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Electric field induced biaxiality and the electro-optic effect in a bent-core nematic liquid crystal

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We report the observation of a biaxial nematic phase in a bent-core molecular system using polarizing microscopy, electro-optics, and dielectric spectroscopy, where we find that the biaxiality exists on a microscopic scale. An application of electric field induces a macroscopic biaxiality and in consequence gives rise to electro-optic switching. This electro-optic effect shows significant potential in applications for displays due to its fast high-contrast response. The observed electro-optic switching is explained in terms of the interaction of the ferroelectric clusters with the electric field. © 2010 American Institute of Physics. [doi:10.1063/1.3280817]

Based on the generalized Maier–Saupé theory, Freiser1 predicted that a reduction in the molecular symmetry could lead to the formation of a biaxial nematic ($N_{b}$) phase in liquid crystals (LCs) in addition to the observed uniaxial nematic phase. The biaxial nematic phase has a secondary director $m$ perpendicular to the primary director $n$; the latter is always prevalent in the nematic phase. From a practical point of view, the switching of the secondary director is at least a factor of 100 faster than the primary one. Therefore such biaxial nematics with improved response offer significant advantages over the conventional nematics in applications for fast displays and photonic devices. The potential for faster devices has led to a significantly increased interest in theoretical, experimental, and computational studies on the biaxial nematic phase. The experimental proof for the existence of the $N_{b}$ phase in a lyotropic LC system was reported in 1980.2 The thermotropic biaxial nematic phase was discovered in 2004 in liquid crystalline polymers,3 organosiloxane tetrapodes,4,5 and in low mass bent-core systems.6–8 The possibility for the existence of a biaxial nematic phase in biaxial parallelepiped (or bricklike) shaped molecules9–11 and in bent-core systems12–14 has been supported by numerous theoretical studies.

Experimental studies on a part of the temperature range of nematic phase of the bent-core LCs suggest the appearance of “cybotactic” (smecticlike) clusters.15–21 In such molecules, two rodlike mesogenic groups linked together through a central unit exhibit a near-$C_{2v}$ symmetry. The presence of biaxial clusters was considered a long time ago by De Vries.22 In bent-core systems, a deviation from the calamitic shape and an existence of a large transverse dipole moment gives rise to unusually strong intermolecular interactions that lead to the formation of ferroelectric domains or clusters. On applying the electric field, the collective alignment of domains leads to a macroscopically biaxial ordering.18 On removal of the field the domains are destabilized, with a corresponding drop in the biaxial order parameter. It is still not clear whether such a nematic LC consists of distinguishable clusters of a lower symmetry phase, or correlated regions with a short-range order.

In spite of the obvious progress made in both experimental and theoretical studies, a number of problems remain to be solved23 and therefore the subject of biaxial nematics continues to be a highly debated and challenging in the field of LCs. Despite numerous claims for the existence of biaxial nematic phase in different systems, electro-optic effect has so far not been demonstrated unambiguously. In this letter, we report results of the experimental study of electro-optic effect in a homeotropically aligned biaxial nematic compound. The observed electro-optic switching is explained in terms of the interaction of the ferroelectric clusters with the electric field.

The bent-core LC sample under study, C$_{6}$-BAN, is synthesized in Halle, Germany. The molecular structure of the studied material is given in Fig. 1. For the optical and electro-optical studies the compound was filled in homeotropically aligned cells of different thicknesses, varying from 4 to 50 $\mu$m. The foil strips of different thicknesses were used as electrodes in order to apply the external electric field parallel to the plane of the glass plates. The gap between the electrodes was fixed to be of the order of $\sim$200 $\mu$m. AL60702 (JSR Japan) was used to achieve the homeotropic alignment.

The optical textures for three different thicknesses (50, 25, and 4 $\mu$m) of the sample are presented in Figs. 1(a)–1(c). On cooling from the isotropic to nematic phase in a thicker (50 $\mu$m) cell, the nematic phase is found to consist

![FIG. 1. (Color online) Microphotographs of the textures near T$_{NI}$: The cell thicknesses are $d=50$ $\mu$m for (a) and (d); $d=25$ $\mu$m for (b) and (e); and $d=4$ $\mu$m for (c) and (f). Textures (d), (e), and (f) are recorded after a period of 1 h from those of (a), (b), and (c).](image-url)
only two-brush Schlieren texture [Fig. 1(a)] which indicates the presence of biaxial nematic phase. Nevertheless this biaxial nematic texture is not stable with time and during a period of one hour it transforms to a rather uniform dark texture [Fig. 1(d)], which nevertheless consists of very small domains of the approximate size of the wavelength of visible light. These considerably reduce the overall extinction of the texture compared with that for the isotropic phase. We assign these domains to cybotactic clusters, which are observed in this sample by x-ray diffraction.24 It has been found that the transition from cybotactic nematic to smectic phases occurs on elongation of the chains and the clusters in this compound are observed with three to four molecules correlated in the transverse direction, and about two molecules correlated parallel to the director.24 Textures in these samples are found to be strongly affected by surfaces. Thus a thin (4 μm) LC cell finally produces a perfect high-extinction homeotropic alignment [Fig. 1(f)]. The textures produced by the cell of intermediate thickness [Figs. 1(b) and 1(e)] are somehow between those for the two previous cells.

