# $K_{13}$-Induced Deformations in a Nematic Liquid Crystal: Experimental Test of the First-Order Theory

## V. M. PERGAMENSHCHIK[a,c], D. SUBACIUS[a] and O. D. LAVRENTOVICH[a,b],*

*aLiquid Crystal Institute and bChemical Physics Program,
Kent State University, Kent, Ohio 44242;
cInstitute of Physics, National Academy of Sciences, Kyiv, Ukraine*

We consider predictions of the so-called "first-order elastic theory" with non-zero elastic constant $K_{13}$ for a nematic cell with tilted molecular orientation. The geometry has been suggested by Faetti to test the validity of different theoretical 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. These deformations can be observed in the measurements of the optical phase retardation; the distinctive feature of the $K_{13}$ effect is non-monotonous behavior of the phase retardation when the magnetic field increases. Despite a number of difficulties, the effect can be tested experimentally. We report experimental data on the behavior of the nematic cell with a high pretilt angle and weak anchoring in a tilted magnetic field. These results show no significant deviation in the monotonous behavior of the phase retardation; therefore, the experiment does not confirm the predictions of the first-order theory when $K_{13} \neq 0$.

*Keywords:* Nematic liquid crystals; elastic constants; splay-bend elasticity; magneto-optics; $K_{13}$; experimental test

## INTRODUCTION

The fundamental formula of the elastic theory of liquid crystals, so-called Frank-Oseen elastic energy functional, has been written in its modern form by Nehring and Saupe [1] more than twenty years ago, but still remains a subject

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*Corresponding author.
Phone: (330) 672-4844; Fax: (330) 672-2796; E-mail: Olavrent@kentvm.kent.edu.

of discussions. The functional is quadratic in derivatives of the director **n**,

$$F_2 = \int dV \left\{ \frac{1}{2} K_{11} (\nabla \cdot \mathbf{n})^2 + \frac{1}{2} K_{22} (\mathbf{n} \cdot \nabla \times \mathbf{n})^2 + \frac{1}{2} K_{33} (\mathbf{n} \times \nabla \times \mathbf{n})^2 \right.$$

$$\left. + K_{13} \nabla [\mathbf{n}(\nabla \cdot \mathbf{n})] - K_{24} \nabla [\mathbf{n}(\nabla \cdot \mathbf{n}) + \mathbf{n} \times \nabla \times \mathbf{n}] \right\}. \quad (1)$$

and contains three standard contributions with the elastic constants $K_{11}, K_{22}$, and $K_{33}$, 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 [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. Note that $F_2$ is nothing else but the leading part of the expansion of the true nematic elastic energy in terms of director derivatives; $F_2$ contains squares of the first derivatives and the second-order derivatives of **n**. A natural idea to bound the free energy functional is to add higher order terms in the free energy expansion [6–9]. 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]. Obviously, the director distribution cannot be found analytically from the infinite-order functional. However, one might assume, since there are no strong deformations, that the family of the director distributions satisfies 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". Note that the equivalence of the infinite-order theory and the first-order theory is not proven.

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 test implies a nematic cell with tilted boundary conditions subjected to a magnetic field **B** directed along **n**. There are two different predictions as to what happens in a finite field.

The model with molecular-scale deformations (in which the $K_{13}$-term is counterbalanced by one positive-definite fourth-order term) predicts that these strong molecular-scale distortions at the boundaries represent the

ground state of the system when $\mathbf{B} = 0$. Since the deformations are strong, non-zero $\mathbf{B}$ of a reasonable strength changes nothing and no experimentally observed features are expected, Figure 1a, b.

