Diffraction in smectic C and nematic with short range smectic C order for oblique incidence of coherent laser light♣

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We describe the application of a laser diffraction technique to the study of electroconvection (EC) in smectics C and nematics with short range smectic C order for oblique illumination with coherent laser light. We have used laser diffraction as a method for the direct quantitative determination of the amplitudes of the director field. We found that oblique incidence in general favours the reflexes of odd numbered order, which indicate the amplitude grating, produced by the director distortion in EC regime. The modification of the standard Carr-Helfrich mechanism, provoked by the diffraction study of the smectic C and nematic with short range smectic C order, was discussed.

1. Introduction

Electroconvection (EC) in homogeneously aligned nematic liquid crystal (NLC) layers, when excited by a *dc* or *ac* voltage, exhibits a simple roll pattern with a wave vector along (normal rolls -NRs) or inclined (oblique rolls - ORs) to the preferential alignment of the director *n* of the system [1-5]. The EC in smectic C liquid crystals (S_C) was studied both experimentally and theoretically in [6,7]. Two EC instabilities were found: "initial" rolls (IRs) and "fundamental" (FRs) with wave vectors normal to or along the direction of *n*.

The most successful technique for the investigation of the EC patterns is the shadowgraph method, but it fails to provide quantitative access to the deflection amplitudes of the director field.

On the other hand, illumination of the distorted EC layer with laser light gives rise to a grating effect, where a number of fringes are observed [8]. The determination of the intensities of these fringes has been carried out by many authors [8-13]. In [9-13], it was indicated that under oblique illumination, the fringes are sufficiently intense, even when the distortion angle is small (very close to the EC threshold).

The goal of the present work is to verify this fundamental result, found in the NLC, for the S_C and N with S_C short range order.

2. Experimental result and discussion

A sketch of the experimental set up is indicated in [14]. We used a low power He-Ne laser (≈1mW). The liquid crystal cell (LCC) was mounted in a Linkam microscope hot stage TMS 600. A photodiode or alternatively a diffusely reflecting screen for 2D camera images, at a distance *l* of approximately 600mm from the LCC, were used for the diffraction analysis. The photodiode aperture was 1mm × 1mm. A circular aperture, 0.5mm in diameter, defined the illumination spot.

We used the LC 4,n-heptyloxybenzoic acids (7OBA) (see [14]).

Figs. 1a,c indicate the FRs and the N with S_C short range order cell rolls (NCRs). The corresponding far-field diffractions are indicated in the same figure (b,d).

We assume a homogeneous, aligned along the *x* axis, LC layer of thickness *d*. The layer was confined between two planes. parallel to the electrodes (*xy)* at *z*=0 and *z*=*d*. When an electrical field *E*=*U*/*d* was applied in the z direction, the conductivity anisotropy of the material, in combination with small fluctuation modes of the *n* tilt, lead to lateral charge separation in the *x* direction and a

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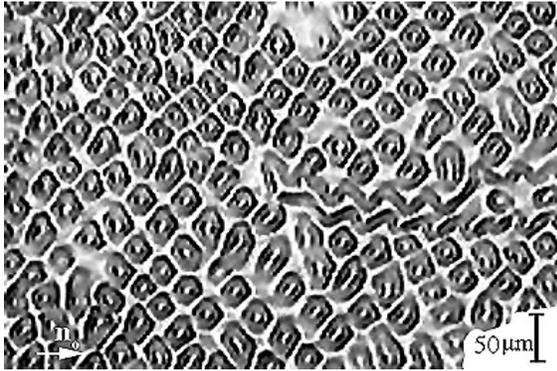
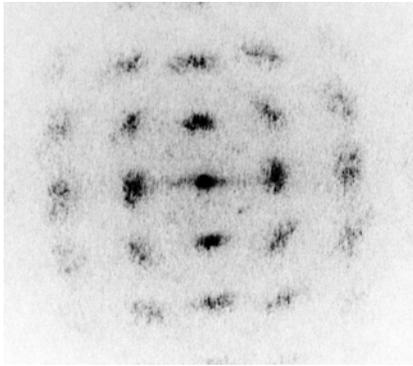periodically modulated flow field, which in turn couples to the **n** field.
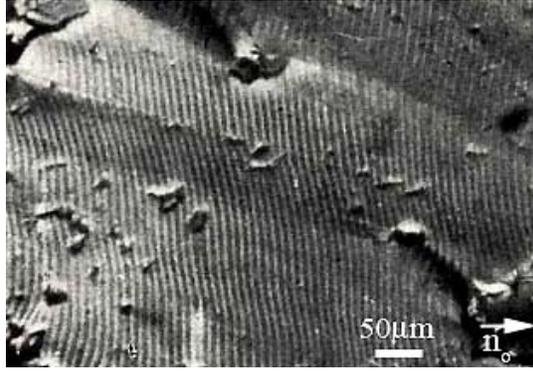
a

b

c

d

Fig. 1. a) The FRs EC structure and b) the corresponding diffraction at α=0° (normal illumination), U=28V,

f=50Hz, d=20μm, T=95°C; c) The NCR's EC structure and d) the corresponding diffraction at α=0°, U=34V, f=500Hz, d=8μm, T=142°C.

