Comment on "Capture-Zone Scaling in Island Nucleation: Universal Fluctuation Behavior"

The Letter [1] proposes a GWS form $g(\alpha) \propto \alpha^{\beta} e^{-b\alpha^2}$ for distribution of capture-zone (CZ) areas, A, for compact islands formed by homogeneous nucleation during surface deposition. Here, $\alpha = A/A_{\rm av}$ where $A_{\rm av}$ is the mean CZ area. Significantly, [1] relates β to the critical size i for stable islands in 2D via $\beta_{\rm GWS} = i + 1$. However, our theoretical and simulation analyses indicate a more complex form for g and a different larger β versus i.

A fundamental theory for CZ areas can be based on the evolution equation for the joint probability [2,3], $N_{s,A}$, for islands of size s with capture zones of area A. A moment analysis summing over s [4] yields an exact evolution equation for the CZ area distribution, $N_A = \sum_s N_{s,A}$, of the form $dN_A/dt = (P_A^+ - P_A + P_A^*)dN_{\rm isl}/dt$. Here, $N_{\rm isl} = \sum_A N_A$ is the island density, P_A is the probability that the (new) CZ of a just-nucleated island overlaps a preexisting CZ of area A, P_A^+ that formation of a new CZ reduces to A the area of a larger preexisting CZ, and P_A^* that a new CZ has area A. Also, $\sum_A P_A = \sum_A P_A^+ = M \approx 4.6$ is the average number of existing CZ's overlapped by the new CZ [3], and $\sum_A P_A^* = 1$. These P's depend on the spatial aspects of island nucleation which occurs predominantly near CZ boundaries [3,5].

We focus on the scaling regime of large $A_{\rm av} = 1/N_{\rm isl}$, where $N_A \approx (N_{\rm isl}/A_{\rm av})g(A/A_{\rm av})$ with $\int g(\alpha)d\alpha = 1$ [3]. We write $P_A \approx M(A_{\rm av})^{-1}p(A/A_{\rm av})$ and $P_A^* \approx (A_{\rm av})^{-1}p^*(A/A_{\rm av})$ with $\int p(\alpha)d\alpha = \int p^*(\alpha)d\alpha = 1$. Since one expects that $P_A \propto N_A$, we set $p(\alpha) = g(\alpha)q(\alpha)$ where $q(\alpha) \sim \alpha^{n\approx 1.5}$ measures the intrinsic probability that a new CZ overlaps an existing CZ of scaled area α [3]. This yields the exact equation [4]

$$2g(\alpha) + \alpha dg(\alpha)/d\alpha = M\langle(1+\alpha'/\alpha)g(\alpha+\alpha')q(\alpha+\alpha')\rangle' - Mg(\alpha)q(\alpha) + p^*(\alpha).$$

Here, $\langle\cdots\rangle'$ denotes an average over the fractional overlap $\mu = \alpha'/(\alpha+\alpha')$ of a new CZ with an existing CZ of scaled area $\alpha + \alpha'$ (thereby creating a CZ of area α), and $\mu_{\rm av} = 0.10$ at 0.1 ML. The complex form of the g-equation precludes simple forms for $g(\alpha)$ (but see [6]), just as the exact equation for the island size distribution precludes popular simple forms for this quantity [3].

For *small-α behavior*, the key is that existing islands with *small* CZ's are *not* required to create small CZ's, contrasting [1]. A new small CZ may come from island nucleation along a line joining $m = 2$ nearby islands or within a triangle of $m = 3$ nearby islands (Fig. 1), none of which have a small CZ. The relative probability for two islands to have small separation $\mathbf{r}$ scales like $(r/r_{\rm isl})^{i+1}$ where $r_{\rm isl} \sim \sqrt{A_{\rm av}}$ is the mean island separation, and for a small pair or triangle with any orientation scales like $P_m \sim (r/r_{\rm isl})(r/r_{\rm isl})^{(m-1)(i+1)}$. The relative probability to nucleate in the target region is $P_{\rm nuc} \sim (r/r_{\rm isl})^{2i+4}$ (cf. [5]), and $p^*(\alpha) \sim P_m P_{\rm nuc}$. In this picture, p^* dominates the right-hand side (RHS) of the g equation so $g(\alpha) \approx (2+\beta)^{-1}p^*(\alpha)$ for small α, and $\beta_m \approx (m+1)\times(i+1)/2 + 3/2$, well above $\beta_{\rm GWS} = i + 1$. The contribution from $m = 2$ likely dominates, but this depends on coverage and island structure. Also, small CZ's can be created differently, e.g., if island C nucleates near a close pair AB and subsequently island D nucleates to enclose C in a small ABD triangle. This corresponds to the P_A^+ term in dN_A/dt. Analysis [4] also indicates large β values for such mechanisms.

