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EFFECTS OF MISCUT SUBSTRATES ON SURFACE MORPHOLOGY OF THIN
FILMS SIMULATED BY DAS SARMA-TAMBORENEA MODEL

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A Thesis Submitted in Partial Fulfillment of the Requirements

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SARMA-TAMBORENEA MODEL) อ.ที่ปรึกษาวิทยานิพนธ์หลัก: ผศ.ดร. ปัจฉา
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แบบจำลองดาสซาร์มา-แทมโบเรเนีย (DT) เป็นแบบจำลองที่ใช้ศึกษากระบวนการปลูกฟิล์ม
บางโดยวิธีโมเลกุลาร์บีมเอพิแทกซี (MBE) ในแบบจำลอง MBE การแพร่ของอะตอมเป็นไปตามความ
น่าจะเป็นของอาร์เรนเนียสซึ่งขึ้นอยู่กับจำนวนพันธะกับอะตอมข้างเคียง และอุณหภูมิของซับสเตรต
แบบจำลอง DT ลดความซับซ้อนในการคำนวณความน่าจะเป็นของอาร์เรนเนียสและได้ใช้กระบวนการ
นับพันธะอย่างเดียวในกระบวนการแพร่ อุณหภูมิของซับสเตรตจะถูกเปลี่ยนแปลงโดยการเปลี่ยนระยะ
การแพร่ ในวิทยานิพนธ์นี้ แบบจำลอง DT กับการเพิ่มระยะการแพร่บนพื้นผิวถูกนำมาใช้ในการศึกษา
การปลูกฟิล์มบางบนมิสคัตซับสเตรต ซึ่งพบว่าการเปลี่ยนแปลงระยะการแพร่สังเกตเห็นไม่ชัดในฟิล์ม
ที่ได้จากแบบจำลองซึ่งไม่สอดคล้องกับผลการทดลอง เราจึงปรับปรุงกฎการแพร่แบบจำลอง DT โดย
เพิ่มความสามารถในการเคลื่อนที่ของอะตอม และผลจากแบบจำลองที่ถูกปรับปรุงนั้นสอดคล้องกับผล
การทดลอง ส่วนแรกเราใช้แบบจำลอง DT ที่ถูกปรับปรุงเพื่อศึกษาผลของระยะการแพร่บนพื้นผิว (ℓ_d)
บนมิสคัตซับสเตรตเมื่อความกว้างของขั้นบันได (L_t) และ ความสูงของขั้นบันได (s) คงที่ เราพบว่า
ฟิล์มสามารถรักษารูปแบบเดิมไว้ได้เป็นเวลายาวนานเมื่อ $\ell_d \geq L_t$ ส่วนที่สองผลของการจัดเรียงตัว
ของมิสคัตซับสเตรตตอนเริ่มต้นถูกนำมาศึกษา ในกรณีแรก ความกว้างของขั้นบันไดถูกแปรค่าและ
ความสูงของขั้นบันไดคงที่เท่ากับหนึ่ง เราพบว่ารูปแบบของซับสเตรตตอนเริ่มต้นสามารถคงอยู่ได้นาน
เมื่อ $L_t \leq \ell_d$ กรณีที่สอง ความสูงของขั้นบันไดถูกแปรค่าและความกว้างของขั้นบันไดคงที่เท่ากับ
ระยะการแพร่บนพื้นผิว $L_t = \ell_d$ เราพบว่า $s > 1$ เหนี่ยวนำให้เกิดความขรุขระขึ้นบนผิวฟิล์ม ยืนยันได้
จากการลดลงอย่างรวดเร็วของค่าความน่าจะเป็นของการคงอยู่

ภาควิชา............ฟิสิกส์............... ลายมือชื่อนิสิต...
สาขาวิชา...........ฟิสิกส์............... ลายมือชื่ออ.ที่ปรึกษาวิทยานิพนธ์หลัก.................
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KEYWORDS: MISCUT SUBSTRATE/ SURFACE MORPHOLOGY/ THIN FILM SIMULATED/ DAS SARMA-TAMBORENEA MODEL

CHANAKAN CHOMNGAM: EFFECTS OF MISCUT SUBSTRATES ON SURFACE MORPHOLOGY OF THIN FILMS SIMULATED BY DAS SARMA-TAMBORENEA MODEL. THESIS ADVISOR: ASST. PROF. CHANAKAN CHOMNGAM, PH.D., 89 pp.

The Das Sarma-Tamborenea (DT) model is a model used to study molecular beam epitaxy (MBE) thin film growth process. In MBE simulations, atomic diffusion follows Arrhenius hopping probability which depends on number of nearest-neighbor bonds and substrate temperature. The DT model simplifies this and uses only bond counting in the diffusion process. Substrate temperature can be varied via the change in the diffusion length. In the thesis, the DT model with long surface diffusion length is used to study thin film growth on miscut substrates. It is found that the change in the diffusion length is not clearly seen in the simulated film, which disagrees with experimental results. We modified the diffusion rule of the DT model by increasing the mobility of atoms and results from the modified DT model agree with experiments. First, using the modified DT model to study effects of surface diffusion length (ℓ_d) on the miscut substrate as the terrace width (L_t) and the step height (s) is fixed, we found that the film can maintain the initial tilt angle for a long time when $\ell_d \geq L_t$. Seconds, the effects of the initial configuration of miscut substrate is studied. The first case, the terrace width is varied and step height is fixed at 1 sites, we found that the initial pattern can survive for a long period of time when $L_t \leq \ell_d$. In the second case, the step height is varied and the terrace width is fixed at $L_t = \ell_d$, we found that $s > 1$ induces roughness on the film surface confirmed by the rapidly decreasing persistence probability.

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Field of Study: ...Physics.......

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CHAPTER I

INTRODUCTION

The word "nanotechnology" has become very important. It affects many types of research. For example, high quality thin film growth technique is required for nano-device fabrications such as electronics semiconductors devices and optical coatings. Many studies and researches, both experiment and theory, concentrate on gaining some understanding about the growth process. Computer simulation is used to study the thin film growth process because it is easier to control the growth conditions such as desorption, bulk vacancies/ overhanging formation, etc. Most studies of thin film growth simulations are performed on an initially flat substrate. However, in reality, it is rare to obtain a completely flat substrate (Pal and Landau, 1996; Tejedor et al., 1999). A substrate is usually miscut leading to a slightly tilted surface. The tilted surface looks like terrace in small scale which is shown in figure 1.1. This substrate is called a miscut substrate or stepped surface (Tejedor et al., 1999) which has been observed during chemical vapor deposition (CVD) (Krishnamurthy et al., 1992; Ishizaki et al., 1996) and molecular beam epitaxy (MBE) of homoepitaxial layer, especially in the $Si/Si_{1-x}Ge_x$ material system (Myslivecek et al., 2002). During the diffusion process, atoms can diffuse to be incorporated at a step edge of the terrace which means the film is grown in step flow (SF) growth mode. The perfect film on a miscut substrate is obtained when the film is grown in perfect SF mode. This is used as an ideal template to build an anisotropic structure such as quantum wires (Videcoq and Pimpinelli, 2001). The topic of growth on patterned substrates has also attracted interest due to its importance on the production of semiconductor devices on nanoscale structures (Gaines et al., 1998; Fukui and Saito, 1987).

In this work, the Das-Sarma Tamborenea (DT) model which is known to generate films with similar statistical properties to films grown by molecular beam epitaxy (MBE) at low temperature (Das Sarma and Tamborenea, 1991) is used to study growth on a tilted substrate. In MBE simulations, atomic diffusion follows Arrhenius hopping probability which depends on number of bonds and substrate temperature. The DT model simplifies this and uses only bond counting in the diffusion process. Substrate temperature can be varied via the change in the diffusion

length. The DT model with long surface diffusion length and a modification to the diffusion rule, called modified DT model, is used for the simulation. A modification of the DT model is done in order to increase the mobility of atoms by increasing number of bond an atom can break which is equivalent to increasing substrate temperature in experiments. In order to study effects of the substrate temperature, the long surface diffusion length noise reduction technique (Chatraphorn and Das Sarma, 2002) is added to the original DT and modified DT model. To do this, the diffusion rule of each model is slightly modified. The noise reduction technique helps to reduce noise which causes roughness of films. This technique was used to study growth on flat substrates (Das Sarma and Ghaisas, 1992; Das Sarma et al., 2001). The noise reduction technique is adapted for growth on miscut substrates in this work.

The purpose of this thesis is to study effects of the miscut substrate on the thin film growth process by simulations and to find the optimum condition for growth on a miscut substrate. Studying effects of the initial configuration of the miscut substrate is done by varying width of the terrace and height of the terrace. Effects of the substrate temperature on growth on a miscut substrate are also investigated. In chapter 2, microscopic thin film growth processes and modes of thin film growth are explained. Models used in this work are also presented in this chapter. In chapter 3, definition of quantities of interest such as the film morphology, interface width, persistence probability and correlation functions are discussed. All results are presented in chapter 4. The results are divided into two parts. The first part shows effects of the surface diffusion length on films grown on substrates with the same initial configuration. In the second part, effects of the initial configuration of the miscut substrate are discussed. Finally, conclusions are given in chapter 5.
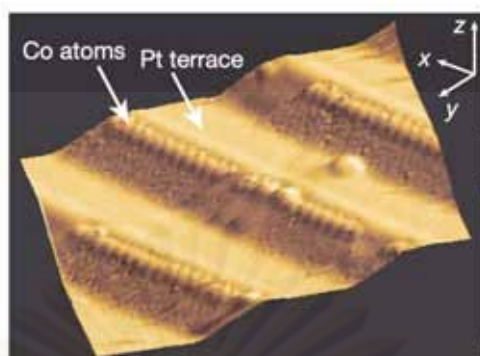
Figure 1.1: STM topographs of the Pt (997) surface. Periodic step structure of Co atoms on Pt terrace held at $T = 260$ K and previously cleaned by ion sputtering and annealing cycles in ultrahigh vacuum (UHV). ("www.nature.com/... /v416/n6878/full/ 416301a.html")

CHAPTER II

THEORY AND MODEL

2.1 Thin film growth process

In order to understand important mechanism in thin film growth, the microscopic processes on the surface is studied. There are three processes. The deposition, surface diffusion and desorption process in the thin film growth. The interplay of these three processes affects quality of the grown film.

2.1.1 Deposition process

In the first process, atoms (or molecules) are randomly dropped on the surface, form bonds and then stick with the surface. This process is the *deposition process* (atom A shown in Figure 2.1). In experiments, deposition process is performed in high vacuum and in ultrahigh vacuum conditions ‒ pressures smaller than 10^{-10} torr. The deposition material is thermally evaporated to be in the form of vapor or beam from a source. In most simulations including ones in this work, each atom is represented by a cubic box and is dropped vertically on the surface. Each cubic consists of only one atom. It represents the simple cubic crystal structure which consists of one lattice point per unit cell. During deposition, atoms are dropped steadily by deposition rate, which is defined as the average number of deposit layers per unit time. In this work, the deposition rate is one monolayer per second (ML/s). If deposit atoms are of the same material as the substrate, it is called *homoepitaxy growth*. On the other hand, growth that deposit atoms grown on a substrate of another material is called *heteroepitaxy growth*. In this work, film is grown in homoepitaxy growth**.**

2.1.2 Surface diffusion process

As an atom is already on the surface, it can break its initial bond and move on the surface. This process is *surface diffusion process* (atom B in Figure 2.1). If an adatom, atom that was just dropped on the surface, is dropped at an unstable site, it diffuses and locates a stable site. The length that atom can diffuse is called surface *diffusion length*, ℓ_d which depends on the substrate temperature (Barabasi and Stanley, 1995). In MBE simulation, as atoms diffuse on the terrace, atomic diffusions follows Arrhenius equation or hopping rate R_n which each surface atom hops continuously. Hopping rate depends on activation energy E_a and the substrate temperature T (Das Sarma and Tamborenea, 1991). The substrate temperature controls the energy of atoms in the system. The Arrhenius hoping probability has an exponential dependence,

$$R_n = R_0 \exp\left(\frac{-E_A}{k_B T}\right). \tag{2.1}$$

where R_0 has a weak temperature dependence by $R_0 = 2k_B T/h$, k_B is the Boltzmann constant, T is the substrate temperature, E_A is calculated from $E_A = E_0 + nE_B$, where E_0 is the activation energy of a free atom (no lateral bonds), E_B is the binding energy per bond and n is the number of nearest-neighbor bonds varying between 1-5 for growth on a two dimensional substrate.

For diffusion rule of the DT model, growth conditions are chosen as E_0=0.3 eV and E_B =1 eV which are semiquantitative consistent with Si and GaAs. It is also set that R=0 for $n \geq 2$. Atomic diffusion is limited to diffuse of atom by number of nearest-neighbor bonds n. So an atom sticks on the terrace and forms two bonds with the surface, it can not diffuse. This condition becomes diffusion rule of the DT model. The site that atom forms two bonds with the surface, is called *kink sites* (atom E is shown in Figure 2.1) and the sites that atom forms three bonds with the surface, is called *trapping sites* (atom D is shown in Figure 2.1). The DT model coincides with molecular beam epitaxy (MBE) at low temperature.

2.1.3   Desorption process

   Desorption process is the opposite process of the deposition process. When an atom is deposited on the surface, it can evaporate from the surface (atom C in Figure 2.1). Desorption process can induces roughness of the film (Xiao and Ming, 1994; Xiao, 1997). The average time that atoms evaporate from the surface, is called *desorption time*, τ (Barabasi and Stanley, 1995), which follows as

$$\tau \propto \exp\left(\frac{E_D}{k_B T}\right). \tag{2.2}$$

where E_D is the characteristic desorption energy to release an atom from the surface, k_B is the Boltzmann constant and T is the substrate temperature. In this work, the growth condition is used similarly to MBE growth at low temperature where the desorption process rarely occurs. Desorption process is then neglected in this work. If the growth process does not allow desorption. it shows that all incoming atoms per unit time or deposition rate equals to sticking atoms per unit time (impingement rate).

2.2   Thin film growth mode

   During the growth processes, the film can be grown in three different modes. Two-dimensional nucleation (*2D*) growth mode, three- dimensional (*3D*) growth or island growth mode and step flow (*SF*) growth mode. They are shown in Figure 2.2.
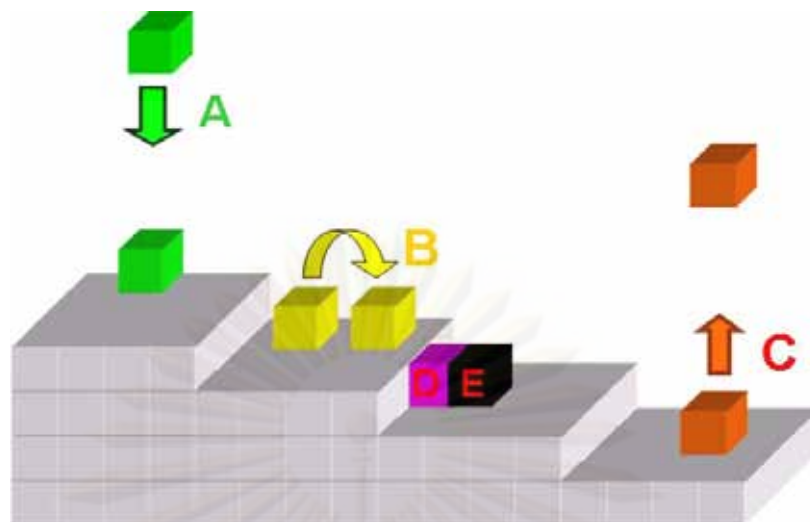
Figure 2.1: Three processes of thin film growth on two dimensions miscut substrate.

Process A represents deposition process.

Process B represents surface diffusion process.

Process C represents desorption process.

2.2.1 Two-dimensional nucleation (*2D*) growth

2D nucleation growth mode is also known as *layer by layer growth.* In this mode, each layer of the thin film is filled up completely before the next layer begins. Layer by layer growth mode can occur on any surface at high temperature when diffusion length is large. If the film is grown by layer by layer growth mode, a smooth film is obtained. In experiments, layer by layer growth mode is studied by the reflection high-energy electron diffraction (RHEED) (Neave et al., 1985 ; Marmorkos and Das Sarma, 1992; Zhang et al., 1997), which is a popular technique for the growth of thin film under ultrahigh vacuum growth condition. In the RHEED pattern or intensity curve, the number of oscillations corresponds to the growing layer and the curve oscillates as new layers are continuously formed. Relations between the growing film and RHEED intensity pattern is shown in figure 2.3. Figure 2.3(a) shows the growing film morphologies and figure 2.3(b) shows the RHEED oscillation pattern. From the figure, the intensity of the RHEED pattern reaches its maximum always at each completely filled layer. In simulations, layer by layer growth mode

observed by an oscillation in the plots of interface width (roughness of film) as a function of growth time t.

2.2.2 Three-dimensional (*3D*) growth

At low temperature or small diffusion length, *3D growth* or island growth mode can occur. The *2D* growth can switch to *3D* growth if a new layer begins forming before the previous layer is completely filled. This growth mode occurs at low temperature with small diffusion length. It leads to island or mound formation. The lateral size of an island is usually 10-50 of the lattice constant (Fachbereich and Gerhard, 1999). A statistic quantity which identifies island formation is the correlation function. If the island formation occurs, the correlation function oscillates. *3D* and *2D* growth mode can occur on any surface, but the third mode which is the step flow growth mode can occur on miscut substrates only.

2.2.3 Step flow growth

In reality, a completely flat substrates is difficult to obtain. It is possible for the substrate to be miscut leading to a slightly tilted surface. In this work, the simulations are performed on miscut substrate which exhibit periodic series of terrace on the films. During growth process at high temperature, adatoms are mobile enough to move directly to step edge of terrace. It seems like the step propagates (flows along the terrace). The mode is *step flow (SF) growth*, which is shown in Figure 2.2(c). In the SF growth mode, there must be a relation between diffusion length ℓ_d and terrace width L_t. If the diffusion length ℓ_d is much less than terrace width L_t, most atoms are deposited on the terrace and form small clusters. The grown film will be very rough. On the other hand, if the diffusion length ℓ_d is more than the terrace width L_t, an adatom have more chance to meet step edge of terrace, so step flow growth mode occurs. Presence of step on substrate can make the growth process complicated. When an adatom descends down the steps on the surface, it has to cross a potential barrier in
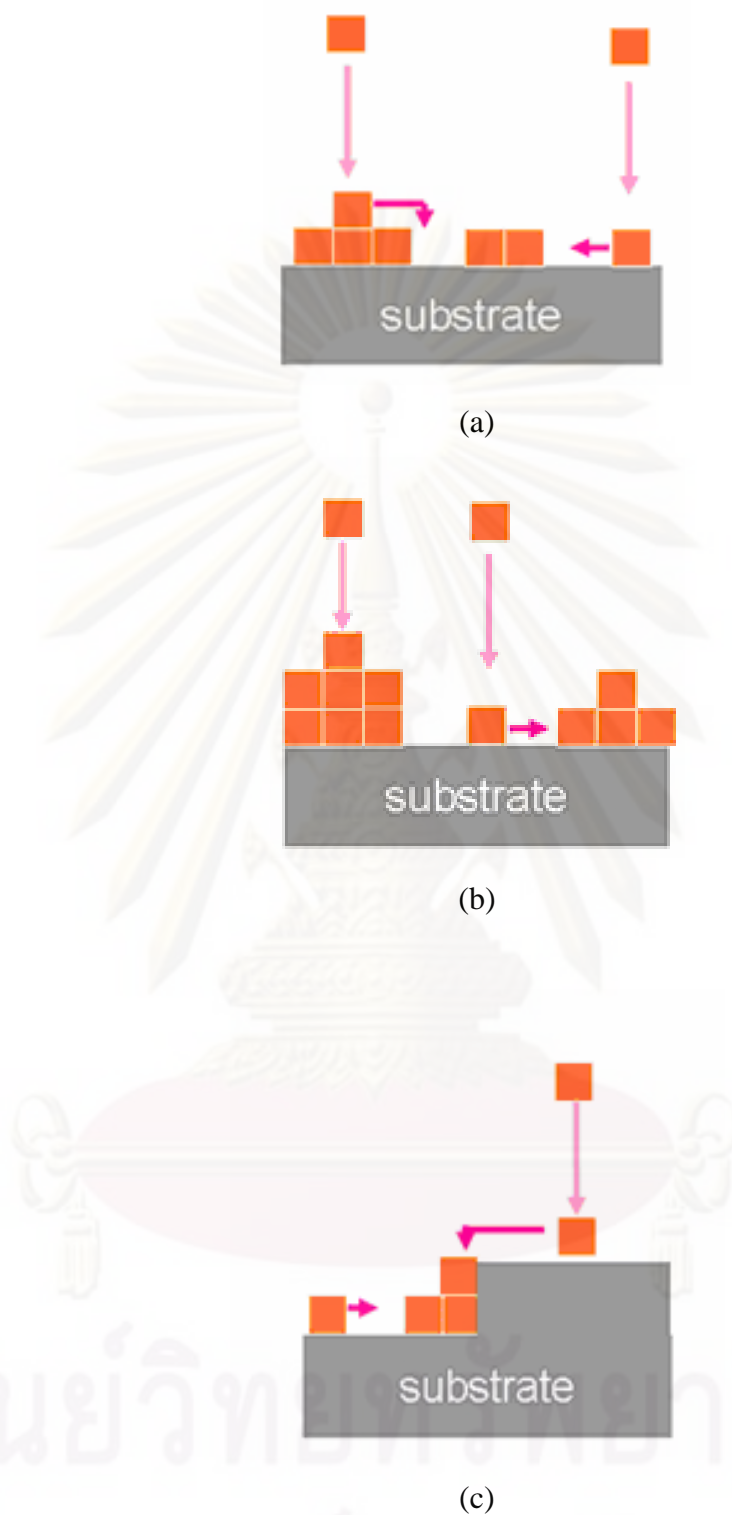
Figure 2.2: Illustration of thin film growth modes as (a) two-dimensional (*2D*) nucleation growth mode, (b) three-dimensional (*3D*) growth mode and (c) step flow (*SF*) growth mode.

t_1

$t_2 > t_1$

$t_3 > t_2$

$t_4 > t_3$

(a)   (b)
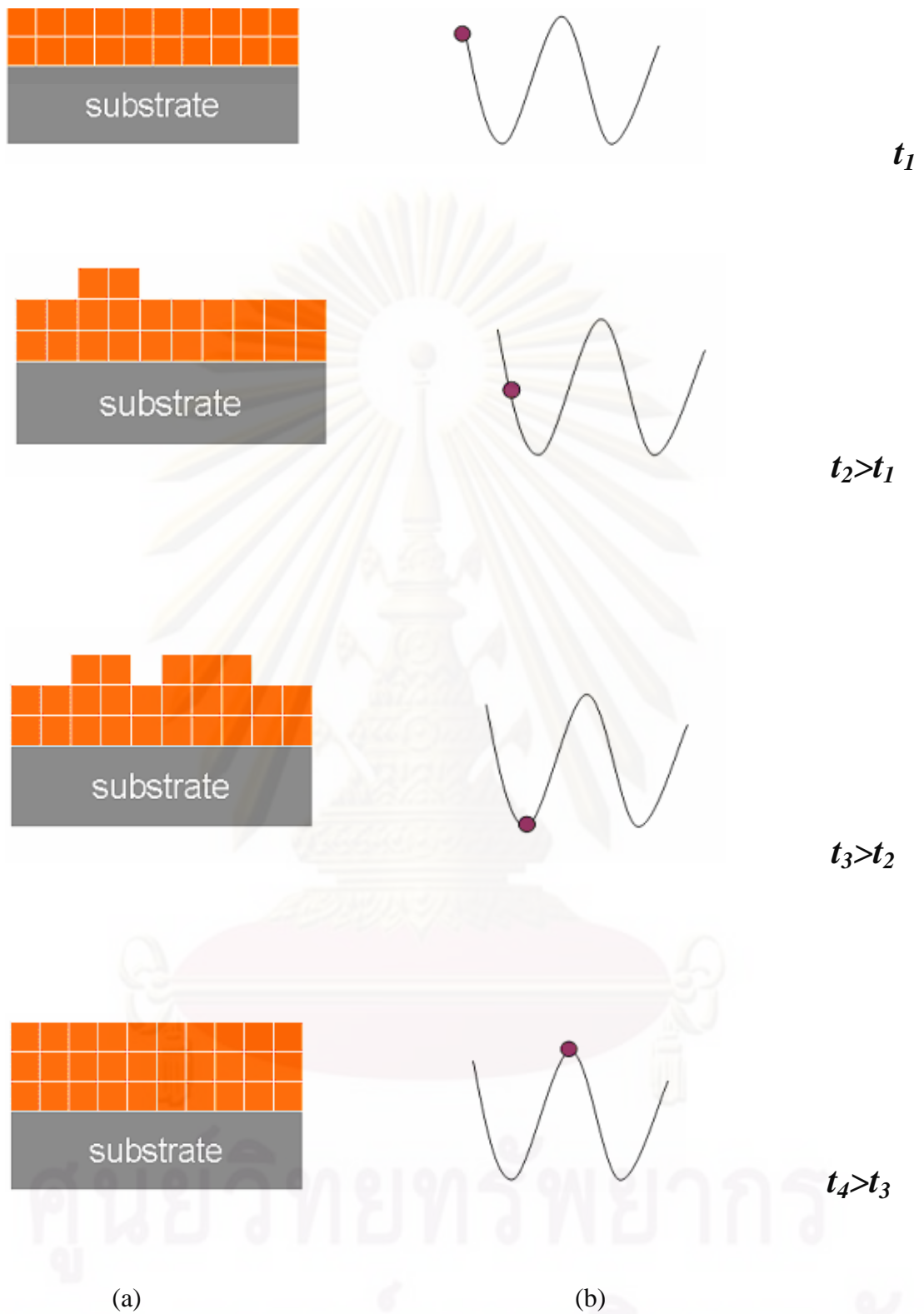
Figure 2.3: Relations between (a) the growing film and (b) reflection high-energy electron diffraction (RHEED) signal.

order to hop onto a different terrace. This potential barrier is called the *Ehrlich-Schwoebel (ES) barrier* (Ehrlich and Hudda, 1966; Felix Otto et al., 2005). The *ES* barrier is a step edge potential of an adatom that diffuses from upper to lower terrace. If the energy of an adatom is not higher than the step edge potential, it can not hop down to the lower terrace. It leads to island formation. In this work, the ES barrier is neglected. Moreover, in miscut substrates, there are sub-processes. If an atom leaves the step edge, this process is called *detachment* (Felix Otto et al., 2005); on the other hand , if it attaches to step edge, this process is called *attachment* (Felix Otto et al., 2005). These are sub-processes due to the presence of steps on a miscut substrate.

2.3   Model

Simulations are used to study microscopic processes in thin film growth because the growth conditions such as desorption and bulk vacancies/overhanging formation can be easily controlled. Growth models are created to imitate the real growth process. These include the detail motion of the deposited atom, subsequent movement on the surface, incorporation on steps or islands, island nucleation and desorption. In this work, a limited mobility growth model (toy model) is used. In a toy model, it follows these conditions - *SOS* constrains is satisfied, the deposition rate is constant and uniform for all sites and an adatom is allowed to diffuse within nearest neighbor sites. There are many toy models such as the Random deposition (RD) (Barabasi and Stanley, 1995), the Wolf-Villain (WV) (Wolf and Villain, 1990) model and the Das Sarma Tamborenea (DT) (Das Sarma and Tamberenea, 1991; Tamborenea and Das Sarma, 1993) model etc. In this work, the DT model is used to study growth on a miscut substrate.

2.3.1 Das Sarma -Tamborenea (DT) model

The Das Sarma Tamborenea (DT) model used in our simulation was introduced by S. Das Sarma and P.I. Tamborenea in 1991 (Das Sarma and Tamborenea, 1991; Tamborenea and Das Sarma, 1993). The model is under solid on

solid (*SOS*) constraints with a simple diffusion rule motivated by results from the Arrhenius hopping probability. From the Arrhenius hopping rate in Eq.(2.1), $R_{n \geq 2}$ are considered to be very small in the DT model. That is, it can be approximated that $R_{n \geq 2} \approx 0$. It is shown in Table 2.1. So when an atom in the DT model is dropped at a kink site(a site with at least 2 nearest-neighbor), it can not diffuse. However, if we consider the ratio $R_{n=2}/R_{n=1}$, we obtain

$$R_{n=2}/R_{n=1} = \exp\left(\frac{-E_B}{k_B T}\right) . \tag{2.3}$$

The ratio $R_{n=2}/R_{n=1}$ depends on the substrate temperature which means T. At low temperature, $R_{n=2}/R_{n=1}$ is very small so most atoms tend to remain at the kink sites. This condition is consistent with the diffusion rules of the DT model which generates films with similar statistical properties to films grown by low temperature MBE technique. The diffusion rule of the DT model is shown in figure 2.4(a). On the other hand, when the temperature is high, $R_{n=2}/R_{n=1}$ is not small enough and R_2 should not be neglected. It is shown in Table 2.1. Since we want to increase the mobility of adatoms, which equivalents higher temperature growth in experiments, we modified the DT diffusion rules by allowing diffusion of a surface atom with $n = 2$ and set $R_{n \geq 3} = 0$. It means that atom can mobile to leave the kink sites, so the most atom tend to remain the *trapping site* (a site with at least 3 nearest-neighbor bonds). The diffusion rule of the modified DT model is shown schematically in figure 2.4(b) for the case when $\ell_d = 1$. From figure 2.4(b), as atoms are randomly dropped on the surface, they can diffuse before being incorporated to the film. If an atom is dropped on a site that provides at least three bonds which is called a trapping site (atom A), it can not diffuse at all. On the other hand, an atom dropped on a site with less than three bonds (atom B, C and D) can diffuse on the surface. During diffusion, if one of the nearest neighbors is a trapping site, atom moves to this site (atom B), otherwise atom diffuses randomly (atom C and D). However, the modified DT model is still a limited mobility growth model in which an adatom is allowed to diffuse within its nearest-neighbor sites only. To further increase the mobility of the atom, long surface

R_n	$T_1 = 500K$	$T_2 = 900K$
$R_{n=1}$	1.64	1.97×10^6
$R_{n=2}$	1.55×10^{-3}	4.95
$R_{n=3}$	1.47×10^{-6}	1.24×10^{-5}
$R_{n=4}$	1.39×10^{-9}	3.12×10^{-11}
$R_{n=5}$	1.32×10^{-12}	7.86×10^{-17}

Table 2.1: Diffusion rate of diffusing atoms which is a function of number of nearest-neighbor bonds and the substrate temperature.

diffusion length noise reduction technique (*NRT*) (Chatraphorn and Das Sarma, 2002) is added to the modified DT model. Increasing diffusion length ($\ell_d > 1$) in simulations is equivalent to increasing substrate temperature in experiments.
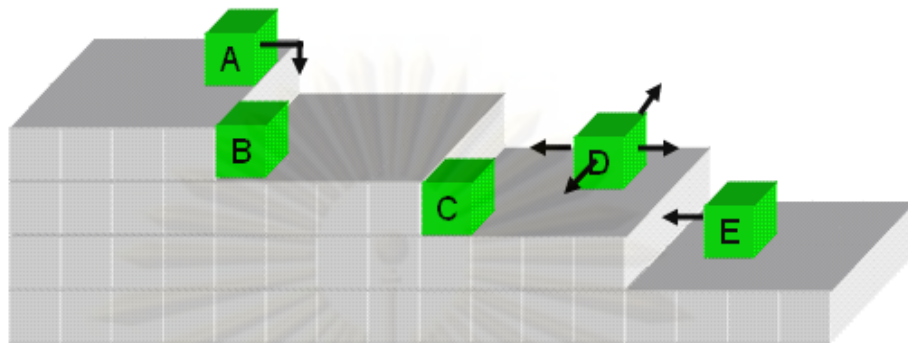
(a)
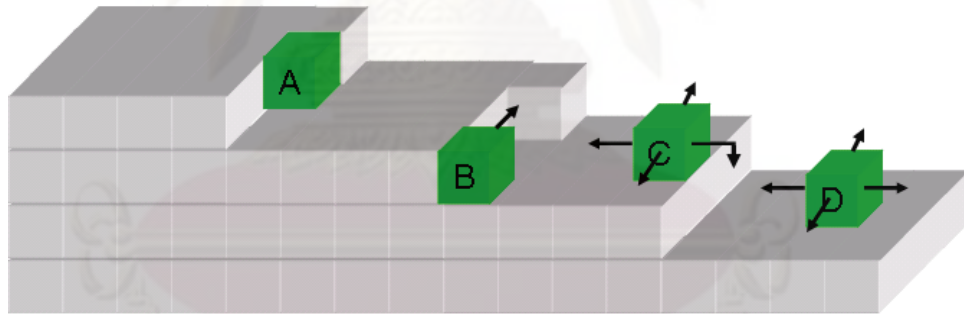
(b)

Figure 2.4: Illustration of diffusion rule in (a) the DT model and (b) the modified DT model on a two dimensional miscut substrate.

2.3.2 Long surface diffusion length noise reduction technique

In diffusion rule of original DT model, an adatom can diffuse to a nearest neighbor sites ($\ell_d = 1$). In reality, however, an adatom can diffuse to a site that is further away. Long surface diffusion length noise reduction technique *NRT* is added to the original DT model. This technique is used to reduce noise which causes roughness of film. To do this, the diffusion length of an adatom is increased to be greater than one ($\ell_d > 1$), which means adatoms have more chance to meet neighboring sites according diffusion rule of the original DT and the modified DT model. This technique helps to generate layer by layer growth mode in the DT and the modified DT model because adatoms can move far enough to form a completely layer. Fronting (sliding) of steps which is step flow growth appears. Although, long surface diffusion is a simulation technique, it connects to substrate temperature in experiments. This is $\ell_d \propto \exp\left(\dfrac{-E_0}{k_B T}\right)$ (Barabasi and Stanley, 1995). The exponent $\gamma (\approx \dfrac{1}{6}$ to $\dfrac{1}{2})$ depends explicitly on the substrate temperature dependent minimum stable island size and can be calculated by stochastic Monte Carlo simulations, E_0 is the activation energy of a free atom, k_B is the Boltzmann constant and T is the substrate temperature. From this equation, increasing diffusion length in simulations is equivalent to increasing substrate temperature in experiments. In NRT technique, there is a parameter ℓ which is the maximum number of times an atom can hop to find the final site under the diffusion rule of the model. For example, in the DT model, if we set the parameter $\ell = 3$, atom can try to find a kink site by hopping three times. We need to know the real distance an atom travels within ℓ hops. This is discussed next.

2.3.3 Random walk

For simple random walk (*RW*) theory, the walker goes from one site to another in d dimensions. A random walk has unity probability of reaching any point as the number of steps approaches infinity. The end-to-end distance is the number of

steps N which is oriented in arbitrary direction. It follows as (Landau and Binder, 2000),

$$\left\langle R^2(N) \right\rangle \approx N^{\frac{1}{2}}. \tag{2.4}$$

From Eq. 2.4, the end to end distance is proportioned to (number of step)$^{1/2}$. If an atom diffuses randomly ℓ times. The distance ℓ_d that an atom can diffuse is $\ell_d \approx \ell^{\frac{1}{2}}$. In this work, the distance ℓ_d is the diffusion length.
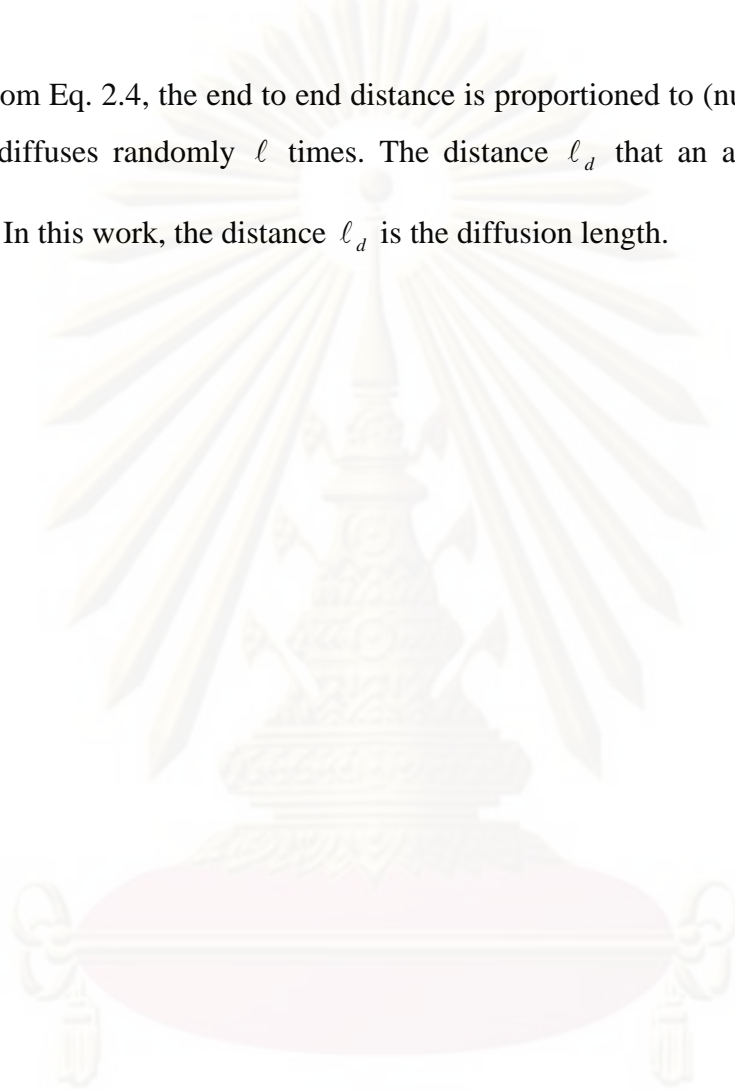
CHAPTER III

Quantities of Interest

In order to understand the quality of the grown films, statistical quantities are used. The quantities of interest in this work are the morphology, the interface width, the correlation functions and the persistence probability. The interface width is used to study the roughness of the film, an information about the characteristic feature and the roughness of the film are given by the correlation functions, and the persistence probability is used to study how long the initial pattern of the film can be maintained.

3.1 Morphology

The film morphology is the first quantity that can be observed from thin film simulations. It shows the general quality of the film such as the roughness/smoothness and shape of the film in macroscopic scale. The morphology of film many look smooth when in fact it is not really smooth. To have a more detailed and complete information on the grown film, statistical quantities are calculated.

3.2 Interface width

The interface width (W) characterizes the roughness of films in the microscopic scale. It is defined as the root mean square height fluctuation of the film surface (Barabasi and Stanley, 1995)

$$W(L,t) = \left\langle \left[\frac{1}{L^2} \sum_{x,y=1}^{L} \left[h((x,y),t) - \langle h \rangle \right]^2 \right]^{\frac{1}{2}} \right\rangle \qquad (3.1)$$

where $h((x,y),t)$ is the surface height of column x and column y at growth time t, the growth time t in the simulation is measured by the average number of deposit layers, d is the dimension of the substrate, L is the substrate size, the bracket $\langle....\rangle$ denotes the ensemble average over many growth simulation to represent many substrates, and $\langle h \rangle$ is the averaged height of the surface or the average film thickness,

$$\langle h \rangle = \frac{1}{L^2} \sum_{x,y=1}^{L} h((x,y),t) . \tag{3.2}$$

In this work, the deposition rate, which is defined as the average number of deposit layers per unit time, is constant. So the average height increases linearly with time,

$$\langle h \rangle \propto t . \tag{3.3}$$

The interface width depends on the substrate size L and growth time t. In the early growth time, the interface width has power law dependence with growth time t,

$$W \propto t^{\beta} \qquad \text{when} \quad t << L^z \tag{3.4}$$

where β is the growth exponent. This region is called growth regime. For a long times, the interface width coverges into constant value or saturate and also has power law dependence with substrate size L,

$$W_{sat} \propto L^{\alpha} \qquad \text{when } t >> L^z \tag{3.5}$$

where α is roughness exponent, which can be obtained from this region. This region is called the saturation region because interface width becomes saturated. Both regions is shown in figure 3.1. The growth time that changes from the growth region to the saturate region is the saturation time (or crossover time, t_s), which depends on the substrate size,

$$t_s \propto L^z \tag{3.6}$$

z is called the dynamic exponent, which relates to β and α,

$$z = \frac{\alpha}{\beta} \ .$$
(3.7)

This equation is called scaling law linking the three exponents α, β, z are called *simulated critical exponents* (Barabasi and Stanley, 1995), which determine the universality class of the model. If different models have the same set of critical exponents, they are in the same universality class. Experimental studies can directly measured roughness by the STM or AFM spectroscopy and theoretical studies calculate the critical exponents from continuum equation (Das Sarma, 1996).

Interface width is the standard deviation of surface height so its value depends on information from the whole film surface. This is called global scaling behavior.

3.3   Correlation functions

The local scaling behavior of the film surface is studied by height-height correlation functions. The height-height correlation functions are functions that characterize the relation between two sites on the surface. Generally, the functions can distinguish a mound or island formation from a kinetically rough (Punyindu, 2000; Chatraphorn et al., 2001). The functions can provide information about the specific feature on film surfaces. Critical exponents can also be extracted from correlation function plots. The height-height correlation functions have two different definitions. At fixed *t*, the first definition is defined in one dimensional substrate growth as

$$G(r_x) = \left\langle \left[\frac{1}{L} \sum_{i=1}^{L} [h(x+r_x) - h(x)]^2 \right]^{1/2} \right\rangle$$
(3.8)
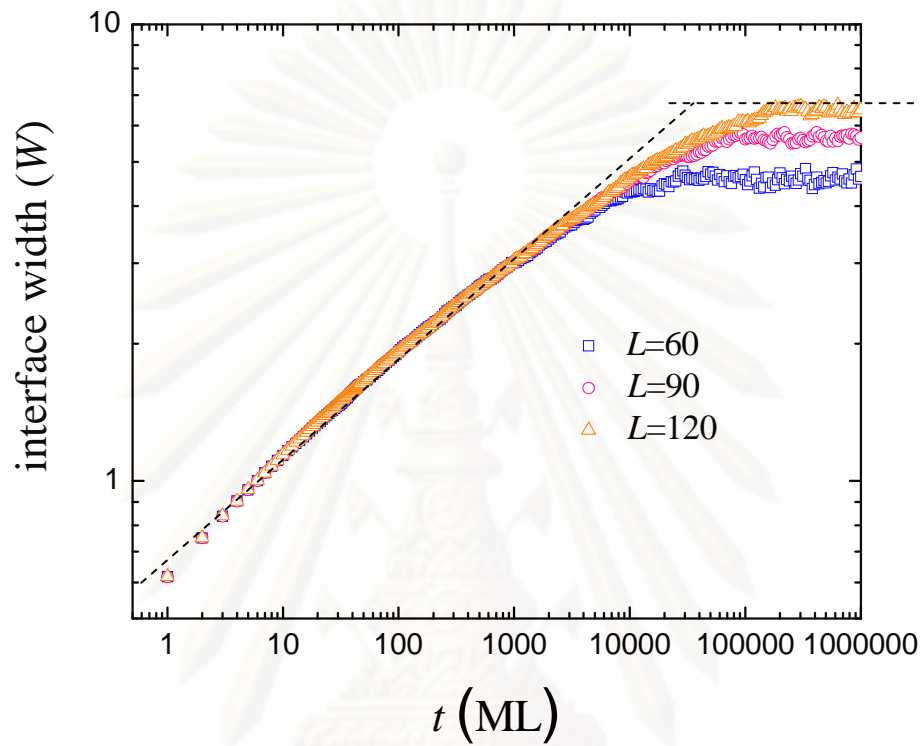
Figure 3.1: *W-t* plot on the two dimensional miscut substrate (the substrate size is 60, 90 and 120 and miscut angle $\theta = 2^o$).

where r is the distance between two sites on the surface, $h(x)$ is the deviation of the surface height from the average height (or height fluctuation) of column x, L is the substrate size, and $\langle ... \rangle$, denotes the ensemble average over many growth simulations using different random numbers for the deposition.

In this work, the simulations are done on two dimensional miscut substrates so the correlation functions are calculated along two independent directions. In the x direction or the tilted direction,

$$G(r_x) = \left\langle \left[\frac{1}{L^2} \sum_{x,y=1}^{L} [h(x+r_x, y) - h(x,y)]^2 \right]^{\frac{1}{2}} \right\rangle . \tag{3.9}$$

In the y direction or the notilted direction,

$$G(r_y) = \left\langle \left[\frac{1}{L^2} \sum_{x,y=1}^{L} [h(x, y+r_y) - h(x,y)]^2 \right]^{\frac{1}{2}} \right\rangle \tag{3.10}$$

where r_x and r_y is the distance between two sites on the surface in the x and y direction respectively.

In the thin film growth processes of original DT model, an adatoms are randomly dropped on the substrate. They can diffuse to a nearest neighbor site if the site provides a lateral bond with a neighboring site. Thus the height of the adatoms is equal to or larger than its neighbors. The height fluctuation expands laterally and there is a mechanism along the surface. So the height of surface are not absolutely independent, but the height of neighbors' sites are correlate. The length which they are correlated, is called *lateral correlation length* ξ. At fixed t, the correlation function $G(r)$ and the lateral correlation length ξ have a power law, which is described by

$$G(r) \quad \propto \quad r^\alpha \qquad\qquad r \ll \xi \tag{3.11}$$

$$G(r) \quad \propto \quad const \qquad\qquad r \geq \xi \tag{3.12}$$

where β is growth exponent and α is roughness exponent. At small r, correlation function has power-law dependence with the distance between two sites on the surface r. At large r, the correlation function coverges into constant value. If the constant value depends on the growth time t, the correlation function G_{sat} is described by

$$G_{sat} \quad \propto \quad t^{\beta} \qquad\qquad r \geq \xi. \qquad (3.13)$$

The roughness exponent can be obtained by either W-t plot and G-r plot at small r. For the DT growth model, this model exhibits anomalous scaling behavior (Chatraphorn, 2000), which makes the difference between the local and global scaling behavior. The difference is roughness exponent which is obtained from the W-t plot, is not equivalent from the G-r plot. If r is varied and t is fixed, the function shows the correlation of the height of two sites on the surface. On the other hand, t is varied and r is fixed, the function can show roughness of the film surface. Typically, the G-r plot has two regions which is shown in figure 3.2. In the first region $r < \xi$, the function depends on r. It shows that the height of sites still correlate in this region for the early time. If the correlation length is much, this region is longer. In the second region $r \approx \xi$, the function saturates. However, the function provides information about the periodic feature of film surface.

The second definitions of the height-height correlation function (Chatraphorn et al., 2000; Das Sarma and Chatraphorn, 2000) have also two functions. In the x direction or the tilted direction,

$$H(r_x) = \left\langle \frac{1}{L^2} \sum_{x,y=1}^{L} h(x,y)h(x+r_x,y) \right\rangle. \qquad (3.13)$$

In the y direction or the notilted direction,

$$H(r_y) = \left\langle \frac{1}{L^2} \sum_{x,y=1}^{L} h(x,y)h(x,y+r_y) \right\rangle \qquad (3.14)$$

where $\langle...\rangle$ denotes the ensemble average (over many growth simulation), *h(x,y)* is the deviation of the surface height from the average height (or height fluctuation) of column *x,y* and *r* is the distance between two sites on the surface.

From the definition, the function approaches its maximum when the two points have the same height. If the $H(r) \propto r$ oscillates, it identifies as mound or island formation on the surface. However, there is not only island formation. There are many feature of film. The function can provide information about the specific periodic feature of the film surface. The function is observed that the $H(r) \propto r$ repeats the periodic pattern of the film surface. In this work, the substrate is miscut substrate that looks like terrace. The $H(r) \propto r$ repeats every step of terrace. If the film surface can keep its original periodic pattern, the $H(r) \propto r$ is the same for a long time. However, the film surface is damaged, the curve of $H(r) \propto r$ changes.

3.4   Persistence probability

When the thin film growth processes continue with specific feature, the pattern of the substrate is deformed during the growth process. The quantity which is used to study how long the original film pattern can persist through growth time, is *persistence probability*. It is a quantity that remaining gives information on ratio of the pattern on the grown films through time t and is defined as (Kallabis and Wolf, 1997)

$$P(t) = \left\langle \frac{1}{L^d} \sum_{x,y=1}^{L} \left[\prod_{s=1}^{t} \delta_{h((x,y)s),h((x,y)0)+s} \right] \right\rangle \tag{3.15}$$

where *h((x,y),s)* is the surface height of column *x* and column *y* at time *s*, *h((x,y),0)* is the surface height at growth time *t*=0 or the original film pattern, δ is the Kronecker delta function $\delta_{ij} = 1$ where i=j and 0 otherwise), Π is a production over time *s*, *d* is
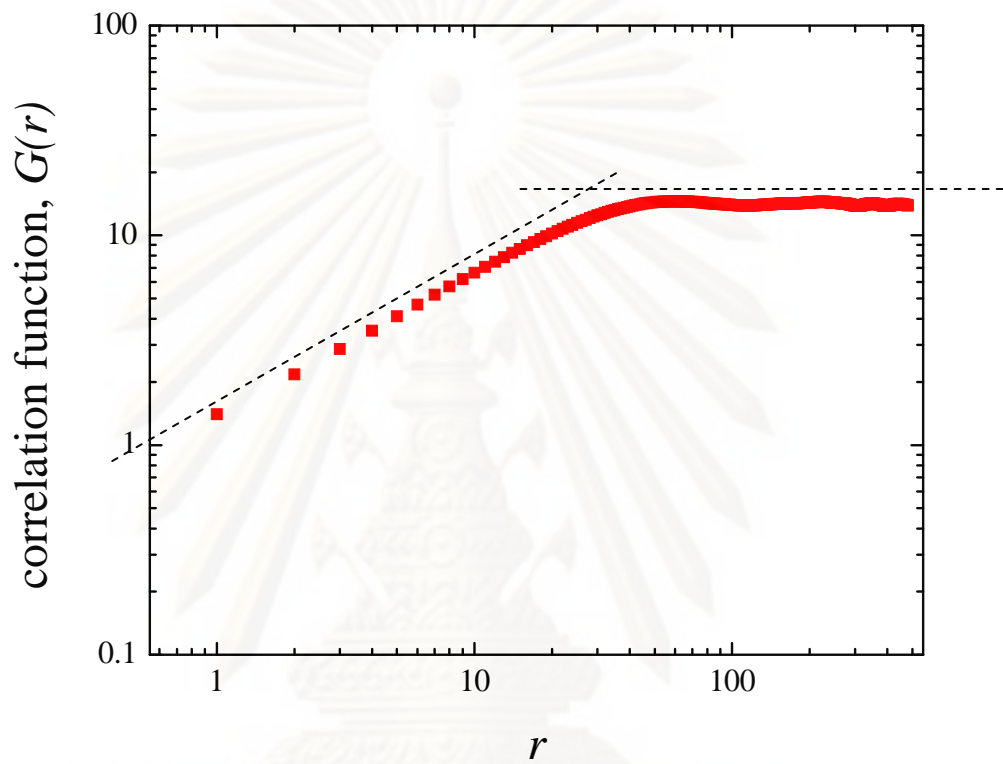
Figure3.2: *G-r* plot on the two dimensional miscut substrate (the substrate size is 960×960 and miscut angle $\theta = 2^{\circ}$) at 10^5 ML.

the dimension of the substrate, L is the substrate size, and the $\langle ... \rangle$, denotes the ensemble average (over many growth simulation)

By definition, the persistence probability is a function of every monolayers t, which starts $t=0$ until the film is grown to t MLs. If the entire sites of film can keep perfectly the original pattern substrate at all time, the persistence probability $P(t)=1$. During ($t=0,...,s$), if the some sites deform with the growth time, the persistence probability becomes zero at this site. Deforming of original pattern film indicates that the original pattern film is damaged. If the surface film is slowly damaged, the persistence probability slowly decreases from $P(t)=1$. On the other hand, if the surface film badly damaged, the persistence probability rapidly decreases from $P(t)=1$.

CHAPTER IV

RESULTS AND DISCUSSION

In this chapter, the simulated results are presented and discussed. The results are divided into two parts. In the first part, effects of the miscut substrate on films grown by the DT model are studied. In this part, we start by using the original DT model. Then we add long surface diffusion length noise reduction technique (NRT) (Chatraphorn and Das Sarma, 2002) to the original DT model to see effects of the substrate temperature. For modeling, we modify the diffusion rule of the DT model in order to increase atomic mobility. The NRT is added on the modified DT model as well. In the second part, we present effects of the initial configuration of the miscut substrate by varying the tilt angle. The goal of this part is to find the optimum conditions for thin film growth on the miscut substrate.

In our work, all simulations are done on a two dimensional substrate with a slightly tilted surface as shown in figure 4.1. The substrate is in the xy plane and the film is grown in the z direction. A simulated thin film growth on the two dimensional substrate can be compared with results from experimental laboratories. It is more realistic than a simulation done on a one dimensional substrate. The substrate size used in this work is $L \times L = 960 \times 960$ sites. Results are ensemble averaged over 10 different runs. From figure 4.1, surface of the miscut substrate looks like terrace. The width of terrace is L_t and the step height of terrace is s. Periodic boundary condition is used in the y direction which does not have any tilt. In the tilt direction (along the x axis), the periodic boundary condition is adjusted to reflect the tilt and is called peculiar condition (Xiao, 1997). Interested quantities are simulated morphology, interface width, persistence probability and correlation functions.
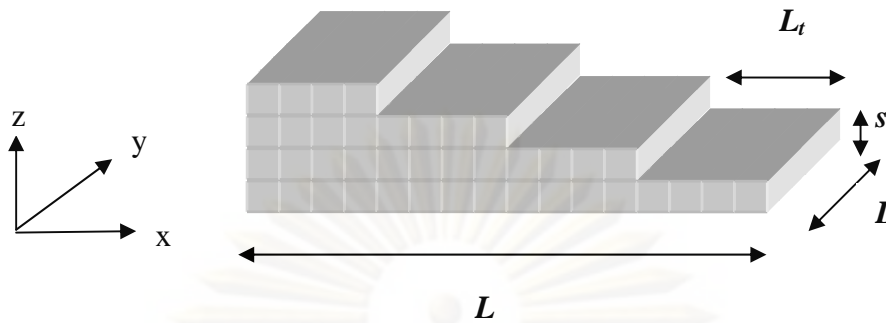
Figure 4.1: The miscut substrate (the substrate size is $L \times L$, the terrace width is L_t and the step height is s).

4.1 Effects of miscut substrate on films grown on substrates with the same initial angle.

In this section, the initial miscut angle ϑ is fixed at $2°$ which agrees with standard miscut specifications for GaAs substrate (Neave et al., 1985). Since the miscut angle is $\theta = \tan^{-1}(s/L_t)$, we choose the width of each terrace L_t to be 30 sites and the terrace height s to be 1. This is called mono-stepped height. Conventionally, most simulations (Pal and Landau, 1996) have been done on mono-stepped height.

4.1.1 Results from the original DT model

4.1.1.1 Interface width

The information on roughness of the surface is given by the interface width W. We plot the interface width with the growth time ($W - t$ plot) and the result is shown in figure 4.2. The curve of the interface width is founded to be separated into three

regions. In the first region ($t < 1$ ML), the slope, which is the growth exponent β, is approximately $\beta_I \approx 0.49$. It corresponds with the growth exponent of the random deposition (RD) model (Barabasi and Stanley, 1995) which is a model that does not include a surface diffusion process. This is understandable because there are only small amount of deposited atoms on the surface in the beginning so a deposited atom has only a very small chance to meet a neighboring site that it can diffuse to according to the diffusion rule of the DT model. So the diffusion process of atoms has very small influence on the growth process at these time intervals. This explains why the growth exponent of the DT model in this region is the same as the growth exponent of the RD model. In the second region ($t > 1$ML), the growth exponent decreases from $\beta = 0.24$ to $\beta = 0.19$. The growth exponent in this region is represented by β_{II} and β_{III} which are shown in figure 4.2 This result agrees with the original DT model on a flat substrate (Das Sarma and Tamberenea, 1991; Das Sarma and Punyindu, 1997). It can be explained that tilt of substrate is damaged in the early time as shown in figure 4.3 This is because, the diffusion length is so small ($\ell_d = 1$) compared with the terrace width. So, the tilted of substrate does not affect for the original DT model. This is cause, the value of β agrees with the original DT model on a flat substrate.

4.1.1.2 Morphologies

Film morphologies of the original DT model simulated on miscut substrates are shown in figure 4.3. Each morphology is a segment of 120×120 sites from the entire substrate (960×960 sites). We found that the roughness of the film surface increases rapidly as the growth time is increased from 1 to 100 monolayers. From these morphologies, it is clear that the initial structure of the tilted substrate is lost from the early time. We can explain that this happens because newly deposited atoms can diffuse only to one of its nearest neighboring sites due to the restrictions in the diffusion rules of the model. This means the mobility of atoms is quite low for the original DT model. So the roughness of film greatly increases with the growth time. This damages the initial pattern on the substrate from the early growth time.
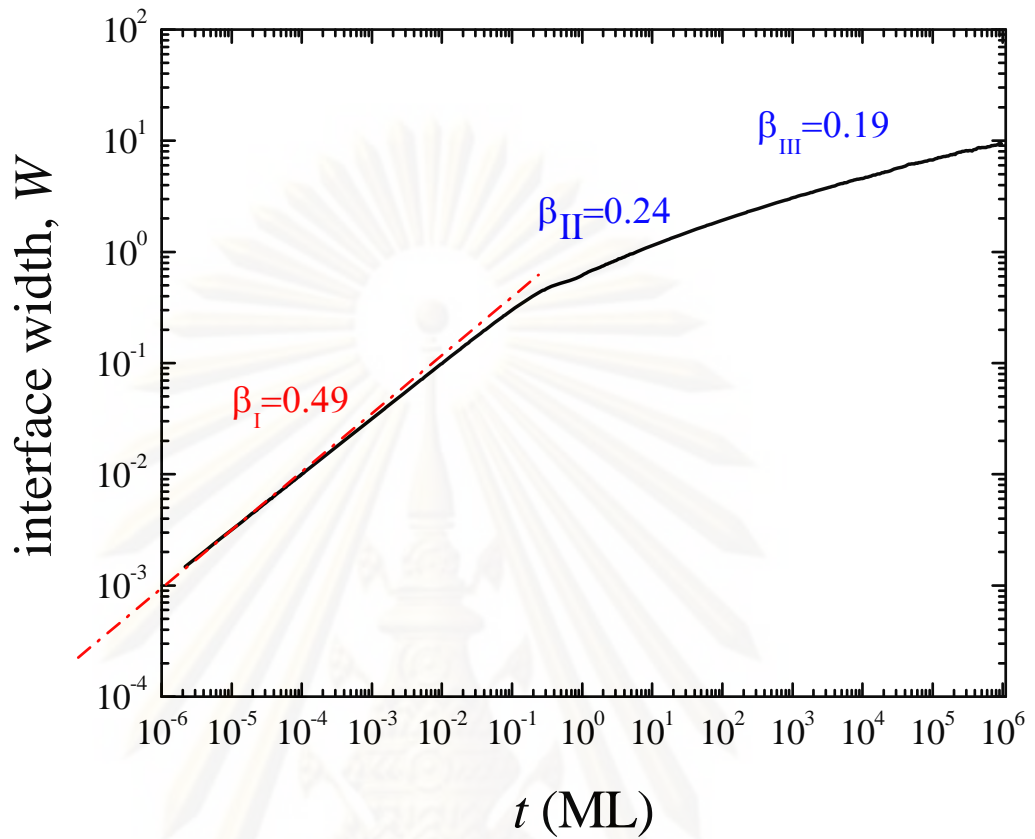
Figure 4.2: *W-t* plot of the original DT model, $L \times L = 960 \times 960$ with initial miscut angle $\theta = 2^o$.
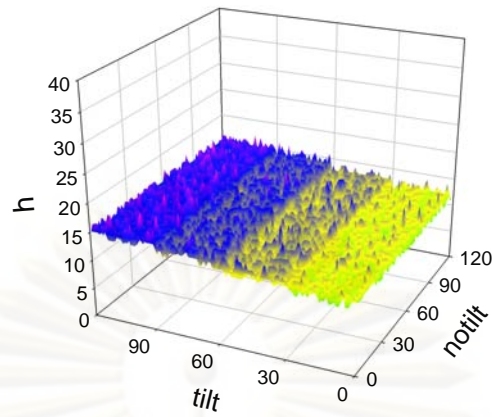
(a) $t = 1$ ML
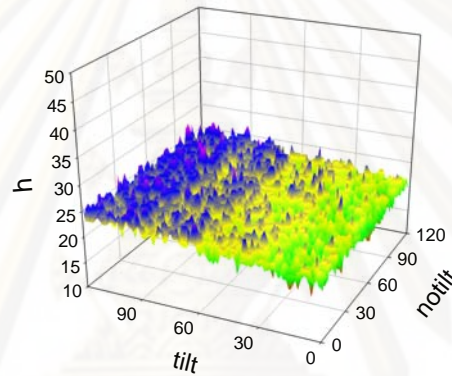
(b) $t = 10$ ML
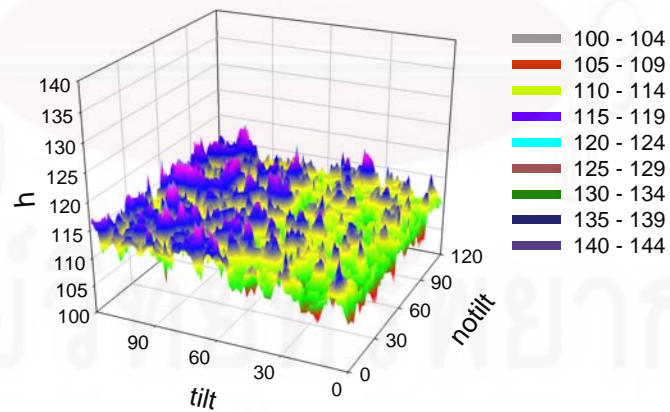
(c) $t = 100$ ML

Figure 4.3: Film morphologies of the original DT model with $\ell_d = 1$ and $L \times L = 960 \times 960$ (a segment of 120×120) at t equals to a) 1 ML, b) 10 ML and c) 100ML.

4.1.1.3 Persistence probability

From the morphologies in figure 4.3, we found that the film is quite rough and can not keep the original pattern. The cause for this is that the atom mobility is too small so most atoms that are deposited on the terrace form small clusters on the terrace instead of diffusing. The result is that the original tilted pattern is destroyed from the early time. We want to investigate how long the film can maintain the initial pattern on its substrate. The quantity that gives this information is the persistence probability $P(t)$. The persistence probability versus the growth time, or $P - t$, plot is shown in figure 4.4. We found that the $P - t$ curve decreases rapidly. The initial pattern on the substrate is totally destroyed within only 20 MLs. In order to keep the original pattern for a long time, long surface diffusion length noise reduction technique (NRT) is used. The results of DT model with NRT will be presented in the next section.

4.1.1.4 Correlation function

The correlation functions are presented in this part. There are two different definitions for correlation functions. The first one is

$$G(r_x, t) = \left\langle \left[1/L^2 \sum_{x,y=1}^{L} [h(x + r_x, y) - h(x, y)]^2 \right]^{\frac{1}{2}} \right\rangle \text{ and the second one is}$$

$$H(r_x, t) = \left\langle \left[1/L^2 \sum_{x,y=1}^{L} [h(x + r_x, y)h(x, y)] \right] \right\rangle. \text{ Both are along the } x \text{ direction. In the } y$$

direction, there are $G(r_y, t)$ and $H(r_y, t)$. The correlation functions depend on the growth time t and distance r which is the distance between two sites on the surface. First, we plot $G(r)$ versus the distance r or $G - r$ plot. This is shown in figure 4.5. In this work, the simulations are done on two dimensional substrates so the correlation functions are calculated along two directions. Figure 4.5(a) shows $G - r$ plot in the tilted direction ($x - axis$). On the other hand, figure 4.5(b) shows the $G - r$ plot in the notilted direction ($y - axis$). From the definition of $G(r)$, if r is varied and t is fixed, the correlation between two sites on the surface is obtained. From the graph, we

found that as r increases, the value of $G(r)$ increases linearly. This means $G(r)$ has a power law dependence with r when r is small. At large r, the value of correlation function coverges into a constant which means $G(r)$ saturates for large r. This happens when r approximately equals the correlation length $\xi (\sim t^{1/z})$, where z is the dynamical exponent which describes the approach to the steady state associated with growth with lateral correlations (Dharmadhikari et al., 1999; Das Sarma and Chatraphorn 2000). By plotting the correlation function at different growth times, the saturation of correlation function in both directions occur quickly at small t which is shown in figure 4.5(a). For example, at 1 ML, $G(r)$ saturates at $r \approx 3$ sites only. This implies that the diffusion process is so small in the early time, so the correlation length is short. But at a large time (10^6 MLs), the correlation length is larger because there is more time for the diffusion process. So, the correlation function saturates at large r. Results of $G(r)$ plot in the tilt direction is the same as in the notilted direction.

Since we are interested in the specific pattern of the growth film, we investigate the second correlation function $H(r)$ (Das Sarma and Chatraphorn, 1999; Chatraphorn, 2000; Das Sarma and Chatraphorn 2000). This correlation function can reveal information of a specific feature on the film. We plot $H(r)$ as a function r, which is shown in figure 4.6. From the graphs of $H(r)$ calculated from both directions, the value of $H(r)$ is maximum at $r = 0$ as it should be by definition. It is also obvious that as t increases, this maximum of $H(r)$ should increases as seen in the graphs. As r increases, $H(r)$ decreases to almost zero for all r in both directions. This means that we do not have any specific feature on the film surface. However, at a large time ($t=10^5$-10^6 MLs), the curve of $H(r)$ slightly oscillates. This shows that there is island formation on the film surface at large t.

The results from both $G(r)$ and $H(r)$ imply the same things. That is, the correlation functions in the tilted direction (figures 4.5(a) and 4.6(a)) are the same as in the notilted direction (figures 4.5(b) and 4.6(b)). This is because films grown by the original DT model are so rough and the initial tilt in the substrate is lost very quickly. In order to see effects of the tilt, we need to increase atom mobility. So in the next section, we present results from our modified DT model with long surface diffusion length NRT.
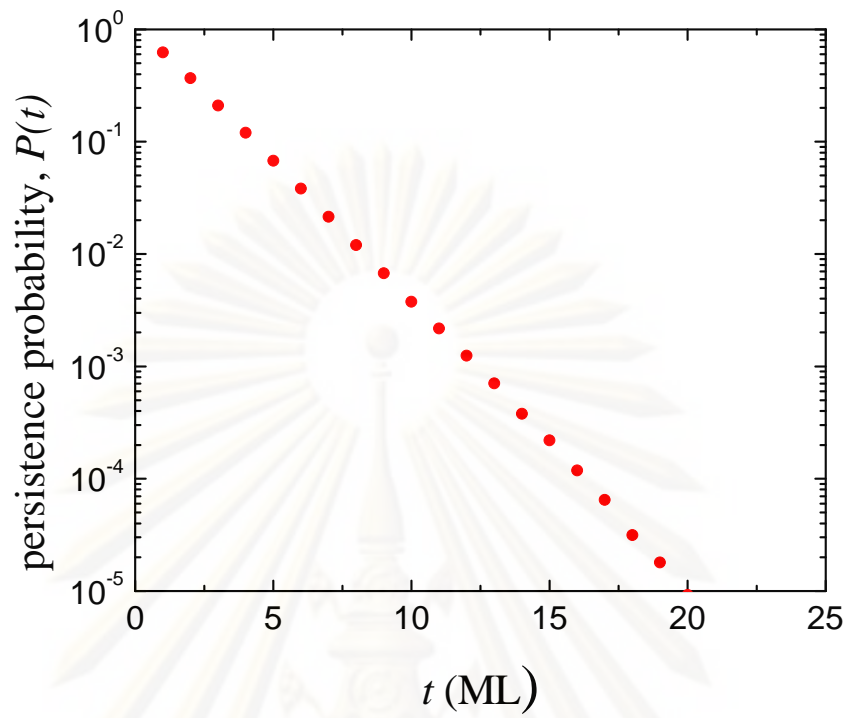
Figure 4.4: *P-t* plot of the original DT model ($\ell_d = 1$), $L \times L = 960 \times 960$ with initial miscut angle $\theta = 2^o$.
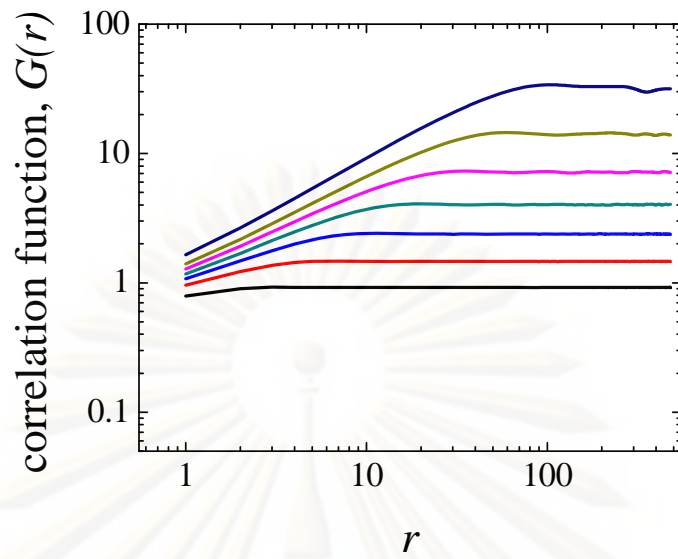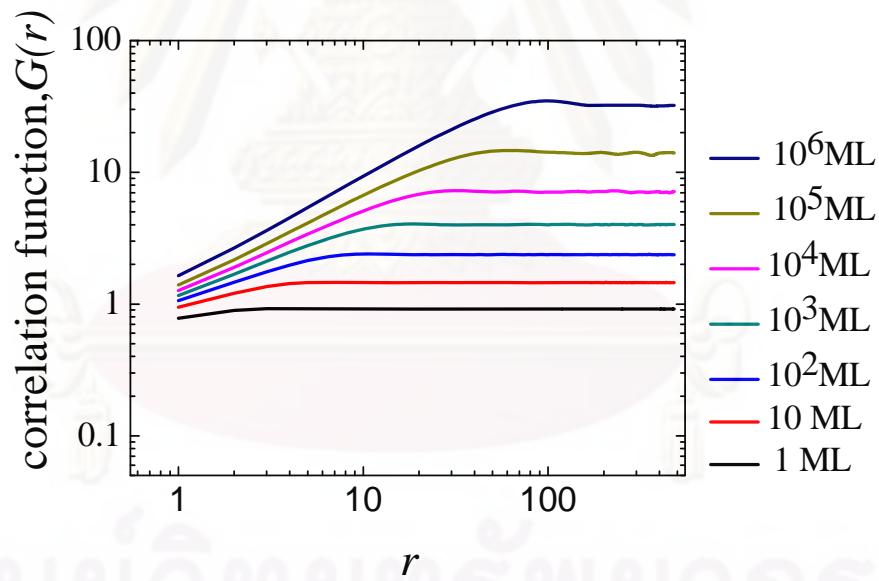
(a) tilted direction

(b) notilted direction

Figure 4.5: *G-r* plot of the original DT model ($\ell_d = 1$) (a) tilt direction and (b) notilt direction as the growth time is increased from $t = 1, 10, 10^2, 10^3, 10^4, 10^5$ and 10^6 MLs.
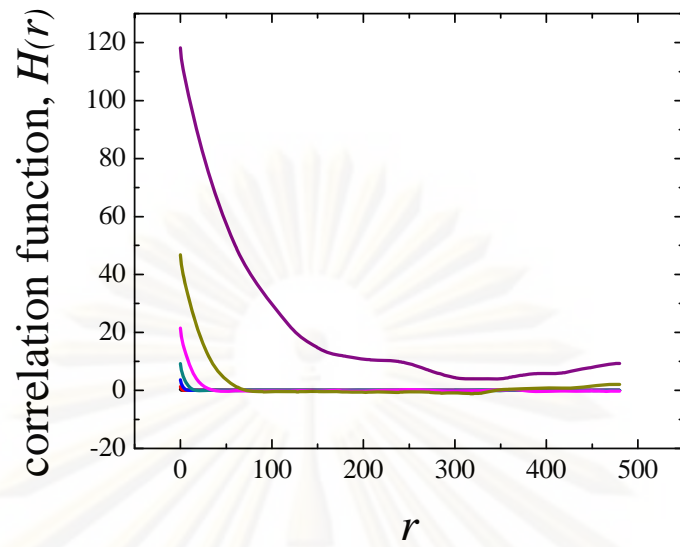
(a) tilted direction
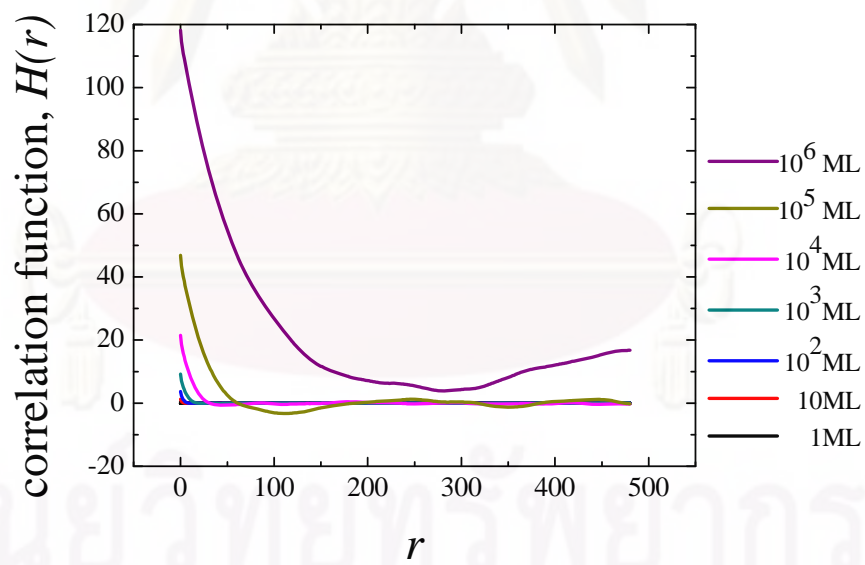
(b) notilted direction

Figure 4.6: *H-r* plot of the original DT model ($\ell_d = 1$) (a) tilt direction and (b) notilt direction as the growth time is increased from $t = 1, 10, 10^2, 10^3, 10^4, 10^5$ and 10^6 MLs.

4.1.2 Results from the DT model with long surface diffusion length

In the previous section, we found that the roughness increases with time for the original DT model because atom can only diffuse to one of its nearest neighboring sites, i.e. diffusion length $\ell_d = 1$. In order to reduce the film roughness, long surface diffusion length noise reduction technique (NRT) is added to the original DT model. The noise reduction technique has been used to produce smooth films grown in the layer by layer growth mode in many models, such as the Eden model (Wolf and Kertesz, 1987), the single step model (Tang, 1992), and the WV and DT model in 1+1 dimensions (Wolf, 1995). This is done by increasing the diffusion length of diffusing atoms. Increasing the diffusion length in simulations is equivalent to increasing the substrate temperature in experiments (Chatraphorn and Das Sarma, 2002). In the NRT, there is a parameter ℓ which is the maximum number of times an atom can hop to find the final site under diffusion rules of the model. According to random walk theory, the end to end distance is proportioned to (number of step)$^{1/2}$ (Landau and Binder, 2000). So, the length that an atom can diffuse (or diffusion length ℓ_d) is $\ell_d \approx \sqrt{\ell}$. Surface diffusion length ℓ_d is defined as the distance between the deposition site and the final incorporation site. In this work, ℓ is varied in simulation in order to change mobility of the moving atoms. However, $\ell_d \approx \sqrt{\ell}$ is specified in the results shown because it is move meaningful in physics.

4.1.2.1 Interface width

From $W - t$ plots shown in figure 4.7, the diffusion length is increased from $\ell_d = 1$ to $\ell_d = 90$ sites. For $\ell_d = 1$, we are back to the original DT model. As NRT is added, atoms can move further from the deposition sites so they have more chance to be incorporated at an edge of existed clusters instead of forming new clusters which leads to roughness of the surface. From the graph, when $\ell_d > 1$, we found that the interface width oscillates as shown in figure 4.7. $W - t$ oscillation appears at large diffusion length (or high temperature). This indicates that it is in the condition called layer by layer growth mode (Larsen and Dobson, 1990; Das Sarma et al., 2001;
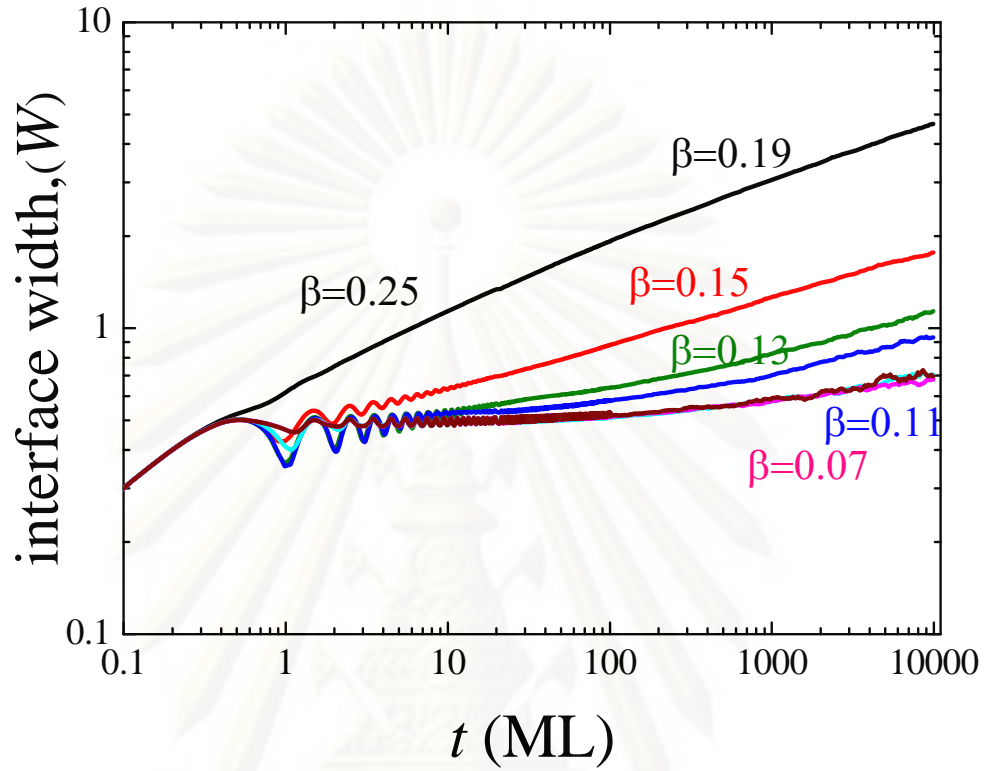
Figure 4.7: *W-t* plot of the DT model as diffusion length is increased from $\ell_d = 1$ (black line), 4 (red line), 10 (green line), 15 (blue line), 30 (pink line), 50 (cyan line) and 90 (brown line).

Chatraphorn and Das Sarma, 2002). In experiments, layer by layer growth mode is studied via the reflection high-energy electron diffraction (RHEED) oscillation pattern (Neave et al., 1985; Shitara et al., 1992; Marmokos and Das Sarma, 1992). The intensity RHEED pattern reaches its maximum at the time instant that a layer is completely filled. However, oscillation pattern in the $W - t$ plot is a reverse of that. The width is at the maximum as the film is grown to a half of each layer and then decreases until it reaches its minimum as the film is at a completely fill layer. The oscillation pattern the $W - t$ plot is reverse to the RHEED pattern because interface width represents roughness of a film, but RHEED pattern represents smoothness of the film.

The amplitude of the oscillation in $W - t$ plots indicates how perfect the layer by layer growth mode really is. Results from figure 4.7 show that this film surface is still imperfect because of the existence of damping in the oscillation. The damping occurs as a result of the fact that diffusion length is still too small so that the previous layer has not been filled up completely when the next layer is grown.

