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Phonon-assisted robust and deterministic two-photon biexciton preparation in a quantum dot

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We investigate both experimentally and theoretically a simple yet more robust and flexible alternative to Rabi oscillation-type biexciton preparation protocols traditionally used for semiconductor quantum dots. The quantum dot is excited by a strong laser pulse positively detuned from the two-photon resonance yielding an on demand initialization of the biexciton state by making use of the phonon-induced thermalization of the photon dressed states. It is shown that for excitation pulses in the picosecond range, a stable and high occupation of up to $C_{XX} = 0.95 \pm 0.02$ is reached. Notably, the generated photons show similar coherence properties as measured in the resonant two-photon scheme. This protocol is a powerful tool for the control of complex solid state systems combining radiative cascades, entanglement, and resonant cavity modes.

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One strong advantage of atomiclike systems, especially semiconductor quantum dots (QD), in the development of quantum computing or communication devices is their ability to deliver on-demand single [1,2] or entangled photon pairs [3,4]. This first step towards the possibility of deterministic quantum operations is a crucial complement to the achievements obtained in the field of quantum information processing [5,6] as well as for tests of fundamental aspects of quantum mechanics [7]. The initial state is usually prepared through a population inversion thanks to a strong coherent pulsed laser field brought to resonance with the two-level system [8–10]. This Rabi oscillation protocol can be very efficient, but is strongly sensitive to fluctuations of the excitation parameters like the pulse area. Other more complex protocols, using chirped laser pulses to populate adiabatically the upper state, are in principle more robust [11], but the degree of population inversion realized in experiments devoted to the single exciton preparation [12,13] stayed below the ideal case. In contrast to real atomic systems, solid state systems experience a coupling to their environment, in particular with the surrounding crystal vibrational modes [14–18]. This has always been considered as a strong limitation to their efficient use because of the occurring decoherence. In particular, phonons have been identified as the cause for the nonideal state preparation using chirped pulse protocols [19–21]. However, it has been recently proposed for semiconductor quantum dots that, when addressed with a controlled off-resonant pulse, this weakness can become an advantage and make the state preparation more efficient, robust, and flexible [22]. The phonons cannot only be used to achieve an inversion in simple two-level systems consisting of the ground $|0\rangle$ and an exciton state $|X\rangle$ [23–25], but also for the initialization of the biexciton state $|XX\rangle$ that forms the upper level of a radiative cascade, which can potentially result in emission of entangled photon pairs [26,27].

In this paper, we investigate the biexciton preparation in a quantum dot through a controlled two-photon excitation scheme taking benefit of the normally undesired carrier-phonon coupling. By tuning the laser energy and pulse length, such that the relaxation processes due to this coupling are most efficient, it is shown that one can transit from a resonant Rabi oscillation regime to an adiabatic, efficient, and robust biexciton state initialization. The results for a sufficiently long pulse are compared to a situation where the pulse length is too short to complete the relaxation and the consequences on the state preparation are studied. The coherence of the resulting emitted photons is evaluated and reveals to be comparable in the Rabi π-pulse case and for the phonon-assisted protocol, adding to efficiency and robustness, a long coherence time. Accompanying our experiment, we have studied theoretically the dynamics of a quantum dot driven by a laser with frequency close to the two-photon resonance, and we find a good overall agreement to the predictions of our simulations. From this, we can conclude that the underlying mechanism of the state preparation is indeed the pure dephasing induced by longitudinal acoustic phonons [28], which we included in our model to account for the solid state environment of the quantum dot. We used an epitaxially grown (In,Ga)As/GaAs QD kept at a temperature of 4.2 K to investigate the photoluminescence under pulsed two-photon excitation. In order to address the biexciton in a resonant or a quasiresonant way, by setting the pulse length at a constant value and by controlling its wavelength, the laser is tailored through a pulse shaping setup. The quantum dot is excited from the side with linearly polarized light and the detection is done after a perpendicularly oriented polarizer in order to reject the scattered laser (for more information concerning sample structure and experimental setup see Ref. [3]). The pulse length as well as the pulse shape was controlled via intensity autocorrelation measurements. More details about the pulse shaping setup are given in Ref. [29].

