Phason strain in an energetic growth model of a quasicrystal

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Energetic growth model of a two-dimensional quasicrystal using Penrose tiles produces phason strain with many topological configurations. These vortexlike structures produce power-law decay of the phason correlation function, with the exponent dependent on the temperature as well as on the interaction energy between the tiles.

I. INTRODUCTION

The mathematical descriptions of quasicrystal^{1,2} show that for a given quasicrystalline symmetry, there is an uncountably infinite number of distinct packing of unit cells. At present, these packings [different local isomorphism (LI) classes] and the unit-cell decoration cannot be determined simultaneously from diffraction data. Steinhardt and co-workers introduce three criteria for the identification of LI classes realizable in solids^{3,4} using growth algorithm: structures should be *restorable*, *growable*, and *rapidly growable*. Since these criteria are chosen to make the packing stable and physically accessible, and that experimental data^{5,6} suggest real quasicrystals always have some defects, a local realization of these criteria is to grow the quasicrystal using energetic models. Atomic decoration can then be explored to compare with experiments. This approach is also supported by the icosahedral glass school,^{7,8} which asserts that a sensible description of quasicrystal is to start from a random packing of clusters with appropriate constraints of local order. The energetic model here attempts to interpolate these two schools: quasicrystalline models and icosahedral glass models, by employing energy parameters to control local order. Here we address only one of the many features revealed by the energetic models: topological disorder which produces power-law decay of the phason correlation function.

In a previous paper,⁹ we have studied the energetic growth of two-dimensional quasicrystals using two kinds of disks, with local configuration dictated by tenfold symmetry. The structures depend critically on the interaction energy parameters between the disks. For certain range of energy parameters, we found a perfect (compact) structure which looks like a multiple-twinned sample describable as the image of projection^{10,11} of a three-dimensional staircase on two dimensions. We now generalize this energetic growth algorithm to the fat and skinny Penrose tiles with angles (72°, 108°) and (36°, 144°). (This choice of units can be generalized to other units such as squares and triangles.¹²) These units are grown with a fixed concentration $C_f = 0.618 = 2 \cos 72^\circ$ and with pairwise interaction energy: $(E_{ff}, E_{fs} = E_{sf}, E_{ss})$. (Here the subscripts *f* and *s* stand for fat and skinny tile.) Although our preliminary exploration in energy space has not found a perfect Penrose tile, nor any quasicrystalline

ground state, we do find interesting topological structures in the phason coordinate space. A standard analysis of phason strain¹³⁻¹⁵ reveals a power-law decay of the phason correlation function with the size of the sample, rather than a logarithmic decay expected from entropic quasicrystal.

II. GROWTH ALGORITHM

Our basic idea is to incorporate local energetic rules in the layer-by-layer growth algorithm. (Layer here refers to the surface perimeter of our two-dimensional cluster.) There are three lists generated in each layer growth. The existing surface atoms form the list of cluster atoms $\{C\}$ and the possible growing points from the surface atoms form the list of potential surface atoms $\{P\}$. (We do not use the term surface atoms because some of the potential surface atoms $\{P\}$ do overlap with neighbors or other P 's during growth, thereby these potential surface atoms are excluded in this algorithm.) The third list keeps track of the inactive bonds on the surface that cannot generate any new tiles because of overlap with existing tiles in the cluster. This is an idealization of quenched growth as adjustments of the neighboring tiles do not occur to accommodate the new tiles. In a given layer, the growth priority is sorted according to the magnitude of their Boltzmann factors which calculation is explained below. This layer-by-layer algorithm hopefully captures some of the features in the Burton-Cabrera-Frank theory of crystal growth.¹⁶

Energetics introduces randomness in our growth process. A random number p determines the composition in the incoming flux and another random number q determines the growth probability of the new tile. If the choices determined by p and by q are compatible, (e.g., p chooses a skinny tile and q chooses either a 36° or 144° turn), and if there is no overlap of the new tile with the existing cluster and with the $\{P\}$ generated before, then the potential surface atoms F and G are created and become part of the list of the cluster atoms $\{C\}$.

