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MAGNETIC FIELD EFFECTS IN A NEMATIC CELL WITH A HIGH TILT ANGLE ("FIRST-ORDER THEORY").

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Abstract Within the framework of so-called "first-order elastic theory" we consider distortions in a nematic cell with tilted molecular orientation subjected to a tilted magnetic field. The geometry has been suggested by Faetti to test the validity of different approaches to the problem of the divergence K_{13} elastic term. According to the first-order elastic theory, in which only the first and second derivatives in the elastic free energy are retained, the K_{13} term causes deformations at the cell surface when a magnetic field is applied along the initial director **n**. We discuss conditions that should be satisfied in order to observe the instability.

INTRODUCTION

The leading Frank-Oseen part of the nematic elastic free energy, written in its modern form by Nehring and Saupe, [1] is quadratic in derivatives of the director **n**,

$$F_2 = \int dV \Big\{ \tfrac{1}{2}K_{11}(\nabla\cdot\mathbf{n})^2 + \tfrac{1}{2}K_{22}(\mathbf{n}\cdot\nabla\times\mathbf{n})^2 + \tfrac{1}{2}K_{33}(\mathbf{n}\times\nabla\times\mathbf{n})^2 \\ + K_{13}\nabla\big[\mathbf{n}(\nabla\cdot\mathbf{n})\big] - K_{24}\nabla\big[\mathbf{n}(\nabla\cdot\mathbf{n}) + \mathbf{n}\times\nabla\times\mathbf{n}\big]\Big\}. \tag{1}$$

The elastic free energy functional F_2 contains three standard contributions with the elastic constants K_{11} (splay), K_{22} (twist) and K_{33} (bend), as well as two divergence

K_{13} and K_{24} terms. It is the divergence term $K_{13}\nabla[\mathbf{n}(\nabla\cdot\mathbf{n})]$ that remains a controversial issue despite decades of discussions.[1-11] The problem is that microscopic theories predict finite K_{13};[6,12-14] however, when $K_{13}\neq 0$, F_2 has no minimum. To deal with a bounded free energy functional, higher order terms in the free energy expansion should be added to F_2. The approaches suggested so far differ in the number of terms added to F_2. The theories that restrict the expansion to a few higher order terms predict strong (molecular-scale) surface deformations.[6,7,9] In the infinite-order theory, the deformations are supposed to be bounded to the standard weak magnitude by all higher order terms.[8] Since there are no strong deformations, the family of the director distributions in this approach can be assumed to satisfy the Euler-Lagrange equations for the functional F_2 alone.[4,8] The theoretical approach in which only F_2 functional is considered, is often called [10] the "first-order elastic theory" (to avoid confusion, note that F_2 contains squares of the first derivatives and the second-order derivatives of director). The equivalence of the infinite-order theory and the first-order theory is not proven yet.

The experimental situation with K_{13} is also murky: different authors have used different approaches in the interpretation of data.[15-21] Moreover, no physical effect caused solely by K_{13} has been reported. To resolve the K_{13} puzzle, one has to find such an effect. A possible experimental test that allows one at least to discriminate the predictions of the first-order approach was suggested by Faetti.[10] The geometry of the problem implies a nematic cell with tilted boundary conditions. The cell is subjected to a magnetic field $\mathbf{B}$ directed along the initial $\mathbf{n}$. There are two different predictions as to what happens in a finite field. The model with molecular-scale subsurface deformations predicts that these molecular-scale deformations of $\mathbf{n}$ exist when $\mathbf{B}$; when $\mathbf{B}\neq 0$, no additional deformations occur. In contrast, the first-order theory which uses exclusively the functional F_2 predicts that the increase of the field results in long-range deformations with maximum at the bounding surface. These deformations occur only when $K_{13}\neq 0$ and only when the initial director orientation is tilted.[10]

In this article we explore the possibility of the experimental detection of the K_{13}-induced deformations in a tilted nematic cell within the frame of the first-order theory. Note that the predictions of this theory may not coincide with the theory in which all the high-order terms are taken into account; the complete analysis would require a resummation of these high-order terms. The one-dimensional geometry of the problem makes another divergence K_{24} term identically zero. We set $K_{11}\neq K_{33}$ (K_{11} and K_{33} were taken to be equal in the Faetti's article[10]), and analyze the optical phase retardation of the cell as a function of the magnetic field, K_{13} value and anchoring strength. We also analyze the role of the possible cell hybridity.

