Equilibrium behavior of the tiling model

Somendra M. Bhattacharjee and Eugene Helfand
AT&T Bell Laboratories, Murray Hill, New Jersey 07974-2070
(Received 31 March 1987)

Equilibrium properties of the tiling model, recently introduced by Stillinger and Weber as a means of studying glass phenomena, are investigated. In the two-dimensional model, a square lattice is covered by tiles of all sizes. The tiles represent domains of well-packed particles and the boundaries between the domains have a positive mismatch energy proportional to the length of the wall. The existence of the thermodynamic limit for this model is proved. It is shown how to obtain bounds for the free energy and the transition temperature from the free energy of semi-infinite strips. A transfer-matrix method is developed for calculating the thermodynamic properties of such semi-infinite strips. The best bound obtained is $\lambda/k_B T_c \geq 0.2459$ from a $9\times\infty$ strip, where λ is the basic energy parameter in the problem. By extrapolation, the transition temperature is estimated as $\lambda/k_B T_c = 0.27002$. By direct counting of states for finite squares, the infinite-temperature entropy is obtained by extrapolation as $s_\infty = 0.314$. A connection between the tiling model and electrical networks is discussed in Appendix A.

I. INTRODUCTION

A tiling model has recently been proposed by Stillinger and Weber[1-3] as an example of a system with glasslike dynamical properties.[4] It is assumed that any amorphous structure can be divided into domains of various sizes, the interiors of which contain only well-packed particles such as quasicrystals.[5] Restructing of these domains, as the temperature is changed, is taken to be the important physical process for the glass transition. The relevant energy parameter for the process is the domain wall energy. A simple statistical mechanical model should, therefore, focus on the packing problem of a collection of domains, with no internal energy or structure, the only energy being the wall energy. Such a model is *the Stillinger-Weber tilting model.*

Numerical simulations and mean-field theories have been used[1-3] to study the equilibrium and kinetic properties of the model in two dimensions. Remarkable similarities with the kinetic behavior of real glass systems have been observed.[2,3] In this paper we study the equilibrium thermodynamic behavior using analytical methods.

The model is defined in Sec. II where the known results of importance to the present work are also summarized. To put things in proper context, the aims and organization of this paper are given at the end of Sec. II.

II. THE STILLINGER-WEBER TILING MODEL

For simplicity, we restrict ourselves, throughout this paper, to two dimensions only. We adopt the notation where an $M\times N$ lattice refers to a square lattice with M unit cells in the x direction and N unit cells in the y direction. The lattice spacing is taken to be unity.

A. The model

The domains, alluded to in the Introduction, are represented by square tiles of all sizes. The tiles are supposed to cover an $N\times N$ square lattice completely without any gap or overlap. Figure 1 shows an example of such a tiling. The boundaries of the tiles (lying along the edges of the lattice) are the domain walls (lines in two dimensions). The states of the model consist of all possible distinct tilings of the lattice. The ground state has the smallest number of domain walls, and for a square lattice it is the tiling by a single $N\times N$ tile.

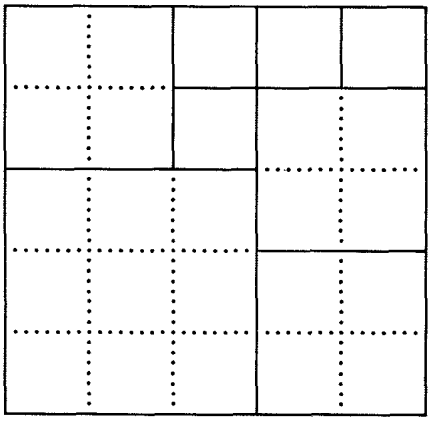

FIG. 1. A tiling of a 5×5 square. The tiles are shown by solid lines and the underlying lattice by the dotted lines. Each tile represents a domain of well-packed particles. The tiles do not have any internal structure. The only energy involved is the energy of the domain walls. The energy for this configuration is $E=26\lambda$.

Let us assign an energy $\frac{1}{2}\lambda$ (≥ 0) per unit length of the perimeter of each tile, i.e, an energy λ per unit length of interface.[6] For any configuration, the energy is given by

$$E=\tfrac{1}{2}\lambda\sum_j 4jn_j\ , \tag{2.1}$$

where n_j is the number of $j\times j$ tiles. (A $j\times j$ tile will, henceforth, be called a j-tile.) The ground state has an energy $2N\lambda$ but the energy per unit cell ($\sim N^{-1}$) vanishes in the thermodynamic of $N\to\infty$.

The thermodynamics is determined by the partition function

$$Z_{N,N}=\sum_{\text{states}}\exp(-\beta E)\ , \tag{2.2}$$

where the subscript of Z refers to an $N\times N$ lattice, with $\beta=1/k_BT$, k_B being the Boltzmann constant and T the temperature. At $T=0$, the system is in its unique ground state but at high temperatures, thanks to larger entropy, the equilibrium configuration consists of random packing of smaller tiles.

