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1990 Europhys. Lett. 11 415

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Re-entrant Phenomenon of a Ferroelectric Liquid Crystal under an a.c. Electric Field.

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(received 19 July 1989; accepted in final form 3 January 1990)

PACS. 61.30 – Liquid crystals.

PACS. 64.70 – Phase equilibria, phase transitions, and critical points.

Abstract. – The T - E phase diagram of a ferroelectric liquid crystal under an a.c. electric field at a fixed frequency $f = 20$ kHz has been determined, and the transition temperatures between the SmA, SmC* and SmC phases have been measured by observing the changes of textures under a polarizing microscope. The experiment shows that the SmC*-SmC transition is not a real phase transition but a simple change of texture, and a re-entrant phenomenon can be found above $E = 2.5 \cdot 10^6$ V/m.

Re-entrant phenomena in liquid crystals have been studied widely in the past several years [1]. The re-entrant phase transition sequence: nematic-smectic A -re-entrant nematic with decreasing temperature has been mostly investigated [2]. Besides, another type of re-entrant phase transition can also take place as a result of the effect of an external field. In ferroelectric liquid crystals the re-entrant distorted SmC* phase was observed under a magnetic field [3] and a d.c. electric field [4, 5]. It seems that such a re-entrant phase transition may probably be observed in the case of the high-frequency a.c. electric field, since it is in essence similar to the magnetic field in coupling with the liquid-crystal anisotropy. However, the mechanism of a.c. unwinding is not yet understood well, although some work has been done on it [6, 7]. In this letter we report on a T - E phase diagram of a ferroelectric liquid crystal under an a.c. electric field at a fixed frequency $f = 20$ kHz. The experimental data indicate that a re-entrant phenomenon can be found indeed above $E = 2.5 \cdot 10^6$ V/m, but it is not a real re-entrant phase transition, and only reflects the texture change.

The material used in this experiment is the chiral ferroelectric mixture FCS 101, kindly provided by the F. Hoffmann-La Roche Company, Switzerland. This mixture presents a SmA phase (from 55.4 °C to 76.1 °C) and a SmC* phase (from 15.5 °C to 55.4 °C). Two flat SnO₂-coated transparent glass plates are separated by mylar spacers with thickness of 25 μm. The electrode surfaces have been rubbed unidirectionally to induce a planar alignment. The sample cell is placed in a hot stage and observed under a Leitz polarizing microscope (ORTHOPLAN-POL). The accuracy in the temperature measurements is better than 0.05 °C. The material is injected in the cell by capillarity in the isotropic liquid. Then

the sample is cooled down slowly (1 °C/h) under an a.c. electric field of 10 V/cm at $f = 20$ Hz. In this way, we can obtain the well-aligned planar SmA texture. Further cooling without the action of the field results in the characteristic parallel stripes pattern of the SmC* phase. An a.c. generator ((0 ÷ 200) kHz) is connected to the glass electrodes in order to apply an a.c. sinusoidal electric field across the sample at a fixed frequency $f = 20$ kHz.

The purpose of our experiment is to plot a T - E phase diagram of the ferroelectric liquid crystal under a high-frequency electric field in order to determine whether there really exists a re-entrant phase transition. Therefore, we measure the temperature and the electric-field dependence of the change of texture under the polarizing microscope to determine the phase boundaries, analogous to the method used in ref. [4]. The domain used for observation is chosen from relatively large domains in a multi-domain focal conic sample. Since the pitch stripes pattern is the characteristic texture of the SmC* phase, it is easy to distinguish C* phase from other phases by observing whether the stripes disappear. When the sample is heated above the SmC*-SmA transition temperature T_{C^*A} , we have a SmC*-SmA second-order phase transition. The stripe spacing increases gradually to a maximum value, then decreases until the stripes vanish, leaving the fan-shaped appearance of the SmA texture. On the other hand, when an a.c. electric field E upon the sample exceeds a critical value E_c at a constant temperature below T_{C^*A} , it will unwind the helical structure of the SmC* phase, and the stripes disappear. It is worth noticing that in this transition process, we observe a kind of colour hydrodynamical flow with the direction perpendicular to the orientation of original parallel stripes. Increasing the voltage, the velocity of the flow increases until the helical texture is unwound. In ref. [8] the authors pointed out that an a.c. electric field can induce a d.c. plug flow of the texture and at higher field the associated viscous shear unwinds the helical texture. The flow observed in our experiment is similar to that described in ref. [8], although these two flows are somewhat different. (This may probably be attributed to the difference of the frequencies. In ref. [8] $f = (10 \div 1000)$ Hz is much lower than that in our experiment.) According to this point of view, the unwound texture is still labelled as the SmC phase, but we keep it in mind that it is not a real SmC phase. After the SmC*-SmC transition comes to an end, the flow will still exist. Further increasing the voltage, the flow becomes more and more drastic and exhibits the characteristic of chaos. So the flow pattern in SmC phase is obviously different from the static fan-shaped SmA texture, which provides us a criterion to distinguish the A and C phases. In this experiment we heat the sample from $T - T_{C^*A} = -16$ °C to $+1$ °C and measure the transition temperature under a fixed field, then we make the sample recover its initial helical texture and repeat the measurement under the different fields. In this way we can determine the phase boundaries.

