Growth and Kinetic Roughening of Quasicrystals and Other Incommensurate Systems

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It is shown that in a simple model of surface dynamics of growing 3D quasicrystals, growth proceeds through the nucleation of steps whose heights h_z diverge like $(\Delta\mu)^{-1/3}$ as the growth-driving chemical-potential difference $\Delta\mu \rightarrow 0$. This large step size leads to very low growth velocities $V_g \propto \exp\{-\frac{1}{3}[\Delta\mu_c(T)/\Delta\mu]^{4/3}\}$. $\Delta\mu_c(T)$ defines a rounded kinetic roughening transition and is nonuniversal. For "perfect-tiling models" I find $\Delta\mu_c(T) \propto T^{-3/2}$ at high temperatures T, which fits recent numerical simulations, while in models with bulk phason Debye-Waller disorder, $\ln(\Delta\mu_c) \propto -\sqrt{T}$. The growing interface is algebraically rough.

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Quasicrystals[1] are intriguing because they force us to consider afresh all of the concepts that have been developed for periodic crystals. This is particularly true of surface roughening.[2-10] In conventional periodic 3D crystals, every facet has a finite roughening temperature[2] T_R, while in three-dimensional quasicrystals T_R is infinite.[7-10]

In this paper, I describe the behavior of a simple *dynamical* model for the growth[11] and kinetic roughening of quasicrystals. This model describes facets in the nonequilibrium case in which the quasicrystal is growing and hence the facets are moving at some finite velocity V_g. I find many important differences between a moving *quasicrystal* facet and a moving crystal one.

(1) The quasicrystal grows by nucleating steps on the surface whose height h_s is a diverging function of the quasicrystal-fluid chemical-potential difference $\Delta\mu$:

$$h_s = a\left(\frac{k_B T}{J}\right)^{1/2}\left(\frac{\Delta\mu_c(T)}{\Delta\mu}\right)^{1/3} \times O(1)\,, \qquad (1)$$

where J is a microscopic interaction energy, a is an atomic length, and $\Delta\mu_c(T)$ will be defined below. The exponent $\frac{1}{3}$ is universal for all quasicrystals whose ratio of fundamental incommensurate lengths is a quadratic irrational.[12] In contrast, crystal surfaces below T_R grow[13] by nucleating steps of height one lattice constant, regardless of $\Delta\mu$.

(2) This large step size leads to slower growth of the quasicrystalline facet, with growth velocity

$$V_g(\Delta\mu) = V_0 \exp\left[-\frac{1}{3}\left(\frac{\Delta\mu_c(T)}{\Delta\mu}\right)^{4/3}\right], \qquad (2)$$

whereas in crystals[13]

$$V_g(\Delta\mu) = V_0 \exp\left[-\frac{1}{3}\left(\frac{\Delta\mu_c(T)}{\Delta\mu}\right)\right]$$

(ignoring algebraic prefactors).

(3) The quantity $\Delta\mu_c(T)$ is nonzero up to arbitrarily high temperatures in quasicrystals, while in crystals[13] $\Delta\mu_c(T)$ vanishes at and above the equilibrium roughening temperature T_R. In quasicrystals, $\Delta\mu_c(T)$ is very sensitive to "phason Debye-Waller disorder"[14,15] in the bulk. In "perfect-tiling" models with no phason Debye-Waller disorder,

$$\Delta\mu_c(T) = \left(\frac{J^5}{(k_B T)^3[\ln(k_B T/J)]^{3/2}}\right)^{1/2} \times O(1) \qquad (3)$$

for $k_B T \gg J$. This result is supported by recent numerical simulations[10] (see Fig. 1). For models which allow phason Debye-Waller disorder, and again for $k_B T \gg J$,

$$\Delta\mu_c(T) \propto \exp[-(k_B T/J)^{1/2} \times O(1)]\,. \qquad (4)$$

(4) The height-height correlation function $C_h(\mathbf{r}) \equiv \langle [h(\mathbf{r}) - h(\mathbf{0})]^2 \rangle$ [where $h(\mathbf{r})$ is the height of the interface at $\mathbf{r}$, and the brackets denote a thermal average] exhibits two crossovers:

$$C_h(\mathbf{r}) \propto \ln(r/a)\,, \quad r \ll \xi_{\mathrm{eq}}(T)\,, \qquad (5)$$

$$C_h(\mathbf{r}) \propto \ln[\xi_{\mathrm{eq}}(T)/a]\,, \quad \xi_K(\Delta\mu, T) \gg r \gg \xi_{\mathrm{eq}}(T)\,, \qquad (6)$$

$$C_h(\mathbf{r}) \propto r^{2\chi}\,, \quad r \gg \xi_K(\Delta\mu, T)\,, \qquad (7)$$

