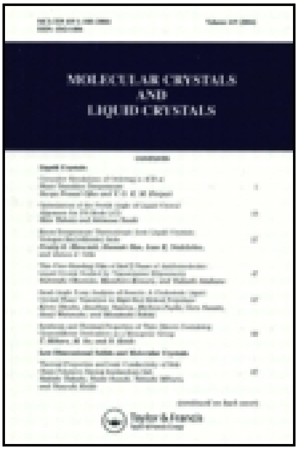

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A Novel Method for Study of a Smectic Phase Obtained from a Strongly-Deformed Nematic Texture

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A NOVEL METHOD FOR STUDY OF A SMECTIC PHASE OBTAINED FROM A STRONGLY-DEFORMED NEMATIC TEXTURE

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Abstract A simple method for the growth of conventional or reentrant smectic phases from strongly-deformed conventional or reentrant nematics, is proposed. It permits the formation of surface-induced and surface-governed conventional and/or reentrant smectic textures which can be studied from the scientific point of view or can be applied in the laser-addressed thermo-optic smectic liquid crystal storage displays for the creation of static figures. The simple method proposed might be applied for study of many smectics arrising either from conventional or reentrant nematic phases under cooling or heating.

INTRODUCTION

The various smectic (Sm) phases are usually obtained and investigated after a slow cooling of the well-aligned nematic (N) phase (in the cases when it exists) down to the corresponding Sm phase. On the other hand, they can be created after slow or rapid (free) cooling of previously-deformed N films. In this way, Cladis and Torza have obtained Sm A textures from initially bent N CBOOA [1] and related their scattering properties with the well-known Kahn's thermo-optic display devices. [2] The influence of the initially-deformed N phase, the surface treatment, the sample thickness and the cooling rate on the type of the Sm scattering textures have been also investigated by Chu and Jacobs. [3] Some problems related to the creation of the Sm A phase from an initially-deformed N phase were discussed by Grinstein and Pelcovits as well. [4] Recently, the method of Cladis and Torza [1] was extended in a simple manner by Hinov who proposed a growth of the Sm phase after slow or rapid cooling of a N with strongly-deformed surface regions and an oriented rest part of the liquid crystal (LC) layer under study[5, 6, 7]. The advantages of this method for a technical application in the thermally-addressed LC displays were also stressed[8].

Our aim is to emphasize on the possibilities of this general and simple method which permits not only the obtaining of very nice and homogeneous Sm scattering textures [5-7] but also can be elaborated for the case of the novel reentrant LCs, the first one obtained by Cladis. [9] The rich polymorphism discovered in a number of conventional or reentrant LCs can also be taken into account . In the first part of the paper we recall the method for the case of conventional LCs and especially emphasize on the LCs with a rich polymorphism. In the second part we extend our method for the case of the reentrant LCs. In the third part we discuss a number of questions which now are open for explanation and give some our qualitative ideas for some of them. The method is illustrated by photos of Sm scattering textures. The possible technic application of this method is discussed in the end of the paper.

A METHOD FOR STUDY OF CONVENTIONAL SMECTIC PHASES OBTAINED FROM A STRONGLY-DEFORMED NEMATIC TEXTURE

The general idea of this method is illustrated in Figure 1. [5-8] The N phase must be initially subject under the action of external forces (electric or magnetic fields, etc.) in such a way that the N must be strongly-deformed in the surface regions and quite well oriented in the rest part of the sample. The direction

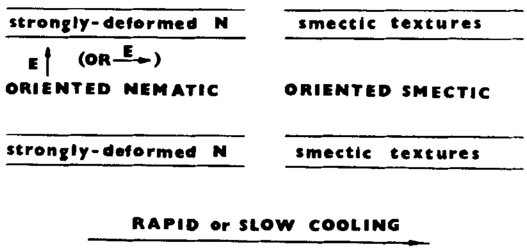

FIGURE 1. A schematic illustration of the novel method proposed for the study of a conventional Sm phase obtained after rapid or slow cooling of a deformed conventional nematic phase.

