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Abstract

The effects of the biased diffusion of particles on the growth of islands for a model of deposition, diffusion and irreversible aggregation of particles forming islands in two dimensions is studied. The direction of the bias is perpendicular to the growth direction of islands and these islands act as obstacles for the diffusing particles that collide with the lateral sides of islands without sticking. Due to the biased diffusion the islands are composed by the agglomeration of monomer chains. Using Monte Carlo simulations we found that the island density exponent continuously increases from to $1/2$ when the bias parameter λ increases from $\lambda = 1$ to ∞, but the chain density exponent $\chi_c \cong 1/3$ is independent of λ. For $\lambda = \infty$, the fully directed diffusion case, an expression for the nucleation rate can be obtained which is in agreement with the numerical results.

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1. Introduction

Since the advent of the electron microscopy techniques which allow us to observe surfaces with atomic resolution, the study of submonolayer deposition has attracted increasing interest in recent years [1]. Several models have been studied from a theoretical point of view [1–16]. In these models, particles which arrive on a surface at a constant flux diffuse before being incorporated into islands or before meeting other isolated particles and forming new islands. Starting with an empty surface and when a coverage of particles θ is reached, the characteristics of the submonolayer formed are analysed. If the islands can neither break or diffuse it is expected that all processes depend only on the ratio R between the diffusion and deposition rates. When R increases, particles perform on average a greater number of hops between the incoming particles. Then, they have a greater probability of reaching pre-existing islands than

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forming new ones. At a fixed coverage θ and for large enough values of R, it is expected that the island density N behaves as

$$N \sim R^{-\chi} \tag{1}$$

where χ is the so-called island density exponent.

In connection with the study of diffusion and growth of silicon islands on a Si(100) surface, a model of aggregation of particles forming dimer chains on a square lattice has been introduced [18]. In this model the diffusion of particles is anisotropic and the diffusion constant in one direction of the square lattices is A times greater than the diffusion constant in the other perpendicular direction (A being the anisotropy parameter of the model). The effects of this anisotropic diffusion on the island density exponent for the case of irreversible aggregation have been recently studied [18]. In order to understand the behaviour of χ the study has been extended to a simpler model in which islands are composed of chains of monomers instead of chains of dimers. It has been found that in a region of four to five orders of magnitude in R, the value of the exponent χ depends on A. This non-universal behaviour can be understood from the analysis of trajectories performed by tracer particles [19, 20].

The aim of this paper is to study the effects of a biased diffusion of particles on the growth of islands. We use a model on a square lattice which is especially attractive for the study of these effects because the direction of the bias (parallel to the y direction of the lattice) is perpendicular to the growth direction of islands (the x direction). Due to the growth rules of the model (explained in section 2), the islands are composed of chains of monomers oriented along the x direction. These islands act as obstacles for moving particles that collide with the lateral sides of these islands without sticking. The length of islands increases as they grow, blocking an increasing number of channels in the y direction.

Let us comment that, although we do not attempt to simulate any specific experiment, in real systems an electric field parallel to the surface can produce relevant effects on the growth of islands due to the biased diffusion of particles on the surface (see e.g. [17]).

The outline of the paper is as follows. In section 2 we define the model. The Monte Carlo results are presented in section 3. From these results we find that islands are composed by the agglomeration of monomer chains and we explain why this agglomeration is due to the biased diffusion of monomers. The value of the exponent χ depends on the bias parameter and the value of the chain density exponent χ_c ($Nc \sim R^{-\chi_c}$, Nc being the density of chains) is independent of it. The density of islands increases with the nucleation of new islands and decreases with the coalescence of close islands. In section 4, for the case of an extremely large value of the bias parameter (fully directed diffusion), and analysing the trajectory of monomers, an expression for the nucleation rate is obtained which is in agreement with Monte Carlo data. Finally, in section 5 we summarize our results.

2. The model

The substrate is represented by square lattice of 3000×3000 sites. Periodic boundary conditions were adopted in order to avoid edge effects.

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

(1) If the site is empty, it is occupied with a particle with probability ϵ.
(2) 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 $P_x = 1/4$, to the upper NN site in the y direction with probability p, or to the lower NN

site in the y direction with probability λp. 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.

