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Effect of rotating magnetic field on orientational dynamics of ferrocholesteric liquid crystals

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ABSTRACT

We present numerical and analytical results describing the dynamics of a helical orientational structure of a ferrocholesteric liquid crystal, i.e., dilute suspension of ferromagnetic nanoparticles in a cholesteric liquid crystal, under the action of a rotating magnetic field. We employ the continuum theory to show how rotating magnetic field can untwist the helical ferrocholesteric structure and induce a ferrocholesteric–ferronematic transition. We analyze the non-stationary and stationary rotation regimes of the helical structure of a ferrocholesteric in a magnetic field. For weak fields, small and large rotational velocities the analytical expressions are obtained for the pitch of the ferrocholesteric helix. In the stationary rotational regime the orientational phase diagram of the ferrocholesteric–ferronematic transition is constructed for different values of magnetic field and angular velocities. It is shown that with increasing these parameters the transition field decreases. The dependence of a ferrocholesteric pitch on the magnetic field and its angular velocity at various material parameters is numerically obtained. We derive the analytical expression describing the divergence law for the pitch of a helix in the pre-transition region.

1. Introduction

In the past ten years, liquid crystal composite media filled with colloidal nanoparticles have attracted increasing attention from researchers. Such nano-additives, anisometric in shape (rod-like or disc-like), significantly change the magneto-electro-optical characteristics of the liquid crystal used in the development of various display devices [1,2]. In these suspensions as a matrix a liquid crystal (LC) is used, i.e. a liquid consisting of anisometric molecules, interconnected by a strong orientational interaction of quadrupole (or lower) symmetry. Above the temperature of transition to the mesophase, the LC suspensions are isotropic, but upon cooling below a certain temperature (the clearing point), a phase transition to the liquid crystal state occurs in the LC matrix and a long-range orientational order arises, so this suspension retains fluidity and acquires a distinct anisotropy of all physical properties.

These materials have a remarkable quality: remaining in the liquid crystal state at room temperatures, they demonstrate the ability to orientate under the external influences and allow to control the orientation of the particles and the properties of the composite system. The use of elongated nanoparticles as a dispersed medium leads to an orientational anchoring between them and the LC matrix. Dipole (ferromagnetic or ferroelectric) particles embedded in the LC matrix are of particular interest, because they allow to reduce the control fields significantly [3].

Among various LC types, cholesteric liquid crystals (CLCs) have attractive properties for their usage as carrier fluids. They have a helical supramolecular structure and, as a result, remarkable optical properties. These materials demonstrate selective light reflection and giant optical activity, which are easily regulated with magnetic and electric fields and temperatures [4], and are widely used in numerous applications (displays, thermometers, optical storage devices, lasers, etc.). These properties can be controlled with the reversible changing of the pitch of a helical structure under the influence of external fields of different nature.

One of the most interesting effects observed in CLCs is the unwinding of the helical structure by an external magnetic field [4]. When a cholesteric is placed in a constant magnetic field **H**, orthogonal to the axis of the helical structure, the helix pitch increases. With the increase of the magnetic field, above the critical value $H_{c0} = (\pi q_0/2)\sqrt{K_{22}/\chi_a}$ the cholesteric helix is untwisted, i.e., the pitch of the helix becomes infinite, and the cholesteric–nematic transition occurs. Here K_{22} is the twist elastic constant, $\chi_a > 0$ is the anisotropy of the diamagnetic susceptibility of the CLC, q_0 is the intrinsic wave number of the helical structure. In a static magnetic field this phenomenon is well studied [5–7]. A recent paper [8] shows that this transition can be caused by a

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rotating magnetic field, as well as the combined action of the shear stress and magnetic field [9].

