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OPTO-ELECTRONICS REVIEW **18**(1), 1–9

DOI: 10.2478/s11772-009-0026-7

Recent advances on single photon sources based on single colloidal nanocrystals

INVITED PAPER

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Single colloidal quantum dots (QDs) are increasingly exploited as triggered sources of single photons. This review reports on recent results on single photon sources (SPS) based on colloidal quantum dots, whose size, shape and optical properties can be finely tuned by wet chemistry approach. First, we address the optical properties of different colloidal nanocrystals, such as dots, rods and dot in rods and their use as single photon sources will be discussed. Then, we describe different techniques for isolation and positioning single QDs, a major issue for fabrication of single photon sources, and various approaches for the embedding single nanocrystals inside microcavities. The insertion of single colloidal QDs in quantum confined optical systems allows one to improve their overall optical properties and performances in terms of efficiency, directionality, life time, and polarization control. Finally, electrical pumping of colloidal nanocrystals light emitting devices and of NC-based single photon sources is reviewed.

Keywords: quantum dots, colloidal nanocrystals, emission efficiency, microcavity optical modes

1. Introduction

Single photon sources (SPSs) are a breakthrough in quantum information due to their possible exploitation in fields such as quantum communication, quantum computation, quantum lithography, and quantum cryptography [1–4]. In particular, quantum cryptography theorizes the secure distribution of a secret key between two partners through quantum photonic states. Also referred to as quantum key distribution (QKD) [5,6], this method is intrinsically secure since a quantum state cannot be read by an eavesdropper without being modified [7]. Since its initial proposal in 1984 [8] and its first experimental demonstration in 1992 [9], QKD has been implemented in many ways [10], exploiting different techniques for the generation of single photons. True single photon sources [11,12] have been demonstrated to outperform key distribution based on attenuated classical laser pulses [13], where multiple photon emission is hard to avoid [14]. Implementations of single photon sources based on individual molecules [15], nitrogen vacancy centres [16] or dopant atoms [17] are rather inefficient due to low emission rates and rapid saturation.

An ideal quantum light source should generate one single photon per excitation pulse, which can be accomplished by using the emission of a single quantum system, such as a single quantum dot [18]. By virtue of their unique electronic structure and quantum confinement, single quantum-dot emitters reliably generate single photons on demand when excited by short optical or electrical pulses. Since 1982, when the concept of quantum dots (QDs) was first proposed by Arakawa and Sakaki [19], their peculiar properties have been applied to a number of photonic devices, and impressive results in the synthesis and application of these powerful nanostructures have been demonstrated. Epitaxial QDs, obtained by the Stranski-Krastanov self-organized growth mode [20], have demonstrated their potential as light sources for ultrafast semiconductor lasers and optical amplifiers (SOAs) for optical communications, showing new functionalities and astonishing performances such as high gain and efficiency, ultra-low threshold current densities and temperature insensitivity [21–23]. A different approach for the fabrication of quantum dots is based on wet chemistry for the synthesis of colloidal QDs. These colloidal nanocrystals (NCs) are good candidates for QDs photonic applications in different fields such as health, energy, environment and aerospace due to their low fabrication costs, high quantum efficiency at room temperature, high versatility in the chemical synthesis, and broad tunable emission range [24]. Both epitaxy and wet chemistry have demonstrated to be effective for the fabrication of single QDs embedded in triggered non-classical sources of single photons. Epitaxial QDs exhibit single photon emission only

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at cryogenic temperature [25], whereas single colloidal NCs based on II-VI compounds exhibit photon antibunching at room temperature and above [26,27], by virtue of their peculiar electronic properties. In order to tailor and enhance the emission of SPSs, single quantum emitters should be embedded inside micro and nanocavities. The insertion of QDs in quantum confined optical systems, such as high quality factor resonators, allows us to improve the overall performances in terms of efficiency and directionality, together with a better control on the polarization characteristics of the emitted photons. Among the above discussed species for single photon generation, self-assembled epitaxial and colloidal QDs can be straightforwardly incorporated into cavities by using standard micro and nanotechnological processing techniques.

