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Magnetic field controlled optical phase retardation in a hybrid nematic cell

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We have studied the phase retardation of linearly polarized light in a hybrid nematic liquid crystal cell. For a certain range of directions of the applied magnetic field the phase retardation is found to change non-monotonically with the magnetic induction. The observed behaviour is described rather well by the standard Frank elastic theory. The corrections resulting from subsurface deformations, which are characteristic both for second order elasticity approach and for surface field theory, are also considered. The analysis of the experimental data suggests that the presence of distortions in the zero-field director configuration is the necessary condition for the non-monotonic phase retardation, which implies that such an experiment could be used for the detection of misalignment of the effective pretilts in a nematic cell.

1. Introduction

Recent articles [1, 2] have considered how the phase retardation of linearly polarized light transmitted through a nematic cell depends on the applied magnetic field. For certain orientations of the field, the phase retardation can exhibit an unusual non-monotonic dependence on the magnetic induction. While it seems that there are several mechanisms that could lead to this peculiar experimentally observed behaviour [3], misalignment of the pretilts at the two substrates (or briefly hybridity) is certainly among the most straightforward.

In this paper, the hypothesis that the non-monotonic phase retardation is caused by the hybridity of the cell is verified both experimentally and theoretically. The experimental part of the study is briefly described in §2. In §3, the problem is analysed in steps to separate the relevant mechanisms from each other. First the consequences of the misalignment of the pretilts in the cell are examined within the standard Frank elasticity theory [4], and then the so-called second order corrections to the elastic theory [5, 6] and the effect of surface fields [7] are considered. The relevance of the experimental method used for the detection of misalignment of the pretilts and subsurface deformations is discussed in §4, and §5 summarizes the results.

2. Experimental

We have prepared a nearly $\pi/2$ hybrid aligned cell by coating two glass substrates with different aligning layers. One layer is rubbed DuPont 2555 polyimide that provides almost planar orientation, the angle between the director and the substrate normal (θ_1) ranging from 88·2° to 88·8°. A spin-coated and cured lecithin layer provides antagonistic normal director orientation ($\theta_2 = 0°$) at the opposite plate. In this cell a very pronounced non-monotonic dependence of the relative phase retardation†, defined by $\Delta\Phi \equiv \Phi(B) - \Phi(B=0)$, on the magnetic induction has been observed for some values of the angle α_0 between the magnetic field and the normal to the cell (around 32·5°) (figure 1).

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†In the following, we will omit the adjective 'relative' since the scaling does not change the (non-)monotonicity of the phase retardation.

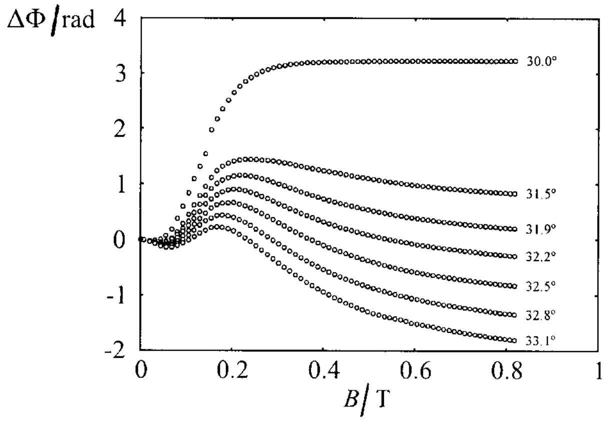

Figure 1. Phase retardation as a function of the magnetic induction for a hybrid cell. Experimental data are taken for different angles α_0 (indicated on the right hand side) between the field and the normal to the cell. As measured independently by the crystal rotation technique for cells with identical bounding plates, $\theta_1 = 88{\cdot}8°$, $\theta_2 = 0°$. Cell thickness $61{\cdot}5 \pm 0{\cdot}8\,\mu m$. All data are measured for light directed at the angle 56° to the cell normal; experimental accuracy corresponds to the size of the data points. Nematic material 4-pentyl-4′-cyanobiphenyl (5CB) was purchased from EM Industries.

