

Separating cascaded photons from a single quantum dot: Demonstration of multiplexed quantum cryptography

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We use cascaded two-photon emission from a single quantum dot in order to double the efficient transmission rate in a quantum key distribution protocol. The energetically nondegenerate photons are separated with a single-photon add/drop filter based on a Michelson interferometer. Time correlation measurements are discussed and compared with numerical simulations.

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Efficient generation of single photons is an important task for modern quantum technology applications such as quantum computation¹ and quantum cryptography.² Recently various single-photon sources based on single quantum emitters such as atoms,^{3,4} molecules,⁵ diamond defect centers,^{6,7} or quantum dots^{8–11} have been realized. Quantum key distribution using the BB84 protocol¹² has been demonstrated using single photons from nitrogen vacancy defect centers in diamond¹³ or from self-organized semiconductor quantum dots.¹⁴ In this application the demands on the single photon source are optical stability, high efficiency, and high emission rate. The overall efficiency can be improved by using nonresonant techniques such as solid immersion lenses¹⁵ or mirrors¹⁰ to enhance the optical collection efficiency or by resonant techniques where the quantum emitters are embedded in microcavities.^{16,17} The latter method exploits the Purcell effect¹⁸ in order to enhance the emission rate in a certain well-defined resonant cavity mode. The Purcell effect can also substantially modify the overall spontaneous emission rate. In quantum cryptography with single photon sources which rely on the decay of an excited state the (modified) spontaneous emission rate determines the maximum transmission rate of quantum bits. In this paper we propose and demonstrate a method to further enhance the effective transmission rate by using multiplexing similar as in classical communication channels.

We exploit an interesting feature of quantum dots which is the occurrence of radiative decay cascades of multiparticle states. In such a cascade a sequence of photons with (in general) different wavelengths is spontaneously emitted. As an example the biexciton (ground state of two confined electrons and two holes), decays via an exciton (single electron-hole pair) eventually to the empty quantum dot state. The energy of the photons from the decay of the biexciton and exciton, respectively, differ by the additional binding energy of the biexciton. This energy is about 4 meV in III-V quantum dots.¹⁹ Biexcitons and even higher excitations are easily excited with an efficiency close to unity.^{20–22} Since the decay of a biexciton prepares the system in the excitonic state it is most likely to find a photon with the exciton energy immediately after the detection of a photon with the biexciton energy. This classical correlation can be used, for example, to identify individual lines in the luminescence spectrum of a quantum dot.²² The emission of the two cascaded photons in

the decay of a single biexciton is thus equivalent to the emission of single photons from two classically correlated single photon sources. If these two photons can be separated, quantum information can be encoded independently on each of the photons. This is the principle of multiplexing which we demonstrate in the following.

A separation of the two photons is in general possible due to their different wavelengths. One way is to use diffractive or refractive optics. These techniques are unfavorable especially in inhomogeneously broadened systems like a sample of self-organized quantum dots. Here the wavelength as well as the wavelength difference between the two photons may vary from quantum dot to quantum dot. This would require a complete realignment of the beam paths for each individual quantum dot under consideration. Moreover, diffractive optics suffer from losses due to diffraction into different orders.

A superior method is to use interferometric techniques, like the one sketched in the upper part of Fig. 1(a). The two photons with different wavelengths λ and $\lambda + \Delta\lambda$ enter a Michelson interferometer with variable arm length. As long as the path difference between the two interferometer arms Δs is significantly smaller than the coherence length s_{coh} , the probability to find a photon at one of the two interferometer outputs ports is $I_{1,2} = \frac{1}{2} \pm \frac{1}{2} \cos(2\pi\Delta s/\lambda)$ (where + and – cor-

