# Square-tiling model for the glass transition: Transfer-matrix approach for the competing energy

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We introduce the correlation length for the square-tiling model for the glass transition through the transfer-matrix approach developed earlier [S. M. Bhattacharjee and E. Helfand, Phys. Rev. A **36**, 3332 (1987)] for this model. We show that this transfer-matrix approach, in the limit when the competing internal energy reduces the model to the hard-square gas problem with nearest- and next-nearest-neighbor exclusion, is equivalent to the conventional row-to-row transfer matrix. Based on this, we propose that the transitions for other values of the coupling constant for the competing term are also nonuniversal, as is known for the hard-square gas problem.

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## I. INTRODUCTION

The strange behavior of both thermodynamic (e.g., specific heat) and dynamic (e.g., diffusion, relaxation time, viscosity, etc.) properties of a liquid as it undergoes a glass transition has been known for a long time, but as yet, no definite consensus has emerged as to whether these are governed by any underlying thermodynamic phase transition [1–4]. As expected, all the three possibilities (yes, no, and maybe) have been proposed so far and are under investigation [1–5]. In such a situation, it seems necessary to study simple models that would mimic some of the glassy behavior and would incorporate what is believed to be the basic mechanism for the transition. The square-tiling model proposed by Stillinger and Weber is such a simplified model [3,6–9].

The model is based on the hypothesis that near the glass transition, clusters or domains of various types and sizes appear, and it is the restructuring of these domains that lead to the special glassy behavior. The domain-wall energy is, therefore, taken to be the most important energy in the problem. In addition, the internal energy of the domain can be different. Such an energy would compete with the domain-wall energy [10–12]. In the two-dimensional model, the domains are taken to be squares of all sizes which tile perfectly a square lattice ("space-filling structure") [6,7]. The energy for a particular configuration can be written in general as

$$E(\{n_j\})=2\lambda\sum_j jn_j+\Theta\sum_j j^{\alpha}n_j\ , \tag{1.1}$$

where $n_j$ is the number of $j\times j$ tiles (to be called $j$-tiles). The first term on the right-hand side is the domain-wall energy with $\lambda/2$ equal to the wall energy per unit length (in units of the lattice spacing), the second term represents the internal energy of the domains with a coupling $\Theta(>0)$ (also called "frustration energy") [12]. The index $\alpha(>2)$, for simplicity, will be taken to be 3. The perfect tiling condition requires

$$\sum_j j^2n_j=N^2\ . \tag{1.2}$$

One may consider further generalization that prefers tiles of a particular (crystallization) size and, may be by incorporating a term controlling the total area covered. Such generalizations will not be considered here.

Monte Carlo (MC) simulations with $\Theta=0$ showed a glasslike dynamics of stretched exponential relaxation, with "Vogel-Fulcher" law for the relaxation time, around a temperature $T_c$ where the system undergoes a *first-order transition* to an inactive low-temperature phase (ground state for all $T<T_c$) [7, 8]. Furthermore, equilibrating the system below $T_c$ turned out to be very difficult as one might expect for a glass. The transition was, therefore, taken as a glass transition. A rigorous approach for calculating bounds to $T_c$ was developed by Bhattacharjee and Helfand [9], and by extrapolation, an estimate of $T_c$ was found which agreed with the MC result.

By studying the ground-state structure and low-temperature expansions of Eq. (1.1), around the special values of

$$\theta=\Theta/2\lambda=[j(j+1)]^{-1}\ ,$$

with integral $j$, it was argued in Ref. [11] that the glass transition point of this model is really a *multiphase* point, where an infinite number of higher-order phase transition lines meet [13]. To set the notation, we recast the argument for the multiphase point in a slightly different way in the spirit of the Pirogov-Sinai theory [14]. The phase diagram at $T=0$ is obtained by minimizing the energy in Eq. (1.1) with $\alpha=3$. First note that the ground state consists of just 1-tiles if $\lambda<0$ for all $\Theta\geq 0$. The other extreme case is $\Theta<0$, when a single $N$-tile will cover the whole lattice, irrespective of the sign of $\lambda$ as shown in Fig. 1(a). Nontrivial structure develops in the positive quadrant for $\Theta>0$ and $\lambda>0$, because the ground state goes from all $j$-tiles to all $(j+1)$-tiles if $\Theta\leq 2\lambda[j(j+1)]^{-1}$ [11,15]. The resultant $T=0$ phase diagram is shown in Fig. 1(a). For very low temperatures, the phase diagram is expected to be [being a two-dimensional (2D) lattice problem] a small perturbation of the $T=0$ diagram [14]. Thanks to the existence [6–9] of a phase transition for $\theta=0$, and $\lambda>0$ and also the transition in a $1\times 1$ and $2\times 2$-tiled system (hard-square lattice-

