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First order phase transition in a model of quasicrystals

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Abstract

We introduce a family of two-dimensional lattice models of quasicrystals, using a range of square hard cores together with a soft interaction based on an aperiodic tiling set. Along a low temperature isotherm we find, by Monte Carlo simulation, a first order phase transition between disordered and quasicrystalline phases.

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(Some figures in this article are in colour only in the electronic version)

1. Introduction

Certain alloys have been shown to exhibit exotic structures in the sense that the structures produce sharp X-ray Bragg peaks with symmetries which cannot arise from any crystal [1]. We model these materials with equilibrium statistical mechanics using a technique due to Levine and Steinhardt [2] which employs an energy ground state associated with an ‘aperiodic tile set’, that is, a set of several geometric shapes which only achieve their densest packings in nonperiodic ways, the best known two-dimensional example being the kite and dart shapes of Penrose [3]. There is very little direct evidence that any equilibrium statistical mechanics model would show a well-defined quasicrystalline phase at positive temperature; we know only one model for which there is (simulation) evidence [4, 5] of such a phase. The theory of solid/fluid phase transitions often emphasizes the role of crystal symmetry, going back at least to Landau [6, 7], and it is of interest to understand how the quasicrystalline state, with its exotic form of symmetry [8], fits into this picture. In this regard it is noteworthy that the simulations in [4, 5] indicate a continuous (high order) transition between the fluid and quasicrystalline phases. We revisit this issue here with slightly more realistic models and find clear simulation evidence of a discontinuous (first order) transition. We contrast our results with related models of hard squares and of Widom–Rowlinson type.

2. The model

Our models are variants of that used in [4, 5], which we describe first. The lattice model of [4, 5] uses 16 types of particles, each corresponding to an element of a certain subset

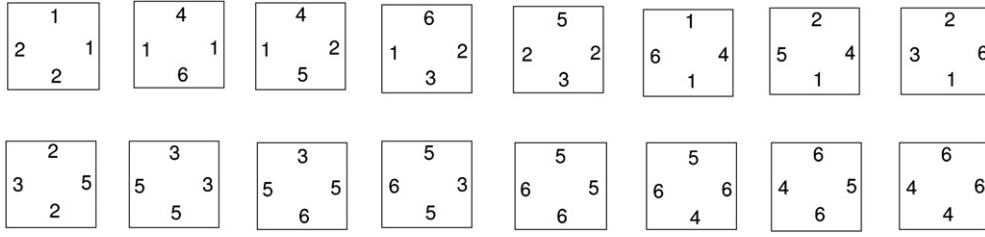


Figure 1. Ammann's 16 Wang tiles.

$S \subset \{1, 2, 3, 4, 5, 6\}^4$ of cardinality 16, associated with an aperiodic tiling set due to Ammann [9]. The model consists of configurations of particles on the square lattice $\mathbb{Z}^2$. It is convenient to interpret each element of S as a unit-edge square *tile*, with its 4 edges colored using 6 possible colors $\{1, 2, 3, 4, 5, 6\}$ in one of the 16 groups of 4 indicated by S ; see figure 1.

The model in [4, 5] requires precisely one particle/tile centered at each lattice site, and each particle interacts with each of its 4 nearest neighbors only, with interaction energy 0 or -1 , with the negative value when the edge colors match. A canonical ensemble is used, but with density fixed at 1. The model then exhibits a continuous phase transition at positive temperature [4, 5], with the low temperature phase showing quasicrystalline structure, including the symmetry [8] it inherits from Ammann's aperiodic tiling set.

We consider variants of the above model, investigating the consequences of two changes: extending the size of the square tiles to integer edge length $w \geq 1$ (they are still 'hard' – no overlap of tiles is allowed); and allowing a full range from 0 to 1 of volume fraction for the particle configurations, in place of the fixed volume fraction of 1 used in [4, 5]. Note that the latter adds a second thermodynamic variable to the model.

The description of the interaction is a bit more complicated than in the $w = 1$ model of [4, 5], since tiles which are close but not touching may still interact, though with lower amplitude. To define the interaction we need some notation. A *tile-state* A is an element of $\mathbb{Z}^2 \times S$, with the first coordinate of A representing its center on the lattice, and the second coordinate representing its edge colors. We define a pairwise interaction H between tiles as follows. For each tile-state A , consider the four 'nearest neighbor' (empty) squares of the same size as A which surround A but do not overlap it, labelled N , S , W and E as in figure 2. If another tile-state A' is disjoint with A but overlaps one of these four squares – it cannot possibly overlap more than one – its energy of interaction with A , $H(A, A')$, is as follows. Assume without loss of generality that A' overlaps N . Then $H(A, A')$ is negative if and only if the north edge of A has the same color as the south edge of A' , and positive otherwise; and has absolute value given by $|H(A, A')| = (3w - d)/(2w)$, where d is the ℓ^1 distance between the centers of A and A' . (The ℓ^1 distance between (x, y) and (x', y') is $|x - x'| + |y - y'|$.) If A' overlaps S , W , or E , then the energy H is defined similarly.

