

Time Resolved Experimental Analysis of the Electric Field Induced Biaxial Order Reconstruction in Nematics

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We present the time resolved experimental characterization of the biaxial switching between two topologically distinct textures of a nematic liquid crystal cell submitted to a strong electric field. This fast electro-optical effect is governed by the electric induced order reconstruction in the nematic bulk, which is an actual challenge for a complete theoretical description of the nematodynamics. The electric measurements across the cell are suitable to this purpose since they well discriminate among the dielectric, the ionic, and the order reconstruction contributions. A phenomenological model describes the experimental data, allowing the measurements of the order reconstruction characteristic time.

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The electro-optical effects of nematic liquid crystals (NLC) are mainly based upon electric field induced texture distortions of these anisotropic fluids. The NLC alignment is usually described by the director field $\mathbf{n}(\mathbf{r})$, where \mathbf{r} is the position and \mathbf{n} represents the average orientation of the molecular long axis. Using this approach, a huge research effort has been done to study the NLC reorientations under electric field, that is, weak distortions, textural bulk bifurcations, and surface bifurcations. The best known textural bulk bifurcation appears above a low threshold characteristic of the well known Fréedericksz transition [1], whereas surface bifurcations are related to the anchoring breaking phenomenon [2,3].

The Fréedericksz transition, and in general the nematic elastic distortions, preserve the starting textural topology. The anchoring breaking, on the contrary, is compatible with an intrinsic textural bistability, because the surface reorientation allows the change of the topology of the starting texture, as in the well known case of the transition between a planar texture and a π -twisted one [4].

Very recently, the controlled nematic order reconstruction under electric field has been presented as a new tool to achieve nematic textural changes with variable topology, also allowing intrinsic textural bistability. The order reconstruction changes the nematic director orientation of $\pi/2$ by exchanging two eigenvalues of the nematic order tensor [5,6]. The average molecular reorientation is now obtained without any director rotation, but with a suitable deformation of the ellipsoid which represents the nematic order.

Recently, this fast transition has been investigated by means of electro-optical experiments and these results have been discussed by using a static model of biaxial melting [7]. Nevertheless, at present, the experimental characterization of this nematic bulk transition lacks details. The intrinsically slow optical response of the nematic material allows showing the existence of the

transition, by means of birefringence measurements [7], but it is not useful for the characterization of the fast dynamics itself. Further theoretical and experimental results have just introduced a full dynamical description based on the nematic order reconstruction [6,8] and a characterization method by means of electric current observations [9].

In this Letter, we present novel experimental observations which allow measuring the order reconstruction characteristic time, a critical parameter to quantify any theoretical model.

Figure 1 shows the transition cascade from the quasi-planar H state to the bent V state in the case of the nematic order reconstruction effect, in an initially slightly splayed nematic layer.

In the H state, the director in the cell is oriented mostly parallel to the plates, while in the V state is aligned perpendicularly, moreover H and V have not the same topology. For a nematic with positive dielectric anisotropy, the transition from H to V happens in the presence of a strong vertical electric field, through the intermediate state I characterized by a thin wall, in the middle of the cell, where we expect that the order reconstruction takes place.

To experimentally investigate this phenomenon, the cell is made by two parallel transparent indium tin oxide (ITO) coated glasses which contain the nematic liquid

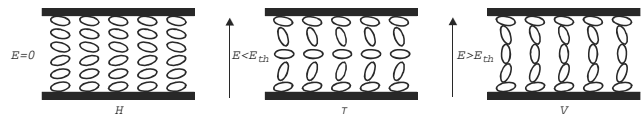


FIG. 1. Transition cascade from the quasiplanar H state to the bent V texture induced by a vertical electric field, via the intermediate I configuration. The I state is characterized by a thin wall, in the middle of the cell, where the order reconstruction occurs.

crystal 5CB (4-cyano-4'-n-pentylbiphenyl), that has a strong positive dielectric anisotropy ($\epsilon_a \sim 12$ at a temperature of about 25 °C [10]). The electrodes on the two boundary plates are 2 mm width lines and they are realized by photolithography of the ITO films: their crossed superposition gives one pixel of about 4 mm² area. The oblique symmetrical anchoring on the two boundary plates is obtained by polymeric coating, i.e., rubbed polyimide, and the resulting anchoring strength is practically infinite, with a small pretilt of few degrees. The cell thickness is $d = 2.40 \pm 0.05 \mu\text{m}$ measured by a standard interference technique before filling it with 5CB.

