Anomalous Dimension and Spatial Correlations in a Point-Island Model

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Abstract

We examine the island size distribution function and spatial correlation function of a model for island growth in the submonolayer regime in both 1 and 2 dimensions. In our model the islands do not grow in shape, and a fixed number of adatoms are added, nucleate, and are trapped at islands as they diffuse.

We study the cases of various critical island sizes $i$ for nucleation as a function of initial coverage. We found anomalous scaling of the island size distribution for large $i$. Using scaling, random walk theory, a version of mean-field theory we obtain a closed form for the spatial correlation function. Our analytic results are verified by Monte Carlo simulations.
Recently there has been much interest in island size distribution and morphology in submonolayer epitaxial growth [1–6]. With the development of real-space imaging methods such as scanning tunneling microscopy (STM) and atomic force microscopy (AFM) and surface-sensitive diffraction methods (e.g. low-energy electron diffraction), several experimental systems have been studied under conditions that favor irreversible aggregation and low island mobility and dissolution. Some examples are homoepitaxial systems such as Fe/Fe(100) [7] Ni/Ni(100) [8] Si/Si(100) [9] Cu/Cu(100) [10] and heteroepitaxial systems such as Pb/Cu(001) [11], Au/Ru(0001) [12] and Ag/Si(111) [13].

In this paper we consider a simplified model in order to look at general features of this type of growth. Our model is a point-island model in which we ignore the island shape, and turn off the flux of adatoms. We put all our adatoms down at once and allow them to aggregate until the evolution stops. We ignore all the complications which arise from shapes such as dendritic or diffusion-limited-aggregation (DLA-like) fractal aggregates [12,14]. We focus here on island size and island-island spatial correlation scaling behavior. We find striking results in the limit of low coverage, in particular a change in the scaling or the island-size distribution as we change the critical island size (defined below). We find that for large critical island size, both the island size distribution ans the island-island correlation obey anomalous scaling.

In our model we start by randomly distributing adatoms on a $d$-dimensional cubic lattice of size $L^d$. Each site is occupied with probability $\theta$. In principle the only natural length scale in this problem is $\theta^{-1/d}$, the initial mean distance between adatoms. Starting at time $t = 0$, adatoms are picked randomly and diffuse by nearest-neighbor hopping. A stable island at site $x$ can be formed if $i$ adatoms meet simultaneously at $x$, and any adatom stepping onto an existing stable island site will be absorbed and becomes immobile. The size $s$ of an island is defined as the number of adatoms it has captured. An island with size smaller than the critical size $i$ can dissociate without any energy barrier, while a stable island cannot dissociate and is immobile. The critical size $i$ defined in this paper differs by one from the critical size usually defined in other literature. We have no incident flux of adatoms
in our model; at the end of the process only stable islands will survive. One motivation for studying this model comes from an experiment \[17\] recently performed where a 50-layer GaAs surface is quenched from a high temperature (600°C) with the flux of adatoms shut off. However, our primary interest is in the model as an example of statistical physics far from equilibrium.

The first important quantity in the description of the asymptotic behavior of this model is the island-size distribution function $N_s$. This is the density of islands of size $s$. Assuming that there exists only one characteristic size in the problem which is the average island size $S$, one may guess a form for this function \[15\]: $N_s(\theta) = Af(s/S)$, where $f(u)$ is a scaling function which also depends on the critical island size $i$. Since there is no flux of atoms in this model, the number of atoms is conserved, hence $\theta = \sum_{s \geq 1} sN_s$. Approximating the sum by an integral, $\theta = AS^2 \int_0^\infty f(u)udu$, which implies $A \sim \theta/S^2$. Therefore one may write:

$$N_s(\theta) \sim \theta S^{-2}f(s/S)$$  \hspace{1cm} (1)

where the scaling function $f(u)$ satisfies $\int_0^\infty f(u)udu = 1$. The total island density is given by $N = \sum_{s \geq 2} N_s$ and the average island size is $S = \sum_{s \geq 1} sN_s / \sum_{s \geq 1} N_s = \theta/N$ where we have used the fact that $N_1 = 0$ at $t = \infty$.

We now assume that the average island size $S$ scales as

$$S \sim \theta^{-z}. \hspace{1cm} (2)$$

Equation (2) may then be rewritten in the form

$$N_s(\theta) = \theta^{1+2z}f(s\theta^z) \hspace{1cm} (3)$$

Hence the total island density has scaling form

$$N \sim \theta^{1+z} \hspace{1cm} (4)$$

As we will see, a naive analysis of the process gives $z = 0$. If $z \neq 0$ we say that we have anomalous scaling.
The second quantity we are interested in is the island-island correlation function $G(r)$, which is the probability of finding two islands separated by a distance $r$. Note that $r \leq 1$ is inaccessible in the model and we define $G(0) = 0$ here. For large distance $r$ the correlation function should approach the total island density $N^2$ since there is no long range order in the system.

