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Single-photon emission at a rate of 143 MHz from a deterministic quantum-dot microlens triggered by a mode-locked vertical-external-cavity surface-emitting laser

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We report on the realization of a quantum dot (QD) based single-photon source with a record-high single-photon emission rate. The quantum light source consists of an InGaAs QD which is deterministically integrated within a monolithic microlens with a distributed Bragg reflector as back-side mirror, which is triggered using the frequency-doubled emission of a mode-locked vertical-external-cavity surface-emitting laser (ML-VECSEL). The utilized compact and stable laser system allows us to excite the single-QD microlens at a wavelength of 508 nm with a pulse repetition rate close to 500 MHz at a pulse width of 4.2 ps. Probing the photon statistics of the emission from a single QD state at saturation, we demonstrate single-photon emission of the QD-microlens chip with $g^{(2)}(0) < 0.03$ at a record-high single-photon flux of (143 ± 16) MHz collected by the first lens of the detection system. Our approach is fully compatible with resonant excitation schemes using wavelength tunable ML-VECSELs, which will optimize the quantum optical properties of the single-photon emission in terms of photon indistinguishability. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4927429]

A light source emitting single photons at high repetition rates is highly desirable with respect to future applications in the field of quantum information technology.[1] Single semiconductor quantum dots (QDs) turned out to be promising candidates for the realization of such non-classical light sources.[2] In particular, the engineering of electrically operated devices has been actively pursued within the nanophotonics community[3–7] and recently, even the feasibility of quantum-key distribution within a 500 m free-space experiment using electrically triggered QD single-photon sources (SPSs) has been demonstrated.[8] Using suitable pulse-generators such SPSs can be triggered electrically with frequencies up to the GHz range[9,10] in order to maximize the single photon flux. On the other hand, quasi-resonant[11,12] or strict-resonant[13–15] excitation is highly desirable with respect to the generation of indistinguishable photons due to reduced temporal jitter and dephasing. These excitation schemes, however, are not easily compatible with electrical current injection schemes[16–18] as in most cases the charge carriers are injected above-band via p-i-n diode structures. Recently, we tackled this issue by utilizing an integrated electrically driven microlaser on the same chip as the non-classical light source,[19] but a very sophisticated device technology is required in this approach. Therefore, in most quantum optics experiments QDs are excited optically using commercial laser systems based on mode-locked Ti:sapphire lasers, which offer ps- or fs-pulse-widths and can be tuned over a broad spectral range. These widely used laser systems, however, are rather bulky, expensive and most importantly have limited repetition rates of typically ~80 MHz, restricting the achievable single photon flux for a given photon extraction efficiency.

In this work, we realize an ultra-bright SPS by optically exciting a deterministically integrated single-QD microlens using a frequency-doubled mode-locked vertical-external-cavity surface-emitting laser (ML-VECSEL). The compact and stable laser system allows us to overcome the limited repetition rates of commercial mode-locked (ML) Ti:sapphire lasers and to excite the single-QD microlens with a pulse repetition rate close to 500 MHz and a pulse width of 4.2 ps at a wavelength of 508 nm. Probing the photon statistics of the emission of a single QD-state at saturation, we demonstrate single-photon emission of the QD microlens with $g^{(2)}(0) < 0.03$ at a record-high single-photon flux of (143 ± 16) MHz collected by the first lens of the detection system.