The non-monotonicity of phase retardation in figure 1 is huge, of the order of 1 rad (cell thickness $d = 61{\cdot}5\,\mu m$) for $B \sim 0{\cdot}1$ T. However, it is possible that the non-monotonic behaviour is also caused by different (subtle) physical mechanisms, which might not all be as prominent as hybridity. In order to avoid the possibility that these are being masked by the effect of misalignment of the pretilts, we will analyse the phenomenon in cells with smaller hybridity and therefore generally less pronounced non-monotonicity. For example, in nematic cells with hybridity $\sim 0{\cdot}1°$ or less, the amplitude of non-monotonicity becomes less than 10^{-2} rad [2]. At this level, the optical changes can also be caused by magnetic suppression of the director fluctuations [8]. As measured by Poggi and Filippini [8], the corresponding change in the phase retardation is about 0·01 rad in a cell of thickness $d = 150\,\mu m$ subjected to $B \sim 0{\cdot}1$ T. In the following, we restrict ourselves to cells of thickness $d \sim 100\,\mu m$ or less and a non-monotonicity of the order of 0·1 rad (which therefore cannot be caused by the Faraday effect and related phenomena).

Experiments were performed for cells with alignment induced by phototransformation of polyvinyl cinnamate layers [9]. The polymers were spin-coated onto glass plates and subjected to polarized UV illumination. The technique is highly sensitive to the conditions of exposure (time, light intensity, polarization, etc.) [10, 11], and thus one might expect to get a slightly hybrid cell even when the plates are treated under similar conditions. However, the advantage is that photoalignment often allows one to avoid strong surface roughness, which is inevitable with other techniques. The surface roughness could cause non-controllable director distortions in the subsurface regions, which would contribute to the optical response to the magnetic field. As defined by atomic force microscopy, the resulting polymer coatings used were rather smooth (roughness amplitude ~ 3 nm at a wavelength of ~ 60 nm). The cell thickness was set by mylar spacers in the range 80–100 μm.

The cells were filled with a nematic material ZLI4801-100 (purchased from EM Industries) with parameters reported by the manufacturer as follows: ordinary refractive index $n_o = 1{\cdot}4832$; extraordinary refractive index $n_e = 1{\cdot}5869$ (both at wavelength $\lambda = 633$ nm); anisotropy of the magnetic susceptibility $\chi_a = 4\pi \times 0{\cdot}61 \times 10^{-7}$. To complete the characterization of the material, we measured the splay elastic constant, $K_1 = 13{\cdot}7 \times 10^{-12}$ N, and the bend elastic constant, $K_{33} = 18{\cdot}3 \times 10^{-12}$ N.

The crystal rotation method modified by van Sprang was used to obtain both the polar tilt angle and the cell thickness [12] and gave the following results for the cell we discuss below: the average value of the director tilt $\theta = 80{\cdot}4° \pm 0{\cdot}2°$ and the corresponding cell thickness $d = 95{\cdot}7\,\mu m$. Note that the crystal rotation technique assumes that the director is absolutely uniform; possible non-uniformity of the director across the cell (caused, for example, by evolution of the photoprocessed layers with time) results in renormalization of the apparent tilt and thickness.

The optical response to the field was measured as follows. For fixed α_0, the phase retardation $\Delta\Phi = \Delta\Phi(B)$ was defined by the Senarmont technique. A rotary stage (angular positioning better than 0·01°) was used to set the cell between the poles of an electromagnet in such a manner that the easy axis, the normal to the cell, and B formed a plane (figure 2). A linearly polarized laser beam (He–Ne, $\lambda = 632{\cdot}8$ nm, modulated at 400–800 Hz)

