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Constantin Rosu^a & Doina Manaila-maximean^a

^a University "Politehnica Bucuresti", Department of Physics, Splaiul Independentei 313, Bucharest, 77206, Romania

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Mechanically Induced Electric Polarization in Polymer Dispersed Nematic Liquid Crystal Films

CONSTANTIN ROSU and DOINA MANAILA-MAXIMEAN

*University "Politehnica Bucuresti", Department of Physics,
Splaiul Independentei 313, Bucharest, 77206, Romania*

We obtained polymer dispersed nematic liquid crystal films E7/PMMA. In order to induce anisotropy and consequently electrical polarization, the films have been hot stretched and pressed.

We studied the dependence of the polarization upon temperature, frequency and amplitude of the applied electric field. High values of the order of $\mu\text{C}/\text{cm}^2$, have been obtained. The frequency dependence of the polarization indicates relaxation times of the order of milliseconds.

Keywords: liquid crystals; hysteresis; polymers; mechanical stretching

INTRODUCTION

Polymer dispersed liquid crystal (PDLC) films have composite structure and consist of micrometer-size liquid crystal (LC) droplets embedded in a solid polymer matrix [1]. PDLC films can be self-supported and flexible since the polymer matrix is plasticized by the dissolved LC molecules. They are considered for applications, such as: light shutters, displays, and electrically controlled polarizers [2,3,4].

The unidirectional alignment of the droplets in a PDLC film can be produced by several methods: polymerization of the matrix in the presence of a strong longitudinal electric or magnetic field, shearing deformation, stretching of the film [6]. Stretching is most effective,

providing a high degree of droplet ordering and anisotropy of the polymer matrix. In these devices the linear coupling between the applied electric field and the induced polarization P_i in paraelectric liquid crystal is used.

In our paper we used the stretching and pressing of the PDLC film, in order to induce a high value of the polarization.

EXPERIMENTAL

The polymer dispersed liquid crystal films have been obtained by the solvent induced phase separation technique [6]. We prepared the liquid crystal/polymer composite by mixing the nematic liquid crystal E7 ($t_{NI}=59^{\circ}\text{C}$) and polymethyl methacrylate (PMMA), in equal ratios by weight. The size of the droplets was ranged between $1\div 5\mu\text{m}$.

To create geometrical anisotropy and induce electrical polarization, the films have been stretched and pressed. By stretching the films unidirectional, the radial symmetry of the droplets is destroyed, and they become elongated. In the stretching process, the polymer matrix also becomes anisotropic. The degree of the film extenuation, p , defined as the ratio between the local film length in the deformed and in the undeformed states, was $p=3$. The film was stretched when the liquid crystal was in its isotropic phase - hot stretching. The stretched films have been pressed and stuck between ITO covered glass plates. The thickness of the cell was $30\mu\text{m}$.

The investigation of the ferroelectric properties of liquid crystals can be carried out by the Sawyer-Tower method, which gives a display of the hysteresis loop on an oscilloscope. This method has been used by Japanese researchers [7]. The polarization is determined by measuring the value obtained by extrapolation of the linear part of the loop. Nevertheless, this method is unsuitable for samples exhibiting higher conductivity. As shown by Diamant ET al. [8], a conductive sample exhibits a loop with rounded edges, which makes impossible to accurately determine the correct value of the polarization. The method initiated by Diamant et al. surmounted these difficulties.

Using a specially built apparatus, hysteresis meter [9], we applied on the sample a.c. electrical signals of different frequencies and amplitudes. For compensating the conductivity and the nonferroelectric capacitance of the sample we used the experimental set-up shown in Figure1.

