# Growth model with disaggregation of islands having an odd number of particles

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A simple model of deposition of particles and growth of point islands in a two-dimensional substrate is introduced and studied. The detachment of particles from islands with an odd number of particles can occur with a probability $P$. The power-law scalings of the island, monomer, and odd island densities are analytically obtained and verified by Monte Carlo simulations. The universality class of the model depends on $P$, and the island density exponent $\chi$ changes from $\chi=1/3$ (for $P=0$) to $\chi=0$ (for $P>0$).

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## I. INTRODUCTION

The fundamental physical processes in the growth of thin films by deposition techniques have been studied from the theoretical and experimental points of view. In particular, the submonolayer deposition on surfaces has attracted a great deal of interest in recent years and scaling laws related to the formed islands have been specially addressed. These studies are important as the island density and distribution may be used to determine a variety of important microscopic parameters in epitaxial growth [1–3].

The recent development of analytical tools such as the scanning tunneling microscope allows us to analyze the island morphology generated at very low coverage. In these studies, semiconductors have attracted a particular interest. In this context the homoepitaxial growth of silicon on Si(100), due to its technological relevance, has been specially studied [4,5]. In the Si/Si(100) system, deposited adatoms diffuse and nucleate when two of them meet forming dimers in a direction perpendicular to the substrate dimers. Islands form by the aggregation of new adatoms to the already formed dimer to form chains. Figure 1 schematically shows a dimer $A$, a dimer chain $B$, and the possible growing sites. Dimers are very stable; their breaking probability is negligible at temperatures below 400 K [6]. In contrast, a single adatom aggregated to an island (this is the case of the particle below site $i$ in chain $B$ of Fig. 1) will detach if another adatom does not arrive (to site $i$) to form a new dimer within the residence time of the single adatom [5]. This detachment delays the growth of islands and modifies its size distribution as the detached adatom can diffuse and form a new island if it meets another diffusing adatom.

Motivated by the detaching mechanism described above, in the present paper we study its effects from a basic theoretical point of view. Our goal is to analyze the scaling properties of the densities of islands and monomers as modified by detachment from islands. To obtain analytical results, we introduce a simple point island model. Interestingly, it is found that scaling laws are radically different from those for a model without detachment.

## II. MODELS

We use two models. One was formulated in a previous work [6] in connection with the growth of silicon dimer chains on Si(100). This model takes into account some characteristics which appear in real systems, such as the anisotropic diffusion and the inhomogeneity of the substrate. Since in the present work we are interested in the detachment effects, a simplified version of that model with isotropic diffusion and homogeneous substrate will be used. In the following this simplified version will be called *dimer model*. In order to obtain analytical results, a second model where islands occupy a single lattice site will be introduced and analyzed. In this *point island model* the detachment rules are defined in agreement with those of the dimer model.

In both models the substrate is represented by a square lattice of $3000\times3000$ sites. Periodic boundary conditions were adopted in order to avoid edge effects. We will assume

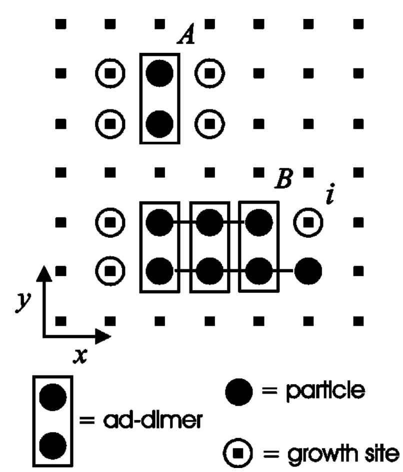


FIG. 1. The substrate (solid square) and the growth of dimer chains. Two particles that become nearest neighbors in the $y$ direction nucleate forming a dimer as shown in structure $A$. Next, particles can aggregate in growth sites to finally form a chainlike structure $B$. Particles not forming dimers can detach with probability $p$ per unit of time, as the one just below the $i$ growth site in the chain $B$. The structure $B$ corresponds to an island with incomplete dimers.

