Ising-Like Transition and Phason Unlocking in Icosahedral Quasicrystals

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Using Monte Carlo computer simulations, we find strong evidence for a novel, finite-temperature, Ising-like transition in the alignment of unit cells in icosahedral quasicrystals. The Ising-like transition may be related to the transition in phason behavior from a locked (Penrose-tiling-like) phase at low temperatures to an unlocked (random-tiling-like) phase at high temperatures.

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In this paper, we present evidence for a finite-temperature phase transition in the alignment of unit cells in icosahedral quasicrystals. The phase transition occurs in models of "energetically stabilized" quasicrystals in which atomic interactions force the structure into an ideal, quasiperiodic arrangement at low temperatures [1]. These quasicrystals are analogous to the Penrose tilings (PTs) in which Penrose matching rules, representing the atomic interactions, force a perfect quasiperiodic configuration of tiles, representing unit cells of atoms. At zero temperature, the ground state of 3D icosahedral quasicrystals displays planar sheets of unit cells normal to the twofold symmetry axes. The unit cells within each sheet are aligned. Above the critical temperature T_I we find that each layer undergoes an Ising-like disordering transition analogous to the 2D Ising transition.

We conjecture that the Ising transition is related to a transition in phason behavior. Phasons are a key feature that distinguishes quasicrystals from periodic crystals [2]. Phasons and phonons are the two types of elastic Goldstone modes exhibited by quasicrystals. Phonons, associated with uniform translations, are common to both periodic crystals and quasicrystals; but phasons are associated with relative translations of incommensurate periodicities unique to quasiperiodic structures. On the microscopic level, the effect of phason excitations is to rearrange the unit cells (analogous to rearranging tiles in a Penrose tiling) [3]. In a 3D Penrose tiling, it is believed that thermal excitation of phason fluctuations is strongly suppressed at low temperatures (in analogy to pinned phasons in the 1D Frenkel-Kontorova model) and the elastic free energy is nonanalytic [$\mathcal{F} \propto |\nabla w|$, where $w(x)$ is the phason elastic field]. Following [4], we refer to this as a "locked phase." Here we explore numerically a possible transition at high temperatures, $T > T_U$, to an "unlocked phase" in which phason excitations are thermodynamically excited and the elastic free energy is proportional to $(\nabla w)^2$. (These properties are similar to "random tilings" (RTs) [4–7].) Specifically, we have studied equilibrium thermodynamics and searched for a transformation in thermal mean square phason fluctuations expected for the unlocked phase.

We present numerical evidence for both the Ising-like transition in unit cell alignment and the phason unlocking transition. The Ising-ordered, phason-locked phase is stable over a finite temperature range in 3D (whereas, in 2D, the locked phase is unstable for any finite temperature [5,6]). Since the Ising-like disorder in the alignment of unit cells occurs through phason rearrangement, we conjecture that the two transitions are coincident, $T_I = T_U$. Our results are consistent with a single critical temperature, although we cannot rule out the possibility that the critical temperatures are different.

Our results are obtained using a Monte Carlo simulation procedure similar to methods used in prior studies of 2D PTs [6] and 3D decagonal phases [8]. Our model of the icosahedral quasicrystal consists of a close packing of prolate and oblate rhombohedra, idealized units representing atomic clusters [1]. The rhombohedra, 3D analogs of Penrose tiles, are packed initially to form a periodic approximant [6,9] to the ideal, icosahedral quasicrystal so that periodic boundary conditions can be employed. Each approximant is characterized by an integer n, where the approximant contains $N_n = 4F_{3n+3}$ rhombohedra in its unit cell. (F_n are Fibonacci numbers.) We use a sequence of increasingly close approximants $n = 3$, 4, 5, and 6 corresponding, respectively, to $N_n = 576, 2440, 10\,336$, and $43\,784$.

