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Improved dielectric and electro-optical parameters of nematic liquid crystal doped with magnetic nanoparticles

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Improved dielectric and electro-optical parameters of nematic liquid crystal doped with magnetic nanoparticles

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This study investigates the effect of magnetic nanoparticles (NPs) on the weakly polar nematic liquid crystal (NLC). Different parameters of dielectric data were measured for both the homeotropic and planar aligned samples as a function of frequency and temperature and the substantial changes have been noticed for the doped systems. Dielectric permittivity has been increased after the dispersion of magnetic NPs in the pure NLC. Dielectric anisotropy has also been influenced by incorporating the magnetic NPs with the NLC molecules. These results were attributed to the dipole–dipole interaction between the magnetic nanoparticles and nematic liquid crystal molecules. Electro-optical study indicated the faster rise time and fall time of the doped systems as compare to pure NLC. Threshold voltage has been calculated and found to be decreased for the doped systems. Moreover, we have also calculated the rotational viscosity and the splay elastic constant for pure and the doped systems. Both the rotational viscosity and splay elastic constant of the doped systems are found to be considerably lower than those of pure NLC. Change in these properties has been explained on the basis of molecular disturbances created by the interaction between the magnetic nanoparticle and LC director. This study reveals that the inclusion of magnetic NPs in weakly polar NLC can be useful to enhance the basic properties of the weakly polar NLC and make it a promising material for many display applications.

Keywords: magnetic nanoparticles, dielectric permittivity, response time, rotational viscosity

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1. Introduction

The investigation on composite liquid crystals (LCs) has been attracting significant attention in the past few decades because of their interesting physical properties. Doping LCs with different nanomaterials like carbon nanotubes (CNTs), dyes, quantum dots (QDs), polymers, etc[1–4] makes it possible to obtain new materials with unique properties. Immense work has been done by researchers to improve the physical properties of composite systems, so that the high performance quality devices with faster switching and low energy consumption can be prepared. Permittivity is a significant parameter of LCs which controls the electro-optical molecular response, anisotropy, and dynamics of the liquid crystal medium.[5] Permittivity is described as an ability of a material to be polarized when exposed to an external electric field. LC possesses anisotropy of physical properties that allows changing the orientation of the LC molecular axis under the influence of an electric field.[6] Dielectric anisotropy is responsible for image quality of liquid crystal displays so it becomes an important parameter for display application. Due to small anisotropy of weakly polar LC, the threshold electric field required to change its orientation is large. Many reports have been presented to study these weakly polar nematic LCs which explain the dielectric properties with the help of dynamics of molecule.[7,8] Oka *et al*. have done the comparative study of weakly polar and non-polar NLCs. They suggested that the strength of dipole moment has little influence on the liquid crystalline properties.[9] Lobo *et al*. performed differential scanning calorimetric and dielectric studies of a composite system of LC that does not possess terminal polar group and found systematic increase in the relaxation frequency.[10]

Suspension of iron oxide (Fe_2O_3) magnetic nanoparticle (NP) in NLC is being a topic of fundamental research interest. Many properties of NLC that are favorable for practical applications can be enhanced by doping ferromagnetic nanoparticles.[11] Fe_2O_3 NPs are low cost, non-toxic materials which have relatively good stability. These nanoparticles affect significantly the dielectric and electro-optical properties of NLCs.[12,13] Fe_2O_3 doped NLC exhibits enhanced dielectric constant, decreased threshold voltage, and shorter response time than the pure NLC.[13] Effect of magnetic NPs in the properties of LC depends upon the shape, size, concentration, and physical properties of the NPs.[14–16] The colloidal suspension of magnetic particles in nematic LCs, known as ferronematic was first theoretically given by Brochard and De Gennes.[17] They pointed out that permanent magnetic moment of magnetic nanoparticle would be coupled with the dielectric movement of director of LC, for more details, read Refs. [18], [19], and [22]. The magnetic nanoparticle dopants induce magnetic capability in LC.[5] There are few other reports which emphasize on physical measurements of LC composites based on ferronematic nanoparticles. Gorkunov and