The observed T - E phase diagram under an a.c. electric field up to $E = 5.0 \cdot 10^6$ V/m is shown in fig. 1. We find that heating leads to the SmC*-SmA phase transition. The A-C* line is within ± 50 mK and independent of the field, as well as that in T - H phase diagram given in ref. [3]. A remarkable feature of the T - E phase diagram in this letter is the existence of a re-entrant SmC* phase above $E = 2.5 \cdot 10^6$ V/m. That is, the critical field E_c has a minimum ($2.5 \cdot 10^6$ V/m) at approximately 1 K below T_{C^*A} . Figure 2 shows the change of texture corresponding to the phase transition sequence under a fixed field (along the dotted line in fig. 1): C*-C-C*-A. The temperature range of the re-entrant C* phase ΔT is narrower and narrower with increasing E , e.g. $\Delta T \sim 50$ mK at $E = 5.0 \cdot 10^6$ V/m. We do not apply the field above $E = 5.0 \cdot 10^6$ V/m for fear of the breakdown. Such re-entrant SmC* phase does not exist in the T - E phase diagram of the same material under a d.c. or a low-frequency a.c. electric field ($f = 50$ Hz), which has been confirmed experimentally [9].

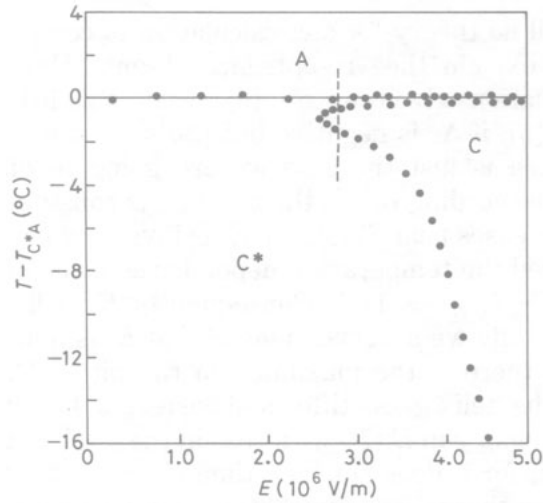


Fig. 1. – The T - E phase diagram of a ferroelectric liquid crystal under an a.c. electric field ($f = 20$ kHz).

Above experimental data show that the T - E phase diagram presented in this letter is different from that obtained by Kondo *et al.* [4] (in their T - E phase diagram the SmA phase exists only on the ordinate, *i.e.* for $E = 0$) and analogous to the T - H phase diagram given by Musevic *et al.* [3]. As the relaxation time of the SmC* phase τ is estimated to be $2 \cdot 10^{-4}$ s, it is passable to take $f = 20$ kHz as a high frequency. Therefore the similarity between the T - E and T - H diagrams is to be expected due to the same essence. However, there is a major difference between them, *i.e.* in the T - E diagram presented here, the SmC*-SmC transition is not a phase transition but a simple texture change, which can also be seen in fig. 1. The difference of nature between the real phase transition SmC*-SmA and the texture change SmC*-SmC is clearly reflected by the independence of the transition temperature *vs.* field. So the «re-entrance» shown in fig. 1 is not a real re-entrant phase transition.