To represent energetics that drives the system towards an ideal, quasiperiodic ground state at low temperature, interactions related to the "alternation conditions" described by Socolar [10] are imposed between rhombohedra. Each rhombohedron lies at the intersection of three criss-crossing "trails" consisting of a string of rhombohedra joined at faces that are parallel to one another. Along any trail, a given prolate or oblate rhombohedron may have one or two possible orientations related by reflection. The alternation condition requires that, for either shape, the orientations must alternate along the trail. Socolar has shown that the conditions are "weak" matching rules which force quasiperiodic translational order. Hence, we assign a finite energy ϵ for each pair of consecutive prolate (or oblate) rhombohedra along a trail which has the same orientation. The energetics ensure that the alternation conditions are satisfied at zero temperature.

A Monte Carlo move entails flipping rhombic dodecahedral elements consisting of two obtuse and two prolate rhombohedra intersecting at a common, interior vertex.

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Flipping dodecahedra is analogous to flipping hexagons in 2D PT or RT simulations. [6] The long axis of each dodecahedron lies along one of the fifteen twofold directions of icosahedral symmetry; see Fig. 1. If we choose a basis, $e_0^{\parallel} = \eta(0,1,\tau), e_1^{\parallel} = \eta(\tau,0,1), e_2^{\parallel} = \eta(1,\tau,0), e_3^{\parallel} = \eta(0,-1,\tau), e_4^{\parallel} = \eta(\tau,0,-1), e_5^{\parallel} = \eta(-1,\tau,0)$, where $\eta = (1+\tau^2)^{-1/2}$, then the dodecahedron with long axis parallel to the x axis has edges along the four basis vectors, $e_1^{\parallel}, e_4^{\parallel}, e_2^{\parallel}$, and $-e_5^{\parallel}$. For each of the fifteen directions, there are two distinct ways of packing four rhombohedra to form the dodecahedron, corresponding to two different interior vertex positions. We assign the two possibilities an Ising-like "spin" value, ± 1. Flipping consists of transforming the dodecahedron from one spin to the other.

Consider the set of dodecahedra with axes parallel to the x axis, say. Each dodecahedron can be assigned to a sheet N_s,

$$N_s \equiv n_1 + n_4 + n_2 - n_5, \tag{1}$$

where the interior vertex point is $\mathbf{v} = \sum_0^5 n_i \mathbf{e}_i^{\parallel}$. N_s is defined such that flipping a dodecahedron changes the n_i, but N_s is unchanged. In perfect 3D PTs, the dodecahedra self-organize into planar "sheets" within which dodecahedra have a common spin orientation. Also, the dodecahedra occupy only a subset of sheets N_s, so that some sheets are densely filled with dodecahedra and some are sparsely occupied. With increasing T, phason excitations (Monte Carlo flipping) disorder the alignment among dodecahedra in each sheet; some original dodecahedra are broken apart and some new dodecahedra are formed. The new dodecahedra can occupy sheets not found in the perfect tiling. We note that the RT phase has a nonzero density of dodecahedra for each N_s (averaging over ensembles) indicative of positional disordering of the dodecahedra.

The Ising-like transition entails a misalignment of dodecahedra spin orientations within sheets. We introduce a new order parameter, the sheet magnetization $m_s \equiv \sum_{i(N_s)} S_{i(N_s)}$, where $S_i = \pm 1$ is the Ising-like value of the ith dodecahedron in sheet N_s. We also define the net magnetization [11],

$$m = \frac{1}{N_d} \sum_{\hat{x}} \sum_{N_s} |m_s|, \tag{2}$$

where N_d is the total number of dodecahedra in the count, and the outer sum is over orientations $\hat{x}$ [12].

For the simulations, we randomly select a vertex and check if it is the interior vertex of a dodecahedron; if it is, we determine whether to flip it or not according to the Metropolis Monte Carlo (MC) procedure assigning energy ϵ for each alternation rule mismatch. For a simulation with N vertices, N random selections comprise one MC step. Several runs of 20 000 MC steps after initial equilibrium stage were performed, and data were gathered every 25 MC steps. The fraction of vertices corresponding to interior points of dodecahedra in the infinite volume limit approach 0.175 at infinite temperature and 0.236 at low temperatures, in agreement with known values for the RT and PT limits, respectively.

