ORIGINAL RESEARCH

Silver nanoparticles dispersed in nematic liquid crystal: an impact on dielectric and electro-optical parameters

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Abstract

In the present study, nematic liquid crystal (NLC) 4′-(Octyloxy)-4-biphenylcarbonitrile dispersed with three different concentrations of silver nanoparticles (Ag NPs) is characterized by electro-optical and dielectric spectroscopy method. The dispersion of Ag NPs into NLC material leads to the change in physical parameters such as dielectric permittivity, photoluminescence, threshold voltage, response time and splay elastic constant. The AC conductivity and threshold voltage of Ag NPs-dispersed composite system have increased significantly. The increase in the AC conductivity for the composite system is attributed to the Ag NPs-assisted induced charge transfer mechanism in the system. Moreover, the increase in AC conductivity value for Ag NPs/NLC composite system discloses its importance in photovoltaic solar cell.

Keywords Nematic liquid crystal · Silver nanoparticles · Dielectric anisotropy · Threshold voltage

Introduction

Liquid crystals (LCs) represent a fascinating class of soft condensed matter where the subtle balance between order and fluidity of the molecules exists. They show a range of exceptional properties that not only are fascinating from basic science point of view, but also hold tremendous potential for various innovative applications [1, 2]. Nematic liquid crystal (NLC) is the simplest of all LC phases and widely used in the ubiquitous liquid crystal displays (LCDs) and many nondisplay applications. With the advancement of technology, LCD also needs advancement such as low power consumption, faster response and greater vision angle. In past few years, much emphasis has been given to tailor the properties of liquid crystals by dispersing suitable nonmesogenic materials (dyes, polymers, nanoparticles, carbon nanotubes, quantum dots, etc.) into the host LC matrix [3–7]. Liquid crystals containing dispersed nanoparticles (NPs) of various shapes and sizes are promising materials for the display and sensor industries, fast electro-optical switchers and shutters, memory cells, tunable filters, nonlinear optical valves for optical processing systems, etc. [8–10]. The coupling of the anisotropic elasticity of LCs to the optical, conductive or magnetic properties of NPs is expected to have an additional benefit with the relevant technologies. It is thus imperative to control NPs interaction with LC molecules so that the macroscopic properties as a whole can be tailored for specific applications.

Among all the NPs, metal NPs are highly studied and extensively used because of their exploitable optical properties. Silver (Ag) NPs are one of the well-known and widely used metal NPs due to their conductivity, chemical stability, intense calorimetric effect at nanoscale range and medical applications [11–13]. The comprehensive study by Vimal et al. revealed that the presence of silver nanoparticles into the pure ferroelectric liquid crystal significantly modified the phase transitions, molecular alignment and various other LC parameters [14]. Singh et al. reported that the dispersion of small amount of Ag NPs in the room-temperature NLC increased the nematic–isotropic transition temperature and decreased threshold voltage [15]. Tripathi et al. doped Ag

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NPs in 6CHBT NLC and found that the dielectric anisotropy, threshold voltage and splay elastic constant have decreased for nanocomposites [16]. The effect of Ag NPs, polyaniline and polyaniline/Ag nanocomposites on NLC material was investigated by our group, and we observed that the isotropic temperature has decreased for all the mixtures and ordering of LC molecules can be tuned by doping of polyaniline/Ag nanocomposites [17]. Fast response of supertwisted nematic cells was achieved with the dispersion of Ag/Pd NPs at low temperature [18]. Recently, Shen et al. presented a very nice review indicating the impact of various kinds of nanodopants on the physical properties of LCs, which is attributed to the different size, shape, dispersibility and functionalization of NPs [19].

In the present work, an effort has been made to study the effect of Ag NPs on the dielectric, optical and electro-optical parameters of NLC (8OCB). To understand the influence of Ag NPs on the molecular dynamics of NLC, we have measured dielectric permittivity and dielectric anisotropy. The voltage dependence of rise time and fall time has been examined for both pure and NPs-dispersed NLC. Further, the splay elastic constant and threshold voltage have been calculated for various concentrations of NPs. The investigated Ag NPs/NLC composites are useful for the development of energy-efficient electro-optical devices.

