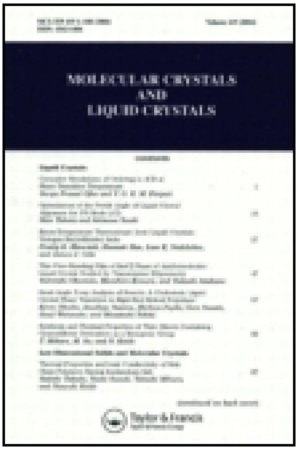

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Dielectric Biaxiality in S_C Host Systems

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AC field stabilisation of ferroelectric S_C* liquid crystals with the chevron layer geometry requires that there is a significant dielectric biaxiality.[1,2] To investigate this, the electric permittivities of an S_C host mixture based on the phenyl pyrimidines and exhibiting an N-S_A-S_C phase sequence are reported. A large relaxation of $\varepsilon_{\parallel}$ occurred, which caused a sign reversal of the dielectric anisotropy with a cross-over frequency of 25 kHz at 25°C. The host electro-optic behaviour was then characterised using extinction angle measurements as a function of applied voltage at frequencies corresponding to $\Delta\varepsilon > 0$, $\Delta\varepsilon \approx 0$ and $\Delta\varepsilon < 0$, and the results explained using a tilted layer model with a biaxial dielectric tensor. The magnitude of the S_C dielectric biaxiality, the layer tilt angle in the planar homogeneous geometry and the cone angle are determined. Measurements for several other hosts commonly used in commercial FLC mixtures are reported and it is shown that in each case the dielectric biaxiality is sufficiently large to cause AC field stabilisation in ferroelectric liquid crystals.

1. INTRODUCTION

The smectic layers of a Surface Stabilised Ferroelectric Liquid Crystal (SSFLC) form a chevron structure[3] (Figure 1a), with a ratio of layer tilt (δ) and S_C* cone angle (θ) which is independent of temperature and typically of magnitude 0.9. This results in a director configuration which is approximately uniform in the surface stabilised state,[4,5] and a white light extinction angle measured from the alignment direction β_0 given by:

$$\cos \beta_0 = \cos \theta / \cos \delta \tag{1}$$

For a typical cone angle of 22.5° and layer tilt of 20° the extinction angle of the zero-field state is 10°.

Application of an electric field which couples to the ferroelectric polarisation P_S causes the director to rotate about the cone, increasing the extinction angle towards the fully switched value β_s[6]:

$$\tan \beta_s = \tan \theta / \cos \delta \tag{2}$$

which is 24° for the typical values of θ and δ used above. The fully switched state is illustrated in Figure 1b. The director is again approximately uniform but tilted out of the plane of the cell by +δ and −δ in either chevron half, except for thin regions close to the surfaces and chevron interface which remain unswitched.[7] On removal of the field, the director relaxes back to the initial state due to the elastic effect of these unswitched regions.

The degradation of optical bistability that this causes may be avoided by AC field stabilisation,[8] a process whereby the switched states are maintained after the switching pulse through application of a high frequency AC field. Previous models of the SSFLC state, such as the bookshelf layer model, included a large amount of tilt of the director out of the cell plane in the relaxed state.[9] The high frequency AC field coupled to the negative dielectric anisotropy of the $S_C{}^*$ material causing the director to rotate about the cone until the out-of-plane tilt was removed and the fully switched state stabilised. With a chevron layer structure, on the other hand, the AC field should stabilise the relaxed state, which is the opposite effect to that observed experimentally. This contradiction has been resolved using dielectric biaxiality of the $S_C{}^*$ phase.[1,2] It is shown in the present work that the measured biaxiality is sufficiently large to cause the AC field effects for several host mixtures. Achiral host mixtures were used throughout so that the coupling between the AC field and the permittivities could be studied without the complications of ferroelectric switching of the director. Differences between the host S_C

