Island Size Distribution with Hindered Aggregation

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Abstract

The study of the effect of hindered aggregation on the island formation process in one-dimensional epitaxial growth. In the proposed model the aggregation of monomers to stable islands is hindered by an additional aggregation barrier, ϵ_a, which decreases the diffusion rate to those islands. As ϵ_a increases the system exhibits a crossover between two different kind of processes, diffusion-limited aggregation (DLA) and attachment-limited aggregation (ALA). The island size distribution, $P(s)$, is calculated by using a self-consistent (SC) set of equations for the capture kernels. We test our analytical model with extensive numerical simulations and previously established results.

1. Introduction

Growth processes provide interesting non-equilibrium phenomena which have been the subject of several studies in recent years. Basically, growth mechanisms involve three different processes: Nucleation, aggregation and mass transport of basic growth units such as atoms and molecules. From now on we will call these basic growth units as monomers.

In standard models of epitaxial growth, the monomers are deposited on the substrate at a constant (controlled) rate F. The temperature of the substrate is usually chosen in such away that the evaporation of deposited monomers can be neglected. Consequently, the growth rate is controlled externally: $\theta = F\,t$ (number of deposited monomers). The critical nucleus size "i" is defined as the size of the largest unstable cluster. Only the islands consisting of $i+1$ or more monomers are completely stable. Islands smaller than $i+1$ are unstable, i.e., the monomers belonging to such islands can diffuse away with diffusion constant $D = D_0 e^{-E_D/K_B T}$. After deposition, the monomers diffuse on the substrate with diffusion constant D and eventually will reach a cluster of monomers with a given size s. If $s = i$ a new island will be formed (nucleation) but if $s > i$ the monomer will attach to the cluster (aggregation). In the point-island model, the islands just occupy one site in the lattice, and their size is simply the number of monomers which belong to the cluster. In contrast, in the extended-island model the islands grow laterally.

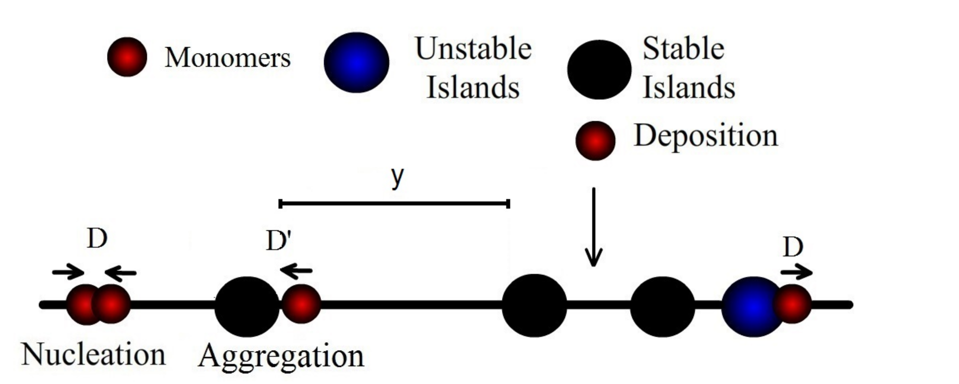

Figure 1: *Cartoon of the system for the point-island model. Two basic reactions are involved: Nucleation and Aggregation. The length of the gap between island edges y is also indicated.*

2. Hindered Aggregation

We consider a model where the absorption of monomers by stable islands is hindered by an additional attachment barrier ϵ_a which decreases the diffusion constant to D'. The associated characteristic length ($D/D' = l_a + 1$) is usually defined to measure the asymmetry between D and D'.

We define the following time scales: the average time between two consecutive deposition events, τ_{dep}, the typical time that a monomer spends inside of a gap, τ_{res}, and the time required for a monomer to reach one of the ends of the gap, τ_{tr}. The behavior of the system depends on the relative value of these time scales. Finally, let L be the average size of the gap between islands.

For weak barriers, ($l_a \ll L$), the aggregation is diffusion limited (DLA) $\tau_{dep} \gg \tau_{res} \approx \tau_{tr} \gg \tau_{dis}$ while for strong barriers, ($L \ll l_a$), the aggregation is attachment limited (ALA) $\tau_{dep} \gg \tau_{res} \gg \tau_{tr} \gg \tau_{dis}$.

The main objective of this work is to calculate the density of stable islands with size s, $P(s)$ for different values of l_a and i.

