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A new technique for direct evaluation of the influence of boundary condition on molecular alignment and dynamics using a transferred ferroelectric liquid crystal film

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Recently, the influence of surface anchoring to the molecular reorientation of liquid crystal (LC) has been measured by electro-optical effect and ellipsometry, which used a sandwich cell geometry containing LC between two glass substrates. In these methods, however, only the averaged value in the film whose thickness was typically several μm could be evaluated.

The smectic liquid crystal has a layer structure and can be used in a freely suspended film (FSF) whose thickness can be varied from two to several thousands of smectic layers. The FSF has attracted considerable attention as a two-dimensional system. In addition, the FSF has air/LC surfaces which induce a characteristic feature of the molecular dynamics.[1,2]

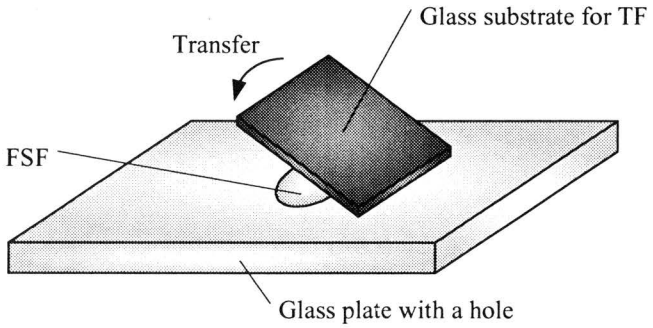


FIG. 1. Schematic explanation of the fabrication of TF.

The FSF can be transferred onto a solid substrate.[3] The transferred film (TF) of a smectic liquid crystal has both air/LC and substrate/LC interfaces, and a thin film with the thickness of the order of the molecule can be realized. Therefore, the TF is suitable for the characterization of the boundary effects on the molecular alignment and dynamics.

The chiral smectic liquid crystal showing tilted phases has a ferroelectricity and possesses a spontaneous polarization, P_s. So far, numerous studies on the evaluation of P_s in a ferroelectric liquid crystal (FLC) have been carried out.[4] The measurement of the polarization reversal current due to P_s is one of the most powerful techniques for the evaluation of the molecular reorientation dynamics. P_s measurement using a FSF geometry has also been reported.

In this study, we measure the polarization reversal current in the TF of FLC and propose a new technique for the direct evaluation of the boundary effects on molecular dynamics in the transferred liquid crystal film. Using this technique, the influences of the air/LC and substrate/LC interfaces can be investigated, because a LC layer of well-controlled thickness can be formed on the substrate. In order to measure the polarization reversal current, the TF was formed on a substrate with an

interdigitated electrode, and the current was measured as a function of film thickness, temperature and measuring frequency.

The FLC used in this study is CS-1029 (Chisso). It shows a chiral smectic C (SmC*) phase between −18 and 73°C, and P_s measured in a bulk sandwich cell is 41nC/cm^2 at 25°C. The FSF was prepared in a hole (5mm in diameter) on a glass plate (30mm×30mm) in the smectic A (SmA) phase, and subsequently the FSF was transferred onto a glass substrate (10mm×13mm) with an interdigitated Al electrode by gently bringing the substrate close to the film in the SmA phase as shown in Fig. 1. The distance between the electrodes was 10μm. The width of each electrode was 10μm, and its total length was 2mm. The number of electrodes was 130. The number of layers N was determined by measuring the reflection spectrum of the FSF in the SmA phase. The polarization reversal charge Q was measured by a conventional triangular wave method.

Figure 2 shows the temperature dependence of Q in TF. As evident from this figure, the temperature dependence is related to the film thickness, that is, the number of layers N. In a thick TF, Q is almost zero above a transition temperature between the SmA and SmC* phases, T_c. With decreasing temperature, Q increases monotonically below T_c, which is the same as the tendency observed in the bulk. Namely, FLC shows the second-order phase transition and P_s in the SmC* phase increases as $P_s \sim (T_c-T)^{\beta}$. β is a critical parameter and has been estimated to be 0.3~0.4 in the conventional sandwich cell.[5] β in the thick TF calculated from Fig. 3 (b) is about 0.4, which coincides with that in the sandwich cell.

