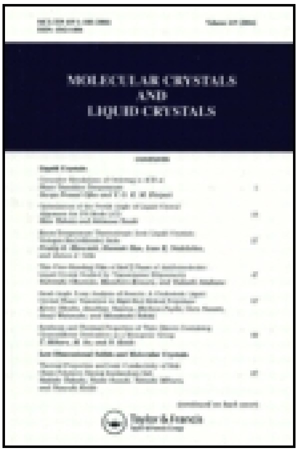

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Linear Light Modulation by a Polymer Dispersed Chiral Nematic

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LINEAR LIGHT MODULATION BY A POLYMER DISPERSED CHIRAL NEMATIC

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Abstract Recently, a new class of light modulators, based on PDLC systems have been reported[1,2]. Here, we present a Polymer Dispersed Chiral Nematic (PDCN) utilizing the linear electro-optic effect in chiral nematics with uniformly aligned helix under an electric field, applied normally to the helix axis[3]. A fairly good linear response has been detected in such PDCN films, containing a chiral nematic mixture 6415 L (Hoffmann La Roche), possessing a short pitch (about 0.3 μm), temperature independent in wide temperature interval (0°C - 50°C). Response time of about 100 μs has been measured.

INTRODUCTION

Polymer dispersed liquid crystals (PDLC)[4] are composed of micron-sized liquid crystal droplets, dispersed in a transparent and optically isotropic polymer matrix. The classical example of PDLC is the one containing nematic which is a field controlled light scattering device. In the off-state the director of the nematic droplets in this device is randomly oriented while in the on-state it becomes uniformly oriented due to the electric field. Thus, the device could be electrically switched between an opaque light scattering "off"-state and a non-scattering fully transparent "on"-state. The scattering-type PDLC could also work in the opposite mode, i.e. the on-state is the scattering state and off-state is the transparent one. In fact, the electro-optic effect in these PDLCs is dielectric by origin and thus it is quadratic with the field. Typically, the response time of the effect is in the millisecond range. Some of the advantages of scattering type PDLCs are that one does not need crossed polarizers and the displays can be made flexible. However, in order to obtain a better scattering state, the PDLC film should have a thickness of more than 10 μm and size of the liquid crystal droplets of less than 1μm. But, the thicker is the layer and the smaller are the droplets, the higher is the threshold voltage of the device. Another disadvantage of these devices is the strong angular dependence of the scattered light by PDLC. There are also PDLCs utilizing chiral nematic liquid crystals giving switchable selective reflection displays[5]. The electric field effects on the droplet structure in polymer dispersed liquid crystals have been studied in details by several authors[6].

Recently, new types of polymer dispersed liquid crystal devices have been presented. They are combining the polymer matrix structure of the classical PDLC and the electro-optic effects of chiral smectic liquid crystals[1, 2]. In these new devices, the linear coupling between the applied electric field and the spontaneous P_s in ferroelectric, or induced polarization P_i in paraelectric liquid crystals, are used. In these systems, the polymer film contains prealigned liquid crystal droplets and it behaves like an optical wave plate with an electrically switchable in-plane direction of the optic axis. Thus, if the device is placed between crossed polarizers, the transmitted light intensity can be modulated by an applied electric field. Ferroelectric and paraelectric types of PDLC exhibit much shorter response times than the classical PDLC (microsecond range for ferroelectric and sub-microsecond range for paraelectric liquid crystals). However, these devices require two crossed polarizers which might be considered as a disadvantage.

There is also another type of linear electro-optic effect which takes place in chiral nematics. As known, a short pitch chiral nematic (pitch of the order of the wave length of transmitted light) with uniformly aligned helix axis, in a so-called stripe-like texture, acts macroscopically as a uniaxial birefringent plate with its optic axis coinciding with the helical axis. An electric field applied normal to the helix axis induces a flexoelectric polarization[7]. As consequence, an effective in-plane rotation of the optic axis, perpendicular to the applied field, takes place. This effect is studied by several authors [3, 8, 9]. For low fields, the induced deviation of the optic axis is linear with the field which gives a linear electro-optic response. The typical response time of the effect is less than 100 μs. For high fields, however, the linearity of the response becomes distorted due to field-induced helix unwinding [8, 9].

Here, we present another type of PDLC system, utilizing the linear electro-optic effect in short pitch chiral nematics; so-called Polymer Dispersed Chiral Nematic (PDCN).

