Quantum Optical Studies on Individual Acceptor Bound Excitons in a Semiconductor

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We demonstrate the generation of triggered single photons at a predetermined and well defined energy using the radiative recombination of single nitrogen-bound excitons in a semiconductor. The nitrogen atoms are embedded in a ZnSe quantum well structure and were excited by nonresonant optical pumping (82 MHz) at low temperature (4 K). We find resolution-limited photoluminescence lines (280 μeV) which display photon antibunching under continuous optical pumping. Our results also suggest that single nitrogen-bound excitons are well suited for cavity quantum electrodynamics experiments.

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Several applications in the field of quantum information science require a single-photon source which is able to generate photons on demand; i.e., single photons can be emitted within short time intervals and a deterministic dwell time between them. Quantum emitters which are able to emit single photons on demand have been actively studied on different material systems such as single atoms [1,2] and single molecules [3,4].

In semiconductors, studies on single-photon generation have been mainly focused on the recombination of single excitons [5–7] and exciton complexes [8] in optically pumped self-assembled quantum dots (QDs). Recently, electrically driven single-photon sources based on a double-barrier *p-n* heterojunction [9] and on a single InAs QD have been realized [10].

For practical application the emission energy of the quantum emitter should be in resonance with a fundamental mode of a microcavity to achieve a high photon collection efficiency. Single-photon emission from an isolated quantum dot on resonance with a whispering gallery mode [5] and with a fundamental mode of a micropillar [11] has been demonstrated. In each of these experiments accidental matching of a QD resonance with the cavity mode was realized because their transition energies are defined only within the relatively broad inhomogeneous linewidth ($\sim$ 30–50 meV) of the corresponding QD ensemble. From this point of view, single quantum dots possess a serious disadvantage. Thus, one would prefer to use an emitter with a well-known and defined energy which can be naturally embedded in a compact solid-state resonator with an appropriate mode.

The concept of single-photon emission from individual impurity centers in semiconductors is very promising. Recently, photon antibunching of the fluorescence light from a single nitrogen-vacancy center in diamond has been observed at room temperature [12,13]. This energetically deep center shows a broad spectral emission (120 nm) with the zero-phonon line at 1.946 eV. Because of a shelving state it is expected that the single-photon pulse rate is limited to stay below 5 MHz [12]. However, triggered single-photon emission has not been verified so far for shallow impurity centers.

In this Letter, we report *triggered single-photon emission* from individual nitrogen-bound excitons in a ZnSe quantum well structure. This approach is highly attractive with respect to coupling the quantum emitter to a high-quality resonator mode since the emission energy of the acceptor-bound exciton in ZnSe is well known and energetically defined within a small range ($\sim$ 1–2 meV), i.e., 1–2 orders of magnitude better than for QDs. Furthermore, we report the experimental observation of photon antibunching under continuous optical excitation. This study not only describes a fundamental quantum optical effect but also provides the lifetime of an individual acceptor bound exciton.

The samples studied were grown by molecular beam epitaxy on GaAs subtrates [14,15]. ZnSe layers of 20 nm (sample A) and 5 nm (sample B) thickness were sandwiched between two 400 nm thick (Zn, Mg) (S, Se) lattice matched barriers. The structures are fully strained as determined by high-resolution x-ray diffraction measurements. Nitrogen doping of the ZnSe layers was achieved using a radio frequency plasma cell. An active acceptor concentration of $N_A - N_D = 3.7 \times 10^{17}$ cm^{-3} was determined by capacitance voltage measurements. Multiple mesa fields were defined by electron-beam lithography with diameters varying from 100 μm down to 95 nm. These patterns were transferred by chemical wet etching into the layer structures [16].

The samples were mounted in a He-flow cryostate (4 K) which was moved by motor-based translation stages and by piezo actuators allowing for scanning across the sample with a spatial resolution of 50 nm. Optical excitation was performed with either a continuous wave (cw) Stilbene-3 dye laser or a frequency-doubled mode-locked Ti:sapphire laser (82 MHz). Both laser systems are operated at a wavelength of 420 nm where electron-hole pairs are generated in the (Zn, Mg) (S, Se) barriers which are subsequently captured by the nitrogen acceptors in the ZnSe layer. The spatially resolved photoluminescence (μ-PL) has been collected with a diffraction limited scanning optical microscope. The collected PL light was dispersed by a 1 m spectrometer and detected by a charged coupled device (spectral resolution: 280 μeV). Photon correlation measurements have been performed with a Hanbury Brown and Twiss setup consisting of a 50/50 beam splitter and two single-photon-counting avalanche photodiodes (APD) with a time resolution of $\sim$700 ps. The number of photon pairs $n(\tau)$ with arrival time separation τ are recorded. Details of this setup are similar to those reported in Ref. [17].

