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Beating of excitonic dressed states in a single quantum dot

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Abstract

We report picosecond control of excitonic dressed states in a single semiconductor quantum dot. A strong laser pulse couples the exciton and biexciton states, to form an Autler-Townes doublet of the neutral exciton transition. The Rabi-splitting, and hence the admixture of the dressed states follows the envelope of the picosecond control laser. We create a superposition of dressed states, and observe the resulting beat: a direct measurement of a Rabi oscillation in *time*-delay rather than the usual power domain.

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Semiconductor quantum dots possess isolated discrete excitonic energy levels, with an atom-like light-matter interaction. The upper and lower excitonic states of any optical transition can be optically coupled, and in the regime where this coupling is far stronger than the rate of coherence loss, a strong-coupling regime occurs where the upper and lower states are mixed to form dressed states. In the frequency domain, signatures of dressed states include: power dependent Rabi splittings [1–3], Mollow triplet [3, 4], and Autler-Townes doublet [1, 3, 5–7]. Since much of the interest in dressed states lies in their possible exploitation for the coherent control of excitonic or spin states in quantum information applications, it is important to achieve dynamic control of dressed states in addition to the previously reported steady-state control. The hallmark of dressed states in the time domain is the Rabi oscillation. For transitions with microsecond coherence times, such as electron spins in quantum dots [8] or some atomic transitions [9], a Rabi oscillation in time can be measured by varying the time-duration of the rf-field. However, for excitonic Rabi oscillations in a semiconductor quantum dot, varying the time-duration of the picosecond laser pulse is non-trivial. Consequently, Rabi oscillations have been indirectly measured as a function of the time-averaged excitation power [10, 11].

Here we report time-resolved measurements of the excitonic dressed states of a single semiconductor quantum dot. A pair of picosecond laser pulses excites a single quantum dot: an intense control pulse to couple the exciton and biexciton states, and a weaker probe to time-resolve the resulting Autler-Townes doublet of the neutral exciton transition. We make two key observations. (1) The Rabi-splitting, and hence the admixture of the dressed states follows the picosecond envelope of the control pulse, indicating that coherent control strategies for quantum dot excitons and spins, such as adiabatic passage [12, 13], based on the picosecond manipulation of the composition of dressed states are feasible. (2) We observe a beat arising from a coherent superposition of the excitonic dressed states. In the excitonic basis, this is a direct time-resolved measurement of a Rabi oscillation between the exciton and biexciton states, achieved by gating the duration of the driving field using a two-color pump-probe scheme. A similar scheme has been utilized in quantum wells [14].

We study a single spatially isolated InGaAs/GaAs quantum dot embedded in the intrinsic regime of an n-i-Schottky diode structure. The dot emits at 1.302 eV, and is excited by two spectrally narrow Gaussian shaped laser pulses derived from a single 150-fs pulse using a pair of pulse-shapers. Absorption of the laser pulses creates electron-hole pairs in the dot, which

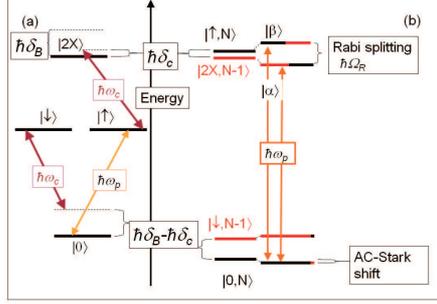


FIG. 1: (color online) Energy-level diagram of a neutral dot. (a) There are four excitonic states: empty $|0\rangle$, exciton spin up/down $|\uparrow\rangle, |\downarrow\rangle$, and the biexciton $|2X\rangle$. (ω_c) The control laser couples the \uparrow - $2X$ states, whilst a π -pulse (ω_p) probes the 0 - \uparrow transition. (b) Dressed states picture. The basis states are labeled $|\zeta, n\rangle$, where ζ is the excitonic state, and n the photon number. The states $|\uparrow, N\rangle, |2X, N-1\rangle$ detuned by $\hbar\delta_c$, are coupled by the Rabi energy $\hbar\Omega_c$ of the control pulse, forming a pair of dressed-states $|\alpha\rangle, |\beta\rangle$ with a Rabi-splitting $\hbar\Omega_R$. The control pulse also couples the $|0, N\rangle$ and $|\downarrow, N-1\rangle$ bare-states far from resonance, resulting in an AC-Stark shift of the $|0, N\rangle$ state.

tunnel from the dot and are detected as a photocurrent [11]. Further details of the setup, and photocurrent spectroscopy of the dot can be found in ref. [15]. The sample temperature is 12 K. A background photocurrent proportional to the incident power is subtracted from all data.

