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Voltage Effect on Cholesteric Liquid Crystals

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Abstract—dc and ac voltage effects on a mixture of cholesteric liquid crystals are studied. The wavelength of the scattering light shifts to the longer wavelength-side with the application of ac voltage at a frequency higher than 20 Hz, while it shifts to the shorter wavelength-side with the increase in dc voltage. Moreover, it is demonstrated experimentally that the modulation of the light can be observed with ac voltage at a frequency lower than 20 Hz. The modulation frequency of the light changes from f to $2f$ at 0.1 Hz with decreasing the applied frequency f . The phenomena such as the modulation factor and the phase lag are observed strongly dependent upon the frequency.

1. Introduction

This paper describes results of some measurements concerning the changes in the wavelength and the modulation of the scattering light when dc and ac voltage is applied to a mixture of cholesteric liquid crystals. Harper⁽¹⁾ reported that the wavelength of the scattering light shifted to shorter wavelength-side with the application of ac and dc voltage in a mixture of cholesteryl chloride, cholesteryl nonanoate and cholesteryl oleyl carbonate. Recently, Kahn⁽²⁾ has observed an electric-field-induced color change in a mixture of the same components as Harper's, in which the color changes from blue to red with increasing dc voltage. We have observed a new color change of the scattering light by ac field, in which the color changes from green to yellow green.

Nevertheless, dc voltages cause a color change similar to that observed by Harper. That is, dc and ac voltages have a different effect on the liquid crystals. This type of color change has not yet been reported. Furthermore, dependence of the modulation upon ac frequency has been observed. Results of the experiment of these phenomena will be described below. A model for an explanation will be proposed.

2. Sample and Cell Preparation

In the present work, cholesteryl erucate (CE) and cholesteryl 2-(2-methoxy, ethoxy)-ethyl carbonate (CMC), which were obtained commercially, are used. In this paper, measurements were performed on a 1 : 1 mixture of CE and CMC which orders cholesterically at room temperature from -2 to 37°C . It was observed with a polarizing microscope that CE alone is easy to take a conical texture, as shown in Fig. 1 and also the planar texture of CE does not scatter a visible light. However a mixture easily scattered a visible light by the addition of CMC and showed a uniform color under crossed nicols, as shown in Fig. 2. Cells examined were made by pressing the mixture of liquid crystals between transparent electrodes coated with tin oxide on a glass surface. The spacing was 25μ and 6μ using a Mylar spacer. The volume resistivity, the dielectric constant and the temperature range are shown in Table 1. The transmission and reflection spectrum of the sample are shown in Fig. 3 and Fig. 4.

TABLE 1 Characteristic Constant of Samples

Sample	CMC	CE	Mixture CE : CMC (1 : 1)
Structure	$\text{C}_{27}\text{N}_{45}\text{OCOOCH}_2\text{CH}_2\text{O}-$ $\text{CH}_2\text{CH}_2\text{CH}_3$	$\text{C}_{27}\text{H}_{45}\text{OCO}(\text{CH}_2)_{11}-$ $\text{CH} : \text{CH}(\text{CH}_2)_7\text{CH}_3$	
Temperature range	$24^{\circ}\text{C}-44^{\circ}\text{C}$	$26^{\circ}\text{C}-41^{\circ}\text{C}$	$-2^{\circ}\text{C}-37^{\circ}\text{C}$
Resistivity (ohm-cm)	10^{11}	10^{14}	$10^{12}-10^{13}$
Dielectric constant	3.4	2.5	3.0

3. Experiments and Results

To study the color change, modulation and transient phenomena of the liquid crystals, the transmission of non-polarized light through the cell was measured. The transmitting light was incident upon a photomultiplier, whose output voltage was amplified and read by a



Figure 1. A focal conic texture of CE under crossed nicols. Thickness: $25\mu \cdot 100\times$.



Figure 2. A planar texture of the mixture of CE and CMC under crossed nicols. Thickness: $25\mu \cdot 100\times$.

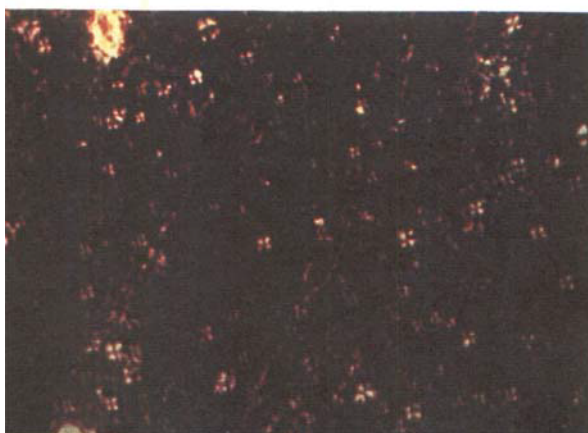


Figure 7. Breakdown of planar texture with Maltese crosses by dc field. Thickness: 25μ . Applied voltage: 40 V. Maltese crosses appear about 35 V under crossed nicols.