3. Analytical Approach

As usual, N_1 is the average density of monomers and N_s is the average density of islands with size s. The time evolution of N_1 and N_s can be determined from classical nucleation theory:

$$\frac{dN_1}{d\theta} = \gamma - (i+1)\Re\sigma_u N_1^{i+1} - \Re N_1 \sum_{s\geq i+1}\bar{\sigma}_s N_s = 1 - \Re N_1/\xi^2 \qquad (1)$$

and

$$\frac{dN_s}{d\theta} = \Re N_1(\bar{\sigma}_{s-1}N_{s-1} - \bar{\sigma}_s N_s) = \Re N_1/\left((i+1)\xi_u^2\right). \qquad (2)$$

where γ is the fraction of the substrate which is not covered by island ($\gamma = 1-\theta+N_1$ and $\gamma = 1-N$ for extended-island and point-island models, respectively). The quantities σ_u and σ_s are the capture kernels for unstable and stable islands ,respectively. Finally, ξ_u and ξ are the capture lengths associated to the capture kernels. The second term of Eq.(1), represents nucleation while the third one aggregation. The first and second terms of Eq.(2), represents the aggregation of monomers to islands with size $s-1$ and s, respectively.

In order to calculate N_s it is necessary to find $\bar{\sigma}_s$ and $\bar{\sigma}_u$ as follows. The time evolution of the density of monomers at the position x inside a single gap with length y, $n_1(x,\theta)$, is given by:

$$\frac{\partial n_1(x,\theta)}{\partial\theta} = 1 + \Re\frac{\partial^2 n_1(x,\theta)}{\partial x^2} - \Re\frac{n_1(x,\theta)}{\xi_u^2}, \qquad (3)$$

where the first term represents deposition, the second one the diffusion of monomers and the third one the nucleation. The boundary conditions are

$$n_1(0,\theta) = l_a\frac{\partial n_1(0,\theta)}{\partial x} \quad \text{and} \quad n_1(y,\theta) = -l_a\frac{\partial n_1(y,\theta)}{\partial x}. \qquad (4)$$

Note that, for $l_a = 0$ and $l_a \to \infty$, there are absorbing and reflecting boundaries, respectively.

The average local monomer density in all the gaps, $\bar{n}_1$, is related with N_1 according to $N_1 = \gamma\bar{n}_1$. Then, multiplying Eq. (1) by γ and subtracting Eq. (3), it is possible to arrive to

$$\frac{\partial^2 n_1(x,\theta)}{\partial x^2} - \xi_u^2\left(n_1(x,\theta) - \frac{\xi_u^2}{\xi^2}N_1\right) \approx 0 \qquad (5)$$

where the approximation $dN_1/d\theta - dn_1/d\theta \approx 0$ has been used. As usual, the capture kernel of an island with size s and gap size y, $\sigma_s(y)$, can be calculated equating the expression for the rate of capture of monomers by an island of size s, $D\sigma_s N_1$, to the microscopic rate of capture $2\,D\,[dn_1/dx]_{x=0}$

$$\sigma_s(y) = \frac{2\alpha^2\xi_u^{-1}\sinh(\tilde{y}/2)}{\gamma\cosh(\tilde{y}/2) + l_a\gamma\xi_u^{-1}\sinh(\tilde{y}/2)} \qquad (6)$$

which implies that

$$\xi^2 = \xi_u^2\left(1 - \frac{2\xi_u N}{\gamma}\,\frac{\tanh\left(\frac{1}{2\xi_u N}\right)}{1 + l_a\xi_u^{-1}\tanh\left(\frac{1}{2\xi_u N}\right)}\right) \qquad (7)$$

A similar procedure can be done to find ξ_u and σ_u.

