Impact of packing fraction on diffusion-driven pattern formation in a two-dimensional system of rod-like particles

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Abstract. Pattern formation occurring in a two-dimensional system of rod-like particles has been simulated using a lattice approach. The rod-like particles were modelled as linear $k$-mers of two mutually perpendicular orientations ($k_x$- and $k_y$-mers) on a square lattice with periodic boundary conditions (torus). Two different models of random sequential adsorption were used to produce an initial homogeneous and isotropic distribution of $k$-mers with different packing fraction values. By means of the Monte Carlo technique, translational diffusion of the $k$-mers was simulated as a random walk, while rotational diffusion was ignored, so, the $k_x$- and $k_y$-mers were considered as individual species. The system tended towards a well-organised nonequilibrium steady state in the form of diagonal stripes for relatively long $k$-mers ($k \geq 6$) and moderate packing fractions ($p_{\text{down}} < p < p_{\text{up}}$, where both the critical packing fractions $p_{\text{down}}$ and $p_{\text{up}}$ were dependent on $k$).

1. Introduction

A variety of complex nonequilibrium phase behaviours, orientational ordering, and self-organisation properties have been experimentally observed in vibrated systems composed of shape-anisotropic (particularly, rod-like) particles in two dimensions, see [1, 2, 3, 4, 5, 6, 7, 8]. These experiments demonstrated that, at a high packing fraction, the shape of the particles is important in their orientational ordering. Numerous examples of patterns and of phase behaviour in granular media together with appropriate references can be found in review [9].

In recent decades, much attention has been paid to the study of systems of linear $k$-mers (particles occupying $k$ adjacent adsorption sites) deposited on 2D lattices. A linear $k$-mer represents the simplest model of a hard-core (completely rigid) rod-like particle with an aspect ratio of $k$. Computer simulations have been extensively applied to investigate percolation and jamming phenomena for the random sequential adsorption (RSA) [10] of $k$-mers (see, e.g., [11, 12, 13] and the references therein).

Recently, diffusion-driven pattern formation in a 2D system of $k$-mers has been studied by means of Monte Carlo (MC) simulations [14, 15]. The $k$-mers were orientated in two mutually

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perpendicular directions. For those systems with equal numbers of species of two different kinds, i.e., $k_x$-mers and $k_y$-mers,

(i) nonequilibrium steady patterns in the form of stripe domains have been observed only when periodic boundary conditions (PBCs) apply in both directions;

(ii) this self-organisation is possible only in fairly dense systems;

(iii) these stripe domains have been observed only for relatively long $k$-mers, $k \geq 6$;

(iv) the relaxation time to reach a well-organised steady-state is drastically dependent on lattice size.

Furthermore, self-organisation can occur in systems with unequal numbers of $k_x$-mers and $k_y$-mers.

However, the effect of the packing fraction on such pattern formation does not appear to have been studied in detail. Particularly, jammed states have been produced using the RSA, as reported in [14, 15]. Note, that, while other mechanisms, e.g., RSA with diffusional relaxation can produce denser systems [16], it is not clear whether self-organisation is possible in such the dense systems.

The present conference paper is devoted to a detailed analysis of how the packing fraction affects pattern formation in a two-species diffusion system. For this, we have examined the effect of the packing fraction on such pattern formation using the above two kinds of RSA to produce initial homogeneous and isotropic states.

The rest of the paper is organised as follows. In section 2, the technical details of the simulations are described and all necessary quantities are introduced. Section 3 presents our principal findings. Section 4 summarises the main results.

2. Details of simulation

In our study, a lattice approach was used, the problem being simulated using a square lattice of size $L \times L$. All calculations were performed using only one lattice size $L = 256$. This size was chosen as the relaxation time increases drastically with increasing lattice size [14, 15]. We used toroidal boundary conditions, i.e., periodic boundary conditions (PBCs) along both the $x$ and $y$ axes.

