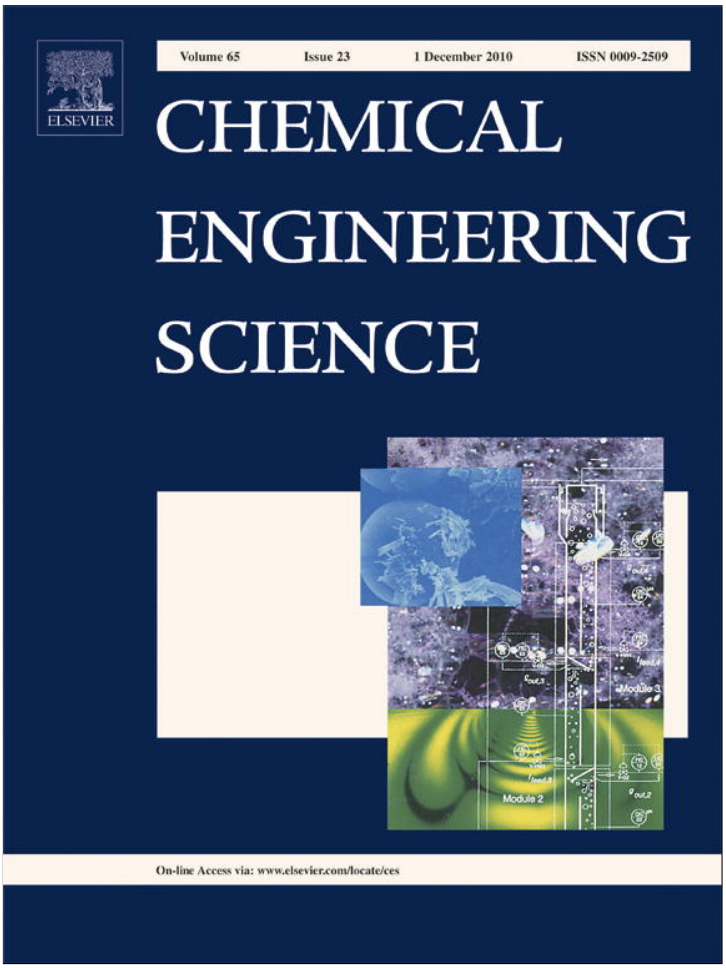

Chemical Engineering Science 65 (2010) 6101–6111

Contents lists available at ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

Dependence of film surface roughness and slope on surface migration and lattice size in thin film deposition processes

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ARTICLE INFO

Article history:
Received 22 July 2010
Received in revised form
24 August 2010
Accepted 25 August 2010
Available online 28 September 2010

Keywords:
Thin film growth
Microscopic modeling
Surface migration
Surface roughness
Surface slope
Lattice size

ABSTRACT

This work focuses on the study of the dependence of film surface roughness and slope on lattice size in thin film deposition processes. Two different models of thin film deposition processes, in both 1D and 2D, are considered: random deposition with surface relaxation model and deposition/migration model. Surface roughness and surface slope are defined as the root-mean-squares of the surface height profile and of the surface slope profile, respectively. Both surface roughness and slope evolve to steady-state values at large times but are subject to different dynamics and scaling properties. A linear and a logarithmic dependence of surface roughness square on lattice size are observed in the 1D and 2D lattice models, respectively, in both the random deposition with surface relaxation model and the deposition/migration model with zero activation energy contribution from each neighboring particle. Furthermore, a stronger lattice-size dependence is found in the deposition/migration model when the migration activation energy contribution from each neighboring particle becomes significant. On the other hand, a weak lattice-size dependence is found for the surface mean slope in all growth models considered, especially at large lattice sizes. Finally, the dynamics of surface roughness and surface slope is studied with respect to different characteristic length scales.

1. Introduction

Thin-film silicon solar cells are currently the most widely investigated and used thin-film solar cells. However, an improved conversion efficiency of the solar energy is essential for a wider application of thin-film silicon solar cells. In this context, research has been conducted on the optical and electrical modeling of thin-film silicon solar cells, which indicates a direct relationship between the light scattering/trapping properties of the thin film interfaces and the conversion efficiencies of thin-film silicon solar cells (Krč et al., 2003; Müller et al., 2004). Recent studies on enhancing thin-film solar cell performance (Zeman and Vanswaaij, 2000; Poruba and Fejfar, 2000; Müller et al., 2004; Springer and Poruba, 2004; Rowlands et al., 2004) have shown that film surface and interface morphology, characterized by root-mean-square roughness (rms roughness, r) and root-mean-square slope (rms slope, m), play an important role in enhancing absorption of the incident light by the semiconductor layers. Specifically, significant increase of conversion efficiency by introducing appropriately rough interfaces has been reported in several works (Tao and Zeman, 1994; Leblanc and Perrin, 1994; Krč and Zeman, 2002). Therefore, it is important to tailor thin film surface morphology characteristics to desired values.

In the context of modeling of thin film growth and surface morphology, two mathematical approaches have been developed and widely used: kinetic Monte Carlo (kMC) methods and stochastic differential equation (SDE) models. KMC methods provide unbiased realizations of thin film growth processes based on pre-defined microscopic rules. The corresponding thermodynamic and kinetic parameters that differentiate the microscopic growth models are obtained from experiments and/or molecular dynamics simulations (Levine et al., 1998; Zhang et al., 2004; Levine and Clancy, 2000; Christofides et al., 2008). However, kMC models are not available in closed form, and thus, they cannot be readily used for feedback control design and system-level analysis. On the other hand, SDE models can be derived from the corresponding master equation of the microscopic process and/or identified from process data. Specifically, methodologies have been developed to construct SDE models and estimate their parameters from first principles (e.g., Haselwandter and Vvedensky, 2006, 2007, 2008) and numerical simulations (e.g., Christofides et al., 2008; Hu et al., 2008, 2009b). The closed form of the SDE models enables their use as the basis for the design of feedback controllers which can regulate thin film surface roughness (Christofides et al., 2008; Hu et al., 2008), film

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0009-2509/$ - see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ces.2010.08.035

porosity (Hu et al., 2009d, 2009b), and film thickness (Hu et al., 2009c).

Different models have been developed to describe thin film growth processes. Specifically, a random deposition with surface relaxation (RDSR) model was introduced by Family (1986). The dependence of surface irregularity on the lattice size of the RDSR model, i.e., scaling properties, was investigated via both numerical simulations and theoretical derivations (Family, 1986; Barabási and Stanley, 1995). A competitive growth model that considers separate deposition and migration processes (deposition/migration model) was further developed to capture the balance of these two processes at different substrate temperatures (Horowitz and Albano, 2003; Ni and Christofides, 2005). The dynamics and the scaling properties of surface roughness in the deposition/migration model were also investigated both numerically and analytically and revealed a temperature dependence (Chou et al., 2009; Chou and Pleimling, 2009). Nevertheless, a close look in the literature indicates that the dynamics and scaling properties of surface slope have not been extensively studied. In a recent work (Huang et al., in press), we focused on a thin film growth process taking place in a 1D triangular lattice and found a weak surface slope dependence on lattice size. The dynamics, scaling properties, and the influence of preferential migration on root-mean-square (rms) slope and roughness of surface height profiles in thin film deposition processes still remain open problems and have both theoretical and practical significance on identifying the dynamic models and controlling thin film surface morphology for solar cells.

Motivated by the above considerations, this work focuses on the study of the dependence of film surface roughness and slope on the lattice size in thin film deposition processes. Both RDSR and deposition/migration models are considered on a square lattice in both 1D and 2D using the solid-on-solid assumption. Kinetic Monte Carlo methods are used to simulate both models. The surface roughness and surface slope are defined as the root-mean-squares of the surface height profile and of the surface slope profile, respectively. We find that both surface roughness and slope evolve to steady-state values at large times but are subject to different dynamics and scaling properties. A linear dependence and a logarithmic dependence of surface roughness square on the lattice size are observed in the 1D and 2D models, respectively, of the random deposition with surface relaxation model and the deposition/migration model with zero activation energy contribution from each neighboring particle. Furthermore, a stronger lattice-size dependence is found in the deposition/migration model with a significant migration activation energy contribution from each neighboring particle. This finding suggests that preferential migration (i.e., surface particles with zero or one nearest neighbor dominate the migration events) results in a stronger dependence of surface roughness on the lattice size in thin film deposition processes. In contrast, a weak lattice-size dependence is found for the surface mean slope in all growth models considered, especially at large lattice sizes. Finally, the different dynamics of surface roughness and slope evaluated under different characteristic length scales are investigated, and the need for spatially distributed control actuation to induce desired roughness and slope levels at large characteristic length scales is demonstrated.