Figure 2(a) indicates the order-disorder transition of

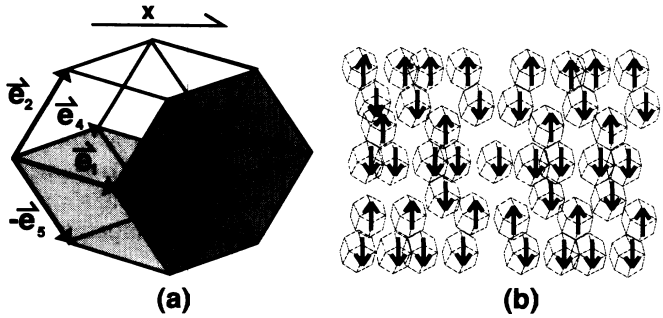

FIG. 1. (a) A rhombic dodecahedron composed of two prolate and two oblate rhombohedra joined at a central vertex (circle). Since the vertex lies on the long axis (along the x axis) off center to the right, the "spin" is defined to be along the x direction. (b) A projection of the 3D simulation at zero temperature, showing only the dodecahedra oriented along the vertical axis. The dodecahedra lie on (horizontal) planar sheets. The arrows indicate the spin. Note that all dodecahedra in a given sheet have the same spin.

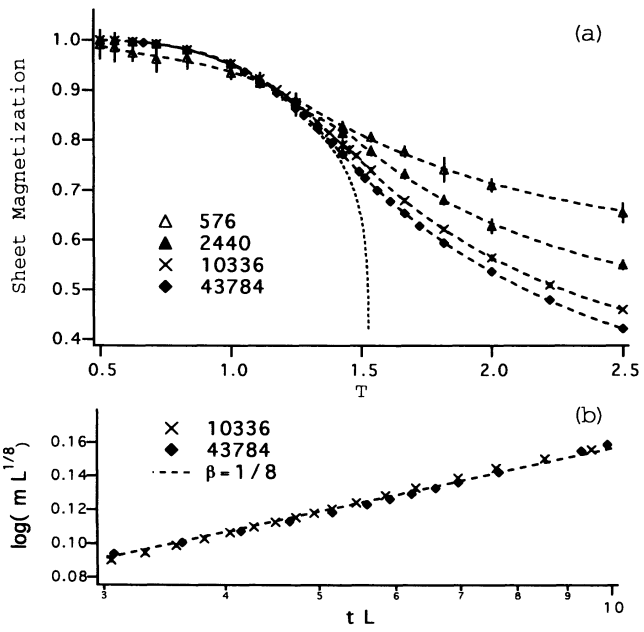

FIG. 2. (a) Sheet magnetization. The dotted lines correspond to an infinite system. (b) Scaling behavior illustrated in plot of $mL^{\beta/\nu}$ vs tL using the 2D Ising values $\beta = 0.125$ and $\nu = 1.0$. The data for the two largest simulations, $N = 43\,784$ and $N = 10\,336$, collapse onto a common line. The critical temperature is estimated from the peak of the susceptibility (Fig. 3). Error bars are smaller than the symbols for most points.

the sheet magnetization. The specific heat appeared to be slowly diverging but no latent heat release was observed. In Fig. 2(b), we show scaling behavior in a plot of $mL^{\beta/\nu}$ vs tL, where $t = (T_I - T)/T_I$ [11]. For 2D Ising exponents, $\beta = 0.125$ and $\nu = 1.0$, the data from the two largest simulations collapse onto a common straight line. (The 3D Ising exponents are $\beta = 0.31$ and $\nu = 0.64$.) Figure 3 shows susceptibility defined by [11]

$$\chi = (\langle N_d m_s^2 \rangle - \langle N_d m_s \rangle \langle m_s \rangle)/k_B T, \quad (3)$$

where $\langle \cdots \rangle$ denotes the ensemble average. The transition temperature T_I and critical exponents below T_I obtained from the largest simulations are $k_B T_I = (1.52 \pm 0.05)\epsilon$, $\gamma = 1.7 \pm 0.1$, where $\chi \sim |t|^{-\gamma}$.

