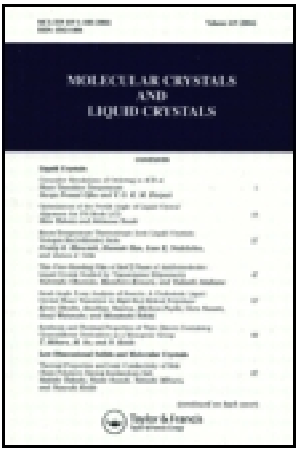

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Determination of the Dielectric Tensor of a Chiral Smectic Liquid Crystal†

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The temperature dependencies of the principal values of the dielectric constant tensor ε_1, ε_2, ε_3; the helical pitch P and the tilt angle θ of a chiral smectic phase have been obtained (for the first time) from measurements of the temperature dependencies of the refractive indices for linearly and circularly polarized light and from the wavelength of the Bragg reflection maximum.

Keywords: chiral smectic, dielectric tensor, helical pitch, refractive index

INTRODUCTION

For the determination of the dielectric tensor it is necessary to find the principal values, ε_1, ε_2, ε_3 of the dielectric tensor, the pitch, P and director tilt angle, θ.[1,2] In the cholesteric phase $\theta = \pi/2$ and hence the determination of the dielectric tensor becomes simple. The temperature dependencies of the principal values of the dielectric tensor of the cholesteric phase have been found (assuming that $\varepsilon_1 = \varepsilon_2$).[3] Up to now, there has been no efficient method for the complete determination of the dielectric tensor of a chiral smectic phase. However some separate measurements have been carried out. Particularly, the tilt angle can be obtained from X-ray diffraction studies and according to Ref. 4 lies in the range 35°–45°.

†Paper presented at the 11th International Liquid Crystal Conference, Berkeley, CA, 30 June–4 July, 1986.

An approach for the direct determination of the ε_1, ε_2, ε_3, θ values for the chiral smectic phase using only optical measurements, is presented in this work.

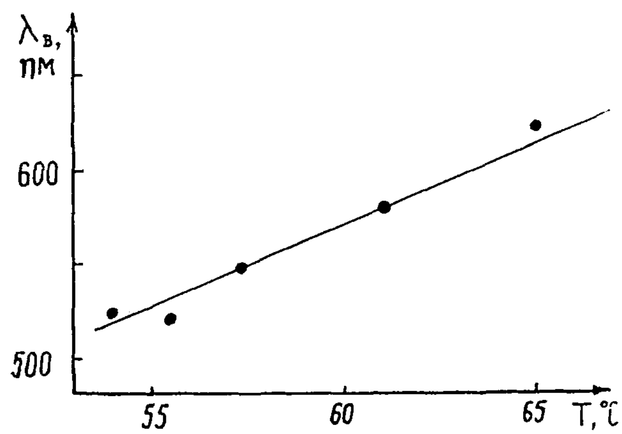
1. THEORY

First we shall give expressions which enable value for ε_1, ε_2, ε_3, P, θ to be obtained from the experimentally determined values: n_{oc}, n_{ec}—ordinary and extraordinary refractive indices, n_R, n_L—refractive indices for right and left polarised light, respectively, λ_B—Bragg wavelength. The equations connecting ε_1, ε_2, ε_3, P, θ with n_{oc}, n_{ec}, n_R, n_L, λ_B can be written in the form:

$$\lambda_B = P\sqrt{\bar{\varepsilon}} \tag{1}$$

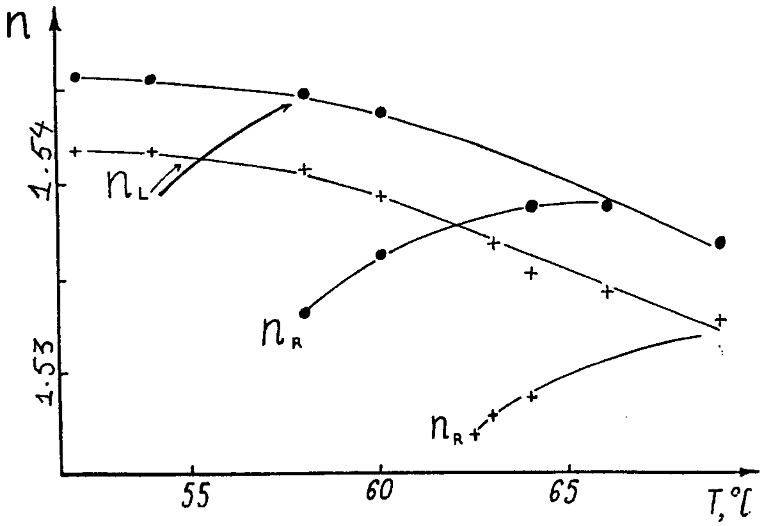
$$\bar{\varepsilon} = \frac{n_R^2 + n_L^2}{2} + \frac{\lambda}{P}(n_L - n_R) \tag{2a}$$

