Chapter I

Introduction and Theory

Recently, there are widely interests and studies on the kinetic surface roughening growth [1, 2, 3, 4, 5]. Especially, the molecular beam epitaxy (MBE) [1] thin film growth, i.e. the epitaxial growth by using MBE technique. This technique receives much interest from both experimentalists and theorists [1, 2] due to the fact that it produces very smooth films. The MBE is also a very important technique used in thin film industrials to grow high quality films. Because the rough surfaces have poor contact properties and unusable for electronic devices, scientists try to understand the behavior of the MBE growth and find a way to describe the kinetic surface roughening growth process.

In this thesis, we use computer simulations to study kinetic nonequilibrium surface roughening growth systems. The computer simulation is a convenient tool to use and it is easy to control or neglect some parameters such as substrate temperature or bonding energy depending on the aim of each study. There are many discrete growth models [1, 3, 4, 5] used to describe MBE growth processes via computer simulations. Among these many discrete growth models, we are most interested in the model that have diffusion rules which followed the *ideal* low temperature MBE growth and solid-on-solid (SOS) constraints (not allowing desorption, overhanging and bulk vacancies formation in the growing films). This model is known as the *Wolf-Villain* (WV) model [3] which will be described in more detail in the next chapter.

In real MBE growth, there is a step-edge potential barrier known as the *Ehrlich-Schwoebel* (ES) barrier [6, 7, 8]. The ES barrier prevents an atom from diffusing down to the lower terrace from the upper terrace (more details in section 2.2). We are interested in studying effects of the ES barrier in WV model. So we add the ES barrier into the WV model by modifying the model diffusion rule.

Then we find the effect of ES barrier on the WV model and compare with the original WV model.

To understand the kinetics properties we must have some background as the tools to study and understand the model. In this chapter, we will introduce some background on scaling hypothesis. This approach has become a standard tool to describe the kinetic surface roughening after Family and Vicsek [9] formulated a scaling law for the ballistic deposition (BD) model in 1985. Moreover, the concept of universality class, continuum growth equations, particle diffusion current, and correlation function are also introduced here as follow:

1.1 Scaling

In order to understand the kinetic surface roughening behavior in MBE growth, we study a quantity which can be used to describe the grown film quantitatively. That quantity is the *surface width*, W, which is the root mean square height fluctuation. It is a standard deviation of the interface height $h(x,t)$ which is a function of the substrate size L and time t, and it is defined as [1]

$$W(L,t) \equiv \sqrt{\frac{1}{L}\sum_{x=1}^{N}[h(x,t)-\bar{h}(t)]^2}, \tag{1.1}$$

where $h(x,t)$ is the height at the site x and at time t above the flat substrate. $\bar{h}$ is the average height of the surface defined as

$$\bar{h}(t) \equiv \frac{1}{L}\sum_{x=1}^{N} h(x,t). \tag{1.2}$$

In the growth process, if the deposited particles arrive on the substrate at a constant rate, the average height of the surface increases linearly with time as

$$\bar{h}(t) \propto t. \tag{1.3}$$

If we plot the time evolution of the surface width in log-log scale, see Fig. 1.1, the surface width has two regimes separated by the time t_c which is called the

crossover time. In the first region (where $t \ll t_c$) we see that the surface width increases as a power law with time

$$W(L,t) \propto t^{\beta}, \tag{1.4}$$

where β is the *growth exponent.* The growth exponent β characterizes the time-dependent dynamics of the roughening process in $t \ll t_c$ regime. As seen in Fig. 1.1, the surface width W does not increase indefinitely. After the crossover time t_c, W saturates. If we plot the width of systems with different substrate size $L =$ 20, 30, 40, 60 respectively, see Fig. 1.2, we find that the saturation value of the surface width, W_{sat}, increases as the substrate size L increases and it also depends on a power law

$$W_{sat}(L) \propto L^{\alpha}, \tag{1.5}$$

where α is called the *roughness exponent.* The roughness exponent α characterizes the roughness of the surface at $t \gg t_c$ regime. Moreover, the crossover time of the systems also increases as the substrate size increases, see Fig. 1.2. The crossover time actually depends on the substrate size L as the power law of L as

