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## Liquid Crystals

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## Optical properties of magnetically doped cholesterics

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We have computed the optical diffraction pattern for linearly polarized light incident normal to the twist axis (phase grating mode) of a magnetically doped cholesteric (ferrocholesteric). The intrinsic Faraday rotation of the magnetic grains results in extra orders of diffraction. Also we find diffraction for any azimuth of the incident vibration. Further, using the Jones  $N$  matrices we have worked out the optical properties for light propagation along the twist axis on the very low wavelength side of the reflection band. We find that the medium behaves very differently from a normal cholesteric.

### 1. Introduction

Cholesteric phases in which magnetic grains are suspended have become important in recent times. Cholesteric phases of rod-shaped molecules in which needle-shaped magnetic grains are aligned along the local director have been realized in the laboratory. There are systems with grains having magnetization parallel to the local director [1] as well as magnetization perpendicular to the local director [2]. The former will give rise to a magnetically doped cholesteric (ferrocholesteric) phase with magnetization gradually twisting with the local director much like a helimagnetic system and in the latter case we get the same phase with the magnetization of the grains parallel to the twist axis. Since these grains can even be optically transparent (for example garnets), their inherent Faraday rotation becomes very important.

The rotatory power due to magnetic grains depends on the direction of propagation of light with respect to the magnetization  $\mathbf{m}$  and is given by

$$\rho = \beta |\mathbf{m}| \cos \theta = \rho_0 \cos \theta$$

where  $\beta$  is a constant and  $\theta$  is the angle between  $\mathbf{m}$  and the direction of propagation. This dependence of the Faraday rotation on  $\theta$  leads to optical properties which are very different from those of the normal cholesterics. We have tried to bring out salient differences between the magnetically doped cholesterics (MDCs) and normal cholesterics.

In §2 we have considered linearly polarized light propagating perpendicular to the twist axis (phase grating mode) of MDCs. The optical periodicity for such a medium is  $P$ , the pitch, instead of  $P/2$  as in cholesterics. This modification of the periodicity gives rise to extra orders (odd orders) of diffraction in addition to those orders (even orders) obtained in cholesterics. In this sense the diffraction pattern is very similar to that of a  $S_C^*$  phase [3, 4].

In §3 we have worked out the properties of the medium for light propagation along the twist axis, far away, on the lower wavelength side of the reflection band.

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We find that depending upon the sign of optical rotation, the medium can act as a Mauguin retarder or a de Vries rotator.

## 2. Light propagation perpendicular to the twist axis

### 2.1. Theory

We consider the magnetization  $\mathbf{m}$  of the grains to be parallel to the local director. We further assume that the medium is locally uniaxial about the local director. The linearly polarized light incident on the medium will see a variation of refractive index along  $Z$ , the twist axis, so that the incident plane wavefront emerges as a periodically corrugated wavefront with fluctuations in azimuth and ellipticity of the state of polarization. As linearly polarized light travels along any layer it splits into two orthogonal elliptic vibrations. The refractive indices of the medium for these two elliptic vibrations are given by [5]

$$\frac{1}{n_R^2} = \frac{1}{2} \left[ (\eta_{\perp}(z) + \eta_{\parallel}(z)) - \left( \sqrt{[(\eta_{\perp}(z) - \eta_{\parallel}(z))^2 + 4\gamma^2]} \right) \right],$$

$$\frac{1}{n_L^2} = \frac{1}{2} \left[ (\eta_{\perp}(z) + \eta_{\parallel}(z)) + \left( \sqrt{[(\eta_{\perp}(z) - \eta_{\parallel}(z))^2 + 4\gamma^2]} \right) \right],$$

where

$$\eta_{\perp}(z) = \frac{\cos^2(\alpha)}{n_1^2} + \frac{\sin^2(\alpha)}{n_2^2},$$

$$\eta_{\parallel}(z) = \frac{1}{n_1^2}.$$

Here  $\alpha = (2\pi/P)z$ , and  $n_2, n_1$  are the refractive indices along and perpendicular to the local director in the absence of Faraday rotation.

The parameter  $\gamma$  is related to the rotatory power  $\rho$  of the medium by the relation

$$\gamma = \frac{\rho\lambda}{(\bar{n})^3\pi}.$$

Here  $\lambda$  is the wavelength of light and  $\bar{n}$  is the mean refractive index of the medium.

