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Magnetic Freedericksz Transition in Ferronematic Layer Under Shear Flow

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Within the framework of continuum theory we analyze the Freedericksz transition in a ferronematic layer under the combined influence of magnetic field and shear flow. The stationary equations for concentration of magnetic particles, planar director and magnetization fields are obtained. We perform numerical calculations of the angles of the director and the magnetization rotation for different values of magnetic field strength, reactive parameter and Ericksen number. We show that in the general case shear flow "smoothes" the Freedericksz transition in ferronematics. For ferronematics with non-flow-aligning nematic matrix we calculate stationary orientations of the director and magnetization in the shear plane.

Keywords Ferronematic; Freedericksz transition; magnetic field; shear flow

1. Introduction

Soft composite materials are of great scientific interest due to novel properties which their separate components do not have. More complex behavior of such materials is caused by additional degrees of freedom and new mechanisms of interaction with external fields. One of them is suspension of magnetic particles in nematic liquid crystals (NLC) named ferronematic (FN). FNs combine high fluidity with strong magnetic properties and belong to the unique class of soft magnetic materials with controlled properties. After Brochard and de Gennes classical work [1] many attempts to introduce ferromagnetic particles in liquid crystals were undertaken in order to increase their magnetic susceptibility, and later these suspensions have been synthesized. In recent years they have been intensively investigated both experimentally and theoretically [2–29]. However, for a long time the influence of flow on ferronematic liquid crystals remained not investigated. Recently, in Refs. [25,26,30] the combined influence of shear flow and magnetic field on the orientational phases in nematics and unconfined FN has been analyzed.

In the present paper we analyze the influence of shear flow and uniform magnetic field on the Freedericksz transition in a ferronematic layer. Within the framework of generalized continuum Ericksen-Leslie theory [12,25,26,31] we obtain the stationary integral equations describing the behavior of ferronematic orientational

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structure under the combined action of shear flow and uniform magnetic field. We consider the case in which the magnetic field is directed in the plane of shear flow. On the boundaries of the ferronematic layer the conditions of rigid planar coupling are imposed. The coupling between the magnetic particles and the director is supposed to be soft and homeotropic.

We perform numerical calculations of the angles of the director and the magnetization rotation for different values of magnetic field strength and Ericksen number. Both flow-aligning and non-flow-aligning liquid crystal matrixes with rod-like molecules are considered.

2. Continuum Theory of Ferronematics

Considering soft coupling between the director and magnetic particles the bulk free energy density of a ferronematic in magnetic field can be written in the following form [1,8]

$$F = F_1 + F_2 + F_3 + F_4 + F_5,$$

$$F_1 = \frac{1}{2} \left[K_1 (\nabla \cdot \mathbf{n})^2 + K_2 (\mathbf{n} \cdot \nabla \times \mathbf{n})^2 + K_3 (\mathbf{n} \times \nabla \times \mathbf{n})^2 \right],$$

$$F_2 = -M_s f \, \mathbf{m} \cdot \mathbf{H}, \quad F_3 = -\frac{1}{2} \chi_a (\mathbf{n} \cdot \mathbf{H})^2,$$

$$F_4 = \frac{k_B T}{v} f \ln f, \quad F_5 = \frac{w}{d} f (\mathbf{n} \cdot \mathbf{m})^2.$$

$$(1)$$

Here K_1 , K_2 , K_3 are the splay, twist, and bend elastic modules of NLC (Frank constants), \boldsymbol{n} is the director, M_s is the saturation magnetization of the ferroparticle material, f is the volume fraction of the particles in a suspension, \boldsymbol{m} is the unit vector directed along magnetization of the suspension, χ_a is the anisotropy of a magnetic susceptibility (we assume that $\chi_a > 0$), v is the volume of a ferroparticle, k_B is the Boltzmann constant, d is the transverse diameter of a ferroparticle, T is the temperature, w is the surface energy density of the NLC molecules coupling with magnetic particles (we assume that w > 0).

