Asymptotic Behavior of the Isotropic-Nematic and Nematic-Columnar Phase Boundaries for the System of Hard Rectangles on a Square lattice

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A system of hard rectangles of size $m \times mk$ on a square lattice undergoes three entropy driven phase transitions with increasing density for large enough aspect ratio $k$: first from a low density isotropic to an intermediate density nematic phase, second from the nematic to a columnar phase, and third from the columnar to a high density sublattice phase. In this paper we show, from extensive Monte Carlo simulations of systems with $m = 1, 2$ and $3$, that the transition density for the isotropic-nematic transition is $\approx A_1/k$ when $k \gg 1$, where $A_1$ is independent of $m$. We estimate $A_1 = 4.80 \pm 0.05$. Within a Bethe approximation, we obtain $A_1 = 2$ and the virial expansion truncated at second virial coefficient gives $A_1 = 1$. The critical density for the nematic-columnar transition when $m = 2$ is numerically shown to tend to a value less than the full packing density as $k^{-1}$ when $k \to \infty$. We find that the critical Binder cumulant for this transition is non-universal and decreases as $k^{-1}$ for $k \gg 1$. However, the transition is shown to be in the Ising universality class.

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I. INTRODUCTION

Hard core lattice gas models of particles interacting only through excluded volume interaction continue to be of interest in Statistical Physics. They are minimal models to study entropy driven phase transitions, have direct realizations in adsorption of gas particles on metal surfaces\cite{1,2}, and are closely related to the freezing transition\cite{3,4}, directed and undirected animals\cite{5,6} and the Yang-Lee singularity\cite{7}. Systems of differently shaped particles on different lattices have been studied both analytically and numerically. Examples include squares\cite{8,9}, hexagons\cite{10,11}, dimers with nearest neighbor exclusion\cite{12}, triangles\cite{13}, tetrominoes\cite{14}, rods\cite{15,16}, rectangles\cite{17}, lattice models for discs\cite{18,19}, and mixtures\cite{20}.

In this paper, we focus on the system of hard rectangles of size $m \times mk$ on the square lattice, where each rectangle occupies $m$ sites along the short axis and $mk$ sites along the long axis, where $k$ is the aspect ratio, and $m$, $k$ are integers. In recent times, there has been renewed interest in this problem when, even though both the low density and the maximal density phases are known to be disordered\cite{31}, the existence of a nematic phase for the hard rod ($m = 1$) system was convincingly demonstrated in simulations for $k \geq 7$\cite{24}, and thereafter proven rigorously for $k \gg 1$\cite{31}. The maximal density phase being disordered implies the existence of a second entropy driven transition with increasing density. This has been established in simulations\cite{25}. While the first transition into the nematic phase is in the Ising universality class\cite{32}, the universality class of the second transition into the high density disordered phase is not that clear\cite{26,33}.

The phase diagram for rectangles with $m > 1$ is even richer, and was recently determined using Monte Carlo simulations for $m = 2, 3$ and $k \leq 7$, and generalized to larger $m,k$ using entropic arguments\cite{26}. For $k \geq 7$, with increasing density, the system transits successively from isotropic (I) to nematic (N) to columnar (C) to solid-like sublattice (S) phases (see Sec. II for a precise definition of the phases). When $k < 7$, the $N$ phase is absent and the system makes a direct transition from the I phase to a C phase, if present. The C phase is absent only for $m = 2$ and $k = 2, 3$, and the system makes a direct transition from the I phase into the S phase. A detailed study of the nature of the transitions may be found in Ref.\cite{26}.

In this paper, we focus on the asymptotic behavior of the isotropic-nematic (I-N) and nematic-columnar (N-C) phase boundaries for large aspect ratio $k$. It was heuristically argued in Ref.\cite{26} that the limit $k \to \infty$ keeping $m$ fixed should correspond to the limit of oriented lines in the continuum, and thus the critical density should be independent of $m$. For this limiting case in three dimensions, the virial expansion truncated at the second virial coefficient becomes exact and the critical density for the I-N transition $\rho^{I-N}_c \approx A_1 k^{-1}$\cite{34,35}. $A_1$ for oriented long rectangles in the two-dimensional continuum can be directly estimated by simulating oriented lines of length $\ell$, for which it is straightforward to show that the critical number density $\approx A_1 \ell^{-2}$. From the simulations of this system with $\ell = 1$, it can be inferred that $A_1 \approx 4.84$\cite{37}. For $m = 1$, by simulating systems with $k$ up to $12$ on the lattice, it has been shown that $\rho^{I-N}_c \propto k^{-1}$\cite{32}. From the value of the critical density for $k = 10$\cite{38}, it can be estimated that $A_1 \approx 5.02$, different from that for oriented lines\cite{37}. There are no such similar studies for $m > 1$.

For the N-C transition, the limit $k \to \infty$ keeping $m$ fixed, corresponds to asking whether oriented long rectangles in the continuum exhibit a C phase. The N-C transition has been studied using a Bethe approxima-
tion that predicts the critical density to be \( \rho_c^{N-C} = A_2(m) + A_3(m) k^{-1} \) when \( k \to \infty \), where \( A_2(m) \leq 1 \) [26]. The approximations being ad-hoc, it is important to have numerical confirmation of these results, but none exists. The other limit \( m \to \infty \), keeping \( k \) fixed corresponds to the continuum problem of oriented rectangles of aspect ratio \( k \), a model that was introduced and studied by Zwanzig using virial expansion [23]. This limit is difficult to study numerically on the square lattice.