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Osipov[19] developed a mean field theory to describe the influence of embedded nanoparticles on the orientational order and isotropic-nematic phase transition temperature of host LC. The basic property of a suspension of rod like γ-Fe_2O_3 particle in the LC MBBA has been reported by Rault *et al.*[20] The discovery of unambiguous ferronematic LC by Mertelj *et al.*[19] has created immense interest in the scientific community. To prevent nanoparticles from agglomeration and sedimentation, nanoparticles are covered with amphiphilic surfactant or ionic polymers.[22] The influence of electric field on the ferronematic depends on the coupling energy and mutual orientation of magnetic nanoparticles and LC molecules. Coupling is the result of interaction of nematic LC with the surface of nanoparticles and due to such coupling, the magnetic moment of magnetic nanoparticle responses to the electric field indirectly.[23] The image sticking behavior of displays could also be improved by doping γ-Fe_2O_3 nanoparticles into the LC materials because of their magnetization induced by the electric field.[24]

In the present paper, an effort has been made to study the dielectric and electro-optical properties of the magnetic NPs doped weakly polar nematic LC. We report the measurement of dielectric permittivity and dielectric anisotropy for understanding the influence of magnetic NPs on molecular dynamics of LCs. Decreased rise time and fall time for the doped system are explained. The threshold voltage for the composite system is decreasing with increasing concentration of NPs. Further the temperature dependences of splay elastic constant and viscosity have been investigated for various concentrations of NPs. For interpreting these results, the mutual influence of spherical magnetic NPs and NLC molecule has been considered.

2. Experimental details

2.1. Materials

Magnetic nanoparticle doped nematic LC composite used in this investigation consists of host NLC 4, 4′-dipentylazoxybenzine (D5AOB). The phase transition sequence of D5AOB is given as Cr $\xrightarrow{22\ ^\circ C}$ N $\xrightarrow{65\ ^\circ C}$ Iso. The chemical structure of the host sample is shown in Fig. 1.

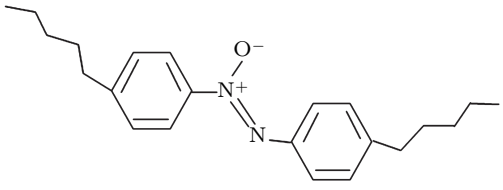


Fig. 1. Chemical structure of D5AOB.

The host sample was purchased from Flintron Laboratory USA. The magnetic spherical nanoparticle used in this study is iron oxide (Fe_2O_3). Figure 2 presents the x-ray diffraction (XRD) pattern of Fe_2O_3 NPs. The crystalline sizes estimated by XRD were found to lie between 10 nm and 20 nm. These NPs were stirred in an ethanol solution for eight hours and then a particular volume was mixed with the particular weight of nematic LC (D5AOB) to prepare four composites of 0.1% wt./wt. (mix1), 0.25% wt./wt. (mix2), 0.5% wt./wt. (mix3), and 0.75% wt./wt. (mix4) of nanoparticles. Each mixture was heated from nematic to isotropic and cooled back to nematic at room temperature. This heating–cooling cycle was repeated till the homogeneous distribution of nanoparticle was not achieved.

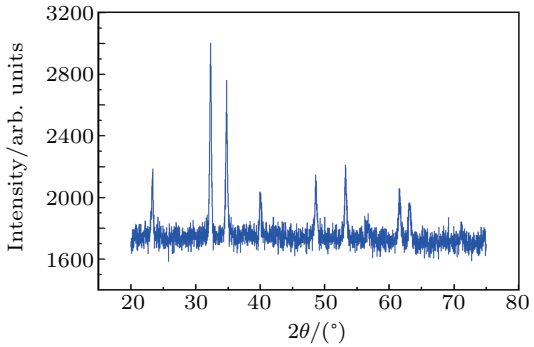


Fig. 2. XRD spectra of Fe_2O_3 nanopaticles.

2.2. Preparation of LC cell

Sandwich type of planner as well as homeotropic sample cells have been used in the present study. These cells were made by using two indium tin oxide (ITO) coated glass substrates. The planner and the homeotropic alignments have been achieved by treating conducting layers with adhesive promoters and coated with polymer nylon (6/6) for planner alignment and lecithin for homeotropic alignment. After drying polymer or lecithin, two substrates were rubbed unidirectional by cotton. The substrates were then placed one over another. The cell thickness was fixed by placing a mylar spacer (2.5 μm) between two substrates and then it was sealed by a UV sealant. The empty sample cells were calibrated by using analytical reagent grade benzene as a standard reference for dielectric studies. After this the assembled cells were filled with the pure and two composites at temperature higher than 65 °C by the capillary method. The detail description of cell fabrication has already described in our previous paper.[25]