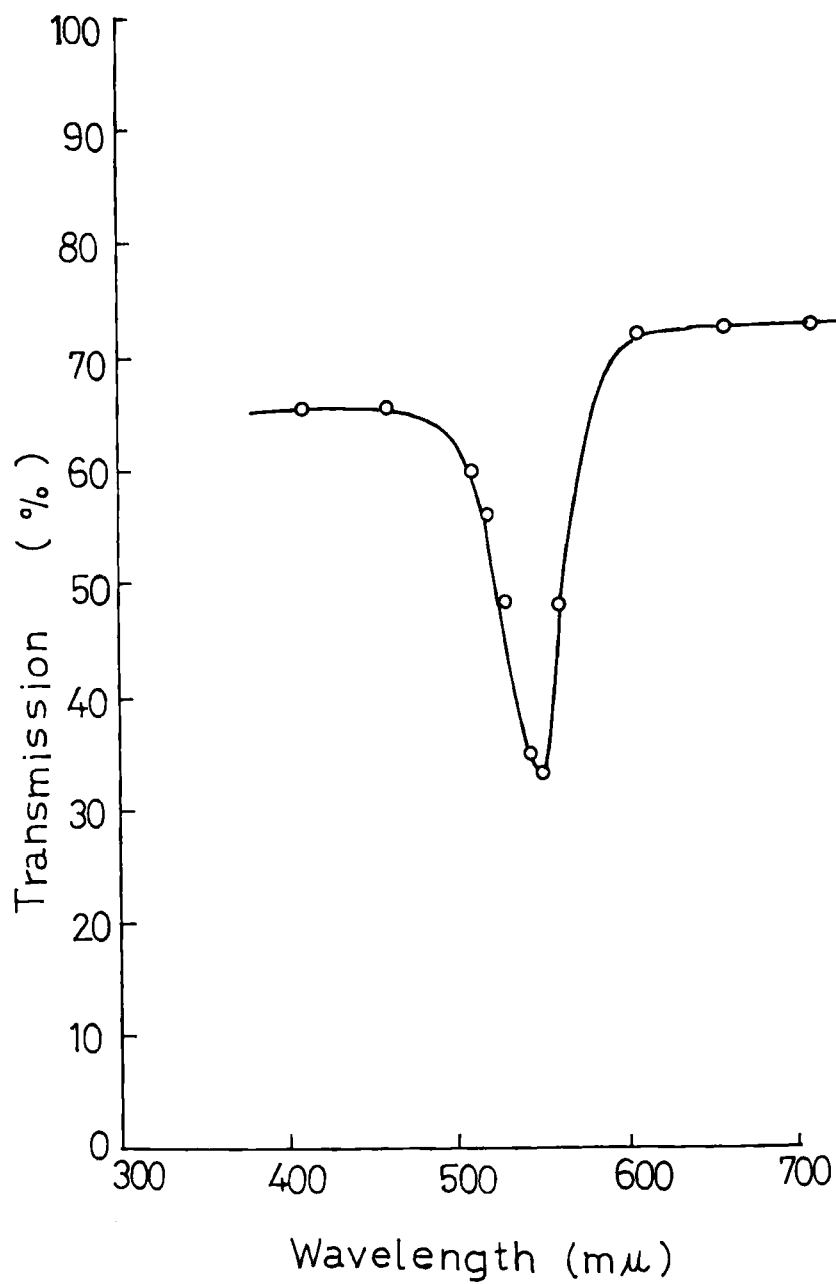


Figure 3. Transmission spectrum. CE : CMC (1 : 1).

