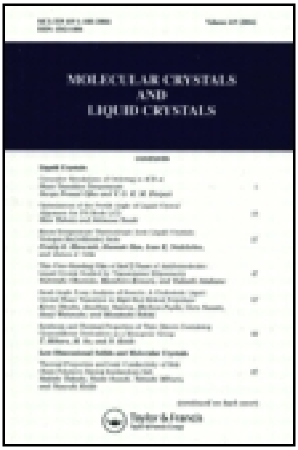

Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl17

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Version of record first published: 17 Oct 2011.

Mol. Cryst. Liq. Cryst., 1987, Vol. 151, pp. 211-223
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Printed in the United States of America

ELECTRIC FIELD INDUCED RELAXATION PROCESSES IN UNWINDING HELICAL TEXTURE OF FERROELECTRIC LIQUID CRYSTALS

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Abstract Under the influence of electric field, helical texture of ferroelectric liquid crystals tends to unwind, giving rise to macroscopic polarization which saturates at a critical field E_C. The helix dynamics in a.c. fields from 0-20KHz has been observed in the compounds having :

1. positive dielectric anisotropy and low permanent polarization,
2. negative dielectric anisotropy and low permanent polarization.

Two relaxation processes have been observed : a "slow" relaxation determined by relaxation time $\tau_s = \gamma(Kq_o^2)-1$ and a "fast" relaxation determined by the relaxation time $\tau_f = \gamma[2\alpha(T_c^*-T)]^{-1}$ (γ is the dynamic viscosity Coefficient). The typical relaxation frequencies for τ_s and τ_f correspond to $\approx$ 400 Hz and $\approx$ 1 KHz respectively. It has been observed that new textures result from the superposition of dynamics caused by helix and undulation instability created by electric field induced charged layers.

INTRODUCTION

Existence of ferroelectric liquid crystals was established by Meyer et al[1]. in 1974. Many of their physical properties have been studied since then.[2-10] Meyer noticed that spontaneous polarization in ferroelectric liquid crystalline materials was lower than that in solid ferroelectrics. Furthermore, this polarization was associated with the helical texture of its molecules inside the smectic layers perpendicular to the molecular director. The angle between the normal z-axis and the director is called tilt angle θ. It has both temperature as well as electric field dependence[2,10] When a chiral smecticC(SmC*) compound is sandwiched between two transparent glass plates, parallel stripes are observed through polarizing microscope under crossed polarizers. The spacing between the stripes gives the pitch Z, of their helical structure in terms of temperature T and electric field E.

The external electric field unwinds the helical texture completely at a critical field E_c which helps in the measurement of polarization P, the elastic constants K, the tilt angle θ and the helical pitch Z. Exact behaviour of the helix in electric field is still not properly understood.

Herein, we study the effects of a.c. fields on the helix pitch in two ferroelectric liquid crystals over 0-20 KHz. The experimental measurements indicate dynamic processes associated with the deformation of the helix. They also emphasize the role of piezo-electric effects in spontaneous polarization. The actual mechanism of helix deformation is still not well understood despite efforts by various researchers[5,8] Electric field induced dynamics of the helix due to relaxation processes has been discussed and principal findings are reported in proceeding sections. The author feels that effects of polarization of the molecular dipoles on relaxation processes at higher frequencies needs further experimental investigation.

RELAXATION PROCESSES

We consider a ferroelectric SmC* liquid crystal molecule in its planar orientation. Director $\underline{n}(r)$ generates a cone with semi-vertical angle θ around z-axis, normal to the smectic layers. Let ϕ be the azimuth angle between field $\underline{E}$ and polarization vector $\underline{P}$. Components of $\underline{n}(r)$ are given by

$$n_x = \mathrm{Sin}\,\theta\ \mathrm{Cos}\phi$$
$$n_y = \mathrm{Sin}\,\theta\ \mathrm{Sin}\phi \qquad (1)$$
$$n_z = \mathrm{Cos}\,\theta$$