2. Description of thin film deposition processes

In this section, two standard different thin film deposition process models are presented: a random deposition with surface relaxation (RDSR) model and a process model involving deposition and surface migration (deposition/migration model). Both deposition models are constructed on a square lattice in both one-dimension (1D) and two-dimensions (2D) using the solid-on-solid (SOS) assumption, where particles land on top of the existing surface particles. Periodic boundary conditions are applied to these lattice models in the directions that are perpendicular to the growth direction. Lattice size is defined as the number of sites in the lateral direction. In the 2D lattice, the lattice sizes in the two lateral directions are the same, i.e., the deposition process models take place on a square lattice in the 2D case.

Fig. 1 shows the lattice models of the thin film deposition processes in both 1D and 2D cases. In Fig. 1, the incident particles are deposited vertically onto the thin film. The surface particles, i.e., the particles on top of each lattice site, are subject to an instantaneous surface relaxation event (the RDSR model) or a migration event (the deposition/migration model). The details of the microscopic events in these two models will be discussed in the following subsections. Kinetic Monte Carlo methods are used to simulate both deposition process models. Specifically, we use the continuous-time Monte Carlo (CTMC) algorithm (Vlachos et al., 1993) to simulate the thin deposition process models.

2.1. Random deposition with surface relaxation model

The RDSR model is a convenient basic model of the thin film deposition process since its microscopic rules are simple and its dynamic behavior is known (Huang et al., in press; Barabási and

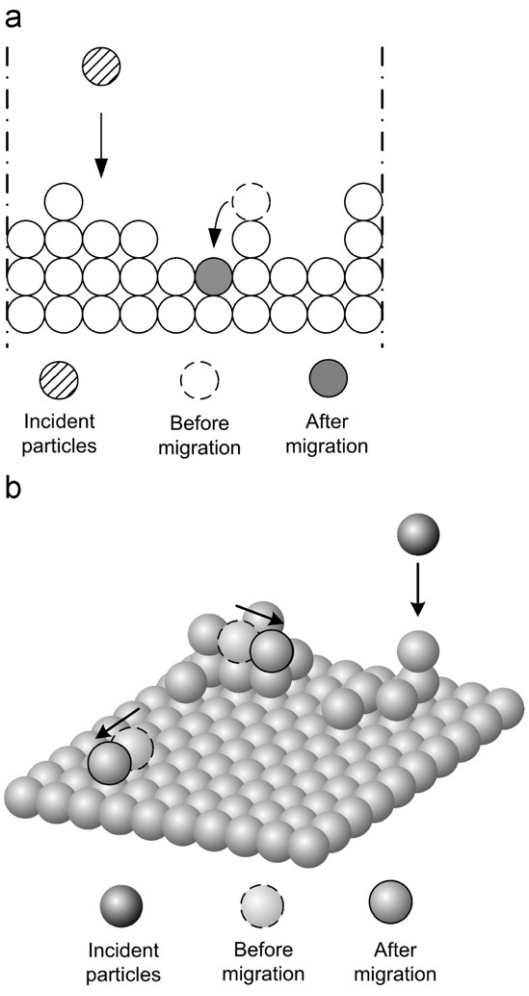

Fig. 1. Thin film deposition processes on (a) a 1D square lattice and (b) a 2D square lattice.

Stanley, 1995; Kardar, 2000). In the RDSR model, there is only one type of microscopic event: the random deposition with surface relaxation event. When a particle is deposited, a site is first randomly chosen among all lattice sites. After the site is determined, an incident particle deposits on the top of the surface particle on that site. Upon deposition, the deposited particle is subject to a surface relaxation event if any of the nearest neighbors of the site is lower than the initial deposition site. When the surface relaxation event is conducted, the deposited particle moves to the neighboring site with the lowest height among its nearest neighboring sites. For the case for which two or more neighboring sites have the same lowest heights, the deposited particle randomly chooses (with equal probability) a neighboring site as its final deposition site. When the lowest height of the nearest neighbors is only one layer lower than the center site after deposition, the deposited particle may stay in the original deposited site subject to the same probability to the one that this particle moves to the lowest nearest neighboring site. We note that the number of nearest neighboring sites varies with respect to the dimension of the lattice model. Specifically, there are two nearest neighboring sites in the 1D square lattice and the number of nearest neighboring sites is four in the 2D case.

In the RDSR model, there is only one macroscopic process parameter that characterizes the deposition process: the deposition rate, W, in units of deposited layers per second. Since random deposition is always followed by surface relaxation of the same deposited particle, the deposition rate, W, does not influence the balance between the deposition and relaxation events. A different W only scales the dynamics of the thin film evolution. Thus, the deposition rate is fixed at $W=1$ layer/s for the RDSR model in all simulations presented in this work.

2.2. Deposition/migration model

In the deposition/migration model (e.g., Ni and Christofides, 2005), the deposition and migration events are separate and independent microscopic events. The deposition event is a random process, i.e., the same random deposition (without surface relaxation) as in the RDSR model in Section 2.1. However, the migration event does not follow immediately the deposition of the particle. Instead, each surface particle, i.e., the top particle of a lattice site, is subject to its own migration event with a probability that depends on its local environment and the substrate temperature. The migration rate (probability) follows an Arrhenius-type law as follows:

$$r_{m,i} = v_0 \exp\left(-\frac{E_s + n_i E_n}{k_B T}\right), \tag{1}$$

where $r_{m,i}$ denotes the migration rate of the i-th surface particle, $v_0 = 2k_BT/h$ is a pre-exponential factor, $n_i=0, 1, 2, \ldots$, is the number of the nearest neighbors in the same layer of the surface particle on the i-th lattice site, E_s and E_n are the contribution to the activation energy barrier from the surface site and from each nearest neighbor, respectively, k_B is Boltzmann's constant, h is Planck's constant, and T is the substrate temperature.

When a surface particle is subject to migration, the particle moves onto a neighboring site with a lower surface height. If two or more neighboring sites have lower height than the height of the initial particle site, the migrating particle randomly moves to one of these neighboring sites with equal probability. We note that when n_i equals the number of nearest neighboring sites (i.e., two for the 1D lattice and four for the 2D lattice), the particle is fully surrounded by other particles and cannot move. Multi-step moves are not included in this deposition/migration model but can be realized via several successive but separate migration events of the same particle.

In the deposition/migration model, the macroscopic process parameters include the deposition rate, W, and the substrate temperature, T. These two process parameters together determine the growth conditions of the thin film as well as its surface morphology. As in the RDSR model in Section 2.1, the deposition rate is fixed as $W=1$ layer/s for the deposition/migration model. The substrate temperature may be varied at different values.

3. Surface roughness

Surface roughness is commonly used to describe the irregularity of surface morphology and measures the vertical deviation of the surface from an ideal, flat surface. In this work, surface roughness is defined as the root-mean-square (rms) of the surface height profile, which is the connection of the centers of the surface particles on all lattice sites. The definition of surface roughness is given as follows:

$$r = \left[\frac{1}{L}\sum_{i=1}^{L}(h_i - \overline{h})^2\right]^{1/2}, \quad \text{1D},$$
$$r = \left[\frac{1}{L^2}\sum_{i=1}^{L}\sum_{j=1}^{L}(h_{i,j} - \overline{h})^2\right]^{1/2}, \quad \text{2D}, \tag{2}$$

where r denotes surface roughness, h_i ($h_{i,j}$), $i=1, 2, \ldots, L$, is the surface height at the i-th (i,j-th) position in the unit of layer, L denotes the lattice size, and $\overline{h} = \frac{1}{L}\sum_{i=1}^{L} h_i$ is the average surface height defined as follows:

$$\overline{h} = \frac{1}{L}\sum_{i=1}^{L} h_i, \quad \text{1D},$$
$$\overline{h} = \frac{1}{L^2}\sum_{i=1}^{L}\sum_{j=1}^{L} h_{i,j}, \quad \text{2D}. \tag{3}$$

To investigate the scaling properties of surface roughness, we perform a series of kMC simulations for the two deposition models for different lattice sizes. Both 1D and 2D lattices are investigated. The surface roughness is computed on the basis of the surface height profile that is obtained from the kMC simulation at each sampling time. For the convenience of observing the lattice size dependence, the roughness square, r^2, is used to express the results. Since the deposition process is a stochastic process in nature, multiple independent kMC simulations (1000–30,000, depending on the level of fluctuations) under the same operating conditions are repeated to generate the expected value of the roughness square. The range of lattice size is chosen as large as possible under the limitation of currently available computing power and the requirement to compute accurate values of expected film surface properties. In this work, all deposition processes start with flat initial surface height profiles.

Remark 1. For each specific point we include in the results, we run a sufficiently large number of simulations, so that the computed expected value does not change with the addition of more simulations for that specific condition. For example, for an expected value that is computed using 10,000 runs, the addition of more runs will not change the location of the point at all; this has been checked for all points. For this reason, we use different number of runs for different conditions because some temperature/deposition rate conditions or variables (roughness/slope) require more runs to get an accurate (one that does not change with the addition of more simulations) expected value.

