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The asymptotic scaling limit of point island models for epitaxial growth

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Abstract

Scaling of the island size distribution in the asymptotic scaling limit $\Theta \to \infty$ will be discussed. In particular, it is shown that for the simplest implementation of a model for epitaxial growth, the so-called point island model, the island size distribution (ISD) does not scale, and in fact becomes singular. The ultimate reason is that in a point island model there is no distinct nucleation phase followed by an aggregation phase. The same argument might be used to explain the existence of scaling in extended island models: Only when the nucleation phase is infinitesimal short compared to the aggregation phase, and there is no significant post-nucleation, do we expect scaling of the ISD.

Keywords: Epitaxy; Scaling; Point islands

Simulation and modeling of submonolayer epitaxy has been an active area of research in the past couple of decades. This has been stimulated by the enormous technological relevance of epitaxial growth of thin films for many devices and applications in optics and microelectronics. Moreover, more realistic simulations have been made possible by the advance of computational facilities together with improved and more efficient algorithms, as well as by the availability of very detailed experimental data. In particular, the atomic resolution of scanning tunneling microscopy (STM) images has been very useful in testing the reliability of many computational results.

A quantity of particular interest is the scaled island size distribution (ISD). It has been shown in experiments [1,2] as well as a large number of computer simulations [3–5] that for irreversible aggregation the ISD scales for different values of the ratio of the diffusion constant D to the deposition flux F, and different coverages Θ according to

$$n_s = \Theta / s_{av}^2 g(s/s_{av}). \tag{1}$$

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In Eq. (1), n_s is the number density of islands of size s, s_{av} is the average island size, and $g(x)$ is a unique scaling function. The shape of $g(x)$ is directly related to the spatial arrangement of islands [6–10], which in turn is determined by the proper spatial fluctuations during the nucleation of islands [11].

Typically, scaling of the ISD is assumed to be scaling with respect to D/F and coverage Θ. An important question that will also be the focus of this paper is the asymptotic scaling limit with respect to these 2 variables. Scaling of the ISD with respect to D/F occurs when D/F is not too small (at least 10^5), and it is expected to become better as D/F increases. Thus, the limit $D/F \to \infty$ (with a fixed value for Θ) is usually referred to as the scaling limit for D/F. The issue is more complicated for scaling with respect to Θ. It is well established that there is no scaling in the nucleation phase. Thus, we only expect scaling with respect to Θ for $\Theta \geqslant \Theta_{crit}$, where Θ_{crit} is the end of the nucleation phase, and can be estimated as $\Theta_{crit} \simeq (D/F)^{-1/2}$. On the other hand, it is also known (from experiments and models with realistic spatial exptent) that scaling breaks down when islands coalesce, which typically starts around $\Theta = 0.2$. Thus, the scaling limit for Θ is only a small regime. Scaling with respect to Θ improves as as Θ increases, since nucleation never stops completely (but becomes less relevant). Thus, the scaling limit for Θ is the ill-defined regime where "Θ is large, but not too large". If one could somehow prevent islands from merging, the true scaling limit for Θ would then also be $\Theta \to \infty$. As described below, this limit can be attained for point islands. Since point islands are also often used to understand basic behavior during epitaxial growth, this is the limit that is the main focus of this paper.

The ISD has proven notoriously difficult to calculate by any means other than simulation. In particular mean field approaches that are typically based on rate equations so far have always shown a narrowing and sharpening of the ISD as D/F and/or Θ increases [12]. In these models in the asymptotic limit that $D/F \to \infty$ the ISD always becomes singular, in contrast to simulations that properly include the spatial fluctuations during the nucleation of islands. Such simulation models are typically based on kinetic Monte Carlo (KMC) simulations.

Two particularly simple types of KMC models have been used in many studies. We will refer to them as point island and extended island models. In a point island model all islands occupy only one lattice site, regardless of the mass that has accumulated within these islands. During the simulation a number is associated with every point island, which counts the number of atoms that have been incorporated into this island, and which we will refer to as the size of the point island. One virtue of and main motivation for such models is that one does not have to worry about shape effects, and that by construction islands cannot coalesce. As a result, it is possible to deposit atoms that amount to many atomic layers and still refer to the sub-monolayer growth regime. Thus, it is possible to study the asymptotic scaling limit $\Theta \to \infty$ within a point island model. In contrast, in extended island models, an island of size s does indeed occupy s lattice sites, and typically coalescence becomes relevant after approx. 20% of a monolayer (ML) have been deposited. There have been many studies where it is claimed that the ISD scales with respect to Θ for either of these models.