2.3. Dielectric measurement

Dielectric measurements were performed by using a computer control impedance gain-phase analyzer (HP-4194 A) in the frequency range from 100 Hz to 40 MHz. Instec hot plate (mK 2000) was used to achieve the desired temperature with an accuracy of ±0.001 °C. The dielectric measurements were carried out as a function of temperature by placing the sample

inside the hot plate. The detail about the dielectric measurement for the liquid crystal has already been reported by us.[26]

2.4. Electro-optical measurement

Optical switching method has been used to measure the response time of nematic liquid crystal.[27] In this method, a square wave input signal (10 V and 1 Hz) was applied to the cell by using a function generator. The input signal of He–Ne laser beam with a wavelength 632 nm was detected by a photo detector (Instec-PD02LI) connected directly to the digital storage oscilloscope (TektronixTDS-2024 C). From the detected shape of wave form, we calculated the rise time (τ_{on}) and fall time (τ_{off}) for pure LC and nano particles doped LC. 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%. The cell was set at an angle of 45° between the crossed polarizer and analyzer for measuring the maximum optical transmittance. Thus, the cell worked as a phase retarder there by altering the polarization of light.

3. Results and discussion

Figure 3 presents the textures of pure NLC and two-composite systems under crossed polarized conditions taken by a polarizing optical microscope. From the figure, it can be seen that the magnetic NPs have been dissolved properly in mix1 and mix2 while some aggregations of magnetic NPs in mix3 and mix4 can be noticed.

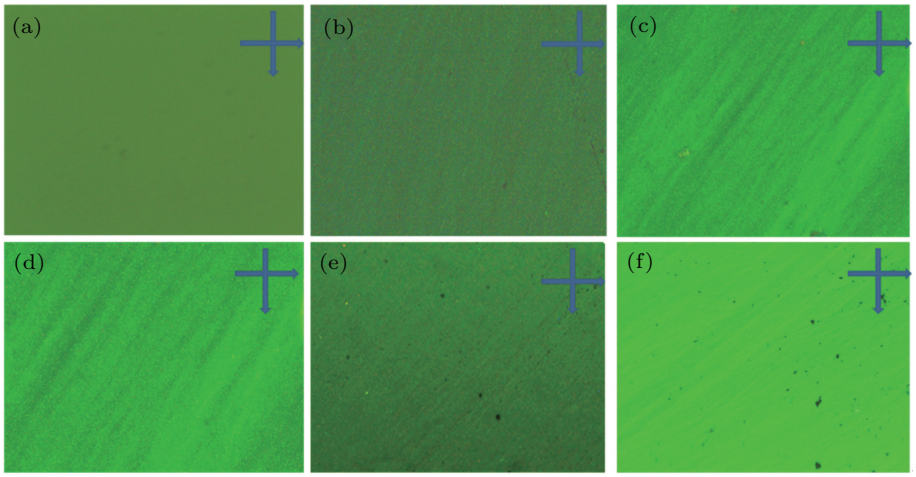


Fig. 3. Textures of (a) unaligned, (b) pure, (c) mix1, (d) mix2, (e) mix3, and (f) mix4 samples.

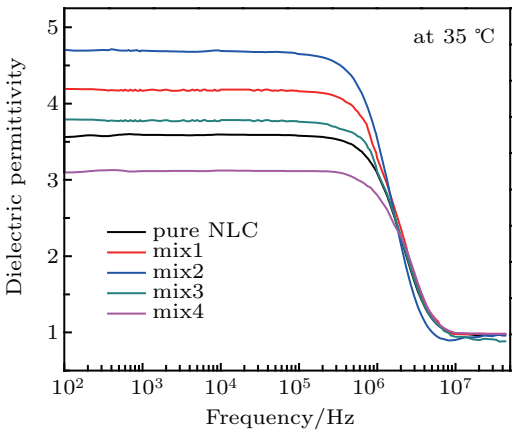


Fig. 4. Variation of dielectric permittivity with frequency for pure and composite systems.