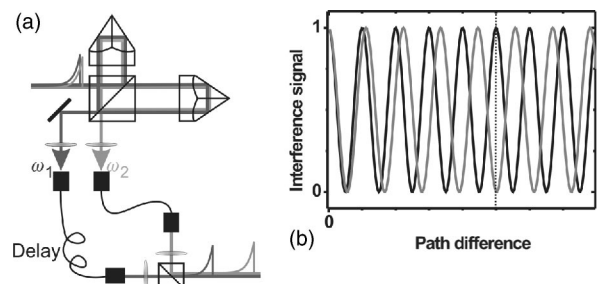


FIG. 1. (a) Sketch of the experimental setup: two photons with different energies enter a Michelson interferometer that consists of a 50:50 beamsplitter and two retroreflectors. The two interferometer output ports are each coupled to an optical fiber, delayed relative to each other with half of the excitation laser repetition time and recombined again at another beamsplitter. (b) Schematics of the intensity interference pattern at one interferometer output for two example wavelengths versus the path difference between the two interferometer arms.

respond to the two ports labeled 1 and 2, respectively). In order to illustrate how this can be used to separate the two photons we plotted the intensity interference pattern $I_1(\Delta s)$ for two different wavelengths in Fig. 1(b). At a certain path difference (position indicated by the dotted line) the two interference patterns are in opposite phase, i.e., each photon interferes constructively at a different output port. As long as $\lambda^2/\Delta\lambda \ll s_{\text{coh}}$, such a situation can always be achieved. Note that this condition simply reflects the spectral distinguishability of the two photons. The smallest path difference for which such a wavelength separation occurs is $\Delta s_0 = \lambda(\lambda + \Delta\lambda)/(2\Delta\lambda) \approx \lambda^2/(2\Delta\lambda)$.

Once this situation is established the two interferometer outputs can be used as two independent single-photon sources. Additionally, each photon can be coupled into an optical fiber, where an arbitrary time delay between the photons can be adjusted. This is also sketched in Fig. 1(a). The result is a train of single photons with twice the rate of the excitation laser. In order to avoid losses at the recombining beamsplitter one could also use a second symmetric Michelson arrangement, so that both partial beams are fully recombined in the same beam path.

In this experiment we used a single-photon source based on single Stranski-Krastanow grown InP quantum dots embedded in a GaInP matrix. The production process and characterization of the sample is described elsewhere.¹⁰ The sample was cooled in a liquid-helium cryostat to 4 K. A frequency doubled titanium-sapphire laser at 396 nm producing 150-fs pulses was used as the excitation source. The emission in the region of 675 nm was collected using a cover-glass corrected objective with 0.75 numerical aperture, providing a spatial resolution of approximately 0.5 μm . Imaging through a pinhole isolated a single quantum dot spatially. Figure 2(a) shows a spectrum of the emission from a single quantum dot under an excitation power of 30 pW/ μm^2 . The excitonic (X) and biexcitonic (X2) emission is clearly distinguished. The assignment to the biexciton and exciton was done by looking on the excitation power dependency of the lines. In order to prove single-photon emission we performed intensity correlation measurements in a Hanbury Brown and Twiss configuration. Two commercial silicon avalanche photodiodes were used, and correlation events were collected into 37-ps time bins. The time resolution of this system was measured to be 800 ps. Correlation measurements under pulsed excitation for both the exciton and biexciton emission are shown as insets in Fig. 2. The number of correlation counts plotted in this figure is proportional to the second-order coherence function $g^{(2)}(\tau)$. The distance between the peaks is 13.2 ns which reflects the time separation of the laser pulses from the 76-MHz mode-locked laser. A clear suppression of the peak around zero time delay is observed. The area of this peak is only 0.17 of the other peaks, which demonstrates the antibunched nature of light from a single quantum dot. A finite background from scattered light prevented this peak from being completely suppressed.

The separation of the two photons from the cascaded emission is now obtained when the Michelson interferometer is inserted in the beam path. After adjusting the arm of the

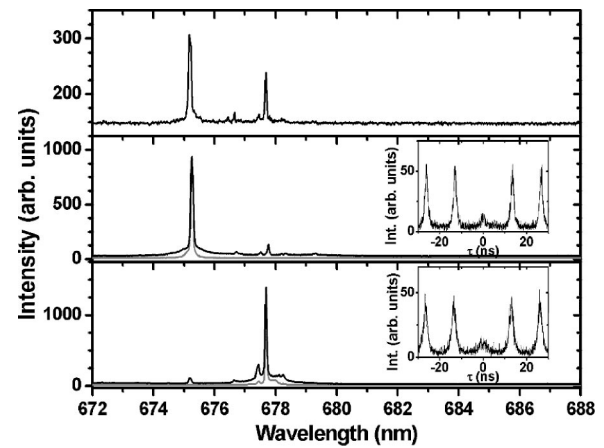


FIG. 2. (a) Unfiltered spectrum of the quantum dot emission. (b) Spectrum of the light emerging from one Michelson output port. (c) The same as in (b) but taken at the other output port. Insets show correlation measurements of exciton and biexciton emission taken after each output port. The gray curves have been obtained after filtering with a bandpass filter.