In this article we will show that for point island models the ISD does not scale in the asymptotic limit $\Theta \to \infty$ (with D/F fixed), and in fact becomes singular, regardless of the value of D/F. We will give an explanation why there cannot be scaling of the ISD for a point island model in this limit. Our argument then might also explain why there is scaling with Θ and D/F of the ISD for extended island models in the limit that $D/F \to \infty$ when Θ is sufficiently large (where the meaning of large Θ is crucial and will be explained below). True scaling can only be obtained in a limit where all island sizes are directly proportional to the size of their corresponding capture area. This can only be the case when there is no more nucleation, and when the island sizes at the end of the nucleation phase are small compared to the island sizes in the scaling limit. In other words, scaling only occurs when the history of the size evolution of the islands up to the end of the nucleation phase can be neglected.

The first question that needs to be answered when considering a *point island* model is: What do we mean when we call an island a *point island*? Mathematically, a point island should not occupy any physical space. But this immediately leads to the next set of questions: How do islands nucleate, and how does an island (of spatial extent zero) ever capture another adatom? Also, from a practical point of view, it is not clear how to implement an island of size zero in a computer code. We have therefore taken the standard approach where a point island actually has the spatial extent of one lattice site. Within our model, an island is nucleated when two atoms are at the same lattice site. An adatom is captured only when it diffuses to exactly that site. In particular, an adatom that is directly adjacent to an existing island is not automatically incorporated into the island. Note that this implies that islands can be nucleated in adjacent lattice sites. This is slightly different from many previous realizations of point island models, where an adatom is incorporated when it is adjacent to an existing island, and as a result islands are separated by at least one lattice site. However, it will become apparent from the explanations below that the conclusions of this article are not dependent on our choice of the definition of a *point island*.

We will also present data for KMC simulations for a model with spatially extended islands. Within this model, atoms are deposited at a rate F, and they can hop to a nearest neighbor site with a rate D. Once 2 atoms are adjacent to each other they become immobile, and form an island. Atoms that diffuse toward an island boundary attach irreversibly. As one additional process we allow singly coordinated edge atoms to diffuse along an island edge at a rate D_{edge} (for islands larger than size 3), to ensure compact island shapes [13]. All data shown for extended islands was obtained with $D_{\text{edge}}/D = 1/1000$ [13] and represents the average over at least 10 independent runs on lattices as large as 2000×2000. We have tested carefully that there are no system size effects in our data.

Our main result for point islands is shown in Fig. 1, where the scaled ISD is shown for different values of D/F and different values of Θ. All data shown is averaged over (at least) 10 statistically independent simulation on lattices of size 800×800 (or larger). It is clear that the ISD is not independent of Θ, and in fact it becomes singular as $\Theta \to \infty$. The explanation for this lack of

