ORIENTATIONAL AND OPTICAL BISTABILITIES IN THE CELL FILLED WITH FERROMAGNETIC PARTICLES

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We have developed an approximate theory of orientational and optical bistabilities in a ferronematic – colloidal suspension of single-domain needle-like ferroparticles in a nematic liquid crystal. The theory is based on a new analytical approach and accounts for the interaction of magnetic moments of particles with an external magnetic field, anchoring-induced ferronematic interaction, and redistribution of the ferroparticle concentration induced by a magnetic field (segregation effect). The homeotropic anchoring of nematic molecules is assumed at the cell bounding surfaces and ferroparticle surfaces. Possibilities to control the bistability effect are discussed.

1. Introduction

In recent years, a lot of attention has been given to suspensions of colloidal magnetic particles in thermotropic nematic liquid crystals (NLC), where mesophases exist in a certain temperature range between the solid state and the isotropic liquid one. Thermotropic ferronematics (FN) are highly dispersed colloidal suspensions of single-domain anisometric (e.g., needle-like) ferro- or ferrimagnetic particles in thermotropic NLCs. These materials have all properties inherent to the liquid-crystal phase. At the same time, the magnetic susceptibilities of these suspensions are by several orders of magnitude higher than that of pure NLCs. Ferronematics were proposed in work [1], where the continuum theory of this system was first developed. Later on, the continuum theory of ferronematics was generalized in [2] to finite anchoring energies between nematic molecules and the ferroparticle surface. The first thermotropic FN was synthesized in work [3]. Chen and Amer [4] were the first able to construct a FN suspension of cylindrical maghemite (γ - Fe_2O_3) ferroparticles embedded in N-(4-methoxybenzylidene)-4-butylaniline (MBBA) and studied the orientation distortion of a FN cell under the action of an external magnetic field. They showed that the system is sensitive even to weak magnetic fields comparable with the Earth's magnetic field ($\sim$ 0.7 Oe). The drawback of this system was its instability which manifested itself in the aggregation of ferroparticles. The lifetime of the system slightly exceeded the measurement time in these experiments. During the last decade, there is significant progress in synthesizing the thermotropic FNs stable to the aggregation [5, 6], in particular in the study of partially aggregated FN systems with the high response to a low applied magnetic field ($\sim$ 5 Oe) [7]. In the present paper, we consider FNs based on thermotropic liquid crystals. Crucial is the assumption of anisometric (elongated) ferroparticles, resulting in the co-directionality of the magnetic moment vector of a ferroparticle and its long axis [8]. The long axes of the ferroparticles rotate in space under the action of an external magnetic field. Therefore, a distortion of the entire LC matrix occurs due to the energy of anchoring between LC molecules and the surface of ferroparticles and due to the long-range interaction between LC molecules. This results in changing the optical properties of a FN. In particular, the effective refractive index changes, and FN turns out to be birefringent. This gives a possibility to create the low-power-consuming sensors of weak magnetic fields and the devices for the image visualization. A distortion of the orientational structure of a FN leads to the redistribution of ferroparticles in a cell. This effect is known as the segregation and is observed under the application of an external magnetic field [1, 2, 9–12]. An increase in the magnetic field strength leads to the migration of colloidal particles toward the region, where the free energy of a FN is minimized. The consequence is an increase in the local concentration of particles. This increase can occur inside the cell, when the same type of anchoring (homeotropic [2, 9, 11] or planar [10]) between nematic molecules and the surfaces of the cell and ferroparticle surfaces takes place. The other possibility is increasing the local particle concentration close to the cell boundaries in the case of the hybrid coupling (e.g., the planar anchoring at the cell boundaries and the homeotropic one at the particle surfaces [12]).