An energy-level diagram of the neutral dot is presented in figure 1(a). There are four excitonic states: vacuum $|0\rangle$, exciton spin-up $|\uparrow\rangle$, exciton spin-down $|\downarrow\rangle$, and the biexciton $|2X\rangle$. The biexciton binding energy $\hbar\delta_B = 1.9$ meV. Due to a slight anisotropy of the dot, the exciton states are coupled by the electron-hole exchange interaction, resulting in linearly polarized energy eigenstates with a fine-structure splitting of $\hbar\delta_{fs} = 15$ μ eV. Two picosecond laser pulses excite the dot, an intense control to couple the up-exciton and biexciton states (\uparrow - $2X$), and a weaker probe to detect the resulting doublet of the neutral exciton transition ($0 - \uparrow$), with center frequencies ω_c, ω_p respectively. Since the fine-structure splitting is small compared with the spectral width of the pulses, the exciton spin up/down states act as energy eigenstates. All of the experiments use cross-circularly polarized control and probe pulses to satisfy the selection rules for creation of a biexciton. Circular polarization also avoids single pulse excitation of a biexciton [16].

The formation of the Autler-Townes doublet is depicted in the dressed states picture shown in fig.1(b), which is a transformation to the rest-frame of the control laser. Here the basis-states $|\zeta, n\rangle$, are labeled by both the excitonic state ζ , and the number of photons n of energy $\hbar\omega_c$ in the mode of the control laser. The upper states $|\uparrow, N\rangle$, and $|2X, N-1\rangle$ are separated in energy by the detuning of the control laser from the \uparrow - $2X$ transition: $\delta_c = (\omega_c - \omega_{\uparrow 2X})$, and optically coupled with the Rabi energy $\hbar\Omega_c$ of the control pulse. Consequently the basis-states are mixed to form a pair of dressed states $|\alpha\rangle$ and $|\beta\rangle$, which anti-cross on resonance, with energies:

$$\Delta E_{upper}^{(\pm)} \approx \frac{\hbar}{2}(-\delta_c \pm \sqrt{\delta_c^2 + \Omega_c^2}) \quad (1)$$

The control pulse also couples the lower states $|\downarrow, N-1\rangle$ and $|0, N\rangle$ far from resonance ($\Omega_c \ll -\delta_c + \delta_B$), and hence the lower states experience an AC-Stark shift of:

$$\Delta E_{lower}^{(\pm)} \approx \frac{\hbar}{2}(-\delta_c + \delta_B \pm \sqrt{(-\delta_c + \delta_B)^2 + \Omega_c^2}) \quad (2)$$

with weak mixing of the basis-states. The probe reveals the Autler-Townes doublet when it is absorbed in resonance with the 0 - α , and 0 - β transitions at $\hbar\delta_p = \Delta E_{upper}^{(\pm)} - \Delta E_{lower}^{(-)}$, where $\delta_p = \omega_p - \omega_{0\uparrow}$ is the detuning of the probe from the $(0 - \uparrow)$ transition.

To observe this energy-level structure, measurements were performed with temporally overlapped pulses, and are presented as grayscale plots of the photocurrent in fig. 2. Spectrally narrow probe and control pulses were used with a spectral FWHM of 0.2 meV, and zero time-delay. The Rabi frequencies of the pulses are calibrated using Rabi rotation measurements for pulses tuned to the $0 - \uparrow$ and \uparrow - $2X$ transitions, where the photocurrent is measured versus incident power [15]. π -pulses are achieved using 0.27 μ W of incident power. The control pulse is tuned on resonance with the \uparrow - $2X$ transition ($\delta_c = 0$), and the photocurrent measured as a function of the detuning δ_p of the probe pulse of pulse-area $\Theta_p = \pi$, where $\Theta_i = \int \Omega_i(t)dt$, ($i \in c, p$). Figure 2(a) presents a grayscale plot of the photocurrent spectra versus the pulse-area of the control pulse. A doublet is clearly resolved with a splitting proportional to the pulse-area of the control pulse. Because the picosecond control pulse can achieve high peak intensities, the observed splitting of greater than 0.4 meV is much larger than the ~ 10 μ eV seen in previous CW observations [1, 3, 5]. The mean energy of the two peaks also exhibits a quadratic increase due to the AC-Stark shift [17].

