

In pursuit of the tetratic phase in hard rectangles

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We numerically investigate two-dimensional systems of hard rectangles at constant pressure through extensive hard-particle Monte Carlo simulations. We determine the complete phase diagram as a function of packing fraction and aspect ratio, which consists of four distinct phases. At very high packing fractions, particles assemble into a smectic phase for all aspect ratios. Rodlike particles with large aspect ratio assemble into an intervening nematic phase, which is displaced by a “tetratic” phase (also called biaxial nematic) for moderately elongated rectangles. Surprisingly, we find evidence that the transition from tetratic to smectic is weakly discontinuous at variance with previously proposed two-step scenarios for the melting of hard particles.

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The emergence of order in microscopic assemblies of hard bodies driven entirely through entropy is a fascinating phenomenon defying the widespread notion that associates entropy with chaos and disorder. The prediction that hard spheres order into a regular crystal structure is one of the early triumphs of computational statistical mechanics [1,2], which was confirmed by the ground breaking experiments of Pusey and van Megen [3]; for a comprehensive review see also [4]. Even earlier, Onsager predicted that hard elongated rods can develop orientational order while positions remain disordered, which highlights the importance of shape [5]. More recent work demonstrates the wide variety of structural and dynamic phases that are unlocked through varying particle shape [6–11].

Another crucial factor is dimensionality [12,13], with two-dimensional materials displaying a number of peculiar properties when compared to their bulk behavior. Indeed, already for hard disks the phase diagram is more involved compared to three dimensions with a hexatic phase intervening between fluid and solid [14,15]. The precise nature of the phase transitions has been a long-standing matter of debate until settled through extensive numerical simulations to be discontinuous (fluid-hexatic) and continuous (hexatic-solid) [16–18]. Anderson and coworkers have performed a comprehensive numerical investigation for regular n -gons [11]. Besides the disklike scenario for large $n \geq 7$ they also find a discontinuous transition from fluid to solid without intervening phase (for hard pentagons with $n = 5$, see also [19,20]) and two continuous transitions (corresponding to the original scenario of the Kosterlitz-Thouless-Halperin-Nelson-Young theory [21,22]) for triangles, squares, and hexagons. Experimentally, granular [23] and colloidal particles with complex shapes [24] have been studied.

The arguably simplest shape in two dimensions is the hard rectangle. While the limits of (rounded) squares [11,25,26] and elongated rods [5,27,28] have been studied extensively, intermediate aspect ratios α have received far less attention. Theoretical phase diagrams based on scaled particle theory [29,30] and density functional theory [31] have been constructed. Partial numerical phase diagrams have been reported and compared to density functional theory [31] and experiments [32] but, somewhat surprisingly, there is no comprehensive numerical phase diagram covering the full density range for small to intermediate aspect ratios and discussing the order of transitions. Here we fill this gap and provide a complete phase diagram as a function of packing fraction and particle aspect ratio featuring four phases that are illustrated in Fig. 1. Intriguingly, we see a clear delineation of a genuine “tetratic” phase [Fig. 1(c)] characterized by a distribution of orientations that shows two peaks. For squares ($\alpha = 1$), Wojciechowski and Frenkel hypothesized that such a phase with fourfold neighbor symmetry would take the place of the more familiar hexatic phase for disks (which have six neighbors) [33]. Donev *et al.* have performed simulations of “dominos” ($\alpha = 2$) and found clear evidence for tetratic order between fluid and solid [34]. In earlier studies of “discorectangular” particles a tetratic phase was not detected [27]. Experimentally, effective hard rectangles have been realized as standing disks on a substrate [35] and vibrated granular matter [32,36], for which tetratic order has been reported. Interest in the precise phase behavior of hard particles in two dimensions is rekindled by the recent progress on the self-assembly of DNA-based tiles [37–39]. Grafted to lipid membranes and endowed with local and specific interactions through DNA hybridization, they open new avenues to functional synthetic nanostructures.

