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

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Se investighează influența unui fascicol laser asupra structurii directorului unui cristal lichid ferocolesteric (FC) confinat într-o celulă homeotropică. Utilizând metoda analitică bazată pe ecuațiile Euler – Lagrange se găseşte o relație între intensitatea fascicolului laser şi raportul de confinare $r = d / p$ (unde d este grosimea celulei şi p este pasul colesteric) la pragul tranziției dintr-o textură de tip nematic (aliniere homeotropă) la o textură de tip colesteric (configurație invariantă la translație cu o răsucire uniformă în plan – TIC). Pe baza acestei corelații şi utilizând valori obişnuite pentru parametrii de material se construieşte elipsa spinodală care separă configurația homeotropică metastabilă de configurația nestabilă TIC.

We investigate the influence of a laser beam on the director structure of a ferrocholesteric liquid crystal (FC) confined in homeotropic cells. Using the analytical method based on the Euler- Lagrange equations we find a correlation between the laser beam intensity 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). Based on this correlation and using some practical values for the material's parameters we construct the spinodal line,(spinodal ellipse) separating the metastable homeotropic configuration from the unstable TIC.

1. Introduction

Ferronematics (FNs) and ferrocholesterics (FCs) are composite materials consisting in a dilute uniform suspension of magnetic grains in a nematic or a cholesteric liquid crystal matrix.

In a continuum approach the FNs or FCs are described by the macroscopic director $\vec{n}$ and the total magnetization $\vec{M}(\vec{r}) = M_s f\vec{m}$, where M_s is the saturation

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magnetization of the particle, f is the concentration of the magnetic particles and $\vec{m}$ the unit vector of the magnetic moment of the ferroparticle. The most essential feature of FNs or FCs is an orientational coupling between the dispersed phase (magnetic particles) and the liquid crystal matrix.

According to the Burylov – Raikher approach [1], [2] the anchoring energy which determines the orientation of the liquid crystal molecules on the ferroparticle surface is finite and, in the equilibrium state, the magnetic moment of the ferroparticle is perpendicular to the liquid crystal director.

In confined structures the contribution to the free energy due to the elasticity, surface anchoring , coupling between components and coupling to the external fields are the same order of magnitude. The competition between these contributions produces a large variety of director structures that can be controlled by material parameters of liquid crystal, dimensions of ferroparticles, the confinement ratio , suitable surface treatment or applied external fields.

In particular, when a FC is confined in cells with different boundary conditions or subjected to an electric, magnetic or laser field, the cholesteric helix can be distorted or even completely unwound. While in the planar anchoring of FCs 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 with decreasing the sample thickness [3]. Therefore, the external fields and confinement ratio $r = d / p$ (d is the cell thickness and p is the cholesteric pitch) are the main parameters whose changing gives the possibility to control the director structures in homeotropic cells.

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 (TIC) with uniform in plane twist when applying an electrical field on a cell containing a cholesteric liquid crystal of negative dielectric anisotropy, with frustration ratio $r < 1$.

The behavior of a confined ferrocholesteric in superposed magnetic and laser fields was studied in [6]. It was found a correlation between the field intensities and the confinement ratio at the limit of the transition from the homeotropic configuration to the TIC. This correlation was discussed as a function of the sign of the magnetic and dielectric anisotropies.

In this paper we study the laser induced transition from the nematic –like texture (homeotropic configuration) to the cholesteric–like one (TIC) in a confined ferrocholesteric liquid crystal using the analytical method based on the Euler- Lagrange equations. We find the possibility to "drive" the transition by changing either the laser beam intensity or the confinement ratio.

2. Theory

We assume a ferrocholesteric liquid crystal bounded by solid transparent walls parallel to the xy plane and located at $z = 0$ and $z = d$. Let us have a laser beam whose vector $\vec{k}$ is parallel to Oz and polarized along Oy.
The cholesteric director $\vec{n}$ and the magnetic moment unit vector $\vec{m}$ are caracterised by the polar angles θ respectively β and azimutal angles (the twist angles) φ respectively γ (Fig.1).

