

## Smoothing in thin-film deposition on rough substrates

T. A. de Assis<sup>1,\*</sup> and F. D. A. Aarão Reis<sup>2,†</sup>

<sup>1</sup>*Instituto de Física, Universidade Federal da Bahia, Campus Universitário da Federação, Rua Barão de Jeremoabo s/n, 40170-115, Salvador, BA, Brazil*

<sup>2</sup>*Instituto de Física, Universidade Federal Fluminense, Avenida Litorânea s/n, 24210-340 Niterói RJ, Brazil*

(Received 25 August 2015; published 24 November 2015)

The evolution of the surface roughness  $W$  of a thin film deposited on a rough substrate is studied with a model of temperature-activated adatom diffusion, irreversible lateral aggregation, and no step energy barrier, in which the main parameter is the ratio  $R$  of diffusion and deposition rates. At sufficiently low temperatures ( $R \lesssim 10$ ), the average number of adatom steps after adsorption is very small, thus  $W$  monotonically increases with time  $t$  due to an approximately uncorrelated deposition at short times. If the temperature is not very low ( $R \sim 10^3$  or larger), smoothing occurs at short times and the Villain-Lai-Das Sarma (VLDS) growth equation governs the long time roughening, which is attained after a crossover time  $t_c$  that increases with the correlation length  $\xi_i$  of the substrate. Scaling arguments predict the dependence of  $t_c$  on temperature and on the substrate production time and the scaling relation for the difference between the roughness of films deposited on rough and flat substrates, in good agreement with numerical results. The effect of temperature is not a direct extension of previous results on flat substrates because the short wavelength fluctuations delay the formation of terraces. For this reason, the effective energy obtained from the dependence of  $t_c$  on  $R$  is 40% of the energy of activated adatom diffusion. A scaling law for the initial smoothing is proposed as  $W/W_i = \Psi(t/t_{c1})$ , with a crossover time  $t_{c1} \equiv R^{-\theta} \xi_i^z$ , where  $W_i$  is the substrate roughness,  $\theta \approx 0.4$ , and  $z$  is the VLDS dynamical exponent. It provides good data collapse if  $W$  is not very small and is suggested to be tested experimentally.

DOI: [10.1103/PhysRevE.92.052405](https://doi.org/10.1103/PhysRevE.92.052405)

PACS number(s): 81.15.Aa, 68.55.-a, 68.35.Ct, 05.40.-a

### I. INTRODUCTION

Models of thin film deposition are widely studied due to the technological relevance of these processes [1]. They usually consider initial flat surfaces, which represent substrates whose fluctuations are of atomic or molecular size, typically below 1 nm. Roughening takes place due to the interplay of the deposition noise and mechanisms of surface relaxation characteristic of the material and substrate-film interactions [2,3]. On the other hand, in the absence of deposition, the smoothing of corrugated surfaces is observed, i.e., they evolve to configurations of decreasing roughness [4–7]. This is a typical situation in thermal annealing [8,9]; in the absence of the deposition noise, a conservative thermal noise brings the system to statistical equilibrium configurations.

A less studied case is the deposition of a thin film over an initially rough or patterned substrate. Majaniemi *et al.* [10] and Nguyen *et al.* [11] addressed this problem with the study of Edwards-Wilkinson (EW) [12] and Mullins-Herring (MH) [13] growth on rough substrates. They show that the roughness and the height-height correlation function can be split in smoothing and roughening terms, respectively dominant at short and long times, with scaling forms of the smoothing term depending on the initial roughness and correlation length. A particularly interesting feature is the possibility of nonmonotonic roughness evolution, which was also observed in unstable film growth on rough substrates [14,15]. An equivalent problem is that of sudden changes in interface growth dynamics, which consider the interplay of the initial and final dynamics in the interface evolution [16–18]. Those

works had a focus on EW and Kardar-Parisi-Zhang (KPZ) [19] growth, with the KPZ-EW changes showing a much slower decay of the roughness due to the initially large correlation length [18].

Here we are interested in deposition of thin films whose dynamics is governed by the competition of deposition and adsorbate diffusion taking place on rough substrates. This type of surface dynamics was not considered in the previous work on sudden changes of interface growth dynamics. We consider a deposition model with temperature-activated adatom diffusion, irreversible lateral aggregation, and no additional step energy barrier [20]. This is a low-temperature approximation of models with reversible aggregation mechanisms, such as the Clarke-Vvedensky (CV) model [21,22]; for a recent comparison with CV model results, see Ref. [23]. These deposition models are represented by the Villain-Lai-Das Sarma (VLDS) [24,25] equation in the hydrodynamic limit [2].

An important question is concerned with the roughness evolution at short times, for which previous work predicted cases of monotonic and nonmonotonic variation in EW and KPZ growth. For deposition in temperatures that are not very low, we will show that smoothing takes place at short times, followed by the expected long time VLDS roughening. The crossover features of the surface roughness, from smoothing to the asymptotic scaling, are addressed by extending the scaling approach of Ref. [18].

However, this problem raises other questions intrinsic to activated diffusion models and important for possible applications. First, we discuss the role of the temperature in the evolution of the roughness. The changes in the surface roughness are shown to scale with the diffusion-to-deposition ratio  $R$  with a nontrivial exponent, which is a consequence of a delay in the propagation of correlations relatively to growth on flat substrates. Second, we discuss how the film roughness

\*thiagoaa@ufba.br

†reis@if.uff.br

can be properly scaled during the smoothing to provide information on the film growth dynamics and on the formation of the rough substrate without requiring the knowledge of its evolution on flat substrates. This leads to a scaling law for smoothing, which is suitable for experimental tests, in contrast to the scaling relations obtained in previous works on the same type of crossover [16–18].

This paper is organized as follows. In Sec. II we present the models and basic concepts and definitions of kinetic roughening theory. In Sec. III we discuss the general trends of the film roughness in deposition on rough surfaces. In Sec. IV we use scaling approaches to study the crossover of the difference of roughness between flat and rough substrates and present numerical results to support them. In Sec. V we propose the scaling for surface smoothing. Section VI summarizes our results and conclusions.

## II. MODELS, ROUGHNESS SCALING, AND SIMULATION

### A. Models of rough substrate production

We consider three growth models on simple cubic lattices for generating the rough substrates. The lattice constant is the unit length. Lattices of lateral size  $L$  are considered, with periodic boundaries in the  $x$  and  $y$  directions. The initial condition in all cases is a flat interface in the plane  $z = 0$ . All models are of solid-on-solid type; i.e., overhang and pore formation is not allowed. Most simulations are performed in lattices with  $L = 1024$ . The time for substrate production is  $s$ .

The first model is the erosion version of the etching model of Mello *et al.* [26], which is hereafter called the E model. At each growth attempt, a column  $(x, y)$  is randomly chosen. Its current height  $h(x, y) \equiv h_0$  is decreased by one unit, i.e.,  $h(x, y) \leftarrow h_0 - 1$ . Subsequently, any neighboring column whose height is larger than  $h_0$  decreases to the height  $h_0$ . These rules are illustrated in the two-dimensional scheme of Fig. 1(a).

One time unit corresponds to  $L^2$  selections of random columns in the E model. The time for the substrate production varies from  $s = 20$  to 200.