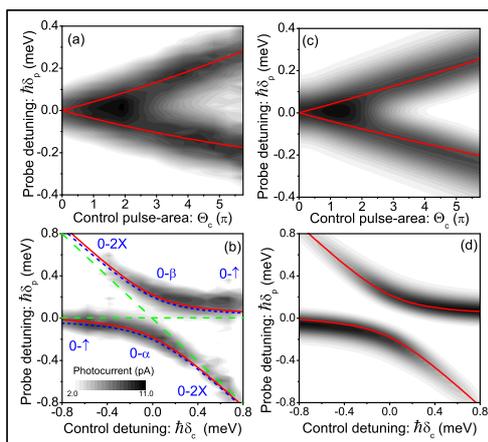


FIG. 2: (color online) Measurements with temporally overlapped spectrally narrow pulses of identical duration. (a) Grayscale plot of photocurrent versus probe detuning, versus pulse-area of control pulse with zero detuning $\delta_c = 0$. (b) Grayscale plot of photocurrent versus probe detuning, versus detuning of a control of pulse-area $\Theta_c = 5\pi$. Green dashed-dot lines indicate energies of the $0-\uparrow$ and $0-2X$ transitions. The red-lines are fits to (eqs. 1, and 2), and blue lines show the fit without AC-Stark shift. (c,d) Numerical simulations of (a,b) with no fitting parameters. A grayscale offset removes the background signal.

To observe the anti-crossing of the dressed states, the pulse-area of the control pulse is fixed at $\Theta_c = 5\pi$, and the photocurrent spectra recorded as the detuning δ_c of the control pulse is varied. The data is shown as a grayscale plot in fig. 2(b), and maps the energies of the dressed states $|\alpha\rangle$, and $|\beta\rangle$, as depicted in fig.1(b). For large detunings of the control laser the photocurrent peaks tend towards the single photon $0 - \uparrow$, and two-photon $0 - 2X$ transitions. The midpoint of the doublet exhibits an AC Stark-shift that increases as the control laser approaches the $0 - \uparrow$ transition. Figures 2(c,d) present numerical simulations [18] of the corresponding experiments (a,b), where the dot is treated as an ideal 4-level atom, using no fitting parameters. As well as the full numerical treatment, fits to equations 1, and 2, are presented as red-lines, with a ‘time-averaged’ Rabi energy $\hbar\Omega_c \rightarrow \langle \hbar\Omega_c \rangle = 80\Theta_c/\pi \mu eV \equiv A\Theta_c$, where Θ_c is the pulse-area of the control pulse. Good agreement between experiment, and theory [18] is achieved.

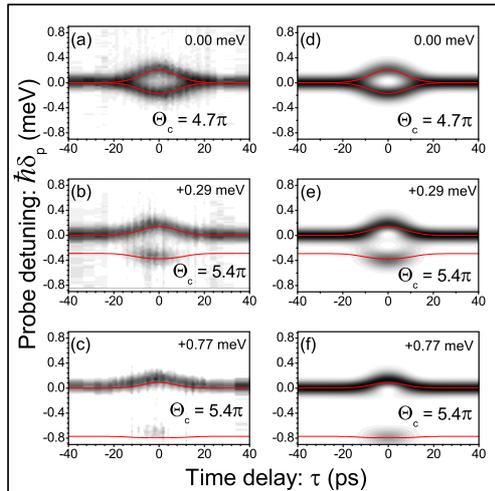


FIG. 3: (color online) Time-resolved measurements of Autler-Townes splitting. (a-c) Grayscale plot of photocurrent versus probe detuning versus time-delay τ at various detunings of the control pulse. Red-lines show the effective Rabi energies of the control pulse in time. (d-f) Simulations of (a-c).