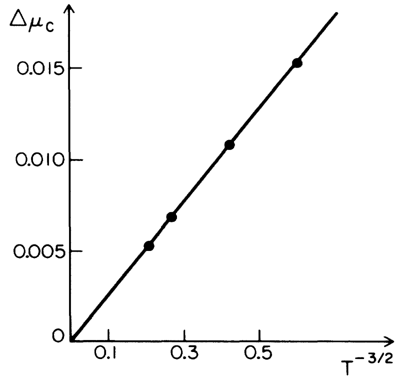

FIG. 1. Plot of the "kinetic roughening threshold" $\Delta\mu_c(T)$ vs $T^{-3/2}$, where $T \equiv k_B T/J$ is the reduced temperature in the perfect-tiling simulation of Ref. 10. The theoretical prediction presented here $\Delta\mu_c(T) \propto T^{-3/2}$ is illustrated by the straight line, while the points represent the results of the simulation.

where a is a microscopic length. Here χ is the universal exponent defined in Ref. 16 for a rather different growth model; its numerical value is[17] $\chi \approx 0.4$. I find

$$\xi_{\mathrm{eq}}(T) = a\frac{k_B T}{J}\left[\ln\left(\frac{k_B T}{J}\right)\right]^{1/2} \tag{8}$$

for perfect-tiling models, while

$$\xi_{\mathrm{eq}}(T) = a\exp[(k_B T/J)^{1/2}\times O(1)] \tag{9}$$

for models with phason Debye-Waller disorder. In both cases

$$\xi_K \propto V_g^{-1}, \tag{10}$$

so the small quasicrystalline growth velocity given in Eq. (2) manifests itself in a long surface correlation length as well. In contrast, crystals exhibit the crossover at ξ_{eq} only in a narrow range of temperatures below the roughening temperature T_R.

These results are expected to apply for any facet normal to an incommensurate direction in *any* incommensurate system, quasicrystalline or not, in which there are precisely two fundamental incommensurate lengths whose ratio is a quadratic irrational. The results do *not* apply for growth of a facet normal to a *commensurate* direction, even in systems which are incommensurate in other directions. For example, these results *do not* apply for facets parallel to the tenfold planes of decagonal quasicrystals,[18] but *do* apply in those systems for facets of any other orientation.

My model is a time-dependent Ginzburg-Landau model,[13]

$$\partial_t h = -\Gamma\,\delta H/\delta h + \eta, \tag{11}$$

where Γ is a kinetic coefficient and $\eta(\mathbf{r},t)$ a zero-mean Gaussian white noise with

$$\langle \eta(\mathbf{r},t)\eta(\mathbf{r}',t')\rangle = 2\Gamma k_B T\,\delta(\mathbf{r}-\mathbf{r}')\delta(t-t').$$

This choice of η guarantees that in the equilibrium ($\Delta\mu = 0$) case the distribution of h relaxes back at long times to the equilibrium distribution for the Hamiltonian H, where

$$H(h) = \int d^2r\left[\tfrac{1}{2}K|\nabla h|^2 + V(h) - \Delta\mu h\right], \tag{12}$$

with $V(h) = \sum_G V_G\cos(Gh)$. Here the sum is over all G's that can be written $G_{m_1,m_2} \equiv G_0(m_1 + m_2\sigma)$, with m_1 and m_2 integers and G_0 the wave number of one of the bulk reciprocal-lattice wave vectors orthogonal to the surface. Here σ is the ratio of incommensurate lengths in the quasicrystal, and is assumed to be a quadratic irrational.[12]

First, I will derive the static, equilibrium ($\Delta\mu = 0$) properties of this model; then, I will treat the dynamics of the nonequilibrium $\Delta\mu \neq 0$ case.

