Dynamics and uniformity of reorientation in liquid crystal cells with PVK alignment layers

Andriy Dyadyusha[1], Malgosia Kaczmarek[2], Sergei Slussarenko[3]

[1]School of Physics and Astronomy, University of Southampton, Southampton SO17 1BJ, UK, E-mail: Andriy@phys.soton.ac.uk, tel: +44 23 8059 3039, fax: +44 23 80593910

[1]School of Physics and Astronomy, University of Southampton, Southampton SO17 1BJ, UK, E-mail: mfk@soton.ac.uk, tel: +44 23 8059 3919, fax: +44 23 80593910

[3]Institute of Physics, National Academy of Sciences, prospect Nauki 46 Kiev, 03028 Ukraine E-mail: serge@iop.kiev.ua

Abstract

We present experimental results on dynamics of Freedericksz transition in liquid crystal cells with alignment layers made of poly(N-vinyl carbazole). DC electric field causes the build-up of, unusually strong, surface charge layers on a liquid crystal-polymer interface that screen liquid crystals from the external field. The process of surface charge build-up and discharge in illuminated areas takes place due to low dark conductivity and high photoconductivity of poly(N-vinyl carbazole), rather than due to its photorefractive nature. If the illuminating beam has a Gaussian profile, reorientation starts from a very small volume, with a clear disclination line in the reorientation spot.

I. Introduction

Cells with different combinations of liquid crystals, photoconductive polymers and sensitisers have been investigated extensively and published in the number of papers [1-9]. One of the most studied geometries is a nematic liquid crystal layer sandwiched between two photoconductive polymer layers, such as poly(N-vinyl carbazole) (PVK) [2]. In this system an active medium, where the change of refractive index takes place (liquid crystal) and the photoactive region (photoconductor) are separated. Two-beam coupling process observed in these structures was explained by the process of reorientation of the bulk liquid crystal due to photorefractive space charge field [4].

While surface effects have primary importance in structures with a polymer-liquid crystal boundary [5, 7], some of their important properties and features have not been investigated in detail yet. In this work we present characteristic features of how the processes on PVK – liquid crystal interface influence the reorientation of liquid crystal molecules. In particular we consider a model explaining the DC field driven Freedericksz transition in systems with poly(N-vinyl carbazole) (PVK) alignment layers, The model is based on the process of build-up of surface charge layers, due to application of DC electric field, and their subsequent discharge with light illumination.

II. Experiment

2.1 Materials and samples

Sample cells had ITO covered glass substrates with different polymer alignment layers such as: undoped PVK, PVK doped with different concentrations of C_{60} and polyimide (PI). Solutions of polymers were spincoated onto clean ITO glass, prebaked and unidirectionaly rubbed with velour cloth to achieve planar homogeneous liquid crystal alignment. The thickness of the layers was of the order 0.1 μm for PVK and below 0.1 μm for PI. The direction of easy axis on a unidirectionally rubbed PVK was found to be orthogonal to rubbing direction with zero pretilt angle.

Our cells were either symmetric – with identical alignment layers or combined - with different combinations of alignment layers on the opposite substrates. We made both planar cells, with easy axis directions on substrates being parallel and twisted cells, with 90^{o} degrees between easy axis directions on substrates. Cells were assembled using 30μm thick Mylar film spacers and filled with undoped E7 liquid crystal.

2.2 Physical properties of PVK.

Poly(vinyl carbazole) chemical structure is presented on figure 1.

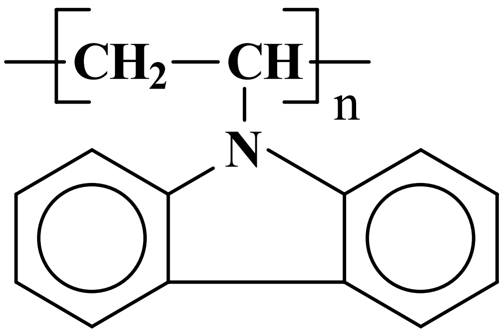

Figure 1. Poly(N-vinyl carbazole) chemical structure.