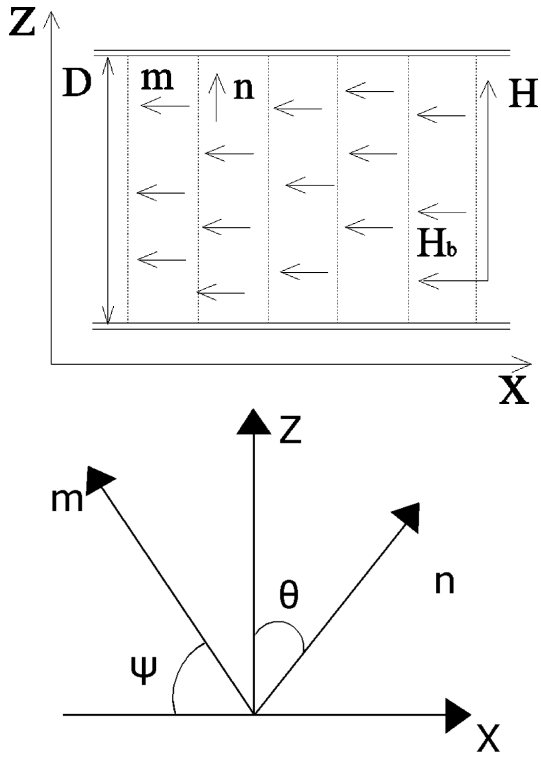

Fig. 1. Schematic of a ferronematic cell

In contrast to the classical Friedericksz transition in a pure NLC, the reorientation of nematic molecules in a FN shows the thresholdless behavior, when applying a magnetic field. When the strength of an external field increases, the nematic cell distortion increases as well. Under specific conditions, the theoretically predicted inverse Friedericksz transition [9–11] can be observed - the nematic response increases at low fields, reaches a maximum, then decreases and reaches zero at a certain value of the magnetic field. At a considerable segregation in the system, the orientational and, as a result, optical bistability effects can also be observed [9–12] In this case, one magnetic field strength value corresponds to two values of the nematic and ferroparticle angle deviation inside the cell. In this paper, we propose an approximate analytic description of the bistability effect and possible methods of its control. We compare the obtained analytic result with numerical computer calculations carried out with the help of accurate formulas [9, 11].

2. Model

We consider a ferronematic cell of thickness D (see Fig. 1) that contains a colloidal suspension of ferroparticles embedded into the nematic host. We suppose the strong homeotropic nematic anchoring at the cell walls (for instance, $W_s > 0.1$ erg/cm^2) and the soft anchoring (see details further) at the ferroparticle surface. This enables us to neglect a change in the nematic director angle at the cell walls under applying a magnetic field.

In the initial state ($H = 0$), the needle-like ferroparticles are homogeneously distributed in the nematic host. The surface of each ferroparticle is treated with surfactants in order to make all magnetic moments be perpendicular to the unperturbed director $\mathbf{n_0}$ (homeotropic coupling). The density of the surface energy anchoring between nematic molecules and ferroparticles W_p satisfies the weak coupling condition $W_p d/K \leq 1$ [2], where K is the mean value of the NLC elastic constants. The size and the shape of ferroparticles are chosen so that they are monodomain, and the magnetic moment of each ferroparticle is directed along its long axis. This assumption is correct for ferroparticles of lengths $L \leq 0.3$ μm with the length-to-diameter ratio $L/d \geq 3$ [8]. We note that the authors of work [4] have investigated the FN which include maghemite needle-like ferroparticles with the length-to-diameter ratio $L/d = 7$. The cell is subjected to an external magnetic field which has components perpendicular, H (control field; we restrict ourselves to the weak fields $H < 500$ Oe), and parallel, H_b, to the cell. The weak field $H_b \leq 0.6$ Oe aligns initially the ferroparticles in one direction at $H = 0$. The bias field holds ferroparticles in the XZ plane. The system is homogeneous in directions of the XY axes. As an example of the FN system, we consider a colloidal suspension of magnetite (Fe_3O_4) ferroparticles in the nematic host 4-cyano-4′-pentylbiphenyl (5CB). To make contact with experiment, we use typical material and experimental parameter values. The value of the anisotropic part of the diamagnetic susceptibility of the nematic is $\chi_a = 1.7 \times 10^{-7}$ [13]. The saturation magnetization per unit volume within an individual colloidal particle is $M_s = 485$ G [9]. The mean ferroparticle concentration is chosen to be $\bar{c} = 1 \times 10^{10}$ cm^{-3}, the particle length $L = 0.2$ μm, diameter $d = L/3$, and volume $v = 7 \times 10^{-16}$ cm^3 that corresponds to the volume fraction $\bar{f} = \bar{c}v = 7 \times 10^{-6}$. The anchoring energy density at the colloidal particle surface is $W_p = 7.6 \times 10^{-3}$ erg/cm^2, $K \approx 6.7 \times 10^{-7}$ dyn [11]. The cell thickness is chosen to be $D = 130$ μm. A free energy functional can be written in the form [2]

