

# Investigation of Increased UV Absorbance, High Quenching Luminescence Efficiency of Quantum Dot Dispersed Nematic Mesogen and Its Relevance in Photonic Applications

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**Abstract:** The dispersion of Cd<sub>1-x</sub>Zn<sub>x</sub>S/ZnS core/shell QD (x=0.85) in nematic liquid crystal into different concentrations has been examined. The spectroscopic parameters such as UV visible absorbance, Photoluminescence (PL) and Fourier Transform Infrared (FTIR) spectra have been observed for pristine and QD dispersed nematic system. The UV visible absorbance spectra brings to light one absorbance peak corresponding to  $\pi$ - $\pi^*$  transition. Increased UV absorbance with decreased band gap for dispersed system reveals the strong photo absorption (i.e. quenched luminescence). In the present study, the blue shift in Photoluminescence has been observed. The remarkable results have been found for 0.1% and 0.25% wt/wt QD dispersed nematic system with an average of 51.7% and 61.7% quenching in PL intensity. The existing theory provides support for our observation of quenching in PL intensity. Full width half maxima (FWHM) obtained from the Gaussian fit have been the support for the quenching in PL. The FTIR spectra explain the symmetric and asymmetric C-H stretching vibrations which confirm the quenched PL intensity. The UV visible absorbance, PL and FTIR studies have been performed in order to disclose the nature of QD dispersed nematic system. The upshot of the present study finds its relevance in the field of various display and photonic applications as well as in the photovoltaic devices.

**Index Term:** Nematic liquid crystal; Quantum dot; UV visible spectroscopy; Photoluminescence; FTIR.

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## I. INTRODUCTION

The unremitting demand for the nanomaterials-liquid crystal composite system and the improvement in the performance of LC based devices has been extensively required by the technological field [1, 2]. The dispersion of several nanomaterials into liquid crystals such as Ferroelectric nanoparticles (NPs) [3], metallic NPs [4] etc has an influential consequence that removes the obstacles which hampered the performance of display devices. The significant alteration in the physical and material parameters has been found with the addition of nanomaterials in liquid crystals. Liquid crystals and its composites have wide range of potential application in opto electronic, photonic and display devices [2, 5-8]. They have immense technological importance in both fundamental and applied aspects [2, 5-9]. Quantum dots have been utilized as dopant in liquid crystals due to its size dependent fluorescence and electrical properties [1]. Semiconductors QD are the smart nanomaterials that attracted much attention of consortium of researchers due to their prospective applications in display and electro optical devices [2, 9]. These nanomaterials are termed as “smart nanomaterials” due to existence of phenomenon of charge transfer, photo or electro-

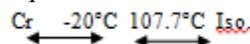
luminescence and self assembly property therefore they have extensive range of application in fabrication of various electro optic and display devices [10-14]. Dispersing nematic liquid crystal with semiconductor QDs changes its dielectric and viscoelastic properties which affect the characteristic of dynamic devices based on these systems [15]. Nanoparticles dispersed nematic systems are the promising candidates for controlling the Photoluminescence (PL) intensity in photonic devices [16]. Change in the luminescence as a result of interaction between donor-acceptor brings into play its application in sensor and biomedical field. Charge separation between the different electron affinities components as a result of excitation of charge donor is widely useful in photovoltaic devices and laser radiation limiters [6, 8, 16]. Various research articles throw light on phenomenon of energy/charge transfer to and from QD. The possibilities for the occurrence of such phenomenon which affect the transfer dynamics have been discussed in it [10-12]. The non radiative energy transfer in molecular system has been described by quantum mechanical theory developed by Forster [11]. It describes the theoretical consideration of energy transfer between QDs. The consideration in dispersion of nanoparticles in LC system is related to the

panorama of controlling their optical and dynamic properties using electric and magnetic fields. The studies on PL spectra of QD dispersed nematic LC system are rare. M.A. Kurochkina and et.al had reported specific features of luminescence quenching in nanoparticles dispersed nematic system [16, 17]. Various possible reasons for the quenching in PL intensity had been described in it. Bae and et.al had elaborated the synthesization process of highly luminescent blue emitting Cd<sub>1-x</sub>Zn<sub>x</sub>S/ZnS core/shell QD [18]. S. Sahai and et.al had discussed the facile synthesis and step by step enhancement of blue Photoluminescence from Ag-doped ZnS quantum dots [19]. Tong et al. have reported an emission based LC display by involving QDs in a self assembled LC gel [20]. The realization of emissive LCDs has been renowned as a matter of fact that QDs introduces enhancement in the luminescence of LC materials. Kumar et al. have observed the remarkable enhancement in PL intensity and shift in the emission band of QD dispersed ferroelectric LC system [21]. D.P. Singh et al had reported the time resolved fluorescence and absence of forster resonance energy transfer in ferroelectric liquid crystal-quantum dots composites [12].