First we discuss the influence of magnetic nanoparticle on the dielectric permittivity of pure and doped systems. Figure 4 presents the variation of parallel component of dielectric permittivity with frequency from 100 Hz to 40 MHz at 35 °C. It can be seen that the permittivity in the nematic phase is almost constant in the frequency range from 1 kHz to 1 MHz and then decreases with frequency, and the variation of permittivity for the composite systems follows the same trend as that for the pure one. Analysis of this plot confirms that with the addition of nanoparticles, the permittivity first increases for lower two concentrations and then decreases for higher concentration. This decreased value of permittivity is more than the pure one for mix4. This effect can be attributed to the mutual influence of magnetic NPs and NLC molecule. Magnetic coupling and elastic coupling are two sources of interactions between NPs and surrounding NLC.[17]

Magnetic particle consisting of magnetic dipole moment will cause a strong dipole–dipole interaction between nanoparticles and surrounding LC molecules.[18,19,22,28] At lower concentration, since the numbers of magnetic nanoparticles are small so the influence of the one nanoparticle on the other one can be negligible. In this situation, the surrounding LC molecules along with the nanoparticle will orient along the long molecular axis so that the net dipole moment of the nano-nematic LC composite is found to be larger than that of the pure NLC and hence results in an increase of dielectric permittivity. For mix2, the numbers of nanoparticles are greater than those of Mix1 so it has more aligned LC molecules and

hence the dielectric permittivity. The dipole moments of the nanoparticle and LC molecule may be in the same or opposite direction. It depends on the type of molecular interaction taking place within the system. At higher concentration, the surface of magnetic nanoparticle plays a crucial role in disturbing the surrounded LC molecular ordering by forming a magnetic coupling with the adjacent magnetic nanoparticles. Because of such perturbation, the net dipole moment of the LC molecules is decreased, hence resulting in the overall decrease of the permittivity. The surfactant used for preventing the agglomeration and segregation also plays a crucial role in disturbing nematic LC molecules by generating elastic forces due to the hyperbolic hedgehog effect.[29,30]

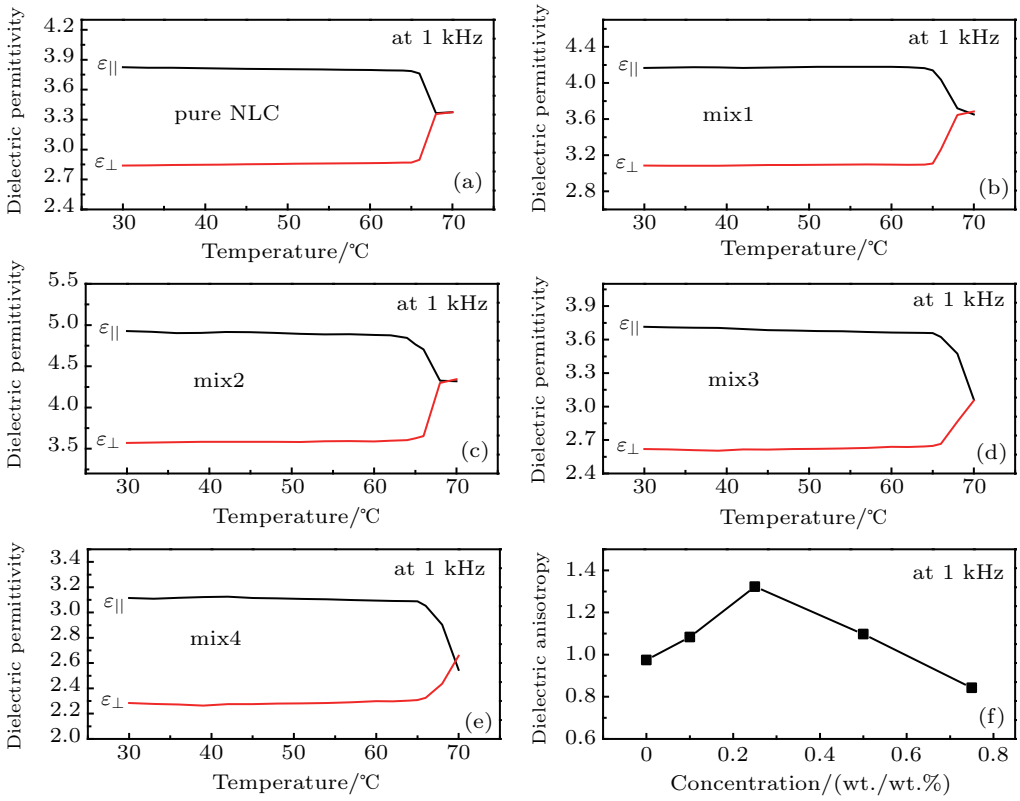


Fig. 5. Variation of dielectric permittivity with temperature: (a) pure NLC, (b) mix1, (c) mix2, (d) mix3, (e) mix4. Panel (f) is the variation of dielectric anisotropy with concentration of dopant (at 35 °C).

