PHYSICAL REVIEW B VOLUME 58, NUMBER 13 1 OCTOBER 1998-I

Properties of random tilings in three dimensions

W. Ebinger, J. Roth, and H.-R. Trebin
Institut für Theoretische und Angewandte Physik, Universität Stuttgart, Pfaffenwaldring 57, 70550 Stuttgart, Germany

(Received 23 January 1998; revised manuscript received 9 March 1998)

Three-dimensional icosahedral random tilings with rhombohedral cells are studied in the semientropic model. We introduce a global energy measure defined by the variance of the quasilattice points in orthogonal space and justify its physical basis. The internal energy, the specific heat, the configuration entropy, and the sheet magnetization (as defined by Dotera and Steinhardt [Phys. Rev. Lett. **72**, 1670 (1994)]) are calculated. Since the model has mean-field character, no phase transition occurs in contrast to matching-rule models. The self-diffusion coefficients closely follow an Arrhenius law, but show plateaus at intermediate temperature ranges, because there is a correlation between the temperature behavior of the self-diffusion coefficient and the frequency of vertices which are able to flip (simpletons). We demonstrate that the radial distribution function and the radial structure factor depend only slightly on the random tiling configuration. Isotropic interactions lead to an energetical equidistribution of all configurations of a canonical random tiling ensemble and do not enforce matching rules. [S0163-1829(98)02937-3]

I. INTRODUCTION

The stability of quasicrystals has been a riddle since their discovery in 1982. Do they form stable or metastable states? Are they stable only at high temperatures? Is the stability due to energetic or due to entropic reasons? The random tiling model of quasicrystals is an abstraction which deals with rigid tiles, thereby neglecting thermal fluctuations and the phonon degrees of freedom. The only dynamic process is the local rearrangement of tiles, called ‘‘flips,’’ ‘‘umklapps,’’ ‘‘zipper’’ moves, depending on the type of tiles and symmetries. The random tiling model has been proposed by Elser[1] and has been studied intensively in recent years. The first author dealing with random rhombohedral tilings in three dimensions was Tang[2] who was interested in diffuse scattering and phason elastic constants. Strandburg[3] calculated the configurational entropy. The state of the art of random tilings was reviewed by Henley.[4] Ebinger[5] studied random tilings at infinite temperatures. Dotera and Steinhardt[6] introduced the concept of sheet magnetization as an order parameter to describe the randomness. Interest in random tilings was renewed by Kalugin and Katz[7] through the new process of flip diffusion. This property has been studied by Jarić and Sǿrensen[8,9] at infinite temperature, and by Joseph[10] and Gähler[11] at finite temperatures. Meanwhile for the energetic interaction of the tiles many different sets of matching rules exist.

In the present work we first deal with quantities that allow to characterize a random tiling (Secs. II and III). The harmonic energy measure is then introduced in Sec. IV and compared with other energy measures. In Sec. V we shortly describe the construction of random tilings. The possibility of a random tiling equilibrium phase transition and how it can be detected is studied in Sec. VI. Results for the special harmonic energy measure are presented in Sec. VIII. The thermodynamic functions, the possiblility of a phase transition and self-diffusion are treated in Sec. VIII A, followed by the structure functions and vertex statistics in Sec. VIII B. Conclusions are drawn in Sec. IX.

II. DEFINITIONS

Quasicrystals are described as cuts through higher-dimensional periodic crystals. The additional dimensions are addressed as orthogonal space $\mathbb{E}^{\perp}$. Quasilattice points $\mathbf{x}$ in the physical space $\mathbb{E}^{\parallel}$ can uniquely be lifted to the higher-dimensional space $\mathbf{X}=\Pi_{\parallel}^{-1}\mathbf{x}$ and then be projected onto the orthogonal space by $\mathbf{y}=\Pi_{\perp}(\mathbf{X})$ $(\Pi_{\parallel}+\Pi_{\perp}=id$ in a proper normalization). The infinitely extended quasilattice thus is contracted into a finite volume called ‘‘acceptance domain’’ or ‘‘atomic hypersurface.’’ The whole procedure of lifting and projecting into the orthogonal space is called ‘‘dualization’’ $\mathbf{y}=\Pi_{\perp}\circ\Pi_{\parallel}^{-1}\mathbf{x}$ of the quasilattice. The higher-dimensional embedding generates new degrees of freedom in addition to the ordinary phonons in periodic crystals, denoted ‘‘phasons.’’

The icosahedral quasilattice lifted into higher-dimensional space forms a three-dimensional hypersurface, called ‘‘de-Bruijn-hypersurface’’ or ‘‘Weiringia roof.’’[12] The hypersurface fluctuates around an average hyperplane $\mathbf{h}(\mathbf{x})=\mathbf{h}_0+\varepsilon_{\text{global}}\mathbf{x}$. The constant quantity

$$\varepsilon_{\text{global}}:=\nabla_{\parallel}\otimes\mathbf{h}(\mathbf{x})=\text{const}$$

is called ‘‘global phason strain’’ and describes the deviation of the slope of the average hyperplane $\mathbf{h}(\mathbf{x})$ from the slope of the physical space. In the case of exact icosahedral symmetry the average hyperplane is running parallel to the physical space and thus $\varepsilon_{\text{global}}=0$.

Long-wavelength deviations from the hypersurface are denoted as ‘‘phason fluctuations’’ and are described in a continuum picture by a phason strain tensor

$$\varepsilon(\mathbf{x}):=\nabla_{\parallel}\otimes\mathbf{h}(\mathbf{x}).$$

According to Henley[13] the fluctuations are governed by a free energy depending quadratically on the phason strain:

$$F=\int d^3\mathbf{x}\ \text{trace}(\varepsilon(\mathbf{x})\otimes\varepsilon(\mathbf{x})^T).$$

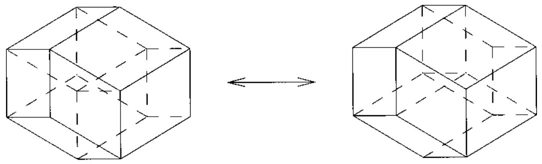

FIG. 1. Simpleton flip.

