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INVITED ARTICLE

Flexoelectric effect in a bent-core mesogen

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The flexoelectricity in a bent-core liquid crystal was evaluated quantitatively using two independent electrooptic methods. The absolute value of the bend flexoelectric coefficient e_3 was measured to be about 15.8 pC m^{-1}. Our results revealed that there is little difference between the flexoelectricity of bent-core mesogens and that of conventional calamitic mesogens, i.e. in the same pC m^{-1} order. The principles of the methods used are reviewed in detail.

Keywords: flexoelectric effect; bent-core liquid crystal; converse flexoelectric effect; flexoelectric instability

1. Introduction

It has been exactly 40 years since the existence of a flexoelectric effect in nematic liquid crystals (LCs) was first described by R.B. Meyer (*1*) in 1969. This effect has been thought to have potential applications for electromechanical devices, including strain gauges, actuators and micropower generators. The flexoelectric effect is defined as the coupling between orientational deformation and electric polarisation. The nematic LC director can be distorted mechanically in a splay or a bending fashion, and a resultant polarisation **P** is created, as shown in Figure 1. This effect is similar to, but not identical with, the piezoelectric effect, which is the phenomenon of the generation of an electric polarisation when stimulated by some external mechanical pressure to solids without having inversion symmetry. The flexoelectric effect originally was called the piezoelectric effect (*1*), but this leads to misunderstanding as the physical origins of the two effects differ from each other. In nematic LCs, pressure does not influence the director **n** and is thus unable to induce distortions and the associated polarisations (*2*). In other words, piezoelectricity is induced by a positional strain (first order spatial derivative), whereas flexoelectricity occurs due to a curvature strain (spatial change of director orientation, second order spatial derivative). For this reason, the terminology 'flexoelectric', which was first proposed by P.G. de Gennes (*2*), has been preferably used until now.

As shown in Figure 1, when the orientation in the nematic phase is uniform, the system is non-polar due to the free rotation of the constituent molecules around their axes and thus the non-polar cylindrical symmetry of the distribution of dipoles (Figure 1(a) and (b)) (*3*). However, a polar axis can arise when a splay and a bending are imposed to the pear-shaped and banana-shaped molecules, respectively (Figure 1(c) and (d)). In this case, the polar order is induced via the closer packing of the molecules. These two geometries involve two corresponding coefficients, namely, splay flexoelectric coefficient e_1 and bend flexoelectric coefficient e_3; and the total flexoelectric polarisation is expressed as:

$$\mathbf{P}_{\mathrm{f}} = e_1\mathbf{n}(\mathrm{div}\ \mathbf{n}) + e_3(\mathrm{curl}\ \mathbf{n}) \times \mathbf{n}. \tag{1}$$

There are various methods to obtain the numerical values of the difference $(e_1 - e_3)$ or the sum $(e_1 + e_3)$ of the two coefficients. The results in some typical calamitic nematics and popular electrooptic methods are summarised in Table 1. All of them range in the pC m^{-1} order and sometimes differ even with the same material, depending on the techniques used. The signs in some values are not able to be determind, and at that time additional experiments are required. An efficient but quite simple method to perform such experiments using asymmetric surface constraints was introduced in Kim and Yokoyama (*17*).

Although one can roughly estimate the individual values of the two coefficients by solving the simultaneous equations constructed by their difference and sum, efforts have also been made to determine them separately. Among these, a new electromechanical technique measuring e_3 has been devised recently by Harden *et al.* (*18*). An oscillatory bending deformation was imposed on a flexible LC cell and then the induced electric current was directly measured. Particularly, for the first time, the value of e_3 of a bent-core nematic

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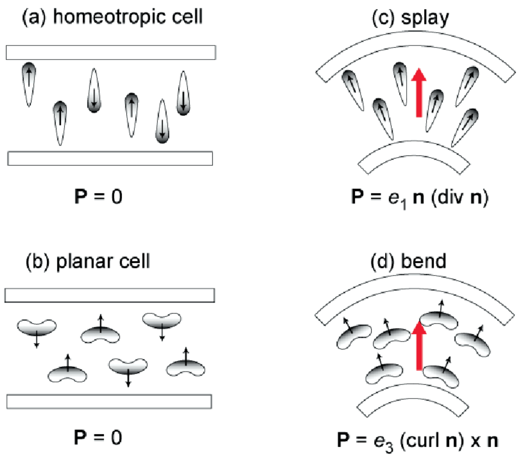

