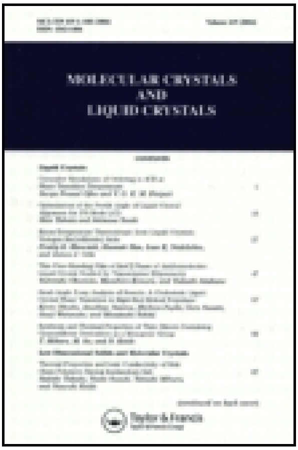


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# Investigation of Interfacial Liquid Crystal Orientation by Reflection Ellipsometry

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# Investigation of Interfacial Liquid Crystal Orientation by Reflection Ellipsometry

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The optical techniques, the Brewster angle reflection method, the dye doped reflection ellipsometry and the total reflection ellipsometry, are proposed to investigate the interfacial Liquid Crystal (LC) director orientation. These methods are based on the analysis of the optical polarization of the reflected light from LC cells.

From the experimental and theoretical results, it was found that these methods are useful to study the interfacial LC director orientation. Comparison of these methods with one another clarifies the features and the advantages of each one.

*Keywords:* ellipsometry; interface; Brewster angle; total reflection

## 1 Introduction

The alignment treatment of the substrates for controlling the liquid crystal (LC) orientation is one of the most important factors which decide the performance of the LC device. There are many alignment techniques but their mechanisms are not yet fully understood. Clarifying the behavior of the LC director at the interface between an alignment film and the LC is not only of physical interest but also of technological importance for display devices. To clear the above, the LC orientation at the interface between an alignment film and the LC has been investigated by many methods, for example, the second harmonic generation[1], the attenuated total reflection method[2], the high field method[3], etc.

Recently, the Brewster angle reflection (BAR) method, the dye doped reflection ellipsometry (DDRE)[4], [5] and the total reflection ellipsometry (TRE)[6] were proposed to investigate the interfacial LC orientation. These ellipsometric methods allow us to analyze the interfacial LC director orientation with a high sensitivity.

In this paper, it is shown that these methods are useful to investigate the interfacial LC director orientation, and these are compared with one another to clarify their features and advantages.

# 2 Principle

## 2.1 Reflected Light from a LC Cell

The conventional LC cell consists of glass substrates, indium tin oxide (ITO) films, alignment films and an LC layer. For simplicity, we assume here that the LC cell is composed of glass substrates and an LC layer. When the light beam is obliquely incident on the LC cell, the reflected light beam consists of followings; the reflection from the interface between air and a front substrate, that from the interface between a front substrate and an LC layer, that from the interface between an LC layer and a rear substrate, and that from the interface between a rear substrate and air. In addition to the above components, the multiple-beam reflection and the multiple-beam interference at an LC layer, whose thickness is uniform in the dimension of the wavelength of the incident beam, can not be neglected. The thickness of a glass substrate is much lager than the wavelength of the incident beam and not uniform in the dimension of the wavelength so that the multiple-beam interference at the glass substrate is ignored.

The reflected light from the interface between a substrate and the LC contains the information about the interfacial LC director orientation. When the external field is applied to the LC cell, the polarized state of the reflected light beam from the interface between a substrate and the LC is changed as the interfacial LC director is reoriented. This fact indicates that the analysis of the reflected light from the interface between a substrate and the LC allows us to investigate the interfacial LC director orientation.

For the investigation of the interfacial LC director orientation with a high sensitivity, it is preferable to detect only the light component which is reflected from the interface between a front substrate and the LC. The light component reflected from the interface between air and a front substrate decreases the sensitivity of the analysis because its amplitude is higher than those of the other reflected light components. The reflected light from the interface between the LC and a rear substrate includes not only the information about the interfacial LC director orientation but also that about the LC director orientation in the bulk, which causes the complication of the analysis. In practice, the existences of an ITO film and an alignment film on the substrate can not be neglected because the multiple-beam reflection and the multiple-beam interference at their films contribute the detected light, seriously[7].

