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The influence of steps on the island distribution function in thin solid film formation

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Abstract

The gamma distribution has been shown to be the simplest function for describing the universal size distribution function of InAs quantum dots on GaAs(001) substrate (Fanfoni *et al* 2007 *Phys. Rev.* B **75** 245312), although there is no reason to believe that it could not be applied to any film growth where the principal feeding of the islands occurs through surface diffusion as proposed, after numerical simulations in the two-dimensional case, by Mulheran and Blackman (1995 *Phil. Mag. Lett.* **72** 55). As far as InAs/GaAs(001) quantum dots are concerned, by fitting the data of Ebiko *et al* (1999 *Phys. Rev.* B **60** 8234) and Krzyzewski *et al* (2002 *Phys. Rev.* B **66** 201302(R)) to the gamma function, $\beta = 4.5$ and 2.9 are the respective returned values. The latter value appears anomalous because for a Poissonian distribution of points (centers of islands), $\beta = 3.5$–3.6. Moreover, the greater the spatial correlation the greater the β value. We prove that the presence of steps alters the distribution of nucleation centers throughout the substrate in such a way that its variance increases with respect to the substrate without steps and, as a consequence, β decreases.

(Some figures in this article are in colour only in the electronic version)

The scale invariance of the nucleus distribution function which emerges during thin film formation has been widely discussed in the literature and was first proposed after a thorough theoretical and numerical analysis for two-dimensional (2D) islands [1–5]. Experimental corroborations soon followed [6–8]. The central result of this analysis can be summed up by the following relation:

$$N_s(\Theta) = \frac{\Theta}{\langle s \rangle^2} f\left(\frac{s}{\langle s \rangle}\right), \tag{1}$$

where N_s is the number density per site of islands of average size $\langle s \rangle$ at coverage Θ and $f(\frac{s}{\langle s \rangle})$ is the scaling function (universal distribution function). Also the form of $f(x)$ has been debated and several proposals have been put forward in the literature [1, 3, 4].

A persuasive physical explanation of the scale invariance has been provided by Mulheran and Blackman [4, 5], who identified it with the invariance of the Voronoi tessellation [9] associated to the nucleation centers. This is all the more true the faster the nucleation process is. This finds a confirmation in the very high value of the parameter $R = D/F$ used in the kinetic Monte Carlo simulations (D is the diffusion coefficient of monomers, F is the impinging flux). The reason is that the faster the nucleation is, the faster the saturation number of nuclei is reached and the faster the asymptotic tessellation is achieved. The fact that the invariance of the Voronoi tessellation warrants a scale invariance depends on the fact that each nucleus grows, on average, proportionally to the area of its associated Voronoi polygon (Vp), that is, to the area on which it exerts influence [10]. Venables and Bell [11] named this area as *the capture zone*. It is quite clear that the distribution of dot sizes must resemble the distribution of the Vps associated to nucleation centers[1]. The latter has been suggested by Kiang [12] who conjectured that the exact 1D

[1] To be right one should consider the Voronoi edge–edge tessellation or the diffusion cells [10] because of dot growth; however, in the case of fast nucleation and low nuclei density, Voronoi tessellation referred to the nuclei is a good approximation [16].

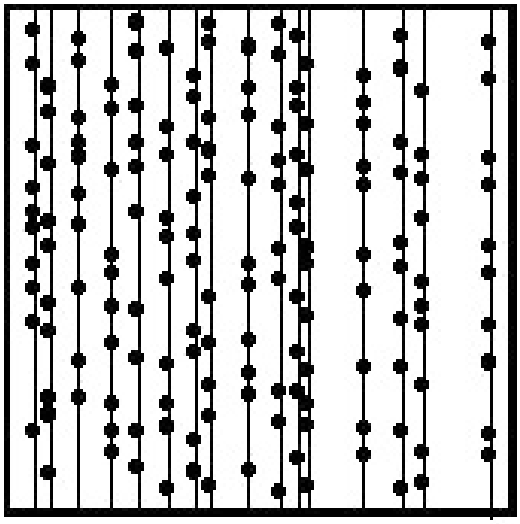

Figure 1. Sketch of the employed model. The lines (steps) are chosen along the x direction on a square lattice possibly with a correlation length s_x. The dots (centers of the islands or quantum dots) are chosen along the lines, possibly with a correlation length d_y.