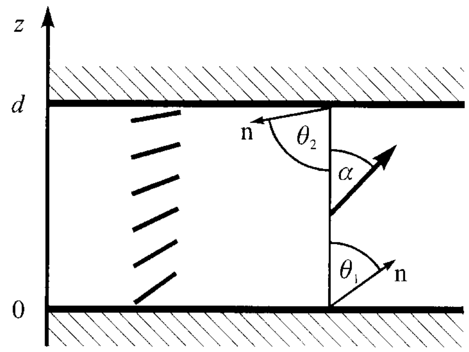

Figure 2. The sample is sandwiched between two plates separated by distance d. The preferred values of the tilt angle at the lower and upper plates are θ_1 and θ_2, respectively; α stands for the angle between the magnetic field and the substrate normal.

was directed normally to the cell. The accuracy of the angular settings was better than 0·1°.

Figure 3 shows a non-monotonic dependence of the phase retardation on the magnetic induction for the cell with parameters specified above. The normal geometry of the light incidence allows one to compare these experimental data with the predictions of different elastic theories since in this case the phase retardation and the director profile across the cell are related by a simple expression:

$$\Phi = \frac{2\pi n_0}{\lambda} \int_0^d \frac{\mathrm{d}z}{(1 - \rho \sin^2 \theta(z))^{1/2}} \tag{1}$$

where $\rho = 1 - (n_o/n_e)^2$ and $\theta(z)$ is the director tilt angle profile.

3. Interpretation of data

Qualitatively, the non-monotonic behaviour of the phase retardation in a hybrid cell and its dependence on the hybridity of the cell can be understood from analytical calculations of $\Delta\Phi$ which are possible when the hybridity $\theta_1 - \theta_2$ is small, of the order of $\sim 1°$. The calculation performed for a Frank–Oseen elastic model with no divergence terms [2] yields

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

where the coefficients a_1, a_2, a_3 depend on the refractive indices, surface orientation, magnetic induction and direction of the applied field; $N \sim (\theta_1 + \theta_2)/2$ is the average director tilt, whereas $A \sim \theta_1 - \theta_2$ is the hybridity of the cell. Clearly, when $\theta_1 < \alpha_0 < \theta_2$, the increase in the magnetic induction would increase the polar angle at one plate and decrease it at the other. These changes would contribute mainly to the quantity $A \sim \theta_1 - \theta_2$ rather than to $N \sim (\theta_1 + \theta_2)/2$. Thus the hybridity is a natural reason for non-monotonic changes in phase retardation. Note, however, that the leading term in the last expression for $\Delta\Phi$ is linear in N; the hybridity starts to contribute only in the second order term $\sim A^2 \sim (\theta_1 - \theta_2)^2$. This is why the corresponding non-monotonicity is rather weak and quickly fades when the hybridity is reduced. In the following text, a quantitative analysis of the phenomenon is presented.

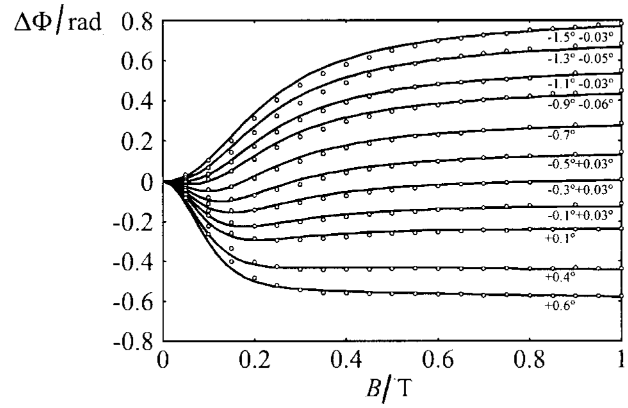

Figure 3. Phase retardation in a moderately hybrid cell as a function of the magnetic induction. Circles are experimental data; solid lines are fits obtained within first order elasticity theory with $\theta_1 = 70{\cdot}3°$, $\theta_2 = 91{\cdot}8°$, $\alpha^* = 79{\cdot}1°$, and $W = 6{\cdot}3 \times 10^{-6}\ \mathrm{J\,m^{-2}}$. The fits are labelled with the orientation of the magnetic field $[(\alpha - \alpha^*) + \text{correction}]$.

