Decoherence of a resonantly driven exciton in a single quantum dot

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Abstract. We report on coherent control of the resonantly driven exciton state in a single quantum dot in the strong coupling regime. With picosecond laser pulses, the two-level system undergoes Rabi oscillations and a given coherent superposition of states can be addressed. Using a pair of phase-locked pulses, optical manipulation is achieved during the coherence time of the qu-bit. The dephasing processes are also studied and we show that in typical quantum dots, the coherence time $T_2$ can be of the same order of magnitude as the effective lifetime of the exciton $T_1$ (a few hundreds of ps) although not reaching the upper theoretical limit of $2T_1$. We conclude that energy relaxation and pure dephasing processes due to virtual scattering with phonons contribute on an equal footing to the loss of coherence.

1. Introduction

Semiconductor quantum dots (QDs) have attracted considerable interest since they appear as promising candidates to realize quantum bits for future quantum information processing. To this aim, a fundamental requirement is the coherent manipulation of QDs, which is severely limited by the decoherence mechanisms arising from the strong interaction with their environment and thus limiting the number of possible quantum operations. Apart from radiative recombination, these decoherence mechanisms are due to interaction with phonons and/or fluctuations of the electrostatic background.

In order to take full advantage of the longest possible coherence time, it is more interesting to drive the lowest lying level of the exciton, since it is the least interacting state with its surrounding. However, resonant optical spectroscopy remains a real experimental challenge. Only recently, resonant luminescence has been investigated by inserting the QDs in microcavities [1]. Photocurrent measurements have also been performed under resonant excitation of single QDs in Schottky diodes [2]. We choose an alternative approach by embedding the QDs in a one-dimensional waveguide for the confinement of the driving-field, so the resonance luminescence can be detected from the waveguide top surface [3]. This experimental configuration combines micro-photoluminescence (µPL) and waveguiding geometry that allows an all-optical control of the driven state. Moreover, the reduced volume of the optical mode in the one-dimensional waveguide, together with a large absorption cross-
section of the semiconductor QD, yields an enhanced light-matter interaction which reduces the effective lifetime of the exciton.

Coherent control (CC) of a single dot has also been achieved [4] using a pair of phase-tailored laser pulses. By addressing different excitation regimes, depending on pulse delay and power, we show that CC provides a useful tool not only to manipulate the excitonic state but also to measure its relaxation-time constants.

2. Resonant Rabi regime

The QDs of interest consist of InAs self-assembled QDs grown by molecular beam epitaxy on a GaAs substrate. The InAs wetting layer and the QDs are embedded in a 1 µm GaAs layer, between two AlGaAs layers. This structure forms a 2D waveguide, the GaAs being its core and the AlGaAs its barrier. 1 µm ridges are then wet-etched on the surface of the sample in order to obtain a 1D waveguide (figure 1). The density of QDs is small enough to be spectrally resolved.

The sample is fixed on the cold finger of a three optical access cryostat and cooled down to 10 K. The excitation is provided by a tunable Ti-Sapphire laser with 1 ps pulses. The pump beam is focused with a large numerical aperture microscope objective, in front of the sample edge. Then, it propagates along the 1D waveguide and interacts with the QDs. The luminescence is then collected from the top surface of the sample by another microscope objective. Since the laser light is confined in the guided mode, the scattered light is greatly suppressed and the resonant luminescence is almost background free, at low pump power. Experimental details can be found elsewhere [3,4].

![Figure 1](a) Schematic experimental guided-wave configuration showing the 1D optical mode and the polarization axes (x and y). (b) µPL maximum intensity extracted from spectra like shown in the inset, for on-resonance excitation in an InAs/GaAs QD demonstrating the Rabi oscillations versus square-root of incident power. The solid curve is a data simulation using the optical Bloch equations.

The optical eigenmodes of the waveguide are linearly polarized (y for TE mode and z for TM) and the QD eigenstates are linearly polarized along x and y. Thus, for low pump power resonant experiments, the QD µPL and the laser should be in principle, copolarized. However, in the case of self-assembled QDs we have very often observed luminescence in both polarization directions, suggesting that the QDs are tilted from the crystallographic axis [5]. The splitting of the exciton fine structure is of the order of 10 µeV and has been measured using a Fabry-Pérot interferometer placed in the luminescence path, because the spectral resolution of our set-up (40 µeV) does not allow spectrally resolving the splitting.

