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R. Balachandran  
*Trinity College Dublin, Ireland*

V.P. Panov  
*Trinity College Dublin, Ireland*

Yuri Panarin  
*Dublin Institute of Technology, [yuri.panarin@dit.ie](mailto:yuri.panarin@dit.ie)*

M. G. Tamba  
*University of Hull*

G. H. Mehl  
*University of Hull*

*See next page for additional authors*

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**Authors**

R. Balachandran, V. P. Panov, Yuri Panarin, M. G. Tamba, G. H. Mehl, J. K. Song, and J. K. Vij

# Flexoelectric Behavior of a Bimesogenic Liquid Crystal

R. BALACHANDRAN,<sup>1</sup> V. P. PANOV,<sup>1,2</sup> YU. P. PANARIN,<sup>1,3</sup>  
M. G. TAMBA,<sup>4</sup> G. H. MEHL,<sup>4</sup> J. K. SONG,<sup>5</sup> AND J. K. VIJ<sup>1,\*</sup>

<sup>1</sup>Department of Electronic and Electrical Engineering, Trinity College,  
University of Dublin, Dublin, Ireland

<sup>2</sup>School of Physics and Astronomy, University of Manchester, Manchester,  
United Kingdom

<sup>3</sup>School of Electronic and Communication Engineering, Dublin Institute of  
Technology, Dublin, Ireland

<sup>4</sup>Department of Chemistry, University of Hull, UK

<sup>5</sup>School of Electronic and Electrical Engineering, Sungkyunkwan University,  
Suwon, Korea

*The effective flexoelectric coefficient and flexoelectric polarization are investigated for a bimesogenic liquid crystal CBC11CB with a twist bend nematic phase ( $N_{tb}$ ). The effective flexoelectric coefficients for CBC11CB in the ordinary nematic phase ( $N$ ) are obtained to be much larger in comparison to conventional calamitics, and is almost twice higher for CBC11CB than that reported for another odd symmetric bimesogen. The flexoelectric polarization ( $P_f$ ) in the  $N$  phase is found to be almost 2.5 times higher than in rod-like molecules. Additionally,  $P_f$  is obtained to be much larger in the  $N_{tb}$  phase than in the nematic phase.*

**Keywords**  $N_{tb}$  phase; bimesogens; flexoelectric coefficient; ULH; flexoelectric polarization

## Introduction

The flexoelectric effect in liquid crystals was first predicted by Meyer [1] as a phenomenon analogous to piezoelectricity observed in crystalline solids. The flexoelectric polarization is expressed in terms of the splay ( $e_1$ ) and bend ( $e_3$ ) flexoelectric coefficients as follows [1]:

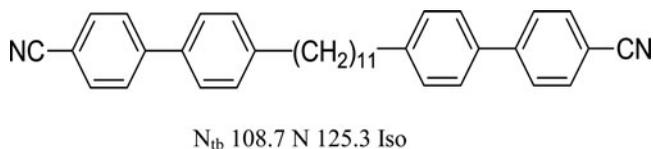
$$P_f = e_1 \mathbf{n}(\nabla \cdot \mathbf{n}) - e_3 \mathbf{n} \times (\nabla \times \mathbf{n})$$

Bimesogens or dimers are liquid crystals composed of two mesogenic units connected via flexible spacers. Altering the parity of the alkyl chain in bimesogens has been reported to give rise to significant odd-even effects in these materials. In some odd bimesogens

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\*Address correspondence to J. K. Vij, Department of Electronic and Electrical Engineering, Trinity College, University of Dublin, Dublin 2, Ireland. E-mail: [jvij@tcd.ie](mailto:jvij@tcd.ie)

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**Figure 1.** Chemical structure and transition temperature of the material under study: CBC11CB.