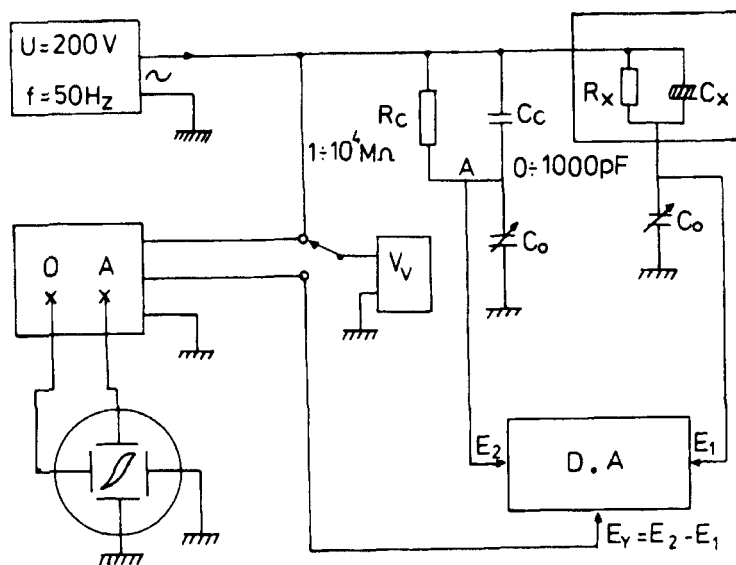


FIGURE 1. Bridge for measurements of ferroelectric hysteresis. OA- output amplifier, DA=differential amplifier, V_v – peak voltmeter. Voltage range 0-200V, $C_0 = 1\text{nF}$, $C_x = 0-1000\text{pF}$, $R_x = 10^4-10^{10}\Omega$.

If U_y is the voltage value needed to reach compensations, then:

$$U_y = k P_s S / C_0 \quad (1)$$

Where P_s is the spontaneous polarization, S is the surface area of the sample, C_0 a fixed capacitance, which should be adjusted in steps in such a way as to stop the saturation of the differential amplifier when the signals E_1 or E_2 are increased, and k is a constant.

The measurements have been performed as follows:

- The electric field was increased until saturation was achieved; if this is done slowly, breakdown can be avoided.
- The free capacitance of the sample should be independent of the applied voltage; otherwise corrections have to be introduced.
- The frequency of the applied voltage was filtered in order to eliminate distortions due to the higher harmonics.

d) In order to follow the temperature dependence of the polarization, the PDLC cell was introduced in an oven, with controlled heating and cooling; the precision of the temperature control was: $\pm 0.01^\circ\text{C}$.

RESULTS AND DISCUSSIONS

Using the described method, we measured the polarization dependence on temperature, applied electric field amplitude and its frequency. For the unstretched PDLC E7/PMMA sample no polarization was observed. For the stretched PDLC E7/PMMA we obtained the following results:

In Figure 2 is represented the dependence of the saturation polarization as a function of temperature. At low temperatures the saturation polarization reaches low values of the order of nC/cm^2 . The polarization increases when increasing the temperature. According to

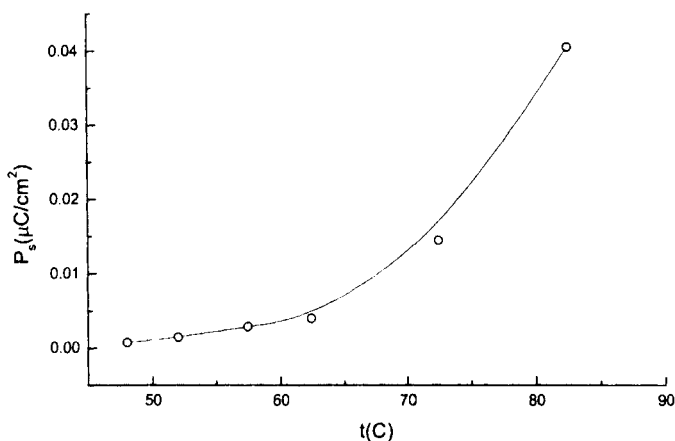


FIGURE 2. Dependence of the saturation polarization on temperature for E7/PMMA stretched sample, at $150\text{ V}_{\text{rms}}$.

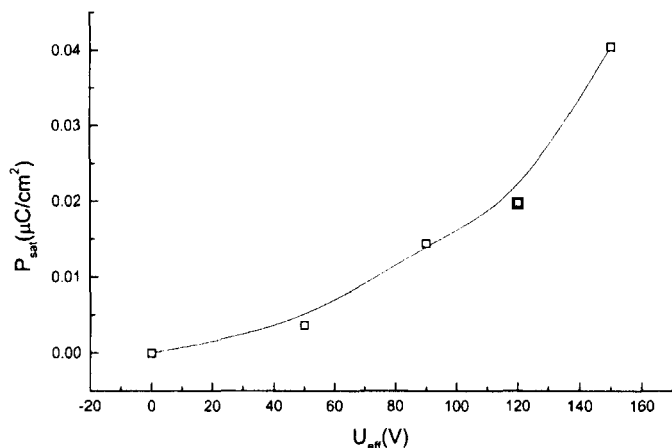


FIGURE 3. Dependence of the saturation polarization as a function of the applied voltage, for E7/PMMA stretched sample, at $f=38$ Hz, $t=82^\circ\text{C}$.

the corresponding liquid crystal studies, we should expect a discontinuity in the temperature dependence of the saturation polarization when the temperature reaches the isotropic phase transition temperature. Because no such discontinuity is noticed, we consider the predominant role of the stretched PMMA in the generation of the polarization. This proves the mechanically induced anisotropy in the PMMA. The continuous temperature increase of the saturation polarization after the isotropic phase transition temperature is due to the great space charge existent in the PMMA [10]; this charge has a maximum at 402K and finally tends to vanish for higher temperatures.

In Figure 3 is shown the dependence of the saturation polarization on the amplitude of the applied voltage. We limited the maximum value of the applied voltage at $150V_{\text{rms}}$ to avoid the heating effects of the sample, which result in hysteresis loop distortions. The saturation polarization versus applied voltage has a parabolic shape. The saturation polarization is absent at $U_{\text{eff}} = 0$, which indicates the absence of a permanent polarization in the sample. In normal ferroelectrics, the dependence $P_{\text{sat}}=f(U_{\text{eff}})$ reaches saturation because the orientation phenomena reach maximum distortion. In our case this

saturation is absent, but we can presume that a short (for avoiding the thermal effects) and high voltage pulse would produce saturation. The frequency dependence of the saturation polarization is shown in Figure 4 for different temperatures. An exponential decrease of the saturation polarization versus frequency is observed. The “cut-off” frequency at which the polarization decreases $e=2.71$ times is approx. 120Hz at temperature $t=82.5^{\circ}\text{C}$, and decreases with the decrease of the temperature. This dependence can be explained by the correlation between the mechanically inertia and the viscosity of the dipoles from the sample. These values for the “cut-off” frequency can be correlated with the time constant of the order of milliseconds, measured in the optical switching studies.

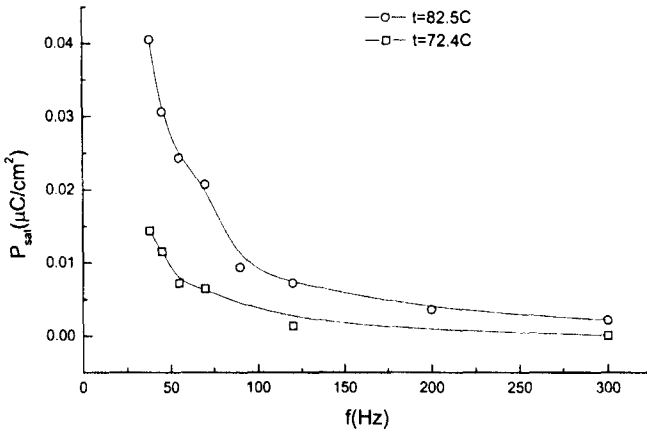
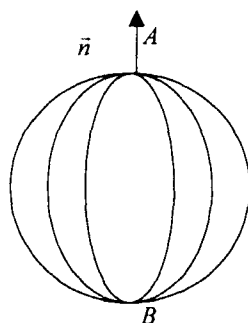


FIGURA 4. Saturation polarization versus frequency for E7/PMMA stretched sample, at 150 V_{rms} .

To understand the polarization induced in the stretched PDLC films, we discuss the changes in the film morphology. As it is known, the nematic phase possesses a long range orientational order, the long axis of the rod-like molecules being aligned along the preferred direction, which can be described by the director \mathbf{n} . The two directions \mathbf{n} and $-\mathbf{n}$ being

FIGURE 5. Droplet with bipolar tangential structure.



equal in the liquid crystals. The preferential direction of the molecules' long axis is modified by the external physical factors (electrical, magnetic or thermal fields). Thus the microstructure studies [11] have shown that in the most widespread configuration the nematic liquid crystal directors in the droplets has a bipolar tangential structure with (Fig.5) with two surface point defects on opposite sides of the droplets (points A and B). Supposing that the droplet directors are confined to two dimensions, the order parameter is defined by the average:

$$S = \langle 2 \cos^2 \theta - 1 \rangle \quad (2)$$

where θ is the angle between the global director \vec{N} of the sample and the Ox axis, chosen in the direction of the stretching (see Figure 6b). The global director \vec{N} is defined as an average on the directors of the droplets.

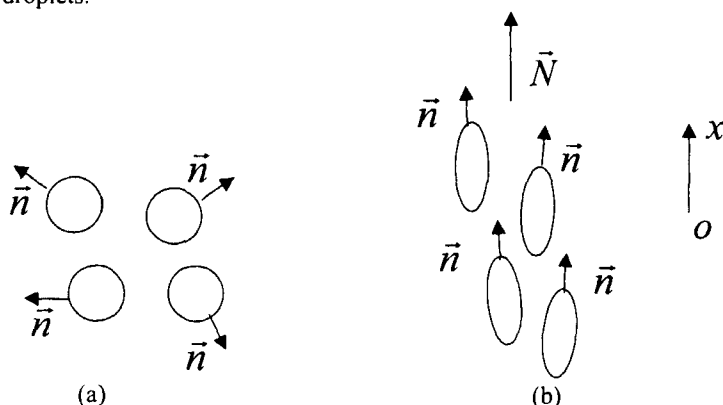


FIGURE 6. Schematic representation of the orientation of the bipolar nematic droplet in the unstretched (a) and in the stretched PDLC films (b).

In the unstressed PDLC matrix, the directors of the sample are randomly oriented (see Figure 5) and the order parameter is zero ($\cos^2\theta=1/2$ and $S=0$). As the mechanical stress may produce the polarization of dielectrics (the piezoelectric effect), similarly the deformations of the nematics induce a nonzero electric polarization. Within the droplet structure, the study is complicated by the liquid crystal/polymer interaction. After the stretching the droplet directors prefer to orient along the major axis of the ellipsoids because the elastic energy of the bipolar nematic structure is a minimum in this position. Accordingly, the order parameter S will be nonzero, leading to an induced polarization P in the stressed material.

CONCLUSIONS

In this paper we presented the results of the hysteresis study on a stretched polymer dispersed nematic liquid crystal E7/PMMA at different temperatures, frequencies and amplitudes of the applied electric field. We conclude that the presence of the induced polarization is correlated with the nonzero value of the order parameter S of the elongated droplets.

The saturation of the polarization with the applied voltage was absent, because the amplitude of the applied electric field is upper limited by the Joule heating.

The frequency dependence of the saturation polarization leads to relaxation times of milliseconds, comparable with the values of the optical switching time.

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