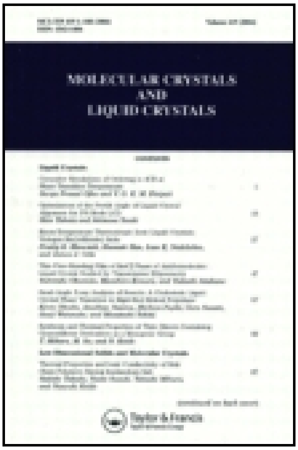

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Study of Nematics-Substrate Coupling Characteristics and Nematics Material Parameters in Dependence on Cell Thickness by Using Wedge Cells of Different Cover Coatings

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The dependence of characteristics of the coupling between nematics and substrates on the planar cell thickness was suggested in the literature. The aim of this study is verification of this hypothesis by exploiting well controlled experimental conditions and stable computational algorithms for finding the characteristics of the nematics-substrate coupling together with the anisotropy of diamagnetic susceptibility and the splay and bend elastic constants via solving inverse problems. Wedge cells (of the wedge angle of the order of few milliradians) were used for studying Fréedericksz' transition of splay-bend type. The phase shift between ordinary and extraordinary rays of light normally incident on cell boundary was used as a physical quantity monitoring the state of the director field inside a cell in selected zones, equivalent to flat-parallel cells of different thicknesses. The optical response was measured as a function of voltage or magnetic field applied to a cell. Experiments were performed with several cells, filled with the 4′-pentyl-4-cyanobiphenyl nematic liquid crystal (5CB), of cover coatings made of different polyimides producing strong anchoring. The magnitudes of the anisotropy of diamagnetic susceptibility, the splay and bend elastic constants and the anchoring angle were found through fitting simulated cell optical transmittance characteristics to the

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measured one for each equivalent cell separately. The apparent dependence of these magnitudes on cell thickness was observed when the fitting was performed to reach the minimal fit error. This effect is interpreted as misleading and corrected by considering the fitting with the same magnitudes of material constants to all measured transmittance characteristics, with a little greater fit errors. The independence of nematics constants and anchoring angle of cell thickness is finally concluded.

Keywords: anchoring characteristics; anisotropy of diamagnetic susceptibility; inverse problem; nematics parameter identification; wedge cell; 4′-pentyl-4-cyanobiphenyl (5CB)

EXPERIMENTAL ARRANGEMENT

Each wedge cell [1,2] was made of glass plates (of size 22×35 mm), coated with indium-tin oxide electrodes and orienting layer of polyimide. Polyimides were spin deposited on the electrode surfaces and polymerised in thermal or photo-induced process [3] and eventually rubbed. The plates were glued without a spacer along one edge and with a spacer (of thickness about 200 μm) along the other. The orientation of the nematics molecules enforced by substrates was parallel to the boundaries and to the wedge edge. The cell was placed in a thermostatic stage between the polariser and analyser crossed in the measurement system, consisting of a He-Ne laser and a microscope with a photodetector, and between pole pieces of an electromagnet. In the normally incident light the interference fringes appeared in the positions of the cell thickness equal to $d_j = \Delta_j n_a^{-1} = j\lambda n_a^{-1}$, corresponding to the difference of the optical path Δ, the phase shift Φ, the light wavelength λ and the birefringence $n_a \equiv n_e - n_o$; $\Phi_j = 2\pi\Delta_j\lambda^{-1}$. In the small neighbourhood of each fringe position a wedge cell can be treated as the planar one, due to very small wedge angle. The intensity of normally incident light transmitted through the cell in the positions of selected interference fringes was recorded as a function of applied voltage and magnetic field. In this way each wedge cell was equivalent to a system of planar cells of different thicknesses. To avoid electric charge transfer effects the low-frequency (1.5 kHz) sinusoidal voltage was used.