We consider FN with low volume fraction f << 1 of ferroparticles and therefore we neglect the interparticle magnetic dipole-dipole interactions in a suspension. The first term F_1 represents the bulk free energy density of elastic deformations of NLC director field (the Oseen-Frank potential). The second F_2 and the third F_3 contributions characterize the interactions of magnetic moments $\mu = M_s v$ of particles (the dipole mechanism of interaction) and diamagnetic NLC-matrix (the quadrupole mechanism of interaction) with an external magnetic field H, respectively. The fourth term F_4 is the contribution of the mixing entropy of the ideal ferromagnetic particle solution. The last term F_5 determines the surface coupling energy of the magnetic particles with NLC-matrix (the coupling energy). We consider FN, which has homeotropic coupling conditions of NLC with magnetic particles $(m \perp n)$ due to w > 0. Under these conditions at H = 0 the director n is normal to the plane where the magnetic moments μ of the particles are placed. If a FN is prepared from an isotropic suspension by cooling in the absence of external fields, the macroscopic magnetization $M = M_s f m$ of the sample is absent. However, it is possible to take off the degeneration of the magnetic moments orientation in a suspension at the transition through the clearing point in the magnetic field. This FN possesses the uncompensated magnetization M in the absence of a magnetic field. Therefore, the description in terms of magnetization is valid for such FN even in zero magnetic field. For this case the coupling energy F_5 is minimized at $m \perp n$.

According to continuum theory [12,31] the ferronematodynamic equations, presenting the balance of forces acting on the fluid and the incompressibility condition, can be written as

$$\rho \frac{d\mathbf{v}}{dt} = \mathbf{\nabla} \cdot \mathbf{\sigma},\tag{2}$$

$$\nabla \cdot \mathbf{v} = 0. \tag{3}$$

Here ρ , v, and σ are the mass density, the velocity, and the total stress tensor of NLC carrier (the embedding of ferroparticles in NLC carrier produce negligible changes in density and stress tensor due to low volume fraction f << 1 of magnetic particles in ferronematic), respectively; $d/dt \equiv \partial/\partial t + v \cdot \nabla$.

The stress tensor σ in Eq. (2) is determined as a sum

$$\sigma = \sigma' + \sigma^{(e)},\tag{4}$$

where viscous stress tensor σ' has the form

$$\sigma'_{ki} = \alpha_1 n_k n_i n_l n_m A_{lm} + \alpha_2 n_k N_i + \alpha_3 n_i N_k + \alpha_4 A_{ki} + \alpha_5 n_k n_l A_{li} + \alpha_6 n_i n_l A_{lk},$$

here the summation over repeated indices is implied. The vector $\mathbf{N} = d\mathbf{n}/dt - \mathbf{\omega} \cdot \mathbf{n}$ represents the rate of change of the director \mathbf{n} relative to the background liquid, $A_{ik} = (\nabla_k v_i + \nabla_i v_k)/2$ and $\omega_{ik} = (\nabla_k v_i - \nabla_i v_k)/2$ are symmetric and antisymmetric parts of the velocity gradients tensor, respectively. The six viscosity coefficients α_s are called Leslie coefficients.

The elastic part of stress tensor (4) known as Ericksen tensor $\sigma^{(e)}$ is given by

$$\sigma_{ki}^{(e)} = -p\delta_{ki} - \frac{\partial F}{\partial(\nabla_k n_l)}\nabla_i n_l,$$

where p is the pressure, δ_{ki} is the Kronecker symbol.

The dynamic equation for the director n [31] has the form

$$\boldsymbol{h}^{(n)} = \gamma_1 \boldsymbol{N} + \gamma_2 \boldsymbol{n} \cdot \boldsymbol{A},\tag{5}$$

where the rotary viscosity coefficients of nematic are given by $\gamma_1 = \alpha_3 - \alpha_2$, $\gamma_2 = \alpha_2 + \alpha_3$. The coefficient γ_1 characterizes the viscous torque associated with an angular velocity of the director, while γ_2 gives the contribution to this torque due to a shear velocity of NLC.