In this paper, by simulating systems of rectangles with aspect ratio \( k \) up to 60 (for \( m = 1 \)), and \( k = 56 \) (for \( m = 2 \) and 3), we show that \( \rho_c^{I-N} \) is proportional to \( k^{-1} \) for \( m = 1, 2 \) and 3. Within numerical error, \( A_1 \) is shown to be independent of \( m \) and equal to 4.80 ± 0.05. To understand better the limit of large \( k \), we study the I-N transition using a Bethe approximation, and a virial expansion truncated at the second virial coefficient. The critical density \( \rho_c^{I-N} \) is obtained for all \( m \) and \( k \). For large \( k \), both theories predict that \( \rho_c^{I-N} \) is independent of \( m \), while \( A_1 = 2 \) in the Bethe approximation and \( A_1 = 1 \) in the truncated virial theory. For the N-C transition, we numerically determine \( \rho_c^{N-C} \) for \( m = 2 \) and \( k \) up to 24. We show that for large \( k \), \( \rho_c^{N-C} \approx 0.73 + 0.23k^{-1} \), consistent with the calculations in Ref. [26]. This shows that a system of oriented rectangles with large aspect ratio in the two dimensional continuum should exhibit both nematic and columnar phases. In addition, we find that the Binder cumulant at the N-C transition is surprisingly dependent on \( k \), and decreases as \( k^{-1} \) with increasing \( k \). However, we show that the transition remains in the Ising universality class.

The rest of the paper is organized as follows. Section II contains a definition of the model, a brief description of the Monte Carlo algorithm, and a definition of the phases and the relevant thermodynamic quantities of interest. In Sec. III we present the numerical results for the I-N transition for \( m = 1, 2 \) and 3. In Sec. IV we numerically determine the asymptotic behavior of the N-C phase boundary for \( m = 2 \) and large \( k \). The Binder cumulant is shown to be non-universal, though exponents continue to be universal. Section V contains calculations of the I-N phase boundary using an ad-hoc Bethe approximation and a truncated virial expansion. Section VI contains a summary and discussion of results.

II. MODEL AND DEFINITIONS

We consider a system of monodispersed hard rectangles of size \( m \times mk \) on a square lattice of size \( L \times L \), with periodic boundary conditions. Each rectangle occupies \( m \) sites along the short axis and \( mk \) sites along the long axis, such that \( k \) is the aspect ratio. A rectangle is called horizontal or vertical depending on whether the long axis is along the x-axis or y-axis. No two rectangles may overlap, or equivalently a lattice site may be occupied by utmost one rectangle. We associate an activity \( z = e^\mu \) to each rectangle, where \( \mu \) is the chemical potential.

We simulate the system in the constant \( \mu \) grand canonical ensemble using an efficient algorithm that involves cluster moves. The implementation of the algorithm for the system of hard rectangles is described in detail in Ref. [26]. The algorithm has been shown to be very useful in equilibrating hard core systems of extended particles at high densities. Other implementations of the algorithm include lattice models of hard rods [25, 39] and hard discs [28] and mixtures of dimers and hard squares [24].

The data presented in the paper corresponds to systems with aspect ratio up to 60 and system sizes up to \( L = 1680 \). In the simulations, we ensured equilibration by confirming that the final state is independent of the initial configuration. The system is typically equilibrated for \( 10^7 \) Monte Carlo steps, and the measurement is broken into 10 statistically independent blocks, each of size \( 10^6 \) Monte Carlo steps.

The system of hard rectangles may exist in one of the four different phases: isotropic (I), nematic (N), columnar (C) and sublattice (S) [20]. In the low density I phase the system neither possess orientational order nor positional order. The N phase breaks the orientational symmetry by preferring a particular orientation, either horizontal or vertical. However, the N phase has no positional order. In the C phase, along with orientational order, the system possesses partial positional order in the direction perpendicular to the preferred orientation. In the C phase of \( 2 \times 2k \) rectangles, if most of the rectangles are horizontal (vertical), then the heads (bottom left corner) of the rectangles mostly occupy either even or odd rows (columns). The high density S phase has positional order along both horizontal and vertical directions, but no orientational order. In this phase the heads of the rectangles preferentially occupy one of \( m^2 \) sublattices [20].

We now define the order parameters and relevant thermodynamic quantities used to study the I-N and the N-C transitions. For the I-N transition, we define an order parameter

\[
Q_1 = \frac{\langle |N_h - N_v| \rangle}{\langle N_h + N_v \rangle},
\]

(1)

where \( N_h \) and \( N_v \) are the number of horizontal and vertical rectangles respectively. \( Q_1 \) is zero in the I phase and nonzero in the N and C phases. For the N-C transition, we define an order parameter only for \( m = 2 \) as our simulations are restricted to this value of \( m \). Generalization to larger \( m \) is straightforward [26]. Let

\[
Q_2 = \frac{\langle |N_{re} - N_{ro}| - |N_{ce} - N_{co}| \rangle}{\langle N_h + N_v \rangle},
\]

(2)

where \( N_{re} (N_{ro}) \) is the number of rectangles whose heads are in the even (odd) rows, and \( N_{ce} (N_{co}) \) is the number of rectangles whose heads are in the even (odd) columns. In the I and N phases, \( N_{re} \approx N_{ro} \), and \( N_{ce} \approx N_{co} \), and
hence \( Q_2 \) is zero. In the C phase, either \( N_{re} \neq N_{ro} \) and \( N_{ce} \approx N_{co} \), or \( N_{re} \approx N_{ro} \) and \( N_{ce} \neq N_{co} \), such that \( Q_2 \) is non-zero.

