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MAGNETIC - FIELD - INDUCED FERRONEMATIC-FERROCHOLESTERIC TRANSITION IN HOMEOTROPIC CELLS

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Este studiată comportarea unui cristal lichid colesteric cu anizotropie negativă ($\chi_a < 0$). Utilizând metoda analitică bazată pe ecuațiile Euler – Lagrange am găsit o relație între inducția cîmpului magnetic B şi raportul de confinare $r = d / p$ (unde d este grosimea celulei şi p este pasul cholesteric) la traziția dintr-o textură de tip nematic (aliniere homeotropă) la o textură de tip cholesteric (configurație invariantă la translație cu o răsucire uniformă în plan – TIC). Această relație este representată în coordonate (r, B)-printr-o eleipsă cu semiaxele mai mici decât în cazul unui crystal lichid pur printr-un factor care depinde de constantele de material şi grosimea celulei. Cu cât sunt mai mici feroparticulelele cu atât sunt mai mici semiaxele elipsei. Deoarece am gasit că unghiul total de răsucire de-a lungul celulei poate fi controlat prin modificarea fie a inducției magnetice fie a raportului de confinare. Rezultatele din lucrare por fi utile în proiecatearea dispozitivelor-optoelectronice.

The behavior of a confined ferrocholesteric liquid crystal with a negative magnetic anisotropy ($\chi_a < 0$) in a magnetic external field is studied. Using the analytical method based on the Euler- Lagrange equations we found the correlation between the field intensity B and the confinement ratio $r = d / p$ (d is the cell thickness and p is the cholesteric pitch) at the threshold of the transition from the nematic-like texture (homeotropic alignment) to the cholesteric-like texture (translationally invariant configuration with uniform in plane twist – TIC). This correlation is represented by an ellipse in (r, B)- coordinates with halfaxes smaller than in case of a pure liquid crystal by a factor depending on the material constants and cell thickness. The smaller are the ferroparticles, the smaller are the halfaxes of this ellipse. We found also that the total twist angle across the cell can be controlled by changing either the magnetic field intensity or the confinement ratio. Our results can be useful in designing magneto-optical devices.

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1. Introduction

Cholesteric liquid crystals (CLCs) display in the ground state a twisted helicoidal director field. When CLCs are confined in the cells with different boundary conditions or subjected to the electric, magnetic or laser fields, the cholesteric helix can be distorted or even completely unwounded; one can obtain a rich variety of director orientations controlled by varying the cell gap thickness d, the pitch p, the intensities of applied fields, the dielectric or magnetic properties of the used liquid crystals or the boundary conditions [1], [2].

While in the planar anchoring of cholesterics the boundary conditions are satisfied by orienting the axis of the helix normal to the plates, no orientation is compatible with homeotropic anchoring. The liquid crystal is therefore frustrated, and the more so when decreasing the sample thickness [1]. The experiment reported in [3] confirm that the textures of the liquid crystal strongly depend on the ratio $r = d / p$. Microscopic observations show the existence of a critical confinement ratio r_c such as: 1) for $r < r_c$ the cholesteric is completely unwound and the director is everywhere normal to the plates (homeotropic nematic phase); 2) for $r > r_c$ fingerprint textures appear composed of cholesteric fingers.

The confinement ratio r can be modified by changing either the thickness or the pitch. It is not convenied to change the cell thickness as a flow perturbing the director field arise. The pitch can be modified by changing the temperature, by introducing additives or by applying an external field.

The electric-field induced phase transition in frustrated cholesteric liquid crystal of a negative dielectric anisotropy was studied in [4], [5]. The authors reported the transition from the homeotropic alignment to the translational invariant configuration with uniform in plane twist when applying an electrical field on a cell with frustration ratio $r < 1$, containing a cholesteric liquid crystal of negative dielectric anisotropy.

The physical properties of CLCs change by introducing nanometric ferromagnetic particles. The behavior of these materials (called ferrocholesterics) under external fields was examined both theoretically and experimentally by many authors [6], [7], [8].