 

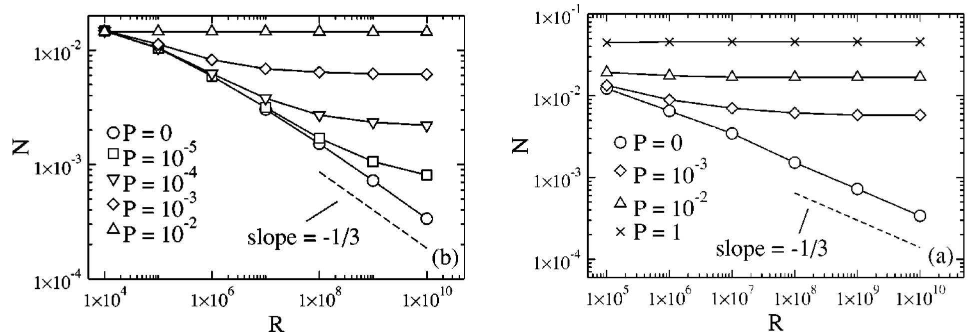


FIG. 2. The island density $N$ against the parameter $R$, in log-log scales, for different detachment probability $P$ (a) corresponds to numerical results obtained from the dimer model and (b) from the point island model, with $\Theta=0.1$ and $\Theta=0.05$, respectively. One can see similar behaviors of $N$ versus large $R$ for both models: (i) for $P\neq 0$, $N$ reaches a constant value that depends on $P$, and (ii) for $P=0$, $N\sim R^{-\chi}$, with $\chi=1/3$ (this result was found for other two-dimensional models with isotropic diffusion [7–11]). The dashed straight lines have slope $-1/3$.

that islands cannot diffuse and that the following processes take place: (a) deposition, (b) diffusion, (c) nucleation, (d) aggregation, and (e) detachment.

The rules for the dimer model are as follows.

(a) Deposition: each empty site of the lattice is occupied by a new particle with probability $\epsilon$ per unit of time.

(b) 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 $q$ per unit of time $t$. If the particle attempts to jump to an occupied site, the jump is not performed and the particle remains at its original position.

(c) Nucleation: if, as a consequence of diffusion or deposition, a particle arrives at an NN site in the $y$ direction of a second isolated particle, these two particles nucleate, forming a dimer. This dimer plays the role of a seed for the dimer chain.

(d) Aggregation: Fig. 1 shows growth sites for different structures. If a diffusing or deposited particle arrives at a growth site, it sticks, increasing the number of particles of the island by 1.

(e) Detachment: each particle of an island which does not belong to a dimer can detach with probability $p$ per unit of time, but particles forming dimers cannot detach. For example, the particle below the $i$ growth site in the chain $B$ of the Fig. 1 can detach from this island. The detached particle is placed on one of the NN empty sites of its original position selected at random or remains at its original site if the NN site selected is occupied.

In the point island model, an island is composed of $s$ particles ($s\geqslant 2$) that occupy a single lattice site. The rule of deposition for this model is the same as for the dimer model. Diffusion is also the same with the exception that if a particle attempts to jump to an occupied site by other particle, these two particles nucleate, forming a new point island with $s=2$ (this is the rule of nucleation). If the particle attempts to jump to an occupied site by a point island, it sticks, increasing the number of particles $s$ of the island by 1 (i.e., $s\rightarrow s+1$, aggregation rule). Ultimately, one particle of an island with an odd number of particles (i.e., $s=3,5,\ldots$) can detach from this island with probability $p$ per unit of time (rule of detachment). The detached particle is shifted to one of the four NN sites of the selected island at random. Particle belonging to islands with an even number of particles cannot detach (even islands cannot break).

Simulations start with the lattice empty and run until the particle density $\Theta$ (i.e., the mean value of particle per lattice site) reaches a desired value. We are interested in coverage low enough to avoid the percolation regime for extended island models [8]. So, the employed values for $\Theta$ are below 0.15. Although, for a fixed particle density, the models present three variable parameters, it is expected that all processes depend only on the relative value of these parameters. Then, there are only two dimensionless independent control parameters, which we have chosen as $R=4q/\epsilon$ and $P=p/4q$. Monte Carlo results were obtained averaging typically over ten samples.

## III. RESULTS

Figure 2(a) shows the island density $N$ (i.e., the average number of island per lattice site) as a function of $R$ for different values of time-independent detaching probability $P$, in log-log scales, for the dimer model. When $R$ increases and $P>0$, the island density reaches a constant value, which depends on $P$. Let us note that there is a different behavior for $P=0$ than for other value of $P\neq 0$, including $P$ close to 0. In the point island model, $P$ only affects the odd islands, i.e., a particle of an odd island can separate from this island with probability $P$, but if the particle belongs to an even island it cannot detach. In the dimer model, islands with completed dimer cannot break (see Fig. 1) and they are represented by the even islands in the point island model. Islands in the dimer model, with incomplete dimers in one or both of their ends, are represented by odd islands in the point model. Figure 2(b) shows $N$ versus $R$ for the point island model in log-log scales and different values of $P$. The behaviors of $N$ as a function of $R$ for different values of $P$ and low particle density for both models are similar. In what follows, for the sake of simplicity, we will focus on the point island model.

