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PERIODIC FRÉEDERICKSZ TRANSITIONS IN POLYMER NEMATICS WITH HIGH ELASTIC ANISOTROPY

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Abstract The polymer nematic liquid crystals can exhibit very high elastic anisotropy, due to their molecular conformation. Usually the twist constant K_{22} can be much smaller (from ~ 0.1 to ~ 0.01 times) than the splay rigidity K_{11} , but also the contrary can happen. In these two cases an external field (either magnetic or electric), properly applied to a well aligned sample, is able to induce the formation of static stripes, due to a Fréedericksz mechanism. In fact, the energetic cost of a mixed deformation is more favorable than the cost of the pure distortion involving only the bigger rigidity, as requested by the usual aperiodic Fréedericksz effect. In the present paper, the critical conditions for the appearance of such kind of static stripes in polymer nematics are presented and discussed.

INTRODUCTION

In the last years, after the pioneering work made by Lonberg and Meyer¹, several authors investigated the static splay-stripes, which can be achieved in nematic liquid crystals subjected to external fields^{2,5} or to opposite boundary conditions⁶⁻⁸ or to both constraints⁹⁻¹⁰.

Moreover, the effect of the surface-like elastic constant K_{24} on the stripes threshold has been pointed out, in the case of hybrid aligned layers^{8,10,11}. In this paper, we deal with the case of polymer nematics, which can exhibit very high elastic anisotropy, due to their molecular conformation. In particular, i., the twist elastic constant K_{22} can be much smaller (from ~ 0.1 to ~ 0.01 times) than the splay elastic constant K_{11} , as for the poly- γ -benzyl-glutamate (PBG) in racemic mixture, dissolved in dioxane and methylene chloride¹²; and, in principle, also the contrary ($K_{11} \approx 0.1 + 0.01 K_{22}$) is expected to happen, ii., for instance in the case of side-chain polymer nematics without flexible spacer¹³, or in the case of combined polymer nematics.

We will treat the important case of splay geometry- undisturbed unidirectional

planar (P-) alignment of the nematic director \mathbf{n} , that is the unit vector parallel to the local average of the molecule long axis. The easy direction at the walls is assumed to be parallel to x-axis, with symmetrical anchoring, both for tilt (out-of-plane polar angle θ) and for twist (in-plane azimuth ϕ). Note that the undisturbed P-alignment exhibits $\theta = 0$, $\phi = 0$ everywhere in the polymer layer. A magnetic field \mathbf{H} normal to the cell walls (i.e. parallel to z-axis) is the destabilizing source in the case i., whereas in the case ii. the magnetic field inducing instability must be in-plane, and parallel to y-axis. This means that in the first situation, a usual aperiodic Fréedericksz transition with starting splay-distortion is expected, whereas in the second one the starting deformation should be pure twist. But if the bulk elastic anisotropy is large, the energetic cost of a mixed deformation of splay-twist may be less than the cost of a pure distortion implying only the greater rigidity. In the following, we present the model and discuss the conditions for the appearance of such a kind of static splay-stripes for the limits, i., $r \equiv K_{22}/K_{11} \rightarrow 0$ and, ii., $r \rightarrow +\infty$ in polymer nematic liquid crystals, pointing out in both cases the role played by the saddle-splay elastic constant K_{24} when the anchoring is weak.

THEORY

Let us consider a cell of thickness d , with wall at $z_0 = -d/2$, $z_1 = d/2$, properly treated in order to induce a P-alignment parallel to the x-axis, with anchoring strengths W_θ for tilt and W_ϕ for twist at both walls (symmetrical anchoring conditions), according to Rapini-Papoular description^{14,15}. The cell is filled by a convenient polymer nematic liquid crystal.

In the frame of the first order continuum theory¹⁶, if a periodic distortion provoked by a magnetic field is present, with period λ along y-axis, the bulk free energy F_b is given by:

$$F_b = d_x \int_0^\lambda dy \int_{-d/2}^{d/2} dz \left\{ \frac{1}{2} [K_{11}s^2 + K_{22}t^2 + K_{33}b^2] + f_H \right\} \quad (1)$$

where $\mathbf{s} \equiv \mathbf{n} \operatorname{div} \mathbf{n}$, $t \equiv \mathbf{n} \cdot \operatorname{rot} \mathbf{n}$, $\mathbf{b} \equiv \mathbf{n} \times \operatorname{rot} \mathbf{n}$ are the splay vector, the twist pseudo-scalar, and the bend vector, respectively, related to the corresponding elastic constants, and $\mathbf{n} = \mathbf{i} \cos \phi \cos \theta + \mathbf{j} \sin \phi \cos \theta + \mathbf{k} \sin \theta$.