The second model is that of Wolf and Villain (WV) [27], which is described in terms of particle deposition. The column

$(x, y)$  of incidence of a new particle is randomly chosen. The numbers of lateral neighbors at the top of this column and at the tops of the four nearest neighbor (NN) columns are calculated. The new particle aggregates at the top of the column with the largest number of lateral neighbors. If there is a tie between column  $(x, y)$  and one or more NN, then the particle aggregates at the former. If there is a tie between one or more NN columns, one of them is randomly chosen. The WV growth rules are illustrated in the two-dimensional scheme of Fig. 1(b).

The third model is the WV model with multiple-hit aggregation. At a given column  $(x, y)$ , the aggregation of a particle is executed only after that column has been chosen for aggregation  $m$  times. The original WV model has  $m = 1$ . We also generated rough substrates with  $m = 10$ , which we call the WV10 model. Large values of  $m$  are expected to produce mounded surface morphology [28,29], which is a common feature in films of several materials.

In the WV models, one time unit corresponds to aggregation of  $L^2$  particles; in the model with noise reduction, it corresponds to  $mL^2$  trials. The time for substrate production with those models varies from  $s = 1000$  to 3000.

### B. Model of film deposition

Here we present the model of deposition, diffusion, and aggregation for thin film growth on rough or flat substrates. We consider an external flux of  $F$  atoms per substrate column per unit time in the negative  $z$  direction. For simplicity, these atoms are also of the size of a lattice site; thus no effect of mismatch with the rough substrates is considered.

The incident atom is adsorbed upon landing above a previously deposited atom (an adatom) or above a substrate site. Adatoms with no lateral and no upper neighbor (i.e., isolated atoms in terraces) are mobile. Their diffusion coefficient  $D$  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 depends on temperature as  $D = \nu_0 \exp(-E_a/k_B T)$ , where  $\nu_0$  is a characteristic frequency and  $E_a$  is an activation energy. The relevant model parameter is the diffusion-to-deposition ratio

$$R \equiv \frac{D}{F} = \frac{\nu_0}{F} \exp(-E_a/k_B T). \quad (1)$$

In the simulations we consider  $\nu_0/F = 10^{14}$ , which is typical of very small atomic flux [22]. The activation energy is fixed as  $E_a = 0.6$  eV, which is close to the values of (100) homoepitaxy of most metals (they vary from  $\approx 0.4$  eV for Ag to  $\approx 0.9$  eV for Rh [22]). Thus, the deposition temperature  $T$  is used as the control parameter in the simulations, although the values of  $R$  are usually shown in the presentation of results and are used in the scaling relations. For each substrate and each temperature, the number of different film configurations grown varies between 100 and 1000.

The thin film growth time  $\tau$  is measured in number of deposited monolayers; i.e.,  $\Delta\tau = 1$  is the time of deposition of  $L^2$  atoms. This is equivalent to stating that  $\tau$  is measured in units of  $1/F$ . Here  $\tau = 0$  corresponds to the initial substrate, which may be rough or flat.

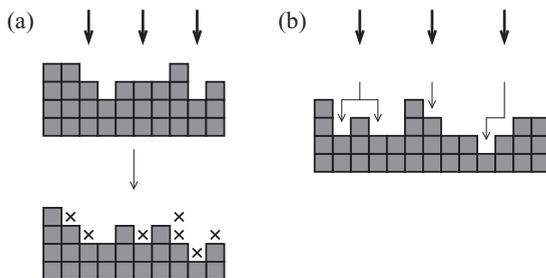


FIG. 1. (a) Growth rules of the E model: the attack of the solution at three columns is indicated by arrows in the upper panel, and the resulting configuration of the solid is shown in the lower panel, with removed particles indicated by crosses. (b) Growth rules of the WV model, in which the columns of particle incidence are indicated by thick arrows and aggregation positions, are indicated by thin arrows. The double arrow of the left case indicates that one of the NN columns will be randomly chosen.

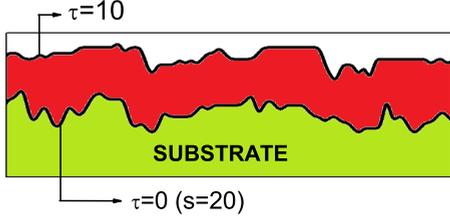


FIG. 2. (Color online) Portion of cross section of lateral size 1024 of a deposit with 10 layers of atoms ( $\tau = 10$ ) deposited at  $T = 310$  K over a rough substrate produced by the E model with  $s = 20$ .

The total time from the beginning of substrate production to the regime of film growth is called  $t$ , in order to match the definition of previous works on sudden changes of growth dynamics [16–18]. Consequently,

$$t = \tau + s. \quad (2)$$

Figure 2 shows a cross section of a deposit with 10 layers of atoms ( $\tau = 10$ ) deposited on a rough substrate produced by the E model with  $s = 20$ , at  $T = 310$  K ( $R \approx 1.7 \times 10^4$ ).

### C. Basics of kinetic roughening

The simplest quantity for characterizing a rough interface is its roughness or interface width, which is defined as the rms fluctuation of the height  $h$ :

$$W = W_2^{1/2} \equiv \left[ \overline{\langle (h - \bar{h})^2 \rangle} \right]^{1/2}. \quad (3)$$

The overbars in Eq. (3) represent spatial averages, and the angular brackets represent configurational averages. Most results in this paper are presented in terms of the squared roughness  $W_2$ .

In this work, our interest is to study the surface evolution in the growth regime, with negligible finite-size effects (far from the steady state). If the surface is initially flat, the roughness is expected to scale as [2,3]

$$W \sim t^\beta, \quad (4)$$

where  $\beta$  is the growth exponent.

The autocorrelation function is useful to characterize the lateral spread of interface fluctuations. It is a function of a distance  $r$  along the substrate and of the time  $t$  and is defined as

$$\Gamma(r, t) \equiv \langle [\tilde{h}(\vec{r}_0 + \vec{r}, t) \tilde{h}(\vec{r}_0, t)] \rangle, \quad r \equiv |\vec{r}|, \quad (5)$$

where  $\tilde{h} \equiv h - \bar{h}$  and the configurational averages are taken over different initial positions  $\vec{r}_0$ , different orientations of  $\vec{r}$ , and different deposits. In lattice models, we restrict the orientation of  $\vec{r}$  to the substrate directions  $x$  and  $y$ .

The correlation length  $\xi(t)$  is frequently estimated as the position of the first zero ( $r_0$ ) or of the first minimum ( $r_m$ ) of  $\Gamma(r, t)$  [30]. However,  $\Gamma(r, t)$  may oscillate with  $r$  before crossing the value  $\Gamma = 0$ . For this reason, we define a correlation length  $\xi_1(t)$  by

$$\Gamma(\xi_1, t) / \Gamma(0, t) = 0.1. \quad (6)$$

$\xi_1$  is slightly smaller than  $r_0$ , except if oscillations of  $\Gamma(r, t)$  appear before it goes to zero. That length is expected to

scale as

$$\xi_1 \approx At^{1/z}, \quad (7)$$

where  $z$  is the dynamical exponent and  $A$  is a model-dependent constant. This scaling is confirmed with good accuracy for several models in the EW and KPZ classes in Ref. [31] and for the models studied here. Instead,  $r_0$  and  $r_m$  show larger deviations from the time scaling of Eq. (7).

The roughness exponent  $\alpha$ , which characterizes the size dependence of the roughness, is related to the other ones as  $\alpha = \beta z$ .

When models of sudden changes in the growth dynamics are considered, the roughness of the interface after the change is usually called  $W_c(t, s)$ . The roughness of the interface grown only with the final dynamics, i.e., without change, is  $W_u(t)$ . In both cases, the interface was flat at  $t = 0$ .

