Effects of a kinetic barrier on limited-mobility interface growth models

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The role played by a kinetic barrier originated by out-of-plane step edge diffusion, introduced by Leal *et al.* [J. Phys.: Condens. Matter **23**, 292201 (2011)], is investigated in the Wolf-Villain and Das Sarma-Tamborenea models with short-range diffusion. Using large-scale simulations, we observe that this barrier is sufficient to produce growth instability, forming quasiregular mounds in one and two dimensions. The characteristic surface length saturates quickly indicating a uncorrelated growth of the three-dimensional structures, which is also confirmed by a growth exponent $\beta = 1/2$. The out-of-plane particle current shows a large reduction of the downward flux in the presence of the kinetic barrier enhancing, consequently, the net upward diffusion and the formation of three-dimensional self-assembled structures.

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I. INTRODUCTION

A rich variety of morphologies can be observed during far-from-equilibrium growth processes and many of them with a potential for technological applications [1–4]. Growth instability can induce three-dimensional mound-like patterns in different types of films such as metals [5–7], inorganic [8,9] and organic [10,11] semiconductor materials to cite only a few examples. Such a growth instability has been mainly attributed to the presence of Ehrlich-Schwoebel (ES) step barriers [12,13] that reduce the rate with which atoms move downwardly on the edges of terraces leading to net uphill flows. Growth instabilities can also emerge from topologically induced uphill currents which depend on the crystalline structure [14] or from fast diffusion on terrace edges [15,16], among other mechanisms [1,2]. The existence of ES barriers is supported by molecular dynamic simulations [17].

Discrete solid-on-solid (SOS) growth models constitute an important approach to investigate the dynamic of kinetic roughening and morphological properties of interfaces. The rules are easily implemented in a discrete space (lattices) rid of overhangs and bulk voids. The role played by ES barriers has been investigated in models with thermally activated diffusion [1,2], the Clark-Vvedenski (CV) model [18,19] being one of the simplest examples, in which any surface adatom can move according to an Arrhenius diffusion coefficient $D \sim \exp(-E/k_BT)$ [3], where E is an energy activation barrier to be overcome in a diffusion hopping. An ES barrier can be included as an additional activation energy for diffusion at the edges of terraces [2]. The effects of a step barrier of purely kinetic origin, namely simple diffusion, were investigated in an epitaxial growth model with thermally activated diffusion [20]. In this model, a particle performing an interlayer movement through steps with more than one monolayer has to diffuse along the columns, perpendicularly to the substrate, instead of attaching directly at the bottom or top of a terrace. This kinetic barrier reduces downhill currents and three-dimensional structures in the form of mounds are obtained at short-time scales even in the case of weak ES barriers where the conventional rule would not lead to mound formation.

Simple models with limited mobility can be used to investigate kinetic roughening [3,4]. Wolf-Villain (WV) [21] and Das Sarma-Tamborenea (DT) [22] models, introduced to investigate molecular-beam-epitaxy (MBE) growth, are benchmarks of this class and have been intensively investigated [23–32]. A variation of the CV model with limited mobility has been considered [33,34] and many features of the original model have been reproduced with this simplified version [35]. Effects of a step barrier were investigated in both WV [36] and DT [37] models introducing two additional probabilities for downward and upward interlayer diffusion with the former larger than the latter, and mound formation was observed in both models. WV and DT models without step barrier were investigated in several lattices [14,38] and it was found that the WV model can present topologically induced mound morphologies on some lattices but not in others while no clear evidence for three-dimensional structures was observed for DT. In one dimension, it is widely accepted that both DT and WV models asymptotically produce self-affine surfaces belonging to nonlinear MBE [32] and Edwards-Wilkinson [39] universality classes, respectively.

It was reported that a kinetic barrier alone does not induce mound morphologies in thermally activated CV-like models [20] but, instead, they exhibit kinetic roughening with exponents consistent with the nonlinear MBE universality class [22,40,41]. Therefore, given the simplicity of limited-mobility growth models and the non-trivial effects of topologically induced uphill currents in DT and WV models, one would

