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Radiative and non-radiative relaxation of excitons in strain-compensated quantum dots

M. Kujiraoka[a,b,*], J. Ishi-Hayase[a], K. Akahane[a], Y. Yamamoto[a], K. Ema[b], M. Sasaki[a]

[a]National Institute of Information and Communications Technology, 4-2-1 Nukui-Kitamachi, Koganei, Tokyo 184 8795, Japan
[b]Department of Physics, Sophia University, 7-1 Kioi-cho, Chiyoda-ku, Tokyo 102 8554, Japan

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Abstract

We have investigated the population dynamics of excitons in strain-compensated InAs quantum dots (QDs) using a pump–probe technique under resonant excitation. Precise control of polarization directions of incident pulses enabled us to selectively estimate population lifetimes for two orthogonally polarized exciton ground states according to polarization selection rules. Measured decay times of the probe transmissions were highly dependent on the polarization directions of the exciton states. We found that the ratio of the decay times for the orthogonally polarized states is in quantitative agreement with the ratio of square of the transition dipole moments. This indicates that radiative recombination processes have a dominant effect on the population dynamics and that non-radiative and spin relaxations are negligible in our QDs. As a result, we can estimate the radiative lifetimes to be 1.0 ± 0.1 and 1.7 ± 0.2 ns for orthogonally polarized exciton ground states.

Keywords: Quantum dots; Exciton radiative lifetime; Optical anisotropy

1. Introduction

Semiconductor quantum dots (QDs) emitting light at optical telecommunication wavelengths have attracted much interest because of their possible applications in optical and quantum communication devices [1,2]. Radiative and non-radiative relaxations of excitons play important roles in determining the performance of such devices.

Recently, the advantages of InAs QDs using an InP(3 1 1)B substrate have been reported [3]. These advantages include a longer-wavelength emission and a defect-free stacked structure compared with QDs on a GaAs substrate. Because of the orientation of (3 1 1)B substrates, QDs grown on an InP(3 1 1)B substrate should show an optical anisotropy, which is analogous to quantum wells grown on a (3 1 1)B substrate [4]. The radiative lifetime of exciton in QDs grown on an InP(3 1 1)B substrate is thus expected to show an in-plane anisotropy reflecting differences of transition dipole moments. The problem is that a polarization-dependent radiative lifetime is difficult to precisely determine from a temporal profile of time-resolved photoluminescence (PL) under non-resonant excitation, which is the most common method to measure a radiative lifetime.

We investigated the population lifetime of excitons in strain-compensated InAs QDs using a polarization-dependent pump–probe technique under resonant excitation. The polarization characteristics provide the detailed information on individual radiative lifetimes for two orthogonally polarized ground states of excitons as well as non-radiative and spin relaxations [5].

2. Experiment

A sample was fabricated using molecular beam epitaxy and contained 150 layers of InAs self-assembled QDs embedded in 60 nm-thick $In_{0.52}Ga_{0.1}Al_{0.38}As$ spacers on an InP(3 1 1)B substrate [3]. To compensate for QD strain

*Corresponding author at: Advanced Communications Technology Group, National Institute of Information and Communications Technology, 4-2-1 Nukui-Kitamachi, Koganei, Tokyo 184 8795, Japan.
Tel.: +81 42 327 5264; fax: +81 42 327 6629.
E-mail address: m-kujira@nict.go.jp (M. Kujiraoka).

