Growth and dynamical roughening of ideal quasicrystal facets

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(Received 6 November 1989)

Quasicrystal growth is studied in three dimensions via Monte Carlo simulation of a solid-on-solid quasiperiodic structure. Our results are consistent with an infinite roughening temperature, and in agreement with equilibrium studies. Surfaces plots and growth rates show that for any temperature T growth takes place via two-dimensional nucleation at sufficiently small chemical-potential driving forces ($\Delta\mu$). However, interfaces dynamically roughen for a large range of T at a nonzero value of $\Delta\mu$ that tends toward zero as T is increased. The average squared height of the growing surfaces is shown to diverge algebraically with the linear system size. Comparison is made with a similar crystal model.

I. INTRODUCTION

Recently, certain quasicrystals that are believed to be bulk equilibrium phases have been synthesized, revealing faceted shapes that beautifully exhibit their noncrystallographic symmetry.[1] To date, theoretical treatments of quasicrystal shapes have focused virtually exclusively on equilibrium properties. Here we present a dynamical study of quasicrystal growth. Specifically, within a particular model we study nucleated growth, which for increased driving force undergoes dynamical roughening. Recent experiments on Al-Mn alloys, discussed below, may have displayed such behavior.[2-4]

We note at the outset that our model is based on a perfect quasicrystal. Most quasicrystals in fact possess disorder[5] characterized by diffraction peaks having finite intrinsic widths characteristic of positional order correlation lengths of several hundred to a thousand angstroms. Notable exceptions appear to be Al-Cu-Fe (Ref. 6) and Al-Cu-Ru (Ref. 7) alloys, which have diffraction peak widths limited by experimental resolution. Strictly speaking then, our work models Al-Cu-Fe, Al-Cu-Ru, and possibly the new decagonal Al-Co-Cu (Ref. 3) classes of materials. The extent to which our conclusions apply to disordered quasicrystals is unclear[8] and is the subject of further investigation[9].

To briefly summarize previous work, we note that several authors have addressed the theoretical question of equilibrium shapes of quasicrystals in two and three dimensions at zero temperature[10-12] and, recently, at nonzero temperatures.[13-16] These studies show that long-range periodic positional order is not necessary for faceting and, in fact, suggests that interfaces in ideal three-dimensional quasiperiodic structures do not roughen at finite temperature. While equilibrium shapes are difficult to observe experimentally, they are important for understanding observed growth shapes. Since much recent experimental effort has focused on growing larger single grains, in particular for x-ray-diffraction studies, the growth behavior of interfaces in quasiperiodic systems has come to be of direct interest. Published theoretical work to date on quasicrystal growth has been limited to two studies.[17] The first showed that perfect Penrose tilings can be grown via strictly local attachment rules.[18] The second paper treats the possibility of spiral growth in quasicrystals[19]. Neither of these studies, however, has directly addressed growth dynamics.

Our dynamical studies consist of Monte Carlo (MC) growth simulations[20] for a simple quasiperiodic solid-on-solid (SOS) model in three dimensions. The results are independent of any surface metastability[16] questions, since the interface moves through a large number of environments in the quasiperiodic system (QPS). We find that, as in periodic structures, optimal (faceted at temperature $T = 0$) interfaces in quasiperiodic systems grow via two-dimensional nucleation at low $\Delta\mu$, and do undergo dynamical roughening at sufficiently large $\Delta\mu$. The temperature dependence of the onset of dynamical roughening is consistent with equilibrium theories[15,21] and simulations[16] that suggest an infinite roughening temperature for quasiperiodic systems.

