Excitation-Induced Dephasing in a Resonantly Driven InAs/GaAs Quantum Dot

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We report on coherent emission of the neutral exciton state in a single semiconductor self-assembled InAs/GaAs quantum dot embedded in a one-dimensional waveguide, under resonant picosecond pulsed excitation. Direct measurements of the radiative lifetime and coherence time are performed as a function of excitation power and temperature. The characteristic damping of Rabi oscillations observed is attributed to an excitation-induced dephasing due to a resonant coupling between the emitter and the acoustic phonon bath of the matrix. Other sources responsible for the decrease of the coherence time have been evidenced, in particular an enhancement of the radiative recombination rate due to the resonant strong coupling between the dot and the one-dimensional optical mode. As a consequence, the emission couples very efficiently into the waveguide mode, leading to an additional relaxation term of the excited-state population.

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In the past decade, important research has been devoted to understanding the mechanisms responsible for dephasing in semiconductor quantum dots (QD). Indeed, coherence is a key issue to address if using QDs as qubits to ensure, for instance, high-fidelity operations in coherent control schemes [1] or obtain a single-photon source with a high degree of indistinguishability [2]. Because of their strong interaction with their environment, the coherence time is not radiatively limited as expected theoretically and pure dephasing processes can occur, depending on the kind of quantum dots and the nature of their host matrix [3]. This can be well understood in the case of incoherent optical pumping of the dot, when using, for instance, nonresonant excitation leading to coupling to continuum states in the wetting layer [4–6] or to multiexcitonic transitions [7]. Strictly resonant pumping of a given optical transition in the dot appears then as the most reliable way to keep the coherence of the state. The first pioneer experiment of resonant pumping with short optical pulses measuring the Rabi oscillations in emission was realized in atomic systems [8]. However, for solid-state emitters, only a few demonstrations have been reported in the literature due to the technical difficulties in performing resonant excitation experiments. Recently, resonant excitation configurations have been achieved, allowing the observation of the resonant luminescence of a single QD [9–13]. Other techniques, such as differential transmission [14], four-wave mixing [15], or photocurrent detection [16,17], also allowed the resonant manipulation of a single QD. Even though, important dephasing occurs observed both in continuous-wave and in pulsed excitation regimes, showing a spectral broadening of the Mollow triplet [18] or a damping of Rabi oscillations [10,15,17], respectively. The damping has been shown to be power dependent [4,10,14,17,19], and the main mechanism for such an excitation-induced dephasing (EID) is the interaction with longitudinal acoustic (LA) phonons that constitute an intrinsic limitation for coherence. Numerous theoretical approaches have been used to investigate the effect of phonon interactions on the coherent manipulation of excitons in QDs, using exact or perturbative methods [20–23], path integral formalism [24], or polaron transform [25,26]. For temperatures below 30 K, which are the usual experimental conditions where single dot spectroscopy is carried out, similar predictions have been found [25] and the weak-coupling regime is enough to describe the physical situation in a first approximation.

In this Letter, we report on resonant luminescence from single QD neutral exciton under picosecond (ps) pulsed excitation. Direct measurements of the radiative lifetime T_1 and coherence time T_2 are reported as a function of temperature and excitation power in order to investigate the different EID mechanisms taking place. The role of phonons in dephasing is definitely dominant, and the observed damping of the Rabi oscillations [17,25] has a quadratic frequency-dependent behavior. This is characteristic of a resonant coupling between the two-level system (TLS) and the phonon bath [21,22]. Moreover, depending on the specific QDs growth conditions and geometry (QDs embedded in micropillars, nanowires, photonic crystal waveguides, etc.), a shaping of the electromagnetic spectral density can give rise to a modification of the spontaneous emission rate [27–30] and influence the coherence properties of the system. In our samples' structure, the dots are coupled to a single-mode 1D waveguide (WG) that modifies the spontaneous emission rate [31] and leads to an additional relaxation of the population. The coupling to the

optical mode is different from one dot to another because of the random distribution of the dots in the InAs layer and, thus, is dot position dependent. Two typical results will be presented: for one kind of dots (hereafter called of "type A") that are not efficiently coupled to the WG mode and for another one (QDs of "type B") where on the contrary the coupling is very efficient.

