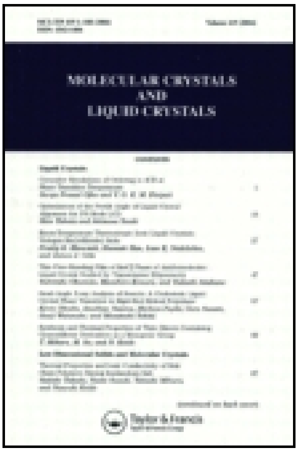

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Freedericksz Transition in Compensated Ferronematic Liquid Crystals

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In the framework of continuum theory, the Freedericksz transition in ferronematic, i.e., suspension of monodomain magnetic particles in nematic liquid crystal, is studied. In the absence of a magnetic field, the suspension is supposed to be compensated, i.e., it contains equal numbers of magnetic particles with magnetic moments oriented in parallel and antiparallel to local director. Spatial distortions of director and concentrational redistribution of magnetic admixture in ferronematic layer under the influence of a magnetic field are studied. Threshold character of magnetic field induced Freedericksz transition from homogeneous to inhomogeneous state is shown. Transition field as a function of ferronematic material parameters is analytically found. Magnetization of ferronematic is studied.

Keywords Ferronematic; Freedericksz transition; liquid crystal; magnetic field; segregation effect

1. Introduction

Ferronematics (FNs) is the name for suspensions of anisometric ferrimagnetic or ferromagnetic particles where the liquid carrier is nematic liquid crystals (NLC). They were predicted theoretically in Ref. [1] that proposed the continuum approach for their description. As opposed to pure NLC being a diamagnetic medium, they are highly susceptible to the influence of the external magnetic fields. Even at low concentrations ($\sim$0.01% vol) of magnetic particles, it is possible to control the orientation of FN with respectively weak magnetic fields ($H \sim 10$ Oe), whereas for the pure NLC, magnetic fields of $H \sim 1$ kOe are needed. FNs are complex fluids, physical properties of which are significantly richer than those of their components. From liquid crystals (LCs), they inherited fluidity and orientational order and hence the anisotropy of physical properties, and from the dispersed medium, high magnetic susceptibility. A distinctive feature of FNs is the orientational bond between magnetic particles and NLC matrix that is called coupling. The presence of this coupling leads to the fact that small impurity particles change physical properties of NLC matrix never essentially disturbing the homogeneous director orientation.

Dispersity of such suspensions is very high: their magnetic phase is formed by monodomain particles of ferromagnetic or ferrite having linear dimensions from fractions of a micron and up to tens of nanometers. So, from macroscopic point of view, FNs can

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be considered as homogeneous anisotropic liquids having high susceptibility to magnetic fields.

The unusual physics of FNs is due to the fact that in contrast to pure LCs, they have not one but two different mechanisms of orientational response to the magnetic field applied. The first one is due to anisotropy of diamagnetic susceptibility of liquid crystalline matrix. The second mechanism arises only in the systems containing dispersed ferromagnetics. Interacting with the magnetic moment of the particle, the field changes orientation of the latter and then coupling forces on the particle-liquid crystal boundary transfer created mechanical rotation to the liquid-crystalline matrix. Existence of two modes of magnetically orientational response generates multiple new effects in FNs which can be interesting not only for fundamental materials science but also very attractive from the practical point of view.

Despite the fact that LCs and liquid-crystalline suspensions have been studied theoretically and experimentally during a long time (from the beginning of 80s of the previous century), a number of problems still remain unsolved. It applies also to the problem of magnetizing of the so-called compensated FNs.

At temperatures higher than the point of transition to mesophase, such suspensions are isotropic; during cooling down phase transition in NCL, matrix takes place initiating long distance orientational order. If cooling down of FN is exercised in the absence of a magnetic field, then the resulting suspension is compensated, i.e., it has macroscopically equal portions of ferroparticles with magnetic moments oriented in parallel and antiparallel toward local director **n** (see Fig. 1). Orientational and magnetic properties of these suspensions are not studied: Ref. [1] offered only the continuum approach for their description and qualitatively studied the behavior of unlimited compensated FN in the magnetic field oriented along the director **n**. The present work is devoted to theoretical study of Freedericksz transition in compensated FN in twist geometry. Proposed consideration is applicable (without taking into account medium polarization) also to electrical analogues of compensated FN—suspensions of ferroelectric particles on the basis of NCL [2–7].

2. Free Energy and Equilibrium Equations

Let FN be located in a layer having thickness L. Axis x of coordinate system will be parallel to bounding plates, whereas axis z—perpendicular to them—origin will be in the center of

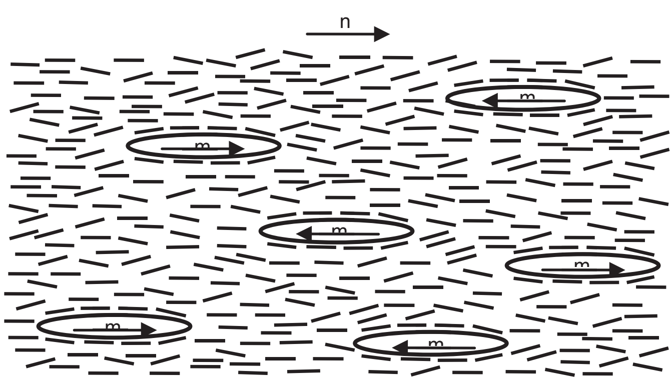

Figure 1. Schematic representation of compensated FN: **n** is the director, **m** is the unit vector of magnetization.