$$t_c \propto L^{z}, \tag{1.6}$$

where z is called the *dynamical exponent.* We can combine the above relations, Eq. (1.4) and Eq. (1.5), into a single relation as [1]

$$W(L,t) \propto L^{\alpha} f(t/L^{z}), \tag{1.7}$$

where this relation is the *scaling relation* for W. The function $f(t/L^{z})$ is called the scaling function. The scaling function has two regimes. The first regime, for the small value of t/L^{z} ($t/L^{z} \ll 1$, implying $t \ll L^{z}$ or $t \ll t_c$) the scaling function increases as a power law as

$$f(t/L^{z}) \propto (t/L^{z})^{\beta}. \tag{1.8}$$

The second regime is when $t \to \infty$ ($t/L^{z} \gg 1$, imply $t \gg L^{z}$ or $t \gg t_c$). This is the saturation regime and we have

$$f(t/L^{z}) \propto constant, \tag{1.9}$$

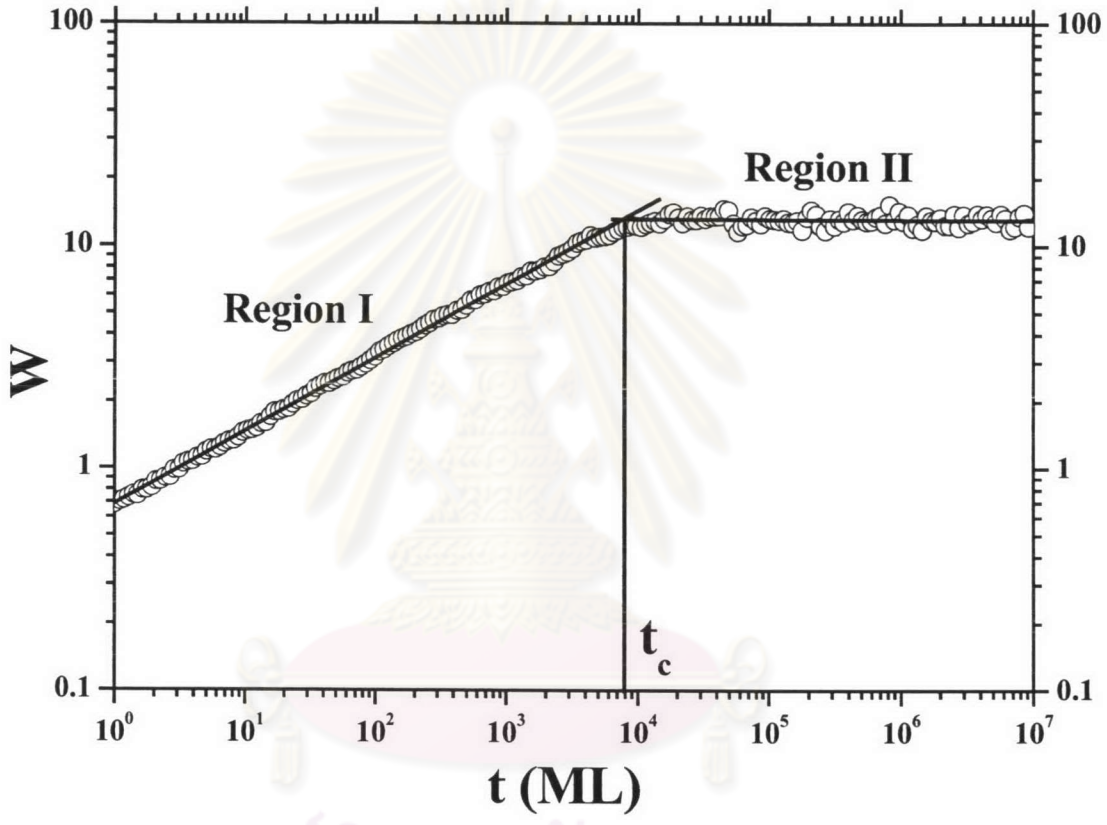

Figure 1.1: The surface width plot versus time of the system with $L = 50$. There are two regimes in this figure. The first one denoted as Region I, the surface width increases with time as a power law. The second regime, Region II, is the saturation regime that start after the crossover time t_c.