Figure 1. Origin of the two flexoelectric coefficients in nematics. (a) Homeotropic cell containing pear-shaped molecules; (b) planar cell with banana-shaped molecules inside; (c) polarisation created by a splay deformation; (d) polarisation created by a bending deformation.

Table 1. Reported values [pC m^{-1}] of flexoelectric coefficients of some calamitic liquid crystals.

Value	Sample	Technique	Ref.
e_1-e_3			
+3.3	MBBA	hybrid-aligned nematic cell	[4]
+14	MBBA	hybrid-aligned nematic cell	[5]
–15	ZLI-4792	hybrid-aligned nematic cell	[5]
|5.7|	BMAOB	homogeneous cell	[6]
+11	5CB	hybrid-aligned nematic cell	[7]
+9.3	E7	twisted nematic cell	[8]
|4.8|	7CB	homogenous cell, uniformly lying helix structure	[9]
e_1+e_3			
–54	MBBA	hybrid-aligned nematic cell	[5]
|23.7|	MBBA	homeotropic cell, interdigited electrode	[10]
–15	MBBA	hybrid-aligned nematic cell	[11]
–3.3	MBBA	homeotropic cell, quadrupolar field	[12]
|21.4|	8OCB	homeotropic cell, interdigited electrode	[10]
+4.7	8OCB	homeotropic cell, quadrupolar field	[12]
+7.4	8OCB	hybrid-aligned nematic cell	[13]
+30	5CB	hybrid-aligned nematic cell	[14]
+35	E7	π-cell	[15]
e_3			
|1.23|	MBBA	homeotropic cell, transverse field	[16]

ZLI-4792 and E7 are nematic mixtures.

LC was determined to be about 35 nC m^{-1}, which is three orders of magnitude greater than those of traditional calamitic nematic LCs. This giant value has motivated us to confirm it by other available methods.

In our work, two independent methods utilising (i) a converse flexoelectric effect (*16*) and (ii) flexoelectric instability were employed (*6*). Both techniques applied to the bent-core mesogen used in the previous study (*18*) gave the same result of pC m^{-1} order, which is not so different from that of calamitic mesogens.

2. Converse flexoelectric effect

2.1 Principle

The principle of the method is explained as follows. When a DC field is applied transversely through the homeotropic cell as indicated in Figure 2, the coupling between the induced flexoelectric polarisation $\mathbf{P}_f$ and the external electric field **E** occurs, leading to the bending deformation of the director. This is the so-called converse flexoelectric effect.

The relation of the physical parameters involved in this effect is expressed as (*16*):

$$\Delta l = \frac{e_3^2}{K_3^2} E^2 n_o \left(1 - \frac{n_o^2}{n_e^2}\right) \frac{d^3}{24} \qquad (2)$$

where Δl is the optical path difference, K_3 the bend elastic constant, E the strength of applied field, d the cell thickness, and n_o, n_e are the ordinary and extraordinary refractive indices, respectively. In the present work, instead of the optical path difference Δl, the phase retardation δ was detected directly by a spectroscopic ellipsometer (Sopra, GESP 5) (at λ = 633 nm). Since we have $\delta = \frac{2\pi \Delta l}{\lambda}$, Equation (2) may be written as:

$$\delta = \frac{2\pi}{\lambda} \frac{e_3^2}{K_3^2} E^2 n_o \left(1 - \frac{n_o^2}{n_e^2}\right) \frac{d^3}{24}. \qquad (3)$$

The best fit of experimental data to Equation (3) leads to the ratio $|e_3/K_3|$ as a fitting parameter.