Given an inverse temperature β , a chemical potential μ (common to all tiles/particle types), and a finite volume $\mathcal{V} \subset \mathbb{Z}^2$, we consider the grand canonical ensemble: the probability $\mathcal{P}_{\beta, \mu}$ on configurations C of tile-states in $\mathcal{V}$ which is given by

$$\mathcal{P}_{\beta, \mu}(C) = \frac{\exp[-\beta(\mathcal{H}(C) - \mu\mathcal{N}(C))]}{Z}, \quad (1)$$

where $\mathcal{N}(C)$ is the number of tiles-states in C , $\mathcal{H}(C) = \sum_{A \neq A' \in C} H(A, A')$ is the total energy of C , and $Z = Z(\mu, \beta)$ is the appropriate normalization. In our simulations we use 'square' volumes $\mathcal{V} = \mathbb{Z}^2 \cap [-k, k]^2$ with k an integer multiple of w . We use boundary conditions as

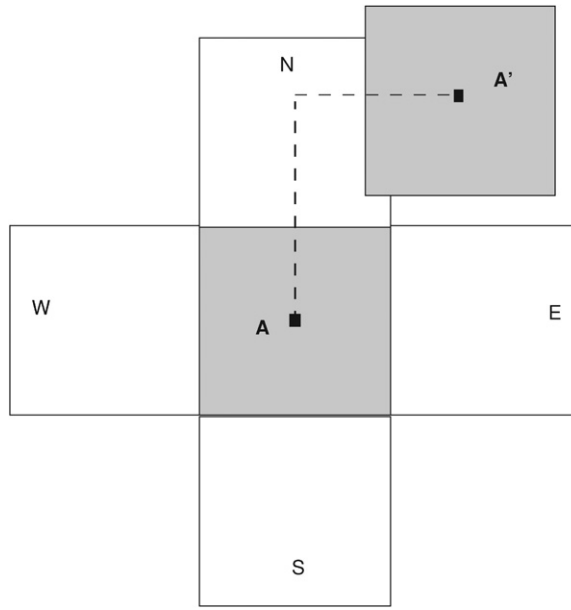


Figure 2. The interaction energy depends on the ℓ^1 distance between particle centers (dashed line).

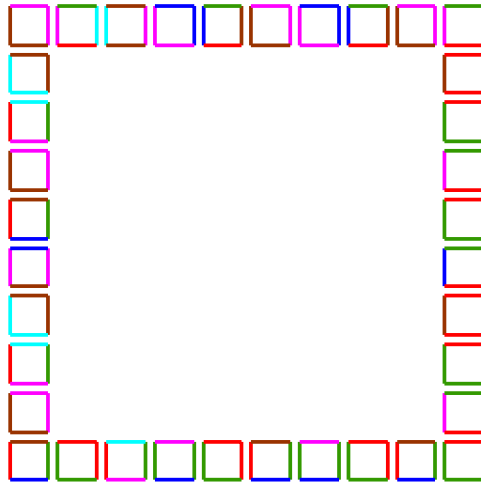


Figure 3. A boundary shell (with edges colored instead of numbered).

follows. Starting from a perfect tiling $\mathcal{T}$ by tile-states, that is, a configuration of tiles covering $\mathbb{Z}^2$ in which all edge colors match, we remove all the tiles except those just outside $\mathcal{V}$; this leaves a (fixed) boundary ‘shell’; see figure 3. (We use 10 000 different boundary shells arising from random translations of a single tiling.)

It remains to define the order parameter, χ . Each tile-state has a ‘long/short pattern’ given by its 4 edge colors, where we call colors 1 and 2 ‘short’ and colors 3, 4, 5 and 6 ‘long’. Note that, for any tile, horizontal edges are either both short or both long, and vertical edges are either both short or both long, so there are 4 possible long/short patterns

for a tile. Any tile-state A centered ‘near’ the site $(wi, wj) \in \mathcal{V}$, namely centered in $[wi - w/2, wi + w/2) \times [wj - w/2, wj + w/2)$, will be said to *agree with* $\mathcal{T}$ if it has the same long/short pattern as the tile-state in $\mathcal{T}$ centered at (wi, wj) . Now, consider the thick ‘shells’ $\mathcal{V}_j = \mathcal{V} - [-j, j]^2$. Given a configuration C in $\mathcal{V}$ and boundary from $\mathcal{T}$, let j^* be the smallest integer such that at least 80% of sites (wi, wj) in $\mathcal{V}_{j^*}$ have a tile-state centered nearby (in the above sense) which agrees with $\mathcal{T}$. The order parameter χ is defined as the corresponding normalized volume, $\chi(C) = |\mathcal{V}_{j^*}|/|\mathcal{V}|$.

