Room-Temperature Single Photon Sources with Fluorescent Emitters in Liquid Crystal Hosts


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Abstract: A single-photon source on demand based on single CdSe quantum-dot fluorescence in a chiral-photonic-bandgap liquid-crystal microcavity manifests itself in observed fluorescence antibunching. The aligned liquid crystal host also provides deterministically polarized fluorescence of single emitters. ©2007 Optical Society of America

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Our project is aimed at developing an efficient, room-temperature single-photon source (SPS) on demand with definite polarization for quantum information, using single-emitter fluorescence in liquid crystal (LC) hosts. This SPS is a room temperature alternative to cryogenic SPSs based on semiconductor heterostructures. LCs can provide both microcavity assembly as well as deterministic polarization of emitted photons. Earlier we reported that oxygen depleted LC significantly reduced bleaching of terrylene molecules [1]. In addition, LC can provide SPS tunability.

In this paper, we report experimental results of two SPSs with different emitters embedded in LC hosts. (1) The first SPS is based on the single colloidal CdSe quantum dots (QD) suspended in a LC host self-assembled in a chiral photonic bandgap structure. This structure can provide not only spontaneous emission enhancement and a diminishing of the fluorescence lifetime, but also circular polarization of definite handedness even for emitters without a dipole moment. In addition, because the refractive index $n$ varies gradually in chiral structures rather than abruptly, there are no losses into the waveguide modes, which arise from total internal reflection at the border between two consecutive layers with different $n$. We report here for the first time fluorescence antibunching of a QD doped in a LC. Earlier we reported fluorescence antibunching of terrylene dye embedded in LC host [1].

Figure 1. (a) Confocal microscope and a Hanbury Brown and Twiss setup. (b) Selective transmission of four different chiral photonic bandgap cholesteric LC hosts (blue lines) and the fluorescence spectrum of the CdSe QDs (red line).

Figure 1 (a) shows the experimental setup for fluorescence antibunching measurements and imaging of single-QD fluorescence. It is based on a home-made confocal microscope and a Hanbury Brown Twiss arrangement with an avalanche photodiode module (APD) in each arm. We illuminated our samples with 76 MHz pulsed, 532 nm light with 6 ps pulse duration. To measure photon statistics and fluorescence lifetime, a TimeHarp 200 computer card with a start and stop channel was used. The samples were prepared by mixing a highly diluted solution of QDs with a monomeric (fluid-like) cholesteric LC (CLC). CLCs with different chiral structure pitch were prepared by selection of proper concentration of chiral additive in nematic. The QD-doped CLCs were planar aligned. Figure 1 (b) shows selective transmission of such chiral photonic bandgap structures. Figure 1 (b) also shows the fluorescence spectrum of CdSe QD with the center of the fluorescence peak near 579 nm.
Figure 2 (a) is a typical confocal fluorescence image of a single QD in a chiral photonic band-gap structure. Note that the dark horizontal stripes in the pattern are the result of single QD blinking, which is a characteristic property of single-QD fluorescence. Figure 2 (b) shows the coincidence count histogram at different interphoton times. It was obtained by using a Hanbury Brown and Twiss setup with two APDs and a start-stop protocol. One sees that the peak at zero interphoton time is clearly smaller than any of the other peaks, which shows an antibunching property, a proof of the single-photon nature of the source.

Figure 2. (a) A typical confocal fluorescence microscope image of a single CdSe QD in a CLC host. (b) Histogram of coincidence counts of a single QD fluorescence in a CLC host showing antibunching.

(2) For the second experiment we used DiI dye in a planar-aligned glassy nematic LC host which is solid at room temperature (see Figure 3 with single-molecule fluorescence image of DiI dye in a glassy nematic LC host). Experimental setup for measurements of deterministic linear polarization of these single emitters is depicted in Figure 4 [2]. In difference with the Figure 1(a) setup, for this experiment we used Witec alpha-SNOM microscope in a confocal transmission mode, a 532-nm laser in a cw-mode, and a polarizing beam-splitter cube. Figure 5 illustrates deterministically polarized fluorescence from aligned single molecules of DiI dye. Polarization anisotropy is defined here [3] as 
\[ \rho = \frac{I_{\text{par}} - I_{\text{perp}}}{I_{\text{par}} + I_{\text{perp}}} \]
where \( I_{\text{par}} \) and \( I_{\text{perp}} \) are fluorescence intensities for polarization components parallel and perpendicular to the alignment direction. These two polarization components in the plane of the sample have been separated with a polarizing beamsplitter cube. Figure 5 shows an assymetric histogram of the polarization anisotropy of 38 dye molecules in the planar aligned glassy nematic LC host. Ref. 3 shows both theoretically and experimentally that in the case of randomly oriented dipole moments of this dye, the histogram is absolutely symmetrical. For DiI dye in a planar-aligned nematic host, the polarization direction of the fluorescence of single molecules is predominantly in the direction perpendicular to the alignment of LC molecules because of the specific molecular structure of this dye [2].

Figure 2. (a) A typical confocal fluorescence microscope image of a single CdSe QD in a CLC host. (b) Histogram of coincidence counts of a single QD fluorescence in a CLC host showing antibunching.

Figure 3. Confocal image of single-molecule fluorescence of DiI dye dopped in a planar-aligned glassy nematic LC. (10 μm x 10 μm scan). Maximum fluorescence wavelength is ∼580 nm.

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Figure 4. Schematics of polarization measurements.

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Figure 5. The histogram of polarization anisotropy of 38 molecules of DiI dye in glassy nematic LC.

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References: