

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

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We investigate a singly charged quantum dot under a strong optical driving field by probing the system with a weak optical field. We observe all critical features predicted by Mollow for a strongly driven two-level atomic system in this solid state nanostructure, such as absorption, the ac-Stark effect, and optical gain. Our results demonstrate that even at high optical field strengths the electron in a single quantum dot with its dressed ground state and trion state behaves as a well-isolated two-level quantum system.

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Quantum dot (QD) nanostructures have been proposed for numerous quantum mechanical applications due to their customizable atomlike 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 [5–7] 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. For an ideal two-level atomic system, it has been shown theoretically [10–12] and demonstrated experimentally [13,14] that the strong coupling leads to interesting spectral features, such as Rabi side bands in the absorption, and strikingly, the amplification of a probe beam, which is known as the Mollow absorption spectrum (MAS).

Because of the unique atomic properties of the QD system, many body effects which dominate the nonlinear optical response in higher dimensional heterostructures are strongly suppressed. Recently, the optical ac-Stark effect has been seen by exciting a neutral QD with a detuned strong optical pulse [15] while the MAS and Mollow triplets [16] have been observed in a single neutral QD [17,18] and a single molecule with intense resonant pumping [19].

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 system comprised of two electrons and a hole. The Fano interference effect, which arises from the coupling between a two-level system with a continuum [20], has been observed in a negatively charged QD [9,21].

The recent study of a single charged QD in the strong coupling regime does not exhibit the typical MAS [22]. All these indicate that interactions with a single charged QD could be more complex due to many-body effects than the electron-hole system reported earlier in neutral dots [17,18]. Interestingly, the results in this Letter 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 for a strongly driven two-level system.

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 line shapes. As the pump beam is detuned from the trion transition, we observe three spectral features: a weak dispersive line shape 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 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 ($|\pm \frac{1}{2}\rangle$) to the trion states ($|\pm \frac{3}{2}\rangle$) with $\sigma \pm$ polarized light excitations. Since the Zeeman sublevels 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 $|S\rangle$ and the excited state as $|T\rangle$, 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

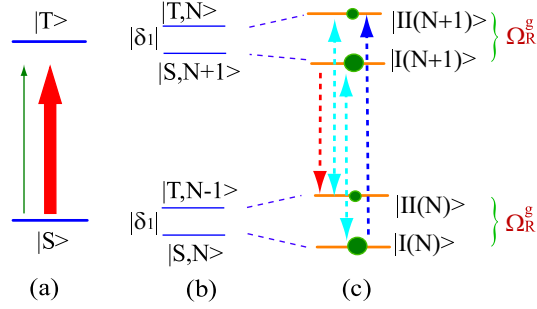


FIG. 1 (color online). (a) The energy level diagram of a trion state at zero magnetic field. The absorption spectrum of the weak probe (thin arrow) is modified by a strong pump field (thick arrow). (b) The uncoupled atom-field states. (c) Dressed state picture of a two-level system driven by a strong optical field. The energy levels outside the picture are not shown. The energy splitting between the dressed states with the same photon number is $\hbar\Omega_R^g$, where Ω_R^g is the generalized Rabi frequency.

significantly modified [10–12]. The absorption coefficient can be obtained by solving the optical Bloch equations to all orders in the pump field and first order in the probe field, as shown in Ref. [11,17]. When the strong pump is on resonance with the trion transition, i.e., the pump detuning $\delta_1 = 0$ and the Rabi frequency Ω_R is much larger than the transition linewidth 2γ , the probe will show a complex Mollow absorption spectrum, which has been discussed in detailed in Ref. [17], 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. 1(b)] map into the dressed states [Fig. 1(c)] when the QD-field interaction is included. In Fig. 1, we assume the pump detuning δ_1 to be negative, $|S\rangle$ and $|T\rangle$ are the quantum dot states, and N is the photon number. Because of the light-matter interaction, one set of the dressed states can be written as [23]

$$|I(N)\rangle = c|S, N\rangle - s|T, N-1\rangle,$$

$$|II(N)\rangle = s|S, N\rangle + c|T, N-1\rangle$$

where $c = \sqrt{\frac{1}{2}(1 - \frac{\delta_1}{\Omega_R^g})}$, $s = \sqrt{\frac{1}{2}(1 + \frac{\delta_1}{\Omega_R^g})}$, and $\Omega_R^g = \sqrt{\Omega_R^2 + \delta_1^2}$ is the generalized Rabi frequency. The energy separation between the dressed states $|I(N)\rangle$ and $|II(N)\rangle$ is $\hbar\Omega_R^g$. As shown in Fig. 1(c), there are three transition frequencies: one centered at the pump frequency ω_1 , and two Rabi side bands centered at frequency $\omega_1 \pm \Omega_R^g$.

