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The growth of one-dimensional islands in two dimensions: Effects of anisotropic diffusion and island sizes on the island density exponent

K I Mazzitello, J L Iguain and H O Mártin†

Departamento de Física, Facultad de Ciencias Exactas y Naturales, Universidad Nacional de Mar del Plata, Funes 3350, 7600 Mar del Plata, Argentina

E-mail: hmartin@mdp.edu.ar

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Abstract. We study a model of deposition, diffusion and aggregation of particles forming one-dimensional islands in a square lattice. The dependence of the island density exponent χ on the anisotropy diffusion parameter A is analysed. It is found that χ continuously decreases from $\chi = \frac{1}{3}$ to $\chi = \frac{1}{4}$ when A increases from $A = 1$ to infinity. This nonuniversal behaviour is a direct consequence of the finite island size and, when $A > 1$, the fast diffusion direction is perpendicular to the growth direction of the islands. For infinite anisotropic diffusion, $A = \infty$, the anomalous result $\chi = 0$ is obtained.

1. Introduction

In recent years much effort has been dedicated to the study of growth of thin films by molecular beam epitaxy. In order to understand the microprocesses associated with deposition, nucleation and aggregation of ad-atoms, several models of growth of islands on two-dimensional substrate have been developed and studied [1]. Some general properties of these models, such us the behaviour of the island density and the scaling of the island size distribution as a function of relevant experimental parameters, have been analysed [2–8].

Recently, a model for diffusion and growth of silicon on Si(100) has been introduced in [9] and some properties of this model related to the universality of the island density exponent is analysed in [10]. More specifically, in the latter is studied a model of deposition, diffusion and irreversible aggregation of particles forming one-dimensional dimerized chains in a square lattice. The particles aggregate at the ends of islands and the probability of sticking is zero at the sides. The dependence of the islands density N, on two independent parameters is explored. These parameters are the anisotropy in the diffusion, A, and the average number of jumps made by every isolated particle in a given time divided by the deposited number of particles per site in the same time, R. For a fixed final coverage of particles and for large enough R, it is expected that the power law $N \sim R^{-\chi}$ be satisfied. For the case of anisotropic diffusion ($A > 1$), the direction of fast diffusion is perpendicular to the island growth direction. As the sticking probability is zero at the sides, the islands block an increasing number of fast diffusion channels as they grow. Then, the model is specially attractive to study the dependence of the *island density exponent* χ on A. It was found that $\chi = \frac{1}{3}$ for $1 \leqslant A < \infty$. The result

† Associate Member of the AS-ICTP.

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$\chi = \frac{1}{3}$ for $A = 1$ was also found for other very dissimilar models in two dimensions (see, for example, [2] for point islands and [3, 4] for dendritic islands). Linderoth [5] also found $\chi = \frac{1}{3}$ for $1 \leqslant A < \infty$ for compact and approximately square islands. All these results give support to the universality with predicts that, for a given space dimension, the value of χ does not depend on the adopted details of models, lattices and diffusion.

In this paper we study a variant of the model of [9] in which the one-dimensional islands are composed of monomers in place of dimers. As reported in section 3 we obtain $\chi = \frac{1}{3}$ for $A = 1$ and a value of χ very close to $\frac{1}{4}$ for $A = 1000$. This suggests that χ changes *smoothly* from $\frac{1}{3}$ to $\frac{1}{4}$ as anisotropy increases from $A = 1$ to the limit $A \to \infty$, provided that A be finite ($A < \infty$). These results were already found by Mo *et al* [6]. Based on point target models, Pimpinelli [7] and Mo *et al* [8] give some justifications for the result $\chi \simeq \frac{1}{4}$ when $A = 1000$ (the main assumption is that for highly anisotropic diffusion the particles describe one-dimensional random walks). For one-dimensional diffusion, which corresponds to $A = \infty$, a point island model gives $\chi = \frac{1}{4}$ [2]. Nevertheless, in [5] it is found that χ is close to $\frac{1}{3}$ for a point island model, with $A = 1000$. Moreover, for the model used in this paper, we obtain $\chi = 0$ for $A = \infty$. This result does not correspond to a point island model. Thus, the results obtained for the monomer chains merit a deeper analysis, and that is the aim of this paper.

This paper is organized as follows. In section 2 we define the model. The Monte Carlo results are presented in section 3. In that section we show how the obtained results are a direct consequence of the combination of the growth rules of the model and the anisotropic diffusion. Finally in section 4 we state our conclusions.