UNIFORM CELL

Consider a nematic cell with plates located at $z_1 = -d/2$ and $z_2 = d/2$. The director is confined to the (x,z) plane of Cartesian coordinates and makes an angle θ with the normal to the cell plates $\mathbf{k}$. The magnetic field is applied in the (x,z) plane at some angle α with respect to $\mathbf{k}$, Fig.1.

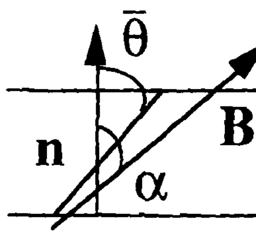

FIGURE 1 Cell geometry

If one restricts the consideration by the Frank-Oseen functional without higher-order (fourth, etc.) terms, the free energy per unit area of the cell is

$$F_2 = \tfrac{1}{2}\int_{-d/2}^{d/2} dz\left[\left(K_{11}\sin^2\theta + K_{33}\cos^2\theta\right)(\theta')^2 + \mu_0^{-1}\chi_a B^2 \sin^2(\theta-\alpha)\right]$$
$$+\tfrac{1}{2}K_{13}\left(\theta_1' \sin 2\theta_1 - \theta_2' \sin 2\theta_2\right) + \tfrac{1}{2}W_1\left(\theta_1 - \overline{\theta_1}\right)^2 + \tfrac{1}{2}W_2\left(\theta_2 - \overline{\theta_2}\right)^2, \tag{2}$$

where $\theta = \theta(z)$ describes director distortions, $\theta' = d\theta/dz$, subscripts 1 and 2 refer to the lower and upper plates, respectively. We consider first the case when the cell is uniform, i.e. the directions of the "easy axis" and the anchoring coefficients at the two plates coincide, $\overline{\theta_1} = \overline{\theta_2} = \overline{\theta}$ and $W_1 = W_2 = W$.

If the field direction is close to the easy axis, $\overline{\theta} \approx \alpha$, the director deformations are weak, which justifies the form of the anchoring terms in Eq.(2). It is convenient to introduce small angles $\overline{\psi} = \overline{\theta} - \alpha$ and $\psi(z) = \theta(z) - \alpha << 1$ and notations $K = K_{11}\sin^2\alpha + K_{33}\cos^2\alpha$, $K_\alpha = K - 2K_{13}\cos 2\alpha$, $q^2 = \chi_a B^2 / K\mu_0$, $u = qd/2$, $l = K/W$.

If one follows the first-order theory, the equilibrium $\psi(z)$ should be found from the Euler-Lagrange equation for the functional (2) alone. We do not discuss here the possible role of the higher-order terms that might completely change the whole analysis.

For small $\psi(z)$, the solution can be found from the leading part of the Euler-Lagrange equation for the functional (2),

$$\psi'' - q^2\psi = 0, \tag{3}$$

as

$$\psi = A\sinh qz + N\cosh qz, \tag{4}$$

where A and N are, respectively, the amplitudes of the modes antisymmetric and symmetric about the middle plane $z = 0$. Substituting (4) into (2) and retaining terms up to the second order in small A and N yield

$$\begin{aligned} F = &\left(\tfrac{1}{2}qK_\alpha \sinh 2u + W\sinh^2 u\right)A^2 + \left(\tfrac{1}{2}qK_\alpha \sinh 2u + W\cosh^2 u\right)N^2 \\ &+ \left[2W\left(\alpha - \overline{\theta}\right)\cosh u - qK_{13}\sin 2\alpha \sinh u\right]N + W\left(\alpha - \overline{\theta}\right)^2. \end{aligned} \tag{5}$$

