U.P.B. Sci. Bull., Series A, Vol. 76, Iss. 1, 2014 ISSN 1223-7027

FERROCHOLESTERIC - FERRONEMATIC TRANSITION IN MAGNETIC AND LASER FIELD

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In this paper we study the behavior of a ferrocholesteric liquid crystal (FCLC) in magnetic and laser external fields, taking into account the influence of the molecular anchoring to the solid boundaries. Using the analytical method based on the Euler-Lagrange equations and the expression of the anchoring energy we find the correlation between the critical fields, the confinement ratio and the anchoring parameters at the threshold of the transition ferrocholesteric-ferronematic. A comparison to the case of the strong anchoring is given.

Keywords: ferronematics, ferrocholesterics, magnetic and optical Freedericksz transition, threshold field, weak anchoring.

1. Introduction

The cholesteric - nematic transition can be induced in one of two ways [1-2]: applying an external field (magnetic, electric and/or optic) or imposing to the sample boundaries conditions that are topologically incompatible with the helicoidal structure of the director field but favour the nematic alignment. These two methods can be combined, diversifying the richness of the observed phenomena. The physical properties of the liquid crystals change when ferromagnetic particles are introduced due to the coupling between the ferroparticles and the liquid crystalline matrix. The obtained composite - a ferroliquid crystal (ferronematic-FN, ferrocholesteric-FC or ferrosmectic-FS) preserves all features of orientational behaviour of liquid crystals at the same time earning the high magnetic susceptibility of the dispersed particles.

For a ferroliquid crystal there are two mechanism of interactions with the external magnetic fields: one caused by the influence of the magnetic fields on the diamagnetic liquid crystal matrix (a quadripolar mechanism) and another caused by the interaction of the magnetic field with the magnetic moment of the ferroparticles (a dipolar mechanism) [3].

The two mechanisms can *compete* if the magnetic moment of the ferroparticle ($\vec{m}$) is perpendicular to the LC director (soft anchoring of the homeoptropic type between the LC molecules and the ferroparticles) or can concur if the two vector are parallel (strong anchoring of the planar type between the LC molecules and the ferroparticles). In both cases the magnetic anisotropy of the LC matrix is supposed to be positive, $\chi_a > 0$.

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The ferronematics and nematic behaviour in external fields have been studied by many authors both theoretically [4-15], and experimentally [16-21].

The authors have underlined the main changes of the physical phenomena due to the ferroparticles intrusion in LC (the modification of the threshold fields, responding times, transitions temperatures, capacitance, etc.).

The behaviour of the ferrocholesterics in external fields have been also studied in many papers [22-25].

The authors have determined the correction to the critical magnetic field producing the transition FC-FN for both situations: the field on and the field off.

The consequences of the magnetic admixture redistribution in a uniform magnetic field i.e. the segregation effect-signalled still in [3] were pointed out in[8-10], [22]. The most important consequence is the tricritical behaviour both ferrocholesteric-ferronematic transition and magnetic Freedericksz transition in ferronematics.

Besides the character of the anchoring between the molecules of LC and ferroparticles (soft or strong) an important role in the behaviour of LC in external fields is played by the character of the anchoring of the LC molecules to the delimiting surfaces of the samples. Rapini and Papoular have introduced the term of "weak anchoring" and have proposed a phenomenological formula for the anchoring energy [26]:

$$g_s = \frac{1}{2} A \sin^2 \theta \qquad (1)$$

where θ_0 is the value of the tilt angle at the boundary and A is the anchoring strength.

In the absence of the external action, the preferred direction of the molecules on the boundary (the so-called easy axis) minimizes the anchoring energy.

Many authors have studied the influence of the weak anchoring on the critical fields producing Freederiksz transition using Rapini-Papoular formula [15], [22] [27-29].

However it has been shown that in some cases results using Rapini-Papoular formula do not agree well with experimental results [30].

Instead, more intricated expressions for the anchoring energy have been proposed in [31-34].

Thus, Young and Rosenblatt [31] have chosen for the anchoring energy per unit area a quadratic form derived from Rapini-Papoular expression:

$$g_s = \frac{1}{2} A \sin^2 \theta_0 \left(1 + \xi \sin^2 \theta_0\right) \qquad (2)$$

Here A is the anchoring strength and ξ is a dimensionless coefficient. They have determined experimentally that the influence of a magnetic field on a tilted nematic can be modelled more accurately using Eq. (2) instead of Eq. (1), when $\xi > 0$.

