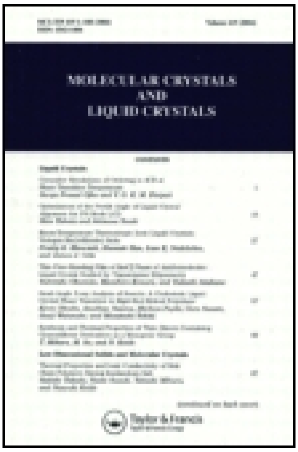

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Switching and Dielectric Behavior of Phase

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Switching and Dielectric Behavior of SmC_{α}^{*} Phase

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Switching and dielectric properties of a compound (S)-(+)-4-(1-methylheptyl) 4-[4-(3-hexanoyloxy prop-1-oxy) benzoyloxy]biphenylate are studied by means of polarization reversal methods and dielectric spectroscopy respectively. Two characteristics polarization peaks have been observed on application of triangular pulse in the SmC_{α}^{*} phase. The separation between two peaks has been decreases with increase in frequency and electric field. These two peaks merge into each other after a critical field of ~200 kV/cm. A relaxation mechanism has been detected under planar orientation of molecules for SmC_{α}^{*} phase in the high kHz region. This mode shows two different behaviors in two temperature ranges.

Keywords Chiral smectic phase; collective processes; dielectric relaxation mode; switching time

1. Introduction

The availability of chiral substances led directly to the discovery of novel and complex structure in liquid crystal systems like SmC_{α}^{*}, SmC_{β}^{*} and SmC_{γ}^{*} phases which are collectively classified as chiral smectic C subphases [1,2]. In these phases, the molecules within each layer are oriented with their long axis along a common direction represented by a tilt (θ) relative to layer normal and an azimuthal angle (α) which precess due to chirality forming a helical structure. On variation of the temperature, these structures with distinct macroscopic properties are formed due to different correlation of azimuthal angles between smectic layers. The structure of SmC_{α}^{*} phase is determined to be incommensurate nano scale helical pitch extended from three to eight smectic layers [3,4]. The azimuthal angle difference ($\Delta\alpha$) between two smectic layers in this phase is found to be vary with temperature and it decreases with decrees in temperature. Hence macroscopic properties of this phase changes with temperature which make its characterisation complicated. When symmetric triangular wave is applied across a capacitor cell filled with material in the SmC_{α}^{*} phase, two polarization peaks are observed in switching process similar to antiferroelectric SmC_{a}^{*} phase [2]. However, tri-state switching behaviour is not observed in this phase

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[1,2]. In the dielectric spectrum, single relaxation mode is observed generally in the SmC_α^* phase for planar oriented sample that exist in high kHz region with moderate value of dielectric strength [5–9]. On the other hand, two relaxation modes are observed for SmC_a^* phase in kHz region with small value of dielectric strength [7–11]. Hence the switching and dielectric characterization are important to ascertain the macroscopic properties and behaviour of SmC_α^* phase. On combining these two characterization techniques only, it is possible to specify the particular chiral smectic phase in new materials. We are reporting here switching and dielectric properties of the compound (S)-(+)-4-(1-methylheptyl)4-[4-(3-hexanoyloxyprop-1-oxy)benzoyloxy]-biphenylate which shows exceptionally wide temperature range of SmC_α^* phase.

2. Experimental

Polarization reversal current technique is applied for switching measurements. A symmetric squire and triangular wave from 1 Hz to 200 Hz applied across the series combination of the standard register ($R_{st} = 100\,k\Omega$) and the sample filled capacitor cell (R_{dc}) of thickness 5 μm. In order to apply sufficient electric field across the sample cell, waves obtained from the function generator (HM5030-4) have been amplified by a high voltage linear amplifier (FLC, model-A400). The applied voltage (peak to peak) and the voltage across R_{st} (and hence resultant current i) are displayed on the vertical inputs CH1 and CH2 of a digital oscilloscope, respectively. (Hameg, model-HM507). The transient current i passing through the C-R combination of the circuit has three components which is written as [12]

$$i = i_R + i_C + i_P = \frac{V}{R} + C\frac{dV}{dt} + \frac{dP_s}{dt} \tag{1}$$

Where, $i_R = V/R$ is the conductive term through the effective resistance $R = R_{dc} + R_{st}$ of the circuit; V is the amplitude of the applied voltage, $i_C = C\frac{dV}{dt}$ is the capacitive term (charge accumulation) and $i_p = \frac{dP_S}{dt}$ is the polarization reversal (switching) current or depolarization current due to the spontaneous polarization P_S. Capacitive term becomes zero when symmetric triangular wave is applied across the sample.