3.1. *First order elasticity*

In the presence of the magnetic field, the Frank free energy density corresponding to planar distortions $[\mathbf{n}(z) = (\sin\theta, 0, \cos\theta)$ with $\theta = \theta(z)]$ is given by

$$f = \frac{1}{2}\left[(K_{11} \sin^2 \theta + K_{33} \cos^2 \theta)\theta'^2 + \frac{\chi_a B^2}{\mu_0} \sin^2(\theta - \alpha) \right] \tag{3}$$

where K_{11} and K_{33} are the splay and bend elastic constants, respectively, $\theta' \equiv \mathrm{d}\theta/\mathrm{d}z$, χ_a is the anisotropy of the magnetic susceptibility, B is the magnetic induction, and α is the angle between B and the layer normal [4]. As usual, the interaction between the director field and the two plates is described by the Rapini–Papoular terms

$$F_S = \frac{1}{2} W \sin^2(\theta - \theta_i) \tag{4}$$

where W is the anchoring strength (assumed to be the same at both plates), whereas θ and θ_i are the actual and the preferred effective tilts at the substrate, respectively; $i = 1$ at $z = 0$ and $i = 2$ at $z = d$. The equilibrium configuration is determined by the minimization of the total free energy, which leads to the Euler–Lagrange equation

$$(1 + \kappa \sin^2 \theta)\theta'' + \frac{1}{2}\kappa \sin(2\theta)\theta'^2 - \frac{1}{2}\xi^{-2} \sin 2(\theta - \alpha) = 0 \tag{5}$$

where $\kappa \equiv K_{11}/K_{33} - 1$ and $\xi = (\mu_0 K_{33}/\chi_a B^2)^{1/2}$ is the magnetic coherence length. The boundary conditions read

$$(1 + \kappa \sin^2 \theta)\theta' - \frac{1}{2} L^{-1} \sin 2(\theta - \theta_1) = 0 \tag{6}$$

at $z = 0$ and

$$(1 + \kappa \sin^2 \theta)\theta' + \frac{1}{2} L^{-1} \sin 2(\theta - \theta_2) = 0 \tag{7}$$

at $z = d$; here $L = K_{33}/W$ is the extrapolation length.

The above system is solved numerically and the tilt angle profiles are used to calculate the phase retardation. In order to minimize the number of adjustable parameters and simplify the fitting procedure, the relative orientation $\alpha - \alpha^*$ (α^* being the reference inclination) of the 11 different directions of the magnetic field at which the phase retardation has been measured is supposed to be known precisely. (However, a brief inspection of the experimental data suggests that this cannot be quite true.) Then the set of unknown parameters is reduced to α^*, θ_1, θ_2, and L. The fits to the experimental data turn out to be rather insensitive to the value of the anchoring strength provided it exceeds $\sim 5\times10^{-6}$ J m^{-2} and for $W=6{\cdot}2\times10^{-6}$ J m^{-2}, the theoretical predictions are closest to the observed phase retardation for $\alpha^*=79{\cdot}1°$, $\theta_1=70{\cdot}3°$ and $\theta_2=91{\cdot}8°$. For larger values of W, the hybridity $\theta_2-\theta_1$ is up to 1° smaller (so that the accuracy of the calculated values of θ_1 and θ_2 is below 1°) whereas α^* is almost independent of the anchoring strength. If W is smaller than 5×10^{-6} J m^{-2}, the actual variation of the tilt angle over the sample becomes too small to achieve considerable non-monotonicity of the phase retardation even for very large hybridity.