The present work deals with the three-dimensional Ammann-Kramer-Penrose tiling and its randomizations. This tiling consists of two different elementary cells, the ''oblate'' and ''prolate'' rhombohedron. Both of them appear in ten different orientations. The six rationally linearly independent unit edge vectors $\mathbf{t}_\alpha$ may be defined as $\mathbf{t}_\alpha = (2/\sqrt{5})(\cos\frac{2}{5}\pi\alpha, \sin\frac{2}{5}\pi\alpha, \frac{1}{2})$ $(\alpha=0,\dots,4)$ and $\mathbf{t}_5=(0,0,1)$. The atomic hypersurface is a rhombic triacontahedron of icosahedral symmetry. The tiling exhibits, among others, a vertex representing lattice points where two prolate and two oblate rhombohedra meet. It is denoted ''simpleton vertex.'' The surface of the cells which touch this vertex is a rhombic dodecahedron. Two possibilities exist how to fill the dodecahedron with tiles. The exchange of one configuration by the other is a ''flip.'' The lattice point jumps a distance which is 0.650 of the edge length of the rhombohedra (Fig. 1). Any nonordered space filling arrangement of rhombohedra without gaps or overlaps is called a ''random tiling.''

III. RANDOM TILING CHARACTERIZATION

1. Variance

A random tiling is characterized by the mean square deviation of the point distribution from the center of mass in orthogonal space. The variance is defined by

$$\Omega = \lim_{N\to\infty} \frac{1}{N}\sum_{j=1}^{N} \left| \mathbf{y}_j - \frac{1}{N}\sum_{i=1}^{N} \mathbf{y}_i \right|^2 = \lim_{N\to\infty} \frac{1}{N}\sum_{j=1}^{N} |\mathbf{y}_j - \mathbf{y}_0|^2. \tag{1}$$

N is the number of quasilattice sites and $\mathbf{y}_0$ is the average of the position vectors $\mathbf{y}_i$ of all dual quasilattice sites. The bigger the phason fluctuations are the larger is the disorder and therefore the random tiling parameter Ω. The value in the pure entropic random tiling ensemble ($T\to\infty$) amounts to 1.73 ± 0.01.[2]

2. Vertex frequency

The lattice points of a quasicrystal are characterized by their local environment. The number of rhombohedra adjacent to a lattice point in an icosahedral quasilattice varies from 4 to 20, the number of edges from 4 to 12. There are 24 ''canonical'' or ''allowed'' vertices,[22] but 5450 vertices may occur in a random tiling. In the three-dimensional rhombohedra tiling flips are possible which only change the frequency of vertices without introducing forbidden ones. This is in contrast to the Penrose tiling in two dimensions. If the degree of randomization has reached a certain level the number of forbidden vertices is going to rise rapidly.

3. Sheet magnetization

The frequency of the simpleton is 23.61% in the ideal rhombohedra tiling. The simpletons are arranged in two-dimensional layers perpendicular to the twofold symmetry axis. Due to the two possibilities to pack the rhombohedra into the simpleton there are two positions for the internal lattice point. The positions may be called ''up'' and ''down'' and attributed a spin value $S=\pm 1$.[6] In the ideal rhombohedra tiling all the spins in a certain layer carry the value $+1$ or -1 which permits the definition of a sheet magnetization of value 1 using a proper normalization. In a random tiling the sheets also exist, but the magnetization is reduced since the spins in one layer are not all aligned. The susceptibility is given by

$$\chi = \frac{1}{T}\langle N_D\rangle \left(\left\langle \frac{M^2}{N_D^2} \right\rangle - \left\langle \frac{M}{N_D} \right\rangle^2 \right).$$

N_D is the number of the dodecahedra and therefore the number of the ''spins'' in the patch. This equation replaces the more cumbersome definition used in Ref. 6.

IV. ENERGY MEASURES

A. Models of stability

Several models currently exist that try to explain the stability of quasicrystals. In the deterministic energy model the internal energy U represents the thermodynamically stabilizing factor. Microscopic forces lead to matching rules[12] or overlapping cluster energies[14,15] that favor an ideal quasicrystalline tiling. In the nondeterministic entropic model stabilized by the entropy S no matching rules exist but the cells of the tiling do not leave any gaps and do not overlap. This is the random tiling model. In between is the semientropic model which is described by a free energy with contributions from internal energy and entropy:

$$F(T) = U(T) - TS(T). \tag{2}$$

The purely energetic model can be regarded as a low-temperature limit, the purely entropic model as a high-temperature limit of the semientropic model.

B. Properties of energy measures

The ideal quasilattice without any violation of matching rules represents the ground state of the energy model, taken at $T=0$. At finite temperatures thermally activated flips exist, mediating the transition between neighboring states. The transition probability is given by the Boltzmann factor, which depends on the energy measure chosen.

Here we deal with the canonical random tiling ensemble. All configurations of the ensemble have the same volume and the same number of vertices due to a constant average slope of the de Bruijn hypersurface. The internal energy $U(T)$ is the ensemble average $U=\langle E\rangle$ of the instantaneous energy E. The specific heat $C_V(T)$ is derived from the variance of the occupied energy levels by the fluctuation-dissipation theorem

$$\langle(\delta E)^2\rangle = \langle E^2\rangle - \langle E\rangle^2 = k_B T^2 C_V = k_B T^2 \frac{\partial U}{\partial T}.$$

The temperature dependence of the entropy density $s(T)$ (precisely: entropy per quasilattice site where large letters indicate total quantities, and small letters denote a quantity per vertex. For periodic approximants with rhombohedral cells the number of cells and vertices is identical) is given by the thermodynamical integration of the specific heat c_V per vertex:

$$s(T)=s_V(T)+\int_0^T dT' \frac{c_V(T')}{T'}+s_0 . \tag{3}$$

The volume contribution $s_V(T)$ vanishes in our case. The ground state entropy is $s_0:=s(T=0)$, and at $T\rightarrow\infty$ we are in the limit of the pure random tiling model with the configuration entropy $s(T=\infty)=:s_\infty$. The temperature variations of u, s, and c_V depend on the energy measure, but not s_∞. If one is interested only in the configuration entropy s_∞, then the specific choice of the energy measure has no physical relevance if the first three of the following four conditions are fulfilled.

(1) The energy of any configuration is unique.

