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Molecular Shape Related Effects in Liquid Crystals

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Liquid crystals are organic materials constituting of molecules characterized by strong shape anisotropy. Many studies unambiguously showed that some important physical effects observed in liquid crystals are strongly dependent on the shape anisotropy of their molecules.

In this talk, some of the molecular shape related effects in liquid crystals, which have a potential for applications in liquid crystal displays (LCDs) and photonic devices, was overviewed. Photo-induced alignment transition from planar to vertical, flexoelectric polarization in short pitch cholesteric liquid crystals, alignment of bent core nematics as well as field driven transition from birefringent to optically isotropic state in these materials are among those effects on which this talk was focused. The role of the molecular shape was discussed with focus on the impact, which it has on the anchoring, flexoelectric, optical and electro-optical properties of the liquid crystals.

Keywords Photoisomerization; alignment transition; bent core nematics; flexoelectric polarization; optically isotropic state

1. Introduction

An important prerequisite to obtain materials exhibiting mesophases is that their molecules possess high degree of shape anisotropy. Generally, the molecules of the liquid crystal materials are divided in three major groups depending on their molecular shape anisotropy:

- a) calamitic – with rod shape molecules
- b) discotic – with disc shape molecules, and
- c) bent core or so called banana molecules, which may have symmetric or asymmetric arms.

I review and discuss here several effects which are closely related to the shape of the molecules and take place in the simplest liquid crystal phase, nematic, as well as in its chiral variant, cholesteric.

Beside the importance of the molecular shape for the appearance of different mesophases, the molecular shape has also a strong impact on the surface as well as on the bulk properties of the liquid crystals.

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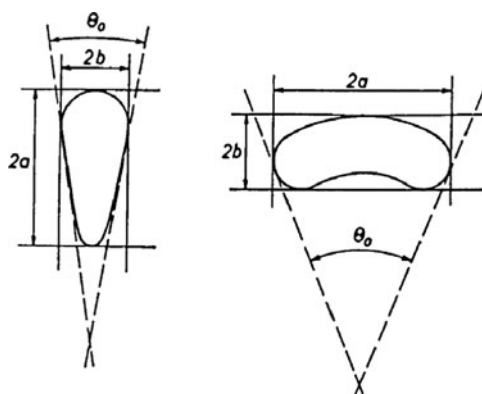
Light-command surface and light-induced alignment transition are good examples of the importance of the shape of the molecules anchored to the solid surface the liquid crystal has contact with. Reversible alignment transitions from vertical to planar or from planar to vertical were demonstrated to be induced by light which resulted change of the shape of the molecules anchored to solid surface from being rod to bend [1].

It has been reported recently that the bent core nematics is very difficult or even impossible to align vertically [2].

Molecular shape asymmetry plays a very important role in the flexoelectric polarization of liquid crystals. Flexoelectricity, as known, is a universal liquid crystal property and it is analogous of the piezoelectric effect in solid materials but quite different in its nature. First described by R. Meyer [3], flexoelectricity means polarization of the liquid crystal as a result of curvature strains. Flexoelectricity is usually pronounced in materials with molecules that in addition to the permanent dipole moment possess also “shape polarity.” The total flexoelectric polarization density in nematics is given by [3]:

$$P_{\text{flexo}} = e_s n(\Delta \cdot n) + e_b n \nabla (\Delta \cdot n), \quad (1)$$

where e_s and e_b are the flexoelectric coefficients for splay and bend elastic deformations, respectively, and n is the liquid crystal director, a unit vector defining the preferred direction of orientation of the liquid crystal molecules. W. Helfrich in 1971 derived the dependence of the flexoelectric coefficients on the molecular shape and permanent dipole moment in their molecular structure [4] according to:



$$e_s = (2p_{\parallel} K_{11}/k_B T) L_s N^{1/3}, \text{ with } L_s = \theta_o (a/b)^{1/3} \quad (2)$$

$$e_b = (p_{\perp} K_{33}/k_B T) L_b N^{1/3}, \text{ with } L_b = \theta_o (a/b)^{2/3} \quad (3)$$