When an external magnetic field acts normal to the helical axis of a ferrocholesteric with a positive magnetic anisotropy ($\chi_a > 0$), the pitch of the cholesteric increases and for a critical value $B_{\uparrow}$, the ferrocholesteric-ferronematic transition takes place. If the magnetic field is perpendicular to the sample walls the obtained alignment is homeotropic. When the magnetic field decreases, the reverse transition occurs for a critical field $B_{\downarrow} < B_{\downarrow}$ [8].

In this paper we studied the behavior of a ferrocholesteric liquid crystal with a negative magnetic anisotropy ($\chi_a < 0$) in an external magnetic fields. We restricted ourselves to the translationally uniform structures which can be described analytically usingEuler- Lagrange equations. We found the correlation between the magnetic field intensity and the confinement ratio at the limit of the transition from the ferromagnetic homeotropic alignment to the translationally invariant configuration with uniform in plane twist (TIC) . Based on this correlation and using some practical values for the material's parameters we draw the spinodal line separating the metastable homeotropic configuration from the instable TIC. We also found the total twist angle across the sample as well as the possibility to control it by changing the magnetic field or/and the confinement ratio.

2. Theory

We assume a ferrocholesteric liquid crystal bounded by solid parallel walls where homeotropic boundary conditions are imposed to the director. The walls are parallel to the xy plane and located at $z = 0$ and $z = d$. According to the Burylov and Raicher hypothesis [9], in the absence of the external field the magnetic moment unit vector $\vec{m}$ of the ferroparticle is perpendicular to the molecular director $\vec{n}$. An uniform magnetic field $\vec{B}$ is applied to the Oz direction. The orientation of the three vectors ($\vec{B}$, $\vec{n}$, $\vec{m}$) is shown in Fig.1.

For the translational invariant configurations, the polar angles (θ and β) and twist angles (φ and γ) are functions of z only and the free-energy density takes the form

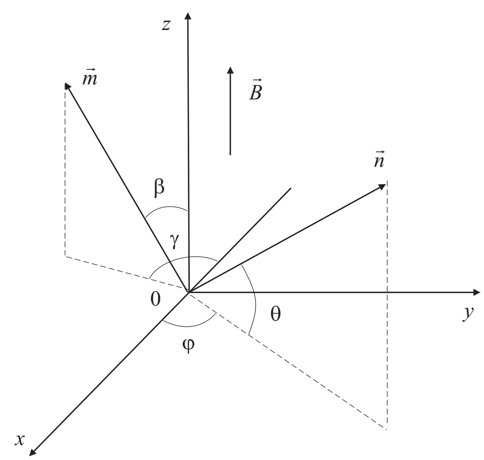

Fig. 1 Orientation of the magnetic moment and molecular director with respect to the static magnetic field $\vec{B}$

$$g = \frac{1}{2}\left(K_1 \cos^2\theta + K_3 \sin^2\theta\right)\dot{\theta}^2 + \frac{1}{2}\cos^2\theta\left(K_2\cos^2\theta + K_3\sin^2\theta\right)\dot{\varphi}^2 - K_2 q_0\left(\cos^2\theta\right)\dot{\varphi} +$$
$$\frac{K_2}{2}q_0^2 - \frac{1}{2}\mu_0^{-1}\chi_a B^2 \sin^2\theta - M_s f B\cos\beta +$$
$$f\frac{W}{a}\left[-\cos\theta\cos\varphi\sin\beta\cos\gamma - \cos\theta\sin\varphi\sin\beta\sin\gamma + \sin\theta\cos\beta\right]^2 + \frac{f k_B T}{V}\ln f \tag{1}$$

Here K_1, K_2, K_3 are the splay, twist and bend elastic constant respectively, χ_a is the magnetic anisotropy, W is the surface density of the anisotropic part of the interfacial energy of the particle-cholesteric boundary, a is the mean diameter of the magnetic particle, f is the volumic fraction of the magnetic particles and M_s is the saturation magnetization of the particles (i.e. the magnetizations $\vec{M} = M_s f\vec{m}$); $q_0 = \frac{2\pi}{p_0}$, p_0 being the cholesteric pitch; $\dot{\theta} = \frac{d\theta}{dz}$, $\dot{\varphi} = \frac{d\varphi}{dz}$.