(2) The quasiperiodic reference tiling is a ground state.

(3) The energy measure is limited from above.

(4) The entropy of the ground state vanishes or is easy to calculate.

The first condition requires an exact law to determine the energy of any microstate. The energy of a fixed microstate must not depend on how it is generated. Two configurations that differ only by a rigid translation have to be energetically identical.

Especially global energy measures are endangered to violate the second condition and to render the quasiperiodic reference state unstable at $T=0$ (see below for the ''quadratic energy measure''). The third condition is a requirement for the integrability of $c_V(T)/T$ as a function of T. If the energy measure is not limited, uncontrolled fluctuations of the energy in the high-temperature limit may exist. If they vary stronger than quadratic with T then

$$\frac{\langle(\delta E)^2\rangle}{T^2}$$

diverges and with it c_V as $T\rightarrow\infty$.

The fourth condition is recommended for practical purposes. The ground state entropy s_0 represents the number of states energetically equivalent to the quasiperiodic ground state. Since it is often complicated to calculate the ground state entropy it is useful to choose an energy measure where s_0 vanishes or is easy to deduce. An example of a complicated case of nonvanishing contribution s_0 has been presented by Baake and Joseph[16] for a two-dimensional octagonal quasicrystal. The locally defined energy measure is 0 for the canonical vertices but positive for forbidden vertices and depends on the vertex type. Unfortunately there exist configurations which are not quasiperiodic, although they do not exhibit forbidden vertices. As a consequence 44% of the total entropy are contributed by the ground state.

An example for a globally defined energy measure with an easily calculable ground state entropy is the ansatz of a harmonic oscillator potential in the orthogonal space as applied in this work. The energy is the sum of the squared distances of the dual quasilattice sites from their center of mass. Up to a factor N it is equal to the variance in the orthogonal space (see Sec. III 1). The precise definition of the energy of the configuration α is

$$E^{(\alpha)}=C\left|\sum_{j=1}^{N}\left|\mathbf{y}_j^{(\alpha)}-\frac{1}{N}\sum_{i=1}^{N}\mathbf{y}_i^{(\alpha)}\right|^2 -\sum_{j=1}^{N}\left|\mathbf{y}_j^{(0)}-\frac{1}{N}\sum_{i=1}^{N}\mathbf{y}_i^{(0)}\right|^2\right|. \tag{4}$$

The index 0 denotes the ideal reference configuration, C is an arbitrary normalization constant.

This energy measure is called the *harmonic energy measure*. The variance for the ideal reference configuration 0 is smaller than the variances for the overwhelming majority of the random tiling configurations α. But there is a tiny minority of configurations with a variance smaller than the value of the ideal tiling. Their atomic hypersurfaces are closer to a sphere than the triacontahedron. Such configurations may play a role at very low temperatures. To avoid energies less than the energy of the ideal tiling we have taken the absolute value in Eq. (4). For zero global phason strain in an ideal tiling this energy measure is not degenerate and thus the second condition is obeyed. But for periodic approximants the N possibilities (N: number of lattice points) to chose the origin of the unit cell yield a ground state entropy $s_0=\ln N/N$ per lattice point. It is obvious that s_0 vanishes in the thermodynamic limit. Strandburg[3] has introduced a similar energy measure called the *quadratic energy measure*

$$E^{(\alpha)}=\sum_{j=1}^{N}|\mathbf{y}_j^{(\alpha)}-\mathbf{M}_{\mathrm{Trial}}|^2. \tag{5}$$

But it was taken relative to $\mathbf{M}_{\mathrm{Tria}}$, the initial center of mass of the triacontahedron in the orthogonal space and not relative to the instantanous center of mass and therefore does not fulfil the criterium of finiteness of the energy measure (third condition). Actually $\mathbf{M}_{\mathrm{Tria}}=0$ was set to zero. Without fixed boundaries of the system the whole distribution of the dual quasilattice points in the orthogonal space may drift and therefore yield a systematic contribution to the energy and is therefore unphysical. In the harmonic energy model this problem has been resolved since now the energy is based on the difference to the center of mass.

An example of a locally defined energy measure is the one that counts the violations of the alternation condition. This condition requires that along a stack of rhombohedra two rhombohedra of the same type and orientation do not occur subsequently. This energy measure was used by Dotera and Steinhardt[6] and Gähler.[11] Other examples are the ''simple energy model'' which assigns the same energy to each forbidden vertex, the ''cluster energy model'' which maximizes the frequency of certain favorite vertices and the ''Tübinger mean-field model.'' All three were used by Joseph in Ref. 10.

The harmonic energy measure provides an excellent model for a quasicrystal, due to the following reasoning: the energy difference of a single jump is

$$\Delta E=E^{(\alpha+1)}-E^{(\alpha)}=CN(|\Omega^{(\alpha+1)}-\Omega^{(0)}|-|\Omega^{(\alpha)}-\Omega^{(0)}|).$$

If the temperature is not too low then the differences are positive and we get

$$\Delta E = CN|\Omega^{(\alpha+1)} - \Omega^{(\alpha)}|.$$

The shift of the center of mass $\mathbf{y}_{cm}^{(\alpha)} = 1/N \Sigma_{i=1}^{N} \mathbf{y}_i^{(\alpha)}$ for a jump of vertex k is

$$\mathbf{y}_{cm}^{(\alpha+1)} - \mathbf{y}_{cm}^{(\alpha)} = \frac{1}{N}(\mathbf{y}_k^{(\alpha+1)} - \mathbf{y}_k^{(\alpha)}) = \frac{\Delta}{N}.$$

$\Delta = 2.753$ is the jump distance in units of the rhombohedra edge length. Since N is large even for rather small samples the shift may be neglected. Thus the energy difference caused by a single jump reads

$$\Delta E = C(\mathbf{y}_k^{(\alpha+1)} - \mathbf{y}_k^{(\alpha)} - 2\mathbf{y}_{cm}) \cdot \Delta \mathbf{y} = 2C(\mathbf{y}_{ck} - \mathbf{y}_{cm}) \cdot \Delta \mathbf{y}.$$

$\mathbf{y}_{ck}$ is the midpoint of the connection between the inital and final vertex position and $\Delta \mathbf{y}$ the respective difference vector. The $\cdot$ represents the scalar product of $\Delta \mathbf{y}$ and the site vector $\mathbf{y}_{ck} - \mathbf{y}_{cm}$. Physically this means that if a vertex jumps *towards* the center of mass, its energy is *negative*. The jump is always accepted, and the randomness is reduced. The same happens for a tangential jump, where the energy difference is zero. But if it jumps *away* from the origin, the energy *increases*, the disorder also increases, and the jump is only accepted within the Boltzmann factor. Thus this energy model favors an ordered quasicrystal, but zero energy modes are allowed. The jump energies are distributed continuously and increase with the distance from the center of mass of the acceptance domain. In a local energy model the energy differences are independent of the position and have one fixed value if the matching rule is violated.