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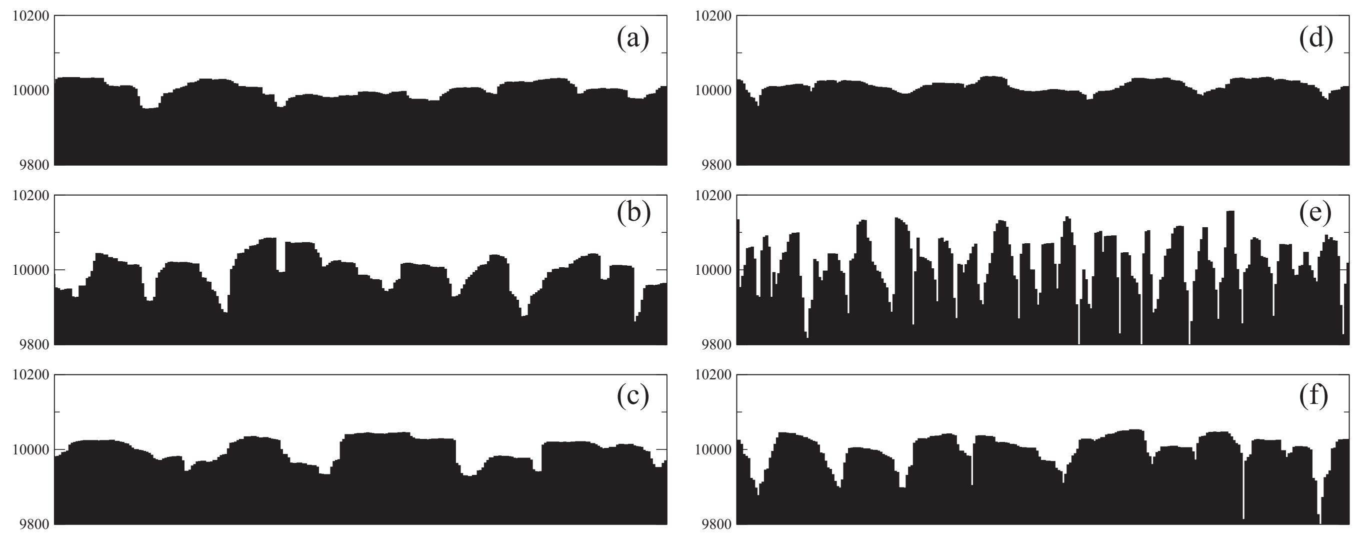

FIG. 1. Interfaces of the WV and DT models in $d = 1$ shown in left and right panels, respectively. Cases (a,d) without and with the kinetic barrier considering (b,e) $N_s = 1$ and (c,f) $N_s = 10$ are shown. All the simulations were done on a lattice of size $L = 2^{10}$ for a deposition time $t = 10^4$.

wonder how they respond to a barrier of purely kinetic origin. In order to fill this gap, we investigate WV and DT models with the introduction of the kinetic barrier proposed in Ref. [20]. We observed mounds in both models in 1+1 and 2+1 dimensions, being more evident for the WV model. The surface coarsening ceases quickly with the saturation of the characteristic surface length and regimes of uncorrelated mound growth are asymptotically observed. Analysis of the out-of-plane currents shows a large reduction of the downhill flux of particles, enhancing surface instabilities and mound formation.

The rest of the paper is organized as follows. The model implementation details are presented in Sec. II. In Sec. III, we discuss the results obtained in the simulations. Our conclusions and some perspectives are drawn in the Sec. IV.

II. MODELS

In all investigated models, the particles are randomly deposited on a d-dimensional lattice of linear size L with periodic boundary conditions under the SOS condition. Results presented in this work correspond to regular chains in $d = 1$ and square lattices in $d = 2$. Other lattices were tested and the central conclusions remain unaltered. The height of the interface at site i and time t is represented by $h_i(t)$ and the initial condition is given by $h_i(0) = 0$ such that the initial interface is flat.

In the WV model with a kinetic barrier investigated in the present work, the growth rule is implemented as follows. At each time step, a position i is randomly chosen. A location i' with the largest number of bonds that a new deposited adatom would have is determined within a set containing i and its nearest neighbors. If the initial position corresponds to the largest number of bonds ($i' \equiv i$), it is chosen as the deposition place and the simulation runs to the next step. In case of multiple options, one is chosen at random. Otherwise, the particle tries to diffuse to the neighbor i' with a probability given by [20]

$$P_{\delta h}(i, i') = \begin{cases} 1, & \text{if } |\delta h| < 2 \\ \frac{1}{|\delta h|}, & \text{if } |\delta h| \geqslant 2 \end{cases}, \tag{1}$$

where $\delta h = h_i - h_{i'}$. With probability $1 - P_{\delta h}(i, i')$ the particle remains at the site i. It is important to mention that Eq. (1) is obtained assuming that the adatom first moves to the top kink of the terrace and then start a unbiased one-dimensional random-walk perpendicularly to the initial substrate, stopping the movement if it either arrives at the bottom or returns to top of the terrace. The result is the solution of a non-directed one-dimensional random walk with absorbing boundaries separated by a distance $|\delta h|$ [42]; see Fig. 1 of Ref. [20] for further details of this diffusion rule. This diffusion attempt is successively applied N_s times departing from the last position of the adatom. A time unit is defined as the deposition of L^d particles.