There are four associated coupled Euler –Lagrange equations associated with boundary conditions ($\theta(0) = \theta(d) = \frac{\pi}{2}$; the angles $\beta(0)$, $\beta(d)$, $\varphi(0)$ and $\varphi(d)$ are arbitrary).

The first Euler-Lagrange equation

$$\frac{d}{dz}\left(\frac{\partial g}{\partial \dot{\gamma}}\right) - \frac{\partial g}{\partial \gamma} = 0 \tag{2}$$

is satisfied when $\gamma = \varphi$ [8].

Then, the part of g that contains β becomes

$$g_\beta = -M_s f B cps\beta + \frac{fW}{a}\sin^2(\theta - \beta) \tag{3}$$

and the second Euler-Lagrange equation

$$\frac{d}{dz}\left(\frac{\partial g_\beta}{\partial \dot{\beta}}\right) - \frac{\partial g_\beta}{\partial \beta} = 0$$

becomes

$$\frac{\partial g_\beta}{\partial \beta} = 0 \tag{4}$$

For small angles β ($\cos\beta \cong 1$, $\sin\beta \cong \beta$) and $\frac{2W}{a} << M_s B$ in Eq. 3 gives

$$g_\beta = -M_s fB + \frac{fW}{a}\sin^2 a \ . \tag{5}$$

The third Euler-Lagrange equation is

$$\frac{d}{dz}\left(\frac{\partial g}{\partial \dot{\varphi}}\right) - \frac{\partial g}{\partial \varphi} = 0 \tag{6}$$

and, with $\frac{\partial g}{\partial \varphi} = 0$ we obtain $\frac{\partial g}{\partial \dot{\varphi}} = C_0$

i.e.

$$-K_2 q_0 \cos^2\theta + \cos^2\theta(K_2\cos^2\theta + K_3\sin^2\theta)\dot{\varphi} = C_0 \tag{7}$$

Eq. 7 has to satisfied for any θ, including $\theta = \frac{\pi}{2}$, which leads to $C_0 = 0$ and

$$\dot{\varphi} = \frac{d\varphi}{dz} = \frac{K_2 q_0}{K_2\cos^2\theta + K_3\sin^2\theta} \tag{8}$$

The total twist angle $\Delta\varphi$ across the cell with thickness d is:

$$\Delta\varphi = \frac{2\pi}{p_0}\int_0^d \frac{K_2}{K_2\cos^2\theta + K_3\sin^2\theta}dz \tag{9}$$

Now we can express the free-energy density only in terms of the angle θ:

$$g\left(\theta,\dot{\theta}\right) = \frac{1}{2}\left(K_1\cos^2\theta + K_3\sin^2\theta\right)\dot{\theta}^2 - \frac{1}{2}\frac{K_2^2 q_0^2\cos^2\theta}{K_2\cos^2\theta + K_3\sin^2\theta}\left(\frac{fW}{a} - \frac{1}{2}\mu_0 B^2\chi_a\right)\sin^2\theta$$
$$-M_s fB + \frac{k_B fT}{V}\ln f + \frac{K_2}{2}q_0^2 \tag{10}$$

In the vicinity of the ferronematic-ferrocholesteric transition $\theta = \frac{\pi}{2} - \xi$, where ξ is a small angle. Consequently $\cos\theta = \sin\xi \cong \xi$; $\cos^2\theta = \xi^2$; $\sin^2\theta = 1 - \xi^2$; $\dot{\theta}^2 = \dot{\xi}^2$ and

$$g\left(\xi,\dot{\xi}\right) = \frac{K_3}{2}\left(1 + \xi^2\frac{K_1 - K_3}{K_3}\right)\dot{\xi}^2 - \frac{1}{2}\frac{K_2^2 q_0^2 \xi^2}{K_3\left(1 + \xi^2\frac{K_2 - K_3}{K_3}\right)} - A\xi^2 + D \tag{11}$$

where

$$A = \frac{fW}{a} - \frac{1}{2}\mu_0^{-1}\chi_a B^2 \tag{12}$$

D incorporates the terms which do not depend on ξ. For $\xi^2 << 1$ we obtain in the first approximation

$$g(\xi,\dot{\xi}) \cong \frac{K_3}{2}\dot{\xi}^2 - \frac{1}{2}\frac{K_2^2 q_0^2 \xi^2}{K_3} - A\xi^2 + D$$

