defined. We consider that the pattern survives only when the exact pattern is reproduced. Any minor changes from the original pattern will kill off the calculated persistence probability right away while in reality a slight “error” on the patterned films we obtain after growth process may still be considered a success. To better reflect this point, we modify the definition of $P(t)$ so that it is more flexible. This new and modified persistence probability is denoted $P_n(t)$ and is defined as

$$P_n(t) = \left\langle \prod_{s=1}^{t} F \right\rangle ,$$

where $$F = \begin{cases} 1 & ; [h(x,0)+s]-\Delta h \leq h(x,s) \leq [h(x,0)+s]+\Delta h \\ 0 & ; otherwise \end{cases}$$

when Δh is the limit of the acceptable “error”. It can be seen from the modified definition that this new $P_n(t)$ will not decay as fast as the original one because the pattern is counted as “survive” even if the obtained height is not exactly the same as the ideal height, as long as it differs from the ideal height, $h(x,0)+s$, by only a small number of layers.

To be more specific, the pattern is considered survived if the simulated height at time s, $h(x,s)$, is in the range from $[h(x,0)+s]-\Delta h$ to $[h(x,0)+s]+\Delta h$. Here Δh is an integer number calculated from the ideal height at that time times ε when ε is the percentage of error we are willing to accept. If $\varepsilon = 0$ then $\Delta h = 0$ and our modified persistence probability $P_n(t)$ returns to the original definition $P(t)$. In Fig. 3.12 we show a plot of $P_n(t)$ as a function of time when we choose the level of acceptable error to be 1%, i.e. $\varepsilon = 0.01$. The data shown here are from the same systems as in the plot of Fig. 3.6. For very small and very large diffusion length ℓ, the original definition and the modified version of persistence probability does not differ much. This is because when the diffusion length is too small, the surface becomes extremely rough and the obtained height is not in the acceptable range because ε is set to be very small. As for the situations when ℓ is too large ($\ell \geq r/2$), the pattern is destroyed quickly and the surface becomes completely flat without any block left, as