of the electric field, which should be with a sufficiently high frequency to avoid the eventual development of a hydrodynamics, depends on the sign of the dielectric anisotropy of the N phase and its initial orientation. Under slow or rapid cooling, the Sm phase initially is formed in the middle oriented part of the LC cell and then in the boundary regions in such a way that very nice and homogeneous Sm textures arise. Very often, intermediate textures such as the stripe-like patterns of Cladis and Torza[1] or nonidentified patterns [5], etc. grow which relax and create the final Sm textures. The stripe-like patterns indicate the coexistence of the N and Sm phases [1] whereas the nonidentified patterns show the transformation of the stripe-like domains into the final confocal domains.[5] We cannot exhaust all the possible intermediate textures between the initially-deformed N and the final Sm textures. This can be done only after the performance of the eventual experiments. However, our duty is to note that there are examples when the intermediate textures cannot appear due to a number of causes such as the value of the latent heat around the corresponding N-Sm phase transition and to the surface tension between the N and Sm phases, etc. We shall focus our attention to these problems below. We again emphasize that a Sm monocrystal appears first in the middle oriented part of the N cell where the transition temperature is higher and then strongly-scattering and regular Sm textures are formed in the strongly-deformed N surface regions where the transition temperature is lower. Consequently, the Sm phase created in the surface regions must match itself both to the already created Sm monocrystal in the middle part of the sample and to the orientation of the LC molecules at the surface determined by the way of preparation of the glass plates confining the LC. This way of the Sm growth has two advantages: first, it is possible to obtain very homogeneous Sm scattering centers with a size of several microns, high density and stability, which are nearly independent both on the temperature inside the Sm interval and on the external forces applied across the N layers under study and second, in some of the cases it is possible the approximate calculation of the interfacial energy of interaction between the Sm molecules and the glass slides or their covering.[5,6] In this way, it is possible to obtain Sm scattering textures with desirable properties for application in the display technique. On the other hand, the proposed method can be successfully applied when LCs with a rich polymorphism are used. Let us recall that the Sm mesophase can be arranged in the following order: S_A - S_D - S_C - S_F - S_B - S_H - S_E - S_G .[10, 11, 12] Of course, in the various LCs different Sm combinations might exist. In our knowlidge, there are a number of LCs with a rich polymorphism such as TBBA [13], TBPA [14], and 50,6 [15], etc. (see also the LCs mentioned in Refs. 16-18). Various LCs with a rich polymorphism and possibilities for application in the display technique were noted by Dubois as well.[19] Our method can be applied to those LCs when the Sm phase follows after the N phase under cooling. According to Sackmann [11] and Petrie [20] the following polymorphism is important (let us mention that a number of novel Sm phases have been recently discovered): dimorphism: S_AN; S_BN; S_CN; S_GN; , trimorphism: S_AS_BN; S_CS_AN; S_GS_AN; S_ES_BN; S_BS_CN; , tetramorphism: $S_ES_BS_AN$; $S_GS_BS_AN$; $S_BS_CS_AN$; $S_FS_CS_AN$; pentamorphism: $S_GS_FS_CS_AN$; $S_GS_BS_CS_AN$ and so on. This polymorphism is important for the application of our method due to the following causes: it would be very interesting the experimental investigation of LCs with a rich polymorphism due to the possible transformation of both the high-temperature Sm textures into the lower-temperature Sm textures and the transformation of the high-temperature oriented Sm phase into the lower-temperature oriented Sm phase in cases when this process is possible. In the more favourable cases , it is possible to obtain hogh-quality Sm scattering textures in a wide temperature range for application in the display technique.