Minimization of $F(A,N)$ with respect to A and N results in the equilibrium $\psi(z)$. If $|K_{13}| < K_{11}/2, K_{33}/2$, then $A = 0$.[22] At the same time, the amplitude N of the symmetric mode is finite for any non-zero $q \sim B$. Namely, the surface value of the tilt $\psi_s = \psi_1 = \psi_2 = N\cosh u$ is

$$\psi_s = \frac{\dfrac{K_{13}}{2K_\alpha}\sin 2\alpha - \left(\alpha - \overline{\theta}\right)(ql)^{-1}\coth(qd/2)}{1 + (ql)^{-1}\coth(qd/2)}. \tag{6}$$

In the limit $q = 0$ the director is not distorted, $\psi_s = \overline{\theta} - \alpha$. However, for $q \neq 0$ and any $0 < \overline{\theta} < \pi/2$, the distortions $\propto K_{13}\sin 2\overline{\theta}$ appear (even for **B** parallel to **n**):

$$\psi_{s,\parallel} = \frac{K_{13}}{2K_\alpha}\frac{\sin 2\alpha}{1 + (ql)^{-1}\coth(qd/2)}. \tag{7}$$

Evidently, the optimal pretilt for the detection of the effect is $\overline{\theta} = 45^o$.

The deviations of **n** can be detected by measuring the phase retardation Φ for light transmitted through the cell. For a normally incident (along z-axis) laser beam of the wavelength λ,

$$\Phi = \frac{2\pi n_o}{\lambda} \int_{-d/2}^{d/2} \left(\frac{n_e}{n} - 1 \right) dz, \tag{8}$$

where

$$n[\theta(z)] = \left[n_o^2 \sin^2 \theta(z) + n_e^2 \cos^2 \theta(z) \right]^{1/2}; \tag{9}$$

n_o and n_e are the ordinary and extraordinary refractive indices, respectively. The change $\Delta\Phi$ in the phase retardation, $\Delta\Phi = \Phi|_B - \Phi|_{B=0}$,

$$\Delta\Phi = \frac{2\pi n_o n_e}{\lambda} \int_{-d/2}^{d/2} \left\{ \frac{1}{n[\theta(z,B)]} - \frac{1}{n(\overline{\theta})} \right\} dz \tag{10}$$

is calculated using Eq. (6). Equation (10) can be used directly to fit numerically the experimental dependencies $\Delta\Phi(B)$ at different α. Note that the transmittive technique is sensitive to the symmetric modes associated with the possible K_{13}-effect. Small imperfections of the cell such as hybridity $\theta_1 \neq \theta_2$ or twist (difference in the azimuthal angles at the two plates, $\varphi_1 \neq \varphi_2$) do not shadow the pure K_{13}-effect. Indeed, the K_{13}-distortions are symmetric, while hybridity and twist correspond to antisymmetric modes. Therefore, even if $\psi_s \sim \delta\theta, \delta\varphi$, the "imperfection" antisymmetric modes contribute only to the order $(\delta\theta)^2, (\delta\varphi)^2$, since the integral Φ above vanishes for an antisymmetric integrand. We consider the effect of hybridity in a greater detail in the next section.

To clarify the effect of K_{13} on $\Delta\Phi$, it is convenient to expand $\Delta\Phi$, retaining terms linear in ψ_s:

$$\Delta\Phi = \pi n_o n_e n_\alpha^{-3} \left(n_e^2 - n_o^2 \right) (d/\lambda) \sin 2\alpha \left[2\psi_s (qd)^{-1} \tanh(qd/2) - \left(\overline{\theta} - \alpha \right) \right], \tag{11}$$

where $n_\alpha = n(\alpha) = \left[n_o^2 \sin^2 \alpha + n_e^2 \cos^2 \alpha \right]^{1/2}$. For $K_{13} = 0$, $\Delta\Phi(B)$ is a monotonous function: as B grows, $\Delta\Phi$ increases for $\alpha > \overline{\theta}$, decreases for $\alpha < \overline{\theta}$ and remains zero when $\alpha = \overline{\theta}$ (Fig.2). This behavior drastically changes when $K_{13} \neq 0$: $\Delta\Phi(B)$ is non-