 

gas problem) [16–18] the multiphase point at $\theta=\lambda=0$ in Fig. 1(a) is expected to get shifted for $T>0$ to the right as in Fig. 1(b). This was further supported by symmetry arguments and finite-size effects [11]. The existence of a multiphase point is attractive because, slightly away from the glass transition point, a small change in the final temperature in a cooling experiment may take the system into different distinct thermodynamic phases. It is tempting to believe that this leads to the apparent diversity in the structures observed during cooling in MC simulations. Could such a sensitive dependence on temperature be true for a real glass?

It is therefore important to understand the behavior of the system with two competing terms as in Eq. (1.1). Approximate theories are of not much help as can be seen from Ref. [12] where even the qualitative results depend crucially on the level of approximation, like no phase transition in the first order but a continuous one in the second order. Mean-field theories are also of not much help because of lack of knowledge of any suitable "unperturbed" Hamiltonian. It is possible to develop good approximation deep in the high-temperature region by choosing $\mathcal{H}_0=\lambda'\sum_j jn_j$ and determining $\lambda'$ through the variational principle. Since this $\mathcal{H}_0$ does not respect the ground-state structure, a mean-field theory based on this will fail in the low-temperature region and therefore cannot be used to study phase transitions.

Our aim in this paper is to develop a procedure through phenomenological renormalization that can be used to study this model. This is done by generalizing the transfer-matrix approach of Ref. [9] for the model with the competing term. We define a correlation length in Sec. II and prove the equivalence of this approach with the conventional row-row transfer matrix as used for the hard-square gas problem [16,18]. In the low-temperature limit when the model is equivalent to the hard-square gas problem, we show that $\xi$ defined here agrees with the length defined in the conventional approach. The application to the other transitions of the phase diagram is then discussed. A summary is given in Sec. III. The Appendix contains an antiferromagnetic spin Hamiltonian for the case for which only the $3\times3$ and $2\times2$ tiles are required.

## II. TRANSFER-MATRIX APPROACH

### A. The matrix and the correlation length

A transfer-matrix method was developed by Bhattacharjee and Helfand in Ref. [9] to study the tiling model with just the $\lambda$ term in Eq. (1.1). A more general form of this matrix has also been discussed there, which can be easily applied to the problem with the $\theta$ term. This transfer matrix $\mathcal{V}$ is different from the usual row-to-row transfer matrix and the thermodynamic behavior is determined by the smallest zero of the determinant $|\mathcal{V}-I|$ as opposed to the largest eigenvalue. The reason for this difference is that $\mathcal{V}$ transfers between properly defined basis states so that a finite number of transfers by $\mathcal{V}$ leads to an ensemble of lattices of various sizes. In that sense, $\mathcal{V}$ can be thought of as the transfer matrix for a grand ensemble from whose partition function one can calculate the partition function for a lattice of definite size. The reader is referred to Ref. [9] for details and only the basic steps of the procedure are outlined here.

We consider a lattice with $N$ bonds in the horizontal direction with periodic-boundary condition and $M$ rows in the vertical direction. For simplicity, we will choose open boundary condition in the vertical direction and take $M\to\infty$. The lattice is tiled by sequentially placing tiles of allowed sizes with the following rules: (i) tiles are placed only on the lowest unoccupied row (LUR) of the upper boundary (UB), (ii) all bonds of the LUR are to be tiled, and (iii) removal of any tile that defines the upper boundary must lower the position of the LUR.