$$\bar{\varepsilon} = \frac{1}{2}\left(\varepsilon_1 + \frac{\varepsilon_2\varepsilon_3}{\varepsilon_2 \sin^2\theta + \varepsilon_3 \cos^2\theta}\right) \tag{2b}$$

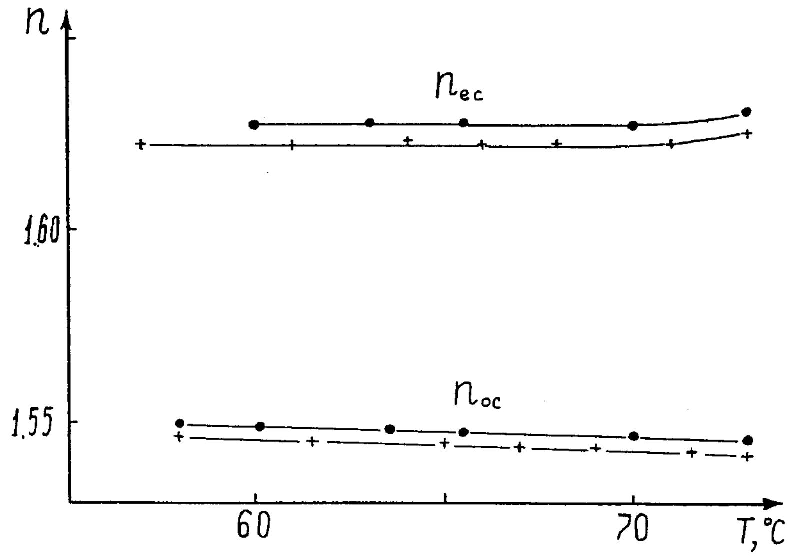


(a)

FIGURE 1 Temperature dependencies of (a) wavelength of the Bragg reflection maximum λ_B, (b) refractive indices for right n_R and left n_L circularly polarised light, (c) refractive indices for the ordinary n_{oC} and extraordinary n_{eC} waves for a chiral smectic phase measured in the chiral smectic phase of liquid space crystalline CE-3.



(b)



(c)

FIGURE 1 (continued)

● $-\lambda = 546$ nm, + $-\lambda = 577$ nm

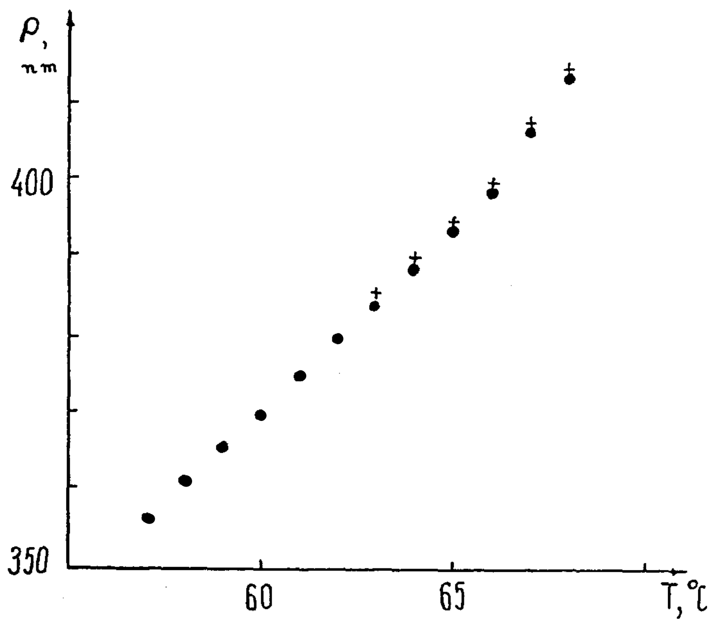
$$\varepsilon_a = \sqrt{\bar{\varepsilon}}\left\{\left[\frac{n_L^2 - n_R^2}{2\bar{\varepsilon}} + \frac{\lambda}{P\bar{\varepsilon}}(n_L + n_R)\right]^2 - \frac{4\lambda^2}{P^2\bar{\varepsilon}}\right\}^{1/2} \quad (3a)$$