Rectangular electric pulses of variable amplitude and fixed width $\tau_p = 1$ ms are applied to the cell, which is connected in series with a 5 k Ω electric resistance: its voltage drop V_r allows to measure the electric current which flows through the sample. The sample is directly observed by means of a polarized microscope. We perform synchronous acquisitions of the electric current and light transmission across the sample, when the field is applied to the cell. The transmitted light intensity is measured by a photomultiplier also connected to the polarized microscope. The plate of the microscope is rotated to align the optical axis of the sample in the starting *H* configuration at 45° with respect to the optical axis of the crossed polarizers. In fact, this is the well known condition of maximum intensity for the transmitted light in presence of a birefringent sample. The electric current data and the light intensity transmitted by the sample are acquired by a PC connected via a GPIB interface.

Figure 2 shows the typical optical and electric responses of our sample at a temperature close to the nematic-isotropic transition, during the electric pulse application: the amplitude of the rectangular pulse is well above the threshold for the *H-V* transition. These experimental data are directly comparable with the experimental observations reported in Refs. [7,9]. The upper signal is the transmitted light intensity and the lower signal is the electric current response, which flows across the sample, vs time. The temporal acquisition starts when the electric field is switched on. During the electric pulse application, in principle, if the field is strong enough, the transmitted light intensity should vanish, due to the molecular dielectric reorientation: the sample, under strong field, is almost homeotropically aligned and hence it should behave like an isotropic material. As expected, the light intensity decreases very fast in correspondence with the rapid molecular dielectric reorientation, i.e., the main peak visible in the current curve. Apparently, the light signal seems to reach its final lower value in correspondence with the dielectric reorientation, but, in fact, it does not fully vanish. As shown in Fig. 2, where the optical response is strongly magnified, it is actually very weak, but it remains practically constant for about

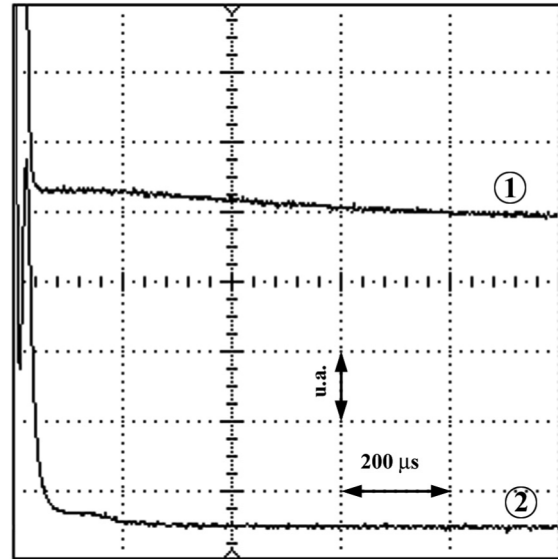


FIG. 2. Typical dynamical response of a thin cell very close to the nematic-isotropic transition temperature, subject to a rectangular electric pulse of 1 ms duration and of amplitude above the threshold for the *H-V* transition. The upper curve 1 is the light intensity transmitted by the sample and the bottom curve 2 is the electric current flowing across the sample. The acquisition starts when the electric field is switched on. The electric pulse duration $\tau_p = 1$ ms is long enough to allow the full evolution of all transient dynamical phenomena.

250 μs and then it decreases to its minimum value. The transient residual birefringence is due to the biaxial wall and only after its breaking the sample birefringence vanishes. The optical observations only allow a rough estimation of the wall disappearing time $\tau \sim 100 \mu\text{s}$ [7], but the current signal also shows that something happens after the dielectric reorientation. The dielectric peak is in fact followed by other weak electric structures for about 150 μs , superposed on the electric current exponential decay. These features also seem to be related to the wall breaking. Moreover the time resolution for the electric signal appears higher than for the optical response. Therefore we perform further electric current observations at lower temperature, where the electric response is stronger and hence they can be better resolved.

