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Dots in rods as polarized single photon sources

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ABSTRACT

We report the evidence of a polarized single photon flux from a colloidal nanoparticle. We analyze, by time and polarization resolved spectroscopy measurements, the polarization behavior of a single CdSe/CdS core/shell dot in rod, achieving a polarization ratio at room temperature of ∼75% and a lifetime of the excited state of ∼11 ns.

1. Introduction

A new generation of integrated-high efficient single photon source (SPS) devices [1,2] was recently developed using epitaxial or colloidal quantum dots (QDs). Wet-chemical synthesized colloidal nanocrystals (NCs) are actually considered among the most promising candidates for these innovative devices, by virtue of their broad tunable emission range, low fabrication costs and high quantum efficiency at room temperature [3,4]. However, the applications of NCs to SPSs devices are limited by some specific features, such as blinking, spectral diffusion, long luminescence lifetime and unpolarized emission [5,6]. Nevertheless, recent studies demonstrated how these limitations could be overcome by properly engineering the shell structure [7] or the shape of the nanoclusters [8–11]. Among shell shapes proposed so far, dots-in-rods (DRs) nanocrystals [9], exhibit high absorption, intense light emission, a quantum yield estimated to be ∼75% (further improvable by adding multiple shells [12]), together with very interesting ensemble polarization properties [9], thus resulting in a very promising

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and efficient single photon source. In 2001 the first evidence of a polarized light emission from colloidal CdSe nanorods was analyzed by imaging techniques in Refs. [8,13] and several theoretical and experimental studies were carried out to explain the dipole moment origin [14,15] and the nature of the polarized emission, usually attributed to the dielectric confinements [16,17] and to the higher levels of valence band [8,11]. In 2005 Thomas et al. [18] studied core/shell CdSe/ZnS DRs at cryogenic temperature showing that these NCs are characterized by several spectral lines with different polarizations. Further studies also revealed the polarized nature of light emitted by elongated CdSe/CdS structures [10,11]. The role of the surface charges in core/shell DRs and NCs is currently under investigation [19,20] and various hypothesis were developed in order to understand the band-structure of CdSe/CdS nanoparticles [21,22], which seems to be shape dependent. To demonstrate non-classical light emission from a colloidal nanocrystal, experiments were performed in early years of 21st century [23–25]; the antibunching from one dimensional core/shell structures at a relatively high temperature ($\sim$220 K) was shown in 2008 by Tribu et al. [26] for epitaxial QDs in nanowires and by Knappenberger et al. [27] for colloidal CdSe/tetradecylphosphonic acid nanorods. However, to the best of our knowledge, at present no evidence of a flux of single, polarized photons on demand from a single DR is reported in literature.

In this work we focus on the polarization features of the antibunched photon flux from a single CdSe/CdS DR, showing that these colloidal particles are among the most promising candidates for low costs, room temperature polarized SPSs. Performing time and polarization resolved photoluminescence spectroscopy (TPRPL) and computing the exciton lifetime with two different techniques, we demonstrate the non-classical behavior and the polarization properties of light emitted by a single CdSe/CdS DR.

2. Methods

CdSe/CdS DRs were prepared by growing a CdS rod-like shell onto a spherical CdSe QD [9] and their dimensions (core diameter of 2.7 nm and shell length of 30 nm) were estimated by a transmission electron microscope (Fig. 1(a)). A low concentration solution of DRs was drop-casted on a glass coverslip and analyzed by a confocal microscopy system. The pulsed laser excitation (circularly polarized, picosecond pulse at a wavelength of 404 nm) was focalized on a single nanorod by a high numerical aperture ($NA = 0.95$) air-objective. The excitation power was chosen in order to avoid bi-exciton and Auger processes contribution as described in [20]. The polarization of the DR emission, analyzed by a half-wave plate and a polarizing cube, was collected by the same objective and sent into a standard high-sensitivity Hanbury–Brown and Twiss setup based on two avalanche photodiodes, shown in Fig. 1(b).

The signals from the photodiodes were sent to a data acquisition card (TimeHarp200) used in two different acquisition modes in order to obtain time resolved measurements and photon antibunching, used to identify a single DR and to characterize its non-classical emission. The photon antibunching was measured in the *start/stop* mode, in which the card records all the stop events in a time window of 4.7 μs, giving access to the coincidence photons histogram (proportional to the autocorrelation function of the emitted signal $g^2(\Delta t)$) with a time resolution of $\sim$1.1 ns. The second acquisition mode (hereafter referred as *collection mode*) records individual photon events with their arrival times and their delays from the synchronization pulse, and it was employed in order to obtain the decay curve with a resolution of $\sim$150 ps.

3. Results and discussion

The TPRPL time traces for detection polarization angles of $\theta = 0°$ and $\theta = 90°$ are shown in Fig. 2(a), (b), (c) for three different timescales ($\theta = 0°(90°)$ means that the detected polarization is parallel (orthogonal) to the direction of maximum intensity). As confirmed by the occurrence histogram (Fig. 2(d)), the dots in rods can be characterized by two emission states: ON, in which the DR is continuously emitting, and OFF, in which the NC is not emitting. Both, the ON and OFF state, are well visible in Fig. 2(d) for $\theta = 0°$, in which the OFF state is represented by the local maximum