We study N identical rectangular particles with aspect ratio α and fixed area σ^2 at constant pressure p moving in two dimensions with periodic boundary conditions [40]. If not stated otherwise, $N \approx 1300$ depending on the box aspect ratio to guarantee a defect-free initial configuration. Particles only interact through their excluded volume and

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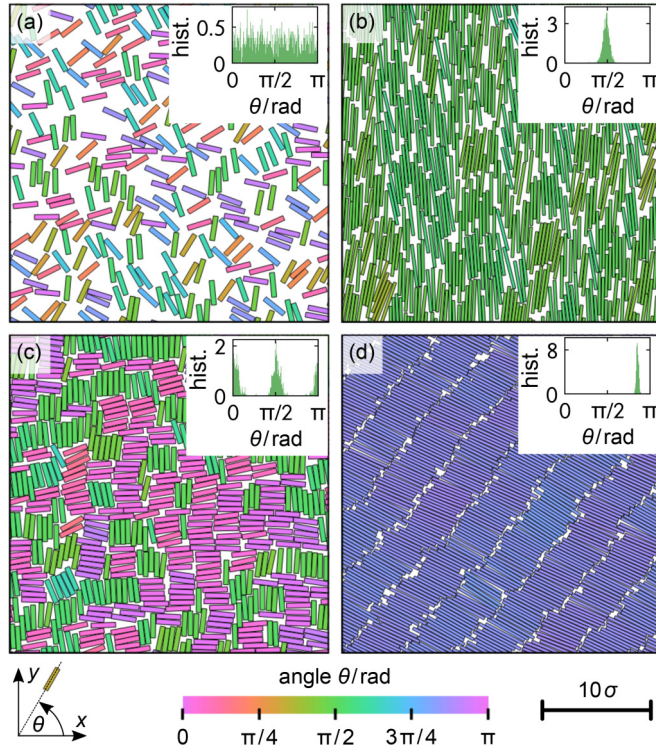


FIG. 1. Simulation snapshots for the four observed phases: (a) isotropic (I) with $\alpha = 5, p = 1.3, \phi \simeq 0.347$, (b) nematic (N) with $\alpha = 15, p = 5.0, \phi \simeq 0.596$, (c) tetratic (T) with $\alpha = 5, p = 8.8, \phi \simeq 0.730$, and (d) smectic (S) with $\alpha = 15, p = 12.8, \phi \simeq 0.812$. The insets show the distribution of the angle θ enclosed with the x axis. Notably, the tetratic phase (c) exhibits two equal peaks, whereas the nematic (b) and smectic (d) phases show the same orientational order and are distinguished by the breaking of translational symmetry in the smectic phase.

we perform extensive hard-particle Monte Carlo simulations utilizing HOOMD-blue [41–44]. Throughout, we employ dimensionless quantities with length unit σ and energies in units of the thermal energy $k_B T$. A single configuration thus comprises the centroid positions $\mathbf{r}_k$ and the angles $0 \leq \theta_k < \pi$ enclosed with the x axis for each particle. Three protocols are implemented distinguished by their initial state: (i) $\mathcal{I}_+$ starts from a disordered state and then the pressure is increased until the target pressure is reached while (ii) $\mathcal{S}_-$ starts from perfect smectic order and the pressure is decreased. (iii) In addition, for $\alpha = 5$ we prepare a perfect “checkerboard” within the tetratic phase (at $\phi \simeq 0.72$) as initial configuration (protocol $\mathcal{T}_\pm$). The checkerboard unit cell is composed of 4×5 rectangles as follows: Always five particles form a small group where all particles point in the same direction, while the nearest-neighbor groups are aligned in the perpendicular direction. All protocols prepare configurations at a target pressure through compressing/expanding a protocol-dependent initial configuration. This is followed by a relaxation run of τ_r Monte Carlo steps, after which we collect configurations for analysis (see Supplemental Material [45] for further details).

To characterize single configurations harvested from the simulations, we employ three order parameters. The first

two are

$$\hat{\psi}_\nu = \left| \frac{1}{N} \sum_{k=1}^N e^{i\nu\theta_k} \right|^2 \quad (1)$$

