Singularities and Spatial Fluctuations in Submonolayer Epitaxy

Amar *et al.* [1] have proposed an extension of rate equations for submonolayer epitaxial growth with which they claim to "solve the long lasting problem of determining the correlations between the size of an island and that of its capture zone." In this Comment, we point out that their solution of these equations does not give the correct scaling of the island-size distribution (ISD) and, because of the way averages over capture zones are carried out, cannot do so even in principle.

The ISD is the central quantity for submonolayer epitaxy, but has proven notoriously difficult to calculate by any means other than simulation. Scaling mandates that the ISD for different coverages θ and values of $R \equiv D/F$, where D is the adatom diffusion constant and F is the deposition flux, collapse onto a universal curve. The ISDs in Ref. [1] are shown only for single values of R and θ, with no evidence presented for data collapse, but data published later (Fig. 6 of Ref. [2]) clearly indicate a narrowing and sharpening of the ISD in comparison with that obtained from simulations, with an "overshoot" of the peak that "increases with coverage and/or R." This manifestly violates the requirements of scaling.

The reason for this is that the spatial arrangement of islands is the overriding factor for determining the growth rates of individual islands and, therefore, the form of the ISD. These growth rates, which are usually described by "capture numbers" in homogeneous rate equations, have resisted all attempts at analytic treatments beyond the mean-field level. The importance of spatial fluctuations of island environments has been demonstrated [3] by simulations with different seeding styles for island nucleation sites. Only by weighing the choice of these sites by the square of the adatom density was agreement obtained with kinetic Monte Carlo simulations; other seeding styles produced *qualitatively* different ISDs. These are the *only* fluctuations that remain in the scaling limit ($R \to \infty$), so this regime determines whether a theory correctly incorporates the spatial correlations between islands.

In a later paper [4], the fluxes of islands of size s across the interval $[s, s+1)$ were calculated directly from simulations and used to determine effective capture numbers. The resulting ISDs, obtained by integrating rate equations with these capture numbers, were statistically indistinguishable from those obtained from simulations. However, replacing these capture numbers with smooth functions proved unsuccessful, ultimately because of the nontrivial cross correlations between island sizes and capture zones [5]. Only for large islands is there a one-to-one correspondence between their sizes and capture zones [4,5], allowing these correlations to be neglected.

As the foregoing makes clear, the ISD is an embodiment of the spatial arrangement of islands. The explicit inclusion of capture zone areas in models of nucleation and aggregation kinetics, whether within a rate equation framework [1] or otherwise [5], addresses this, but at a substantial cost: The creation of new capture zones and the accompanying fragmentation of existing capture zones caused by nucleation must be somehow specified. The inherently stochastic nature of nucleation [3] leads to the dispersion of the island-size distribution about the most probable size, which thereby suppresses the development of singularities as $R \to \infty$. The calculations in Ref. [1] were carried out with an *ad hoc* mean-field description for the correlations between capture zones and island sizes wherein the nth island has a capture area that is proportional to the average area per island, which is $1/n$. This clearly leads to *some* spreading of the island-size distribution but, as R increases, the level of agreement between the resulting ISDs and kinetic Monte Carlo simulations diminishes [2]. Thus, the results presented in Ref. [1] are not typical of the theory as a whole. The essential point is that ISDs are fluctuation-dominated entities, so even the detailed mean-field theory proposed by Amar *et al.* [1] only delays the appearance of singularities.

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