The second moment of the order parameter \( \chi_i \) and the Binder cumulant \( U_i \) are defined as,

\[
\chi_i = \langle Q_i^2 \rangle L^2, \quad (3a)
\]
\[
U_i = 1 - \frac{\langle Q_i^4 \rangle}{3\langle Q_i^2 \rangle^2}, \quad (3b)
\]

where \( i = 1, 2 \). The thermodynamic quantities become singular at the transition. Let \( \epsilon = (\mu - \mu_c)/\mu_c \), where \( \mu_c \) is the critical chemical potential. The singular behavior is characterized by the critical exponents \( \beta, \gamma, \nu \) defined by \( Q_i \sim (-\epsilon)^\beta, \epsilon < 0, \chi_i \sim |\epsilon|^{-\gamma}, \) and \( \xi_i \sim |\epsilon|^{-\nu} \), where \( \xi_i \) is the correlation length, \( |\epsilon| \to 0 \), and \( i = 1, 2 \). The critical exponents may be obtained numerically through finite size scaling near the critical point:

\[
U_i \approx f_u(\epsilon L^{1/\nu}), \quad (4a)
\]
\[
Q_i \approx L^{-\beta/\nu} f_q(\epsilon L^{1/\nu}), \quad (4b)
\]
\[
\chi_i \approx L^{\gamma/\nu} f_\chi(\epsilon L^{1/\nu}), \quad (4c)
\]

where \( f_u, f_q, \) and \( f_\chi \) are scaling functions.

III. ASYMPTOTIC BEHAVIOR OF THE ISOTROPIC–NEMATIC PHASE BOUNDARY: NUMERICAL STUDY

In this section we investigate the asymptotic behavior of the I-N phase boundary for \( m = 1, 2, \) and 3 by numerical simulations and show that \( \rho_{c}^{I-N} = A_1 k^{-1} \) when \( k \gg 1 \), where \( A_1 \) is independent of \( m \). Since there are two symmetric N phases (horizontal and vertical), the I-N transition for the system of hard rectangles is continuous and belongs to the Ising universality class for all \( m \). We determine the critical density \( \rho_{c}^{I-N} \) from the point of intersection of the curves of Binder cumulant with density for different system sizes. A typical example is shown in Fig. 1 where the variation of \( U_i \) with density \( \rho \) is shown for three different system sizes when \( m = 1 \) and \( k = 32 \). The Binder cumulant data are fitted to a cubic spline to obtain a smooth and continuous curve for each \( L \). This allows us to determine the point of intersection or \( \rho_{c}^{I-N} \) more accurately. In the example shown in Fig. 1 the curves for Binder cumulants for three different system sizes cross at \( \rho = \rho_{c}^{I-N} \approx 0.152 \) and the value of the critical Binder cumulant \( U_i^c \approx 0.61 \). We find \( U_i^c \approx 0.61 \) for all values of \( m \) and \( k \) that we have studied, consistent with the value for the two dimensional Ising model [10].

We simulate systems with aspect ratio up to \( k = 60 \) for \( m = 1 \) and \( k = 56 \) for \( m = 2 \) and 3. The critical density \( \rho_{c}^{I-N} \) obtained from the Binder cumulants are shown in Fig. 2. The data are clearly linear in \( k^{-1} \) for large \( k \), confirming that \( \rho_{c}^{I-N} = A_1 k^{-1} \), \( k \gg 1 \). In addition, the data for \( m = 1, 2, 3 \) asymptotically lie on the same straight line, showing that \( A_1 \) is independent of \( m \). We estimate \( A_1 = 4.80 \pm 0.05 \).

IV. ASYMPTOTIC BEHAVIOR OF THE NEMATIC–COLUMNAR PHASE BOUNDARY: NUMERICAL STUDY

In this section, we numerically study the N-C phase transition for \( m = 2 \) and determine the asymptotic behavior of the critical density \( \rho_{c}^{N-C} \) for large \( k \). When \( m = 2 \), the N-C transition belongs to the Ising universality class for all \( k \) and the corresponding critical densities are determined from the intersection of Binder cumulant curves for different system sizes as discussed in Sec. III.

The critical density \( \rho_{c}^{N-C} \) decreases to a constant with increasing \( k \) (see Fig. 3). We obtain \( \rho_{c}^{N-C} \approx \)

FIG. 1. (Color online) The variation of the Binder cumulant \( U_i \) with density \( \rho \) for three different system sizes. The lines are cubic splines, fitted to the data. The value of \( U \) at \( \rho = \rho_c \) is \( \approx 0.61 \). The data are for \( m = 1 \) and \( k = 32 \).

FIG. 2. (Color online) The variation of the critical density for the I-N transition \( \rho_{c}^{I-N} \) with \( k^{-1} \) for \( m = 1, 2 \) and 3. The straight line is \( 4.80 k^{-1} \).
$\beta/\nu$ curve when scaled as in Eq. (4) with Ising exponents for the Ising universality class. The data for the critical Binder cumulant is approximately those of the two dimensional Ising model. To do so, we use the ratio of the underlying lattice \[40\]. In the latter case, the critical Binder cumulant is a function of the aspect ratio of the isotropic Ising model on rectangular lattice, where coupling constants along the $x$ and $y$ directions \[41\], and the isotropic Ising model on rectangular lattice, where the critical Binder cumulant is a function of the aspect ratio of the underlying lattice \[40\]. In the latter case, $U_2^c \approx 2.46\alpha^{-1}$, where $\alpha$ is the aspect ratio of the lattice \[40\]. Thus, nominally $k \approx 1.8\alpha$.

