

SELF-CONSISTENT RATE-EQUATION APPROACH TO NUCLEATION AND GROWTH IN POINT/EXTENDED ISLAND MODELS OF 1-D HOMOEPITAXY

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ABSTRACT

A self-consistent rate equation (RE) approach to submonolayer growth on a one-dimensional surface is presented. This approach explicitly takes into account the existence of gaps between clusters and can successfully predict the coverage dependence of the average densities of monomers N_1 , and clusters, N . It also implies an unusual dependence for the monomer-monomer capture number σ_1 as a function of monomer density. To obtain the island size-distribution, a second set of mean-field equations is used describing the evolution of the size-dependent capture zones and leading to explicit size- and coverage-dependent capture numbers. The solution of this fully self-consistent RE approach is then compared with kinetic Monte Carlo results.

INTRODUCTION

Molecular beam epitaxy (MBE) offers the possibility of atomic-scale controlled production of thin films, high quality crystals, and nanostructures. Recently, new experimental methods, such as scanning tunneling microscopy (STM), and reflection high energy electron diffraction (RHEED) have made possible the real-time probing of microscopic details of the surface during the early stages of thin-film growth [1]. This has led to a renewed experimental interest in submonolayer growth [2] - [12]. It has also stimulated considerable theoretical work that has led to a better understanding of the scaling properties of the island density and island-size distribution in epitaxial growth [13]-[26].

Although homoepitaxial submonolayer growth is usually two-dimensional, it is well known that during heterogenous deposition on a cleaved surface, nucleation occurs preferentially on steps rather than on the free surface, leading to one-dimensional growth. For example, the spatial distribution of gold atom clusters on steps of atomic size on UHV cleaved sodium chloride crystal substrates has been measured by Gates and Robins [27]. They have shown that this distribution can be scaled into a time-invariant form and have derived a theoretical expression [28], [29] in good agreement with experimental results. The decoration of steps by clusters has also been used to study the behavior of surface steps, and recently it has been suggested that this offers the possibility of controlled growth of nanoclusters on cleavage steps [17]. Recently it has also been argued that for systems with an extremely high anisotropy, such as the 2×1 -row-reconstructed Si(001) surface, it is also possible to have a quasi one-dimensional MBE [30].

Here we present a self-consistent rate-equation (RE) approach to one-dimensional submonolayer growth that explicitly takes fluctuations into account in order to accurately predict average quantities such as the average monomer density and island density as functions of coverage. To obtain the island size-distribution, a second set of mean-field equations is used to describe the evolution of size-dependent capture zones, leading to explicit size- and coverage-dependent capture numbers. A numerical solution of the resulting rate-equations leads to predictions for the island-size distribution which agree well with simulations in contrast to the standard RE approach.

KINETIC MONTE CARLO (KMC) SIMULATIONS

We have studied two models for irreversible submonolayer growth in one-dimension - a point-island model and a more realistic extended-island model. In both models atoms are deposited

randomly on an initially empty line of M sites at a rate F and may diffuse to nearest-neighbor sites with hopping rate D_h . In the point-island model an island occupies a single site. When a monomer moves onto a site occupied by another monomer, an island is nucleated. Similarly, a monomer moving onto a site occupied by an island is absorbed and the island size increases by one. This corresponds to the irreversible growth of “3D” vertical islands along a step. In the extended-island model the islands are allowed to grow laterally along the step-edge by irreversible attachment of atoms at nearest-neighbor sites. Atoms with one or more nearest-neighbors are assumed to be immobile while monomers landing on top of an island diffuse on the surface until they fall off an edge and are incorporated into the island. Monomers landing on another monomer are immediately incorporated into the first layer and nucleate a dimer.

Similar models have been previously studied [16], [17] using a somewhat different rate-equation approach which takes into account island capture zones. In this approach the scaled gap-distribution (corresponding to the distribution of gaps between islands) was measured via simulations at late-times and then used as an input to the rate-equations (see below). In contrast we have developed a self-consistent rate-equation formulation which does not assume scaling and in which the gap-distribution is directly calculated from the rate-equations.

