Phase diagram of a bidispersed hard-rod lattice gas in two dimensions

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received 26 October 2015; accepted in final form 23 December 2015
published online 11 January 2016

PACS 64.60.De – Statistical mechanics of model systems (Ising model, Potts model, field-theory models, Monte Carlo techniques, etc.)
PACS 05.50.+q – Lattice theory and statistics (Ising, Potts, etc.)
PACS 64.70.mf – Theory and modeling of specific liquid crystal transitions, including computer simulation

Abstract – We obtain, using extensive Monte Carlo simulations, virial expansion and a high-density perturbation expansion about the fully packed monodispersed phase, the phase diagram of a system of bidispersed hard rods on a square lattice. We show numerically that when the length of the longer rods is 7, two continuous transitions may exist as the density of the longer rods is increased, keeping the density of shorter rods fixed: first from a low-density isotropic phase to a nematic phase, and second from the nematic to a high-density isotropic phase. The difference between the critical densities of the two transitions decreases to zero at a critical density of the shorter rods so that the fully packed phase is disordered for any composition. When both the rod lengths are larger than 6, we observe the existence of two transitions along the fully packed line as the composition is varied. Low-density virial expansion, truncated at the second virial coefficient, reproduces features of the first transition. By developing a high-density perturbation expansion, we show that when one of the rods is long enough, there will be at least two isotropic-nematic transitions along the fully packed line as the composition is varied.

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Introduction. – Entropy-driven transitions in systems of rod-like particles have long been an active area of theoretical and experimental research. Experimental realizations of such systems include tobacco mosaic virus \cite{1}, \textit{fd} virus \cite{2–4}, silica colloids \cite{5,6}, boehmite particles \cite{7,8}, DNA origami nanoneedles \cite{9}, liquid crystals \cite{10} and adsorbed gas molecules on metal surfaces \cite{11–15}. A system of hard spherocylinders in three-dimensional continuum undergoes a transition from an isotropic phase to an orientationally ordered nematic phase as density is increased. A further increase in density leads to a smectic phase with partial translational order and a solid phase \cite{10,16–20}. Lattice models of hard rods, of interest to this paper, also have a rich phase diagram in two dimensions, while not much is known in three dimensions.

Consider monodispersed hard rods on a two-dimensional lattice, where each rod occupies \(k\) consecutive lattice sites along any of the lattice directions and no two rods may overlap. When \(k = 2\) (dimers), the system is known to be disordered at all densities \cite{21–24}. When \(k \geq 7\), there are, interestingly, two transitions: first, from a low-density disordered to an intermediate-density nematic phase and second, from the nematic to a high-density isotropic phase \cite{25,26}. While the first transition belongs to the Ising (three-state Potts) universality class for the square (triangular) lattice \cite{27}, the universality class of the second transition remains unclear with the numerically obtained critical exponents differing from those of the first transition, though a crossover to the Ising exponents at larger length scales could not be ruled out \cite{26,28}. Exact analysis, restricted to a rigorous proof for the existence of the first transition when \(k \gg 1\) \cite{29} and the exact solution on a Bethe-like lattice \cite{30}, does not shed any light on the second transition. The fully packed limit of monodispersed rods is disordered, may be mapped onto a height model with a \((k - 1)\)-dimensional height field, and has power law orientational correlations \cite{21,31,32}. 
Polydispersity in length of the particles is hardly avoidable in experiments and results in features such as fractionation, two distinct nematic phases and nematic-nematic or isotropic-nematic-nematic phase coexistence [7,8]. Some of these features may be obtained using the density functional theory, virial expansion or Monte Carlo simulations in the continuum [33–42]. The lattice counterpart is less studied and the phase diagram is mostly unexplored. Although, different theories like the fundamental measure theory [43,44] and the scaled particle theory [45] have been developed for studying mixtures of hard particles, a detailed numerical simulation for the hard-rod system with restricted orientations is still lacking. The existence of demixing transitions are predicted within these theories. A particular model of polydispersed rods with a rod of length $k$ having a weight $z_i^2 z_i^{-k-2}$, where $z_i$ is the fugacity of an internal (endpoint) monomer was shown to undergo an isotropic-nematic transition using transfer matrix methods [46]. When $z_i = \sqrt{z_i}/2$, the model may be solved exactly by mapping it to the two-dimensional Ising model [47]. A second transition to the high-density disordered phase is absent [46]. However, in this model, densities of different species cannot be changed independently.

