Effects of the sticking probability on the scaling of the island density in a point island model

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The behavior of the island density exponent χ for a model of deposition, nucleation, and aggregation of particles, forming point islands with a sticking probability p in one dimension, is analyzed. Using Monte Carlo simulation we found that χ depends on p. For $p=1$ we obtain $\chi \cong \frac{1}{4}$, the well-known result for perfect sticking and one-dimensional diffusion. Interestingly, as p is decreased, χ adopts higher values. Possible reasons for this behavior are addressed. The universal result for a one-dimensional diffusion, $\chi = \frac{1}{4}$, is expected to be recovered, for all p, only in the asymptotic regime.

I. INTRODUCTION

The study of submonolayer deposition on surfaces has attracted a great deal of interest in recent years. From an experimental point of view, this is due to the recent development of scanning tunneling microscopy, which allows us to analyze the island morphology generated by the molecular beam epitaxy technique at very low coverage [1–3]. In order to understand the microscopy mechanics responsible for the formation of these island morphologies, several models of deposition, diffusion, nucleation, and aggregation of particles forming islands in a two-dimensional substrate were recently developed and studied [3–10]. From a basic theoretical point of view, the studies focused on finding scaling laws in the island density distributions and scaling functions in the island size distributions [4,6–9,11–19]. In all these models, particles are deposited at random with a constant flux and, after deposition, each particle performs a random walk. *The end of the diffusion for a particle occurs when it sticks to another diffusing particle* (a nucleation process takes place, and a new island is created) or to a preexisting island (an aggregation process). A time-independent parameter of the models is the ratio R between the mean number of jumps per unit of time performed by an isolated particle and the mean number of deposited particles per substrate site and unit of time. Starting with an empty substrate, the final island structure for each specific model depends on the final coverage of particles on the substrate θ and R. In most models, islands can neither break nor diffuse. In the following we will restrict ourselves to immobile islands and irreversible nucleation and aggregation.

When R increases, each particle performs on average a greater number of hops before the incoming of new particles. Then they have a greater probability of reaching pre-existing islands than forming new ones. At a fixed coverage θ, and for large enough values of R, the island density N (the average number of islands per site) satisfies the following power law [6–9]

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

where χ is the so-called *island density exponent*. In many models the nucleation and aggregation processes take place at the first encounter of a particle with another particle or island. In other words, the sticking is *perfect* (i.e., its occurs with probability one).

For the case of islands occupying single lattice sites (point islands) and perfect sticking, the following was found: (i) $\chi = \frac{1}{4}$ when the deposited particles perform a one-dimensional diffusion between its deposition and its attachment to another particle or island, and (ii) $\chi = \frac{1}{3}$ when the particles perform a two-dimensional diffusion [6,7]. The one- or two-dimensional character of the diffusion can be determined by computing the mean value $S(n_s)$ of different visited sites by a deposited particle after n_s steps. For n_s around the mean value of steps $\langle n_s \rangle$, performed between its deposition and the end of diffusion, $S(n_s)$ behaves as n_s^α, where $\alpha = \frac{1}{2}$ or 1 for one- or two-dimensional diffusion, respectively. Interestingly, some models show an intermediate behavior, with $\frac{1}{2} < \alpha < 1$. For example, in a two-dimensional substrate with randomly distributed repulsive impurities (a case in which the available space for diffusion is reduced), an exponent $\frac{1}{4} < \chi < \frac{1}{3}$ was found in an intermediate regime of R [11]. Another interesting case arises when the diffusion is anisotropic in a two-dimensional substrate (i.e., the diffusion constant in one direction is much greater than the diffusion constant in the perpendicular direction). Due to this anisotropy, $\alpha = \frac{1}{2}$ for small n_s, and $\alpha = 1$ for large n_s. As the density of islands N decreases when R increases [see Eq. (1)], $\langle n_s \rangle$ increases because the particles need a larger number of steps to stick to an island. Then a crossover from a one-dimensional regime (at intermediate values of R) to a two-dimensional regime (at very large values of R) was found [7].

