Solid State Communications 149 (2009) 1479–1484

Contents lists available at ScienceDirect

Solid State Communications

journal homepage: www.elsevier.com/locate/ssc

Strong optical field study of a single self-assembled quantum dot

Xiaodong Xu [a,1], Bo Sun [a], P.R. Berman [a], Duncan G. Steel [a,*], Daniel Gammon [b], L.J. Sham [c]

[a] The H. M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, MI 48109, USA
[b] Naval Research Laboratory, Washington, DC 20375, USA
[c] Department of Physics, University of California San Diego, La Jolla, CA 92093, USA

ARTICLE INFO

Article history:
Received 4 February 2009
Accepted 27 April 2009 by the Guest Editors
Available online 18 May 2009

PACS:
78.67.HC
42.50.Gy
42.50.Hz
78.47.Fg

Keywords:
A. Nanostructure
D. Optical properties
E. Nonlinear optics

ABSTRACT

We review the investigation of a single quantum dot driven by a strong optical field. By coherent pump-probe spectroscopy, we demonstrate the Autler–Townes splitting and Mollow absorption spectrum in a single neutral quantum dot. Furthermore, we also show the typical Mollow absorption spectrum by driving a singly charged quantum dot in a strong optical coupling regime. Our results show all the typical features of an isolated atomic system driven by a strong optical field, such as the AC stark effect, Rabi side bands and optical gain effect, which indicate that both neutral and charged quantum dots maintain the discrete energy level states even at high optical field strengths.

1. Introduction

Semiconductor Quantum dots (QDs) are customizable solid state analogs of atoms [1]. Due to their unique atomic like physical properties, QDs have been proposed in numerous quantum mechanical based applications, one of which is optically driven quantum information processing and computation [1,2]. Significant research progress has been made in this direction, such as initial qubit state preparation [3–8], coherent optical manipulation of a single qubit [9–15], and quantum logic gates based on QD qubits [13,16,17].

One topic that has drawn a lot attention recently is the investigation of QD systems driven by strong optical fields [18–25]. The coupling strength between the light field and QD transition is represented by the Rabi frequency $\Omega_R = \frac{\mu \cdot E_x}{\hbar}$, where μ is the transition dipole moment and E_x is the optical field strength. The QD system is driven in the strong coupling regime if Ω_R is comparable or larger than the transition linewidth. In atomic systems, it has been shown that the strong coupling can lead to interesting spectral features, such as triplets in the emission spectrum, known as Mollow triplets [26], and an optical gain effect in the absorption spectrum [27,28], known as Mollow absorption spectrum (MAS) [29].

The opportunities for observing these quantum optics effects in the QD system come from its atomic properties, which suppresses the many body effects which exist in higher dimensional semiconductor systems. In addition, a QD has a much larger dipole moment compared to atomic transitions. The research progress on the strong coupling between laser light and QDs has grown rapidly in the last couple of years. Autler–Townes splitting [30], optical AC stark effect, Mollow triplets, and MAS all have been demonstrated for a single neutral exciton [18,21], bi-exciton [20,22], and charged QD systems [19,25].

In this paper, we use coherent pump-probe spectroscopy to study the dynamics of a strongly driven quantum dot. For a single neutral QD, we utilize the V system formed by the two polarized neutral exciton states. A strong pump is fixed to be resonant with one exciton transition. If the weak probe scans across the coupled exciton transition, we observe two Lorentzian lineshapes, known as the Aulter–Townes (AT) doublet. If the weak probe is on the same transition as the strong pump, we observe MAS, which is composed of a weak center peak and two Rabi side bands with dispersive lineshapes. The negative part of the

* Corresponding author.
E-mail address: dst@umich.edu (D.G. Steel).
1 Present address: Center for Nanoscale Systems, Cornell University, Ithaca, New York 14853, USA.

