

Atomic size effects in continuum modeling

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Continuum modeling of many physical systems typically assumes that the spatial extent of an atom is small compared to the quantities of interest and can therefore be neglected. We show that this is valid only asymptotically. For many applications of practical interest, the spatial extent of a discrete atom cannot be neglected. We have developed a model for the description of epitaxial growth based on the levelset method, and find that we can accurately predict quantities such as the island densities, if we implement boundary conditions in a region with atomic width, rather than just on a line without any spatial extent. Only in the limit of very large islands and island spacings can this be neglected.

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Epitaxial growth and many other physical phenomena in materials sciences occur on time and length scales that span many orders of magnitude. For example, typical feature sizes for many opto-electronic devices might be up to micrometers in size, and the typical growth time for such devices is minutes up to hours. On the other hand, the physics that ultimately control the morphology of an epitaxially grown device occur on the atomic level, i.e., on length scales of the order of Å, and time scales that reflect the typical atomic vibration frequencies of approximately 10^{-13} s.

An ideal model to describe epitaxial growth would seamlessly link all the relevant time and length scales. However, such a model does not currently exist. Atomistic models of various degrees of sophistication resolve the atomic length and time scales. They are therefore very valuable in understanding the microscopic mechanisms during epitaxial growth. However, such models are computationally not efficient enough to also describe the more mesoscopic and macroscopic scale. It is therefore an important task to link the relevant information from atomistic models to more macroscopic, continuum-type models, so that the strengths of each approach is incorporated in a hybrid model.

Continuum models are typically valid only in an asymptotic regime, so that they do not resolve the atomic scale [1,2]. The size of an atom is typically assumed to be negligible, since it is small in size compared to the continuum scale. For example, boundary conditions are enforced on a line, even though in reality the adatom concentration has to be realized in an area that at a minimum reflects the atomic lattice constant. This is a good assumption if the typical sizes of interest in the model are many orders of magnitude larger than the atomic lattice constant. Often, however, we are interested in mesoscopic length scales where the size of the large objects such as islands or clusters of atoms is only 1–2 (and not many) orders of magnitude larger than the atomic size.

In this Rapid Communication, we show that the discrete size of atoms cannot be neglected in continuum-type models of epitaxial growth under typical growth conditions. We describe a formalism that allows us to include the spatial extent

of atoms through a boundary condition that is enforced in a boundary region of width a , which is the lattice constant or lateral size of an atom.

Our model is based on the levelset method [3,4], which is a general technique for simulating the motion of moving boundaries. Its applicability to the description of epitaxial growth has been shown elsewhere [5–7]. Numerical details of the method are described in Ref. [8]. The main component of our model is that a (zero thickness) boundary curve, such as the boundary of an island, can be represented by the set $\varphi=0$, called the *levelset*, of a smooth function φ , called the *levelset function*. For a given boundary, this function evolves according to

$$\frac{\partial \varphi}{\partial t} + v_n |\nabla \varphi| = 0, \quad (1)$$

where v_n is the velocity of the moving boundary in the outward normal direction. This velocity contains all the physical information of the simulated system, and is computed by solving the diffusion equation for the adatom concentration $\rho(\mathbf{x}, t)$

$$\frac{\partial \rho}{\partial t} = F + D \nabla^2 \rho - 2 \frac{dN}{dt}, \quad (2)$$

where F is the deposition flux, D is the surface diffusion constant, and the last term on the right hand side is the rate of nucleation of new islands on the surface. The velocity of the island boundaries is determined by the flux of adatoms into the island boundary, and is given by

$$v_n = a^2 D (\mathbf{n} \cdot \nabla \rho^- - \mathbf{n} \cdot \nabla \rho^+). \quad (3)$$

The superscripts (+) and (–) label the contributions from above and below the island boundary.