Once the free parameters are determined, it becomes clear that one must allow for the inaccuracy of the relative orientations of the magnetic field $\alpha-\alpha^*$. If these are adjusted to compensate for the experimental inaccuracy (the adjustments not exceeding 0·05°, which is below the inaccuracy of angular setting in the experiment $\sim$0·1°), the matching of the experimental and the theoretical predictions is very good (figure 3).

3.2. *Subsurface deformations*

3.2.1. *Second order elasticity*

As shown by Nehring and Saupe, the free energy density could contain another symmetry-allowed elastic contribution, the so-called splay–bend term $K_{13}\nabla\,(\mathbf{n}\nabla\,\mathbf{n})$ [13]. In order to incorporate this type of deformation into the Frank elastic energy consistently, the second order elastic term $K^*(\theta'')^2$ must also be included, and the resulting free energy functional leads to strong subsurface deformations of the director field [5, 14, 15]. Could the phase retardation experiment be sensitive to such distortions and, consequently, to the value of K_{13}?

Using Gauss' theorem, the splay–bend term can be transformed into the surface contribution of the form $2K_{13}\theta_i'\sin(2\theta_i)$ so that the corresponding Euler–Lagrange equation differs from equation (5) only by the term $-\delta^2\theta^{(\mathrm{IV})}$ on the left hand side; here $\delta\equiv(K^*/K_{33})^{1/2}$ is of the order of the molecular length [14]. On the other hand, there are four boundary conditions instead of two:

$$\begin{aligned}&\delta^2\theta'''+[R\cos(2\theta)-(1+\kappa\sin^2\theta)]\theta'\\&\quad+\frac{1}{2}L^{-1}\sin 2(\theta-\theta_1)=0\\&\delta^2\theta''-\frac{1}{2}R\sin(2\theta)=0\end{aligned}\tag{8}$$

at $z=0$, and

$$\begin{aligned}&\delta^2\theta'''+[R\cos(2\theta)-(1+\kappa\sin^2\theta)]\theta'\\&\quad-\frac{1}{2}L^{-1}\sin 2(\theta-\theta_2)=0\\&\delta^2\theta''-\frac{1}{2}R\sin(2\theta)=0\end{aligned}\tag{9}$$

at $z=d$. Here $R\equiv K_{13}/K_{33}$.

Strong subsurface deformations, predicted by the above corrections to the first order elastic theory, are almost insensitive to external forces (e.g. a magnetic field), provided these are moderate. This implies that the concept of nominal pretilt θ_i is quite irrelevant and should be replaced by the effective pretilt, the difference between the two being related to the sign and the magnitude of the splay–bend elastic constant K_{13} and the second order elastic constant K^*.

The phase retardation is an integrated quantity and quite independent of the details of the subsurface variation of the tilt angle, which is limited to a region of thickness $\delta\ll d$, λ [15]. This means that any choice of W, K^*, K_{13}, θ_1, and θ_2 that gives rise to the same effective pretilts will produce the same phase retardation. Indeed, by performing the fits with different values of K_{13}, one ends up with a number of equally good theoretical descriptions of the experimental data. Depending on the sign and the magnitude of the splay–bend elastic constant, the nominal hybridity might be either larger or smaller than in the case of first order theory, but, as already pointed out, this quantity is not really informative itself. For example, for $W=2\times10^{-3}$ J m^{-2}, $K_{13}=-0{\cdot}3K_{33}=-5{\cdot}5\times10^{-12}$ N, and $K^*=10^{-10}K_{33}d^2=1{\cdot}7\times10^{-25}$ Nm^2 (i.e. $\delta\simeq1$ nm), second order theory gives best fits with $\theta_1=76{\cdot}7°$, $\theta_2=90{\cdot}6°$, and $\alpha^*=78{\cdot}5°$. As in the preceding case, the relative orientations of the magnetic field, $\alpha-\alpha^*$, must be slightly adjusted, the magnitude of the corrections not exceeding 0·04°. (The figure with the fits to the data is virtually indistinguishable from figure 3 and is not presented in the paper.) This indicates that the splay–bend and the second order elastic constants cannot be accurately determined by the phase retardation experiment.