$$F = \int\limits_V \left\{ \frac{1}{2} \left[K_1 (\nabla \cdot \mathbf{n})^2 + K_2 (\mathbf{n} \cdot \nabla \times \mathbf{n})^2 + \right.\right.$$

$$+\ K_3(\mathbf{n}\times\nabla\times\mathbf{n})^2\big] - \frac{1}{2}\chi_a(\mathbf{n}\cdot\mathbf{H_s})^2 + \frac{fk_{\rm B}T}{v}\ln f -$$

$$-M_s f(\mathbf{m}\cdot\mathbf{H_s}) + f\frac{W_p}{d}(\mathbf{n}\cdot\mathbf{m})^2\Big\}\, dV, \qquad (1)$$

where $K_{1,2,3}$ are Frank elastic constants, and $\mathbf{H_s} = \mathbf{H} + \mathbf{H_b}$. The terms in square brackets are the Frank elastic energy density of the nematic. The remaining terms are, respectively, the direct magnetic energy density, contribution of the mixing entropy of an ideal ferroparticle solution at a given temperature T, magnetic energy of colloidal particles, and anchoring-induced ferronematic interaction. We ignore the magnetic dipole-dipole interaction energy of ferroparticles which disappears at low ferroparticle concentrations $f < 10^{-4}$ [14]. In this geometry, the nematic director and the magnetic director (a unit vector in the direction of the ferroparticle magnetization) are given by expressions $\mathbf{n} = (\sin\theta, 0, \cos\theta)$ and $\mathbf{m} = (-\cos\psi, 0, \sin\psi)$, respectively. The quantities $\theta \equiv \theta(z)$ and $\psi \equiv \psi(z)$ are the angles of deviation of the nematic and magnetic directors, respectively, from their initial directions at $H = 0$. The boundary conditions are $\theta(0) = \theta(D) = 0$. In this case, the free energy functional in the two-constant approximation $K_1 = K_3 = K$ reduces to

$$F = \int_0^D \left[\frac{1}{2}K\left(\frac{d\theta}{dz}\right)^2 - \frac{1}{2}\chi_a(H\cos\theta - H_b\sin\theta)^2 +\right.$$

$$+\ \eta(z)\frac{\bar{f}k_{\rm B}T}{v}\ln\eta(z) - M_s\eta(z)\bar{f}(H\sin\psi + H_b\cos\psi) +$$

$$\left. +\ \eta(z)\frac{\bar{f}W_p}{d}\sin^2(\theta-\psi)\right]\, dz, \qquad (2)$$

where $\eta(z) = f(z)/\bar{f}$. The function $\eta(z)$ describes a change in the local volume fraction of colloidal particles induced by the segregation effect and obeys the ferroparticle-number conservation $\int_0^D \eta(z)dz = D$. In what follows, we neglect the second term in (2) connected with the direct magnetic-nematic interaction. This term is small as compared with the fourth (linear in field) one in weak fields $H < 500$ Oe. The dimensionless form of functional (2) is as follows:

