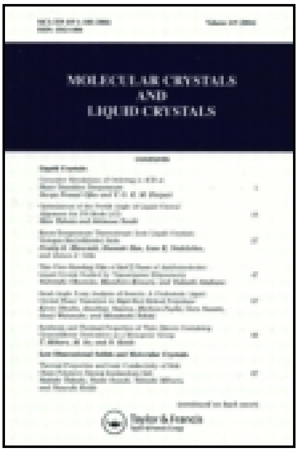

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Channelled Spectrum Due to Homeotropic Nematic Samples Undergoing Electric Field Induced Molecular Orientations

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Channelled Spectrum Due to Homeotropic Nematic Samples Undergoing Electric Field Induced Molecular Orientations

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We have observed the formation of the channelled spectrum in unpolarized light when a homeotropic nematic liquid crystal sample, undergoing molecular reorientations, is placed in a grating spectrometer. As the sample ceased to behave like a mono-crystal above the threshold of the Freedericksz transition due to the reorientational degeneracy of the molecules, domains are nucleated which are characterized by their optic axes pointing in different directions. The sample becomes infested with point singularities or umbilics and it can be considered to represent an ensemble of randomly distributed mono-crystal domains. This quasi-polycrystal model of the sample has been used to explain the interference in unpolarized light of the ordinary and extraordinary waves emerging from it. We give the theory of these interference bands in the spectrum of a white light source, produced by a spectrometer where no external polarizers were used around the sample. Experimental results will be discussed by taking into consideration the frequency and the visibility of the interference bands.

1. INTRODUCTION

In a certain type of interference experiments[1] involving white light illumination, one finds that every time the optical path difference becomes an integral multiple of a wavelength, there results an extinction of the corresponding wavelength. Thus, if a spectrometer with its entrance slit parallel to the direction of the fringes is used, the spectrum of the light source becomes channelled. These interference bands are equally spaced in spectroscopic wave number. While observing interference phenomena due to crystal plates, it is usually mandatory that the birefringent sample be placed between a polarizer and an analyser. The latter is used to render parallel the vibrations of the waves emerging from the plate in order to make them interfere. The nature of the resulting interference fringe pattern depends on the optical path difference between the ordinary and the extraordinary waves. The channelled spectrum is obtained by placing the birefringent plate surrounded by two crossed polarizers in the spectrometer as before.

In the course of our research on the tunable Talbot bands using electric field induced Freedericksz transitions in nematic liquid crystals,[2] we have observed the channelled spectrum like phenomenon in unpolarized light. A homeotropic nematic sample was subjected to an electric field and was examined in a grating spectrometer. Dark bands characteristic of the optical path difference due to the induced birefringence were seen in the absence of external polarizers. We have attempted to explain the phenomenon by assigning a quasi-polycrystal structure to the nematic sample above the threshold. The invariance of the optic axis produces a degeneracy in the reorientational behavior of the nematic molecules and this is dependent on the type and the initial alignment of the molecules. In the case of a homeotropic nematic sample of negative dielectric permittivity, there exists an infinite degeneracy when the molecules tend to pass to a planar configuration. The molecules are nucleated into a large number of domains around point defects associated with a radial optic axis distribution. Numerous studies relating to these point singularities or the umbilics have been described in the literature.[3-6]

Section 2 gives an experimental observation of the channelled spectrum in unpolarized light. After recapitulating the model describing the optic axis distribution in a quasi-polycrystal sample in Section 3, we present the theory of the channelled spectrum produced without external polarizers in Section 4. Finally, Sections 5 and 6 are devoted to a discussion of the results obtained and the conclusions drawn from them respectively.

2. OBSERVATION OF CHANNELLED SPECTRUM IN UNPOLARIZED LIGHT

We have used the optical setup shown in Figure 1 for observing the channelled spectrum due to nematic liquid crystal samples in unpolarized light. The sample has been placed in a spectrometer upstream with respect to its dispersive element which in the present case was

