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G. Hauck[a]; H. D. Koswig[a]; U. Labes[a]
[a] Forschungsgruppe Flüssigkristalle (WIP), Berlin, Germany

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Electric field induced structure formation in free standing ferroelectric films

by G. HAUCK, H. D. KOSWIG* and U. LABES
Forschungsgruppe Flüssigkristalle (WIP), Hausvogteiplatz 5/7,
D-1086 Berlin, Germany

Free standing ferroelectric films in a rotating electric field have been investigated. Depending upon the applied voltage and the period of the field rotation different structure formations could be observed. At high fields, a homogeneous orientation is obtained. In the case of very low fields, a schlieren texture results. Under special conditions, a stable ring system appears. The dynamics of these rings have been studied. The dependence of the structure formation on the relation between the impulse length of the applied field and the reorientation time is discussed.

1. Introduction

In free standing films of smectic liquid crystals, different types of structural defects are possible (points, disclinations, π-walls) [1, 2]. These defects may be influenced by electric fields [1]. The formation of domains and π- as well as 2π-walls has been observed in the S_C^* phase at low frequency fields [3]. Effects of electro-convection and structure formation could be detected in S_A samples with enlarged conductivity [4]. Recently, we reported on systems of dark and bright rings, closed disclination walls like bubble domains, which can be created in ferroelectric liquid crystals (FLC) by a rotating electric field in a four electrode experimental set-up [5].

In this paper we want to discuss the conditions under which ring systems and other structures are formed in a rotating electric field. We present also some observations concerning the connection between the motion of the FLC molecules on the smectic cone and the structure formation. First measurements of the dynamic behaviour of ring systems will be offered.

2. Experimental

Free standing films stretched over a hole have been investigated for the S_C^* phase. Four electrodes are arranged perpendicular to each other on the sample, as shown in figure 1 [5, 6]. They offer the possibility of applying a rotating electric field, which results from rectangular voltage impulses, parallel to the film surface. In our experiments, the films had a diameter of 1 mm and a thickness of 500–800 nm. In principal, thinner films can be used but the optical contrast is lower. The upper limit of the field strength was $E_{max} = 1\,kV\,cm^{-1}$, and the period of rotation of the field T, which is the four-fold duration of a single impulse, was between 10 s and 0·4 ms (frequency $f = 1/T$ between 0·1 Hz and 2·5 kHz). The experiments were carried out on a hot stage in a polarizing microscope.

The optical response was controlled by a charge coupled device (CCD) camera with video recording, and the optical transmission by a photodetector connected with an

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* Author for correspondence.

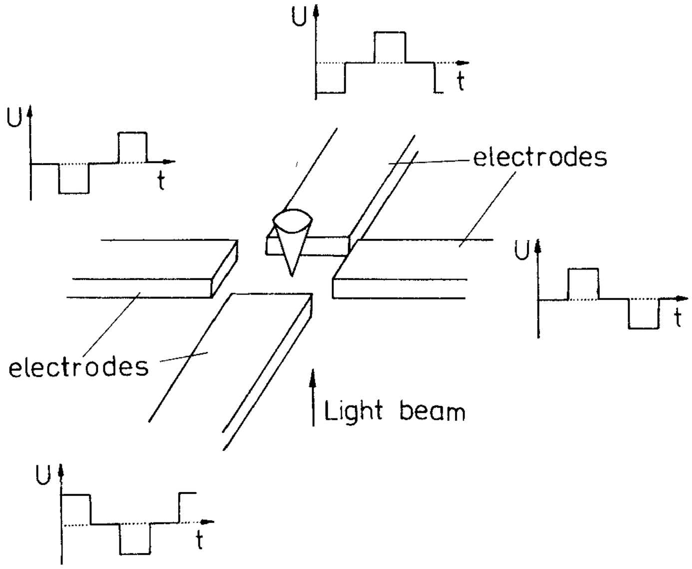

Figure 1. Experimental set-up with impulses applied to the electrodes.

oscilloscope. The symmetry relations between the smectic layers and the field are the same as in the bookshelf structure of thin ferroelectric cells. The rotating field induces a motion of the director on the molecular cone of the S_C^* phase (Goldstone-mode). The time of a field induced 90° turn is $\tau \propto \eta/(PE)$ [7], where η is the rotational viscosity and P the spontaneous polarization. τ has been derived from the optical transmission curves. We used in the experiments two FLC mixtures which exhibit the S_C^* phase at room temperature: 15/2 and FE1, both from MLU Halle, with spontaneous polarizations of 34·7 and 11·3 nC cm^{-2}, respectively. The pitch values of the mixtures at 24°C were 2·3 and 8·5 μm, respectively. The thickness of the film used is smaller than this, and, therefore, a nearly unwound system can be assumed.

