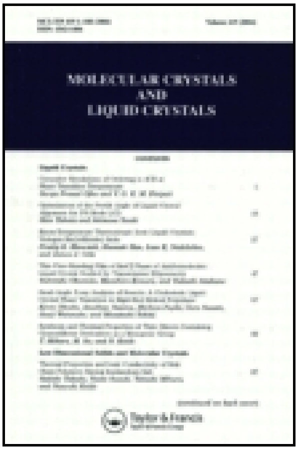

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Characteristics of Transient Light Scattering in Ferroelectric Liquid Crystals

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The rise and decay times of the transient light scattering (TSM) at the instant of the polarity reversal of applied voltage, become shorter at high voltage but also strongly depend on material and cell thickness. There exists some critical thickness at which the decay characteristics change drastically. The wave form of the transmitted light and the wave length dependence of the transmission are found to be dependent on the repetition rate of voltage reversals. The results are discussed in terms of the molecular reorientation by the applied field. However, all the characteristics can not be explained by the picture of dynamics of single molecule and the importance of the existence of the differently oriented regions which are influenced by the surface are suggested.

Keywords: ferroelectrics, liquid crystal, ferroelectric liquid crystal, electro-optic effect, light scattering, optical switching

1. INTRODUCTION

Electro-optic devices utilizing ferroelectric liquid crystals have recently great attention, because their response time is shorter than that of the nematic type devices by many orders of magnitude.

Among various electro-optic effects so far reported,[1–4] the SSFLC (surface stabilized ferroelectric liquid crystal) effect[3] has been studied most extensively. Though this effect has many advantages like a low threshold voltage, memory effect and very high response speed, it has also several disadvantages. For example, it is necessary to prepare ultra-thin cells of thickness less than several μm which is not so easy practically, especially for large area devices. Also an optical polarizer must be used, which restricts the transmittance and also the viewing angle.

We have already reported several types of electro-optic effect in ferroelectric liquid crystals which do not need any optical polarizer.[1,4,5] In the first type of effect,[1] the difference of the transmission light intensity through a homogeneously aligned cell between wound and unwound states of the helicoidal structure was utilized. We have also proposed a second type of electro-optic effect[4,5] which utilizes the change of the light transmission by the violent molecular motion at the instant of the reversal of the molecular alignment (domain switching) by the polarity reversal of the applied field. In this type of device, the high transmission state was obtained by the application of dc field or by the memory effect after the application of dc voltage, and the low transmission opaque state was obtained during the polarity reversal of the applied voltage. When it was necessary to obtain the opaque state for a long time, it could be established by repeating the polarity reversals. This type of electro-optic effect was named the TSM (Transient Scattering Mode) effect.[4,5] It has already been reported by us that the response time is very short and strongly dependent on material and various other conditions.[6]

In this paper, we will discuss the detailed characteristics of the TSM effect. The study of the TSM effect is also important from fundamental view point to understand the ferroelectric liquid crystal, especially the domain switching.

In principle, this TSM effect originates in the light scattering, therefore it should be appropriate to study the scattered light itself. However, here we will mainly discuss the decrease of the transmission by the scattering, because this transmission change is important in practical light switching element and display devices.

2. EXPERIMENTAL

Several types of ferroelectric liquid crystals have been used in this study; DOBAMBC (*p*-decyloxybenzylidene-*p*'-amino-2-methylbutylcinnamate), DOBA-1-MBC (*p*-decyloxybenzylidene-*p*'-amino-1-methylbutylcinnamate), DOBA-1-MPC (*p*-decyloxybenzylidene-*p*'-amino-1-methylpropylcinnamate) and 3M2CPOOB ((2S,3S)-3-methyl-2-chloropentanoic acid 4',4''-octyloxybiphenyl ester). Details of the preparation of these materials and also the fundamental properties of these materials have been already reported.[7-10] These ferroelectric liquid crystals were sandwiched between two nesa coated glass plates with the appropriate distance whose surfaces had been rubbed in a direction to establish a homogeneous alignment.

The change of the transmission of He-Ne laser light through the cell (without optical polarizer) by the applied voltage was monitored by a photo-diode. For the study of spectral dependence of the TSM effect, an optical absorption spectrometer Hitachi 330 was used and the sample was set in the optical path of this spectrometer. Therefore, in this case the absolute transmission level can not be compared with other cases.

