Decoherence processes during optical manipulation of excitonic qubits in semiconductor quantum dots

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Using photoluminescence spectroscopy, we have investigated the nature of Rabi oscillation damping during optical manipulation of excitonic qubits in self-assembled quantum dots. Rabi oscillations were recorded by varying the pulse amplitude for fixed pulse durations between 4 ps and 10 ps. Up to five periods are visible, making it possible to quantify the excitation dependent damping. We find that this damping is more pronounced for shorter pulse widths and show that its origin is the nonresonant excitation of carriers in the wetting layer, most likely involving bound-to-continuum and continuum-to-bound transitions.

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The current topic of quantum computation presents a wide range of challenges to physical science,[1] particularly the search for candidates for solid-state quantum bits (qubits). Semiconductor quantum dots (QDs) are attractive because they possess energy structures and coherent optical properties similar to, and dipole moments larger than, those of atoms.[2,3] Efforts in the past few years have led to successful observations of Rabi oscillations (ROs) of excitonic states,[4–6] the hallmark for optical manipulation of qubits in QDs. However, all found that ROs damped out very quickly when the external field is increased. Because QDs contain a macroscopic number of atoms, this strong decoherence process must be due to unwanted coupling to other degrees of freedom. Identification of the underlying mechanism is difficult precisely because of this macroscopic nature. Yet such understanding plays the most crucial role in future development of quantum information technology in semiconductors, because strong laser fields imply high qubit rotation frequencies. Through manipulations of high quality factor excitonic qubits in InGaAs QDs, we have studied the underlying mechanism for decoherence processes during optical manipulation, i.e., processes occuring while the qubit is being actively rotated by strong fields. More specifically, we have found that this strong decoherence process is manifested through indirect excitations of carriers in the wetting layer (WL).

Self-assembled $In_{0.5}Ga_{0.5}As$ QD (SAQD) samples were grown by molecular beam epitaxy.[7] These QDs have an average lateral size, height, and dot-to-dot distance of 20–40 nm, 4.5 nm and 100 nm, respectively. Three excitonic levels are involved in this study [Fig. 1(a)]. The laser is resonant with the transition from the exciton vacuum state $|0\rangle$ to the first excited exciton state $|1\rangle$. The population in state $|1\rangle$ is known to relax nonradiatively to the exciton ground state $|2\rangle$ before radiative recombination to $|0\rangle$ occurs and is detected as the PL signal.[8,9]

The qubit formed by $|0\rangle$ and $|1\rangle$ is manipulated by a pulsed Ti:sapphire laser with the pulse width adjustable from 4 to 10 ps. The PL is collected along the direction normal to the sample (maintained at 5 K), dispersed by a spectrograph and imaged using a liquid nitrogen cooled two-dimensional array detector. Shown in Fig. 1(b) is the spectral image of the particular QD used for this study on an unprocessed sample. Light with linear polarization was used in such a way as to excite only one transition in the fine-structure split doublet.[10]

Figure 1(c) shows the PL intensity as a function of the square root of the average laser intensity. In each data series, the laser pulse width is fixed while the intensity is varied over several orders of magnitude. The oscillations of the PL intensity correspond to the ROs described above that the population of state $|1\rangle$ undergoes as a function of the input pulse area, $\theta=(\mu/\hbar)\int_{-\infty}^{\infty}\varepsilon(t')dt'$,[11] which is proportional to the square root of the average laser intensity. Here μ is the transition dipole moment (measured to be 40 Debye) and $\varepsilon(t)$ the electric field envelope. Furthermore, the variation of the periodicity in the three oscillations of Fig. 1(c) agrees quantitatively with the fact that the pulse area should be proportional to the square root of the pulse width at the same average laser power. One also observes that at a fixed pulse width τ_p the RO amplitude is damped out as θ increases. This is emphasized in Fig. 1(d) where we plot the amplitudes extracted from the oscillations in Fig. 1(c) at $\theta=\pi$, 2π, etc., on a logarithmic scale. In addition, the smaller the pulse width τ_p, the faster the RO is damped out when the laser intensity is increased. However, at $\theta<2\pi$, the smaller pulse width actually results in larger RO amplitude.

