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Dynamic scaling in thin-film growth with irreversible step-edge attachment

F. D. A. Aarão Reis[*]
Instituto de Física, Universidade Federal Fluminense, Avenida Litorânea s/n, 24210-340 Niterói, RJ, Brazil

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We study dynamic scaling in a model with collective diffusion (CD) of isolated atoms in terraces and irreversible aggregation at step edges. Simulations are performed in two-dimensional substrates with several diffusion to deposition ratios $R\equiv D/F$. Data collapse of scaled roughness distributions confirms that this model is in the class of the fourth-order nonlinear growth equation by Villain, Lai, and Das Sarma (VLDS) with negligible finite-size effects, while estimates of scaling exponents show some discrepancies. This result is consistent with the prediction of a recent renormalization group approach and improves previous numerical works on related models. The roughness follows dynamic scaling as $W=L^{\alpha}/R^{1/2}f(\xi/L)$, with correlation length $\xi=(Rt)^{1/z}$, where z is the dynamic exponent. We also propose a limited mobility (LM) model where the incident atom executes up to G steps before a new atom is adsorbed, and irreversibly aggregates at step edges. This model is also shown to belong to the VLDS class. The size of the plateaus in the film surface increases as $G^{1/2}$ and the lateral correlation scales as $G^{1/2}t^{1/z}$. The time evolution of the roughness reproduces that of the CD model if an equivalent parameter $G\sim R^{2/z}$ is chosen. This suggests the possibility of using LM models with tunable diffusion length to simulate processes with simultaneous diffusion of many atoms. A scaling approach is used to justify exponent values and dynamic relations for both models, including the significant decrease of surface roughness as R or G increases.

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I. INTRODUCTION

Thin film growth by molecular beam epitaxy motivated the proposal of many atomistic and continuous models because it is one of the most important techniques to produce high quality films with smooth surfaces [1]. The main parameter to determine the film surface morphology is the ratio $R\equiv D/F$ between the free adatom diffusion coefficient D and the atomic flux F, but other quantities also play an important role, such as lateral binding energies and barriers at step edges [2,3]. In low temperatures, one frequently assumes irreversible adatom attachment to island edges or to other atoms at the same height, consequently R and the surface coverage determine scaling properties—in submonolayer growth, this is the case of models with critical island size $i=1$ [2,4]. Growth models that incorporate the competition between deposition and diffusion are usually called full or collective diffusion (CD) models. Most works on CD models focus on a particular application and estimate physical quantities such as diffusion coefficients or energy barriers in films with a small number of layers [2]. On the other hand, for studying scaling properties, one frequently simplifies the growth dynamics by introducing limited mobility (LM) models, where the final position of aggregation of an atom is chosen immediately after its deposition, i.e., before another atom is deposited. In most cases, adatom diffusion in a LM model is restricted to nearest neighbor sites, but this is not a general rule.

Dynamic scaling is intensively studied theoretically and experimentally because it helps to find the essential physical mechanisms of the film growth, independently of the details of the interactions [2,3]. The set of scaling exponents determines the universality class of the process, which is usually connected to a stochastic equation [2,3]. With LM models, this connection can frequently be done by inspection of their symmetries [3,5], by numerical methods, and sometimes by analytical methods. On the other hand, the situation with CD models is more complicated. Most numerical works suggest temperature-dependent effective exponents and the possibility of anomalous scaling [6–9]. However, a recent renormalization study [10–12] of a CD model with reversible aggregation of all adatoms showed that it belongs to the universality class of the fourth-order nonlinear growth equation proposed by Villain, Lai and Das Sarma (VLDS) [13,14]. The numerical study of the renormalization flux [11] explained the long crossover from the linear to the nonlinear growth exponent observed in simulations by Wilby *et al.* [15], and helps to understand the apparently nonuniversal behavior observed in previous works.

Since numerical simulations are widely used to study film growth models, a deeper analysis of dynamic scaling of CD models is justified, in the light of the recent advance in the area. Other relevant questions may also be addressed, such as the role of the diffusion-to-deposition ratio and the use of LM models to approximate collective diffusion processes.