The implementation of the DT model with kinetic barrier is similar. The difference is that diffusion to the nearest neighbors is performed only if the adatom does not have lateral bounds and any neighbor with a number of bonds higher than 1 can be chosen with equal chance as the target site.

III. RESULTS

The one-dimensional simulations were carried out on chains with up to $L = 2^{14}$ sites and evolution times of up to $t = 10^7$. In the two-dimensional case, the simulations were done in systems of sizes up to $L = 2^{10}$ and times up to $t = 10^6$. The averages were performed over 100 independent runs.

Figures 1 and 2 show interfaces obtained in simulations in one- and two-dimensional substrates, respectively. Surfaces for the original WV and DT models without and with ($N_s = 1$ or $N_s = 10$) kinetic barriers are compared. In both dimensions, the irregular morphologies without a characteristic length observed in the original versions change to structures separated by valleys that present a well-defined characteristic

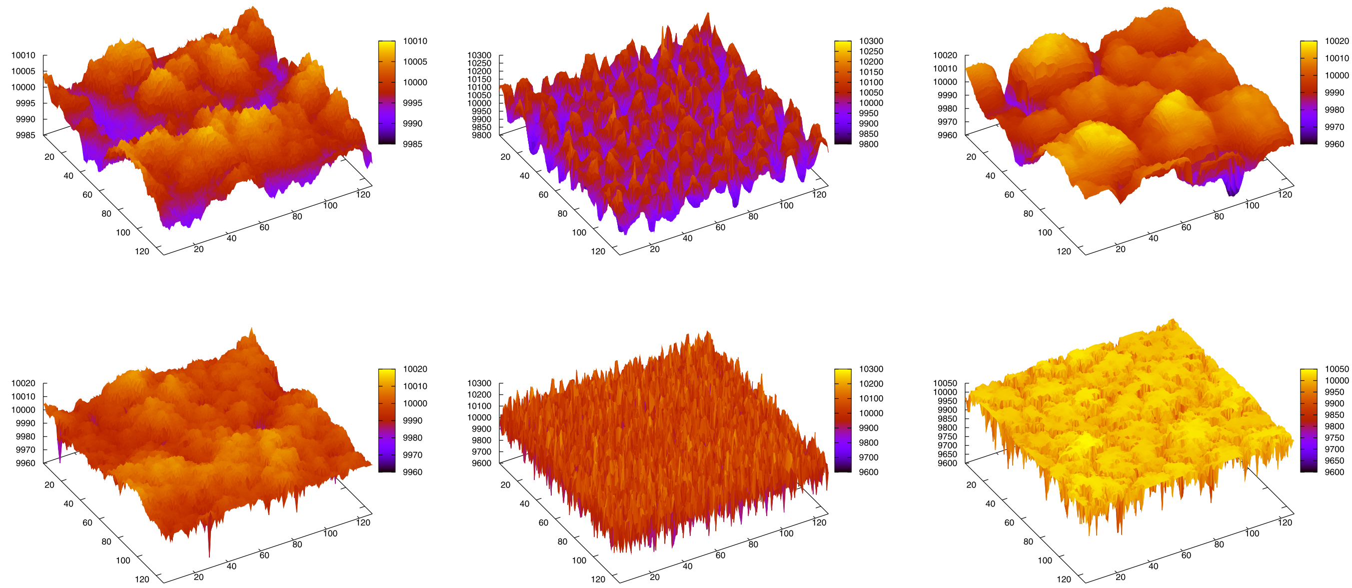

FIG. 2. Interfaces obtained using the WV and DT models in $d = 2$ are shown in top and bottom panels, respectively. The case without (left) and with the kinetic barrier considering $N_s = 1$ (center) and $N_s = 10$ (right) are shown. All simulations were done on square lattices of size $L = 2^9$ and a deposition time $t = 10^4$.

length. We also observe that an increase in the value of N_s reduces valley deepness and increases the characteristic width of the mounds. The effects of the kinetic barrier seem to be stronger in two than one dimension. A remarkable change in the profiles happens when just one hop to nearest neighbors is allowed in the DT model with kinetic barrier, as can been seen in Fig. 1(e). Surfaces become columnar with a high aspect ratio (height/width). Such a behavior is reminiscent of the very strict rule for diffusion in the DT model when a single lateral bound is enough to irreversibly stick the adatom on a site. In the WV case, where diffusion happens more readily, mound morphologies with quasiregular structures emerge more clearly.

