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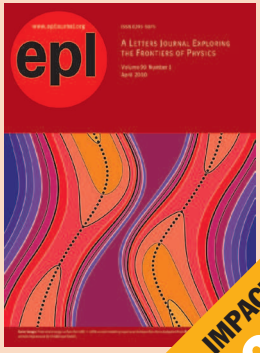

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Director field in plastic crystals

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Abstract – The director field in a plastic crystal is calculated by particle-resolved Monte Carlo computer simulations of two-dimensional, slightly anisometric hard spherocylinders exposed to an external periodic substrate potential. We investigate the structure of the director field in the Wigner-Seitz cell and find a topological defect structure that can be controlled with the substrate potential. At zero potential we find a charge $-1/2$ defect at the corners besides the expected defect in the centre with unit topological charge. When switching the substrate potential on, the corner defects are surrounded by three satellite defects which bear charge $-1/2$, too. Additionally, we then find two charge $+1/2$ defects on each edge of the unit cell. Finally, within a simplified model, we obtain a qualitative explanation for this defect structure. Our predictions can in principle be verified by using particle-resolved experiments of colloidal plastic crystals.

At appropriate thermodynamic conditions, systems composed of non-spherical particles can self-assemble into meso-phases [1], which show both liquid- and solid-like order [2,3]. This is due to the fact that the translational and the orientational degrees of freedom exhibit different kinds of ordering. One of the emerging meso-phases is a plastic crystal (or rotator crystal) in which the averaged centre-of-mass positions of the particles are ordered on a crystalline lattice while there is no global orientational order (polar or nematic).

Plastic crystals are found for mesoscopic colloidal particles [4–6] and for molecules [7,8] both in three dimensions and in two-dimensional sheets. In fact, colloidal particles can be strongly confined to almost two dimensions [9–11] and molecular plastic-crystalline monolayers are typically observed on structured substrates [12]. In general, plastic crystals have special elasto-mechanical properties, represent ideal orientational glass formers [13–15] and are —in their colloidal realization— promising candidates for photonic crystals [16,17]. Clearly, plastic crystals are only expected for slightly anisometric particles in order to allow for an almost unhindered rotation around the lattice sites. However, the particles fluctuate around the mean lattice positions and the rotational degree(s) of freedom of a particle that is displaced from the perfect lattice position is confined by the neighbouring particles resulting in a preferred mean orientation. The latter can be regarded as a director field $\hat{\mathbf{n}}(\mathbf{r})$ of a plastic crystal which is lattice periodic, *i.e.*, defined in the Wigner-Seitz cell of the crystal, where $\hat{\mathbf{n}}$ is a unit vector and $\mathbf{r}$ a given position.

The director field $\hat{\mathbf{n}}(\mathbf{r})$ in plastic crystals can bear topological defects [18] as any director field in liquid-crystalline phases (*e.g.*, in the nematic phases). The defect structure has been studied in great detail for two-dimensional nematics confined on a sphere (so-called nematic bubbles or shells) [19–21] and in three-dimensional nematic droplets (so-called "tactoids") [22–24] as well as for spherical colloidal particles immersed in a nematic solvent [25–27]. However, there is much less work done in the plastic-crystalline phase. The important point to mention here is that for plastic crystals the director field $\hat{\mathbf{n}}(\mathbf{r})$ varies on the microscopic scale, *i.e.*, on the scale of the particle extensions. In the macroscopic context of elasticity theory, the director is defined as the average orientation within a subvolume of the system containing many particles. From a microscopic point of view, the director is defined as an average over the orientations of single particles during many visits of their centres of mass to a subvolume much smaller than their size.

In most of the approaches, the director field is assumed to be completely disordered [28] or only globally directed as a response to external fields [29] or the orientation has

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been modelled on a purely phenomenological level [30]. As far as we know, the only work which addresses the director field in bulk plastic crystals in detail from a more microscopic point of view including a discussion of the topological defects structure is ref. [31]. In their approach, Achim *et al.* apply a coarse-grained phase-field-crystal model [32] designed for liquid-crystalline phases [33,34] to a two-dimensional plastic crystal. A defect with positive unit topological charge was found at the lattice positions while there is a defect with topological charge $-1/2$ in the corners of the Wigner-Seitz cell.