The sample used for the fabrication of deterministic QD microlenses was grown by metal-organic chemical vapor deposition on GaAs (001) substrate. After a 300 nm thick GaAs buffer layer, a distributed Bragg reflector (DBR) consisting of 23 alternating $\lambda/4$-thick layers of AlGaAs (77.0 nm) and GaAs (65.7 nm) is grown. Next, 65 nm of GaAs is deposited followed by a low-density layer of self-organized InGaAs QDs which are realized in the Stranski-Krastanow growth mode. Finally, the QDs are capped by 400 nm of GaAs. The thickness of the GaAs capping layer is optimized for the realization of microlenses with high photon

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extraction efficiency. The actual QD microlens is then fabricated deterministically onto a single pre-selected InGaAs QD via a recently established *in-situ* electron beam lithography technique.[20,21] Here, cathodoluminescence spectroscopy in combination with 3D *in-situ* electron-beam writing and subsequent plasma etching is used to precisely define a monolithic microlens on top of the pre-selected QD. The overall alignment accuracy is 34 nm.[22] The resulting QD microlenses (cf. Fig. 1(c)) feature enhanced photon extraction efficiencies as reported recently in Ref. 21.

To optically excite the QD microlens with high pulse repetition rates, we implemented a compact laser system emitting 508-nm ps-pulses based on a ML-VECSEL (see Fig. 1(a)) into a standard micro-photoluminescence (μPL) spectroscopy setup (Fig. 1(b)). The laser system itself is mainly composed of an optically pumped VECSEL chip in combination with a semiconductor saturable-absorber mirror (SESAM) and a nonlinear crystal made out of beta-barium borate (BBO) for frequency conversion via second-harmonic generation (SHG). The VECSEL chip consists of 10 (InGa)As quantum wells equally spaced by $\lambda/2$ (GaP)As barrier layers. The DBR consists of $24^1/_2$ pairs of quarter wavelength GaAs/(AlGa)As layers. Details of the semiconductor layer structure can be found in Ref. 23. Such a chip is then mounted to a water-cooled heat-sink and optically pumped by a continuous-wave (CW) diode laser emitting at 808 nm.[24,25] The frequency-doubled emission of the ML-VECSEL is coupled to the μPL setup via a cold mirror. Here, a microscope objective (MO) with a numerical aperture of 0.4 focuses the laser light onto the QD microlens,

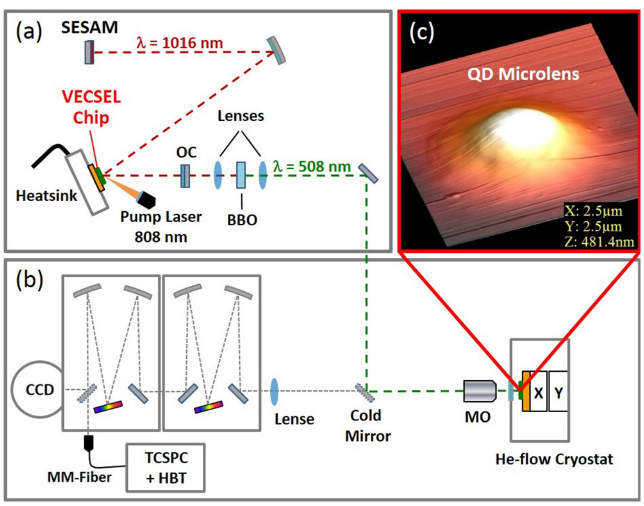

FIG. 1. Schematic of the experimental setup used to operate a quantum dot (QD) single-photon source at high repetition rates of 494 MHz using a mode-locked vertical-external-cavity surface-emitting laser (ML-VECSEL). (a) The laser system comprises an optically pumped VECSEL chip operating at 1016 nm in combination with a semiconductor saturable-absorber mirror (SESAM) and a nonlinear crystal (BBO) for second-harmonic generation of 508 nm light pulses (OC: output coupler). (b) Micro-photoluminescence setup: Emission of the QD sample is collected via a microscope objective (MO) serving as first lens of the detection system and spectrally analyzed using a double-grating spectrometer. Time-resolved and Hanbury-Brown and Twiss (HBT) type measurements can be performed at a second output port of the spectrometer. (c) Atomic force microscopy image of a QD microlens fabricated via *in-situ* 3D electron beam lithography. In the experiments, a deterministic single QD microlens with a base diameter of 2 μm acted as single-photon emitter.

