



Time-resolved fluorescence and absence of Förster resonance energy transfer in ferroelectric liquid crystal-quantum dots composites

D.P. Singh ^{a,b,*}, S. Pandey ^b, R. Manohar ^{b,**}, S. Kumar ^c, G.H. Pujar ^d, S.R. Inamdar ^d

^a *Unité de Dynamique et Structure des Matériaux Moléculaires, EA 4476, Université du Littoral Côte d'Opale, F-59140 Dunkerque, France*

^b *Liquid Crystal Research Lab, Physics Department, University of Lucknow, Lucknow 226007, India*

^c *Raman Research Institute, Bangalore 560080, India*

^d *Department of Physics, Karnatak University, Dharwad 580003, India*

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ABSTRACT

The present article deals with the interaction of octadecylamine (ODA) capped cadmium selenide quantum dots (CdSe QDs) and two ferroelectric liquid crystals (FLCs) having different values of spontaneous polarization (Ps). The pristine FLCs and FLC-QDs composites have been investigated by using steady-state and time-resolved fluorescence spectroscopic techniques. Stokes shift and the preferred alignment of FLC molecules in the presence of QDs are found to depend on the secondary order parameter of FLCs (i.e. Ps). The secondary fluorescence emissions have been strongly influenced by the concentration of QDs. The Förster resonance energy Transfer (FRET) was absent in both the composites. This article discloses the role of various factors, like relative orientation, dipolar interaction between donor and acceptor and the proximity of QDs (> 10 nm), which might be responsible for the absence of FRET. FLC-QDs composites do not form a new class of luminescent material though the fluorescence properties of host FLCs are tailored in the presence of QDs by means of the change in molecular alignment. The change in lifetimes of FLC-QDs composites has not been observed.

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1. Introduction

Size dependent fluorescence and electrical properties of quantum dots (QDs), due to their quantum confinement effects, make them suitable to be used as dopant in liquid crystals (LCs) [1]. The physical properties and material parameters of LCs are found to be altered significantly with the addition of QDs [2–5]. Semiconductor QDs are the most attractive nanomaterials for the phenomenon of charge transfer, photo or electro-luminescence and self-assembling property, therefore they have been used in a wide range of applications in the fabrication of various devices [6]. Transient fluorescence and ultra short decay lifetime are focal attractive features of QDs which have been subjected to an extensive research [7,8]. Various research articles have served the phenomenon of charge/energy transfer to and from QDs and various possible reasons which affect the transfer dynamics for the

different combinations of QDs [9,10]. It is also reported that the photoluminescence intermittency of QDs has been influenced due to the presence of dielectric medium [11,12]. The effects of dynamic quenching and in particular the process of Förster resonance energy transfer (FRET) are now widely studied in photo chemistry and photo-biology, wherein, signal intensity is often used to quantify FRET efficiency. Fluorescence lifetime is a more reliable parameter largely independent of the fluorescence intensity and fluorophore concentration. Since this process is affiliated with an energetically unstable state, fluorescence lifetime can be sensitive to a great variety of internal factors defined by the fluorophore structure and external factors like temperature, polarity, and the presence of fluorescence quenchers. Fluorescence lifetime is the time required by a population of excited fluorophores to decrease exponentially to N/e via the loss of energy through fluorescence and other non-radiative processes, therefore; the monitoring of fluorescence lifetime of LCs in the presence of QDs is an important process to study the FRET mechanism. Time resolved fluorescence measurements provide significant and precise information about the molecular interactions in comparison to the steady state measurements. The charge and energy transfer in QDs-LC composites has been investigated recently to analyze the influence of QDs on the fluorescence property of LCs and the

* Corresponding author at: Unité de Dynamique et Structure des Matériaux Moléculaires, EA 4476, Université du Littoral Côte d'Opale, F-59140 Dunkerque, France.

** Corresponding author.

E-mail addresses: dpsr4u@gmail.com (D.P. Singh), rajiv.manohar@gmail.com (R. Manohar).

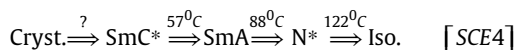
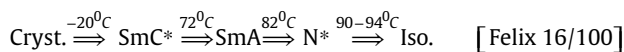
transfer of physical quantities like charge and energy between the skeleton of QDs and LCs [6,13,14] but these studies are only limited with nematic and cholesteric LCs. These observations on Smectic C* (SmC*) have not been reported so far.

In the present article, we have investigated the interaction of ODA capped CdSe QDs and two FLCs having different values of spontaneous polarization with the help of polarizing optical microscopy (POM), steady-state and time-resolved fluorescence spectroscopic techniques. Interestingly, the fluorescence (Förster type) resonance energy transfer was found to be absent in the FLC-QDs composites. In QDs and doped QDs, FRET is commonly observed phenomenon but in the FLC-QDs composite, even with the variation of QDs concentration, FRET was not observed. The main focus of the present article is to point out the factors which influence the occurrence of FRET phenomenon. We conclude that the FLCs do not form a new class of luminescent material after the addition of QDs because we have observed almost same lifetimes for them.