Let $p_s^{(0)}(y;\theta)$ be the gap length distribution of islands with size s at a coverage θ. The time evolution of $p_s^{(0)}(y;\theta)$ is given by

$$\frac{dp_{i+1}^{(0)}(y;\theta)}{d\theta} = \frac{dN}{d\theta}\delta\left(y - \langle y\rangle\right) - \Re N_1\sigma_{i+1}(y)p_i^{(0)}(y;\theta) \qquad (8)$$

and

$$\frac{dp_s^{(0)}(y;\theta)}{d\theta} = \Re N_1\left(\sigma_{s-1}(y)p_{s-1}^{(0)}(y;\theta) - \sigma_s(y)p_s^{(0)}(y;\theta)\right). \qquad (9)$$

The solution can be written as

$$p_s^{(0)}(y;X_y) = X_y^{s-(i+1)}e^{-X_y}/\left(y^2(s-(i+1))!\right), \qquad (10)$$

where $X_y = \int_{\theta_y}^{\theta}\Re N_1(\theta')\sigma(y;\theta')d\theta'$. Finally, the capture kernels can be approximated by $\sigma_s(y) \approx \sigma_s(y')$ where $y_s' = \gamma y_s^*/\sum_s y_s^* N_s$ and $y_s^* = \sum_y y p_s^{(0)}(y)/\sum_y p_s^{(0)}(y)$. It worth to note that for extended islands the size of the islands have to be taken explicitly into account by $\sigma_s \approx \sigma(y_s' - (s-\bar{s})/2)$. Henceforth, this analytical procedure will be called self-consistent (SC) approach.

4. Results

The time (coverage) evolution of N_1 and $N = \sum_{s>i}N_s$ is shown in Fig 2. For $l_a = 0$ the system is in the DLA regime while for $l_a = 250$ is in the ALA regime.

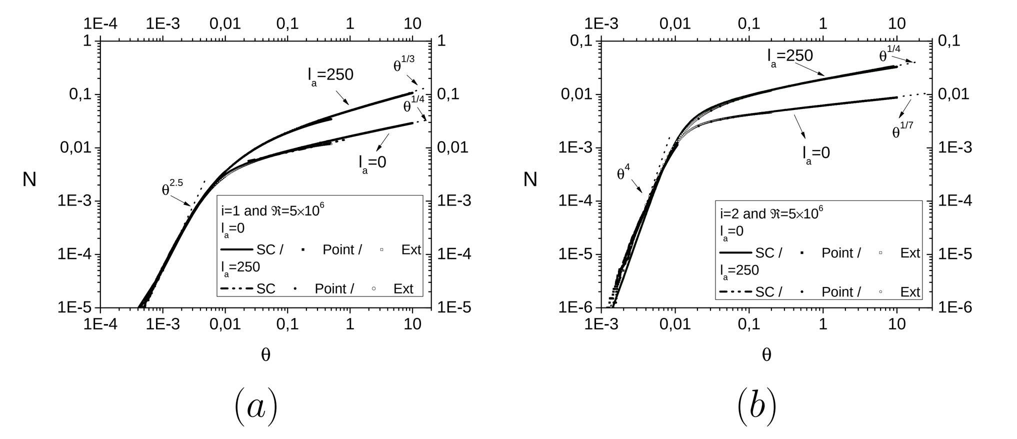

Figure 2: *Coverage evolution of the island density N for two different attachment barriers: $l_a = 0$ and $l_a = 250$. The cases $i = 1$ and $i = 2$ are shown in left and right panels respectively. Filled symbols corresponds to point-islands and open symbols to extended-islands.*

The behavior of N_s for DLA and ALA regimes are shown below.

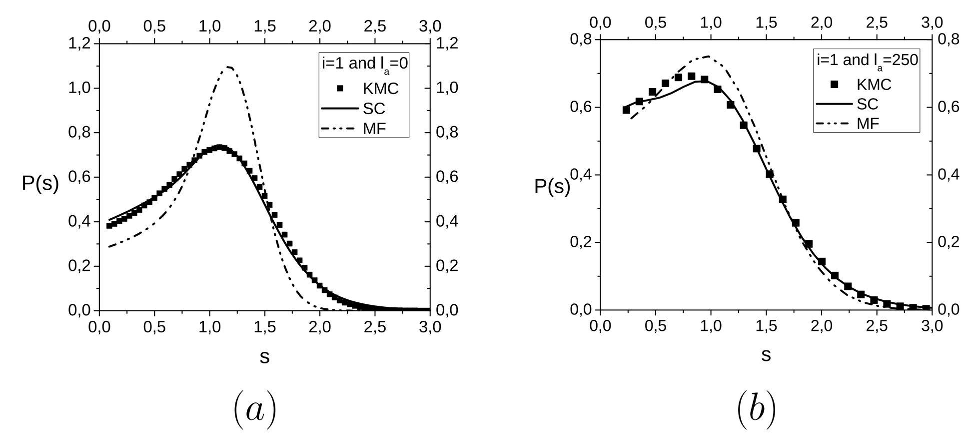

Figure 3: *Island size distribution for $i = 1$ with two different attachment barriers, $l_a = 0$ and $l_a = 250$ for point-islands. The parameters used are $\theta = 0.25$ and $\Re = 5\times10^6$. Symbols correspond to kMC simulations while continuous lines to the SC approach. Dotted lines corresponds to MF (In the MF approach $\sigma_s \approx \bar{\sigma}$).*