A standard tool to characterize the morphology of interfaces in growth process is the height-height correlation function defined as [2,15,38]

$$\Gamma(\mathbf{r}) = \langle \tilde{h}(\mathbf{x})\tilde{h}(\mathbf{x}+\mathbf{r})\rangle_x. \tag{2}$$

Here, $\tilde{h}(\mathbf{x})$ is the height interface at position $\mathbf{x}$ relative to the mean height and $\langle\ldots\rangle_x$ denotes an average over the surface. The height-height correlation for $\mathbf{r} = 0$ is related to the interface width by

$$\sqrt{\langle\Gamma(0)\rangle} = w. \tag{3}$$

Here, $\langle\ldots\rangle$ denotes an average over independent runs. A self-affine interface is characterized by a height-height correlation function that goes monotonically to zero while those characterized by mounds exhibit oscillatory behavior around 0. In the latter case, the first zero of $\Gamma(r)$, denoted by ξ, is a characteristic lateral length of the mounds in the surface.

Figure 3 shows the height-height correlation function for the WV model with kinetic barrier in one- and two-dimensional substrates. The curves clearly exhibit oscillatory behavior even for averages over 100 independent samples. Conversely, the irregular oscillatory behavior observed for the original WV model shown in the insets of Fig. 3 is lumped after averaging. Therefore, interfaces obtained with kinetic barrier are characterized by the formation of quasiregular mound structures differently from those obtained using the original model that exhibits irregular structures within the intervals of size and time we investigated. These plots also show a coarsening of the mounds represented by the first minimum displacement at the early growth times.

The effect of the parameter N_s in WV model is shown in Fig. 4. As indicated by the interface profiles shown in Figs. 1 and 2, the characteristic lateral length increases with N_s in both dimensions. The correlation function for the DT model follows a qualitative similar dependence with N_s, as can be seen in Fig. 5 where the effects of time and number of diffusion steps in the correlation function of the DT model are shown. However, the mounds are much less evident than those obtained in the WV model. However, the correlation functions still present the typical oscillatory behavior of mounded structures that is preserved after the averaging over 100 independent samples. Besides, the typical widths of the mounds in the DT model are much smaller than those of WV. It is important to note that the correlation function of the original DT model also presents an irregular oscillatory behavior as does the WV model.

Figure 6 shows the time evolution of the interface width for both models in one and two dimensions. The main panels and insets present the results for the WV and DT models, respectively, including or not the kinetic barrier. The interface width is expected to scale as $w \sim t^\beta$, where β is the growth exponent [3]. The short time dynamics of both WV and DT models is well described by the linear version of the MBE equation [40,41]

$$\frac{\partial h}{\partial t} = -\nu\nabla^4 h + \lambda\nabla^2(\nabla h)^2 + \eta \tag{4}$$

with $\lambda = 0$ where η is a non-conservative Gaussian noise [40,41,43]. This result is confirmed in Fig. 6 where the

