Growth coalescence shapes for islands during metal (100) homoepitaxy

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Abstract
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Keywords
film, geometry, molecular dynamics, molecular model, Monte Carlo method, surface property

Disciplines
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Comments
Growth coalescence shapes for islands during metal (100) homoepitaxy

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During submonolayer homoepitaxy, instability in the shapes of growing two-dimensional islands can develop due to the diffusion-limited aggregation of deposited adatoms at their edges. However, in metal (100) systems, periphery diffusion is typically efficient, quenching this shape instability, and resulting in simple near-square or near-rectangular shapes of isolated islands. Despite this feature, growth coalescence shapes resulting from collision of two or more growing islands are nontrivial. These coalescence shapes are elucidated here by developing three complementary formulations: (i) suitable atomistic lattice-gas models analyzed by kinetic Monte Carlo simulation; (ii) deterministic rate equations for the dynamics of kinks along island step edges; and (iii) continuum theories for step-edge evolution. Characterization of coalescence shapes is important as they affect interlayer transport during multilayer growth. Such a characterization is also necessary to enable coarse-grained modeling of film growth with a realistic treatment of the evolution of island edge morphologies, e.g., using level-set methods.

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I. INTRODUCTION

Two-dimensional (2D) metal islands formed during deposition on fcc (111) or hcp (0001) surfaces at lower temperatures often exhibit dendritic or fractal forms, very different from their compact equilibrium shapes. Island shape relaxation in these systems is not efficient enough to quench the shape instability associated with diffusion-limited aggregation (DLA) of deposited adatoms with these growing islands. Such shape relaxation is typically mediated by periphery diffusion (PD) of adatoms at island edges, and detailed models with suitably “high” PD barriers have recovered irregular island shapes observed in specific epitaxial thin film systems.

In contrast, for deposition on fcc (100) or bcc (100) surfaces, and particularly for homoepitaxy, typically individual 2D metal islands are near-square or near-rectangular resembling their equilibrium shapes. Edges are aligned with the close-packed [110] step directions. It is recognized that these compact near-square shapes are due to both efficient PD of singly-coordinated edge adatoms along [110] step edges and to efficient rounding of kinks along the step edge by these adatoms to reach higher coordinated sites (i.e., doubly coordinated kink sites). Thus, relatively little attention has been paid to the issue of the “less interesting” growth shapes in such systems.

However, even in such metal (100) systems with simple compact shapes of individual islands, it remains to develop a comprehensive understanding of the shape of coalescing pairs or larger clusters of islands during growth under continued deposition. We call this phenomenon “growth coalescence.” In some previous work, it has been termed “static coalescence,” presumably to reflect the feature that the centers of the islands are roughly static during deposition. This phenomenon should be distinguished from so-called “dynamic coalescence” of diffusing islands in the absence of deposition. The latter has been modeled in detail for metal (100) homoepitaxial systems. Except for small island sizes, behavior can be described quantitatively by a two-dimensional, anisotropic version of a Mullins continuum theory for shape relaxation via PD. By default, a Mullins-type treatment is often adopted for growth shapes or growth coalescence. This is not generally valid, as we clarify in subsequent discussion.

Growth coalescence shapes in metal (100) homoepitaxy are important in determining interlayer transport and thus film evolution during multilayer growth. Their influence on film roughness is particularly clear given the expected presence of nonuniform (orientation-dependent) Ehrlich-Schwoebel step-edge barriers to downward transport. Specifically, the step-edge barrier along open or kinked [100] step edges should be negligible compared with the small but significant barrier on close-packed [110] step edges. Thus, any degree of rounding of coalescence shapes away from [110] orientations will facilitate downward transport.

A natural goal of detailed atomistic studies of growth coalescence would be to develop appropriate coarse-grained or continuum theories for the evolution of these shapes. Such formulations of growth coalescence are needed as input to coarse-grained modeling of the overall process of island formation during deposition. In such modeling, island-edge locations are often described by continuous curves, and the flux of diffusing adatoms aggregating with growing islands is obtained from analysis of an appropriate boundary value problem for continuum deposition-diffusion equation. However, an appropriate continuum partial differential equation (PDE)–based theory of island growth and coalescence shapes is also needed to facilitate implementation of realistic, system-specific models. In particular, such theories are essential in order to fully exploit the potential of the level-set approach that is geared to conveniently track edge locations during island coalescence. Also, for recently developed geometry-based-simulation algorithms, one needs to provide “by hand” a specification of growth coalescence shapes in order to treat the regime of higher coverages where island coalescence and percolation are prevalent.
One such model involves efficient kink rounding (EKR) only for single-atom high kinks, and negligible rounding for multiple-height kinks including “global corners” of square islands.\(^7\) The motivation for this model is that kink rounding may occur through a facile two-atom exchange process, a pathway that is less likely available at global corners. Then, growing islands adopt near-rectangular (rather than just near-square) shapes, as different numbers of adatoms are in general impinging and accumulating on the four different sides of the island.\(^19\) In our simulations of this model, diffusing atoms reaching the edge of the island are immediately moved to a “nearby” kink site, provided that this is possible without rounding a global corner. In the rare cases where no kink can be reached, e.g., for a perfect rectangular island, the atom is left at its impact site and acts as the nucleus for a new layer.

For the EKR model, one could further distinguish between two cases: the zero extra “kink Ehrlich-Schwoebel” barrier to round kinks (EKR\(_0\)) versus a “small” extra barrier (EKR\(\delta\)). For the zero extra barrier, the aggregating atom is typically captured by a kink on the step edge either immediately to the left or right of the aggregation site. The probabilities to be captured at the left or right neighboring kink site reflect only the relative distances to these kinks, irrespective of whether the adatom has to round a kink to reach those sites. The precise form of these probabilities is determined by analysis of the problem of a 1D random walk between two traps: they vary linearly with the distance from the kink.\(^13\) For the “small” extra barrier, one can imagine that adatoms will preferentially attach to a kink which can be reached without kink rounding.\(^20\)

For growth of individual near-square or near-rectangular islands, there is no significant difference between EKR\(_0\) and EKR\(\delta\). Growth of each of the four sides occurs in a layer-by-layer fashion, new layers being nucleated once a side is completed. However, for growth coalescence of pairs of islands that collide roughly corner-to-corner, typically adatoms aggregating in the neck region can reach one neighboring kink site without kink rounding, and the other by kink rounding (see Figs. 1 and 2). This produces a significant difference between the behavior for EKR\(_0\) and EKR\(\delta\) models in the neck region. For EKR\(_0\), adatoms will round kinks roughly half the time in attaching to double-coordinated kink sites. For EKR\(\delta\), they will never round kink sites in the neck region (so local behavior is equivalent to models with no kink rounding); see Sec. III.

Another reasonable model involves efficient corner rounding (ECR), irrespective of whether corners are single-atom high kinks, or multiple-height kinks or “global” corners of square islands. Here growth of individual islands occurs by nucleation and completion of one side at a time, so the picture is similar to EKR, except that islands tend to be more square as mass transport occurs between the four sides. However, for growth coalescence, there is a significant difference as growth of neck regions is now fed by atoms depositing at distant points on outer straight edges on the perimeter (see Sec. III (B)).