It should be noted that this method is validated only if these three conditions are satisfied (*19*):

(1) The anchoring force in the vicinity of the glass substrate should be sufficiently small.
(2) The dielectric anisotropy $\Delta\varepsilon$ of LCs must be as small as possible to avoid the influence of

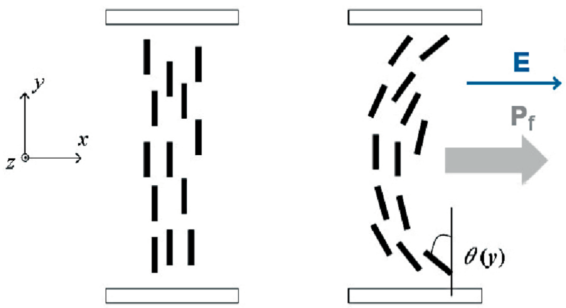

Figure 2. Molecular alignment of a homeotropic cell for the measurement of the bend flexoelectric coefficient.

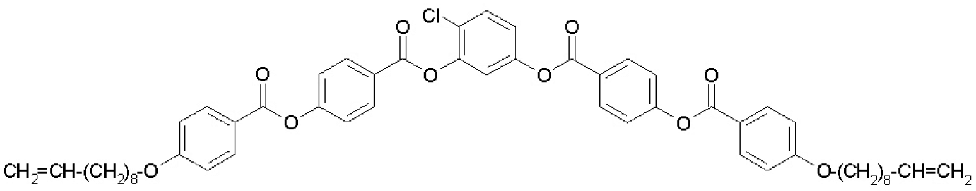

Figure 3. Chemical structure of compound ClPbis10BB.

Freedericksz transition, which would turn the bending geometry into the initial homeotropic alignment.

(3) The applied field should not be too high because the flexoelectric effect is proportional to the electric field (*E*), whereas the dielectric torque is proportional to the second power of the electric field (E^2).

2.2 *Experiment*

The bent-core compound called ClPbis10BB (Figure 3) was used in the present study. Some thermodynamic and electrooptic properties of this nematic have been reported (*20, 21*). This material has a relatively low transition temperature and a fairly wide N phase: Iso 74.5°C N 49.5°C Cry as observed using a polarising microscope upon cooling. The LC was introduced into a homeotropically aligned (JALS-204) cell, which contained two parallel 12-μm thick strips of aluminium foil serving as spacers and electrodes with a gap of 2 mm. With such a design, it was possible to apply a uniform transverse field throughout the LC bulk. A 633-nm beam was incident in the middle of the two strips of aluminium foil of the sample cell at 71.5°C, and the retardation was measured along the transmission direction (*22, 23*).

2.3 *Results and discussion*

When the sample was cooled down from the isotropic phase, the texture as shown in Figure 4(a) was observed. For a short while, all the defects disappeared and a completely dark N domain was obtained. We note that Equation (3) is only valid in the case of a uniaxial material, while bent-core mesogens are generally thought to have biaxiality. Thus we also confirmed ClPbis10BB's uniaxiality by observing its N phase through a conoscopic polarising microscope (see Figure 5). The uniaxiality was also confirmed earlier (*20*).

The experimental results are shown in Figure 6. We used a least-squares method to fit the experimental data to Equation (3) and found that $|e_3|$ of ClPbis10BB was about 15.8 pC m^{-1}, with no significant temperature dependence down to 65°C. Here, $K_3 = 5.22 \times 10^{-12}$ N as determined from a preliminary experiment of Freedericksz threshold voltage, and we assumed that $n_o = 1.58$ and $n_e = 1.63$. To prove the validity of this method, we also performed the same experiment with

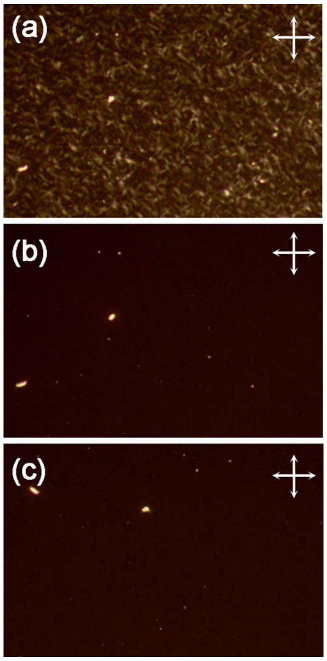

Figure 4. Nematic phase of ClPbis10BB at 71.5°C. (a) Thread-like texture, (b) uniform darkness and (c) unchanged darkness when the sample was rotated indicating a perfect homeotropic orientation.

