Single photon emission from a site-controlled quantum dot-micropillar cavity system

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We demonstrate the deterministic integration of single site-controlled quantum dots (SCQDs) into micropillar cavities. Spatial resonance between single positioned QDs and GaAs/AlAs micropillar cavities was achieved using cross markers for precise SCQD-cavity alignment. Cavity effects are clearly reflected in an enhanced photoluminescence intensity when tuning SCQD emission lines through the fundamental cavity resonance. Single photon emission from a spatially and spectrally coupled SCQD-resonator system is confirmed by photon autocorrelation measurements yielding a $g^{(2)}(0)$ value of 0.12. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3097016]

Integration of site-controlled quantum dots (SCQDs) (Ref. 1) into optical resonators is an important step toward the deterministic exploitation of cavity quantum electrodynamic (cQED) effects[2] on a semiconductor platform. The long range ordering of SCQDs combined with accurate alignment procedures[3] facilitates not only high yield device fabrication but is also a prerequisite for the realization of more complex nanodevices.[4–6] Recently, the integration of SCQDs into photonic crystal (PhC) cavities has been demonstrated by several groups.[7–9] Whereas PhC cavities promise severe advantages for on chip in-plane quantum information processing,[5] micropillar cavities are promising candidates for nonclassical light sources due to very efficient and directed vertical emission behavior and the straightforward way of realizing electrical pumping. Furthermore, micropillar cavities can be regarded as a model system for the exploration of cQED effects with QDs.[10,11] The underlying cQED effects rely on a spectral and spatial overlap of the emitters with the resonator modes. While spectral resonance can be achieved via several methods,[10,12,13] deterministic spatial resonance has only recently been achieved by postgrowth alignment of micropillars to randomly grown QDs.[14] As spatial control is of major importance, the accurate control over the QD location in the micropillar makes the deterministic integration of SCQDs into micropillar cavities highly desirable.

In this letter, we report on a technology to integrate SCQDs into micropillar devices, resulting in spatially and spectrally resonant SCQD-resonator systems acting as triggered single photon sources. In contrast to the integration of SCQDs into photonic crystal cavities, sample growth as well as device alignment is much more challenging in the distributed Bragg reflector (DBR) structures. The possibility of retrieving the QD positions must be assured after the overgrowth of GaAs/AlAs material forming the upper DBR with a thickness of several microns. Furthermore, in PhC cavities the spectral position of the cavity mode can be mainly controlled by the processed geometry and is therefore rather independent of the growth. The spectral resonances of micropillar cavities are, in contrast, almost fixed by the Bragg condition in the planar cavity and can only be slightly changed with the pillar diameter. Therefore the sample growth itself has to be performed in such a way that the planar cavity almost matches the SCQD PL already. These challenges make the growth and patterning of spectrally resonant SCQD-micropillar cavity systems very demanding. Yet, taking into account a possible temperature tuning range of a single QD in the order of 1 meV and a full width at half maximum of the photoluminescence (PL) spectrum of 23 meV, as presented in this letter, the possible yield of resonant QD-resonator systems is on the order of 3%.

It is interesting to compare this estimated yield with the expected one for samples based on randomly distributed QDs. Herein, the yield of spatially resonant randomly grown QD-resonator systems with a spatial deviation of less than 200 nm from the cavity center with a comparable surface density can be estimated to a value of 0.5%, which is clearly exceeded by our approach.

To realize a spatially resonant SCQD-micropillar system we have established the following fabrication technology. In the first molecular beam epitaxy growth step the bottom part of AlAs/GaAs planar microcavities, consisting of the bottom DBR section and the lower half of the central GaAs one λ cavity was prepared. Then, micrometer sized cross markers for device alignment and low density (from 500 nm to 2 μm period) square arrays of nanoholes serving as nucleation centers for the QD growth were processed on the sample in several lithography and etch steps (see Ref. 3 for details).