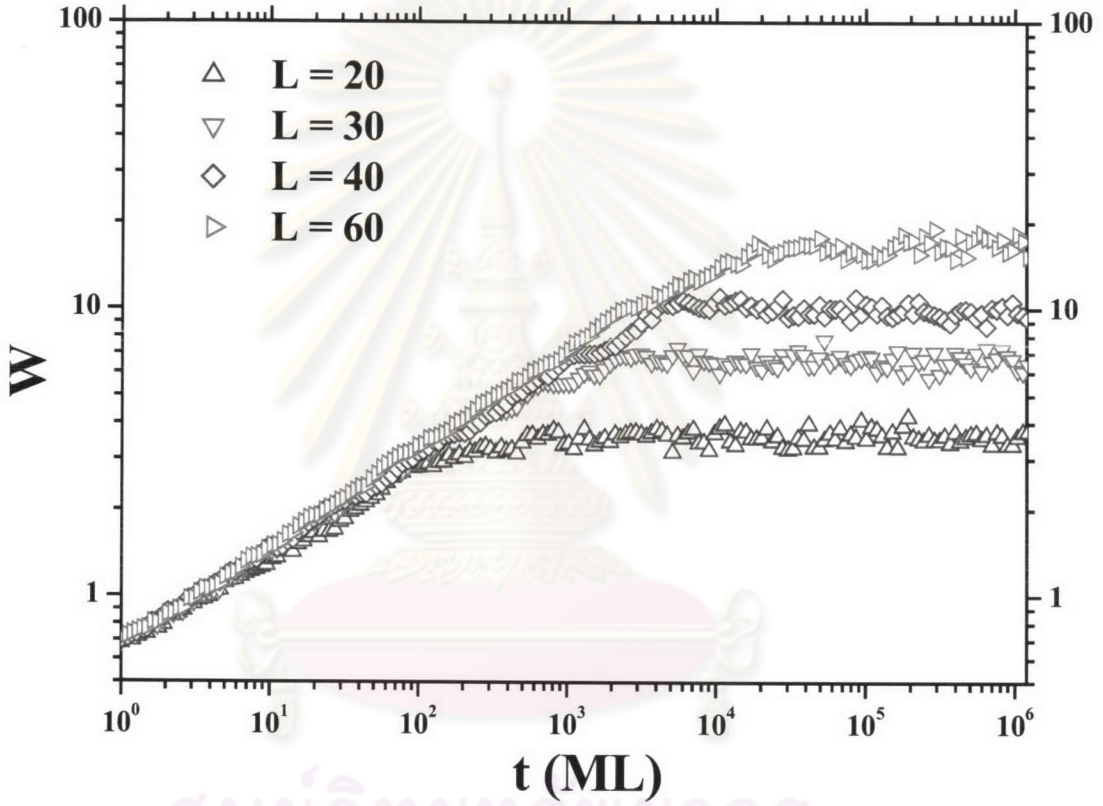

Figure 1.2: The surface width plot versus time t of the systems with different substrate size L = 20, 30, 40, 60 respectively. The saturation regime and the crossover time increases as the substrate size L increases.

for this limit.

The crossover time t_c is at the boundary between these two regimes, so it satisfies both Eq. (1.4) and Eq. (1.5). From this, if we approach the crossover time t_c from the left hand side we find

$$W(t_c) \propto t_c^{\beta}, \tag{1.10}$$

corresponding to Eq. (1.4). On the other hand, if we approach t_c from the right hand side we find

$$W(t_c) \propto L^{\alpha}, \tag{1.11}$$

corresponding to Eq. (1.5). From these two relations we can write

$$t_c^{\beta} \propto L^{\alpha}. \tag{1.12}$$

Substitute t_c from Eq. (1.6) we obtained

$$(L^z)^{\beta} \propto L^{\alpha}, \tag{1.13}$$

which yields

$$z = \alpha/\beta. \tag{1.14}$$

Eq. (1.14) links the three exponents α, β, z. This means there are only two independent exponents. This relation is valid for growth processes which follow the scaling relation Eq. (1.7). All of these critical exponents (α, β, z) define the *universality class* of a system, as will be discussed in more details in the next section.

