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Ultrafast Semiconductor Quantum Optics

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1. Introduction

Since the "photon" was introduced by Albert Einstein in 1905, a large number of quantum optical experiments has been successfully performed with these quanta of light (Zeilinger et al., 2005). Also the interaction of single photons with atomic or molecular systems has been studied in great detail (Weiner & Ho, 2003). However, from an applications point of view, devices operating under high vacuum conditions are difficult to handle. Therefore, highly integrated quantum optical systems based on the solid-state are worthwhile studying. In this review we are focusing on semiconductor quantum dots.

2. Semiconductor quantum dots

Semiconductor quantum dots are sometimes also called "artificial atoms" due to their discrete energy states (Weisbuch & Vinter, 1991; Bányai & Koch, 1993; Bimberg et al., 1999; Bratschitsch & Leitenstorfer, 2006). If nanocrystals made of semiconductors become so small that the de Broglie wavelength of an electron is comparable to the crystal dimensions, a "quantum dot" is created. The electron is confined in the narrow potential well and resembles a particle in a box with discrete energy levels. The electronic energy states of the quantum dot may be populated with electrons and holes. If the energetically lowest lying electron and hole form a bound complex – similar to an electron and proton in a hydrogen atom - a so called "exciton" is formed. If electron and hole recombine, energy in form of a photon is released, i.e. a semiconductor quantum dot constitutes a nanoscopic light emitter. At the beginning of the 21st century it has been shown that a quantum dot is a single photon source (Michler et al., 2000; Santori et al., 2002). Recently, it has been demonstrated that photons emitted by a single quantum dot may be entangled (Akopian et al., 2006; Stevenson et al., 2006). These two experimental findings are basic building blocks for quantum optical experiments with semiconductor quantum dots. Up to now, most quantum optical experiments have been performed in the steady state with narrowband lasers (Atature et al., 2006; Xu et al., 2007; Kroner et al., 2008).

2.1 Ultrafast quantum optics with a single semiconductor quantum dot

Semiconductor quantum dots are also promising systems for robust and scalable quantum information processing (Li et al., 2003; Petta et al., 2005). Ultrafast sequences of coherent quantum operations may be envisioned with femtosecond light pulses, if the involved quantum states are separated by at least tens of meV. Therefore, small quantum dots with

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high confinement potentials are favorable. Due to their large Coulomb correlation energies, CdSe quantum dots are ideal candidates.

We have performed two-color femtosecond pump-probe spectroscopy on a single self-assembled CdSe/ZnSe quantum dot (Sotier et al., 2009). The transient quantum dynamics was probed with resonant excitation and detection (Fig. 1).

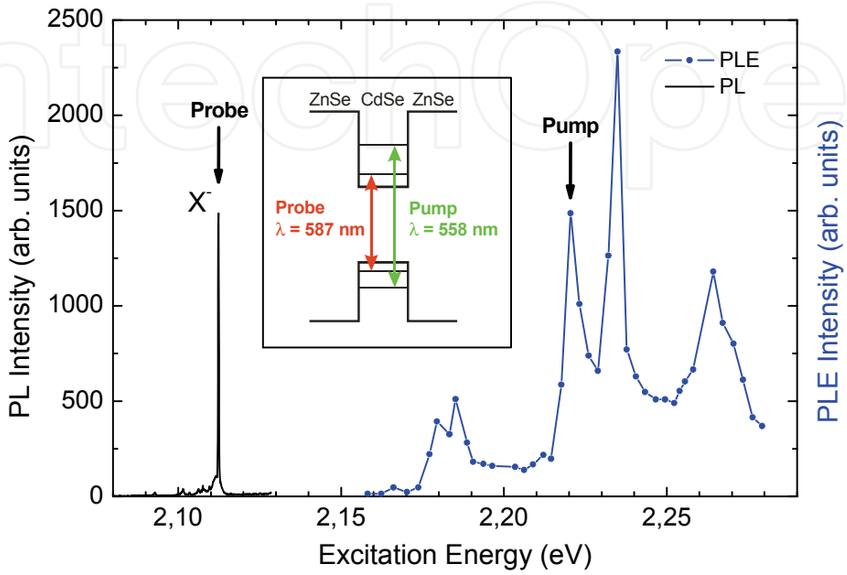


Fig. 1. Photoluminescence (PL) and photoluminescence excitation spectrum (PLE) of a single CdSe/ZnSe quantum dot, Inset: Schematic energy level structure of the quantum dot with indicated pump and probe center wavelength.