In this work recent advances on single photon sources based on single colloidal quantum dots are reviewed. We first recall basic concepts related to the optical properties of colloidal nanocrystals, with special emphasis on their photon antibunching, blinking and spectral diffusion phenomena. We then discuss the technological approaches for isolating and positioning single colloidal quantum dots, an essential requirement for practical realization of single nanocrystal SPSs. The third section reports on the approaches to significantly enhance the emission efficiency of SPSs by coupling the emission of single nanocrystals to microcavity optical modes. The successful achievement of this challenging technological target leads to devices with enhanced performances in terms of extraction efficiency, low divergence and improved emission efficiency in both weak and strong coupling regimes. Finally, possible approaches for the electrical pumping of single colloidal nanocrystal SPS devices are discussed. Electrically driven triggered sources of single photons are highly attractive since they would bring compact SPSs to practical application, without requiring expensive and sizeable light excitation sources.

2. Single colloidal quantum dots as sources of single photons

Semiconductor colloidal nanocrystals synthesized by wet chemical approaches as sources of single photons have been extensively studied in the last years because of their low fabrication costs, high quantum efficiency and photostability also at room temperature [28]. Broad tunability of their emission wavelength from the visible to the infrared has been readily achieved by virtue of the high versatility in the chemical synthesis, which allows for excellent control over NCs size, shape and composition [29–32].

Typical nanocrystals are nanometer-sized spherical core--shell structures. The role of the shell is to engineer the band structure of the nanostructure and to passivate the core surface, thus reducing surface defects, such as dangling bonds, which dramatically affect their efficiency and photostability. Due to their absorption continuum at energies above the exciton transition they can be excited by a variety of light sources. Recombination from higher excited states, multi--exciton and charged excitons in careful experimental conditions is typically negligible because of strong Auger processes [33], and the emission of single photons can be made highly probable only from single exciton recombination.

In spite of their excellent quantum efficiency and photostability, colloidal nanocrystals, when observed at the single particle level, exhibit fluorescence blinking and spectral diffusion. The first effect consists in the random switching of fluorescence between bright ("on") and dark ("off") states [34]. Such a fluctuation of the luminescence over time has been attributed to charge transfer or charge escape from the dot, which leads to a free charge in the NC, preventing any possible radiative recombination for times in the order of milliseconds. A strategy to control and completely prevent blinking has been recently proposed by Mahler and coworkers [35], who show that a thicker shell in CdSe-CdS NCs allows one to control the blinking behaviour, suggesting that well-designed shells are the key parameters for obtaining non-blinking QDs. Non blinking behaviour has been also obtained in ternary CdZnSe/ZnSe NCs, designed as a radial graded alloy of CdZnSe into ZnSe [36].

Spectral diffusion consists of random spectral jumps ascribed to charge movements or, more generally, to instability in the nanoscale environment of the nanocrystals, which cause strong fluctuations of local electric fields [37,38]. The resulting linewidth of single nanocrystals is broadened by the time integration of NCs emission at slightly shifted wavelengths. High temporal resolution is necessary to observe, in a single quantum dot, the evolution of spectral diffusion [39]. In CdSe/ZnS single colloidal nanocrystals, a spectral diffusion of 4 μeV over a time scale of 200 μs was observed for an emission peak having a linewidth as narrow as 6.5 μeV [40].

The good emission efficiency at room temperature, good stability and advances in high spatial resolution spectroscopic techniques has allowed in the last years a deeper understanding of the peculiar optical properties of QDs, by probing NCs at single particle level [34,37].

Evidence of efficient single photon emission at room temperature corroborates the potential of these nanostructures as active medium of single photon sources. A Hanbury Brown and Twiss setup is normally used to confirm the non-classical nature of the single QD emission through the second-order correlation function,

$$g^{(2)}(\tau) = \frac{< I(t+\tau)I(t) >}{< I(t+\tau) >< I(t) >}$$

i.e., an intensity-intensity correlation function, having information on both photon statistics and dynamics of the light generated from the nanocrystals. Figure 1(a) shows the scheme of a Hanbury Brown and Twiss setup. Single photons impinging on a beam splitter are split towards two fast avalanche photodiodes (APDs), which allow to measure directly $g^{(2)}(\tau)$, by detecting the delay between the arrival events. The absence of coincident detection events on the two detectors is the fingerprint of a one-by-one photon

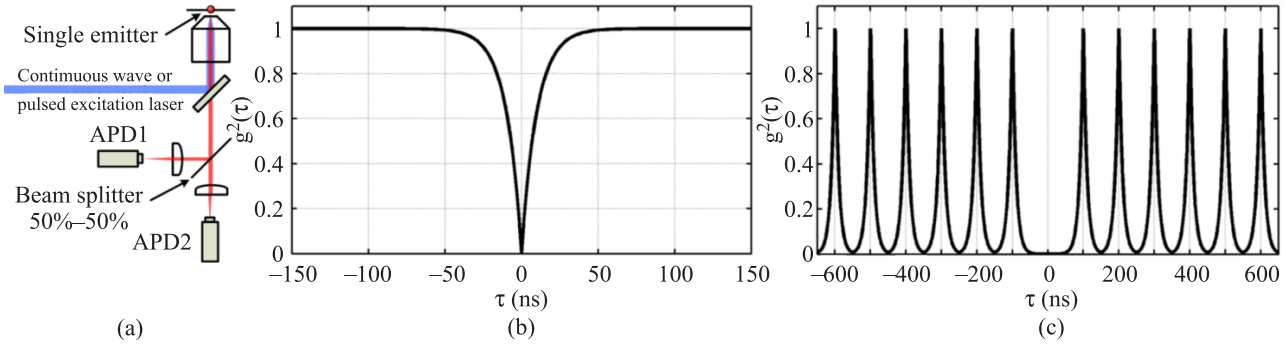

Fig. 1. Schematic representation of the Hanbury Brown and Twiss setup (a), typical behaviour of the second order autocorrelation function for continuous (b) and pulsed excitation (c).

emission, i.e., photon antibunching. In most single photon sources at room temperature, this behaviour is manifested by a dip around zero time delay ($\tau = 0$) in the second-order correlation function [Fig. 1(b)]. When using a pulsed excitation, the second-order correlation function consists of an array of short peaks with the period of the pumping sources and the evidence of single photon emission is the absence of the peak at zero time delay [Fig. 1(c)].

Most of the photon antibunching reports in the literature are based on II-VI NCs constituted by a CdSe spherical core embedded in either ZnS or CdS shells. In CdSe/CdS NCs, by virtue of a low lattice mismatch, the influence of defects on the radiative recombination process is reduced, as well as the influence of surface traps on single excitons, which usually tend to increase the average radiative lifetime of the system. The full control on the growth parameters in the wet chemical synthesis process was exploited to synthesize different NCs such as rods, dot in rods, tetrapods and also dimers [29,31,32,41,42]. Most of these nanocrystals demonstrated to be effective sources of single photons. In particular nanorods, i.e., elongated NCs, and CdSe/CdS dot-in--rods (DR), obtained by surrounding a spherical CdSe core with an elongated CdS shell [31,43] appear to be very promising quantum emitters, by virtue of their relatively short lifetime [44,45] and electrical dipole-moment oriented along the rod axis [29,43] which leads to a higher degree of linear polarization [29,43,45].

This feature, invaluable for a SPS device, allows a deterministic photon polarization to be achieved, as required in BB84 and B92 cryptographic algorithms [8,46], without recurring to polarizers to encode information. In Ref. 47, it is shown that the polarization control of single photons can be achieved by using dot-in-rods rotated at different angles, envisioning a strategy to develop polarization controlled, low cost and highly efficient room temperature single photon sources.

3. Localization of single quantum dots

Both epitaxial self-organized quantum dot and colloidal nanocrystals have been used as active media of single photon sources. Both being based on self organization, one of the most challenging tasks is the control over the QD position, in order to align the nanoemitter to a microcavity or to fabricate arrays of SPS devices in specific locations.

