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# Coherent optoelectronics with single quantum dots

**A Zrenner[1], P Ester[1], S Michaelis de Vasconcellos[1], M C Hübner[1], L Lackmann[1], S Stufler[1] and M Bichler[2]**

[1] Universität Paderborn, Department Physik, Warburger Straße 100, D-33098 Paderborn, Germany
[2] Walter Schottky Institut, Technische Universität München, Am Coulombwall, D-85748 Garching, Germany

E-mail: zrenner@mail.upb.de

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**Abstract**
The optical properties of semiconductor quantum dots are in many respects similar to those of atoms. Since quantum dots can be defined by state-of-the-art semiconductor technologies, they exhibit long-term stability and allow for well-controlled and efficient interactions with both optical and electrical fields. Resonant ps excitation of single quantum dot photodiodes leads to new classes of coherent optoelectronic functions and devices, which exhibit precise state preparation, phase-sensitive optical manipulations and the control of quantum states by electrical fields.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

From a physical point of view, all currently available active key components for information technology rely on incoherent phenomena in view of the associated electronic states in a solid. The use of coherent phenomena for the implementation of quantum information technology is expected to give plenty of room for advanced developments in the future [1]. The basic building blocks within this technology are quantum bits (qubits), which are coherent quantum-mechanical two-level systems.

Within the current contribution we concentrate on low-dimensional semiconductor systems, where it has become possible during the last 15 years to handle and control single electrons, spins and excitons (see, for example, [2–7]). Furthermore we focus on excitons in semiconductor quantum dots (QDs), which will be treated as two-level systems throughout this contribution. Recently coherent population oscillations, so-called Rabi oscillations [8], have been demonstrated in the exciton [9–14] and bi-exciton [15] population of single QDs. Low temperature dephasing times for excitons in self-assembled QDs have been shown to exceed several hundred picoseconds, basically allowing for sufficiently high numbers of coherent manipulations with picosecond pulses, as required for applications in the field of quantum cryptography [16].

The experimental results presented in this contribution have been obtained from self-assembled $In_{0.5}Ga_{0.5}As$ QDs embedded in n-i Schottky diodes, which were grown by molecular beam epitaxy on a (100)-oriented $n^+$-GaAs substrate. The QDs are embedded in a 360 nm thick intrinsic GaAs layer, 40 nm above the n-doped GaAs back contact. A semitransparent Schottky contact is provided by a 5 nm thick titanium layer. The optical selection of a single QD is done by electron-beam-written shadow masks with apertures from 100 to 500 nm (see [11] for details). A schematic view of such a single QD photodiode is shown in figure 1(a). In this paper we report experimental results which have been obtained under the condition of resonant excitation from a Ti:sapphire laser of either the s-shell or the p-shell of a single QD at $T = 4.2$ K.

## 2. Coherent manipulation of the ground state

In detailed studies of the single exciton ground state by photocurrent (PC) spectroscopy, the ground state transition appears as an extremely narrow resonance with fully resolved fine structure splitting [17–19]. The observed width of the lines ($\sim 3.5\ \mu$eV) is lifetime-limited, resulting from a tunneling time of about 300 ps, which applies for the data shown in figure 1(b). Each of the fine structure split lines can be complementary selected or suppressed by proper orientation of the linear polarization of the resonant laser excitation.

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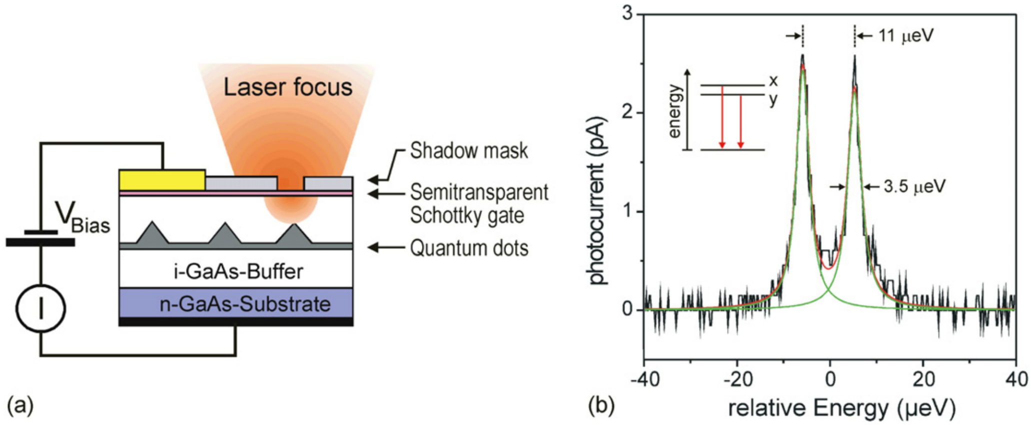