3. RESULTS AND DISCUSSION

Figure 1 (a) and (b) show typical wave forms of the transmitted light through the cell for a single polarity reversal and repeated polarity reversals of applied voltage, respectively. As is evident from this figure, the transmission intensity decreases at the instant of the polarity reversal and by repeating the polarity reversal the opaque state can be established for any time interval. As is evident also from this figure, the rise time is very short. It should also be noted that the rise time is strongly dependent on the material. The rise time should be related to both the magnitude of the spontaneous polarization and the viscosity. Indeed as shown in Figure 2, the material with large

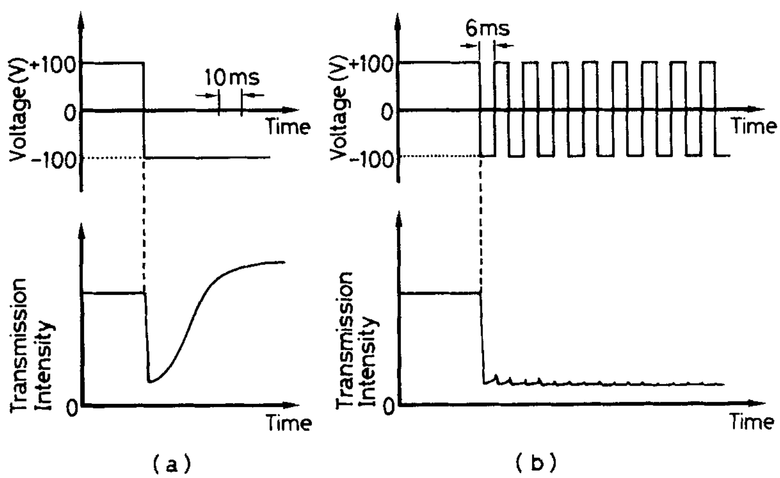

FIGURE 1 Response to a single polarity reversal (a) and a repeated polarity reversals (b) of applied voltage in the 100 μm cell of DOBAMBC.

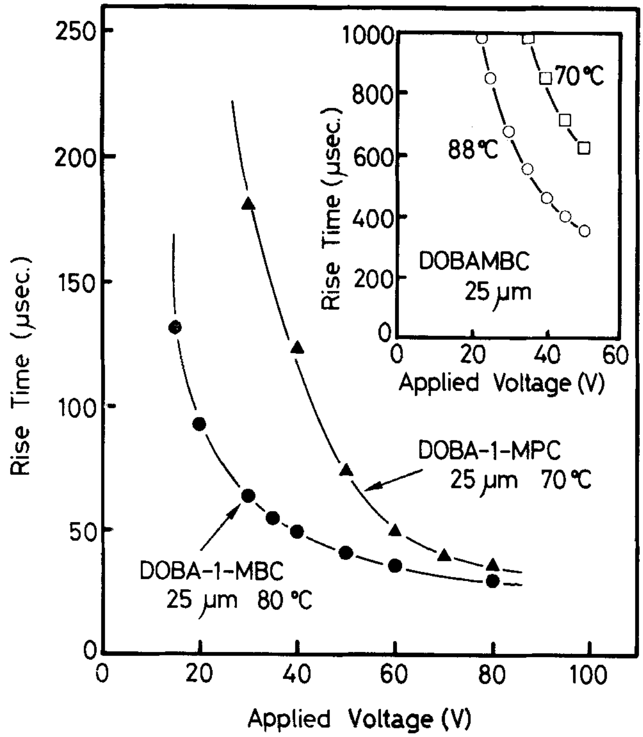

FIGURE 2 Voltage dependences of rise times of the transient light scattering in DOBA-1-MBC and DOBA-1-MPC. Inset: Those in DOBAMBC.

spontaneous polarization shows faster response. For example, DOBA-1-MBC whose spontaneous polarization P_s was about 4×10^{-8} C/cm^2 indicated about one order of magnitude shorter rise time compared with DOBAMBC of P_s around 3×10^{-9} C/cm^2. DOBA-1-MPC also indicated a short rise time.