2. The model and the Monte Carlo simulation

The substrate is represented by a square lattice of 1000×1000 sites and lattice constant a. Periodic boundary conditions were adopted in order to avoid the edge effects.

At each Monte Carlo step, one site of the substrate is randomly chosen. The following situations may appear:

(i) If the site is empty, it is occupied with a particle with probability ϵ.
(ii) If the site is occupied and both its nearest neighbour (NN) sites in the x direction are empty, the particle tries to jump to any of its NN sites in the x direction with probability P_x, and to any of its NN sites in the y direction with probability P_y. If the particle attempts to jump in the y direction and the chosen NN site is occupied by another particle, the jump is not performed.
(iii) If the site is occupied and any of its NN sites in the x direction is also occupied, nothing happens. Particles located at NN sites in the x direction are bounded and form islands.

Simulations start with the lattice empty and run until the density per site of deposited particles reach a desired value Θ. We are interested in coverage low enough to avoid the percolation regime [3]. So, employed values for Θ are below 0.1.

In this model, sites can be occupied, at most, by only one particle. Islands grow irreversibly by aggregation of particles in the x direction and particles do not stick at the lateral side of one-dimensional islands.

Although, for a fixed final coverage, the model presents three variable parameters (ϵ, P_x, and P_y), it is expected that all processes depend only on the relative value of these probabilities. Then there are only two independent relevant parameters, which we have chosen as $A \equiv P_y/P_x$ and $R \equiv 2(P_x + P_y)/\epsilon$. Figure 1 shows, as an example, a final resulting island structure corresponding to a simulation performed with $R = 10^9$, $A = 1000$ and $\Theta = 0.05$.

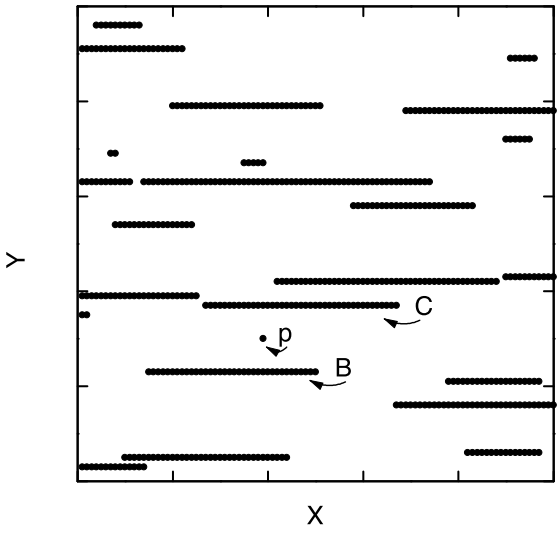

Figure 1. A representative sample of 100×100 sites from a lattice of 1000×1000 sites obtained with our Monte Carlo simulation for $R = 10^9$, $A = 1000$ and $\Theta = 0.05$. Filled circles represent particles and p is an example of isolated particles that perform quasi one-dimensional diffusion in the x direction because of the collisions with the lateral sides of islands, in this case B and C.

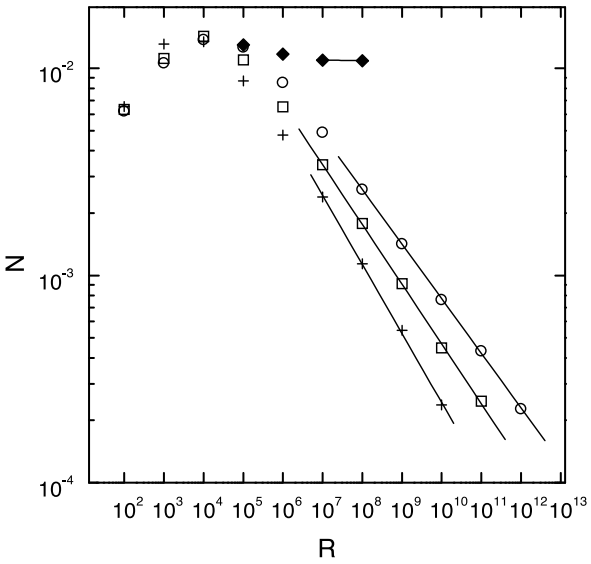

Figure 2. The average island density, N, against R in log–log scale for different values of the diffusion anisotropy A. $A = 1$ (crosses), $A = 10^2$ (open squares), $A = 10^3$ (open circles) and $A = \infty$ (diamonds). Lines correspond to the asymptotic behaviour and their slopes are stored in the second column of table 1.