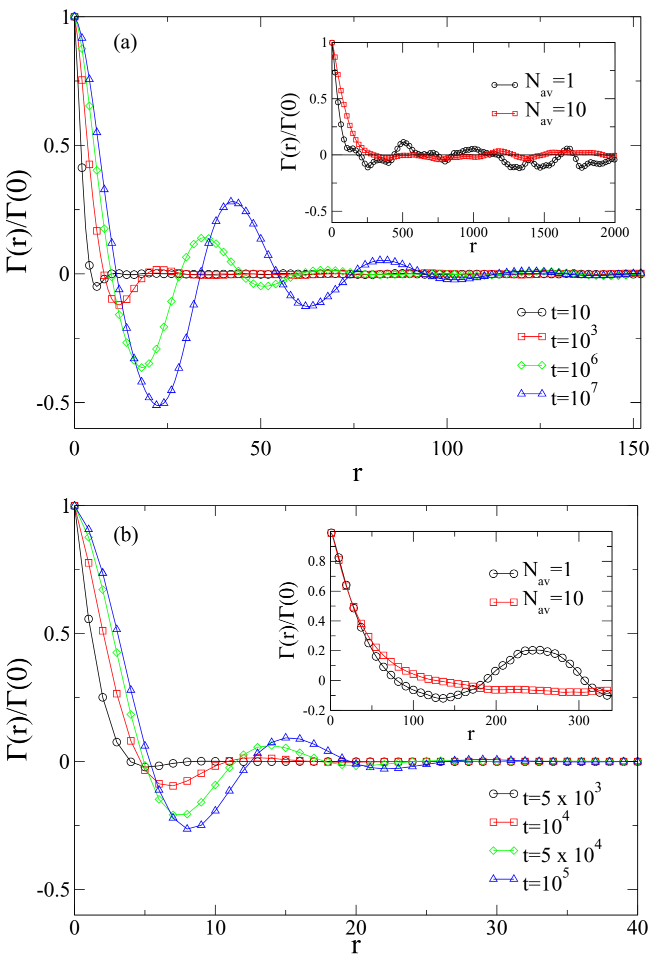

FIG. 3. Main panels: Height-height correlation function for the WV model with kinetic barrier at distinct times indicated in the legends for (a) one- and (b) two-dimensional substrates. The number of steps is $N_s = 1$. The averages were computed over $N_{\mathbf{av}} = 100$ independent runs. Insets: Correlation functions averaged over $N_{\mathbf{av}} = 1$ and 10 samples for the original WV model at time $t = 10^5$ showing that the oscillations observed in single samples are not due to regular structures.

short time behavior is consistent with the growth exponents $\beta = 3/8$ in $d = 1$ and $\beta = 1/4$ in $d = 2$ expected for the linear MBE universality class [3]. It is worth mentioning that these models may undergo crossovers to different universality classes in the asymptotic limit, depending on the dimension and model [29–31,39,44,45]. The curves in Fig. 6 are consistent with crossovers to different universality classes at long times. One expects that original DT is asymptotically consistent with the non-linear MBE equation with $\lambda > 0$ [31,32,46], for which $\beta \approx 1/3$ and $1/5$ in $d = 1$ and $d = 2$, respectively,[1] while crossovers to the Edwards-Wilkinson universality class with $\beta = 1/4$ in $d = 1$ and $\beta = 0$ (logarithmic

[1]The exponents $\beta = 1/3$ and $1/5$ are predictions of the one-loop renormalization group [40,41]. Two-loop calculations [47], however, predict corrections where the growth exponents are slightly smaller than these values.

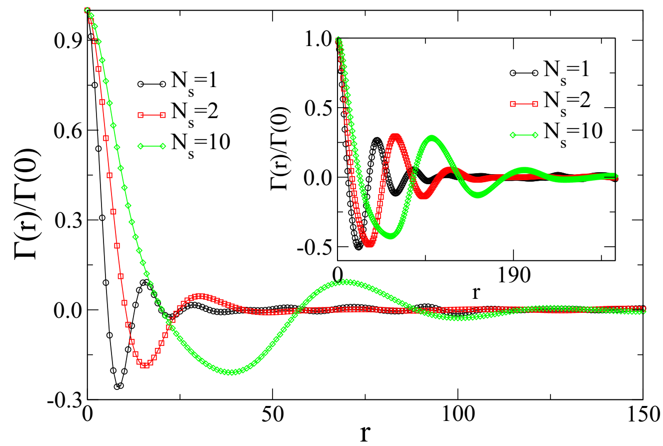

FIG. 4. Main plot: Height-height correlation function dependence with the parameter N_s (indicated in the legends) for the WV model with kinetic barrier in two-dimensional substrates at a time $t = 10^5$. Inset: Same as the main plot for one dimension at a time $t = 10^7$. Curves correspond to averages over 100 independent samples.

growth) in $d = 2$ are expected for the original WV model [30,39]. The simulations with the kinetic barrier, however, depart from the original dynamics after a transient which increases with the diffusion of particles. For long times, an evolution consistent with an uncorrelated growth described by $\frac{\partial h}{\partial t} = \eta$, characterized by a growth exponent $\beta = 1/2$ [3], is observed. This observation can be rationalized as follows. At long times, mounds interact weakly since the kinetic barrier reduces drastically the inter-mound diffusion. Consider the idealized case of plateaus of size L_0 with infinity barriers at their edges. A particle initially adsorbed on the top of a plateau will never slide down to its bottom. So, the probability that this plateau receives R particles after one unity of time (deposition

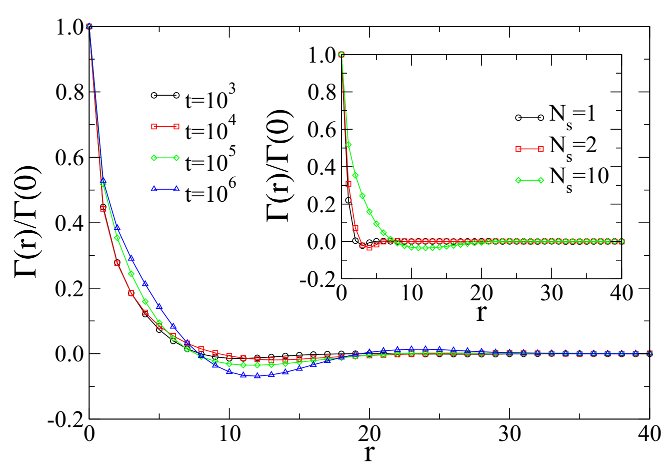

FIG. 5. Main plot: Correlation function for the DT model in two-dimensional substrates for distinct times shown in the legends and fixed $N_s = 10$. Inset: Correlation function for DT model with kinetic barrier in two dimensions at a fixed time $t = 10^5$ and different values of N_s shown in the legends. Curves correspond to averages over 100 independent samples.

