Novel smectic phases and orientational order switching in two-dimensional liquid crystalline system

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ABSTRACT
Liquid crystalline (LC) materials are ubiquitous in our modern life. Most of the applications where LC materials find their use depend on maintaining the orientation of the constituent LC particles. In this article, we report a recipe to control the molecular orientation. We have done a computer simulation study of a two-dimensional (2D) liquid crystalline material consisting of soft ellipses. The pair interaction is modelled by using the Gay–Berne potential. The 2D LC material is set to interact with an underlying substrate which varies periodically in one-dimension (1D) only. By controlling the substrate parameters like periodicity and strength, we showed that a nematic phase LC material transitions into different novel phases. We finally show that by applying a substrate with large periodicity, the substrate strength can be used as a control parameter to steer the LC director.

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1. Introduction

Colloidal systems have received considerable attention as a model system to study various equilibrium phenomena. Different types of melting and ordering phenomenon appear when periodic substrate is applied to 2D colloidal systems. That is why 2D systems with an underlying periodicity have received much attention consistently in the last few decades [1–10]. In experiments where colloids are interacting with one-dimensional (1D) substrates, created using interfering laser beams, a novel laser-induced freezing was observed [11–17]. When the colloids are placed on an optically created one-dimensional periodic substrate (1DPS), several distinct crystalline and smectic ordering appear as a function of substrate strength and periodicity. Phenomena of freezing and melting have been studied both experimentally [18–22] and theoretically [23–29]. Such studies predicted a rich variety of phases, for example locked smectic, floating smectic, floating solids, and modulated liquids [30–39].

Depending upon the relationship between the particle-lattice parameters and the underlying substrate parameters, there arises the possibility of a commensurate-incommensurate phase transition phenomenon. The possibility and implications of such phase transitions are highly relevant to the surface and thin film condensed matter physics. A commensurate state occurs when specific length scales of the particle lattice match the periodicity of the underlying substrate [40]. In contrast, an incommensurate phase will appear otherwise. However, a commensurate-incommensurate phase transition may also depend on the substrate’s strength. For strong substrates, a chunk of the particles locks into a configuration commensurate with the substrate, while the remaining particles would form excitations such as vacancies or domain walls [41]. In two dimensions, the incommensurate phase is a floating phase without complete long-range order [42]. Particles maintain their intrinsic lattice structure at weak substrate potential and float on top of the substrate and the lattice ordering remains preserved at intermediate substrate strengths. However, at high substrate strength, there can be periodic distortions or rotations of the particle lattice with respect to the substrate lattice [43–47]. Systems that exhibit commensurate-incommensurate phases transition include vortices in type-II superconductors [37,41,48–52] and colloids [46,53–59]. As a consequence of substrate effects, a wide variety of systems also exhibit pattern formation as ordered stripes [60–64]. Such stripe morphologies appear in superconducting vortex systems, colloids [65–68], magnetic systems [69] and multi-layered superconductors [70]. Numerous studies have been done to understand how substrate–particle interactions can give rise to such patterns.

This article reports our study of a liquid crystalline (LC) system under the influence of an underlying substrate. Here, the LC system is a two-dimensional monolayer supported by a one-dimensional periodically varying substrate. We varied the substrate periodicity and strength and have observed a variety of interesting ordering of the particles and their dynamics. We modelled the LC system as a collection of soft ellipses with the Gay–Berne (GB) potential describing the interaction between a pair of ellipses. In one of our recent work, we have shown that with a substrate having moderate strength, such a system undergoes a pinning–depinning transition as the substrate periodicity is changed [71]. In the present work, we have observed a few novel LC phases, whose existence is owed not only to the anisotropic nature of the constituent particles and the role played by their orientational order but also to the substrate parameters. We also observed that the system’s dynamics undergo a substrate periodicity and strength-dependent transition from a dynamically locked state to a modulated-floating state. In addition, we also observed a phenomenon of orientation-switching of smectic-stripes at very high periodicity and strength. All these observations are made from the molecular dynamics simulations of the LC system in canonical ensemble conditions.

This paper is arranged as follows: In Section 2 we present the details of the pair potential for the LC material and the substrate. In Section 3 we describe the details of our numerical approach. In Section 4 we discuss and present the results of our simulations. Finally, in Section 5 we summarise the results with some future outlook on the problem.