The first aim of this paper is to study dynamic scaling of a CD model with irreversible adatom attachment to lateral and upper neighbors, i.e., where only isolated atoms at terraces are mobile. Simulations in two-dimensional substrates ($2+1$-dimensional model) are performed, which is the most relevant case for applications. The comparison of roughness distributions provides the strongest evidence that this model belongs to the VLDS class for any R, even using data of small lattices. On the other hand, estimates of growth and roughness exponents are slightly below the VLDS values, in agreement with previous numerical works. We also discuss the role of the diffusion-to-deposition ratio R on the dynamic

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[*] reis@if.uff.br

scaling relation, showing a drastic reduction of surface roughness as R increases.

Subsequently, we study a limited mobility (LM) model that mimics the dynamics of the above CD model. In order to tune the diffusion length in the LM model, an adsorbed atom executes up to G random steps and stops only if it encounters a lateral neighbor during its diffusion. We show that this model is also in the VLDS class and that results of the CD model can be reproduced by an equivalent LM model with $G \sim R^{0.6}$. This is a nontrivial result, since the effective number of steps of free atoms in the CD model is much smaller than R. It also suggests that extensions of the LM model may be used to speed up simulation of realistic models with large R.

Finally, we will use scaling arguments to predict the time evolution of the correlation length in the CD and the LM model and to explain their connection and the significant decrease of surface roughness as R or G increases.

The rest of this paper is organized as follows. In Sec. II, we will define the CD and LM models, present the most common stochastic equations for growth processes with diffusion and review the basics of dynamic scaling. In Sec. III, we will show numerical results for the CD model. In Sec. IV, we will show numerical results for the LM model. In Sec. V, we present scaling arguments to explain the role of R and G in the dynamic scaling relations. In Sec. VI, we present our conclusions.

II. ATOMISTIC MODELS, STOCHASTIC GROWTH EQUATIONS, AND DYNAMIC SCALING

All models studied here are of solid-on-solid type. Deposition occurs in a two-dimensional substrate of linear size L, with the lattice parameter as the length unit. The surface is flat at $t=0$. There is an external flux of F atoms per site per unit time. In our simulations, we take $F=1$ for simplicity, so that one layer of atoms is deposited in one time unit.

In the CD model, an incident atom is adsorbed upon landing above a previously deposited atom or a substrate site. All adsorbed atoms with no lateral and no upper neighbor (i.e., isolated atoms in terraces) diffuse with coefficient D, which is the number of random steps to neighboring columns per unit time. If an adatom has a lateral or an upper neighbor, then it is permanently aggregated at that position.

The diffusion coefficient D is expected to depend on temperature as $D=\nu_0 \exp(-E_s/k_B T)$, where ν_0 is a frequency of order 10^{12} Hz and E_s is the activation energy in a flat surface. However, since we are not interested in studying a particular growth process, we will use $R \equiv D/F$ as the model parameter in the rest of this paper.

In real processes, the ratio R may vary of several orders of magnitude. For this reason, most simulation works are performed with R ranging from 10^4 to 10^9. Under these conditions, almost all simulation time is spent with adatom diffusion and the substrate size has to be much larger than the diffusion length $\sim\sqrt{R}$ during one layer deposition. Thus, simulations with large R are frequently limited to the submonolayer regime. However, here we are interested in studying deposits with many layers, up to the regime of roughness saturation. Consequently we simulated smaller values of R, ranging between 10^1 and 10^4. Since the basic dynamic mechanisms of the model are the same for any R, a single universality class is expected.

In simulations of the CD model, the adsorption of each new atom occurs in a time interval $1/L^2$, using $F=1$. Subsequently, R/L^2 steps of randomly chosen free atoms are performed. Since R/L^2 is usually not integer, and it may be small for large L, we keep its fractional part for determining the number of steps after the next adsorption events. This represents the correct value of the ratio R.