Figure 1(a) shows a photoluminescence spectrum under above band-gap excitation. The exciton (1.4212 eV) and biexciton lines (1.4189 eV) are separated by the biexciton binding energy (2.3 meV). Direct excitation of the biexciton is obtained by setting a shaped laser in resonance between the exciton and the biexciton [see Fig. 1(b)]. Although the biexciton and the ground state are not directly dipole coupled, the dynamics induced by a resonant laser field results in Rabi-type oscillations where similar to a directly coupled two-level

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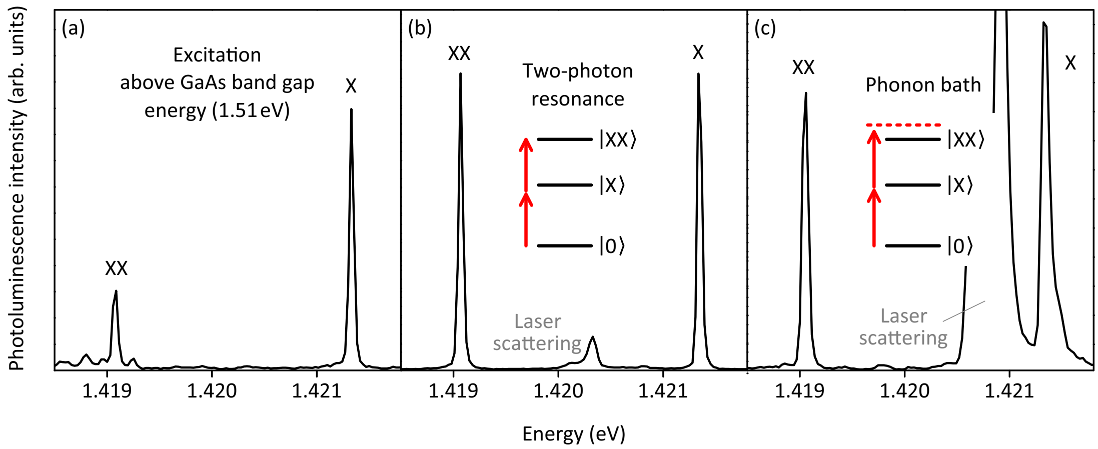

FIG. 1. (Color online) Single QD emission spectrum (a) under nonresonant above band gap excitation, (b) under resonant two-photon biexciton state excitation, and (c) with phonon-assisted 13-ps pulsed, 0.65-meV positively detuned excitation.

system the oscillation frequency scales with the square-root of the laser intensity [10]. Therefore the final inversion obtained after a pulse of finite length also oscillates between ± 1 as a function of the excitation power. These Rabi oscillations were probed with power dependent PL intensity measurements where the results for 13-ps pulses are shown as blue dots in Fig. 2(a). The solid lines are the result of a numerically exact real-time path-integral simulation [30], which allows us to calibrate the scaling of the measured intensity by fitting the theoretically predicted Rabi oscillations for resonant excitation to the corresponding experimental data. Because the size of the quantum dot and the strength of the chirp of the laser pulse where not directly measured, we treated these quantities as fitting parameters. We would like to note that the path-integral approach gives us the ability to solve our model of an optically driven quantum dot without further approximations taking into account arbitrary multiphonon processes as well as all non-Markovian effects. Further information about our model and other system parameters [31] used in the calculations are given in Ref. [32].

The first maximum of the blue curve for resonant excitation in Fig. 2(a) indicates the inversion of the biexciton population and the corresponding pulse will be referred to as a π pulse. At the π-pulse power, the $|XX\rangle$ occupation is estimated to be $C_{XX}^{\mathrm{res}} = 0.94 \pm 0.01$, whereby the error is calculated by means of a least square analysis using the experimental data and the theoretically calculated curves. Two other noticeable features of these oscillations are the decrease of the Rabi period with increasing pulse area and a damping of the amplitude. The first characteristic is a signature of the two-photon excitation process [10] and the damping is due to a coupling to phonons [33,34]. The average value of the oscillation is above 0.5 due to an imperfection of the excitation pulse, which is weakly chirped in the pulse shaping setup. For a positive sign of the chirp and low temperatures, such an increase of the biexciton population has already been predicted [35]. However, here it should be noted that according to our simulations the frequency sweep mostly affects the dynamics for resonant excitation, while for detuned laser pulses targeted in this paper the chirp only has a very small effect.

