Deterministic Photon Pairs and Coherent Optical Control of a Single Quantum Dot

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The strong confinement of semiconductor excitons in a quantum dot gives rise to atomlike behavior. The full benefit of such a structure is best observed in resonant excitation where the excited state can be deterministically populated and coherently manipulated. Because of the large refractive index and device geometry it remains challenging to observe resonantly excited emission that is free from laser scattering in III/V self-assembled quantum dots. Here we exploit the biexciton binding energy to create an extremely clean single photon source via two-photon resonant excitation of an InAs/GaAs quantum dot. We observe complete suppression of the excitation laser and multiphoton emissions. Additionally, we perform full coherent control of the ground-biexciton state qubit and observe an extended coherence time using an all-optical echo technique. The deterministic coherent photon pair creation makes this system suitable for the generation of time-bin entanglement and experiments on the interaction of photons from dissimilar sources.

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Deterministic single photon sources exhibit the property of emitting one and only one photon with a very high probability at a desired time. Nonclassical light sources like single photon and entangled photon pair sources are needed for linear optical quantum computation [1], long-distance quantum communication [2], and protocols like teleportation [3] or entanglement swapping [4]. Such sources have been demonstrated in stimulated emission of cavity-coupled atoms [5] or heralded down-conversion sources [6,7]. Quantum dots are proven sources of single photons, cascaded photons, and entangled [8] photon pairs. To be used for quantum information processing, resonant excitation [9,10] and coherent manipulation [11] are essential.

Quantum dots can be used as a source of coherently created photon pairs if they are excited through two-photon resonant excitation of the biexciton [12]. Owing to the difficulty of separating the excitation laser light from the emission, so far biexciton emission through two-photon excitation has only been shown in II/VI quantum dots [13], characterized with an order of magnitude higher biexciton binding energy. Nevertheless, in these experiments the emission was not collected and the statistics of the photons were not characterized. Furthermore, II/VI systems are characterized by emission of visible photons that are less suitable for quantum communication applications compared to III/V quantum dots, which emit in the IR wavelength range. In III/V systems, only photocurrent measurements [14–16] were performed following the two-photon excitation. In contrast, in the present work, we resonantly excited a III/V quantum dot system and through a combination of techniques we collected single photons and measured their statistics with high efficiency and virtually no background as shown in Fig. 1.

We present results obtained through resonant two-photon excitation [13–15] of the biexciton state of a single InAs/GaAs quantum dot embedded in a microcavity. In particular, we used pulsed laser light (4 ps) to coherently excite a single quantum dot using the lateral wave-guiding mode of a planar microcavity [17] [Fig. 2(b)]. The two-photon excitation was performed via a virtual level half way in energy between the exciton and biexciton [Fig. 2(a)]. Additionally, we performed all-optical full coherent control of the ground-biexciton state superposition. Further, to our knowledge for the first time, we applied an interferometric picosecond sequence of pulses to perform a spin echo measurement on the ground-biexciton qubit and through this recovered the coherence of the qubit (T_2 time). The coherent manipulation of such a ground-biexciton qubit is essential for state preparation. Entanglement schemes like time-bin entanglement require such state preparation, which encodes the entanglement in the system [18]. Using time-bin schemes to create entangled photon pairs from a quantum dot would combine robustness of the time-bin entanglement [19,20] with the state purity of the quantum dots [21]. In addition time-bin entanglement is not affected by the fine structure splitting of quantum dots, the major obstacle for high quality polarization entanglement.

Our sample contained self-assembled InAs quantum dots of low density ($10\ \mu m^{-2}$) grown by molecular beam epitaxy. The quantum dots were embedded in a 4λ distributed Bragg reflector microcavity with a cavity mode at $\lambda = 920$ nm. The sample was kept in a helium flow-cryostat temperature stabilized to 5.0 ± 0.1 K.