The dynamic equation for the unit magnetization vector m [12] has the form

$$\boldsymbol{h}^{(m)} = (\gamma_{1p}\boldsymbol{M} + \gamma_{2p}\boldsymbol{m} \cdot \boldsymbol{A})f \tag{6}$$

where γ_{1p} and γ_{2p} are the rotary viscosity coefficients of the ferroparticles in a suspension. The vector $\mathbf{M} = d\mathbf{m}/dt - \mathbf{\omega} \cdot \mathbf{m}$ characterizes the rate of change of the magnetization vector \mathbf{m} relative to the background NLC carrier.

The molecular fields $h^{(n)}$ and $h^{(m)}$ in Eqs. (5) and (6) are given by

$$h_i^{(n)} = -\frac{\partial F}{\partial n_i} + \nabla_k \frac{\partial F}{\partial (\nabla_k n_i)}, h_i^{(m)} = -\frac{\partial F}{\partial m_i} + \nabla_k \frac{\partial F}{\partial (\nabla_k m_i)}.$$

The director n and magnetization m are the unit vectors, therefore the variation of the free energy F [see Eq. (1)] should be produced under auxiliary conditions $n^2 = 1$ and $m^2 = 1$ by Lagrange coefficients method.

The diffusion equation for the ferroparticles in NLC carrier (the particle-number conservation law) [12] can be written in the following form

$$\frac{\partial f}{\partial t} + \nabla \cdot (V_f f) = 0. \tag{7}$$

Here $V_f = -B_t \nabla (vF^{(m)}/f)$ is the velocity of ferroparticles relative to the NLC carrier, B_t is the diffusion coefficient, v is the volume of a particle, $F^{(m)} = F_2 + F_4 + F_5$ are the terms of the total free energy (1) caused by the embedding of ferroparticles into the NLC carrier.

3. Basic Equations for Ferronematic Under Shear Flow

We consider a ferronematic layer of thickness D sandwiched between two parallel plates moving relative to each other with the constant velocity U (see Fig. 1). In the middle of the layer we place the origin of the rectangular coordinate system with x axis directed along the trajectory of moving plate and the z axis directed perpendicular to the plates. The bottom plate in the given system of coordinates is motionless. We consider rigid planar coupling of the director n (the unit vector determining

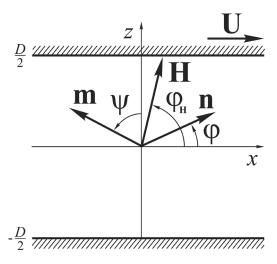


Figure 1. Orientation of a ferronematic layer in the magnetic field H.

the direction of preferred orientation of nematic molecules axes) on the boundaries of a layer; the anisotropy of diamagnetic susceptibility χ_a is supposed to be positive. Let the homogeneous magnetic field be enclosed to the ferronematic in the shear plane $\mathbf{H} = H(\cos\varphi_H, 0, \sin\varphi_H)$. Then the director \mathbf{n} and the unit vector of magnetization \mathbf{m} can be written as:

$$\mathbf{n} = (\cos \varphi(z), 0, \sin \varphi(z)), \qquad \mathbf{m} = (-\sin \psi(z), 0, \cos \psi(z)). \tag{8}$$

We use the linear approximation of a velocity field in the layer [30]:

$$\mathbf{v} = \left[\frac{U}{D} \left(z + \frac{D}{2} \right), \ 0, \ 0 \right].$$

We choose the thickness D of the layer as the unit of length and introduce the following dimensionless parameters:

$$h = HD\sqrt{\frac{\chi_{a}}{K_{1}}}, \quad b = \frac{M_{\sqrt{f}D}}{\sqrt{\chi_{a}K_{1}}}, \quad k = \frac{K_{3}}{K_{1}},$$

$$\lambda = -\frac{\gamma_{2}}{\gamma_{1}}, \quad a_{1p} = \frac{\gamma_{1p}}{\gamma_{1}}, \quad a_{2p} = \frac{\gamma_{2p}}{\gamma_{1}},$$

$$Er = \frac{U\gamma_{1}D}{2K_{1}}, \quad \sigma = \frac{w\bar{f}D^{2}}{dK_{1}}, \quad \bar{f} = \frac{N\nu}{V}, \quad \varsigma = \frac{\bar{f}k_{B}TD^{2}}{K_{1}\nu}.$$
(9)

Here h is the dimensionless magnetic field strength, b characterizes the mechanism of magnetic field influence on FN [9]. Also we define the parameter of elastic anisotropy k, the reactive parameter λ , the coefficients a_{1p} and a_{2p} representing the ratio of rotary viscosity coefficients of ferroparticles and NLC; σ characterizes the energy of magnetic particles coupling with the NLC-matrix, ς is the segregation parameter, and Er is the Ericksen number.

