Smectic A herringbone patterns

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Abstract. Two qualitatively different SmA structures exhibiting herringbone-type layer patterns, to which we refer as the Defectless Smectic Herringbone (DSH) and the Dislocation Decorated Smectic Herringbone (DDSH) pattern are studied by a Landau-de Gennes-Ginzburg mesoscopic approach. Liquid crystal structures are described in terms of a nematic director field and a smectic complex order parameter. It is demonstrated that, in the proximity of the N-SmA phase transition, a melting of smectic layers could be realised even for relatively weakly-tilted smectic layers (i.e. \( \theta_t \approx 10^0 \)) for type I SmA phase. The width of melted region could be relatively large with respect to bulk values of the smectic characteristic lengths. In addition, a critical value of \( \theta_t \) is determined at which a DDSH pattern is expected to appear.

1. Introduction
Recent years have brought increasing interest in soft nanocomposites [1]. Typically, such systems consist of a soft matrix containing nanoparticles, where each component contributes a specific desired system’s property. In particular these materials have great potential to play important role in a wide variety of future tunable and multifunctional nanodevices [1,2].

Various liquid crystalline (LC) phases are often used as soft matrices [3,4]. Their main advantageous properties are softness, liquid character, optical transparency and anisotropy as well as an enormously large variety of different structures that they could exhibit. On the other hand, added nanoparticles (NPs) are used either to quantitatively anomalously enhance anisotropic LC properties [5] or to induce qualitatively new features [1].

Furthermore, LC structures could be exploited to trigger ordering [6,7], spatial localization and/or assembling [8,9,10,11,12,13] of immersed NPs within desired attracting regions. In those studies it was demonstrated that localized elastic distortions are very efficient attractors for appropriate (or adequately surface decorated) NPs. In view of potential applications it is of great interest to form different well controlled LC elastically distorted patterns as potential traps for NPs. In particular, well defined localisation of NPs forming linear nanowire-like structures could be achieved in elastically distorted LC structures exhibiting smectic translational ordering [11,12,13]. Recently, it was demonstrated [14] that the technique of scribing a substrate confining LC using the stylus of an atomic force microscope could be exploited to manipulate the smectic ordering in plan-parallel cells. Using this technique herringbone-type smectic layer structures were induced where the imposed bending of smectic layers efficiently enforced a localized melting of smectic ordering.
In this contribution we focus theoretically on structural properties of smectic herringbone-type patterns. We utilize a mesoscopic Landau-de Gennes-Ginzburg approach [3,15] in order to determine key parameters controlling the width of localized regions in which smectic ordering is melted. The paper is organized as follows. In Sec. 2 we introduce the model. The geometry of the problem and the parametrization used to describe smectic ordering are both introduced in Sec. 3. In Sec. 4 we numerically analyze the smectic patterns for cases without dislocations. The conditions at which dislocations could appear are estimated in Sec. 5. In the last section our results are summarized.

2. Model
The Landau-de Gennes-Ginzburg mesoscopic model [3,15] is used in standard terms of the nematic director field $\vec{n}$ and the complex smectic A (SmA) order parameter $\psi = \eta e^{i\phi}$. The unit pseudo-vector $\vec{n}$ (i.e. orientations $\pm \vec{n}$ are physically equivalent) points along local uniaxial orientational ordering. The translational order parameter $\eta$ measures the degree of the smectic layer ordering. The phase $\phi$ determines the position and orientation of smectic layers. The local smectic layer normal vector is given by $\vec{v} = \nabla \phi / |\nabla \phi|$. The smectic order parameter is related to the LC mass density spatial variation via the equation $\rho = \rho_0 (1 + \psi + \psi^*)$, where $\rho_0$ stands for the mass density in the nematic phase.

In terms of $\vec{n}$ and $\psi$ we express the free energy density [3,15] as $f = f_{n} + f_{c}^{(s)} + f_{c}^{(a)} + f_{c}^{(e)}$. The nematic elastic free energy is expressed as

$$f_{c}^{(e)} = K_1 \left( \nabla \cdot \vec{n} \right)^2 + K_2 \left( \vec{n} \cdot \nabla \times \vec{n} \right)^2 + \frac{K_3}{2} \left| \vec{n} \times \nabla \times \vec{n} \right|^2 ,$$

where the positive quantities $K_1, K_2$ and $K_3$ are the splay, twist and bend Frank nematic elastic constants, respectively. This contribution enforces a homogeneous alignment of $\vec{n}$ along a symmetry breaking direction.