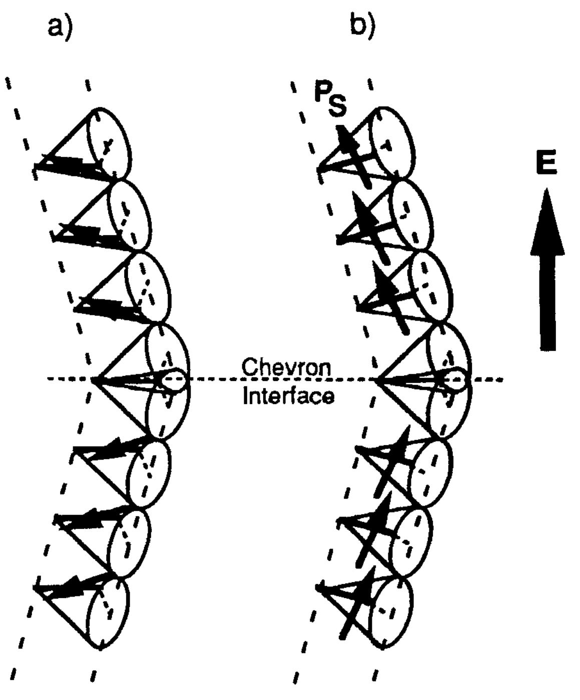


FIGURE 1 The S_C(*) chevron layer geometry in a) the relaxed and b) fully switched states.

and ferroelectric S_C* are otherwise unimportant for the present work, and this is represented by S_C(*) where appropriate.

2. THE S_C(*) COORDINATE GEOMETRY

Choosing the S_C(*) principal axes parallel to the director (ε_3), parallel to the C_2 symmetry axis (ε_2) and orthogonal to both of these (ε_1) as shown in Figure 2a, the permittivity may be written in the form[6]:

$$\varepsilon = \varepsilon_1 + \Delta\varepsilon \cdot \sin^2\zeta + \partial\varepsilon \cdot \cos^2\delta \cdot \cos^2\varphi \qquad (3)$$

where ζ is the out-of-plane tilt (Figure 2b), and φ the azimuthal angle of the projection of the director onto the layer. $\varphi = 0$ is chosen to coincide with the azimuthal angle of the fully switched director. The dielectric anisotropies $\Delta\varepsilon$ and $\partial\varepsilon$ are defined by:

$$\Delta\varepsilon = \varepsilon_3 - \varepsilon_1; \quad \partial\varepsilon = \varepsilon_2 - \varepsilon_1 \qquad (4)$$

The in-plane tilt angle β is defined as the angle between the alignment direction and the projection of the director into the cell plane (Figure 2b), and is given by:

$$\tan\beta = \frac{\cos\varphi \cdot \sin\theta}{\sin\delta \cdot \sin\varphi \cdot \sin\theta + \cos\delta \cdot \cos\theta} \qquad (5)$$

Assuming that the director profile is approximately uniform at all field strengths then β corresponds to the measured extinction angle.

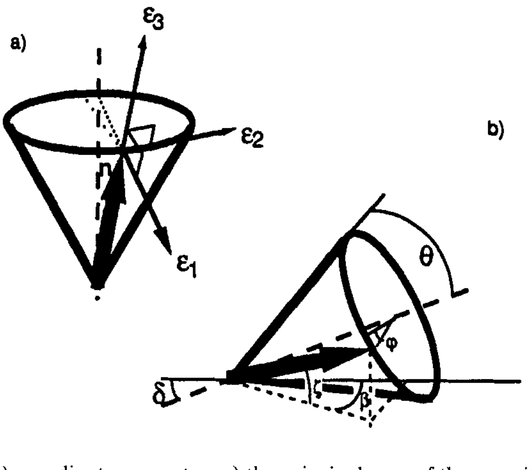
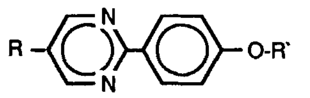


FIGURE 2 The S_C(*) coordinate geometry: a) the principal axes of the permittivity and b) definition of the angular quantities.

3. DIRECT OBSERVATION OF DIELECTRIC BIAXIALITY

The effect of varying $\Delta\varepsilon$ from positive to negative was investigated using a two-frequency host[1,2] based on the phenyl pyrimidines (PYP)[10]:



R, R' alkyl groups

I 72 N 70 S_A 55 S_C 18 K (°C)

The permittivities measured in planar homogeneous ($\varepsilon_\perp$) and homeotropic ($\varepsilon_\parallel$) alignment cells are shown as a function of temperature for several frequencies in Figure 3. At 25°C, the dielectric anisotropy changes sign (i.e. $\Delta\varepsilon = 0$) at 25 kHz. Figure 4 shows the change in extinction angle with RMS voltage for frequencies corresponding to $\Delta\varepsilon > 0$ (1 kHz), $\Delta\varepsilon = 0$ (25 kHz) and $\Delta\varepsilon < 0$ (100 kHz).