In Ref. [18] the case of finite-size islands consisting of linear chains of monomers oriented in one direction of a square lattice was studied. The growth sites of an island are located at both ends of the monomer chain, when a particle arrives at these sites, it always aggregates to the island. Also, nucleation takes place at the first encounter of two diffusing particles. Then we are dealing with a model of perfect sticking. The lateral sides of these islands act as obstacles, block-

ing the diffusion in the perpendicular direction of the chains. The effects of diffusional anisotropy, where diffusion is easier in the direction perpendicular to chains than in the direction of the chains, were analyzed. It was found that, in an intermediate region of R, the exponent χ changes smoothly from $\frac{1}{3}$ to $\frac{1}{4}$ when the diffusion changes from isotropic ($\alpha \cong 1$) to highly anisotropic ($\alpha \cong \frac{1}{2}$). It is expected that in the asymptotic regime, $R \rightarrow \infty$, $\chi = \frac{1}{4}$ independently of anisotropy [19]. The value of the exponent $\chi = \frac{1}{4}$ for both the intermediate and asymptotic regimes is due to the presence of long monomer chains, because the particles are obliged to perform a one-dimensional diffusion.

In the present paper we show that the above-mentioned non universal behavior of the exponent χ, in the intermediate region of R, can also be obtained with an imperfect sticking. More specifically, we introduce a point island model in a one-dimensional lattice in which the nucleation and aggregation processes take place with a sticking probability $p(0 < p \leq 1)$. We found that χ depends on p. However, the reason for this dependency is quite different from those for the above-mentioned examples. As we are working with a one-dimensional lattice, the particles always perform a one-dimensional diffusion (i.e., $\alpha = \frac{1}{2}$). In this paper we explain why the sticking probability p is responsible for the nonuniversal behavior of χ.

II. MODEL

The model is defined on a one-dimensional lattice with periodic boundary conditions (a ring) in order to avoid edge effects. In the simulation we use lattices of at least 10^6 sites. Once deposited, the particles perform random walks and form islands. The islands are immobile, and are composed of two or more particles that occupy a single site (i.e., we are dealing with a point island model). The islands cannot break and grow irreversibly by aggregation. The formation of a new island and aggregation take place with a probability $p(0 < p \leq 1)$. More specifically, the rules of the deposition, diffusion, nucleation, and aggregation processes are as follows.

(i) Deposition: each empty site of the lattice is occupied by a new particle with probability ε, per unit of time t.

(ii) Diffusion: an isolated particle (i.e., a particle not bounded to an island) attempts to jump to any of its nearest neighbor (NN) sites with probability $q = \frac{1}{2}$ per unit of time t.

(iii) Nucleation: if, as a consequence of diffusion, a particle tries to jump from a site i to a NN site occupied by a another isolated particle, these two particles nucleate with a probability p, forming a new island of two particles. With a probability $1-p$ the first particle jumps over the second, arriving at the NN site of the second particle opposite to site i. If there are two or more consecutively occupied sites, the particle continues jumping in the same direction, until it finds the first empty site where it is deposited.

(iv) Aggregation: if, as a consequence of diffusion, a particle tries to jump from site i to a NN site occupied by an island, this particle aggregates to the island with probability p, increasing the number of particles in the island by 1. With a probability $1-p$, the particle jumps over the island and
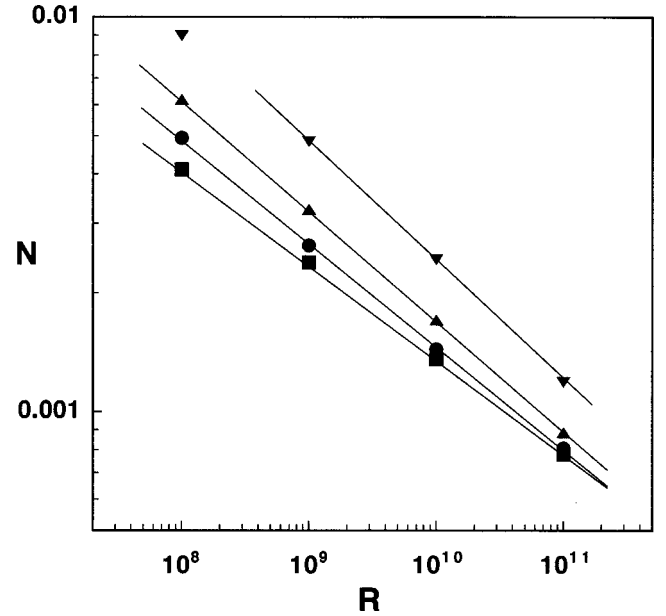

