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SI: Substrate induced freezing, melting and depinning transitions in two-dimensional liquid crystalline systems

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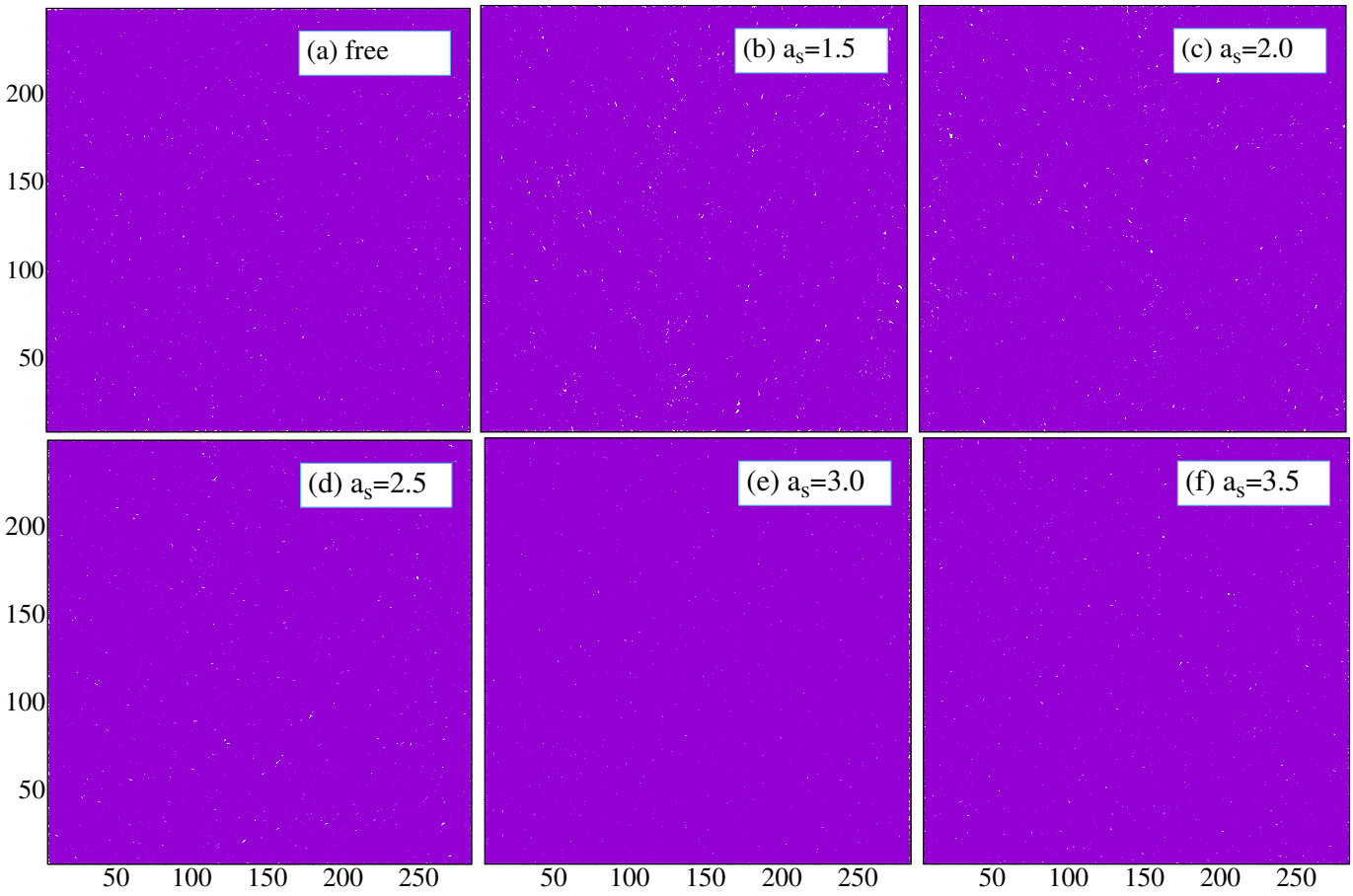
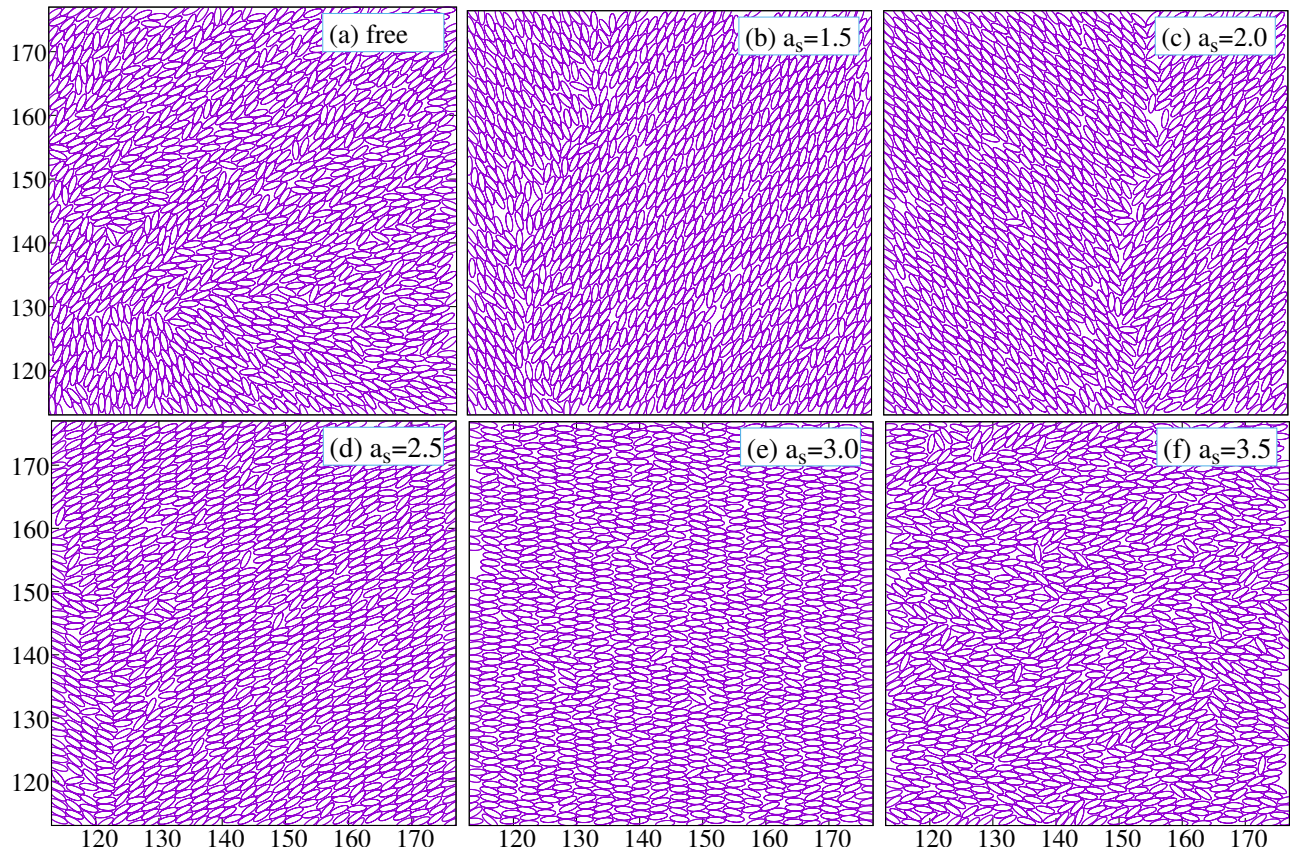
Abstract

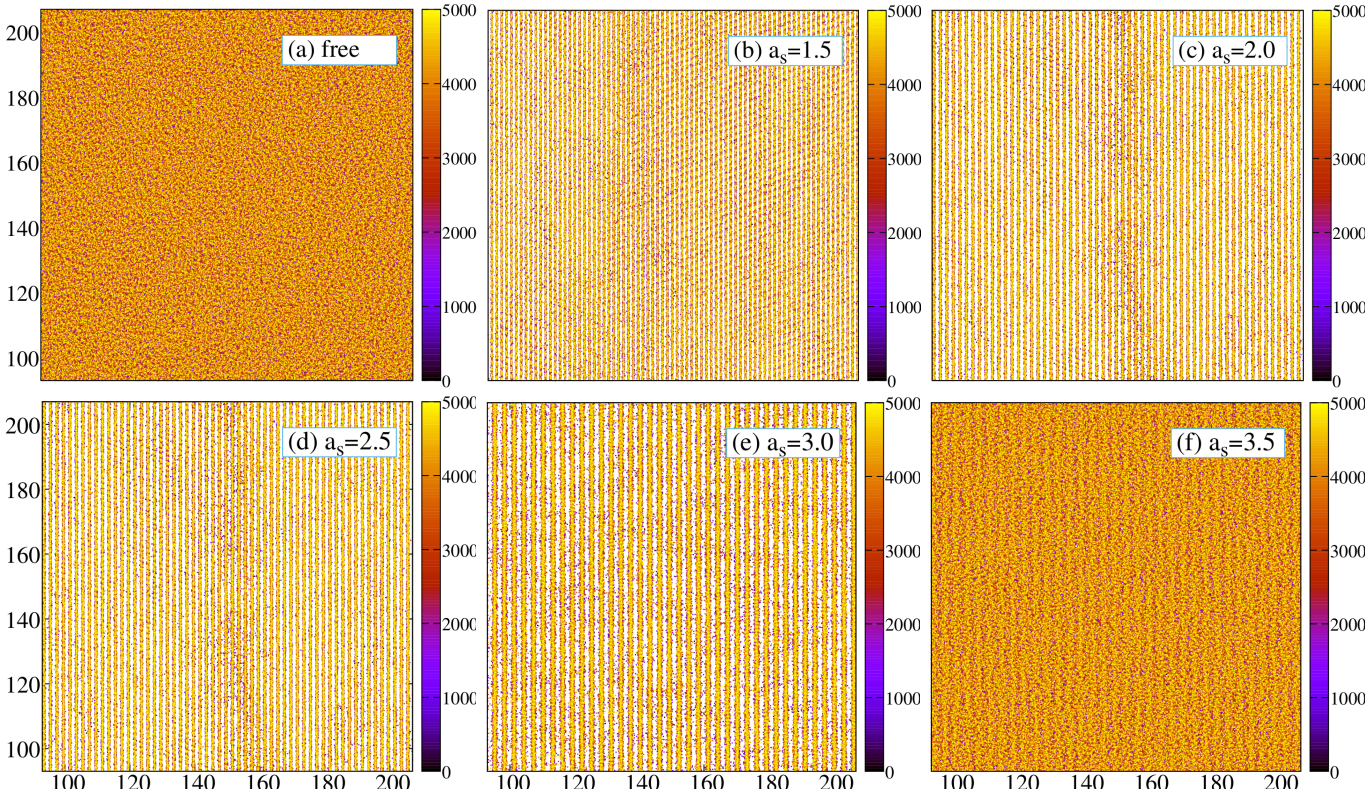
We use molecular dynamics simulations to investigate the ordering phenomena in two-dimensional (2D) liquid crystals over the one-dimensional periodic substrate (1DPS). We have used Gay-Berne (GB) potential to model the interaction between a pair of liquid crystalline (LC) particles. The underlying substrate potential with which the GB particles interact