In this letter, we address the director field of a plastic crystal by a particle-resolved Monte Carlo computer simulation [35–38] of two-dimensional hard spherocylinders [39] which are only slightly anisotropic. For these particles, a plastic crystal with a triangular lattice is found at intermediate densities. We apply a triangular external potential to the system, which increases the stability of the plastic crystal. The external potential mimics the particle-substrate interactions for the molecular case [12], but can also be realised by superimposing laser-optical fields for the colloidal case [40,41]. It constrains the translational fluctuations which destroy long-range positional ordering in two spatial dimensions [42]. For zero substrate potential amplitude we confirm in general the gross defect structure obtained in ref. [31] for a triangular crystal. When the potential is switched on, however, the fine structure is different. In detail, we find the same charge $+1$ topological defect at the lattice position and $-1/2$ defects at the Wigner-Seitz cell corners as ref. [31]. However, the $-1/2$ defects at the Wigner-Seitz cell corners are surrounded by three satellite defects which bear the topological charge $-1/2$. The total topological charge neutrality required for a crystal with no global orientational order dictates that there are additional defects with positive topological charge. We find them in the form of charge $+1/2$ defects on the edges of the Wigner-Seitz cell. This defect structure is qualitatively explained in terms of a cell model for the plastic crystal. Only if the field is smeared close to the Wigner-Seitz cell corner by long wavelength fluctuations (as achieved for vanishing substrate potential), the rough structure predicted in ref. [31] is recovered. Our predictions can in principal be verified by using particle-resolved experiments of colloidal (lyotropic) plastic crystals. By exploiting the dependence of the defect structure on the substrate potential, it might be possible to use a plastic crystal as an optical switching device [43].

Model and simulation technique. – We consider a two-dimensional triangular plastic crystal of hard spherocylinders on a substrate potential, which supports the triangular lattice, see fig. 1 for a schematic representation. Due to the well-known Landau-Peierls instability, positional order decays logarithmically with the system size in two-dimensional crystals. To prevent this instability and to have a well-defined lattice on which our director field can be measured,

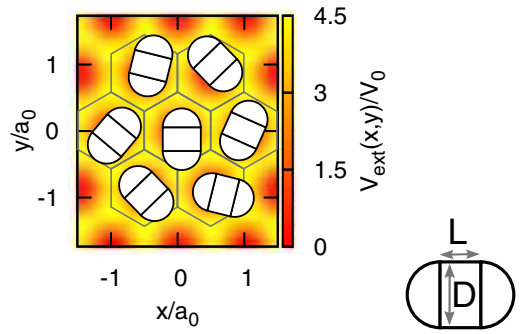

Fig. 1: (Color online) A schematic representation of the system (not to scale): a two-dimensional triangular plastic crystal of spherocylinders of length L and diameter D under the influence of a small-amplitude hexagonal substrate potential.

we impose the hexagonal substrate potential given by $V_{\text{ext}}(\mathbf{r}) = V_0\,[3 - \cos(\mathbf{k}_1 \cdot \mathbf{r}) - \cos(\mathbf{k}_2 \cdot \mathbf{r}) - \cos(\mathbf{k}_3 \cdot \mathbf{r})]$ [41], where $\mathbf{r} = (x, y)$ is the position of a particle and $\mathbf{k}_1 = [2\pi/a_0](1, 1/\sqrt{3})$, $\mathbf{k}_2 = [2\pi/a_0](0, 2/\sqrt{3})$ and $\mathbf{k}_3 = [2\pi/a_0](1, -1/\sqrt{3})$ are the reciprocal lattice vectors of a hexagonal lattice with lattice constant a_0. Unless mentioned otherwise, the amplitude V_0 was set to $0.1 k_B T$ and the number of particles to $N = 10000$ throughout this work. With these values for V_0 and N, system size effects were negligible. We apply standard Monte Carlo simulations [44] at fixed number of particles N, area A and temperature T. Periodic boundary conditions in a rectangular simulation box are employed in both directions. We initialise the spherocylinders with random orientations on ideal triangular lattice sites and then perform 10^6 equilibration moves per particle to obtain an equlibrium plastic crystal. Subsequently, $24 \cdot 10^6$ moves per particle are performed to gather statistics. The packing fraction is defined as $\eta \equiv N A_p / A$, where $A_p \equiv LD + \pi D^2/4$ is the area of a two-dimensional spherocylinder. The local nematic order parameter S and the nematic director were obtained by diagonalizing the nematic order parameter tensor [45] measured locally for particles at a certain position $\mathbf{r}$. We have considered spherocylinder aspect ratios of $0.1 \leqslant L/D \leqslant 0.25$ and packing fractions in the range $0.73 \leqslant \eta \leqslant 0.74$.