Let $n_s(t)$ be the density of islands (i.e., the average number of islands per lattice site) with $s$ particles, at time $t$. Note that $n_1(t)$ is the monomer density at time $t$. The total densities of even islands $N_E$ and odd islands $N_O$ are given by

$$N_E = \sum_{s_{even}} n_s(t) \quad \text{and} \quad N_O = \sum_{s_{odd}} n_s(t), \tag{1}$$

where $s_{even}(s_{odd})$ runs over all even (odd) values of $s$, with $s \geqslant 2$ (i.e., $s_{even}=2,4,\ldots;\ s_{odd}=3,5,\ldots$).

It is possible to derive a set of differential equations for $n_s$ by analyzing all possible processes that can take place in a time interval $dt$. Thus,

$$\frac{dn_1}{dt} = \epsilon[1-(n_1+N_E+N_O)] - kn_1^2 - n_1 k\sum_{s\geqslant 2} n_s + p\sum_{s_{odd}} n_s, \tag{2}$$

$$\frac{dn_s}{dt} = n_1 k(n_{(s-1)} - n_s) + pn_{(s+1)}, \quad s=2,4,6,\ldots, \tag{3}$$

$$\frac{dn_s}{dt} = n_1 k(n_{(s-1)} - n_s) - pn_s, \quad s=3,5,7,\ldots, \tag{4}$$

where $\epsilon$ is the particle deposition rate per empty site and unit of time, $p$ is the detaching probability from odd islands per unit of time, and $k$ governs the monomer attachment rate to islands. In general, $k$ depends on size island [8] but here we are dealing with point islands. In the following we consider a low particle density regime and then $[1-(n_1+N_E+N_O)]\cong 1$.

Dividing the above equations by $\epsilon$ and summing over all even and odd islands in Eqs. (3) and (4), respectively, Eqs. (2)–(4) can be rewritten as

$$\frac{dn_1}{d\Theta} \cong 1 - \frac{k}{\epsilon}n_1^2 - \frac{k}{\epsilon}n_1 N + \frac{p}{\epsilon}N_O, \tag{5}$$

$$\frac{dN_E}{d\Theta} = \frac{k}{\epsilon}n_1(n_1+N_O-N_E) + \frac{p}{\epsilon}N_O, \tag{6}$$

$$\frac{dN_O}{d\Theta} = \frac{k}{\epsilon}n_1(N_E-N_O) - \frac{p}{\epsilon}N_O, \tag{7}$$

where $N_E$ and $N_O$ are even and odd island densities, respectively [defined in Eq. (1)], $N$ is the total island density, and $\Theta=\epsilon t$ is the particle density, i.e., the number of particles deposited per site ($d\Theta=\epsilon dt$). Summing Eqs. (6) and (7) one obtains

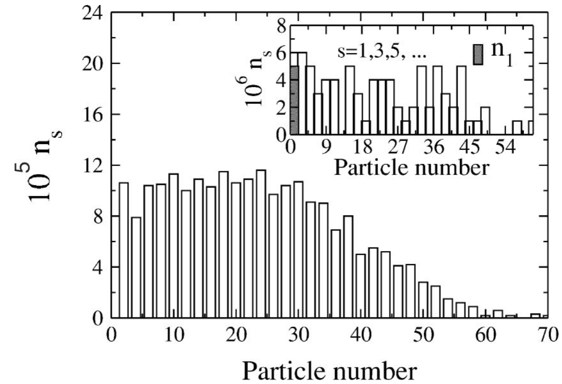


FIG. 3. Even island density distribution ($n_s, s=2,4,6,\dagger$) vs particle number for $R=10^{10}$, time-independent detaching probability $P=p/4q=10^{-4}$, and particle density $\Theta=0.055$. The inset shows the odd island density distribution and the monomer density. Even island density $N_E$ is much greater than odd island density $N_O$ for large values of $R$. $N_E=2.247\times10^{-3}$ while $N_O=7.6\times10^{-5}$. $P$ only affects the odd islands. Particle deposition and odd islands are the source of monomers. These results were obtained for only one sample.

$$\frac{dN}{d\Theta} = \frac{k}{\epsilon}n_1^2. \tag{8}$$

Equation (8) indicates that $N$ grows irreversibly by nucleation. An island forms when two particles meet and will remain as an island, since there is no detachment for even islands.