The BAR method, the DDRE and the TRE are proposed to observe the reflected light component from a front substrate.

## 2.2 The Brewster Angle Reflection Method

The schematic diagram of the reflection based on the BAR method is shown in Fig. 1.

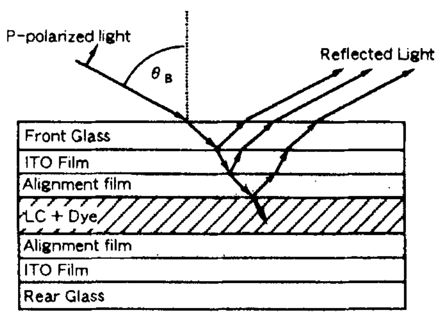



Figure 1: Schematic diagram of the reflection based on the BAR method.

In Fig. 1, a LC cell consists of glass substrates, ITO films, alignment films and a dye-doped LC . When the wavelength of the incident beam is in the absorption band of the dye, the traveling beam is completely absorbed by the bulk of LC without passing through a LC cell. Then, the reflected light beam is that from a front substrate but not from a rear substrate in a sandwiched LC cell. The reflected light from the interface between an alignment film and LC depends on the interfacial LC director orientation so that the analysis of it allows us to study the interfacial LC director orientation.

When the incident angle from air into a glass substrate is the Brewster angle of the glass ($\theta_B$; derived by Snell's law), P-polarized light transmit through the air-glass interface without the reflection. Then, the detected light components consist of those reflected from the glass-ITO film interface, the ITO film-alignment film interface, and the alignment film-LC interface.

By the BAR method, the analysis of the detected light intensity allows us to investigate the interfacial LC director orientation.



## 2.3 The Dye-Doped Reflection Ellipsometry

The DDRE requires the dye doped LC cell as well as the BAR method. When the light beam whose wavelength is in the absorption band of the dye is incident on the dye doped LC cell, the reflected light components consists of followings; the reflection from the air-front glass plate interface, that from the front glass plate-ITO film interface, that from the ITO film-alignment film interface, that from the alignment film-LC interface and the multiple reflection and the multiple interference at films. By the DDRE, the interfacial LC director reorientation is observed as the change in the polarization states when the external field is applied to the LC cell. The details of the DDRE are shown in ref. [4],[5].

## 2.4 The Total Reflection Ellipsometry

In a LC cell, if the total reflection occurs at the interface between an ITO film and an alignment film, or that between an alignment film and the LC, the reflected light contains the information about the interfacial LC director orientation. In practice, an LC cell with a glass substrate whose refractive index is higher than that of the LC is required. When the incident angle from a glass into the ITO film is larger than the critical angle $\theta_c$ ($\theta_c$ depends on refractive indices of the LC and the LC orientation near the interface), the light beam is totally reflected at the interface between the ITO and alignment films or that between the alignment film and the LC.

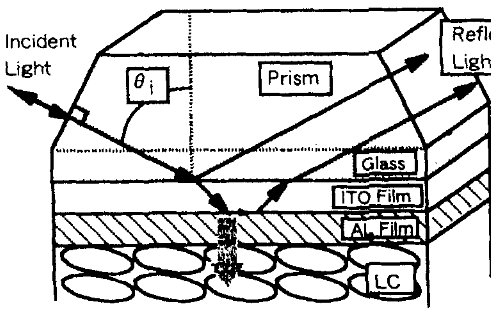



Figure 2: Schematic diagram of total reflection from a LC cell.

The schematic diagram of the total reflection from an LC cell is shown in Fig. 2. The prism has the same refractive index as the glass substrate and is attached to it with a matching oil which also has the same refractive index. The prism allows the light component reflected from the interface between air and the glass plate to be eliminated so that the analysis of the LC director orientation can be made more sensitive. In Fig. 2, it is assumed that the refractive index of the ITO film is higher than those of the other materials in the LC cell and the refractive index of the alignment film is nearly the same as that of the LC. Then, the total reflection occurs at the interface between an ITO film and an alignment film. The light which is transmitted through the ITO film penetrates into the alignment film and the LC layer with amplitude attenuation so that the reflected light contains information on the interfacial LC director orientation. Thus, the TRE method is applicable to the investigation of the interfacial LC director orientation.