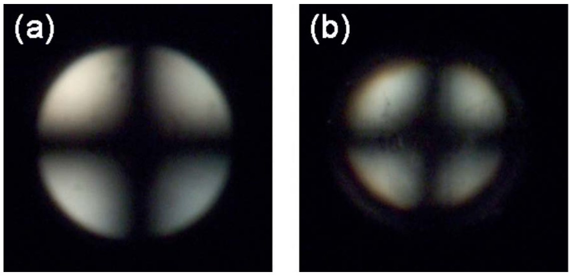

Figure 5. Conoscopic figures of homeotropic cells of (a) MBBA and (b) ClPbis10BB, with the same alignment layer JALS-204 (JSR).

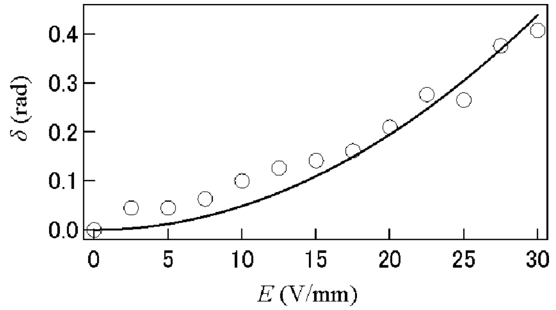

Figure 6. Dependence of phase retardation on the electric field in ClPbis10BB at 71.5°C. Open circles represent the experimental data and the solid line is the best fit to Equation (3).

MBBA (at 22°C). In this case, an absolute value of $|e_3|$ of around 2.50 pC m^{-1} was obtained, which agrees quite well with the result reported previously (*16*).

Comparing two results of the same bent-core compound ClPbis10BB, it should be noted that our present result is more than three orders of magnitude smaller than that reported previously (*18*). However, our result is quite close to the molecular statistical calculation proposed by Helfrich (*24*) in 1971. It is possible to calculate the macroscopic polarisation by taking into account the dipole moments μ of individual molecules. For a banana-shaped molecule shown in Figure 7, the numerical estimate of e_3 can be expressed as (*24*):

$$e_3 = \frac{\mu_\perp K_3}{2k_B T}\theta_0\left(\frac{b}{a}\right)^{2/3} N^{1/3} \tag{4}$$

where $\mu_\perp$ is the transverse component of the dipole moment, k_B the Boltzmann constant, T the absolute temperature, θ_0 the opening angle, N the density of the molecules, and a and b are the length and width of a molecule, respectively. If we assume that for a typical nematic, $k_BT = 5 \times 10^{-21}$ J, $b/a = 1/2$, and $N = 10^{27}$ m^{-3}, and consider that $\mu_\perp$ of a bent-core molecule is about 3 D (*25*) (note that 1 D is around 3.33564×10^{-30} C m), and $\theta_0 = 1.05$ (about 60°), then with $K_3 = 5 \times 10^{-12}$ N, the value of e_3 is estimated to be approximately 1.1 pC m^{-1}. The similarity of pC m^{-1} of e_3 and e_1 is discussed in detail in Helfrich (*24*).

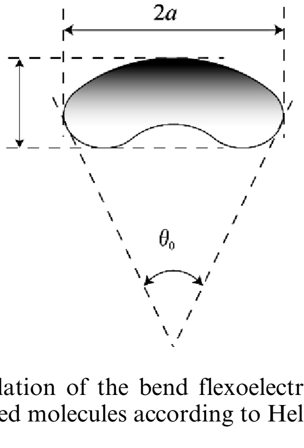

Figure 7. Calculation of the bend flexoelectric coefficient for banana-shaped molecules according to Helfrich [24].