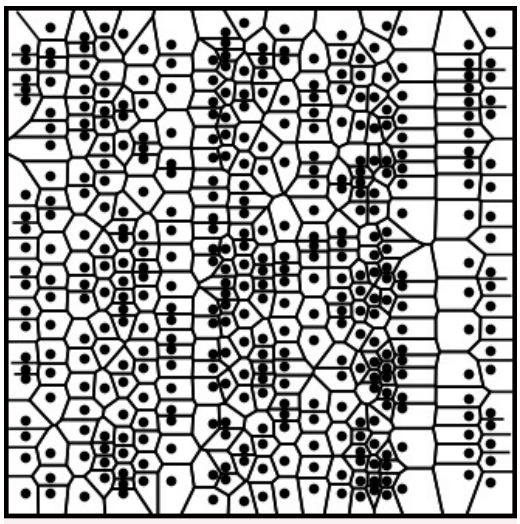

Figure 2. Example of Voronoi tessellation for a 1000 × 1000 lattice with 20 steps and 20 dots per step. The code, on the contrary, on a square lattice 6000 × 6000, chooses 3600 dots distributed along 60 steps, 60 dots per step.

distribution form is valid in any dimension; the distribution is

$$f_\beta(x) = \frac{\beta^\beta}{\Gamma(\beta)} x^{\beta-1} \mathrm{e}^{-\beta x}, \tag{2}$$

where $\beta \in \mathbb{R}$ is a parameter and $\Gamma(x)$ is the Euler's gamma function, $x = s/\langle s \rangle$. It is also evident that once the nuclei become larger and larger, tessellation begins changing noticeably and the invariance is lost even before coalescence or impingement takes place.

The capture zone interpretation of the scale invariance promptly implies that its emergence is not limited to a 2D island growth, but can be extended to the 3D case as well, provided the number of monomers making up islands is taken into account. This is exactly what Ebiko *et al* [13] have shown to hold in the case of InAs quantum dots on GaAs(001) substrate. Their results have been subsequently confirmed by Krzyzewski *et al* [14] and Arciprete *et al* [15]. In all these studies the authors used the Amar and Family (AF) [3] universal function to justify the experimental results. Nevertheless, in a recent paper [16] it has been demonstrated experimentally that, when the nucleation is a reasonably fast process, conditions come into play for which the scale invariance directly follows; during thin film formation characterized by nucleation and growth, in a certain range of coverage. The authors demonstrate that the equation (2) is the more appropriate distribution function, in contrast to other studies [13–15]. Incidentally, Ebiko *et al* [13] explicitly declare that the AF function follows the experimental behavior only qualitatively. In [16] the invariance is proved by fitting the experimental data to equation (2) and showing that the β parameter remains constant within a certain range of coverage.

In [13, 14] it is displayed that the scaled distributions of dot volume collapse, within the experimental error, into the same curve, yet no fit to any function is worked out. We have digitized and fitted to equation (2) the distributions showed in figure 3(b) of [13] and figure 2(d) of [14]. While the distribution of [13] returns $\beta \cong 4.5$, the same as in [16], that of [14] returns $\beta \cong 2.9$. At first sight, this seems a peculiar result given that $\beta \cong 3.6$ [4] corresponds to a Poissonian spatial distribution of nuclei.

This brief report is devoted to demonstrating that a similar value is not strange at all, and may be due to the presence of steps, where, it has been established by now, that nucleation takes place [17].

