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นางสาวจินดารัตน์ แย้มวงษ์

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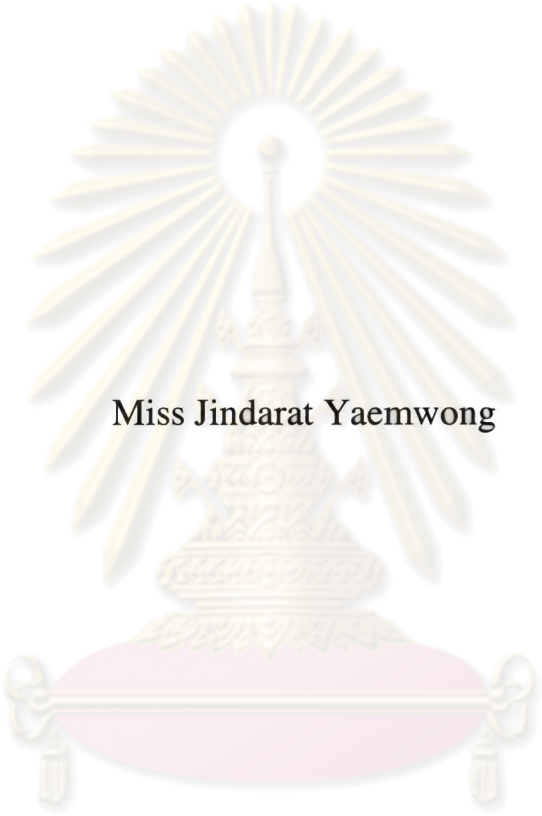
สาขาวิชาฟิสิกส์ ภาควิชาฟิสิกส์
คณะวิทยาศาสตร์ จุฬาลงกรณ์มหาวิทยาลัย
ปีการศึกษา 2553

MONTE CARLO SIMULATIONS OF STEP FLOW IN MOLECULAR BEAM EPITAXY GROWN SURFACES

Miss Jindarat Yaemwong



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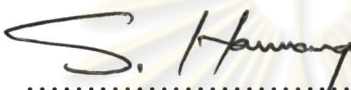
By	Miss Jindarat Yeamwong
Field of Study	Physics
Thesis Advisor	Assistant Professor Patcha Chatraphorn, Ph.D.

Accepted by the Faculty of Science, Chulalongkorn University in Partial Fulfillment of the Requirements for the Master's Degree



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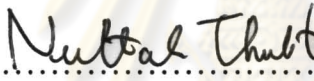
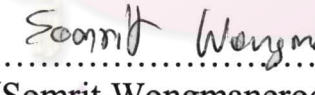


..... Chairman
(Assistant Professor Nuttakorn Thubthong, Ph.D.)

..... Thesis Advisor
(Assistant Professor Patcha Chatraphorn, Ph.D.)



..... Examiner
(Somrit Wongmanerod, Ph.D.)

..... External Examiner
(Assistant Professor Wandee Onreabroy, Ph.D.)

จินดารัตน์ แย้มวงษ์ : การจำลองแบบมอนติคาร์โลของการไหลแบบขั้นบันไดในผิวที่ปลูกโดยโมเลกุลาร์บีมเอพิแทกซี. (MONTE CARLO SIMULATIONS OF STEP FLOW IN MOLECULAR BEAM EPITAXY GROWN SURFACES) อ. ที่ปรึกษาวิทยานิพนธ์หลัก : ผศ.ดร. ปัจฉา ฉัตราภรณ์, 59 หน้า.

การจำลองการปลูกฟิล์มบางแบบโมเลกุลาร์บีมเอพิแทกซีบนซับสเตรตแบบขั้นบันได ถูกศึกษาโดยใช้เทคนิคมอนติคาร์โล พื้นผิวแบบขั้นบันไดประกอบด้วยแผ่นเรียบแบนกว้างเท่ากันแต่ละขั้นแบ่งโดยขอบบันไดที่มีความสูงแตกต่างกันซึ่งเป็นชนิดของซับสเตรตที่ใช้กันอย่างกว้างขวางในกระบวนการผลิตฟิล์มบาง ซับสเตรตชนิดนี้เป็นที่รู้จักกันดีว่าสามารถใช้ในการผลิตฟิล์มบางคุณภาพสูงเมื่อปลูกในโหมดการไหลแบบขั้นบันได จุดประสงค์ของงานนี้คือการหาเงื่อนไขสำหรับการปลูกฟิล์มบางในโหมดการไหลแบบขั้นบันไดบนพื้นผิวแบบขั้นบันไดและศึกษาคุณสมบัติเชิงสถิติของการปลูกฟิล์ม ในงานนี้ใช้แบบจำลองดาส ซาร์มา-แทมโบเรเนียและแบบจำลองแฟมิลี่ แบบจำลองทั้งสองใช้ศึกษากรณีการเคลื่อนที่แบบจำกัดได้ดี เราได้ปรับปรุงแบบจำลองเล็กน้อยโดยใช้เทคนิคการลดความหยาบของฟิล์มด้วยการเพิ่มระยะการแพร่ซึ่งทำให้การเคลื่อนที่ของอะตอมในแบบจำลองเปลี่ยนแปลงไป เนื่องจากการเคลื่อนที่ของอะตอมขึ้นอยู่กับอุณหภูมิของการปลูกฟิล์ม การเปลี่ยนแปลงระยะการแพร่ที่เป็นผลมาจากอุณหภูมิของการปลูกฟิล์มในการศึกษานี้จึงสามารถตรวจสอบได้ ปริมาณที่คำนวณหาคือฟังก์ชันสหสัมพันธ์เชิงเวลา, การแจกแจงความน่าจะเป็นของการคงอยู่และการดำรงอยู่ ผลจากการจำลองแสดงว่าโหมดการไหลแบบขั้นบันไดจะเริ่มเกิดเมื่ออุณหภูมิสูงพอจนกระทั่งระยะทางการแพร่ของอะตอมมีค่าประมาณครึ่งหนึ่งของความกว้างขั้นบันได กระบวนการเชิงอะตอมที่พบในการปลูกฟิล์มในโหมดการไหลแบบขั้นบันไดคืออะตอมทับถมกันที่ขอบบันไดและอะตอมกระจายตัวบนเนินของขั้นบันได ทั้งการแจกแจงความน่าจะเป็นของการคงอยู่และการดำรงอยู่จะลดลงตามเวลา เลขชี้กำลังของความน่าจะเป็นของการคงอยู่ซึ่งก็คืออัตราความน่าจะเป็นที่ลดลงจะมีค่าน้อยเมื่อปลูกฟิล์มในโหมดการไหลแบบขั้นบันได

ภาควิชา..ฟิสิกส์..... ลายมือชื่อนิสิต.....

สาขาวิชา..ฟิสิกส์..... ลายมือชื่อ อ.ที่ปรึกษาวิทยานิพนธ์หลัก.....

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5172241223 : MAJOR PHYSICS
KEYWORDS : VICINAL SURFACE / STEP-FLOW GROWTH / DAS-SARMA TAMBORENEA MODEL / FAMILY MODEL / TEMPORAL CORRELATION FUNCTION / PERSISTENCE PROBABILITY DISTRIBUTION / SURVIVAL PROBABILITY DISTRIBUTION

JINDARAT YAEMWONG : MONTE CARLO SIMULATIONS OF STEP FLOW IN MOLECULAR BEAM EPITAXY GROWN SURFACES. ADVISOR : ASSISTANT PROFESSOR PATCHA CHATRAPHORN, PH.D., 59 pp.

Molecular beam epitaxy growth on vicinal substrates are studied using the Monte Carlo simulation technique. A vicinal surface consists of equal-width flat terraces of different height separated by step edges. It is a type of substrate widely used in thin film fabrication. It is well-known that high quality films can be obtained when the growth is in step flow mode. The aim of this work is to find growth conditions that lead to step flow growth on vicinal substrates, and to study statistical properties of the grown films. Two discrete growth models, the Das Sarma-Tamborenea and the Family models, are used for this task. Both models have been well-studied for limited mobility cases. A slight modification, adding long surface diffusion length noise reduction technique, is made so atomic mobility can be varied in these models. Since the atomic mobility depends directly on the growth temperature, varying the diffusion length in the study means effects of the growth temperature can be investigated. Quantities of interest are morphology, temporal correlation function, persistence probability and survival probability distribution. The simulated results show that the step flow mode begins when the growth temperature is high enough so that the atomic diffusion length is approximately half of the terrace width. Atomic processes found during the step flow mode are deposition at the step edges and diffusion on the terraces. Both persistence and survival probability distributions decrease with growth time. The persistence exponent, which is the rate that the probability decreases, is small when the growth is in the step flow mode.

Department : Physics Student's Signature .....
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Chapter I
Introduction

The surface study plays an important role for technology development. Surface physics is crucial in computer chip manufacturing processes (Stasevich and Einstein, 2007). Thin film growth is a well-known technique for creating nanoscale electronic device. Figure 1.1 shows gold surface used in thin film growth.

The roughness in thin film growth process depends on orientation of the chosen substrates. The substrates can be categorized to either flat or vicinal (or stepped) substrate. The atoms that are incorporated on the flat substrates make the film rough and can create mounds on the films (Vilone, Castellano and Politi, 2005). Stepped surface, however, is an efficient substrate in thin film growth technology. Presently, there are many research groups working on stepped surfaces (Constantin et al., 2007; Balykov and Voigt, 2005; Zhang, et al., 2006). In molecular beam epitaxial growth, stepped substrates are used in fabrication of semiconductor films. Au nanowire is an example of electronic devices grown on Mo stepped surfaces (Yakovkin, 2005). Interaction and dynamics of atoms on the film surface as well as evolution of morphologies such as steps on this surface are investigated. It was found that atoms deposited on stepped substrates can be incorporated to be a part of the film easier than those on the flat substrates. This is why stepped substrates are widely used in experiments and this is why we chose to study the growth on this type of substrates.

During the growing process, diffusion of surface atoms depends on the rate of the deposition flux and the diffusion rate that varies with the growth conditions (Schinzer et al., 1999). The temperature is clearly one of the main factors in these thin film growth processes. Diffusion rate varies with the substrate temperature according to the Arrherius diffusion rate (Ehrlich and Hudda, 1966; Schwoebel and Shipsey, 1966):

$$R = \nu_0 \exp\left(\frac{-E_x}{k_B T}\right) \qquad (1.1)$$

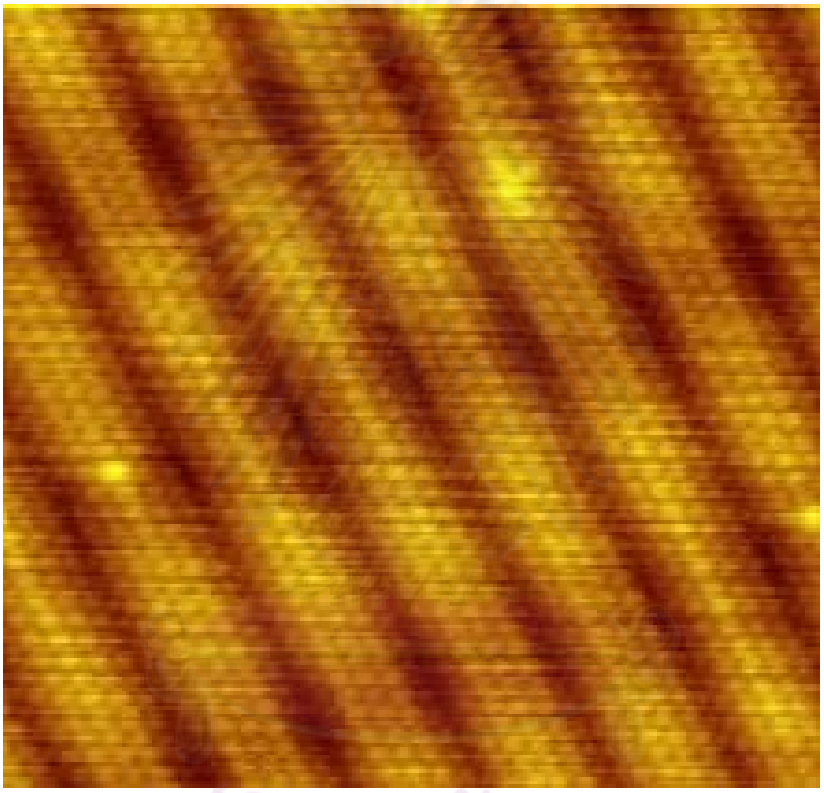


Figure 1.1 : Gold (100) surface from scanning tunneling microscopy. (Erwinrossen, 2008 : online)

where ν_0 is a typical atom vibration frequency, T is the substrate temperature and E_x is the hopping barrier. The barrier E_x includes a term due to interaction with the substrate E_d and interactions with each lateral nearest neighbor atom E_a :

$$E_x = E_d + nE_a \tag{1.2}$$

where n is the number of nearest neighbor (Hamouda, Pimpinelli and Phaneuf, 2008). When the temperature is high, atoms diffuse with high diffusion rate making deposition flux small by comparison. From the Arrherius diffusion rate, it can be seen that an atom at a site with large n has small hopping rate. So the probability that atoms get incorporated at the step edge, which provides at least two nearest neighbors, is much larger than incorporation on the terraces.

In the step-flow growth mode, the steps move slowly over the terraces through the deposition of each monolayer (Stasevich and Einstein, 2007). Average width of the terrace is constant during the step-flow growth process (Zhu, 1998). Because diffusion length increases with growth temperature, higher growth temperature contributes to better step-flow during this epitaxial growth process. When the step-flow growth mode is stable, island formations are negligible (Hong et al., 2005). The grown thin films look very smooth and maintain the “look” of the initial substrate with this technique.

The diffusion processes can be described by the temporal correlation function (Karim et. al., 2004). The temporal correlation function depends on how step and terrace exchange atoms (Giesen-Seibert et al., 1995). Microscopic mechanisms are studied from atomic steps on crystal surface. Time-dependent of step fluctuation function and the persistence probability distribution are used to consider the diffusion activity along the steps (Verhoeven and Frenken, 2007). Persistence probability distributions are quantities that can predict and describe the characteristic of step position. Survival probability distributions are calculated in the growth processes. These probabilities can be used to classify mode of thin film growth (Constantin et al., 2004). Step fluctuation in nanostructure can be investigated via survival probability.

Through experimental methods in macroscopic scale using scanning tunneling microscopy (STM)), low energy electron microscopy (LEEM) and reflection electron microscopy (REM), some understanding of microscopic processes of step fluctuation are obtained (Giesen-Seibert et al., 1993). From experimental results of Cu(1 1 79) surface using

temperature between 310 K and 600 K, the log-log plots of the temporal correlation function and time show that the correlation functions increase with time with a power law relation. The data in this process cannot be observed when the temperature is higher than 600 K (Giesen-Seibert et al., 1995). This limit in experiments encourages simulation researchs. The simulation technique can be used to explain and predict the results when temperature increases. The SOS models can be used to describe specific growth process of MBE (Schinzer et al., 1999). Monte Carlo simulation was used to simulate morphologies.

Thin films grown on stepped surfaces by MBE in step-flow growth mode are studied in this work. We use the Das-Sarma Tamborenea (also known as DT) model (Das Sarma and Tamborenea, 1991; Tamborenea and Das Sarma, 1993) and the Family model (Family, 1986) to simulate thin film. The substrate temperature is the varying factor in this work. The temperature in these models is varied by adjusting the number of time an atom can diffuse. The most interesting issue in this work is to find growth conditions to grow films with similar structure as the initial stepped surfaces. The time fluctuation of steps is studied with the mean-square step displacement fluctuations or the temporal correlation function. The persistence and the survival probability distributions are used to show evolution of morphology and describe step motion.

The models that were used in this research are explained in chapter 2. Chapter 2 also describes the definitions of quantities of interest. In chapter 3, we clarify the process of computer simulation to calculate and analyze data in this study. The results and discussion are shown in chapter 4. Finally, in chapter 5, we conclude the results that we have achieved in this work.

Chapter II
Theory

2.1 Models

There are many atomistic models introduced for the purpose of studying thin film growth process (Barabasi and Stanley, 1995). These models have different diffusion rules, created in order to investigate different complicated behavior of atoms in the growth process. In the rules of the models, complex conditions are simplified and only one or two aspects are investigated in detail. In this thesis, two such models are chosen : the Das Sarma-Tamrenea (DT) model (Das Sarma and Tamborenea, 1991) and the Family model (Family, 1986). The DT model is a simple bond-counting model which has been shown to reproduce similar results as the complicated and time-consuming MBE model in basic growth conditions (Das Sarma and Tamborenea, 1991). Here, it is chosen as a model to study MBE growth on vicinal substrates with adjustable growth temperature. The Family model, on the other hand, is known to produce very smooth surface in two dimensional substrates (Family, 1986). It is chosen for this work to compare with the DT model although the diffusion rules of the Family model do not have a real connection to MBE growth process. Both models are under solid-on-solid (SOS) constrain which means evaporation, bulk vacancy and overhanging are not allowed.

