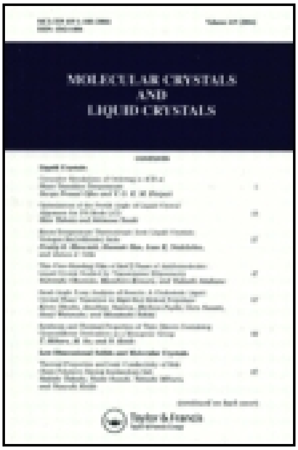

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# ENHANCED FLEXOELECTRIC SWITCHING BEHAVIOUR IN THE CHIRAL NEMATIC PHASE

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# Enhanced Flexoelectric Switching Behaviour in the Chiral Nematic Phase

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In order to develop materials that exhibit enhanced flexoelectric switching in the chiral nematic phase we have identified mesogenic units that display inherently strong flexoelectric coupling capabilities. Here we examine the oxycyanobiphenyl (OCB) moiety: homologues from the *n*OCB series exhibit significant electro-optic switching effects when doped with a highly chiral additive. Here we have examined lower dielectric anisotropy materials, since they allow the flexoelectric response to be extended to high field amplitudes. We show that dielectric coupling strength can be low in symmetric bimesogenic molecules. The flexoelectric response of such a molecular structure is tested by doping a homologue from the series CBO*n*OCB with a chiral additive: very significantly we find that the optic axis is rotated through $2\phi=45°$ in <50 μs on reversing the polarity of the field (amplitude $E=\pm6$ V $\mu m^{-1}$). Subsequently we have synthesized room temperature chiral nematic materials that exhibit $2\phi\geq90°$ at $E\approx10$ V $\mu m^{-1}$.

*Keywords:* chiral nematic; cholesteric; flexoelectricity; dielectric anisotropy; electro-optic; bimesogen

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## INTRODUCTION

Flexoelectric coupling [1] in a short pitch ($P<1$ μm) chiral nematic phase can engender a fast (≤100 μs), temperature-independent birefringence switching effect that is linear with applied field amplitude.[2] The electro-optic effect stems from the flexoelectrically-induced in-plane rotation $\phi$ of the optic axis, which is associated with the helical structure of the phase, as the amplitude of the field **E** applied across the uniaxial lying helix texture is varied.

While a strong flexoelectric coupling mechanism is clearly very important to the design of molecular structures for the flexoelectric switching effect, the impact of dielectric coupling must also be recognised.[2,3] For optimal performance, a large deflection of the optic axis through an angle $\phi$ from its unperturbed position must be achieved before dielectric coupling can act either to realign the mean helix axis or to unwind the helical structure. The equation for the free energy $f$ of a chiral nematic subjected to an electric field, illustrates the individual coupling processes' dependencies on the applied field:

$$f = \frac{1}{2}\left[K_{11}(\nabla\cdot\mathbf{n})^2 + K_{22}\left(\frac{2\pi}{P} + \mathbf{n}\cdot\nabla\times\mathbf{n}\right)^2 + K_{33}(\mathbf{n}\times\nabla\times\mathbf{n})^2\right]$$
$$-\frac{\varepsilon_0\Delta\varepsilon}{2}(\mathbf{n}\cdot\mathbf{E})^2 - \mathbf{E}\cdot(e_s\mathbf{n}\nabla\cdot\mathbf{n} + e_b\mathbf{n}\times\nabla\times\mathbf{n})\,. \qquad (1)$$

The first term in Eq. 1 gives the elastic free energy density of the undisturbed chiral nematic. $K_{11}$, $K_{22}$ and $K_{33}$ are the elastic constants of splay, twist and bend respectively. The second term is the contribution from the coupling between the electric field **E** and the dielectric anisotropy, $\Delta\varepsilon = \varepsilon_{\|} - \varepsilon_{\perp}$. Note that the dielectric coupling mechanism is quadratic in $E$. The third term describes the effect of the flexoelectric coupling between the field and the material. The parameters $e_s$ and $e_b$ are the flexoelectric coefficients associated with splay and bend director deformation. In this paper we use the sign convention devised previously.[4] Flexoelectric coupling is linear in $E$ and is therefore dominant at low fields for lower $\Delta\varepsilon$ materials.

The rotation of the optic axis induced by flexoelectric coupling is related to the natural helical pitch $P$ of the phase via[2]

$$\tan\phi = (eEP)/(2\pi K), \qquad (2)$$

where $K$ and $e$ are the effective elastic constant and flexoelectric coefficient of the phase, respectively: $K = (K_{11}+K_{33})/2$ and $e = (e_s+e_b)/2$. The characteristic response time $\tau$ of flexoelectro-optic switching[5] is given by $\tau = P^2\gamma/(4\pi^2 K)$, where $\gamma$ is the effective viscosity associated with the helix distortion.