For the case of irreversible aggregation, a dimer (consisting of two atoms) is the smallest stable island, and the nucleation rate is

$$\frac{dN}{dt} = D \sigma_1 \langle \rho^2 \rangle, \quad (4)$$

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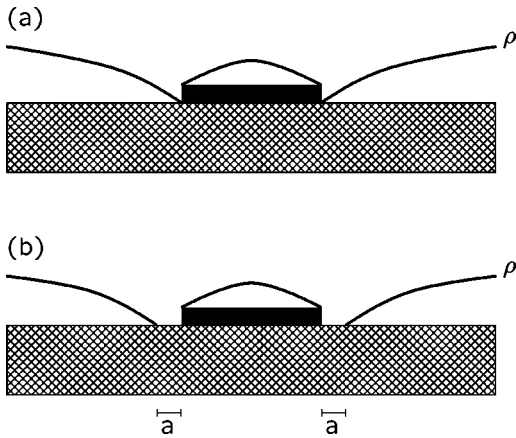


FIG. 1. Schematic representation of the different boundary conditions and the corresponding adatom concentrations as obtained from the solution of Eq. (2): (a) The original boundary condition that neglects the spatial extend of the atomic lattice constant; and (b) the proposed implementation of the boundary condition that reflects a region of atomic width a along the island boundaries with $\rho=0$.

where $\langle \cdot \rangle$ denotes the spatial average of $\rho(\mathbf{x}, t)^2$, and σ_1 is the adatom capture number [9]. While the time of nucleation is chosen deterministically in our model according to Eq. (4), the position of a new nucleus is chosen with a probability that is weighted with the local value of $\rho(\mathbf{x}, t)^2$, as determined by Eq. (2). The importance of these spatial fluctuations has been discussed in Ref. [7].

In order to complete the model, a boundary condition for ρ needs to be specified. For the case of irreversible aggregation, every atom that reaches an island boundary remains at the island boundary. Thus, a physical meaningful boundary condition is $\rho=0$ at the island boundary. Such a boundary condition was implemented for the results presented in Ref. [7]. This boundary condition and the corresponding solution of the diffusion equation are schematically shown in Fig. 1(a).

However, such a boundary condition implies that the adatom concentration is nonzero everywhere except exactly on the boundary; in particular, in a region that is less than a lattice constant a away from the boundary (cf. Fig. 1). There is a fundamental difficulty associated with this: due to the discrete lattice structure of the substrate, there can never be a free adatom within one lattice spacing of the island boundaries. Thus, the boundary condition $\rho=0$ precisely at the island boundary is not completely accurate.

The error associated with this is small in a true continuum problem (where the island separation is very large compared to a lattice constant). However, for typical conditions during epitaxial growth, the island spacing is only 1–2 orders of magnitude larger than the lattice constant. For example, for $D/F=10^6$, the saturated island density for irreversible aggregation is approximately 0.003, which translates into an (average) distance between island centers of approximately 18 lattice constants. Thus, the boundary region of interest where ρ should be zero is not negligible compared to the inter-island distances.

In this Rapid Communication we show how the boundary condition can be improved, and how it affects the results obtained from our island dynamics levelset method. In our model, we assume a boundary condition where ρ is set equal to zero in a region around an island boundary that has width a . This idea and the corresponding adatom concentration are shown in Fig. 1(b). This is a particularly good model for a case with fast edge diffusion, where the island boundaries are straight. For a system with large atomic roughness along the island edges one might have to choose a region with a width that is larger than a . We also note that an alternative approach would be the implementation of a higher order boundary condition along the island edges, i.e., $\rho - a\mathbf{n} \cdot \nabla \rho^- = 0$ on the terrace side and $\rho=0$ on top of the island. However, we found that such an approach is numerically less accurate.

We average the velocities along each island boundary, which guarantees the compactness of the islands. Thus, the velocity v_n is now computed as

$$v_n = \frac{Da^2}{\Gamma_1} \left[- \int_{\Gamma_1} \mathbf{n} \cdot \nabla \rho^+ d\Gamma + \int_{\Gamma_2} \mathbf{n} \cdot \nabla \rho^- d\Gamma \right] + F \frac{\Delta A}{\Gamma_1}. \quad (5)$$

The integrals are along the island boundary of length Γ_1 , and the curve that is a distance a away from the boundary with length Γ_2 , and the additional term in Eq. (5) describes the direct impingement of atoms onto the boundary region of size ΔA .