Experimental details

Materials

Positive dielectric anisotropic nematic liquid crystal 8OCB has been used as host material in the present investigation. The phase transition sequence of nematic LC is as follows:

Cryst. $\longrightarrow$ 51°C Nematic $\longrightarrow$ 77°C Iso.

Silver nanoparticles capped with hexanethiol and having the spherical size less than 10 nm have been used as a dopant to disperse into the pure NLC 8OCB.

Preparation of LC sample cell and NPs–LC composites

The LC sample cells were prepared by using two optically active indium tin oxide (ITO)-coated flat glass substrates through the photolithography. The planar alignment has been achieved by the treatment with nylon (6/6) and rubbing unidirectionally. Mylar spacer has been used to maintain a constant thickness of the sample cell. For homeotropic alignment, the glass substrates were coated with a dilute solution of lecithin (cetyltrimethylammonium bromide). The empty sample cells have been calibrated using analytical reagent (AR) grade benzene (C_6H_6) as a standard reference. The details of LC cell fabrication have been already reported by our group [20].

Three composites were prepared by dispersing an appropriate amount of Ag NPs 0.5 wt./wt.% (Mix.1), 1 wt./wt.% (Mix.2) and 2 wt./wt.% (Mix.3) into the pure NLC material. For the preparation of homogeneous mixture of NPs-dispersed nematic liquid crystal, we have first dissolved the NPs in propanol-2 solution and then the suitable amounts of NPs and NLC were taken and homogenized in an ultrasonic mixer for 2 h. The assembled sample cells were filled with pure NLC and NPs-NLC composites by capillary action at above the isotropic temperature of the NLC material. The uniform mixing of NPs in NLC was checked by the polarizing optical microscope (Radical RXLr-5).

Dielectric and electro-optical measurements

The dielectric spectroscopy of pure NLC and NPs-dispersed NLC has been carried out in a frequency range of 100 Hz–40 MHz using a computer-controlled impedance gain/phase analyzer (HP 4194A). The value of dielectric permittivity varies with the frequency of the applied AC field. At very low and high frequencies (< 50 Hz and $> 10^6$ Hz), data obtained from the instruments suffer from some inadequacies. To remove these inadequacies, we fit the data by using correction terms for low- and high-frequency regime. The frequency range from the hundred to kilo hertz suffers minimum inadequacies for nematic liquid crystal, so we choose any frequency in between this range to present our data and in this particular study we have chosen 5 kHz for data representation. The temperature has been maintained by a computer-controlled INSTEC hot plate with an accuracy of ± 0.5 °C.

To study the electro-optical behavior of the pure NLC and Ag NPs-dispersed NLC, an arbitrary square wave of frequency 5 Hz has been applied to the planar aligned cell using a function generator (Tektronix AFG-3021B). A He–Ne laser beam of wavelength 632 nm was used as the input signal, and the output signal was detected by a photodetector (Instec-PD02LI) fed to a digital storage oscilloscope (Tektronix TDS-2024C) in an electrical form. The rise time (τ_{on}) and fall time (τ_{off}) for the pure NLC and Ag NPs-dispersed NLC were calculated by using the following equations.

$$\tau_{on} = t_{90} - t_{10} \text{ and } \tau_{off} = t_{10} - t_{90} \tag{1}$$

τ_{on} is the time required for the transmittance to rise from 10 to 90% and τ_{off} is the time required for the transmittance to fall from 90 to 10% [21].