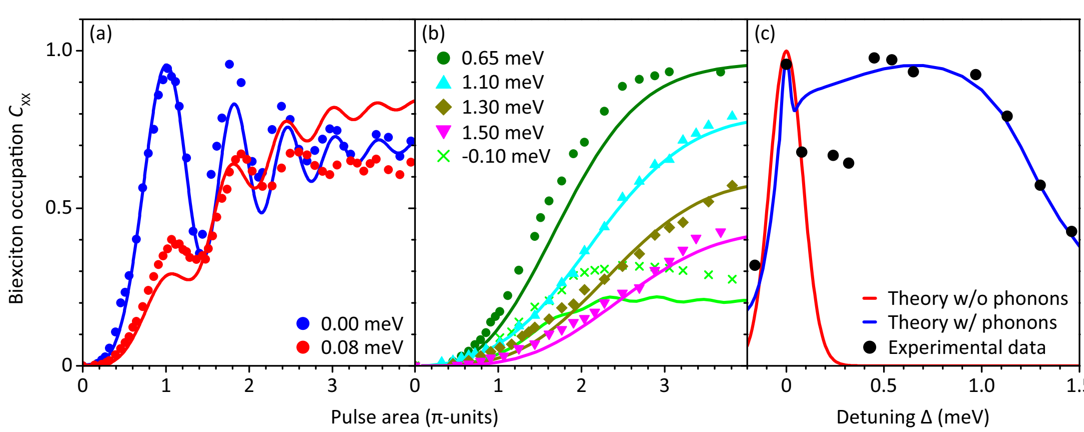

FIG. 2. (Color online) (a) Biexciton occupation C_{XX} as obtained from comparison with simulations (see text) vs renormalized pulse area with excitation pulse length of 13 ps superposed to path-integral simulation results for different laser detunings from the TPBR: in resonance represented in blue, 0.08-meV detuning in red, (b) 0.65-meV detuning in green, 1.1-meV detuning in light blue, 1.3-meV detuning in dark yellow, 1.5-meV detuning in magenta, -0.1-meV detuning in light green. (c) Maximum biexciton occupation (see text) reached for different laser detunings from the TPBR. The points represent occupations measured in the experiment and the full lines are results of the simulations (in blue with phonon coupling included in the calculations, in red without any phonon coupling).

Let us now focus on the case of off-resonant excitation of the QD exemplarily shown in the spectrum in Fig. 1(c) for 0.65-meV detuning. The laser detuning Δ is referred to the two-photon biexciton resonance (TPBR) and in our measurements ranges from -0.1 up to 1.5 meV. For such detuned excitation in a system with few isolated, discrete levels it is a well known result that the amplitude as well as the mean value of the Rabi oscillations rapidly decrease as the laser frequency is increased. In fact, for a small positive detuning of $\Delta = +0.08$ meV, shown in Fig. 2(a) in red color, reduced Rabi oscillations are still visible and when the detuning is increased, they disappear completely. However, as both our calculations and our experimental data show, there is also an overall increase of the biexciton population for positive detunings. This can be seen even more clearly in Fig. 2(b), which shows the power dependence of the biexciton population, for a few more selected detunings. The state initialization is most efficient between 0.45- and 1.00-meV detuning (only the 0.65-meV detuning data are shown in dark green), where the measured $|XX\rangle$ occupation at high-pulse area ($C_{XX}^{\text{det}} = 0.95 \pm 0.02$) is similar to the one reached with the resonant π pulse ($C_{XX}^{\text{res}} = 0.94 \pm 0.01$). In this range, the occupation is not only stable against small changes of the excitation frequency, but also shows a pronounced region where the biexciton population stays unaffected by fluctuations of the laser power, which is a clear advantage compared to the traditional resonant π-pulse scheme. For larger detunings (light blue for 1.1 meV, dark yellow for 1.3 meV, violet for 1.5 meV), the preparation becomes less efficient consistent with our calculations.

The detuning dependence of the biexciton occupation can be seen in more detail in Fig. 2(c), where we have plotted the maximum biexciton population measured in the laser power range between 0 π and 4 π (black dots) for the whole series together with the corresponding results of the simulations (blue line). Also shown are results of calculations where the exciton-phonon coupling was disregarded (red line). The strong influence of the environment visible from the discrepancy between the blue and the red line on the one hand, and the close overall agreement between the experimental data and the calculations including the phonon interaction on the other hand, provide clear evidence that the state preparation can be attributed to the carrier-phonon coupling. To understand the physics behind this feature, it is important to note that a phonon-induced relaxation is possible as the bare electronic states become dressed by the laser field. This relaxation can lead to a thermal occupation of the photon dressed states, which for positive detunings yields a high biexciton population [22,36]. For negative detunings [also shown in Fig. 2(b) in light green], the energetic order of the dressed states changes, and thus the biexciton state is no longer the final state of the relaxation at low temperatures, which explains the steep decrease of the biexciton population seen in Fig. 2(c) for $\Delta < 0$. Around the two-photon resonance, we can see a sharp peak in Fig. 2(c) as the maximum occupation in the observed pulse area interval is determined by the height of the π-pulse peak. When further increasing the laser frequency the $|XX\rangle$ population decreases until the transition from the resonant Rabi oscillation scheme to the off-resonant phonon-assisted state preparation occurs. In the

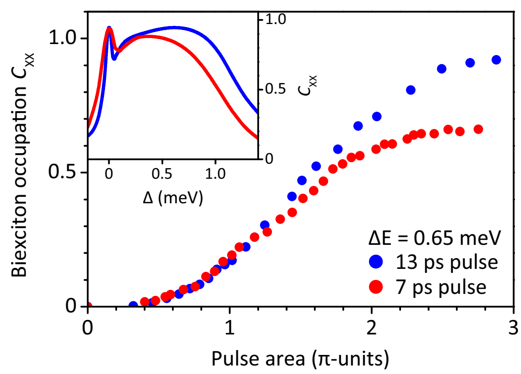

FIG. 3. (Color online) Measured biexciton population vs renormalized excitation power for excitation pulse length of 13 ps (blue dots) and 7 ps (red dots) at laser detuning of 0.65 meV. (Inset) Maximal biexciton occupation as a function of the detuning for excitation with an unchirped pulse of length 13 ps (blue) and 7 ps (red).

experiments, the population stays below the calculated values for detunings between 0.08 and 0.32 meV. While it is, so far, not clear why the quantitative agreement between theory and experiment is not as perfect as for detunings below and above this range, the qualitative behavior is still as expected. In particular, C_{XX} steeply decreases from the maximum at zero detuning, but always stays at a finite value well above what is predicted without phonons (cf. the red curve). Moreover, for higher detunings, a wide plateau is reached where the phonon-assisted relaxation is most efficient and therefore ideal for deterministic state initialization. The maximal efficiency of the phonon coupling in this region is due to the resonance of the most pronounced phonon energies to the transitions between the relevant dressed states. For even higher detunings, the splitting between the dressed states becomes too large and hence the phonon relaxation does not take place efficiently yielding lower and lower values for the maximal biexciton population as is nicely seen in both experiment and theory. It should be noted that the resonance structure described above has the same origin as the nonmonotonic dependence of the phonon induced damping of Rabi oscillations on the pulse area [34,37,38].

In order to test the influence of the excitation pulse length, the same experiment was carried out for shorter excitation pulses of 7-ps width. Figure 3 shows the power dependencies at $\Delta = 0.65$ meV detuning for excitation pulse widths of 13 ps (blue dots) and 7 ps (red triangles), respectively. The biexciton occupation at high pulse areas becomes much less efficient for the shorter excitation pulse (7 ps) and whatever the detuning used, the occupation of the biexciton obtained with short pulses stays below 0.62 and is therefore insufficient for schemes requiring photons on-demand (data are shown in Ref. [39]). The same tendencies are also found in the theory as seen from the inset of Fig. 3, where as in Fig. 2(c), but for an unchirped excitation, the detuning dependence of the maximal biexciton occupation is plotted for pulse durations of 13 ps (blue) and 7 ps (red), respectively. While the maximal attainable biexciton occupation is practically independent of the pulse length for resonant excitation, it is significantly

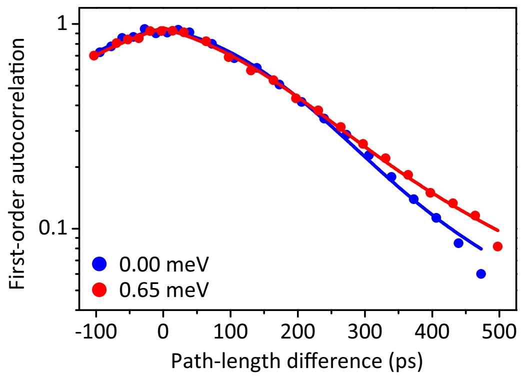

FIG. 4. (Color online) Biexcitonic photon first-order interference visibility vs time delay, under resonant π-pulse excitation (blue dots), under phonon-assisted, 0.65-meV detuned, 3π-pulse excitation (red dots). Full curves are fits of the Fourier transform of a Voigt profile to the experimental data from which the coherence times (277 ± 8 ps in resonance, 271 ± 7 ps for the 0.65-meV detuned excitation) are extracted.

reduced at larger positive detunings for the 7-ps pulse. The calculations support the conclusion that robust and efficient preparation can be obtained provided that the pulse is long enough to relax the system to the energetically lowest dressed state during the pulse, whereas under too short excitation no robust biexciton preparation can be achieved.

In order to check the usefulness of the protocol in quantum optics applications, we investigated the coherence of the generated photons and compared them to coherence times observed in resonance. Figure 4 displays the measured coherence time curves under 13-ps resonant π pulses (blue) and 0.65-meV detuned excitation (at large pulse area, i.e., around 3π, in red). For both excitation schemes, the first-order autocorrelation functions are nearly identical. The full curves are Fourier transforms of a Voigt profile from which we extracted a coherence time of 277 ± 8 and 271 ± 7 ps, respectively. This is in contrast with measurements made with above band-gap excitation pulses which resulted in significantly lower coherence times ($\tau = 114 \pm 4$ ps [3]), because of the electronic fluctuations generated in the surrounding of the quantum dots [40]. This conservation of the coherence obtained in resonance is of crucial importance since the coherence of the emitted photons is a decisive parameter determining their degree of indistinguishability [41,42].

In summary, it is demonstrated experimentally in this paper that one can obtain a robust biexciton preparation with near unity occupation probability and a long coherence time by using a simple protocol involving excitations detuned from the two-photon resonance. Comparing with theoretical results, we find a good agreement revealing that the preparation is due to phonon-induced relaxation processes. Applied to the optical preparation of biexcitons in quantum dots, this is a particularly flexible and efficient scheme for the initialization of entangled photon states. Since this protocol leaves the TPBR free from laser scattering, it is particularly suitable for a recently proposed two-photon emission ($|X\rangle$ and $|XX\rangle$) in a large Q-factor cavity mode set between $|X\rangle$ and $|XX\rangle$, which was demonstrated as highly entangled whatever the $|X\rangle$ fine structure splitting is [43]. The approach presented in this paper for the biexciton preparation would solve practical problems encountered in such two-photon emission observations in QD-photonic crystals systems [44] enabling clean photon statistic measurements.

Note added in proof. Recently, we became aware of related results by two other groups [45,46].

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