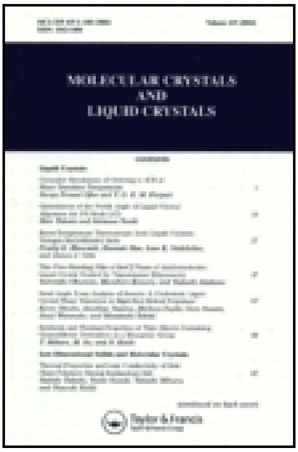

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Praveen Malik [a], Ashok Chaudhary [a], Rohit Mehra [a] & K. K. Raina [b]

[a] Department of Physics, Dr. B. R. Ambedkar National Institute of Technology, Jalandhar, Punjab, India

[b] Materials Research Laboratory, School of Physics and Materials Science, Thapar University, Patiala, Punjab, India

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Dielectric Studies and Memory Effect in Nanoparticle Doped Ferroelectric Liquid Crystal Films

PRAVEEN MALIK,[1] ASHOK CHAUDHARY,[1] ROHIT MEHRA,[1] AND K. K. RAINA[2]

[1]Department of Physics, Dr. B. R. Ambedkar National Institute of Technology, Jalandhar, Punjab, India
[2]Materials Research Laboratory, School of Physics and Materials Science, Thapar University, Patiala, Punjab, India

Dispersed liquid crystal materials consist of silica nanoparticles doped into ferroelectric liquid crystal have been prepared and studied in smectic C phase. It is found that dielectric permittivity decreases with increasing the silica concentration. Goldstone mode has been observed at a relaxation frequency of ~250 Hz in the smectic C* phase. Relaxation frequency increases with increasing the silica concentration and temperature in the smectic C* phase and decreases as we approaches toward the SmA phase. Bias voltage dependence on permittivity indicates a memory state.*

Keywords Dielectric permittivity; ferroelectric liquid crystal; Goldstone mode; memory; relaxation frequency; silica nanoparticles

1. Introduction

Dispersed liquid crystal materials have attracted significant interest due to their practical and technological application in displays [1–3]. Recently, dispersion of nanoparticle (NPs) in liquid crystals (LC) has shown considerable improvement in existing properties of LC material. Dispersion of various NPs e.g., CdSe, $BaTiO_3$, $Sn_2P_2S_6$, Au, Pd, MgO, ZnO, SiO_2, TiO_2, etc. into liquid crystals have shown tremendous interest due to unique thermal, mechanical and electrical properties of nanoparticles [4–13]. Among the variety of nanoparticles, silica nanoparticle in LCs materials are currently used for memory based application [14–16]. In the beginning, dispersion of NPs into nematic liquid crystals was focused on the dielectric and phase transition studies. In the present work, efforts have been made to study the effect of silica nanoparticle on the dielectric studies of ferroelectric liquid crystal (FLC) in the frequency range of 50 Hz – 1 MHz. The effect of bias voltage on dielectric permittivity and memory state is also a subject of our current interest.

Address correspondence to Praveen Malik, Department of Physics, Dr. B. R. Ambedkar National Institute of Technology, Jalandhar 144 011, Punjab, India. Tel.: +91-9888382021; Fax: +91-181-2690 320; E-mail: pmalik100@yahoo.com

2. Experimental

FLC material trade name KCFLC10R (obtained from Kingston Chemicals, UK) [17] has been used as host material. The phase sequence of FLC is following:

$$Cr. \overset{5^\circ C}{\longleftrightarrow} SmC^* \overset{64.5^\circ C}{\longleftrightarrow} SmA \overset{99.5^\circ C}{\longleftrightarrow} N^* \overset{112^\circ C}{\longleftrightarrow} I$$

Silica nanoparticle (SNP) (M/S Sigma Aldrich, India) has particle size of 5–15 nm and molecular weight equal to 60.08 [18]. SNP in different concentrations (0.01, 0.02, 0.03 wt./wt.% of FLC) was doped into FLC. To get a completely homogenize mixture of SNP and FLC, both materials were properly mixed at room temperature and than ultrasonication was done at a frequency of 36 kHz for one hour. Planar aligned cells of thickness 12 μm made of indium tin oxide (ITO) coated glass substrates were used. A solution of nylon 6/6 and m-cresol was spin coated on ITO coated glass substrates and then uni-directionally rubbed for planar alignment. The prepared homogenize mixtures were filled in empty sample cells by capillary action at the isotropic temperature of FLC. Filled sample cells were placed in hot stage (model THMS600) interfaced with temperature controller (model Linkam TP94). The dielectric studies have been carried out in SmC* phase in the frequency range from 50 Hz to 1 MHz by LCR meter (model Fluke-PM6306). The sample cells were calibrated with air and benzene as standard reference [19].

3. Results and Discussion

3.1. *Dielectric Studies*

Dielectric spectroscopy is an important tool to understand the molecular relaxation process in liquid crystals. The complex dielectric permittivity in terms of real (ε') and imaginary (ε'') parts is given by the relation [20]:

$$\varepsilon^*(\omega, \tau) = \varepsilon'(\omega, \tau) - i\varepsilon''(\omega, \tau) \tag{1}$$

Where $\omega = 2\pi f$ is the angular frequency of applied electric field and τ is the relaxation time. The dielectric responses consist of Goldstone mode (GM) which arises due to fluctuation in azimuthal angle in the SmC* phase and is given by [21]:

$$\varepsilon^*(\omega, \tau) = \varepsilon_\infty + \frac{\Delta\varepsilon_{GM}}{(1 + i\omega\tau)^{1-\alpha_{GM}}} \tag{2}$$

Where, $\Delta_{GM} = \varepsilon_0 - \varepsilon_\infty$, α_{GM} are dielectric strength and distribution parameter for GM mode, respectively. ε_0, ε_∞ are the low and high frequency dielectric permittivity. The temperature dependence of the real and imaginary parts of the dielectric permittivity at 200 Hz and 1 kHz for different silica doped samples are shown in Figure 1. Figure 1(a, b) shows that at 200 Hz, ε' and ε'' increases up to SmC* and decreases near SmC*-SmA transition temperature. Similar temperature dependence behaviour on permittivities was also observed at 1 kHz with lower value which can be clearly seen in Figure 1(c, d). Figure 1(a) depicts that permittivity decreases with increasing the

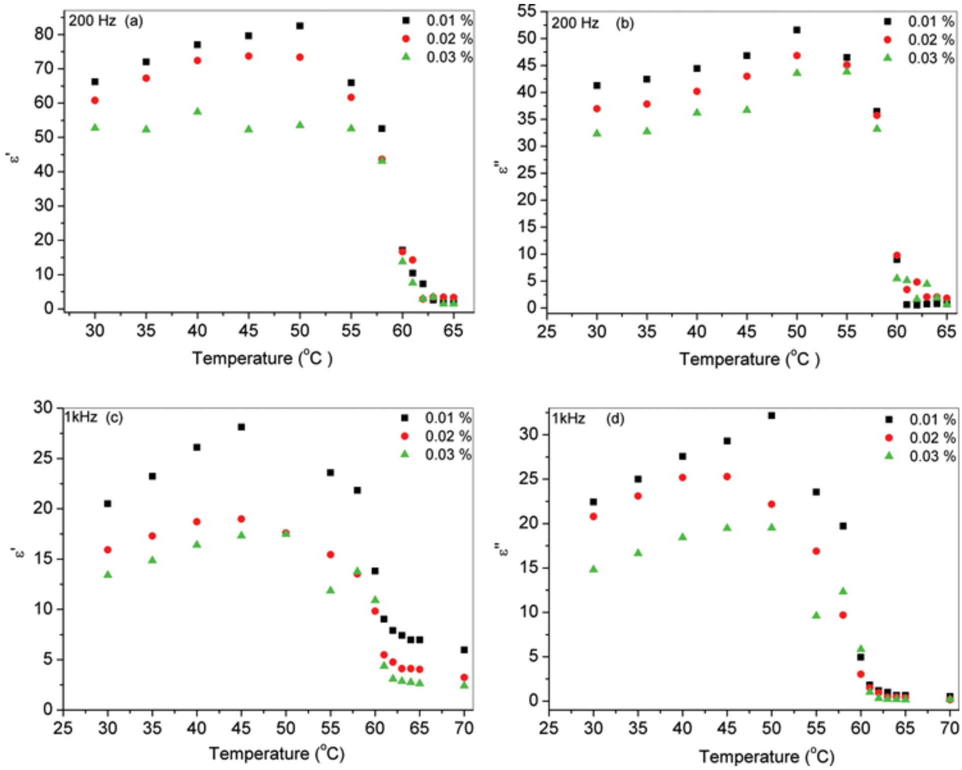

Figure 1. Temperature dependence of the dielectric permittivity (a, b) at 200 Hz and (c, d) at 1 kHz for silica doped samples. (Figure appears in color online.)

concentration of silica. This decrease in permittivity with increasing the silica concentration may be due to the strong intra-molecular forces between FLC and silica molecules. At high concentration of silica (0.03%), FLC molecules are highly disordered due to strong anchoring forces and hence results a lower permittivity than 0.01 and 0.02% doped samples.