The typical way to realize a single photon source based on self organized QDs is to inspect the sample, purposely fabricated with low-density of quantum dots ($<10^9$ cm^{-2}), and after having localized an isolated area with a single nanostructure inside, to build a device using very high resolution fabrication techniques [48–50]. The nucleation sites, and therefore the surface density of epitaxial QDs, are usually controlled by carefully tuning the Stranski Krastanov growth parameters. Colloidal NCs are typically diluted to nanomolar concentrations and dropcasted on glass cover slides, obtaining, after evaporation of solvents, isolated nanocrystals with an average distance among them in the order of microns.

These approaches, though well-suited for the characterization through confocal microscopy of photon antibunching behaviour in single nanostructures, are not appropriate for the realization of controlled arrays of SPS to be embedded in QKD commercial systems.

In order to localize epitaxial QDs, fabrication of site--controlled pre-patterned recesses in the substrate and regrowth has been successfully exploited [51] to embed single QDs inside micropillar vertical cavities [52] and 2D photonic crystal membranes [53], but poor control over emission wavelength and lack of single photon emission up to room temperature prevent their practical use.

Recently, a new approach has made possible the pinpointing of single colloidal quantum dots by direct electron beam lithography. The controlled localization of ordered arrays of single colloidal nanocrystals was demonstrated in Ref. 54 by dispersing a specific concentration of nanocrystals inside a negative high-resolution electron-beam resist after precipitation and re-dilution in methyl-isobutyl-ketone (MIBK) solvent. Ensembles of nanocrystals embedded in electronic resists can be easily patterned by means of traditional lithographic processes, since the presence of semiconductor clusters in the matrix does not significantly affect the sensitivity of the polymeric host and, at the same time, the emission properties of the nanocrystals are not influenced by the interaction with the electron beam [55]. To obtain on average one nanocrystal inside a nanosized pillar of the NC/resist blend, direct e-beam patterning is employed

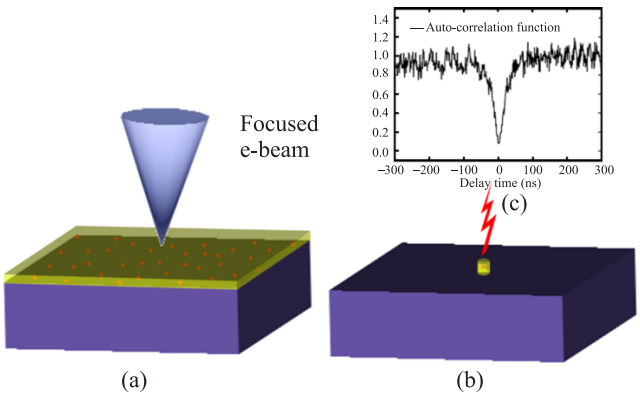

Fig. 2. Schematic representation of the localization of single colloidal quantum dots by direct electron beam lithography of a resist/NCs blend (a). By a careful control of molar density of the NCs dispersed in the resist and pillar volume it is possible to isolate single QDs (b). Autocorrelation function of an isolated CdSe/ZnS QD of coincidence counts of fluorescence from a single QD in microcavity (c).

[Figs. 2(a) and 2(b)]. Through a careful control of molar density of the NCs dispersed in the resist and pillar volume, i.e., blend layer thickness and pillar diameter, it is possible to obtain a very high probability of having localized single colloidal NCs, whose photon antibunching behaviour was confirmed through a confocal microscope and Hanbury-Brown and Twiss setup [Fig. 2(c)].

It is worth mentioning a few more methods for trapping single colloidal nanocrystals which rely on electrostatic trapping in nanogaps [56] or by electrostatic biological self-aligned placement [57]. In the latter case, the self-aligned positioning of a nanocrystal in the cage-shaped negatively charged protein is followed by the trapping of the protein on specific positively charged ultra small positions. Electrostatic trapping is very attractive remarkably for nanocrystals exhibiting a high dipole moment, such as asymmetric dot-in-rods.