If the observed doublet is due to the control laser mixing the exciton and biexciton states, the doublet should disappear the instant the control laser is switched off, whereas if the doublet arises due to a carrier mediated nonlinearity it would persist following the turn-off of the control laser. To test this, we performed photocurrent spectra measurements versus the time-delay τ , where for positive time-delay the probe pulse precedes the control pulse. The data is presented in figs. 3(a-c), for various detunings of the control pulse, with the pulse-areas Θ_c given in fig.3. The red-lines trace the time-evolution of the energies of the dressed states found by substituting $\langle \hbar\Omega_c \rangle = (A\Theta_c)\exp(-(\frac{\tau}{2T_c})^2)$ into eqs 1, and 2. $\langle \hbar\Omega_c \rangle$ is proportional to the envelope of the electric-field of the laser with time durations $T_c = 5.6$ ps, $T_p = 4.2$ ps determined from spectral and autocorrelation measurements of the control, and probe lasers respectively. In the case of zero detuning, fig. 3(a), a symmetric splitting following the Gaussian pulse-shape of the control laser is observed. As the detuning is increased, the eye-like pattern seen in the grayscale plot of fig. 3(a) evolves into a continuous line with a kink, and a satellite peak close to the two-color two-photon absorption resonance. At positive time delays the pulse sequence creates a biexciton, and at negative

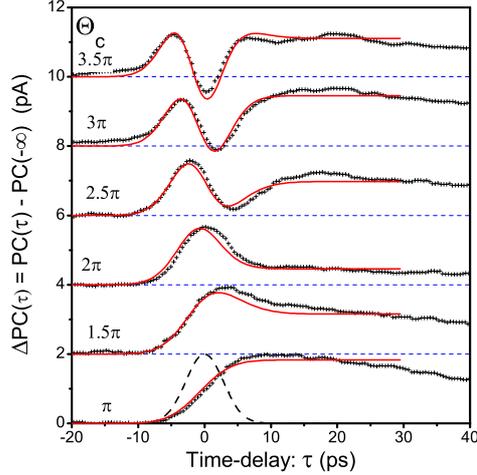


FIG. 4: (color online) Beats between dressed states of \uparrow and $2X$, or time-resolved measurements of a exciton-biexciton Rabi oscillation. The change in photocurrent $\Delta PC(\tau) = PC(\tau) - PC(-\infty)$ is measured as a function of the time-delay τ , for a slow control and fast probe pulses ($T_c = 4$ ps, $T_p = 1.072$ ps) for various pulse-areas Θ_c of the control pulse. Lines show fits to data. Dashed line traces envelope of control pulse.

delays an exciton, resulting in higher photocurrent at positive delay. The oscillations that appear in fig. 3(a-c) are an artifact of the sampling interval of the experiment. Figures 3(d-f) present 4-level atom simulations [18], showing close agreement with the theory using the same experimentally determined parameters as in fig. 2. The doublet clearly follows the envelope of the control pulse, providing strong evidence that the doublet is formed by optically coupled states only. Figure 3 shows that the Rabi-splitting, and hence the admixture of the dressed states of a semiconductor quantum dot can be manipulated on a picosecond timescale. This is an important step towards the realization in quantum dots of control strategies, such as adiabatic passage [12, 13] that rely on the dynamic control of the composition of dressed states. Compared with conventional control strategies based on Rabi rotations, adiabatic passage should be less sensitive to dephasing and errors in the control sequence, leading to higher fidelity quantum gates.