The calculation of growth probability involves a simple Hamiltonian for the pairwise interaction between nearest neighbors. From a given cluster bond AB , we calculate four probabilities values: $P_{36}, P_{72}, P_{144}, P_{108}$ which sum to unity. Let us consider the growth of a skinny tile $ABFG$ obtained from the cluster bond AB by making a

36° turn. The potential surface bond FG is determined by computing the interaction energy plus the chemical potential for four cases: (1) F and G already exist, (2) F is new and G already exists, (3) F already exists and G is new, and (4) F and G are both new. For each case, the Boltzmann factor B_{36} is given by $\exp(-E/T)$ with $E = E_{36} + u$.

Here u is the pairwise interaction energy between tiles. Many-body effects are incorporated in the formation of the tiles and longer-range correlations between tiles are ignored. The pairwise potential takes into account the local curvature effect of the growing surface through the number of nearest-neighboring tiles. The E_{36} term is the chemical potential associated with a skinny tile obtained by a turn of 36°. It is an energy parameter controlling anisotropic growth. The interesting case of anisotropic growth, which often occurs in a real system, will be addressed in a separate paper. Here we address only the isotropic case by setting $E_{36} = E_{144} = E_{72} = E_{108} = 0$. After calculating in a similar way the values of B_{144} , B_{72} , and B_{108} , we get the normalized Boltzmann probabilities: P_{36} , P_{144} , P_{72} , P_{108} . By dividing the unit interval into four sections with length proportional to the Boltzmann probabilities, and using q , we determine which kind of turn is in order, thereby the new potential surface atoms F and G .

We go through the surface layer of the clusters till we form the new surface layer with the list of potential surface atoms. Usually this new surface layer contains part of the old surface layer, as inactive bonds often exist. These inactive bonds will form the perimeter of the defective regions eventually, when the potential surface atoms in some future layers close up the defective region. These regions are defects which can be analyzed using the classification scheme of Wu and Szeto¹⁷ (See Fig. 1).

III. SIMULATION RESULTS

Five sets of energy parameters are used for the growth simulation: $(E_f, E_{fs} = E_{sf}, E_{ss}) = (1, 1, 1)$, $(10, 1, 1)$, $(1, 10, 1)$, $(1, 1, 10)$, and $(10, 1, 10)$. For each set, 50 samples are generated for 31 layers (≈ 2000 vertices and ≈ 1300 tiles). The phason width given by $W_h(N) = \langle 1/N \sum_i [\mathbf{h}(\mathbf{r}_i) - \bar{\mathbf{h}}]^2 \rangle$ is measured. Here $\langle \rangle$ means average over 50 samples, N is the number of vertices in the cluster, and $\bar{\mathbf{h}}$ is the mean value over the cluster of the phason coordinate $\mathbf{h}(\mathbf{r}) = \sum_{j=1}^5 n_j \mathbf{e}_j^\parallel$. Here $\mathbf{r} = \sum_{j=1}^5 n_j \mathbf{e}_j^\parallel$. The basis vectors $\mathbf{e}^\parallel$ and $\mathbf{e}^\perp$ are given by

$$\mathbf{e}_j^\perp = \cos \left[(j-1) \frac{4\pi}{5} \right] \hat{x} + \sin \left[(j-1) \frac{4\pi}{5} \right] \hat{y} \quad (1)$$

and

$$\mathbf{e}_j^\parallel = \cos \left[(j-1) \frac{2\pi}{5} \right] \hat{x} + \sin \left[(j-1) \frac{2\pi}{5} \right] \hat{y} . \quad (2)$$

In Fig. 2 the phason strain for $(1,1,1)$ and $(1,10,1)$ at $T=1$ and $T=100$ is plotted. For reference, phason strain for the random tiling model at equilibrium^{15,18} is illustrated in Fig. 3. These two figures show very different phason strains. If we go back to the postulate of Elser¹⁹ and Henley²⁰ on the square-gradient entropic phason free energy given in the absence of dislocations by

$$F = \frac{K}{2k_B T} \int d^2 \mathbf{r} [\nabla_i h_j(\mathbf{r})]^2 , \quad (3)$$

where K is the elastic constant, we arrive at the following size dependence of $W_h(N)$:

$$W_h(N) = \frac{1}{2\pi K} \ln N + \text{const} . \quad (4)$$

FIG. 1. Growth of nine layers for the $(1,10,1)$ sample at $T=1$ and $C_f=0.618$. The initial seed is the unmarked fat tile and the defects are darkened. The layer number runs from 0 to 9.