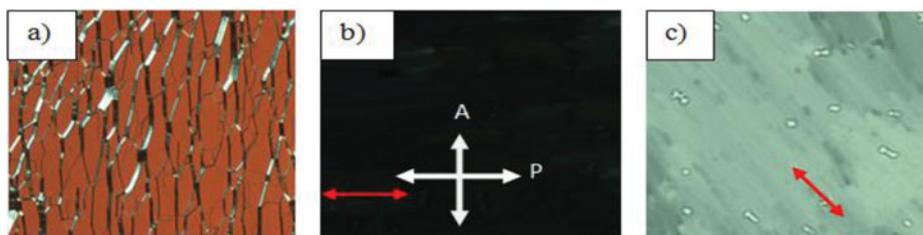
there exists a second nematic phase in addition to the higher temperature ordinary nematic phase ( $N$ ). Recent studies [2] have indicated that this phase could indeed be the twist-bend ( $N_{tb}$ ), similar to the ones predicted by R.B. Meyer [3] and I. Dozov [4]. The theoretical studies pertaining to the mysterious yet fascinating  $N_{tb}$  phase is still in its initial stages. However, the parameters of ordinary nematic phase in these materials, the equations for which are well established, could also reveal useful information related to the underlying  $N_{tb}$  phase.

## Material Under Study

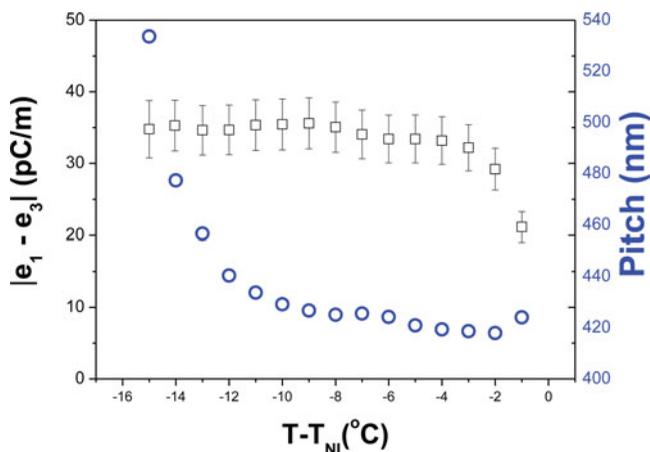
The material under investigation, CBC11CB is a symmetric dimer, synthesized in Hull, which consists of two cyanobiphenyl units linked by an alkyl chain of eleven methylene groups. The chemical structure and phase transition temperature of the sample is shown in Fig. 4.1. The material, on cooling from the isotropic phase, exhibits a transition to the ordinary nematic phase ( $N$ ) and to the second nematic phase ( $N_{tb}$ ) on further lowering the temperature.

## Flexoelectric Measurements

The measurement of the effective flexoelectric coefficient  $|(e_1 - e_3)|$  was carried out by exploiting the flexoelectro-optic effect observed in the uniform lying helix (ULH) geometry of a chiral nematic [5]. Since the material under investigation is achiral, the cholesteric state is induced by doping the sample with a chiral dopant (3% by weight R5011, Merck, Korea) of large helical twisting power ( $\sim 100 \mu\text{m}^{-1}$ ). The ULH structure is not formed spontaneously on cooling a planar cell from the isotropic phase due to the minimum free energy configuration of a chiral nematic being a Grandjean texture. Hence, the cell was



**Figure 2.** (a) Cholesteric texture obtained in a planar cell under crossed polarizers, on cooling from the isotropic phase without field applied, at  $T - T_{NI} = -3^\circ\text{C}$ . (b) and (c) depict the ULH textures captured by a 20x lens; the optic axis (depicted by the red arrow) in each case is aligned at angles of  $0^\circ$  (b) and  $45^\circ$  (c) with respect to the polarizer P.