It is well known that nitrogen atoms in ZnSe are substitutionally incorporated at selenium site (N_{Se}) where they act as shallow acceptor centers. Accordingly, the PL spectra of ZnSe:N are dominated by the nitrogen-bound exciton emission (I_1^N). This is shown exemplarily in Fig. 1 (bottom spectrum) for an ensemble averaged spectrum taken at an unstructured region of sample A where the I_1^N transition is visible at 2.7961 eV with a full width at half maximum (FWHM) of 1.8 meV. The N_{Se} centers are unambiguously identified by the spectrum of their excited states which has been analyzed from the corresponding two-hole-transitions of the I_1^N line as already presented in Ref. [14]. The heavy-hole exciton (hh) can be observed as a relatively weak emission peak at 2.8065 eV, indicating an efficient capture of the generated electron-hole pairs by the nitrogen atoms. Because of confinement effects in sample B with the 5 nm ZnSe:N layer, the I_1^N and hh emission is blueshifted to 2.815 and 2.831 eV, respectively (not shown).

The μ-PL spectra show several spectrally sharp lines due to bound-exciton transitions at individual nitrogen atoms (Fig. 1). The number of these lines decreases with decreasing mesa diameter, and their relative intensities are comparable in the smaller structures (< 300 nm). This is expected for optical transitions at individual centers as long as additional nonradiative recombination channels are not involved, e.g., near the inactive surface layer of the mesas. For the smallest mesa diameters, just one strong resolution-limited line dominates the whole spectrum. The true linewidth could be considerably smaller; i.e., the measured value of 280 μeV is an upper limit. In addition, a broad emission band is visible in the unstructured sample on the low-energy side of the I_1^N transition as can be seen in the bottom spectrum of

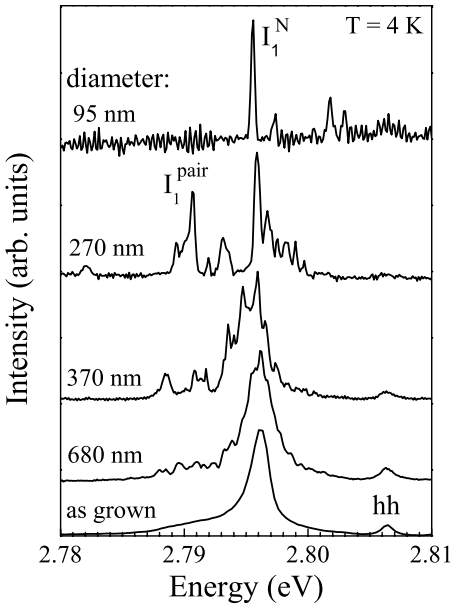

FIG. 1. μ-PL spectra of sample A for different mesa diameters under cw excitation with 500 W/cm^2. All curves are normalized and shifted vertically for a better representation. $T = 4$ K.

Fig. 1. This band evolves likewise with decreasing mesa diameter showing several sharp lines referred to as I_1^{pair}. The I_1^{pair} emission is attributed to the recombination of excitons bound to $[N_{Se}-N_{Se}]$ pairs with different spatial separations located on individual sites of the discrete lattice. This common low-energy wing of the acceptor-bound exciton transition has already been observed in the ensemble averaged PL of, e.g., GaP:N [18], GaAs:Be [19], and CdTe:Cu [20] while the recombination model has been introduced by Street and Wiesner [18].

The observation of a discrete emission line cannot be taken as an evidence that the observed system consists of a single quantum emitter. In contrast, photon correlation measurements such as reported below provide a reliable method for deciding whether or not the observed system is a single two-level quantum emitter. The measured photon-count distributions $n(\tau)$, in the energy region of the acceptor-bound exciton emission are shown in Fig. 2(a) for an unstructured sample part and in Fig. 2(b) for a mesa structure with 160 nm diameter. Both traces are normalized to the expectation value for completely uncorrelated photons obeying a Poissonian arrival time distribution. Trace 2(a) is flat over the full measurement time due to the contribution of many bound-exciton emitters. In contrast, trace 2(b) exhibits a clear dip in the correlation counts for a time delay $\tau = 0$, indicating strong photon antibunching.