RESULTS AND DISCUSSION

One of the most important factors concerning the linear electro-optic effect in chiral nematics is to create a texture with a uniform alignment of the helix axis, oriented at 90 degrees with respect to the PDCN film normal. The uniform in-plane orientation of the helical axis is crucial for the effect. Generally speaking, in the conventional sandwich cells (liquid crystal layer confined between two glass plates), a chiral nematic can adopt two types of texture, Grandjean, with the helix axis perpendicular to the confining surfaces, and fingerprint, with the helix axis parallel to the surfaces. By choosing homeotropic surface conditions in a sandwich cell one usually may obtain a fingerprint texture. Then, by unidirectional shear of the cell substrates, a uniform in-plane alignment of the helix axis can be achieved. In such a texture, the helix is aligned along unique direction, perpendicular to the shear. This texture is called stripe texture and in the large pitch chiral nematics, placed between crossed polarizers, it appears as a sequence of dark and bright bands[10, 11]. On the other hand, only for chiral nematics with positive dielectric anisotropy, one can use also planar boundary conditions giving Grandjean texture. In this case, an electric field has to be applied in order to obtain a fingerprint texture. However, it might be difficult without field to keep for enough long time the stripe texture achieved by mechanical shear because of the tendency of the liquid crystal to relax back to the initial Grandjean texture.

Moving from the conventional sandwich cell to the polymer dispersed liquid crystal system, we have to underline some major differences. First, in the sandwich cell, the liquid crystal layer is confined between two substrate surfaces whereas PDCN consists of a lot of single microsized liquid crystal droplets where the total interface area is much greater compared to the one in the sandwich cell. Thus, the influence of surface-liquid crystal interactions on the electro-optic response in PDCN is expected to be strongly enhanced. Second, the effective strength of the electric field in the droplets may differ substantially from the strength of the applied field to the PDCN film[12]. Third, the symmetry of the sandwich cell and the one of the droplets are different. The last one makes impossible the achievement of the required unidirectionally aligned stripe texture of chiral nematic in the droplets and thus no linear electro-optic response at all. Obviously, one has to assure similar conditions in the droplets to those in the sandwich cell in order to obtain a maximum response. If we want to transform the symmetry of the single droplet to the one of the sandwich cell we have to deform in a proper way the PDCN film and thus the droplets as well-for instance, by stretching and/or by pressing it. In the sample preparation two general steps are of major importance - creating homeotropic surface conditions and deforming the droplets. The last step gives the uniform alignment of the chiral nematic.

The PDCN films were made by dispersing the chiral nematic liquid crystal mixture 6415L (Hoffmann-La Roche), containing less than 3%wt of lecithin, in Poly(vinyl)butyral (PVB)(Aldrich). The mixture 6415L is a short pitch material which have a temperature independent pitch of 0.3 μm between 0°C and 50°C. Samples were prepared using the Solvent Evaporation Induced Phase Separation (SIPS) technique. Lecithin was dissolved in the liquid crystal at room temperature. PVB was added to this mixture to form the final composition of 59 % PVB, 38% 6415L and 3% lecithin, by weight. Chloroform was used to dissolve completely all the components and to obtain a homogeneous solution which was spread onto a glass substrate. On solvent evaporation at room temperature, an opaque membrane was formed and then it was removed from the support and imposed on mechanical treatment, in order to align the chiral liquid crystal within the droplets. By pressing the PDLC film one creates geometric anisotropy of the droplets. Thus, the droplet surfaces which are parallel to the PDLC film are enlarged whereas the others perpendicular to it are diminished. Of course, just pressing the PDLC film does not give any preferable direction of the director since we then have a radial symmetry in each droplet and a degenerate director alignment is expected. To destroy the radial symmetry of the droplets we stretch the film unidirectionally thus elongating the droplets in the stretching direction (see Figure 1). The stretching of the film, however, gives two important results. First, it creates a liquid crystal flow inside the droplets, similar to the shear flow in the sandwich cell. Secondly, the stretching process also creates anisotropy of the polymer matrix which may affect the polymer-liquid crystal surface interactions in each droplet. Consequently, in presence of homeotropic surface conditions, by unidirectional stretching we are able to orient the