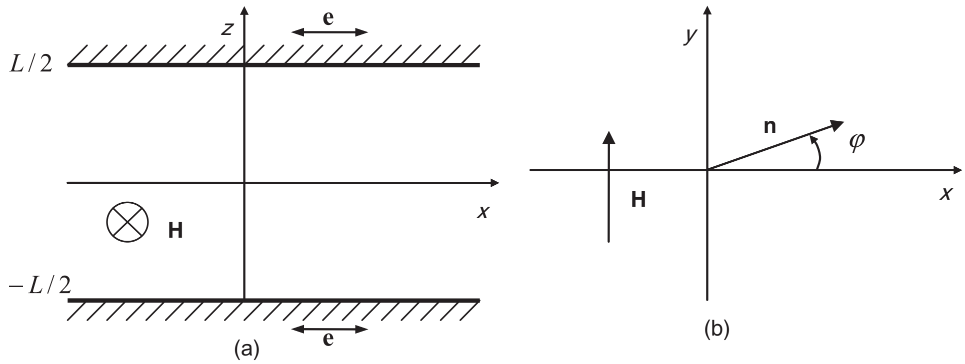

Figure 2. Planar ferronematic layer in an external magnetic field. The choice of coordinate system.

the layer [see Fig. 2(a)]. We assume that there is rigid planar anchoring of director to the boundaries of the layer, i.e., director is fixed at the boundary and directed along the axis of easy orientation $\mathbf{e} = (1, 0, 0)$. We also consider the anchoring of magnetic particles to LC matrix to be absolutely rigid and planar. Let us direct magnetic field $\mathbf{H} = (0, H, 0)$ in parallel to the boundaries of the layer along axis y. The expression for the FN free energy reads

$$F = \iiint F_V dV. \tag{1}$$

Here spatial density of the FN free energy has the following form [1, 8]

$$\begin{aligned} F_V &= F_1 + F_2 + F_3 + F_4, \\ F_1 &= \frac{1}{2}[K_1 (\nabla \cdot \mathbf{n})^2 + K_2 (\mathbf{n} \cdot \nabla \times \mathbf{n})^2 + K_3 (\mathbf{n} \times \nabla \times \mathbf{n})^2], \\ F_2 &= -\frac{1}{2}\chi_a (\mathbf{n} \cdot \mathbf{H})^2, \quad F_3 = -M_s (f_+ - f_-) (\mathbf{n} \cdot \mathbf{H}), \\ F_4 &= \frac{k_B T}{v} (f_+ \ln f_+ + f_- \ln f_-), \end{aligned} \tag{2}$$

where K_1, K_2, and K_3 are the Frank elastic moduli; $\mathbf{n}$ is the LC director; M_s is the saturation magnetization of the particle substance; f_+ and f_- are volume fractions of particles with magnetic moments $\boldsymbol{\mu} = M_s v \mathbf{m}$ (here $\mathbf{m}^2 = 1$), directed in parallel ($\mathbf{m} \equiv \mathbf{n}$) and in antiparallel ($\mathbf{m} \equiv -\mathbf{n}$) to the local director **n,** respectively; χ_a is the anisotropy of LC diamagnetic susceptibility (we assume $\chi_a > 0$, so the director tends to rotate in the direction of the field); v is the particle volume; k_B is the Boltzmann constant; and T is the temperature. We assume that suspension is compensated, i.e., in the absence of magnetic field, it contains equal fractions of ferroparticles with magnetic moments directed along $\mathbf{n}$ or $-\mathbf{n}$ ($f_+ |_{H=0} = f_- |_{H=0} \equiv \bar{f}/2$, where $\bar{f} = Nv/V$, N is the number of magnetic particles in a suspension, and V is the FN volume), so total FN magnetization equals to zero (see Fig. 1). We assume $\bar{f} << 1$, which allows to neglect interparticle magnetic dipole–dipole interactions in a suspension.

The term F_1 in the expression (2) represents the free energy density of elastic deformation of the director field (the Oseen–Frank potential). The second (F_2) and the third (F_3) contributions characterize the interaction of diamagnetic nematic and magnetic moments of particles with external magnetic field **H**, respectively. The fourth term (F_4) describes the contribution of the entropy of the suspension particles "ideal gas" mixing.

In our case, the deformation of the orientational structure in the field corresponds to twist and the solution can be sought in the form

$$\mathbf{n} = [\cos\varphi(z), \sin\varphi(z), 0]\,. \tag{3}$$

Here $\varphi(z)$ is the angle of the director deviation from the easy orientation axis $\mathbf{e} = (1, 0, 0)$ [see Fig. 2(b)].

Let us choose the thickness of the layer L as a unit of length and define dimensionless quantities: the coordinate $\zeta = z/L$, the field strength $h = HL\sqrt{\chi_a/K_2}$, reduced volume fractions $g_\pm = f_\pm/\bar{f}$, and dimensionless material parameters [9]

$$b = \frac{M_s \bar{f} L}{\sqrt{K_2\chi_a}}, \quad \kappa = \frac{k_B T \bar{f} L^2}{K_2 \nu}. \tag{4}$$

