LAB 3-4, PHY434. Single Photon Source: Confocal Microscope Imaging of Single-Emitter Fluorescence and Hanbury Brown and Twiss setup for Photon Antibunching Measurements

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ABSTRACT

We present results indicating antibunching of two CdSe quantum dot single photon sources that fluoresce at 800nm and 705nm. We obtained antibunching histograms in TimeHarp using the Hanbury Brown and Twiss setup for photon antibunching. To do this, we prepared QD samples with concentrations of 10 nanoMol, 1 nanoMol, and 100 picoMol and placed the QD samples in a cholesteric liquid crystal host, which we exposed to 632.8nm HeNe laser light in a raster scanning confocal microscope setup. Observations of the quantum dot fluorescence were made with Avalanche Photo-Diodes through LabVIEW, and an EM-CDD camera through Andor-Solis.

Keywords: single photon source, single emitter, Hanbury Brown and Twiss, antibunching, cholesteric liquid crystal, photonic bandgap material, fluorescence lifetime

1 INTRODUCTION AND BACKGROUND

1.1 *Single Photon Sources*

Quantum information, communication, and security are growing fields of research whose practical implementation in computation, communication, banking, and security require the use of single photon sources to provide provably secure keys [1]. Single-emitters are a completely quantum prediction, as they are equivalent to a zero-valued second order correlation function at t=0, $g^{(2)}(0) = 0$. This is not possible for a classical light source like a laser. Hence, even a greatly attenuated laser beam is not a source of antibunched light. There are currently several options to create room temperature single photon sources. These include single molecules, di-molecules, semiconducting colloidal quantum dots (e.g. in nanocrystals), and color centers in nanodiamonds. In this lab, we used colloidal quantum dots.

To achieve a single photon source, we can distribute a certain concentration of QD solution with a host bandgap material such as cholesteric liquid crystals. By focusing a laser beam on the sample, if single-emitter are isolated in the CLC-host, we can achieve single excitations of the electrons in an emitter and the relaxation of that CdSe emitter will give off fluorescent single-photon light. A schematic of this process is shown in Figure 1.

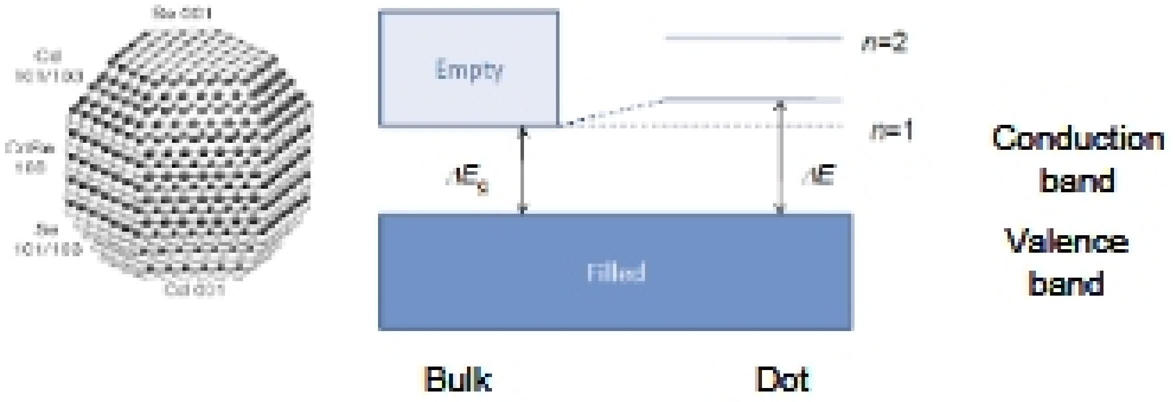

Fig. 1 CdSe QD energy level diagram of excitation and fluorescent emission

1.2 *Photon Antibunching*

Figure 2 shows a schematic of the Hanbury Brown and Twiss photon antibunching setup; they were the first researchers to observe correlations between pairs of photons in 1956 [2]. The first experiment that demonstrated the existence of photon antibunching was conducted by Kimble, Dagenais, and Mandel in 1977 [3]. This was evidence of the quantization of the electromagnetic field. In contrast, the classical EM field is simply one that behaves according to Maxwell's equations. For classical fields, the correlations between the intensities of the transmitted I_T and reflected I_R beams at a beam splitter are given by the *degree of second-order (temporal) coherence*, or $g^{(2)}{}_{T,R}(\tau) = \dfrac{\langle I_T(t+\tau)I_R(t)\rangle}{\langle I_T(t+\tau)\rangle\langle I_R(t)\rangle}$, which is a function of the delay time τ between the intensity measurements. We have a stationary light source so we can interpret the averages as ensemble averages and not time averages. Thus, for our 50/50 beam splitter we can assume the form:

