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Dependence of the molecular orientational states on the surface conditions for the ''V-shaped switching'' in a ferroelectriclike liquid crystal sample

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The three-component mixture showing the V-shaped switching in a rubbing cell is investigated using a temperature gradient cell (TGC) where the epitaxial method of alignment, but without an alignment layer, is used. The molecular alignment and the switching characteristics in a TGC are compared with the rubbing sandwich and the open cells. The uniform or the twisted state is observed in the TGC, depending on the surface condition whether indium-tin oxide (ITO) or glass, while the uniformlike state with the average optical axis parallel to the smectic layer normal emerges only in the rubbing cell. The cancellation of the surface polarization charges by the surface free charges on the ITO surfaces determines the molecular alignment state in the TGC. From these results, we conclude that the nonpolar in-plane anchoring plays an important role in giving a uniformlike state in the rubbing cell.

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In the tilted chiral smectic liquid crystal (LC) cells, the interaction between liquid crystalline chirality and surface polarization causes many interesting and varying interfacial phenomena. Along with the nonpolar molecule-surface interaction, this polar interaction provides a useful means to define a preferential spatial orientation of the molecules in a thin cell. Conventionally, these interactions have been investigated in detail for the ferroelectric liquid crystal (FLC) cells. The appearance of different ferroelectric liquid crystalline states such as the uniform, the twisted and the helical states are governed by the surface-molecule interactions [1–3].

Recently, a unique uniformlike ferroelectric state was observed in the V-shaped switching cells [4–7]; here the averaged optical axis at an applied voltage $V=0$ is almost parallel to the layer normal l similar to that in a helical cell. Consequently, very low transmittance is observed under crossed polarizers [see arrow in Fig. 4(b)]. We have shown that this apparent uniformlike state originates from the very small twisted state along the cell thickness (2–3 μm) [8,9]. First, the molecules near the rubbed surface are aligned along the rubbing axis **r** due to the strong in-plane anchoring by the rubbing treatment [10]; the angle between **l** and **r** is ~12°. This is confirmed in both cells, the open cell with an-air molecule boundary [8] and the one-side rubbing sandwich cell [9]. Second, the small twisted molecular orientation [8] is determined by the balance between this in-plane anchoring [10] and the polar anchoring on the unrubbed surface. The calculated azimuthal angle, ϕ (the angle that the spontaneous polarization **P** makes with the surface normal) is $\sim 0.41\pi$ on one surface and $\phi=\sim\pi+0.41\pi$ on the second surface [8,9].

On the other hand, other studies have concluded that the electrostatic bulk property in the thin cell induces the uniformlike molecular state. According to Rudquist *et al.* [7], **P** in the bulk is aligned uniformly parallel to the surface ($\phi=\pi/2$) by self-interaction of polarization space charge even though the molecules are abruptly twisted in the very thin surface regions ($\phi=\mathbf{0}$ and π). In addition, more recently, Clark *et al.* [11] have suggested that the completely uniform alignment of **P** with $\phi=\pi/2$ from surface to surface arises from the electrostatic effect in the limit of high **P**. Ikeda *et al.* [12] have recently suggested the modified electrostatic model where the smectic layer twists on approaching both surfaces from the center of the cell. Here the layer twist was introduced to compensate the abrupt molecular twisted state from the bulk to the surface region as was suggested by Rudquist *et al.* [7].

The electrostatic model [7,11,12] for explaining the emergence of the uniformlike ferroelectric state is different from our model [8,9] in that the electrostatic model ignores the molecule-surface interactions including the in-plane anchoring [10]. In this paper, we investigate the molecular alignment obtained in a temperature gradient cell (TGC) using the epitaxial method of alignment [13]. We can obtain a high quality planar alignment on the ITO (indium-tin oxide) surface *without rubbing treatment*. Hence the optical properties in the TGC and the rubbing cell need to be compared for understanding the origin of the uniformlike molecular alignment in the rubbing cell.

