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Characterization of the electroclinic effect of smectic LCs in terms of the transitional dielectric constant in the S_A phase

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The electric-field dependence of the dielectric constant of some materials which show a strong electroclinic effect in the S_A phase, has been studied by measurement of the transitional dielectric constant. We suggest that the observed variation of the dielectric constant may originate from the electric field induced dielectric biaxiality; this is inherent in a material that exhibits the electroclinic effect.

1. Introduction

Ferroelectric liquid crystals (FLCs) have attracted researchers' interests for their useful features, such as bistability and fast response speed. In addition to this, the electroclinic (EC) effect of smectic liquid crystals is interesting for the application of fast electrooptic devices. The EC effect has been observed either in a S_A phase containing chiral centres [1] or in a S_C^* phase [2].

Our group has developed an instrument which is capable of measuring the temporal variation of the dielectric constant of surface stabilized ferroelectric liquid crystal (SSFLC) cell [3]. This instrument is suitable for observing the dynamics of FLC molecular motion.

In this paper, the transitional dielectric constant of some substances which show a strong EC effect in the S_A phase is studied by measurement of the transitional dielectric constant (TDC) [3]. It is found from these investigations that the variation of the transitional dielectric constant is related to a large electroclinic coefficient in these substances, and the reciprocal of the dielectric constant, measured at 40 kHz, varies linearly with the applied electric field. These phenomena may be explained by considering the appearance of the biaxiality in the S_A phase.

2. Experimental

The substances used were 4-(1-methylheptyloxycarbonyl)phenyl 4'-octyloxybiphenyl-4-carboxylate (MHPOBC) and 764E (supplied by Merck Ltd.), which show a strong EC effect in the S_A phase [2, 4]. For comparison, we used another material ZLI-3654 (Merck Ltd.), which does not exhibit the EC effect. The phase sequences of these materials are as follows [5]:

MHPOBC

$$C \xrightarrow[84°C]{} S^*_{C_A} \xrightarrow[118{\cdot}3°C]{} S^*_{C_\gamma} \xrightarrow[119{\cdot}2°C]{} S^*_C \xrightarrow[120{\cdot}7°C]{} S^*_{C_\alpha} \xrightarrow[122°C]{} S_A \xrightarrow[148°C]{} I.$$

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764E

$$S_C^* \underset{26°C}{\text{———}} S_A \underset{72°C}{\text{———}} N^* \underset{92°C}{\text{———}} I.$$

ZLI 3654

$$S_C^* \underset{62°C}{\text{———}} S_A \underset{76°C}{\text{———}} N^* \underset{86°C}{\text{———}} I.$$

The sample FLC cell was of the ordinary sandwich-type and consisted of two ITO-coated glass plates whose inner surfaces were coated with pre-rubbed polyimide films. The LC molecules were aligned homogeneously, so that the smectic layers were perpendicular to the glass substrate in the S_A phase. The thickness of the LC medium was 1·0 μm.

To characterize these FLC cells, the electrooptic (EO) response was measured, using crossed polarizer geometry, by applying a triangular voltage to the sample cells. The voltage ranged from 10 $V_{p\text{-}p}$ to 100 $V_{p\text{-}p}$. The transitional dielectric constants of these materials were measured with the instrument reported in the previous paper [3]. The measuring frequency for the dielectric constant was 40 kHz for detecting the soft mode response.