On the other hand, it is worth noting that a recent Monte Carlo simulation also showed that there was no significant flexoelectric enhancement for bent-core nematics (*26*). Meanwhile, recently, Aziz *et al.* (*27*) doped a nematic liquid crystalline mixture (MLC6437-000) with some banana-shaped dopants (8 wt%) and quantitatively evaluated the mean flexoelectric coefficient $(e_1+e_3)/2$ using the uniformly lying helix technique (*9*). Unfortunately, in contrast to the initial intention, the flexoelectricity in the doped mixtures decreased compared with that of the host. This result again does not support the very large flexoelectricity in bent-core mesogens reported earlier (*18*). Thus we believe there is little difference in the flexoelectric coefficients between bent-core and rod-like mesogens.

3. Flexoelectric instability

3.1 Principle

In order to confirm the result described above, we also performed another method making use of flexoelectric instability. Flexoelectric instability describes the appearance of a longitudinal domain or parallel-stripe domain in planar nematic films when an electric field (DC or AC at low frequencies) is applied vertically to the director, resulting in periodic distortions in the sample. The deformation of the director for one-dimensional geometry and that for two-dimensional geometry, which corresponds to the longitudinal domain, are shown schematically in Figure 8,

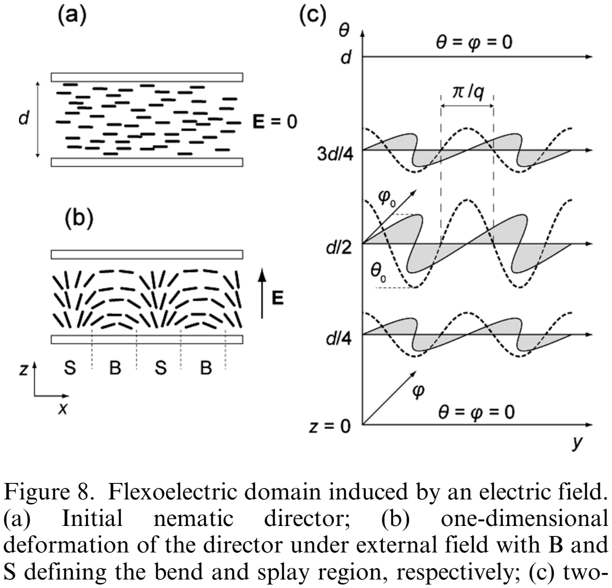

Figure 8. Flexoelectric domain induced by an electric field. (a) Initial nematic director; (b) one-dimensional deformation of the director under external field with B and S defining the bend and splay region, respectively; (c) two-dimensional flexoelectric distortion.

where θ and ϕ denote the tilt angles in plane xz and plane xy, respectively (*28*); $\theta = \theta_0 \cos(qy) \cos\frac{\pi z}{d}$ and $\varphi = \varphi_0 \sin(qy) \cos\frac{\pi z}{d}$.

The theoretical formula for the threshold voltage U_{th} of the domain formation using one elastic constant $K = K_i$ approximation was first derived by Bobylev *et al.* (*6*). Later, Schiller *et al.* (*29*) also proposed another improved formula taking into account the elastic anisotropy of nematic LCs. However, in our work, we adopted the former to confirm the pC m^{-1} order of the flexoelectric coefficients of bent-core mesogens experimentally determined in Section 2.

Denoting the layer thickness, domain period and dielectric anisotropy of the nematic LC by d, w_{th}, $\Delta\varepsilon$, respectively, U_{th} and w_{th} are expressed as follows:

$$U_{th} = \frac{2\pi K}{|e^*|(1+\mu)} \tag{5}$$

$$w_{th} = \frac{d}{\pi}\sqrt{\frac{1+\mu}{1-\mu}} \tag{6}$$

with $\mu = \Delta\varepsilon.\varepsilon_0.K/e^{*2}$ and $e^* = e_1 - e_3$; $K = K_i$. Hence from Equation (6), it is possible to determine the absolute value of the difference $|e_1 - e_3|$ by measuring w_{th}, provided that K and $\Delta\varepsilon$ are already known. It should be emphasised that in order to observe the flexoelectric instability, it is important to choose a compound with a small $\Delta\varepsilon$. In addition, in the case of bent-core LCs, which have negative dielectric anisotropy $\Delta\varepsilon < 0$ in general, the frequency of the field should not be so high as to prevent the electrohydrodynamic instability (electroconvection effect) (*21, 30*).