where p_{\parallel} and p_{\perp} are the net dipole moment parallel and perpendicular to the molecule long axis, respectively. The degree of the molecular asymmetry is described by the coefficients L_s , for drop shape molecules, and L_b , for banana (bent) shape molecules, with a and b being the length and width of the molecule, respectively. θ_o represents the opening angle for drop shape molecule and the kink angle in the bent core molecule. K_{11} and K_{33} are splay and bend elastic constant, respectively. N is the number density of the molecules (Avogadro constant) and k_B is the Boltzmann constant. From Eq. 2 and 3 it follows that the bend flexoelectric coefficient e_b has stronger dependence on the molecular asymmetry, represented by the ratio a/b , than e_s , and thus being more sensitive to changes in the molecular shape.

Closely related to the molecular shape anisotropy is the peculiar optical behavior of the liquid crystals with bend shape presented recently [5, 6]. With such molecular shape are the molecules of the bimesogenic antiferroelectric liquid crystals (AFLC). Their molecules are dimers whose arms form a bow having an angle close or equal to 90° . When such AFLC is aligned in bookshelf texture, with smectic layers oriented normal to the cell substrates and with molecules lying parallel to substrates, then such a sample exhibits optically isotropic state when the angle between the molecular arms becomes 90° since the birefringence of the arms is compensating each other. As we shall see, the same behavior was found in bent core nematics and their mixtures with calamitic nematics.

2. Experimental Results and Discussion

As known, there is a group of liquid crystal materials, so called photochromic liquid crystals, whose molecules undergo reversible *trans* to *cis* photo-isomerization. Typical representative of this group are azo-compounds.

As a first example for the important impact of the molecular shape on the physical properties of the nematic liquid crystal, it will be given the light-induced alignment transition due to photoisomerization of nematic liquid crystal azo-dye: 4-hexyloxy-(4'-hexyl)azobenzene with structure presented in Fig. 1. This azo-dye undergoes *trans*- to *cis*-isomerization upon illumination with UV light transforming the linear form of the *trans*-isomer into V shaped *cis*-isomer (see Fig. 2). In addition to the molecular shape transformation also the molecular net dipole moment changes resulting in higher polarity of the azo-group [7]. The *cis*-isomer is generated under UV illumination in the volume as well as at the surface and its concentration depends on the exposure time. However, the concentration of *cis*-isomer at the surface increases much faster with the exposure time than the one in the volume since the *cis*-isomer possesses a larger transverse dipole moment than the *trans*-isomer and their adsorption on the substrate surface is more favorable (selective adsorption) (see Fig. 3a). Due to the molecular V shape, the *cis*-isomer does not favor the liquid crystalline molecular order. Therefore, the nematic state transforms into isotropic above a critical concentration of the *cis*-isomers in the liquid crystal volume [8]. Below this critical *cis*-concentration, however, a transition from the initial planar to vertical alignment is observed to take place in a cell whose substrates are hydrophilic, as the one covered by SiOx, for instance (c.f. Fig. 3b). Due to the V shape, the *cis*-isomer anchored to the surface will affect the anchoring of the liquid crystal in a similar way as the surface-active agents do and above a certain surface concentration, the *cis*-isomer will promote a vertical (homeotropic) alignment of the nematic liquid crystal [9]. Until the critical surface concentration of the *cis*-isomers for planar to homeotropic alignment transition is reached, there is a continuous change of the anchoring strength with the UV-exposure time due to the selective adsorption process of *cis*-isomers onto the substrate surface [10] (see Fig. 4).

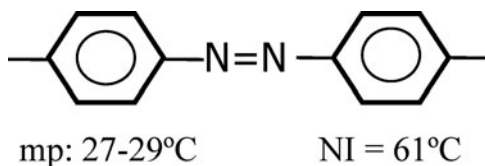


Figure 1. Azo-dye 4-hexyloxy-(4'-hexyl)azobenzene possessing a nematic phase.