3. Results

In figure 2 is shown, in log–log scales, the behaviour of the island density as a function of R for different values of A. The values of the exponent χ obtained from the straight lines are

Table 1. Island density exponent corresponding to several values of A (first column) and obtained in four different ways. Asymptotic values of the slope of $\log N$ versus $\log R$ (second column see figure 2) and of $\log N$ versus $\log(R/\log R)$ (fourth column). Exponents calculated from equation (7) (third column) and equation (8) (fourth column). The relative errors of all these results are below 5%.

A	χ	$\chi_{\bar{n}}$	χ'	$\chi'_{\bar{n}}$
1	0.33	0.31	0.33	0.33
10^2	0.29	0.28	0.30	0.29
10^3	0.26	0.26	0.27	0.27
∞	0	0	0	0

shown in the second column of table 1. The infinitely anisotropic case is very special and will be discussed later. For $A = 1$ we obtain $\chi = 0.33 \pm 0.01$ which is in agreement with the value $\chi = \frac{1}{3}$ obtained for other two-dimensional models with isotropic diffusion [2–5, 10]. For $A = 10^3$ we obtain $\chi = 0.26 \pm 0.01$, a value which is very close to $\chi = \frac{1}{4}$, which corresponds to the point island model in one dimension. These results show that χ continuously changes as R increases.

Let us now try to understand this behaviour. Remember that the particles do not stick at the lateral sides of islands; then these sides act as obstacles for them. For example, the particle p in figure 1 will collide many times with islands B and C. Then this particle will perform an *effective* one-dimensional diffusion in the x direction (not in direction of fast diffusion) before it can escape from the region limited by the former two islands. In order to study this effect we compute the square root of mean-square displacements, Δx and Δy, in the x and y direction, respectively, for an isolated random walker (a tracer particle) with anisotropic diffusion A, as a function of the number of jumping steps n. This particle starts the walk in a randomly chosen empty site of a substrate. This substrate was previously obtained applying the Monte Carlo rules of the model up to the final coverage Θ was reached. This random walker does not stick at the ends of islands but always diffuses. For this tracer particle, we also compute the mean number of distinct visited sites $S(n)$ as function of n.

Figure 3 shows Δx and Δy as a function of n in log–log scales, for $R = 10^9$, $A = 1$ and $R = 10^9$, $A = 10^3$. The dashed straight line corresponds to Δy for a random walk on an empty substrate (a lattice without islands). At short times, the random walk behaves as in an empty substrate. After that it encounters obstacles that delay displacement in the y direction. At long times the random walk recovers the behaviour $\Delta y \propto \sqrt{t}$. Δx is, in practice, not modified by the presence of islands, because they are obstacles of small cross section. Note that, for $A = 10^3$, there is a region of n in which Δy is almost constant, and then the random walker performs an effective one-dimensional diffusion in the x direction.

The exponent χ is related to the way monomers explore the substrate. It can be shown, following [2], that N and the densities of monomers, N_1, satisfy the relations

$$\frac{\mathrm{d}N}{\mathrm{d}t} \sim \frac{N_1}{\tau}\left(\frac{N_1}{N}\right) \tag{1}$$

$$\frac{\mathrm{d}N_1}{\mathrm{d}t} \sim \frac{\epsilon}{\Delta t} - \frac{N_1}{\tau} \tag{2}$$

when $N_1 \ll N$, an assumption valid when $R \gg 1$ (the case we are interested in), where τ is the lifetime for monomers and Δt is the time associated with each Monte Carlo step. Numerical integration of equations (1) and (2) shows that a quasi-stationary regime exists