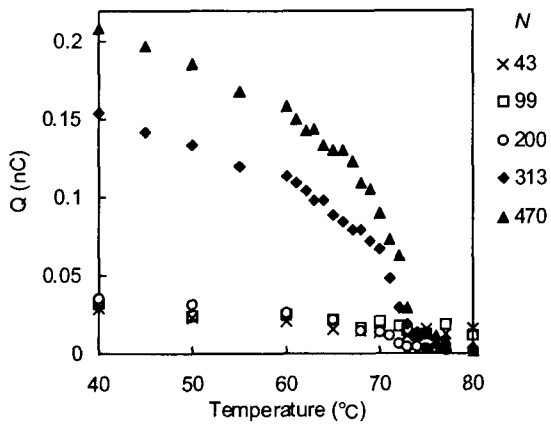
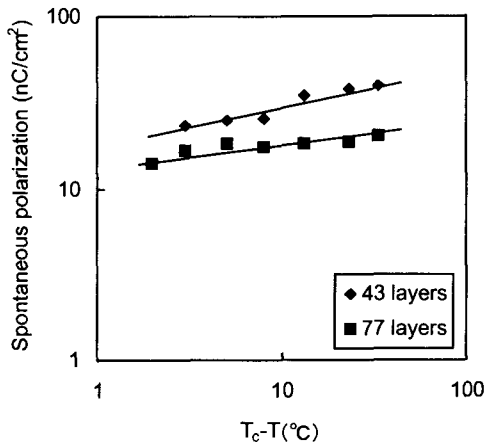
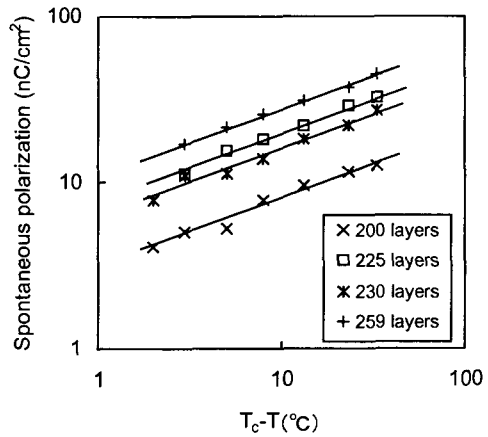


FIG. 2. Temperature dependence of Q in TF.



(a)



(b)

FIG. 3. The Tc-T dependence of Ps.
((a) Thin film, (b) Thick film).

In a thin TF, however, Q is not zero even above T_c and does not show a marked temperature dependence even below T_c as shown in Fig. 2. In addition, β in the thick TF calculated from Fig. 3 (a) is about 0.2. In other words, the temperature dependence of Q in the thin film is different from that in the thick TF and the sandwich cell. It is well known that T_c differs between the bulk and the surface in FSFs. We measured the dependence of T_c on the number of layers N in the TF by X-ray diffraction. However, no conspicuous change of T_c was observed in the TF used in this study (N>52 layers). Therefore, it is considered that there is a critical number of layers N_c to induce the influence of air/LC interface in the TF.

Figure 4 shows the N dependence of Q in the SmC* phase (40°C). Above N=200, Q increases monotonically with increasing N. This indicates that P_s is independent of N. In other words, P_s is not influenced by the substrate/LC interface in the thicker film (N>200). On the other hand, in the thinner film (N<200), Q is almost constant in spite of change in N.

The existence of the critical thickness is confirmed also in an electro-optical study. The optical transmission change upon application of a triangular voltage in a TF has been investigated. Figure 5 shows the N dependence of the threshold field of the transmission change. As evident from this figure, although the threshold field is almost zero above N=200 in the same manner as for the thicker FSF, it increases under 200 layers with decreasing N. This indicates that the molecular motion is restricted by the substrate/LC boundary effect in a film thinner than 200 layers.