As also shown in Figure 3, 3M2CPOOB with P_s of 3×10^{-7} C/cm^2 also showed a short rise time. The difference of viscosity between 3M2CPOOB and DOBA-1-MBC should explain why the rise time in 3M2CPOOB was not so fast compared with DOBA-1-MBC as expected from the difference of P_s between them.

In contrast to a fast rise time, the decay time in DOBAMBC was relatively long as also evident in Figure 1. The decay time should also become short at high field, because the domain switching will be accomplished in a shorter time at higher field. However, the decay time should be also related to the time of the settling of the associated violent molecular motion.

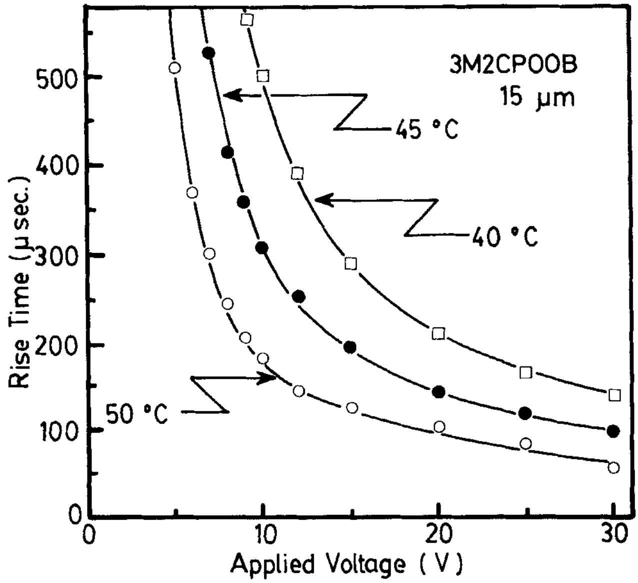

FIGURE 3 Voltage dependence of decay time of the transient light scattering in 3M2CPOOB.

It was also made clear that the decay time is strongly dependent on the molecular structure. As shown in Figure 4, DOBA-1-MBC which showed high relaxation frequency (tens of kHz) of dielectric constant indicated very short decay time. The decay time of DOBA-1-MPC with also higher relaxation frequency (several kHz) than that of DOBAMBC (several hundred Hz) showed much shorter decay time but a little longer decay time compared with DOBA-1-MBC. This also should indicate that the time of the settling of the violent molecular motion has a close relationship with the relaxation time of the materials. This interpretation seems to be reasonable because the relaxation frequency of the dielectric constant in ferroelectric liquid crystals is considered to be related with the relaxation of the winding and unwinding motions of the helicoidal structure.

As already mentioned, to establish the opaque state for a long time interval, it is necessary to repeat many polarity reversals. To obtain a stable opaque state with small ripple, it is very important to select an appropriate pulse width and repetition rate of polarity reversal. Indeed, the observed wave form of the transmission intensity was strongly dependent on the repetition rate.

Figure 5 indicates the case of DOBAMBC for various repetition

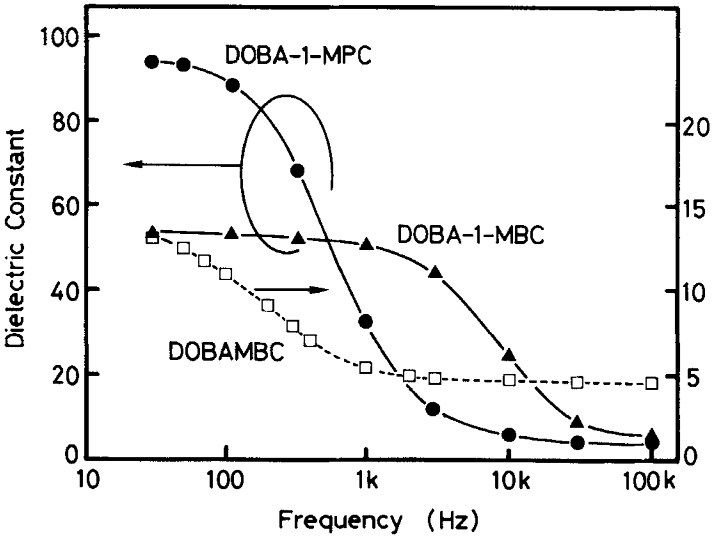

FIGURE 4 Frequency dependences of dielectric constants of DOBAMBC, DOBA-1-MBC and DOBA-1-MPC.

rates of polarity reversals. For example, in the case of small repetition rate (Figure 5(a)), a sharp pulsive peak of the transmission decrease was observed at each instant of polarity reversal. However, when the repetition frequency became high, the wave form was deformed seriously. As shown in Figure 5 (b) and (c), the scattering intensity seems not to be the same for all the instant of polarity reversals but changes alternatively. Finally at the high repetition rate, the frequency of the appearance of the strong scattering became half of the frequency of polarity reversal (Figure 5 (d)).