**Figure 1.** (a) Schematic view of a single QD photodiode. Optical access to a single QD is provided via a shadow mask and a semitransparent Schottky gate. (b) Photocurrent spectrum of the single exciton ground state with fully resolved fine structure splitting.

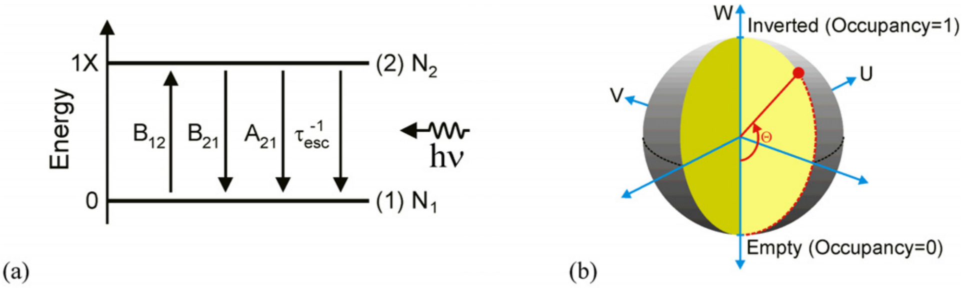

**Figure 2.** (a) Fundamental processes in an exciton two-level system at finite electric fields. In addition to the rates of absorption ($B_{12}$), stimulated emission ($B_{21}$) and spontaneous emission ($A_{21}$), the tunneling rate ($\tau_{\mathrm{esc}}^{-1}$) also has to be considered. (b) Bloch sphere representation of a resonant, coherent excitation in a dot. The rotation angle Θ is proportional to the pulse area and to the oscillator strength of the ground state transition.

As indicated in figure 2(a) we attempt to describe the ground state exciton transition as a two-level system in a semiconductor. Detailed knowledge and understanding of this system will be necessary if we plan to apply exciton two-level systems as qubits in future coherent devices for quantum information processing. The integration of QDs in the active region of a photodiode plays thereby an important role. The new parameter bias voltage ($V_{\mathrm{B}}$) allows, first of all, an electric measurement of the occupancy of a given QD. For internal electric fields beyond ∼35 kV $\mathrm{cm}^{-1}$ (for the InGaAs/GaAs system) excitons can be ionized, which corresponds to a state projection of the exciton by tunneling. The resulting single-particle states (electron and hole) can be easily detected as a PC signal in a quantitative way. At the same time, the diode arrangement allows us to tune the energy of an exciton by the bias voltage $V_{\mathrm{B}}$ (the Stark effect). A single QD photodiode can therefore be described as an exciton two-level system with a connection to an external electric circuit. With resonant ps laser excitation a single QD photodiode is therefore expected to perform as a coherent optoelectronic device [20].

The most fundamental experiment in the coherent regime aims for the observation of Rabi oscillations. As sketched in figure 2(b), resonant ps laser excitation results in a controlled manipulation of a two-level system, which can be displayed on the Bloch sphere (displayed here in the rotating frame). For a two-level system with a given oscillator strength, the rotation angle Θ of the Bloch vector is thereby proportional to the pulse area (or excitation amplitude).

The occupancy of the upper level of a two-level system under coherent resonant excitation is given by $\sin^2(\Omega t/2)$ [21, 22], where the Rabi frequency $\Omega$ is proportional to the square root of the laser intensity and $t$ corresponds to the pulse length. A $\pi$ pulse thereby results in a complete inversion of the two-level system. We define the pulse area, i.e. the rotation angle $\Theta = \Omega t$, by adjusting the excitation amplitude rather than the pulse length.