2.1.1 Das Sarma-Tamborenea Model

In the DT model (Das Sarma and Tamborenea, 1991; Tamborenea and Das Sarma, 1993), an atom is deposited at a random site on the substrate. If it is deposited at a site with at least two bonds, it cannot diffuse. If an atom has only one bond at the deposition site, it diffuses randomly to one of its nearest neighbors in order to increase bonds. Although the atoms diffuse to increase bonds, the final sites do not necessary have the maximum bonds (Das Sarma and Tamborenea, 1991). The DT diffusion rules are expressed in figure 2.1. The white space shows the substrate and the gray boxes show diffusing atoms. Atom **a** diffuses to the right because there is two bonds. Here, there is only one bond

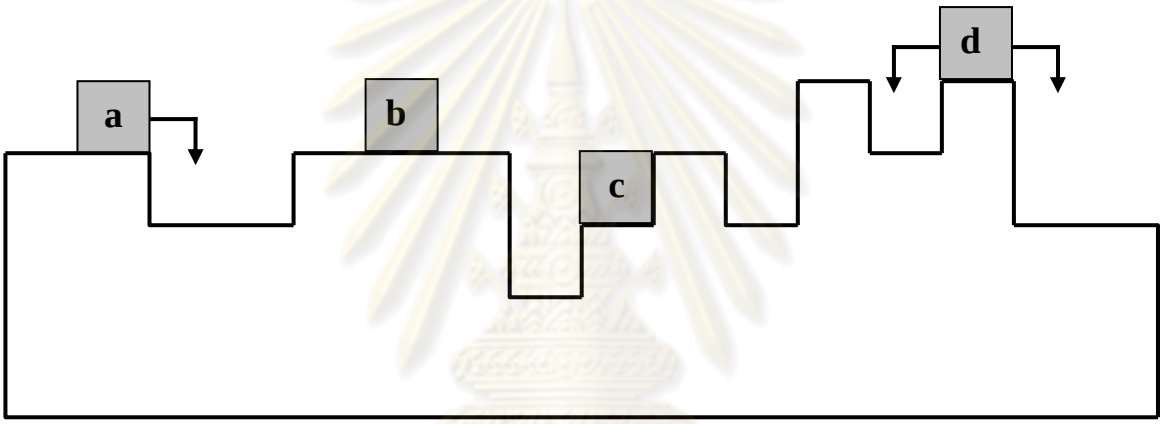


Figure 2.1 : Diagram of atom diffusion with diffusion rule of the DT model**.**

at the left neighbor and at the deposition position. Atom **b** cannot diffuse to any site because both neighbors provide only one bond each. Atom **c** cannot diffuse to other positions because there are already two bonds at its site. Atom **d** chooses randomly one site from the two neighbors. The probabilities of diffusing to one of two neighbors are the same. After the diffusing atom diffuses once, it is incorporated at the final site and stays there permanently. The whole deposition/diffusion process is repeated until the film is grown to the preferred thickness.

2.1.2 Family Model

In the Family model (Family, 1986), atom diffuses to find a nearby site with the minimum height. The atoms can diffuse following the diffusion rule of the Family model as shown in figure 2.2. Atom **a** must diffuse to the right which is the lowest nearest neighbor site. Atom **b** cannot diffuse to nearest neighbor sites because the sites are at the same level as the deposition site of atom **b**. Atom **c** chooses to diffuse to the left site following the diffusion rule. Atom **d** chooses the right site with the minimum height. With the Family diffusion rule, atoms can diffuse one time before incorporation.

2.2 Long Surface Diffusion Length Noise Reduction Technique (NRT)

The noise in thin film growth is a cause of roughness in the grown film. It can be decreased by noise reduction technique (NRT). The film is much smoother when NRT is applied in the simulation process. There are minor modification in the diffusion rules for NRT (Punyindu, 2000). Normally, a diffusing atom diffuses only one time. NRT is based on increased temperature in this work. If substrate temperature increases, atoms have enough energy to break bonds from its deposited site. The diffusing atoms are allowed to search as far as "l_d" (diffusion length) sites away from its deposition site (Das Sarma and Punyindu, 1997). Diffusion length in this case is larger than normal. It is found that the diffusion length $l_d \propto \exp\left(\frac{-E_x}{k_B T}\right)$ (Barabasi and Stanley, 1995), showing that l_d increases with the substrate temperature T. It leads to reduced noise and smoother films. Thin film growth on flat substrates reaches layer-by-layer growth mode (Punyindu, 2000) and we speculate that growth on stepped substrates will be in the step-flow growth mode.

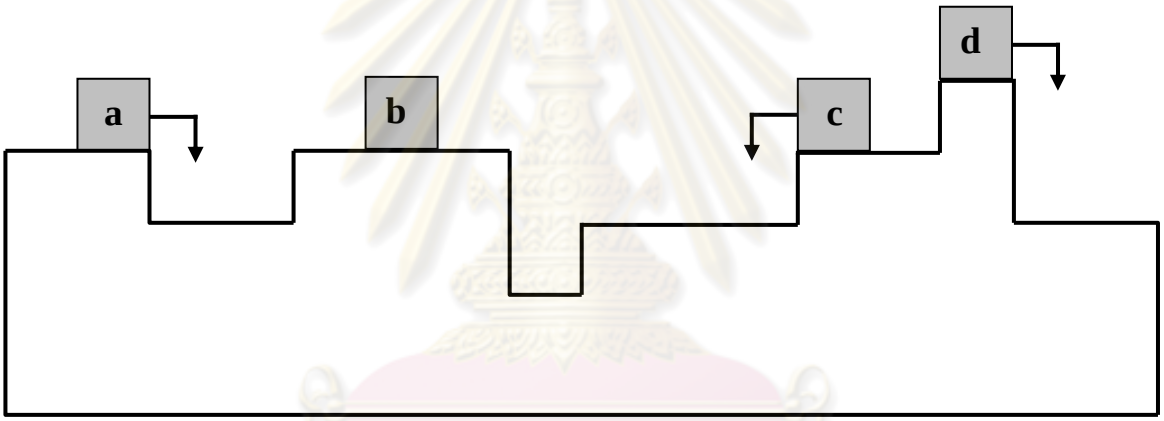


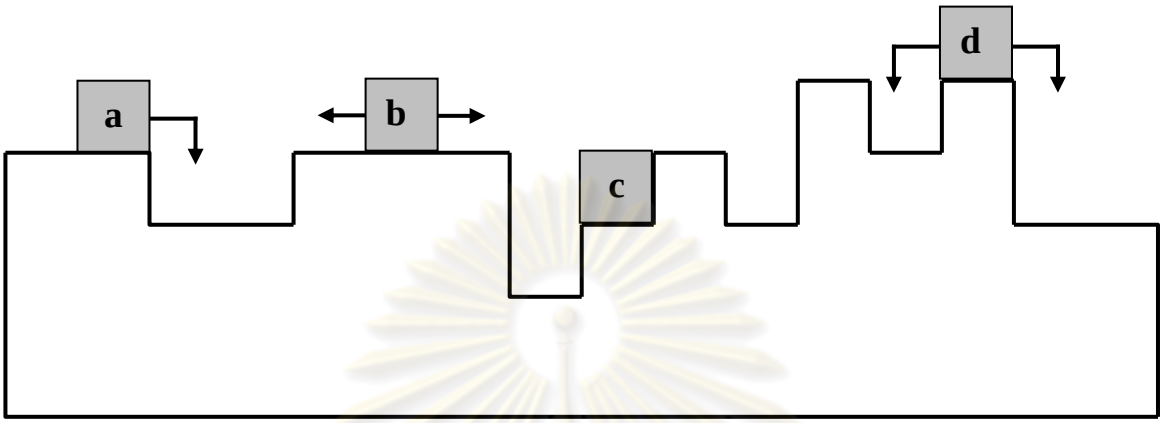
Figure 2.2 : Diagram of atom diffusion with diffusion rule of the Family model**.**

2.2.1 DT with NRT

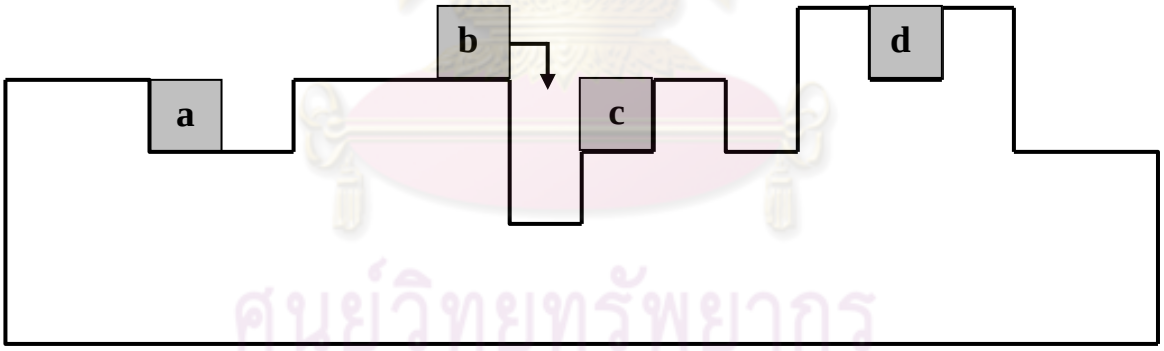
Atom behavior for increased temperature in this DT model is adapted. Figure 2.3(a) shows the atoms diffuse the first time with the DT diffusion rule for increased substrate temperature. Atom **a** cannot diffuse again after the first diffusion because its bonds is already two. At the first diffusion in figure 2.3(a), atom **b** can diffuse to one of the nearest neighbor site with the same probabilities. The site that is randomly chosen has one bond. Atom **b** has enough energy to break bond for long diffusion length. It must choose one of the neighbors. If the temperature is increased, some atoms can diffuse the second time following the diffusion rule as shown in figure 2.3(b). Atom **b** diffuses to the site as shown in figure 2.3(b) after the first diffusion. In the second diffusion, atom **b** diffuses to right site. This site keeps the diffusion rule and correct diffusion length. Atom **c** deposits at the site in figure 2.3(a)-(b) following DT diffusion rule. Atom **d** cannot diffuse in the second time because it already has three bonds.

2.2.2 Family with NRT

Figure 2.4(a) shows the Family model with NRT. The growth at elevated temperature is explained in figure 2.4(b) with the second diffusion. Atom **a** can diffuse to the right for increasing diffusion length. Atom **b** can diffuse again following the Family diffusion rule. Atom **c** must diffuse to the left in figure 2.4(b) for longer diffusion length. Obviously, atom **d** must diffuse to the site in figure 2.4(b).

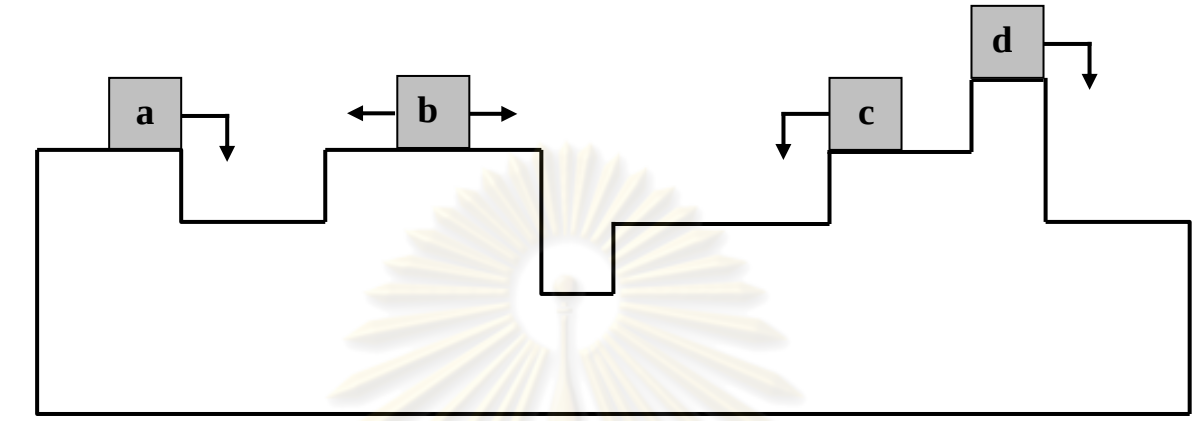


(a)

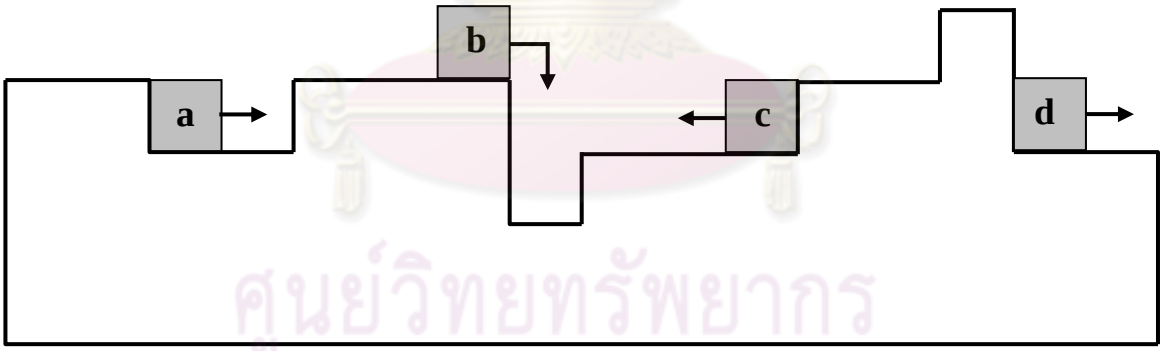


(b)

Figure 2.3 : The thin film growth process in step-flow growth mode using the DT model; (a) The DT atoms diffuse at the first time and (b) The DT atoms diffuse at the second time.



(a)



(b)

Figure 2.4 : The thin film growth process in step-flow growth mode using the Family model; (a) The Family atoms diffuse at the first time and (b) The Family atoms diffuse at the second time.

2.3 Step-Flow Growth Mode

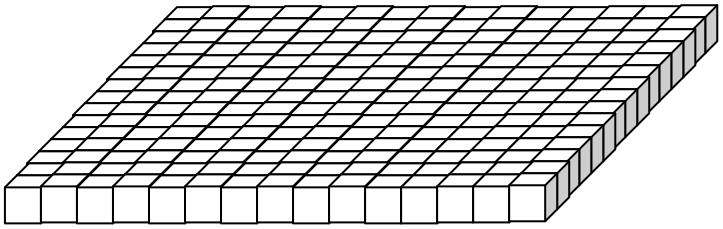
In general, a substrate can be classified to either a flat surface or a vicinal surface. The examples of substrate are shown in figures 2.5(a)-(c).

There is forming of flat surface in figure 2.5(a) that is rather smooth. The surface has high symmetry orientation. When the interaction energy increases, it becomes the surface as figure 2.5(b). The atomic level can be observed on the surface. If there are proper long-ranged interactions, it will be vicinal surface as shown in figure 2.5(c) (Jeong and Williams, 1999).

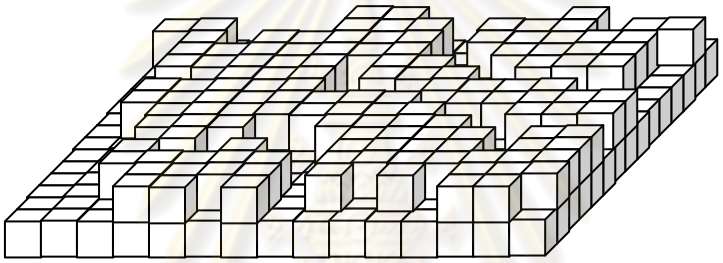
The vicinal surface, or the stepped surface, is an efficient type of substrates in MBE. A stepped surface has a small misorientation. This surface declines from the flat surface by a few degree. Figure 2.6 show (Vilone et al., 2005; Hanbucken et al., 1993) a stepped surface which consists of consecutive atomic flat terraces that line up on top of another terrace. The step edges are divided by boundary lines of different surface height. The height difference is usually one atomic unit (Prestipino and Tosatti, 2001; Stasevich and Einstein, 2007). For the ideal stepped substrate, there is monatomic layer (Giesen, 2001). Figure 2.6 shows the ideal stepped surface. There are not kink sites in the initial stepped surface or the ideal stepped surface. There are parallel regular steps with same distance for each terrace. The ideal stepped surface has one atomic unit of height. The stepped surface makes up from steps and kink formations as shown in figure 2.7. Meandering step can be occurred for the real vicinal substrate as shown in figure 2.5(c) and 2.7.

The vicinal surfaces are used both in theoretical and experimental study. The vicinal surfaces of Si(111) are used experimentally in epitaxial growth (Hanbucken et al., 1993). From the works of Hanbucken, it was found that the (111) terrace reconstruction affects to step formation. The Mo(112) and W(112) surfaces were used to study surface relaxation and stability of surface (Yakovkin, 2005). From the works of Petrova and Yakovkin, the (111) surfaces are the most proper for kinetic adsorption studying of oxygen (Petrova and Yakovkin, 2007).