with $\nu = 2, 4$ and $0 \leq \hat{\psi}_\nu \leq 1$. These measure orientational order with $\hat{\psi}_\nu \simeq 0$ if the distribution of orientations is uniform. Both the nematic and smectic phase are characterized by a distribution of orientations exhibiting a single peak corresponding to the preferred orientation, the unit-length director. In the smectic phase [Fig. 1(d)], the distribution is much narrower than in the nematic phase and smectic layers form separated by a small gap. Importantly, for two peaks $\hat{\psi}_4$ becomes nonzero while $\hat{\psi}_2 \simeq 0$, whereas for a single peak both order parameters are nonzero. Through the third-order parameter ψ_x , we measure the breaking of translational symmetry in the smectic phase (details on its construction can be found in the Supplemental Material [45]). In addition, we record the actual area $\hat{A}$ for each configuration, from which we determine the packing fraction $\phi = N/(\hat{A})$. While we control the pressure p in simulations, in the following we will mostly report ϕ as the control parameter. To extract the phase behavior, we calculate the expectation values $\psi_\nu = \langle \hat{\psi}_\nu \rangle$ of the order parameters and their susceptibilities

$$\chi_\nu = \langle \hat{\psi}_\nu^2 \rangle - \langle \hat{\psi}_\nu \rangle^2, \quad (2)$$

where the brackets $\langle \cdot \rangle$ denote the equilibrium expectation value. To improve the statistical efficiency and to interpolate between pressures, we employ the *multistate Bennett acceptance ratio* method (MBAR) [46,47]. In the following, we identify transitions through peaks of the susceptibilities.

Figure 2(a) shows the susceptibility χ_2 as a function of packing fraction ϕ for hard rectangles with $\alpha = 5$ and $\alpha = 15$. We see that there is a clear peak for both aspect ratios indicating a transition toward orientational order [Fig. 1(b)]. However, the nature of the transition is different in both cases. For strongly elongated rectangles ($\alpha = 15$), the peak of χ_2 occurs at low packing fraction $\phi^* \simeq 0.36$ and ψ_2 [Fig. 2(b)] exhibits a smooth increase for both protocols $\mathcal{I}_+$ and $\mathcal{S}_-$. At these packing fractions, the translational order parameter ψ_x does not show any signal [45]. This continuous transition thus occurs from the isotropic phase to the nematic phase (IN). At a larger packing fraction also the translational order parameter rises (its susceptibility peaks at $\phi^* \simeq 0.73$ [45]) indicating the transition to the smectic phase [Fig. 1(d)]. Across this second transition, the nematic order parameter $\psi_2 \simeq 1$ remains a smooth function.

In contrast, for small aspect ratio $\alpha = 5$ the order parameter ψ_2 in Fig. 2(b) displays a sigmoidal profile as expected for a discontinuous transition. Further evidence comes from the strong hysteresis between compression ($\mathcal{S}_-$) and expansion ($\mathcal{I}_+$) route, where for the latter $\psi_2 \simeq 0$ shows no signs of ordering. Importantly, the jump of ψ_2 coincides with the jump of the translational order parameter ψ_x [45]. We thus conclude that this jump corresponds to a transition into the smectic phase. From the behavior of ψ_2 alone one might be tempted to identify the other phase as isotropic since ψ_2 exhibits a single jump. However, turning to χ_4 in Fig. 3(a) reveals another narrow peak at a smaller packing fraction $\phi^* \simeq 0.68$ before entering the smectic phase. Configurations