From equations of ferronematodynamics (2), (3), (5)–(7) in the case of $\partial n/\partial t \equiv 0$ we obtain the set of stationary equations for the director n and magnetization m, and the distribution function f of magnetic particles. Using the dimensionless variables (9) these equation can be written as

$$K(\varphi)\frac{d^2\varphi}{dz^2} + \frac{1}{2}\frac{dK}{d\varphi}\left(\frac{d\varphi}{dz}\right)^2 - \frac{1}{2}h^2\sin 2(\varphi - \varphi_H) - \sigma QE(\varphi, \psi)\sin 2(\varphi - \psi) - Er(1 - \lambda\cos 2\varphi) = 0,$$
(10)

$$bh\cos(\psi - \varphi_H) - \sigma\sin 2(\varphi - \psi) + \bar{f}Er(a_{1p} - a_{2p}\cos 2\psi) = 0, \tag{11}$$

$$f = \bar{f}QE(\varphi, \psi), \qquad Q^{-1} = \int_{-1/2}^{1/2} E(\varphi, \psi) dz,$$
 (12)

with rigid planar boundary conditions

$$\varphi(-1/2) = \varphi(1/2) = 0. \tag{13}$$

Here the following notations are introduced:

$$K(\varphi) = \cos^2 \varphi + k \sin^2 \varphi, \qquad E(\varphi, \ \psi) = \exp\left\{-\frac{bh}{\varsigma}\sin(\psi - \varphi_H) - \frac{\sigma}{\varsigma}\sin^2(\varphi - \psi)\right\}. \tag{14}$$

The set of coupled equations (1)–(7) describing the dynamics of ferronematic shows that shear flow influences the director, the magnetic particles and the density of ferroparticles [see Eq. (7)]. In the stationary case Eq. (7) gives the distribution of ferroparticles concentration in the form (12). As it is seen from Eq. (12), shear-induced-inhomogeneity in m and n leads to an inhomogeneity in the density of the ferroparticles. This effect is known as segregation effect (see Ref. [1]).

Let's make the estimations of dimensionless quantities (9), using typical values of material parameters for nematic liquid crystals and magnetic particles [12,28, 29]. Assuming for a ferronematic based on the liquid crystal 5CB $\chi_a \sim 10^{-7} \text{SGSE}$ units, K_1 , $K_3 \sim 10^{-6} \text{dynes}$, $T \sim 300 \text{K}$, γ_1 , $\gamma_2 \sim 10^{-1} \text{ P}$, γ_{1p} , $\gamma_{2p} \sim 1 \text{ P}$, $\bar{f} \sim 10^{-6}$, $M_s \sim 10^2 \text{ G}$, $w \sim 10^{-2} \text{ dyn/cm}$, $d \sim 10^{-5} \text{cm}$ and supposing the layer thickness $D \sim 10^{-2} \text{cm}$, $U \sim 10^{-2} \text{cm/s}$, we obtain $\lambda \sim 1$, a_{1p} , $a_{2p} \sim 10$, $Er \sim 10$, $\sigma \sim 10^{-1}$, $b \sim 10$ and $\varsigma \sim 10^{-1}$. As is seen from these estimations $\bar{f}Er < \sigma$, so it is possible to neglect from Eq. (11) the term characterizing the influence of flow on the orientation of magnetic particles.