The simulation results for $d = 1$ dimension are summarized as follows. The simulation involved 1000 runs on lattices of 1000 sites, with periodic boundary conditions. We use critical size $i$ ranging from 2 to 4, and initial coverage $\theta$ ranging from 0.02 to 0.2. The results for $N_s(\theta)$ are shown in Fig 1. A data collapse shows that $z = 0$ for $i = 2$ and, $z = 0.03$ for $i = 3$, but $z = 0.2$ for $i = 4$. We will argue below that this corresponds to anomalous scaling for $i \geq 4$. For $i = 2$, $N_s$ can be compared to an exact solution of a mean field rate equation: $N_s = e^{-1}\theta(s - 1)/s!$ which fits the simulation curve. We can see that the formation of islands mostly comes from the nucleation events: two adatoms meet each other and become an island. The aggregation events are rare. On the other hand, for large $i$, nucleation events are less probable than aggregation events.

The $z = 0$ result can be understood by naive scaling theory, because there is only one characteristic length scale, and $N_s$ and $\theta$ should have the same dimension, $L^{-1}$, where $L$ is the system size. Suppose we rescale the lattice constant: $a \rightarrow \lambda a$ ($\lambda > 1$) while keeping the number and distribution of the initial adatoms fixed. This operation will reduce the initial coverage of adatoms to half its original value. Now, if we let the adatoms still perform the unit length hopping then it would be reasonable to assume that the final number of islands with size $s$ will be the same as before. Thus $N_s$ is half of the previous value, that is $N_s \sim \theta$ or $z = 0$.

The fact that $z \neq 0$ for $i \geq 4$ implies that the lattice constant plays a relevant role in this case, since it is the only other length in the problem. The exponent $z$ is analogous to an anomalous dimension in statistical physics. If we include the fact that $\theta$ has the dimension of $L^{-d}$ then the proper scaling theory suggests that
\[ N_s(\theta) = a^{-2dz\theta^1+2z} f(s\theta^za^{dz}) \]  

where \( a \) is the lattice constant which has the dimension of length and \( d \) is the dimension of the substrate. Similarly, the average island size \( S \) and total island density \( N \) have the scaling form

\[ S \sim a^{dz}\theta^{-z} \]  

and

\[ N \sim a^{dz}\theta^1+z \]

One interesting property of this model is seen when \( \theta \to 0 \) for fixed \( a \), namely \( S \to \infty \) for \( z > 0 \). It seems that there is only one giant island finally for an initially sparsely distributed system. This happens when \( i \geq 4 \) in \( d = 1 \).

We can attempt a qualitative explanation of our results. There are three fundamental physical processes in our model: diffusion, nucleation and aggregation. The final island distribution is the result of local (esp. in \( d = 1 \) dimension) competition between nucleation and aggregation. It is well known that in \( d = 1 \) the probability for a particle to eventually return to its random walk origin is always 1, but the reunion of \( i \) particles is not always certain. In fact, Fisher \[19\] showed that, in \( d = 1 \), this reunion has probability 1 when \( i < 4 \) but smaller than 1 when \( i \geq 4 \). We believe that this is reflected in our simulation in that once an island (for \( i \geq 4 \)) is formed it will sweep up many adatoms. Adatoms have small probability to group in 4 before they are absorbed by an existing island.

The simulation for \( d = 2 \) involves 1000 runs on a 100 times 100 lattice. The critical size varies from 2 to 4 and the initial coverage from 0.02 to 0.2. The data for \( N_s(\theta) \) are shown in (Fig 4). A data collapse shows that \( z = 0 \) for \( i = 2 \), \( z = 0.24 \) for \( i = 3 \) and \( z = 0.65 \) for \( i = 4 \). We have been able to extend Fisher’s argument to \( d = 2 \). We find that the probability for 3 walkers to meet is less than 1 in \( d = 2 \). Our simulation indeed shows anomalous scaling for \( i \geq 3 \).
Further, we studied the island-size distribution for $i = 2$ in $d$ up to 4. Interestingly, we found that $z$ is always 0 and the island-size distribution function $N_s(\theta)$ agrees with the rate equation result $N_s = e^{-1}\theta(s - 1)/s!$ [18]. The rate equation result is obtained under the assumption that the “capture number” of an island is independent of its size $s$ [18]. However, recently some authors [21] pointed out that this “capture number” depends on island size $s$ even in a point-island model. They simulated a point-island model with external flux of adatoms and obtained the “capture number” for each class of island by measuring the capture event per unit time for each class of island. They found that the capture number depends on the island size $s$ for large island while for small island this number is a constant. Since we turn off flux of adatoms in our model we do not have ($i = 2$ case) enough adatoms to form very large island. Thus the the constant “capture number” assumption is reasonable in our case.