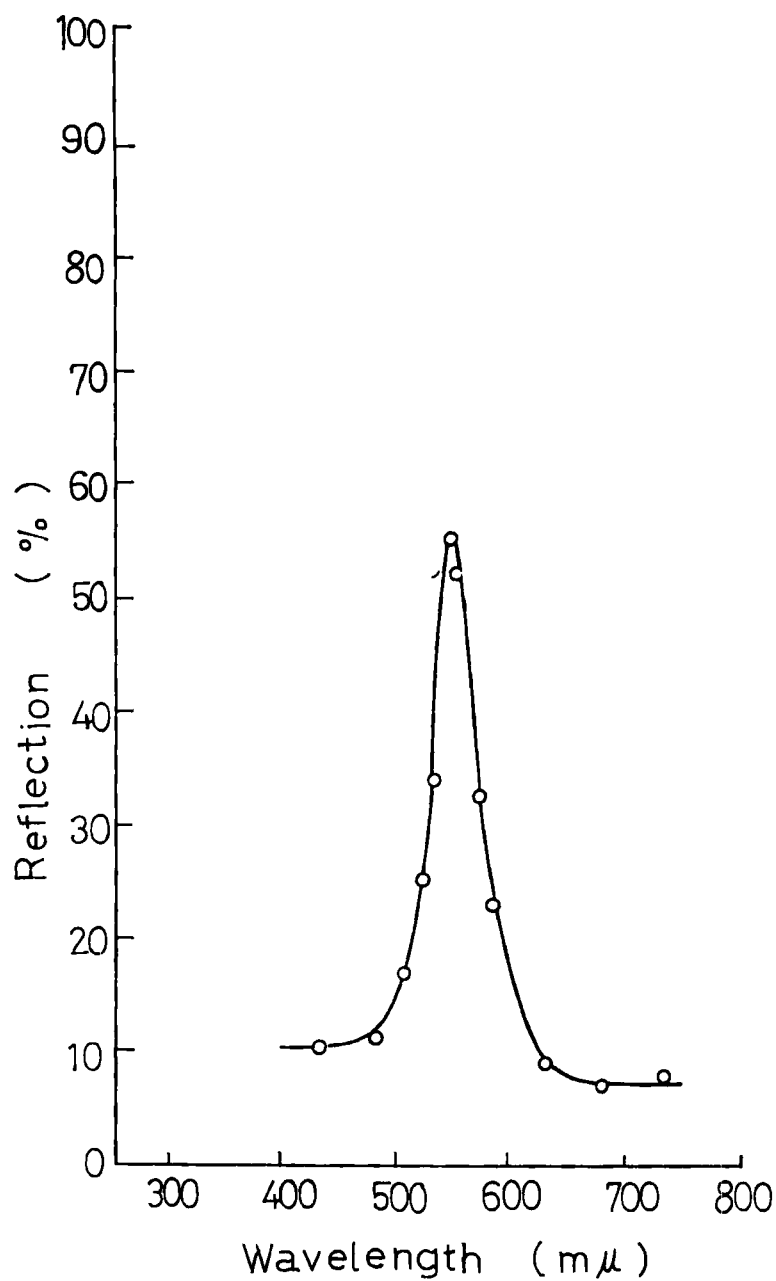


Figure 4. Reflection spectrum. CE : CMC (1 : 1).

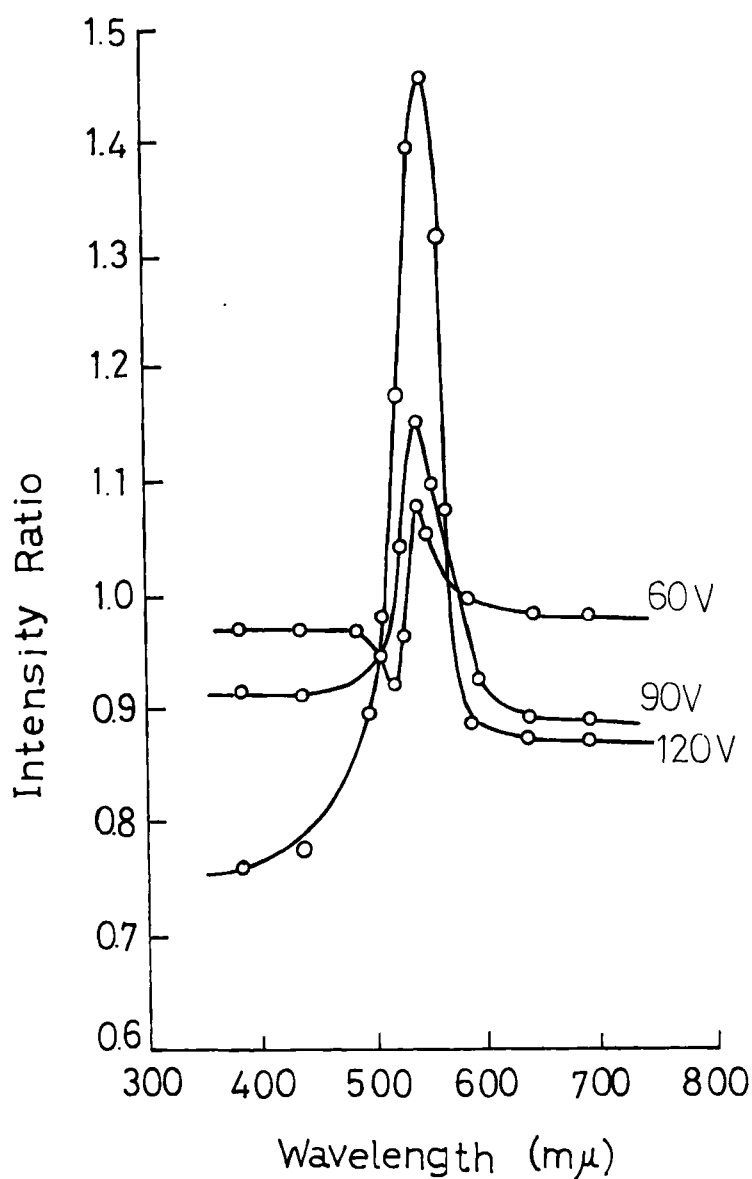


Figure 5. Spectral dependence of intensity ratio ($= T_{on}/T_{off}$) of transmitting light when dc voltage is applied. Thickness: 25μ .

