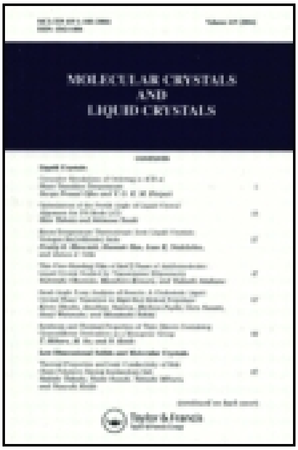

Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

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Version of record first published: 17 Oct 2011.

To cite this article: Francesco Simoni , Roberto Bartolino , Giovanni Barbero , Patrizia Aiello & Barbara Valenti (1984): Field Induced Molecular Reorientation in the Hybrid Nematic Cell, Molecular Crystals and Liquid Crystals, 113:1, 303-319

To link to this article: http://dx.doi.org/10.1080/00268948408071691

Mol. Cryst. Liq. Cryst., 1984, Vol. 113, pp. 303-319
0026-8941/84/1134-0303/$20.00/0

Printed in the United States of America

FIELD INDUCED MOLECULAR REORIENTATION IN THE HYBRID NEMATIC CELL

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Abstract In this paper we review a general theoretical treatment of the interaction between a static or optical electric field and the molecular director of a hybrid aligned nematic liquid crystal. Preliminary experimental data are reported in the two cases for the first time.

INTRODUCTION

The interaction of a nematic liquid crystal with an electric or a magnetic field has been a subject of study for a long time[1]. Application of a weak field can induce a molecular reorientation because of the dielectric and magnetic anisotropy: and these effects are enhanced in monocrystals of nematic samples, i.e. in uniformly oriented samples. In fact the applied field induces a torque on the molecules depending on the reciprocal orientation between the external field and the local director. A distortion of the nematic structure is induced until the elastic torque balances the externally imposed torque. If the time dependent behaviour is considered one should also take into account the viscous torque, but in the following we will consider only steady state conditions.

The effects of the interaction of static external fields with a uniformely oriented nematic slab (homeotropic or planar) was studied in the early 30's by V. Fredericksz[2] and the more used treatment of the problem was developed by A. Saupe[3]. A review of these effects is reported in ref. 4.

A typical feature of this interaction in homeotropic samples is that molecular reorientation occurs for field values higher than a critical one. At this threshold we have a transition to the distorted structure very similar to a second order phase transition[5]. It can be studied using a Landau-De Gennes expansion of the free energy using as parameter the maximum deviation of the molecules from the umperturbed state.

Most of the studies on these so called Fredericksz transitions have been made for static or low frequency fields, but recently the optically induced Fredericksz transition has been discovered[6] and many different nonlinear optical effects have been observed in nematic liquid crystals[7]. A wide class of fundamental features are present in these phenomena. Besides that, both static and optical effects are very interesting for applications. The electrooptical behaviour (EBC effect) is used in liquid crystal displays[8], while the nonlinear optical response seems promising to be used in bistable devices and phase conjugators[9].

Recently new interest in this field has arisen because of the intensive study of a particular geometrical configuration of a nematic cell: the hybrid aligned nematic cell (HAN), where even in the unperturbed state the molecular alignement is not uniform, but continuosly deformed. In this case the liquid crystal has two different orientations imposed on the boundaries, homeotropic on one and planar on the other side: in this way a uniform splay+bend deformation is imposed. New fundamental features are observed in the HAN cell. The flexoelectric effect[10] not dependent on the molecular shape[11] but only on the molecular alignement has been discovered.

Moreover the HAN cell has no threshold for molecular reorientation[12] due to either static or optical field and the nonlinear optical properties of a nematic liquid crystal are enhanced by this configuration[13] . On the other hand this alignement is strongly dependent on the surfaces conditions[14] and it seems suitable to study the effects of surface anchoring on a liquid crystal[15].

In this paper we review a general theoretical treatment of the interaction between electric field (static or optical) and molecular director for the HAN cell and we report the first preliminary experimental data obtained in the two cases.