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1.2 Universality Class

The universality class of a growth system is defined by the set of critical exponents (roughness exponent α, growth exponent β, and dynamical exponent z). It is used to characterize the *asymptotic* (at long time, $t \to \infty$, and long distance, $x \to \infty$) properties of the kinetic roughening in epitaxial growth. In general, we need only two exponents, i.e. α and β, to define the universality class of a system because

the other exponent (z) can be represented in the form of α and β as $z = \alpha/\beta$, Eq. (1.14). There are only a few known universality classes describing asymptotic growth properties in many nonequilibrium surface growth models. There are four types [10] of universality classes for kinetic surface roughening in epitaxial growth.

1. Kardar-Parisi-Zhang (KPZ) universality

The growth process will belong to the Kardar-Parisi-Zhang (KPZ) universality class [11] if the process allows desorption, overhang, and bulk vacancies in the growing films. The critical exponents are known [11, 12] in 1+1 dimensions (one dimensional substrate) to be $\alpha = 1/2$, $\beta = 1/3$, and $z = 3/2$. In 2+1 dimensions (two dimensional substrate), the critical exponents are known approximately by numerical simulations to be $\alpha \approx 0.39$, $\beta \approx 0.24$, and $z \approx 1.61$.

2. Edwards-Wilkinson (EW) universality

The Edwards-Wilkinson (EW) universality [13] describes a growth system with no overhang, no vacancy and no desorption in the process. This type of growth is a conserve growth which is under the SOS conditions. Atoms try to diffuse down to lower terraces during EW growth. The critical exponents are exactly known in both 1+1 and 2+1 dimensions [1, 10] with $\alpha = 1/2(0)$, $\beta = 1/4(0)$, and $z = 2(2)$ in 1+1(2+1) dimensions. In 2+1 dimensions, the exponents $\alpha = \beta = 0$ predicts very smooth surfaces. The EW universality class describes the smoothest morphology in both 1+1 and 2+1 dimensions because the values of the roughness exponent (α) and the growth exponent (β) are the smallest values possible.

3. Mullins-Herring (MH) universality

The Mullins-Herring (MH) universality [14, 15] also describes a system under SOS constraints. The critical exponents in 1+1(2+1) dimensions are [10] $\alpha = 3/2(1)$, $\beta = 3/8(1/4)$, and $z = 4(4)$. The exponents α and β in MH universality are the largest values among these four universality classes. They imply that the growth morphology has the most kinetic surface roughness. The dynamical exponent $z = 4$ is a very large value. So it takes a long time for the surface width to saturate.

4. Molecular Beam Epitaxy (MBE) universality

The critical exponents for the MBE universality class are known by calculation [16], $\alpha = 1(2/3)$, $\beta = 1/3(1/5)$, and $z = 3(10/3)$ in d = 1+1(2+1) dimensions.

1.3 Continuum Growth Equations

To understand the kinetic surface roughening phenomena, we want to make a connection between results obtained from the discrete growth model simulations and the continuum growth equation of motion which is used to describe the growing interfaces on the coarse-grained scale. In the continuum growth equation approach, we want to find the equation for the time derivative of the surface height $h(x,t)$ which can be written in the form [1]

$$\frac{\partial h(x,t)}{\partial t} = G(h,x,t) + \eta(x,t), \tag{1.15}$$

where $G(h,x,t)$ is a general function that depends on the interface height $(h(x,t))$, position (x), and time (t) and $\eta(x,t)$ is the fluctuations due to the random deposition process. To derive the stochastic growth equation of motion, all terms must follow symmetry principles [1, 12] of the problem:

1. Invariance under translation in time.

The growth equation must be invariant under the translation $t \to t + \delta t$ because it must not depend on where we define the origin of time. So terms that depend explicitly on t cannot be included and the survived terms are the time derivative of h, $\partial h/\partial t$, and its higher order terms because they are invariant under translation in time.