The first-order theory which uses exclusively the functional $F_2$ and the Euler-Lagrange equation based on $F_2$, predicts that the initial state is strictly uniform (Fig. 1c); there are no spontaneous deformations when $\mathbf{B} = 0$. The increase of the field results in long-range deformations with maximum at the bounding surface (Fig. 1d). 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 possiblity 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 by rigorous resummation; it might happen that in some geometries a naive approach based on the first-order theory would lead to qualitatively correct results while in other situations it would not. We analyze the predictions of the first-order theory and conditions under which the experimental detection of the effect predicted by this theory is possible. Experimental results show no significant manifestation of the effect predicted by the first-order theory.
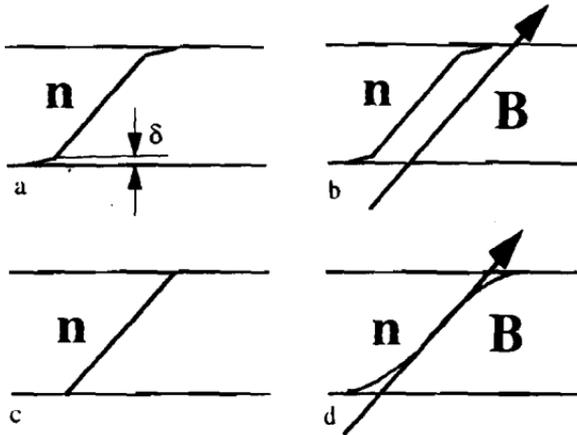


FIGURE 1   A nematic cell with tilted boundary conditions in zero (a,c) and non-zero (b,d) magnetic field applied parallel to the easy axis. Director distribution is shown schematically as predicted by the theory with strong molecular deformations (a,b) and by the first-order theory (c,d). The scale of deformation $\delta$ in (a) is molecular; the scale of deformations in (d) is macroscopic.

## FIRST-ORDER THEORY

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 $\theta$ with the normal $\mathbf{k}$ to the cell plates. The magnetic field is applied in the $(x,z)$ plane at some angle $\alpha$ with respect to $\mathbf{k}$. The one-dimensional geometry makes the twist and divergence $K_{24}$ terms identically zero. In the first-order theory, the free energy per unit area of the cell is

$$F_2 = \frac{1}{2} \int_{-d/2}^{d/2} dz \left[ (K_{11}\sin^2\theta + K_{33}\cos^2\theta)(\theta')^2 + \mu_0^{-1}\chi_a B^2 \sin^2(\theta - \alpha) \right]$$

$$+ \frac{1}{2}K_{13}(\theta'_1\sin 2\theta_1 - \theta'_2\sin 2\theta_2) + \frac{1}{2}W_1(\theta_1 - \bar{\theta}_1)^2 + \frac{1}{2}W_2(\theta_2 - \bar{\theta}_2)^2, \quad (2)$$

where $\mu_0$ is the magnetic constant, $\chi_a$ is the anisotropy of diamagnetic susceptibility, $W$ is the anchoring coefficient, $\theta = \theta(z)$ describes director distortions, $\theta' = d\theta/dz$, finally, the subscripts 1 and 2 refer to the lower and upper plates, respectively. We consider a uniform cell, in which the directions of the "easy axis" and the anchoring coefficients at the two plates coincide, $\bar{\theta}_1 = \bar{\theta}_2 = \bar{\theta}$ and $W_1 = W_2 = W$.

If the field direction is close to the easy axis, $\bar{\theta} \approx \alpha$, the deformations are weak, which justifies the form of the anchoring terms in Eq.(2). It is convenient to introduce small angles $\bar{\psi} = \bar{\theta} - \alpha$ 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 is found from the leading part of the Euler-Lagrange equation for the functional (2), $\psi'' - q^2\psi = 0$, as

$$\psi = A\sinh qz + N\cosh qz, \quad (3)$$

where $A$ and $N$ are, respectively, the amplitudes of the modes antisymmetric and symmetric about the middle plane $z = 0$. Substituting (3) into (2) and

retaining terms up to the second order in small $A$ and $N$ yield

$$F = \left(\frac{1}{2}qK_\alpha\sinh 2u + W\sinh^2 u\right)A^2 + \left(\frac{1}{2}qK_\alpha\sinh 2u + W\cosh^2 u\right)N^2$$

$$+ [2W(\alpha - \bar{\theta})\cosh u - qK_{13}\sin 2\alpha\sinh u]\dot{N} + W(\alpha - \bar{\theta})^2. \qquad (4)$$

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

$$\psi_s = \frac{\dfrac{K_{13}}{2K_\alpha}\sin 2\alpha - (\alpha - \bar{\theta})(ql)^{-1}\coth(qd/2)}{1 + (ql)^{-1}\coth(qd/2)}. \qquad (5)$$

In the limit $q = 0$ the director is not distorted, $\psi_s = \bar{\theta} - \alpha$. However, for $q \neq 0$ and any $0 < \bar{\theta} < \pi/2$, the distortions $\propto K_{13}\sin 2\bar{\theta}$ appear (even for **B** parallel to **n**). Evidently, the optimal pretilt for the detection of the effect is $\bar{\theta} = 45^0$.