2. Model system

2.1. Gay–Berne pair potential

The Gay–Berne (GB) potential is a generalisation of the Lennard-Jones (LJ) potential defining the interaction between anisotropic molecules. [72–76]. The generalisation is made by incorporating the positional and orientational dependence of the LJ potential for the anisotropic particles. Moreover, the orientational dependency is included in such a way that the potential has elliptical symmetry.

\[
U_{\text{GB}}(\mathbf{u}, \mathbf{u}_j, \mathbf{r}_j) = 4\epsilon(\mathbf{u}, \mathbf{u}_j, \mathbf{r}_j) \left[ \frac{\sigma_0}{(\mathbf{r}_j - \mathbf{u}_j) + \sigma_0} \right]^{12} - \left( \frac{\sigma_0}{(\mathbf{r}_j - \mathbf{u}_j) + \sigma_0} \right)^6. \tag{1}
\]
Here $r_{ij}$ is the vector displacement between the $i$th and $j$th particles; $\mathbf{u}_i$ and $\mathbf{u}_j$ are the unit vector representing the direction of the principal vector of $i$th and $j$th ellipses as described in Figure 1. The parameters $\sigma(\mathbf{u}_i, \mathbf{u}_j, \mathbf{r}_{ij})$ and $\epsilon(\mathbf{u}_i, \mathbf{u}_j, \mathbf{r}_{ij})$ respectively describe the range and depth of the GB potential [77–80]. The orientation-dependent length scale (range) parameter $\sigma(\mathbf{u}_i, \mathbf{u}_j, \mathbf{r}_{ij})$ is expressed as:

$$
\sigma(\mathbf{u}_i, \mathbf{u}_j, \mathbf{r}_{ij}) = \sigma_0 \left[ 1 - \chi \left\{ \frac{\left( (\mathbf{u}_i + \mathbf{u}_j) \cdot \mathbf{r}_{ij} \right)^2}{1 + \chi (\mathbf{u}_i \cdot \mathbf{u}_j)} + \frac{\left( (\mathbf{u}_i - \mathbf{u}_j) \cdot \mathbf{r}_{ij} \right)^2}{1 - \chi (\mathbf{u}_i \cdot \mathbf{u}_j)} \right\} \right]^{1/2}.
$$

(2)

Here $\chi$ is the shape anisotropy parameter of the potential and $\sigma_0$ defines the molecular diameter. The strength anisotropy function $\epsilon(\mathbf{u}_i, \mathbf{u}_j, \mathbf{r}_{ij})$ is defined by

$$
\epsilon(\mathbf{u}_i, \mathbf{u}_j, \mathbf{r}_{ij}) = \epsilon_0 \epsilon'(\mathbf{u}_i, \mathbf{u}_j) \epsilon''(\mathbf{u}_i, \mathbf{u}_j),
$$

(3)

with

$$
\epsilon(\mathbf{u}_i, \mathbf{u}_j) = (1 - \chi^2(\mathbf{u}_i, \mathbf{u}_j)^2)^{-1/2}
$$

(4)

and

$$
\epsilon'(\mathbf{u}_i, \mathbf{u}_j, \mathbf{r}_{ij}) = \left[ 1 - \chi \frac{\left( (\mathbf{u}_i + \mathbf{u}_j) \cdot \mathbf{r}_{ij} \right)^2}{1 + \chi (\mathbf{u}_i \cdot \mathbf{u}_j)} + \frac{\left( (\mathbf{u}_i - \mathbf{u}_j) \cdot \mathbf{r}_{ij} \right)^2}{1 - \chi (\mathbf{u}_i \cdot \mathbf{u}_j)} \right]^{1/2}.
$$

(5)

The interaction anisotropy parameter in Equation (5) is defined as $\chi' = \left( \kappa^{1/\mu} - 1 \right) / \left( \kappa^{1/\mu} + 1 \right)$. The parameter $\epsilon_0$ sets the energy scale of the potential. Thus, GB potential includes four main parameters ($\kappa, \kappa', \mu, \nu$), and they define a family of such GB potentials. Here, $\kappa$ defines the geometric aspect ratio of particles ($\sigma_j/\sigma_i$), and $\kappa'$ is the ratio of the potential well depths for the side-by-side and end-to-end configuration of a pair of interacting GB ellipses. Parameters $\nu$ and $\mu$ determine the effective strength of the potential. We have chosen the respective values of four parameters as $(3, 5, 2, 1)$ [81], as this is the most commonly chosen GB potential reported in the literature. Note that $\kappa = 3$ implies each GB ellipse is of size $3\sigma_0$ and $\sigma_0$ along with its major and minor axes, respectively.