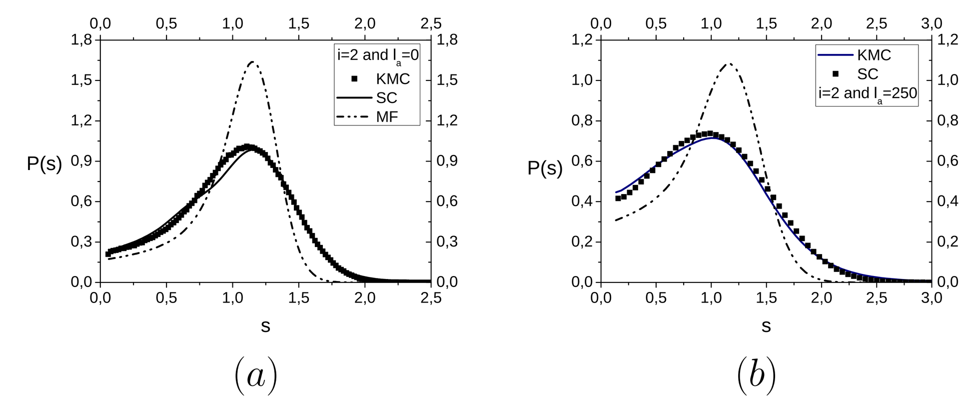

Figure 4: *Island size distribution for $i = 2$ with two different attachment barriers, $l_a = 0$ and $l_a = 250$ for point-islands. The parameters used are $\theta = 0.25$ and $\Re = 5\times10^6$. Symbols have the same meaning as in Fig. 3.*

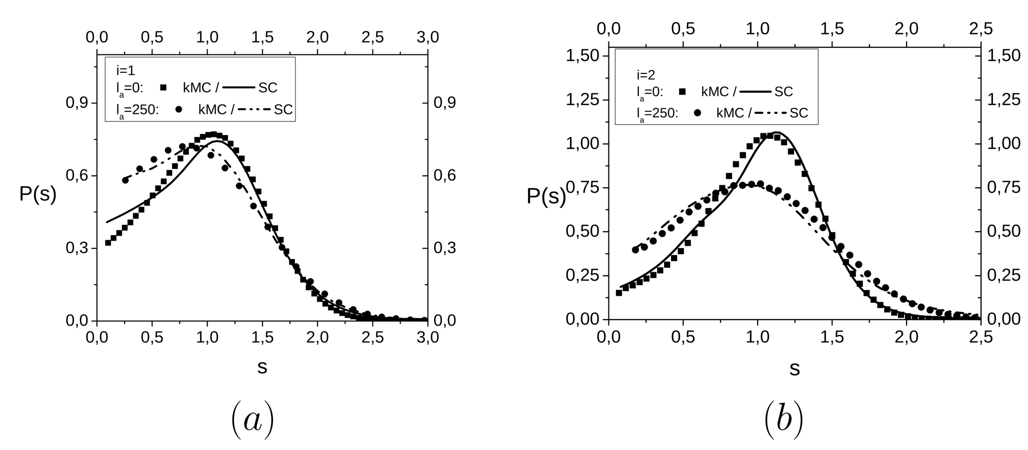

Figure 5: *Island size distribution for $i = 1$ and $i = 2$ with two different attachment barriers, $l_a = 0$ and $l_a = 250$ for extended-islands. The parameters used are $\theta = 0.25$ and $\Re = 5\times10^6$. Symbols have the same meaning as in Fig. 3.*

5. Conclusions

The additional attachment barrier changes the functional form of $P(s)$. Since $P(s)$ is usually measurable experimentally, we can use it to actually calculate microscopic parameters of the model such ϵ_a. Even in the case of strong barriers the island size dependence of the capture kernels have to be taken into account in order to describe $P(s)$.

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