A Single Charged Quantum Dot in a Strong Optical Field: Absorption, Gain, and the AC Stark Effect

Xiaodong Xu,1 Bo Sun,1 Erik D. Kim,1 Katherine Smirl,1 P. R. Berman,1 D. G. Steel,1⊥A. S. Bracker,2 D. Gammon,2 and L. J. Sham3

1The H. M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan 48109, USA
2The Naval Research Laboratory, Washington D.C. 20375, USA
3Department of Physics, The University of California-San Diego, La Jolla, California 92093, USA

(Dated: March 5, 2008)

We investigate a singly-charged quantum dot under a strong optical driving field by probing the system with a weak optical field. When the driving field is detuned from the trion transition, the probe absorption spectrum is shifted from the trion resonance as a consequence of the dynamic Stark effect. Simultaneously, a gain sideband is created, resulting from the coherent energy transfer between the optical fields through the quantum dot nonlinearity. As the pump detuning is moved from red to blue, we map out the anticrossing of these two spectral lines. The optical Bloch equations for a stationary two-level atom can be used to describe the numerous spectral features seen in this nano solid state system.

Quantum dot (QD) nano-structures have been proposed for numerous quantum mechanical applications due to their customizable atom-like features [1]. One important application involves using these QDs as the building blocks for quantum logic devices [2]. An electron spin trapped inside a QD is a good candidate for a quantum bit (qubit) since it is known to have long relaxation [3] and decoherence times [4, 5]. Recently, the electron spin coherence has been optically generated and controlled [6, 7] in ensembles of QDs. The initialization of the electron spin state in a single QD has also been realized by optical cooling techniques [8, 9].

One important task is to understand and control the physical properties of a singly-charged QD in the strong optical field regime, i.e. the light-matter interaction strength is much larger than the transition linewidth, under both resonant and nonresonant excitation. Given the recent work on optically driven neutral quantum dots in strong fields [10, 11] demonstrating many features similar to atomic systems, it is clear that a negatively charged quantum dot has similarities to a negative ion. However, the excited state of a dot is a many body structure comprised of two electrons and a hole. Interestingly, the results in this paper show that strong field excitation tuned near resonance in a negatively charged dot leads to changes in the absorption spectrum that are in excellent agreement with theory.

In the time domain, the strong field interaction leads to the well-known Rabi oscillations [12, 13, 14, 15]. In the frequency domain, it will introduce Rabi side bands in the absorption, and strikingly, the amplification of a probe beam. This phenomenon has been studied theoretically [16, 17, 18] and demonstrated experimentally in atomic systems [19, 20]. Recently, these effects have also been observed in quantum dot and molecular systems [10, 11, 21, 22]. The optical AC Stark effect has been seen by exciting a neutral QD with a detuned strong optical pulse [21] while the Mollow absorption spectrum and Mollow triplets [23] have been observed in a single neutral QD [10, 11] and a single molecule with intense resonant pumping [22], respectively. However, the study of the singly-charged QDs in the strong field regime has been very limited at the single dot level [24].

In this letter, we investigate a singly-charged QD under a strong optical driving field with both on and off-resonant pumping. When the strong pump is on resonance with the trion transition, a triplet appears in the probe absorption spectrum with a weak center peak and two Rabi side bands with dispersive lineshapes. As the pump beam is detuned from the trion transition, we observe three spectral features: a weak dispersive lineshape centered at the driving field frequency flanked by an AC Stark shifted absorption peak and a Raman gain side band. Our results reflect the coherent nonlinear interaction between the light and a single
quantum oscillator, and demonstrate that even at high optical field strengths, the electron in a single quantum dot with its ground state and trion state behaves as a well isolated two-level quantum system. It is a step forward toward spin based QD applications.

Assuming the trion can be considered as a simple two-level system in the absence of the magnetic field, the only optically allowed transitions are from the spin ground states (|±1/2⟩) to the trion states (|±3/2⟩) with σ± polarized light excitations. Since the Zeeman sub-levels of the electron spin ground state are degenerate, as are the trion states, both trion transitions are degenerate. We then use the two-level optical Bloch equations to model the trion system. For simplicity, we labeled the electron spin ground state as state |S⟩ and the excited state as |T⟩, as shown in Fig 1(a).

It is known that in a two-level system driven by a strong optical field, the absorption of the weak probe beam is significantly modified [16, 17, 18]. By solving the optical Bloch equations to all orders in the pump field and first order in the probe field, we obtain the absorption coefficient of the probe beam as [17]

$$\alpha = \text{Im}[\frac{\alpha_0 \gamma_T A(i \gamma_T A^* \Omega + i \Delta) + \Delta(\frac{\Omega_R^2}{2} - \gamma B + i (B + i \Delta) \delta_1 + \delta_1^2)}{(\gamma_T^2 A A^* + i \Omega_R^2 \gamma \Delta)(B^2 + \delta_1^2) + \gamma \Omega_R^2 B + \gamma T(\Omega_R^2 (\gamma + B)(\gamma B + \delta_1^2) + i \gamma B^2 \Delta + i \delta_1^2 \Delta(B^2 + \gamma^2 + \delta_1^2))}]$$

