Robust Quantum Dot Exciton Generation via Adiabatic Passage with Frequency-Swept Optical Pulses

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The energy states in semiconductor quantum dots are discrete as in atoms, and quantum states can be coherently controlled with resonant laser pulses. Long coherence times allow the observation of Rabi flopping of a single dipole transition in a solid state device, for which occupancy of the upper state depends sensitively on the dipole moment and the excitation laser power. We report on the robust population inversion in a single quantum dot using an optical technique that exploits rapid adiabatic passage from the ground to an excited state through excitation with laser pulses whose frequency is swept through the resonance. This observation in photoluminescence experiments is made possible by introducing a novel optical detection scheme for the resonant electron hole pair (exciton) generation.

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Photon correlation measurements along with resonant laser scattering have established the atomlike character of the interband transitions in quantum dots [1–3]. Excitation of a two level system by a short, intense laser pulse can induce an oscillation of the system between the upper and lower state during the pulse, at the Rabi frequency $\Omega(t) = \mu A(t)/\hbar$ where μ is the dipole moment of the transition and $A(t)$ the electric field envelope of the laser pulse. Any quantum state (qubit) manipulation scheme benefits from the long coherence times in quantum dots [4,5] and necessitates fast and robust initial state preparation [6–8]. In principle a two level system can be initialized in the upper state with a maximum fidelity of 100% if the laser power is optimized in order to induce exactly half a Rabi oscillation during the pulse duration, a so-called π pulse [9–11]. Although Rabi oscillations observed in a single dot or for individual atoms [12] are a beautiful example of strong coupling between laser light and a single dipole, this commonly used technique presents two major drawbacks: (i) the upper state population is highly sensitive to fluctuations in the system, such as laser power, and (ii) in measurements on dot ensembles, an inhomogeneous distribution of dipole moments and transition energies among dots requires different laser intensities and frequencies for inducing a full population inversion in the dot ensemble.

Here we show that these drawbacks can be overcome by inducing an adiabatic passage (AP) with frequency-swept laser pulses. Unlike Rabi cycling, AP is robust against small-to-moderate variations in the laser intensity, detuning, dipole moment, and interaction time [13]. Chirped radiofrequency (rf) pulses are commonly used in nuclear magnetic resonance (NMR) based imaging [14]. In the context of semiconductor quantum dots adiabatic population transfer has been induced by a slow variation of the electrostatically defined confinement potential [15]. For applications in atomic physics and chemistry for quantum states separated by frequencies in the optical domain, chirped laser pulses, in strong analogy to NMR, have been used to induce complete population transfer via AP [13,16]. The intriguing combination of excitation with a linearly chirped and a transform limited pulse has also been proposed for control of both population and coherence in multilevel systems [17]. Our experiments demonstrate the power of adiabatic population transfer between individual quantum states in condensed matter, paving the way for stimulated Raman AP (STIRAP) [13,18] in single dots, efficient spin state preparation [19,20] and quantum condensation of excitons in a microcavity [21].

The two level system investigated here is composed of a ground state and neutral exciton X^0 state of an individual InAs quantum dot in a GaAs matrix embedded in a Schottky diode structure with a 25 nm tunnel barrier as in [22]. The experiments are carried out at 4 K in a confocal microscope connected to a spectrometer and a Si charge coupled device camera. Application of a gate voltage enables deterministic loading of individual electrons. To minimize the laser stray light on the detector we employ dark field techniques and linear cross polarization.