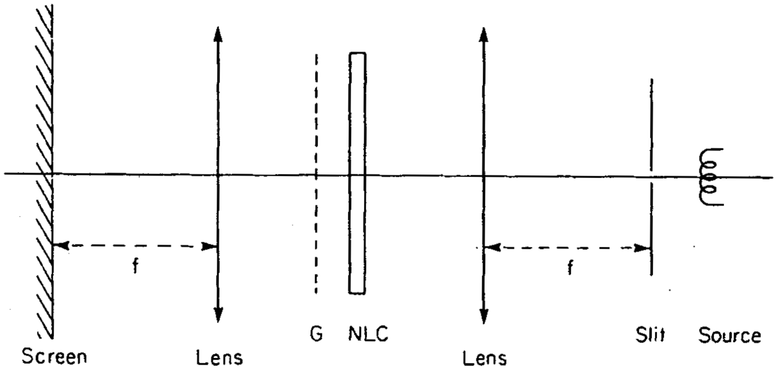

FIGURE 1 Optical arrangement of the experimental set-up.

a transmission grating. The channelled spectrum of the white light source is observed on screen S located in the conjugate plane of the spectrometer entrance slit. We used methoxybenzylidene-p-butylaniline (MBBA), having an anisotropy of the dielectric permittivity $\Delta\epsilon = -0.5$ and available commercially from Aldrich Chemicals, in our experiment. The parallel cell, shown in Figure 2, was formed by using a mylar spacer having a thickness of 24 μm between two glass plates. Transparent electrodes were deposited on the inner walls of the glass plates by coating them with tin oxide. The homeotropic orientation of the nematic on both interfaces was achieved by treating the cell walls with a thin layer of cetyltrimethyl ammonium bromide (CTAB). The liquid crystal cell was subjected to an a.c. electric field by applying voltage on the electrodes.

The interaction of the nematic molecules or equivalently of the optic axis of the liquid crystal sample with electric fields is a well known phenomenon. In the experiment under consideration, the mo-

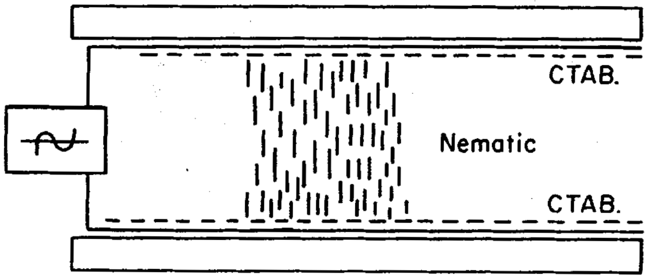

FIGURE 2 Sample geometry.

lecular reorientation occurred above the Freedericksz transition threshold of about 4.5 volts rms at 1 kHz. We have varied the a.c. voltage applied to the cell up to 30 volts rms by keeping its frequency constant at 1 kHz. The pattern on the screen consists of the usual dispersed image of the slit source for applied voltages up to at least a couple of volts above the threshold value. Above 7 volts rms, the spectrum became channelled by the appearance of vertical dark bands parallel to the grating lines. The behaviour of the channelled spectrum has been examined by increasing the applied voltage on the cell gradually to 30 volts rms. The frequency of the dark bands which is directly related to the effective crystal birefringence increases as the voltage on the sample is increased. As regards the visibility of the dark bands, it is also found to increase with the increase in the applied voltage. In addition to this, for a given value of the applied voltage the visibility of the dark bands has been found to vary a great deal from one sample to another. This point will be discussed in a subsequent Section. Figure 3 shows a photograph of the channelled spectrum corresponding to a sample at 20 volts rms.

The formation of channelled spectrum of a white light source due to a birefringent crystal plate, is attributed to the interference of the ordinary and extraordinary waves emerging from the crystal plate. In this case the sample is illuminated by a linearly polarized beam

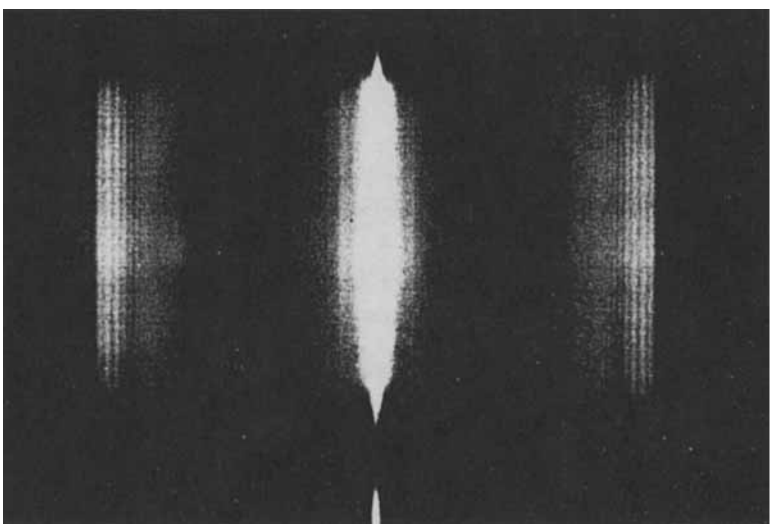

FIGURE 3 Photograph showing the channelled spectrum in absence of polarizers. See Color Plate I A.