II. DYNAMICAL ROUGHENING

It is well known that faceted interfaces in crystals can undergo a roughening transition as a function of temperature, in which the flat facets disappear, becoming curved and microscopically rough.[22] Facets can also roughen under growth conditions as a function of the chemical potential driving force ($\Delta\mu$) through a process called dynamical roughening.[23]

A faceted interface at T less than the roughening temperature T_R has a nonzero step free energy η. For such a system with a chemical potential at $\Delta\mu$ above coexistence, a nucleus of height h has a critical diameter d_c for growth given by[23]

$$d_c = \frac{c_2\eta}{c_1\Delta\mu\rho h}, \tag{1}$$

where ρ is the density, and c_1 and c_2 are shape-dependent

constants, equal to $\pi/4$ and $\pi/2$, respectively, for a cylindrical nucleus. For $\Delta\mu$ sufficiently small the in-plane correlation length $\xi_{\parallel}$ (Ref. 24) is less than d_c, and growth takes place by two-dimensional nucleation with a free-energy barrier $E_A = c_2\eta d_c$. The growth rate is exponential in $-E_A$ and therefore also in $-1/\Delta\mu$ from the Arrhenius equation.[23,25] For sufficiently large $\Delta\mu$, $d_c < \xi_{\parallel}$, $k_BT > E_A$, and thermal fluctuations provide nuclei of sufficient size such that the growth is no longer exponential but linear in $\Delta\mu$ (as for a rough surface at $T > T_R$). Let the $\Delta\mu$ for which growth changes from linear to exponential be $\Delta\mu_c$. Then from Eq. (1) for the critical diameter, we have the crossover driving force

$$\Delta\mu_c \sim \frac{\eta}{\xi_{\perp}\xi_{\parallel}} \sim \frac{\eta^2}{k_BT\xi_{\perp}}, \tag{2}$$

where we have taken the height h of the relevant steps[26] (which is just a lattice spacing for crystals) to be of the order of the perpendicular correlation length, $\xi_{\perp}$ (Refs. 16 and 27) for the quasiperiodic system. For a faceted interface, the correlation lengths are related by[28]

$$\xi_{\perp}^2 \sim \frac{T}{\sigma}\ln\xi_{\parallel}, \tag{3}$$

where σ is the microscopic surface stiffness. For a crystal, $\xi_{\parallel}$ diverges as $T \to T_R$ from below according to

$$\xi_{\parallel} \sim \exp\left[A/(T_R - T)^{1/2}\right], \tag{4}$$

where A is a constant.

III. SIMULATION RESULTS AND DISCUSSION

As in our recent paper[16] on the equilibrium properties of interfaces in quasiperiodic structures, we abandon the perfect Penrose tiling in favor of a simpler structure based on a tetragonal lattice. In analogy to the Penrose tiling, where tiles can be added to a surface within a lane (of quasiperiodic width[14]) at no additional cost in surface energy, we construct the "lattice" by quasiperiodically spacing layers normal to $\hat{\mathbf{z}}$ that cost no energy to cross (free layers), between layers that do cost energy to cross (bond cost layers). We define J as the nonzero bond energy. Any given layer in the lattice will quasiperiodically have zero or one additional layer through which an interface can wander at no additional energy cost [see Fig. 1(a)]. For comparison, simulations were performed for an entropic crystal model[29] (ECM) consisting of one free layer per every two bond cost layers [Fig. 1(b)].

Employing the SOS constraint disallowing vacancies in the bulk or overhangs on the surface, the interface is described by integer column heights $z(\mathbf{r})$. A single MC move consists of sequentially selecting a site on the surface, choosing to add or remove one particle, and calculating the change in energy, $\Omega \equiv \Delta E_b - \Delta\mu\ \Delta N$, for the proposed move. ΔE_b is the change in broken bond energy, and $\Delta N = \pm 1$ is the change in particle number. MC moves with Ω less than or equal to zero were automatically accepted, while moves that cost energy $\Omega > 0$ were accepted with probability $\exp(-\beta\Omega)$ by comparison with a random number.

Simulations were performed on surfaces of 45×45 sites using periodic boundary conditions. Surfaces of up to 141×141 sites were studied for finite-size effects. Surfaces were equilibrated for 2×10^4 MC moves per site at $\Delta\mu = 0$ before growing. The surface-averaged column heights, $\langle z \rangle_r$, were monitored as a function of time, t, measured in units of the number of MC passes through the lattice.