$$g^{(2)}{}_{T,R}(0) = g^{(2)}{}_{I,I}(0) = \frac{\langle I_I(0)^2\rangle}{\langle I_I(0)\rangle^2} = g^{(2)}(0).$$

for our the second order coherence at zero time delay. But the Cauchy-Swartz inequality allows one to show that the above implies: $g^{(2)}(0) \geq 1$, classically. The quantum mechanical equivalent form of the preceding shows that for a single-emission of a photon, we should have $g^{(2)}(0) = 0$, but anything that violates the classical inequality above is considered a quantum effect known as antibunching. The antibunching derivation follows:

$$g^{(2)}{}_{T,R}(0) = \frac{\langle \hat{n}_T\,\hat{n}_R\rangle}{\langle \hat{n}_T\rangle\langle \hat{n}_R\rangle} = \frac{\langle a^*{}_T a^*{}_R\; a_T a_R\rangle}{\langle a^*{}_T a_T\rangle\langle a^*{}_R a_R\rangle} = \frac{\langle \hat{n}_I(\hat{n}_I - 1)\rangle}{\langle \hat{n}_I\rangle^2} = g^{(2)}{}_{I,I}(0) = g^{(2)}(0)$$

$$\text{which, for a single incident photon, } n_I = 1 \Rightarrow g^{(2)}(0) = 0$$

where the above comes from assuming the EM field is quantized and the rewriting the creation and annihilation operators so that the number operator of the incident field determines the second order correlation. Thus, we see that for a single incident photon, we expect a minimum at $g^{(2)}(\tau = 0)$. The 1977 experiment from [6] achieved $g^{(2)}(0) = 0.4$.

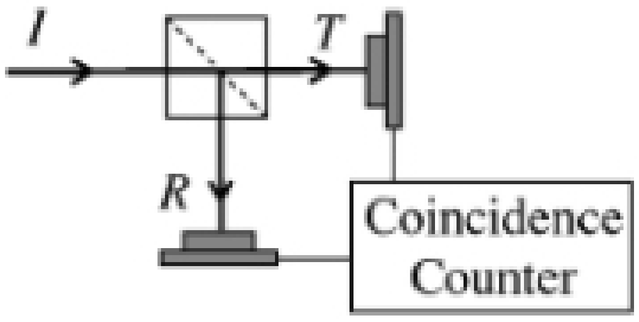

Fig. 2: Hanbury Brown and Twiss setup for photon antibunching

1.3 *Cholesteric Liquid Crystal, used as a 1D Photonic Bandgap Material*

Cholesteric liquid crystals (CLCs) have rod-like molecules having a short chiral tail that, for circularly polarized light with counter-handedness to the rotation of the CLC, induces a globally periodic and helical structure in the CLC which makes the CLC behave like a 1D photonic bandgap material after it is given a unilinear planar-aligned shearing force. The pitch, P_0, is the distance over which a 360° rotation in the CLC structure is made, which is displayed in Fig. 3A.

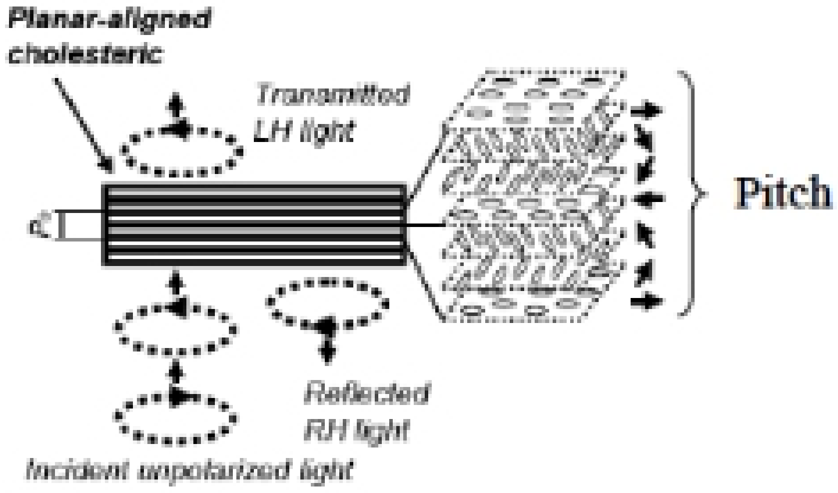

Fig. 3A Schematic CLC periodic structure, and transmission/reflection within CLC wavelength bandgap

Cholesteric liquid crystals are perfect reflectors under the Bragg condition within $\Delta\lambda = \lambda_0 \, \Delta n / n_{avg}$ in a wavelength band centered on $\lambda_0 = n_{avg} P_0$, where n_{avg} is the average of the ordinary and extraordinary refractive indices of the CLC.