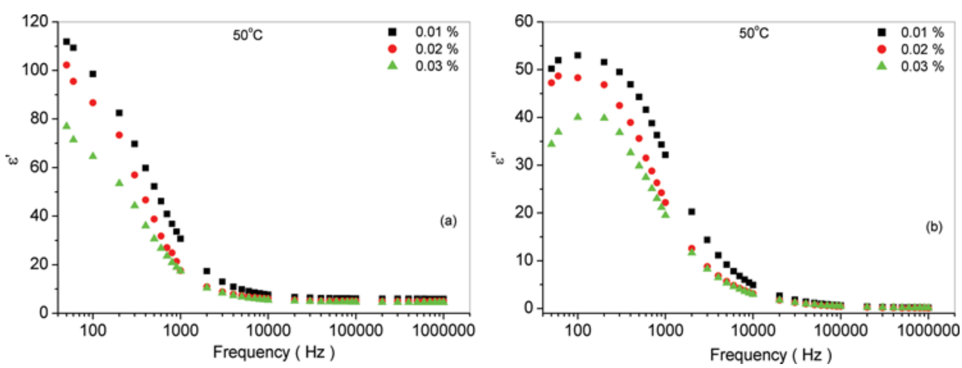

Figure 2. Frequency dependence of the dielectric permittivity for different silica concentrated samples at 50°C. (Figure appears in color online.)

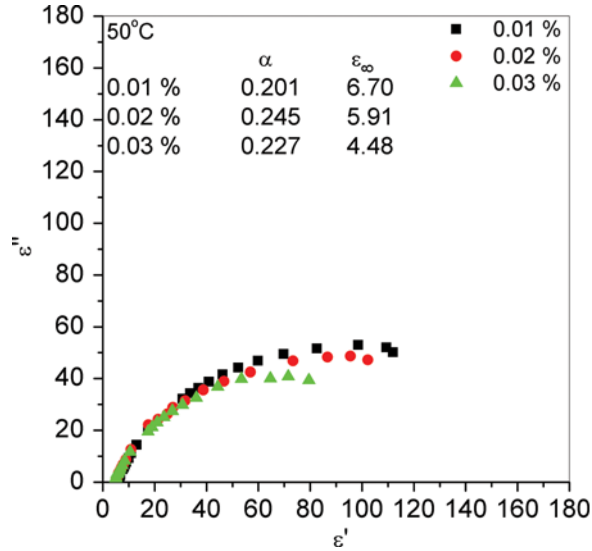

Figure 3. Cole-Cole plots in SmC* phase at 50°C. (Figure appears in color online.)

The frequency dependence on dielectric permittivity (ε', ε'') in all samples at 50°C are shown in Figure 2. Figure 2(a) shows that permittivity decreases with increasing the silica concentration however at higher frequencies ($\geq$1 kHz) no significant variation in permittivity was observed. Figure 2(b) follow a typical frequency dependence on ε'' which infers that single relaxation peak appears in the low frequency region ~250 Hz. The appearance of single peak in low frequency domain suggests a Goldstone mode. However some other expected relaxation modes like soft mode,

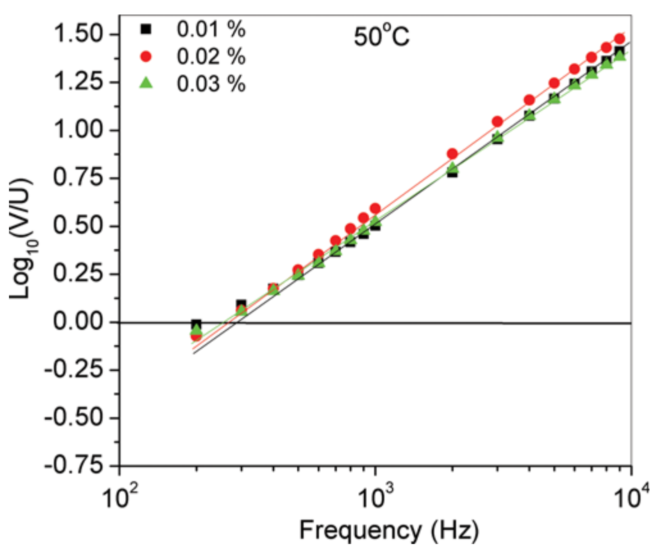

Figure 4. Frequency versus $\log_{10}$ (V/U) for measurement of relaxation frequency and distribution parameter at 50°C. (Figure appears in color online.)