A definitive time domain observation of the Autler-Townes doublet is the observation of a beat arising from a coherent superposition of the dressed states $|\alpha\rangle$ and $|\beta\rangle$. In the excitonic

basis, this is a Rabi oscillation in time between the exciton and biexciton states. The principle of the measurement is as follows. The dot is excited with a slow ($T_c = 4$ ps, $\delta_c = 0$) control pulse to split the dressed states, and a fast ($T_p = 1$ ps, $\delta_p = 0$, $\Theta_p = \pi$) probe pulse to create a superposition of dressed states. Consider the limit where the spectral width of the probe is far greater than the peak Rabi frequency of the control pulse ($2\sqrt{\ln(2)} \gg \Omega_c T_p$). When the probe pulse precedes the control at a time-delay τ with respect to the control pulse, it creates an equal superposition of the dressed states: $|\uparrow\rangle = (|\alpha\rangle + |\beta\rangle)/\sqrt{2}$. Due to the Rabi splitting the dressed states $|\alpha\rangle, |\beta\rangle$ accumulate a relative phase-shift $\theta(t, \tau) = \int_{-\tau}^t \Omega_c(t') dt'$, resulting in a beat in the excitonic basis. Consequently the final state ($t = \infty$) as measured in photocurrent oscillates between the exciton and biexciton states as the time-delay is varied.

Figure 4 presents time-resolved measurements of the exciton-biexciton Rabi oscillation for various pulse-areas of the control pulse. The change in photocurrent, $\Delta PC(\tau) = PC(\tau) - PC(-\infty)$, versus time-delay τ is plotted. The spectral width of the probe is a compromise between time resolution, and background signal. At low pulse-areas ($\Theta_c < 3\pi$) the data can be understood in terms of a time-resolved Rabi oscillation. For a π -pulse the photocurrent exhibits a step, followed by a decay due to a 110-ps electron tunneling time, and a slow fine-structure beat. The step can be reproduced using a simplistic two-level atom picture, where $\Delta PC \propto \sin^2(\theta(\infty, \tau)/2)$ (not shown). The 1.5π -pulse exhibits a maximum and the photocurrent then decreases to half the maximum. For the 2π -pulse the signal reaches a maximum at a time-delay of zero, but due to dephasing does not fully return to zero. For pulse-areas of up to 3π the data is consistent with a rotation through an angle equal to the pulse-area Θ_c . The oscillation occurs only for the time duration of the control laser, and exhibits an increased frequency at higher pulse-areas. The interpretation of these measurements as a time-resolved Rabi oscillation, begins to falter at pulse-areas $\Theta_c > 3\pi$, most notably where the change in photocurrent goes negative close to zero time-delay. Here the spectral overlap of the probe (FWHM 0.8 meV), and the doublet with a peak Rabi-splitting of 0.53 meV is reduced, inhibiting the absorption of the probe.

The main features of the data in fig. 4, namely the oscillation and the inhibited absorption can be reproduced using an idealized 4-level atom model with field independent decay rates (not shown). However, to fit the damping of the Rabi oscillation an additional phenomenological excitation induced dephasing (EID) term [19] was used: $\dot{\rho}_{\uparrow 2X}|_{damp} = -D\Omega_c^2(t)\rho_{\uparrow 2X}$. A good fit to all of the data is obtained for a single value of the EID-time $D = 73 \pm 8$ fs,

corresponding to a dephasing rate of $1/(24 \text{ ps})$ for $\hbar\Omega_c = 0.5 \text{ meV}$. For reasons beyond the scope of this letter, we attribute the EID in our sample to acoustic phonons [20].

Figure 4 demonstrates a two-color pump-probe scheme for direct time-resolved measurements of a Rabi oscillation in a single dot. Unlike Rabi rotation measurements in the power domain, the temporal information has not been integrated out. The close agreement between experiment and theory demonstrates that the Rabi frequency of the \uparrow - $2X$ transition is proportional to the amplitude of the control field on a sub-picosecond timescale. With improved time-resolution and higher excitation powers, it should be possible to resolve the field dependence of the excitation induced dephasing. Figure 4 demonstrates the creation of a coherent superposition of two dressed states, and the ability to control their relative phase by gating the control field with a second probe pulse.

To summarize, an intense laser pulse couples the exciton and biexciton states to form an Autler-Townes doublet in a neutral quantum dot. We observe the doublet in both time and frequency domains. The Rabi-splitting and hence the admixture of the dressed states follows the envelope of the control pulse. Finally we observe a beat arising from a coherent superposition of the excitonic dressed states, a Rabi oscillation in time, instead of the usual power domain. This work shows that the dressed states of semiconductor quantum dots may be optically manipulated on a picosecond timescale, paving the way for promising control strategies based on the manipulation of dressed states.

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