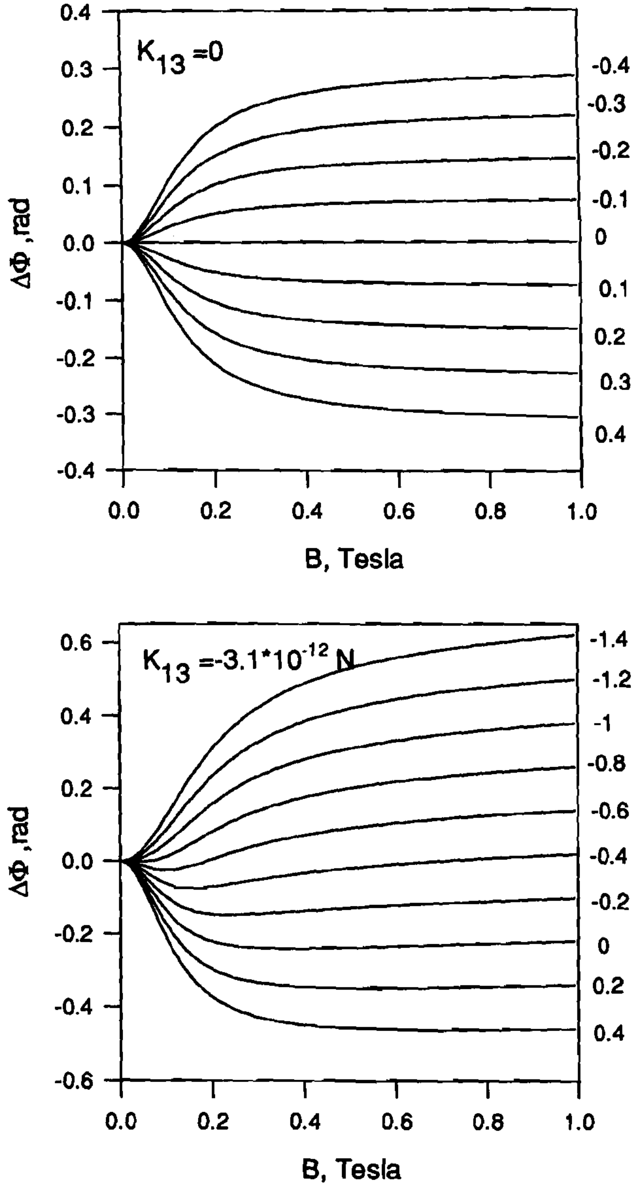

FIGURE 2 Phase retardation as a function of the magnetic field and the angle $\overline{\psi} = \overline{\theta} - \alpha$ (shown in degrees) for $K_{13} = 0$ (above) and $K_{13} = -3.1 \times 10^{-12}$ N (below); other parameters are as follows: $n_o = 1.4832$, $n_e = 1.5869$, $\lambda = 633$ nm; $d = 96 \mu$m, $\chi_a = 4\pi \times 0.61 \times 10^{-7}$, $K_{11} = 13.7 \times 10^{-12}$N, $K_{33} = 18.3 \times 10^{-12}$N, $\overline{\theta} = 80.4°$, and $W = 1.4 \times 10^{-6}$ J / m^2.

monotonous with a minimum for $K_{13} < 0$ (Fig. 2) and a maximum for $K_{13} > 0$. Only for $B \to \infty$ the phase retardation curves asymptotically approach those calculated for $K_{13} = 0$. The non-monotonous behavior is especially pronounced for $\alpha \approx \overline{\theta}$.

