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1 Introduction

The control over the self-organization of colloidal particles is a problem of both fundamental and practical interest.^{1–10} One promising approach is the use of chemically or otherwise patterned substrates, where the particle–substrate interaction is spatially dependent and may even change with time. Spatial patterns on substrates may be created using, for example, lithographic methods,^{11,12} magnetic domains,^{13,14} chemical coating,^{15–17} or DNA-mediated functionalization of interfaces.¹⁸

It has been shown that the equilibrium phases of colloidal particles are affected strongly by substrates with spatial patterns. For example, patterns may induce new surface phases¹⁹ and crystalline structures,²⁰ or affect the wetting properties of the surfaces.²¹ The collective (non-equilibrium) dynamical properties are also affected. For example, in the limit of irreversible adsorption, a simple pattern of pits distributed in a square-lattice arrangement induces either local or long-range order, depending on the size of the pits and the distance between them.²²

For simplicity, most of the previous works have considered isotropic particles but, in general, the interparticle interaction is anisotropic. Anisotropy may result, for example, from the

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Crossover from three- to six-fold symmetry of colloidal aggregates in circular traps

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At sufficiently low temperatures and high densities, repulsive spherical particles in two-dimensions (2d) form close-packed structures with six-fold symmetry. By contrast, when the interparticle interaction has an attractive anisotropic component, the structure may exhibit the symmetry of the interaction. We consider a suspension of spherical particles interacting through an isotropic repulsive potential and a three-fold symmetric attractive interaction, confined in circular potential traps in 2d. We find that, due to the competition between the interparticle and the external potentials, the particles self-organize into structures with three- or six-fold symmetry, depending on the width of the traps. For intermediate trap widths, a core-shell structure is formed, where the core has six-fold symmetry and the shell is three-fold symmetric. When the width of the trap changes periodically in time, the symmetry of the colloidal structure also changes, but it does not necessarily follow that of the corresponding static trap.

individual particle shape,^{23–27} heterogeneous distribution of charges,^{28–30} or functionalization of the particle surface.^{31–41} In these cases, the final structures should depend not only on the symmetries of the pattern but also on those of the interparticle potential. In a recent study the equilibrium properties of particles with three-fold symmetric attractive interaction adsorbed on patterned substrates were considered.⁴² The properties of the pattern strongly affect both the percolation properties and the type of network in which the particles self-assemble.

Here, we investigate how the dynamics of self-organization of colloidal particles with anisotropic interparticle interactions is affected by the presence of spatial patterns. These patterns result from a square lattice arrangement of (Gaussian) attractive traps with a characteristic width on an otherwise flat substrate. We show that, the structure of the colloidal aggregates, on the substrate, depends strongly on the width of the traps. We consider also traps with a time dependent width and show that the dynamics may differ significantly from that of the static traps.

We introduce the model and the relevant parameters in Section 2. In Section 3 we present the results and we draw some conclusions in Section 4.

2 Model

We consider a monodisperse suspension of spherical (colloidal) particles, where the particle–particle interaction is a superposition of an isotropic repulsion and a three-fold symmetric attraction. Following ref. 43, we describe this pairwise interaction by

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Fig. 1 Schematic representation of the attractive Gaussian potential traps distributed in a square-lattice arrangement. The traps are separated by a distance of ten particle diameters (d_p) and have a width (a) $R_W = 2$, (b) $R_W = 4$, and (c) $R_W = 6$ in units of the particle diameter (d_p). The potential strength is in units of ε .

decorating the surface of the spherical particles with three patches distributed along the equator. The patch-patch attractive interaction has a Gaussian form given by,

$$U_{\text{patch/patch}}(r_{\text{p}}) = -\varepsilon \exp[-(r_{\text{p}}/\sigma)^{2}], \qquad (1)$$

where $r_{\rm p}$ is the distance between the center of the patches, ε is the interaction strength that sets the energy scale, and $\sigma = 0.1$ the width of the Gaussian in units of the effective particle diameter $d_{\rm p}$ (which sets the length scale).