GENERAL THEORY OF THE HAN CELL

The hybrid aligned nematic cell is depicted in fig. 1. It is characterized by a different alignement on the two boundary walls. We choose the homeotropic alignement on the first surface ($z = -d/2$) and the planar one on the second surface ($z = d/2$). The $\hat{z}$ direction is chosen normal to the cell boundaries and $z = 0$ at the midpoint of the sample. The orientation of the molecular director $\hat{n}$ is given by the angle ϕ between $\hat{n}$ and $\hat{z}$.

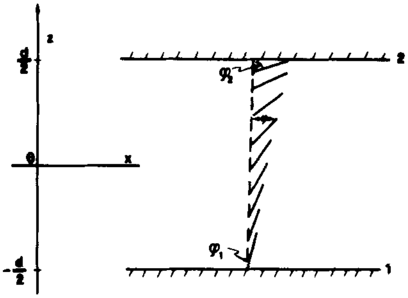

FIG. 1

The total free energy for unit surface area can be written as

$$\tilde{F} = \int_{-d/2}^{d/2} F(\phi,\ \phi_z)dz + F_s(\phi) \qquad (1)$$

where $F(\phi,\phi_z)$ is the bulk free energy density and $F_s(\phi)$ is the surface free energy density due to the anchoring on the walls; $\phi_z = \frac{\partial\phi}{\partial z}$. We apply to (1) a second order variational expansion:

$$\tilde{F}(\phi+\eta) = \tilde{F}(\phi) + \delta\tilde{F}(\phi;\eta) + \delta^2\tilde{F}(\phi;\eta) \qquad (2)$$

where $\phi' = \phi + \eta$ is a new function infinitely close to ϕ. Then from the minimum conditions $\delta\tilde{F} = 0$, $\delta^2\tilde{F} > 0$, we get the well known Euler-Lagrange equations and the boundary conditions:

$$\frac{\partial F}{\partial \phi} - \frac{d}{dz}\frac{\partial F}{\partial \phi_z} = 0 \qquad -\frac{d}{2} \leqq z \leqq \frac{d}{2} \qquad (3)$$

$$\frac{\partial F}{\partial \phi_z} + \frac{\partial F_S}{\partial \phi} = 0 \qquad z = \frac{d}{2} \qquad (4a)$$

$$\frac{\partial F}{\partial \phi_z} - \frac{\partial F_S}{\partial \phi} = 0 \qquad z = -\frac{d}{2} \qquad (4b)$$

Now we have to work out $F(\phi,\phi_z)$ and $F_S(\phi)$ for the HAN cell in order to find out the stable configurations under an applied electric field.

From the continuum theory we have the general expression of the free energy density

$$F = \frac{1}{2} K_{11}(\mathrm{div}\ \hat{n})^2 + K_{22}(\hat{n}\cdot\mathrm{rot}\ \hat{n})^2 + K_{33}(\hat{n}\wedge\mathrm{rot}\ \hat{n})^2$$

$$+ F_E + F_S \qquad (5)$$

where K_{ii} $(i = 1,2,3)$ are the Frank's elastic constant and $\hat{n} = \hat{z}\cos\phi + \hat{x}\sin\phi$ the molecular director. F_E is the free energy density due to interaction with the field, which will be specified later on for the static and the optical case; F_S is the surface interaction term and it will be discussed later.

Interaction with optical field

Let us consider a light wave at normal incidence on the sample, with $\overline{E} \equiv (E_x,0,E_z)$ and $\overline{H} \equiv (0,H_y,0)$. We can keep the plane wave approximation if the beam diameter is much larger than the sample thickness. In this case we have also $\frac{\partial}{\partial x} = \frac{\partial}{\partial y} = 0$ for any component of the field.

In the optical case the molecular reorientation is due only to the torque applied by the electric field, but in the free energy density we have a contribution from the magnetic field as well. We will follow the procedure of Ong[16] which overcomes the problems of the correct application of eq.(3) to this case, but we suggest the reading of a paper by Zel'dovich[17] devoting a long discussion to it.