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

$$\rho_{I,I} = \frac{c^4}{c^4 + s^4}, \quad \rho_{II,II} = \frac{s^4}{c^4 + s^4}.$$

It is clear when $\delta_1 < 0$, the dressed state $|I(N)\rangle$ is more populated than the dressed state $|II(N)\rangle$. In Fig. 1(c), the size of the dots on states $|I(N)\rangle$ ($|I(N+1)\rangle$) and $|II(N)\rangle$ ($|II(N+1)\rangle$) indicates their population. Therefore, the transition centered at $\omega_1 + \Omega_R^g$ represents probe absorption [the rightmost dashed line in Fig. 1(c)], and the transition centered at $\omega_1 - \Omega_R^g$ is probe gain due to the population inversion of the dressed states [the leftmost dashed line in Fig. 1(c)]. The gain process, in its simplest form, can also be considered as a three photon process, in which two pump photons are absorbed at frequency ω_1 and a third photon is emitted at frequency $\omega_1 - \Omega_R^g$ [14]. The middle dashed lines indicate the probe frequency is close to the pump frequency and the secular approximation fails. These can give rise to a dispersive line shape [14,24].

The experiment is performed on a singly charged self-assembled InAs QD embedded in a Schottky diode structure. The detailed sample information can be found in Ref. [17,25]. The sample is located in a continuous helium flow magneto cryostat at a temperature of 5 K. By varying the dc gate voltage across the sample, the charge state of the dot can be controlled [25,26] and the transition energies can be electrically tuned using the dc Stark effect [27]. When the dc Stark shift is modulated by a small ac voltage, the changes in the transmission signal can be detected at the modulation frequency by a phase-sensitive lock-in amplifier.

By setting the voltage modulation amplitude to about 16 times the transition linewidth, we avoid complexities associated with smaller modulations [27]. The data taken directly correspond to the absorption. To obtain the Mollow absorption spectrum, two continuous wave (cw) lasers are used. In the pump-probe experiment, we set both beams to be linearly polarized with orthogonal polarization. By filtering out the pump beam with a polarizer in front of the detector, we can measure the probe absorption only.

We first set the pump detuning δ_1 to be zero and scan the probe frequency across the trion transition frequency ω_o . Figure 2(a) shows the probe absorption line shape with a pump intensity of 95 W/cm². Instead of a Lorentzian absorption line shape in the absence of the pump, as shown at the bottom of the Fig. 2(a), the line shape of the probe beam in the presence of a strong pump beam shows a complex structure [28]: a tripletlike absorption pattern appears with one weak central structure and two Rabi side bands with dispersive line shape. The observation of the Rabi side bands is a signature of the optical generation of 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 intensity. The largest Rabi splitting we achieved in the experiment is about $2 \times \hbar\Omega_R = 13.2 \mu\text{eV}$, which corresponds to switching between the ground and trion states at a frequency of 1.6 GHz and is limited only by the current experimental configuration.