FIG. 1. The island density for different sticking probabilities p as a function of R. Squares correspond to $p=1$, circles to $p=0.02$, triangles to $p=0.005$, and inverted triangles to $p=0.001$. The island density exponent χ is 0.241 ± 0.005, 0.257 ± 0.005, 0.281 ± 0.005, and 0.305 ± 0.005 for $p=1$, 0.02, 0.005, and 0.001, respectively.

arrives at the NN site of the island opposite to site i. If there are two or more consecutively occupied sites, the particle continues jumping in the same direction until it finds the first empty site where it is deposited.

III. RESULTS AND DISCUSSIONS

The island density N as a function of R in log-log scales for different values of p, extracted from our Monte Carlo simulations, are shown in Fig. 1. From the slope of the straight lines we found χ varying from 0.241 ± 0.005 for $p=1$ to 0.305 ± 0.005 for $p=0.001$. Note that for $p=1$ we obtain a value of χ very close to $\frac{1}{4}$ which, as mentioned, is the expected value for the island point model in one dimension with a perfect sticking probability. The numerical result is slightly lower than $\frac{1}{4}$, as previously reported [6,7]. The analytical result $\chi = \frac{1}{4}$ can be reached only for extremely large values of R.

The data of Fig. 1 clearly show that the behavior of N as a function of R strongly depends on p. We now explain why for large enough values of R (not shown in the figure) the effects of p must finally become irrelevant. Let us consider a one-dimensional lattice where the sites are located at $m = 0, \pm 1, \pm 2, \pm 3,...$ Let $W(N_s, m)$ be the average number of times that a site m has been visited by a random walker after N_s steps, starting at the origin $m=0$. At each step the random walker jumps to any of its NN sites with a probability $\frac{1}{2}$. Then [20]

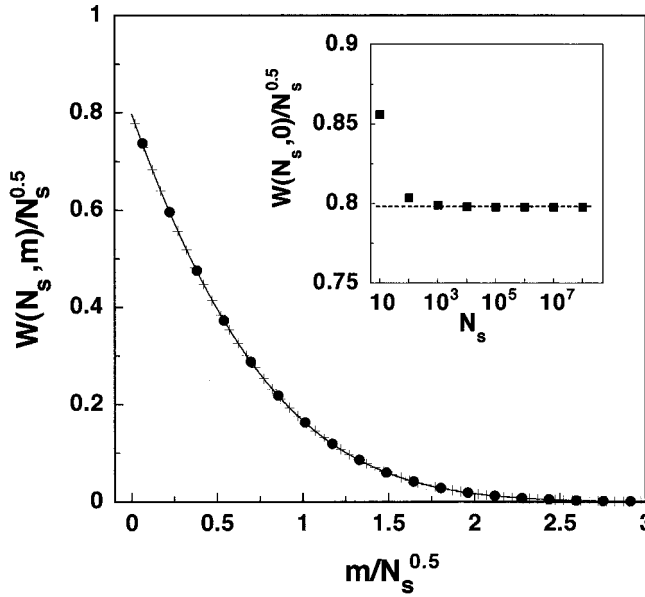$$W(N_s, m) = \sum_{M \geq m}^{N_s} \frac{M!}{[(M-m)/2]![(M+m)/2]!}(1/2)^M,$$

