Rabi Oscillations of Excitons in Single Quantum Dots

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Transient nonlinear optical spectroscopy, performed on excitons confined to single GaAs quantum dots, shows oscillations that are analogous to Rabi oscillations in two-level atomic systems. This demonstration corresponds to a one-qubit rotation in a single quantum dot which is important for proposals using quantum dot excitons for quantum computing. The dipole moment inferred from the data is consistent with that directly obtained from linear absorption studies. The measurement extends the artificial atom model of quantum dot excitonic transitions into the strong-field limit, and makes possible full coherent optical control of the quantum state of single excitons using optical π pulses.

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Semiconductor quantum dots (QD's) are solid state structures on the 10 nm (nanometer) scale that have been shown to have properties similar to those of individual atoms at the 0.1 nm scale. Numerous demonstrations of the atomlike properties of single QD's have recently included entanglement [1], excited state spectroscopy [2,3], and photon antibunching [4]. The identification of quantum dots as "artificial atoms" is compelling based on such studies, but there remain additional signatures of atom-light interactions that require further experimental study. These signatures would make this analogy more complete and differentiate these systems from higher-dimensional semiconductor structures characterized by many-body physics. Such studies are necessary for progress in both nanoscale semiconductor physics and device development.

Of interest to this work are the quantum mechanical features of discrete level systems that are observed in the limit of strong optical excitation. One such well-known phenomenon is Rabi oscillations, which are observed in resonantly driven two-level systems. They are a direct result of the nonlinearities inherent in a two-level system and have no classical analog [5]. Rabi oscillations are the sinusoidal time evolution of the population difference in a two-level system that occurs at the Rabi frequency (the electric dipole interaction matrix element) for time scales short compared to the dephasing time. They can also be described as a sinusoidal dependence of the population difference on pulse area (the time-integrated Rabi frequency). In the frequency domain, these oscillations translate into the well-known three-peak fluorescence spectrum [6]. In higher-dimensional and impurity-doped semiconductors, Rabi oscillations have been the focus of recent theoretical [7] and experimental [8–11] studies.

In this Letter we report the first observation of Rabi oscillations from excitons confined to single QD's. The results are remarkable in that they demonstrate the coherent manipulation of the QD state and a one-qubit rotation in QD-based quantum logic gates [12,13]. The oscillations are observed at relatively low intensity due to the large oscillator strength (compared to atomic systems) of QD excitons, as independently determined by our group and discussed elsewhere [14]. While many-body effects in a quantum well fundamentally change many aspects of Rabi oscillations [10], particularly the lack of saturation of the excitation density, exciton-exciton interaction effects in a single dot actually sharpen frequency selection for one-exciton transitions and make the Rabi oscillation more atomiclike even though the dot is much larger than an atom.

The experiments are performed on QD's formed naturally in a thin (4 nm) GaAs layer grown between two 25 nm $Al_{0.3}Ga_{0.7}As$ layers. Two-minute growth interruptions at each $GaAs/Al_{0.3}Ga_{0.7}As$ interface lead to the formation of monolayer-high islands which tend to localize the heavy-hole excitons in the GaAs layer [2,15,16]. The islands are elongated in the $[\bar{1}10]$ direction, leading to linear polarization selection rules for the excitonic transitions, as indicated in Fig. 1(a). Excitons in isolated QD's are probed in transmission through ~ 0.5 μm apertures at 6 K in structures similar to those used in [1,17].

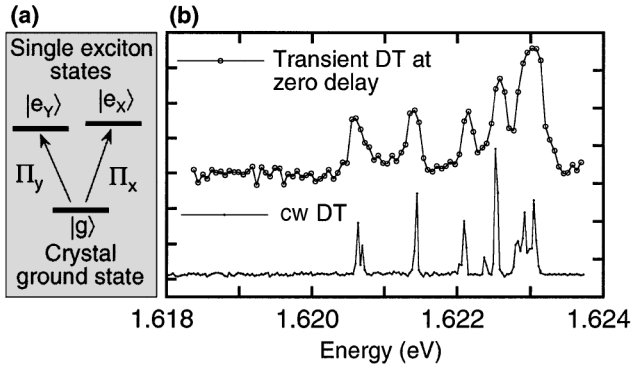

FIG. 1. (a) The energy level structure in the excitonic picture. The polarization selection rules for the fine structure doublet are orthogonal linear for these samples, denoted by Π_x and Π_y. (b) DT spectrum of single QD excitons obtained through a ~ 0.5 μm aperture using pulsed (top) and cw (bottom) lasers. The pump and probe are both Π_y polarized for the data in (b).

The transient nonlinear spectroscopy techniques used are an extension of high-resolution cw nonlinear spectroscopy recently demonstrated on single QD states [17]. A 76 MHz tunable mode-locked dye laser is used to both pump and probe single QD excitonic transitions with a time resolution determined by the pulse width, T, about 6 ps for these experiments. This pulse width represents a compromise between two competing constraints: too much bandwidth resulting from a shorter pulse will cause excitation of multiple QD states, whereas a longer pulse will diminish the temporal resolution necessary to see decaying dynamic effects, such as Rabi oscillations. That is, we require $\hbar/(\Delta\varepsilon) < T < 1/\gamma$, where $\Delta\varepsilon$ is the typical energy spacing between different QD states ($\sim$1 meV for 0.5 μm apertures) and γ is the dephasing rate for QD excitonic transitions, previously measured to be $\sim 1/30$ ps^{-1} in these samples [2,17].

The pump pulse $\mathbf{E}_1(t)$ is used to excite a single QD exciton, $\rho_{ee}(t)$, which then decays back to the crystal ground state, ρ_{gg}, at the relaxation rate, Γ. A weak probe pulse $\mathbf{E}_2(t)$ is delayed with respect to the pump by a time τ and, upon absorption by the excitonic resonance, creates an induced nonlinear optical polarization field, $\mathbf{P}^{(\mathrm{NL})}(t) = 1/2[P^{(\mathrm{NL})}(t)e^{-i\Omega t} + \text{c.c.}]\hat{\boldsymbol{\epsilon}}_P$, where

$$P^{(\mathrm{NL})}(t) \propto \int_{-\infty}^{t} E_2(t')[\rho_{gg}(t') - \rho_{ee}(t')]e^{(i\delta-\gamma)(t-t')}\,dt'. \tag{1}$$

$\mathbf{E}_j(t) = 1/2[E_j(t)e^{-i\Omega t} + \text{c.c.}]\hat{\boldsymbol{\epsilon}}_i$, ω is the excitonic transition frequency, and $\delta = \Omega - \omega$ is the laser detuning. Time-integrated homodyne detection of the polarization field with the probe field represents a differential transmission (DT) signal that is proportional to the level of excitation induced by the pump. Amplitude modulation at $\sim$1 MHz of both the pump and probe allows for the use of phase-sensitive lock-in detection at the difference of the modulation frequencies.

Figure 1(b) shows a transient DT spectrum obtained at low pump powers at zero pump-probe delay ($\tau = 0$). Also shown for comparison is the high-resolution cw DT spectrum, showing that the resonances in transient DT correspond to excitons localized to single QD's. Both spectra were obtained using linearly copolarized Π_y fields. The broadening of the resonances in transient DT is consistent with the bandwidth of the 6 ps pulse ($\sim$350 μeV), whereas the width of the resonances in the cw DT is given by the homogeneous linewidth, γ.

The strong-field response of individual QD excitons can be investigated by monitoring the DT as a function of both the probe delay (τ) and the pump field strength (E_1). A strong pump field can contribute to excess detector noise as well as mix with the probe at the detector, leading to an undesirable background signal. To avoid these complications, the Π_x transition of the exciton fine structure doublet is probed while the Π_y transition (see Fig. 1) is pumped. The fine structure splitting is typically $\sim$10 μeV, easily covered by the bandwidth of the picosecond laser. A polarizer is then used to block the strong pump before the detector. As shown in [1], coupling between the fine structure states $|e_x\rangle$ and $|e_y\rangle$ through the common crystal ground state $|g\rangle$ can be understood in terms of the well-studied atomic three-level system [18]. This is because the binding energy of the biexciton state keeps the exciton to biexciton transition well detuned from the resonant ground state to exciton transition. The excitonic energy levels are shown in Fig. 1(a), and imply that the DT obtained in this way is proportional to the probability that a $|e_y\rangle$ exciton has been created.

Figure 2(a) shows the DT as a function of probe delay, for various pump powers, obtained by tuning the center wavelength of the laser to a single excitonic QD resonance. Each case may be separated into a part driven by the pulse, which is consistent with Rabi rotation in a two-level system, and a part after the pulse duration, which is due to relaxation. The decay time at the lowest power is a measure of $1/\Gamma$ for the $|e_y\rangle$ exciton state. The level of excitation induced by the pump is given by $\sin^2[\Theta(t)/2]$, where $\Theta(t) = (\boldsymbol{\mu}_{eg} \cdot \hat{\boldsymbol{\epsilon}}_1)/\hbar \int_{-\infty}^{t} E_1(t')\,dt'$ and $\boldsymbol{\mu}_{eg}$ is the electric dipole moment for the excitonic transition. Thus, the differential probe absorption (transmission) is minimal (maximal) for $\Theta(t) = \pi, 3\pi, 5\pi, \ldots$. For a pump pulse with a power that corresponds to $\Theta(\infty) = \pi$, the DT signal increases until the pump pulse is completely gone, and it then decays at the relaxation rate. For powers that correspond to $\pi < \Theta(\infty) < 2\pi$, the DT signal will increase, then decrease as the exciton is stimulated back to the crystal ground state. The remaining probability of finding an