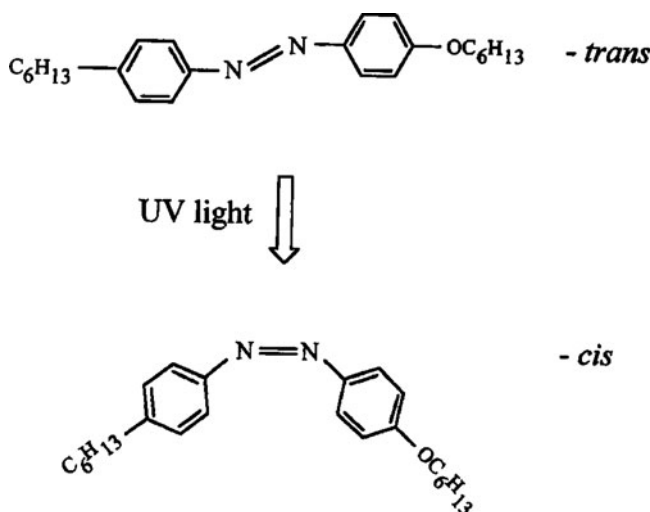


Figure 2. Photoisomerization of 4-hexyloxy-(4'-hexyl)azobenzene upon illumination with UV light of $\lambda = 365$ nm.

The importance of the molecular shape for the alignment of the nematic liquid crystals has been recently demonstrated [2]. It is found that the substrate surface treatment, which usually promoted vertical alignment of the rod like molecules of calamitic nematic liquid crystals, does not promote such an alignment of bent core nematics due to the bent shape of the molecules, as shown in Fig. 5. Even mixtures of bent core nematics (60 wt%) and calamitic nematics (40 wt%) could not be aligned vertically. This is because once the bent core molecules are adsorbed onto the surface they screen the rest of the liquid crystal bulk from the substrate surfaces treated to promote vertical alignment.

Another important example, demonstrating the impact of the molecular shape on the physical properties of nematic liquid crystals is the light-induced changes in the flexoelectric properties of photochromic nematic liquid crystals.

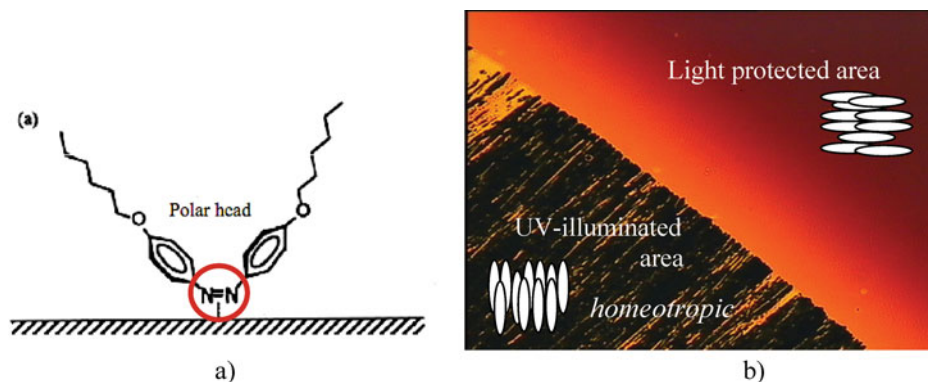


Figure 3. a) Selective adsorption of the *cis*-isomer of 4-hexyloxy-(4'-hexyl)azobenzene upon illumination with UV light of $\lambda = 365$ nm. b) Cell with light protected and illuminated area, respectively. In the protected area the alignment is planar whereas in the illuminated area is vertical (homeotropic).

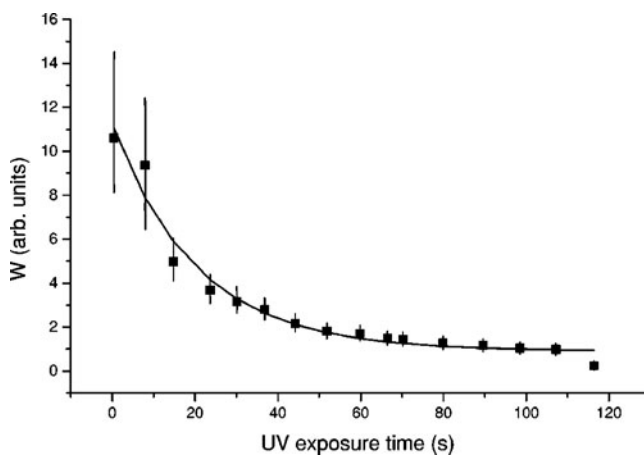


Figure 4. Anchoring strength as a function of the illumination with UV light of $\lambda = 365$ nm.