### 2.2. Underlying substrate

The 1DPS with which the GB ellipses interact is as described below:

$$
V_{1D}(x, y) = \frac{A_1}{2} \left[ \sin \left( \frac{2\pi x}{a_s} \right) - 1 \right].
$$

(6)

Here, $(x, y)$ represents the position of the centre of mass of GB ellipses. The parameter $a_s$ represents the periodicity of the sinusoidal substrate potential, and the parameter $A_1$ represents the substrate strength, i.e. strength of the attractive interaction between the GB ellipses and the substrate. Note that the substrate–particle interaction depends only on the ellipses’ coordinate $(x, y)$, and there is no particle orientational dependency included in the substrate potential. Figure 2 is a cartoon representation of the system of GB ellipses interacting with the underlying substrate.

### 3. Simulation details

To simulate the GB ellipses with the underlying substrate, we have used the LAMMPS package [82–84] with GPU acceleration. Simulations are conducted with $N = 10,000$ particles contained in a rectangular box of size $A = L_x \times L_y$. We have chosen $L_y = \sqrt{3} L_x / 2$ to ensure that a system with hexagonal positional symmetry can be accommodated with the box. The periodic boundary condition is applied in both $x$ and $y$-directions. $L_x$ is so chosen that the substrate remains periodic in $x$-direction. For this, we have carefully chosen $L_x = n \times a_s$ with $n$ being an integer. While calculating the potential between a pair of GB ellipses, we have used a cut-off radius equal to $4\sigma_0$ to enhance the computational efficiency. We have varied substrate periodicity $a_s$ from 4.04 up to 16 and the substrate strength $A_1$ from 10 to 200. Our simulations begin with a nematic system having an orientational order. To achieve this, we have chosen the temperature and density to be $T = 0.8$ and $\rho = 0.3$, respectively. However, getting a good initial configuration at this relatively high density is not easy. Therefore, first, we took a highly diluted system at

![Figure 1.](image-url) (Colour online) Cartoon snapshot of the system of Gay-Berne ellipses with their directors, $\mathbf{u}_i$ as used in the Equation (1) to define the GB pair potential.
very low pressure and density. Then, we simulate the system as an \( NpT \)-ensemble and set the pressure \( p \) to get an initial configuration with desired density \( (\rho) \) and temperature \( (T) \). Generally, we wait for \( 1.5 \times 10^7 \) MD steps in the \( NpT \)-ensemble to reach a well-equilibrated system. Next, we switch the ensemble to \( NVT \). At this stage, the simulation is continued for another \( 2 \times 10^6 \) MD steps to equilibrate the system in the \( NVT \) ensemble condition. Next, we introduce the substrate to the 2D system. Extreme care needed to be taken while introducing the substrate, as the system might get frustrated if the substrate with a given strength is applied at once. So, once again, to get a well-equilibrated system with the underlying substrate, we ramp up the substrate strength \( A_1 \) very slowly, and for this, we followed the following steps: in the first step, we ramped the substrate strength from 0 to \( A_1 \) in \( 10^6 \) MD steps and then in the second step, we anneal the system by doubling the systems’ temperature \( (T) \) in 5 steps and then reducing the temperature in 5 steps. We also ramped the substrate strength simultaneously while increasing or decreasing the temperature \( (T) \). We waited for \( 2 \times 10^6 \) MD steps during each annealing step. After all these steps, finally, we further simulate the system for \( 2 \times 10^6 \) MD steps in \( NVT \)-ensemble with fixed substrate strength \( A_1 \) to get the final well-equilibrated system. To collect the statistics, the production run of the system was for \( 2 \times 10^6 \) MD steps, of which every \( 10^2 \)th microstate was saved for calculating the ensemble averages. As a final note, we have used the standard \( lj \)-unit system in our calculations, where the energy and length are scaled in the units of \( \varepsilon_0 \) and \( \sigma_0 \), respectively.

