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Hysteresis of Optical Transmission in Ferroelectric Liquid Crystal by Winding and Unwinding Motions of Helical Structure

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Characteristics of the electro-optic effect originating in the transition between wound and unwound states of the helicoidal structure have been studied in detail. The fast response of the decrease of the transmission at low voltage application has been related to the helical pitch and explained in terms of the reorientation of the surface layers and the disturbance of the boundary between surface layer and the interior helicoidal structure. Extremely low threshold voltage and short response time are found to be realized with a ferroelectric liquid crystal of large spontaneous polarization. A method to reduce the decay time and also a self bias effect have been proposed.

Keywords: ferroelectrics, liquid crystal, ferroelectric liquid crystal, electrooptic effect, optical switching

I. INTRODUCTION

Recently, ferroelectric liquid crystals have attracted great interest as the first non-solid ferroelectrics and also as a new type of material which can be used as a fast electro-optic element. Already several types of applications of ferroelectric liquid crystals as optical-switching and display devices have been reported.^{1,2,3,4} The first proposal

of the electro-optic effect in ferroelectric liquid crystals was done many years ago by us, in which the difference of the optical transmissions between wound and unwound states of a helicoidal structure of ferroelectric liquid crystal was utilized. However, because of the relatively high threshold voltage and low response speed, this effect has not attracted much attention and most efforts in the industry have been exerted to the study of a surface stabilized ferroelectric liquid crystal (SSFLC) cell.3 This SSFLC cell has several advantages like a short response time and low threshold voltage, but it needs the use of optical polarizers and ultra-thin cells less than several µm which is practically not so easy. On the other hand, the effect utilizing winding and unwinding transition does not need any polarizer and also thick cells can be used. However, the characteristics and mechanisms of the winding and unwinding effect of the helicoidal structure have not yet been completely revealed. It will be also very important to study the detailed characteristics of this effect for the understanding of the fundamental aspect of ferroelectric liquid crystals and the dynamics of the helicoidal structure.

In this paper, we will report on the characteristics of this effect and also will explain that by the selection of appropriate material an extremely low threshold voltage and a high response speed can be realized even by this simple effect.

II. EXPERIMENTAL

Two types of materials are used in this study; DOBAMBC(p-decyloxybenzylidene-p'-amino-2-methylbutylcinnamate) and 3M2CPOOB ((2S,3S)-3-methyl-2-chloropentanoic acid-4',4"-octyloxybiphenyl ester). Synthesis and purification procedures of these materials were already reported in our previous papers. 1.5.6.7

The ferroelectric liquid crystals were sandwiched between two nesa coated glass plates whose surfaces were rubbed in a direction to obtain homogeneous alignment. The distance between electrodes was fixed by the thickness of the spacer sheets (films of polyethyleneterephthalate). The sample cell was set in the optical path of a He-Ne laser (6328 Å) and the change of the transmission by the application of the electric field was monitored by a photodiode.

For the study of the fundamental process of the dynamical response of the helicoidal structure, a Fast Fourier Transform Analyzer (VS-3310A) was introduced for the first time as shown in Figure 1.

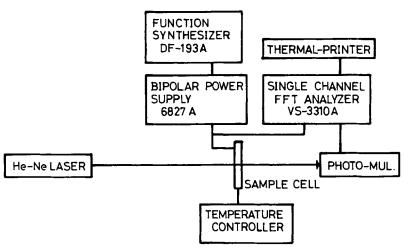


FIGURE 1 Experimental set up for the measurement of the Fast Fourier Transform analysis.

III. RESULTS AND DISCUSSION

Figure 2 indicates the dependence of the transmission intensity through the cell on the applied voltage. As evident from this figure, in the low voltage range the transmission decreased slightly and then above some threshold voltage, V_{th} , it increased remarkably tending to saturation. Such a behaviour in a ferroelectric liquid crystal was reported by us many years ago.¹

The threshold voltage, V_{th} , therefore also the threshold field E_{th} , was a function of the sweep rate of the applied voltage. When the sweep rate was decreased, E_{th} decreased as shown in Figure 3(a). However, as long as the sweep rate dV/dt was less than 0.1V/sec, characteristics such as curve (1) in this figure were obtained. Therefore, the experiments were performed with the sweep rate of 0.1V/sec.