C. Pair interactions

All energy measures discussed in Sec. IV B contain a certain kind of arbitrariness since they do not depend on interatomic interactions but on the assignment of an energy penalty to randomized lattice configurations. Here we present test interactions which are motivated by pairwise interactions between atoms sitting on certain sites of the quasilattice (called ''decoration''). We test whether the energetic equidistribution postulated in the random tiling model is justificable.

The potential interactions used are the 12-6 Lennard-Jones potential modified by a cutoff function to guarantee a smooth behavior at the cutoff radius ($r_c = 4$ in units of rhombohedra edges). The potential energy u can be directly calculated from the radial distribution function in the case of pair interactions $v(r)$:

$$u = \sum_{r=0}^{r_c} 4\pi r^2 g(r) v(r). \tag{6}$$

The results of the test are presented in Sec. VIII B 1. It turns out that different random tiling samples do not show a significant difference in their internal energy u. The reason is the following: The distances between the vertices or atoms, if the quasicrystal is decorated, can be subdivided into two classes. The distances within a rhombohedron, on its faces, and along its edges are only permuted, therefore their frequency does not change at all. The frequency of distances between vertices in neighboring rhombohedra may change. Since all distances between nearest neighbor vertices or atoms and also most of the distances between next-nearest neighbors belong to the first class, the radial distribution function is nearly unchanged for the short distances. Since pair interactions are strong between nearest neighbors a change in potential energy is also expected to be small. Furthermore, the potential is weak at larger distances, where the changes of the radial distribution function are stronger.

V. CONSTRUCTION OF RANDOM TILINGS

A. Monte Carlo method

Simpleton flips directed in their frequency by the Monte Carlo method allow the generation of equilibrium configurations at a given temperature. The randomization of the quasilattice is realized by a sequence of such flips which usually are associated with a change in energy. The flips play the role of elementary transition paths between different states of a system in contact with a heat bath.

We are assuming that the method is ergodic.[17] Although this has not yet been proven, there is no hint up to now of the opposite. The method has already been used extensively to generate and study equilibrium random tiling quasicrystals. It is the only one known that produces equilibrium ensembles.

A simpleton flip does not generate or destroy tiles, it only rearranges them. Therefore the frequency of both types of rhombohedra in the tiling is remaining constant. The average orientation of the hypersurface and the global phason strain are not changed.

During the Monte Carlo simulation lattice points are selected at random. If a simpleton vertex is hit, the energy of the original and the flipped configuration are compared. If the energy decreases, the flip is always carried out. If it increases, it is performed with a probability of $\exp(-\Delta E/k_B T)$.

B. Boundary conditions

Quasicrystals do not permit periodic boundary conditions. Hence one has to work with a finite patch of a quasicrystal. Then there are two possibilities to deal with the surface: either keep it fixed (fixed boundaries) or identify opposite sides (periodic approximants).

In the case of fixed boundaries the surface lattice points are not allowed to move whereas in approximants all surface lattice points are mobile. Therefore the configuration entropy per lattice point with fixed boundaries is smaller than the entropy of approximants. The configuration entropy of the approximant, on the other hand, differs only slightly from the value of the ideal tiling. The frequency of vertices also is not much different between an ideal quasicrystal and an approximant tiling. The frequency of the simpleton, however, in patches with fixed boundaries may grow to up to 30% of the bulk vertices. For a discussion of the entropy dependence on boundary conditions see Ref. 18.

Periodic approximants show matching rule violations already in the ground state. They exhibit a periodic superstructure and for very small samples a remarkable deviation of the vertex frequencies. The intrinsic global phason strain changes the flip diffusion properties at low temperatures as the ground state phasons generate zero energy modes. The sheet magnetization (see Sec. III 3) in the case of a cubic approximant is useful only for the three twofold directions parallel to the cubic cell axis. The acceptance region is no longer homogeneously filled but has a lattice structure.

VI. RANDOM TILING TRANSITION AND STRUCTURE FUNCTIONS

Signals for a phase transition can be obtained from thermodynamic functions like u, c_V, and s. In a plot of the specific heat c_V vs temperature T for different sample sizes c_V should diverge in the case of a second order phase transition.

The Binder order parameter

The Binder order parameter[19] $B^{(N)}(T)$ for a given sample size N and temperature T is defined by

$$B^{(N)}(T):=1-\frac{M_4^{(N)}(T)}{3(M_2^{(N)}(T))^2},$$

where $M_2^{(N)}(T)$ and $M_4^{(N)}(T)$ are the second and fourth moment

$$M_k^{(N)}(T)=\int d\mu(p(\mu)^{(N)}(T))\mu^k$$

of the probability $p(\mu)$ of a microstate of the sheet magnetization μ belonging to a macrostate with sheet magnetization M. The Binder order parameter $B^{(N)}(T)$ is plotted as a function of T and yields a set of curves parametrized by N. A unique intersection point of the curves points to a second order phase transition and yields the transition temperature.

VII. SIMULATION PROCESS AND MODELS STUDIED

Thermodynamical quantities are sampled by storing their values at regular intervals of length Δt during the simulation. Data of microconfigurations are stored every $1000\cdot\Delta t$ Monte Carlo step. An initial configuration (e.g., an ideal Ammann-Kramer-Penrose approximant) was equilibrated over a typical thermalization time of 10^3 to 10^4 steps before the real simulation was started. The thermalization time was chosen according to the saturation behavior of the energy, for example. At low temperatures longer equilibration and simulation times and sampling intervals were used. The length Δt was checked by the decay of the autocorrelation functions. For high temperatures Δt was set typically about 50 Monte Carlo moves per lattice point, while for lower temperatures 200 moves were taken due to temporal correlations. The entire duration of a simulation run at given temperature typically has been about $25000\cdot\Delta t$ to $30000\cdot\Delta t$ simulation steps.

