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### Some Mechanical Treatments for Alignments of Smectic Liquid Crystals

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# Some Mechanical Treatments for Alignments of Smectic Liquid Crystals

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We show the results on the planar alignment of chiral smectic liquid crystals made by mean of three different mechanical treatments. The orientational processes of these technics are explained on the basis of the orientation induced by a planar shearing of the samples. In addition we report on interesting realignment effects induced dynamically by the changing of the frequency of the sample thickness vibration.

**Keywords:** *smectic l.c., alignments of l.c., uniaxial stresses, chiral smectic l.c.*

## INTRODUCTION

For scientific investigations and for practical applications, it is very important to obtain good alignments of smectic liquid crystals. Contrary to nematics, this problem cannot be solved generally only by surface treatments. For this reason in smectics other treatments were investigated intensively. The advantages and limits of these different alignment technics were summarized e.g. in References 1, 2.

Recently a special shear method<sup>2</sup> was worked out which provided good planar alignment in a broad range of sample thickness (up to  $100 \div 150 \mu\text{m}$ ) with mono-domain areas of about one square centimeter.

According to this method one can obtain homogeneously planar aligned smectic liquid cells by applying a shear at the isotropic-smectic A, or cholesteric-smectic A phase transition temperature, when the smectic droplets (so called “batonnets”) are freely floating in the isotropic or cholesteric fluid.

The optimum displacement was found to be in the order of sample thickness. In that experiment, shearing was produced by connected one of the bounding plates to a membrane of a loudspeaker, while the other plate was fixed. The loudspeaker was excited by a square wave of few Hz-s.

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As an extension of this method we investigated other types of mechanical treatments. These can be divided into three groups.

a.) Induced sample thickness vibrations at the phase transition temperature of the highest temperature smectic phase  $T_{tr}$ , in the highest temperature smectic phase, below  $T_{tr}$ .

b.) Uniaxial static stresses (dilative and compressive as well) at  $T_{tr}$  and below  $T_{tr}$ .

c.) Static and alternating bending of the bounding plates at  $T_{tr}$  and below  $T_{tr}$ .

Our results can be regarded as a check of statements of Reference 2, and provide useful simple alignment technics in different geometries. These results can be useful if the shear is not applicable, or if we want planar alignment with e.g. radial layer distribution.

Furthermore, we report on an interesting dynamical realignment effect which appeared in high amplitude sample thickness vibration above a certain frequency threshold.

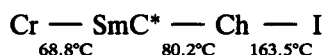
## EXPERIMENTAL

### Materials

For our experiments we used the following two liquid crystal compound.

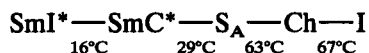
1. (+)—4 — 6-alkyloxy phenil 4-(2"-methilbutyl) biphenyl 4'-carboxilate. This is a BDH<sup>3</sup> product and designated CE3.

Its phase sequence is<sup>3</sup>:



as described by the procedures.<sup>3</sup> Within the measuring errors we observed the same phase transition temperatures.

2. A binary mixture called FK4 synthetized in the Central Research Institute for Physics in Budapest. This is made by two compounds of 4- (2'-methylbutyloxy)-phenil-4-alkyloxy-benzoate homologues (MBOPEnOBA) containing 60% by weight of the compound with  $n = 8$  and 40% of  $n = 12$ . The phase sequence of FK4 is the following<sup>4</sup>:



## EXPERIMENTAL SET-UP

In case of a.) and b.) type investigations the bounding glass plates were mounted in a Fabry-Perot like holder with no spacer (see Figure 1). The cell was similar to

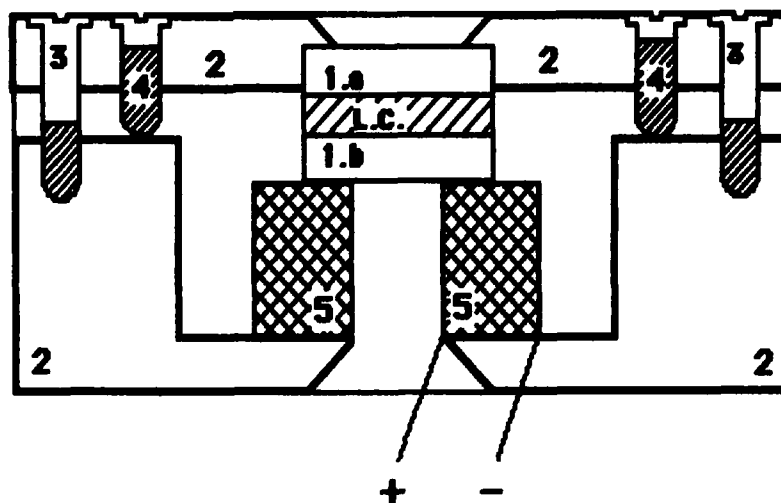


FIGURE 1 Schematic of the sample holder used in experiments type a.) and b.).