Results and discussion

The variation in conductivity with the change in frequency in disordered solids, such as polycrystalline semiconductors, polymers, glasses and transition metal oxides, is an important parameter in transport mechanisms. Frequency spectrum of conductivity explains the role of disordering in transport of ions in such systems. In above-mentioned disordered systems, universal law is applied to understand the behavior of composites having foreign particles. It has been observed that at low frequencies, conductivity is constant (illustrated as σ_{dc}), while above a critical frequency (f_o), it becomes strongly frequency dependent and written as σ_{AC}. The Jonscher's universal description employs the following explicit expression for $\sigma(f)$:

$$\sigma(f) = \sigma_{dc} + \sigma_{ac} \text{ Or } \sigma(f) = \sigma_{dc} + kf^n \tag{2}$$

Here, k is constant and the exponent n has the limits $0 < n < 1$ for disordered solids. With the help of impedance spectroscopy of the material, we get to know about the permittivity and dielectric loss as a function of frequency and temperature. By drawing and fitting the Cole–Cole plot, we get the value of DC conductivity (σ_{dc}) which remains constant throughout the frequency spectrum as it does not interfere with the dipoles [22, 23]. However, the AC conductivity (σ_{ac}) varies with the frequency and can be further written as [24]:

$$\sigma_{ac} = 2\pi f \varepsilon_o \varepsilon'' \tag{3}$$

Here, ε_o is permittivity of the free space, ε'' represents the dielectric loss and f is the frequency of the applied field. The AC conductivity has been calculated for the pure and composite system by using above formula. Figure 1 presents
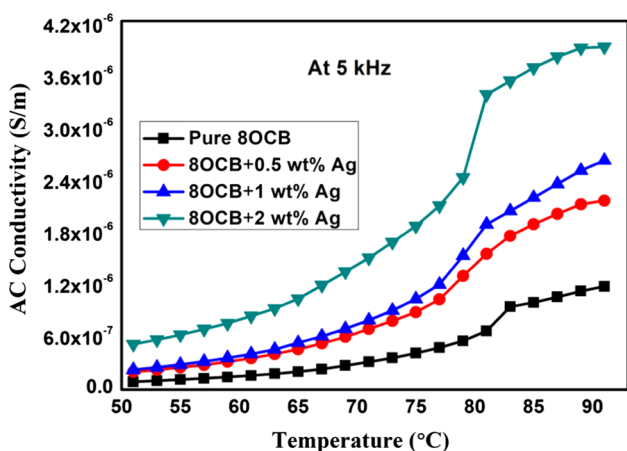
the AC conductivity versus temperature curve for pure NLC 8OCB and Ag NPs-dispersed composite systems.

From the figure, it is observed that the AC conductivity of composite system has been enhanced as compared to pure NLC, which may be due to the presence of metal nanoparticles. It has been suggested by Urbanski et al. [25] that the ligand molecules from the organic capping layer of nanoparticles act as trap for ionic impurities. A partial release of these impurities occurs, while the ligand shell interacts with the uniaxial nematic host material. According to their results, the releasing of these impurities is the most likely source for the increased conductivity of doped system. As the temperature increases, the AC conductivity of pure and composites also increases in the same manner. This is due to the increase in the kinetic energy of the silver nanoparticles.

Figure 2 indicates the photoluminescence (PL) intensity of the pure 8OCB and Ag NPs-dispersed composites. A sharp emission peak has been found at 362 nm, and no shift in emission peak was observed for composites. But the PL intensity of the pure NLC has been quenched after the addition of silver NPs. This quenching in luminescence of the composites shows a dependence on the concentration of Ag NPs. Absorption of major portion of PL emission of the pure NLC material by the Ag NPs might be responsible for reduction in the PL intensity of the composites. When Ag NPs are dispersed in the NLC material, NP–NP dipolar interaction takes place along with NLC–NLC dipolar interaction. The momentum originating from these interactions reduces the charge transfer between Ag NPs and LC molecules, which results in the reduced transfer of photonic energy between the LC molecules and NPs. This can be termed as nonradiative excitation energy transfer from LC molecules to Ag NPs.