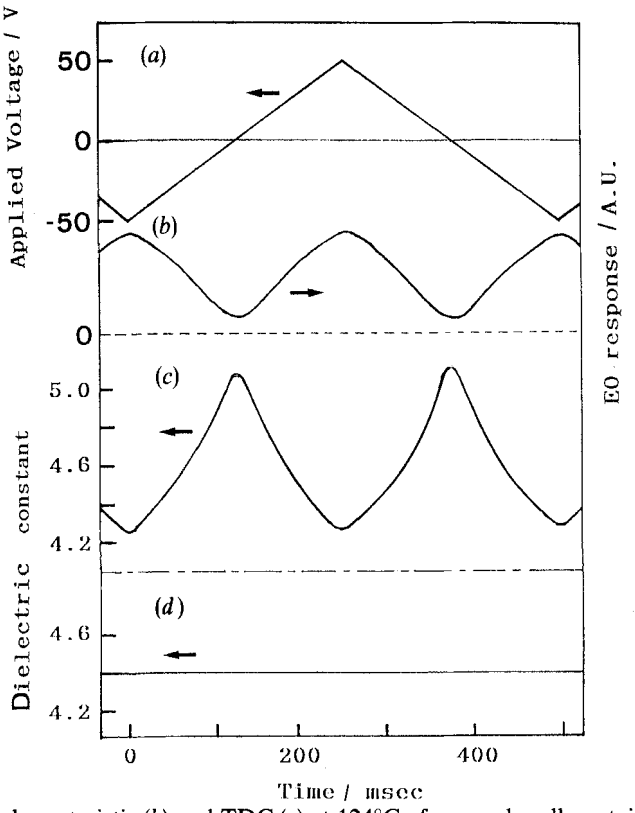

Figure 1. EO characteristic (*b*) and TDC (*c*) at 124°C of a sample cell containing MHPOBC, and the TDC (*d*) at 64°C of a ZLI 3654 sample. The TDC response also varies with the EO response; this result indicates that the apparent dielectric constant varies with the tilt angle.

3. Results and discussion

Figure 1 represents the results of the EO characteristics (see figure 1 (*b*)) and the TDC (see figure 1 (*c*)) of the sample of MHPOBC in the S_A phase. For comparison the TDC of a conventional FLC ZLI 3654 (see figure 1 (*d*)) is also shown. The applied field was 100 $V_{p\text{-}p}$ and the temperature was just above the phase transition point. The MHPOBC sample shows a remarkable electroclinic effect in the EO response (see figure 1 (*b*)). Note that the TDC response also varies with the EO response; this result indicates that the dielectric constant normal to the glass substrate varies with the tilt angle. On the other hand, the TDC response of the ZLI 3654 sample (see figure 1 (*d*)) is almost flat, indicating no change in molecular configuration under the electric field. Such dielectric behaviours are also observed for the 764E sample, as shown in figure 2. These results suggest that LC materials which exhibit the EC show significant dielectric response.

The temperature dependence of the reciprocal of the dielectric constant is shown in figure 3. The sample is MHPOBC and the applied voltage $E = 0$ V (*a*) and $E = 50$ V (*b*). These behaviours are in agreement with theoretical predictions (Curie–Weiss Law) [1, 6] near the phase transition point. It is to be noted that, when the electric field was applied (trace (*b*)), the critical transition point shifts to higher temperature.

In order to understand the variation of the dielectric constant, which depends on the tilt angle, figure 4 shows the temperature dependence of the reciprocals of the electroclinic coefficient K (closed circle) and the dielectric constant variation (closed triangle), $\delta\varepsilon$, for the MHPOBC sample. $\delta\varepsilon$ is defined as

$$\delta\varepsilon = \varepsilon_{E=0\,\mathrm{V}} - \varepsilon_{E=50\,\mathrm{V}}, \tag{1}$$

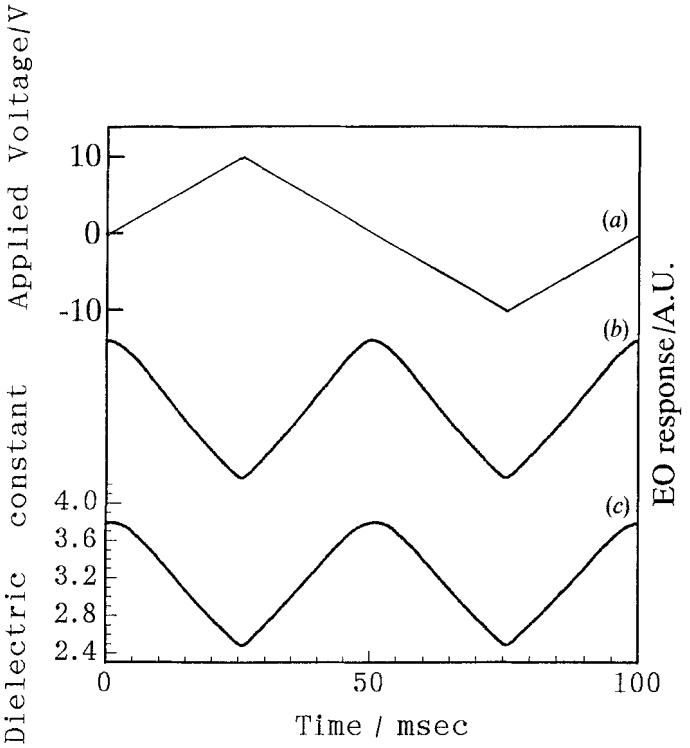

Figure 2. EO characteristics (*b*) and TDC in the S_A phase (*c*) at 30°C of a sample cell containing 764E. The LC materials which exhibit the EC effect show significant dielectric response.