We have also studied the island-island spatial correlation function. The results are shown in Fig. 2. Starting from 0 at $r = 0$ the island-island correlation function increases to an asymptotic value which is the square of the total island density. The value of $G(r)$ near $r = 0$ implies that close to an island further nucleation is suppressed. Adatoms are bound by the island edge before they meet another adatom. The depletion in the population of nearby pairs of islands is enhanced with increasing $i$. This depletion leads to a ring structure or “splitting” in the diffraction profile of the specular beam [20]. For $i = 2$ we get a functional form of $G(r)$ which in good agreement with the simulation. Our argument for $i = 2$ is as follows.

Since an island is formed by an adatom and its nearest neighbor (essentially by definition for $i = 2$) one can approximate $G(r)$ (for islands) by

$$G(r) \sim \sum_{n=2}^{\infty} p_n(r)$$

(8)

where $p_n(r)$ the probability of initially (at time $t = 0$) finding an $n$-th nearest neighbour adatom at distance $r$ from a given adatom. This probability is given by the Poisson distribution for an randomly distributed system,
\[ p_n(r) \sim (r\theta)^ne^{-r\theta}/n! \] (9)

Therefore we get

\[ G(r) \sim 1 - e^{-r\theta}. \] (10)

Generally we propose a closed form for \( G(r) \)

\[ G(r) \sim 1 - e^{-r\theta} - \ldots - (r\theta)^{i-2}e^{-r\theta}/(i-2)!. \] (11)

From Fig. 3 we can see that this functional form of \( G(r) \) fits the simulation for \( i \) up to 4 in \( d = 1 \).

The results for the island-island spatial correlation function in \( d = 2 \) are shown in Fig. 2. For \( i = 3, 4 \) near \( r \sim 0 \) we only see the linear behavior of \( G(r) \). This doesn’t agree with the mean-field theory we gave above. This can be understood by noting the topological difference between \( d = 1 \) dimension and \( d \geq 2 \) dimension, namely in \( d \geq 2 \) dimension an adatom can go around an island to meet another adatom. This behavior reduces the depletion effect of island in higher dimensions.

In our model we ignore the shape of the island. We expect that for small initial coverage the shape of island will not affect the results we obtained, especially the universal index \( z \) for the island size distribution. However, for large initial coverage we enter the percolation regime where this assumption no longer holds. It will be interesting to study a more realistic model for a large (but still below the percolation critical value) initial coverage.

Another interesting issue is the critical island size \( i \). In our model we assumed that island with size larger than \( i \) are stable and immobile. In experiment the peripheral adatoms of an island always have the chance to break the bond to the island. This will make the definition of critical island size ambiguous. By changing the breaking energy we can effectively tune the critical island size \( i \) continuously. It will be interesting to see when the anomalous behaviour appears.

In sumary, in this paper we studied a new point-island model in both 1 and 2 dimensions for various critical island size and different initial adatom coverage. We investigated
the island size distribution function for various initial coverages, critical island sizes and in different dimensions. We proposed a scaling form for the island size distribution function and found anomalous behaviour for this function when critical island size is large. This can be understood by the many body random walk theory, namely the reunion probability of \( i \) random walkers is smaller than 1 when \( i \) is large enough. A search for experimental manifestation of this effect would be interesting. We also studied island-island spatial correlation function. Using a version of mean field theory we obtain a closed form for the island-island spatial correlation function in 1 dimension. Monte-Carlo simulation agrees with the analytic results.

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FIGURES

FIG. 1. simulation results of \( N_s(\theta) \) and data collapse, \( d = 1 \). We choose system size \( L = 10000 \) and number of runs 1000. Here initial coverage \( \theta \) varies from 0.02 to 0.2. The circle symbol stands for \( \theta = 0.02 \) and square for \( \theta = 0.04 \) etc.

FIG. 2. simulation results of \( G(r) \) , \( d = 1 \) and \( d = 2 \). \( G(r) \) is normalized here so that \( G(\infty) = 1 \). Here initial coverage \( \theta = 0.1 \) and number of runs 1000. The system size \( L \) is 10000 for \( d = 1 \) and 100 for \( d = 2 \)

FIG. 3. fit of \( G(r) \) using the functional form from mean field theory, \( d = 1 \). Here initial coverage \( \theta = 0.1 \) and number of runs 1000. The system size \( L \) is 10000.

FIG. 4. simulation results of \( N_s(\theta) \) and data collapse, \( d = 2 \). We choose system size \( L = 100 \) and number of runs 1000. Here initial coverage \( \theta \) varies from 0.02 to 0.2. The circle symbol stands for \( \theta = 0.02 \) and square for \( \theta = 0.04 \) etc.