Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

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Online publication date: 29 June 2010

To cite this Article Giesselmann, F. , Langhoff, A. and Zugenmaier, P.(1997) 'Preliminary communication Dispersion of the optical axes in smectic C* liquid crystals', Liquid Crystals, 23: 6, 927 — 931
To link to this Article: DOI: 10.1080/026782997207885
URL: http://dx.doi.org/10.1080/026782997207885

LIQUID CRYSTALS, 1997, VOL. 23, NO. 6, 927–931

Preliminary communication

Dispersion of the optical axes in smectic C* liquid crystals

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(*Received 31 March 1997; accepted 27 June 1997*)

Optical axes dispersion denotes the dependence of the orientation of the indicatrix and the optical axes on the wavelength of light. Theory predicts optical axes dispersion in optically biaxial phases with low crystallographic symmetry, like the C_{2h}-symmetry of SmC or the C_2 symmetry of SmC* phases. The magnitude of this axes dispersion was measured electro-optically for two liquid crystal materials exhibiting SmC* phases using several wavelengths of light. Far below the phase transition temperature, the wavelength dispersion of the optical tilt is about 1–2° (5–10% of the total tilt) over the range of visible wavelengths.

The local symmetry of SmC and helically unwound SmC* phases corresponds to monoclinic C_{2h} and C_2 symmetry, respectively. The optical indicatrix is a triaxial ellipsoid with two circular sections and, hence, two privileged wave normal directions for which there is no double refraction. These are the optic axes of SmC or SmC*, and the phases are said to be *biaxial*. The biaxial indicatrix is described by three refractive indices n_α, n_β, and n_γ which define the lengths of the principal (semi-) axes of the indicatrix (figure 1). The optical biaxiality $\delta n = n_\beta - n_\alpha$ of SmC or SmC* phases was found to be small ($\approx 10^{-3}$) [1] and is often neglected, introducing a uniaxial approximation [2].

According to Neumann's principle of crystal physics [3], one of the three orthogonal principal axes of the indicatrix *must* coincide with the C_2 symmetry axis which defines the *b*-crystallographic axis of the SmC or SmC* structure. This is the one and only axis of the indicatrix which is fixed in space for every wavelength λ of the electromagnetic spectrum. The direction of the other two orthogonal axes of the indicatrix depends in general on the wavelength of light, i.e. the orientation of the indicatrix depends on the wavelength of light considered. With varying wavelength of light, the indicatrix is free to rotate around the C_2 symmetry axis (figure 2). This is a well-known phenomenon in crystal physics which is called *dispersion of the optic axes*, and it applies to all crystals of the monoclinic and triclinic system† [4].

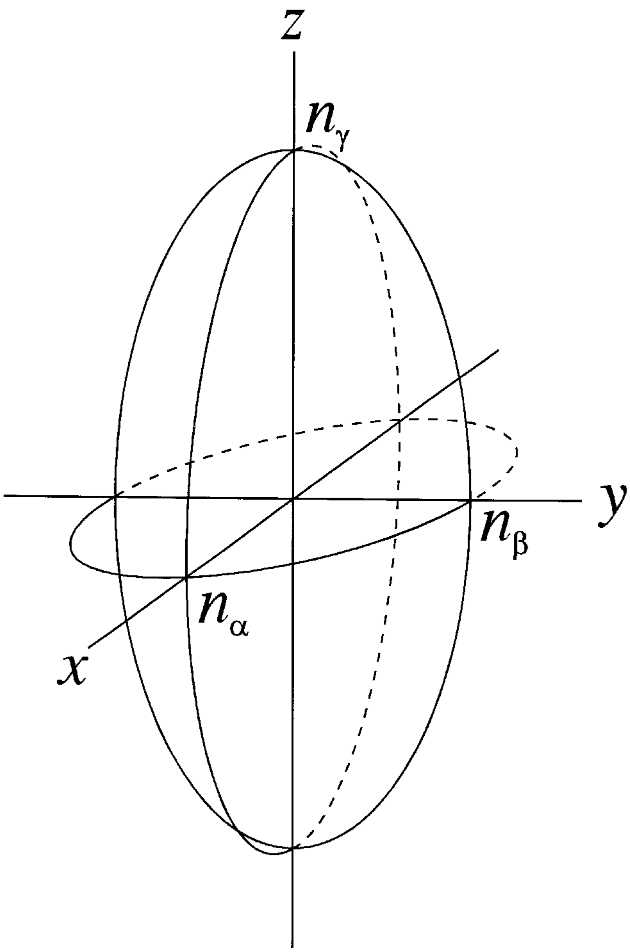

Figure 1. Indicatrix of an optically biaxial medium. The lengths of the principal semi-axes *x*, *y* and *z* are defined by the refractive indices n_α, n_β, and n_γ.

*Author for correspondence.

†In the orthorhombic system, the three principal axes of the indicatrix coincide with the three crystallographic axes and are fixed in space, whatever the wavelength [4].

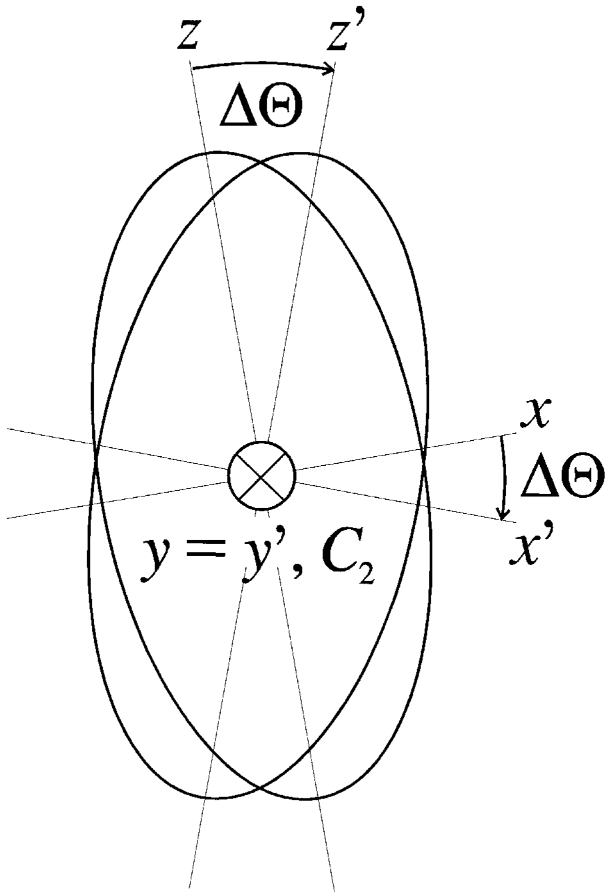

Figure 2. Axes dispersion in SmC and SmC* phases results in a tilt of the indicatrix around the C_2 symmetry axis by an angle $\Delta\Theta$ that depends on the wavelength of the light.

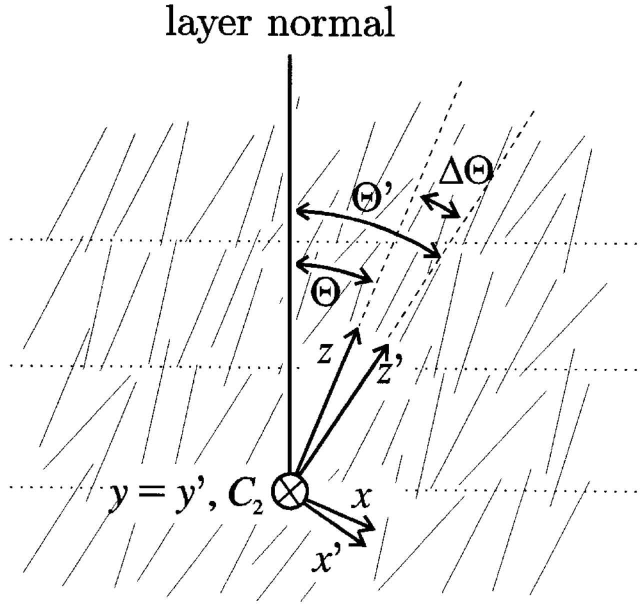

Figure 3. One of the principal axes of the indicatrix, which is perpendicular to the C_2 symmetry axis, defines the smectic director ($z=\hat{\mathbf{n}}$, $z'=\hat{\mathbf{n}}'$). Due to the axes dispersion, different optical director tilt angles Θ and Θ' are detected with red and blue light.