which is mounted onto the cold-finger of a liquid-helium-flow cryostat for cooling the sample to a temperature of 20 K. The same MO serves as the first lens of the detection system and collects the emission of the QD-microlens chip. The collimated emission is then spectrally analyzed using a double-grating spectrometer with attached charge-coupled device (CCD) camera enabling a spectral resolution down to 25 μeV. To perform time-resolved (TR) measurements, a multi-mode (MM) fiber attached to a second output port of the spectrometer is coupled to a Si-based avalanche photodiode (APD) with a timing resolution of 40 ps. The photon statistics of the QD emission is analyzed by means of photon-autocorrelation measurements using a 50:50 MM fiber-coupled beam-splitter (BS) and two Si-APDs in a Hanbury-Brown and Twiss (HBT) type setup with an overall timing resolution of 380 ps. Both the HBT and TR setup utilize time-correlated single-photon counting (TCSPC) electronics for coincidence measurements.

Prior to single QD experiments, the utilized laser system was analyzed in terms of its spectral and temporal emission features. Fig. 2(a) displays the spectrum of the ML-VECSEL emission after SHG, which shows a full width at half maximum of 0.2 nm. The maximum output power of the laser after the BBO crystal was measured to be 5 μW. A TR measurement of the ML laser emission recorded via a Si-APD with a timing resolution of 40 ps is depicted in Fig. 2(b). Here, a pulse train with equidistant pulse separations of 2.025 ns can be observed corresponding to a pulse repetition rate of 494 MHz. The shape of a single laser pulse of the 1016 nm VECSEL emission recorded with a commercial intensity autocorrelator is shown in Fig. 2(c), which exhibits a pulse width of 4.2 ps. Next, the ML-VECSEL was used to optically excite the QD sample. Fig. 3(a) shows a μPL spectrum of a single-QD microlens under ML-VECSEL excitation at a laser power of 1.3 μW. The spectrum is dominated by the emission of the positively charged trion state X^+ at an emission energy of 1.335 eV. The assignment of the emission to a specific QD state was carried out via polarization and power dependent measurements as described in Ref. 26. At lower energies around 1.331 eV, the emission of other excitonic states of the same QD is visible. The exceedingly bright emission of the X^+ state (maximum count rate of 65 kHz per CCD-pixel) in comparison to other QD states is

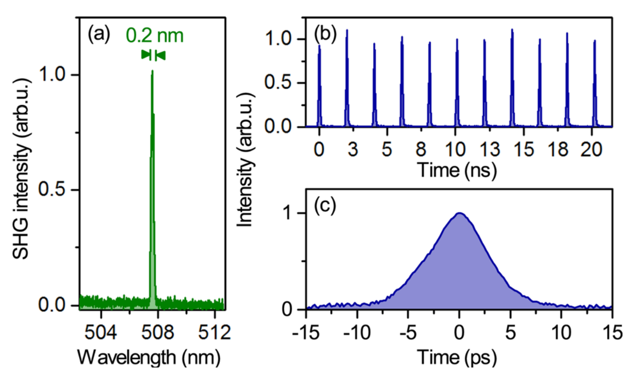

FIG. 2. (a) SHG spectrum of the frequency-doubled VECSEL emission under mode-locked operation at 494 MHz. (b) Corresponding time-resolved measurement on a laser pulse train. (c) Autocorrelation measurement of the 1016 nm VECSEL emission revealing a pulse width of 4.2 ps.