Growth coalescence becomes prevalent at coverages \(\theta \geq 0.25\) monolayers (ML) where a significant fraction (equal to \(\theta\)) of atoms are deposited on top of islands. Thus, we must
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FIG. 2. Detailed schematic of the upper left neck region for the precise corner-to-corner coalescence geometry in Fig. 1. $L_0$ is the edge length of the individual square islands at the point of coalescence. S denotes the symmetry point, and C denotes the corner point which separates the outer edge and neck region. The terminology for kink positions, terrace widths, aggregation flux, etc., corresponds to that used in our DKD formalism in Sec.2.2.

also specify the treatment of (downward) interlayer diffusion for atoms landing on top of the islands. Then the existence of an additional Ehrlich-Schwoebel (ES) step-edge barrier to downward transport becomes important. Three basic possibilities that one might consider are no ES barrier (corresponding to uninhibited interlayer transport); nonuniform ES barrier (set to zero along kinked or open step edges); and infinite ES barrier (corresponding to no interlayer transport).

Finally, it is also important to emphasize that in these island growth studies, the flux of adatoms diffusing on the terrace and aggregating with the island edge is nonuniform along the edge. It is enhanced at protruding corners of islands, and inhibited at concave neck regions formed during coalescence. The opposite is the case for the flux of atoms reaching the step edge from on top of islands. These features will affect island growth shapes, complicating their analysis. Thus, sometimes it is instructive to compare island growth behavior with simpler benchmark models where this aggregation flux is taken as uniform along the island perimeter.

B. Modeling based on deterministic kink dynamics

For growth coalescence of islands that collide corner-to-corner, a simplifying feature is that extended portions of the step edge in the key neck region have kinks of one “sign,” i.e., they form a staircase (see Fig. 2). Thus, the situation is analogous to the higher-dimensional problem of step propagation on vicinal surface, especially in the approximation where steps are assumed straight. Consequently, we can adopt the formalism used for these problems. This analogy is further aided since we will see that the step edges during coalescence are for the most part oriented not far from a close-packed direction corresponding to the edge of the square island in the initial configuration. This direction is denoted here as $x$. Thus, typical kink separations in the $x$ direction are many times the surface lattice constant, $a$, and position of the $n$th kink, $x_n$, can be regarded as a continuous variable (although $x_n$ is actually some discrete multiple of $a$). Below, we assume that the step-edge height (denoted by $h$) increases in the orthogonal direction with increasing $x$, for the portion of the step edge under consideration. To be specific, the terrace to the immediate right of $x_n$ is taken to have a height $h=na$ above the initial step height. See again Fig. 2.

Let the flux of adatoms aggregating with the step edge at position $x$ be denoted by $F(x)$, measured in atoms per adsorption site per unit time. In general, $F(x)$ denotes the total flux attaching from both sides of the step, although in our numerical studies below, we consider only the case of an infinite ES barrier where atoms attach just from the lower side (cf. Fig. 2). Then, the evolution equations for these step positions have the form

$$\frac{dx_n}{dt} = V_n = -\int_{x_{n-1}}^{x_n} dx F(x) \text{ for EKR0},$$

$$\frac{dx_n}{dt} = V_n = -\int_{x_{n-1}}^{x_n} dx F(x)(x-x_{n-1})/(x_n-x_{n-1})$$

$$-\int_{x_n}^{x_{n+1}} dx F(x)(x_{n+1}-x)/(x_{n+1}-x_n) \text{ for EKR0}.$$  

Here, $V_n$ denotes the velocity (in the $x$ direction) of the $n$th kink. The EKR0 equation incorporates the linear variation of capture probabilities with the distance from the kink. For the benchmark models with uniform $F$, these reduce to forms

$$\frac{dx_n}{dt} = V_n = F(x_{n-1}-x_n) \text{ for EKR0},$$

$$\frac{dx_n}{dt} = V_n = F(x_{n-1}-x_{n+1})/2 \text{ for EKR0},$$

analogous to equations for step propagation on vicinal surface with infinite and zero Ehrlich-Schwoebel step-edge barriers, respectively.

An instructive alternative formulation focuses on the terrace widths, $l_n = x_{n+1} - x_n$. In the context of our study, suitably normalized $l_n$ corresponds to a height distribution (in the direction orthogonal to $x$) for the portion of the island step edge under consideration. This follows since the height of the portion of the step corresponding to the terrace of width $l_n$ is $h=na$, where again $a$ is the lattice spacing (see Fig. 2). Evolution equations can be obtained from those above for the $x_n$. For the case of uniform $F$, these have the forms

$$\frac{dl_n}{dt} = F(l_{n-1} - l_n) \text{ for EKR0},$$

$$\frac{dl_n}{dt} = F(l_{n-1} - l_{n+1})/2 \text{ for EKR0}.$$  

One must also impose suitable boundary conditions on these equations for values of $n$ corresponding to the limits of the
region of the island step edge under consideration (e.g., C and S in Fig. 2). These conditions are somewhat complicated and specific to the island geometry and the model under consideration. Thus, a detailed discussion is deferred until Sec. III and the Appendixes.

The root mean square roughness, $W$, of the portion of the step edge under consideration in the direction orthogonal to $x$ can be expressed in terms of the terrace widths as

$$W^2 = a^2 \sum_n (n-n_{av})^2 l_n / \sum_n l_n,$$

(5)

where $n_{av} = \sum_n n l_n / \sum_n l_n$.

Here $n_{av}$ gives the average height of the portion of the step edge of interest. In our study, $W$ is used as a useful measure of the degree of rounding of the step edge in the step region during growth coalescence.

C. Continuum modeling of the terrace width distribution

Here, we assume that a smooth curve $x = x(h,t)$ is fit through the portion of the vertically discrete step edge of interest (see Fig. 1), so that $l_n = l(h,t) = \delta x(h,t)/\partial h$, for each $n$, with $h = na$. The function $l(h,t)$ denotes the continuous step width or step-edge height distribution for which we will obtain a continuum Fokker-Planck-type evolution equation (FPE) by taking the continuum limit of the equations in Sec. II (B) for the $l_n$. One can use a Taylor expansion to obtain

$$l_{n+1} = l(h \pm a,t) = l(h,t) \pm a \frac{\partial l(h,t)}{\partial h} + \frac{a^2}{2} \frac{\partial^2 l(h,t)}{\partial h^2} \pm \frac{a^3}{6} \frac{\partial^3 l(h,t)}{\partial h^3} + \cdots,$$

(6)

If $F = F(x)$ denotes the total flux of atoms attaching at the step edge, then for uniform or slowly varying $F$, the $l_n$ equations of Sec. II (B) yield the FPE

$$\frac{\partial l(h,t)}{\partial t} \approx -Fa \frac{\partial l(h,t)}{\partial h} + \frac{a^2}{2} \frac{\partial^2 l(h,t)}{\partial h^2} + \cdots \quad \text{for EKR}\delta,$$

(7)

$$\frac{\partial l(h,t)}{\partial t} \approx -Fa \frac{\partial l(h,t)}{\partial h} + \frac{a^3}{6} \frac{\partial^3 l(h,t)}{\partial h^3} + \cdots \quad \text{for EKR}0,$$

(8)

In general, one must also specify appropriate boundary conditions for these equations at the end of this portion of step edge of interest, e.g., $C$ and $S$ in Fig. 2 (see Sec. III and the Appendixes). The significant role of the boundary conditions is particularly clear for Eq. (8) since integration for all of $-\infty < h < \infty$ would lead to the development of Airy-function-like oscillations corresponding to unphysical negative values of $l(h,t)$. Finally, it is useful to note that one can readily change the independent variable in Eq. (7) or Eq. (8) to $\partial h = h - h^*(t)$, for suitable $h^*$, noting that

$$\frac{\partial l(h,t)}{\partial t} \approx - \frac{\partial l(h^*,t)}{\partial t} \frac{\partial h^*}{\partial t} \frac{\partial l(h^*,t)}{\partial h^*} + \cdots.$$

Thus, choosing $h^*(t) = Fat$ (where growth from the initial configuration starts at time $t=0$) will shift to a reference frame moving with the typical step-edge height, thus eliminating the drift term in the FPE (see Appendix A). Choosing $h^*(t) = h^0(t)$, the height of the corner, $C$, in Fig. 2, will modify the drift term in the FPE for $l(\partial h,t)$ and allow more convenient treatment of the boundary condition at $C$ with fixed $\partial h = 0$.

Next, we provide some general observations regarding the FPE (7) and (8). The first “drift” term in the FPE describes an overall increase in step edge height at rate $Fa$. With no additional terms, this would preserve the form of the terrace width distribution. The second term for EKR$\delta$ in Eq. (7) describes the “diffusive spreading” of the terrace width (or step height) distribution. This behavior is expected by analogy with the rapid roughening corresponding to Poisson-type growth observed in multilayer growth systems with no interlayer transport. An alternative derivation of Eq. (7) can be found in Ref. 24. This diffusive spreading term is absent for EKR0, consistent with the expectation of slower growth of the terrace width or step-height distribution for unrestricted kink rounding (or unrestricted interlayer transport for multilayer growth).

As in Sec. II (B), to quantify growth behavior, we consider the roughness, $W$, of the portion of the step edge under consideration in the direction orthogonal to $x$. Analogous to Eq. (5) in Sec. II (B), $W$ can be expressed as

$$W^2 = \int dh(h-h_{av})^2 l(h,t),$$

(10)

where $h_{av} = \int dh l(h,t) / \int dh l(h,t)$ is the average height of the portion of the step edge of interest. Again, we will use $W$ to measure rounding of the step edge during growth coalescence. After transforming Eqs. (7) and (8) to remove the drift term, it is clear that the form of these equations is consistent with “intrinsic” scaling behavior $W \sim (Ft)^{\beta}$, with $\beta = 1/2 (1/3)$ for EKR$\delta$ (EKR0) (see Appendix A). However, we shall see below that at least for EKR$\delta$, the effect of the boundary condition at $C$ dominates to induce more rapid roughening with $W \sim Ft$. This difference between EKR$\delta$ and EKR0 will be reflected in more rounded coalescence shapes for the former.

D. Continuum modeling of the evolution of step edge height

By analogy with treatments of multilayer film growth, traditional continuum modeling would utilize an evolution equation for a continuous step height function, $h(x,t)$, of a continuous variable $x$, for suitable portions of the step edge.
Again, $h$ is measured orthogonal to the $x$ direction. This equation might be expected to have the form\textsuperscript{25}

$$
\frac{\partial h(x,t)}{\partial t} = a F(x) - \frac{\partial J}{\partial x} + \eta,
$$

(11)

where $F$ denotes the total flux of attaching adatoms, and $J$ denotes the lateral mass current in the $x$ direction along the step edge. Also $\eta$ denotes deposition noise, which will not be significant here as growth is effectively deterministic. Alternatively, it is sometimes instructive to utilize an evolution equation for the local slope of the step edge, $m(x,t) = \partial h(x,t)/\partial x$, which can be obtained from Eq. (11) by differentiation with respect to $x$. Then, $h(x,t)$ can be recovered from $m(x,t)$ by integration. The root-mean-square roughness, $W$, of the portion of the step edge under consideration with average height $h_{av} = \int dx h(x,t)/\int dx$ is now naturally expressed as

$$
W^2 = \frac{\int dx [h(x,t) - h_{av}]^2}{\int dx}.
$$

(12)

Usually the flux $J$ in Eq. (11) is decomposed into nonequilibrium ($J_{ne}$) and equilibrium ($J_{eq}$) components, where the latter has a Mullins-type form $J_{eq} = \sigma \partial \delta h / \partial x$ with step-edge mobility $\sigma$, and step-edge stiffness $\beta$\textsuperscript{13,14,25}. However, in our models, and in more general models with effectively irreversible trapping at kinks, $\sigma$ vanishes and the term $J_{eq}$ is absent, i.e., $J_{ne}$ dominates $J_{eq}$ for such growth processes. In general, $J_{ne}$ can include a component with this Mullins form which is associated with nucleation of new terraces when two diffusing edge adatoms meet\textsuperscript{26}. However, for our models, this component is also absent, as a result of the lack of nucleation due to very rapid PD.

For EKR$\delta$, it is clear that an uphill current exists due to the lack of kink rounding in the neck region.\textsuperscript{25} In the simplest approximation, $J_{ne}$ equals $F(x)$ times one-half the local terrace length $l(x,t) = a/m(x,t)$.\textsuperscript{24} Thus, one has $J = J_{ne} = a F(x) l(x,t)/2$. For EKR$0$, there is no net uphill current, and the analysis is more subtle. Thus, a more systematic approach is desired, starting with the equations from Sec. II (B).

One could attempt to obtain continuum equations of the desired form for $m(x,t) = a/l(x,t)$ from the FPE in Sec. II (C) by applying a suitable Lagrange-type transformation, where one writes $l(h,t) = l(x(h,t),t)$. As a result of this procedure, one obtains

$$
\frac{\partial m(x,t)}{\partial t} = F a^3 \left[ \frac{m_{ss}}{2m^2} - \frac{(m_s)^2}{m^3} \right]
$$

for EKR$\delta$,

(13)

$$
\frac{\partial m(x,t)}{\partial t} = F a^3 \left[ - \frac{m_{sss}}{6m^3} + \frac{3m_t m_{ss}}{2m^2} - \frac{2(m_s)}{m^3} \right]
$$

for EKR$0$.

(14)

Here, the subscript $x$ denotes $\partial/\partial x$. Of course, no noise term appears in these equations since we are starting from a deterministic theory for kink motion. The derivation of Eqs. (13) from (7) was presented in Ref. 24, where it was also noted that this equation is consistent with the heuristic derivation of the evolution equation for $h(x,t)$ presented above for EKR$\delta$.

Since this Langrange transformation procedure is not particularly transparent, a more direct derivation of evolution equations for $h(x,t)$ is instructive. To this end, we adopt the strategy of Refs. 27 and 28, and now suppose that the continuous smooth $h = h(x,t)$ is fit through the vertically discrete step edge so that $h(x_{n+1},t) - h(x_n,t) \approx a$ for each $n$. Then, we use a Taylor expansion to obtain

$$
a = h(x_{n+1}) - h(x_n) = l_n \frac{\partial h(x_n)}{\partial x} + \frac{(l_n)^2}{2} \frac{\partial^2 h(x_n)}{\partial x^2} 
$$

$$
+ \frac{(l_n)^3}{6} \frac{\partial^3 h(x_n)}{\partial x^3} + \ldots,
$$

and a similar result

$$
-a = h(x_{n-1}) - h(x_n) = -l_{n-1} \partial h(x_n)/\partial x + \ldots
$$

involving $l_{n-1}$. Solving (15) for $l_n$ yields

$$
l_n = \frac{a}{h_s(x_n)} - \frac{a^2 h_{xs}(x_n)}{2 h_s(x_n)^3} + \frac{a^3 h_{xss}(x_n)}{2 h_s(x_n)^5}
$$

$$
- \frac{a^3 h_{xxs}(x_n)}{6 h_s(x_n)^4} + \ldots.
$$

(16)

For $l_{n-1}$, one obtains a similar expression with a change of sign in the second term.

Using the key relation\textsuperscript{27,28}

$$
\frac{\partial h(x_n,t)}{\partial t} \approx -V_n \frac{\partial h(x_n,t)}{\partial x},
$$

(17)

one can obtain the desired evolution equation. Specifically, for EKR$\delta$ where $-V_n = Fl_{n-1}$, using the expression indicated above for $l_{n-1}$, one finds that

$$
\frac{\partial h(x,t)}{\partial t} \approx a F + \frac{a^2 F h_{xs}(x,t)}{2 h_s(x,t)^2}
$$

for EKR$\delta$.

(18)

Equation (18) agrees exactly with the result of the heuristic derivation described above based on identifying the relevant uphill current, and is consistent with Eq. (13). For EKR$0$, where instead $-V_n = F(l_{n-1} + l_n)/2$, the second-order terms cancel in the $\partial h/\partial t$ equation, consistent with vanishing of the net uphill current, and one obtains

$$
\frac{\partial h(x,t)}{\partial t} \approx a F + \frac{a^3 F h_{xss}(x,t)}{2 h_s(x,t)^4} - \frac{a^3 F h_{xxs}(x,t)}{6 h_s(x,t)^3}
$$

for EKR$0$.

(19)
Equation (19) is consistent with Eq. (14), but we note that the $O(a^7)$ terms do not have a conservation form. This reflects the feature that the boundary conditions will be important in enforcing mass conservation (see the Appendixes).

III. PRECISE CORNER-TO-CORNER GROWTH COALESCENCE OF ISLAND PAIRS

As indicated in Sec. II, there are many reasonable variations of models for growth coalescence with efficient PD. We will focus on the “simplest” case denoted EKR (efficient kink rounding) in Sec. II with no interlayer transport. However, we will also comment briefly on other cases. We consider a single “initial” island geometry: two square clusters each of size $L \times L$ atoms are arranged corner-to-corner in the center of a larger square lattice of $4L \times 4L$ sites. We perform simulations of deposition, diffusion, aggregation, and island growth and coalescence on the larger lattice for we impose periodic boundary conditions. Thus, the larger lattice represents the “capture zone” for a specific pair of islands in a full simulation of the formation of many islands during deposition.

A. EKR with no interlayer transport

First, in Fig. 3, we present kinetic Monte Carlo (KMC) simulation results showing two snapshots in the sequence of shapes during growth coalescence for both the EKR0 and EKR$\delta$ models. Both cases correspond to large islands with $L=100$, and the shapes are “geometric” with little rounding. Second, in Fig. 4, we show KMC results for the variation of the coalescence shape with increasing system size, $L$, for fixed additional coverage $\delta \theta = 0.1$ since the time of initial coalescence. It is clear that growth shapes are more rounded for smaller sizes, particularly for the EKR$\delta$ model. For comparison with these deposition models including diffusion-mediated aggregation, we also perform simulations of benchmark models where atoms are added to all periphery sites of the growing island at a uniform rate. KMC results in Fig. 5 for the EKR0 and EKR$\delta$ versions of the benchmark models for $L=100$ and $\delta \theta = 0.1$ should be compared with corresponding results from the realistic models in Fig. 3. It is clear that growth coalescence shapes for the benchmark models are more rounded, especially for EKR$\delta$.

Next we present results from the deterministic kink dynamics (DKD) treatments and compare these predictions against the results of atomistic simulations. We focus on the EKR$\delta$ model where the coalescence shapes are more rounded, and thus more interesting. However, we also present limited results for EKR0. Such DKD modeling ex-
hibits reflection symmetry about a line at 45° through the center of the neck of the coalescing pair of islands. Thus, for the evolution equations of Sec. II, we prescribe a boundary condition at $S$ along this symmetry line that determines the creation of new layers in the center of the neck—see Fig. 2. There is also a separate boundary condition required to specify the completion of layers at the sharp corner, $C$, where the neck region meets the straight outer edge, and to specify the motion of this corner—see again Fig. 2. The specific form of these boundary conditions is described in detail in Appendix B for the corner $C$, and in Appendix C for the symmetry point, $S$.

In Fig. 6, we compare DKD and atomistic simulation results for the EKR$\delta$ model including diffusion-mediated aggregation for different island sizes with $\delta \theta=0.1$. The DKD results correctly recover coalescence shapes and, specifically, the enhanced rounding for smaller sizes. We emphasize that these DKD analyses incorporate the nonuniform rate, $F(x)$, of aggregation of deposited atoms with the island perimeter, the specific form of which is described in Sec. IV. In Fig. 7, we compare DKD and atomistic simulation results for the benchmark version of the EKR$\delta$ model where atoms are added with equal probability at all island perimeter sites for different island sizes. Again we set $\delta \theta=0.1$. Here, the coalescence shapes are much more rounded, even for large sizes, and again the DKD results recover coalescence shapes and enhanced rounding for smaller sizes. For a more quantitative analysis, one can examine the evolution of the roughness, $W$, of the portion of the step edge between the corner, $C$, and symmetry, $S$, points, where $W$ is defined in Sec. II. Results in Fig. 8 compare DKD and atomistic simulation results for $W$ versus coverage increment, but just for the benchmark EKR0 and EKR$\delta$ models where roughening or rounding is greatest.

The agreement is generally good. The discrepancy for small sizes likely comes from ambiguity in determining $W$ for the atomistic simulations. In atomistic modeling, fluctuations lead to uncertainty in the appropriate assignment of the corner and “symmetry” points (noting that individual simulations do not display symmetry).

### B. Other models

Here, we first consider the behavior of EKR models with choices of Ehrlich-Schwoebel step-edge barriers that are different from the uniform infinite barrier of Sec. III. In Fig. 9, we show KMC simulation results for the behavior of both EKR0 and EKR$\delta$ models including diffusion-mediated aggregation for zero step-edge barrier, and for a nonuniform step-edge barrier (zero barrier on open or kinked step edges, and infinite barrier along close-packed step edges). These coalescence shapes are presented for $L=100$ and $\delta \theta=0.1$ ML.