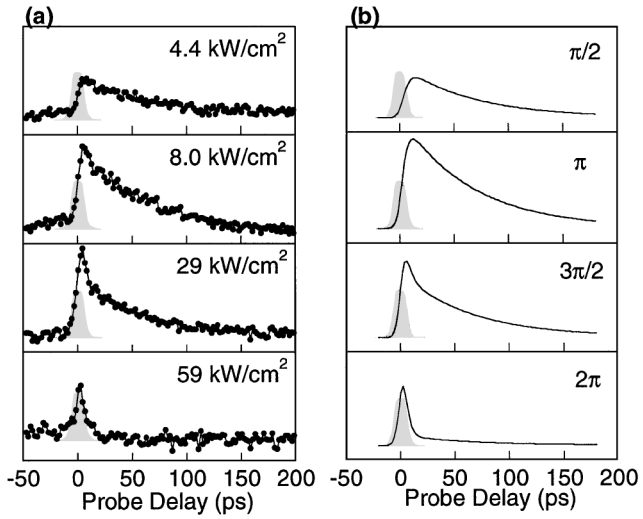

FIG. 2. DT vs probe delay for various pump powers, obtained from a single excitonic QD state. The experimental data are shown in (a), with the peak intensity for the pump pulse shown in the upper right of each plot. The calculated DT is shown in (b), with the pump power expressed as a total pulse area in the upper right of each plot. The pulse autocorrelation is shown for reference with each plot. The vertical axis is the same for all the plots in (a) and for all the plots in (b).

exciton then decays to the ground state at the usual relaxation rate. At $\Theta(\infty) = 2\pi$, $\sin^2[\Theta(\infty)/2] = 0$, so the exciton that was created with unity probability by the first half of the pump pulse is then stimulated (or driven) back to the crystal ground state by the second half of the pulse.

The calculated DT as a function of probe delay illustrates this behavior [Fig. 2(b)]. Here, the pump power is indicated in each plot by the total pulse area, $\Theta(\infty)$. The calculation is based on a numerical solution for $[\rho_{gg}(t) - \rho_{ee}(t)]$ from the density matrix equations with a hyperbolic secant pump pulse (consistent with the measured pulse autocorrelation and spectrum) used as the source term. This population difference is then substituted into Eq. (1), and the time integral of the product of the nonlinear polarization and the probe pulse is evaluated to account for homodyne detection. The pulse width and relaxation rate for the calculation were taken from the data. The agreement between the measured DT and calculated DT further establishes that the data is consistent with the onset of Rabi oscillations.

The Rabi oscillations can be seen more explicitly by examining the DT as a function of E_1. The DT for fixed probe delays such that $T < \tau < 1/\Gamma$ will show oscillations as the pump field strength is increased, since for probe delays longer than the pulse width the level of excitation is proportional to $\sin^2[\Theta(\infty)/2]$. Figure 3(a) shows such data obtained from a single excitonic QD resonance. The DT is measured at two different delays, approximately 10.5 ps and 18.5 ps. Both data sets show an oscillatory behavior, with the first peak corresponding to a pump pulse with $\Theta(\infty) \approx \pi$, the first trough corresponding to $\Theta(\infty) \approx 2\pi$, and so on. The oscillations imply that the excitation of the QD resonance can be controlled by the strength of the pump pulse and are consistent with the DT shown in Fig. 2.

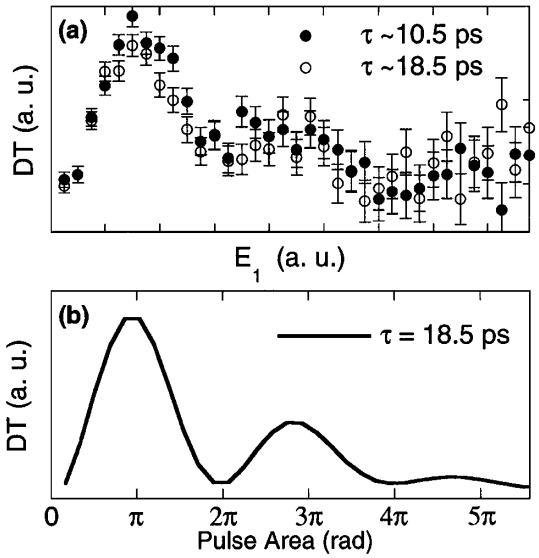

FIG. 3. (a) Measured DT vs pump field amplitude for $\tau = 10.5$ ps and $\tau = 18.5$ ps. Both data sets show behavior consistent with Rabi oscillations for the excitonic QD state under investigation. (b) Calculated DT for an 18.5 ps probe delay, including a continuum of delocalized excitons in the dynamics.