The dielectric data are acquired for the 5 and 10 μm thick planar aligned sample during cooling cycle in the frequency range of 1 Hz to 10 MHz using frequency response analyzer (Solartron, model SI-1260) coupled with a Solartron dielectric interface (model-1296). An ac electric field of 0.5 V_{rms} is applied through electrodes.

3. Results and Discussion

The investigated compound is following phase sequences as obtained by DSC measurements at the scanning rate of 2.5°C/min in the cooling cycle [13].

I (98.8°C) SmA^* (75.4°C) SmC_α^* (53.2°C) Cr2 (50.1°C) Cr1

The above mentioned phase sequence is also confirmed by optical textures and miscibility studies.

When the symmetric wave (Squire and triangular) of sufficiently high amplitude is applied across the sample, it reorients the dipoles between two polarization states (i.e., either parallel or anti parallel to the applied electric field). The molecular

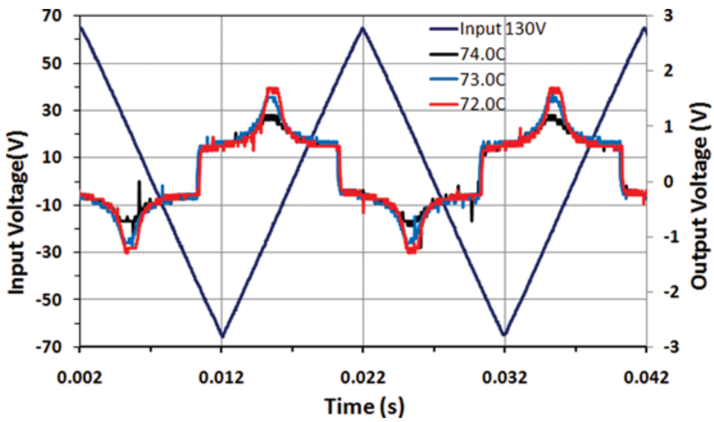

Figure 1. Input triangular waveform across the series combination of cell filled with materials and 100 kΩ standard resistor on channel I (50 Hz, 130 Vpp) and the resulting voltage waveform across 100 kΩ standard resistor on channel II.

realignment is observed as a hump (polarization reversal current) in the output voltage/current. In the orthogonal SmA* phase, polarization in each smectic layer is cancelled due to symmetry property, hence no polarization hump is observed in this phase.

Below SmA*-SmC_{α}^{*} transition, when a triangular pulse of 130 Vpp (peak to peak) and 50 Hz frequency is applied, a weak hump due to orientation of dipole is observed as shown in Figure 1. The strength of this hump increases on lowering the temperature but not significantly. At 71.5°C, the polarization hump split into two peaks which increase significantly on lowering the temperature as shown in Figure 2. This indicates two different behaviour of the SmC_{α}^{*} phase in the studied compound. On lowering the temperature, these two peaks grown fully and are separated further. The hump area of these two peaks also increases on lowering the temperature.

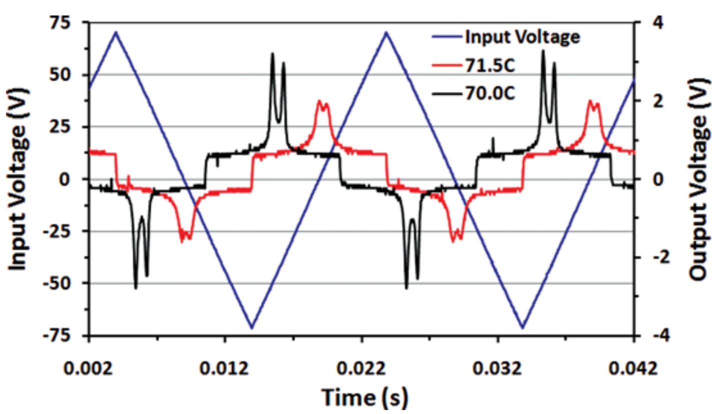

Figure 2. Emergence of two polarization peaks in SmC_{α}^{*} phase. 70.0°C data was shifted on time scale for clarity.

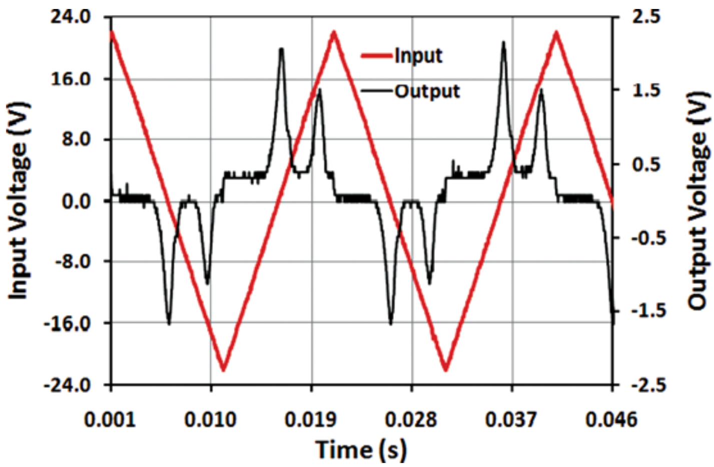

Figure 3. Fully grown two polarization peaks at 65.0°C of SmC_{α}^{*} phase. (Figure appears in color online.)