What is the phase diagram for lattice models of polydispersed rods? Does polydispersity preserve the second phase transition into a high-density disordered phase? Is the fully packed line still disordered or could there be regions with nematic order? In this letter, we address these questions by determining the phase diagram of bidispersed 2-7, 6-7, and 7-8 mixtures using extensive Monte Carlo simulations and studying generic bidispersed mixtures using low-density virial expansions and high-density perturbation expansions close to full packing. In particular, we show that the second transition at high densities persists, and if one of the rod lengths is large enough, the system at full packing will exhibit at least two isotropic-nematic transitions as the ratio of densities of the two species is varied.

**Model and the Monte Carlo algorithm.** – Consider a bidispersed system of rods of length $k_1$ and $k_2$ on a square lattice of size $V = L \times L$ with periodic boundary conditions, where each rod is either horizontal or vertical. A horizontal (vertical) rod of length $k_i$ (where $i = 1, 2$) occupies $k_i$ consecutive lattice sites along the $x$ ($y$) axis. Each site may be occupied by at most one rod. A fugacity $z_i$, $i = 1, 2$, where $\mu_i$ is the corresponding reduced chemical potential.

We simulate this model using a constant fugacity grand canonical Monte Carlo algorithm involving cluster moves. This algorithm is an adaptation of the scheme that was quite efficient in equilibrating systems of monodispersed long rods [26,48]. Variants of this algorithm have been used to study systems of hard rectangles [49–51], disks on square lattice [52] and mixtures of squares and dimers [53]. We briefly discuss the algorithm here.

Choose at random a row or column of the lattice. If a row is chosen, all the horizontal rods on that row are removed, while the rest of the configuration is kept unchanged. The row now consists of intervals of empty sites separated by the sites occupied by vertical rods. These empty intervals are re-occupied with a new configuration of horizontal rods consistent with equilibrium probabilities. If, instead of a row, a column is chosen, a similar evaporation-deposition operation is done with vertical rods. The calculation of these equilibrium probabilities reduces to a one-dimensional problem.

Let $\Omega_i(z_{k_1}, z_{k_2}; \ell)$ be the grand canonical partition function of a one-dimensional chain of length $\ell$ with open boundary conditions. The probability that the first site of the one-dimensional chain is occupied by the left-most site of a rod of length $k_i$ is $p_i^\ell = z_i \Omega_i(z_{k_1}, z_{k_2}; \ell - k_i)/\Omega_i(z_{k_1}, z_{k_2}; \ell)$, where $i = 1, 2$. The partition functions $\Omega_i(z_{k_1}, z_{k_2}; \ell)$ obeys the recursion relation $\Omega_i(z_{k_1}, z_{k_2}; \ell) = \sum_{i=1}^{\ell} z_i \Omega_i(z_{k_1}, z_{k_2}; \ell - k_i) + \Omega_i(z_{k_1}, z_{k_2}; \ell - 1)$ for $\ell \geq \min(k_1, k_2)$, with the boundary conditions $\Omega_i(z_{k_1}, z_{k_2}; \ell) = 1$ for $\ell = 0, 1, \ldots, \min(k_1, k_2) - 1$ and $\Omega_i(z_{k_1}, z_{k_2}; \ell) = 0$ for $\ell < 0$. The partition function of a one-dimensional chain of length $\ell$ with periodic boundary condition, $\Omega_p(z_{k_1}, z_{k_2}; \ell)$, is easy to determine once $\Omega_i(z_{k_1}, z_{k_2}; \ell)$ is known. It obeys the recursion relation $\Omega_p(z_{k_1}, z_{k_2}; \ell) = \sum_{i=1}^{\ell} z_i k_i \Omega_i(z_{k_1}, z_{k_2}; \ell - k_i) + \Omega_i(z_{k_1}, z_{k_2}; \ell - 1)$. The recursion relations may be solved exactly for $\Omega(z_{k_1}, z_{k_2}; \ell)$ and $\Omega(z_{k_1}, z_{k_2}; \ell)$. The list of relevant probabilities $p_i^\ell$ for all $\ell \leq L$ are stored in order to reduce computational time.