The core-core interaction is repulsive and given by,

$$U_{\text{part/part}}(r) = \frac{A}{k} \exp\left[-k\left(r - d_{\text{p}}\right)\right],\tag{2}$$

where *r* is the distance between the center of the particles, A = 0.25 (in units of ε/d_p), and k = 0.4 is the screening length (in units of d_p).

To confine the particles to the surface of a planar substrate, we implemented the method described in ref. 43. The surface pattern consists of attractive potential traps, distributed in a square-lattice arrangement (see Fig. 1), with a Gaussian form,

$$U_{\text{part/trap}}(r) = -3\varepsilon \exp[-(r/R_{\text{W}})^2], \qquad (3)$$

where ε is the strength of the patch–patch interaction (see eqn (1)) and $R_{\rm W}$ is the width (range) of the trap. The potential is truncated at a distance of 10 particle diameters ($d_{\rm p}$) from the center of the trap. As shown in Fig. 1, although the minimum of the traps is kept fixed, the effective potential landscape depends not only on $R_{\rm W}$, but also on the distance between the center of the traps, as in some regions the particles interact simultaneously with more than one trap. Since the particle–trap interaction is always attractive, this implies that the net force acting on a particle is lower if the traps overlap. We impose periodic boundary conditions along the *x*- and *y*-directions. This pattern can be realized experimentally, for instance, using a setup of multiple optical tweezers arranged on a square array, by passing a laser beam through diffractive optical elements.⁴⁴

To resolve the trajectory of individual particles, we perform Langevin dynamics using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS).⁴⁵ Particles are spherical with mass m and inertia I and the patches on their surface have negligible mass. The translational and rotational motion of the particles is described by the following equations,

$$m\dot{\vec{v}}_i(t) = -\nabla_{\vec{r}_i} U - \frac{m}{\tau_t} \vec{v}_i(t) + \sqrt{\frac{2mk_B T}{\tau_t}} \vec{\xi}_i^i(t), \qquad (4)$$

and

$$I\vec{\omega}_{i}(t) = -\nabla_{\vec{\theta}_{i}}U - \frac{I}{\tau_{\rm r}}\vec{\omega}_{i}(t) + \sqrt{\frac{2Ik_{\rm B}T}{\tau_{\rm r}}}\vec{\xi}_{\rm r}^{i}(t), \qquad (5)$$

where, \vec{v}_i and $\vec{\omega}_i$ are the translational and angular velocities of particle *i*. The translational and rotational damping times are given by $\tau_t = 0.02 \sqrt{(md_p^2/\varepsilon)}$ and $\tau_r = 10\tau_t/3$ for spherical colloids. $\vec{\xi}_t^i(t)$ and $\vec{\xi}_r^i(t)$ are stochastic terms taken from a truncated random distribution of zero mean and standard deviation of one unit.⁴⁶ *U* is the total potential with contributions from the particle–particle and particle–trap interactions, and therefore it depends on both the positions \vec{r}_i and orientations $\vec{\theta}_i$ of all particles i = 1...N in the system. Note that, although the particles are on a planar substrate, they can still rotate in three dimensions.

3 Results

Particles are initially distributed, without overlapping, uniformly at random on the substrate with a given particle number density ρ , defined as the number of particles per unit volume. Simulations were performed at a reduced temperature $T^* = k_{\rm B}T/\varepsilon$, where *T* is the thermostat temperature, ε the strength of the patch-patch interaction and $k_{\rm B}$ the Boltzmann constant. Unless otherwise stated, we rescale the time by the Brownian time $\tau_{\rm B} = d_{\rm p}^{-2}/D_{\rm t}$, where $D_{\rm t}$ is the translational diffusion coefficient $D_{\rm t} = k_{\rm B}T\tau_{\rm t}/m$. $\tau_{\rm B}$ is related to the typical time taken by a particle to diffuse in an area $d_{\rm p}^{-2}$, considering an overdamped regime.⁴⁷ All results are averages over (i) 10 different realizations and (ii) all traps in each realization.

