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One-Dimensional Structures in Ferrocholesteric Film with Weak Homeotropic Anchoring on the Layer Boundaries

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We consider one-dimensional structures of a ferrocholesteric film with weak anchoring of homeotropic type on the boundaries of the layer. We assume that the external magnetic field is oriented normal to the plane of the layer and diamagnetic anisotropy of cholesteric liquid crystal matrix is positive. We find that two kinds of ordering can exist in the layer with weak homeotropic anchoring on its boundaries: ferronematic (FN) and ferrocholesteric (FC) states. It is shown that variation of the surface anchoring strength between the cholesteric molecules and layer boundaries can induce the FN-FC transition, and can lead to a change of the phase transition character from the first order to second order one. The critical parameters of the transition between FC and FN states are determined as a function of FC material parameters. At present study of orientational and magnetic properties of FN-FC transition the so-called segregation effect are taken into account: the concentration redistribution of the magnetic admixture under the influence of a magnetic field.

Keywords: ferrocholesteric; ferronematic; weak anchoring

INTRODUCTION

Ferrocholesterics (FC) represent dilute magnetic suspensions on the base of cholesteric liquid crystal (CLC). The solid phase of FC consists of needle-like ferromagnetic particles with the length-to-width ratio of 10 : 1. The magnetic moment $\mu = M_s v m$ of each particle is rigidly linked to main axis m of the needle-like grain due to the magnetostatic shape anisotropy, here M_s is the saturation magnetization of a grain, v is the grain volume, $m^2 = 1$. We assume that magnetic particles are embedded into the CLC-matrix in such way that their long axes m are oriented parallel to the local director n of the CLC-matrix, and on the surfaces of ferroparticles the rigid anchoring of the planar type takes place. The volume fraction f of the magnetic admixture is assumed to be very small ($f \leq 10^{-4}$), therefore the

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solid phase of FC can be treated as the ideal gas of non-interacting magnetic particles. The presence of small amount of ferroparticles doesn't change a character of orientational ordering of this dispersion media, therefore FC, having strong enough magnetic properties, in the rest of features behaves like usual CLC. FC is characterized by two mechanisms of interaction with the external magnetic field: quadrupolar one (regarded with the influence of magnetic field on the diamagnetic CLC-matrix) and dipolar (caused by the influence of magnetic field on ferroparticles) mechanism, whereas the pure CLC has only quadrupolar mechanism. We assume that CLC-matrix has positive anisotropy $\chi_a > 0$ of diamagnetic susceptibility. In that case, the director \mathbf{n} of the CLC-matrix and the long axes of magnetic particles tend to orient along the magnetic field direction.

In this paper we study one-dimensional FC structures in the layer with soft homeotropic anchoring between the molecules and the boundaries of the layer and the influence of the anchoring strength, the thickness of the layer and magnetic field strength on the ferrocholesteric - ferronematic transition.

FREE ENERGY OF A FERROCHOLESTERIC

The z -axis is chosen to be perpendicular to the layer surfaces, so $z = 0$ corresponds to the bottom boundary, and $z = d$ - to the upper one, here d is the layer thickness. The external magnetic field is directed along z axis: $\mathcal{H} = (0, 0, \mathcal{H})$. Under the action of a field \mathcal{H} the distorted director field is assumed to be

$$\mathbf{n} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta), \quad \mathbf{m} \parallel \mathbf{n}. \quad (1)$$

The tilt angle θ is measured from the layer normal. The functional of FC free energy can be written in the following form [1, 2]:

$$\frac{\mathcal{F}}{Sq_0K_{22}} = \int_0^D dz \left\{ \frac{[(\theta')^2(1 - \Delta_1 \sin^2 \theta) + (\phi')^2(1 - \Delta_2 \sin^2 \theta)]}{2(1 - \Delta_2)} - \phi' \sin^2 \theta \right\} - \frac{H^2}{2} \cos^2 \theta - \frac{H\xi f}{f_0} \cos \theta + \frac{\kappa f}{f_0} \ln f \Bigg\} - \frac{1}{S} \oint \frac{w}{2} \cos^2 \theta dS, \quad (2)$$

