Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

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To cite this Article Kumar, A. , Prakash, J. and Biradar, A. M.(2010) 'Non-linear behaviour of phason mode with bias field in ferroelectric liquid crystals', Liquid Crystals, 37: 3, 247 — 253
To link to this Article: DOI: 10.1080/02678290903506032
URL: http://dx.doi.org/10.1080/02678290903506032

Liquid Crystals,
Vol. 37, No. 3, March 2010, 247–253

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Non-linear behaviour of phason mode with bias field in ferroelectric liquid crystals

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(Received 23 April 2009; final version received 23 November 2009)

The decrease in Goldstone mode relaxation frequency with applied dc electric field in a deformed helix ferroelectric liquid crystal (DHFLC) is observed at room temperature. This decrement in relaxation frequency is observed at very low dc electric fields (below the threshold voltage, i.e. around 2 V). This behaviour is compared with that of conventional ferroelectric liquid crystals (FLCs) where the relaxation frequency increases with the applied dc electric field. It is observed that a low dc electric field is insufficient to unwind the helix completely due to the higher rotational viscosity of the DHFLC material. Moreover, the DHFLC molecules take more time in the winding (Smectic C–Smectic C*) process, compared to the FLC molecules, which take comparatively less helix winding time because of their low viscosity. The decrease in relaxation frequency in DHFLCs with bias has been attributed to the critical balancing amongst the electric torque, elastic constraints and rotational viscosity in a certain regime of the electric field not exceeding the threshold field.

Keywords: deformed helix ferroelectric liquid crystal; relaxation phenomenon; Goldstone mode; rotational viscosity

1. Introduction

There has been a tremendous growth in the research field of ferroelectric liquid crystals (FLCs) from both the theoretical and experimental point of view since their discovery by Meyer *et al.* [1]. Dielectric relaxation studies in a large number of FLCs have been reported [2–9]. These studies give an idea of the molecular orientations and relaxation mechanisms in the case of FLCs. In the chiral smectic C (SmC*) phase of FLCs, the molecules are arranged in a helical manner because the director precesses about the smectic layer normal with a constant inclination (tilt angle) of the director. The dielectric relaxations in the SmC* phase can be described by two relaxation modes [2] such as the Goldstone mode, which is due to the phase fluctuations, and the soft mode, which is due to fluctuations in the tilt angle. Moreover, there may be some additional modes produced by the dynamics of the molecules. Among all FLCs, the deformed helix ferroelectric liquid crystal (DHFLC) has become more prominent since their discovery [10] due to their fast response, low driving voltage, easily achievable alignment and grey-scale generation capability in the field of liquid crystal displays [11]. Furthermore these materials have very short pitch ($\sim$0.3–0.7 μm), high rotational viscosity and large spontaneous polarisation. We report here the decrement in the Goldstone mode relaxation frequency with the applied field in DHFLCs, which has been attributed to critical balancing amongst the electric torque, elastic constraints and rotational viscosity in a certain range of the electric field.

2. Experimental

Sample cells for the above studies were made from indium tin oxide (ITO) coated, optically flat (having flatness $\lambda/2$), glass substrates. For homogeneous alignment, the substrates were treated first with adhesion promoter and nylon (6/6) and then unidirectional rubbing was performed on both surfaces to ensure the planer alignment [12, 13]. The thickness of the sample cells was maintained using 3.5 μm Mylar spacers. In this study, a DHFLC material (FLC 6304, Rolic, Switzerland) and a FLC (CS1016, Chisso, Japan) were used. The phase sequence of these materials is as follows:

$$\text{cryst} \xleftrightarrow{-14^\circ\text{C}} \text{SmC}* \xleftrightarrow{60.5^\circ\text{C}} \text{SmA} \xleftrightarrow{64^\circ\text{C}} \text{iso} \qquad (\text{FLC6304})$$

$$\text{cryst} \xleftrightarrow{-21^\circ\text{C}} \text{SmC}* \xleftrightarrow{56^\circ\text{C}} \text{SmA} \xleftrightarrow{67^\circ\text{C}} \text{N} \xleftrightarrow{73^\circ\text{C}} \text{iso} \qquad (\text{CS1016})$$