PVK is a transparent thermoplastic material, with good thermal and chemical stability [10]. Pure PVK is a good insulator in the dark and under visible light illumination. It becomes photoconductive upon exposure to ultraviolet light. PVK has proved particularly efficient as photoconductive material being a charge-transporting polymer with good hole conductivity and high concentration of active charge transport sites (carbazole groups) [11]. Doping with trinitrofluorene (TNF) or C_{60} shifts the absorption of PVK into the visible band via the formation of a charge transfer states [2].

2.3 Geometry of experiment

The schematic diagram of our experimental set up is shown at figure 2.

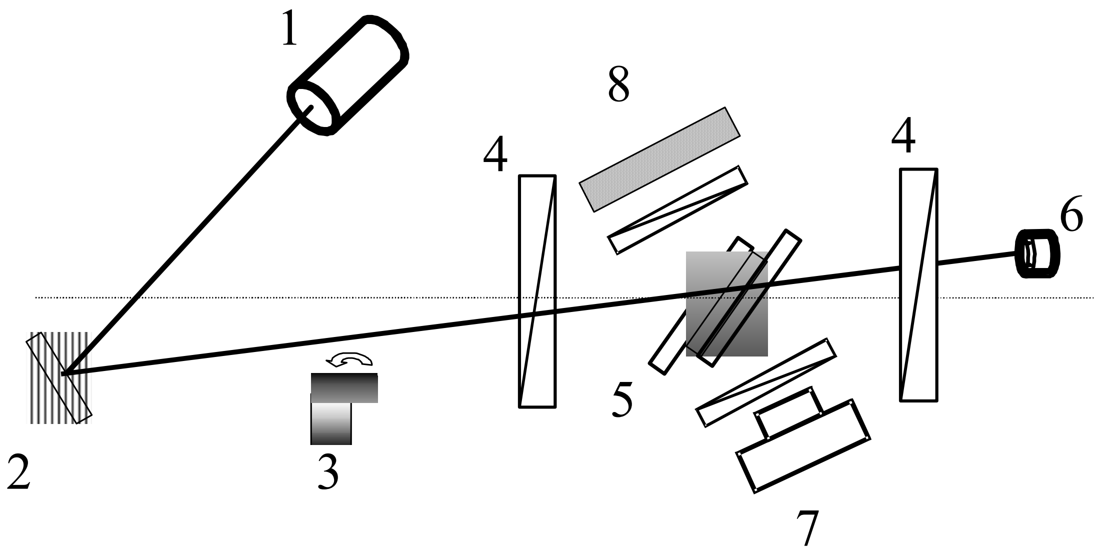

Figure 2. Experimental setup: 1 - laser, 2 – mirror, 3 – shutter, 4 – polarizer, 5 – liquid crystalcell, 6 – photodiode, 7 – camera, 8 – backlight

He-Ne lasers emitting light at either 543 nm or 632.8 nm were used as light sources. A single laser beam, with intensity in the range of 10μW – 2mW, was collimated and had a diameter of 2mm. The incident angle was varied from 0 to 90^o (towards the plane of the substrate). The transmittance of the laser beam, passing through a liquid crystal cell placed between polarisers, was monitored on a photodiode. Laser beam was turned on/off with a shutter connected to an electromagnet relay. DC and AC electric fields were applied from a programmable power supply and a waveform generator. Sample was connected to power supply through a computer controlled electromagnetic relay allowing us to switch instantly ON and OFF electric field as well as to close the sample's circuit. The structure and appearance of an illuminated spot on a sample was simultaneously monitored through second pair of polarisers and backlight and then recorded on a CCD camera. Monitoring of the illuminated area of the cell enabled us to record the dynamics and uniformity of light and electric field driven reorientation.

III. Results and discussion

3.1 Dynamics of Freedericksz transition

Freedericksz transition depends not only on the type of electric field applied (AC or DC), but also on the type of alignment layers. For most polymers, for example PI, there is some, but not significant difference between AC and DC field driven reorientation. Namely, DC Freedericksz transition threshold is usually higher than that with AC field. This is due to the build-up of surface charge layers on liquid crystal – alignment layer interfaces.