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

$$\Phi = \frac{2\pi n_0}{\lambda}\int_{-d/2}^{d/2}\left(\frac{n_e}{n} - 1\right)dz, \qquad (6)$$

where $n[\theta(z)] = [n_0^2\sin^2\theta(z) + n_e^2\cos^2\theta(z)]^{1/2}$; $n_0$ and $n_e$ are the ordinary and extraordinary refractive indices, respectively. The field-induced change of the phase retardation, $\Delta\Phi = \Phi|_B - \Phi|_{B=0}$, is

$$\Delta\Phi = \frac{2\pi n_0 n_e}{\lambda}\int_{-s/2}^{d/2}\left\{\frac{1}{n[\theta(z,B)]} - \frac{1}{n(\bar{\theta})}\right\}dz. \qquad (7)$$

The expected dependencies $\Delta\Phi(B)$ are shown in Figure 2 for the following parameters (taken for the typical nematic material 5CB): $n_o = 1.530$ and $n_e = 1.708$ at $\lambda = 633$ nm [23]; $\chi_a = 113.5 \times 10^{-12}\text{kg}^{-1}\text{m}^3$, $K_{11} = 6.2 \times 10^{-12}$ N, $K_{33} = 8.2 \times 10^{-12}$ N[24] and $d = 68.0\,\mu\text{m}$, $\bar{\theta} = 73.3^0$, $W = 4.5 \times 10^{-6}$ J/m$^2$. For $K_{13} = 0$, $\Delta\Phi(B)$ is a monotonous function: as $B$ grows, $\Delta\Phi$ increases for $\alpha > \bar{\theta}$, decreases for $\alpha < \bar{\theta}$ and remains zero when $\alpha = \bar{\theta}$. This

behavior drastically changes when $K_{13} \neq 0$: $\Delta\Phi(B)$ is non-monotonous with a minimum for $K_{13} < 0$ and a maximum for $K_{13} > 0$. Only for $B \rightarrow \infty$ the phase retardation curves asymptotically approach those calculated for $K_{13} = 0$. The non-monotonous behaviour is especially pronounced for $\alpha \approx \bar{\theta}$. The amplitude of non-monotonity is about 0.1 rad at $B = 0.1$ Tesla. Note that with the parameters given above the changes in $\Delta\Phi$ 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 [25] the phase retardation change in about 0.01 rad at $B = 0.1$ Tesla for a 150μm thick cell filled with the nematic 7CB.