The material was introduced into the sample cells by means of capillary action at an elevated temperature, which ensures that the filling takes place in the isotropic phase. The dielectric measurements of the

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ISSN 0267-8292 print/ISSN 1366-5855 online

DOI: 10.1080/02678290903506032
http://www.informaworld.com

sample cells were taken using an impedance analyser 6540A (Wayne Kerr, UK) on the application of a low ac measuring voltage of 1 V. The effect of different low-frequency measuring voltages at a constant bias of 1 V was also tested. The determination of the material constants such as the spontaneous polarisation and rotational viscosity was carried out by using an automatic liquid crystal tester (ALCT-P Instec, USA). The optical micrographs of the cells were taken using a polarising optical microscope (Ax-40, Germany). The tilt angle measurements at different dc biases were also performed with the help of a polarising optical microscope.

3. Theoretical background

Two collective relaxation modes (the Goldstone and soft modes) connected with the director reorientation motion and other two polarisation modes lying in the high-frequency regime connected with the distortion of the electronic configuration are observed in almost all types of FLC materials. The expression governing the contribution of Goldstone and soft modes to complex dielectric permittivity [14–18] can be written as

$$\varepsilon^*(\omega, T) = \varepsilon_\infty + \Delta\varepsilon_G(T)\Big/(1 + i\omega\tau_G)^{1-\alpha_G} + \Delta\varepsilon_S(T)\Big/(1 + i\omega\tau_S)^{1-\alpha_S} + \frac{\sigma}{j \in_0 \omega}, \quad (1)$$

where $\tau_i = 1/2\pi f_i\,(i = \mathrm{G, S})$ is the relaxation time for the molecules of the corresponding mode, $\Delta\varepsilon_i = \varepsilon_{0i} - \varepsilon_{\infty i}$ represents the dielectric strength of the corresponding mode, α is the distribution factor, G and S indicate the Goldstone and soft modes respectively, $\in_0$ is the electric permittivity of free space and σ is the electric conductivity.

In the SmC* phase, when distortion in the helical structure takes place, it returns to its undistorted state with a certain characteristic time τ_G due to the elastic torque followed by the opposite dissipative viscous forces. In the presence of an external electric field, the electric torque on the molecules comes into play and hence the viscous elastic equation becomes [8]

$$K_\phi \sin^2\theta \frac{\delta^2\phi}{\delta^2 Z} - \gamma_\phi \sin^2\theta \frac{\delta\phi}{\delta E} = P_S E \sin\theta, \quad (2)$$

where K_ϕ is elastic constant, $\boldsymbol{\gamma}_\phi$ is the rotational viscosity and P_s is the spontaneous polarisation. The following expressions relate the dielectric strength $\Delta\varepsilon$ and the relaxation frequency f_G in the SmC* phase to the spontaneous polarisation P_s and tilt angle θ:

$$\Delta\varepsilon = \frac{P_s^2}{2\varepsilon_0 K_\phi q^2 \theta^2}, \quad (3)$$

$$\tau_G = \frac{\gamma_\phi}{K_\phi q^2}, \quad (4)$$

where q is the magnitude of the wave vector of the helical pitch in the SmC* phase and ε_0 is the free space dielectric permittivity.