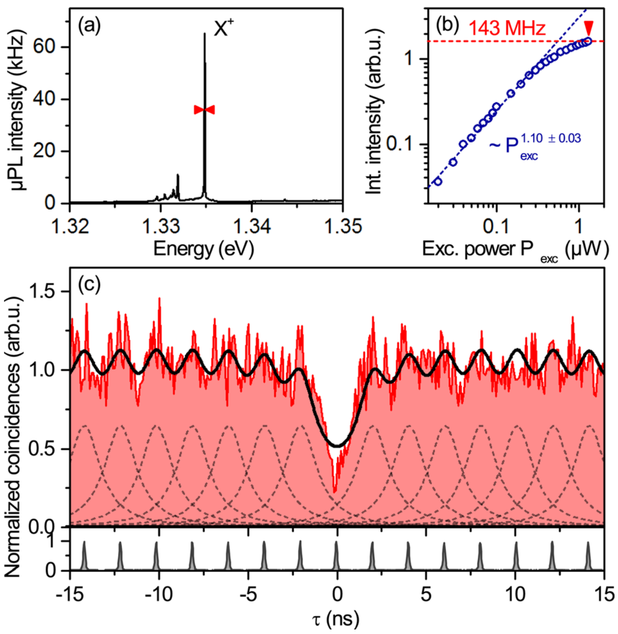

FIG. 3. Analysis of the emission of a deterministic QD microlens excited by a frequency-doubled ML-VECSEL at a repetition rate of 494 MHz. (a) μPL spectrum of the single QD microlens, revealing bright emission of a positively charged exciton state (X^+) at an excitation power of $P_{exc} = 1.3\,\mu$W. (b) Spectrally integrated μPL intensity of the X^+ emission in dependence on the ML-VECSEL excitation power. The dashed blue line corresponds to a linear fit to the experimental data. The maximum X^+ emission intensity is reached at 1.3 μW corresponding to saturation of the QD state (dashed horizontal line). At this working point, the single-photon flux emitted by the QD into the first lens of the setup amounts to 143 MHz. (c) Photon-autocorrelation histogram measured on the X^+ emission at saturation (spectral filtering is indicated by arrows in (a) as well as the working point in (b)). The coincidence data reach a minimal value of 0.22. The model curve (solid black line) reveals an upper bound to the antibunching value of $g^{(2)}(0) < 0.03$, clearly proving single-photon emission. The lower panel shows for comparison the photon-autocorrelation histogram measured on the ML-VECSEL emission.

attributed to an intrinsic p-type background doping during growth, which is typically observed in this type of QD sample.[20,26] To probe the achievable occupation of the X^+ state, excitation-power dependent μPL spectra have been evaluated. In Fig. 3(b), the integrated intensity of the X^+ emission is depicted as a function of the ML-VECSEL excitation power at 508 nm in logarithmic scaling. An almost linear excitation power dependence with a slope of 1.10 ± 0.03 is observed up to an excitation power of 0.3 μW. Further increasing the excitation power results in saturation of the X^+ state, until the maximum emission intensity is reached at $P_{exc} = 1.3\,\mu$W (indicated by red arrow in Fig. 3(b)). In order to gain insight into the photon statistics of the QD emission under these excitation conditions, we performed measurements of the second-order photon-autocorrelation $g^{(2)}(\tau)$. For this purpose, the spectrally filtered emission of the X^+ state (indicated by red arrows in Fig. 3(a)) was coupled to the fiber-based HBT setup. The excitation power of the ML-VECSEL was set to 1.3 μW for saturation of the QD state, resulting in a combined detection rate of 71.5 kHz at the APDs. The resulting coincidence histogram of $g^{(2)}(\tau)$ is presented in the upper panel of Fig. 3(c). A strongly reduced number of coincidences with a measured value of 0.22 can be observed at zero time delay ($\tau = 0$), which indicates single-photon emission of the QD microlens. Furthermore, coincidence maxima with a periodicity of 2.025 ns, corresponding to the pulse repetition rate of the ML-VECSEL, can be clearly identified at finite time delays. This gets even more evident by comparing the data with the $g^{(2)}(\tau)$ histogram measured directly on the frequency-doubled emission of the ML-VECSEL, which is shown as a reference in the lower panel of Fig. 3(c). The strongly overlapping coincidence peaks observed in this measurement are due to the high repetition rate of the excitation laser generating pulses with a separation close to the lifetime ($\sim$1.5 ns) of the QD state. To quantitatively extract the suppression of two-photon emission events, we modeled the data with equidistant photon pulses represented by Lorentzian profiles with 2.3 ns full-width at half maximum and a pulse separation according to the 494 MHz repetition rate of the exciting laser. The area of the pulses was assumed to be constant, except for the zero-delay peak. The fitted model function (cf. Fig. 3(c)) nicely retraces the experimental data and allows to deduce an upper bound for the antibunching of $g^{(2)}(0) < 0.03$ by dividing the area of the almost vanishing zero-delay peak by the area of the exemplarily displayed peak at finite time delay. Such strong suppression of two-photon emission events unambiguously proves the triggered emission of single-photons from the deterministic QD microlens even at saturation of the QD state.