2. Invariance along the growth direction.

The growth equation must be invariant under the translation $h \to h + \delta h$ because it must be independent of where we define the level $h = 0$. So terms that depend explicitly on h cannot be included and the survived terms are the combinations of ∇h, $\nabla^2 h$, ..., $\nabla^n h$ terms.

3. Invariance along the direction perpendicular to the growth direction.

The growth equation must be invariant under the translation $x \to x + \delta x$ because it should be independent of the actual value of the position x on the substrate (where we assume that the substrate is homogeneous). So there cannot be any terms that depend explicitly on x.

4. Invariance under rotation and inversion symmetry about the growth direction.

The growth equation must be invariant under the transformation $x \rightarrow$ -x. This symmetry eliminates the odd order derivative ∇h, $\nabla(\nabla h)^2$, ... terms from the growth equation. The survived terms are $(\nabla h)^2$, $\nabla^2 h$ and their combination terms.

The general form of the stochastic continuum equation after we eliminate all terms that violate any symmetry listed above is [1, 12]:

$$\frac{\partial h(x,t)}{\partial t} = \nabla^2 h + \nabla^4 h + ... + \nabla^{2n} h + (\nabla^2 h)(\nabla h)^2 + ... + (\nabla^{2k})(\nabla h)^{2j} + \eta(x,t), \quad (1.16)$$

where n, k, j are positive integer numbers.

Because we are interested in the asymptotic properties of the interfaces, the higher order terms are less important when comparing with the lower orders terms. We can rewrite Eq. (1.16) in the most general form for conserved epitaxial growth including all terms up to the fourth order as [2, 10, 17]:

$$\frac{\partial h}{\partial t} = \nu_2 \nabla^2 h - \nu_4 \nabla^4 h + \lambda_{22} \nabla^2 (\nabla h)^2 + \lambda_{13} \nabla (\nabla h)^3 + \eta(x,t). \quad (1.17)$$

Note that h represents the height fluctuation around the average surface height, h = h - $\bar{h}$, rather than the actual height itself. This is because we do not include a term representing the deposition flux in the equation. From the renormalization point of view, the most relevance term in Eq. (1.17) is $\nabla^2 h$ if $\nu_2 \neq 0$ [18]. This term is known as the Edward-Wilkinson (EW) term [13] and lead to the EW asymptotic universality class that we discussed in the previous section. The stochastic growth equation that includes only the EW term is

$$\frac{\partial h}{\partial t} = \nu_2 \nabla^2 h + \eta(x,t), \quad (1.18)$$

and it is generally called the EW equation [13]. If $\nu_2 = 0$, the growth Eq. (1.17) can still leads to the EW universality class because it was found [17, 18, 19] that the $\lambda_{13} \nabla (\nabla h)^3$ term is the higher order form of the EW term whenever $\lambda_{13} \neq 0$.

The extension of the EW equation to include the nonlinear second order term is proposed by Kardar, Parisi and Zhang [11]

$$\frac{\partial h}{\partial t} = \nu_2 \nabla^2 h + \lambda_2 (\nabla h)^2 + \eta(x,t), \quad (1.19)$$

where we called Kardar-Parisi-Zhang (KPZ) equation [11] and this equation lead to the KPZ universality class that describes nonconserved growth process. In general, this equation is used to describe the asymptotic behavior of nonconserved MBE growth that allows desorption, overhanging, and bulk vacancies in a growing film.

In the case when $\nu_2 = \lambda_{13} = 0$, the most relevance term is the $\nabla^2(\nabla h)^2$ term and the growth Eq. (1.17) become

$$\frac{\partial h}{\partial t} = -\nu_4 \nabla^4 h + \lambda_{22} \nabla^2 (\nabla h)^2 + \eta(x,t). \tag{1.20}$$

This is the nonlinear fourth order growth equation, which is used to describe the MBE growth [1, 2] and is called the MBE growth equation. This equation leads to the MBE universality class. The linear form of Eq. (1.20) is called the Mullins-Herring surface diffusion equation [2, 10, 14, 15, 19] which is in the form

$$\frac{\partial h}{\partial t} = -\nu_4 \nabla^4 h + \eta(x,t), \tag{1.21}$$

This equation leads to the MH universality class.