A maximum extinction angle was observed for frequencies where $\Delta\varepsilon > 0$ as the director moves from close to the bottom of the cone at V = 0, through the maximum at the side of the cone, and towards zero as the cone apex is approached, for which the permittivity component parallel to the applied field is largest. For typical values of θ and δ, the maximum extinction angle roughly corresponds to that predicted by Equation 2. Simultaneous solution of Equations 1 and 2 allows θ and δ to be determined, as shown in Figure 5.

The AC field induced electro-optic effect at the frequency for which $\Delta\varepsilon = 0$ requires that the S_C is biaxial and that ε_2 is greater than ε_1. Thus, the AC field couples to the biaxial permittivity tensor to maximise the component of ε_2 parallel

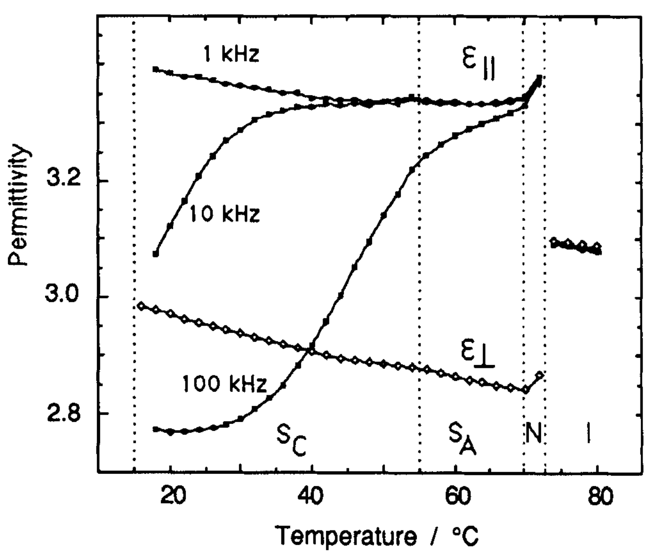


FIGURE 3 Temperature dependences of the homeotropic ($\varepsilon_\parallel$) and planar homogeneous ($\varepsilon_\perp$) permittivities for the PYP host at several frequencies.

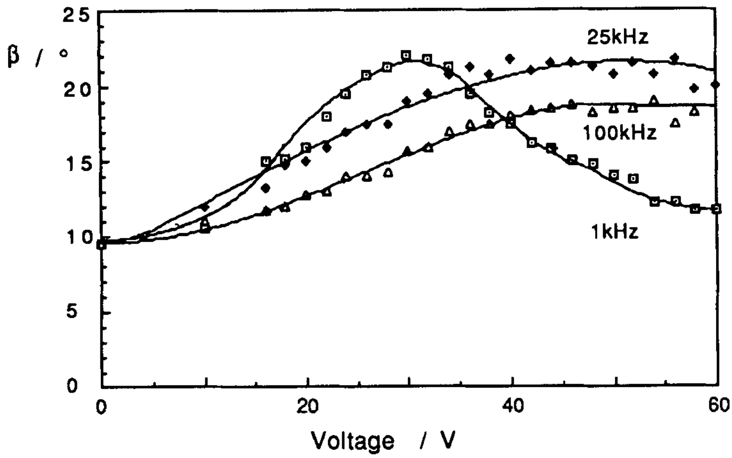


FIGURE 4 Extinction angle vs. RMS voltage at frequencies corresponding to $\Delta\varepsilon > 0$ (1 kHz), $\Delta\varepsilon \approx 0$ (25 kHz), and $\Delta\varepsilon < 0$ (100 kHz).

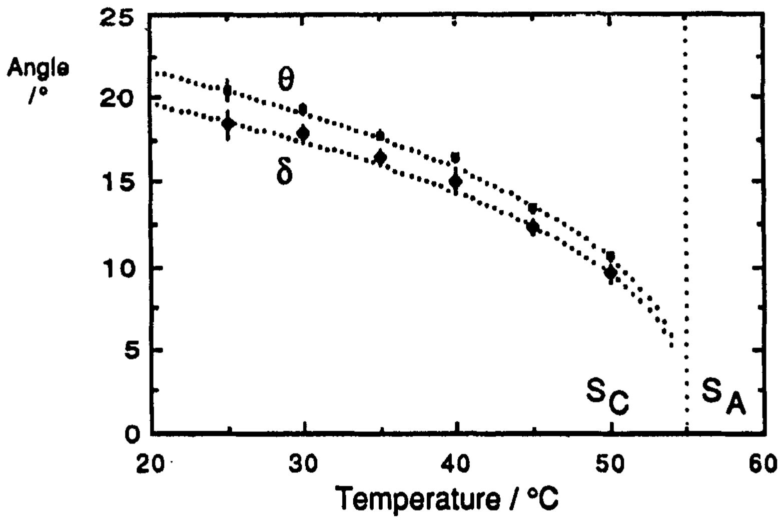


FIGURE 5 Cone and layer tilt angles for the PYP host determined from the extinction angle measurements of Figure 4.