The expected dependencies $\Delta\Phi(B)$ are shown in Fig.2 for the following parameters: $n_o = 1.4832$ and $n_e = 1.5869$ at $\lambda = 633$ nm; $\chi_a = 4\pi \times 0.61 \times 10^{-7}$, $K_{11} = 13.7 \times 10^{-12}$N, $K_{33} = 18.3 \times 10^{-12}$N (which are close to the material ZLI 4108-100) and $d = 96\mu$m, $\overline{\theta} = 80.4^o$, $W = 1.4 \times 10^{-6}$ J / m^2. The amplitude of phase retardation non-monotonity is about 0.1 rad at $B = 0.1$ Tesla and should be easily detected. Note that with the parameters given above the optical retardation changes are one order of magnitude stronger than those potentially caused by suppression of the nematic fluctuations in the magnetic field: As measured by Poggi and Filippini,[23] the phase retardation change is about 0.01 rad at $B = 0.1$ Tesla for a 150μm thick cell filled with the nematic material 7CB.

THE ROLE OF THE ANCHORING COEFFICIENT.

An important point is that the K_{13}-induced deformations can be detected only if the anchoring strength is sufficiently small. Figure 3 illustrates the role of the anchoring coefficient W in the behavior of the phase retardation, calculated from Eq. (11): A smaller W enhances the non-monotonity of $\Delta\Phi(B)$. If W is larger than $W = 10^{-5}$ J / m^2, the effect becomes virtually undetectable (the amplitude of the non-monotonity is less than 10^{-2} rad and effects such as suppression of the nematic fluctuations in the magnetic field should be taken into account).

Note that in the problem under consideration one deals with a tilted equilibrium orientation of the director and determination of the anchoring coefficient is a challenging problem itself. For example, a simple and reliable method recently suggested by Gu, Uram and Rosenblatt [12], which is based on the dielectric Fréedericksz transition in the wedged cell, can be applied only to a strictly homeotropic or planar cells. When director is tilted with respect to the electric field, the Fréedericksz transition has no threshold.

In principle, W in the tilted cell can be defined from the fitting procedure for the phase retardation (10), together with K_{13}. The measured W value can be checked for reliability by using the fact that the K_{13}-effect, if it exists, should show up only in a narrow angular region $|\alpha - \overline{\theta}| \le 1^o$. For $|\alpha - \overline{\theta}| > 1^o$ the function $\Delta\Phi(B)$ becomes monotonous since the usual (diamagnetic) director reorientation along the field becomes much stronger than the K_{13}-effect. The last circumstance allows one to estimate W from the region, say, $2^0 \le |\alpha - \overline{\theta}| \le 6^o$ where K_{13} has practically no influence. Another way to check the reliability of the measured W is to compare two independent W