Uniform solutions of Eqs. (10)–(12) have been analyzed earlier in Refs. [25,26]. In the present paper we consider non-uniform solutions for the director and magnetization. We integrate Eqs. (10)–(12) over the layer thickness with the boundary conditions (13). Then the stationary solutions describing the disturbed states of the director and the magnetization, and concentration distribution of magnetic particles in the middle of the layer, can be found from the following set of equations:

$$\frac{1}{2} = \pm \int_0^{\varphi_0} \left[\frac{K(\varphi)}{\Phi(\varphi, \psi)} \right]^{\frac{1}{2}} d\varphi, \tag{15}$$

$$\frac{1}{2Q} = \pm \int_0^{\varphi_0} E(\varphi, \psi) \left[\frac{K(\varphi)}{\Phi(\varphi, \psi)} \right]^{\frac{1}{2}} d\varphi, \tag{16}$$

$$bh\cos(\psi - \varphi_H) - \sigma\sin 2(\varphi - \psi) = 0, \tag{17}$$

$$f = \bar{f} Q E(\varphi, \psi), \tag{18}$$

where

$$\Phi(\varphi, \, \psi) = h^2 \left[\sin^2(\varphi - \varphi_H) - \sin^2(\varphi_0 - \varphi_H) \right]
+ 2Er(\varphi - \varphi_0) + Er\lambda(\sin 2\varphi_0 - \sin 2\varphi) + 2Q\varsigma \left[E(\varphi_0, \, \psi_0) - E(\varphi, \, \psi) \right],$$

and the functions $K(\varphi)$ and $E(\varphi, \psi)$ are determined by Eq. (14), $\varphi_0 = \varphi(0)$ and $\psi_0 = \psi(0)$ are the angles of orientation of the director and the magnetization in the middle of the layer. Equations (15)–(16) with the "plus" sign give the solutions with positive values (while "minus" sign —with negative) of the angle of the director rotation.

Thus, Eqs. (15)–(18) determine the concentration f of magnetic particles, the director angle φ and the magnetization angle ψ of a ferronematic under shear flow as functions of the magnetic field strength h, the angle φ_H of magnetic field orientation, the coupling energy σ , the anisotropy of elasticity k, the segregation parameter ς , the reactive parameter λ , the parameter b and the various values of Ericksen number Er.

4. Shear and Magnetic Field Influence on Ferronematic Structure

We direct the magnetic field H = (0, 0, H) perpendicularly to the plane of FN layer, therefore $\phi_H = \pi/2$ (see Fig. 1). The results of numerical calculations of Eqs. (15)–(18) are shown in Figures 2–4.

In the shear absence (Er = 0, solid line in Fig. 2) and at $h < h_c^{FN}$ the ferronematic layer has uniform planar structure with homeotropic coupling between the director and magnetization ($m \perp n$). Here the critical field h_c^{FN} is determined by the equation

$$(h_c^{FN})^2=\pi^2+rac{2\sigma b h_c^{FN}}{2\sigma+b h_c^{FN}}.$$

With the increase in the field the uniform state becomes unstable, and for $h > h_c^{FN}$ the distorted structure appears, i.e., at $h = h_c^{FN}$ the Freedericksz transition takes place. In this case two symmetric solutions describing clockwise $(\varphi_0 < 0)$ and anti-clockwise $(\varphi_0 > 0)$ director rotation correspond to the distorted state [see solid lines in Figs. 2–4]. In this state the vectors \mathbf{m} and \mathbf{n} are no longer perpendicular, that corresponds to the so-called angular ferronematic phase [21]. As the field increases, the director and the magnetization tend to align along the field direction $(\varphi_0 \to \pm \pi/2, \ \psi_0 \to 0)$. Thus, the planar $(\mathbf{m}||\mathbf{n})$ ferronematic phase [21] can be achieved at $h \to \infty$ only.

The presence of shear stress $(Er \neq 0)$ does not change the threshold character of the transition only when the absolute values of rotary viscosity coefficients are equal $(\lambda = 1)$. In this case the critical field h_c^{FN} is the same as the one for the static transition(Er = 0), but the symmetry of non-trivial solutions is now broken (Fig. 2,

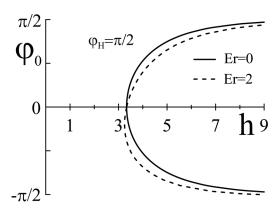


Figure 2. The angle φ_0 of the director **n** orientation as a function of a magnetic field strength **h** for different values of Ericksen number Er and $\sigma = 1$, k = 1, $\varsigma = 5$, b = 1, $\lambda = 1$.