1) Bounding glasses; 2) Sample holder from metal; 3) and 4) Steel screws for setting the sample thickness; 5) P4-68 type piezoelectric ceramics.

that used in the measurement of elasticity in  $S_A$ ,<sup>5</sup> thus a system of six steel screws ensured the rigidity of the holder and controlled the sample thickness.

The sample were heated by electric current applied on the  $\text{SnO}_2$  electrodes, and visually observed by a microscope. There is a temperature gradient in the sample. At boundary edges the sample was found to be colder by about  $0.5^\circ\text{C}$  than in the centre. For the sample thickness variation a sequence of 40 pieces of P4-68 type piezoelectric ceramic joined together mechanically in series and electrically in parallel.

The parallelism of bounding plates were controlled better than  $10^{-3}$  rad.

The sample thickness is initially determined by Mylar spacers which were than removed.

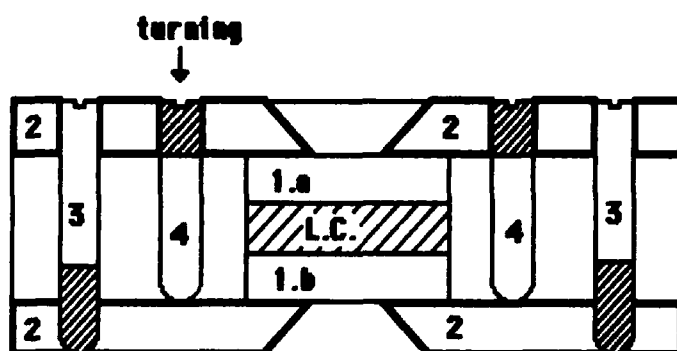


FIGURE 2 Schematic of sample holder used in the experiment type c.).

1) Bounding glasses; 2) Sample holder from metal; 3) and 4) Steel screws for setting the sample thickness and changing the parallelism.

In the a.) type measurements the sample thickness was  $d = 100 \pm 5 \mu\text{m}$ . At such large sample thicknesses the elastic reaction of the holder is negligible and the applied displacement is proportional to the applied voltage being equal to the free-space displacement of ceramic.<sup>6</sup> At the temperatures where we made the measurements  $\pm 100 \text{ V}$  on the ceramics corresponded to  $\pm 2.2 \mu\text{m}$  sample thickness variation. The precise value of the electric voltage-displacement coefficient is given by producer ("Quartz & Silice" in France) and was not controlled by us.

In the measurement b.) we used samples with thickness  $d = 10 \pm 2 \mu\text{m}$ . In such sample thickness the displacement due to the applied voltage on the ceramics is very small,<sup>6</sup> and the applied voltage  $U$  is proportional to the force  $F$  applied on the sample. From the data given by the producer:

$$F/U = 6.7 \text{ N/V}.$$

For the purpose of measurement c.) we used an experimental set-up which is similar to the one sketched in Figure 1, but without ceramics. The parallelism of the bounding plates was changed by turning a little back and forth one of the screws (see Figure 2).

The temperature control and the observation methods was similar to the one used in the former two cases.

## RESULTS

Without mechanical influences the CE3 sample cooled from the cholesteric phase formed a roughly homeotropic  $\text{SmC}^*$  phase, while the FK4 sample cooled from the cholesteric formed a focal conic  $\text{S}_\text{A}$  phase.

### I. PLANAR ALIGNMENT TREATMENTS

#### a. Sample Thickness Vibration

Applying a square-wave voltage  $U = 50 \div 100 \text{ V}$  with  $f = 1 \div 20 \text{ Hz}$  on the ceramic while cooling down slowly (c.c.a.  $20^\circ\text{C/h}$ ) the sample from Ch to  $\text{SmC}^*$  (or Ch to  $\text{S}_\text{A}$ ), always planar samples were created with quasi radial symmetry in the orientation.