Such a difference of E_{th} with the sweep rate can be explained if the main response above E_{th} (increase of transmission) was a much slower process compared with the first smaller response below E_{th} (decrease of transmission). The main response above E_{th} has been interpreted in terms of the transition from the helicoidal structure under no or low bias voltage to the unwound state at higher voltage above E_{th} . Namely, it is well known that the ferroelectric liquid crystals form a helicoidal structure under free state due to the chirality of the molecule. Under the application of a high voltage, the heli-

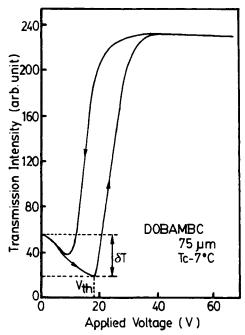


FIGURE 2 Typical voltage dependence of the transmission intensity through the DOBAMBC cell.

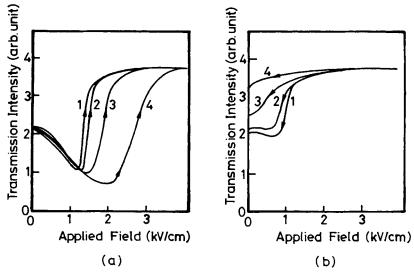


FIGURE 3 Field dependence of the transmission intensity through the DOBAMBC cell with 100 µm thickness for various sweep rates; 1;0.1, 2;1.0, 3;10, 4;100 (V/sec).

coidal structure is unwound and turns into the uniform monodomain structure. The change of the light scattering due to this transition between two states results in the change of the light transmission.

The threshold field was related to the spontaneous polarization P_s by the equation

$$E_{th} \propto K/L^2 P_s \tag{1}$$

where K and L are the torsional elastic constant for the helix and the pitch of the helix, respectively.

On the other hand, the origin of the first small response below $E_{\rm th}$ has not been completely explained until now, though the importance of the surface layer has been suggested.

The transmission change in the case of lowering voltage is also shown in Figure 3(b). In this case, to obtain a sweep rate independent result, the sweep rate must be also less than 0.1V/sec. It is evident from both Figures 2 and 3 that there exists remarkable hysteresis in the voltage dependence of the transmission. In some cases, however, we observed the interesting and unexplained result shown in Figure 4, in which the transmission intensity continued to increase in the case of lowering the voltage until the second threshold V_{th2} . Such characteristics were observed in the case of thin cells and at high temperatures, though their origin has not been clarified.

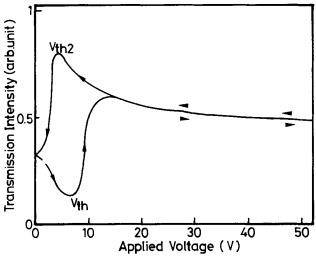


FIGURE 4 Voltage dependence of the transmission intensity through the DOBAMBC cell with 25 μm thickness at 90 °C.

It should be also pointed out that the absolute transmission level depends on the condition of the molecular alignment and on the temperature.

When the step voltage was applied to the cell, the transmission changed as shown in Figure 5, which depended on the condition of voltage application. At low voltage, the transmission decreased as shown in Figure 5(a) which corresponds to the low voltage range below V_{th} in Figure 2. In the case of higher voltage application, the transmission decreased at first and then increased remarkably as shown in Figure 5(b). Superposition of the step voltage on the small bias voltage only gave an increase of the transmission as shown in Figure 5(c). These behaviours are consistent with voltage dependence of the transmission shown in Figure 2.

It should be noted that the response time of the decrease of the transmission (Figure 5(a) and the initial part of Figure 5(b)) is very short, less than several hundred µsec. As evident from Figure 6, this response time of the fast part decreases with increasing field. Such a fast response was also already reported by us.¹

On the other hand, the main response (increase of transmission) is relatively slow. Figure 7 shows the voltage dependence of the response time of this slow main component. This result is consistent with those of Figure 3. Namely, this result supports the interpretation of the sweep rate dependence of hysteresis shown in Figure 3.

As already mentioned the main slow response was well interpreted

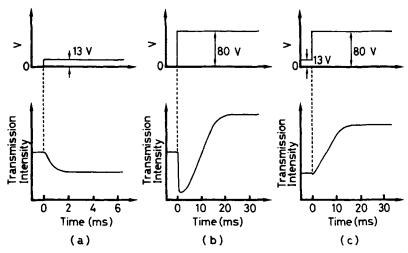


FIGURE 5 Response to various step wise voltages in the 75 µm cell of DOBAMBC.