3. Results and discussion

At high amplitudes of the biasing electric field, the whole area of the film is homogeneously aligned and rotates with a constant angular velocity which depends on the frequency f of the field [6]. This macroscopic flow may be the result of defects in the layer structure. It seems possible that the field acts on edge dislocations (or pairs of π disclinations) inducing in this way the rotation of the film. This high field behaviour corresponds to region **D** in figure 2. Besides this cooperative, macroscopic, circular flow, there exists the individual motion of the director on the molecular cone of the S_C^* phase, which is characterized by τ.

Under special field parameters (applied voltage U, rotation time of the field T), the homogeneous orientation of the film becomes unstable (below the curve $\mathbf{U}_2$ in figure 2). A central or spiral system of rotating, dark and bright, ring-shaped regions (bubble domain) appears immediately after switching on the field (see figure 3 (*a*)). The different transmission (dark and bright regions) between crossed polarizers indicates a variation of the azimuthal angle and, therefore, a change of the director position on the cone. This behaviour corresponds to a phase delay between the rotating field and the induced orientation of the director. The phase delay is probably the reason for the formation of the domain structure with a zig–zag arrangement of the director in the radial direction

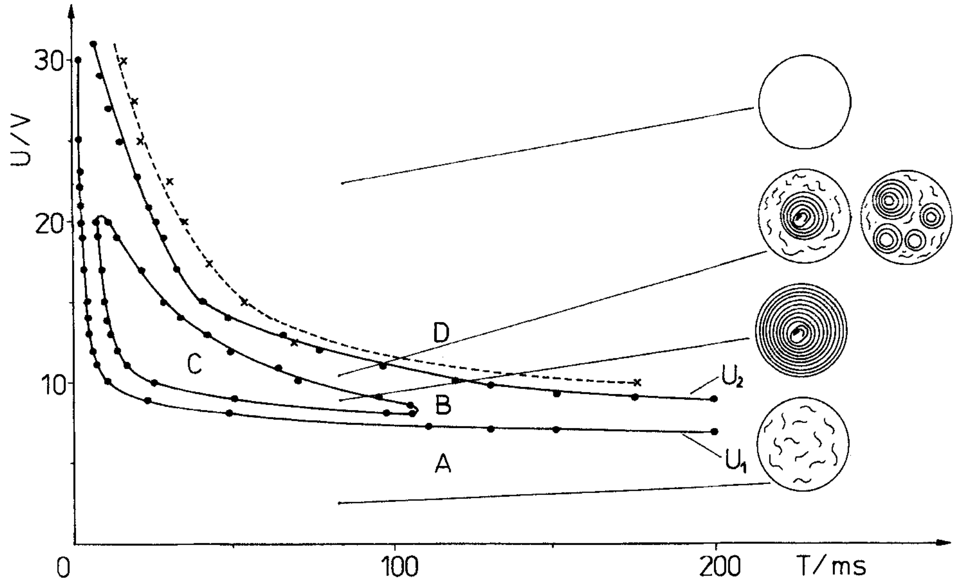

Figure 2. Dependence of structure formation on the applied voltage U and the period of the field T for the mixture 15/2 (24°C); **A**: schlieren texture; **B**: unstable structure formation; **C**: stable ring systems; **D**: homogeneous orientation; U_1 and U_2: limits of structure formation. The dashed line represents the four-fold reorientation time 4τ.

[5]. The range of parameters for a stable structure formation over the whole area of the film (see figure 3 (*a*)) is given by the region **C** in figure 2. The field parameters under which the bubble domain is formed depend on the temperature and the spontaneous polarization of the liquid crystal material.