An important point is that the $K_{13}$-induced deformations can be detected only if the anchoring strength is sufficiently small. If $W$ is larger than $W = 10^{-5}$ J/m$^2$, the amplitude of the non-monotonity becomes less than $10^{-2}$
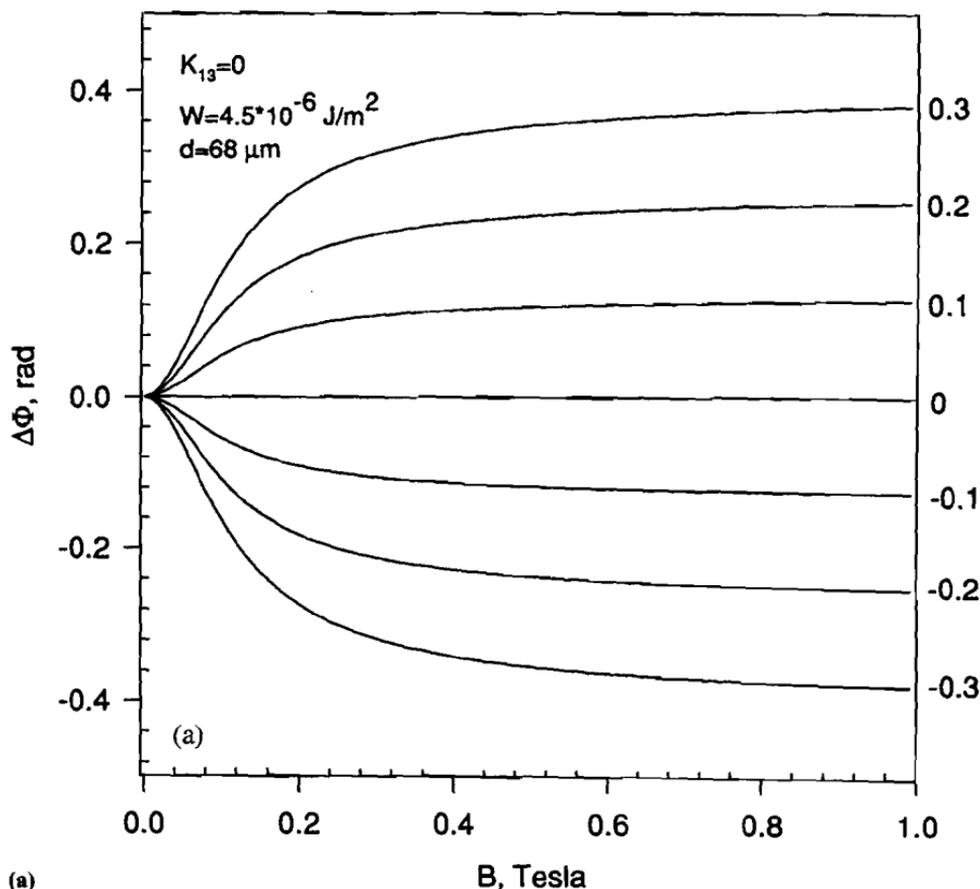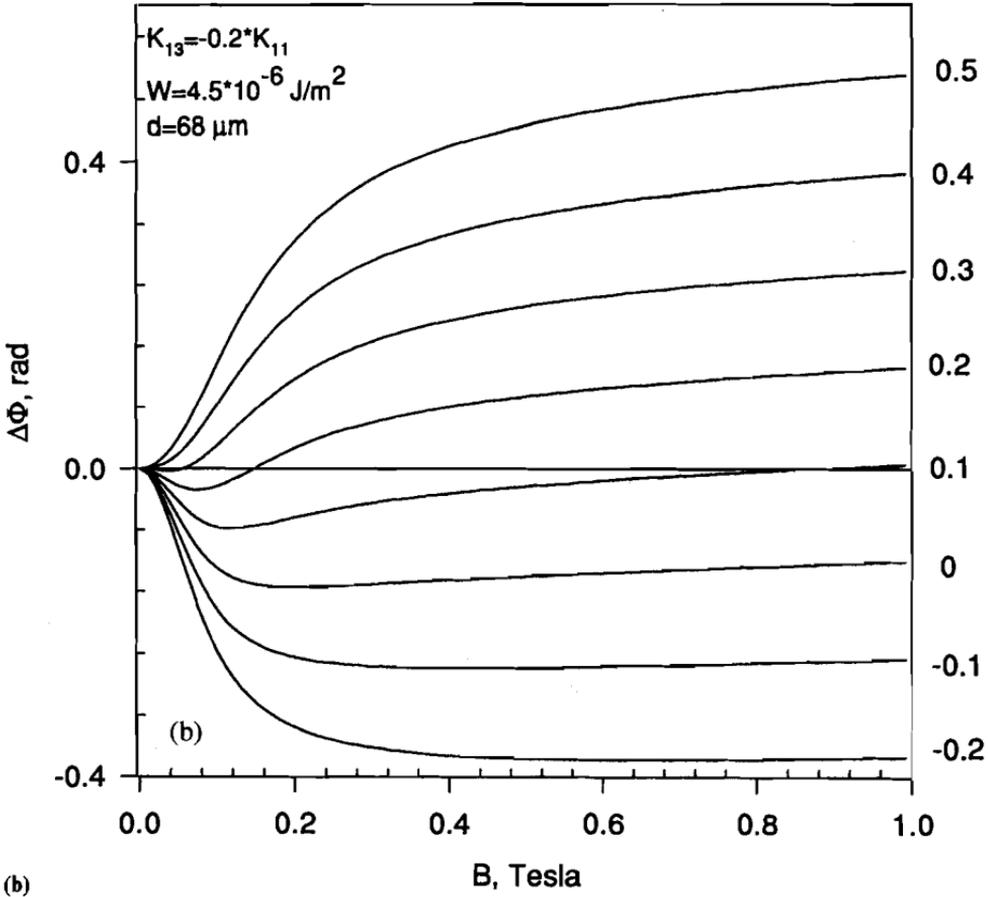


