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Optical diffraction in twisted liquid-crystalline media—phase grating mode

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We have calculated the diffraction of light perpendicular to the twist axis in a chiral smectic C liquid crystal. In contrast to a cholesteric liquid crystal, in a chiral smectic C liquid crystal we find extra orders which form the odd orders in the diffraction pattern. For an incident linearly polarized light, at a general azimuth, these odd orders are linearly polarized and the even orders are elliptically polarized. The intensities of the odd orders are always independent of the azimuth of the incident light, while this is possible for even orders only at a particular tilt angle of the chiral smectic C liquid crystal. Also, for the incident vibration parallel or perpendicular to the twist axis the odd orders are polarized in the orthogonal linear state with respect to incident vibration, while the even orders are in the same linear state.

1. Introduction

Cholesteric and chiral smectic C liquid crystals exhibit strong reflection bands in the Bragg reflection mode [1]. However, Oldano [2] showed that the chiral smectic C liquid crystal has features very different from that of a classical cholesteric. These new features are not lost even in the uniaxial approximation for the local index ellipsoid. He has shown that the polarization states of the waves propagating through the medium are generally smooth functions of the tilt angle except at an angle $\theta_{\rm rev}$ at which drastic changes occur. In view of this result, we can except a chiral smectic C liquid crystal to behave very differently from classical cholesterics even in the phase grating mode: propagation perpendicular to the twist axis.

Optical diffraction in heterogeneous media can be understood as a consequence of phase fluctuations. Raman and Nath were the first to solve [3] this problem in the context of ultrasonic diffraction of light. Cholesteric and chiral smectic C liquid crystals are also examples of such media. These can be looked upon as spontaneously twisted nematic and smectic C phases respectively. In cholesterics, the experimentally observed optical diffraction [4] in a direction normal to the helical axis can be treated in an analogous manner [5] to diffraction in a phase grating. This is possible only when the electric vector of the incident light has a component perpendicular to the cholesteric twist axis. On the other hand in the chiral smectic C the medium acts as a phase grating for any azimuth of the incident light and results in a diffraction pattern. To our knowledge, this problem does not appear to have been addressed in the literature. In this paper we consider optical diffraction normal to the twist axis in cholesteric and chiral smectic C liquid crystals. We have also worked out the implications of absorption.

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Our treatment shows that the Raman and Nath theory predictions concerning intensities and phases [6], which are implied in non-absorbing cholesterics [5], are valid only at very low values of layer birefrigence. However, for realistic values of birefrigence, the intensities and phases of the different orders are very different. Introducing linear dichroism alters the phases and the strengths of the diffraction peaks.

A chiral smectic C liquid crystal behaves very differently in the phase grating mode. Here we get extra orders of diffraction compared to cholestrics which arise due to the pitch, P, being the optical period instead of $P/2$ as in cholesterics. These extra orders are surprisingly always linearly polarized and at an angle $(\pi/2)-\phi$ for any azimuth ϕ (with respect to the twist axis) of the incident linearly polarized light. Also, interestingly, the intensities of these orders are independent of ϕ. The even orders are generally elliptically polarized and they become linearly polarized only when the incident light is linearly polarized parallel or perpendicular to the twist axis. The intensities of the even orders are sensitive functions of ϕ. However, at a particular value of the tilt angle equal to θ_c, the intensities of these even orders are independent of ϕ. The introduction of linear dichroism alters the intensities of the various orders. Here the odd orders continue to be linearly polarized, but the azimuths and ellipticities of the even orders are affected.

2. Theory

2.1. *Cholesterics*

Considering incident light of amplitude A_0 linearly polarized at an arbitrary azimuth ϕ with respect to the twist axis, this will have a component $A_0 \sin \phi$ of the electric vector perpendicular to the cholesteric twist axis. For this component, the refractive index of the medium is a periodic function of the position. In such a case an incident plane wavefront emerges as a periodically corrugated wavefront. As in the Raman and Nath theory we assume the wavelength of the phase fluctuation to be large compared to its amplitude and the wavelength of light. Hence local refractions in the light rays are neglected. Earlier treatment of this problem [5] assumed the refractive index variation to be sinusoidal giving rise to a sinusoidally corrugated wavefront. Under such an approximation the results of the Raman and Nath theory of ultrasonic diffraction of light [3] can be directly employed to obtain the diffraction pattern. However, in reality, the periodic refractive index variation is more complicated and one has to obtain the diffraction pattern by finding the Fourier transform of the periodically corrugated emergent wavefront.

The refractive index n_z for the vibration perpendicular to the twist axis at any point z is given by

$$\frac{1}{n_z^2}=\frac{\sin^2\alpha}{n_1^2}+\frac{\cos^2\alpha}{n_2^2},$$

where

$$\alpha=\frac{2\pi}{P}z.$$

n_1 and n_2 are the principal refractive indices parallel and perpendicular to the local director, respectively. The emergent wavefront is described by

$$U(z)=A_0\sin\phi\exp(i2\pi n_z t/\lambda),$$

where t is the thickness of the sample. The diffraction pattern is given by the Fourier transform of $U(z)$

$$F(K)=\int_{-\infty}^{\infty} U(z)\exp(-iKz)\,dz,$$

where

$$K=\frac{2\pi\sin\Theta}{\lambda},$$

Θ being the angle of diffraction.

We make use of the fact that $U(z)$ is a periodic function that can be obtained as a convolution of the periodic array of δ functions with $V(z)$ representing the emergent wavefront for one pitch. Therefore the diffraction pattern $F(K)$ is nothing but a product of the Fourier transform of the δ array with that of $V(z)$.

For the component parallel to the twist axis, the emergent wavefront continues to be planar suffering an uniform phase retardation of $2\pi n_2 t/\lambda$. This does not result in any diffraction and contributes only to the central order which consequently will be elliptically polarized. By taking $\hat{n}_1=n_1+ik_1$ *and* $\hat{n}_2=n_2+ik_2$, the effects of the linear dichroism can be easily worked out.

2.2. *Chiral smectic C*

In a chiral smectic C we have a spontaneously twisted smectic C. Here the index ellipsoid spirals about the twist axis at a constant angle θ, the tilt angle of the smectic C. This is always a triaxial ellipsoid. In our treatment the incident plane wavefront is assumed to be linearly polarized.

For local biaxiality we use Oldano's model [2]: one of the principle directions is along the long axis of the molecule. The second one is along the local two-fold axis and the third is perpendicular to these two.

In this case the index tensor $[a]=[\varepsilon]^{-1}$ is given by

$$[a]=[a_{ij}],$$

where

$$\begin{aligned}
a_{11}&=b_1\cos^2\alpha+\sin^2\alpha(b_2\sin^2\theta+b_3\cos^2\theta),\\
a_{12}&=a_{21}=\tfrac{1}{2}(b_2-b_3)\sin\alpha\sin 2\theta,\\
a_{13}&=a_{31}=\tfrac{1}{2}[b_1-(b_2\sin^2\theta+b_3\cos^2\theta)]\sin 2\alpha,\\
a_{22}&=b_2\cos^2\theta+b_3\sin^2\theta,\\
a_{23}&=a_{32}=\tfrac{1}{2}(b_3-b_2)\cos\alpha\sin 2\theta,\\
a_{33}&=b_1\sin^2\alpha+(b_2\sin^2\theta+b_3\cos^2\theta)\cos^2\alpha,
\end{aligned}$$

where

$$b_1=\frac{1}{n_1^2},\quad b_2=\frac{1}{n_2^2}\quad\text{and}\quad b_3=\frac{1}{n_3^2}.$$

n_1, n_2 and n_3 are the refractive indices along the local principal axes.

At any layer, the central elliptic section of the index ellipsoid perpendicular to the direction of propagation (x axis) gives the refractive indices μ_1 and μ_2 for the two permitted linear orthogonal vibrations along the major and minor axes. These are given by

$$\mu_1 = [a_{22}\cos^2\psi + a_{23}\sin 2\psi + a_{33}\sin^2\psi]^{-1/2},$$

$$\mu_2 = [a_{22}\sin^2\psi - a_{23}\sin 2\psi + a_{33}\cos^2\psi]^{-1/2},$$

where ψ is the angle between one of the principal axes of the central elliptic section and the twist axis (z axis). It is given by

$$\psi = \tfrac{1}{2}\tan^{-1}\left[\frac{a_{23}}{(a_{22}-a_{33})}\right].$$

As light travels along any layer it gets resolved into two orthogonal linear vibrations along the major and minor axes of the elliptic section of the layer. However, the orientations of these principal vibrations will periodically vary along the twist axis. When light emerges from the system, we have from each smectic layer, two linear orthogonal vibrations. The azimuths and amplitudes of these vibrations vary periodically along the twist axis. To calculate the diffraction pattern in such problems [7] where the emergent wavefront has polarization fluctuations, we mathematically resolve at every point on the wavefront, the two emergent linear vibrations, along the two chosen orthogonal linear states. Therefore we end up with two periodically corrugated but orthogonally polarized wavefronts given by

$$U_{\|}(z) = A_{\|}\exp[\mathrm{i}\Phi_{\|}(z)],$$

$$U_{\perp}(z) = A_{\perp}\exp[\mathrm{i}\Phi_{\perp}(z)],$$

where $\Phi_{\|}(z)$ and $\Phi_{\perp}(z)$ are the phase fluctuations respectively in the two wavefronts. They result in two diffraction patterns described respectively by their individual Fourier transforms. These are evaluated in the same manner as described previously in cholesterics. The net vibration in any direction is obtained by adding coherently these two diffraction patterns.

If Δ is the phase difference between these two vibrations in any order and I_1, I_2 are their intensities then the ellipticity ω and azimuth λ of the resultant vibration are given by the standard formulae [8]

$$\tan 2\lambda = \cos\Delta\tan A,$$

$$\tan 2\omega = \tan\Delta\sin 2\lambda,$$

where A is given by $\tan A/2 = \sqrt{(I_1/I_2)}$.

3. Results

3.1. *Cholesterics*

The earlier study [5] on the diffraction in these systems assumed a sinusoidal fluctuation in the refractive index n_z. This results in the well known Raman and Nath diffraction pattern [3] regarding intensity and phases. This is valid only for small values of layer birefringence. Our treatment yields the following new results:

(i) the intensity ratios do not follow the Bessel function law predicted by Raman and Nath theory;

(ii) the phases of the odd and even orders of diffraction are no longer equal to even and odd multiples of π (or $\pi/2$) respectively [6]; this is due to the non-sinusoidal nature of the corrugated wavefront;

(iii) for high birefrigence values, the lower orders are extremely weak in intensity and most of the intensity appears in the higher orders.

We have also worked out the diffraction pattern in absorbing cholesterics (k and $\Delta k \approx 10^{-3}$). Here we find that:

(i) linear dichroism, as can be expected, decreases the overall intensity as compared with that of the non-absorbing case; however, it relatively suppresses certain of the strong diffraction peaks and enhances some of the weak ones;
(ii) at the same mean absorption, an increase in linear dichroism increases the intensities of the various orders.

3.2. *Chiral smectic C*

Our results very clearly bring out the differences between the chiral smectic C and the cholesteric diffraction patterns.

3.2.1. *Odd orders (extra orders)*

Since the optical period in a chiral smectic C liquid crystal corresponds to a 2π rotation of the local director, the diffraction pattern has extra orders compared to cholesterics. These extra orders form the odd orders of diffraction. For a tilt angle θ, in the range $0<\theta<\pi/2$ these orders are always linearly polarized. For $\phi=0$ and $\phi=\pi/2$ their azimuths are orthogonal to that of the incident linearly polarized light. For any other value of ϕ, their azimuths are at $\lambda=(\pi/2)-\phi$ to that of the incident light. In view of this, at $\phi=\pi/4$ the azimuth of the incident linearly polarized light and those of the odd orders are parallel.