varies sinusoidally in one direction only. At a given temperature and density of the GB system, we varied the substrate's periodicity (a_s) but fixed the substrate strength. We observed that with a small value of a_s, an underlying substrate helps to stabilize a disordered LC nematic phase to a 2D solid phase. However, for an intermediate range of a_s, the system melts and transitions to a modulate-smectic. Finally, with a further increase in a_s, the system undergoes a structural depinning transition and returns to an LC nematic phase like a free system with no substrate. We argue that a three-way interplay of the energies arising from orientation-dependent particle-particle and particle-substrate interaction makes it possible for the system to undergo substrate-periodicity-dependent multiple phase transitions in the GB LC system.



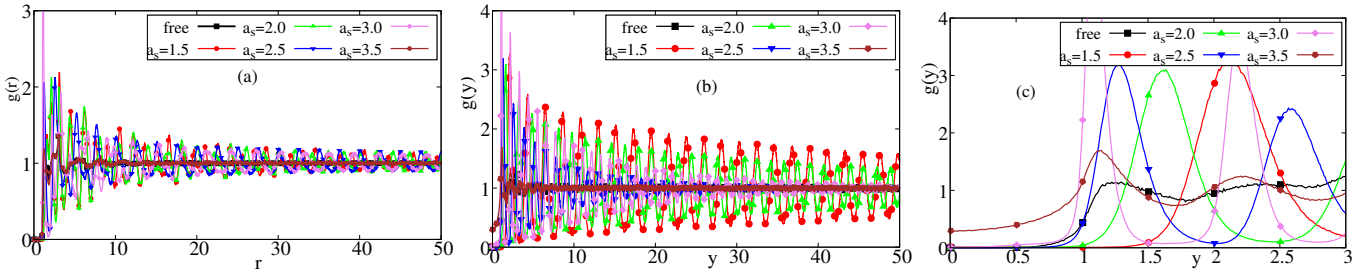
SI Fig. S1: The snapshots of GB liquid crystalline system at number density $\rho = 0.30$, $N = 22532$ and temperature $T = 0.8$. The system size is $L_x = \sqrt{3}L_y/2 = 294.0878$. The subfigure (a) is of a free system without any underlying substrate. Other snapshots are of systems with underlying substrate having different substrate periodicity, a_s as (b) $a_s = 1.5$, (c) $a_s = 2.0$, (d) $a_s = 2.5$, (e) $a_s = 3.0$, and (f) $a_s = 3.5$. A central part of this snapshots were shown in Fig. 1 in the main article.