Michelson interferometer according to the line separation of $\Delta\lambda = 2.5$ nm, the two spectral lines are directed separately into the two output ports. Figure 2(b) shows the spectrum taken right after one of the two output ports: Only the excitonic emission is present while the biexcitonic emission is completely suppressed. In the other port the situation is reversed as can be seen in Fig. 2(c). With this the exciton and biexciton can be suppressed in the according port by 90.5% and 91.4%, respectively. The total transmission of each separated lines was 77%, where the main losses result from reflections from the uncoated optical surfaces. Additional spectral filtering using a 0.5-nm (full width at half maximum) bandpass filter behind each output was then performed to completely suppress background and excitation light [gray curves in Figs. 2(b) and 2(c)].

The light from the two output ports was now coupled into two multimode fibers where the different lengths of the fibers provided a relative time delay of 6.6 ns. The total coupling loss was 40%. Figure 3 shows correlation measurements of the light from the two fibers separately [(a) and (b)] as well as of the light from the two fibers together [Fig. 3(c)]. The excitation powers are set as in the measurements displayed in Fig. 2, so that the exciton and biexciton transitions are similarly strong. Again a clear reduction of the correlations around the time origin to a fraction of 0.2–0.4 relative to the neighboring peaks is observed in (a) and (b) similar as in the insets in Fig. 2. The distance between the peaks in (a) and (b) is 13.2 ns and reflects again the time separation of the 76-MHz laser pulses. In contrast to this Fig. 3(c) shows a peak-to-peak distance of only 6.6 ns which means that the photon emission rate is doubled. Still a clear antibunching is visible.

The figure also demonstrates nicely the limit of the emission rate of a single-photon source based on spontaneous emission. As the photo detectors cannot distinguish between the energy of the two photons, the same correlation measurement would have been obtained if single photons from only

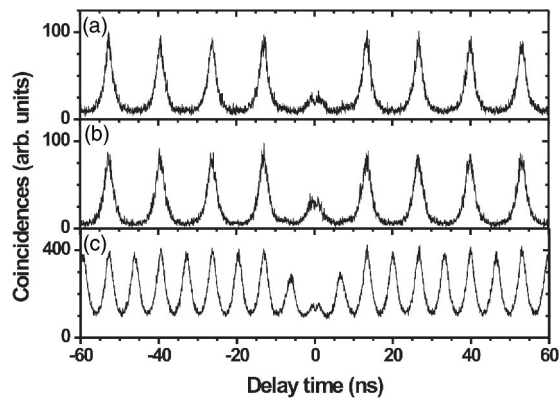


FIG. 3. Intensity correlation of the excitonic (a) and biexcitonic (b) spectral line after coupling to a fiber. Both single-line correlations were obtained by blocking one of the interferometer arms. (c) Intensity correlation function of the multiplexed signal.

the excitonic transitions had been recorded, but with a doubled excitation rate. In both situations the time period between excitation events approaches the spontaneous lifetime of the dots which is for the excitonic and biexcitonic transition on the order of 1 ns as reflected by the width of the peaks. Thus the peaks start to overlap and form a constant background which produces the offset in Fig. 3(c). However, we want to stress that in our experiment we can use the light from both fibers independently (e.g., also with a relative time delay of zero) and thus indeed double a possibly transmission rate in a quantum cryptography experiment.