The second model studied in this work is of LM with tunable diffusion time. Only the last incident atom moves on the surface, executing up to G random steps to nearest neighbor columns. If it encounters a lateral neighbor during its diffusion, it permanently aggregates at that position. Otherwise, it permanently aggregates at the final position after the G steps. As far as we know, this growth model was not studied before. However, the idea of tuning the diffusion time in a LM model is not new; for instance, it was proposed in the Family model [16] (deposition with relaxation to smaller heights) and in an extension [7] of the model of Das Sarma and Tamborenea [17].

Our simulations of the LM model are also restricted to small values of G, not only for reaching long times but also for the interest to compare results with the CD model.

Note that both the CD and the LM models studied here allow downhill movement at terrace edges without additional energy barries (Ehrlich-Schwebel) [18]. This movement always lead to permanent aggregation at the step edges. For this reason, the inverse process (uphill movement) is not possible.

In the hydrodynamic limit, diffusion dominated processes are expected to be described by a fourth order equation of the form [3]

$$\frac{\partial h}{\partial t} = \nu_4 \nabla^4 h + \lambda_4 \nabla^2 (\nabla h)^2 + \eta(\vec{x},t), \tag{1}$$

where $h(\vec{x},t)$ is the height at position $\vec{x}$ and time t in a d-dimensional substrate, ν_4 and λ_4 are constants and η is a Gaussian (nonconservative) noise. The contribution of the average external flux (a constant term) was omitted in Eq. (1). The linear version ($\lambda_4=0$) is the Mullins-Herring (MH) equation [19], while the nonlinear case is the Villain-Lai-Das Sarma equation [13,14].

If atom diffusion leads to frequent downhill movements, surface tension may be the dominant mechanism, thus the second order linear equation of Edwards and Wilkinson [20] provides the suitable hydronamic description: $\frac{\partial h}{\partial t} = \nu_2 \nabla^2 h + \eta(\vec{x},t)$. Nonlinear terms in this equation are expected if the growth mechanisms lead to excess velocity of Kardar-Parisi-Zhang (KPZ) type [21], but this is not the case in the present work.

The most important geometrical quantity to characterize the film surface is the roughness (or interface width)

$$W \equiv \langle \overline{(h-\bar{h})^2} \rangle^{1/2}, \tag{2}$$

where the overbars represent spatial averages and the angular brackets represent configurational averages. For short times, it scales as

$$W \sim t^{\beta}, \tag{3}$$

where β is called the growth exponent. For long times, in the steady state, the interface width saturates at

$$W_{sat} \sim L^{\alpha}, \tag{4}$$

where α is called the roughness exponent. The crossover time from the growth regime to the steady state scales as

$$t_{\times} \sim L^{z}, \tag{5}$$

where

$$z = \alpha/\beta \tag{6}$$

is the dynamical exponent. These relations can be condensed in the dynamic scaling relation originally proposed by Family and Vicsek [22]

$$W = L^{\alpha} f(t/L^{z}), \tag{7}$$

where f is a scaling function. Extensions of this scaling relation may include one or more model parameters [23–25], as will be shown in Sec. III.

We also measured the roughness distributions in the steady states of our models. $P(w_2)$ is defined as the probability density of the squared roughness of a given configuration to lie in the range $[w_2, w_2+dw_2]$ [26–28]. It is expected to obey the scaling relation

$$P(w_2) = \frac{1}{\sigma}\Psi\left(\frac{w_2 - \langle w_2\rangle}{\sigma}\right), \tag{8}$$

where $\sigma \equiv \sqrt{\langle {w_2}^2\rangle - \langle w_2\rangle^2}$ is the rms deviation of the squared roughness and Ψ is a universal scaling function. Recent works on KPZ models [29,30] showed that comparison of roughness distributions is a much more accurate method to determine the universality class of a growth model than measuring scaling exponents. There are alternative approaches, such as using space-time correlations and response functions [31], but the above quantities are sufficient for the current investigation.