If $\overline{k}$ is the wave vector of the light the electric and magnetic contributions to the free energy density are

$$F_e = F_m = \frac{\overline{S}\cdot\overline{k}}{2\omega} \tag{6}$$

therefore in this case

$$F_E = \frac{\overline{S}\cdot\overline{n}}{c} = \frac{S_z n(z)}{c} \tag{7}$$

where $\overline{S}$ is the Pointing vector and

$$|n| = \frac{\sqrt{\varepsilon_{\perp}\varepsilon_{\parallel}}}{(\varepsilon_{\perp}\sin^2\phi + \varepsilon_{\parallel}\sin^2\phi)^{1/2}} \tag{8}$$

$\varepsilon_{\parallel}$ and ε are the dielectric constants parallel and perpendicular to the molecular director. Eq.(8) is very convenient to use because it can be shown that $\overline{S}$ is not uniform in the sample, but S_z is constant, then in eq.(8) only the actual refraction index n is dependent on z. Moreover because of this property we can identify S_z with the intensity I of the wave impinging on the liquid crystal sample, then

$$F_E = \frac{In(z)}{c} \tag{9}$$

which after minimization gives a term

$$\frac{\partial F_E}{\partial\phi} = \frac{\Delta\varepsilon\sqrt{\varepsilon_{\parallel}\varepsilon_{\perp}}}{c}\, I\, \frac{\sin\phi\cos\phi}{(\varepsilon_{\perp} + \Delta\varepsilon\,\cos^2\phi)^{3/2}} \tag{10}$$

with $\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$.

Interaction with static electric field

We set a static voltage applied to the boundaries, inside the sample the electric field is not uniform but it can be considered to keep an orientation normal to the boundaries, i.e. $E \equiv E(z)\cdot\hat{z}$. The free energy term is well known in this case:

$$F_E = -\frac{\Delta\varepsilon}{8\pi}(\hat{n}\cdot E)^2 - \frac{\varepsilon_\perp}{8\pi}|E|^2 \tag{11}$$

According to Landau the forces acting on the molecular director can be obtained using eq.(3) keeping E as constant. This is because eq.(3) is locally valid and at any fixed z, E has a well defined value. Therefore after minimization we get

$$\frac{\partial F_E}{\partial\phi} = -\frac{\Delta\varepsilon}{4\pi}E^2(z)\sin\phi\cos\phi \tag{12}$$

From Maxwell equations we have div $\bar{D} = 0$, then $D_z = \text{const}$ and

$$E(z) = \frac{D_z}{\varepsilon_\perp + \Delta\varepsilon\cos^2\phi(z)} \tag{13}$$

This way the director-field interaction will lead to

$$\frac{\partial F_E}{\partial\phi} = -\frac{\Delta\varepsilon}{4\pi}D_z^2\frac{\sin\phi(z)\cos\phi(z)}{|\varepsilon_\perp + \Delta\varepsilon\cos^2\phi(z)|^2} \tag{14}$$

In order to relate the constant D_z to the actual applied voltage we need to use the relationship $V = D_z\int\frac{dz}{\varepsilon_\perp + \Delta\varepsilon\cos^2\phi}$

Surface interaction

The director-surface interaction included in F_S has to be considered in order to distinguish between different kinds of boundary orientations. For the hybrid alignement we take the following expression

$$F_s = \frac{1}{2} W_1 \sin^2\phi_1 + \frac{1}{2} W_2 \cos^2\phi_2 \qquad (15)$$

W_1 and W_2 are the anchoring energies on the surfaces. From (15) we get the boundary conditions necessary to calculate ϕ_1 and ϕ_2, applying eq.(4a) and (4b):

$$\left(\frac{\partial F_K}{\partial \phi_z}\right)_i + \left(\frac{\partial F_E}{\partial \phi_z}\right)_i - W_i \sin\phi_i \cos\phi_i = 0 \qquad (16)$$

where i = 1 at z = - d/2, i = 2 at z = d/2. Of course eq. (16) is necessary to know the actual director orientation in the sample only when finite anchoring energy is considered, otherwise $\phi_1 = 0$, $\phi_2 = \pi/2$.