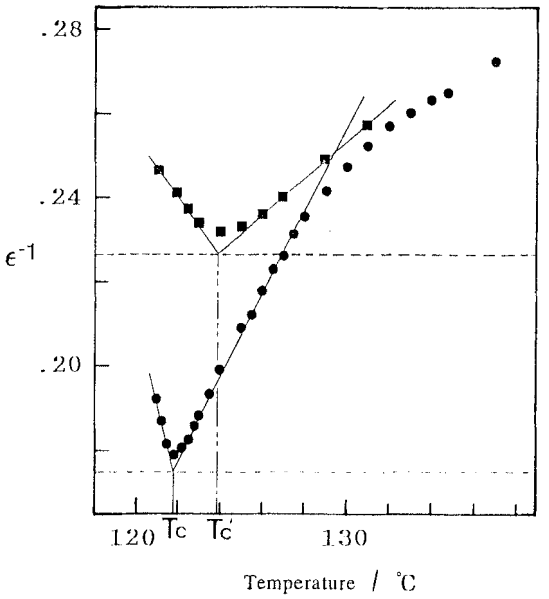

Figure 3. Temperature dependence of the reciprocal dielectric constant at the external applied voltages (*a*) $E=0$ V (●) and (*b*) $E=50$ V (■). These behaviours are in agreement with theoretical predictions.

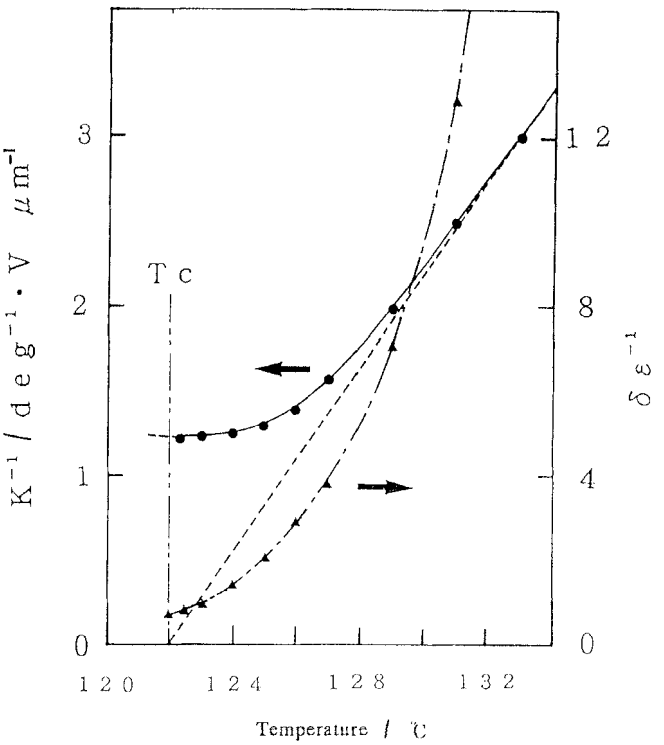

Figure 4. Temperature dependence of the reciprocals of the electroclinic coefficient K (●) and the dielectric constant difference $\delta\varepsilon$ (▲). The electroclinic coefficient K (the tilt angle θ) and the dielectric constant $\delta\varepsilon$ may have a strong correlation.

where $\varepsilon_{E=50\,V}$ and $\varepsilon_{E=0\,V}$ are the dielectric constants with an applied voltage of 50 V and without applied voltage, respectively. The reciprocal of the EC coefficient (denoted as K^{-1}) is estimated at the applied voltage of 50 V. Theoretical consideration of the Landau-type free energy leads to the following relation [1, 2, 6],

$$\theta = K \cdot E, \tag{2}$$

where θ is the tilt angle of the LC molecules, and K is written by

$$K = \frac{\chi_c}{a(T - T_c)} \propto \frac{1}{(T - T_c)}. \tag{3}$$

Thus K^{-1} is proportional to $(T - T_c)$. As shown in figure 4, however, the experimental value of K^{-1} (●) gradually deviates from the theoretical curve (dashed line) on approaching the critical temperature T_c. This is because the electric field dependence of the tilt angle θ tends to saturate near the critical point. For such a large EC effect, field induced layer distortion from a uniform to something like a chevron structure may be caused. On the other hand, the temperature dependence of $\delta\varepsilon^{-1}$ has a similar tendency to that of K^{-1}; this similar behaviour indicates that the electroclinic coefficient K (the tilt angle θ) and the dielectric constant $\delta\varepsilon$ have a strong correlation.