To evaluate the local structure formed by the colloidal particles, we measure the local *k*-fold bond order parameter of the *i*-th particle,⁴⁸

$$\phi_{k=\{3,6\}}^{(i)} = \left| \frac{1}{\max\{N_{l}, k\}} \sum_{j=1}^{N_{l}} e^{-ik\theta_{ij}} \right|,$$
(6)

where N_i is the number of neighbors around the particle within a cut-off radius $r_{\text{cut}} = 13$ in units of the particle diameter (d_p) . θ_{ij} is the angle between the vector connecting particles *i* and *j* and the *x*-direction (parallel to the substrate). *k* is a parameter related to the local symmetry such that, ϕ_6 is one for perfect sixfold symmetry and ϕ_3 is one for perfect three-fold symmetry. We then define n_k as the fraction of particles with a value of ϕ_k above a specific threshold that we set to 1/2.

In what follows, we analyze first the dynamics at a temperature $T^* = 0.0625$ which is below the $3D^{49}$ and $2D^{50}$ gas–liquid critical temperature (note that the 2D bulk phase diagram may be the most relevant for our system). Then, we consider traps with a time-dependent width at temperatures $T^* = \{0.0625, 0.075, 0.1, 0.125\}$, corresponding to a range of temperatures encompassing the limits of irreversible and reversible bonds.

3.1 Constant potential traps

Fig. 2 shows the time evolution of the fraction of particles with a local (a) three- (n_3) and (b) six-fold (n_6) symmetry, for different values of the width of the traps $(R_W = 2, 4 \text{ and } 6)$ but the same initial particle number density $\rho = 0.25$. This density was chosen to be well below the six-fold equilibrium configuration and also below the percolation threshold taken from ref. 42.



Let us focus on the case $R_W = 6$ (squares), which is the largest value that we have considered. Both n_3 and n_6 increase in time as the particles accumulate in the potential traps. However, the asymptotic value of n_3 is about two orders of magnitude larger than n_6 , as most particles have a local three-fold symmetry, in line with the symmetry of the patch-patch attractive potential. By contrast, when the width of the trap is reduced, the values of n_3 and n_6 are comparable. For $R_W = 2$, the fraction of particles with a local six-fold symmetry is even larger than that of particles with a three-fold symmetry (*i.e.*, $n_6 > n_3$). This can be explained by the following mechanism. Due to the particle-trap interaction, particles are dragged towards the center of the potential traps, increasing the local density there. The dragging forces are stronger at lower values of R_W (see eqn (3)). Thus, while for $R_{\rm W}$ = 6 the symmetry of the aggregates resembles that of the particle-particle attractive potential, at a lower pressure that corresponds to a lower packing, for $R_W = 2$, the attractive particletrap forces favor an increase in the local density (packing), with a corresponding increase of the local pressure in the region of the trap, leading to the emergence of the six-fold symmetry.

Fig. 2 shows that the dynamics for traps with $R_W = 4$ exhibits a non-monotonic behavior of n_3 . Fig. 3 depicts snapshots of the



Fig. 2 Time dependence of the fraction of particles with a local (a) three-(n_3) and (b) six-fold (n_6) bond order parameter above 1/2 for different traps with different widths, namely, $R_W = \{2, 4, 6\}$. Simulations are performed on a substrate of linear size L = 40 (in units of the particle diameter d_p). The expected values of n_6 for isotropic colloids (no patches) and traps of width $R_W = \{2, 4, 6\}$ are, respectively, $n_6 = \{0.47, 0.40, 0.04\}$.

Fig. 5 Bottom: Time dependence of the fraction of particles (r_3) with a local three-fold bond order parameter above 1/2 for traps with $R_W = 4$ and particle number density $\rho = 0.25$. Simulations are performed on a substrate of linear size L = 40. Top: Snapshots of the structure at different times (as marked in the bottom plot). Blue and green particles have a local six- and three-fold bond order parameter above 1/2, respectively. In black are the particles with none of the two local bond order parameters above the threshold.

structure at different instants, as pointed out in the plot in the bottom of the same figure. Initially (in I), the particles are randomly distributed in space, without overlapping. As the potential traps are switched on, the particles are dragged towards the center of the traps, establishing bonds with other particles. The value of n_3 increases (from I to II), since the particle–particle attractive interaction favors three bonds per particle (green particles). As more particles are attracted to the traps (from II to III), the fraction of particles with six neighbors in the center of the trap (blue particles) increases and the value of n_3 decreases. As the aggregates grow (from III to IV), the outer particles are under weaker trap forces than the inner ones, favoring again the particle–particle bonds over packing.