A METHOD FOR STUDY OF THE CONVENTIONAL AND REENTRANT SMECTIC PHASES OBTAINED FROM A STRONGLY-DEFORMED NEMATIC REENTRANT TEXTURE

The discovery of the reentrant N phase by Cladis [9] was very important from the scientific point of view. For instance the following sequence of LC phases : $I - N - S_A - N_{re} - S_{A(re)} - K$ give rise to a number of fundamental questions for the structure of the N and S_A reentrant phases[23](a number of papers dealt with this problem and measurements of various characteristic parameters of the reentrant phase such as the order parameter S and the value of the optical birefringence, etc. were made). The rich polymorphism of the reentrant LCs was recently reviewed by Nguyen Huu Tinh [24] in accordance with the results obtained in the Bordeaux LC Group in France and Halle LC Group in DDR. The reentrant LCs enlarge very much the possibilities for application of the novel method . Our general scheme for this case is shown in Figure 2. The comparison of the two schemes shown in Figures 1 and 2 clearly points out the apparent advantages of the novel method proposed for the case of the reentrant LCs. The Sm phase can be studied not only after slow or rapid cooling of the reentrant N phase but also after slow or rapid heating of the same reentrant N phase. The intermediate reentrant and/or conventional N and Sm phases are not shown since these textures depend drastically on the sequence of the LC phases under study. It is important to emphasize on several sequences of LC phases. Let us start with the LCs syntesized and investigated by the Halle LC Group in DDR. Simultaneously, we shall comment on the possible application of the method.

a) $I - S_A - N_{re} - S_B - S_E - K$ [25]

In this case, using a strongly-deformed reentrant N phase in the boundary regions and an oriented reentrant N phase in the rest part of the samples, it is possible to study both the S_A and S_B phases simultaneously.

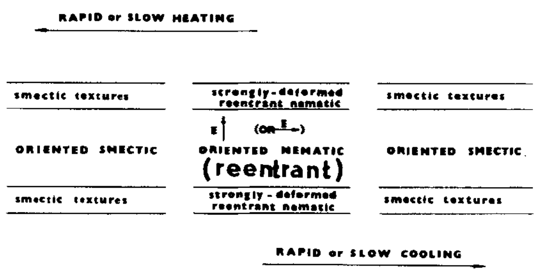

FIGURE 2. A schematic illustration of the novel method proposed for the study of conventional and reentrant Sm phases obtained after rapid or slow cooling or heating of a deformed reentrant N phase.

However, let us stress that the tilted Sm phases such as S_C , S_{B_t} , E_t etc.,[26] should be investigated after deformation of previously-twisted reentrant N layer: i.e. in this case the complex bend-twist-splay deformations must exist in the boundary regions.[7]

b) $I - N - S_A - N_{re} - S_C - K$ [27]

In this case it is possible to investigate both the S_A and S_C phases. As in the previous case, the S_A and S_C phases can be investigated simultaneously upon heating or cooling of a previously-deformed twisted reentrant N layer, respectively. Of course, we can investigate separately the S_A and S_C phases from a different deformed reentrant N phase. For instance, it is sufficient to use a previously-deformed splay-bend reentrant N to study the S_A phase.

c) $I - N - S_A - S_C - N_{re} - K$ [27]

Now we can investigate only the S_C phase, however upon heating of the previously-deformed twisted reentrant N phase.

d) $I - N - S_A - N_{re} - S_{A(re)} - K$ [28, 32]

This case of a phase sequence is very interesting since it gives the possibility to compare the conventional and reentrant Sm textures obtained from the reentrant N phase.

e) $I - N - S_A - S_C - N_{re} - S_B - S_E - K$ [29]

In this case it is possible to study simultaneously the S_C and S_B phases after heating or cooling of the previously-deformed reentrant N phase, respectively.

f) $I - N - S_A - S_C - N_{re} - S_{C(re)} - K$ [28]

Again we have a very interesting phase sequence. Now, it is possible to compare the conventional and reentrant S_C textures.

g) $I - S_A - N_{re} - K$; $I - S_A - S_C - N_{re} - K$;
$I - S_A - N_{re} - S_C - K$ [30]

With such LCs we can investigate the S_A phase upon heating of the previously-deformed reentrant N phase, the S_C phase after heating of the previously-deformed N reentrant phase and the S_A phase after heating of the previously-deformed N phase. The last case is similar to the case b). However, the S_A phase can now be obtained after cooling of the I phase.