At $T = 25$ °C, we apply a rectangular electric pulse ($E = 8.9 \times 10^6$ V/m, $\tau_p = 1$ ms) above the threshold $E_{\text{th}} \approx 6.9 \times 10^6$ V/m for the *H-V* transition [9].

Figure 3 shows now only the current response of the sample. The first dielectric peak is well represented in the main frame, while the following weak electric structures are also 10 times magnified in the box. It is evident that the first molecular reorientation effect is followed by other two phenomena, whose signatures are the two peaks visible in the smaller box. These two peaks are related to the other two electric phenomena that can take place in the cell in the presence of the external electric field: the

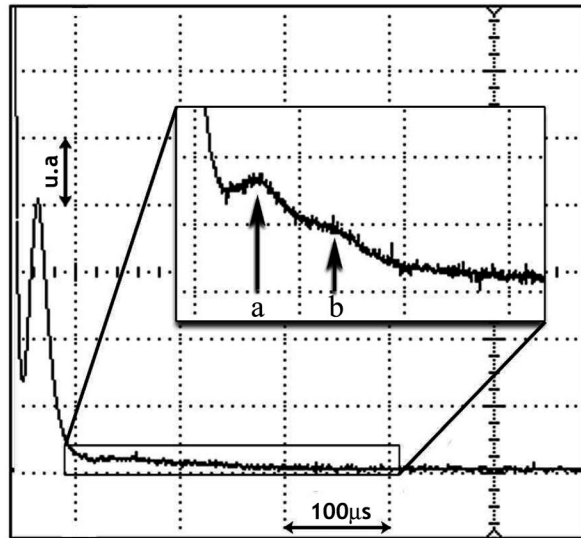


FIG. 3. Electric current response of a $2.40 \mu\text{m}$ thick cell at $T = 25^\circ\text{C}$, submitted to a rectangular electric pulse of 1 ms duration and of amplitude $E = 8.9 \times 10^6 \text{ V/m}$, above the threshold $E_{\text{th}} \approx 6.9 \times 10^6 \text{ V/m}$ for the H - V transition. The large peak due to the dielectric molecular reorientation is visible in the main frame. The two peaks due to the nematic order reconstruction a and to the ion current b are well resolved in the smaller box, where the signal is 10 times magnified.

ion current effect and the induced order reconstruction of the thin biaxial wall present in the transient I state.

The temporal resolution of the current signal is actually much higher than the correspondent response of the birefringence, but it is fundamental to distinguish the ion and the order reconstruction contributions.

For a good time resolved analysis of these phenomena, one needs a wide response of the electric current dynamics. For the order reconstruction, at fixed pulse duration, the useful range for the applied electric field E has a lower limit corresponding to the transition threshold $E_{\text{th}} \approx V_{\text{th}}/d$ and an upper limit due to the fact that, if the order reconstruction is too fast, its current response is hidden inside the dielectric peak. The larger range is obtained at lower temperature and this is another reason to perform our measurements at $T = 25^\circ\text{C}$, quite far from the nematic-isotropic transition for 5CB.

To distinguish the ion and the order reconstruction contributions the raw current signals have been fitted by using the sum of an exponential decay and multiple Lorentzians: we extract the peaks temporal delays measured with respect to the starting time $\tau = 0$ when the electric field is switched on. The current peaks observations are performed during the electric pulse application as its duration $\tau_p = 1 \text{ ms}$ is long enough to allow the full evolution of all transient phenomena.