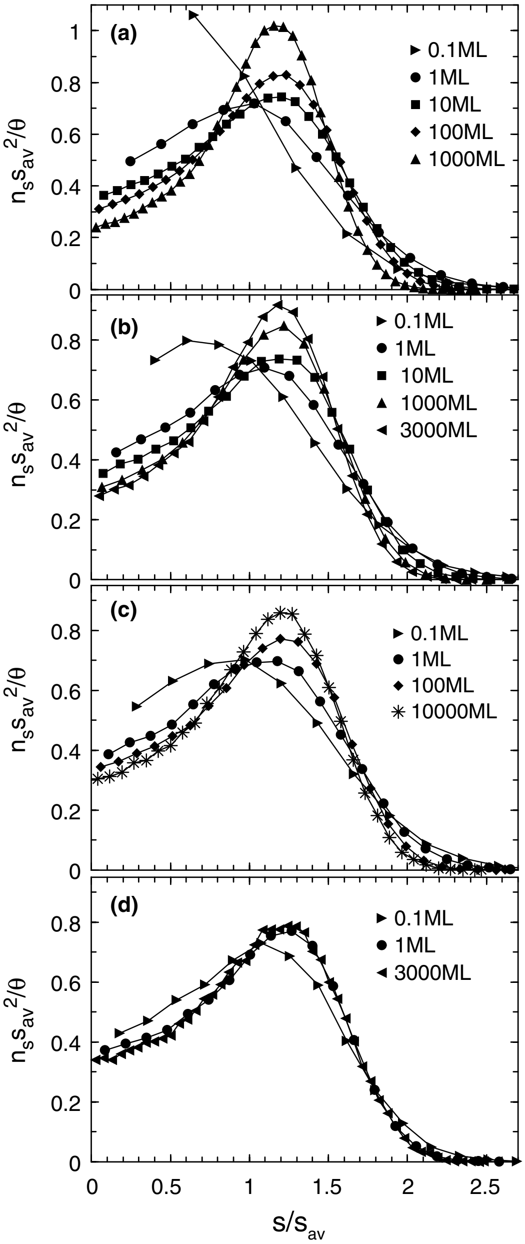

Fig. 1. The scaled ISD for $D/F = 10^4$ (a), $D/F = 10^5$ (b), $D/F = 10^6$ (c), and $D/F = 10^7$ (d) at different coverages Θ. Note the different scale in (a).

scaling is the following: The island density does not saturate in a point island model, but increases with coverage according to $N \sim \Theta^{1/3}$ [14]. As a result, the distribution of capture areas around the islands changes whenever a new island is nucleated. Thus, the correspondence between island size and size of the related capture area is destroyed whenever there is a new nucleation event. But this correspondence (that islands grow exactly according to the fixed size of their related capture area) is ultimately the requirement for the ISD to scale (cf. below).

Regardless of the exact value of D/F, there will be a value for Θ when an island has been nucleated at every lattice site. This can be seen in Fig. 2, where we plot the island density as a function of coverage for different values of D/F. Clearly, the island density increases according to $N \sim \Theta^{1/3}$ until it approaches one, where it bends over and saturates at one island at every site. At this point (which will happen for a finite Θ for any value of D/F) every atom is immediately incorporated because it directly hits an island, and the problem reduces to a Poisson process. In the limit that $\Theta \to \infty$, the Poisson distribution will always dominate the ISD, and the evolution of the ISD up to this point is negligible. However, for a Poisson distribution the width scales according to $\Theta^{1/2}$, so that the width of the scaled Poisson distribution scales according to $\Theta^{-1/2}$. On the other hand, the amplitude of the scaled Poission distribution increases linearly with Θ. Thus, the scaled ISD approaches a δ-function. This is illustrated in Fig. 3, where we show the ISD for different coverages in the extreme limit that $D/F = 0$.

The main conclusion of this article is that the ISD for any value of D/F as shown in Fig. 1 has to become singular in the asymptotic limit and approach the ISD shown in Fig. 3. Of course one might argue that the asymptotic limit $\Theta \to \infty$ is not physical for a point island model. Nevertheless, we believe that it is important to understand the scaling of the point island model in the true asymptotic limit if one wants to understand scaling for other models. One might wish to re-define the asymptotic limit for point islands as the limit where Θ is large, but not too large, such that N is significantly less than 1. It can be seen from Fig. 1 that the sharpening of the ISD is delayed as D/F increases. Thus, there is a regime where the ISD (almost) scales, and this regime extends as D/F increases. But in our view this is not true scaling in the asymptotic limit; in the true asymptotic limit $\Theta \to \infty$ the approach of the ISD to a Poisson distribution is inevitable, and scaling has to break down.

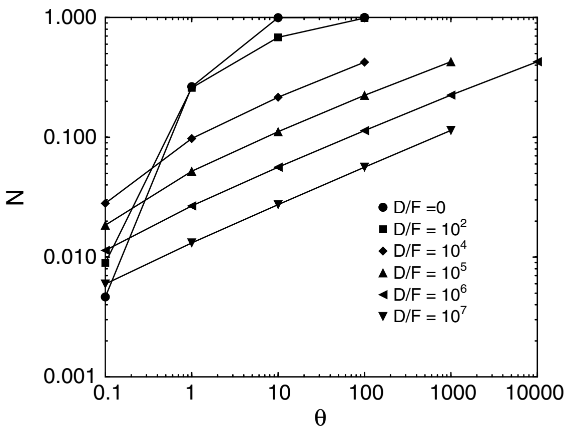

Fig. 2. The total island density N as a function of coverage Θ for different values of D/F. The saturation value of $N = 1.0$ corresponds to one island per lattice site.