Due to the photo-isomerization, the molecular net dipole moment is usually changing whereas the molecular shape may or may not change. By studying the impact of the photo-isomerization process on the physical properties of the liquid crystals one could evaluate and at the same time separate the role of the molecular shape and the one of the net molecular moment on these properties and related to them effects in liquid crystals, such as flexoelectric polarization, for instance.

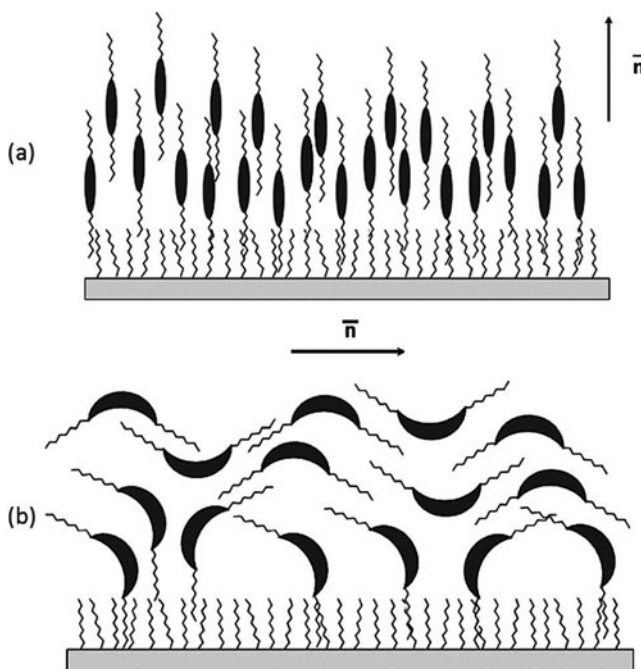
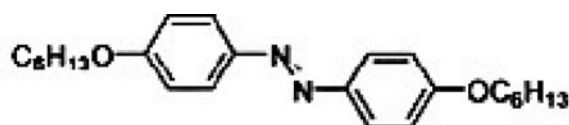


Figure 5. Alignment of rod like (a) and bent core (b) molecules by polyimide alignment layer for promoting vertical alignment of calamitics.

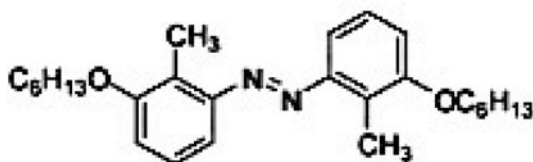
The liquid crystalline azo dye 4-hexyloxy-(4'hexyl)azobenzene shown in Fig. 1 is dissolved in the commercial nematic mixture E7 (BDH) in ratio 95 wt%(E7)/5 wt%(azo dye). The azo dye undergoes *trans*- to *cis*-isomerization under illumination with UV light with wavelength of $\lambda = 365$ nm. As seen, the molecular shape changes from being linear in *trans*-conformation to become strongly bent in the *cis*-conformation. The net molecular dipole moment also changes from 0.5D in *trans*-conformation to 3.5D in *cis*-form. The study of the light-induced changes of the flexoelectric polarization was carried out in a cell with hybrid aligned nematic (HAN) configuration, according the method described in [11]. It was found that the magnitude of $(e_s - e_b)/K$, representing the flexoelectric polarization where K is the average elastic constant, increased with 40% after UV illumination [12]. Such an increase of $(e_s - e_b)/K$ corresponds to increase of the flexoelectric polarization of the pure azo dye more than an order after illumination with UV light. It should be noted here that usually the *cis*-isomer does not favor the liquid crystalline order, therefore only up to certain percentage this isomer could be appropriate for use.