The exponents of the (linear) EW and MH classes are exactly known; the EW class in $d=2$ has logarithmic scaling ($\alpha=0$, $\beta=0$) and the MH class has $\alpha=1$ and $\beta=0.25$. On the other hand, for the (nonlinear) VLDS class, the best estimates are calculated with atomistic models. The conserved restricted solid-on-solid (CRSOS) models [32–35] are suitable for this purpose because they were clearly shown to belong to the VLDS class [36,37] and their data have small finite-size corrections. The most accurate estimates of roughness and dynamic exponents [35] are very close to the one-loop values $\alpha=2/3$ and $z=10/3$ [3,13,38].

III. NUMERICAL RESULTS FOR THE COLLECTIVE DIFFUSION MODEL

In Fig. 1 we show the time evolution of the surface roughness for four values of R and different substrate sizes. Two lines with the expected slopes of the VLDS class ($\beta \approx 0.2$) [35,38] and of the MH class ($\beta=1/4$ exactly) are also shown in Fig. 1. Note that the roughness significantly decreases as R increases. For $R=10^4$, despite the long simulation time ($t=10^5$) and large size ($L=1024$), the roughness is always of order unity.

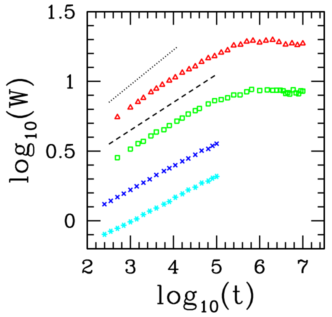

FIG. 1. (Color online) Time evolution of the surface roughness in the CD model with $R=10$ (triangles), $R=10^2$ (squares), $R=10^3$ (crosses), and $R=10^4$ (asterisks). The dotted line has slope 0.25 and the dashed line has slope 0.2.

For $R=10$ and $R=100$, the slopes of the growth regimes are close to 0.2. For larger R, the slope is slightly below 0.2, but the values of the roughness are very small, thus there may be strong corrections to the dominant scaling in Eq. (3). We recall that the EW roughness increases logarithmically, thus this possibility is ruled out.

For $R=10^2$, we calculated the saturation roughness for several sizes L, as shown in Fig. 2(a). A least-squares fit gives $\alpha \approx 0.64$, which is close to the VLDS exponent $\alpha \approx 2/3$ [35,38], but very far from the MH and EW values ($\alpha=1$ and $\alpha=0$, respectively). We estimated effective roughness exponents following the same lines of Ref. [35], but the number of data points is too small to provide a reliable extrapolation.

In Fig. 2(b), we show the scaled roughness distributions for $R=10^2$ and $R=10^3$, in lattice size $L=128$, and the distribution for the CRSOS model in size $L=64$ [39]. The exact distribution for the MH equation (linear, fourth order) [27] is

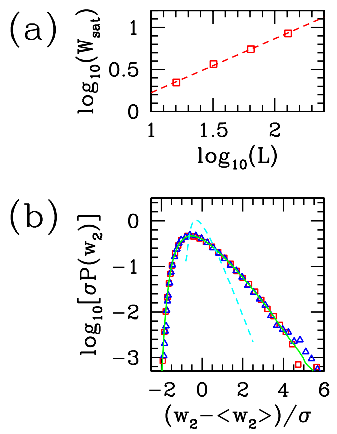

FIG. 2. (Color online) (a) Saturation roughness as a function of the system size for the CD model with $R=10^2$. The dashed line is a least-squares fit of the data, with slope 0.64. (b) Scaled roughness distributions for the CD model with $R=10^2$ (squares) and $R=10^3$ (triangles), both in $L=128$, for the diffusive CRSOS model in $L=64$ [39] (solid curve), and for the MH equation [27] (dashed curve).

