Simultaneous excitation of N single colloidal quantum dots

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We report on the implementation of a new microscopy device that allows us to selectively excite up to 20 quantum dots simultaneously and collect their fluorescence. We use it to excite and observe single CdSe/ZnS quantum dots, which are known to be single photon emitters at room temperature. Complex excitation timings can be realised. The microscope works in confocal mode with a resolution of 2 micrometers and a field of view of 20 micrometers. We discuss applications of our device in quantum optics (imaging with incoherent single photons) and quantum cryptography (wavelength multiplexed quantum cryptography and "multi-colour" quantum cryptography).

Introduction

Driven by the development of Quantum Information Science and the perspective of technological applications, much of the work in quantum optics from the late nineties has been devoted to single-photon state generation, photon-pair state generation and photonic entanglement. The fast development of the field has been facilitated by the parallel development of photonics and material science (new high quality materials became available) and nanoscale engineering. Therefore the field is now called Quantum Photonics (QP).
The next step in the development of QP is certainly *scalability*. Large numbers of qubits and large scale entanglement are required for performing non trivial quantum computing tasks. Even for quantum communication, which is known as more tractable than quantum computing, scalability is needed if one wants to build a quantum communication network that is more elaborated than the famous two-end Alice/Bob channel. From a practical point of view, scalable QP requires one to be able to simultaneously address and manipulate large numbers of quantum emitters (single-photon or photon-pair emitters, depending of the application).
The race towards scalable QP has started. In 2008, Zhang et al. demonstrated a technique for creating highly ordered 2D arrays of single CdSe (II-VI semiconductor) colloidal quantum dots (QDs) [1]. These kind of QDs have been previously proven [2, 3] to be very efficient *room-temperature* single-photon emitters. Multiexcitonic emission is inhibited by Auger recombination and radiative efficiency of the remaining exciton can be as high as 98%. To get an even better control on the single-photon emission mode, Qualtieri et al. inserted CdSe/ZnS QDs in vertical micro-cavities arranged in a two-dimensional array [4]. They measured anti-bunching from single emitters and strong cavity-induced reduction of the photon duration. However, in Zhang's and Qualtieri's works, the key-question of scalability ("How to operate many single-photon sources simultaneously?") is not answered.

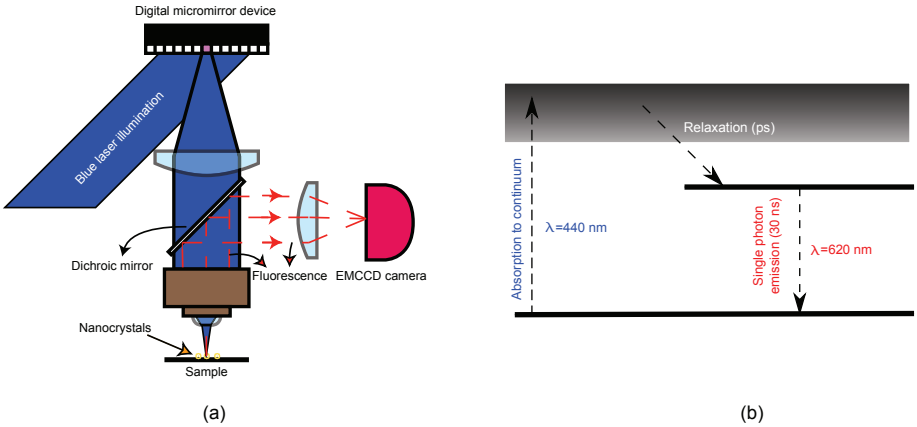

Figure 1: (a) Outline of the experimental setup. (b) Simplified "3-level" approximation of the CdSe/ZnS electrodynamics.