In addition to the evaporation-deposition moves, we also implement a flip move [49]. We choose a site at random. Only if it is the bottom-left corner of a block of size $(k_1 \times k_2)$ containing $k_1$ aligned parallel horizontal (vertical) rods, it is replaced by a similar block of $k_1$ aligned parallel vertical (horizontal) rods. One Monte Carlo (MC) move contains $2L$ evaporation-deposition moves and $L^2$ flip moves. All the numerical results presented in this paper are obtained using a parallelized version of the algorithm.

The largest system size that we simulate is $L = 560$. A single data point in the phase diagrams (total of 47 data points), has been obtained using (on an average) 30 runs of Monte Carlo simulation. In a typical run, we equilibrate the system for $5 \times 10^6$ Monte Carlo steps following which the relevant quantities are averaged over $3 \times 10^6$ Monte Carlo steps. We ensure equilibrium by confirming that the same equilibrium state is reached from two different initial conditions: one corresponding to a nematic phase and the other to a disordered phase.

**Results.** – We study three different mixtures: 2-7, 6-7, and 7-8. These choices were made for the following reasons. A monodispersed system of hard rods shows phase transitions only when the rod length $k \geq 7$. Rods of length 2 and 6 being the smallest and largest lengths that do not show a nematic phase, studying 2-7 and 6-7 allows us to obtain the trend for intermediate lengths. To study the
Fig. 1: (Color online) Snapshots of the 7-8 mixture at (a) low-density isotropic phase ($\rho_1 \approx 0.187$, $\rho_8 \approx 0.311$, $\mu_7 = -2.0$, $\mu_8 = -1.4$), (b) intermediate-density nematic phase ($\rho_7 \approx 0.372$, $\rho_8 \approx 0.441$, $\mu_7 = 1.5$, $\mu_8 = 2.0$) and (c) high-density isotropic phase ($\rho_7 \approx 0.055$, $\rho_8 \approx 0.922$, $\mu_7 = 7.2$, $\mu_8 = 11.0$).

The horizontal and vertical rods of length 7 are colored red and black, and the same of length 8 are colored blue and green respectively.

Fig. 2: (Color online) Phase diagram in the $\mu$-plane for mixtures of (a) 2 and 7, (c) 6 and 7, and (e) 7 and 8, and in the $\rho$-plane for (b) 2 and 7, (d) 6 and 7, and (f) 7 and 8. The data points (solid circles) are obtained from Monte Carlo simulations. The shaded regions are guides to the eye and correspond to regions with nonzero nematic order.

Bidispersed hard-rod gas

Phases. For a particular bidispersing mixture, we obtain the complete phase diagram by simulating the system at different values of $\mu_k$ and $\rho_k$. The chemical potentials and densities are determined from the crossing of the curves of the Binder cumulant as a function of the chemical potential or density for different system sizes. The phase diagrams for 2-7, 6-7, and 7-8 mixtures are shown in fig. 2. The shaded regions in the phase diagrams correspond to ordered N phases, while the empty regions correspond to I phases with no orientational order.

A system of monodispersing dimers ($k = 2$) does not show any phase transition. Thus, when $\rho_2 \gg \rho_7$ (for the 2-7 mixture), we do not expect any phase transition. When $\rho_2$ or $\rho_7$ is small enough, we observe two transitions as $\mu_7$ or $\rho_7$ is increased: first from a low-density I phase to an intermediate-density N phase and second from the N phase to a high-density I phase (see fig. 2(a) and (b)), as seen for the system of monodispersing rods of length $\geq 7$. The difference between the two critical densities decreases as $\rho_2$ is increased and beyond a critical $\rho_2$, no transitions are observed. On the other hand, when $\rho_7$ is kept fixed and $\rho_2$ is increased, at most one transition is present. One may go from the low-density I phase to the high-density I phase continuously without crossing any phase boundary, suggesting that the high-density I phase is a re-entrant low-density I phase [28].