When the pump is resonant with the lowest lying exciton state in the dot and as the pump power is increased, we observe oscillations of the luminescence, demonstrating strong coupling between the QD and the driving field (figure 1 b). The number of periods is limited by the laser scattering as the pump power is increased, but we also observe that the oscillations are damped. Very recently, in [6] interaction with acoustic phonons has been proposed as the main source for the intensity damping of Rabi oscillations (RO). However, in our case the damped RO can be well reproduced only by
introducing a power-dependent phenomenological parameter in the population term of the Bloch equations [3]. This means that there is a very fast relaxation of the population as power is increased. It has also to be related to the increase of the spontaneous emission rate observed in the resonant time-resolved experiments. Indeed, the exciton effective radiative lifetime is reduced by about 50% for InAs dots. We ascribe this effect to an enhanced coupling of the emitted photons to the guided 1D mode arising from a modification of the refractive index that leads to a reduction of the propagating pulses group velocity. This phenomenon of “slow light” is due to self-induced transparency of the dipole coupled to the one-dimensional optical mode, a similar effect observed in QDs coupled to photonic crystals [7]. Recently, slow light effects have been investigated in QDs coupled to photonic crystals waveguides exploiting the waveguide dispersion [8]. In our case, the waveguide dispersion leads to a very small modification of the refractive index, only a few %.

Rabi oscillations experiments allow addressing one qu-bit, i.e. to choose the proper superposition of the two quantum levels. The final state \( |\psi\rangle = \cos \left( \frac{\theta}{2} \right) |0\rangle + \sin \left( \frac{\theta}{2} \right) |1\rangle \) is completely determined by the pulse area \( \theta \), which is the time integrated Rabi frequency: \( \theta = \int_{\infty}^{\infty} \Omega(t) dt = \int_{\infty}^{\infty} \frac{\mu E(t)}{\hbar} dt \), \( \mu \) being the dipole moment of the transition and \( E(t) \) the pulse envelope. For typical InAs QDs, the transition dipole moment is 25 D and the Rabi coupling constant is of the order of one meV.

3. Coherent control of a single qu-bit

The optical manipulation of the prepared state can then be achieved using a pair of phase-locked pulses. The delay between the pulses \( \delta \), and the relative phase \( \phi \) are well controlled using a stabilized Michelson interferometer on a scale of \( \lambda / 100 \). The most interesting case for investigating the coherence properties of the system is when \( \pi/2 \) pulses excite the QD. The first pulse creates a state with the maximum coherence and the second one, depending on its relative phase, can interfere constructively or destructively, thus enhancing or annihilating the dipole oscillation [4]. Measuring the resonant luminescence as a function \( \phi \) and \( \delta \) yields the total coherence time \( T_2 \) of the state. Figure 2 shows an example of coherent control experiments, where the population oscillation is observed as a function the phase \( \phi \) for a given delay. The broad peak on which is sitting the sharp \( \mu \text{PL} \) line is due to the spectrally broad laser line (1 ps laser pulse corresponds to a 0.5 meV wide spectral line).

![Figure 2 (a). Resonant luminescence spectra of a QD pumped by two \( \pi/2 \) pulses in phase \((\phi = 0)\) and out of phase \((\phi = \pi)\) delayed by 10 ps. (b) The population oscillates as a function of \( \phi \) at a given delay \( \delta \).](image)

The contrast between maximum \((\phi = 0)\) and minimum \((\phi = \pi)\) luminescence as a function of delay \( \delta \), is shown in figure 3. The exponential decay depends on both the state coherence time \( T_2 \) and its effective lifetime \( T_1 \) and has the following expression: \( C(\delta) = \frac{e^{-\delta/T_2}}{2 - e^{-\delta/T_1}} \). Independent time-resolved
measurements allow fixing the effective lifetime $T_1$, thus the only adjustable parameter is the coherence time $T_2$. In several dots, we find that the coherence time is of the same order of magnitude as the effective lifetime.

![Figure 3](image)

Figure 3. Quantum interference contrast as a function of delay $\delta$ (solid circles). The solid line corresponds to a fit with fixed $T_1=400$ ps (see text). The dephasing time obtained is $T_2=160$ ps.

The decoherence probability rate, $1/T_2$ can be written as: $$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T^*},$$ $T_1$ represents the exciton effective lifetime (radiative recombination and inelastic scattering with phonons) and $T^*$ represents the elastic pure dephasing processes (virtual phonons, and environmental charge fluctuations). For the ground exciton state that we are probing, phonon absorption at low temperature has been calculated to be very inefficient [4], typically of the order of 1 ns. Thus $T_1$ is limited only by emission of photons. As $T_2$ is less than the theoretical upper limit of $2T_1$, then pure dephasing processes are also relevant. In the example given above, $T^*$ is of the order of 250 ps. Due to low enough temperature, polarons are not formed, as confirmed by the absence of lateral wings in the emission line. Therefore, the pure dephasing contribution is due to virtual phonon scattering mechanisms or environmental charge fluctuations if they exist.

4. Conclusion
Coherent control in the strong coupling regime has been achieved on the ground exciton state in a single QD. Resonant excitation in the 1D guided-wave geometry allow to observe long coherence time of the ground exciton state and faster exciton radiative lifetime. These are basic requirements for realizing single indistinguishable photon sources with high generation rate. The role of acoustic phonons in the dephasing processes has to be further investigated and CC experiments as a function of temperature are currently carried out. For future applications, the collection efficiency remains a challenge and new improvements of the structures design have to be made.

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