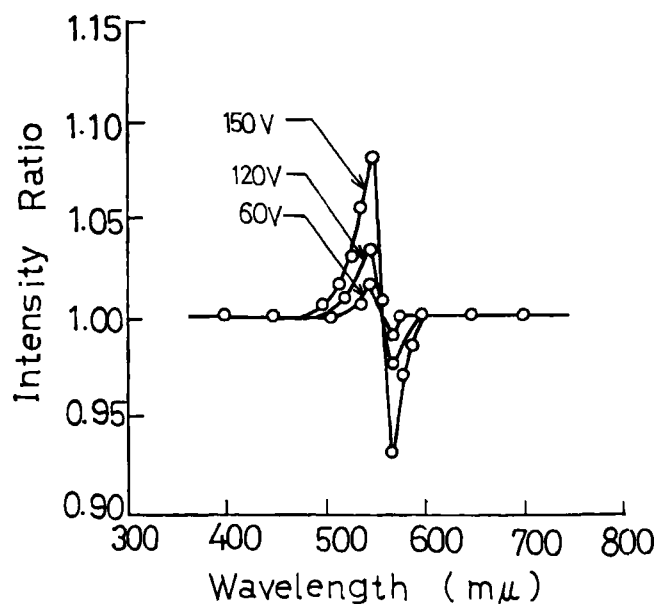


Figure 6. Spectral dependence of intensity ratio ($= T_{on}/T_{off}$) of transmitting light when ac voltage is applied. Thickness: $25\ \mu$. Frequency: 100 Hz.

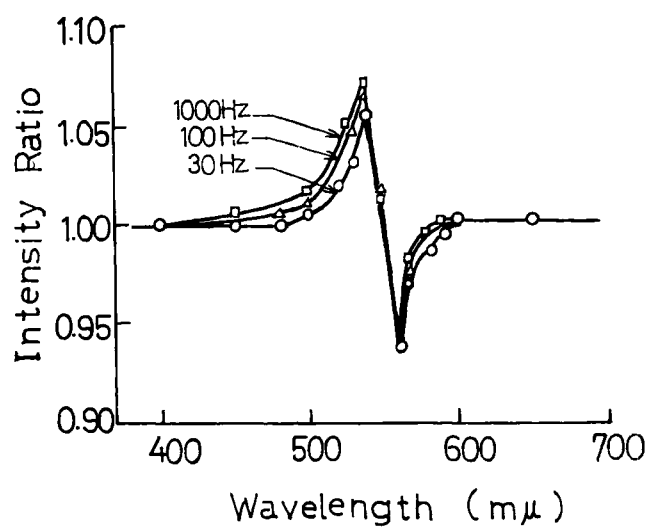


Figure 8. Spectral dependence of intensity ratio ($= T_{on}/T_{off}$) of transmitting light for different frequency. Applied voltage: 150 V. Thickness: $25\ \mu$.

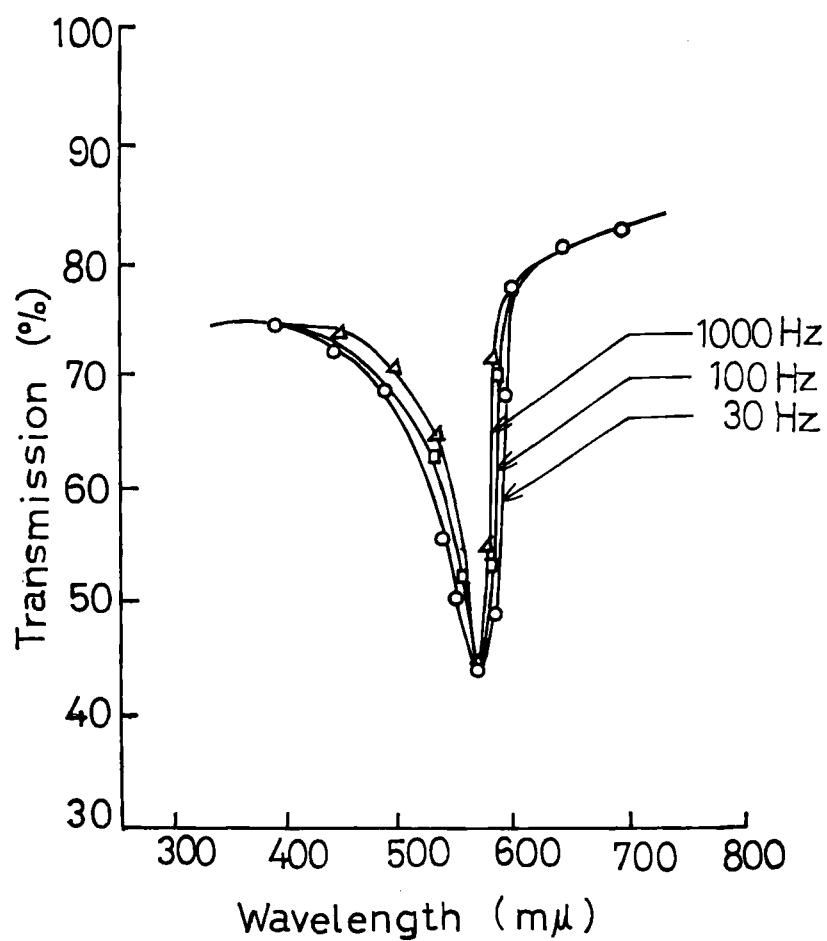


Figure 9. Spectral dependence of transmission for different frequency. Applied voltage: 150 V. Thickness: 25 μ .