(3) 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 chains of monomers.

Simulations start with the lattice empty and run until the density per site of deposited particles reaches a desired value θ. We are interested in coverage low enough to avoid the percolation regime [6], so the employed values of θ are $\theta \leqslant 0.2$. The bias parameter λ ($1 \leqslant \lambda \leqslant \infty$) is a free parameter of the model. We normalized the jumping probabilities by demanding that $2P_x + p + \lambda p = 1$. Then $p = 1/2(\lambda + 1)$. For $\lambda = 1$ the isotropic diffusion case is recovered and $\lambda = \infty$ corresponds to a fully directed diffusion. The drift velocity of particles v_{drift} is given by the difference between the probability of jumping downwards (λp) and upwards (p). Then $v_{\text{drift}} = (\lambda - 1)p = (\lambda - 1)/2(\lambda + 1)$ ($v_{\text{drift}} = 0, 1/2$; for $\lambda = 1, \infty$, respectively).

In this model, sites can be occupied, at most, by only one particle. Chains grow irreversibly by aggregation of particles in the x direction and particles do not stick at the lateral sides of chains. We consider that two chains connected by NN distances in the y direction belong to the same island (see figure 1(a)). In this way an island can be formed by one isolated chain (a chain without NN chains in the y direction) or a group of chains connected by NN distances in the y direction.
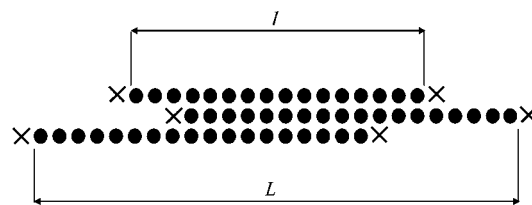
(a)
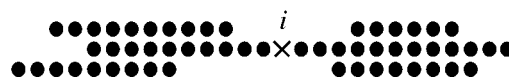
(b)
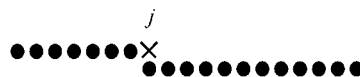
(c)

Figure 1. (a) A typical island composed of K chains of monomers ($K = 3$ in the present case). We denote the length of islands by L and the length of monomer chains by l. The crosses indicate the growth sites. If one particle arrives at one of these sites it irreversibly aggregates to the island. Two examples of coalescence are shown in (b) and (c). If a particle arrives at site i (case (b)) or j (case (c)), coalescence between the two close islands will occur.
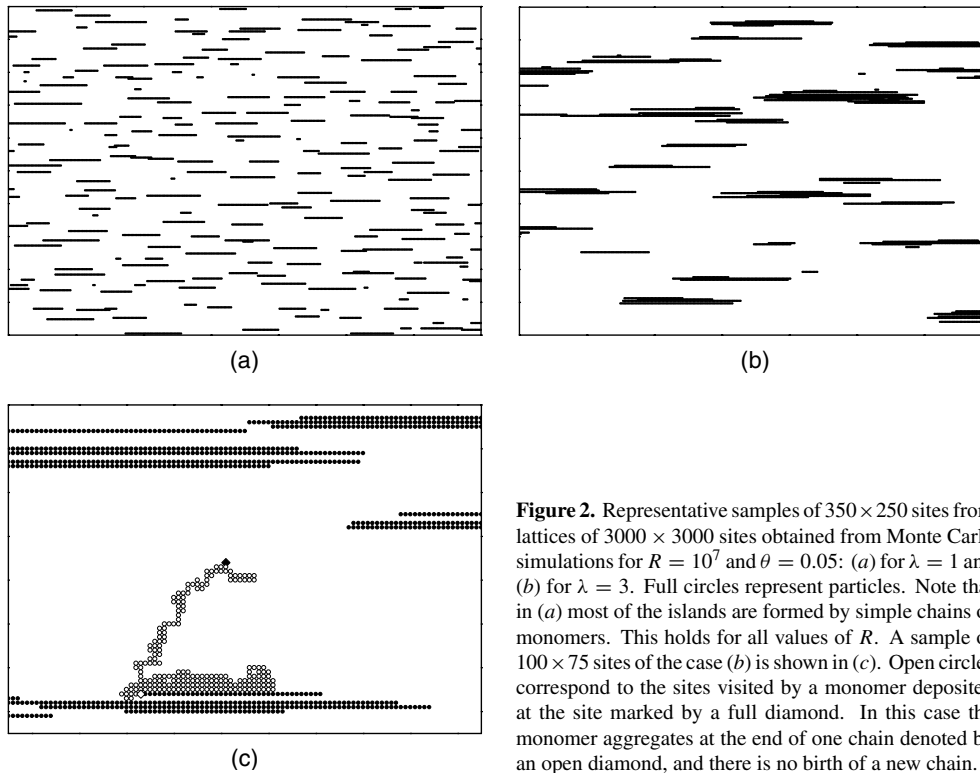
(a)