At equilibrium, structure of the SmC* is defined by a uniform twist $\frac{d\phi}{dz} = q_0$ resulting in a helicoidal conical texture, $q_0 = \frac{2\pi}{Z}$, Z being the helix pitch. In an electric field $\underline{E}$, free energy density of SmC* phase is given by[3]

$$F = F_0(\theta) + \frac{1}{2} K\theta^2 \left(\frac{d\phi}{dz} - q_0\right)^2 - \chi\,\mu_p \theta E\ \mathrm{Cos}\phi \qquad (2)$$

where $F_0(\theta) = \frac{1}{2} a\theta^2 + \frac{1}{4} b\,\theta^4$ (3)

is the contribution due to Landau free energy. $a = \alpha(T-T_c^*)$, $\alpha > 0$ and $b > 0$ are the associated saturation terms. Second term in Eq.(2) is due to pure twist of the layers and the third term because of coupling between $\underline{E}$ and the permanent polarization $\underline{P} \propto \mu_p\theta$. K is the twist elastic constant, χ the dielectric susceptibility and μ_p the piezo electric coefficient. Flexoelectric coefficient and the dielectric anisotropy $\Delta\epsilon$ has been neglected in Eq.(2). For low frequency fields, "flexo" contribution arising from large area distortions will be reduced due to screening of polarization charges by ionic impurities in the liquid crystal. At higher frequencies, $\Delta\epsilon$ is expected to be reduced[4]. In equilibrium, ϕ dependent part of free energy:

$$\Delta F = \frac{1}{2} K\theta^2 \left(\frac{d\phi}{dz} - q_o\right)^2 - \chi \mu_p \theta E \cos\phi \qquad (4)$$

For $\frac{dz}{d\phi} = \frac{1}{q_o}$, minimization of above equation with respect to $\phi(z)$ gives

$$K \theta \frac{d^2\phi}{dz^2} + \chi \mu_p E \sin\phi = 0 \qquad (5)$$

In an alternating field $E = E_0 \exp(i\omega t)$, the equation of helix untwisting dynamics becomes

$$(K \theta + I) \frac{d^2\phi}{dz^2} - \gamma\theta \frac{d\phi}{dt} + \chi \mu_p E \sin\phi = 0 \qquad (6)$$

Where I is the moment of inertia of the molecule and γ, the dynamic viscosity coefficient of the liquid crystal. Solution of the above equation, neglecting the inertial term, is given by

$$\phi = q_o Z + A \exp i(\omega t + \alpha) \sin q_o Z \qquad (7)$$

which after substitution in Eq.(6) gives

$$A \exp(i\alpha) = \frac{\chi \mu_p E_o \tau}{\theta \gamma (1 + i\omega\tau)} \qquad (8)$$

where

$$A = \frac{\chi \mu_p E_o}{K q_o^2 \theta (1 + \omega^2\tau^2)^{\frac{1}{2}}} \qquad (9)$$

$$\tau = \frac{\gamma}{K q_o^2} \qquad (10)$$

and

$$\tan\alpha = (\omega\tau) \qquad (11)$$

τ is the relaxation time of the molecule. The dielectric relaxation dispersion due to "soft" as well as "Goldstone" mode and the Landau energy in the SmC* becomes

$$\Delta \epsilon_c^*(\omega) \approx \frac{2\pi \mu_p^2}{K q_o^2 (1 + i\omega\tau) + 2\alpha(T_c^* - T)} \qquad (12)$$

for $(T_c^* - T) < 1^oK$.

Eq. (10) and (12) indicate that there are two relaxation processes : a "slow" one determined by the relaxation time

$$\tau_s = \frac{\gamma}{Kq_o^2} \tag{13}$$

and a "fast" one with relaxation time

$$\tau_f = \frac{\gamma}{2\alpha(T_c^*-T)} \tag{14}$$

The "fast" relaxation is governed by the temperature dependence of the helix pitch and is due to the polar perturbation in SmC* and SmA phases. These relaxations are reflected in the polarization under threshold field E_c. This should give it higher values at frequencies corresponding to the two relaxations. Beresmev et.al[11]. also had observed this phenomena.

ELECTRIC FIELD INDUCED HELIX DYNAMICS

Dynamics of the helix in an alternating field was investigated in two ferroelectric liquid crystal Compounds ;

1) p- decyloxy benzylidene p'-amino 2-methyl butyl cinnamate (DOBAMBC)

2) p-decyloxy benzylidene p'-amino 2-methyl butyl α- Cyamo cinnamate (DOBAMBCC).

Their molecular structure , phase transition temperatures and dielectric anisotropy is illustrated in Fig. 1. DOBAMBC has positive dielectric anisotropy and low permanent polarization whereas latter has a negative dielectric anisotropy and low permanent polarization. The compounds were sandwiched in planar orientation between two SnO_2 coated glass plates seperated by Mylar spacers. Thermal microscopy was studied using a Leitz polarizing microscope and Mettler FP_5 + FP_{52} temperature controller programmer maintaining accuracy up to