4. Coupling of colloidal QDs to microcavities

The presence of a cavity strongly affects the density of available photon states (PDOS) of the environment. When an emitter is placed inside a cavity, many properties of its light emission can be strongly modified. Spontaneous emission rate and radiative decay can be engineered and enhanced, radiative pattern and divergence angles can be modified to improve collection efficiency and optical matching with optical fibers, spectral emission can be narrowed or filtered and the emission polarization can be controlled.

Micro and nanocavities embedding nanoemitters are the subject of intense studies for the realization of ultra-low threshold microlasers, or cavity quantum electrodynamics (CQED). CQED systems allow us fundamental studies of coherent interactions of confined electromagnetic fields (cavity photons) and microscopic dipoles (single nanoemitters) [58–61].

The task of aligning a single nanoemitter to a cavity is very challenging since its single exciton transition must be coupled to a resonant mode of a high quality factor (Q-factor) microcavity. When this is achieved, the efficiency and emission properties of a single colloidal NC are significantly improved. The coupling between the nanoemitter and the cavity can be either strong or weak depending on the coupling parameter,

$$g = \frac{\mu_{dm}}{\hbar}\sqrt{\frac{\hbar\omega}{2\varepsilon_0 V}}$$

where V is the cavity mode volume, ε_0 is the vacuum dielectric constant, and μ_{dm} is the dipole moment of the transition. Asymmetric NCs, such as nanorods and dot-in-rods, are particularly suitable for the achievement of strong coupling regime since they show a strong dipole moment and increased dipole oscillator strength [62].

Weak coupling enhances the spontaneous emission, whose rate is given by the Purcell factor

$$F_p = \frac{3}{4\pi^2}\frac{Q}{V}\left(\frac{\lambda}{n}\right)^3.$$

When a nanocrystal is inserted into the microcavity and its eigenfrequency ω_{QD} is resonant with the high-Q cavity mode ω_{cav} and $g^2 > (\gamma_{cav} - \gamma_0)^2/16$, being γ_0 and γ_{cav}, the field decay rate in free space and inside the cavity, respectively, the confined photonic and electronic states interact by coherently exchanging energy. This leads to a new strongly coupled quantum-mechanical system which evolves with a coupling strength defined by the vacuum Rabi splitting [63–68].

In both strong and weak coupling, the target is to reduce the modal volume of cavity and to carefully tune its Q-factor as it is typically possible in photonic crystal nanocavities. When F_p is greater than 1, the emitter radiates faster in the cavity than in free space and the emission is enhanced only as long as the system remains in the weak coupling regime.

The coupling is maximized when the emitter is placed in the maximum of the electric field intensity; however, this is hard to be fulfilled for point-like emitters, since it requires a high accuracy in nanocrystals positioning inside an ultra-small volume.

Doping of vertical microcavities with an ensemble of NCs has been demonstrated by different techniques, by embedding the nanocrystals in two distributed Bragg reflectors (DBRs) [69] and by focused ion beam (FIB) post-milling to obtain micropillars [70] or by imprint lithography [71].

However, very few papers in the literature report on single colloidal nanocrystals embedded in microcavities and efficiently emitting triggered single photons at room temperature.

Coupling of a single colloidal quantum dot with photonic cavities has been achieved by exploiting whispering gallery modes generated on the surface of glass and polymer microspheres [72]. Artemyev *et al.* report on the coupling of

a single CdSe/ZnS NCs chemically bound to the surface of a single glass microsphere, reporting the Purcell factor F_p ~10. The same group reported on a strongly coupled CQED system consisting of an anisotropically-shaped CdSe nanocrystal coupled to a single photon mode of a polymer microsphere, showing a vacuum Rabi splitting between 30 and 45 μeV in a microsphere cavity, slightly deformed to remove mode degeneracy [61].

Although glass spherical cavities localize very high quality factor modes, their use as SPS is hindered due to their relatively large mode volume and their poorly collimated photon emission.