FIGURE 2

FIGURE 2    Phase retardation as a function of the magnetic field and the angle $\bar{\psi} = \bar{\theta} - \alpha$ (shown in degrees on the right side) for $K_{13} = 0(a)$ and $K_{13} = -0.2K_{11}(b)$; other parameters are indicated in the text.

rad and other subtle effects such as suppression of fluctuations in the field should be taken into account.

Another concern about the possible detection of the $K_{13}$-induced distortions is the hybridity of the cells, $\delta\bar{\theta} = \bar{\theta}_2 - \bar{\theta}_1 \neq 0$. When the hybridity is large, $|\delta\bar{\theta}| \sim 10^{\circ}$, it can mimic the $K_{13}$ effect. Galatola and Ziherl [26] found this mimic effect in numerical calculations. However, smaller hybridity, of the order of few degrees, does not shadow the possible $K_{13}$ effect, because the transmittive technique is sensitive to the symmetric modes. This observation is quite important, since the $K_{13}$-distortions are symmetric, while hybridity or small twist between the plates correspond to antisymmetric modes. Therefore, even if $\psi_s \sim \delta\bar{\theta}, \delta\varphi$, the "imperfection" antisymmetric modes contribute only to

the order $(\delta\bar{\theta})^2, (\delta\varphi)^2$ (the integral $\Phi$ above vanishes for an antisymmetric integrand). As shown in Ref.[27], the change in the phase retardation of the hybrid cell caused by the magnetic field reads as

$$\Delta\Phi \sim a_1 N + a_2(A^2 + N^2) + a_3(N^2 - A^2), \tag{8}$$

where $a_1\, a_2$ and $a_3$ are some coefficients depending on the refractive indices, easy direction and field orientation, and $N \sim (\bar{\theta}_1 + \bar{\theta}_2)/2, A \sim \bar{\theta}_2 - \bar{\theta}_1$ are small amplitudes of the symmetric and antisymmetric modes, respectively. Note that the antisymmetric mode contributes only to the order $(\bar{\theta}_2 - \bar{\theta}_1)^2$. The leading term in Eq.(8) is the term linear in $N$ which is defined by the average tilt rather than by the difference in the tilt at the two plates. If, for example, $\bar{\theta}_1 < \alpha < \bar{\theta}_2$, 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 $\delta\bar{\theta} = 2^0 (\bar{\theta}_1 = 80^0$ and $\bar{\theta}_2 = 78^0)$, $d = 80\mu$, 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 $0.2^0$ [27].

## EXPERIMENT

To favor detection of the predicted effect the cell should satisfy the following requirements: (1) small anchoring coefficient, preferably less than $10^{-5}\mathrm{J/m^2}$; (2) high tilt angle $\bar{\theta}$ (optimally $45°$, since the amplitude of the $K_{13}$-instability is proportional to $\sin 2\bar{\theta}$); (3) smooth substrate to maintain a constant value of the actual surface angle $\bar{\theta}$ and to prevent suppression of the $K_{13}$-deformations by profile-induced distortions. Taken together, these requirements represent an extraordinary difficult challenge, to say the least. Our initial results [28] performed for cells with rather small $\bar{\theta} \sim 10°$ gave an ambiguity in the discrimination of the possible $K_{13}$ and hybridity [26] mechanisms. The alignment methods that give higher 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. We did not observe non-monotonous behavior at these SiO-coated substrates, even with $\bar{\theta} \sim 60^0$, but these results were not conclusive since the roughness of the substrate was of the order of 100 Å or