In the equilibrium case ($\Delta\mu = 0$) the linearized renormalization-group (RG) recursion relations for the V_G's follow from standard treatments[2] of the sine-Gordon model:

$$\frac{d\tilde{V}_G}{dl} = \left(2 - \frac{k_B T G^2}{4\pi K}\right)\tilde{V}_G, \tag{13}$$

where $\tilde{V}_G \equiv V_G/k_B T$. Since the V_G's suppress fluctuations of the interface, it will remain smooth if any of the V_G's renormalize to infinity. Since quasicrystals have V_G's at arbitrarily small G, this will always happen; hence[7,8] there is no roughening transition.

The $\tilde{V}_G$ that *first* renormalizes to $O(1)$ dominates the physics; I will define its wave vector as $G_c(T)$. From (13), it follows that a given $\tilde{V}_G = O(1)$ at a length scale

$$\xi_G = a[\tilde{V}_G(l=0)]^{-\lambda_G^{-1}}, \tag{14}$$

where $\lambda_B \equiv 2 - k_B T G^2/4\pi K$ and a is a microscopic length of order an interatomic distance. Physically, ξ_G is the length scale at which $\tilde{V}_G$ starts to have an appreciable effect on fluctuations of the interface. The smallest ξ_G ($\equiv \xi_{\mathrm{eq}}$) at a given temperature is, therefore, the length scale above which the equilibrium interface appears smooth; for $L < \xi_{\mathrm{eq}}$, the interface will be logarithmically rough. This is the crossover at ξ_{eq} described by Eqs. (5) and (6). The value of G at which ξ_G is minimized is $G_c(T)$. To proceed, I need to specify the bare $\tilde{V}_G(l=0)$'s. I will consider two classes of models here.

In the first, the perfect-tiling models,[4-6,9,10] the quasicrystal-liquid interface is modeled by first removing the tiles from a semi-infinite region of a perfect quasicrystalline tiling of space. Surface configurations are now generated by adding (or removing) tiles to (or from) this interface, following the rule that tiles are always added only in places where tiles were present in the original infinite tiling. Different surface configurations are now assigned different energies according to some local rule (e.g., by counting "broken bonds"[4-6]), and the interface evolves by conventional Monte Carlo dynamics.[9,10]

The long-distance properties of the interface in this model can be shown[19] to be described by the Hamiltonian (12) and the dynamical equation (11), with

$$\begin{aligned}\tilde{V}_{G(m_1,m_2)} &= O(1)\times\frac{V_0 G_0^2}{k_B T m_2^2}\\ &= O(1)\times\frac{V_0}{k_B T}G^2(m_1,m_2)\end{aligned} \tag{15}$$

for the choice of m_1 and m_2 that gives the largest $\tilde{V}_{G(m_1,m_2)}$ with a $G(m_1,m_2)$ within a few factors of σ of a given G. Here $V_0 \equiv V_{G_0}$ and $G_0 \sim a^{-1}$, where a is the tile size. Using this in Eq. (14), treating G as a continuous variable (which only leads to errors of a few factors of σ), and minimizing over G gives

$$G_c(T) = \left(\frac{4\pi K}{k_B T}\right)^{1/2}\Big/\left[\ln\left(\frac{k_B T}{J}\right)\right]^{1/2} \tag{16}$$

and Eq. (8) for $\xi_{\mathrm{eq}}(T)$, with $J \equiv Ka^2$.

My second model, the phason Debye-Waller disorder model, actually applies to any case in which the $V_{G(m_1,m_2)}$'s in the Hamiltonian equation (12) vanish faster than $\exp[-c\,|G_\perp(m_1,m_2)|]$ as the "phason momentum" $G_\perp(m_1,m_2) \equiv G_0(m_2 - m_1\sigma)$ associated with G goes to infinity. This behavior is precisely what is expected for systems with thermally excited phasons[14,15] in the bulk. Such a dependence of V_G on $G_\perp$ guarantees that the V_G's will be very small at small G, since any small G necessarily has a large $G_\perp$ associated with it. In particular,[20] $|G_\perp| > O(G_0^2) \times |G|^{-1}$. It follows that for small G, $V_G \leq V_0 e^{-CG_0/|G|}$ for some constant C of order unity.