The cell width in the x-direction is d_x and the stripes wavevector is parallel to the y-axis, with the spatial period λ . The free energy density f_H is connected to the

coupling between the nematic director and the external field. We stress the fact that the analogy between the effect of a magnetic field \mathbf{H} on a nematic with positive susceptibility anisotropy χ_a , and the effect of an electric field \mathbf{E} on a non-conducting nematic with positive permittivity anisotropy ϵ_a is rather poor¹⁷. Actually, it is necessary to include in the second case the flexoelectric coupling^{18,19} and to consider the back-field contribution, which renders the interaction a non local one²⁰. Here we consider only the magnetic coupling: hence $f_H = -\frac{1}{2}\chi_a(\mathbf{n} \cdot \mathbf{H})^2$.

The surface free energy is dependent on the anchoring and on the saddle-splay:

$$F_s = d_x \int_0^\lambda dy [f_w - (K_{22} + K_{24})(\mathbf{s} + \mathbf{b}) \cdot \mathbf{u}_s] \quad (2)$$

\mathbf{u}_s being the normal unit vector at the boundary, according to Gauss' theorem (i.e., $\mathbf{u}_s = -\mathbf{k}$ for $z_0 = -d/2$, $\mathbf{u}_s = \mathbf{k}$ for $z_1 = d/2$).

The anchoring free energy areal density f_w reads in covariant form²²

$$f_w = -a(\mathbf{n} \cdot \mathbf{i})^2 + b(\mathbf{n} \cdot \mathbf{k})^2 \quad (3)$$

at the boundary, with $\{a, b\} > 0$, the anchoring strengths for tilt and for twist being $W_\theta = 2(a + b)$, $W_\phi = 2a$, respectively.

If the magnetic field is lower than the threshold H_p for the periodic Fréedericksz transition, the nematic remains undistorted in the P-configuration: instead, the periodic pattern takes place, if $H_p < H_a$, where H_a is the threshold for the usual aperiodic Fréedericksz transition. We will show that this actually happens, and, furthermore, that surprisingly $H_p \rightarrow 0$ for the bulk elastic ratio $r \rightarrow 0$, describing an intrinsic instability of the P-alignment for polymer nematics with very high elastic anisotropy.

Anyway, for finding the correct solution of the physical problem, we keep in mind that every system moves, in the space of the virtual configurations, towards its equilibrium state, characterized by a minimum of the total free energy. Moreover, in the equilibrium state there is a complete balancing of all torques acting on the system, both in the bulk and at the surface. The first balancing is described by the Euler-Lagrange equations, the latter one by the boundary conditions.

We consider a situation of small distortion, with $H > H_p$ but really close to H_p ; this means that it is possible to linearize the distortion torques with respect to both θ and ϕ , thus expanding the free energy density to the second order.

Notice that at this point the bend contribution vanish, since it is of the 4-th order in θ , ϕ , and their first derivatives: hence, the bulk linearized free energy reads⁴:

$$F_b = \frac{1}{2} d_x \int_0^\lambda dy \int_{-d/2}^{d/2} dz \{K_{11}(\phi_y + \theta_z)^2 + K_{22}(\theta_y - \phi_z)^2 - \chi_a H^2 \theta^2\} \quad (4)$$

while the surface linearized free energy at the (i-1)-th substrate (i=0 for $z = z_0$, i=1 for $z = z_1$) is given by:

$$F_{si} = \frac{1}{2} d_x \int_0^\lambda dy [W_\theta \theta_i^2 + W_\phi \phi_i^2 + 2(-1)^i (K_{22} + K_{24})(\theta_i \phi_{yi} - \phi_i \theta_{yi})] \quad (5)$$

In (4) and (5) the subscript y, z means first derivative with respect to the indicated variable.