FIG. 2. Phason strain for a (1,1,1) sample with 31 layers at (a) $T=1$ (b) $T=100$ and a (1,10,1) sample at (c) $T=1$ and (d) $T=100$. The concentration of fat tiles is 0.618. The number of sites, N , are (a) 1748, (b) 1749, (c) 2313, and (d) 1840.

However, Fig. 2 shows regions resembling vortices in a two-dimensional XY magnet, but the magnitude of $\mathbf{h}^\perp$ is not a constant. A better description is to treat the phason coordinate $\mathbf{h}$ as the projection of a Heisenberg spin $\mathbf{S}$ of fixed length onto the XY plane so that the magnitude of the projection, $(S_x^2 + S_y^2)^{1/2}$ is $|\mathbf{h}|$. With this mapping in mind, Fig. 2 shows that dislocations, and very possibly topological excitations such as Belavin-Polyakov solitons,²¹ exist at finite temperature growth.

The soliton solution of Belavin and Polyakov makes use of the stereographic mapping of our two-dimensional space (the plane) onto the unit sphere $|\mathbf{v}|=1$ and a subsequent mapping of this sphere into the unit sphere in the *spin space*, $|\mathbf{s}|=1$. To each point $z=x+iy$ of the plane, the coordinates of the corresponding point on the sphere $|\mathbf{v}|=1$ is given by

$$x + iy = \frac{v_1 + iv_2}{1 - v_3}. \quad (5)$$

Similarly for the spin $\mathbf{s}(\mathbf{r})$, we can define the variable

$$w = w_1 + iw_2 = \frac{s_1 + is_2}{1 - s_3}. \quad (6)$$

In homotopy theory, the mapping from the sphere S^2 to S^2 is classified by a soliton number or topological charge Q . It can be shown that for spin

$$\begin{aligned} \mathbf{S}(\mathbf{r}) &= s_1 \hat{x} + s_2 \hat{y} + s_3 \hat{z} \\ &= \sin\theta \cos\phi \hat{x} + \sin\theta \sin\phi \hat{y} + \cos\theta \hat{z}, \end{aligned}$$

this topological charge (soliton number) is

FIG. 3. Phason strain for an (8,5) approximant entropic quasicrystal in equilibrium after 3000 Monte Carlo steps. $N = 521$.

$$\begin{aligned}
 Q &= \frac{1}{4\pi} \int dx dy \sum_{\mu\nu} \epsilon_{\mu\nu} \mathbf{S}(\mathbf{r}) \cdot [\partial_\mu \mathbf{S}(\mathbf{r}) \times \partial_\nu \mathbf{S}(\mathbf{r})] \\
 &= \frac{1}{4\pi} \int dx dy \sin\theta \left[\frac{\partial\theta}{\partial x} \frac{\partial\phi}{\partial y} - \frac{\partial\theta}{\partial y} \frac{\partial\phi}{\partial x} \right] \\
 &= \frac{1}{\pi} \int \frac{d^2r}{(1+|w|^2)^2} \left[\frac{\partial w}{\partial z} \frac{\partial w^*}{\partial z^*} - \frac{\partial w}{\partial z^*} \frac{\partial w^*}{\partial z} \right]. \quad (7)
 \end{aligned}$$

Here the index μ and ν runs from 1 to 2 corresponding to the x and y position coordinates.

This conjecture is borne out by a calculation of effective soliton number Q_{eff} using a normalized phason strain. For the set of all phason vectors ($\mathbf{h}(\mathbf{r})$) in a finite-grown sample, we first find out the one (say $\mathbf{h}^*$) with maximum magnitude h_{max} and use it to define our Heisenberg spin vector. We can choose our spin coordinate with $\hat{z}_s$ as $\mathbf{h}^*$. Next we can work out the polar angle and azimuthal angle of all other $\mathbf{h}$. The set of phason coordinates $[n_1(i), n_2(i), n_3(i), n_4(i), n_5(i)]$ for the i th vertex of a given tile then corresponds to a two-dimensional vector shown as an arrow (a unit magnitude Heisenberg spin) attached to the physical coordinates of the i th vertex. For a given tile with four vertices labeled by i , one can compute the solid angle subtended by the four phason vectors by choosing a counter clockwise convention when going through the four vertices of the tile. The solid angle is then normalized by the tile area, and the effective soliton number Q_{eff} is the effective number of times the spin spherical surface (which is the sum of the normalized solid angles), wraps around the physical space of the grown structure. This will be exact when the tile edges go to zero and when the size of the grown sample goes to infinity.