here we have introduced the notations for dimensionless layer thickness $D = q_0 d$ and dimensionless magnetic field strength $H = \mathcal{H}/q_0 \sqrt{\chi_a/K_{22}}$. The dimensionless parameters defined here are expressed as $\Delta_1 = (K_{33} - K_{11})/K_{33}$, $\Delta_2 = (K_{33} - K_{22})/K_{33}$, $\xi = M_s f_0 / (q_0 \sqrt{K_{22} \chi_a})$, $\kappa = k_B T f_0 / (v K_{22} q_0^2)$, $w = W / (q_0 K_{22})$. In the above formulas, K_{ii} are Frank elastic constants, $q_0 = 2\pi/p_0$ is the wave vector of intrinsic helical pitch p_0 in a zero field,

k_B is the Boltzmann constant, T is a temperature, W is a surface energy density of the CLC molecules anchoring with the layer boundaries, S is the surface area of the layer boundary. The prime in (2) denotes a derivative with respect to dimensionless coordinate $\tilde{z} = q_0 z$, and the sign tilde over the coordinate \tilde{z} will be omitted below. The term in square brackets in Eq.(2) is the energy density, caused by the elastic distortion of the CLC-matrix director field. The second and third terms in (2) describe the quadrupolar and dipolar mechanisms of FC interaction with the external magnetic field, respectively. The fourth term in (2) is the mixing entropy contribution for the dilute suspension of non-interacting magnetic particles. The last integral term represents the surface interaction of CLC molecules with the layer boundaries written in Rapini form. We consider the case when the anchoring energy on both layer boundaries is equal and $W > 0$.

Minimization of the total free energy (2) of FC over $f(z)$, $\theta(z)$ and $\phi(z)$, under the condition of particles number constancy in the layer $\int f dV = Nv$, leads to the following equations, describing the equilibrium state of FC:

$$f = f_0 Q \exp \left\{ \frac{\xi H}{\kappa} \cos \theta \right\}, \quad Q^{-1} = D^{-1} \int_0^D \exp \left\{ \frac{\xi H}{\kappa} \cos \theta \right\} dz, \quad (3)$$

$$z = \int_{\theta_S}^{\theta(z)} \sqrt{A(\theta_m, \theta)} d\theta, \quad \phi(z) - \phi(0) = (1 - \Delta_2) \int_{\theta_S}^{\theta(z)} \frac{\sqrt{A(\theta_m, \theta)}}{1 - \Delta_2 \sin^2 \theta} d\theta, \quad (4)$$

here

$$\begin{aligned} A(\theta_m, \theta) = & (1 - \Delta_2 \sin^2 \theta_m)(1 - \Delta_2 \sin^2 \theta)(1 - \Delta_1 \sin^2 \theta) / \left\{ (1 - \Delta_2)^2 \times \right. \\ & \times (\sin^2 \theta_m - \sin^2 \theta) \left[1 - \frac{H^2}{(1 - \Delta_2)} (1 - \Delta_2 \sin^2 \theta_m)(1 - \Delta_2 \sin^2 \theta) \right] + \\ & + 2\kappa Q (1 - \Delta_2)(1 - \Delta_2 \sin^2 \theta_m)(1 - \Delta_2 \sin^2 \theta) \left[\exp \left\{ \frac{\xi H}{\kappa} \cos \theta_m \right\} - \right. \\ & \left. \left. - \exp \left\{ \frac{\xi H}{\kappa} \cos \theta \right\} \right] \right\}. \end{aligned} \quad (5)$$

The values of angles $\theta_S \equiv \theta(z=0)$ and $\theta_m \equiv \theta(z=D/2)$ in (4) and (5) are obtained from following equations:

$$\int_{\theta_S}^{\theta_m} \sqrt{A(\theta_m, \theta)} d\theta = \frac{D}{2}, \quad \frac{(1 - \Delta_2)^2 A(\theta_m, \theta_S) \sin^2 2\theta_S}{4(1 - \Delta_1 \sin^2 \theta_S)^2} = \frac{1}{w^2}, \quad (6)$$

here the last expression results from the weak homeotropic boundary conditions on the boundaries of the layer. Eqs.(3) describe the concentration

redistribution of the magnetic admixture induced by magnetic field (the segregation effect [1]), the angles $\theta = \theta(z)$ and $\phi = \phi(z)$ are determined by Eqs.(4). Taking into account Eqs.(1) and (3), we obtain the equation for the reduced magnetization $M = \mathcal{M}/(M_s f_0) = mf/f_0$, this expression enables us to determine the components of the reduced magnetization, averaged over the thickness of the layer:

$$\langle M_x \rangle = \langle M_y \rangle = 0, \quad \langle M_z \rangle = \frac{2Q}{D} \int_{\theta_s}^{\theta_m} \cos \theta \exp \left\{ \frac{\xi H}{\kappa} \cos \theta \right\} \sqrt{A(\theta_m, \theta)} d\theta. \quad (7)$$