We stress the fact that: i. the saddle-splay K_{24} only affects the surface condition, and thus only the boundary conditions²³; ii. the saddle-splay distortion is only dependent on the derivative with respect to a direction (y-axis) parallel to the surface, thus giving no troubles from the point of view of the variational calculus, as demonstrated for the first time in ref. 16 and recently recognized by Pergamenschchik²⁴.

i. Bulk elastic ratio $r \rightarrow 0$

i.1. $K_{22} \rightarrow 0$, $K_{11} \neq 0$ and finite

Introducing the reduced free energy and the relevant free energy density, defined as $G = 2F/(K_{11}d_x)$ and $g = 2f/(K_{11}d_x)$ respectively, eq.(4) writes:

$$G_b = \int_0^\lambda dy \int_{-d/2}^{d/2} g_b dz \quad (6)$$

$$g_b = (\phi_y + \theta_z)^2 - h^2 \theta^2$$

where the reduced field h is given by $h = (\chi_a/K_{11})^{1/2}H$, and has the meaning of the inverse magnetic coherence length. In eq.(6) is shown, that only splay- and magnetic contributions survive in the bulk free energy.

Let us define the extrapolation lengths for tilt and for twist, according to deGennes-Kléman^{25,26} as $L_\theta = K_{11}/W_\theta$, $L_\phi = K_{11}/W_\phi$ - note that $L_\phi = K_{22}/W_\phi$ is useless, since it vanish for every value of W_ϕ except for the case of very weak anchoring $W_\phi \rightarrow 0$. Eq.(5) reads:

$$G_s = \int_0^\lambda f_s dy \quad (7)$$

$$f_s = L_\theta^{-1} \theta_i^2 + L_\phi^{-1} \phi_i^2 + 2(-1)^i \kappa (\theta \phi_y - \phi \theta_y)$$

where f_{si} is the surface free energy, referred to a unit area of the boundary ($i=0,1$ for $z=z_0, z_1$), and $\kappa \equiv K_{24}/K_{11}$.

The common procedure of minimization of the generalized total functional $G_{tot} \equiv G_b + G_s$ provides the two Euler-Lagrange equations:

$$\begin{aligned}\theta_{zz} + \phi_{yz} + h^2\theta &= 0 \\ \phi_{yy} + \theta_{yz} &= 0\end{aligned}\quad (8)$$

the first equation giving the balancing in the bulk of the torques polar component, the second one the balancing of the torques azimuthal component. Note that the magnetic contribution affects only the polar balancing, and that the equations are coupled.

Since the boundary conditions are symmetrical, it is enough to consider the substrate $z_0 = -d/2$. Hence the boundary conditions are derived as

$$\begin{aligned}(1 - 2\kappa)\phi_{y_0} + \theta_{z_0} - L_\theta^{-1}\theta_0 &= 0 \\ 2\kappa\theta_{y_0} - L_\phi^{-1}\phi_0 &= 0\end{aligned}\quad (9)$$

The algebraic eqs.(9) give also the balancing of the torques, at the surface $z=z_0$, referred to the substrate unit area. Note that in the polar balancing (first equation) the tilt- and twist- elastic torques are equilibrated by the tilt- anchoring and by the saddle- torques whereas in the azimuthal equation (second one), only the twist-anchoring and the saddle- torques are in competition.

Aperiodic solution

The usual Fréedericksz transition is recovered imposing $\phi = 0$ everywhere and $\theta = \theta(z)$. Hence (8) becomes simply

$$\theta_{zz} + h^2\theta = 0 \quad (8')$$

with the boundary condition

$$\theta_{z_0} - L_\theta^{-1}\theta_0 = 0 \quad (9')$$

where the saddle- splay and the twist- anchoring are uninfluent, as expected.

Then the solution is obtained as

$$\theta = c_1 \cos hz \quad (10)$$

which, put into eq.(9'), gives the generalized Rapini-Papoular equation, already found in ref. 28:

$$hL_\theta = \cot(hd/2) \quad (11)$$

The value $h = h_a$ satisfying eq.(11) provides the threshold for the usual Fréedericksz transition. It is noticeable that the implicit function $h_a(L_0)$ is monotonically decreasing, reaching the asymptote $h_a = 0$ for $L_0 \rightarrow \infty$. Hence the maximum of $h_a(L_0)$ is attained for strong anchoring $L_0 = 0$ and is given by $h_a = \pi/d$, as expected²⁵.