Not surprisingly, coalescence shapes without an infinite step edge barrier are more rounded due to downward transport from atoms landing on top of the island, at least in the case of the nonuniform barrier where atoms deposited on top of the island tend to descend in the neck region. Note that downward
transport does not dramatically change the coalescence shapes in this regime. Finally, in Fig. 10, we show one example of KMC simulation results for the ECR model in which adatoms aggregating with the outer edges of the island round the global corners and accumulate in the neck region. As expected, this results in the neck region “growing out” rapidly.

IV. DISCUSSION

A principle observation from the above studies is that in the regime of large island size, growth coalescence shapes adopt a simple near-geometric form, i.e., overlapping squares with negligible rounding of the neck region. In fact, coalescence shapes are quite geometric even for moderate sizes. We now describe two factors that contribute to this behavior.

First, as indicated in Sec. II, and discussed further in the Appendixes, one can think of evolution of suitable portions of the neck region in terms of multilevel growth in a direction orthogonal to the original step edge. Then, the degree of rounding of the neck region is reflected by the degree of roughening in these multilevel growth models. Specifically, if $W$ denotes the roughness, one expects evolution of the form $W \sim a(Ft)^{\beta}$, where $t = 0$ denotes the “initial coalescence time” when square islands meet. The degree of rounding is naturally measured by the ratio $R = W/L$. Thus, if one considers behavior after deposition of a fixed coverage increment $\delta \theta$, where $Ft = \delta \theta L$, one concludes that $R = W/L \sim a(\delta \theta)^{\beta}L^{-(1-\beta)}$. Thus, in situations where this intrinsic scaling is realized with $\beta < 1$, such as for the simple geometry with fixed boundaries in Appendix A, it follows that $R \to 0$, as $L \to \infty$. For more realistic boundary conditions describing the moving corner $C$, one has $\beta = 1$ for the benchmark model (see the appendices), so another factor contributes to the observed geometric shapes.

This second factor, which impacts behavior for both the EKR$\delta$ and EKR0 models incorporating diffusion-mediated aggregation, is that the flux of attaching adatoms is highly nonuniform. Given the nature of diffusion-limited aggregation, one expects that this flux will be highest at the lower leftmost (or upper rightmost) global corner in Fig. 1, then diminish in the middle of this outer edge before increasing again to a high value at the global corner $C$ bordering the neck region in Fig. 2. Indeed, this behavior is observed, as shown in Fig. 11(a). Of more relevance in determining growth shapes is that this flux will vary from a high value at the corner, $C$, to a very low value at the neck, $S$, in Fig. 2. This variation along the growing “upper” step edge in Fig. 2, as determined directly from simulation, is shown explicitly in Fig. 11(b). This result was incorporated in the DKD analysis of Sec. III. More specifically, the simulation data were fitted by the smooth dashed curves shown, and that fit was used in the DKD analysis. It is clear that the greatly enhanced tendency to attach near the corner $C$ will facilitate the completion of lower terraces before the growth of upper terraces, thus tending to make growth smoother (i.e., a less rounded neck region).

Precise corner-to-corner coalescence of equal-sized islands considered in Sec. III is of course a special case of the general coalescence phenomenon. It is thus appropriate to consider more general cases and identify which features of our above analysis are generic, and which need modification. Specifically, we consider near-corner-to-corner coalescence and side-to-side coalescence of unequal size clusters, as illustrated in Fig. 12. The former is the most common situation experimentally, and in fact many examples with almost equal-sized cluster pairs are observed. The treatment of these general cases is analogous to that for precise corner-to-corner coalescence with equal-sized clusters in that the step edge is naturally divided into portions that are either straight outer edges or neck regions. The neck regions are further divided into portions bordered by corners, $C$, and by a “midpoint,” $M$, of the neck, defined to have a local orientation aligned at $45^\circ$ to a close-packed step edge. This midpoint, $M$, plays the role of the symmetry point, $S$, in Sec. 3. In this way, $x$ axes

FIG. 11. Variation along the island edge of the scaled aggregation flux for diffusion-mediated aggregation with an island pair. Flux along: (a) the outer edge; (b) the inner edge associated with the neck region. The island pair geometry is taken at the point of coalescence.
Since there is no symmetry, the ultimate of its motion follows in the regime of geometric growth velocities determine the direction of motion of \( M \). The local slope is zero for straight segments of unequal sized clusters. Results from KMC simulations for the EKR\( \delta \) model with \( \delta \theta = 0.1 \) ML for \( L_1 = 100 \) and \( L_2 = 50 \).

Can be chosen for each portion of the step edge so that the length of the straight portion from the ratio of the area of its subcapture zone to that in Sec. III and Appendix B. However, determination of neck regions of the shape observed in direct atomistic simulation is not geometric in the limit of large sizes, but rounded to an extent determined primarily by the step-edge energetics.

FIG. 12. Near-corner-to-corner coalescence (left) and side-to-side coalescence (right) for unequal sized clusters. Results from KMC simulations for the EKR\( \delta \) model with \( \delta \theta = 0.1 \) ML for \( L_1 = 100 \) and \( L_2 = 50 \).

Determinations and treatments of the corners, \( C \), is similar to that in Sec. III and Appendix B. However, determination of the location of \( M \) presents an additional complication since there is no symmetry (unlike for \( S \)). A reasonable estimate of its motion follows in the regime of geometric growth where the two portions of each neck region are also straight. By estimating the rate of growth of each straight portion from the ratio of the area of its subcapture zone to the length of the straight portion (from \( C \) to \( M \)). The relative growth velocities determine the direction of motion of \( M \) (which deviates from 45° from the close-packed directions for unequal growth velocities).

V. CONCLUSIONS AND GENERALIZATIONS

In conclusion, we have analyzed growth coalescence shapes during metal (100) homoepitaxy for the regime of efficient periphery diffusion (PD) and kink rounding. We have applied the kinetic Monte Carlo simulation of suitable atomistic models, a formulation incorporating deterministic kink dynamics (DKD), and a fully continuum formulation. Little attention has been paid previously to this regime where the DLA island shape instability is absent. Using either DKD or fully continuum formulations, we elucidate the simple geometric shapes observed in direct atomistic simulation studies. Our treatment presents not just the appropriate DKD or continuum evolution equations, but also emphasizes the treatment of boundary conditions at corners and at midpoints of neck regions of the coalescence shapes. More detailed discussion and analysis of boundary conditions is provided in the Appendices. We describe the treatment of corners, and symmetry or midpoints, in the DKD formalism. We derive basic constraints in the evolution of corners in the continuum formalism. However, the evolution of corners and midpoints is nontrivial, and it remains to develop a complete theory or prescription.