We were able to resonantly excite, manipulate and read out a single semiconductor quantum dot with femtosecond time resolution. A two-color Er:fiber laser with excellent noise performance (Tauser et al., 2003; Moutzouris et al., 2006; Moutzouris et al., 2007) is key to these first resonant pump-probe measurements on a single-electron system. The quantum dot was resonantly pumped into the p-shell with laser pulses of a duration of 750 fs. The transient transmission at the charged exciton ("trion") transition was probed by a time-delayed 190 fs pulse (Fig. 2). At a negative delay of -2 ps small oscillatory signatures around the fundamental exciton resonance are discernible, which are related to the perturbed free induction decay (Fig. 2). At +2 ps a bleaching of the exciton transition is observed due to Coulomb renormalization (Fig. 3). In the following 20 ps, the positive transmission signal increases by a factor of 1.5 and reaches a constant plateau that slowly decays with the radiative lifetime of the trion of 480 ps. As the hot exciton relaxes to its ground state (Fig. 3), the original transition is restored, but now under inversion conditions. A time delayed probe pulse causes stimulated emission, corresponding to an increase in differential transmission. The thermalization rate is governed by the electron spin (Fig. 3): the 20-ps onset of stimulated emission corresponds to the relaxation of a singlet electron which is generated by 50% of the pump pulses.

Operating the single semiconductor quantum dot in the nonlinear regime, the ability to change the number of quanta in a femtosecond light pulse by exactly ±1 is demonstrated (Sotier et al., 2009).

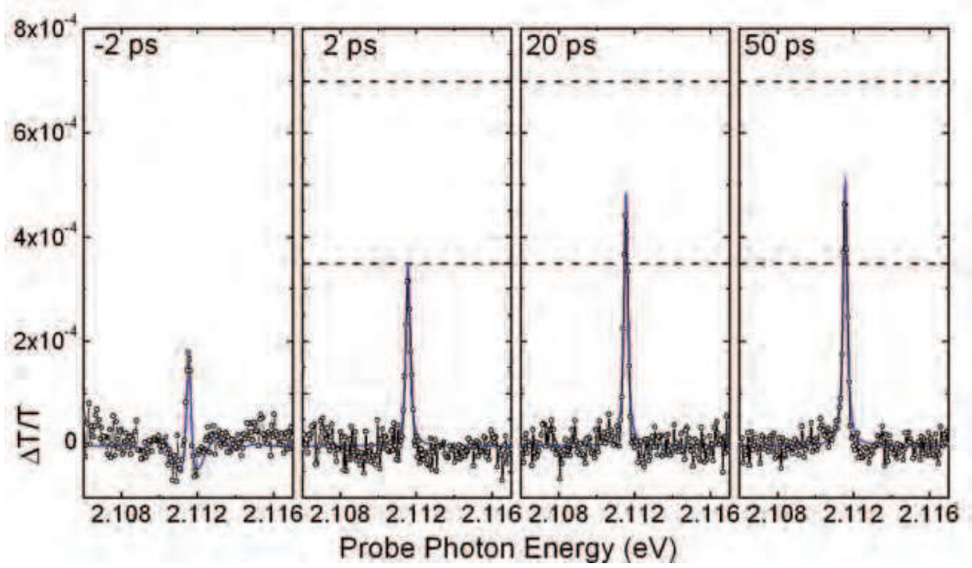


Fig. 2. Spectrally resolved transmission change through a single CdSe/ZnSe quantum dot for different pump-probe time delays t_D. The experimental data are represented by the hollow circles, while fits based on the optical Bloch equations for a two-level system are given by the blue lines. At negative time delays the perturbed free induction decay is visible. At positive time delay ultrafast Coulomb renormalization and spin-dependent electron relaxation is observed.