B. Known results

Mean-field theories, similar to the Flory-Huggins theory[7] of polymeric systems, predict[1,2] a first-order transition at a temperature T_c ($=1/k_B\beta_c$) below which the system is frozen in its ground state. The resulting free energy is shown in Fig. 2 (curve a). T_c has also been estimated from Monte Carlo simulations. These estimates are given in Table I which also gives the estimates for the infinite temperature ($\beta=0$) entropy per unit cell, s_∞. This entropy is related to the total number of partitions, Ω_{tot}, in the following way:

$$s_\infty=\lim_{N\to\infty}\left[\frac{1}{N^2}\ln\Omega_{\text{tot}}\right]\ . \tag{2.3}$$

From the time required to equilibrate the system at low temperatures ($\beta>\beta_c$), it has been inferred that β_c is also

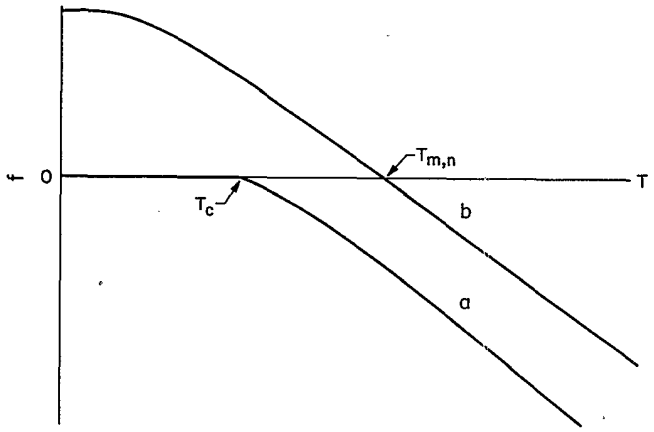

FIG. 2. The free energy per unit cell, in the thermodynamic limit, is shown schematically by curve a. The free energy is zero for $T<T_c$. T_c is the first-order transition temperature. Curve b is the schematic form of the upper bound $f_{m,n}$ (or f_m) obtained in Sec. III (or IV). $f_{m,n}$ (or f_m) is positive at $T=0$. $T_{m,n}$ gives an upper bound for T_c.

TABLE I. $\beta_c\lambda$ and s_∞.

	Flory[a]	Improved Flory[a]	Monte Carlo[a]	This paper
$\beta_c\lambda$	0.326	0.23	0.268–0.271	0.270 02
s_∞	0.244	0.299	0.3155	0.314

[a]Reference 2.

the "ideal" glass transition temperature. (We will be using T and β interchangeably.)

C. Aims and organization

A free energy of the type shown in Fig. 2 (curve a) is not uncommon[8] in statistical mechanics. In some cases, this behavior turns out to be an artifact[9] of the Flory-Huggins approximation. It is, therefore, important to prove or disprove the existence of a phase transition in the tiling model. This is one of the focal themes of this paper.

In Sec. III the existence of the thermodynamic limit is proved, and an upper bound for the free energy is constructed from the free energy of an $m\times\infty$ strip. This bound also gives a bound for T_c. In Sec. IV T_c is determined by extrapolation of a sequence of such bounds from different values of m. That such an extrapolation is meaningful is shown in Sec. III. The free energy of an $m\times\infty$ strip is calculated by a transfer-matrix method in Sec. IV. The cases of $2\times\infty$ and $3\times\infty$ are explicitly worked out in Sec. IV as examples of the technique. From the number of tilings of finite squares, s_∞ is estimated in Sec. V. Section VI is the summary and conclusion. In Appendix A a connection between the tiling model and electrical networks is point out.

III. BOUNDS FOR FREE ENERGY AND T_c

In this section we construct an upper bound for the free energy of an $N\times N$ lattice. As a corollary of this, we prove the existence of the thermodynamic limit. Furthermore, an upper bound for T_c is obtained.

A. Formulation

Let us divide the $N\times N$ lattice into smaller $m\times n$ blocks and tile each block independently, keeping the relative positions of the blocks fixed in the lattice. We will primarily be interested in the $n=N$ case, in the limit $N\to\infty$. For generality we keep n in the following formulation.

The configurations generated by the above construction form a subset of all the possible tilings of the $N\times N$ lattice. Therefore, at any T,

$$Z_{N,N}\geq(Z_{m,n})^{N^2/mn} \tag{3.1}$$

from which one has

$$f_{N,N}\leq f_{m,n}\ , \tag{3.2}$$

where the free energy per unit cell is defined by the relation

$$f_{m,n}=-\frac{k_B T}{mn}\ln Z_{m,n} \ . \tag{3.3}$$

1. Existence of the thermodynamic limit

Taking $m=n$, we have, from Eq. (3.2),

$$f_{2n,2n}\le f_{n,n} \tag{3.4}$$

for any n. Taking $n=2^p$ where p is a positive integer and writing

$$f^{(p)}=f_{n,n} \ ,$$

we can form *a monotonically decreasing sequence* $\{f^{(p)}\}$ of the free energy as the size increases. Note that $\{f^{(p)}\}$ is bounded above by $f_{2,2}$. It is shown in Appendix B that $f_{n,n}$ is *bounded below* by the free energy of the four-state Potts model.[10] Since a thermodynamic limit $(n\to\infty)$ is known[10] to exist for the Potts model, the sequence $\{f^{(p)}\}$ is bounded below. This proves that

$$f\equiv\lim_{n\to\infty} f_{n,n}$$

exists at all temperatures. The limiting free energy will be denoted by $f(T)$ without any subscript.