One fundamental question arises: "Does the extra damping occur only during the manipulation pulse or does it persist even after the pulse is over?" To answer this question we performed wave-packet interferometry experiments under different excitation intensities on the same QD. The quantum interference amplitude is measured while varying the time

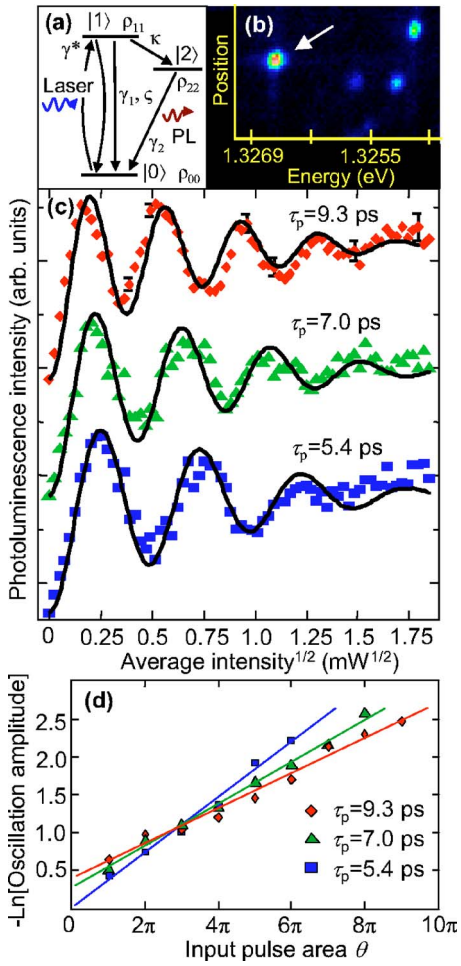

FIG. 1. (Color online) ROs of the upper state in the excitonic three-state system and its PL detection. (a) QD energy diagram. The QD is resonantly excited to the first excited excitonic state $|1\rangle$. The population that relaxes nonradiatively to the excitonic ground state $|2\rangle$ is eventually emitted and detected as the PL signal. The different decay channels and their rate constants are denoted by arrows. (b) Spectral image of QDs excited at 1.3418 eV. The QD investigated (arrow) is well isolated, both spatially and spectrally. The total vertical dimension is about 10 μm. (c) ROs for different pulse widths. The PL from the $|2\rangle$ to $|0\rangle$ transition was recorded while the average intensity was varied for fixed pulse width τ_p. The fit (solid lines) was obtained by numerical integration of the density matrix equations using a pure dephasing term proportional to the intensity. (d) Negative logarithm of the oscillation amplitude plotted versus the input pulse area θ. The data points are taken from the peaks and valleys of the ROs shown in (c), corresponding to the points where $\theta=n\pi$. The straight lines are guides to the eye to show the qualitative trend of the damping at different pulse widths.

delay between the pulses for a given single pulse input area. This measurement probes the decoherence rate in the time interval *between* two laser pulses. A detailed description of this procedure is given elsewhere for both linear[3] and nonlinear excitation regimes.[6] We find that T_2 decreases by a factor of 2 from $\theta \ll \pi$ ($T_2=48\pm5$ ps) to $\theta=2.5\pi$ ($T_2=24\pm5$ ps). This shows that the RO damping of Fig. 1(c) indeed originates in an excitation dependent dephasing term which persists even after the end of the pulse.