SELF-CONSISTENT RATE-EQUATION ANALYSIS

The rate equation approach [13], [31] involves a set of deterministic coupled reaction-diffusion equations describing the time (coverage) dependence of average quantities. The RE variables are the average densities of monomers, N_1 , and of islands of size $s \geq 2$, N_s , where s is the number of atoms in the island. A general form of these equations, valid for both point and extended island models may be written,

$$\frac{dN_1}{d\theta} = \gamma - 2N_1 - 2R\sigma_1 N_1^2 - RN_1 \sum_{s \geq 2} \sigma_s N_s \quad (1)$$

$$\frac{dN_s}{d\theta} = RN_1 (\sigma_{s-1} N_{s-1} - \sigma_s N_s) + k_{s-1} N_{s-1} - k_s N_s \quad \text{for } s \geq 2. \quad (2)$$

In these equations, the terms with σ_s correspond to the capture of monomers by other monomers or by existing islands, while the terms with k_s (where $k_s = s$ for extended islands and $k_s = 1$ for point islands) correspond to the deposition of adatoms directly on islands of size s . The first two terms on the RHS of (1) correspond to the deposition flux minus the direct impingement, with $\gamma = 1 - \theta + N_1$ for extended islands and $\gamma = 1 - N$ for point islands. Here $R = D_h/2F$ since the diffusion constant is half of the hopping rate.

Once σ_s is known, equations (1) and (2) can be numerically solved to find the coverage dependence of densities N_s . However, if we write $N = \sum_{s \geq 2} N_s$ and identify $2\sigma_1 N_1 = 1/\xi_1^2$ and $1/\xi^2 = 1/\xi_1^2 + \sum_{s \geq 2} \sigma_s N_s$, we may obtain a pair of simpler equations for the island-density N and monomer density N_1 ,

$$\frac{dN_1}{d\theta} = \gamma - 2N_1 - RN_1/\xi^2 \quad (3)$$

$$\frac{dN}{d\theta} = RN_1/2\xi_1^2 + N_1. \quad (4)$$

These equations can be numerically integrated once the “nucleation length” ξ_1 and “monomer capture length” ξ are known.

Self-Consistent Calculation of Capture Numbers σ_s

We have developed a self-consistent approach to the calculation of the capture numbers σ_s which extends the mean-field approach of Bales and Chrzan [25] in two-dimensions to take into

account correlations between the size of an island and its local environment. We consider the following diffusion equation for the monomer density $n_1(x, \theta)$ in a gap of length y_s near an island of size s (the length of a gap is defined as the number of sites between the two clusters bounding the gap),

$$\frac{\partial n_1}{\partial \theta} = 1 - 2n_1 + R\nabla^2 n_1 - Rn_1/\xi_1^2 \quad (5)$$

with boundary conditions appropriate for irreversible growth $n_1(0) = 0$, $n_1(y_s) = 0$. Here the first two terms on the RHS correspond to deposition while the last two terms correspond to diffusion and nucleation respectively. Since γ represents the fraction of sites that are not occupied by islands, the proper renormalization of the monomer density is $\gamma\bar{n}_1 = N_1$, where \bar{n}_1 is the gap average of $n_1(x)$. Multiplying (5) by γ and subtracting (3) one obtains:

$$\nabla^2 n_1 - \xi_1^{-2} (n_1 - \alpha^2 N_1) = 0, \quad (6)$$

where $\alpha^2 = \xi_1^2/(\xi^2\gamma)$. Solving for $n_1(x)$, then calculating the flux $2D[dn_1/dx]_{x=0}$ of atoms near the island and comparing it with the corresponding RE term $D\sigma_s N_1$ leads to an expression for σ_s ,

$$\sigma_s = \frac{2\xi_1}{\gamma\xi^2} \tanh(y_s/(2\xi_1)) \quad (7)$$

where y_s is the gap-length for an island of size s .