For the QPS, Fig. 2 shows average surface height as a function of the MC lattice passes (time) for a range of driving forces, $\Delta\mu$, at $k_BT/J = 0.6$. The existence of the plateaus at smaller $\Delta\mu$ suggests that new layers are formed via nucleation. Figure 3 shows the surface during the creation of the plateau near MC time 10 000 at $\Delta\mu/J = 0.085$ in Fig. 2. These surfaces clearly demon-

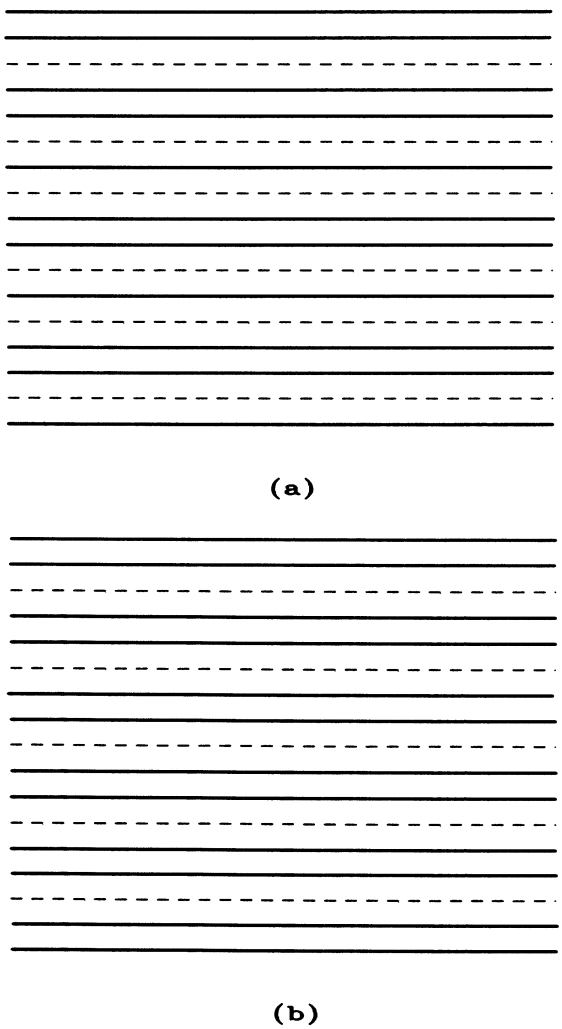

FIG. 1. Sample sequence of free layers (dashed lines) among bond cost layers (solid lines) for (a) the QPS, and for (b) the ECM.

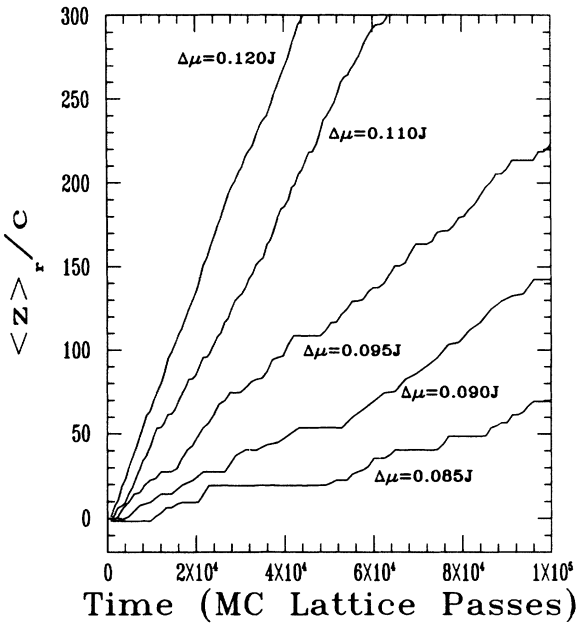

FIG. 2. The average surface height $\langle z \rangle_r / c$ as a function of time (MC passes through the lattice) at $k_B T/J = 0.6$ for several $\Delta\mu/J$. (c is a lattice constant.)

strate two-dimensional nucleation and subsequent growth of the new layer, which was completed by 11 000 MC lattice passes. Plateaus similar to those in Fig. 2 were observed at all temperatures up to $k_B T/J = 2.8$ for sufficiently small $\Delta\mu$ implying that equilibrium surfaces in the QPS are faceted at least up to these temperatures. The height difference between plateaus shows the step height at a given temperature and are in agreement with the relevant step heights investigated in Ref. 16.