Here we use the value $H_q = L^{-1}\sqrt{K_2/\chi_a}$ as a unit of field strength. It is chosen from the energy balance of elastic deformations F_1 and diamagnetic contribution F_2 [see Eq. (1)]. At $H \geq H_q$, orientational distortions arise because of magnetic anisotropy of NLC matrix. Similarly, from the balance of elastic F_1 and dipole F_3 contributions (magnetic particles energy in a magnetic field) in the free energy density can be found another characteristic field $H_d = K_2/(M_s \bar{f} L^2)$. In this case, for $H \geq H_d$, the distortions in FN are caused by interaction of magnetic particles with external magnetic field (dipole mechanism). The parameter $b = H_q/H_d$ represents the ratio of two characteristic fields H_q and H_d and characterizes the mechanism of magnetic field influence on FN [9]. For $b >> 1$, when $H_d << H_q$, orientational distortions in weak field are caused by dipole mechanism, and in case $b << 1$ $\left(H_d >> H_q\right)$, they are caused by quadrupole mechanism. At $H \approx H_0 = M_s \bar{f}/\chi_a$, the terms F_2 and F_3 become of the same order and predominant mechanism of magnetic field influence on FN changes from dipole to quadrupole (or vice versa). Segregation parameter $\kappa = (L/\lambda)^2$ represents a square of ratio of two characteristic lengths [9]—the layer thickness L and segregation length $\lambda = (\nu K_2/k_B T \bar{f})^{1/2}$ [1]. For $\kappa >> 1$, the segregation effect is weak, because characteristic size of segregation area is less than thickness of the layer.

Substituting Eq. (3) into Eq. (1), we find for the dimensionless free energy $\tilde{F} = FL/(K_2 S)$ the expression of the following form

$$\tilde{F} = \int_{-1/2}^{1/2} \left(\frac{1}{2}\left(\frac{d\varphi}{d\zeta}\right)^2 - \frac{1}{2}h^2 \sin^2\varphi - bh\,(g_+ - g_-)\sin\varphi + \kappa\,(g_+ \ln g_+ + g_- \ln g_-) \right) d\zeta, \tag{5}$$

where S is the boundary planes area.

The equilibrium state corresponds to the minimum of free energy [Eq. (5)], the latter being functional with respect to functions $\varphi(\zeta)$ and $g_\pm(\zeta)$. Minimization of the free energy [Eq. (5)] over $\varphi(\zeta)$ gives the equation for the angle of director orientation:

$$\varphi'' + \frac{1}{2}h^2 \sin 2\varphi + hb\,(g_+ - g_-)\cos\,\varphi = 0. \tag{6}$$

Hereafter, the prime denotes the derivative with respect to ζ. Condition of rigid planar coupling of the director with the boundary $\mathbf{n} \,||\, \mathbf{e} = (1, 0, 0)$ takes the following form:

$$\varphi(-1/2) = \varphi(1/2) = 0. \tag{7}$$

Equilibrium distribution of magnetic particles $g_\pm(\zeta)$ in a layer is found by minimizing free energy [Eq. (5)] over g_+ and g_- with the additional condition of constancy of the magnetic particles number N in a suspension

$$\int_V (f_+ + f_-)\, dV = N\nu,$$

or in terms of reduced volume fractions

$$\int_{-1/2}^{1/2} (g_+ + g_-)\, d\zeta = 1. \tag{8}$$

Minimizing free energy [Eq. (5)], we obtain

$$g_\pm(\zeta) = Q \exp\left\{\pm\frac{bh}{\kappa}\sin\varphi(\zeta)\right\}, \quad Q^{-1} = \int_{-1/2}^{1/2} 2\cosh\left\{\frac{bh}{\kappa}\sin\varphi(\zeta)\right\} d\zeta. \tag{9}$$

Formula (9) describe the so-called effect of segregation [1]—the phenomenon of the accumulation of magnetic particles in those parts of a layer where their magnetic energy is minimal.

Note that the distribution of impurity particles [Eq. (9)] differs from that found in Refs. [2–4], where instead of integral condition [Eq. (8)] of particle number constancy in the suspension, the relation $g_+ + g_- = 1$ is used.

The set of Eqs. (6) and (9) with boundary conditions [Eq. (7)] admits the uniform solution

$$\varphi(\zeta) \equiv 0, \quad g_+(\zeta) = g_-(\zeta) \equiv 1/2, \tag{10}$$

corresponding to the initial planar texture of FN ($\mathbf{n} \,||\, \mathbf{e} \perp \mathbf{H}$). As shown later, this solution becomes unstable at fields exceeding a certain threshold, called the Freedericksz field.

Along with uniform solution [Eq. (10)], the system of equations admits also nonuniform solution for the director and the concentration. To find it, we multiply Eq. (6) by φ'. As a result we obtain

$$\frac{d}{d\zeta}[(\varphi')^2 - h^2\cos^2\varphi + 2\kappa(g_+ + g_-)] = 0. \tag{11}$$

In the center of the layer, the angle of director deviation is maximal ($\varphi' = 0$ at $\zeta = 0$), for this reason, the first integral of Eq. (11) takes the following form:

$$\varphi' = \pm\mathcal{R}^{-1/2}(\varphi). \tag{12}$$

Here

$$\mathcal{R}^{-1}(\varphi) = h^2(\cos^2\varphi - \cos^2\varphi_0) + 2\kappa(g_{0+} + g_{0-} - g_+ - g_-), \tag{13}$$

and introduced notations $g_{0\pm} \equiv g_{\pm}(\varphi_0)$ and $\varphi_0 \equiv \varphi(0)$ for the reduced volume fractions of ferroparticles and for the angle of director rotation in the center of the layer, respectively.

Integration of Eq. (12) for $\zeta > 0$ with boundary conditions [Eq. (7)] gives an implicit dependence $\varphi(\zeta)$:

$$\int_0^{\varphi(\zeta)} \mathcal{R}^{1/2}(\varphi)\, d\varphi = \frac{1}{2} - \zeta. \tag{14}$$

For definiteness, we have chosen the plus sign in Eq. (14), which corresponds to counterclockwise ($\varphi_0 > 0$) rotation of the director.