$$F = \int_0^1 d\tilde{z}\left[\frac{1}{2}K\left(\frac{d\theta(\tilde{z})}{d\tilde{z}}\right)^2 + \tilde{\eta}(\tilde{z})t\ln\tilde{\eta}(\tilde{z}) -\right.$$

$$-\tilde{\eta}(\tilde{z})\left[h\sin\psi(\tilde{z}) + h_b\cos\psi(\tilde{z})\right] +$$

$$\left. +\tilde{\eta}(\tilde{z})w\sin^2\left(\theta(\tilde{z}) - \psi(\tilde{z})\right)\right], \qquad (3)$$

where $t = \frac{k_{\rm B}T\bar{f}D^2}{Kv}$ is the dimensionless temperature (roughly, the ratio of the thermal energy of colloidal particles to the nematic elastic free energy density) [9, 11], $w = \frac{\bar{f}D^2}{K}\frac{W_p}{d}$ the dimensionless coupling, $h = \frac{\bar{f}M_sHD^2}{K}$ the dimensionless magnetic field, $h_b = \frac{\bar{f}M_sH_bD^2}{K}$ the dimensionless bias magnetic field, and $\tilde{z} = \frac{z}{D}$ the dimensionless coordinate. In these variables, the normalization condition of the function $\eta(z)$ has the form $\int_0^1 \eta(\tilde{z})d\tilde{z} = 1$. Note that the parameter t controls the extent of segregation in the system: it decreases, as the segregation increases, and vanishes in the limiting case $t \to \infty$ [9]. The increase in the control magnetic field strength leads to the rotation of ferroparticles, which induces the nematic director field distortion in the presence of the nematic anchoring at the particle surface. The accurate numerical computations [10, 11] show that the two stable states for a nematic orientation (orientational bistability) can arise at $t < t_c$ in a certain control field range. Here, $t_c \approx \frac{w_c}{2}\left(\frac{w}{w_c}\right)^3$ is a certain value of the dimensionless temperature which depends on the value of w; $w_c = \pi^2/2$. A change in the bias field strength H_b leads to a change in the width of the control field range, where the bistability is observed. This provides us with the possibility to control the bistability effect.

3. Finding the Equilibrium State of a Ferronematic

For further solving the problem, we introduce a new variable $\gamma(\tilde{z}) = \pi/2 - \psi(\tilde{z})$ and expand $\gamma(\tilde{z})$ in a Fourier series [9–11], by restricting ourselves to the first term $\gamma(\tilde{z}) = \gamma_0\sin\pi\tilde{z}$. The same procedure can be used for $\theta(\tilde{z}) = \theta_0\sin\pi\tilde{z}$. The parameter $\eta(\tilde{z})$ can be represented as two terms of the Fourier series $\eta(\tilde{z}) = 1 - 2s\cos 2\pi\tilde{z}$ [9–11], where the segregation parameter $0 \leqslant s \leqslant 1$ can be treated as the order parameter describing the redistribution of ferroparticles within the colloidal volume. The values $s = 0$ and $s = 1$ correspond, respectively, to the unsegregated and perfectly segregated states. After substituting the truncated Fourier series for $\gamma(\tilde{z})$, $\theta(\tilde{z})$ and $\eta(\tilde{z})$ into the scaled free energy functional (3), expanding the trigonometric and logarithmic functions into the power series in γ_0, θ_0 and s up to the fourth-order terms, and integrating over $\tilde{z}$, we derive the following