4.1.2.2 Morphologies

Morphologies shown in figure 4.8 are snapshots at half layers and full layers of a film grown with $\ell_d = 15$ sites. All of them are grown in layer by layer growth mode according to the *W-t* plot in figure 4.7 because the oscillation in the *W-t* plot can still be seen for $\ell_d = 15$ in this time range ($t \le 3 \, \text{MLs}$). If the film is grown in perfect layer by layer mode, the film should be at half-filled layer at $t = 0.5$, 1.5 and 2.5 ML (figure 4.8(a), (c) and(e)). In the same way, morphologies at $t = 1$, 2 and 3 ML (figure 4.8(b), (d) and (f)) should be at completely filled layer. However, from the morphologies in figure 4.8, we found that the film is not grown to exactly half-filled layer when $t = 0.5$, 1.5 and 2.5 MLs. At $t = 1$, 2, and 3 MLs, the film does not form complete layer either. We see some voids in the existed layer and a few small clusters appear as the next layer is beginning to form. It corresponds with the existence of damping and small amplitude in the *W-t* oscillation in figure 4.7.

Morphologies from $\ell_d = 50$ sites are shown in figure 4.9. It is clear that morphologies from this large ℓ_d system are much better than the ones with $\ell_d = 15$

sites. The film seems to be able to better maintain the initial tilted pattern of the substrate. At $t = 1, 2$, and 3 MLs, we do not see any cluster popping up on the existed terraces. However, the edge of each step is not sharp but has a meandering form (Wu et al., 1993; Xiao, 1997; Myslivecek et al., 2002; Rusanen et al., 2003). So the film is still not in the perfect layer by layer mode as can be confirmed by the damped oscillation in W-t plot in figure 4.7. In fact, from the W-t plot, the oscillations of growth systems with $\ell_d \geq 30$ have smaller amplitudes (more damped) than the ones from $\ell_d \leq 15$. This can be from the meandering of the step edge which results in a lot of voids on the film surfaces shown in figure 4.9. This is quite puzzling because at $\ell_d = 50$ sites, the diffusion length is already significantly larger than the terrace width L_t which is fixed at $L_t = 30$ sites. For $\ell_d > L_t$, the grown film should keep the exact substrate. There should not be any meandering at the step edge. The W-t oscillation should not be damped. To further understand this, morphologies at 100 MLs of films grown with $\ell_d = 15$ sites and $\ell_d = 50$ sites are studied. The morphologies are shown in figure 4.10 and 4.11 respectively. It is clear that the two morphologies are very similar despite the fact that one is from a system with $\ell_d < L_t$ and the other is from a system with $\ell_d > L_t$.

Another strange results observed here is when ℓ_d is increased from 30 sites to 95 sites, there is no change in the W-t curve and morphologies. From the $W - t$ graph, it is clear that the growth exponent β decreases from 0.19 to 0.11 as the diffusion length is increased from 1 to 15. This is because increasing the diffusion length reduces roughness of the films. However, this is true only when ℓ_d is in the range $1 \leq \ell_d \leq 15$. When ℓ_d is increased further ($30 \leq \ell_d \leq 95$), the $W - t$ plot and growth exponent β in figure 4.7 remain unchanged. The value of β is constant at $\beta = 0.07$. It is unusual because increasing the diffusion length is equivalent to increasing mobility of atom. So the growth exponent β should decrease with increasing ℓ_d. This problem is discussed in more details in section 4.1.3.

(a) $t = 0.5$ ML

(b) $t = 1$ ML

(c) $t = 1.5$ ML

(d) $t = 2$ ML

(e) $t = 2.5$ ML

(f) $t = 3$ ML

Figure 4.8: Snapshots of film morphologies of the original DT model with $\ell_d = 15 < L_t$ from substrate size $L \times L = 960 \times 960$ (a segment of 120×120) at (a) 0.5 ML (b) 1ML (c) 1.5ML (d) 2ML (e) 2.5 ML (f) 3 ML.

Figure 4.9: Snapshots of film morphologies of the original DT model with $\ell_d = 50 > L_t$ from substrate size $L \times L = 960 \times 960$ (a segment of 120×120) at a) 0.5 ML b) 1ML c) 1.5ML d) 2ML e) 2.5 ML f) 3 ML.

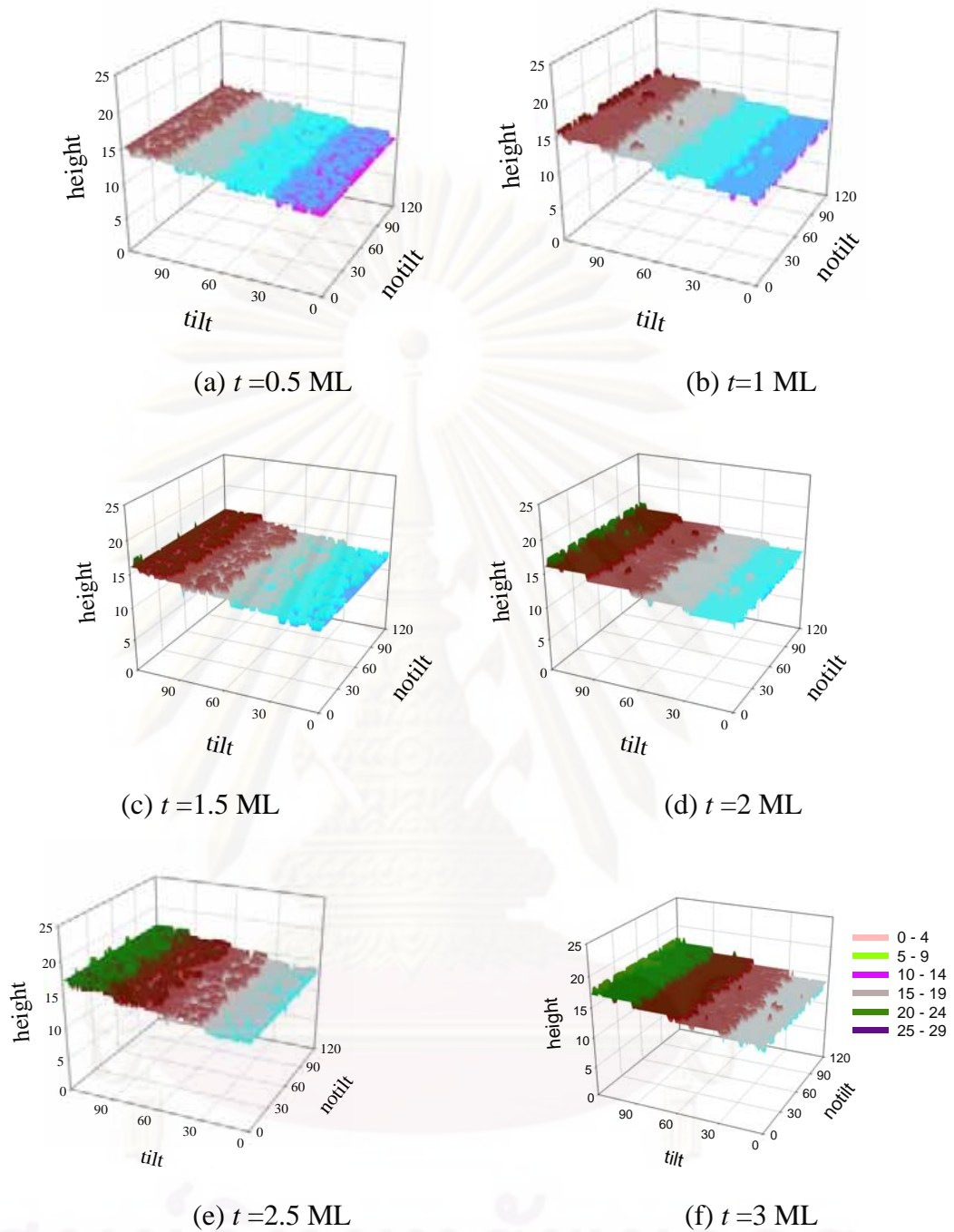**Figure 4.10:** Film morphologies of the DT model with $\ell_d = 15 < L_t$ from substrate size $L \times L = 960 \times 960$ (a segment of 120×120) at 100 ML.
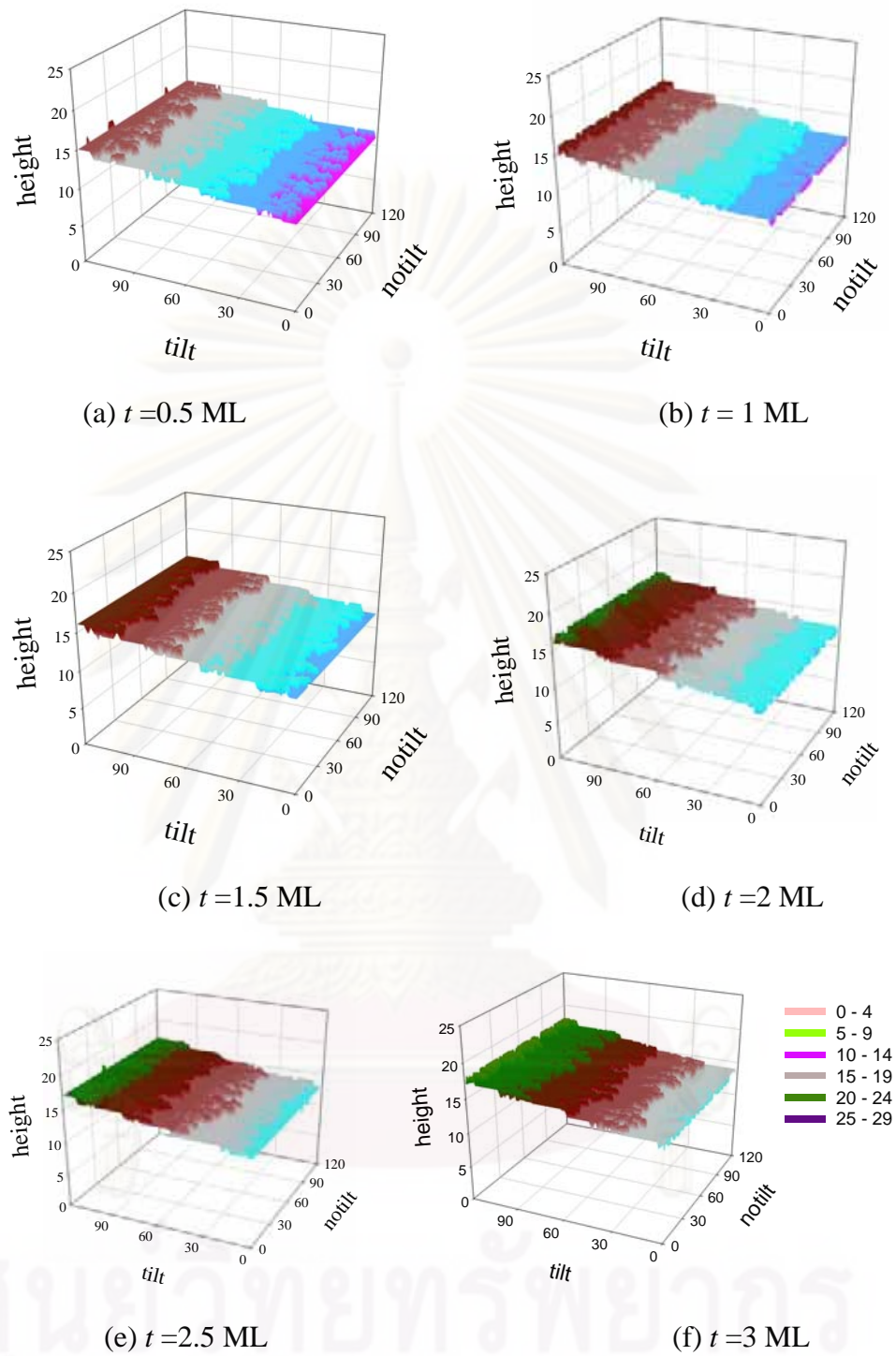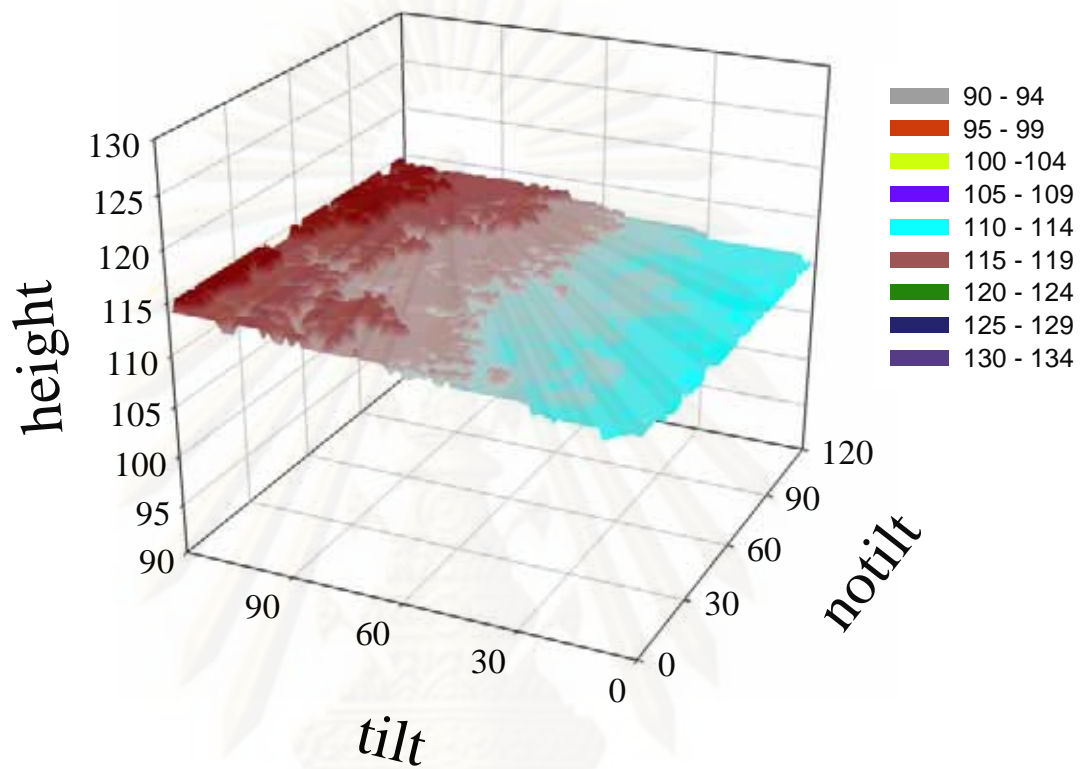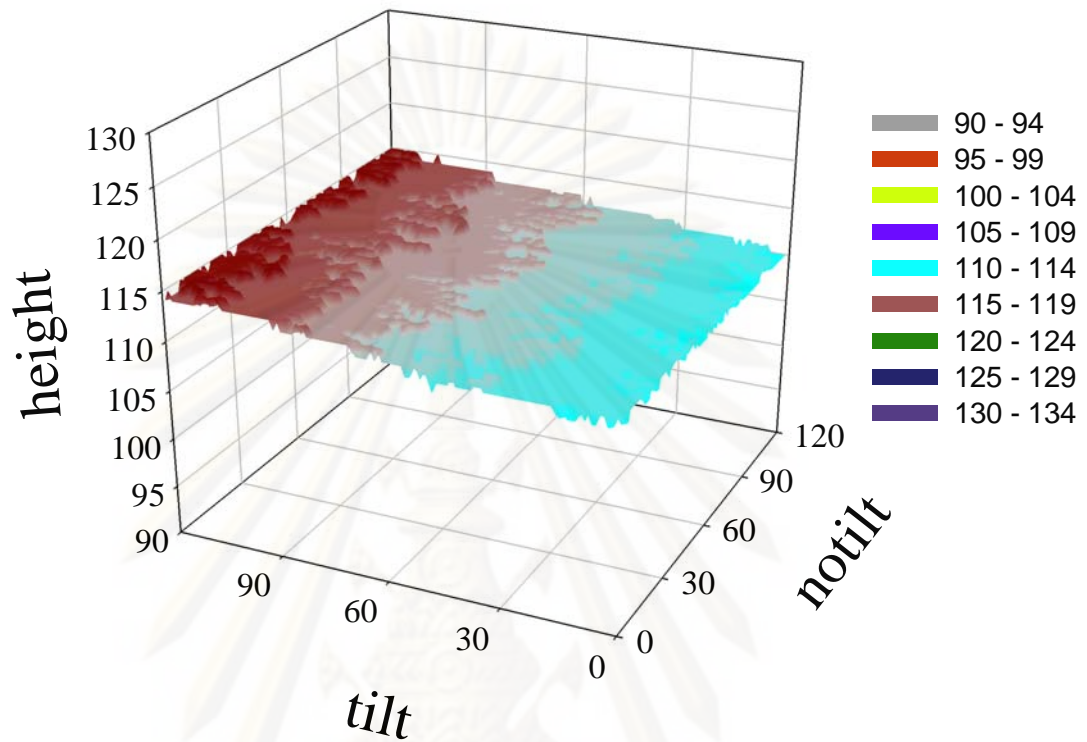
Figure 4.11: Film morphologies of the DT model with $\ell_d = 50 > L_t$ from substrate size $L \times L = 960 \times 960$ (a segment of 120×120) at 100 ML.

4.1.2.3 Persistence probability

A plot of persistence probability versus time ($P - t$ plot) is shown in figure 4.12. From the graph, as the diffusion length ℓ_d is increased in the range $1 \le \ell_d \le 15$, the persistence probability decay rate become smaller. This indicates that the film can keep the original pattern better at large ℓ_d. It corresponds with the oscillation pattern in $W - t$ plot in the range $1 \le \ell_d \le 15$. However, the film loses its original pattern from the early time because of the existence of damping in the $W - t$ oscillation. For $30 \le \ell_d \le 95$, the $P - t$ curves provide practically the same results in this range. This is consistent with the problem found in the $W - t$ plot and morphologies earlier.

4.1.2.4 Correlation function

Finally, the correlation function versus the distance or $G - r$ plot is shown in figure 4.13 (tilt direction) and in figure 4.14 (notilted direction). For results from the $G - r$ plot in the original DT model, there is no specific feature on the tilted direction. It implies that atom can diffuse to nearest neighboring site so the tilted of substrate does not have a strong influence on the original DT results. As the NRT is added to the original DT model, effects of the tilted of substrate can be seen. From figure 4.13, information about specific feature of the miscut substrate appears at all time t. We found that the curve of specific feature changes as the growth time is increased from 1 to 10^3 ML. In the early time (1 ML), the curve of $G - r$ plot looks like periodic square oscillation for all ℓ_d. The curve of periodic square oscillation repeats at every step of terrace. That is, the length between the square oscillations represents the length of the terrace. This is because the height used for the calculation of correlation function is the deviation of the surface height from the average height or height fluctuation. So, the height of the initial miscut substrate is taken away. However, the periodic of film surface remains to be seen. If the film can keep perfectly the initial pattern substrate, the periodic square oscillation which has the same size and shape (in figure 4.13(a)) is obtained. If the amplitude of the oscillation is constant, it indicates
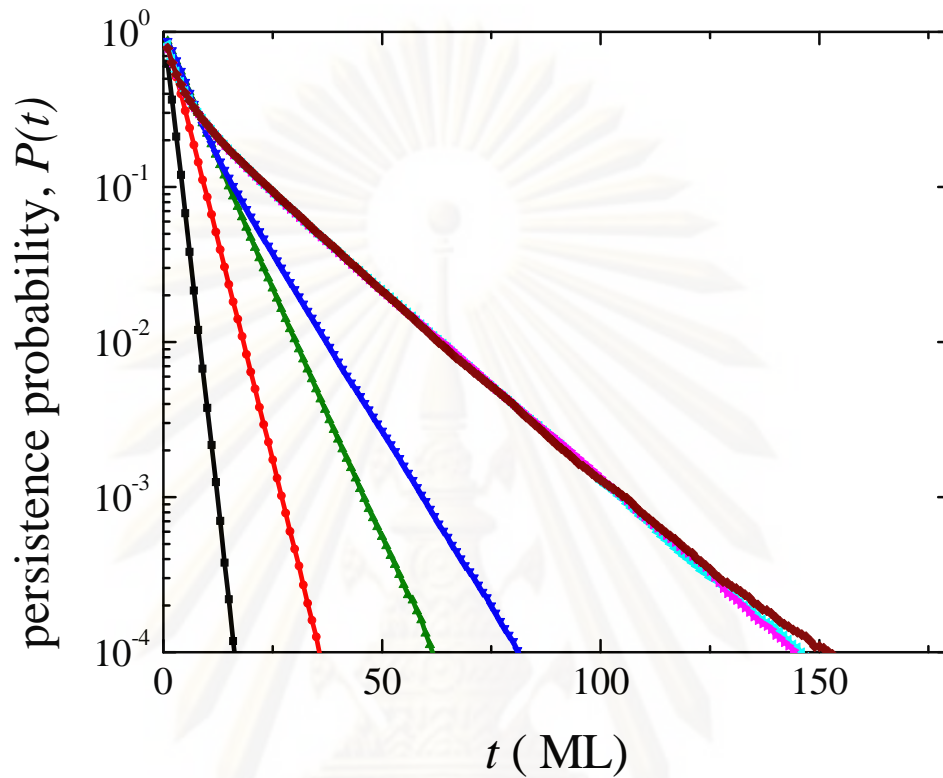
Figure 4.12: *P-t* plot of the DT model as diffusion length is increased from ℓ_d =1 (black line), 4 (red line), 10 (green line), 15 (blue line), 30 (pink line), 50 (cyan line) and 90 (brown line).

(a) t=1 ML

(b) t=10 ML

(c) t=10^2 ML

(d) t=10^3 ML

Figure 4.13: *G-r* plot of the original DT model in the tilted direction as the diffusion length ℓ_d is increased from 10 (black line), 15 (red line), 30 (blue line), 50 (green line) and 90(pink line) sites.

Figure 4.14: *G-r* plot of the original DT model in the notilted direction as the diffusion length ℓ_d is increased from 10 (black line), 15 (red line), 30 (blue line), 50 (green line) and 90(pink line) sites.

perfect flat terraces. On the other hand, if the tilted of the substrate are slightly different, the curve of specific feature is damped which can be seen in figure 4.13(b) for ℓ_d=10 and figure 4.13(c)(d) for all ℓ_d. At large time (10^3 ML), damped oscillation occurs at all ℓ_d which indicates that there are small clusters and islands on the terrace. From the graph of $G(r)$ at $t \geq 10$ MLs, we found that the film can not keep the initial configuration of the tilt substrate because the oscillation of the G-r curve is damped within a very short time. Furthermore, the diffusion length is increased in the range $\ell_d \geq 30$, the curves of correlation function provide the same results in both direction. It corresponds with the results from the morphology, the interface width and the persistence probability for the DT model.