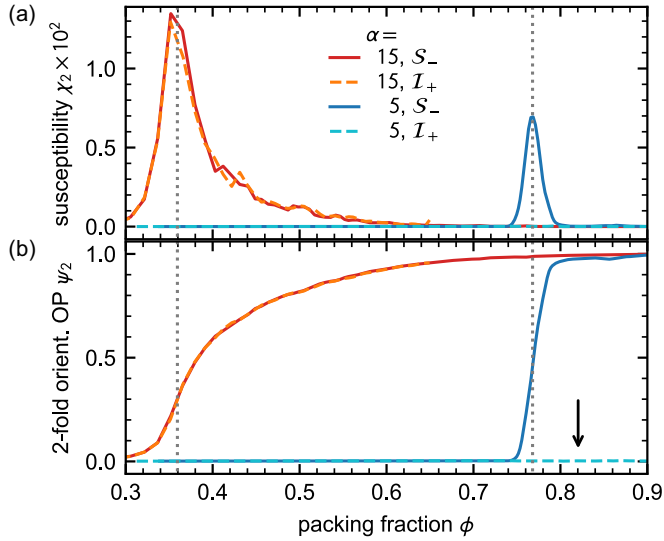


FIG. 2. Orientational order measured through Eq. (1) with $\nu = 2$ for high ($\alpha = 15$) and low ($\alpha = 5$) aspect ratios from expansion (S_- , solid lines) and compression (I_+ , dashed lines) protocols. (a) Susceptibility χ_2 . Peaks that we identify with transitions are indicated by gray dashed lines: continuous isotropic-nematic (IN) for $\alpha = 15$ at $\phi^* \simeq 0.36$ and discontinuous tetratic-smectic (TS) for $\alpha = 5$ at $\phi^* \simeq 0.77$; see discussion in main text. (b) Order parameter ψ_2 . Note the strong hysteresis for $\alpha = 5$, where no signal is found along the expansion route (arrow).

for $\phi > 0.68$ [Fig. 1(c)] exhibit short-range translational order together with orientational ordering along two perpendicular axes. Consequently, the distributions of orientations show two approximately equal populations separated by $\pi/2$. We thus associate this peak with the transition between the isotropic and a genuine *tetratic* phase. Across the tetratic-smectic (TS) transition, now ψ_4 remains smooth.

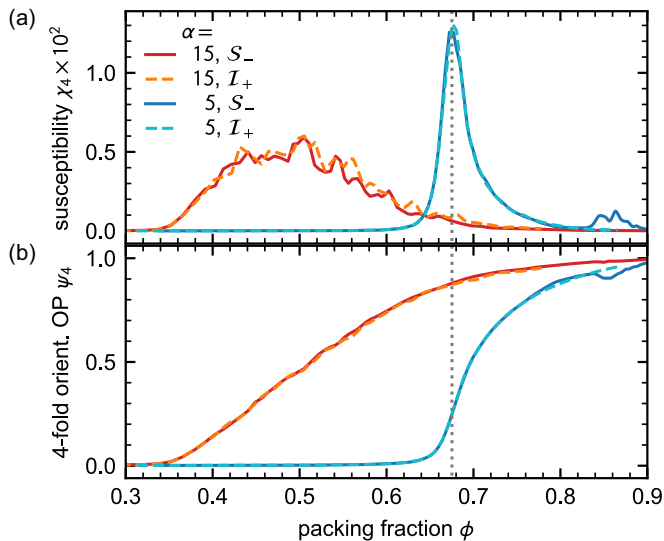


FIG. 3. Analogous to Fig. 2 but plotting (a) χ_4 and (b) ψ_4 . The peak of χ_4 (gray dashed line) corresponds to the isotropic-tetratic (IT) transition. The very broad peak of χ_4 for $\alpha = 15$ lies within the nematic phase and is not identified as a transition.

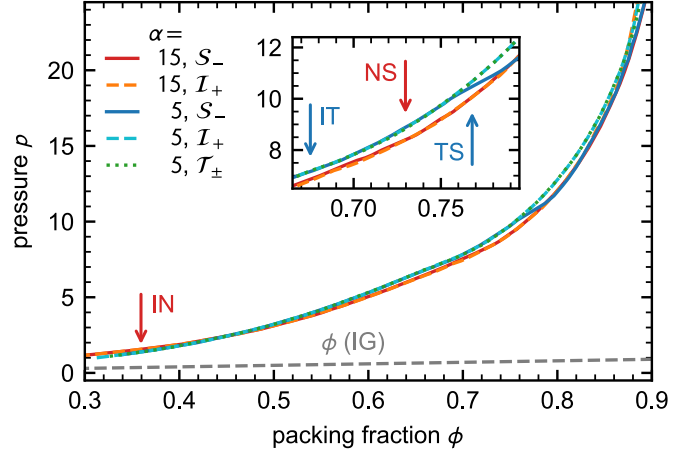


FIG. 4. Equation of state. Pressure $p(\phi)$ plotted as function of the average packing fraction ϕ . The gray dashed line shows the ideal gas pressure. The inset zooms into the range of packing fractions where phase transitions occur (arrows).

We note that ψ_4 also increases for $\alpha = 15$ [Fig. 3(b)], but with a rather gentle slope and a corresponding wide peak of the susceptibility χ_4 [Fig. 3(a)]. Crucially, ψ_4 alone is not sufficient to distinguish tetratic order since it becomes nonzero also for nematic order. Only the combination of nonvanishing ψ_4 together with $\psi_2 \simeq 0$ is indicative of tetratic order characterized by a bimodal angle distribution. We thus conclude that there is no tetratic phase anymore for $\alpha = 15$, and that the order for the transition to the smectic phase has changed from discontinuous for small aspect ratios α to continuous for large α . Before addressing intermediate aspect ratios, we turn to the nature of the transition into the smectic phase for small aspect ratios.