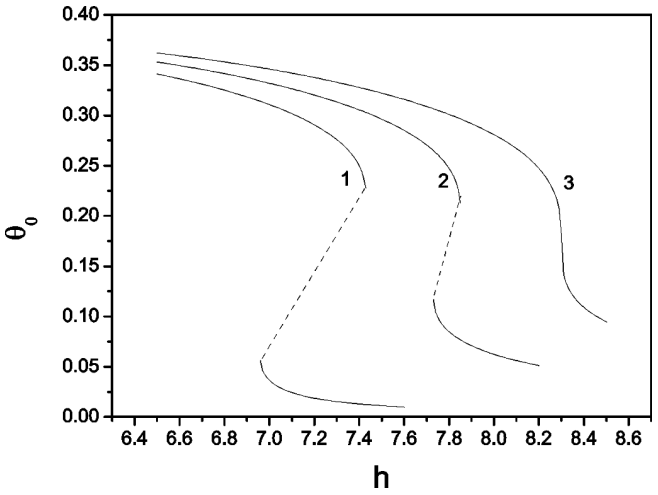

Fig. 2. Dependence of the deviation of the nematic angle $\theta_0(h)$ on the control field calculated from Eqs. (5), (6), and (7) for different values of the bias field. *1* – $h_b = 0.01$, *2* – $h_b = 0.08$, *3* – $h_b = 0.15$

approximate expression for the variable part of the cell free energy:

$$F \approx -(\theta_0+\gamma_0)^2 w(1+s) + (\theta_0+\gamma_0)^4 w\left(\frac{1}{4}+\frac{s}{3}\right) -$$

$$-\frac{h}{32}\gamma_0^4 + \frac{h}{2}\gamma_0^2 + w_c\theta_0^2 - \frac{h}{24}\gamma_0^4 s + 2ts^2 + \frac{h}{2}\gamma_0^2 s -$$

$$-\frac{4h_b}{3\pi}\gamma_0 s + \frac{4h_b}{9\pi}\gamma_0^3 + \frac{8h_b}{15\pi}\gamma_0^3 s + ts^4. \qquad (4)$$

Further, we find a minimum of the free energy, by using the standard condition $\frac{\partial F}{\partial\theta_0} = \frac{\partial F}{\partial\gamma_0} = \frac{\partial F}{\partial s} = 0$. From equation $\frac{\partial F}{\partial\theta_0} - \frac{\partial F}{\partial\gamma_0} = 0$, we express θ_0 in terms of the variables s and γ_0:

$$\theta_0 \approx \frac{h\gamma_0}{2w_c}(1+s) - \frac{2h_b}{\pi w_c}\left(1+\frac{2s}{3}\right) -$$

$$-\frac{h}{16w_c}\gamma_0^3\left(1+\frac{4s}{3}\right) + \frac{2h_b}{\pi}\gamma_0^2\left(1+\frac{2s}{5}\right). \qquad (5)$$

The condition $\frac{\partial F}{\partial\theta_0} = 0$ yields the equation

$$2(\theta_0+\gamma_0)w(1+s) - 4(\theta_0+\gamma_0)^3 w\times$$

$$\times\left(\frac{1}{4}+\frac{s}{3}\right) - 2w_c\theta_0 = 0. \qquad (6)$$

The equation for the last variable γ_0 can be found directly from $\frac{\partial F}{\partial s} = 0$:

$$4ts(1+s^2) + \left(\frac{8h_b}{15\pi}+\frac{4\theta_0}{3}\right)\gamma_0^3 + \left(-w+\frac{h}{2}+2\theta_0^2 w\right)\gamma_0^2 +$$

$$+\left(\frac{4\theta_0^3 w}{3} - \frac{8h_b}{3\pi} - 2\theta_0 w\right)\gamma_0 - \theta_0^2 w = 0. \qquad (7)$$

Here, we neglect the terms of orders γ_0^4 and θ_0^4. As a result, we obtain a system of algebraic transcendent equations in the variables θ_0, γ_0, and s (5, 6, 7) suitable for our analysis.