Since, in the above described case, both the molecular shape and the net molecular order are changing due to photoisomerization, it is difficult to separate the role of each of them in the enhancement of the flexoelectric polarization. In order to do that another experiment involving two azo dyes, namely, 4-4'-azo and 3,3'-azo, were used as dopants. These dyes are possessing very similar molecular structure but they are exhibiting different photo isomerization behavior [13]. The dyes' molecular structure and their *cis*-form are shown in Fig. 6. Each of these dyes was dissolved in a short pitch cholesteric liquid crystal host material with 5 wt% concentration of the dye. The cholesteric liquid crystal host was a binary mixture of short pitch cholesteric CF1 ($p = 0.3\mu\text{m}$) (kindly supplied by M. Schadt, Hoffmann La Roche & Co.) and 8OCB (octyloxycyanobiphenyl) at ratio 89.5 wt% and 10.5 wt%, respectively.

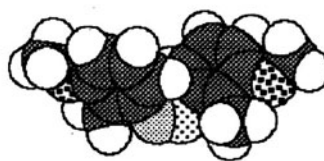
The cholesteric material CF1 has temperature independent short pitch and exhibits a peculiar property, i.e., a sign reversal of $(e_s - e_b)$ at temperature of about 28°C , thus possessing zero flexoelectric polarization at this temperature. Therefore, this compound is useful to be used as a chiral host material for evaluating the flexoelectric polarization of liquid crystals. In the host mixtures were dissolved of about 5 wt% of each azo dye. Such a low percentage of the dyes was chosen in order to avoid a light-induced nematic to isotropic phase transition. Whereas the dyes' net molecular moments have very similar magnitudes, before (0.1D) and after photo-isomerization (about 5D), their molecular shape is completely different after photo-isomerization, as seen in Fig. 6. Such a photo-isomerization behavior of the azo dyes with almost identical molecular structures enables the evaluation of the impact on the molecular shape of these dyes on the flexoelectric polarization of their liquid crystal mixtures. The changes of the flexoelectric polarization due to photo-isomerization were evaluated by means of the method described in [14]. The method is based on the flexoelectro-optic effect in cholesteric liquid crystals oriented in uniform lying helix (ULH) texture and it has an important advantage over the existing methods since a high frequency electric field is applied and therefore no any complications related to generation and separation of charges occur when this method is applied. It was found that the changes in the molecular shape from linear *trans*-form to bend *cis*-form resulted in an increase of the flexoelectric polarization $(e_s - e_b)/K$ with 20% whereas the increase of the magnitude of the molecular net dipole moment due to photo-isomerization, which in this case was more than one order, the increase of the flexoelectric polarization was only 4% [13]! Hence we may conclude that the molecular shape anisotropy plays a vital role for the flexoelectric polarization of the liquid crystal. The higher is the molecular shape asymmetry the higher is the flexoelectric polarization. The mentioned above 20% and 40% increase of the magnitude of the flexoelectric polarization

**4,4'-azo**

K 86 °C S 98 °C N 110 °C I

**3,3'-azo**

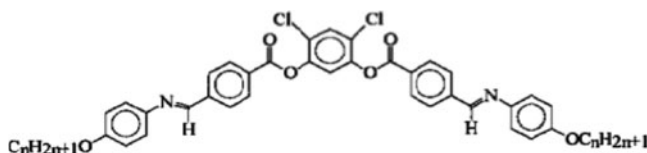
K 58 °C S 105 °C I

**cis-4,4'-azo****cis-3,3'-azo****Figure 6.** Molecular structure of 4,4'-azo and 3,3'-azo dyes and their *cis*-isomers.

in two photochromic nematic mixtures containing 5 wt% different azo dyes, indicate that, due to photoisomerization resulting in change of the molecular shape from rod to bend like, the flexoelectric polarization of the pure azo dopants, increase more than an order (about 30 times).

Another similar evaluation of the influence of the molecular shape on the flexoelectric polarization was performed with a short pitch cholesteric host in which is dissolved bent core nematic as a dopant. The host mixture contained 90 wt% short pitch cholesteric CF1 and 10 wt% MBBA. In this mixture is dissolved 5 wt% bent core dopant (kindly supplied by W. Weissflog) with the structure shown in Fig. 7. The measurements are performed with the method described in [14]. The obtained magnitude of $(e_s - e_b)/K$ after doping with the bent core dopant was found to increase from $17 \times 10^{-3} \text{ C/Nm}$ to $56 \times 10^{-3} \text{ C/Nm}$ i.e. 5 wt% concentration of the dopant increased the magnitude of $(e_s - e_b)/K$ of about 3 times, which corresponds to more than one order of magnitude, i.e., about 60 times larger flexoelectric polarization of the pure dopant material than the one of the short pitch host [15]. This result is in a good agreement with the recently performed studies on the flexoelectricity of bend core nematics reported in [16].