The RG shows that any such model behaves at long distances like one with

$$\tilde{V}_G(l=0) = V_0 e^{-CG_0/|G|} \tag{17}$$

at small G, again for the right choice of (m_1,m_2) and with C of order unity. Equation (17) for $V_G(l=0)$ can now be used in Eq. (14) for ξ_g. Again, the minimization of ξ_G over G can be performed essentially treating G as a continuous variable, since there are guaranteed to be some G's within a few factors of σ of the continuous minimum. This yields

$$G_c(T) = 2(2\pi K/3k_BT)^{1/2}, \tag{18}$$

and Eq. (9) for ξ_{eq}, again with $J \equiv Ka^2$.

Let us now turn to the $\Delta\mu \neq 0$ dynamics.

In *crystals* described by the kinetic model Eq. (11), growth proceeds via the nucleation and growth of steps on the surface.[13] If the step height h_s is equal to the spatial period a of the crystal, a circular step of radius r will lower the energy of the system by $E_1 = -\Delta\mu A h_s = -\pi r^2 a \Delta\mu$. Choosing the step height to be a was crucial in that such a step leaves the periodic potential $V(h)$ unchanged.

The Hamiltonian *does* change on the step perimeter P, leading to a step energy $E_2 = \epsilon_s P = 2\pi\epsilon_s r$, where the step energy per unit length ε_s depends on K and $V(h)$.

Summing the perimeter and areal energies gives the energy of the nucleated step, $E_s(r) = -\pi r^2 \Delta\mu a + 2\pi\epsilon_s r$. This energy has the canonical form for a nucleating droplet,[13] with a "nucleation barrier" $E_b = \pi\epsilon_s^2/\Delta\mu a$ equal to the maximum of $E_s(r)$. In simple dynamical models like Eq. (11), thermal tunneling over this barrier occurs at a rate[13]

$$\gamma \propto \exp\left[-\frac{E_b}{k_BT}\right] = \exp\left[-\frac{\pi\epsilon_s^2}{\Delta\mu a k_BT}\right],$$

which vanishes very rapidly as $\Delta\mu \to 0$.

How is this picture modified for quasicrystals? At the very first step: No matter what the step height h_s, the potential $V(h_s)$ cannot be zero [choosing the zero of potential to be the minimum of $V(h)$ over all h], since $V(h)$ is not a periodic function of h. Thus, in addition to the $\Delta\mu$ term, the areal-energy change will always have a piece proportional to $V(h_s)$. Furthermore, if the interface before the step was sitting in a favorable (i.e., fairly low-energy) local minimum of $V(h)$, then any step h_s that only carries the interface to a nearby minimum will in general lead to $V(h_s) > \Delta\mu h_s$, so the step actually raises the energy by an amount proportional to the step area, rather than lowering it. Such a step can clearly never grow to infinity. The only steps that *do* grow are those with h_s large enough that $V(h_s) < \Delta\mu h_s$, for only then can such a step eventually lower the system's energy as it grows laterally. The smallest nucleation barrier will occur for the smallest h_s for which this condition can be satisfied;[19] hence, steps of this height will nucleate fastest and dominate the growth.

The dependence of h_s on $\Delta\mu$ is universal for all models with quadratic irrational σ's. Keeping only the two largest terms in the potential at a given temperature T, I have

$$V(h) = -\tilde{V}_{G_c(T)} k_BT\{\cos[G_c(T)h] + \cos[G_c(T)h/\tau]\}, \tag{19}$$

where I have specialized to the case $\sigma = \tau$ and ignored unimportant factors of order unity.

The problem of minimizing this potential is readily seen to be equivalent to the problem of finding an integer m that is as near as possible to being an integral multiple of τ. Fibonacci numbers are the best choice.[20] Choosing $m = F_k$, the kth Fibonacci number, one can readily show that

$$\frac{F_k}{\tau} = F_{k-1} - \frac{(-1)^k \tau^{-k}}{1+\tau^2} \approx F_{k-1} - \frac{(-1)^k}{5F_k}, \tag{20}$$

where the second, approximate, equality holds for large k or F_k.