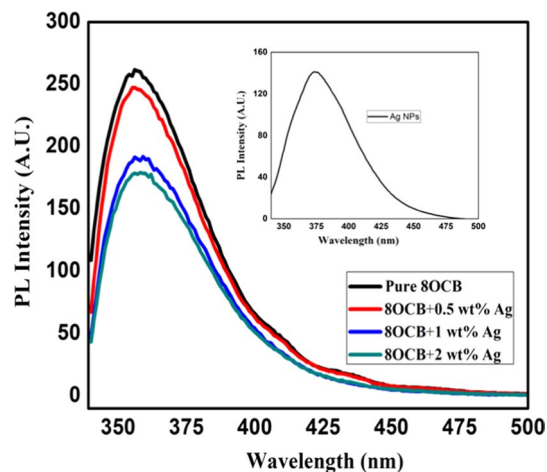Fig. 1 AC conductivity versus temperature graph for pure NLC 8OCB and 8OCB/Ag NPs composites

Fig. 2 Photoluminescence curve for pure NLC 8OCB and 8OCB/Ag NPs composites

Figure 3 represents the variation of parallel and perpendicular component of dielectric permittivity with the temperature. These components have been calculated with the help of method given in Ref. [26]. Since the NLC used in the present study is a positive dielectric anisotropic material, the parallel component of dielectric permittivity (ε_l) will always show higher value as compared to perpendicular component of dielectric permittivity ($\varepsilon_\perp$). As the temperature increases, the difference between parallel and perpendicular components of permittivity becomes narrower. It is interesting to note that the presence of Ag NPs in the pure 8OCB causes a decrement in the parallel and perpendicular component of permittivity. The reduction in the dielectric permittivity of the composite system may be accredited to the strong antiparallel dipolar interaction between the NLC molecules and NPs and change in the orientation of the NLC molecules also. Ag NPs develop antiparallel dipole moment [27–29], and hence resultant dipole moment of the composite system decreases, which results in the decrement in permittivity. On increasing the dopant concentration, the antiparallel dipolar interaction between the NPs and the LC molecules becomes high, which is responsible for affecting the permittivity.

Figure 4 shows the variation in $\varepsilon_\perp$ with respect to the concentration of the dopant. For the similar physical conditions, a LC molecule demonstrates different values of $\varepsilon_\perp$ for different orientations and the minimum value of $\varepsilon_\perp$ is found for perfectly aligned planer sample cell. From the figure, it can be observed that $\varepsilon_\perp$ is decreasing continuously with the increasing concentration of dopant, which suggests that the system becomes more ordered as we increase the concentration of Ag NPs in the host NLC material. The self-assembling nature of NPs improves the molecular alignment of the system as coupling between the NP–LC is higher in comparison with the coupling between the LC–LC molecules.
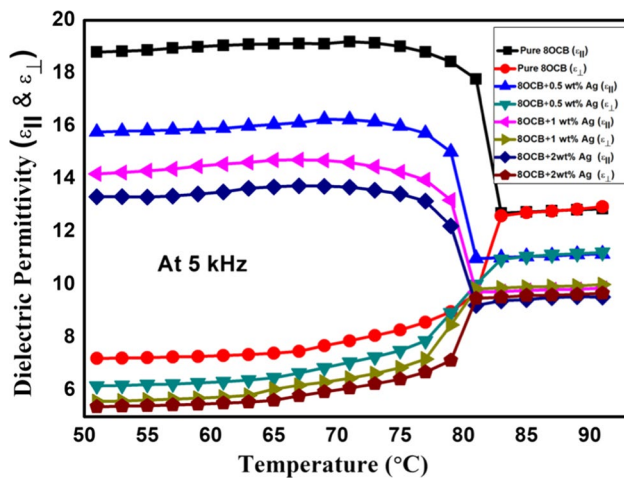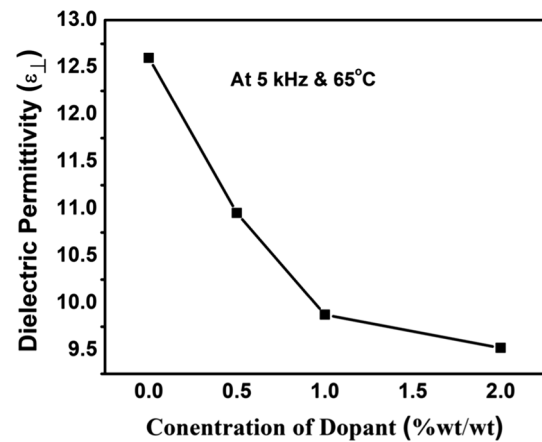