$$\varepsilon_a = \frac{1}{2}\left(\varepsilon_1 - \frac{\varepsilon_2\varepsilon_3}{\varepsilon_2 \sin^2\theta + \varepsilon_3 \cos^2\theta}\right) \quad (3b)$$

$$n_{oc}^2 = \frac{1}{2}(\varepsilon_1 + \varepsilon_2 \cos^2\theta + \varepsilon_3 \sin^2\theta) \quad (4)$$

$$n_{\varepsilon c}^2 = \varepsilon_2 \sin^2\theta + \varepsilon_3 \cos^2\theta \quad (5)$$

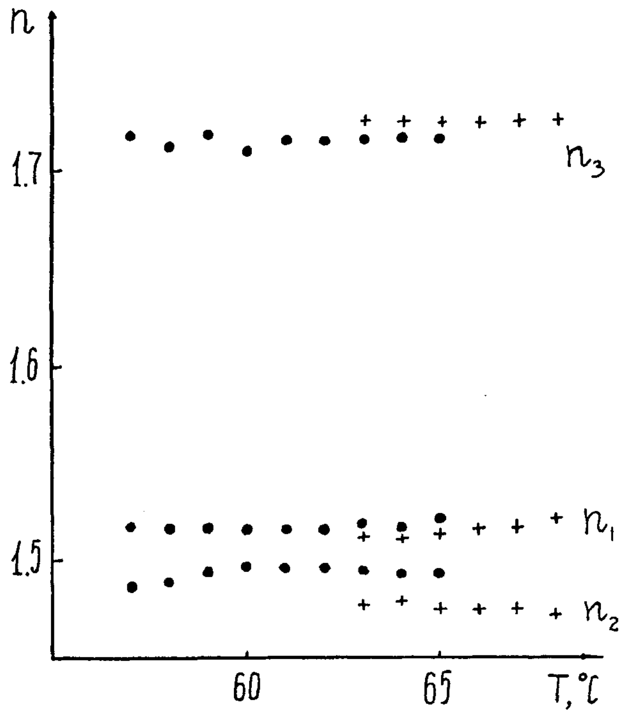
where ε_a and $\bar{\varepsilon}$—are the anisotropy and the mean value of the dielectric permittivity, respectively. These equations are obtained from the formulas given in Refs. 1–3, 5 by simple transformations. Equations (4) and (5) are valid if $P < \lambda$, where we are far from the Bragg



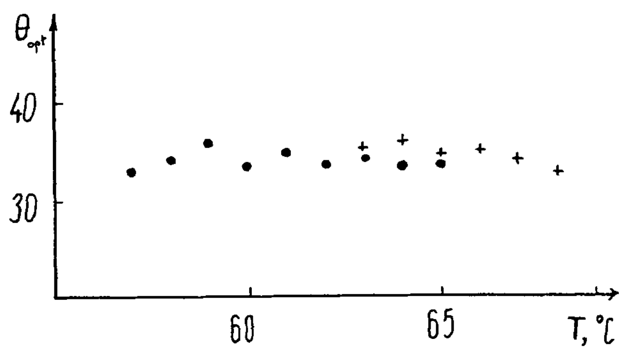
(a)

FIGURE 2 Temperature dependencies for the (a) helical pitch P, (b) principal values of the dielectric constant tensor $n_1 = \sqrt{\varepsilon_1}$, $n_2 = \sqrt{\varepsilon_2}$, $n_3 = \sqrt{\varepsilon_3}$, (c) tilt angle, θ calculated for the chiral smectic phase of liquid crystalline CE-3.