The results of the theoretically calculated asymptotic exponents [19] from Eq. (1.18) through Eq. (1.21) which corresponds with the four asymptotic universality classes are listed in Table 1.1.

Dimensions	**1+1**			**2+1**		
Exponent	α	z	β	α	z	β
$\nabla^2 h$	1/2	2	1/4	0(log)	2	0(log)
$(\nabla h)^2$	1/2	3/2	1/3	~ 0.4	~ 1.67	~ 0.24
$\nabla^4 h$	3/2	4	3/8	1	4	1/4
$\nabla^2(\nabla h)^2$	1	3	1/3	2/3	10/3	1/5

Table 1.1: The theoretical asymptotic exponents in d = 1+1 and d = 2+1 dimensions for various continuum growth equations.

1.4 Particle Diffusion Current

For our nonequilibrium growth model, we consider the model as a conserved growth model. It implies that in our growth process there are mass and volume conservation. Consequently, the surface fluctuation of the growing films can be described by the continuity, coarse-grained equation as [2]

$$\frac{\partial h}{\partial t} = -\nabla \cdot J + \eta(x,t), \tag{1.22}$$

where J is a conserved *particle diffusion current* which is parallel to the horizontal average surface height direction [2, 20]. Comparing Eq. (1.22) to Eq. (1.17), the particle diffusion current, including terms up to the fourth order, is written in the form

$$J = -\nu_2 \nabla h - \lambda_{13}(\nabla h)^3 + \nabla[\nu_4 \nabla^2 h - \lambda_{22}(\nabla h)^2]. \tag{1.23}$$

The particle diffusion current, J, in this equation is a function of the derivatives of h and a leading term is the ∇h term which is the local inclination. The current can be measured directly by the study of growth on tilted substrates [20].

To measure the particle current, we simplify the current in Eq. (1.23) and keep only the most relevance term as

$$J = -\nu_2 \nabla h, \tag{1.24}$$

where ν_2 is the EW coefficient. The growth process is started on a tilted substrate. By "tilted substrate", it means the substrate has a non-zero inclination $\tan\theta$ and the substrate height at any position x and initial time $t = 0$ is set to be

$$h(x) = x \cdot \tan\theta, \tag{1.25}$$

for growth on one dimensional substrate (where the standard initial condition is $h(x) = 0$) as shown in Fig. 1.3. We count the numbers of diffusing atoms during growth process simulations. If an atom hops in the *uphill(downhill)* direction (represented by black arrow in Fig. 1.3), it contributes to a positive(negative) current which is shown by the red(green) arrow in Fig. 1.3(b). For atoms that do not move (stick at the deposited site) after the deposition, the current from

these atoms are zero. After the growth process is completed, we calculate the net current from the following equation:

$$J_{avg} = \frac{|J_u| - |J_d|}{n}, \tag{1.26}$$

where J_{avg}, $|J_u|$ and $|J_d|$ are the net current, the total uphill current and the downhill current in the system respectively. n is the total number of the deposited atoms. Then we consider the net current J_{avg} as follows. If the net current is positive the system is said to have an *uphill current* but if the net current is negative the system has a *downhill current.* Krug *et al.* [20] suggested that this is a powerful method to determine the true asymptotic behavior of model by determining if the EW term ($\nabla^2 h$) should be included in the continuum equation describing the model. Sign of the coefficient ν_2 in Eq. (1.17) can also be determined by this method. For a system with an *uphill* current, $j > 0$ (implying $\nu_2 < 0$), we have an unstable surface with mound formation or instabilities where as in a system with a *downhill* current, $j < 0$ (implying $\nu_2 > 0$), the surface is stable and belongs to the EW asymptotic universality class. But there is a limit to this method because there are growth models that this method does not work. One example is the Das Sarma-Tamborenia (DT) model [4, 5] that has an average zero particle diffusion current [2] which implies $\nu_2 \equiv 0$ in the model. For the case of $\nu_2 = 0$, we cannot find the true asymptotic universality class of these models by using this method. In a model with zero current, we can only conclude that the EW term is not in the continuum equation describing the model [2]