Mean-Field Solution for ξ_1 and Fluctuations in One-Dimension

In order to calculate the capture number σ_s the gap-size distribution $P(y_s)$ and nucleation length ξ_1 must both be known. To calculate ξ_1 we have used a mean-field approach. In this approach we consider the overall rate of capture of monomers by both other monomers and islands in terms of an overall ‘‘screening length’’ ξ . The resulting equation for $n_1(x)$ is the same as Eq. (6), but with ξ_1 replaced by ξ . Assuming that at late-time the rate of monomer deposition is balanced by the rates of monomer capture and island-nucleation one obtains,

$$\xi_1 = \frac{1}{2} \left(\frac{R\gamma}{N_1} \right)^{1/4} \quad (8)$$

It turns out that this ‘‘late-time’’ mean-field expression for ξ_1 works extremely well in predicting the rate of nucleation of dimers from monomers at *all* times, including very early times. It also implies the unusual result for the monomer capture cross section,

$$\sigma_1 = \left(\frac{4}{RN_1\gamma} \right)^{1/2} \quad (9)$$

which implies that due to the existence of fluctuations in one-dimension σ_1 depends on the monomer density. In the low-coverage limit for which $N_1 \simeq \theta$, it also implies that,

$$N \simeq (4/5) R^{1/2} \theta^{5/2} \quad (10)$$

In the absence of fluctuations, the simplified rate-equations (3) and (4) may now be integrated, using Eq. (8) and the relation $1/\xi^2 = 1/\xi_1^2 + \sum_{s \geq 2} \sigma_s N_s$, if one assumes $y_s = \langle y \rangle = \gamma/N$ in Eq. (7) to obtain σ_s . However, due to fluctuations in one-dimension, we must average over the distribution of gap sizes y (we neglect for the moment any dependency on the cluster size s) in order to self-consistently find the ‘‘screening length’’ ξ . In particular, we may write,

$$1/\xi^2 = 1/\xi_1^2 + (2N\xi_1/\gamma\xi^2) \langle \tanh(y/2\xi_1) \rangle_{P(y)} \quad (11)$$

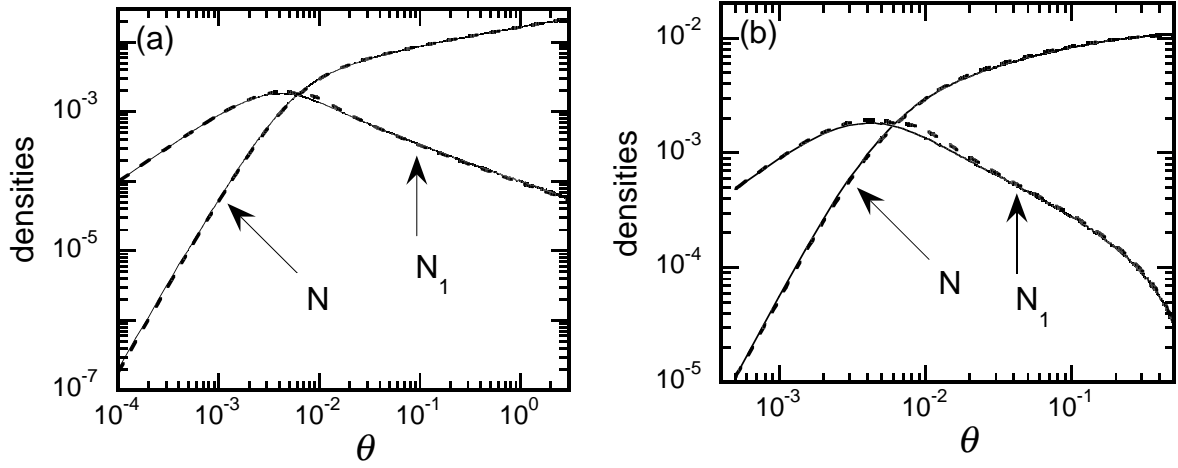


Fig. 1. KMC (solid lines) and RE (dotted lines) results for the island density N and for the monomer density N_1 as a function of coverage for $D_h/F = 10^7$: (a) point-island; (b) extended-island.