By solving the above equations γ_ϕ may be written as [14]

$$\gamma_\phi = (1/4\pi\varepsilon_o \Delta\varepsilon f_G) \cdot (P_s/\theta)^2 \quad (5)$$

where f_G represents the Goldstone mode relaxation frequency. From Equation (5) the relaxation frequency can be determined to be

$$f_G = (1/4\pi\varepsilon_o \Delta\varepsilon \gamma_\phi) \cdot (P_s/\theta)^2. \quad (6)$$

4. Results and discussions

Figure 1 shows the real and imaginary parts of the complex dielectric permittivity for the DHFLC (Figures 1(a) and (b)) and FLC (Figures 1(c) and (d)) materials as a function of frequency at different biases. It can be seen clearly from Figures 1(a) and 1(c) that the dielectric permittivity (ε') for DHFLCs is larger than that of FLCs. The DHFLC material possesses an ultra short pitch providing much denser packing of dipoles in a small volume of the sample, ensuring the large values of the spontaneous polarisation due to the alignment of the dipoles on the application of the electric field. Hence the value of ε' for DHFLCs is larger [19] than that of FLCs. When we compared the dielectric loss (ε'') in DHFLC and FLC materials (Figures 1(b) and (d)) we observed that the relaxation frequencies are larger in DHFLCs compared to those in FLC material. Moreover, the relaxation frequency increases with bias in the case of the FLC material. The large value of spontaneous polarisation in the case of the DHFLCs is clearly reflected in Figure 2 where it has been compared with the spontaneous polarisation value of FLCs.

We also measured the effect of different low-frequency measuring voltages on the dielectric permittivity and dielectric loss in DHFLC material at constant bias of 1 V. The changes in the real and imaginary parts of the permittivity at different ac measuring voltages at a fixed bias (1 V) are shown in Figure 3(a) and (b). The dielectric permittivity increases with ac measuring voltage. The dielectric loss also increases with ac measuring voltage, which gives an indication