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originating from the lattice mismatch, the composition of spacers was precisely tailored and an InP(3 1 1)B substrate was used. This strain compensation technique enabled us to control the emission wavelength and to increase the interaction length by stacking up to 150 QD layers. Corresponding effective QD density was up to $\sim 1.3 \times 10^{13}$ dot/cm^2. The stacked structure of our sample is advantageous because it enhances weak nonlinear signals [6]. From an atomic force microscopy image, the shape of our QDs is found to be an ellipse that is elongated in the $[\bar{2}\,3\,3]$ direction, which is one of the crystal axes in the (3 1 1)B surface. The average-lateral size of QDs was estimated to be 51 nm in the $[\bar{2}\,3\,3]$ direction and 39 nm in the $[0\,1\,\bar{1}]$ direction, which is orthogonal to the $[\bar{2}\,3\,3]$ direction. Due to the broken rotational symmetry, exciton ground states are split into two orthogonally polarized non-degenerate states, whose polarizations are in the $[\bar{2}\,3\,3]$ and $[0\,1\,\bar{1}]$ directions (See Fig. 1). We use the abbreviation ***x*** (***y***) to represent the $[0\,1\,\bar{1}]$ ($[\bar{2}\,3\,3]$) direction. The backside of our sample was antireflection-coated to prevent multiple reflections.

A transmission-type pump–probe technique was used to investigate exciton population dynamics. The optical excitation consisted of 1.1 ps optical pulses generated by an optical parametric oscillator. The wavelength of the excitation pulses was tuned to the central wavelength of exciton ground-state emission, 1468 nm, as shown in Fig. 1. The excitation density of pump pulses was set at 16 kW/cm^2, which corresponds to the excited exciton density of less than one exciton per one QD. The excitation density of probe pulses was set at 0.5% of that of the pump pulses. Due to the weakness and narrowband width of the excitation, contributions of biexciton formations were ignored. The pump and probe pulses were linearly co-polarized. By controlling the polarization direction of incident pulses, we could selectively excite either of the ***x***- or ***y***-polarized states. All measurements were performed at 3 K.

3. Results and discussion

Fig. 1 shows the polarized PL spectra under non-resonant excitation. The PL spectra have maximum and minimum intensities in ***y*** direction (solid line) and ***x*** direction (dashed line), respectively. The degree of polarization defined as $(I_y - I_x)/(I_y + I_x)$ is evaluated to be approximately 16%. This in-plane anisotropy in QDs is due to the asymmetry of the QD shape and anisotropic strain [7].

Figs. 2 and 3 show the change of probe transmission for various polarization directions. The signal profile is highly dependent on the polarization direction of excitation pulses. In Fig. 2, we note that the oscillatory signal is observed only when the polarization direction is rotated 45° (Fig. 2(b)) with respect to ***x*** direction. This oscillation

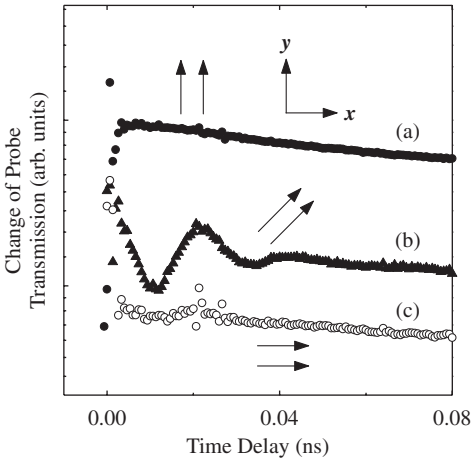

Fig. 2. Change of probe transmission at 3 K when polarization directions were in (a) ***y*** and (c) ***x*** directions and (b) directions rotated 45° with respect to ***x*** direction. Arrows represent polarization directions of incident pulses.

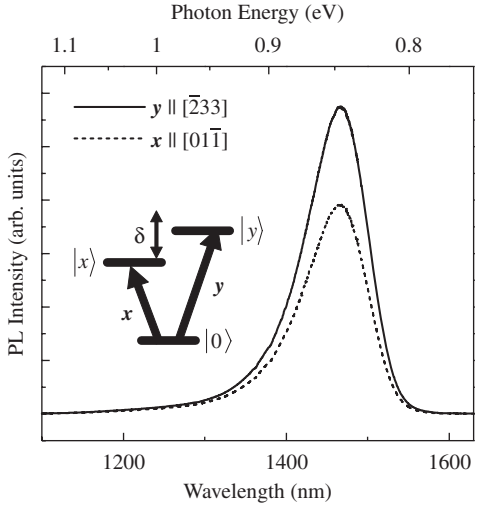

Fig. 1. Polarization dependence of photoluminescence spectra at 3.2 K under non-resonant laser excitation. Inset: energy-level diagram and optically allowed transitions related to exciton ground states.