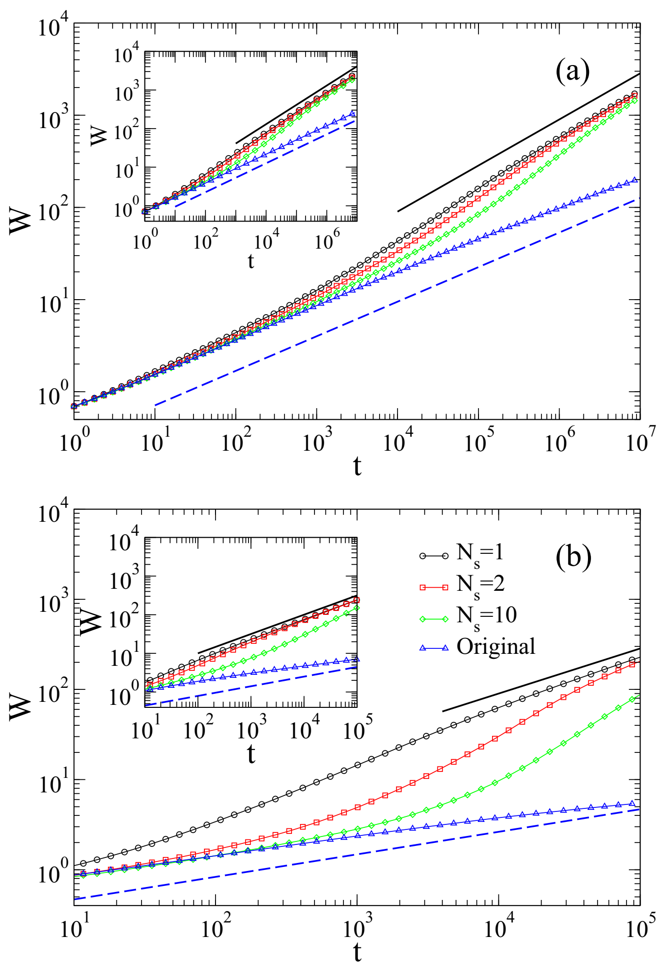

FIG. 6. Time evolution of the interface width w for WV (main panels) and DT (insets) models grown on (a) one- and (b) two-dimensional substrates. Both simulations with the kinetic barrier (using N_s values indicated in the legend) and the original version are shown. In (a), dashed and solid lines are power-laws with exponents 3/8 and 1/2, respectively, in both main panels and insets. In (b), the exponents of the dashed and solid lines are 1/4 and 1/2, respectively.

of L particles) is a binomial distribution

$$P(R) = \binom{L}{R} p^R (1-p)^{L-R} \simeq \frac{1}{\sqrt{2\pi L_0}} e^{-\frac{(R-L_0)^2}{2L_0}}, \qquad (5)$$

where $p = L_0/L$ is the probability that a particle is deposited on this terrace and $1 \ll L_0 \ll L$ is assumed in the Gaussian limit in the right-hand side of Eq. (5). We argue that this situation is similar to the weakly interacting mound observed in our simulations.

In addition, as can be seen in Fig. 7, the characteristic lateral lengths of simulations with kinetic barrier saturate after an initial transient in values that increase with the parameter N_s while the models without barrier present coarsening with $\xi \sim t^{1/z}$ [3]. The saturation implies that the aspect ratio (height/width) of the mounds remains increasing with time and the surface does not present slope selection forming columnar growth. This property is also reflected in the asymptotic interface width scaling as $w \sim t^{1/2}$. As explained previously, it can be interpreted as an uncorrelated evolution