2. Bound on T_c

The inequality in Eq. (3.2) is maintained in the limit $N\to\infty$, because the right-hand side is independent of N. Therefore

$$f(T)\le f_{m,n}(T) \ . \tag{3.5}$$

[The T dependence was suppressed in Eqs. (3.1)–(3.4).] Since $f_{m,n}(T)$ is an analytic and monotonic function of T, with

$$f_{m,n}(T=0)=E_{\text{ground state}}>0 \ , \tag{3.6}$$

it will have a form shown in curve b of Fig. 2. If $f(T)$ is of the form of curve a of Fig. 2, then we must have

$$T_c\le T_{m,n} \tag{3.7a}$$

or, equivalently,

$$\beta_c\ge\beta_{m,n} \ , \tag{3.7b}$$

where $T_{m,n}$ (or $\beta_{m,n}$) is the temperature at which

$$f_{m,n}(T)=0 \ , \tag{3.8a}$$

$$Z_{m,n}(T)=1 \ . \tag{3.8b}$$

A simple physical interpretation can be given to the above construction. We are basically studying the stability of the ground state (in the limit $N\to\infty$), against small length scale fluctuations. $T_{m,n}$ is the temperature at which the entropic contribution from further subdivision of the $m\times n$ blocks (i.e., from fluctuations up to a length scale determined by the minimum of m and n) is balanced by the boundary wall energy of the blocks.

3. Example: $m=n=2$

As an example, we take $m=n=2$, so that

$$Z_{2,2}=x^{-1}(1+x^{-1}) \ , \tag{3.9}$$

where

$$x=\exp(4\beta\lambda) \ . \tag{3.10}$$

From (3.8b), $T_{2,2}$ is determined by the equation

$$x^2-x-1=0 \tag{3.11}$$

giving

$$\beta_c\lambda\ge\tfrac{1}{4}\ln\tau=0.1203\cdots \ , \tag{3.12}$$

where $\tau=(1+\sqrt{5})/2$ is the golden mean.

B. Monotonicity of the sequence of bounds

For simplicity, let us now restrict ourselves to the case $n=N$ with $N\to\infty$. Note that Eq. (3.5) is satisfied as an *equality* in the limit $m\to\infty$. If we can obtain the partition function $Z_{m,n}$ of $m\times N$ strips for different values of m, we will have a sequence of T_m's ($N\to\infty$ in the subscript is omitted) from which T_c can be extracted by extrapolation. We now argue that such an extrapolation is meaningful.

A straightforward generalization of Eq. (3.1) for a $2m\times N$ strip gives

$$Z_{2m,N}(T)>[Z_{m,N}(T)]^2 \ . \tag{3.13}$$

Since the partition function is a positive definite and monotonically increasing function of the temperature, the solution T_{2m} [see Eq. (3.8b)] of

$$Z_{2m,N}(T)=1 \tag{3.14}$$

is less than the solution of

$$[Z_{m,N}(T)]^2=1 \ ,$$

which, incidentally, is the equation determining T_m for the $m\times N$ strip. [Note that the monotonicity of the partition function guarantees that there is only one physical solution of Eq. (3.8b).] This shows that the subsequence $\{T_\nu\}$ where $\nu=2^p m$ (p an integer) is monotonically decreasing. One can, in fact, convince oneself that any such sequence with $\nu=j^p m$, for any positive integer p, is monotonic. Because of the existence of the thermodynamic limit, all of these subsequences will have the same limit as that of the whole sequence $\{T_m\}$. We will assume, as will be shown to be the case, that $\{T_m\}$ is also monotonic.

IV. BOUNDS FROM STRIPS: TRANSFER-MATRIX APPROACH

In this section we develop a transfer-matrix approach for calculating the free energy per unit cell of an $m\times N$ strip, in the limit $N\to\infty$,

$$f_m=\lim_{N\to\infty}\left[\frac{1}{mN}\ln Z_{m,N}\right] \ . \tag{4.1}$$

From Eq. (3.8a), $f_m=0$ will give a bound for T_c.

A. Generating functions

We define a new partition function $Q_{m,k}$ for an $m \times k$ strip as

$$Q_{m,k}(z,y)=\sum z^{\sum_j n_j} y^{-\sum_j j n_j} , \tag{4.2}$$

where $y=\exp(2\beta\lambda)$, z is the fugacity for tiles, and the summation is over all possible tilings. One can, in fact, introduce fugacities z_j for j-tiles and define

$$Q_{m,k}(\{z_j\},y)=\sum \left[\prod_j z_j^{n_j}\right] y^{-\sum j n_j} \tag{4.3}$$

from which the average numbers $\langle n_j \rangle$ of j-tiles can be calculated. Equation (4.3) reduces to Eq. (4.2) by setting $z_j=z$ for all j. Note also that

$$Z_{m,k}(T)=Q_{m,k}(z=1,y) . \tag{4.4}$$

A generating function $G(t,z,y)$ for $Q_{m,k}$ is now defined as

$$G(t,z,y)=\sum_{k=0}^{\infty} Q_{m,k}(z,y)t^k . \tag{4.5}$$

Thus function G can be evaluated by a transfer-matrix method as we show next.