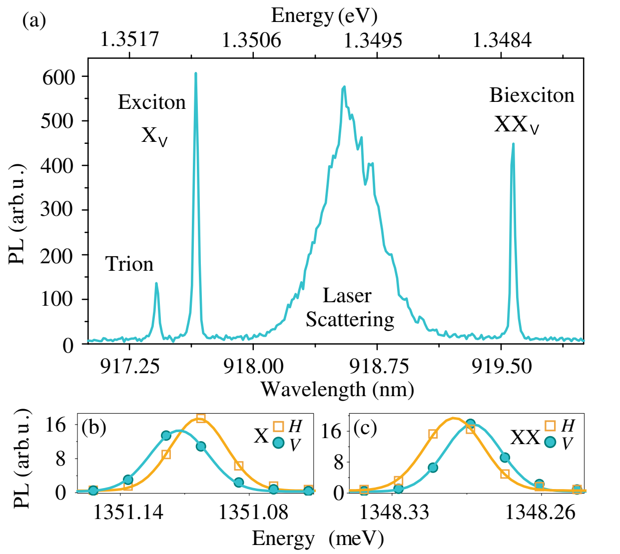

FIG. 1 (color online). Photoluminescence under resonant two-photon excitation. (a) Photoluminescence spectra of the V-polarized cascade and a suppressed H-polarized excitation laser. (b), (c) Emission from V and H-polarized cascades under H-polarized excitation. Solid lines are Gaussian fits to the data showing the fine structure splitting of the exciton states.

The excitation pulses were derived from a 76 MHz Ti: sapphire laser. To spectrally limit the scattered laser light, the pulse length was conveniently adjusted by a pulse shaper, which consisted of two diffraction gratings and a slit placed in-between them [Fig. 2(b)]. The excitation light was focused onto the sample from the side. Here the distributed Bragg reflector structure of the sample acted as a waveguide for the excitation laser. The emission was collected from the top (orthogonal to the excitation plane) using a microscope objective. The emission (biexciton and exciton photons) was spectrally separated in a home-built spectrometer and coupled into single mode fibers.

The levels involved are the ground ($|g\rangle$), exciton state ($|x\rangle$), and biexciton state ($|b\rangle$). The level scheme is shown in Fig. 2(a). The energy differences between the ground state and exciton state, and between the exciton state and biexciton state are not equal due to the increased biexciton binding energy E_b with respect to the single exciton. This electronic configuration allows for a two-photon excitation process where the pump laser is not resonant to any of the single photon transitions, while the two-photon process is resonant. The Hamiltonian used to describe this system is given as

$$H = \frac{\hbar\Omega_l(t)}{2}(\sigma_{gx} + \sigma_{gx}^\dagger) + \frac{\hbar\Omega_l(t)}{2}(\sigma_{xb} + \sigma_{xb}^\dagger) + \hbar\sigma_{xx}(\Delta_e - \Delta_b) - 2\hbar\sigma_{bb}\Delta_b. \quad (1)$$

Here, $\Omega_l(t)$ is the Rabi frequency of the pump laser driving both single photon transitions. The transition operators or projectors are given as $\sigma_{(i,j)} = |i\rangle\langle j|$. The detuning, $\Delta_e = E_b/2\hbar = 2\pi \times 335\,\text{GHz}$, between the two-photon transition virtual level and the exciton energy. To drive the two-photon transition off-resonantly we define the detuning (Δ_b), the difference between the two-photon resonance and the driving laser energy.

The emission probability of a resonantly driven system shows an oscillatory behavior as a function of the excitation pulse area known as Rabi oscillations. In our measurements we confirm the two-photon resonant excitation and the coherent nature of the excitation of the two-level system by observing an oscillation in emission power as a