where $\gamma_T$ ($\gamma$) is the population (coherence) decay rate of the trion state, $\delta_1 = \omega_1 - \omega_o$ is the detuning of the pump frequency $\omega_o$ from the trion transition $\omega_1$, $\Delta = \omega_2 - \omega_1$ is the probe frequency $\omega_2$ detuning from the pump, $A = \gamma + i \delta_1$, $B = \gamma + i \Delta$, $\alpha_0$ is a constant, $\Omega_R = \mu \cdot E_{\text{pump}}/\hbar$ is the Rabi frequency of the pump field, $\mu$ is the dipole moment matrix element and $E_{\text{pump}}$ is the pump field strength.

When the strong pump is on resonance with the trion transition ($\delta_1 = 0$ and $\Omega_R >> \gamma$), the probe will show a complex Mollow absorption spectrum, which has been discussed in detailed in Ref [10], where a neutral exciton has been studied with a strong resonant pumping.

When the pump detuning is larger than the transition linewidth, the physics can be understood in the fully quantized dressed state picture. The uncoupled QD-field states (Fig. 1b) map into the dressed states (Fig. 1c) when the QD-field interaction is included. In Fig. 1 we assume the pump detuning $\delta_1$ to be negative, |S⟩ and |T⟩ are the quantum dot states, and $N$ is the photon number. Due to the light-matter interaction, one set of the dressed states can be written as [25]

$$|I(N)⟩ = c |S, N⟩ - s |T, N - 1⟩$$
$$|II(N)⟩ = s |S, N⟩ + c |T, N - 1⟩$$

where $c = \sqrt{\frac{1}{2}(1 - \delta_1^{1/2})}$, $s = \sqrt{\frac{1}{2}(1 + \delta_1^{1/2})}$, and $\Omega_R^2 = \sqrt{\Omega_R^2 + \delta_1^2}$ is the generalized Rabi frequency. The energy separation between the dressed states $|I(N)⟩$ and $|II(N)⟩$ is $\hbar \Omega_R^2$. As shown in Fig. 1(c), there are three transition frequencies: one centered at the pump frequency $\omega_1$, and two Rabi side bands centered at frequency $\omega_1 \pm \Omega_R^2$.

Assuming $\Omega_R^2 >> \gamma$ and using the secular approximation, the steady state solutions for the dressed state population are

$$\rho_{II} = \frac{c^4}{c^4 + s^4}, \rho_{II,II} = \frac{s^4}{c^4 + s^4}.$$
We first set the pump detuning $\delta_1$ to be zero and scan the probe frequency across the trion transition frequency $\omega_1$. Figure 2(a) shows the probe absorption lineshape with a pump intensity of $95 \text{ W/cm}^2$. Instead of a lorentzian absorption lineshape in the absence of the pump, as shown at the bottom of the Fig. 2(a), the lineshape of the probe beam in the presence of a strong pump beam shows a complex structure [30]: a triplet-like absorption pattern appears with one weak central structure and two Rabi side bands with dispersive lineshape. The observation of the Rabi side bands is a signature of the optical generation of a single dot trion Rabi oscillations. The inset in Fig. 2(a) shows the Rabi splitting of the side bands as a function of the pump detuning. The largest Rabi frequency we achieved in the experiment is about $2\pi \times 1.6 \text{ GHz}$, limited only by the current experimental configuration.

The negative part of the absorption lineshape demonstrates gain of the probe beam. Since the pump is resonant with the trion transition, there is no population inversion in the steady state of the trion system in any picture. The gain effect comes from the coherent energy exchange between the pump and probe beams through the QD nonlinearity. We define the efficiency of the probe gain as the ratio of the amplitude of the negative absorption to the probe absorption in the absence of the strong pump. The probe gain efficiency corresponding to a pump intensity of $95 \text{ W/cm}^2$ is 5.3 %. The earlier work by Kroner et. al. [24] in a negatively charged dot did not show gain in their spectrum though they saw many of the other features consistent with strong field excitation. They attribute this difference to possible effects of dephasing.

As we tune the pump laser frequency away from the trion transition, the dispersion-like lineshapes of the Rabi side bands evolve into three spectral features: one weak central structure with a dispersive lineshape and two Rabi side bands with Lorentzian lineshapes. Figure 2(b) displays the probe absorption spectrum as a function of the pump detuning with a fixed pump intensity of $95 \text{ W/cm}^2$.