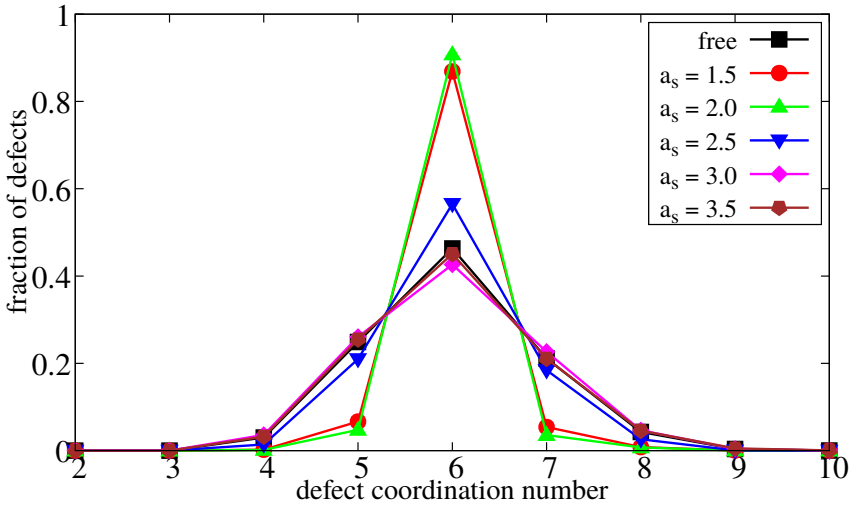
SI Fig. S2: Snapshot of GB ellipses on smaller scale at number density $\rho = 0.30$, and temperature $T = 0.8$. The system size is $L_x = \sqrt{3}L_y/2 = 294.0878$. (a) Snapshot for the free system (without the underlying substrate). Other snapshot shows the system with different substrate periodicity (a_s) as (b) $a_s = 1.5$, (c) $a_s = 2.0$, (d) $a_s = 2.5$, (e) $a_s = 3.0$, and (f) $a_s = 3.5$.



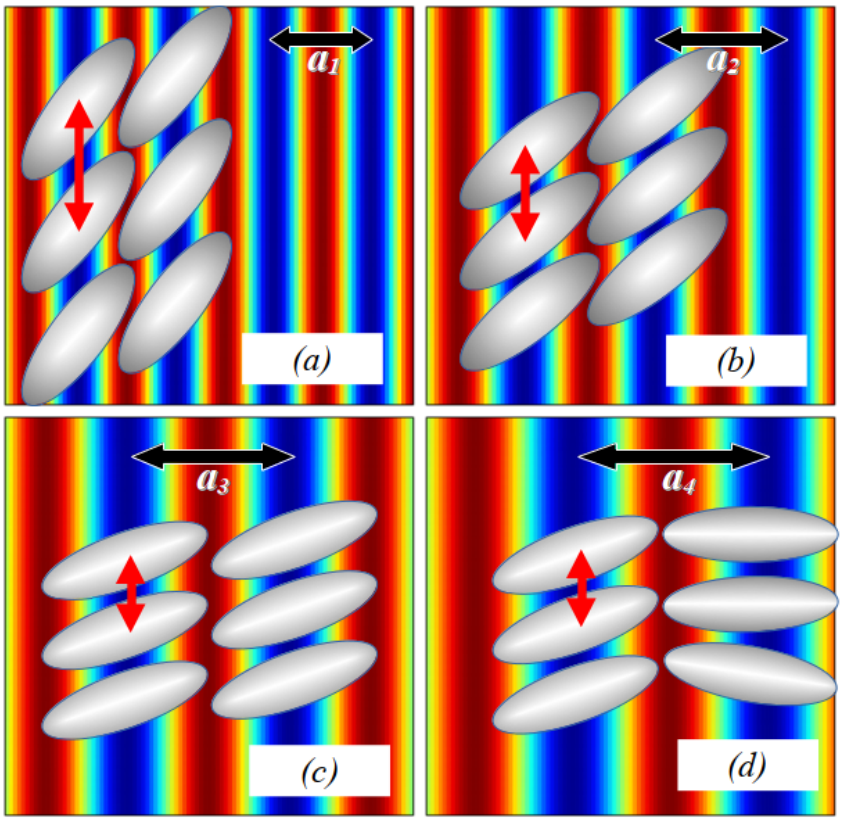
SI Fig. S3: Plots are showing the time evolution of the positions of the GB ellipses. Only the central part of the system is plotted for clarity. Here a total of 101 snapshots taken at a gap of 10000 over 10^6 MD steps are plotted together. Each GB ellipse is represented with a dot that is colored according to the time of the corresponding snapshot. The color bar attached next to the figures is showing the time range. Note that the MD step size $\Delta t = 0.005$ was taken during the simulations. Sub-figure (a) shows that particle positions are uniformly distributed in a (free) system without substrate. All the colors are mixed across the system, indicating a uniformly disordered state. However, as the systems are subjected to an underlying 1D sinusoidal (along x-direction) substrate, the particles arrange themselves in columns according to the position of the underlying substrate minima. Thus, their positions are restricted along the substrate minima (mixing of colors along the columns). When the underlying substrate has a periodicity $a_s = 3.5$, as shown in the sub-figure (f), the system's behavior is very similar to a disordered state as in sub-figure (a).