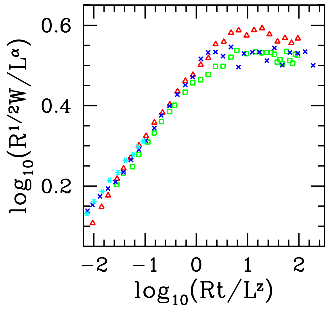

FIG. 3. (Color online) Scaled roughness versus scaled time for the CD model with $R=10$ in $L=64$ (triangles), $R=10^2$ in $L=128$ (squares), $R=10^3$ in $L=128$ (crosses), and $R=10^4$ in $L=1024$ (asterisks), using $\alpha=2/3$ and $z=10/3$.

also shown in Fig. 2(b) for comparison. The distribution for the EW equation is a delta function [not shown in Fig. 2(b)]. The excellent data collapse with the CRSOS curve confirms that the CD model is indeed in the VLDS class. The distributions for the CD model in size $L=64$ [not shown in Fig. 2(b)] also collapse very well with the curves for $L=128$, which indicates small finite-size effects.

These results lead to the conclusion that the difference between the estimated exponents and the VLDS values are only due to finite-size effects. However, it is very difficult to obtain accurate results in larger sizes because simulation times become very large (note the large dynamical exponent $z=10/3$).

Given that the class of the model is VLDS, now we analyze the effect of the diffusion-to-deposition ratio R on dynamic scaling by collapsing data for different values of D and L. We propose the extension of the Family-Vicsek relation (7) as

$$W=\frac{L^{\alpha}}{R^{x}}f(R^{y}t/L^{z}), \tag{9}$$

with $\alpha=2/3$, $z=10/3$, and exponents x and y to be determined. Similar scaling forms are found in models with competitive growth dynamics [23,24]; a recent example is shown in a model of random deposition with surface relaxation containing a temperaturelike parameter [25].

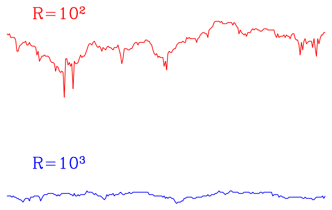

FIG. 4. (Color online) Cross sectional views of the surfaces of films grown with two values of R and $L=256$, at $t=10^6$.

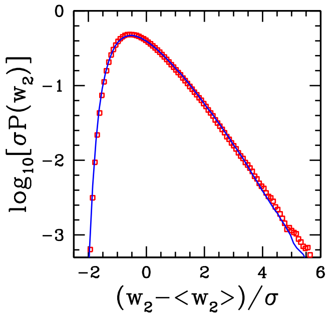

FIG. 5. (Color online) Scaled roughness distributions for the LM model with $G=10$ in $L=128$ (squares) and for the diffusive CRSOS model in $L=64$ [39] (solid curve).

In Fig. 3 we show that a good data collapse is obtained with $x=0.5$ and $y=1$ in Eq. (9), using data for four different values of R. A significant deviation in Fig. 4 is observed only in the steady state for $R=10$, probably due to finite-size corrections—we used $L=64$ in that case. With those estimates of x and y in Eq. (9), we have

$$W=\frac{L^{\alpha}}{R^{1/2}}f(\xi_{CD}/L), \tag{10}$$

where ξ_{CD} is a lateral correlation length given by

$$\xi_{CD}=(Rt)^{1/z}. \tag{11}$$

The scaling exponent $x=0.5$ is not trivial and represents a drastic decrease in the roughness as R increases; in the steady state, we have $W_{sat}\sim L^{\alpha}/R^{0.5}$. Figure 4 shows cross sectional views of the film surface for $R=10^2$ and $R=10^3$ that illustrate this feature. As R increases, the size of the plateaus increases, and the steps between flat regions and the density of surface cracks decrease. The scaling ideas presented in Sec. V, in connection to the LM model, explain the exponent values obtained here.

IV. NUMERICAL RESULTS FOR THE LIMITED MOBILITY MODEL

The estimates of scaling exponents β and α for this model are similar to those of the CD model. For $G=10$, we obtain $\beta\approx 0.2$ in the growth regime and $\alpha\approx 0.60$ from the saturation roughness. Consequently, there is no improvement of previous numerical results by estimating those exponents. However, comparison with the roughness distribution of the CRSOS model provides again the strongest evidence of VLDS scaling, as shown in Fig. 5.

