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## Pattern Formations of Smectic-A Domains Grown from an Isotropic Phase in Cano Wedges

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Pattern formation processes and equilibrium shapes of smectic-A domains grown from an isotropic phase in the Cano wedges have been observed in the mixture of octyloxycyanobiphenyl and dodecyl alcohol. The Cano wedges enable us to observe cell thickness ( $L$ ) dependence of the pattern formation. In the region of  $L > 10\mu\text{m}$ , three dimensional growth and equilibrium shapes that have been reported earlier in the  $50\mu\text{m}$ -thick cells were observed. On the other hand, in the region of  $L < 10\mu\text{m}$ , the observed patterns are essentially characterized by plane curves. For instance, circular filaments, which were rarely found for  $L > 10\mu\text{m}$ , developed from buckled filaments were reproducibly observed as equilibrium shapes. In addition, the pattern formation processes in the thickness range of  $1 < L < 4\mu\text{m}$  are found to be different from those of  $L > 4\mu\text{m}$ . The influence of electric-field application to the equilibrium shapes in the Cano wedges is also examined.

**Keywords:** pattern formation; equilibrium shape; smectic-A liquid crystals; plane curves; Cano wedge

### INTRODUCTION

A smectic-A (Sm-A) phase grown from an isotropic ( $I$ ) phase exhibits a rich variety of spatial patterns and there are some geometrically interesting equilibrium shapes of a Sm-A phase in an  $I$  phase such as spheres, tori and circular filaments<sup>[1-3]</sup>. In addition, thermo-temporal evolutions of a Sm-A phase are also interesting examples of self-organization processes<sup>[4,5]</sup>. Thus the pattern formation of Sm-A domains has attracted much attention.

We have studied time evolution and equilibrium shapes of Sm-A do-

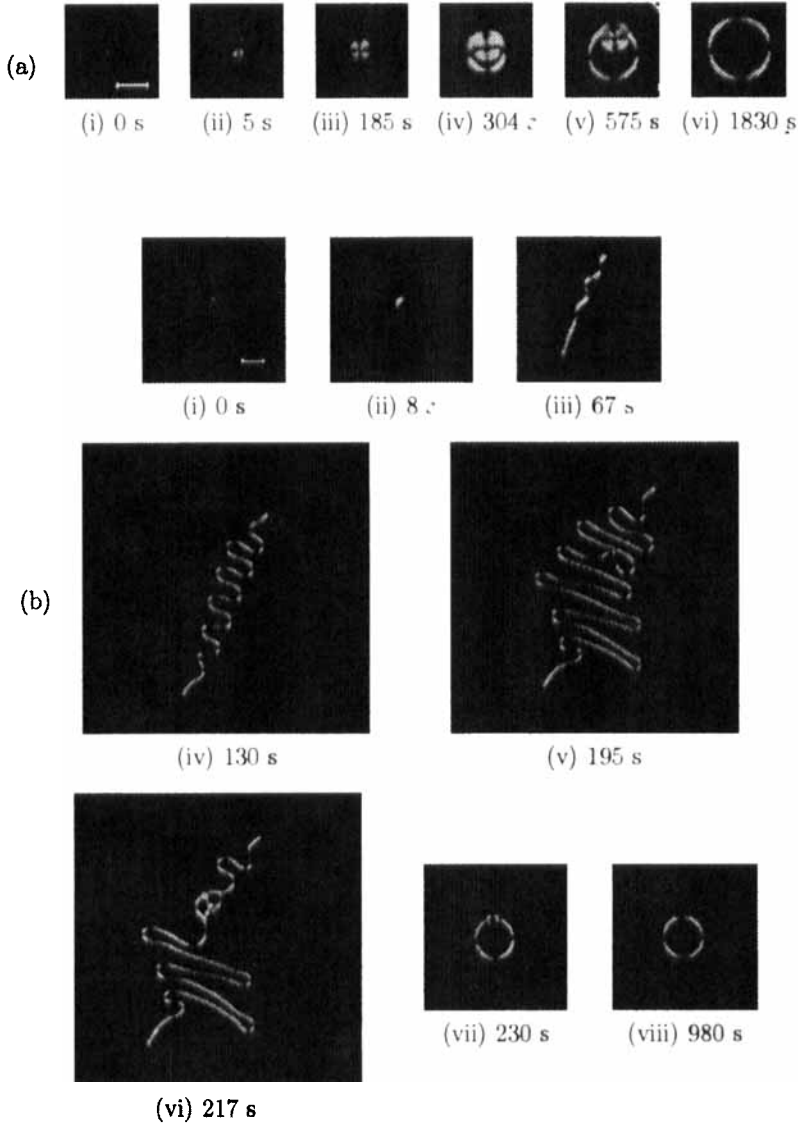


FIGURE 1 continued

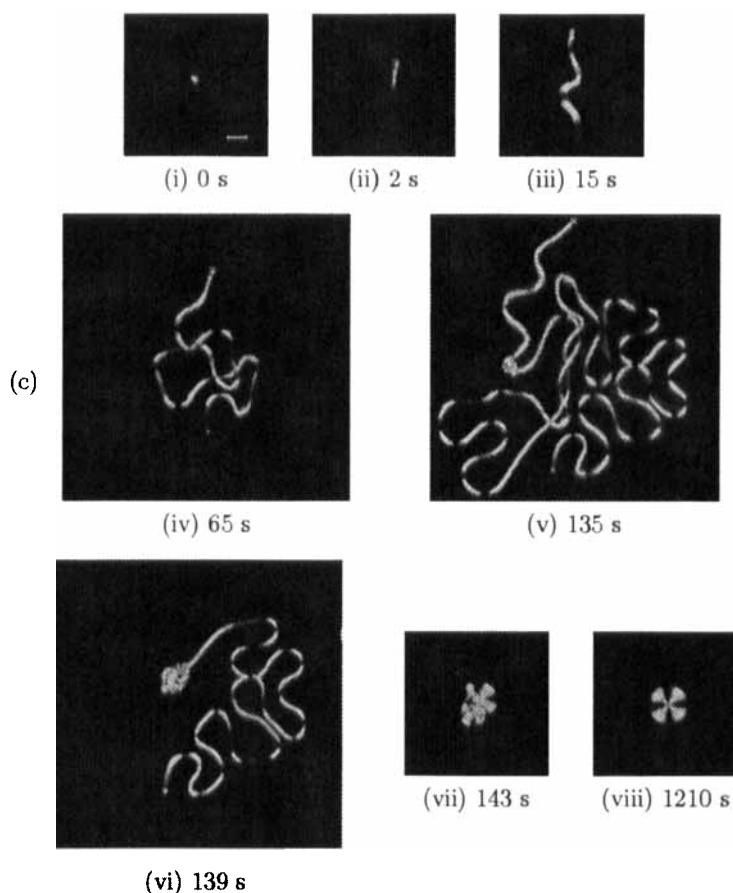


FIGURE 1 Growth sequence of Sm-A domains from an *I* phase in the mixture of 8OCB and DODA (the molar concentration of 8OCB is 40 %) for different thickness regions: (a)  $1 < L < 4\mu\text{m}$ , (b)  $4 < L < 10\mu\text{m}$  and (c)  $10 < L < 15\mu\text{m}$ . The cooling rate from the *I* phase was  $-0.2^\circ\text{C}/\text{min}$ . The bars indicate  $20\mu\text{m}$ .

mains grown from an *I* phase both theoretically and experimentally, and have derived the shape equations for Sm-A filaments<sup>[5]</sup>. The shape equations have successfully explained the patterns of plane-curve Sm-A filaments. However, such plane-curve Sm-A filaments were difficult to reproducibly observe in cells whose thickness is about  $50\mu\text{m}$  (such cells have

been often used for the observation of patterns in Sm-A LC's<sup>[1,4]</sup>). It is expected that the plane-curve filaments become observable with reducing cell thickness. However, the influence of cell thickness on the pattern formations of a Sm-A phase in an *I* phase has not been studied yet.

In this paper, we report on the cell thickness (*L*) dependence of pattern formations and of equilibrium shapes using the Cano wedges. We find that the pattern formation processes of a Sm-A phase are greatly influenced by cell thickness: the three-dimensional growth patterns are changed into the two-dimensional patterns, characterized by plane-curves, with decreasing cell thickness. We also report on the influence of the electric-field application on the equilibrium shapes observed at different cell thicknesses.

## EXPERIMENT

The LC material used here was the binary mixture of octyloxycyanobiphenyl (8OCB) and dodecyl alcohol (DODA) whose phase diagram has been reported<sup>[1]</sup>. For a molar concentration (>20 %) of DODA, the nematic phase is suppressed and the *I* and Sm-A phases coexist over a fairly wide temperature range (for example, 30–42 °C for 60 % of DODA studied here). Wedge-shaped cells with a 10mm×10mm active region were constructed and filled with the mixture of 8OCB and DODA. A 16μm spacer was inserted into one end of the cell while on the other side, the glass plates were in contact. The length of the active region and the distance from the point of contact to the 16μm spacer were measured. The thicknesses were found to vary from 1 to 15μm using geometrical calculations. The polyimide layers were coated on the surfaces of the glass plates for homogeneous alignment. To apply external voltage to the wedge-shaped cells, indium-tin oxide (ITO) precoated glass plates were used. The sample temperature was controlled using a hot stage with an accuracy of ±0.002 °C (Instec HS1-i). Pattern formations of Sm-A domains were observed with a polarizing microscope (Nikon X2TP-11) equipped with a color video camera (Sony DXC-107A).

## RESULTS AND DISCUSSION

### Growth processes

The cells with 40 % of 8OCB were cooled from the *I* phase at -0.2 °C/min, and the cooling was stopped at 36.0 °C in the coexisting region of the Sm-

A and I phases for the observation of growth processes. Figure 1 shows the growth sequence of Sm-A domains from an I phase in the mixture of 8OCB and DODA for three different thickness regions.

In all the thickness regions, Sm-A phase first appears in the form of droplets on cooling the I phase of the mixture. In the region (a), after the droplets attain a radius comparable to the cell thickness, the droplets transform into disk-like form of Sm-A domains. When the radii of the disk-like domains become  $20\text{--}30\mu\text{m}$ , they change into circular filaments. In the region (b), after the droplets attain a radius of  $3\text{--}4\mu\text{m}$ , they start elongating and form filaments with a constant radius of  $\sim 3\mu\text{m}$ . During growth, the filaments continuously buckle and then become wavy. The filaments are metastable, and eventually transform into circular filaments. In the region (c), after the droplets attain a radius of  $3\text{--}4\mu\text{m}$ , they start elongating and form filaments, as observed in the region (b). The filaments continuously buckle to take on a serpentine form and subsequently become convoluted. The filament finally collapses forming compact domains<sup>[1-5]</sup>. We note that the Sm-A filaments observed in the region (b) and (c) grow rapidly in length but not in diameter.

It is evident that in the region (c), the three dimensional growth is observed while in the regions (a) and (b), the growth patterns are essentially characterized by plane curves. It is also evident that the growth pattern in the region (a) is greatly different from that in the regions (b) and (c) in the sense that the buckled filaments are not formed and, instead, disk-like texture is observed. In addition, the growth velocity of the disk-like texture in this region is much slower than that of the filaments in the regions (b) and (c).

We note that the growth sequence and the equilibrium shape of Sm-A domains are essentially the same for two different substrates: we examined the effect of the alignment layers on the growth sequence and the equilibrium shapes, and found no difference in the shapes between polyimide alignment layers and ITO precoated substrates. This indicates that the contribution of the anchoring of the Sm-A domains is negligible in the present study.

### **Equilibrium shapes**

As shown in Figure 1, the equilibrium Sm-A domains are circular filaments for  $1 < L < 10\mu\text{m}$  and concave forms for  $L > 10\mu\text{m}$ . In addition to these single domains, we observe the new domains that consist of the two circular filaments for  $1 < L < 10\mu\text{m}$  and of the two concave domains for  $L > 10\mu\text{m}$ . These observation is shown in Figures 2(d)-2(f), together with

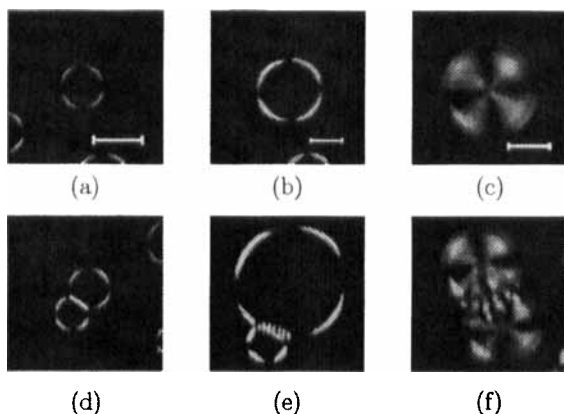


FIGURE 2 The equilibrium shapes of Sm-A domains in an *I* phase at three different thickness regions in the wedge-shaped cell, observed with the analyzer and polarizer crossed (a)–(f). The molar concentration of 8OCB is 40 % in the mixture of 8OCB and DODA, and the observation is carried out at 36.0 °C. Single Sm-A domains and contacted Sm-A domains are shown in (a)–(c) and (d)–(f), respectively. (a),(d)  $1 < L < 4\mu\text{m}$ , (b),(e)  $4 < L < 10\mu\text{m}$ , (c),(f)  $10 < L < 15\mu\text{m}$ . The bars indicate  $20\mu\text{m}$ .

the single domains (Figures 2(a)–2(c)). It is interesting to note that the domains in Figures 2(d)–2(f) are just in contact with each other and do not fuse. This will be obvious from the experimental results, shown in the next subsection. We also note that the texture in the contacted area for  $1 < L < 4\mu\text{m}$  is the same as that for  $4 < L < 10\mu\text{m}$ , although the growth processes in these cell thickness ranges are quite different, as shown in Figures 1(a) and 1(b) (high magnification observation shows that the feature of the contacted area in Figure 2(d) is similar to that in Figure 2(e)). It is evident from the observation that in the range of  $1 < L < 10\mu\text{m}$ , the director distributions are changed in the contacted area, reflecting the Sm-A layer undulation. It is likely that in the equilibrium shapes of Figures 2(d)–2(f), the increase in the elastic energy due to the Sm-A layer undulation is compensated by the decrease in the Sm-A/Sm-A interface energy.