THEORETICAL OVERVIEW

Plane stationary deformations of the nematics in a wedge cell in the position of an interference fringe were induced by aligning covers and external electric or magnetic field, with the field vectors, the anchoring direction and the director being all in the same plane. The optical transmittance of a cell was recorded as a function of voltage applied to the cell electrodes or magnetic field. The rate of increasing the magnitude of an external field was sufficiently small to avoid dynamical effects during the registration of each characteristics. In the small vicinity of a fringe position a wedge cell can be treated as the flat-parallel nematic layer and described in one-dimensional approximation, with good accuracy. Let, in the Cartesian co-ordinates, the Oxz plane be the deformation plane, with the Oz axis perpendicular to the layer, the electric field vector in the Oz direction (as being induced by a voltage applied to the cover electrodes) and the magnetic field vector $\vec{B}$ forming the angle ψ with the Oz axis. All physical quantities are assumed to depend only on z, in the layer bounded by planes $z = 0$ and $z = d$, e.g.:

$$\vec{n}(z) = (\cos\vartheta(z), 0, \sin\vartheta(z)), \quad \vec{E}(z) = (0, 0, E(z)),$$
$$\vec{B}(z) = (B\sin\psi, 0, B\cos\psi).$$

The driving equations [1,2,4] describe the director field vector $\vec{n}$ through the tilt angle ϑ (between $\vec{n}$ and the Ox axis):

$$(K_{11}\cos^2\vartheta + K_{33}\sin^2\vartheta)\vartheta'' + (K_{33} - K_{11})\sin\vartheta\cos\vartheta\vartheta'^2 + \frac{\varepsilon_0\varepsilon_a\varepsilon_e^2 U^2 \sin\vartheta\cos\vartheta}{d^2\left(\varepsilon_\perp\cos^2\vartheta + \varepsilon_{||}\sin^2\vartheta\right)^2} + \frac{\chi_a B^2}{\mu_0}\sin(\vartheta+\psi)\cos(\vartheta+\psi) = 0$$

$$\varepsilon_e = \left\{\frac{1}{d}\int_0^d \frac{1}{[\varepsilon_\perp\cos^2\vartheta(z) + \varepsilon_{||}\sin^2\vartheta(z)]}\,dz\right\}^{-1}$$

where: K_{11}, K_{33} are the splay and bend Frank elastic constants, $\varepsilon_\perp$, $\varepsilon_{||}$ are the electric permittivities, $\varepsilon_a = \varepsilon_{||} - \varepsilon_\perp$, χ_a is the anisotropy of diamagnetic susceptibility, ε_e is the effective electric permittivity of the layer, U is the voltage applied to the layer. In the case of the aligning coatings identical on both covers, like studied here, function $\vartheta(z)$ should satisfy the symmetric boundary condition which can be written down in the form [1,2,4]

$$\vartheta(0) = \vartheta(d) = \Theta(T_b)$$

where T_b is the torque per unit area. This torque is transmitted from the deformed nematics bulk to the layer boundaries and for a function

$\vartheta(z)$, satisfying the driving equations in the flat-parallel layer, can be calculated by the following formula [4]:

$$T_b = \int_0^d \left[(K_{11} - K_{33}) \cdot \sin\vartheta(z) \cdot \cos\vartheta(z) \cdot \vartheta'(z)^2\right] dz$$
$$+ \int_0^d \left[\frac{\varepsilon_0 \varepsilon_a \varepsilon_e^2 U^2 \sin\vartheta(z) \cdot \cos\vartheta(z)}{d^2[\varepsilon_\perp \cos^2\vartheta(z) + \varepsilon_{||} \sin^2\vartheta(z)]^2} + \frac{\chi_a B^2}{\mu_0} \sin(\vartheta(z) + \psi) \cdot \cos(\vartheta(z) + \psi)\right] dz$$