In [33] and [34] the authors have shown that $\xi < 0$ leads to a first-order Freedericksz transitions in a NLC cell. Eq. (2) was used also in [13] related to the behaviour of a ferronematic in magnetic field. A similar form of the anchoring energy per unit area was taking into account [35] for modelling the bidirectional surface anchoring of a planar cholesteric layer.

In our actual study we focus our attention on the influence of the weak anchoring on the FC-FN transition in superposed magnetic and laser fields, using for the anchoring energy per unit area the expression (2).

Unlike the situation studied in [22] where the authors have considered the liquid crystal molecules and the ferroparticles to be strong anchored (the magnetic moment of the ferroparticle and the nematic director are parallel), in our work we consider the soft anchoring between the two types of molecules and consequently the magnetic moment of the ferroparticle is perpendicular to the director [4-7].

We obtain a correlation between the critical fields, the confinement ratio (the ratio between the cell thickness and the cholesteric pitch) and the anchoring strength at the threshold of the transition FC-FN. This correlation is discussed both for positive and negative values of the magnetic anisotropy.

2. Theory

We consider a FC liquid crystal confined between two plates parallel to the xy plane and placed at $z = d$ and $z = -d$.

The configuration of the molecular director $\vec{n}$ and the unit vector of the magnetic moment of the ferroparticle $\vec{m}$ in the presence of the static magnetic field $\vec{B}$ and the laser beam is presented in Fig. 1 where

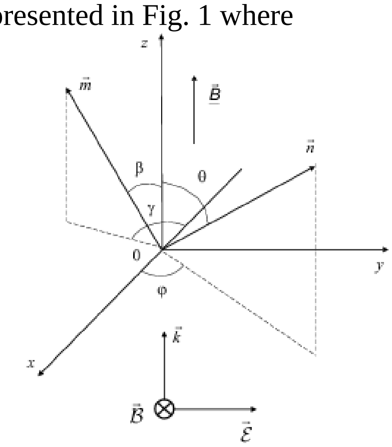


Fig. 1 The tilt angles of the vectors $\vec{n}$, and $\vec{m}$ in the presence of the static field $\vec{B}$ and the laser beam (⎯ ≐ and k) are the characteristic vectors othe electromagnetic wave.

In this geometry the free energy density expression in the bulk of the system is [3], [6], [22], [36]:

$$g_s = [\frac{1}{2}\left(K_1\sin^2\theta + K_3\cos^2\theta\right)\dot{\theta}^2 + \frac{1}{2}\left(K_2\sin^2\theta + K_3\cos^2\theta\right)(\sin^2\theta)\dot{\varphi}^2 -$$

$$-K_2 q_0\left(\sin^2\theta\right)\dot{\varphi} + \frac{K_2}{2}q_0^2] - \frac{1}{2}\frac{\chi_m}{\mu_0}B^2\cos^2\theta - M_s fB\cos\beta +$$

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

$$+\frac{fk_BT}{V}\ln f - \frac{I\left(\varepsilon_{\|}\varepsilon_{\perp}\right)^{1/2}}{\left(\varepsilon_{\perp} + \varepsilon_a\cos^2\theta\right)^{1/2}}$$

Here K_1, K_2 and K_3 are the splay, twist and bend elastic constant, respectively; $\varepsilon_{\|}$, $\varepsilon_{\perp}$ are the dielectric constants (parallel and perpendicular to $\vec{n}$), ε_a and χ_a are the dielectric and magnetic anisotropy respectively, W is the surface density of the anisotropic part of the interfacial energy at the ferroparticle-cholesteric boundary, a is the mean diameter of the magnetic particles f is their volume fraction and M_s is the saturation magnetization of the particles, I is the mean volumic density of the electromagnetic energy of the light (connected to the intensity of the light J by the relation $J = cI$; $q_0 = 2\pi / p_0$, p_0 being the cholesteric pitch; $\dot{\theta} = \frac{d\theta}{dz}$ and $\dot{\varphi} = \frac{d\varphi}{dz}$.