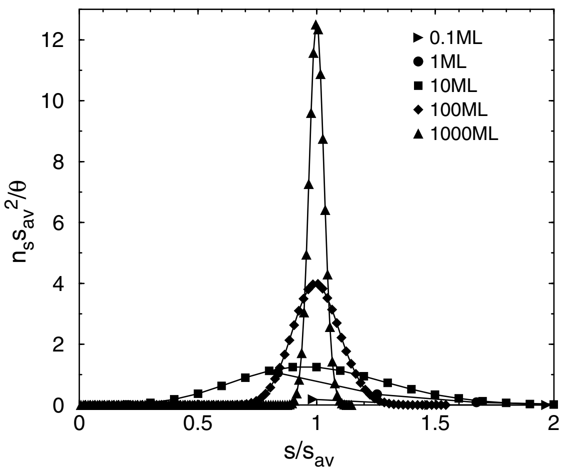

Fig. 3. The scaled ISD in the extreme limit of no diffusion $D/F = 0$ at different coverages Θ. Note the scale of the y-axis when comparing to Fig. 1.

Our results suggest that the sharpness of the ISD is directly related to the island density N. For example, we can see that the ISD for $D/F = 10^4$ has a peak height of approx. 0.85 for $\Theta = 10^2$ ML. A very similar shape (and peak height) is reached for $D/F = 10^5$ and $D/F = 10^6$ at coverages of $\Theta = 10^3$ ML and $\Theta = 10^4$ ML, respectively. This can be explained as follows: For point islands, the island density evolves according to $N \sim (D/F)^{-1/3}\Theta^{1/3}$. Thus, increasing D/F by a factor of 10 leads to the same N if one also increases Θ by a factor of 10. The shape of the ISD for different values of D/F is a function of the coverage, but it is approximately the same if the ratio of D/F to Θ is kept fixed. Thus, we can estimate that an ISD with a peak height of 0.85 would be attained for $D/F = 10^7$ for a coverage of $\Theta \simeq 10^5$ ML [cf. Fig. 1(d)]. This is 100 times larger than the coverages shown. In other words, we expect the distribution function for $D/F = 10^7$ to approach a singular shape at a time that is just beyond a reasonable computational time. The main point is that this sharpening *has to occur* at large enough Θ for *any* value of D/F.

So far we have discussed that there is no scaling of the ISD for different values of Θ for a fixed D/F. But one might ask: Is there at least scaling of the ISD for different D/F at a fixed value of Θ? But from the previous discussion it is clear that this can not be the case: At a fixed Θ, the number of islands that have been nucleated is different for different values of D/F (cf. Fig. 2). But since all ISDs ultimately become singular as $\Theta \to \infty$, one could rephrase this and say that the ISD for different D/F is at a different stage toward becoming singular at a fixed coverage. Our results show that one should only expect data collapse if one keeps the ratio of D/F to Θ (or, equivalently, the island density N) fixed. But different values for this ratio lead to different ISDs. Thus, at fixed Θ and different D/F the ISD changes. Of course, as explained above, this effect is small in some intermediate quasi-scaling regime, when N is significantly less than 1, and D/F is sufficiently large.

At this point one might object that the explanation given above is just an effect of the discrete size of our realization of point islands. Indeed, we believe that the distribution of capture areas of true point islands (that have no spatial extent) might scale and be non-singular. However, there is still continuous nucleation, which continuously re-arranges the capture areas of the islands, and thus destroys the requirement for scaling of the ISD. Furthermore, any numerical implementation of a point island that has physical justification in one way or another has to attribute a discrete size to the point island. Another possible objection is that one might obtain scaling if one takes the limit $D/F \to \infty$ first, and then looks at the behavior of the ISD as $\Theta \to \infty$. We believe that the question of "which limit to take first" might be an interesting question to be studied elsewhere, but is not the focus of this paper. Furthermore, it is impossible to realize this limit in any model.