In this paper, we demonstrate a principle that allows us to address a large number of QDs in a very flexible way. Excitation is achieved by optical means (absorption of short-wavelength pump radiation). Pump radiation can be directed on a given QD on demand, allowing complex time patterns. The number of QDs that are excited can be varied in time without any other restrictions than the number of QDs available in the field of view of our device. And most importantly, our system does not require the QDs to be arranged in any specific array (like in [1, 4]), though they must lay on a planar surface. In the following, we describe our scheme, present experimental results, then explain some foreseen applications of our device in quantum cryptography and quantum optics.

Experimental setup and results

Our experimental setup is represented on Fig. 1(a): CdSe/ZnS nanocrystals are excited and observed using a home-made fluorescence microscope. The same objective lens is used to focus the pump beam (blue laser, central wavelength: 440 nm) on the sample and collect photons emitted by the nanocrystals (central wavelength: about 620 nm). The pump beam is produced by a laser diode (PicoQuant, LDH-D-C-440M) that can operate either in cw or pulsed (500 ps) mode. The nanocrystals are observed using a back-illuminated electron-multiplication CCD camera (Andor iXon^{EM}+ 897). Our nanocrystals have a diameter of about 1 nm. When nanocrystal are so small, Auger recombination is very efficient and leads to a blockade effect that prevents multiple excitons in the QD [2]. Therefore, the nanocrystals approximately behave as the 3-level system represented on Fig. 1(b). The absorption of a pump photon is followed by a picosecond-fast non-radiative relaxation to a long-lived (30 ns) single-exciton state: single-photon emission results from the quasi-exclusively radiative decay of this level.
The pump laser beam is modulated in amplitude using a digital micro-mirror array (Texas Instrument, DMD Discovery 4000) made of 1024 x 768 10.8µm-sized flat mirrors. The mirrors have two stable orientations, that we call "on/off", and can be addressed individually. In the "on" state, a mirror reflects light on the optical axis of the microscope. Light reflected by a mirror in the "off" state does not enter the optical system. The optics of our microscope are positioned in such a way that the micro-mirror plane is conjugated (in the sense of image formation) to the nanocrystal plane. This creates a one-to-one corre-

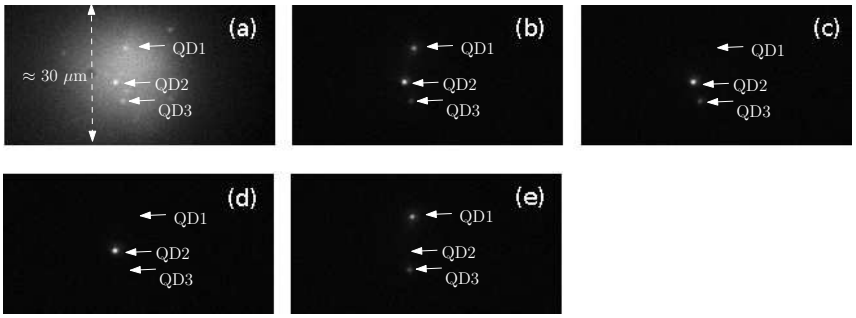

Figure 2: (a) To observe the entire field of view, all the micro-mirrors of the array are switched on. (b) Only mirrors addressing quantum dots QD1, QD2, and QD3 are switched on. (c) QD2 and QD3 are on, QD1 is off. (d) Only QD2 is on. (e) QD1 and QD3 are on, QD2 is off.

spondence between mirror coordinates on the micro-mirror array plane and nanocrystal positions in the sample plane. By switching the right mirrors in the "on" state we can select the nanocrystals that we want to illuminate. The microscope is confocal in the sense that no pump light reaches regions of the sample that are void of QDs.
The ability of this device to selectively excite an arbitrary number of QDs is demonstrated on Fig.2. In Fig. 2(a), all the micro-mirrors of the array are switched on. This allows us to observe the entire filed of view (about 30 μm) and map the position of the QDs on our samples to micro-mirror coordinates. In Fig. 2(b), all micro-mirrors have been switched off except those illuminating the three quantum dots of interest. The numerical aperture of our objective lens (0.4) and imperfections in the setup currently limit the resolution of the microscope to about 1.5 μm for illumination and to less than 1 micron for imaging. In Fig. 2(c)-(e), we show that we can arbitrary select those QDs that we want to address.