The rod-like particles were presented as linear $k$-mers. Previous data had evidenced that self-organisation is not observed for short $k$-mers ($k < 6$) [14]; accordingly in this work, the length of the $k$-mers was varied from 6 to 12. Longer particles were excluded from consideration due to an expected essential finite-size effect (the ratio $L/k$ becomes too small for $k > 12$). Isotropic deposition of the $k$-mers was ensured, i.e., $k$-mers oriented along the $x$ and $y$ directions ($k_x$-mers and $k_y$-mers, respectively) were equiprobable in their deposition. This corresponded to the zero value of a mean order parameter of the system, defined as

$$s = \frac{|N_y - N_x|}{N_y + N_x}, \quad (1)$$

where $N_x$ and $N_y$ are the numbers of sites occupied by the $k_x$-mers and $k_y$-mers, respectively.

The diffusion of $k$-mers was simulated as a random walk. Only translational diffusion was taken into consideration. This is essentially the case for fairly dense systems in the jamming state, where rotational diffusion is impeded, especially for large values of $k$. Undoubtedly, for dilute systems, rotational diffusion can occur, however, this was ignored in our study.

There are two possibilities of random walk [17]. In the first case, the particles are “blind”, i.e. a particle chooses one of the four possible directions and tries to move in this direction. If the attempt is unsuccessful, the particle does not try to find a different direction available for its movement. Such behaviour obeys detailed balance condition [18]. In the second case, a
particle is “myopic”, i.e., it consistently tries all four possible directions in a random order until its first successful attempt to move or until all possibilities have been tried. Note, that this type of kinetics can violate the detailed balance condition and drives the system to a nonequilibrium steady state \[18\]. Nevertheless, this approach is natural for “intellectual” particles, e.g., biological species that are looking for life resources, and for pedestrians. Thus, such particles can also be called “intellectual” because they choose one of the possible directions to move, if any exists, with equal probability.

An arbitrary \(k\)-mer was randomly chosen at each step and a translational shift by one lattice unit along either the longitudinal or the transverse axis of the \(k\)-mer was attempted. All four possible directions to shift the \(k\)-mer were attempted in a random order until a direction was found in which displacement of the particle is possible, or until all possible directions had been exhausted \[17\].

The time required for an attempted displacement of the total number of \(k\)-mers in the system, \(N\), was taken as the MC time unit. Time counting was started from the value of \(t_{\text{MC}} = 0\), being the initial moment (before diffusion), and the total duration of the simulation was \(10^7\) MC time units. There are two possibilities of random choice of the next particle \[19\]. The first possibility is to perform a random permutation of all \(N\) particles and then consistently to go through these randomly reordered particles. In this case, each particle is chosen once, and only once, at each MC step. The second possibility is a selection with return. In this case, at the current MC step, some particles may be chosen several times while others may end up not being chosen at all. Both algorithms lead to the same final states but the intermediate dynamics of the systems may differ \[19\]. The latter algorithm was used in \[14, 15\] and in the present research.

The RSA model \[10\] was used to produce an initial homogeneous distribution of linear \(k\)-mers. These \(k\)-mers were deposited randomly and sequentially, and their overlapping with previously placed particles was forbidden, i.e. excluded volume interaction was assumed. The packing fraction, \(p\), was varied in the range \(p \in [0.1, p_j]\), where \(p_j\) was the packing fraction at jamming. In the jammed state, no additional \(k\)-mer can be placed because the presented voids are too small or of inappropriate shape. To avoid confusion, here and below, we will use \(p_j\) for the packing fraction of the jammed state produced solely by standard RSA.

We additionally studied the effect of the packing fraction on self-organisation when using the extended RSA approach for obtaining packing fractions \(p > p_j\). To produce such a dense state, we used a two-step algorithm. 

At the first step, RSA was used to produce a jammed state. 

At the second step, the particles were allowed to diffuse. Due to diffusion, some additional voids can occur. When a void is large enough, one additional particle was obligatorily deposited onto the lattice. The action resembles the purpose of shaking a full sugar bowl when we want to add more sugar to it. This additional deposition was stopped when the packing fraction reached the necessary value.