The fourth Euler-Lagrange equation

$$\frac{d}{dz}\left(\frac{\partial g}{\partial \dot{\xi}}\right) - \frac{\partial g}{\partial \xi} = 0 \tag{13}$$

becomes

$$\dot{\xi}^2 + \left(\frac{2A}{K_3} + \frac{K_2^2 q_0^2}{K_3^2}\right)\xi = 0 \tag{14}$$

The solution of this equation is:

$$\xi = C_1 \sin(Rz) + C_2 \cos(Rz) \tag{15}$$

where

$$R^2 = \frac{2A}{K_3} + \frac{K_2^2 q_0^2}{K_3^2} \tag{16}$$

and C_1 , C_2 are determined using the boundary conditions:

$\xi(0) = 0$ leads to $C_2 = 0$

$\xi(d) = 0$ leads to $Rd = \pi$, $R^2 = \dfrac{\pi^2}{d^2}$

Substituting A from Eq. 12 and introducing $r = \dfrac{d}{p}$ we get:

$$\frac{r^2}{\left(\dfrac{K_3}{2K_2}\right)^2} - \frac{B^2}{\dfrac{\mu_0 K_3 \pi^2}{\chi_a d^2}} = 1 - \frac{2fW}{K_3 a}\frac{d^2}{\pi^2} \tag{17}$$

This is the equation relating the critical values of the confinement ratio r and the field intensity B inducing the transition from the homeotropic nematic – like alignment to a cholesteric –like translational invariant configuration with uniform plane twist (Fig. 2).

3. Discussion

In Eq. 17 we pointed out the critical values of parameters r and B producing a Freedericksz transition in a pure liquid crystals [10]. So for $r_F << \frac{K_3}{2K_2}$ the transition cholesteric- nematic “without field” occurs [11].

The value $B_F = \frac{\pi}{d}\sqrt{\frac{\mu_0 K_3}{\chi_a}}$ is the threshold field for the same transition in a pure liquid crystal with positive magnetic anisotropy.

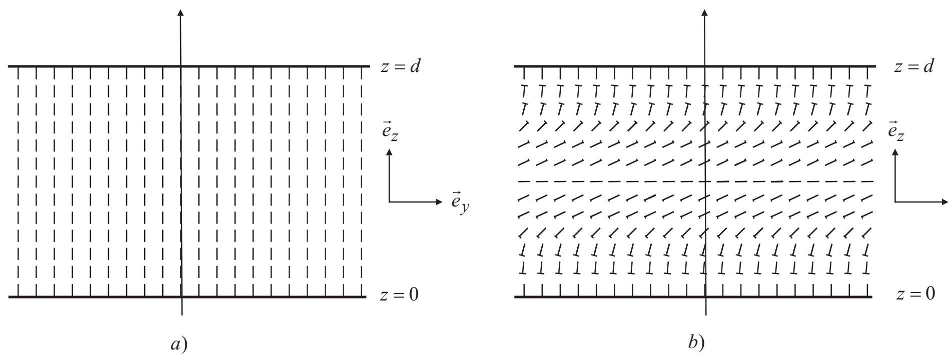

Fig. 2 Representation of the homeotropic (a) and TIC (b) alignment. Molecules are represented as nails pointing out of the image

When introducing the ferromagnetic particles these critical values change. Let us suppose that the confinement ratio is smaller that the critical one for the ferrocholesteric liquid crystal and in the whole sample the arrangement is homeotropic.