### 4. Results and discussion

The overarching objective of this work is to investigate the effects of the particle shape anisotropy of an LC material on its phase behaviour when the system is subjected to an underlying periodic substrate. As a nematic phase contains the orientation order (a material property borne out of particle shape anisotropy) but no positional order, it becomes natural to choose the system’s initial state as nematic. Thus, we hypothesise that LC materials’ orientational order will be essential in determining the system’s final configuration when a substrate is applied. To this end, we have set the temperature and density of the system as \( T = 0.8 \) and \( \rho = 0.3 \), respectively, to make sure the initial phase remains nematic. The configuration of the system (without substrate) is as shown in Figure 3(a). This system then acts as the initial state for all the cases as we vary the periodicity and strength of the underlying substrate. The snapshot (b) in the middle panel of Figure 3 shows a Voronoi diagram of the system. Each ellipse in (a) is now replaced by a solid dot with a colour that represents its number of nearest neighbours. The latter is determined from the Voronoi construction of the system. The detailed colour code is mentioned in the caption of Figure 3. The presence of particles having other than 6-fold hexagonal symmetry (i.e. particles with colours other than blue) indicates a positional defect. As this free system is in the nematic phase, the system is filled with particles having different local positional symmetries (mixing of different colours in Figure 3(b)). Further, to characterise the positional order of the system, we have calculated the static structure factor \( S(k) \), which is generally an excellent tool in studying the positional order in a system, especially when there are complex ordering is present. The \( S(k) \) is calculated using the following definition \([41,85–87]\):

\[
S(k) = \frac{1}{N} \left\langle \sum_{i=1}^{N} \sum_{j=1}^{N} e^{-i(k \cdot (r_i - r_j))} \right\rangle \\
= \frac{1}{N} \left\langle \sum_{i=1}^{N} \cos(k \cdot r_i) \right|^2 + \left\langle \sum_{i=1}^{N} \sin(k \cdot r_i) \right|^2 \right).
\]
Here \( \mathbf{k} \) represents the wave vector, and \( N \) the number of GB ellipses present in the system. The right panel (c) in Figure 3 is the projected map of \( S(\mathbf{k}) \) of the same free system as in Figure 3(a). Because the nematic system does not contain any structural order, hence the \( S(\mathbf{k}) \) in Figure 3(c) has only one bright spot at \( k = 0 \).

With our tools established as in Figure 3 for the study, in the following we discuss the results when the free nematic LC system is subjected to a substrate whose periodicity, \( a_s \), is varied as 4.04 \( \leq a_s \leq 16 \) and strength, \( A_1 \) as 10 \( \leq A_1 \leq 200 \). The thermodynamic parameters like temperature (\( T = 0.8 \)) and number density (\( \rho = 0.3 \)) are identical for all the systems considered henceforth. Please note that, in the following, some of the substrate periodicity values are not exactly integer but are very close. This is because our simulation recipe begins with the \( NpT \) ensemble and is followed by \( NVT \). In the \( NpT \) ensemble, the system’s volume fluctuates, so we cannot precisely control the system size. As a result, the substrate periodicity (applied during the \( NVT \) simulations) could not be matched exactly equal to integer values. We believe that these minor differences (with respect to the nearest integer values) in \( a_s \) would not essentially change the observations made in our report.

