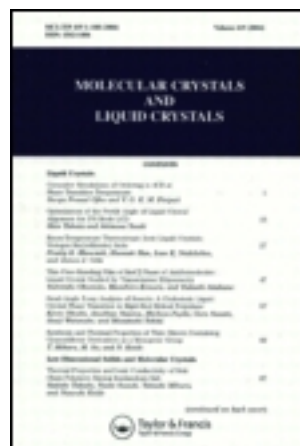




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Magnetic Field Induced Orientational Bistability in a Ferronematic Cell

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The equilibrium states of a suspension of single-domain ferroparticles in a nematic liquid-crystalline homeotropic cell subject to an external magnetic field are studied. We predict the existence of magnetic field induced orientational bistability in such a system in a magnetic field $\sim 10^2$ Oe. The existence of the bistability phenomenon is governed by conditions on the cell thickness and on the director anchoring energy. The effect can be controlled using a small bias magnetic field normal to the unperturbed director. The director reorientation in a magnetic field causes an effective change in refraction index, which enables the orientational bistability to be exploited in optical devices.

Keywords: liquid crystal; ferroparticle; magnetic field; bistability

INTRODUCTION

In the last ten years there has been increasing interest in liquid crystals (LC) containing magnetic particles. These systems are both of general-physical interest and have considerable potential for use in technical, in part optical, applications^[1–4]. The key property of a nematic crystal doped with single-domain ferromagnetic particles, known as a ferronematic (FN), is the inherent high magnetic susceptibility making it possible to exert control of its orientational texture using weak (< 10 Oersteds) magnetic fields.

Magnetic suspensions in nematic liquid crystals were proposed and considered by Brochard and de Gennes^[1]. Much later thermotropic FN's were synthesised by Chen and Amer^[5] using a MBBA LC matrix. The initial continuum theory^[1] was generalized by Burylov and Raikher^[6] to include a finite nematic anchoring energy at the ferro-particle surface. This provided an explanation for the absence of co-alignment of the nematic director and averaged local magnetization in the FN, as was found experimentally (see, for example, Ref. ^[7]). The generalized theory has been confirmed in experiments, among them one on 8CB-based FN with almost spherical magnetite particles ^[2].

Since these early studies, peculiarities have been noted in behavior of field dependencies of birefringence in ferronematics containing needle-like magnetic particles. The phase retardation between the extraordinary and ordinary rays exhibits saturation or nonmonotonic behavior in the fields ≥ 10 Oe, depending upon cell thickness and the concentration of ferroparticles. There are phase retardation oscillations caused by ferroparticle coagulation as well as a monotonic increase in the phase retardation followed by a decrease for fields ≥ 40 Oe. These were confirmed by numerical simulations reported in Ref. ^[6]. In this paper, we demonstrate the consequences of this feature for a homeo-

tropic FN cell. Specifically, we predict the existence of a new phenomenon: magnetic field induced orientational bistability in nematic liquid crystals containing monodomain ferromagnetic particles.

FORMULATION OF THE PROBLEM

Let us consider a cell of thickness D filled with FN. We suppose homeotropic nematic anchoring of finite strength W_0 and W_D at the lower ($z = 0$) and upper ($z = D$) cell planes, respectively. We also suppose an anchoring energy of strength W_P at the ferroparticle surfaces favoring soft homeotropic anchoring. The ferroparticles will be modelled as needle-like monodomain ferrite grains of length L and diameter $d \approx L/7$ significantly greater than the nematic molecule size. The cell is initially subjected to a small bias magnetic field H_b parallel to the cell planes. In this initial state ferroparticles are homogeneously distributed over the cell volume and their magnetic moments are perpendicular to the unperturbed director $\mathbf{n}_0$. The sample is magnetized up to saturation, which is attained for the ratio of magnetic field energy to heat energy $\rho_b = M_s \nu H_b / k_B T \geq 10$ [6]. Here M_s is the saturation magnetization of the particle substance, ν is the volume of the particle. For $\gamma\text{-Fe}_2\text{O}_3$ particles $M_s \sim 340$ G, $\nu \sim 2 \times 10^{-15}$ cm³ and we have $\rho_b \geq 10$ at $H_b \geq 0.6$ Oe and $T = 25^\circ$ C.