After removing the native oxides[3] overgrowth of the prepatterned substrate was performed. In order to optimize the optical quality of the SCQDs, three layers of InAs were stacked on top of the nanoholes as depicted schematically in Fig. 1(a). As shown in a representative atomic force microscopy image on uncapped SCQDs in Fig. 1(b), well ordered QD arrays have been obtained for the stacked InAs layers in agreement with other reports.[1] The overgrowth was initiated by an 8 nm thick GaAs buffer layer followed by the first InAs layer. The second and third InAs layers were separated by 10 nm thick barriers. The InAs layers have been deposited at a rate of 0.005 nm/s and a substrate temperature of 530 °C. In the first InAs layer, we deposited a nominal amount of 2.5 ML of semiconductor material. Studies on uncapped InAs layers grown on nucleation centers in this manner revealed neither distinct QD nucleation on the nano-

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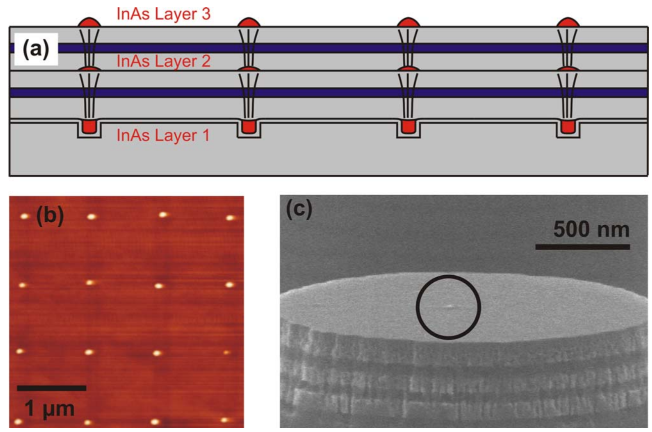

FIG. 1. (Color online) Growth of position controlled QD arrays and deterministic device integration. (a) Sketch of the growth scheme applied for stacked, position controlled QDs. (b) Atomic force micrograph of uncapped SCQDs in the third deposited InAs layer. (c) SEM image showing the device alignment of a micropillar cavity and a single SCQD.

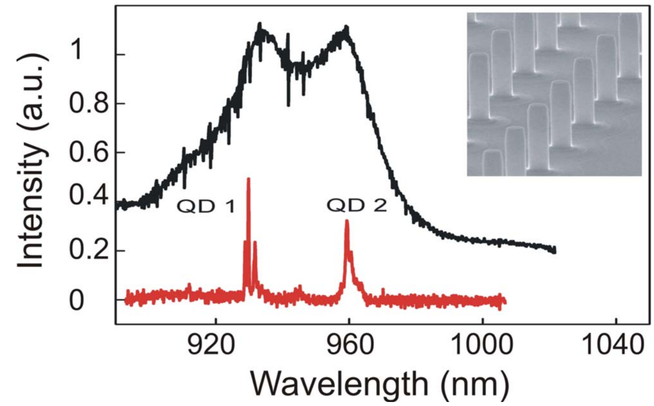

FIG. 2. (Color online) Characterization of a cavity structure containing ordered arrays of SCQDs. The ensemble luminescence of the SCQDs (recorded at 30 K after removing the top DBR section) exhibits two separated peaks (black, upper graph) matching the two distinct peaks in the representative μPL spectrum of a microcavity device (red, lower graph).

holes nor at interstitial positions, which indicates that noticeable desorption takes place at this high temperature. Although no pronounced QD formation is observed in this first layer, comparison with reference samples revealed the importance of the deposited indium for the ordered QD stacking in the subsequent layers. In the second and third InAs layer, 3.3 MLs of InAs were deposited in order to initiate QD formation at the predetermined sites by strain coupling. Both, the first and the second InAs layers were capped by 2 nm of GaAs followed by an *in situ* annealing step at 560 °C for 2 min. This particular growth routine shifts the emission of the QDs in the first and second layer towards higher energies[15] in order to spectrally detune their emission from those SCQDs located in the topmost third layer envisaged for the coupling to the resonator. A statistical analysis of the uncapped QDs on the surface yields a mean QD height of 10 nm, while less than 6% of the QD sites remain unoccupied and 2.5% of all QDs nucleate at interstitial positions for a lattice period of 1 μm.