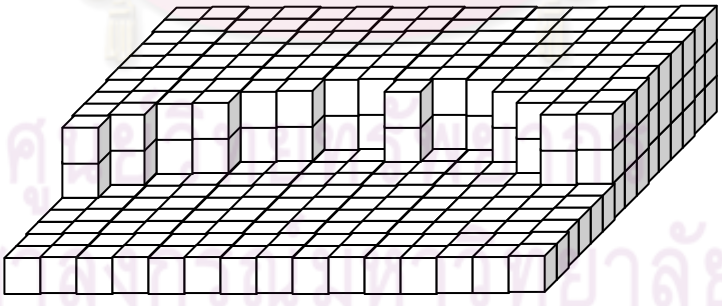
The step-flow growth mode is chosen as a main consideration of this thesis. This step-flow growth mode can be distinguished by considering behavior of the step edges. The constant average terrace width and the advancing step edges are characteristic of this mode. The step edges must evolve in the vicinal direction. Thin films that are grown in the step-flow growth mode have smooth terraces (Hong et al., 2005).



(a)



(b)



(c)

Figure 2.5 : The examples of surface; (a) The flat surface, (b) The surface in high energy and (c) The vicinal or stepped surface.

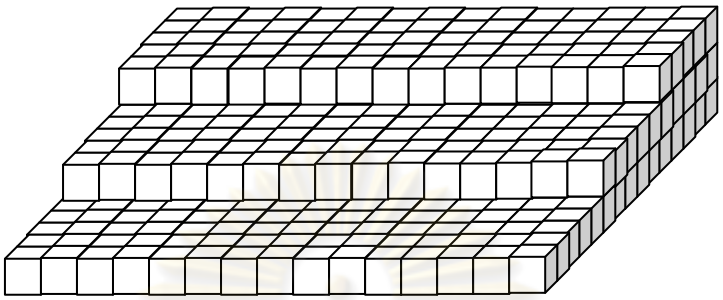


Figure 2.6 : Vicinal surface or stepped surface**.**

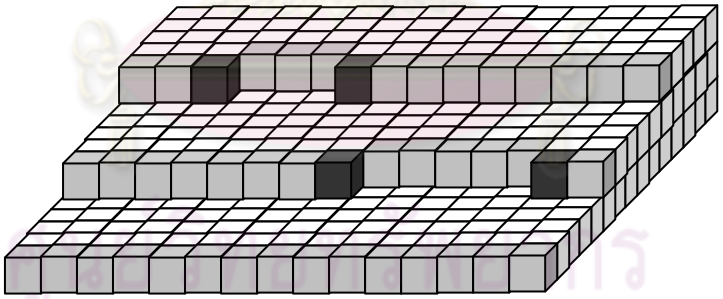


Figure 2.7 : Step and kink formation on the vicinal surface**,** The grey boxes represent step formation and the back boxes represent kink formation**.**

Figure 2.8 shows top view of perfect growth in step-flow growth mode. The perfect growth indicates that all the positions of step edge move equally to vicinal direction. Figure 2.8(a) shows morphology at initial (0 ML). One monolayer is the time where the film is grown exactly likes the initial substrate. The step edges are parallel straight lines. Each shade of step expresses different atomic height. The shade of color shows 4, 3, 2 and 1 atomic unit of height respectively from left side. At 0.5 ML in figure 2.8(b) for perfect step-flow growth, the step edges are still parallel straight. These lines move to a half of each terrace with screw boundary condition. When a film is grown perfectly, top view of the step edges shows parallel straight lines as in figure 2.8(c) with 1 ML. The terrace widths do not change but the step edges advance in step-flow growth process (Zhu, 1998). Figure 2.9 shows imperfect step-flow growth mode. For imperfect step-flow growth in figure 2.9(b)-(c), these new edges are not straight lines. There are meander step edges as shown in 2.9(b)-(c).

To grow thin film in this growth mode, temperature of the substrate and deposition rate must be carefully controlled. If the temperature is high or the deposition rate is small, the atoms have high hopping rate. This means that the atoms have long diffusion length. The atoms can reach step edges. This process is in step-flow growth mode. By contrast, if the temperature is low or the deposition rate is large, the atoms have low hopping rate. The atoms may not have enough energy to break the bonds formed with the substrate since they were deposited. If the diffusion length is short, atoms will form island on the terrace as can be seen in figure 2.10. Figure 2.10 shows snap shot of graphene islands. The island are occurred in the thin film growth process in experimental work (Coraux et al., 2009). This work is under SOS constraint in step-flow growth. This means that bulk vacancy, overhanging and void defect are not allowed here.

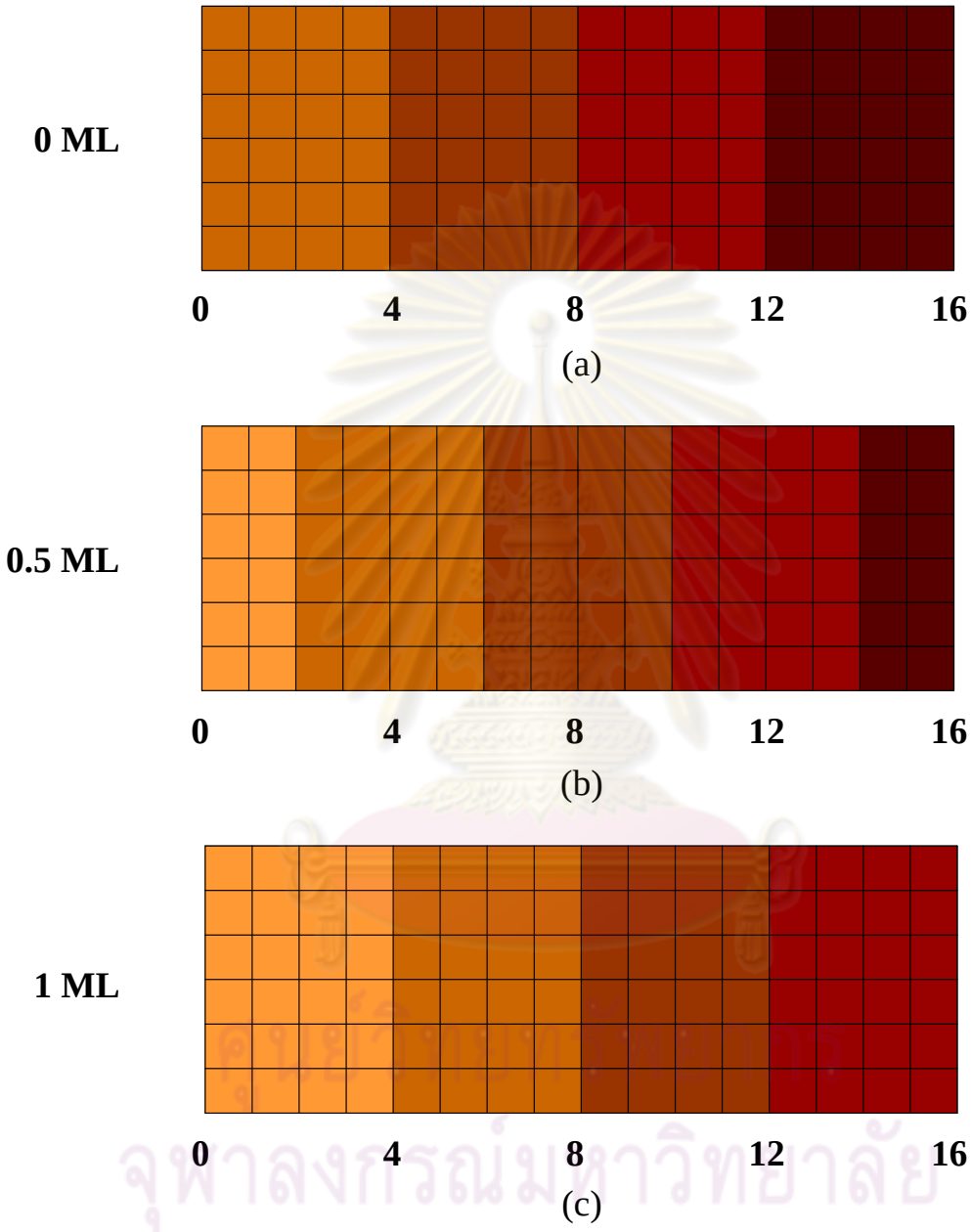


Figure 2.8 : Top view morphology in step-flow growth with perfect growth; (a) The initial stepped surface, (b) Film morphology at 0.5 ML and (c) Film morphology at 1 ML.

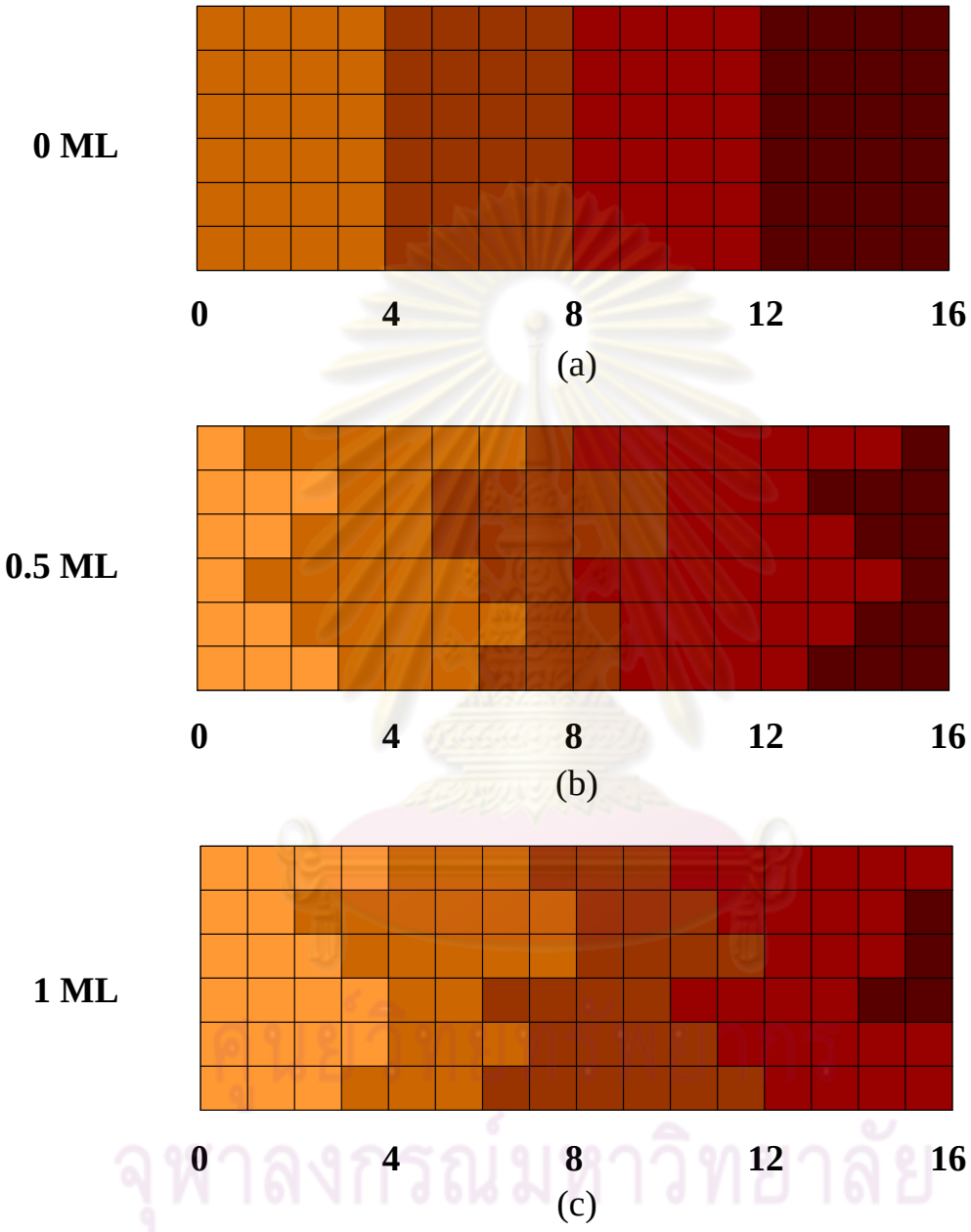


Figure 2.9 : Top view morphology in step-flow growth with imperfect growth; (a) The initial stepped surface, (b) Film morphology at 0.5 ML (d) Film morphology at 1 ML.

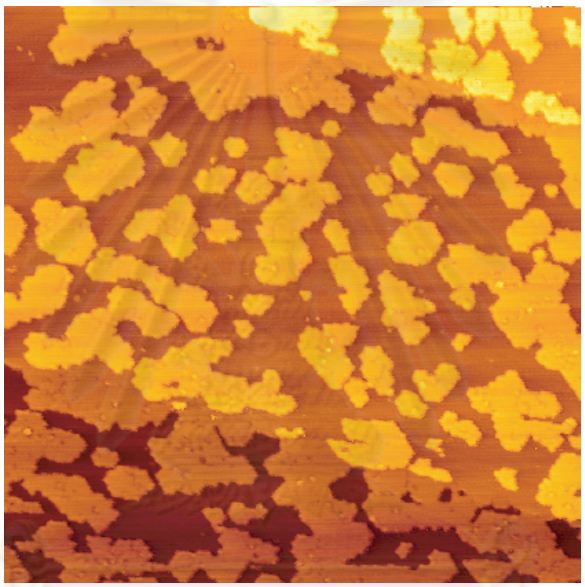


Figure 2.10 : A snap shot of graphene island on the terrace from experimental work. (Coraux et. al., 2009)

2.4 Quantities of Interests

2.4.1 Morphologies

Morphology is structure of film from growth process. The morphology shows evolution of atoms in the growth process. When conditions are adjusted, changed films can be seen clearly by morphology. The morphologies that are investigated will be presented in chapter 4. In the chapter 4, there are both morphologies on flat and stepped surface of all applied technique in this work. For thin film growth in step-flow growth mode, top view morphologies are important and show clearer than three dimensional view. The morphologies show meaningful results that are calculated and interpreted.

2.4.2 Temporal Correlation Function

The step dynamic with monatomic height on vicinal surface is interesting for both theoretical and experimental researches. Spatio-temporal evolution and step fluctuation are determined by kinetic parameter. It is used to observe mass transport (Szalma et al., 2006). The step fluctuation is widely defined by correlation function for equilibrium case (Szalma, Selke and Fischer, 2001). This correlation function measures the distance of fluctuation in space or time (Giesen, 2001). Definition of the correlation function is

$$G(y,t) = \left\langle \left[x(y, t + t_0) - x(y_0, t_0) \right]^2 \right\rangle \tag{2.1}$$

where x is step edge at time t and position y and y is position that parallel with step edge.

Kinetic parameters in the correlation definition can be described by figure 2.11(a). The advance step motion is shown as figure 2.11(b) with 1 ML (Jeong and Williams, 1999). The x coordinate expresses atoms of same step. The x-axis shows direction of atoms that diffuse to the end of step in vicinal direction. The y-axis shows equivalent direction. This y coordinate is used to observe diffusion each step. On the other hand, type of this function explains the fluctuation in direction of parallel to step (Barbier et al., 1996). In usual case, y_0 and t_0 are set at zero (Giesen, 2001).

