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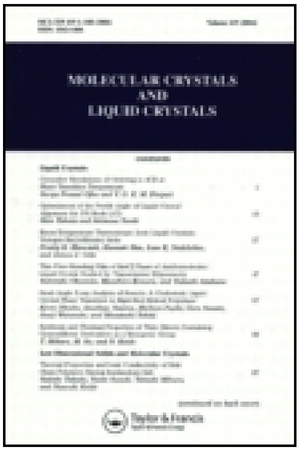

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Optical Activity of Small Pitch Chiral Smectic C Liquid Crystals

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OPTICAL ACTIVITY OF SMALL PITCH CHIRAL SMECTIC C LIQUID CRYSTALS

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Abstract Recently it has been shown that chiral smectics C liquid crystals with pitch shorter than the light wavelength are optically active with maximum rotatory power for light propagating orthogonally to the helix axis. We report the first experimental evidence of this fact.

Natural optical activity (or gyrotropy) has been associated, since its discovery, to helically shaped structures. In the last decades it has been recognized that gyrotropy comes in general from molecules having the shape of helices or segments of an helix[1], or from molecules arranged in such a way to give helically shaped structures. However, exact analytic expression for the gyrotropic properties of helicoidal structures are available only for the particular case of light propagating along the helix axis of cholesteric-like media[2,3], and are well approximated by de Vries' equation[2]. It has been recently shown that even simpler expressions are valid for small pitch smectics C*, for any direction of the light beam[4]. In the limit $p \ll \lambda$ a smectic structure whose helix axis is along the z-coordinate is well approximated by a homogeneous medium whose dielectric tensor and gyrotropy pseudo tensor are given by

$$\boldsymbol{\varepsilon}^{\text{eff}} = \begin{pmatrix} \tilde{\varepsilon}_o & 0 & 0 \\ 0 & \tilde{\varepsilon}_o & 0 \\ 0 & 0 & \tilde{\varepsilon}_e \end{pmatrix} + i g_\perp \begin{pmatrix} 0 & 0 & -m_y \\ 0 & 0 & m_x \\ m_y & -m_x & 0 \end{pmatrix}, \tag{1}$$

$$\mathbf{g} = \begin{pmatrix} g_\perp & 0 & 0 \\ 0 & g_\perp & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{2}$$

where p is the helical pitch, $\mathbf{m} = \mathbf{k}/k_0$,

$$\tilde{\varepsilon}_e = \varepsilon_o + \varepsilon_a \cos^2 \alpha\ , \quad \tilde{\varepsilon}_o = \varepsilon_o(1 + \varepsilon_e/\tilde{\varepsilon}_e)/2\ , \tag{3}$$

$$g_\perp = -\frac{p}{\lambda}\frac{\varepsilon_a^2}{8\tilde{\varepsilon}_e}\sin^2(2\alpha)\,, \tag{4}$$

α is the tilt angle of the structure, ε_e and ε_o are the principal values of the local dielectric tensor of the periodic medium and $\varepsilon_a = \varepsilon_e - \varepsilon_o$ is the dielectric anisotropy.

The effective medium is uniaxial in both the real and imaginary parts of the dielectric tensor (*bi-uniaxial*), and the gyrotropy pseudo tensor **g** depends on a single parameter $g_\perp$, as shown by Eq. (2). The index $\perp$ refers to the optical axis, which is coincident with the helix axis.

The gyrotropy parameter $g_\perp$ depends on the square of the local dielectric anisotropy of the medium. Its sign is therefore the same for media having positive and negative local anisotropy. It is maximum for $\alpha = 45°$, zero for $\alpha = 0°$ and $\alpha = 90°$ (namely for a cholesteric liquid crystals). The absence of the parallel component $g_\parallel$ means that the optical activity reaches its maximum value for light propagating *orthogonally to the helix axis* and is zero for parallel light. This property is quite unexpected, since it is well known that, for $p > \lambda$, smectics C* display for parallel light a huge rotatory power, which is given by an equation having the same structure of de Vries' equation for cholesterics. A residual optical activity for parallel light survives for $p < \lambda$, but decreases as $(p/\lambda)^3$, becoming practically negligible, with respect to the orthogonal component, for $p < \lambda/5$ [4,5].