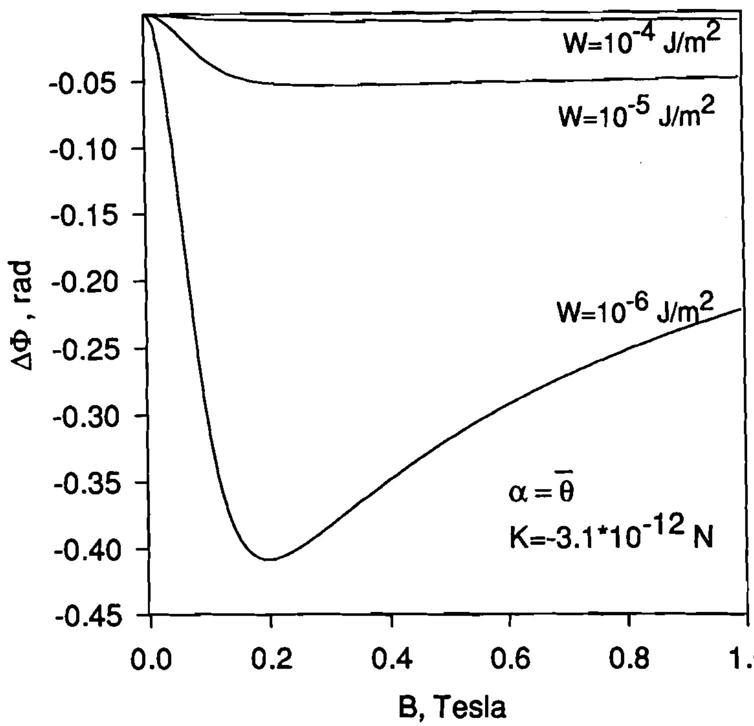

FIGURE 3 Nonmonotonity in phase retardation at $\alpha = \overline{\theta}$ fades as the anchoring strength W increases; other parameters are the same as in Fig.2..

values, calculated from the dependence $\Delta\Phi(\alpha)$ when B=const and from the dependence $\Delta\Phi(B)$ when α=const.[26] The analytical expression for $\Delta\Phi(B, W)$ can be obtained by expanding $\Delta\Phi$ in Eq.(10) in terms of the small angle $\overline{\psi}$, retaining now both the linear and quadratic terms, and neglecting K_{13} contribution:

$$\Delta\Phi = \frac{4\pi a}{\lambda g q}\left[b\overline{\psi}(\tanh u - ug) + c\overline{\psi}^2\left(\frac{2u + \sinh 2u}{4g\cosh^2 u} - ug\right)\right], \tag{12}$$

where $g = 1 + lq\tanh u$ and the constants a, b and c are defined by the indices of refraction and the direction of the magnetic field α:

$$a = \frac{n_o n_e\left(n_e^2 - n_o^2\right)}{2n_\alpha^5},\ b = n_\alpha^2 \sin 2\alpha,\ c = n_e^2\cos^2\alpha - n_o^2\sin^2\alpha + \left(n_e^2 - n_o^2\right)\cos^2\alpha\sin^2\alpha.$$

One can alternatively use a slightly different expression for $\Delta\Phi$ derived in Ref. [26] with a, b and c defined through $\overline{\theta}$; however, α can be defined more accurately than $\overline{\theta}$.

Another concern about the possible detection of the K_{13}-induced distortions in the light-transmission mode is the hybridity of the cells. Below we consider the effect and show that small hybridity $\sim 1^0$ does not affect the observed phenomena for the parameters chosen above. However, when the hybridity becomes of the order of $\sim 10^0$, it can mimic the K_{13} effect. Galatola and Ziherl [24] were the first to find this mimic effect in numerical calculations of the phase ratardation.

HYBRID CELL.

The free energy per unit area of the hybrid cell is given by Eq.(2) where the surface angular and anchoring parameters are generally different. To simplify the analysis, we put K_{13}=0 and consider how the hybridity influences the phase retardation and the measurements of the anchoring coefficient.

If the hybridity is small, $|\overline{\theta}_1 - \overline{\theta}_2|, |\psi| << \overline{\theta}_1, \overline{\theta}_2, \alpha$, one can still employ the solution of type (4) of the equation (3). Substituting (4) into Eq. (2) and retaining terms up to the second order in small A and N yield

$$\begin{aligned} F = {} & \tfrac{1}{2}\left[qK\sinh 2u + (W_1 + W_2)\sinh^2 u\right]A^2 + \tfrac{1}{2}\left[qK\sinh 2u + (W_1 + W_2)\cosh^2 u\right]N^2 \\ & + \tfrac{1}{2}AN\Delta W\sinh u - A\Delta J\sinh u + NJ\sinh u + \tfrac{1}{2}W_1\overline{\psi}_1^2 + \tfrac{1}{2}W_2\overline{\psi}_2^2, \end{aligned} \tag{13}$$

where $\Delta W = W_2 - W_1$, $J = W_1\overline{\psi}_1 + W_2\overline{\psi}_2$, $\Delta J = -W_1\overline{\psi}_1 + W_2\overline{\psi}_2$. Minimization of $F(A, N)$ with respect to A and N results in the equilibrium $\psi(z)$ with amplitudes

$$A = \frac{2r\Delta J\sinh u - J\Delta W\sinh u\cosh^2 u}{4rs - (\Delta W)^2\sinh^2 u\cosh^2 u},\ N = \frac{2sJ\cosh u - \Delta J\Delta W\sinh^2 u\cosh u}{4rs - (\Delta W)^2\sinh^2 u\cosh^2 u}. \tag{14}$$