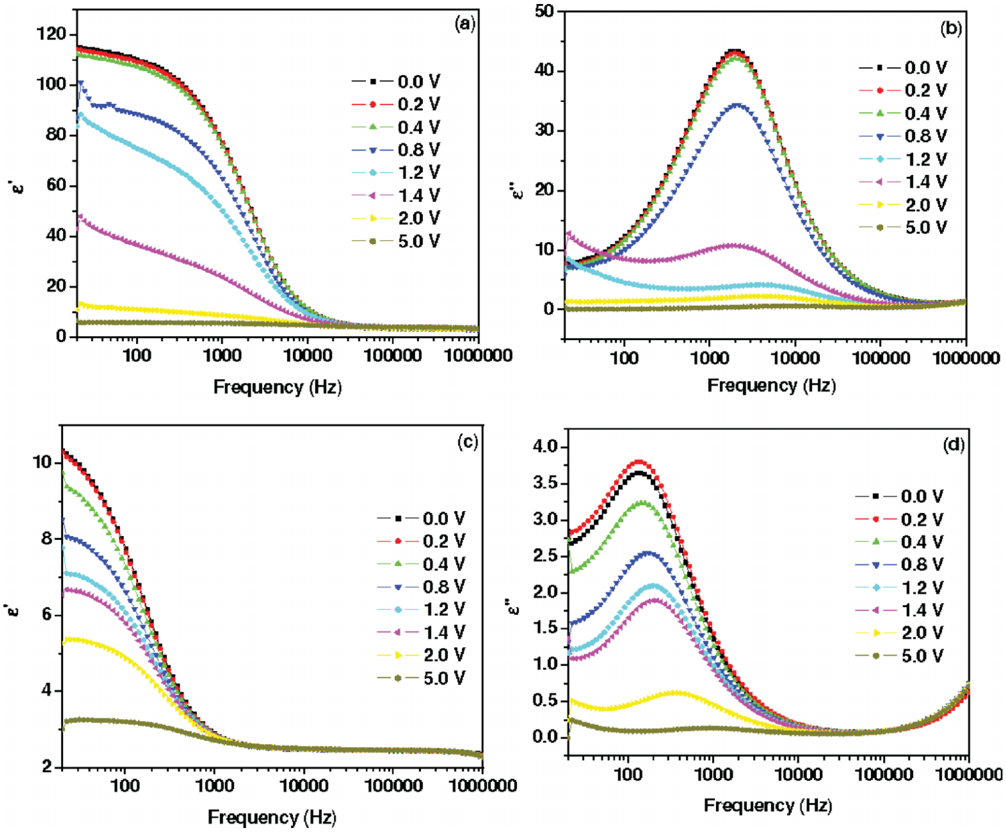

Figure 1. (Colour online) (a) Dielectric permittivity and (b) dielectric loss for DHFLCs, and (c) dielectric permittivity and (d) dielectric loss for FLCs as a function of the log of frequency (log ν) at different biases in 3.5 μm thick cells at room temperature.

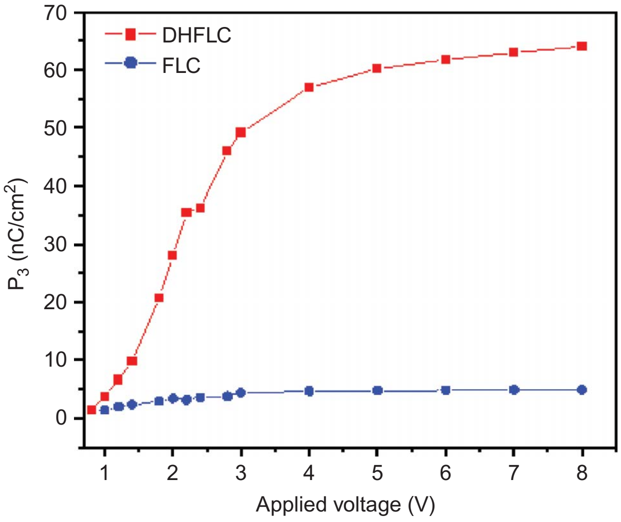

Figure 2. The variation of spontaneous polarisation with applied voltage for DHFLC and FLC cells.

of the enhancement in the conductivity. Hence, it will influence the relaxation frequency which decreases with an increase in the measuring voltage. To obtain the relaxation frequency of the Goldstone mode, dielectric data were analysed by fitting the experimental data to the Cole–Cole function. The bias dependence of the relaxation frequencies of the Goldstone mode for DHFLCs and FLCs are presented in Figures 4(a) and (b), respectively. The decrement in relaxation frequency continues up to a certain value of the applied voltage below the threshold voltage. The decrement in relaxation frequency can also be seen through the behaviour of the dielectric decrement with bias, which is shown in Figure 4(c). In the case of DHFLCs, the low field causes only a distortion of the helix, which explains the fast electro-optic response and the fast relaxation when the field is removed. Above a certain threshold field, the helix becomes completely unwound. So, beyond this certain voltage, i.e. the threshold voltage, the relaxation frequency increases as the applied voltage is increased. On the other hand, it is a well-known fact that the relaxation frequency either remains independent of bias or increases with applied field in the case of conventional FLCs [20, 21]. It is worth noting here that the voltage range for FLCs in which the relaxation frequency remains almost invariant is the same as that where the relaxation frequency decreases with bias in the DHFLC material. Moreover, the magnitude of the

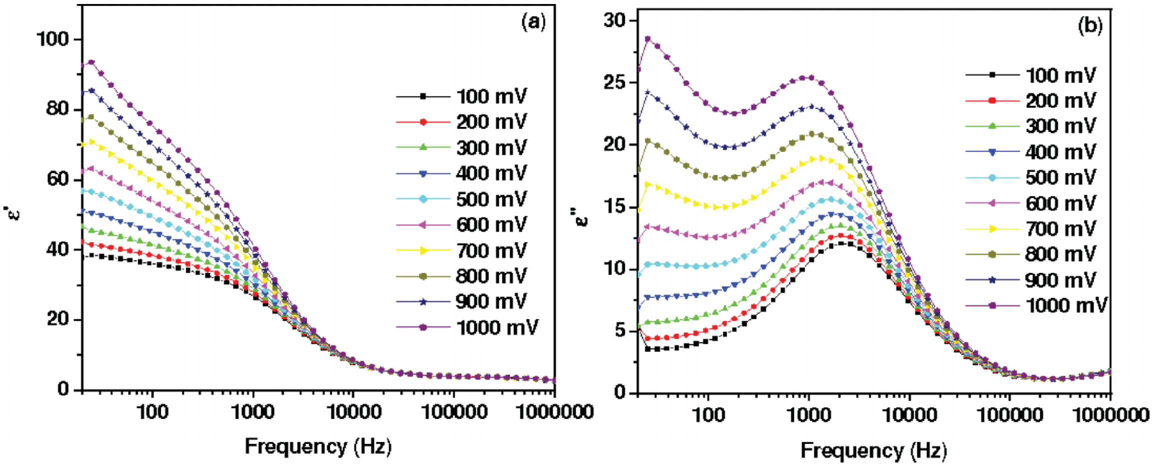

Figure 3. (Colour online) (a) Dielectric permittivity and (b) dielectric loss for DHFLCs as a function of the log of frequency (log ν) at different measuring voltages at a fixed bias of 1 V in 3.5 μm thick cells at room temperature.