(b)

(c)

Figure 2. Representative samples of 350×250 sites from lattices of 3000×3000 sites obtained from Monte Carlo simulations for $R = 10^7$ and $\theta = 0.05$: (*a*) for $\lambda = 1$ and (*b*) for $\lambda = 3$. Full circles represent particles. Note that in (*a*) most of the islands are formed by simple chains of monomers. This holds for all values of R. A sample of 100×75 sites of the case (*b*) is shown in (*c*). Open circles correspond to the sites visited by a monomer deposited at the site marked by a full diamond. In this case the monomer aggregates at the end of one chain denoted by an open diamond, and there is no birth of a new chain.

For fixed values of θ and λ it is expected that all processes depend only on the ratio R between the jumping rate and the deposition rate, $R = (2P_x + p + \lambda p)/\epsilon = 1/\epsilon$. Then the relevant parameters of the model are θ, λ and R.

3. Monte Carlo results

Figure 2 shows typical examples of island structures obtained from simulations performed with $R = 10^7$, $\theta = 0.05$ and $\lambda = 1$ and 3. For $\lambda > 1$ islands formed for more than one chain of monomers appear. These agglomerations of chains are due to the biased diffusion of isolated monomers (monomers with the four NN sites empty). An isolated monomer moves, on average, downwards. If during this movement the monomer reaches a lateral side of one island, the movement downwards is stopped. This is because there is no interaction between NN particles in the y direction and the monomer collides with the lateral sides of chains without sticking. Then the monomer will diffuse in a region close to and above the island (see figure 2(*c*)). If now, as a consequence of diffusion or deposition, a particle arrives at a NN site in the x direction of the monomer, the two particles become bounded forming a new chain of two particles. This new chain is formed near the old island. It is possible that the new chain is formed above and upon the old island (i.e. at a NN distance in the y direction of the old island). In this case the number of chains of this island is increased by one ($K \rightarrow K + 1$, a new chain is formed but not a new island). From the point of view of islands, this process corresponds to an aggregation of two particles to the island. Note that in the case of $\lambda = \infty$ the monomer cannot jump upwards (because $p = 0$). Then the new chain will be necessarily formed above and upon the old one.
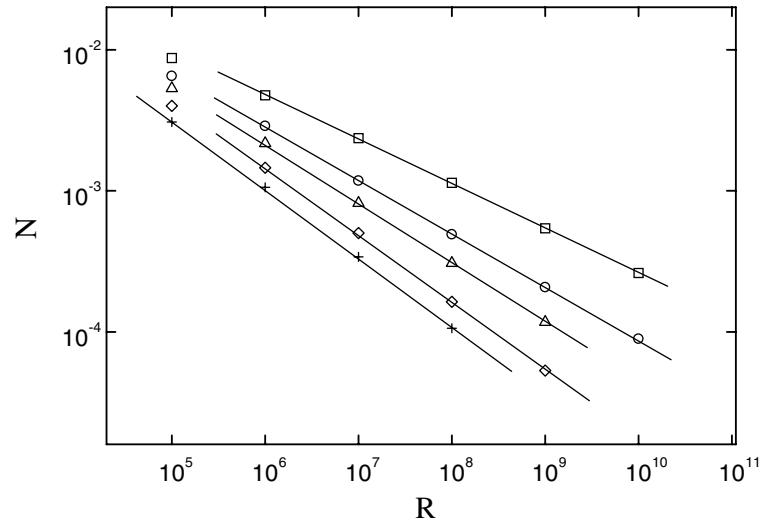
Figure 3. The density of islands N versus R in log–log scales for $\theta = 0.05$ and different values of λ. $\lambda = 1$ (squares), 2 (circles), 3 (triangles), 7 (diamonds) and ∞ (crosses). The exponents χ obtained from the straight lines are shown in table 1. The typical error bars of the Monte Carlo data are approximately the same size as the symbols shown for each point.

