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## Flow-Controlled Alignment of Smectic Liquid Crystal by “Grooves” and “Ribs”

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A new cell structure has been proposed, in which parallel micro grooves are etched on glass substrates and a pair of them are adhered to ribs in parallel alignment to the direction of grooves. Uniform smectic orientations have been achieved in this cell structure with both the grooves and ribs, and excelled the orientation in cells with rubbed polyimide. However, the best orientations have never been achieved within the cells using only grooves or only ribs. The important role of ribs is considered to control the shape and moving rate of liquid crystal-air boundaries when liquid crystals were introduced into the cell.

**Keywords:** flow-controlled alignment; smectic liquid crystal; grooves; ribs; temperature gradient cooling

### INTRODUCTION

We have developed temperature-gradient cooling as an alignment method of smectic liquid crystals (LCs) such as ferro- and anti-ferroelectric LCs<sup>[1,2]</sup>. This method provided superior uniform orientation free from zigzag defects typical to chevron smectic layers. However, in

the cells treated with this method, there were many scratches and zigzag defects caused by small particles caught in the rubbing process. This method has so far been applied to cells with rubbed polyimide layers.

A new cell structure has been developed, eliminating the need for rubbing process. Grooves are formed on the surface of glass substrates that contact with the LCs. Ribs which perform the function of spacers are also formed parallel to the direction of the grooves.

The purposes of this paper is; (a) to show that uniform orientations in smectic LCs are achieved in the cell, described above, combined with temperature gradient cooling; (b) to examine the geometrical and material conditions for the ribs and grooves; (c) to discuss qualitatively the roles of the ribs and grooves, in particular focusing on ribs.

## EXPERIMENTAL

### Preparation of Cells

90mm squared glass substrates coated with Indium Tin Oxide (ITO) available from Japan Sheet Glass Co., Ltd. were used in this experiment. The thickness of the ITO layer was 100nm. The grooves were formed by etching the ITO layer. The width of etched and non-etched parts was equal and the pitch of grooves was 2 to 8  $\mu\text{m}$ . The depth of the grooves were controlled from 10 to 50nm by changing the etching period. Figure 1(a) shows an AFM image of grooves (depth; 50nm). The ribs were fabricated with the usual photolithography process by use of a nega-type resist, which included epoxy resins functionalized with acryl groups. The ribs with width 20 $\mu\text{m}$ , height 1.8 $\mu\text{m}$  and pitch 80-300 $\mu\text{m}$  were formed parallel to the grooves on the substrate. Another substrate with grooves was completely adhered to the ribs on the former substrate by an appropriate heat and press treatment. The standard cell structure is illustrated in Figure 1(b). The geometrical dimensions and materials of the ribs and grooves were changed, if necessary, in order to clarify influences on LCs orientations.

The LCs in the nematic phase were introduced into the cells by capillary effect. Felix SCE9 from Höchst, CS1014 from Chisso and K24 from Merck were used as smectic LCs, and K15 from Merck as a

nematic LC for reference. They are listed in Table 1.

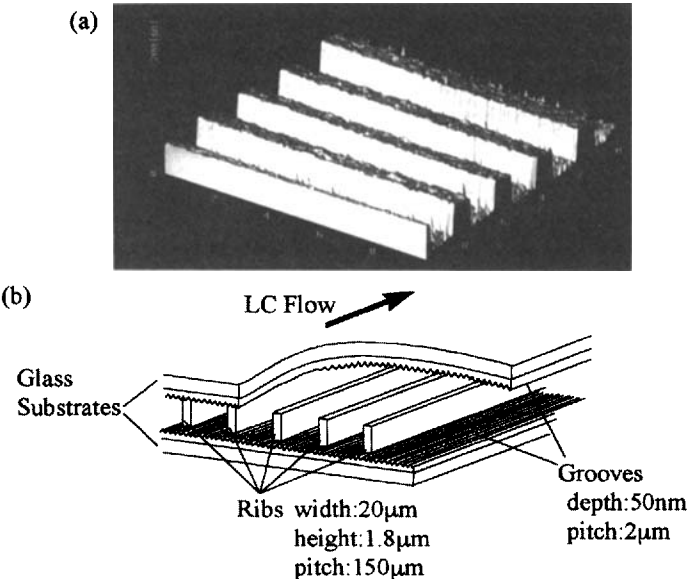


FIGURE 1 (a) AFM image of grooves (depth: 50nm), (b) Standard cell structure schematically.  
See Color Plate XV at the back of this issue.

TABLE 1 Liquid crystals used.

LC	Transition temperature/°C				Filling temperature/°C <sup>*a</sup>
	Iso	N	SmA	SmC	
SCE9	118-113	86	64		102
CS1014	81	69	54		75
K24	41	34			37
K15	35				25

<sup>\*a</sup>a Filling temperature means the temperatures at which LCs were introduced into the cells.

Temperature Gradient Cooling

The cell filled with LC was placed on a temperature-controllable hot

plate and the upper side of the cell was covered with a thick aluminum plate to keep the temperature of the cell uniform. The cell was removed parallel to the grooves from the hot plate into air temperature at 2–4 mm/min. The temperature of the cell was monitored by sensors attached on the glass substrates and a steep temperature gradient was applied around the edge of the hot plate.

#### Observation of the LC-Air Boundries

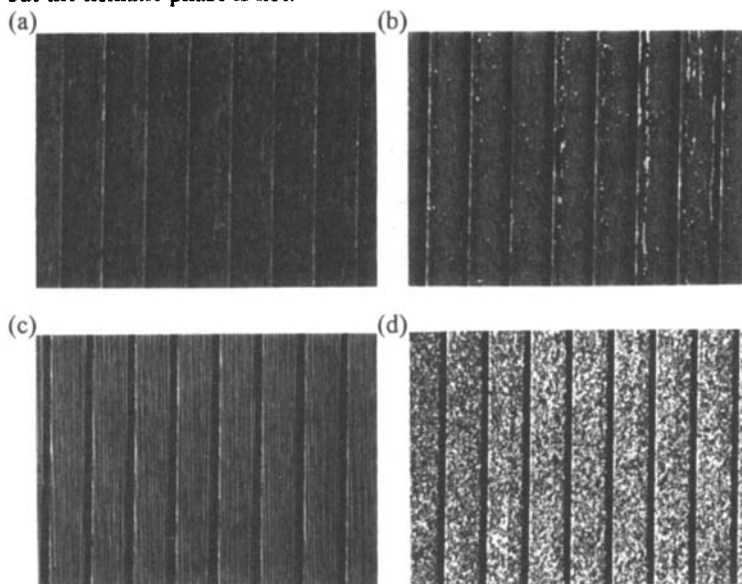
The boundaries between air and LCs that were flowing into the space between ribs in a cell in the nematic phase were observed under a polarized microscope. The directors were determined from the observations of the extinction parts of the flowing LC and the texture changes in the cooling process to the smectic phase. Under crossed nicols condition, the directors in the extinction part should correspond to the direction of analyzer or polarizer. The direction in which the director lay, analyzer or polarizer, was determined from the texture observation of smectic layers.

### RESULTS AND DISCUSSION

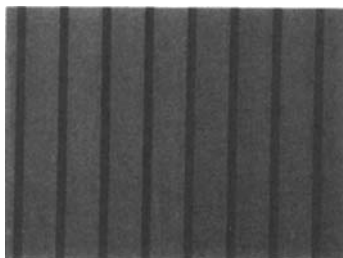
#### Smectic Layers Orientation

Figure 2 shows microscopic photographs of initial orientations after four kinds of LCs in Table 1 were introduced into the cell with ribs pitch 150  $\mu\text{m}$ , height 1.8  $\mu\text{m}$ , grooves pitch 2  $\mu\text{m}$  and depth 50 nm, followed by gradual cooling to room temperature. Three kinds of smectic LCs basically provided better initial orientations with the layer normal direction parallel to the grooves, though there were slight differences among the orientations and small zigzag and line defects in those as seen in Figure 2. Temperature gradient cooling remarkably improved the initial orientations of SCE9 as shown in Figure 2(a) and eliminated these defects as shown in Figure 3. These orientations were superior to those in polyimide rubbed cells. On the other hand, the initial orientation of K15, nematic LC, was inferior to that in polyimide rubbed cell. The smectic phase is capable of self organization as a result of molecular interactions,

but the nematic phase is not.