The doping of CLCs with nanoparticles of various nature leads to a significant increase in their optical and opto-electronic characteristics, increasing birefringence, dielectric and magnetic anisotropy, which allows to reduce the control fields [10]. Composite materials with magnetic nanoparticles dispersed in the CLC are called ferrocholesterics (FCs). In such systems, due to the orientational anchoring between the needle-like magnetic particles and the CLC matrix, the magnetization vector is spirally twisted in space around some axis. In this respect, an FC is a liquid-crystalline analog of helicoidal ferromagnets or antiferromagnets. Unlike solid ferro- or antiferromagnets, the magnetic moments in FCs are capable of spatial displacement, migrating to those regions of the sample where their magnetic and orientational energies are minimal (the so-called segregation effect [11]). FCs are attractive materials for the applications due to opportunity to change the helicity of their texture and, thereby, to modulate the spectral composition of the reflected light with the help of weak external magnetic fields.

FCs were theoretically predicted by F. Brochard and P.G. de Gennes [11]. The first synthesized ferrocholesterics [12] were not stable. However, in recent years due to the success in the synthesis of helical ferrosuspensions there have appeared a lot of experimental works [13–22], where various properties of these new soft magnetic materials are studied. Available theoretical studies are devoted to the research of the static properties of helicoidal ferromagnetic LC suspensions [23–29], while the dynamic properties of these media are still poorly understood [30,31].

In this paper we theoretically study the unwinding of the helical orientational structure of a ferrocholesteric liquid crystal placed in a rotating magnetic field.

2. Basic equations

As noted above, ferrocholesterics are dilute suspensions of single-domain needle-like particles of a ferromagnet in a CLC. The chirality of CLC molecules leads to the rotation in the direction of preferred orientation of their long axes, resulting in the formation of a spiral structure. The unit vector $\mathbf{n}$ along the direction of preferred orientation is called a director. In the free state, the director of the FC is twisted in space around an axis called the axis of the FC helix. The magnetic field applied to the FC affects both the magnetic moments of the ferroparticles (the dipole mechanism) and the diamagnetic cholesteric matrix (quadrupole mechanism). If the diamagnetic anisotropy of the matrix is positive, and planar anchoring conditions are created on the surface of magnetic particles (i.e., the molecules of the liquid crystal are parallel to the surface of the needle-like ferroparticle), the quadrupole mechanism, along with the dipole mechanism, contributes to the magnetization of the FC in the direction of the field. A magnetic field applied perpendicular to the axis of the spiral structure of an FC, orients the director along the field and increases the pitch of the helical structure. An increase in the field leads to the unwinding of the helix, and at the critical field strength the FC pitch diverges and the FC phase changes to the ferronematic (FN) phase, i.e., the field induces the FC–FN phase transition. Unlike a solid helicoidal ferromagnet, in an FC the process of the helical structure unwinding is complicated by the fact that the role of the anisotropy axis (the optical axis) is played by the CLC director, which itself is oriented by the magnetic field. In the FC, a change in the direction of the director orientation leads to the spatial modulation of the dielectric permittivity, and hence, of the refractive index, which makes it possible to control the optical response.

Let us study the dynamics of the orientational structure of the FC in an external magnetic field, assuming the distribution of the magnetic particles over the volume V of the suspension to be homogeneous with the volume fraction $f = Nv_p/V \ll 1$, where N is the number of particles in the suspension, v_p is the volume of the particle. We assume that the anchoring energy between the CLC molecules and the magnetic particles is large, which corresponds to the so-called rigid anchoring between the director and the magnetization [11]. In this case, the orientational behavior of the liquid-crystalline and magnetic subsystems of the suspension can be described by one vector – the director $\mathbf{n}$. The dynamics of an FC is described by continuum theory [11,4].

