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, $\alpha = A/A_{\text{av}}$ where A_{av} is the mean CZ area. Significantly, [1] relates β to the critical size i for stable islands in 2D via $\beta_{\text{GWS}} = i + 1$. 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 $dN_A/dt = (P_A^+ - P_A + P_A^*) dN_{\text{isl}}/dt$. Here, $N_{\text{isl}} = \sum_A N_A$ 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 $A_{\text{av}} = 1/N_{\text{isl}}$, where $N_A \approx (N_{\text{isl}}/A_{\text{av}}) g(A/A_{\text{av}})$ with $\int g(\alpha) d\alpha = 1$ [3]. We write $P_A \approx M (A_{\text{av}})^{-1} p(A/A_{\text{av}})$ and $P_A^* \approx (A_{\text{av}})^{-1} p^*(A/A_{\text{av}})$ 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]

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

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 $\mu_{\text{av}} = 0.10$ 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 $(r/r_{\text{isl}})^{i+1}$ where $r_{\text{isl}} \sim \sqrt{A_{\text{av}}}$ is the mean island separation, and for a small pair or triangle with any orientation scales like $P_m \sim (r/r_{\text{isl}})(r/r_{\text{isl}})^{(m-1)(i+1)}$. The relative probability to nucleate in the target region is $P_{\text{nuc}} \sim (r/r_{\text{isl}})^{2i+4}$ (cf. [5]), and

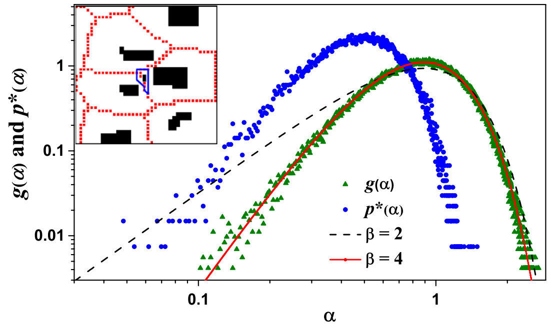

FIG. 1 (color online). Simulation data for $g(\alpha)$ and $p^*(\alpha)$ for $i = 1$ at 0.1 ML. Fits: $\beta = 2$, $n = 2$ (GWS) and $\beta = 4$, $n = 1.5$ (GG) [6]. Inset: smallest new CZ from $\sim 10^5$ cases.

$p^*(\alpha) \sim P_m P_{\text{nuc}}$. 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 $\beta_{\text{GWS}} = i + 1$. 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.

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. $\beta_{\text{GWS}} = 2$. 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. $\beta_{\text{GWS}} = 1$.

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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 $g \sim \alpha^{\beta} e^{-b\alpha^n}$.