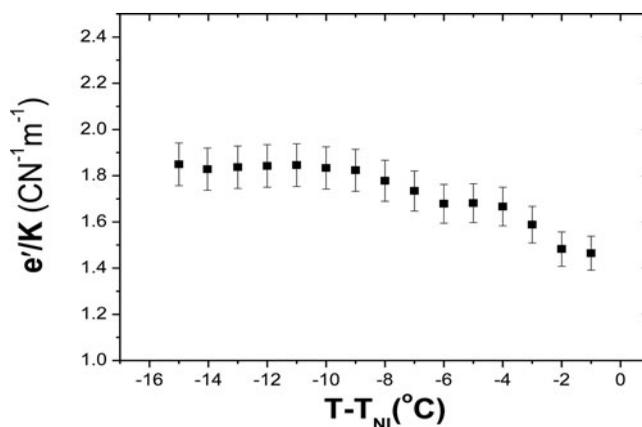


**Figure 3.** Temperature dependence of the pitch and the effective flexoelectric coefficient.

cooled at a rate of  $0.1^{\circ}\text{C}/\text{min}$  under the influence of a moderate electric field ( $E \sim 2 \text{ V}/\mu\text{m}$ ) at a frequency of 100 Hz. Once the ULH (Fig. 2(b, c)) is formed, an electric field applied perpendicular to the helicoidal axis of the ULH along the substrate results into in-plane deviation of the optic axis caused by the flexoelectric coupling [5]. For small electric fields  $E$ , the tilt angle  $\phi$  is linearly related to the applied electric field by:

$$\tan \phi = \frac{e' p}{2\pi K} E$$

$e$  and  $K$  are the flexoelectric and effective elastic constants, respectively. These are defined as:  $e' = \frac{(e_1 - e_3)}{2}$ ,  $K = \frac{K_{11} + K_{33}}{2}$ ,  $p$  is the helicoidal pitch of the helix. The optical pitch of the helix was determined in a planar cell by the measurement of the selective reflection using a UV/VIS spectrometer, and the tilt angle is measured using a method described in ref. [6]. The value of the effective elastic constants was taken from our previous measurements [7] on the same material.

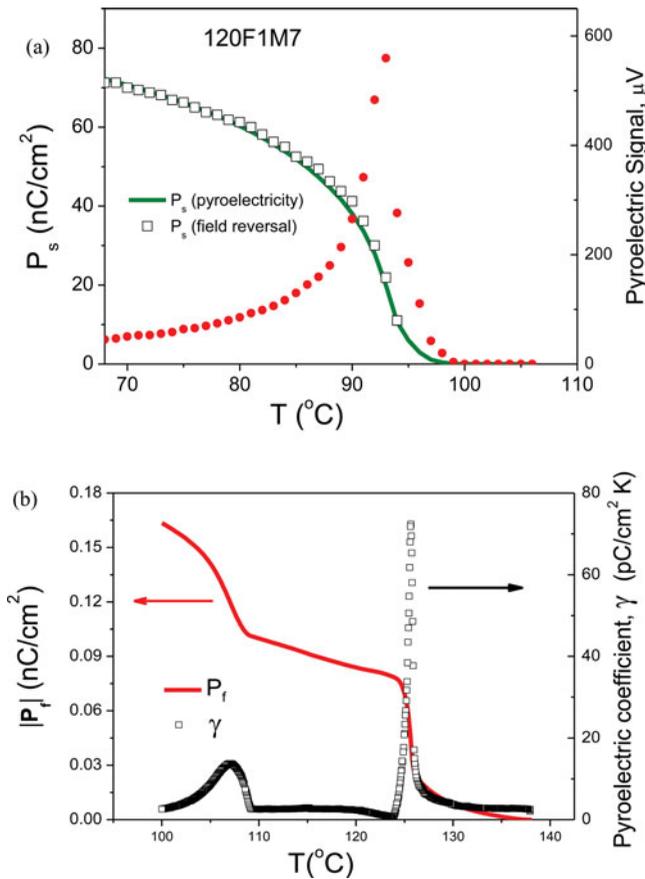


**Figure 4.** Flexo-elastic ratio as a function of reduced temperature.

Figure 3 shows the variance of the optical pitch  $p$  and  $|(e_1 - e_3)|$  in the  $N^*$  phase as a function of the reduced temperature. The value of  $|(e_1 - e_3)|$  increases from 21 pC/m at  $T - T_{NI} = -1^\circ\text{C}$  to  $\sim 35$  pC/m at the lowest temperature for which these measurements were performed.  $|(e_1 - e_3)|$  increases marginally at higher temperatures and becomes nearly independent of it as the temperature is decreased further. The quality of alignment and the resulting signal was seen to degrade significantly at the temperatures approached the  $N_{tb}$  phase transition, rendering it nearly impossible to carry out measurements at these points.

For a well-known liquid crystal 5CB, the effective flexoelectric coefficient was determined to be 7.10 pC/m at  $T = 25^\circ\text{C}$  [8], which is smaller by a factor of almost 5 in comparison to the value obtained for CBC11CB. It is also almost twice greater than bimesogens [9] in which  $N_{tb}$  phase is absent.

