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Dependence of Film Surface Roughness on Surface Migration and Lattice Size in Thin Film Deposition

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Abstract— **This work focuses on the study of the dependence of film surface roughness on surface migration and lattice size in thin film deposition processes. Two different models of thin film deposition processes, in both one-dimension and two-dimensions, are considered: random deposition with surface relaxation model and deposition/migration model. Surface roughness is defined as the root-mean-squares of the surface height profile and is found to evolve (starting from a flat initial surface zero value) to steady-state values at large times. A linear and a logarithmic dependence of surface roughness square on lattice size are observed in the one-dimensional and two-dimensional 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.**

I. 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 desired for a wider application of thin-film silicon solar cells. In this direction, 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 [13], [18]. Recent studies on enhancing thin-film solar cell performance [18], [20]–[22], [25] 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 [14], [15], [23]. Therefore, it is important to tailor thin film surface morphology characteristics to desired values.

In the context of modeling and control of thin film surface morphology, two modeling approaches have been developed and widely used: kinetic Monte Carlo (kMC) methods and stochastic differential equation (SDE) models. KMC methods were initially introduced to simulate thin film microscopic processes based on the microscopic rules. The corresponding thermodynamic and kinetic parameters are obtained from experiments and molecular dynamics simulations [2], [16], [17], [26]. Since kMC models are not available in closed form, 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., [3]–[5]) and numerical simulations (e.g., [2], [7], [8]). 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 [2], [7], film porosity [8], [10], and film thickness [9]. However, the influence of preferential migration on RMS roughness of surface height profiles in thin film deposition processes has not been studied.

Motivated by the above considerations, this work focuses on the study of the dependence of film surface roughness on the surface migration and lattice size in thin film deposition processes. Two different models of deposition processes are considered: a random deposition with surface relaxation model and a process model involving deposition and surface migration. Both models are constructed on a square lattice in both one-dimension and two-dimensions using the solid-on-solid assumption. Kinetic Monte Carlo methods are used to simulate both models. In the random deposition with surface relaxation model, a just-deposited particle is allowed to instantaneously relax to a neighboring site of lower height. The deposition/migration process model involves a random particle deposition event and a surface particle migration event. The probability of migration of a surface particle depends on the substrate temperature and the number of the neighboring particles. Each neighboring particle contributes equally to the activation energy of the migrating particle. Surface roughness is defined as the root-mean-square of the surface height profile. A linear dependence and a logarithmic dependence of surface roughness square on the lattice size are observed in the one-dimensional and two-dimensional 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 neighbor-

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ing particle. 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 in thin film deposition processes.

II. Description of thin film deposition processes

In this section, two different thin film deposition process models are introduced: 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 bounded by the periodic boundaries. In the two-dimensional lattice, the lattice sizes in both lateral directions are the same, i.e., the deposition process models take place on a square field in the two-dimensional 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 highest particles of the lattice sites, 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 [24] to simulate the thin deposition process models.

A. 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 [1], [11], [12]. 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 one-dimensional square lattice and the number of nearest neighboring sites is four in the two-dimensional case.

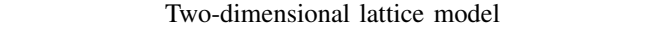

Fig. 1. Thin film deposition processes in the one-dimensional square lattice (upper plot) and the two dimensional square lattice (lower plot).

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.

B. Deposition/migration model

In the deposition/migration model [19], the deposition and migration events are separated into two independent microscopic events. The deposition event is a random deposition, i.e., the same random deposition (without surface relaxation) as in the RDSR model in Section II-A. 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:

$$r_{m,i} = \nu_0 \exp\left(-\frac{E_s + n_i E_n}{k_B T}\right), \tag{1}$$

where $r_{m,i}$ denotes the migration rate of the i-th surface particle, $\nu_0 = 2k_BT/h$ is a pre-exponential factor, $n_i = 0$, 1, 2, …, is the number of the nearest neighbors in the same layer of the surface particle on the ith 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 to a neighboring site with a lower surface height. If two or more neighboring sites are lower than the surface height, 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 and cannot move.