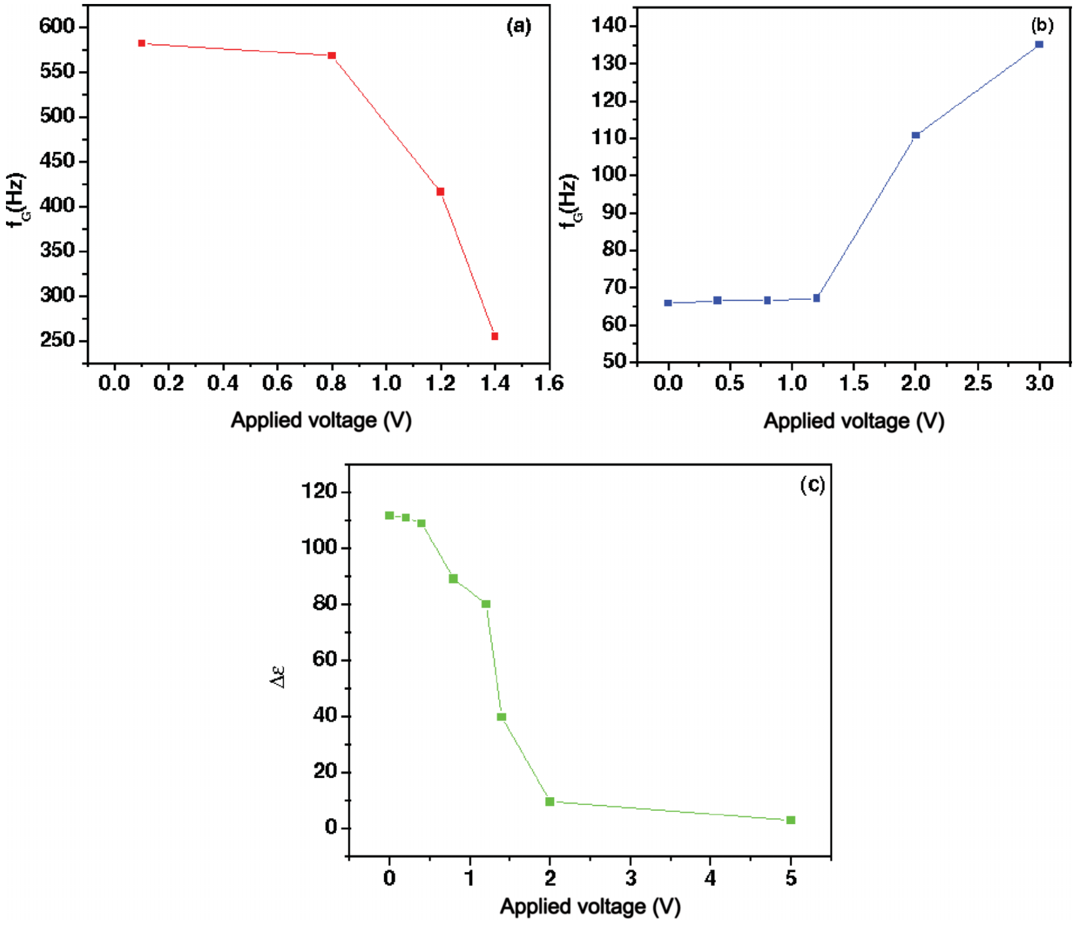

Figure 4. (Colour online) The behaviour of (a) the calculated Goldstone mode relaxation frequency for DHFLCs, (b) the calculated Goldstone mode relaxation frequency for FLCs and (c) the dielectric decrement for DHFLCs with different voltages at room temperature.

relaxation frequency in the case of the DHFLCs is somewhat greater than that of the FLCs. This large difference in the magnitude of the relaxation frequency can be explained by Equation (6), as the value of P_s/θ is much larger for DHFLCs as reflected in the inset of Figure 5.

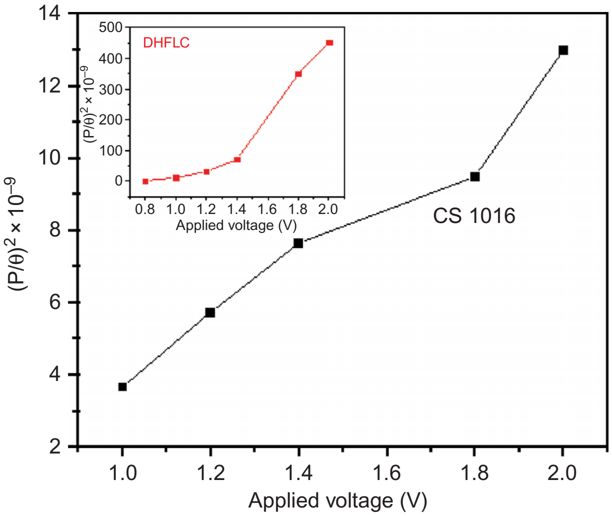

Figure 5. The variation of $(P_s/\theta)^2$ with applied voltage for the FLC cell. The behaviour for DHFLCs is shown in the inset.

The reason behind the decrement of the Goldstone mode relaxation frequency in DHFLCs lies in the mechanism of the unwinding/winding of the helix. It was reported earlier that the unwinding of the helix takes place rapidly as compared to the winding, i.e. the rise time (time taken in the unwinding) is less than the decay time (time taken in the winding) [22]. Moreover, it is expected that unwinding takes place under the constraint of applied dc electric field (beyond the threshold value) whereas the winding of the helix occurs in a natural way that depends on the material parameters such as the elastic constant, rotational viscosity etc. In DHFLC material, the contribution of viscosity to the winding process will be more prominent in comparison to FLCs. The viscosity is a property of the bulk. The switching in the DHFLC material is due to the combination of bulk and surface effects meaning that the viscosity plays an important role in the switching phenomenon of DHFLCs. In contrast, switching in FLCs is mainly due to surface effects [23] and hence the viscosity does not play a major role in the switching phenomenon of FLCs [24]. When we apply the dc bias field parallel to the smectic layers, a deformation in the helical structure occurs due to the alignment of the polarisation vector in the field direction. The molecules at the surfaces of the substrate may be more sensitive to the field in comparison to those wound in the bulk and they tend to align parallel to the surfaces [24]. On further increasing the bias, the molecular orientation in the bulk switches to add its contribution to the molecular orientation–reorientation process.