Thus, the essential variables describing the EC structures are the spatially modulated charge distribution q(x,y,z) and the director tilt angle θ(x,y,z). At the critical field E_{th}, stabilizing elastic and dielectric torques on **n** are overcome by destabilizing hydrodynamic torques. The equations describing the EC mechanism yield two dynamic regimes: conduction and dielectric [1-5]. This is the Carr-Helfrich (CH) mechanism [1]. The modification of this mechanism for S_C was described in [6,7].

The linear stability analysis of the torque balance in conventional Ns gives, for the **n** deflection: $\theta(x,z)=\theta_t\cos(k_x x)\cos(k_z z)$, where k_x is the periodicity of the pattern. In an *ac* field, q and θ have qualitatively different dynamic behaviours. However, for the diffraction experiment, only the **n** modulation is relevant and accessible.

We will present briefly the main formulae for the diffractions under normal and oblique illumination in the N state, to analyse the experimental results for FRs and NCRs.

For normal illumination (laser beam incidence at an angle α=0), it was found that the displacement of the rays after propagating through the LCC and the conservation of energy gives the intensity *I* in terms of the incident intensity I_o, and the amplitude of the light ($E^2\sim I$) in the exit plane, and leads to the dependence [12]:

$$\frac{I}{I_o}=\frac{E^2(x)}{E_o^2}=\frac{1}{1+2\dfrac{k_x(n_e^2-n_o^2)}{k_z n_o^2}\theta_t\sin k_x x}, \quad (1)$$

where n_o, n_e are the ordinary and extraordinary refractive indices. In the case of small θ, the differences between the exit point and entry point of the light can be neglected, and the periodicity of the phase grating can be written as $\Delta\phi(x)=\Delta\phi_{max}\cos(2k_x x)$. This is twice that of the director field θ(x), and therefore one expects diffraction reflexes from the phase grating to appear only at even orders *m*. On the other hand, the periodicity of the amplitude grating is the same as that of the **n** field, and it contributes also to odd order reflexes. With the assumption E(x)=const., the fringe intensities at angles ψ_m are described by Bessel functions $J_{m/2}$ with the amplitude of the laser light phase modulation in the argument (see[10-12])

$$\frac{I_m(\theta_t)}{I_o}=\frac{|E(\psi_m)|^2}{|E(\psi_o)|^2}=J^2_{m/2}(\Delta\phi_{max})\approx\frac{\Delta\phi^m_{max}}{2^m[(m/2)!]^2}. \quad (2)$$

Using the phase φ(x) in dependence on the amplitude of the **n** deflections

$$\Delta\phi(x_o)\approx-\frac{k_L n_e d(n_e^2-n_o^2)}{8n_o^2}\theta_t^2\cos 2k_x x_o \quad (3)$$

(x_o–the entering point, k_L laser light wave number) which is the result for straight transmission, and the result that the lateral phase difference along curved paths at the exit position for small θ_t is basically the same as that for this straight transmission, leads to the quantitative relation between the normalized diffraction intensity and the director tilt amplitude at the second order reflex [12]:

$$\frac{I_2(\theta_t)}{I_o} = \left(\frac{k_L n_e d(n_e^2 - n_o^2)}{16 n_o^2}\right)^2 \theta_t^4, \quad (I_2 \ll I_o). \qquad (4)$$

For oblique illumination under a tilt α, experiments [9-13] show that the diffraction profile has a strong dependence on this angle. As proved in [10,11], the intensity I_m of the m_{th} order diffraction fringe in this case is [11] $I_m = \beta_m[J_m(Q\theta_t)]^2$, where J_m is the Bessel function of the first order m. β_m and Q are quantities that depend on the refractive indices of the LC, the angle α and the geometry of the spatial distortion of layer. It is important to note that (i) - the coefficient β_m depends slightly on the order m of the fringe, as well as on the system parameters, but is independent of θ_t, and (ii) - the quantity Q depends only on the system parameters, being completely independent of the fringe order and θ_t. Furthermore, the quantity Q is proportional to the ratio d/λ.

The symmetry of the diffraction angles is broken in the case of oblique incidence ($\psi \leftrightarrow -\psi$). In addition to this symmetry breaking, an extra phase $\Delta\phi_e(\psi)$ appears. The condition allowing for this additional phase for constructive interference on the second even order spot ($m>0$, $\psi_m>0$) is [12]

$$\lambda_{ph}\sin(\psi_m+\alpha) = \Delta\psi_o + m2\pi/k_L, \qquad (5)$$

where $\psi_m = \arcsin[(2\pi m/\lambda_{ph}k_L) + \sin\alpha] - \alpha; \qquad (6)$

and for $\psi_m<0$ and $m<0$ is

$$\psi_m = \arcsin[(2\pi|m|/\lambda_{ph}k_L) - \sin\alpha] + \alpha, \qquad (7)$$

where λ_{ph} is the wave length of the phase modulation of exiting light.