III. Surface roughness

Surface roughness is the most commonly used quantity to describe the 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:

$$\begin{aligned} r &= \left[\tfrac{1}{L}\textstyle\sum_{i=1}^{L}(h_i - \bar{h})^2\right]^{1/2}, && \text{one-dimension,} \\ r &= \left[\tfrac{1}{L^2}\textstyle\sum_{i=1}^{L}\sum_{j=1}^{L}(h_{i,j} - \bar{h})^2\right]^{1/2}, && \text{two-dimensions,} \end{aligned} \tag{2}$$

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 $\bar{h} = \frac{1}{L}\sum_{i=1}^{L} h_i$ is the average surface height defined as follows:

$$\begin{aligned} \bar{h} &= \tfrac{1}{L}\textstyle\sum_{i=1}^{L} h_i, && \text{one-dimension,} \\ \bar{h} &= \tfrac{1}{L^2}\textstyle\sum_{i=1}^{L}\sum_{j=1}^{L} h_{i,j}, && \text{two-dimensions.} \end{aligned} \tag{3}$$

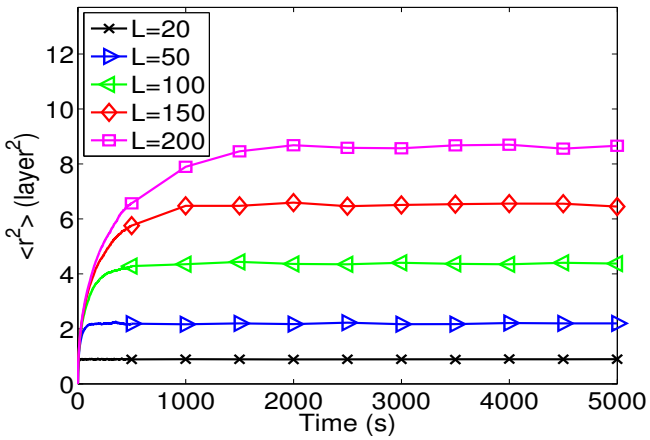

Fig. 2. Profiles of the expected surface roughness square at different lattice sizes, $L = 20$, 50, 100, 150, 200; 1D RDSR model with $W = 1$ layer/s.

To investigate the scaling properties of surface roughness, we carry out a series of kMC simulations for the two deposition models for different lattice sizes. Both one-dimensional and two-dimensional 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 and for comparing with the dynamic equation model, 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 (1,000–30,000, depending on the level of fluctuations) under the same operating conditions are repeated to generate the expected value of the roughness square. In this work, all deposition processes start with flat initial surface height profiles.

A. Random deposition with surface relaxation model

We first look at 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. 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 (the deposition duration is small), all roughness profiles evolve similarly. As time increases, the roughness profiles in Fig. 2 increase and approach their respective steady-state values at large times. 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 $L = 200$ require a longer time frame (2000 s) to reach their respective steady-states.

To further investigate the scaling properties of surface roughness of the deposition process, Fig. 3 shows the 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

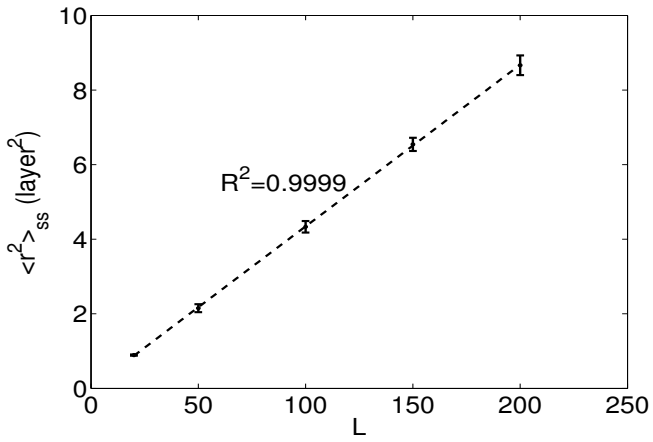

Fig. 3. Linear dependence of the expected steady-state value of surface roughness square on the lattice size; 1D RDSR model with $W = 1$ layer/s.