In the center of the layer ($\zeta = 0$), the angle $\varphi = \varphi_0$ and Eq. (14) take the following form

$$\int_0^{\varphi_0} \mathcal{R}^{1/2}(\varphi)\, d\varphi = \frac{1}{2}. \tag{15}$$

Passing in expression (9) for Q from integration over coordinate to integration over the angle φ with the help of relation (12), we obtain the equation for Q in the following form:

$$\int_0^{\varphi_0} (g_+ + g_-)\, \mathcal{R}^{1/2}(\varphi)\, d\varphi = \frac{1}{2}. \tag{16}$$

Magnetization of FN has the form $\boldsymbol{\mathcal{M}} = M_s(f_+ - f_-)\,\mathbf{n}$. Let us introduce the dimensionless magnetization $\mathbf{M} = \boldsymbol{\mathcal{M}}/(M_s \bar{f}) = (g_+ - g_-)\,\mathbf{n}$ and define the average magnetization across the thickness of the layer by relation

$$\langle \mathbf{M} \rangle = \frac{1}{L} \int_{-L/2}^{L/2} \mathbf{M}\, dz = \int_{-1/2}^{1/2} \mathbf{M}\, d\zeta. \tag{17}$$

Using Eq. (12), we replace the integration variable in Eq. (17) and then obtain the following expressions for averaged over the layer thickness components of magnetization

$$\langle M_x \rangle = 2 \int_0^{\varphi_0} (g_+ - g_-)\ \mathcal{R}^{1/2}(\varphi)\ \cos\varphi\ d\varphi, \quad \langle M_y \rangle = 2 \int_0^{\varphi_0} (g_+ - g_-)\ \mathcal{R}^{1/2}(\varphi)\ \sin\varphi\ d\varphi. \tag{18}$$

Thus, Eqs. (14)–(16), (18), and boundary conditions [Eq. (7)] determine the angle $\varphi(\zeta)$ of the director rotation, distribution $g_{\pm}(\zeta) = f_{\pm}(\zeta)/\bar{f}$ of magnetic admixture concentration, and average components of magnetization $\langle M_x \rangle$, $\langle M_y \rangle$ in FN layer depending on magnetic field h and dimensionless parameters b and κ.

Let us estimate the dimensionless quantities [Eq. (4)], using typical material parameters of NLC and magnetic particles [10–14]. For FN based on LC 5CB, we have $\chi_a = 1.67 \times 10^{-7}$, $K_2 = 3 \times 10^{-7}$ dyn, $T = 298$ K, $\bar{f} = 2 \times 10^{-7}$, $M_s = 500$ Gs, $\nu = 1.5 \times 10^{-16}$ cm^3, and assuming thickness of the layer $L = 2.5 \times 10^{-2}$ cm, we get $b \approx 10$, $\kappa \approx 0.1$. Small values of parameter κ tell about the importance of magnetic segregation effects in the problem considered.

3. Freedericksz Transition

As noted above, equations of orientational equilibrium [Eqs. (6), (7), and (9)] have uniform solution $\varphi(\zeta) \equiv 0$ and $g_+(\zeta) = g_-(\zeta) \equiv 1/2$, which corresponds to a planar texture of FN ($\mathbf{n} \,||\, \mathbf{e} \perp \mathbf{H}$). However, such a state becomes unstable if external magnetic field exceeds a certain threshold value h_c, known as Freedericksz field [15]. In the vicinity of h_c, the values of $\varphi(\zeta)$ are small, so that from Eq. (8) in the lowest order, we obtain

$$g_+ - g_- = 2Q\frac{bh}{\kappa}\varphi, \quad Q = \frac{1}{2},$$

and Eq. (6) takes the form

$$\varphi'' + h_c^2\varphi + hb\,(g_+ - g_-) = 0.$$

As a result, we get the following equation for $\varphi(\zeta)$:

$$\varphi'' + h_c^2\left(1 + \frac{b^2}{\kappa}\right)\varphi = 0. \tag{19}$$

Nontrivial solution of Eq. (19) with boundary conditions (7) has the following form:

$$\varphi\ (\zeta) = \varphi_0 \cos(\pi\zeta)\,; \tag{20}$$

It exists at $h > h_c$, where h_c is defined by

$$h_c^2 = \frac{\pi^2}{1 + b^2/\kappa}. \tag{21}$$

From the formula (21), it can be seen that Freedericksz field h_c, as it should be in the geometry under consideration, does not change if the direction of magnetic field is reversed. At $\bar{f} \equiv 0$, it coincides with Freedericksz field ($h_c = \pi$) for pure nematic [15]. The presence of compensated magnetic admixture lowers the threshold of the transition ($h_c < \pi$), which is characteristic also for suspensions of ferroelectric particles in the NLC [4]. We further note that Freedericksz threshold field h_c decreases significantly in the dipole mode ($b >> 1$) and at low κ, when the segregation phenomena are particularly important.