A. System sizes and interaction types

For the simulations we have prepared cubic approximants with 136, 576, 2440, 10336, and 43784 points. These correspond to generation $n=3$ to 7. Further simulations to fill the gaps between the sizes have been carried out with fivefold approximants with 890 and 1440 lattice points. The influence of the boundary condition has been analyzed with finite patches containing 4403 lattice points and 3507 cells. The influence of the initial conditions has been checked by using ideal quasicrystals or nonequilibrium random tiling samples as starting points. Both samples types lead to the same ensemble averages if equilibrated correctly.

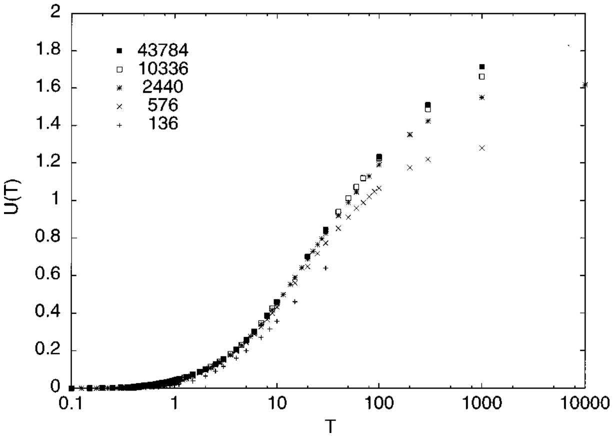

FIG. 2. Internal energy for the harmonic energy model. The sizes of the samples are indicated by the number of quasilattice points.

VIII. RESULTS

A. Thermodynamic functions

1. Internal energy, specific heat, entropy

The internal energy grows monotonously, and a clear saturation becomes visible (Fig. 2). The saturation value depends on the size of the sample. The limit value can be derived from limits of the variance: $\Omega(T=\infty)=1.73\pm0.01$ (Ref. 2) leads to u between 1.97 and 2.05. The specific heat shows an anomaly typical for few-level systems. It has an additional bump above the maximum (Fig. 3). Such a behavior is known for example for three-level systems with sufficiently separated levels. We have mapped the distribution of the energy levels for $T=\infty$ assuming equidistribution of the

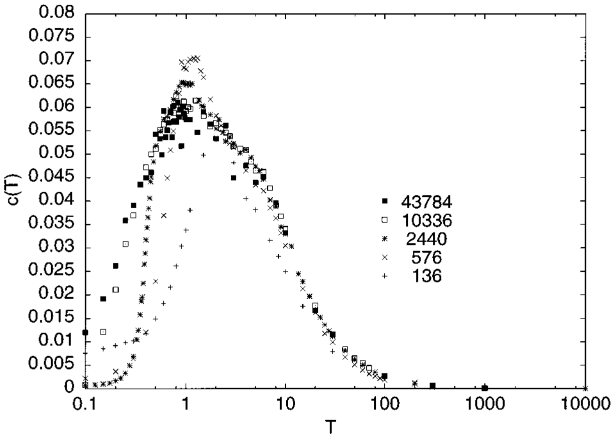

FIG. 3. Specific heat of the harmonic energy model. The sizes of the samples are indicated by the number of quasilattice points.

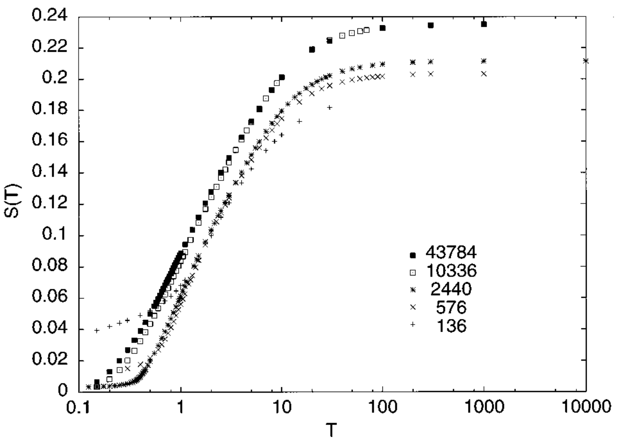

FIG. 4. Configurational entropy of the harmonic energy model. The sizes of the samples are indicated by the number of quasilattice points.

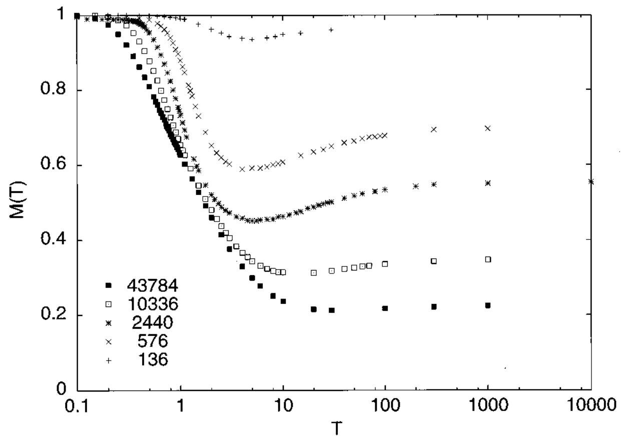

FIG. 5. Sheet magnetization of the harmonic energy model. The sizes of the samples are indicated by the number of quasilattice points.

occupancy. No indication of discrete energy levels was found, only an asymmetry of the distribution with a smaller slope at higher energies could be observed. There are other energy measures[11] which exhibit no visible asymmetry in the energy distribution and no bump in the specific heat. The asymmetry, however, is in our opinion not a clear explanation for the bump.

The value of the maximum of c_V is not significantly dependent on sample size. The ground state entropy vanishes for large N. The increase in s_∞ is caused only by the growing width of maximum of c_V (Fig. 4). The entropy values at $T=\infty$ are listed in Table I. Since no divergence occurs with increasing sample size, we conclude that there is no second order phase transition.