For changes in linear growth equations, the difference in the squared roughness of the changed growth and of the pure growth,

$$\Delta W^2(t, s) \equiv |W_c^2(t, s) - W_u^2(t)|, \quad (8)$$

was exactly calculated in Ref. [17]. When the change takes place in the growth regime of the initial and the final dynamics [16–18], the general scaling form of  $\Delta W^2$  is

$$\Delta W^2 \sim s^c \tau^{-\gamma}, \quad (9)$$

where  $c$  and  $\gamma$  depend on both dynamics.  $\Delta W^2$  corresponds to the smoothing terms in Refs. [10,11].

For real systems applications, the time interval  $s$  of production of the rough substrate is usually meaningless. In this case, the relevant comparison is that between the roughness of the film grown on the rough substrate,  $W_r(\tau) \equiv W_c(t, s)$  [see Eq. (2)], and that of the film grown on the flat substrate,  $W_f(\tau) \equiv W_u(t - s)$ . Thus, we define the difference of squared roughness of films grown on those substrates as

$$\Delta W_{rf}^2(\tau) \equiv |W_r^2(\tau) - W_f^2(\tau)|, \quad (10)$$

i.e., with the same film growth time  $\tau$  for flat and rough substrates.

Note that  $\Delta W_{rf}^2(\tau) = |W_c^2(t, s) - W_u^2(t - s)|$ , which coincides with  $\Delta W^2(t, s)$  only for  $s = 0$ . However, for  $\tau \gg s$ , the scaling in Eq. (9) is also expected for  $\Delta W_{rf}^2(\tau)$ .

### D. Universality classes

When growth is dominated by surface diffusion, it is expected to be described by the VLDS equation [2,24,25]

$$\frac{\partial h(\vec{r}, t)}{\partial t} = v_4 \nabla^4 h + \lambda_4 \nabla^2 (\nabla h)^2 + \eta(\vec{r}, t), \quad (11)$$

where  $h(\vec{r}, t)$  is a coarse-grained height variable in a  $d$ -dimensional substrate,  $v_4$  and  $\lambda_4$  are constants, and  $\eta$  is a Gaussian, nonconservative noise. The contribution of the average external flux is omitted. The linear version of Eq. (11) ( $\lambda_4 = 0$ ) is the Mullins-Herring (MH) equation [13].

The temperature-activated diffusion model studied here is in the VLDS class, as confirmed by numerical simulation [20]. The scaling exponents of this class in  $d = 2$  are  $\beta \approx 0.2$  and  $z \approx 3.33$  [32]; for a discussion of small corrections to these values, see Ref. [33].

The E model is represented by the KPZ equation in the hydrodynamic limit:

$$\frac{\partial h}{\partial t} = \nu_2 \nabla^2 h + \lambda_2 (\nabla h)^2 + \eta(\vec{r}, t), \quad (12)$$

where  $\nu_2$  is a surface tension and  $\lambda_2$  is an excess velocity. The EW equation [12] corresponds to the KPZ equation with  $\lambda_2 = 0$ . In  $d = 2$ , recent numerical results for KPZ give  $\alpha \approx 0.39$  and  $\beta \approx 0.24$  [34]. The corresponding estimate of the dynamical exponent is  $z \approx 1.63$ , which will be considered in this work.

The universality class of the WV model in  $2 + 1$  dimensions is controversial. Simulations of the original model ( $m = 1$ ) in short times or small lattices give exponents close to the MH values  $\beta = 0.25$  and  $z = 4$ . This is the expected behavior in the present work. A crossover to EW scaling is suggested at longer times [35–37]. Simulations of the model with  $m = 5$  show mounded surface morphology and growth exponent near 0.30, with no evidence of crossover to EW scaling [28,29]. Thus, an effective MH scaling is also expected in our simulations with relatively short times.

### III. ROUGHENING VERSUS SMOOTHING

Figure 3 shows the roughness  $W$  as a function of the time of film deposition  $\tau$  in three different temperatures, starting with a substrate produced by the E model with  $s = 100$ . The roughness of this substrate is  $W_{\text{sub}} \approx 4.0$ , which corresponds to 1–2 nm for most metals and semiconductors and slightly more for larger molecules. The results for growth with an initial flat substrate are also shown in Fig. 3 for comparison.

At low temperature ( $T = 233$  K,  $R \approx 10$ ), the roughness increases with time since the beginning of the film deposition. The initial surface has a large number of kink sites, and the mobility of deposited particles is small; thus the initial growth takes place approximately as a random uncorrelated deposition, in which the roughness rapidly increases [2]. Below we will show additional results to support this interpretation. For high temperatures ( $R \approx 4 \times 10^3$  or larger in Fig. 3),

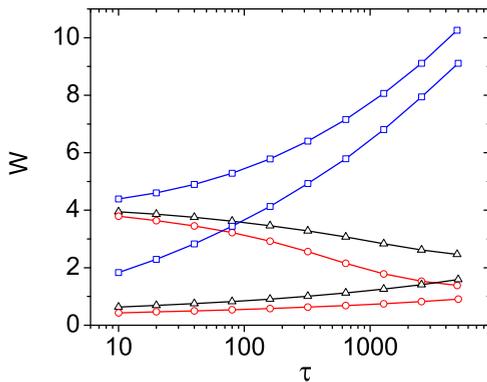


FIG. 3. (Color online) Roughness as a function of the film deposition time at temperatures 233 K ( $R \approx 10$ ; blue squares), 290 K ( $R \approx 4 \times 10^3$ ; black triangles), and 330 K ( $R \approx 7 \times 10^4$ ; red circles). For each temperature, the lower curve is for deposition on flat substrates and the upper curve is for deposition on rough substrates produced by the E model with  $s = 100$ .

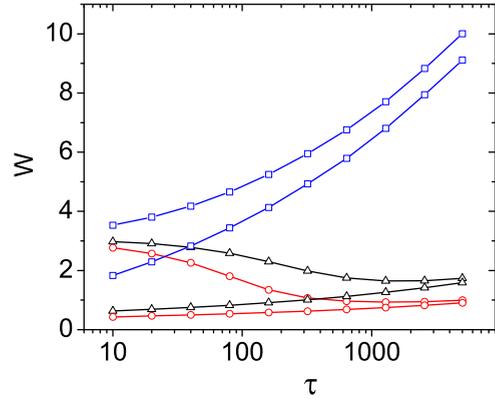


FIG. 4. (Color online) Roughness as a function of the film deposition time at temperatures 233 K ( $R \approx 10$ ; blue squares), 290 K ( $R \approx 4 \times 10^3$ ; black triangles), and 330 K ( $R \approx 7 \times 10^4$ ; red circles). For each temperature, the lower curve is for deposition on flat substrates and the upper curve is for deposition on rough substrates produced by the WV model with  $s = 3000$ .

the smoothening effect of the adparticle diffusion leads to a slow decrease of the roughness. Even after deposition of a large number of layers ( $\approx 5000$ ), smoothing persists, with no evidence that the roughness evolution will change this trend and begin to increase [11,18]. However, curves for deposition on rough and flat substrates at the same temperature seem to converge to the same value at long times.

Figure 4 shows the roughness of films deposited on substrates produced by the WV model at  $s = 3000$  as a function of  $\tau$ . The substrate has roughness  $W_{\text{sub}} \approx 3.0$ . Results for growth in a flat substrate are also shown. At short deposition times, Fig. 4 also shows that low-temperature deposition leads to time-increasing roughness, and high-temperature deposition leads to time-decreasing roughness. However, the high-temperature decrease is much faster than that observed in Fig. 3. For this reason, when a number of monolayers between 1000 and 2000 is deposited, the roughness reaches a minimum and subsequently increases. Again, this is consistent with the trend of the roughness of the film grown in the rough substrate to converge to that of the film grown in the flat substrate.