larger. The results were also negative for polyimides coatings (e.g., material SE 610, Nissan Chemical Ind.) with $\bar{\theta} \sim 75-85^0$; here again the results might not be conclusive since the anchoring energy was rather high, $W > 10^{-5} \text{J/m}^2$. Finally, we found the photoalignment technique to be the most suitable method of surface coating to favor the appearence of the possible $K_{13}$-effect. Two alignment materials based on polysiloxane and polyvinylcinnamate were used. The polymers were spin-coated onto glass plates and subjected to polarized UV-illumination. As defined by atomic force microscopy, the polymer coatings were rather smooth (roughness amplitude less than 30Å at the wavelength 600 Å). Pairs of identically treated plates separated by distances $\sim 30-80\,\mu\text{m}$ were used to assemble the cells with uniform director tilt. The cells were filled with 5CB (pentylcyanobiphenyl, EM Industries) and cemented with a high temperature epoxy.

We report our results for two cells with tilted boundary conditions: No.1 and No.2. The cells were initially characterized by a crystal rotation method to determine both the tilt angle and the thickness of the filled cell [29]: $\bar{\theta} = (73.3 \pm 0.1)^0$, $d = (68.0 \pm 0.9)\,\mu\text{m}$ for No.1 and $\bar{\theta} = (68.85 \pm 0.1)^0$, $d = (54.5 \pm 0.9)\,\mu\text{m}$ for No.2. The cells were placed in oven (temperature control better than 30 mK), which in turn was mounted on an automatic rotary stage (angular positioning better than 0.01°) between the poles of an electromagnet. The cell were set in such a manner that the easy axis, the normal to the cell, and $B$ formed a plane. The accuracy of the angular settings was better than 0.1⁰. A linearly polarized laser beam (He-Ne, $\lambda = 632.8\,\text{nm}$, modulated at 400-800 Hz) was used to measure the phase retardation.

The optical response of the cells to the magnetic field applied closely to the easy axis is shown in Figures 3 and 4. For fixed $(\alpha - \bar{\theta})$, $\Delta\Phi(\mathbf{B})$ was defined using the experimental set-up described in Ref. [30]. The technique [30] allows also an independent measurement of the anchoring coefficient. We found $W = (4.5 \pm 2.0) \times 10^{-6} \text{J/m}^2$ for the cell No.1 and $W = (2 \pm 1) \times 10^{-6} \text{J/m}^2$ for the cell No.2.