The second correlation function $H(r)$ versus the distance r or $H - r$ plot is shown in figure 4.15 and 4.16. The specific feature can be found in the tilt direction 4.15(a) at the early time corresponding with the first correlation function $G(r)$. From the definition of $H(r)$, the value of $H(r)$ is at its maximum at r=0. As the diffusion length is larger, the value of $H(r=0)$ becomes smaller. It implies that the multiplicity of heights of two sites is smaller because of the large diffusion length. The value of $H(r)$ (multiplicity of heights at different sites) can be either positive or negative because the height fluctuation is used in the calculation. As r increases, the information about the periodic feature of the film appears in the tilt direction which is shown in figure 4.15. In the notilted direction (figure 4.16), there is no specific feature on the film surface because the value of $H(r)$ is close to zero for all r. In the tilt direction, as r increases, the multiplicity of heights between two sites on the surface decreases so that the value of $H(r)$ decreases. As r equals the terrace length (L_t), the peak position of $H(r)$ occurs. We found that the peak of multiplicity of heights repeats at every step of terrace. This is because when the diffusion length ℓ_d equals L_t, most of the atoms can move to a step edge of the terrace. So the majority of atoms deposit at a step edge of the terrace. This causes the multiplicity of the heights to increase at each step edge of terrace so the peak position repeats itself at every step edge of the terrace. As $r > L_t$, the value of $H(r)$ decreases and becomes approximately zero. As r equals to L_t again, the peak position occurs. The peak positions are separated by the terrace length L_t. In the result of $H(r)$ in the tilt.
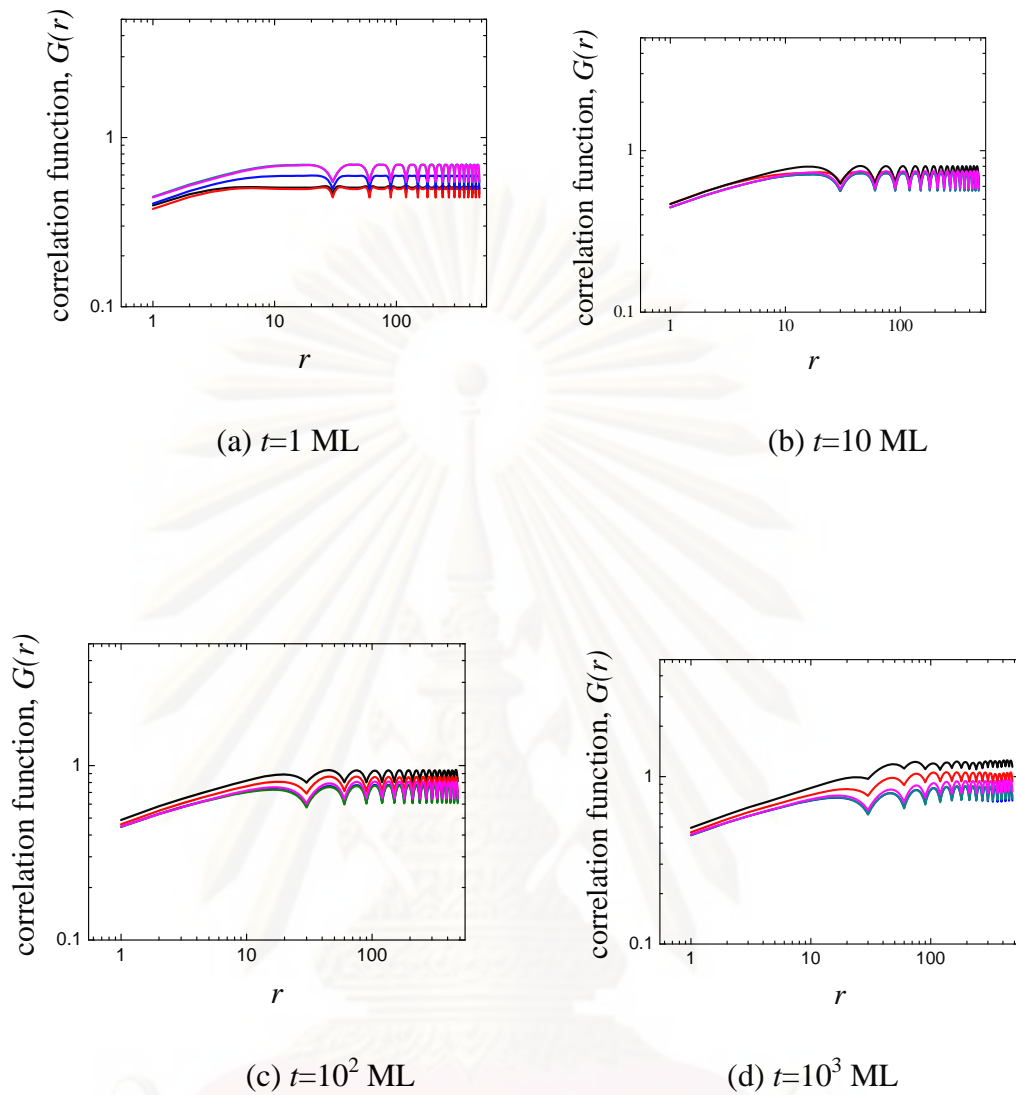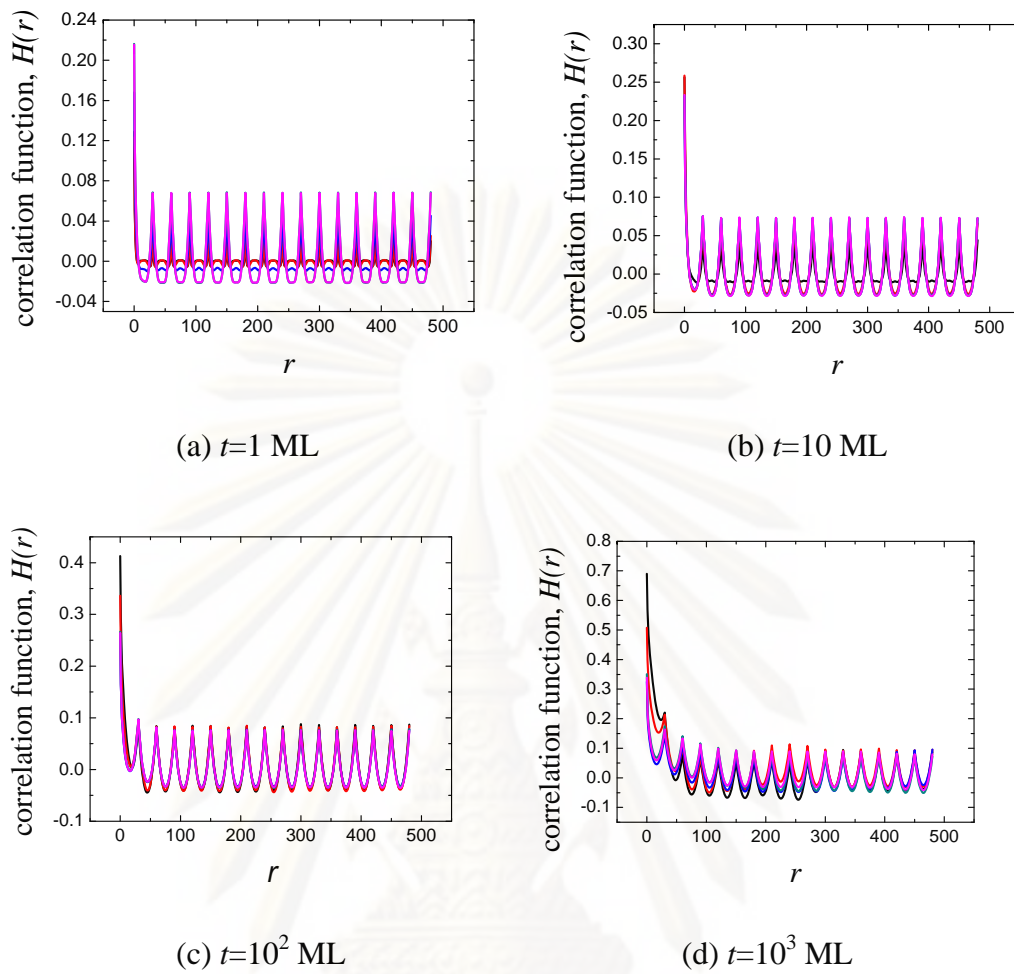
Figure 4.15: *H-r* plot of the original DT model in the tilted direction as the diffusion length ℓ_d is increased from 10 (black line), 15 (red line), 30 (blue line), 50 (green line) and 90(pink line) sites.
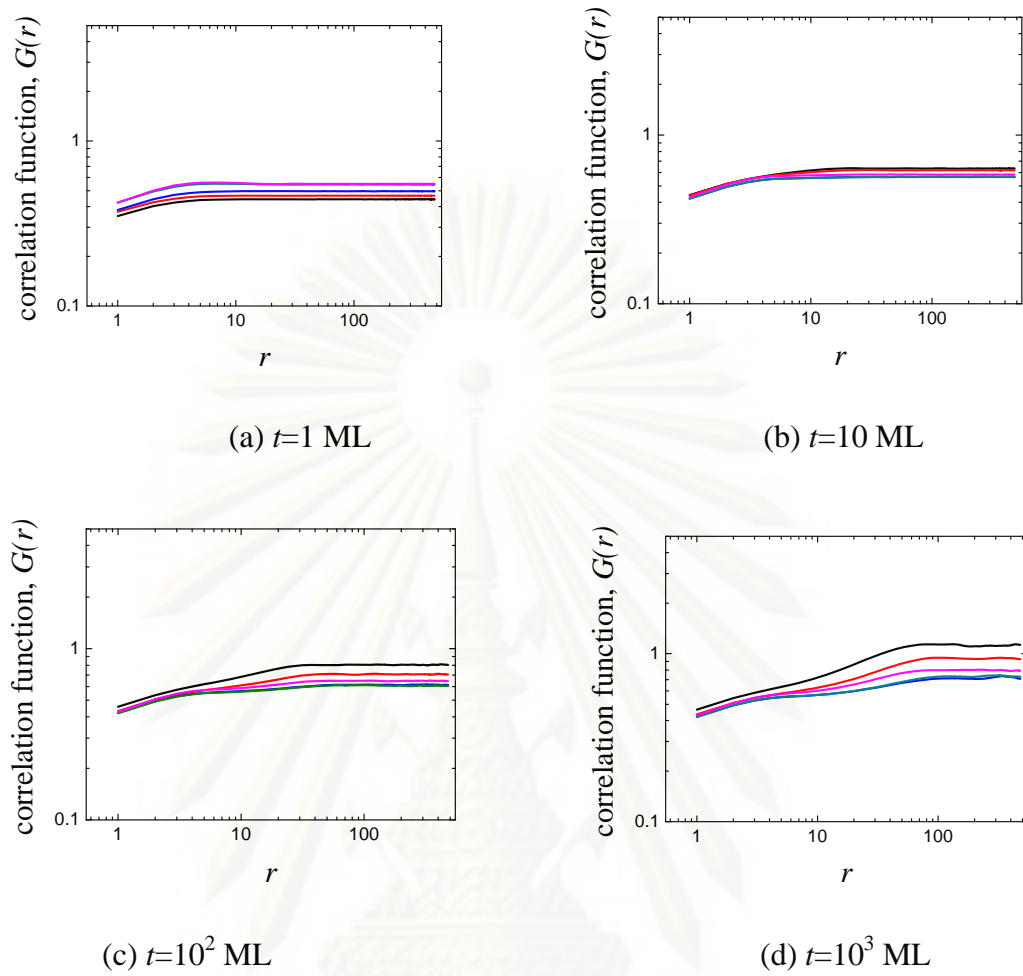
(a) t=1 ML

(b) t=10 ML

(c) t=10^3 ML

(d) t=10^4 ML
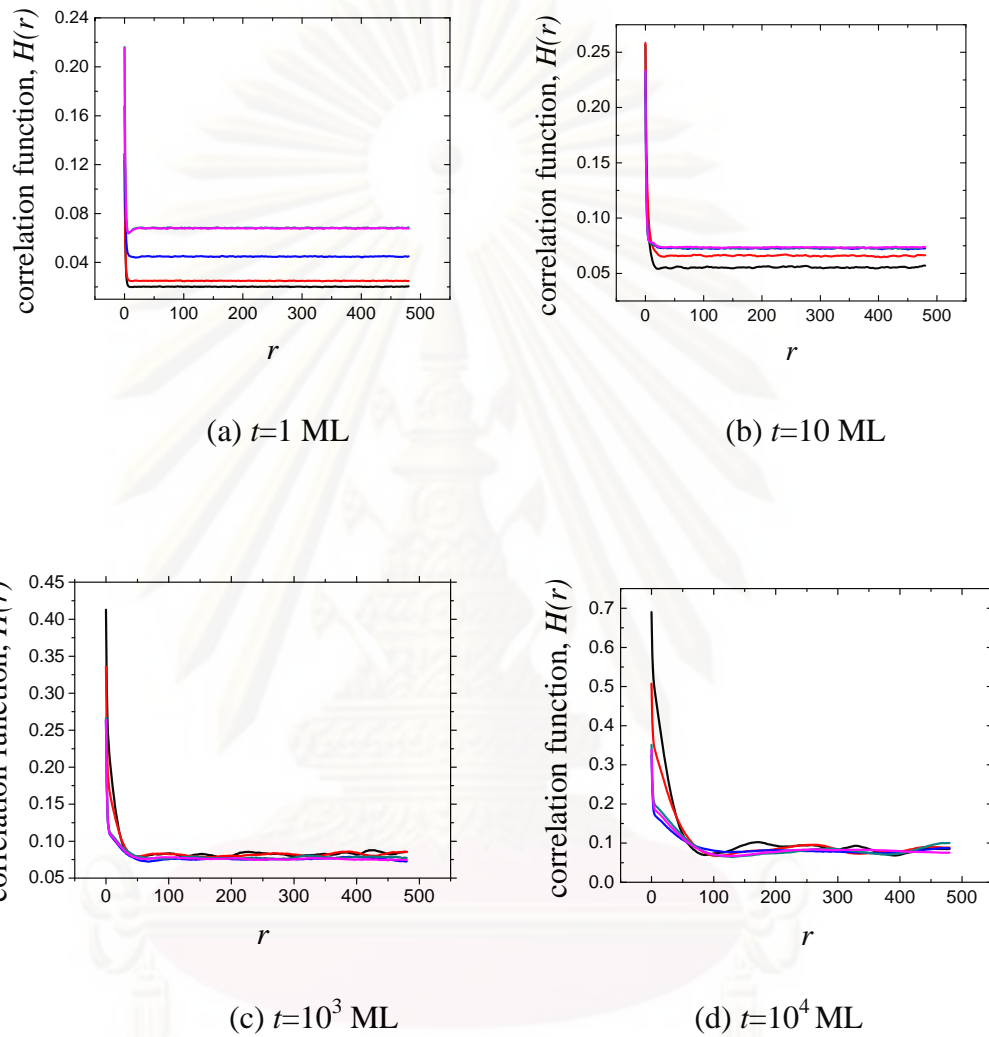
Figure 4.16: *H-r* plot of the original DT model in the notilted direction as the diffusion length ℓ_d is increased from 10 (black line), 15 (red line), 30 (blue line), 50 (green line) and 90(pink line) sites.

direction, damping of oscillation occurs in the early time although the diffusion length is large.

For the DT model with long surface diffusion length, all results are quite unrealistic. The curves of $W-t$, $P-t$, $G-r$ and $H-r$ plots are the same for ℓ_d=30,50 and 90 sites. A possible cause is the diffusion rule of the DT model that was designed for low mobility diffusion. In the diffusion rule of the DT model, an atom that has at least one lateral bond with a neighboring site does not move because it does not have enough energy to break away from the existed bonds. When we increase the diffusion length $30 \leq \ell_d \leq 95$, the intention is to increase the mobility of atoms. In this situation, atoms with more energy have much more chance to leave the kink site. But that is not allowed in the diffusion rule of the DT model. In this work, we modify the diffusion rule of the DT model to improve on this. The modified DT model allows atoms to leave kink sites by breaking two existed bonds. In the modified DT model, if atom is at a trapping site with at least two lateral bonds with its neighboring sites, it still can not move. Results from this modified DT model are presented in section 4.1.3.

4.1.3 Results from the modified DT model with long surface diffusion length

From 4.1.2, the results from the DT model with long surface diffusion length are quite unrealistic and a possible cause is the diffusion rule of the DT model itself. In the diffusion rule of the DT model, if an atom has at least one lateral bond with a neighboring site, it is not allowed to move. As we add the long surface diffusion length NRT to the DT model, it is equivalent to increasing the substrate temperature in experiments. When the substrate temperature is high, energy of surface atoms also becomes high leading to two outcomes?. The first is that the atom mobility is large and the atom can travel far from its initial deposition site, i.e. long surface diffusion length. At the same time, the second is that atom with more energy should be able to break the initial bonds. So atoms have much more chance to leave the kink site. But the diffusion rule of the DT model with long surface diffusion length is not consistence. The model used for 4.1.2 is equivalent to an experiments with high substrate temperature that allows only the first outcome (long surface diffusion length) while the second outcome (atom can break more bonds) is not included. So, the curves of $W - t$, $P - t$, $G - r$ and $H - r$ are the same for ℓ_d=30,50 and 90, which is not believable. In this work, we modify the diffusion rule of the DT model to improve on this. The modified DT model allows a diffusing atom to leave a kink site by breaking two bonds. However, an atom at a trapping site (a site with at least two lateral bonds) still can not move away. This implies that the energy of the surface atom is larger than atoms in the original DT model, but not too high. The long surface diffusion length NRT is also added to this modified DT model.

4.1.3.1 Interface width

$W - t$ plot of the modified DT model with NRT is shown in figure 4.17. As ℓ_d is increased from 10 to 95 sites, we found that the amplitude of the oscillation increases due to the perfect layer by layer growth mode. As $\ell_d \geq L_t$, the $W - t$ oscillation continues for a very long time and there is only a very small damping in
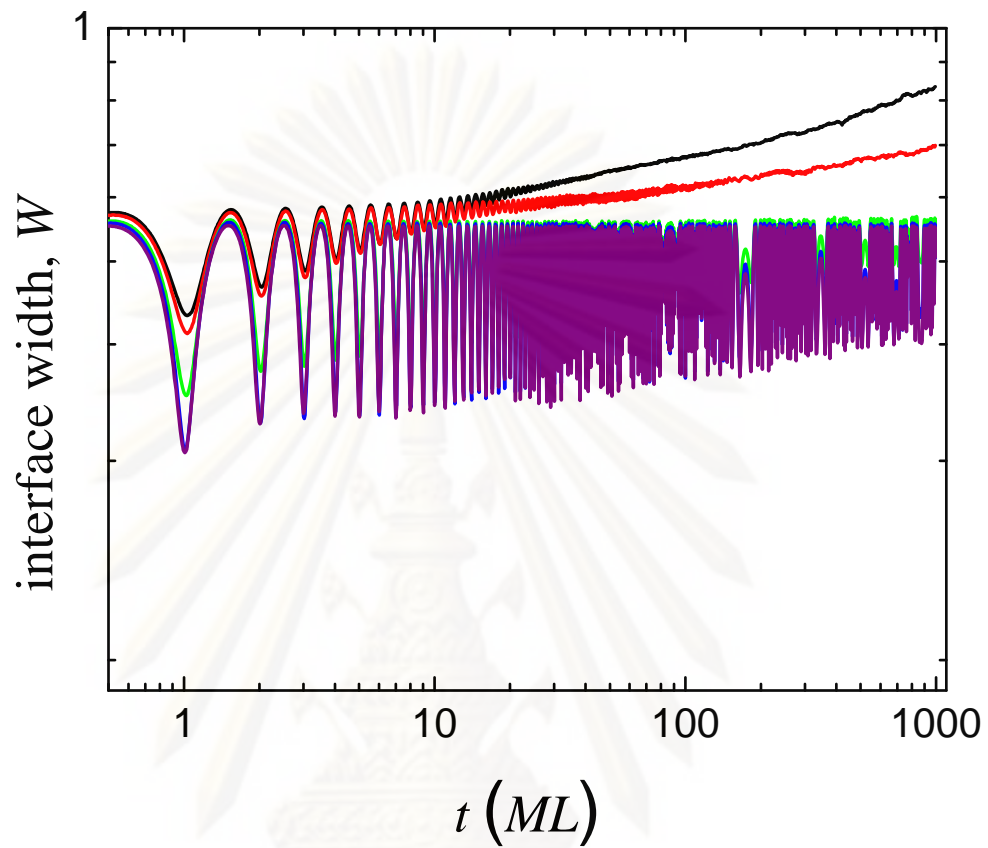
Figure 4.17: *W-t* plot of the DT model as diffusion length is varied from $\ell_d = 10$ (black line), 15 (red line), 30 (green line), 50 (blue line) to 95 (purple line).

the oscillation. From the results of the modified DT model, the perfect layer by layer growth mode is obtained as $\ell_d \geq L_t$.

4.1.3.2 Morphologies

Film morphologies of the modified DT model are shown in figure 4.18 and figure 4.19. These morphologies show that increasing the diffusion length reduces the roughness of films and help maintaining tilt in the substrate. As we have discussed before, when the diffusion length is increased, the atom can move further away from the deposition site and the layer by layer growth can be obtained. As $\ell_d = 10 < L_t$, which can be seen in figure 4.18, the film is very rough. The initial tilt of the substrate can not be clearly seen in this condition. As $\ell_d = 50 > L_t$, shown in figure 4.19, atoms can move to the step edge of each terrace and form a complete layer. The initial tilt of the substrate can be clearly seen under this condition. Comparison between the film morphology of the DT model in figure 4.11 and the film morphology of the modified DT model in figure 4.19 at $\ell_d \geq L_t$, we found that the initial tilt of the substrate of both models can be seen. However, in the DT model, the film does not form complete layers, but in the modified DT model, we found that the film surface forms complete layers. We can explain that the diffusion rule of the DT model is the leading cause of the voids in the film surface. This is because, if atom is at a kink site, it can not move. So, there is much empty space in the film surface. It leads to voids on the film surface. In the modified DT model, the void in the film surface does not appear. This is because, as the final site is a trapping site (a site with at least 2 lateral bonds), atom can diffuse to fill a void. It corresponds with the figure 4.19, the perfect layer by layer growth mode or smooth film is obtained as $\ell_d \geq L_t$. Snapshots of the film surface with the condition that $\ell_d \geq L_t$ and in layer by layer growth mode regime are shown in figure 4.20. The film forms completely filled layers at 1, 2 and 3 ML. It shows that the film is grown in perfect layer by layer growth mode as $\ell_d \geq L_t$. If the film is grown in this mode, the film morphology should be perfectly smooth. From the morphologies in the range $1 \leq t \leq 3$ ML, each layer of thin film fills up completely

Figure 4.18: Film morphologies of the modified DT model with $\ell_d = 10 < L_t$ from substrate size $L \times L = 960 \times 960$ (a segment of 120×120) at 100 ML.

Figure 4.19: Film morphologies of the modified DT model with $\ell_d = 50 > L_t$ from substrate size $L \times L = 960 \times 960$ (a segment of 120×120) at 100 ML.

(a) *t*=0.5 ML

(b) *t*=1 ML

(c) *t*=1.5 ML

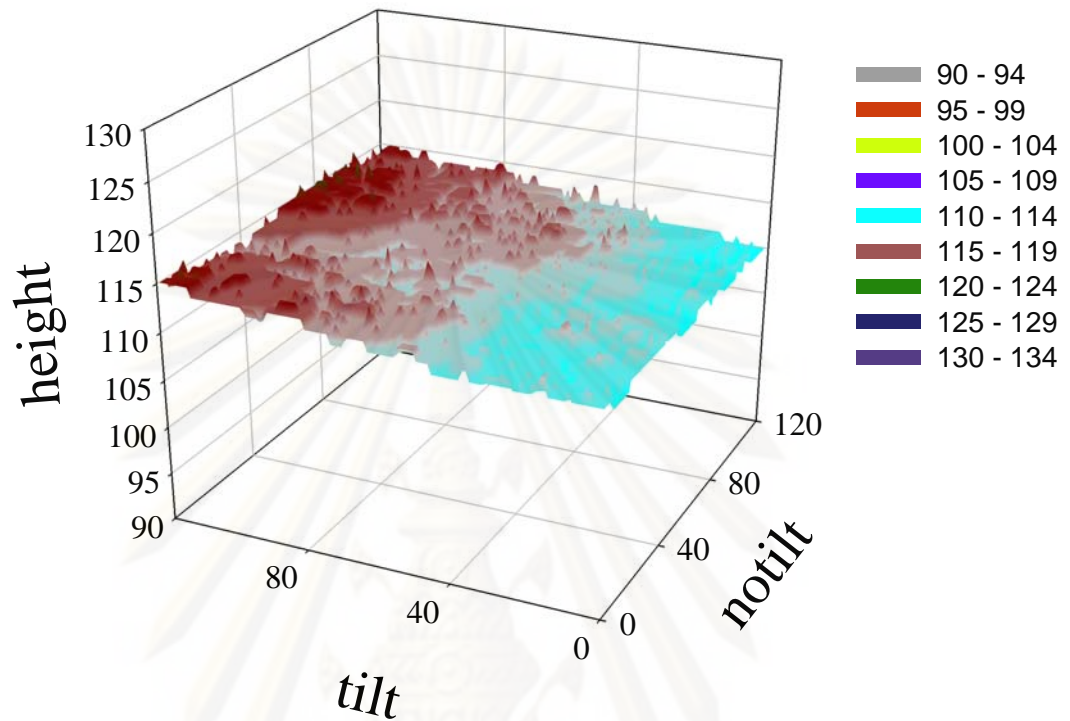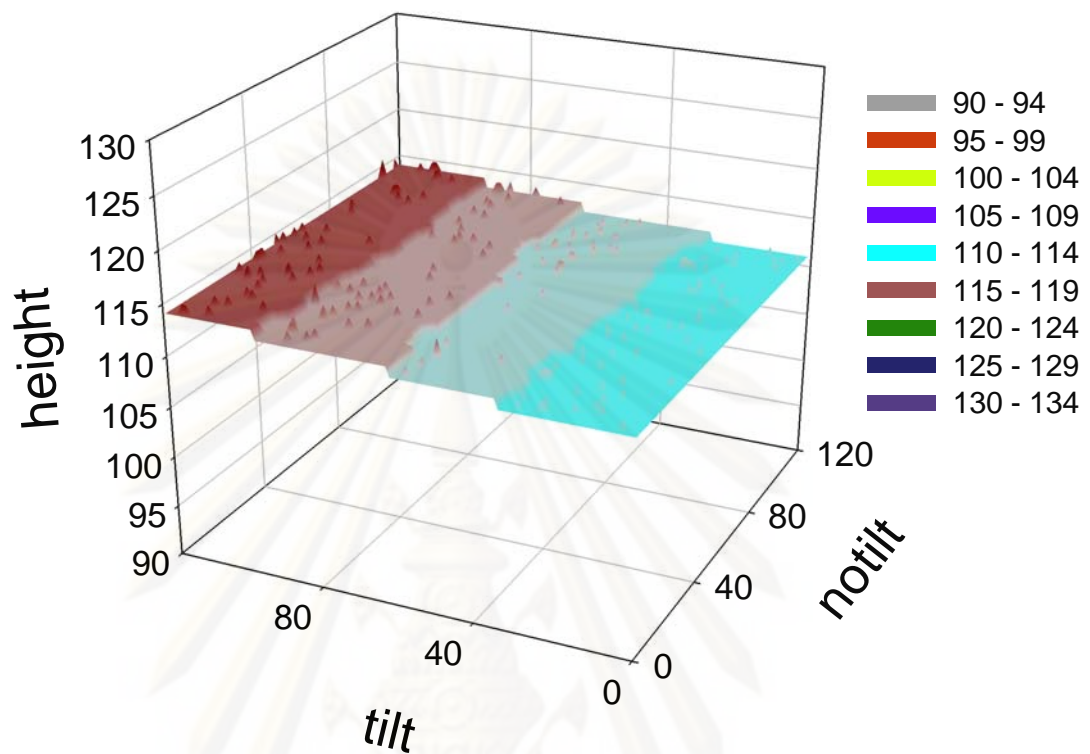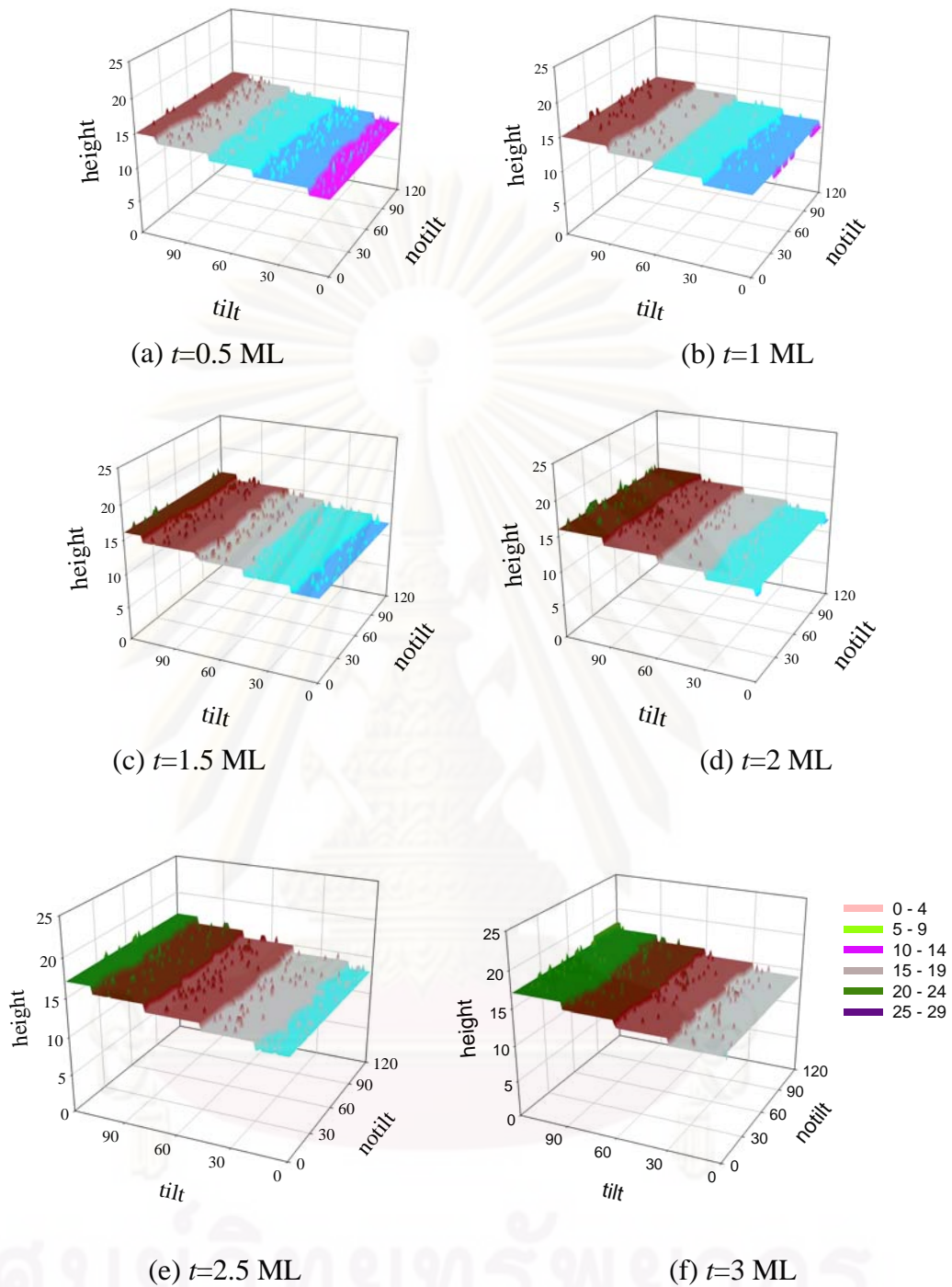(d) *t*=2 ML

(e) *t*=2.5 ML

(f) *t*=3 ML

Figure 4.20: Snapshots of film morphologies of the modified DT model with $\ell_d = 50 < L_t$ from substrate size $L \times L = 960 \times 960$ (a segment of 120×120) at a) 0.5 ML b) 1ML c) 1.5ML d) 2ML e) 2.5 ML f) 3 ML.

before the next layer begins and there is no void on the surface. This result confirms that the film is grown in perfect layer by layer growth mode for a long time as $\ell_d \geq L_t$. It corresponds with $W - t$ plot shown in figure 4.17.