For example, in the case of the repetition rate of Figure 5(c) in DOBAMBC, at the instant of the first polarity reversal at T_0, the strong light scattering occurred, resulting in the remarkable decrease of the transmission. But even after the polarity of applied voltage had been completely reversed, the scattering did not disappear in a short time within the negative polarity of applied voltage, which means that the domain switching had not been completed. This suggests that the molecules have not been completely aligned at the instant of the second reversal at T_1. In that case, the direction of the dipole moment of individual molecule is different, resulting in the cancellation of dipole moments each other and decreasing of the net spontaneous polarization, just similar to the case of the helicoidal structure. Therefore, the effective force exerted by the field E, $P_s{\cdot}E$

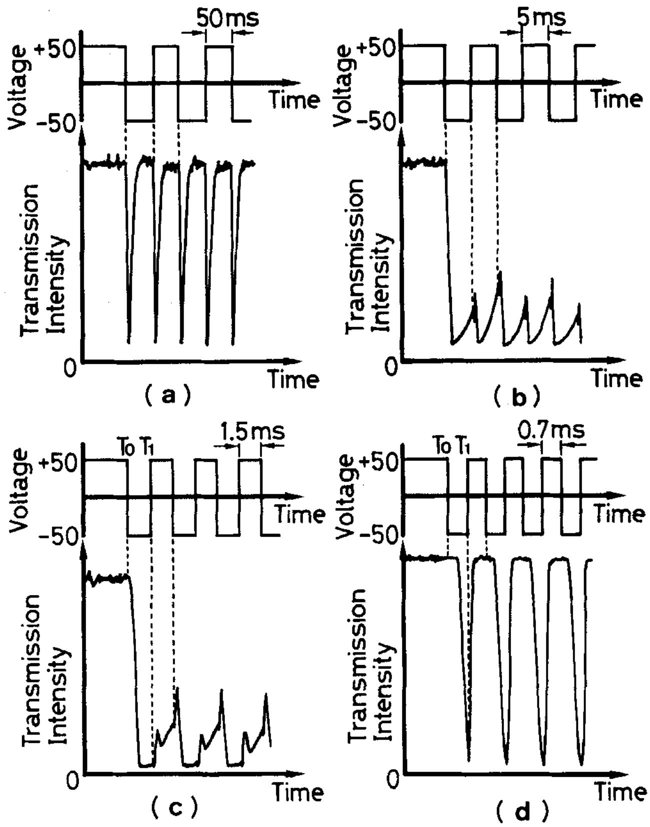

FIGURE 5 Responses to a repeated polarity reversals of applied voltage in the 50 μm cell of DOBAMBC for various repetition frequency: (a) 10 Hz, (b) 100 Hz, (c) 300 Hz and (d) 700 Hz.

torque, should be less than the case of completely aligned polarization, which should result in the less turbulence by induced molecular reorientation and smaller scattering at time T_1 as observed in Figure 5 (c).

In the case of much higher repetition rate like Figure 5 (d), at the instant of the first polarity reversal T_0, the molecular alignment will not be so much disturbed, because of the delay of molecular reorientation comparable to the pulse width, though the transmission intensity decreases to some extent. The mean polarization will not change, even though the net P_s should be less than for the completely aligned case. Therefore, at the second polarity reversal at time T_1 the molecules should align, coming back to the original direction in a very short time. By these processes the repetition frequency of strong scattering becomes half of the frequency of the polarity reversal of the applied voltage.