To observe the orientational bistability phenomenon, an external variable magnetic field H of moderate strength (~ 50 - 150 Oe) should be applied to the cell normally to its planes. The orientational texture of FN and the choice of the reference frame are shown in Figure 1. A magnetic field H rotates the ferroparticles producing distortions within the LC matrix. The volume fraction f of the particles, at given thickness of the cell, will be considered to exceed the critical value $f^* \sim 2K\nu / (\pi d L W_P D^2)$, where K is the mean value of the nematic

elastic constants, yielding the collective behavior of the particles [1,6]. Induced distortions of the particles in this case are imparted to the LC matrix bringing about macroscopic changes in the sample texture.

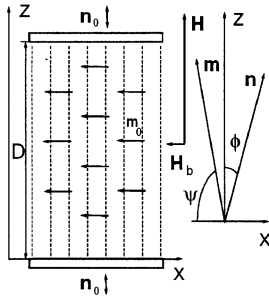


FIGURE 1 Schematic of the FN cell.

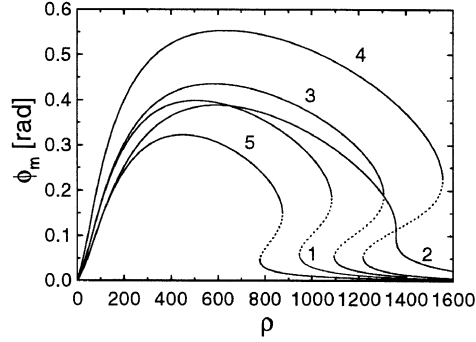


FIGURE 2 The angle ϕ_m vs ρ for $p = 0$
 (1) $D/\lambda = 0.1$, $w_0 = w_D = 3$, $\sigma = 200$, $\rho_b = 10$, (2) the same values, but $\rho_b = 68$.
 (3) $D/\lambda = 0.1$, $w_0 = w_D = 3$, $\sigma = 220$,
 (4) $D/\lambda = 0.12$, $w_0 = w_D = 3$, $\sigma = 200$,
 (5) $D/\lambda = 0.1$, $w_0 = 1000$, $w_D = 3$, $\sigma = 200$; $\rho_b = 10$.

The FN equilibrium state at given $\mathbf{H}_p = \mathbf{H} + \mathbf{H}_b$ can be determined by minimizing the full free energy functional[6]:

$$\begin{aligned}
 F = & \int_V \left\{ \frac{1}{2} \left[K_1 (\operatorname{div} \mathbf{n})^2 + K_2 (\mathbf{n} \cdot \operatorname{curl} \mathbf{n})^2 + K_3 (\mathbf{n} \times \operatorname{curl} \mathbf{n})^2 \right] \right. \\
 & - \frac{1}{2} \chi_a (\mathbf{n} \cdot \mathbf{H}_p)^2 + \frac{f k_B T}{\nu} \ln f - M_s f (\mathbf{m} \cdot \mathbf{H}_p) + \frac{f W_p}{d} (\mathbf{n} \cdot \mathbf{m})^2 \Big\} dV \\
 & + \int_{S_0} \frac{W_0}{2} (\mathbf{n} \times \mathbf{n}_0)^2 dS + \int_{S_D} \frac{W_D}{2} (\mathbf{n} \times \mathbf{n}_0)^2 dS
 \end{aligned} \quad (1)$$

in combination with condition $\int_V f dV = \bar{f}V$, where $\mathbf{m}$ is the unit vector in the direction of the sample magnetization, K_j are elastic constants, χ_a is the anisotropic part of the diamagnetic susceptibility of nematic and $\bar{f}$ is the mean particle volume fraction. The volume integral in Eq. (1) describes the FN bulk free energy in a magnetic field^[6]. The last two terms are the nematic surface potentials.

In this geometry, the director n and the unit vector $\mathbf{m}$ are given by $\mathbf{n} = [\sin \phi, 0, \cos \phi]$, $\mathbf{m} = [-\cos \psi, 0, \sin \psi]$, where $\phi = \phi(z)$ and $\psi = \psi(z)$. In what follows we use the parameters intrinsic to this system: $\lambda = (K_3 \nu / 2 \bar{f} k_B T)^{1/2}$ (the FN magnetic coherence length^[6]) and dimensionless parameters: $\sigma = \nu W_P / d k_B T$, $\rho = M_s \nu H / k_B T$, $w_0 = W_0 D / K_3$, $w_D = W_D D / K_3$, $p = K_1 / K_3 - 1$, and $\kappa = \chi_a k_B T / 2 \bar{f} \nu M_s^2$. Minimization of the functional (1) implies a set of equations for equilibrium profiles $\phi(z)$, $\psi(z)$, $f(z)$ and boundary conditions on the cell planes (see details in Ref. ^[6]). These equations were solved numerically.