Table 1. The exponents χ, χ_c and α for $\theta = 0.05$ and different values of λ. The island and chain density exponents χ, χ_c are obtained from Monte Carlo simulations. The exponent α is obtained from equation (4). The relative error bars are below 2%.

λ	χ_{ef}	χ_c	$\alpha = \chi - \beta$
1	0.310	0.310	0
2	0.377	0.297	0.080
3	0.415	0.300	0.115
7	0.470	0.301	0.169
15	0.480	0.296	0.184
1000	0.486	0.301	0.185
∞	0.487	0.302	0.185

Figure 3 shows $\log N$ as a function of $\log R$ for $\theta = 0.05$ and different values of λ. The exponents χ obtained from the straight lines are shown in table 1. Let us note that χ continuously increases with λ from $\chi \cong 1/3$ for $\lambda = 1$, to $\chi \cong 1/2$ for $\lambda = \infty$.

Let $\overline{K}$ be the average number of chains per island (see figure 1(a)). Then

$$N_c \sim \overline{K} N \tag{2}$$

where N_c is the density of chains. Following equation (1) it is expected that

$$N_c \sim R^{-\chi_c} \tag{3}$$

where χ_c is the chain density exponent. From equation (2) $\overline{K} \sim R^{\alpha}$, with

$$\alpha = \chi - \chi_c. \tag{4}$$

The values of the exponents χ, χ_c and α obtained for $\theta = 0.05$ and for different values of λ are shown in table 1. Note that the value of χ_c is close to $1/3$ and is independent of the value of λ. In figure 4 $N_c R^{0.30}$ is plotted against R in log–log scales for $\lambda = \infty$ and different values of coverage θ. One can also observe here that χ_c does not depend on θ. From these and other
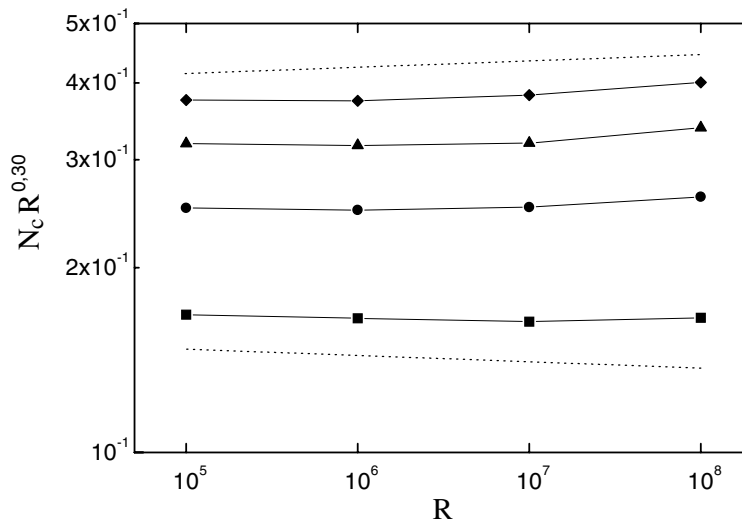
Figure 4. Log–log plot of $N_c R^{0.30}$ against R in log–log scales for $\lambda = \infty$ and different values of the coverage θ. $\theta = 0.05$ (squares), 0.10 (circles), 0.15 (triangles) and 0.20 (diamonds). Error bars are approximately the same size as the symbols. The dashed straight lines have slopes of ± 0.01 and have been drawn to guide the eye.