With respect to the computation of the errors of the computed mean values, we follow the following procedure: we group the simulations for a specific condition into 10 different groups (for example, in the case of running 10,000 simulations, we form 10 groups of 1000 simulations each) and compute the mean of each group first and then the standard deviation of these 10 means which is equal to the size of the error bar. This exact procedure is repeated for all points included in the results.

3.1. Random deposition with surface relaxation model

In the RDSR model of the deposition process, the deposition rate is fixed at $W=1$ layer/s for all simulations. The lattice size ranges from 20 to 200 sites. Fig. 2 shows the evolution profiles of the expected surface roughness square. All profiles in Fig. 2 start from zero, since the surface is assumed to be flat at the beginning of the deposition process. At the initial stages of deposition (the time is sufficiently small), all roughness profiles evolve similarly. As time increases, the roughness profiles in Fig. 2 increase and approach their respective steady-state values. It is evident from Fig. 2 that the lattice size strongly influences the dynamic behavior of surface roughness. The roughness square of the RDSR model with a larger lattice size takes longer time to reach a higher steady-state value. Thus, the roughness profiles for $L=150$ and 200 require a longer time frame ($\sim$2000 s) to reach their respective steady-states.

To further investigate the scaling properties of surface roughness of the deposition process, we examine the behavior of surface roughness square v.s. lattice size. Fig. 3 shows the L-dependent expected steady-state values of surface roughness square, $\langle r^2 \rangle_{ss}$, with respect to the lattice size. A clear linear dependence on the lattice size is observed in Fig. 3. This linear lattice-size dependence is consistent with the dynamic equation, the Edwards–Wilkinson (EW) equation, of the RDSR model (Kardar, 2000; Hu et al., 2009a).

Subsequently, we study the scaling properties of the 2D RDSR model. Figs. 4 and 5 show the profiles of the expected roughness square and the lattice-size dependence of the expected steady-state values of the roughness square of the 2D RDSR model. The evolution profiles in Fig. 4 have similar dynamics as the ones in the 1D system shown in Fig. 2. This similarity is because the surface height dynamic behavior of the RDSR model belongs to the EW universality class, irrespective of the dimension of the

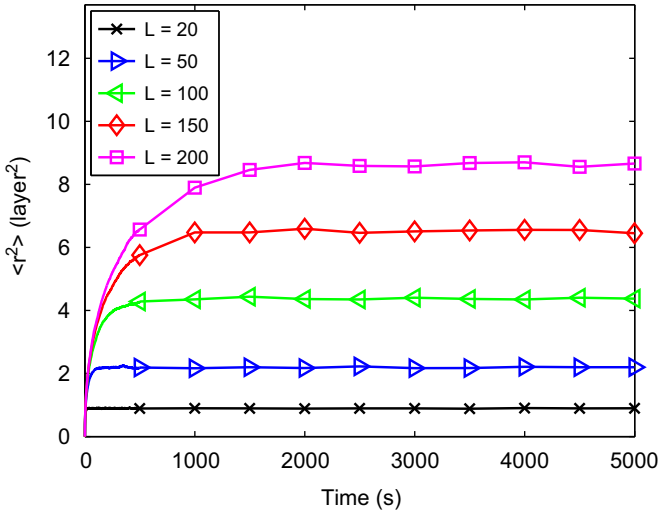

Fig. 2. Profiles of the expected surface roughness square at different lattice sizes, $L=20$, 50, 100, 150, and 200; 1D RDSR model with $W=1$ layer/s.

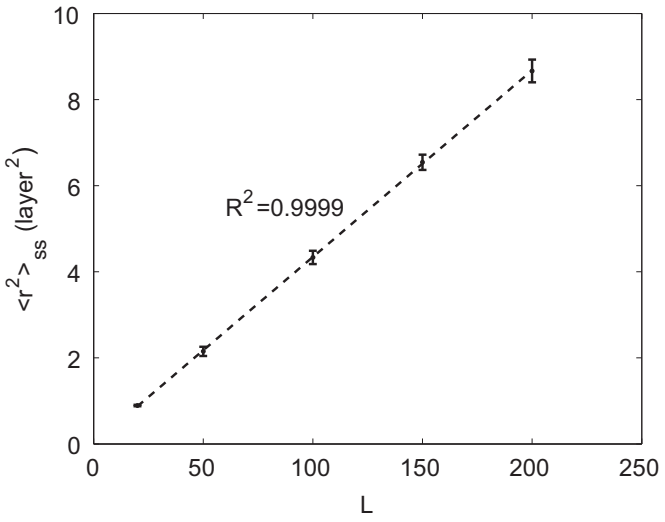

Fig. 3. Dependence of the expected steady-state value of surface roughness square on the lattice size; 1D RDSR model with $W=1$ layer/s.

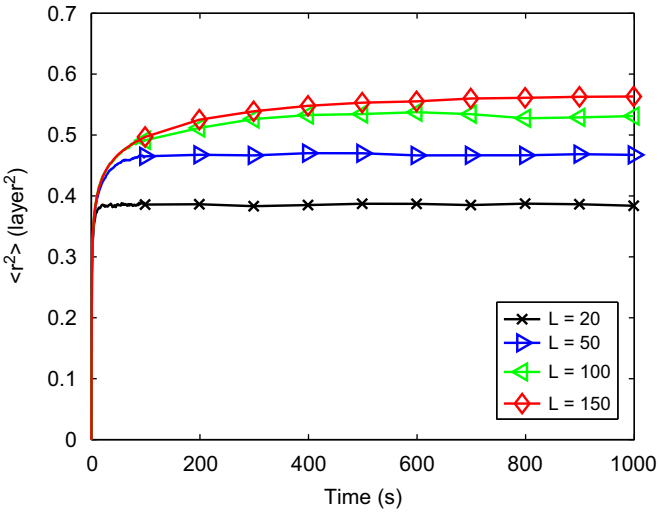

Fig. 4. Profiles of the expected surface roughness square at different lattice sizes; 2D RDSR model with $W=1$ layer/s.

model. A logarithmic dependence on the lattice size can be seen in Fig. 5. A semi-log plot is used in Fig. 5 with a least-square-fitted line to show clearly the logarithmic dependence. The logarithmic lattice-size dependence of roughness square is consistent with the EW dynamics, from which a dependence of zeroth order on lattice size can be derived (which is consistent with logarithmic dependence) (Kardar, 2000; Horowitz and Albano, 2003).

However, by comparing Figs. 2 and 4, it can be seen that there is a difference in the dynamics between the 1D and 2D RDSR models. The roughness square in the 2D RDSR model reaches a lower steady-state value at a shorter time, given the same lattice size. The different dynamic behavior of the 2D RDSR model is due to the extra dimension for the surface relaxation, i.e., the deposited particles have more freedom of migrating in the 2D model than in the 1D model. Thus, it takes less time for the surface roughness in the 2D model to reach a steady-state, which can be thought of as an equilibrium state between the deposition and migration events. Furthermore, the extra dimension for the surface relaxation also leads to different lattice-size dependence of surface roughness; a linear dependence in the 1D RDSR model

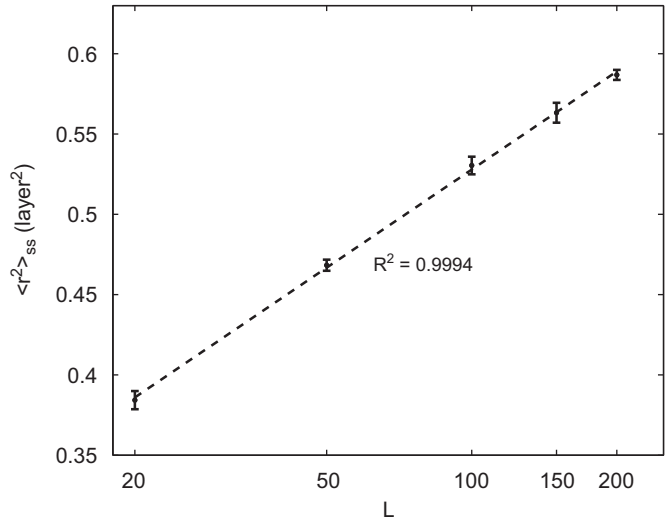

Fig. 5. Dependence of the expected steady-state value of surface roughness square on the lattice size; 2D RDSR model with $W=1$ layer/s.