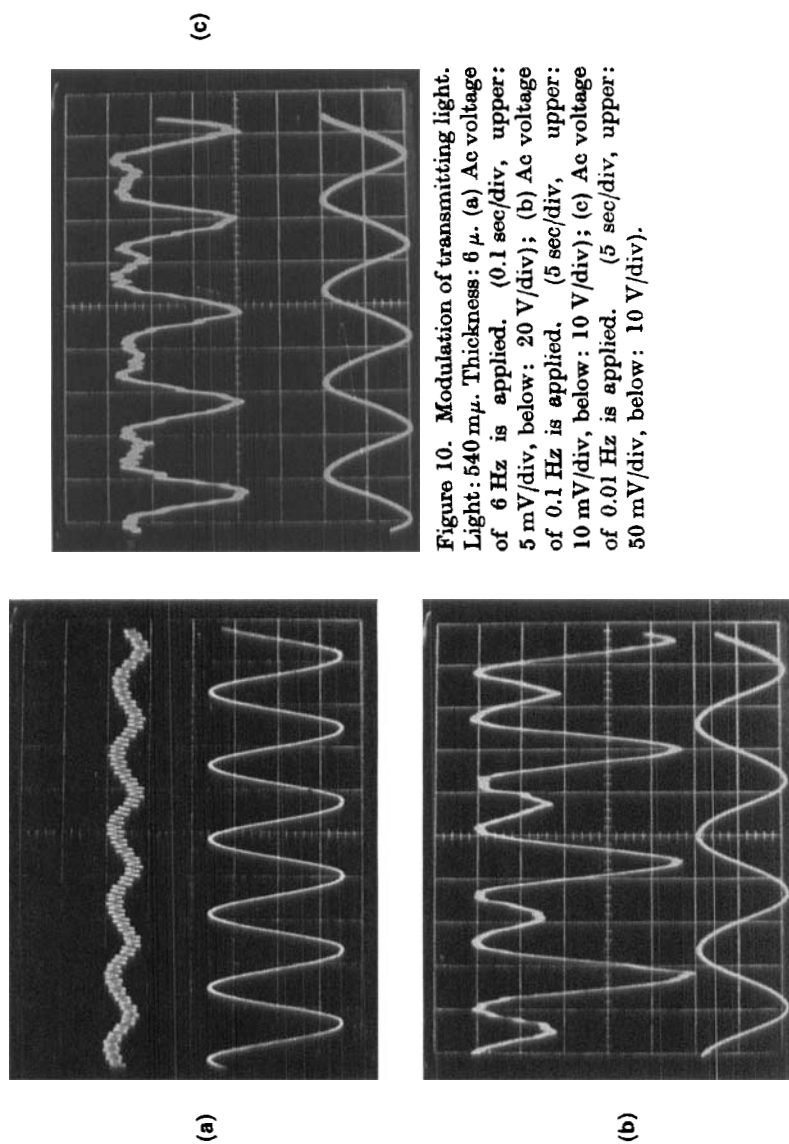


Figure 10. Modulation of transmitting light. Light: 540 m μ . Thickness: 6 μ . (a) Ac voltage of 6 Hz is applied. (0.1 sec/div, upper: 5 mV/div, below: 20 V/div); (b) Ac voltage of 0.1 Hz is applied. (5 sec/div, upper: 10 mV/div, below: 10 V/div); (c) Ac voltage of 0.01 Hz is applied. (5 sec/div, upper: 50 mV/div, below: 10 V/div).

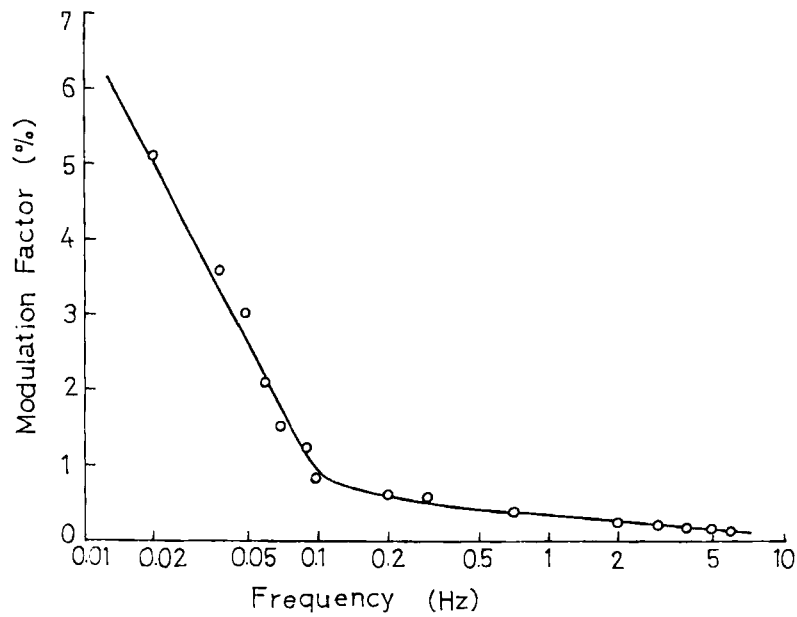


Figure 11. Modulation factor as a function of frequency. Light: 540 m μ . Voltage: 20 V. Thickness: 6 μ .