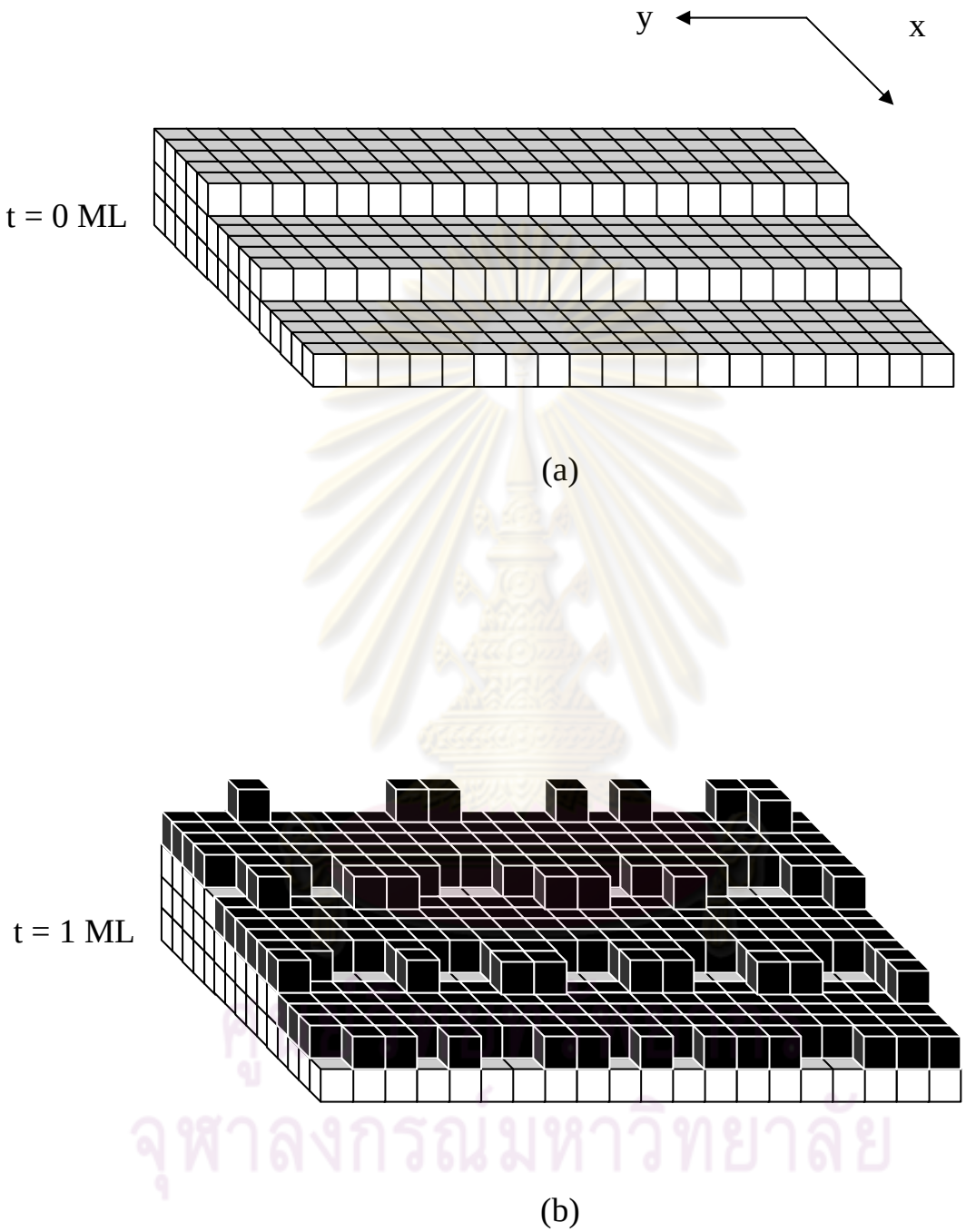


Figure 2.11 : Example of grown thin film ; a thin film grown at 0 ML is shown as figure (a) and a thin film grown at 1 ML is shown as figure (b)

The microscopic fluctuation can be studied with the spatial and the temporal fluctuation. The correlation function is classified to the spatial and the temporal correlation. The spatial correlation function is used to measure step-step interaction and the kink formation energy. The space-dependent correlation function is used to describe a step motion following random walk. While the temporal correlation identifies the activity energy and diffusion processes (Karim et. al., 2004).

Images of time were shown with the temporal correlation function (Giesen-Seibert et al., 1995). Step displacement along the edges with the time dependent correlation function was considered (Goff et al., 1999). The time-dependent correlation function is the ensemble average of the mean square difference of the step edge. A position was fixed at different time from the initial time (Constantin et. al., 2007). The temporal correlation function, a quantity that describes time evolution of the fluctuation of step edge position $x(t)$ (Hong et. al., 2005; Dougherty et. al., 2004), is defined as (Constantin et. al., 2007)

$$G(t) = \left\langle \left[x(t + t_0) - x(t_0) \right]^2 \right\rangle \tag{2.2}$$

A single fixed position on a step edge or y_0 and initial time or t_0 are set at zero. In simple microscopic processes, the temporal correlation function is expected to vary with time as (Jeong and Williams, 1999).

$$G(t) \propto t^{1/n} \tag{2.3}$$

where n is constant that depends on types of diffusion processes. The accurate meaning can use to describe for growth processes of the theory, the experiments and the real models from computer simulation.

Both experimental and theoretical data show that the temporal correlation function varies by power law of time (Szalma et. al., 2001).

2.4.3 Persistence Probability Distribution

In stochastic process, there are probabilities to explain details more than the correlation function (Constantin et al., 2007). These probabilities provide complementary characteristic of dynamic. The probabilities are used as statistical description in the stochastic system (Constantin et al., 2007). The probabilities are the persistence probability distribution and the survival probability distribution. The persistence and survival

probabilities depend on time range. They are used to explain flowing of atoms with the time in experiment, numerical and theory.

These probabilities can be classified to time-dependent and space-dependent of persistence and survival probability, respectively (Constantin et al., 2007).

The persistence probability is an interesting quantity that is studied in this work. This persistence is called the temporal persistence probability; $P(x,t)$. It is the probability that the step edge position does not return to the initial position over all time. Definition of the temporal persistence probability distribution is shown (Constantin et al., 2007)

$$P(x,t) \equiv \text{ Prob } \{x \neq x_{ref}(t_0)\} \tag{2.4}$$

where x is the position of atoms that deposit on the surface. x_{ref} is reference position. The atoms cannot cross the reference position. t_0 is the initial time.

The initial position is a dominant reference for calculating this probability. If the reference is standard for organizing the probability, it can be arranged to negative and positive temporal persistence probability. The negative temporal persistence probability is counted in the negative area. The deposited atom position is lower than the reference. It goes to negative identity direction. In the same way, the negative temporal persistence probability is calculated in the positive area. It is counted when the atom goes far away from the reference. It stays in specific range. Definition of negative and positive temporal persistence probability distribution are

$$P_{-}(x,t) \equiv \text{ Prob } \{x < x_{ref}(t_0)\} \tag{2.5}$$

and,

$$P_{+}(x,t) \equiv \text{ Prob } \{x > x_{ref}(t_0)\} \tag{2.6}$$

The persistence probability tends to decrease with time (Constantin et al., 2007; Constantin et al., 2004; Dougherty et al., 2002). The temporal persistence probability scales with time as

$$P(t) \propto t^{-\theta_p} \tag{2.7}$$

where θ_p is the temporal persistence exponent. It is used to investigate behavior of atoms in step-flow growth mode. The persistence exponent shows probability of step edge to vicinal direction. It is divided to be negative and positive temporal persistence exponent.

This temporal persistence probability varies with time to the power of negative theta. The temporal persistence exponents are calculated both exponents in this research. If theta is high, the persistence probability is small. This case means that probability of atom flowing to the reference is large. If theta is low, the persistence is large. Flowing probability of atom to the reference is small.

Moreover, the persistence exponent can be characterized into the transient persistence exponent and the steady-state persistence exponent (Constantin et al., 2007; Dougherty et al., 2002). These types of persistence are classified by time range as standard. If time range for probability distribution calculation is smaller than saturation time, the persistence probability is transient. If time range is larger than the saturation time, the persistence probability is steady-state (Constantin et al., 2004).

2.4.4 Survival Probability Distribution

The temporal survival probability; $(S(x,t))$ is defined from step deviation of average reference line. The reference lines are shifted with average values of step positions at each monolayer. The survival is probability of step edge position that does not cross to the average reference line in a time interval (Constantin et al., 2007).

$$S(x,t) \equiv \text{ Prob } \{x \neq \bar{x}_{ref}(t)\} \tag{2.8}$$

$\bar{x}_{ref}(t)$ is an average reference position changing with time in a monolayer.

The persistence probability calculation use only initial reference line. The reference of the survival probability follows to time being average value of step (Constantin et al., 2007; Tao et al., 2007).

So the temporal survival probability should be vary with time, The survival probability rather likes the persistence probability. The temporal survival can be classified to negative and positive like the temporal persistence. The negative and positive survival probability distributions are

$$S_{-}(x,t) \equiv \text{ Prob } \{x < \bar{x}_{ref}(t)\} \tag{2.9}$$

$$S_{+}(x,t) \equiv \text{ Prob } \left\{x > \bar{x}_{ref}(t)\right\} \tag{2.10}$$

There is no temporal survival probability distribution exponent definition (Constantin et al., 2004).

Chapter III
Computational Simulation

In this work, stochastic process in thin film growth is studied via computer simulations. The Monte Carlo technique is used in the modeling process. The Monte Carlo technique is a type of simulation used in statistical processes or random processes. It follows probabilistic behavior. Random numbers are used in this technique (Landau and Paez, 1997). Usually, Monte Carlo simulations require large scale computing facility (Giordano, Weir and Fox, 2003).

Here, discrete growth models are simulated using Monte Carlo algorithm created by the C++ language. A cluster consists of 10 computers. Each with Core2 Duo of CPU, 3.0 GHz of speed and 4 GB of RAM is used for this large scale simulations.

In this chapter, computational simulation conditions are presented. Techniques and conditions to grow thin film in step-flow growth mode and calculating methods of temporal correlation function, persistence and survival probabilities are included in this chapter.

3.1 Computational Simulation Conditions

The ideal stepped substrates of size 100×100 lattice sites are used in this research. There are five steps in the substrate. The width of each step is twenty atomic units. The total growth time for every simulation in this research is fixed at 10^6 MLs.

Considering figure 3.1, periodic boundary condition is employed in the y-axis which is the direction without the tilt. Screw boundary condition is used in the x-axis which is the tilt direction. Screw boundary condition, also known as helical boundary condition (Karim et. al., 2004), is similar to periodic boundary condition except that the height is adjusted to be consistent with the tilt. From figure 3.1, when an atom crosses boundary A from left to right, it will be at position B. In the same manner, if an atom moves across the boundary B in backward direction, it will be placed at position A.

From NRT, $l_d \propto \exp\left(\frac{-E_x}{k_B T}\right)$ (Barabasi and Stanley, 1995), the diffusion length varies with the substrate temperature. When the substrate temperature is high, atom has more energy. The atom can break away from its site and diffuses many times. This leads to a large diffusion

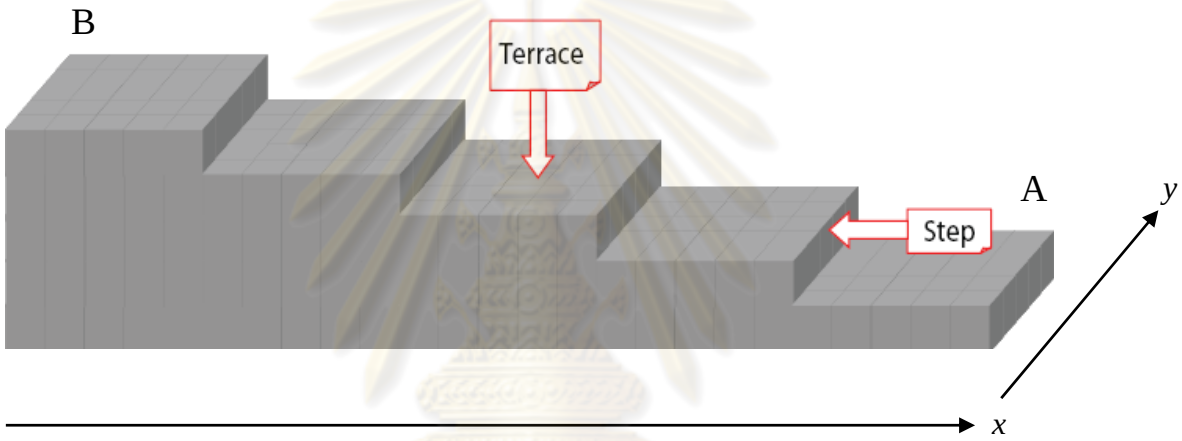


Figure 3.1 : Vicinal surface or stepped surface**.**

length. Adjusting number of diffusions is the technique used to reflect the change of the substrate temperature. Number of diffusions used here are 1, 100, 400 and 900 hops.

3.2 Calculation Methods

3.2.1 Temporal Correlation Function

The step edge positions $x(t)$ at the initial time $t_0 = 0$ are fixed at the positions $x(t=0) = 0$, 20, 40, 60, 80 and 100 sites. The value of y position used in this work is fixed at zero. Figure 3.2 shows an example of calculation at fixed y in side view. Figure 3.2(a) shows the stepped surface at 0 ML. This stepped surface consists of four step and the width of each step is three sites. The initial edges of the first and the second step are shown in figure 3.2(a) at $x_1(t=0) = 3$ and $x_2(t=0) = 6$ respectively. From figure 3.2(b), the grey boxes show atoms of the first and the second step that are grown after 1 ML. The new step edge positions are $x_1(t=1) = 4$ and $x_2(t=1) = 8$ respectively. These positions are used in the calculation of temporal correlation function. These functions are collected after each monolayer deposition at all steps. Average value of the temporal correlation function from all steps and many substrates is calculated.

3.2.2 Persistence and Survival Probability Distribution

This section presents concepts of temporal persistence and survival probabilities calculating method. Initial substrate consists of five steps. Each step has terrace with the width of 20 atomic units as shown in figure 3.3. The shades of color show height levels from high (light color) to low (dark color). The straight lines at 20, 40, 60, 80 and 100 in figure 3.3 show initial step edges. These lines are taken as references. Negative and positive temporal persistence are considered from atoms that diffuse within a specific range of 10 atomic units. If the diffusion length of the atom is shorter than the reference range (approximately 10 atomic units), the persistence will be negative. On the contrary, if the atom diffuses further than the reference range (about 10 atomic units), the persistence will be positive.

The reference lines for the calculation of the survival probability distribution differ from the initial references. The references for the survival probability distribution are averaged values following the time

evolution of the film growth. They advance with time and diffusion length.

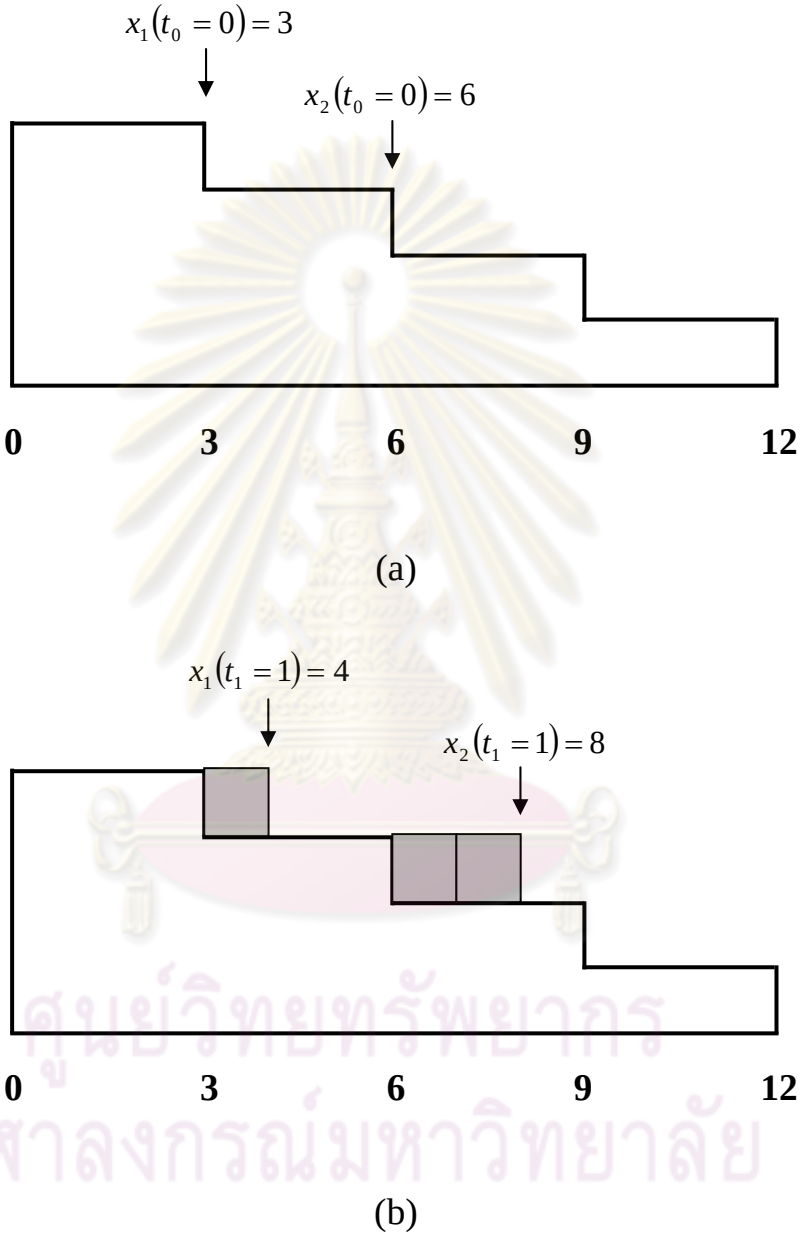


Figure 3.2 : Example for the calculation of the temporal correlation function**.**

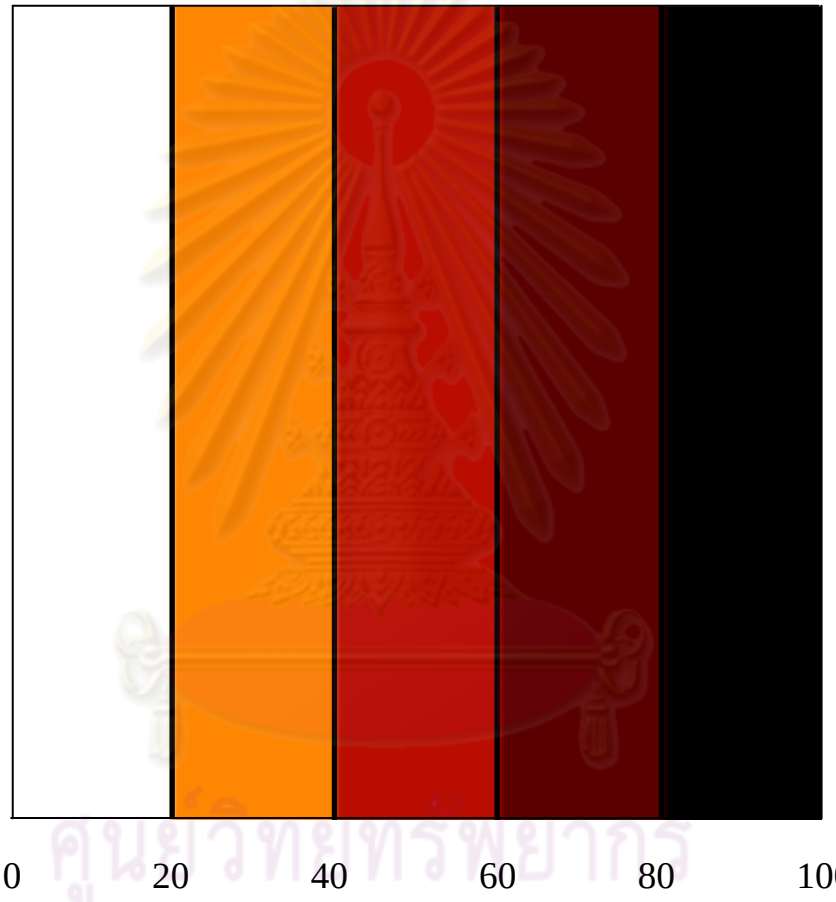


Figure 3.3 : Top view of vicinal surface or stepped surface at 0 ML.