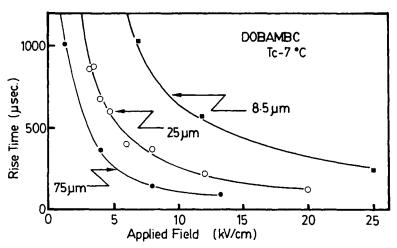


FIGURE 6 Field dependence of the response time of the fast component in DOBAMBC.

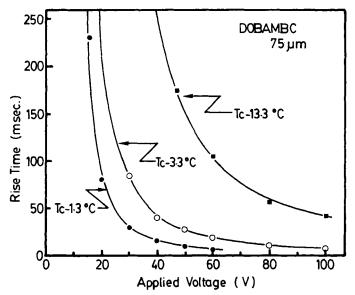


FIGURE 7 Voltage dependence of the response time of the slow main component in DOBAMBC.

in terms of the unwinding of the helicoidal structure. However, the fast small response has not been clarified in its origin. We found that the magnitude of the decrease of the transmission δT (fast response) defined as shown in Figure 2 was relatively small just below T_C (transition between chiral smectic C and smectic A phases). Then indicating a sharp peak, again it decreased with decreasing temperature, tending also to saturate as shown in Figure 8. If we compare this Figure with the temperature dependence of the helical pitch shown in Figure 9, it is quite clear that their temperature dependences have many points of resemblance. These results clearly indicate that the decrease of the transmission has some relationship to the pitch of the helix.

The fast response can be also related to the reorientation of the molecular alignment in the layer near the electrode surface. Generally, in the surface layer, the helicoidal structure should be unwound

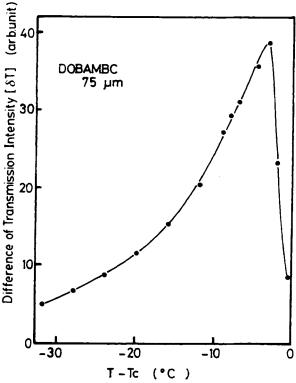


FIGURE 8 Temperature dependence of the transmission decreasing δT in DOBAMBC.

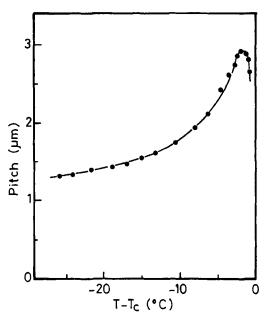


FIGURE 9 Temperature dependence of the helical pitch of DOBAMBC.

and an uniform alignment should be obtained. In such a case, the P_sE torque works quite effectively, which should result in the fast response because of this large torque. Decrease of the transmission should originate from the change of the thickness of the surface layer, or the deformation of the boundary between the surface layer and the interior helicoidal structure. The applied field will induce a fluctuation of the boundary due to the reorientation of the surface layer, which may result in the decrease of the transmission in a short time as observed.

For a full understanding of the dynamics of the helicoidal structure and the domain switching it is necessary to study the characteristics of the response in the time and the space and the frequency domains. Therefore, we started to study the change of the transmission intensity at the instant of the voltage application of the reverse polarity with the experimental system already shown in Figure 1. In this case, the transmitted light through the cell was monitored by a photomultiplier and the signal was analyzed using the Fast Fourier Transform Analyzer VS-3310A. Figure 10 shows an example of the observation of the transmitted light with this apparatus. As evident from this Figure, we can observe the details of the dynamics directly. A detailed study utilizing this system is now under progress.

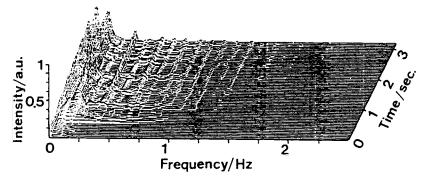


FIGURE 10 One example of the observation of the transmission change through the cell of DOBAMBC with the FFT analyzer.