● $-\lambda = 546$ nm, + $-\lambda = 577$ nm



(b)



(c)

FIGURE 2 (continued)

● $-\lambda = 546$ nm, + $-\lambda = 577$ nm

reflection band. The solution of equations (1)–(5) gives the following expressions:

For the helical pitch:

$$P = \frac{\lambda\,(n_R - n_L) + [\lambda^2(n_R - n_L)^2 + 2(n_R^2 + n_L^2)\lambda_B^2]^{1/2}}{n_R^2 + n_L^2} \tag{6}$$

For principal values of the dielectric tensor:

$$\varepsilon_1 = \bar{\varepsilon} + \varepsilon_a \tag{7}$$

$$\varepsilon_2 = B - \sqrt{B^2 - A} \tag{8}$$

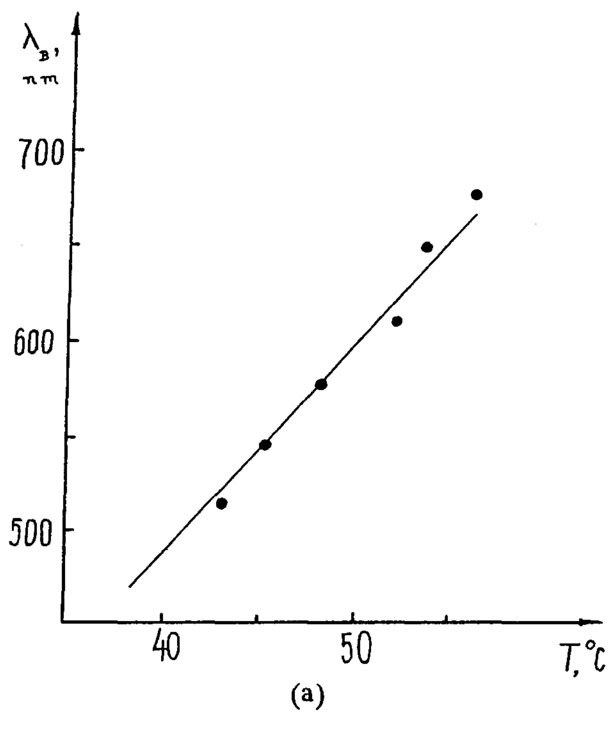


FIGURE 3 Temperature dependencies of (a) wavelength of the Bragg reflection maximum λ_B, (b) refractive indices for right n_R and left n_L circularly polarised light, (c) refractive indices for ordinary n_{oC} and extraordinary waves n_{eC} for a chiral smectic phase measured in the chiral smectic phase of the mixture 90% CE-3 + 10% TDO-BAMBCC.