3. Simulations and results

We ran Markov-chain Monte Carlo simulations of the model with tile widths $w = 1, 2$, and 3 . For each tile width w we simulated volumes $|\mathcal{V}| = (20w)^2, (40w)^2, (60w)^2, (80w)^2$ and $(100w)^2$. (We will also use the notation $V = |\mathcal{V}|/w^2$, so V represents the maximum possible number of tile-states in a single configuration.)

The Monte Carlo steps were designed to produce the target distribution of equation (1). The basic Monte Carlo step involves a choice between four types of moves: moving a particle (locally), changing a particle type, removing a particle, and adding a particle. As usual in grand canonical Monte Carlo simulation, given a configuration C a trial configuration C' is introduced, effecting one of the moves described above, and is accepted with probability determined by the desired limiting distribution, that is, equation (1).

More precisely, the basic Monte Carlo step is as follows. Let $C(n)$ be the configuration at the n th step of the Monte Carlo chain, and choose a random lattice site x . Select $k \in \{1, 2, 3, 4\}$ with probability p_k , where the p_k are the probabilities of attempting the four types of Monte Carlo moves, with the p_k summing to 1 and $p_3 = p_4$. (We choose the p_k somewhat arbitrarily.) Without loss of generality suppose $x \in R := [wi - w/2, wi + w/2) \times [wj - w/2, wj + w/2)$. First suppose $k \in \{1, 2\}$. If there is no tile-state centered in R , then $C(n+1) = C(n)$; otherwise, let A be the (unique) tile-state centered in R , and produce a trial configuration $C(n)'$ by: if $k = 1$, moving the center of A to a site in R chosen uniformly at random; if $k = 2$, replacing A with a different tile-state centered at the same lattice site. Now suppose $k = 3$. If there is a tile-state A centered in R , then $C(n)'$ is obtained by removing A ; otherwise $C(n+1) = C(n)$. Finally suppose $k = 4$. If there is no tile-state centered in R , then let $C(n)'$ be the configuration obtained from $C(n)$ by adding a tile-state chosen uniformly at random and centered at x ; otherwise $C(n+1) = C(n)$. In cases where a trial configuration $C(n)'$ is introduced, we take $C(n+1) = C(n)'$ with probability $Q = \min(1, q)$; otherwise we take $C(n+1) = C(n)$. Here q is given by

$$q := \min\{1, e^{\beta[\mathcal{H}(C) - \mathcal{H}(C') + \mu^*(\mathcal{N}(C') - \mathcal{N}(C))]} \}, \quad (2)$$

where we have written C for $C(n)$ and C' for $C(n)'$, and take $\mathcal{H}(C') = \infty$ for trial configurations C' with overlapping tiles. Here $\mu^* = \mu + \beta^{-1} \ln(16w^2)$ is chosen so that the Monte Carlo configurations $C(n)$ have limiting probability distribution given by equation (1). (The value μ^* arises because of the following. Starting with a configuration C , the probability of proposing a trial configuration C' which removes tile-state A from C is $16w^2$ times the probability that, starting with the configuration C' , a trial configuration $C'' = C$ is proposed which adds the tile-state A to C' .)

In our simulations we slowly increase μ , starting from low μ , using the final configuration in the simulation of each μ as the starting configuration in the simulation of the next μ , and starting with an initial configuration which is a vacuum. Our simulations of each value of μ for the largest volume, $V = 100^2$, have 10^{10} Monte Carlo steps for $w = 1$ and $w = 2$, and 2×10^{10} steps for $w = 3$. The simulations have between 50 to 100 different values of μ , with our longest