However, there is an important difference between the predictions of the standard Frank theory and second order elasticity. As mentioned above, the former gives practically identical fits for $W\lesssim5\times10^{-6}$ J m^{-2}. In second order elastic theory the lower limit of the relevant anchoring strengths is significantly larger: for the above values of K_{13} and K^*, W must exceed $\sim2\times10^{-3}$ J m^{-2}, and if K^* is increased by a factor of 100 (which means that $\delta\simeq10$ nm), the lower limit of W values that give good fits to the data is about 2×10^{-4} J m^{-2}. This means that the anchoring strengths required by second order theory are about two orders of magnitude larger than those from Frank elasticity. This is a direct consequence of the well-known fact that within second order theory the splay–bend term produces intrinsic anchoring that renormalizes the strength of the external anchoring and the corresponding easy axis [14]. The strength of the intrinsic anchoring $W_{\mathrm{int}}\sim K_{13}^2/K_{33}\delta$ is consistent with the above estimates of the lower limit of W.

3.2.2. *Surface field approach*

Strong subsurface deformations can also occur due to surface electric fields resulting from, for example, selective adsorption of ions [7]. Such a field is localized to the subsurface layer and if **E** is perpendicular to the substrate, the corresponding free energy density is given by $-\frac{1}{2}\varepsilon_0\varepsilon_{\mathrm{a}}E^2(z)\cos^2\theta$, where ε is the anisotropy of the electric susceptibility and E is the strength of the surface electric field. This effect can be taken into account by adding

$$\frac{1}{2}u(z)\sin^2\theta \tag{10}$$

to the standard Frank free energy density. The model surface field profile is often of the form

$$u(z)=u_0\frac{\mathrm{ch}\left[(z-d/2)/\Lambda\right]}{\mathrm{ch}\left[(d/2)/\Lambda\right]}, \tag{11}$$

where u_0 is the strength of the surface field and Λ is the characteristic penetration length (in the case of selective ion adsorption, the Debye screening length).

The above theory does not modify the usual first order elasticity formalism significantly: one only has to add a term $-\frac{1}{2}U(z)\sin 2\theta$ [where $U(z)\equiv u(z)/K_{33}$] to the left-hand side of the Euler–Lagrange equation (5). For a given set of parameters of the surface field (i.e. u_0 and Λ), the equilibrium tilt angle profiles are found numerically and the fits to experimental data are very similar to those obtained in the frameworks of first and second order theories. For $W=2\times10^{-4}$ J m^{-2}, $\Lambda=2\times10^{-3}d=0{\cdot}19$ µm, and $u_0=2{\cdot}5\times10^4K_{33}/d^2=$ 50 N m^{-2}, the best fit is obtained with $\theta_1=73{\cdot}0°$, $\theta_2=91{\cdot}0°$, and $\alpha^*=78{\cdot}7°$. (Again, since the fit itself is practically identical to figure 3, it would not be very informative to show this in a separate figure.) As in the case of second order elasticity, there is a range of values of u_0, Λ, and W that give virtually the same fits, so that none of these three parameters of the set-up can be extracted from the experimental data.

4. Discussion

The non-monotonic behaviour of the phase retardation in a nematic cell seems to be intimately related to a system characterized by a distorted zero-field director configuration. Within first order elasticity, this occurs only in a hybrid cell. On the other hand, both second order corrections to the elastic theory and the surface fields, which are characterized by strong subsurface deformations, also give rise to non-monotonic phase retardation in the absence of hybridity, although for physically relevant values of the second order elastic constant and strength of the surface field, the non-monotonicity would occur only for strong magnetic fields.