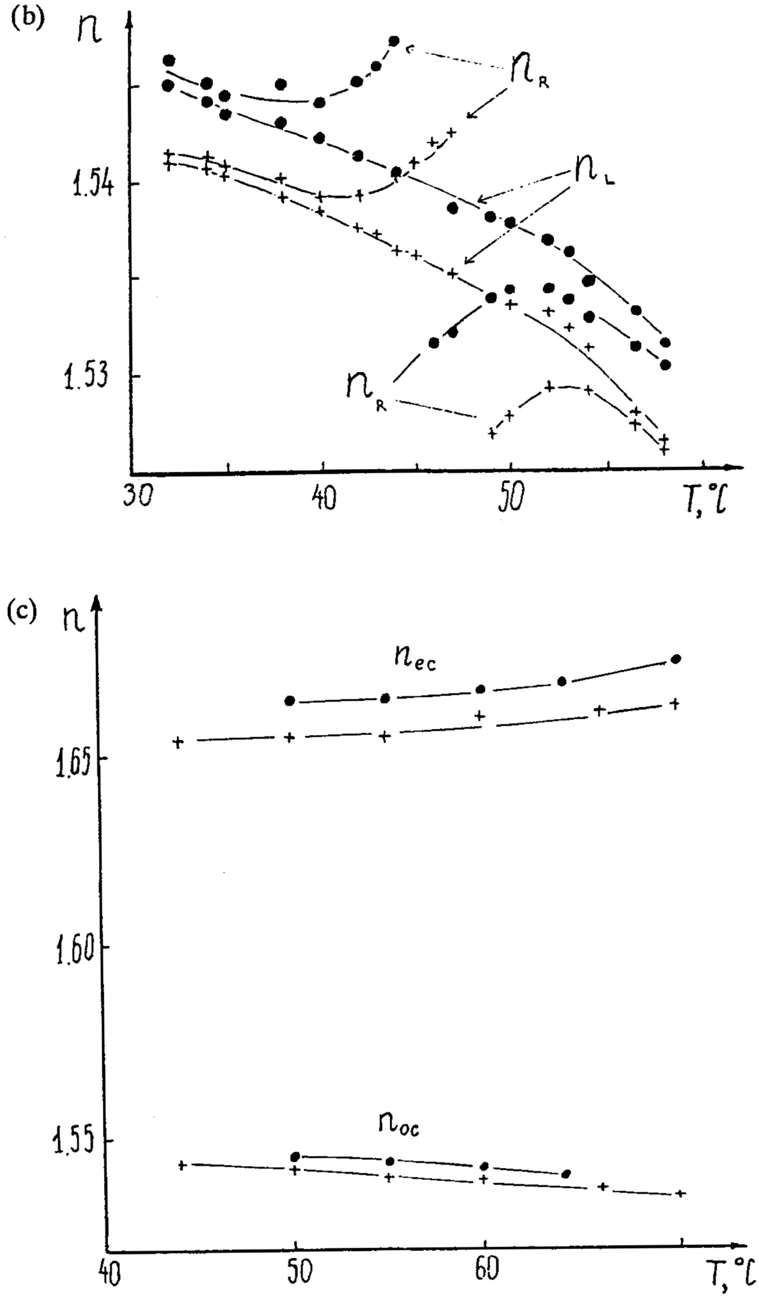
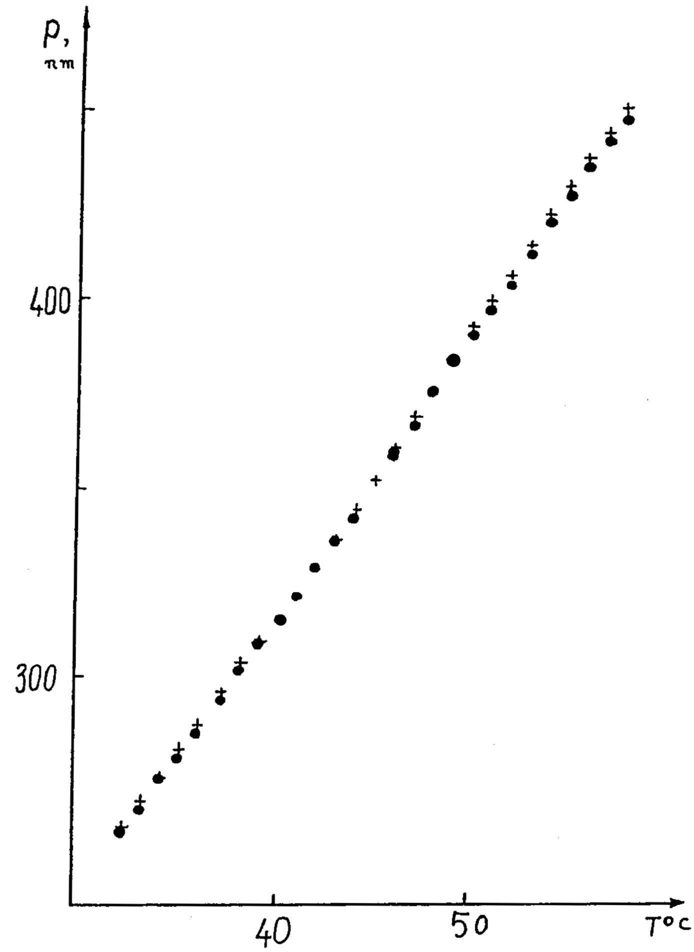


FIGURE 3 (continued)

● $-\lambda = 546$ nm, + $-\lambda = 577$ nm

$$\varepsilon_3 = B + \sqrt{B^2 - A} \tag{9}$$

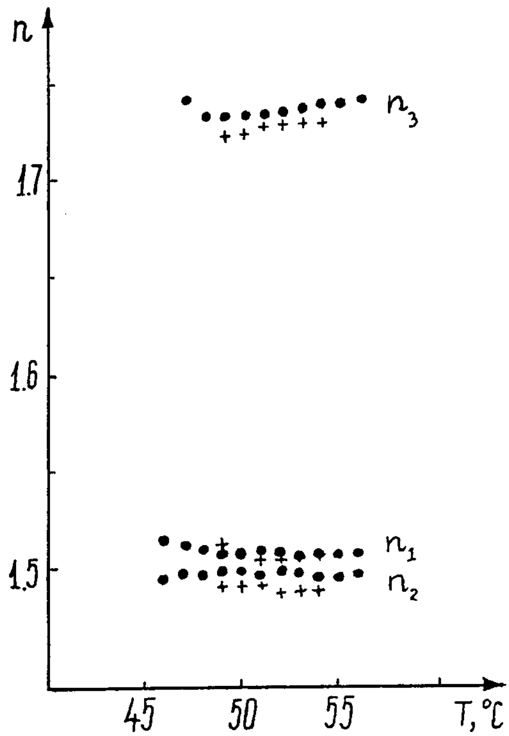
where $A = n_{ec}^2(\bar{\varepsilon} - \varepsilon_a)$; $B = \frac{1}{2}(2n_{oc}^2 + n_{ec}^2 - \bar{\varepsilon} - \varepsilon_a)$ and $\bar{\varepsilon}$ and ε_a are calculated from (2b) and (3b).