To date, strong flexoelectric switching effects at low field amplitudes have been reported[6,7] in materials usually with positive dielectric anisotropy. At higher fields the promising flexoelectric performance of such materials, however, has been limited by dielectric coupling causing the helical structure to be unwound. So, for example, in asymmetric bimesogens composed of an OCB group connected to the chiral mesogen estradiol via a flexible aliphatic spacer,[7] it was found that values of $\phi \approx 10°$ were recorded before the helical structures were unwound at $E \approx 10$ V $\mu m^{-1}$. Large rotation angles have been reported in one material with very low positive dielectric anisotropy (TM216).[8]

However, the flexoelectric effect in this material is weak: a deflection of $\phi = 30°$ was recorded only on the application of an inordinately large field ($E \approx 130$ V $\mu m^{-1}$).

In order to design molecules specifically for flexoelectric coupling we have found it expedient to identify mesogenic units that undergo strong flexoelectric coupling mechanisms and then to combine these groups in bimesogenic structures so that the molecular dielectric anisotropy is minimised. Here we will describe the results which lead to the synthesis of new bimesogens displaying $\phi = 45°$ at $E \approx 9$ V $\mu m^{-1}$.[9]

## RESULTS AND DISCUSSION

A number of proprietary chiral additives (supplied by Merck UK Ltd.) have been employed as dopants in achiral nematic liquid crystals in the course of this work. Used at concentrations in the range 4 to 6% by weight, these dopants are assumed to have little effect on the elastic, dielectric and flexoelectric properties of the host materials. The structures of the three nematic host materials discussed in this paper are shown in Figure 1. We have chosen to examine the monomesogen 7OCB, the bimesogen CBO8OCB, and we have synthesized new symmetric bimesogens specifically for this work.[9] The table illustrates the thermo-optic transition temperatures of the chiral mixtures, together with their pitch lengths, which were measured using the Cano wedge technique or (in the case of the high temperature material CBO8OCB) was estimated, at the reduced temperatures $T_r$ ($T_r$=$T_{meas}$-$T_{clear}$) shown. Also given in the table are published values for the $\Delta\varepsilon$ in the host materials 7OCB[10] and CBO8OCB[11] at the reduced temperatures shown. The electro-optic properties of the chiral nematics confirm that although Δε is positive in all three materials, its magnitude in the monomesogen is significantly higher than it is in the bimesogens.

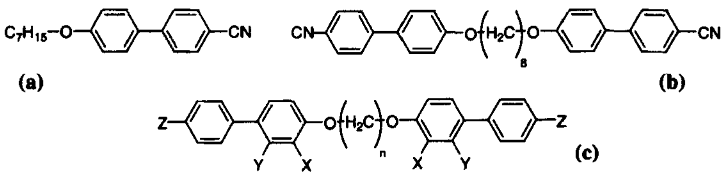

FIGURE 1 Chemical structures of (a) 7OCB, (b) CBO8OCB, and (c) a novel symmetric bimesogen (X, Y, Z represent functional groups).

| Host material | Transition temperatures of mixture, °C | Pitch length of mixture, nm ($T_r$, °C) | Δε ($T_r$, °C) |
|---|---|---|---|
| 7OCB | Cr 55 N* 69 BP 70 I | 295 (-7) | +8.8 (-8) |
| CBO8OCB | Cr 175 N* 205 I | 700 (-22) | +3.3 (-27) |
| New bimesogen | Cr <20 N* 57 I | 890 (-22) | Low[12] |

TABLE The phase properties of the chiral nematic materials.

The flexoelectric effect was observed in 6 μm spaced cells. Electric fields were applied across the filled cells by means of transparent electrodes deposited on the inner cell walls. The square waveforms of the applied fields were usually in the frequency range 10 Hz – 100 Hz, and varied in amplitude from zero up to ±10 V$\mu$m$^{-1}$. The procedure for measuring tilt angles and response times has been described previously.[7] The flexoelectric switching effect is observed with the chiral nematic helix perpendicular to both the applied field and the viewing direction, *i.e.* with the uniaxially-lying helix form of the focal conic texture. The alignment technique is also described elsewhere.[7]