4. Landau Expansion

In the vicinity of Freedericksz transition field h_c, where a nonuniform orientation of the director takes place, the free energy [Eq. (5)] can be expanded into power series in $\varphi(\zeta) = \varphi_0 \cos(\pi\zeta)$, where $\varphi_0 << 1$. In the fourth order of expansion in φ_0, the free energy [Eq. (5)] gets the form of Landau expansion

$$\tilde{F} = \tilde{F}_0 + \frac{\alpha}{2}(h_c - h)\,\varphi_0^2 + \frac{\beta}{4}\varphi_0^4 + \cdots, \tag{22}$$

where

$$\tilde{F}_0 = -\kappa \ln(2)\,, \quad \alpha = \frac{\pi^2}{h_c}, \quad \beta = \frac{\pi^2}{4} + \frac{b^4 h_c^4}{16\kappa^3}.$$

Here h_c is defined by Eq. (21). Minimization of free energy [Eq. (22)] over φ_0 gives the expression for the angle of director rotation in the center of the layer:

$$\varphi_0 = \pm\sqrt{\frac{\alpha}{\beta}(h - h_c)}. \tag{23}$$

This formula shows that nonzero solutions, corresponding to inhomogeneous phase, exist only at $h > h_c$. Value of h_c defines, thus, the threshold field (Freedericksz field), above which the orientational distortions take place in previously homogeneous FN. Formula (23) defines the magnitude of the director field orientational distortions in the vicinity of orientational instability threshold [see Fig. 3(a)]. It represents the so-called normal bifurcation, for which the angle of the director rotation in the center of the layer at $h > h_c$ increases continuously from zero as $(h - h_c)^{1/2}$; such behavior in the Landau theory is typical for the second order transitions. Therefore, Freedericksz transition in FN is the second order phase transition (inhomogeneous phase is stable at $h > h_c$) as in pure LC.

5. Numerical Results

Figures 3–7 represent the results of numerical solution for Eqs. (14)–(16) and (18) of orientational and magnetic state of FN layer for $b = 10$ and $\kappa = 5$ (in this case $h_c = 0.69$). Figure 3 represents dependence of the director deviation angle φ_0 and ferroparticles concentration g_{0+} and g_{0-} in the center of the layer on the applied magnetic field h. At $h \le h_c = 0.69$, Eqs. (15) and (16) have the solution $\varphi_0 = 0$ and $g_\pm = 1/2$ that corresponds to the homogeneous planar FN texture, i.e., the director is oriented along the axis of easy orientation at the layer boundaries ($\mathbf{n} \,||\, \mathbf{e} \perp \mathbf{H}$), the particles are uniformly dispersed throughout the volume. This state becomes unstable when the field exceeds the threshold value h_c. Further increase of the magnetic field leads to orientational distortions in FN ($\varphi_0 \neq 0$), which appear as the second-order transition (the order parameter is given by $\sin^2\varphi_0$), similar to classical Freedericksz transition in pure LC [15]. With the field increasing, the angle of the director deviation from the axis of easy orientation in the center of the layer also increases and asymptotically tends in the field direction $\varphi_0 = \pi/2$, while the concentration g_{0-} of the particles, oriented against the director, decreases drastically,

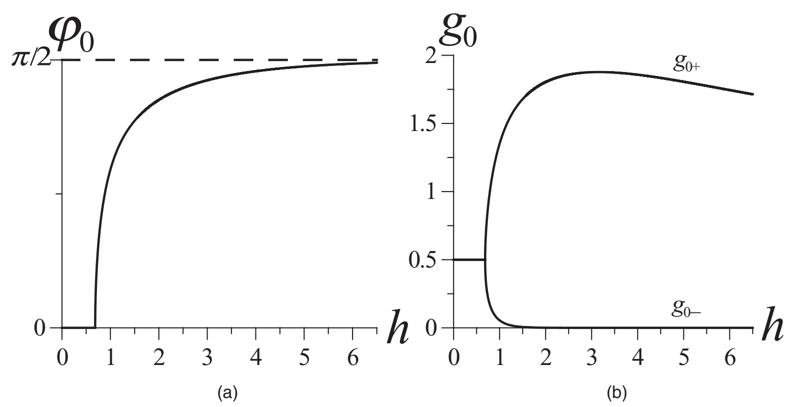

Figure 3. (a) Director orientation angle φ_0, (b) concentration of magnetic particles directed along (g_+) and against (g_-) director in the center of the layer as a function of the magnetic field h.

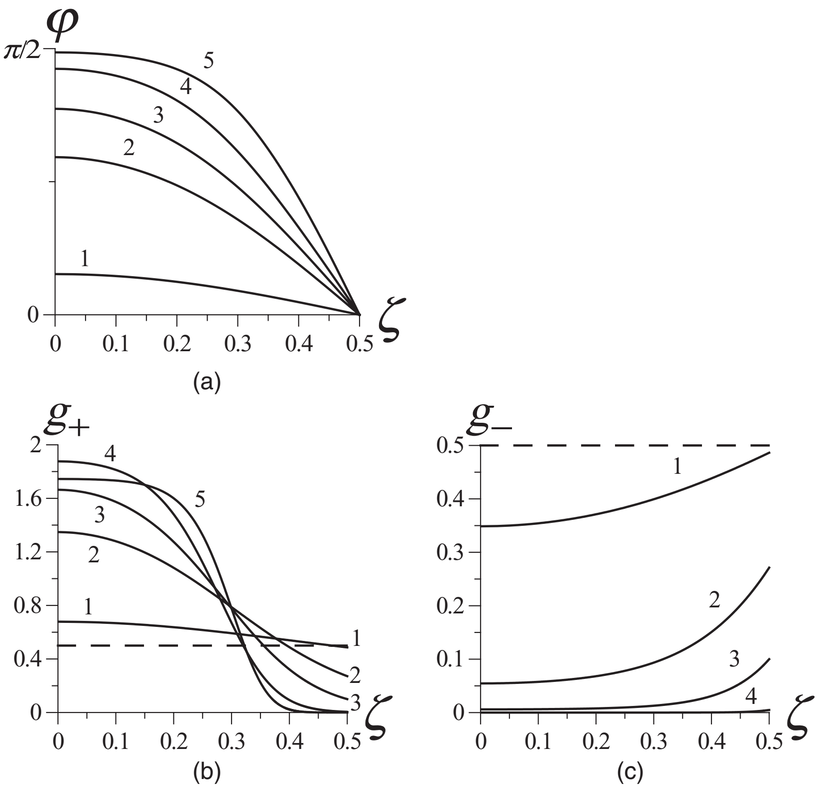

Figure 4. Director orientation angle (a) and concentration of ferroparticles with magnetic moments, directed along (b) and against (c) the director in a layer of FN. Curve 1, $h = 0.7$; 2, $h = 1$; 3, $h = 1.5$; 4, $h = 3$; 5, $h = 6$. Dashed line $g_+ = g_- = 1/2$ corresponds to $h \leq h_c$.