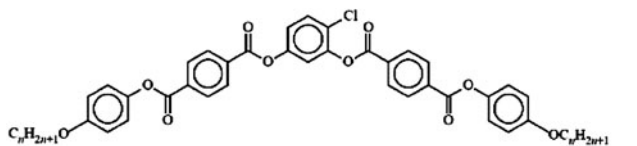
The last but not the least important example of the impact of the molecular shape is the electro-optical behavior of bent core nematics and their mixtures with calamitic nematics.



with $n = 12$

Figure 7. Molecular structure of the bent core dopant.

Bent core (BC) liquid crystal molecules are molecules which consist of two arms, which could be symmetric or asymmetric, connected by a central group in such way that the arms make an angle. Thus, their molecular shape looks like banana (symmetric arms) or hockey stick (asymmetric arms). Such an example is 4-chloro-1,3 Phenylene bis{4-(9 decenyloxy)-1,1-biphenyl-4 carboxylate (CIPbis10BB) whose molecular structure (see Fig. 8) possesses a net dipole moment, parallel to the Cl atom, inclined of about 45° with respect to the central phenyl ring [17]. The opening angle between the molecular arms is 137° . This material has negative dielectric anisotropy ($\Delta\epsilon = -1.7$), according to [18], which means that the planar alignment of the BC nematic liquid crystal in a conventional sandwich cells will be only stabilized by an electric field applied across the cell gap due to the dielectric coupling of the negative dielectric anisotropy $\Delta\epsilon < 0$ of the BC nematic with the applied dc electric field. However, it was found that the dc field induced out-of-plane switching of the bent core molecules contrary to what was expected, no switching at all [19]. As the origin of the out-of-plane switching of the molecules of the bent core nematic was suggested to be the induced bias of the free rotation of the bent core molecules around their long axis, due to the coupling between the molecular net dipole moment and applied dc electric field. Another important factor for the out-of-plane switching is the presence of molecular clusters in the nematic phase of the bent core material, possessing a short-range ordering, the so-called cybotactic group with smectic order with size of 30–50 nm [20]. These clusters may exist even in the mixtures of BC nematic with up to 40 wt% calamitic (with rod like molecules) nematic. It was assumed that these clusters might possess a surface polarization [21] due to the smectic layering. Then the applied dc field may result in out-of-plane switching of the clusters. Hence, these two factors, bias of the molecular free rotation around their long axis and the orientation of the clusters by the applied dc electric field will result in out-of-plane orientation of the bent core molecules. Importantly, such an out-of-plane switching of the bent core molecules is accompanied by orientation of the two bent core molecule's arms in a plane (molecular plane), parallel to the molecular long axis,



$n = 10$; Cry 93°C N 96°C (on heating); Iso 100°C N Cry 80°C (on cooling)

Figure 8. Molecular structure of the bent core nematic 4-chloro-1,3 phenylene bis{4-(9 decenyloxy)-1,1-biphenyl-4 carboxylate (CIPbis10BB) and the temperature interval of the nematic phase.

which makes the bent core nematic biaxial. Therefore, once the dc electric field is applied, the bent core nematic starts to behave optically likewise the bimesogenic AFLCs, due to the similarity in their molecular structures [6, 19]. Hence, the optical properties of the bent core nematic can be described, likewise the bimesogenic AFLC, as a sum of two biaxial dielectric ellipsoids each of them representing the optical property of the mesogenic arms of bent core molecules. According to Ref. [6], this sum is given by:

$$\varepsilon_{anticl} = \frac{1}{2} [\varepsilon_{anticl}(+\theta) + \varepsilon_{anticl}(-\theta)] = \begin{bmatrix} \varepsilon_{\perp} \cos^2 \theta + \varepsilon_{\parallel} \sin^2 \theta & 0 & 0 \\ 0 & \varepsilon_{\perp} & 0 \\ 0 & 0 & \varepsilon_{\perp} \sin^2 \theta + \varepsilon_{\parallel} \cos^2 \theta \end{bmatrix} \quad (4)$$

where $\varepsilon_{\text{sincl}}(+\theta)$ and $\varepsilon_{\text{sincl}}(-\theta)$ are the respective (biaxial) dielectric tensors of each arm and θ is the half of the angle between the molecular arms.