4. Orientational and Optical Bistabilities

The expressions we have obtained in the previous section allow us to analyze a state of the system under the influence of a given external magnetic field. In what follows, we will consider the case of the weak coupling between the nematic and ferroparticles ($w < w_c$) using the terminology accepted in [9–11]. Exactly in this regime, the inverse Friedericksz transition arises. As was shown in [9–11], the orientational bistability can appear on the decreasing branch of the nematic angle dependence on the magnetic field if $t < t_c$. In our calculations, the following values of the dimensionless parameters were used: $w = 2$, $t = 0.1$, and $t_c \approx 0.164$. Figure 2 illustrates the $\theta_0(h)$ dependence numerically calculated from Eqs. (5), (6), and (7). The figure demonstrates that one value of the control field h corresponds to two values of the nematic deviation angle. There are two specific switching points on the $\theta_0(h)$ curves, where the abrupt transition between two stable branches of solution occurs. The segment of the $\theta_0(h)$ dependence between two switching points corresponds to the physically unstable states of a ferronematic. On the lower branch of the solution at high values of h, the nematic director is almost perfectly oriented along the control field. A decrease in the control field leads to a deviation of the nematic director from this direction. When this deviation reaches the certain value, a jump to the upper branch of $\theta_0(h)$ (the first-order phase transition [11, 12]) occurs. The dependences $\gamma_0(h)$ and $s(h)$ have the same form and are not presented here. An orientational distortion of the FN structure under the influence of the control magnetic field leads to a redistribution of the ferroparticle concentration in the cell, which is described by the dependence $s(h)$. When the value s reaches zero, the ferroparticles become homogeneously distributed over the cell volume.

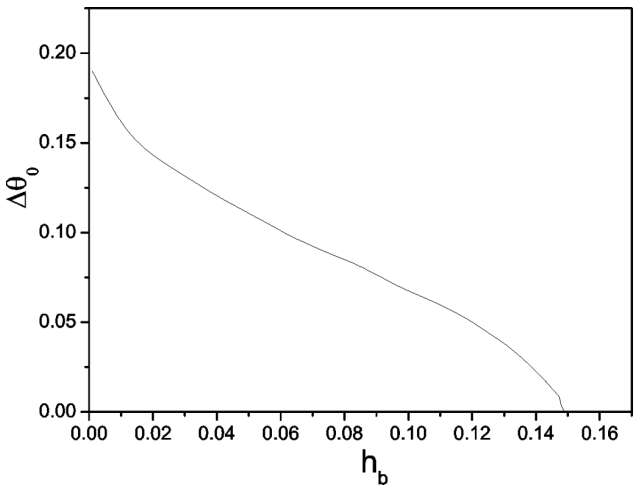

Fig. 3. Dependence of the distance between switching points of the nematic angle deviation $\Delta\theta_0(h_b)$ on the bias field h_b, calculated from Eqs. (5), (6), and (7)

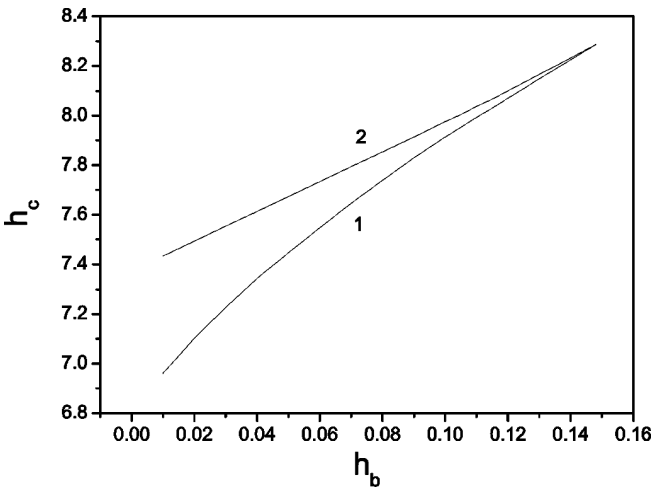

Fig. 4. Dependence of the switching point positions on the bias field calculated from (8). *1* – lower branch of $\theta_0(h)$, *2* – upper branch of $\theta_0(h)$

The intermediate values $0 < s < 1$ orrespond to an inhomogeneous distribution of ferroparticles in the cell (segregation effect). With increase in the control magnetic field, the ferroparticles migrate to the center of the cell minimizing the free energy of the FN. Our analysis of the dependences $\theta_0(h)$, $\gamma_0(h)$, $s(h)$ show that an increase in the bias field h_b induces a decrease of the distance between the switching points for all variables. The specific value of h_b exists, where the orientational bistability disappears, and one value of the control field h corresponds to one value for each of the three system parameters taken separately. The response of the system to a weak bias magnetic field is considerable. This feature makes it possible to control the orientational bistability in ferronematics.