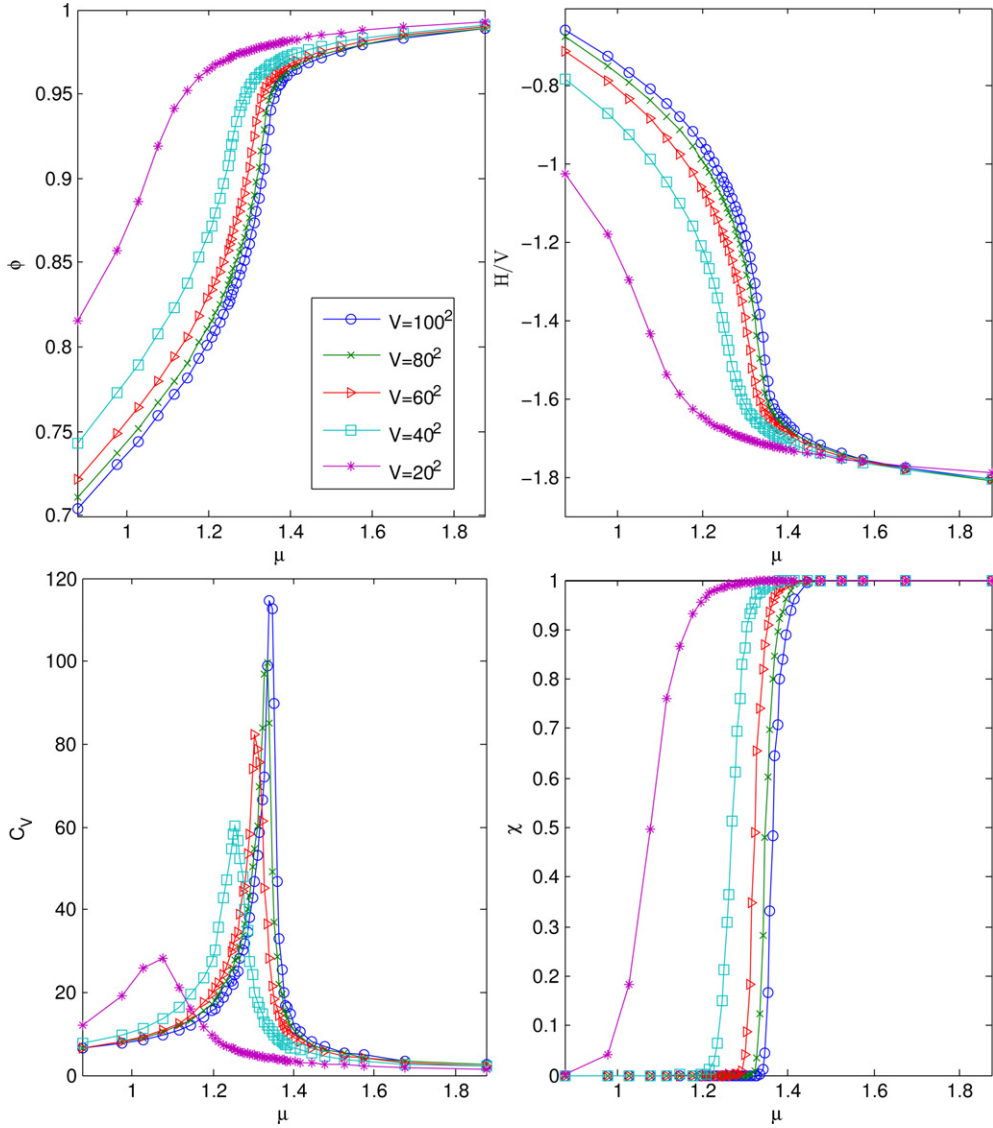


Figure 4. Data for $w = 2$ and $\beta = 1.5$.

simulations, for the model with $w = 3$, having a total length of 2×10^{12} Monte Carlo steps. The lengths of our simulations of smaller volumes were scaled relative to the largest volume; for instance the simulation of each μ for $V = 60^2$ and $w = 3$ had $(60^2/100^2) \times 2 \times 10^{10}$ Monte Carlo steps. To check that our simulations have reached equilibrium, we performed some simulations which ran ‘backwards’, starting from ground states and large μ ; we checked that the resulting data was in good agreement with our simulations starting from vacuum. To smooth the data curves we ran 100 (independent) copies of each simulation.