**FIGURE 2** Initial orientations for LCs listed in Table1; (a) SCE9, (b) CS1014, (c) K24, (d) K15  
See Color Plate XVI at the back of this issue.

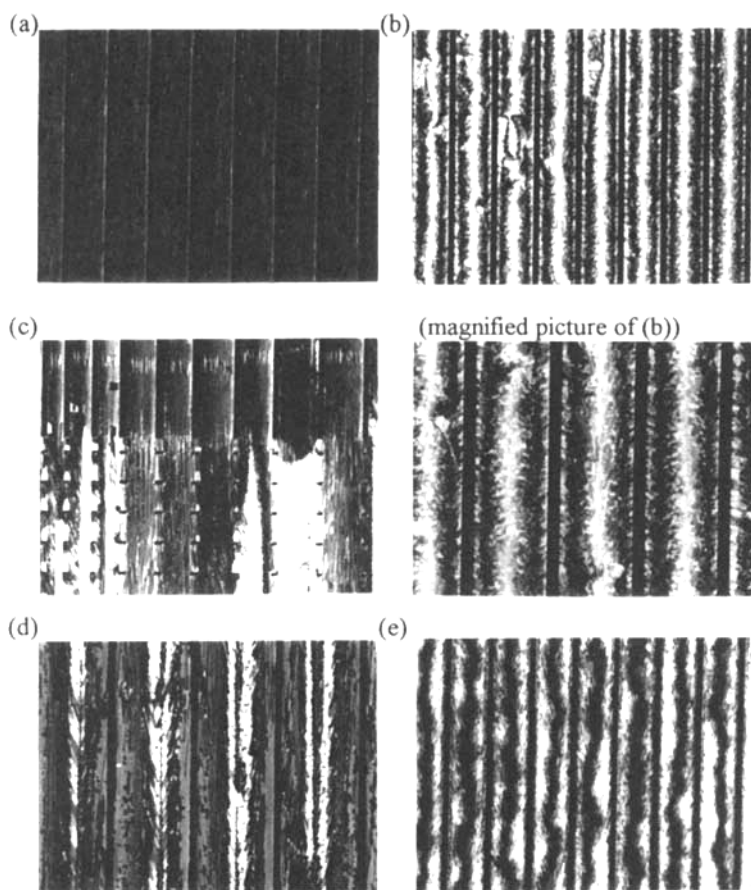


**FIGURE 3** Orientation for SCE9 after temperature gradient cooling.  
See Color Plate XVII at the back of this issue.

**The Effect of Geometrical Dimensions and Types of Grooves and Ribs**

Figure 4 shows the initial orientations of SCE9 for five cells with

different geometrical dimensions and types of grooves and spacers. Their conditions are listed in Table 2. We refer (a) in Table 2 as a standard.



**FIGURE 4** Initial orientation of SCE9 in smectic phase for five cells with different dimensions that are listed in Table 2. The photographs (a)–(e) corresponds to (a)–(e) in Table 2, respectively.

See Color Plate XVIII at the back of this issue.