The phase diagram for the 6-7 mixture (see fig. 2(c) and (d)) is quite similar to that of the 2-7 mixture. The area of the nematic region is larger for the 6-7 mixture, showing that longer rods favor orientational ordering. Unlike the 2-7 mixture, now there are regions where the system undergoes two transitions when $\rho_7$ is kept fixed and $\rho_8$ is varied. The fully packed line remains disordered for all compositions of 2-7 and 6-7 mixtures. We expect a qualitatively similar phase diagram for mixtures with $k_1 < 7$ and $k_2 = 7$.

Now, consider the 7-8 mixture. This case is different from the above two as there are two critical points on each of the two axes $\rho_7 = 0$ and $\rho_8 = 0$ (see fig. 2(e) and (f)). For small values of $\rho_7$ or $\rho_8$, two transitions are observed. The high-density I phase is separated from the low-density I phase by a region of N phase. It raises the question as to whether the system is disordered at full packing as seen for 2-7 and 6-7 mixtures. The algorithm that we use does not equilibrate the system at full packing. Instead, by simulating the system close to full packing, we find that the phase boundaries, separating the N phase from the high-density I phase approach the $\rho = 1$ line, and appear to terminate at two separate points (see fig. 2(f)), suggesting that there are two transitions along the $\rho = 1$ line.

All the transitions that we observe are continuous as we do not observe any jump in the density or in the nematic order parameter near the transitions. Although, we do
not observe any demixing or fractionation, we do not rule out the possibility of its existence when the aspect ratios of the two species are very different.

**Virial expansion.** We now determine the phase diagram of the system from a standard low-density virial expansion for multiple species truncated at the second virial coefficient [19].

Let \( N_i \), where \( i = 1, 2 \) and \( j = h \) (horizontal), \( v \) (vertical), denote the number of rods of length \( k_i \) with orientation \( j \). The partition function of a system of \( N \) rods in a volume \( V \) is then given by

\[
Q_N = \frac{V^N}{N!VN} \sum_{\{N_i\}} \frac{N!}{\prod_i N_i!} \exp[-\phi(\{N_i\})],
\]

where the prime denotes the constraint \( \sum_{i,j} N_{ij} = N \), and \( \phi \) is the reduced excess free energy for a given distribution of lengths and orientations:

\[
\exp[-\phi_N(\{N_i\})] = \frac{1}{V^N} \sum_{R} \exp(-\beta U_N),
\]

where \( U_N \) is the total interaction energy, and \( R \) denotes all possible positions. Let \( x_i^j = N_i^j / N \) denote the fraction of rods of length \( k_i \) with orientation \( j \). For large \( N \), \( V \), (1) may be written as

\[
Q_N = \int_0^1 \prod_i dx_i e^{-NF(x_i^j)\delta(\sum_{i,j} x_i^j - 1)},
\]

where \( F \) is the free energy per particle:

\[
F(\{x_i^j\}) = \ln \frac{4\theta}{e} + \sum_{i,j} x_i^j \ln x_i^j + \frac{\theta}{2} \sum_{i,j} (x_i^j)^2(2k_i - 1) + \theta \sum_i k_i^2 \prod_j x_i^j + \theta(k_1 + k_2 - 1) \sum_i \prod_j x_i^j + \theta(x_1^1 x_2^1 + x_2^2 x_1^2)k_1k_2 + O(\theta^2).
\]

For given densities of the two species, the free energy \( F \) in (7) may be expressed in terms of the nematic order parameters of the two species denoted by \( \psi_{12} = (x_2^1 - x_1^1)/(x_1^2 + x_2^2) \) and \( \psi_{23} = (x_2^2 - x_3^2)/(x_2^2 + x_3^2) \). The phase for a given number density is obtained by minimizing \( F \) with respect to \( \psi_{12} \) and \( \psi_{23} \). We find the existence of only the low-density isotropic-nematic (I-N) transition, similar to the solution of the monodisperse system on a Bethe-like lattice [30]. The I-N phase boundary may be found by solving the equation \( \partial^2 F / \partial \psi_{12}^2 = \partial^2 F / \partial \psi_{13}^2 = 0 \), and we obtain

\[
\frac{(k_1 - 1)^2}{k_1} \rho_{k_1}^2 + \frac{(k_2 - 1)^2}{k_2} \rho_{k_2}^2 = 2.
\]

By setting \( \rho_{k_2}^2 = 0 \), we obtain the critical density \( \rho_{k_1}^c = 2k_1/(k_1 - 1)^2 \), as found earlier for the monodisperse system [51]. For the monodisperse system, the I-N transition exists for lengths larger than 3.