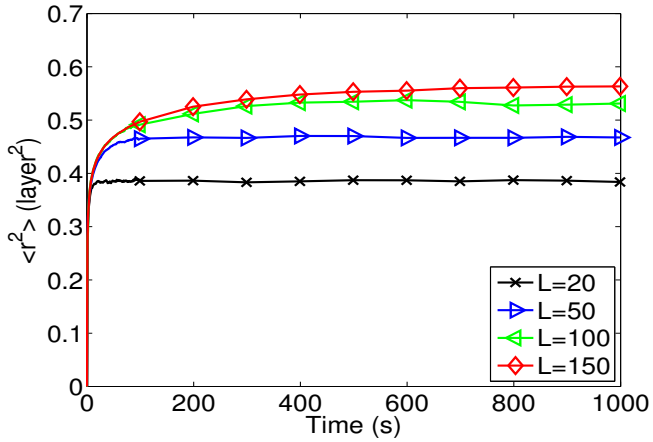

Fig. 4. Profiles of the expected surface roughness square at different lattice sizes; 2D RDSR model with $W = 1$ layer/s.

equation, the Edwards-Wilkinson equation, of the RDSR model [6].

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. By comparing Figs. 2 and 4, it can be seen that, although the roughness profiles evolve similarly in both 1D and 2D RDSR models, the roughness square in the 2D RDSR model has faster dynamics and lower steady-state values. 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 in the 2D model to reach a steady-state, which is the balance-point between the deposition and migration events.

Furthermore, the lattice-size dependence of surface roughness of the 2D RDSR model is different from the 1D model; as is shown in Fig. 5. A logarithmic dependence on the lattice

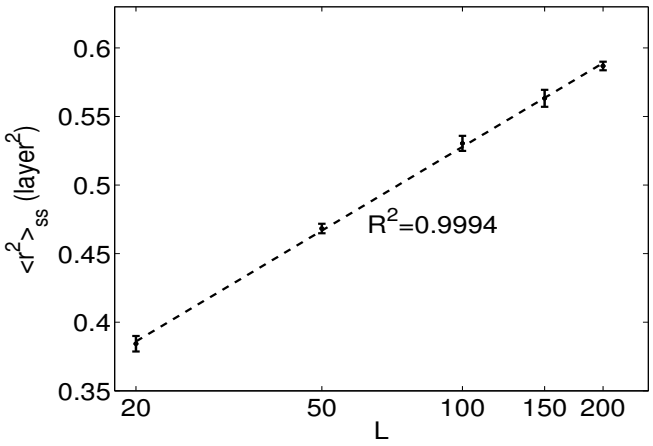

Fig. 5. Logarithmic dependence of the expected steady-state value of surface roughness square on the lattice size; 2D RDSR model with $W = 1$ layer/s.

size can be seen in Fig. 5. A semi-log plot is used in Fig. 5 with a fitted line to show clearly the logarithmic dependence.

B. 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 [6] in the two-dimensional 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, $\langle r^2 \rangle_{ss} \sim \exp(L)$; while the 2D dependence is quasi-linear, $\langle r^2 \rangle_{ss} \sim O(L)$.

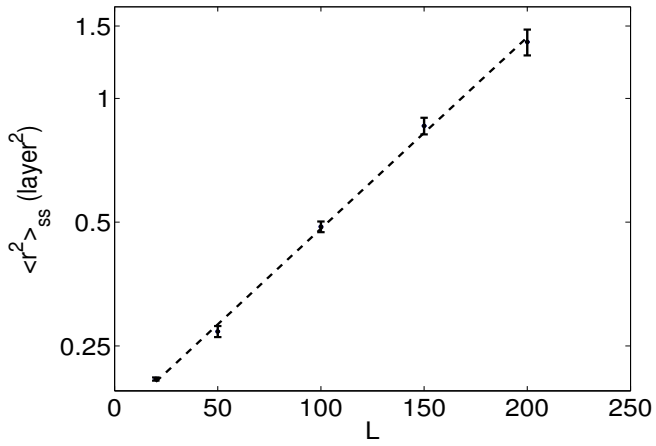

Fig. 6. Dependence of the expected steady-state value of surface roughness square on the lattice size; 1D deposition/migration model with $E_n = 0.6$ eV, $T = 680$ K, $W = 1$ layer/s.