In figure 4.21, Simulated film morphologies using increased diffusion length (figure 4.21(a)) are compared with experimental film morphologies with increased substrate temperature (figure 4.21(b)) (Myslivecek et al., 2002). The experimental images are 150×150 nm^2 from STM measurements. For the experimental film, Si (001) substrate is miscut 0.66^o along an azimuth [1 1 0] direction and the surface of film looks like terrace. As the substrate temperature is increased, the tilted of substrate can be seen clearer. It agrees with the simulated film which show better morphologies for larger ℓ_d. It confirms that increasing the diffusion length in the simulation is equivalent to increasing the substrate temperature in experiments and results from the modified DT model better agree with experiments. This shows that the inconsistency between the original DT results and experimental results arise from the limitation presents in the diffusion rule of the original DT model.

4.1.3.3 Persistence probability

The $P - t$ plot shown in figure 4.22 corresponds with the morphology results. As the diffusion length is increased from ℓ_d=10 to 90 sites, we found that the curve of $P - t$ plot decreases slowly from $P(t)$=1. For large ℓ_d, the rate that $P(t)$ decrease becomes small. If the film can keep perfectly the original pattern substrate, the value of persistence probability will be $P(t)$=1. It shows that the persistence of film approach perfect persistence as the diffusion length is increased.

4.1.3.4 Correlation function

Finally, the correlation function versus the distance r ($G - rplot$) is shown in figure 4.23 and figure 4.24 (tilted and notilted direction, respectively). We found that the $G - r$ plot has a power law dependence with r in tilt and notilt direction at all ℓ_d.
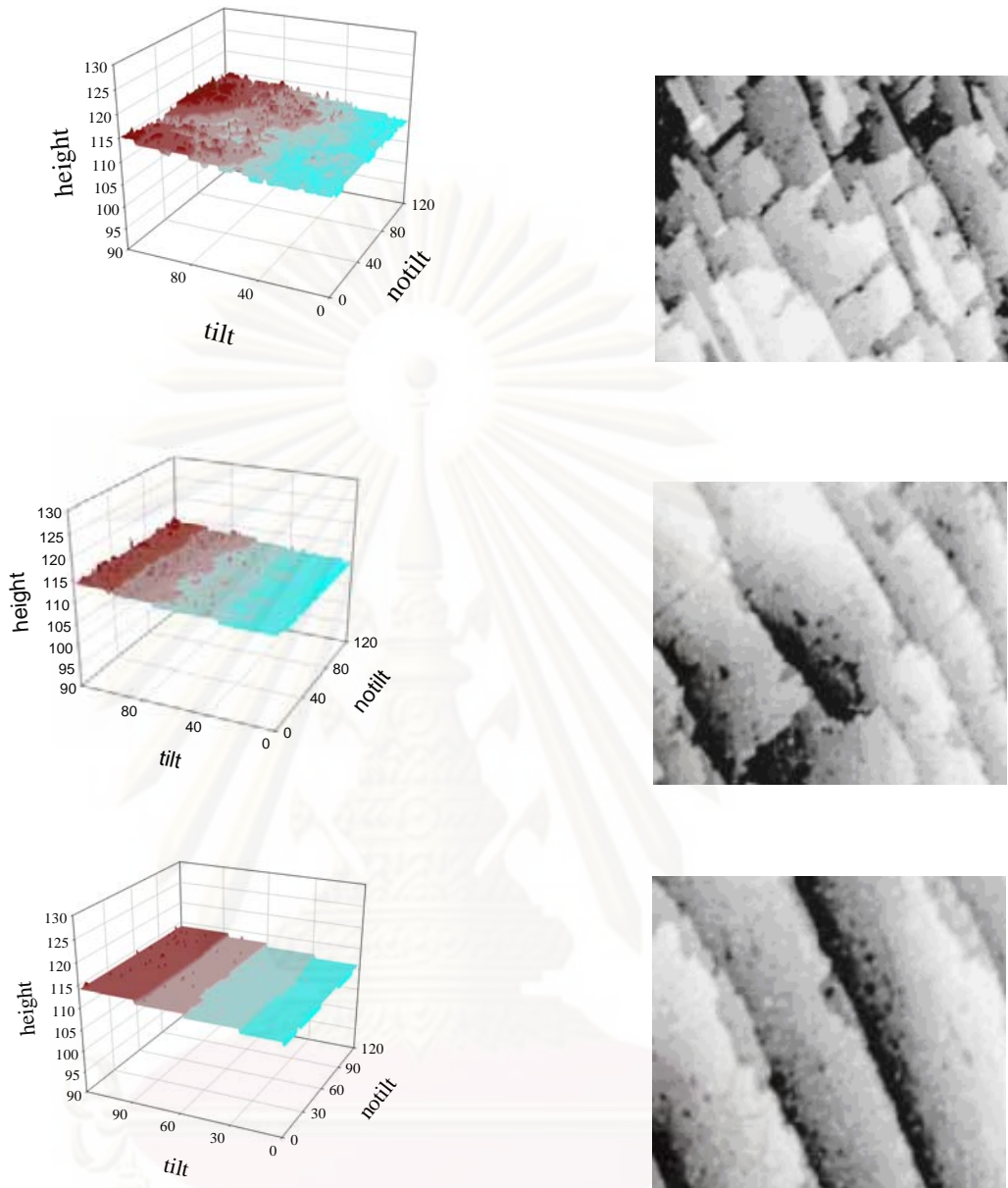
(a) simulated film   (b) experimented film

Figure 4.21: Simulated and experimental film morphologies (a) and (b) respectively. Increasing the diffusion length ℓ_d=10, 30 and 95 sites (top to bottom) in the simulated film and increasing substrate temperature T =400, 490 and 550 $^{\circ}$C (top to bottom) in experimental film (Myslivecek et al., 2002). The experimental images are 150×150 nm^2 from STM measurements. For the experimental film, Si (001) substrate is miscut 0.66° along an azimuth [1 1 0] direction.
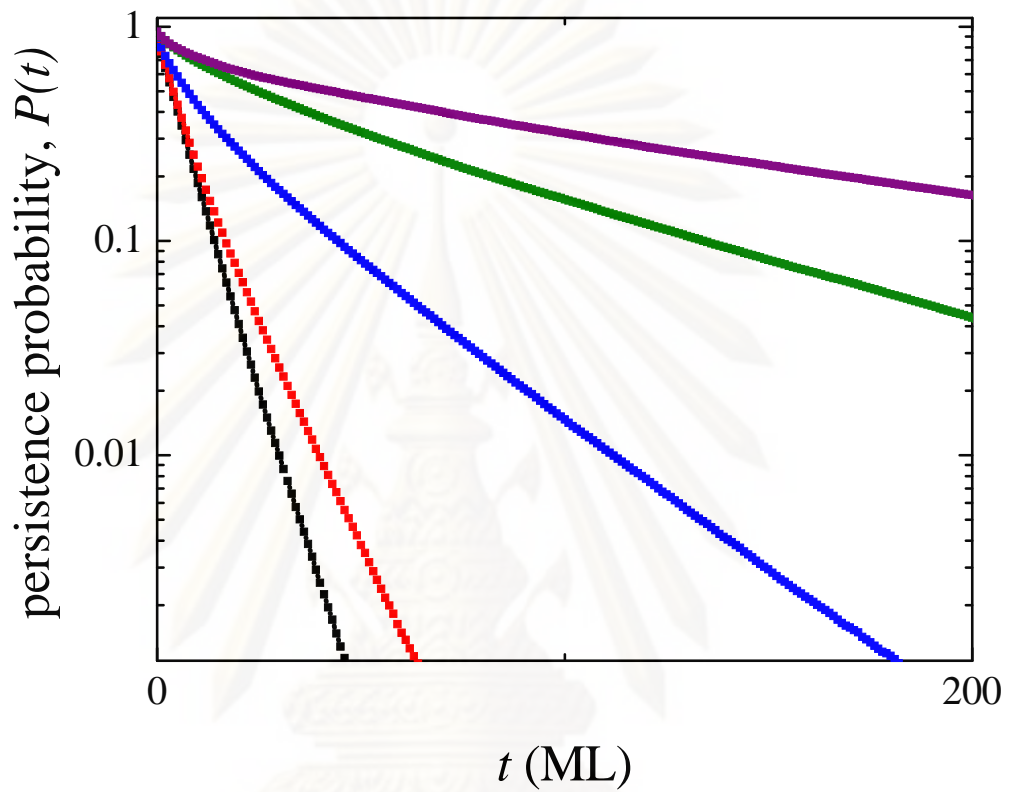
Figure 4.22: *P-t* plot of the DT model as diffusion length is increased from $\ell_d=10$ (black line), 15 (red line), 30 (blue line), 50 (green line) and 95 (pink line).

Figure 4.23: *G-r* plot of the original DT model in the tilted direction as the diffusion length ℓ_d is increased from 10 (black line), 15 (red line), 30 (blue line), 50 (green line) and 95 (pink line).

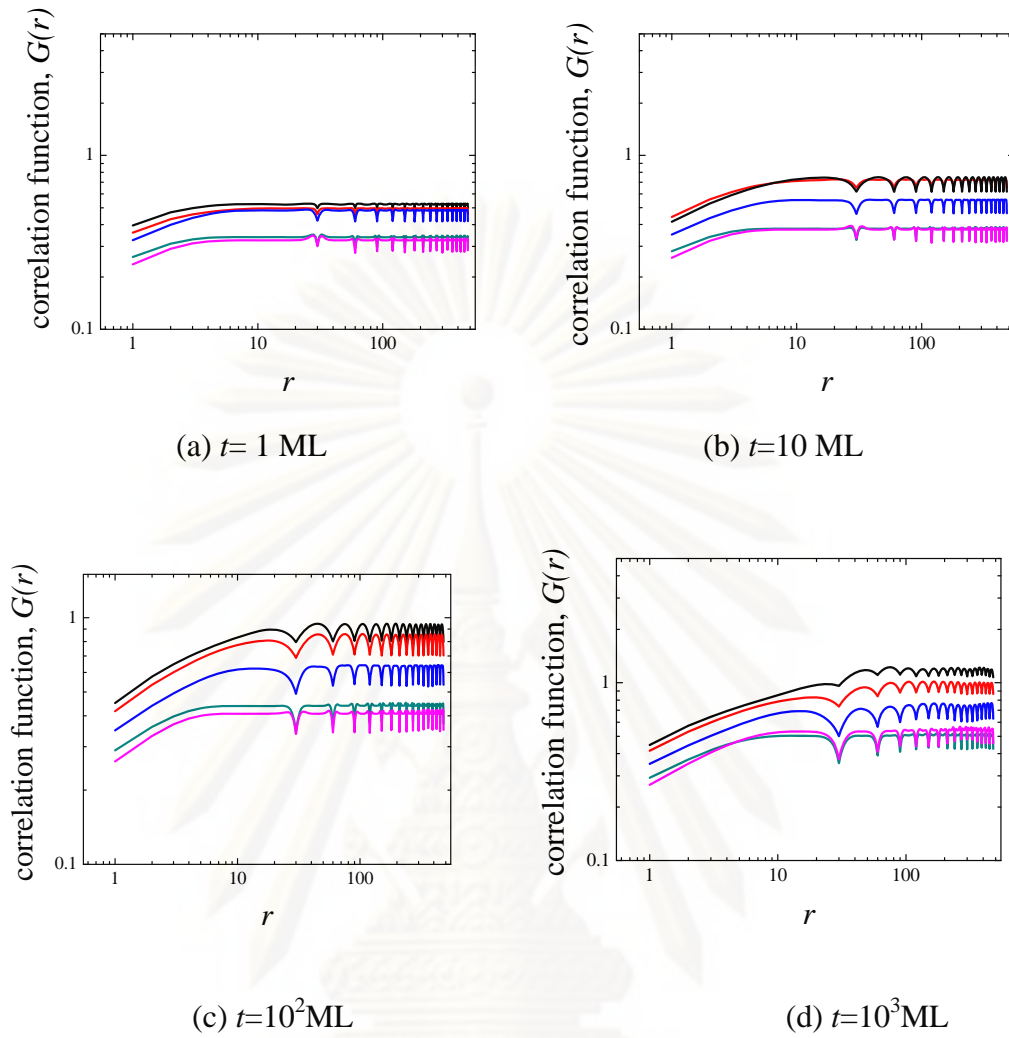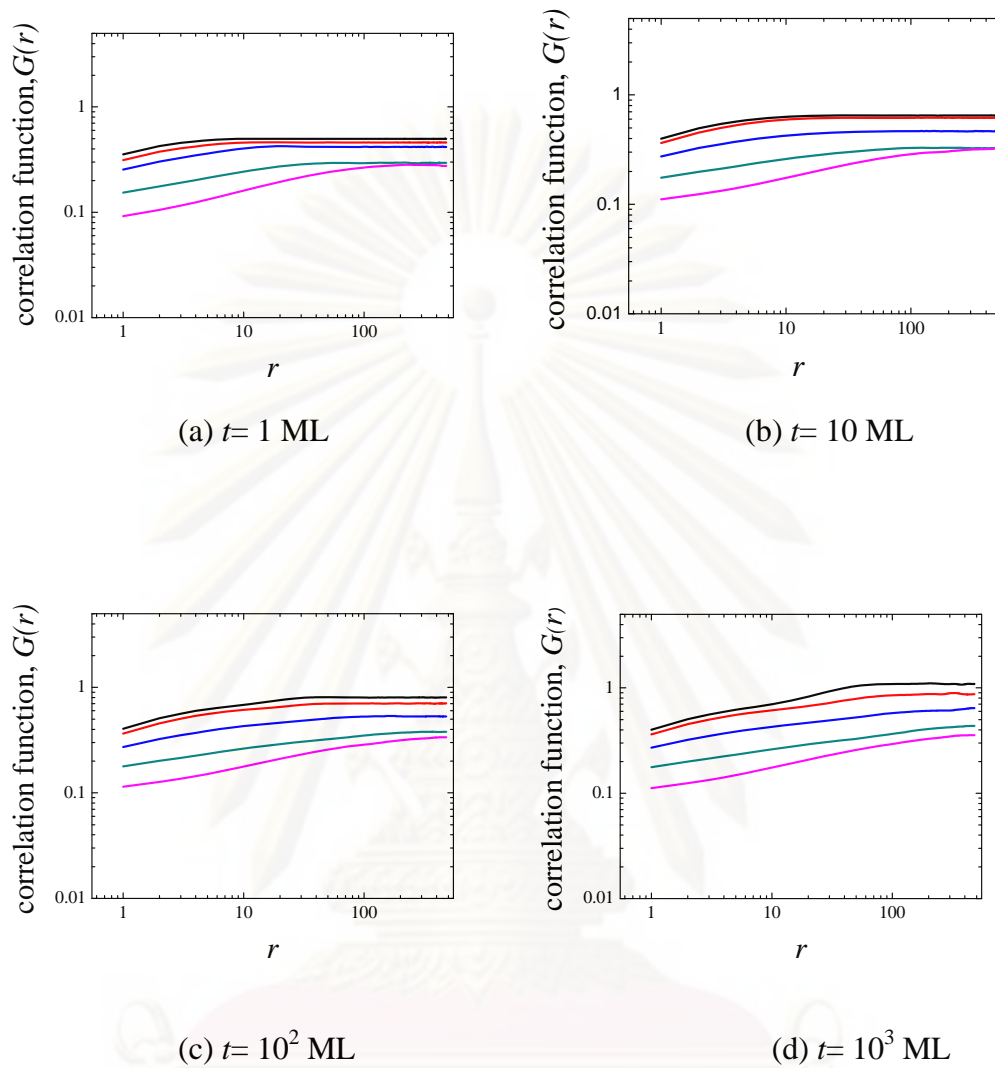**Figure 4.24:** *G-r* plot of the original DT model in the notilted direction as the diffusion length ℓ_d is increased from 10 (black line), 15 (red line), 30 (blue line), 50 (green line) and 95 (pink line).

This region occurs as $r \ll \xi$. This region is long if the diffusion process can expand for a long distance. From the graph of $G - r$ in tilted and notilted direction, this region

which the function has a power law with r, is longer as the diffusion length increases from ℓ_d=10 to 95 sites. This is because, as the diffusion length is increased, the expansion of correlation length ξ is longer. So, this region is long at large diffusion length. Moreover, as the diffusion length is increased from 10 to 95 sites, we found that the value of correlation function decreases. We see that the value of *G(r)* is smallest at ℓ_d=95 sites in both directions. It implies that increasing the diffusion length reduces the different heights between two sites on the film surface. Moreover, in the result of the modified DT model, the value of *G(r)* follows order of the diffusion length. It differs from the correlation function of DT results, which is disorderly with increasing diffusion length. For large r, the correlation function coverges into constant. This region is called saturated steady state which occurs for $r \approx \xi$. In this region, we found the periodic square oscillation in the tilt direction for t= 1 to 10^3 MLs when $\ell_d \geq L_t$. It implies that most atoms can move to step edge of the terrace indicating layer by layer growth for $\ell_d \geq L_t$. These results agree with the layer by layer growth mode in *W-t* plot. On the other hand, as $\ell_d < L_t$, the damped oscillation occurs since 10 ML. For notilted direction, information of specific feature of film surface does not appear. The value of *G(r)* decreases as the diffusion length is increased. This result is the same in the tilt direction.

For second definition of correlation function *H(r)* (in figure 4.25 and figure 4.26), At r=0, we found decreasing the maximum value of *H(r)* as the diffusion length is increased. The reason is increasing the diffusion length reduces the multiplicity of heights between two sites on the film surface. As r increases, in the tilt direction, the periodic square oscillation which agrees with the *G-r* plot but the curve of *H(r)* overturns *G(r)*, is obtained. The cause of overturning of graph between the *G-r* and *H-r* plot is, the *G-r* is plotted on the log-log scale, but the *H-r* is plotted on linear scale. However, the results of *H(r)* agree with *G(r)* for the modified DT results. This is the original periodic pattern substrate persists as $\ell_d \geq L_t$ for a long time. From the graph of *H(r)* in tilted direction, the square periodic oscillation still remains from t=1 to 10^3 MLs. It shows that the layer by layer growth persists as long as 10^3 MLs for $\ell_d \geq L_t$. In the notilted direction, there is not information specific feature of film. But at ℓ_d=10 (in figure 4.26 (d), small oscillation of *H(r)* appears. It shows that there is small

mound or island on the film surface because of ℓ_d is too small. So, there is formation of island on the terrace. Small oscillation of *H(r)* can be seen in the DT result as $\ell_d = 1$. Generally, the average mound height *H* is calculated by $H = G(r=0)^{\frac{1}{2}}$ (Chatraphorn, 2000; Chatraphorn et al., 2001). In this work, the average mound is neglected because existence of formation of island is very tiny.

All results of the modified DT model show that increasing the diffusion length has influenced on the simulated film. As the diffusion length is increased in the range $30 \le \ell_d \le 90$, the grown film is smoother. It agrees with the experimental film. Moreover, we found that the film is grown in layer by layer growth mode for along time when $\ell_d \ge L_t$. This condition, the grown film can maintain the initial configuration tilted substrate for a long time. In this work, modified diffusion rule of DT model is appropriate to increase long surface diffusion length in this range. However, if the diffusion length is further increased, the modified DT model may be unusable. Limitation of diffusion rule of the modified DT model occurs if diffusion length is too much.

Figure 4.25: *H-r* plot the of original DT model in the tilted direction as the diffusion length ℓ_d is increased from 10 (black line), 15 (red line), 30 (blue line), 50 (green line) and 95 (pink line).
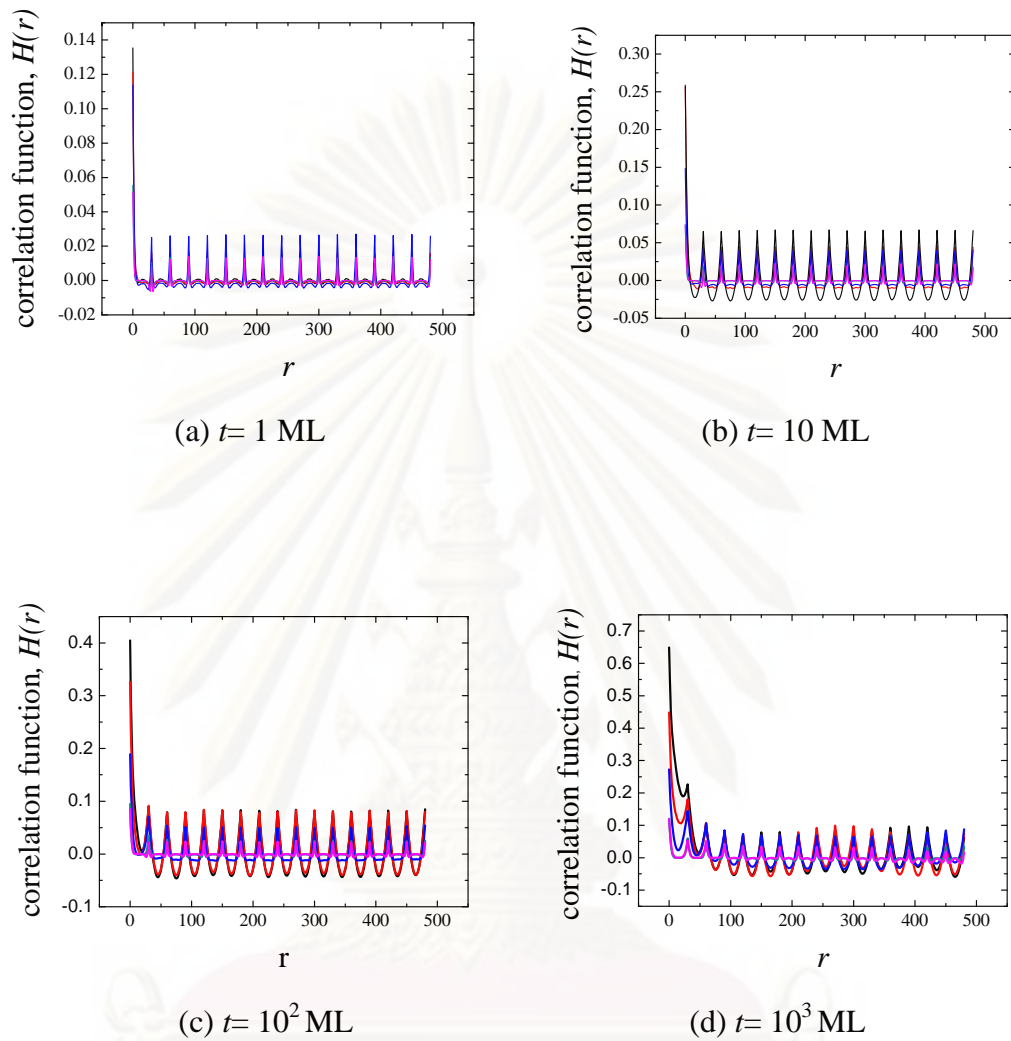
Figure 4.26: *H-r* plot of the original DT model in the notilted direction as the diffusion length ℓ_d is increased from 10 (black line), 15 (red line), 30 (blue line), 50 (green line) and 95 (pink line).
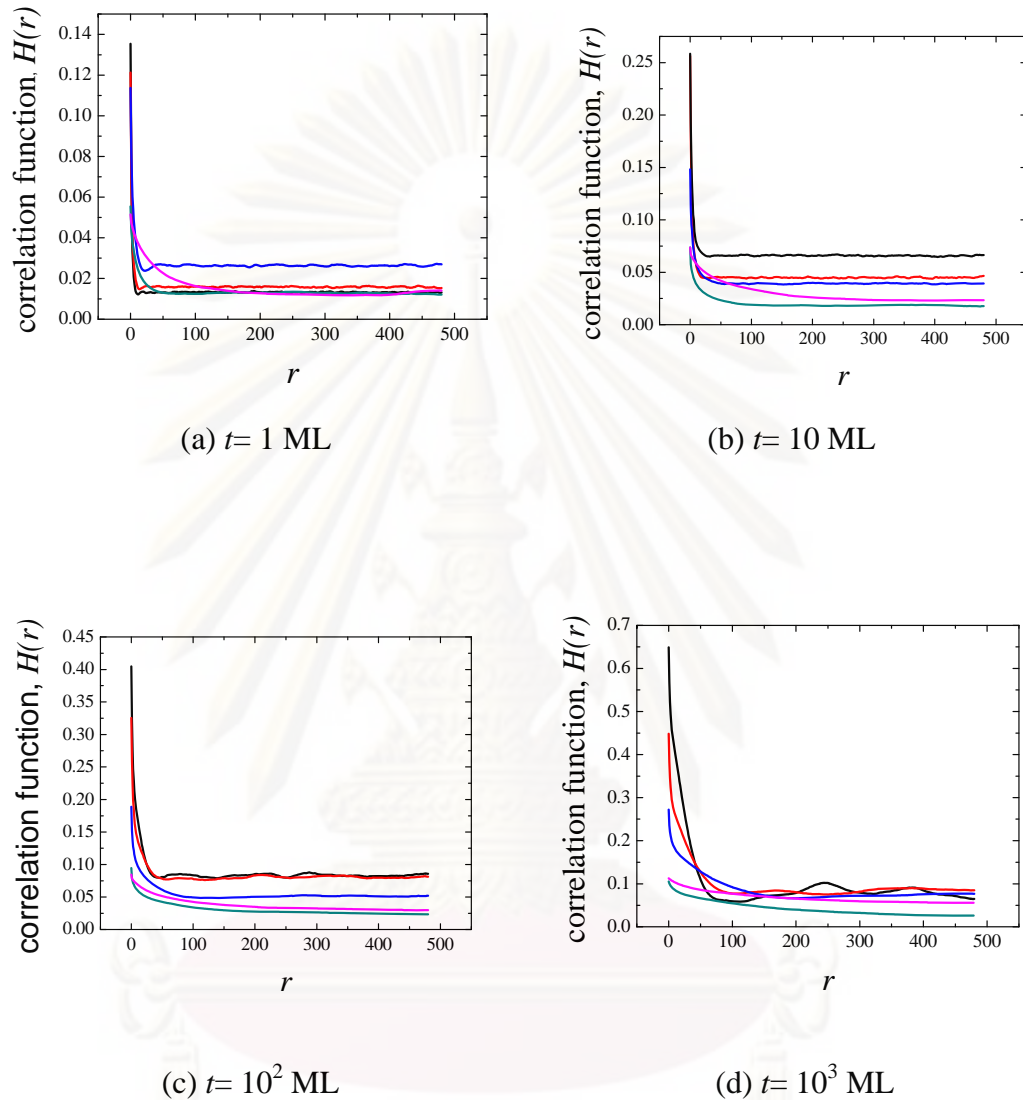
4.2 Effects of the initial configuration of a miscut substrate on thin film growth simulation

In this section, effects of the initial configuration of the miscut substrate are studied in order to find the optimum conditions for growth on a miscut substrate. The modification to the diffusion rule of the Das Sarma Tamborenea (DT) model with long surface diffusion length is used in this section. The substrate size is $L \times L = 960 \times 960$ sites and the diffusion length ℓ_d is fixed at 30 sites in all simulations. The initial configuration of the miscut substrate is shown in figure 4.1. Since the miscut angle is defined as $\theta = \tan^{-1}(s/L_t)$, studying effects of the initial configuration of the miscut substrate can be done by varying the terrace width (L_t) and the terrace height (s). In the first case, the terrace width L_t is varied from 20, 30, 96 to 480 sites and step height s is fixed at 1 site. For second case, the step height s is varied from 1, 3 to 5 sites and the terrace width L_t is fixed at 30 sites. Since the diffusion length is fixed at $\ell_d = 30$ sites, $\ell_d = L_t$ in this case.