Let an uniform magnetic field $\mathbf{H} = H(\cos\omega t, \sin\omega t, 0)$ rotates around the axis of the FC helix z with the angular velocity ω, here t is the time. In this case, the director one can search in the following form:

$$\mathbf{n} = [\cos\varphi(z, t), \sin\varphi(z, t), 0], \tag{1}$$

here φ is the angle of the director and magnetization orientation, measured from the x-axis. In the problem under consideration, the orientational distortions correspond to twist deformations of the director, so the gradients of the flow velocity $\mathbf{v}$ are absent, and there is no backflow effects [32]. Then the equation for the director motion is as follows [4]:

$$\gamma_1 \frac{\partial n_i}{\partial t} = -\frac{\partial F}{\partial n_i} + \nabla_k \frac{\partial F}{\partial(\nabla_k n_i)}, \tag{2}$$

where γ_1 is the coefficient of the rotational viscosity of a liquid crystal [32], and F is the bulk density of the free energy of an FC in a magnetic field [11]

$$F = F_1 + F_2 + F_3, \tag{3}$$

$$F_1 = \frac{1}{2}[K_{11}(\nabla\cdot\mathbf{n})^2 + K_{22}(\mathbf{n}\cdot\nabla\times\mathbf{n} + q_0)^2 + K_{33}(\mathbf{n}\times\nabla\times\mathbf{n})^2],$$
$$F_2 = -\frac{\chi_a}{2}(\mathbf{n}\cdot\mathbf{H})^2, \quad F_3 = -M_s f\,\mathbf{n}\cdot\mathbf{H}.$$

Here K_{ii} are elastic constants, q_0 is the wave number of an unperturbed helical structure of a CLC (further we assume that $q_0 > 0$), $\chi_a > 0$ is the anisotropy of the diamagnetic susceptibility, M_s is the saturation magnetization of the material of impurity particles, v_p is the volume of a magnetic particle, and f is the volume fraction of the magnetic particles.

In Eq. (2) we imply summation over repeated tensor indices, and variation of the free energy F is performed under the constraint $\mathbf{n}^2 = 1$.

The term F_1 in the free energy density (3) corresponds to the energy of the orientational-elastic deformations of the director field, the contribution F_2 is the energy density of the magnetic field $\mathbf{H}$ interaction with the CLC, and F_3 is the energy density of the interaction of magnetic particles with the magnetic field. We neglect magnetic dipole-dipole interactions, because the concentration of magnetic particles is assumed to be small ($f \ll 1$).

Taking into account the expression (1) for the director field, the density of free energy (3) can be written as follows

$$F = \frac{K_{22}}{2}\left(\frac{d\varphi}{dz} - q_0\right)^2 - \frac{\chi_a H^2}{2}\cos^2(\varphi-\omega t) - M_s f H\cos(\varphi-\omega t). \tag{4}$$

In this case the equation of the director motion (2) with the help of Eq. (1) leads to the following equation for the angle φ:

$$K_{22}\frac{\partial^2\varphi}{\partial z^2} - \frac{\chi_a H^2}{2}\sin 2(\varphi-\omega t) - M_s f H\sin(\varphi-\omega t) = \gamma_1\frac{\partial\varphi}{\partial t}. \tag{5}$$

We choose the value q_0^{-1} as the unit of length and define the dimensionless coordinate $\zeta = q_0 z$, and also the dimensionless time $\tau = \omega t$. As the unit of the field, we choose the value $H_0 = q_0\sqrt{K_{22}/\chi_a}$ and determine the dimensionless field strength $h = H/H_0$. In a pure CLC, i.e., at $f = 0$, the cholesteric–nematic transition field is equal to $\pi H_0/2$ [4]. The value of H_0 can be obtained from the balance of the contributions F_1 and F_2 in the free energy (3). Thus, H_0 determines in order of magnitude the FC–FN transition field if the unwinding of the FC helical structure is ensured by the quadrupole (diamagnetic) mechanism F_2 [23]. Along with H_0, another characteristic field $H_d = K_{22}q_0^2/(M_s f)$ can be determined for FC from the balance of contributions F_1 and F_3 in free energy (3). In the case of the dipole (ferromagnetic) mechanism domination F_3, the FC–FN transition field will have the order of H_d [23].

We introduce the following dimensionless quantities: