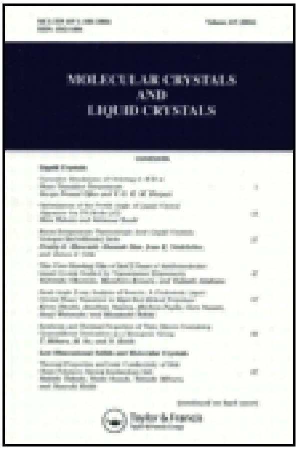

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The Relationship between Pretilt Angle of Liquid Crystal and Optical Anisotropy of Alignment Film

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We investigated the relationship between the pretilt of liquid crystal and the optical anisotropy of three kinds of polyimide films (PI-A, PI-B and PI-C) rubbed under various conditions. The optical anisotropy of the rubbed films was detected by reflection ellipsometry, and was characterized according to anisotropic dielectric constants, the tilt of the principal dielectric axis and the thickness of the molecular oriented layer. The optical anisotropy of the rubbed PI-A and PI-C films was largely dependent on the rubbing condition. A monotonic decrease in pretilt was observed as the difference between the ordinary and extraordinary dielectric constants of the molecular oriented layers. In the case of PI-B, the optical anisotropy is changed very little when rubbing conditions were changed. The correlation between the optical anisotropy in PI-B and pretilt was unclear. It was concluded that the influence of molecular orientation on pretilt is strongly dependent on the polyimide type.

Keywords: polyimide; rubbing; pretilt; reflection ellipsometry; alignment film

Controlling the pretilt angle of liquid crystal is essential in fabricating high-quality liquid crystal displays (LCDs). It is widely held that liquid crystal aligns as a result of intermolecular interaction to polyimide molecules[1, 2, 3], and that the pretilt angle is largely influenced by the polyimide molecular alignment induced by rubbing. Molecular orientation of polyimide usually generates optical anisotropy in rubbed polyimide films. As a result, we studied the relationship between the pretilt angle and optical anisotropy of polyimide films rubbed under various conditions to understand the influence of polyimide molecular orientation.

All samples were prepared on glass substrates (Corning 7059). Three kinds of polyiamic-acid (PI-A, PI-B, and PI-C: Nissan Chemical, Fig.1) were coated on the substrates by using a spinning coater. Imidization was performed by heating for 60 min at 250°C after pre-baking for 15 min at 80°C. Rubbing conditions varied according to tuning rotation speed(f), circumference contact length (L), transferring speed (v) of the rubbing roller, and the cumulative number of rubs (N). The molecular orientation of the rubbed polyimide film was characterized by using an ellipsometer (MARY-102: 1mW He-Ne laser). The incident angle was 50°. The pretilt angle of the liquid crystal (ZLI-2293, Merk Japan) in the 25-μm-thick cells were measured by using the crystal rotation method [4].

Figure 1: Structures of studied polyimides

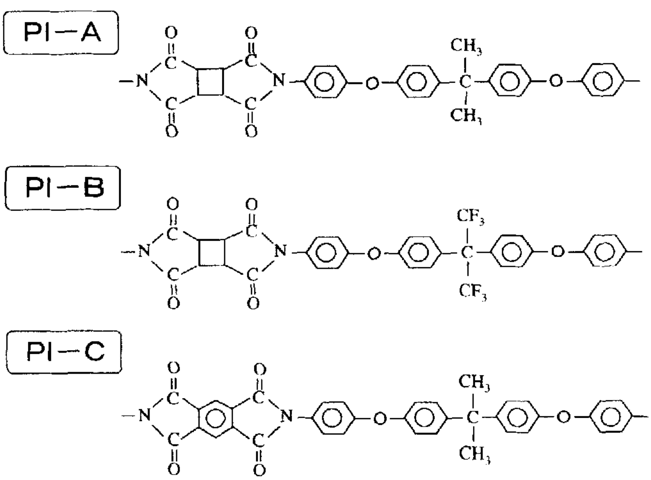


Figure 2 shows the observed polarizations for a strongly rubbed sample (f=392 rpm, L=1.12 mm, V=4.36 mm/s, N=3) and a weakly rubbed one (f=135 rpm, L=1.12 mm, V=4.36 mm/s, N=3). The anisotropy of the polarizations of both samples were clearly visible in the phase difference (Δ) and amplitude ratios (ψ) of the S and P components of the reflected light. Figure 2 clearly indicates that the optical anisotropy of polyimide films depends on the rubbing conditions.

The anisotropy of the rubbed polyimide film was determined based on the observed polarizations. We approximate the upper molecular oriented layer as an optically uniaxial medium and the lower random layer as an isotropic one [5, 6, 7, 8, 9, 10]. The upper uniaxial layer is characterized by four parameters: two anisotropic dielectric constants ε_e and ε_o, the tilt of the principal dielectric axis θ, and the thickness d_u. The lower random layer can be described by the isotropic dielectric constant ε_i and thickness d_l. The polarization of the reflected light is calculated by using all six parameters in 4x4 matrix formalism [11, 12]. The six parameters can be optimized by using the non-linear least-square-method to fit the observed polarization to the calculated one. We assumed that the isotropic dielectric constant ε_i is the average of the two anisotropic dielectric constants ε_e and ε_o. In truth there were only five optimized parameters.

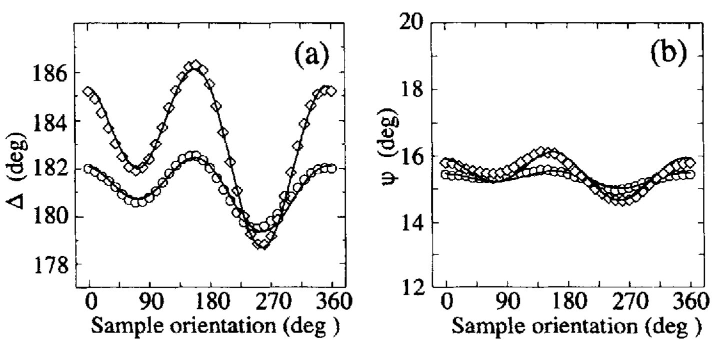


Figure 2: Observed polarizations of strongly and weakly rubbed PI-A. Open circles are observed polarizations for weakly rubbed film, and open diamonds are for strongly rubbed ones. Solid curves are

The solid curves in Figures 2(a) and 2(b) are calculated polarizations based on optimized parameters. In the case of the strongly rubbed sample, the optimized values of the two anisotropic dielectric constants (ε_e and ε_o) were 2.89 and 2.72, and the tilt of the principal dielectric axis (θ) was 35.9°. The thickness of the molecular oriented layer (d_u) was 35.0 nm. The dielectric constant (ε_i) was determined as 2.78, and the thickness of the random layer (d_l) was 58.0 nm. The good fit between the calculated and observed polarizations shows that our two- layer model does a good job of approximating the optical structure of a rubbed polyimide film. The parameters of the weakly rubbed sample were determined as ε_e=2.80, ε_o=2.74, θ=32.8°, d_u=35.7 nm, ε_i=2.76 and d_l= 66.7 nm. These results show that the optical structure of rubbed polyimide film is largely dependent on the rubbing condition.

We focused on the relationship between pretilt and the parameters characterized molecular-oriented layer. We introduced a parameter defined as $(\varepsilon_e-\varepsilon_o)/\varepsilon_i$ as an indicator of polyimide molecular ordering. We observed that the pretilt angles decreased monotonically as increased $(\varepsilon_e-\varepsilon_o)/\varepsilon_i$ in PI-A and PI-C, as shown in Figure 3. There seems to be linear relationship between pretilt angle and $(\varepsilon_e-\varepsilon_o)/\varepsilon_i$ in PI-A and PI-C. The correlation coefficient between pretilt and $(\varepsilon_e-\varepsilon_o)/\varepsilon_i$ for PI-A was -0.96, and for PI-C, was -0.79. In the case of PI-B, the dependence of the pretilt angle on $(\varepsilon_e-\varepsilon_o)/\varepsilon_i$ was unclear; the correlation coefficient between pretilt and $(\varepsilon_e-\varepsilon_o)/\varepsilon_i$ was rather small, as shown in Table I.

Figure 3: Pretilt versus $(\varepsilon_e-\varepsilon_o)/\varepsilon_i$ for PI-A, PI-B, and PI-C