Chapter IV
Results and Discussion

In this chapter, we show our simulated films grown by the DT model and the Family model on flat and stepped surface. Morphologies, temporal correlation function, persistence probability distribution and survival probability distribution are presented respectively.

4.1 Morphologies

In this part, the results are shown in array format. The row of the array represents number of diffusions while the column of the array represents paused growth time. The height of the film surface can be seen from different shade of color, the lighter the color the higher the film surface.

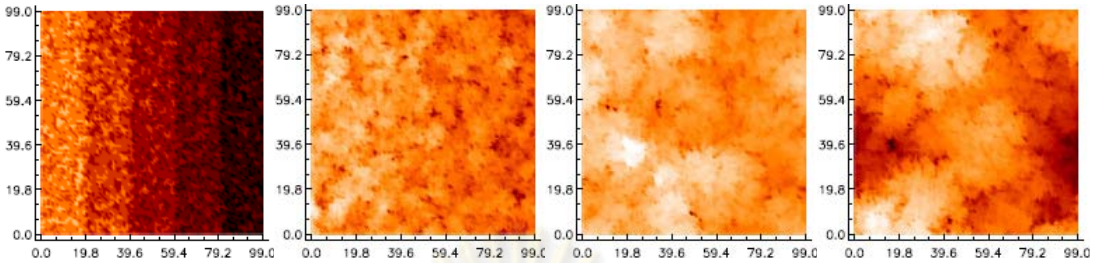
Figure 4.1(a), (b), (c) and (d) show three dimensional plot of morphologies of the DT films grown on stepped surface with 1, 100, 400 and 900 hops respectively. For the basic growth, diffusion length is one, the film loses their step pattern very quickly. This is because the growth does not follow step-flow growth mode. From the figure 4.1(a), we can see that the film surface of 1 hop at $t = 10^2$ MLs has almost the same height. It looks as if the film is grown on flat substrate. For 100 hops shown in figure 4.1(b), the step-flow growth mode begins. The steps of thin film of 100 hops can be seen but do not clear. For 400 hops and 900 hops in figure 4.1(c) and (d), at $t = 10^4$ MLs and $t = 10^6$ MLs, we can see step and flowing of atoms from the growth. For larger mobility ($n \geq 100$ hops), atom can diffuse to the end of each step that follows step-flow growth mode.

Figure 4.2 shows top view morphologies of the DT films grown on stepped substrate. For the observed film grown with 1 hop at 1 ML in figure 4.2(a), the steps are still formed. At 10^2 ML in Figure 4.2(a) does not show clear steps but the film still inclines. At 10^4 and 10^6 MLs, the films incline but we cannot see steps in figure 4.2(a). From this figure, we can see the similar films as they are grown on flat substrate. For figure 4.2(b), deposited atom can diffuse 100 times before another atom is

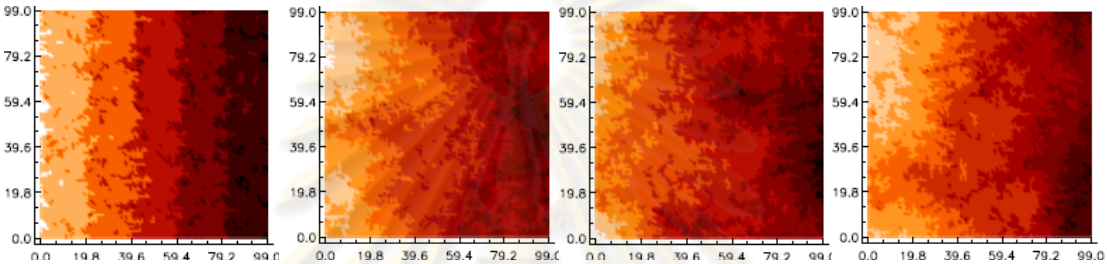
Figure 4.1 : Three dimensional morphologies of the DT films grown on stepped surface.

released to the film. Thin film at 1 ML shows atom flow from step to step according to color of steps. Although the straights of steps cannot be seen clearly, the film can maintain the incline up to 10^6 MLs. The step-flow growth mode begins from 100 hops in figure 4.2(b). The next morphologies are the case of 400 hops. Here we can see the step clearly throughout the growth. Step-flow growth mode is activated because of temperature added to the growth process. Newly arrival atom can diffuse to the end of the step. In this case, the film can keep the shape of substrate better than those grown at low temperature. For 900 hops in figure 4.2(d), we can see obviously both steps and flowing of steps to vicinal direction. We set the temperature to be high in this growth process. Therefore, atoms can diffuse far enough to reach the end of steps. When time increases, straight lines of steps cannot be seen explicitly. For 400 hops and more, the morphology shows step explicitly for all ranges of growth time. In vertical view of figure 4.2, each column shows thin films at same time with different mobilities. Steps of thin films can be seen obviously for all mobilities at 1 ML. For 1 hop of 1 ML steps of thin film are the clearest because atoms can diffuse only one time. The atoms do not flow fast and the terraces of each step are the roughest. More mobilities or more roughness at this time, atoms can diffuse many times. There are more probabilities to flow to the step edges. The views of morphologies at 10^2 MLs, for 1 hop, steps are absence and the film looks very rough. The atoms fill up and form island on the terraces of stepped surface. The film rather likes the morphology on flat surface at the same time. The step-flow growth happens for large mobilities from 100 to 900 hops. At 10^4 MLs, the film of 1 hop looks just like thin film on flat substrate at the time in figure 4.3(a). The new step edges of large mobilities at this time are longer than the step edges at previous time. The step-flow growth can occur in large mobilities. Steps flow faster than those at the previous time. The morphology of thin film of 1 hop is the same as thin film that is grown on the flat surface at 10^6 MLs shown in figure 4.2(a). The films of large mobilities are grown in step-flow growth mode. There are longer new step edges than usual.

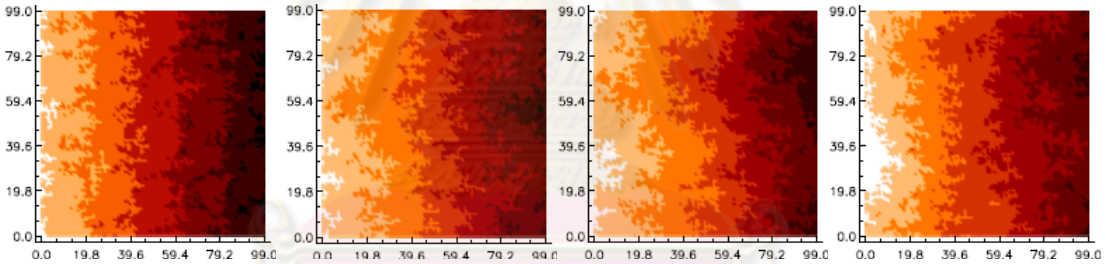
Considering morphologies of the film in figure 4.3(a)-(d) from small to large mobility in each column while the growth time is fixed. The films that has large atomic mobilities are smoother than the film that has at small atomic mobilities. Atoms can diffuse and fill the holes on the films when it has large mobilities. They can find more proper position than small mobilities. Subsequent growth processes in more time show atom diffusion following diffusion rule of the model. From figure 4.3, step flow growth mode does not happen certainly when the thin films are



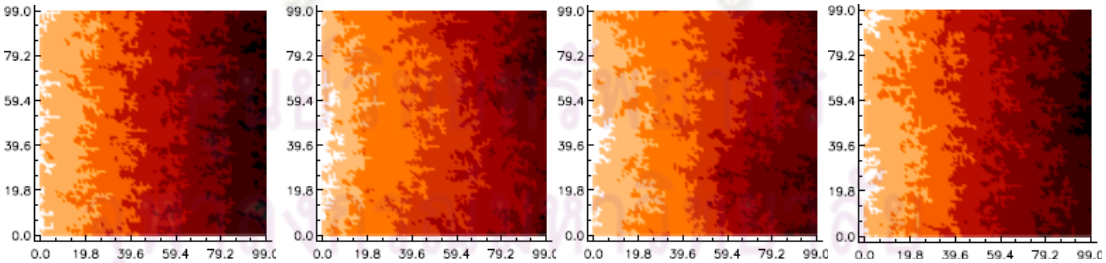
(a) 1 hop



(b) 100 hops



(c) 400 hops



(d) 900 hops

1 ML 10^2 MLs 10^4 MLs 10^6 MLs

Growth time

Figure 4.2 : Top view of the DT films grown on stepped surface.

Figure 4.3 shows 3D Morphology from the DT films grown on flat surface.

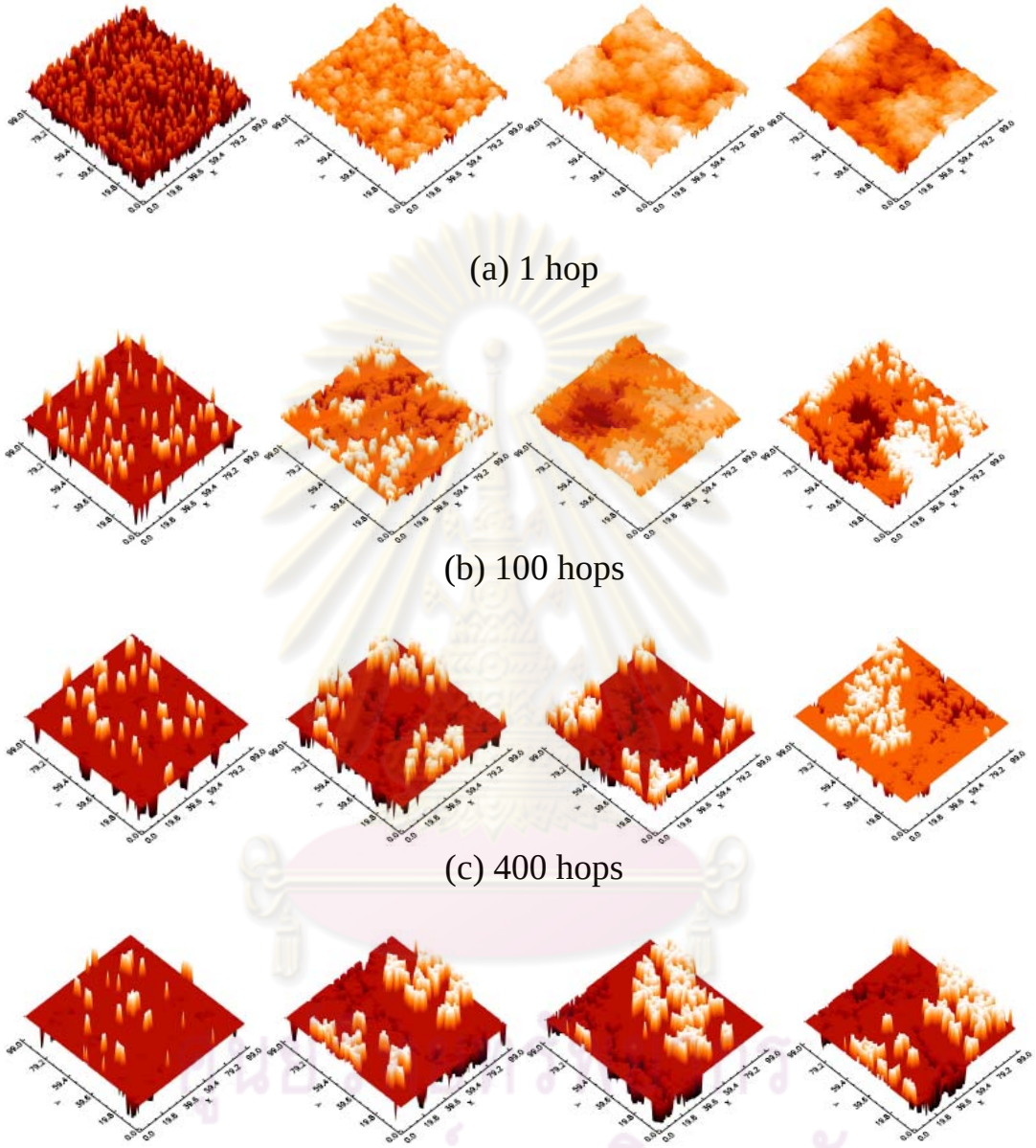


Figure 4.3 : Three dimensional morphologies of the DT films grown on flat surface.

grown on the flat surfaces for all mobilities.

Figure 4.4(a)-(d) shows top view morphology of the DT films of 1, 100, 400 and 900 hops grown on flat surface. Here films do not grow in step-flow growth mode. The thin films with large mobilities are smoother than small mobilities. This can be seen from figure 4.4(d) that the thin film at 900 hops has smoothest surface.

The three dimension morphologies grown on stepped surface with the Family model from 1 to 900 hops are shown in figure 4.5(a)-(d). Both three dimension view (figure 4.5(a)-(d)) and top view (figure 4.6(a)-(d)) show flowing of atoms to the ends of steps. Considering the morphologies each mobility, starting at 1 ML in figure 4.5, step edge can be seen clearly. The morphologies of thin film for 1 hop from 10^2 to 10^6 MLs are very rough as shown in figure 4.5(a). The film inclines but the step edges cannot be seen at large time for 1 hop. Obviously, step-flow growth mode cannot be seen for 1 hop. The step-flow growth mode cannot happened when atoms rarely move to the edges. To observe thin film at 100 hops in figure 4.5(b), step-flow growth starts at this mobility. Atoms spread over and new edges of steps almost not identify definitely positions from the start of the growth to the growth at 10^6 MLs. The morphology at 100 hops shows that atom can distribute over the next terraces. Typically, the maximum range that atoms can diffuse is just a half of step width. This result makes the step width increases wider than the width of the film with larger number of hops. For 400 hops in figure 4.5(c) atoms can diffuse with maximum probability to the edges of steps. The maximum diffusion length equals twenty sites. These morphologies with this mobility show clearer step edges than 100 hops. When considering morphologies of 900 hops in figure 4.5(d), the maximum probability of diffusing atoms is larger than original step size. At this point, new steps can be seen clearly at all time. These results are similar as the growth with 400 hops. Subsequently, morphologies at the same time for all mobilities are studied with column in figure 4.5. The first time, step can be seen for all mobilities. The morphologies at the next time show atom arrangement better than previous time. This time is long to notice step edges in growth process range. One different thing that is observed from all mobilities is the indistinction of step edges for 1 hop. At the first time, the different thing is the roughest surface on each terrace at 1 hop. The terraces of each step for 1 hop are not smooth. For more monolayer, at 1 hop flowing of step can be seen but step edges cannot be seen certainly. At longer time, step edges of 400 and 900 hops are smoother than 1 hop and 100 hops. To observe at final time, the