A typical result of this process can be seen on Figure 3. This picture represents a  $0.35 \times 0.25 \text{ mm}^2$  area of the CE3 sample with thickness  $d = 100 \mu\text{m}$ . The parallel stripes—representing the helical structure—are perpendicular to the helical axis.

The orientation of the layers is parallel to the direction of flow, generated in a transient during the strain application because of the incompressibility (constant volume) of the liquid crystal. In our experiment we used samples where the bounding plates were square shaped with the side length of 2 cm.

Generally the following layer field was created due to the above mentioned process (see Figure 4).

In the middle of the sample a homeotropic stripe remained with a width is about  $30 \mu\text{m}$  and a length is about 0.5 cm (cross sectioned stripe on the Figure 4).

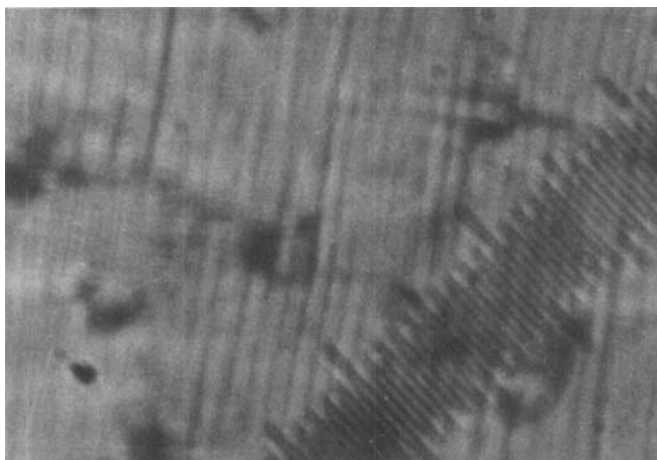


FIGURE 3 Typical result of the alignment by sample thickness vibration at  $T_{tr}$ . The picture represents a  $0.35 \times 0.25 \text{ mm}^2$  area of the CE3 sample with thickness  $d = 100 \text{ }\mu\text{m}$  at  $T = 77^\circ\text{C}$ . The parallel stripes are perpendicular to the helical axis and the parallel to the flow velocity. On the right lower part of picture a scale can be seen. See Color Plate I.

In the other parts the sample was planar. The parallel stripes on the figure show the direction of smectic layers. The area of the largest monodomains was typically  $1 \text{ cm}^2$ .

On the basis of the ideas presented in Reference 2 we can explain the orientation mechanism as follows. In case of  $d = 100 \text{ }\mu\text{m}$  sample thickness with the area of the bounding plates  $A = 4 \text{ cm}^2$  the sample thickness variation, ranged between

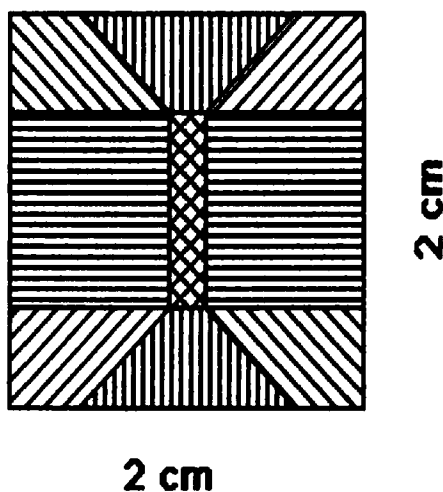


FIGURE 4 Schematic representation of the created smectic layer field due to the sample thickness vibration (upper view). In the middle of the sample the homeotropic stripe remained (cross sectioned stripe). In the other part the sample is planar. The parallel stripes show the direction of smectic layers.

$\delta = 1 \mu\text{m}$  and  $\delta = 2 \mu\text{m}$ , causes a Poiseuille flow normal to the thickness variation with an amplitude of  $S = 100 \div 200 \mu\text{m}$ . This is in the order of sample thickness  $d$ , which was found to be the optimal shear amplitude in Reference 2.

Due to symmetry conditions there is no flow in the centre of the sample, therefore (in the case of CE3) homeotropic alignment occurs (in case of FK4 the centre of the sample was found to be in focal conic state).