The continuum theory based on Eqs.(3) - (7) predicts the existence of two phases. The first one corresponds to uniform homeotropic ferronematic (FN) phase: $\mathbf{n} = (0, 0, 1)$ and $\mathbf{m} = (0, 0, 1)$. The second one is the helical FC phase, with the helix axis oriented along the z axis. It is the so-called translationally invariant configuration (TIC) [3]-[5]. We assume that the initial state of the system corresponds to uniform homeotropic FN phase, i.e. the main axes of the CLC molecules and magnetic moments of ferroparticles are oriented perpendicular to the layer plane. In that case, the sample magnetization is maximum $\langle M_z \rangle = 1$ and the concentration of magnetic particles is constant in the whole bulk $f = f_0 = Nv/V$. Let consider a distortion of this FN order and further phase transition between FN and FC states.

THE FERRONEMATIC-FERROCHOLESTERIC PHASE TRANSITION

The appearance of FC ordering is accompanied by small distortions of the initial FN state, i.e. the tilt angle $\theta \ll 1$, therefore the critical parameters of the phase transition between FN and FC states can be obtained from the zero order of the power series expansion of Eqs.(3) and (6) in θ_m :

$$D = 2 \arctan \left(w \sqrt{\frac{1 - \Delta_2}{1 - \Delta_2 - H^2 - \xi H}} \right) / \sqrt{(1 - \Delta_2)(1 - \Delta_2 - H^2 - \xi H)}. \quad (8)$$

There are two parameters: the layer thickness D and the magnetic field strength H , the variation of which can cause the phase transition between FN and FC states. The expression (8) for given values of material parameters Δ_1 , Δ_2 , ξ , w , and magnetic field strength H , determines the critical value D_c of the layer thickness, such that the FC ordering can be possible for $D > D_c$. On the other hand, if the layer thickness D is fixed and the magnetic field is an external variable parameter, then Eq.(8) determines the critical value of the magnetic field strength H_c , and the FN - FC phase

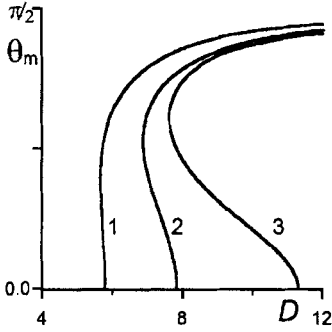


FIGURE 1: Variation of θ_m as a function of the layer thickness D for $\Delta_1 = -0.2$, $\Delta_2 = 0.3$, $w = 5$, $H = 0.5$ and $\kappa = 0.1$. Curve 1 - $\xi = 0.2$, 2 - $\xi = 0.5$, 3 - $\xi = 0.7$.

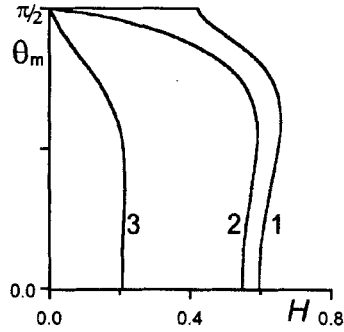


FIGURE 2: Variation of θ_m as a function of the strength of magnetic field H for $\Delta_1 = -0.2$, $\Delta_2 = 0.3$, $w = 1.4$, $D = 4.5$ and $\kappa = 0.1$. Curve 1 - $\xi = 0.0$, 2 - $\xi = 0.1$, 3 - $\xi = 1.5$.

transition takes place at $H < H_c$. In the limit of rigid anchoring $w \rightarrow \infty$ and $\xi = 0$ (pure CLC) the Eq.(8) gives a known from [3] - [6] critical value of the layer thickness for the transition between the homeotropic nematic and cholesteric states.