results obtained for other values of θ and λ not shown here, we can conclude that the chain density exponent χ_c for $0.05 \leqslant \theta \leqslant 0.20$ is independent of λ and θ, and that the increases of the island density exponent χ with λ is due to the agglomeration of chains (see equation (4)). For $\lambda = 1$ the drift velocity is zero and there is no agglomeration ($\alpha = 0$, see table 1): the islands are composed by single chains and then the island density exponent is equal to the chain density exponent. For $\lambda = \infty$ the exponent χ seems to be also independent of θ. We have obtained $\chi = 0.487, 0.496, 0.498$ and 0.495 for $\theta = 0.05, 0.10, 0.15$ and 0.20, respectively. As was explained in [19] the result of $\chi \cong 1/3$ for $\lambda = 1$ (the case $A = 1$ there corresponds to $\lambda = 1$ here) is due to the two-dimensional diffusion performed by monomers and it is well known that for a two-dimensional non-biased diffusion a two-dimensional exponent $\chi = 1/3$ is expected [5, 10, 19].

4. Nucleation rate for $\lambda = \infty$

For $\lambda = \infty$ the monomers cannot jump upwards. Let us consider a monomer which reaches an island. As was mentioned in section 3, one possibility is that the monomer will aggregate to the island by finding another monomer and forming a new chain of two particles above and upon the old island (and $K \rightarrow K+1$). Another kind of aggregation takes place if the monomer arrives at one of the growth sites of the island (see figure 1(*a*)).

The coalescence of islands is another process that occurs. As the coverage θ increases, the length of islands also increases and it is possible that two islands become separated by only one empty site in the x direction (see figure 1(*b*)). Now, if a particle arrives at this empty site, the two islands coalesce and form one single island. Another example of coalescence is shown in figure 1(*c*). If a particle arrives at site j the two islands will coalesce.

On the other hand, a nucleation of islands takes place when a particle arrives at a NN site in the x direction of an isolated monomer. Then the probability of nucleation forming new
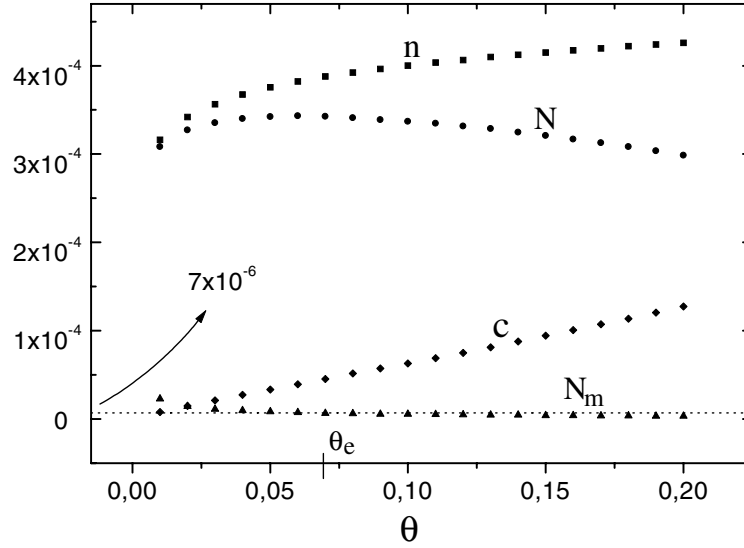
Figure 5. (*a*) The island N, nucleation n, coalescence c and monomer N_m densities as a function of coverage θ for $R = 10^7$ and $\lambda = \infty$. The dashed straight line corresponds to the mean value of N_m.

islands is proportional to the density of isolated monomers, N_m, and the probability that a particle collides with an island is proportional to $N\overline{L}$, where $\overline{L}$ is the average length of islands (see figure 1(*a*)). $\overline{L}$ corresponds to the cross section for an isolated monomer diffusing from the top with a drift velocity perpendicular to the direction of L. When a monomer reaches an island it is highly probable that either an aggregation to this island or a coalescence between close islands will occur. Then the probability of nucleation, P_n, of new islands behaves as $c N_m/(c N_m + \overline{L} N)$, where c is a constant (c depends on λ but in this section we are working with $\lambda = \infty$) which takes into account that monomers are moving downwards and islands are fixed. We will use this relation later on.