In liquid crystal physics, the optical director $\hat{\mathbf{n}}$ is related to the average direction of the molecular long axis and defines another principal axis of the indicatrix associated with an extreme value of the refractive index (usually the highest index of refraction). In SmC or SmC* phases the director is perpendicular to the C_2 axis which defines the only axis of the indicatrix that is fixed in space for every wavelength. According to the considerations of the preceding paragraph, the direction of the remaining orthogonal principal axes and, thus, the direction of the optical director depends on the wavelength of light in the case of SmC and SmC* liquid crystals (figure 3). The same holds for the director tilt angle Θ which is the angle included by the (optical) director $\hat{\mathbf{n}}$ and the smectic layer normal (figure 3).

So far, only symmetry has been considered in the prediction of optical axes dispersion for SmC or SmC* phases. Symmetry considerations do not allow prediction of the magnitude of a certain effect. In this study, we report the experimental detection of optical axes dispersion which is determined by wavelength dependence of the optical director tilt angle in ferroelectric SmC* phases where the tilt angle is easily measured by electro-optical switching experiments. The results also account for non-chiral SmC, since axes dispersion is an effect of the monoclinic symmetry and not a chirality effect.

In a helically unwound SmC* phase, the director is related to the long axis of the indicatrix and can be switched by an external electric field in a proper bookshelf geometry by twice the optical tilt angle Θ [5]. For our measurements we used an electro-optical technique which was first proposed by Bahr and Heppke [6]: the FLC sample is filled into a thin liquid crystal cell with ITO electrodes and switched in the surface-stabilized state with bookshelf configuration by an a.c. electric square field. The transmissions of up- and down-states of the spontaneous polarization between crossed polarizers are recorded for several wavelengths of light on rotating the cell around the direction of the light by an angle ϕ_o, which is the orientation of the cell with respect to the (fixed) polarization plane of the incident light. Under optically uniform and saturated switching conditions, the transmissions of the up- and down-states for a certain wavelength are given by two $\sin^2 2\phi$ functions, which exhibit a phase shift δ to each other of twice the optical tilt angle Θ for that wavelength (figure 4). The transmissions were measured simultaneously for multiple wavelengths of light using an optical multi-channel

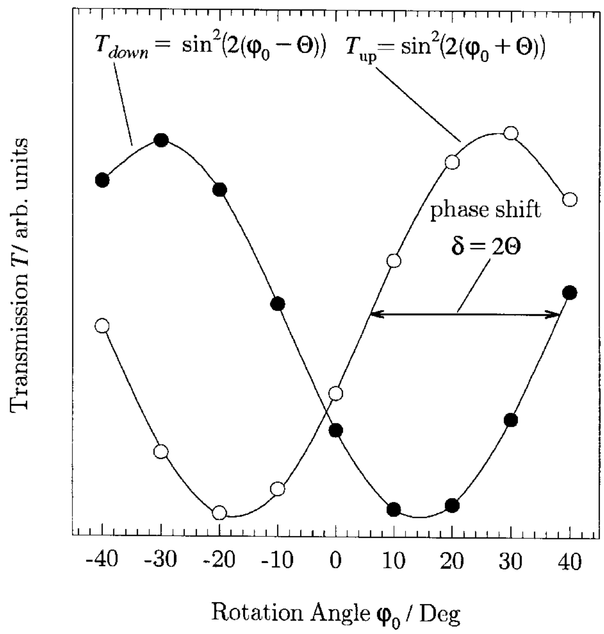

Figure 4. Determination of the optical director tilt angle Θ by electro-optical measurements.

analyser (LS 2000, Alton Instruments) which was mounted on the photo-tube of the polarizing microscope. Overall, the accuracy of this method can be estimated to $\pm 0{\cdot}2°$.

The measurements were carried out on the FLC compound 4{β-{-[(R)-2-chloro-3-methylbutanoyloxy]-phenyl}ethyl}phenyl 4-octyloxybenzoate (D8) with the mesophase sequence [7] (temperatures in °C):

SmF* 83·7 SmI* 92·9 SmC* 125·6 SmA* 129·2

TGBA* 129·4 N* 134·5 I

The isotropic compound was filled by capillarity into 2 μm cells from E. H. C., Tokyo, with transparent electrodes (ITO) and coated with an antiparallel rubbed polyimide alignment layer. The sample was aligned in the bookshelf geometry by switching the FLC using a high electric square field of 10–15 V μm^{-1} amplitude and 200 Hz frequency for several minutes. This high-field treatment [8] was repeated throughout the measurements after every change of temperature in order to avoid the reformation of chevrons due to variations of the smectic layer spacing with temperature. The removal of chevrons was checked by the elimination of the characteristic zig–zag defects during the high-field treatment. During the transmission measurements, the director was switched by an electric square field of 1 Hz frequency and 2 V μm^{-1} amplitude.

Similar measurements have been carried out for the commercial mixture FLC 6430 from Hoffmann-La Roche. The results obtained were comparable with those from D8 which are those reported here.

The wavelength dispersion of the optical tilt angle $\Theta(\lambda)$ is depicted in figure 5 for selected temperatures T. The results clearly establish a significant dependence of the optical tilt and, thus, a considerable degree of optical axes dispersion in the SmC* phase of D8 at temperatures below 122°C. The optical tilt angle increases with decreasing wavelength:

$$\frac{d\Theta}{d\lambda} < 0. \tag{1}$$

The extent of optical axis dispersion increases with

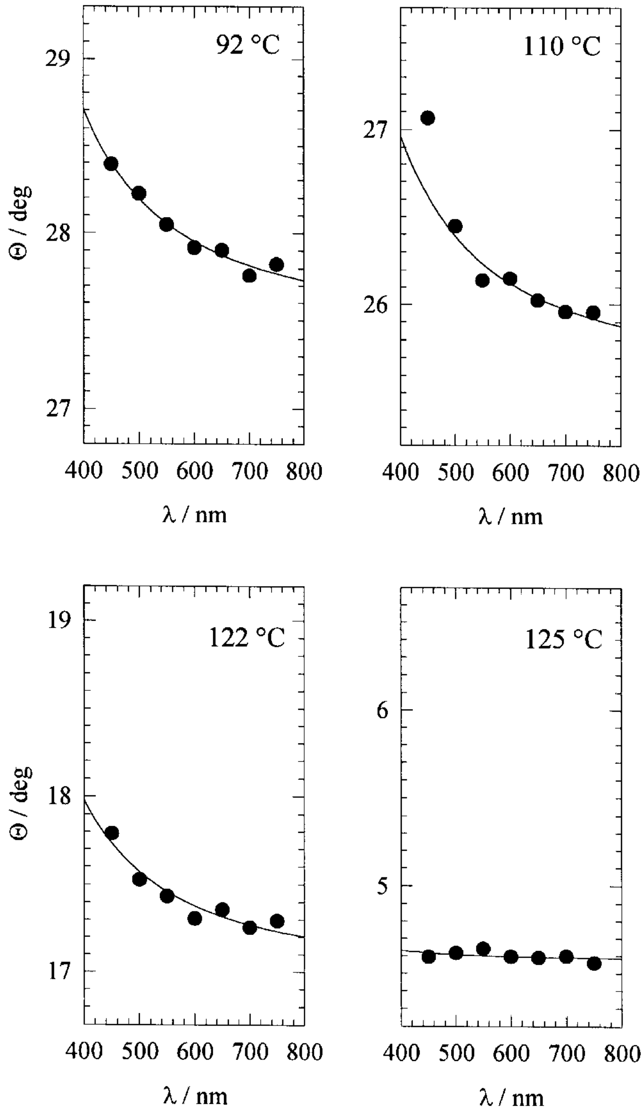

Figure 5. Dispersion of the optical tilt angle Θ with wavelength of light λ at selected temperatures for the SmC* phase of D8.

decreasing temperature and increasing tilt angle:

$$-\frac{d\Theta}{d\lambda} \propto \Theta(T). \qquad (2)$$

This is shown in figure 6 where the optical tilt angle measured for red (700 nm) and blue (400 nm) light is depicted versus the temperature T. Far below the phase transition temperature and at tilt angles of about 20–30°, the dispersion of the optical tilt is about 1·0–1·5° over the range of visible wavelengths, which is about 5% of the total tilt. An even higher dispersion (≈10% of the total tilt) was obtained for FLC 6430.