As it is follows from the equations of state, the phase transition between FN and FC states can be both first order transition and second order one. In the case of the first order transition the FC state occurs at $D = D_c$ as the layer thickness increases, but the inverse FC - FN phase transition, as the thickness of the layer decreases, takes place at some value $D < D_c$, i.e. there is a hysteresis behavior (see curves 2 and 3 in Fig.1). The similar situation occurs in the case of the first order transition, which arise from the variation of the magnetic field strength (see curves 2 and 3 in Fig.2): the FN - FC transition takes place at $H = H_c$ as the magnetic field strength decreases, but the inverse FC - FN transition happens at $H > H_c$ (the value of H_c is determined from Eq.(8) as a function of FC material parameters and layer thickness D). The dependence of angle θ_m on the layer thickness D (Fig.1) and on the magnetic field strength H (Fig.2) is a many-valued function in the case of the first order transition, and the area of this multiplicity determines the hysteresis region. In the case of the second order transition between FC and FN states, the transition occurs at $D = D_c$ (for the transition caused by a variation of thickness) or at $H = H_c$ (for the transition forced by

variation of the magnetic field). The conditions of the hysteresis behavior can be obtained from the second order of the power series expansion of Eqs.(3) and (6) in small θ_m . For the case of phase transition forced by the variation of the layer thickness, the condition of hysteresis behavior has the following form

$$f_D = \frac{\pi}{16\sqrt{1-\Delta_2}(1-\Delta_2-H^2-\xi H)^{3/2}} \left\{ 4(\Delta_2-1)[3\Delta_2+\Delta_1-1+H^2\frac{(1-\Delta_1)}{(1-\Delta_2)}] - \frac{\xi^2 H^2}{\kappa} - H\xi(1-4\Delta_1) \right\} < 0, \quad (9)$$

and for the case of phase transition caused by the variation of magnetic field strength the condition can be written as

$$f_H = \left\{ \left[H_c^2 \left[\frac{\xi^2}{\kappa} + 4(1-\Delta_1) \right] + \xi H_c(1-4\Delta_1) - 4\Delta_2[3\Delta_2+\Delta_1-4] - 4(1-\Delta_1) \right] \right\} / 8(\xi + 2H_c) > 0. \quad (10)$$

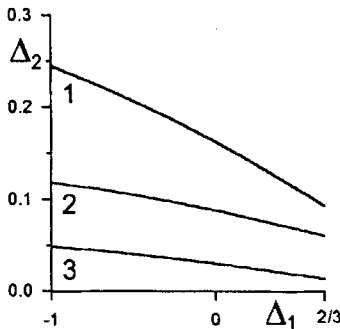


FIGURE 3: The single sign areas of functions f_D on the parameters field Δ_1 and Δ_2 for $H = 0.5$ $w = 1$, and $\kappa = 0.1$. Curve 1 - $\xi = 0.5$, 2 - $\xi = 1.0$, 3 - $\xi = 1.2$.

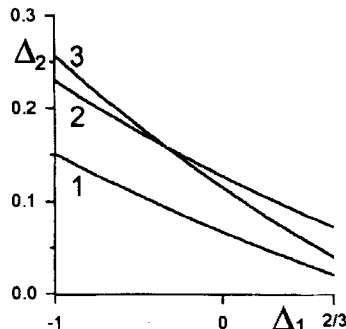


FIGURE 4: The single sign areas of functions f_H on the parameters field Δ_1 and Δ_2 for $D = 5$ $w = 1$, and $\kappa = 0.1$. Curve 1 - $\xi = 0.0$, 2 - $\xi = 0.5$, 3 - $\xi = 2.5$.

Here, owing to complicated form of functions f_D and f_H for the case of weak anchoring, the expressions (9) and (10) are written for the limiting case of rigid anchoring, when $w \rightarrow \infty$. For the particular case of pure cholesteric ($\xi = 0$) the condition (9) gives the form: $(3\Delta_2 + \Delta_1 - 1) < 0$,

which coincides with one obtained in [4]. The single sign areas of functions f_D and f_H , for the case of weak anchoring, are depicted in Figs.3 and 4. Each of the curves divides the plane of the parameters into two regions: in the region above the curve the phase transition between FN and FC states is the first order one; the second order transition takes place for the value of the parameters corresponded to the region, which is below the curve. The curves themselves determine the coordinates of the so-called three-critical points corresponded to the change of the phase transition character.