Figure 1 demonstrates how the packing fraction increases with time for one particular case. In the case of unlimited additional deposition, fairly dense packing fraction \(p \approx 1.25p_j\) could be reached for \(6 \leq k \leq 12\). It was found that additional particle deposition leads to suppression of self-organisation. Preliminary studies had shown that the initial structure is actually frozen, but becomes more and more dense during the additional deposition of particles.

After reaching the desired packing fraction, only diffusion of the particles was allowed. We will use the abbreviation RSADA for this process of extended RSA with diffusion and additional adsorption.

To characterise the temporal evolution of the system under consideration, several quantities were monitored at each given MC step.
(i) The normalised number of clusters, \( n \), i.e., the current value divided by the value at the initial state, \( t_{MC} = 0 \). In percolation theory, a cluster is a group of neighbouring occupied sites [20]. In our particular case, a cluster is a group of \( k \)-mers of the same kind, i.e., \( k_x \)-mers or \( k_y \)-mers, that are connected with one another. Two \( k \)-mers are considered to be connected when there is at least one pair of neighbouring sites that belong to different \( k \)-mers. The clusters built of \( k_x \)-mers and clusters built of \( k_y \)-mers were accounted separately and then averaged. We used the Hoshen–Kopelman algorithm [21] to count the clusters.

(ii) The local anisotropy, \( s \), i.e., the order parameter was calculated using (1) in a window of \( l \times l \) sites and averaged over the entire set of windows. We used \( l = 2^{-n}L \), \( n = 1, 2, \ldots, 5 \). The principal analysis was performed for \( l = L/4 \). For this window size, we calculated ratios of each final value of \( s \) at \( t_{MC} = 10^7 \) to its initial value at \( t_{MC} = 0 \).

(iii) The fraction of interspecific contacts \( n_{xy}^* = n_{xy}/(n_{xy} + n_x + n_y) \), where \( n_{xy} \) is the number of interspecific contacts between the different sorts of \( k \)-mers (i.e., \( k_x-k_y \)), \( n_x \) and \( n_y \) are the numbers of intraspecific contacts between \( k \)-mers of the same kind (i.e., \( k_x-k_x \) and \( k_y-k_y \), respectively). For convenience of comparison, we additionally used the ratios of the final value of \( n_{xy}^* \) at \( t_{MC} = 10^7 \) to its initial value at \( t_{MC} = 0 \).

(iv) The shift ratio, \( R \), i.e., the ratio of the number of shifts of the \( k \)-mers along the transverse axes to the number of shifts along their longitudinal axes during one MC step.

(v) The electrical conductivities, \( \sigma \). We used the method described in [14] to transform the system under consideration into a random resistor network (RRN). The Frank–Lobb algorithm [22] was applied to calculate the electrical conductivity of such RRNs, (see figure A1 for details of the calculations).

All these quantities were averaged over 100 independent statistical runs for each pair \((p, k)\).

3. Results

We examined the effect of the packing fraction, \( p \), on pattern formation. Figure 2 presents examples of the final patterns \((t_{MC} = 10^7)\) at different values of \( p \) for \( k = 9 \). Up to \( p = 0.4 \), no regular patterns occurred despite visible rearrangement of the system (figure 2(a)–(d)). Then, stripe domains formed when the value of the packing fraction, \( p \), was between \( p_{down} = 0.5 \) and the jamming concentration, \( p_j \), (figure 2(e)–(h)). Slightly above \( p_j \) (\( p \approx 1.02p_j \)), stripe domains


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did not form within $10^7$ MC steps (figure 2(j)), nevertheless, rearrangement of the system was still continuing. For larger values of $p$, the mobility of the $k$-mers was effectively constrained, namely, a $k$-mer could perform small longitudinal irregular oscillations with zero mean square displacement (figure 2(j)).

Figure 2. Examples of systems at $t_{MC} = 10^7$ for different values of the packing fractions, $p$. $k = 9$, $L = 256$. (a) $p = 0.1$, (b) $p = 0.2$, (c) $p = 0.3$, (d) $p = 0.4$, (e) $p = 0.5$, (f) $p = 0.6$, (g) $p = 0.7$, (h) $p = p_j \approx 0.743$, (i) $p = 0.75$, (j) $p = 0.925$.