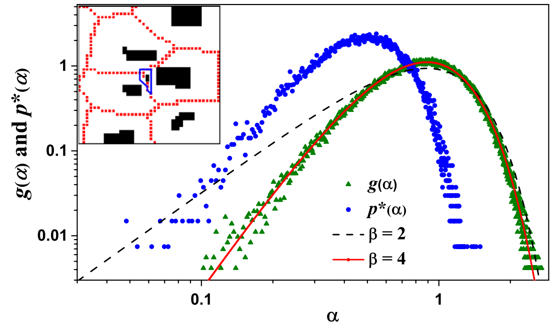

FIG. 1 (color online). Simulation data for $g(\alpha)$ and $p^*(\alpha)$ for $i = 1$ at 0.1 ML. Fits: $\beta = 2$, $n = 2$ (GWS) and $\beta = 4$, $n = 1.5$ (GG) [6]. Inset: smallest new CZ from $\sim 10^5$ cases.

Extensive simulation data for $i = 1$ (3×10^5 CZ's) for compact islands at 0.1 ML supports the above type of relation between g and p^*. An excellent fit for small α (but also for the entire g) is $\beta \approx 4$ with $n = 1.5$ [6] cf. $\beta_{\rm GWS} = 2$. See Fig. 1. For $i = 0$ (3×10^5 CZ's) at 0.1 ML, we find $\beta \approx 3$ with $n = 1.3$ cf. $\beta_{\rm GWS} = 1$.

Work supported by NSF Grant No. CHE-0809472 (Y. H., J. W. E.) and by NSF China Grant 10704088 (M. L.).

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Received 8 January 2010; published 9 April 2010
DOI: 10.1103/PhysRevLett.104.149601
PACS numbers: 68.35.−p, 05.10.Gg, 68.55.A−, 81.15.Aa

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[6] Integration for large α gives $g \sim e^{-M\int q(\alpha)/\alpha d\alpha} \sim e^{-b\alpha^n}$, suggesting a generalized gamma (GG) fit $g \sim \alpha^{\beta} e^{-b\alpha^n}$.

Pimpinelli and Einstein Reply: In [1], we proposed analyzing the capture-zone (CZ) distribution of islands in submonolayer epitaxial growth by fitting with the generalized Wigner surmise (GWS) [2]: $P_\beta(s) = a_\beta s^\beta \exp(-b_\beta s^2)$; s is the CZ area A over its mean $\langle A \rangle$, and β is the sole adjustable parameter. Our mean-field (MF) argument for $P_\beta(s)$ also suggested that β was the size of the smallest stable nucleus of an island, $i+1$ (i.e., i is the critical nucleus), in dimensions $d \geq 2$, and $2(i+1)$ in $1d$. $P_\beta(s)$ fits experimental data at least as well as the alternatives. Furthermore, much (but not all) Monte Carlo data supported the deduced value of β in terms of i for $1d$ and $2d$. However, more thorough analysis and numerical testing was clearly warranted.

Recently, Amar's [3] and Evans's groups [4] [SSA and LHE, respectively] have taken up this challenge and produced extensive numerical data, SSA for two models of point islands in $d = 1, 2, 3, 4$ [5], and LHE for compact islands in $2d$, the case more appropriate for comparison with experiment. Space limits our focus here to $2d$. Both groups' results differ notably from our MF description, arguably reminiscent of using mean field for critical phenomena. Specifically, with $i = 1$ and fractional coverage $\theta = 0.1$, SSA found for both point-island models that β was closer to 3 than our MF-predicted $\beta = 2$. Up to $\theta \geq 0.4$, β did not change with θ, but β decreased modestly as D/F, the ratio of the rates for atom hopping and for deposition, ramped up over 10^5–10^{10}, reaching $\beta \approx 2.8$ as $D/F \to \infty$ [6].

For compact islands with $i = 1$, LHE's data is likewise better described by $\beta \approx 3$ than 2—cf. Fig. 1. Also, the variance is that of a GWS with $\beta = 2.97$. LHE's data for $i = 0$ is even closer to $\beta = 2$, and the variance yields $\beta = 1.90$. Both SSA and LHE find $\beta \approx i + 2$ accounts for the data better than $i + 1$. However, the distribution is more skewed than $P_\beta(s)$. LHE find the optimal fit occurs with a distribution between GWS and the oft-used gamma distribution $G_\alpha(s)$ [7]. The log-log plot in their Fig. 1 suppresses this exponential factor for small s; their plot supports $\beta \approx 4$. We advocate emphasizing data near the peak, where the count rate is highest and the fractional error is smallest. This procedure is especially warranted when dealing with experimental data, in which the number of CZs is 2–3 orders of magnitude smaller than in these simulations. Figure 1 shows that $\beta = 3$ describes the overall data better than $\beta = 4$, especially regarding width and peak height [6]. Fits with $P_3(s)$ and $G_7(s)$ are comparable [as are fits of LHE's unpublished data for $i = 0$ by $P_2(s)$ and $G_5(s)$].

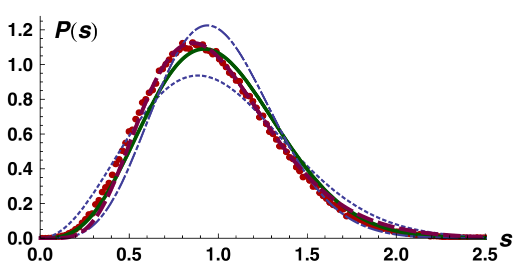

FIG. 1 (color online). Plots of LHE's numerical data [red dots] for the CZD ["$g(\alpha)$"] for $i = 1$ (their Fig. 1) and $P_n(s)$, $n = 2$ [dotted, blue line], 3 [solid, green line], and 4 [dash-dotted, blue line], along with $G_7(s) \propto s^6 e^{-7s}$ [dashed, purple line].

In [1], we assumed that the nucleation probability $\propto n^{i+1}$, where n is the adatom density. We then wrote $n \propto \bar{n}A/\langle A \rangle \equiv \bar{n}s$. Thus, the nucleation rate NR $\propto \bar{n}^{i+1}s^{i+1}$. But NR is also $\propto \bar{n}^{i+1}P(s)$. Thus, $P(s) \propto s^{i+1}$. SSA's and LHE's simulations imply that this argument is insufficient. We go beyond MF for small adatom coverage, thereby showing that larger exponents of s can arise.

In $2d$, the adatom density $n(r) \propto R^2 - r^2$, with $R_i < r < R$, where R and R_i are the radii of the CZ and island, respectively. Then, we find the total NR by integrating between these two radii, but $R_i \to 0$ for point islands, as well as for compact islands at small coverage; hence,

$$\int_{R_i \to 0}^{R} dr r[n(r)]^{i+1} \propto R^{2i+4} \propto A^{i+2} \Rightarrow P(s) \propto s^{i+2},$$

consistent with $\beta \approx 3$ (2) for $i = 1$ (0) in $2d$ [8].

The main points are that $P_\beta(s)$ accounts well for CZD, with physical information in β. The addend to i turns out to be larger than the MF prediction of 1, closer to 2, in this fascinating problem. In many experimental instances, the question is whether β changes, e.g., when impurities are added to the system [9].

Work at UMD supported by the NSF-MRSEC, Grant No. DMR 05-20471. We thank J. W. Evans for sharing LHE's data, and him and J. G. Amar for fruitful discussions.

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Received 18 February 2010; published 9 April 2010
DOI: 10.1103/PhysRevLett.104.149602
PACS numbers: 68.35.−p, 05.10.Gg, 05.40.−a, 81.15.Aa

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