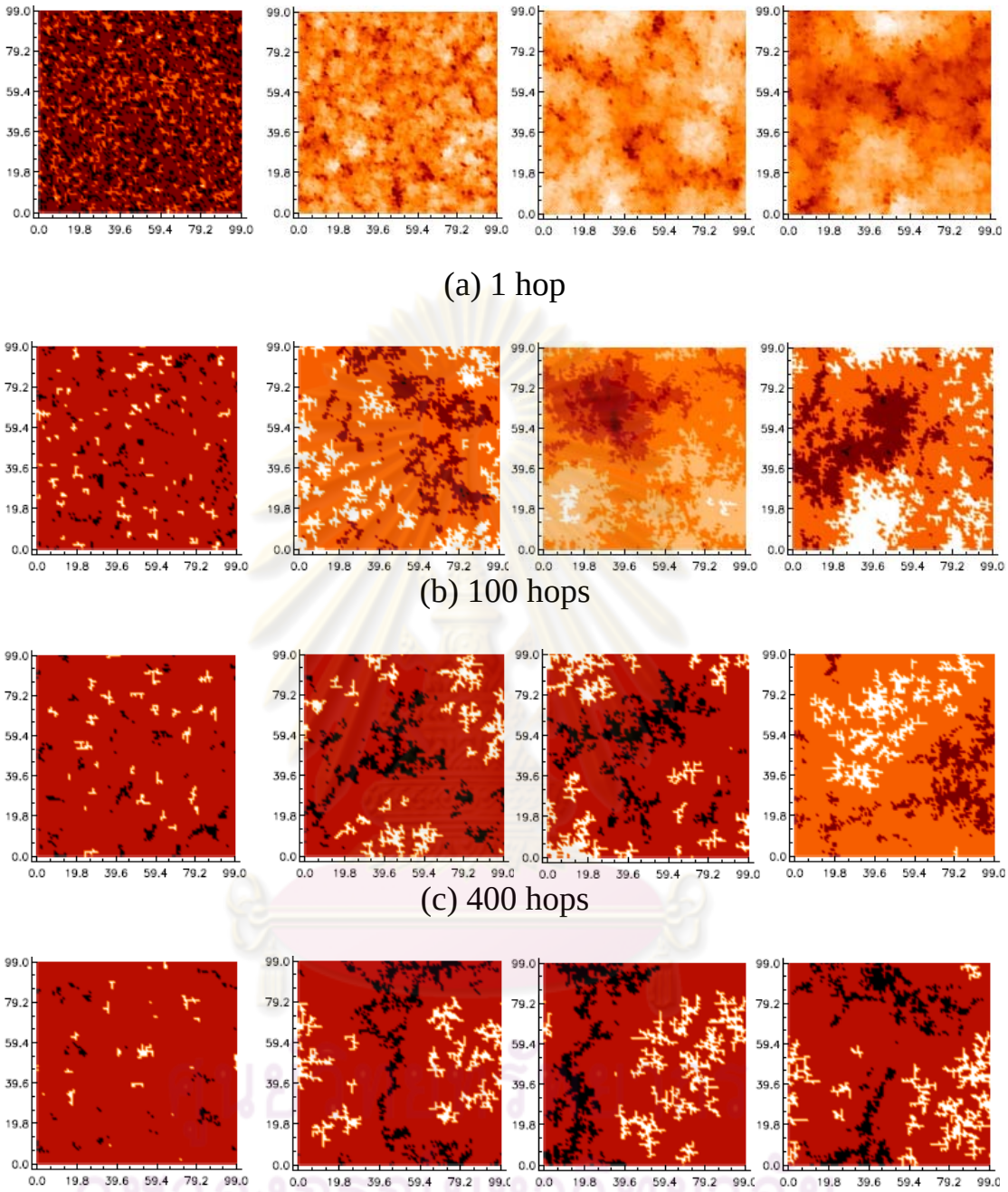


Figure 4.4 : Top view of the DT films grown on flat surface.

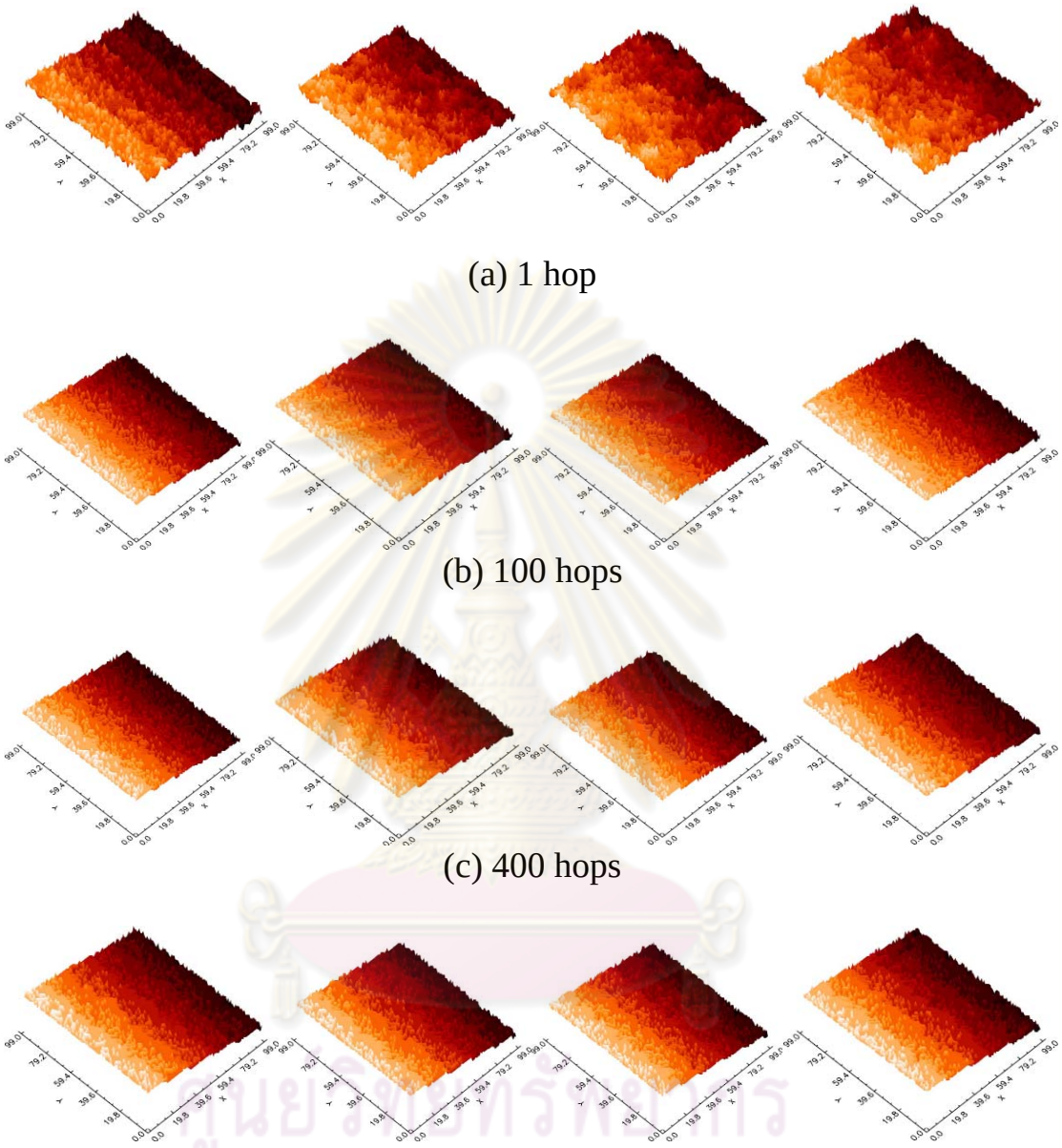
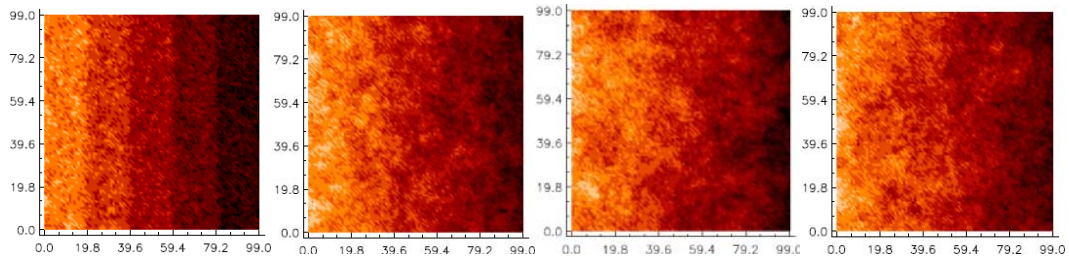
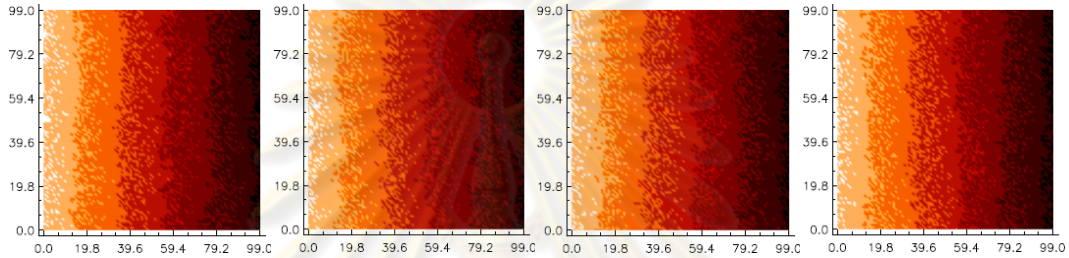


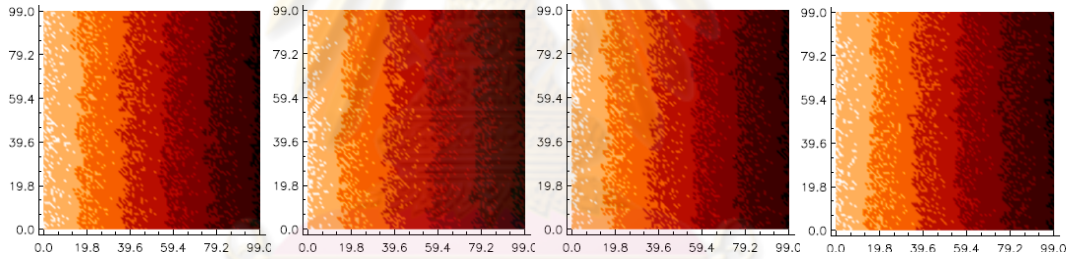
Figure 4.5 : Three dimensional morphologies of the Family films grown on stepped surface.



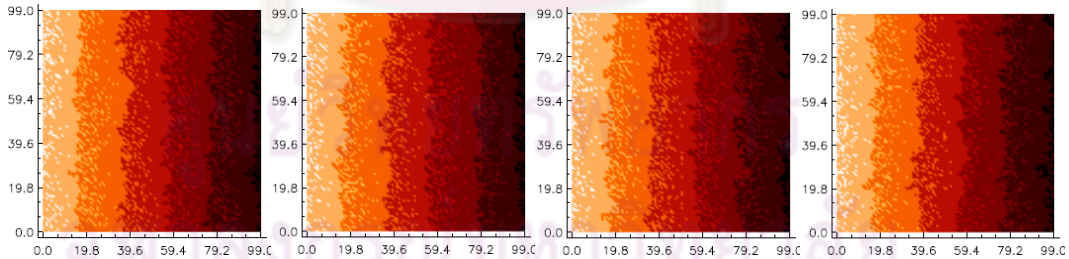
(a) 1 hop



(b) 100 hops



(c) 400 hops



(d) 900 hops

1 ML 10^2 MLs 10^4 MLs 10^6 MLs

Growth time

Figure 4.6 : Top view of the Family films grown on stepped surface.

morphologies are rather stable. Obviously, step-flow growth mode does not appear in the growth of 1 hop. The mode can be seen for the growth of at least 100 hops. One hundred hops or a half of step diffusion make non-uniform step edges in the growth process. Four hundred hops make the proper step-flow growth mode. Step-flow growth mode can happen certainly at 900 hops. Figure 4.5 and figure 4.6 correspond to above discussion.

4.2 Temporal Correlation Function

Relations of temporal correlation and time of the DT model and the Family model are shown in figure 4.7 and figure 4.8, respectively. The simple trend of the temporal correlation function should be linear with time. Figure 4.7 and 4.8 show relation of the temporal correlation for long time. In this time, the curve of temporal correlation function saturates. The films are reset each monolayer in step-flow growth mode. The functions in short time are considered. Figure 4.9-4.10 show log-log plot of the temporal correlation function and short time of the DT and the Family model respectively. In figure 4.9, the lines show that the functions grow with time. To analyze the exact slope of the curves we show the exponents $(1/n)$ of the temporal correlation function in table 4.1. These results show increasing energy of atoms. These results correspond to the growth process in step-flow growth mode. Lower activation energy occurs at low temperature. The activation energy is higher in high temperature.

Considering table 1 for the DT model, the exponents are transition from low to high temperature. The exponents for the DT model are characterized in four cases which are the exponents of 1 hop, 100 hops, 400 hops and 900 hops. These types of atomic processes can be seen from the morphologies. From the morphologies in figure 4.1(a) and 4.2(a), atoms are in low temperature so they have low energy. Atoms for 1 hop cannot diffuse far from their incorporated sites. The sites are mostly at step edges because of the diffusion rule of the DT model. To be on top of atoms are shown by morphologies of 1 hop in figure 4.1(a) and 4.2(a). The atoms start to flow in 100 hops case in expressing with figure 4.1(b) and 4.2(b). Atoms in this case have enough energy to flow a little. They can diffuse on terraces. They incorporate at new step edges. However atoms cannot cross terraces, diffusion length is less than terrace width.
For higher energy case where an atom can diffuse approximately 400

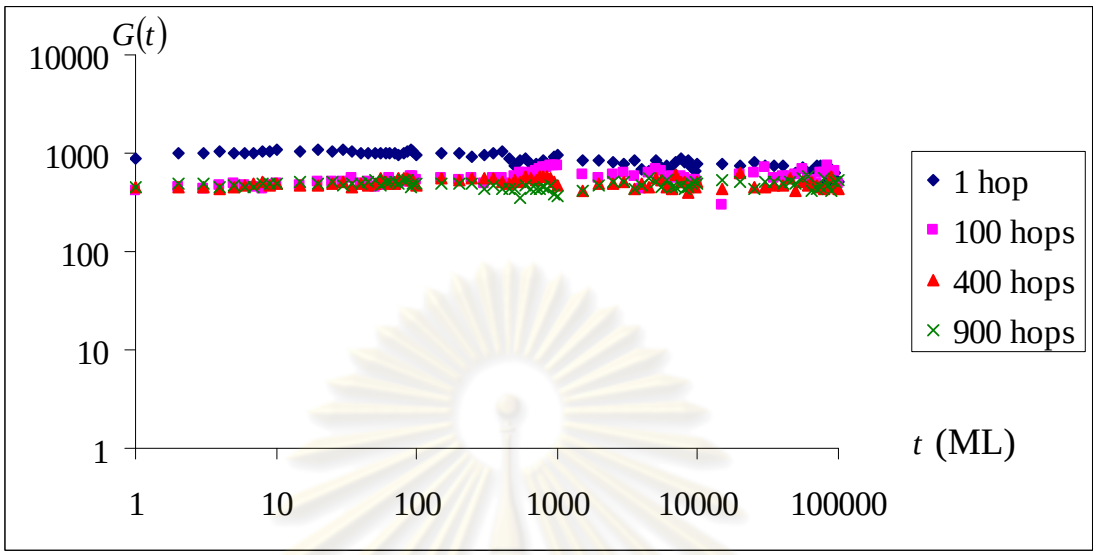


Figure 4.7 : Log-log plots of the temporal correlation function of the DT films grown on stepped surfaces**.**

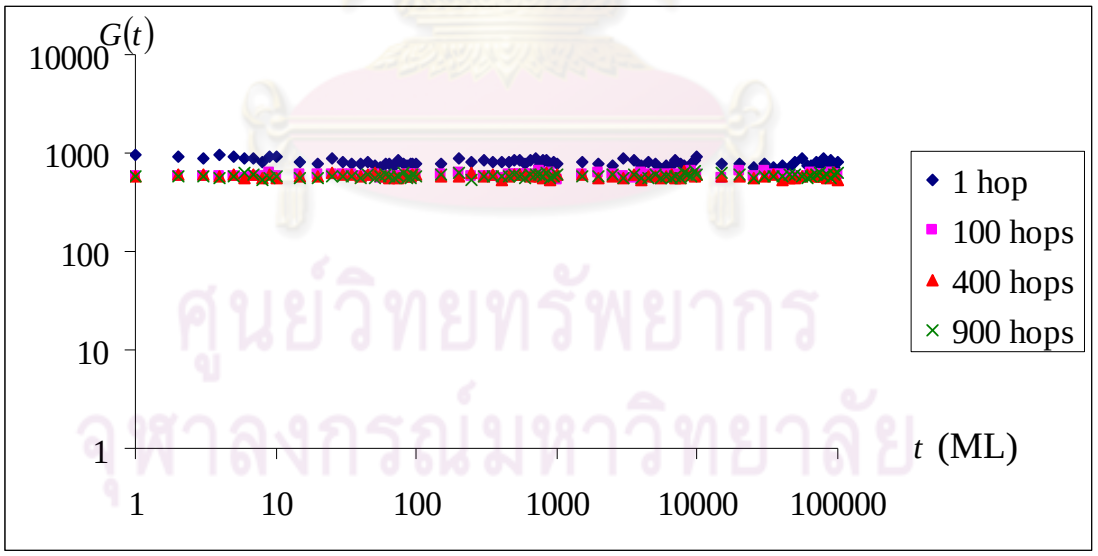


Figure 4.8 : Log-log plots of the temporal correlation function of the Family films grown on stepped surfaces**.**

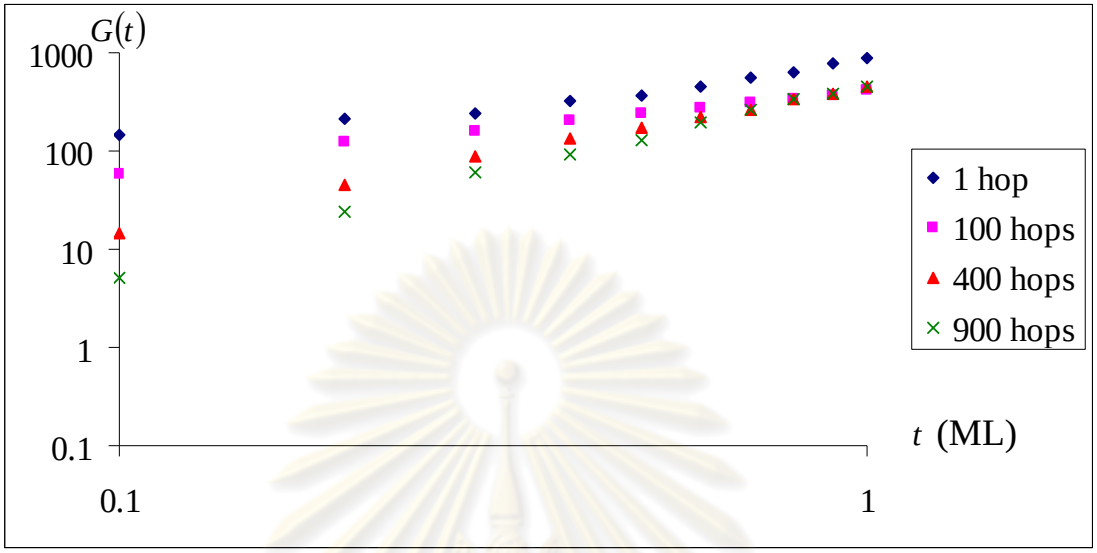


Figure 4.9 : Log-log plots of the initial time temporal correlation function of the DT films grown on stepped surfaces**.**

Figure 4.10 : Log-log plots of the initial time temporal correlation function of the Family films grown on stepped surfaces.

Table 4.1 : Exponents of the temporal correlation function of the DT films and the Family films.

Number of hops	Exponent of temporal correlation function $(1/n)$	
	DT model	Family model
1	0.79	0.88
100	0.82	0.62
400	1.46	0.85
900	1.92	0.91

times, atoms have high energy. They can diffuse to another step edge because diffusion length in this case is about step width. There is probability for diffusion to another step. The largest diffusion length in this work is 30 atomic units. In this case, atoms can diffuse to another terrace. These meaning of atomic processes support applying temperature in the growth processes. The step-flow growth mode can occurs in high temperature. To consider exponents of the temporal correlation function of the Family model, figure 4.10 shows trend of the temporal correlation in the short time. There is transition atomic process when the films are grown in previous time range for all hops. Diffusion rules of the Family model bring these results. The exponents of the temporal correlation of the Family model are shown in table 4.1. These results are different from the DT model. From figure 4.5 and 4.6, the morphologies at the lowest temperature are quit different from the morphologies at high temperature. Atomic process of 1 hop in figure 4.5(a) and 4.6(a) is the diffusion process. In this process, atoms diffuse along step edges. According to the Family diffusion rule atoms must choose the lowest site ; therefore, it looks as if there is no flow. Because of this, 1 hop of the Family model is different from 1 hop of the DT model. Consider the case of 100 hops, diffusion length is a half of step width. Atoms can flow and incorporate to be a part of the film in a far distant from its deposition site. Since the diffusion of atom by this model does not depend on the number of bonds, all surface atoms can diffuse on the terrace. The result from this process can be seen in figure 4.5(b) and 4.6(b). For 400 hops case, atoms still find the lowest sites. They have twenty atomic of diffusion length. Atoms can incorporate twenty diffusion lengths far from old site of deposition. The sites that they incorporate are not necessary at step edges. Figure 4.5(c) and 4.6(c) show this atomic process. Atoms of 900 hops show similar behavior as those of 100 and 400 hops. Atoms can diffuse far to thirty atomic units and they can incorporate at any sites lower than its original position. Atomic processes for the Family model are categorized into two cases, the case of high temperature and that of low temperature. Behavior of atoms is rather same in high temperature. The atoms diffuse on the terraces. For low temperature, atoms are on top of others at the step edges. These results correspond to trend of the temporal correlation function as shown in figure 4.10. There is rather same trend of 100-900 hops series.

4.3 Persistence and Survival Probability Distribution

The persistence probabilities are calculated in both negative and positive cases for all hops. Figure 4.11 shows the log-log plots of negative and positive persistence probability versus time for all hops of the DT model. Both positive and negative persistence probabilities for every case decrease with time. Atoms have small probabilities that cannot return to the references when time passes. From figure 4.11, slopes of negative and positive persistence distribution are not far so the details will be discussed when table 4.2 is shown.

Survival probability distributions are shown in figure 4.12. The figure 4.12 is plots of negative and positive survival probability versus time in the growth process. The survival probabilities of all mobilities decrease with growth time both negative and positive. They are less different both negative and positive.

Figure 4.13 and 4.14 shows comparing of persistence and survival probability distributions of the DT model. Decrease rates of $p(t)$ and $s(t)$ are approximately the same both negative and positive. The references or step edges of persistence calculation are constant. The DT atoms flow not far so the references for calculating of $p(t)$ are not far different from average references of $s(t)$.

Table 4.2 shows values of negative and positive persistence exponents when the diffusion length is changed, from short to long diffusion length. The results are consistent with the morphologies shown in figure 4.1 or 4.2. Atoms can flow to step edges. Atoms diffuse to step edges following step-flow growth mode. The step-flow growth occurs in 100-900 hops. Both negative and positive persistence exponents decline with more mobilities in step-flow growth as shown in table 4.2. The probabilities for finding new sites are added in higher temperature. These results correspond to step-flow growth.

Figure 4.15 shows trend of negative and positive persistence and time of the Family model. Both negative and positive persistence decreases with growth time like persistence probabilities in the DT model. Atoms have more probabilities to return the references with time. Negative and positive persistence are not far each hop.

The survivals of all mobilities decrease almost both positive and negative as shown in figure 4.16. This means that there are less probabilities of step edge position being beyond the average reference with growth time for all mobilities.

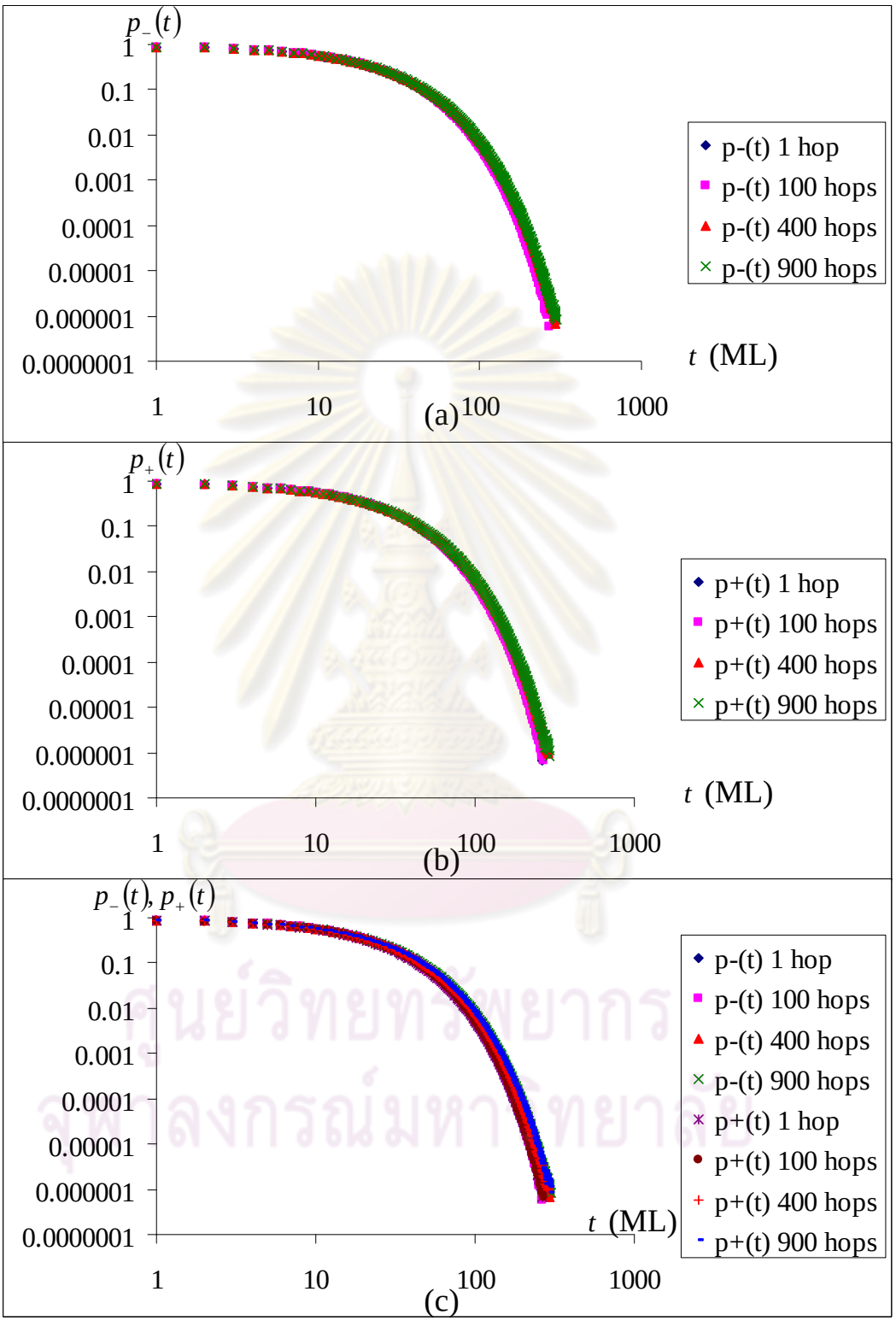


Figure 4.11 : Log-log plots of negative vs. positive persistence distribution of the DT films on stepped surfaces**.**

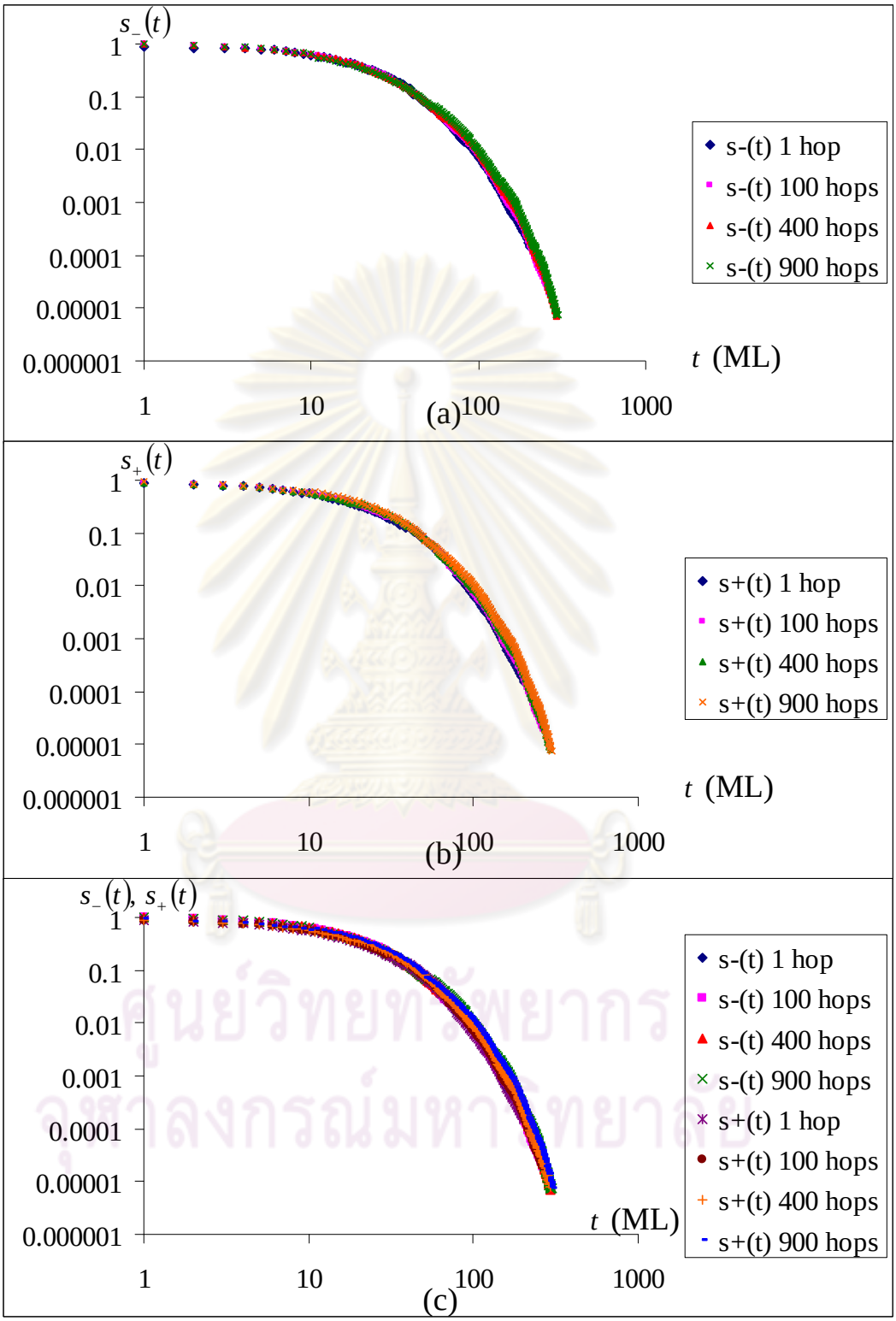


Figure 4.12 : Log-log plot of negative vs. positive survival distribution of the DT films on stepped surfaces**.**

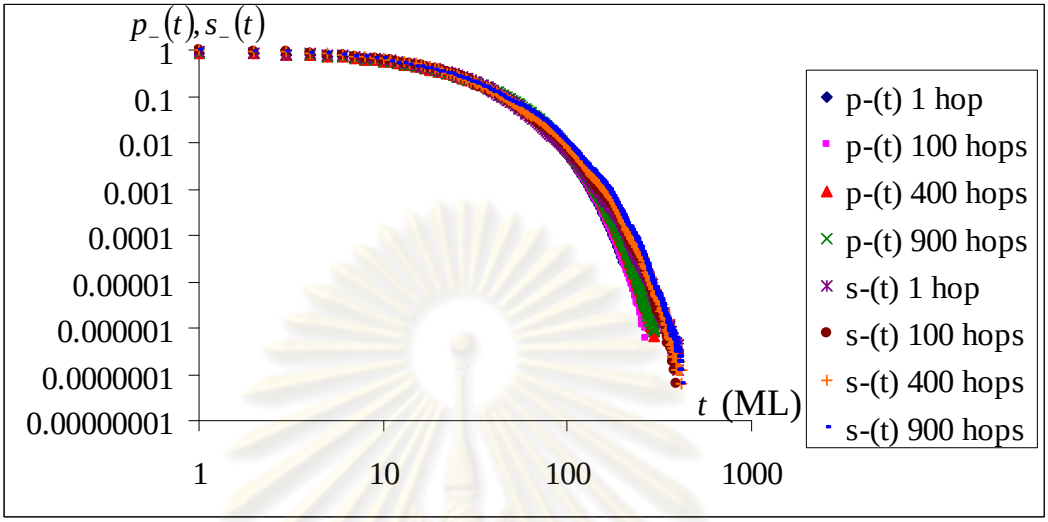


Figure 4.13 : Log-log plots of negative survival distribution vs. persistence distribution of the DT films on stepped surfaces**.**

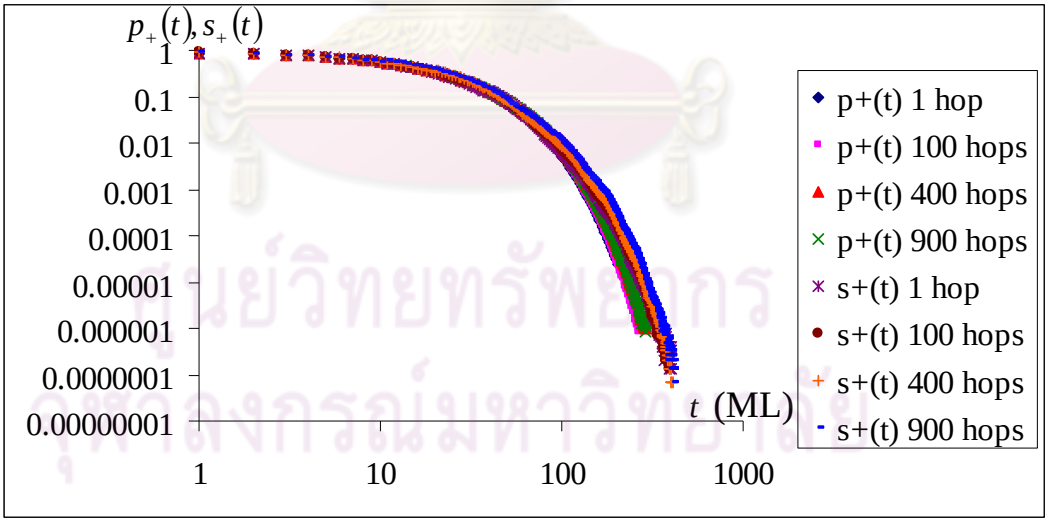


Figure 4.14 : Log-log plots of positive survival distribution vs. persistence distribution of the DT films on stepped surfaces**.**