Figure 3 demonstrates some examples of variations of the relative number of contacts, $n_{xy}^*$, (a) and of the normalised number of clusters, $n$, (b) vs MC steps, $t_{MC}$, for different values of the packing fraction, $p$, with a fixed value of $k$, $k = 9$. Packing fractions $p = 0.5, 0.6, 0.7$ show pattern formation, whereas no pattern formation was observed for smaller values of packing fractions. Both $n_{xy}^*(t_{MC})$ and $n(t_{MC})$ curves show a stepwise decrease between $t_{MC} = 10^5$ and $t_{MC} = 10^6$ when their packing fractions correspond to pattern formation.

Figure 3. Examples of the dependencies of the relative number of contacts, $n_{xy}^*$, (a) and the normalised number of clusters, $n$, (b) vs MC steps, $t_{MC}$, for different values of the packing fraction, $p$, with a fixed value of $k$, $k = 9$. 

\[\text{Figure 2. Examples of systems at } t_{MC} = 10^7 \text{ for different values of the packing fractions, } p. \] 
\[k = 9, L = 256. \ (a) p = 0.1, (b) p = 0.2, (c) p = 0.3, (d) p = 0.4, (e) p = 0.5, (f) p = 0.6, (g) p = 0.7, (h) p = p_j \approx 0.743, (i) p = 0.75, (j) p = 0.925. \]

\[\text{Figure 3 demonstrates some examples of variations of the relative number of contacts, } n_{xy}^*, (a) \text{ and of the normalised number of clusters, } n, (b) \text{ vs MC steps, } t_{MC}, \text{ for different values of the packing fraction, } p, \text{ with a fixed value of } k, k = 9. \] 

\[\text{Packing fractions } p = 0.5, 0.6, 0.7 \text{ show pattern formation, whereas no pattern formation was observed for smaller values of packing fractions. Both } n_{xy}^*(t_{MC}) \text{ and } n(t_{MC}) \text{ curves show a stepwise decrease between } t_{MC} = 10^5 \text{ and } t_{MC} = 10^6 \text{ when their packing fractions correspond to pattern formation.} \]
Figure 4 demonstrates how the relative number of contacts, $n_{xy}^*$, and the normalised number of clusters, $n$, varies with the packing fraction, $p$, for $k = 12$. The occurrence of stripe domains is not accompanied by any characteristic change in the curves. Hence, these quantities are of little value for monitoring self-organisation and its characterisation.

By contrast, the local order parameter offers clear evidence of long-range pattern formation (figure 5). Significant growth of the order parameter after $10^5$ MC steps is a result of cluster coarsening. This effect is pronounced for packing fractions $p = 0.5, 0.6, 0.7$ but almost imperceptible for smaller packing fractions values.

The transition from a homogeneous steady-state to a patterned one appears to be continuous, i.e., the stripe domains become less pronounced for smaller initial packing fractions. For some initial packing fractions, the steady-state looks quite homogeneous (figure 2(d)) whereas imperfect stripe domains start to form at larger initial packing fractions (figure 2(e)). All the quantities of interest change smoothly as the packing fraction changes (figure 3). However, near the critical packing fraction, $p_{down}$, changes in the local order parameter are more visible (figure 6). This suggests the local order parameter is a useful quantity, suitable for building a
phase diagram. For this purpose, we used the relative local order parameter, \( s^* = s_f/s_i \), where \( s_i = s(0) \), \( s_f = s(10^7) \).

Figure 6. Example of the dependency of relative local order parameter, \( s^* = s_f/s_i \), vs the packing fraction, \( p \), for \( k = 12 \).