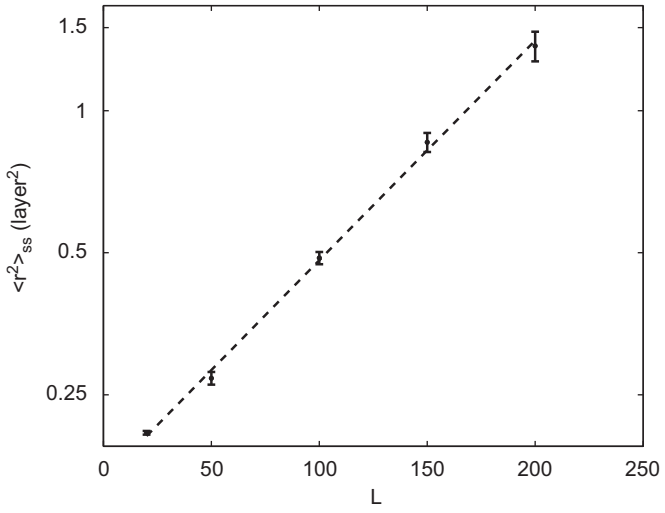

Fig. 6. Dependence of the expected steady-state value of surface roughness square on the lattice size; 1D deposition/migration model with $E_n=0.6$ eV, $T=680$ K, $W=1$ layer/s.

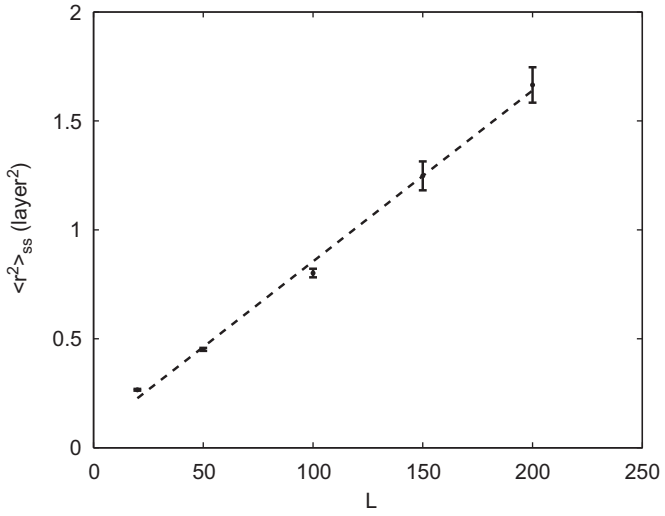

Fig. 7. Dependence of the expected steady-state value of surface roughness square on the lattice size; 2D deposition/migration model with $E_n=0.6$ eV, $T=650$ K, $W=1$ layer/s.

and a logarithmic dependence in the 2D case, as shown in Figs. 3 and 5.

3.2. Deposition/migration model

In the RDSR model, the ratio between the particle deposition and relaxation rates is fixed and the particles after the deposition/relaxation process cannot move. However, in the deposition/migration model, the deposition and migration events are independent and the particles on the surface are subject to migration unless fully surrounded by nearest neighbors. The thin film surface morphology is the result of a complex interplay between adsorption and migration events. Thus, the surface roughness of the deposition/migration model may have different dynamic behavior, i.e., time of approaching the steady-state value, from the one of the surface roughness in the RDSR model. To carry out the kMC simulations of the deposition/migration model, the values of the activation energy barriers are chosen to be consistent with silicon thin films (Hu et al., 2009a) in the 2D lattice model and are taken as follows: $E_s=1.2$ eV and $E_n=0.6$ eV. The operating conditions for the deposition/migration model are chosen, so that the resulting surface roughness under these operating conditions is close to the one in the RDSR model; $T=680$ K and $W=1$ layer/s for the 1D model and $T=650$ K and $W=1$ layer/s for the 2D model.

In the deposition/migration model, the dynamic behavior of surface roughness square is similar to the one in the RDSR model; the profile of roughness square increases from zero and approaches a steady-state value at large times. However, the scaling properties of roughness are different in the two deposition process models. Figs. 6 and 7 show the lattice-size dependence of $\langle r^2\rangle_{ss}$ in the 1D and 2D deposition/migration process models with $E_n=0.6$ eV, respectively. By comparing to the dependence of $\langle r^2\rangle_{ss}$ in the RDSR models in Figs. 3 and 5, both 1D and 2D deposition/migration models have a stronger roughness dependence on the lattice size. The 1D lattice-size dependence is quasi-exponential, $\langle r^2\rangle_{ss}\sim\exp(L)$ (Fig. 6, please note the scale in the vertical axis is the logarithmic scale); while the 2D dependence is quasi-linear, $\langle r^2\rangle_{ss}\sim O(L)$ (Fig. 7).

This stronger dependence of surface roughness on the lattice size has a correlation with a larger difference of the rates of the various migration possibilities of the surface particles. Here the migration possibilities refer to the dimension of the lattice and the classifications of surface particles with respect to the number of neighboring particles (particles belonging in different classes are associated with different migration rates when $E_n\neq 0$). In the 1D lattice, the migration of surface particles is limited to one direction; while in the 2D lattice, the surface particles have an extra dimension to migrate. Thus, the roughness dependence on the lattice size in the 1D model is stronger (linear) than in the 2D model (logarithmic). Similarly, in the deposition/migration model with a non-zero E_n, the surface particles are classified according to the number of nearest neighbors, n_i, in Eq. (1). The particles in different classes have different migration rates; a larger migration rate for the class with a smaller n_i. Thus, the surface particles with less nearest neighbors are more likely to migrate than the particles with more nearest neighbors. However, in the RDSR model, all surface particles have the same migration probabilities, since the relaxation event is executed instantaneously following a random deposition. This difference of the migration possibilities (classification of migration particles) in the deposition/migration model results in a stronger lattice-size dependence than in the RDSR model.

To further support the correlation between the scaling properties and the difference of the migration possibilities, kMC simulations are carried out for the deposition/migration model with $E_n=0$ eV. In the deposition/migration model, a zero E_n indicates no additional energy barrier from each nearest neighbor, and thus, all particles have the same migration rates. From a physical significance point of view, $E_n=0$ implies that the rate of migration of each surface particle is only dependent on temperature and the activation energy E_s of the surface site and it is independent of the number of nearest neighbors of the particle; this scenario is appropriate in the case where the nearest neighbor interactions of the surface particle are very weak relative to the bonding of the particle with its site. Lower substrate temperatures ($T=480$ K for the 1D model and $T=460$ K for the 2D model) are selected for the operating conditions due to the lower total activation energy barriers compared to the deposition/migration model with $E_n=0.6$ eV. Figs. 8 and 9 show the lattice-size dependence of $\langle r^2\rangle_{ss}$ in the 1D and 2D deposition/migration models with $E_n=0$ eV, respectively.

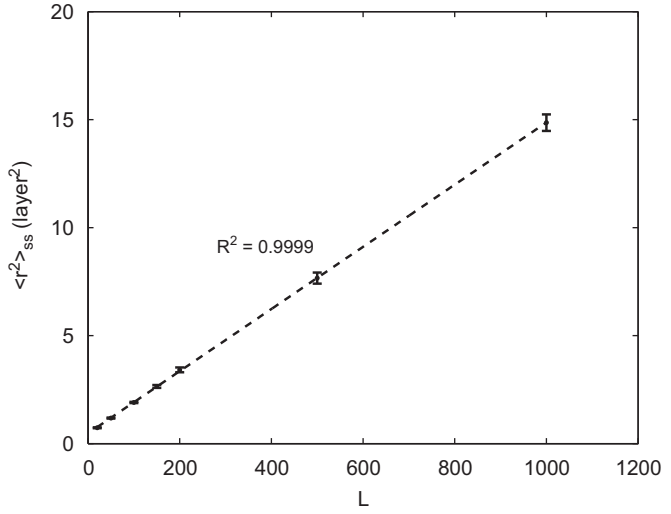

Fig. 8. Dependence of the expected steady-state value of surface roughness square on the lattice size; 1D deposition/migration model with $E_n=0$ eV, $T=480$ K, $W=1$ layer/s.

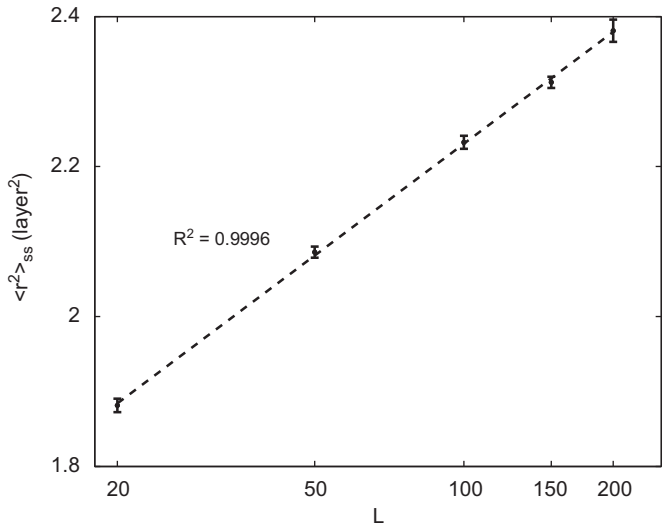

Fig. 9. Dependence of the expected steady-state value of surface roughness square on the lattice size; 2D deposition/migration model with $E_n=0$ eV, $T=460$ K, $W=1$ layer/s.