When $R_W = 4$, the increase in n_6 is in fact a transient. As shown in Fig. 4, although n_6 initially increases due to packing, it eventually decreases at longer times. Particles in the aggregates relax slowly to form domains with six-fold symmetry with strong particle–particle bonds along the grain boundaries (see inset of Fig. 4). As a result, the fraction of particles with six-fold symmetry is reduced and the value of ϕ_k for a large fraction of the particles is below the threshold for both k = 3 and 6.

Fig. 5 shows the number density (ρ) of particles around the center of the trap for R_W = 4. Fig. 5(a) and (b) show $\rho(r,n_3)$ and



Fig. 4 Time dependence of the fraction of particles (n_6) with a local sixfold bond order parameter above 1/2 for traps with $R_W = 4$ and particle number density $\rho = 0.25$. Simulations are performed on a substrate of linear size L = 40. Snapshots are examples of clusters at the point where the maximum occurs (left) and at the end of the simulation (right). Blue and green particles have a local six- and three-fold bond order parameter above 1/2, respectively. In black are the particles where none of the two local order parameters are above threshold. The zoomed region at the top shows the lines of defects (red-dotted line) between six-fold regions (green-dotted line).



Fig. 5 (a) Number density of particles with a local six-fold bond order parameter above 1/2, measured from the center of the traps at times $t = \{0.20, 0.50, 1.13, 7.50\}\tau_{\rm B}$. (b) Number density of particles with a local three-fold bond order parameter above 1/2, measured from the center of the traps at times $t = \{0.20, 0.50, 1.13, 7.50\}\tau_{\rm B}$. Solid lines are averages over neighboring points on the left and right to show the overall tendency. Simulations were performed on a square substrate of size L = 40.

 $\rho(r,n_6)$, respectively. Note that the maxima of n_3 and n_6 occur at different positions. This difference corroborates the hypothesis that the local three- and six-fold symmetric structures are formed in different regions. In the center of the trap, the particles self-organize (pack) with six-fold symmetry, while in the perimeter most particles form three bonds with other particles. The time evolution of the radial distribution function shows that, at early times, both structures form near the center of the trap independently. As time evolves, a separation of the structures is observed, with the six-fold structure in the center of the trap and the three fold one in the perimeter. Asymptotically, the six-fold peak of the radial distribution function decreases slightly due to the rearrangement discussed above.

3.2 Time-dependent potential traps

The results reported above suggest the design of a device, where the symmetry of the aggregates may be dynamically switched from three to six fold through the width of the potential traps. The effectiveness of such a device, however, depends on how the rate of the symmetry change compares to the different timescales involved, namely those related to bond breaking/ formation and rotational/translational diffusion.^{47,51}

We considered time dependent trap widths, as shown in Fig. 6(a). We start with traps with $R_W = 2$ and periodically increase R_W linearly to $R_W = 6$ and then reduce it back (linearly)



Fig. 6 (a) Time dependent width of the potential traps (R_W) that varies periodically between two and six. (b) Time dependence of the fraction of particles (n_6) with a local six-fold and (n_3) three-fold bond order parameter above 1/2 for traps with the same oscillating width R_W , particle number density $\rho = 0.3$, and reduced temperature $T^* = 0.125$. Here, time is rescaled in units of the period of the oscillation. Simulations are performed on a square substrate of size L = 40. The solid line is the equilibrium order parameter for a potential of constant width $R_W = 4$. Simulation snapshots at the two limiting potential widths (c) $R_W = 6$ and (d) $R_W = 2$ for the oscillating traps. Blue and green particles have a local six- and three-fold bond order parameter above 1/2, respectively. In black are the particles where none of the two local order parameters are above the threshold.