The VLDS scaling is not an obvious result for the LM model. The kinetic rules of our model with $G=1$ resemble those of the model of Das Sarma and Tamborenea (DT) [17], where the incident atom permanently aggregates at a site with at least one lateral neighbor, chosen only among the column of incidence and its nearest neighbors. However, simulations of the DT model in $d=2$ suggest EW scaling [40,41]. We also recall that Ref. [7] proposed an extended DT model in $d=1$ where the incident atom searches for the aggregation position in a neighborhood of size l around the column of incidence. That model is also similar, though not equal to our LM model, and Ref. [7] suggested that it had anomalous scaling.

In Fig. 6(a) we show results for four values of R and the values of G that give nearly the same roughness as a function of time. For $R\leq 10^3$, we compare data in $L=256$. For $R=10^4$ and $G=60$, we compare data in $L=1024$ because

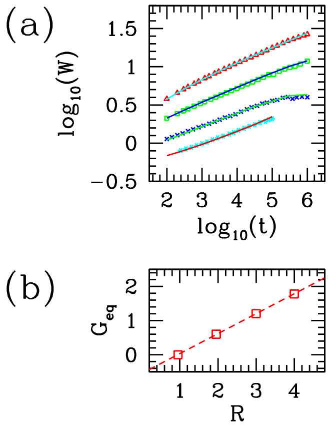

FIG. 6. (Color online) (a) Time evolution of the surface roughness: CD model with $R=9$ in $L=256$ (triangles), $R=90$ in $L=256$ (squares), $R=10^3$ in $L=256$ (crosses), and $R=10^4$ in $L=1024$ (asterisks); solid curves are for the LM model, from top to bottom with $G=1$ in $L=256$, $G=4$ in $L=256$, $G=16$ in $L=256$, and $G=60$ in $L=1024$. (b) Equivalent parameter G of the LM model as a function of the parameter R of the CD model. The dashed line is a linear fit.

roughness saturation is attained at short times for small lattices; however, due to the larger size, only data until $t=10^5$ were calculated in this case. Since the roughness evolution of the CD model can be reproduced by the LM model with an equivalent parameter G_{eq}, simulation of realistic CD models may be faster with the use of equivalent LM models with tunable number of steps.

In Fig. 6(b) we show G_{eq} as a function of R and a linear fit which gives

$$G_{eq} \approx 0.28R^{0.58}. \tag{12}$$

This means that large values of the diffusion-to-deposition ratio are represented by much smaller values of G. We understand that G_{eq} is a measure of the average number of steps of an adatom before aggregation, while the meaning of R is more complicated due to the many atom dynamics involved in the CD model. The scaling relation (12) implies that the effective number of steps is much smaller than R, particularly for large R. The scaling exponent in Eq. (12) is discussed in Sec. V.

V. SCALING RELATIONS

In the CD model, the correlation length of Eq. (11) is obtained by replacing the growth time by Dt (taking $F=1$ and unit lattice size). This is the natural variable to represent the faster adatom dynamics as D increases. On the other hand, the exponent $z>2$ in Eq. (11) represents the subdiffusive propagation of correlations, which is a universal VLDS feature.

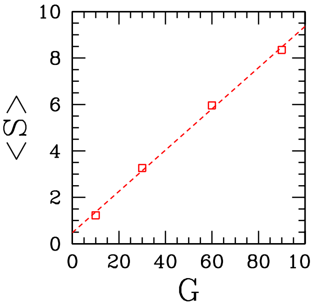

FIG. 7. (Color online) Average number of steps of free atoms as a function of G in the LM model. The dashed line is a linear fit of the data.

In the LM model, G has the role of determining the size of terraces because there is only one adatom moving at each time. This is confirmed in Fig. 7, where we plot the average number of steps $\langle S\rangle$ before aggregation as a function of G. We observe a linear increase in that plot for $G>10$, with some deviations for smaller G (where $\langle S\rangle$ is quite small).