Results. – The director field within the Wigner-Seitz cell obtained from the simulation can be seen in fig. 2. Results are shown here for a spherocylinder aspect ratio of $L/D = 0.2$ and a packing fraction of $\eta = 0.74$. However, director fields for $L/D = 0.1, 0.15, 0.25$ and $\eta = 0.73, 0.74$ respectively show the same overall structure. At a topological defect, all directors are equally probable or equivalently the nematic order parameter is zero [46]. Therefore, the positions of the defects are those positions for which $S = 0$ in fig. 2. The charge (or winding number) of the topological defects can be determined by investigating the local nematic director field near the defect. The resulting defect structure is schematically

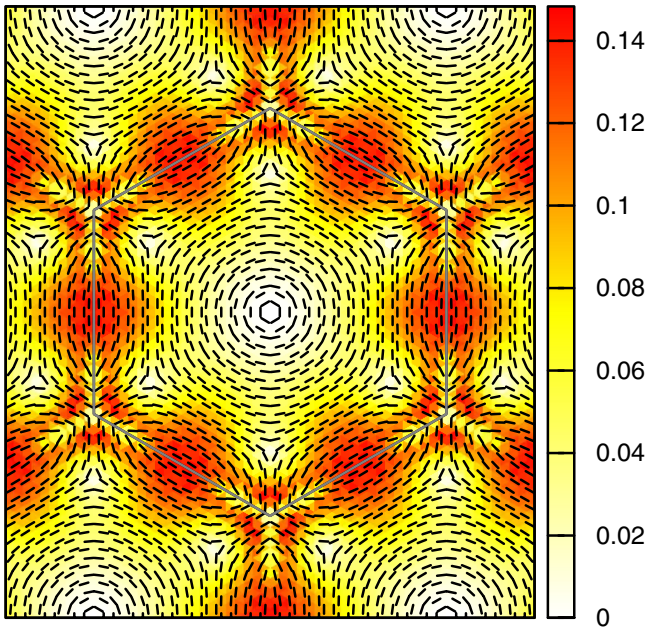

Fig. 2: (Color online) Director field obtained from Monte Carlo simulations for spherocylinders with aspect ratio $L/D = 0.2$ at a packing fraction of $\eta = 0.74$ superimposed on a color plot of the nematic order parameter field S. At the positions of topological defects S goes to zero.

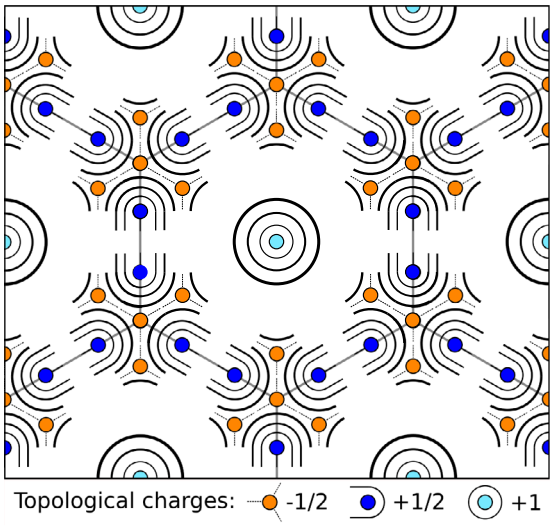

Fig. 3: (Color online) A schematic representation of the topological defect structure in the two-dimensional triangular plastic crystal of hard spherocylinders, also compare to the Monte Carlo results in fig. 2. Topological charges of positive winding number are denoted by blue dots, where the lighter dot in the centre has charge +1 and the darker blue dots on the edges of the unit cell have winding number +1/2. The orange dots denote defects with topological charge −1/2.

depicted in fig. 3. As mentioned above, the net charge of the unit cell is always zero for a plastic crystal. To determine the defect positions quantitatively we plot S along a path connecting the Wigner-Seitz cell's symmetry