0038-1098/$ – see front matter © 2009 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ssc.2009.04.042

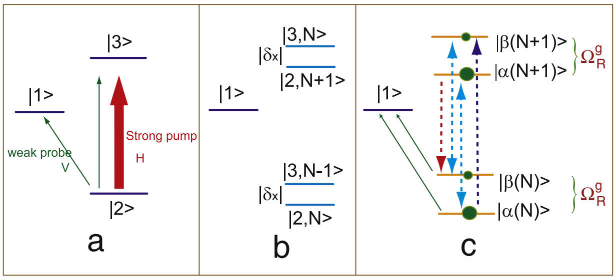

Fig. 1. Bare state and dressed state pictures of a QD driven by a strong optical field. (a) A strong pump is in resonance with transition H and a weak beam probes either transition H or transition V. Dressed state picture (b) without (c) and with light-transition coupling. The dots represent the dressed state populations with a red detuned pump.

lineshape indicates the probe gain effect. We further demonstrate the MAS of a single charged QD with the pump beam both on and detuned from resonance. When the pump is detuned, the probe absorption spectrum has three spectral features: one weak dispersive lineshape at the pump frequency, one AC stark shifted Rabi side band due to the absorption, and one Rabi side band showing a probe gain effect. All the experimental observations can be explained by the standard optical Bloch equations used for atomic systems. Our results not only demonstrate the coherent nonlinear interaction between laser light and a single QD oscillator, they also show that both neutral and singly-charged QDs maintain discrete energy levels even when driven by a strong optical field.

The paper is organized as following. We first give the physical model and theoretical explanation of the AT splitting and optical MAS. Then we present the experimental results of a strongly optical driven neutral QD. Finally, we will discuss the MAS of a singly charged QD.

2. Theory

When an atomic transition is driven by a strong optical field, shown in Fig. 1(a), the atomic states are dressed by the laser fields, as shown in Fig. 1(b), where N is the number of laser photons. Taking into account the coupling between the light and the atomic transition, the system can be represented by the dressed states in Fig. 1(c) [31], where the separation of the dressed states in each manifold is the coupling strength $\hbar\Omega_R^g$, where Ω_R^g is the generalized Rabi frequency. In order to solve for the probe absorption analytically, we follow the method developed in Refs. [29,32] and describe our system with the optical Bloch equations $\frac{d\hat{\rho}}{dt} = \left[\hat{H}, \hat{\rho}\right] + Decay$, where $\hat{\rho}$ is the density matrix and $\hat{H}$ is the Hamiltonian.

We first consider the case where the weak beam probes the transition from state 2 to state 1. By solving the density matrix equations to all orders of the pump and to first order of the probe, we get the weak probe absorption [18]

$$\alpha_{AT} = \alpha_o \mathrm{Im}\left\{ \mathrm{i}\frac{4\left[\gamma_{13} - \mathrm{i}\left(\delta_p - \delta_x\right)\right]\rho_{22}^{(0)} - 2\mathrm{i}\Omega_R\rho_{32}^{(0)}}{4\left(\gamma_{21} - \mathrm{i}\delta_p\right)\left[\gamma_{13} - \mathrm{i}\left(\delta_p - \delta_x\right)\right] + \Omega_R^2} \right\} \tag{1}$$

where $\alpha_\circ$ is a constant, $\gamma_{13} = \frac{\gamma_1+\gamma_3}{2} + \Gamma_{13}$, $\gamma_{21} = \frac{\gamma_1}{2} + \Gamma_{21}$, $\gamma_{23} = \frac{\gamma_3}{2} + \Gamma_{23}$, Γ_{ij} is a dephasing rate of transition ij, γ_i is the population relaxation rate of state i, $\delta_x = \omega_{32} - \omega_x$, $\delta_p = \omega_{12} - \omega_p$, $\omega_x(\omega_p)$ is the pump (probe) field frequency, and ω_{ij} is the atomic frequency of transition ij. Under the condition of $\Omega_R >> \gamma_i$, Γ_{ij}, the probe absorption profile separates into two Rabi side bands located at E_+ and E_-, where $E_\pm = \frac{1}{2}\left(\delta_x \pm \sqrt{\Omega_R^2 + \delta_x^2}\right)$, as shown by the dressed state picture in Fig. 1(c). These two side bands are known as the Autler-Town doublet. If δ_x is larger than Ω_R, the absorption peaks are AC stark shifted. If the pump is tuned to be resonant with transition ω_{23}, i.e. $\delta_x = 0$, the two peaks are symmetric, separated in energy by $\hbar\Omega_R$.