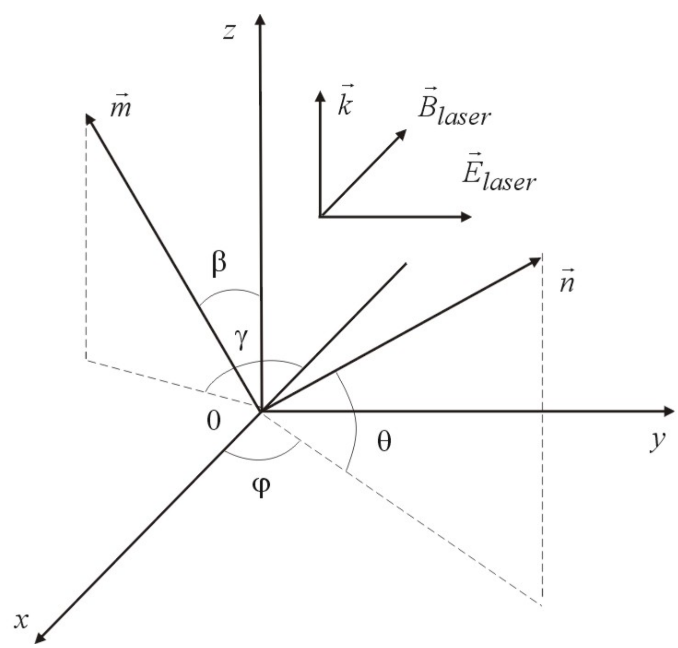

Fig. 1 Orientation of the magnetic moment and molecular director with respect to the laser field

We restrict our study to the translationally uniform structures where the four angles are functions of z only. In this geometry the free-energy density takes the form [6], [7]:

$$g = \frac{1}{2}\left(K_1 \cos^2\theta + K_3 \sin^2\theta\right)\dot{\theta}^2 + \frac{1}{2}\cos^2\theta\left(K_2\cos^2\theta + K_3\sin^2\theta\right)\dot{\varphi}^2 - K_2 q_0\left(\cos^2\theta\right)\dot{\varphi} +$$
$$\frac{K_2}{2}q_0^2 + f\frac{W}{a}\left[-\cos\theta\cos\varphi\sin\beta\cos\gamma - \cos\theta\sin\varphi\sin\beta\sin\gamma + \sin\theta\cos\beta\right]^2 +$$
$$\frac{fk_BT}{V}\ln f - \frac{I\left(\varepsilon_{II}\varepsilon_{\perp}\right)^{1/2}}{\left(\varepsilon_{\perp} + \varepsilon_a\sin^2\theta\right)^{1/2}} \tag{1}$$

Here K_1, K_2, K_3 are the splay, twist and bend elastic constant respectively, $\varepsilon_{II}, \varepsilon_{\perp}$ are the dielectric constants parallel and perpendicular to $\vec{n}$, respectively; $\varepsilon_a = \varepsilon_{II} - \varepsilon_{\perp}$ is the dielectric anisotropy assumed positive, 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, I`is the mean volumic density of the electromagnetic energy of the light (connected with the light intensity J by the relation $J = cI$), $q_0 = \frac{2\pi}{p_0}$, p_0 being the cholesteric pitch; $\dot{\theta} = \frac{d\theta}{dz}$, $\dot{\varphi} = \frac{d\varphi}{dz}$.

There are four 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 $\gamma = \varphi$ [7].

Then, the part of g that contains β becomes

$$g_\beta = \frac{fW}{a}\left(\sin\theta\cos\beta - \cos\theta\sin\beta\right)^2 \tag{3}$$

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 ($\cos\beta \cong 1$, $\sin\beta \cong \beta$) Eq. 4 has two roots $\beta_1 = \tan\theta$ (unsuitable at $\theta \to \frac{\pi}{2}$) and $\beta_2 = -\cot\theta$ wich leads to

$$g_\beta(\theta) = \frac{fW}{a}\frac{1}{\sin^2\theta}. \tag{5}$$

The free energy density becomes independent on φ and the third Euler-Lagrange equation admits the prime integral

$$\left(\frac{\partial g}{\partial\dot{\varphi}}\right) = C_0 \tag{6}$$

i.e.