The results are then compared to an atomistic kinetic Monte Carlo (KMC) simulation of a simple cubic, solid-on-solid model. Such a model includes the same physical processes as our levelset model. In particular, the two external physical parameters are also the diffusion constant D and the deposition flux F , and we choose identical values for the comparison of the two models. Very fast edge diffusion is included to ensure that the islands obtained from the simulations are also compact [10]. The details of the KMC simulations are described in Ref. [7].

The results obtained for the island densities from the levelset method with the original and improved boundary conditions and from the KMC simulations are shown in Fig. 2 for different values of D/F . The values of D/F shown here represent the range of these parameters under typical experimental conditions. All data shown was obtained on a lattice of lateral size 180 a , with a numerical resolution of 512 gridpoints. We tested carefully that all the results shown are converged with respect to system size and numerical resolution. The data represent the averages over 20–50 statistically independent simulations. The KMC data was obtained on (at least) ten independent lattices of lateral size 600 a for each value of D/F .

The island densities are decreased with the new boundary condition. The reason for this is the following: If ρ is set to zero in a boundary region (rather than just on a line), there is a certain fraction on the substrate where ρ is zero. This implies that $\langle \rho \rangle$ (the average adatom concentration) is reduced, which in turn reduces the nucleation rate. This can also be seen in Fig. 3, where the averaged adatom densities are

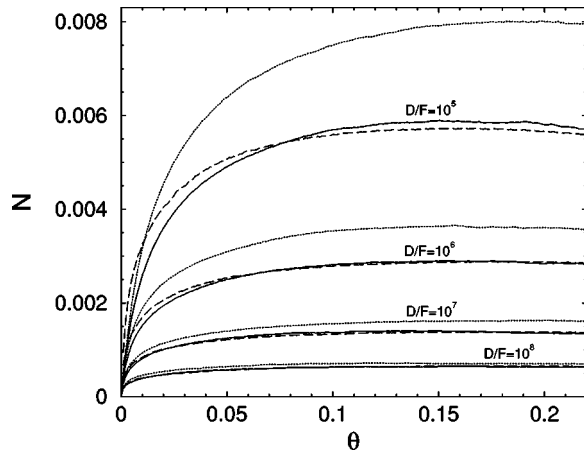


FIG. 2. Island densities as a function of coverage (in monolayers) obtained with the original boundary condition (dotted lines) and the improved boundary condition (solid lines) in comparison with results obtained from KMC simulations (dashed lines). Results are shown for different values of D/F .

shown. The difference in the island densities is largest for smaller value of D/F (approximately 25% for $D/F=10^5$), and gets progressively smaller for increasing values of D/F (less than 5% for $D/F=10^8$). We expect this discrepancy to vanish in the limit $D/F \rightarrow \infty$, since in that limit the atomic size can be neglected compared to the average island size.

The most striking result in Fig. 2 is, however, the extremely good quantitative agreement of the island densities obtained with the new boundary condition in comparison to those obtained from the atomistic KMC simulations. This illustrates that the proper treatment of the discrete size of an atom in a continuum-type description can significantly improve the qualitative and *quantitative* agreement of the quantities of interest in the physically relevant regime of D/F . For epitaxial growth under typical growth conditions, the discrete size of an atom cannot be neglected in a continuum-

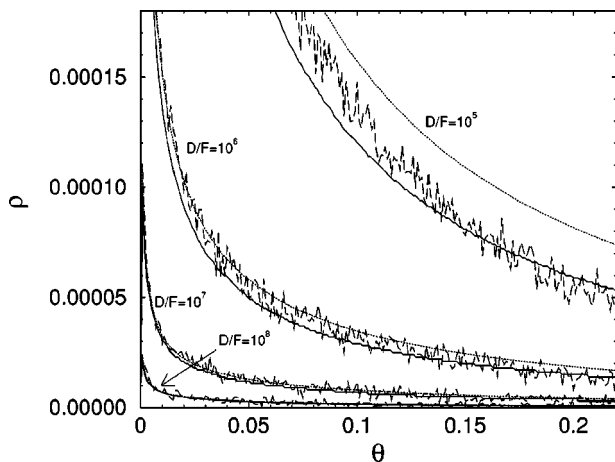


FIG. 3. Adatom densities as a function of coverage (in monolayers) obtained with the original boundary condition (dotted lines) and the improved boundary condition (solid lines) in comparison to results obtained from KMC simulations (dashed lines). Results are shown for different values of D/F .