The lower switching points in Fig. 2 are found from the analytic expressions (5), (6), and (7). Their positions are almost equal to those obtained by the numerical minimization of functional (3); positions of the upper switching points differ by 30%-40% that can be explained by using the approximate method of expansion in a series. We note that the bistability predicted with the approximate expressions 5), (6), and (7) disappears at lower values of the bias field h_b as compared with the case of the numerical minimization of functional (3). Figure 3 shows the dependence of the distance between switching points for the function $\theta_0(h)$ on the bias field h_b. This dependence calculated from Eqs. (5), (6), and (7) is in a qualitative agreement with the results of numerical calculations in [9, 11].

We can derive an expression which describes the position of the switching points for different values of the bias field h_b (see Fig. 4). This expression can be obtained from the condition for the derivatives at the points of stability loss to tend to infinity. Let us introduce the following designations: $\varphi(\theta_0, \gamma_0, s) = -2w(1+s) + 12(\theta_0+\gamma_0)^2 w\left(\frac{1}{4}+\frac{s}{3},\right)$ and $\psi(\theta_0, \gamma_0) = -2(\theta_0+\gamma_0)w + \frac{4}{3}w(\theta_0+\gamma_0)^3$. Then the switching points can be described by the equation

$$\left(\varphi + h + hs - \frac{3h}{8}\gamma_0^2 + \frac{8h_b}{3\pi}\gamma_0 + \frac{16h_b}{5\pi}\gamma_0 s\right)\left[(\varphi + 2w_c) \times\right.$$

$$\left.\times\,(4t - 12ts^2) - \psi^2\right] + \left(\psi + h\gamma_0 - \frac{h}{6}\gamma_0^3 - \frac{8h_b}{3\pi} + \frac{8h_b\gamma_0^2}{5\pi}\right) \times$$

$$\times \left[2\varphi\psi - (\varphi + 2w_c)\left(\psi + h\gamma_0 - \frac{h}{6}\gamma_0^3 - \frac{8h_b}{3\pi} + \frac{8h_b\gamma_0^2}{5\pi}\right)\right] -$$

$$-\varphi^2(4t - 12ts^2) = 0. \qquad (8)$$

The direct consequence of the orientational bistability in the FN system is the optical one. The magnetic-field-induced orientational structure of the FN turns out to be birefringent to a light beam normally incident on the lower cell wall (in parallel to the Z-axis). The optical phase lag of rays can be calculated from the expression

$$\delta = \frac{2\pi}{\lambda}\int_0^D (n - n_0)dz, \qquad (9)$$

where λ is the wavelength of the normally incident laser light; n_0 is the ordinary index of refraction of

