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PERSISTENCE IN THIN FILM GROWTH ON PATTERNED SUBSTRATES

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จุดมุ่งหมายในการปลูกฟิล์มบางบนแผ่นรองรับที่มีรูปแบบให้มีคุณภาพสูงคือ ต้องรักษารูปแบบให้อยู่ได้นานตลอดกระบวนการปลูกฟิล์ม แบบจำลองทางคอมพิวเตอร์ได้ถูกนำมาใช้ในการหาเงื่อนไขที่เหมาะสมที่ทำให้รูปแบบคงอยู่ได้เป็นเวลานาน ในที่นี้เราคำนวณปริมาณการคงอยู่ของรูปแบบที่เวลาหนึ่งๆได้จากความน่าจะเป็นของการคงอยู่ เพื่อให้รูปแบบคงอยู่ได้นาน เราได้เพิ่มเทคนิคการลดสิ่งรบกวนในแบบจำลองดั้งเดิม อันได้แก่ เทคนิคมัลติเพิลฮิต (multiple hit technique, $m > 1$) และเทคนิคการเพิ่มระยะการแพร่บนพื้นผิว (long surface diffusion length technique, $\ell > 1$) เพื่อหวังว่ารูปแบบมีการคงอยู่เป็นเวลานาน ในงานวิจัยนี้ เราพบว่าเทคนิคแรกใช้ได้ดีในการปลูกฟิล์มบนแผ่นรองรับที่เรียบ แต่ไม่ดีสำหรับการปลูกฟิล์มบนแผ่นรองรับที่มีรูปแบบ ส่วนเทคนิคที่สอง ระยะการแพร่บนพื้นผิว (ℓ) ที่ไกลขึ้นในการจำลองทางคอมพิวเตอร์นั้นจะเทียบได้กับอุณหภูมิของแผ่นรองรับที่สูงขึ้นในการทดลอง เราพบว่ารูปแบบจะคงอยู่ได้เป็นเวลานานเมื่อ ℓ เพิ่มขึ้น อย่างไรก็ตามก็มีข้อจำกัดของค่า ℓ ที่จะเป็นได้ สุดท้ายเราได้เสนอนิยามของความน่าจะเป็นของการคงอยู่ของรูปแบบใหม่ แทนที่นิยามเดิมที่มีข้อจำกัดมากเกินไป ดังนั้นนิยามเดิมจึงไม่สะท้อนค่าที่เป็นจริงที่เราสังเกตได้จากลักษณะพื้นผิว การศึกษาของเรานี้จะนำไปสู่ข้อสรุปที่ว่า การคงอยู่ได้นานของรูปแบบจะขึ้นกับสองปัจจัย ได้แก่ ความเรียบของส่วนที่ราบในรูปแบบ และรูปร่างเค้าโครงของรูปแบบ

ภาควิชา ฟิสิกส์ ลายมือชื่อนิสิต .....

สาขาวิชา ฟิสิกส์ ลายมือชื่ออาจารย์ที่ปรึกษา.....

ปีการศึกษา 2547

4472471223 :MAJOR PHYSICS
KEY WORDS: MULTIPLE HIT NOISE REDUCTION TECHNIQUE / LONG SURFACE DIFFUSION LENGTH NOISE REDUCTION TECHNIQUE / PERSISTENCE PROBABILITY

SUWAKAN PIANKORANEE : PERSISTENCE IN THIN FILM GROWTH ON PATTERNED SUBSTRATES. THESIS ADVISOR : PATCHA CHATRAPHORN, PH.D., 58 pp. ISBN 974-17-6396-4.

The purpose of high quality thin film growth on patterned substrates is to maintain the pattern during deposition process. A simulation growth model is used here to determine appropriate conditions in which the pattern can remain for a long time. Two noise reduction techniques, *multiple hit* and *long surface diffusion length*, are introduced into the original model. In this work, we find that the former technique, which works well in growth on flat substrates, is not good for patterned substrate growth. The latter, the longer surface diffusion length ℓ in computer simulations, is equivalent to higher substrate temperature in experiments. We find that the pattern persists for longer period of time when ℓ is increased. However, there is a limit to how large ℓ can be. Finally, we suggest a new definition of persistence probability instead of the original one which is too strict and hence does not truly reflect what we observe in the morphology. Our studies lead to a conclusion that the long-lived persistence of the pattern depends on two factors: the smoothness of the flat part in the pattern and the outline shape of the pattern.

Department Physics — Student's signature .....

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Chapter 1

Introduction

The physics of surfaces and interfaces has become increasingly more important in the field of condensed matter. Profound knowledge of surface and interface effects is essential for the understanding of a wide variety of phenomena, many of which belong to the general field of solid state physics. Growing interest in nanostructures and the attempt to obtain an atomic-scale understanding have further emphasized the importance of surface and interface physics.

Modern techniques of controlling crystal surfaces, e.g. etching techniques, can imprint structures down to the atomic scale on a substrate. Then, the substrate can be made to have a specific structure on its surface as seen in Fig. 1.1. We call this substrate a "patterned substrate". In most cases the aim of thin film growth on patterned substrates is to produce high quality films. Typically, "high quality" in thin films means very smooth films since rough surfaces have poor contact properties and cannot be used in most applications. Besides smoothness, the pattern must also be kept after the growth process. Consequently, there is a need to determine suitable parameters that give information about the pattern so appropriate conditions in which the pattern can remain as time evolves during the growth process.

Recently there has been increasing interest in thin films produced by molecular beam epitaxy (MBE) (Siegert and Plischke 1994; Das Sarma and Ghaisas 1992; Wolf and Villain 1990; Das Sarma and Tamborenea 1991; Kim and Das Sarma 1994; Lai and Das Sarma 1991). In this thesis, we study the growth con-

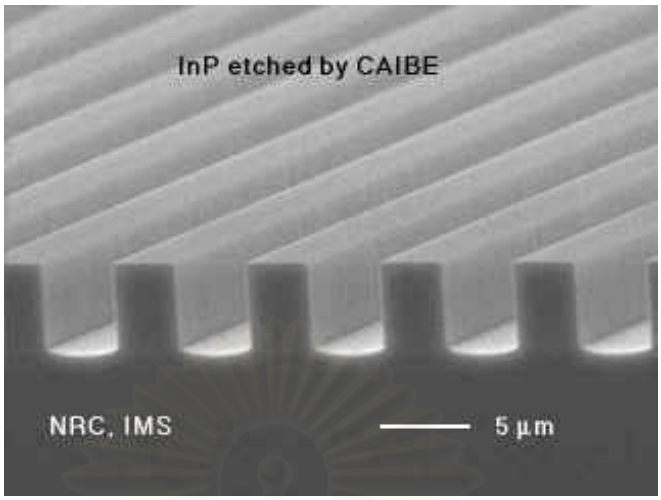


Figure 1.1. InP film was etched on its surface to produce the pattern in atomic scale. (This figure is copied from http://www.oxfordplasma.de/process/inp_etch.htm)

ditions for ideal low temperature MBE without both defect and desorption (evaporation), and there are many discrete growth models (Siegert and Plischke 1994; Das Sarma and Tamborenea 1991; Tamborenea and Das Sarma 1993; Das Sarma and Ghaisas 1992; Wolf and Villain 1990; Kim and Das Sarma 1994) describing this type of growth process. Simulations using these discrete models are carried out according to the given growth rules that govern physical properties of surface growth. Here, the Das Sarma-Tamborenea (DT) model (Das Sarma and Tamborenea 1991; Tamborenea and Das Sarma 1993) is chosen. Generally, the DT model produces very rough thin films because there is an unavoidable stochastic noise. In order to obtain high quality thin films, the noise must be reduced. Two noise reduction techniques, the long surface diffusion length (Chatraphorn and Das Sarma 2002) and the multiple hit (Punyindu and Das Sarma 1998) noise reduction technique, are introduced into the original DT model. According to previous works (Das Sarma and Tamborenea 1991; Wolf and Villain 1990; Das Sarma et al. 1996; Family 1986; Das Sarma and Chatraphorn 1997; Punyindu and Das Sarma 1998), these noise reduction techniques were used to study flat patterned substrate growth; therefore, we expect that these noise reduction techniques will also be suitable for a substrate that is initially not flat. We hope this knowledge

can help us improve the real growth on a patterned substrate.

The outline of the thesis is as follows. In Chapter 2, basic concepts related to crystal growth are discussed. MBE is also briefly introduced in this chapter. Chapter 3 gives a detail of discrete growth models. These include the noise reduction techniques. In Chapter 4, we present simulation results of the study for two types of initial patterned substrates: flat patterned substrate and periodic patterned substrate. The results including the noise reduction techniques are discussed. Finally, a conclusion is offered in Chapter 5.

Chapter 2

Theoretical Aspect

In this chapter the basic concepts of surface growth are introduced. Issues such as deposition, desorption and surface diffusion processes are addressed here. These topics are limited under conditions of molecular beam epitaxy (MBE), which is also introduced in the last section.

2.1 Concept of surface growth

In order to understand the surface growth, we review relevant processes in microscopic atomic picture. There are three basic processes that take place on the interface: *deposition* of an atom onto the surface, *desorption* from the surface, and *surface diffusion*. Note that in this section, we consider the growth on initially flat substrates and treat a real structure of an atom as a simple cubic.

2.1.1 Deposition

Deposition is the process of adatoms (arriving atoms) from the vapor being put on the surface (see Fig. 2.1). A deposition material is thermally evaporated from a reservoir – an atomic source. It forms a beam of neutral atoms or molecules that have thermal velocities. The beam (or vapor) is directed toward the crystal surface. Typically, particles in the beam do not collide or react chemically with each other before they reach the surface. A deposited particle is dropped randomly

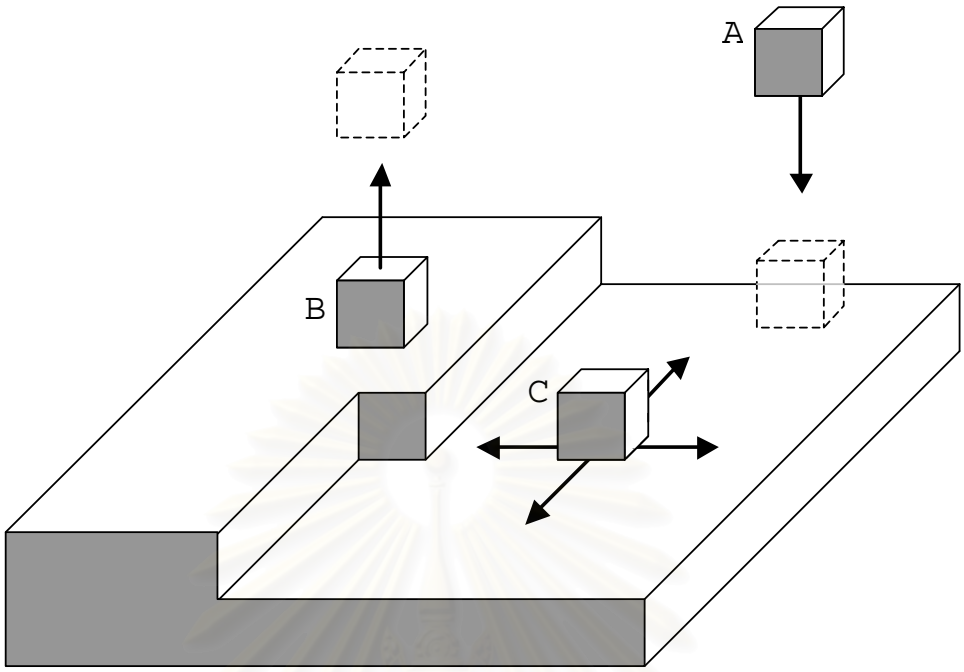


Figure 2.1. Three basic atomic processes during surface growth. atom **A** is *deposited* at a random position on the surface, forming bonds with the surface atoms, and sticks. Some deposited adatoms may *desorp* and leave the surface such as shown in atom **B**. Atom **C** *diffuses* in random directions on the surface to search for an energetically most favorable position.

on the surface site, then it forms bonds with its neighboring particles on the surface and sticks.

2.1.2 Desorption

A process that competes with the deposition is the *desorption*. Desorption is the process in which some deposited adatoms (or molecules) on the surface evaporate from the interface (see Fig. 2.1). The average time that one atom takes from deposition to desorption, is called a *lifetime* of a deposited adatom. The lifetime, τ, was found to depend on the Arrhenius law (Barabási and Stanley 1995)

$$\tau \sim \exp\left(\frac{E_D}{k_B T}\right), \tag{2.1}$$

where E_D is the energy barrier for desorption, T is the substrate temperature, and k_B is the Boltzmann constant.

The lifetime of a deposited adatom corresponds to a desorption rate. The shorter lifetime makes the larger desorption rate. According to Eq. (2.1) the desorption rate increases if the substrate temperature is increased. Moreover, the desorption probability depends on a strength of bonds between the atom on the surface and the crystal surface. The strength of bonds can be expressed in terms of the amount of energy E_D. The adatom must overcome E_D to leave the surface. The strength of the bonds depends on the type of the atom and the local surface geometry of the surface where the atom sticks.

Under ideal molecular beam epitaxy growth condition, the substrate temperature is set not too high. For many material, the desorption rate is very low compared to the deposition rate. Therefore, in our studies we assume that the desorption is negligible.

2.1.3 Surface diffusion

In molecular beam epitaxy growth, deposited adatoms diffuse on crystal surface in random directions to search for the most energetically favorable positions (see Fig. 2.1). When an adatom is deposited randomly on the surface site, it binds with surface atoms. In order to diffuse, the adatom must break the bonds. If the bond is not too strong, it can be broken easily and the adatom can continue to diffuse. The diffusion length can be quite large, and it depends on the substrate temperature and the binding energy.

In a microscopic point of view, a diffusive step of an atom is integral multiples of the crystal lattice. To diffuse to the next lattice position, the adatom must overcome the energy barrier E_0. This barrier exists between two neighboring positions as schematically illustrated in Fig. 2.2.

The diffusion rate or hopping rate is defined as an average number of jumps

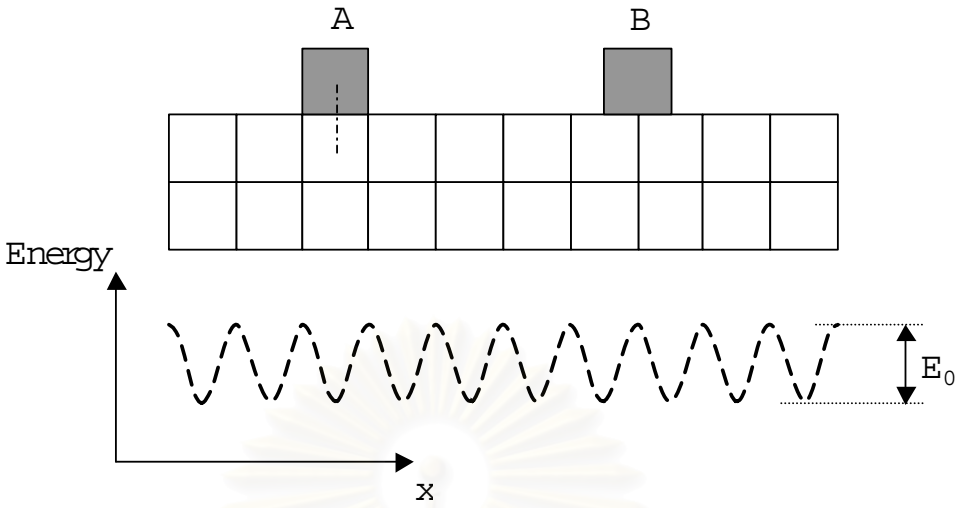


Figure 2.2. Schematically illustration in one dimension of diffusion (Barabási and Stanley 1995). Atom **A** forms one bond with a surface atom. It is in an energetically favorable position. This corresponds to a minimum energy. In order to diffuse, it must move past an unstable position such as shown in atom **B** where is energetically unstable. This position corresponds to a maximum energy. An adatom can diffuse only if it has excess energy E_0.

of an atom in a unit time interval. Diffusion rate, D, of a free surface atom depends exponentially on the substrate temperature and follows the Arrhenius law (Barabási and Stanley 1995):

$$D \sim \exp\left(-\frac{E_0}{k_B T}\right), \tag{2.2}$$

when E_0 is the substrate bonding energy.