These elliptic vibrations have ellipticity given by

$$\omega_R = \frac{1}{2} \tan^{-1} \left[ \frac{2\gamma}{\eta_{\parallel} - \eta_{\perp}} \right] \quad \text{and} \quad \omega_L = \pi/2 - \omega_R.$$

The elliptic vibration can be mathematically resolved at each point of the emergent wavefront into two linear vibrations polarized along and normal to the twist axis. This results in two periodically corrugated, orthogonally linearly polarized wavefronts given by

$$U_{\parallel}(z) = A_{\parallel}(z) \exp [i\psi_{\parallel}(z)],$$

and

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

where  $A_{\parallel}(z)$  and  $A_{\perp}(z)$  are the amplitude fluctuations and  $\psi_{\parallel}(z)$  and  $\psi_{\perp}(z)$  are the phase fluctuations of these wavefronts, respectively. We assume that the wavelength of the corrugation is large compared to its amplitude. The diffraction patterns due to

these two wavefronts are given by their individual Fourier transforms. The complete diffraction pattern is obtained by coherently adding the diffraction pattern due to the two wavefronts.

## 2.2. Results and discussion

Using the above theory we have computed the diffraction pattern for experimentally realisable parameters.

The Faraday rotation due to the magnetic grains results in diffraction for any azimuth  $\phi$  (with respect to the twist axis) of the incident, linearly polarized light, unlike cholesterics where it occurs only for  $0 < \phi \leq \pi/2$ . Also the MDC has a periodicity of  $2\pi$  due to the magnetization  $\mathbf{m}$  which will result in extra orders of diffraction. We find that for  $\phi = 0$  or  $\pi/2$  the odd orders are linearly polarized in the state orthogonal to that of the incident light whereas the even orders are linearly polarized in the same state as that of the incident light. We also find that for any other general azimuth  $\phi$  in the range  $0 < \phi < \pi/2$  the odd and even orders are in general elliptically polarized.

The computed diffraction patterns with intensity as a function of scattering vector are shown in figures 1(a), (b), 2(a) and (b). Each set gives patterns corresponding to a given value of the Faraday rotatory power  $\rho$  and for  $\phi = 0, \pi/4$  and  $\pi/2$ . We find the interesting result that the intensity of any odd order is independent of the azimuth of the incident light and only varies with  $\rho$ . The Faraday

