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CONTROL OF ELECTRO-OPTIC MODULATION IN POLYMER-DISPERSED FERROELECTRIC LIQUID CRYSTALS

JAE-HOON KIM, KYEHUN LEE†, and SIN-DOO LEE*
Physics Department, Sogang University, C. P. O. Box 1142, Seoul, Korea
†Samsung Electronics Co., Ltd., Kiheung, Yongin, Kyungki, Korea

Abstract Electro-optic (EO) modulation in polymer-dispersed ferroelectric liquid crystals (FLCs) has been studied as a function of the droplet size in a confined geometry. It is found that an optimum droplet size exists for the best alignment of molecules as well as smectic layers which produces the maximum EO modulation. The alignment quality depends critically on the shape and size of FLC droplets. The stable structure formed inside the FLC droplets results from a competition between the elastic energy stored in a confined geometry and surface interactions at the FLC-polymer interface.

INTRODUCTION

Inhomogeneous composite materials consisting of liquid crystals (LCs) and polymers have been attracted considerable attention for a basic understanding of a confinement effect as well as for practical applications in the area of flat panel displays.[1,2] One of them is a polymer-dispersed ferroelectric liquid crystals (PDFLCs) made up of micron-sized FLC droplets formed in a continuous polymer matrix.[3,4]

The physical properties of PDFLCs are generally affected by a number of parameters such as the compatibility between a polymer matrix and a LC used, the mixing ratio of two materials, the intrinsic molecular chirality, the droplet size and its shape. Among them, of particular importance is anisotropic interfacial interactions between the droplets and the polymer matrix in a confined geometry which depends primarily on the chemical properties and the interfacial ordering of the materials used. In PDFLCs, the helix unwinding process in addition to the collective molecular reorientation will play a role in producing an electro-optic (EO) effect. Therefore, it is important to study the relationship between the FLC droplet structure formed in a polymer matrix and the associated EO response of PDFLCs. Moreover, a basic understanding of the aligning mechanism for FLC droplets is of great interest for practical applications.

In this paper, we report on the EO response of PDFLCs with various droplet sizes in a confined geometry. It is found that a PDFLC made up of 3μm droplets exhibits the maximum EO effect, and its switching time is on the order of millisecond. Some aspects of the confinement effect will be described in terms of the droplet size.

EXPERIMENTAL

PDFLCs consist of micro-sized FLC droplets formed in a continuous polymer matrix. In this study, PDFLCs were prepared using a polymerization induced phase separation process during which a phase separation by photo-polymerization occurred in a homogeneous mixture of FLCs and a prepolymer. A ultraviolet (UV) light is commonly used for photo-polymerization.[5] The FLC material used was CS1024 of Chisso Petrochemical Co. which has a relatively long helical pitch (20 μm) in the smectic C* phase. It undergoes the smectic A - smectic C* (Sm A-Sm C*) phase transition. The prepolymer used for a matrix was NOA61 of Norland Co. which is a UV-curable blend of monomers, oligomers, and a photoinitiator. The mixing ratio of NOA61 to CS1024 was 8 to 1 in weight. The mixed solution became uniform at 120°C in the isotropic state. The UV intensity was varied up to 15 mW/cm^2 for controlling the size of the FLC droplets in the polymer matrix.

The sample cell was made with conductive indium-tin-oxide coated glasses which were treated with polyimides. The thickness of the polyimide layer was about 300 Å. The polyimide layers of the cell were unidirectionally rubbed, so that the alignment of both NOA61 polymers and FLC molecules in droplets was produced to some extent.[4] The cell gap was maintained by glass spacers of 10μm thick.

The EO measurements on PDFLCs were carried out by monitoring the change in the transmitted intensity of the light through the cell between crossed polarizers. The sample cell was placed such that one of the crossed polarizers made an angle of 22.5° with respect to the optic axis of the cell so that the maximum transmission was obtained. All the measurement were performed at room temperature.

RESULTS AND DISCUSSION

We first examine the size effect on the quality of alignment and the EO modulation in PDFLCs. We prepared four different samples with the droplet size of about 1μm, 3μm, 5μm, and 7μm in diameter by varying the intensity of UV irradiation from $0.1 mW/cm^2$ to $15 mW/cm^2$. Fig. 1 shows the results for the FLC droplet size formed in the polymer matrix as a function of the UV intensity. As shown in Fig. 1, the droplet size decreases with increasing the UV intensity. Since the polymerization rate of prepolymer increases with increasing the UV intensity, the higher intensity produces the smaller droplets. The critical temperature for the Sm A - Sm C* transition in the PDFLC becomes lower by $(5 \sim 10)^{\circ}C$ than that in the bulk FLC cell, indicating that the size effect should be taken into account for PDFLCs.[6] Note that the shift of the critical temperature becomes larger as the droplet size decreases.