Thus, if emitting wavelengths are within the CLC bandgap there is suppressed emission. But at the band-edge, emission is enhanced. This is analogous to a laser cavity, since at the bottom of the stop-band the periodic structure acts like completely reflecting mirrors. At the band-edge, however, it acts like partially transmitting mirrors. Figure 3B, taken from the lab manual, shows the plot of a CLC spectral transmittance with a bandgap; one also sees the fluorescence spectrum of CdSe quantum dot (579nm peak wavelength) in the CLC-host, which has a maximum at the bang-edge.

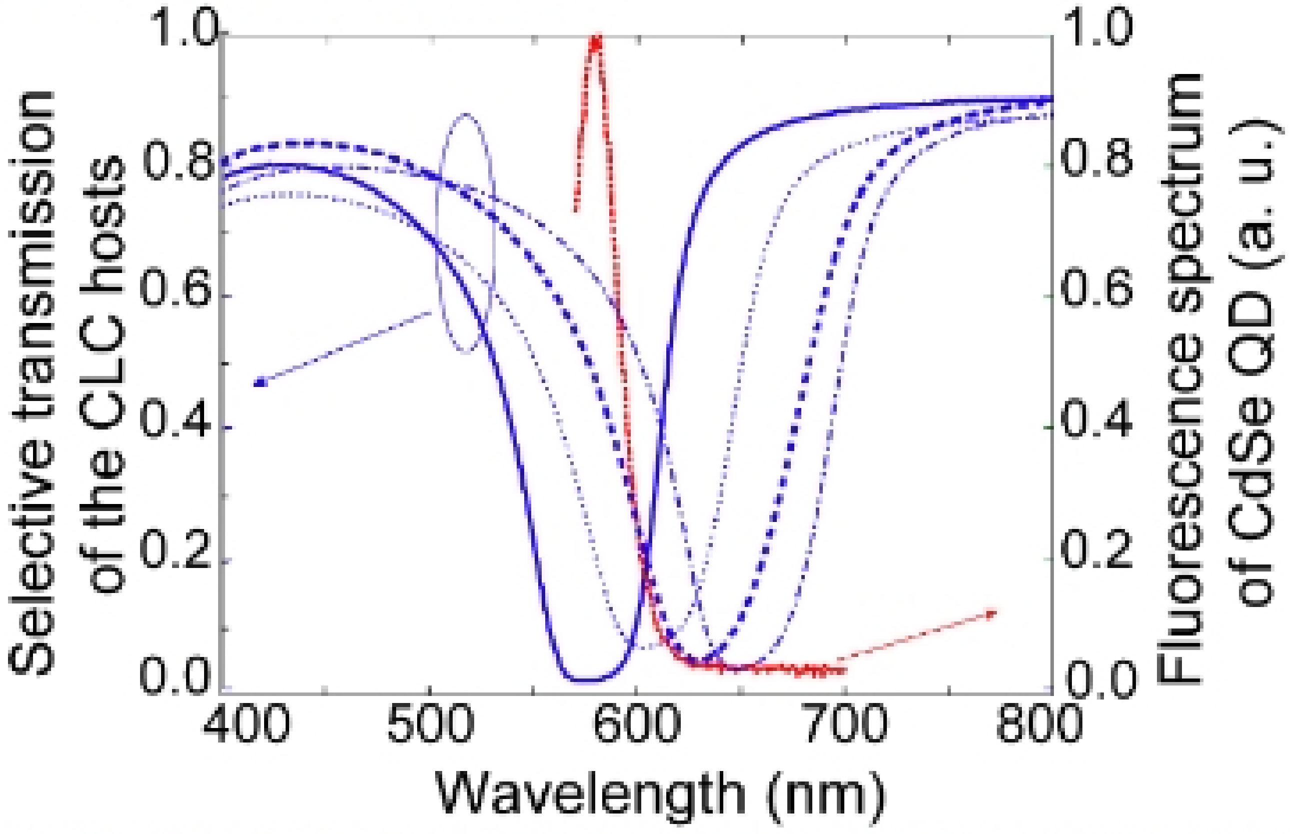

Figure 3 B. Selective reflection curves of several cholesteric liquid crystal layers (blue curves). Red curve shows fluorescence spectrum of CdSe quantum dots.

2 EXPERIMENTAL SETUP AND PROCEDURE

2.1 *Experiment Setup*

Fig. 4 shows our experimental setup. Both a red 632.8nm Helium-Neon (HeNe) laser with 5 mW average power, and a diode-pumped solid-state 532nm laser with 6 ps pulse duration and 76 MHz pulse repetition rate at 40 mW average power were used. We took histographic data for demonstration of fluorescence antibunching from our CdSe quantum dots using the HeNe laser, and took data for the CdSe QDs fluorescence lifetimes using the pulse laser.