Further simulations with fixed boundaries and the cubic energy measure show that the internal energy also saturates although the energy measure is unlimited. (The cubic energy measure is defined as the quadratic energy measure [Eq. (5)], except that the exponent 2 is replaced by 3.) The entropy at very large temperature approaches $s_\infty \approx 0.1194 \pm 0.015$, lower than the value for periodic boundary conditions. The reason for both observations are the fixed boundaries which reduce the number of available configurations. If the quadratic energy measure is used with periodic boundary conditions, it is not clear whether the internal energy saturates since the energy measure is again unlimited. The decay of the specific heat is slower which is also a consequence of the energy measure. The entropy at very large temperatures approaches $s_\infty \approx 0.2621 \pm 0.015$.

TABLE I. Some thermodynamic parameters for the harmonic interaction model with periodic boundary conditions. The first column provides the generation for cubic approximants, the second column the number of lattice points, and the following columns list the entropy s_∞ and the variance $\Omega(T=\infty)$. The rows ''890'' and ''1440'' denote pentagonal approximants.

n	Size	s_∞	$\Omega(T=\infty)$
3	136	0.1816878	1.475 ± 0.015
4	576	0.2032964	1.56 ± 0.01
	890	0.2086351	1.60 ± 0.02
	1440	0.2076728	1.615 ± 0.02
5	2440	0.2114427	1.635 ± 0.01
6	10336	0.2349887	1.675 ± 0.01
7	43784	0.2367582	1.70 ± 0.01
∞	Limit	0.24 ± 0.01	1.73 ± 0.01

2. Simpleton magnetization, simpleton susceptibility, Binder order parameter

The temperature dependence of the energy fluctuations and the specific heat do not clearly rule out a phase transition, even if there is not a divergence of the maximum of c_V. The reason may be that the intrinsic divergence of the specific heat with sample size, if any, is very weak. For further insight we calculated the sheet magnetization M and the susceptibility χ since the latter may show a much more pronounced divergence behavior.

The magnetization saturates at high T, and the minimum value decreases with size (Fig. 5). At higher temperatures between $T=3$ and 10 it increases again. We attribute this behavior to the finite size of the samples. The value of the maximum of the susceptibility grows almost linearly with the generation n and moves to lower T (Fig. 6). It is not certain that the relation $\chi_{\max}(n) \propto (n-n_0)$ is valid for $n>7$. If

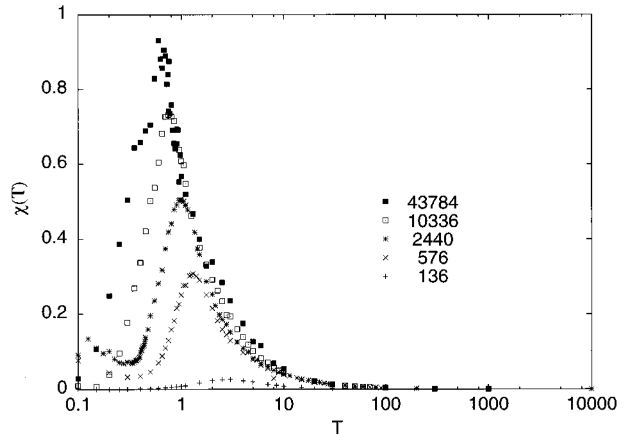

FIG. 6. Susceptibility of the sheet magnetization of the harmonic energy model. The sizes of the samples are indicated by the number of quasilattice points.

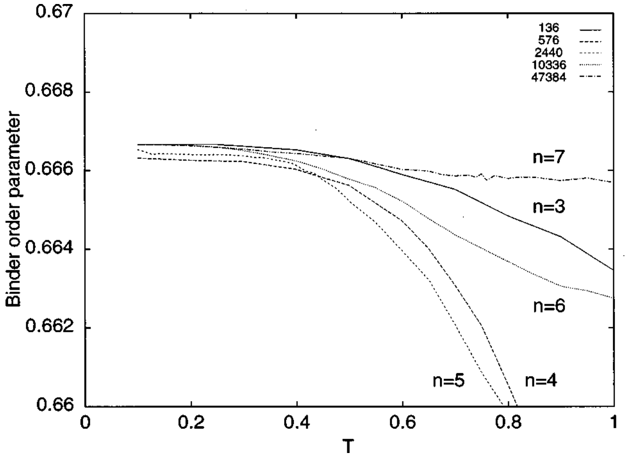

FIG. 7. Binder order parameter for the sheet magnetization of the harmonic energy model. The sizes of the samples are indicated by the number of quasilattice points and the generation parameters n.

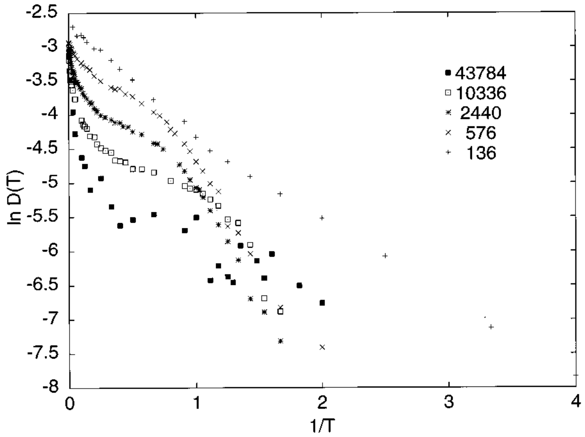

FIG. 8. Self-diffusion coefficient of the harmonic energy model. The sizes of the samples are indicated by the number of quasilattice points.

yes, this would be a slow divergence [more precisely: $\chi_{\max}(N)$ is about proportional to $\tau\sqrt[3]{N}$ where N is the number of lattice cells and $\tau=(\sqrt{5}+1)/2$].