This torque is balanced by the torque originated from the boundary interactions that is equal to the first derivative of the boundary free energy per unit area [4] (with respect to the boundary value $\vartheta(0)$ or $\vartheta(d)$). In computations it is more convenient to use a function $\vartheta(0) = \Theta(T_b)$, as above, instead of an inverse one $T_b = \Theta^{-1}(\vartheta(0))$. In case of strong anchoring the boundary values of the tilt angle are constant and equal to the anchoring angle ϑ_0: $\vartheta(0) = \vartheta(d) = \vartheta_0$. Function $\Theta(T_b)$ should be non-decreasing; in this work it is modelled as a non-decreasing cubic spline. Such a modelling is applied to investigate which type of anchoring is dealt with, weak or strong. The strong anchoring is treated as the special case of the weak one, corresponding to a constant function $\Theta(T_b)$. The strong anchoring was realised from experimental data analysed below for all polyimides used as aligning substrates.

Based on the equations given above, the inverse problem can be formulated. Every electric or magnetic characteristics of a nematic cell as a birefringence system, i.e., the dependence of the phase shift between ordinary and extraordinary rays of light (normally incident and transmitted through the nematics layer) on external fields, $\varphi = \varphi(U; B, \psi)$,

$$\varphi = \frac{2\pi}{\lambda} d \left[\frac{1}{d} \int_0^d \frac{n_e \cdot n_o}{\sqrt{n_e^2 \cdot \sin^2\vartheta(z) + n_o^2 \cdot \cos^2\vartheta(z)}} dz - n_o\right],$$

contains the information about material constants $K_{11}, K_{33}, \chi_a, \Theta$ (where Θ is a function characterising the nematics-substrate coupling and not the nematics itself; e.g., a cubic spline modelling $\Theta(T_b)$). Let $p = (K_{11}, K_{33}, \chi_a, \Theta)$ denote the set of unknown material parameters (where, e.g., Θ states all coefficients of a non-decreasing cubic spline). For any sequence of measurements $(\varphi_e(U^i; B^i, \psi^i))_{i=1}^n$ one can calculate a corresponding sequence of values of light phase shift

$(\varphi_c(U^i; B^i, \psi^i; p))_{i=1}^n$ (by assigning some values to the material parameters). By minimising the similarity functional [1,2]

$$S(p) = \left\{ \frac{1}{n} \sum_{i=1}^{n} \left[\frac{\varphi_e(U^i; B^i, \psi^i) - \varphi_c(U^i; B^i, \psi^i; p)}{\varphi_e(U^i; B^i, \psi^i))} \right]^2 \right\}^{\frac{1}{2}}$$

as the objective function of independent variables p, one approximately finds the unknown magnitudes of material parameters p as corresponding to the minimum. The minimisation should stop when $S(p)$ achieves the magnitude corresponding to errors in the phase shift values experimentally determined, that is usually greater than the least-possible one. The values of the similarity functional $S(p)$ can be interpreted as the mean relative error of approximating each point of studied characteristics.

RESULT OF EXPERIMENT

The dependence of characteristics of the coupling between nematics and substrates on the planar cell thickness was concluded from