Figure 5(a) shows the temperature dependence of dielectric permittivity for pure and different concentration doped NLCs at the frequency of 1 kHz. For each concentration, there are two diagrams, the upper one belongs to the parallel component of dielectric permittivity and the lower one for the perpendicular component of dielectric permittivity. It can be observed that by increasing the temperature, the parallel component of dielectric permittivity is decreasing and conversely the perpendicular component of dielectric permittivity is increasing. Considerable increase in dielectric permittivity at both the perpendicular and parallel components for mix1 and mix2 has been noticed while mix3 and mix4 show decrease in dielectric permittivity in both types of components. Dielectric anisotropy is described as the difference between $\varepsilon_{||}$ and $\varepsilon_{\perp}$ ($\Delta\varepsilon = \varepsilon_{||} - \varepsilon_{\perp}$) . The components $\varepsilon_{||}$ and $\varepsilon_{\perp}$ represent the dielectric permittivity for the applied electric field in parallel and perpendicular directions of molecular orientation, respectively. The NLC used in this study is a positive dielectric anisotropic material (i.e., $\Delta\varepsilon > 0$). Figure 5(d) shows the variation of dielectric anisotropy with the concentration of NPs. It can be seen from the figure that dielectric anisotropy ($\Delta\varepsilon$) has been increased for mix1 and mix2, and for mix3 and mix4 the anisotropy has been decreased. The value of $\Delta\varepsilon$ for mix4 is found to be lower than that of pure NLC. The dielectric anisotropy is proportional to the rotational order parameter S of the LC. According to the Maier and Meier theory,[31] the dielectric anisotropy is given by

$$\Delta\varepsilon = \varepsilon_{||} - \varepsilon_{\perp} = \frac{NhF}{\varepsilon_0}\left[\Delta\alpha S - \frac{F\mu^2}{2KT}(1 - 3\cos^2\beta)S\right], \quad (1)$$

where ε_0 is the permittivity of free space, N is the number density, h and F are the internal field factors, $\Delta\alpha$ is the anisotropy of polarizability, KT is the thermal energy, β is the angle between the molecular net permanent dipole moment and the long molecular axis of the molecule, and S is the rotational order parameter.

It means that at lower concentration, the magnetic NPs can improve the order parameter of NLC molecule. The dielectric anisotropy of the LC also depends on the molecular polarizability and the change of the effective dipole moment

of the LC molecules. The doped nanoparticles will cause new orientation of resultant dipole moment of LC molecule with respect to long molecular axis. There is some additional permanent dipole moment in D5AOB, which contributes slightly more in the perpendicular direction than in the parallel direction. The variation in dielectric anisotropy with concentration is the resultant of the net contribution of dipole moment and polarizability towards $\varepsilon_{||}$ and $\varepsilon_{\perp}$ permittivity. At higher concentration, the decreased dielectric anisotropy has been observed. The dielectric anisotropy also depends upon angle β, $\beta = \tan^{-1}(\mu_t/\mu_l)$, where μ_t and μ_l are the transverse and longitudinal components of dipole moment of NLC molecule. The decreased value of $\Delta\varepsilon$ may be due to decrease in the parallel component of dipole moment more than the perpendicular component. The values of $\varepsilon_{||}$ and $\varepsilon_{\perp}$ of D5AOB having weakly polar terminal groups are much smaller than those having strong polar terminal group.

To study the electro-optical properties of pure and composite systems, we have determined the rotational viscosity and splay elastic constant. The splay elastic constant is obtained from Fredrick's threshold voltage, i.e., $V_{th} = \sqrt{\frac{K_{11}\pi^2}{\Delta\varepsilon\varepsilon_0}}$, where V_{th} is the threshold voltage calculated from electro-optical study, K_{11} is the splay elastic constant, and ε_0 is the permittivity of free space. This relation clearly indicates that the threshold voltage strongly depends upon the splay elastic constant and dielectric anisotropy. The threshold voltage is an important parameter of NLCs. Figure 6 shows the variation of threshold voltage with concentration of magnetic nanoparticles in the pure NLC. The threshold voltages are 4.3 V, 3.5 V, 2.8 V, 3.6 V, and 3.8 V for pure, mix1, mix2, mix3, and mix4 systems, respectively. It is following the same trend as the dielectric anisotropy so the change in threshold voltage can be attributed to the change of dielectric anisotropy. Reduced voltage causes the reduced electrical consumption, thus making the NLC a potential material for display applications.