The states that define the transfer matrix are the configurations of the upper boundary that one may get using the above rules. These states can be uniquely specified by a sequence of $N$ integers $(n_1,n_2,\ldots,n_N)$, $n_\alpha\in[0,N-1]$, where $n_\alpha$ would give the height of the $i$th bond of the UB as measured from its LUR (for which $n=0$). Each time a new UB is created, LUR is redefined as 0.

If going from a state $|\{n_\alpha\}\rangle$ to $|\{n'_\alpha\}\rangle$ requires $p_j$ of $j$-tiles (with weight $z_j$ for each $j$-tile), covering completely $w$ rows in the process, then the transfer matrix is defined as

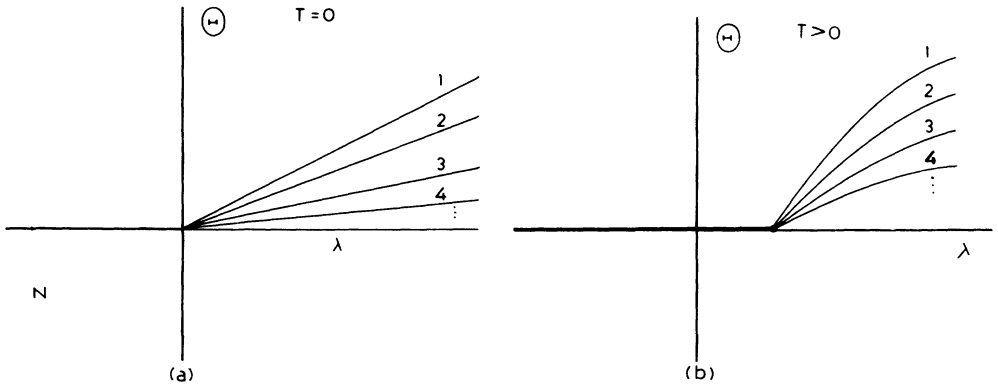


FIG. 1. (a) Phase diagram at $T=0$. The numbers $1,2,\ldots,N$ represent the size of the tile in the ground state. (b) The phase diagram at $T>0$.

$$\langle\{n'_\alpha\}|\mathcal{V}|\{n_\alpha\}\rangle=\sum_{\text{allowed tilings}}\prod_j z_j^{p_j}t^w\ , \tag{2.1}$$

where the sum is over all possible tilings that can take the unprimed state to the primed one, and $t$ is the weight for each completely covered row. Note that such tilings may have not only different $\{p_j\}$ but also different $w$.

For the $N\times M$ lattice under consideration, the partition function would come as

$$Z_{N,M}=\frac{1}{2\pi i}\oint\frac{1}{t^{M+1}}\langle 0|(\mathcal{V}-I)^{-1}|0\rangle dt\ , \tag{2.2}$$

where $I$ is the identity matrix and $|0\rangle$ denotes the state with perfectly horizontal boundary as the first and the last row of the lattice and the contour encloses the origin of the complex $t$ plane without enclosing any singularities of $(\mathcal{V}-I)^{-1}$. The singularities of $(\mathcal{V}-I)^{-1}$ are just poles at the zeros of

$$|\mathcal{V}-I|=0\ . \tag{2.3}$$

The contour can be deformed into small circles around the above zeros only, such that, in the limit $M\to\infty$, the smallest zero $t_0$ contributes giving

$$\lim_{M\to\infty}\frac{1}{M}\ln Z_{N,M}=\ln t_0\ . \tag{2.4}$$

For finite $M$,

$$Z_{N,M}=\sum_{i=0}^{L}c_i t_i^M\ , \tag{2.5}$$

where $L$ is the number of zeros. The approach to the thermodynamic limit is, therefore, given by $(|t_1|/t_0)^M$, $t_1$ being the second smallest root. A correlation length can, therefore, be defined as

$$\xi=\left[\ln\frac{|t_1|}{t_0}\right]^{-1}\ . \tag{2.6}$$

This is reminiscent of the definition of the correlation length in the conventional transfer-matrix approach.

Once the correlation length is defined, the transition point can be estimated by using phenomenological renormalization [19]. In this approach, if $\xi_M$ is the correlation length for an $M\times\infty$ lattice, then the transition point corresponds to the "temperature" at which

$$\frac{\xi_M}{\xi_L}=\frac{M}{L}\ . \tag{2.7}$$

We use this to locate the transition temperatures in the low-temperature limit.