Although $U_2^c$ varies with $k$, we confirm that the critical exponents for the N-C transition remains the same as those of the two dimensional Ising model. To do so, we determine the critical exponents for the system with $m = 2$ and $k = 13$ using finite size scaling. For this example, critical Binder cumulant is $\approx 0.35$, noticeably different from that for the Ising universality class. The data for the Binder cumulant $U_2$ for different system sizes intersect at $\mu_c \approx 1.00$ [see fig. 3(a)]. We find that the data for $U_2$, $Q_2$ and $\chi_2$ for different system sizes collapse onto a single curve when scaled as in Eq. (4) with Ising exponents $\beta/\nu = 1/8$, $\gamma/\nu = 7/4$, and $\nu = 1$ [see Fig. 3(b)–(d)]. We thus conclude that, though the critical Binder cumulant is non-universal, the transition is in the Ising universality class.

V. ESTIMATION OF THE I-N PHASE BOUNDARY USING ANALYTICAL METHODS

In this section we obtain the asymptotic behavior of the isotropic-nematic phase boundary for large $k$ using analytical methods. In the absence of an exact solution, we present two approximate calculations: first a Bethe approximation:
approximation and second a virial expansion truncated at the second virial coefficient.

A. Bethe Approximation

The Bethe approximation becomes exact on tree like lattices. For \( m = 1 \), the model was solved exactly on the 4-coordinated random locally tree like layered lattice (RLTL) to obtain \( \rho_{I-N}^{-N} = 2/(k-1) \) \( 33 \) or \( A_1 = 2 \). The RLTL also allows an exact solution to be obtained for more complicated systems like repulsive rods \( 33 \). However, a convenient formulation of the problem of hard rectangles on the RLTL is lacking. Therefore, we resort to an ad-hoc Bethe approximation introduced by DiMarzio to estimate the entropy of hard rods on a cubic lattice \( 41 \), and later used for studying the statistics of hard rods on different lattices \( 38, 45, 46 \). However, a straightforward extension of this method to a system of rectangles suffers from the enumeration result depending on the order in which the rectangles are placed. A scheme that overcomes this shortcoming was suggested in Ref. \( 47 \) and was implemented by us to study the N-C transition \( 26 \). Here, we adapt the calculations to study the I-N transition.

The I-N phase boundary can be determined if the entropy as a function of the densities of the horizontal and vertical rectangles is known. We estimate the entropy by computing the number of ways of placing \( N_x \) horizontal and \( N_y \) vertical rectangles on the lattice.

First, we place the horizontal rectangles on the lattice one by one. Given that \( j_x \) horizontal rectangles have been placed, the number of ways of placing the \((j_x + 1)^{th}\) horizontal rectangle may be estimated as follows. The head of the rectangle may be placed in one of the \((m - m^2 k_{j_x})\) empty sites, where \( M \) is the total number of lattice sites. We denote this site by \( A \) (see Fig. 6). For this new configuration to be valid, all sites in the \( m \times m k \) rectangle with head at \( A \) should be empty. Given \( A \) is empty, we divide the remaining \((m^2 k - 1)\) sites in three groups: \((m k - 1)\) sites along the line \( AB \), \((m - 1)\) sites along the line \( AC \), and the remaining \((m - 1)(m k - 1)\) sites \( (D \) is an example). Let \( P_x(B|A) \) be the conditional probability that \( B \) is empty given that \( A \) is empty. Then the probability that \((m k - 1)\) sites along the line \( AB \) are empty is \([P_x(B|A)]^{m k - 1}\), where the subscript \( x \) denotes the direction \( AB \). In writing this, we ignore all correlations beyond the nearest neighbor. Likewise, the probability that \((m - 1)\) sites along the line \( AC \) are empty is \([P_y(C|A)]^{m - 1}\), where \( P_y(C|A) \) is the conditional probability that \( C \) is empty given \( A \) is empty. Let \( P(D|B \cap C) \) denote the conditional probability that \( D \) is empty given that \( B \) and \( C \) are both empty. Then, the probability that the remaining \((m - 1)(m k - 1)\) sites are empty may be approximated by \([P(D|B \cap C)]^{(m - 1)(m k - 1)}\). Collecting these different terms together, we obtain the number of ways to place the \((j_x + 1)^{th}\) horizontal rectangle

\[
\nu_{j_x+1} = (M - m^2 k_{j_x}) \times [P_x(B|A)]^{m k - 1} [P_y(C|A)]^{m - 1} \times [P(D|B \cap C)]^{(m - 1)(m k - 1)}.
\]  

(5)

It is not possible to determine these conditional probabilities exactly. However, they may be estimated by assuming that the rectangles are placed randomly. Given \( A \) is empty, either \( B \) might be empty or occupied by a horizontal rectangle (as no vertical rectangles have been placed yet) in \( m \) ways. Thus, given \( A \) is empty, the probability that \( B \) is also empty, is

\[
P_x(B|A) = \frac{M - m^2 k_{j_x}}{M - m^2 k_{j_x} + m j_x}.
\]  

(6)