It is remarkable to note that voltage required to switch the dipole in two polarization states decreases on decreasing the temperature as shown in Figure 3. At 65°C two peaks are fully grown on application of 42 V peak to peak only. It suggests that structure becomes soft for orientation of dipoles and switching of polarization states on lowering the temperature.

It is also observed that the separation between two peaks decreases upon increasing the strength of the applied pulse. Figure 4 shows output waveform with increasing input voltage at 65°C. The two peaks merge into each other after a critical field of ~200 kV/cm at each temperature.

The observation of two characteristic polarization peaks in the switching studies and their electric field dependence indicates the structure under studied is SmC_{α}^{*}

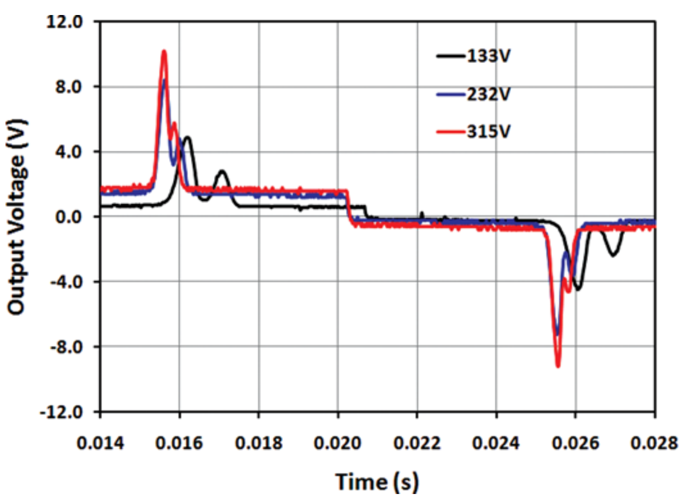

Figure 4. Emergence of two polarization peaks with electric filed at 65°C of SmC_{α}^{*} phase.

phase. However, two characteristic polarization peaks is also observed in antiferroelectric SmC_a^* phase. To ascertain the structure of the phase, we have also carried out dielectric measurement of the compound under planar anchoring. In the dielectric spectrum of SmA^* and SmC_α^* phases, single relaxation mode is observed. The measured dielectric spectra in SmA^* and SmC_α^* phases are well described by single Debye and Cole-Cole type behaviors respectively as shown in Figure 5 [13]. To get the dielectric parameters of the observed mode, the measured data is analyzed using generalized Cole-Cole equation [14–16]

$$\varepsilon^* = \varepsilon' - j\varepsilon'' = \varepsilon'(\infty) + \sum_i \frac{(\Delta\varepsilon)_i}{1 + (j\omega\tau_i)^{(1-h_i)}} + \frac{A_1}{\omega^n} + \frac{\sigma(dc)}{j\varepsilon_0\omega} - jA\omega^m \tag{2}$$

where $\varepsilon'(\infty)$ is high frequency limiting value of relative permittivity, $\Delta\varepsilon_i$, τ_i and h_i are dielectric strength, relaxation time (inverse of angular relaxation frequency) and