Figure 7 presents a phase diagram for the systems under study in the \((k,p)\)-plane. In the phase diagram, the region with diagonal stripe formation in steady state at \( t_{MC} \gtrsim 10^7 \) is marked as SD (this region is coloured from red to light blue in the online version); the region that corresponds to an absence of stripe domains is marked as NSD (this region is coloured in lilac in the online version); the transient region is marked as T (this region is shown in dark blue in the online version). No diagonal stripes can be observed at small packing fractions below some critical value, \( p_{down} \). This critical value, \( p_{down} \), decreases with increased value of \( k \).

Figure 7. Phase diagram in the \((k,p)\)-plane; SD corresponds to the values of \( k \) and \( p \) when stripe domain formation occurs; NSD corresponds to the values of \( k \) and \( p \) when no stripe domain formation is observed; T corresponds to the transient region.

It was argued in [18] that diagonal stripe domains can be produced only by “myopic particles. By contrast, “blind particles demonstrate segregation into two regions of arbitrary shapes. Nevertheless, this problem needs additional study.

Pattern formation affects the connectivity between domains, and this can lead to a change in electrical conductivity. The conventional model is more suitable for describing the initial reorganisation of the system under consideration. When the packing fraction slightly exceeds the
percolation threshold, the system in its initial state, i.e., a disordered system, is a conductor. For this particular case, reorganisation of the system leads to a decrease in the electrical conductivity due to decay of the percolation cluster. Figure 8 demonstrates the conductor–insulator phase transition for $k = 10$ and $p = 0.5$. This packing fraction is a little greater than the percolation threshold, $p_c \approx 0.47$. By contrast, for $p = 0.7$ (essentially greater than the percolation threshold) and $p = 0.4$ (somewhat below the percolation threshold), diffusion does not lead to a phase transition from conductor to insulator.

Figure 8. Examples of the temporal dynamics of electrical conductivity for $k = 10$ and different values of the packing fraction, $p$, $L = 256$. (a) Conventional model, (b) Insulating ends model.

The insulating ends model can catch long-range pattern formation. This model demonstrates quite different behaviour. For small values of the packing fractions, the system is an insulator throughout simulation, whereas for values of the packing fraction close to the jamming concentration, there is an insulator–conductor phase transition. This transition occurs due to the formation of dense stripe domains. Inside such domains, lateral contacts of $k$-mers ensure electrical conductivity in both directions.

4. Conclusion

Diffusional reorganisation in two-dimensional systems of rigid rod-like particles has been simulated using a lattice approach. Rod-like particles were presented as linear $k$-mers of two mutually perpendicular orientations on a square lattice with periodic boundary conditions (torus). An initial homogeneous distribution of $k$-mers was produced using random sequential adsorption. The packing fraction, $p$, was varied in the range from 0.1 to its jamming value, $p_j$. By means of the Monte Carlo technique, translational diffusion of the $k$-mers was simulated as a random walk. In our model, a particle obligatorily moves when it has any possibility of changing its location. For $k \geq 6$, the system tends toward a well-organised nonequilibrium steady state in the form of diagonal stripes, when the packing fraction exceeds a critical value.

Additionally, we studied the behaviour of high-density systems ($p > p_j$) produced using random sequential adsorption with diffusion and additional adsorption. In such systems, diffusivity is quite small. This results both in a significant increase of the relaxation time and in a suppression of reorganisation. Even for $p \approx 1.02p_j$, stripe domain formation was not observed within $10^8$ MC steps.

It is of note, that pattern formation was absent both in dilute systems and in high-density systems. Stripe domain formation was observed only for systems of intermediate density. There
is a curious analogy with social systems, viz., social self-organisation is possible when the average number of social links per person is large enough (high population density) and individual freedom is simultaneously ensured.

Figure 9 presents the phase diagram obtained using both models.

![Phase diagram](image)

**Figure 9.** Phase diagram in the \((k,p)\)-plane.

**Appendix**

To transform the system under consideration into an RRN, we associated each bond between two empty sites of the lattice with conductivity \(\sigma_m\), each bond between two sites belonging to the same \(k\)-mer with conductivity \(\sigma_k\), each bond perpendicular to the \(k\)-mer with conductivity \(2\sigma_t\), and each end bond of the \(k\)-mer with conductivity \(2\sigma_e\) (figure A1(a)). The resulting conductivities between the two different sites \(i\) and \(j\) were calculated as

\[
\sigma_{ij} = \frac{2\sigma_t\sigma_j}{\sigma_i + \sigma_j}
\]

All possible combinations are presented in figure A1(b).