The Binder order parameter clearly shows that there is no intersection point (Fig. 7) and therefore no phase transition at finite temperatures takes place. The trend of $B^{(N)}(T)$ changes at $n=5$, similar to the magnetization behavior. For matching rules energy models a phase transition between the locked phase, where the matching rules are obeyed, and the unlocked phase, which is a random tiling, have been predicted by Henley[4] and confirmed by Dotera and Steinhardt[6] and Gähler.[11] From the standpoint of this phase transition type the harmonic energy model is neither locked nor unlocked. The mean value of the Hamiltonian does not correspond to a spontaneous symmetry breaking, rather it is set by an ''external field'' $-V\Sigma_i|\mathbf{y}_i-\mathbf{y}_0|^2$ [see also Eq. (1)]. An analogy is a ferromagnet which is kept in a constant magnetic field. Because of the field, there is never a sharp symmetry breaking or a sharp qualitative change in the state; if it is cooled down from high temperature, there is always some magnetization, and at $T=0$ a particular state determined by the applied field is reached.

3. Self-diffusion coefficient

The mean square displacement $\langle(\mathbf{r}(t)-\mathbf{r}(0))^2\rangle$ grows linearly with time t, indicating a normal diffusion behavior and allowing the calculation of the diffusion coefficient D. At temperatures lower than $T\approx 1$ the extrapolation is difficult due to large fluctuations. D becomes unmeasurably small below $T=0.5$. The diffusion coefficient forms a plateau at $T\approx 1$ for $n=4,5,6$ in the Arrhenius plot (Fig. 8). There may be several reasons for this behavior.

First, there are energy barriers between different tiling configurations due to the harmonic energy measure, which at low temperatures lower the mobility of lattice points for flips of higher energy. In the range of the plateau the probability for a flip only occasionally suffices to overcome the barriers which play no role at high temperatures.

Second, a phase transition may occur which changes the slope in the Arrhenius plot. Gähler has also observed a change of the slope in case of the energy measure with the alternation condition which turns out to be a phase transition since other response functions like the susceptibility and the specific heat definitely yielded a divergence at the same temperature.

Last, there is an explanation which comes from the fact that the number of flippable lattice points (number of simpletons) changes with temperature. We can distinguish four ranges.

(1) The number of simpletons is about 23% in the range $0\lesssim T\lesssim 1$.

(2) In the range $1\lesssim T\lesssim 10$ we find a nearly logarithmic decrease of the number of simpletons. The plateau of $D(T)$ is clearly seen here.

(3) In the range $10\lesssim T\lesssim 100$ the approach of the frequency of the simpleton to a constant value leads to an increase of the negative slope of $D(T)$.

(4) Above $T\gtrsim 100$ the number of simpletons is constant at $\approx 17.5\%$.

In the approximants the behavior of $D(T)$ is obscured to some degree by zero energy modes caused by periodic boundaries. These modes become less and less important at larger sizes, but suppress the plateau for small sample sizes.

Within the framework of the random tiling model we have studied only flip diffusion. No other diffusion mechanism, in particular vacancy diffusion, can be introduced in this way. The latter mechanism is expected to be the dominant diffusion process at least above 600 °C. This would mean that quasicrystals behave like crystalline metals and alloys. But there are experimental indications that the new mechanism plays a role below 600 °C temperature,[21] since there is a sharp kink at about 400 °C and a smaller slope of the Arrhenius law at low temperatures.

Since we have no possibility to relate the experimental temperature scale and the scale of the Monte Carlo simulations we do not know where or if the plateau observed in our simulations is found in nature. We do not know either if the reduction of the number of flippable vertices plays a role in experiment. If we identify our low temperature part with the low temperature part of experiment, the plateau and the increased diffusion constant at high temperatures are hidden by the vacancy diffusion. But if we identify our high tempera-

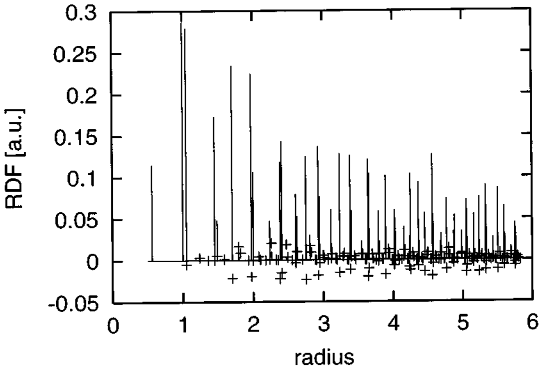

FIG. 9. Radial density function (RDF) of the rhombohedra quasilattice points for an approximant of size $n=5$. The peaks represent the values for the ideal tiling; the crosses denote the difference for the random tiling at $T=\infty$.

ture part with the low temperature part of experiment, we would expect that the diffusion behavior in a temperature range lower than currently measured becomes increasingly more complicated.

The diffusion behavior may also be rather complex if the quasicrystal is a random tiling and a phase transition to the perfect quasicrystal occurs at finite temperatures. This would cause a second kink in the Arrhenius law. But if the transition is at zero temperature as in our model, no change in slope should be observed.

B. Structure functions

1. *Radial density function, radial structure factor, pair interaction energy*

Structure functions have been calculated for samples of size $n=6$, containing 10336 lattice points and for the binary decoration with 54120 atoms.[20] There are only small changes in the partial radial density function $g(r)$ of the binary tiling. The largest differences of about 12% occur for bonds between two large atoms, but they are the least significant ones for stability due to their small number. The energetically most important nearest-neighbor bonds change less in all cases, since a large portion of them lives *within* the rhombohedra and thus is only transferred to another place by a flip, but the frequency itself does not change. The differences we observe in nonequilibrium random tilings are somewhat larger, but still very small. Figure 9 shows the radial distribution function for the monatomic sample (vertices occupied only) of size $n=5$. The maxima are the different atom shells, the crosses denote the changes for a random tiling at $T=\infty$. It is obvious that the changes at small distances are small.

The radial structure factor $I(k)$, the Fourier-transform of $g(r)$, shows a trend similar to the radial density function. Due the smallness of the samples there are rather large finite size effects which prevent quantitative comparison.

We further observe a very small dependence of the pair energy function [see Eq. (6)] on size and T due to the relation between the pair energy and the pair distribution function. It is interesting to note that the potential energy of the random tiling ensemble is at $T=\infty$ about 0.4% lower than for the ideal quasicrystal for a large class of Lennard-Jones-like potentials, whereas the fluctuations within the random tiling ensemble are of the order of 0.1%. Therefore simple pair potentials obviously favor energetical equidistribution, i.e., the random tiling model.