SI Fig. S4: Panel (a): The pair correlation function only along radial r-direction. Panel (b) and (c): The pair correlation function only along y-direction. From (b), it is clear that $g(y)$ is decaying algebraically in systems with $a_s = 1.5$ and 2 while in all other cases its decaying exponentially. The plots in (c) show only the first peaks of $g(y)$ for different systems. It can be seen that the first peaks for the disordered states at $a_s = 3.5$ and free system are very close to $y = 1$ as expected. However, for other systems, the first peak in $g(y)$ shifts towards $y = 1$ as the substrate periodicity increases. With $a_s = 1.5$, the nearest neighbor distance was the highest. This can be understood when we look at the corresponding snapshot in Fig. 1 (in the main article), which indicates that the GB ellipses are oriented at an angle with the horizontal. Hence, the nearest neighbor distance is longer than 1. As the a_s is increased, the ellipses reorient themselves more and more horizontal, and consequently, the minimum pair distance also decreases. The smaller the a_s, the substrate minimums are closer, hence the substrate restricts the particles within a narrower vertical column (y-direction), and the column themselves are also closer. This means that with smaller a_s, the GB ellipses have to orient themselves to place themselves along the narrowly spaced substrate minimums to minimize the substrate-particle interaction energy. Conversely, the higher the a_s, the further the minimums, and hence the GB ellipses have higher leverage in terms of their relative orientation, and hence they can position themselves further away to minimize the pair energy (GB end-to-end energy is lesser than the side-to-side).



SI Fig. S5: The plot is showing the defect distribution in different systems. The distributions are nearly identical for the *free* system and system with a substrate having periodicity $a_s \geq 3.0$. Systems with substrate periodicity a_s equal to 1.5 and 2.0 are highly stabilized as the fraction of 6-folded particles nearly equals 1, and very few defects (5, 7, 4, 8 folded particles) are present. However, as the substrate periodicity a_s nears 3.0, the fraction of 6-folded particles decreases dramatically, and the system depins and melts into a disordered state.



SI Fig. S6: A cartoon picture is showing the substrates with the possible configuration of some GB ellipses. The underlying color map is of the substrate, such that the dark red and dark blue representing the substrate maxima and minima respectively. The substrates have periodicity in increasing order as $a_1 < a_2 < a_3 < a_4$. The horizontal black double arrows indicate the substrate periodicity, increasing from (a) to (d). The vertical red double arrows indicate the separation between the ellipses along the vertical direction, decreasing as the substrate periodicity increases. In (a), the substrate minima are close, and as a result, ellipses have to be in an oblique orientation. In (b), substrate minima separation is increased compared to that in (a) as $a_2 > a_1$. Hence ellipses in (b) can reorient a little towards the horizontal. In (c) and (d), the vertical separations between ellipses are the same and are further less than that in (a) or (b). In (c) and (d), the substrate minima are enough separated that the ellipses in a particular column can reorient independently of the ellipses in the adjacent column. All these repositioning and reorientation of ellipses, as substrate periodicity is changed, is to achieve a minimum configuration in a competition between the ellipse-ellipse (side-to-side and end-to-end) interaction energy and ellipse-substrate interaction energy.