On the other hand, in a thin film, P_s could

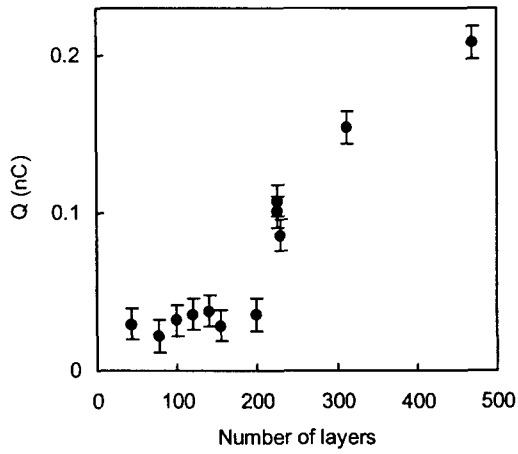


FIG. 4. N dependence of Q in the SmC* phase (40°C).

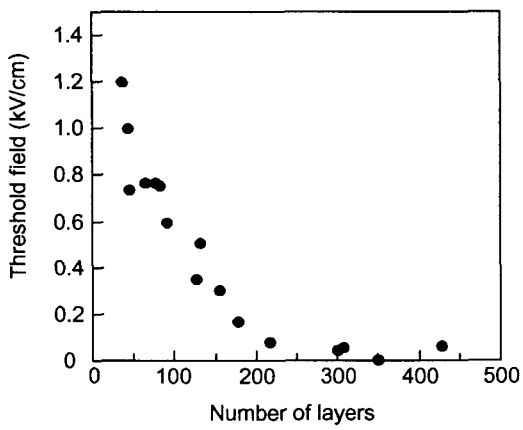


FIG. 5. Threshold field of the transmission change as a function of N (25°C).

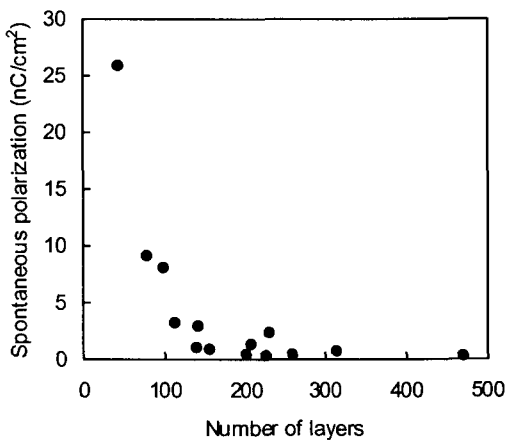


FIG. 6. N dependence of P_s in the SmA phase (80°C).

be observed even above T_c as shown in Fig. 6. This result may be due to the surface clinic effect of the molecules at the air/LC interface. In a FSF, the molecules in the vicinity of the air/LC interfaces tilt even in the SmA phase. As a result, polarization coupled to the molecular tilt is induced near the air/LC interface. In the TF, the region exhibiting the induced polarization is restricted to the vicinity of the air/LC interface and the molecules near the substrate/LC interface are aligned perpendicular to the surface. Another explanation for the enhancement of P_s in the thinner TF is a flexoelectric contribution to the polarization. The flexoelectric effect is based on the director deformation of the polar molecules. As mentioned above, at the air/LC interface, the molecules tilt with respect to the surface normal. In contrast, at the substrate/LC interface the molecules show a tendency to align homeotropically, that is, perpendicular to the surface. Therefore, the bend deformation of the director occurs along the surface normal. As a result, flexoelectric polarization is induced and the non-zero in-plane component of the polarization appears even in the SmA phase.

In conclusions, we successfully realized a new method for direct evaluation of the boundary effects of the air/LC and the substrate/LC interfaces on the molecular alignment and dynamics using a thin transferred ferroelectric liquid crystal film. From the N dependence of Q below the Curie temperature, it was found that the anchoring effect suppressed the spontaneous polarization in a film thinner than approximately 200 layers. This result corresponds to the result of the electro-optic experiment. In addition, the polarization reversal current has been observed even above a Curie temperature in a thin film, which is more enhanced in thinner film. It was interpreted as follows the thin film had larger value of an averaged tilt angle of liquid crystal molecules compared with that of the thick film because of the surface clinic effect. The flexoelectric contribution to the polarization in the SmA phase was also discussed.

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