2. *Variance, vertex statistics*

The alteration of the variance is roughly linear with respect to the internal energy u. This is obvious if one compares their definition in Eqs. (1) and (4). Their temperature variation is therefore also similar.

The change in the statistics of the vertices with temperature is much more important as we have seen in the discussion of the diffusion behavior in Sec. VIII A 3. The changes of the vertex frequencies (notation as in Ref. 22) can be summed up as follows.

(1) The simpleton vertex 1 (452) decreases from 23.61% to 17.5%.

(2) Vertex 2 (561) increases from 23.61% to 29% at $T=8$ and then decreases to 26%.

(3) Vertex 3 (670) decreases from 23.61% to less than 10%.

(4) The sum of frequencies of vertex 1 and vertex 2 is roughly constant.

(5) The forbidden vertices (all together) increase from 0% to more then 40%.

(6) The vertex with full icosahedral symmetry and 20 adjacent prolate rhombohedra (vertex 24, in Ref. 22 also called twelvefold site) decreases in frequency from 1.2% close to extinction.

We note that the statistical error of the vertices statistic is largely independent of the sample size and already for $n \geqslant 4$ very small.

Vertex 1 is the most important one since it is the only vertex that can be flipped. But the change of vertices 2 and 3 is also not negligable, since these two vertices are neigbors of vertex 1 and are generated or destroyed by a flip. Since the total frequency of 1 and 2 is roughly constant, but the frequency of vertex 3 decreases with increasing temperature, this further contributes to a reduction of the diffusion constant described in Sec. VIII A 3.

3. *Diffraction patterns*

Diffraction patterns (Bragg scattering without the diffuse part characteristic for random tilings) have been calculated for a sample of size $n=5$, containing 2440 lattice points. The analysis has been carried out for several temperatures up to infinity for planes perpendicular to two-, three-, and fivefold direction. The intensities are only weakly dependent on temperature. The reason is that acceptance domain acts as a filtering function which changes only weakly with temperature. The result is consistent with Tang's observation of limited phason fluctuations in three dimensions.[2]

IX. CONCLUSIONS

We have studied the properties of the semientropic random tiling model for the harmonic energy measure. The results are summarized as follows.

(1) The sheet magnetization decreases to a minimum with

the temperature and slightly grows to a saturation value. In the thermodynamic limit it should vanish without an intermediate minimum. The susceptibility diverges slowly, the maximum shifts to smaller temperatures. The Binder order parameter does not exhibit a unique intersection point. This behavior indicates a phase transition already at $T=0$.

(2) The self-diffusion coefficient displays a plateau in the central temperature range indicating energy barriers for certain flips due to the harmonic energy measure. On the other hand there exist correlations between the temperature dependence of the self-diffusion coefficient and the frequencies of simpletons per lattice point. The Arrhenius plot is deviating strongly from that assumed by Kalugin and Katz:[7] where these authors are plotting a steep increase, we are observing the plateau. This is due to the fact that there is only one flippable vertex type in our model and its frequency *must* decrease. The result is a reduction of the diffusion vehicles, a possibility not taken into account by Kalugin and Katz.

(3) Radial structure functions depend only weakly on the configuration. This is due to the rigidness of the cells. Pair interactions realize an equidistribution of all configurations—a possible realization of the pure entropic random tiling model.

The behavior of c_V and χ obviously depends strongly on the type of energy measure used. The alternation condition seems to work much better[11] — maybe as consequence of its closer similarity with Ising interaction models in comparison with the harmonic energy measure. However, for the simple energy model[10] it was also not possible to decide if a phase transition occurs, since no divergence of the specific heat could be observed.

ACKNOWLEDGMENTS

The authors are very indebted to Franz Gähler and Dieter Joseph for helpful discussions. Chris Henley has contributed the example with the ferromagnet.

---

[1] V. Elser, Phys. Rev. Lett. **54**, 1730 (1985).

[2] L. H. Tang, Phys. Rev. Lett. **64**, 2390 (1990).

[3] K. J. Strandburg, Phys. Rev. B **44**, 4644 (1991).

[4] C. L. Henley, in *Quasicrystals - The State of the Art*, edited by D. P. DiVincenco and P. J. Steinhardt (World Scientific, Singapore, 1991), p. 429.

[5] W. Ebinger, Diploma thesis, Stuttgart, 1991.

[6] T. Dotera and P. J. Steinhardt, Phys. Rev. Lett. **72**, 1670 (1994).

[7] P. Kalugin and A. Katz, Europhys. Lett. **21**, 921 (1993).

[8] M. V. Jarić and E. Sørensen, Phys. Rev. Lett. **73**, 2464 (1994).

[9] M. V. Jarić and E. Sørensen, in *Proceedings of the 5th International Conference on Quasicrystals*, edited by C. Janot and R. Mosseri (World Scientific, Singapore, 1995), p. 363.

[10] D. Joseph, in *Proceedings of the 5th International Conference on Quasicrystals* (Ref. 9), p. 605.

[11] F. Gähler, in *Proceedings of the 5th International Conference on Quasicrystals* (Ref. 9), p. 236.

[12] N. G. de Bruijn, Kon. Akad. Wetensch. Proc. Ser. **A84**, 38 (1981).

[13] C. L. Henley, J. Phys. A **21**, 1649 (1988).

[14] P. Gummelt, Geometriae Dedicata **62**, 1 (1996).

[15] H. C. Jeong and P. J. Steinhardt, Nature (London) **382**, 431 (1996).

[16] M. Baake and D. Joseph, Phys. Rev. B **42**, 8091 (1990).

[17] F. Gähler (private communication).

[18] D. Joseph and M. Baake, J. Phys. A **29**, 6709 (1996).

[19] K. Binder and D. W. Heermann, *Monte Carlo Simulations in Statistical Physics* (Springer, Heidelberg, 1998).

[20] J. Roth, R. Schilling, and H.-R. Trebin, Phys. Rev. B **41**, 2735 (1990).

[21] R. Blüher, Diploma thesis, Stuttgart, 1997; R. Blüher, P. Scharwaechter, and W. Frank, Phys. Rev. Lett. **80**, 1014 (1998).

[22] C. L. Henley, Phys. Rev. B **34**, 797 (1986).