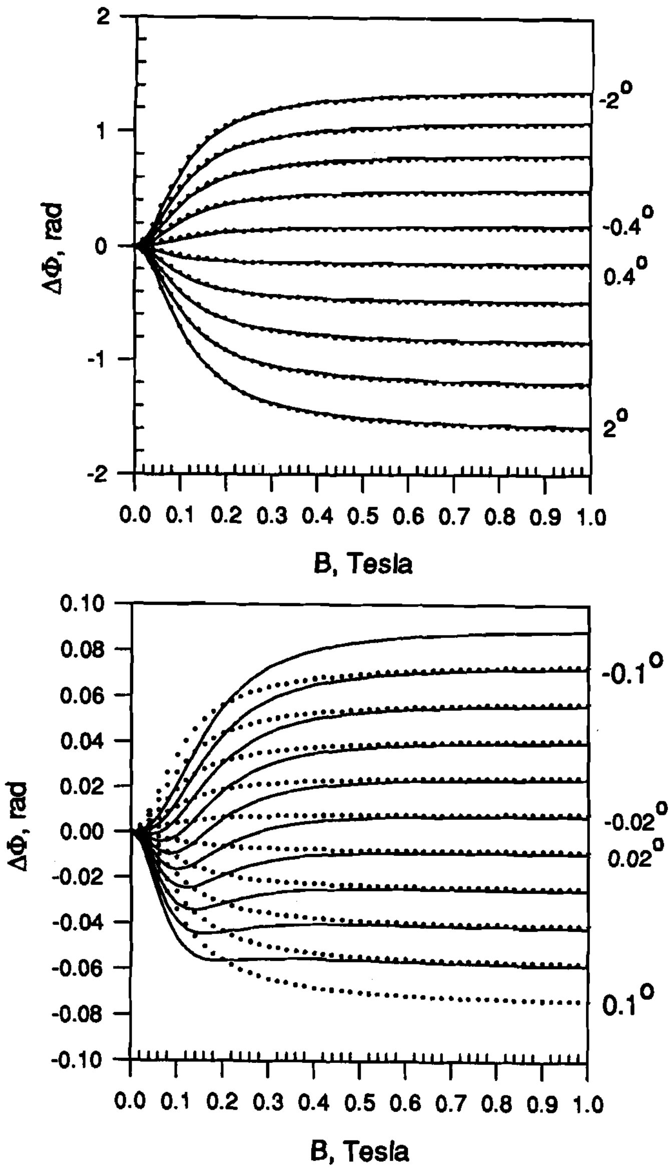

FIGURE 4 Phase retardation as a function of the magnetic field and the angle $\overline{\psi} = \overline{\theta} - \alpha$ (in degrees) for a hybrid cell with $\overline{\theta}_1 = 14^o$, $\overline{\theta}_2 = 6^o$ (lines) and for a uniform cell with $\overline{\theta} = 10.15^o$ (dots); two different scales are shown.
$K_{13} = 0$, $n_o = 1.53$, $n_e = 1.708$, $\lambda = 633$nm; $d = 60\,\mu$m, $K_{11} = 6.2\times10^{-12}$N, $K_{33} = 8.2\times10^{-12}$N, $\chi_a = 4\pi\times1.13\times10^{-7}$, $W = 10^{-6}$ J / m^2.

Here $r = \frac{1}{2}qK\sinh 2u + (W_1 + W_2)\cosh^2 u$, $s = \frac{1}{2}qK\sinh 2u + (W_1 + W_2)\sinh^2 u$. Note that the amplitude A is defined primarily by the difference in the tilt angles while N is defined by an average tilt angle (as easy to verify by assuming $W_1 = W_2$).