In order to obtain a planar cell without rubbing treatment, we have used the special ITO-coated glasses patterned as in Figs. 1(a) and 1(b). Figure 1(a) has two ITO regions independent of each other. One is for the electrode (E1) and the other for the local heater: Fig. 1(b) has one ITO region for

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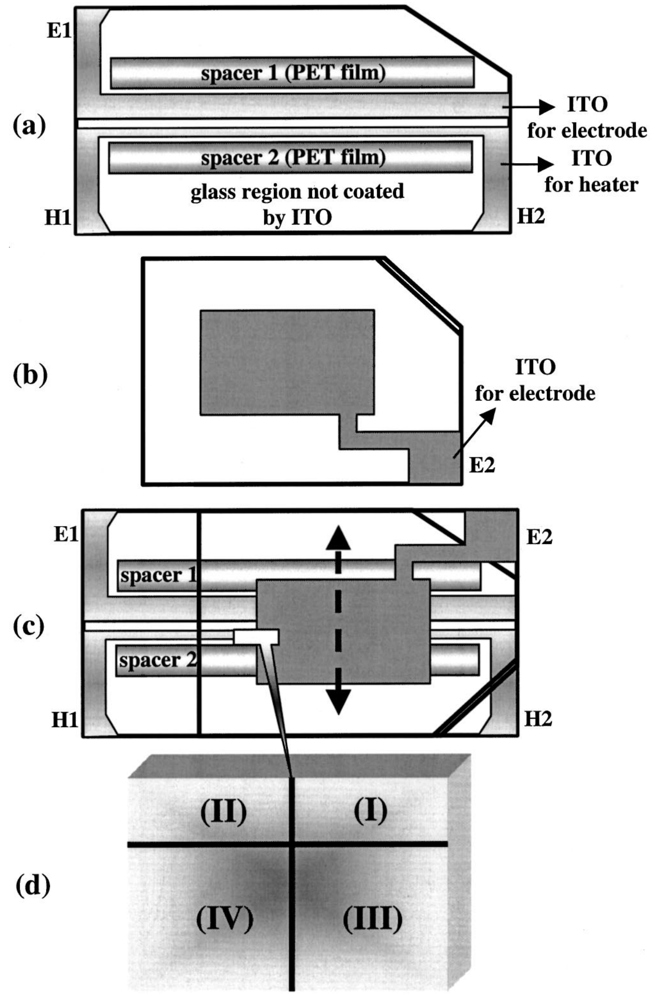

FIG. 1. Schematic diagram for a temperature gradient cell (TGC). (a) Lower substrate, (b) upper substrate, (c) assembled cell, (d) the region with different surfaces: (I) ITO and ITO, (II) ITO and glass, (III) glass and ITO, (IV) glass and glass.

the electrode (E2). After assembling two ITO-coated glasses as in Fig. 1(c), when an electric current flows between H1 and H2, ITO works as a heating element and then the temperature gradient is produced around the ITO region along the dotted arrow [Fig. 1(c)]. The cell gap was controlled by a polyethylene terephthalate (PET) film with a thickness of 2 μm. The edge of the PET film cut by a sharp knife also acts as a nucleus for the growth of the crystal. The Inui mixture (Tokyo mixture) [5] was inserted into the cell at the isotropic phase [the phase sequence: AF [20–43 °C: coexistence with SmX^*), SmX^* (64 °C), SmA (68.5 °C) iso]. By lowering the temperature of the hot stage on which the TGC is fixed, the monodomain of the SmA phase (the smectic layer normal l parallel to the edge) starts forming from the sharp edge of the spacer 1 (PET film) in Fig. 1(c). The temperature of the hot stage is set to decrease at the rate of 0.1 °C per min. from 55 °C. The ITO heating element with resistance 800 Ω is turned on with 20 V applied across it. Due to this local heating, there is a strong temperature gradient along the direction shown by the dotted arrow in Fig. 1(c). When the phase transition, iso-SmA, occurs at the edge of the spacer 1, the temperature of the hot stage is $\sim$40 °C; however, the sample close to this heating element is still isotropic. After the entire sample between two PET films was changed to a liquid crystalline phase, the local heating in the TGC was ceased, and the temperature of the hot stage is raised slowly as appropriate.