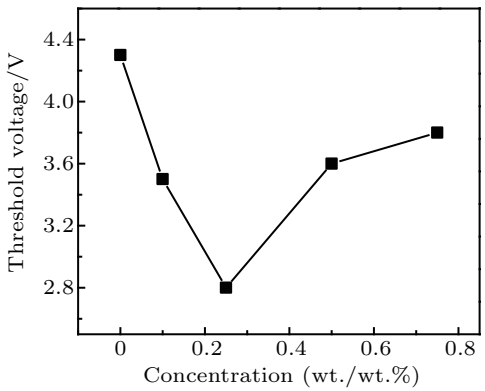


Fig. 6. Variation of threshold voltage with concentration for pure and composite systems.

The splay elastic constant and rotational viscosity of nematic LC are given by

$$K_{11} = \left(\frac{V_{th}}{\pi}\right)^2 \Delta\varepsilon\varepsilon_0, \tag{2}$$

$$\gamma = \frac{\tau_0 K_{11}\pi^2}{d^2}, \tag{3}$$

where d is the cell thickness and τ_0 is the relaxation time.

Figures 7 and 8 show the temperature dependence of splay elastic constant and rotational viscosity for pure and nanoparticle doped composite systems. It is observed that K_{11} and γ both have been decreased for the composite systems. According to the mean field theory,[32] both K_{11} and γ are directly proportional to S^2. The decrease of S in the composite system is very small. Hence the decreases of K_{11} and γ cannot be due to the decrease of S alone, as both decreases more than the expected decrease of S. The splay elastic constant basically depends upon the threshold voltage and dielectric anisotropy. So this large amount of decrease in K_{11} may be attributed to the decrease of V_{th}. The splay elastic constant also depends upon the number and size of the molecule. Rotational viscosity of aligned LC represents an internal friction among liquid crystal directors during the rotation process. The magnitude of rotational viscosity depends on the molecular structure of constituents, intermolecular association, temperature, and the presence of ions. The decrease in rotational viscosity in our case may be due to the decrease of K_{11} and intermolecular association between the nanoparticles. The response time of the LC is linearly proportional to the rotational viscosity so it is an important parameter for many electro-optical devices. Faster switching of nematic LCs is an important factor to improve the quality of LCDs.

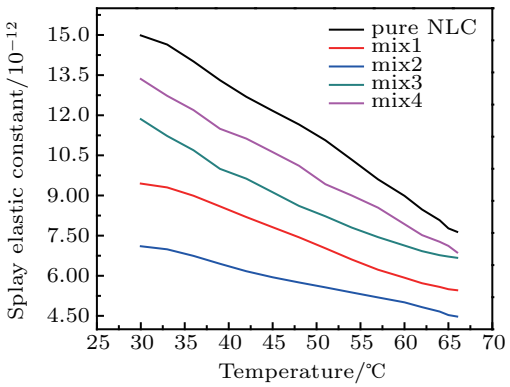


Fig. 7. Variation of splay elastic constant with temperature for pure and composite systems.

The above study clearly suggests that the response time should reduce for the doped system. Figure 9 and 10 show the variation of rise time and fall time with temperature, respectively. It is observed that both are decreasing as the temperature is increasing. Rise time and fall time depend upon the viscosity and the order parameter. Viscosity and order parameter always decrease with temperature. The slowest response

is observed for the undoped system which exhibits rise time of 8.1 ms and fall time of 81.3 ms at room temperature. The faster response time is observed for mix2, having doping concentration of 0.25%, in this case the nematic LC exhibits rise time of 4.2 ms and fall time of 74.5 ms at room temperature. It can be concluded that at particular concentration (0.25%) of magnetic nanoparticles, the faster switching can be achieved.

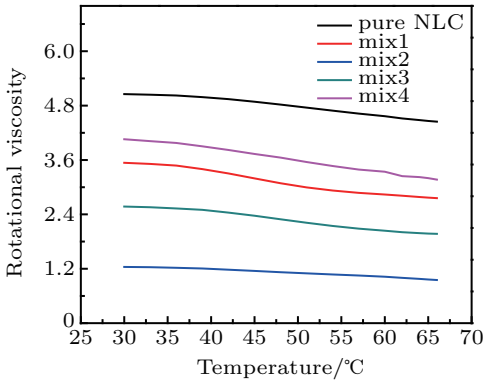


Fig. 8. Variation of rotational viscosity with temperature for pure and composite systems.