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

which must be satisfied for any θ, including $\theta = \frac{\pi}{2}$. Therefore $C_0 = 0$ and

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

Substituting Eq. 8 in Eq. 1 we obtain the free – energy density in terms of the angle θ only

$$g(\theta,\dot{\theta}) = \frac{1}{2}(K_1\cos^2\theta + K_3\sin^2\theta)\dot{\theta}^2 - \frac{1}{2}\frac{K_2^2 q_0^2\cos^2\theta}{K_2\cos^2\theta + K_3\sin^2\theta} + \frac{fW}{a}\frac{1}{\sin^2\theta} - \frac{I(\varepsilon_{II}\varepsilon_\perp)^{1/2}}{(\varepsilon_\perp + \varepsilon_a\sin^2\theta)^{1/2}} + \frac{k_B fT}{V}\ln f + \frac{K_2}{2}q_0^2 \tag{9)}$$

In the vicinity of the transition from the homeotropic alignment (nematic– like state) to the translationaly invariant configuration with uniform in plane twist (ferrocholesteric – like alignment) $\theta = \frac{\pi}{2} - \xi$, 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

$$g(\xi,\dot{\xi}) = \frac{K_3}{2}\left(1 + \xi^2\frac{K_1 - K_3}{K_3}\right)\dot{\xi}^2 - \frac{1}{2}\frac{K_2^2 q_0^2 \xi^2}{K_3\left(1 + \xi^2\frac{K_2 - K_3}{K_3}\right)} + \frac{fW}{a}(1-\xi^2)^{-1} - I\varepsilon_\perp^{1/2}\left(1 - \frac{\varepsilon_a}{\varepsilon_{II}}\xi^2\right)^{-\frac{1}{2}} + \frac{k_B fT}{V}\ln f + \frac{K_2}{2}q_0^2 \tag{10}$$

For $\xi^2 << 1$ ignoring higher order terms we can obtain:

$$g\left(\xi,\dot{\xi}\right)\cong\frac{K_3}{2}\left(\dot{\xi}\right)^2+\xi^2\left(-\frac{1}{2}\frac{K_2^2q_0^2}{K_3}+\frac{fW}{a}-I\frac{\varepsilon_\perp^{1/2}\varepsilon_a}{\varepsilon_{II}}\right)+A, \qquad (11)$$

A incorporating the terms which do not depend on ξ

The fourth Euler-Lagrange equation

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

leads to:

$$\left(\frac{d^2\xi}{dz^2}\right)^2+\left(\frac{K_2^2q_0^2}{K_3^2}-\frac{2fW}{aK_3}+I\frac{\varepsilon_\perp^{1/2}\varepsilon_a}{\varepsilon_{II}K_3}\right)\xi=0. \qquad (12)$$

The solution of this equation is:

$$\xi(z)=C_1\sin(Rz)+C_2\cos(Rz) \qquad (13)$$

where

$$R^2=\frac{K_2^2q_0^2}{K_3^2}-\frac{2fW}{aK_3}+I\frac{\varepsilon_\perp^{1/2}\varepsilon_a}{\varepsilon_{II}K_3} \qquad (14)$$

and C_1 , C_2 are integration constants. Using the boundary conditions $\xi(0)=\xi(d)=0$ we obtain $C_2=0$ and $Rd=\pi$.

Substituting R from Eq. 14 and introducing the confinement ratio $r=\frac{d}{p}$ we find the relation between the confinement ratio and the laser beam intensity at the threshold of the transition from the homeotropic ferronematic-like alignment to the ferrocholesteric – like translational invariant configuration:

$$\frac{r^2}{\left(\frac{K_3}{2K_2}\right)^2}+\frac{\sqrt{I}^2}{\frac{K_3\varepsilon_{II}\pi^2}{\varepsilon_a\varepsilon_\perp^{1/2}d^2}}=1+\frac{2fW}{K_3a}\frac{d^2}{\pi^2} \qquad (15)$$

3. Discussion

In Eq. 15 we pointed out the critical values of parameters r and I producing a Freedericksz transition in a pure liquid crystal. So for $r_F<<\frac{K_3}{2K_2}$ the transition cholesteric- nematic "without field" occurs [8], [9].

The value $I_F=\frac{K_3\varepsilon_{II}\pi^2}{\varepsilon_a\varepsilon_\perp^{1/2}d^2}$ is the threshold laser beam intensity for the same transition [3]. Introducing the ferromagnetic particles these critical values change.

Let us suppose that the confinement ratio is smaller that the critical one and in the whole sample the arrangement is homeotropic.