B. Transfer matrix

In order to evaluate the partition function by a transfer-matrix approach, we must build up the tiling state in some unique, Markovian, and repetitious fashion (stationary Markoff process). To do so, we define certain intermediate states in the construction which we call *basis states* or just *states*. This is done in part 1. See also Figs. 3 and 4 for further explanations. The transfer matrix is constructed in part 2.

1. Basis states

We start building up the strip of width m from a straight horizontal line, at level zero, by adding tiles (of size 1×1 up to $m\times m$) with base on this horizontal line only, until level zero is full. A typical configuration achieved is illustrated in Fig. 3 for $m=10$. In this configuration, we identify the new *base line* as the lowest unoccupied level, level 1 in Fig. 3. The shape of the upper boundary at this point is called a *state* (renaming the base line level 0). We then add tiles to the base line only, until it is full. A new base line (at level 2) is identified (and renumbered zero) as the lowest unoccupied level, and the boundary shape is another state as illustrated in Fig. 3. This process is repeated w times, ending with a flat upper boundary. The intermediate states correspond to one or more specific types of tilings that achieve that state. Tilings corresponding to defined states must be such that removal of any tile lowers the base line level, i.e., the tile cannot be attached to the base line or a higher level.

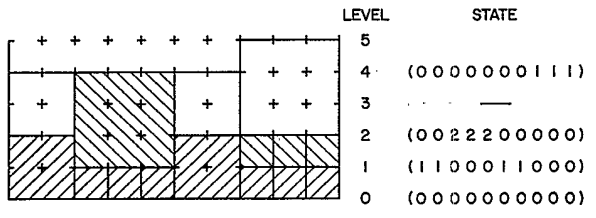

FIG. 3. Building up an $m\times\infty$ strip. $m=10$ for this figure. The underlying lattice points are indicated by $+$. Tiles shaded similarly are added at the same step. Note that the new base line corresponds to the lowest upper boundary of a tile added. The states produced are also given. There is no state at level 3.

It is easy to generate the states but not uniquely. The tiling of the uppermost level can be made of tiles of size $l_1,l_2,\ldots,l_r$ with $r\le m$, such that $\sum_{i=1}^{r} l_i=m$. There are $P_m=2^{m-1}$ such ordered partitions of the integer m. (See Appendix C for a proof.) Corresponding to each such partitions, there is a state such that a tile of size l_i may have its upper boundary at levels from 0 to l_i-1 above the base line. In this way, its lower boundary will be below the base line, as required by the definition. Also, at least one of the r tiles must have its upper boundary at the base line. Several such tile configurations may produce the same upper boundary shape as illustrated in Fig. 4. For $m=4$, all the possible upper tile configurations have been drawn. That figure also shows all the unique states.

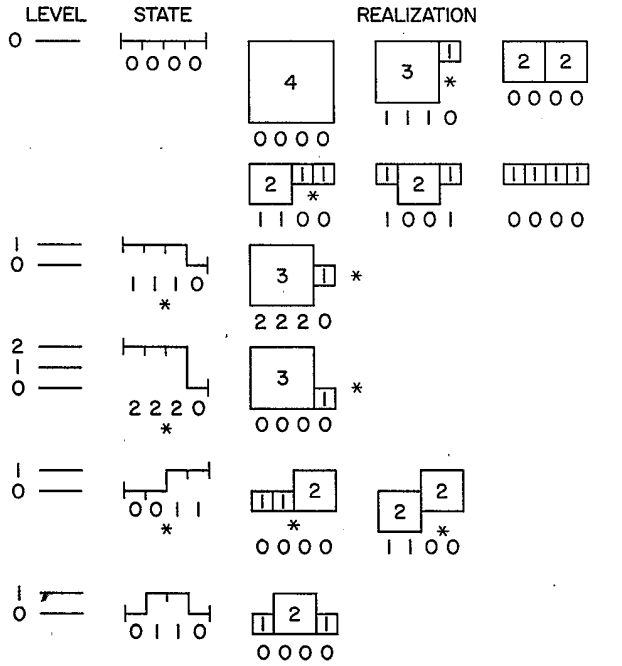

FIG. 4. All the basis states for $m=4$. The levels each state occupies are shown at the left. The possible tilings that can generate a particular state are shown on the right. The old state from which the state in question is obtained is also indicated. * indicates that another state (or tiling) is obtained by left-right inversion. For example, in addition to 1110 there is state 0111.

A basis state can be represented by explicitly stating the height of each lattice edge of the boundary above the base line with 0 representing the base line. Hence, the intermediate states for an $m \times \infty$ strip can be denoted by m integers $(a_1, a_2, \ldots, a_m)$ with $a_i \in [0, m-1]$ for each i. This notation is used in Figs. 3 and 4. We have not been able to obtain a general formula for the number of basis states B_m. Table II lists these numbers for sizes up to $m=10$.