Similarly, if \( A \) is empty, \( C \) might be empty or it might be occupied by any of the \( m k \) sites on the longer axis (passing through \( C \)) of a horizontal rectangle. Thus the probability that \( C \) is empty, given \( A \) is empty is given by

\[
P_y(C|A) = \frac{M - m^2 k_{j_x}}{M - m^2 k_{j_x} + m j_x}.
\]  

(7)

Next we estimate \( P(D|B \cap C) \). If we follow a similar approach to calculate \( P(D|B \cap C) \), the resultant entropy becomes dependent on the density of the horizontal and vertical rectangles, and thus asymmetric with respect to \( N_x \) and \( N_y \). To overcome this shortcoming, we follow the Bethe approximation proposed in Ref. \( 47 \) and assume

\[
P(D|B \cap C) \approx \frac{P_x(C|D)P_y(B|D)}{P_xB(B)}.
\]  

(8)
where
\[ P_{xy}(B|C) = \frac{M - m^2kj_x}{M - m(m - 1)kj_x + (m - 1)j_x}. \] (9)
is the probability that \( C \) is empty given \( B \) is empty. It can be easily seen that
\[ P_x(C|D) = P_x(B|A), \quad (10a) \]
\[ P_y(B|D) = P_y(C|A). \quad (10b) \]

After placing \( N_x \) horizontal rectangles we would like to determine the number of ways in which \( N_y \) vertical rectangles may be placed on the lattice. Given \( N_x \) horizontal rectangles and \( j_y \) vertical rectangles have already been placed, we estimate \( \nu_{j_y+1} \), the number of ways to place the \((j_y + 1)^{th}\) vertical rectangle, using the same procedure as above. Now, we may choose an empty site \( A \) (see Fig. 6) randomly in \((M - m^2kN_x - m^2kj_y)\) ways to place the head of the \((j_y + 1)^{th}\) vertical rectangle. As the vertical rectangles have their longer axis along \( y \)-direction, it can be easily seen that
\[ \nu_{j_y+1} = (M - m^2kN_x - m^2kj_y)[P_y(C|A)]^{mk-1} \]
\[ \times [P_x(B|A)]^{m-1}[P(D|B \cap C)]^{(m-1)(mk-1)}. \] (13)

The expressions for the conditional probabilities will now be modified due to the presence of both horizontal and vertical rectangles. If \( A \) is empty, \( C \) may be empty or occupied by one of the \( mk \) sites on the long axis (passing through \( C \)) of a horizontal rectangle, or by one of the \( m \) sites on the short axis (passing through \( C \)) of a vertical rectangle. Hence, given \( A \) is empty, the probability that \( C \) is also empty is
\[ P_y(C|A) = \frac{M - m^2kN_x - m^2kj_y}{M - mk(m-1)N_x - mk(m-1)kj_y}. \] (14)

Similarly, the probability of \( B \) being empty, given \( A \) is empty, is
\[ P_x(B|A) = \frac{M - m^2kN_x - m^2kj_y}{M - m(m-1)N_x - mk(m-1)kj_y}. \] (15)

Now the probability that \( B \) is empty, given \( C \) is empty, is
\[ P_{xy}(B|C) = \frac{M - m^2kN_x - m^2kj_y}{M - (mk-1)(m-1)(N_x + j_y)}. \] (16)

\( P(D|B \cap C) \) is determined using Eqs. (8) and (10). Substituting Eqs. (14)–(16) into Eq. (13), we obtain \( \nu_{j_y+1} \). The total number of ways to place \( N_y \) vertical rectangles, given that \( N_x \) horizontal rectangles have already been placed, is then

\[ \Omega_x = \frac{1}{N_x} \prod_{j_x=0}^{N_x-1} \nu_{j_x+1}. \] (11)

Substituting Eqs. (9)–(10) into Eq. (6), we obtain \( \nu_{j_y+1} \). \( \Omega_x \) is given by

\[ \Omega_x = \frac{1}{N_x} \prod_{j_x=0}^{N_x-1} \nu_{j_x+1}. \] (12)

As all the horizontal rectangles are indistinguishable, the total number of ways to place \( N_x \) of them is

\[ \Omega = \frac{1}{N_x} \prod_{j_x=0}^{N_x-1} \nu_{j_x+1}. \] (13)

The total number of ways to place \( N_x \) horizontal and \( N_y \) vertical rectangles on the lattice is given by

\[ \Omega = \Omega_x \Omega_y. \] (18)

Let \( \rho_x \) and \( \rho_y \) be the fraction of the sites occupied by the horizontal and the vertical rectangles, given by

\[ \rho_i = \frac{m^2kN_i}{M}, \quad i = x, y. \] (19)

Using Eqs. (12) and (17), the entropy of per site in the thermodynamic limit may be expressed in terms of \( \rho_x \) and \( \rho_y \) as
The transition density for the I-N transition is denoted by \( \rho_{c}^{I-N} \).

\[
s(\rho_x, \rho_y) = \lim_{M \to \infty} \frac{1}{M} \ln \left( \Omega_x \Omega_y \right)
= - \sum_{i=x,y} \frac{\rho_i}{m^2 k} \ln \frac{\rho_i}{m^2 k} - [1 - \rho_i] \ln [1 - \rho_i] - \left[ 1 - \frac{(m - 1)(mk - 1)}{m^2 k} \rho \right] \ln \left[ 1 - \frac{(m - 1)(mk - 1)}{m^2 k} \rho \right]
+ \sum_{i=x,y} \left[ 1 - \frac{(mk - 1)}{mk} \rho + \frac{(k - 1)}{mk} \rho_i \right] \ln \left[ 1 - \frac{(mk - 1)}{mk} \rho + \frac{(k - 1)}{mk} \rho_i \right],
\]

Asymptotic behavior of \( \rho_{c}^{I-N} \) is given by

\[
\rho_{c}^{I-N} = \frac{2km}{mk^2 + m - k - 1}.
\]

Thus, \( A_1 = 2 \).