To explain the observed alignment profile, we say that it is determined by the symmetry of sample geometry: partly due to surface tension of substance at bounding plate edges, and mainly due to the temperature profile. As the temperature was the coldest at the edges, the smectic phase grows from starting at edges. According to the aligning effect of cooling from spacer edge (epitaxial growth<sup>7</sup>) the smectic layer direction was determined not only by the flow direction,<sup>2</sup> but was influenced also by the direction of the temperature gradient.<sup>7</sup>

On the basis of this explanation we guess that—in case of cylindrical bounding plates—radial planar alignment would be established (except of the centre). In some applications this alignment would be very useful.

We investigated the aligning effect of the sample thickness vibration below  $T_{tr}$  also. In this case initially homeotropic  $\text{SmC}^*$  (or focal conic  $S_A$ ) was created in the whole volume of the sample. The above mentioned vibration process was found to be effective in a similar way also in this case.

Due to the vibration, at locations where the homeotropic alignment was not perfect, planar alignment was getting to be established. After about 15 minutes more than a half of the sample was in planar state. However by this process we never got perfect planar alignment even after one hour vibration.

This observation allows us to compare the effect of shearing below  $T_{tr}$  (this method was reported first by N. A. Clark and S. T. Lagerwall<sup>8</sup>) and at  $T_{tr}$  (as reported in Reference 2). Our results show, that the mechanical treatments at  $T_{tr}$  are much more effective than below  $T_{tr}$ .

The effect of sample thickness vibration  $3^\circ\text{C}$  below  $T_{tr}$  is shown on Figures 5 for CE3.

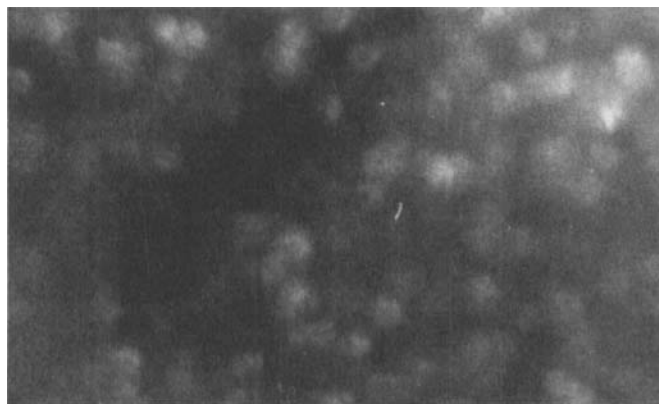
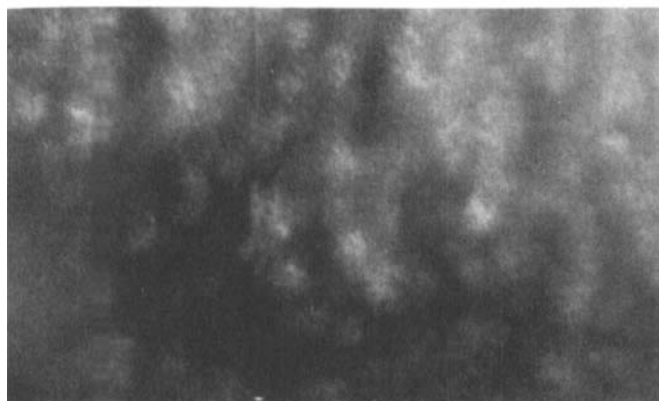
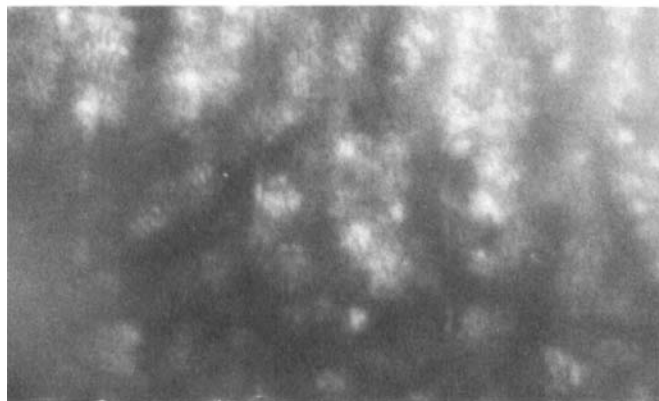
Figure 5a shows the situation at the beginning of the thickness vibration (with the conditions  $f = 1 \text{ Hz}$  and  $\delta = \pm 2 \mu\text{m}$ ); the result of this process after 5 minutes (and after 10 minutes) is shown in Figure 5b (and on Figure 5c).