Figure 2 shows the textures obtained from the TGC. Quite good alignment was obtained in the SmA phase [Fig. 2(a-1) and (a-2) at 65 °C]. Figure 2 (a-2) was taken at the position, where the stage was rotated 45° from (a-1). Meanwhile, in the SmX* phase, we can observe two kinds of uniform states $U1$ and $U2$. $U1$ and $U2$ in Fig. 2(a-3) at 50 °C, show extinction position when the stage is rotated $\sim$26° from Fig. 2(a-3) to the left and the right, respectively. However, each uniform domain under a high electric field has the apparent tilt angle of $\sim$34°. These two different angles can be explained by the existence of the chevron structure and the pretilt of $\mathbf{P}$ on the surfaces [14–16].

In Fig. 2(b), we have also observed the texture in the region that has four boundaries. As indicated in Fig. 1(d), the region (I) has the ITO-coated surfaces in both upper and lower substrates as in Fig. 2(a). On the contrary, three other regions (II), (III), and (IV) have different surfaces from the region (I). In the SmA phase, the same monodomain was obtained regardless of the surface condition [Fig. 2 (b-1), (b-2), and (b-3]. However, in the SmX^* phase, each region with different surface conditions shows different molecular orientational states. First, two different uniform states are found in region (I) with the ITO on both surfaces in the same way as shown in Fig. 2(a). Second, in regions (II) and (III) with the ITO only on one surface, one uniform state ($U1$ or $U2$) predominates. Thirdly, the region (IV) with glass on both surfaces shows two different twisted states $T1$ and $T2$.

In the cell with the ITO-liquid crystal interface, the emergence of the different states under different surface conditions has been analyzed by Clark *et al.* [11]. Here surface free charges in addition to surface anchoring play an important role in determining the molecular orientation along the cell thickness. They have classified the molecular state according to the magnitude of $\mathbf{P}$; that is, with increasing $\mathbf{P}$, the normal uniform state ($\phi=\mathbf{0}$ or π on both surfaces), the twisted state ($\phi=\mathbf{0}$ on one surface and π on the second) and another uniform state ($\phi=\pi/2$) emerge successively. Accordingly, they have predicted the uniform state with $\phi=\pi/2$ in the sample with higher $\mathbf{P}$ such as the Inui mixture ($\mathbf{P}=40\sim90\,\mathrm{nC\,cm^{-2}}$) investigated in this paper. As shown in Fig. 2, the uniformlike state with $\phi=\pi/2$ did not emerge in the TGC.

We examine our results based on their suggestion. The schematic diagram for explaining the role of the surface free charges on the ITO surfaces is given in Fig. 3. To simplify the system, qualitatively, we ignore the chevron structure and the pretilt of $\mathbf{P}$ observed in the real TGC. Generally, since the in-plane anchoring does not exist in the TGC because of the absence of the rubbing treatment, the surface energy is given as $W_s=-\gamma_p\cos\phi-\gamma_n\cos^2\phi$, γ_p and γ_n correspond to the polar and nonpolar anchoring strength coefficients [17]. The surface polarization charges (thick sym-