The tilt angle dependence [equation (2)] of the dispersion of the indicatrix may be explained in the following way. The tilt is considered as an order parameter which measures the change in symmetry during the transition from the uniaxial SmA/SmA* phase to the biaxial SmC/SmC* phase. Thus, the tilt provides a direct measure of biaxiality. From the molecular point of view, the tilt affects the biasing of rotation around the molecular long axis, which is also reflected by the biaxiality. The sign of the optical tilt dispersion [equation (1)] is related to the UV absorption of the mesogenic cores. If the wavelength decreases from red to blue, the absorption frequency of the mesogenic cores is approached more and more closely, and the direction of maximum refractive index, which defines the long axis of the indicatrix and the optical director, approaches the direction of the mesogenic cores. According to the Boulder model [9], the tilt of the mesogenic cores is higher than the tilt of the complete molecule, which explains why the optical tilt is higher for blue than for red light.

As a result of the monoclinic symmetry, the optical properties of SmC and SmC* liquid crystals are affected through (1) the optical biaxiality $\delta n = n_\beta - n_\alpha$, which was shown to be negligibly small at optical frequencies [1], and (2) the optical axes dispersion, which is now observed to the considerable extent of about 5% of the optical tilt angle, at least for the compounds investigated in this study. It is interesting to note that axes dispersion seems to be the more pronounced expression of biaxiality in SmC* liquid crystals at optical frequencies. The observed axes dispersion is relevant to the theory and application of ferroelectric liquid crystals, whilst taking into account the following considerations.

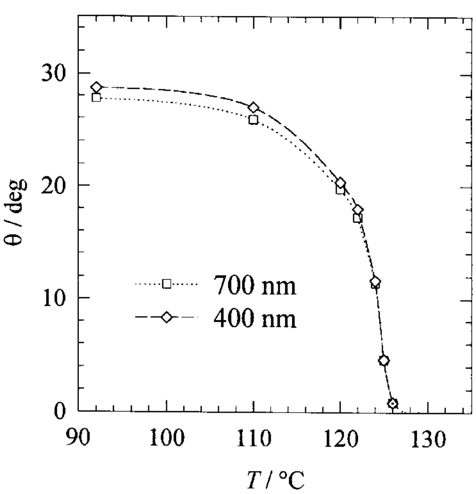

Figure 6. Optical tilt angle Θ of D8 for red (λ=700 nm) and blue (λ=400 nm) light as a function of temperature T.

(1) It is well-known, that the director determined by X-ray experiments differs from the optical director; the situation becomes even more confused if the optical director itself depends on the wavelength of light. The existing director concept seems to be oversimplified in the case of monoclinic biaxial phases.
(2) The choice of the optical tilt angle as the primary order parameter of SmC or SmC* phases is unsatisfactory. The free energy is usually expanded in terms of the (optical) tilt in a Landau-type description; hence the free energy does not depend on the wavelength, but the optical tilt does, and the expansion coefficients have also to depend on the wavelength of light.
(3) FLC materials are characterized by their optical tilt. If the desired accuracy should be less than about 1°, monochromatic light with a specified wavelength has to be used.
(4) Complete extinction of a ferroelectric liquid crystal display between crossed polarizers can, in principle, only be achieved for a single wavelength, leading to a reduction of the contrast and involving chromatic deviations. Nevertheless, a shorthand calculation shows that the effect has no relevance in most cases: if the orientation of the indicatrices for red and blue light differs by $\Delta\Theta = 1°$ and the red light is set to extinction, the transmission for blue light is about $\sin^2(2\Delta\Theta) \approx 0{\cdot}1\%$.

The consequences of the tilt angle dispersion on the order parameter and the Landau theory of SmC liquid crystals will be considered in a subsequent paper.

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