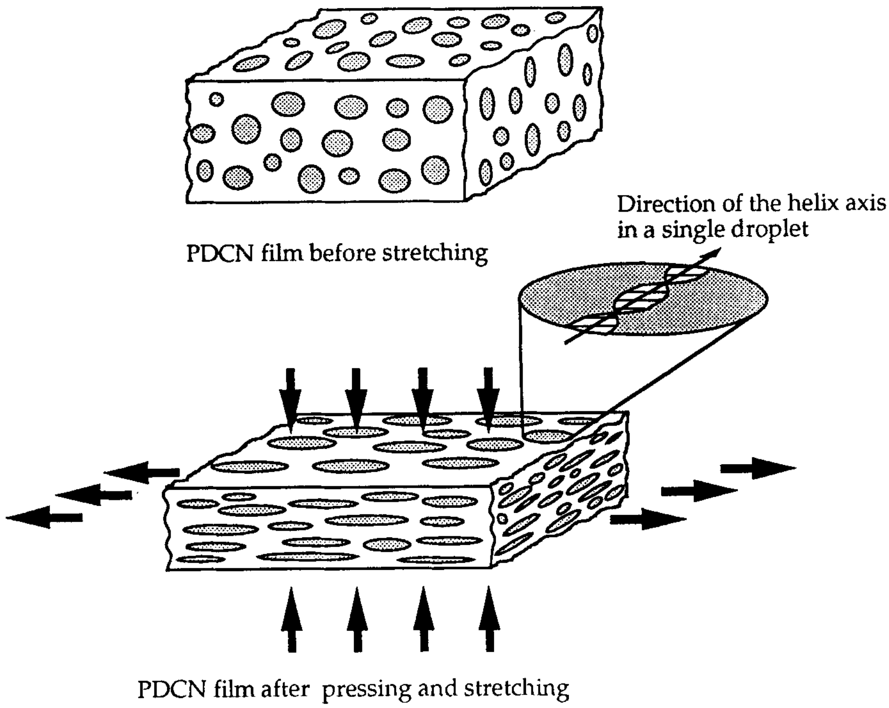

FIGURE 1 PDCN film before and after pressing and stretching. The helix axis in the prolonged and flattened out droplets orients perpendicularly to the stretching direction.

helix axis in a unique direction. Then the stretched films were sandwiched between two glass substrates, covered with transparent conducting layers. For a good contact between the PDCN film and the glass substrates, the latter were covered with thin layer of an epoxy glue (Bostik, Boston S.p.a , Italy). So, the measured total thickness of PDCN film together with the glue was about 10 μm. For obtaining homeotropic surface conditions one can use as a polymer matrix polysiloxane RN 783 (Nissan) instead of PVB and Lecithin.

A photograph of the PDCN film with the droplets clearly prolonged shape along the stretching direction is shown in Figure 2. However, the uniform alignment of the helix in the droplets is far from the perfect one and therefore the contrast in the transmitted light intensity is very small. More investigations of the director field in the polymer dispersed chiral nematic droplets have to be done to find a way for improvement of the uniform helix alignment in PDCN films.

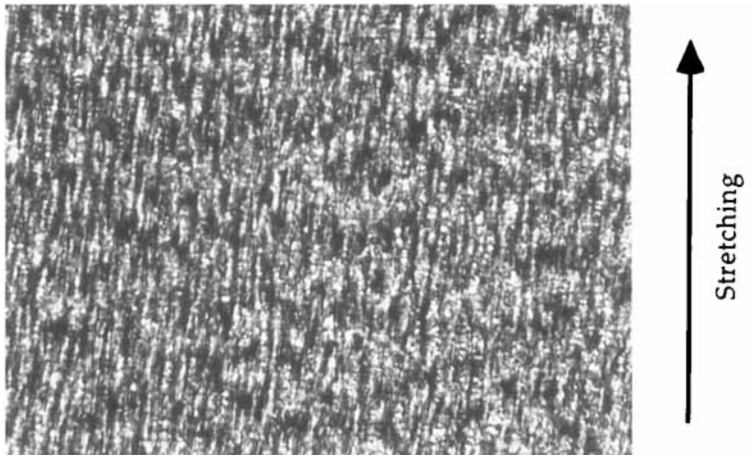

FIGURE 2 Photograph of the PDCN film. The droplets are prolonged in the stretching direction See Color Plate VI.