Pioneering work on Rabi oscillations in single semiconductor quantum dots used differential transmission [6,7] and photocurrent techniques [10,11] for detection following resonant optical excitation. To probe resonant exciton generation in emission poses the problem of laser stray light at the detection wavelength reaching the detector. Implementation of up-conversion techniques [23], waveguide sample structures [3], or nonresonant emitter–cavity coupling [24] have allowed this problem to be circumvented. Using charge tunable structures allowed us to implement a novel detection scheme that does not suffer from a strong background signal as photocurrent. The generated X^0 population is probed by monitoring the negatively charged exciton X^- emission, which plays the role of a spectator state at lower energy; see energy level diagram in Fig. 1(d). In a small window of voltage, at a time τ_{in} (which we estimate to be ≈ 70 ps [25]) the photogenerated X^0 becomes an X^- by grabbing an electron from the Fermi sea in the sample back contact. Radiative emission of the X^- leaves behind a single electron which tunnels out of the dot, allowing again X^0 absorption [26]. Photocurrent experiments [10,11] are carried out at the low bias end of the X^0 charging plateau, our optical detection scheme can in principle be implemented in the exact same structures, carefully scanning the high bias end of the X^0 charging plateau.

In a first step, the population transfer achieved via Rabi cycling between the ground state $|0\rangle$ and the exciton state $|X^0\rangle$ will serve as a reference for the robust exciton generation via an AP for the same quantum dot. The lifetime T_1 and the maximum coherence time $T_2 = 2T_1$ of the optically created X^0 are in our case limited by the tunneling time τ_{in}. To observe Rabi oscillations, τ_{in} should be longer than the pulse duration $\tau_0 \simeq 3$ ps (FWHM) of spectral width $\simeq 0.5$ meV from the pulsed Ti-Sa laser. The photogenerated X^0 population undergoes clear sinusoidal oscillations when plotted versus the pulse area $\Theta = \int_{-\infty}^{t} \Omega(t')dt'$ where $t \gg \tau_0$ [6], see Fig. 2(a). This time integrated Rabi frequency is proportional to the square root

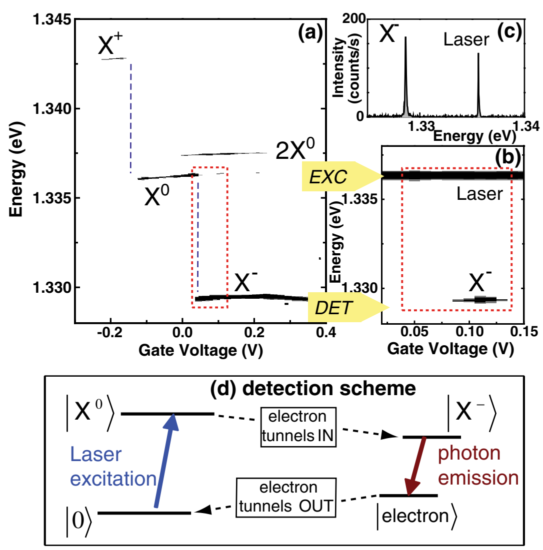

FIG. 1 (color online). (a) Contour plot of the PL intensity at 4 K as a function of gate voltage applied to the sample for nonresonant excitation in the wetting layer; white <50 counts, black ≥ 1000 counts. The positively charged exciton X^+, and the neutral exciton X^0, biexciton $2X^0$ and the negatively charged exciton X^- were identified through gate voltage dependence and fine structure analysis. (b) Contour plot PL as a function of voltage with a narrowband cw laser resonant with the X^0 transition. The X^- transition appears for a gate voltage range of about 50 mV. (c) Spectrum when the laser is resonant with the X^0 transition leading to X^- emission. (d) Energy level diagram with X^0 absorption monitored via X^- emission.