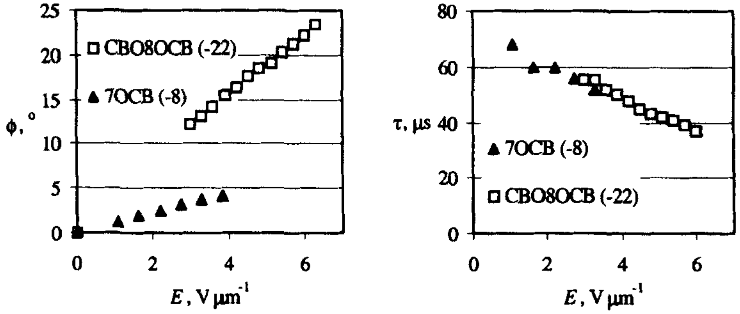

FIGURE 2 Switching properties of doped 7OCB and CBO8OCB measured at the reduced temperatures ($T_r$) shown in parentheses.

Figure 2 shows the electro-optic responses of the 7OCB and CBO8OCB mixtures as functions of applied field amplitude, measured at the reduced temperatures indicated. The relationship between $\phi$ and $E$ in these mixtures is linear, in agreement with Eq. 2. The values of $\phi$ exhibited by the CBO8OCB mixture at a given applied field are larger by a factor of 3.5 than those exhibited by the 7OCB mixture. This difference is mainly due to the longer pitch of the bimesogenic mixture (Eq. 2), which is a factor of ≈2.3 times larger than the pitch of the 7OCB mixture at these temperatures. The values of the ratio $e/K$ in these mixtures are rather similar and large, at 0.5 and 0.6 C $N^{-1}$ $m^{-1}$ in 7OCB and CBO8OCB respectively, indicating that the behaviour of the bimesogen is likely to be governed by the individual responses of the terminal mesogenic units. Most strikingly, the values of $\phi$ recorded before helix-unwinding are significantly higher in the bimesogenic material, owing to its lower dielectric anisotropy. The CBO8OCB mixture is the first short pitch chiral nematic material to exhibit such a strong response at low field amplitudes ($\phi = 22.5°$ at $E = 6$ V $\mu m^{-1}$). The response times of these materials as a function of applied field amplitude are also given in Figure 2. As $E$ is increased the value of $\tau$ decreases, in agreement with previous observations.[5] Both materials respond very rapidly, over tens of microseconds, to variations in applied field amplitude. We believe that the fast response of the bimesogen is attributable to the high temperature (183 °C) at which the measurements were made.

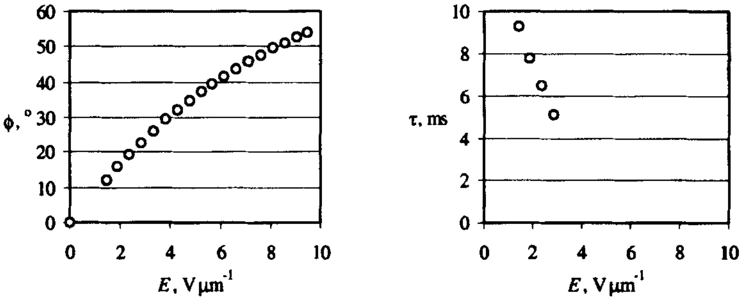

FIGURE 3 Behaviour of the new material at $T_r$ = -22 °C. Note that response times are measured only for $\phi < 22.5°$.

Although the electro-optic behaviour of the CBO8OCB mixture is impressive, the chiral nematic phase exists at a rather high temperature. We have subsequently synthesized room temperature nematic symmetric bimesogenic materials. The behaviour of one mixture containing the new bimesogens is illustrated in figure 3. The response time of this mixture is in the millisecond range for optic axis deflections of $2\phi < 45°$, diminishing to microseconds as the field is increased to 10 V $\mu m^{-1}$. We are currently investigating the possibility of increasing the mobility of the phase by altering the form of the terminal mesogenic units. The magnitude of $\phi$ at a given applied field amplitude is similar to that observed in those materials containing the OCB mesogenic unit, although, owing to the stability of the lying helix texture, very large deflections of the optic axis are now possible. For example $2\phi = 110°$ for $E = \pm 9.5$ V $\mu m^{-1}$ is measured at $T_r = -22$ °C: such a response has not been reported in any other chiral nematic material. The value of $e/K$ in the bimesogen at this temperature is 1.0 C $N^{-1}$ $m^{-1}$, which is twice as large as the values observed in the OCB-containing materials. To conclude, we would suggest that the exceptional flexoelectric coupling behaviour of bimesogenic nematics warrants wider investigation.

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