If the magnetic anisotropy is negative ($\chi_a < 0$) the magnetic field perpendicular to the walls perturbs the homeotropic alignment pushing the system to perform the transition to TIC with uniform in plane twist. In this case Eq. (17) describes a spinodal ellipse in the plane r , B (Fig. 3) .

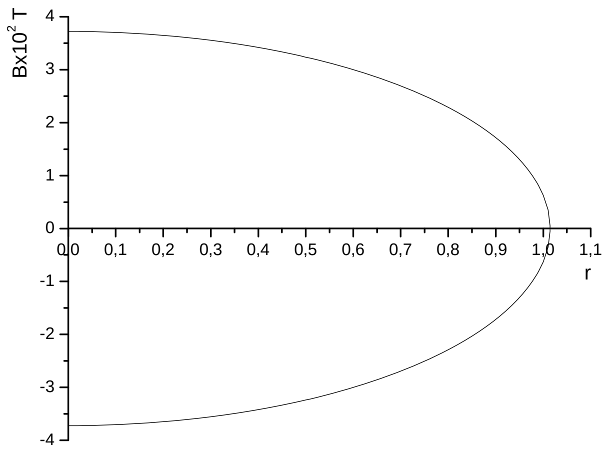

Fig. 3 Magnetic induction B function of r

The half-axes of the ellipse are smaller that in the case of a pure cholesteric by a factor of $\left(1-\frac{2fW}{K_3 a}\frac{d^2}{\pi^2}\right)^{1/2}$

For the values of the parameters r and B inside the ellipse the homeotropic arrangement is metastable. Values of the parameters outside the ellipse correspond to a locally unstable equilibrium: this is due to the growth of a ferrocholesteric phase..

A branch of this ellipse is drawn in Fig. 3. We used the following values for material parameters: $\chi_a = 7\times10^{-7}$, $W = 5\times10^{-10}$ N/m; $f = 10^{-3}$; $a = 3\times10^{-9}$ m; $K_1 = 17.2\times10^{-12}$ N; $K_2 = 7.51\times10^{-12}$ N; $K_3 = 17{,}9\times10^{-12}$ N; $d = 400$ μm.

It follows from Eq. 9 that the total twist angle $\Delta\varphi$ has a minimum value $\Delta\varphi_{\min} = 2\pi r\frac{K_2}{K_3}$ at the threshold of the transition ($\cos^2\theta \cong \xi^2$, $\sin^2\theta \cong 1-\xi^2$ and $\xi << 1$) and a maximum one, $\Delta\varphi_{\max} = 2\pi r$ when B is much higher that the threshold value ($\theta \to 0$).

Therefore, for a given cell it is possible to control the twist angle by changing the field intensity B .This fact is important for magneto-optical devices as the twist angle control the optical phase retardation.

4. Conclusion

In this paper we analyzed the influence of a magnetic field on the director orientation of a ferrocholesteric liquid crystal with negative magnetic anisotropy, confined in a cell with homeotropicaly alignment imposed at the walls.

We found analytically the correlation between the critical magnetic field $\vec{B}$ and the confinement ratio $r = d / p$ at the threshold of the transition from the nematic –like configuration (homeotropic alignment) to the cholesteric-like texture (translationaly invariant configuration with uniform in plane twist). Based on this correlation and using some usual values for the material parameters we drawn the phase diagram which is an ellipse in the plane r, B. For values of r and B inside the ellipse the homeotropic configuration is metastable while for values of r, B outside the ellipse the transition to TIC takes place. The main difference between our setup and a pure liquid crystal is the presence of an additional term in the right size of Eq. 17. The effect of this term is to shrink the size of the spinodal ellipse by a factor of $\left(1-\frac{2fW}{K_3 a}\frac{d^2}{\pi^2}\right)^{1/2}$.

We may conclude that the mean diameter of the ferroparticles is essential in the behavior of the cell.

We found also that the total twist angle across the cell can be varied between a lower and an upper limit by changing the magnetic field or/and the confinement ratio.

Our results can be useful in designing of magneto-optical devices such as light shutters and displays.

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