4.2.1 Effects from the size of terrace width

4.2.1.1 Surface morphologies

Film morphologies from the modified DT model with ℓ_d=30 sites and L_t is varied from 20 to 480 sites are shown in figure 4.27. Film morphologies shown in figures 4.27(a) and (b) are examples of growth with near-perfect persistence. From the morphologies with $L_t < \ell_d$ in figures 4.27(a) and (b), shown after 100 MLs depositions, the initial configuration of the substrate can clearly be seen. However, when $L_t > \ell_d$ as shown in figures 4.27(c) and (d), small islands appear on the whole surface because the diffusion length is so small compared to the terrace width. It can be explained that when $\ell_d \geq L_t$, most atoms can move to step edge of the terrace so the initial pattern can survive for long period of time but when $\ell_d < L_t$, atoms cannot

(a) L_t= 20 sites

(b) L_t= 30 sites

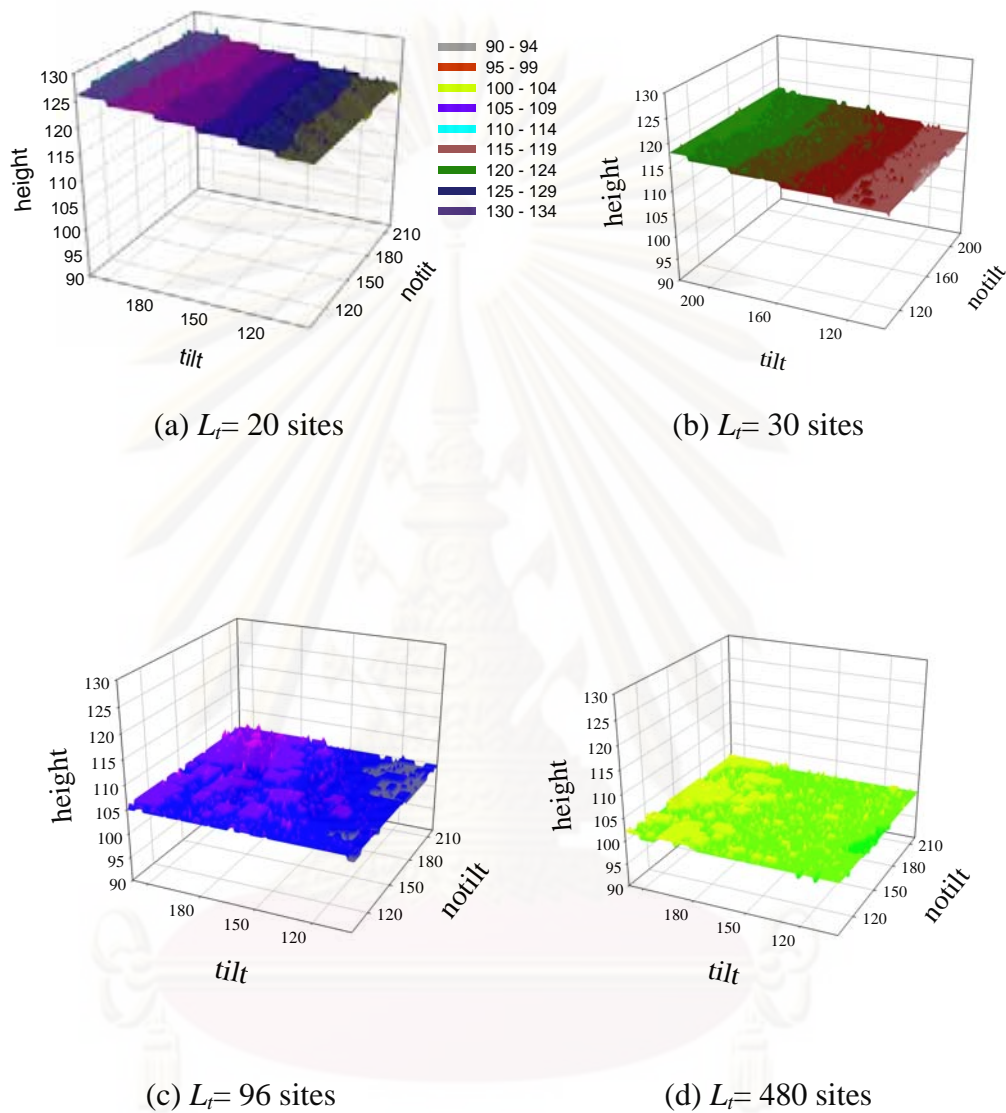(c) L_t= 96 sites

(d) L_t= 480 sites

Figuren4.27 : Film morphologies after 100 MLs deposition with ℓ_d =30 sites as the terrace width L_t is varied from (a) 20 sites, (b) 30 sites, (c) 96 sites and (d) 960 sites and s is fixed at 1 site.

reach the step edge and so they form new clusters-islands on the terrace. This is consistent with the *P-t* plot in figure 4.28.

4.2.1.2 Persistence probability

The persistence probability versus time or $P-t$ plot is shown in figure 4.28. From the graph, we found that the value of persistence probability for large L_t ($L_t = 96$ and 480 sites) rapidly drops to $P(t)=0$ in the early time. On the other hand, the value of persistence probability drops much slower from perfect value $P(t)=1$ for small L_t ($L_t =30$ and 20 sites). If the persistence probability remains at $P(t) =1$, it shows that the film can keep a completely set of initial pattern for the whole time. This perfect persistence occurs when $L_t \leq \ell_d$. These results are consistent with film morphologies in figure 4.28.

4.2.1.3 Correlation function

The calculated correlation functions are shown in figure in figure 4.29. The correlation functions are calculated when films are grown to 100 MLs. The plots in figure 4.29 exhibit a periodic pattern corresponding to the steps on the films along the *x* axis which is the direction of the tilt. On the other hand, there is not specific pattern in the $G(r_y)$ plots in figure 4.30. In the tilt direction, the periodic curve is shown in the saturated steady state. From the graph, we found that the flat square periodic curve is obtained since *t*=1 until t=10^2 MLs in the range $L_t \leq \ell_d$ .The square periodic oscillation indicates perfectly flat terrace. If the film is grown perfectly flat surface, the film is grown in the mode of the layer by layer growth. Due to most atoms form completely filled-layers and the small clusters little occur. However, as *t*=10^3 MLs, we found that the damped oscillation occurs at all ℓ_d . Damped oscillation
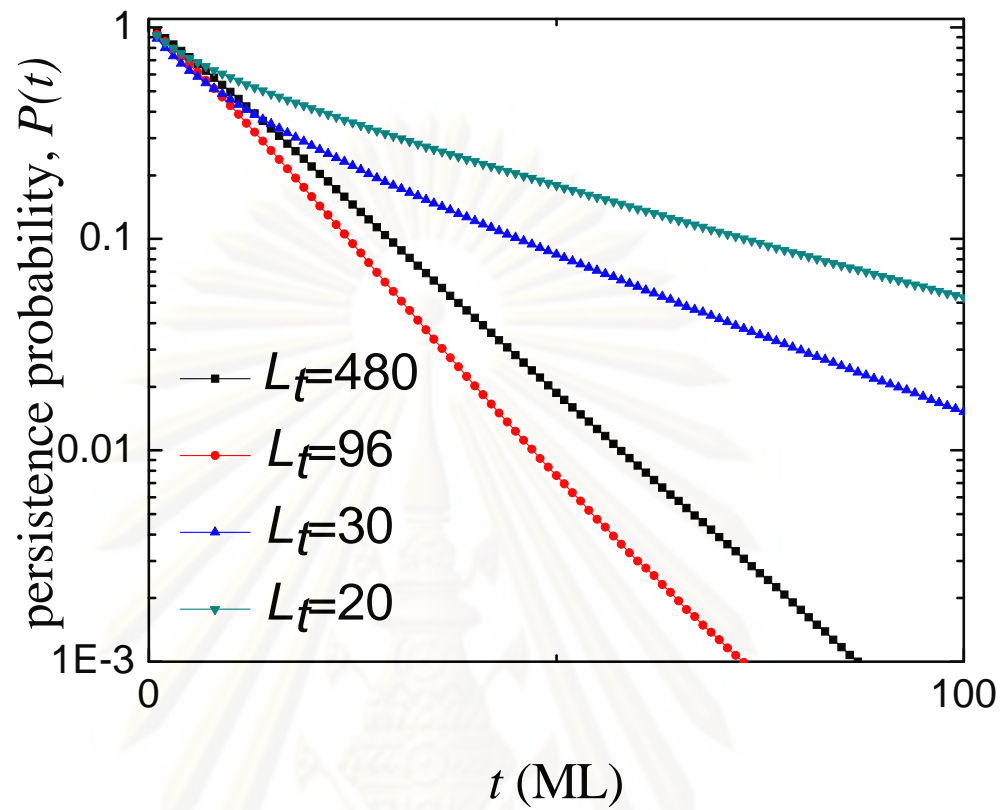
Figuren4.28: *P-t* plot as the length of terrace is varied from L_t =20, 30, 96 to 480 sites and step height is fixed at 1 site.

(a) $t= 1$ ML

(b) $t= 10$ ML

(c) $t= 10^2$ ML

(a) $t= 10^3$ ML
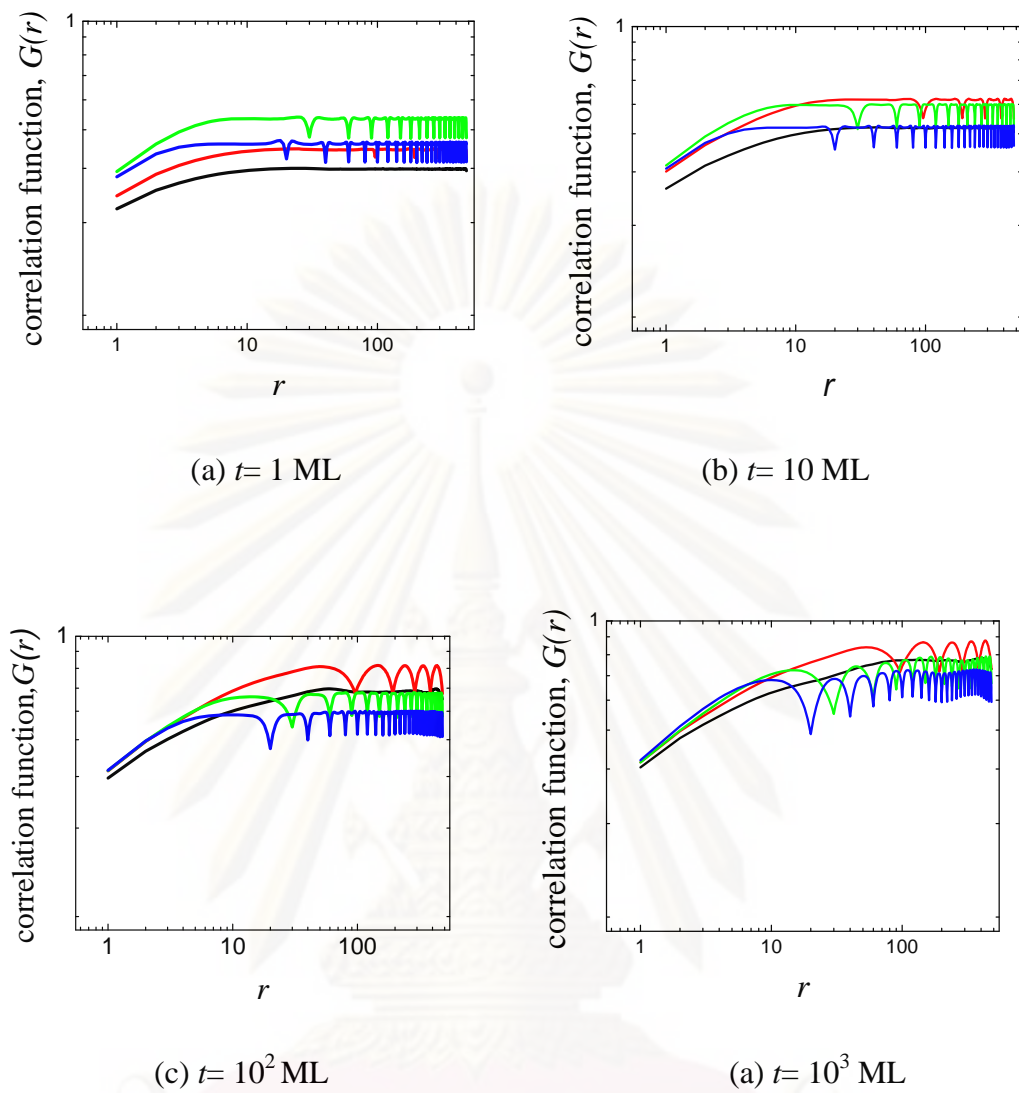
Figure 4.29: *G-r* plot in the tilted direction as the terrace width is varied from $L_t = 20$ (blue line), 30 (green line), 96 (red line) to 480 (black line) and the step height is fixed at 1 site.
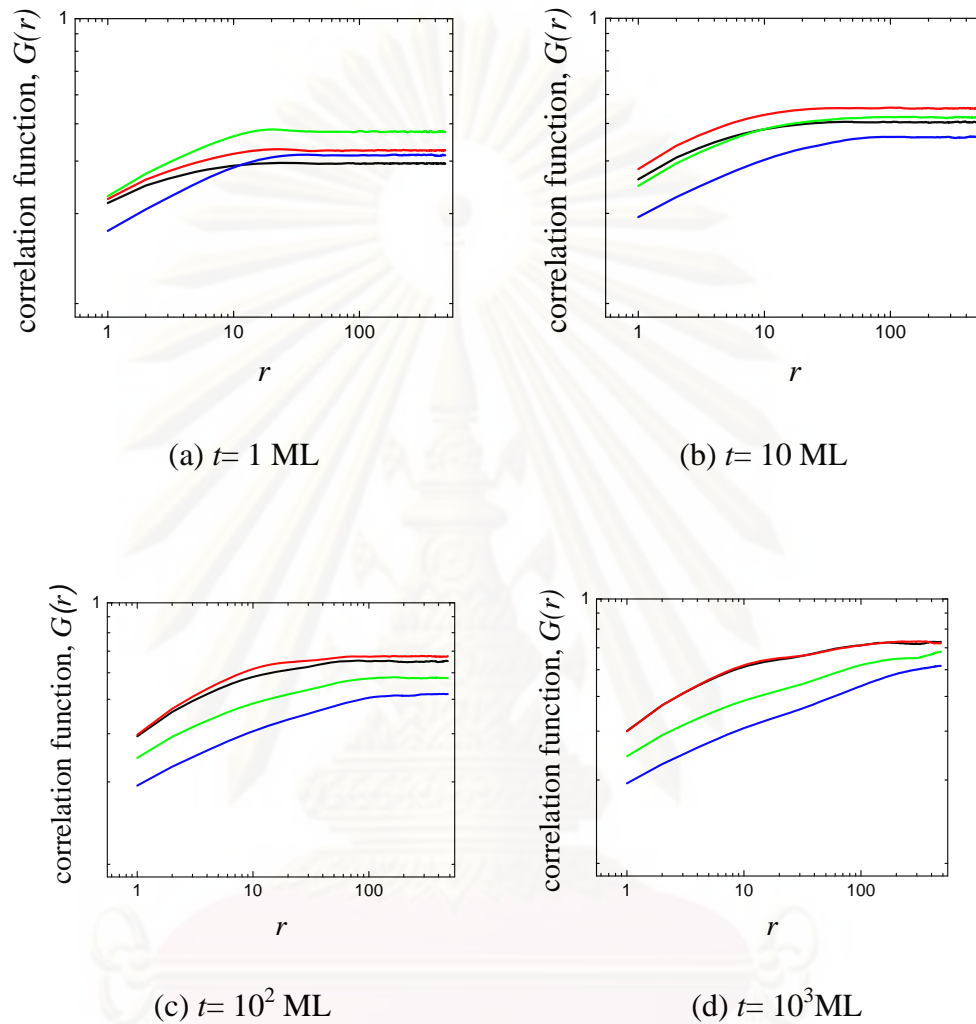
Figure 4.30: *G-r* plot in the notilted direction as the terrace width is varied from $L_t = 20$ (blue line), 30 (green line), 96 (red line) to 480 (black line) and the step height is fixed at 1 site.

indicates that small island disperses throughout the terrace. We consider the value of $G(r)$ as the terrace width is increased from 20 to 480 sites. We found that the value of $G(r)$ increased as the terrace width is increased. But result of $G(r)$ at 1 ML oppose other time. As we discussed, in the beginning, there is a few atoms on the surface. Tilted of substrate does not affect on film. For the notilted direction (figure 4.30), we found that the results of G-r plot in notilted direction is the same in the tilt direction for all ℓ_d. But, the difference is, there is not specific feature in the notilted direction. Interestingly, the first region that the function in notilted direction depends with r, we found that this region is longer as the terrace width decreases. It can explain as terrace width L_t is small compared with the surface diffusion length ℓ_d, correlation length ξ is long. On the other hand, if the terrace width is large compared with surface diffusion length ℓ_d, this region is short. Due to correlation length ξ is so small at large L_t. These results correspond with the modified DT with long surface diffusion length results.

For second definition of correlation function $H(r)$, H-r plot is shown in figure 4.31 and 4.32. Figure 4.31 presents $H(r)$ in tilted direction and figure 4.32 presents $H(r)$ in notilted direction. We found that the maximum value of $H(r)$ at $r = 0$ increases with time in the tilted and the notilted direction because of the very different height of two sites on the surface. However, the maximum value of $H(r)$ at $r=0$ decreases as decreasing the terrace width. Because, reducing roughness which causes the difference heights of film as the terrace width L_t is smaller than the surface diffusion length ℓ_d. From figure 4.31, there is not the curve of specific feature for large L_t (L_t =480) (in figure 4.31(a)). It shows that as $L_t \gg \ell_d$ induces the damaging to periodic pattern of film. Due to the roughness rapidly increases in this condition $L_t \gg \ell_d$. As we look $H(r)$ in the tilt direction, we found the results of $H(r)$ agrees with $P(t)$ and $G(r)$ results. This is, the initial pattern substrate can survive for a long period of time for $L_t \le \ell_d$. Because of the square periodic oscillation is shown in the interval time between 1 to 100 MLs. At 1000 MLs, the damped oscillation occurs at all L_t.

(a) $t = 1$ ML

(b) $t = 10$ ML

(c) $t = 10^2$ ML

(d) $t = 10^3$ ML
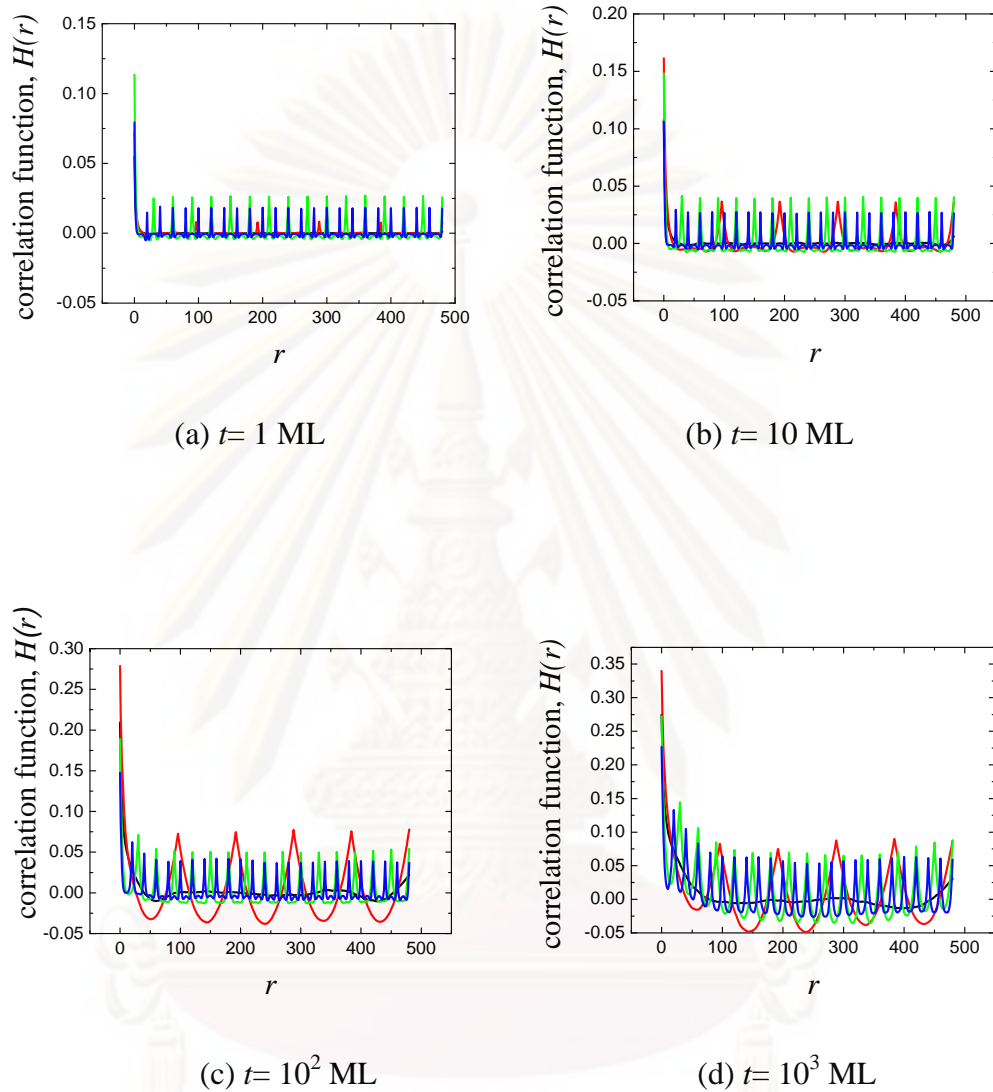
Figure 4.31: Correlation function with the distance r or H-r plot in the tilted direction as the terrace width is varied from $L_t = 20$ (blue line), 30 (green line), 96 (red line) to 480 (black line) and the step height is fixed at 1 site.

(a) $t = 1$ ML

(b) $t = 10$ ML

(c) $t = 10^2$ ML

(d) $t = 10^3$ ML

Figure 4.32: $H\text{-}r$ plot in the notilted direction as the terrace width is varied from $L_t = 20$ (blue line), 30 (green line), 96 (red line) to 480 (black line) and the step height is fixed at 1 site.
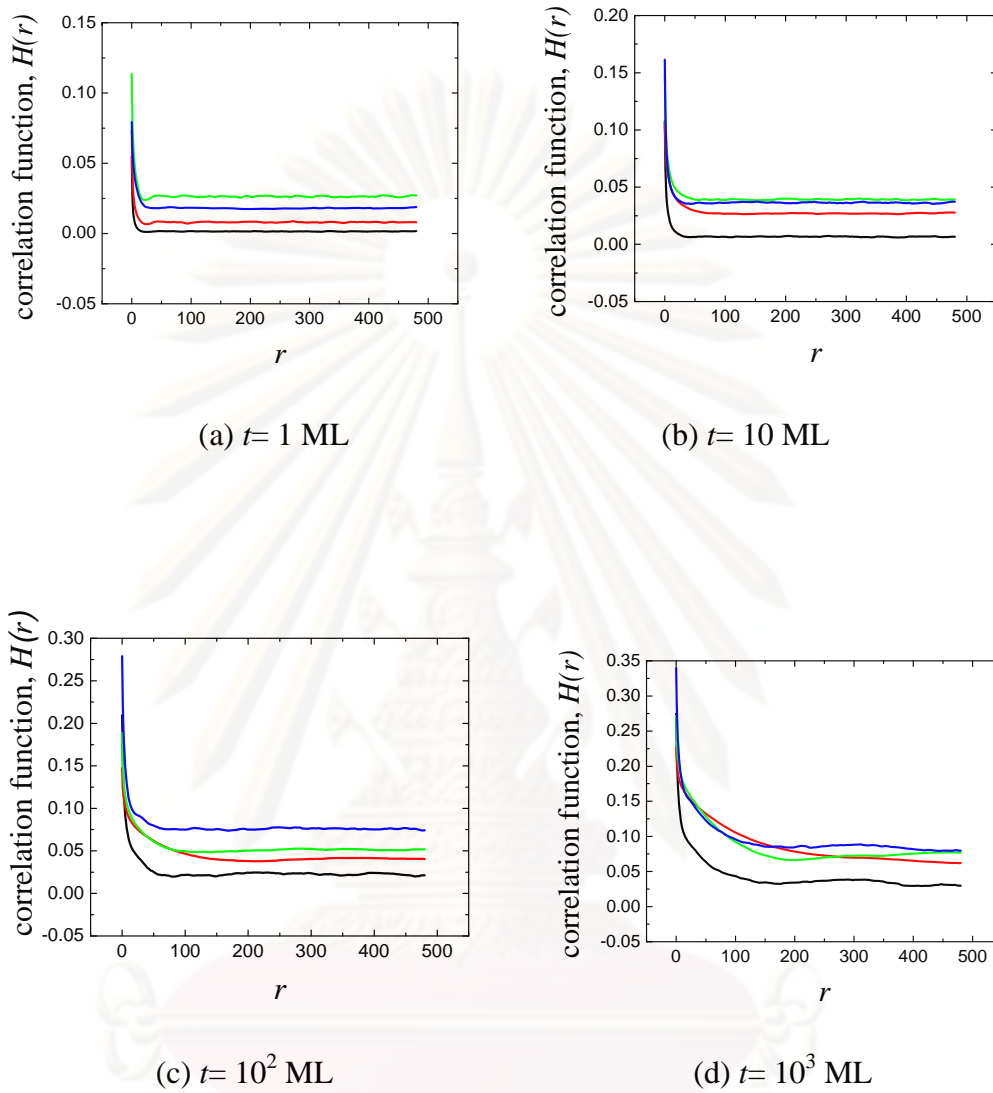
4.2.2 Effects from the size of the terrace height

In the second case, the step height is varied from 1, 3 to 5 sites and the terrace width is fixed at 30 sites which equals to the diffusion length ℓ_d.