The possibility of layer distortion was studied for the S_C^* phase instead of the S_A phase for the 764E sample, as shown in figure 5. Some anomalous behaviour is found such that the TDC response and optical intensity vary before and after the molecular switching based on the Goldstone mode. However conventional FLC substances (for

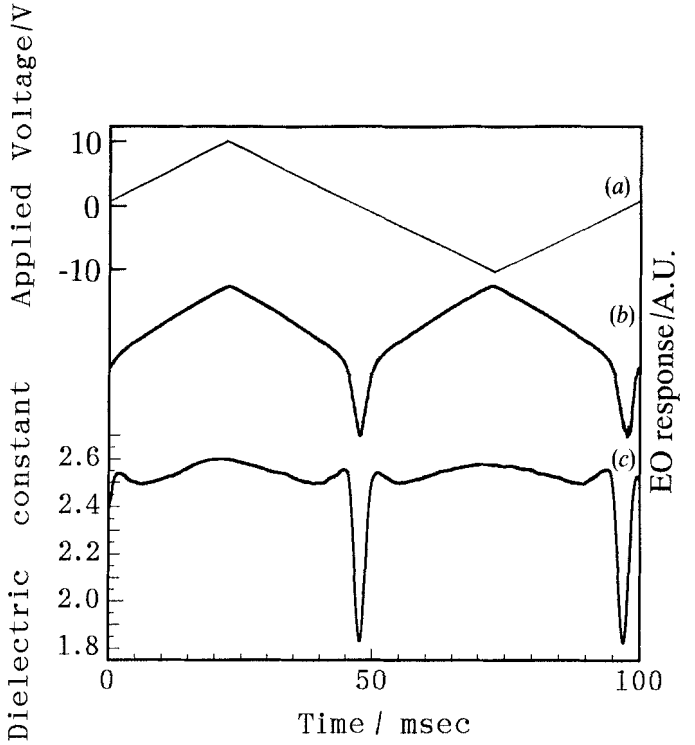

Figure 5. EO characteristic (*b*) and TDC (*c*) at 20°C of a 764E sample in the S_C^* phase. The variation of the dielectric constant may be caused by layer distortion.

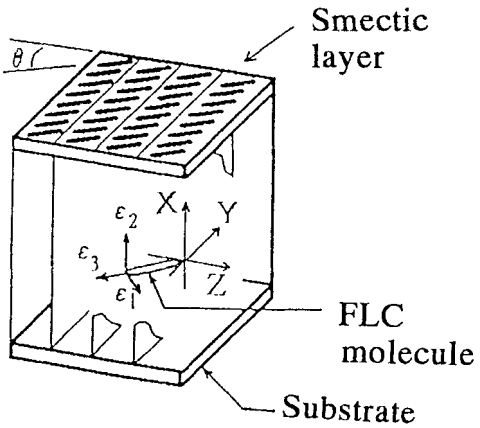

Figure 6. Schematic description of the principal dielectric constants ε_1, ε_2, and ε_3.

example ZLI 3654) did not exhibit these dielectric variations, but showed only a sharp peak [3]. This results suggests that the variation of the dielectric constant is also caused by layer distortion. Therefore we should consider the dielectric biaxiality for the S_A phase when we estimate the dielectric constant, similarly to the case of the conventional S_C^* phase [7, 8].

In order to understand the TDC behaviour, it is useful to consider the dynamical molecular motion in a S_A phase. It is known that a LC molecule in the S_A phase rotates around its molecular long axis. Therefore the S_A phase is uniaxial. When an electric field is applied parallel to the S_A layer, the electric dipole of the molecule tends to align parallel to the electric field. As a result of this interaction, the molecular rotation along the molecular long axis is hindered and the molecule is tilted. Thus the system can be considered as being biaxial.

Now we can define the principal dielectric constant for the S_A phase just as for a S_C^* phase [7, 8] in such a way that ε_3 is parallel to the director **n**, ε_2 is parallel to the applied field, and ε_1 is normal to these axes, as shown in figure. 6. Then ε_2 and ε_1 have the same values under the no-bias condition. On the other hand, applying the electric field to the sample results in different values for ε_2 and ε_1 due to the appearance of the biaxiality, as mentioned previously. Thus the variation of the dielectric constant is also caused by layer distortion, since the observed dielectric constant decreases with the layer tilt angle which may be caused by the strong EC effect. Therefore the measured value of $\delta\varepsilon$ varies with the molecular tilt angle. We finally point out that lowering the sample temperature enhances the electric field-induced molecular tilt, leading to larger TDC in the S_A phase.

4. Conclusion

The transitional electric field dependence of the dielectric constants of smectic LC substances which show a strong electroclinic effect in the S_A phase have been studied by measurement of the transitional dielectric constant. The observed dependence of the dielectric constant on the applied voltage is understood by considering the electric field-induced biaxiality of the LC medium.

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