RESULTS OF SIMULATIONS AND DISCUSSION

The distortions within the LC matrix increase initially with the magnetic field strength H . When $\rho \geq (1.5 - 2)\sigma$, two types of orientational deformations can occur, depending on cell thickness^[8]. For $D > D_{cr} = (\pi \lambda / \sigma^{1/2}) C$, as ρ increases ferroparticles tend to align parallel to $\mathbf{H}$, forming a high particle concentration zone inside the cell. Here C is a factor whose analytical expression is known only for very large and very small values of w_0 and w_D as compared to unity^[8]. Note that $D_{cr} = D^* \pi C$, where $D^* = \lambda / \sigma^{1/2}$ is the critical value of the cell thickness^[8], above which the collective behavior of the particles takes place. When $D^* < D < D_{cr}$, the nematic director is decoupled from the ferroparticles with increasing ρ and the concentration tends to

be homogeneous. In the transitional region between small and strong decoupling the FN director possesses two steady-state spatial orientations in some range of the magnetic field strengths.

We have performed numerical simulations for cells of thickness $D^* < D < D_{cr}$ such that the nematic director is decoupled from ferroparticles in strong enough magnetic field and orientational bistability can occur. As an illustration we have considered a ferronematic with rod-like magnetite particles with $d \sim 70$ nm and $L \sim 500$ nm ($\rho/H \approx 16$ Oe $^{-1}$ at 25 $^{\circ}$ C). H_b should be ≥ 0.6 Oe ($\rho_b \geq 10$) so as to magnetize FN nearly perfectly. For chosen value of $\sigma = 200$, W_P is 3×10^{-2} erg cm $^{-2}$, which is compatible with known values for real systems (see, for example, Ref. [6]). To ensure the collective behavior of the ferroparticles, the thickness of the cell must exceed the critical value D^* . It follows that the ratio D/λ must be greater than $\sigma^{-1/2}$. This imposes the restriction that $D^2 f > K_3 \nu / 2 \sigma k_B T$. For MBBA-based FN $K_3 \approx 7.5 \times 10^{-7}$ dyn and we have $D^2 f > 8.8 \times 10^{-11}$ cm 2 . As a consequence, for $D = 340$ μ m the mean volume fraction of ferroparticles $\bar{f} = \bar{c} \nu$ must be greater than 7.6×10^{-8} ($\bar{c}$ is the mean number concentration). In our calculations we put $\bar{f} = 1.5 \times 10^{-7}$ and $\chi_a = 0.97 \times 10^{-7}$.

The set of equilibrium equations was solved numerically for the actual ferronematic system. The solutions obtained were tested for stability with respect to perturbations. A numerical investigation showed that the solutions corresponding to the middle part of the hysteresis loop are unstable.

Figure 2 shows the behavior of the hysteresis loop for different values of parameters of the system. The hysteresis loop shifts towards larger values of ρ and the loop area enlarges as σ and/or D/λ increase (curves 1, 3, 4). Curves 1 and 5 illustrate the effect of weak and strong

anchoring at the lower orienting surface of the cell. An increase in H_b leads to a reduction in the loop area and the disappearance of the hysteresis at specific $H_b = H_b^*$ (curves 1 and 2). This permits the phenomenon to be controlled using small bias magnetic field (~ 4.25 Oe in Figure 2).

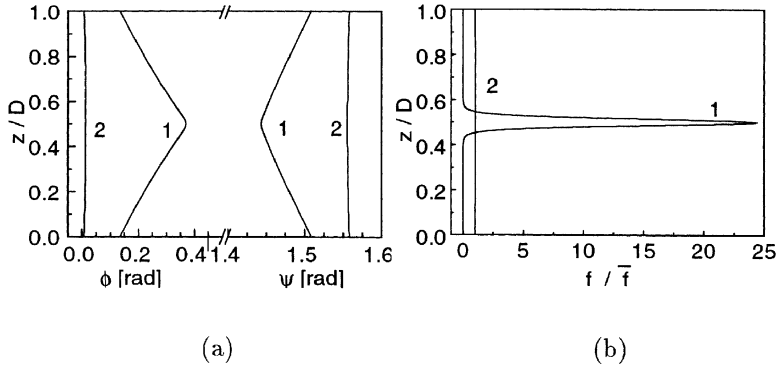


FIGURE 3 Calculated (a) orientational and (b) concentration profiles at $D/\lambda = 0.12$, $w_0 = w_D = 3$, $\sigma = 200$, $\rho_b = 10$, $p = 0$ and $\rho = 1400$. Curves 1 and 2 correspond to the upper and lower parts of the hysteresis loop.