In the so-called conventional model, \(\sigma_k = \sigma_t = \sigma_e = 10^6\) a.u. and \(\sigma_m = 1\) a.u. In the so-called insulating ends model, \(\sigma_k = \sigma_t = 10^6\) a.u. and \(\sigma_m = \sigma_e = 1\) a.u.

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Figure A1. Transformation of a lattice with $k$-mers into an RRN ($k = 3$).

References
[1] Narayan V, Menon N and Ramaswamy S 2006 J. Stat. Mech. Theor. Exp. 2006 P01005 ISSN 1742-5468
[2] Galanis J, Harries D, Sackett D L, Losert W and Nossal R 2006 Phys. Rev. Lett. 96 028002 ISSN 0031-9007
[3] Aranson I S, Volson D and Tsimring L S 2007 Phys. Rev. E 75 051301 ISSN 1539-3755
[4] Galanis J, Nossal R, Losert W and Harries D 2010 Phys. Rev. Lett. 105 168001 ISSN 0031-9007
[5] Yadav V and Kudrolli A 2012 Eur. Phys. J. E 35 104 ISSN 1292-895X
[6] Börzsönyi T and Stannarius R 2013 Soft Matter 9 7401–7418 ISSN 1744-6848
[7] Müller T, de las Heras D, Rehberg I and Huang K 2015 Phys. Rev. E 91(6) 062207 ISSN 2470-0045
[8] González-Pinto M, Borondo F, Martínez-Ratón Y and Velasco E 2017 Soft Matter 13(14) 2571–2582 ISSN 1744-6848
[9] Aranson I S and Tsimring L S 2006 Rev. Mod. Phys. 78(2) 641–692 ISSN 0034-6861
[10] Evans J W 1993 Rev. Mod. Phys. 65(4) 1281–1329 ISSN 0034-6861
[11] Centres P M and Ramirez-Pastor A J 2015 J. Stat. Mech. Theor. Exp. 2015 P10011 ISSN 1742-5468
[12] Kuriata A, Polanowski P and Sikorski A 2016 Macromol. Theor. Simul. 25 360–368 ISSN 1521-3919
[13] Budinski-Petković L, Lončarević I, Dujak D, Karač A, Šćepanović J R, Jakšić Z M and Vrhovac S B 2017 Phys. Rev. E 95(2) 022114 ISSN 2470-0045
[14] Lebovka N I, Tarasevich Y Y, Gigiberiya V A and Vygornitskii N V 2017 Phys. Rev. E 95(5) 052130 ISSN 2470-0045
[15] Tarasevich Y Y, Laptev V V, Burmistrov A S and Lebovka N I 2017 J. Stat. Mech.: Theory Exper. 2017 093203 ISSN 1742-5468
[16] Fusco C, Gallo P and Petri A and Rovere M 2001 J. Chem. Phys. 114 7563–7569 ISSN 0021-9606
[17] Selinger R B and Stanley H E 1990 Phys. Rev. A 42(8) 4845–4852 ISSN 1050-2947
[18] Patra S, Das D, Rajesh R and Mitra M K 2018 Phys. Rev. E 97(2) 022108 ISSN 2470-0045
[19] Landau D P and Binder K 2014 A Guide to Monte Carlo Simulations in Statistical Physics 4th ed (Cambridge: Cambridge University Press) ISBN 9781107074026
[20] Stauffer D and Aharony A 1992 Introduction to Percolation Theory (London: Taylor & Francis)
[21] Hoshen J and Kopelman R 1976 Phys. Rev. B 14(8) 3438–3445 ISSN 2469-9950
[22] Frank D J and Lobb C J 1988 Phys. Rev. B 37(1) 302–307 ISSN 2469-9950