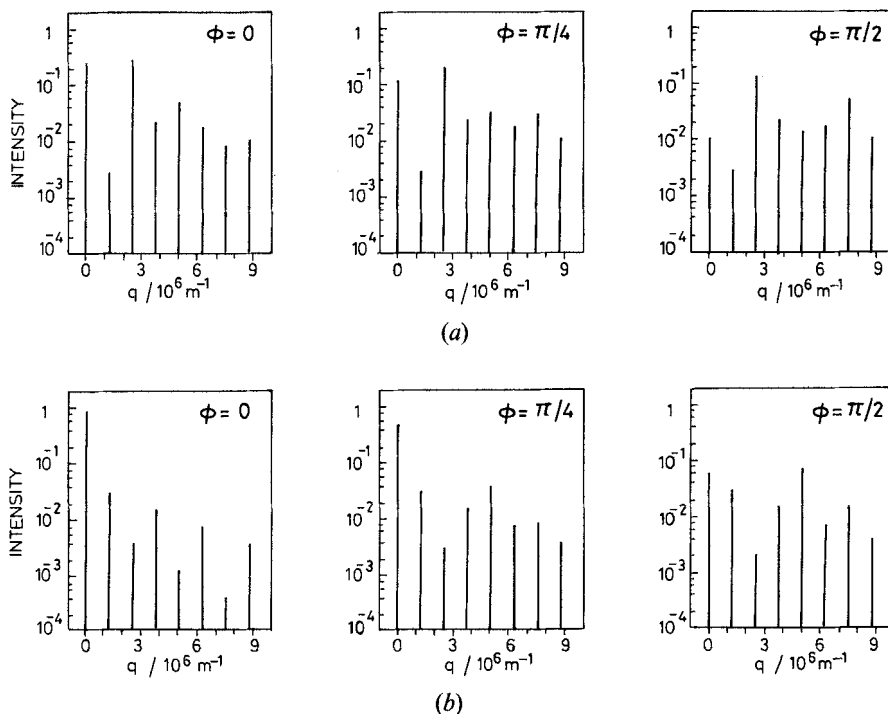


Figure 1. Computed diffraction pattern in a MDC showing intensity as a function of scattering vector  $q$  for  $\lambda = 0.633 \mu\text{m}$ ;  $\Delta n = 0.07$ ;  $n_1 = 1.535$ ;  $n_2 = 1.605$ ;  $P = 5 \mu\text{m}$ ; sample thickness ( $t$ ) =  $20 \mu\text{m}$ . For (a)  $\rho_0 = 1.92 \times 10^3 \text{ rad cm}^{-1}$ , and for (b)  $\rho_0 = 3.84 \times 10^2 \text{ rad cm}^{-1}$ .

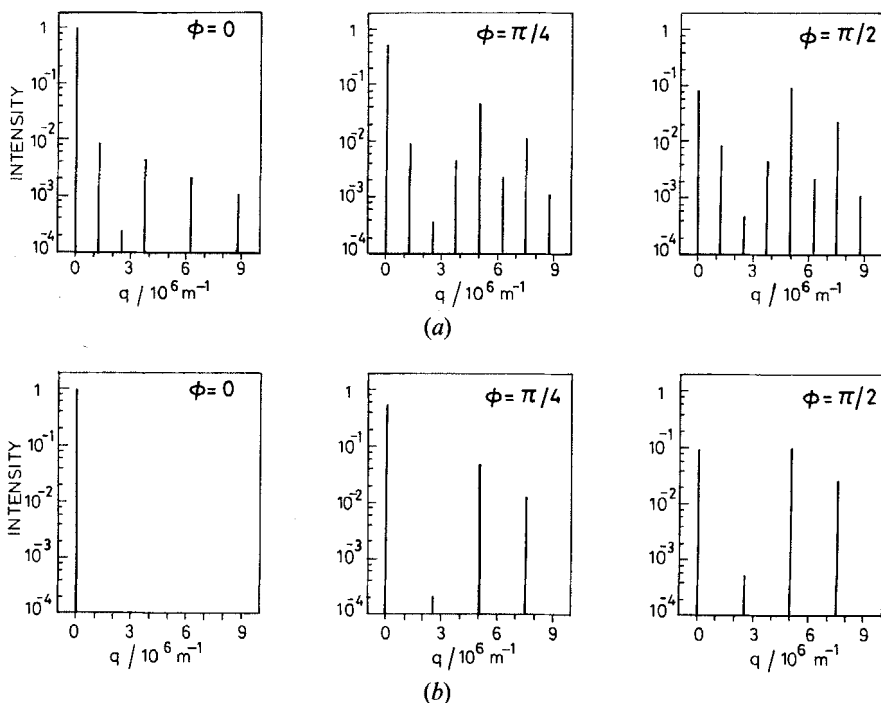


Figure 2. Computed diffraction pattern for the same values of  $\lambda$ ,  $\Delta n$ ,  $n_1$ ,  $n_2$ ,  $P$  and  $t$  given in figure 1. For (a)  $\rho_0 = 1.92 \times 10^2 \text{ rad cm}^{-1}$ , and for (b)  $\rho_0 = 0$ , i.e. a normal cholesteric.

rotation not only results in extra orders but also alters the intensities of the even orders as can be seen from the figures. All these features are seen even at extremely low values of  $\rho$  (see figure 2(a)). It should be noted that in the diffraction patterns calculated for higher values of  $\rho$ , the intensities of the higher orders grow at the expense of lower orders (see figure 1(a)). For comparison we give in figure 2(b) the diffraction patterns for zero Faraday rotation (i.e. a normal cholesteric). As is to be expected, in this case the odd orders do not exist at all for any value of  $\phi$  and for  $\phi = 0$  the entire pattern degenerates to the zeroth order. We would like to remark that in many respects the intensity and polarization features of the odd orders of diffraction pattern are very similar to those found for the  $S_C^*$  phase [3].

### 3. Light propagation parallel to the twist axis

#### 3.1. Theory

In this case we assume that the magnetic grains are parallel to the local director but with  $\mathbf{m}$  along the twist axis. The medium at any point acts as a linearly birefringent plate having Faraday rotation.

In the Jones matrix formulation, the  $N$  matrix for such a plate is given by [6]

$$\mathbf{N}_0 = \begin{bmatrix} -ik + ig_0 & -\rho \\ \rho & -ik - ig_0 \end{bmatrix},$$

where  $2g_0$  is the phase retardation per unit thickness,  $k$  is the wave vector in the medium and  $\rho$  is the Faraday rotatory power. We assume the medium to be twisting along the  $Z$  axis. Then the  $N$  matrix of a layer at  $z$  is given by

$$\mathbf{N} = S(q_0 z) \mathbf{N}_0 S(-q_0 z),$$

where  $q_0 = 2\pi/P$ ,

$$S(q_0 z) = \begin{bmatrix} \cos q_0 z & -\sin q_0 z \\ \sin q_0 z & \cos q_0 z \end{bmatrix}$$

so that the Jones  $M$  matrix for the entire sample can be written as

$$\mathbf{M} = S(q_0 z) \exp \{ [\mathbf{N}_0 - q_0 S(\pi/2)] z \}.$$

If the electric vector of the incident light is

$$\mathbf{E} = \begin{bmatrix} E_x \\ E_y \end{bmatrix},$$

then the emergent electric vector is given by

$$\mathbf{E}' = \begin{bmatrix} E'_x \\ E'_y \end{bmatrix} = \mathbf{M} \begin{bmatrix} E_x \\ E_y \end{bmatrix} = \mathbf{M} \mathbf{E}.$$

### 3.2. Results and discussion

#### 3.2.1. Case 1

$|\rho - q_0| \gg |g_0|$  with  $\rho$  and  $q_0$  of opposite signs (i.e. when the direction of propagation of light is opposite to that of  $\mathbf{m}$ ). Then

$$\mathbf{M} = \exp(ikz) S(\rho z).$$

The medium, to a very good approximation, acts as a pure rotator, i.e. there will be a rotation in the plane of polarization of the incident light entering the medium.

#### 3.2.2. Case 2

$|\rho - q_0| \ll |g_0|$  with  $\rho$  and  $q$  of the same sign (when the propagation of light is in the same direction as that of  $\mathbf{m}$ ). Then

$$\mathbf{M} = \exp(ikz) \begin{bmatrix} \cos q_0 z & -\sin q_0 z \\ \sin q_0 z & \cos q_0 z \end{bmatrix} \begin{bmatrix} \exp(ig_0 z) & 0 \\ 0 & \exp(-ig_0 z) \end{bmatrix}.$$

This leads to Mauguin's solution, i.e. the incident vibration splits into two linear orthogonal vibrations polarized along and perpendicular to the local director. As these vibrations travel they follow the director as in a twisted nematic.

Therefore depending on the propagation of light along or opposite to the direction of  $\mathbf{m}$  the medium can act as a Mauguin retarder or as a de Vries rotator, respectively. For example, this happens for a MDC of pitch  $\approx 30 \mu\text{m}$ ,  $\rho_0 \approx 2.0 \times 10^3 \text{ rad cm}^{-1}$  and birefringence  $\Delta n = 0.025$ . Such a medium between two appropriately aligned polaroids can act as an optical diode, i.e. transmitting light in one direction and blocking it completely in the opposite direction.

Interestingly, the condition  $|\rho - q_0| \gg |g_0|$  need not imply that  $|q_0| \gg |g_0|$ . Here this condition can also be satisfied for small values of  $q_0$ , i.e. for large values of pitch. Thus the existence of Faraday rotation can lead to a de Vries limit even for a medium of large pitch. This is contrary to the case of normal cholesterics where the de Vries limit is reached only for very small pitch values.

Further the condition  $|(\rho - q_0)| \ll |g_0|$  does not mean that  $|q_0| < |g_0|$ . In fact  $|q_0| > |g_0|$  is also possible. In this situation, in the absence of Faraday rotation, the solution will not go over to Mauguin's limit, but to the de Vries limit [7].

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