InAs/GaAs self-assembled QDs were grown by MBE on a planar [001] GaAs substrate and embedded in a two-dimensional GaAlAs transverse single-mode waveguide [32]. Micrometer ridges are etched on the top surface to reduce the volume of the optical mode and enhance the light-matter interaction. In contrast to the case of dots embedded in microcavities, there is no need here to match the cavity mode energy with the QD emission in order to achieve the strong coupling regime. The QDs are excited by ps pulses provided by a tunable mode-locked Ti:sapphire laser and coupled into one ridge. The laser polarization is such as resonant excitation addresses a single eigenstate of the fine structure-split exciton states [33]. More experimental details can be found in Refs. [10,32]. In this geometry, the laser is confined in the guided mode and the single QD luminescence is collected from the ridge top surface by a confocal microphotoluminescence (μPL) detection setup. The scattered light is then greatly suppressed, and at low pump power the resonant luminescence is almost laser background free.

The nonlinear interaction between the TLS and the resonant field gives rise to the well-known Rabi oscillation (RO) of the excited level population. In pulsed excitation, the emission intensity oscillates as a function of the pulse area, which is proportional to the square root of the incident laser power [34]. A typical RO of a QD emitting at 930.3 nm is shown in Fig. 1. Two main features appear on this curve, the rapid damping and the limited number of oscillations that can be recorded. The former behavior is related, as will be discussed in the following, to the excitation-induced nonlinear coupling between the optically driven dipole and the phonon modes. The latter problem is due in fact to the residual scattered laser, which becomes important when the pump power is increased. This is one of the main difficulties when studying the resonant PL as compared to photocurrent measurements where numerous oscillations can be observed [16,17]. Because of the finite radiative lifetime and the limited coherence time of the system, a damping of RO is expected. We measured by time-resolved PL the on-resonance lifetime T_1 for this dot and found it to be 800 ps. The coherence time T_2 was also measured by coherent control experiments [35] and found to be on the order of $T_1/2$. Nevertheless, using the standard optical Bloch equations for a TLS with constant T_1 and T_2 is not enough to accurately simulate the experimentally observed damping, as shown in Fig. 1 by the blue dotted curve. A power-dependent dephasing has to be introduced in the model to explain the rapid damping of RO. For resonantly driven QDs, we expect that the coupling to phonons should be the most relevant dephasing process [20–26]. The RO behavior can then be more accurately adjusted as shown in Fig. 1 by the red solid curve.

Thereafter, we performed μPL experiments with resonant ps pulses by varying the sample temperature from 7 to 30 K. Above 30 K, the resonant emission intensity becomes too weak to be observed. In Fig. 2, we show two RO curves at 10 and 20 K for another dot of type A. As expected, the damping has increased with temperature. To identify whether the interaction with acoustic phonons is the only EID mechanism, we modeled the exciton-LA phonon coupling using a perturbation approach [17,19–21]. The decoherence rate is found to be proportional to the square of the bare Rabi frequency Ω_R, which is characteristic of an EID process. The optical Bloch equations in the rotating wave approximation then take the form [34,36]

$$\dot{\sigma}_{11}(t) = -\frac{i}{2}\Omega_R(t)(\sigma_{10}(t) - \sigma_{01}(t)) - \frac{\sigma_{11}(t)}{T_1},$$
$$\dot{\sigma}_{01}(t) = \frac{i}{2}\Omega_R(t)(\sigma_{11}(t) - \sigma_{00}(t)) - \left(\frac{1}{T_2^0} + \kappa(T, \Omega_R(t))\right)\sigma_{01}(t). \quad (1)$$

The diagonal terms $\sigma_{00}(t)$ and $\sigma_{11}(t)$ stand for the population of the fundamental and excited state, respectively, whereas the off-diagonal terms $\sigma_{01}(t)$ and $\sigma_{10}(t)$ stand for the coherence of the quantum state. $\Omega_R(t)$ is the time-dependent Rabi frequency of the laser pulse assumed to

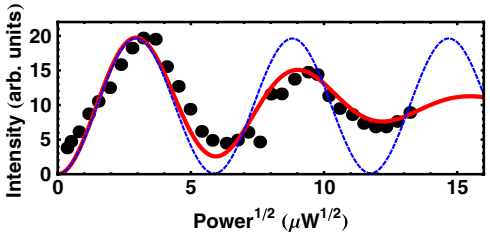

FIG. 1 (color online). Resonant Rabi oscillation of the population in one particular QD of type A at 7 K: the black dots correspond to the emission intensity of the exciton plotted as a function of the square root of the pump power. The blue dotted curve is a fit using the bare optical Bloch equations, and the red solid curve includes coherence relaxation (see text).