The appearance of TIC ferrocholesteric ordering distorts the initial uniform FN state, and as it is mentioned above, consequently the average sample magnetization $\langle M_z \rangle$ decreases. As it can be seen from Fig.5, this decreasing can happen both continuous way (the second order transition, see curve 1) and discontinuous one (the first order transition, see curves 2 and 3). The many-valued region of $\langle M_z \rangle$ as a function of the thickness D determines the hysteresis area. The breaking of the FN ordering causes the non-uniformity of magnetic admixture distribution in the layer and as a result, the areas of high concentration of ferroparticles appear near the boundaries of the layer (see Fig.6). The magnetic particles, under the influence of the external magnetic field, tend to accumulate in those parts of the layer, where their energy has minimum (the segregation effect). The intensity of this process, as it can be seen from Fig.6, grows up if the magnetic field strength increases, because the ferroparticles tend to be oriented along the magnetic field direction.

THE PLANAR ORDERING

In the absence of the magnetic admixture ($\xi = 0$) or at zero magnetic field ($H = 0$) Eqs.(3)-(6) admit the solution $\theta(z) \equiv \pi/2$, describing the cholesteric ordering in the whole bulk: the helical axis is oriented along the z axis, the helical pitch is equal to the one of the undisturbed CLC $p_0 = 2\pi/q_0$. The appearance of the planar ordering is a consequence of weak anchoring on the layer boundaries. The planar state in the whole cell's bulk can't be formed in the limit $w \rightarrow \infty$, so the planar ordering, in this case, is formed in the central part of the layer just for $D \rightarrow \infty$. The threshold parameters at which the appearance of the planar ordering becomes possible, can be obtained in zero order of the power series expansion in $(\pi/2 - \theta_m)$ of Eqs.(3)-(6). The threshold value of the anchoring energy is determined by the following expression:

$$w_s = \frac{\sqrt{(1 - \Delta_1)[1 - H^2(1 - \Delta_2)]}}{(1 - \Delta_2)} \tanh \left\{ \frac{D}{2} \sqrt{\frac{1 - H^2(1 - \Delta_2)}{1 - \Delta_1}} \right\}. \quad (11)$$

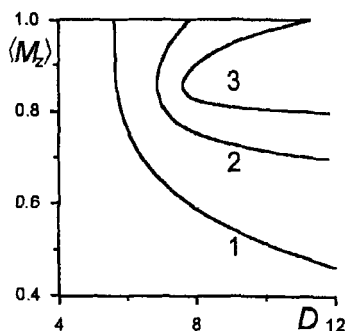


FIGURE 5: Average magnetization of FC versus the layer thickness for $\Delta_1 = -0.2$, $\Delta_2 = 0.3$, $H = 0.5$, $w = 5$, and $\kappa = 0.1$. Curve 1 - $\xi = 0.2$, 2 - $\xi = 0.5$, 3 - $\xi = 0.7$.

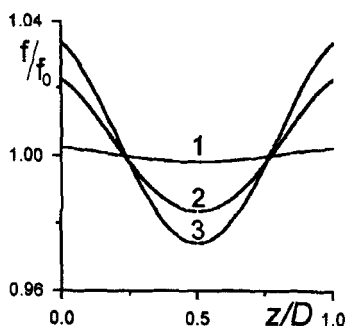


FIGURE 6: Concentration of the magnetic admixture versus the coordinate z for $\Delta_1 = -0.2$, $\Delta_2 = 0.3$, $D = 4.5$, $\xi = 0.5$, $w = 5$, and $\kappa = 0.1$. Curve 1 - $H = 0.01$, 2 - $H = 0.1$, 3 - $H = 0.3$.

For $w \geq w_*$ TIC cholesteric ordering takes place. The appearance of planar ordering in pure CLC is depicted in Fig.2 (curve 1). The magnetic field strength, which induces this type of ordering, can be determined from Eq.(11), as a function of the layer thickness D and material parameters w , Δ_1 , and Δ_2 .

Acknowledgments

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