## 3 Experiments

For the BAR method and the DDRE, the LC used was 4-pentyl-4'-cyanobiphenyl (5CB) with a small amount (5 wt.%) of dye dopant which was *p*-dimethylaminoazobenzene (DAB). DAB is a dichroic dye and has a strong absorption band at wavelength $\lambda$ = 419 nm. The glass substrates used were coated with a solution of polyvinyl-alcohol (PVA) on the ITO film. Their surfaces were rubbed unidirectionally after baking them. The homogeneously aligned cells were constructed with different gaps, which were enough thickness to absorb the travelling light in the bulk of the LC.

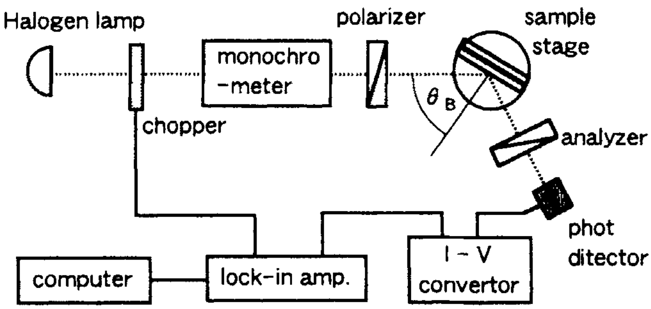



Figure 3: Experimental setup for BAR method.

Fig. 3 shows the experimental setup for the BAR method. By the monochromator, the light beam from the halogen lamp was monochromatized (419 nm). A polarizer and an analyzer were set to transmit only P-polarized light. The P-polarized light was incident on the LC cell at the Brewster angle (56.8°) and whose incident plane was parallel to the rubbing direction of the front substrate. For the BAR method, the experimental setup was very simple as shown in Fig. 3. To observe the azimuthal orientation of the interfacial LC director, the LC cell used was a 45° TN cell and fixed to the sample stage which was rotated about the surface normal of the substrate.

For the DDRE, the ellipsometer used was M-150 (JASCO Corporation) whose system consisted of a polarization-modulated spectroscopy with a photoelastic modulator. The polarization-modulated light was incident on the LC cell at the angle of 45° and whose incident plane was parallel to the rubbing direction of the front substrate.

The theoretical analysis can be performed by means of the $4 \times 4$ matrix method which automatically takes the multiple reflection and the multiple interference at the films and a LC layer into account. The LC orientation was calculated by applying Frank's elastic continuum theory[8]. The optical parameters used in the theoretical analysis are shown in Table 1.

Table 1: Parameter used in simulation.

| Material | Refractive index | Extinction coeff. |
|---|---|---|
| 5CB (600 nm) | 1.726 (∥) | 0.0 (∥) |
| | 1.537 (⊥) | 0.0 (⊥) |
| 5CB+Dye (419 nm) | 1.795 (∥) | 0.0064 (∥) |
| | 1.562 (⊥) | 0.0183 (⊥) |
| Glass (for BAR, DDRE) | 1.53 | 0.0 |
| (for TRE) | 1.80 | 0.0 |

# 4 Results and Discussion

## 4.1 Results for BAR method

By the BAR method, the reflected light intensity from the front substrate of the 45° TN cell whose substrates were rubbed bare glasses was measured. Turning over the cell, that from the rear substrate of the cell was also measured. For normalization, the measured reflectance was divided by the maximum reflectance. The polar plots of the normalized


(a) Rubbing direction 0 [deg]   (b) Rubbing direction 45 [deg]

Figure 4: Polar plots of normalized reflectance as a function of rotation angle about the substrate normal.