### 4.1. Substrate periodicity \( a_s = 4.04 \), and 4.97

We first consider the system with substrate periodicity \( a_s = 4.04 \), and the results are shown in Figure 4. At \( A_1 = 10 \), the system has a long-range orientational order as the ellipses are globally aligned. However, there is no positional order in the system; hence, the system is still in the nematic phase (Figure 4(a)). To illustrate the lack of positional order in the system, the Voronoi diagram is shown in Figure 4(b). The colour-coding in Figure 4(b) is the same as that in Figure 3(b). The first two images in Figure 3 (free system) and Figure 4 are almost identical, except that the nematic director of the system is now parallel to the horizontal \( (x\text{-axis}) \). The latter has happened because the underlying substrate forces the particles to remain aligned in vertical columns. However, the substrate strength \( A_1 = 10 \) is not yet high enough to change the system structurally; hence, the system remains in the nematic phase. This slight alignment of the ellipses in vertical columns shows up as few distinct sharp-bright spots along \( k_y = 0 \) line and around \( k = 0 \) point in the structure factor \( S(\mathbf{k}) \) (Figure 4(c)). In addition, for the system with \( (a_s = 4.04, A_1 = 10) \), there is a slight hallow around the \( k = 0 \) point that is more prominent along \( k_y \) (vertical) direction then in the \( k_x \) (horizontal) direction. This modification in \( S(\mathbf{k}) \) along \( k_y \) direction is an indication that ellipses are getting more ordered along \( y \)-direction and this also apparent from the real-space snapshot in Figure 4(a). Further comparison between the free system and system with the substrate \( (a_s = 4.04, A_1 = 10) \) is presented in the supplementary information (SI) sheet (SI Fig. S3).

As our next experiment, we increase the substrate strength to \( A_1 = 50 \), and we observe a dramatic change in the structural property of the system. The ellipses are aligned in vertical columns along the substrate minima and oriented horizontally (Figure 4(d)). However, interestingly, a few ellipses reside over the substrate maxima and are oriented vertically. Such a configuration of ellipses is a discovery for the liquid crystalline material. We may call such substrate modulated configurations as the crossed-smectic phase, where two types of layering are present: the majority of the ellipses are arranged in vertical layers (or columns) with their orientation
the given are perpendicular ($\theta = 90^\circ$), 3), 0 T. A particle under Figure 1.988 be of top. In fact, strength, remain very become quite counter-intuitive. In all cases, however, the substrate periodicity is kept the same as $a_z = 4.04$. All other thermodynamic parameters of the system are the same as that in Figure 3.

perpendicular to the layers, while the rest of the ellipses are also arranged in layers in between the other layers but with their orientation parallel to the layers. The presence of the minority ellipses along the substrate potential crests is quite counter-intuitive. Usually, ellipses would prefer to be at the substrate troughs to minimise the energy, but given the temperature ($T = 0.8$), density ($\rho = 0.3$), and the substrate strength ($A_1 = 50$) of the system, it may become energetically less favourable for the ellipses to be very close to each other and hence some ellipses may remain in the crests. When we increased the substrate strength, we indeed found that some of the ellipses, in fact, migrated to the troughs from the crests and populated the troughs more densely, as we discuss just below. In terms of the structure factor calculations (Figure 4(f)), the number of bright spots (peaks) has increased along the $k_y = 0$ line indicating strong layering of the particles along $x$-direction. In addition, there are now $S(k)$ peaks at non-zero $k_y$ values too, indicating the regular arrangement of ellipses in individual layers. However, the positions of the ellipses in different layers are not correlated; hence, the $S(k)$ peaks at higher non-zero $k_y$ values are smeared. These structural properties of this substrate-induced crossed-smectic phase get enhanced when we increase the substrate interaction strength to $A_1 = 200$ as shown in Figure 4(g–i). In terms of essential features, though, there is not much difference between the $A_1 = 50$ and $A_1 = 200$ cases, except that the number of ellipses that are orientationally in crossed position along the substrate crests has reduced as $A_1$ is increased from 50 to 200, and this is because some of the ellipses got accommodated in the trough layers as an increase in substrate strength made it energetically more favourable to be in troughs as we mentioned above.