2. Construction of the transfer matrix

The rule for adding new tiles at the boundary is to put them only on the base line, covering it *completely,* so that a new base line has to be defined. Figure 3 shows an example for $m=10$. It is easy to check that the set of basis states is closed under this rule of transformation.

In any state $|j\rangle$, if q_j is the number of zeros (or the total length of the base line), then the above rule guarantees that the total perimeter of all the tiles next added is $4q_j$. The Boltzmann factor associated with this addition, from Eq. (2.1), is y^{-q_j}, where, as before, $y=\exp(2\beta\lambda)$.

The elements V_{ij} of the transfer matrix $\underline{V}$ is associated with tilings that take state $|j\rangle$ to state $|i\rangle$ and is given by

$$V_{ij}=\left[\sum_{L\in(j\to i)} t^{k(L)} z^{n(L)}\right] y^{-q_j} , \tag{4.6}$$

where the summation is over the tilings L that take $|j\rangle$ to $|i\rangle$, $k(L)$ is the number of complete rows added by that tiling, and $n(L)$ is the number of tiles added. Recall that t and z are generating function variables introduced in Eqs. (4.5) and (4.2) to keep count of the total number of rows and tiles, respectively. Examples of $\underline{V}$ are given in Sec. IV D.

C. Connection to thermodynamic

1. Partition function and free energy

After w applications of $\underline{V}$, the partition function for all states with w intermediate base line is

$$\langle 0|\underline{V}^w|0\rangle ,$$

where $|0\rangle$ is the flat state (00...0). The number of intermediate base lines is *not* physically significant, but one can recover the generating function G of Eq. (4.5) by summing over w

$$G(t,z,y)=\sum_{w=0}^{\infty}\langle 0|\underline{V}^w|0\rangle$$

$$=\langle 0|(\underline{I}-\underline{V})^{-1}|0\rangle , \tag{4.7}$$

where $\underline{I}$ is the identity matrix. The partition function $Q_{m,k}$ is recovered as the coefficient of t^k by Cauchy's theorem as

$$Q_{m,k}=\frac{1}{2\pi i}\oint\frac{1}{t^{k+1}}\langle 0|\underline{A}^{-1}|0\rangle dt , \tag{4.8}$$

$$\underline{A}=\underline{I}-\underline{V} , \tag{4.9}$$

TABLE II. The number of basis states B_m.

m	2	3	4	5	6	7	8	9	10
B_m	1	3	8	18	42	97	223	506	1145

and the contour encloses the origin of the t plane and no singularities of $\langle 0|\underline{A}^{-1}|0\rangle$. To give each tiling equal weight, irrespective of the number of tiles, set $z=1$ in Eq. (4.8).

The contour of the Cauchy integral in Eq. (4.8) may be deformed out to infinity and the result for $Q_{m,k}$ written as an integral about the singularities of $\langle 0|\underline{A}^{-1}|0\rangle$. Since the elements of $\underline{A}$ are finite polynomials in t the singularities of $\langle 0|\underline{A}^{-1}|0\rangle$ are poles at the zeros of the determinant $|\underline{A}|$. Since we are interested in long strips, $k\to\infty$, the dominant contribution comes from the smallest t for which

$$|\underline{A}(t, z=1, y)|=0 ; \tag{4.10}$$

call it t_0. We assume that this is always real and positive, although we do not have a proof other than the requirements of the physics. Thus the free energy per cell, from Eqs. (4.1) and (4.4), is given by

$$f_m=\frac{k_B T}{m}\ln t_0 , \tag{4.11}$$

where t_0 is a function of y only.

2. Transition temperature

The bound for the transition temperature T_c, from Eq. (3.8a), is obtained by equating f_m to zero, which is equivalent to setting $t_0=1$. Therefore the bound for T_c comes from the equation

$$|\underline{I}-\underline{V}(t=1, z=1, y)|=0 . \tag{4.12}$$

Numerical results are presented in Sec. IV E.

3. Other thermodynamic properties

Other thermodynamic properties can be easily extracted from the transfer matrix. To do this, we introduce the spectral representation as

$$\langle 0|\underline{A}^{-1}|0\rangle=\sum_p\frac{\langle 0|p\rangle\langle p|0\rangle}{a_p(t,z,y)} , \tag{4.13}$$

where p labels the right and left eigenvector of A, $|p)$ and $(p|$, corresponding to eigenvalue a_p. The partition function is then determined by $t_0(z,y)$ which is the smallest t for which an eigenvalue $a_p=0$. Let p_0 be the label for that eigenvalue.

To determine the density of tiles (reciprocal of the average tile size), we use

$$\rho=\lim_{k\to\infty}\left[\frac{1}{km}\frac{\partial}{\partial z}\ln Q_{m,k}(z,y)\right]\Bigg|_{z=1}$$

$$=-\frac{1}{m}\frac{\partial}{\partial z}\ln t_0(z,y)\Bigg|_{z=1} . \tag{4.14}$$

The required derivative is easily evaluated by

$$\left[\frac{\partial t_0}{\partial z}\right]_{a=0} = -\left.\frac{(\partial a/\partial z)_t}{(\partial a/\partial t)_z}\right|_{t=t_0,\,z=1} \tag{4.15a}$$

and

$$\frac{\partial a}{\partial \nu} = -(p_0 \,|\, \partial \underline{V}/\partial \nu \,|\, p_0)\ , \tag{4.15b}$$

where ν is t or z.