The solution of these equations, for $k/\epsilon \gg 1$, $p/k \ll 1$, and low particle density, is as follows. When $k/\epsilon \gg 1$, a quasistationary regime exists in which $n_1 \ll N$, $dn_1/d\Theta \cong 0$, [7,12] and also $dN_O/d\Theta \cong 0$ for $p\neq 0$ (these behaviors were verified numerically). Then, from Eq. (5) we can write

$$\frac{k}{\epsilon}n_1 N - \frac{p}{\epsilon}N_O \cong 1. \tag{9}$$

In two dimensions and for isotropic diffusion, the attachment rate of monomers $k\sim D$ [8], where $D$ is the diffusion rate constant of single particles (in unit of hops per unit of

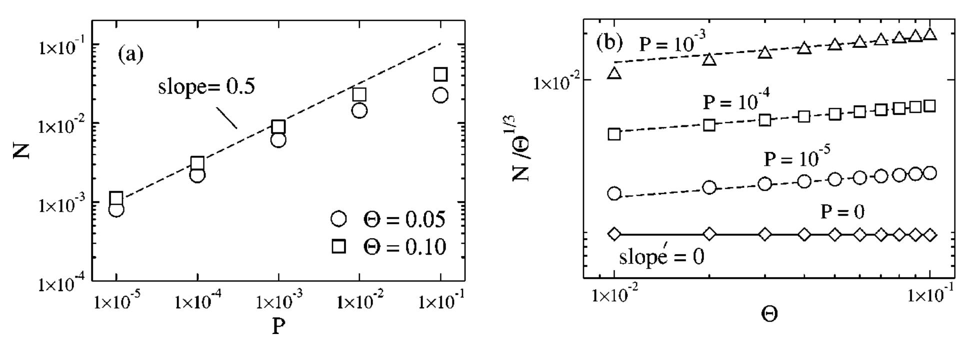


FIG. 4. Log-log plots of (a) the island density $N$ vs the time-independent detaching probability $P$ (for two different values of $\Theta$) and (b) $N\Theta^{-1/3}$ vs $\Theta$ (for different values of $P$). In both cases $R=10^{10}$. In (a), the dashed straight line has slope 0.5, while in (b) dashed lines have slopes $1/6$. These values are in accord with the exponent of Eq. (10). For $P\ll 1$, the exponent does not depend on $\Theta$.

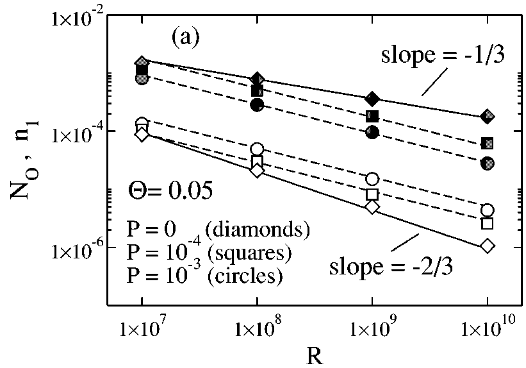


FIG. 5. The odd island density $N_O$ (filled symbols) and the monomer density $n_1$ (open symbols), in log-log scales, vs $R$ for particle density $\Theta = 0.05$. The dashed straight lines have slopes $-0.5$. This value corresponds to the exponent of $R$ in Eqs. (11) and (12).

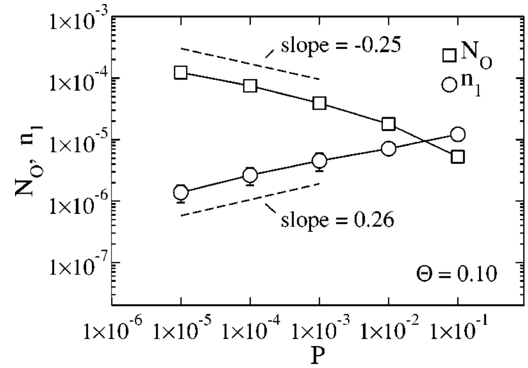


FIG. 7. Plot of numerical results of odd island density $N_O$ (squares) and monomer density $n$ (circles) vs detachment probability $P$, in log-log scales, for $R = 10^{10}$ and $\Theta = 0.1$.