Periodic solution

In the case of periodic Fréedericksz transition the starting slope of the distortion has to be searched by imposing

$$\begin{pmatrix} \theta \\ \phi \end{pmatrix} = \begin{pmatrix} \Theta \\ \Phi \end{pmatrix} \exp[i(\alpha z + \beta y)] \quad (12)$$

and by substituting (12) into Euler - Lagrange eqs.(8), one immediately obtains $h = 0$, no matter what values are assumed by β , $\alpha(\beta)$. This actually indicates that the threshold h_p for the appearance of the saddle-stripes vanishes: but, in order to obtain this point, it is necessary to consider the bulk torques equations for r small but not equal to zero⁴. Following such a procedure, we get

$$\alpha^2 = \frac{h^2}{2} - \beta^2 \pm \frac{1}{2} \left[h^4 + 4 \frac{1-r}{r} \beta^2 h^2 \right]^{1/2} \quad (13)$$

for $h > h_p$ and close to h_p . If the hypothesis: $[h_p \rightarrow 0 \text{ when } r \rightarrow 0]$ does work self-consistently, eq.(13) shall provide

$$\alpha^2 = -\beta^2 \pm \beta h_p / \sqrt{r} \quad (14)$$

with

$$h_p / \sqrt{r} = C > \beta \quad (15)$$

C being a convenient positive constant, to be found numerically.

If (14) and (15) are true, then the solutions for θ and ϕ , shifted each other by $\pi/2$, are obtained as:

$$\begin{aligned} \theta &= (A \cos kz + B \cosh kz) \cos \beta y \\ \phi &= (a \sin kz + b \sinh qz) \sin \beta y \end{aligned} \quad (16)$$

since for a given value of β four values of α are derived, and precisely

$$\begin{aligned} \alpha &= \pm \sqrt{(C - \beta)\beta} \equiv \pm k \\ \alpha &= \pm i \sqrt{(C + \beta)\beta} \equiv \pm iq \end{aligned} \quad (17)$$

with $q > k$ and $q > \beta$.

It is interesting to note that the same solutions (16) are derived for $r \neq 0$: in this case $\{k, q\}$ are also dependent on the reduced field h - see eq.(13). By putting (16) into the Euler - Lagrange eqs. valid in the case $r \neq 0$ ⁴, we found the conditions of self-consistency:

$$\begin{aligned} R &\equiv \frac{a}{A} = \frac{(1-r)k\beta}{\beta^2 + rk^2} \\ T &\equiv \frac{b}{B} = \frac{(1-r)q\beta}{-\beta^2 + rq^2} \end{aligned} \quad (18)$$

which, in the limit $r \rightarrow 0$, provide

$$\begin{aligned} R &= \frac{k}{\beta} \\ T &= -\frac{q}{\beta} \end{aligned} \quad (19)$$

This means that in eqs.(16) only two integration constants are independent. By putting (16) with the constraints (19) into the boundary conditions (12), a homogeneous system is deduced, giving no trivial solutions for the two integration constants, if the following dispersion relation is satisfied:

$$D \equiv \beta \sin \frac{kd}{2} \sinh \frac{qd}{2} \begin{vmatrix} l_\theta k - \cot \frac{kq}{2} & -l_\theta q - \coth \frac{qd}{2} \\ l_\phi \beta \cot \frac{kd}{2} - \frac{k}{\beta} & l_\phi \beta \coth \frac{qd}{2} + \frac{q}{\beta} \end{vmatrix} = 0 \quad (20)$$

with $l_\theta \equiv 2\kappa L_\theta$, $l_\phi \equiv 2\kappa L_\phi$.

Eq. (20) provides implicitly the wavevector constant C - and hence k and q - just above the threshold for the periodic pattern vs. β , l_θ , l_ϕ .