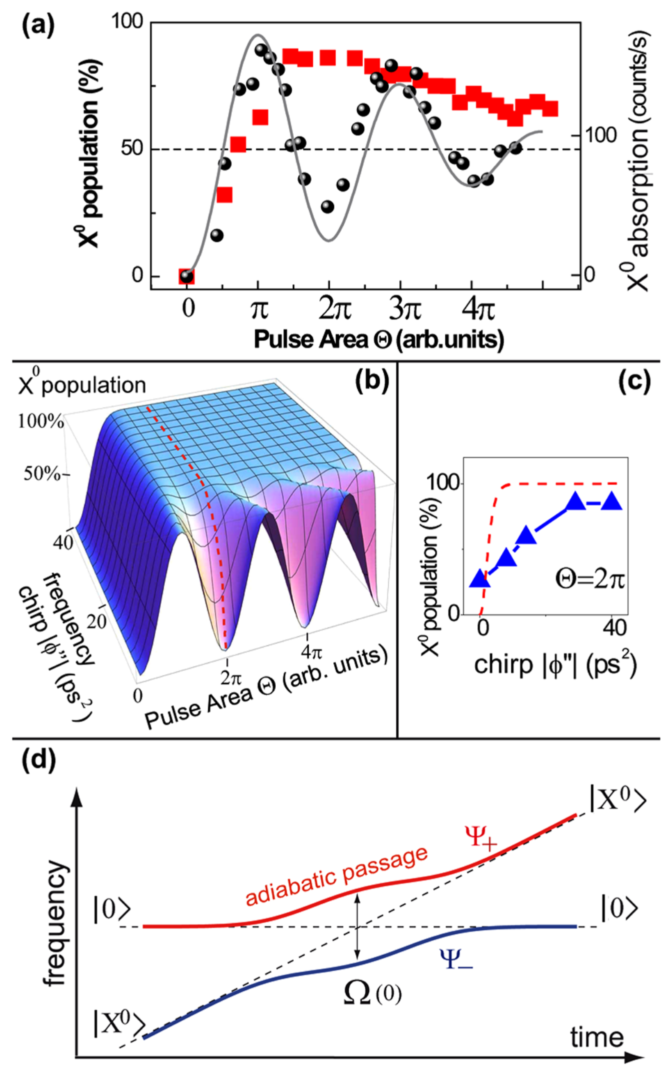

FIG. 2 (color online). (a) Rabi oscillations (black circles data point, gray line is a fit with model adapted from [11]) for excitation with Fourier transform (FT) limited pulse and AP for strongly chirped laser with $\phi'' = -40$ ps^2 (red squares). (b) Numerical calculation [see Eq. (4)] of the X^0 population as a function of laser pulse area Θ for different chirps up to $|\phi''| = 40$ ps^2, the maximum value used in the experiment. The presented graph is symmetrical with respect to a change in sign of ϕ''. Dashed red line shows X^0 population for constant $\Theta = 2\pi$. (c) Blue triangles: experimentally achieved X^0 population as a function of frequency chirp for a $\Theta = 2\pi$ pulse, dashed red line [same as in (b)] shows calculated X^0 population for constant $\Theta = 2\pi$. (d) Eigenfrequencies of the dressed states $|\Psi_+(t)\rangle$ and $|\Psi_-(t)\rangle$ given by $1/2[-\Delta(t) \pm \sqrt{\Delta(t)^2 + \Omega(t)^2}]$ [see Hamiltonian in Eq. (3)] calculated as a function of time for $\Theta = 5\pi$ and $\phi'' = -40$ ps^2.

of the average laser intensity. The strictly resonant excitation in our experiment would lead without damping to oscillations of the X^0 population between 0 and 100% [11]. A first maximum for the X^0 population of about 90% is observed for a pulse area equivalent to the rotation angle of the system of $\Theta = \pi$; see Fig. 2(a). The contrast of the Rabi oscillations does not change significantly over the bias voltage range (0.07 to 0.12 V) which results in X^- emission upon X^0 excitation. As $\tau_{\rm in}$ does change when the tunnel barrier height is changed, this observation provides another indication that $\tau_{\rm in} \gg \tau_0$. Symmetric damping of the Rabi oscillations as a function of pulse area indicates an excitation power dependent dephasing process, as, for example, the interaction with acoustic phonons [11]. The gray line in Fig. 2(a) is based on the model in [11], i.e., allowing for transitions between the two dressed states (see below), taking into account that in our experiments the thermal energy is comparable to or smaller than the Rabi splitting $\hbar\Omega$ (here in the order of 1 meV). The comparison between these calculations and the data serve to express the X^0 population in percent.