For $w = 2$ and $w = 3$ we find clear evidence of discontinuous (first order) phase transitions. At the transitions the average volume fraction ϕ and energy per volume $\mathcal{H}/V$ exhibit developing jump discontinuities as V increases; see figures 4–5. Furthermore the (volumetric) heat capacity, C_V (defined as the derivative of $\mathcal{H}/V$ with respect to $T = 1/\beta$),

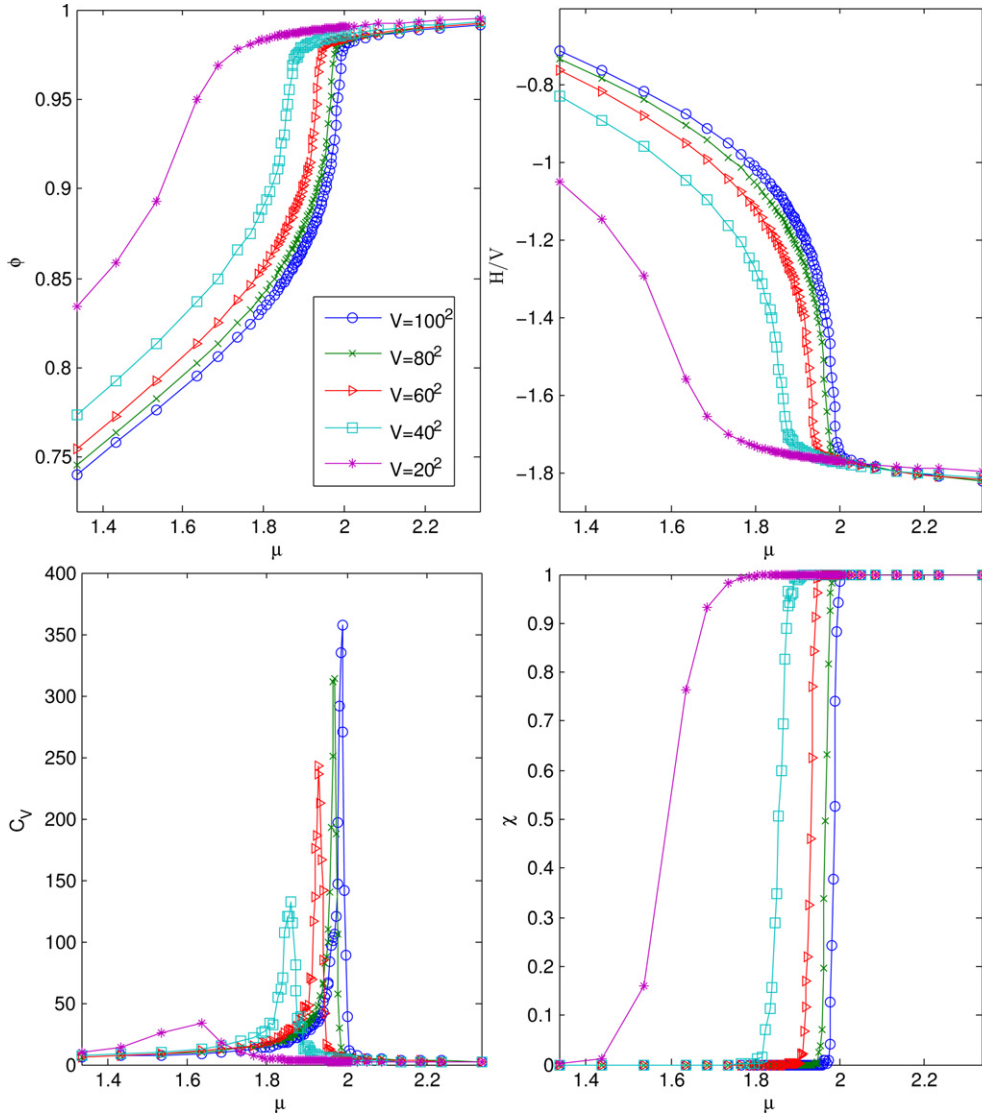


Figure 5. Data for $w = 3$ and $\beta = 1.5$.

exhibits a developing delta function as V increases (see figures 4–5). From our data in figure 6, for the $w = 1$ model, there appears to be a continuous transition (at $\beta = 1.5$ near $\mu = 0.8$), from a disordered fluid phase to a phase with quasicrystalline order, similar to the transition in [4, 5] of the canonical ensemble at density 1.

From our data in figure 6, for the $w = 1$ model, there appears to be a continuous transition (at $\beta = 1.5$ near $\mu = 0.8$), from a disordered fluid phase to a phase with quasicrystalline order, similar to the transition in [4, 5] of the canonical ensemble at density 1.