Let us begin with the more interesting phase sequences discovered and investigated by the Bordeaux LC Group in France. It is necessary to point out that some phase sequences have been simultaneously discoveied and investigated by the Bordeaux LC Group in France and by the Halle LC Group in DDR.

h) $I - N - S_A - S_C - N_{re} - S_{A(re)} - K$ [31]

In this case we can simultaneously study the conventional S_C phase and the reentrant S_A phase.

e) $I - N - S_A - N_{re} - K$ [24]

This is very interesting case because it permits the comparison of conventional and reentrant N textures. This can be done after the study of the S_A textures obtained either after cooling of the conventional N phase or after heating of the reentrant N phase.

DISCUSSION

We like to outline some general problems closely related to the novel method proposed for the study of conventional or reentrant N and Sm phases. First, we stress that the obtaining of useful Sm textures depends on many causes, some of which will be discussed here. The following circumstances are more important: the value of the initial N or N_{re} splay-bend, splay-twist-bend or only twist deformations in the boundary regions their depth of penetration inside the LC layers, the existence of cybotactic groups[5-7, 33], the value of the latent heat at the corresponding phase transition closly related to the type of the phase transition[1], etc. In our opinion, the rapid cooling is more interesting case since it usually is accompanied by stripe-like patterns which after relaxation give rise to the formation of the final Sm textures. Let us again stress that the rate of the cooling can change the type of the phase transition.[7] It is clear that our method is able to give very interesting and useful Sm scattering textures when LCs with a small latent heat around the phase transitions are used. It is also clear that the case of the reentrant phase is more favourable for application of this method. Let us at this point stress that the small latent heat at the corresponding reentrant phase transitions is evident from the many experimental results obtained up to now. It is well-known that heterogeneous and homogeneous nucleation theories exist[1, 35, 36]. In accordance with the results of Armitage and Price [35, 36] let us recall that the boundary surfaces induce alignment of the molecules and therefore act as heterogeneous nucleation sites. These authors dealt also with the supercooling which usually is characteristic for the first-order phase transition. The general question is related to the possible kind of the nucleation of the Sm phase in the middle oriented part of the samples and in the boundary regions. The answer depends on the nature of the phase transition which in turn depends on the rate of the cooling and the impurity content [37], etc. On the other hand, the very impurities can surve as nucleation centers around which the nucleation sites might be formed,etc. Let us again stress the claims of Armitage and Price[36] for the possible way of growth of the LC phase. It depends on the supercooling, heat flow and temperature distribution, interfacial tension between the low-temperature phase sites and the rest high-temperature phase, surface alignment properties, elastic distortion energy and pressure changes. From our recent experimental results [5-7] we understood that the type of the phase transition closely related to the existence of cybotactic groups is the most important cause which determines the way of the Sm growth. The glass plate topography is important for the type, distribution and homogeneity of the Sm textures. Consequently, the Sm surface forces[5, 6, 38] are very important for the size and the distribution of the Sm scattering centers. In some of the cases, it is possible to calculate approximately the value of the surface energy[5, 6]. According to us , this is the only possible way which permits to evaluate the strength of the surface coupling of the Sm molecules with the glass plates or their covering. Second, we should emphasize that the thermal properties of the glass plates and of the very LC are important as well[5, 8, 39]. Finally, let us again mention that tilted Sm textures can be obtained from initially-deformed twisted N layers. [7] It is easy to understand that the weak Θ-polar surface forces cannot overcome the tilt of the LC molecules due to the very Sm structure. In the case of an initially-deformed twisted N layer, the cybotactic groups play a crucial role for the obtaining of already known and novel Sm textures. [7]

SOME ILLUSTRATIONS OF THE NOVEL METHOD

Although we have applied this method to three separate cases [5-7] we like , for completness, to illustrate the method either by already obtained Sm textures or by Sm textures which are original.

a) Let us start with the case of CBOOA [5]. The N layer was initially planar. The coupling of the LC molecules with the glass plates treated by SiO under vacuum evaporation was strong. A high-frequency electric field with a different strength was applied across the the N layer and then the N phase was subjected under a rapid cooling. The application of a voltage of 3 V (U_{th} = 1,1 V, where U_{th} is the threshold voltage, d = 26 microns) at a cooling rate of 15^0 C/min led to the formation of the well-known honeycomb texture shown in Figure 3a [1]. The raising of the voltage above 5 V, led to the formation of confocal domains shown in Figure 3b (for the details of the explanation see Ref. 5)