The Gibbs free energy of the system is

$$G = S\int_{-d}^{+d} g_B\left(\theta,\dot{\theta},\varphi,\dot{\varphi},\beta,\dot{\beta},\gamma,\dot{\gamma}\right)dz + SA\sin^2\theta_0\left(1+\xi\sin^2\theta\right)$$

(S is the substrates area). There are four coupled Euler-Lagrange equations with associated boundary condition [37]:

$$\left(K_1\sin^2\theta_0 + K_3\cos^2\theta_0\right)\left(\frac{d\theta}{dz}\right)_{z=\pm d} = A\sin\theta_0\cos\theta_0\left(1+2\xi\sin^2\theta_0\right) \tag{4}$$

The angles $\beta(\pm d)$, $\varphi(\pm d)$, $\gamma(\pm d)$ are arbitrary. In the middle of the cell ($z=0$), $\theta(0)=\theta_m$, $\dot{\theta}=0$; we assume the distortions to be symmetric $\theta(z)=\theta(-z)$

Following the considerations made in [23] and [25] from the first three Euler-Lagrange equations referring to γ, β, φ we obtain:

$$\gamma = \beta; \quad \cos\beta = 1; \quad \dot{\varphi} = \frac{K_2 q_0}{K_2 \sin^2\theta + K_3 \cos^2\theta}$$

and:

$$g_B\left(\theta,\dot{\theta}\right) = \frac{1}{2}\left(K_1 \sin^2\theta + K_3 \cos^2\theta\right)\dot{\theta}^2 - \frac{1}{2}\frac{K_2^2 q_0^2 \sin^2\theta}{K_2 \sin^2\theta + K_3 \cos^2\theta} + \\ + \left(\frac{fW}{a} - \frac{\chi_a B^2}{2\mu_0}\right)\cos^2\theta - M_s fB + \frac{k_B fT}{V}\ln f + \frac{K_2}{2}q_0^2 - \frac{I\left(\varepsilon_{\|}\varepsilon_{\perp}\right)^{1/2}}{\left(\varepsilon_{\perp} + \varepsilon_a \cos^2\theta\right)^{1/2}} \tag{5}$$

The fourth Euler-Lagrange equation will be:

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

and will be solved with associated boundary condition (4).

As $g_B\left(\theta,\dot{\theta}\right)$ does not depend explicitly on z, we have:

$$g_B\left(\theta,\dot{\theta}\right) - \dot{\theta}\frac{\partial g_B\left(\theta,\dot{\theta}\right)}{\partial\dot{\theta}} = C \tag{6}$$

The constant C is determined from the conditions in the middle of the cell. After some calculations we obtain:

$$K_3\left(1 + \eta\sin^2\theta\right)\left(\frac{d\theta}{dz}\right)^2 + \left(\sin^2\theta - \sin^2\theta_m\right)T = 0 \tag{7}$$

where

$$T = \frac{K_2^2 q_0^2}{K_3}\frac{1}{\left(1 + \eta_1 \sin^2\theta\right)\left(1 + \eta_2 \sin^2\theta_m\right)} + \left(\frac{2fW}{a} - \frac{\chi_a B^2}{\mu_0}\right) + I\frac{\sqrt{\varepsilon_{\|}}\varepsilon_a}{\varepsilon_{\perp}} \\ \eta_1 = \frac{K_1 - K_2}{K_3} \quad \text{and} \quad \eta_2 = \frac{K_2 - K_3}{K_3} \tag{8}$$

Now, we do the substitution:

$$\sin\lambda = \frac{\sin\theta}{\sin\theta_m} \tag{9}$$

From Eq. (7) we obtain:

$$dz = \sqrt{\frac{K_3\left(1 + \eta_1 \sin^2\lambda \sin^2\theta_m^2\right)}{T}}\frac{d\lambda}{\sqrt{1 - \sin^2\lambda\sin^2\theta_m}} \tag{10}$$

and, integrating Eq. (10):

$$\int_{-d}^{0} dz = \int_{\lambda_0}^{\pi/2} G\left(\lambda,\theta_m\right) d\theta \tag{11}$$

where

$$G\left(\lambda,\theta_m\right) = \sqrt{\frac{K_3\left(1+\eta_1 \sin^2\lambda \sin^2\theta_m\right)\left(1-\sin^2\lambda\sin^2\theta_m\right)^{-1}}{T}} \tag{12}$$