(2)

FIG. 2. The average number of times divided by $N_s^{0.5}$ that a site m is visited by a random walker that starts at the origin after N_s steps. The curve corresponds to $N_s = 10^5$, circles correspond to $N_s = 10^3$, and crosses correspond to $N_s = 10^4$. The inset shows the average number of times divided by $N_s^{0.5}$ that the starting site ($m = 0$) is visited as a function of N_s. Note that the same value is obtained for $N_s \gtrsim 10^3$.

where the sum runs over even values of M, if m is even, or odd numbers of M, if m is odd. In Fig. 2, $W/N_s^{1/2}$ is shown as a function of $m/N_s^{1/2}$ for $N_s = 10^3$, 10^4, and 10^5, and $m \geq 0$. For large values of M, each term of the sum can be approximated by a Gaussian distribution, and the sum by an integral [20]. After an appropriate change of variables, $W/N_s^{1/2}$ is a function of $m/N_s^{1/2}$, in agreement with the data collapse shown in Fig. 2. Let $m_1(N_s)$ be the maximum value of m for which $W(N_s,m) \geq 1$ holds, or equivalently $W(N_s,m_1 + 1) < 1$. For $-m_1 \leq m \leq m_1$ all sites have been visited at least once. Then $m_1 = S(N_s) \sim N_s^{1/2}$.

Let us now consider the aggregation process in the island point model which is dominant in the range of interest because $N \gg n$, where N and n are the density of islands and monomers, respectively. A deposited particle that, after a diffusion process, arrives for the first time at a NN site of an island i, has a probability p to aggregate. Before a successful aggregation takes place, this particle will attempt to aggregate on average, $1/p$ times. For example, one possibility is that the particle tries λ/p times to aggregate to island i and $(1-\lambda)/p$ times to aggregate to another island j $(0 \leq \lambda \leq 1)$. For a given value of p we define N_{sp} such that $1/p \cong W(N_{sp},0)$. Let $l_p = m_1(N_{sp}) \sim N_{sp}^{1/2}$. From Fig. 2, $W(N_{sp},0) \sim N_{sp}^{1/2}$ for large N_{sp} and then $l_p \sim (1/p)$, for small p. Let us assume that we are working with a value of R such that the average distance between consecutive islands is $l = 1/N \gg l_p$. In this condition it is highly probable that the particle aggregates to the first found island (i.e., the island i) since the distance between this island and any other one (as island j) is very large. In other words, the particle will attempt to aggregate to the first island $1/p$ times before finding

any other island (i.e., $\lambda = 1$). As m_1 and $W(N_s,0)$ increase with N_s, aggregation is not expected for $N_s < N_{sp}$ for the above-specified values of p and R. The fact that aggregation occurs at the first found island, as in the case of $p = 1$, holds for even larger values of R, because the average distance between consecutive islands increases with R [note that $l = 1/N$ and N decreases with R; see Eq. (1)]. Also l_p is the maximum distance that the particle can travel after it arrives at the first island. Then, for $l \gg l_p$ the effects of p become irrelevant, and the same value of the island density N must be obtained for $p = 1$ and $p < 1$. The behavior of the smaller value of R, denoted by R_c, at which the collapse occurs can be estimated as follows. At R_c, the mean distance between islands, l_c, must be $l_c \gg l_p$. That is, $l_c = a l_p$, with $a \gg 1$. For $l \sim l_c$ we have the one-dimensional behavior $l_c \sim R^\chi$, with $\chi = \frac{1}{4}$. Then, $R_c \sim (a/p)^4$ for small values of p. Although the confirmation of this relation is beyond our computational facilities, the numerical results shown in Fig. 1 give support to all the above reasoning. From this figure we expect, for large enough values of R (not shown in the figure), that the curves of N corresponding to $p < 1$ and $p = 1$ collapse onto one curve. For decreasing values of p the collapse will occur at increasing values of R.