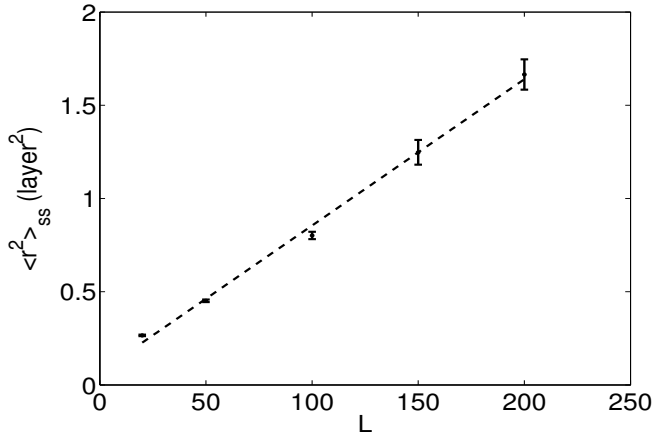

Fig. 7. Dependence of the expected steady-state value of surface roughness square on the lattice size; 2D deposition/migration model with $E_n = 0.6$ eV, $T = 650$ K, $W = 1$ layer/s.

This stronger dependence of surface roughness on the lattice size has a correlation with a larger difference of the 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.

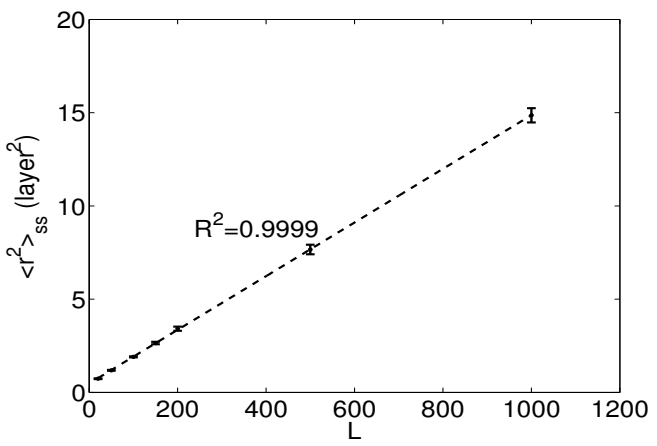

Fig. 8. Linear dependence of the expected steady-state value of surface roughness square on the lattice size; 1D deposition/migration model with $E_n = 0$ eV, $T = 480$ K, $W = 1$ layer/s.

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. 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. 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.

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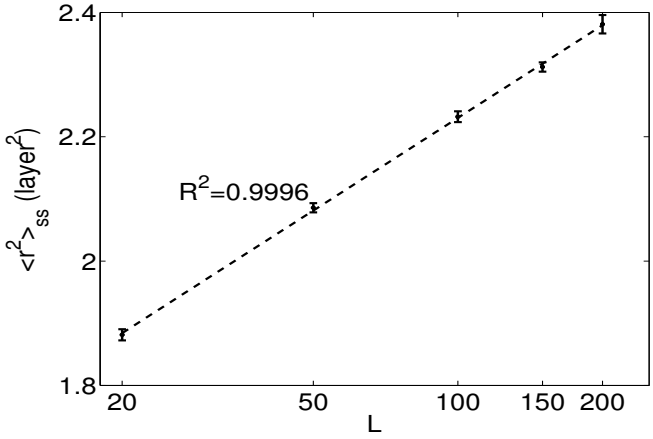

Fig. 9. Logarithmic dependence of the expected steady-state value of surface roughness square on the lattice size; 2D deposition/migration model with $E_n = 0$ eV, $T = 460$ K, $W = 1$ layer/s.

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