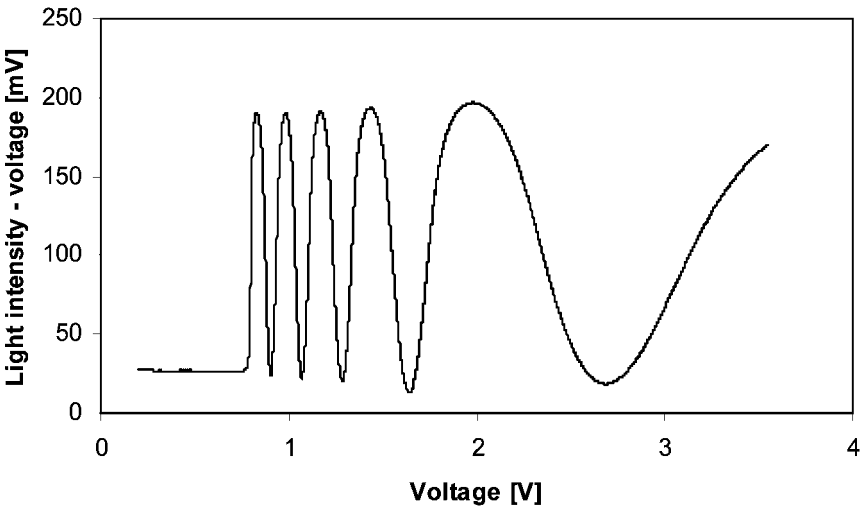

FIGURE 1 Intensity of normally incident light (expressed as equivalent voltage), transmitted through the 5CB-PVCN20 cell, as a function of voltage, recorded for the 6th fringe corresponding to the cell thickness 20.8 μm at the temperature 22.7°C.

measurements of the saturation voltage [5]. Some partial explanation of this phenomenon was proposed later [6] by taking into account the surface polarisation of nematics, but it still remains an open question. The following results verify this hypothesis negatively.

The wedge cells were filled with 4′-pentyl-4-cyanobiphenyl (5CB). Two polyimides, the commercial Nissan SE130 polyimide (NSE) and the photo-aligning poly(vinyl cinnamate), exposed to ultraviolet (from the same radiation source) during 10 or 20 minutes (PVCN10 or PVCN20), were used as aligning substances. The orienting surfaces were prepared by rubbing in the direction parallel to the wedge edge.

The intensity of light, transmitted through the birefringence system, as a function of a magnetic field B perpendicular to the cell boundaries ($\psi = 0$) and as a function of a voltage U (i.e., of an electric field), was recorded for twenty nine interference fringes in the three cells (4–20 m and 4–20e for 5CB-PVCN20, 6–13e for 5CB-PVCN10 and 5–11e for 5CB–NSE), corresponding to the cell thickness in the range 13.9–69.4 μm, at the temperature close to 22.7°C (in the range inferior to 0.1°C), stabilised with the variations inferior to 0.1°C. The

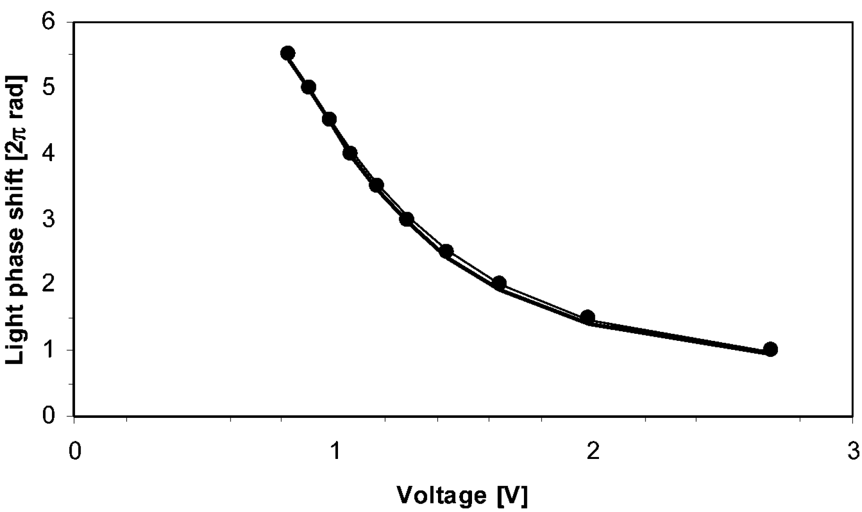

FIGURE 2 Phase shift between ordinary and extraordinary rays of transmitted light (in 2π radian units) as a function of voltage, determined after the characteristics recorded for the 6th fringe in the 5CB-PVCN20 cell (shown in Fig. 1), corresponding to the cell thickness 20.8 μm (circles), and calculated with the best-fit parameters $K_{11} = 6.12$ pN, $K_{33} = 9.94$ pN, $\vartheta_0 = 8.6$ mrad (thin line, $S(p) = 0.014$) and with the averaged ones $K_{11} = 6.33$ pN, $K_{33} = 8.59$ pN, $\vartheta_0 = 10$ mrad (thick line, $S(\bar{p}) = 0.032$).