A distinct feature of the probe absorption spectrum is that one of the side bands shows purely negative “absorption”, which is the gain effect. Using the pump detuning at $-1.5 \text{ GHz}$ as an example (the bottom curve of Fig. 2(b)), there is an absorption peak located at $\omega_1 + \Omega_{R}^g$. This is an AC stark shifted absorption peak. The side band centered at $\omega_1 - \Omega_{R}^g$ is negative, which signifies the amplification of the probe beam. In lowest order perturbation theory, this reflects a three photon Raman gain effect: the QD absorbs two pump photons at frequency $\omega_1$ and emits a photon at $\omega_1 - \Omega_{R}^g$. The frequency at which gain occurs can be tuned by adjusting the pump detuning. As expected, if the pump detuning is positive, the probe sees gain at $\omega_1 + \Omega_{R}^g$. The data with pump detuned $+0.3 \text{ GHz}$ is shown at the top of Fig. 2(b). A gain peak is clearly observed for the positive detuning of the probe. It has been shown theoretically that the maximum gain occurs at the absolute value of the pump detuning $|\delta_1| = \Omega_{R}^g/3$ provided $\Omega_{R}^g >> \gamma$

![FIG. 2: (color online)(a) Top curve: trion Mollow absorption spectrum at a pump intensity of $95 \text{ W/cm}^2$ with resonant pumping. Bottom curve: a probe beam absorption spectrum with no pump. Inset: the Rabi splitting of the side bands as a function of the pump detuning. (b) Trion Mollow absorption spectrum with various pump detuning with a fixed pump intensity of $95 \text{ W/cm}^2$. Two Rabi side bands are clearly observed, where one is the AC Stark shifted absorption peak and the other shows gain. (c) The spectral position of the Rabi side bands as a function of the pump detuning. We use the trion transition energy as the zero point. The anti crossing feature of the Rabi side bands is demonstrated as the pump is detuned from the red to the blue of the trion transition. (d) The energy separation of the Rabi side bands as a function of the pump detuning. The solid blue line is the fits by the formula $2\sqrt{\delta_1^2 + \Omega_{R}^g}$.](image)

For the pump detuning $-0.3 \text{ GHz}$, the data shows a probe gain of 9.7 %, which is much larger than under resonant pumping with the same intensity. When the probe frequency is nearly degenerate with the pump beam, there is also a small dispersive structure in the probe absorption spectrum, as shown in Fig. 2(b).
The solid lines in Fig. 2(b) are theoretical fits of the data to Eq. (1). The fits yield $\gamma_T/2\pi$ and $\gamma/2\pi$ of $(580 \pm 90)$ MHz and $(350 \pm 35)$ MHz, respectively. Since $\gamma_T$ is almost twice $\gamma$, the amount of pure dephasing in this QD is statistically insignificant compared with the error bars. These fits show that our results can be well reproduced by the Optical Bloch equations and that the singly charged QD behaves like a single isolated atomic system.

Figure 2(c) shows the spectral positions of the Rabi side bands as a function of the pump detuning. In the plot, we use the trion transition frequency $\omega_{\text{tr}}$ as the zero energy point. Figure 2(c) clearly illustrates the anti crossing behavior of the Rabi side bands. The separation between the two peaks at zero pump detuning represents the interaction strength between the light and QD, equal to the Rabi frequency. The dotted curves in the plot are the theoretical predictions of the peak positions as a function of the detuning, which is in good agreement with the measurements. The laser light induced transition energy shifts at the large pump detuning are a demonstration of the dynamic, or AC Stark effect.

We extracted the energy separation of the side bands from the data and plotted it as a function of the pump detuning in Fig. 2(d). The solid blue line is the fit by the expression $2\sqrt{\Omega^2_R + \delta^2}$ and gives $\Omega_R/2\pi = (1.5 \pm 0.1) \text{ GHz}$. Since $\Omega_R = \mu \cdot E_{\text{pump}}/\hbar$, we infer the trion dipole moment of $(25 \pm 2) \text{ D}$. The trion dipole moment we calculated is similar to the reported neutral exciton dipole moment [10].

The Einstein A coefficient, or spontaneous emission rate, is [32]

$$\gamma_{sp} = \frac{9n^5}{(2n^2 + n_{QD}^2)^2} \frac{\omega_0^3 \mu^2}{3\pi \epsilon_0 \hbar c^3}$$

(2)

where $\gamma_{sp}$ is the spontaneous emission rate in the vacuum, $n$ and $n_{QD}$ are the refractive index of the medium and the QD, respectively. By inserting the parameters into Eq. (2), we get a spontaneous emission rate of $2\pi \times 130$ MHz, which corresponds to a trion radiative life time of 1.2 ns. Assuming there is no pure dephasing in the QD, as we shown earlier, then the trion transition linewidth is about 130 MHz, which is smaller than what we extracted from our previous fits. Also, the low power single beam absorption data yields a transition linewidth of 600 MHz, which is much larger than what we calculated from the Einstein A coefficient. This discrepancy could come from the spectral diffusion process, which broadens the trion transition linewidth [10, 33].

In summary, we have shown that an electron trapped inside a QD with its ground state and the excited two electron and one hole state behaves as an isolated quantum system even in the strong field limit by observing the optical Mollow absorption spectrum as well as the AC Stark effect.

The behavior is well described by the solutions to the optical Bloch equations for a two-level system and show that the state of the electron can be switched at a rate of $2\pi \times 1.6$ GHz with low power cw diode lasers.

This work is supported by the U.S. ARO, AFOSR, ONR, NSA/LPS, and FOCUS-NSF.

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* Electronic address: dst@umich.edu

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