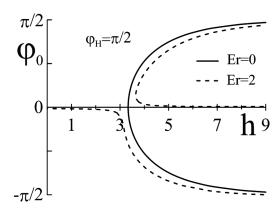


Figure 3. The angle φ_0 of the director **n** orientation as a function of a magnetic field strength h for different values of Ericksen number Er and $\sigma = 1$, k = 1, $\varsigma = 5$, b = 1, $\lambda = 0.9$.

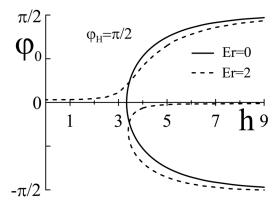


Figure 4. The angle φ_0 of the director **n** orientation as a function of a magnetic field strength h for different values of Ericksen number Er and $\sigma = 1$, k = 1, $\varsigma = 5$, b = 1, $\lambda = 1.2$.

dashed line). The shear flow leads to the breakdown of invariance of Eqs. (15)–(18) under the replacement $\varphi_0 \to -\varphi_0$, $\psi_0 \to -\psi_0$. The trivial solutions $\varphi_0 = \psi_0 = 0$ still exist for any values of magnetic field strength h. The branches of solutions corresponding to the non-uniform states of the director are displaced downwards. Numerical calculations show, that for $\lambda \neq 1$ shear stress "smoothes" the orientational transition. In this case the shear stress acts in the same way as the external field in Landau theory of phase transitions.

In Figure 3 the angles of the director and magnetization orientation in a ferrone-matic with non flow-aligning NLC-matrix ($0 \le \lambda < 1$) are shown. In this case the trivial solution describing the non-perturbed configuration of the director disappears at imposing of shear stress. Two branches of solutions (Fig. 3, dashed lines) appear, describing the clockwise and counter-clockwise rotation of the director. Non-trivial solutions in the top semi-plane ($\varphi_0 > 0$, $\psi_0 > 0$) appear at greater h values, than in static Freedericksz transition for ferronematics (Fig. 3, continuous lines).

In a ferronematic with flow-aligning ($\lambda \ge 1$) NLC-matrix the shear flow, as well as in the previous case, leads to "smoothing" of the transition, but the continuous

branch of solutions now lays in the top semi-plane ($\varphi_0 > 0$) and exists for any h (Fig. 4, dashed lines). This is due to the fact that in the magnetic field absence the shear flow orients the director under a positive angle to the direction of flow. Imposing of the field succeeded by the increase in its strength leads to growth of φ_0 positive values. Besides, there are two more branches of solutions in the bottom semi-plane ($\varphi_0 < 0$). The more intensive the shear flow is, the stronger the orientational transition in a considered configuration "smoothes."

5. Conclusion

Within the framework of continuum theory we have analyzed the Freedericksz transition in a ferronematic layer under the combined influence of magnetic field and shear flow. Magnetic field has been applied in the shear plane. We have imposed the rigid planar coupling conditions for the director on the boundaries of the layer and the soft homeotropic ones on the surfaces of the magnetic particles. The linear approximation of a velocity field in the layer has been used. Taking into account the segregation effects we have obtained the stationary integral equations for the concentration of magnetic particles and planar director and magnetization fields.

We have performed numerical calculations of the angles of the director and the magnetization rotation for different values of magnetic field strength and Ericksen number. Both flow-aligning and non-flow-aligning liquid crystal matrixes have been considered. In the case when the magnetic field is directed perpendicular to the layer we have shown that only the angular ferronematic phase [21] with acute angle between the director and magnetization exists. The shear flow leads to symmetry loss of the solutions describing the disturbed states of the director and the magnetization. The existence of non-disturbed solutions for the director and magnetization is possible only when the value of reactive parameter equals one. For any other values of reactive parameter the "smoothing" of phase transition between orientational states takes place. For ferronematics with non-flow-aligning NLC-matrix we have obtained the solutions describing the stationary orientations of the director and the magnetization in the shear plane.

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