To demonstrate the feasibility of deterministically integrating SCQDs into micropillar cavities, devices have been fabricated on reference structures containing uncapped SCQDs as can be seen in Fig. 1(c).[16] The pillars, containing 25 mirror pairs in the bottom DBR and the lower half of a one λ-thick cavity were aligned with respect to the same reference cross markers on the sample as the nanoholes and the SCQDs, respectively. Based on preceding work in which the accuracy of the alignment procedure has been evaluated in detail,[3] the exemplary image in Fig. 1(c) demonstrates the capability of the established technology to place single SCQDs laterally in the center of micropillar structures.

For the integration into full micropillar cavities the alignment marker edges have to be well maintained even after overgrowth, which amounts to ~2 μm for the presented sample. Therefore, we examined the evolution of the cross markers for different overgrowth thickness (not shown). From the obtained results we conclude that the alignment accuracy is still sufficient even after deposition of up to 4 μm deposited material. This layer thickness is typical for the top DBR section of a high quality factor micropillar cavity with ~23 mirror pairs.[10]

In order to demonstrate cQED interaction in a spatially resonant SCQD-micropillar cavity system, we integrated a stack of three InAs layers in a planar AlAs/GaAs microcavity structure. The cavity was sandwiched between 25 mirror pairs in the bottom DBR and 12 mirror pairs in the top DBR. To probe the ensemble PL, the top DBR was removed. The corresponding spectrum plotted in Fig. 2 shows two clearly separated peaks at 930 and 960 nm, which we attribute to the second QD layer (930 nm) and the third QD layer (960 nm) showing the accomplished spectral detuning.

Detailed optical studies of spatially resonant QD-micropillar systems were performed at low temperature (10–50 K). Taking into account the excellent position control of the QD formation process [cf. Fig. 1(b)] and the high alignment accuracy demonstrated in Fig. 1(c) one can expect spatially deterministic SCQD-micropillar cavity coupling. In fact, each processed micropillar with a diameter below 2 μm (see inset of Fig. 2) patterned on a SCQD array with a period of 1 μm is expected to contain with high probability of $>\sim 90\%$ only one single SCQD emitting at about 960 nm. An exemplary μPL spectrum of a microresonator with a diameter of 1 μm is shown in Fig. 2. Two features, one at 959 nm and the other at 929 nm, indicate that only two spectrally detuned SCQDs are contained in the pillar.

Figure 3 summarizes PL results obtained on a 1 μm diameter micropillar cavity. Figure 3(b) shows exemplarily the emission spectrum at 42 K. Lorentzian fitting to the cavity mode yields a quality factor of ~1700. According to transfer matrix simulations, the Q-factor of an empty, planar resonator with the given number of mirror pairs and the experimentally determined layer thicknesses yields a Q-factor of about 1800, which is in reasonable agreement with the experimentally extracted value. The linewidth of the single SCQD emission lines is 280 μeV. It is worth noting that the SCQD linewidths are even somewhat better than reported values on InAs SCQD samples grown under optimized conditions.[17] Despite the cavity effect, we see herein an indication of an improved optical quality of the topmost SCQDs owing to the larger separation to the prepatterned interface.