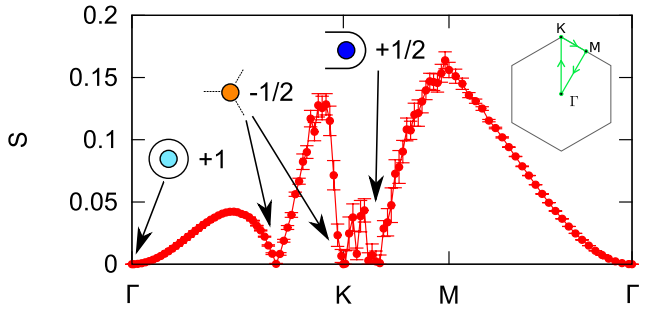

Fig. 4: (Color online) Nematic order parameter S along a path connecting the symmetry points of the Wigner-Seitz cell (the path is shown in the inset). It must vanish at Γ and K due to symmetry, but it does so additionally between Γ and K and between K and M in the many-particle Monte Carlo simulation.

points. This is shown in fig. 4. The symmetry points are the centre (Γ-point), the corner (K-point) and the centre of the edge (M-point). Again, points of vanishing S along the path reveal the defect positions. Defects at the Γ-point and at the K-point are dictated by the symmetry. They appear in the form of a +1 vortex defect in the cell's centre and a $-1/2$ wedge disclination in each corner. A $-1/2$ wedge disclination between Γ and K and a $+1/2$ wedge disclination between K and M appear in addition to the symmetry-dictated defects. A point worth noting is that the structure away from the defects appears to display as little bending in the local director field as possible given the presence of the defects. For instance, the directors near the line connecting the satellite defect positions are surprisingly parallel to this line. This is in agreement with the phase field crystal theory of ref. [31], in which a bending term, that penalises locally non-parallel directors, can be found in the expression for the free energy.

For small systems ranging from $N = 144$ to $N = 1024$ particles, the same defect structure was found (that of fig. 3) regardless of the potential strength V_0. However, when we set the substrate potential amplitude $V_0 = 0$ and increase the system size further, the defects move closer to each other and eventually merge, showing the combined charge. At a system size of $N = 10000$, only the symmetry-dictated defects at the Γ and K points can be observed. This coarsened structure was also obtained within the phase field crystal model in ref. [31]. Clearly, the decreased positional order for large system sizes due to the Landau-Peierls instability leads to a smearing of the director field which causes the coarsening of the defect structure. By applying a substrate potential amplitude of $V_0 = 0.1 k_B T$, we can suppress the Landau-Peierls instability normally incurring in two-dimensional crystals. That allows us to obtain the complex defect structure in fig. 3 also for large system sizes.

To understand qualitatively how the defect structure arises in our system we put now forward a simplified, yet topologically equivalent model. A single spherocylinder

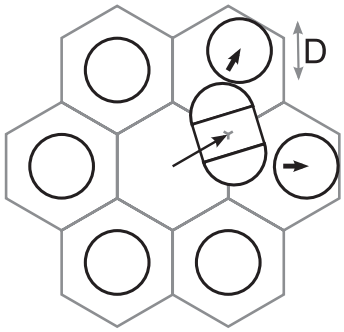

Fig. 5: In a simplified model, one spherocylinder of aspect ratio L/D is confined by its nearest neighbours on the hexagonal lattice, which are approximated as disks of diameter D. Two of the neighbouring disks have to be displaced as indicated to allow the spherocylinder to reach the Wigner-Seitz cell corner. (The particles and displacements are not drawn to scale.)

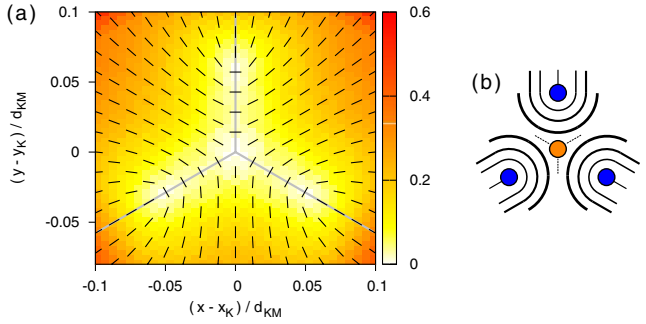

Fig. 6: (Color online) The director field obtained from the simplified model contains the same defect structure near the Wigner-Seitz cell corner. (a) Results from the calculation are qualitatively the same as the results from the many-particle Monte Carlo simulation in fig. 2. (b) A schematic representation of the observed defects. The x and y distances from the corner at (x_K, y_K) are measured in units of d_{KM}, the distance between the K and M points.