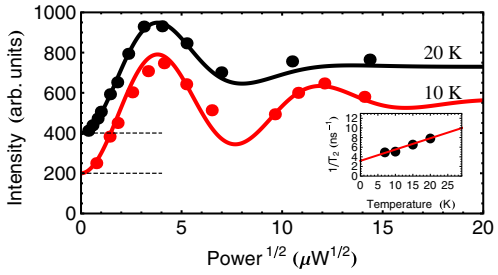

FIG. 2 (color online). Rabi oscillation of a type A QD at 10 and 20 K in red and black curves, respectively. The inset shows the linear dependence of the decoherence rate with temperature for excitation with $\pi/2$ pulses (see text).

be a hyperbolic secant with field amplitude E_L and pulse width τ. The pulse area θ is defined by $\theta = \int_0^\tau \Omega_R(t)dt = \int_0^\tau ((\mu E_L)/\hbar)\mathrm{sech}(t/\tau)$, μ being the dipole moment of the transition. The pulse area is therefore proportional to the square root of the pump power. This proportionality coefficient is the only adjustable parameter for fitting the experimental RO. T_1 is measured experimentally and kept constant. T_2^0 accounts for the radiatively limited coherence time plus any other additional pure dephasing time. $\kappa(T, \Omega_R(t))$ is the damping rate due to phonon coupling, which takes the simple form for temperatures above 10 K of

$$\kappa(T, \Omega_R(t)) \approx KT\Omega_R^2(t) = \frac{(\mathcal{D}_e - \mathcal{D}_h)^2}{4\pi\hbar^2\rho c_s^5} k_B T \Omega_R^2(t).$$

The proportionality coefficient K depends only on the properties of the surrounding GaAs bulk material that can be found in the literature [23]: the effective band-gap deformation potential $(\mathcal{D}_e - \mathcal{D}_h) \approx -8.5$ eV, the material density $\rho \approx 5.4\ \mathrm{g\cdot cm^{-3}}$, the sound velocity $c_S \approx 5110\ \mathrm{m\cdot s^{-1}}$. We can then estimate $K_{\mathrm{theo}} \approx 10\ \mathrm{fs\cdot K^{-1}}$. In fact, at temperatures lower than 10 K, the linear dependence is not valid anymore because $k_B T \leq \hbar\Omega_R/2$ [17,25]. By adjusting numerically the RO curves at different temperatures with Eqs. (1), we find that the damping parameter $\kappa(T)$ has a linear dependence with temperature as expected (not shown here). The characteristic slope is $K_{\mathrm{exp}} \approx 25\ \mathrm{fs\cdot K^{-1}}$ which agrees with the above estimation of K_{theo}. Moreover, the coherent control measurements as a function of temperature allowed us to determine the coherence time T_2 temperature dependence. A linear variation has been found for the decoherence rate: $1/T_2 = 1/T_2^0 + KT\Omega_R^2$, in agreement with the model (see inset in Fig. 2). In order to fit the data we took $K = K_{\mathrm{exp}}$ and left Ω_R as an adjustable parameter found to be consistent with $\pi/2$ pulses. When extrapolating at zero temperature, it appears that the coherence time is not radiatively limited. Indeed, T_1 has been measured to be 600 ps for this dot but at zero temperature $T_2^0 \approx 330\ \mathrm{ps} \neq 2T_1$. Thus, it seems that another source of pure dephasing exists with a rate comparable to the coupling to phonons on the order of $2.2\ \mathrm{ns^{-1}}$. This additional dephasing has been discussed by different groups [37–39] and is likely due to the dot fluctuating electrostatic environment that is a consequence of charge trapping in the vicinity of the dot. This supplementary mechanism has been evidenced only for 20% of the studied dots, showing again the influence of the dot specific environment. In the case of EID due to phonons, the luminescence intensity $\mathcal{L}$ tends to the limiting value $\mathcal{L}_\infty = \mathcal{L}_{\mathrm{max}}/2$ for high pump power corresponding to the stationary occupation of one half [34,36]. This is the case for the dots of type A like that shown in Figs. 1 and 2. In our experiments, neither a renormalization of the Rabi frequency with increasing temperature [17] nor a revival of the RO with higher pulse area was observed, as predicted theoretically by Vagov and co-workers [24]. The reasons for that may be due to experimental limitations because the luminescence