In case of PVK alignment layers these dependencies are drastically and qualitatively different, mainly due to high potential of surface charge layers. We observed that AC field driven transition was similar in all samples, irrespective of their structure. Namely, the reorientation was uniform across the illuminated areas of the samples and did not depend on the presence of photosensitizing dopants in PVK, light intensity (in the limit of weak illumination as used in our experiments) or the number (one or two) PVK layers. A typical example of AC field induced reorientation is shown in figure 3.

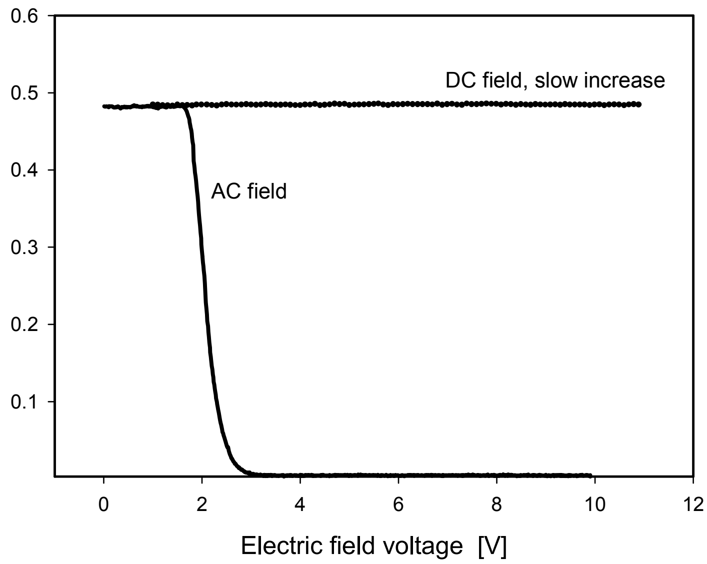

Figure 3. **Normalised intensity of light transmissted through a cell with increasing magnitude of electric field. The solid line presents the case of AC field and the dotted line of slowly-increasing DC field. AC field frequency = 1kHz**

Freedericksz transition is observed with a threshold of about 2V of AC field. We measured the dynamics of AC field driven ON-OFF transition (figure 4 a).

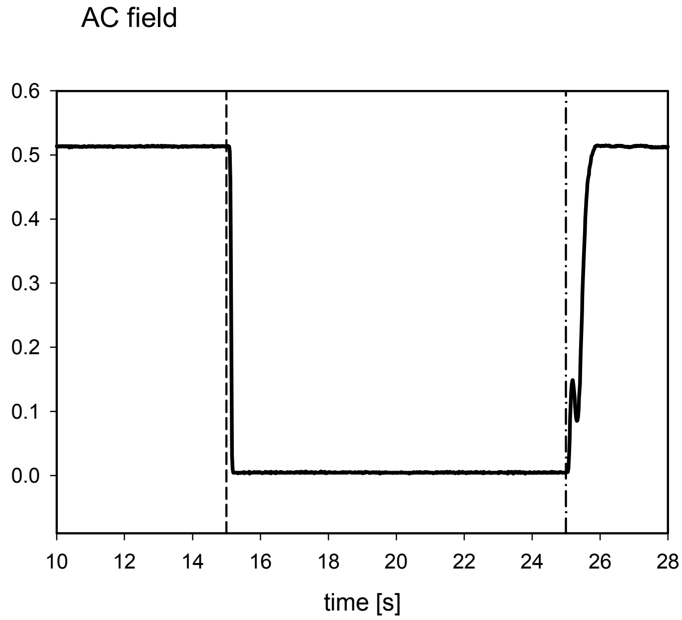

Figure 4 a Dynamics of AC field induced transmission. AC field = 10 V. The field was switched on at t_1=15 seconds (dashed line) and switched off at t_2=25 seconds (dash-dot line).

In the OFF state the circuit between sample cell electrodes is closed and the AC field is switched off. In the ON state cell electrodes are connected to power supply and field is applied. Following the initial change in the molecules alignment, as long as the AC field was applied, the transmission remained constant in time.

DC field driven Freedericksz transition in liquid crystal cells with PVK alignment layers is strongly depended on the experimental geometry, light intensity and the presence of sensitisers. Furthermore, its dynamics was completely different from a typical transient, DC field induced transition, observed in cells with most conventional alignment layers, including PI. This effect was particularly pronounced in cells with pure PVK layers and our experimental studies we concentrated in these types of structures. We tested, in detail, AC and DC field induced transition in twisted nematic cells with rubbed polyimide on one substrate and pure PVK on the other.

If DC field was increased slowly, either continuously or in small steps, no transition was observed up to 56 V. The dashed line shown in figure 3 demonstrates how transmission remained unaffected by increased DC field.

When DC was switched instantly, to a certain non-zero value, a strong transient transition was observed (figure 4 b).

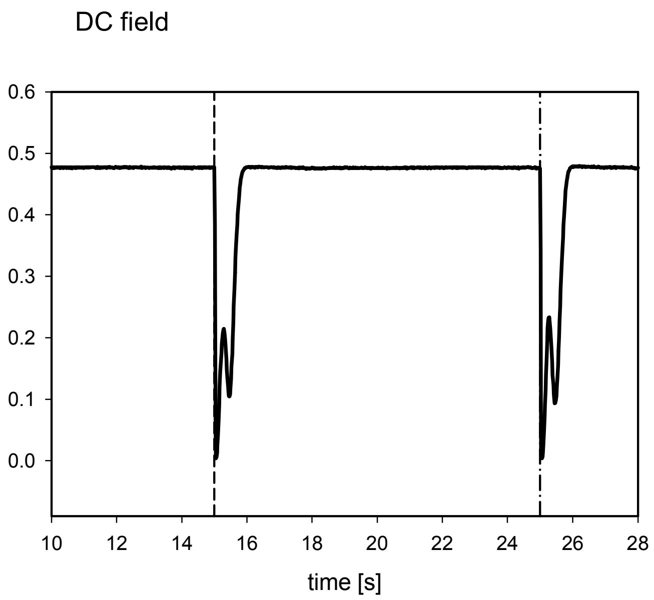

Figure 4 b Dynamics of DC field induced reorientation. DC field = 20 V. The field was switched on at t_1=15 seconds (dashed line) and switched off at t_2=25 seconds (dash-dot line).

In the case of our cells, this value had to be above 10 V. Following a transient change, transmission returned to its original value, namely to the level before application of DC field.

We explored then two ways in which the DC field could be switched off. Second transient transition was observed when DC was switched off (figure 4 b) by disconnecting the electrodes from a power supply and instantly connecting them together. In this way the charge present on ITO layers was discharged. The remaining, uncompensated electric field was equal to surface charge field accumulated on PVK-liquid crystal interfaces.

In other case, after switching off the DC field, the cell electrodes were not connected to one another (after disconnecting from power supply). The charge on ITO layers remained and continued to be compensated by the surface charge on PVK. As a result, the second transient (OFF) transition was not observed.

The transient reorientation was approximately uniform across the sample and viewing it though the polarisers on a CCD camera, it resembled a flash. The transient DC field induced transition was, in fact, the same in both types of cells we investigated, namely in symmetrical and combined cells.

We can explain the mechanism of DC field and light induced transition by the accumulation of charges on the PVK-liquid crystal interface and the screening of the liquid crystal bulk from the applied, external DC electric field. The buildup of surface charge was not very fast, limited probably by low conductivity of liquid crystal. Characteristic time of this process is of the order of seconds. Value of the electric field applied directly to the liquid crystal layer was equal to the difference between external DC field and surface charge field. In case of slowly increasing DC field, an instant difference between the values of external DC and surface charge fields can be smaller than the Freedericksz transition threshold and hence no reorientation of liquid crystal director takes place. However, if an external electric field is applied rapidly, the build-up of surface charge field cannot follow this instantaneous increase, unless the field applied is low. In our case, transient transition was not observed up to 10 V. For such small DC fields, irrespective of the method of its application, the surface charge field builds-up fast enough to screen the external field and there is no change in transmission on application of the DC field.

For stronger DC fields (above 10V), applied instantly, surface charge field takes longer to develop and unscreened external electric field induce liquid crystal bulk reorientation. As time progresses, surface charge screening layer grows stronger and eventually screens the external field, causing liquid crystal director to return to the original distribution as before the field was applied. The transmission shown on figure 4b confirms this change in the orientation of liquid crystal molecules as surface charge layers develop.

Such dynamics of Freedericksz transition only applies if the wavelength of illuminating beam is far from absorption band of PVK and so the conductivity of PVK layer does not change with illumination.

3.2 The effect of PVK photoconductivity

When the sample is illuminated, the surface charge layer accumulated due to the DC field presence can be partially or completely discharged, leading to penetration of external DC field into the LC bulk. This can be achieved by either illuminating cells with ultraviolet light (cells with pure PVK) or by illuminating with visible light (cells with doped PVK). Doping PVK layer with C_{60}, makes the system extremely photosensitive and photoconductive under visible light. In this case Freedericksz transition is sensitive even to ambient light. It is, therefore, important to control ambient light or backlight intensity. The incident light beam is not only testing the changes in reorientation, but also inducing them. While the reorientation can be initiated with either UV or visible light, it is more convenient to use visible light.

In cells with PVK doped with C_{60}, the DC transition threshold depended on light intensity of the testing beam, namely it decreased with increasing incident intensity. In our experiment, the incident beam had Gaussian intensity distribution, which meant that the transition started in the center of the beam, radially extending towards the edges of the spot. When the DC field increased, the size of reoriented spot increased as well. As a result, the transmittance through the cell (or birefringence) measured on a photodiode was not only an average over the thickness of the sample, but also over the diameter of the reoriented spot. Moreover, the distribution of the liquid crystal director reorientation in a cell was three-dimensional. Due to light induced reorientation, there is dependence of director distribution on all x,y,z coordinates, not only on z, like in a typical one-dimensional Freedericksz transition.

The Freedericksz transition threshold is an exponential function of light intensity and is only limited by the saturation of PVK photoconductivity or complete discharge of surface charge in bright areas, depending on which effect takes place first.

Our model of surface charge buildup in a liquid crystal cell with PVK:C_{60} can be understood better via a comparison with the xerography process. In xerography, a photoconductor layer, typically 10 - 50 µm thick (for example doped PVK) is deposited onto conductive, grounded substrate [12]. In the first step, a photoconductor is charged by corona discharge up to 1000 V. and this charging occurs in the dark. PVK or PVK:C_{60} is capable of maintaining this surface charge for considerable time, because its dark conductivity is low. In the next step, the photoconductor is exposed to an image. In bright areas surface charge is discharged to the grounded substrate via induced photoconductivity. Finally, the photoconductor with surface charge distribution corresponding to the initial image is used to attract toner particles and transfer the image onto paper.

In our liquid crystal cell a PVK layer (0.1 μm thick) is also deposited onto conductive (ITO covered) substrate. What is different to xerography process is that instead of air there is a liquid crystal layer and another ITO covered substrate. Moreover, instead of a corona device, an external DC field is applied to ITO electrodes. In our experiments we applied up to 56 V – the limit set by our power supply. PVK surface was charged up to the value of external potential, if we neglected a potential drop on the other elements of the cell. We want to stress that in our model, and according to our results, surface charge build up is only due to external DC field. It builds-up efficiently in the dark, so this process is completely different from previous reports where surface charge was attributed to photorefractive effect [2, 4].

For surface charge to develop, the high resistivity (or low dark conductivity) of PVK is an essential parameter. While in xerography, surface is charged before image exposure, in a liquid crystal cell charging occurs all the time when it is connected to external DC field. Discharge also occurs while PVK is illuminated. The result of this simultaneous charge-discharge process is a steady-state balance of surface charge field.

This qualitative model agrees with experimental results. For example, testing in our cells AC field induced Freedericksz transition, we observed that, as expected, charges do not accumulate on the PVK layer and as a result the transition was uniform across the cell.

The essential role of PVK – the accumulation of screening surface charge- can be confirmed by observing DC induced Freedericksz transition in an area where PVK was removed. This can be just a simple scratch on the substrate surface. In the area where PVK is absent, such as at and near the scratch, there should be no significant surface charge, so an external DC field can easily reorient liquid crystal near the scratch. We observed Freedericksz transition in the vicinity of the scratch at low external DC, which confirms our model.

3.5 Symmetry of reorientation and disclination lines.

We observed an interesting effect in a symmetric cell having both PVK:C_{60} photoconductive substrates. Photos of reoriented spot and the proposed director distribution are presented in figure5.