Next, we consider the case when substrate periodicity is $a_z = 4.97$. Like the system with ($a_z = 4.97$, $a_s = 4.97$), here also, we find that the system with ($a_z = 4.97$, $a_s = 4.97$) remains in nematic state (data
4.2. Substrate periodicity \( a_s = 6, 6.93, 8.08, \) and 9

In Figure 6, we present the data for the systems with substrate periodicity \( a_s = 6 \), substrate strength \( A_1 = 50 \) (Figure 6(a)), and 200 (Figure 6(d)). The system transitions from substrate modulated crossed-smectic phase (Figure 4) to a double-layered smectic phase. In this phase, two smectic layers reside in a substrate trough without any cross-oriented layers between two neighbouring double layers (see SI Fig. S1). This is somewhat expected because the system’s substrate periodicity \( a_s = 6 \) is double the size aspect ratio of the GB ellipses (\( \kappa = 3 \)). At lower substrate strength (\( A_1 = 50 \)), the ellipses within each double-layered smectic column are loosely bound and continue to move horizontally (see SI movie M6). However, they remain restricted within the local substrate trough. At \( A_1 = 200 \), ellipses are more compact and exhibit high dynamic anisotropy. They are hardly mobile in the horizontal direction but very fast-moving in the vertical direction, i.e. along the substrate troughs (further discussions in Section 4.5). The particle’s mobility analysis of the systems indicates that the double-layers in neighbouring columns at \( A_1 = 50 \) are still coupled. In contrast, at \( A_1 = 200 \), double-layers are not coupled, and that is why they can move very fast along substrate troughs in vertical directions (up or down) independent of their neighbouring double-layer columns.

In terms of orientational order, there is a phase change between the systems \( A_1 = 50 \) and \( A_1 = 200 \). The double-layered smectic phase at \( A_1 = 200 \) has layer directors that are not perpendicular to the double-layers but make an angle less than 90°. This angle between the layer director and double-layer is also not fixed. Within a single column of double-layer, there are domains with different directors. For example, \( \text{PQ and RS} \) are the two directors of the two neighbouring double-layer domains and are oriented with an angle \( \theta \) and \( \pi - \theta \) respectively, as shown in Figure 6(d). Moreover, the director of the double-layer domains in two
neighbouring columns may or may not be parallel (see the directors of the different double-layers as indicated by the blue arrows attached to the horizontal red line in Figure 6(d)).

The structure factor calculations of the two systems are also indicative of their structural and dynamic dissimilarities (Figure 6(c,f)). The double layering of the ellipses in the systems shows up as the bright spots along the $k_y = 0$ line in $S(k)$, and these spots are less in numbers in $A_1 = 50$ (Figure 6(c)) than in $A_1 = 200$ (Figure 6(f)) as the double-layers are more ordered along $x$-direction in the latter case. The slightly smeared bright spots in Figure 6(f) at non zero $\pm k_y$ values are further indicative of the regular ordering of the ellipses in individual double layers in $y$-direction (for real space observations, see the SI movie M6).

As we increase the substrate periodicity further to $a_x = 6.93$, $8.08$, and $9$, we find that the system’s overall behaviour repeats the trend that we saw in the case of $a_x = 4.04$, $4.97$ and $6$. The corresponding real-space snapshots, Voronoi diagram, and structure factor are provided in the supplementary information sheet (SI Fig. S4 and S5). For the system with $a_x = 6.93$, we again see substrate modulated crossed-smectic phase at $A_1 = 50$, very similar to the system with $(a_x = 4.04, A_1 = 50)$ (Figure 4(d)), but now the smectic layers are double-layered (SI Fig. S4). The major change in structural properties of the system arrives when the substrate periodicity is set to $a_x = 9$. At $a_x = 9$ and $A_1 = 50$, the smectic layers become triple-layered (Figure 7(a)). This is again expected as the substrate periodicity is three times the size aspect ratio of the ellipses. However, not all smectic columns are triple-layered; some are still double-layered, surrounded by crossly oriented ellipses. In each column, there are both double- and triple-layered smectic domains. However, these multiply-layered smectic columns are not as ordered as it was in the case of $a_x = 6$ (Figure 6(a)), which is also apparent from the structure factor data shown in Figure 7(c). At $A_1 = 200$, the smectic layers are comparatively more ordered (Figure 7(d)). The corresponding structure factor plot has more bright spot along $k_y = 0$ line (positional order along $x$-direction) and also more bright spot, though smeared, at non zero $k$ values (Figure 7(f)).