Between the limiting voltages U_1 and U_2, and outside of **C**, some types of structures may arise: small bubble domains at different places of the film, several domains simultaneously (see figure 3 (*b*)), or a system consisting of some rings of large diameter in the outside part of the film. These structures in the region **B** in figure 2 are unstable. They can grow to enclose the whole area of the film, or they may be destroyed. Below the U_1 curve an inhomogeneous schlieren texture is visible which moves slowly in the direction of the field rotation (see figure 3 (*c*)). The U_1 and U_2 curves are independent of the thickness of the film.

To explain the reason for the structure formation, the time for a field-induced 90° turn of the director on the cone τ is given in figure 2 (dashed line). The value 4τ is drawn there to provide a better comparison with T, which indicates the 360° rotation of the field. The 4τ curve and the curve between regions **D** and **B** are situated very close together. This indicates that a homogeneously oriented film which rotates in the field direction is realized if τ is smaller than the impulse length ($T/4$), or if both values are nearly equal to each other. At sufficient field strength, a homogeneous orientation of the molecules in the film is enforced. This orientation rotates synchronously with the field. The relation between the impulse length and the reorientational behaviour of the director is schematically shown in figure 4.

If $\tau > T/4$, the field is switched to the next direction until the director is able to attain the previous position. There is a delay of the orientation with respect to the field. This phase delay is probably the reason for the spontaneous formation of the ring structure with a zig-zag arrangement of the director with respect to the field direction. At very

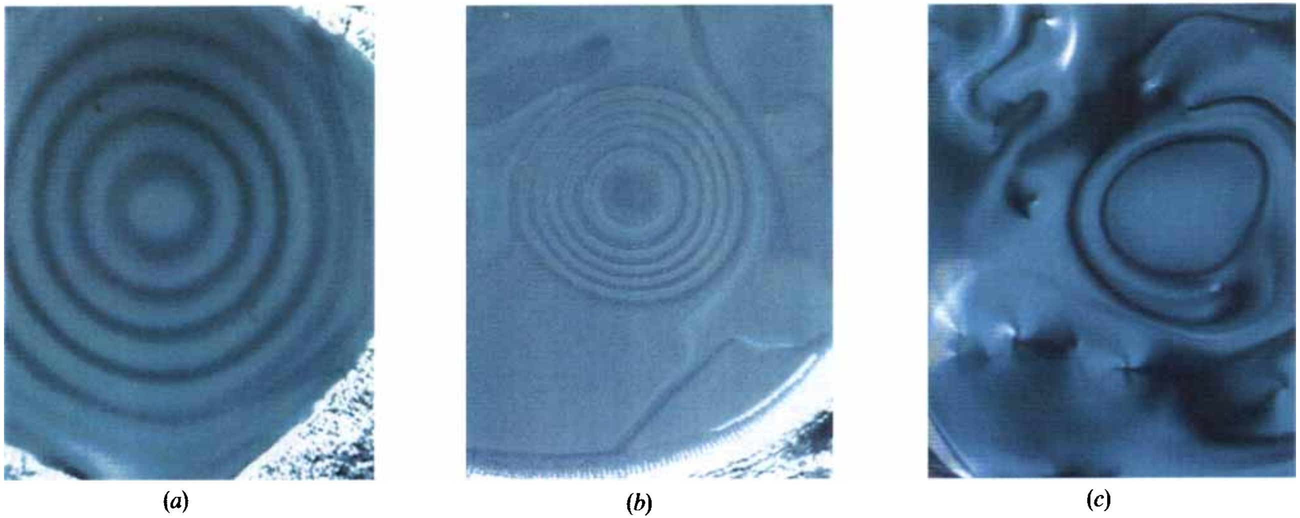
(*a*) (*b*) (*c*)

Figure 3. Structures in the mixture 15/2: (*a*) stable ring system which fills the whole area of the film; (*b*) small domain; (*c*) schlieren texture.

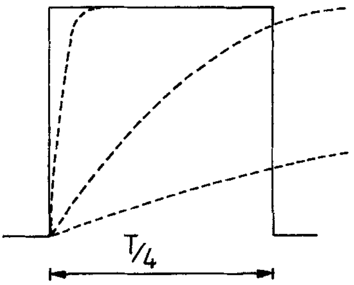

Figure 4. Relation between impulse length ($T/4$) and reorientation behaviour.