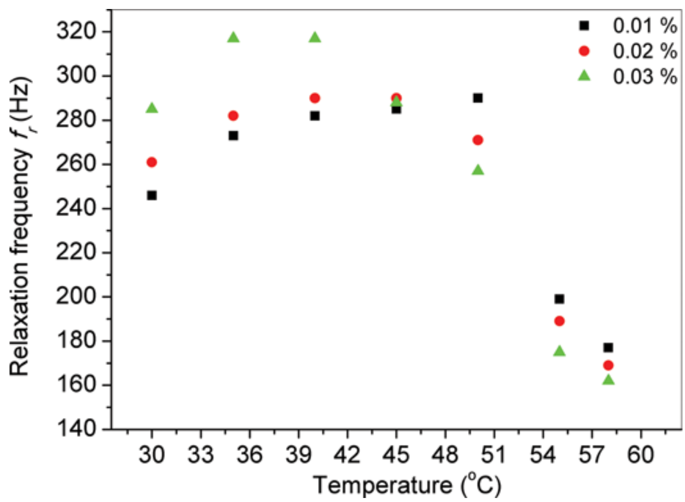

Figure 5. Temperature dependence of the relaxation frequency at different silica concentration samples. (Figure appears in color online.)

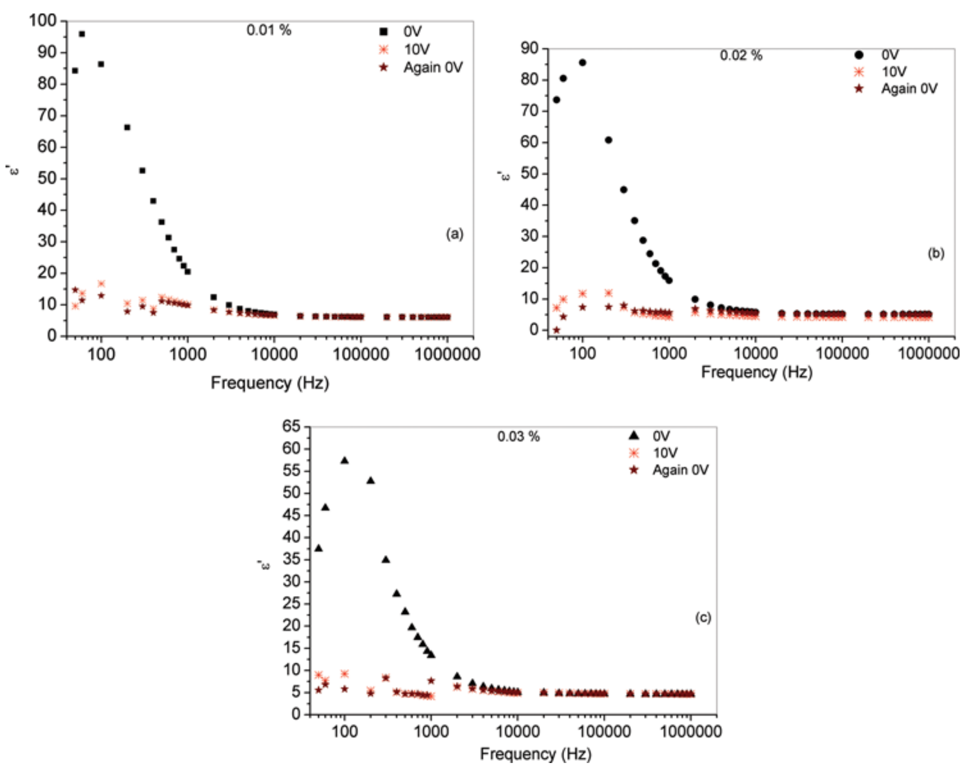

Figure 6. Dielectric permittivity as a function of frequency at 0 V, 10 V and again 0 V at 30°C for (a) 0.01%, (b) 0.02%, and (c) 0.03% doped samples. (Figure appears in color online.)

surface like mode, domain mode, new relaxation mode may be seen in the SmA-N* transition temperature, in different surface configuration and bias effects. A relaxation behaviour in the form of Cole-Cole plot obtained from Eq. (2) in SmC* phase at 50°C is seen in Figure 3.