The smectic condensation free energy density reads

$$f_{c}^{(s)} = \alpha_0 (T - T_c) \left| \rho \right|^2 + \frac{\beta}{2} \left| \psi \right|^2 .$$

Here $\alpha_0$ and $\beta$ are positive material constants and we consider the case of 2nd order bulk nematic-SmA phase transition at $T = T_c$. Below $T_c$ the condensation term imposes the bulk equilibrium value of the translational order parameter

$$\eta_{eq} = \sqrt{\frac{\alpha_0 (T_c - T)}{\beta}} .$$

while $\eta_{eq} (T > T_c) = 0$.

The smectic elastic term is given by

$$f_{c}^{(e)} = C_1 \left| \left( \vec{n} \cdot \nabla - i q_0 \right) \psi \right|^2 + C_\perp \left| \left( \vec{n} \times \nabla \right) \psi \right|^2 .$$

Here the wave vector $q_0 = 2\pi / d_0$ determines the bulk intrinsic smectic layer distance $d_0$ which is enforced by the positive smectic compressibility elastic constant $C_\perp$ [15,16]. On the other hand, the positive smectic bend elastic constant $C_\perp$ enforces alignment of $\vec{n}$ along the smectic layer normal [15,16].

The quantity $f_0$ includes the free energy density contributions of the remaining degrees of freedom.
This relatively simple free energy density expression is sufficient to describe key phenomena of our interest. In following calculations we neglect the LC elastic anisotropy. We set $K_1 = K_2 = K_3 = K$ and $C_6 = C_\perp = C$. Note that on approaching the SmA phase, by decreasing temperature in the nematic phase, the nematic twist and bend nematic elastic constant diverge as a result of the smectic pre-transitional fluctuations [3,15,16]. They enable the temporal formation of SmA-like domains (their characteristic size is approximated by the smectic order parameter correlation length), which resist to the nematic twist and bend deformations. However, in the SmA phase one can use “bare” values of $K_2$ and $K_3$ in our mesoscopic modelling, because the nematic twist and bend deformations are suppressed by the smectic constants, represented by $C$.

In addition to $d_0$, the important material dependent lengths of the systems are the smectic order parameter correlation length $\xi$ and the nematic penetration length $\lambda$ [3]. The quantity $\xi$ estimates the distance over which the smectic degree of order recovers if it is locally perturbed. On the other hand, $\lambda$ estimates the distance over which $\hat{n}$ roughly recovers its orientation along the smectic layer normal $\hat{v}$ if it is locally reoriented from this direction. Below $T_c$ in bulk their values are estimated by [3]

$$\xi = \frac{\xi_0}{\sqrt{|r|}},$$

$$\lambda = \frac{\lambda_0}{\sqrt{|r|}},$$

$$r = (T - T_c)/T_c$$

is the reduced temperature, $\xi_0 = \sqrt{C/\alpha_0 T_c}$, and $\lambda_0 = \sqrt{\beta C q_0}$. The temperature independent “smectic” Ginzburg parameter [3,14,15]

$$\kappa = \frac{\lambda}{\xi} = \frac{\lambda_0}{\xi_0},$$

is also playing an important role in our study. Namely, it distinguishes between the type I and type II SmA phase [15], corresponding roughly to materials characterized by $\kappa < 1/\sqrt{2}$ and $\kappa > 1/\sqrt{2}$, respectively. In the latter case dislocations are relatively more easily introduced into SmA patterns if a smectic layer frustration is enforced.