time). Since we defined in Sec. II that $R = 4q/\epsilon$, $k/\epsilon \sim R$. For $p \neq 0$, large values of $R$ (i.e., $k/\epsilon \gg 1$), and large enough particle density, $n_1, N_O \ll N_E$ (see Fig. 3) and then $N = N_O + N_E \cong N_E$. Substituting $N_E$ by $(N - N_O)$ in Eq. (7) (note that $dN_O/d\Theta \cong 0$) and using Eq. (9) one obtains $(k/\epsilon)n_1 N_O \cong 1/2$. Then, $(k/\epsilon)n_1 N \gg 1$ and using Eqs. (8) and (9), $dN/d\Theta \cong p/(2kN)$. Integrating the previous equation from $\Theta = 0$ up to the final value $\Theta$, we find

$$N \sim \left(\frac{p}{k}\Theta\right)^{1/2} \sim (P\Theta)^{1/2}, \qquad (10)$$

where $P$ was defined in Sec. II as $P = p/4q \sim p/k$. Thus, $N$ does not depend on $R$, for $R \gg 1$ and $P \neq 0$, as seen in Fig. 2(b).

When $R \gg 1$ and $p \neq 0$ ($R$ is proportional to the ratio between the diffusion and deposition rates), it is expected that $N$ reaches the possible maximum value, i.e., $N = \Theta/2$. Although $N$ remains essentially constant, it depends on $\Theta$ and also on $P$. In Fig. 4(a), $N$ as a function of $P$, in log-log scales for two values of $\Theta$ and $R = 10^{10}$, is plotted. For $P \ll 1$ numerical results are in accord with Eq. (10). However, as $P$ increases, for low particle density (for example, $\Theta = 0.1$) there are not enough particles to form many islands. In this case, numerical results separate from the straight line with slope 0.5, as it can be seen in Fig. 4(a), and $N$ tends to its maximum value which is lower or equal to $\Theta/2$. In spite of this restriction, $N$ behaves as $\Theta^{1/2}$ for $P \ll 1$, $R \gg 1$, and low particle densities [see Fig. 4(b)] as Eq. (10) predicts.

From Eqs. (8) and (10), $n_1$ can be determined as

$$n_1 \sim \left(\frac{p}{k}\right)^{1/4} \left(\frac{k}{\epsilon}\right)^{-1/2} \Theta^{-1/4} \sim P^{1/4} R^{-1/2} \Theta^{-1/4}. \qquad (11)$$

Finally, from $(k/\epsilon)n_1 N_O \simeq 1/2$ and Eq. (11) it follows that

$$N_O \sim P^{-1/4} R^{-1/2} \Theta^{1/4}. \qquad (12)$$

Figure 5 shows plots of $n_1$ and $N_O$ as functions of $R$ for $\Theta = 0.05$, and Figs. 6(a) and 6(b) show plots of $N_O \Theta^{-1/3}$ and $n_1 \Theta^{1/3}$ versus $\Theta$ for $R = 10^{10}$. The influence of $P$ on the exponents is clearly shown. The numerical simulations confirm the behavior of $n_1$ and $N_O$ with $R (R \gg 1)$ and $\Theta$ given in Eqs. (11) and (12).

In Fig. 7 $n_1$ and $N_O$ are plotted against $P$, in log-log scales, for $\Theta = 0.1$. From the numerical results obtained for $10^{-5} \leq P \leq 10^{-3}$, we found the exponents $-0.25 \pm 0.03$ and $0.26 \pm 0.03$ for $n_1$ and $N_O$ as functions of $P$, respectively [see Eqs. (11) and (12)].