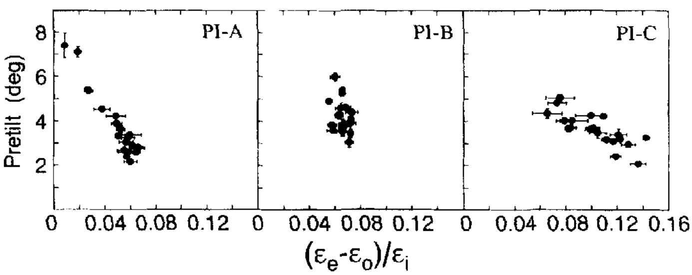
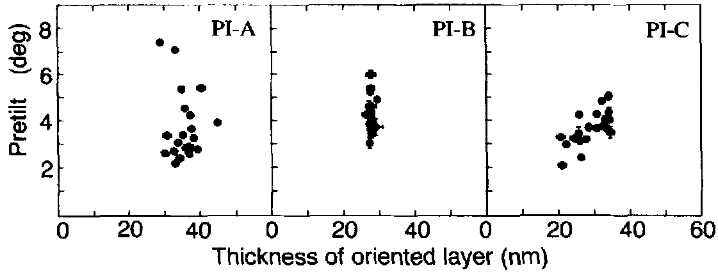


Table I: Correlation coefficients between pretilt and three parameters of rubbed polyimide film ((ε_e-ε_o)/ε_i, thickness of molecular-oriented layer and tilt of dielectric axis).

	$(\varepsilon_e-\varepsilon_o)/\varepsilon_i$	Thickness of molecular oriented layer	Tilt of dielectric axis
PI-A	−0.96	−0.48	−0.13
PI-B	−0.36	−0.02	0.00
PI-C	−0.79	−0.83	0.70

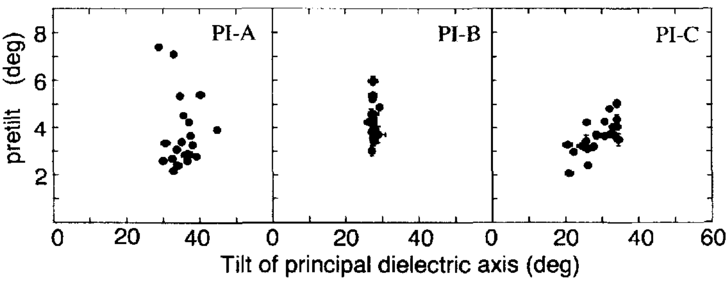
Figure 4 shows the pretilt versus the thickness of the molecular oriented layers of PI-A, PI-B and PI-C. The pretilt in PI-A seems to vary independently of the thickness of the polyimide molecular-oriented layer around 35 nm. There is no detectable variation of pretilt over 40 nm. We, therefore, concluded that there is no relationship between the pretilt angle and the thickness of the molecular-oriented layer in PI-A. In the case of PI-B, the correlation coefficient between the pretilt and the thickness of the molecular-oriented layer was -0.02, and there was no correlation between the pretilt and the thickness of the molecular- oriented layer. A monotonic decrease in pretilt was observed as the thickness of the molecular-oriented layer of PI-C increased. This is shown in Figure 4. The correlation coefficient between the pretilt and the thickness of the molecular-oriented layer was -0.83.

Figure 4: Pretilt versus thickness of molecular-oriented layer



The pretilt and tilt of the dielectric axes of PI-A, PI-B, and PI-C were in the same direction, and the tilt of the dielectric axis has some influence on pretilt. However, there was no detectable relationship between the pretilt and tilt of the dielectric axes of PI-A or PI-B. The correlation coefficient for the pretilt and tilt of PI-A's dielectric axis of was -0.13. PI-B's pretilt varied between 3.0° and 6.0°. Despite this, the tilt of the dielectric axis was relatively constant around 28°. The correlation coefficient for the pretilt and tilt of PI-B's dielectric axis was 0.01. For PI-C, by contrast, the pretilt angle increased as the tilt of the principal dielectric axis increased.

Figure 5: Pretilt versus tilt of dielectric axis



As mentioned, the influence of the optical anisotropy of a rubbed polyimide film on pretilt angle differs widely among PI-A, PI-B and PI-C. In the case of PI-C, it seems that the tilt of the dielectric axis, $(\varepsilon_e-\varepsilon_o)/\varepsilon_i$, and the thickness of the molecular oriented layer influence the pretilt angle. By contrast, in PI-B, anisotropy of rubbed film did not influence pretilt. We see that the influence of polyimide molecular orientation on pretilt is strongly dependent on the polyimide type. Furthermore, the absence of correlation between pretilt and PI-B's molecular orientation suggests that some unknown factor controls pretilt.

In summary, we investigated the relationship between the pretilt angle and optical anisotropy of rubbed polyimide film by using reflection ellipsometry. We found that the influence of polyimide molecular orientation on pretilt is strongly dependent on the polyimide type, and that there is some unknown factor directing pretilt angle separate from the molecular orientation of polyimide.

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