### 3. Parameterisation and Euler-Lagrange equations

For reasons of computational convenience we introduce the scaled order parameter $\psi = \eta / \eta_{eq} = \tilde{\eta} e^{i\theta}$, and the dimensionless free energy density $\tilde{f} = f / (\alpha_0 (T_c - T) \eta_{eq}^2)$. We henceforth discard the tildes and express $f$ in terms of the system’s characteristic lengths (which are in principle known) as

$$f = -\eta^2 + \eta^2 \frac{\lambda_0^2 \xi^2}{2} \left| \nabla \hat{n} \right|^2 + \xi^2 \left( (\hat{n} \cdot \nabla - i \nu_0) \psi \right)^2 + \left( (\hat{n} \times \nabla) \psi \right)^2$$

within the SmA phase temperature regime. Note that the temperature dependence is hidden in the smectic material dependent lengths $\lambda$ and $\xi$, see Eq.(5) and Eq.(6).

In our study we focus on LC ordering within the herringbone-type smectic structure [14] depicted in the figure 1 in the Cartesian coordinate system $(x,y,z)$. The symmetry of the problem allows us to restrict to the $(x,z)$ plane. Relatively far (i.e., $x \gg \lambda$) from the herringbone’s tip we set $\hat{n}$ to be tilted for a tilt angle $\theta$ with respect to the $z$–axis and the smectic layer distance $d$ equals the equilibrium smectic layer distance $d_0$, i.e. $d = d_0$. On the other hand, LC molecules within the tip (at $x=0$) are aligned along the $z$-
axis. Consequently, smectic layers tend to be locally dilated in this region. Namely, in absence of dislocations the smectic periodicity at \( x=0 \) is given by \( q = 2\pi / d = q_0 / \cos \theta_0 \). This smectic pattern roughly mimics the experimentally obtained herringbone-type patterns studied in [14].

In such LC configurations two qualitatively different equilibrium patterns could be formed, to which we refer as the Defectless Smectic Herringbone (DSH) pattern and the Dislocation Decorated Smectic Herringbone (DDSH) pattern. In the DSH pattern the smectic order parameter tends to be depressed at the herringbone tip and one dimensional \( \eta_1(x) \) spatial variation is expected. In the DDSH pattern edge dislocations are introduced in order to partially relive the layer dilatation-imposed strain at the tip giving rise to two-dimensional spatial variation in degree of smectic ordering (i.e. \( \eta = \eta(x,z) \)). The former and latter patterns are expected in type I and type II Sm\( A \) structures [15], respectively.

In this paper we focus on cases without dislocations. Consequently, it is convenient to parametrize the smectic phase factor as follows [16]

\[
\phi = q(z + u), \tag{9}
\]

where \( q = 2\pi / d \) with \( d = d_0 / \cos \theta_0 \) describes smectic layer dilatation at \( x=0 \) due to bending of smectic layers and \( u \) represents the smectic layer displacement field. Furthermore, we express the nematic director field as

\[
\hat{n} = (\sin \theta, 0, \cos \theta). \tag{10}
\]

In DSH patterns the symmetry of the problem suggests \( \theta = \theta(x) \) and \( u = u(x) \), therefore

\[
\begin{align*}
\phi &= -\eta^2 + \frac{\eta^2}{2} + \frac{q_0^2 \lambda^2 \xi^2}{2} \left( \frac{\partial \theta}{\partial x} \right)^2 + \\
&\quad + \xi^2 \left( \frac{\partial \eta}{\partial x} \right)^2 + \eta^2 \left( q \left( \sin \theta \frac{\partial u}{\partial x} + \cos \theta \right) - q_0 \right)^2 + q^2 \left( \cos \theta \frac{\partial u}{\partial x} - \sin \theta \right)^2
\end{align*}
\]

(11)

One can minimize \( f \) with respect to \( \frac{\partial u}{\partial x} \), yielding the relation

\[
\frac{\partial u}{\partial x} = \frac{q_0}{q} \sin \theta. \tag{12}
\]

With this in mind we finally obtain

\[
\begin{align*}
f &= -\eta^2 + \frac{\eta^2}{2} + \frac{q_0^2 \lambda^2 \xi^2}{2} \left( \frac{\partial \theta}{\partial x} \right)^2 + \\
&\quad + \xi^2 \left( \frac{\partial \eta}{\partial x} \right)^2 + \eta^2 \left( q - q_0 \cos \theta \right)^2.
\end{align*}
\]

(13)