The characteristics of the response of the TSM effect were also confirmed to be strongly influenced by the cell thickness. As shown in Figure 6, the rise time becomes longer with decreasing cell thickness, especially at very thin cell it increases steeply. The decay time also depends strongly on thickness as shown in Figure 7. As is evident from this figure, the decay time decreases with decreasing thickness and then in a very thin cell the decay time again becomes longer. Namely there is some thickness where the shortest decay time is observed. Similar anomalous behavior was also observed in other materials. In DOBA-1-MPC, the minimum became more pronounced as shown in Figure 8.

From these results, it is clear that there exists some critical distance at which the situation of the light scattering changes remarkably. For

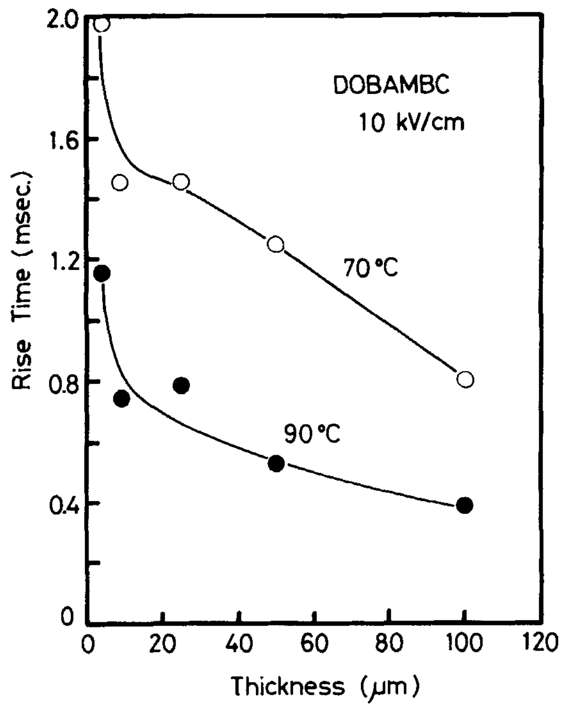

FIGURE 6 Dependence of rise time of the transient light scattering in DOBAMBC on cell thickness.

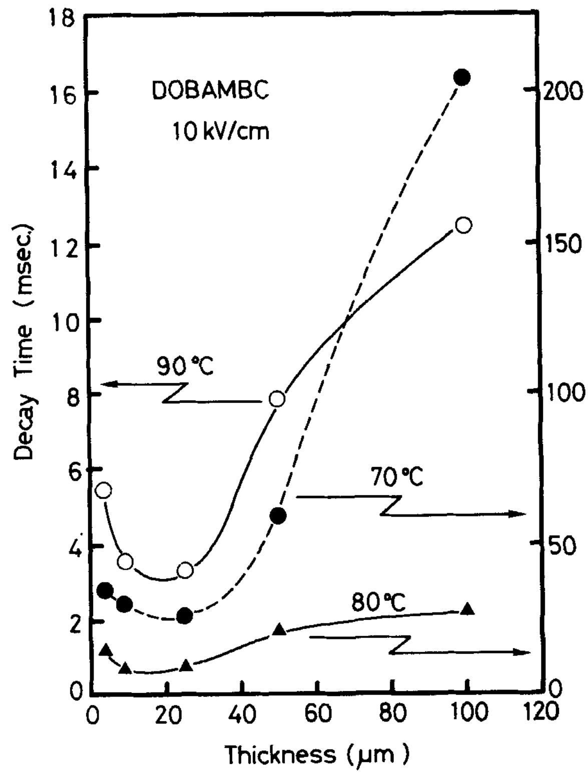

FIGURE 7 Dependence of decay time of the transient light scattering in DOBAMBC on cell thickness.

the full understanding of the scattering mechanism this effect will be very important.