For this reason, the average distance traveled by an adatom before aggregation is proportional to $G^{1/2}$. Thus, the length of the plateaus in the film surface is also of order $G^{1/2}$. The same factor is expected to appear in the scaling of the lateral correlation length,

$$\xi_{LM} \sim G^{1/2}t^{1/z}. \tag{13}$$

The equivalence between the CD and LM models is observed if both correlation lengths scale in the same form. Matching Eqs. (11) and (13), we obtain

$$G \sim R^{2/z}, \quad \frac{2}{z} = 0.6. \tag{14}$$

This exponent is in excellent agreement with the numerical value of Sec. IV [Eq. (12)].

The LM model features also shows that the strong decrease in surface roughness as G (or R) increases comes from two contributions, as follows.

Since the plateaus in the two-dimensional substrate have length of order $l_G \sim G^{1/2}$, they have area of order $l_G^2 \sim G$. This is the order of the number of aggregated atoms in the plateau. The random fluctuations of this number are of order $G^{1/2}$, and relative fluctuations of order $G^{-1/2}$. These fluctuations appear in the borders between the flat regions, corresponding to formation of small hills and valleys. Since the atoms have unit size, they appear in the surface roughness amplitude as a factor $G^{-1/2}$. This argument is similar to those of Refs. [23,24] for competitive growth models.

The increase in the size of the plateaus also reduces the roughness in a length scale of order l_G. The reduction factor is the local roughness for $G\approx 1$ in a length l_G, which is ${l_G}^{\alpha} \sim G^{\alpha/2}$.

Combining the effects of smoothing larger regions and reducing the size of steps, the parameter G leads to an overall reduction in surface roughness by $G^{1/2}G^{\alpha/2}=G^{(\alpha+1)/2}$. The dynamic scaling relation reads

$$W = \frac{L^{\alpha}}{G^{(\alpha+1)/2}} f(G^{1/2}t^{1/z}/L). \tag{15}$$

The values of α and z for the VLDS class [3,35,38] give an exponent $(\alpha+1)/2 \approx 5/6$ which agrees with numerical values obtained from data collapse methods (similar to Fig. 3).

The connection between the CD and LM models [Eq. (14)] leads to a dynamic scaling relation for the CD model in the form of Eq. (9) with $x=(\alpha+1)/z\approx 1/2$ and $y=1$. These values are in excellent agreement with the results in Fig. 3 for the CD model.

VI. CONCLUSION

We studied a CD model with irreversible step edge attachment and diffusion-to-deposition ratio R, and a LM model with maximum number of steps G after adsorption of a new atom, with the same aggregation rule. Simulations in two-dimensional substrates were performed. Estimates of scaling exponents are close to the values of the VLDS class and comparison of scaled roughness distributions provide a stronger support to that conclusion. This confirms the recent analytical connection of collective diffusion models and stochastic equations, and improves previous numerical results. The VLDS scaling of the LM model contrasts with the evidence of EW scaling in the DT model, which has similar stochastic rules. The CD model shows a remarkable decrease of the roughness as $R^{1/2}$, and a scaling of the lateral correlation length as $(Rt)^{1/z}$. In the LM model, the correlation length increases as $G^{1/2}t^{1/z}$. The roughness evolution of the CD model is reproduced if the equivalent parameter $G_{eq}\sim R^{2/z}$ is used. This connection and the smoothing of the surface as G increases explain the numerical values of exponents obtained in the dynamic scaling relations.

Previous works on CD models suggested R-dependent exponents and anomalous scaling in $1+1$ and $2+1$ dimensions [6–9]. This was certainly a consequence of corrections to the dominant VLDS scaling. Our work shows that comparison of roughness distributions is the best procedure to avoid the effects of those corrections. Such comparison may also be useful in the study of some growth models of porous films that are also in the VLDS class [42,43], in contrast to the usually expected KPZ scaling.

The most common LM models have diffusion steps restricted to nearest neighbors, while the CRSOS models allow an infinitely large number of steps. For this reason, they provide results very different from the CD models, which in turn are more realistic for diffusion dominated growth. However, the LM models have the advantage of being simpler for simulation work. Thus, the possibility of using a LM model with tunable diffusion length to reproduce features of a CD model is promising, for instance in extensions to reversible lateral aggregation.

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