Writing $h = (2\pi/G_c)F_k + \epsilon$ with $\epsilon \ll 1$, expanding the cosines in (19), taking advantage of the facts that F_{k-1} is an integer and $1/F_k \to 0$ as $F_k \to \infty$, and minimizing over ϵ, I obtain

$$\begin{aligned} V(h) &\approx \frac{\tilde{V}_{G_c(T)} k_BT}{F_k^2} \times O(1) \\ &= \left(\frac{1}{G_c h}\right)^2 \tilde{V}_{G_c} k_BT \times O(1). \end{aligned} \tag{21}$$

The optimal step size h_s is that value of h for which this first becomes $< \Delta\mu h_s$. Thus, it is the first Fibonacci number multiple of $2\pi/G_c(T)$ larger than h_c, where

$$\tilde{V}_{G_c} k_BT / G_c^2 h_c^2 = \Delta\mu h_c. \tag{22}$$

Since successive Fibonacci numbers differ only by factors of τ, h_s will be within a factor of τ of the solution h_c of (22); using this and

$$G_c^2 \sim (K/k_BT) \times \text{logarithmic factors},$$

I arrive, up to logarithmic factors, at Eq. (1) for h_s, with

$$\Delta\mu_c(T) = \frac{\tilde{V}_{G_c} k_B T}{\sqrt{G_c}} \left(\frac{K}{k_B T} \right)^{3/4} . \tag{23}$$

To calculate the nucleation rate I need the nucleation barrier. The energy gain due to nucleating the step is

$$E_1 = -\Delta\mu h_s r^2 \times O(1) . \tag{24}$$

I still need the step energy per unit length ϵ_s. Note that, since $h_s \gg a$, a step large compared to the microscopic lattice constant must be taken. Such a step consists[19] of a series of smaller steps of height $2\pi/G_c(T)$, with total step energy per unit length

$$\epsilon_s(h_s) = h_s (K\tilde{V}_{G_c(T)} k_B T)^{1/2} \times O(1) . \tag{25}$$

The total energy of a step is therefore given, up to factors of $O(1)$, by

$$\begin{aligned} E(r) &= \epsilon_s(h_s) r - \Delta\mu h_s r^2 \\ &= h_s [r (K k_B T \tilde{V}_{G_c(T)})^{1/2} - \Delta\mu r^2] . \end{aligned} \tag{26}$$

Maximizing this over r gives the nucleation barrier

$$E_b = \frac{K k_B T \tilde{V}_{G_c(T)}}{\Delta\mu} h_s = k_B T \left(\frac{\Delta\mu_c(T)}{\Delta\mu} \right)^{4/3} ,$$

where I have used Eq. (1) for h_s and ignored factors of $O(1)$.

From E_b, the step nucleation rate follows from $\gamma \propto \exp(-E_b/k_B T)$.

Inserting the expressions derived earlier for $\tilde{V}_G$ and $G_c(T)$ in the two models (phason Debye-Waller and perfect tiling) into Eq. (23) yields Eqs. (3) and (4) for $\Delta\mu_c$.

Once a step has grown substantially larger laterally than the nucleation radius r_c, it subsequently grows radially with a velocity V which is proportional to $\Delta\mu$. This type of growth model falls into the category of polynuclear growth (PNG) models, which have been studied extensively for crystals.[17,21,22] Combining the results of these studies with those obtained here, I find that the growth velocity of the interface is given by

$$V_g = h_s (\gamma V^2)^{1/3} \propto \exp\left[-\frac{1}{3} \left(\frac{\Delta\mu_c(T)}{\Delta\mu} \right)^{4/3} \right] . \tag{27}$$

Standard results[17,21,22] for the PNG model also imply the algebraic form for the height-height correlation function given in Eq. (7), with[21]

$$\xi_K = \left(\frac{V}{\gamma} \right)^{1/3} \propto \exp\left[\frac{1}{3} \left(\frac{\Delta\mu_c(T)}{\Delta\mu} \right)^{4/3} \right] . \tag{28}$$

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