In this respect, photonic-crystal structures [73,74] are promising candidates for the trapping of light in ultrasmall volumes with high Q-factor [75–77]. Two- and three-dimensional photonic crystal microcavity architectures were also exploited by Wu *et al.* [78], who coupled PbSe nanocrystals to a silicon PC membrane, and by Lodhal *et al.* [79] who used a titania inverse opal as host photonic crystal lattice.

Photonic crystal cavities, providing a well collimated emission, are more suited for the realization of SPS, but the crucial step for their fabrication, i.e., the positioning of single colloidal nanocrystals in the resonant path of the cavity modes, is difficult to be achieved.

While the demonstration at low temperature of single photon emission from a single epitaxial quantum dot coupled to cavity modes has been reported both in micro/nanopillars vertical cavity resonators [80] and in two-dimensional photonic crystal resonators [48], the localization of single colloidal NCs inside a microcavity resulted to be more complex, due to the more difficult handling of these nanoemitters. Recently, the experimental observation of fluorescence from single colloidal nanocrystals in microcavities by both doping of a chiral liquid-crystal microcavity and also by a microcavity fabricated by spin coating of several polymeric layers and dispersing a nanomolar concentration of NC in the waveguiding layer was reported [81]. A further step toward the practical realization of single NC based SPS was reported in Ref. 82. Qualtieri *et al.* showed for the first time the occurrence of photon antibunching at room temperature from single colloidal CdSe/ZnS nanocrystals inserted in an ordered array of vertical microcavities. The approach relies on the single NC localization technique obtained by direct electron beam lithography on top of a SiO_2/TiO_2 Bragg mirror [82]. Subsequently, the array of localized single quantum emitters was clad by a second DBR consisting of four alternating quarter-wavelength thick layers of TiO_2/SiO_2.

Figure 3 shows the schematic of the fabricated microcavity and the antibunching demonstration [Fig. 3(c)] from single nanocrystals in microcavity at room temperature.