Fig. 4 Variation in dielectric permittivity with different concentrations of dopant

Therefore, these self-assemblies offer a local ordering to the system, which in turn provides the better alignment of molecules in the planar geometry and causes a lower permittivity value.

The variation in dielectric anisotropy with respect to temperature is depicted in Fig. 5. Static dielectric anisotropy ($\Delta\varepsilon$) of the nematic phase of LC material is governed by the Maier and Meier theory [30]:

$$\Delta\varepsilon = \frac{NHF}{\varepsilon_0}\left[\Delta\alpha - \frac{F}{2kT}\mu^2\left(1 - 3\cos^2\beta\right)\right]S \qquad (4)$$

where N is the number density of molecules (the number of molecules per unit volume), H and F are internal field factors, $\Delta\alpha$ is the anisotropy of the polarizability, kT is the thermal energy, μ is the resultant dipole moment of the
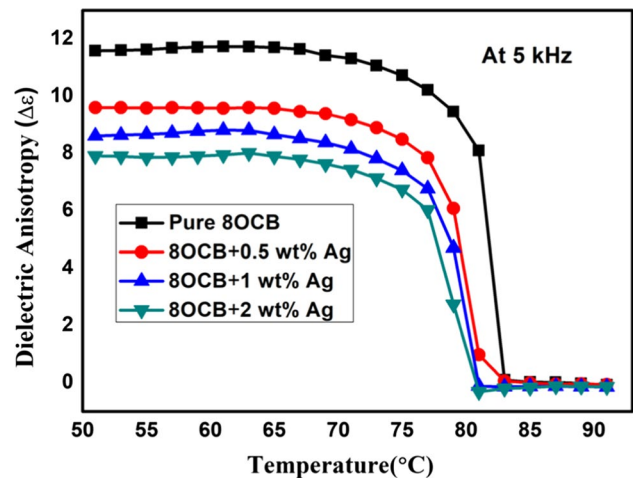

Fig. 3 Dielectric permittivity versus temperature graph for pure NLC 8OCB and 8OCB/Ag NPs composites

Fig. 5 Dielectric anisotropy versus temperature curve for pure NLC 8OCB and 8OCB/Ag NPs composites

molecules, β is the angle between the dipole moment and long molecular axis of the molecule and S is rotational order parameter. According to the theory, the value of $\Delta\varepsilon$ is mainly determined by the N, $\Delta\alpha$, μ and S. N is expected to decrease for the doped system, due to which $\Delta\varepsilon$ must decrease. The dielectric anisotropy depends on the interaction strength of the LC with an applied electric field and is therefore an important parameter which can influence the threshold voltage. Each composite of 8OCB NLC dispersed with Ag NPs exhibits the decrement in the dielectric anisotropy. The more the concentration of Ag NPs in the pure 8OCB, the more the decrement in the observed dielectric anisotropy. The reason for this decrement is various factors; (1) spherical shape of the nanoparticles, (2) the net dipole moment of the system decreases after doping of the Ag NPs in the 8OCB. The Ag NPs dipole correlation is not in favor of the dipole moments of the pure 8OCB. Due to opposite or inclined orientation of dipole moments, the net dipole moments of Ag NPs are not get aligned parallel to the dipole moments of 8OCB molecules. This decreased dipole moment may cause the decrement in the dielectric anisotropy of Ag NPs-dispersed 8OCB sample. Therefore, according to Maier and Meier theory, if dipole moment and order parameter decrease, then, consequently, the dielectric anisotropy also decreases.