A $\pi$ pulse typically corresponds to an average laser power on the sample of about 2 $\mu$W at a pulse length of 2.3 ps and a repetition frequency of $f_{\mathrm{Laser}} = 80$ MHz. If the tunnel efficiency of our device was 100%, each $\pi$ pulse would contribute to the PC with one elementary charge, resulting in a maximum value of $I = f_{\mathrm{Laser}}e = 12.8$ pA [11].

In figure 3(a) we show the upper level occupancy measured in the PC as a function of the excitation pulse area. At the highest excitation intensities the system undergoes almost nine full inversions within each laser pulse. We used here circular polarized light in order to suppress bi-exciton generation not only by spectral separation but also by Pauli blocking [23–26]. The data displayed in figure 3(b) was obtained for $V_{\mathrm{B}} = 0.6$ V, which results in a tunneling-induced

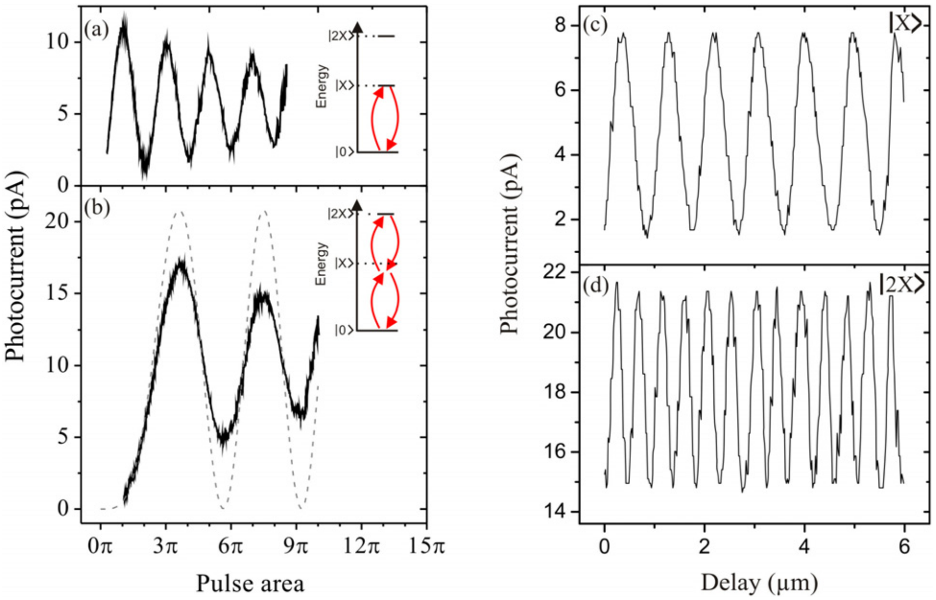

**Figure 3.** (a) Rabi oscillations of the one-photon ground state exciton transition, measured by PC spectroscopy. (b) Two-photon Rabi oscillations as measured in the PC (solid line) and as predicted by theory [15] (dashed line). Results from two-pulse interferometry are shown in (c) for the one-photon transition and in (d) for the two-photon transition.

dephasing time of $T_2 \approx 100$ ps. The nature of the damping of pulse-area-dependent Rabi oscillations has recently been the subject of intensive theoretical work (see [27–30] and references therein).

In an analogous way we have performed an equivalent experiment for the resonant two-photon bi-exciton transition, for which we changed to a laser energy of $E_{|XX\rangle}/2$, which corresponds to $E_{|X\rangle} - E_{B,XX}/2$ (where $E_{B,XX}$ is the bi-exciton binding energy) and to linear polarization (see the inset in figure 3(b)) [31]. This shift by $E_{B,XX}/2 = 1.38$ meV is considerably larger than the ps laser linewidth of 0.6 meV. The corresponding PC signal versus the square root of the power is plotted in figure 3(b) over the same $x$ axis as in figure 3(a). For those conditions we clearly observe two-photon Rabi oscillations of the $|XX\rangle$ state [15]. The power dependence, including the rather slow start at low pulse area, is in good qualitative agreement with theory [15]. We thereby show that the QD behaves very much like a quantum-mechanical model system even in the more complex case of two mutually coupled two-level systems. Additional and unambiguously convincing evidence for the previously discussed one- and two-photon Rabi oscillations is provided by two-pulse interferometry as shown in figures 3(c) and (d). Whereas in the one-photon case the interferogram exhibits a $\lambda$ periodicity as a function of the optical pulse delay, a clear and also expected $\lambda/2$ periodicity is observed for the two-photon case.