Due to the polar coupling of the molecular net dipole moment and the polarization of the cybotactic groups with the applied dc electric field, the BC molecules will switch out-of the plane thus the molecular plane adopts a tilt with respect to the substrate, which increases with the field. This tilt seems to saturate at 45° tilt of the BC molecules i.e. as much as the inclination of the net dipole moment in the BC molecular structure with respect to the central phenyl ring is. Out-of-plane switching of the bent core molecules from field-off planar alignment strongly affects the optical appearance of the bent core nematic sample (sandwich cell filled with bent core nematic). The bright field-off state of the samples, inserted between two crossed polarizers with the preferred alignment direction oriented at 45° with respect to the polarizers, continuously changes to dark state with the applied dc voltage (c.f. Fig. 9) and at higher voltage becomes again slightly birefringent with grayish optical appearance (see Fig. 10). Importantly, the dark state, which appeared as a result of the out-of plane switching of the molecules of the BC nematic from the planar alignment, did not change its contrast upon rotation of the cell in between the crossed polarizer around the normal of the cell substrates, i.e., it appeared optically isotropic (Fig. 9).

This is because for the optical properties of the sample is important the angle between the projections of the bent core molecules' arms on the cell substrates. When increasing the applied dc voltage, the tilt of the molecular plane increases whereas the angle between the molecules' arms projection decreases from 137°, in field-off state, to reach 90° at certain voltage (in the case shown in Fig. 10 this voltage is 4.2 V). Then according to Eq. 4 the birefringence of the sample becomes zero, i.e., the sample exhibits optically isotropic state.

Increasing further the applied voltage results in decreasing of the angle between the molecules' arms projections which in turn give rise to birefringence, i.e., the completely dark state transforms to a grayish one exhibiting low birefringence (see Fig. 10). It should be noted here that in the bright state the slow axis of the sample is oriented along the director preferred direction of alignment, i.e., along the rubbing direction. In the dark state, however, the slow axis of the sample is oriented perpendicular to the cell substrates therefore the cell appears to be optically isotropic when rotated in between crossed polarizers. At higher voltages, when the sample shows weak birefringent state (grayish state), the slow axis is parallel to the substrate but perpendicular to the rubbing direction. This was confirmed by means of inserting λ red optical plate between the crossed polarizers at 45° with respect to the transmission direction of the polarizers [6, 22].

Similar electro-optical behavior was found for binary mixtures of bent core nematics with calamitic nematics containing up to 40 wt% of the calamitic nematic compound [22].