This theoretical prediction has been experimentally proved on the smectic C* compound ZKS-2502, with pitch $p = 0.17\ \mu$m, tilt angle $\alpha = 26°$ and effective optical anisotropy $\Delta n = 0.16$. The measurements have been performed on a sample with the helix axis perpendicular to the glasses, to investigate another important point concerning periodic media. The electromagnetic eigenwaves within any periodic medium are in general Bloch waves. This means that strictly speaking the medium can never be treated as homogeneous. When we consider an equivalent homogeneous medium, we simply neglect all the Fourier components of the Bloch wave, except the zeroth-order one. This is a general feature of periodic media, also valid for usual crystals. The fact that crystals are generally considered in optics as homogeneous media poses the questions: why the higher Fourier components can generally be neglected, and in what cases they can play some non negligible role? These questions can be easily answered in the particular case of chiral smectics, which are periodic in only one direction. To this purpose, we have considered a sample confined between the planes $z = 0$ and $z = d$, where d is an integer multiple of the pitch, and a light beam in the plane (x, z). The cartesian components of the director are given by

$$n_x = \sin\alpha\cos\varphi\,, \quad n_y = \sin\alpha\sin\varphi\,, \quad n_z = \cos\alpha\,, \tag{5}$$

where $\varphi = qz + \varphi_0$ and $q = 2\pi/p$. The optical properties of the sample are summarized by the transfer Berreman matrix $U(d,\varphi_0)$ which gives the electromagnetic field at $z = d$ as a function of the field at $z = 0$ [6]. The matrix depends on the phase constant φ_0, which defines the average direction of the molecules at $z = 0$ with respect to the incidence plane of light, and also defines the phase constants of the neglected Fourier components. The transfer matrix of the homogeneous model[4] is obtained by averaging $U(d,\varphi_0)$ over φ_0. For samples with the helix axis

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not perpendicular to the boundaries, the averaging over φ_0 is automatic, and the neglected Fourier components play no role. The higher order Fourier components can play a non negligible role only in samples with boundary planes orthogonal to the axes of the reciprocal lattice.

The experiment has been performed by sending TM and TE linearly polarized light, with $\lambda = 0.6328$ μm on a sample of thickness $d = 13.6$ μm with incidence angle between $-50°$ and $+50°$. The polarization of the transmitted light is in general elliptical and has been analyzed with a Glan-Taylor polarizer. We have measured the ratio between the minimum and the maximum intensity obtained by rotating the analyzer, namely the square of the amplitude ratio of the ellipse axes (Fig. 1a and 2a) and the angle between the main axis of the ellipse and the polarization direction of the incident light (Fig. 1b and 2b). In Fig. 1 we have plotted the experimental values, averaged over φ_0, for TE input polarization (circles), the theoretical values given by the homogeneous model by assuming as material parameters the nominal ones (dotted lines), with $n_o = 1.5$, and the theoretical values obtained by assuming a tilt angle $\alpha = 30°$ (solid lines) instead of the nominal one, to show the strong dependence of the output polarization on the material parameters.

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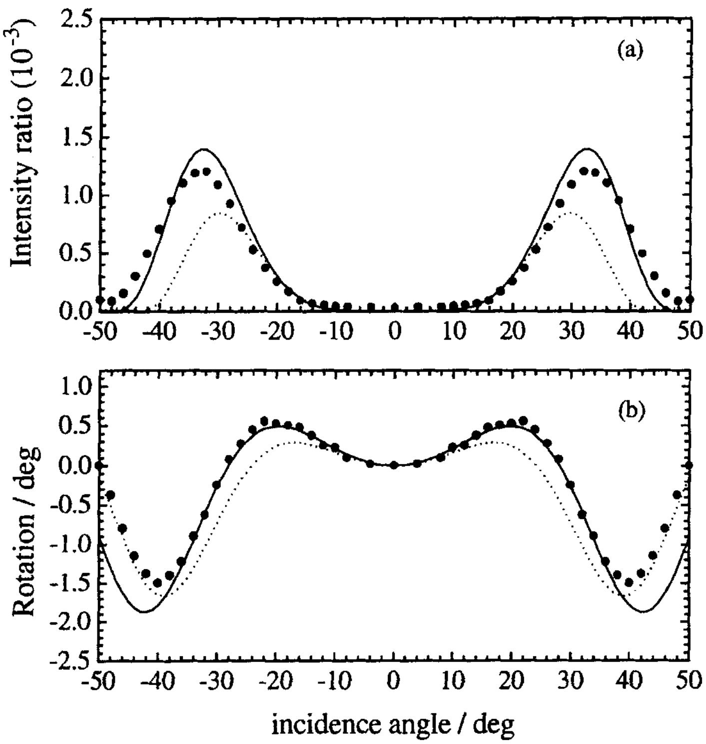

FIGURE 1 Polarization parameters (squared ratio of the ellipse axes (a) and axes rotation (b)), averaged over φ_0, of the transmitted beam for incident TE polarization: experimental data (circles), theoretical data with tilt angle $\alpha = 26°$ (dotted lines) and $\alpha = 30°$ (solid lines).