## 3. Coherently excited single-photon emitters

The general concept of the so-far employed excitation scheme for semiconductor-based single-photon emitters is based on strong incoherent, non-resonant excitation (either by optical or electrical pumping) and a sequential decay cascade with final single-photon emission [32]. Different renormalization energies of the various multi-exciton states thereby allow for spectral isolation of the photon emitted by the decay of the last exciton from the ground state of the QD. Strong excitation and the formation of multi-exciton states are necessary to avoid the case of missing population within the statistical fluctuation of the created exciton number for this incoherent process.

Single-photon emission from QDs had been reported first by Michler *et al* [7], followed by many other groups employing both optical and electrical pumping. Single-photon emitters and recently also sources for entangled photons [33–35] are of special interest in the fields of quantum cryptography and quantum information research. Very recently it has been shown that incoherent pumping schemes result in a degradation of the main figure of merit, the second-order correlation function $g^{(2)}(t)$, if high repetition rates of photons on demand are required [36]. One way out of this problem can be the application of an excitation scheme, which provides us, with high probability, just one bright exciton per excitation cycle.

As pointed out before, coherent state preparation allows for the generation of a defined optical polarization which, in the case of $\pi$ pulse excitation, results after projection in a single (and bright) exciton occupancy. The coherent manipulation of the single-exciton ground state has been shown to work very well, with up to 95% efficiency [11, 19], but it cannot be directly applied here due to the spectral overlap of ps excitation and single-photon emission. In order to spectrally separate the emission from the ps excitation, we suggested therefore to perform, for example, p-shell Rabi flopping with subsequent relaxation to the ground state, followed by single-photon emission [37].

In figure 4(a) we show a photoluminescence excitation (PLE) spectrum of an InGaAs QD with a p-shell transition ($|x_p\rangle$) about 30 meV above the ground state. In cw PL experiments this transition shows, with high accuracy, the

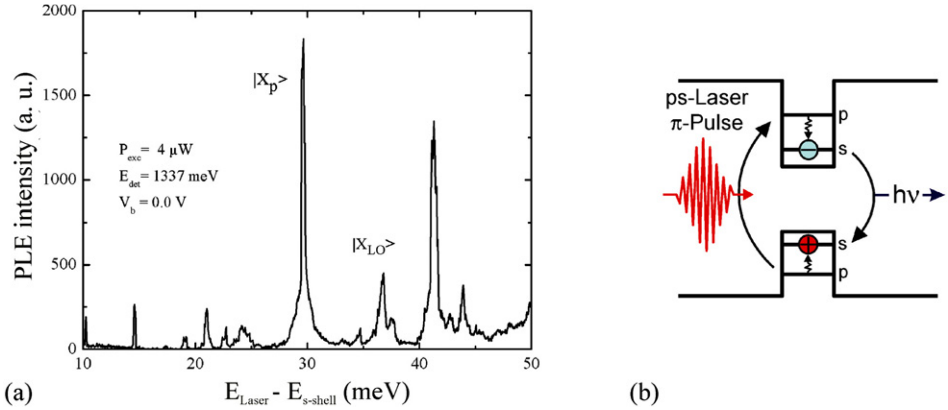

**Figure 4.** (a) PLE-spectrum of an InGaAs QD. The strongest peak at about 30 meV is assigned to the p-shell absorption ($|X_\mathrm{p}\rangle$). The resonances around 36.7 meV correspond to (LO) phonon-assisted absorption ($|X_\mathrm{LO}\rangle$). (b) Excitation scheme for a coherently excited single-photon emitter.

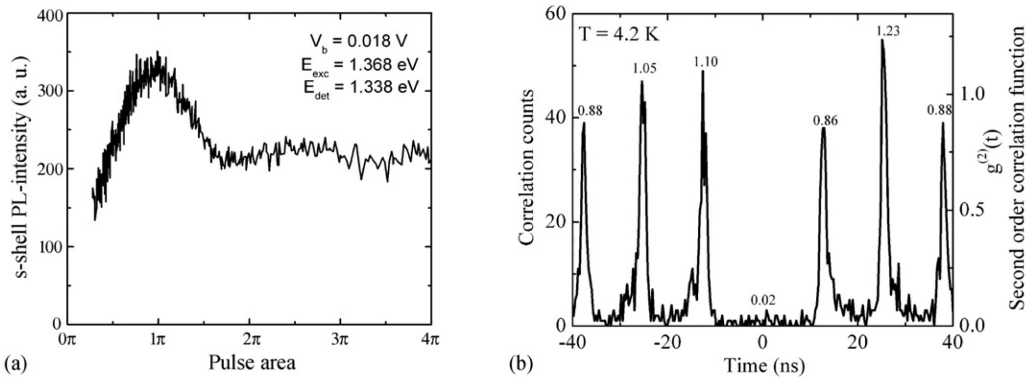

**Figure 5.** (a) p-shell Rabi oscillations detected in the ground state PL. The Rabi oscillations have a clearly developed $\pi$ pulse maximum, followed by substantial damping at higher pulse areas. (b) Photon correlation measurement under the condition of $\pi$ pulse (2 $\mu$W) excitation in the p-shell. The central peak of the periodic pattern is strongly suppressed, which proves clean single-photon emission at a repetition rate of 80 MHz.

nonlinear saturation behavior, which is expected for two-level systems [21, 38, 39].

Based on those findings we have performed the basic excitation scheme shown in figure 4(b), which starts with $\pi$ pulse excitation and Rabi flopping in the p-shell, followed by p- to s-shell relaxation and spontaneous recombination in the s-shell. Coherent state preparation by Rabi flopping, as applied here, is probably one of the most efficient ways for deterministic creation of a single, bright exciton, as required for single-photon emission. At the same time it is the minimal required excitation, which also avoids the creation of multi-excitons or charged states in a QD.

In figure 5(a) we show a Rabi oscillation of the p-state measured in the s-shell PL. The PL intensity shows a clear nonlinear behavior, whereby the oscillations imply that the excitation of the QD can be controlled by the strength of the incident laser field. The measurements exhibit a strong damping of the oscillation, but the first maximum is clearly visible. The data obtained here are comparable to the results reported in, for example, [9]. The damping indicates enhanced dephasing rates compared to ground state Rabi oscillations [19] (see also figure 3(a) for comparison). The dephasing time of the p-shell exciton was measured to be 6 ps by two-pulse interferometry. A $\pi$ pulse for the p-shell transition typically corresponds to an average laser power on the sample of about 2 $\mu$W at a pulse length of 2.3 ps and a laser repetition frequency of $f_\mathrm{Laser} = 80$ MHz.

Assigning the strongly damped part of the Rabi data shown in figure 5(a) (between pulse areas of about $2\pi$ and $4\pi$) to the incoherent limit of 50% projected exciton occupancy, we are able to conclude, that the efficiency of the coherent state preparation for the $\pi$ pulse condition is about 76% in our current data.

For the proof of the quality of the resulting single-photon emission a Hanbury-Brown and Twiss (HBT) [40] set-up with two similar avalanche photodiodes was used. The time correlation measurement was performed by a time-to-amplitude converter with a resolution of 300 ps. It gives the probability that a photon is detected at a time $t$ and another photon at a time $(t + \tau)$. The normalized second-order correlation function $g^{(2)}(\tau)$, which is the most important figure of merit for single-photon emitters, is given by $g^{(2)}(\tau) = \langle I(t)I(t+\tau)\rangle/(\langle I(t)\rangle\langle I(t+\tau)\rangle)$, where $I(t)$ corresponds to the emission intensity [41]. Under continuous excitation, a