In the present paper PL, UV-Visible and band gap evaluations have been performed in order to disclose the nature of QD dispersed nematic system. Cd<sub>1-x</sub>Zn<sub>x</sub>S/ZnS core/shell QD used here has value of  $x=0.85$ . The reason for the choice of particular value of  $x=0.85$  is being the high blue emission obtained [18]. The quenching in PL intensity with increasing concentration and interaction between guest QD and host nematic LC have been analyzed and discussed. The core finding in the investigation reports the quenching in PL with increased UV absorbance and small band gap for QD dispersed nematic system that find application in photovoltaic, photonic and display devices.

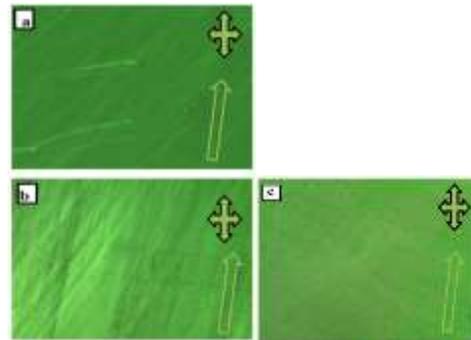
## II. EXPERIMENTAL DETAILS

We have used the positive dielectric anisotropy nematic liquid crystal mixture LC- 2020 [2, 22] which is laterally fluorinated 4'-alkylphenyl-4-isothiocyanatotolanes and exhibits the phase sequence as follows:



The synthesis of the nematic mesogen LC-2020 was carried out by our collaborating research group Institute of Chemistry, Warsaw Poland [2, 22]. Cd<sub>1-x</sub>Zn<sub>x</sub>S/ZnS core/shell QD used here for the dispersion, are semiconducting quantum dots with  $x=0.85$  and size 8.5 nm (core diameter 5nm and shell diameter 3.5 nm) [6]. The QD dispersed nematic system have been prepared by taking an appropriate amount in the weight ratio (0.1% wt/wt and 0.25% wt/wt MIX 1 and MIX 2) of the QDs into pure

nematic and then homogenized with an ultrasonic mixer at 108o C for 1 hour and uniform dispersion of QD was ensured by polarizing micrographs at room temperature 30o C as shown in Fig.1 [2]. The LC sample cells of thickness 6μm used to fill the dispersed mixtures have been fabricated by using highly conducting optically transparent sputtered indium tin oxide (ITO) glass substrates.



**Fig.1. The polarizing optical micrographs (POMs) of (a) pristine nematic LC (b) QD dispersed nematic system MIX 1 and (c) MIX 2 under crossed polarizer condition at 30oC [Ref. 2]. (a), (b), (c) corresponds to homogenous distribution of QD in nematic system. Crossed arrows show under crossed polarizer condition whereas tilt arrow represents the rubbing direction.**

UV visible absorption study of pristine and QD dispersed system have been carried out using UV visible spectrophotometer (ELICO, SL 210) for a wavelength interval of 250-700 nm at room temperature in spectrum mode [23]. In the present study, the optical band gap of the QD dispersed nematic system was evaluated with the help of Tauc plot [24]. The curve between  $(\alpha h\nu)^2$  versus  $(h\nu)$  has been plotted to calculate the optical band gap which has been obtained by extrapolating the linear portion of the plot on the  $(h\nu)$  axis, here  $\alpha$  is the absorption coefficient calculated by using Lambert beer law [25],  $h$  is the plank's constant,  $\nu$  is the corresponding frequency ( $\nu = hc/\lambda$ ,  $c$  is the speed of light,  $\lambda$  is the wavelength) and  $(h\nu)$  is the photon energy. The Photoluminescence spectra of pristine and QD dispersed system have been carried out using Cary eclipse fluorescence spectrophotometer (Agilent technologies) in scan mode using 5 nm slit width and 350 nm excitation wavelength [23]. The FTIR study has been carried out using Fourier transform infrared spectrophotometer (IR Affinity-1 Shimadzu) in the wavelength range of 400-4000 cm<sup>-1</sup>. In the experiment background scan and reference scan have been filtered [23]. The detailed information about the experimental design has already been reported by our group [2, 23].