and g_{0+} behaves nonmonotonically: it rapidly increases above the Freedericksz threshold and then asymptotically tends to unit. Thus, with the increase of the field, the particles, magnetic moments of which are directed along the director, accumulate in the center of the layer.

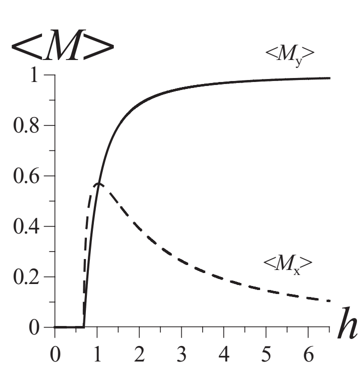

Figure 5. Average ferronematic magnetization: dashed curve, $\langle M_x \rangle$; solid curve, $\langle M_y \rangle$.

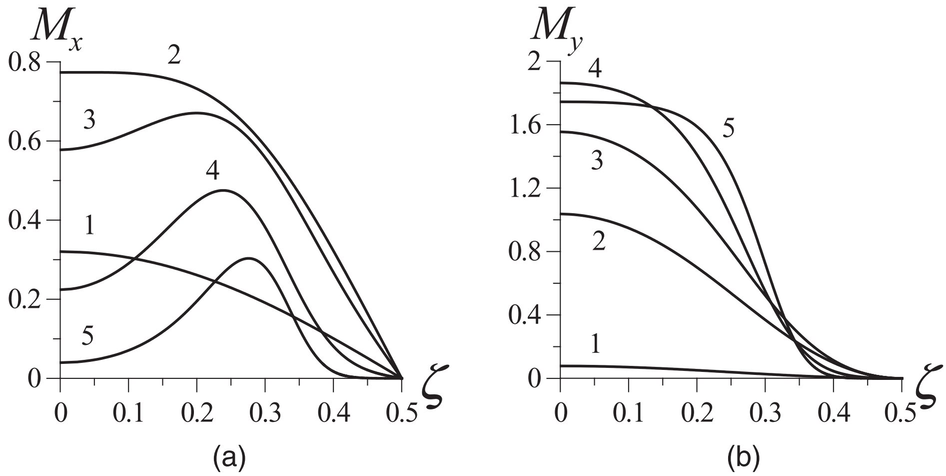

Figure 6. (a) x-component and (b) y-component of magnetization in FN layer. Curve 1, $h = 0.7$; 2, $h = 1$; 3, $h = 1.5$; 4, $h = 3$; 5, $h = 6$.

Figure 4 represents the director rotation angle $\varphi(\zeta)$ and magnetic particles concentrational distribution $g_{\pm}(\zeta)$ in a FN layer at various values of the field $h > h_c = 0.69$, obtained by numerical solving Eqs. (14)–(16). At $h \leq h_c$, FN is in homogeneous phase [$\varphi(\zeta) = 0$ and $g_{\pm} = 1/2$—see dashed lines in Figs. 4(b) and 4(c)]. At $h > h_c$, orientational and concentrational inhomogeneities appear. With the increase of the field, director is oriented in the direction of the field [see Fig. 4(a)] and ferroparticles with magnetic moments parallel to the director start to migrate from the boundaries of the sample to its center—the so-called segregation effect [see Fig. 4(b)]. Ferroparticles with unfavorable orientation of magnetic moments migrate to the boundaries of the layer where the director is oriented

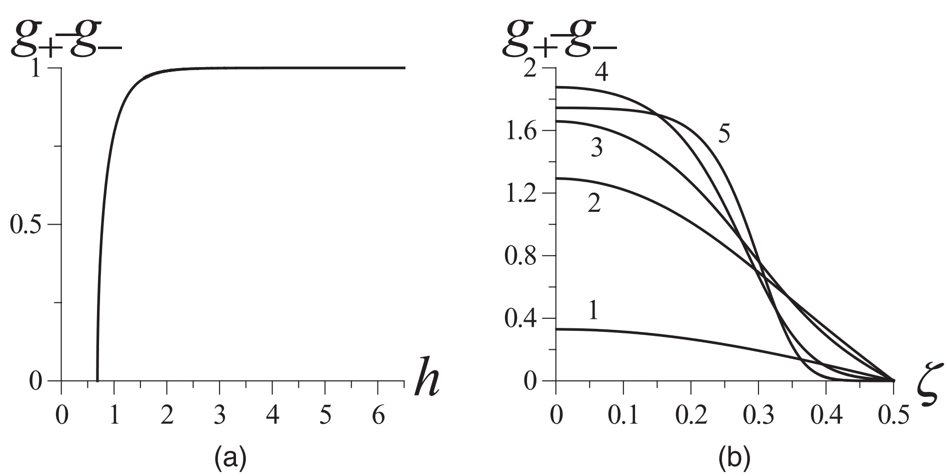

Figure 7. (a) Difference between volume fractions of magnetic particles in the center of a layer as a function of magnetic field; (b) difference between volume fractions of magnetic particles in FN layer in different magnetic fields. Curve 1, $h = 0.7$; 2, $h = 1$; 3, $h = 1.5$; 4, $h = 3$; 5, $h = 6$.

along the axis of easy orientation, but their concentration g_- is significantly reduced with the increase of the field [see Fig. 4(c)]. In strong fields ($h >> h_c$), concentration of particles, with magnetic moments oriented against the director, tends $g_- \to 0$ and $g_+ \to 1$ in most parts of the layer, for the exception of narrow near-wall zone, from which particles migrated to the center of the layer.