4.2.2.1 Surface morphology

Film morphologies are shown in figure 4.33. These films are grown from 0 ML to 100 MLs. From film morphologies in figures 4.33(b) and (c), new steps are formed between original steps and the step height becomes one. Because the modified DT model is under solid on solid constrain, so atoms can not allow overhang. If an atom needs to hop down to the lower terrace, atom can not overhang. So, fronting steps appears as $s>1$. Forming new steps between the former steps is the cause of the rapid decreases in the persistence probability which is shown in figure 4.34.

4.2.2.2 Persistence probability

The *P-t* plot is shown in figure 4.34. In this situation with a fixed, $L_t = \ell_d$, the film should be able to keep the initial structure for a long time from previous results. But, from the graph of *P-t*, the rapid reduction of persistence probability occurs as $s>1$. It shows that higher step of terrace which is more than mono-stepped height induces roughness of the film surface. This result is the same as film morphologies results.

4.2.2.3 Correlation function

The quantity which gives information of specific feature of film G(r), is discussed as shown in figure 4.35 (tilted direction) and in figure 4.36 (notilted direction). In tilt direction, at small *r*, the function has linearly dependent upon *r*. we found that the length of this region nearly equals all *s*. But, the value of *G(r)* increases
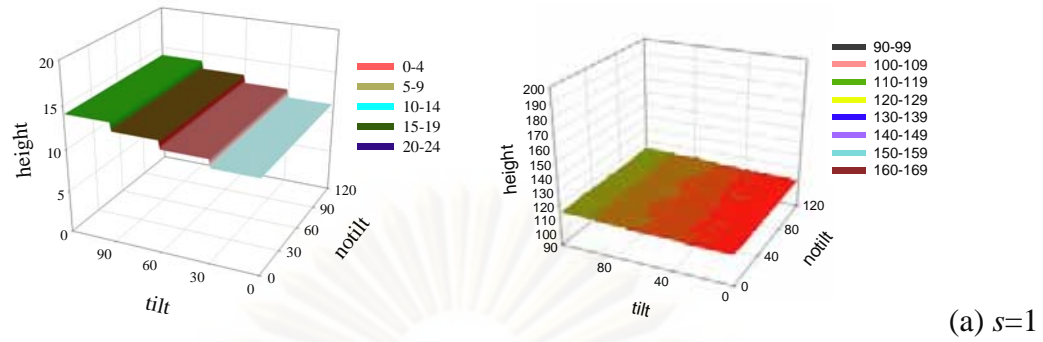
(a) *s*=1
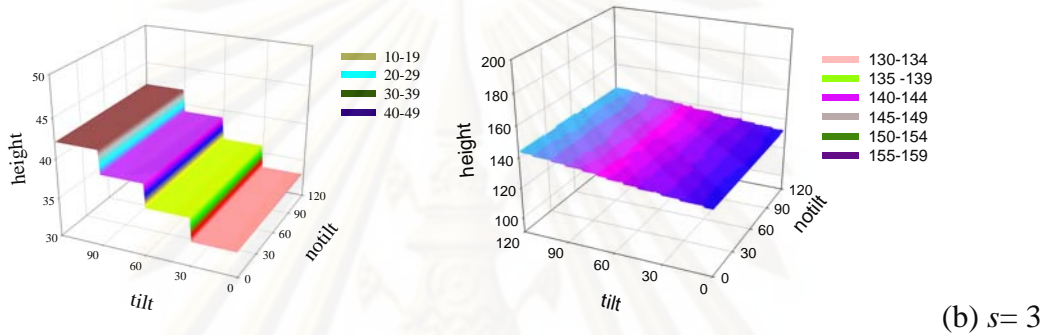
(b) *s*= 3
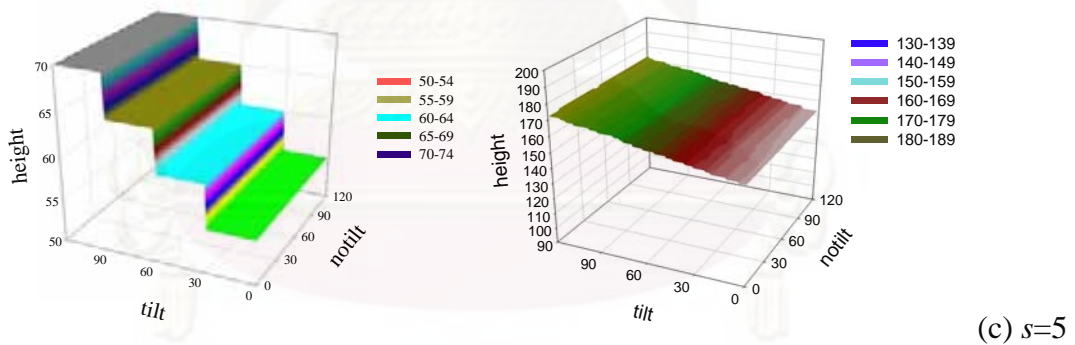
(c) *s*=5

t=0 ML   *t*=100 ML

Figuren4.33: Film morphologies from *t*=0 to100 MLs (left to right) deposition with ℓ_d =30 sites as the step height *s* is varied from (a) 1 sites, (b) 3 sites and (c) 5 sites and L_t is fixed at 30 site.
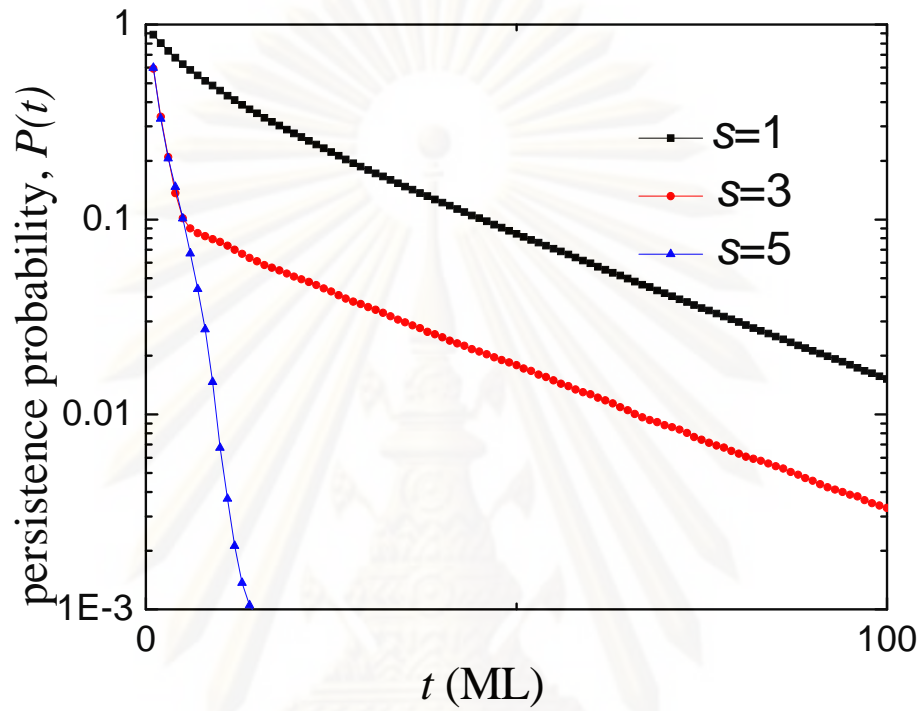
Figuren4.34: *P-t* plot as step height is varied from s=1 (black line), 3 (red line) to 5 (blue line) and terrace length is fixed at 30 site.

(a) $t = 1$ ML

(b) $t = 10$ ML

(c) $t = 10^2$ ML

(d) $t = 10^3$ ML
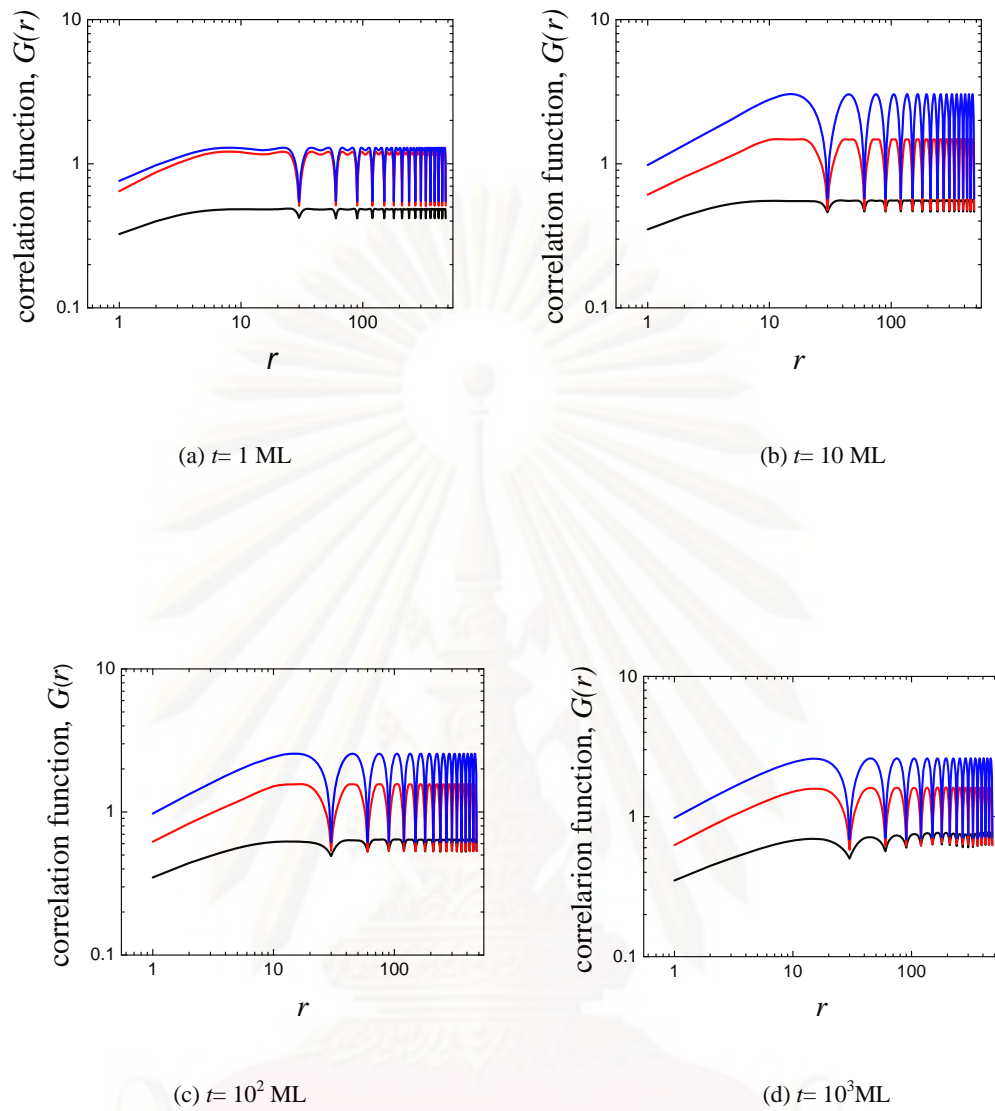
Figure 4.35: *G-r* plot in the tilted direction as the step height is varied from $s = 1$ (black line), 3 (red line) to 5 (blue line) sites and the terrace width is fixed at 30 site.

Figure 4.36: *G-r* plot in the notilted direction as the step height is varied from *s* =1 (black line), 3 (red line) to 5 (blue line) sites and the terrace width is fixed at 30 site.

with increasing the step height. It shows that as *s*>1, induces the difference of height (roughness) on the film surface. As we look *G-r* plot in the notilted direction, we found that slope of curve of *G-r* increases with increasing *s*. The slope in the range which *G(r)* depend on *r*, is roughness exponent $\alpha$. If roughness exponent is much, it shows that the film is quite rough. Increasing of *s* induces roughness of film. For *G-r* plot in saturated steady state, damping oscillation occurs in the early time for *s*>1. But *s*=1, the damped oscillation occurs in the large time (10^3 MLs).

The result of *H(r)* corresponds to these results. *H-r* plot in the tilted and notilted direction is shown in figure 4.37 and 4.38, respectively. In the tilt direction, we found that the multiplicity of heights clear increase as *s*>1 due to the difference height of two sites on the film. As *s*=5, we found that each peak position is quite high. If the multiplicity of height is high, it indicates that the height of sites differ greatly. On the other hand, for mono-stepped height (*s*=1), the periodic curve has the same size and shape remains at all time. Because, this condition ($\ell_d \geq L_t$ and *s*=1) is backed into the previous result. So, the initial pattern substrate persists for a long time in this condition. *H-r* plot in the notilted direction as shown in figure 4.38 provides the same result. This is, the multiplicity height increases as *s*>1. At *r*=0, the maximum value of *H(r)* in both direction is larger as *s*> 1. It shows that this condition (*s*>1) stimulate roughness of film.

From all results of the first case, decreasing the terrace width which equals to increasing the number of the terrace, reduces the roughness of film. As $L_t \leq \ell_d$, the remaining initial configuration of tilt substrate can be clearly seen. This result agrees with result of DT model with long surface diffusion length. It mentioned that film is grown in layer by layer growth mode for a long time as $\ell_d \geq L_t$. As we looks variable *s*, we can conclude that *s*>1 induces roughness of film surface because the grown film tries to reduce the height of terrace as *s*>1. So, the initial structure has changed rapidly. From the results of both cases, it agrees with studying dynamic behavior during growth on surface by varying tilt (Pal and Landau, 1996). This work shows that increasing the number of terrace suppresses the surface roughness and the surface roughness is larger when the height of terrace is high.

(a) t= 1 ML
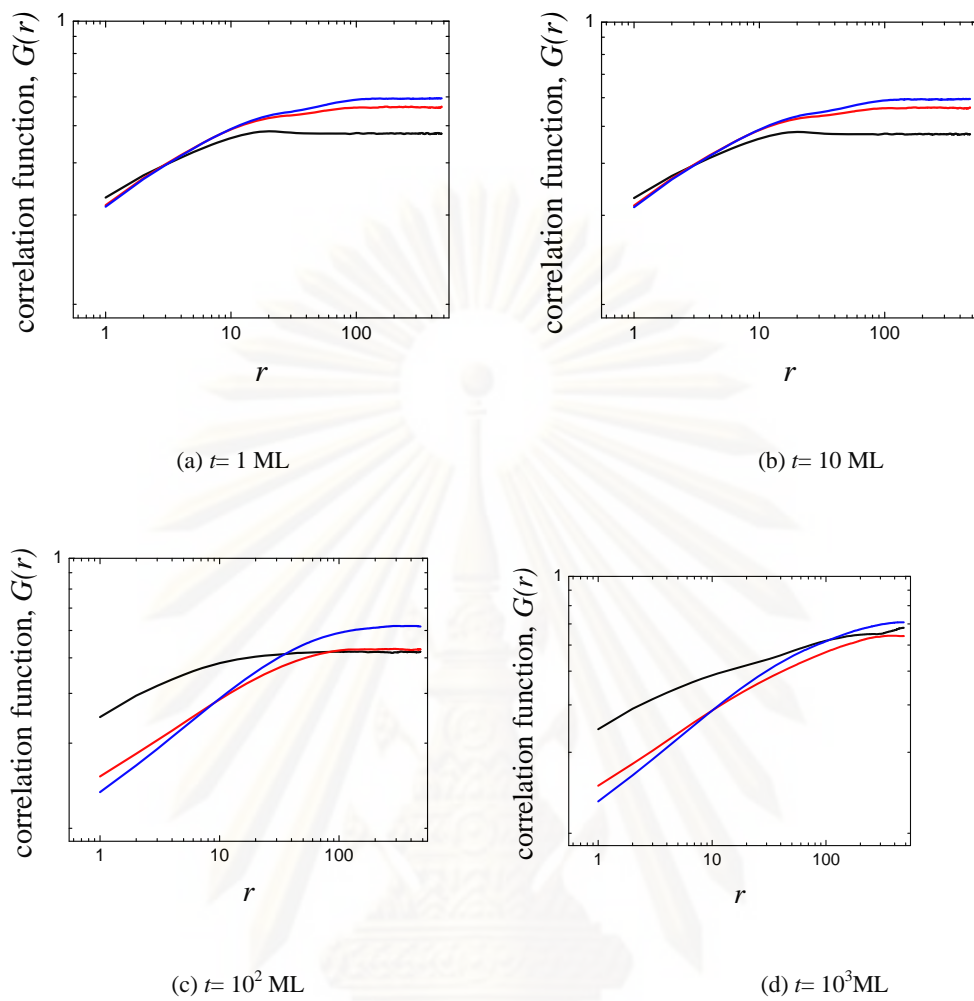
(b) t= 10 ML

(c) t= 10^2 ML

(d) t= 10^3ML

Figure 4.37: *H-r* plot in the tilted direction as the step height is varied from s =1 (black line), 3 (red line) to 5 (blue line) sites and the terrace width is fixed at 30 site.

(a) $t= 1$ ML

(b) $t= 10$ ML

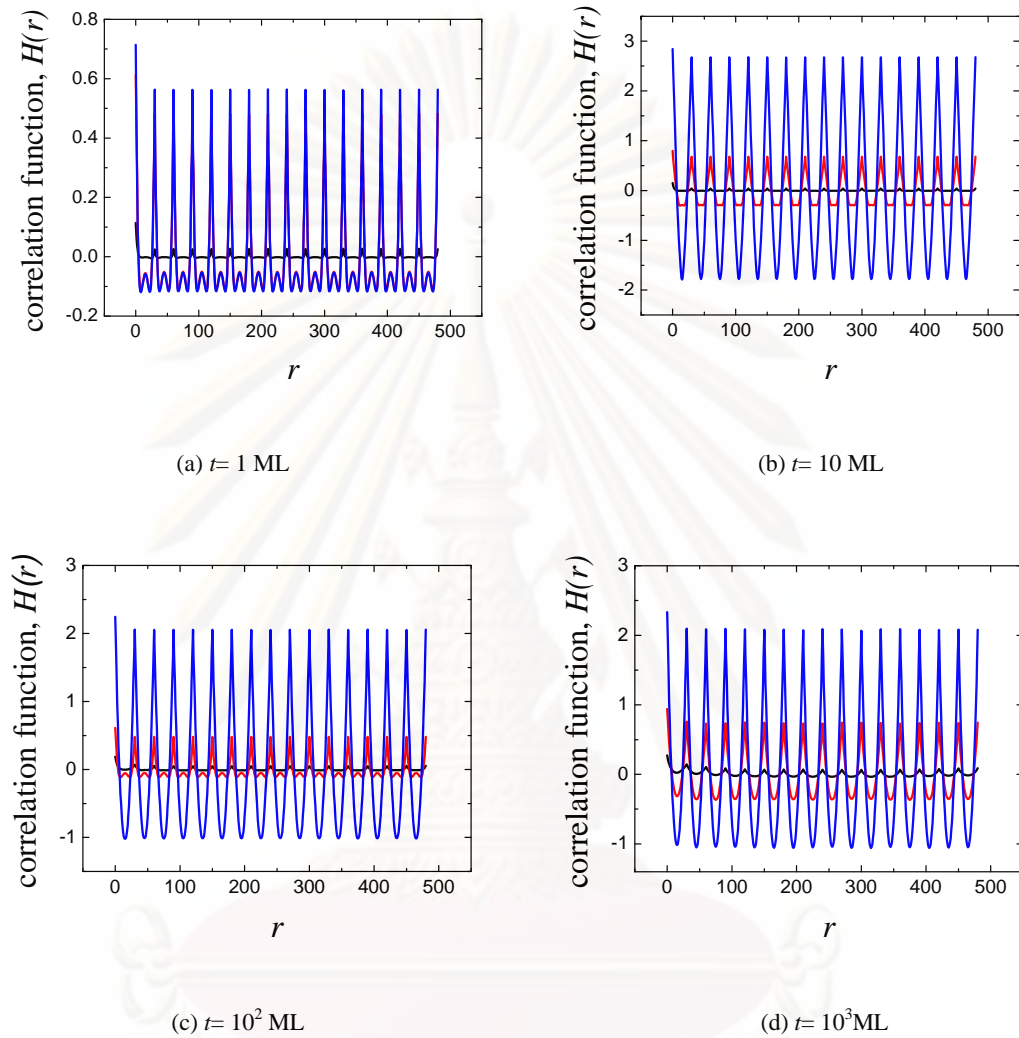(c) $t= 10^2$ ML

(d) $t= 10^3$ ML

Figure 4.38: *H-r* plot in the notilted direction as the step height is varied from *s* $=1$ (black line), 3 (red line) to 5 (blue line) sites and the terrace width is fixed at 30 site.
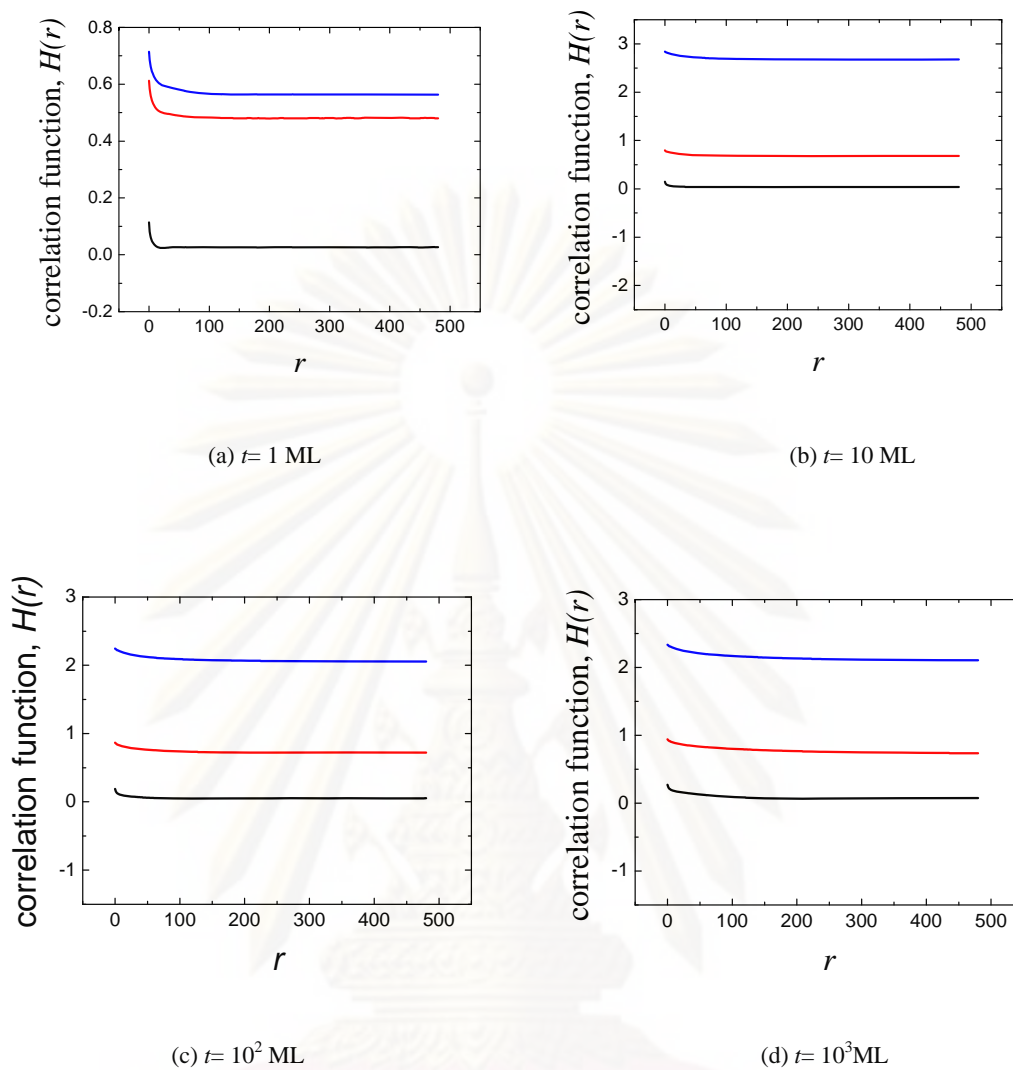
CHAPTER V

CONCLUSIONS

In the thesis, the main purpose is to study effects of the miscut substrate on the thin film growth process and to find the optimum condition for growth on a miscut substrate. The Das Sarma- Tamborenea (DT) model is used to study. The results agree with the original DT model on a flat substrate. To see effect of the substrate temperature, long surface diffusion length noise reduction technique is added to the original DT model. In the NRT technique, we define the parameter ℓ as a maximum number of times an atom can hop to find the final site under diffusion rule of the model. According to random walk theory, the end-to-end distance is proportioned to (number of step). Results from the DT model with long surface diffusion length on substrates with the same initial angle, we found that the change in the diffusion length is not clearly be seen in the simulated films. As the diffusion length is increased, the morphology is still rough which disagrees with experimental results. Statistical quantity such as the interface width, persistence probability and correlation functions corresponds morphology result. This is the curve of interface width, persistence probability and correlation functions provide the same results at various diffusion length. These results are quite unrealistic and unbelievable. A possible cause is limitation in the diffusion rule of the DT model. Therefore, a modification to the diffusion rule of the DT model is used for the simulations.

When using the modified to study, we found that the results of the modified DT model better agree with experiments. Using the modified DT model for the thin film growth on a tilted substrates, we found that the surface morphology is smoother when diffusion length increases corresponding statistical quantities, surface morphology, interface width persistence probability and correlation functions. Moreover, the perfect layer by layer growth mode is obtained and the film can maintain the initial angle for a long time when the diffusion length substrate temperature is high: $\ell_d \geq L_t$. This condition is optimum for the grown on the same

initial miscut angle. On the other hand, the film is quite rough because there is some void and small cluster for the film surface as $\ell_d < L_t$.

Effects of the initial configuration tilted substrate, The persistence probability and film morphology show that, as the terrace width (L_t) is varied and the step height of the terrace (s) is fixed, the film can maintain the initial tilted configuration for a long time when $L_t \leq \ell_d$. For correlation functions, specific pattern can be seen for a long time in this condition. On the other hand, when the terrace width (L_t) is fixed and the step height (s) is varied, we found that initial configurations with $s > 1$ can not be maintained through growth process and the persistence probability of the grown films quickly decay. Information of the specific feature on the miscut substrates changes with time at large s. In conclusion, the optimum conditions for the miscut substrate are $L_t \leq \ell_d$ and s=1 because the films simulated which that conditions are grown in near perfect step flow growth mode.

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Conference Presentations:

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2009 C. Chomngam and P. Chatraphorn. Effects of initial configuration of a miscut substrate on thin film growth simulations. Siam Physics Congress 2009, Cha-Am, Phetchburi, Thailand (19-21 March 2009).

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