Using the same quantum dot and detection techniques presented in Fig. 1, we present in what follows a successful implementation of AP to prepare robustly a single quantum dot in an exciton state X^0. The key change lies in the excitation pulse: the system is excited resonantly by an initially Fourier transform limited laser pulse that passed a compressor composed of two reflection gratings with 2000 lines/mm [27] before reaching the sample. As a result, the frequency in the excitation pulse sweeps now linearly in time [16], with an instantaneous pulse frequency $\omega_i(t) = \omega_0 + 2bt$, where b is the sweep rate. The frequency sweep should be slow enough such that the system is at any given time during the interaction in an eigenstate [16]. Results for the X^0 absorption as a function of pulse area are plotted on Fig. 2(a). Excitation with a resonant Fourier transform limited pulse results in Rabi oscillations, whereas excitation with a chirped pulse with the same central frequency is strikingly different, showing no oscillations of the prepared X^0 population. For $\Theta = \pi$, the chirped pulse does not yet create an efficient population transfer $|0\rangle \rightarrow |X^0\rangle$. This is achieved for a pulse area $\geq 1.5\pi$, beyond which the created X^0 population changes very little with increasing power [28]. This is a clear signature of a robust population inversion via an adiabatic process. AP is a coherent process. The disappearance of the Rabi oscillations due to decoherence would result in a pulse area (laser power) dependence in Fig. 2(a) that would slowly approach from below, but never exceed a 50% X^0 population probability. The AP data in Fig. 1(a) shows for $\Theta \simeq 1.5\pi$ an X^0 population of 90%, i.e., as the highest value observed for Rabi oscillations. The X^0 population decreases for $\Theta \geq 1.5\pi$ with increasing laser power. It is reasonable to assume that acoustic phonon emission is possible to go from one eigenstate to the other, as in the case of Rabi oscillations [11]. This could explain why the generated X^0 population seems to decrease from the initial 90% when using higher laser power, and model calculations to clarify this effect are under way.

To further demonstrate the influence of the laser chirp on the X^0 population, we plot in Fig. 2(c) the measured X^0 population created for a 2π pulse as a function of the applied chirp. The lowest value of only about 25% for the damped Rabi oscillations rises gradually to about 90% for an AP with maximum chirp, with intermediate values for smaller chirps.

Figure 2(b) shows a calculation of the amount of chirp necessary to pass from the Rabi oscillation regime to robust AP in the absence of damping. The electric dipole interaction of the laser pulse with the two level system $|0\rangle$ and $|X^0\rangle$ separated in energy by $\hbar\omega_0$ is

$$W(t) = -DE(t) = -\mu E(t)(|0\rangle\langle X^0| + |X^0\rangle\langle 0|). \qquad (1)$$

The real electric field of the excitation pulse $E(t)$ can be written as $E(t) = \frac{1}{2}[A(t)e^{-i\omega_0 t} + A^*(t)e^{i\omega_0 t}]$, where ω_0 is the laser frequency in the case of our resonant experiment. In what follows we assume a Gaussian pulse shape [29]:

$$A(t) = \frac{\Omega(t)}{2\mu} = \frac{\Theta}{2\sqrt{\pi}\mu\sqrt{\Gamma}} \exp(-t^2/\Gamma). \qquad (2)$$

τ_0 corresponds to the Fourier limited FWHM pulse duration in intensity while ϕ'' is the second derivative of the spectral phase of the electric field at the laser frequency and $\Gamma = \frac{\tau_0^2}{2\ln 2} - 2i\phi''$. Because of the chirp ϕ'' the excitation frequency sweeps linearly in time during the pulse with a rate $b = \frac{8\phi''(\ln 2)^2}{\tau_0^4 + 16(\phi'')^2(\ln 2)^2}$. This leads for the system-laser interaction to a time-dependent detuning $\Delta(t) = 2bt$ assuming the laser is resonant at $t = 0$. In our setup the double grating compressor introduces a negative chirp ϕ'' proportional to the distance d between the two gratings in the compressor [16]. The AP is carried out with d large enough to produce a chirp as strong as $\phi'' = -40$ ps^2 leading to a stretched pulse of $\tau \simeq 40$ ps, where the chirped pulse width is given by $\tau^2 = \tau_0^2 + (4\phi'' \ln 2/\tau_0)^2$. In our case $\tau < \tau_{\rm in}$, and simulations show that only for τ clearly longer than $\tau_{\rm in}$ AP becomes impossible.