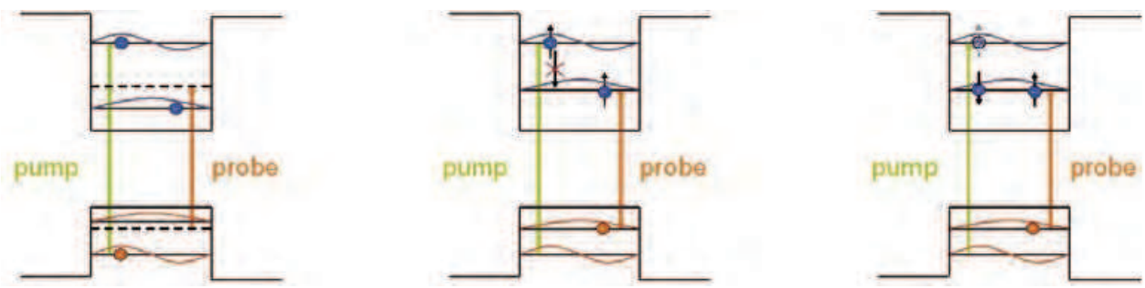


Fig. 3. Schematic drawing of the electronic configurations relevant for the differential transmission signals at positive time delays.

3. Nanophotonic elements for increased light-matter coupling

The experiments presented so far have been performed with at least thousands of photons. To reach the limit of only a single photon manipulating a single electron, the coupling of light with a wavelength of a few hundred nanometers into the object of nanometer dimensions, such as a semiconductor quantum dot, a color center in diamond, or a molecule has to be optimized. We follow two main routes to reach this goal: optical microcavities and metallic nanoantennas.

3.1 Dielectric optical pillar microcavities

We succeeded in embedding colloidal CdSe/ZnSe quantum dots in a planar and pillar microresonator operating in the visible regime (Kahl et al., 2007). A planar dielectric cavity

is formed by two Bragg mirrors, each consisting of sputtered pairs of alternating TiO_2 and SiO_2 layers. The semiconductor nanocrystals are embedded in the central cavity layer in SiO_2 surroundings via a spin-on glass technique (Fig. 4).

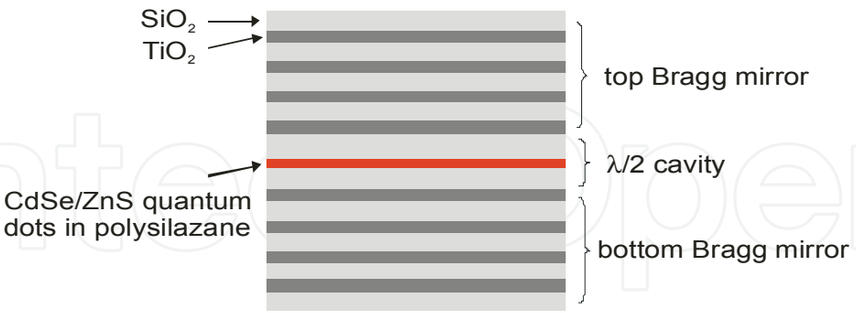


Fig. 4. Schematic drawing of a planar cavity with embedded colloidal semiconductor quantum dots.

Subsequently, micropillars with diameters in the range of 5 µm down to 600 nm are milled out of the planar resonator via a focused ion beam (Fig. 5).

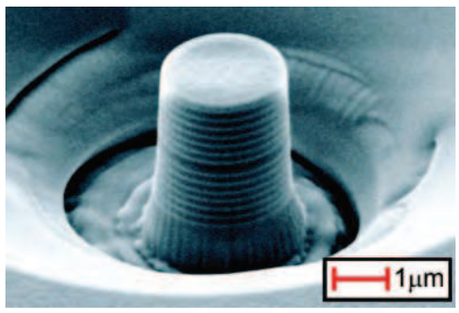


Fig. 5. Scanning electron microscopy (SEM) image of a dielectric micropillar with embedded colloidal CdSe/ZnS quantum dots.