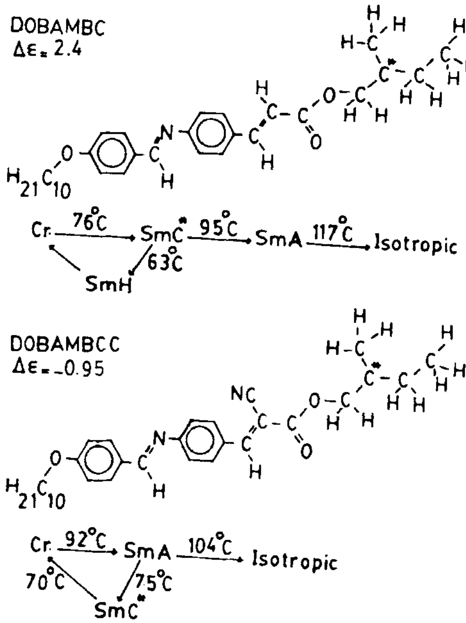


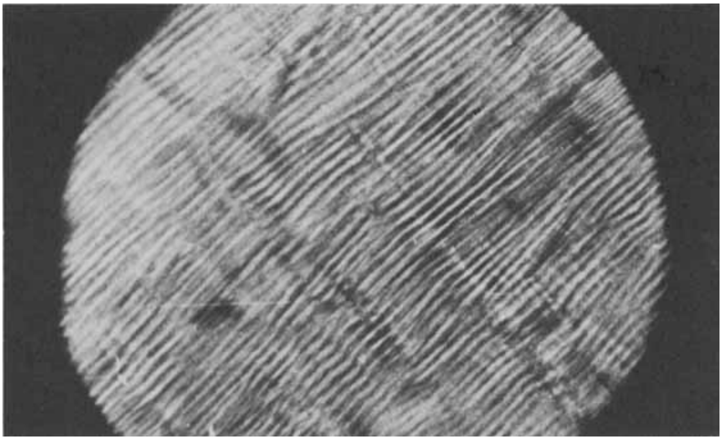
FIGURE 1. Molecular structure, phase transition temperatures and dielectric anisotropy of the Compounds studied.

0.1°C. Helix texture was observed at SmA — SmC* transition temperature with the appearance of colours due to the selective reflections from the helical stripes. It can be unwound by external fields.
A.c. field at different frequencies was applied through a frequency generator and an amplifier across the SnO_2 coated electrodes. Observations for unwinding the helix completely were recorded at a particular frequency and temperature. The average of three such

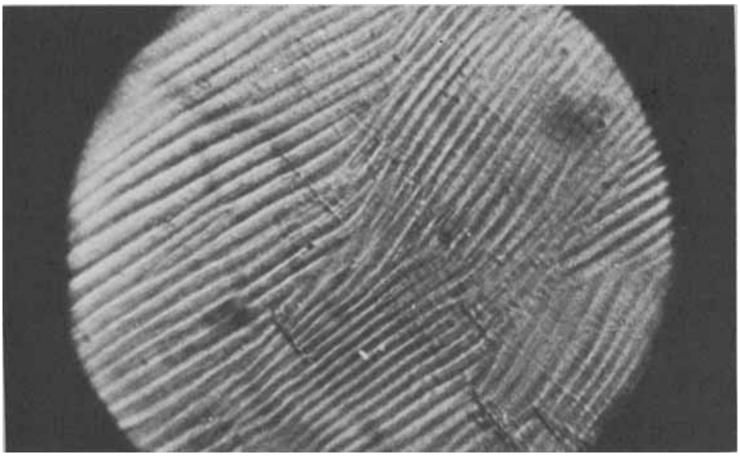
measurements gave the exact value of E_c.

NEW TEXTURE

We noted that "slow" and "fast" relaxation processes were expected to reflect their influence on E_c.



(a)



(b)

FIGURE 2. Dynamics of helical texture in SmC* phase (a) appearance of π- defect entering the texture at 200Hz (b) λ- texture formed at 500Hz.

Clark[12] suggested a new electric field - induced-mechanical instability of a monodomain SmC*. He also predicted the existence of charged layers due to layer distortion by large electric fields. For values close to E_c, bend distortions and layer displacements were reported to be sinusoidal. Such distortions and resultant charged layers would give rise to a new kind of helix dynamics as shown in Fig.2. Behaviour of the monodomain sample for applied voltage Vr.m.s. = 10 Volts, f = 200Hz, in Fig.2(a), shows that helix starts deforming and the parallel stripes texture became distorted. It was interesting to note that a π-type defect entered the texture and stripes were seen oscillating. At higher values of Vr.m.s. and frequency = 500Hz, a new type of texture was seen as in Fig. 2(b). This resembles with William's domains but is somewhat different because of dynamics of the charged layers resulting from undulation instabilities. The stripes became wavy because of the superposition of two types of motions of the helical texture; one due to the unwinding of the smectic layers in SmC* and the second that of charged layers. A new texture demonstrates a λ - pattern. When the a.c. field is further increased motion of the charged layers dominated and a new type of pattern appeared which disappears completely at threshold value E_c.