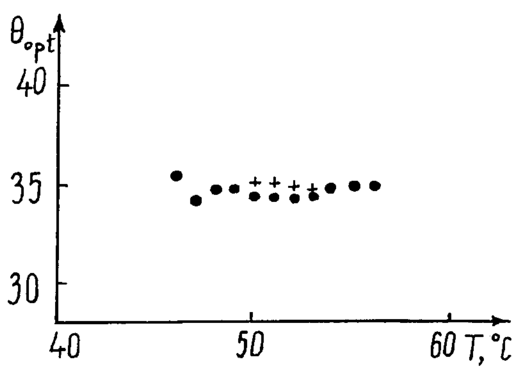
(a)

FIGURE 4 Temperature dependencies for (a) helical pitch P, (b) principal values of the dielectric constant tensor $n_1 = \sqrt{\varepsilon_1}$, $n_2 = \sqrt{\varepsilon_2}$, $n_3 = \sqrt{\varepsilon_3}$, (c) tilt angle, θ calculated for the chiral smectic phase of the mixture 90% CE-3 + 10% TDO-BAMBCC

● – λ = 546 nm, + –λ = 577 nm



(b)



(c)

FIGURE 4 (continued)

● $-\lambda = 546$ nm, + $-\lambda = 577$ nm

For tilt angle

$$\theta = \arcsin\left(\frac{n_{ec} - \varepsilon_3}{\varepsilon_2 - \varepsilon_3}\right)^{1/2} \tag{10}$$

Using expressions (6)–(10) we obtain the temperature dependencies ε_1, ε_2, ε_3, P, θ of a chiral smectic phase.

2. EXPERIMENTAL RESULTS

The chiral smectic phase of the 4-p-hexyloxyphenyl ester of 4′-(2″-methylbutyl) biphenyl-4-carboxylic acid (CE-3) gives a convenient selective reflection in the visible range and this phase was therefore chosen for study. Selective light reflection in the chiral smectic phase was studied experimentally for both normal and oblique incidence I4,6-9I. A previous attempt to obtain information about the dielectric tensor from reflection spectra is described in Ref. (10). But it was not possible to determine precisely the complete set of optical parameters (ε_1, ε_2, ε_3, P, θ). The helical structure prevents the direct measurements of refractive indices, as had been noted in Ref. (11).

In the present work, measurements of λ_B, n_o, n_e, n_L, n_R were carried out to calculate ε_1, ε_2, ε_3, P, θ. Temperature dependence of these values are given in Fig. 1. Bragg wavelength is determined from data of selective reflection; n_L and n_R are measured by a goniometer using the wedge method described in Ref. (3), and n_{oc}, n_{ec}—by an Abbè refractometer. All measurements were made on the chiral smectic phase.

Temperature dependencies of pitch P, of indices of refraction $n_1 = \sqrt{\varepsilon_1}$, $n_2 = \sqrt{\varepsilon_2}$, $n_3 = \sqrt{\varepsilon_3}$ and of tilt angle θ are shown in Fig. 2a, Fig. 2b, Fig. 2c respectively. Similar dependencies, measured and calculated for a mixture 90% CE-3 + 10% TDOBAMBCC, are presented in Fig. 3 and Fig. 4 (TDOBAMBCC—P tetradecyloxy-benzylidene P′-amino 2 methyl butyl α cyano cinnamate).

3. DISCUSSION

The results obtained from this work show that the complete dielectric tensor of a chiral smectic phase (ε_1, ε_2, ε_3, P, θ) can be determined easily from optical measurements and that this method gives higher precision than alternative methods.[10] The suggested approach allows us to measure the local biaxility of a chiral smectic phase which was not possible before.

For both pure CE-3 and for a mixture, θ is independent of temperature over the whole temperature range investigated. This result agrees with de Vries' statement, that for smectic liquid crystals without a smectic A phase, such as CE-3, $\frac{d\theta}{dT} = 0$.[12]

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