III. RESULTS AND DISCUSSION

Fig.2. A. represents the variation of UV absorbance over whole wavelength range in spectrum mode with concentration %wt/wt of core/shell QD in nematic liquid crystal LC-2020 in 3 dimensions. Panel B illustrates 2-D plot for the UV-visible absorbance of pristine LC and QD dispersed nematic system on wavelength scale. The inset of the panel B shows the absorbance for core/shell QD. It has been found that UV absorbance increases for QD dispersed system than that of pristine LC. The increment in the magnitude of absorbance is due to charge transfer between the mesogenic molecules and core/shell QD which generates net charge on QD resulting in the permanent dipole moment in them [26]. There is a formation of charge transfer complexes in the doped system due to electronic coupling between the HOMO (highest occupied molecular orbital) of the donor and LUMO (lowest unoccupied molecular orbital) of the acceptor molecule [26]. It is clear from the figure that the centre of absorption peaks was found at 348 nm. It has been attributed to  $\pi-\pi^*$  excitation of  $\pi$  electron system. The low absorption of UV light on either side of absorption peak implies that this region is transparent for UV absorption [26]. The broadness in the peak has been noticed which arises from the absorption of radiation over the band of wavelength (i.e. band spectrum). The origin of band spectrum is probably due to the close packing of the molecules in pristine and QD dispersed nematic system. Since the molecules are closely packed in the system, therefore they exert influence on each other which disturbs the numerous energy levels and combine the sharp spectral lines into bands.

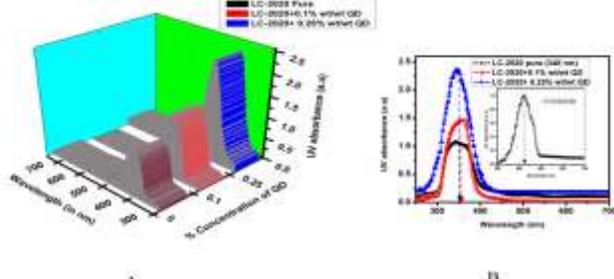


Fig.2. A. 3-D Plot represents the variation of UV absorbance over whole wavelength range in spectrum mode with concentration %wt/wt of core/shell QD in nematic liquid crystal LC-2020. XY face is represented by grey color, X axis depicts wavelength in nm, Y-axis represents UV absorbance and on Z-axis %wt/wt concentration of QD in LC- 2020 is plotted. XY and YZ plane are shown by green and cyan color respectively. Fig. B. represents the 2-D plot of same figure A. it depicts the UV visible spectra of pristine LC and QD dispersed nematic system for both mixtures as a

function of wavelength. Inset in the graph represents absorbance spectra for core/shell QD.

Fig.3 shows the plot for  $[(\alpha h\nu)]^2$  and optical band gap. For pristine LC and QD dispersed nematic system the magnitude of optical band gap has been found to be 3.25 eV, 3.20 eV and 3.15 eV respectively. The nature of plot discloses the direct allowed transition. The shape of the transition provides insight into the electron interband transition. The transition region is the absorption edge. The decrement in the band gap for QD dispersed nematic system is explained on the basis of Mott and Davis model [24, 26] according to which shallow states causes band gap to decrease. The increased UV absorbance with decreased band gap implies that the electron transfer from lower state to higher state takes place with strong photo absorption therefore emission should reduce for QD dispersed system (i.e. luminescence quenching). This has also been evident from the PL result discussed in the later part of the paper. Hence our study for UV absorbance and Band gap supports the phenomenon of quenching in PL intensity. The UV-visible absorbance study finds its application in the use of QD dispersed nematic composites for UV light storage devices and fabrication of photovoltaic devices. These devices directly generate electricity by the absorption of UV light via an electronic process that occurs naturally in semiconducting materials, electrons in these materials are released by the solar energy which can be induced to travel through an electric circuit [28-31].