As already stressed, if $K_{13} = 0$ and $\alpha = \bar{\theta}$, the first-order theory predicts that there are no director deviations for any value of the magnetic field and thus $\Delta\Phi = 0$; the position $\alpha = \bar{\theta}$ corresponds to the classical "magnetic null" position used in measurements of the pretilt angles [31]. If $\mathbf{B}$ is tilted away from the easy axis, $\Delta\Phi(\mathbf{B})$ should be monotonous (Fig. 2a). Experimental data, Figure 3,4, are consistent with the monotonous behavior of $\Delta\Phi(\mathbf{B})$. Figure 3 shows $\Delta\Phi(\mathbf{B})$ measured in cell No.1 for different angles $\alpha$. Figure 4 illustrates $\Delta\Phi(\mathbf{B})$ for the smallest possible misalignment between the field and the easy axis for both cells tested. In Figure 4a, solid lines correspond to the
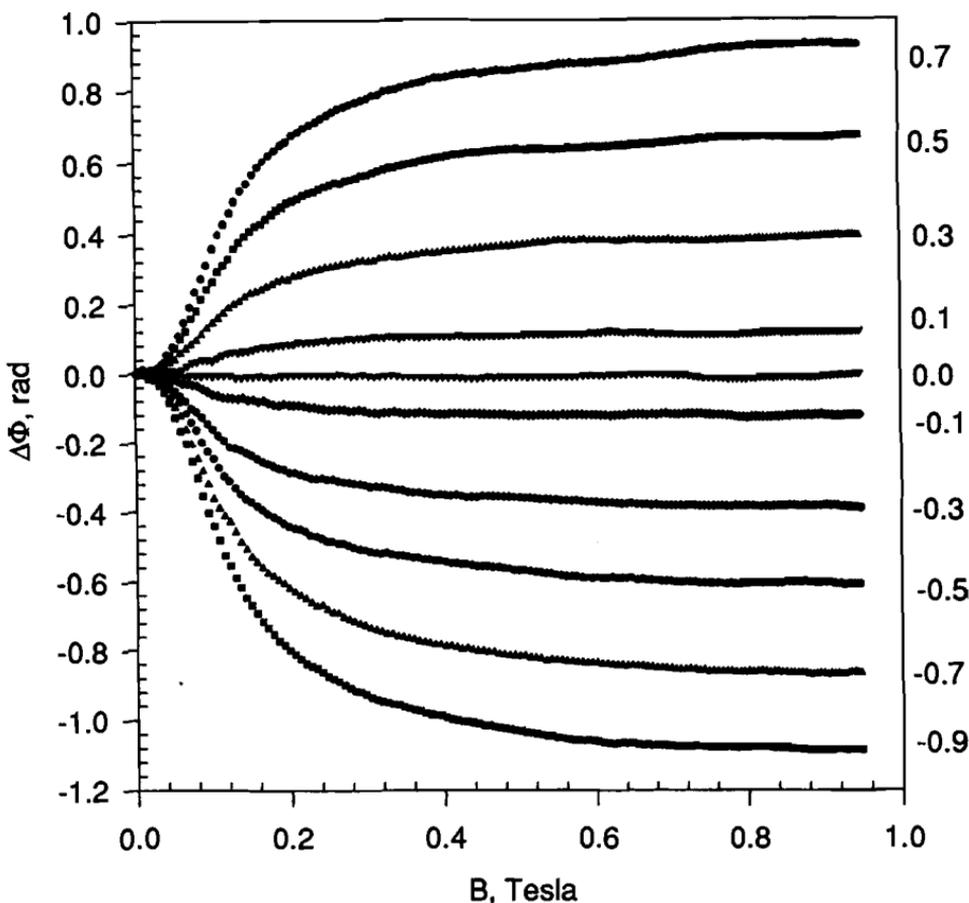
FIGURE 3    Phase retardation vs. magnetic field oriented at different angles with respect to the "easy direction" as measured for the cell No. 1.

expected $\Delta\Phi(\mathbf{B})$ for two non-zero $K_{13}$ values, $K_{13} = -0.05K_{11}$, and $K_{13} = -0.2K_{11}$. In Figure 4b the solid lines correspond to the expected $\Delta\Phi(\mathbf{B})$ for different $(\alpha - \bar{\theta})$ when $K_{13} = 0.05K_{11}$. The absence of significant deviations in $\Delta\Phi(\mathbf{B})$ (larger than $\sim 10^{-2}$ rad) was additionally verified for different directions of the testing light beam, including the directions normal and parallel to the magnetic field.