Euler Equation and solution

We are now able to write the Euler equation that we need to calculate the director distribution along the sample, that is $\phi(z)$. Eq.(3) gives:

$$(1 - K\sin^2\phi)\frac{d^2\phi}{dz^2} - K\sin\phi\cos\phi\left(\frac{d\phi}{dz}\right)^2 + \frac{\tilde{E}}{K_{33}} = 0 \qquad (17)$$

where $K = (K_{33} - K_{11})/K_{33}$.

$\tilde{E}$ is given by eq.(10) in the optical case and eq.(14) in the static case. In the following we will distinguish between them by subscript o and s.

A first integration of (17) gives:

$$(1 - K\sin\phi)\phi_z^2 + f_{o,s}(\phi) = c^2 \qquad (18)$$

with

$$f_o(\phi) = \frac{2I}{cK_{33}} \frac{1}{\left(1 + \frac{\Delta\varepsilon}{\varepsilon_\perp}\cos^2\phi\right)^{\frac{1}{2}}}$$

$$f_s(\phi) = \frac{D_z}{4\pi K_{33}} \frac{1}{(\varepsilon_\perp + \Delta\varepsilon\cos^2\phi)}$$

and c^2 is an integration constant to be determined by the boundary conditions (16) which become

$$2\,\frac{K_{33}}{W_i}\left|(1 - K\sin^2\phi_i)(c^2 - f_{o,s}(\phi))\right|^{\frac{1}{2}} = \sin 2\phi_i \qquad (19)$$

(i = 1) at $z = -\,d/2$, $i = 2$ at $z = d/2$.

By integration of (18) we get

$$\int_{\phi_1}^{\phi_2} \left|\frac{1 - K\sin^2\phi}{c^2 - f_{o,s}(\phi)}\right|^{\frac{1}{2}} d\phi = d \qquad (20)$$

Through eqs.(19) and (20) we calculate ϕ_1, ϕ_2 and c^2. It is then possible to obtain the tilt angle distribution as a function of the applied field from the integral equation:

$$\int_{\phi_1}^{\phi(z)} \left|\frac{1 - K\sin^2\phi'}{c^2 - f_{o,s}(\phi')}\right|^{\frac{1}{2}} d\phi' = z + \frac{d}{2} \qquad (21)$$

Eq.(21) is the final result of the theoretical approach, from that we calculate $\phi(z)$ for the possible different conditions. The function $\phi(z)$ at each applied field is necessary to compute the expected value of the measured quantity as we shall see in the next section.

PRELIMINARY EXPERIMENTAL DATA

The optical methods give the more direct way to study the described phenomena of molecular reorientation because the index of refraction of the extraordinary ray travelling in the sample is strictly dependent on the director spatial distribution. Therefore the physical parameter to be measured to get information about it is the induced phase variation on the extraordinary ray by the liquid crystal sample. For normal incidence it is given by

$$\delta = \frac{2\pi}{\lambda} \int_{-d/2}^{d/2} (n(\phi) - n_o)dz \qquad (22)$$

The usual way to measure δ is to detect the trasmitted light intensity when the sample is placed between crossed polarizers. For each value of the applied field the different director distribution will lead to a different value of δ. For example in the static case with $\Delta\varepsilon < 0$ we would expect an increase of the phase shift towards the maximum value given by the planar texture, while in the case with $\Delta\varepsilon > 0$ the phase shift approaches zero as the field is increased, because for the homeotropic texture $\delta = 0$.

In fig. 2 we report the function $\delta(E)$ calculated for the optical induced reorientation in a nematic sample of MBBA of 10mμ.