phase shift between the ordinary and extraordinary rays as a function of an external field was determined by finding the positions of the extreme points of transmittance characteristics, corresponding to integer multiples of π rad (as shown in Figs. 1 and 2 for the sixth interference fringe of the 5CB-PVCN20 cell). The errors of determining these extreme positions were rather less than 3 mV or 3 mT, mostly about 1 mV or 1 mT; they are equivalent to the relative errors of determining the effective birefringence up-to 5%, mostly up-to 1%.

Firstly, the computations were done to achieve approximately the best-possible fit of the characteristics, calculated separately for each equivalent flat-parallel cell, to the experimental ones. The best results were obtained by approximating the dependence of the boundary tilt angle on the torque density as constant functions (resulting from modelling $\Theta(T_b)$ as non-decreasing cubic splines) for each of the three studied systems (5CB-PVCN20, 5CB-PVCN10 and 5CB-NSE). Secondly, the characteristics were re-calculated for all fringes with the

TABLE 1 The Magnitudes of the Splay and Bend Elastic Constants, Anisotropy of Diamagnetic Susceptibility and Anchoring Angle Determined as the Solutions of the Inverse Problems for the 5CP-PVCN20 Equivalent Cells after Magnetic Characteristics, by Minimising the Similarity Functional to the Least-Possible Value. The Last Column Displays the Values of this Functional Obtained for Characteristics Simulated with the Averaged Magnitudes of the Material: $K_{11} = 6.33\,\mathrm{pN}$, $K_{33} = 8.59\,\mathrm{pN}$, $\chi_a = 1.266 \cdot 10^{-6}$, $\vartheta_0 = 10\,\mathrm{mrad}$

j	d [μm]	K_{11} [pN]	K_{33} [pN]	χ_a [$\times 10^6$]	ϑ_0 [mrad]	$S(p)$	$S(\bar{p})$
4	13.9	5.98	14.00	1.41	5.6	0.007	0.049
5	17.4	6.12	12.59	1.41	15.6	0.020	0.043
6	20.8	6.21	11.06	1.38	15.0	0.021	0.035
7	24.3	6.27	10.18	1.37	12.4	0.019	0.032
8	27.7	6.47	8.91	1.30	8.1	0.015	0.015
9	31.1	6.44	8.57	1.30	6.5	0.013	0.014
10	34.5	6.43	8.57	1.30	6.2	0.012	0.013
11	38.2	6.75	8.56	1.35	4.8	0.013	0.016
12	41.6	6.42	8.63	1.29	3.6	0.010	0.013
13	45.1	6.51	7.59	1.24	5.5	0.019	0.028
14	48.6	6.42	8.56	1.30	2.5	0.009	0.011
15	52.0	6.43	8.52	1.30	3.0	0.008	0.011
16	55.5	6.07	7.73	1.20	2.6	0.010	0.015
17	59.0	6.51	7.83	1.26	3.5	0.010	0.019
18	62.4	6.53	7.83	1.26	3.3	0.008	0.019
19	65.9	6.57	7.45	1.23	4.3	0.011	0.026
20	69.4	6.71	6.53	1.19	5.7	0.017	0.041