Figure 5 represents the components of the average magnetization [Eq. (17)] of FN as a function of the magnetic field. It is evident that with the increase of the field over the threshold h_c, FN ceases to be compensated and the sample becomes magnetized. In the vicinity of h_c, director deviations from the axis of easy orientation are small and expressions [Eq. (18)] for $\langle M_x \rangle$ and $\langle M_y \rangle$ can be expanded in power series in $\varphi = \varphi_0 \cos(\pi\zeta)$, where φ_0 is defined by the formula (23). In the lowest order of expansion in φ_0, we find

$$\langle M_x \rangle = \frac{2}{\pi}\frac{bh}{\kappa}\varphi_0, \quad \langle M_y \rangle = \frac{1}{2}\frac{bh}{\kappa}\varphi_0^2, \tag{24}$$

and after substitution of (23) into (24), we finally obtain

$$\langle M_x \rangle = 8bh_c\sqrt{\frac{\kappa\,(h-h_c)}{h_c\left(4\pi^2\kappa^3 + b^4h_c^4\right)}}, \quad \langle M_y \rangle = \frac{8\pi^2 b\kappa^2\,(h-h_c)}{4\pi^2\kappa^3 + b^4h_c^4}. \tag{25}$$

These formulas describe in Fig. 5 the behavior of average magnetizations over the threshold h_c. In Fig. 5, it is explicitly shown that following the increase of the magnetic field, $\langle M_y \rangle$ tends to saturation, in which magnetic moments of ferroparticles are directed along the field h, whereas $\langle M_x \rangle$ demonstrates nonmonotonic response, connected with remagnetization of "unfavorably" oriented subsystem. Above the threshold of the Freedericksz transition, magnetization in the direction perpendicular to the applied field $\langle M_x \rangle = \langle (g_+ - g_-)\,n_x \rangle$ increases rapidly and reaches the extremum, and then, following the field growth, asymptotically approaches to zero. The extremum is related to magnetic reversal of "unfavorable" subsystem.

Figure 6 represents magnetization distribution in the layer for different values of the magnetic field. With the increase of the field, as it was already noted, increases the concentration $g_+(\zeta)$ of favorably orientated toward the field magnetic particles and their migration to the mid-layer. For this reason, spatial distribution of magnetization component $M_y(\zeta)$ qualitatively seems similar to $g_+(\zeta)$ [see Fig. 4(b)], because magnetization in the direction of the field is determined by the particles, magnetic moments of which are oriented along the director. For $M_x(\zeta)$, spatial distribution has a more complicated pattern: as magnetic field grows, magnetic particles quantum g_+ increases in the center [see Figs 3(b), 4(b), and 7(a)], which explains the appearance of a minimum in Fig. 6(a). Presence of the extremum on the curve $M_x(\zeta) = (g_+ - g_-)\cos\varphi(\zeta)$ and its shift to the boundaries of the layer as the field grows are due to the decrease of the volume fraction of "unfavorably" orientated particles because of their migration to the boundaries of the layer [see Fig. 7(b)].

We now discuss the nonmonotonic behavior of concentration g_{0+} as a function of applied magnetic field [see Figs 3(b) and 4(b)]. Above the threshold of the Freedericksz transition in FN layer, the distortions of the director orientation arise and the segregation phenomena take place. Recall that this phenomenon consists in accumulation of magnetic particles in those areas of the layer where their magnetic energy has a minimum, i.e., where the director is aligned along the field. In the considered geometry, this corresponds to the middle of the layer. As it is seen from Figs 3(a) and 4(a), with the field increasing the angle φ_0 of the director deviations in the middle of the layer grows, and in accordance with

Eq. (9), the concentration of the magnetic particles with magnetic moments oriented along the director (“+ family”) in the middle of the layer increases [see Fig. 4(b), curves 1−4]. At $h \geq 3.5$, the angle φ_0 becomes close to $\pi/2$, i.e., the magnetic particles of the “+ family” in the middle of the layer are oriented along the field. Further growth of the field leads to a broadening of the region in which the angle of the director deviations is close to $\pi/2$, i.e., the orientation of the director is close to the field direction [see Fig. 4(a), curve 5]. For this reason, the magnetic particles of the “+ family” occupy an increasing part of the layer [Fig. 4(b), curve 5], that due to conservation law [Eq. (8)] leads to decrease of their volume fraction in the middle of the layer.