The phase diagrams for two different mixtures (2-6 and 4-6), obtained from the virial expansion are shown in fig. 3. The shaded (empty) regions correspond to N (I) phases. While the theory predicts the existence of an I-N transition at full packing \( (\rho = 1) \) for 2-4 mixture as the ratio of the densities of the two species is varied (see fig. 3(a)), for the 4-6 mixture, the fully packed line is always nematic (see fig. 3(b)).

The phase diagram obtained from the virial expansion differ from those obtained by simulations. It was shown in ref. [51] that in two dimensions, the higher-order even virial coefficients contribute and cannot be neglected even in the limit \( k_i \to \infty \). Thus, truncating the expansion of the reduced excess free energy at the second virial coefficient is only a reasonable approximation at very low densities. The inclusion of the third virial coefficient is quite straightforward. While for finite \( k_i \), it introduces small corrections to the critical densities, it does not contribute in the limit of a large aspect ratio. The question as to whether a phase separation
or demixing transition exist for an arbitrary mixture is not clear and may be addressed using a bifurcation analysis after the careful consideration of the higher-order coefficients.

Expansion about the pure state along the fully packed line. The fully packed line can neither be numerically studied with the algorithm used in this paper nor with the low-density virial expansion. Instead, we calculate the entropies of the I and N phases as a perturbation expansion about the fully packed monodisperse system. For simplicity, let $k_1$ and $k_2$ be mutually prime. We approximate the N phase as one where all the rods point in one direction. The arrangement of these rods is a simple combinatorial problem and the entropy per unit site $s_{\text{nem}}$, in terms of the number densities of the two species $\theta_{k_1}$ and $\theta_{k_2}$, is

$$s_{\text{nem}} \approx (\theta_{k_1} + \theta_{k_2}) \ln(\theta_{k_1} + \theta_{k_2}) - \theta_{k_1} \ln \theta_{k_1} - \theta_{k_2} \ln \theta_{k_2},$$

where $\sum_i k_i \theta_{k_i} = 1$. Expanding (9) for small $\theta_{k_2}$ we find

$$s_{\text{nem}} \approx -\theta_{k_2} \ln \theta_{k_2} + O(\theta_{k_2}), \quad \theta_{k_2} \to 0.$$  (10)

To estimate the entropy of the I phase, we break the lattice into $L/k_1$ horizontal strips of width $k_1$. The partition function $\mathcal{L}_0$, when only rods of length $k_1$ are present, is then

$$\mathcal{L}_0 = 2k_1 \omega_p(L)^{L/k_1},$$  (11)

where $\omega_p(L)$ ($\omega_o(L)$) is the partition function for a strip of length $L$ with periodic (open) boundary conditions, and the factor $2k_1$ accounts for the two orientations and translational invariance. Clearly, $\omega_o(L) = \omega_p(L-1) + \omega_o(L-k_1)$ with solution $\omega_o(L) = a_0 \lambda^{L}$, where $\lambda^{k_1} - \chi^{k_1-1} - 1 = 0$. Likewise, $\omega_p(L) = a_0 \chi^L$.

Consider defects consisting of rods of length $k_2$. To make the system fully packed, a minimum of $k_2$ such rods are required. If $k_1$ and $k_2$ were not mutually prime, this number would change. The smallest contribution to the partition function is when these $k_1$ rods are either horizontal and directly on top of each other, forming a block of size $k_1 \times k_2$ or vertical and one on top of the other, forming a line of length $k_1k_2$. Denoting the contribution from these defects as $\mathcal{L}_1$, we obtain

$$\frac{\mathcal{L}_1}{\mathcal{L}_0} = \frac{L^2 \omega_{k_1}}{k_1} \left[ \frac{\omega_o(L-k_2)}{\omega_p(L)} + \frac{\omega_o(L-1k_2)}{\omega_o(L-k_2)} \right].$$  (12)

Substituting for $\omega_p$ and $\omega_o$ in terms of $\lambda$, we obtain

$$\frac{1}{L^2} \ln \frac{\mathcal{L}}{\mathcal{L}_0} = \frac{\omega_{k_1}}{k_1} \ln \left[ \frac{\theta_{k_2} \lambda^{k_2}}{a_0/a_p + (a_0/a_p)^{k_2}} \right] + O(\omega_{k_1}).$$  (13)