The dependence of $\langle r^2\rangle_{ss}\sim\exp(L)$ in the deposition/migration model with zero E_n on the lattice size, shown in Figs. 8 and 9, is consistent with the dependence found in the RDSR model; both models have linear lattice-size dependence in the 1D lattice and logarithmic lattice-size dependence in the 2D lattice. The close relationship of the surface roughness dependence on the lattice size for the RDSR model and the deposition/migration model with $E_n=0$ is expected, since the surface relaxation model has a migration step that is always performed if the neighboring sites have lower height, and this is also achieved in the deposition/migration model with E_n being 0. Furthermore, the roughness is higher in the deposition/migration model v.s. the RDSR model because the relaxation step takes place in every deposition event in the RDSR model.

Remark 2. Regression coefficients R^2 were only added to the plots where linear dependence can be proved analytically (using closed-form equations (stochastic PDEs) of the surface evolution profile) to hold for any lattice-size range (Figs. 3, 5, 8 and 9). Specifically, in previous works (e.g., Barabási and Stanley, 1995; Hu et al., 2009a), the linear dependence of $\langle r^2\rangle_{ss}$ on lattice size L for RDSR models has been proved analytically for any lattice-size range, so in the present paper regression coefficients were added in all related plots (Figs. 3 and 5) to support this conclusion. In addition, regression coefficients were also added to Figs. 8 and 9 to show that the scaling properties of the deposition/migration model with $E_n=0$ are similar to the ones of the RDSR model. For all the other plots, although quasi-linear or quasi-exponential dependences were observed locally (for the range of lattice size used in the simulations) from the results, there is no proof that these types of dependences hold globally (for any range of lattice size), so regression coefficients were not added.

4. Surface slope

Although rms roughness can be used to capture the height deviations of a thin film surface profile, neither surface slope nor surface height correlation between different surface locations can be captured by the rms roughness. Under certain assumptions for surface height distribution functions, these additional surface characteristics can be simplified to the quantities of surface mean slope and auto-covariance length; both of which can be connected to rms roughness via a simple equation (Davies, 1954). In this work, the surface mean slope is defined in a fashion similar to surface roughness as follows:

$$m=\left[\frac{1}{L}\sum_{i=1}^{L}h_{s,i}^2\right]^{1/2},\quad \text{1D},$$
$$m=\left[\frac{1}{L^2}\sum_{i=1}^{L}\sum_{j=1}^{L}h_{s,i,j}^2\right]^{1/2},\quad \text{2D}, \tag{4}$$

where m denotes the rms slope and $h_{s,i}$ ($h_{s,i,j}$), is the surface slope at the i-th (i,j-th) lattice site, which is a dimensionless variable. The surface slope, $h_{s,i}$ ($h_{s,i,j}$), is computed as follows:

$$h_{s,i}=h_{i+1}-h_i,\quad \text{1D},$$
$$h_{s,i,j}=h_{i+1,j}-h_{i,j},\quad \text{2D}. \tag{5}$$

Due to the use of PBCs, the slope at the boundary lattice site ($i=L$) is computed as the slope between the last lattice site (h_L) and the first lattice site (h_1). It is necessary to point out that the surface slope in the 2D lattice is calculated as the slope in 1D. Due to the isotropy of the lattice model, the surface slope can be obtained in either dimension of the lattice, i.e., ($h_{i+1,j}-h_{i,j}$) or ($h_{i,j+1}-h_{i,j}$).

To be consistent with the roughness plots, the mean slope square, m^2, is used to present the results.

4.1. Random deposition with surface relaxation model

The evolution profiles of the expected mean slope square in the RDSR model, both 1D and 2D, are obtained from the same kMC simulation data for the roughness profiles in Section 3. Fig. 10 shows the profiles of the expected mean slope square at different lattice sizes in the 1D and 2D RDSR models. From Fig. 10, it can be seen that the mean slope square of the RDSR model has a smaller value and faster dynamics than the roughness square. The different dynamic behavior of the slope is because the correlation between the heights of two adjacent lattice sites in the slope definition in Eq. (4) is higher than the correlation between the heights of a lattice site and the average height in the roughness definition in Eq. (2) (Huang et al., in press).

The dependence of the mean slope square on the lattice size is also different from the dependence of the roughness square; see Fig. 11 for the lattice-size dependence of the mean slope square. The expected steady-state value of the mean slope square, $\langle m^2 \rangle_{ss}$, has a very weak dependence on the lattice size in both

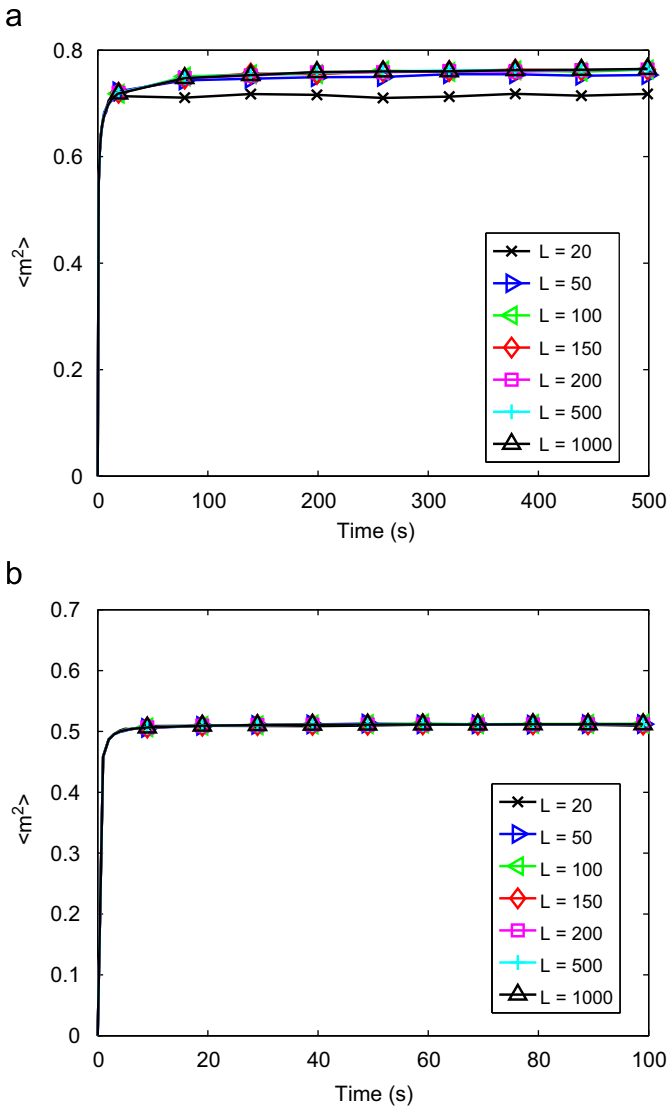

Fig. 10. Profiles of the expected mean slope square at different lattice sizes: (a) 1D RDSR model and (b) 2D RDSR model with $W=1$ layer/s.

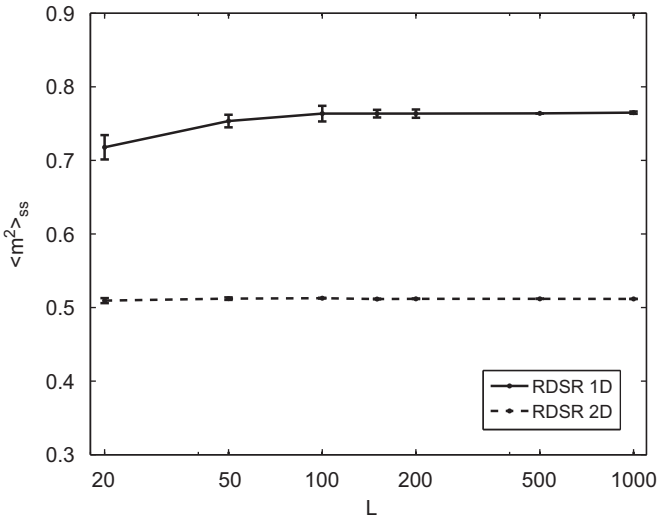

Fig. 11. Dependence of the expected steady-state value of mean slope square on the lattice size; 1D and 2D RDSR models with $W=1$ layer/s.