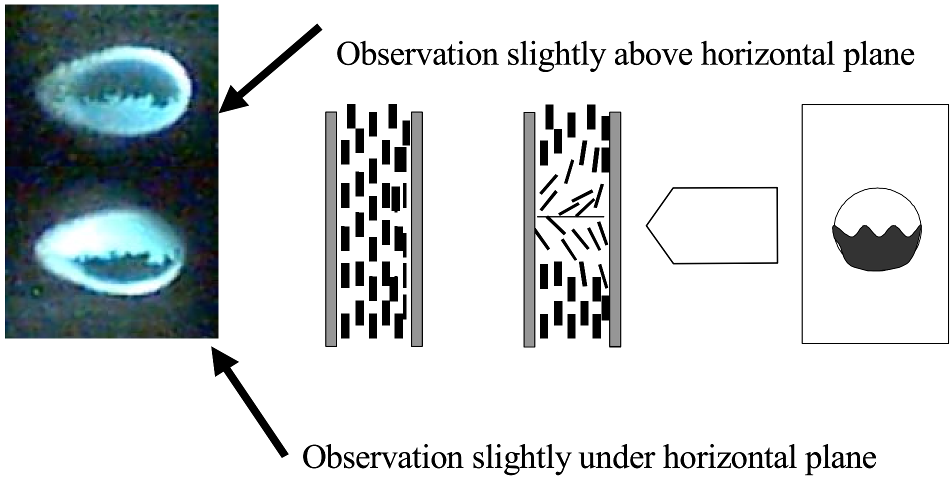

Figure 5 Photographs of the reoriented spot under DC field and laser illumination with schematic diagrams of the proposed director distribution

As described above, the reorientation starts in the centre of the beam and, typically for Freedericksz transition, in the middle of a liquid crystal layer. Due to the symmetry of the system and the equal probability of director fluctuations, the reoriented spot splits into domains with the opposite tilt of the director. As reorientation originates from a very small volume, it is likely that the number of domains will be small, as low as two, for example. Our cell had initial planar alignment and circular (or spherical) shape of reoriented volume of liquid crystal molecules. This small, reoriented volume was surrounded by planar liquid crystal that remained in the initial alignment. Therefore, a stable condition of such system would include a disclination dividing the reoriented area in the middle of the spot and orthogonal to initial director direction. We observed such reorientation pattern, which was easily achieved by increasing light intensity at a fixed external DC field or by increasing DC field at a fixed light intensity. The disclination line was clearly visible, as shown on photographs presented in figure 5. They were taken with viewing directions slightly up and down from normal to the cell placed between crossed polarisers. In this case, DC field was well above the threshold of the Freedericksz transition and liquid crystal was reoriented in the whole layer inside the spot.

The symmetrical change of transmittance is the evidence of the opposite tilt in domains that had a director reoriented closely to homeotropic alignment. Increasing/decreasing of external DC field (or light intensity) leads to corresponding increase/decrease in the diameter of the reoriented spot and the length of the disclination line. This presence of a disclination and its characteristics were well reproducible.

IV. Conclusions

In conclusion, we reported the characteristic features and dynamics of light and electric field induced Freedericksz transition in a liquid crystal cell with alignment layers made of PVK and PVK doped with fullerene (C_{60}) as a photosensitiser. The Freedericksz transition observed in such cells is different than that observed in cells with standard alignment layers, such as polyimide. This is due to high photoconductivity and low dark conductivity of PVK. DC field induced Freedericksz transition depends on light intensity and its dynamics could be explained by the build-up of a surface charge layer and its subsequent discharge with light illumination. The build-up of surface-charge layers is relatively slow so only a steady-state, external DC field can create them. The threshold of Freedericksz transition depended on intensity, the profile of applied electric field and the quality of PVK layer. The disclination line observed inside the reoriented spot was the result of balance and interplay between the initial and induced reorientation of liquid crystal molecules. In general, liquid crystal cells with PVK+C_{60} alignment-command substrates are extremely photosensitive at visible light and their transmission properties can be well controlled with light and electric field.

V. Acknowledgments

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VI. References

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