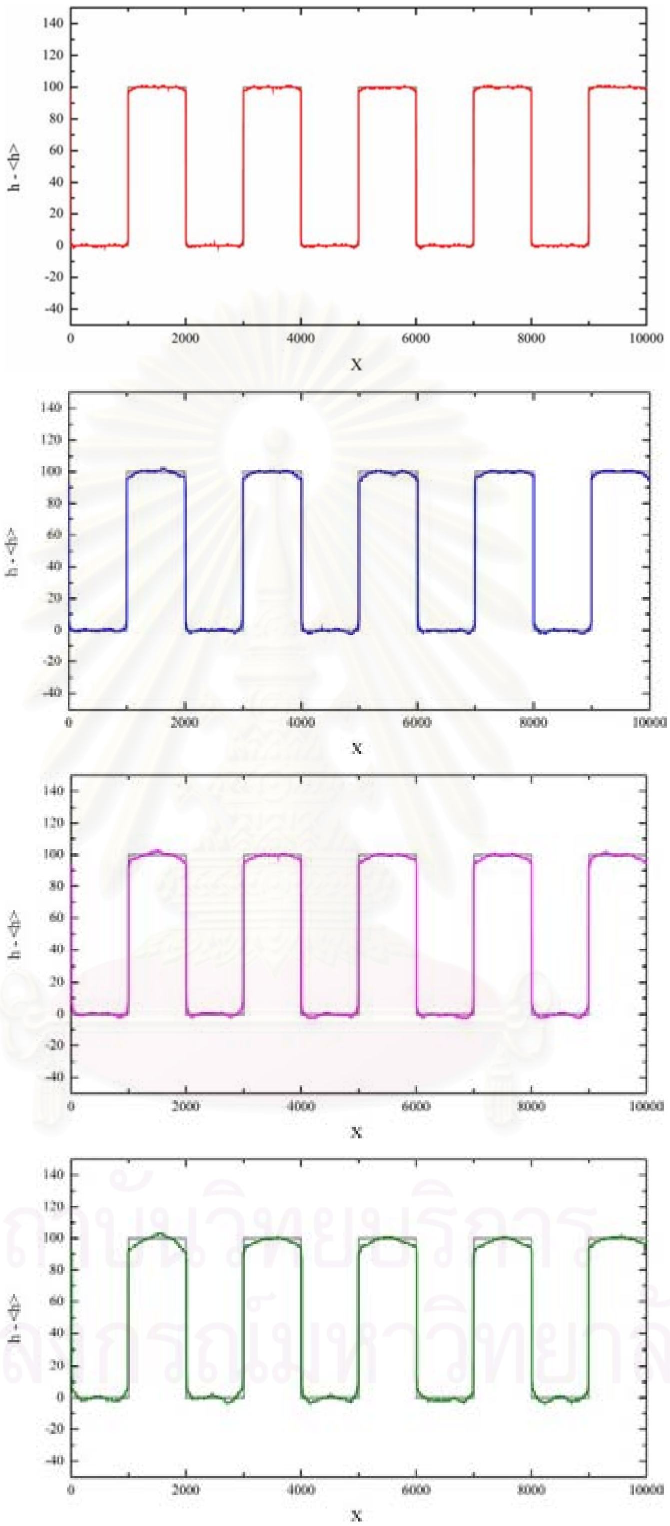

Figure 3.11 Dynamical morphologies of the periodic patterned substrate growth with $r = 1000$, $h_0 = 100$ and $\ell = 50$ at $t = 10$ MLs (a), $t = 50$ MLs (b), $t = 100$ MLs (c), and $t = 200$ MLs (d).

discussed earlier. So, again, the obtained height is not within the acceptable range. $P(t)$ and $P_n(t)$ yield practically the same values. However, for a moderate ℓ ($\ell = 50$ and 100 in the plots), it is clear that the modified persistence probability decays much slower than the original "strict" version. In Fig. 3.13, our results with various flexibility are shown. In the plot, we concentrate only on the moderate $\ell = 50$ case and the percentage of acceptable error is varied from $0\%, 1\%, 5\%$ to 10%. As expected, the larger ε yields the better persistence probability $P_n(t)$. It is very important to emphasize that our modified definition for the persistence probability does not change the behavior of the grown film at all. Rather, it changes the behavior of the persistence probability to agree better with the obtain morphology.

Although the new definition makes the persistence probability more flexible, it has a problem as can be seen in Fig. 3.13. For example, when $\varepsilon = 1\%$, before 50 ML the plot of $P_n(t)$ are the same as $P(t)$ (which is $P_n(t)$ with $\varepsilon = 0\%$). This is because when $t < 50$ ML, the calculated $\Delta h < 0.5$ and is rounded down to be zero since Δh has to be an integer. As time increases, Δh increases to be nonzero and the modified and the original version of the persistence probability start giving different values. This rounding number of the computer causes abrupt changes in $P_n(t)$ as seen in Figs. 3.12 and 3.13.

To avoid the above mentioned problem, we use another way to calculate the modified persistence probability. Instead of using the percentage of ideal height at each time step, Δh is chosen as a fixed constant for the whole process. Fig. 3.14 shows the re-modified persistence probability when $\Delta h = 1$ ML. Here we can see that when we accept the error of just 1 layer, the modified persistence probability decays much slower than the original definition (in Fig. 3.6) and the curves are smooth without any broken line as in Fig. 3.12. When the diffusion length is too large, we have seen from the morphology in Fig. 3.7 that the pattern is destroyed quickly and any definition of persistence probability reflects that as all version of $P(t)$ and $P_n(t)$ decay at a dramatic rate.

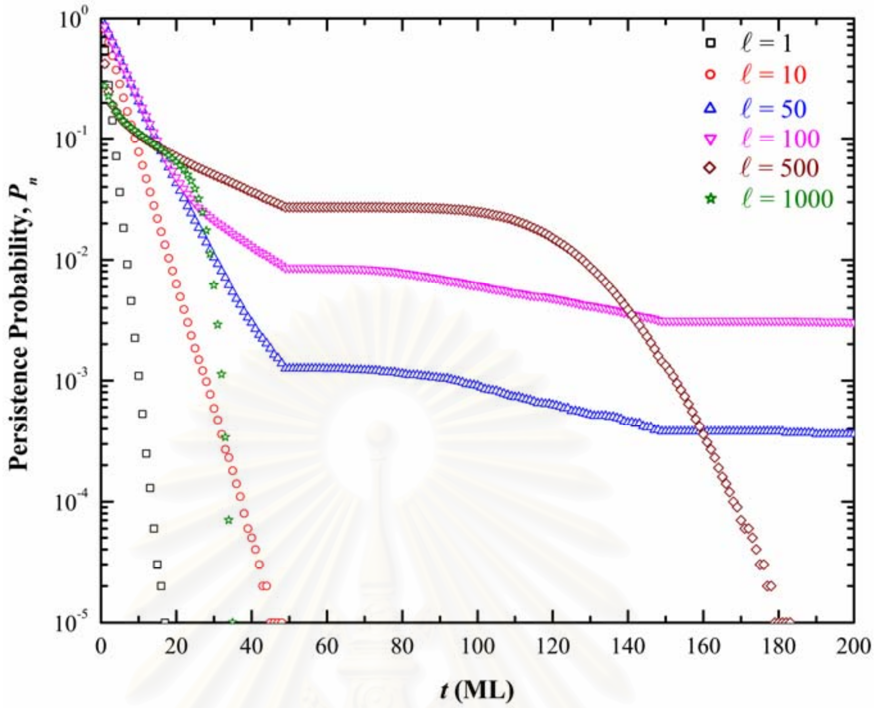

Figure 3.12 The modified persistence probability versus time plot for the periodic patterned growth when $\varepsilon = 1\%$.