We would like to conclude our discussion about point islands with the following remark: A lot of effort has been spent on constructing analytic models to reproduce scaling [15,16], and the simplest model that is often employed is a point island model. Such models are typically based on mean field rate equations. There has been some recent controversy [17,18], and it is our belief that such models can never properly describe the scaling of the ISD. In fact, our results presented in this article strongly suggest that at least for point island models one should never expect such scaling, since we have shown that the ISD becomes singular in the asymptotic limit.

We now turn our discussion to the scaled ISD for spatially extended islands. Two fundamental questions that need to be raised are: Is there scaling of the ISD at all, and why is there scaling? We will certainly not answer these questions here, but we believe that the results discussed above shed some new light on these issues. From the preceding discussion it is clear that there cannot be scaling if there is continuous nucleation. But when the spatial extent of islands is taken into account and D/F is large enough, all the islands are nucleated during an infinitesimally short time interval, followed only by a subsequent logarithmic increase in the island density. Thus, there is no reason a-priori that scaling should be ruled out. Fig. 4 shows the scaled ISD for different values of D/F and different coverages for such a model. It is evident that the scaling with D/F is excellent. Close

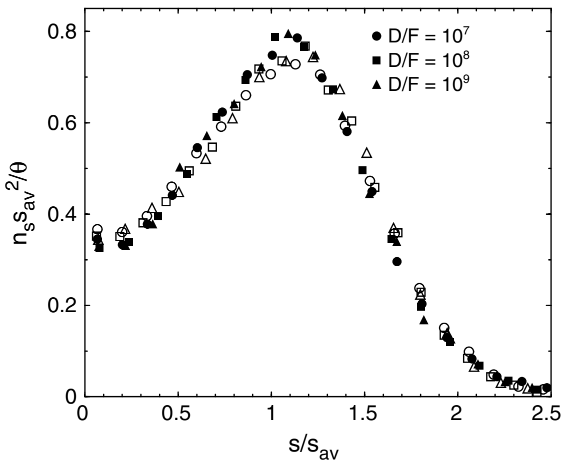

Fig. 4. The scaled ISD in an extended island model at different values of D/F and different coverages. Closed (open) symbols correspond to $\Theta = 0.2$ ML ($\Theta = 0.1$ ML).

inspection of the data indicates that the peak of the data for coverages of $\Theta = 0.1$ ML (open symbols) is slightly below the data for $\Theta = 0.2$ ML (closed symbols). This small discrepancy might be caused by one (or both) of the following: (i) the length of the nucleation phase is still too long for the chosen values of D/F, or (ii) the logarithmic correction to the island density is not yet negligible. We believe that both of these will be removed in the limit $D/F \to \infty$. But we note that it is not possible computationally to increase D/F much beyond what we have shown, and that therefore one cannot study the coverage-ranges over many orders of magnitude, as we did for the point island model above.

Scaling of the ISD in the limit $D/F \to \infty$ is obtained for extended islands because the following necessary condition is met: Essentially all islands are nucleated in a sufficiently short time interval, such that the size of the islands in the scaling limit is much larger than it was at the end of the nucleation phase. Thus, after a certain amount of time, the size of the islands is completely independent of their sizes at the end of the nucleation phase. It only depends on the spatial arrangement of the islands, or on the distribution of the corresponding capture zones. We note that the data of Mulheran and Blackman [7] suggests that the distribution for stationary capture areas (that do not change as the islands grow) scales according to a more peaked gamma distribution. It is not clear at this point whether the ISD in Fig. 4 approaches this gamma distribution for $D/F \to \infty$. But it would only happen for values of D/F that are orders of magnitude larger than what can be simulated with current computational resources. More work is needed to truly understand the scaling limit of spatially extended islands.

In this article we have shown that the ISD for point island models does not scale in the asymptotic scaling limit. We have given an explanation why a point island model has to become singular for $\Theta \to \infty$ for any value of D/F. In contrast, it appears that spatially extended islands do scale in the scaling limit, and the reason is that there is no continuous nucleation of new islands.

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