Figure 6 shows the optical micrographs at various dc voltages for the DHFLC material. It can be seen from the figure that the texture of the DHFLC samples changes (colour and intensity) when they are viewed under a polarising optical microscope, due to distortion in the helical structure. Figure 7 shows the corresponding micrographs of the FLC material in which no remarkable change is observed. The behaviour of the Goldstone mode relaxation frequency (f_G) in the presence of dc bias can be determined with the help of Equation (6), which shows the dependence of f_G on the material constants such as θ, $\Delta\varepsilon$ and γ_ϕ. The values of P_s and θ for both the materials change with the bias in such a way that their combined contribution $(P_s/\theta)^2$ follows the same trend. The variation of $(P_s/\theta)^2$ with the applied bias can be seen clearly from Figure 4. It is also observed that the value of the dielectric strength ($\Delta\varepsilon$) decreases with electric field for both the materials and follows the same trend.

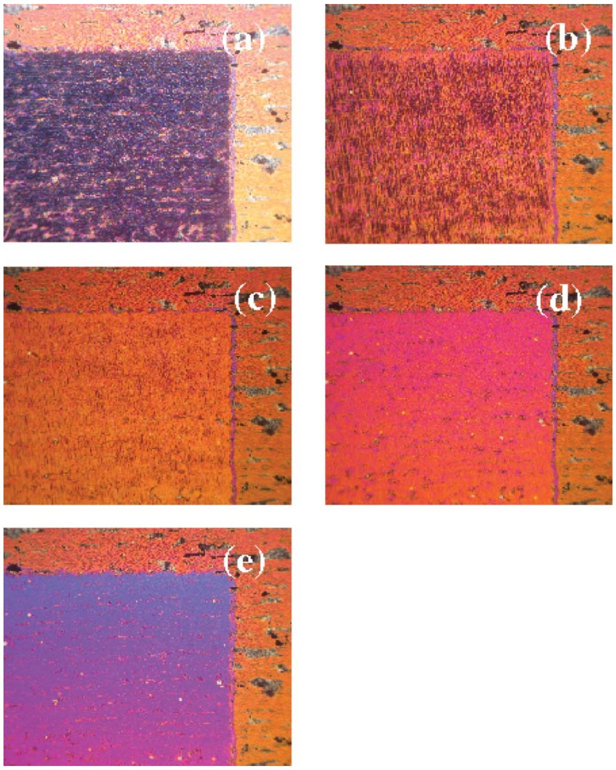

Figure 6. Optical micrographs of the 3.5 μm DHFLC cell at room temperature where the bias voltage is changed from 0 to 2.5 V: (a) 0 V, (b) 0.9 V, (c) 1.7 V, (d) 2.1 V and (e) 2.5 V.

The decrement in f_G in the case of DHFLCs is due to the high rotational viscosity possessed by the material in comparison to FLCs. Due to the higher viscosity, the effect of viscous forces on the molecules dominates over the electric torque on the molecules in the winding process resulting in comparatively a much larger decay time. The variation of the rotational viscosity with the dc bias for both the DHFLC and FLC materials is shown in Figure 8. One can see