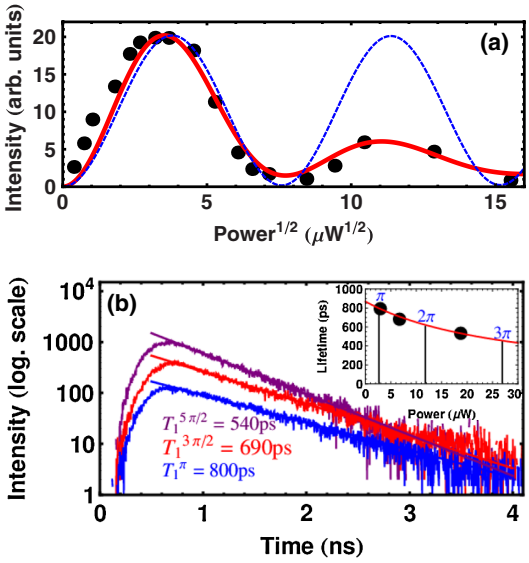

FIG. 3 (color online). (a) Resonant RO of the population in a type B QD. The red solid curve includes excitation-induced dephasing and population relaxation (see text). (b) Time-resolved μPL for different pulse areas. The inset shows the values of the radiative lifetime (black dots) as a function of pump power; the red solid curve represents a numerical fit of this dependence scaling as the inverse of the pump power.

becomes too weak to be detected above 30 K and the resonant laser completely blurs the emission of the dots for large pulse areas.

Another typical trend that we found is that at high pump power $\mathcal{L}_\infty < \mathcal{L}_{\mathrm{max}}/2$, and even $\mathcal{L}_\infty \to 0$ in some cases, such as that shown in Fig. 3(a). These dots are the ones denoted as type B. In this situation, the excitation-induced pure dephasing is not the only mechanism explaining the damping, and an additional power-dependent relaxation term for the population has to be taken into account. This would be equivalent to a term that enhances the emission rate as a function of the pump power. A similar behavior has been reported in previous resonant experiments in thickness fluctuations interface dots [10] embedded also in 1D waveguides. We have corroborated this result by performing resonant time-resolved μPL experiments as a function of the pump power. Indeed, for type B QDs, the radiative lifetime gets shorter as the pump power increases, whereas in the case of type A QDs the radiative lifetime does not vary significantly with power. Figure 3(b) shows the time-resolved μPL of the exciton in the same type B QD excited on resonance with π, $3\pi/2$, and $5\pi/2$ pulses. The radiative lifetime is reduced from 800 to 540 ps, respectively. The inset in Fig. 3(b) shows a numerical fit (solid curve) of the power-dependent radiative lifetime (black dots), scaling as $T_1 = T_1^0/(1 + \alpha T_1^0 P)$. T_1^0 accounts for the power-independent lifetime, P is the pump power, and α is an adjustable parameter that is also used to fit the RO. Thus, the lifetime is inversely proportional to the pump power, i.e., to the square of the Rabi frequency. This additional relaxation term has been added phenomenologically in

the Bloch equations, substituting T_1 in Eq. (1) by its power-dependent expression defined previously. The oscillation curve in Fig. 3(a) is now accurately fitted.