$$\sin\lambda_0 = \frac{\sin\theta_0}{\sin\theta_m} \tag{13}$$

At the limit of the transition $\theta_m \to 0$ from Eq. (11) we obtain:

$$d = \left(\frac{\pi}{2} - \lambda_0\right)\sqrt{\frac{K_3}{\frac{K_2^2 q_0^2}{K_3} + \left(\frac{2fW}{a} - \frac{\chi_a B_c^2}{\mu_0}\right) + I_c \frac{\sqrt{\varepsilon_{\parallel}}\varepsilon_a}{\varepsilon_{\perp}}}} \tag{14}$$

where B_c and I_c are the critical values of the two fields. From Eq. (4) we have:

$$K_3\left(1+\eta\sin^2\theta_0\right)\left(\frac{d\theta}{dz}\right)_{z=\pm d} = T\left(\sin^2\theta_m - \sin^2\theta_0\right) \tag{16}$$

Eqs. (15) and (16), with the substitution (13) lead to:

$$\tan\lambda_0 = \frac{\sqrt{K_3}}{A}\sqrt{\frac{K_2^2 q_0^2}{K_3} + \left(\frac{2fW}{a} - \frac{\chi_a B_c^2}{\mu_0}\right) + I_c \frac{\sqrt{\varepsilon_{\parallel}}\varepsilon_a}{\varepsilon_{\perp}}} \tag{17}$$

Eliminating λ_0 between Eqs. (14) and (17), introducing the confinement ratio $r = 2d / p_0$ and naming

$$Y_c = \sqrt{\frac{\pi^2 K_2^2 r^2}{K_3 d^2} + \left(\frac{2Wf}{a} - \frac{\chi_a B_c^2}{\mu_0}\right) + I_c \frac{\sqrt{\varepsilon_{\parallel}}\varepsilon_a}{\varepsilon_{\perp}}} \tag{18}$$

we obtain:

$$\cot\left(\frac{Y_c d}{\sqrt{K_3}}\right) = \frac{\sqrt{K_3}}{A} Y_c \tag{19}$$

This is the equation relating the critical values of I, B and confinement ratio at the threshold of the transition from the cholesteric - like alignment to the nematic-like alignment.

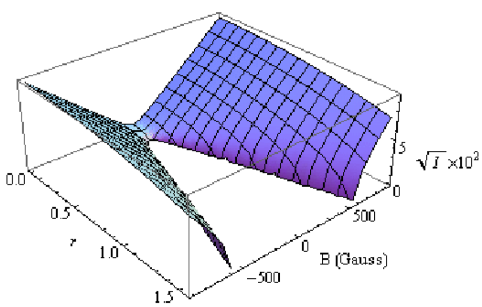
3. Discussion

a) In the case of the rigid anchoring $A \to \infty$, $\cot\left(\frac{Y_c d}{\sqrt{K_3}}\right) \to 0$ and

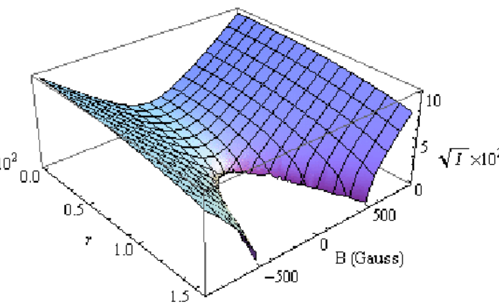
$$\frac{Y_c d}{\sqrt{K_3}} = \frac{\pi}{2} \tag{20}$$

Eq. (20) is now similar to Eq. (18) from ref. [25]

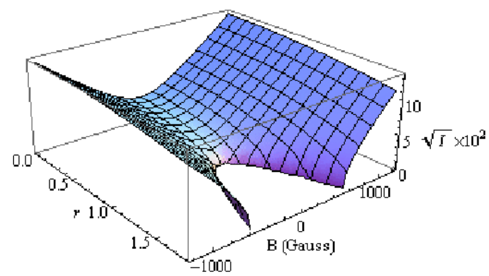
b) If both anisotropies are positive, the magnetic external field and the electric field of the laser beam have opposite effects. While the magnetic field performs the homeotropic alignment, the laser breaks it. This can be used in laser writing memory device.