To proceed we need to know the gap distribution $P(y)$. However, at late times we can expand $\tanh(z) \simeq z - z^3/3$ and solve for ξ to obtain,

$$\xi^{-2} \rightarrow 12N^2 / (\gamma^2 \langle Y^3 \rangle) \quad (12)$$

where $Y = y / \langle y \rangle$ and $\langle Y^3 \rangle \simeq 1.6$ from KMC simulations. This implies that at late-time, the mean-field prediction for $\sum_{s \geq 2} \sigma_s N_s = \sigma N = 1/\xi^2 - 1/\xi_1^2$ must be corrected by a factor of $\langle Y^3 \rangle \simeq 1.6$ to obtain the correct result. Using this correction and Eq. (8) for ξ_1 , we have integrated Eqs. (3), (4). As can be seen in Fig. 1, we find excellent agreement with simulation results for both point- and extended island models, over the whole range of coverages.

Evolution Equations for the Gap Distributions

To obtain the island size distribution, one needs to go beyond mean-field theory and consider the dependence of the gap-size distribution on the cluster size s . We begin by defining $G_s(\theta, y)$ as the number of gaps of size y near clusters of size s at coverage θ . Taking into account the generation of new gaps by nucleation and the aggregation of clusters, one can write a general set of evolution equations for the set of functions G_s in the following form,

$$\frac{dG_2}{d\theta} = -R N_1 \sigma(y) G_2 + \frac{R N_1}{\xi_1^2} \delta(y, \beta \frac{\gamma}{N}) \quad (13)$$

$$\frac{dG_s}{d\theta} = R N_1 \sigma(y) [G_{s-1} - G_s] \quad (14)$$

where $G_s \equiv G_s(\theta; y)$, $\sigma(y) = \frac{2}{\xi \gamma} f_y$, and

$$f_y = \frac{\xi_1}{\xi} \tanh \frac{y}{2\xi_1}. \quad (15)$$

In these equations we have assumed that at coverage θ the nucleation events produce, on average, gaps of length $\bar{y} = \beta \gamma / N$, where γ / N is the average gap size, and β is a constant factor which takes into account fluctuations in the gaps between islands (determined from the KMC simulations to be 0.75). We note that the break-up of larger gaps due to nucleation is neglected in Eq. (14). If one considers the equations above for a given y , then $\delta(y, \frac{\gamma}{N}) \rightarrow \delta(\theta, \theta_y)$, where

θ_y is the solution of the equation $y = \beta \frac{\gamma}{N(\theta_y)}$. For $\theta < \theta_y$ one has $G_2 \equiv 0$, $G_s \equiv 0$ for all s , while for $\theta \geq \theta_y$ one has $G_2 > 0$ (and some of $G_s > 0$).

To solve Eqs. (13),(14), we note that the factors f_y are slowly varying with θ , and assume that $f_y \simeq \text{const}$. A change of variable from θ to $x = \int_{\theta_0}^{\theta} d\phi \frac{2}{\xi} RN_1$, leads to:

$$\frac{dG_2}{dx} = -f_y G_2 + B_y \delta(x, x_y) \quad (16)$$

$$\frac{dG_s}{dx} = f_y [G_{s-1} - G_s] \quad (17)$$

where $x_y = x(\theta_y)$, and $B_y = (\frac{RN_1}{\xi^2})_{x=x_y}$. Eq. (16) has the exact solution:

$$G_2(x; y) = B_y \exp[-f_y(x - x_y)] H(x - x_y) \quad (18)$$

where $H(z)$ is the step function ($H(z) = 1, z \geq 0; H(z) = 0, z < 0$). In the limit of large x and s , it has been shown by Zinsmeister [32] that the system given by Eq. (17) has the approximate solution,

$$G_s(x; y) = G_2[x - (s - 2)/f_y; y] \quad (19)$$

For a given x and a given s , the average $f_s = \langle f_y \rangle_y$ can be computed as: $f_s = \frac{\sum_y f_y G_s(x; y)}{\sum_y G_s(x; y)}$.