and the complex amplitudes of the emergent waves are combined by the analyser placed downstream from the crystal plate. Thus, the use of a pair of crossed polarizers around the crystal plate is absolutely essential to produce the required interference fringes which channel the source spectrum. Evidently, the dark bands are characterized by the optical path difference between the ordinary and extraordinary beams and their visibility is maximum when the crystal optic axis bisects the angle between the axes of the two polarizers. In the beginning, the formation of the channelled spectrum in our experiment where no external polarizers were used around the sample, has been quite intriguing. However, a plausible explanation of the phenomenon has been given by taking into consideration the fact that the sample above the threshold consists of an ensemble of a large number of mono-crystal domains with their optic axes pointing in different directions. We now present the model describing the optic axis distribution in such quasi-polycrystal nematic samples and consider the propagation of a plane wave through them.

3. OPTIC AXIS DISTRIBUTION IN QUASI-POLYCRYSTAL SAMPLES

A homeotropic nematic sample with a negative anisotropy dielectric permittivity behaves like a perfect monocrystal below the threshold of the Freedericksz transition. Above the threshold of the applied electric field in the direction perpendicular to the cell walls, the molecules tend to realign themselves perpendicular to the electric field. Due to the degeneracy in the molecular realignment, there does not exist a preferred direction in which the molecules may lie in the plane of the plates. The molecules are, thus, nucleated in multiple domains. The optic axis direction of such a molecular distribution varies continuously and it can be defined with the help of the in-plane angle β and the out-of-plane angle ψ as shown in Figure 4. A mapping of the optic axis distribution over the XY-plane of the sample can be obtained experimentally by examining the sample under a polarizing microscope in monochromatic light. One obtains two sets of lines corresponding to the equal in-plane tilt angle ($\beta = \beta_o$) and the equal out-of-plane tilt angle ($\psi = \psi_o$) which we will refer as I-lines and O-lines respectively. The O-lines surround the umbilics whereas the I-lines join two defects. For any set of I-lines (β_o) joining two umbilics, there always exists another set of I-lines joining the same two defects but associated with the value $\beta = (\beta_o + \pi/2)$. This has been shown in Figure 5. This special feature of the crystal structure fulfills the role of combining the extraordinary and ordinary waves emerging from the sample for the purpose of observing the interference between them. The polarization direction of the ordinary wave for one set of I-lines is parallel to that of the extraordinary wave for the other set of I-lines, thus allowing the interference between them to take place.

4. THEORY

As shown in the Figure 2, the sample is illuminated at normal incidence with an unpolarized, polychromatic plane wave. The role of the grating consists of spreading out the wavelengths and for the sake of simplicity we will assume the incident wave to be monochromatic in the foregoing calculation. An unpolarized wave can be considered to be a superposition of an infinite number of isotropically distributed linearly polarized waves. The total intensity due to such a distribution will then consist of an incoherent addition of all polarization components. A rigorous analytical expression of the electric field of each of these components is beyond of the scope of this paper and we will use an approximate calculation in our analysis which is adequate for explaining the phenomenon. We consider the crystal plate to contain a distribution of umbilics, as defined previously. The electric field of the transmitted wave is determined by the local properties of the crystal plate.

An incident wave with its polarization direction inclined at an angle α with respect to the X-axis, as shown in the Figure 4, is written in terms of its electric field components as:

$$E_{i,x} = A_o \cos\alpha \tag{1a}$$

$$E_{i,y} = A_o \sin\alpha \tag{1b}$$

It produces a transmitted wave at a point $M(x,y)$ given by the electric field components:

$$E_{t,M,x} = A_o[\cos(\beta_M - \alpha)\cos\beta_M \exp(i\Phi_o) - \sin(\beta_M - \alpha)\sin\beta_M \exp(i\Phi_{eM})] \tag{2a}$$

$$E_{t,M,y} = A_o[\cos(\beta_M - \alpha)\sin\beta_M \exp(i\Phi_o) + \sin(\beta_M - \alpha)\cos\beta_M \exp(i\Phi_{eM})], \tag{2b}$$