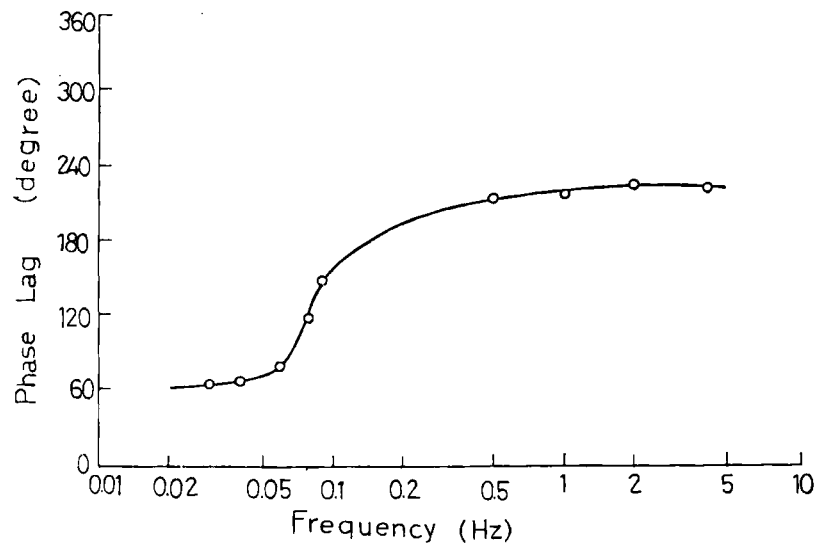


Figure 12. Phase lag as a function of frequency. Light 540 m μ . Voltage: 20 V. Thickness: 6 μ .

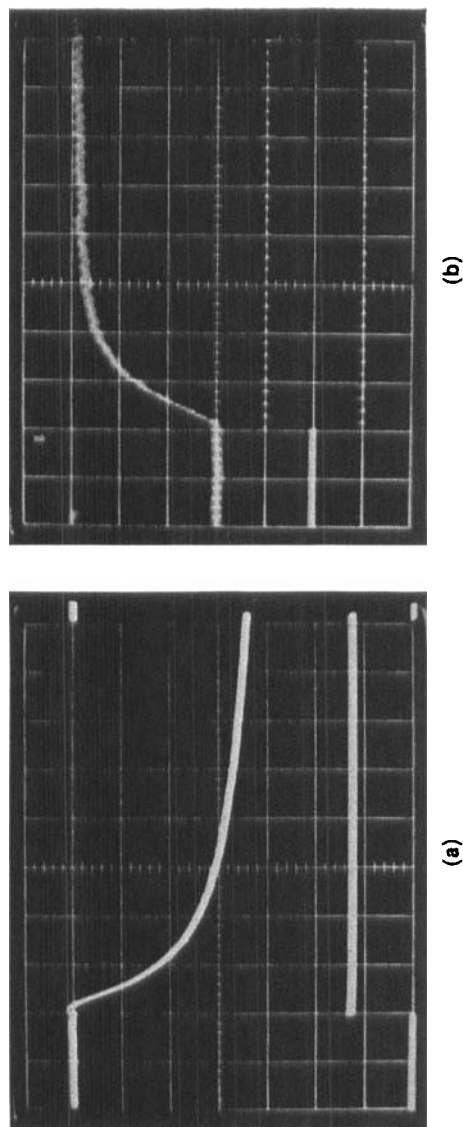


Figure 13. Transient waveform of transmitting light. Light: 540 m μ . Thickness: 25 μ . (a) De step voltage is applied. (1 sec/div, upper: 20 mV/div, below: 20 V/div); (b) Ac voltage of 20 Hz is applied. (0.2 sec/div, upper: 10 mV/div, below: 50 V/div).