The adatom continues to diffuse as long as it is on the surface, until it meets another atom (e.g. an edge of an island or a terrace). As shown in Fig. 2.3, atom **B** and **C** stick to an edge of a terrace, and form additional bonds with atoms at the edge of the terrace. The probability of diffusion is decreased since an adatom must overcome the energy barrier E_0 and must break the additional bonds. In the most simple picture, the only factor affecting the diffusion rate is the number of lateral nearest neighbors . Therefore, Eq. (2.2) becomes

$$D \sim \exp\left[-\frac{(E_0 + nE_N)}{k_B T}\right], \tag{2.3}$$

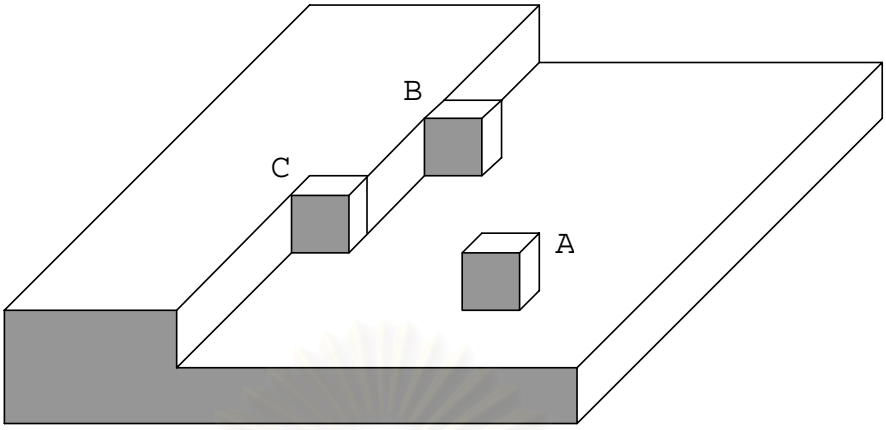


Figure 2.3. Atom **A** can diffuse easiest since it must break only one bond with a surface atom below in order to diffuse. As for atom **B** and **C**, They stick to an edge of a terrace, and hardly diffuse. Because they need more energy to break the substrate bond and the additional bonds (one bond for atom **B** and two bonds for atom **C**).

where n is the number of lateral nearest neighbors and E_N is the nearest neighbor binding energy.

This relation shows that the hopping rate is exponentially suppressed by n. An adatom finds the edge of a terrace or island and thus it remains there for a long time. The diffusion rate also depends exponentially on the substrate temperature. At low T, the adatom remains at its deposition site. However, when the substrate temperature is increased, the bonds can be broken more easily and the adatom can diffuse.

2.2 Molecular beam epitaxy growth

Epitaxial growth is a nonequilibrium process which a new layer on a surface is arranged to form the same crystal structure as the substrate. One technique to achieve such growth that we concentrate in this study is *molecular beam epitaxy* (MBE).

MBE technique is used in the fabrication of high quality films, e.g. optoelectronic devices, quantum structures, etc. There are several sources with varying incoming flux rates that can be used to grow complex compounds with precise elemental compositions. In this technique, the neutral atoms or molecules are thermally evaporated from a source and form a beam that have thermal velocities. This beam (or vapor) is directed toward a heated substrate with a slow growth rate. In general, the film is grown one monolayer at a time. A monolayer means average film growth fills up one layer. MBE experiments are usually performed under ultra-high vacuum conditions (pressures smaller than 10^{-10} torr) to minimize impurities from unwanted gases such as H_2, CO_2, CO and H_2O, which affect the quality of the growing film.

There are three main mechanisms that affect surface growth: deposition, desorption, and surface diffusion. In MBE growth, desorption is usually negligible when compared to the deposition rate. Therefore, studying MBE growth is focused on a competition between deposition and surface diffusion. Surface diffusion depends on the magnitude of the binding energy and the substrate temperature. The binding energy cannot be modified in experiment unless the substrate is changed. So the parameters that can be controlled experimentally are the deposition rate and the substrate temperature T. In this study, the film is grown with slow deposition rate of 1 monolayer per second. Below we consider the surface of an arbitrary film that occurred by adjusting the substrate temperature:

1. At relatively high T (within the constraint that desorption from the growth front should be negligible, so T cannot be arbitrarily high), the surface

diffusion length is very large comparing with the size of an existing island or an existing terrace. Thus an adatom can diffuse to find the edge of the island or the terrace, where it is an energetically favorable position. In this case the new layer is not formed until the previous one is completed. This growth occurs via the two dimensional *layer-by-layer* mechanism. Films from this growth have smooth surface.

2. At relatively low T, the surface diffusion length is decreased and becomes smaller than the size of an existing island or an existing terrace. An adatom can still diffuse but may not be able to go far enough to find an edge of an island or a terrace because the mobility of adatoms is low. adatoms will meet and nucleate a new island on top of the surface. This makes the surface becomes kinetically rough. The growth in this case is called *three-dimensional* growth.

3. At very low T, the surface diffusion length is very small. It is actually smaller than the lattice spacing. In this case, only the deposition process affects the surface growth. The deposited adatom cannot diffuse and they stick at their random deposited site. Thus, the film has a very rough noisy surface.

Experimentally, the morphology (the form or shape of a surface in macroscopic point of view) of the surface can be observed by the *Reflection High Energy Electron Diffraction* (RHEED). Under layer-by-layer growth on a flat substrate, the oscillating behavior of the diffraction intensity is observed as shown in Fig. 2.4. The period of these oscillations corresponds to the time that is used to form each complete monolayer. Figure 2.5 shows that the intensity is at the maximum when a film fills up one monolayer completely. As new atoms arrive on the new layer, the intensity decreases. It is at its minimum when the film grows a half of a layer. And then the intensity increases again when the film begins to form a new complete monolayer. The amplitude of the oscillations is damped as growth proceed, but the period of the oscillation remains constant.

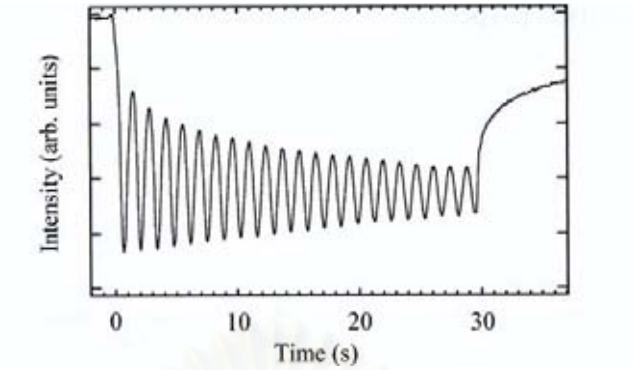


Figure 2.4. Schematic illustration shows the layer-by-layer growth oscillation observed by reflection high-energy electron diffraction (RHEED) (Braun 1999).

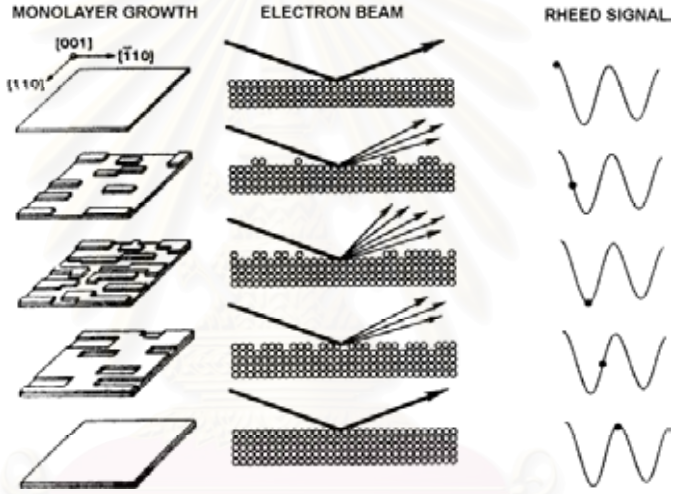


Figure 2.5. The formation of a single complete monolayer corresponding RHEED oscillation signal is shown (Elshabini-Raid and Barlow 1998).

The substrate has a strong influence on the growth in MBE technique. If both film and substrate are the same material, the lattice parameters are perfectly matched and there is no interfacial bonding strain. The process which the film and the substrate are identical is called *homoepitaxy*. On the other hand, when films and substrates are composed of different kind of materials, the growth is called *heteroepitaxy*. The focus of this study is on the former – the homoepitaxial growth.

Chapter 3

Growth Simulations

3.1 Discrete growth models

Discrete growth models in computer simulations are used to study complicated behaviors of atomistic epitaxial growth. Experimentally, we cannot control some effects such as desorption, defects, etc., but in computer simulations, we can choose to study only a few particular mechanism at a time. This is clearly the advantage of discrete growth models. In this work, we study effects of the surface diffusion process to the growth surface. There are many discrete growth models, e.g. MBE model (Das Sarma et al. 1996; Lanczycki 1995), Das Sarma-Tamborenea (DT) (Das Sarma and Tamborenea 1991; Tamborenea and Das Sarma 1993), Wolf-Villian (WV) (Wolf and Villain 1990). Each model has its own restriction and diffusion rule that govern the physical properties of the surface growth. In this work, all our simulations are carried out using the DT model. However, we also describe the MBE model for comparison and to point out why we choose the DT model.

3.1.1 MBE model

In molecular beam epitaxial (MBE) growth, the diffusion process on a growing surface is the dominant smoothening mechanism and noise fluctuation inherent in the deposition beam is the roughening mechanism. As discussed in Chapter 2, all

surface adatoms (not just the most recently deposited atom) can hop at any time (not only at the time of its deposition) with the Arrhenius hopping rate. This rate is an exponential function of the activation energy and the substrate temperature T. The activation energy of a diffusing adatom depends on bonds formed by the adatom with its neighboring atoms. The most realistic simulation for this study is the MBE model.

In the MBE model, both deposition and diffusion are taken to be stochastic processes. They are simulated by various random number generators. Therefore, the diffusion process becomes more complicated since there are more than one mobile atom on the growing surface. Sometimes this model is referred to as full temperature dependent activated diffusion MBE growth model in order to differentiate it from limited mobility growth models which are our main interest in this work.

The diffusion process of the MBE model depends on the activation energy according to the Arrhenius law, hence, simulations of such model require a lot of computational time and cannot be carried out for large systems. These reasons make the model very difficult.

3.1.2 Das Sarma-Tamborenea model

In order to study large systems, Das Sarma and Tamborenea introduced DT model (Das Sarma and Tamborenea 1991; Tamborenea and Das Sarma 1993), an extremely simple instantaneous relaxation limited mobility conserved discrete growth model of ideal MBE growth under random vapor deposition nonequilibrium growth conditions. This model came from the observation of one dimensional ($d = 1$) MBE growth. Note that by"one dimensional", we mean that the growth process is being done on a one dimensional substrate. The convention notation for this is $d = 1$. However, DT model has two crucial differences from the one dimensional MBE growth model: (1) there is only one rate, namely, the depo-

sition rate (and no diffusion or hopping rate), and (2) each atom is allowed to diffuse only once and not continuously. The model is also under solid-on-solid (SOS) constraint, i.e. overhangs, bulk vacancies and desorption are not allowed. This constraint makes the model conserved since the volume of the growing film equals the volume of the incoming flux. DT model is easier because only the most recently deposited adatom can move. The moving atom will move according to diffusion rule of DT model instantaneously to find its final site within finite mobility length. (So the model is called limited mobility model). According to previous works, DT model was studied (Das Sarma and Tamborenea 1991; Das Sarma et al. 1996; Das Sarma and Chatraphorn 1997; Punyindu and Das Sarma 1998) extensively in thin film growth on flat substrates. It was shown that this model can explain MBE growth reasonably well. Moreover, this growth model is the low to intermediate substrate temperature version (Das Sarma and Tamborenea 1991; Tamborenea and Das Sarma 1993) of the full temperature dependent activated diffusion MBE growth model (Tamborenea and Das Sarma 1993; Das Sarma et al. 1996) because morphologies and scaling exponents[†] of DT model agree quantitatively with MBE model in this temperature range. However, MBE model can be used for both homoepitaxial and heteroepitaxial growth while DT model can be used only in homoepitaxial growth studies.

In order to simplify for growth simulations, a simple cubic lattice is used instead of the real structure of the solid. Usually, discrete atomic positions are determined from integral multiples of the lattice constant. Here, we set the lattice constant to be one. The deposition rate F (growth rate) is fixed as one monolayer (ML) per second. It means that in one second, the film is filled up with an average of one layer with $N = L^d$ atoms, where L is substrate size, and d is substrate dimension in $(d + 1)$-dimensional space (1 is referred to film growth dimension which is perpendicular to the substrate). In this work, we measure time in unit of

[†]The set of scaling exponents α, β and $z = \alpha/\beta$ for a discrete growth process defines a dynamic universality class which characterizes asymptotic properties of kinetic surface roughening associated with that discrete growth process.

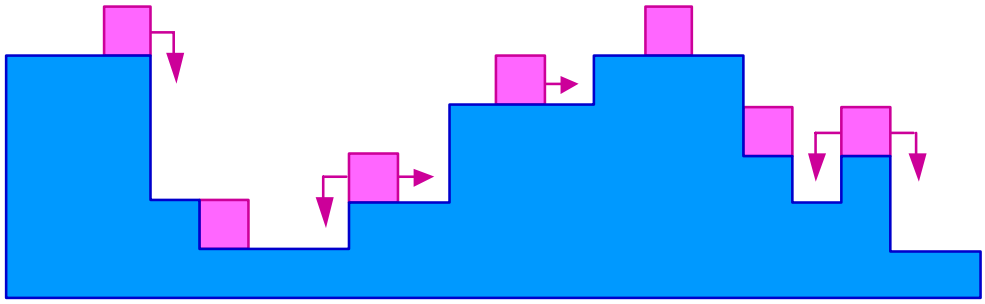


Figure 3.1. The diffusion rule for DT model with $\ell = 1$ in one dimensional substrate.

monolayer. This will be described in more detail in the next chapter. Moreover, all of our simulations are done with periodic boundary conditions along the substrate. Hence, the topology of the substrate in one dimensional substrate is a circle.

In the DT model, at every time step deposited adatoms are dropped vertically, randomly and sequentially (one atom at a time at a randomly chosen spatial position) on a d-dimensional substrate. After the deposition, each adatom is allowed to move immediately only once within finite lateral surface diffusion length to its final incorporation site according to the diffusion rule. After the adatom chooses its final site, it is permanently incorporated at that site and cannot move for the rest of the growth time. Note that the surface diffusion length ℓ is measured in lattice units. If $\ell = 0$, we got the *random deposition* (RD) model where deposited adatoms cannot diffuse at all and will be incorporated at their random deposition sites. This model corresponds to MBE growth at extremely low substrate temperature. In that situation, deposited adatoms have insufficient thermal energy in order to diffuse; hence they stick at their deposition sites. The RD model is the simplest atomistic growth model in computer simulations because we randomly choose a position of a deposited adatom and add its height by one.