The probe absorption spectrum changes dramatically if the probe scans the same transition as the pump. Again, solving the optical Bloch equations to all orders of the pump and to first order in the probe, we get the absorption coefficient of the probe beam as in Box I, where $\Delta = \delta_x - \delta_p$ is the probe detuning from the pump, $A = \gamma_{23} - \mathrm{i}\delta x$ and $B = \gamma_{23} + \mathrm{i}\Delta$. When the pump is in resonance and Ω_R is larger than γ_{23}, we can get a simplified form for equation in Box I. If $\delta_p \simeq \pm\Omega_R$, we find

$$\alpha_{MS} = \pm\alpha_o \frac{\gamma_3}{\Omega_R} \frac{\left(\delta_p \mp \Omega_R\right)}{4\left(\delta_p \mp \Omega_R\right)^2 + \left(\gamma_3 + \gamma_{23}\right)^2}. \tag{2}$$

Eq. (2) shows that there are two dispersive line shapes as a function of the probe frequency centered at $\pm\Omega_R$ with their zero crossings at $\delta_p = \pm\Omega_R$. When $\left|\delta_p\right| > \Omega_R$, the probe experiences absorption. Interestingly, the probe absorption is negative given $\left|\delta_p\right| < \Omega_R$, i.e. the probe sees gain. Since the pump field is in resonance, this corresponds to a gain effect without a population inversion in any picture. The wings of the dispersive line shapes also provide nearly constant gain near $\delta_p = 0$. However, the gain is canceled by a weak absorptive component exactly at $\delta_p = 0$. Explicitly, near $\delta_p = 0$ one finds

$$\alpha_{MS} = -\frac{\alpha_o}{2}\left(\frac{\gamma_{23}}{\Omega_R}\right)^2 \frac{\gamma_3}{\gamma_{23}} \left(\frac{\delta_p^2/\gamma_{23}}{\delta_p^2 + \gamma_{23}^2}\right).$$

The above calculation of the MAS is for a resonant pump field. If the pump is detuned from the atomic transition, the probe absorption lineshapes are quite different. When the pump detuning $|\delta_x|$ is comparable or larger than transition linewidth, a simple physical picture can be derived in the fully quantized dressed state picture. Assuming the pump is tuned to the red of the atomic transition, the quantized dressed states can be written as [31]

$$|\alpha\,(N)\rangle = c\,|2, N\rangle - s\,|3, N-1\rangle$$
$$|\beta\,(N)\rangle = s\,|2, N\rangle + c\,|3, N-1\rangle$$

where $c = \sqrt{\frac{1}{2}\left(1 + \frac{\delta x}{\Omega_R^g}\right)}$, $s = \sqrt{\frac{1}{2}\left(1 - \frac{\delta x}{\Omega_R^g}\right)}$ and the generalized Rabi frequency $\Omega_R^g = \sqrt{\Omega_R^2 + \delta_x^2}$.

The population of the dressed states can be solved by ignoring the coherence between the dressed states. The steady state solutions are

$$\rho_{\alpha,\alpha} = \frac{c^4}{c^4 + s^4}, \qquad \rho_{\beta,\beta} = \frac{s^4}{c^4 + s^4}.$$

The above solutions show that under the negative detuning of the pump, $\delta_x > 0$, the dressed state $|\alpha\,(N)\rangle$ is more populated than $|\beta\,(N)\rangle$. In Fig. 1(c), we use the size of the dots to represent the dressed state population. There are three transitions shown by Fig. 1(c). Since $|\alpha\,(N)\rangle$ is more populated than $|\beta\,(N+1)\rangle$, the transition centered at $\omega_x + \Omega_R^g$ is an AC stark shifted absorption peak (the purple dashed line). The state $|\alpha\,(N+1)\rangle$ is more populated than $|\beta\,(N)\rangle$and hence, the population inversion of the dressed states induces a probe gain centering at $\omega_x - \Omega_R^g$ (the red dashed line). When the probe frequency approaches the pump, there is a weak dispersive lineshape (light blue dashed line). This feature cannot be derived from the previous method, as it relies on the coherence between the dressed states.

3. Experiments

The experiment is performed on single self-assembled QDs embedded in a Schottky diode structure [33,34]. There is an Al