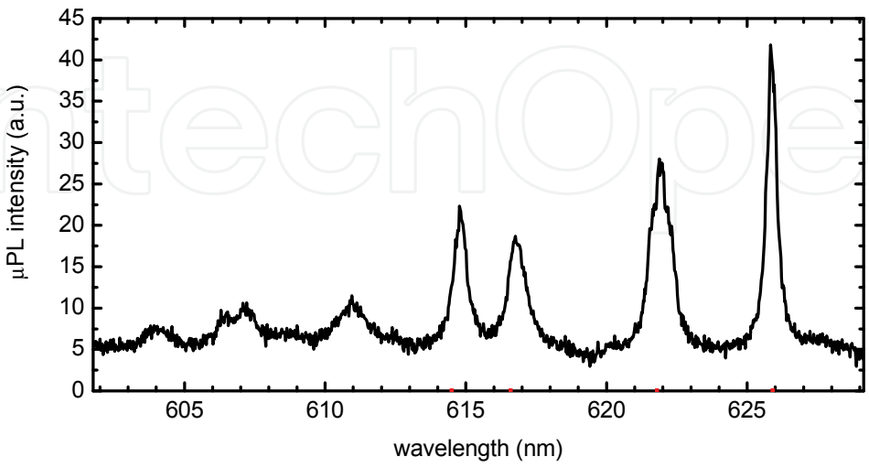


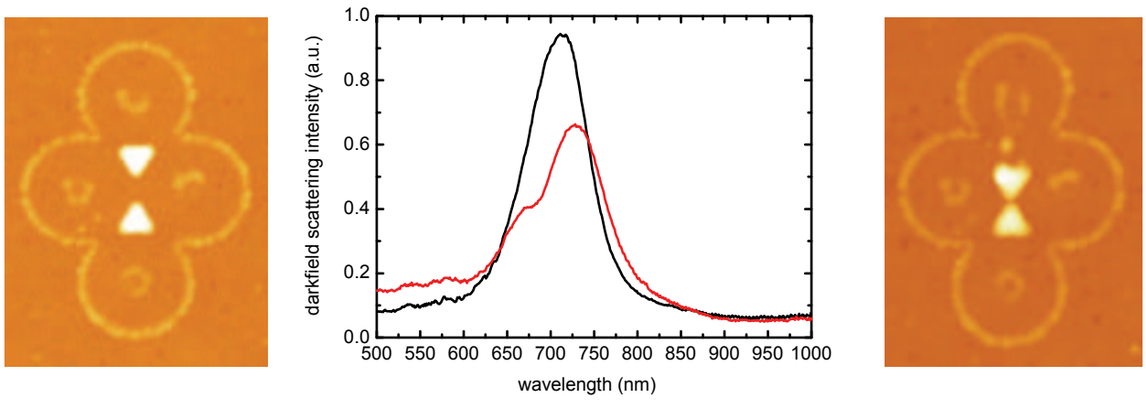
Fig. 6. Photoluminescence emission from a pillar resonator of 3 µm diameter with embedded colloidal CdSe/ZnS quantum dots, showing distinct microcavity modes.

A broadband light transmission measurement through a single micropost cavity shows the lowest cavity mode blueshifting with decreased pillar diameter. This effect directly demonstrates three-dimensional light confinement. Higher cavity modes are identified in the photoluminescence emission from an ensemble of quantum dots (Fig. 6).
The spectral position of these resonances may be calculated by modelling the pillar cavity as a waveguide with an effective refractive index. The theoretical results are in excellent agreement with the experimentally observed pillar mode positions and patterns.
We have also extended our studies to the ultraviolet regime of the electromagnetic spectrum. To minimize absorption in the mirrors of the resonator, HfO_2 and SiO_2 were used as high and low refractive index materials. We manufactured the first high-quality dielectric pillar microcavities with embedded colloidal ZnO quantum dots in the ultraviolet (Thomay et al., 2008). The semiconductor ZnO possesses excellent optical properties (Klingshirn, 2007). Both the large bandgap of ZnO of 3.4 eV and its exciton binding energy of 60 meV will be strongly beneficial to room-temperature operation of future single-photon devices. ZnO has a low refractive index as compared to other semiconductors, which facilitates light extraction. This compound is also a promising candidate for spintronics applications (Janssen et al., 2008). ZnO quantum dots have shown long electron spin dephasing times even at room temperature (Liu et al., 2007) and have been successfully doped with magnetic ions (Kittilstved & Gamelin, 2006). In the future, the quantum dot ensemble used in this study may be replaced by sputtered ZnO quantum dots (Mayer et al., 2009), magnetically doped ZnO nanocrystals, or other UV emitters.