For original DT model ($\ell = 1$), deposited adatoms are dropped on the substrate and move following DT diffusion rule (see Fig. 3.1). A deposited adatom is allowed to move only if it has no lateral nearest neighbor in the same layer (i.e., the deposited adatom has a nearest neighboring bond with the atom underneath it that is necessary to satisfy the SOS constraint). If a deposited adatom has more

than one nearest neighbor bond at its deposition site, it will be sticked at that place. For a deposited adatom which has one nearest neighbor bond, it diffuses instantaneously by one site within surface diffusion length to its final position. At this position, the adatom must have more local coordinate numbers than at the deposition site. This is equivalent to increasing the number of nearest neighbor bonds it forms with other atoms. If there are many possible final sites that satisfy the requirement to increase the number of bonds compared with at the deposition site, the deposited adatom chooses one of those sites with equal probability. If there is no other site within the surface diffusion length that satisfies the diffusion rule, then a deposited adatom is incorporated at its deposited site. When adatoms are incorporated, they cannot diffuse again.

It is important to emphasize that deposited adatoms in DT model search for final sites with higher coordination numbers compared to the original deposition site. The final sites are not necessarily the local sites with maximum coordination numbers. In other words, adatoms try to *increase*, but *not necessarily maximize*, the local coordination number. DT diffusion rule also allows only downward diffusion of the adatoms (can move on the same layer or move down) but not necessarily to height minima as follow gravitational rule – because the electronic force is more dominant than the gravitational force. For this reason, the step roughening is occurred on the surface.

Before concluding this section, it should be explained that the similarity between the DT results and the low temperature MBE results does not come as a surprise. The fact is, the diffusion rule for the DT model is set following extensive studies of the low temperature MBE model. In low temperature MBE simulations, it was found that eventhrough all surface atoms are allowed to move, most of the times atoms without lateral bonding are the one that move (Das Sarma and Tamborenea 1991). So the simple diffusion rule of the DT model was created with the intention to mimic behavior of atoms in low temperature MBE model from the beginning.

3.2 Noise reduction techniques

In DT growth simulation, there is an unavoidable stochastic noise during the growth processes. The noise cause the kinetic roughness on the surface. This shot noise comes from the fact that adatoms are dropped randomly on deposition sites. Moreover, there is a noise associated with the stochastic diffusion process. The adatom will select randomly one of such possible sites, if there are many possible final sites that satisfy DT diffusion rule. This randomness produces the noise in the simulation as well. The noise associated with the stochastic diffusion also contributes to kinetic roughness, but the shot noise associated with the incident beam fluctuations is more dominant roughening mechanism. In practice, the rough surface is not required. To obtain smooth MBE growth characterized by *layer-by-layer* growth, where each layer of the growing film is essentially filled up completely before the next layer deposition begins, we need to reduce the noise and enhance diffusion. Two noise reduction techniques (NRTs) are introduced in the growth simulation: *long surface diffusion length* (Chatraphorn and Das Sarma 2002) NRT and *multiple hit* (Punyindu and Das Sarma 1998) NRTs. By these techniques, the diffusion rules of DT model are still the same as original DT model.

3.2.1 Long surface diffusion length

The long surface diffusion length NRT is to increase the surface diffusion length, i.e., to increase the maximum lateral length that the adatoms can move. This technique is obvious to improve the smoothness of the growth surface. The adatoms with longer diffusion length will have more chances to incorporate into the most appropriate site (e.g. the site that has two or more nearest neighbors). As an example, Fig. 3.2(a) shows a morphology of $\ell = 1$ limited mobility growth model. We see that it does *not* exhibit any layer-by-layer epitaxy since the deposited adatoms with $\ell = 1$ can move only one step so that many layers are partially filled and the surface roughness is increased. If we increase the surface diffusion length

($\ell > 1$) as shown in Fig. 3.2(b), we see that the deposited adatoms can move far enough to form the complete layer (within the surface diffusion length) and so the layer-by-layer growth is obtained.

Although the long surface diffusion length is a technique in computer simulation, it relates to the substrate temperature T in experiment. The surface diffusion length ℓ depends on growth conditions, i.e., the deposition rate F and the diffusion rate D, as a power law function (Ghaisas and Das Sarma 1992; Das Sarma 1997),

$$\ell \sim \left(\frac{D}{F}\right)^{\gamma}. \tag{3.1}$$

The exponent γ ($\sim \frac{1}{6} - \frac{1}{2}$) depends explicitly on the substrate temperature dependent minimum stable island size and can be calculated by stochastic Monte Carlo simulations or kinetic rate-theoretic arguments. The diffusion rate depends exponentially on the substrate temperature via the Arrhenius law as shown in Eq. (2.2) [‡]. From Eq. (3.1), Eq. (2.2) and setting $F = 1$ monolayer per second, we get

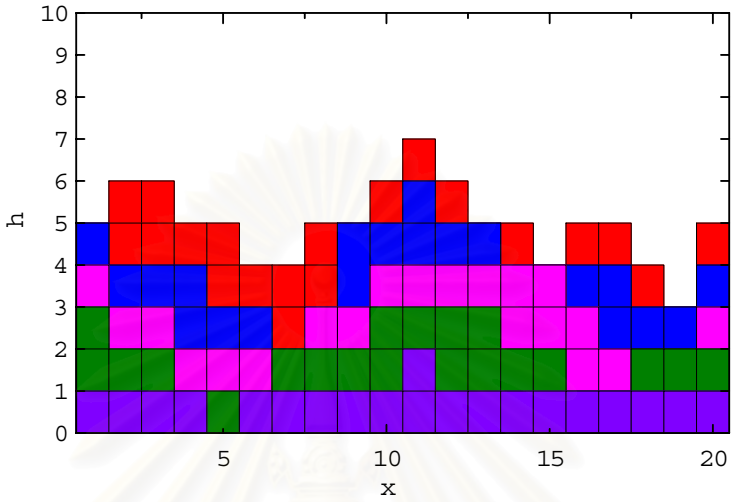
$$\ell \sim \exp\left(-\frac{\gamma E_0}{k_B T}\right), \tag{3.2}$$

According to Eq. (3.2), we see that ℓ depends explicitly on T and conclude that the film has a smooth surface when the film is grown at sufficiently high substrate temperature. Note that the substrate temperature cannot be too high because our model is assumed to have the constraint that desorption from the growth front should be negligible.

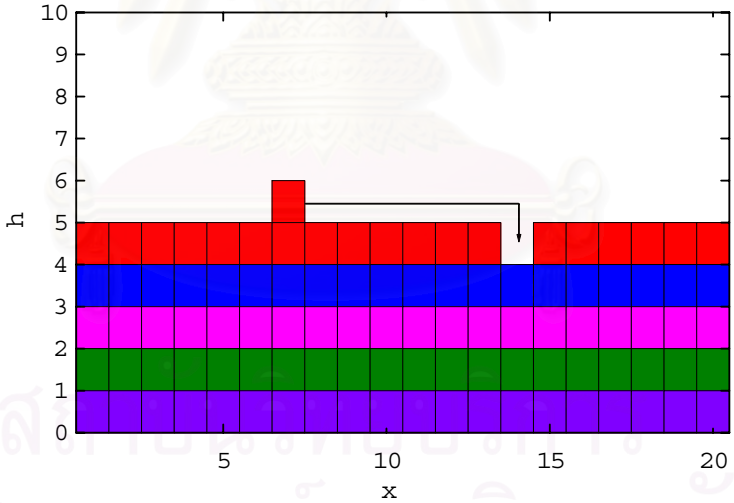
3.2.2 Multiple hit noise reduction

The multiple hit NRT involves the acceptance of only a fraction of the allowed atomistic deposition events in simulations (Punyindu and Das Sarma 1998). This technique is characterized by multiple hit factor m, which is chosen as an integer

[‡]We choose this equation instead of Eq. (2.3) because in the DT model only the free surface adatom without any lateral bonding are allowed to move.



(a)



(b)

Figure 3.2. Schematic plots showing morphologies of substrate size $L = 20$. (a) For $\ell = 1$. (b) For $\ell = 10$.

number. Each surface site has its own counter, C_i. When deposition event occurs and then adatom moves to its final preferred site i according to DT diffusion rule, the counter C_i is increased by one (see Fig. 3.3). In original DT model with $m = 1$, the height h_i of the preferred site is increased by one. However, in DT model with multiple hit NRT, its diffusion rule is slightly modified in such a way that height of the preferred site h_i remains the same and the counter of that preferred site is the quantity that is increased instead. The deposition event at a particular site is accepted and h_i is increased only when a counter C_i reaches a multiple hit factor $m > 1$. Therefore, this technique is called multiple hit NRT since the deposition event becomes a true deposition process only if that site is hit m times. After the true deposition, the counter at that particular site is reset to zero, and the whole multiple hit process is repeated.

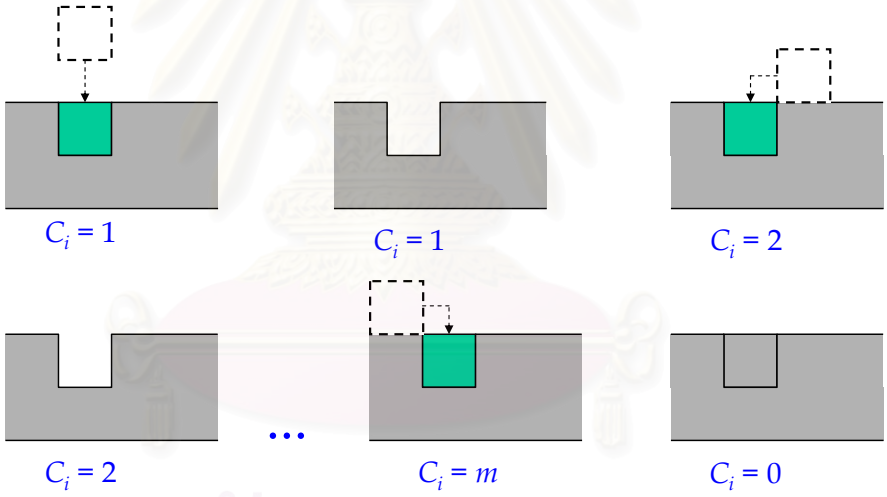


Figure 3.3. Schematic plots showing the multiple hit process.

Although the multiple hit NRT is a pure computational simulation, it works in a similar way as the long surface diffusion length in some aspects. However, we found that in higher substrate dimension, the latter is difficult and extremely time consuming. So that in some works, it is more appropriate to using the multiple hit NRT.

Chapter 4

Simulation Results

The main purpose of this thesis is to determine optimized conditions for thin film growth. In this chapter we present simulation results of two different types of initial patterned substrates: completely flat patterned substrates and periodic patterned substrates, characterized by size of a periodic pattern. The *flat patterned* substrate, on the one hand means no pattern or a perfectly smooth substrate. On the other hand, it can be thought of as a periodic patterned substrate with an infinitely large size of a periodic pattern. For the *periodic patterned* substrate, our simulation results show that a pattern persistence depends on pattern size. This leads to the condition for optimized pattern survival. The model presented here is not meant to describe any particular system in detail, but to give a general idea of underlying mechanisms affecting a characteristic feature size on a patterned substrate growth. In the following, persistence of a pattern will be discussed for the simplest case that diffusion across a step edge from an upper to a lower terrace is not constrained by an Ehrlish-Schwoebel barrier. In addition, the system we study here is homoepitaxial growth which means elastic strain between substrate and film is negligible. Note that all simulations studied here were done with DT model on (1+1)-dimensional substrate of size 10 000 (to aviod the finite size effect).

4.1 Growth on flat patterned substrates

Growth on flat substrate means that the growth process starts at initial height $h(x, 0) = 0$ for all sites $x = 1, \ldots, L$. In our work, flat patterned substrate growth is studied through surface morphologies, interface width W, and persistence probability P of the grown films. The first two quantities are used to describe the smoothness/roughness of the surface. The surface morphology can be seen clearly from the growing film, but it does not give sufficient information. Therefore a new indicator, the interface width (W), is introduced. The last quantity, the persistence probability (P), is used to determine how long the film pattern can survive through time. Below, these issues are discussed in more detail.

4.1.1 Surface morphology

In order to study the growth on flat patterned substrate, the smoothness/roughness of the surface is very important. For this reason, we first consider morphologies of films. Fig. 4.1 illustrates how a typical surface morphology for the original DT model (diffusion length $\ell = 1$ and multiple hit factor $m = 1$) evolves as the deposited time t increases from 10 to 1000 MLs. We see that the dynamical morphology is more and more rough at larger time. In this case, an adatom only has the ability to diffuse to its nearest neighbors so it cannot search for the most appropriate site. As a result, the roughness grows in time. Typically, we do not want a rough film. In order to get rid of the roughness, two noise reduction techniques (NRT) introduced in the previous chapter are used: the long surface diffusion and the multiple hit NRTs.

Fig. 4.2(a) and Fig. 4.2(b) represent surface morphologies in the same deposited time $t = 100$ MLs for various values of surface diffusion length ℓ and multiple hit factor m respectively. It is clearly visible from the figures that the surface morphologies are smoother as the value of ℓ or m is increased. The reason for this is that, in both cases, the adatom has more chance to diffuse to its suitable