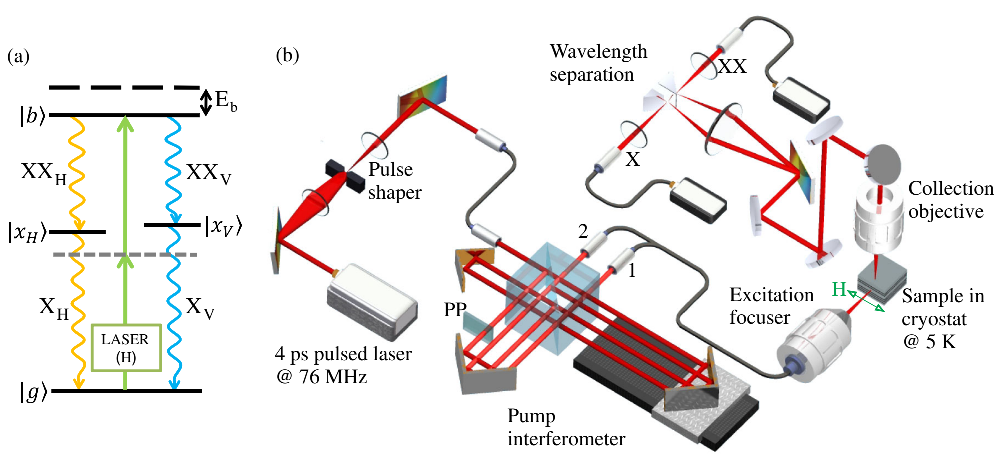

FIG. 2 (color online). Excitation-emission scheme and experimental setup. (a) Energy level scheme. A pulsed laser with energy $E_{\text{exc}} = (|b\rangle - |g\rangle)/2$ and linearly polarized (H) along the cleaved edge of the sample, coherently couples the ground ($|g\rangle$) and the biexciton ($|b\rangle$) states through a virtual level (dashed gray line). Biexciton recombination takes place through the intermediate exciton states ($|x_{H,V}\rangle$) emitting biexciton ($XX_{H,V}$) and exciton ($X_{H,V}$) photons. Biexciton binding energy (E_b) results in $(|x_{H,V}\rangle - |g\rangle) > E_{\text{exc}} > (|b\rangle - |x_{H,V}\rangle)$. (b) Experimental setup: consists of excitation, collection, and detection part. Pump interferometer output fiber couplers labeled 1 and 2 collect double pulses and triple pulses, respectively, for coherent control experiments. A glass phase plate (PP) on the short arm of the interferometer controls the intensity of the echo pulse.

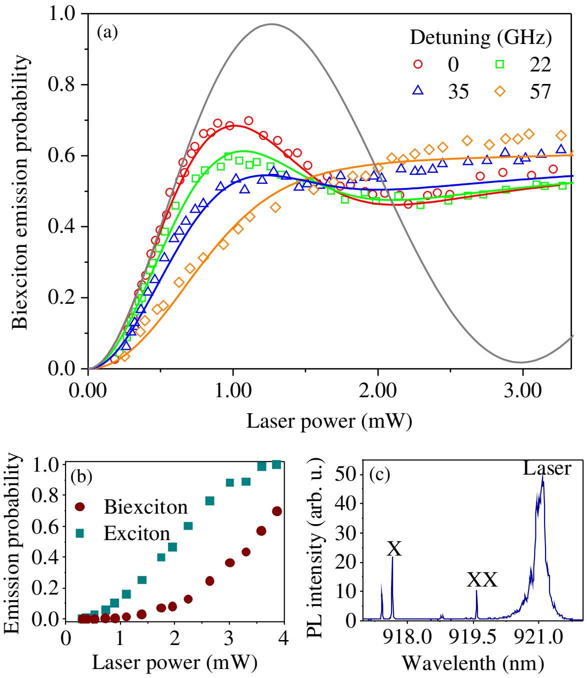

FIG. 3 (color online). (a) Power dependence of the biexciton emission probability for various excitation laser detunings $[E_{\rm exc} - (|b\rangle - |g\rangle)/2]/\hbar$ from the two-photon resonance. Solid lines are simulations. (b) Power dependence of biexciton and exciton photons under incoherent two-photon excitation, far detuned from two-photon biexciton state resonance towards lower energy. (c) Photoluminescence spectra from the 3 mW measurement point of (b).

function of laser power [22–24] [Fig. 3(a)]. In the graphs the emission intensities are all normalized by the same numerical factor to the theoretically predicted values of the emission probability (see Supplemental Material [25]), which are plotted as solid lines.

The Rabi oscillations result from an exchange of the population between the ground state and the biexciton state; however, the limited number of oscillations in Fig. 3(a) is the result of decoherence. Any process that transfers the population to another state will destroy the coherence in the population exchange and therefore damp the Rabi oscillations. An obvious possibility could be the spontaneous radiative decay from the biexciton to the exciton level whose lifetime we have measured to be 405 ps. Nevertheless we are not affected by this kind of dephasing because we use laser pulses that are 2 orders of magnitude shorter.

A second possibility is the damping of the Rabi oscillations due to the proximity of the virtual level to the exciton state (detuned by $\Delta_e = 2\pi \times 335\,\text{GHz}$ or 1 meV). The gray theory line in Fig. 3(a) shows that this is a very minor effect. A third possible dephasing process could be based on interaction with lattice phonons whose energies ($k_B T \sim 400\ \mu$eV) could transfer the population from the virtual two-photon resonance to the exciton state [26]. However, at sufficiently high detuning this process should cease due to insufficient energy of the lattice phonons. In our case the virtual level is approximately $5k_B T$ away from the exciton line, which significantly reduces the probability for this process.