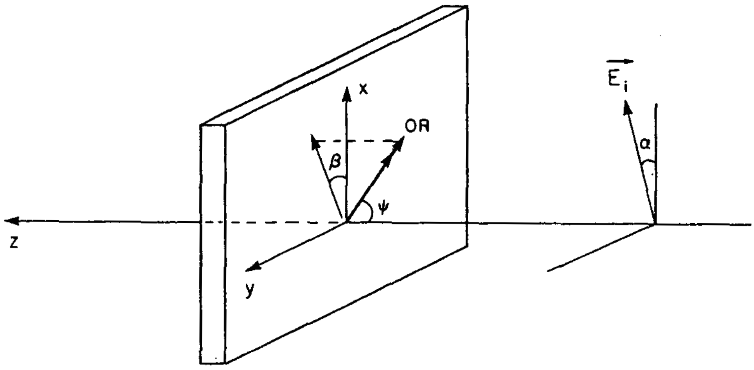

FIGURE 4 Definition of reference angles. E_i: Electric field of the incident beam. *OA*: Optic axis.

where β_M is the in-plane inclination at the point M and Φ_o and Φ_{eM} are the phase retardations for the ordinary and the extraordinary waves respectively.

In general, the phase retardation of the crystal plate depends on the local out of plane orientation of the optic axis. In the focal plane of the lens, the resultant electric field is the sum of contributions of all points in the pupil. Referring to the optic-axis mapping, the point M belongs to the O-line (ψ_M) and the I-line (β_M) associated with the umbilic located in their proximity. Also, there always exists another point M' belonging to the same O-line ($\psi_M = \psi_{M'}$) and the I-line characterized by $\beta_{M'} = \beta_M + \pi/2$ and associated with the same umbilic (Figure 5). Thus, the electric field associated with the two

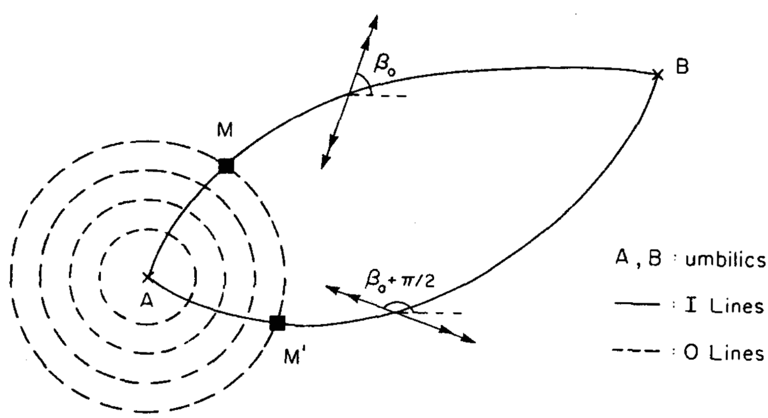

FIGURE 5 Mapping of the optic axis distribution in the sample (see text for the definition of I-lines and O-lines).

points M and M' of the pupil is given by:

$$E_{t,M,M',x} = E_{t,M,x} + E_{t,M',x} = A_o\cos\alpha[\exp(i\Phi_o) - \exp(i\Phi_{eM})] \quad (3a)$$

$$E_{t,M,M',y} = E_{t,M,y} + E_{t,m',y} = A_o\sin\alpha[\exp(i\Phi_o) - \exp(i\Phi_{eM})]. \quad (3b)$$

These field components are independent of β_M and the total complex amplitude due to all contributing points belonging to an O-line surrounding an umbilic is proportional to the electric field given by Eq. 3. The total complex amplitude on the screen contributed by an area limited by an O-line containing a point P is obtained by integrating Eq. 3 along any I-line from the umbilic to the point P. We have:

$$E_{u,x} = A_o\cos\,\alpha A_u \quad (4a)$$

$$E_{u,y} = A_o\sin\,\alpha A_u \quad (4b)$$

$$A_u = \pi \int_0^{R_p} [\exp(i\Phi_o) - \exp(i\Phi_{eM})]\rho d\rho, \quad (5)$$

where the subscript u refers to the umbilic under considerations and R_p is the curvilinear abscissa of the point P on the I-line. Experimentally, the phase factor Φ_{eM} associated with the I-line is found to vary rapidly in the core region of the umbilic, whereas it remains practically constant elsewhere over the curvilinear abscissa. As a consequence to this, we can rewrite Eq. 5 as:

$$A_u = C_u + S_u \sin(\Delta\Phi_p/2)\exp[i(\Phi_o + \Phi_{ep})/2], \quad (6a)$$

where

$$C_u = \pi \int_0^{R_c} [\exp(i\Phi_o) - \exp(i\Phi_{eM})]\,\rho d\rho \quad (6b)$$

and

$$\Delta\Phi_p = \Phi_o - \Phi_{ep}. \quad (6c)$$

Here R_c is the curvilinear abscissa of the core region, S_u is the area contained between the O-lines R_c and R_p and Φ_{ep} is the extraordinary phase retardation of that area. Now the total amplitude on the screen can be obtained by a discrete summation over the umbilics:

$$E_x = A_o \cos \alpha[\Sigma A_u] \tag{8a}$$

$$E_y = A_o \sin \alpha[\Sigma A_u]. \tag{8b}$$

We assume that each point P on the umbilic corresponds to the extreme value of the extraordinary phase retardation along an I-line. This implies that the Eqs. 8 account for a major part of the total surface area and that the phase retardation $\Delta\Phi_{ep}$ is constant for each umbilic area. Thus, the total intensity observed on the screen is given by the modulus squared of the complex amplitude in Eqs. 8:

$$I = I_o|\Sigma A_u|^2, \tag{9}$$

where

$$\Sigma A_u = (\Sigma C_u) + (\Sigma S_u) \sin(\Delta\Phi_p/2) \exp[i(\Phi_o + \Phi_{ep})/2]. \tag{10}$$

Clearly, the intensity given by Eq. 9 does not depend on the direction of the polarization of the incident beam. Therefore, for an unpolarized incident beam, the intensity transmitted by the birefringent sample is a periodic function of its birefringence. Thus the channelled spectrum can be observed on the screen without the necessity of using any polarizers in the light beam. The visibility of the fringe pattern is determined by the nature of the intensity dependence on the factor $\sin(\Delta\Phi_p/2)$ as well as on other experimental parameters as discussed below.

5. DISCUSSION

Generally, the first term on the right hand side of Eq. 10 is a complex number, the modulus of which is proportional to the total area (ΣS_c) of the core region. Thus, the fringe visibility will essentially depend on the ratio between the sample area (ΣS_u) where the phase retardation is constant over the total area ($\Sigma S_u + \Sigma S_c$) of the sample. Now, as the size of the core region is inversely proportional to the applied voltage, an increase in the voltage across the sample results in an improvement in the fringe visibility. This has been confirmed experimentally. Furthermore, the overall size of the area occupied by the core regions depends on the density of these defects. The sample observation under a microscope in monochromatic light showed that the area occupied by the cores becomes larger if the density of defects is smaller. The loss in the visibility of the dark bands in the channelled spectrum has been observed experimentally for samples having a low defect density. According to our first estimation, the period of the dark bands of the channelled spectrum varies directly as a function of the applied voltage and is a function of the maximum sample extraordinary phase retardation Φ_{ep} away from the core region. In effect, it is determined by the phase retardation averaged over the length of the defect away from the core and not by its maximum value. However, as the applied voltage increases, the mean value of the phase retardation approaches its maximum value as given by the maximum birefringence of the nematic. Thus, the visibility and the spatial frequency of bands increase as the applied voltage is increased above the threshold value. A steady final state is reached which corresponds to the channelled spectrum of the birefringent sample with its optic axis lying in the plane of the sample. The exact relation that might exist between the position of the bands, their visibility and the applied voltage would require more rigorous calculations by taking into consideration the exact structure of the nematic and other experimental parameters. Finally, the visibility of the dark bands is dependent on the degree of coherence of the illuminating light source in the usual manner.

6. CONCLUSION

A homeotropic nematic liquid crystal cell undergoing molecular reorientation under the influence of an external electric field shows defects characterizing the transition regions between various molecular domains. The presence of such disclinations has been used to explain the formation of channelled spectrum due to birefringent nematic samples in the absence of external polarizers. The experimental study using a nematic sample subjected to an a.c. electric field, when placed in a grating spectrometer exhibited channelled spectrum without requiring any external polarizers. Whereas the frequency of the bands is dependent on the sample birefringence, their visibility is inversely proportional to the size of the defect cores nucleated during their reorientation. The visibility increases if the density of the defects increases. Finally, the fact that a nematic liquid crystal sample free from any structural defects does not show any bands in the spectrum of a white light source, this phenomenon can be used for checking the quality of nematic preparations.

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