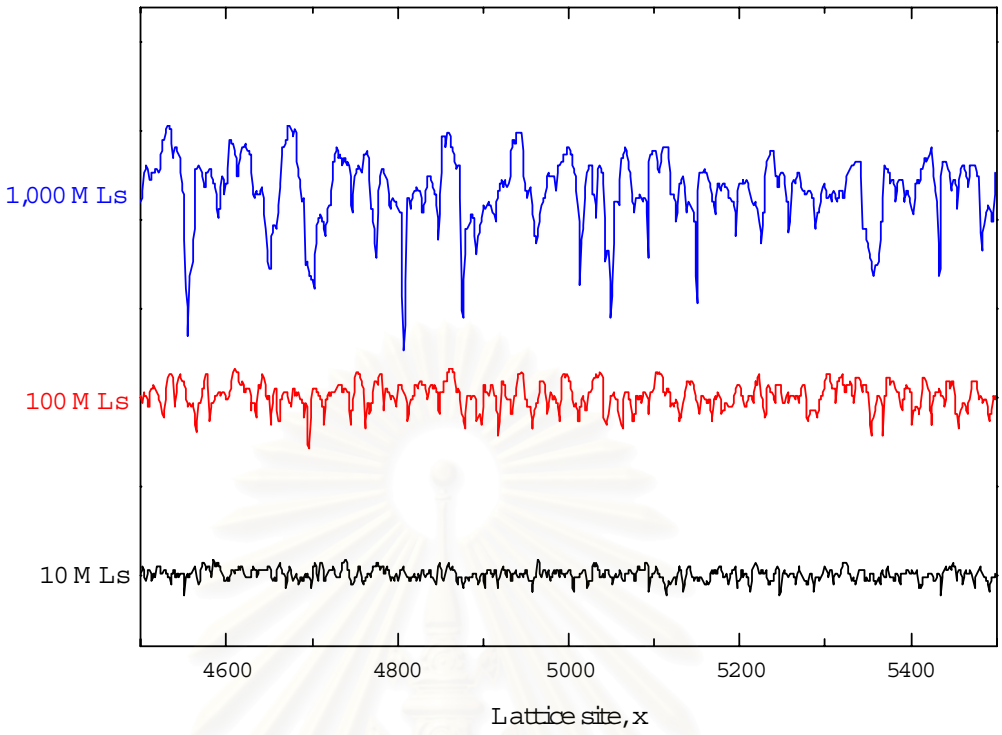
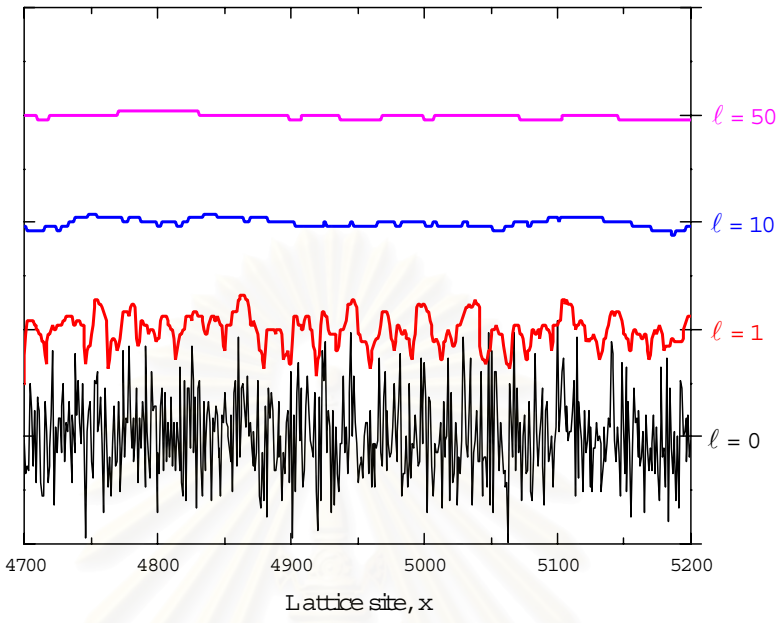
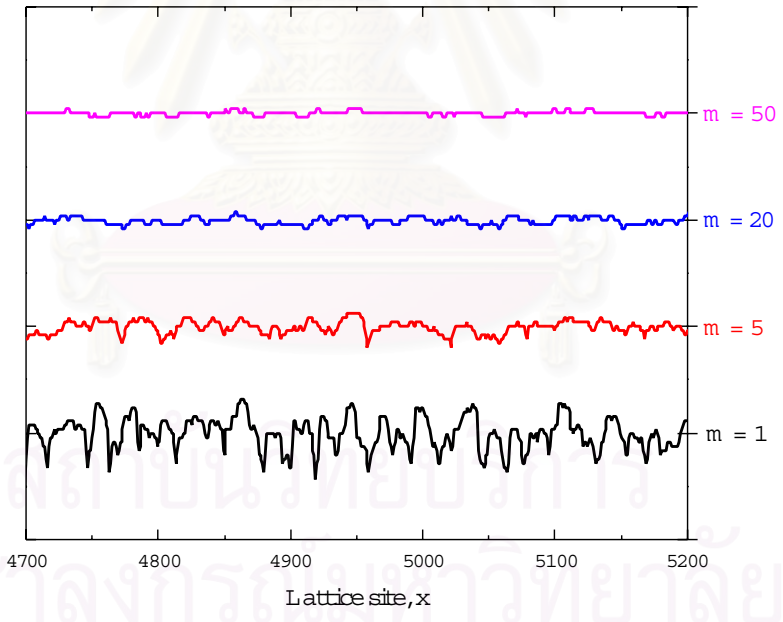


Figure 4.1. Dynamical morphology shows evolution of the kinetically rough interface at $t = 10$, 100 and 1000 MLs for the original DT model, cutting from a middle section of the substrate.

site, and consequently high surface steps and deep grooves shown in the original DT model in Fig. 4.1 are reduced. It is also evident that the morphologies in Fig. 4.2(a) and Fig. 4.2(b) are equivalent – in other words, the morphology of the DT model with long surface diffusion length NRT is similar to the morphology of the DT model with multiple hit NRT.



(a)



(b)

Figure 4.2. Morphologies at $t = 100$ MLs (a) for RD model ($\ell = 0$), original DT model ($\ell = 1$), and DT model with long surface diffusion length NRT ($\ell > 1$, $m = 1$), and (b) for original DT model and DT model with multiple hit NRT ($\ell = 1$, $m > 1$).

4.1.2 Interface width

Although the smoothness/roughness of the growing surface is clearly seen from the surface morphology, to obtain more detailed information of the evolution of the film growth, fluctuations in the film height through time t must be considered. Here, we use the dynamic interface width W as an indicating parameter. The interface width is defined as the root mean square fluctuation in height (Barabási and Stanley 1995):

$$W(t) = \left\langle \left(h - \bar{h}\right)^2 \right\rangle^{1/2}, \tag{4.1}$$

where $h = h(x,t)$ is height of a interface at the lateral position x at time t, $\bar{h}$ is the average film thickness and the angular brackets represent the average over substrate site L. We note that the average film thickness is $\bar{h} = Ft$ [†] where F is growth rate.

Fig. 4.3 illustrates $W(t)$ plots for RD and original DT model. As mentioned before, the growing surface is rougher at longer time so W should go up as time is increased. In both cases, $W(t)$ increases monotonically in the growth time t. It means that as time runs up, there is a wilder fluctuation around the average height, or the morphology is rougher at larger time. Thus, this growth is the kinetically rough growth mode.

The interface width plot with applying NRTs is shown in Fig. 4.4 for the long surface diffusion length NRT, and in Fig. 4.5 for the multiple hit NRT. In the case of the long surface diffusion length NRT, oscillation of $W(t)$ manifests at $\ell > 1$. These oscillations in $W(t)$ correspond to the layer-by-layer growth mode (Chatraphorn and Das Sarma 2002). The interface width W reaches its peak when a film is filled by a half layer. As new adatoms are dropped and grown on this half layer, the $W(t)$ decreases and reaches its lowest when the film grows to a complete layer. Then $W(t)$ increases to the highest point again. These layer-by-layer oscillations of the interface width are in agreement with the RHEED

[†]In this work, we define $F = 1$ and then $\bar{h} = t$; therefore, we can measure time through an average film thickness in unit of monolayer (ML).

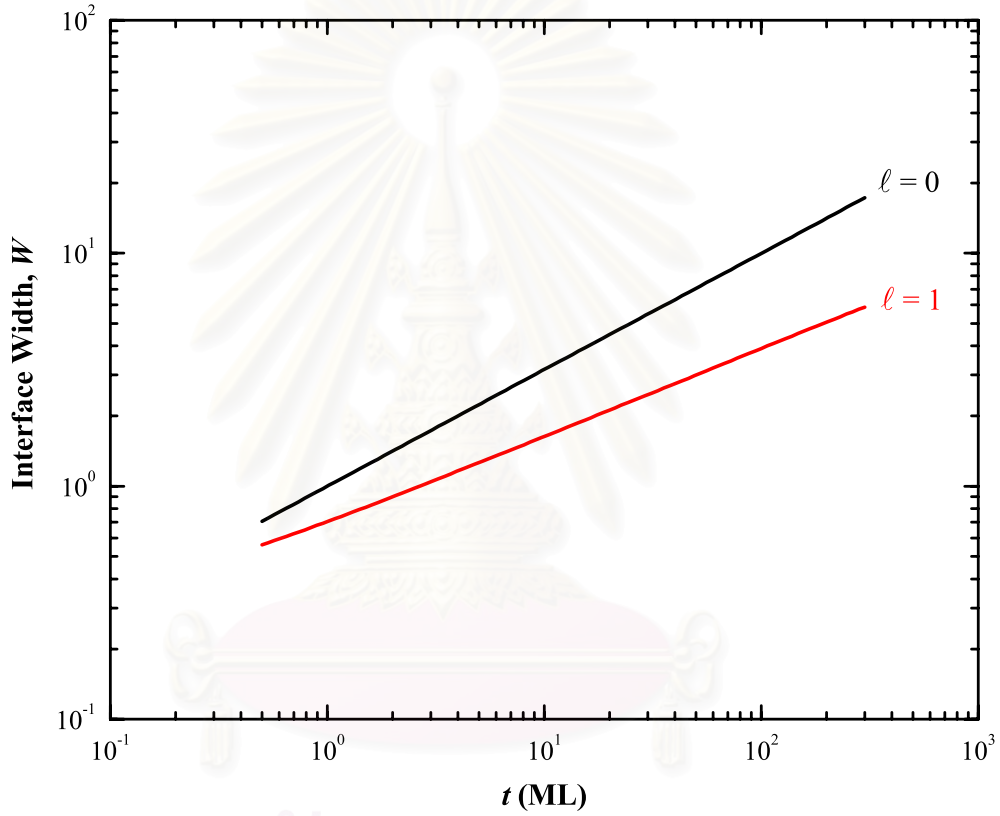


Figure 4.3. The interface width W plots as a function of growth time t on a log-log scale for the RD and the original DT model.

intensity oscillations at high substrate temperature T from various experimental studies (Braun 1999).

For long time process, amplitude of $W(t)$ oscillation decreases continuously until the oscillations disappear. Instead layer-by-layer growth, kinetic roughening is observed. So this growth is a finite size (both spatially and temporally) transient phenomenon. The amplitude of the $W(t)$ oscillation decreases gradually until it disappears and the layer-by-layer growth becomes the kinetically rough. At an early time, the surface adatom diffusion plays a major role in surface growth. Therefore deposited adatoms can move to search for their best sites to be incorporated there and the surface is grown in layer-by-layer growth mode. At long enough time scales (and for large enough lateral system sizes, to prevent saturation), the shot noise intrinsic in the incident deposition beam fluctuations will win out to damp the layer-by-layer oscillations. The multilayered kinetically rough growth will emerge. The length of the initial transient regime depends on the surface diffusion length ℓ, which is equivalent to the growth temperature (substrate temperature T in experiment).

For the multiple hit NRT (Fig. 4.5), the interface width $W(t)$ also oscillates for longer time when m is larger; hence, we can simulate the thin film growth in the layer-by-layer growth mode by using the multiple hit NRT. Comparison between these two figures, The oscillations from Fig. 4.4 do not differ notably from Fig. 4.5. That means the growing surface becomes smoother when value of either ℓ or m is larger. From this, we can conclude that the reduction of noise in simulation, or the increase of substrate temperature T in experiment, is the key to success in growth on flat substrate.

Furthermore, since $W(t)$ from the DT model with multiple hit NRT exhibits qualities that are very similar to the DT model with the long surface diffusion length NRT described above, we can replace the long surface diffusion length NRT with the multiple hit NRT in order to save computational time in higher substrate dimensions.

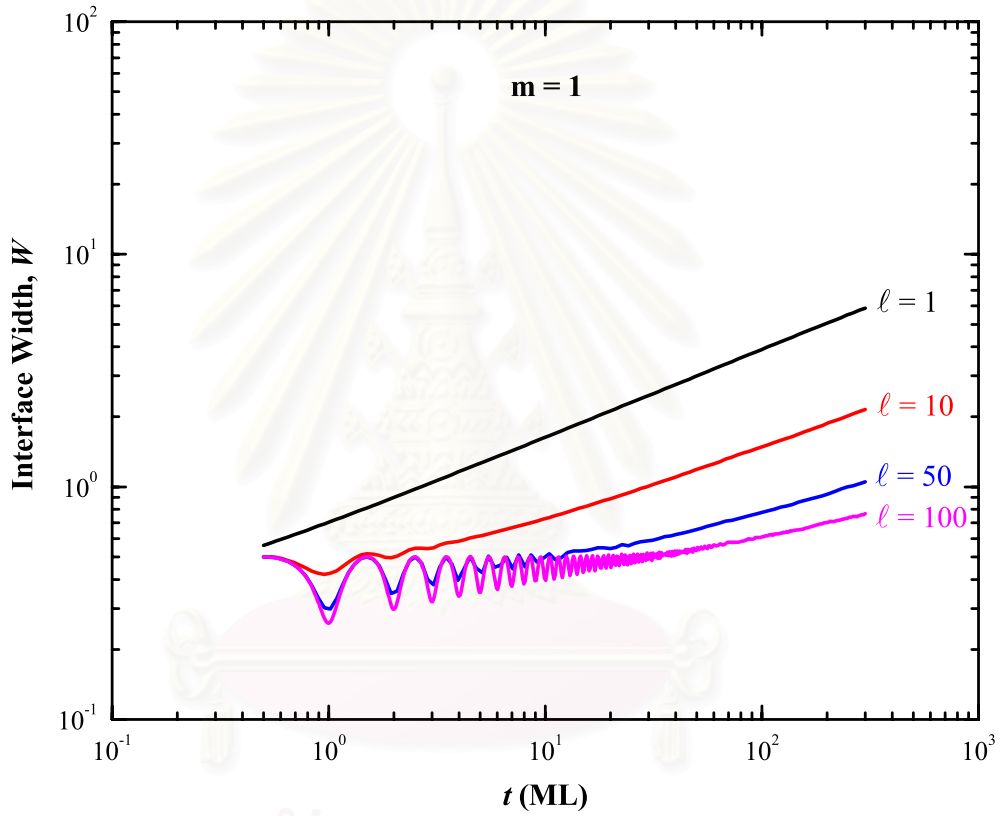


Figure 4.4. The interface width $W(t)$ plots against the deposited time t for various values of ℓ.

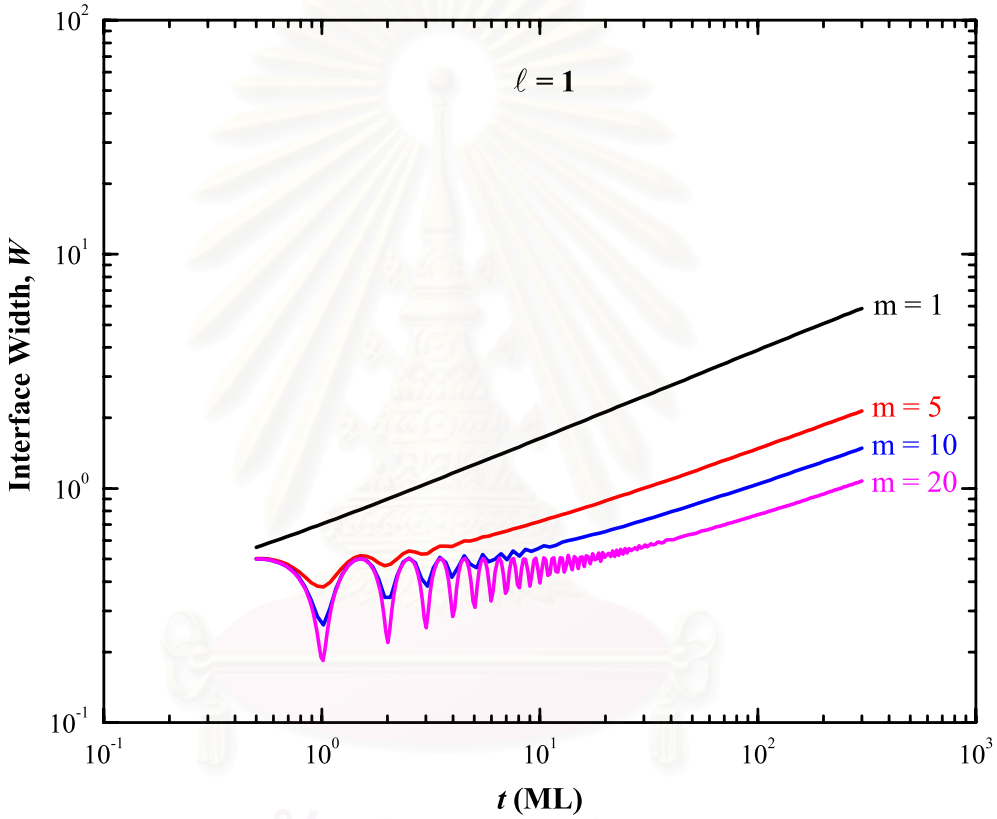


Figure 4.5. The interface width $W(t)$ plots against the deposited time t for various values of m.

