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Dynamics of InP/(Ga,In)P quantum-dot single-photon emitters

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Abstract. The dynamics of single photon emitters based on InP/(Ga,In)P quantum dots (QDs) has been studied by time-resolved photoluminescence (TRPL) and photon correlation spectroscopy (PCS) up to 50 K. The increase of temperature produces marked effects on the exciton photoluminescence (PL) decay time (τ_X) and on the anti-bunching time (τ_R) in the second order photon correlation function. Both times are found to depend on the QD size. A competition of the thermally activated Dark-to-Bright (D→B) exciton transition and the thermal excitation of the carriers to- and from the QD give a good qualitative understanding of the observed results. Resonant excitation at the QD p-states produces a marked decrease of τ_R together with the appearance of time bunching in a longer time scale.

Keywords: Quantum Dot, Single Photon Emitter, Dark Exciton.

PACS: 78.67.Hc, 78.55.Cr, 42.5.Ar

INTRODUCTION

Semiconductor quantum dots (QDs) are very good candidates for optoelectronic devices, especially for single photon emitters (SPEs) in quantum information applications. It is of great importance to understand the intrinsic processes governing the QD emission to realize an efficient SPE device. In this work we study the optical properties of InP/(Ga,In)P QDs and in particular the influence of temperature, QD size and excitation energy on a single QD dynamics.

RESULTS AND DISCUSSION

Optical emission of single QDs has been studied by time-resolved photoluminescence (TRPL) and photon correlation spectroscopy (PCS) up to 50 K. Self-assembled InP/(Ga,In)P QDs grown by molecular beam epitaxy were selected from the high energy tail of the ensemble PL emission in order to study single QDs with reduced influence of the neighboring ones. Three emission lines assigned to the exciton (X), charged exciton (CX) and biexciton (XX) have been observed in the micro-PL spectra for all QDs under study. The relative size of the QD was inferred from the values of the fine structure splitting Δ_{FS} and the biexciton binding energy E_b^{XX} [1]. In this work we present results for two representative, single QDs with

different sizes. The excited states (p-states) for each QD have been identified by micro-PL excitation (PLE) measurements. We observe several peaks in the PLE spectra, which are indicative of excited states and of phonon-assisted absorption. Using the PLE results we have measured TRPL and PCS under non resonant excitation, below the wetting layer (WL) absorption, or in resonance with a given excited state, what minimizes the influence of carrier diffusion on the measured times. The TRPL and PCS results indicate that both the X decay time, τ_X , and anti-bunching time, τ_R , are approximately twice longer for X as those obtained for XX. With increasing excitation power τ_X increases due to XX formation which delays the recombination of X. For the smaller QD τ_X is approximately twice longer than that of the larger QD. The trend of the τ_X values confirm the QD relative size inferred from PL, in agreement with calculations by Wimmer *et al.* [2].

We have observed marked differences in the TRPL and PCS temperature behaviour depending on the QD size. Upon increasing temperature, both τ_X and τ_R increase in the small QD (Fig. 1(a) and 1(c)). This is a result of the thermal excitation of carriers, which occurs at low activation energies (E_{th}). E_{th} can be obtained from the temperature dependence of the PL intensity ratio of X and XX [3]. For the small QD E_{th} is approximately 7 meV. In the larger QD (Fig 1(b)

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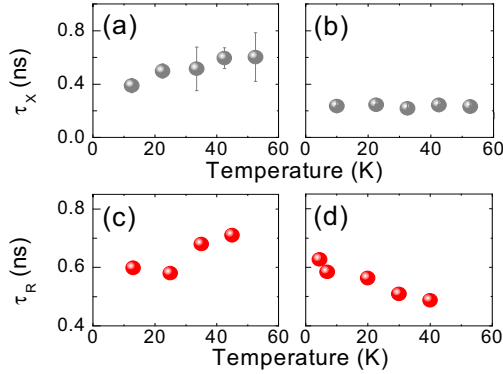


FIGURE 1. Temperature dependence of the exciton decay time τ_X and anti-bunching time τ_R for a small (a and c) and a large (b and d) QDs. The difference in the τ_X and τ_R behavior between both QDs reflects the competition of two thermally activated processes due to the different QDs sizes (see text).

and 1(d) this activation energy is higher (30 meV) and the D→B exciton transition dominates, reducing τ_R . The τ_X value of the large QD is independent of temperature as a result of the compensation between the effects: 1) the D→B transition, which reduces τ_X , and 2) the thermal scattering of carriers in the QD, which delays their arrival at the ground state and therefore increases τ_X .

For the small QD we have measured the autocorrelation function of the X emission under quasi-resonant excitation at the p-state. The experiment was run at 15 K and for different values of the excitation energy detuning ($\delta = E_p - E_{ex}$) in the range from -0.5 meV to 0.6 meV. The p-state is about 23.6 meV above the detection energy at X ($E_{det} = 1.861$ eV). Both anti-bunching and bunching peaks are observed in the $g^{(2)}(\tau)$ plot (Fig. 2(a)). A narrowing of the anti-bunching peak is observed for $\delta \rightarrow 0$. To estimate the anti-bunching rate Γ_R ($\Gamma_R = \tau_R^{-1}$) we use a phenomenological equation for $g^{(2)}(\tau)$ [4]:

$$g^{(2)}(\tau) = 1 - (1 + a) \cdot e^{(-\tau|\Gamma_R)} + a \cdot e^{(-\tau|\Gamma_D)}, \quad (1)$$

where Γ_D is the bunching rate and a is a fitting parameter. Γ_R has a maximum at $\delta \rightarrow 0$ following the PLE curve (Fig. 2(b)). The explanation for this behavior could be an increase of the absorption probability and in consequence a faster electron - hole pair (EHP) formation while $\delta \rightarrow 0$. Thus, the recharge process after emission of the previous photon is shorter than for off-resonance excitation. In addition, excitation below WL reduces the influence of carrier diffusion on the measured rates. A slight increase of Γ_R with excitation power (P_{ex}) is also observed

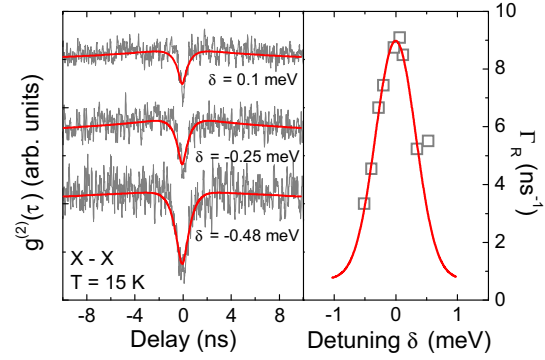


FIGURE 2. (a) Second order correlation function $g^{(2)}(\tau)$ under quasi-resonant excitation at the p-state for three different excitation energy detuning δ ; (b) anti-bunching rate Γ_R (\square) vs. detuning δ and Gaussian fit (—).

although keeping the dependence on δ shown in Fig. 2. The Γ_R increase with P_{ex} can also be explained in terms of increased probability of EHP presence in the p-state under stronger optical excitation.

Summarizing, we have presented a study of TRPL and PCS in InP/GaInP single QDs. We observe different temperature behaviour of τ_X and τ_R depending on the QD size. In small QD the thermal excitation of carriers increases both characteristic times. In the large QDs the D→B transition dominates and compensates the thermal scattering. This decreases τ_R and keeps τ_X independent of temperature. The PCS results reveal the dependence of the single QD dynamics on the excitation energy. The Γ_R increase as the detuning to the p-state decreases has been discussed in terms of the EHP formation probability in the QD excited state.

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