The response time from off to on state, τ_{on} (rise time), and that from on to off, τ_{off} (fall time), are defined as

$$\tau_{on} = \frac{\gamma d^2}{\varepsilon_0 \Delta\varepsilon V^2 - K_{11}\pi^2}, \tag{4}$$

$$\tau_{off} = \frac{\gamma d^2}{K_{11}\pi^2}. \tag{5}$$

From Eq. (4), it is clear that τ_{on} depends on viscosity, dielectric anisotropy, and threshold voltage. Dielectric anisotropy is showing very small variation, so τ_{on} will depend on viscosity and field. Viscosity is decreasing with increasing temperature and concentration as well. Thus the behavior of fall time shown in Fig. 10 is an outcome of the above mentioned two events. From Eq. (5), it can be seen that the fall time is proportional to γ and K_{11}. Since both γ and K_{11} are decreasing with temperature, it means the visco-elastic (γ/K_{11}) term is not much affecting the fall time, thereby resulting in very mild decrease in fall time. But there is a significant change in fall time. This change might be due to the suppression of screening effect. There exists an interaction between impurity ions and Fe_2O_3 nanoparticle.[13] Under an applied voltage, the impurity ions near the alignment layer get trapped by NPs. This trapping mechanism abates the screening effect by triggering few impurity ions to flow to the two sides of the substrate when the voltage is on. In off state, the attraction between the alignment layer and LC molecule increases and results in fast response of fall time. The slow response of rise time and fall time of mix3 and mix4 as compare to mix1 and mix2 may be due to aggregation, generally observed at higher concentrations, which causes a hindrance in the movement of NLC molecules.

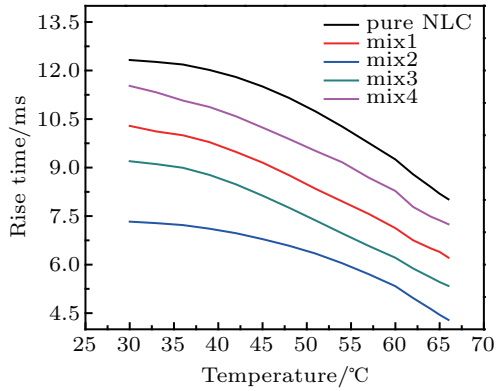


Fig. 9. Variation of rise time with temperature for pure and composite systems.

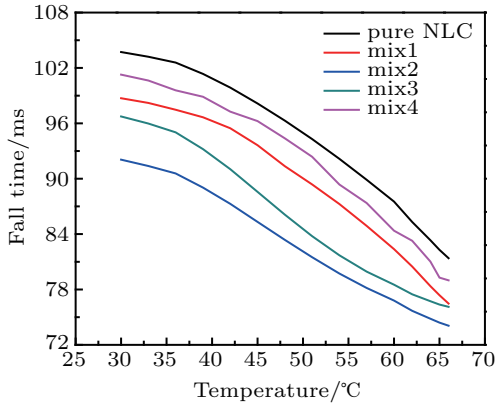


Fig. 10. Variation of fall time with temperature for pure and composite systems.

4. Conclusion

In the present paper, we have studied the dielectric and electro-optical properties of weakly polar nematic NLC doped with magnetic nanoparticles. The dielectric permittivity, dielectric anisotropy, threshold voltage, rotational viscosity, splay elastic constant, and response time have been measured for pure and doped systems. Dielectric study shows the considerable increase in dielectric permittivity for lower concentration and decrease for higher concentration, which may be attributed to the coupling between the magnetic moment of magnetic nanoparticles and the director of nematic liquid crystal. Dielectric anisotropy shows reasonable variation in all concentrations, which has been explained with the help of Maier and Meier theory. In electro-optical study, we have found decreased rise time and fall time for both composite systems. Decreased rise time and fall time increases its applicability in display devices by improving its switching behavior. The threshold voltage has been decreased, which is good from application point of view. Reduction of threshold voltage will enable reduced electrical energy consumption. The splay elastic constant and the viscosity have also been decreased with increasing both the concentration and the temperature. From overall results, we can conclude that, at particular concentration (i.e., 25%) of magnetic nanoparticle, the highly improved

characteristic features of NLC material can be achieved.

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