4.1.3 Persistence of film pattern

In layer-by-layer growth mode, reproduction of an initially flat pattern on a substrate is nearly perfect. However, the thin film pattern cannot keep its original pattern for a long time since layer-by-layer growth is a finite size transient phenomenon. The question is how long and how much the pattern can persist as time evolves during the growth. In order to answer this, the persistence probability P is defined. This quantity is used to determine the fraction of the pattern that propagate through time t MLs. It is defined as (Kallabis and Wolf 1997)

$$P(t) \equiv \left\langle \prod_{s=1}^{t} \delta_{h(x,s),h(x,0)+s} \right\rangle_L \tag{4.2}$$

where $h(x,s)$ is height of the growing surface at time s at the lattice site x, δ is the Kronecker delta function which $\delta_{i,j} = 1$ when $i = j$ and 0 otherwise, $\prod$ denote a product of δ over time t, and the angular brackets $\langle \ldots \rangle$ represent the average over the whole substrate site L.

By definition, $P(t)$ means survival of the pattern through time t is counted only when the initial patterned configuration, $h(x,0)$, is reproduced every time after each monolayer deposition until the film is grown to t MLs. As mentioned before, the thin film cannot maintain its original pattern for a long time, so $P(t)$ will eventually decrease. Figure 4.6 shows a plot of persistence probability $P(t)$ as a function of time t for the original DT model (ℓ and $m = 1$). The calculated $P(t)$ from this plot decays rapidly which means the pattern disappears quickly.

In order to keep the pattern for long time, the NRTs are used. Having applied the NRTs, we expect to obtain similar results. Figure 4.7 and Figure 4.8 illustrate how $P(t)$ evolves for the long surface diffusion length NRT and the multiple hit NRT respectively. As the value of ℓ or m is increased, $P(t)$ gives smaller slope, indicating better persistence. This is due to the fact that the thin film grows in layer-by-layer mode for long time at large value of ℓ or m. This is to say that the pattern can survive forever if $\ell \to \infty$ or $m \to \infty$.

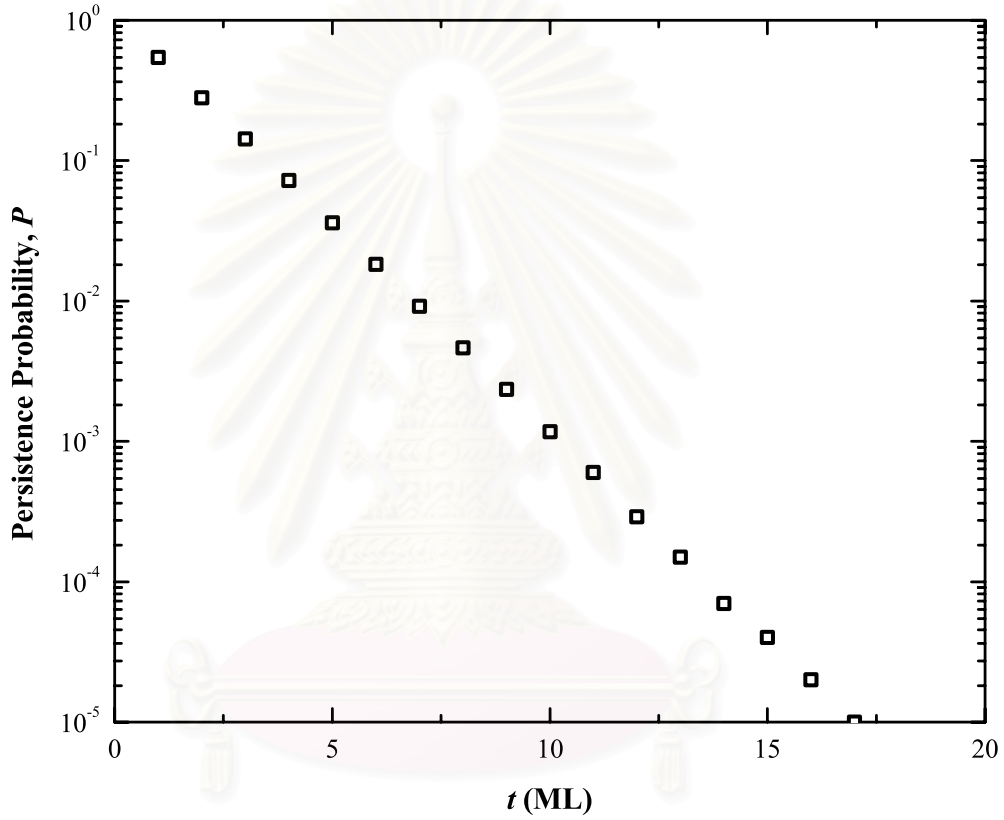


Figure 4.6. The persistence probability P plots as a function of deposition time t from a (1+1)-dimensional flat patterned substrate for the original DT model, i.e., $\ell = 1$ and $m = 1$.

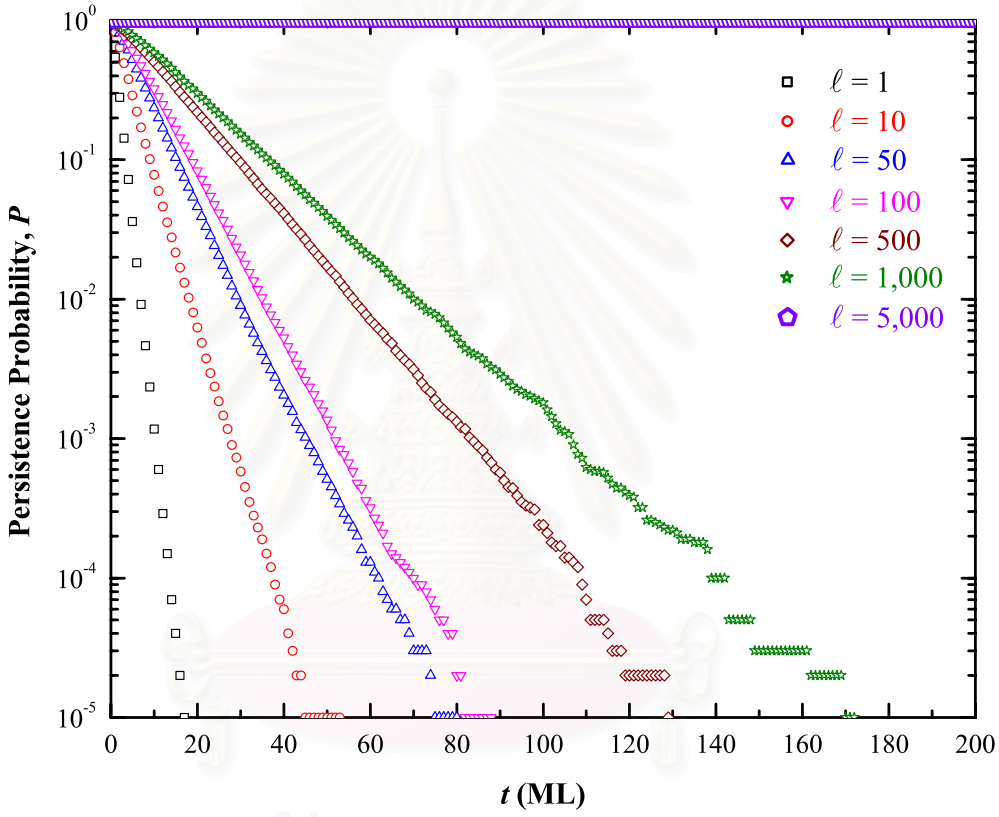


Figure 4.7. Plot of $P(t)$ against t for DT model with the long surface diffusion length NRT ($m = 1$ and $\ell > 1$).

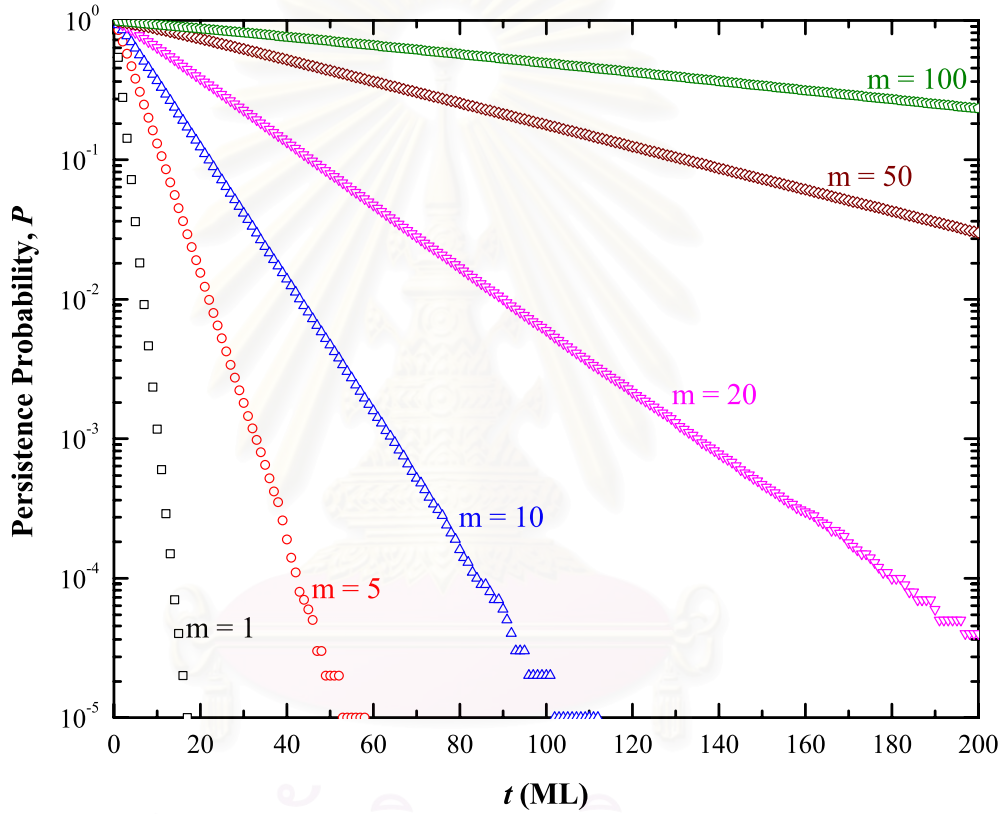


Figure 4.8. Plot of $P(t)$ for DT model with the multiple hit NRT ($\ell = 1$ and $m > 1$).

4.2 Growth on periodic patterned substrates

In this section, the more complicated problems concerning patterned growth are discussed. A specific structure (pattern) is fabricated on the substrate before growth process. This type of growth is an especially interesting case because it may offer a new method to fabricate highly ordered nanostructures. In this work, the initial periodic pattern is chosen as a series of blocks of width size r lattice sites. The height of each of these blocks is h_0 MLs and the blocks are placed at an equal interval throughout the substrate (see Fig. 4.9). We investigate how a periodic pattern propagates through deposited time t MLs. Below we present our simulation results from DT model with the long surface diffusion length and the multiple hit NRTs.

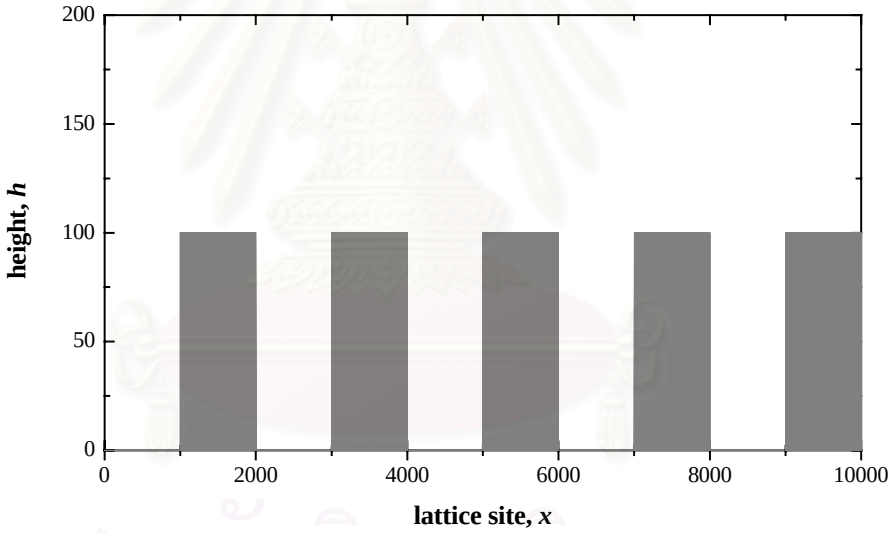


Figure 4.9. Schematic plots of morphology of the periodic patterned substrate, which an initial width r and height h_0 of each block is fixed 1000 lattices and 100 layers respectively.

4.2.1 Applying the long surface diffusion length noise reduction technique

According to growth on initially flat pattern substrates in previous section, the long surface diffusion length NRT can make the flat pattern persisting in a very long time. With this knowledge, we hope that this technique will help us to achieve the same thing in periodic patterned substrate growth. Firstly, we expect that the size of the periodic blocks should have some effects on the pattern persistence. To be more specific, we expect that the persistence probability should decay as the diffusion length approach half the size of each block ($\ell = r/2$). This is due to the fact that adatoms prefer to be incorporated at the bottom edges of blocks where they can increase their number of bonds following DT diffusion rule. As $\ell = r/2$, none of the adatom will be able to nucleate on top of blocks and we lost the pattern as the empty space between blocks are filled up quickly. To prove this, the periodic patterned substrate with $r = 1000$ and $h_0 = 100$ is chosen. Figure 4.10 shows our calculated $P(t)$ results which turn out to be different from our expectation. We loosely divide our results into three ranges of ℓ, comparing to the value of the block size r.

1. At $\ell \ll r$ ($\ell = 1$ and $\ell = 10$), $P(t)$ decays nearly exponentially in the same way as for the initially flat patterned substrate. This is because the diffusion length is so small compared to r that the adatoms do not "see" the pattern, so they are hardly affected by the blocks. Nevertheless, the pattern vanishes through kinetic roughening because the film grows in approximate layer-by-layer mode at very early time, and then the roughness emerges and wins out soon because ℓ is very small. Consequently, the quality of the pattern becomes worse quickly.