To estimate the transition values of μ for $w = 1, 2, 3$, we searched for the parameters μ^* which optimized the least-squares linear fit of a log-log graph of $(\mu^* - \mu(V))$ versus V , where $\mu(V)$ is the x -coordinate of the maximum of C_V . (The optimization minimized the sum of the squares of the distances between the values of $\log(\mu^* - \mu(V))$ and the corresponding values

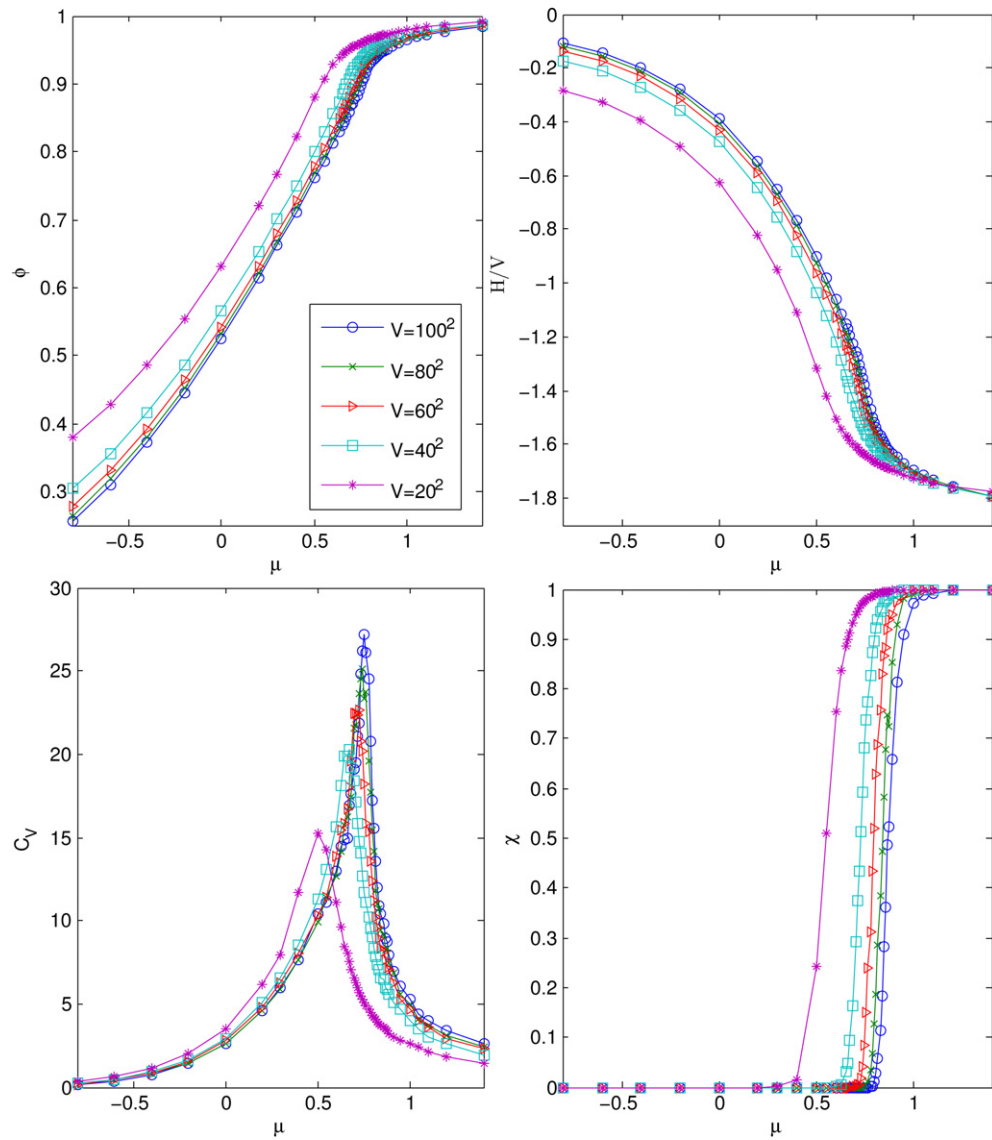


Figure 6. Data for $w = 1$ and $\beta = 1.5$.

of the fitting line.) We estimated the transitions occur at: $\mu^* = 0.789$ for $w = 1$; $\mu^* = 1.389$ for $w = 2$; and $\mu^* = 2.066$ for $w = 3$.

To quantify the developing delta function in C_V noted above for $w = 2, 3$ we measure, from figures 4 and 5, the maximum value h of C_V divided by the ‘width’ of the graph of C_V at $h/2$ (see figure 7). The order parameter χ also develops a discontinuity at the transition, signaling the onset of quasicrystalline symmetry: see figures 4–5.