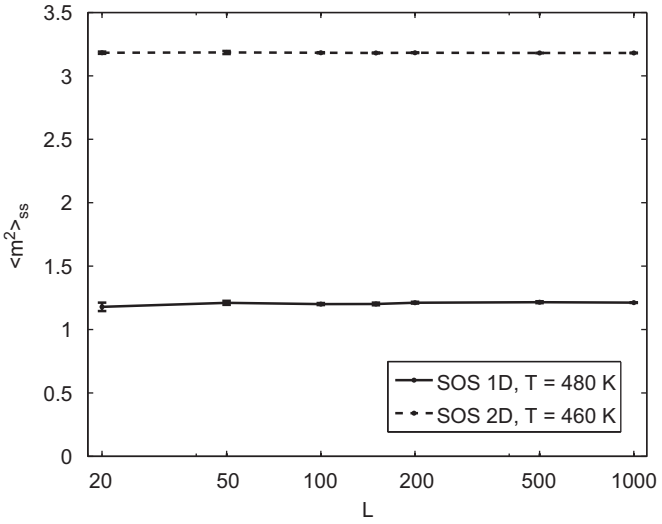

Fig. 12. Dependence of the expected steady-state value of mean slope square on the lattice size; 1D and 2D deposition/migration models with $E_n=0$ eV, $T=480$ K for the 1D model and $T=460$ K for the 2D model, and $W=1$ layer/s for both models.

1D and 2D RDSR models; $\langle m^2 \rangle_{ss}$ converges to a fixed value. To show the weak dependence, Fig. 11 is generated in a semi-log plot, which indicates that the dependence is weaker than logarithmic dependence.

4.2. Deposition/migration model

In the deposition/migration model, the weak lattice-size dependence of the mean slope square can be also observed. As shown in Section 3.2, the deposition/migration model with $E_n=0$ eV exhibits a consistent dynamic behavior and scaling property of surface roughness with the RDSR model. A similar weak dependence of the mean slope square on the lattice size is found in the deposition/migration model with $E_n=0$ eV; see Fig. 12.

However, in the deposition/migration model with non-zero E_n, the lattice-size dependence of the mean slope square is slightly stronger. Fig. 13 shows the dependence of $\langle m^2 \rangle_{ss}$ on the lattice size in the deposition/migration model with $E_n=0.6$ eV for both

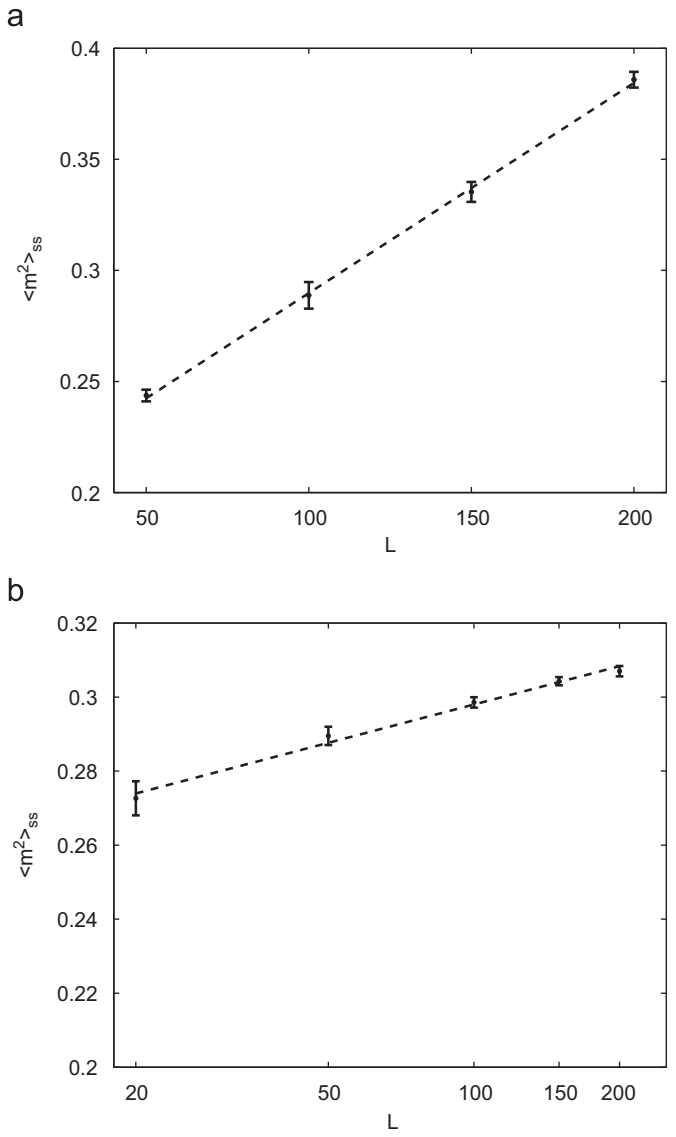

Fig. 13. Dependence of the expected steady-state value of mean slope square on the lattice size: (a) 1D deposition/migration model with E_n=0.6 eV, T=610 K, and W=1 layer/s and (b) 2D deposition/migration model with E_n=0.6 eV, T=650 K, and W=1 layer/s.

1D and 2D lattices. In the 1D deposition/migration model with non-zero E_n, the lattice-size dependence is quasi-linear; in the 2D model, the dependence is quasi-logarithmic. Overall, the mean slope square has a weaker dependence on the lattice size than the roughness square, especially at large lattice sizes.

5. Applications to light trapping efficiency

In this section, the influence of the surface morphology characteristics, i.e., the surface roughness and slope, on the light reflectance/transmittance is investigated in a light scattering (Rayleigh scattering) process. When the incident light goes through a rough interface, the light is divided into four components: specular reflection, specular transmission, diffused reflection and diffused transmission; see Fig. 14 (Tao and Zeman, 1994; Leblanc and Perrin, 1994). Under the assumptions of normal surface height distribution and correlation, the total reflectance of a beam of monochromatic light at normal incidence

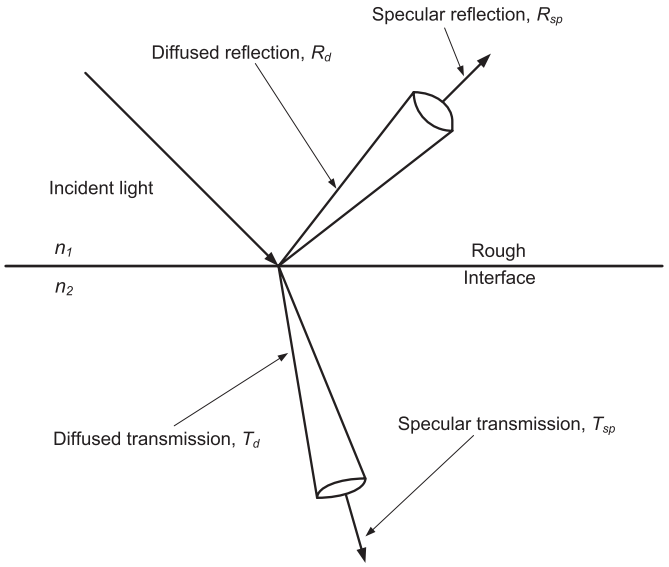

Fig. 14. Light scattering at a rough interface: specular reflection, R_{sp}, diffused reflection, R_d, specular transmission, T_{sp}, and diffused transmission, T_d. n_1 and n_2 are the refractive indices of the two substances above and below the rough interface, respectively.

to a rough surface, which is denoted by R, can be approximately calculated as follows (Davies, 1954):

$$R = R_0 \exp\left(-\frac{4\pi r^2}{\lambda^2}\right) + R_0 \int_0^{\pi/2} 2\pi^4 \left(\frac{a}{\lambda}\right)^2 \left(\frac{r}{\lambda}\right)^2 (\cos\theta+1)^4 \sin\theta \exp\left[-\frac{(\pi a \sin\theta)^2}{\lambda^2}\right] d\theta, \quad (6)$$

where R_0 is the reflectance of a perfectly smooth surface of the same material, θ is the incident angle, λ is the light wavelength, and a is the auto-covariance length of the interface, which can rewritten as a ratio between the rms roughness and the rms slope, $a=\sqrt{2}r/m$ (Bennett and Porteus, 1961). Eq. (6) shows that both r and m have a strong influence on the intensity of light reflection (and light transmission) at the surface/interface. The distributions of the four components of light reflectance and transmittance are also affected by r and m (Krč and Zeman, 2002, 2004) even though this dependence cannot be expressed by an approximate equation like the one of Eq. (6). For thin-film solar cells, specifically, a maximum or minimum of the light reflectance is desired for a certain surface/interface. For example, the top surface of the solar cell favors a minimum reflectance so as to absorb the incident light as much as possible. The objective of an optimal light trapping efficiency can be achieved by attaining certain values of r and m during the manufacturing of thin-film solar cells.

The mean surface slope investigated in this work is defined on the basis of the slope profile computed from the surface heights of the adjacent lattice sites; see Eq. (4). Thus, the characteristic length scale of the mean surface slope computed using Eq. (4) is the atomic/molecular diameter ($\sim$0.25 nm). However, the wavelength of visible light (400–700 nm) is much larger than the diameters of atoms/molecules; it may result in different surface slope under the larger length scale. Thus, it is necessary to compute the mean surface slope over the length scale of the light wavelength.