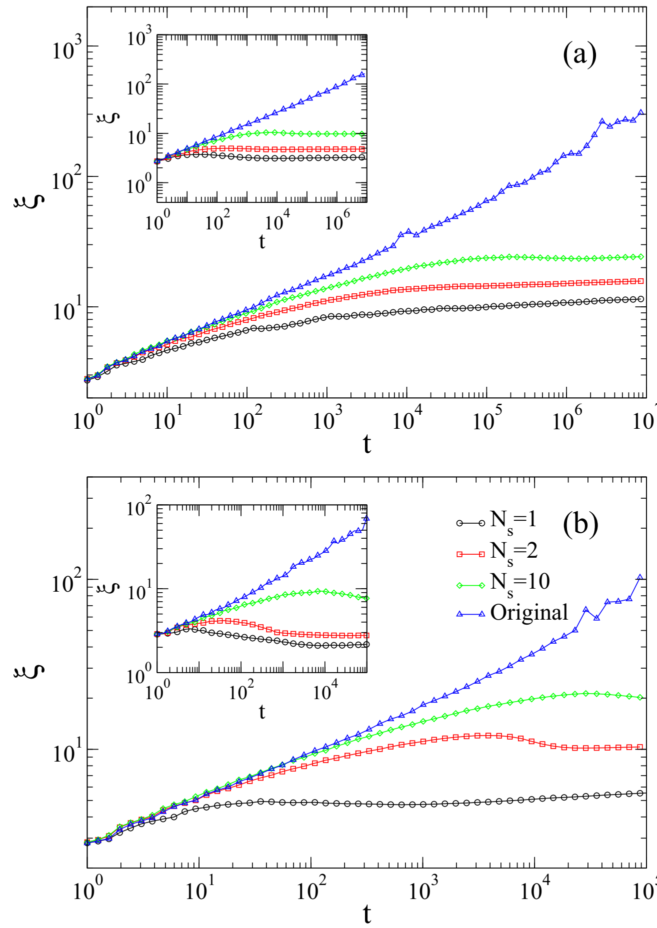

FIG. 7. Characteristic length of mounds ξ for WV (main plots) and DT (insets) models with and without the kinetic barrier in (a) one- and (b) two-dimensional substrates for different values of the parameter N_s indicated in the legend.

of the columns, in which the 1/2 exponent comes out. The results shown in the insets of Figs. 6 and 7 corroborate that the DT model presents the same behavior of the WV model despite of the mounds are less evident in the former.

Instability and mound formation can be investigated considering the surface currents [48,49]; see [50] for details. In this work, we investigated the out-of-plane component of the current defined as [51]

$$J_z = \frac{1}{N} \sum_{(i,j)} \mathrm{sgn}(\delta h) D(i,j) P_{\delta h}(i,j), \qquad (6)$$

where $\mathrm{sgn}(x) = 1$ for $x > 0$, $\mathrm{sgn}(x) = -1$ for $x < 0$, and $\mathrm{sgn}(0) = 0$ is the definition of the sign function, $P_{\delta h}(i,j)$ is given by Eq. (1), and $D(i,j)$ is the rate of hopping attempts from site i to j and depends on the investigated model. The sum runs over all N pairs of nearest neighbors of the lattice. Let n_i be the number of lateral bonds of site i and $n_i^{\max}$ the largest number of bonds among the nearest neighbors of i. For the WV model, $D(i,j)$ is given by

$$D(i,j) = \begin{cases} 1/q_i^{\mathrm{WV}}, & \text{if } n_j = n_i^{\max} \text{ and } n_i < n_i^{\max} \\ 0, & \text{otherwise} \end{cases}, \qquad (7)$$