same values of the nematics parameters K_{11}, K_{33}, χ_a and the three functions characterising the nematics-substrate coupling, determined as the means from the results of the preliminary computations; these characteristics were compared ones again to the experimental ones. The material constants of 5CB (at 22.7°C) were following: $\varepsilon_{||} = 19.04$, $\varepsilon_{\perp} = 6.50$, $n_o = 1.5383$, $n_e = 1.7206$ determined from the previous separate measurements [1,2] and $K_{11} = 6.33$ pN, $K_{33} = 8.59$ pN, $\chi_a = 1.266 \cdot 10^{-6}$ obtained as the averages from the preliminary solutions of the inverse problems. The anchoring angles ϑ_0 were estimated as the averages from approximations obtained in preliminary computations, characterising the three cases of very strong anchoring with value ϑ_0 equal to 10 mrad for 5CB-PVCN20, 10 mrad for 5CB-PVCN10 and 11 mrad for 5CB-NSE. The results of computations for the 5CB-PVCN20 cell are shown in Tables 1 and 2 and also in Figures 1 and 2, for the 5CB-PVCN10 cell if Table 3 and for the 5CB-NSE cell in Table 4. The equivalent cell thickness was determined by using the birefringence $n_a \equiv n_e - n_o$, what produced error

TABLE 2 The Magnitudes of the Splay and Bend Elastic Constants and Anchoring Angle Determined as the Solutions of the Inverse Problems for the 5CP-PVCN20 Equivalent Cells after Electric Characteristics by Minimising the Similarity Functional to the Least-Possible Value. The Last Column Displays the Values of this Functional Obtained for Characteristics Simulated with the Averaged Magnitudes of the Material Parameters: $K_{11} = 6.33$ pN, $K_{33} = 8.59$ pN, $\vartheta_0 = 10$ mrad

j	d [μm]	K_{11} [pN]	K_{33} [pN]	ϑ_0 [mrad]	$S(p)$	$S(\bar{p})$
4	13.9	6.04	11.13	2.1	0.011	0.055
5	17.4	5.98	10.40	7.1	0.012	0.035
6	20.8	6.12	9.94	8.6	0.014	0.032
7	24.3	6.19	9.57	9.7	0.016	0.026
8	27.7	6.35	9.07	9.9	0.017	0.024
9	31.2	6.34	9.09	10.7	0.019	0.024
10	34.7	6.29	8.80	9.2	0.011	0.012
11	38.2	6.33	8.59	10.5	0.011	0.011
12	41.7	6.37	8.39	13.1	0.013	0.016
13	45.1	6.76	7.95	12.3	0.027	0.031
14	48.6	6.37	8.31	13.2	0.013	0.018
15	52.6	6.37	8.26	14.3	0.008	0.020
16	55.6	6.28	8.42	13.2	0.012	0.019
17	59.0	6.28	8.40	12.8	0.013	0.019
18	62.4	6.37	8.25	13.7	0.012	0.018
19	65.9	6.34	8.25	15.2	0.012	0.022
20	69.4	6.36	8.23	16.6	0.012	0.023

TABLE 3 The Magnitudes of the Splay and Bend Elastic Constants and Anchoring Angle Determined as the Solutions of the Inverse Problems for the 5CP-PVCN10 Equivalent Cells after Electric Characteristics by Minimising the Similarity Functional to the Least-Possible Value. The Last Column Displays the Values of this Functional Obtained for Characteristics Simulated with the Averaged Magnitudes of the Material Parameters: $K_{11} = 6.33\,\mathrm{pN}$, $K_{33} = 8.59\,\mathrm{pN}$, $\vartheta_0 = 10\,\mathrm{mrad}$

j	d [μm]	K_{11} [pN]	K_{33} [pN]	ϑ_0 [mrad]	$S(p)$	$S(\bar{p})$
6	20.8	6.30	8.78	5.8	0.009	0.013
8	27.8	6.24	8.78	9.7	0.011	0.012
9	31.2	6.23	8.69	11.9	0.012	0.015
10	34.7	6.33	8.66	11.3	0.015	0.015
11	38.2	6.28	8.56	11.4	0.011	0.016
12	41.7	6.44	8.41	14.3	0.014	0.014
13	45.1	6.44	8.33	13.9	0.009	0.019