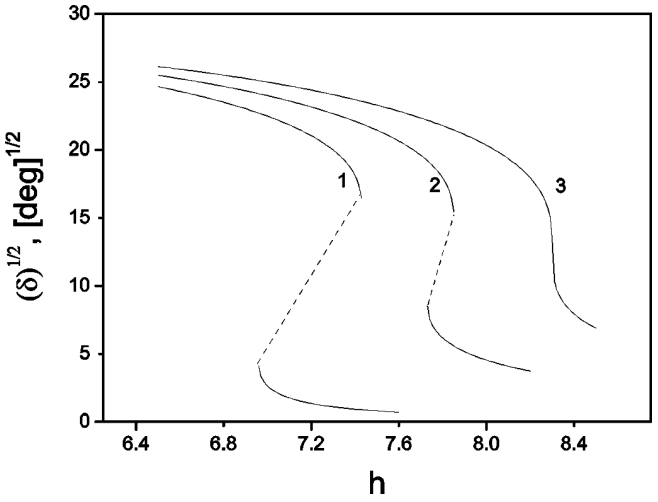

Fig. 5. Dependence of the phase lag of ordinary and extraordinary rays $\sqrt{\delta(h)}$ on the control field for different values of the bias field *1* – $h_b = 0.01$, *2* – $h_b = 0.08$, *3* – $h_b = 0.15$

the nematic; and n is the effective index of refraction which depends on the nematic angle $\theta_0(z)$. The dependence of n on z is given by the well-known expression $n^{-2} = n_0^{-2}\cos^2\theta(z) + n_e^{-2}\sin^2\theta(z)$, where n_e is the extraordinary index of refraction of the nematic. After substituting n in Eq. (9) and nondimensionalizing, we have

$$\delta = \frac{2\pi D}{\lambda}\int\limits_0^1 \left\{ \frac{n_0 n_e}{\left[n_e^2\cos^2\theta(z) + n_0^2\sin^2\theta(z)\right]^{1/2}} \right\} d\tilde{z}, \quad (10)$$

where $\theta(\tilde{z}) = \theta_0 \sin\pi\tilde{z}$ is the approximation of $\theta_0(z)$ with the Fourier series terms that are used in our analytic calculations. To make contact with experiment, we use the following parameter values: $n_0 = 1.5309$, $n_e = 1.7063$, and $\lambda = 0.6328$ μm [13]. Figure 5 illustrates the field dependence of the phase lag of rays calculated from Eqs. (5), (6), and (7). The effect of the optical bistability and its relationship to the orientational bistability are clearly seen from Figs. 2 and 5.

5. Conclusions

In this paper, we have developed an approximate theory of orientational and optical bistabilities in the ferronematic system, which appears at a significant segregation in the system when $t < t_c$. We have obtained the simple analytic expression (4) for the FN free energy allowing us to give the analytic description of this effect. We have found analytic expressions for calculating the nematic director distortion angle θ, the magnetic director distortion angle ψ, and the segregation order parameter s as functions of the applied external magnetic field in the bistability range. The approximate method developed in this paper enables us to give the qualitative description of the orientational and optical bistabilities in the ferronematic system. It is shown that positions of the lower switching points in hysteretic curves calculated from the approximate analytic theory are close to those calculated from the numerical minimization of the model functional (3); the upper switching points differ by $30\% - 40\%$ from those derived numerically. Changing the value of the weak bias field h_b, we can vary the distance between the switching points and thus to control the bistability effect. The high magnetic susceptibility of the system and the ability to control the bistability effect by means of a weak bias magnetic field open prospects to produce magnetic-sensitive sensors, magnetic switchers, and devices for processing and information storage in near future.

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ОРІЄНТАЦІЙНА ТА ОПТИЧНА БІСТАБІЛЬНІСТЬ У КОМІРЦІ З ФЕРОМАГНІТНИМИ ЧАСТИНКАМИ

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Р е з ю м е

Побудовано наближену теорію ефекту орієнтаційної та оптичної бістабільності в феронематику – колоїдній суспензії однодоменних стрижеподібних ферочастинок у нематичному рідкому кристалі. Теорія ґрунтується на новому аналітичному підході і враховує взаємодію магнітних моментів частинок з зовнішнім магнітним полем, індуковану зчепленням нематика з поверхнею ферочастинок взаємодію і індукований магнітним полем перерозподіл концентрації ферочастинок (ефект сегрегації). Припускається, що на обмежуючих поверхнях комірки та поверхнях ферочастинок реалізуються гомеотропні умови зчеплення молекул нематика. Обговорено можливості керування явищем бістабільності.