The contrast between the slow convergence in Fig. 3 and the rapid convergence in Fig. 4 cannot be related to the value of the roughness of the substrates, since it changes only 25% from the E model at  $s = 100$  to the WV model at  $s = 3000$ . Instead, this difference is related to the correlation length behavior. The low-temperature case deserves a separate discussion, as explained below.

In Fig. 5 we show  $\xi_1$  as a function of  $\tau$  for deposition at 310 K (a typical high temperature in the present context) in three different substrates: the flat one and those produced by E ( $s = 100$ ) and WV ( $s = 3000$ ) models. In flat substrates, the correlation length increases with the VLDS exponent  $1/z \approx 0.3$  [20]; after deposition of the maximal thickness of 5000 monolayers, it reaches  $\xi_1 \approx 60$ . The correlation length of the WV model substrate is small ( $\xi_1 \approx 25$ ). Thus, the film deposition is able to create longer correlations since short times, so that  $\xi_1$  rapidly attains the VLDS scaling. On the other hand, the correlation length of the E model substrate is  $\xi_1 \approx 60$ . Thus, even after deposition of many layers ( $t \approx 5000$ ), the

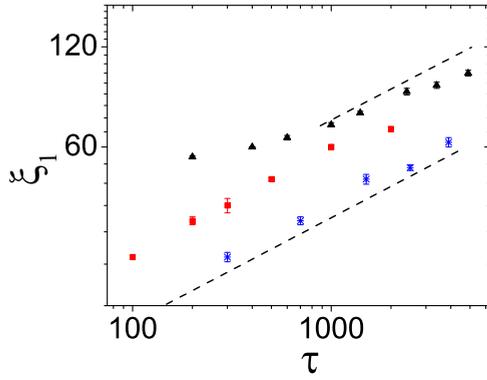


FIG. 5. (Color online) Correlation length  $\xi_1$  as a function of the film deposition time at 310 K ( $R \approx 1.7 \times 10^4$ ) in three different substrates: the flat one (blue asterisks) and those produced by WV ( $s = 3000$ ) (red squares) and E ( $s = 100$ ) (black triangles) models. Error bars are also shown. The dashed lines have slope 0.3.

VLDS kinetics is not able to create correlations in much longer distances. Indeed, for deposition on E model substrates, Fig. 5 shows that  $\xi_1$  does not follow the expected scaling at the longest times. This analysis shows that the correlation length of the substrate fluctuations is the main quantity to determine the rate in which the roughness converges to that of deposition on a flat substrate.

Now we turn to the case of low-temperature deposition. In Figs. 6(a) and 6(b), we show the autocorrelation function  $\Gamma(r, \tau)$  as a function of  $r$  for several times at  $T = 233$  K ( $R \approx 10$ ) in the substrates produced respectively by the E model and the WV model. In both cases,  $\Gamma$  increases in time for small  $r$  but has small changes for larger  $r$ , so that  $r_0$  and  $r_m$  slowly vary in time; correspondingly,  $\xi_1$  will show an apparently unphysical time decrease. It means that only short wavelength fluctuations increase, while long wavelength fluctuations are approximately constant. This is consistent with our interpretation of an approximately uncorrelated deposition, i.e., a deposition with negligible surface diffusion.

Additional support to this interpretation follows from a related deposition model with a step number parameter  $G$  that was studied in Ref. [20] (with flat substrates). It was connected to the present model by the relation  $G \approx 0.3R^{0.58}$ , and the average number of steps of an adatom was  $N_s \approx 0.1 G$  [20]. For deposition at  $T = 233$  K, these relations give an estimate  $N_s \approx 0.1$ , suggesting that most atoms do not move after adsorption; this is uncorrelated growth. At 290 K, the same relations give  $N_s \approx 4$ . Thus, smoothing of a rough substrate is related to a non-negligible adatom mobility at nanoscopic level ( $N_s > 1$ ), even if diffusion lengths are very small.

#### IV. CROSSOVER SCALING OF ROUGHNESS DIFFERENCE

In this section we study the scaling relations for the difference between the roughnesses of the films grown on rough and flat substrates,  $\Delta W_{r,f}^2(\tau)$ . The scaling arguments of Ref. [18] are extended to predict its time decay and the effect of the initial roughness and correlation length (Sec. IV A).

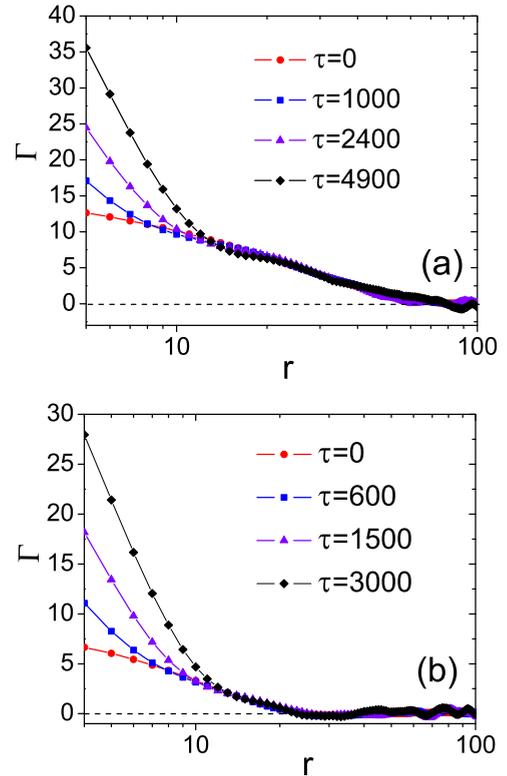


FIG. 6. (Color online) Autocorrelation function as a function of the distance  $r$  at several scaled times, for deposition at  $T = 233$  K ( $R \approx 10$ ) on substrates produced by (a) the E model ( $s = 100$ ) and (b) the WV model ( $s = 3000$ ). The horizontal dashed lines indicate  $\Gamma(r, \tau) = 0$ .

Numerical examples are presented in Sec. IV B. Subsequently, we discuss the role of temperature (or the ratio  $R$ ) on that smoothing term (Sec. IV C), which is very different from its role on roughness scaling with an initially flat substrate due to the effect of short-range corrugation on the adatom mobility.

##### A. Scaling approach

Here we extend the scaling arguments of Ref. [18] for predicting the characteristic time of the convergence of the roughness to its value in the flat substrate. We refer to the scaling exponents of the model for production of the substrate with subindex  $i$  (initial dynamics;  $z_i, \alpha_i, \beta_i$ ) and no index is used in the exponents of the film deposition model (VLDS class;  $z, \alpha, \beta$ ).

Substrate features are related to the production time  $s$ . At this time, Eq. (4) gives a roughness

$$W_i \sim s^{\beta_i}, \quad (13)$$

and Eq. (7) gives a correlation length

$$\xi_{1i} \approx A_i s^{1/z_i}, \quad (14)$$

where  $A_i$  is a model-dependent constant related to substrate production process. If film deposition takes place on an initially flat substrate, the correlation length increases as [20]

$$\xi_{1f} \approx A_f \tau^{1/z}, \quad A_f \sim R^{1/z}, \quad (15)$$

with the VLDS exponent  $z \approx 3.33$  [32]. If the substrate is rough, the correlation length is expected to increase with the same dynamical exponent, as

$$\xi_{1r} \approx A_r \tau^{1/z}. \quad (16)$$

The amplitude  $A_r$  is also related to  $R$ , but it is different from  $A_f$ , as will be discussed in Sec. IV C.