The stretching procedure may be performed either when the liquid crystal is in the isotropic phase, hot stretching, or in the chiral nematic phase, cold stretching. The hot stretching eliminates the flow alignment of the liquid crystal during the stretching while the cold stretching includes it. Both kinds of stretching, however, give droplets with anisotropic form and with anisotropic boundary conditions at the liquid crystal-polymer interfaces.

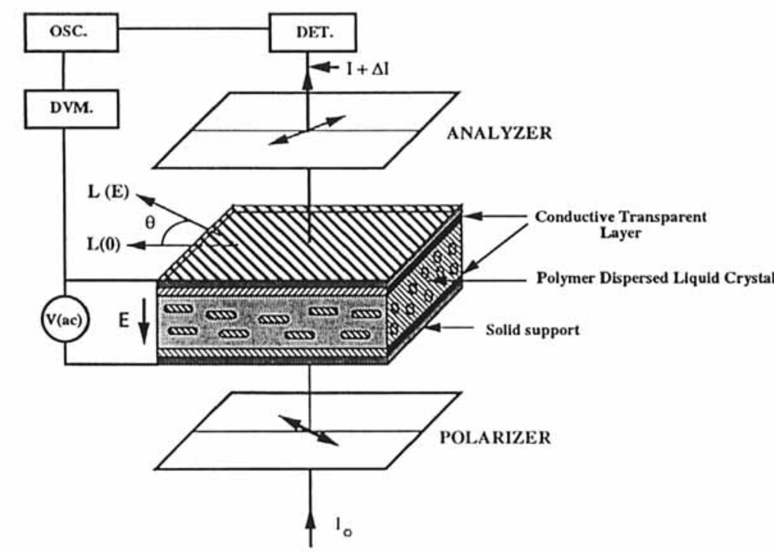

FIGURE 3 Experimental set-up

The optic and the electro-optic characteristics of PDCN films were studied in the set-up depicted in Figure 3. Investigations of the phase shift of