We have developed a theory of equilibrium shapes of Sm-A domains in *I* phase: we derived the shape equations for the equilibrium shapes by the variation of the sum of the volume free energy change due to *I*-Sm-A transition, the surface energy of Sm-A/*I* interface, and the curvature elastic energy of Sm-A domains with respect to either the normal direction of



the surfaces of the compact domains or the normal and binormal direction of the filaments<sup>[3,5]</sup>. The shape equations have successfully explained the equilibrium shapes of Sm-A domains such as circular filaments (Figure 2(a) and 2(b)) as well as compact domains (Figure 2(c)). In addition, the shape equations have predicted the features of the growth pattern of the filaments that are essentially characterized by plain curves. For instance, as shown in Figure 1(b)(iv), a feature that both ends of a buckled filament are straight can be predicted<sup>[5]</sup>. However, the reproducible observation of such plain curves has been rather difficult in cells whose thickness is about 50  $\mu\text{m}$ . We note that the continuous variation in cell thickness using the wedge-shaped cells enables us to identify the influence of the cell thickness on the pattern formation processes of Sm-A LC's in *I* phase. The experimental results of plane-curve Sm-A phase observed in the thickness range of  $1 < L < 10 \mu\text{m}$  would be very important for further theoretical studies.

#### **Influence of electric field application on the equilibrium shapes**

We examine here the effect of electric-field application on the equilibrium shapes shown in Figures 2(d)-2(f) (no changes in the equilibrium shapes shown in Figures 2(a)-2(c) were induced by the electric-field application). For this purpose, the polyimide layers were coated on the surfaces of the ITO precoated glass plates for electrical insulation as well as homogeneous alignment. Careful electrical insulation is necessary near the point of contact in the wedge-shaped cells.



FIGURE 3 Equilibrium Sm-A domains in an *I* phase at 36.0°C observed with the analyzer and polarizer crossed ( $L = 5\mu\text{m}$ ) before (a) and after electric-field ( $2.0\text{V}/\mu\text{m}$ ) application (b). The bar indicates  $20\mu\text{m}$ .

The square-wave voltage (100Hz) was applied to the wedge-shaped cells. Above certain threshold voltages, the contacted domain is separated into two single domains as shown in Figure 3 (we show the results at  $L = 5\mu\text{m}$  only, but the same phenomena are observed in the other thickness

regions). This observation demonstrates that the Sm-A domains shown in Figure 3(a) are just in contact with each other. The threshold electric-fields at  $L = 2, 5, 9$  and  $13\mu\text{m}$  are 2.7, 2.0, 1.2 and  $1.1\text{V}/\mu\text{m}$ , respectively.

The interpretation of the equilibrium shapes in Figures 2(d)-2(f) and their electric-field induced separation in Figure 3 is an interesting theoretical issue. We consider that these phenomena would also be explained in terms of the theory mentioned in the previous subsection by taking account of Sm-A/Sm-A interface energy and/or electric-field induced dipole-dipole interaction.

## CONCLUSIONS

We have studied the influence of cell thickness on the pattern formation processes in the binary mixture of 8OCB and DODA using the Cano wedges. Three dimensional growth of Sm-A filaments and subsequent formation of compact domains on the substrate surfaces are observed in the cell thickness range of  $L > 10\mu\text{m}$ . These observation is consistent with that reported earlier. In the cell thickness range of  $4 < L < 10\mu\text{m}$ , the two dimensional growth of Sm-A phase (the observed patterns are characterized by plane curves) is observed. The equilibrium shapes in this range are circular filaments. Such plane curves of Sm-A phase has been difficult to observe reproducibly in cells whose thickness is about  $50\mu\text{m}$ , and are good examples for the demonstration of our theory developed for filamentary growth of soft matters. The same equilibrium shapes are found in the cell thickness range of  $1 < L < 4\mu\text{m}$  but their growth process is completely different from that in the cell thickness range of  $4 < L < 10\mu\text{m}$ . We also find that the two circular filaments are in contact with each other, and that the contacted circular filaments are equilibrium shapes of Sm-A phase and are separated by the application of electric field to the cell.

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