The temperature dependence of the flexo-elastic ratio  $e'/K$  is plotted in Fig. 4.13.  $e'/K$  is seen to be weakly dependent on temperature at higher temperatures, and becomes nearly independent of it at lower temperatures. The ratio is observed to increase from  $\sim 1.58$  to  $\sim 1.85 \text{ CN}^{-1}\text{m}^{-1}$  as the temperature is reduced.



**Figure 5.** (a) Temperature dependence of the pyroelectric signal (red circles), and spontaneous polarization of 120F1M7 measured using field reversal (black squares) and pyroelectric technique (green line). (b) Temperature dependence of the pyroelectric signal (black squares) and flexoelectric polarization (red line) of CBC11CB.

## Flexoelectric Polarization

The flexoelectric polarization  $P_f$  is obtained by integrating the pyroelectric coefficient,  $\gamma$ , measured from a 7.7  $\mu\text{m}$  hybrid cell (HAN) filled with CBC11CB (experimental details in ref. [10]). In a HAN cell, one of the substrate is coated for planar alignment and the other for homeotropic. In such a configuration the director field has a permanent splay-bend deformation which generates a flexoelectric polarization  $P_f$ .

The absolute value of  $P_f(T)$  cannot be calculated by the pyroelectric measurements alone as the time rate of change of temperature of the sample due to the heating by the light pulse is unknown. The calibration of the pyroelectric signal, required to obtain the absolute value of the pyroelectric coefficient for CBC11CB, was provided by a well-known ferroelectric liquid crystal 120F1M7 whose spontaneous polarization was determined independently using the well-known current repolarization technique. The pyroelectric signal generated for 120F1M7 was recorded using the same measurement conditions as for CBC11CB. The integrated pyroelectric signal was then scaled to the value of  $P_s$ , evaluated using the field reversal method, at a particular temperature (Fig. 5a).

Figure 5b shows the temperature dependence of the pyroelectric signal and the absolute value of flexoelectric polarization  $P_f$ . The peaks that appears at  $T = 126.6^\circ\text{C}$  and  $107^\circ\text{C}$  (on cooling the liquid crystal) correspond to the *Iso-N* and *N-N<sub>tb</sub>* phase transitions, respectively. The magnitude of  $P_f$  increases on transition from the isotropic phase and is seen to grow marginally from 0.08 to 0.10  $\text{nC}/\text{cm}^2$  as the temperature is further reduced in the nematic phase. It is observed to increase steadily on cooling at the transition into the *N<sub>tb</sub>* phase. The value reaches almost 0.17  $\text{nC}/\text{cm}^2$  at the lowest temperature for which the measurements were carried out. Our results show that the *N<sub>tb</sub>* phase has a large flexoelectric polarization compared to the classical nematic phase, by a factor of almost 2. This is also possibly the source of the electroclinic effect observed in this phase by Meyer et. al. [11]. These results on CBC11CB also indicate that splay deformations may influence the macroscopic effects exhibited by the twist-bend nematic phase especially close to the transition temperature. This implies that as the macroscopic structure evolves from the nanostructure, splay deformations can also have a role especially in exhibiting macroscopic effects.

## Conclusion

We have obtained the effective flexoelectric coefficient,  $|(e_1 - e_3)|$  in the nematic phase and flexoelectric polarization in both the ordinary nematic and twist nematic phase of a bimesogen CBC11CB. The value of  $(e_1 - e_3)$  ranged from  $\sim 21\text{--}35$   $\text{pC}/\text{m}$ , and was observed to be nearly independent of temperature after an initial increase below the isotropic-nematic transition. The effective flexoelectric coefficients for CBC11CB were obtained to be much larger in comparison to conventional calamitics and almost twice higher for CBC11CB than that reported for another odd symmetric bimesogen. The flexoelectric polarization in the *N* phase was found to be almost 2.5 times higher than in rod-like molecules. Additionally,  $P_f$  was obtained to be much larger in the *N<sub>tb</sub>* phase than in the nematic phase. This is possibly the source of the electroclinic effect observed in this phase.

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