Finally the process that can best explain the damping of the Rabi oscillations in our data originates from competing nonresonant two-photon excitation processes that involve the quantum dot, for example, the creation of an electron-hole pair with one carrier trapped in the dot and one free in the host material. Evidence for this explanation comes from observing exciton and biexciton emission with the excitation laser red detuned with respect to the virtual level [Fig. 3(c)]. In agreement with this proposed mechanism of an incoherent two-photon process we find that this emission has a superlinear population power dependence [Fig. 3(b)], which indicates a nonlinear excitation process. We do not exclude the existence of a two-photon excitation in the surrounding material, but this process would not cause the damping of the Rabi oscillation but rather a background in the photoluminescence signal. The theoretical curves [Fig. 3(a)] include the dephasing caused by incoherent two-photon processes and fit the data very well for all values of the detuning of the excitation laser to the two-photon resonance.

The resonantly created photon pairs emitted in the cascade allow us, like in the case of spontaneous parametric down-conversion, to use the ratio of the coincidental detections of both cascaded photons to the number of single events to estimate the total detection efficiency (2.7‰). Using the detection efficiency we obtain a rate of created excitations or photon pairs of 18 MHz at the maximum of the highest Rabi oscillation (π-pulse). This reduction in the rate of excitations from the 76 MHz laser repetition rate can be attributed to two sources: decoherence of the excitation process observable in the damping of the Rabi oscillation and blinking [27] of the quantum dot, which is evident in the autocorrelation measurement through the decreased correlation peaks at long delay times [Fig. 4]. Because of the resonant nature of the excitation process the measurements of the autocorrelations of both exciton and biexciton photons show full suppression of multiphoton events. Without subtraction of the background caused by the coincidence events between signal and the detector dark counts, the autocorrelation parameters are 0.012(1) for the exciton and 0.0314(4) for the biexciton. With background subtraction the autocorrelations of the exciton and biexciton are 0.0073(8) and 0.024(3) at zero delay. For this particular polarization we detected single photons with a rate of 24 kHz in a single mode fiber and photon pairs with a rate of 62 Hz. From our total detection efficiency (2.7‰) and the quantum efficiency of the detectors (20%) we can estimate the collection efficiency from the emitter to the single mode fiber coupled detectors to be 1.35%. We estimated single mode fiber coupling alone to be higher than 60%.

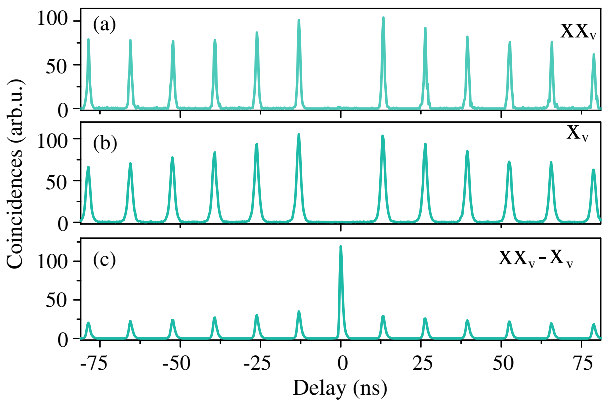

FIG. 4 (color online). (a), (b) Autocorrelation measurement of the V-polarized biexciton and exciton photons in resonant excitation. (c) Cross correlation coincidence counts between the biexciton (start) and exciton (stop) photons. All the data shown here are presented without background subtraction.

The coherence of the excitation process enables us to manipulate the phase of the ground-biexciton state superposition. To perform coherent control of this qubit we use a sequence of two consecutive pulses that are derived by feeding pulsed laser light into a Michelson interferometer [pump interferometer in Fig. 1(b)]. With this method we observe Ramsey interference fringes in both exciton (see Supplemental Material [25]) and biexciton [Fig. 5] emission. The power of the first pulse in the Ramsey sequence is adjusted to reach half of the maximal biexciton state population ($\pi/2$-pulse). The second pulse, delayed in time, can either map the population back to the ground state or further increase it to the biexciton state, depending on the relative phase between the pulses. When the time delay between the pulses is shorter than the pulse coherence we observe direct interference of the laser pulses [Fig. 5(b)]; when the delay exceeds the coherence length of the laser we observe interference originating from the atomic superposition. The observed visibility of the Ramsey fringes decays with 179 ps (T_2^*) for the biexciton [Fig. 5] and 182 ps (T_2^*) for the exciton (see Supplemental Material [25]).