Why does a distorted zero-field director profile necessarily lead to a non-monotonic phase retardation? This can be most easily visualized by the following argument. Suppose that the director field in question is non-uniform for $B=0$ and that the anchoring is not strong. As the magnetic induction is increased, the nematic director becomes more and more uniform and aligned with the magnetic field, i.e. $\lim_{B\to\infty}\theta(z)=\alpha$. Now there must exist a special orientation of the magnetic field such that the phase retardations of the zero-field distorted configuration $\theta_0(z)$ and the high-field uniform cell $\theta(z)=\tilde{\alpha}$ match, $\tilde{\alpha}$ being given by

$$\tilde{\alpha}=\arcsin\left(\frac{1}{\rho}\left\{1-d^2\left[\int_0^d\frac{\mathrm{d}z}{(1-\rho\sin^2\theta_0(z))^{1/2}}\right]^{-2}\right\}\right)^{1/2}. \tag{12}$$

If the magnetic field is oriented along this 'null' direction, the phase retardation has to saturate at 0 as $B\to\infty$. Since $\Delta\Phi(B=0)=0$, it must have either a minimum or a maximum at some finite value of B and thus exhibit a non-monotonic dependence. Therefore, this type of behaviour of the phase retardation should occur in every cell characterized by a non-uniform director field for $B=0$. However, the range of orientations of the magnetic field that give rise to non-monotonic $\Delta\Phi(B)$ may be very narrow so that the phenomenon might not be easily detectable. An example of non-monotonic phase retardation in a nonhybrid cell described by second order theory is presented in figure 4.

5. Conclusions

This experiment revealed an interesting feature of non-uniform nematic cells in an external aligning field: for a certain range of orientations of the field, the phase retardation of the transmitted light will exhibit non-monotonic dependence on its strength, which is consistent with the findings of a similar study of a nematic confined to a wedge cell [16]. If the non-monotonicity occurs at moderate magnetic fields ($B\lesssim1$ T), the two effective pretilts must be different, which means that such an experiment could be used for the determination of the hybridity of nematic cells. In this case, the

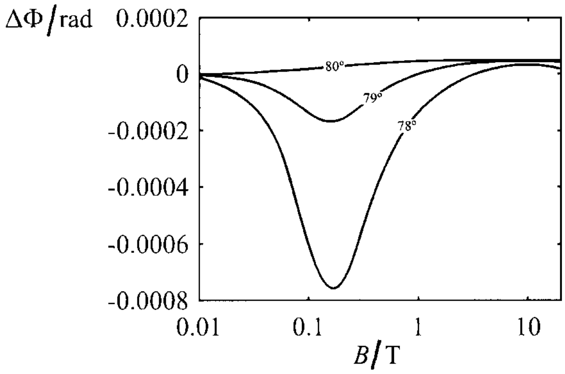

Figure 4. Second order theory prediction of the phase retardation for $\theta_1 = 80{\cdot}0°$ and $\theta_2 = 78°$, 79°, and 80°. (Other parameters: $W \to \infty$, $K_{13} = -0{\cdot}3K_{33} = -5{\cdot}5 \times 10^{-12}$ N, and $K^* = 10^{-10}K_{33}d^2 = 1{\cdot}7 \times 10^{-29}$ Nm^2.) The curves are labelled with the θ_2 value. In each case, the inclination of the magnetic field is such that $\lim_{B \to \infty} \Delta\Phi = 0$. Should the non-monotonicity occur due to subsurface deformations, the maximum magnitude of the phase retardation is generically rather small.

behaviour of the optical phase retardation can be explained in terms of standard Frank theory with $K_{13} = K_{24} = 0$, which implies that the contribution of subsurface deformations is not detectable so that the values of the corresponding model parameters (K_{13}, K^*; u_0, Λ) cannot be extracted from the experimental data. On the other hand, the non-monotonic behaviour that would appear at higher fields might be caused by strong (subsurface) deformations, implying that their existence could be proven by an experiment similar to ours, but carried out in strong magnetic fields, which would therefore also serve as a test of the second order elastic theory. However, the theoretical analysis of the phenomenon at high fields would also include some subtle effects, e.g. the variation of the scalar order parameter due to the applied field [8].

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