of aspect ratio L/D is confined in a cage of nearest neighbours approximated as disks of diameter D. To allow the spherocylinder to reach the corner of the unit cell, two of the neighbours have to be displaced from their lattice sites, as indicated in fig. 5. Such displacements are readily available due to the characteristic softness of the phonon in any plastic crystal at moderate density [47]. Subsequently, the director at each position near the corner of the Wigner-Seitz cell is determined by Monte Carlo integration over the directions of the spherocylinder. The resulting director field, which has been symmetrised using the rotation and mirror symmetries at the corner of the Wigner-Seitz cell, can be seen in fig. 6. It shows the same defect structure near the corner as the many-particle Monte Carlo simulation. This indicates that the director field near the K-point in the many-particle system is indeed created by a two-particle displacement similar to the one depicted in fig. 5. However, this is a qualitative model and not designed for complete agreement with the many-particle Monte Carlo simulation. The $+1/2$ defect is predicted to lie between K and M, but there is no quantitative agreement with figs. 2 and 4, since the positions of the defects do not coincide. Note also the difference in scale for the nematic order parameter compared to fig. 2. Additionally, the simplified model for the configuration of neighbours in fig. 5 can only provide information close to the Wigner-Seitz cell edge. Other configurations of neighbours are responsible for the director field at other points in the unit cell.

The analogy between the simple model and the many-particle system allows us to explain the observed defect structure. For most positions of the spherocylinder within the unit cell, its neighbours form a closed "front" at the packing fractions where the crystal is stable. Minimisation of the distance between the spherocylinder and this front of neighbours requires the spherocylinder to align parallel to the front. However, when the spherocylinder moves towards a corner of the unit cell it drives the two closest neighbours apart from each other, opening a gap in the front of neighbours. To insert itself into this gap, the spherocylinder must assume an orientation perpendicular to the front of neighbours. Accordingly, the spherocylinder changes its orientation when it approaches the corner. On the symmetry lines between Γ and K and between M and K the director has to be either perpendicular or parallel to the line in question. If it rotates, it has to perform a 90-degree rotation. The only topological defect types compatible with such a rotation are those with half-integer winding number. Thus, the charge $-1/2$ defect between Γ and K appears naturally and the charge $+1/2$ defect between M and K follows from the topological charge neutrality of the unit cell. The structure of fig. 3 is then the most obvious structure that both minimises bending in the director field and obeys the symmetries of a hexagonal plastic crystal.

Conclusions. – In conclusion, we have calculated the director field of a plastic crystal using the Monte Carlo simulation of two-dimensional hard spherocylinders exposed to an external substrate potential. A non-trivial topological defect structure of the director field was found. There are integer topological defects at the lattice position and charge $-1/2$ defects in the Wigner-Seitz cell corners, where the latter are surrounded by three $-1/2$ satellite defects. Finally, defects with topological charge $+1/2$ are found on the edges. We investigated the origin of these defects qualitatively within a simplified model. Basically they result from a pair displacement of the neighbouring particles, that allows the spherocylinder access to the corner.

The most promising set-up to verify our results both qualitatively and quantitatively is for colloidal suspensions confined to a quasi–two-dimensional substrate [5,10,40]. In an experimental real-space study, the director field can be obtained from the orientations and positions of the colloids using the methods described above.

Different aspects would be interesting for future work: first, three-dimensional plastic crystals should be addressed by computer simulations. In the 3d case, many more possibilities for defect structures arise [18] including defect lines. Since three-dimensional crystals exhibit true long-range positional order no substrate potential is required in that case. Second, it would be challenging to apply a microscopic theory like the density functional theory [11] to hard spherocylinders in the plastic-crystalline phase and to resolve the director field completely. The fundamental measure approach which has been recently constructed for hard anisotropic particles by Mecke and Hansen-Goos [48,49] is a promising theory since it has recently been successfully applied to hard spherocylinders [48,50,51]. Finally, a growing plastic crystal, say out of a disordered phase would provide an exciting set-up to see the internal topological defects emerge: it is unclear at the moment how the generation of defects is coupled to the development of positional order. The phase-field-crystal model [31,33] would be a good starting point to study these dynamical non-equilibrium questions.

* * *

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