Growth rates, $v(\Delta\mu)$, were measured as $\Delta\langle z \rangle_r / t$, where $\Delta\langle z \rangle_r$ is the change in the surface-averaged height during a time t. Figure 4 shows sample growth rates $v(\Delta\mu)$ for several temperatures for both the QPS and the ECM.[30] For the QPS, $v(\Delta\mu)$ is exponential at low $\Delta\mu$ at all temperatures, but becomes linear at sufficiently high $\Delta\mu$, indicating that the QPS dynamically roughens. We note, however, that at high temperatures only a very small $\Delta\mu$ is required to dynamically roughen the interface. Similarly, the ECM shows dynamical roughening at low T. On the other hand, for the ECM, we find $v(\Delta\mu)$ is always linear in $\Delta\mu$ for $k_B T/J \geq 1.2$, indicating a roughening transition between $k_B T/J = 1.0$ and 1.2, in agreement with the results of Ref. 16.

The crossover driving force, $\Delta\mu_c$, is graphed as a function of temperature for the QPS and the ECM in Fig. 5. The error bars indicate the width of the crossover of $\Delta\mu_c$ from an exponential variation in $-1/\Delta\mu$ to one linear in $\Delta\mu$. The deviation of the QPS curve from the ECM curve provides independent evidence consistent with an infinite roughening temperature for the QPS. For the ECM, $\Delta\mu_c$ goes to zero as T approaches T_R from below as[23]

$$\Delta\mu_c \sim \exp[-2A/(T_R - T)^{1/2}], \quad (5)$$

from (1) and (4). On the other hand, for the QPS, Fig. 5 suggests that $\Delta\mu_c \to 0$ only as $T \to \infty$, consistent with $T_R = \infty$. This might be expected from (2), since $\xi_{\|} \to \infty$ as $T \to \infty$, provided that η not diverge too fast. In fact,