We believe that the power dependence of the lifetime is specific to the coupling between the dipole emission and the 1D WG mode. The WG modifies the structure of the electromagnetic environment, changes the emission properties of the dipole, which is inside, and as a consequence modifies the emission rate [31,40]. This modification is not uniform for all the optical modes but occurs only with the WG resonant mode. We can assume in a first approximation that the emission rate to all other radiative modes remains unchanged. If we define the fraction of spontaneously emitted light into a given mode, the so-called coupling factor β, we can write the total *resonant* emission rate $\gamma_R = 1/T_1$ as $\gamma_R = (1-\beta)\gamma_0 + \beta(\gamma_0 + \gamma_{wg}) = \gamma_0 + \beta\gamma_{wg}$. γ_0 is the spontaneous emission rate in the sample, and γ_{wg} is the modified emission rate due to the resonant coupling to the 1D optical mode.

As a consequence, the modified emission rate γ_{wg} will be power dependent. To show it qualitatively, we may use the results of cavity QED in the strong coupling regime [28,41,42]. Although our QDs are not coupled to cavity photons, they are resonantly coupled with those of the 1D single-mode WG in the nonlinear Rabi regime. The modification of the emission rate allows us to define an enhancement factor F, which is the ratio between γ_{wg} and γ_0, F being analogous to the well-known Purcell factor F_p [43]. In the strong coupling regime, the generalized Purcell factor reads $F_p = (4g^2)/(\gamma_c\gamma_0)$, with γ_c the intracavity decay rate and g the light-matter coupling [28,42]. The strong coupling regime holds when $g > \gamma_c, \gamma_0$. In pulsed experiments, the relevant parameter equivalent to $1/\gamma_c$ is the pulse width τ, equal to 2 ps in our case. The coupling parameter g is related to the Rabi frequency Ω_R and, thus, to the number of photons N ($N \gg 1$) in the pulse [31]: $(2g)^2 = \Omega_R^2 = N(2g_0)^2$, g_0 being the single photon coupling. Therefore, $\gamma_{wg} = N(2g_0)^2\tau$, and the modified rate scales like the pump power, which is exactly what we observe experimentally. The coupling factor β can be estimated using the value of the fitting parameter α and γ_R defined above. β is characteristic of the WG structure, and in the case of 1 μm^2 ridges dimensions, we find a few percent (3% in the case of the studied dot). Achieving a large β factor by reducing the WG cross section dimensions would be of great interest for the development of very efficient single-photon sources. Although in our etched structures it is rather difficult to reduce the ridge dimensions, this can be obtained in other geometries such as those in nanowires [29] or photonic crystal waveguides [30], where very large β coupling factors up to 95% have been reached. In our sample structure, the fact that γ_{wg} varies from dot to dot is related to the light-matter coupling g_0, which depends mainly on the position of the dot with respect to the maximum of the field amplitude. Therefore, reducing the dimensions of the WG would have as a consequence the enhancement of the resonant light-matter coupling due to the larger overlap between the dot absorption cross section and the cross section of the optical mode.

We have presented two extreme cases of dots, types A and B with $\mathcal{L}_\infty \to \mathcal{L}_{\max}/2$ and $\mathcal{L}_\infty \to 0$, respectively. However, different situations have been encountered with $\mathcal{L}_\infty$ ranging between these two values. For all dots, the damping rate K caused by phonons is of the same order of magnitude whereas the population relaxation rate α is related to the modification of the emission rate and thus dot dependent. The resonant coupling to the 1D WG mode is therefore responsible for the differences observed in the oscillations damping.

In summary, we have shown the dominant role of acoustic phonons in the EID processes that inherently limit the coherence of the system. The linear temperature dependence of the measured decoherence rate supports this result. For a certain number of dots well coupled to the 1D waveguiding mode, an additional damping related to an excitation-induced relaxation of the population is observed. The spontaneous emission rate enhancement scaling with the pump power acts as a leak of population that can lead in certain cases to a complete vanishing of the resonant luminescence at high excitation. Further investigation with longer than 2 ps pulses is currently being carried out, since interesting effects have been predicted theoretically. In particular, a low quality of Rabi oscillations has been calculated for ultrashort pulses whereas a decrease of the damping has been predicted with longer pulses [20,21]. This could be a way to minimize the EID and achieve more reliable coherent operations.

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