In order to understand the behavior of the exponent χ for different values of p, we compute $\Delta x/l$, where Δx is the square root of the mean-square displacement performed by a particle between its deposition and the end of the diffusion due to nucleation or aggregation. As $n \ll N$, the value of Δx is dominated by aggregation. For $p = 1$ the ratio $\Delta x/l$ is not expected to depend on l. The particle is deposited between two islands and then, since $p = 1$, it cannot escape from the region limited by these two islands. For $p < 1$ the particle can escape and aggregate to other islands outside this region. Therefore, for a specific value of l, $\Delta x/l$, corresponding to $p < 1$, is greater than or equal to $\Delta x/l$ corresponding to $p = 1$. In Fig. 3 we plot $\Delta x/l$ as a function of l in log-log scales for different values of p. We expect that for large enough values of l all these curves will collapse because, as l becomes greater than l_p, the particle, as in the case of $p = 1$, will not escape from a region limited by the two consecutive islands where it was deposited. From Fig. 3, we can write

$$\Delta x/l \sim l^{-\varepsilon}, \qquad (3)$$

where ε is an effective exponent which depends on p. From the above-mentioned collapse of $\Delta x/l$, it can be expected that $\varepsilon \to 0$ when $l \to \infty$ for all values of $p > 0$.

We now focus on the relation between the exponents ε and χ. Due to nucleation and aggregation processes, the density of monomers, n, decreases per unit of time as n/τ, where τ is the lifetime of monomers. Since we are dealing with a point island model, the probabilities of nucleation and aggregation of a monomer are $P_n \sim n/(n+N)$ and $P_a \sim N/(n+N)$, respectively. Note that these relations hold for all values of p because, in our model, the nucleation and aggregation are affected in the same way by p. Assuming $n \ll N$, as occurs for large values of R, one has [6,11,18]
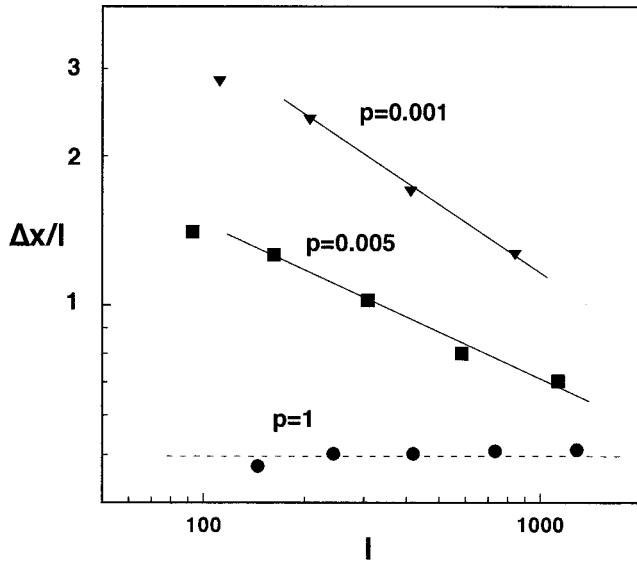
FIG. 3. The ratio of the square root of the mean-square displacement in the x direction and the average distance between consecutive islands l as a function of l for different values of p. The exponent ε [see Eq. (3)] is 0.31 ± 0.07 for $p = 0.005$ and 0.44 ± 0.07 for $p = 0.001$. The region of l shown corresponds to $10^7 \leq R \leq 10^{11}$ for $p = 1$ and 0.005 and $10^8 \leq R \leq 10^{11}$ for $p = 0.001$. Δx and l are measured in terms of the distance between nearest-neighbor sites of the lattice.