to the field. Figure 4 shows that the electro-optic effect is larger at the frequencies for which $\Delta\varepsilon = 0$ than for $\Delta\varepsilon < 0$. This is because the applied field couples to the negative $\Delta\varepsilon$ opposing the out-of-plane tilt which is induced as the director is switched by the effect of the biaxiality. In this manner, the AC field behaviour of negative S_C and S_C* materials arises from the competition between the uniaxial and biaxial anisotropies, $\Delta\varepsilon$ and $\partial\varepsilon$. It is important, therefore, to measure the three S_C(*) permittivities and to show that the biaxiality is sufficiently large to overcome the effect of the negative $\Delta\varepsilon$.

4. MEASUREMENT OF THE THREE S_C PERMITTIVITIES

The $S_C(^*)$ director configuration of the planar homogeneous and homeotropic geometries are represented in Figure 6a and b, respectively. The measured permittivities ε_p and ε_h are then[6]:

$$\varepsilon_p = \varepsilon_2 - \partial\varepsilon \sin^2\delta/\sin^2\theta \tag{6}$$

and

$$\varepsilon_h = \varepsilon_1 + \Delta\varepsilon \cos^3\theta \tag{7}$$

where values of θ and δ were taken from Figure 5. Determination of three permittivities is made possible using the average permittivity $\bar{\varepsilon}$ defined as:

$$\bar{\varepsilon} = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3, \tag{8}$$

and found by linear extrapolation from the uniaxial nematic and smectic A phases, (where $\varepsilon_1 + \varepsilon_2 = 2\varepsilon_\perp$ and $\varepsilon_3 = \varepsilon_\parallel$).

The results for the PYP host are shown in Figure 7. The permittivities for this and other host systems commonly used in ferroelectric liquid crystal mixtures taken from Reference 6 are quoted in Table I at the reduced temperature T_{AC}-T = 30°C. Recent optical measurements[11] of a ferroelectric S_C^* mixtures based on one of these hosts predict that the magnitude of ∂ε which causes AC field stabilisation is similar to that of Table I.

5. THE IMPORTANCE OF DIELECTRIC BIAXIALITY

Assuming that the applied voltage induces motion of the director about the cone through changes of azimuthal angle φ and without change in either δ or θ, then at high voltages the azimuthal angle tends towards φ_∞:

$$\sin \varphi_\infty = \frac{\tan \delta \cdot \cos \theta \cdot \sin \theta}{\sin^2\theta - \partial\varepsilon/\Delta\varepsilon} \tag{9}$$

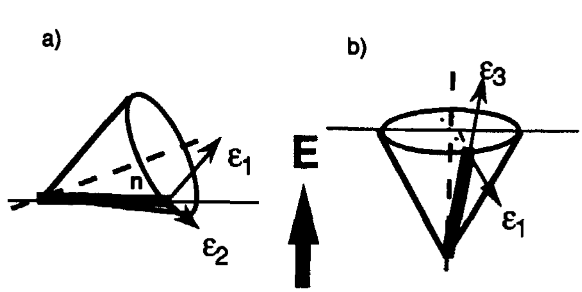


FIGURE 6 Schematic diagrams of the director configuration in a) planar homogeneous and b) homeotropic alignment geometries.

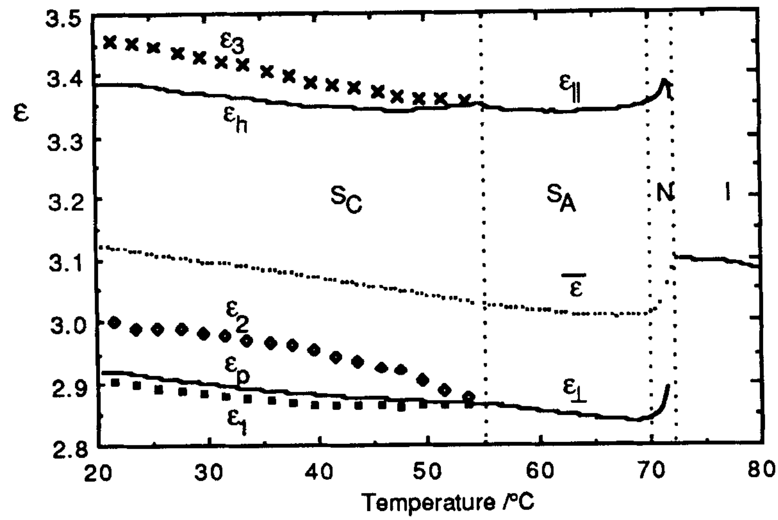
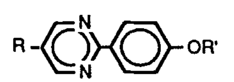
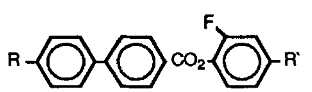
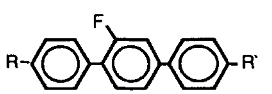
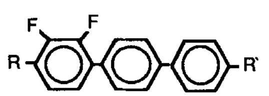