Nonmonotonic behavior of M_x and $\langle M_x \rangle$ in the dependence of applied magnetic field (see Figs 5 and 6) is easy to understand from Figs 4(a) and 7(a), and from the definition of $M_x = (g_+ - g_-)\cos\varphi(\zeta)$. As shown above, for $h > h_c$, the particles whose magnetic moments are oriented along the director begin to accumulate in the middle of the layer [see Fig. 4(b)], while the concentration of the particles, whose magnetic moments are directed against the director, decreases not only in the middle but also over the whole layer [see Fig. 4(c)]. We only note that in the fields slightly above the Freedericksz field, the difference $(g_+ - g_-)$ increases faster [see Fig. 7(a)], than $n_x = \cos\varphi$ decreases. For this reason, above h_c, the magnetization increases with the field increasing, according to Eq. (25). However, at $h \approx 1$, most of the magnetic admixture is concentrated near the middle of the layer [see Figs 3(b) and 7]. In this case, as shown in Figs 7(a) and 3(a), the difference between the concentrations of subsystems $(g_+ - g_-) \to 1$ and the angle φ_0 tends to $\pi/2$, so $\cos\varphi$ decreases. Consequently, at $h \geq 1$, the behavior of the magnetization is mainly determined by the director angle, so the growth of $\langle M_x \rangle = \langle (g_+ - g_-)\cos\varphi(\zeta) \rangle$ is replaced by a decrease (see Fig. 5, dashed curve), and $\langle M_y \rangle = \langle (g_+ - g_-)\sin\varphi(\zeta) \rangle$ continues to grow, trying to saturation.

6. Conclusions

A new class of soft materials—colloidal suspensions of ferromagnetic or ferroelectric particles in LCs—have attracted much attention of researchers. They are interesting not only from the standpoint of practical applications in information display devices, but also with the physical point of view as a medium, the physical properties of which are much richer than properties of each of its components separately. Many of these remarkable properties are related to the elastic distortions of the optical axis of LC that arise when an external field is applied to the crystal and colloidal particles embedded in it. In this paper, we used the model of the collective response of FN suspension to the applied field [1].

We studied the magnetic field induced Freedericksz transition in a flat layer of compensated FN whose boundary conditions included rigid planar coupling. The coupling of magnetic particles with the LC matrix was assumed to be absolutely rigid, and the long axes of the magnetic grains were in parallel and antiparallel to the director. The magnetic field was applied so that the resulting deformation of the orientational structure corresponded to pure twist. With the help of continuum theory, the equations were derived, describing the distortions of the director, the distribution of magnetic particles, and the magnetization of FN in an external magnetic field. It is shown that orientational distortions in the increasing field have threshold character (the Freedericksz transition), and analytical expression for the Freedericksz field has been obtained. It was found out that the Freedericksz transition in compensated FN is the second-order phase transition. It is shown that the presence of compensated magnetic admixture decreases the transition field compared to pure LC. Spatial distortions of the director, concentration of magnetic admixture and magnetization of

FN has been obtained numerically as functions of magnetic field. Near the Freedericksz transition field, the analytical expressions for the director, magnetic susceptibility, and the average FN magnetization were obtained.

We have predicted that segregation effect leads to nontrivial magnetic response of a FN. This effect is the main reason for the magnetization of compensated FN. With the increasing of magnetic field initially, the concentration of the particles which magnetic moments are directed along the director increases in the middle of the layer, and the concentration of the particles with magnetic moments directed opposite to the director is decreased. With further field increasing, the region, where the particles accumulate, extends from the center of the layer to the periphery. Thus, the effect of segregation leads to the fact that under the magnetic field influence, the FN in the threshold manner ceases to be compensated and is magnetized in the field direction. We have predicted also the nonmonotonic behavior of the concentration of "+ family" particles and the transverse magnetization with the field increasing.

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References

[1] Brochard, F., & de Gennes, P. G. (1970). *J. de Physique (France)*, *31*, 691.
[2] Reshetnyak, V. (2004). *Mol. Cryst. Liq. Cryst.*, *421*, 219.
[3] Reshetnyak, V. Yu., Shelestiuk, S. M., & Sluckin, T. J. (2006). *Mol. Cryst. Liq. Cryst.*, *454*, 201.
[4] Shelestiuk, S. M., Reshetnyak, V. Yu., & Sluckin, T. J. (2011). *Phys. Rev. E*, *83*, 041705.
[5] Reznikov, Yu., Buchnev, O., Tereshchenko, O., Reshetnyak, V., Glushchenko, A., & West, J. (2003). *Appl. Phys. Lett.*, *82*, 1917.
[6] Li, F., Buchnev, O., Cheon, C. I., Glushchenko, A., Reshetnyak, V., Reznikov, Yu., Sluckin, T. J., & West, J. L. (2006). *Phys. Rev. Lett.*, *97*, 147801.
[7] Cook, G., Reshetnyak, V. Yu., Ziolo, R. F., Basun, S. A., Banerjee, P. P., & Evans, D. R. (2010). *Optics Express*, *18*, 17339.
[8] Burylov, S. V., & Raikher, Y. L. (1995). *Mol. Cryst. Liq. Cryst.*, *258*, 123.
[9] Zakhlevnykh, A. N. (2004). *J. Magn. Magn. Mater.*, *269*, 238.
[10] Zakhlevnykh, A. N., & Makarov, D. V. (2007). *Mol. Cryst. Liq. Cryst.*, *475*, 233.
[11] Makarov, D. V., & Zakhlevnykh, A. N. (2008). *J. Magn. Magn. Mater.*, *320*, 1312.
[12] Makarov, D. V., & Zakhlevnykh, A. N. (2010). *Phys. Rev. E*, *81*, 051710.
[13] Zakhlevnykh, A. N., & Makarov, D. V. (2011). *Mol. Cryst. Liq. Cryst.*, *540*, 135.
[14] Zakhlevnykh, A. N., & Semenova, O. R. (2011). *Mol. Cryst. Liq. Cryst.*, *540*, 219.
[15] Blinov, L. M., & Chigrinov, V. G. (1994). *Electrooptic Effects in Liquid Crystal Materials*, Springer-Verlag: New York.