As illustrated in Sec. III, we expect the crossover to the final dynamics to occur when  $\xi_{1r} \sim \xi_{1i}$ . In this situation, the correlations created in the substrate production are replaced by correlations of the film growth dynamics. Matching Eqs. (14) and (16) gives the crossover time  $t_c$  as

$$t_c \sim \left( \frac{A_i}{A_r} \right)^z s^\kappa \sim R^{-\theta} s^\kappa, \quad (17)$$

with

$$\kappa = \frac{z}{z_i} \quad (18)$$

and  $\theta$  obtained by power counting from

$$R^\theta \sim A_r^z. \quad (19)$$

Here note that  $A_i$  does not depend on  $R$  in Eq. (17).

The convergence of the roughness to its value in the flat substrate is determined by the decay of  $\Delta W_{rf}^2(\tau)$ , which scale as Eq. (9) at long times. The expected crossover scaling is

$$\Delta W_{rf}^2 \approx s^c F(x), \quad (20)$$

where  $c$  is a constant and  $F$  is a scaling function of the variable

$$x \equiv \frac{\tau}{t_c}. \quad (21)$$

Immediately after the film deposition begins,  $\tau$  is very small and  $W_r \gg W_f$ . In this case,  $x \ll 1$  and  $F(x) \rightarrow \text{const}$ . Using Eq. (4), we obtain

$$c = 2\beta_i. \quad (22)$$

At long times ( $\tau \gg t_c$ ), the expected to decay of Eq. (9) leads to

$$F(x) \sim x^{-\gamma} \quad (23)$$

for  $x \gg 1$ .

Reference [10] had formerly proposed a scaling form for EW or MH smoothening terms which is equivalent to Eq. (20), with the same scaling of  $t_c$  on the initial correlation length. References [17,18] showed that the value of exponent  $\gamma$  depends on the final dynamics and on the choice of coefficient of the growth equation to be changed. For instance, in changes from an EW growth to another EW growth,  $\gamma = d/2 + 1$  if only the surface tension changes and  $\gamma = d/2$  if only the noise amplitude changes [17]. In many cases, the conditions  $\Delta W \ll W_u$  or  $\Delta W_{rf} \ll W_u$  apply at long times, and a simple time-shift hypothesis in Eq. (4) leads to [18]

$$\gamma_{ts} = 1 - 2\beta. \quad (24)$$

However, this is not a universal result. It works for changes in the noise amplitude of the EW equation but fails for changes only in the surface tension due to a particular cancellation of terms in the difference of square roughness [16].

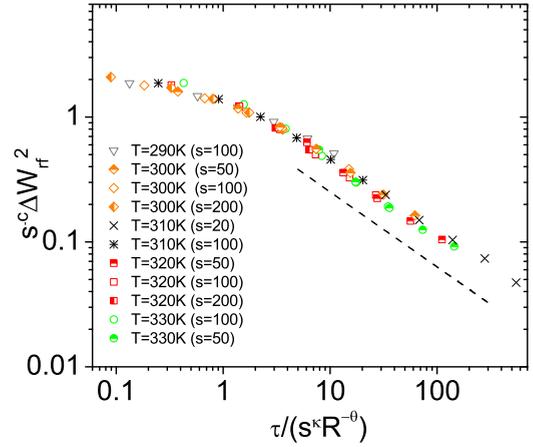


FIG. 7. (Color online) Scaled square roughness change as a function of scaled time for deposition in various temperatures and substrates produced by the E model in different times  $s$ . Scaling exponents are  $c = 0.4$ ,  $\kappa = 2.04$ , and  $\theta = 0.4$ . The dashed line has slope  $-0.6$ .

## B. Numerical results

Figure 7 shows the scaled square roughness change  $s^{-c} \Delta W_{rf}^2$  as a function of the scaled time  $\tau / (s^\kappa R^{-\theta})$ , as suggested by Eq. (20), for deposition at several temperatures and several substrates produced by the E model ( $W_i$  varies from  $\approx 3$  to  $\approx 4$ ). The collapse of data in a single curve was obtained in three steps. First,  $c = 0.4$  was chosen for collapsing the data for short  $\tau$ . This value is slightly below the one predicted by Eq. (22) using  $\beta = 0.24$  [34]. The discrepancy is expected due to corrections to scaling in short time KPZ growth. Subsequently, the value  $\kappa \approx 2.04$  [Eqs. (17), (20), and (21)] was chosen because it is consistent with the dynamical exponent values  $z \approx 3.33$  of the VLDS class [32,33] and  $z \approx 1.63$  of the KPZ class [34]. These two steps lead to collapse the data for the same  $R$  and different  $s$ . The third step was the choice of  $\theta = 0.4$ , which provides the collapse of the data for different  $R$ .

The long crossover observed in Fig. 3 is related to the large value of the crossover exponent  $\kappa$ , which is a consequence of the small dynamical exponent of the substrate production process (KPZ). If the temperature dependence of the correlation length amplitudes  $A_f$  and  $A_r$  was the same [Eq. (15)], then  $\theta = 1$  would be obtained from Eq. (19). However, the numerical estimate of  $\theta$  is very different from this value. This represents a much weaker temperature effect on the roughness change. If the crossover time is written in terms of an effective Boltzmann variable as  $t_c \sim \exp(E_{ef}/k_B T)$ , then we obtain  $E_{ef} = \theta E_a$ . This means that the apparent activation energy is only 40% of the actual activation energy of surface diffusion.

Figures 8(a) and 8(b) show the scaled square roughness change  $s^{-c} \Delta W_{rf}^2$  as a function of the scaled time  $\tau / (s^\kappa R^{-\theta})$  for deposition in several temperatures and substrates produced by the WV and WV10 models, respectively. The estimate  $\kappa = 0.83$  is consistent with the short time MH scaling of the WV model ( $z_i = 4$ ) and VLDS scaling of the film deposition model ( $z \approx 3.33$ ), using Eqs. (17), (20), and (21). It shows that the scaling approach originally proposed in Ref. [18] for

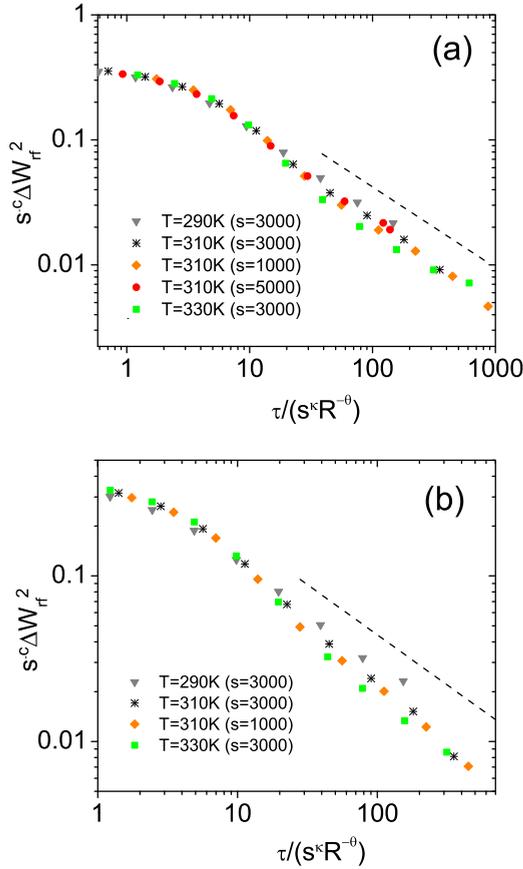


FIG. 8. (Color online) Scaled square roughness change as a function of scaled time for deposition in various temperatures and substrates produced in different times  $s$  by the WV model with (a)  $m = 1$  and (b)  $m = 10$ . Scaling exponents are  $c = 0.4$ ,  $\kappa = 0.83$ , and  $\theta = 0.4$ . The dashed line has slope  $-0.6$

$z > z_i$  (rephrased in Sec. IV A) is also valid for  $z < z_i$ . In this case, the exponent  $\kappa < 1$  explains the rapid crossover observed in Fig. 4.