The results presented above lead us to the conclusion that the experiment does not show any clear manifestation of the spontaneous director deformations predicted by the first-order theory with non-zero elastic constant $K_{13}$.
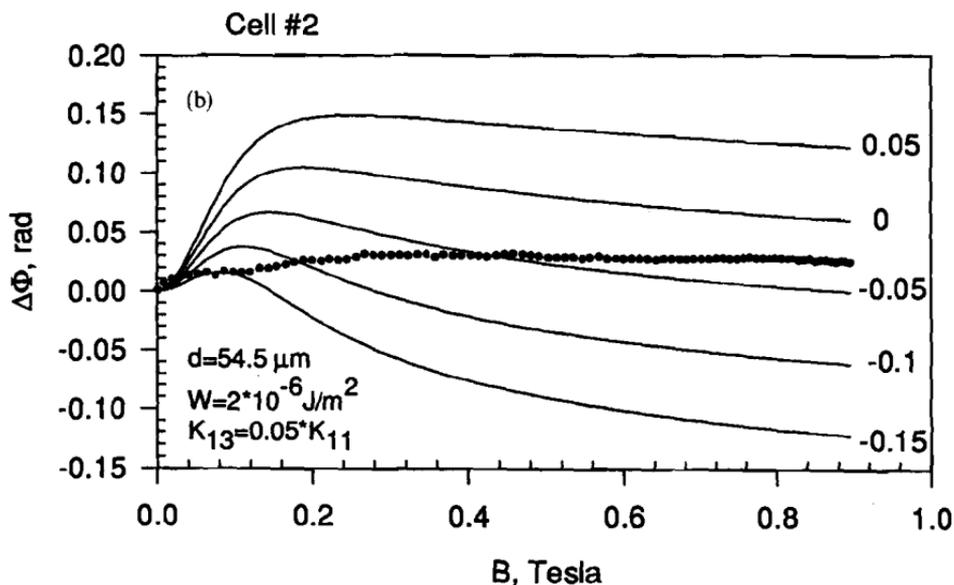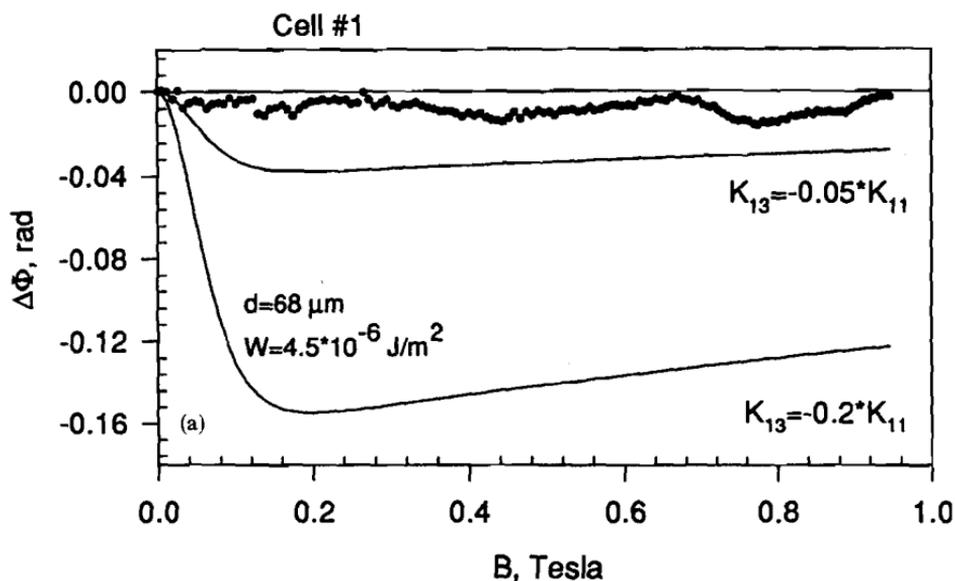
FIGURE 4 Experimental dependence (dots) of the phase retardation as a function of the magnetic field oriented as close to the "easy direction" as possible: (a) cell No. 1; solid lines are theoretical predictions of the first-order theory for two different values of $K_{13}$ and fixed angle $\bar{\psi} = \bar{\theta} - \alpha = 0$ between the "easy axis" and the magnetic field; (b) cell No. 2; solid lines are theoretical predictions of the first-order theory for different $\bar{\psi} = \bar{\theta} - \alpha$ (shown in degrees on the right side) and fixed $K_{13} = 0.05 K_{11}$.

## CONCLUSION

Formal application of the first-order theory to a cell with tilted n predicts that the initial uniform structure becomes unstable when the magnetic field is applied along n provided $K_{13} \neq 0$. The deformations can be detected in optical retardation $\Delta\Phi$ experiments; the distinctive feature is a non-monotonous behavior of $\Delta\Phi(B)$.

We prepared nematic cells using different alignment techniques to satisfy rather challenging requirements of the experimental test: high surface tilt, weak surface anchoring, and smooth orienting coating. No significant deviations that might have been caused by the $K_{13}$ elastic term in the first-order theory were detected in the experiments.

There are two possible conclusions to these results: either (1) $K_{13}$ is zero (the data above give the restriction $|K_{13}| < 0.05K_{11}$ or $K_{13} = (0.0 \pm 0.3) \times 10^{-12}$ N in the first-order theory) or (2) the first-order theory does not describe adequately the behavior of the cells. The first-order theory might oversimplify the situation: for example, it assumes that the spontaneous deformations caused by $K_{13}$ are completely suppressed by the higher-order terms. Resummation of higher-order terms in the free energy functional [32] should be one of the necessary steps to clarify the situation. This work is in progress.

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