The coefficient determinant D vanish if either

$$\cot \frac{kd}{2} \tanh \frac{qd}{2} = -\frac{k}{q} \quad (21)$$

or

$$\beta = \sqrt{W_\theta W_\phi} / 2K_{24} \quad (22)$$

Eq. (21) implicitly provides the normalized field $C \equiv h/\sqrt{r}$ as a function of the wavenumber β : the minimum C_m of such a function gives the threshold value β_p for the principal mode of the periodic pattern - in our case h_p goes to zero as well as \sqrt{r} -, and consequently the threshold value k_p and q_p (see fig.1).

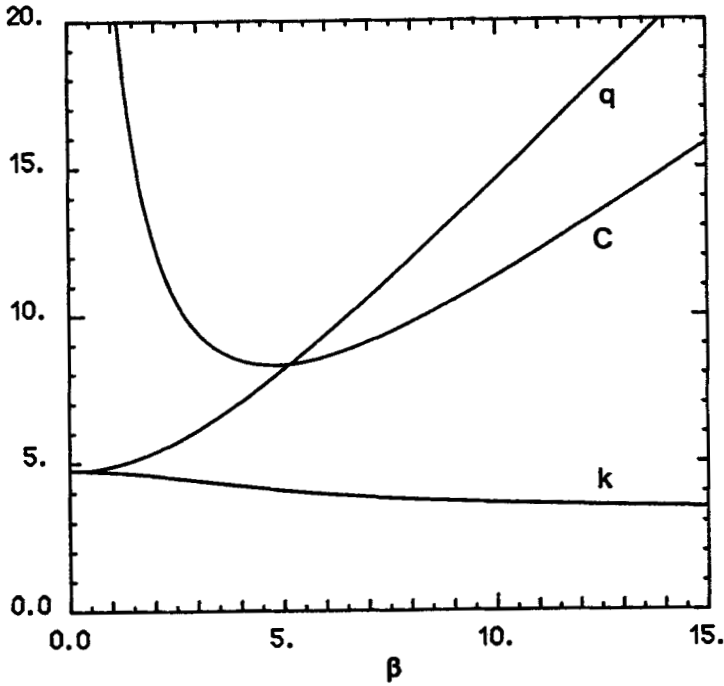


Fig. 1: Behavior of C , k and q as a function of β .

The predicted results are $\beta_p(r=0) \approx 1.5 \frac{\pi}{d}$, $k_p(r=0) \approx 1.3 \frac{\pi}{d}$, $q_p \approx 2.5 \frac{\pi}{d}$: they are consistent with ref.4. Note that the principal domain mode is independent of the anchoring energy and of the surface - like elasticity. But eq. (22) suggests that another mode of the periodic pattern may be present, provided $K_{2d}/\sqrt{W_6 W_\phi}$ is not vanishing: such a secondary mode appears only for materials having $r \approx 0$, and strongly depends on K_{2d} and on the anchoring strengths.

i.2 $K_{22} \neq 0$ and finite, $K_{11} \rightarrow \infty$

Defining now the reduced free energy and the corresponding reduced free energy density as $G^* \equiv 2F/(K_{22}d_x)$, $g^* \equiv 2f/(K_{22}d_x)$ the second equation of system (6) becomes

$$g_b^* = (\phi_y + \theta_z)^2 \quad (6^*)$$

independent of the reduced field h , whereas the second equation of system (7) is given by:

$$f_{si}^* = \mathcal{L}_\theta^{-1} \theta_i^2 + L_\phi^{-1} \phi_i^2 + 2(-)^i (1 + \kappa) (\theta_i \phi_{yi} - \phi_i \theta_{yi}) \quad (7^*)$$

where the extrapolation lengths are $\mathcal{L}_\theta \equiv K_{22}/W_\theta$, $L_\phi \equiv K_{22}/W_\phi$ and the surface - to - bulk elastic ratio is $\kappa^* \equiv K_{24}/K_{22}$. Hence the Euler- Lagrange equations read

$$\theta_{zz} + \phi_{yz} = 0$$

$$\phi_{yy} + \theta_{yz} = 0 \quad (8^*)$$

that give simply the extremality of the splay distortion $\partial(\text{div}\mathbf{n}) = 0$, whereas the boundary conditions if $\mathcal{L}_\theta \neq 0$ are obtained as

$$\phi_{yo} + \theta_{zo} = 0$$

$$-(1 + 2\kappa^*) \theta_{yo} + L_\phi^{-1} \phi_o - \phi_{zo} = 0 \quad (9^*)$$

Note that the first equation of system (9^{*}) is implicitly satisfied by every solution of system (8^{*}). We stress the fact that "a priori" there is no field threshold, due to the absence of h in Euler- Lagrange eqs.