Hence, an intriguing analogy is found between a phase transition in the alignment of unit cells in d dimensions and the Ising transition in $d-1$ dimensions. In 3D (2D), the quasicrystal contains dodecahedra (hexagons) arranged in planar sheets (linear trails). In each sheet (trail), the interactions, whether alternation conditions or Penrose matching rules, induce alignment of the dodecahedron (hexagon) spins within each layer [10]. Overall, the structure consists of grids of parallel sheets (trails) with different grids oriented along different symmetry directions. The grids are not completely equivalent to an ensemble of $d-1$ dimensional Ising models: (1) The interaction within a layer is not nearest neighbor; rather, it is communicated through finite-range alternation conditions (or matching rules). (2) There are interactions between sheets (trails) with different orientations where they cross one another. However, these differences have no apparent affect on the critical behavior. Quantitatively, the critical exponents found here for the 3D transition ($\beta = 0.125 \pm 0.010$; $\gamma = 1.7 \pm 0.1$) are in good agreement with the exponents for the 2D Ising model

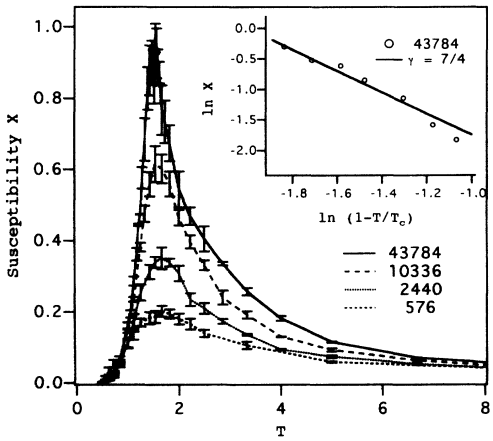

FIG. 3. Susceptibility. T_I is estimated as 1.52 ± 0.05. The inset shows a portion of log-log plot below T_I for the $N = 43\,784$ system. The solid line corresponds to $\gamma = 7/4$, the value for the 2D Ising exponent.

(and clearly distinct from the 3D Ising model, $\beta = 0.31$ and $\gamma = 1.25$). Numerically, our largest 3D model gives results comparable to 10×10 2D Ising simulations. In 2D, the unlocking transition for PTs is at zero temperature [6], as is the 1D Ising transition.

The Ising analogy does not capture all aspects of the structural difference between the two phases, though. There appears to be an associated positional disordering of the dodecahedra in 3D. At low temperatures, we find that the dodecahedra lie in a restricted subset of sheets N_s; hence, very high densities of dodecahedra are found for some values of N_s, and very low densities for other values. At the phase transition, so far as we can resolve, the sheets with high density of dodecahedra break up through the destruction of some dodecahedra and the creation of new ones in different sheets. The net result is that the dodecahedra become rather homogeneously spread in N_s.

To study the unlocking transition, we compute the temperature-dependent behavior of the mean-square phason fluctuations as a function of system size, L. In a 3D packing of rhombohedra, each vertex position can be expressed as $\mathbf{x} = \sum_{i=0}^{5} n_i \hat{\mathbf{e}}_i^{\parallel}$, where n_i is an integer. The associated value of the phason variable is $\mathbf{w}(\mathbf{x}) = \sum_{i=0}^{5} n_i \hat{\mathbf{e}}_i^{\perp}$, where $\hat{\mathbf{e}}_i^{\perp}$ are the complementary, "perpendicular space" unit vectors [4]. Then, the mean square phason fluctuation for a system with N tiles and linear dimension L is [13]

$$\langle w^2 \rangle_L = \left\langle \frac{1}{N} \sum_x \left[\mathbf{w}(\mathbf{x}) - \frac{1}{N} \sum_{\mathbf{x}} \mathbf{w}(\mathbf{x}) \right]^2 \right\rangle,$$

where the brackets denote ensemble average. In the unlocked phase, $\langle w^2 \rangle_L$ *increases* with L (for sufficiently large L) as $\alpha - (\beta/L)$, where α and β are temperature dependent. Unlocked behavior is usually associated with "random tilings" [4–7] in which no Penrose matching rules are imposed and, consequently, there is no long-range alignment of unit cells. Although all close-packed configurations of unit cells have equal energy at zero temperature, it is conjectured that finite-temperature phason excitations select out quasicrystalline order as the state of highest entropy and, consequently, lowest free energy. Here we find that energetically stabilized quasicrystals display this behavior at sufficiently high temperature, as had been suggested previously [5,6].