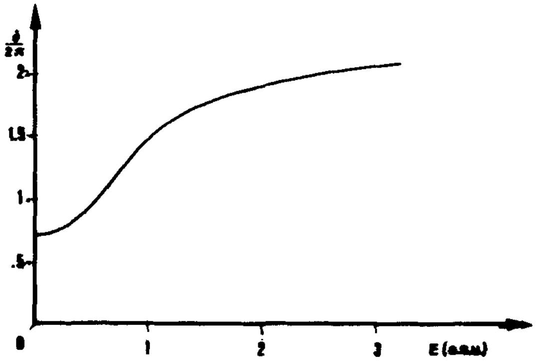

FIG. 2

By studying the interaction of a laser field with the nematic structure it is possible to use the nonlinear optical effects either to measure δ in a different way or to measure the nonlinear dielectric constant which is dependent on the molecular orientation as well.

The effects of self-phase modulation allows to make a direct measurement of δ just counting the number of interference rings N appearing in the beam waist after the passage through the sample. This measurement has the advantage to be straightforward even if it is not very sensitive to small variations, because $\delta = 2\pi N$ and only variation of 2π can be observed. A direct and more precise detection can be done on the nonlinear dielectric constant[13]:

$$<\delta\varepsilon> = \frac{1}{d} \int_{-d/2}^{d/2} \left\{ \frac{\Delta\varepsilon}{2} |\varepsilon^{2} - \left(\frac{\Delta\varepsilon}{2}\right)^{2} | \cdot \frac{\cos 2\phi(z,o) - \cos 2\phi(z,E)}{\left|\varepsilon + \frac{\Delta\varepsilon}{2}\cos 2\phi(z,E)\right| \left|\varepsilon + \frac{\Delta\varepsilon}{2}\cos 2\phi(z,o)\right|} \right\} dz$$

$<\delta\varepsilon>$ can be measured through the self-focussing effect arising in the crystal measuring the beam diameter on a screen far from the sample. In this way it is possible to have the inverse focal lenght of the nonlinear lens equivalent to the liquid crystal at different laser intensities, which is proportional to the nonlinear dielectric constant. The absolute value of it can be calculated knowing the geometry of the experiment. In the following we report preliminary data taken on hybrid aligned sample of MBBA at room temperature.

The homeotropic orientation was obtained by coating the optical glass with a Silane polymer (ODS by Kisso Corp). The planar alignement was obtained by dipping the glass in a 0.25% solution of FORMVAR in dichloroetane. Using this treatment a strong anchoring on the walls can be assumed.

Interaction with optical field

One of the most interesting features of the HAN cell is the expected absence of threshold for the field induced molecular reorientation.

Such a behavior is demonstrated in fig. 3 where the inverse focal length of the liquid crystal is reported vs the laser intensity.

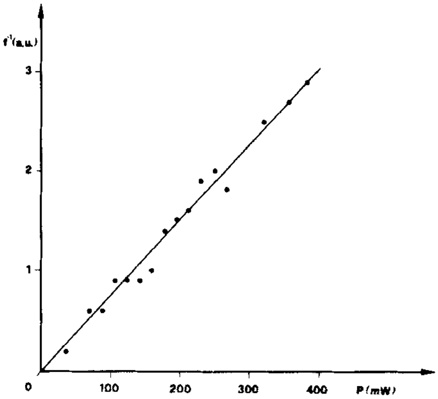

FIG. 3

From this figure it is evident that no threshold exists for the onset of the nonlinear optical effect, that is for the optical induced reorientation. Moreover it is confirmed the expected[13] linear dependence on the intensity in the low excitation region. This feature of the HAN cell is very important for possible applications if we compare it to the homeotropic cell, where we always need to reach the Fredericksz's threshold to induce the molecular rearientation.

The measurements of the induced phase shift can be a test of the presented theory. A first qualitative check of it has been done using the effect of self phase modulation to measure δ. In fig. 4 the number of observed rings is reported vs the laser intensity for a sample thickness of 70μm. In this case the experimental geometry is slightly different because the sample is placed in the focal waist of a lens (f = 25cm), while in the former case the sample was placed 4 cm before it, thus the measured laser power corresponds to different local intensities.