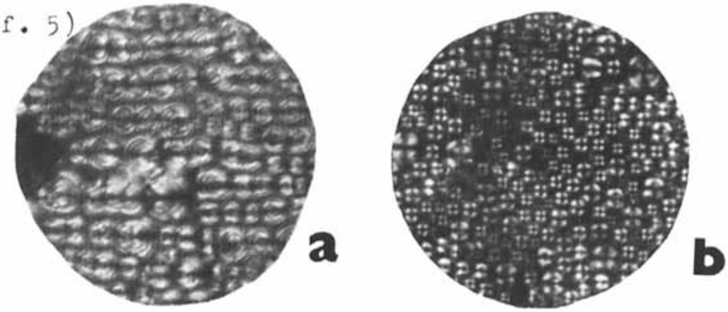

FIGURE 3. Various Sm A textures arrising in the LC CBOOA layer 26 microns thick after the application of a voltage across the N phase folloed by rapid cooling: a) U = 3 V(f = 5 kHz), cooling rate 15^0 C/min, crossed nicols; b) U larger than 5 V

b) The second example is with the LC NPOB which was subjected under the action of a transversal high-frequency electric field.[6] In this case the N phase was initially homeotropic. The coupling between the LC molecules and the glass plate substrate being clear or coated by lecithin was weak. The electric field was applied in a transversal direction due to the positive dielectric anisotropy of the N phase of the LC under study. At a low voltage (U = 100 V, l = 1 mm, f = 5kHz d = 10 microns) we obtained Sm texture very similar to the honeycomb texture [1] (see Figure 4a).

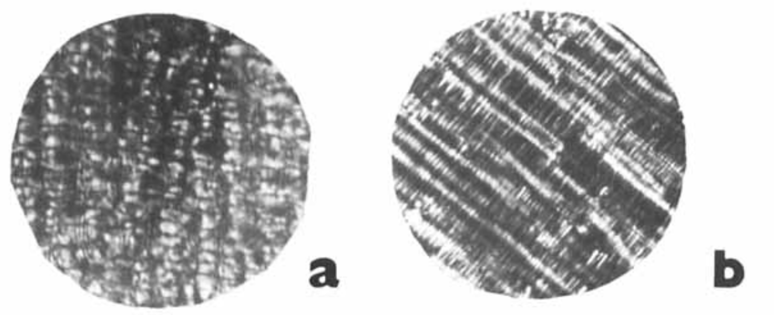

FIGURE 4. Various Sm A textures arrising in a NPOB LC layer of 10 microns after the application of a transversal voltage across the N phase followed by rapid cooling: a) The distance between the metal foils was 1 mm, U = 100 V, f = 5 kHz, rapid cooling rate, crossed nicols, the electric field was applied along the nicol bisectrix; b)U= 700 V the electric field was applied along the polarizer.

Before the obtaining of this texture, the well-known stripe-like patterns were noted. Consequently, this Sm texture has been created due to the coexistence of the Sm and N phases resulting in the formation of a corugated N-Sm A interface and its relaxation. The raising of the voltage up to 500 V led to the formation of regular patterns in the boundary regions consisting of confocal pairs of ellipse-hyperbola with a Marignan-Malet-Parodi association .[6] (see Figure 4b)

c) The final illustration which we like to show concerns the Sm C phase.[7] The LC under study NOBA was initially twisted in the N state. The electric field was applied normal to the glass plates. After the application of a voltage with a sufficient strength (larger than 100 V, d = 26 microns, f = 10 kHz) we obtained for the first time optically-observable edge-dislocations shown in Figure 5 (some of the lines are usual walls). [7] In this way, the application of the novel

FIGURE 5. Observation of novel optically-observable edge-dislocations in a NOBA LC layer 26 microns thick, U = 200 V, f = 20 kHz

method leads to the formation of nice and homogeneous Sm scattering textures which can be either used in the thermally-addressed LC display devices [8] or can be novel Sm textures, especially in the reentrant case.