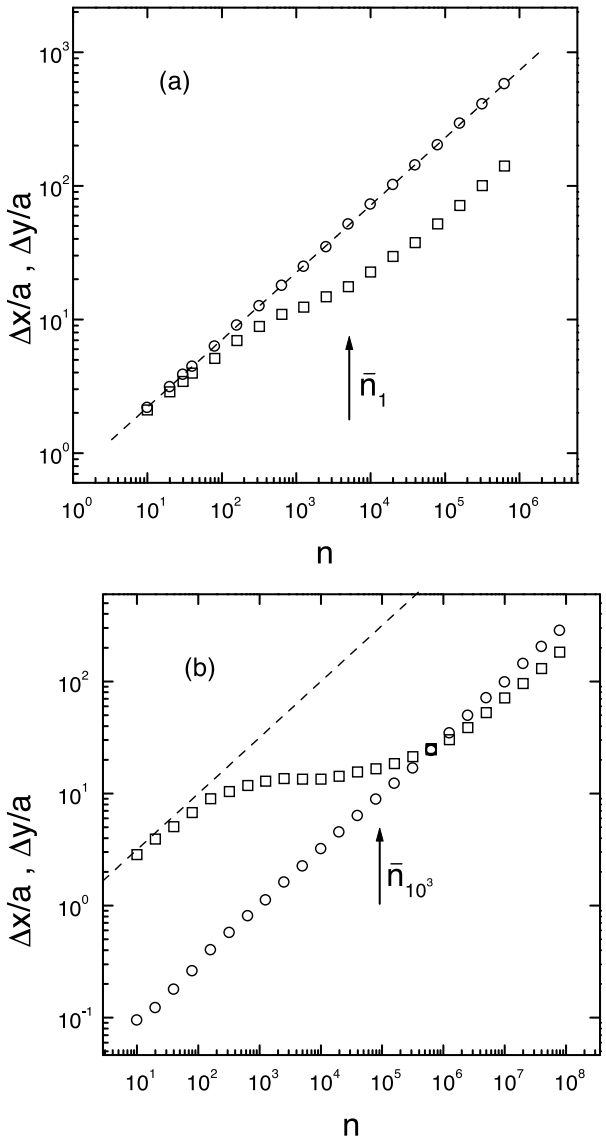

Figure 3. The square root of the mean-square displacement, Δx (open circles) and Δy (open squares), against the number of steps, n, for particles diffusing with anisotropy A on a substrate with an island configuration obtained by means of Monte Carlo simulations of the model for A, R and Θ. (*a*) Corresponds to $A = 1$, $R = 10^9$ and $\Theta = 0.05$, (*b*) corresponds to $A = 10^3$, $R = 10^9$ and $\Theta = 0.05$. The dashed lines correspond to Δy for the case of free diffusion (i.e. on an empty lattice). For the meaning of $\overline{n_i}$ see figure 4. The Monte Carlo results where obtained averaging over 10^3 samples.

where $\mathrm{d}N_1/\mathrm{d}t \sim 0$ [2], and in such a case (1) becomes

$$\frac{\mathrm{d}N}{\mathrm{d}t} \sim \frac{\epsilon^2}{\Delta t^2}\tau N^{-1}. \tag{3}$$

On average, each monomer visits a number N^{-1} of distinct sites between the instant of

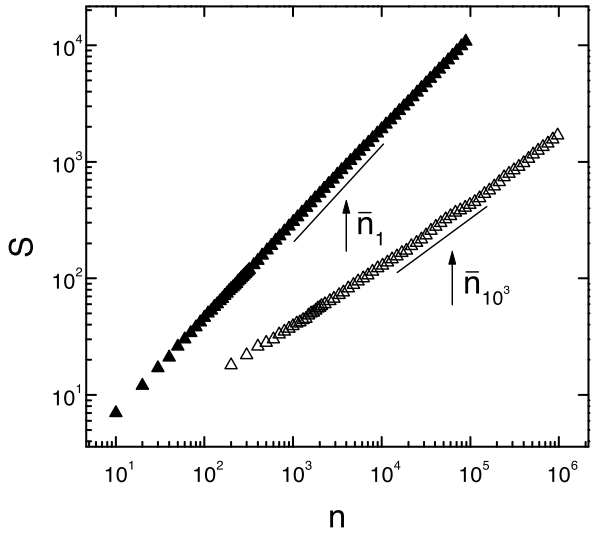

Figure 4. The mean number of distinct visited sites S against the number of steps for particles diffusing with anisotropy $A = 1$ (filled triangles) and $A = 10^3$ (open triangles) on substrates obtained by means of Monte Carlo simulations with the values of A, R and Θ corresponding to figures 3(*a*) and (*b*), respectively. The values of $\overline{n_i}$ were obtained graphically using equation (4) with the values of N extracted from figure 2. The slopes of the straight lines correspond to the exponent α of equation (5). These Monte Carlo results where obtained averaging over 10^3 samples.