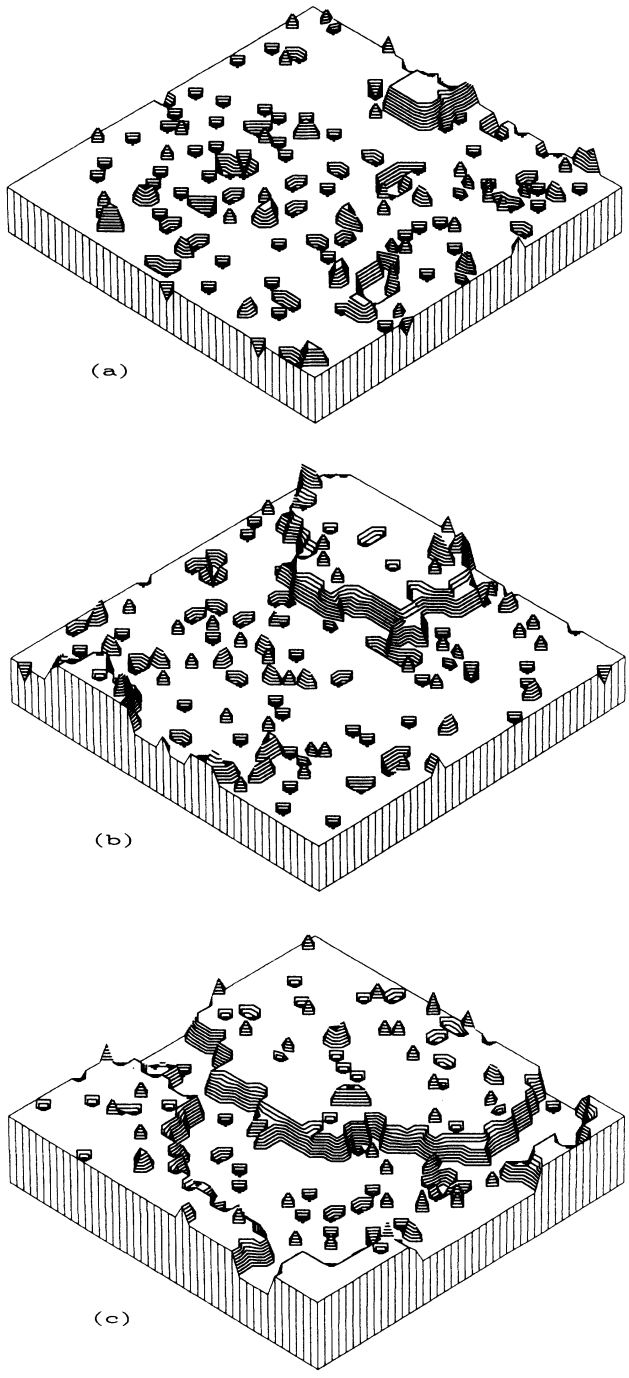

FIG. 3. Perspective view of 45 × 45 surfaces in the quasiperiodic structure at $k_B T/J = 0.6$ and $\Delta\mu/J = 0.085$ at (a) 9800, (b) 10 200, and (c) 10 600 MC lattice passes. Only fluctuations across bond cost layers are shown.

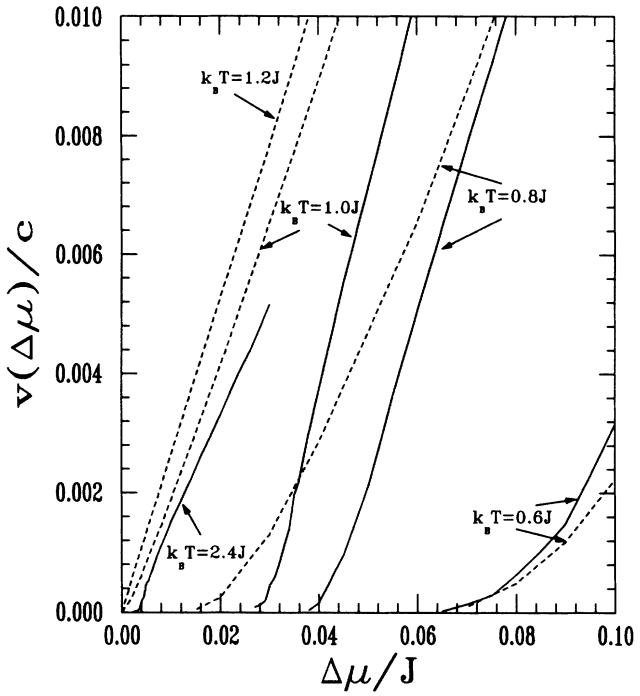

FIG. 4. Growth rates $v(\Delta\mu)/c$ for a range of temperatures. The dashed lines are for the ECM and the solid lines are for the QPS.

$\eta \rightarrow$ const as $T \rightarrow \infty$, since it is bounded from above by the step energy, and equilibrium studies in Refs. 16 and 31 show that step energies tend to a constant as $T \rightarrow \infty$. In addition, recent renormalization-group calculations by Toner[32] also show that $\eta \rightarrow$ const as $T \rightarrow \infty$. More precisely, after modifying the expression for the crossover driving force (2) (Ref. 26), Toner has predicted[32] that $\Delta\mu_c(T) \rightarrow 0$ as $T^{-3/2}$ for $T \rightarrow \infty$, consistent with our results at the higher temperatures,[32] and in contrast to Eq. (5).