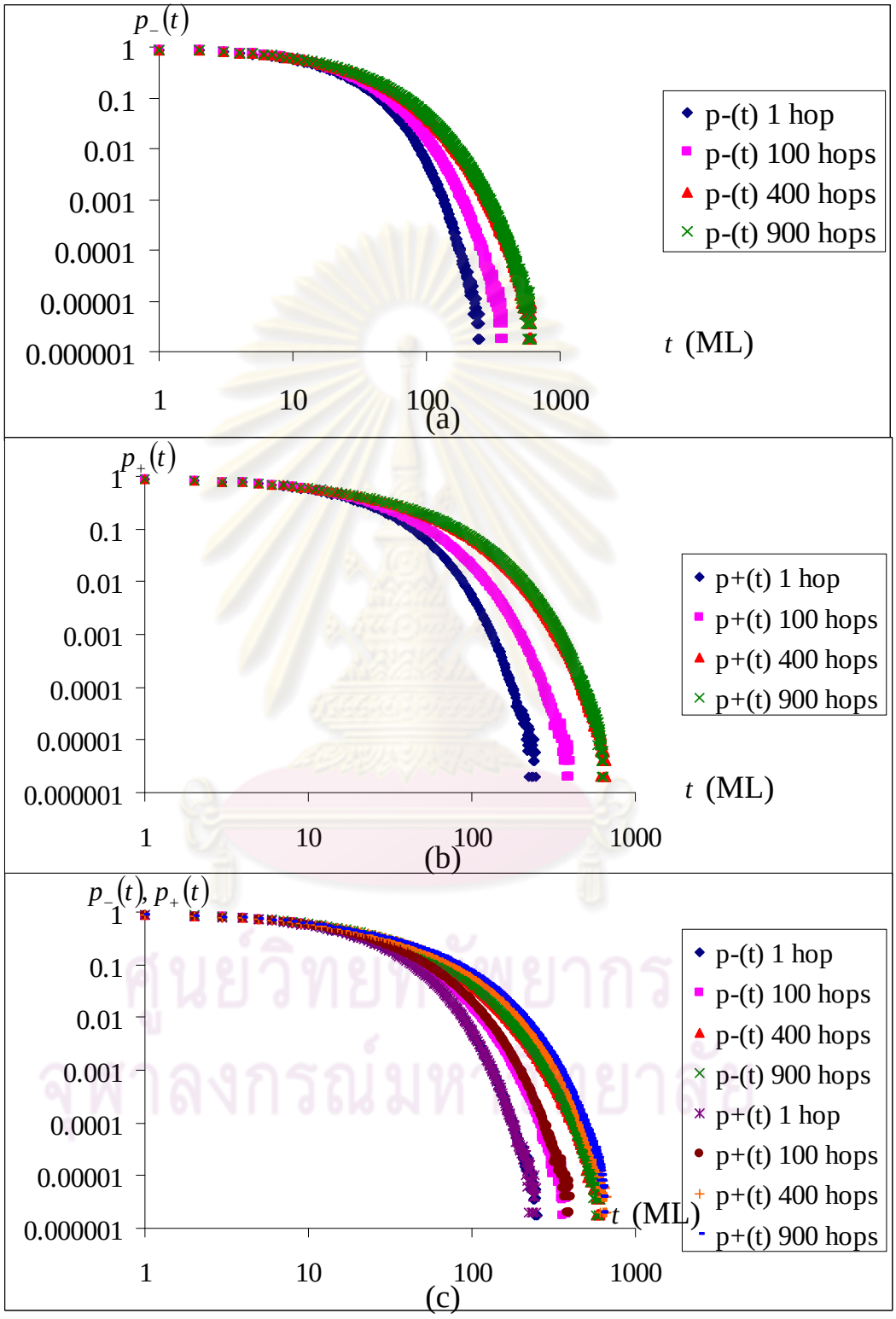


Figure 4.15 : Log-log plots of negative vs. positive persistence distribution of the Family films on stepped surfaces**.**

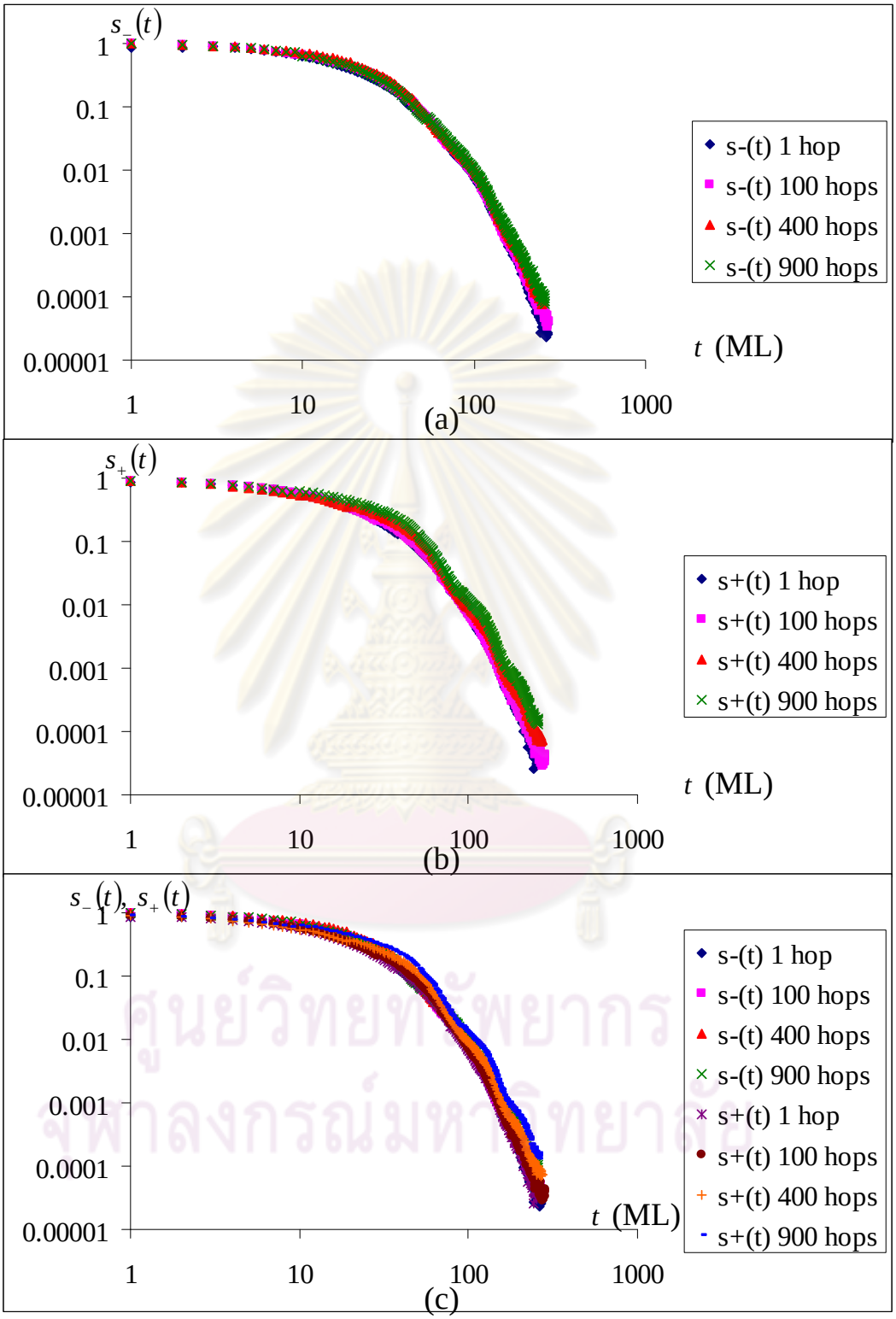


Figure 4.16 : Log-log plots of negative vs. positive survival distribution of the Family films on stepped surfaces**.**

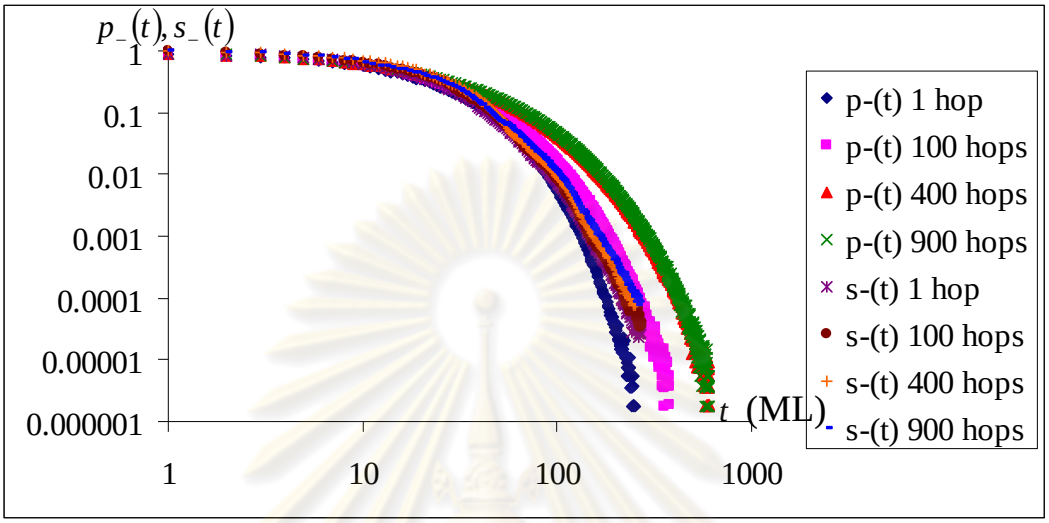


Figure 4.17 : Log-log plots of negative survival distribution vs. persistence distribution of the Family films on stepped surfaces**.**

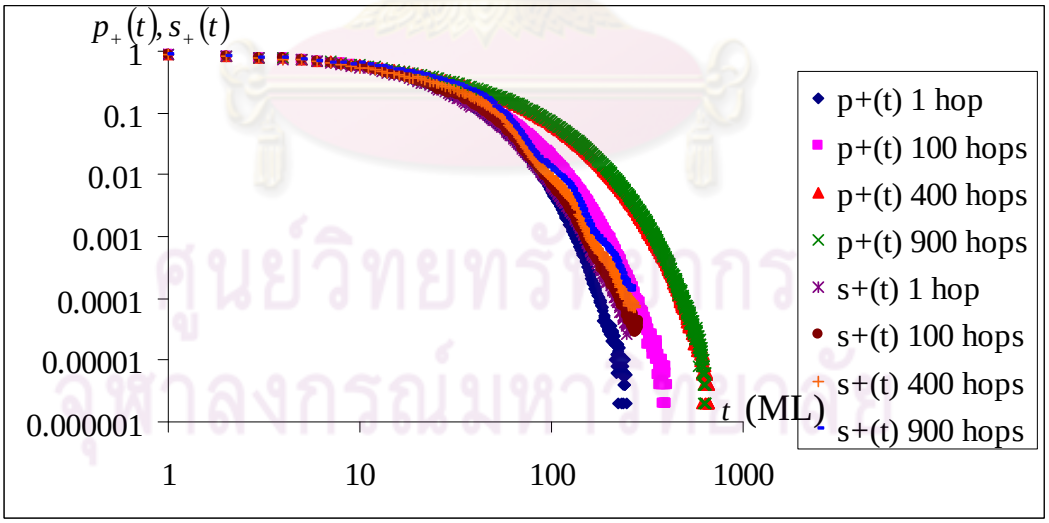


Figure 4.18 : Log-log plots of positive survival distribution vs. persistence distribution of the Family films on stepped surfaces**.**

Figure 4.17 and 4.18 show that persistence and survival probabilities of the Family atoms. The survival probabilities decrease faster than persistence probabilities. The references of survival are average values of new step edges each time. The average references grow with time. Atoms hardly escape from the evolution references. The Family atoms diffuse further than the DT atoms.

The persistence probability exponents of the Family model are in the table 4.2. These results can be described with figure 4.5 and 4.6. A little flowing of atoms happen for the Family model since 1 hop but step edges cannot be separated. This interesting issue occurs in growth process of the Family model. Because of the diffusion rule of the Family model, number of bond is not important. Most atoms must incorporate at step edges for 1 hop. It looks like flowing. Table 4.2 shows the trend of both positive and negative exponents of the Family model. Each curve decreases when the number of hops or temperature increases as the curve of the DT model, even though the value of exponents are not the same. They rapidly decrease with temperature. This shows that the step-flow growth mode can be happened well for high temperature. These results correspond to step edge position going forward with high temperature.

Table 4.2 shows both exponents of the DT and the Family model. The exponents of the Family model decrease greatly with temperature. Since diffusion rule of the Family model, atoms can flow any sites on lower terrace. The atoms have more probabilities to flow far from the references. We compare exponents between of the DT and the Family at the same hop since 100 hops (step-flow growth begins). The DT diffusion rule effects to incorporated sites of atoms. The DT atoms mostly deposit on edges. Even if high temperature is applied, the DT atoms cannot diffuse as far as the Family atoms.

Table 4.2 : Negative and positive temporal persistence exponents.

Number of hops	DT model		Family model	
	θ_p^-	θ_p^+	θ_p^-	θ_p^+
1	8.99	8.97	8.99	9.26
100	9.18	9.24	5.60	5.57
400	8.32	8.38	3.54	3.45
900	8.07	8.10	3.51	3.31

Chapter V
Conclusion

In summary, the growth of thin film on vicinal substrate is studied using the 2+1 Das Sarma-Tamborenea model and the 2+1 Family model. This growth process is a stochastic process that the Monte Carlo technique can be used to simulate thin films. The goal of this study is to see the influence of substrate temperature on the step-flow growth mode. Quantities of interest are morphologies, temporal correlation function, persistence probabilities and survival probabilities. The morphologies show how the film surfaces look like. This helps us to understand some atomic activities during the growth process. The atomic processes are further investigated via the temporal correlation function. The flow of atoms at the step edges is observed via the persistence and survival probabilities of the step edge positions in the films.

From the shown morphologies, it is clear that for the DT model, the step-flow growth mode begins when the temperature is elevated. The step-flow growth occurs easier for the Family films because the Family diffusion rules, even without the noise reduction technique, help generate very smooth films.

Calculated correlation function of thin films grown on stepped surfaces of both the DT and Family model increase with time. The type of atomic process that occurs depends on temperature and diffusion rule of chosen model. The values of slope of $G-t$ plots are used to predict transition of atomic process from low to high temperature. Atomic processes of the DT and Family model are characterized for both low and high temperature cases. There are deposition of atoms at the edges in low temperature simulations and diffusion of atoms on the terraces in high temperature simulations. Large diffusion length makes large distribution for the Family model.

To calculate persistence probability of thin films grown on stepped surfaces is another objective of this work. Probabilities decrease with time for all models. Atoms have more chance to return to the initial value when time is large. The experimental results (Constantin et al., 2007) confirm power law relation of the persistence probability distribution for Al/Si(111) surface steps at 770, 870 and 970 K. Persistence probability exponents for all models decrease with temperature or diffusion length. Larger diffusion length leads to better “step-flow” growth. From the experimental results (Dougherty et al., 2002), Si(111) at temperature

between 770 and 970 K is used to find different exponents in different physical mechanisms. The persistence exponents of the Family atoms decrease greatly. The Family diffusion rule support atoms in diffusion further than the DT atoms. Negative persistence exponents are not as far as positive persistence exponent. Negative and positive survival probability distributions have the same trend. The persistence and survival probability distributions are compared. The persistence distributions decrease with the same rate as survival distributions in the DT model because the DT atoms cannot diffuse far away. For the Family atoms, the survival probabilities decrease faster than the persistence probabilities. The Family atoms can flow to a far distance so the average references follow the atoms. The Family atoms cross these average references more than the initial references. This point supports the evolution of references of survival.

The last objective of this work is to find conditions for step-flow growth mode in thin films grown on stepped surfaces by Monte Carlo simulations. These above conclusions correspond to step-flow growth mode, atoms flow to the end of step starting at 100-900 hops. The number of diffusion is adjusted by adding substrate temperature. The substrate temperature is added for proper diffusion length. The step-flow growth mode happens when the diffusion length is a half of terrace width and more. The best step-flow growth occurs when the diffusion length equals terrace width. If the diffusion length is larger than the terrace width, the film is smoother but there is little influence to step-flow growth mode. The results correspond to the smooth films that are grown in this mode. These results are consistent with theoretical (Dougherty et al., 2004; Giesen, 2001) and experimental (Constantin et al., 2007; Giesen-Seibert et al., 1995) researches.

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Vitae

Jindarat Yaemwong was born on January 3, 1982. She received her Bachelor of Science (Cum Laude) in Physics in 2004 and her Graduate Diploma in teaching in 2005 from Burapha University. She continued her Master study in Physics at Chulalongkorn University in 2008.