The aggregate surface slope, m_{Δ}, is computed similarly to the original mean surface slope of Eq. (4), but on the basis of the averaged surface height profile, $h_{\Delta,i}$, which is defined on the basis

of the aggregation of Δ surface sites as follows in 1D:

$$h_{\Delta,i} = (h_{i\Delta+1} + h_{i\Delta+2} + \cdots + h_{(i+1)\Delta})/\Delta, \quad i = 0,1,\ldots,L_\Delta - 1, \tag{7}$$

where $h_{\Delta,i}$ denotes the averaged surface height, Δ denotes the aggregation size, i.e., the number of lattice sites used to calculate the averaged surface height, and L_Δ denotes the number of aggregated surface heights. For the wavelength of visible light and silicon thin-film solar cells, the corresponding aggregation size, Δ, is around 2000. The computation of aggregated surface mean slope, m_Δ, is given as follows in 1D:

$$m_\Delta = \left[\frac{1}{L}\sum_{i=1}^{L_\Delta}\left(\frac{h_{\Delta,i} - h_{\Delta,i+1}}{\Delta}\right)^2\right]^{1/2}. \tag{8}$$

The dynamics of the aggregate surface slope is dependent on the characteristic length scale, Δ. To show this dependence, kMC simulations of the 1D deposition/migration model with $E_n = 0$ eV and $L = 10{,}000$ are carried out at $T = 430$ K and $W = 1$ layer/s. The mean slope square, m_Δ^2, is calculated from the surface height profile from the kMC simulations at different length scales. Fig. 15 shows the profiles of the expected mean slope square, $\langle m_\Delta^2 \rangle$ under different characteristic length scales, Δ. It is evident from Fig. 15 that the larger the characteristic length scale, the smaller the mean slope square and the slower the dynamics of evolution (i.e., the longer the time to reach the steady-state value).

Furthermore, Fig. 16 shows the dependence of the steady-state value of the expected mean slope square, $\langle m_\Delta^2 \rangle_{ss}$, on the characteristic length scale, Δ. This dependence is quasi-quadratic on $1/\Delta$, i.e., $\langle m_\Delta^2 \rangle_{ss} \sim 1/\Delta^2$. Following this dependence, the corresponding mean slope square for Δ around 2000 is very small ($\langle m_\Delta^2 \rangle_{ss} \sim 10^{-5}$). This close-to-zero value of the mean slope square reveals a smoothly changing surface profile with respect to a relatively large characteristic length scale. The smoothness of the surface profile persists at larger lattice sizes as well, due to the very weak lattice-size dependence of the mean slope square. This small mean slope square under larger length scales is partly because the operating conditions are spatially uniform during the entire deposition process, i.e., the same deposition rate or substrate temperature is applied throughout the spatial domain. Thus, spatially distributed operating conditions (implemented via spatially distributed control actuators) are necessary for the purpose of optimizing thin film light reflectance/transmittance by

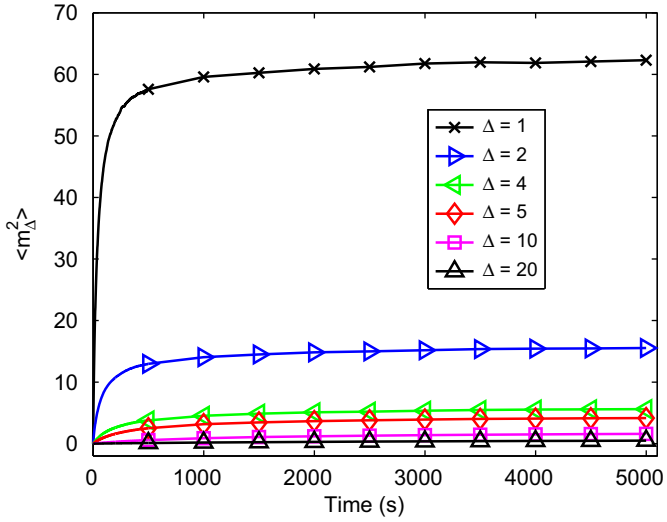

Fig. 15. Profiles of the expected mean slope square under different characteristic length scales; 1D deposition/migration model with $E_n = 0$ eV, $T = 430$ K, and $W = 1$ layer/s.

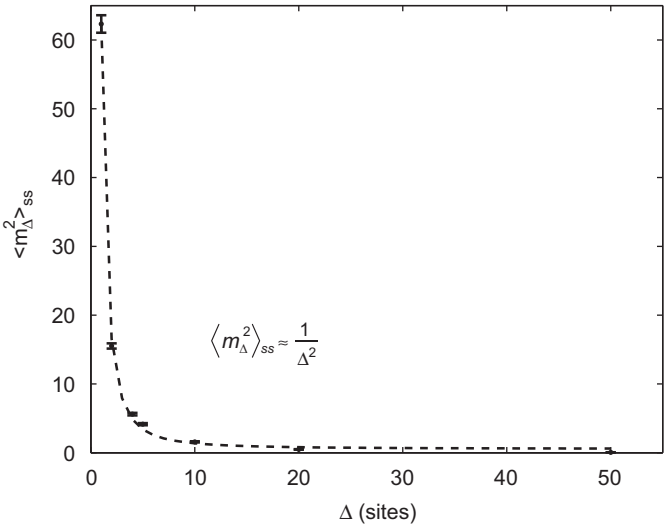

Fig. 16. Dependence of the steady-state value of the expected mean slope square on the characteristic length scale (symbols) and the fitted quadratic dependence on the characteristic length scale (dashed line); 1D deposition/migration model with $E_n = 0$ eV, $T = 430$ K, and $W = 1$ layer/s.

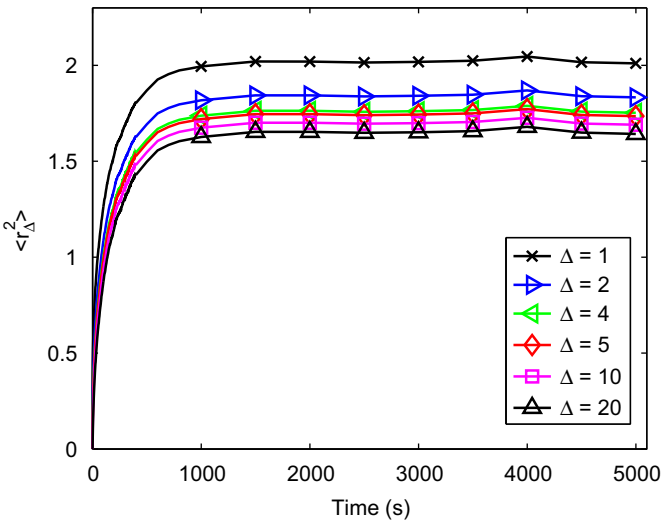

Fig. 17. Profiles of the expected surface roughness square under different characteristic length scales; 1D deposition/migration model with $E_n = 0$ eV, $T = 500$ K, and $W = 1$ layer/s.

manipulation of film surface roughness and slope at length scales relevant to visible light wavelength.

On the other hand, the surface roughness behaves differently from the surface slope at large length scales. Specifically, the new surface roughness, r_Δ, is computed as the surface roughness in Eq. (2) but on the basis of the average surface height profile computed from Eq. (7). The surface roughness has a weaker dependence on the characteristic length scale, Δ, than the surface slope. To show this weak dependence, kMC simulations of the 1D deposition/migration model with $E_n = 0$ eV are carried out at $T = 500$ K and $W = 1$ layer/s. A smaller lattice size, $L = 500$, is used in the kMC simulations because it takes much longer time for the surface roughness square than the mean slope square to reach the steady-state value at larger lattice sizes. Fig. 17 shows the profiles of the expected surface roughness square, $\langle r_\Delta^2 \rangle$, under different characteristic length scales, Δ. It can be seen that the surface roughness square attains smaller steady-state values for larger

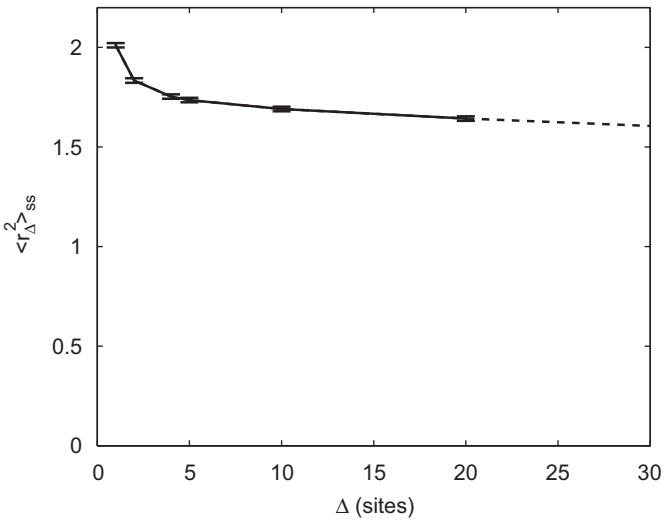

Fig. 18. Dependence of the steady-state value of the expected surface roughness square on the characteristic length scale (symbols); 1D deposition/migration model with $E_n=0$ eV, $T=500$ K, and $W=1$ layer/s.

length scales. However, the surface roughness square does not drop as prominently as the mean slope square when the characteristic length scale increases. This weak dependence of the steady-state value of the expected surface roughness square, $\langle r_\Delta^2 \rangle_{ss}$, on the characteristic length scale, Δ, is also shown in Fig. 18. The weak dependence on the length scale indicates a different behavior of surface roughness: the high frequency components in the surface height profile (i.e., local ripples below the characteristic length scale) contribute less significantly to the surface roughness than to the surface slope. Therefore, smooth surface height profiles with respect to large length scales have a very small mean slope square but considerable surface roughness square. Due to the insensitivity of the surface roughness to the high frequency components, spatially distributed control actuators may have less influence on the surface roughness while achieving the desired surface slope under certain characteristic length scales. This decoupled relationship between the surface roughness and slope can be utilized in the controller design for improving the light trapping efficiency of thin-film solar cells. This problem will be studied in future research work.