### B. Hard-square gas: equivalence

We now show the equivalence of the state-state transfer matrix (STATM) introduced in the preceding section with the conventional row-row transfer matrix (ROWTM), for a problem where both can be used, namely, the hard-square lattice-gas problem with nearest-neighbor (NN) and next-nearest-neighbor (NNN) exclusion. This will also show that the correlation length defined by Eq. (2.7) is consistent with the conventional definition from the row-row transfer matrix. We follow Runnels and Combs [16] in constructing the conventional ROWTM. Incidentally, the hard-square gas problem arises in the low-temperature limit of the tiling model around $\theta\equiv\Theta/\lambda=\frac{1}{2}$. The fugacity for the $2\times2$ tiles on the $1\times1$ tile background is given by $z=\exp(-4\beta\lambda\epsilon)$, where $\epsilon=\theta-\frac{1}{2}$, in the double limit $\epsilon\to0$, $\beta\to\infty$, with $z$ finite.

The conventional ROWTM is most easily described in the lattice-gas language. Since the 1-tiles can be placed in one and only one way, once the 2-tiles are in place, one can altogether ignore them. The lattice-gas variable (0 or 1) would then tell us whether a site on a row has a 2-tile which is being replaced by an occupied central site with vacant NN and NNN sites (0). The ROWTM is, therefore, defined with respect to the states of the rows and these states can be represented by sequences of 0 and 01, the maximum number of 1 being $[N/2]$, the integer part of $N/2$. Since 1 must have 0 as its NN, it follows that the number of states $d_n$ for a row of $N$ sites is related to the numbers for smaller $N$'s as $d_N=d_{N-1}+d_{N-2}$ (Fibonacci sequence).

For the STATM, the states are again described by 0 and 1, and $\mathcal{V}$ completes one row at each operation unless it is placing all 2-tiles on the $|0\rangle$ state (see Fig. 2.). Thanks to this row-by-row buildup by $\mathcal{V}$ at each step, the configuration of UB can as well be represented by the state of the completed row, except for the all 2-tile case with even $N$. There are two such configurations which are to be counted in $|0\rangle$ as opposed to the (010101...) and (101010...) states for RTM. Therefore if $d'_N$ is the number of basis states for STATM, then $d'_N=d_N-2$.

Labeling the row states for the ROWTM as $|1\rangle\equiv101010\ldots,|2\rangle\equiv010101\ldots,|3\rangle\equiv00000\ldots$, etc, the ROWTM will have $\langle3|R|1\rangle=\langle3|R|2\rangle=z$, $\langle1|R|3\rangle=\langle2|R|3\rangle=z^2$, $\langle1|R|\text{any other}\rangle=\langle\text{any other}|R|1\rangle=\langle2|R|\text{any other}\rangle=\langle\text{any other}|R|2\rangle=0$. The eigenvalue equation $|R-\lambda I|=0$ can be written as $|\tilde{R}-1|=0$, where

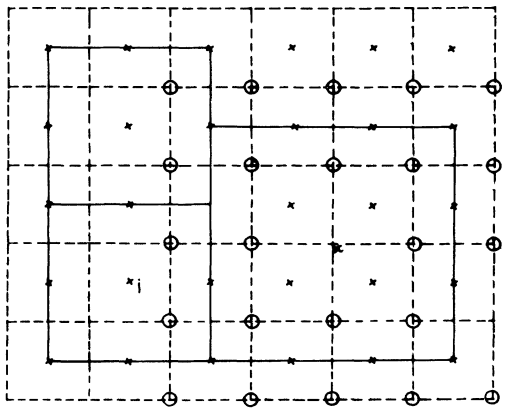

FIG. 2. 2-tiles and 3-tiles on the lattice. For clarity only a few tiles are shown. Dashed lines represent the dual lattice, × indicates sites on the real lattice, and ○ sites are the first- and second-shell neighbors enclosing site $\alpha$ on the dual lattice.