deposition and the instant of aggregation for this monomer. So, we can write

$$S(\overline{n}) \sim \frac{1}{N} \tag{4}$$

with $\overline{n} = 2((P_x + P_y)/\Delta t)\tau$. If, furthermore,

$$S(\overline{n}) \sim \overline{n}^{\alpha} \tag{5}$$

then (3) can be integrated to obtain [11]

$$N \sim R^{-\chi_{\overline{n}}}\Theta^{\chi_{\overline{n}}} \tag{6}$$

with

$$\chi_{\overline{n}} = \frac{\alpha}{1+2\alpha}. \tag{7}$$

The quantity α appearing in (7) is, according to (4) and (5), the local value of the slope of $\log S$ as a function of $\log n$ around the point $\overline{n}$ where $S(\overline{n}) = 1/N$. So, values for α were obtained graphically from figure 4, using the corresponding values for N extracted from the asymptotic regime of figure 2. The values of $\chi_{\overline{n}}$ obtained from equation (7) are shown in the third column of table 1, and are in agreement with the results of χ obtained from the slopes of straight lines of figure 2. Let us note that the values of α (and then $\chi_{\overline{n}}$) might depend on R; nevertheless the values of $\chi_{\overline{n}}$ obtained are independent of the value of R used. For example, for $A = 10^3$ we obtain $\chi_{\overline{n}} = 0.260, 0.262, 0.268$ and 0.257 for $R = 10^9$, 10^{10}, 10^{11} and 10^{12}, respectively. These results and the straight line shown in figure 2 confirm that the value $\chi = 0.26$ for $A = 10^3$ corresponds to region of an R extended, at least, over five orders of magnitude. The open question is whether in the asymptotic regime $R \to \infty$ one obtains the two-dimensional value $\chi = \frac{1}{3}$. The simulation for very large values of R is beyond our computational facilities. Nevertheless, for $10^8 \leqslant R \leqslant 10^{12}$ an effective one-dimensional

diffusion (the plateau shown in figure 3(*b*)) is obtained. We also compute the ratio between the average island length $\langle l \rangle$ and Δx at $\overline{n}$, $\eta \equiv \langle l \rangle / \Delta x(\overline{n})$. A value of $\eta > 1$ means that, on average, the particles cannot escape from obstacles. We obtain $\eta = 4.3$, 4.4, 4.1 and 4.5 for $R = 10^9$, 10^{10}, 10^{11} and 10^{12}, respectively. Then the value $\chi = 0.26$ for $A = 10^3$ can be interpreted as a consequence of the effective one-dimensional diffusion (remember that $\chi = \frac{1}{4}$ for a point island model in one dimension), and there is no indication that this value tends to $\chi = \frac{1}{3}$ as R increases (at least for $10^8 \leqslant R \leqslant 10^{12}$).

Taking into account the logarithmic correction which appears in two-dimensional systems we replace, in the x-axis of figures 2 and 4, R and n by $R/\log R$ and $n/\log n$, respectively. From these new figures (not shown here) we obtain the exponents χ' and α'. Following the same procedure used to obtain equation (7), one has

$$\chi'_{\overline{n}} = \frac{\alpha'}{1 + 2\alpha'}. \tag{8}$$

The obtained values of χ' and $\chi'_{\overline{n}}$ are shown in table 1. One can see the agreement between the values of the island density exponents obtained by different methods.

Finally, we discuss the case of infinitely anisotropic diffusion. For $A = \infty$, and long times, the diffusion along the y direction is blocked by obstacles and $\Delta x = 0$ because the probability of jumping along the x direction is zero ($P_x = 0$). Then, the random walker does not perform an effective one-dimensional diffusion as in the case of finite values of A. Now Δy and $S(n)$ reach saturation constant values at large times. We obtain $\alpha = 0$ and then, from equation (7), $\chi_{\overline{n}} = 0$. But the assumptions used to obtain equation (7) could be unsatisfied. For example, the number of diffusing monomers could be large. Nevertheless, from figure 2 we also found that $\chi = 0$. This is because for large enough R, particles visit all their accessible sites between obstacles before finding another new deposited particle, and then increasing R will not change the island number density. The same anomalous result $\chi = 0$ for $A = \infty$ was found in [10] for a model of growth of dimerized chains.

4. Conclusions

In this paper we obtain for $1 < A < \infty$, nonuniversal (i.e. A-dependent) results for the island density exponent χ. These values can be considered as effective exponents (χ_{eff}), in the sense that they could be different from the exponent corresponding to the asymptotic regime $R \to \infty$. Nevertheless, the values of these χ_{eff} hold in a region of four to five orders of magnitude in R, and we do not find any indication that these values change as R increases. Then it is possible that $\chi = \chi_{eff} = \frac{1}{4}$ in the limit $R \to \infty$. Unfortunately, the confirmation of this possibility is beyond our computational facilities.