When \( m = 1 \), the critical density \( \rho_{c}^{I-N} = 2/(k - 1) \), which matches with the exact calculation of \( \rho_{c}^{I-N} \) for the system of hard rods of length \( k \) on the RLTL. It reflects that the Bethe approximations becomes exact on the RLTL. For \( m = 1 \), the nematic phase and hence the I-N transition exists for \( k \geq k_{\min} = 4 \). While for \( m = 2 \) and \( 3 \), \( k_{\min} = 3 \), for \( m \geq 4 \) the nematic phase exists even for \( k = 2 \).

### B. Virial Expansion

In this subsection we determine \( \rho_{c}^{I-N} \) using a standard virial expansion truncated at the second virial coefficient. We closely follow the calculations of Zwanzig for oriented hard rectangles in the continuum. The excess free energy of the system of hard rectangles (relative to the ideal gas) may be expressed in terms of the virial coefficients and the density. We truncate the series at the second virial coefficient and study the I-N transition in the limit \( k \to \infty \).

Consider a system of \( N \) rectangles on the square lattice of volume \( V \). Each rectangle may be oriented along two possible directions. Setting \( \beta = 1 \), the configurational sum of the system is given by

\[
Q_N = \frac{1}{N!2^N} \sum_{\mathbf{R}} \sum_{\mathbf{u}} \exp(-U_{\mathbf{R}}),
\]

where the sum over all possible positions and directions are denoted by \( \sum_{\mathbf{R}} \) and \( \sum_{\mathbf{u}} \) respectively, \( U_{\mathbf{R}} \) is the total interaction energy of all rectangles. The excess free energy
energy (relative to the ideal gas) \( \phi_N \) of the system of rectangles having fixed orientations is defined by

\[
\exp[-\phi_N(u)] = \frac{1}{V^N} \sum_{\mathcal{R}} \exp(-\beta U_N). \tag{26}
\]

As the rectangles having same orientation are indistinguishable, \( \phi_N \) depends only on the fractions of the rectangles pointing along the two possible directions. If the number of rectangles oriented along direction \( i \) is denoted by \( N_i \), we may rewrite the Eq. (25) using Eq. (26) as

\[
Q_N = \frac{V^N}{N!^2} \sum_{N_1, N_2=0}^{N} \frac{N!}{N_1!N_2!} \exp(-\phi_N(N_1, N_2)) \delta_{N_1+N_2,N} = \sum_{N_1=0}^{N} \sum_{N_2=0}^{N} W(N_1, N_2), \tag{27}
\]

where \( \delta_{N_1+N_2,N} \) takes care of the constraint that the total number of rectangles is \( N \) and \( W \) is given by

\[
W(N_1, N_2) = \frac{V^N}{2^N N_1!N_2!} \exp[-\phi_N(N_1, N_2)]. \tag{28}
\]

In the thermodynamic limit \( N \to \infty \) and \( V \to \infty \), the above summation may be replaced by the largest summand \( W_{\text{max}} \) with negligible error. Thus the configurational free energy per particle is given by

\[
f = - \lim_{N,V \to \infty} \frac{1}{N} \ln Q_N = - \lim_{N,V \to \infty} \frac{1}{N} W_{\text{max}}. \tag{29}
\]

The fractions of rectangles pointing in the \( i \)-direction is denoted by \( x_i = N_i/N \), such that \( (x_1 + x_2) = 1 \), and the number density of the rectangles is given by \( N/V = \rho/m^2k \), where \( \rho \) is the total fraction of occupied sites. Equation (29) for the free energy may be expressed in terms of \( x_1 \) and \( x_2 \) as

\[
f(x_1, x_2) = -1 + \ln 2 + \ln \rho + 2 \sum_{i=1}^{2} x_i \ln x_i + \frac{1}{N} \phi_N(\rho, x_1, x_2). \tag{30}
\]

The virial expansion of the excess free energy \( \phi_N \), for a composition \( \mathbf{x} = (x_1, x_2) \) of the rectangles is given by

\[
-\frac{1}{N} \phi_N(\rho, \mathbf{x}) = \sum_{n=2} B_n(\mathbf{x}) \left( \frac{\rho}{m^2k} \right)^{n-1}, \tag{31}
\]

where

\[
B_n(\mathbf{x}) = \frac{1}{n!} \int \prod f
= \frac{1}{n!} \int_{0}^{n} \binom{n}{j} x_1^{n-j} x_2^j B(n-j, j)
= \frac{1}{n!} \sum_{j=0}^{n} \frac{B(n-j, j)}{(n-j)!^2} x_1^{n-j} x_2^j. \tag{32}
\]

where \( \int \prod f \) is the standard abbreviation for the cluster integrals over the irreducible graphs consist of \( n \) rectangles with composition \( \mathbf{x} \) and \( f \) denotes the Mayer functions, defined as

\[
f = \exp(-U) - 1,
\]

where \( U \) is the interaction energy. Due to the hard core exclusion, we have \( U = \infty \) for any intersection or overlap among the rectangles, otherwise \( U = 0 \). Hence

\[
f = \begin{cases} 
-1, & \text{for any intersection} \\
0, & \text{otherwise}
\end{cases} \tag{34}
\]

\( B(n-j, j) \) denotes the sum of the irreducible graphs for the composition where \( (n-j) \) rectangles are oriented along the \( x \)-direction and \( j \) rectangles are along the \( y \)-direction.