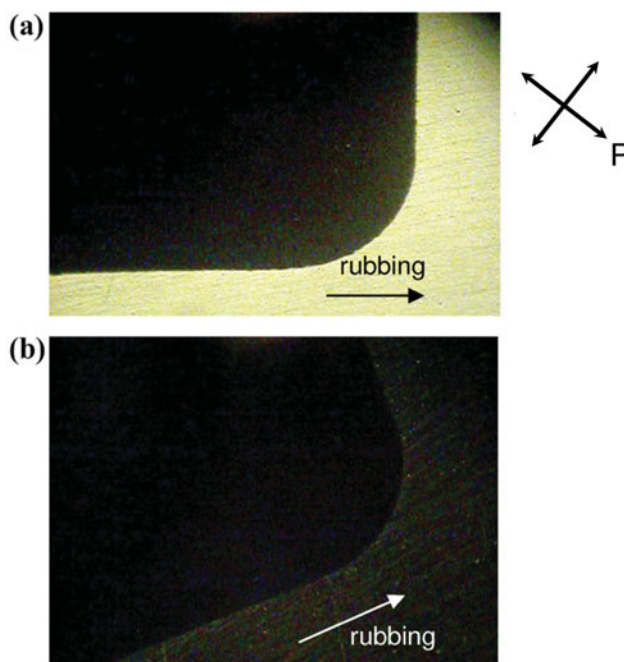


Figure 9. Photograph of a cell with cell gap of $1.87 \mu\text{m}$, containing the BC nematic material from Fig. 8, at 88°C . The cell is inserted between crossed polarizers with optic axis at a) 45° and b) 0° with respect to the transmission direction of the polarizer. Dc electric field of $2.5 \text{ V}/\mu\text{m}$ is applied to the cell. As seen, the dark state in the pixel is superb and it does not change when the cell is rotated at 45° between the crossed polarizers, indicating thus field-induced optically isotropic state in the pixel.

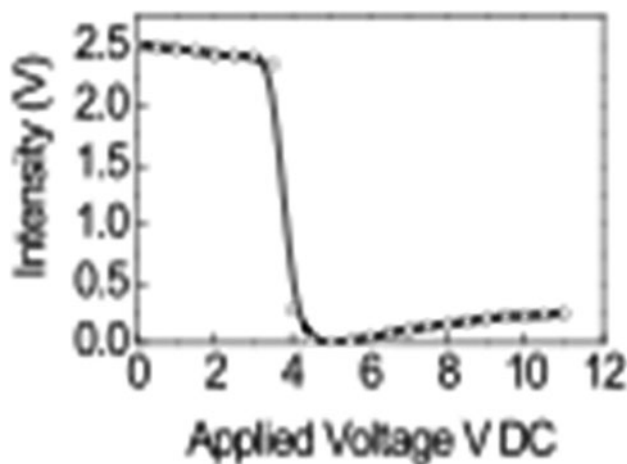


Figure 10. The light transmitted intensity versus the applied voltage for the sample from Fig. 9 at the same temperature 88°C . The minimum intensity of the transmitted light was achieved at applied field of about $2.5 \text{ V}/\mu\text{m}$ at which the cell exhibited isotropic optical properties. With the further increase of the field the isotropic optical state was transformed to slightly birefringent (grayish) state.

3. Conclusion

The overview made in this paper shows that the molecular shape has an important impact on the physical properties of the nematic liquid crystals and the effects related to these properties. The examples given here, unambiguously demonstrate the important role of the molecular shape.

Surface and bulk liquid crystal properties showed strong dependence on the molecular shape. The photo-induced transition from planar to vertical (homeotropic) alignment indicated that the molecular shape of the molecules adsorbed onto substrate surface, the liquid crystal has contact with, strongly affect the alignment of the molecules in the liquid crystal bulk. The *cis*-isomers, due to their V shape, trigger alignment transition from planar to vertical alignment when their surface concentration exceed a certain level.

Strong impact of the molecular shape of bent core molecules on the vertical alignment of the bent core nematic and their mixtures with calamitic nematic was also found. It became clear that the conventional alignment methods applied for calamitic nematics to obtain vertical alignment do not promote such alignment of bent core nematics.

Molecular shape was found to have a strong impact on the flexoelectric properties of the nematic liquid crystals. The bent shape of the molecules may increase the magnitude of the flexoelectric polarization $(e_s - e_b)/K$ more than one order. Therefore the bent core nematic seems to be a promising candidate for liquid crystals with enhanced flexoelectric polarization.

Due to the polar coupling between the net molecular dipole moment and polarization of the clusters with the applied dc electric field, the out-of-plane switching of the bent core nematics and their mixtures with calamitic nematics, gave rise to an optically isotropic state in these materials at low magnitude of the applied voltage. The generated optically isotropic state enhances substantially the contrast of the sample containing a bent core nematic or a mixture of bent core and calamitic nematic.

Importantly during the switching process under the applied dc field, the bent core nematic liquid crystal sample slow axis changed its position between three orthogonal positions: a) parallel to the substrate plane and along the rubbing direction, b) perpendicular to the substrate and c) parallel to the substrate and perpendicular to the rubbing direction.

All the reviewed effects have a great potential for application in liquid crystal displays and photonic devices, each of them within a specific niche.

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