HELIX UNWINDING

Typical characteristics of the field frequency are shown in Fig.3 for DOBAMBC and DOBAMBCC.
In the former case, Fig.3(a), we notice that E_c increases rapidly near the "slow" relaxation frequency

and falls there-after. At $\Delta T = 12.0^{o}C$, E_c increases from $0.8 \times 10^4 Vcm^{-1}$ to $1.5 \times 10^4 Vcm^{-1}$ for 600Hz. At

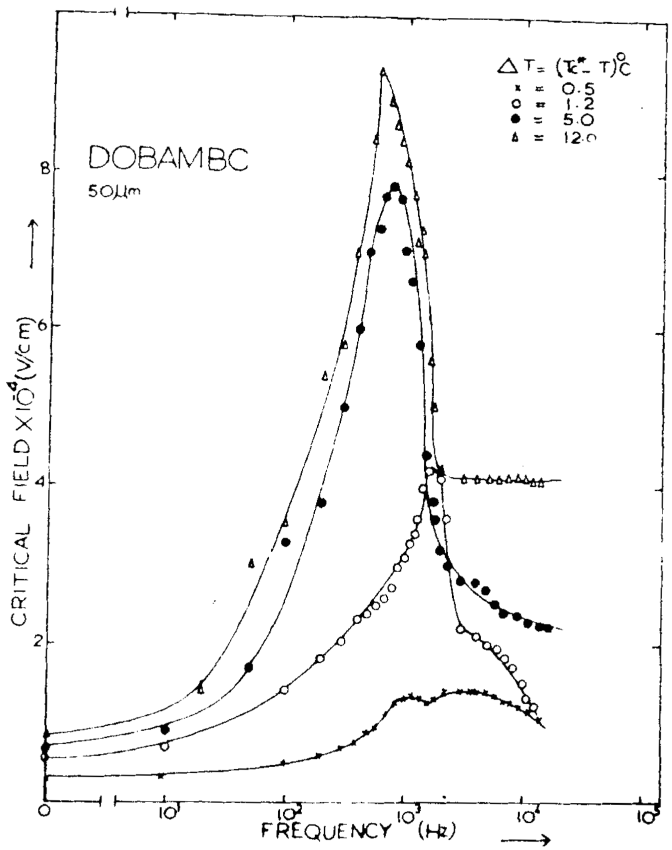


FIGURE 3 (a) A.C. E_c as a function of frequency for DOBAMBC

higher frequencies E_c falls to $4.2 \times 10^4 Vcm^{-1}$ for 2KHz and saturates thereafter. It is in agreement with Vistin et.al.[13] E_c indicates saturation at higher frequencies. Relaxation Corresponding to the "fast" mode could not be observed because of limitations on frequencies.

Frequency dependence of E_c for DOBAMBCC in SmC* phase is shown in Fig.3(b). The two figures reveal