Figure 4 illustrates $\langle w^2 \rangle_L$ vs $1/L$. For $T > 2.5$, $\langle w^2 \rangle_L$ *increases* with L as $\alpha - (\beta/L)$ (approaching α from below), from which we infer unlocked phason behavior. For $T = 1.0$ (or below), $\langle w^2 \rangle_L$ *decreases* monotonically with L towards the zero-temperature Penrose value, 1.24. Although there is no rigorous elasticity theory for describing the locked phase, this behavior is consistent with locked phason fluctuations and inconsistent with an unlocked phase. Hence, we infer an unlocking transition in the range $1.0 \lesssim T \lesssim 2.5$. Note that the increase in $\langle w^2 \rangle_L$

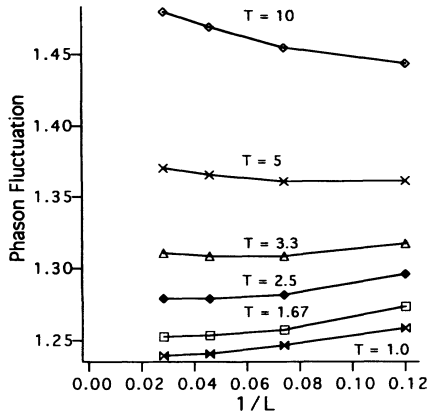

FIG. 4. The mean square phason fluctuations $\langle w^2 \rangle_L$ vs $1/L$ where L is the linear dimension ($\propto N^{1/3}$) and $N = 576, 2440, 10\,336$, and $43\,784$. For high temperatures, $\langle w^2 \rangle_L$ increases as $\alpha - (\beta/L)$, implying unlocked behavior, whereas for low temperatures it decreases.

with L only sets in for sufficiently large L, where the requisite L increases as T approaches the critical temperature. (For example, the $T = 10$ curve increases monotonically, but the $T = 3.3$ increases only for $N > 2440$ or $1/L < 0.07$.) Our limited computer capacity prevents us from extending the curves towards larger L and thereby more precisely determining the transition temperature.

In summary, we have found numerical evidence for both an Ising-like transition in the alignment of unit cells and a phason unlocking transition. The Ising transition implies a hitherto unknown symmetry change that appears to have no effect on the long-range translational or orientational order. The phason unlocking transition had been postulated previously [3,6] on the basis of comparison with the unpinning transition in the 1D Frenkel-Kontorova [14] (FK) model. In the FK model, balls connected by springs are draped on a sinusoidally corrugated potential. The phason mode, in which the balls are translated with respect to the potential, may be pinned or unpinned depending upon the coupling strength between the balls and the potential. The analogy break is suspicious, though, since the quasicrystal is no analogy to the corrugated substrate. Our conjecture is that a better understanding of the unlocking transition can be obtained by relating it to the Ising-like transition and the associated symmetry breaking. In fact, we suspect that there is only a single transition, $T_I = T_U$, and that the growth of phason fluctuations is directly linked to the disordering in the alignment of unit cells. At this point, our numerical simulation results are consistent with this conjecture, but we cannot rule out the possibility of two different transitions. Through future numerical and analytic work, we hope to clarify the nature of the phase transitions and explore the consequences of the Ising-like order for other physical properties of quasicrystals.

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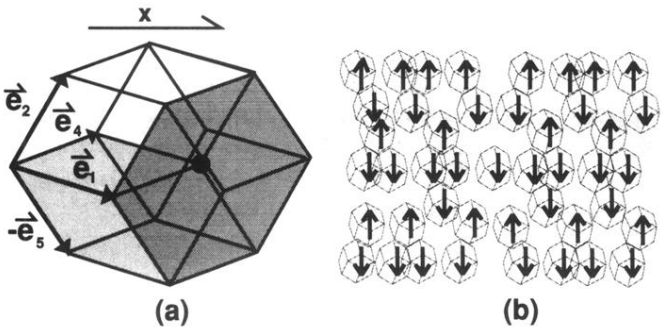

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