We showed that $(k/\epsilon)n_1 N \gg 1$ [see paragraph above Eq. (10)]. With Eqs. (10) and (11) this inequality becomes equivalent to $RP^{3/2} \gg 1$. Thus, to be consistent with our as-

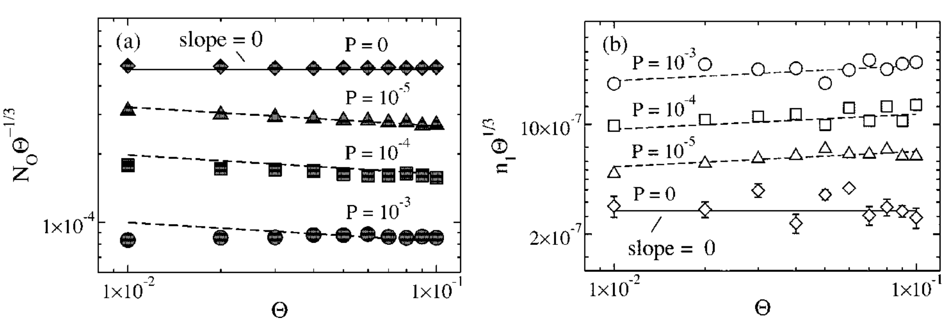


FIG. 6. Numerical results of (a) $N_O \Theta^{-1/3}$ and (b) $n_1 \Theta^{1/3}$, in log-log scales, against $\Theta$, for $R = 10^{10}$. These results correspond to different values of $P$: $P = 0$ (diamonds), $10^{-5}$ (triangles), $10^{-4}$ (squares), and $10^{-3}$ (circles). The dashed straight lines have slopes $-1/12$ and $1/12$ in (a) and (b), respectively. These values are consistent with the exponent of $R$ in Eqs. (12) and (11), respectively.

sumptions, it must be stated that Eqs. (10)–(12) hold in the asymptotic regime $R \gg 1$, $R^{-2/3} \ll P \ll 1$, and low particle density $0.01 \leqslant \Theta \leqslant 0.1$.

The above results correspond to $P \neq 0$. When $P=0$, monomers cannot detach from islands and there is no difference between the behavior of even and odd island densities. In this case $N_O \sim N \sim R^{-\chi} \Theta^{\chi}$ and $n_1 \sim R^{-2\chi} \Theta^{-\chi}$ with $\chi = 1/3$ for $R \gg 1$ and low particle density $(0.05 \leqslant \Theta \leqslant 0.15)$ [7]. These results can be obtained taking $p=0$ in Eqs. (2)–(4) and considering $dn_1/d\Theta \sim 0$ for $R \gg 1$ and low particle density. These results are completely different from those previously obtained [see Figs. 2(b), 4(b), 5, 6(a), and 6(b)]. That is to say, the power-law behaviors of $N$ and $n_1$ with $R$ and $\Theta$ (in the limit $R \to \infty$ and low particle density) for time-independent detachment probability $P=0$ and $P \to 0$ [see Eqs. (10) and (11)] are different. In other words, $P$ is a relevant parameter in the sense that for $P=0$ and $P>0$ the point island model belongs to different universality classes.

The obtained Monte Carlo results can be qualitatively extended to the dimer model, as long as the mean size of the islands is not too large. Then, for this model we expect that the scaling laws of Eqs. (10)–(12) hold, where $N_O$ corresponds to the island density of incomplete dimer chains.

## IV. FINAL REMARKS

We have introduced a model in which islands with an odd number of particle can disaggregate with probability $P$. Interestingly, we found that the cases $P=0$ and $P>0$ belong to different universality classes. The first case corresponds to a model with irreversible aggregation and in the second one detachment is allowed. There are other models that include the disaggregation of particles from islands, for example, the case in which islands with more than $j$ particles are stable was considered (for more details see Refs. [2,9]). In this case, for $j=1$ all islands are stable, for $j=2$ only dimers can break; and for $j=3$ only dimers and trimers can break. Considering isotropic sticking and islands with finite size, the island density behaves as $N \sim R^{-\chi_j}$, where $\chi_j = j/(j+2)$ is the so-called *island density exponent*. Thus, in this model, the breaking of island implies the increase of $\chi_j$ ($\chi_j$ increases with $j$). Let us stress that, conversely, the island density exponent $\chi$ of our model behaves in the opposite direction. For irreversible aggregation $(P=0)$ $\chi = 1/3$, and when detachment is allowed $(P>0)$ $\chi = 0$ [see Eq. (10) and Fig. 2(a)].

In summary, a model of deposition, nucleation, aggregation, and detachment of particles in a two-dimensional substrate is introduced and studied. For $P>0$ this model belongs to a new universality class. In the asymptotic regime $(R \gg 1)$ and for small enough values of $P$ and $\Theta$, the theoretical found power-law scalings for the island, monomer, and odd island densities [see Eqs. (10)–(12)] are in agreement with Monte Carlo results [see Figs. 4–7].

## ACKNOWLEDGMENTS

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