It is not surprising the lack of symmetry between the case ($K_{22} \rightarrow 0$, $K_{11} \neq 0$ and finite) and the case ($K_{22} \neq 0$ and finite, $K_{11} \rightarrow +\infty$). In fact, in the first situation the field can induce a tilt, working against the splay- rigidity, whereas in the latter case the infinite splay- rigidity prevents any finite field from having any influence.

Aperiodic solution

No Freedericksz transition is ever possible, since eq. (8^{*}) simply gives

$$\theta_{zz} = 0 \quad (8'^*)$$

with the boundary condition

$$\theta_{zo} = 0 \quad (9'^*)$$

Hence the aperiodic solution is naïvely the undeformed state $\theta = 0$, $\phi = 0$, irrespective of the anchoring energy, and of the applied field.

Periodic solution

Let us look for a solution of the form

$$\theta = A \cos \alpha z \cos \beta y$$

$$\phi = a \sin \alpha z \sin \beta y \quad (23)$$

By putting (23) into the first integral of Euler - Lagrange equation

$$\theta_z + \phi_y = C_1 \quad (24)$$

where $C_1 = 0$ due to the first eq. of system (9*), we get an infinite set of solutions for $\alpha A = \beta a$. The solution $\alpha = \beta$, $a = A$ gives, through the second equation at the boundary:

$$2K_{24}/W_\phi = \frac{1}{\beta} \tan(\beta d/2) \quad (25)$$

providing the maximum threshold wavevector β_{PM} at the appearing of the periodic domains as a function of K_{24} , W_ϕ . In particular, in the case of strong torsional anchoring $W_\phi = \infty$, we have $\beta_{PM} = 2\pi/d$, consistently with ref.4. Here the periodic pattern starts irrespectively of the value of the applied field h , since the periodic distortion does not affect the bulk free energy, due to eq. (24). Thus, the undeformed P-alignment is intrinsically unstable for polymer nematics with K_{11} very large, and W_θ finite.

Let us now consider the case of strong tilt anchoring, where $W_\theta \rightarrow \infty$ as well as K_{11} . The first boundary condition (9*) changes and becomes:

$$\phi_{y0} - \theta_o + \theta_{zo} = 0 \quad (9^{**})$$

This means that, ensuring the solutions of (8*) the vanishing of $(\phi_y + \theta_z)$, also θ_o must be zero. Hence

$$\alpha_p = \pi/d \quad (26)$$

with infinite possible values of β_p , provided the torsional anchoring is very weak ($L_\phi \rightarrow \infty$).

ii. Bulk elastic ratio $r \rightarrow \infty$

This case, symmetrical with respect to the case i., turns out to be described by the same equations, provided the following transformations have been made:

$$K_{11} \leftrightarrow K_{22}, \quad W_0 \leftrightarrow W_\phi$$

$$\theta \leftrightarrow -\phi$$

$$\mathbf{H} = H\mathbf{k} \leftrightarrow \mathbf{H} = H\mathbf{j} \quad (27)$$

Hence, the results previously obtained are exactly the same: it is only necessary to interchange "splay" with "twist" and vice-versa.

CONCLUSIONS

We considered the possibility of building-up a P-aligned polymer nematic cell, in the case of very large bulk elastic anisotropy K_{11}/K_{22} (or K_{22}/K_{11}). We found that it is not allowed to reach such a simple undisturbed alignment, since it is unstable with respect to a periodically distorted one, ensuring the absence of splay (or of twist) everywhere. This means that a long chain polymer is expected to align with the single molecules lightly tilted, i.e. with an interconnected texture, instead of preferring an unidirectional planar orientation.

If the twist constant K_{22} (or K_{11}) vanish, there are two possible modes of the domain texture, one of which does not depend on the anchoring and on the surface-like elastic constant K_{24} . On the contrary, if K_{11} (or K_{22}) diverges, the domain texture essentially depends on the torsional anchoring strength rescaled by K_{24} .

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