3.2 *Experiment*

The compound ClPbis10BB was injected into a 5-μm thick homogeneous cell (EHC) with antiparallel rubbing directions in the isotropic phase by capillary suction. Both inner faces of the glass plates were coated with transparent indium tin oxide so that the vertical field could be applied. The cell was then cooled slowly by a temperature controller with an accuracy of 0.1°C and fixed at 74°C, which was half a degree below the Iso-N transition temperature, resulting in a good homogeneous alignment of LC molecules. An AC field was then applied and the texture was observed using a polarising microscope.

3.3 *Results and discussion*

Figure 9(a) shows the texture of ClPbis10BB under a 5-Hz sinusoidal field. We chose an observed temperature not far from the transition temperature, so that the material was in the low dielectric anisotropy $\Delta\varepsilon$ regime, since $\Delta\varepsilon$ increases with decreasing temperature (*31, 32*). Indeed, when the temperature was lower than 74°C, such a longitudinal domain vanished and we could not reproduce it with any other frequencies or field strengths. A similar parallel-stripe domain was also observed by Wiant *et al.* (*21*) when the authors studied the electroconvection effect in the same bent-core liquid crystal, although they did not extract the value of e^*.

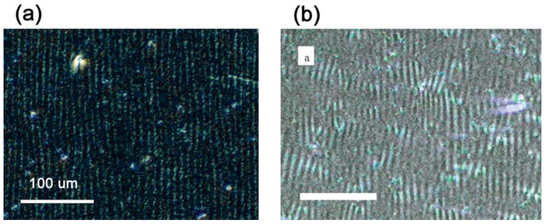

Figure 9. (a) Flexoelectric domain observed in a 5-μm thick cell under a 5-Hz, 4.5-V$_{\text{rms}}$ sinusoidal field; (b) similar domain observed in the same compound in Wiant *et al.* [21], with the length scale being 100 μm. The rubbing directions are in the vertical direction in both photomicrographs.

It should be noted that all our attempts to obtain the flexoelectric domain with some homogeneous alignment layers available at hand, such as SE-7492 (Nissan) and AL-1254 (JSR), failed. We succeeded in observing the flexoelectric domain only when the commercial EHC cell was used, which somehow reproduces similar boundary condition as in the work of Wiant *et al.* (*21*). This coincidence means that the surface anchoring plays an important role on this effect, as discussed by Hinov (*33*) many years ago, when the author studied flexoelectric domains under different boundary conditions.

From Figure 9(a), we found w is about 8.3 μm and using Equation (6), $|e^*| = |e_1 - e_3|$ could be estimated to be about 6 pC m^{-1}. Here we adopted the values $\Delta\varepsilon = -1.65$ and $K = (K_1+K_3)/2 = 2.30 \times 10^{-12}$ N for our calculation (*21*), and assumed that the change in these parameters was negligible. This result implies that e_3 of ClPbis10BB is in the pC m^{-1} order, confirming the result determined by the converse flexoelectric method described in Section 2.

4. Conclusion

Flexoelectricity in a bent-core nematic LC was studied by two independent electrooptic methods. The experimental results revealed that there is not much difference in the flexoelectricity of bent-core mesogens compared with conventional calamitic mesogens. The bend flexoelectric coefficient was measured to be about 15.8 pC m^{-1}, which is about a factor larger than MBBA. This can be explained by the different transverse dipole moments in the two LCs. Another possibility might be the easier and thus stronger close packing of constituent molecules when they have a bent shape. Studying the existence of cybotactic clusters in the nematic phase of bent-core LCs may clarify this intermolecular effect. In fact, such cluster formation has been reported recently in the nematic phase (*34*) and even in the isotropic phase (*20, 35*) of bent-core LCs.

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