The relaxation frequency (f_r) and distribution parameter (α) for Goldstone mode in silica doped samples have been calculated using the following Cole-Cole equations [22]:

$$\frac{V}{U} = (\omega\tau)^{1-\alpha} \tag{3}$$

$$V = [(\varepsilon_0 - \varepsilon')^2 + (\varepsilon'')^2]^{1/2} \tag{4}$$

$$U = [(\varepsilon' - \varepsilon_\infty)^2 + (\varepsilon'')^2]^{1/2} \tag{5}$$

The frequency versus $\log_{10}(V/U)$ at a specific temperature follow an Arrhenius type behaviour in the form of straight line and shown in Figure 4 [22,23]. The intercept of straight line and slope on X-axis corresponds to relaxation frequency and distribution parameter. The calculated relaxation frequency as a function of temperature

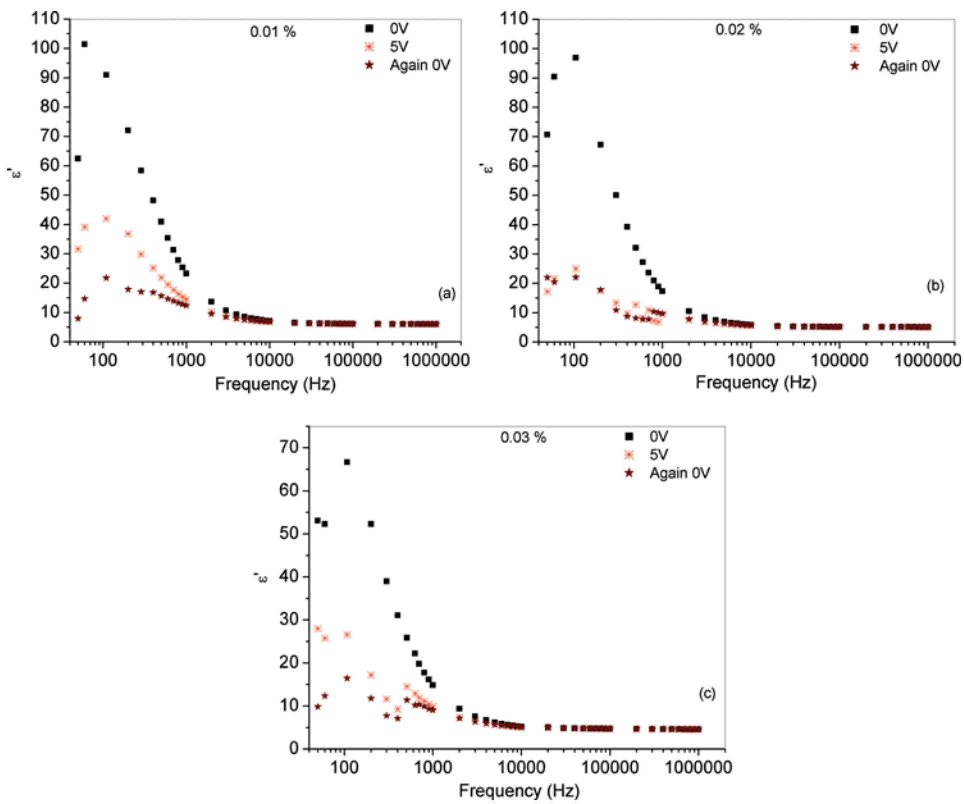

Figure 7. Dielectric permittivity as a function of frequency at 0 V, 5 V and again 0 V at 35°C for (a) 0.01%, (b) 0.02%, and (c) 0.03% doped samples. (Figure appears in color online.)

at different silica concentration in SmC* phase is shown in Figure 5. Figure 5 shows that f_r is temperature dependent and increases with increasing the temperature as well as silica concentration. However an interesting point is that up to 45°C, f_r increases and beyond 45°C, it continuously decreases near the SmA phase with increasing the temperature and silica concentration.