Finally,  $\theta = 0.4$  is the same exponent used to collapse the data of films grown on the substrates produced by the E model (Fig. 7). This is consistent with a universal scaling of the crossover time on the temperature-like variable  $R$  in conditions of surface smoothening.

The long time decays in Figs. 7, 8(a), and 8(b) have the same exponent  $\gamma \approx 0.6$ . This is consistent with the prediction of the time-shift hypothesis, Eq. (24), which gives  $\gamma_{ts} \approx 0.6$  for this system. This is not an obvious result in the case of substrates produced by the E model, since  $\Delta W_{rf}^2$  is not much smaller than  $W_f^2$ .

A subtle point in this analysis is that the scaling variables can be written independently of the substrate production time  $s$ . The variable  $\Delta W_{rf}^2$  compares the roughness of the films grown in rough and flat substrates for the same deposition time  $\tau$ , and powers of  $s$  may be replaced by the roughness and the correlation length. On the other hand, the variable  $\Delta W^2$  [Eq. (8)] commonly used in works on changes of interface growth dynamics [16–18] is implicitly connected to  $s$ . Using this variable, the best data collapse is obtained with approximately the same exponents  $c$ ,  $\theta$ , and  $\kappa$  for all

substrates. However, the long time slope of the scaling plots are  $\approx -0.6$  for substrates produced by the E model and  $\approx -1.32$  for substrates produced by the WV model (plots not shown here).

The experience with EW-EW changes [16,17] shows that the value of  $\gamma$  may depend on both the initial and the final dynamics, although the time-shift hypothesis disregards the initial one. The apparently nonuniversal exponent  $\gamma$  in the scaling of  $\Delta W^2$  possibly is another example. However, no analytic approach for a KPZ-VLDS change is currently accessible to clarify this point.

### C. Temperature effects

If the thin film is deposited on a flat substrate, the formation of terraces is observed for large  $R$  (high temperatures). The equivalence with a limited mobility deposition model shows that the average lateral size of the terraces is  $l_F \sim R^{1/z}$  [20]. This is schematically illustrated in Fig. 9(a). The corresponding terrace area is of order  $l_F^2$ . Since these terraces are dynamically formed during the growth, the mobile adatoms deposited on them are not expected to nucleate new terraces (or islands); instead, most of these atoms will migrate to the boundaries and attach there. Thus, for simplicity, the scheme of Fig. 9(a) does not represent the mobile adatoms on the terraces.

The rough substrates considered here (produced by E or WV model) may have large correlation lengths, but they are locally rough; thus the lateral size of terraces  $l_i$  is very small. This is illustrated in Fig. 9(b); see also the substrate profile in Fig. 2. The diffusion length of the adatoms on this type of surface is much smaller than the diffusion length on the landscapes of Fig. 9(a) because lateral aggregation is much more frequent. Thus, the propagation of the lateral correlations is slower in the rough substrates.

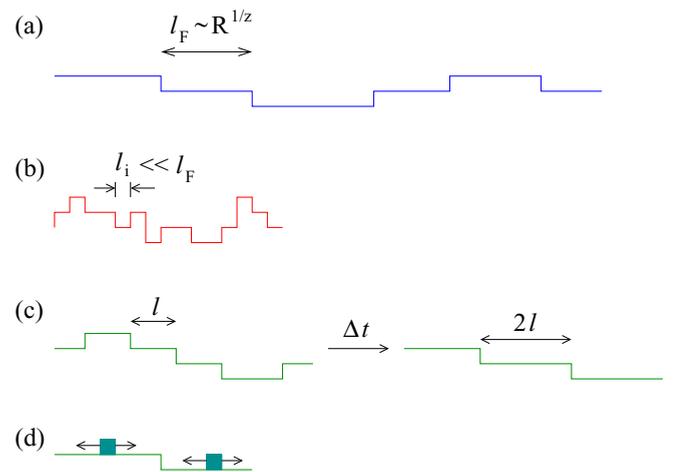


FIG. 9. (Color online) (a) Terrace formation at a high temperature; (b) profile of a locally rough substrate, such as those produced by E or WV model showing a small lateral size of terraces; (c) increase of the lateral size of terraces on the rough substrates in a time interval  $\Delta t$  in which the average size increases from  $l$  to  $2l$ ; (d) adatom diffusion on terraces before reaching the step edges.

In deposition on flat substrates, the correlation length scales as Eq. (15); i.e., it is a function of the time variable  $X \equiv Rt$  [20]. In deposition on rough substrates, the slower correlation involves a rescaled time variable  $Y \equiv X/t_\times$ , as observed in other systems with crossovers between different universality classes. A typical example is the competition of uncorrelated deposition and some limited mobility model (e.g., the E and WV models), in which  $t_\times$  is the time necessary to correlate neighboring column heights [38]. In the present case, the rescaling factor  $t_\times$  is the time necessary for the formation of the same structures that appear in the growth on flat substrates, i.e., the time of formation of the terraces shown in Fig. 9(a). The example of Fig. 2 confirms the trend of formation of terraces during film deposition on a rough substrate.

Figure 9(c) shows the coarsening of terraces in a time interval  $\Delta t$ , in which the average size increases from  $l$  to  $2l$ , with  $l < l_F$ . The area of a terrace increases or decreases by a total amount of order  $\Delta\mathcal{A} \sim l^2$  during this time. This is a consequence of random attachments of adatoms to steps at the same level or below. Those adatoms are deposited on neighboring terraces and diffuse on them until reaching the step edges, as shown in Fig. 9(d). For  $l < l_F$ , as explained above, the nucleation of new terraces by those mobile adatoms can be neglected.

For the above reasons, the area of a terrace changes as a random walk with unit steps. At a given terrace, the addition of a single adatom takes place in a time of order  $t_1 \sim 1/\Delta\mathcal{A}$  in units of monolayers (or  $1/F$  units). Thus, the random increase or decrease of its area by one unit takes place as a random walk with diffusion coefficient  $D_A \sim 1/t_1 \sim l^2$ . For the terrace area in Fig. 9(c) to increase  $\Delta\mathcal{A}$ , we have  $(\Delta\mathcal{A})^2 \sim D_A \Delta t$ , which gives  $\Delta t \sim l^2$ . Thus, since  $t_\times$  is the time interval for formation of the terraces of size  $l_F$ , we obtain

$$t_\times \sim l_F^2 \sim R^{2/z} \quad (25)$$

(coarsening of terraces with  $l \ll l_F$  takes a much smaller time, thus it does not contribute to the scaling of  $t_\times$ ).