To fully capture the dynamics of RO damping, the excitonic state is described by a three-level system where the laser exclusively interacts with the $|0\rangle \rightarrow |1\rangle$ transition. The middle state $|2\rangle$ acts as a shelving state whose time integrated population $\int_0^\infty \rho_{22} dt$ is proportional to the PL intensity. The density matrix equations read

$$\frac{d\rho_{11}}{dt} = i\frac{\Omega}{2}(\rho_{10}-\rho_{01}) - (\gamma_1+\kappa+\zeta)\rho_{11},$$

$$\frac{d\rho_{00}}{dt} = -i\frac{\Omega}{2}(\rho_{10}-\rho_{01}) + \gamma_1\rho_{11} + \gamma_2\rho_{22} + \zeta\rho_{11},$$

$$\frac{d\rho_{01}}{dt} = -i\delta\rho_{01} - i\frac{\Omega}{2}(\rho_{11}-\rho_{00}) - \left(\frac{\gamma_1+\kappa+\zeta+2\gamma^*}{2}\right)\rho_{01},$$

$$\frac{d\rho_{22}}{dt} = -\gamma_2\rho_{22} + \kappa\rho_{11},$$

where $\Omega=\Omega(t)=\mu\varepsilon(t)/\hbar$ is the Rabi frequency, $\delta=\omega_0-\omega$ is the detuning from the resonance frequency ω_0 of the $|0\rangle$ to $|1\rangle$ transition (ω is the laser frequency) and $\gamma_{1,2}, \kappa, \zeta, \gamma^*$ are damping terms whose effect is depicted in the energy diagram of Fig. 1(a). γ_1 and γ_2 denote the radiative recombination rates of state $|1\rangle$ and $|2\rangle$, respectively. κ is the decay rate from state $|1\rangle$ to state $|2\rangle$ which primarily occurs via phonon emission.[9] γ^* describes pure dephasing (dephasing without population relaxation) and ζ is an additional decay rate from state $|1\rangle$ to state $|0\rangle$ that accounts for all other processes that scatter the exciton out of state $|1\rangle$ without decaying into $|2\rangle$. Radiative lifetimes in our sample are generally larger than 500 ps so that $\gamma_1, \gamma_2 \ll 1/\tau_p$ and thus they play no significant role in the dephasing process. The overall dephasing rate of $|1\rangle$ then becomes

$$\frac{1}{T_2} = \frac{\kappa+\zeta}{2} + \gamma^*.$$

From numerical integration of the density matrix equations at exact resonance ($\delta=0$) and with the initial conditions $\rho_{11}=\rho_{22}=0$ and $\rho_{00}=1$ one finds that there can be no decay of the RO amplitude with intensity unless the excited state dephasing rate increases with intensity. Throughout we assume that at low intensity, $\kappa=2/T_2^{(0)}$ where $T_2^{(0)}$ is obtained from wave-packet interferometry.

In principle, any of the three parameters, κ, ζ, and γ^* can depend on the excitation intensity and result in intensity dependent damping of ROs. However, each affects the damping in a different way as shown in Fig. 2. We simulated the ROs with either κ, ζ, or γ^* proportional to the intensity in such a way as to give rise to proper intensity dependent damping. Clearly, the oscillations only damp out symmetrically when γ^* is intensity dependent. We have found that all three curves in Fig. 1(c) can be fitted with a pure dephasing term of the form $\gamma^*=c\cdot I/\tau_p \propto \Delta\lambda \cdot I$ where $\Delta\lambda \propto 1/\tau_p$ is the laser bandwidth, I is the average laser intensity, and $c=0.4$ mW^{-1} [solid lines in Fig. 1(c)]. While we cannot completely exclude a more complicated relationship between γ^* and I and/or a combination of intensity dependent parameters we

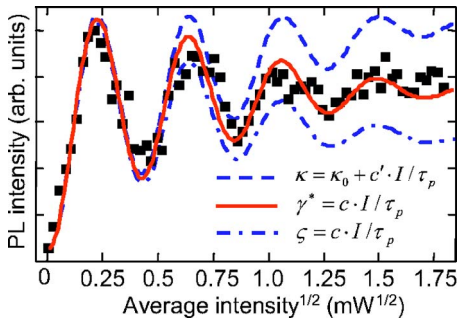

FIG. 2. (Color online) Simulated ROs assuming various intensity dependent decoherence terms. Plotted is the case when either $\kappa = \kappa_0 + c' \cdot I / \tau_p$ (dashed curve), $\zeta = c \cdot I / \tau_p$ (double-dashed curve), or $\gamma^* = c \cdot I / \tau_p$ (solid curve). The data for $\tau_p = 7.0$ ps is plotted as a reference (squares).