a) $A = 10^{-7}$ J/m^2



b) $A = 5\times10^{-7}$ J/m^2



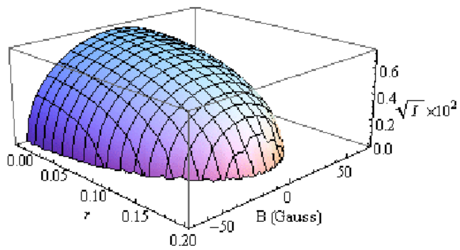
c) $A \to \infty$

Fig. 2 Phase transition (FC-FN) diagram for several values of anchoring constant A when $\chi_a > 0$

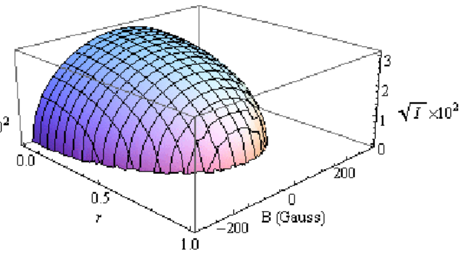
c) If the magnetic anisotropy is negative $\chi_a < 0$ the magnetic and the laser fields will have the same tendency: to perform the FC - FN transition. In both cases ($\chi_a > 0$ and $\chi_a < 0$) Eq. (19) represents a surface in 3D system of coordinates: $\sqrt{I}$, B, r

We have used the following material parameters: $\varepsilon_{\|} = 2.89$; $\varepsilon_{\perp} = 2.25$; $f = 10^{-3}$; $\chi_a = 7\times10^{-7}$; $W = 5\times10^{-10}$ N/m; $a = 3\times10^{-9}$ m; $K_1 = 17{,}2\times10^{12}$ N;

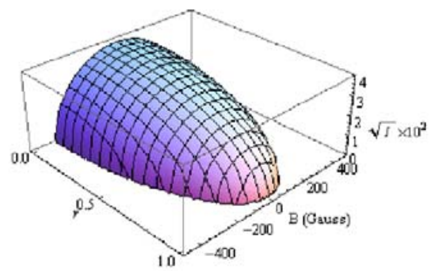
$K_2 = 7,5\times10^{-12}$ N; $K_3 = 17,9\times10^{-12}$ N and $d = 200$ μm and we have plotted the surfaces described by Eq. (19) for $\chi_a > 0$ (Fig. 2) and $\chi_a < 0$ (Fig. 3)



a) $A = 10^{-7}$ J/m^2



b) $A = 5\times10^{-7}$ J/m^2



c) $A \to \infty$

Fig. 3 Phase transition (FC-FN) diagram for several values of anchoring constant A when $\chi_a < 0$.

We can observe that, for $\chi_a > 0$ the surfaces are open, while for $\chi_a < 0$ the surfaces are closed. The values of the two fields "inside" this closed surface do not perform the transition: the FC arrangement is metastable. The values of both fields "outside" this surface perform the FC - FN transition. The critical values of both fields are controlled by the ratio r. As the parameter A increases, the critical fields and the critical ratio r increase. For all finite values of A the surfaces close for $r < 1$; this means either that the FC must have a great pitch or that the cells must be thicker. A similar results was obtained in [38].

d) The final results of our calculation (Eq. (19) does not contain the parameter ξ introduced in Eq. (2). The same result was obtained in [33] and [13], therefore for this level of the discussion the initial formula of Rapini and Papoular is sufficient.

4. **Conclusions**

In this paper we have studied analytically the transition from the cholesteric to the nematic arrangement of a confined liquid crystal matrix containing ferromagnetic particles and subjected simultaneously to a magnetic field and an optical beam. Unlike our previous studies, we have assumed a finite anchoring energy of the molecules of the liquid crystal to the cell walls. For the anchoring energy we have used the formula proposed by Young and all. We have obtained a correlation between the critical fields, the confinement ratio and the anchoring strength at the threshold of the transition FC-FN. Based on this correlation and using some practical values for the material and device parameter we have plotted phase diagrams in a system of $\sqrt{I}$, B , r coordinates for two cases: the positive and the negative values of the magnetic anisotropy. We have used several anchoring strengths and have concluded that the fields needed for the transition increase as the anchoring strength increases.

R E F E R E N C E S

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