To model our photon correlation experiment, we use a simple three-state model recently presented by Michler *et al.* [21]. In order to correct for the time resolution of the APD's the normalized count distribution $\bar{n}(\tau)$ is fitted

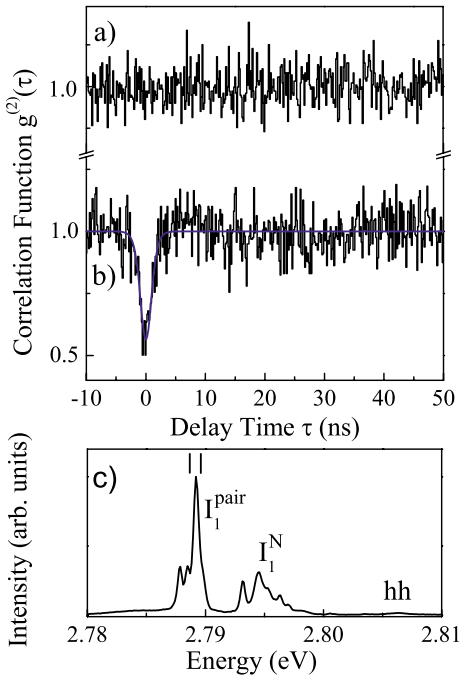

FIG. 2 (color online). Measured distribution of coincidence counts $n(\tau)$ for an unstructured part (a) and an etched mesa structure with 160 nm diameter (b) of sample A under cw excitation with 500 W/cm^2. The solid line is a fit of the correlation function $g^{(2)}(\tau)$. The corresponding μ-PL spectrum for trace (b) is shown in (c). The spectral window for the correlation measurement of trace (b) has been set to the I_1^{pair} emission peak at 2.7891 eV with a FWHM of 800 μeV as marked by the vertical bars. $T = 4$ K.

with the correlation function $g^{(2)}(\tau) = 1 - ae^{-|\tau|/t_m}$ convolved with a Gaussian time distribution with a FWHM of 700 ps. The parameter a accounts for the background of uncorrelated photons present in the measurement, and t_m is the antibunching time constant. The fitted $g^{(2)}(\tau)$ is shown as a solid line in Fig. 2(b). The value of $1 - a = g^{(2)}(0)$ obtained from the fit amounts to 0.10, i.e., $g^{(2)}(0) < 0.5$, and this indicates that the measured photon antibunching stems dominantly from a single quantum emitter. However, the measured $g^{(2)}(0)$ does not reach its theoretical minimum of zero. This is caused by the presence of background stray light and the contribution of photons from an additional weak discrete line emitting within the selected spectral window [compare Fig. 2(c)]. The time constant t_m for the exponential decay of the antibunching was determined to be 590 ps. In the limit of a low excitation power t_m directly corresponds to the lifetime of the quantum emitter [17,21]. The excitation power used for recording trace 2(b) is a factor of 40 lower than the saturation power of the bound-exciton emission determined from an unstructured part of sample A, providing that t_m is a good estimate for the lifetime of the single emitter. The fitted value for t_m of the I_1^{pair} is slightly longer compared to literature values for ensemble average measurements of the I_1^N on strain relaxed layers (430 ps) [22]. However, further systematic time-resolved measurements on individual I_1^{pair} and I_1^N lines are necessary, which is beyond the scope of this Letter.

In order to demonstrate that individual shallow impurities in a semiconductor are well suited for the implementation in photon turnstile devices, triggered single-photon emission has been verified using pulsed Ti:sapphire excitation. Figure 3 shows the measured unnormalized correlation function $G^{(2)}(\tau)$ of the I_1^N transition for an unstructured part 3(a) and a 95 nm mesa structure 3(b) of sample A, and for a 95 nm mesa structure of sample B 3(c). As expected, the measured $G^{(2)}(\tau)$ for the ensemble [Fig. 3(a)] exhibits peaks at integer multiples of the repetition time of 12 ns with negligible signal in between the peaks corresponding to a Poissonian photon statistic. Additionally, this demonstrates a successful locking of the photon emission to the pulsed excitation. In contrast to trace 3(a), the peaks at $\tau = 0$ are highly suppressed for traces 3(b) and 3(c), providing strong evidence for single-photon emission [5].

The areas of the zero delay peaks for traces 3(b) and 3(c) are 46% and 19% of those at other delay times, respectively. This indicates an approximate decrease in multiphoton emission pulses by a factor of 5 for trace 3(c) as compared to a Poissonian light source of the same average intensity. The corresponding normalized μ-PL spectra for the correlation measurements of Figs. 3(a)–3(c) are shown in Figs. 3(d)–3(f), respectively. The energetically flat background under the peak area of the discrete lines has been estimated to about 41% for Fig. 3(e) and 9% for Fig. 3(f), indicating that the residual uncorrelated photons at $\tau = 0$ are mainly caused due to insufficient suppression of background light. Therefore, the rate of multiphoton emission could be further suppressed by reducing the background light from the sample which is caused by laser stray light and due to the contribution of the low-energy wing of the barrier luminescence.