1.5 Correlation Function

In the study of thin film growth, sometimes it is difficult to distinguish a mounded surface morphology from a dynamically rough one. A useful tool that can help us distinguish between mound formation and dynamically rough is the calculation of the height-height correlation function [2, 21, 22]

$$G(\mathbf{r}) = \langle h(\mathbf{x}) h(\mathbf{x} + \mathbf{r}) \rangle_x, \tag{1.27}$$

where h is the deviation of the surface height from the average height and $r = |\mathbf{r}|$ is the distance between two sites on the substrate.

If we calculate the height-height correlation function $G(r)$ and find an oscillation of $G(r)$ as a function of r, see Fig. 1.4, it implies [2, 21, 22] regular mound formation on the surface. If there is no oscillation, it implies no mound formation on the surface. Moreover, we can obtain an average mound height (H) from a $G(r)$ versus r plot through the definition [2, 23]

$$H(t) = \sqrt{G(r=0)}. \tag{1.28}$$

Conventionally, the smallest distance r which makes $G(r) = 0$, i.e. the distance of the first zero crossing of $G(r)$, is taken to be an average mound radius. The average mound radius, R, scales with growth time as [2, 23]

$$R(t) \propto t^n, \tag{1.29}$$

where $n \simeq z^{-1}$ ($= \beta/\alpha$) is the coarsening exponent which describes how the individual mound size increases in time.

1.6 Overview of the Thesis

In this chapter, we introduced the theoretical background needed for the study of far from equilibrium computational models for MBE growth. The Wolf-Villain (WV) model [3] is a simple model that we are interested in this study. It is a model with an instantaneous diffusion process. We are also interested in the effect of the step-edge potential barrier, known as the Ehrlich-Schwoebel (ES) barrier [6, 7] in the WV model. The details description of the model is presented in Chapter 2.

The results of our simulations in 1+1 dimensions of both WV model and WV model with ES barrier are presented in Chapter 3. The results of the particle current and correlation functions are also shown here. The conclusions of our work are presented in Chapter 4. In this last chapter, the true asymptotic universality class of the WV model is also discussed.