A major advantage of using a ML-VECSEL for the excitation of a single quantum emitter, as demonstrated in this work, is given by the prospect and achievement of significantly higher excitation repetition rates compared to conventional laser systems such as Ti:sapphire lasers. To quantitatively analyze the emission of the QD microlens in terms of the single-photon flux, we experimentally determined the detection efficiency η_{setup} of our experimental setup.[21] This was achieved by focusing a CW diode laser tuned to the emission wavelength of the X^+ state onto a gold mirror mounted in the cryostat. The laser was attenuated using neutral-density filters in front of the monochromator to achieve APD count-rates at the HBT setup similar to those observed for the QD emission. Taking into account the laser power, the reflection of the gold mirror, the transmission of the cryostat window, the attenuation of the density filters, and the maximal count-rates on the APDs, we determined our setup efficiency to $\eta_{setup} = (0.50 \pm 0.06) \times 10^{-3}$. The X^+ state excited at saturation ($P_{exc} = 1.3\,\mu$W) showed a combined detection rate on the APDs of $R_{det} = (71.5 \pm 2.0)$ kHz, which corresponds to a single-photon flux F_{SPS} emitted into the microscope objective of $F_{SPS} = R_{det}/\eta_{setup} = (143 \pm 16)$ MHz. Taking into account the repetition rate of the ML-VECSEL of 494 MHz, this corresponds to a photon extraction efficiency of the deterministic single QD microlens of (29 ± 3)%. The demonstrated single-photon flux of 143 MHz represents a significant improvement compared to the previous reports on SPSs based on tapered nanowires[27] as well as optically[28] and electrically[29] triggered QD micropillar cavities.

In summary, we realized an ultra-bright QD SPS by using a frequency-doubled ML-VECSEL operating at 494 MHz pulse repetition rate to excite a single QD deterministically integrated within a monolithically fabricated microlens. This compact and long-term-stable laser system allows us to overcome the repetition rate limit of standard commercial ML Ti:sapphire lasers and to achieve record-high single-photon fluxes of (143 ± 16) MHz collected by the first lens of the setup, which corresponds to a photon extraction efficiency of $(29 \pm 3)\%$. Probing the photon statistics of the emission of a single QD-state at saturation, we demonstrate triggered single-photon emission of the microlens with $g^{(2)}(0) < 0.03$. This first proof of principle experiment carried out under above-band excitation points out the high potential of our approach, which can be extended in the future towards resonant excitation of QD-microlenses or other QD based nanophotonic devices. This could be accomplished by deterministically matching the wavelength of respective QD transitions to the emission of a ML-VECSEL, or by exploiting wavelength-tunable ML-VECSELs in the future. In this light, even the generation of indistinguishable photons at unprecedented emission rates seems to be feasible. Further improvements towards practical SPSs might include spectral tuning via piezoelectric actuators[30] or the integration of microlenses onto arrays of site-controlled QDs.[31,32] Moreover, it would be interesting to combine our approach with materials providing stable room-temperature operation,[33,34] or alternatively by utilizing compact table-top cryocoolers.[26]

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