**TABLE 2** Geometrical dimensions and types of grooves and spacers in five cells.

	Grooves		type	Spacers	
	pitch[ $\mu\text{m}$ ]	depth[nm]		pitch[ $\mu\text{m}$ ]	height[ $\mu\text{m}$ ]
(a)	2	2	ribs	150	1.8
(b)	no grooves		ribs	150	1.8
(c)	2	2	dots(no ribs)	150	1.8
(d)	2	2	ribs	300	1.8
(e)	2	2	ribs	150	0.8

In the standard case (Table2 (a)), a better initial orientation was achieved as shown in Figure 4(a). First, grooves were removed from the standard (Table 2(b)). Resultant orientation was radially disturbed as shown in Figure.4(b). Secondly, the type of spacers were changed from rib to dot (Table 2(c)). The orientation was greatly disturbed as shown in lower part of Figure 4(c). It is obvious from the above results that better initial orientations could never be achieved in the cell with either grooves or ribs only .

Thirdly, the pitch of ribs was expanded to 300 $\mu\text{m}$  (Table 2(d)). The orientation became disturbed, despite the presence of grooves, with directors inclined to the grooves at the center point of space between ribs as shown in Figure 4(d). Finally, rib height was lowered to 0.8 $\mu\text{m}$  (Table 2(e)). Radially disturbed orientation like Figure 4(b) was obtained as shown in Figure 4(e). There appeared to be almost no effect with or without grooves. From these results, there were the cases that good orientations were not attained, even if both grooves and ribs were arranged in the cells.

Then we examined the range of the pitch and depth of grooves, and the pitch and height of ribs that provided good orientations for SCE9, and considered the roles of grooves and ribs.

### Roles of grooves

Table 3 shows the results of initial orientations of SCE9 for three values of the depth and pitch of grooves. It was evident that deeper depth and shorter pitch attained better orientation. The desirable depth and pitch were greater than 20nm and less than 2 $\mu\text{m}$ , respectively.

TABLE 3 Initial orientations of SCE9 for three values of, (a) the depth of grooves, (b) the pitch of grooves. The other parameters of the cell were the same as the standard in Table2(a)

(a) Depth of grooves[nm]	Orientation	(b) Pitch of grooves[μm]	Orientation
10	bad	2	good
30	good	4	not good
50	good	8	bad

It is well known that surface shape has considerable effect on nematic orientation<sup>[3-5, 7]</sup>. Berreman<sup>[5]</sup> calculated the difference between elastic energy for molecules aligned parallel to grooves and that for normal to grooves. The difference,  $\Delta F$ , is given by the equation,

$$\Delta F=(\pi^3 K/2\lambda)(2a/\lambda)^2$$

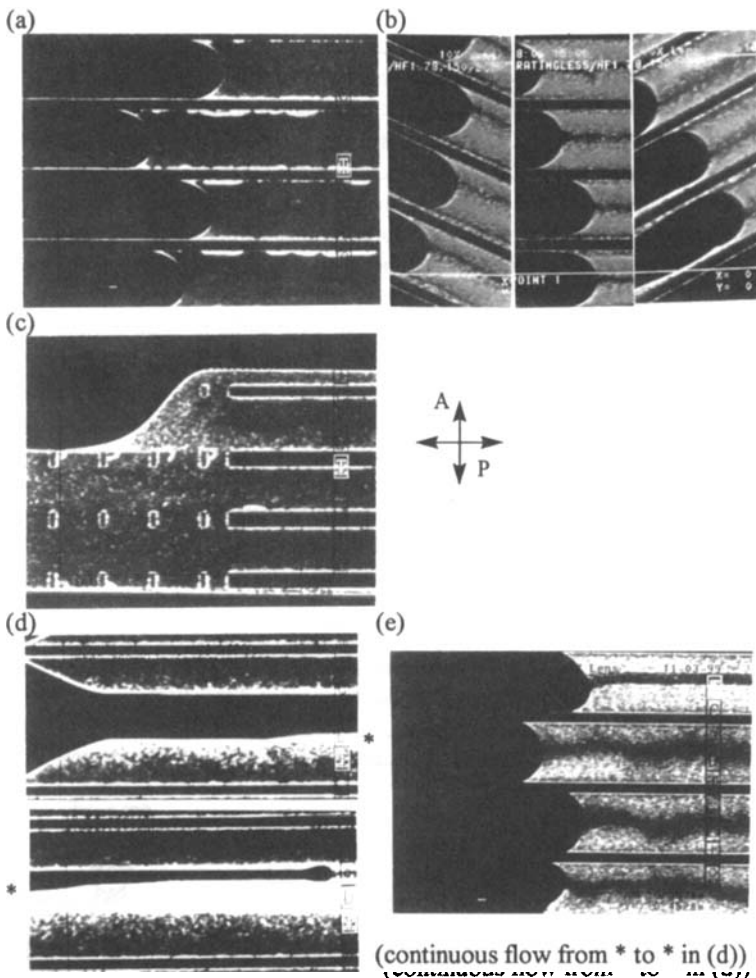
, where  $K$  is an elastic constant,  $a$  is a half of depth,  $\lambda$  is the pitch, and the shape of grooves are described by a sinusoidal function. The elastic energy for molecules aligned parallel to grooves is lower than that for molecules aligned normal to grooves. Present results agreed well with Berreman's equation and a role of grooves is to make directors lie parallel to them.