It is appropriate to comment on the more general situation for two-dimensional island growth during deposition where PD and kink rounding are not so efficient to allow all edge adatoms to reach kink sites rather than meeting to nucleate new layers. Then the DLA instability is not completely quenched. However, here we restrict our attention to the regime where it is sufficiently weak that individual island growth shapes are still fairly compact (rather than highly fractal or dendritic), but not equilibrated. This situation applies for metal (100) systems at sufficiently low temperatures, and metal (111) systems at higher temperatures. In this regime, one could adopt phenomenological continuum equations appropriate this intrinsically far-from-equilibrium growth process. These equations must account for the effect of a kink-rounding barrier in producing destabilizing mass currents, as well as for various other features such as nucleation-generated Mullins-type terms and symmetry-breaking terms.

Finally, we wish to emphasize that neither in the extreme regime of efficient PD and kink rounding, nor in the regime where these processes are somewhat inhibited, can one automatically adopt a Mullins-type continuum description of step-edge evolution as is often assumed. Generally, nonequilibrium mass currents will dominate the Mullins-type current associated with unrestricted PD, including kink escape. One can either attempt to obtain the appropriate equations directly from the atomistic models usually via heuristic arguments, or from simple kink dynamics models, as in Sec. III. An alternative to the latter is to consider more complete models that simultaneously treat the density of diffusing periphery atoms appropriately coupled to the kink dynamics.

Given the above criticism of the application of Mullins-type equations for nonequilibrium growth processes, we should emphasize that for treatment of postdeposition shape relaxation, an anisotropic Mullins approach is effective, provided island sizes are not too small. In particular, this is the case for postdeposition sintering of near-square islands that collide corner-to-corner. In this case the evolving shape is not geometric in the limit of large sizes, but rounded to an extent determined primarily by the step-edge energetics.

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APPENDIX A: BOUNDARY CONDITIONS FOR STEP-EDGE EVOLUTION—SIMPLE GEOMETRY WITH FIXED BOUNDARIES (0<\( x < L_a \))

Rather than immediately dealing with the complicated geometry of corner-to-corner coalescence of islands, it is instructive to first consider a simplified model for step-edge evolution: deposition at rate \( F(x) \) on a terrace of fixed width, 0 <\( x < L_0 \), where the end at \( x = 0 \) is “free,” and the other end at \( x = L_0 \) is bordered by an infinite vertical “frozen” step edge. See Fig. 13 where the height of the step edge at \( x = 0 \) is denoted by \( h^0 \). Below, for simplicity, we just consider the case of constant \( F(x) = F_0 \), although some basic features of model behavior are more general. Also \( t = 0 \) denotes the time when growth starts from the initial configuration.
FIG. 13. Schematic of the step edge profile for the simple geometry of Appendix A with fixed boundaries in the x-direction.

1. EKR$\delta$ model (kink-rounding barrier)

The EKR$\delta$ model is particularly simple in this geometry. Since there is no interlevel transport (every deposited atom attaches to the kink in the right side in the same layer), it is immediately clear that for constant $F$ one has a Poisson distribution of terrace heights. Thus, in the atomistic formulation, the mean value of the width of the $n$th terrace of height $n\alpha$ satisfies

$$
\langle \lambda_n \rangle = L_0 (Ft)^n \exp \left[ -Ft / n! \right] \approx L_0 (2\pi Ft)^{-1/2} \times \exp \left[ -\left( n - Ft \right)^2 / (2Ft) \right], \quad \text{for large } Ft. \quad (A1)
$$

This result does not provide information on fluctuations about the average profile. For example, what is the typical value of $h^0$, $\langle h^0 \rangle$, versus $t$ for the height $h^0 = a n^0$ shown in Fig. 13? The result depends on $L_0$ since obviously $\langle n^0 \rangle = F t$ for $L_0 = 1$, and $\langle n^0 \rangle$ drops progressively below $Ft$ for increasing $L_0$. For large $L_0$ and large $F t$, a simple estimate would come from the condition that

$$
\sum_{n=0}^{n^0} \langle \lambda_n \rangle = a, \quad \text{so } \text{erfc} \left( \frac{(Ft - \langle n^0 \rangle)}{(2Ft)^{1/2}} \right) \approx 2aL_0. \quad (A2)
$$

In the DKD formulation of this model, the rate equations (4) for the (deterministic) terrace widths become

$$
\frac{dl_0}{dt} = -FL_0, \quad \frac{dl_n}{dt} = F(l_{n-1} - l_n)(n > 0) \quad \text{for EKR}\delta.
$$

Using a continuum formulation (7) for EKR$\delta$ for the terrace width distribution, $l(h,t)$, for constant $F$, one finds scaling forms for the solutions as $t \to \infty$. In terms of $\delta h = h - aFt$, one has

$$
l(h,t) \sim L_0 (Ft)^{-\beta} K((Ft)^{-\beta} \delta h / a),
$$

$$
K'''(y) + y K'(y) + K(y) = 0 \quad \text{for EKR}\delta \quad (A4)
$$

where $\beta = 1/2$.

One finds that $K(y) \approx \exp(-y^2/2)$, and $W \sim a(Ft)^{1/2}$ for EKR$\delta$. From this result, one can also show that $h(x,t) \sim aFt + a(2Ft)^{1/2} \text{erf}^{-1}(2x/L_0 - 1)$. The agreement between these continuum predictions and simulations of the atomistic model is demonstrated in Ref. 20. As an aside, “natural” boundary conditions [where $l(h,t) \to 0$, as $h \to \infty$, or $K(y) \to 0$, as $y \to \infty$] at $x = 0$ and $x = L_0$ is satisfied automatically for long times, and thus the uphill current $J$ also vanishes at both boundaries.

2. EKR0 model (no kink-rounding barrier)

The EKR0 model in this simple geometry is both non-trivial and instructive. In the DKD formulation of this model, the lowest (leftmost) terrace does repeatedly shrink to zero width (in finite time), as in the atomistic model (so $h^0 > 0$ in Fig. 13). Define $t_m$ as the time when the leftmost terrace of width $l_m$ and height $ma$ shrinks to zero. Thus, $h^0$ changes from $ma$ to $(m+1)a$ as $t$ increases above $t_m$. Then, for $t_m < t < t'_m$ (where $h^0 = ma$), the rate equations (4) become

$$
l_n = 0 \quad \text{for } n < m, \quad \frac{dl_n}{dt} = -F l_n - \frac{F l_{n+1}}{2}, \quad \text{for EKR0}. \quad (A5)
$$

Our numerical solution of these equations reveals that $W \sim a(Ft)^{1/3}$, and the terrace width distribution displays a scaling form for $t \to \infty$ consistent with the continuum formulation discussed next.