Another interesting feature in Fig. 3(c) is the varying height of the peaks in the correlation measurement at higher delay times τ . Similar modulations of the peak height have been observed also in a conventional single photon source, where only one transition is used.²³ This effect has been attributed to different probabilities to populate the excitonic state from a charged or an uncharged quantum dot in case of off-resonant and resonant optical excitation, respectively. In our experiment the correlations between adjacent correlation peaks are more complex since a start-stop event may be caused by either a biexcitonic photon followed by a delayed excitonic photon (delay of half of the laser repetition time of 6.6 ns) or vice versa. After detection of the first photon the quantum dot contains an uncharged exciton or it is empty. In both cases this influences the probability to detect the second photon. In order to study this we have performed Monte Carlo simulations that are based on a cascade model similar as in Ref. 23, but including the delayed correlation measurement technique used in our experiment. The details of the model go beyond the scope of this paper and will be published elsewhere.²⁴ Figure 4 displays two typical results from this simulation. The decay rates that are used are $0.71 \times 10^9 \text{ s}^{-1}$ and $0.83 \times 10^9 \text{ s}^{-1}$ for the biexciton and exciton decay, respectively, and are taken from fits to the autocorrelation measurements of these transitions. When exciting above the barrier band gap free charges are excited and trapped into the quantum dot creating both charged and uncharged excitons and biexcitons. Figure 4 is a numerical simulation for correlations between unequally (a) and equally (b) charged transitions, respectively.

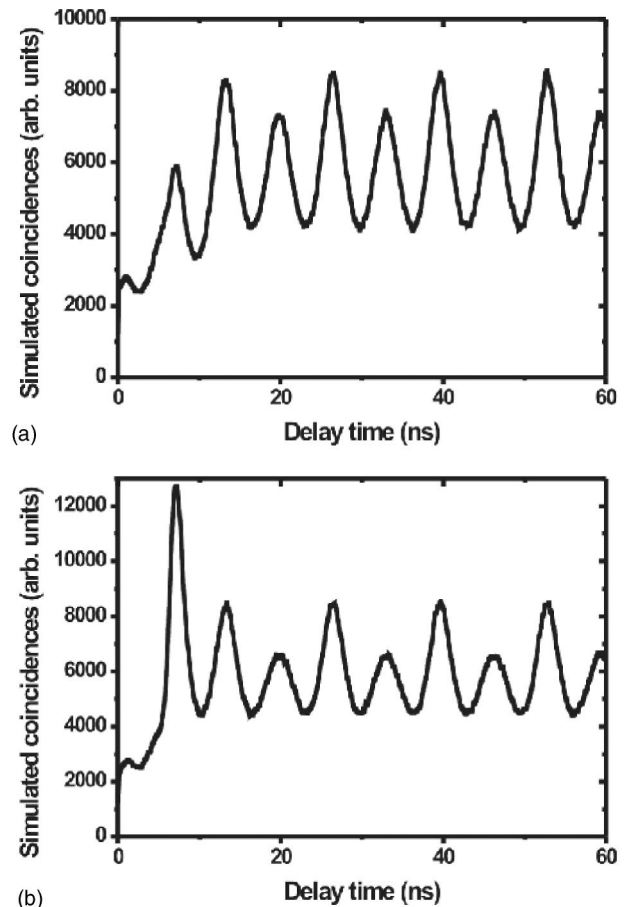


FIG. 4. Results obtained from a Monte Carlo simulation: (a) for a combination of differently and (b) equally charged exciton and biexciton states.

An important application of cascaded photon generation and subsequent separation is multiplexing in single-photon quantum cryptography. In the BB84 protocol¹²⁻¹⁴ information is encoded in the polarization state of the transmitted photons. If there is no correlation between the encoded polarization and the energy of adjacent photons then the energy of the photons is irrelevant. A possible implementation of multiplexing into the BB84 protocol is shown in Fig. 5(a). Following the common notation sender and receiver are called Alice and Bob, respectively. The Michelson interferometer acts as an add/drop filter for single photons, where the two photons are separated. When the quantum dot emission is unpolarized, additional polarizers have to prepare the photons in a well defined initial state. Two electro-optical modulators (EOM's) randomly encode a certain polarization state on each photon individually. At the latest here any polarization correlation between the two photons is destroyed, thus providing independent qubits. The photon streams are recombined in an additional Michelson interferometer and sent via a common transmission line. At the detection the photons are separated and independently detected in randomly chosen bases. In this way the transmission rate will be doubled.