Figure 4 shows the equation of state $p(\phi)$ (note that the pressure is the control variable and ϕ is determined from the average area), again for $\alpha = 5$ and $\alpha = 15$ below and above the onset of the tetratic phase. As expected, no signature of the continuous transitions is seen in these curves. This is different for the TS transition for $\alpha = 5$, where we confirm the hysteresis in the equation of state: The expansion route S_- switches from the smectic branch to the tetratic branch, whereas the compression route I_+ continues along the (now metastable) tetratic branch, corroborating our identification of this transition as discontinuous. We note, however, that the crossing between smectic and tetratic branch is rather gradual, which we attribute to our simulations being performed in the constant-pressure ensemble. Moreover, a closer look reveals that in trajectories sampled in the neighborhood of the TS transition a relaxation time of $\tau_r \sim 10^7$ Monte Carlo steps is insufficient to reach the stationary regime and the twofold orientation order parameter ψ_2 (and also ψ_x) decreases further as τ_r is increased (see Supplemental Material [45] for a more detailed discussion on finite-time and finite-size effects).

To gain further insight into the nature of the tetratic-smectic (TS) transition at $\alpha = 5$, we employ MBAR to calculate the probability $P(A)$ of the fluctuating area, from which we determine the free energy $F(\phi) = -\ln P(\phi)$ of the packing fraction (see Supplemental Material [45]). Note that for these runs, the relaxation time of $\tau_r \sim 10^8$ Monte Carlo steps is

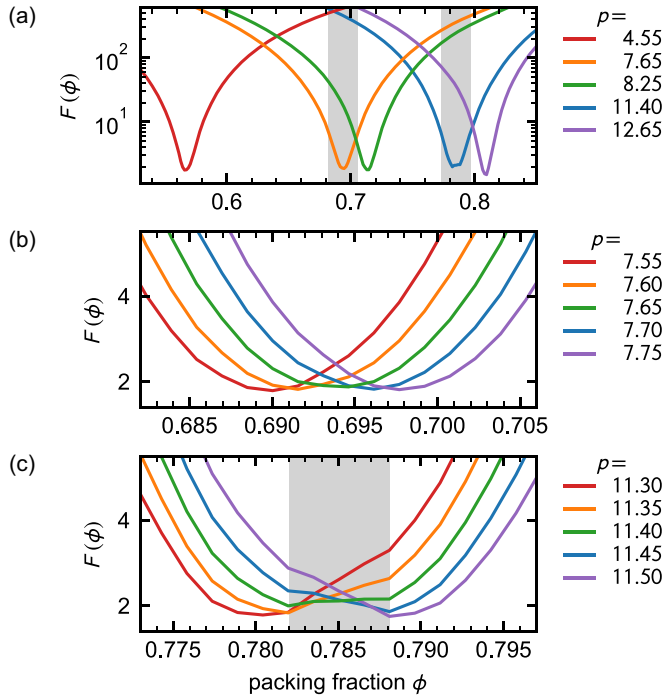


FIG. 5. Free-energy profiles $F(\phi)$ of the packing ϕ for $\alpha = 5$ and protocol S_- . (a) Selected pressures covering a wide range of ϕ . We zoom into the two shaded areas: (b) Across the IT transition ($p^* \simeq 7.65$, green), the position of the minimum of $F(\phi)$ evolves continuously from a high to a low value of ϕ . (c) Across the TS transition ($p^* \simeq 11.40$, green), we now observe a signature of a bimodality within a narrow pressure range corroborating a discontinuous transition. The shaded area indicates the coexistence region.

one order of magnitude longer. Figure 5(b) shows free-energy profiles as we increase the pressure p across the first transition from isotropic to tetratic. Distributions are clearly unimodal and their minimum smoothly varies with pressure as one would expect for a continuous phase transition. This changes for higher pressures shown in Fig. 5(c), where we see signs for a nonconvex distribution. Around the transition pressure $p^* \simeq 11.40$, the minimum of $F(\phi)$ jumps. These features further corroborate our identification of the TS transition as discontinuous. The coexistence region is very narrow, $\Delta\phi \simeq 0.006$, and the peak of the susceptibility remains a good indicator for the location of the phase transition [45].