believe the above choice to be most reasonable. Thus we conclude that the RO damping during optical manipulation is primarily due to the additional pure dephasing term induced at high excitation intensity. This behavior also rules out the mechanism resulting from coupling to delocalized excitons, proposed in Ref. 5 for interface fluctuation QDs (IFQDs) since that mechanism will take the excitonic state out of the QDs and will give rise to totally different overall behavior. This is not surprising since the energy confinement in SAQDs is much higher than that in IFQDs. We note that although pure dephasing does not play an important role in IFQDs,[12,13] its manifestation has been reported in SAQDs.[14]

What could be the underlying mechanism? The lattice mediated dephasing model proposed in Ref. 15 showed that the RO amplitudes decrease with the laser intensity. However, the pulse width dependence is inconsistent with our experimental observation. Biexcitonic excitation is another possibility[16] since the Rabi energy $\hbar\Omega$ in our experiments could be close to the biexciton binding energy (typically a few meV). However, experiments performed using circularly polarized light to suppress biexciton excitation showed almost identical intensity dependent behavior, thus ruling out this possibility. Interdot localized-exciton interactions (dipole-dipole) were also considered. However, theoretical calculations[13] showed that the interaction energy is only a few μeV at a typical interdot distance, too small to give rise to significant damping.

Below we propose that the observed RO damping in our system is due to indirect excitation of carriers in the WL. Recent work[17] has clarified the origin of the continuous absorption background related to the WL and attributed the broad resonances seen in single dot PL excitation (PLE) spectra[9,18] to bound-to-continuum and continuum-to-bound transitions. Such indirect excitation channels involving a hole (electron) in the WL and an electron (hole) in other QDs can exist, despite weak transition dipole moments (the wave functions of the electronic states in the QDs decay rapidly into the WL) [Fig. 3(a)]. Since ROs are excited under very strong excitation, these low probability channels can be excited. Once the carriers in the WL are excited, they provide a dephasing channel for the excitonic states in the QD that exhibit ROs. The linear dependence on the intensity for RO damping (one carrier type is sufficient to cause dephasing)