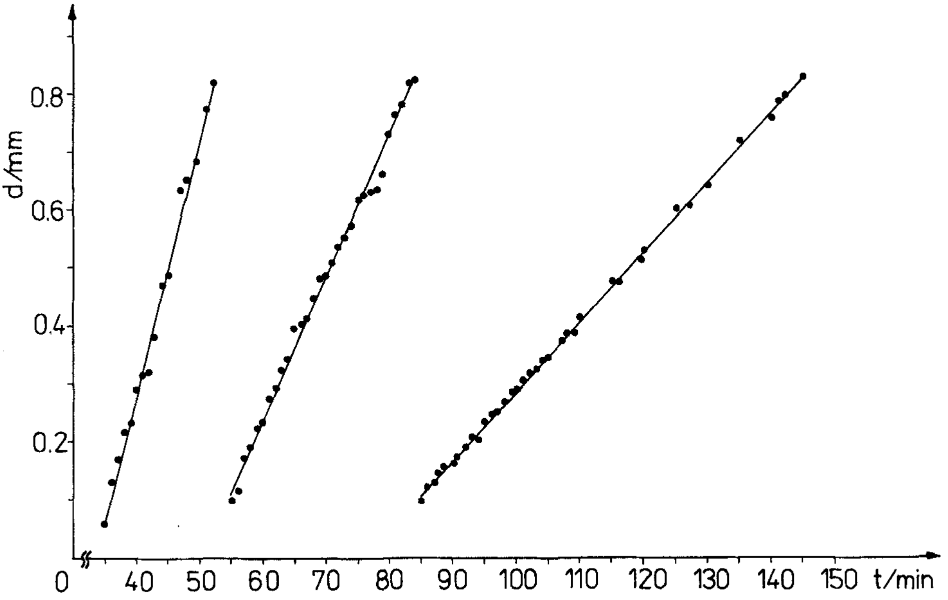

Figure 5. Increase in diameter d of three different rings with the time t (application of the field at $t=0$) for the mixture 15/2 at 24°C; $U=12\,V$, $T=20\,ms$.

long switching times (lower case in figure 4), a defined relation between the direction of field rotation and the direction of director movement on the cone does not exist. The switching in different areas of the schlieren texture is not synchronous.

After the initial formation of a quasi-stationary system, new ring-shaped regions may be created, dependent on the applied voltage, in the centre of the film. Their diameter d increases, and they are destroyed at the border of the film. Figure 5 shows a linear increase of d for the mixture 15/2. The field has been switched on at $t=0$. The left curve in figure 5 corresponds to the growth of an arbitrarily selected ring-shaped region in the ring system. It is created as an area of finite extension in the rotating centre of the system. The area enlarged, and, after the creation of the next area, the selected region becomes ring-shaped. If the selected region is destroyed at the border of the film (upper end of the left curve), the development of the region with the smallest diameter at that moment is further investigated. The curves never intersect the time axis because of the finite extension of the regions at their creation. The slope of the curves is dependent on U and the temperature. The increase decelerates for later created rings. In the mixture FE1, this expansion of the diameter turns out to be non-linear (see figure 6). In the