2. Materials and methods

Two different commercial FLC mixtures viz. Felix 16/100 (spontaneous polarization (P_s) and tilt angle (θ) values; $P_s = 10.5 \text{ nC/cm}^2$, $\theta = 27.1^\circ$ at 25°C) and SCE4 ($P_s = 6.1 \text{ nC/cm}^2$, $\theta = 23^\circ$ at 25°C) were used as host material whereas CdSe QDs of 3.5 nm (diameter) capped with octadecylamine ligand was used as dopant

[15]. Both FLCs are commercial mixtures; therefore their chemical structures are not available. The phase sequences of both the FLCs are as follows:



The dopant was dispersed in FLCs in 0.1 and 0.3 wt% and termed as F1 (Pure FLC Felix 16/100), F1QD 0.1 (Felix 16/100 + CdSe QDs 0.1 wt%), F1QD 0.3 (Felix 16/100 + CdSe QDs 0.3 wt%), S1 (Pure FLC SCE4), S1QD 0.1 (SCE4 + CdSe QDs 0.1 wt%) and S1QD 0.3 (SCE4 + CdSe QDs 0.3 wt%). The homogenous dispersion of FLCs and QDs was obtained by the ultrasonic mixing of composites for 3 h at room temperature (RT). All the optical measurements were carried out at RT (25°C).

Pure FLCs and FLC-QDs composites were dissolved in methanol (HPLC grade). The UV-vis absorption spectra of all samples were obtained using JASCO UV-vis NIR Spectrophotometer (Model V-670) whereas fluorescence spectra were recorded using Spectrofluorometer (JY Horiba, FloroMax-4).

The fluorescence lifetimes were measured employing picosecond time domain spectrometer based on Time Correlated Single Photon Counting (TCSPC) technique (IBH Jobin Yvon 6.1) described elsewhere [16]. FLCs and FLC-QDs composites were excited at 278 nm (F series) and 290 nm (S series), respectively using

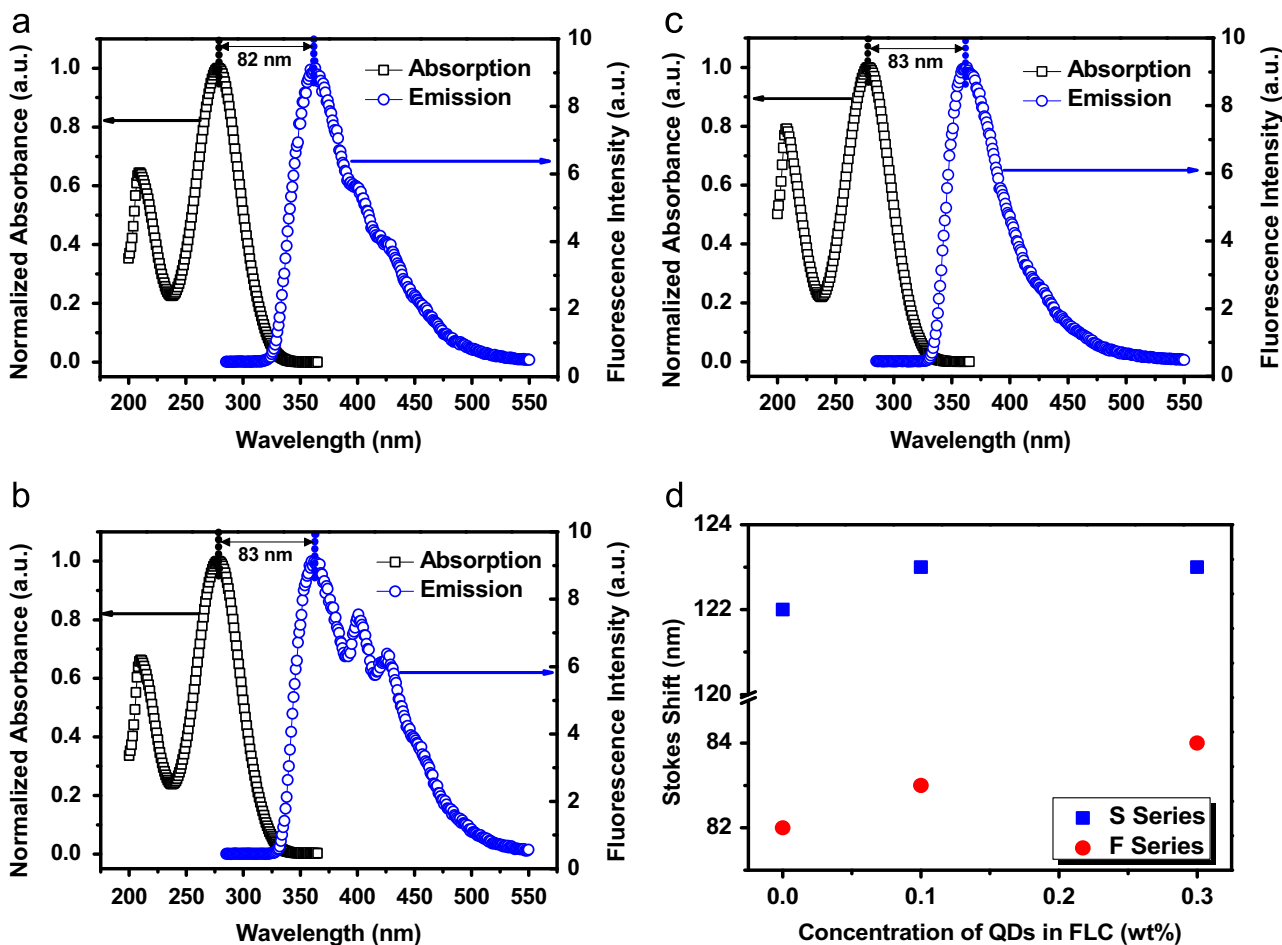


Fig. 1. Normalized UV absorbance and fluorescence of (a) Felix 16/100 [F1], (b) 0.1 wt%, (c) 0.3 wt% CdSe QDs dispersed in FLC whereas (d) represents the Stokes shift in FLC-QDs composites with the variation of QDs concentration. Absorbance study was performed in colloidal state (at RT) by taking HPLC grade methanol (mg/ml) as solvent. The excitation wavelength was 278 nm for all samples.

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