Smectic-glass transition in a liquid-crystal cell with a “dirty” substrate

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Abstract – We explore the smectic liquid-crystal order in a cell with a “dirty” substrate imposing random pinning. Within harmonic elasticity we find a subtle three-dimensional surface disorder-driven transition into a pinned smectic glass, controlled by a three-dimensional Cardy-Ostlund–like fixed line, akin to a super-rough phase of a two-dimensional $xy$ model. We compute the associated random substrate-driven distortions of the smectic-glass state, identify the characteristic length scales on the heterogeneous substrate and in the bulk, and discuss a variety of experimental signatures.

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Introduction. – Over the past several decades there has been considerable progress in understanding the phenomenology of ordered condensed-matter states subject to random heterogeneities, generically present in real materials [1]. More recently, attention turned to systems where the heterogeneity is confined to a surface [2–4], an important example of which is a liquid-crystal cell with a “dirty” substrate [5–8]. These surface-disordered systems are of considerable interest and exhibit phenomenology qualitatively distinct from their bulk-disordered counterparts.

The schlieren texture commonly observed in nematic cells [9] is a manifestation of such surface pinning in nematic cells. Recent experimental studies also include photo-alignment and dynamics in self-assembled liquid crystalline monolayers [10,11], as well as memory effects and multistability in the alignment of nematic cells with heterogeneous random-anchoring substrates [5]. The existence of the corresponding phenomena in smectic liquid crystals has recently been revealed in ferroelectric smectic-C cells in the book-shelf geometry [7,8]. This latter system was found to exhibit long-scale smectic-layer distortions, demonstrated to be driven by collective random surface pinning, and awaits a detailed description. Among many puzzling features observed in this system is the onset of broadening of the X-ray smectic peak as the temperature is reduced toward the smectic A-C transition.

A schematic of such a smectic liquid-crystal cell is illustrated in fig. 1, with a “dirty” front substrate imposing two types of surface disorders: surface heterogeneity that pins the nematic director $\hat{n}$ in random planar orientation (random orientational disorder), without a zenithal component, and the surface pinning that imposes a
quenched random potential on the layer position (random positional disorder). Generically such random surface pinning distorts the smectic layers and for sufficiently strong pinning can induce topological defects. As a first treatment of this problem, in this letter we focus on the simpler weak-disorder limit, where topological defects are absent or sufficiently dilute, so that a purely elastic description suffices.

In this letter we study the correlations of surface disorder-driven distortion of the smectic layers within a harmonic elastic description. To summarize our findings, we find that the conventional smectic order is always unstable in the presence of even infinitesimal random surface pinning. It is limited to a highly anisotropic finite domain (see fig. 1), characterized by Larkin-like length scales \( \xi \sim \xi_s^2 / \lambda \) (\( \lambda \) along the smectic-layer normal), with \( \lambda = \sqrt{K/B} \) a material-dependent length set by the bend and compressional elastic moduli \( K \) and \( B \) that is (away from the nematic-smectic transition) typically comparable to the layer thickness \( a \) [12]. On scales smaller than the domain size, the disorder effects are weak and can be treated perturbatively. In the plane of the substrate the pinning leads to power-law growth of the smectic-layer distortions and the associated short-range smectic order, that heal exponentially with the distance (beyond the domain size) from the substrate into the bulk.

To understand the behavior on longer scales where pinning is dominant, we employ the functional renormalization group (FRG) to account for random-potential nonlinearities [13]. We find a three-dimensional (3D) Cardy-Ostlund–like (CO) [14] phase transition at a nontrivial fixed line of disorder strength \( \Delta_s(T) \propto (T_g - T) \) for \( T < T_g \). The width of the X-ray scattering peak \( 1/\xi \), as given in eq. (18), is plotted as the top curve.

\[
H_{\text{pin}} = \frac{1}{\xi_s} \partial_z^{-1} [\Delta_f(T)]^\gamma
\]

\[
\Delta_f \propto \frac{K}{\delta} \partial_z^2 u^2 + \frac{B}{2} \partial_z u^2 + H_{\text{pin}},
\]

where \( u(x,y) \) is the distortion of the smectic layers at a point \( r = (x,y) \), and in 3D, \( x = (x,z) \) spans a 2D plane parallel to the substrate. Even though the nonlinear elasticity is known to be important in pure [16] and bulk disordered smectic systems [17], detailed analysis [15] shows that in this surface-disordered system, it is relevant below lower critical dimensions \( d_{\text{lc}}^g = 4 \) for the random orientational disorder and \( d_{\text{lc}}^g = 6 \) for the random positional disorder, but it is less relevant than the harmonic elasticity at distances below a length \( \xi_{NL} \) much larger

\[
\xi_s \sim \xi_{NL}^2 / \lambda
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\Delta_f \sim \xi_{NL}^2 / \lambda
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than the domain size \((\xi_{NL} \gg \xi_x)\). Thus, in the range of interest we can safely ignore the existence of nonlinearity. The pinning imposed by the surface disorder on the front \((y = 0)\) substrate is given by [15]

\[
H_{\text{pin}} = - \int d^{d-1}x \left[ (\hat{n} \cdot g(x))^2 + U(x)\rho(x) \right]
\approx - \int d^{d-1}x \left[ h(x) \partial_x u + V(u, x) \right],
\]

(4)

including the coupling between the nematic director \(\hat{n}\) and the local random pinning axis, \(g(x)\), as well as the coupling between the smectic density \(\rho(x)\) and the local random scalar potential \(U(x)\). These are determined by the substrate’s local chemical and physical structure (composition, roughness, rubbing, etc.). In the second line of eq. (4), we approximated this weak pinning by specializing to the smectic state, with the layer normal \(\hat{n}\) taken along the \(\hat{z} + \hat{\mathbf{n}}\) and \(\hat{\mathbf{n}} \approx -\nabla u\). This leads to \(h(x) \approx -2q_x(x)g_x(x)\) and \(V(u, x)\) a linear functional of the random potential \(U(x)\) (for more details see refs. [15,17]). \(h(x)\partial_x u\) captures the surface random orientational pinning of nematogens and smectic-layer normals, while \(V(u, x)\) encodes random pinning of smectic-layer positions, respectively associated with orientational and positional irregularities of an unrubbed substrate. Without loss of generality we take these random potentials to be characterized by zero-mean Gaussian distributions, with the variances

\[
h(x)h(x') = \Delta f \delta_{d-1}(x - x'),
\]

\[

\]

\[
V(u, x)V(u', x') = R_u(u - u')\delta_{d-1}(x - x'),
\]

where \(\delta_{d-1}(x)\) is a short-range function set by the scale on the order of the molecular size. Its precise form has no qualitative affect on the long scale (longer than its range) behavior which is our focus here [15].

Since the surface disorder is confined to the front substrate at \(y = 0\), there are no nonlinearities in the bulk \((y > 0)\) of the cell. Consequently, it is convenient to eliminate the bulk degrees of freedom \(u(x, y)\) in favor of the distortion on the heterogeneous substrate, \(u_0(x) \equiv u(x, y = 0)\). For \(T = 0\), we eliminate \(u(x, y)\) by solving the Euler-Lagrange equation

\[
K \nabla_y^4 u - \frac{\partial^2}{\partial y^2} f(x)\delta(y) = 0,
\]

with \(f(x)\) representing the boundary condition which imposes \(u_0(x)\) on the substrate, and obtain [15]

\[
u(q_x, q_y, y) = u_0(q_x, q_y)e^{-b q_x^2 + \frac{q_y^2}{2}\lambda} \frac{\sqrt{\lambda^2 q_x^4 + q_y^2 + \lambda q_y^2}}{\sqrt{\lambda^2 q_x^4 + q_y^2 - \lambda q_y^2}} \sin \left( y \sqrt{\lambda^2 q_x^4 + q_y^2 - \lambda q_y^2} \right) + \cos \left( y \sqrt{\lambda^2 q_x^4 + q_y^2 - \lambda q_y^2} \right).
\]

Substituting (7) into (3) and integrating \(y\) across a thick cell, \(0 \leq y < \infty\), the bulk energy reduces to the surface functional

\[
H_{\text{surface}}[u_0] = \int \frac{d^{d-2}q_x dq_y}{(2\pi)^{d-1}} \frac{1}{2} \Gamma_q |u_0(q_x, q_y)|^2 - \int d^{d-1}x \left[ h(x)\partial_x u_0 + V[u_0, x] \right],
\]

(8)

confined to the random substrate at \(y = 0\), with

\[
\Gamma_q = B\sqrt{2\lambda}\sqrt{\lambda^2 q_x^4 + q_y^2} \sqrt{\lambda^2 q_x^4 + q_y^2 + \lambda q_y^2}.
\]

\[
\]

\[
C_0(q) = C_T(q) + C_\Delta(q) = \frac{T}{\Gamma_q} + \frac{\Delta f q_x^2 + \Delta v}{\Gamma_q},
\]

(10)

where at long scales the first thermal contribution is subdominant to the random pinning.