As the total fraction \( x_1 + x_2 = 1 \), we set

\[
x_1 = x,
\]

\[
x_2 = 1 - x. \tag{35}
\]

We consider up to the second virial coefficient and truncate the expansion in Eq. (31) at first order in \( \rho \). From the definition of the virial coefficients in Eq. (32), we can easily infer that they are symmetric in the following way:

\[
B(n_1, n_2) = B(n_2, n_1). \tag{36}
\]

Using Eq. (32) and the above symmetry property of \( B(n_1, n_2) \), we can rewrite Eq. (31) as

\[
-\frac{1}{N} \phi_N \approx \frac{1}{2V} B(2, 0) \left( \frac{\rho}{m^2k} \right) (2x^2 - 2x + 1)
+ \frac{1}{V} B(1, 1) \left( \frac{\rho}{m^2k} \right) (2x - 2x^2) + O(\rho^2). \tag{37}
\]

Now we evaluate the virial coefficients. From Eq. (31) we can see that \( f \) has nonzero contributions only when the rods intersect. Thus the calculation of the virial coefficients on a lattice turns out as the problem of counting the number of disallowed configurations. By definition

\[
B(2, 0) = B(0, 2) = \int d^2 R_1 \int d^2 R_2 f_{12}(2, 0)
= -V \times (2mk - 1) \times (2m - 1), \tag{38}
\]

where \( (2mk - 1) \times (2m - 1) \) is the number of disallowed configurations when both the rectangles are oriented along the same direction [see Fig. 3(a)]. Similarly

\[
B(1, 1) = \int d^2 R_1 \int d^2 R_2 f_{12}(1, 1)
= -V \times (m + mk - 1)^2, \tag{39}
\]

where \( (m + mk - 1)^2 \) is the number of disallowed configurations when the two rectangles are oriented along different directions [see Fig. 3(b)].
The free energy reduces to

\[ f(\rho) = \frac{1}{N} \phi_N \approx \frac{1}{2} (2x^2 - 2x + 1) \rho (2m - 1)(2mk - 1) - (2x - 2x^2) \rho (m + mk - 1)^2 + O(\rho^2). \]  

Now substituting Eq. (40) in Eq. (30), the expression for \( \rho \) is minimized by solving Eq. (42) for \( B \).

The Bethe approximation and the virial theory predicts the number of diagrams are of order one. Hence, it may be determined from the N phase to a C phase and third from the C phase to a high density S phase [26]. In this paper we study the I-N and the N-C transition when \( k \gg 1 \). From extensive Monte Carlo simulations of systems with \( m = 1, 2 \) and 3, we establish that \( \rho_c^{I-N} \approx A_1/k \), for \( k \gg 1 \), where \( A_1 \) is independent of \( m \) and is estimated to be \( 4.80 \pm 0.05 \). The numerical value being consistent with that obtained from simulation of oriented lines [37]. The maximum value of \( k \) studied in the paper is 60, earlier simulations having been restricted up to \( m = 1 \) and \( k = 12 \) [48]. The I-N transition was also studied analytically using an ad-hoc Bethe approximation and a truncated virial expansion. Both these theories support the numerical result \( \rho_c^{I-N} \approx A_1/k \), for \( k \gg 1 \), where \( A_1 \) is independent of \( m \).

The preferred state at any fixed density is obtained by minimizing the free energy \( f(x) \) with respect to \( x \). For \( \rho < \rho_c^{I-N}, \) \( f(x) \) is minimized for \( x = 1/2 \), corresponding to the isotropic phase, and beyond \( \rho_c^{I-N}, \) \( f(x) \) is minimized for \( x \neq 1/2 \), corresponding to the nematic phase. Thus the system undergoes a transition from an isotropic phase to a nematic phase with increasing density. The I-N transition is found to be continuous with the critical density \( \rho_c^{I-N} \). The expansion of the free energy \( f(x) \) as a power series in \( x \) about \( x = 1/2 \) contains only even powers, and thus the critical density \( \rho_c^{I-N} \) may be determined by solving

\[ -\frac{d^2}{dx^2} f(x)|_{x=1/2} = 0. \]  

By solving Eq. (42) for \( \rho_0 \), we find

\[ \rho_c^{I-N} = \frac{2km^2}{1 - 2(1 + k)m + 2(1 + k^2)m^2}. \]  

The asymptotic behavior of \( \rho_c^{I-N} \) is given by

\[ \rho_c^{I-N} \approx \begin{cases} \frac{1}{3} + \frac{mk}{1 + mk} + O(k^{-3}), & k \to \infty, m \text{ fixed}, \\ \frac{1}{3} + \frac{k(1+k)}{1+k^2} + O(m^{-2}), & m \to \infty, k \text{ fixed}. \end{cases} \]  