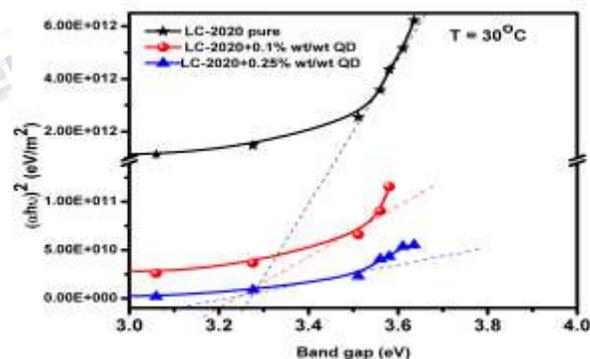
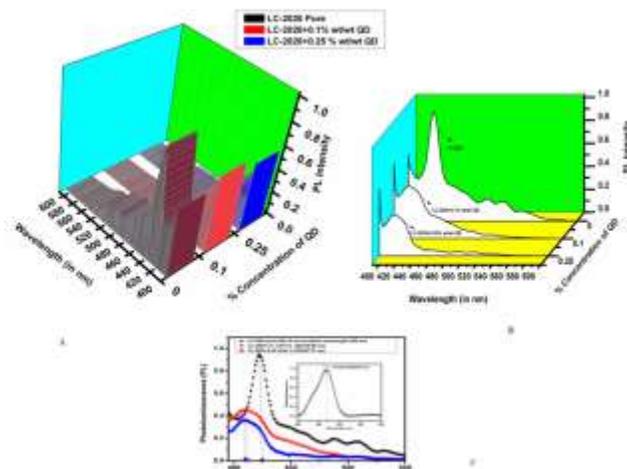


Fig.3. The plot for  $(\alpha h\nu)^2$  versus band gap for pristine LC and QD dispersed nematic system for both mixtures.

Fig.4. A. Represents the variation of normalized PL intensity over whole wavelength range with concentration %wt/wt of core/shell QD in nematic liquid crystal LC-2020 in 3 dimensions. It shows quenched PL spectra, panel B shows the blue shift in PL spectra in 3 dimensions. However, panel C depicts the normalized Photoluminescence (PL) spectra of pristine and QD dispersed nematic system with variation of

concentration in 2 dimensions. The excitation wavelength for PL is taken to be 350 nm. It is clear from the figure that the maximum PL intensity has been observed at a wavelength of 422.61 nm, 410.80 nm and 407.97 nm respectively for pristine LC, MIX 1 and MIX 2. This has been attributed to blue shift in PL spectra. The PL intensity in both QD dispersed system (MIX 1 and MIX 2) has been quenched by 51.7% and 62.7% as compared to pristine LC. It can be interpreted that both Core/shell QD (inset) and pristine nematic LC have emission bands which can be combined constructively so that some portion of the PL of pristine LC might be absorbed by QD to give quenched PL intensity for dispersed system. In addition to this the presence of QD significantly modifies the surface topology which leads to change in PL emission [6]. PL luminescence quenching suggested that the interaction between LC molecules and core/shell QD leads to non radiative recombination of excitons in photo excited LC molecules. This has also been evident from the FTIR spectra discussed later part of the paper. The broadness in the PL peaks is due to the fact that the photons on either side of emission peaks have smaller probability of reabsorption than the photons at the peak center.



**Fig.4.A.** 3-D Plot represents the variation of PL intensity over whole wavelength range with concentration %wt/wt of core/shell QD in nematic liquid crystal LC-2020 which shows the quenching in PL intensity. XY face is represented by grey color, X axis depicts wavelength in nm, Y-axis represents PL intensity and on Z-axis %wt/wt concentration of QD in LC- 2020 is plotted. XY and YZ plane are shown by green and cyan color respectively. Panel B. 3 D plot represents the quenching in PL intensity with the variation of both wavelength and concentration %wt/wt of QD in nematic LC- 2020. The plot represents the blue shift in wavelength along with quenched PL intensity. Panel C.

*Depicts the 2-D plot for Photoluminescence spectra of pristine LC-2020 and QD dispersed nematic system for both mixtures as a function of wavelength. Inset in the graph shows the PL spectra of core/shell QD.*