The average (over the surface and over ensembles) squared surface height $\langle z^2 \rangle$ was measured as a function of the linear system size L for $k_BT/J = 1.0$ and $\Delta\mu/J = 0.03$. As shown in Fig. 6, over a range of L from 51 to 141 lattice sites, $\langle z^2 \rangle$ diverges almost linearly with L. This algebraic divergence, even under conditions of nucleated growth, is expected for both crystals and quasicrystals according to recent theories.[32,33] However, in our simulations of the ECM, $\langle z^2 \rangle$ diverged at most logarithmically with L, where the most extreme divergence was observed, as expected, under conditions of high temperature and large driving force. This difference between the QPS and the ECM is unexpected and is under further study.

Finally, we note that a columnar morphology has been observed in the decagonal phase of Al-Mn alloys, with 10 facets appearing parallel to the 10-fold axis.[2,3] In

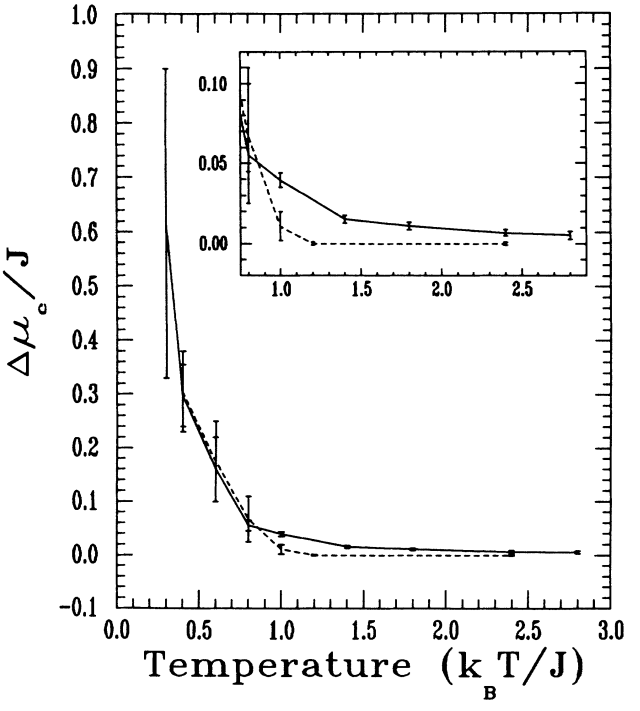

FIG. 5. $\Delta\mu_c/J$ as a function of temperature. The solid and dashed lines correspond to the QPS and ECM, respectively. Error bars indicate the width of the crossover region. The inset is a detail of the curves at the higher temperatures.

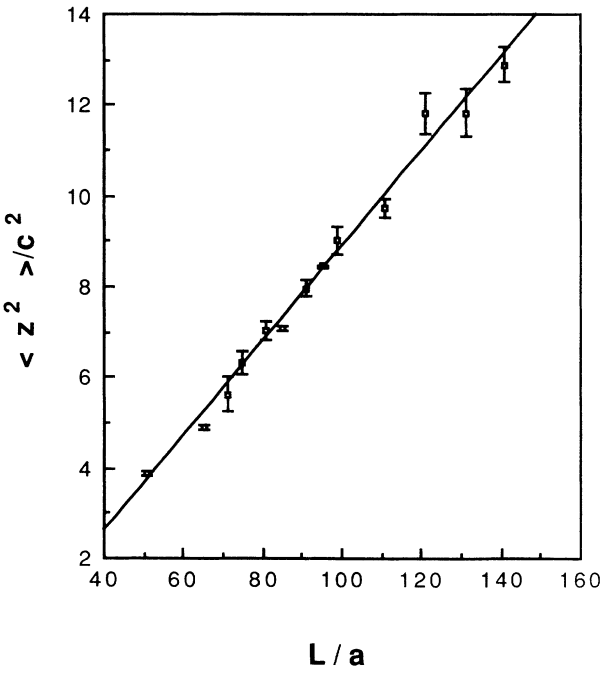

FIG. 6. Average squared surface height, $\langle z^2 \rangle / c^2$, as a function of the linear system size L/a. The error bars represent standard deviations over two to four independent simulations. The straight line is a least-squares fit to the data. (a and c are lattice constants.)

an ideal tiling model, these facets are not expected to roughen in equilibrium.[15] However, other morphologies have been observed with a facet normal to the 10-fold axis but curved around the axis, where 10 facets would be expected in equilibrium.[4] Our dynamical studies indicate that these observations are consistent with the ideal tiling model if dynamical roughening plays a role.

In conclusion, we have simulated the growth of an interface in a three-dimensional quasiperiodic system and find that at all finite temperatures growth proceeds via two-dimensional nucleation at sufficiently small chemical potential driving forces $\Delta\mu$. This result provides independent evidence consistent with an infinite T_R in quasiperiodic systems. Likewise, at all temperatures, the interface dynamically roughens at sufficiently large $\Delta\mu \equiv \Delta\mu_c$. Since $\Delta\mu_c \to 0$ as $T \to \infty$, dynamical behavior at high temperatures is indistinguishable from that of a rough crystal for most practical purposes. Dynamical roughening can resolve apparent inconsistencies between the observed morphologies of decagonal quasicrystals and predictions based on ideal tiling models. Finally, the average of the squared height of the growing surface was seen to diverge algebraically with the linear system size (L), in contrast to the logarithmic divergence observed for the crystal model simulated.

ACKNOWLEDGMENTS

We are very grateful to John Toner for numerous valuable discussions, for sending us some of his calculations, and for a copy of his work prior to publication. We have also benefited from his encouragement and interest in our work. Thanks also to David DiVincenzo for introducing one of us (J.A.J) to John Toner's work. We also thank David Huse for stimulating conversations. We gratefully acknowledge the Ohio Supercomputer Center for a generous grant of supercomputer time. One of us (J.A.J) acknowledges the support of The Ohio State University Graduate School. This work was supported in part by National Science Foundation Grant No. DMR-84-04961.

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[24] $\xi_\parallel$ is defined via the correlation function $G(r) = \langle [z(\mathbf{r}) - z(0)]^2 \rangle$, where $z(\mathbf{r})$ is the interface height at position $\mathbf{r} = (x, y)$. In the faceted (nonrough) phase, $G(r) = K \ln(r/a)$ for $r << \xi_\parallel$, where K is a temperature-dependent constant, and a is a lattice constant. For $r >> \xi_\parallel$, $G(r) = K \ln(\xi_\parallel/a)$. Physically, $\xi_\parallel$ is the lateral size of typical fluctuations in $h(\mathbf{r})$.

[25] J. Toner (Ref. 32) predicts the nucleated growth rate in quasiperiodic systems to be exponential in $-\Delta\mu^{-4/3}$. Our study is consistent with either result, and, primarily due to finite-size effects, no attempt is made here to distinguish the difference.

[26] J. Toner (Ref. 32) has rederived the steps leading to (2) taking the relevant nucleation step h to be a series of steps

of height $\xi_\perp$ (for small $\Delta\mu$), where $h \sim (\Delta\mu)^{-1/3}$ due to the quasiperiodicity of the system.

[27]The perpendicular correlation length, $\xi_\perp$, is a measure of the equilibrium interface width, and is defined as $\xi_\perp^2 = \lim_{r\to\infty}\langle[z(\mathbf{r}) - z(0)]^2\rangle$.

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[30]No significant size dependence was observed for the growth rates in the dynamically rough regime. On the other hand, in the slower growing nucleated regime, growth rates were up to a factor of 3 times larger on a 141×141 surface compared to the 45×45 surface at the lowest temperatures and driving forces, with agreement improving with increased driving force and temperature. However, since the growth rates are relatively small in the nucleated regime as compared to the dynamically rough regime, these size effects do not change our conclusions.

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