reflectance as a function of the rotation angle about the substrate normal are shown in Fig. 4 (a) and (b). In Fig. 4, (a) and (b) are the results of the front substrate and the rear substrate, respectively. The closed circles and the solid line are the experimental results and the theoretical ones, respectively. The rubbing direction on the rear substrate was 45°, whereas that on the front substrate was 0°. For theoretical analysis, the LC orientational distribution was assumed that the interfacial director aligned parallel to the rubbing direction. The experimental result was in good agreement with the theoretical one and they have the peak of the reflectance along the rubbing direction. When the incident plane is parallel to the interfacial LC director, the refractive index difference between the glass and LC is the largest so that the amplitude reflectance at the interface between the glass and LC was maximum. As mentioned before, the detected light is that from the interface between a glass and an LC which depends on the refractive index difference between a glass and an interfacial LC director. From the experimental result and theoretical one, the BAR method is useful to investigate the interfacial LC director orientation.

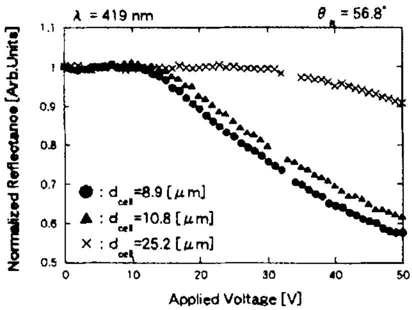
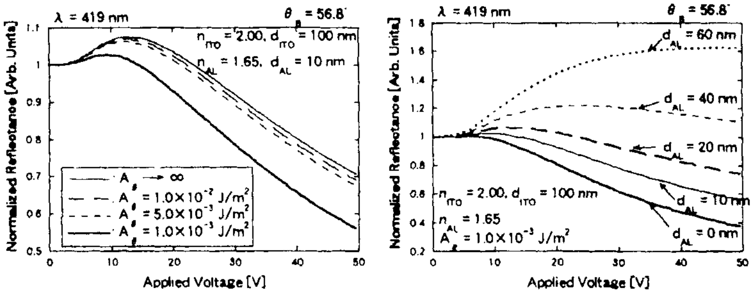



Figure 5: The normalized reflectance in static response to the applied voltage for different cell thicknesses.

Applied voltage dependence of the normalized reflectance of homogeneously aligned cells with different cell thicknesses, 8.9, 10.8, 25.2$\mu$m, were shown in Fig. 5. In Fig. 5, the reflectance versus applied voltage reflects the interfacial LC director reorientation with a sensitivity. From Fig. 5, it was found that the interfacial LC director is anchored more strongly to the substrate as the gap of cell is thicker. The reason is that the orientational deformation of LC directors is smaller across the LC layer as the gap of cell is thicker.

The theoretical results of the normalized reflectance versus applied voltage for different anchoring strengths and different alignment film thicknesses are shown in Fig. 6 (a) and (b), respectively. It is found

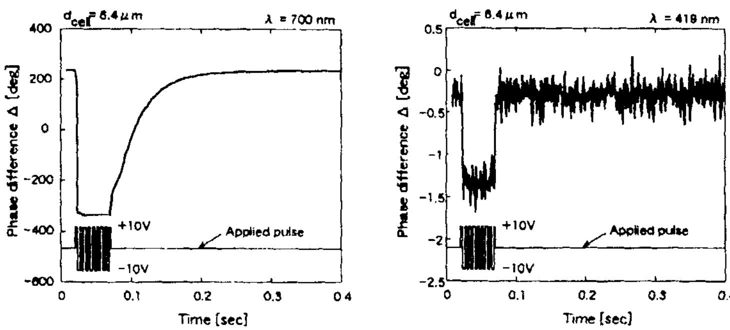


(a) dependence on anchoring strength
(b) dependence on alignment film thickness

Figure 6: Normalized reflectance versus applied voltage.

that the normalized reflectance versus applied voltage depends on the anchoring strength. In Fig. 6 (b), the influences of the multiple beam reflection and the multiple beam interference at the alignment film on the normalized reflectance are serious. The influences of the ITO film is also serious. If refractive indies and thicknesses of an ITO film and an alignment film are measured, comparison of the experimental results with the theoretical ones allows us to estimate the anchoring strength.