The quenching in PL intensity for dispersed system is also confirmed from the Gaussian fit of PL spectra as shown in Fig.5. The normalized PL intensity has been fitted with Gaussian equation for pristine and QD dispersed system. The inset of Fig.5 shows the Gauss fit for core/shell QD. It is clear from the fitted curve that the centre of PL peaks was obtained at 423 nm, 411 nm and 408 nm for pristine LC, MIX 1 and MIX 2 respectively which is in consistent with the experimental plot for PL discussed earlier. FWHM obtained from Gaussian fit has been plotted in Fig.6. It shows that FWHM for QD dispersed nematic system has been increased. For core/shell QD, full width half maxima (FWHM) is 23 nm which is less than 25 nm. Therefore, it has narrow spectral band width with high PL Quantum yield and strong band edge emission [6, 18]. FWHM for pristine LC has been found to be ~ 13 nm where as for MIX 1 and MIX 2 it is found to be 35.39 nm and 39.44 nm respectively. Lower magnitude of FWHM ( $FWHM \leq 25$  nm) of pristine LC is attributed to near band emission and high PL quantum yield as it results sharper peak in PL spectra as compared to dispersed system (shown in Fig.4) [12, 27]. The increment in FWHM ensures that QD dispersed nematic system have high PL quenching efficiency for the reason that as the magnitude of FWHM increases ( $FWHM > 25$  nm) the broadness in the peak also increases which results in quenched PL intensity due to surface level defect transitions and suppression of band edge emissions [27]. Inset of Fig.6 shows that PL emission peak reduces with increase in concentration of QD i.e. it shifts towards the lower wavelength side (blue shift). Therefore, the inverse proportionality in the variation of PL emission peaks and FWHM satisfy the blue shift in PL spectra. This blue shift in emission peak corresponds to change in quantum confinement effects associated with QD. The outcome of quenching in PL intensity makes QD behaves as quencher for dispersed system which has been suitable for the use in liquid crystal optical devices for imaging and processing data as well as developing LC technology [17]. The PL emission in visible region (near 400 nm of violet color) might find its use for UV to visible light conversion thus it has been inferred that the present study has relevance in the photonic applications [28-31].

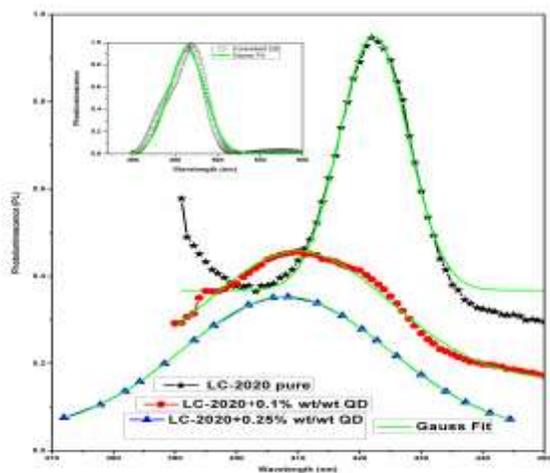


Fig.5. Gaussian fit of Photoluminescence as a function of wavelength for pristine LC and QD dispersed nematic system for both the mixtures. Inset in graph represents Gauss fit of core/shell QD.

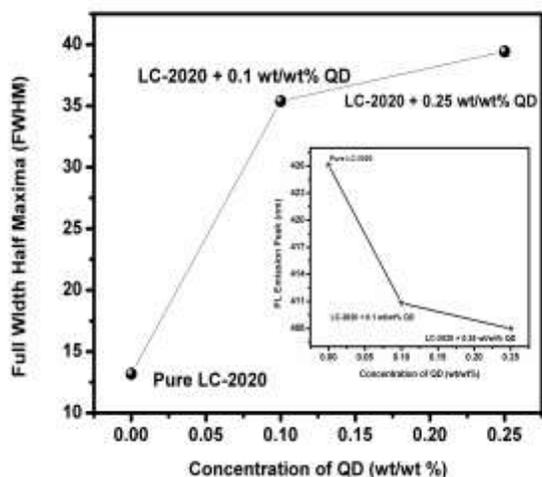
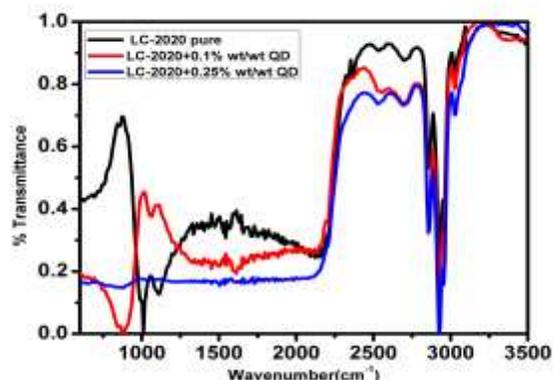


Fig.6. Variation of full width half maxima (FWHM) as a function of % wt/wt concentration of QD in LC system. Inset in the graph shows PL emission peak for different concentrations.