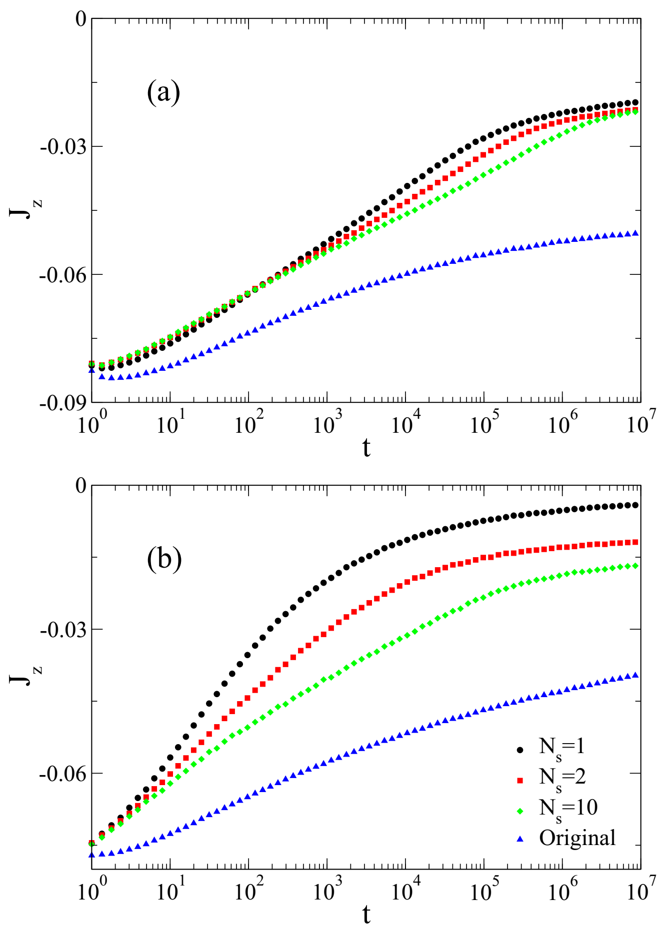

FIG. 8. Evolution of the out-of-plane current for (a) WV and (b) DT models grown in one-dimensional substrates. Models with the kinetic barrier using $N_s = 1$, 2, and 10 steps (indicated in the legend) and the original version are shown.

where q_i^{WV} is the number of nearest neighbors with $n_i^{\max}$ lateral bonds. We can express $D(i, j)$ for the DT as

$$D(i,j) = \begin{cases} 1/q_i^{\mathrm{DT}}, & \text{if } n_j > 0 \text{ and } n_i = 0 \\ 0, & \text{otherwise} \end{cases}, \qquad (8)$$

where q_i^{DT} is the number of nearest neighbors with at least one lateral bond. The quantity J_z is the average interlayer diffusion rate per site.

The currents for simulations in $d = 1$ are presented in Fig. 8. All versions in both $1 + 1$ and $2 + 1$ dimensions are characterized by a current with a downward (negative) flux with the intensity decreasing monotonically. Considering the last decade of time, we estimated the current J_∞ for $t \to \infty$ using a regression with a simple allometric function in the form

$$J_z = J_\infty + a t^{-\gamma}, \qquad (9)$$

where a and γ are parameters. In all cases with step barrier, we obtained asymptotic small negative currents with a non-universal value of γ. The results can be seen in Table I. The absolute currents for the standard models are considerably

TABLE I. Parameters J_∞ obtained in the regression using the Eq. (9) in the last decade of data of the out-plane current curves ($t > 10^6$ for $d = 1$ and $t > 10^5$ for $d = 2$).

	$d = 1$		$d = 2$	
	WV	DT	WV	DT
$N_s = 1$	−0.0015	−0.0033	−0.042	-5×10^{-5}
$N_s = 2$	−0.0019	−0.011	−0.052	-3×10^{-4}
$N_s = 10$	−0.020	−0.014	−0.053	-5×10^{-4}
Original	−0.048	−0.023	−0.050	−0.030

larger than in the cases with a barrier. The values for the DT model with a barrier are very small indicating that this current could be actually null in the asymptotic limit as observed in thermally activated diffusion models with ES step barriers [51]. In the case of the WV model, the current values may indicate the same asymptotic behavior, but our present accuracy does not allow a conclusion on this issue.

IV. CONCLUSIONS

In this work, we investigate the effects of a purely kinetic barrier caused by the out-of-plane step edge diffusion [20] on limited-mobility growth models. The cases of studies were the benchmark models of Wolf-Villain [21] and Das Sarma–Tamborenea [22]. Large-scale simulations were performed considering one- and two-dimensional substrates. It was observed that the introduction of the kinetic barrier induces the formation of quasiregular mound structures differently from those obtained with the original models that forms irregular (self-affine) structures in the interface. The kinetic barrier stabilizes the mound width, leading to the formation of quasiregular structures. The interface width in models with kinetic barriers has an initial regime similar to the original models. However, a growth exponent very close to $\beta = 1/2$ is observed for asymptotically long times. Also, the characteristic lateral length saturates after a transient that depends on the number of steps that an adatom can perform before irreversibly stick in a position. These results are consistent with mounds evolving independently. The dynamics in both one- and two-dimensional substrates are characterized by a strong reduction of downward current with respect to the original models. The downward flux has an intensity decreasing monotonically to an asymptotic value that seems to be null for the DT model and small for WV, the latter being possibly subject to strong crossover effects in the present analysis.

A central contribution of this work is to show that a very simple mechanism neglected in a previous analysis, in which particles also diffuse in the direction perpendicular to the substrate, is able to change markedly the surface morphology of basic growth models with limited mobility. Our results are qualitatively very similar to those obtained when an explicit step barrier, with a smaller probability to move downward, is considered [36]. Particularly, asymptotic mound morphology has been reported for limited mobility models in $d = 2$ without barriers with the application of the noise reduction method [38]. Our results corroborate this scenario since a small perturbation induces mound instability in this kind of processes while it alone does not produce mounds in models with thermally activated diffusion [51].

We expect that the concepts investigated in this work will be applied to more sophisticated models and aid the understanding of pattern formation in film growth and the production of self-assembled structures for technological applications.

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