The rescaled time variable for deposition on the rough substrate is

$$Y \equiv \frac{X}{t_\times} \sim R^{1-2/z} t. \quad (26)$$

The correlation length scaling involves the VLDS exponent  $z$  as

$$\xi_{1r} \sim Y^{1/z} \sim (R^{1-2/z} t)^{1/z}. \quad (27)$$

Using the VLDS exponent  $z \approx 3.33$ , Eqs. (19) and (27) give

$$\theta = 1 - 2/z \approx 0.4. \quad (28)$$

This result is in excellent agreement with the numerical estimate obtained by data collapse in Sec. IV B.

## V. SCALING LAW FOR SMOOTHENING

The scaling features of the height differences defined in Eqs. (8) or (10) is very difficult to be tested experimentally because it requires independent measurements for deposition on two different substrates: a flat one and a rough one. For applications, it is generally not interesting to perform this

type of comparison; instead, the main interest is usually to understand the roughness scaling of films deposited on substrates of different materials of technological relevance. For this reason, here we show how the crossover scaling can be expressed without quantities characterizing the ideal case of deposition on flat substrates. In other words, we look for a scaling law for smoothening, analogously to Eq. (4), which applies for roughening.

Figures 3 and 4 show that the roughness of the substrate and of the films deposited on it at short times are much larger than the roughness of films deposited on the flat substrate. This is the typical behavior for  $\tau \lesssim t_c$ . The corresponding differences in the square roughness [Eqs. (8) or (10)] are dominated by the roughness of the film deposited on the rough substrate, so that  $\Delta W_{rf}^2(\tau) \approx W_r^2(\tau)$ . From Eqs. (13) and (22), the substrate roughness is  $W_i \sim s^{c/2}$ , thus the scaled roughness difference shown in Figs. 7 and 8 is approximately

$$r_w(\tau) \equiv \frac{W_r^2(\tau)}{W_i^2}. \quad (29)$$

This is simply the ratio between the current and the initial square roughness.

Now consider the crossover time in the scaling relation (20), with definitions in Eqs. (17) and (21). That time may be written as

$$t_{c1} \equiv R^{-\theta} \xi_{1r}^z \quad (30)$$

or as

$$t_{c2} \equiv R^{-\theta} W_i^a, \quad a \equiv \frac{z}{\alpha_i}, \quad (31)$$

which depend on the initial correlation length or the initial roughness, respectively. These quantities can be calculated independently of the knowledge of the substrate production process. Note that both crossover times involve exponents of the film deposition process, but the latter [Eq. (30)] also involves an exponent of the substrate production process ( $\alpha_i$ ).

These results imply that the reduced roughness  $r_w$  is a function of  $\tau/t_{c1}$  or  $\tau/t_{c2}$  for short times, in which  $r_w$  is not very small:

$$r_w = \Psi(\tau/t_{c1}) \quad (32)$$

or

$$r_w = \Phi(\tau/t_{c2}), \quad (33)$$

with scaling functions  $\Psi$  and  $\Phi$ .

For deposition on substrates produced by the E model, Figs. 10(a) and 10(b) show  $r_w$  as a function of  $\tau/t_{c1}$  and  $\tau/t_{c2}$ , respectively. Those plots consider  $\theta = 0.4$  and the values of  $z$  ( $t_{c1}$ ) and  $a$  ( $t_{c2}$ ) that give the best data collapses. In Fig. 10(a) the VLDS and KPZ estimates  $z \approx 3.33$  [32,33] and  $\alpha_i \approx 0.39$  [34] give  $a = 8.5$  in Eq. (31), which is only 2.4% larger than the above numerical estimate. In Fig. 10(b) the numerical estimate of  $z$  is very close to the VLDS value; this result and the independence of  $t_{c1}$  on substrate production exponents are two important advantages of the scaling variable  $\tau/t_{c1}$ .

Figures 11(a) and 11(b) show  $r_w$  as a function of  $\tau/t_{c1}$  and  $\tau/t_{c2}$ , respectively, for deposition on substrates produced by the WV model. In this case, the values of  $a$  and  $z$  that provide the best data collapses are consistent with the VLDS exponent

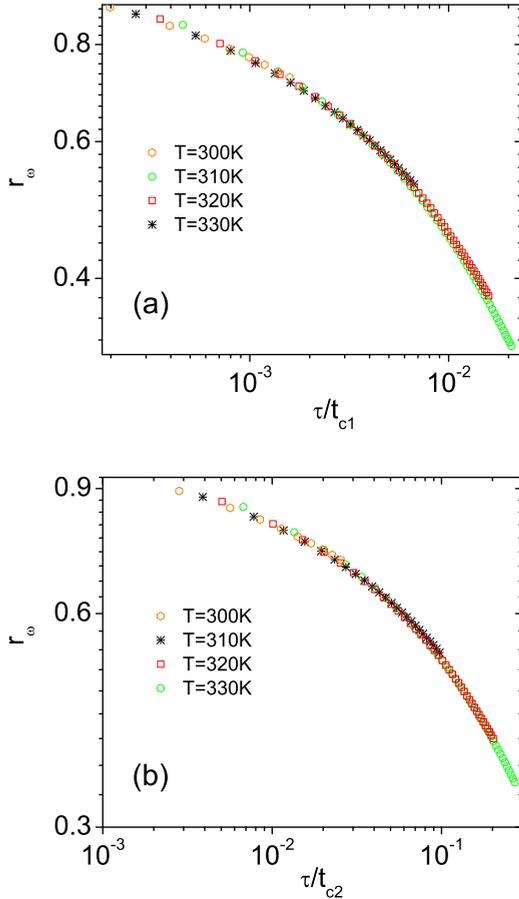


FIG. 10. (Color online) Reduced roughness as a function of (a)  $\tau/t_{c1}$  and (b)  $\tau/t_{c2}$  for deposition on substrates produced by the EV model. The scaling exponents are  $\theta = 0.4$ ,  $a = 8.3$ , and  $z = 3.33$ .

$z \approx 3.33$  and the MH exponent  $\alpha_i = 1$  (which is the effective scaling of WV model in short times).

The data collapse in the above plots show smoothening by factors between 3 and 5, which represent significant changes in the film roughness. The ranges of scaled times varied between 1.5 and 2 orders of magnitude. These results strongly suggest experimental tests of relations (32) and (33) (particularly the former) for smoothening in film growth on rough substrates.

## VI. CONCLUSION

We studied the evolution of surface roughness during thin film deposition on a rough substrate using a model of temperature-activated adatom diffusion, irreversible lateral aggregation, and no additional step energy barrier. Previous works on deposition on flat substrates showed that this model was in the VLDS class and that the roughness was not very different from that of the (reversible) Clarke-Vvedensky model if the temperature is not very high.

