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A new method for calculation of island-size distribution in submonolayer epitaxial growth

P.P. Petrov [a,b,*], W. Miller [a], U. Rehse [a], R. Fornari [a]

[a] Leibniz Institute for Crystal Growth, Max-Born-Str. 2, 12489 Berlin, Germany
[b] Department of Mathematics and Informatics, University of Sofia, J. Bourchier Blvd. 5, 1164 Sofia, Bulgaria

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ABSTRACT

In this paper, a novel method for calculation of island-size distribution in one-dimensional submonolayer epitaxial growth based on difference-differential rate equations is introduced. Moreover, precise analytic expressions for the first several Taylor coefficients of the distribution (with respect to the ratio of diffusion and deposition rates) are obtained and the corresponding error term is estimated.

1. Introduction

In recent years, the problem of characterization of island-size distribution in submonolayer epitaxial growth has become one of the central problems in theoretical physics. On one hand, its deep significance is determined by the practical reason that submonolayer structures can strongly influence the morphology of the growing multilayered film. In particular, the one-dimensional (1D) lattice gas model can successfully represent nucleation and growth at step edges [1–3] as well as step bunching in the step-flow regime (where the monomer is the step itself) (see e.g. [4]). On the other hand, the beauty of this essentially mathematical problem itself has challenged the attention of mathematicians and theoretical physicists. Traditionally, the efforts to obtain a (numerical) solution to it, are based on two approaches. The first one goes back to the paper of von Smoluchovski [5] and relies on studying the so called rate equations [6,7]. The second one is based on kinetic Monte Carlo simulations [8–10]. Usually the authors combine both methods and compare the results of simulations with rate equation analyses and experiments [11–17]. Heuristic predictions have been made for the main quantities of interest which, in most of the cases, are island- and capture zone-size distributions and their dynamic scaling properties [3,18]. An excellent introductory review on the topic can be found in [19].

In the present paper, we introduce a new approach for calculation of island-size distribution. It is based directly on *exact difference-differential* rate equations and gives the possibility of calculating the distribution with high precision. For clarity of the presentation, we describe our method for the case of 1D nucleation and *critical island size 1*. Let us remind, that the critical island size is one less than the size of the smallest stable island. It should be noted that this method is closer to reality than other methods considering the so called *point islands* or differential rate equations with respect to a continuous island-size variable. We also believe that the suggested approach can be successfully generalized for calculating of island-size distribution on a two-dimensional substrate.

* Corresponding author at: Leibniz Institute for Crystal Growth, Max-Born-Str. 2, 12489 Berlin, Germany. Tel.: +49 30 6392 3078; fax: +49 30 6392 3003.
E-mail address: petrov@ikz-berlin.de (P.P. Petrov).

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2. Problem statement and solution

First, we describe the problem and introduce some notations. We consider 1D lattice of sites which are supposed to be unoccupied at time $t = 0$. The time is normalized by the adsorption of one monolayer at the lattice. Thus, atoms are raining (*adsorbed*) upon the sites at a rate 1 per site and diffusing at a rate D until the lattice is completely filled up. At any moment $t \in [0,1]$ every site is occupied or unoccupied. The question we want to answer is: *What is the mean (statistical) number of appearances of certain finite pattern of sites at fixed time t?* More precisely, the statistical distribution of the so called *islands* or *chains* is very important for the applications, i.e. patterns of type $\circ\bullet\cdots\bullet\circ$, where $\circ$ and $\bullet$ denote unoccupied and occupied site, respectively. Similar problem was studied in [20] by means of computer simulations in the post-deposition regime and higher critical island size.

Let us introduce the following notations for some patterns, which we will often use throughout the paper.

$P_{b_1 b_2 \cdots b_k} := P_{b_1 b_2 \cdots b_k}(D;t)$ will denote the number of appearances (per site) of the pattern $b_1 b_2 \cdots b_k$ at time t, $b_i \in \{\circ,\bullet\}$. Note that $P_{b_1 b_2 \cdots b_k}(D;t)$ coincide with the probability for pattern $b_1 b_2 \cdots b_k$ at some fixed k consecutive sites. Let

$$I_k(D;t) := P_{\circ\underbrace{\bullet\cdots\bullet}_{k}\circ}(D;t)$$

be the probability for island of size k and introduce also the notations

$$H_k(D;t) := P_{\underbrace{\bullet\cdots\bullet}_{k}}(D;t)$$

and

$$Y_k(D;t) := P_{\circ\circ\underbrace{\bullet\cdots\bullet}_{k}}(D;t).$$

We will often use also the following simple property of the probabilities P:

$$P_{\circ b_2 \cdots b_k} + P_{\bullet b_2 \cdots b_k} = P_{b_2 \cdots b_k}. \tag{1}$$

In the case under consideration – that of critical island size 1 – an atom is not allowed to move if it possesses an occupied neighbor. Our aim is to find an approximate expression for the island-size probability I_k. To this aim we suppose that $I_k(D;t)$ is an analytic function of the diffusion rate D and therefore, possesses a Taylor representation:

$$I_k(D;t) = t^k(1-t)^2 \sum_{n=0}^{\infty} \varphi_{n,k}(t)\frac{D^n}{n!}. \tag{2}$$

In this way, the problem of calculation of island-size distribution is reduced to the problem of finding analytic expressions for the Taylor coefficients $\varphi_n := \varphi_{n,k}$ in Eq. (2). Let us note first, that we can immediately write down the coefficient φ_0 since it corresponds to the simplest case of critical island size 0 ($D = 0$). Since the probability for a fixed site to be occupied (unoccupied) at a time t is equal to t (resp. $1-t$), we have

$$I_k(0;t) = t^k(1-t)^2, \tag{3}$$

i.e. $\varphi_0(t) = 1$.

Throughout the paper, the notation P' for a probability function $P(D;t)$ will be reserved for the partial derivative with respect to t, while the partial derivative with respect to D will be denoted as usual by $\frac{\partial}{\partial D}P(D;t)$. In order to simplify the calculations (and the form of Taylor coefficients) we will consider the processes of adsorption and diffusion on the 1D lattice *backwards*. In other words, we will suppose that initially all the sites are occupied and the atoms begin to desorb at rate 1 per site and diffuse at rate D. The new time parameter $u = 1 - t$ will be ranging again from 0 to 1 and attachments of atoms to an island, i.e. movings of the form

$$\bullet\circ\bullet\cdots\bullet \mapsto \circ\bullet\bullet\cdots\bullet$$

will not be allowed. For the sake of simplicity, we will use the notation $P(D;u)$ for $P(D;1-u)$, where P is any probability function.

In order to find the coefficients $\varphi_n(t)$ we will write first a differential equation for the probability H_k of k-tuples of consecutive occupied sites. The following equation holds true:

$$H_k'(D;u) = -\frac{k}{1-u}H_k - DY_k. \tag{4}$$

Indeed, since the pattern $\underbrace{\bullet\cdots\bullet}_{k}$ changes only with desorbing of some of its atoms or with moving of the leftmost, resp. rightmost atom in the patterns $\circ\circ\underbrace{\bullet\cdots\bullet}_{k}$ and $\underbrace{\bullet\cdots\bullet}_{k}\circ\circ$, then the rate of its variation is given by Eq. (4).

By making use of $H_k(D;0) = 1$ and $H_k(D;1) = 0$, it follows from Eq. (4) that