Keeping in both sums the dominant term for which the exponential is equal to 1 (the term for which $x - x_y - (s - 2)/f_y = 0$) leads to the following equation for f_s :

$$f_s = \frac{\xi_1}{\xi} \tanh \left[\frac{\beta \gamma \left(x - \frac{s-2}{f_s} \right)}{2 \xi_1 N \left(x - \frac{s-2}{f_s} \right)} \right]. \quad (20)$$

Solving this equation, it follows that the size-dependent capture numbers $\sigma_s = \frac{2}{\xi \gamma} f_s$ are determined, and the full rate-equations (1) and (2) can be integrated to find the island size distributions.

Fig. 2 shows rate-equation results for the island size distribution ($R = 10^7$) obtained using the gap-evolution equations. Also included for comparison are ‘‘mean-field’’ rate-equation results in which the gap-size is assumed to be the same for all islands. While there is poor agreement between the ‘‘mean-field’’ RE results and simulations there is relatively good agreement between the rate-equation predictions using the gap-evolution equations and the KMC results. The overestimate in the ‘‘tail’’ of the distribution for large s/S_{av} is most likely due to the neglect of the break-up of larger gaps, and work is in progress to include these terms in Eqs. (13), (14).

CONCLUSIONS

We have developed a self-consistent rate equation approach that explicitly takes into account the existence of gaps between clusters. This approach has been used to derive a new expression for the monomer-monomer capture number σ_1 and to accurately predict the coverage dependence of the average densities of monomers and clusters. To obtain the island size-distribution, we have proposed a second set of mean-field equations describing the evolution of the size-dependent capture zones. This second system was solved in closed form, and the solution was used to derive the size- and coverage-dependent capture numbers. Finally, we have shown that such a fully self-consistent RE approach leads to a significant improvement over mean-field results and in particular to reasonably good agreement with KMC results for the island size distribution. In

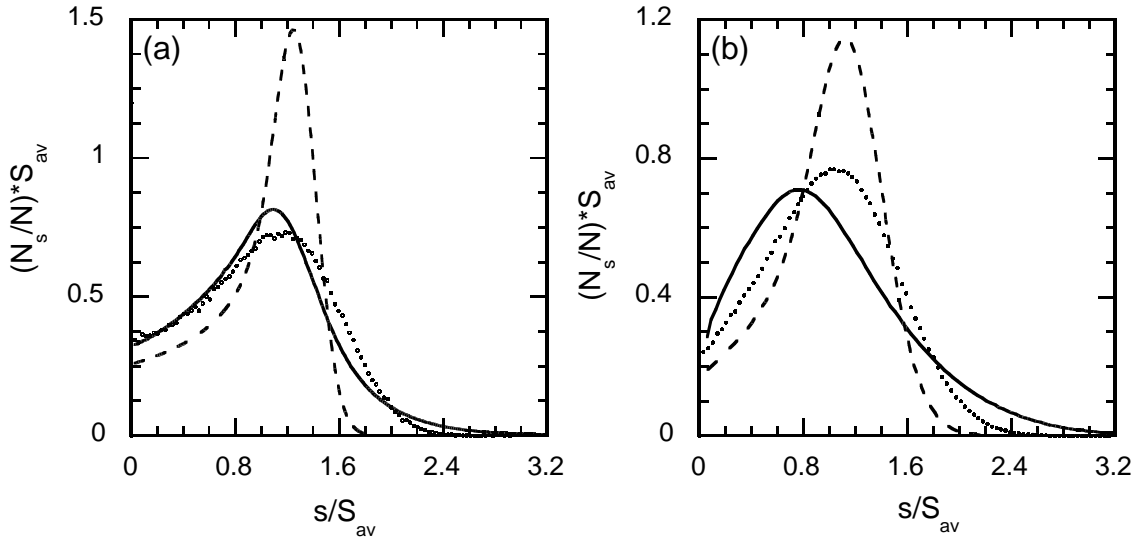


Fig. 2. Island size distributions for: a) point island model at coverage $\theta = 1.0$, b) extended islands model at coverage $\theta = 0.3$, and $D_h/F = 10^7$. For both plots the symbols are KMC results, the solid lines are our RE prediction, and the dotted lines are the MF results.

future work we plan to extend this approach by including the break-up of large gaps, and to develop a similar one for the very important case of 2-d submonolayer growth.

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