since the anchoring angle was greater than zero, but for very small ϑ_0 (like in the analysed cells) this error was negligible. The precise estimation of errors in the magnitudes of the nematics parameters is difficult. Comparing results of intermediate measurements, computations and simulations, one can expect the relative errors of these magnitudes smaller than the following: 0.001 in n_o, n_e; 0.01 in $d, \varepsilon_\perp, \varepsilon_{||}$; 0.03 in K_{11}; 0.05 in K_{33} and 0.1 in χ_a, ϑ_0.

The results of computing the material parameters for 4th and 5th interference fringes are less reliable due to small number of extreme points of the transmittance characteristics which can be taken into account. The values of the similarity functional $S(p)$ corresponding

TABLE 4 The Magnitudes of the Splay and Bend Elastic Constants and Anchoring Angle Determined as the Solutions of the Inverse Problems for the 5CP-NSE Equivalent Cells After Electric Characteristics by Minimising the Similarity Functional to the Least-Possible Value. The Last Column Displays the Values of this Functional Obtained for Characteristics Simulated with the Averaged Magnitudes of the Material Parameters: $K_{11} = 6.33\,\mathrm{pN}$, $K_{33} = 8.59\,\mathrm{pN}$, $\vartheta_0 = 11\,\mathrm{mrad}$

j	d [μm]	K_{11} [pN]	K_{33} [pN]	ϑ_0 [mrad]	$S(p)$	$S(\bar{p})$
5	17.3	6.33	8.84	9.5	0.005	0.011
7	24.3	6.38	8.70	9.3	0.008	0.012
8	27.7	6.44	8.43	14.7	0.007	0.008
10	34.7	6.50	8.18	14.6	0.011	0.013
11	38.2	6.53	8.10	13.8	0.012	0.014

to the averaged values of material parameters $K_{11}, K_{33}, \chi_a, \vartheta_0$, presented in the last column of each table, are at the level of the averaged relative error of determining the light phase shift, between 0.01 and 0.03. It means that with the accuracy, which could be achieved in the experiments presented in this work, the dependence of the magnitudes of anisotropy of diamagnetic susceptibility, splay and bend elastic constants and anchoring angle on cell thickness is not observed. Moreover it follows that the mathematical model presented above describes well the analysed phenomena.

CONCLUSION

A wedge cell of planar nematics alignment enables measurements of optical response (i.e.,the transmittance and the effective birefringence) to external magnetic or electric field, substituting a set of flat-parallel cells of different thicknesses and with the same cover surface treatment. The thickness of each equivalent flat cell may be precisely determined irrespectively of possible bend of cell covers, what makes the measurements be more precise.

The method (with the mathematical model) presented above makes possible the quantitative determination of the characteristics of nematics-substrate interaction simultaneously with the determination of the nematics constants (i.e., the unknown magnitudes of bulk material parameters), both in the case of weak anchoring and in the case of strong anchoring. For obtaining the reliable results it is necessary to take into account few different characteristics measured in a wide range of voltage or magnetic field magnitudes, enforcing stationary states of a nematics inside a cell.

The dependence of the material constants and anchoring angle on the cell thickness is evidently apparent, due to instability typical for inverse problems. The characteristics computed with the averaged magnitudes of the nematics parameters and anchoring angle fit well the experimental data, with the error at the acceptable level estimated after the errors in both the experimental data and the preliminary computations. In general, the best fit of simulated characteristics to measured ones does not correspond to the reliable solution of the inverse problem; the minimisation of the similarity functional must be stopped when its value achieves a specific level, corresponding to the errors in experimental data. The same instability, as the intrinsic feature of this inverse problem, is dealt with irrespectively of the method for determining the nematics-substrate coupling characteristics. The use of a wedge cell makes it possible to overcome the influence of these spurious effects, as it was described above.

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