Yokoyama *et al.*<sup>[6]</sup> proposed the flow-induced orientation mechanism. In this mechanism, anisotropic surface shapes of obliquely evaporated SiO layers promoted LCs to flow to the specific direction. The molecules which orientated to the flow direction were absorbed onto the SiO layers. In the present case, the grooves may also regulate the flow direction, resulting in molecules lying parallel to the flow direction.

Role of ribs: the shape of LC-air boundaries

Better orientation was not achieved in the cell with only the grooves, as noted above, and the presence of ribs was inevitable. We considered this point from observations of LC-air boundaries. Figure 5 shows snap shots of LC-air boundaries of the flowing LC, SCE9, in the nematic phase. The corresponding resultant initial orientations in smectic phase are already

shown in Figure 4.



**FIGURE 5** LC-air boundaries of SCE9 in nematic phase for five cells with different dimensions that are listed in Table 2. The photographs (a)-(e) corresponds to (a)-(e) in Table 2 and Figure 4, respectively.

See Color Plate XIX at the back of this issue.

Figure 5(a) shows the LC-air boundary was kept arched and the directors were parallel to the direction of the polarizer. The direction of the directors was determined to be parallel to the grooves, from the observation of the corresponding orientation in smectic layers, after cooling. With no grooves, as seen from Figure 5(b), though the LC-air boundary took the arched form, the extinction part changed along the arched boundary. The LC molecules were likely to align normal to the boundary in the cell without grooves. In the case that the type of spacers were changed from ribs to dots, or the rib pitch became wide, the LC-air boundary did not take an arched form to exhibit disturbed orientation near the agitated LC-air boundary, as shown in Figure 5(c) and Figure 5(d), even if there were grooves. The cause of these worst orientations is considered to be that LC molecules have a tendency to align normal to the LC-air boundaries, against the effect of grooves.

These disturbed orientations near LC-air boundaries in nematic phase did not change after the cells were filled completely and gradually cooled to room temperature, and then further re-heated to the isotropic phase and gradual re-cooling to smectic phase. Disturbing the LC-air boundary led to the smectic disturbed orientation, because the orientations near the boundary were memorized on the substrates. Thus to control the shape of the LC-air boundary is one of the important roles of ribs.

TABLE 4 Initial orientations of SCE9 for five values of; (a) the pitch of ribs, (b) the height of ribs. The other parameters of the cell were the same as the standard in Table 2(a).

(a)Pitch of ribs[μm]	Orientation	(b) Height of ribs[μm]	Orientation
80	bad	0.8	bad
100	good	1.1	not good
150	good	1.8	good
200	not good	2.4	good
300	bad	2.7	bad

In order to clarify the range of the rib pitches providing better initial orientation, the results of orientations for the five values of rib pitches are shown in Table 4(a). Favorable range of pitch was from 100 to 150μm.