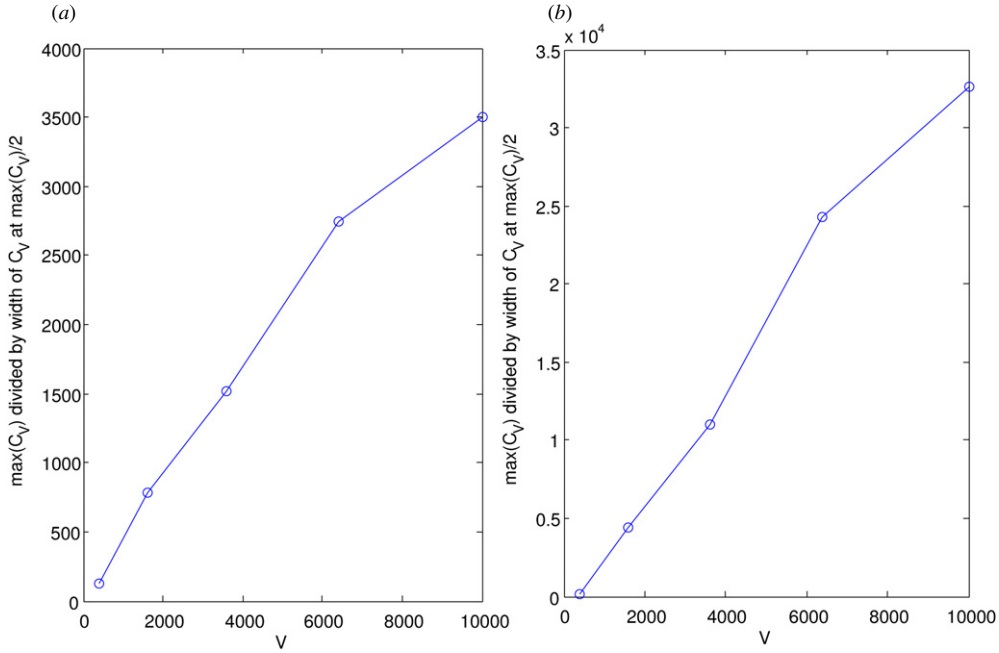


Figure 7. Evidence of C_V developing into a delta function for $\beta = 1.5$ and (a) $w = 2$, and (b) $w = 3$.

4. Comparisons

We next compare our results with related models, in particular hard squares and Widom–Rowlinson models on $\mathbb{Z}^2$; see [10] for a review of, and earlier references to, the former and [11] for a review of, and earlier references to, the latter.

At sufficiently high temperature the soft interaction between our particles must become negligible compared to the hard core so we first review what is known about hard square systems.

For hard squares on $\mathbb{Z}^2$ the energy is zero for all configurations so it is convenient to use a grand canonical ensemble with fixed temperature 1 and variable chemical potential μ . Hard squares with edge size $w = 1$ (‘point hard core’) do not have a phase transition as μ is varied, since the variables at each site, ‘occupied’ or ‘unoccupied’, are independent. For fixed $w \geq 2$ the densest configurations, of volume fraction 1 and corresponding to $\mu \rightarrow \infty$, are degenerate, allowing parallel rows or columns of squares to slide independently in what is called a ‘columnar’ phase. It is generally accepted from simulation [10] that the model with $w = 2$ has a continuous transition at volume fraction about 0.92. We only know of one paper on the model with $w = 3$ [10]; the authors claim a first order freezing transition beginning at a density above 0.91, but are unable to show developing discontinuities in any thermodynamic quantities.

We now compare our model to hard squares on $\mathbb{Z}^2$. Consider a version of our quasicrystal model with $w \geq 2$ but in a canonical ensemble with volume fraction fixed at 1. Based on the results of [4, 5], we expect this model to have a columnar phase at high temperature, and a quasicrystalline phase at low temperature, with a transition between them at some unknown temperature T_c . Now considering the same model except at lower volume fraction or chemical