The doping concentration of donors and acceptors in semiconductors can be controlled within a wide range, especially 3 orders of magnitude lower than in our approach. Thus, single donor or acceptor atoms could in principle be embedded into nanostructured microcavities. Because of the well-known and sharp transition energies of the impurity-bound excitons, these systems are highly attractive for cavity-QED experiments in an all-semiconductor nanostructure. At this point it is interesting to discuss whether the strong coupling regime can be reached with a single impurity embedded in a microcavity. In order to observe the Rabi splitting for a single emitter, the Rabi energy $\hbar\Omega = dE_{\max}$ should be larger than the arithmetic average of the emitter and the mode linewidth. With current technology a maximum electric field per photon of $E_{\max} \sim 10^5$ V/m can be reached in semiconductor microcavities [23]. Because of their higher electrical dipole moment donor bound excitons may be even more suitable for cavity-QED experiments

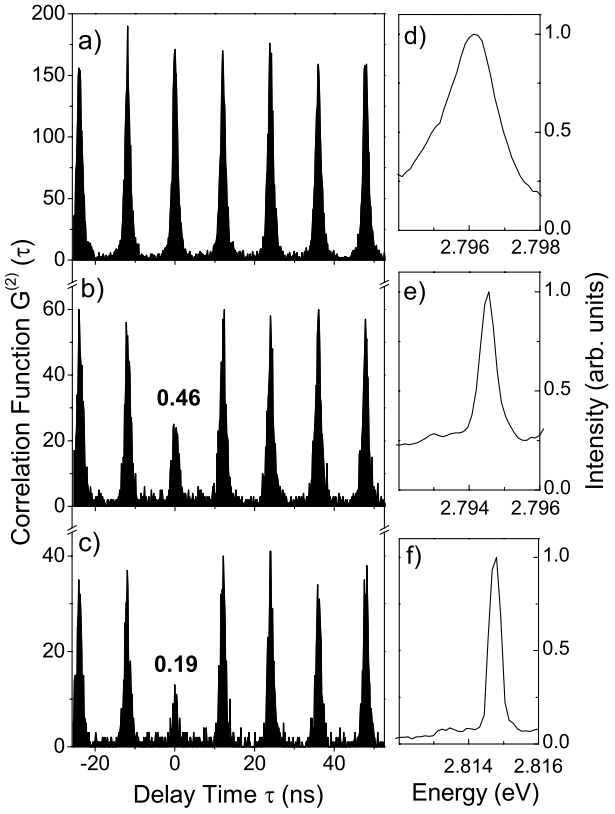

FIG. 3. Measured unnormalized correlation function $G^{(2)}(\tau)$ in the energy region of the I_1^N transition for (a) an unstructured part (ensemble average) and for a 95 nm mesa structure of sample A (b) and sample B (c) under pulsed excitation with a Ti:sapphire laser (82 MHz). The corresponding normalized μ-PL spectra for traces (a)–(c) are shown in (d)–(f), respectively, recorded with an energy resolution of 500 μeV as used for the correlation measurements. $T = 4$ K.

than acceptor-bound excitons studied in this work. The electric dipole moment d, e.g., of the donor bound-exciton transition (I_2) in GaAs can be estimated from its recombination lifetime ($\tau = 179$ ps, Ref. [24]) to 1.75×10^{-28} Cm which results in a Rabi energy of 110 μeV. This value is larger than both the linewidth, e.g., for a high-Q whispering gallery mode (~ 100 μeV) and the linewidth of the I_2 emission in GaAs (e.g., values of ~ 40 μeV have already been reported for the ensemble emission [25]). Furthermore, we point out that the electrical dipole moment for the I_2 emission in ZnSe is $d = 2.3 \times 10^{-28}$ Cm ($\tau = 40$ ps, Ref. [26]). If we assume state-of-the-art quality factors of GaAs cavities ($Q \sim 10000$), even enlarged Rabi energies of $\hbar\Omega = 144$ μeV could be obtained in the ZnSe material system. Thus, we envision that a single donor bound exciton could provide a solution to achieve the challenging goal of the strong coupling regime in a semiconductor microcavity.

In conclusion, we have investigated the photon emission of nitrogen atoms embedded in mesa etched ZnSe:N quantum well structures by use of spatially resolved μ-PL and by photon correlation measurements. The μ-PL spectra show several spectrally sharp lines due to bound-exciton transitions at individual nitrogen atoms. These separate resolution-limited lines (FWHM: 280 μeV) display photon antibunching under continuous optical excitation. In addition, triggered single-photon emission has been verified under pulsed optical excitation (82 MHz) with a decrease in multiphoton emission up to a factor of 5 as compared to a Poissonian light source. This demonstrates that individual shallow impurities in a semiconductor are well suited for quantum optics experiments.

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