Fig.7 shows the FTIR spectra for pristine and QD dispersed system. It explains the molecular dynamics of QD dispersed nematic system and supports our observation for quenching in PL luminescence. The non radiative recombination process deduced from PL results for QD dispersed system is also

confirmed from FTIR. The PL luminescence quenching is evident from anharmonic molecular vibrations of C-H group of alkyl chain which acts as photo excitation energy acceptor. Therefore this transition is non-radiative electron-vibrational energy transfer [16]. It is clear from the figure that the absorption occurs at the resonant frequencies where the frequency of absorbed radiation matches the vibrational frequency of bond or group. Since the vibrational energy states of nematic molecules are closer than the electronic energy level thus lower energy photon are sufficient to cause vibrational changes. The normalized transmittance of pristine nematic material has been reduced slightly after the dispersion of QD. This is attributed to absorbance of incident light by the QD. The emergence of peaks or bands at 800-1250  $\text{cm}^{-1}$  is intrinsic bands in all thermotropic LCs [6]. The bands in the interval of 650-1500  $\text{cm}^{-1}$  reveals that the presence of QD in nematic system not only seizes some intrinsic vibrations but also originates few extrinsic vibrations. The appearance of dominant peak at 864  $\text{cm}^{-1}$  has been observed for pristine LC, whereas for MIX 1 the dip in the same peak has been noticed and the disappearance of this peak takes place for MIX 2. The appearance of this peak has been attributed to the presence of ionic impurities in pristine LC medium whereas for QD dispersed nematic system (i.e. MIX 1 and MIX 2). The dip and the disappearance have been associated to the reduction in the ionic impurities due to the adsorption of these impurities on the shell surface of core/shell QD [2]. The emergence of extrinsic bands for MIX 1 near 1000  $\text{cm}^{-1}$  has been originated due to the strong interaction between nematic molecules and QD [2] while these bands disappears for MIX 2 because of the dominant interaction between QD molecules [2]. The dip in the pristine LC near 1000  $\text{cm}^{-1}$  has been due to the effective interaction between nematic molecules [2]. The presence of peak at 2353  $\text{cm}^{-1}$  has been due to the presence of C-O single bond group in LC medium. 1500-4000  $\text{cm}^{-1}$  is the functional group regimes which are the characteristic of specific kind of bonds and used for the identification of functional groups. There are vibrational bands of C-F bond,  $\text{C}\equiv\text{C}$  triple bond and -NCS group ( $\text{N}=\text{C}=\text{S}$ ) in nematic mesogen. Triple bonds have small intensity and are observed in the range 2200-2250  $\text{cm}^{-1}$ , -NCS group are observed in the range of 2050-2100  $\text{cm}^{-1}$  [8]. The dominant peaks at 2862  $\text{cm}^{-1}$ , 2885  $\text{cm}^{-1}$  and 2931  $\text{cm}^{-1}$  are due to symmetric & asymmetric C-H stretching vibrations for saturated aliphatic chain. 2400-2800  $\text{cm}^{-1}$  regime probably indicates the origin of interaction between core/shell QD and pristine LC-2020. Peaks near 3000  $\text{cm}^{-1}$  have been appeared due to the interaction between core/shell QD and aromatic core of pristine LC which rearranges C-H vibrations.



**Fig.7. Graphical representation of FTIR spectra for pristine LC and QD dispersed nematic system for both mixtures in transmission mode on wave number scale at 30oC.**

#### IV. CONCLUSION

In concise, the present paper reveals the influence of QD on certain optical properties of nematic liquid crystal. The alterations in nematic liquid crystal properties are strongly dependent on the dopant concentration. A remarkable increase in UV absorbance along with the decrement in band gap and quenched PL spectra for higher concentration of QD dispersed nematic system has been the core finding in the present investigation. The results of UV visible absorbance and band gap suggested that emission should reduce for QD dispersed system (i.e. PL quenching) due to strong photo absorption. This has been confirmed from the PL spectra and FWHM values obtained from the Gaussian fit of PL curve. Increased UV absorbance and quenched PL in QD dispersed nematic system discloses the effect of prominent interaction between aromatic rings of nematic LC molecules and QD which was also confirmed from FTIR spectra. Asymmetric and symmetric C-H stretching vibrations are confirmed from FTIR spectra which had been the support for the quenched PL intensity. Reduced band gap, increased UV absorbance and PL quenching of QD dispersed system have gained true profit interest substantially vested at the time of applications in fabrication of photovoltaic devices, UV to visible light conversion, UV light storage devices and LC based optical devices. The outcome of the present investigation provides its application in the field of photonic and photovoltaic devices.

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