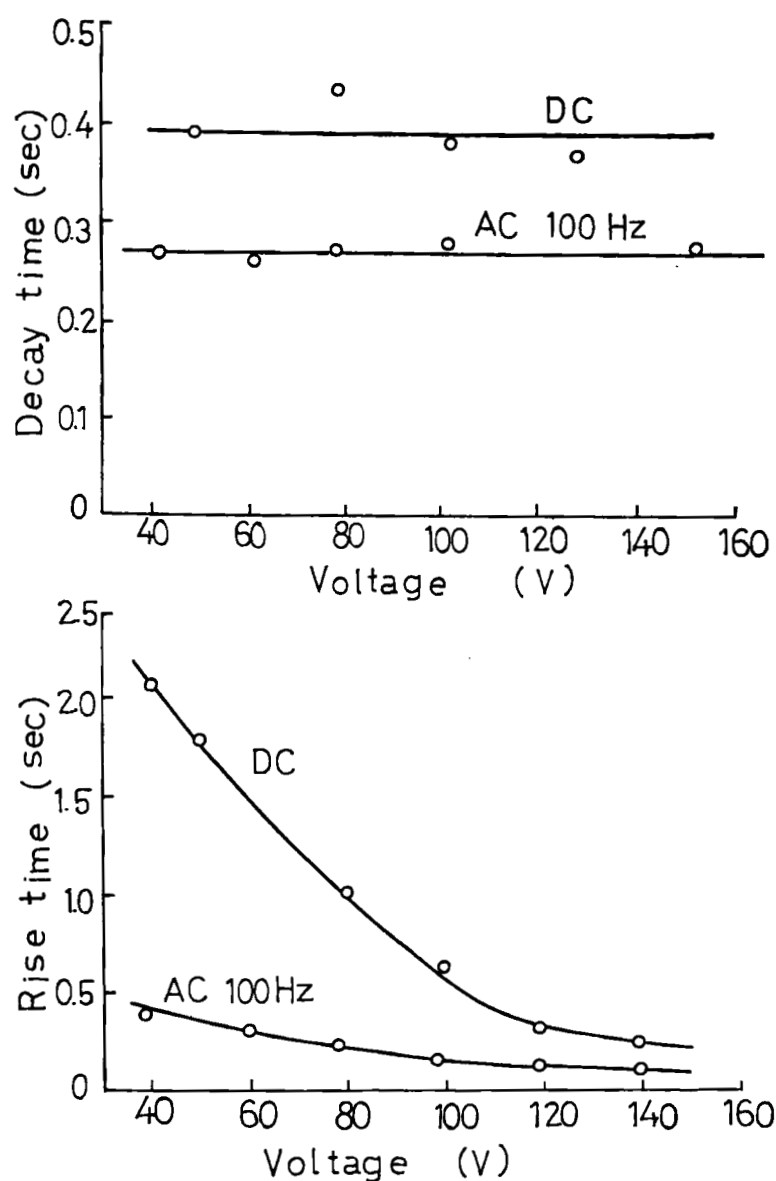


Figure. 14. Rise time and decay time as a function of applied voltage. Ac frequency: 100 Hz. Thickness: $25\ \mu$. Light: $540\ m\mu$.

synchroscope. Most measurements were performed at room temperature. Instead of measuring the scattering light, the transmitting light was measured because the maximum scattering light wavelength can be obtained from the curve of transmission. For example, compare Fig. 3 with Fig. 4.

(A) COLOR CHANGE WITH APPLICATION OF VOLTAGE

Effects of dc, ac voltage and ac frequency on the color change were investigated. Ac voltage with frequency above 20 Hz caused the shift of the scattering wavelength from green to yellow green. This also was identified with the naked eye. On the contrary, dc voltage caused the shift of the scattering wavelength from green to blue. That is, the direction of the shifts with ac and dc voltage was opposite. When dc voltage was increased up to about 2×10^4 V/cm, liquid crystals turned clear and the light scattering ceased, while the scattering was still observed for ac voltage. After removal of the dc voltage, a storage effect, in which the transmittance does not return to the initial state, was observed. Such an effect was also observed by L. Melamed and D. Rubin⁽³⁾ and F. J. Kahn⁽²⁾ with a mixture of cholesteryl chloride, cholesteryl nonanoate and cholesteryl oleyl carbonate. No storage effect was found with ac voltage. The spectral changes of the transmitting light are shown in Fig. 5 and Fig. 6, where the intensity ratio ($= T_{\text{on}}/T_{\text{off}}$) is that of transmitting light intensities with and without the application of voltage. These curves have different characteristics in the case of ac and dc voltage. Ac voltages have caused a spectral change only in a narrow region from $450 \text{ m}\mu$ to $560 \text{ m}\mu$, while dc voltages have caused it over a visible region.

The peak and dip wavelength observed in Fig. 6 did not shift with increasing ac voltage, though the change in the intensity ratio became larger. On the other hand, the dip disappeared with increasing dc voltage while the peak was increased. This means the spectral curve of transmission became flat and the scattering ceased. This is due to the breakdown of the planar texture⁽²⁾ with the appearance of Maltese crosses.⁽⁴⁾ The picture of Maltese crosses is shown in Fig. 7. Lastly, the intensity ratio depends upon frequency between 20 and 10^3 Hz for the mixture in 6μ thickness. That is, the half-width of the spectrum transmittance curve becomes nar-

rower with increasing frequency, while the curve shifts to the longer wavelength-side with increasing ac voltage. This may be significant to discuss the displacement of molecules in the cell. The results are shown in Fig. 8 and Fig. 9.

(B) MODULATION OF THE TRANSMITTING LIGHT

When an ac voltage of frequency f was applied to the liquid crystal, the transmitting light modulated was observed. The modulation frequency was $2f$ below 0.1 Hz of applied voltage, and was f between 0.1 Hz and 20 Hz. Above 20 Hz, the modulation was not observed. The entire process could be observed with the naked eye. The results are shown in Fig. 10, where the upper curve represents the waveform of the transmitting light and the lower the waveform of the applied voltage. An incident light of $540\text{ m}\mu$ was used because, at that wavelength, the change in the light intensity with applied voltage was larger. At any wavelength other than $540\text{ m}\mu$, results obtained were similar, though the transmitting light was somewhat varied.