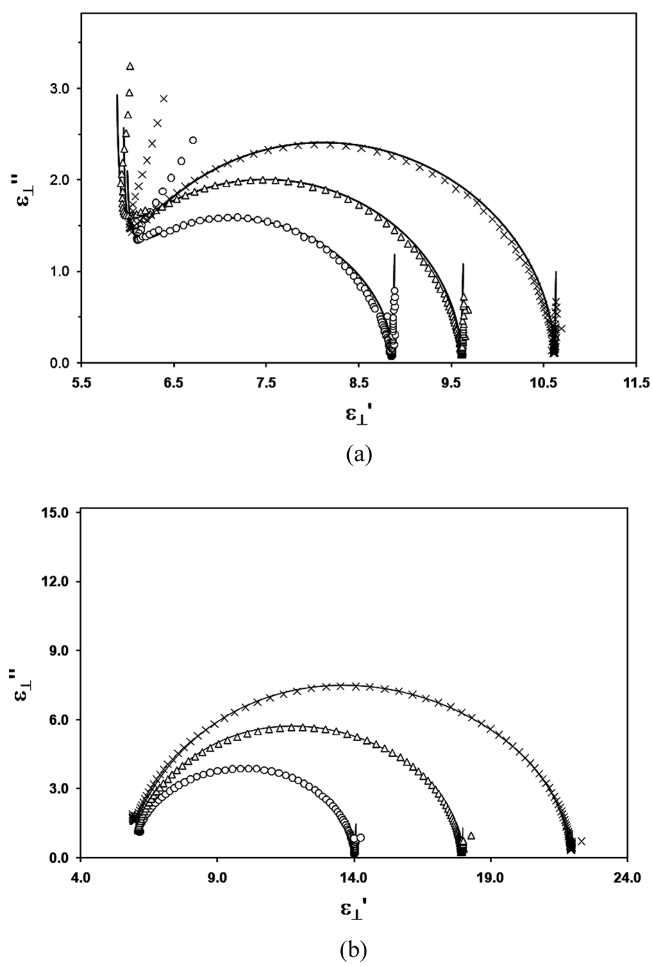

Figure 5. Cole-Cole arcs in (a) SmA^* phase at 78.0°C (circle), 76.5°C (triangle) and 75.0°C (cross) and (b) SmC_α^* phase at 74.1°C (circle), 72.0°C (triangle), and 70.2°C (cross). Solid line with the experimental data shows best fitting of equation (2).

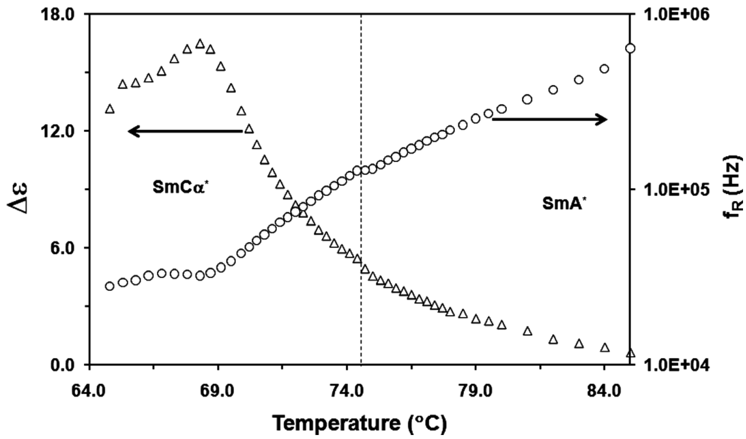

Figure 6. Variation of relaxation frequency (open circle) and dielectric strength (open triangle) of observed mode with temperature in the SmA* and SmC$_\alpha^*$ phases.

symmetric distribution parameter ($0 \leq h_i \leq 1$) of ith mode respectively. The third and fourth terms in Eq. (2) represent the contribution of electrode capacitance and ionic conductance at low frequencies where A_1 and n are constants [12]. σ(dc) is the ionic conductance. The fifth term in Eq. (2) is added to partially account for ITO effect, where A and m are constants as for as correction terms are small. The dielectric strength ($\Delta\varepsilon$) and frequency (f_R) of the relaxation modes observed in the SmA* and SmC$_\alpha^*$ phases are shown in Figures 6.

In the SmA* phase, $\Delta\varepsilon$ increases whereas f_R decreases continuously with decrease of temperature. This mode is associated with director tilt fluctuation of molecules in the smectic layers i.e., soft mode [5–11]. On passing through SmA*-SmC$_\alpha^*$ transition, clear changes in the slopes of f_R and $\Delta\varepsilon^{-1}$ have been observed (see Fig. 5). On lowering the temperature of the sample in the SmC$_\alpha^*$ phase, f_R of this mode decreases moderately and $\Delta\varepsilon$ increases sharply with decrease in temperature till 69.0°C. This is in distinction to the dielectric behavior of SmC$_a^*$ phase, where two relaxation modes are observed with small and constant value (~1) of dielectric strength [8–11]. Below 69.0°C in the SmC$_\alpha^*$ phase, f_R becomes approximately invariant with temperature but $\Delta\varepsilon$ decreases slowly with decrease in temperature. This clearly indicates that the behaviour of observed mode in SmC$_\alpha^*$ phase below 69.0°C is different as compared to those observed above 69.0°C. Similar observation is also visible during switching studies.

4. Conclusions

- Switching and dielectric studies confirmed SmC$_\alpha^*$ phase in the investigated compound below SmA* phase.
- Characteristics polarization peaks of SmC$_\alpha^*$ phase have been observed on application of triangular pulse below 4°C to SmA*- SmC$_\alpha^*$ transition.
- The separation between two peaks has been decreases with electric field. These two peaks merge into each other after a critical field of ~200 kV/cm at each temperature.

- In the SmC_{α}^{*} phase, observed relaxation mode shows two different behaviour in different temperature ranges.
- Switching and dielectric studies confirm that SmC_{α}^{*} phase shows two different macroscopic properties at high and low temperature region of the investigated compound.

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