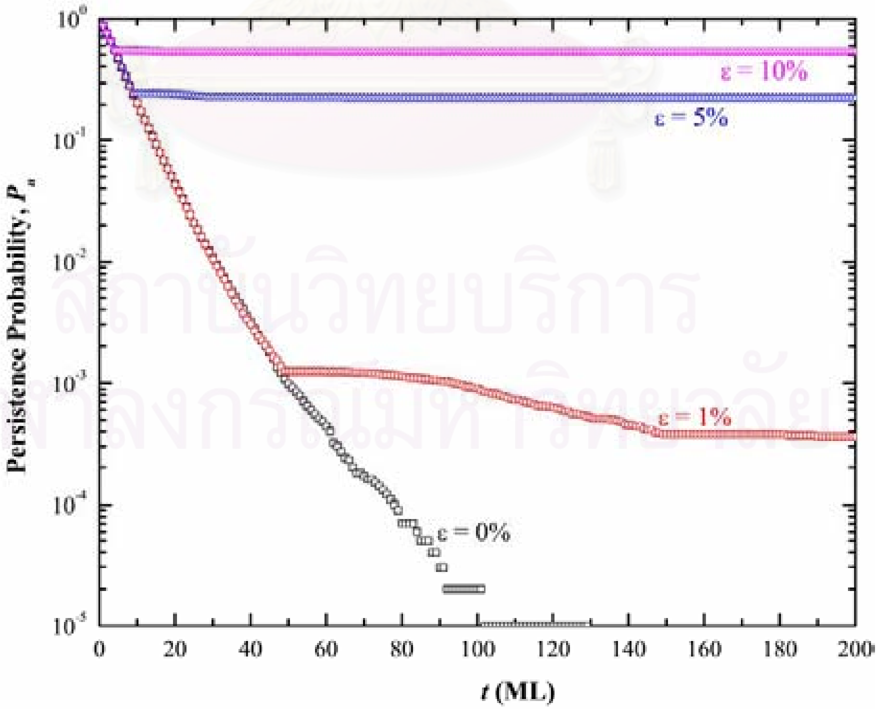

Figure 3.13 The modified persistence probability versus time plot for the periodic patterned growth with various ε.

With the new definition of $P_n(t)$, a new question comes up as how we should choose a proper value for Δh. In another word, we have to decide how much "error" we should allow. This seems to be a type of question that does not have one correct answer. This depends on the goal of each experiment, i.e. how accurate the film has to be for the purpose of that experiment. It will be up to the judgment of one who uses $P_n(t)$ as a parameter to determine the survival rate.

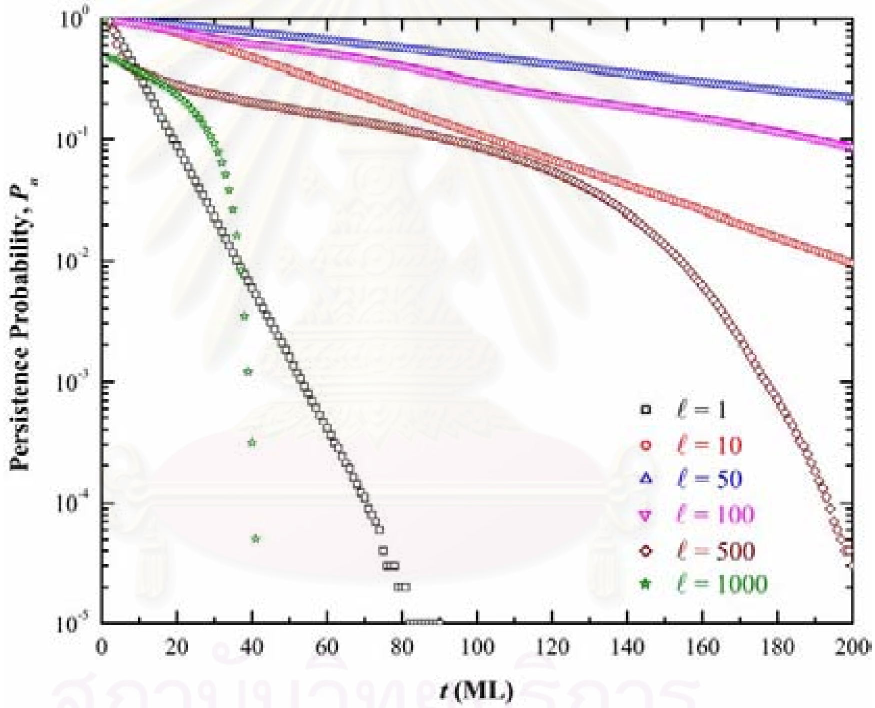

Figure 3.14 The modified persistence probability versus time plot for the periodic patterned growth with $\Delta h = 1$.