3.2 Metallic optical nanoantennas

An alternative way of coupling single nanoobjects to the light field is metallic nanostructures. These so-called "optical antennas" are one million times smaller than the radio antenna analogues. Successful fabrication and operation of these devices has only been demonstrated recently (Muhlschlegel et al., 2005; Schuck et al., 2005).
We succeeded in tuning the length and feedgap of a single gold bowtie antenna by precise nanomanipulation ("tuneable nanoantenna") with the tip of an atomic force microscope (AFM) while at the same time monitoring the optical response of the nanostructure via dark-field scattering spectroscopy (Merlein et al., 2008). The gold bowtie optical nanoantenna was fabricated via a colloidal nanomask. The bowtie nanoantenna consists of two gold nanotriangles separated by a tip-to-tip distance of 85 nm (Fig. 7a). One arm of the nanoantenna was moved with the tip of an AFM, so that the total length of the nanoantenna and hence its feedgap were gradually reduced (Fig. 7a and c).
In contrast to all previous experiments on nanoantenna structures, we were able to study the evolution of the optical properties of the same single nanoobject as a function of the geometry. In our experiment, the feedgap was precisely controlled on a nanometer scale, while the other experimental parameters were kept constant. Most importantly, the shape of the nanoantenna arms was not altered. Reducing the antenna gap dramatically changed the scattering spectrum. The original single plasmon peak split into two distinct resonances (Fig. 7b). The optical properties of the metal nanoantenna with tuneable gap may be understood in the following way. At a large feedgap of 85 nm, the dark field scattering spectrum is the superposition of the spectra of the two individual nanotriangles. When the gap decreases, the metal antenna arms begin to couple electrodynamically. The charge density distribution in one triangle at any given time acts on the other arm and vice versa.

As a result, collective plasmon modes in direction of the long antenna axis are formed. In the experiment, two resonances were clearly observed. The exact three-dimensional shape of the nanoantenna is the reason for this effect. Breaking the symmetry of the antenna arms in vertical direction due to tilted sidewalls causes a splitting of the dipole mode into two distinct resonances, which are both of dipole type. In contrast to earlier work on nanoantennas, the full three-dimensional shape has to be taken into account to understand their optical properties. This finding will be particularly important if these antenna structures are resonantly coupled to single nanoemitters, such as semiconductor quantum dots, diamond nanocrystals, or single molecules. These results have also opened the door to new nano-opto-mechanical devices, where mechanical changes on the nanometer scale control the optical properties of photonic structures.



(a) (b) (c)

Fig. 7. Atomic force microscope images (a and c) and dark field scattering spectra (b) of a single gold bowtie optical nanoantenna with feedgap of 85 nm (black line) and 5 nm (red line).

4. Conclusion

In conclusion, we have presented a semiconductor quantum dot as a solid-state system to perform femtosecond quantum optics experiments. The ultrafast dynamics in a single semiconductor quantum dot reveals instantaneous Coulomb renormalization, perturbed free induction decay, and unexpected single-photon gain. It is now possible to prepare, manipulate, and readout this single-electron system on a femtosecond timescale. When driving the dot in the nonlinear regime, it is possible to add single photons to a femtosecond laser pulse. Finally, we have demonstrated means of increasing the coupling of light with objects of nanometer dimensions. We presented dielectric microcavities operating both in the visible and ultraviolet. Alternatively, metal nanoantennas were fabricated. They work as optical counterparts of the well-known radio antennas and concentrate light in the antenna feedgap. We have demonstrated a tunable bowtie nanoantenna and investigated its linear optical properties. We believe that progress in nanotechnology in the next years will further enable us to tailor the interaction of light with single nanoemitters to enter the regime of few-photon femtosecond physics.

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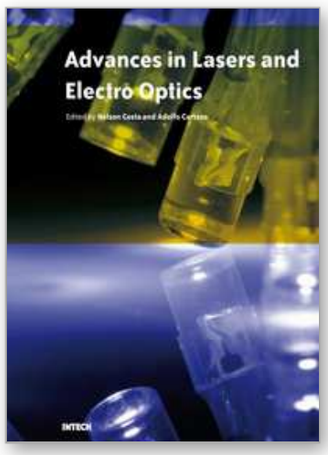

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Lasers and electro-optics is a field of research leading to constant breakthroughs. Indeed, tremendous advances have occurred in optical components and systems since the invention of laser in the late 50s, with applications in almost every imaginable field of science including control, astronomy, medicine, communications, measurements, etc. If we focus on lasers, for example, we find applications in quite different areas. We find lasers, for instance, in industry, emitting power level of several tens of kilowatts for welding and cutting; in medical applications, emitting power levels from few milliwatt to tens of Watt for various types of surgeries; and in optical fibre telecommunication systems, emitting power levels of the order of one milliwatt. This book is divided in four sections. The book presents several physical effects and properties of materials used in lasers and electro-optics in the first chapter and, in the three remaining chapters, applications of lasers and electro-optics in three different areas are presented.

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