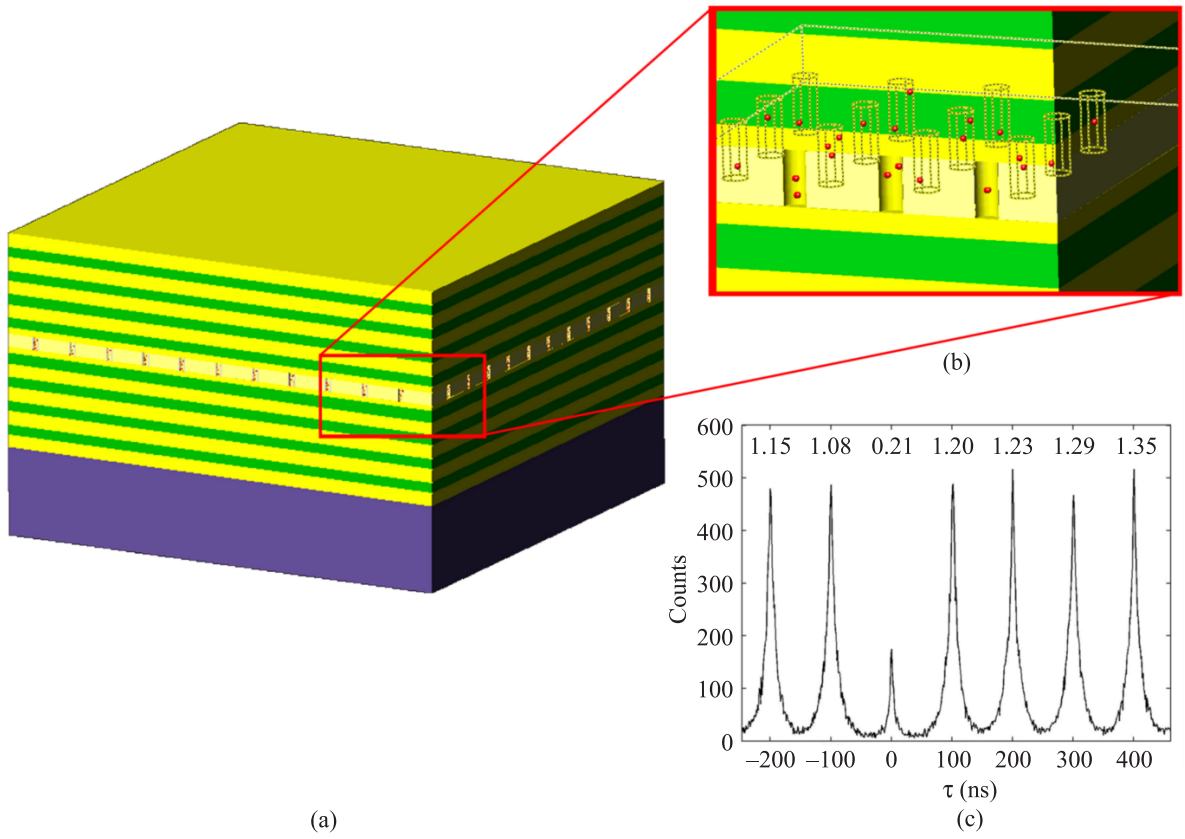

Fig. 3. Schematic of single colloidal quantum dots microcavity array obtained by direct electron beam lithography and SiO_2/TiO_2 DBR deposition (a,b). Histogram of coincidence counts of fluorescence from a single QD in microcavity (reproduced with permission from Ref. 82) (c).

5. Electrical pumping of single photon sources based on colloidal nanocrystals

Colloidal NCs have demonstrated their potential in many applications due to their aforementioned peculiar properties. Most experimental results on photon antibunching are based on optical pumping of NCs, but recently different approaches for electrical pumping/driving of colloidal nanocrystals embedded in light emitting devices or solar cells are being pursued. Injection of carriers in colloidal NCs is not as straightforward as for epitaxial QDs, where these nanostructures can be directly embedded in p-n junctions.

Direct injection of charge carriers in colloidal nanocrystals is typically limited due to the presence of an insulating organic capping layer on the nanocrystals. An indirect approach for the electrical pumping is based on the insertion of NCs inside a device which transfers either radiatively or non-radiatively the accumulated electrical energy to colloidal nanocrystals. The latter mechanism, called Förster resonant energy transfer (FRET), is based on a donor-acceptor non-radiative coupling and is governed by the spectral overlap between the emission spectrum of the donor species with the absorption spectrum of an acceptor and by the distance between them. In this process of non-radiative transfer, described for the first time by Theodor Förster in 1948 [83,84], the excess energy of the donor is transferred to the acceptor by a dipole-dipole interaction in a purely non-radiative process whose rate is given by the formula

$$R_{ET} = \frac{9c^4\kappa^2}{8\pi n^4 \tau_r}\frac{1}{d^6}\int_0^\infty \frac{f_D(\omega)\alpha_A}{\omega^4}d\omega\,, \qquad (1)$$

where c is the speed of light, n is the refractive index of the medium, κ^2 is the parameter given by the mutual orientation of the donor to acceptor dipoles (for isotropic orientation equal to 2/3), and τ_r is the radiative lifetime of the donor and the integral is the "overlap integral" which takes into account the overlap between the donor emission spectrum $f_D(\omega)$ and the absorption spectrum of the acceptor $\alpha_A(\omega)$. The most significant factor is the distance d between the acceptor and the donor, the FRET rate being proportional to the inverse of the sixth power of d.

For very short donor-acceptor distances, shorter than the extent of dipoles, higher order effects prevail, but their discussion is beyond the scope of this review.

FRET has been exploited in the last years in many demonstrations of LEDs based on hybrid technologies and embedding colloidal nanocrystals.