The change in the phase retardation of the hybrid cell caused by the magnetic field can be found as an expansion in terms of small A and N:

$$\begin{aligned}\Delta\Phi &= \pi n_o n_e n_\alpha^{-3}\left(n_e^2 - n_o^2\right)(d/2u\lambda) \\ &\times\left\{2N\sinh u\sin 2\alpha + \left[\cos 2\alpha + \frac{3\sin^2 2\alpha\left(n_e^2 - n_o^2\right)}{4n_\alpha^2}\right]\right. \\ &\times\left.\left[\frac{\sinh 2u}{2}\left(A^2 + N^2\right) + u\left(N^2 - A^2\right)\right]\right\}.\end{aligned} \tag{15}$$

Note that the antisymmetric mode contributes only to the order $\left(\delta\overline{\theta}\right)^2$. The leading term in the last equation is the term linear in N which is defined by the average tilt in the cell rather than by the difference in the tilt at the two plates. If, for example, $\overline{\theta_1} < \alpha < \overline{\theta_2}$, the increase in the magnetic field would increase the polar angle at the lower plate and decrease it at the upper plate; the average value would change less than the difference in the polar angles. This is why a small hybridity does not cause substantial non-monotonity of $\Delta\Phi(B)$. For example, numerical calculations with $\delta\overline{\theta} = 2^\circ$ ($\overline{\theta_1} = 10^\circ$ and $\overline{\theta_2} = 12^\circ$), $d = 80\mu$, and material parameters given above show that the amplitude of $\Delta\Phi(B)$ non-monotonity is less than 0.01 rad and this non-monotonity appears only in a narrow region of about 0.2°. Figure 4 gives another example for a material with parameters close to that of pentylcyanobiphenyl (5CB).

CONCLUSION

We analyzed the behavior of the nematic cell with tilted boundary conditions under the action of the magnetic field. The formal use of the first-order theory leads to the conclusion that initial uniform director orientation becomes unstable when the magnetic field is applied along the director. We do not consider the possible role of the higher-order terms; note that a rigorous analysis would require at least a resummation of all higher -order terms in the expansion of the nematic free elastic energy. According to the analysis performed within the framework of the first order theory, if the K_{13}-instability exists, it shows up as a symmetry-breaking spontaneous deformations. The deformations can be detected in the measurements of the optical phase retardation $\Delta\Phi$ for light transmitted through the cell; the distinctive feature of the K_{13}-instability is a non-monotonous behavior of $\Delta\Phi$ when the applied field increases. If the field is directed along the initial director, the K_{13}-driven surface deviation occur at low field and decrease $\Delta\Phi$ when K_{13} is negative (or increase $\Delta\Phi$ when K_{13} is positive); at higher fields, the diamagnetic reorientation of the molecules in the bulk tends to restore the initial value of $\Delta\Phi$. Provided the cell is uniformly aligned, the amplitude of non-monotonous changes in $\Delta\Phi$ can reach 0.1 rad for $(50-100)\,\mu$m thick cell if K_{13} is of the order of $(0.1-1)\,K$.

The instability can be hindered by surface anchoring: for example, the characteristic non-monotonity in $\Delta\Phi(B)$ becomes practically undetectable when the anchoring coefficient is higher than $10^{-5}\,\mathrm{J/m^2}$. The second problem is the requirement of a high tilt angle $\overline{\theta}$ (optimally 45^o), since the amplitude of the K_{13}-instability is proportional to $\sin 2\overline{\theta}$. Note also that the substrate should be also smooth enough to maintain a constant value of the actual surface angle $\overline{\theta}$. All these anchoring requirements are rather hard to satisfy simultaneously: the alignment methods that give a high pretilt (e.g., oblique evaporation of SiO) are known to provide strong anchoring, $W > 10^{-5}\,\mathrm{J/m^2}$, and also result in a rough profile of substrates.

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