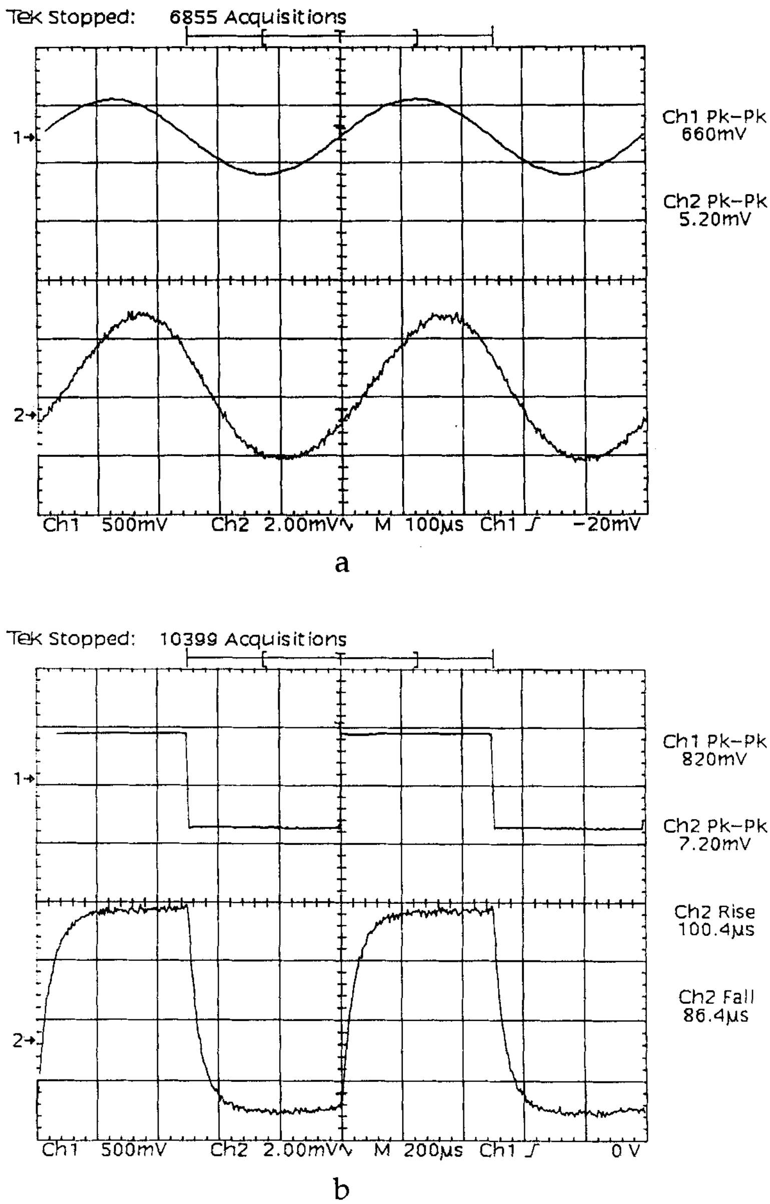

FIGURE 4 a) The electro-optic response (lower curve) for a cold stretched PDCN film under an applied sine-wave voltage (upper curve) at room temperature, f = 2 kHz and b) the response times for a bipolar applied voltage.

transmitted light through the hot and cold stretched samples show that the helix axis was oriented perpendicular to the stretching direction, situation similar to the one in the sandwich cell where the helix axis orients in a direction perpendicular to the shearing. The main reason for the helix to be oriented perpendicularly to the stretching direction seems to be the stretching-induced anisotropy of the polymer matrix. This idea is supported by the fact that both hot and cold stretched films show unidirectional alignment of the helix axis. The polymer chains due to the stretching become more or less straightened out and aligning along the stretching direction. We hereby introduce a preferable direction of the liquid crystal molecules at the chiral nematic-polymer interface (similar to the effect of rubbed polymer surfaces). Obviously the final shape of the liquid crystal droplets is another factor which certainly play an important role.

In Figure 4a is shown the electro-optic response in a cold stretched sample, between crossed polarizers, under a sine voltage at frequency 2 kHz and amplitude 66 volts peak to peak (V_{pp}). Despite the slight phase shift, the response is clearly linear because of the linear deviation of the optic axis with the applied field. The response times were measured to be of order of 100 μs as shown in Figure 4b. The field induced deviation of the optic axis for the same sample is shown in Figure 5. The hot stretched samples were found to give similar results as the cold stretched ones.

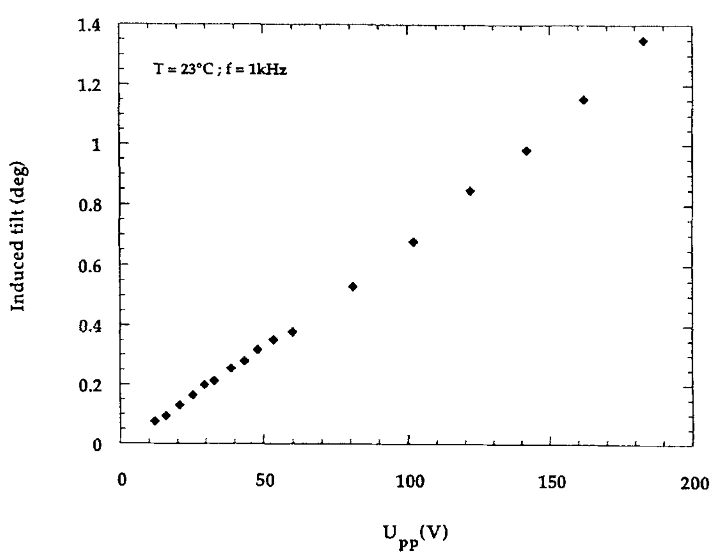

FIGURE 5 Field induced deviation of the optic axis in a cold stretched PDCN film as a function of the applied voltage.