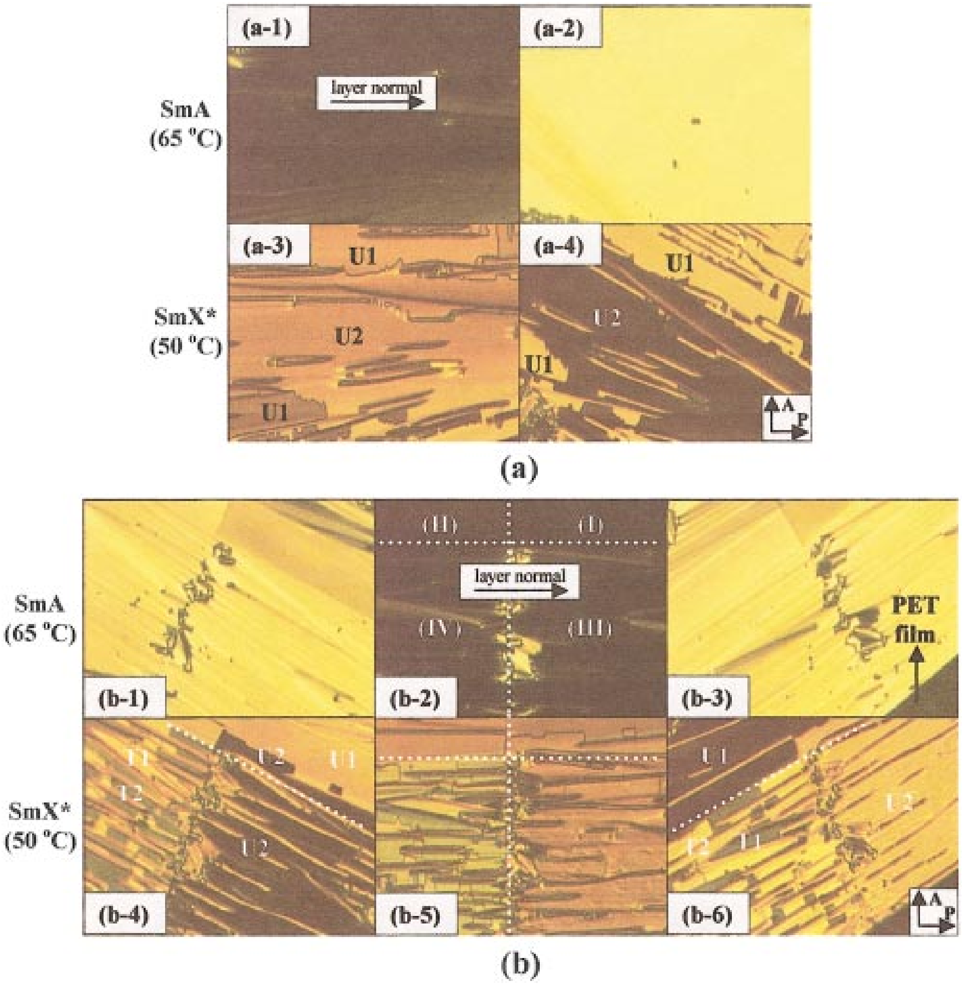

FIG. 2. (Color) Textures in the TGC. In (a-2) and (a-4), the stage was rotated ~45° and ~26° from (a-1) and (a-3), respectively. (b) was observed in the region shown in Fig. 1(d). The stage in (b-1) and (b-3) was rotated by ~30° with respect to (b-2). And the stage in (b-4) and (b-6) rotated to find the extinction position of $U1$ and $U2$ from (b-5). Textures in (a-3) and (a-4) correspond to region (I) in Fig. 1(d).

bols) in Fig. 3 (I) are compensated with the surface free charges (thin symbols) so γ_n predominates in W_s. Consequently the minimum energy is degenerated at $\phi=0$ and π and we observe two uniform states. For Fig. 3 (II) and (III), the surface polarization charges on the ITO surface are compensated by the surface free charges, however, they remain on the glass surface. Therefore the molecule-surface polar interaction on the glass surface determines the molecular tilting sign, and as a consequence one uniform state predominates depending on the position of the glass surface. The emergence of the twisted states in Fig. 3 (IV) is quite natural because the molecule-surface polar interactions on both glass surfaces compete with each other.

The results in the TGC, without any aligning layer, show the sensitivity of the Inui mixture to the surface interactions. In Table I, we summarize the static molecular states observed in the three different cells. If the electrostatic energy controls the molecular orientation of the Inui mixture, the uniformlike state [see Fig. 3 (V)] must emerge in both the rubbing cell and the TGC [11] because the molecule-surface interaction can be ignored compared with the polarization charge self-interaction. In addition, even the open cell with