Applications of the scheme

Quantum cryptography is a natural application for any single-photon source since the very security of quantum private-key distribution relies on the uniqueness of quanta sent through the communication channel [5]. Our source has the unique feature that many emitters can be addressed simultaneously and that, the emitters being colloidal nanocrystals, the central wavelength of each emitters depends on its composition and size. Mixing

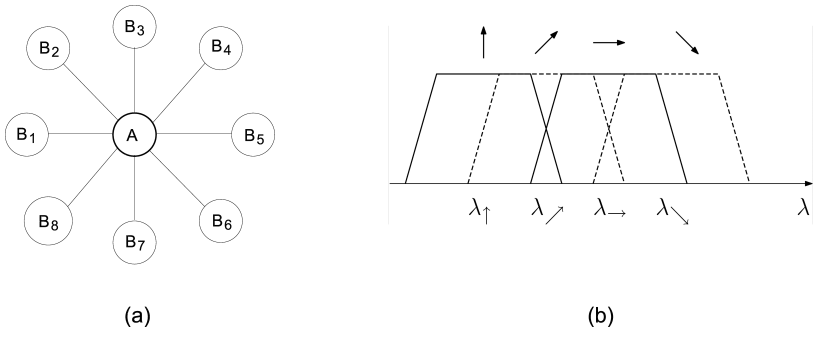

Figure 3: (a) A quantum cryptography network with a central server A and n nodes B_i ($i \in \{1, ..., n\}$). (b) Wavelength coding for quantum cryptography.

on the same sample nanocrystals emitting single-photons in n non-overlapping wavelength bands would transform our device into a wavelength multiplexed single-photon server for a star-shaped quantum cryptography network as the one represented on Fig. 3(a). In this network, photons with a given wavelength λ_i are uniquely associated to a definite $A - B_i$ channel using wavelength multiplexers.
A second way of using the wavelength tunability of the source for quantum cryptography consists in coding qubits in the wavelength itself. The BB84 protocol uses two pairs of mutually orthogonal states $(|\uparrow\rangle, |\rightarrow\rangle)$ and $(|\nearrow\rangle, |\searrow\rangle)$ with maximal overlap. As showed on Fig. 3(b), the same idea can be implemented using wavelength coding. Channels $\lambda_\uparrow$ and $\lambda_\rightarrow$ ($\lambda_\nearrow$ and $\lambda_\searrow$) are orthogonal and channel $\lambda_\nearrow$ ($\lambda_\rightarrow$) has a maximal overlap with both of them. However, the last channel $\lambda_\searrow$ ($\lambda_\uparrow$) only overlaps with channel $\lambda_\rightarrow$ ($\lambda_\nearrow$). The consequence of that difference and how to adapt BB84 have to be investigated.
Quantum imaging with incoherent photons is a third application of our apparatus. It takes advantage of the fact that single photons can be emitted on demand from definite and known sites. The basic idea behind quantum imaging is that the effective de Broglie wavelength of a N-photon quantum state is λ/N rather than λ, the wavelength of individual photons. To take advantage of that, one needs to control the position of the sources (our source allows that) and the detectors relatively to the object that one wants to image. This quantum optical technique permits to beat the standard diffraction limit and could become relevant to optical lithography [6].

Conclusion

We demonstrate a new device allowing to excite simultaneously individual nanocrystals with unprecedented flexibility and showed how this apparatus can be used for *scalable* quantum photonics applications to quantum information processing.

This work received the financial support from the Belgian F.R.S-FNRS under the FRFC grant 2.4.638.09F and from the Interuniversity Attraction Poles program of the Belgian Science Policy Office, under grant IAP P6-10 “photonics@be”. One of the authors (E.B.) also thanks the F.R.S-FNRS for personal support.

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