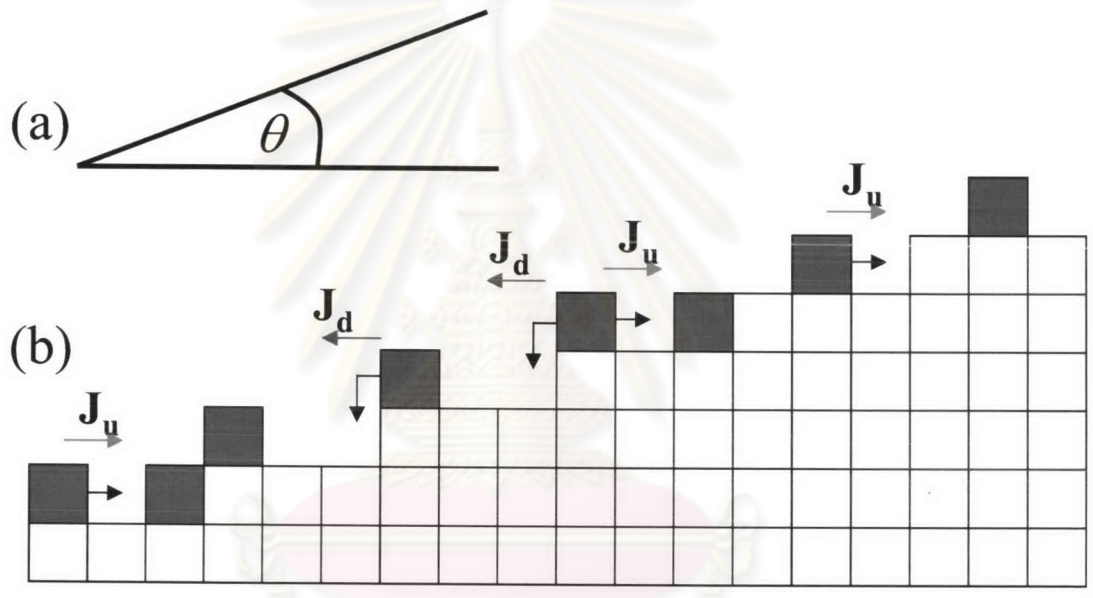

Figure 1.3: (a) The tilted substrate with the slope $\tan\theta$. (b) The method to contributes the particle diffusion current to uphill (red arrow) and downhill (green arrow) directions.

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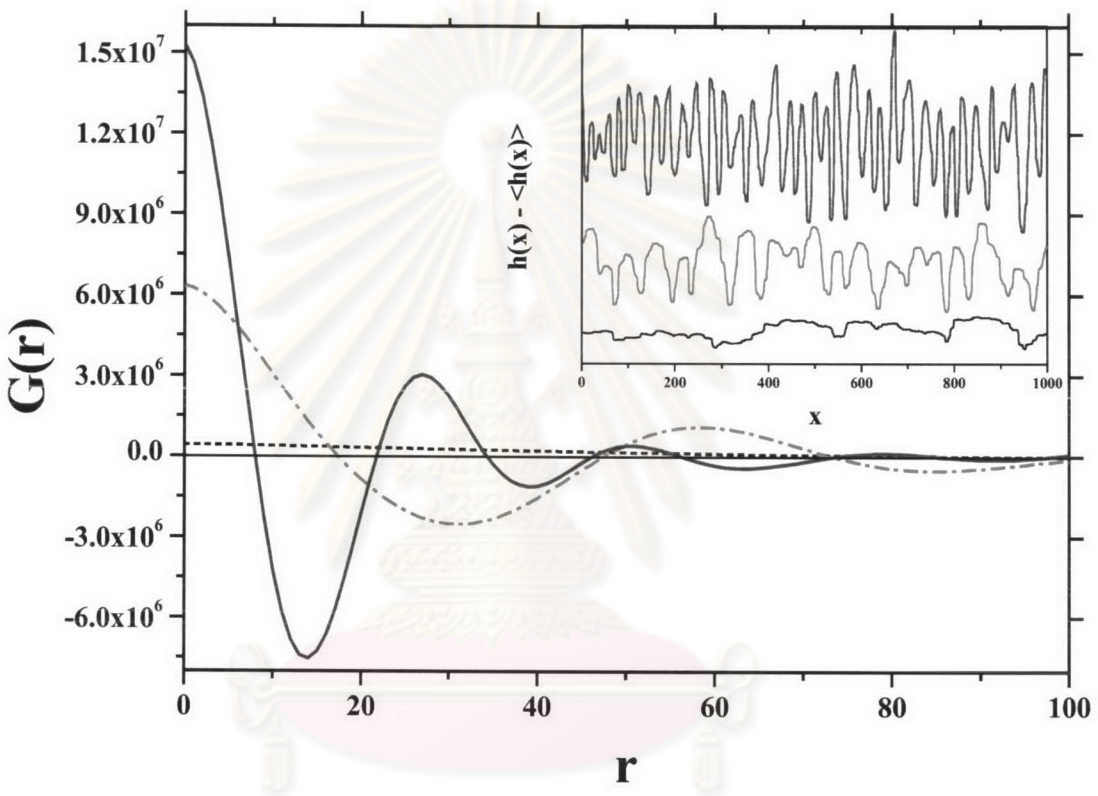

Figure 1.4: The height-height correlation function $G(r)$ shows the oscillations as a function of r. It implies mound formation on the surface (the blue line, the red dash and the dotted lines). But if there is no oscillations (the black short dash line), it implies no mound formation on the surface. Inset: The morphologies of the systems that use to calculate $G(r)$.