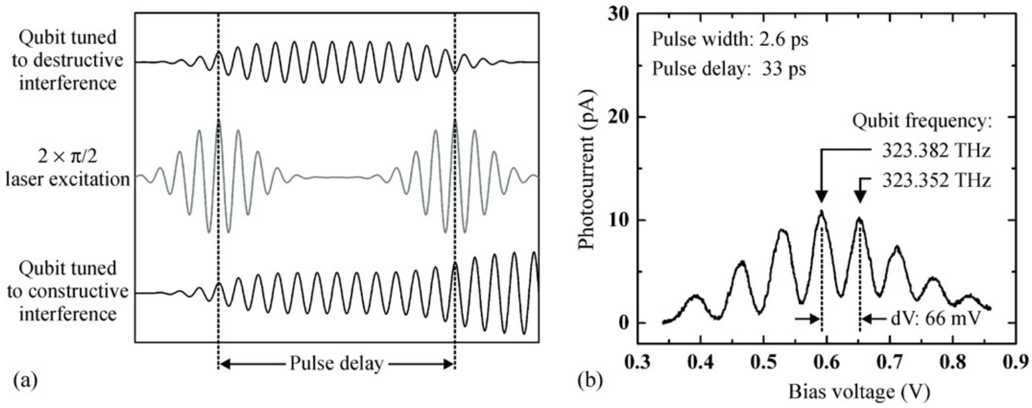

**Figure 6.** (a) Schematic picture of the phase relation between a coherent QD polarization (upper and lower curves) and two laser pulses (central curve). On arrival of the second pulse the phase of the QD in the detuned case is exactly opposite compared to the resonant case. This results in a strong variation of the final state occupancy although the detuning would be almost negligible for single-pulse excitation. (b) Photocurrent spectrum double-pulse excitation with 33 ps delay. The envelope of the signal corresponds to the spectrum of a single pulse, centered at 323.382 THz. When tuning the transition energy of the quantum dot, we observe interference oscillations (Ramsey fringes).

single quantum emitter would generate a dip in the histogram at zero delay time ($\tau = 0$). This means that the system must be re-excited after emission, before a second photon can be emitted. This effect is known in the literature as antibunching.

In our experiments the use of pulsed excitation leads to a recurring pattern of peaks, which are separated in time by the pulse repetition period of the mode-locked Ti:sapphire laser of about 12.5 ns. Each detected photon can be correlated with a photon generated from one of the next laser pulses. Due to the fact that there is at most one photon for each laser pulse, there cannot be a correlation at zero delay time. Therefore, single-photon emission will lead to a missing peak at zero delay time, which means that $g^{(2)}(\tau = 0)$ should vanish.

Corresponding data for $\pi$ pulse excitation is shown in figure 5(b). Due to the design of the investigated sample (no Bragg mirrors and light collection through a near-field shadow mask) the detected count rate is accordingly low. As a matter of fact, however, the $g^{(2)}$ value of the central peak is almost zero for the condition of 80 MHz $\pi$ pulse excitation [37]. Our results therefore indicate that the concept of coherent state preparation is very useful for the development of high repetition rate and highly deterministic single-photon emitters.

## 4. Ramsey fringes: electric-field-controlled quantum interference

The basic effect of moderate detuning in coherent experiments is an incomplete inversion for $\pi$ pulse excitation and a slight modification of the Rabi frequency [21]. The spectral sensitivity of a two-level system is substantially enhanced though, by applying two phase-controlled pulses separated by a fixed time delay. This effect was first described by N F Ramsey and is well known from atomic optics [42]. Due to numerous applications of the effect in precision spectroscopy and atomic clocks, Ramsey was awarded the 1989 Nobel Prize.

With the pulse sequence shown schematically in figure 6(a) we like to examine the influence of detuning for the case of weakly detuned double-pulse excitation. We consider two $\pi/2$ pulses, which result, depending on their relative phase, in a pure $|1\rangle$ or $|0\rangle$ final state. A first $\pi/2$ pulse brings the two-level system into a superposition state between $|0\rangle$ and $|1\rangle$ which falls on the equator of the Bloch sphere. Since the Bloch sphere is displayed in the rotating frame of the laser field, the Bloch vector will perform a precession around the equator of the Bloch sphere with $\Delta\omega = \omega_{\mathrm{QD}} - \omega_{\mathrm{Laser}}$. Within the time delay $\tau_{\mathrm{delay}}$ between the two laser pulses the phase difference between the laser field and the two-level system proceeds therefore proportional to time. Depending on the momentary phase of the two-level system after the delay time, the second $\pi/2$ pulse will rotate the Bloch vector to its final state, which is the state $|0\rangle$ or $|1\rangle$ for the case of destructive or constructive interference, respectively (see figure 6(a)). As a function of detuning, the phase will change correspondingly and hence lead to an oscillation of the final state between $|0\rangle$ and $|1\rangle$ (Ramsey fringes). In general the final state will oscillate as a function of the detuning $\Delta\omega$ with a period of $2\pi/\tau_{\mathrm{delay}}$. This means that the frequency of the spectral fringes increases directly proportional to the delay time between the two pulses [42, 43].

In figure 6(b) we show a Ramsey fringe for a fixed delay time of 33 ps. The envelope of the data corresponds to the power spectrum of the ps laser [43]. The oscillation is caused by the voltage-dependent detuning of the QD resonance with respect to the fixed laser energy (ps laser centered at 1337.4 meV), as described above. Within an energy interval of $\Delta E = 0.124$ meV we observe $n = \tau_{\mathrm{delay}}\Delta E/h = \tau_{\mathrm{delay}}/33$ ps periods of the oscillation, where $h$ is Planck's constant. Expressed in frequency this means, that two oscillators, the laser with 323.382 THz and the two-level system with 323.352 THz (one fringe period away), run out of phase by $2\pi$ within 33 ps. Within this delay time, the coherent polarization of the two-level system has performed about 10 670 oscillation periods, one less compared to the reference laser field. As indicated in figure 6(b), only a small change in bias voltage is required to electrically sweep the quantum system between two adjacent conditions of constructive interference ($\mathrm{d}V = 66$ mV for a 33 ps pulse delay).

For delay times longer than the dephasing time the spectral resolution of such electrically controlled Ramsey experiments can be increased beyond the continuous excitation limit (i.e. the homogeneous linewidth) [43]. In view of quantum information processing and coherent optoelectronics we preferentially consider the case of delay times shorter than the dephasing time. An excitation with two $\pi/2$ clock pulses can then be applied to enable the electrically controlled manipulation of the quantum state of a two-level system.

## 5. Summary

In summary we have reported on the coherent optical and optoelectronic properties of a single QD in a photodiode. In the previous sections we have described this system as an exciton two-level system with electrical contacts. Compared to conventional photodiodes, single QD photodiodes exhibit resonant spectral sensitivity, which can be tuned over a wide range by the quantum-confined Stark effect. With resonant ps laser excitation new and so far unattained coherent functionalities also become available. Of particular interest for future applications is therefore (i) the phase-sensitive response on multiple laser pulses, (ii) the availability of coherent state preparation and manipulation in an optoelectronic device and (iii) the availability of quantitative (photocurrent), projective quantum measurements.

By means of single QD photodiodes it becomes possible to transfer one- or two-photon optical excitations of single quantum systems into deterministic electric currents. This new optoelectronic functionality provides a link between the world of coherent optical excitations and electric currents on the single-electron level. In future this will allow for the efficient electric readout of exciton qubits and for (optically triggered) deterministic single-electron sources. Applied to the field of single-photon emission, the coherent control of excited state transitions has been shown to be a promising route towards deterministic high repetition rate and high fidelity single-photon sources.

The Ramsey effect can be applied for precision measurements and for the electrically induced manipulation of single QDs. In the context of quantum information processing this will allow us to implement voltage-controlled state preparation (as demonstrated here) and even electrically controlled quantum gates, if fast electric signals are applied within the delay time between the laser pulses. Single QD photodiodes and related devices can therefore help us to establish a new generation of coherent optoelectronics, which could have a substantial impact on future information technologies.

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