To demonstrate this application a simplified proof-of-principle experiment was set up [Fig. 5(b)]. In this setup the

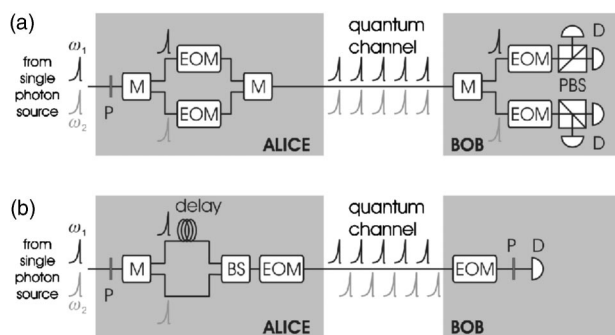


FIG. 5. Possible implementations of the multiplexer into the BB84 protocol. In both schemes the polarization is modulated between straight and circular polarization using an polarizer P with an electro-optical modulator (EOM). Bob's detection side is realized by another EOM, a polarizing beamsplitter PBS, and detectors D. ω_1 and ω_2 indicate the two energies of the photons and M is the Michelson arrangement. In (a) the two photons are recombined without time delay, in (b) a time delay is introduced and the two beams are then recombined with a beamsplitter BS, like it is done in the experiment.

excitonic and biexcitonic photons from the micro-PL setup were sent to Alice, where they were separated, fiber-coupled, and delayed. As a simplification we replaced the second add/drop Michelson (which combines the photons again) by a 50:50 beamsplitter. With this we doubled the photon pulse rate, but reduced the average photon number per pulse. Additionally, Bob's detection consisted of a second EOM, an analyzing polarizer and an APD, where the EOM was randomly switched between the two bases. Since only in one-fourth of the cases Bob measures in the same state as Alice, this results in a reduction of the effective count rate of 50% compared to a scheme with two detectors. In this experiment we used a pulsed diode laser ($\lambda=635$ nm, pulse width 125 ps) with a repetition rate of 10 kHz, which was adopted to the modulation rate of the EOM driver. In this configuration two-photon events can create a possible insecurity, but in our setup the collection efficiency is estimated to be $p \approx 10^{-3}$, the probability to collect two adjacent photons is $p_{11}=p^2 \approx 10^{-6}$ and thus much smaller than the probability to collect one and lose the next, $p_{10}=p(1-p) \approx 10^{-3}$. At higher

collection efficiencies the setup in Fig. 5(a) is favored.

Due to these simplifications the achieved transmission rates are smaller than in the optimized configuration from Fig. 5(a). Nevertheless, it successfully demonstrates a doubling of the transmission rate with respect to the same setup without multiplexer. Altogether we work with the following parameters: After an additional electronic gating of Bob's detector the rate of usefully exchanged photons is found to be 30 s^{-1} , whereas the dark count rate is reduced to 0.75 s^{-1} . The probability to transmit photons through the two EOM's with crossed polarizations was measured to be 6.8%. After comparing Alice's and Bob's keys an error rate of 6% was found. With these parameters the number of secure bits per pulse after Lütkenhaus²⁵ is 5×10^{-4} .

The Michelson add/drop could find applications in linear optical quantum computation (LOQC).¹ Since gates in LOQC have only limited success probabilities, parallel processing may increase the efficiencies of the gate or at least improves the statistical significance of a computational result. Multiphoton decay cascades in single quantum dots have also been proposed as a possible source for entangled photon pairs.²⁶ Here the photons are polarization entangled and in general not energetically degenerated. The method which we demonstrated allows the spatial separation of the two photons without destroying their entanglement, so that they can be used subsequently in a multitude of experiments and applications.

In conclusion we proposed to use multiphoton cascades as multiple single-photon sources. We realized this idea by establishing an add/drop filter based on a Michelson interferometer for separating the photons from the biexciton-exciton cascade in a single quantum dot. By recombining the delayed photons we achieved a doubling of the effective single-photon emission rate. As a proof-of-principle we implemented this in a BB84 quantum cryptography experiment.

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