Two sets of temporal peaks positions are clearly distinguishable in Fig. 4. One of them exists below and above the critical threshold E_{th} of the nematic order reconstruc-

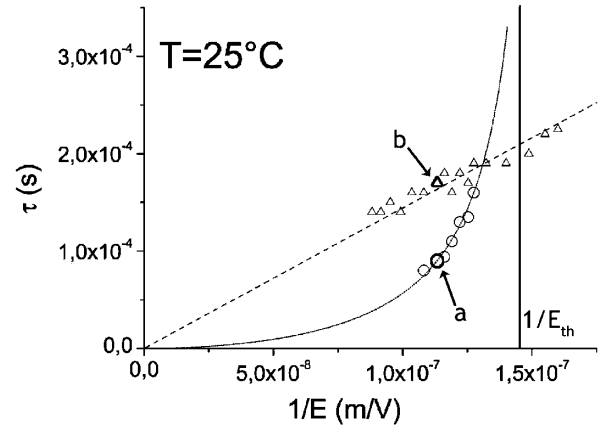


FIG. 4. Plot of the temporal peaks positions in the electric signal after the dielectric reorientation. The linear family is related to the ion current (triangular points); the other set is connected to the nematic order reconstruction (circular points). The highlighted points correspond to the two peaks a and b in the small box of Fig. 3. The continuous lines are best fits of the experimental data.

tion: it shows linear behavior as function of $1/E$. Moreover, this series is well fitted by a straight line which crosses the origin of the $(\tau, 1/E)$ plot, hence one can recognize the expected behavior of the ion current in the cell. Evaluating the slope from the linear fit, we can estimate the ion mobility, $\mu \sim 1.6 \times 10^{-5} \text{ cm}^2/\text{V s}$, comparable with other data found in literature [11].

The second peaks family shows a completely different behavior. It appears only if the electric field exceeds the threshold E_{th} and its dependence on $1/E$ is not linear. Hence this second faster series must be the signature of the nematic order reconstruction.

Coming back to Fig. 3, we are now sure that the peak a is related to the wall breaking and the peak b is due to the induced ion electric current. The two correspondent experimental points are also highlighted in Fig. 4.

To phenomenally describe the electric induced nematic order reconstruction, we consider that the driving force is the dielectric one $\propto E^2$ less the threshold value $\propto E_{\text{th}}^2$ and the nematic order viscously follows this force. Hence, the typical delay time τ will evolve as

$$\frac{1}{\tau} = \frac{1}{\tau_{\text{th}}} \left(\frac{E^2 - E_{\text{th}}^2}{E_{\text{th}}^2} \right), \quad (1)$$

where τ_{th} is the transition characteristic time. The fit of the second peak series of Fig. 4 has been evaluated by using the Eq. (1). We have obtained $\tau_{\text{th}} = (80 \pm 20) \mu\text{s}$. τ_{th} represents the characteristic time that the nematic phase employs to leave the thermodynamical equilibrium at $T = 25^\circ\text{C}$ under the action of the applied electric field. In principle, the system is also characterized by the intrinsic time τ_s , required to rebuild the new nematic order in the vertical direction. This time has been evaluated by

Lelidis and Durand and it is actually very short, $\tau_s \sim 0.1 \mu\text{s}$ [12]. Therefore it can be neglected in this first analysis.

Up to now we have described only the very fast transition from the initial H state to the electrically induced V state, via the intermediate texture I . When the electric field is removed, the nematic texture V is not a stable configuration. It continues to evolve on a longer time scale towards a π -twisted configuration with the same topology. This slower spontaneous viscoelastic relaxation appears because common nematic materials are characterized by a twist elastic constant K_2 lower than the bend elastic constant K_3 [1]. Similar effects have been observed since about 20 years ago, unfortunately without a convincing explanation [13].

To conclude, we have experimentally investigated the tensorial order parameter dynamics in the nematic phase of 5CB by studying the electric induced nematic order reconstruction in a slightly splayed thin cell. For the first time, we have obtained direct data on the characteristic time for the exchanging of two eigenvalues of the nematic order tensor \bar{Q} . This exchange allows the transformation of the quasiplanar H state to the V configuration, as represented in Fig. 1, without any director rotation. Thanks also to the viscoelastic properties of 5CB, in practice, we can transform the quasiplanar texture H in the π -twisted state T , with the same topology of V , by realizing the control of an electro-optical effect that could be exploited for practical devices. Moreover, the starting texture H and the final configuration T are separated by a topological barrier which avoids, in absence of defects, spontaneous relaxations of the system.

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