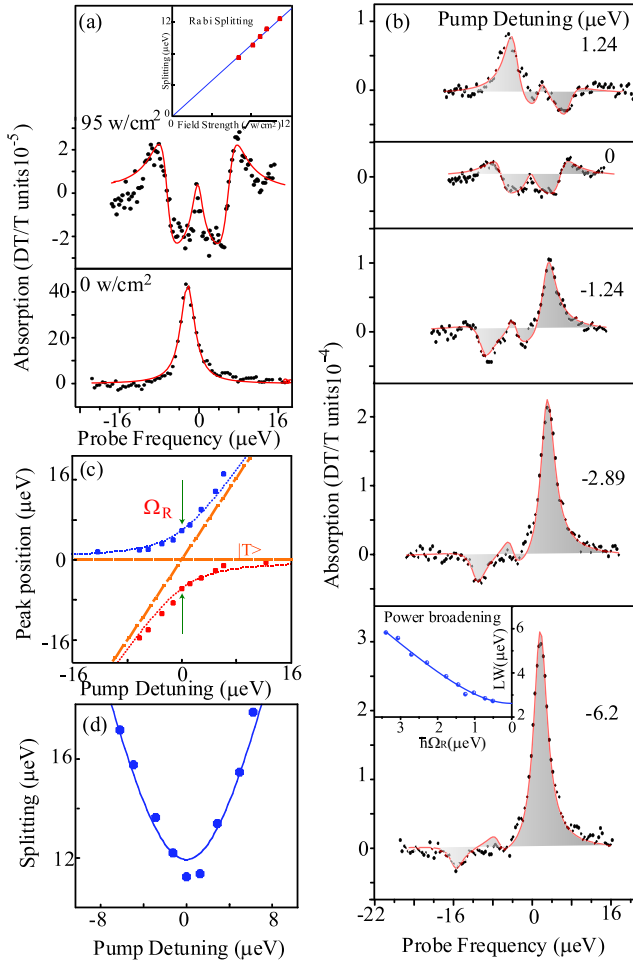


FIG. 2 (color online). The zero of the laser energy scale is 1318.797 meV. (a) Top curve: trion Mollow absorption spectrum at a pump intensity of 95 W/cm² with resonant pumping. Bottom curve: a probe beam absorption spectrum with no pump. Inset: the Rabi splitting of the side bands as a function of the pump intensity. (b) Trion Mollow absorption spectrum with various pump detuning with a fixed pump intensity of 95 W/cm². Two Rabi side bands are clearly observed, where one is the ac-Stark shifted absorption peak and the other shows gain. Inset: the trion linewidth of the one beam absorption spectrum as a function of laser Rabi frequency. It clearly shows the power broadening effect. (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 anticrossing 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 line is the fit.

The negative part of the absorption line shape 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 W/cm² is 5.3%. The earlier work by Kroner *et al.* [22] did not observe the typical spectral features for a isolated two-level system, such as the dispersive side bands with optical gain effect, and they attribute this difference to possible effects of dephasing.

As we tune the pump laser frequency away from the trion transition, the dispersionlike line shapes of the Rabi side bands evolve into three spectral features: one weak central structure with a dispersive line shape and two Rabi side bands with Lorentzian line shapes. Figure 2(b) displays the probe absorption spectrum as a function of the pump detuning with a fixed pump intensity of 95 W/cm².

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 $\hbar\delta_1 = -6.2 \mu\text{eV}$ 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 ω_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 $\hbar\delta_1 = 1.24 \mu\text{eV}$ are 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 $\Omega_R^g \gg \gamma$ [29]. For the pump detuning $\hbar\delta_1 = -1.24 \mu\text{eV}$, 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 trion decay rate $\hbar\gamma_T$ and decoherence rate $\hbar\gamma$ of $(2.4 \pm 0.4) \mu\text{eV}$ and $(1.45 \pm 0.15) \mu\text{eV}$, respectively. We also performed power dependent one beam absorption measurements. The extracted linewidth is plotted in the inset of Fig. 2(b) as a function of laser Rabi frequency, which clearly shows the power broadening effect. The fit with a equation of $\text{FWHM} = 2\hbar\gamma\sqrt{1 + \Omega_R^2/(\gamma\gamma_T)}$ yields respectively $\hbar\gamma_T$ and $\hbar\gamma$ of $(2.2 \pm 0.1) \mu\text{eV}$ and $(1.35 \pm 0.1) \mu\text{eV}$, which agrees well with the extracted parameters from the MAS. Since γ_T is almost twice γ , these fits show that our results can be well reproduced by the optical Bloch equations and that there is no significant pure dephasing in the QDs.

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 ω_o as the zero energy point. Figure 2(c) clearly illustrates the anticrossing 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 line is a fit by the expression $2\sqrt{\Omega_R^2 + \delta_1^2}$ and gives $\hbar\Omega_R = (6.2 \pm 0.4 \mu\text{eV})$. Since $\Omega_R = \mu E_{\text{pump}}/\hbar$, we infer the trion dipole moment of $(25 \pm 2) D$. The trion dipole moment we calculated is similar to the reported neutral exciton dipole moment [17].

The Einstein A coefficient, or spontaneous emission rate, is [30]

$$\gamma_{\text{sp}} = \frac{9n^5}{(2n^2 + n_{\text{QD}}^2)^2} \frac{\omega_o^3 \mu^2}{3\pi\epsilon_o \hbar c^3} = \frac{9n^5}{(2n^2 + n_{\text{QD}}^2)^2} \gamma_{\text{spo}} \quad (1)$$

where γ_{spo} 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. (1), we obtain a spontaneous emission rate of $\hbar\gamma_{\text{sp}} = 0.54 \mu\text{eV}$, which corresponds to a trion radiative life time of 1.2 ns. Assuming there is no pure dephasing in the QD, as we have shown earlier, then the trion transition linewidth is about $\hbar\Gamma_T = 0.54 \mu\text{eV}$, which is smaller than what we extracted from our previous fits from MAS or power broadening data. This discrepancy could come from the spectral diffusion process, which broadens the trion transition linewidth [17,31].

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