The electro-optical response of LC molecules under the influence of electric field is the major characteristics used in industrial applications. To estimate the performance of the liquid crystal dispersions for display device application, we have measured response time of the sample. The electro-optical switching of 8OCB molecules in the nematic phase is recorded as a function of voltage at 5 Hz frequency of applied field. The voltage variation of rise time and fall time of the pure 8OCB and Ag NPs-dispersed samples is shown in Fig. 6a, b. The rise time and fall time decrease on increasing the applied electric field. It is also noticed that the rise time and fall time increase on increasing the dopant concentration. This increment in the rise and fall time is due to the hindrances produced by the spherical Ag NPs. When we increase the dopant concentration in the pure 8OCB NLC, it means more Ag NPs are added into the NLC material. Consequently, more hindrances are produced by Ag NPs on the movement of 8OCB molecules. Therefore, the rise time and fall time (response time) increase considerably of the composite system. The response time is related to the elastic constant of the samples. The strong π–π electron interaction between 8OCB molecules and Ag NPs might be responsible for the increase in the rise time.

The total response time can be given as

$$\tau_{total} = \tau_{rise} + \tau_{fall} \tag{5}$$

The threshold voltage (V_{th}) is an imperative parameter of the NLCs. Figure 7 represents the variation in V_{th} with
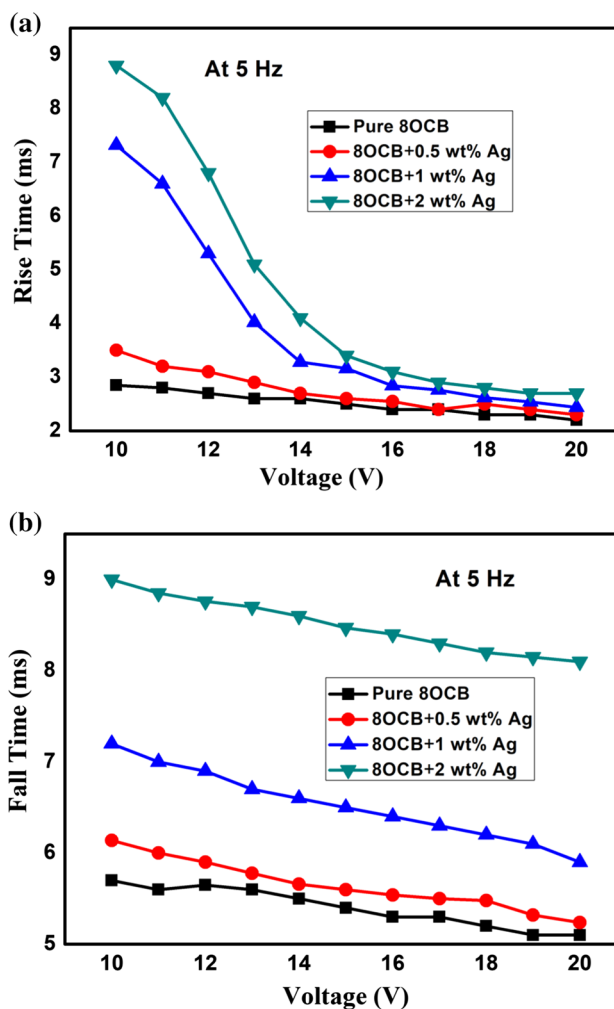
(a)

(b)