$$\frac{dn}{dt} \cong \varepsilon - \frac{n}{\tau}, \tag{4}$$

$$\frac{dN}{dt} \cong (n/\tau)(n/N). \tag{5}$$

The average number of steps $\langle n_s \rangle$ performed by a deposited particle until the end of the diffusion, due to nucleation or aggregation processes, is

$$\langle n_s \rangle = 2q\tau = \varepsilon R \tau. \tag{6}$$

For $R \gg 1$ a quasistationary regime exists for which $dn/dt \sim 0$ (which is verified numerically). From Eqs. (4)–(6), and using $d\theta = \varepsilon dt$, one obtains

$$\frac{dN}{d\theta} \cong \frac{\langle n_s \rangle}{RN}. \tag{7}$$

Considering $\langle n_s \rangle \propto \Delta x^2$ and $l = 1/N$, Eq. (7) can be written as

$$\frac{dN}{d\theta} \sim \frac{1}{RN^3}\left[\frac{\Delta x}{l}\right]^2. \tag{8}$$

N cannot be expressed as a function of R and θ integrating Eq. (8) from $\theta = 0$, because $\Delta x/l$ is an unknown function of θ and because for $\theta \cong 0$ this equation is no longer valid (for example, the inequality $n \ll N$ is not true). Now, assuming that, in the region of interest, $0.03 \leq \theta \leq 0.15$, the exponent χ does not depend on θ [i.e., $N = f(\theta)R^{-\chi}$, which numerically

was found as a very good approximation], $dN/d\theta \sim N$. From Eqs. (3) and (8) the relation $N \sim R^{-\chi}$ is obtained, with

$$\chi = 1/(4 - 2\varepsilon). \tag{9}$$

As already mentioned, $\Delta x/l$ does not depend on l for $p = 1$ (i.e., $\varepsilon = 0$; see Fig. 3), and Eq. (9) predicts the well-known one-dimensional exponent $\chi = \frac{1}{4}$ for perfect sticking. For other values of p this equation is in good agreement with numerical results. For example, from the slopes of the straight lines of Fig. 3 we obtain $\varepsilon = 0.31 \pm 0.07$ and 0.44 ± 0.07 for $p = 0.005$ and 0.001, respectively. Using Eq. (9) we can determine $\chi = 0.30 \pm 0.02$ and 0.32 ± 0.02. These results must be compared with $\chi = 0.281 \pm 0.005$ and 0.305 ± 0.005 determined from Fig. 1.

We also expect that $\varepsilon \to 0$ for $l \to \infty$ (i.e., $N \to 0$) which from Eq. (9) implies that, in the asymptotic regime, $R \to \infty$, the one-dimensional exponent $\chi = \frac{1}{4}$ is recovered for all values of $p > 0$. Finally, note that for $p = 0$ there is neither nucleation nor aggregation in the model ($N = 0$ for all R), and then the limit $p \to 0$ is different from the case $p = 0$.

IV. SUMMARY AND CONCLUSIONS

In this paper a point island model with a sticking probability $0 < p \leq 1$, that affects both, the nucleation and aggregation processes, was introduced and studied. In an intermediate region of R, of four orders of magnitude, the island density exponent χ was found to depend on p (see Fig. 1). For $p = 1$ we obtain $\chi \cong 0.24$, which is in agreement with the well-known result $\chi = \frac{1}{4}$ for perfect sticking and one-dimensional diffusion. Although the deposited particles perform a one-dimensional diffusion for all values of p, the exponent χ increases when p decreases.

For $p < 1$, the particles have a probability to escape from the region limited by two consecutive islands where they were deposited. With the arguments of Sec. III and the analysis of the function W (see Fig. 2), it can be concluded that, for a fixed value of p, this probability decreases as the average distance between consecutive islands l increases. Then $\Delta x/l$ decreases with l (see Fig. 3), and from Eqs. (3) and (9) an exponent $\chi > \frac{1}{4}$, that depends on the specific value of p, is obtained.

In the asymptotic regime, $R \to \infty$, it is expected that $\chi = \frac{1}{4}$ for all values of p; then the universality is recovered. In short, before a successful attachment occurs, the particle tries to aggregate, on average, $1/p$ times. With the function W, we showed that for large values of R the mean distance between islands becomes so large that the deposited particle must stick to the first island that it finds, as in the case of $p = 1$.

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