$$\tilde{R}_{ij}=\begin{cases}R_{ij} & \text{for } i\neq j\\ R_{ii}/\lambda & \text{for diagonal elements .}\end{cases}$$

Trivial transformation, then, reduces this $d_N\times d_n$ determinant to $d'_N\times d'_N$ eliminating the first and second row and columns. The new $\langle 3|R|3\rangle$ element is $\lambda^{-1}+2z^2\lambda^{-2}-1$ which is also the $\langle 0|\mathcal{V}-I|0\rangle$ element if we use $\lambda^{-1}=t$. All other elements of $\tilde{R}-I$ and $\mathcal{V}-I$ are the same, showing that the two are equivalent.

The above equivalence establishes that the correlation length defined by Eq. (2.6) agrees, in this case, with the conventional definition of this length [16]. It was noted by Kinzel *et al.* that in the ROWTM formalism the two largest eigenvalues determining correlation length $\xi$ for the hard-square gas problem belong to the block of the matrix that transforms like the identity (under the 2D rotation group) [18,16]. This leads to the nonuniversal behavior unlike the hard-square gas with only nearest-neighbor interaction [17]. The latter problem has an Ising-type critical behavior, and it can easily be verified that no STATM can be constructed for it.

In the STATM procedure, it is easy to check that the relevant zeros should come from the identitylike block that transforms because for the $2\times\infty$ lattice, the transfer matrix is just a $1\times 1$ scalar. Since the symmetry property is not expected to change with the lattice size, hence for any $N$, the two zeros will always come from these identitylike blocks. As is well known [16], this block is obtained by considering only those states which are not translationally related. Our numerical calculations, using the phenomenological renormalization as discussed in Sec. II A, agree with the results of Ref. [18].

We therefore hypothesize that if, in the STATM approach, there is one lattice for which the transfer matrix is a $1\times 1$ scalar, then the correlation length will be determined by the zeros coming from the identitylike block, and the transition will be nonuniversal.

### C. 3-tiles and 2-tiles

In order to study the phase diagram near $\theta=\frac{1}{6}$ at which the ground state changes from 2-tiles to 3-tiles, we consider the low-temperature limit, such that only $3\times 3$ excitations over the $2\times 2$ background need to be considered. With $\theta=\frac{1}{6}+\epsilon$, the fugacity for the 3-tiles is $z=\exp(-18\epsilon\beta\lambda)$ which is to be held fixed in the double limit $\epsilon\to 0$ and $T\to 0$. Note that the 3-tiles occur in combinations of 4 replacing nine 2-tiles.

The construction of the STATM is now straightforward. For a $6\times\infty$ lattice that can accommodate both 3- and 2-tiles, it is a $1\times 1$ matrix so that

$$|\mathcal{V}-I|=3t^3z^2+2t^2-1 . \tag{2.8}$$

The next lattice to be considered is $12\times\infty$, for which the identitylike block of the matrix is $6\times 6$, the relevant equation being

$$12t^9z^8+12t^7z^4+3t^6z^4(z^4-4)-10t^5z^4 +3t^4z^4-2t^3(2z^4-1)-2t^2-t+1=0 . \tag{2.9}$$

As a check for $z=0$ both the equations reproduce, as they should, the correct entropy $\frac{1}{2}\ln 2$ per row. Similarly the correct entropy is also reproduced in the $z\to\infty$ limit. The occurrence of the pair $\pm 1/\sqrt{2}$ of zeros requires that the length of the strip be even. No conclusion regarding the transition point can, however, be drawn from these two lattices because phenomenological renormalization gives two solutions. This is not surprising because the smallest allowed lattice generally does not help in phenomenological renormalization, as is well known for the Ising model [19]. Unfortunately, the rapid growth of the size of the matrix (next relevant width of the lattice is 18) forbids us from extending this to bigger lattices. However, the use of the hypothesis tells us that the phase transition will also be a nonuniversal one similar to the hard-square gas problem.

## III. CONCLUSION

We proved the equivalence of the transfer matrix developed in Ref. [9] with the conventional row-row transfer matrix for the hard-square gas problem to which the square-tiling model reduces in the low-temperature region around $\theta=\Theta/2\lambda=\frac{1}{2}$. We then argued that the transition for the $3\times 3$–$2\times 2$ around $\theta=\frac{1}{6}$ is nonuniversal as is known for the hard-square gas problem. The small lattices studied could not locate the transition point, calling for studies of larger lattices.