The feature of the above-linearized (Larkin) approximation is that it predicts the range of its own validity, limited to small scales where the distortion \(u_0(x)\) remains small. Standard analysis in 3D gives \(\langle u_0(x) \rangle \approx \int \frac{\Delta f q_x^2 + \Delta v}{\Gamma_q} d^4q \), that we find to diverge with substrate extent \((L_x, L_z)\) for surface orientational disorder, \(\Delta f\), for \(d \leq d_c^f = 4\), and for surface positional disorder, \(\Delta v\), for \(d \leq d_c^v = 6\). Thus, below these lower critical dimensions \((e.g., for a physical 3D cell)\), an arbitrarily weak surface disorder destabilizes long-range smectic order.

We denote the scales at which these distortions grow to the order of the smectic period \(a\) as \(\xi_x\) and \(\xi_z\) [18], characterizing the size of the finite, highly anisotropic smectic domains. In the regimes of dominant orientational pinning we find

\[
\xi_f^c = c B^2 \lambda^3 a^2 \approx \sqrt{\lambda \xi_x^f}.
\]

For the dominant surface positional disorder we instead find

\[
\xi_v^c = \left( 3c \frac{B^2 \lambda^3 a^2}{\Delta_v} \right)^{1/3} \approx \sqrt{\lambda \xi_x^v},
\]

(12)

where \(c = \frac{4\pi^2}{\pi^2} \approx 34.6\). Utilizing the relation (7) between surface and bulk distortions indicates that they decay into the bulk exponentially \(\sim e^{-y/\xi_x}\).

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Physics beyond domain size. – On length scales longer than the domain size \( \xi_{x,z} \), the disorder imposed distortions \( v_0(\mathbf{x}) \) are large, invalidating the random force (Larkin) model, and requiring a nonperturbative treatment of surface random pinning in its fully nonlinear form (4). As with bulk disorder problems, this can be done systematically using an FRG treatment [13,17,19].

Employing the FRG analysis [15], we find that for a physically relevant 3D cell at finite \( T \), the problem significantly simplifies, allowing us to focus on the dominant lowest harmonic component of the variance function

\[
R_v(u) = \Delta_v \cos(\theta_0 u)/q_0^2, \tag{13}
\]

with \( q_0 = 2\pi/a \), and all higher harmonics irrelevant at long scales [15].

To treat this leading random pinning nonlinearity we employ the standard momentum shell RG [14,15,20], integrating out perturbatively (to one loop) in the surface disorder nonlinearities the short-scale modes with support in an infinitesimal momentum shell \( a^{-1} e^{-\delta k} < q_2 < a^{-1} \).

The result of this coarse-graining is summarized by the flow equations for the surface disorder strengths,

\[
\partial_t \hat{\Delta}_f = \hat{\Delta}_f + \pi^2 \Delta_v^2, \tag{14}
\]

\[
\partial_t \hat{\Delta}_v = \left( 3 - \frac{T q_0^2}{2 \pi B \lambda} \right) \hat{\Delta}_v - \hat{\Delta}_v^3, \tag{15}
\]

where \( \hat{\Delta}_f = 8\pi^4 a/\xi_f^4 \propto \Delta_f \) and \( \hat{\Delta}_v = 6\pi^2 (a/\xi_v)^3 \propto \Delta_v \) are dimensionless measures of the two types of disorders.

A straightforward analysis of eq. (15) shows that \( \hat{\Delta}_v(\ell) \) exhibits two qualitatively distinct long-scale behaviors. For \( T > T_g = 6\pi B \lambda/q_0^2 \) the effective positional pinning vanishes \( \Delta_v^* = 0 \), and for \( T < T_g \) it flows to a nontrivial fixed line \( \Delta_v^* = 3(1 - T/T_g) \), as illustrated in fig. 2. Then, for large \( \ell \) we replace \( \hat{\Delta}_f(\ell) \) inside (14) by its fixed point value \( \hat{\Delta}_f^* \) and solve the resulting equation for \( \hat{\Delta}_f(\ell) \), finding

\[
\hat{\Delta}_f(\ell) = \left[ \hat{\Delta}_f + \pi^2 (\hat{\Delta}_f^*)^2 \right] e^\ell - \pi^2 (\hat{\Delta}_f^*)^2. \tag{16}
\]

Although this predicts the same \( e^\ell \) asymptotics throughout, the prefactor depends on the long-scale value of the effective positional pinning \( \Delta_v^*(T) \), distinct in the two phases, \( \propto (T_g - T)^2 \) for \( T < T_g \) and vanishing for \( T > T_g \).