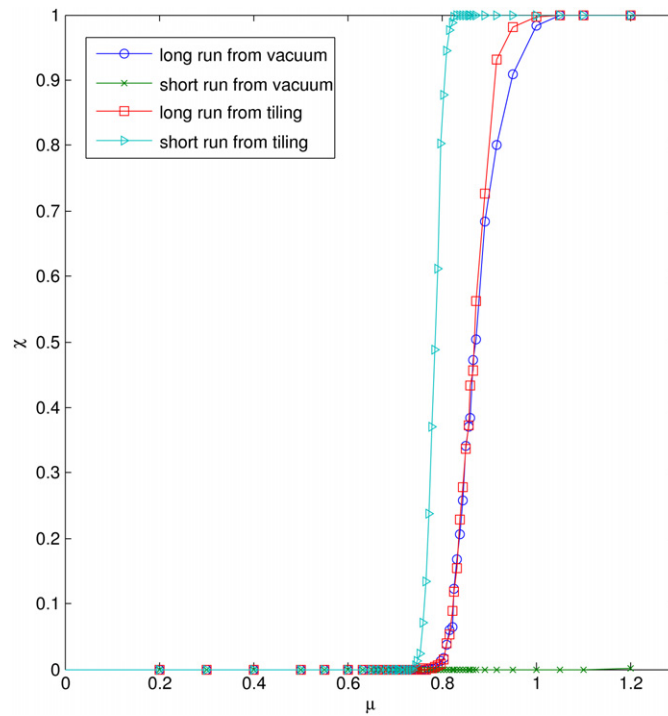


Figure 8. Comparison of order parameter values of long and short runs starting from both vacuum and perfect tiling, for $\beta = 1.5$, $w = 1$ and $V = 100^2$. (The long runs are 100 times as long as the short runs; the short runs have 10^8 Monte Carlo steps.)

potential, one expects the transition to the quasicrystalline phase to move to lower temperature since the soft interaction should have less effect as the density is lowered. We conclude that above temperature T_c our model should behave qualitatively like hard squares, with disordered and columnar phases, but without a phase with quasicrystalline order.

Absent from the above discussion is the effect on entropy, in our model, of the multiple types of particle. This effect is emphasized in Widom–Rowlinson models, which we now discuss. The multitype Widom–Rowlinson model with q colors, on $\mathbb{Z}^2$, is defined by the hard core condition that nearest neighbor sites cannot be occupied by different colors. The main conclusion of such work is the existence (for large enough q) of two types of high density phase transitions: one transition at which spatial symmetry is broken, producing different population densities for the two sublattices, but without breaking the symmetry between particle types, and a second (discontinuous) transition at higher density at which the symmetry between particle types is broken, so that one particle type dominates the density [11]. These models, with diamond-shaped instead of square hard cores, are not simply related to our models, but there are two interesting variants treated by Georgii and Zagrebnov [12] which are closely related. The first model has a square-shaped hard core, with square width $w = 2$, between particles of different color; the second model adds to this a smaller diamond-shaped hard core between nearest neighbor particles of the same color. One could think of these models as employing a hard core of size $w = 2$ between *all* particles, with a ‘cancelling’ (or attractive) interaction between particles of like color on sites at separation $\sqrt{2}$ for the second model, and for sites at separation 1 and $\sqrt{2}$ for the first model. Thus the models can be thought of

as hard squares with an added particle-type-dependent short range attraction, somewhat like our quasicrystal models. Both Widom–Rowlinson models show only one transition, which is discontinuous. For the first model the high density phase breaks particle type symmetry but not spatial symmetry, and for the second model the high density phase breaks the symmetry of both space and particle type. In particular the second model seems close to our ($w = 2$) quasicrystal model, and our results bear this out, with a discontinuous transition corresponding to broken spatial and particle-type symmetries. The correspondence might be closer if the attraction and repulsion in our model was a hard rather than soft interaction, as in [13].

It would have been useful to simulate our model over a range of temperatures, but we found this prohibitively expensive in computation time. For this reason we investigated a technique noted in [14], in which the initial state for the simulation is an energy ground state. However this technique proved to be unreliable here unless the runs were of comparable length to those starting from vacuum, thus yielding no advantage (see figure 8). (We suspect that simulations starting from an energy ground state get restricted to a narrow portion of phase space more easily than those starting from the vacuum.)

5. Conclusion

We have introduced an extension of a two dimensional lattice gas model of a quasicrystal [4, 5]. That model was studied as a function of temperature at fixed density 1, with simulations showing a continuous phase transition between a disordered (fluid) phase and a phase with quasicrystalline order. When we introduce vacancies into the model we find that the basic character does not seem to change: there still appears to be only the two phases, disordered and quasicrystalline, with a continuous transition between them.

We then generalized the model by extending the range of the hard core as well as allowing variation in the density, and found, by simulation along a (low temperature) isotherm, that the model then exhibits a *discontinuous* transition between a low density disordered and high density quasicrystalline phase. The high density quasicrystalline phase breaks symmetries of space (a square sublattice is preferentially populated) and particle type (the particles inherit the quasicrystalline boundary structure). Along an isotherm at sufficiently high temperature we expect the model to exhibit the transition of simple hard squares (continuous for $w = 2$ and possibly discontinuous for $w = 3$), between the low density disordered and high density columnar phases. At fixed high density or chemical potential we expect a transition from the quasicrystalline phase to a columnar phase as temperature is raised from zero.

This family of models with unusual symmetry adds a special flavor to the more traditional lattice gas models with hard core. Its complicated multiparticle structure seems at present to defy proof of a transition, and is also expensive to simulate. Further progress in filling out the phase structure of the model would be highly desirable.

Acknowledgment

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