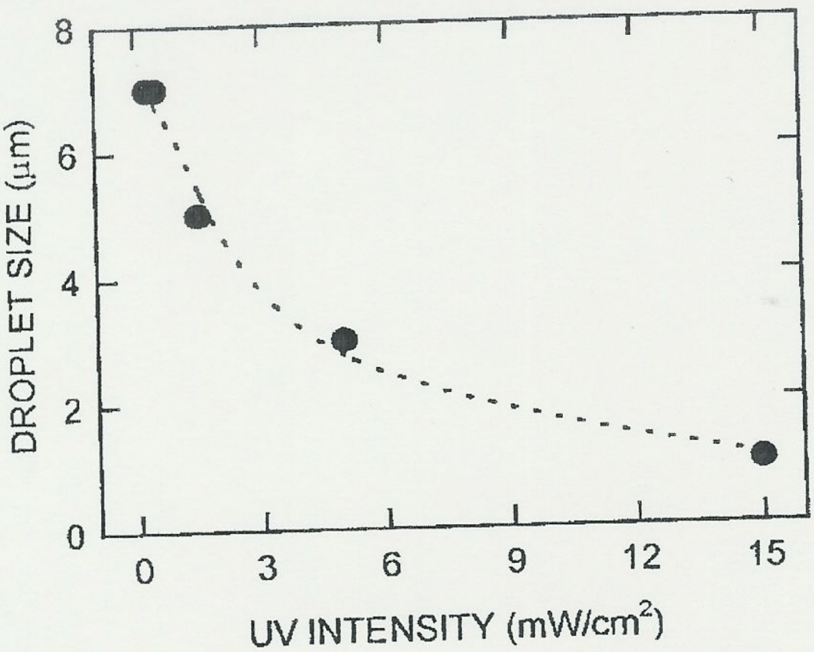

Figure 1: Droplet size as a function of the UV intensity.

From the scanning electron microscopic observation, it was found the FLC droplets of $1\mu m$ were found to be fairly symmetric and uniformly distributed between two substrates. However, the droplets of $7\mu m$ were mostly distributed near the substrates and their shapes were quite irregular. The amount of the elastic distortions involved depends on the gradient of the droplet curvature, and thus the larger gradient of the curvature (for the smaller droplet size) and the more irregular shape make molecular alignment more distorted in the droplets. This implies that there should exist an optimum droplet size between $1\mu m$ and $7\mu m$ for our PDFLCs. In fact, the optimum size was found to be $3\mu m$, which will be discussed later on. Moreover, the alignment in the droplets will be also affected by other intrinsic material parameters such as the helical pitch, the molecular tilt, the spontaneous polarization, and the phase sequence.

We now discuss the EO modulation of the PDFLCs of different droplet sizes. Measurements were made with a square wave voltage at 50Hz whose amplitude was varied. Fig. 2 shows the optical transmission through the PDFLCs of different droplet sizes.

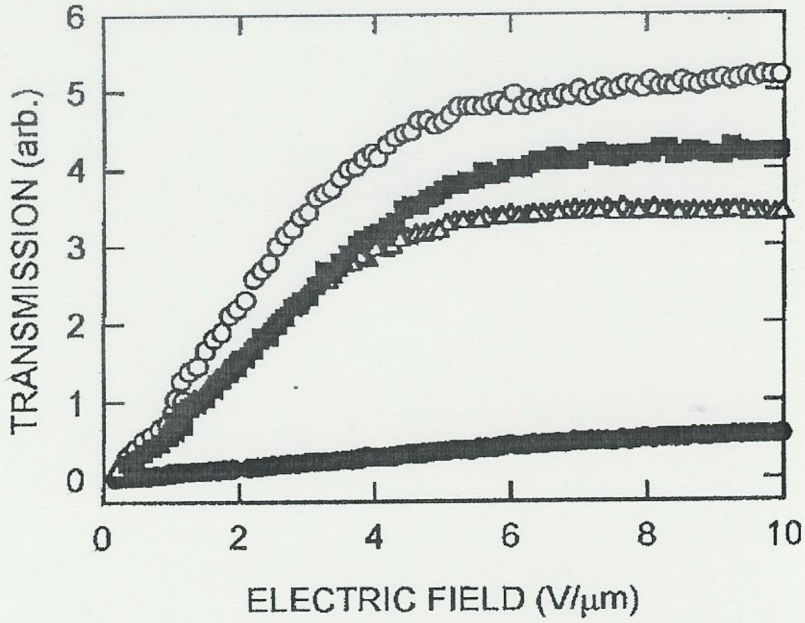

Figure 2: The optical transmission as a function of the electric field through PDFLCs of different droplet sizes. The filled circles, open circles, filled squares, and open triangles represent the droplet size of 1μm, 3μm, 5μm, and 7μm, respectively.