The oscillation period in Fig. 3(a) is proportional to $|\mu_{eg}|$, the dipole moment of the $|g\rangle \rightarrow |e_y\rangle$ transition. Measuring this dipole moment from the data is therefore predicated upon an accurate knowledge of the pump field shape and amplitude inside the sample. An approximate calculation using a refractive index of 3.6 and a hyperbolic secant pulse shape yields a dipole moment of about 75 Debye. This value compares well with predictions [19,20], based on dots in this size range [2], and recent measurements [14] of transition strengths of single QD excitons. The extremely large dipole moment associated with these transitions (ground state atomic dipoles are on the order of a few Debye) allows for the use of lower pulse energies to observe strong-field effects.

The data show the oscillatory behavior expected for a two-level system, but this simple model alone does not predict the observed decay of the oscillations with increasing pulse area. Separate ensemble measurements show an unexpected increase in the exciton relaxation rate with pump intensity (data not shown). Such an effect could be due to a local heating effect, but seems unlikely due to the low average powers used for these measurements. More likely, the increased decay rate arises from exciton-exciton interactions that occur between the resonantly excited QD exciton and weakly localized excitons excited off-resonance. Evidence for the simultaneous existence of both strongly and weakly localized excitonic states has been seen in photoluminescence (PL) imaging studies of interface fluctuation QD's. These studies report broad resonance features (degenerate with sharp PL lines) which emerge with an increase in power, suggesting the presence of delocalized (or weakly localized) excitons [21]. Reports of biexponential photon echo decays have also been attributed to a class of delocalized excitons nearly degenerate with localized excitons [22].

To model this observed increase in scattering, an equation of motion for an incoherent population of delocalized states has been added to the Lindblad equation for the density matrix of the three states in Fig. 1(a). The localized exciton can be scattered by the delocalized excitons, which are characterized by a density that increases with laser intensity. This results in an intensity dependent relaxation term. Such delocalized states could also contribute to a shift in the exciton resonance frequency. However, the density dependent scattering was found to be more effective in causing the damping in the DT, so the effects of the shift were neglected. Figure 3(b) shows the calculated DT, which reproduces the qualitative features of the data in Fig. 3(a). The properties of delocalized excitons concerning Coulomb scattering and the optical absorption coefficient were assumed to be close to the two-dimensional case. The absorption coefficient for the delocalized excitons is $\alpha = 0.1\%$ and the scattering rate is $\Gamma(t) = 10 + \beta n_{\mathrm{del}}(t)\ \mu\mathrm{eV}$, where $\beta = 0.75 \times 10^{-7}\ \mu\mathrm{eV\,cm^2}$ [23]

and n_{del} is the density of delocalized excitons. Even if the total population of delocalized excitons is small, the DT can be sensitive to a change in the scattering rate of a few tens of μeV for the localized exciton. For the pump powers shown in Fig. 2(a), this effect does not result in a significant change in the relaxation rate. (Γ has increased by less than 20% for pump powers corresponding to a π pulse.) It is important to note that, according to the model, the origin of the decay of the Rabi oscillations is due to an increase in the decay rate of the exciton state (i.e., the diagonal density matrix elements). The increased decay also contributes to an increase in the dephasing rate (decay of the off-diagonal density matrix elements), but this increase (which can be suppressed in the model) does not by itself lead to the observed decay.

The above analysis supports the conclusion that the excitation induced Coulomb scattering is the origin of the decay of the Rabi oscillations. However, in light of the recent report of the effects of nearby two-exciton (e.g., biexciton) states on Rabi oscillations [13], the density matrix was extended to include the two-exciton state, with either positive or negative binding energy. Including this state, while ignoring the effects of delocalized states, shows a change of the apparent Rabi period, but no corresponding decay. Such a result is not surprising, since an increase in the exciton relaxation rate must be the result of an interaction with a continuum of states.

In summary, the observed Rabi oscillations show that ultrafast π pulses can be used to coherently manipulate the QD excitations at the single dot level, creating new opportunities for experiments in semiconductor nanostructures. For example, due to both their strong-field behavior and their large dipole moments, these types of QD's could serve as the electronic transition coupled to a mode of a semiconductor microcavity, an inherently strong-field interaction [19,24]. Also, the use of these QD's as qubits in an all-optical semiconductor quantum logic gate [12,13] is now possible, whereby π and $\pi/2$ pulses are used for ultrafast coherent single-qubit rotations. The decay of the oscillation is not fundamental, but is likely due to the presence of nearby states that are not well confined; quantum dots with stronger confinement are expected to be less susceptible to these effects.

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