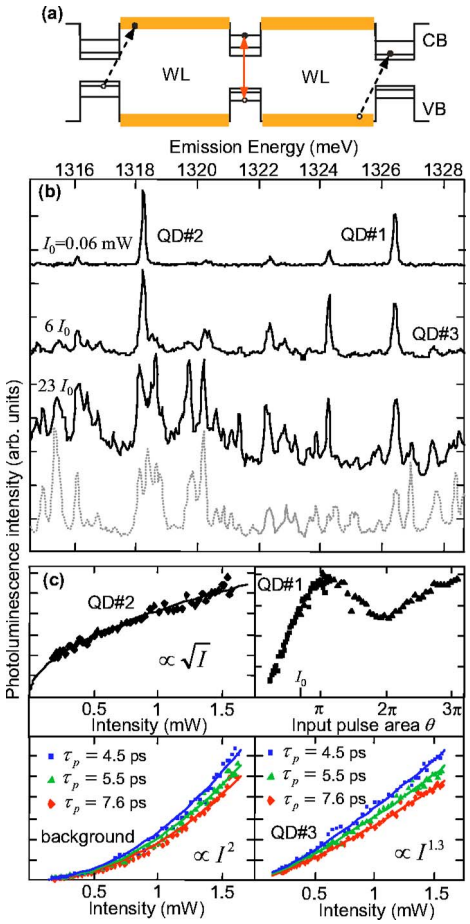

FIG. 3. (Color online) PL spectra of QDs under a 1 micron aperture and their power dependence. (a) Band diagram along a direction perpendicular to the growth direction. The dark bands between dots represent a continuum of states resulting from the WL. The dashed arrows indicate the transitions that can occur at high intensity and are likely responsible for the superlinear dependence of the background signal. (b) Resonant and nonresonant PL spectra. The dots were excited resonantly at ~1340 meV at intensities I_0 =0.06 mW, $6I_0$ and $23I_0$, top to bottom, respectively (solid lines) and above band at ~1650 meV (dashed line). (c) Intensity dependence of peaks denoted by QD#1, QD#2, QD#3 in (a) and the background signal. The PL of peak QD#1 is plotted versus θ. For peak QD#3 and the background signal, for which the PL grows superlinearly with intensity, the experiment was repeated for three laser pulse widths τ_p=4.5 ps (squares), τ_p=5.5 ps (triangles), and τ_p=7.6 ps (diamonds). The smaller the pulse width (the larger the spectral width), the stronger the PL intensity.

and their behavior with the pulse width, i.e., spectral width, is consistent with coupling to a continuum of states. Note that due to composition fluctuations in the wetting layer,[19] there exists a smooth transition between highly localized states (quantum dots) and delocalized quasicontinuum states. The states involved in the present mechanism play the role of a continuum in the sense that they possess broad absorption resonances.[9,18]

To verify that such processes indeed occur, we probed QDs within a submicron shadow mask under varying excitation conditions. In this case, at most about ~150 QDs can be

excited so that resonant and above-band PL spectra can be conveniently compared. Figure 3(b) shows part of the PL spectrum under resonant excitation ($\sim$1.33 eV) for three increasing intensities (solid lines) I_0=0.06 mW, $6I_0$, and $23I_0$, from top to bottom, respectively. At low power, only QDs with their excited states in resonance with the laser emit. Most other QDs have energy states far from resonance and cannot be excited. Similarly, direct excitation of excitons in the WL is not possible because the absorption edge is far above the laser frequency. However, when the laser intensity is increased, transitions involving the excitation of one electron in the WL and one hole in the off-resonance QD, or vice versa, can occur which allows to populate the off-resonance QDs and the WL, as is evident by the fact that at higher intensity, more off-resonance QDs emit until the resonant spectrum coincides with the spectrum excited above the band edge [dashed curve in Fig. 3(b)]. In fact, as is shown in Fig. 3(c), only the peaks present at low intensity are truly excited coherently. For instance, the QD state labelled by QD#1 undergoes RO while the PL from QD labelled QD#2 increases with the square root of the intensity. In contrast, the PL from another QD (labelled QD#3) and the background "wetting layer" emission increases superlinearly with intensity. More interestingly, such a PL displays a very similar pulse width, i.e., spectral width dependence as the RO damping rate [Fig. 1(d)]. This is consistent with multievent processes (I^n intensity dependence) involving a continuum as described in Ref. 17. The carriers thereby created in the WL interact with the exciton undergoing RO leading to intensity dependent dephasing.

In summary, we have investigated the decoherence processes occurring during strong field manipulation of an excitonic qubit in a SAQD. For the first time, observation of rotations of up to 10π pulse area makes it possible to quantify the RO damping, including its variation with the pulse width. Together with the direct dephasing time measurement in the nonlinear regime, we have shown that this damping, which persists even after the end of the pulse, is related to a coupling to broad resonances and is consistent with the mechanism proposed by Vasanelli *et al.* This mechanism involves bound-to-continuum and continuum-to-bound transitions that cause carriers to populate the wetting layer even though the laser is near resonance with interband QD transitions. Therefore, if one can prevent the WL formation, then the major source of decoherence will also be suppressed, thus raising the quality factor of qubits to a practical regime. This might be achieved in the future by employing chemically synthesized QDs that can be grown essentially defect free with near unity quantum efficiency and do not involve a wetting layer.[20] Unfortunately, coherent properties such as quantum interference or Rabi oscillations have not yet been demonstrated in these systems, most likely due to the unwanted blinking effects related to surface passivation. Once these problems have been overcome, these QDs have strong potential to replace MBE-grown QDs as excitonic qubits.

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