A coherent system is Fourier transform limited with $T_2 = 2T_1$. When compared with the lifetimes (T_1) of the states of 405 ps (biexciton) and 771 ps (exciton) the Ramsey coherence times of the ground-biexciton qubit are shorter, and thus probably affected by phase noise. In addition, the contrast decays as a Gaussian exponential, which further indicates the presence of inhomogeneous broadening. To investigate the nature of the noise we performed a echo measurement. For this we used the Michelson interferometer in a double-pass configuration capable of delivering three consecutive pulses necessary for the spin-echo sequence ($\pi/2$, π, $\pi/2$). Here, the middle pulse was a result of interference of the first and second interferometer passes. Consequently, using a phase control we could adjust the middle pulse to have twice the intensity of the other pulses in the sequence ($\pi/2$-π-$\pi/2$ sequence). We controlled the coarse delays of the interferometer by a motorized linear stage and fine adjustments by a piezoelectric transducer.

We observed an increase in the coherence time for the biexciton ($T_2 = 375$ ps) and exciton ($T_2 = 388$ ps). The measured values along with the preserved Gaussian decay indicate the presence of high frequency noise, which could not be refocused by the spin echo technique. The initial interference visibility in the Ramsey measurement was 0.67 while the same visibility in the echo measurement was 0.4. We attribute this decrease as well as the Ramsey interference not being equal to unity to the damping processes explained above.

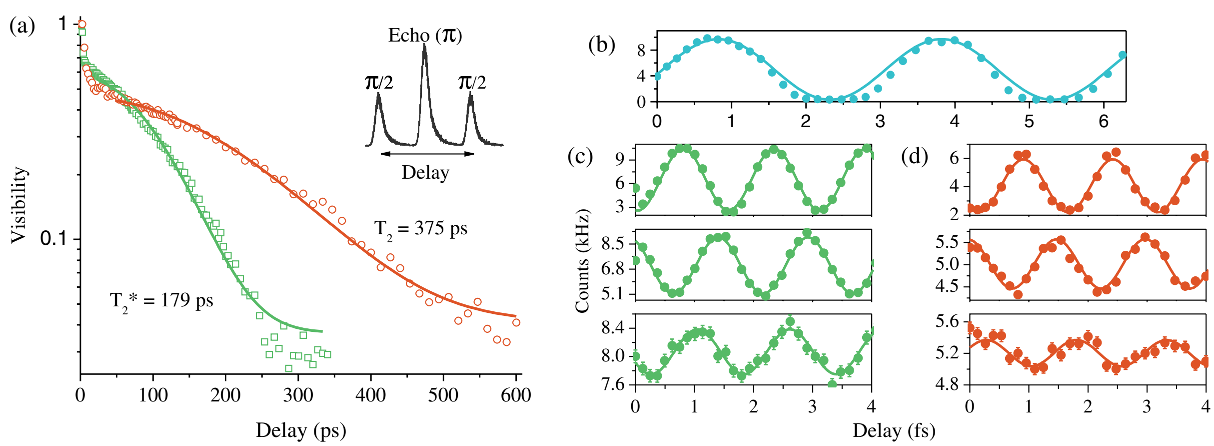

FIG. 5 (color online). (a) Visibility of Ramsey interference in the two-pulse (green open squares) and three-pulse echo sequence (red open circles) coherent control experiment monitored with the biexciton photons. (b) Laser interference at 0 ps coarse delay. (c) Ramsey interference fringes at coarse delays of 8, 107, and 240 ps. (d) Ramsey interference fringes after the echo pulse at coarse delays of 45, 339, and 472 ps. Both Ramsey and echo sequence measurements do not reach zero visibility due to the shot noise of $\sqrt{N}$ counts. Error bars for the shot noise are visible only for long delays.

To summarize, we have shown laser-scattering-free emission of coherent photon pairs created by pulsed, resonant, two-photon excitation. Using picosecond optical pulses we performed coherent manipulation of the phase of the ground-biexciton qubit monitored in photoluminescence with a visibility of 67%. The coherence time of the qubit was more than doubled by an echo sequence. Our photon statistics measurements prove the complete suppression of multiphoton emission events resulting in one of the cleanest single photon sources ever demonstrated [28–31]. Furthermore, the biexciton photons are created resonantly and the emission process does not involve spontaneous scattering of phonons, which makes the biexciton wave packet jitter-free. This property increases the indistinguishability [32] of the biexciton photon, which is an essential condition for the interaction of flying qubits and for linear optical quantum computation schemes. Deterministic creation of single photons and single photon pairs in combination with polarization entanglement [8] is a step forward in the realization of quantum information protocols with quantum dots. The possibility to coherently transfer the phase of the excitation light to the excited system makes the presented excitation scheme suitable for the creation of time-bin entanglement from quantum dots [18].

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