In the practical case, the contrast is very important. As shown in Figure 9, the contrast in DOBAMBC is already found to be very high. However, the contrast is found to be strongly dependent on the materials. As shown in Figure 10, in 3M2CPOOB, a relatively high contrast was observed. DOBA-1-MPC, on the other hand, indicated smaller value than those of these two cases. The difference of the contrast among materials suggests that the scattering is dependent not only on the difference of the spontaneous polarization but also on other factors like the helicoidal pitch, the tilt angle, disclination, influence of the surfaces and so on. It should also be noted that there is large difference in the threshold voltage between

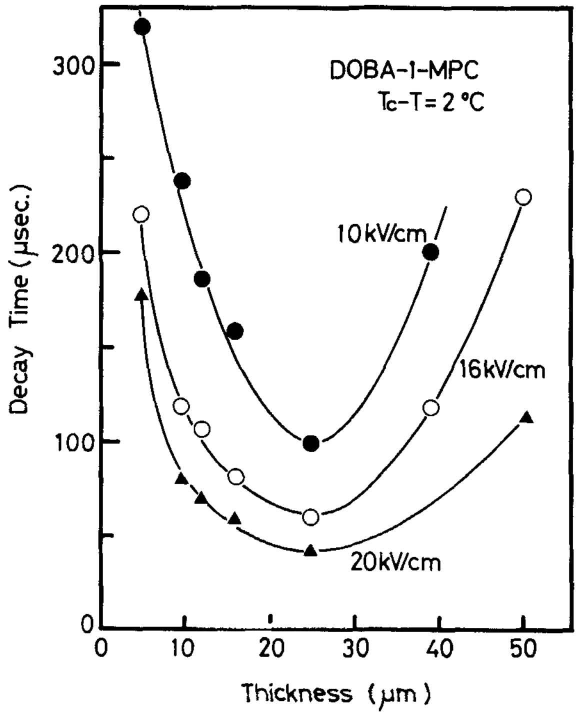

FIGURE 8 Dependence of decay time of the transient light scattering in DOBA-1-MPC on cell thickness.

DOBAMBC and 3M2CPOOB as shown in Figures 9 and 10. The threshold voltage of the TSM effect is related to that of unwinding the helicoidal structure. Therefore, 3M2CPOOB with a large spontaneous polarization has a low threshold voltage.

In the practical application as fast electro-optic devices, it is important to clarify the wavelength dependence of the transmission and switching characteristics. It was observed that the transmission was dependent on the wavelength and also this wavelength dependence was influenced remarkably by the repetition rate as shown in Figure 11. The wavelength dependence should reflect the mechanism of the scattering. From these curves, it is clear that for each wavelength there exist some repetition conditions at which the minimum transmission can be observed as shown in the inset of Figure 11. This also should be related to the size of the scattering center and also dynamics of the scattering centers. These results can not be completely explained by the simple scattering due to the reorientation of the in-

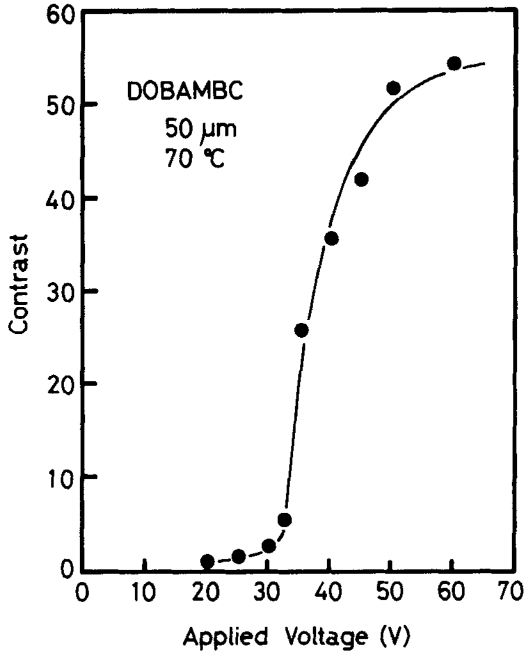

FIGURE 9 Voltage dependence of contrast of the transient light scattering at a single polarity reversal in the 50 μm cell of DOBAMBC.

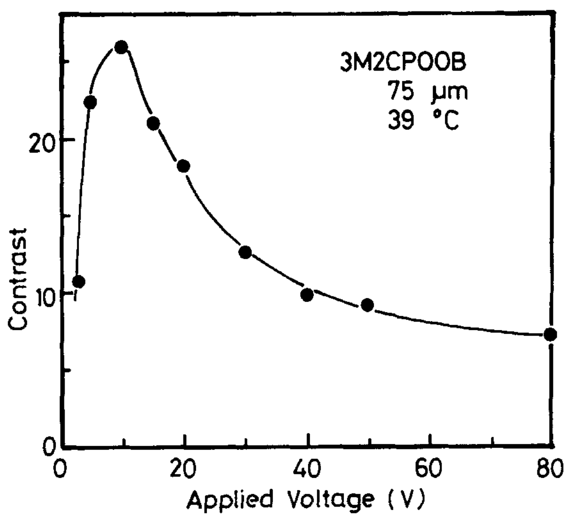

FIGURE 10 Voltage dependence of contrast of the transient light scattering at a single polarity reversal in the 75 μm cell of 3M2CPOOB.