Fig. 6 **a** Rise time versus voltage curve for pure NLC and its nanocomposites. **b** Fall time versus voltage curve for pure NLC and its nanocomposites
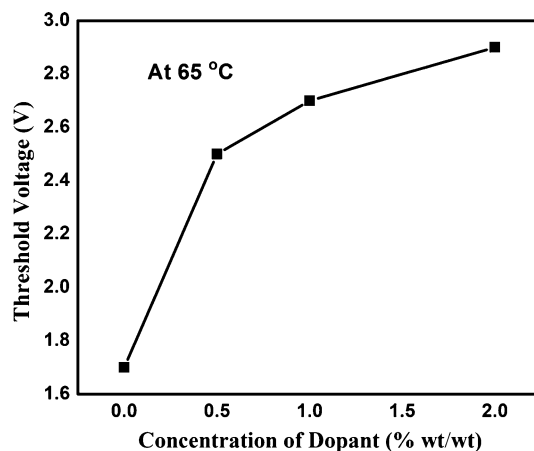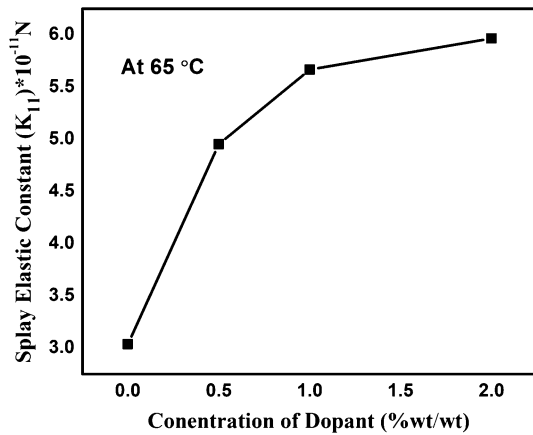
Fig. 7 Variation in threshold voltage with different concentrations of dopant

Fig. 8 Variation in splay elastic constant with different concentrations of dopant

different concentrations of dopant in the pure NLC material 8OCB. These voltages were determined by measuring the intensity of transmitted light of sample cells as a function of applied voltage. With the application of step change AC voltage and then detecting the change in transmitted light intensity, we have measured the threshold voltage (the voltage under which the transmitted intensity is changed by the 10%). Freedricksz's threshold voltage (V_{th}) is related to the splay elastic constant (K_{11}) and dielectric anisotropy ($\Delta\varepsilon$) by the following relation [21]:

$$V_{th} = \left[\frac{k_{11}\pi^2}{\varepsilon_0|\Delta\varepsilon|}\right]^{1/2} \tag{6}$$

The above relation clearly indicates that the threshold voltage strongly depends upon the splay elastic constant and dielectric anisotropy. From Fig. 7, we can see that the threshold voltage of composites is higher than that of pure NLC. The inverse relation between the threshold voltage and dielectric anisotropy explains the increment in the value of threshold voltage. Since the dielectric anisotropy has been decreased after the addition of Ag NPs to pure NLC, an increment in value of threshold voltage has been observed for the composite systems.

The variation in splay elastic constant as a function of dopant concentrations is depicted in Fig. 8. The splay elastic constant is an important parameter which can govern the switching of nematic LC in display devices. K_{11} is determined from the threshold voltage and dielectric anisotropy. From the figure, we can see that the value of K_{11} increases on increasing the concentration of Ag NPs in the pure NLC matrix. The splay elastic constant basically depends upon the threshold voltage and also depends upon the number and size of the molecule. So, the enhancement

in the value of K_{11} for composites may be attributed to the increment in the threshold voltage of composites.

Conclusions

In brevity, we had demonstrated the immense effects of silver nanoparticles on the dielectric, electro-optical and luminescence properties of host NLC material. The AC conductivity of all the nanocomposites has increased due to the presence of metal nanoparticles. The dielectric permittivity of the composite system is reduced, which may be attributed to the strong antiparallel dipolar interaction between the NLC molecules and NPs. The photoluminescence intensity of the pure NLC is quenched after the addition of Ag NPs into it due to absorption of major portion of PL emission by the Ag NPs. These properties of the composites make them suitable toward the development of novel materials for different applications.

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