The density of individual tile sizes can be ascertained by returning to a formalism with individual fugacities $z_1, z_2, \ldots$ in the transfer matrix, as mentioned earlier [Eq. (4.3)]. By varying these z_i's from unity the thermodynamics of a system at constant tile numbers can be determined. Since the energy is a unique function of these tile numbers, there is no temperature dependence but one can count the number of ways of tiling a lattice with n_1 1-tile , n_2 2-tile, etc.

The energy and specific heat are related to derivatives with respect to y, which again can be obtained from the spectral representation of $\underline{A}$. Details will not be given.

D. Examples

In this section we explicitly consider two cases: (1) $m=2$ and (2) $m=3$, as examples of the method developed so far.

1. $m=2$

For $m=2$, there is only one basis state, namely, 00. The transfer matrix is, therefore, a scalar. From Eq. (4.6), it is given by

$$V=(t^2 z_2 + t z_1^2) y^{-2}\ . \tag{4.16}$$

Here z_1 and z_2 are the fugacities for 1- and 2-tiles, which we retain separately because of the simplicity of this case.

The bound for T_c is obtained from Eq. (4.12) as

$$2y^{-2}=1 \tag{4.17}$$

This gives $\exp(2\beta_m \lambda)=2$ or

$$\beta_c \lambda \geq \tfrac{1}{4}\ln 2 = 0.173\cdots\ , \tag{4.18}$$

which is a remarkable improvement over Eq. (3.12), for a 2×2 square.

The full thermodynamics for the $2\times\infty$ strip can also be obtained from Eq. (4.16). The relevant zero of $1-V$ is

$$t_0 = \frac{[(z_1^4 + 4z_2 y^2)^{1/2} - z_1^2]}{2z_2}\ . \tag{4.19}$$

From Eq. (4.11), the free energy is obtained, by setting $z_1=z_2=1$, as

$$f_2 = \tfrac{1}{2} k_B T \{\ln[(1+4y^2)^{1/2}-1] - \ln 2\}\ . \tag{4.20}$$

If ρ_1 and ρ_2 are the average densities of 1- and 2-tiles, then

$$\rho_1 \equiv \frac{1}{2}\frac{\partial}{\partial z_1}(-\ln t_0)\bigg|_{z_1=z_2=1} = (1+4y^2)^{-1/2} \tag{4.21a}$$

and

$$\rho_2 \equiv \frac{1}{2}\frac{\partial}{\partial z_2}(-\ln t_0)\bigg|_{z_1=z_2=1} = [1-(1+4y^2)^{-1/2}]/4\ . \tag{4.21b}$$

Note that $\rho_1 \to 0$ and $\rho_2 \to \frac{1}{4}$ as $y\to\infty$ (i.e., $T\to 0$) as they should.

2. $m=3$

For $m=3$, the basis states are (i) 000, (ii) 011, and (iii) 110. Taking, as before, z_i as the fugacity for i-tiles ($i=1,2,3$), the transfer matrix is given by

$$\underline{V} = \begin{bmatrix} (t^3 z_3 + t z_1^3) y^{-3} & t z_1 y^{-1} & t z_1 y^{-1} \\ t z_1 z_2 y^{-3} & 0 & 0 \\ t z_1 z_2 y^{-3} & 0 & 0 \end{bmatrix}\ . \tag{4.22}$$

The bound for T_c is obtained from Eq. (4.12) as the solution of

$$y^4 - 2y - 2 = 0\ . \tag{4.23}$$

This equation has only one real positive solution at $y=1.4945\ldots$, giving the bound

$$\beta_c \lambda \geq 0.2009\cdots\ . \tag{4.24}$$

E. Sequence of bounds

The transfer matrices for $m=4$ to 9 have been generated on the computer and the zero of the corresponding determinant, Eq. (4.12), found numerically.

The results for $x=\exp(4\beta_m\lambda)$ are plotted against $1/m$ in Fig. 5 (set b). These points are fitted to a curve

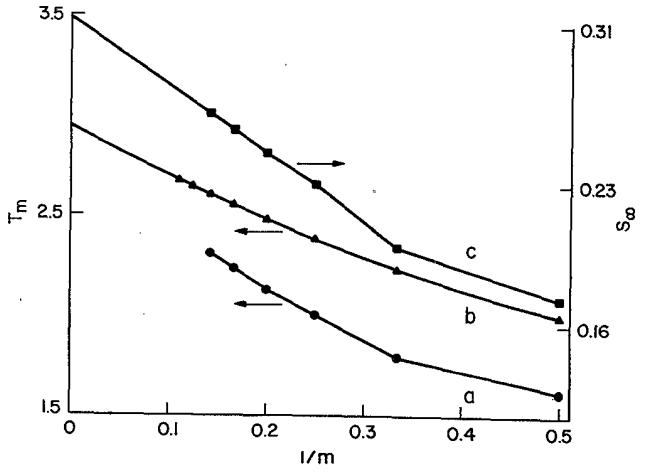

FIG. 5. $1/m$ extrapolation plots for T_c and s_∞. a, bounds $T_{m,m}$ on T_c from $m\times m$ squares from $m=2$ to $m=7$. b, T_m from $m\times\infty$ strips. The solid line is obtained by fitting Eq. (4.25). c, the entropy per unit cell, from $m\times m$ squares up to $m=7$. The intercepts in b and c on the y axis give values for the bulk.