While we have presented numerical evidence for the TS transition to be discontinuous, one might still wonder what stabilizes the tetratic over the smectic phase. The tetratic phase is clearly characterized by a bimodal distribution of orientations, but it also shares positional order with the smectic phase. Instead of perfect layers, we find a network of domains that are orientated perpendicular. Blocks of α particles can flip collectively and form a lattice. Of course, perfect periodic order in two dimensions is suppressed by long-wavelength fluctuations (Goldstone modes) [12,13]. Moreover, for layer dislocations in the smectic phase that cost a finite free energy there is a density c_D of these defects with a corresponding length $\xi_D \sim c_D^{-1/2}$ (certainly larger than the system sizes studied here) beyond which the smectic phase fractures into domains [48]. Treating the system as blocks (superparticles)

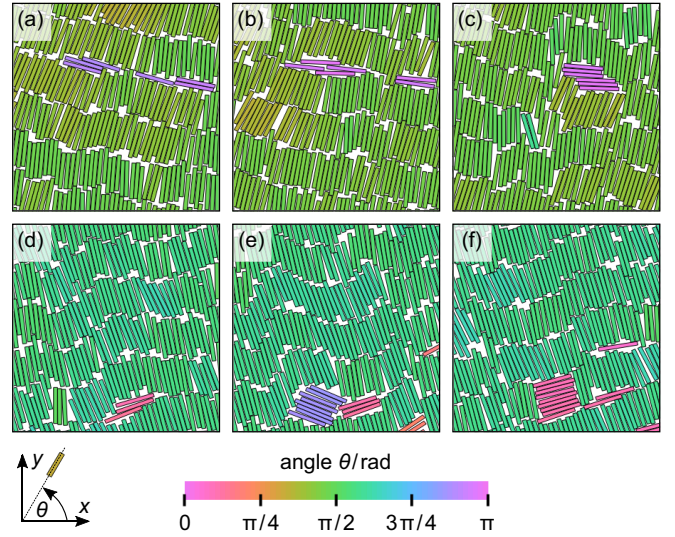


FIG. 6. Melting of the smectic phase occurs via two mechanisms: (a)–(c) Particles enter the gap between smectic layers and aggregate into a block oriented perpendicular or (d)–(f) collective flip of multiple particles. Each sequence of simulation snapshots shows early frames originating from a single trajectory of the protocol S_- at $\phi \simeq 0.74$ with $\tau_r \sim 10^8$.

of α rectangles, the degeneracy of the two perpendicular orientations raises the entropy of the tetratic phase by roughly $(1/\alpha)\ln 2$ per particle [34] (whereas the smectic phase is denser). This entropy gain diminishes as the aspect ratio increases. Because of this entropic gain one might wonder whether the tetratic phase should be the stable phase up to close packing (at least for sufficiently small aspect ratios). For squares the transition from tetratic to smectic has already been established [11]. Moreover, the equation of state [Fig. 4] shows that the smectic phase has a smaller pressure and thus is the thermodynamically stable phase. By ordering into the smectic phase, each particle has access to (a tiny bit) more area, and this increase of entropy outweighs the entropy gain from block flips.

Figure 6 gives some insight into how the smectic phase “melts” into the tetratic phase. Essentially, we observe two mechanisms: First, single particles can enter the gap between smectic layers [Figs. 6(a)–6(c)]. The sequence of simulation snapshots reveals that these particles propagate along the gaps and can assemble into a block that is orientated perpendicular to its local smectic neighborhood. Second, such a perpendicular block can also emerge more directly by means of a collective flip of several rectangles [Figs. 6(d)–6(f)], which occurs also in our hard-particle Monte Carlo simulation simulations utilizing only local rotation and translation moves. Additionally, the sequence of simulation snapshots resulting in Fig. 6(f) demonstrates that the block can grow by incorporating further gap travelers.