(C) MODULATION FACTOR AND PHASE LAG

Cholesteric liquid crystals have a large viscosity.⁽⁶⁾ Thereby, a phase lag between the applied voltage and the modulation light may be expected. The light was found to be approximately modulated in the following expression

$$I = A \sin(\omega t - \delta) + (I_0 - A) \quad 0.2\pi < \omega < 40\pi$$

$$I = A \sin(2\omega t - \delta) + (I_0 - A) \quad \omega < 0.2\pi$$

where I_0 is the transmitting light intensity before the application of voltage, ω the angular frequency of the applied voltage, δ the phase lag between the applied voltage and the observed light, A the amplitude of the observed light. At $\omega < 0.2\pi$, the amplitude appears asymmetrically, that is, the bigger and smaller amplitude appears alternately. The phase lag may occur because of the viscosity which works against the displacement of molecules and may be dependent upon the velocity of the displacement. Thus, frequency dependence of the phase lag and the modulation factor $2A/I_0$ were measured. The results are shown in Fig. 11 and Fig. 12. It is found that the frequency dependence of the modulation factor and

the phase lag is different at lower and higher frequencies than 0.1 Hz respectively, at which the modulation frequency is altered. The modulation factor rapidly decreases at higher frequencies.

(D) TRANSIENT WAVEFORM WITH AC AND DC VOLTAGE

As described previously, when an ac frequency was above 20 Hz, the light could not follow alternating of voltage. The response was similar as when dc step voltage was applied, as shown in Fig. 13, though the transmitting light of $540\text{ m}\mu$ was opposite in the direction of the change. This direction depends upon wavelength of transmitting light and applied voltage. The speed of the response observed at a frequency above 20 Hz, was measured by the time required for the change in intensity to reach 63% of its maximum value. The results are as follows.

- (1) The rise time was usually longer than the decay time for both ac and dc voltage. (Fig. 14)
- (2) For dc voltage, the decay time was independent of the applied voltage, but the rise time was dependent upon the voltage.
- (3) For ac voltage, the decay time was independent of both voltage and frequency, but the rise time became a little shorter with increasing both voltage and frequency.

4. Discussion

In most of our experiments, mechanical disturbance has been given to the cell prior to measurements. Because cholesteric liquid crystals have two states of order, as shown by Adams, Haas and Wysoki.⁽⁶⁾ They have shown that cholesteric liquid crystals have two states of order when those are placed between two glass plates. They call these two states "disturbed" and "undisturbed" because the transition from the one to the other can be caused by shear force. In the disturbed state, the helical ordering axes tend to be aligned normal to the surface of glasses, while in the undisturbed state, the axes tend to be aligned parallel to the surface. Also Fergason⁽⁷⁾ assumed local domains, which are anisotropic plates stacked in a helical fashion and are distributed in the cell, as Bragg scattering sites. We will discuss color change due to dc and ac voltage, and also optical modulation in the following section.

(A) COLOR CHANGE

There is a big difference between transmitting spectral curves on the application of dc and ac (> 20 Hz) field. That is, dc field makes the spectral curve broad and wide with the increase in intensity ratio. By the application of dc field, a large part of local domains will be reverted in a sense from the disturbed state to the undisturbed state but some domains will remain in the disturbed state. In the former domains, when the angle between directions normal to the glass surface and parallel to the helical ordering axis increases, the light scattering will disappear and the transmitting will increase. The latter domains still remaining in the disturbed state may lie in a lower field than threshold field. This means that each planar structure existing in sample has not always the same threshold field for the disappearance. This is confirmed by a microscopic observation.

According to Meyer's theory⁽⁹⁾ that describes the decrease in the helical pitch due to dc field, such domains may be affected by dc field. As a result color change to shorter wavelength-side occurs. This is clearly seen in the curve for 60 V in Fig. 5. On the other hand, by the application of ac field (> 20 Hz), the transmitting spectral peak was shifted to a longer wavelength by about $20\text{ m}\mu$, with preserving the sharpness of spectral curve. This is interpreted to mean that a large part of local domains are ordered in a state rotated by a slight angle from the disturbed state. In other words, ac voltage has an ordering effect of making uniform the perturbation of orientation of the local domains. Such effect becomes large with the increase in ac frequency, as seen in Fig. 9. It seems that such ordering due to ac field makes the color change of the scattering light to longer wavelength-side.

(B) OPTICAL MODULATION

The optical modulation at frequency $2f$ observed in a lower frequency than 0.1 Hz is explained with a model based on induced dipoles. Namely, local domains or Bragg scattering sites can vibrate two times within one cycle of the applied voltage. Such vibration may be difficult with the increase in frequency.

The optical modulation at frequency f in the range between 20

and 0.1 Hz, therefore, must be caused by other effect. Such an effect can not thoroughly be understood. However, it is likely that a carrier transport causes an ordering effect. An electric current in one direction may disorder the domains near the glass wall, and a current in the opposite direction may order the disordered domains, probably due to the wall effect. This means that domains near one of the glass walls are ordered and those near other wall are disordered. Scattering light intensity may be affected, to a certain extent, by the ordering or disordering of these domains near the wall at the side of the incident light. Therefore, transmitting light also may be affected. Such an ordering due to a carrier transport will be much lower than that due to induced dipoles. This explains the slight modulation factor in this frequency range (see Fig. 11). The asymmetric waveform appearing in the $2f$ modulation is probably due to the overlap of this current-ordering effect.

Our experimental results suggest that a further increase in frequency (> 20 Hz) causes only a slight reorientation without the vibration of scattering sites. In this range color change without modulation is only observed as described earlier.

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