Thus we find that the surface-disordered smectic cell exhibits a 3D Cardy-Ostlund–like [14] phase transition at \( T_g = 6\pi B \lambda/q_0^2 \), between a high-temperature phase, where at long scales the surface positional pinning is smoothed away by thermal fluctuations, controlled by the orientational disorder and thermal fluctuations, and a low-temperature smectic-glass pinned phase, controlled by the nontrivial translational-pinning fixed line, \( \Delta_v^*(T) \). This transition to the surface-pinned state is analogous to the super-rough phase of a crystal surface grown on a random substrate [20], the vortex glass phase of flux line vortices confined to a plane in a type-II superconductor [21,22], and to the 3D smectic liquid crystals pinned by a random porous environment such as aerogel [17]. Similar to a number of bulk examples (the Cardy-Ostlund model [14,17,20,21] and the Kosterlitz-Thouless transitions), here too the phase transition at \( T_g \) does not display experimentally observable thermodynamic singularities of conventional phase transitions. It is thus detected through the change in the asymptotic behavior of the correlation functions.

To explore the physical distinctions between the two phases, we compute the smectic correlation function, utilizing the RG matching method to relate it at long scales (where disorder dominates over elasticity and must be treated nonperturbatively) to short scales, where perturbative analysis with effective, length-scale–dependent couplings suffices. We thereby find

\[
C(q) = \Delta_f \left[ 1 + \frac{\xi_f^2}{8\pi^2 a^2 (\Delta_v^*)^2} q_2^2 \right], \tag{17}
\]

We note that because of the additional factor of \( q_2 \), the direct contribution of the positional disorder (a consequence of the fixed line) is subdominant at long scales even for \( T < T_g \), where \( \Delta_v^* \) is relevant. Thus, remarkably, at long scales the smectic-layer distortions are controlled by the orientational pinning with the effective temperature-dependent strength, \( \Delta_f(T) = \Delta_f(T_g) \), but with scaling identical to that of the short-scale linearized model, as summarized by eqs. (1), (2).

Experimental predictions. – Such an asymptotically growing phonon correlation function translates into short-range correlated smectic order, with an anisotropic X-ray scattering peak of widths, \( 1/\xi_{x,z} \), and growing as \( 1/(k_z - q_0)^3 \) around \( q_0 \). The temperature-dependent onset of the enhanced orientational pinning for \( T < T_g \) leads to a nonanalytic dependence of the smectic peak width along \( k_z \),

\[
1/\xi_x(T) \sim 1/\xi_z^2 \sim \delta_f(T), \tag{18}
\]

as illustrated in fig. 2. We suggest that this prediction provides a plausible explanation for the precipitous X-ray peak broadening observed upon cooling of a smectic liquid-crystal cell with a random substrate [7,8]. Although the compressional modulus \( B(T) \) is also known to decrease on the approach to the smectic A-C transition [23], this variation is smooth away from \( T_{AC}, T_{NA} \) and can therefore be distinguished from the above onset behavior at \( T_g \). More systematic experimental and theoretical studies are necessary to explore this further [15].

Using eq. (7), together with a scaling analysis, we predict that the short-range smectic order persists arbitrarily deep into the bulk, but with the correlation length at depth \( y > \xi_z \) set by \( y \) itself. For weak surface pinning smectic-layer distortions are extended beyond the visible wavelength, validating the Mauguin limit, where linear light polarization “adiabatically” follows the local optic
axis. Under such conditions, transmission polarized light microscopy directly probes the smectic-layer tilt $\partial_x u_0(x, z)$ on the heterogeneous substrate [15], similar to a surface-disordered nematic cell [4], and can be used to test our predictions.

Summary. – In summary, we studied the stability and distortions of smectic order inside a thick liquid-crystal cell with a random heterogeneous substrate, but neglecting the possible role of disorder-induced dislocations and elastic nonlinearities that we believe is valid for weak pinning [15]. We found that even arbitrarily weak random surface pinning reduces smectic order to finite sizedomains. By considering the behavior on longer scales we predicted the existence of a 3D Cardy-Ostlund–like transition at $T_g$ from a weakly disordered smectic for $T > T_g$ (where on long scales the surface positional pinning is averaged away by thermal fluctuations), to a low-temperature disorder-dominated smectic glass, where pinning is enhanced below $T_g$. We leave a number of interesting and challenging questions to a future publication [15].

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