We note that the sample thickness vibration induces planar alignment in the cholesteric phase also. Several minutes treatment induces broken planar textures in the whole sample, which become more homogeneous after letting the sample to relax. The result of this process is shown on Figure 6.

## b. Static Stresses

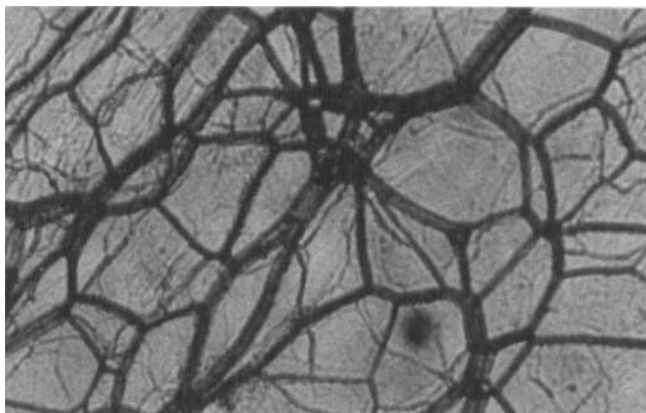
According to the Le Chatelier-Brown principle in any system the response to an external influence is such that the effect of external influence should be decreased by producing an appropriate rearrangement of the system.

In case of smectic liquid crystals the effect of external stress is decreasing if the sample forms a planar alignment due to the flow in the layers. On the basis of this idea we can suppose, that under the influence of static stresses (compressive or dilative) the planar alignment is preferable for the smectic liquid crystals.

**Fig. 5a****Fig. 5b****Fig. 5c**

**FIGURE 5** Representation of the process of the sample thickness vibration at  $T = 75^\circ\text{C}$  ( $T_v = 80.2^\circ\text{C}$ ) of the sample CE3 with sample thickness  $d = 100\mu\text{m}$ . **a)** The homeotropic alignment at the starting time of the vibration process. **b)** The texture with increasing planar part (white places) after 5 minutes of sample thickness vibration with the amplitude of  $\delta = 2\mu\text{m}$  and frequency of  $f = 1\text{ Hz}$ . **c)** The texture after 10 minutes of the sample thickness vibration. The pictures represent  $0.25 \times 0.2\text{ mm}^2$  areas. See Color Plate II.

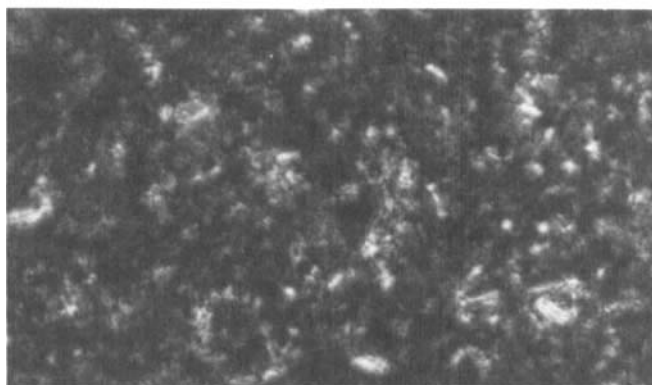




**FIGURE 6** Broken planar texture of cholesteric phase of CE3 after half an hour of a ten minutes duration sample thickness vibration treatment ( $\delta = 2 \mu\text{m}$ ,  $f = 3 \text{ Hz}$ ). The picture shows an  $0.35 \times 0.25 \text{ mm}^2$  area. See Color Plate III.



**Fig. 7a**



**Fig. 7b**

**FIGURE 7** Representation of the result of the static press on a  $d = 10 \mu\text{m}$  thickness film of CE3. **a)** The homeotropic sample at the  $\text{Sc}^*$  phase ( $T = 74^\circ\text{C}$ ) created without any external treatment. **b)** Focal conic  $\text{Sc}^*$  sample at  $T = 75^\circ\text{C}$  appeared due to the  $p = 10^5$  Pascal press applied while the sample cooled down from the Ch phase to the  $\text{Sc}^*$  phase. See Color Plate IV.