If the dielectric anisotropy is positive ($\varepsilon_a > 0$) the electric field of the electromagnetic wave perturbs the homeotropic alignement pushing the system to perform the transition to translational invariant configuration with uniform in plane twist. The total twist angle across the cell can be obtained from Eq. 8:

$$\Delta\varphi = \frac{2\pi}{p}\int_o^d \frac{K_2}{K_2\cos^2\theta + K_3\sin^2\theta}dz$$

This angle is minimum $\Delta\varphi_{\min} = 2\pi r\frac{K_2}{K_3}$ at the threshold of transition or maximum $\Delta\varphi_{\max} = 2\pi r$ for laser beam intensity much higher that the threshold value (when $\theta \to 0$).

Eq.15 describes a spinodal ellipse in the $r - \sqrt{I}$ plane. This ellipse defines the limit of metastability of the homeotropic phase: for r and $\sqrt{I}$ inside this ellipse the uniform homeotropic configuration is metastable, while outside the ellipse, it is a locally unstable equilibrium.

In an idealized cell with infinitely strong homeotropic anchoring and no pretilt, the transition from the homeotropic to TIC is rather a bifurcation than a phase transition (in the usual thermodynamic sense) [3]. A branch of the ellipse (15) is drawn in Fig. 2 (the full line).We used following values for material parameters: $\varepsilon_{II} = 2{,}89$, $\varepsilon_{\perp} = 2{,}25$, $W = 5\times10^{-10}$ 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.

On the same figures, the dashed line corresponds to the spinodal ellipse in the case of a pure cholesteric liquid crystal (obtained from Eq. 15 for $f = 0$). One can see that the presence of the ferroparticles increases slightly the laser beam intensity necessary for the transition.

Any how the transition occurs for laser beam intensities ($J = cI$) of order of 100-120 W/cm^2 which are experimentally accessible.

For a similar cell containing the same ferrocholesteric , if the homeotropic alignment is obtained using a magnetic field, for the same value of the confinement ratio, the necessary laser beam intensity for the transition is greater [6].

Our results can be useful in designing of optical devices such as light shutters or laser writing devices.

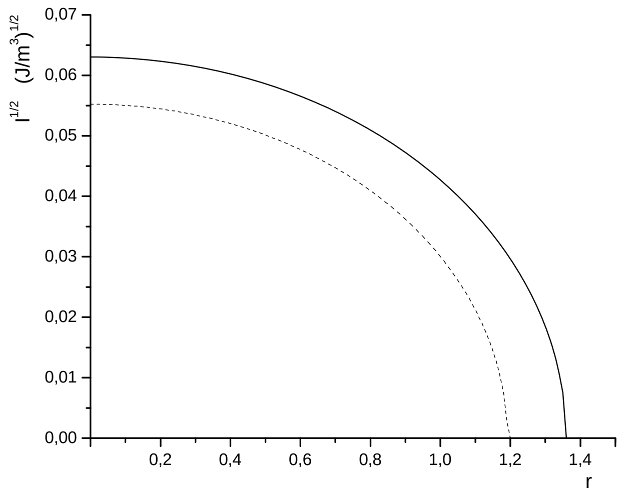

Fig. 2 The spinodal ellipses for ferrocholesteric (full line) and pure liquid crystal (dashed line)

4. Conclusion

In our paper we studied analytically the transition from the nematic –like to the cholesteric –like textures of a confined liquid crystal matrix containing ferromagnetic particles, subjected to a laser beam.

Using Euler-Lagrange equation with adequate boundary conditions we found a correlation between the critical laser beam intensity and the confinement ratio at the threshold of the transition from the homeotropic arrangement to TIC.

This correlation can be seen as a phase diagram in the (r, $\sqrt{I}$) plane having the shape of an ellipse (spinodal ellipse). For values of r and I inside the ellipse the homeotropic configuration is metastable while for values outside the ellipse the transition to TIC takes place.

The difference between our setup and a pure liquid crystal is the presence of an additional term in the right size of the Eq. 15. The effect of this term is to broaden the size of the ellipse, i.e. to increase the critical confinement ratio and the critical intensity . In spite of this increase, many practical devices use ferroparticles of ferronematics, taking advantage of their other "good " properties.

Our results can be useful in designing of practical devices such as light shutters or laser writing devices.

A more detailed study of the interaction between a laser beam and a ferrocholesteric (or ferronematic) should probably consider a new term in Eq. 1, due to action of the magnetic field of the laser beam on the magnetic moment of the ferroparticles.

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