Orientation and concentration profiles inside the FN cell are shown in Figures 3 (a) and 3 (b), respectively, for orientational bistability region. In strong enough magnetic field under condition $D < D_{cr}$ both the nematic director and the magnetic moment of each particle are aligned normally to the planes of the cell and parallel to $\mathbf{H}$ (strong nematic director decoupling; curves 2 in Figure 3 (a)). In this case, the concentration of ferroparticles is uniform within the cell (curve 2 in Figure 3 (b)). As magnetic field decreases, the contribution of the particle-to-director anchoring energy increases. The torque exerted on the nematic by the particle rises, restoring the initial homeotropic

orientation of a ferroparticle in a nematic matrix. An increase in the angle β between local $\mathbf{m}(z)$ and $\mathbf{n}(z)$ results in distortion of the LC matrix. Once the turning point in the hysteresis loop is reached, the system turns into a new stable state and the LC matrix distortion rises sharply (see Figure 3 (a)); ferroparticles transfer inward from the planes of the cell minimizing their orientational energy. Consequently, the ferroparticles gather within a narrow region inside the cell (the segregation effect^[1,6]; see Figure 3 (b)). The degree of the FN layer compression is controlled by entropy springiness of the ferroparticle solution.

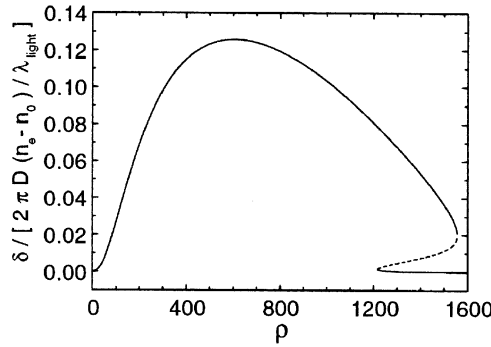


FIGURE 4 Phase retardation vs normalized magnetic field at $\lambda_{\text{light}} = 514.5$ nm, $D/\lambda = 0.12$, $w_0 = w_D = 3$, $\sigma = 200$, $\rho_b = 10$ and $p = 0$.

With increasing magnetic field, the energy goes into rotating the ferroparticles and the consequent decoupling of the nematic director. A decrease in stored elastic energy and magnetic energy of the LC matrix is accompanied by a decrease in the angle ϕ . With decoupling of the nematic director the angle β decreases and the FN stratification decreases. A predominance of the expenditure of energy into rotating

the ferroparticles over decoupling of the nematic director terminates in sharp decoupling of the director. An increase in H_b causes the braking torque to rise. This involves a redistribution of the contributed energies and, as noted above, starting with some H_b^{cr} sharp decoupling changes to smooth one (see Figure 2). In the latter case, there is no hysteresis.

A flat layer of FN is birefringent for light beam propagating along the z-axis. The phase retardation between the extraordinary and ordinary rays depends on the distortion of the LC matrix and exhibits bistable behavior (see Figure 4).

CONCLUSIONS

In conclusion, we have extended previous studies on macroscopic properties of ferronematics in external magnetic field caused by orientational interactions on the ferroparticles surfaces^[8] to the demonstration of new possible effects in a moderate magnetic field. Specifically, we predict the existence of orientational hysteresis in a homeotropic ferronematic cell governed by conditions on the cell thickness and on the director anchoring energy at the ferroparticle surface and planes of the cell. A direct consequence of this effect is magnetic field induced orientational bistability in such a system. The phenomenon under consideration can occur if the thickness of the cell is in the range from D^* to $D_{cr} = D^* \pi C$, where $D^* = (K_3 d / 2 \bar{f} W_P)^{1/2}$, and the magnetic field $H > H_c > \sqrt{2} W_P / M_s d + H_b$. The transition between strong and slightly distorted states of the LC matrix is sharp and is accompanied by sharp change in optical phase retardation of a FN layer. Small bias magnetic fields (< 10 Oe), parallel to the planes of the cell, influence the width of the hysteresis loop, thereby enabling the effect to be controlled. Reasonably strong bias magnetic fields suppress the hysteresis. This effect can be used under optimized conditions in bistable devices,

in polarization analysis, and also for investigating the properties of liquid crystals.

Acknowledgments

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