VI. SUMMARY AND DISCUSSION

For \( k \geq 7 \), the system of long, hard rectangles of size \( m \times mk \) on the square lattice undergoes three entropy driven phase transitions with density: first from a low density I phase to an intermediate density N phase, second from the N phase to a C phase and third from the C phase to a high density S phase [26]. In this paper we study the I-N and the N-C transition when \( k \gg 1 \). From extensive Monte Carlo simulations of systems with \( m = 1, 2 \) and 3, we establish that \( \rho_c^{I-N} \approx A_1/k \), for \( k \gg 1 \), where \( A_1 \) is independent of \( m \) and is estimated to be \( 4.80 \pm 0.05 \). The numerical value being consistent with that obtained from simulation of oriented lines [37]. The maximum value of \( k \) studied in the paper is 60, earlier simulations having been restricted up to \( m = 1 \) and \( k = 12 \) [48]. The I-N transition was also studied analytically using an ad-hoc Bethe approximation and a truncated virial expansion. Both these theories support the numerical result \( \rho_c^{I-N} \approx A_1/k \), for \( k \gg 1 \), where \( A_1 \) is independent of \( m \).

The Bethe approximation, while taking into account nearest neighbor correlations, ignores other correlations and there appears to be no systematic way of improving the calculations to obtain better estimates of \( A_1 \). On the other hand, the virial expansion truncated at the second virial coefficient is known to become exact in three dimensions when \( k \to \infty \). But in two dimensions, higher order virial coefficients contribute significantly. To confirm this, we computed the higher order virial coefficients. As \( B_2 \sim k^2 \) [see Eqs. (49)], in the limit \( k \to \infty, B_2 \to \rho/k \sim O(1) \). We can rewrite Eq. (41) as

\[ -\frac{1}{N} \phi_N(x) \approx B_2(x) \frac{\rho m^2 k}{k^2} + B_3(x) \frac{\rho m^2 k}{k^3} + B_4(x) \frac{\rho m^2 k}{k^4} + O(\rho^4). \]  

When \( k \gg 1 \), it can be verified that \( B_3 \sim O(k^3) \) and hence \( B_3/[B_2]^2 \sim O(1/k) \). Quite interestingly we find \( B_4 \sim O(k^6) \) and \( B_4/[B_2]^3 \sim O(1) \). Thus \( B_4 \) will have non negligible contribution to \( \rho_c^{I-N} \). In general, \( B_2 \sim O(k^{4n-2}) \), implying all the even virial coefficients will have non negligible contributions. Usually, the number of diagrams required to compute higher order virial coefficients increase rapidly with order. However, here the number of diagrams are of order one. Hence, it may be possible to determine \( A_1 \) exactly by taking into account all the even virial coefficients.

We also numerically investigated the asymptotic behavior of \( \rho_c^{N-C} \) for \( m = 2 \) and find \( \rho_c^{N-C} \approx 0.73 + 0.23k^{-1} \) when \( k \gg 1 \), which is in qualitative agreement with the prediction of the Bethe approximation: \( \rho_c^{N-C} \approx A_2 + A_3/k \), for \( k \gg 1 \), presented in Ref. [26]. For larger \( m \), we expect the transition to become first order, however the asymptotic results is likely to be qualitatively the same. Taking the limit \( k \to \infty \) keeping \( m \) fixed
corresponds to a system of thin, long hard rectangles in the continuum. Thus, we expect the N-C transition to persist in continuum models.

Density functional theory calculations for a system of hard rectangles with restricted orientation in the continuum, confined in a two dimensional square nanocavity, predicts that the system will exhibit nematic, smectic, columnar and solid like phases, where the solid-like phase has both orientational and complete positional order [48]. In contrast, we do not find any evidence of smectic or solid-like phases when \( m \) or \( k \) tend to \( \infty \), the continuum limit. In particular, on lattices the maximal density phase of a monodisperse system does not have orientational order [24, 50]. It would therefore be important to verify the phase diagram of hard rectangles with restricted orientation in two dimensional continuum through direct numerical simulations, similar to the simulations for rectangles with continuous orientation [49–52].

We showed that the critical Binder cumulant for the N-C transition decreases as \( k^{-1} \) with increasing the aspect ratio \( k \) of the rectangles. The critical Binder cumulant in the Ising model on rectangular geometry decreases as \( \alpha^{-1} \), where \( \alpha \) is the aspect ratio of the lattice [40]. Whether a mapping between \( k \) and \( \alpha \) exists is an open question. Curiously, the critical Binder cumulant is zero when \( k \rightarrow \infty \) (or \( \alpha \rightarrow \infty \)). In the Ising model, this has been interpreted as the absence of transition on one-dimensional geometries [40]. However, the hard rectangle system shows a transition at \( k \rightarrow \infty \). It is possible that in this limit, the fluctuations at the transition become gaussian.

The critical density for the high-density C-S transition was argued to be of the form \( 1 - a/(mk^2) \) for \( k \gg 1 \), where \( a \) is a constant [24]. However, we could not numerically verify this claim as it becomes difficult to equilibrate the system at densities close to one due to the presence of long-lived metastable states. Thus, the Monte Carlo algorithm needs further improvement. One possible direction is the modification suggested in Ref. [29], where fully packed configurations are simulated using transfer matrices.

The hard rectangle model may be generalized in different directions. Including attractive interaction results in phases with broken orientational and transitional symmetry even for dimers [53, 54]. Such phases may also be seen in mixtures of hard particles, for example dimers and squares [29]. Another generalization is to study polydisperse systems. In the continuum, polydispersity may result in reentrant nematic phase or two distinct nematic phases [55, 56]. It would be interesting to see which features persist in the lattice version of rods [57] or rectangles. These are promising areas for further study.

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