4.3. Substrate periodicity $a_x = 16$

We have seen so far that for a given substrate strength ($A_1$) with an increase in substrate periodicity ($a_x$), the system transitions into different types of modulated smectic phases. However, note that such transition occurs only after a threshold $A_1$ and $a_x$ (Figure 4). The appearance of crossed-smectic, double-layered smectic, and triple-layered smectic phases as a function of $(a_x,A_1)$ deserves to ask how many such layering can effectively be reached and whether we may derive any functionality.
from such phenomena. To address this, we have investigated the systems with substrate periodicity $a_s = 16$ and substrate strength $A_1 = 50$, 200, 1000 and 2000. The corresponding results are presented in Figure 8. At $A_1 = 50$, ellipses form quintuple-layered 2D-solid like phase, where a quintuple-layered column is present at each substrate minima (Figure 8(a)). With $a_s = 16$, a quintuple-layered column is expected as $a_s$ is very close to the five times the particle size ratio. Each quintuple-layered column consists of loosely bound solid domains (blue dots with hexagonal symmetry in the corresponding Voronoi diagram of the system as shown in Figure 8(b)). The boundaries of the solid-like domains within a column are highly volatile, and in addition, ellipses are exchanged frequently between neighbouring columns as if each column is behaving like a grand canonical ensemble. Because of these two phenomena, there is no long-range positional order in the system, also evident from the structure factor calculations shown in Figure 8(c). Next, at $A_1 = 200$, the solid-like domains disappear, and each column transitions into a multiple-layered smectic like phase. Each column is now filled with smectic domains; most are quintuple-layered with director nearly parallel to horizontal, and the rest are variable-layered smectic domains. The actual number of layers in the latter type of domains depends on the direction of the corresponding domain director; the higher the angle between the domain-director and horizontal, the higher is the number of smectic-layer in a particular domain. In principle, when the director angle would be $90^\circ$, the numbers of smectic-layers should be infinity, i.e. the domain will span the entire column. In our simulations, we observed up to quindecuple-layered domains with director angle $\sim 110^\circ$ (or equivalently $70^\circ$) with the horizontal in the system with $A_1 = 200$ (Figure 8(d) and SI Fig. S2). The presence of the hallow of rings in the corresponding structure factor of the system (Figure 8(f)) indicates that the presence of large numbers of smectic domains oriented randomly in different columns destroys any long-range order in the system.

However, when we increased the substrate strength further to $A_1 = 1000$, almost all quintuple-layered smectic domains disappeared. Instead, all domains are now aligned diagonally (both forward and backward), and the average number of layers in a domain has increased, therefore the average domain size has also increased (Figure 8(g)). With the increase in $A_1$, the smectic domains get squeezed further towards the substrate minima (column centre); as a result, the domains rotate towards the substrate minima (y-axis), nearby domains merge, and the domain size, i.e. the number of smectic layers in a domain increases. In this process, the overall energy of the system gets minimised. An increase in domain size stabilises the system further, and consequently, in the corresponding structure factor of the system, bright spots appear (though smeared), and the rings get brighter (Figure 8(i)). Diagonal
alignment of the smectic domains in real-space gets translated into diagonally brighter rings in the structure factor.

As a trivial next step, we increased the substrate strength further to $A_1 = 2000$, and as a result, the average smectic-domain size increased further (Figure 8(j)). Some domains span a column almost entirely, and those smectic-layers in these domains are also nearly horizontal, i.e. domain directors are almost vertical (see SI Fig. S2). This is interesting because we now have a recipe to control the directionality of the smectic director in an LC system. In a 2D system of a given LC material with a particular aspect ratio of the constituent particles, we have two control parameters: the 1D substrate periodicity $a_s$ and substrate–particle interaction strength ($A_1$). By varying these two parameters suitably, we can switch the orientational order of an LC system. This would have applications in the surface science domain and LC display and sensor technology.
4.4. Commensurate phases

From the above analysis, we could identify that in some of the phases, the substrate periodicity and strength made it possible for the GB ellipses to get registered with the underlying substrate and form an ordered state. For example, when the periodicity \((a_s = 6\) or\(9\)) matches the ellipses’ size aspect ratio, the system transitions into a modulated multi-layered smectic phase. Such a phase can be regarded as a commensurate phase, and such transition will be an example of commensurate-incommensurate phase transition. The phases like the crossed-smectic phase arise because the substrate periodicity does not match the usual smectic-layer periodicity in the LC system, and hence the cross-oriented ellipses appear in between the multi-layered smectic stripes.