$$x = a + \frac{b}{m} + \frac{c}{m^2} , \tag{4.25}$$

which gave $a=2.94496$, $b=-2.596$, and $c=1.411$. The fitted curve is shown in Fig. 5 by solid line b. The coefficient a gives the estimate for x, and hence for $\beta_c\lambda$, as $m \to \infty$. The value we find is

$$\beta_c\lambda = 0.270\,02 \cdots , \tag{4.26}$$

which is to be compared with the values given in Table I.

For high m, the analysis of the transfer matrix can be simplified using the symmetry of $\underline{V}$ with respect to left-right inversion of the states. For comparison, the bounds obtained from $Z_{m,m}$ are also shown in Fig. 5 (set a). This sequence shows odd-even oscillation.

V. INFINITE-TEMPERATURE ENTROPY

We have determined the partition function of $m \times m$ squares for $m=3-7$ by using the analogy with electrical circuits (see Appendix A) and also by the method of Sec. IV. From these, the entropy per unit cell at $T=\infty$, as defined in Eq. (2.3), has been obtained. These are shown in Fig. 5 (set c) plotted against $1/m$. Extrapolation to $m \to \infty$ gives

$$s_\infty = 0.314 , \tag{5.1}$$

which is to be compared with the previous estimates given in Table I.

Figure 5 shows that the approach to the thermodynamic limit is algebraic with size. Because of the analyticity of $f(T)$ above $T > T_c$ a similar algebraic approach is expected for all $T > T_c$. In contrast, we found rapid exponential looking approach $[f(T)=0]$ for $T < T_c$. This difference in behavior is a manifestation of the singularity at $T=T_c$.

VI. CONCLUSION

In this paper we proved the existence of the thermodynamic limit and obtained upper bounds for the free energy of the tiling model. From these bounds, upper bounds for T_c are also determined. The best bound is

$$\beta_c\lambda \geq 0.2459 \cdots ,$$

which is obtained from the free energy of a $9 \times \infty$ strip. The partition functions for such strips are obtained by a transfer-matrix method as developed in Sec. IV. From extrapolation of the bounds for T_c, from $m \times \infty$ strips with $m=2$ to $m=9$, T_c is estimated as

$$\beta_c\lambda = 0.270\,02 \cdots .$$

Similar extrapolation of the total number of tilings of $m \times m$ squares, up to $m=7$ gives

$$s_\infty = 0.314 .$$

These values of $\beta_c\lambda$ and s_∞ agree well with those obtained by numerical simulations as given in Table I.[2,3] We have noted that the approach to the bulk behavior looks algebraic for $T > T_c$ but exponential for $T < T_c$.

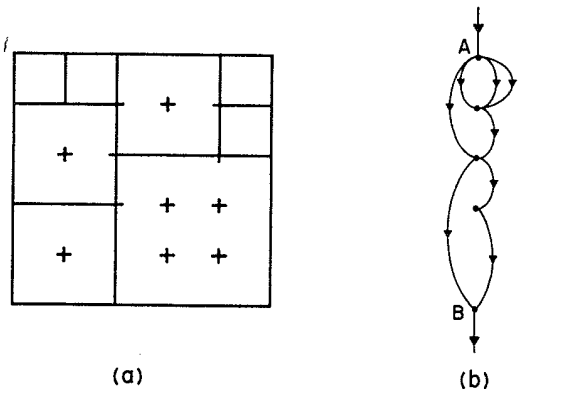

FIG. 6. (a) A particular tiling of a 5×5 lattice. (b) The corresponding electrical network. Note that Fig. 1 has the same network as (b). The multiplicity associated with (b) is 4.

APPENDIX A: CONNECTION WITH ELECTRICAL NETWORKS

Take any configuration of a finite $N \times N$ lattice, as shown in Fig. 6(a). Replace each horizontal line of the lattice by a point. Connect two points A and B by a unit resistance if the two corresponding horizontal lines are connected by a square. Delete all isolated points. The result is Fig. 6(b).

Let us now put arrows on the links of Fig. 6(b) so that they flow from upper horizontal lines to lower ones. Assign a number to the link equal to the size of the square. If a current N flows between the two extreme points A and B, then it is easy to convince oneself, from the tiling rules, that Kirchhoff's laws are obeyed.