The application of these characteristics associated with the winding and unwinding of the helicoidal structure has been proposed by one of the author. However, because of the relatively high threshold voltage and low response speed, a device utilizing this effect has not been developed. However, the threshold field is related to the spontaneous polarization P_s by the equation (1). Therefore, a much lower threshold field than in DOBAMBC can be expected in a material with large spontaneous polarization. Indeed, as shown in Figure 11, 3M2CPOOB with large spontaneous polarization showed very low threshold voltage and therefore threshold field less than 1 kV/cm. In a relatively thin cell (15 μ m) and at higher temperatures V_{th} became less than 1 V, which is smaller by more than one order of magnitude, than that of DOBAMBC. The response time was also found to be very short (less than several hundred μ sec) as shown in Figure 12.

It should also be remembered that the decay time of the transmission change related to the transition from unwound to wound states is much longer than the rise time (from wound state to unwound state) which can be partly related to the relaxation time of the Goldstone mode (winding and unwinding motions of helix in this case). Therefore, in the materials with large P_s and a high relaxation frequency it should be possible to establish an electro-optic device with low threshold voltage and the higher response speed.

There are also other ways to reduce the decay times. As already proposed by us with the name of TSM effect, ⁴ a strong light scattering can be obtained by the polarity reversal of the applied voltage. In this effect, an opaque state was obtained in a very short time less

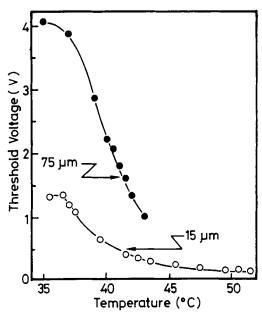


FIGURE 11 Temperature dependence of the threshold voltage in the 3M2CPOOB cell.

than several tens of μ sec. Therefore, by applying the pulse voltage of the inverse polarity to terminate a dc voltage as shown in Figure 13, much faster decay time should be obtained due to the superposition of TSM effect on the unwinding effect. For establishing high response speed, it is necessary to choose appropriate pulse height and width. It is also effective to apply a pulse trains as shown in Figure 14.

To obtain the high transmission (transparent) state in the thick cell, it is necessary to apply a dc voltage. However, we also reported that by applying voltage pulse trains with appropriate height and interval, the transparent state can be maintained.¹⁰

We would like also to propose to use the capacitance effect of the liquid crystal cell itself. If the dielectric constant of the ferroelectric liquid crystal were large enough, by opening the circuit or terminating with a high impedance after applying the dc voltage to the unwound state, the high transmission state can be maintained for long time due to the self bias effect originated from the capacitance of the element itself as shown in Figure 15.

In summary, characteristics of the electro-optic effect originating in the transition between wound and unwound states of the helicoidal

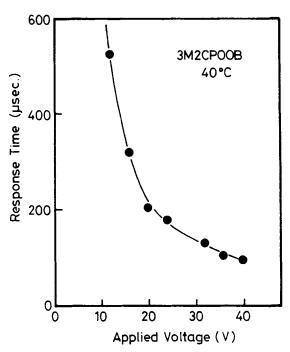


FIGURE 12 Voltage dependence of the response time in the 3M2CPOOB cell with 15 μm thickness.

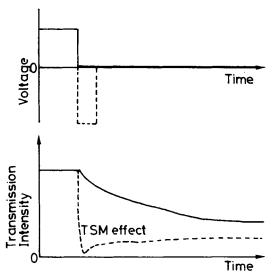


FIGURE 13 Ideal proposal of a method of reducing the decay time by the application of TSM effect.

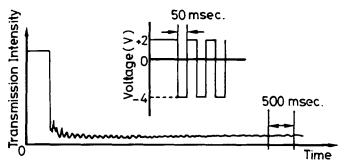


FIGURE 14 Typical wave form of the transmission intensity through the cell of 3M2CPOOB for the application of pulse trains at the end of dc voltage in order to reduce the decay time.

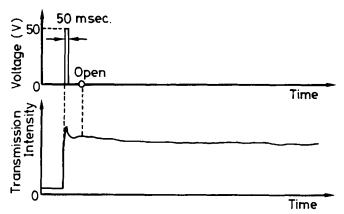


FIGURE 15 Typical wave form of the transmission intensity through the cell of 3M2CPOOB for the condition of open-circuit after the application of a pulse voltage.

structure were studied in detail. The fast response of the decrease of the transmission at low voltage application has been related to the helical pitch and explained in terms of the reorientation of the surface layer and the fluctuation of the boundary between the surface layer and the bulk helical structure. Extremely low threshold voltage and a short response time were realized with ferroelectric liquid crystals with large P_s . A method to reduce the decay time and also a self bias effect were proposed.

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