4.5. Mobility in modulated phases

In Section 4.2, we had a very brief mention of the mobility of the constituent GB ellipses in the system. In the following, we have presented some important observations that we have made in this study with different \(a_s\) and \(A_1\). In the upper panels of Figures 9 and 10, we have shown the time evolution of the displacement of the ellipses along \(y\)-direction, i.e. along the substrate minima, and in the lower panels, the real-space path traced by the ellipses is shown. There are 14 randomly chosen particles whose movement history is presented. Please refer to Figs. S6 to S14 in the SI sheets to find the mobility data (i.e. \(x(t), y(t)\), and the real-space path) for all the systems considered in our study. In terms of particle mobility, the most interesting finding is that at high substrate strength (e.g. \(A_1 = 200\)), when the substrate periodicity \((a_s)\) is an integer multiple of the particles’ size-anisotropy \((\kappa = 3)\), then the smectic-layers in modulated columns begin to move very fast along the substrate minima. For example, at \(a_s = 6\) and \(9\) (panel (e) in Figures 9 and 10), the particle mobility is an order magnitude higher than that at \(a_s = 4.04\), 6.93 or 8.08. However, not all smectic-columns move at the same speed (panel (f) in Figures 9 and 10). For a better understanding, please refer to the SI Movie M6 and M15. At lower substrate strength \((A_1 = 50)\), the smectic-layered columns are not entirely decoupled from neighbouring columns. Column decoupling also remains incomplete when the substrate periodicity \((a_s)\) is not an integer multiple of \(\kappa\), even at higher substrate strength \((A_1 = 200)\). Due to this incomplete decoupling of the neighbouring columns, the particles’ mobility is limited at \(a_s = 4.04, 4.97, 6.93\) and \(8.08\). Again, to observe this better, please refer to the SI Movie M1–16. Once the column decoupling is complete, as is the case when \(a_s = 6\) and \(9\) and \(A_1 = 200\), the system transitions into a substrate-induced modulated multilayered-smectic phase. In this phase, the system gets divided into several columns around the substrate minima, and each column consists of a multilayered smectic phase. Such

![Figure 9](image)

**Figure 9.** (Colour online) The mobility graphs of the GB ellipses under the influence of an underlying 1D periodic substrate with strength \(A_1 = 200\) and periodicities, \(a_s = 4.04, 4.97\) and \(6\). Upper panels: the time evolution of the displacement of the ellipses along \(y\)-direction, i.e., along the substrate minima. Lower panels: the real-space path traced by the ellipses in time. There are 14 randomly chosen particles whose movement history is presented here.
decoupled columns of LC material can move independently along the substrate minima without any influence from the neighbouring columns. The exact velocity (both direction and magnitude) of a particular smectic column along substrate minima would depend on the random fluctuation in the kinetic energy of ellipses when the substrate was applied.

5. Summary

We have done a computer simulation study of a liquid crystalline system interacting with an underlying one-dimensional substrate. We varied the substrate parameters like periodicity ($a_s$) and strength ($A_1$) and observed various novel smectic phases. The most exciting cases appear when the substrate strength is set to $A_1 = 200$, and the substrate periodicity is varied from $a_s = 4.04$ to 9. When the substrate periodicity is close to an integer multiple of the particle size ratio ($a_s = n \times \kappa$, $n$ being an integer), a modulated multilayered-smectic phase appears that is commensurate with the substrate. With other substrate periodicities (i.e. $n$ is a fraction), crossed smectic phases appear where two types of layers with orthogonal orientations exist. When we increased the substrate periodicity to about five times the particle size ratio, we found that the director of the multilayered-smectic columns reorients as a function of the substrate interaction strength. This indicates that for a 2D LC system, a one-dimensional substrate with large $a_s$, the $A_1$ can be used to control the orientation of a smectic phase. It would be interesting to see if our recipe to control in-situ the molecular orientation in LC material is realisable or not in an actual experimental setup.

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Disclosure statement

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Author’s contributions

DD conceptualised the work. Bharti performed the simulations and prepared the code for data analysis. Both DD and Bharti contributed towards the preparation of the manuscript.
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