The effect of electric field on the textures is studied in a 4 μm cell with foil spacers as electrodes. A square-wave field 100 Hz is applied across the electrodes in the plane of a glass plate. The cell was placed between the crossed polarizers with an angle of 45° between the electric field direction and polarizer axis. Initially for a rather small electric field (<0.3 V/μm), no significant effect is observed and the LC remains optically uniaxial. On the application of a stronger electric field, the cell exhibits electro-optical switching from dark to bright state and reverse when the field is removed (Fig. 2). The electro-optic response shows dynamics with different speeds; slow and fast. These suggest coexistence of the two different processes. This suggestion is supported by observations under a polarizing microscope (Fig. 3). Figure 3(a) shows the texture for an applied electric field of 0.5 V/μm. On the removal of the field, the cell relaxes to an apparently initial macroscopically uniaxial state [Fig. 3(c)].

The magnitude of the induced biaxiality was measured using a tilting optical compensator inserted in the polarizing microscope. This allows one to measure rather small values of the optical retardation for a specific region of the cell, which in our case is the center between the electrodes of the foil (Fig. 3). Figure 4(a) shows the dependence of the induced biaxiality (δn) as a function of electric field for different temperatures using a homeotropically aligned cell of thickness 4 μm. Initially (after a threshold field of ~0.3 V/μm is applied), the induced biaxiality increases gradually with electric field and then saturates to a constant value.

The magnitude of the induced biaxiality decreases with a reduction in temperature, which shows that this phase disappears at temperatures below 97 °C. Figure 4(b) presents the dependence of the induced biaxiality (δn) as a function of frequency for an electric field of 0.7 V/μm. On cooling from 106 to 97 °C, the cut-off frequency of the electro-optic
A question arises as to what is the phase assignment for tempera-
tion processes. Both spectra show a relaxation process
molecular axes corresponding to the molecular relaxation around the short
frequency range of 100 Hz to 10 MHz. The dashed-line denotes fitting of the
two relaxation processes and the dotted-line shows the conductivity for a
temperature of 100 °C.

The dielectric study was performed on a planar cell of
9 μm thickness using a broadband high resolution dielectric
spectrometer (Novocontrol GmbH, Germany). Experiments
are performed on cooling the sample from 115 to 60 °C. Figure 5 shows the dielectric loss spectra in the frequency of
10 Hz–10 MHz where the EO response disappears. Dielec-
tric loss spectra (ε″) were fitted to the two relaxation pro-
cesses. Both spectra show a relaxation process (P1) corre-
sponding to the molecular relaxation around the short
molecular axes (flip-flop mode) similar to other uniaxial
nematics. The spectra at a temperature of 96 °C can well be
fitted by a single relaxation process.

The second spectra for a temperature of 100 °C show an
additional process (P2), Fig. 5. This process is observed in
the same temperature range (97–106 °C), where the fast EO
response is present. Therefore we can relate this definitely to
the polar dielectrically active process observed in the EO
switching. The two switched-ON states of the opposite po-
larities (+/-) are optically indistinguishable for the reason that
the switching mechanism involves the polar reorientations of
the microscopic biaxial domains in the direction of the ap-
plied electric field irrespective of its direction. Therefore the
EO effect (Fig. 4) is observed between the field induced
biaxial (ON) state and the uniaxial (OFF) state.

For temperatures below 97 °C, the sample exhibits nei-
ther the dielectric process (P2) nor the fast electro-optic re-
response. This indicates the absence of the polar clusters. Nev-
evertheless neither texture nor the DSC shows any phase
transition at 97 °C. The cybotactic clusters have also been
found to exist in the temperature range below 97 °C.24 The
question arises as what is the phase assignment for tempera-
tures below 97 °C. The plausible explanation is a gradual
formation of cybotactic clusters with antiferroelectric order-
ing as the temperature decreases. This process lowers the
fraction of the clusters with ferroelectric orderings and hence
the magnitude of the EO (Fig. 3) and dielectric amplitude (Δε, Fig. 5) are reduced significantly. At a temperature of
97 °C all ferroelectric clusters transform to antiferroelectric
ones. The coexistence of both ferroelectric and antiferroelec-
tric clusters is in agreement with the results of Liao et al.16
An additional (polar) relaxation process in dielectric spectra
is observed even in the absence of the bias voltage (Fig. 5
and Ref. 16) and the intensity of this process increases sig-
nificantly after application of large bias field due to field
induced antiferroelectric to ferroelectric transition16 and for-
mation of ferroelectric clusters.

In summary, results on nematic phase for a bent-core LC
system show that the nematic phase containing biaxial clus-
ters is macroscopically uniaxial but the biaxiality is shown to
be induced by the electric field. The electro-optic switching
is shown to occur via the short axes. Such LCs, in their
intermediate induced biaxial phase can be exploited for ap-
plications due to fast field-induced (~1 ms) switching be-
tween the uniaxial and biaxial states thus leading to a new
concept for the devices.

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