2. At $\ell < r$ ($\ell = 50$, 100 and 500), a decline of $P(t)$ is separated into two time regimes. At the beginning of the process, $P(t)$ decays exponentially with smaller slope at higher ℓ (except $\ell = 500$). Afterward, $P(t)$ falls slower and

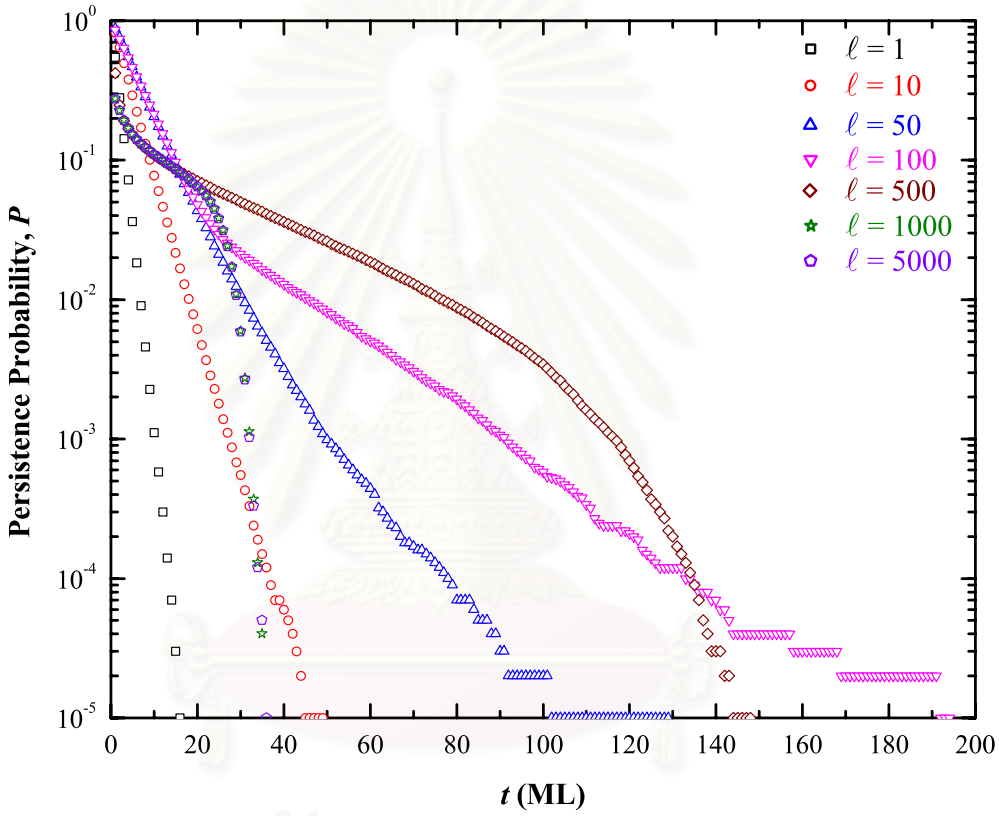


Figure 4.10. Plot of $P(t)$ as a function of t for periodic patterned substrates with $r = 1000$ and $h_0 = 100$ for the selected values of ℓ. All data are fixed at $m = 1$.

persists for long time especially at large ℓ. This can be explained that the adatoms, are able to move a long distance (within ℓ) on the surface, detect the periodic pattern, so they prefer to be incorporated at the bottom edge of blocks in order to increase their numbers of bond. As a result of that, some of them which are deposited on top of the block close to the edge move down to the bottom part. This causes a sharp decrease in $P(t)$. However, this mechanism increases the initial width of blocks, r until the blocks are wide enough for incorporation of adatoms on the top surface. At this point, the diffusion process is more dominant than the effect of the blocks. And since ℓ is quite large, the film can keep the pattern for long deposition time.

3. At $\ell \gtrsim r$ ($\ell = 1000$ and 5000), $P(t)$ decays dramatically since all adatoms cannot nucleate on top of the blocks. They diffuse to bottom part before the nucleation on top takes place. As an example, Fig. 4.11 shows the morphology of the initial periodic pattern $r = 1000$ for $\ell = 1000$. Although the width of blocks is widened, adatoms does not stay on top of the blocks because ℓ is too large. Eventually, the blocks are coalesced and the information of the pattern is completely lost.

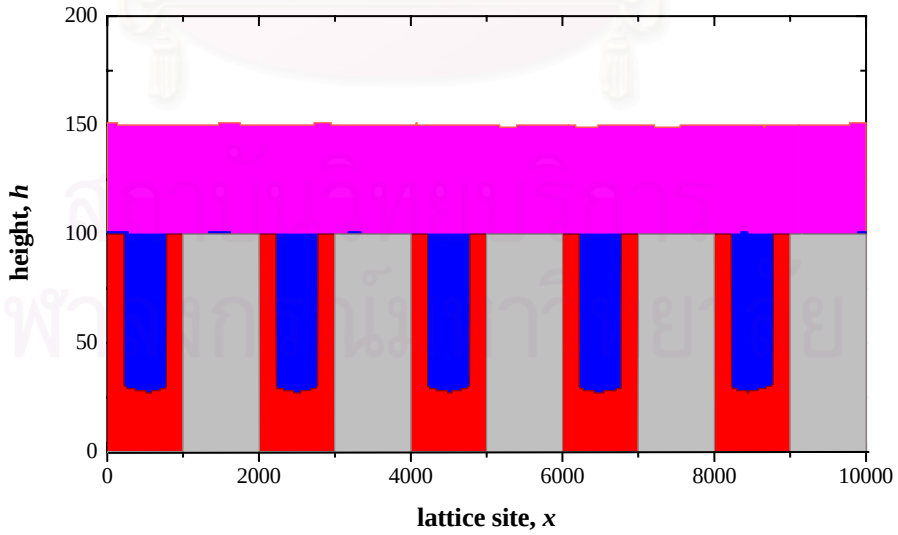


Figure 4.11. The morphologies show the growing film surface for $\ell = 1000$ at $t = 30$ (red), 50 (red and blue) and 100 (red, blue and pink) MLs.

According to our results, diffusion length that is too small (case (i)) or too large (case (iii)) are not a good choice for periodic patterned growth. Appropriate ℓ should lie in case (ii). We will focus on this range to find the optimal value of ℓ for long-lived persistence of pattern. Fig. 4.12 illustrates that the persistence of the pattern decays quickly at $\ell > 500 = r/2$. It indicates that the optimal value of ℓ must be less than $r/2$. Figure 4.13(a) and Figure 4.13(b) are presented here for $r = 200$ and 500 respectively with the same $h_0 = 100$. Results shown in the figures confirm our discussion above.

In order to examine how height of blocks affect the persistence of the pattern, Fig. 4.14(a) and Fig. 4.14(b) show $P(t)$ for $h_0 = 50$ and 200 respectively. Both systems are fixed at width of the blocks $r = 1000$. In the limit that $\ell \gtrsim r/2$, the calculated $P(t)$ of the periodic pattern with $h_0 = 200$ persists for longer time than the periodic pattern with $h_0 = 50$. This is because the empty region in the pattern with smaller h_0 will be filled completely after a deposition of 25 MLs (half of h_0) for very large ℓ. On the contrary, a periodic pattern with $h_0 = 200$ will be completely filled after deposition of 100 MLs. In other words, when h_0 is larger (or the blocks are taller), it takes longer deposition time to completely destroy the pattern so $P(t)$ decays slower. Thus we can conclude that the periodic pattern can survive longer when height of the blocks is larger. Although height of the blocks of the periodic pattern has an impact on the calculated $P(t)$ when ℓ is very large, it does not determine the critical value of ℓ.

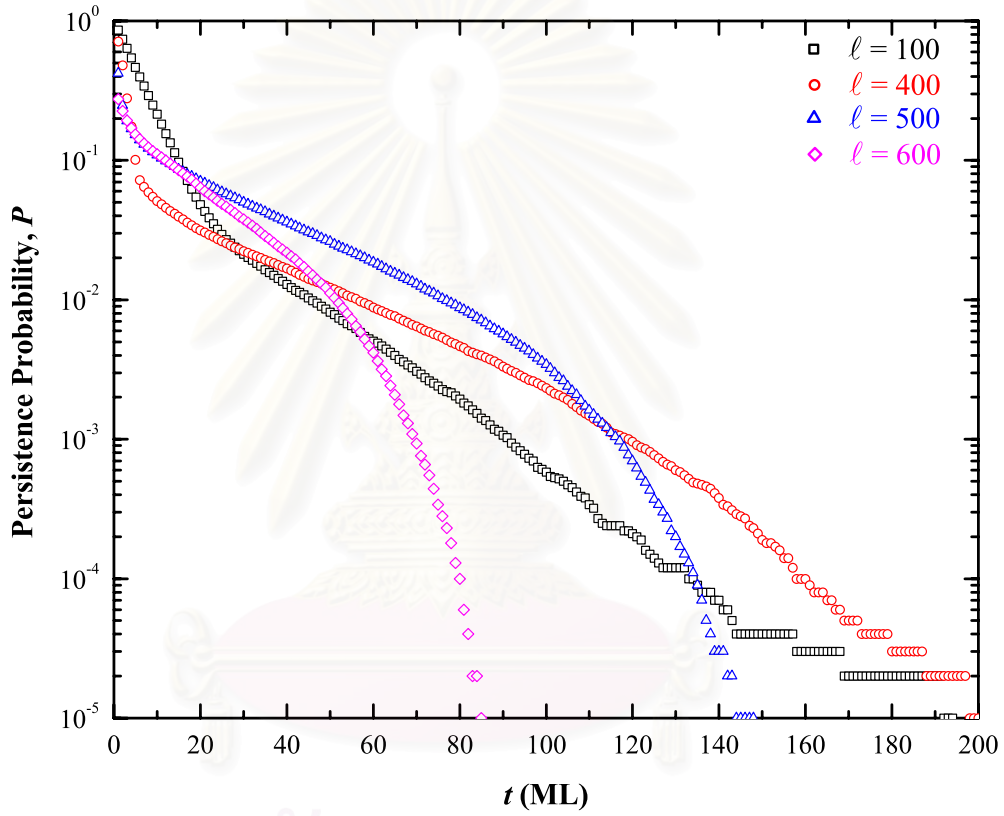
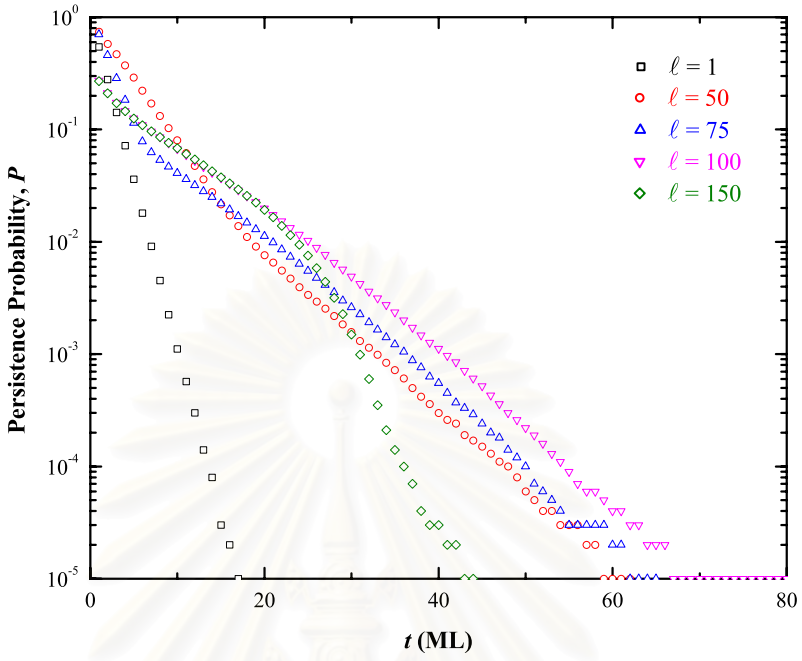
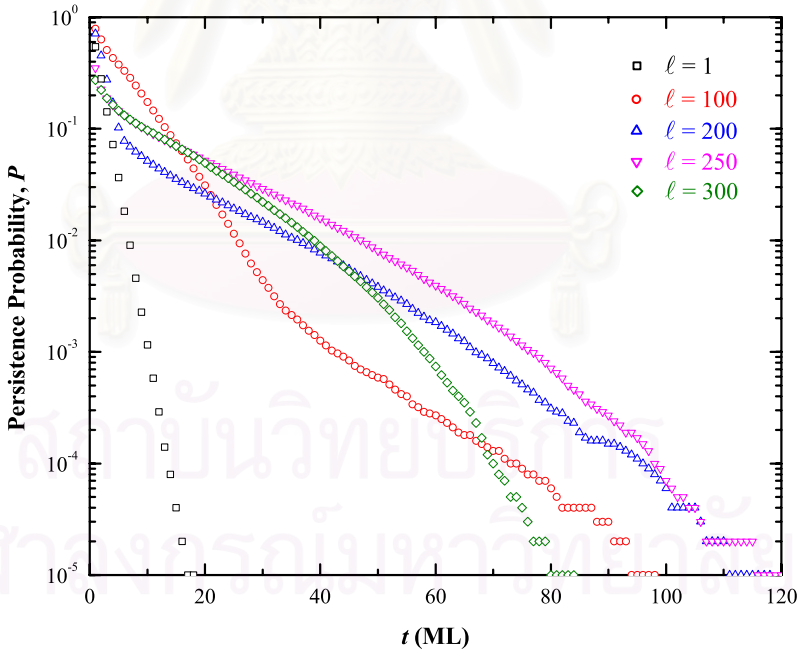


Figure 4.12. Plot of $P(t)$ for periodic patterned substrates with $r = 1000$ and $h_0 = 100$ for the selected values of ℓ.

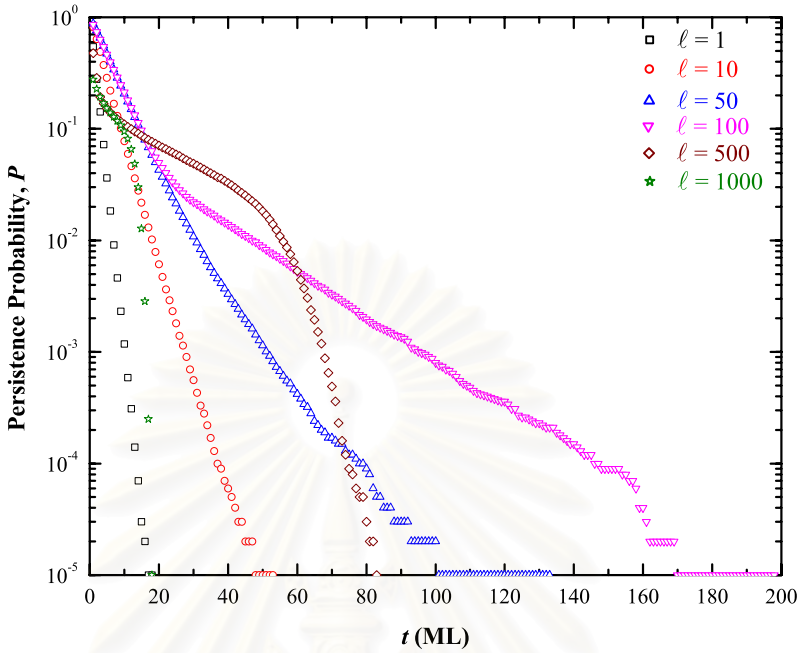


(a)

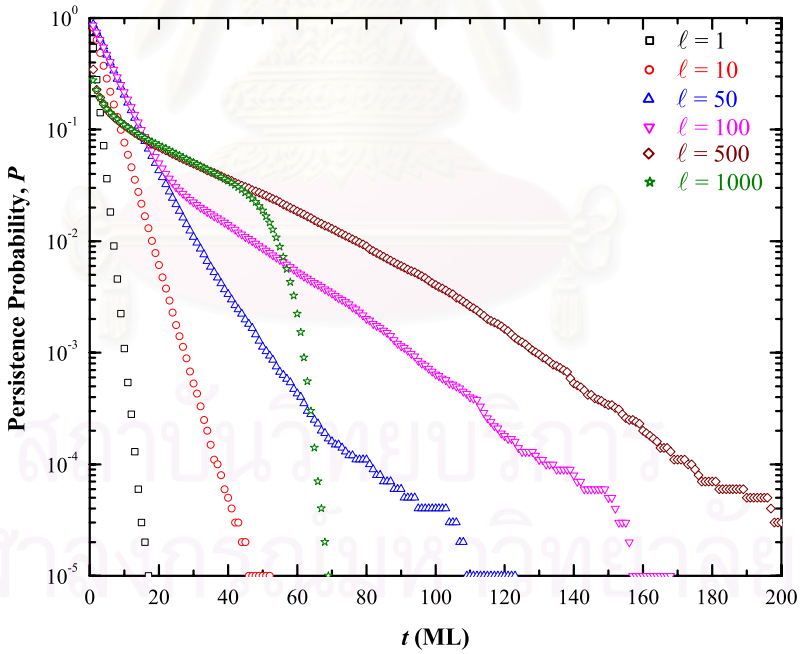


(b)

Figure 4.13. Plot of $P(t)$ as a function of t for periodic patterned substrates with h_0 = 100: (a) r = 200 and (b) r = 500.



(a)



(b)

Figure 4.14. Plot of $P(t)$ as a function of t for periodic patterned substrates with $r = 1000$: (a) $h_0 = 50$ and (b) $h_0 = 200$.