Chapter 4 Conclusions

In this project, we have created a computer code for a discrete model to simulate MBE growth process on a substrate with a specified initial condition. The model used here is a well established MBE growth model, the DT model, that has been studied extensively in the flat substrate growth. The model is modified to simulate growth on a substrate with a specific predetermined pattern. Two initial conditions were studied: a flat pattern and a periodic pattern. In the case of periodic pattern, the initial substrate has a series of equally sized block located at an equal interval.

For the flat patterned growth, we found that the film can remain flat when it is grown in layer-by-layer mode. This can be achieved by increasing the mobility of surface atom, i.e. increasing the surface diffusion length of the moving atom. In experiments, the long surface diffusion length is a result of high substrate temperature. The layer-by-layer mode can be identified via an oscillatory interface width. The quantity we used here to determine the pattern survival rate is the persistence probability. We found that the probability decays exponentially in time for the flat patterned growth. The decay is more rapid in a system with a shorter diffusion length (or lower substrate temperature). For longer diffusion length, the persistence probabilities decay slower which indicate that the pattern can survive for a longer period of time. When the diffusion rate is long enough, the persistence probability remains a constant throughout growth time, the film is grown in layer-by-layer mode for the whole time and the surface is totally smooth.

For the periodic pattern substrate, we found that there are two crucial factors that determine how long the pattern can survive. The first one is the ability of the film to stay smooth in the flat parts which are the top terraces of the blocks and the lower terraces between each block. The second factor is the film ability to keep the shape/outline of the pattern, i.e. the width and height of the blocks. Similar technique of increasing surface diffusion length is used here in the periodic patterned case as had been done with the flat patterned study. We found that although the longer diffusion length helps keep the flat parts smooth, just as we found in the flat substrate growth, longer diffusion length destroys the outline of the pattern. Since we need to keep both the smooth parts and the block size, the surface diffusion length needs to be chosen in a compromised manner: not too short and not too long. If the diffusion length is very short, the block size stays approximately the same as the original size but the flat parts turn extremely rough quickly and the persistence probability decreases very quickly. If the diffusion length is too long, the empty space between blocks are filled up with atoms and soon the film becomes flat and the original pattern is totally lost. The optimal value of diffusion length that we found here is a number that is slightly less than half of the block width. So wider blocks are easier to maintained because we can choose a large diffusion length and the flat parts can be maintained for a long time. The is in contrast to a pattern with narrow blocks that we need to use small diffusion length and the flat parts turn into rough surfaces in a short time. We have also found that the original height of the blocks does not effect the way the diffusion length should be selected, but it effects the decay rate of the persistence probability. Taller blocks survive better. So in conclusion, when the original pattern has a bigger scale (both in width and height), it can last longer.

In this work, we have shown that the conventional definition of the persistence probability is not consistent with the obtained morphology. Since only the reproduction of the exact pattern is considered, the persistence probability decays very quickly even when the pattern can still be seen in the grown film. In this work, we introduce a modification to the persistence probability definition in order to count the pattern when it is just slightly off from the exact pattern. We found that the modified version agree better with the quality of the pattern seen on the simulated films.

Finally, we note that our results from both flat patterned and periodic patterned systems with limited mobility ($\ell = 1$ only) agree well with previous work [8-9], i.e. the persistence probability decreases with growth time with a power law relation, leading to a straight line with a negative slope when $P(t)$ is plotted versus time on a log-log scale. However, the pioneer work on patterned substrate growth [8-9] concentrated on statistical behavior of $P(t)$ so the studies [8,9] involve large-scale simulations to investigate asymptotic behavior and scaling relations of $P(t)$ but do not include any noise reduction technique. In our work that concentrate on determining important parameters that control the persistence of the pattern, we utilized the long surface diffusion noise reduction technique in order to make the model more realistic. Our large ℓ results, though cannot be compared with other work in the literature, yield interesting results which we have reliable reasons to support them as already discussed in the previous chapter. The modification of the persistence probability definition in our work was done in response to comments from experimentalists. We believe this is a new result that does not have any published work to compare with as well.

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