This connection shows that each tiling can be represented by a planar network. However, the mapping is many to one. Each network is associated with a multiplicity factor that gives the number of tilings that can be generated from it. This number can be obtained from the distinct arrangements of all possible paths from A to B, each obeying Kirchhoff's law. In doing so, one can split a point, again respecting Kirchhoff's law, as in Fig. 7 and arrange the new paths. Drastic reduction in counting occurs if one considers graphs with no cut vertices and then combines them. Further simplification occurs if special symmetries are taken into account. (A similar problem is known in graph theory as the "squar-

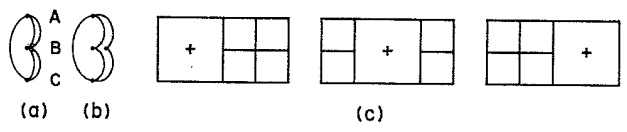

FIG. 7. Splitting a point. The degeneracy associated with (a) is obtained by splitting the point B as in (b). Note that, in doing so, Kirchhoff's law has not been violated. The multiplicity is 3. The tilings associated with this multiplicity are shown in (c).

ing the square" problem.[11]) Therefore, the tiling configurations can be obtained by solving a set of linear equations involving positive integers only.

APPENDIX B: LOWER BOUND FOR $f_{n,n}$

It is shown in Ref. 2 that four colors are necessary for coloring the tiling model. From the four-color conjecture,[12] we know that four colors are sufficient.

Let us put a four-component spin (representing the four colors), σ_i, at the center of each cell i of the $n\times n$ lattice (i.e., at the lattice points of the dual lattice). Once the tiling configuration is colored, the energy can be obtained from the Hamiltonian

$$H=\lambda\sum_{\langle ij\rangle}[1-\delta(\sigma_i,\sigma_j)]+2n\lambda\ , \tag{B1}$$

where $\delta(\sigma_i,\sigma_j)=1$ if $\sigma_i=\sigma_j$ and zero otherwise, and the summation is over the nearest neighbors $\langle ij\rangle$. Equation (B1) is the Hamiltonian of a four-state Potts model up to an additive constant. Writing $Z^{(4)}_{n,n}$ for the partition function obtained from H by summing over all spin configurations, we have

$$Z^{(4)}_{n,n}>Z_{n,n}\ , \tag{B2}$$

because of gross overcounting of states; (i) the Potts model can have domains which are not necessarily squares, and (ii) each tiling configuration can be colored in more than one way, each way corresponding to a distinct spin configuration. Therefore $f_{n,n}$ is bounded below by the free energy per spin of the four-state Potts model [Eq. (B1)] on a square lattice with n^2 lattice points.

APPENDIX C: PROOF OF $P_m=2^{m-1}$

Suppose an integer m can be partitioned into exactly n positive integers, not necessarily all distinct, in P^n_m ways. We define

$$P^n_m=0 \text{ if } n>m \text{ or } n=0\ . \tag{C1}$$

P_m, the total number of partitions into not more than m integers, is

$$P_m=\sum_{n=1}^{m}P^n_m\ . \tag{C2}$$

We also have

$$P^n_m=\sum_{i=1}^{m}P^{n-1}_{m-i} \tag{C3}$$

because the first integer can have any value $i\in[1,m]$ and $(m-i)$ can be partitioned into $n-1$ integers in P^{n-1}_{m-i} ways. Therefore

$$\begin{aligned}P_m&=\sum_{n=1}^{m}\sum_{i=i}^{m}P^{n-1}_{m-i}\\&=\sum_{i=1}^{m}\left[\sum_{n=1}^{m}P^{n-1}_{m-i}\right]\\&=\sum_{i=1}^{m}P_{m-i}=2P_{m-1}\end{aligned} \tag{C4}$$

by using (C1). The solution satisfying $P_1=1$ is

$$P_m=2^{m-1}\ . \tag{C5}$$

[1]F. H. Stillinger and T. A. Weber, Ann. N.Y. Acad. Sci. **484**, 1 (1986).
[2]T. A. Weber, G. H. Fredrickson, and F. H. Stillinger, Phys. Rev. B **34**, 7641 (1986).
[3]T. A. Weber and F. H. Stillinger, Phys. Rev. B (to be published).
[4]R. Zallen, *Physics of Amorphous Solids* (Wiley, New York, 1983).
[5]D. Shechtman, I. Belch, D. Gratias, and J. W. Cahn, Phys. Rev. Lett. **53**, 1951 (1984).
[6]One can study crystallization in this model by preferring a particular size of the tiles over the others. See Refs. 1 and 2 for details. We do not consider such possibilities in this paper.
[7]P. J. Flory, *Principles of Polymer Chemistry* (Cornell University, Ithaca, 1953).
[8]J. F. Nagle, Proc. R. Soc. London, Ser. A **337**, 569 (1974).
[9]S. M. Bhattacharjee, Phys. Rev. B **34**, 1624 (1986).
[10]F. Y. Wu, Rev. Mod. Phys. **54**, 235 (1982).
[11]R. L. Brooks, C. A. B. Smith, A. H. Stone, and W. T. Tutte, Duke Math. J. 7, 312 (1940). For a recent review, see P. J. Federico, in *Graph Theory and Related Topics,* edited by J. A. Bondy and U. S. R. Murty (Academic, New York, 1979).
[12]T. L. Satty and P. C. Kainen, *The Four Color Problem: Assaults and Conquest* (McGraw-Hill, New York, 1977).