A possible explanation of the dependence of χ_{eff} on A is as follows. Let us consider a random walk with an anisotropic diffusion A much greater than one (then $\Delta y \gg \Delta x$ at all times). In this case a crossover number of step $n_c = A$ exists ($\Delta x(n_c) \simeq a$). At n much smaller than n_c, the random walk behaves as in one dimension ($\alpha = \frac{1}{2}$). At n much greater than n_c, the random walker performs a two-dimensional diffusion ($\alpha = 1$). Now if the average number of steps $\overline{n}$ of a monomer is lower than n_c, one would expect to obtain the one-dimensional exponent $\chi_{eff} = \frac{1}{4}$ (see equation (7)). Although we actually found $\chi_{eff} = \frac{1}{4}$ for $A \gg 1$, it was also obtained that $\overline{n} \gg A$ (see figure 4 where $\overline{n}_{1000}$ is close to 10^5 for $A = 1000$) which would implicate $\chi = \frac{1}{3}$. So the above-mentioned argument is wrong. Note that in this reasoning, we are dealing with a random walk in an empty lattice and that for $n < n_c$ the one-dimensional diffusion is in the y direction. In a real system, there exist islands that play the role of obstacles (see figure 1). Due to these obstacles, monomers perform an effective

one-dimensional diffusion, but in the x direction (see figure 3(b) where there is a region of n in which $\Delta y \simeq$ constant). Due to this effect, one finds $\chi_{eff} \simeq \frac{1}{4}$ for $A = 10^3$, as in an one-dimensional system.

The results presented here show that $\chi_{eff} = \frac{1}{4}$ in the limit $A \to \infty$. On the other hand, for $A = \infty$ one finds $\chi = 0$. This last result is a direct consequence of the fact that the particles cannot escape from the column where they have landed, and that their motions are limited by the nearest obstacles in the y direction. In contrast, for a finite value of A, they perform an effective one-dimensional diffusion in the x direction.

Let us finally comment that in [11] is also found a nonuniversal behaviour of the exponent χ for a point island model in two dimensions. This behaviour is a consequence of the substrate where the particles diffuse. In this substrate there are impurities with a repulsive monomer–impurity interaction. These impurities act as obstacles reducing the space where the particles diffuse. Then, a random walker performs an anomalous diffusion (i.e. $\alpha < 1$) in this substrate. Due to this anomalous diffusion, the exponent χ (see equation (7)) smoothly changes with the concentration of impurities. The main difference between this model and ours is that in the former the obstacles are already present at the beginning of simulations, whereas in the latter they appear as a consequence of the dynamics.

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References

[1] See, for example
King D A and Woodruff D P 1997 *Growth and Properties of Ultrathin Epitaxial Layers* (Amsterdam: Elselvier)
Barabási A-L and Stanley H E 1995 *Fractal Concepts in Surface Growth* (Cambridge: Cambridge University Press)
[2] Bartlet M C and Evans J W 1992 *Phys. Rev.* B **46** 12 675
[3] Amar J G, Family F and Pui-Man Lam 1994 *Phys. Rev.* B **50** 8781
[4] Family F and Amar J G 1995 *Mater. Sci. Engng* B **30** 149
[5] Linderoth T R, Mortensen J J, Jacobsen K W, Laegsgaard E, Stensgaad I and Besenbacher F 1996 *Phys. Rev. Lett.* **77** 87
[6] Mo Y W, Kleiner J, Webb M B and Lagally M G 1992 *Phys. Rev. Lett.* **69** 986
[7] Pimpinelli A, Villain J and Walf D E 1992 *Phys. Rev. Lett.* **69** 985
[8] Mo Y W, Kleiner J, Webb M B and Lagally M G 1992 *Surf. Sci.* **268** 275
[9] Iguain J L, Mártin H O and Aldao C M 1996 *Phys. Rev.* B **54** 8751
[10] Iguain J L, Mártin H O and Aldao C M 1999 Effects of diffusional anisotropy and substrate inhomogeneity on lengths distribution scaling of dimerized chains *Phys. Rev.* B **59** 4596
[11] Liu S, Böing L, Detch J and Metiu H 1995 *Phys. Rev. Lett.* **74** 4495