We have computed Q_{eff} for fifty 25-layer samples with energy parameter $(E_{\text{ff}}, E_{\text{fs}}, E_{\text{ss}}) = (1, 1, 1)$. We found that

TABLE I. Fifty 25-layer samples with energy parameter $(E_{\text{ff}}, E_{\text{fs}}, E_{\text{ss}}) = (1, 1, 1)$.

Temperature	Effective Q	Error in Q
1.00	2.003 36	0.244 58
5.00	2.065 62	0.353 05
10.00	2.028 07	0.231 53
50.00	2.005 13	0.267 60
100.00	2.041 38	0.262 82

$Q_{\text{eff}} \approx 2$ for many cases but never zero²² (Table I). Other samples also show similar nonzero Q_{eff} . This suggests that we should generalize the analysis of phason fluctuation about the perfect quasicrystalline value ($\mathbf{h}^1(\mathbf{r}) = \text{const.}$) for the thermodynamic stability and translational order to include these topological configurations. A power-law scaling, $W_h(N) \approx N^\eta$, is expected.

We find that the quality of fit for $W_h(N)$ vs N with the power law is better than the logarithmic fit. (For the logarithmic fit, only the large- N values are used in the fit, as the inclusion of small- N values gives a worse result.) The values of the slope $1/(2\pi K)$ in [Eq. (4)], and η are shown in Fig. 4 and Fig. 5 for various temperature. Temperature dependence in the slope in both figures exist, in disagreement with the prediction of the random tiling model.²³ The (1,10,1) case shows the most pronounced temperature dependence of the slope in either fit, possibly due to the formation of chemical order between the skinny and fat tiles. The results here are similar to the strain accumulation²⁴ in the growth model of Elser.¹⁹

We have also checked if the dislocations and topological excitations are destroying the tenfold symmetry by a

FIG. 4. Slope of logarithmic fit $1/2\pi K$ vs temperature T . [$\circ$], (1,1,1); [$+$], (1,1,10); [H], (1,10,1); [$*$], (10,1,1); [$\times$], (10,1,10).

FIG. 5. Slope of power-law fit η vs temperature T . [$\circ$], (1,1,1); [$+$], (1,1,10); [H], (1,10,1); [$*$], (10,1,1); [$\times$], (10,1,10).

linear fit $\mathbf{h}(\mathbf{r}) = \mathbf{M}\mathbf{r} + \mathbf{b}$ for the phason strain.¹⁹ If tenfold symmetry is preserved, then $\mathbf{M}$ vanishes. We use the best-fit values of the invariants combinations $\alpha^2 + \beta^2$ and $\gamma^2 + \delta^2$ for the values of $\mathbf{M}$ in the form¹³

$$\mathbf{M} = \begin{pmatrix} \alpha + \gamma & \beta + \delta \\ \beta - \delta & \gamma - \alpha \end{pmatrix}. \quad (8)$$

We find that these invariants decrease rapidly to zero as one increases the size of the cluster. This observation to-

gether with diffraction pattern confirm that our growth model in general does preserve tenfold symmetry. However, this tenfold symmetry can be broken in certain extreme cases of very low temperature and/or large difference in interaction energy.

IV. DISCUSSION

In conclusion, we have constructed an energetic model for the growth of a quasicrystal using Penrose tiles and a layer-by-layer algorithm. The results suggest that the presence of dislocations and topological disorder introduce an important correction to the purely entropic description of the phason fluctuations and that these excitations yield a power-law dependence of the width of the phason coordinates on the size of the system. Furthermore, the flexibility of the energetic model allows one to perform more exploration in energy space for various grown samples, thereby bridging the gap between the two schools of thought: (1) one should approach the structure from a perfect quasicrystal structure (such as generated by the generalized dual method) and introduce defects later or¹⁻³ (2) one should approach the structure from a random packing glassy-type structure and introduce local order.^{15,19,20,23} For comparison with the experiments, one can use different models for decoration²⁵ to look at such details as the diffraction pattern and diffuse scattering intensities.²⁶

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