Using Eq. (1-7), and assuming that the EC mechanism in S_C and N with short range S_C order differs from the standard CH mechanism only in the direction of the charge distribution (confined in the layers) and in the type of the *n* deflection relative to the *xz* plane, we present in Tables 1,2 the compact experimental results.

The Table 1 concerns the fundamental S_C domains, and indicates that for the normal illumination the even order fringe intensity dominates over the odd one and the deflection of *n* is maximal. Under oblique illumination, even a very small tilt angle provokes comparable even and odd order fringe intensities. The effect is more emphasized at $\alpha > \pm 10°$, where the odd order intensities dominate. It is very close to U_{th}. An intriguing result is the monotonic reduction of the wavelength of the phase modulation of the exiting light λ_{ph} with increasing α. For a primary beam at $\psi=0$, $\Delta\phi_e(\psi)$ vanishes. The symmetry breaking leads to a

slight shift of the diffraction spots, and more obviously, to a change of the intensities of the even and odd order spots.

Table 1. The FRs diffraction parameters for α and U variations. $d=20\mu m$, $f=50Hz$, $T=95°C$.

$\alpha,°$	0	2	-2	5	-5	10	-10
$I_1^{+1},\%$	0,5	0,5	0,7	0,3	0,6	0,3	0,7
$I_1^{-1},\%$	-	0,7	0,4	0,5	0,3	0,5	0,5
$I_2,\%$	0,9	0,8	0,9	0,9	1,1	0,6	0,4
U_{th}, V	24	21	20,5	21	20,5	21	20
U, V	28	26	26,5	25	25	23	22
$\theta_t^{+1},°$	29	12	14	4	5	3	5
$\theta_t^{-1},°$	-	14	11	5	4	4	4
$\theta_t^{+2},°$	35	15	16	7	7	5	4
$\lambda_{ph},\mu m$	-	7	7	6	6	4	4

Table 2. The NCRs diffraction parameters for α and U variations. $d=8\mu m$, $f=500Hz$, $T=142°C$. I_o represents the intensity at $\theta=0$.

$\alpha,°$	0	5	-5	10	-10	15	-15
I_2, %	0,8	0,8	1,7	1,2	1,3	1	1,7
U_{th}, V	29	29	27	29	27	29	27
U, V	34	32	30	31	29	31	29
$\theta_t^{+2},°$	42	20	29	14	15	9	11
$\lambda_{ph},\mu m$	-	11	11	7	7	5	5

The most obvious results are the quantitative changes of the diffraction intensities with the rotation angle α. Whereas the odd order maxima, which are mainly generated by the amplitude modulation, slightly vary with the tilt of the cell, the even order spot intensities show a noticeable decrease.

The differentiation between the even and odd orders, however, ceases to have any significance, because of the lack of symmetry compared to the normal incidence case. This suggests an equal importance in the contributions of the ray deviation and $\Delta\phi$ to the effect. One can note that at small deformation angles, the odd fringes have comparable to or even greater than the even fringe intensities.

For an obliquely illuminated LC grating for a LC layer with a weak distortion, where the fringes are very weak when the incidence is normal, one has spectacular grating effects by gradually increasing the angle of incidence. An important characteristic of the oblique incidence case is the asymmetry of the fringes, that is, the fringes with orders m and $-m$ have in general, different powers. We also found that for a sufficiently thick sample, the asymmetry is of lesser importance. Thus, an obliquely illuminated LC grating is a much more sensitive diffraction tool, because a small distortion angle is adequate for the formation of intense fringes.

Table 2 concerns the N with short range S_c order. We analyse this phase in the high-temperature region, where the smectic order fluctuation does not dominate [14]. The

more important effect now is that near to the threshold (smaller *n* distortions) the second order fringes are more noticeable at bigger cell tilts.

Therefore, we do not find exactly the same effect, both in FRs and NCRs, as that in the classical NRs, where the standard CH mechanism works, namely the odd order maxima exhibit a minimum in the non-tilted cell, whereas the even order spots show qualitatively opposite behaviour. We, however, found a more unifying characteristic: the difference between the cases of normal and oblique incidence is that the contribution of the deviation of the ray to the fringe power is also important. like that of the phase disturbance. This common characteristic leads mainly to the most important effect: for a LCC layer with a weak distortion (note that θ_t could be less than 3° for α bigger than 10°) the intense fringes are observed at gradually increasing angles of incidence. The small difference between the oblique illumination in classical NRs with that in FRs and NCRs indicates that the CH mechanism is important for the FR in S_C and in N with short range S_C order, but the modifications suggested in [6,7] must be more precise, specially in accounting for the charge distribution inside the layers.

3. Conclusions

We have used laser diffraction as a detection method for the direct quantitative determination of the amplitudes of the director field. An important aspect in the diffraction experiment is the strong dependence of the diffraction image on small deviations from normal laser beam incidence. The dependence of the diffraction pattern upon the angle of incidence of the laser beam was discussed. The basic characteristics of the effect, namely the much more intense fringes obtained from a weakly distorted layer, compared with those formed in the normal incidence case, the asymmetry of the fringes, and the equal importance of the even and odd-order fringes were discussed.

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