4.2.2 Applying the multiple hit noise reduction technique

Another technique used to reduce noise is the multiple hit NRT. As shown in Sec. 4.1, this technique is equivalent to the long surface diffusion length NRT for flat patterned substrate growth. Therefore, we can replace the long surface diffusion length NRT with the multiple hit NRT in order to save computational time. Below, for the periodic patterned substrate growth, we investigate whether the multiple hit NRT can be used to replace the long surface diffusion length NRT.

Fig. 4.15 illustrates the calculated $P(t)$ for the same system as in Fig. 4.10. It shows that the calculated $P(t)$ decays exponentially for all values of m and the decay is slower as m is larger. These results are different from Fig. 4.10, which is simulated with long surface diffusion length NRT. This can be explained as following. Since the multiple hit NRT is a process that consider only local configuration because deposited atoms are able to move only within nearest neighboring length ($\ell = 1$), so size of the blocks of the periodic pattern does not have any effect on the diffusion process. Thus, applying the multiple hit NRT should not be reasonable for any patterned substrate growth. Consequently, we cannot use the multiple hit NRT in thin film growth on periodic patterned substrate.

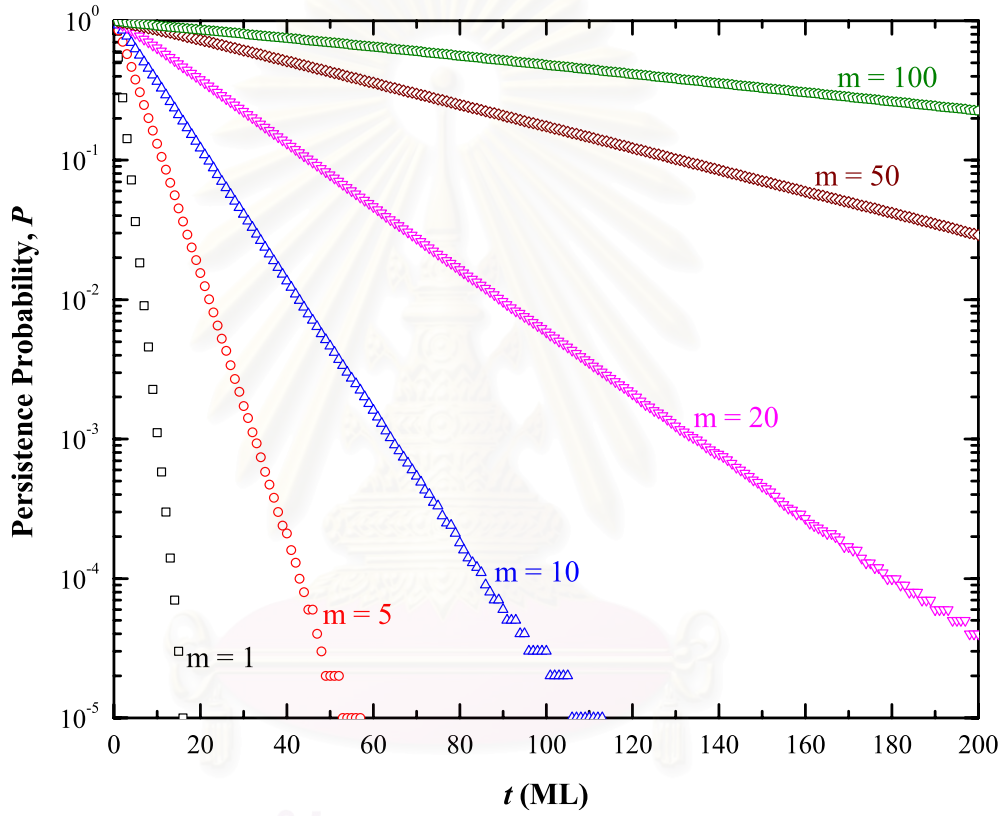


Figure 4.15. Plot of $P(t)$ as a function of t for periodic patterned substrates of $r = 1000$ and $h_0 = 100$ for the selected values of m. All data are fixed at $\ell = 1$.

4.3 New Definition of $P(t)$

All simulation results we have shown so far are calculated from Eq. (4.2), in which the pattern is counted as survive only when the exact pattern is reproduced. This gives the calculated $P(t)$ a very strict definition. Hence, the value of $P(t)$ decreases quickly whereas the morphology still somewhat resembles the original substrate. As an example, Fig. 4.16 shows dynamical morphologies of a film grown on a substrate with periodic pattern with $r = 1000$, $h_0 = 100$ and the diffusion length is $\ell = 50$. At $t = 200$ MLs, the pattern which is a series of blocks, can still be seen quite clearly, but the plot of $P(t)$ shows that the information of the pattern is all disappeared at that time. It means that this definition of $P(t)$ does not agree with the observed morphology. From experimental point of view, this raises a question of how reliable the calculated $P(t)$ is.

Therefore, we suggest a new definition of $P(t)$, which is more flexible. It is denoted $P_n(t)$ and is defined as

$$P_n(t) \equiv \left\langle \prod_{s=1}^{t} F_{\Delta h}(s) \right\rangle_L \tag{4.3}$$

Where

$$F_{\Delta h}(s) = \begin{cases} 1 & \text{if } [h(x,0)+s] - \Delta h \le h(x,s) \le [h(x,0)+s] + \Delta h \\ 0 & \text{otherwise.} \end{cases} \tag{4.4}$$

This new definition makes $P_n(t)$ decays slower compare with the original persistence probability (see Fig. 4.17). This is because the pattern is still counted as survive even when height of the film, $h(x,s)$, differs from the ideal height, $h(x,0)+s$, by a small amount up to Δh. Here Δh, chosen as an integer number, is calculated from the ideal height at that time, $h(x,0)+s$, times ϵ when ϵ is the percentage of "error" we are willing to accept. If $\epsilon = 0$ then $\Delta h = 0$, and $P_n(t)$ returns to the original definition $P(t)$. In Fig. 4.18 we show a plot of $P_n(t)$ as a function of t for varying ϵ when ℓ is fixed at $\ell = 50$. Our results show that $P_n(t)$ persists longer as ϵ is larger. A question comes up as how we can choose a proper

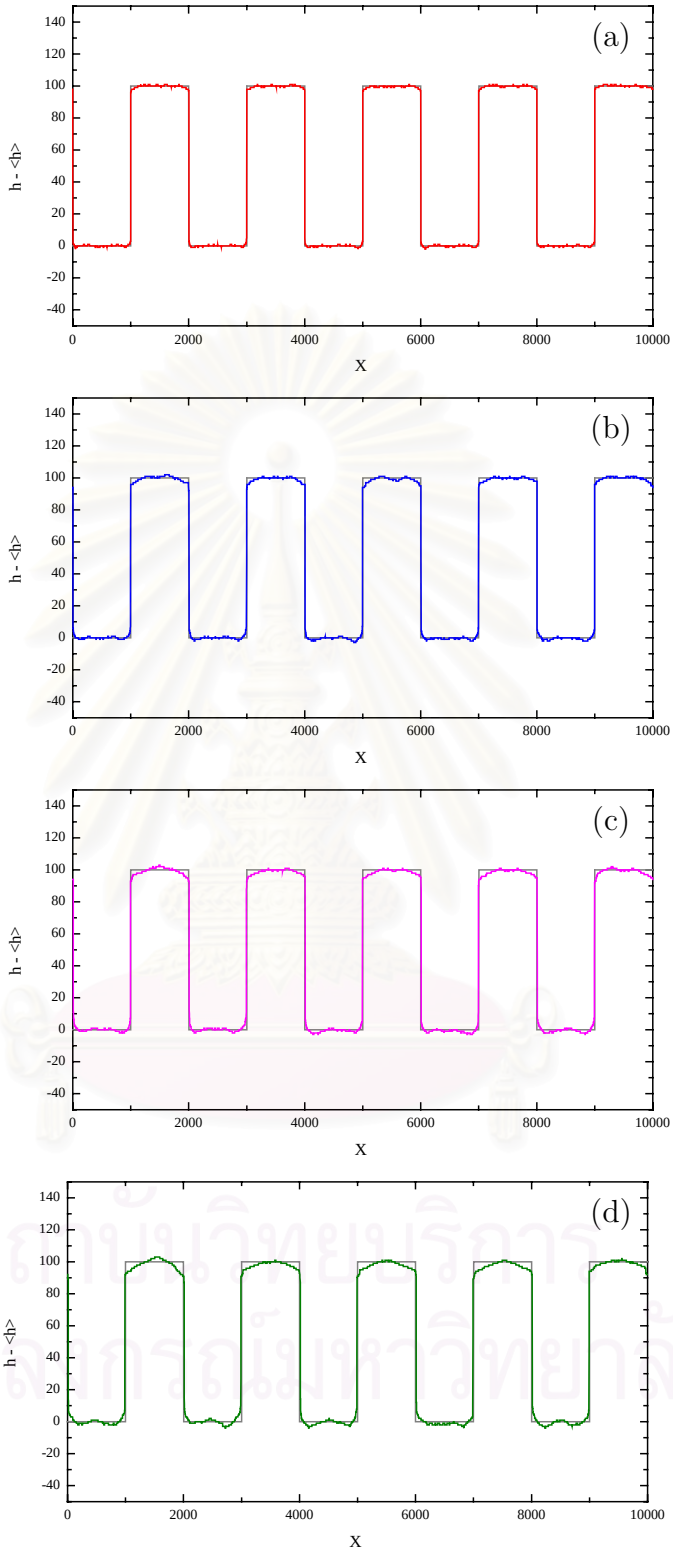


Figure 4.16. Dynamical morphologies of the periodic patterned substrate with $r = 1000$ and $h_0 = 100$ for $\ell = 50$ at growth time $t = 10$ MLs (a), 50 MLs (b), 100 MLs (c) and 200 MLs (d).

value for ϵ. For this question we do not have strict 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 is important to emphasize that our modified definition for the persistence probability $P_n(t)$ does not change the behavior of the film growth, but it changes the behavior of the persistence probability in order to agree better with the morphology.

Although the new definition makes the persistence probability more flexible, it is not without a problem. As shown in Fig. 4.17, for $\epsilon = 1\%$, before 50 MLs the plots of $P_n(t)$ are same as $P(t)$ shown in Fig. 4.10, and then the decay almost stops except when ℓ is greater than $r/2$. In this case, the reason that $P_n(t)$ is not different from $P(t)$ when $t < 50$ MLs is that the value of Δh obtained from ϵ is less than one layer ($\Delta h < 0.5$[‡]) so $\Delta h = 0$ and $P_n(t) = P(t)$. Nonetheless, as t increases, Δh increases to be larger than zero and $P_n(t)$ starts behaving differently from $P(t)$. This means the obtained values of $P_n(t)$ have abrupt changes every time Δh increases, and Δh increases in a step depending on the way the number is rounded in our program. So the behavior of $P_n(t)$ we see in Fig. 4.17 does not actually reflect the real changes in the pattern. To address this problem, we modify the definition of $P_n(t)$ in Eq. (4.3). Instead, Δh is chosen as a fixed constant for whole deposition process. Figure 4.19 shows a modified $P_n(t)$ plot for $\Delta h = 1$ layer.

The modified $P_n(t)$ plot has similar trend as seen in $P(t)$ plot (Fig. 4.19) except for $\ell = 50$ and 100. For the original definition, $P(t)$ of $\ell = 50$ is worse than that of $\ell = 100$, which is reasonable. This is simply because surface with $\ell = 50$ is rougher than with $\ell = 100$. On the other hand, the shape of pattern with $\ell = 100$ is lost faster than the other (see Fig. 4.20). However, when the modified $P_n(t)$ is used in order to determine the persistence of pattern within the range from $[h(x,0) + s] - \Delta h$ to $[h(x,0) + s] + \Delta h$, $P(t)$ of $\ell = 50$ is better. This is because

[‡]In our program we denote that if the number after decimal point is less than five, it is rounded down; otherwise, it is rounded up.

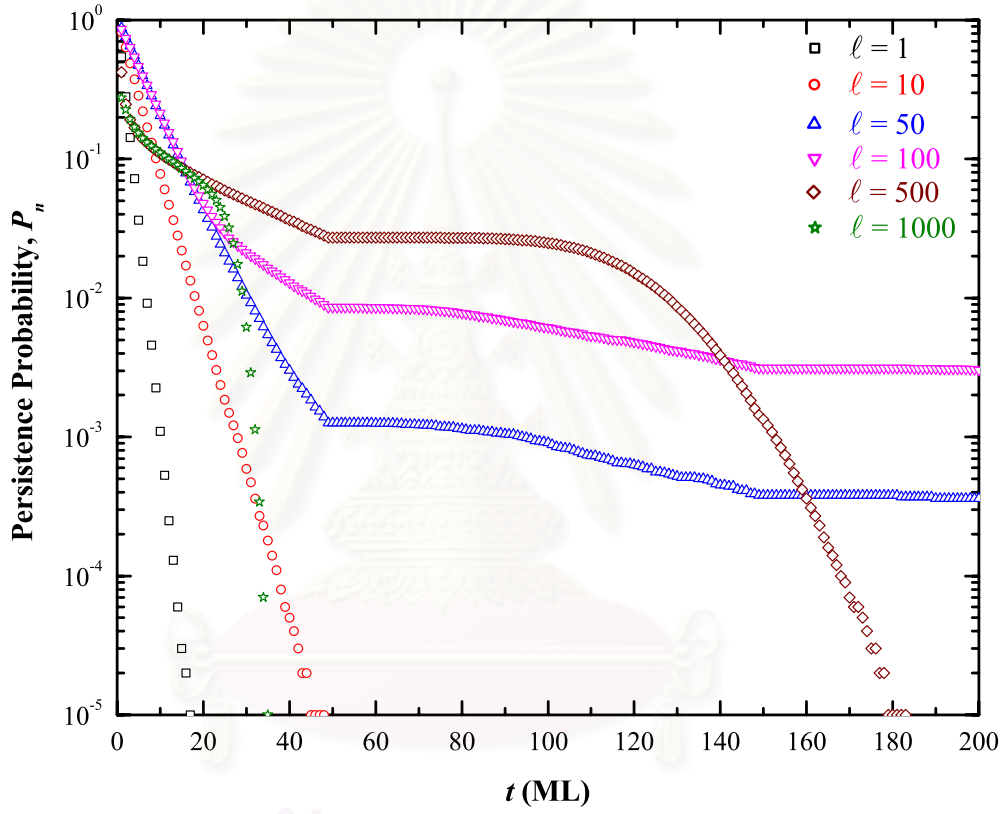


Figure 4.17. Plot of new persistence probability, $P_n(t)$, for the periodic patterned substrate with $r = 1000$ and $h_0 = 100$ for $\epsilon = 1\%$.