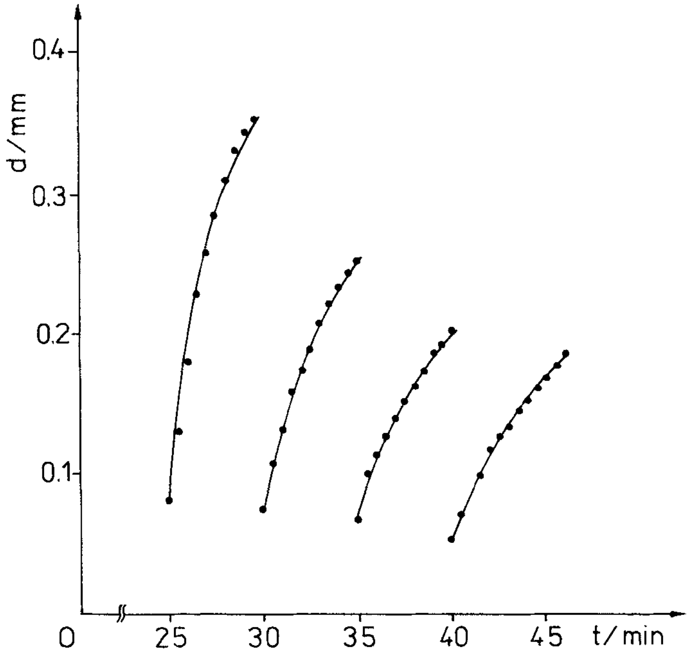

Figure 6. Increase in ring diameter *d* for the mixture FE1 (24°C); $U=17$ V, $T=1$ ms.

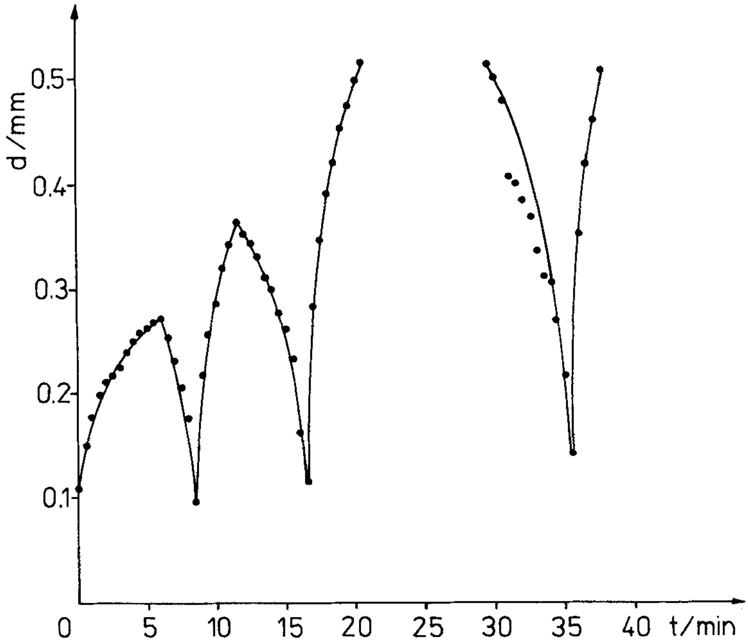

Figure 7. Variation in diameter *d* of a ring for the mixture FE1 (24°C) on successively applying (increasing *d*) and switching-off the field (decreasing *d*); $U=17$ V, $T=1$ ms.

mixture 15/2 the increase could be also non-linear if the rings were able to grow to greater diameters than those defined by the geometric limits of the sample. In this case, the curves in figure 5 would be only the nearly linear initial portions.

The expansion of the rings is not connected with a radial transport of the FLC substance, but the quasi-stationary orientational order in the ring system is shifted by the electric field. If the field is switched off, the growth of d stops and a reduction commences. In figure 7, the behaviour of one defined ring is drawn: after application of the field ($t=0$), the diameter of the spontaneously formed ring grows. Then the field is switched off and d decreases. The reduction in the diameter does not turn out in a square manner, as observed in [1]. Now, the field is applied again, and a new expansion starts. On successive switching of the field, the corresponding expansions and reductions of the ring are accelerated. The velocities of expansion and reduction are dependent on the excess energy of the walls and the viscous properties of the FLC [8].

In thin FLC cells with an electrode arrangement identical to that for the free standing films, we were not able to induce structure formation in the manner discussed with our limited field strength ($E_{\max}=1\,\mathrm{kV\,cm^{-1}}$) and frequency range. The surface forces between the FLC molecules and the solid surface are probably too strong. But, the synchronous motion of the director on the smectic cone takes place.

We could also observe the formation of ring systems in free standing films of S_C phases. Their behaviour is somewhat different from those for the ferroelectric mixtures and they appear at lower frequencies. The reorientation of the molecules is determined only by the dielectric properties of the liquid crystal. The admixture of a conducting dopant does not change the behaviour of the S_C substance. This observation gives rise to the assumption that the structure formation is not connected with the transport of free charges, as reported for the vortex pattern formation in a two electrode arrangement [4].

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