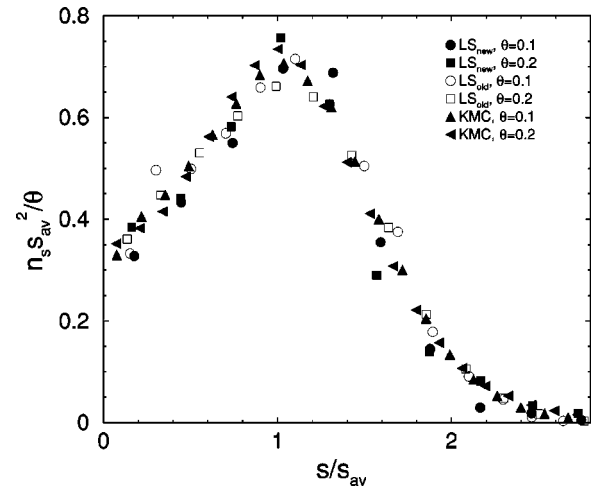


FIG. 4. Scaling of the island size distribution obtained with the original boundary condition (LS_{old}) and the improved boundary condition (LS_{new}) in comparison with results obtained from KMC simulations. n_s is the density of islands of size s , and s_{av} is the average island size. Results represent data for $D/F=10^6$ at different coverages Θ .

type description. In passing, we would like to mention that our results also recover the well known scaling result [11] $N \approx (D/F)^{-\chi}$, with the scaling exponent $\chi = 1/3$ for irreversible aggregation. In fact, on a log-log plot that spans several orders of magnitude, the difference between the two methods is rather small, and there is essentially no effect on χ .

There is a small discrepancy in the time evolution of the island density between the new levelset results and the KMC results. This discrepancy is more pronounced for smaller values of D/F , and vanishes as D/F increases. The reason for this discrepancy is not fully understood. We speculate, however, that it might be because of the following: In our model, islands are nucleated according to Eq. (4). This implies that islands are nucleated sequentially, and that the sequence of nucleation events is deterministic. In an atomistic simulation, the sequence of nucleation events is not the same. We believe that in particular for small values of D/F , temporal fluctuations in the nucleation might become relevant. Several islands might be nucleated (almost) simultaneously, leading to a faster approach of the island density to its saturation value. This becomes less relevant for larger values of D/F , where fewer islands are present on the surface, and where the length of the nucleation phase decreases. We also know from previous work [7] that what matters most is *where* islands are seeded, and not necessarily *when*. Almost all islands are nucleated in a very short initial phase, often called the nucleation phase [12], and grow in the so-called aggregation phase. This growth is mainly determined by the distribution of the capture areas [13–15], which are formed as a result of the spatial distribution of the nuclei.

We also checked that not only averaged quantities such as the island density exhibit the correct behavior, but that in fact we also properly describe the spatial correlations. We therefore examined the distribution of island sizes on the surface. It is well established that the island size distribution scales according to $n_s = \Theta / s_{av}^2 g(s/s_{av})$ [16], where n_s is the den-

sity of islands of size s , Θ is the coverage, s_{av} is the average island size, and $g(x)$ is a scaling function that has a unique form for the case of irreversible aggregation. In Fig. 4 the scaling of the entire island size distribution in comparison to results obtained with the original model and the atomistic KMC simulations are shown. The results obtained with the different boundary conditions are essentially indistinguishable, and both agree very well with the results obtained from the KMC simulations.

In conclusion, we have shown that we can include the discrete size of the atomic lattice constant in a continuum-type model that is based on partial differential equations. Our results show that for epitaxial growth, where the size of typi-

cal features of interest is 10–100 atomic lattice constants, the discrete extent of a boundary region cannot be neglected. Thus, any model that attempts to link models that are valid on different time and length scales needs to carefully evaluate the validity of approximations made, such as an approximation to neglect the size of an atom in comparison to the size of the larger features.

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