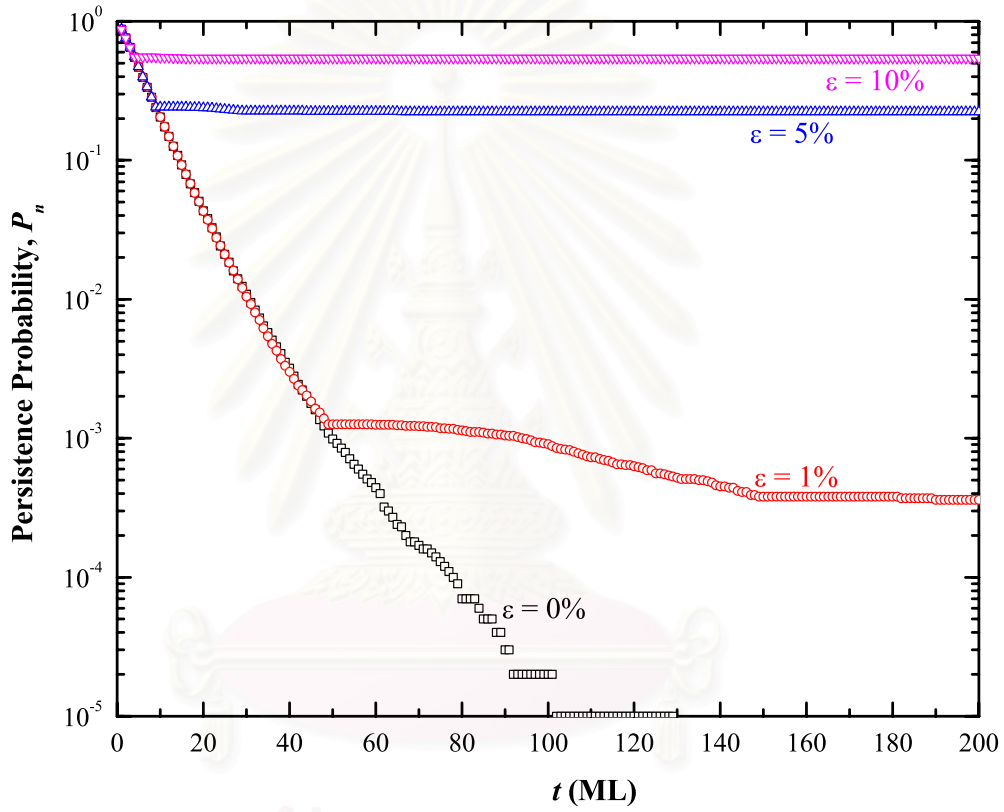


Figure 4.18. Plot of $P_n(t)$ for the periodic patterned substrate with $r = 1000$, $h_0 = 100$ and $\ell = 50$ for the selected values of ϵ.

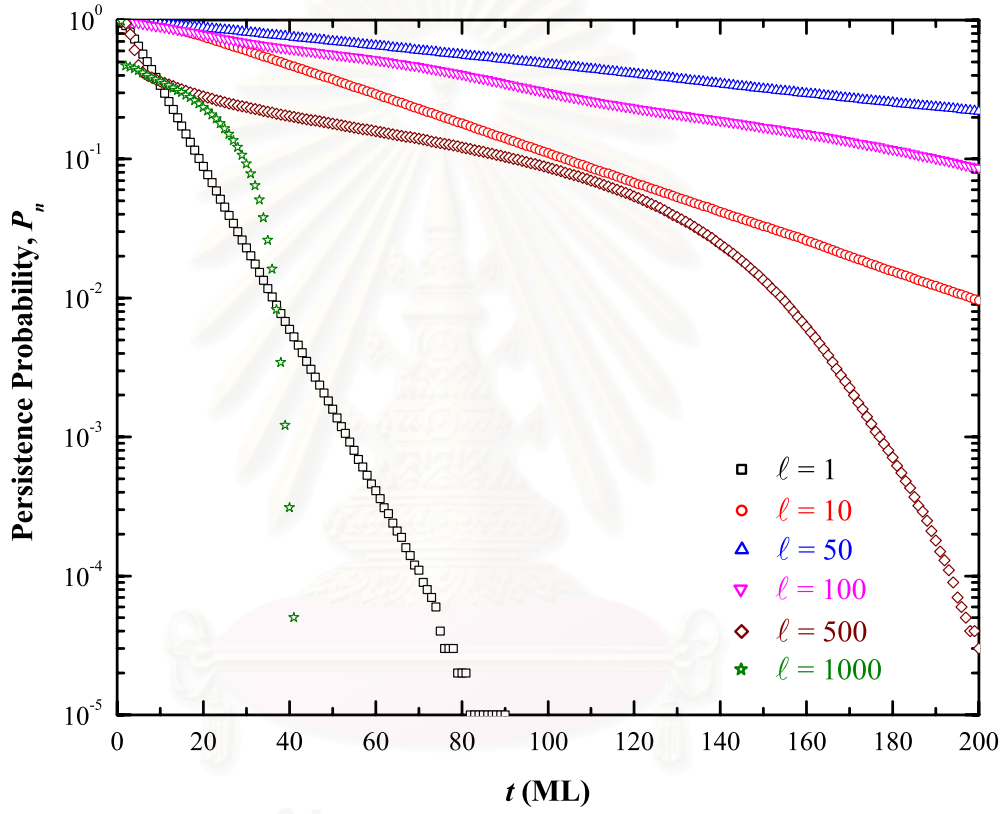
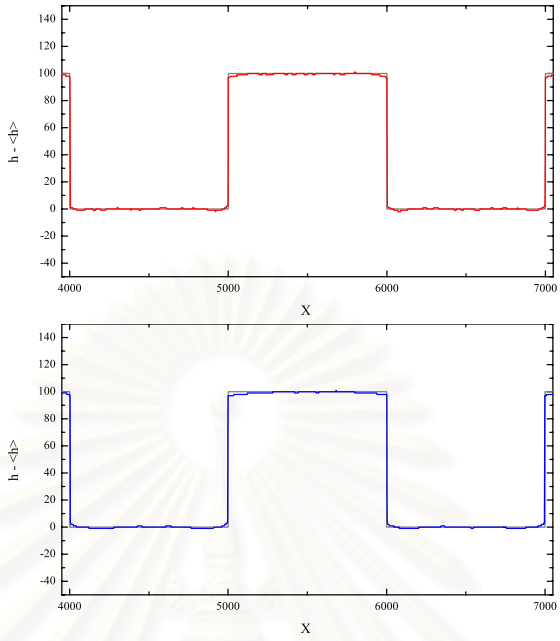
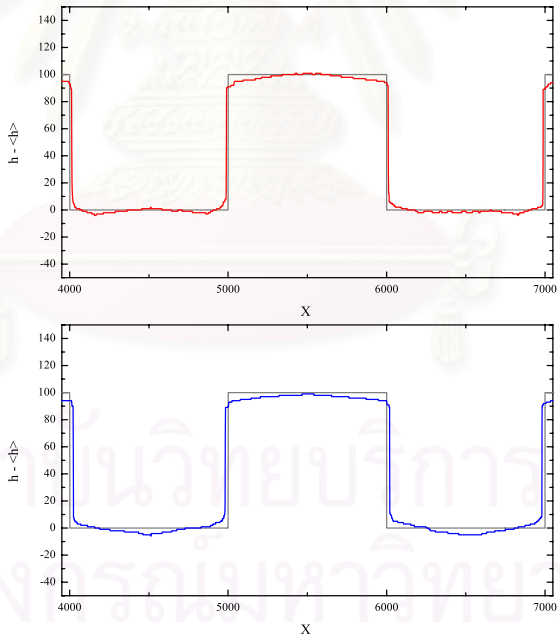


Figure 4.19. Plot of new modified persistence probability, $P_n(t)$, for the periodic patterned substrate with $r = 1000$ and $h_0 = 100$ for $\Delta h = 1$.



(a)



(b)

Figure 4.20. Comparing the Morphologies of the periodic patterned substrate with r = 1000 and h_0 = 100 between ℓ = 50 (red line) and ℓ = 100 (blue line) (a) at growth time t = 10 MLs and (b) at growth time t = 200 MLs.

the shape of the pattern with $\ell = 50$ resembles the substrate closely whereas the pattern with $\ell = 100$ loses information from the substrate as t increases. Since in the growth with $\ell = 100$, most atoms falling on top of the blocks can diffuse down to the lower edge of the blocks, the blocks in the substrate become wider and less tall compare with the original pattern.

To sum up, the long-lived persistence depends on two factors: smoothness[§] of the flat part and shape of the pattern. In order to grow smooth film, ℓ should be increased; but on the other hand, large ℓ destroys the shape of pattern since adatoms prefer to be incorporated at the lower edges of blocks. So far patterned growth, we need to choose the diffusion length to be large enough to have a relatively smooth surface, but we also need to be careful that ℓ should not be too large in order to keep the sharp of the pattern.

[§]According to Chapter 2 the smoothness of film surface is concerned with a competition between deposition and surface diffusion, but in our work the deposition rate, F, is fixed 1 ML/second. Therefore, we attend the diffusion process only.

Chapter 5

Conclusions

In this work, the effect of a patterned substrate on adatom nucleation during epitaxial growth is studied using DT model in (1+1)-dimension. In this approach, each atom is assumed to be a square lattice, and the growth is under SOS constraint, i.e. overhangs, bulk vacancies and desorption are not allowed. In addition, we do not include the influence of elastic strain and Ehrlich-Schwoebel (ES) barriers. Two initial substrate patterns are chosen: a flat patterned substrate, and a periodic patterned substrate with equal sized blocks characterized by the width of blocks, r, and the height of blocks, h_0. For the flat patterned substrate, it can be viewed as the periodic patterned substrate with $r = 0$ or $r \to \infty$. In order to study the effect of a periodic patterned substate, two noise reduction techniques (NRTs) are applied to the model, namely the long surface diffusion length NRT in which we vary the value of the surface diffusion length ℓ, and the multiple hit NRT where we vary the value of the multiple hit factor m. The results of the two NRTs are compared.

For the flat patterned substrate, we have shown that films can be grown in layer-by-layer mode as we increase ℓ or m, with the result that the flat patterned thin film persists a very long time. Our results also show that the two NRTs are equivalent. Moreover, there is no limit for these values in computer simulations. In a physical meaning, the larger ℓ corresponds to the higher substrate temperature, T, whereas the multiple hit NRT is used only as a trick in simulations. Hence, we can say that the flat patterned substrate has long-lived persistence as the film

is grown at sufficiently T (but not too high as desorption occurs at very high T). However, in higher substrate dimension increasing ℓ is difficult and extremely time consuming in simulations; therefore, the multiple hit NRT is used instead.

For the periodic patterned substrate, the film growth is not only based on the surface diffusion process but also dependent on the characteristics of the pattern, i.e. r and h_0. At small ℓ (low T) the exact pattern cannot be reproduced. However, at large ℓ (compared with r) the persistence of the pattern also decays dramatically. We have presented that all adatoms with large ℓ cannot nucleate on top of the blocks. On the contrary, they hop down to fill empty space between each blocks. Increasing ℓ, therefore, is limited by r. This is also a valid conclusion for the flat patterned substrate characterized by $r \to \infty$. We also found that the multiple hit NRT cannot be used for the periodic patterned substrate growth because we cannot include the effect of r in the process.

Moreover, we have pointed out that the original definition of the persistence probability, $P(t)$, which counts the fraction of the exact pattern that is reproduced after each time step, is too strict. At small ℓ, $P(t)$ plot decays sharply while a shape of the pattern can be maintained for a reasonably long time. In some cases, this "slightly off" pattern, with some roughness in the originally flat part of the substrate, can still be acceptable. To this end, we have improved the definition of $P(t)$ in order to be more flexible, and finally we have modified the new definition of persistence probability, $P_n(t)$. We found that the behavior of $P_n(t)$ as a function of time t agrees with our observed morphology better than the original $P(t)$. Furthermore, we found that the long-lived persistence of the pattern depends on two factors: smoothness of flat part and shape of the pattern.

In this thesis we did not include the effect of ES barriers on the patterned growth. Since it is well known that such energy barriers occur in many experimental systems, it should be interesting to study their effect. One expects that the persistence of the pattern under the influence of ES barriers is similar to the persistence without ES barriers for small ℓ because adatoms would rather be in-

corporated into the same layer than diffuse to the lower part. The intention in future work is to extend the DT model in (2+1)-dimensions to study the above-mentioned cases in more detail.

References

Barabási, A.-L. and Stanley, H. (1995). *Fractal Concepts in Surface Growth*. New York: Cambridge University Press.

Braun, W. (1999). *Applied RHEED Reflection High-Energy Electron Diffraction During Crystal Growth*. Germany: Springer.

Chatraphorn, P. P. and Das Sarma, S. (2002). Layer-by-layer Epitaxy in Limited Mobility Nonequilibrium Models of Surface Growth. *Phys. Rev. E* 66: R4863.

Das Sarma, S. (1997). Dynamic scaling in epitaxial growth. *aiXiv:cond-mat* 9705118v2.

Das Sarma, S. et al. (1996). Scale invariance and dynamical correlations in growth models of molecular beam epitaxy. *Phys. Rev. E* 53: 359.

Das Sarma, S. and Chatraphorn, P. P. (1997). Dynamic scaling in a (2+1)-dimensional limited mobility model of epitaxial growth. *Phys. Rev. E* 55: 5361.

Das Sarma, S. and Ghaisas, S. V. (1992). Solid-on-Solid Rules and Models for Nonequilibrium Growth in 2+1 Dimensions. *Phys. Rev. Lett.* 69(26): 3762.

Das Sarma, S. and Tamborenea, P. I. (1991). A New Universality Class for Kinetic Growth: One-Dimensional Molecular-Beam Epitaxy. *Phys. Rev. Lett.* 66(3): 325.

Elshabini-Raid, A. and Barlow, F. (1998). *Thin Film Technology Handbook*. New York: McGraw-Hill.

Family, F. (1986). Scaling of rough surfaces: effects of surface diffusion. *J. Phys. A* 19(8): L441.

Ghaisas, S. and Das Sarma, S. (1992). Surface Diffusion Length Under Kinetic Growth Conditions. *Phys. Rev. B* 46(11): 7308.

Kallabis, H. and Wolf, D. (1997). Growth of Patterned Surfaces. *Phys. Rev. Lett.* 79(24): 4854.

Kim, J. M. and Das Sarma, S. (1994). Discrete models for conserved growth equations. *Phys. Rev. E* 72: 2903.

Krug, J. (2002). Four Lectures on the Physics of Crystal Growth. *Physica A* 313: 47–82.

Lai, Z.-W. and Das Sarma, S. (1991). Kinetic growth with surface relaxation: Continuum versus atomistic models. *Phys. Rev. E* 66: 2348.

Lanczycki, C. (1995). *Nonequilibrium Interface Growth: Roughening, Coarsening and Scale Invariance*. Ph. D. Thesis, Faculty of the Graduate School of the University of Maryland.

Ohring, M. (1992). *The Material Science of Thin Films*. San Diego: Academic Press, Inc.

Punyindu, P. (2000). *Understanding Kinetic Surface Roughening Using Local, Discrete, Nonequilibrium Growth Models*. Ph. D. Thesis, Faculty of the Graduate School of the University of Maryland.

Punyindu, P. and Das Sarma, S. (1998). Noise Reduction and Universality in Limited Mobility Models of Nonequilibrium Growth. *Phys. Rev. E* 57: 041601.

Siegert, M. and Plischke, M. (1994). Solid-on-solid models of molecular-beam epitaxy. *Phys. Rev. E* 50: 917.

Tamborenea, P. I. and Das Sarma, S. (1993). Surface-diffusion-driven kinetic growth on one-dimensional substrates. *Phys. Rev. E* 48: 2575.

Wolf, D. E. and Villain, J. (1990). Growth with surface diffusion. *Europhys. Lett.* 13: 389.

Yang, H.-N.; Wang, G.-C.; and Lu, T.-M. (1994). Instability in Low-Temperature Molecular-Beam Epitaxy Growth of Si/Si(111). *Phys. Rev. Lett.* 73(17): 2348.

Vitae

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S. Piankoranee and P. Chatraphorn. Noise reduction techniques in thin film growth on patterned substrate. *8th Annual National Symposium on Computational Science and Engineering*, Suranaree University of Technology (21-23 July 2004): CP-O05

S. Piankoranee and P. Chatraphorn. Persistence in thin film growth on patterned substrates. *12th Annual Academic Conference*, Faculty of Science, Chulalongkorn University (18-19 March 2004): PH 5

2003 S. Piankoranee and P. Chatraphorn. Persistence in thin film growth on patterned substrates. *29th Congress on Science and Technology of Thailand*, Khon Kaen University (20-22 October 2003): SD-490