We have carried out our calculations both for local uniaxial ($n_1=n_2\neq n_3$) and local biaxial ($n_1\neq n_2\neq n_3$) symmetries. Figures 1 and 2 depict the diffraction pattern for the uniaxial approximation while figures 3 and 4 show the results for the biaxial approximation. We can clearly see that in both approximations the computed intensities of the different odd orders do not change on changing the azimuth ϕ of the incident light. They depend only on the tilt angle θ of the chiral smectic C phase.

3.2.2. *Even orders*

These are generally elliptically polarized. However, for $\phi=0$ or $\phi=\pi/2$, they are linearly polarized parallel to the incident vibration. As shown in figures 1 to 4, the $\phi=0$ polarization mode incident on the structure with $\theta=\pi/2$ (i.e. cholesterics) does not yield any diffraction pattern in contrast to θ in the range $0<\theta<\pi/2$ (i.e. chiral smectic C) which yields a rich diffraction pattern. For a given θ the different even orders have different intensities, azimuths and ellipticities which are very sensitive to the value of ϕ. However, we get an intriguing result at a particular tilt angle equal to θ_c for the chiral smectic C. At this value of θ, the intensities of the various even orders (computed up to the eighth order) are independent of ϕ. This is depicted in figure 4, second row. For the set of parameters that we used θ_c turns out to be very nearly $\pi/8$. Generally θ_c happens to be less than $\pi/4$ and hence experimentally realizable. Our calculations indicate that θ_c is non-zero for the biaxial approximation and tends to zero for the uniaxial approximation. For any given value of n_3, there is an upper limit for (n_1-n_2) beyond which such a phenomenon does not exist. Also, at a fixed value of (n_1-n_2), θ_c increases with a decrease in the value of n_3 and vice versa.

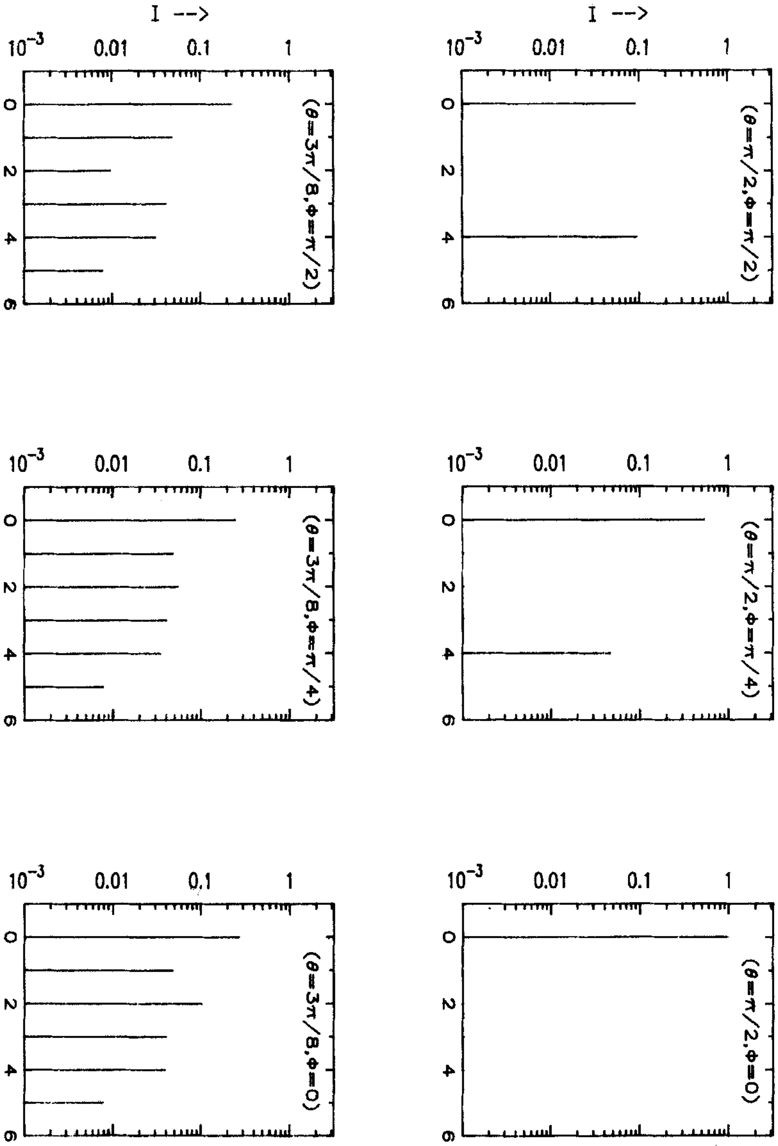

Figure 1. Intensities (I) of the various diffraction orders indicated by the integers on the x axis as a function of the tilt angle (θ) of the uniaxial structure and the azimuthal angle (ϕ) of the incident linearly polarized light computed for $n_1 = n_2 = 1{\cdot}535$, $n_3 = 1{\cdot}605$, $P = 10\,\mu$m, $t = 20\,\mu$m and $\lambda = 633$ nm. Cholesterics (i.e. $\theta = \pi/2$) exhibit diffraction with only even orders which are linearly polarized perpendicular to the twist axis. However, the central order is linearly polarized only for $\phi = \pi/2$. In the chiral smectic C (i.e. $0 < \theta < \pi/2$) the odd orders are always linearly polarized with the even orders in the elliptic state.

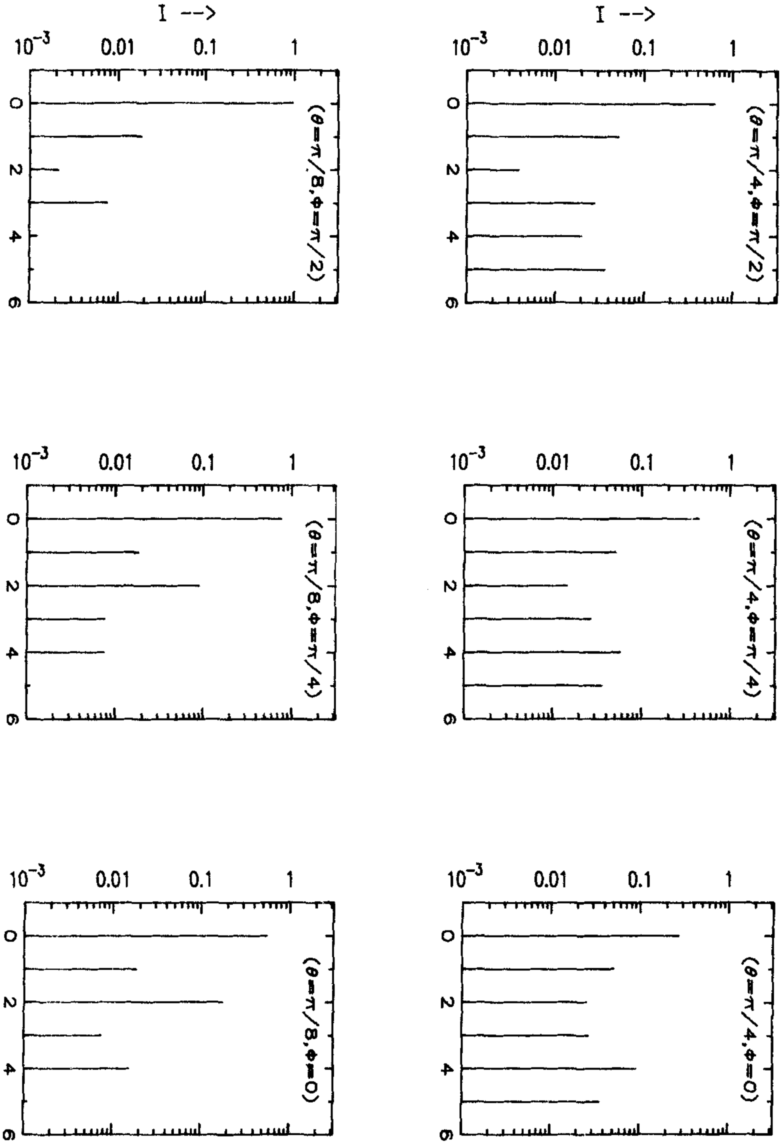

Figure 2. Chiral smectic C diffraction orders indicated by the integers on the x axis (for $n_1 = n_2 = 1{\cdot}535$, $n_3 = 1{\cdot}605$, $P = 10\,\mu\text{m}$, $t = 20\,\mu\text{m}$ and $\lambda = 633\,\text{nm}$) for smaller angles of tilt angle θ.

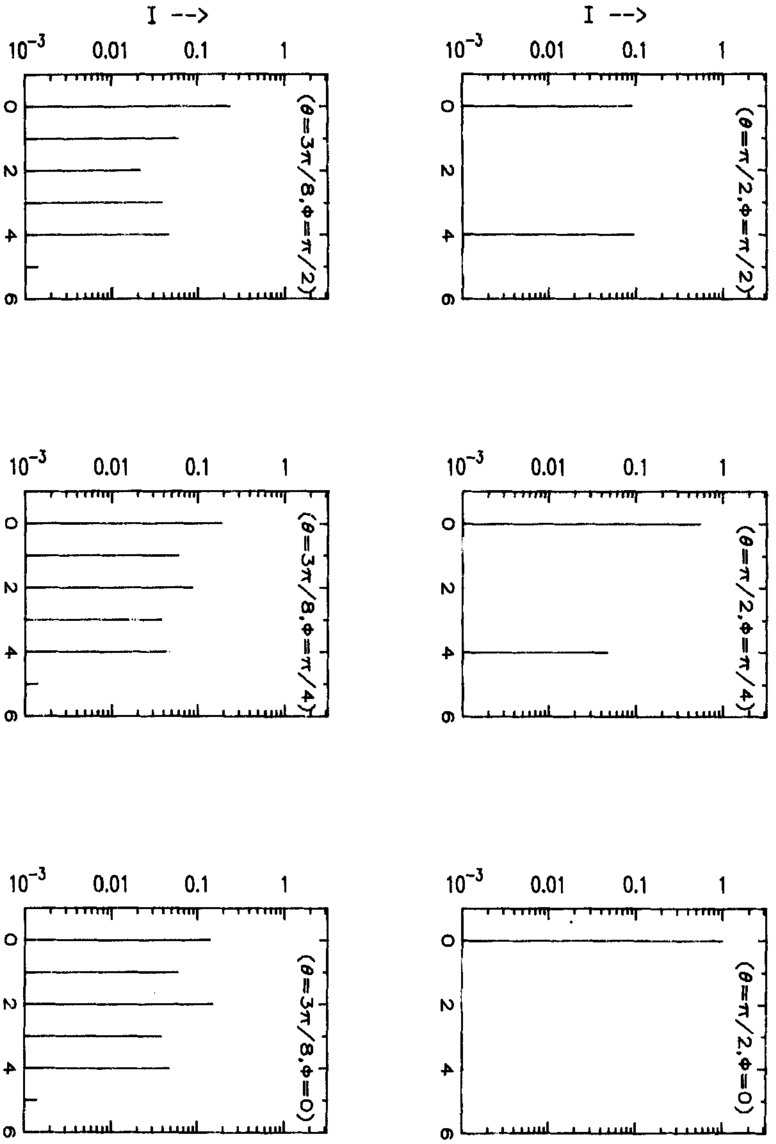

Figure 3. Intensities (I) of the various diffraction orders shown as integers on the x axis as a function of the tilt angle (θ) of the biaxial structure and the azimuthal angle ϕ of the incident linearly polarized light computed for $n_1 = 1{\cdot}535$, $n_2 = 1{\cdot}545$, $n_3 = 1{\cdot}605$, $P = 10\,\mu$m, $t = 20\,\mu$m and $\lambda = 633$ nm.

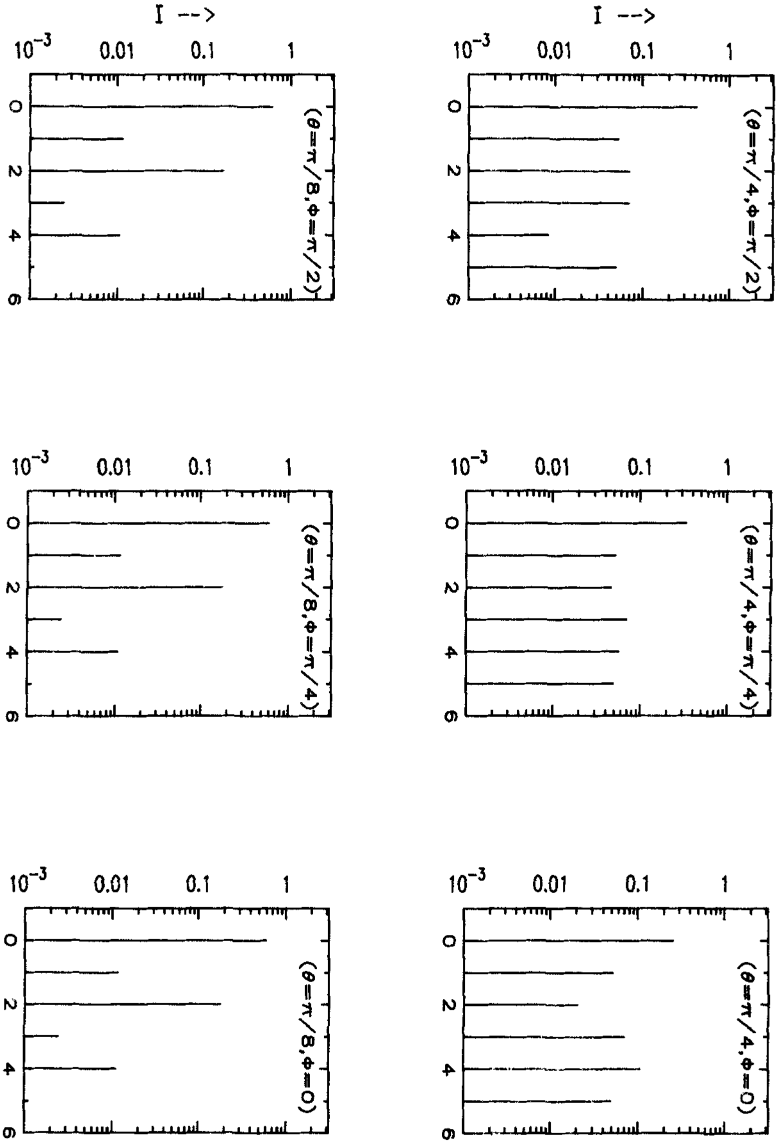

Figure 4. Chiral smectic C diffraction orders shown as the integers along the x axis (for $n_1 = 1{\cdot}535, n_2 = 1{\cdot}545, n_3 = 1{\cdot}605, P = 10\,\mu\text{m}, t = 20\,\mu\text{m}$ and $\lambda = 633$ nm) for smaller angles of tilt angle θ. Notice that for $\theta = \pi/8(\approx \theta_c)$ the intensities (I) of the even orders are independent of ϕ. However, as one can see from figures 1 to 4, for any θ, the intensities of the various odd orders are independent of ϕ.

3.2.3. *Effect of wavelength*

The variation in birefrigence Δn affects the intensities of the various orders. However, variation in the wavelength λ affects both the intensities and positions of the different orders.

3.2.4. *Effect of absorption*

Introduction of linear dichroism (k and $\Delta k \approx 10^{-3}$) redistributes the intensities of the different orders with respect to the non-absorbing case. However, the odd orders continue to be in the same linear polarization state, while those of the even orders are altered. For $\phi = 0$ or $\pi/2$, the even orders are in the same polarization state as the incident vibration and the odd orders are in the orthogonal state.

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