The boundary became disturbed when the rib pitch exceeded  $150\mu\text{m}$ . On the contrary, many cracks appeared for rib pitch less than  $100\mu\text{m}$ . The short pitch of ribs prevents the glass substrate from bending, and thus the volume shrinkage of LCs with decreased temperature is not compensated by substrate bending, giving rise to cracks.

#### Role of ribs: the moving rate of the LC-air boundaries

Figure 5(e) is a snap shot of LC-air boundaries moving into the cell with  $0.8\mu\text{m}$  rib height. As seen, the LC-air boundaries are kept arched, but the orientation near the boundary is radially disturbed as in the case of no grooves (Figure 5(b)). The corresponding initial orientation is shown in Figure 4(e), and also resembles that of the cell without grooves (Figure 4(b)).

Then, initial orientations were examined for five values of rib height as listed in Table 4(b). Better orientations were in the range of  $1.1\text{--}2.4\mu\text{m}$ . A lot of voids were observed for rib height exceeding  $2.4\mu\text{m}$ . The cause of voids is similar to that of cracks as described in the case of narrow rib pitch. The radially disturbed orientation was observed in the cell with rib height less than  $1.1\mu\text{m}$ .

Other examples showing orientations and flowing boundaries similar to Figure 4(e) and Figure 5(e) are observed in the cell with ribs made in a posi-type resist ribs (MP1400 from Shipley) and a photosensitive polyimide (Photoneece from Toray). These results indicated that the role of ribs are not only to control the LC-air boundary.

We then focused on the moving rate of the LC-air boundaries. Table 5 shows the results, indicating that the higher moving rate provided better orientations. Moving rates for the posi-type resist (Table 5(b)) and photosensitive polyimide (Table 5(c)) were slow compared to the nega type resist as a standard (Table 5(a) corresponding to Table 2(a)). The cause of the slow rate in the posi-resist is attributed to worse wettability than nega-resist, as determined by our contact angle measurements. On the other hand, the polyimide has better wettability than the nega-resist. Despite better wettability, better orientation was not achieved in the cell with polyimide ribs. This is because, from the microscope observation, LC molecules moved with a higher rate only along the rib side, but the

arched LC-air boundary moved with slower rate. The nega-type resist possessed appropriate wettability, giving rise to a high moving rate. However, when the cross section of the space was made small by reducing rib height (Table 5(d) corresponding to Table 2(e)), the flow rate became slow on account of high resistance to LC moving.

TABLE 5    Moving rates of the LC (SCE9)-air boundaries in nematic phase for four cells. The other parameters of the cell were the same as the standard in Table 2(a).

	Material of ribs	Height of ribs[μm]	Moving rate[μm/sec]	Orientation
(a)	nega-resist	1.8	18.5	good
(b)	posi-resist	1.8	4.1	bad
(c)	photosensitive polyimide	1.8	7.3	bad
(d)	nega-resist	0.8	5.6	bad

Yokoyama *et al.* noted that in flow-induced orientation on normal evaporated SiO<sub>2</sub>, LCs aligned parallel to the flow direction and a higher flowing rate provided better orientations<sup>[6]</sup>. Our results and the data by Yokoyama indicate that the flow rate is important to orientations in the nematic phase, and one of the roles of ribs is considered to control the flow rate.

CONCLUSIONS

When smectic LCs were introduced during the nematic phase, followed by temperature gradient cooling, excellent smectic orientations were achieved in the cell which had periodical grooves and ribs parallel to the grooves. The required conditions, which may slightly depend on LC materials used, for grooves are that depth is greater than 20nm and pitch is less than 2μm, while for ribs height is between 1.1 and 2.4μm and pitch is 100 and 150μm. The choice of appropriate materials for ribs is very important.

The roles of grooves are considered to be 1) surface shape effect and 2) the control of flow direction. The roles of ribs are to 1) control the

shape of LC-air boundaries and 2) control moving rates of the boundaries into the space between ribs.

The benefits of the present method are not only to achieve excellent orientation beyond that in the cells with rubbed polyimide, but the possibility to use a variety of materials, including inorganic, as an alignment layer.

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