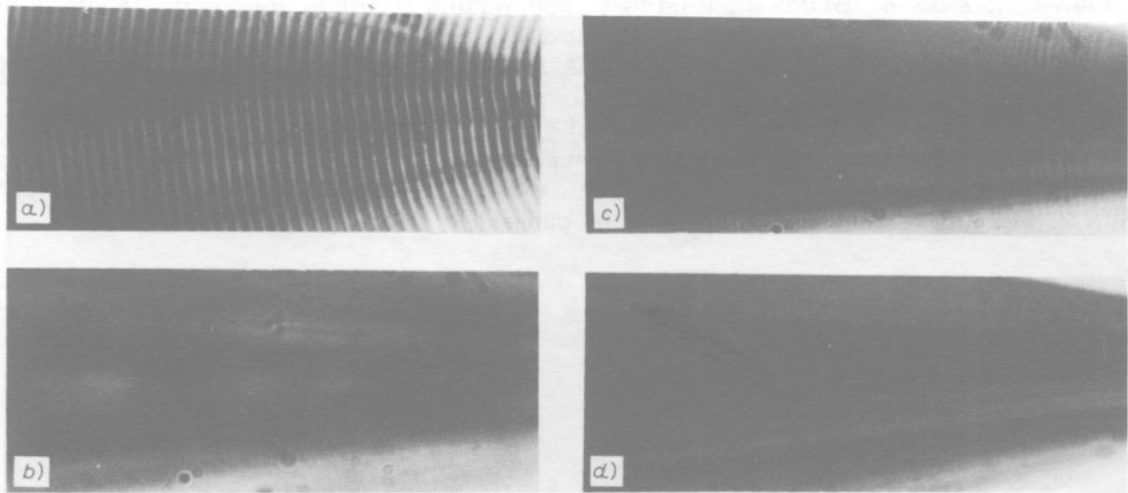


Fig. 2. – Micrographs of a transition sequence (along the dotted line in fig. 1). $E = 2.8 \cdot 10^6$ V/m, $T_{C^*A} = 55.40$ °C. a) $T = 52.40$ °C, SmC* phase. b) $T = 54.85$ °C, SmC phase. c) $T = 55.25$ °C, SmC* phase. d) $T = 56.45$ °C, SmA phase.

Up to now, there is still no theory for a.c. calculation to compare with the experimental results. It is difficult to explain the «re-entrance» bump. Here we try to analyse this problem using the calculation given in ref. [6]. For FCS 101, one has the dielectric anisotropy $\Delta\epsilon \leq 0$. In ref. [6], if $\Delta\epsilon$ is negative but $|\Delta\epsilon| \approx 0$, one can obtain the critical field $E_c = (\pi^4/4)(K_0 \theta/P_0 Z^2)$, which is just the case we are going to discuss. If we neglect the variation of the ratio P_0/θ (according to the theory, P_0/θ is constant, but recent experiments show that P_0/θ slightly decreases near T_{C^*A}), the E_c is inversely proportional to the square of pitch Z^2 . We have measured the temperature dependence of the pitch and we find the pitch reaches a maximum at $T - T_{C^*A} \sim -1^\circ\text{C}$. Consequently E_c will have a minimum for the above formula. Experimentally we observed indeed that E_c is minimal at approximately the same temperature where there is the maximum in the pitch. Close to T_{C^*A} , the pitch is decreasing very rapidly, the helix gets stiffer and correspondingly E_c increases sharply. In this way we connect the anomaly in $E_c(T)$ with that in the pitch of the helix $Z(T)$. However, such analysis is still rough, for it does not take the influence of the frequency into account. Therefore, a more complete theoretical explanation is indeed needed to reveal the anomaly in the critical field $E_c(T)$.

Finally, we would like to point out that the flow associated with the texture change is very complex. For both cases of the focal conic sample and the well-aligned one, the flow patterns are different. We believe that the defects may play an important role. The related problems will be discussed in a separated paper.

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The authors are grateful to Dr. SCHADT for kind supplying the good quality sample. This work was supported by the National Natural Science Foundation of China under the grant No. 1870714.

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