Using a continuum formulation (8) for EKR0 for $l(h,t)$, for constant $F$, and setting $\delta h = h - aFt$, one finds $t \to \infty$ scaling forms

$$
l(h,t) \sim L_0 (Ft)^{-\beta} K((Ft)^{-\beta} \delta h / a),
$$

$$
K'''(y) + 2yK'(y) - 2K(y) = 0 \quad \text{for EKR0} \quad (A6)
$$

where $\beta = 1/3$. Our normalization is consistent with the condition $\int_0^{h^0} \delta h l(h,t) = aL_0$ (see below). This scaling relation also shows that $l(h = h_0, t) \sim a(Ft)^{-1/3}$, so the height profile, $h(x,t)$, becomes steep at the corner $C$. The solutions of Eq. (A6) for $K(y)$ for EKR0 are Airy-function-like, but imposition of appropriate boundary conditions excludes oscillations and negative values. Figure 14 demonstrates the consistency of results from DKD simulations and from this scaling theory (A6), using boundary conditions for the latter as described in detail below.
Finally, we discuss in more detail the boundary conditions in the continuum formulation for EKR0. The "natural" boundary condition at \( x = L_0 \) is satisfied automatically [see Eq. (A1)]. Thus, we focus on the boundary condition at the "free" end \( x = 0 \), which can be elucidated through a moment analysis of the basic evolution equation (8) for \( l(h,t) \). Below, we set \( h^0 = h(x=0,t) \), \( l^0 = l(h = h^0,t) \), etc., and \( K_0 = K(h = h^0) \). Note that since \( l(h,t) = (\alpha\partial/\partial h)x(h,t) \), one has

\[
\int_{h^0}^{\infty} dh l(h,t) = a \int_{0}^{L_0} dx = aL_0,
\]

\[
\int_{h^0}^{\infty} dh h l(h,t) = a \int_{0}^{L_0} h dx = a^2 F t L_0.
\]  

The second condition just says the area of deposited material up to time \( t \) is \( a F t L_0 \). Differentiating these relations with respect to \( t \) and rearranging yields

\[
\int_{h^0}^{\infty} dh \frac{\partial l(h,t)}{\partial t} = h^0 \frac{\partial}{\partial t} h^0,
\]

\[
\int_{h^0}^{\infty} dh h \frac{\partial l(h,t)}{\partial t} = h^0 h^0 \frac{\partial}{\partial t} h^0 + a^2 F L_0.
\]  

The integrals in Eq. (A8) can also be evaluated from Eq. (7) using integration by parts. Analysis of the first integral yields the relation

\[
h^0_t - F a = a^3 F \left( \frac{1}{l^0_h} \right) \frac{\partial}{\partial t} \left( F t \right)^{-2/3} a F K_0'' \frac{6}{K_0}.\]

The second integral in Eq. (A8) yields again the expression (A9) for \( h^0_t - F a \), but with an additional term, \( - F a^3 l^0_{hh} (h^0 F t) \). Thus, for consistency, one must set \( l^0_{hh} = 0 \) and \( K_0'' = 0 \). It is also clear that \( K_0'' < 0 \), since \( h^0 < a F t \). The result (A9) is consistent with Eq. (A6), which implies that \( h(x,t) = F a t \sim a(F t)^{1/3} \), and thus that \( h^0 - F a t \sim a(F t)^{1/3} \).

APPENDIX B: BOUNDARY CONDITIONS FOR STEP-EDGE EVOLUTION—MOVING OUTER STEP EDGE IN A SIMPLE GEOMETRY

In the geometry for corner-to-corner coalescence of islands shown in Fig. 1 or 2, the straight left edge of the islands (and thus the corner \( C \)) moves leftward at roughly constant velocity. In benchmark models with constant attachment rate per site, \( F \), it is easy to see that a new layer on this left edge is completed due to aggregation of adatoms every \( 1/F \) time units. We wish to first test the effect of this moving edge on the shape of the upper step edge without introducing the full complexity of corner-to-corner coalescence geometry (in particular, treatment of the neck \( S \)). To this end, we consider a simpler problem considering deposition at rate \( F \) on a terrace of initial width \( L_0 \), where the end at \( x = L_0 \) is bordered by an infinite vertical "frozen" step edge (as in Appendix A), but now the end at \( x = 0 \) is moving left at rate \( F a t \) (see Fig. 15 where the height of the step edge at \( x = 0 \) is again denoted by \( h^0 \)). We believe that step-edge behavior in this simpler model near the corner \( C \) will carry over to the more complex corner-to-corner coalescence geometry.

1. **EKR\( \delta \) model (kink rounding barrier)**

The EKR\( \delta \) model is no longer simple due to the moving boundary. Detailed behavior at the corner \( C \) in Fig. 15 is determined by a the following competition: (i) completion of the left edge at rate \( F \), causing \( C \) to move left, and the leftmost terrace on the upper step edge to grow; and (ii) shrinkage and completion of this leftmost terrace on the upper step edge by incorporation of atoms depositing at rate \( F \), causing \( C \) to move up. Simulations show that as a consequence of the fluctuation-dominated competition, \( C \) moves up at a rate \( 0.22 a F t \) and left at rate \( a F t \) (i.e., completion of the leftmost terrace on the upper edge is less efficient than completion of the left edge, as should be expected).

In the DKD formulation of this model, the leftmost terrace of width \( l_m \) and height \( h^0 = ma \), shrinks to zero at finite time \( t_m \). A convenient formulation of evolution for \( t_m - t < t_m \) is obtained by
Here, we have added a gain term, \( +Fa \), to the \( l_m \) equation to reflect the moving left edge, and \( t_m \) is determined by when \( l_m \) decreases to \( a(1+\epsilon) \), for suitably chosen \( \epsilon \), which recovers \( h_0^t = 0.22Fa \). At this time, \( l_m \) is set to zero, and the current value of \( l_{m+1} \) is increased by \( +a \). Results for the terrace width distribution and the step height profile are shown in Fig. 16. The feature that \( h^0 \) is only a fraction of the mean step height, \( h_{av} \sim aFt \), means that \( W \sim aFt^{1/2} \) (cf. Appendix A), i.e., the effect of the boundary condition dominates the intrinsic scaling of \( W \).

Next we consider the continuum formulation for EKR\( \delta \) for constant \( F \). We note that solutions to the PDE (7) for \( l(h,t) \) do exist with the scaling form \( l(h,t) \sim L_0(Ft)^{1-\beta}K((Ft)^{-\beta}dh/a) \), where \( \beta = 1/2 \), consistent with \( f_{h}^{\infty}dhl(h,t) \sim aFt \) (see below). However, these solutions require that \( h^0 - Fat - a(Ft)^{1/2} \), which is not satisfied here (see above). Thus, scaling solutions do not apply here, but instead, one can directly integrate the PDE (7) after transforming to a reference frame moving with \( C \), and prescribing a suitable constant “terminal” terrace width \( l^0 = l(h=h_0,t)(>a) \). Note that this behavior is quite distinct from that for EKR\( \delta \) in Appendix A1 where \( l^0 \to 0 \), as \( t \to \infty \). Results are consistent with those from the DKD treatment in Fig. 16.