From the above discussion one has

$$N = n - c \tag{5}$$

where n and c are the density of nucleation and coalescence, respectively (i.e. the total number of nucleation and coalescence processes per lattice site which have been taking place from the beginning of the simulation with an empty lattice up to the point of obtaining the final structure at the coverage θ).

In figure 5 N, n, c and N_m as a function of θ for $R = 10^7$ are plotted. One can see that for small values of θ, N increases with θ, and N takes its maximum value at $\theta_e \cong 0.07$. For larger values of θ, and due to coalescence processes, N decreases.

Let us now obtain the rate equation for nucleation [5, 10, 19]. Let τ be the lifetime of isolated monomers between their deposition and nucleation or collision with an island. Then

$$\frac{dN_m}{dt} \cong (1 - \theta)\frac{\epsilon}{\Delta t} - \frac{N_m}{\tau} \tag{6}$$

where Δt is the time associated with each Monte Carlo step. We are neglecting the case where a monomer is deposited at a NN site of an island or another monomer. But the probability of these events is negligible compared to $(1 - \theta)\epsilon/\Delta t$. The decrease of N_m/τ monomers per

unit of time in equation (6) is due to nucleation, coalescence and aggregation processes. Then $dn/dt \sim P_n N_m/\tau$, where P_n is the probability of nucleation. As mentioned above, P_n behaves as $c\, N_m/(c\, N_m + \overline{L} N)$. For large values of R one has $N_m \ll N$ (see figure 5) and also $1 < \overline{L}$. Then $P_n \sim N_m/(\overline{L} N)$ and

$$\frac{dn}{dt} \sim \frac{N_m}{\tau} \frac{N_m}{\overline{L} N}. \tag{7}$$

Also, for $R \gg 1$ a quasi-stationary regime exists where $dN_m/dt \simeq 0$ (see figure 5). Then from equation (6) one has $N_m/\tau \simeq (1-\theta)\epsilon/\Delta t$. Using the average number of steps, $\overline{n}$, performed by an isolated monomer $\overline{n} = (2P_x + p + \lambda p)\tau/\Delta t = \epsilon R\tau/\Delta t$, one obtains from equation (7)

$$\frac{dn}{dt} \sim (1-\theta)^2 \frac{\epsilon}{\Delta t} \frac{\overline{n}}{R} \frac{1}{\overline{L} N}. \tag{8}$$

Due to the fully directed diffusion of isolated monomers, $\overline{n}$ is proportional to the average distance between two consecutive islands in the y direction. Since the islands are randomly distributed this distance is $\overline{K}/\theta$. Note that in this reasoning we are considering the collision of isolated monomers with islands and not the nucleation processes. This is a good approximation because $N_m \ll \overline{L} N$. Finally, using $d\theta = (1-\theta)\epsilon\, dt/\Delta t$ one obtains

$$\frac{dn}{d\theta} \sim \frac{(1-\theta)\overline{K}}{\theta R \overline{L} N}. \tag{9}$$

In figure 6 $(dn/d\theta)[(1-\theta)\overline{K}/\theta R\overline{L}N]^{-1}$ is plotted as a function of R for different values of θ. From these Monte Carlo results one can conclude that equation (9) gives a good approximation of $dn/d\theta$ for $\lambda = \infty$. The results are close to a straight line of slope zero indicating that $dn/d\theta$ behaves as $R^{\alpha+\chi-1}/\overline{L}$. For large values of R the density of monomers is much smaller than the coverage θ, and then $\theta \simeq \overline{l}\overline{K}N$. Assuming that $\overline{L}$ and $\overline{l}$ have approximately the same behaviour one obtains $dn/d\theta \sim R^{-\gamma}$, with $\gamma \simeq 1 - 2\alpha$. From Monte Carlo data we found that this is a reasonably good approximation for the exponent γ. Note that we cannot obtain n as a function of R and θ integrating equation (9) because $\overline{K}$, $\overline{L}$ and N are unknown functions of θ.