We attribute the occurrence of several emission lines in Fig. 3(a) in a spectral range of a few meV to the excitonic (X), the positively charged (X+), and the biexcitonic (XX) emission of one single SCQD, as supported by power dependent measurements (not shown). The increased intensity ob-

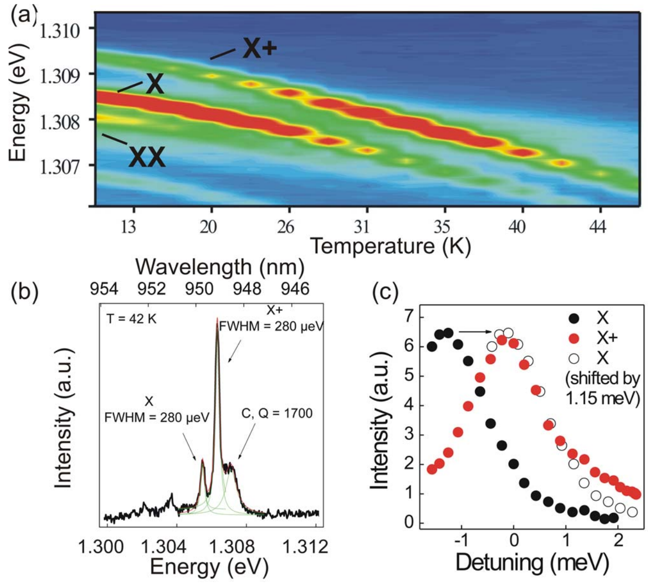

FIG. 3. (Color online) (a) Contour plot of a tuning series of a single QD through the cavity resonance by temperature. (b) PL spectrum recorded at 42 K. (c) Integrated intensities of the $X+$ emission line (filled red circles) and the X transition (filled black circles). For better comparison, the X transition is additionally shifted by 1.15 meV (black open circles).

served for the $X+$ transition compared to the X transition in Fig. 3(b) is ascribed to cavity enhanced emission.

The cavity induced enhancement of the emission can be controlled by temperature tuning, as evident from Fig. 3(a). As can be seen from the intensity plot versus detuning in Fig. 3(c), the X and $X+$ emission lines show an almost identical resonance behavior associated with an enhancement of emission by the same factor. This indicates that both emission lines originate from a single SCQD. These characteristic features demonstrate a weakly coupled SCQD-micropillar cavity system. It is interesting to note that cavity emission is almost absent under weak excitation conditions when no transition line of the SCQD is in resonance with the cavity mode. This is a clear advantage of the present system in comparison to QD-microcavity systems including randomly grown QDs. The latter suffer typically from strong background emission[18] from a large number of off-resonant QDs illuminating the cavity mode which complicates the application of such systems as single photon sources.[19]

To prove the feasibility of a coupled SCQD-micropillar system acting as a single photon source, we performed photon autocorrelation measurements.[18] The SCQD is excited by a mode locked Ti:sapphire laser tuned to an emission wavelength of 760 nm and providing 150 fs wide pulses at a repetition rate of 82 MHz. The photon statistics of the cavity emission was studied at zero detuning of $X+$ with respect to the cavity mode in order to enhance the coupling of spontaneous emission into the cavity mode.[2] The corresponding second order autocorrelation function shown in Fig. 4 reveals strong antibunching as expected from a nonclassical light emitter. In particular, single photon emission with a low probability of multiphoton events is reflected by the observed value of $g^{(2)}(0)=0.12<1/2$. This result is very promising with respect to the application and integration of site-controlled QDs in quantum light sources.

In conclusion, we have demonstrated the integration of single, site-controlled QDs into micropillar cavities. Both the

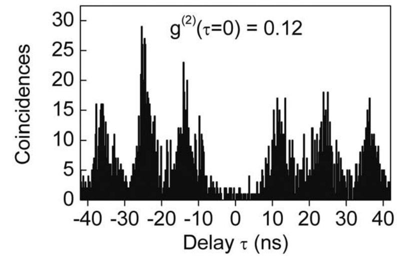

FIG. 4. Photon autocorrelation measurement of the SCQD emission line ($X+$) on resonance with the cavity mode. A $g^2(0)$ value of 0.12 was determined.

interaction of a QD with the cavity mode and single photon emission of the QDs were demonstrated in our spatially and spectrally coupled cavity-SCQD system. We believe that these results can be considered as a major step in the field of semiconductor based quantum information processing.

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