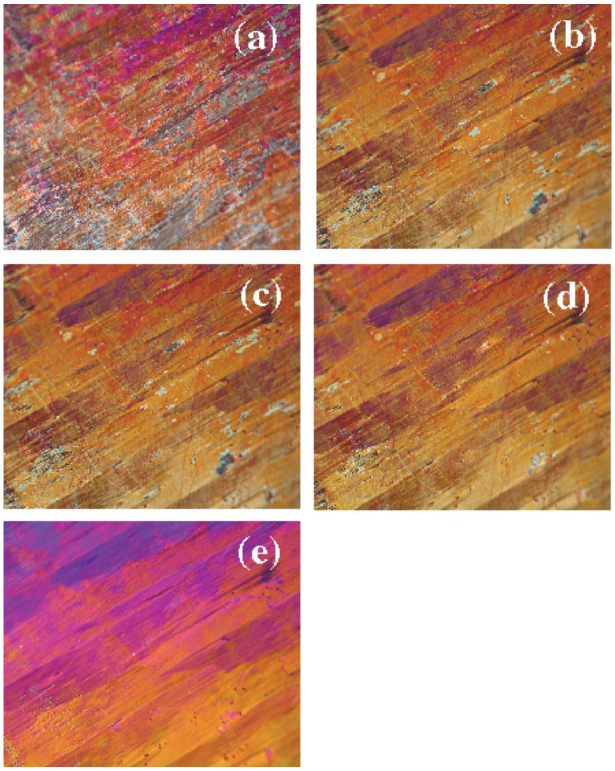

Figure 7. Optical micrographs of the 3.5 μm FLC cell at room temperature where the bias voltage is changed from 0 to 20 V: (a) 0 V, (b) 0.8 V, (c) 1.2 V, (d) 2.8 V and (e) 20 V.

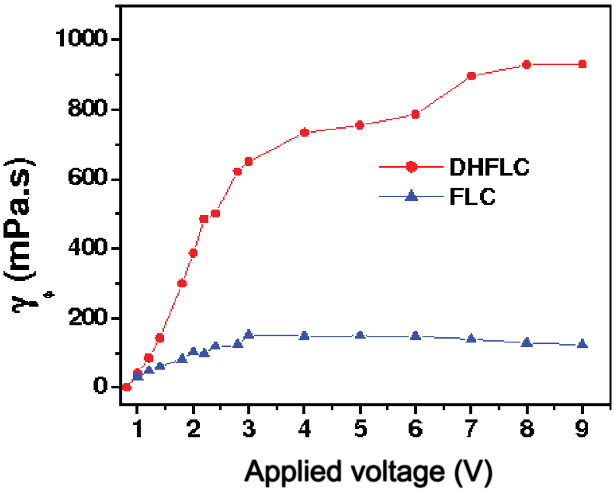

Figure 8. The variation of rotational viscosity with applied voltage for DHFLC and FLC cells.

clearly that the change in rotational viscosity with bias is sharp for the DHFLC material whereas it is gradual for the FLC material. Also the amplitude of viscosity is larger for the DHFLC material. When we increase the bias gradually from zero, the rotational viscosity increases rapidly and at the same time $(P_s/\theta)^2$ increases gradually, giving the possibility of a critical balancing of the elastic constant and rotational viscosity, which yields the decrement in f_G (Equation (6)) in a DHFLC material which is not observed in FLCs. However, in the case of surface stabilised ferroelectric liquid crystals (SSFLCs), it is observed that the relaxation frequency decreases with applied voltage. Equation (5) cannot be applied to SSFLC cells where the helix is suppressed by surface interactions and the molecular director is no longer modulated along the normal to the smectic layers [25].

5. Conclusions

The behaviour of the Goldstone mode relaxation frequency in a low dc electric field regime below the threshold has been studied for a DHFLC material and compared with that of a conventional FLC material. It has been concluded that the decrement in relaxation frequency for the DHFLC material is a critical phenomenon that takes place due to the higher rotational viscosity providing an interesting balance with the elastic constant, which results in a decrement of the relaxation frequency. This study may be a useful tool to help understand the role of the rotational viscosity and surface effects in the helix winding and unwinding processes in FLCs.

Acknowledgements

The authors sincerely thank Dr Vikram Kumar, Director, National Physical Laboratory, for continuous encouragement and interest in this work. We sincerely thank Dr S.S. Bawa, Dr Indrani Coondoo, Mr Amit Choudhary and Ms Anu Malik for fruitful discussions. We are grateful to Dr Poonam Silotia of the University of Delhi, India for useful discussions. The authors AK and JP are grateful to UGC, New Delhi and CSIR, New Delhi, respectively, for financial assistance.

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