Performing a Legendre transform of the free energy $-\ln \mathcal{L}$ to obtain entropy in terms of density, we find

$$s_{\text{iso}} \approx \frac{\ln \lambda + \theta_{k_2}}{k_1} \ln \left[ \frac{\theta_{k_2} \lambda^{k_2}}{a_0/a_p + (a_0/a_p)^{k_2}} \right] + \ldots .$$  (14)

For large $k_1$, the calculation based on strips gives a good estimation of the entropy. In this limit, $\ln \lambda \approx k_1^{-1} \ln k_1$ [25]. Equating the entropies for $N$ and $I$ phases (see (10) and (14)), we obtain that along the fully packed line, the system undergoes an isotropic-nematic transition at $\theta_{k_2} \sim k_1^{-2}$. Given that the fully packed phase of a monodispersed system is isotropic, we expect that there are at least two I-N transitions along the fully packed line, when $k_1$ is very large.

Discussions. – In this letter, we determined the phase diagram of a system of bidispersed hard rods on a square lattice using Monte Carlo simulations, virial expansion and high-density perturbation expansion. Numerically, the phase diagrams of three different mixtures (2-7, 6-7 and 8) were determined. For any $2 \leq k_1 \leq 6$ and $k_2 = 7$, the system at full packing is always disordered and the phase diagram is expected to be qualitatively similar to that of the 6-7 or 2-7 mixture. When $k_1, k_2 \geq 7$, we expect the phase behavior to be qualitatively similar to that of the 7-8 mixture, and predict the existence of two transitions at full packing. The low-density virial expansion is able to reproduce the low-density $I$-$N$ transition but does not work well at high densities. When one of the rod lengths is high enough, the high-density perturbation expansion predicts the existence of two I-$N$ phase transitions along the fully packed line. This prediction could not be verified numerically as the algorithm used in the paper is unsuitable for studying the fully packed line. Although, we do not find any demixing transitions for the mixtures that are studied, we do not rule out its existence for other mixtures. The results from the virial expansion may be reproduced by solving exactly the model on tree-like lattices. However, the high-density disordered phase is absent for hard rods, similar to that seen for the monodispersed case [30]. With soft repulsive interactions [28], it is possible to reproduce the features seen in the Monte Carlo simulations, and will be addressed in a future publication.

Monodispersed hard rectangles have a richer phase diagram than rods, with up to three density-driven transitions: from isotropic to nematic to columnar to a solid-like phase [49–51, 54]. A simple mixture of dimers and squares in a 3-10 lattice using Monte Carlo simulations, virial expansion and high-density perturbation expansion. Numerically, the phase diagrams of three different mixtures (2-7, 6-7 and 8) were determined. For any $2 \leq k_1 \leq 6$ and $k_2 = 7$, the system at full packing is always disordered and the phase diagram is expected to be qualitatively similar to that of the 6-7 or 2-7 mixture. When $k_1, k_2 \geq 7$, we expect the phase behavior to be qualitatively similar to that of the 7-8 mixture, and predict the existence of two transitions at full packing. The low-density virial expansion is able to reproduce the low-density $I$-$N$ transition but does not work well at high densities. When one of the rod lengths is high enough, the high-density perturbation expansion predicts the existence of two I-$N$ phase transitions along the fully packed line. This prediction could not be verified numerically as the algorithm used in the paper is unsuitable for studying the fully packed line. Although, we do not find any demixing transitions for the mixtures that are studied, we do not rule out its existence for other mixtures. The results from the virial expansion may be reproduced by solving exactly the model on tree-like lattices. However, the high-density disordered phase is absent for hard rods, similar to that seen for the monodispersed case [30]. With soft repulsive interactions [28], it is possible to reproduce the features seen in the Monte Carlo simulations, and will be addressed in a future publication.
shows a line of critical points with continuously varying exponents [53]. Polydisperse d rectangles should thus have a complicated and richer phase diagram and represent a promising area for future study.

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The simulations were done on the supercomputer Annapurna at the Institute of Mathematical Sciences.

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