## 4.2 Results for DDRE

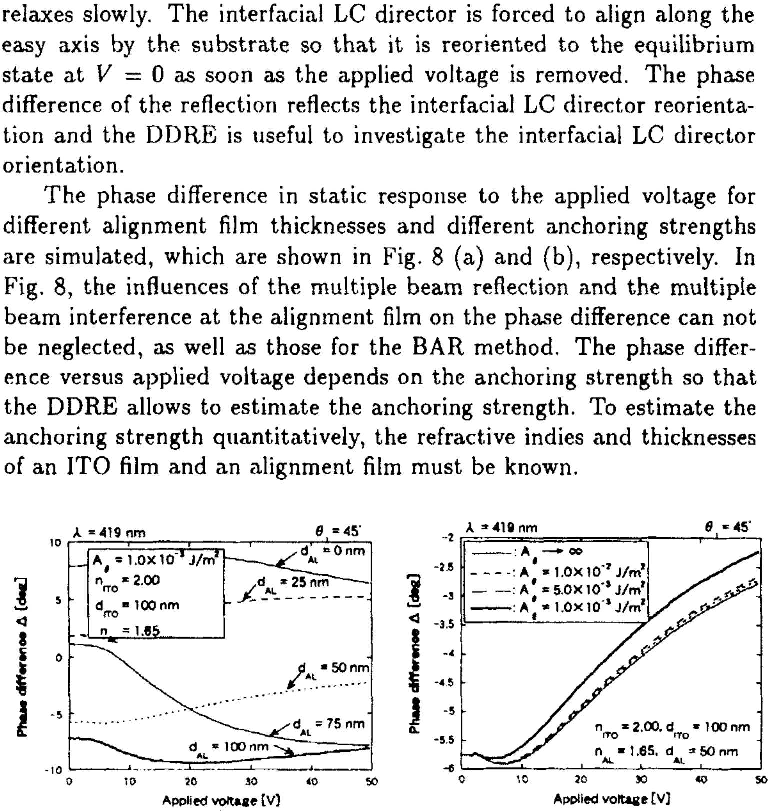


(a) Phase difference of transmission
(b) Phase difference of reflection

Figure 7: The phase difference in dynamic response to applied voltage.

By the transmission ellipsometry and the DDRE, the dynamic response of the phase difference to applied voltages are shown in Fig. 7 (a) and (b), respectively. The applied voltage was a pulse of 10 V at the frequency 1 kHz. For the decay process, the phase difference of the reflection changes to the initial state (the phase difference at $V = 0$) as soon as the applied voltage is removed whereas that of the transmission relaxes slowly. The interfacial LC director is forced to align along the easy axis by the substrate so that it is reoriented to the equilibrium state at $V = 0$ as soon as the applied voltage is removed. The phase difference of the reflection reflects the interfacial LC director reorientation and the DDRE is useful to investigate the interfacial LC director orientation.

The phase difference in static response to the applied voltage for different alignment film thicknesses and different anchoring strengths are simulated, which are shown in Fig. 8 (a) and (b), respectively. In Fig. 8, the influences of the multiple beam reflection and the multiple beam interference at the alignment film on the phase difference can not be neglected, as well as those for the BAR method. The phase difference versus applied voltage depends on the anchoring strength so that the DDRE allows to estimate the anchoring strength. To estimate the anchoring strength quantitatively, the refractive indies and thicknesses of an ITO film and an alignment film must be known.




(a) dependence on thickness of alignment film (b) dependence on anchoring strength

Figure 8: Phase difference in static response to the applied voltage.