to $R_{\rm W} = 2$. Let us discuss the dynamics at $T^* = 0.125$, the highest temperature considered here, here the oscillations have a period $\nu^{-1} = 20\tau_{\rm B}$. In Fig. 6(b) we plot the fraction of particles with a local six- and three-fold symmetry, as a function of time. Here, for convenience, time is rescaled in units of the period of the oscillation of $R_{\rm W}$ (ν^{-1}). At this temperature, the local structure oscillates between three-fold and six-fold symmetry, with the same frequency and phase of $R_{\rm w}$ (see snapshots Fig. 6(c) and(d)). A local minimum in n_6 is observed at $R_{\rm W} = 2$ indicating that the optimal width of the trap to promote local six-fold symmetry is around three particle diameters ($d_{\rm p}$).

Fig. 7(b) shows the time dependence of n_6 (black squares) for the same oscillating R_W at a slightly lower temperature ($T^* = 0.1$). In this case, n_6 oscillates with a period $\nu^{-1} = 16\tau_B$, but the phase is shifted by approximately 1/4 of the period. To investigate the dependence on the initial conditions, we applied the same oscillating trap R_W , starting at $R_W = 6$ (red circles in Fig. 7). The same shift is observed, and the periodic behavior at longer times is clearly independent of the initial conditions.

When the temperature is reduced further, namely, $T^* = \{0.0625, 0.075\}$ (corresponding to periods of $\nu^{-1} = \{10, 12\}\tau_B$), we observed a similar shift in the (weaker) oscillations and a marked dependence



Fig. 7 (a) Time dependent width of the potential traps (R_W) which varies periodically between two to six. (b) Time dependence of the fraction of particles (n_6) with a local six-fold bond order parameter above 1/2 for oscillating traps with initial widths $R_W(0) = 2$ (squares) and $R_W(0) = 6$ (circles) for a particle number density $\rho = 0.3$ and reduced temperature $T^* = 0.1$. Here, time is rescaled in units of the period of the oscillation. Simulations are performed on a square substrate of size L = 40. The solid line gives the equilibrium order parameter for a fixed potential of $R_W = 4$.



Fig. 8 Time dependence of the fraction of particles (n_6) with a local six-fold bond parameter above 1/2 for traps with R_W varying periodically from two to six with initial trap widths $R_W(0) = 2$ and $R_W(0) = 6$ (same as Fig. 7(a)) and reduced temperatures $T^* = 0.0625$ (squares) and $T^* = 0.075$ (circles), for a particle number density $\rho = 0.3$. Here, time is rescaled in units of the period of the oscillation. Simulations are performed on a square substrate of size L = 40.

on the initial conditions. This is shown in Fig. 8, at different temperatures (different symbols) and initial conditions (top and bottom curves, respectively). The timescales of the relaxation mechanisms (bond break/formation and translation/rotational diffusion) depend strongly on temperature. Thus, at sufficiently low temperatures, the rate of change of $R_{\rm W}$ is too fast preventing the particles to relax to the structures expected for the corresponding static traps $R_{\rm W}$.

4 Conclusions

We have studied the dynamics of spherical colloidal particles on a surface in the presence of circular potential traps. The attractive interaction between the particles has three-fold symmetry. However, in the presence of potential traps, we observe a crossover from the expected local three-fold symmetry to a six-fold one when the width of the traps is reduced. For intermediate values of the trap width, we find a core-shell structure, where the symmetry in the core is six fold, while that in the shell is three fold. Note that, for a fixed distance between the centers of the traps, increasing the width of the traps corresponds to smoothing out the external potential landscape. Thus, we expect similar results when the strength of the potential is changed, at fixed width.

For traps with oscillating widths, we find that the final structure may deviate significantly from the thermodynamic one, when the relaxation timescales are comparable to the period of the oscillations. In this limit, the final structures depend on the rate of change of the trap width, the thermostat temperature, and the initial conditions.

Conflicts of interest

There are no conflicts to declare.

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