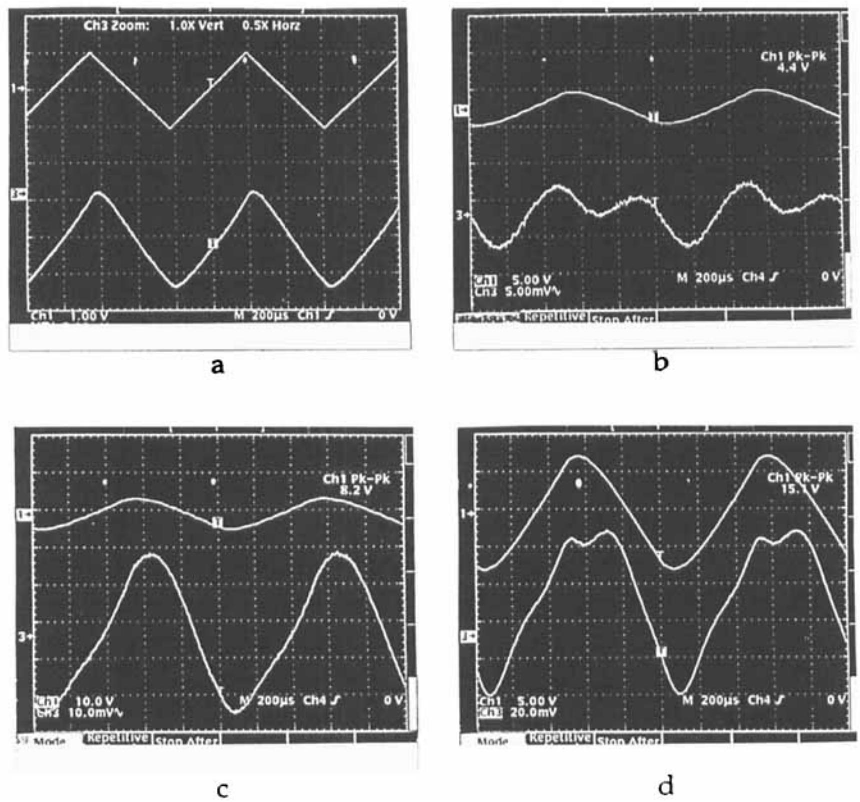

FIGURE 6 Electro-optic response in a PDCN film containing the chiral nematic mixture 6415L with $\Delta\varepsilon > 0$.
a) U_{PP} = 200V. The response has a linear character.
b) U_{PP} = 440V. The dielectric torque, switching the position of the helix, strongly affects the linearity of the response.
c) U_{PP} = 820V. A complete uniform alignment of the helix axis is achieved, and the response is again linear.
d) U_{PP} = 1500V. The response is disturbed by field induced helix unwinding

A chiral nematic with positive dielectric anisotropy (6415L has $\Delta\varepsilon>0$) can be transformed from Grandjean to fingerprint texture by applying an external electric field. This also means that if the uniform planar alignment of the helix axis has partially relaxed to Grandjean texture, one can restore the planar alignment of the helix by applying an electric field. In Figure 6 is shown the electro-optic response of a PDCN film for different values of the applied voltage. In Figure 6a (U_{pp} = 200V) the response has clearly a linear character. Increasing the applied voltage, however, the linear response is strongly affected from field induced reorientation of the helix within the domains where the helix axis has a tilted position with respect to the film plane, Figure 6b (U_{pp} = 440V). In this case, the dielectric torque is changing the position of the helix from tilted to quasi-planar. This type of switching is quadratic with the field and gives such an impact on the detected response. At high enough voltages the texture in the droplets is fully transformed to a stripe texture, Figure 6c (U_{pp} = 820V). This gives again a linear electro-optic response but with much higher amplitude and contrast than before. Consequently, from the very beginning, the liquid crystal alignment in the droplets has not a perfect stripe texture. However, the applied electric field improve the uniformity of this texture within the droplets. At U_{pp} = 1500V, Figure 6d, a dip in the response takes place, which suggests a field induced unwinding of the helix[8, 9]. The linear electro-optic effect in short pitch chiral nematic could be strongly affected by the field induced unwinding of the helix at high fields. In PDCN, however, even at very large applied voltages (> 2000 V_{pp}) the helix in the droplets was not completely unwound. It has to be pointed out that the critical voltage of helix unwinding in a 2 μm thick sandwich cell was about 50 V_{pp}[8, 9]. Either the electric field inside the droplets of PDCN film is not the same as the applied electric field[12] and/or the surface-liquid crystal interactions strongly support the director distribution in the droplets. The experimental results show also that in the sandwich cells, the induced deviation of the optic axis (about 10 degrees peak to peak[8,9])was larger than in PDCN films, probably for the same reasons as those just mentioned.

CONCLUSION

PDCN film seems to be promising for applications in the field of light modulators. It is flexible and its response time is very short. This system is a rather complex one since a lot of factors may affect the electro-optic response of PDCN. The choice of polymer material, chiral nematic liquid crystal and mechanical treatment are among them. Surface-liquid crystal interactions are also playing an important role. Therefore, further studies of this system are necessary to be performed in order to optimize it.

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