5. Conclusions

The Monte Carlo results show that the chain density exponent χ_c does not depend on λ ($1 \leqslant \lambda \leqslant \infty$), at least in the range of R used in this work and for $0.05 \leqslant \theta \leqslant 0.20$, and the value of χ_c is close to $1/3$. For $\lambda = 1$ the same result for χ_c was found in [19] (the case $A = 1$ there corresponds to $\lambda = 1$ here) and can be explained due to the two-dimensional non-biased diffusion performed by monomers. The increase of the island density exponent χ with λ is due to the agglomeration of chains. This agglomeration appears because the chain growth direction (the x direction) is perpendicular to the direction of the drift velocity (the y direction) and there is no interaction between NN particles in the y direction (the probability of sticking at the sides of chains is zero). The agglomeration exponent α increases from $\alpha = 0$ to $\alpha \simeq 0.19$ when λ increases from $\lambda = 1$ to ∞. As $\chi = \chi_c + \alpha$, the exponent χ increases from $\chi \simeq 1/3$ to $1/2$. All these results hold for $10^5 < R < 10^{10}$ and for coverage $0.05 \leqslant \theta \leqslant 0.20$.

All the exponents (χ, χ_c and α) were obtained for $R \gg 1$ and in a region of four to five orders of magnitude in R. Nevertheless, all these must be considered as effective exponents because their values can change in the asymptotic regime $R \to \infty$. Unfortunately, to work with $R > 10^{10}$ is beyond our computational facilities.

It has also been found that the coalescence of islands plays an important role in the growth of islands. The coalescence appears at very low coverage θ and its effects increase with
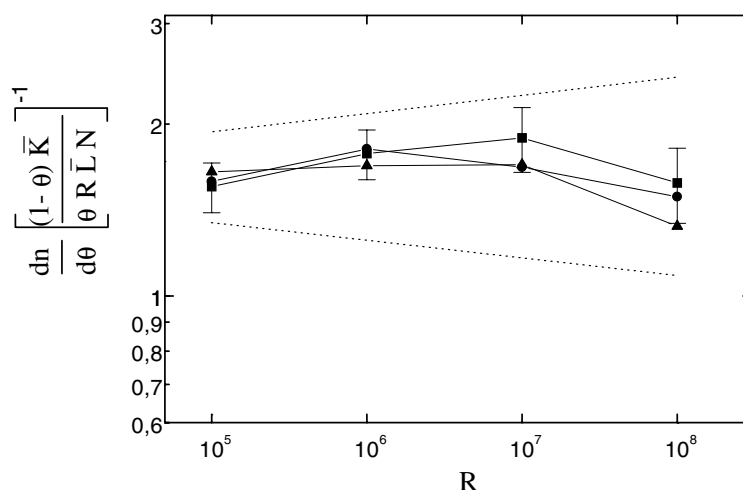
Figure 6. $dn/d\theta[(1-\theta)\overline{K}/\theta R\overline{L}N]^{-1}$ versus R in log–log scales for $\lambda = \infty$ and different values of the coverage θ: $\theta = 0.05$ (squares), 0.10 (circles) and 0.16 (triangles). For $\theta = 0.05$ the error bars are shown and using a least squares fit a straight line with a slope of 0.012 ± 0.024 is obtained. Note that the error bars for each point are larger than those shown in figures 3 and 4. The reason for these large error bars is due to the numerical calculation of $dn/d\theta$. The dashed straight lines have slopes of ± 0.03 and have been drawn to guide the eye.

θ. For larger values of θ, and at a constant value of R, the coalescence rate is greater than the nucleation rate and the density of islands N decreases as a function of θ. This general behaviour was found for all λ (for $\lambda = \infty$ see figure 5). Note that we have worked in the region of θ which is far from the percolation regime and even in this region the coalescence is relevant. To our knowledge, coalescence effects have not been taken into account in previous studies of irreversible aggregation models in two-dimensional substrates at very low coverage. Finally, for the case of $\lambda = \infty$, monomers cannot jump upwards. When a monomer reaches an island it is highly probable that either an aggregation to this island or a coalescence between close islands will occur. Analysing the trajectories of monomers and the probability that they reach an island, an expression for the nucleation rate was found (see equation (9)) which is in agreement with numerical results (see figure 6).

Acknowledgments

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