

P-79: Model of Freedericksz Transition and Hysteresis Effect in Polymer Stabilized Nematic Liquid Crystal Configurations for Display Applications

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Abstract

The critical field of the Freedericksz transition and optical hysteresis is investigated for Polymer Stabilized Nematic Liquid Crystals as a function of polymer concentration. The results are described by a simple phenomenological model, which accounts for the change in polymer network morphology as the concentration of polymer is increased.

1. Introduction

Polymer Stabilized Liquid Crystals (PSLC) are composed of a polymer network dispersed in a nematic liquid crystal. They are promising materials for many display configurations. Small amounts of reactive monomer polymerized in a nematic liquid crystal create an aligned polymer network that under some circumstances can provide a memory effect [1]. The polymer network structure captures the details of the nematic director prior to photo-polymerization. In this paper we present our results on the influence of the polymer network on the switching behavior of a homogeneously aligned nematic liquid crystal. We show that the critical field of the Freedericksz transition, as well as the width of optical hysteresis, increases with the polymer network concentration, and that this dependence on polymer concentration can be described by a phenomenological model.

Previous models described the critical field of the Freedericksz transition in PSLC systems, which to some extent took into account the polymer morphology as collection of planes [2] or bundles [3] parallel to the substrates, or collectively as an effective field [4]. These models nicely describe the behavior of the critical field in the low concentration limit. The model of the polymer matrix as a series of planes perpendicular to the x-, y-, and z-axes of the Cartesian coordinate system was introduced in [5]. Current models do not explain the dependence of the critical field on the polymer concentration especially in the high concentration limit. We explain the behavior of the critical field of the Freedericksz transition in liquid crystal-polymer systems by introducing a phenomenological model, which takes into account the morphology of the polymer network. Also we explain the optical hysteresis effect and its dependence on concentration in such systems.

2. Result

Consider a planar cell of PSLC sandwiched between two transparent indium tin oxide (ITO) coated glass plates with a $5\mu\text{m}$ cell gap thickness. The glass substrates are coated with rubbed polyimide such that the nematic directors on both surfaces are anchored parallel to the plates. Two types of PSLC

were prepared from low molecular weight nematic liquid crystal BL038 (EM Industries), diacrylate monomers LC242 (BASF) for the first type and RM257 (EM Industries) for the second type, and a low percentage of photo-initiator Darocur1173 (Ciba) was used so that polymerization can be carried out in the UV. When a diacrylate monomer is polymerized in a liquid crystal solvent, the orientation and order of the resultant network depends on the orientation and order of the liquid crystal. So we assume that the polymer network has planar alignment, which is identical to the alignment of liquid crystal. In our studies we investigated the concentration range where the monomer is totally dissolved in liquid crystal at room temperatures ($<6\%$ of monomer by weight). We also make sure that the polymerization is complete by UV curing the samples for sufficiently long time.

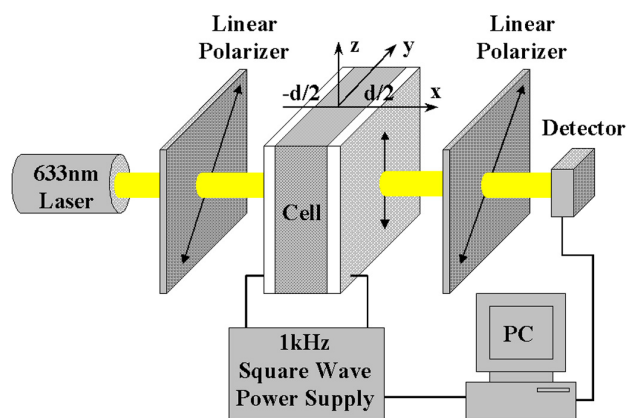


Figure 1 The experimental set up for measuring the transmission properties of a PSLC cell. The cell is situated between the cross polarizers oriented at 45° with respect to the nematic director inside the cell.

A schematic illustration of the experimental set up for measuring the transmission properties of a PSLC cell is presented in Figure 1. We consider the arrangement of a cell between a pair of parallel polarizers. The nematic director in the cell is oriented at 45° relative to the transmission axes of the polarizers. ITO glass plates serve as the electrodes to apply 1kHz square ac-voltage V . In the field-off state, the cell exhibits a phase retardation $\Gamma (=2\pi/\lambda(n_e - n_o)d = 4\pi)$, where $n_e (=1.799)$, $n_o (=1.527)$ are the extra-ordinary and ordinary refractive indices of the BL038 respectively, $d (=5\mu\text{m})$ is the cell gap thickness, and $\lambda (=633\text{nm})$ is the wavelength of the incident laser light. Due to the uniform alignment of the liquid crystal and the polymer network, the transmission light intensity of the cell $I (=1/2\cos^2(\Gamma/2))$ is maximum in the field-off state with such arrangement of polarizers.

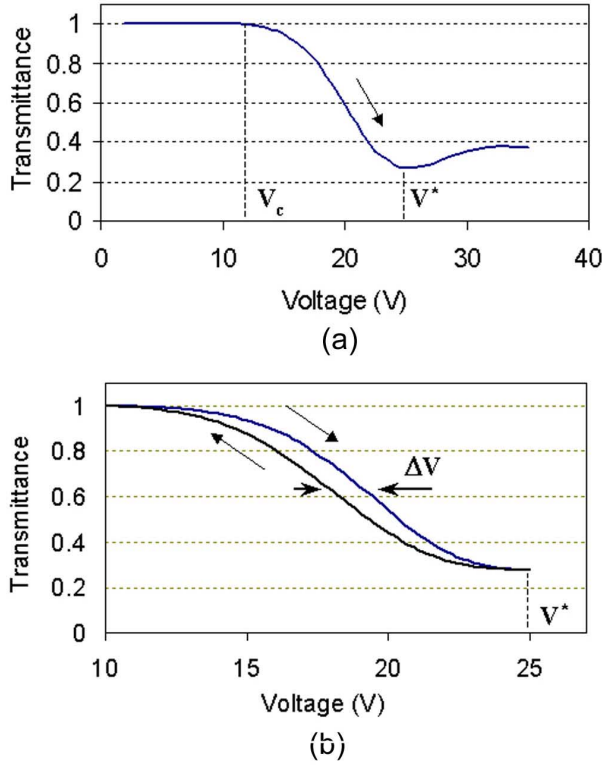


Figure 2 Scans of transmittance versus applied voltage V . The arrows point in the direction of increasing or decreasing field. V_c is the critical voltage, V^* is the maximum applied voltage for optical hysteresis experiment, which corresponds to the first minimum in the transmission intensity, and ΔV is the width of optical hysteresis measured at the median value of the transmittance.

A transparent cell can be switched to an opaque state when an electric field is applied. This is due to the creation of the domains of the nematic director in the cell [6]. By application of the field only a part of the molecules become reoriented. Those, which are in the close vicinity to the polymer network, remain less influenced due to anchoring. In this way domains with different orientations of the nematic director can be created. The phase retardation of the reoriented molecules becomes $\Gamma = 2\pi/\lambda \int (n_e(\theta) - n_o) dx$, where the integral is over the domain size and $n_e(\theta)$ is given by:

$$\left(\frac{1}{n_e(\theta)} \right)^2 = \left(\frac{\sin \theta}{n_o} \right)^2 + \left(\frac{\cos \theta}{n_e} \right)^2 \quad (1)$$

The angle $\theta = \theta(x, y)$ is the tilt angle of the director with respect to the z -axis, which depends on the position x, y of the molecule in the domain and the field strength. The transmittance $T (= I(V)/I(V=0))$ of the cell in the field-on state is affected by two factors, which are the scattering by the nematic director domains and the change in the phase retardation. The change in the phase retardation gives rise to oscillations in the transmittance, which we employ in the optical hysteresis experiment. For this purpose we continue the light transmission intensity versus voltage scans

until the maximum voltage V^* , where the light transmission intensity reaches its minimum for the first time (Figure 2(a)), to insure that for each concentrations of the polymer in PSLC cells the phase retardation is the same at the maximum applied voltage. For each concentration voltage was increased and decreased at the same rate. By monitoring the transmittance of the cells as a function of the applied voltage we determine the critical voltage V_c of the Fredericksz transition, and hence the critical field $E_c = E_c(c)$ for each concentration c of the polymer. Secondly, for each concentration we measure the width of the optical hysteresis loop, which we define as the difference in voltage $\Delta V (= V_{up} - V_{down})$ between V_{up} , when the voltage is increased, and V_{down} , when the voltage is decreased, at the median value of the transmittance (Figure 2(b)).

The results of the critical field of the Fredericksz transition for both PSLC are shown in Figure 3, where we normalized our data to the critical field $E_0 (= \pi/d \sqrt{K/(\epsilon_a \epsilon_0)}) = 0.24 \text{ V}/\mu\text{m}$ for BL038 in planar geometry without the polymer network ($c=0$), where $\epsilon_a (= \epsilon_{||} \epsilon_{\perp} = 16.4)$ is the dielectric anisotropy of the BL038, ϵ_0 is the permittivity of the free space, and $K=20.7 \text{ pN}$ is the elastic constant for BL038 in one constant approximation.

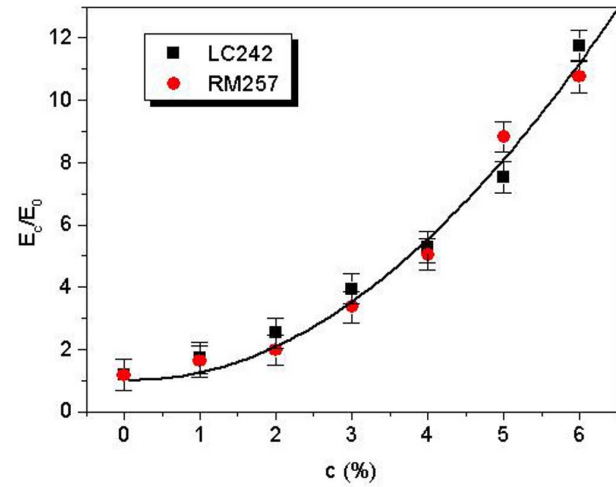


Figure 3 Critical fields versus concentration for two PSLC prepared using BL038 and diacrylate monomers LC242 (BASF), and RM257 (EM Industries). E_c is the critical field normalized to the critical field E_0 in the planar cell filled with pure BL038. Solid curve represents the fitting based on the model.

To explain the critical field behavior for arbitrary concentration, we assume that the polymer network is composed of polymer fibrils oriented parallel to the nematic director of the field-off state, which depending on the concentration may or may not be cross-linked. At low concentration the fibrils are not cross-linked (Figure 4(a)), although at higher concentration, the fibril assembly can be thought of as a formation similar to interpenetrated polymer planes [5], due to cross-linking. These polymer planes resemble cylinders with their axes parallel to the director of the nematic liquid crystal in the field-off state (Figure 4(b)). A Scanning Electron Microscope (SEM) study of the polymer networks [3,7] shows excellent agreement with this

concept of polymer morphology. We approximate the polymer network at any polymer concentration as forming tubes of rectangular cross-section and long axes parallel to the nematic director in the field-off state (Figure 4). In the field-on state the size of the rectangles equals to the size of the domains of the nematic director. We discuss the Fredericksz transition in the PSLC cell with the cell gap thickness d in context of the transition in the rectangle with sides of length a and b .

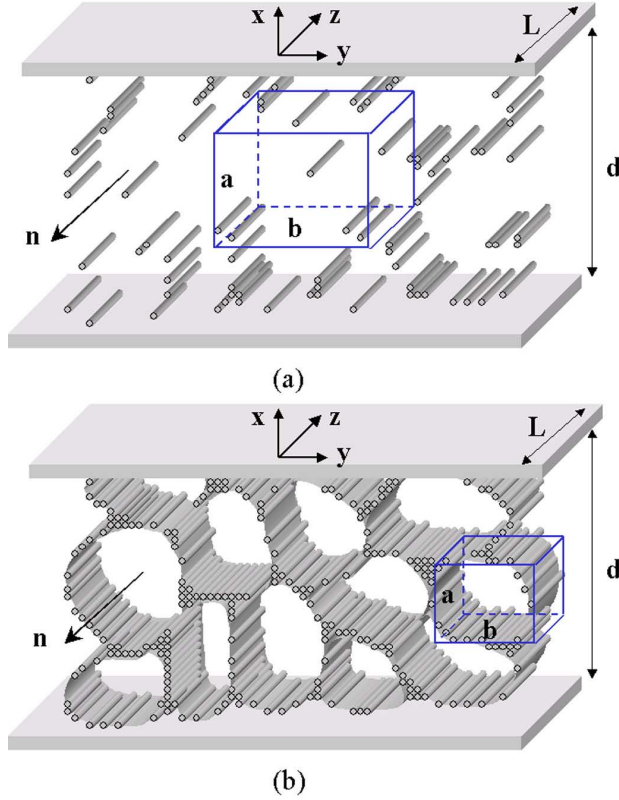


Figure 4 At low concentration the polymer network is shown as a collection of fibrils parallel to the nematic director n in the field-off state (z -axis). At high concentration the cross-linked polymer form hollow tubes. The electric field E was applied along the x -axis perpendicular to the glass substrates. We approximate the cross-section of the cylinders by rectangles as shown.

Based on elastic continuum theory, the total free energy of the nematic liquid crystal can be written as:

$$F = \frac{L}{2} \int (K \left(\frac{\partial \theta}{\partial x} \right)^2 + K \left(\frac{\partial \theta}{\partial y} \right)^2 - \epsilon_a \epsilon_0 E^2 \sin^2 \theta) dx dy \quad (2)$$

where $\theta(x,y)$ is the distortion angle with respect to the z -axis, and L is the lateral size of the cell. By minimizing the total free energy (2), we obtain the differential equation for the distortion angle:

$$\frac{\partial^2 \theta}{\partial x^2} + \frac{\partial^2 \theta}{\partial y^2} + \frac{\epsilon_a \epsilon_0 E^2}{K} \sin \theta \cos \theta = 0 \quad (3)$$

The critical field of Fredericksz transition in a rectangle of sides a and b in the x and y directions respectively is obtained by solving (3) with the strong anchoring boundary conditions: $\theta=0$ at $x=0, a$ and $y=0, b$. We find:

$$E_c = \sqrt{\frac{\pi^2 K}{\epsilon_a \epsilon_0} \left[\left(\frac{1}{a} \right)^2 + \left(\frac{1}{b} \right)^2 \right]} \quad (4)$$

This result can be understood by balancing the torque of the external electric field and the torques induced by distortions in both x and y direction. We assume that the lengths a and b diminish with increasing concentration as: $1/a = 1/d + 1/\xi$ and $1/b = 1/\xi$, where the characteristic length ξ is determined by the polymer network and it depends on the concentration c as $\xi = dA/c$. The parameter A is a material property, related to the number of polymer fibrils per unit volume. With finite surface anchoring, we also need to include the surface anchoring energy. We assume that the surface anchoring energy per unit area takes the Rapini-Papoular form $f_s = 1/2 W \sin^2 \theta$ [8], where W is the surface anchoring strength. The modified boundary conditions at finite anchoring are [8,9]:

$$\begin{aligned} \left(\frac{d\theta}{dx} \right)_s \pm \frac{W}{K} \sin \theta \cos \theta &= 0, \\ \left(\frac{d\theta}{dy} \right)_s \pm \frac{W}{K} \sin \theta \cos \theta &= 0 \end{aligned} \quad (5)$$

where the derivatives are taken on the surfaces of the polymer. Hence the critical field with finite anchoring can be written approximately as:

$$E_c = \sqrt{\frac{\pi^2 K}{\epsilon_a \epsilon_0} \left[\left(\frac{1}{d} + \frac{1}{\xi + 2K/W} \right)^2 + \left(\frac{1}{\xi + 2K/W} \right)^2 \right]} \quad (6)$$

This expression is valid when $K/W < 10a$ and $K/W < 10b$ [10]. The surface anchoring strength W is also a function of the polymer concentration c . We assume that it depends on the concentration as: $W = Kc^2/(dB)$ since the surface density of polymer fibrils scales as c^2 . The parameter B is also a material parameter, which characterizes the polymer-liquid crystal interface. Finally the critical field as a function of the polymer concentration takes the form:

$$E_c = \frac{\pi}{d} \sqrt{\frac{K}{\epsilon_a \epsilon_0} \left[\left(1 + \frac{c^2}{Ac + 2B} \right)^2 + \left(\frac{c^2}{Ac + 2B} \right)^2 \right]} \quad (7)$$

At low concentrations ($c \rightarrow 0$), the critical field in equation (7) gives the original Fredericksz critical field $E_c = E_0 = \pi/d \sqrt{K/(\epsilon_a \epsilon_0)}$. By fitting the experimental data to

the critical field of expression (7) we obtain $A=0.2$ and $B=2.4$. The fit is shown in Figure 3 by the solid line. The value of the parameter A shows that even at concentrations $c \sim 1\%$, the relative polymer spacing is much smaller than the cell gap d , which is in the agreement with the SEM pictures of the PSLC [3,7].

The dependence of the optical hysteresis width on polymer concentration is shown in Figure 5. We believe that the presence of optical hysteresis is due to the extremely high relaxation time of the polymer network, which is due in turn to the elastic strain induced on the polymer network by the electric field. Clear dependence of the optical hysteresis on concentration indicates the dependence of the morphology of the polymer network on concentration. We are currently working on a model to describe this phenomenon.

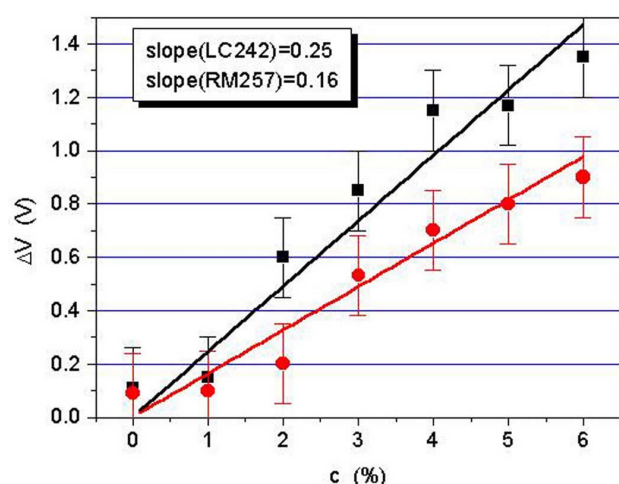


Figure 5 Dependence of the optical hysteresis width on concentration. $\Delta V_{LC242}=0.25c$ and $\Delta V_{RM257}=0.16c$ for PSLC prepared using LC242 and RM257 respectively.

3. Summary

This contribution represents a comprehensive approach to Polymer Stabilized Liquid Crystals, and underlines the details behind their critical field of switching and hysteresis effect. Polymer networks are being investigated for a number display related reasons such as to make Liquid Crystal Display materials more robust for flexible display applications, to create a bistable memory effect, and increase the response time in in-plane switching displays. Our basic model and results are very relevant in all of these current display related orientations.

4. Acknowledgement

This work was supported by the National Science Foundation under grants DMR-9875427 (G. P. C and P. A. K.) and DMR-9873849 (N. V. P. and R. A. P.).

5. References

- [1] Liquid Crystals in Complex Geometries, Edited by G. P. Crawford and S. Zumer, Taylor & Francis 1996
- [2] C.-C. Chang, L.-C. Chien, and R. B. Meyer, Phys. Rev. E **56**(1), 595-599 (1997)
- [3] R.-Q. Ma, D.-K. Yang, Phys. Rev. E **61**(2), 1567-1573 (2000)
- [4] M. J. Escuti, C. C. Bowley, G. P. Crawford, S. Zumer, Appl. Phys. Lett. **75**(21), 3264-3266 (1999)
- [5] R. A. M. Hikmet, H. M. J. Boots, Phys. Rev. E **51**(6), 5824-5831 (1995)
- [6] R. A. M. Hikmet, J. Appl. Phys. **68**(9), 4406-4412 (1990)
- [7] D.-K. Yang, L.-C. Chien and Y. K. Fung in Liquid Crystals in Complex Geometries, Edited by G. P. Crawford and S. Zumer, Chapter 5, 109-114, Taylor & Francis 1996
- [8] A. Rapini, M. J. Papoular, J. Phys. (Paris), Colloq. C4, **30**, C4-54 (1969)
- [9] J. Nehring, A. R. Kmetz, T. J. Scheffer, J. Appl. Phys. **47**, 850 (1976)
- [10] L. M. Blinov, V. G. Chigrinov, Electrooptic Effects in Liquid Crystal Materials, Chapter 3, 111-113, Springer 1993