ONE EXAMPLE FOR DISPLAY APPLICATION [8]

It is evident that the method proposed for the formation of high-quality Sm textures with storage can be applied in the thermally-addressed Sm LC displays. The operation of such a display, for example with CBOOA, is illustrated in Figure 6. The initial transparent homeotropic Sm A case, designated by (1) can be obtained after the application of a sufficiently high AC voltage (above 30 U_{th} for the CBOOA case, where U_{th} is the Frederiks threshold voltage) across the LC area , followed by the heating-cooling cycle. The second step is the decrease of the voltage to between 3 U_{th} and 10 U_{th} and the local heating of the homeotropic transparent Sm A LC to the I phase by an infrared laser beam or by other means. This is revealed by

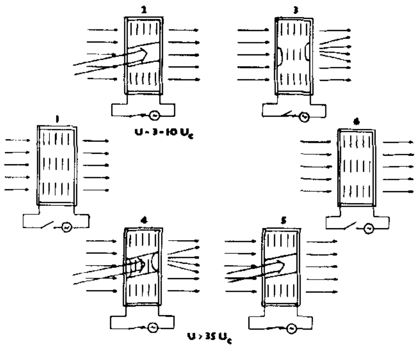

FIGURE 6. Operation of the thermally-addressed display for the CBOOA case for creation of static figures: "WRITING" cycle (1)-(2)-(3), "ERASING" cycle (4)-(5)-(6).

the position (2) of the LC shown in Figure 6. The LC goes back to the Sm A phase after the removal of the heating source. The rapid (free) cooling then leads to the formation of a homeotropic Sm A phase in the middle part of the cell and two strongly-scattering surface regions with confocal domains which are usually homogeneously distributed (see Figure 3b) with a high density. Their size and density, as noted, depends crucially on the treatment of the glass plates and the strength of the AC voltage applied across both the N and Sm A phases. The appearance of the electrically surface-induced confocal domains is SURFACE-CONTROLLABLE, VERY STABLE, REPEATABLE and only SLIGHTLY DEPENDENT on the VARIATION of the TEMPERATURE INSIDE

THE SM A INTERVAL. Neither the increase or removal of the applied voltage changes the Sm A textures obtained. It is clear that the "WRITING" cycle is effected by the (1)-(2)-(3) successive states of the LC as shown in Figure 6. The "ERASURE" of the stored Sm A confocal domains can be local or total and each of these cases is a highly voltage assisted thermal process. This is shown by the (4)-(5)-(6) states of the LC, also illustrated in Figure 6. Initially, the scattering regions are heated by an infrared laser beam up to the I phase (positions (4) and (5) of the LC cell shown in Figure 6). The "ERASURE" cycle is accompanied by the application of a high-frequency AC voltage, which must ensure the final transparent homeotropic Sm A phase as shown in positions (4) and (5) on Figure 6. The final position (6) is equivalent to the initial position (1) and is obtained after the removal of the voltage. Consequently, the "WRITING" (1)-(2)-(3) cycle and the "ERASING" (4)-(5)-(6) cycle are THERMALLY and ELECTRICALLY assisted (Figure 6). The full "WRITING"-"ERASING" cycle is effected by the (1)-(2)-(3)-(4)-(5)-(6) states of the LC.

CONCLUSION

We propose a simple method for the formation of surface-induced and surface-governed conventional and reentrant Sm textures which can be studied from the scientific point of view or can be applied in the laser-addressed thermo-optic Sm LC storage displays for creation of static figures. These Sm displays can work either in transmissive or reflective modes. Since the created Sm textures depend on the type of the phase transition and the latent heat this method is very favourable for application for the reentrant Sm and N phases. The existence of cybotactic groups is very important as well. Still to be explained are the distribution of the heat flow [40, 41] now under novel conditions, as well as a quantitative answer for the direct relation between the topography of the glass plates and the type and distribution of the Sm textures in each of the cases under study, etc. The simple method proposed can be applied for study of many Sms arrising either from conventional or reentrant N phases under cooling or heating.

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