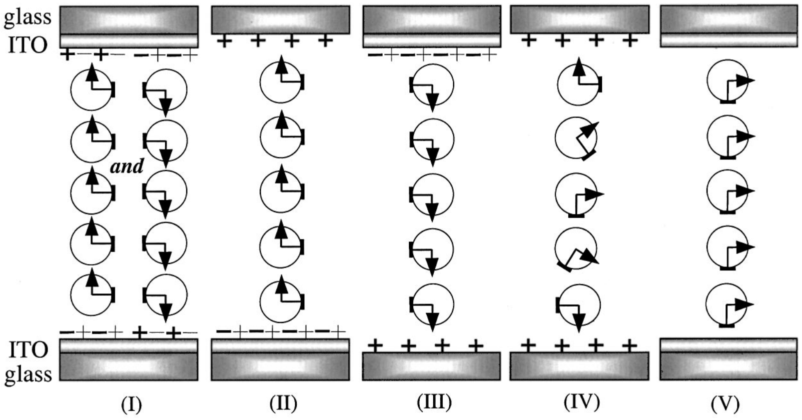

FIG. 3. Schematic explanation of the states observed in Fig. 2(b). Thick symbols denote the surface charges due to **P** and thin symbols denote the free charges on electrodes. (V) shows the space charge stabilized uniform state as suggested by Clark *et al.* [11], however it did not emerge in the TGC.

TABLE I. The molecular states of the Inui mixture observed in different cells.

	One-side rubbing sandwich cell	open cell	TGC
layer normal axis **l** formed in Sm*A*	~12° away from the rubbing axis **r**	~12° away from the rubbing axis **r**	Parallel to the edge of the PET film
average optical axis in Sm*X**	layer normal **l**	rubbing axis **r**	~26° away from the layer normal **l**

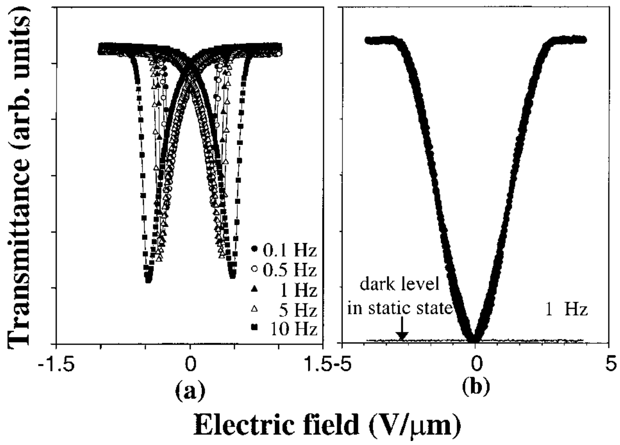

FIG. 4. The switching pattern for a triangular signal (a) for the TGC, (b) for the rubbing cell coated by polyimide RN1199. In the bistable switching in (a), the critical field for uniform state increases due to the ionic effect by increasing frequency. The V-shaped switching in (b) is almost independent of frequency in the range given.

an air-molecule boundary would have to show a common surface-stabilized ferroelectric state close to $\phi=\mathbf{0}$ or π instead of $\phi=\sim 0.41\pi$ [8]. Therefore, it is quite obvious that the electrostatic model alone cannot be applied to the explanation of the uniformlike state observed in the rubbing cell of the Inui mixture; conversely, it can be explained by the in-plane anchoring [10], and the small twisted state. The result that was used to suggest the twisted layer structure in Ref. [12] also supports the existence of the molecular twisted state in the rubbing cell.

Figure 4 shows the switching pattern in the TGC and the rubbing cell. The bistable switching characteristic in the SSFLC (surface-stabilized ferroelectric liquid crystal) cells [1,17,18] is observed in the TGC, while the V-shaped switching emerges in the rubbing cell [8]. It is, therefore, suggested that the uniformlike state due to the in-plane anchoring is the prerequisite for the V-shaped switching.

In summary, the Inui mixture shows the uniformlike state in the rubbing cell where the average optical axis is parallel to **l**. In the TGC without the rubbing treatment (and aligning material), however, the normal uniform or the twisted state is observed because of the cancellation of the surface polarization charges by the surface free charges; we obtain the common uniform state ($\phi=\sim\mathbf{0}$ or $\sim\pi$) for the ITO-coated surfaces whereas for the glass surfaces, the twisted state is obtained. It follows, therefore, that the electrostatic model by space charge stabilization alone cannot be applied to the Inui mixture. Accordingly, the uniformlike state emerging in the rubbing cell of the Inui mixture [8], can be understood in terms of the in-plane anchoring due to the rubbing treatment and from the properties of the mixture.

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