Putting everything together, Fig. 7 shows the resulting phase diagram for hard rectangles as a function of average packing fraction ϕ and aspect ratio α . Our numerical evidence suggests the following orders: IT, IN, and NS are classified as continuous, while TS and TN exhibit a discontinuous character. For large aspect ratios the nematic phase occupies a wide

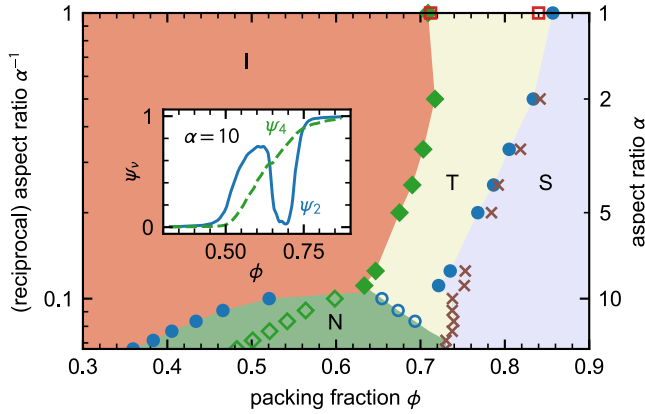


FIG. 7. The phase diagram of hard rectangles. Closed symbols indicate the location of peaks in the susceptibilities: χ_2 (●), χ_4 (◆), and χ_x (×). The open blue symbols correspond to the second peak of χ_2 while the open green symbols indicate the position of the wide peak of ψ_4 inside the nematic phase; cf. Fig. 3. Also shown are the phase boundaries for squares ($\alpha = 1$, □) taken from Ref. [11]. The inset shows $\psi_{2,4}$ for $\alpha = 10$ (using $\tau_r \sim 10^8$ Monte Carlo steps and the smectic protocol S_-).

range of packing fractions but narrows and finally terminates at $\alpha \simeq 9$. In the inset of Fig. 7, we show the orientational order ψ_2 at $\alpha = 10$. We see that ψ_2 rises as the system enters the nematic phase ($\psi_4 \simeq 0$) but then drops to zero before rising again but this time ψ_4 is already close to unity. We thus observe three peaks; the first is associated with the IN transition and the second with the NT transition. For smaller aspect ratios $\alpha < 10$, the second peak vanishes and the position of the first peak jumps to coincide with the emergence of translational order (the TS transition). For larger aspect ratios the second peak moves to larger packing fractions but the drop becomes less pronounced.

To conclude, we have numerically determined the full phase diagram for hard rectangles in two dimensions as a function of packing fraction ϕ and small to intermediate aspect ratios $1 \leq \alpha \leq 15$. We identify four phases: isotropic (no orientational and no translational order), nematic (uniaxial orientational order), smectic (uniaxial orientational and

translational order), and tetratic (biaxial orientational order). The nematic phase in hard rectangles terminates at a finite aspect ratio $\alpha \simeq 9$, below which it is replaced by the tetratic phase. The line of susceptibility maxima between nematic and tetratic order meets the phase boundary of the isotropic phase in a special “tricritical” point that deserves further investigation beyond the scope of this work. Another interesting question is the fate of the tetratic phase for large aspect ratios, and whether it also terminates or tapers into a very thin stripe between nematic and smectic phases.

The order parameters ψ_v we have used are proxies that probe the distribution of orientations and are convenient for efficient sampling in finite systems. Strictly speaking, for squares one has to turn to correlations of bond order to identify the “ n -tic” phase. The underlying degeneracy is lifted for rectangles and orientational order (as measured through ψ_v) is sufficient to identify the various phases. Still, as shown in Fig. 7, applying our local order parameters to squares allows to identify phase boundaries that agree very well with the results from Ref. [11]. The reason is that the TS transition coincides with the “unfreezing” of orientations [45].

Increasing the system size toward the thermodynamic limit, the unit director will slowly vary spatially (a consequence of “gapless” Goldstone modes), and global order parameters will fail for Kosterlitz-Thouless-type continuous transitions. The same holds in principle for the transition to the smectic phase, but now the condensation into layers frees a tiny portion of the area per particle and the transition becomes discontinuous. We thus find a novel scenario in hard rectangles for such two-step melting: the isotropic-tetratic transition is continuous (versus the discontinuous isotropic-hexatic transition for hard disks) while the tetratic-smectic transition is discontinuous (versus continuous hexatic-solid for hard disks).

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