The substrate has been represented by a 6000 × 6000 square lattice where 3600 points, representing the centers of the dots (DC), have been chosen with the following criteria. Along the x-axis the code chooses randomly 60 values: they are the coordinates of the steps. Along the y-axis in correspondence to each step coordinate, 60 values are chosen at random. In figure 1(a) rescaled sketch has been reported merely to clarify graphically the structure of the model employed. Once the dot coordinates are established, the program generates the associated Voronoi tessellation and the relative distribution of the areas of the Voronoi cells (VCs) are calculated. In figure 2 has been reported a typical Voronoi diagram, that, so as to make the picture clearer, refers to a 1000 × 1000 lattice where 20 steps and 20 dots per step have been chosen. In order to reduce the noise of a single distribution the above-mentioned procedure has been repeated ten times and from the average distribution fitted to equation (2), the β value has been determined. In all, we determined three values of β for as many average distributions and their mean and half maximum deviation provide the final β and its error, respectively. The average value is $\beta = 1.54 \pm 0.04$ which has to be compared with $\beta \cong 3.5$ received when nucleation takes place at random throughout the entire substrate. The variances of the two distributions are, respectively, 0.65 and 0.29 (figure 3). In other words, this result suggests that in the presence of steps, there is a larger variety of cell areas and consequently a greater disorder. The latter statement can be demonstrated as follows.

Table 1. Values of β as a function of spatial correlation among steps (s_x) and among dots (d_y). In this case $d = s_x = d_y$ (see the text).

d	0	10	20	30	40	50	60
β	1.54 ± 0.04	1.93 ± 0.09	2.45 ± 0.06	3.2 ± 0.1	4.65 ± 0.15	6.5 ± 0.4	10.9 ± 0.7

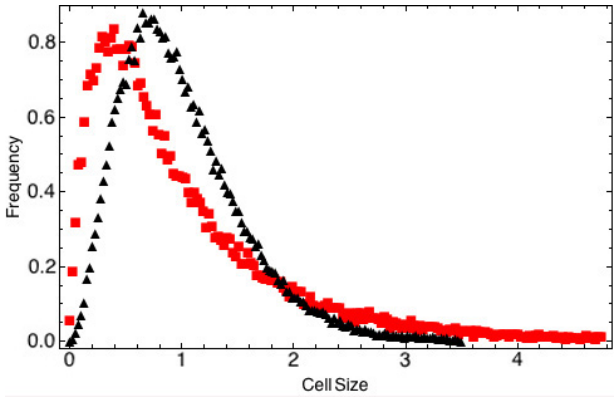

Figure 3. Comparison between the distribution function of the Voronoi areas obtained from stepless (triangles) and stepped (squares) substrates. The variances are 0.29 and 0.65, respectively. In both cases all the correlation lengths are zero.

Let us consider a square of area A and the stochastic variable N_1 which describes the number of DCs contained in A. Let us figure out the probability that $N_1 = n$, where $0 \leqslant n < \infty$. The step coordinates are independent of those of DCs. Inside A there can be k steps each of which contains n_i DCs in such a way that $\sum_{i=1}^{k} n_i = n$; moreover $1 \leqslant k < \infty$. Summing up over all k values and adding the probability of finding no steps inside A, one ends up with

$$P_{m_x,m_y}(N_1 = n) = \chi_n e^{-m_x} + e^{-m_x}\left[\frac{m_x m_y^n}{n!}e^{-m_y} + \sum_{(n_1,n_2,\ldots,n_k)_n} \frac{m_x^k m_y^{\sum_i^k n_i}}{n_1!n_2!\ldots n_k!}\frac{e^{-km_y}}{k!}\right]$$
$$= e^{-m_x}\left[\chi_n + \frac{m_y^n}{n!}\sum_{k=1}^{\infty} m_x^k \frac{k^n}{k!}e^{-km_y}\right] \quad (3)$$

where $m_x = \rho_x\sqrt{A}$ and $m_y = \rho_y\sqrt{A}$ being ρ_x and ρ_y the number densities per unit length along x and y direction, respectively. $\chi_n = 1$ if $n = 0$ while $\chi_n = 0$ if $n \neq 0$. $(n_1, n_2, \ldots, n_k)_n$ indicates the set of all the k-tuples of integers such that $\sum_i^k n_i = n$. In contrast, the probability of finding $N_2 = n$ DCs inside a square of area A for a Poissonian distribution of DCs is

$$P_m(N_2 = n) = \frac{m^n}{n!}e^{-m} \quad (4)$$

where $m = \rho_x\rho_y A$. It is then possible to calculate the variance of the two expressions equation (3) and equation (4) in the case $m = m_1 = m_2 = 1$ obtaining $\mathrm{Var}(N_1) = 2.97$ and $\mathrm{Var}(N_2) = 1$, respectively. This result confirms that in the presence of steps, disorder increases.

The determination of the Voronoi tessellation can also be performed for correlated dots, the centers of which are chosen according to the rule that two steps coordinates cannot lie closer than a distance s_x while two y-dot coordinates cannot lie closer than a distance d_y on a given step.

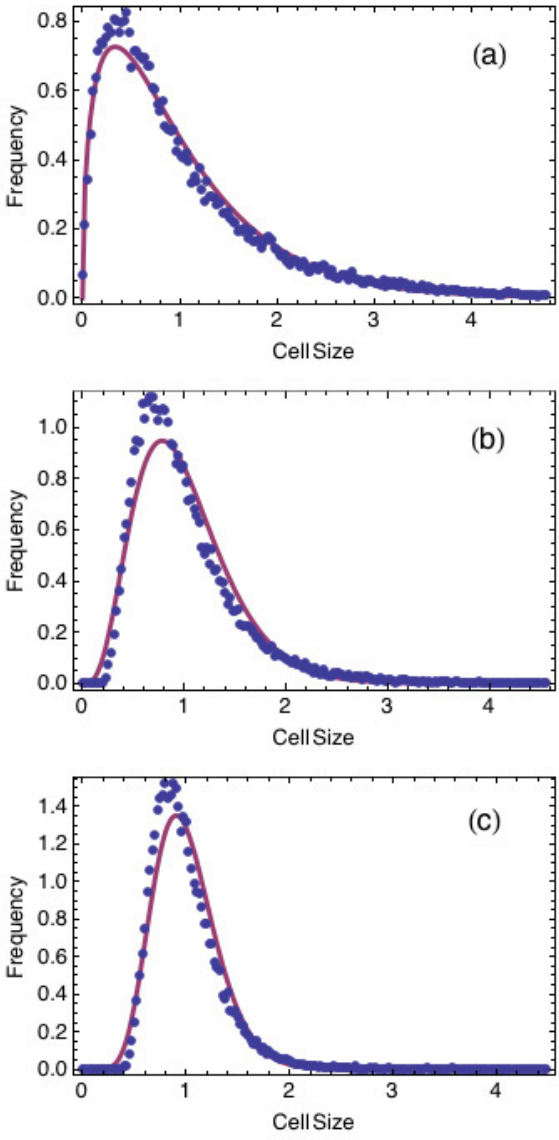

Figure 4. Simulations and relative curve fit for:
(a) $s_x = d_y = d = 0$; (b) $s_x = d_y = d = 30$; (c) $s_x = d_y = d = 60$.

In table 1 we have reported the value of β for some spatial correlations, when $s_x = d_y = d$. These are measured in lattice units.

We need to note that equation (2) is not particularly suitable for describing the VC area distribution as the degree of correlation increases. Incidentally, this is true for a stepless substrate as well. In figure 4 we report, just to give an idea, three distributions and as many curve fits for as many correlation lengths.

Before concluding the article I would like to quote a very recent paper by Pimpinelli and Einstein [18] who proposed a new simple distribution function: the generalized Wigner surmise distribution, in substitution of equation (2). As a matter of fact, it is worth underlining again that the exact analytical form of the distribution function for the Voronoi diagram is known only for 1D case and that distribution function equation (2) is only an arbitrary extension of the 1D solution for higher dimensions. Although their proposal needs checking, it could constitute a valuable advance because it makes it possible to determine, for example, the dimension of the critical nucleus.

In conclusion, we have demonstrated that on a stepped substrate the distribution function of island size is altered with respect to that on a stepless substrate becoming larger. This result makes it possible to explain the 'anomalous' β value got from the distribution published in [14]. We have learned how distribution of steps influences the size distribution of islands or dots in thin film growth.

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