## 4.3 Results for TRE

The phase difference in static response to the applied voltage for different thicknesses of the alignment films and different thicknesses of the

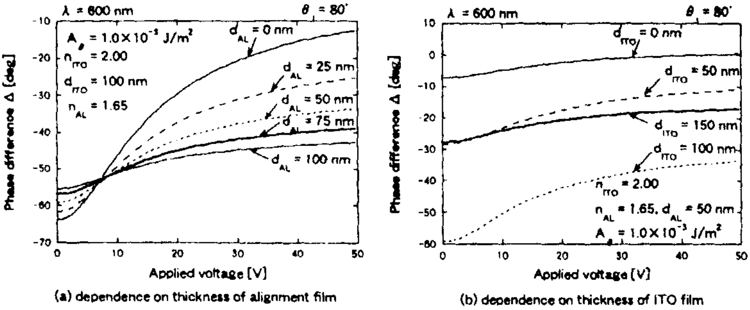



Figure 9: Phase difference in static response to the applied voltage.

ITO films are simulated, and are shown in Fig. 9 (a) and (b), respectively. It was assumed that the cell was filled with 5CB whose thickness was 6.0$\mu$m. It is seen that the phase difference versus applied voltage depends monotonically on the thickness of the alignment film. The reason is that total reflection occurs at the interface between the ITO and alignment films such that the amplitude of light which penetrates into the LC layer is weaker with a thicker alignment film.

In Fig. 9(b), the reflected light includes multiple reflection and multiple interference at the ITO film so that the dependence of the phase difference versus applied voltage on $d_{\text{ITO}}$ is not monotonic.

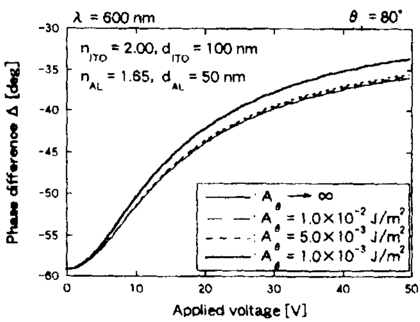



Figure 10: Phase difference in static response to the applied voltage for different values of the anchoring strengths.

The phase difference in static response to the applied voltage for different values of the anchoring strengths are simulated, and are shown in Fig. 10. Even under high electric fields, the interfacial LC director is fixed to the substrate when the polar anchoring strength is $\infty$. Then, the phase difference versus applied voltage is always constant if the reflected light includes only information about the interfacial LC director orientation. However, the phase difference versus applied voltage for strong anchoring strength varies, which means that the reflected light includes information not only on the interfacial LC director orientation but also on the bulk LC director orientation in the vicinity of the interface. Although the TRE method can not be used to observe the LC director orientation only at the interface between the alignment film and an LC, it is applicable to the estimation of the polar anchoring strength of the LC cell with high sensitivity.

### 4.4 Discussion on their Advantages

The experimental setup based on the BAR method is simpler than those of the DDRE and the TRE. As for the experimental sensitivity, the DDRE and the TRE using the polarization modulated spectroellipsometry are superior to the BAR method so that these ellipsometries allows to analyze the anchoring strength, quantitatively.

The BAR method and the DDRE allow us to observe the reflected light which contains the information about the LC director orientation at the front interface between an alignment film and the LC but requires the use of a LC cell which is doped with dichroic dye in order to eliminate the reflected light from the rear interface. Although the influence of the dichroic dye on nematic LC orientation is probably negligible, that on ferroelectric smectic LC orientation is not. On the other hand, the TRE method does not necessitate the use of the dye doped LC cell. As the penetration depth of the evanescent light in total reflection is of the order of the wavelength, the TRE allows us to use a thin cell, for example, the surface stabilized ferroelectric LC cells.

## 5 Conclusion

The BAR method, the DDRE and the TRE are proposed to investigate the interfacial LC director orientation. These methods are based on the idea that the reflected light from the interface between the substrate and the LC reflects the interfacial LC director orientation. From the experimental and theoretical results, it was found that these methods are useful to investigate the interfacial LC director orientation. By taking the multiple beam reflection and the multiple beam interference at the films into account, these methods allow to estimate the polar anchoring strength quantitatively even if the polar anchoring is rather strong.

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