At very low temperatures, the roughness monotonically increases because the average number of adatom steps after adsorption is very small, which leads to an approximately uncorrelated deposition. If the temperature is not very low, smoothening takes place at short times. The expected long time

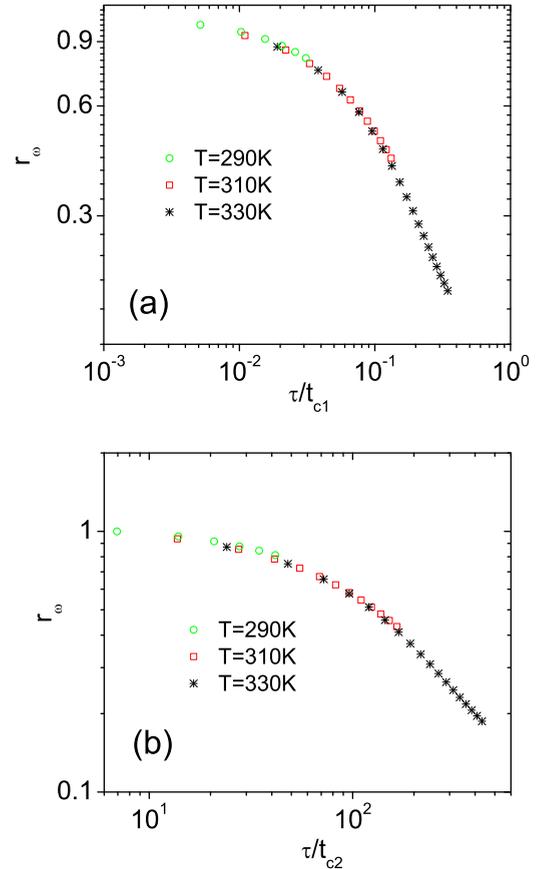


FIG. 11. (Color online) Reduced roughness as a function of (a)  $\tau/t_{c1}$  and (b)  $\tau/t_{c2}$  for deposition on substrates produced by the WV model with  $m = 1$ . The scaling exponents are  $\theta = 0.4$ ,  $a = 1.0$ , and  $z = 3.33$ .

VLDS roughening is observed only after a crossover time that increases with the correlation length of the initial rough pattern. A scaling approach is used to predict the relation between the crossover time and the time of substrate production. The corresponding scaled time provides excellent collapse of numerical data for the difference in the roughnesses of films growing on rough and flat substrates. These results extend those of previous works on KPZ-EW changes of interface growth dynamics but suggest the use of the film deposition time in the roughness difference and in the scaled time instead of the total interface growth time, which includes substrate production. The effect of temperature on the crossover time cannot be predicted by a direct extension of the results on flat substrates because the initial rough substrate leads to a delay in the formation of terraces and the consequent propagation of correlations. A scaling approach is able to capture these corrections and predict a relation between the crossover time and the diffusion-to-deposition ratio  $R$  in agreement with the numerical data.

We also suggest a scaling law for smoothening, in which the surface roughness and the deposition time are scaled by factors that depend only on the initial roughness and initial correlation length. It provides good data collapse for smoothening by a factor up to 5, depending on the substrate and temperature, and for more than one decade in time. This

scaling may be experimentally tested without information on the film deposition on flat substrates nor information on substrate production time, which is an advance over the scaling relations formerly proposed for sudden changes in interface growth dynamics [16–18].

#### ACKNOWLEDGMENTS

F.D.A.A.R. acknowledges support from CNPq and FAPERJ (Brazilian agencies). T.A.d.A. also acknowledges support from CNPq under Grant No. 150874/2014-6.

- 
- [1] M. Ohring, *Materials Science of Thin Films: Deposition and Structure*, 2nd ed. (Academic Press, New York, 2001).
- [2] A. L. Barabási and H. E. Stanley, *Fractal Concepts in Surface Growth* (Cambridge University Press, Cambridge, 1995).
- [3] J. Krug, *Adv. Phys.* **46**, 139 (1997).
- [4] J. Lapujoulade, *Surf. Sci. Rep.* **20**, 195 (1994).
- [5] Z. Jiang and C. Ebner, *Phys. Rev. B* **40**, 316 (1989).
- [6] M. V. Ramana Murty and B. H. Cooper, *Phys. Rev. B* **54**, 10377 (1996).
- [7] S. Bustingorry and P. M. Centres, *Phys. Rev. E* **84**, 011613 (2011).
- [8] M. S. Hoogeman, M. A. J. Klik, R. van Gastel, and J. W. M. Frenken, *J. Phys.: Condens. Matter* **11**, 4349 (1999).
- [9] J. Erlebacher, M. J. Aziz, E. Chason, M. B. Sinclair, and J. A. Floro, *Phys. Rev. Lett.* **84**, 5800 (2000).
- [10] S. Majaniemi, T. Ala-Nissila, and J. Krug, *Phys. Rev. B* **53**, 8071 (1996).
- [11] T. T. T. Nguyen, D. Bonamy, L. Phan Van, J. Cousty, and L. Barbier, *Europhys. Lett.* **89**, 60005 (2010).
- [12] S. F. Edwards and D. R. Wilkinson, *Proc. R. Soc. London* **381**, 17 (1982).
- [13] W. W. Mullins, *J. Appl. Phys.* **28**, 333 (1957); C. Herring, in *The Physics of Powder Metallurgy*, edited by W. E. Kingston (McGraw-Hill, New York, 1951), pp. 143–179.
- [14] J. Krug and M. Rost, *Phys. Rev. B* **60**, R16334 (1999).
- [15] C. Castellano and J. Krug, *Phys. Rev. B* **62**, 2879 (2000).
- [16] Y.-L. Chou, M. Pleimling, and R. K. P. Zia, *Phys. Rev. E* **80**, 061602 (2009).
- [17] Y.-L. Chou and M. Pleimling, *J. Stat. Mech.* (2010) P08007.
- [18] T. A. de Assis and F. D. A. Aarão Reis, *Phys. Rev. E* **89**, 062405 (2014).
- [19] M. Kardar, G. Parisi, and Y.-C. Zhang, *Phys. Rev. Lett.* **56**, 889 (1986).
- [20] F. D. A. Aarão Reis, *Phys. Rev. E* **81**, 041605 (2010).
- [21] S. Clarke and D. D. Vvedensky, *J. Appl. Phys.* **63**, 2272 (1988).
- [22] J. W. Evans, P. A. Thiel, and M. C. Bartelt, *Surf. Sci. Rep.* **61**, 1 (2006).
- [23] T. A. de Assis and F. D. A. Aarão Reis, *J. Stat. Mech.* (2015) P06023.
- [24] J. Villain, *J. Phys.* **11**, 19 (1991).
- [25] Z.-W. Lai and S. Das Sarma, *Phys. Rev. Lett.* **66**, 2348 (1991).
- [26] B. A. Mello, A. S. Chaves, and F. A. Oliveira, *Phys. Rev. E* **63**, 041113 (2001).
- [27] D. E. Wolf and J. Villain, *Europhys. Lett.* **13**, 389 (1990).
- [28] P. P. Chatrathorn, Z. Toroczkai, and S. Das Sarma, *Phys. Rev. B* **64**, 205407 (2001).
- [29] S. Das Sarma, P. P. Chatrathorn, and Z. Toroczkai, *Phys. Rev. E* **65**, 036144 (2002).
- [30] D. Siniscalco, M. Edely, J. F. Bardeau, and N. Delorme, *Langmuir* **29**, 717 (2013).
- [31] F. D. A. Aarão Reis, *J. Stat. Mech.* (2015) P11020.
- [32] F. D. A. Aarão Reis, *Phys. Rev. E* **70**, 031607 (2004).
- [33] H. K. Janssen, *Phys. Rev. Lett.* **78**, 1082 (1997).
- [34] J. Kelling and G. Ódor, *Phys. Rev. E* **84**, 061150 (2011).
- [35] J. Krug, M. Plischke, and M. Siegert, *Phys. Rev. Lett.* **70**, 3271 (1993).
- [36] M. Siegert, *Phys. Rev. E* **53**, 3209 (1996).
- [37] A. Chame and F. D. A. Aarão Reis, *Surf. Sci.* **553**, 145 (2004).
- [38] F. D. A. Aarão Reis, *Phys. Rev. E* **73**, 021605 (2006).