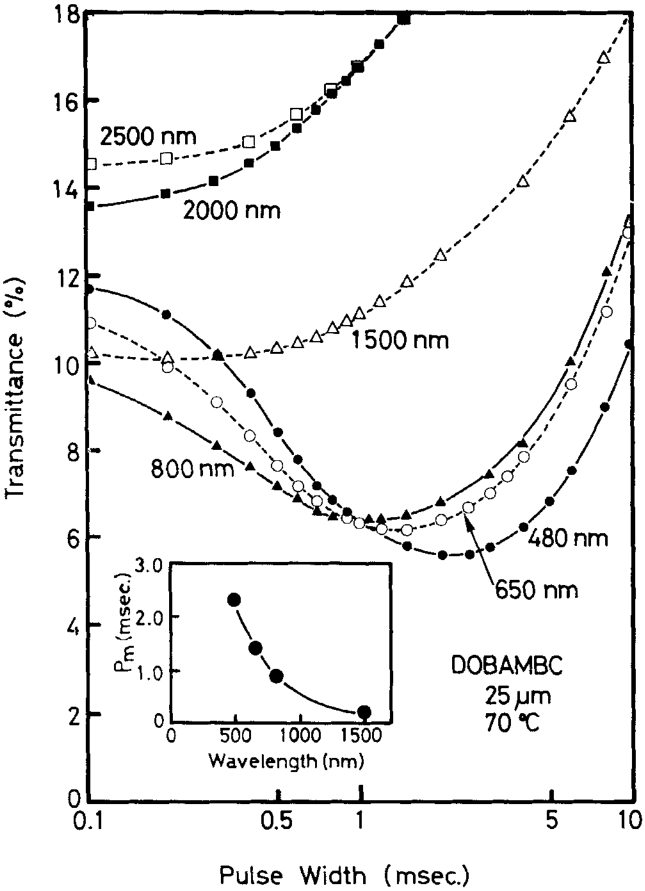

FIGURE 11 Dependence of the transmission through the cell applying a repeated polarity reversals on pulse width. Inset indicates dependence of *Pm* on wavelength. *Pm* is the pulse width at which the minimum transmission is observed for each wavelength.

dividual molecule. For a full understanding, the detailed mechanisms of the appearance of the domain reversal and its progress should be taken into consideration and is now under study. Namely, not only the dynamics of each molecule under applied voltage but also the existence of the differently oriented regions and their dynamics which should be influenced by the surfaces of wall should be taken into consideration.

SUMMARY

The results of the present experimental study on the transient light scattering at polarity reversals are summarized as follows.

1. The rise time and the decay time of the TSM effect became shorter with increasing voltage but depended strongly on the material. The materials with the large P_s shows shorter times, but they seem to also depend on the difference of viscosity.
2. The decay time of the material with a high dispersion frequency of dielectric constant indicated shorter decay time, which indicates that the decay time is partly related to the settling of the violent molecular motion.
3. The response of TSM cell by the repetition of the polarity reversals depended also on the repetition rate. The characteristic response wave form was explained in terms of dynamics of the molecular motion.
4. The rise time increased with decreasing cell thickness. On the other hand, the decay time decreased with decreasing cell thickness but it again increased below some critical thickness, which also depends on the material. This fact indicates that the scattering should change remarkably below the critical cell thickness.
5. The transmission and switching characteristics of the TSM operation were found to depend on the wavelength of light and the repetition rate of the applied voltage pulse.
6. These characteristics were tentatively explained in terms of molecular reorientation under the polarity reversal and molecular relaxation. However, it was pointed out that other factors like helicoidal pitch, tilt angle, disclination, influence of the surfaces etc. should be also included for the full understanding. The existence of differently oriented regions and their dynamics should also have a strong influence on the scattering.

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