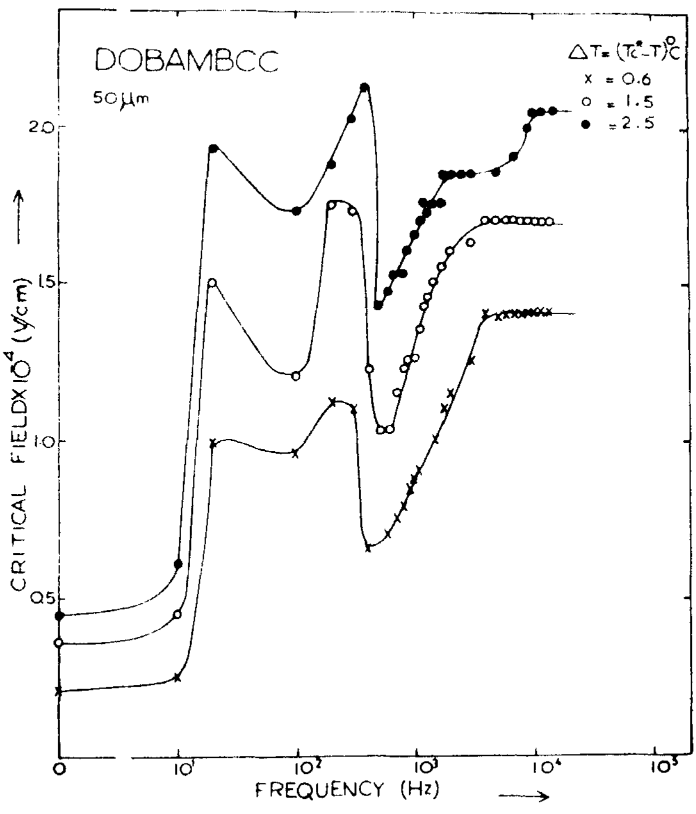


FIGURE 3 (b) A.C. E_c as a function of frequency for DOBAMBCC

different effects at high frequencies. In this case relaxation frequency corresponding to the high frequency peak shifts towards lower frequency side on increasing ΔT unlike in the first sample. Critical field E_c saturates at higher frequencies. The "slow" relaxation peak in DOBAMBCC corresponds to 200Hz and second at 400Hz much below the one observed in the former compound.

The low frequency behaviour of E_c can be explained on the basis of contribution of permanent polarization giving rise to the same unwind state of the helix as for d.c. field. As the frequency is increased, the induced polarization becomes responsible for helix unwinding and hence higher E_c values. The shift of "slow" relaxation peak towards higher frequencies suggests weak coupling between different molecules at higher temperatures and hence easy follow on of the field frequency of the molecules. The molecular structure of these compounds as shown in Fig.1 is also expected to give rise to relaxation phenomena due to branched polarization of the molecules.

CONCLUSION

The a.c. field effects indicate "slow" and "fast" relaxation in ferroelectric liquid crystals. The relaxation corresponding to "piezo" effects is expected in the MHz range.

A new texture has been observed as a result of superposition of helix and charged layer dynamics. The dielectric anisotropy effects on E_c suggest that at high frequencies it is lower for compounds with $\Delta\epsilon>0$ and higher for those processing $\Delta\epsilon<0$.

POSSIBILITIES FOR FURTHER INVESTIGATION

Mechanism of interaction between field frequency and the dipoles of ferroelectric phase needs more understanding despite efforts by various researchers[3,14,15]

A model for a.c. E_c is yet to be proposed which could explain the dynamics of the helix and the measurement of polarization associated with its deformation. The contribution of dielectric anisotropy and "flexo" effects in free energy density for unwinding the SmC* helix is to be investigated.

λ– texture depicted in Fig.2(b) needs further explaination. Dynamics of stripes at high frequencies is expected to find use in display devices. The high frequency region of the critical field remains unexplained. There is need to investigate frequency dependence of E_c in the MHz range.

The author is grateful to Dr. D.S. Parmar for many helpful discussions. These investigations were carried out as a part of DST (India) project at Kashmir University. Interest of Dr. N.C. Nigam and Dr. B.M. Nayer is thankfully acknowledged.

REFERENCES

1. R.B.Meyer, L. Liebert, L. Strzetcki and P.Keller, J. Phys. (Paris) Lett., 30, 69 (1975)
2. Ph. Martinot - Lagarde, J.Phys. (Paris), 37-C3, 129 (1976)
3. B. I. Ostrovskii, A.Z. Rabinovich, A.S. Sonin and B.A. Strukov, Sov. Phys. JETP, 45, 912 (1978)
4. D.S. Parmar and Ph. Martinot-Lagarde, Ann. Phys. (Paris), 3, 275 (1978)
5. R. Blinc and B. Zeks, Phys. Rev. A, 18, 740 (1978)
6. S. Garoff and R.B. Meyer, Phys. Rev., A, 19, 388 (1979)

7. Ph. Martimot Lagarde, R. Duke and G. Durand, Mol. Cryst. Liq. Cryst., 75, 249 (1981)
8. Ph. Martinot-Lagarde and G. Durand, J. Phys. 42, 269 (1981)
9. N.A. Clark, M.A. Handschy and S.T. Lagerwall, Mol. Cryst : Liq. Cryst., 94, 213 (1983)
10. D.S. Parmar, K.K. Raima and J. Shanker, Mol. Cryst Liq. Cryst., 103, 77 (1983)
11. L.A. Beresnev, L.M. Blimov and F.B. Sekolova, JETP Lett., 28, 340 (1978)
12. N.A. Clark, Appl. Phys. Lett., 35, 688 (1979)
13. L.K. Vistim, S.P. Chumakova and Z.B. Rajabova Advances im Liquid Crystal Research and Applications, edited by L. Bata, (Pergamom Press, Oxford, 1980).
14. B. Zeks, A. Levestik and R. Blinc, J. Phys. Collog. 40, C-3, 402 (1979)
15. Ph. Martimot - Lagarde and G. Durand, J. Phys. Lett., 41, L-43 (1980)