3.2. *Memory Effect*

Figures 6(a–c) show the frequency dependence on dielectric permittivity (ε') at different bias voltages (0 and 10 V) at 30°C. At 0 V, high value of ε' was notice in the SmC* phase which is due to the GM contribution. It is found that the value of ε' decreases as the bias voltage is changed from 0 V to 10 V. This sudden decrease in ε' is due to the suppression of GM mode and corresponding molecules reach in unstable state. It is found that again switch off the bias voltage (0 V) and hold the sample in off condition for ~60–120 seconds, the ε' value does not much change and remained as it was in the presence of bias voltage of 10 V. This confirms that FLC molecules did not come back in their stable state as they were before applying the bias and hence sample shows a memory state. Similar type of memory effect was also observed at (0, 5 and again 0 V) at 35°C as shown in Figure 7(a–c). The influence of bias voltages (1, 3, 5 and 10 V) at different silica amount are also presented in Figure 8(a–c). It can be seen that in the presence of bias effect permittivity decreases to a minimum value due

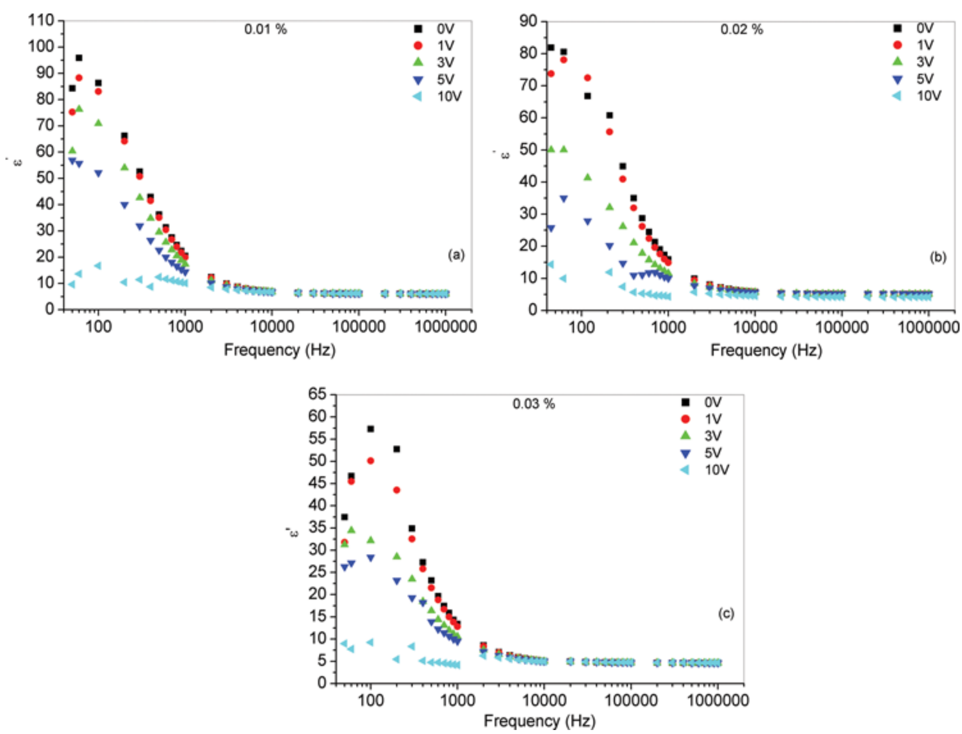

Figure 8. Bias voltage dependence as a function of frequency at 30°C for (a) 0.01%, (b) 0.02%, and (c) 0.03% doped samples. (Figure appears in color online.)

to the suppression of GM mode. Authors believe that to support and confirm the memory effects in silica doped samples, some additional studies i.e., optical parameters, textures and aging effect are also required.

4. Conclusions

In summary, silica nanoparticles in different concentration were doped into FLC material were prepared and studied. The dielectric spectroscopy method has been applied to study their properties in the SmC* phase. Dielectric permittivity decreases with increasing the silica concentration. Goldstone mode was also observed in the low frequency range at ~250 Hz. The decrease in permittivity is due to the strong anchoring forces develop between higher concentrated silica and FLC molecules. Our results show that relaxation frequency increases with increasing the temperature from room to SmC* and decreases as we approaches towards $T_{C_A^*}$. Bias voltage dependence on dielectric permittivity indicates that silica doped FLC samples show a memory effect. However some additional experiment and related studies are also required to confirm the memory phenomena for practical applications.

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