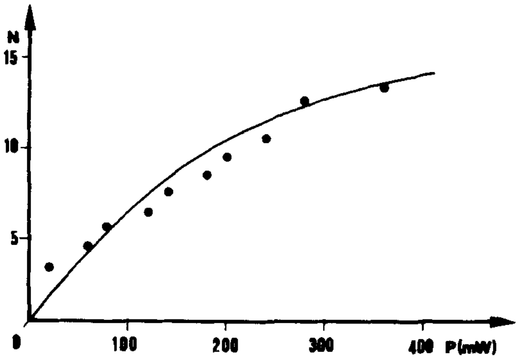

FIG. 4

The continuous line is derived from eq.(22), matching the maximum number of observed rings with the saturation value of δ. The agreement is very good if one considers the low resolution of this measurement and the difficulty to count the rings in the central bright spot of the beam.

More sensitive measurements of phase shift, using a second weak laser beam as probe are under way.

Interaction with static field

In fig. 5 we report a typical transmission curve at different voltages for a 10μm sample placed between crossed polarizers. In this case the electric field is normal to the

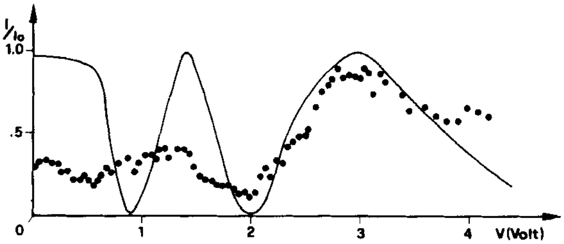

FIG. 5

glass walls, but the low frequency dielectric anisotropy is negative thus the reorientational effect is similar to that observed in the previous optical case and the expected final configuration is almost planar.

Also in this case of course no threshold is necessary to induce variation in the transmitted light, while in a homeotropic sample of the same thickness the Fredericksz transition occurs at 3.2 volts. We observe in Fig. 5 an oscillatory behaviour before a saturation where hydrodinamical instabilities start, at V = 4.2 volts, much before than in the corresponding homogeneous cell (either planar or homeotropic). The continuous lines have been calculated from eq.(22) for an ideal hybrid cell in the strong anchoring approximation and using the relation $I = I_o \sin^2 \frac{\delta}{2}$ where I_o is the intensity of the incident light.

Three maxima are expected before saturation as observed in the experiment and using as parameters the position of the third maximum we find good agreement with the experimental data for the position of the second and third peak and the minimum between them.

These data suggest that the d.c. case is qualitatively different from the optical case in such a way that a parallel formulation of the two problems may not be adeguate. The first correction to be introduced in this case should be due to the flexoelectric effect.

We should consider the polarization induced by the molecular deformation:

$$\overline{P} = e_1 \hat{n} \cdot (\mathrm{div}\ \hat{n}) - e_3 (\mathrm{rot}\ \hat{n}) \wedge \hat{n}$$

where e_i are the flexoelectric constants.

In eq. (11) we would have an additional term

$$F_{flex} = - \int_o^d \overline{P_z} \cdot \overline{E}(z)\, dz$$

linearly dependent on the electric field.

A quantitative analysis is under way including also the flexoelectric term in the free energy density.

CONCLUSIONS

We have presented the theory of field induced molecular reorientation in a hybrid aligned cell necessary to calculate the director orientation inside a sample when interaction with an optical or static electric field is considered.

Then we have reported the preliminary experimental data in both cases and we have shown a good qualitative agreement with the theoretical previsions. Detailed experimental data and more quantitative analysis of them will be reported elsewhere.

About the possible flexoelectric contribution in the d.c. case we greatly appreciate the fruitful discussion with G. Durand during the Bovec Conference.

Aknowledgements are due to the technical aid by G. Perri, C. Prete, B. De Nardo.

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