The corresponding equilibrium Euler-Lagrange equations read

\[
\begin{align*}
\xi^2 \frac{\partial^2 \eta}{\partial x^2} &= -\eta + \eta^3 + \eta \left( \frac{q}{q_0} - \cos \theta \right)^2 d_0^2 \xi^2, \tag{14}
\end{align*}
\]

\[
\lambda^2 \frac{\partial^2 \theta}{\partial x^2} = 2\eta^2 \left( \frac{q}{q_0} - \cos \theta \right) \sin \theta. \tag{15}
\]
Due to the symmetry of the problem (see figure 1) the boundary conditions at \( x=0 \) read \( \frac{\partial \eta}{\partial x} = 0 \) and \( \theta = 0 \). At \( x=R \), where we set \( R > \max\{\lambda, \xi\} \), we impose \( \frac{\partial \eta}{\partial x} = 0 \) and \( \theta = \theta_i \). The latter conditions enforce an equilibrium smectic ordering at this distance. The equilibrium equations are solved numerically for a given set of parameters.

In our simulations in studying DSH patterns it is expected that \( \eta(x) \) exhibits a minimal value at \( x=0 \) and recovers its bulk degree of ordering roughly at the distance \( x = \xi \). On the other hand \( \theta(x) \) monotonously increases from \( \theta(x = 0) = 0 \) to the value \( \theta(x = R) = \theta_i \). One intuitively expects that on increasing \( x \) the condition \( \hat{n} \parallel (\sin \theta_i, 0, \cos \theta_i) \) would be realized at \( x = \lambda \). However, in our simulations \( \eta \) and \( \theta \) might recover the SmA bulk-like structural properties at different distances. Namely, \( \xi \) and \( \lambda \) are estimated for “ideal” reference conditions. In deriving the expression for \( \xi \) the SmA layer position and \( \hat{n} \) are undistorted. Similarly, in the expression for \( \lambda \), the SmA layers are assumed to be undistorted, i.e., they form an uniform stack of layers with thickness \( d = d_0 \) and \( \eta = \eta_0 \). In our cases there are deviations from these conditions. Furthermore, the smectic phase factor and director field are expected to respond on different length scales in ideal condition. Let us recall that \( \phi \) plays the role of “gauge” field [4] in the SmA phase which is known to respond to imposed frustrations on geometrically-imposed scales [4]. In contrast, \( \hat{n} \) is expected to recover from locally induced distortions on the distance \( \lambda \) [15]. However, in our configurations both fields are distorted and coupled in a complex way as indicated in Eq.(12). For this reason we introduce the effective smectic order parameter correlation length \( \xi_{\text{eff}} \) and \( \lambda_{\text{eff}} \) as the distances from \( x=0 \), where the conditions \( \eta_{\text{bulk}} - \eta(\xi_{\text{eff}}) = \frac{1}{2} \left( \eta_{\text{bulk}} - \eta(0) \right) \) and \( \theta(\lambda_{\text{eff}}) = \frac{1}{2} \theta_i \) are realized, respectively. \( \left( \xi / d_0 = \lambda / d_0 > 3 \right) \) the smectic melting becomes pronounced.

![Figure 1](image_url)

**Figure 1.** Schematic presentation of a herringbone-type SmA pattern in the Cartesian \((x,z)\) plane. Smectic layers are tilted for an angle \( \theta_i \) with respect to the z-axis. LC molecules are forced to be aligned along the local normal of a smectic layer. The tip of the structure is placed at \( x=0 \). At the tip \( \hat{n} \) is aligned parallel with the z-axis.
4. Defectless Smectic Herringbone pattern

We study first the DSH patterns. Of our special interest are conditions at which melting at the tip of a DSH pattern is essential. We consider both type I and type II SmA LCs where we vary $\theta_i$ as well as the temperature. We probe values $\theta_i = 10^\circ$ and $\theta_i = 20^\circ$ corresponding to the minimal and maximal tilt angle experimentally realized in DSH-type structures in the type I SmA studied in [14]. In our simulations a relatively low temperature deep in the SmA phase roughly corresponds to $\min \{\xi, \lambda\} = d_o$.

On approaching the 2nd order N-SmA phase transition both $\lambda$ and $\xi$ tend to diverge. In figure 2 we first plot the spatial dependences of $\theta / \theta_i$ (dashed lines) and $\eta / \eta_{eq}$ (full lines) for $\kappa = 1$, corresponding to the crossover regime between the SmA I and II behaviour. A relatively large tilt angle ($\theta_i = 20^\circ$) and consequently the strong elastic strain at the tip of DSH patterns are imposed. At a relatively low temperature ($\xi / d_o = \lambda / d_o = 1$) smectic melting at the tip is relatively weak. Furthermore, distortions in $\eta$ and $\theta$ recover on scales roughly given by $\xi$ and $\theta$ as expected. However, at higher temperatures ($\xi / d_o = \lambda / d_o > 3$) the smectic melting becomes pronounced.

![Figure 2. $\theta(x)$ (solid lines) and $\eta(x)$ (dashed lines) dependencies for different temperatures (thin lines: $\xi / d_o = 1$; thick lines: $\xi / d_o = 3$; very thick lines: $\xi / d_o = 5$). $\theta_i = 20^\circ$, $\kappa = 1$.](image)

In figures 3 we compare $\theta(x)$ and $\eta(x)$ responses in the type I and type II SmA phases at a relatively low temperature ($\min \{\xi, \lambda\} = 1$). For $\theta_i = 20^\circ$ and type I SmA (see figure 3a) the smectic order parameter is relatively reduced (for about 90% for $\kappa = 0.2$) at the tip. For the imposed lengths $\xi / d_o = 5$ and $\lambda / d_o = 1$ we obtain $\lambda_{eff} \sqapprox 5.5d_o$. On the other hand, only relatively weak departures from $\eta_{eq}$ are observed for $\kappa = 5$ (the imposed lengths $\xi / d_o = 1$ and $\lambda / d_o = 5$) where we observed $\xi_{eq} \sqapprox 12d_o$. 


Some further type I patterns (by imposing $\xi / d_o = 10$ and $\lambda / d_o = 1$) are shown in figure 3b for $\kappa = 0.1$. For $\theta_i = 20^\circ$ we obtain in this case a pronounced smectic melting at the tip and $\xi_{\text{eff}} \leq 20d_o$. On the contrary, for $\theta_i = 10^\circ$ the imposed strain is too weak to melt the smectic ordering at relatively low temperatures. In figure 4 we demonstrate the changes in $\eta(x)$ and $\theta(x)$ behavior on increasing temperature towards the N-SmA phase transition for $\theta_i = 10^\circ$ and $\kappa = 0.1$, where the initial configuration obtained for $\lambda / d_o = 1$ is shown in figure 3b. One sees that, on increasing $T$ (and consequently the value of $\lambda$), the degree of smectic ordering progressively decreases. A complete smectic melting at $x=0$ is reached for $\lambda \approx 5d_o$. Above this value of $\lambda$, the width of melted region, represented by $\xi_{\text{eff}}$, begins to apparently increase on increasing $T$. Therefore, our study reveals that, for the type I SmA, a total melting of smectic ordering can be realized at the tip of the herringbone-type structures even for relatively small values of the layer tilt (i.e., $\theta_i \leq 10^\circ$) that could be experimentally realized using AFM technique [14].

Figure 3. $\theta(x)$ (solid lines) and $\eta(x)$ (dashed lines) dependencies. (a) thin line: $\kappa = 5$, thick line $\kappa = 0.2$; $\theta_i = 20^\circ$. (b) thin line: $\theta_i = 10^\circ$, thick line $\theta_i = 20^\circ$; $\kappa = 0.1$. 
5. Dislocation Decorated Smectic Herringbone patterns

In the following we derive the critical condition for which edge dislocations [3,17] are introduced into samples in order to partially relax the imposed elastic strain at the tip of herringbone-type patterns. For this purpose we estimate the free energy costs of the competing DDSM and DSM patterns. The critical condition is estimated by the energy balance of these patterns.