The results of this section also suggest that given a monochromatic light of wavelength λ, the light trapping efficiency of a thin film should be computed using surface roughness and slope values corresponding to an aggregate length scale which is on the order of λ; this implies that the lattice size to be used to carry out this simulation should be at least two orders of magnitude larger than the one corresponding to a length λ to minimize boundary effects (i.e., the use of periodic boundary conditions) on the computed properties.

Finally, we note that while the results for aggregate roughness and slope in Figs. 16 and 18 have been computed for $E_n=0$ (which corresponds to the case where the migration rates of all surface particles are the same), the reduction of the aggregate roughness and slope with increasing aggregation length will continue to hold for non-zero E_n value; however, the exact shape of the curves will depend on the specific E_n values.

6. Conclusions

This work focused on the study of the lattice-size dependence and dynamic behavior of thin film surface roughness and slope under different migration rates, lattice sizes and characteristic length scales. Two thin film deposition process models were used in this study: the RDSR model and a deposition/migration model. Both surface roughness and slope evolve to steady-state values at large times but are subject to different dynamics and scaling properties. A linear and a logarithmic dependence of surface roughness square on the lattice size were observed in the 1D and 2D lattice models, respectively, of the RDSR model and of the deposition/migration model with zero energy barrier contribution from neighboring particles. Furthermore, a stronger lattice-size dependence was found in the deposition/migration model with a significant contribution to the energy barrier from neighboring particles. This finding suggests that preferential migration (i.e., surface particles with zero or one nearest neighbors dominate the migration events) results in a stronger dependence of surface roughness on the lattice size. Contrary, a weaker lattice-size dependence was found for the surface mean slope, especially at large lattice sizes. Finally, the necessity of spatially distributed control action was demonstrated for the purpose of roughness and slope control at large characteristic length scales.

Acknowledgement

Financial support from NSF, CBET-0652131, is gratefully acknowledged.

References

Barabási, A.-L., Stanley, H.E., 1995. Fractal Concepts in Surface Growth. Cambridge University Press, New York.
Bennett, H.E., Porteus, J.O., 1961. Relation between surface roughness and specular reflectance at normal incidence. Journal of the Optical Society of America 51, 123–129.
Chou, Y.-L., Pleimling, M., 2009. Parameter-free scaling relation for nonequilibrium growth processes. Physical Review E 79, 051605.
Chou, Y.-L., Pleimling, M., Zia, R.K.P., 2009. Changing growth conditions during surface growth. Physical Review E 80, 061602.
Christofides, P.D., Armaou, A., Lou, Y., Varshney, A., 2008. Control and Optimization of Multiscale Process Systems. Birkhäuser, Boston.
Davies, H., 1954. The reflection of electromagnetic waves from a rough surface. Proceedings of the Institution of Electrical Engineers 101, 209.
Family, F., 1986. Scaling of rough surfaces: effects of surface diffusion. Journal of Physics A: Mathematical and General 19, L441–L446.
Haselwandter, C.A., Vvedensky, D.D., 2006. Stochastic equation for the morphological evolution of heteroepitaxial thin films. Physical Review B 74, 121408.
Haselwandter, C.A., Vvedensky, D.D., 2007. Renormalization of stochastic lattice models: basic formulation. Physical Review E 76, 041115.
Haselwandter, C.A., Vvedensky, D.D., 2008. Renormalization of stochastic lattice models: epitaxial surfaces. Physical Review E 77, 061129.
Horowitz, C.M., Albano, E.V., 2003. Relationships between a microscopic parameter and the stochastic equations for interfaces evolution of two growth models. The European Physical Journal B 31, 563–569.
Hu, G., Huang, J., Orkoulas, G., Christofides, P.D., 2009a. Investigation of film surface roughness and porosity dependence on lattice size in a porous thin film deposition process. Physical Review E 80, 041122.
Hu, G., Lou, Y., Christofides, P.D., 2008. Dynamic output feedback covariance control of stochastic dissipative partial differential equations. Chemical Engineering Science 63, 4531–4542.
Hu, G., Orkoulas, G., Christofides, P.D., 2009b. Modeling and control of film porosity in thin film deposition. Chemical Engineering Science 64, 3668–3682.
Hu, G., Orkoulas, G., Christofides, P.D., 2009c. Regulation of film thickness, surface roughness and porosity in thin film growth using deposition rate. Chemical Engineering Science 64, 3903–3913.
Hu, G., Orkoulas, G., Christofides, P.D., 2009d. Stochastic modeling and simultaneous regulation of surface roughness and porosity in thin film deposition. Industrial & Engineering Chemistry Research 48, 6690–6700.
Huang, J., Hu, G., Orkoulas, G., Christofides, P.D., in press. Dynamics and lattice-size dependence of surface mean slope in thin film deposition. Industrial & Engineering Chemistry Research, doi:10.1021/ie10012w.
Kardar, M., 2000. Roughness and ordering of growing films. Physica A 281, 295–310.
Krč, J., Smole, F., Topič, M., 2003. Analysis of light scattering in amorphous Si:H solar cells by a one-dimensional semi-coherent optical model. Progress in Photovoltaics: Research and Applications 11, 15–26.
Krč, J., Zeman, M., 2002. Experimental investigation and modelling of light scattering in a-Si:H solar cells deposited on glass/ZnO:Al substrates. Material Research Society 715, A13.3.1–A13.3.6.
Krč, J., Zeman, M., 2004. Optical modeling of thin-film silicon solar cells deposited on textured substrates. Thin Solid Films 451, 298–302.

Leblanc, F., Perrin, J., 1994. Numerical modeling of the optical properties of hydrogenated amorphous-silicon-based p-i-n solar cells deposited on rough transparent conducting oxide substrates. Journal of Applied Physics 75, 1074.

Levine, S.W., Clancy, P., 2000. A simple model for the growth of polycrystalline Si using the kinetic Monte Carlo simulation. Modelling and Simulation in Materials Science and Engineering 8, 751–762.

Levine, S.W., Engstrom, J.R., Clancy, P., 1998. A kinetic Monte Carlo study of the growth of Si on Si(1 0 0) at varying angles of incident deposition. Surface Science 401, 112–123.

Müller, J., Rech, B., Springer, J., Vanecek, M., 2004. TCO and light trapping in silicon thin film solar cells. Solar Energy 77, 917–930.

Ni, D., Christofides, P.D., 2005. Multivariable predictive control of thin film deposition using a stochastic PDE model. Industrial & Engineering Chemistry Research 44, 2416–2427.

Poruba, A., Fejfar, A., 2000. Optical absorption and light scattering in microcrystalline silicon thin films and solar cells. Journal of Applied Physics 88, 148–160.

Rowlands, S.F., Livingstone, J., Lund, C.P., 2004. Optical modelling of thin film solar cells with textured interfaces using the effective medium approximation. Solar Energy 76, 301–307.

Springer, J., Poruba, A., 2004. Improved three-dimensional optical model for thin-film silicon solar cells. Journal of Applied Physics 96, 5329–5337.

Tao, G., Zeman, M., 1994. Optical modeling of a-Si:H based solar cells on textured substrates. 1994 IEEE First World Conference on Photovoltaic Energy Conversion, Conference Record of the 24th IEEE Photovoltaic Specialists Conference-1994 (Cat.No.94CH3365-4), vol. 1, p. 666.

Vlachos, D.G., Schmidt, L.D., Aris, R., 1993. Kinetics of faceting of crystals in growth, etching, and equilibrium. Physical Review B 47, 4896–4909.

Zeman, M., Vanswaaij, R., 2000. Optical modeling of a-Si:H solar cells with rough interfaces: effect of back contact and interface roughness. Journal of Applied Physics 88, 6436–6443.

Zhang, P., Zheng, X., Wu, S., Liu, J., He, D., 2004. Kinetic Monte Carlo simulation of Cu thin film growth. Vacuum 72, 405–410.