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A best fit of the experimental data has not been done, because the sample was not perfectly mono domain owing to surface irregularities and non uniform thickness over the light beam. However the discrepancy between the experimental and the theoretical data are largely within the estimated deviation coming from the intrinsic uncertainty of the nominal values of the material parameters (tilt angle, pitch, local dielectric anisotropy, sample thickness) and from the non perfect TE polarization of the incident polarization (a small TM component was indeed present). In the absence of optical activity, the homogeneous model would give a transmitted beam with the same polarization state of the incident beam. The effect of chirality on the output polarization is rather complex, since the internal eigenwaves are in general elliptically polarized. The medium transforms linear into elliptic polarization, and at the same times rotates the principal axes of the ellipse. The oscillatory behavior of the rotation angle apparent in Fig. 1b is not due to a sign reversal of the gyrotropy parameter, but to the progressive dephasing of the TE and TM components. This can be easily understood by recalling that, for linear polarization, a $(2n+1)\pi$ dephasing, with integer n, simply reverses the polarization direction with respect to the incidence plane.

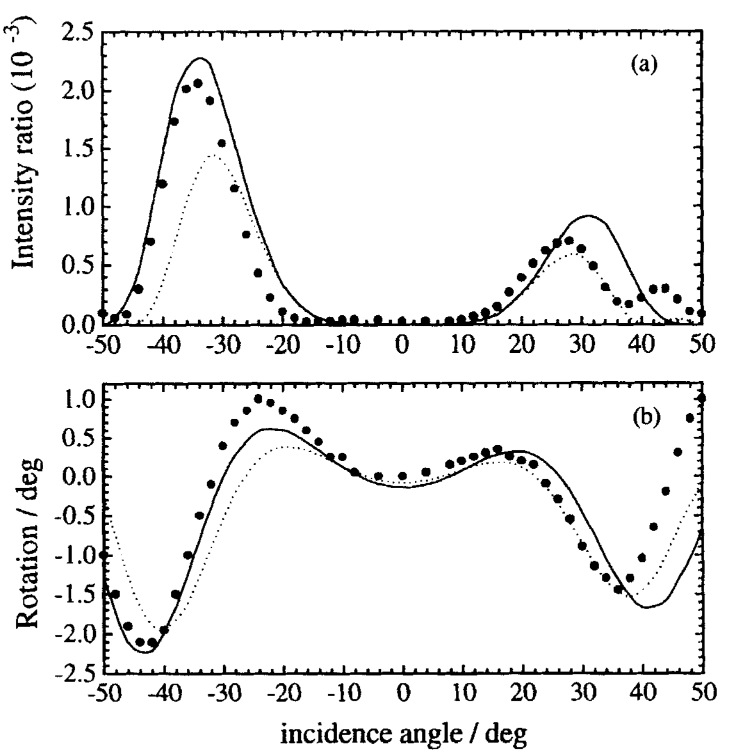

FIGURE 2 Same as fig. 1, but for $\varphi_0 = 60°$.

Figure 2 refers to a phase constant $\varphi_0 = 60°$. It shows that for our sample the higher order Fourier components play indeed a strong role and the homogeneous model is of limited usefulness. The theoretical curves have been obtained by a numerical integration of Maxwell's equations, with the same parameters of Fig. 1. This is in our opinion a most important point, which evidences the limits of validity of any homogeneous model for periodic structures and crystals.

In conclusion, our preliminary experimental results confirm the existence of optical activity in small pitch smectics C*. It is in particular to be noticed that the rotatory power is practically zero at normal incidence, namely for light propagating along the optical axis, and increases by increasing the incidence angle, in agreement with the model defined by Eq. 1. As a final comment, we observe that this rotatory power is not a small effect, and cannot be neglected in experiments with small pitch smectics C*. In fact in our sample the gyrotropy parameter $g_\perp$, for He-Ne light, is 5×10^{-3}, corresponding to a maximum rotation rate of nearly 1 degree per μm.

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