## ACKNOWLEDGMENT

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## APPENDIX: A SPIN REPRESENTATION FOR THE 3- AND 2-TILE PROBLEM

It is known that the hard-square problem with NN and NNN exclusion can be obtained as an extreme limit of an Ising antiferromagnet, which has been studied extensively using various techniques [18]. In this appendix, we show that the 3-tile and 2-tile problem that occurs near $\theta=\Theta/2\lambda=\frac{1}{6}$ can also be described by a suitable antiferromagnet, though it will be a little bit more complicated because it requires spins both on the real lattice and on the dual lattice.

An inspection of Fig. 2 will show that any configuration of the 2-tiles and 3-tiles lattice can, as well, be represented by lattice-gas variables (0 or 1) sitting on the real lattice for the $2\times 2$ tiles and on the dual lattice for the $3\times 3$ tiles. We will use greek indices to represent the lattice sites and the variables on the dual lattice while roman letters for the real lattice.

Let $\tau_0$ and $t_i$ be the lattice-gas variables sitting at the dual site $\alpha$ and the real site $i$. The partition function, taking into account the hard-core restriction, can now be written as

$$Z=\sum_{\tau,t}\prod_{\substack{[\alpha\beta]_{12}[i\alpha]_{12},\\ [ij]_1}}(1-\tau_\alpha\tau_\beta)(1-t_it_j)(1-t_i\tau_\alpha)z^s ,$$

where $s=\sum_\alpha\tau_\alpha$ and the factors would be zero if both the involved sites are occupied. The product is over the $\alpha,\beta$

pairs, if they are first- and second-shell neighbors as explained in Fig. 2. $\beta$ lies on the first or the second enclosing square on the dual lattice. This comes from the hard-core restriction of the 3-tiles. Similarly the 2-tile restriction translates into a single shell exclusion for $ij$ pairs. However, an occupied $i$ site on the real lattice excludes lattice-gas particles from the neighboring two shells of the dual lattice. Similar is the case for an occupied dual site. The last factor is the weight factor for the 3-tiles, $z$ for each one of them. The partition function requires a summation over all the lattice-gas variables with the perfect tiling condition:

$$4\sum_i t_i + 9\sum_\alpha \tau_\alpha = N^2 .$$

The above partition function can be represented by a spin Hamiltonian. Using $s_i = 2t_i - 1$ and $\sigma_\alpha = 2\tau_\alpha - 1$, the mutual exclusion can be represented by an antiferromagnetic interaction, so that for each type of bond that appears in the partition function we will have an antiferromagnetic coupling. For simplicity we choose all the couplings to be the same. The tiling problem will be recovered in the limit when this coupling goes to infinity. The resultant Hamiltonian, incorporating the perfect tiling condition, is

$$H = J\sum_{[i,j]_1} s_i s_j + J\sum_{[\alpha,\beta]_{12}} \sigma_\alpha\sigma_\beta + J\sum_{[i,\alpha]_{12}} s_i\tau_\alpha - \mu\sum\tau_\alpha ,$$

where the sums are over pairs which are mutually on the excluded shell, the last term is the chemical potential for the 3-tiles controlling its concentration. As we already said, one can consider a more general Hamiltonian with different couplings for different types of pairs. It is easy to generalize spin representations for $j$- and $(j+1)$-tiles at a cost of distant-neighbor interactions.


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[15] We like to point out the importance of the unrestricted sum over all the domains in the Hamiltonian. To get the phase diagram of Fig. 1(a), it is imperative that the sum in the $\Theta$ term be over the whole range of $j$. It is easy to see that a lower cutoff like $\sum_{j=a}^{\infty} j^3 n_j$ would remove all $j < a$ tiled phases from the phase diagram. In contrast, an upper cutoff for the competing term would completely wash out the phase diagram because the ground state will always be the largest tile as in the $\theta = 0$ case. In this situation, the $\theta = 0$ transition is expected to occur for all $\lambda$.

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