An approach to nanocrystal-based, electrically pumped light emitting devices exploits hybrid organic/inorganic structures, in which only a small fraction of the charge carriers is directly delivered to nanocrystals through the organic network [85,86]. Coe *et al.* demonstrated efficient electrical injection in a colloidal quantum dot LED embedding a single monolayer of QDs between two organic thin films [85]. The organic layers transport charge carriers to the vicinity of the QD monolayer. Exciton generation on QDs occurs via two parallel processes, direct charge injection and non-radiative energy transfer from excitons close to the single QD monolayer, and formed in organic molecules in grain boundaries, interstitial spaces, and voids. These excitons then undergo FRET energy transfer to the lower-energy QD sites, where they recombine radiatively. Based on the same approach, the only evidence of electroluminescence from a single colloidal quantum dot is reported in Ref. 87, where no evidence of photon antibunching is reported, however.

The performance of these hybrid devices is limited by the low carrier mobility in both the organic and nanocrystal components, and by the poor stability of the organic molecules. Moreover, single nanocrystal single photon sources, triggered by electrical pulses, are still to be demonstrated.

Most of the reports on FRET in the past years have taken into account an ensemble of NC as acceptors. When single nanocrystals are used one has to deal with the reduced transition width, which becomes narrower and narrower as the temperature is decreased. This behaviour, together with the possibility of shifting the transition in asymmetric rod-like NCs through the quantum confined Stark effect (QCSE) [88], has been exploited in Ref. 89 to switch on and off the energy transfer between a dye molecule and an asymmetric dot in rod NC [90].

Finally, it is worth mentioning the radiative energy transfer, which is a technique used in traditional colour conversion schemes based on phosphors. The emission of a photon from a semiconductor standard LED is followed by absorption and re-emission by an optically active material, which is typically a phosphor but can also be replaced

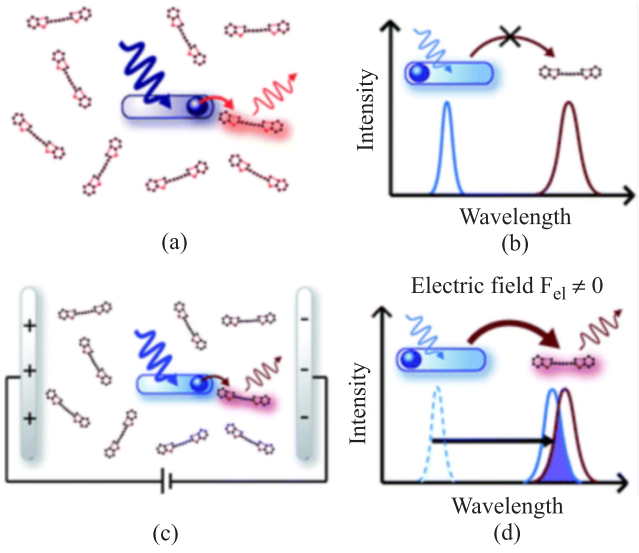

Fig. 4. Electrical control of energy transfer in a single FRET couple consisting of a dye molecule and an asymmetric dot in rod NC (a). Each NC is, in principle, adjacent to one acceptor molecule (b). At low temperatures, however, the electronic transitions of donor and acceptor are too narrow to enable a sufficient spectral overlap, required for efficient FRET (c). Application of an electric field to the NC-dye mixture can facilitate FRET by Stark-shifting the emission of the NC into resonance with the absorption of an adjacent dye molecule (reproduced with permission from Ref. 90) (d).

by nanocrystals. This approach has been already exploited for the demonstration of white light LEDs based on colloidal NCs on nitride-based quantum wells LEDs [91]. The quantum wells can be pumped electrically in the same way a common LED is pumped but the efficiency of the energy transfer is very poor, typically several orders of magnitude lower than non-radiative FRET transfer [92].

6. Conclusions

This review has discussed recent achievements and technological approaches for the realization of single photon sources based on single colloidal nanocrystals, a building block for future quantum information. Despite the body of work done so far, many issues still need to be faced and some of the technological problems to be solved. Manipulation, embedding and positioning inside the microcavities and efficient electrical pumping of single colloidal nanocrystals are technological subjects discussed in this paper, which still need to find optimal solution. However, the impressive results and astonishing performances reported in the literature in the last years make triggered sources of single photons closer and closer to their practical application in quantum communication and quantum cryptography.

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