Finally, we discuss in more detail the boundary conditions in this continuum formulation for EKR\( \delta \). The condition at \( x=L_0 \) is as in Appendix A. The condition at the “free” end \( x=0 \) can be elucidated through a moment analysis of Eq. (7) for \( l(h,t) \). Since \( l(h,t) = (a\partial l/\partial h)x(h,t) \), one has

\[
\int_{h_0}^{x} dh l(h,t) = a \int_{-aFt}^{t_0} dx = aL_0 + aFt,
\]

\[
\int_{h_0}^{x} dh h l(h,t) = a \int_{-aFt}^{t_0} dx = a^2FtL_0 + \frac{a(aFt)^2}{2} + a^2F \int_{0}^{t} dt' h^0(t'). \tag{B2}
\]

The second expression accounts for the amount of material deposited on the upper step edge, in addition to that accumulated for \( h>0 \) from aggregation with the left step edge. Using the first expression to determine \( f_{h}^{\infty}dh(\partial l/\partial t)l(h,t) \) and comparing with the result obtained from integrating Eq. (7) yields the relation

\[
h^0 = aF(1-\alpha) - (a^2F/2)(l_0^0/l_0) \sim aF(1-\alpha), \tag{B3}
\]

since \( l_0^0 \to 0 \) from the numerical analysis. Since also \( h^0 = 0.22Fa \), one concludes that \( l_0^0 \sim 1.3a \), consistent with the results of Fig. 16. Finally, a similar analysis of the second expression yields the same expression for \( h^0 \) except for an additional term \( a^2F/(2h^0) \), which vanishes as \( t \to \infty \). Since the approximate evolution equation (7) is only second order in \( h \), one cannot exactly satisfy both conditions above. However, the solution of Eq. (7) still describes model behavior well.

2. EKR\( 0 \) model (no kink rounding barrier)

The EKR\( 0 \) model is again nontrivial. In the DKD formulation of this model, the leftmost terrace of the upper step edge again repeatedly shrinks to zero width (in finite time). To appropriately describe this behavior, one can simply adopt the rate equations from Appendix A for EKR\( 0 \), except that one adds a gain term \( +Fa \) to the \( dl_m/dt \) equation reflecting movement left at rate \( aF \) of the left step edge. Our numerical solution of these equations (not shown) indicates that \( l^0 \) initially decreases, achieving a plateau value of \( l^0 \approx 43a \), for \( Ft > 150 \), where \( h^0 - aFt \to 0.02aFt \). Initially, one has \( W \sim a(Ft)^{\beta} \) with \( \beta = 0.4 \), but eventually a linear increase must occur. Note that this behavior is quite distinct from that for EKR\( 0 \) in Appendix A2 where \( l^0 \to 0 \) as \( t \to \infty \).

Next we consider the continuum formulation for EKR\( 0 \) for constant \( F \). Solutions to the PDE (8) for \( l(h,t) \) exist with the scaling form \( l(h,t) \sim L_0(Ft)^{1-\beta}K((Ft)^{-\beta}dh/a) \), where \( \beta = 1/3 \), consistent with \( f_{h}^{\infty}dhl(h,t) \sim aFt \) [see Eq. (B2)]. However, these solutions require that \( h^0 - Fat \sim a(Ft)^{1/3} \), which is not satisfied here (see above). Thus, scaling solutions do not apply here, although they do provide a reasonable description of initial behavior.

Finally, we discuss in more detail the boundary conditions for EKR\( 0 \). The boundary condition at \( x=L_0 \) is natural, and that at \( x=0 \) can be elucidated through a moment analysis of Eq. (8) for \( l(h,t) \). The results (B2) still hold here. Using the first expression to determine \( f_{h}^{\infty}dhl \), and comparing with the result obtained from integrating Eq. (8) yields the relation

\[
\int_{h_0}^{x} dh l(h,t) = a \int_{-aFt}^{t_0} dx = aL_0 + aFt,
\]

\[
\int_{h_0}^{x} dh h l(h,t) = a \int_{-aFt}^{t_0} dx = a^2FtL_0 + \frac{a(aFt)^2}{2} + a^2F \int_{0}^{t} dt' h^0(t'). \tag{B2}
\]
\[ h_t^0 - aF = - \frac{a^2 F}{l_0} + \frac{a^3 F l_0^0}{6} \frac{h}{l_0}. \]  \hspace{1cm} (B4)

Analysis of the second integral yields Eq. (B4) with an additional term, \(-a^3 F l_0^0 (6h)^0 \), on the right-hand side. Thus, for consistency, one must set \( l_0^0 = 0 \). Assuming the last term in Eq. (B4) does not dominate, this relation is consistent with the behavior of \( l_0 \) and \( h_0 \) obtained from the DKD formulation.

**APPENDIX C: BOUNDARY CONDITIONS FOR STEP-EDGE EVOLUTION—NECK REGION FOR CORNER-TO-CORNER COALESCENCE**

In atomistic modeling for the precise corner-to-corner coalescence problem with equal-sized islands, the mean coalescence shape will of course display reflection symmetry about a line at 45° through the neck. Of course, individual simulations will display fluctuations or deviations from this symmetric shape. In modeling using deterministic kinetics dynamics (DKD), we build in this symmetry condition by hand, and thus consider only a portion of the step edge on one side of the symmetry line shown in Fig. 17 (or Fig. 2). This requires imposing a suitable boundary condition at the symmetry line, as we now describe. We describe only the case of constant \( F \).

The key requirement is the appropriate prescription of the creation of a new terrace of height \( h = (n + 1)a \) and width \( l_{n+1} \) at the symmetry line in the middle of the neck above the existing highest terrace in Fig. 17(i). For the EKR0 model, we write

\[
\frac{dl_n}{dt} = F(l_{n-1} - l_{n+1})/2 \text{ for } m = n - 1, n - 2, \ldots,
\]

\[
\frac{dl_{n+1}}{dt} = F(l_n + l_{n+1})/2, \quad a \frac{dm_n}{dt} = F l_n/2. \quad (C1)
\]

Here \( m_n \) denotes the mass (in atoms) deposited on the \( m \)th terrace that accumulates at the kink on the symmetry line leading to formation of a new higher-level terrace. When \( m_n \) reaches a value of unity, \( m_{n+1} \) a new terrace is created with \( l_{n+1} \geq 0 \) as shown in Fig. 17. Specifically, at this time, we do the following: (a) reduce the current value of \( l_n \) by \( a \), and change its evolution equation to \( \frac{dl_n}{dt} = F(l_n - l_{n+1}) \). (b) introduce \( l_{n+1} \) with initial value zero satisfying \( \frac{dl_{n+1}}{dt} = F l_n/2 \).

In closing, we note that the DKD formulation developed here can be extended to treat creation of new layers at the midpoint, \( M \), of nonsymmetric neck regions. Here one must simultaneously integrate equations for the terrace widths for portions of the step edge on both sides of \( M \). One must now also prescribe the direction of motion of \( M \). This is done so as to maintain equal terrace widths on both sides. Finally, we note that in the continuum formalism, one has \( \partial h/\partial x = 1 \) at \( M \), but further analysis is needed to determine the trajectory of \( M \).


19 The rates for aggregation with each of the edges can be quantified in terms of a partition of the capture zone for each island into four subcomponents. See M. C. Bartelt, C. R. Stoldt, C. J. Jenks, P. A. Thiel, and J. W. Evans, Phys. Rev. B 59, 3125 (1999).

20 The range of (small) values of the kink rounding barrier consistent with this behavior can be determined from the formulations in Refs. 6, and 7.