As already discussed above, the maximum transmission was obtained for the PDFLC of 3μm droplets. This results from a competition between the elastic energy stored in a confined geometry and surface interactions at the FLC-polymer interface. The transmission exhibits a nearly linear behavior at relatively low fields while it becomes to saturate with increasing the field above 4V/μm. The physical origin comes from the unwinding of the helix and the rotation of the molecules with keeping the tilt angle fixed in the Sm C* phase. Note that for the PDFLC of 1μm droplets, the transmission is very small and almost linear in the field since a large curvature elasticity reduces the response of the molecules and the smectic layers inside the droplet to the field.

Fig. 3 shows the rising time in the field-driven state for PDFLCs of different droplet sizes as a function of the electric field E. As reported previously,[4] the rising time is given by $\tau_{on} = \eta/[PE + K(\ell^2 - 1)/a^2]$ where η is the relevant viscosity, K the effective elastic constant, and P the spontaneous polarization. Here, $\ell = a/b$ denotes the aspect ratio of the droplet with a the semi-major axis and b the semi-minor axis.

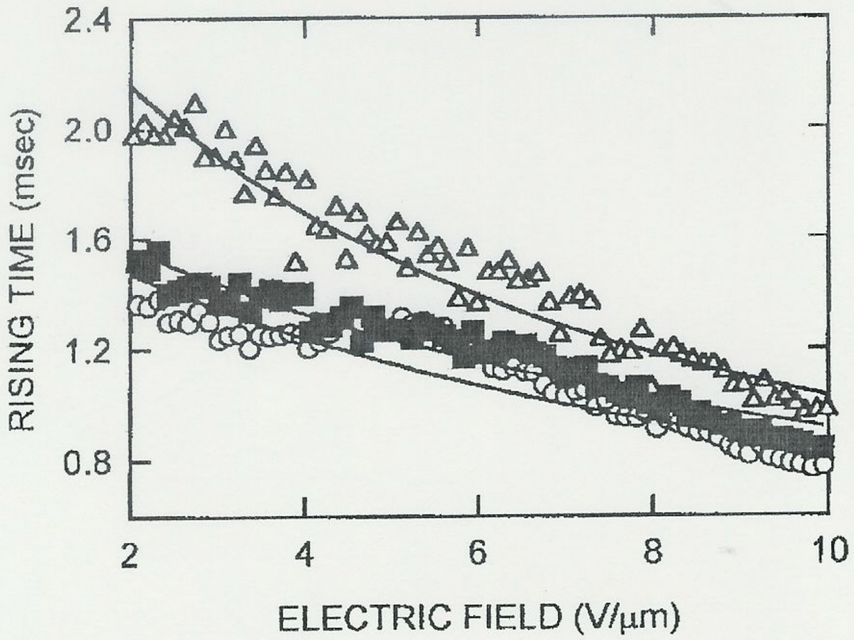

Figure 3: The rising time as a function of the electric field for PDFLCs of different droplet sizes. The open circles, filled squares, and open triangles represent the droplet size of 3μm, 5μm, and 7μm, respectively.

Since the relaxation time in the field-off state $\tau_{off} = \eta a^2/K(\ell^2 - 1)$, the rising time τ_{on} can be rewritten as $1/(PE/\eta + 1/\tau_{off})$. The solid lines in Fig. 3 are the least-square fits of the data. The fitted values of P/η, τ_{off}, and η for PDFLCs of different droplet sizes are collected in Table I. The spontaneous polarization, independently measured using the triangular method, was $P \approx 1$ nC/cm^2, which is an order of magnitude smaller than that in the bulk FLC. This is, of course, one of the confinement effects.[6] The viscosity was found to be in the range of $(0.14 \sim 0.18)$ kg/ms, which is in good agreement with the literature value.[7] As shown in Table 1, the relaxation time τ_{off} decreases with decreasing the droplet size since a larger elastic energy will be stored in a smaller droplet.

CONCLUDING REMARKS

We have studied how the size and shape of FLC droplets in a confined geometry in-

Table 1: The fitted values of material parameters for PDFLCs of different droplet sizes

	3 μm	5 μm	7 μm
P/η ($\times 10^{-3}$Cs/kgm)	5.81±0.30	6.41±0.26	6.91±0.20
τ_{off}($\times 10^{-3}$s)	1.68±0.04	2.02±0.05	3.26±0.11
η (kg/ms)	0.18±0.01	0.16±0.01	0.14±0.01

fluence the macroscopic EO modulation in PDFLCs. An optimum droplet size exists for uniform alignment of the molecules as well as smectic layers formed in a polymer matrix. It is suggested that anisotropic interactions at the polymer/FLC interface play an important role in the structural and dynamical properties of PDFLCs. As the droplet size decreases, the hindrance of the collective molecular rotation becomes profound, and thus the relaxation phenomenon in the droplet appears to be quite different from that in the bulk FLC.

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