The Hamiltonian in the frequency modulation frame using the rotating wave approximation, applicable for the laser powers used here, may be written as [16]

$$H = \hbar\begin{pmatrix} 0 & \frac{\Omega(t)}{2} \\ \frac{\Omega^*(t)}{2} & -\Delta(t) \end{pmatrix}. \qquad (3)$$

To evaluate the X^0 population as a function of time, that corresponds to the element $\rho_{11}(t)$ of the 2×2 density matrix $\rho(t)$, we solve numerically the Liouville equation

$$i\hbar\frac{d\rho(t)}{dt} = [H, \rho(t)]. \qquad (4)$$

In Fig. 2(b) we plot the solutions of Eq. (4) for a time $t > \tau$ varying the pulse area Θ and the frequency chirp ϕ''. For the picosecond pulse used in our experiment a chirp $|\phi''| > 20$ ps^2 of positive or negative sign, achieves robust population transfer via AP in our nondamped calculation. For example, the calculated X^0 population as a function of ϕ'' for $\Theta = 2\pi$ shows the same tendency as the experiments for different values of $|\phi''|$ in Fig. 2(c). In theory there is no upper limit for a useful $|\phi''|$, in practice an excessively strong chirp (long pulse duration) should be avoided to ensure the system remains coherent during the light-matter interaction [4].

An intuitive interpretation of the observed AP comes from the dressed state picture [2,3,30]. The eigenstates $|\Psi_+(t)\rangle$ and $|\Psi_-(t)\rangle$ and the corresponding eigenenergies of the two level system coupled to an electromagnetic wave are obtained by diagonalizing the Hamiltonian H [Eq. (3)]. During an AP the system evolves slowly enough so it does not change eigenstate population. It is the composition of the eigenstate that changes as the laser frequency sweeps past resonance. In our case the sweep rate $b < 0$, so an AP is possible following the dressed state $|\Psi_+(t)\rangle$ in Fig. 2(d). Asymptotically, for times long before and after the pulse, each dressed state becomes uniquely identified with a single unperturbed state and complete adiabatic inversion $|0\rangle \rightarrow |X^0\rangle$ occurs. Considering the time evolution, critical points occur when the two dressed states are close in energy so diabatic transitions are possible [31] in our case via emission of acoustic phonons to go from the $|\Psi_+(t)\rangle$ to $|\Psi_-(t)\rangle$ as observed in the case of Rabi oscillations [11]. Changing the sign of the chirp (sweep rate) will lead to AP along the lower energy dressed state $|\Psi_-(t)\rangle$. In this case dephasing is only possible due to phonon absorption, which is very unlikely at 4 K. So by changing the sign of the chirp the population inversion achieved could be even closer to 100% than in the present case.

In future experiments it would be intriguing to verify if controlled, resonant exciton generation improves the fidelity of single photon interference experiments [32]. Another interesting extension of the presented measurements is biexciton creation via an adiabatic passage to allow controlled generation of entangled photon pairs [33,34]. The robustness of the adiabatic passage presented here is a starting point for optical manipulation of qubits based on quantum dots coupled through tunable tunnel barriers [35,36] or for a single dot in a transverse magnetic field [8,37]. In both scenarios a Λ system is formed for which STIRAP can be envisaged [13,18].

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Note added.—After completing this work, we became aware of similar results obtained with photocurrent detection [38].

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