We investigated the influence of static stresses up to  $p = \pm 2 \times 10^6 \text{ N/m}^2$  without any surface treatments. The pressure was determined by measuring the force was imposed externally on bounding plate.

In case of CE3 during the Ch-SmC\* phase transition the stress caused a focal conic sample (instead of the homeotropic one). The influence of static stress  $p = 10^6 \text{ Pascal}$  is represented in Figures 7a and 7b.

In case of FK4 the applied stress at the Ch-S<sub>A</sub> phase transition created a focal conic texture, similarly to the case without external influences. However due to the stress the average size of the monodomains increased to about  $5 \div 6$  times larger dimension.

The effect of stress and dilation was found to be similar.

We think that applying a static stress combined with surface treatments (i.e. making some direction to be preferred) this treatment would provide a better homogeneous planar alignment. However, we found in practice that dynamical treatments are more effective.

### c. Alternating Bending

Because of the constant volume of the liquid crystal when changing the parallelism of the bounding plates a flow occurs in the direction of the sample thickness gradient.

Varying alternately the parallelism (e.g. simply by a screw driver) so that a amplitude of flux was in the order of sample thickness a homogeneous planar alignment was created. A typical alignment obtained by this method is shown in Figure 8. (CE3 sample with the sample thickness  $d = 20 \mu\text{m}$ ).

If the amplitude of the induced flow was much larger than the sample thickness, mainly homeotropic alignment occurred. (Compare this fact to the statements of Reference 2).

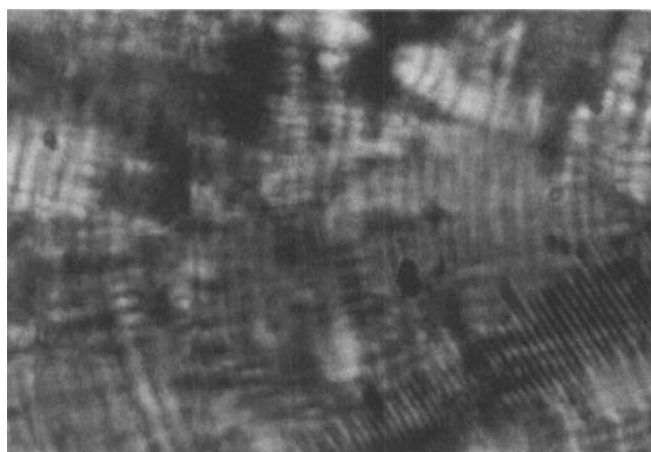


FIGURE 8 Planar CE3 liquid crystal with  $d = 20 \mu\text{m}$  sample thickness in Sc\* phase at  $T = 72^\circ\text{C}$ . The sample was aligned by alternating bending during the Ch-Sc\* phase transition. The stripes are due to the helical structure of the material (the stripes are parallel to the bending direction). The scale in the left hand side of the picture shows that the photo was taken from a  $0.2 \times 0.35 \text{ mm}^2$  area. See Color Plate V.

## II. REALIGNMENT EFFECT DUE TO THE SAMPLE THICKNESS VIBRATION AT HIGH FREQUENCIES

We investigated the frequency dependence of the sample thickness vibration on the alignments. The following behaviour was found for different ranges of amplitude of thickness variation  $\delta$  and vibration frequency  $f$ .

a.  $\delta < 0.9 \mu\text{m}$  and any frequency

- the effect of vibration on the alignment was very weak at all frequencies.

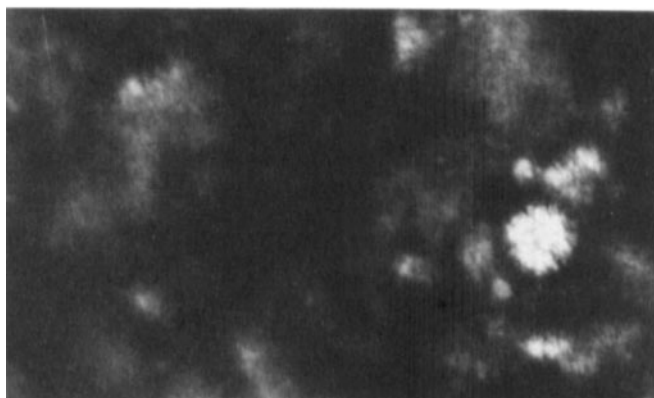
b.  $1 \mu\text{m} < \delta < 2 \mu\text{m}$  and  $f < 20 \text{ Hz}$

- the planar alignment was preferred;

$f > 20 \text{ Hz}$



**Fig. 9a**



**Fig. 9b**

**FIGURE 9** Realignments due to sample thickness vibration. **a)** Result of the sample thickness vibration of amplitude  $\delta = 2 \mu\text{m}$  and of frequency  $f = 100 \text{ Hz}$ . Intensive irregular shape flows occurred. ( $T = 75^\circ\text{C}$ ,  $d = 100 \mu\text{m}$ ,  $0.25 \times 0.35 \text{ mm}^2$  area) **b)** Result of the sample thickness vibration of amplitude  $\delta = 2 \mu\text{m}$  and of frequency of  $f = 4.2 \text{ kHz}$ : as a result of the vibration of homeotropic alignment occurred from the planar one. ( $T = 75^\circ\text{C}$ ,  $d = 100 \mu\text{m}$ ,  $0.25 \times 0.35 \text{ mm}^2$  area). See Color Plate VI.

- the alignment practically was not disturbed;

c.  $\delta > 2 \mu\text{m}$  and  $f < 20 \text{ Hz}$

- the planar alignment was preferred;

$$20 \text{ Hz} < f < 4 \text{ kHz}$$

- irregular shape flows occurred (see Figure 9a);

$$f > 4.2 \text{ kHz}$$

- homeotropic alignment was created again (see Figure 9b).

This alignment was found to be stable: decreasing the frequency, or switching off the vibration the homeotropic alignment remained stable.

At  $f = 4.2 \text{ kHz}$  the realignment was very fast ( $1 \div 2$  seconds) suggesting that at this frequency there is an inner resonance of the system material, glasses + oven A.S.O.

This surprising effect was not observed earlier e.g. during the investigation of shear on the alignment. The reason is probably that in earlier works (e.g. Reference 2) an amplitude of  $100 \mu\text{m}$  at  $f = 4 \text{ kHz}$  was impossible to be produced.

We note that similar realignment effect was found by us also in cholesteric phase.<sup>9</sup>

In Figure 10 we report qualitatively a sketch of the reorientation processes we observed. The transitions between different regimes have been not investigated, and they are only qualitatively putted to better understand.

For the time being we cannot give a detailed theoretical explanation of this effect. We believe that this realignment is a close connection with the viscoelastic behaviour of the smectic layers; until about  $20 \text{ Hz}$  the smectic layers response as a viscous liquid. (That is why the planar alignment is preferred at low frequencies).

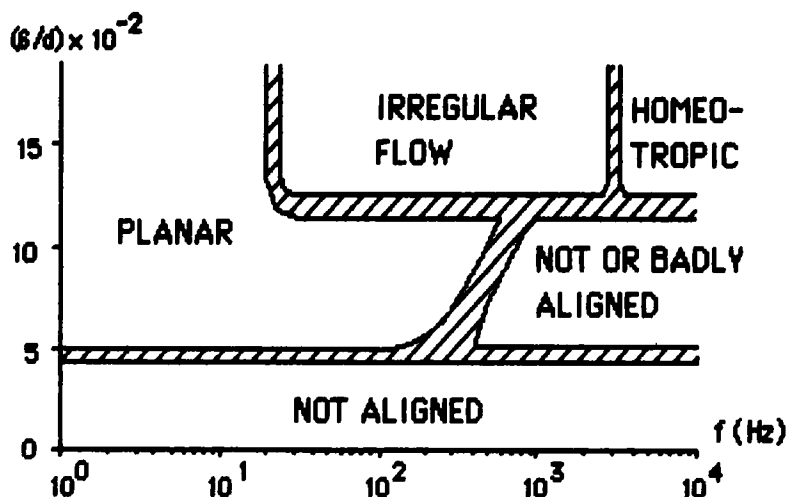


FIGURE 10 Schematic sketch of reorientation process.

As the frequency increases the smectic layers behave more and more rigidly, thus at high frequencies energetically there is no difference between the homeotropic and planar alignment.

(The homeotropic alignment is preferred in this case, because due to the surface interactions without mechanical treatments the material tends to be in homeotropic state).

### Acknowledgment

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