The basic steps of this approximate derivation are as follows. We take into account that at the herringbone tip the smectic layers tend to be dilated. Namely, geometrical reasons enforce the layer distance \( d = d_0 / \cos \theta \) which is larger than the equilibrium spacing \( d_0 \). Consequently, the layer bending compressibility free energy density penalty at \( x=0 \) equals to [17]

\[
f_{\eta} \cap C (q - q_0)^2 .
\]  

This layer distortion roughly extends over the distance \( \lambda \) from the tip along the \( z \)-axis. Assuming a homogeneous structure over a distance \( h \) along the \( z \)-axis the resulting total elastic free energy penalty reads

\[
\Delta F_{\text{DSH}} \cap C (q - q_0)^2 \lambda h .
\]  

Next we assume that an edge dislocation appears within the distance \( h \). We assume that the presence of the dislocation totally relaxes the layer strain at the tip. Therefore, for \( d = d_0 \) at \( x=0 \) the distance within which one dislocation is inserted equals to

\[
h = \frac{d_0}{1 - \cos \theta} .
\]  

The relationship follows from the condition \( h(q_0 - q) = 2\pi \). Consequently, the compressibility penalty approximately vanishes at the tip area. However, the necessary topological presence of the edge dislocation introduces in the \( (x,z) \) plane a region of surface area \( 2\pi \xi^2 \) where the translational ordering is melted. The related total condensation penalty \( \Delta F_{\text{DDSH}} \) is approximated by

\[
\Delta F_{\text{DDSH}} \cap \left( \frac{\alpha(T - T_c)}{\beta} \right)^2 \frac{\xi^2}{\pi} .
\]  

From the condition \( \Delta F_{\text{DDSH}} = \Delta F_{\text{DSH}} \) an estimate is obtained for the critical tilt angle \( \theta^{(c)}_\lambda \) where the DDSH-type pattern could appear

\[
\theta^{(c)}_\lambda \cap \arccos \left( 1 - \frac{1}{q_0^2} \right).
\]  

In the limit \( \lambda \to \infty \) it follows \( \theta^{(c)}_\lambda = 0 \) because melting does not cost any energy at the phase transition.

6. Conclusions

We have studied theoretically and numerically herringbone-type patterns in the liquid-crystalline SmA phase. This study has been inspired by experimental work [14] in which surprisingly broad region of melted smectic order at the tip of herringbone-type structures was reported in type I SmA. By using the
atomic force microscope, nanopatterned substrates that enforce such structures were prepared. In that study the smectic Ginzburg parameter was estimated in a range between 0.1 and 0.2. The tilt angles, characterising the herringbone patterns, were ranged between \( \theta = 10^\circ \) and \( \theta = 20^\circ \). The width of the melted region was substantially larger than the relevant bulk penetration or the smectic order parameter correlation length. Recent studies reveal that such localised melted regions could efficiently trap appropriate nanoparticles [8,9,10,11,12,13]. Therefore, it is of interest to understand the key parameters controlling the existence and the width of localised melted regions in these patterns.

In our study we have used Landau-de Gennes Ginzburg mesoscopic approach [3] in standard terms of the nematic director field and the complex smectic order parameter. We used minimal model in effectively two-dimensional system exhibiting the 2nd order N-SmA phase transition to test the impact of material parameters and temperature on the width of the smectic melted region at the tip of herringbone patterns. We used single elastic constant approximations where the nematic and the smectic elasticities were approximated by the constants \( K \) and \( C \), respectively.

In studying the degree of smectic melting in Defectless Smectic Herringbone (DSH) patterns we have assumed one-dimensional spatial variations \( \eta(x) \) and \( \theta(x) \). We analyzed the impact of the material, geometrical properties (represented by \( \kappa \) and \( \theta_i \), respectively) and the temperature on LC patterns. Our simulations reveal that relatively wide melted regions, with respect to the predicted bulk values of the relevant material characteristic lengths, could be realized even for relatively small tilt angles \( (\theta \leq 10^\circ) \) for high enough temperatures in the type I SmA phase. Furthermore, we estimate conditions for which edge dislocations are expected to penetrate smectic patterns, giving rise to the Dislocation Decorated Smectic Herringbone (DDSH) pattern. In this derivation we assumed that main free energy penalties in DSH and DDSH patterns arise due to dilatation of smectic layers at the herringbone tip and the melting of smectic ordering within the cores of dislocations, respectively.

In near future we plan to study trapping of appropriate NPs to DSH and DDSH patterns both theoretically and experimentally. The former structures display planar trapping walls while the latter patterns consist of linear traps consisting of edge dislocations. We will calculate the mechanical forces on NPs immersed to different LC regions using the continuum approach presented in [17].

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