FIGURE 7 Temperature dependence of the PYP biaxial permittivities.

TABLE I

Permittivities for four typical host mixtures at T_{AC}-T = 30°C[6]

Host Structure	Acronym	ε_1	$\Delta\varepsilon$	$\partial\varepsilon$	$\Delta\varepsilon/\partial\varepsilon$
	PYP	2.94	+0.45	+0.11	+4.1
	MBF	3.78	-0.58	+0.24	-2.4
	FTP	3.13	+0.08	+0.13	+0.6
	DiFTP	4.62	-1.59	+0.86	-1.8

Substituting values for φ_∞ into Equation 5 gives β_∞, which corresponds to the extinction angle of the fully AC field stabilised state and is plotted as a function of $\Delta\varepsilon/\partial\varepsilon$ in Figure 8. With large positive anisotropies, the director is at equilibrium at the top of the cone, and $\beta_\infty = 0$. For low, positive values of $\Delta\varepsilon/\partial\varepsilon$, the role of

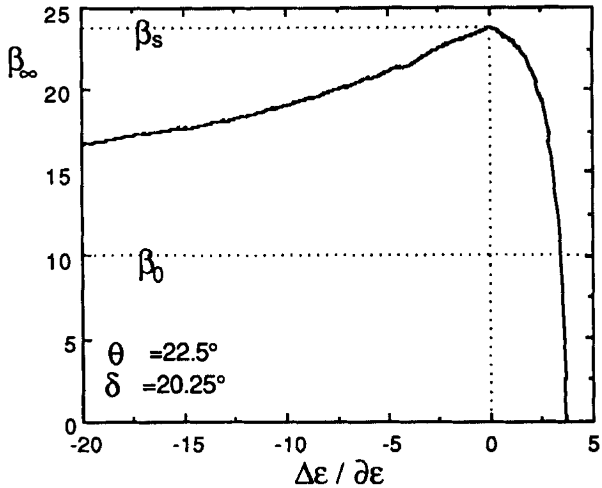


FIGURE 8 The importance of dielectric biaxiality. Even for large negative values of $\Delta\varepsilon/\partial\varepsilon$ significant AC field stabilisation is possible.

the biaxiality is more important and β_∞ rises sharply. For zero and negative $\Delta\varepsilon$ the AC field effect is dominated by the biaxiality, and a significant degree of field stabilisation occurs even for a $\Delta\varepsilon$ which is many times greater than $\partial\varepsilon$. This is because, where δ/θ is close to unity, relatively large changes of β may occur without inducing significant out-of-plane tilt.

The director orientation which maximises the permittivity is related to the ratio $\Delta\varepsilon/\partial\varepsilon$ and allows the extinction angle of the fully switched state to be predicted. For a given voltage, this director orientation depends on the magnitudes of $\Delta\varepsilon$ and $\partial\varepsilon$, and on the relevant elastic constants. Without an adequate continuum theory for the $S_C(^*)$ phase, however, the electro-optic characteristic cannot be modelled fully.

6. CONCLUSIONS

Dielectric biaxiality plays an important role in the electro-optic behaviour of both the S_C and S_C^* phases. Optimisation of $\Delta\varepsilon$ and $\partial\varepsilon$ is desirable for their successful application in FLC devices. For example, materials with large positive $\partial\varepsilon$ will AC field stabilise at lower voltages. It is likely that $S_C(^*)$ materials with large negative $\Delta\varepsilon$ will also exhibit large positive $\partial\varepsilon$ because both anisotropies are related to the magnitude of the transverse molecular dipole moment. The adverse effect of a negative $\Delta\varepsilon$ on AC field stabilisation is small compared with the advantageous effect of an increased dielectric biaxiality. Therefore, continued effort is required to develop suitable materials with large transverse molecular dipole moments.

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