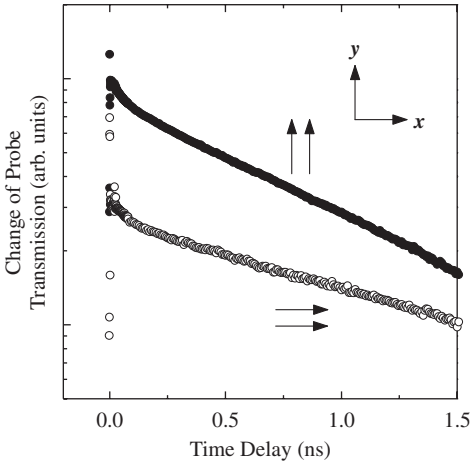

Fig. 3. Change of probe transmission at 3 K for ***y*** (filled circles) and ***x*** polarizations (open circles). Arrows represent polarization directions of incident pulses.

almost vanishes for x- or y-polarized pulses (See Figs. 2(a) and (c)). These results confirm that x- or y-polarized pulses enable us to selectively excite either of the two-exciton ground states. This is because the oscillation is due to a quantum beat, which results from a simultaneous excitation of x- and y-polarized excitons. The period of the oscillation estimated by fitting the data to a damped harmonic oscillation function was 23 ps, which corresponds to an energy difference of 180 μeV. The damping of the quantum beat originates from the fluctuation in splitting energy [8].

Fig. 3 shows the changes in probe transmission for x-(open circles) and y-(filled circles) polarized pulses measured for time delays of up to 1.5 ns. The intensity and decay time of exponentially decaying components are highly dependent on the polarization direction of excitation pulses. By fitting single exponential functions, signal decay times for x and y polarizations (τ_x and τ_y) were estimated to be $\tau_x = 1.7 \pm 0.2$ and $\tau_y = 1.0 \pm 0.1$ ns, respectively.

When determining the reason for decay in the probe transmission, a resonant weak excitation allows us to exclude the effect of carrier relaxations from energetically upper states to exciton ground states. Excluding this effect is a crucial advantage compared with time-resolved PL under non-resonant excitation [9]. The effect of biexciton formations can also be excluded. In our experiments, a signal decay time is determined by radiative recombination processes as well as spin relaxations and non-radiative recombination processes [5]. In general, a radiative lifetime T_1 is inversely proportional to the square of a transition dipole moment $|\mu|^2$. In our sample, the ratio of the square of the transition dipole moments $|\mu_y|^2/|\mu_x|^2 = 1.75 \pm 0.04$ was independently estimated by polarization-dependent four-wave mixing measurements [10]. The ratio is approximately consistent with the ratio of the decay times, i.e., $\tau_x/\tau_y = 1.7 \pm 0.4$. This comparison demonstrates that the decay time of probe transmission is determined by the radiative lifetime T_1 of selectively excited ground states. Therefore, we conclude that ${T_1}^x = 1.7$ ns for x-polarized state and ${T_1}^y = 1.0$ ns for y-polarized state. Our results also indicate that spin and non-radiative relaxations are negligible in our QDs. This is a great advantage to improve the emission efficiency in a luminescent diode and a single-photon emitter using QDs. Furthermore, the reduced relaxation channels would contribute to elongate the coherence time, which is one of most important factors in quantum information devices. Therefore, our QDs have many advantages for application in optical and quantum information devices.

4. Conclusions

We performed a polarization-dependent pump–probe spectroscopy under weak resonant excitation in InAs strain-compensated QDs on an InP(3 1 1)B substrate. The decay times of probe transmissions showed large in-plane anisotropy. From our quantitative analysis, we found that radiative lifetime is the dominant effect acting on the population dynamics due to negligible spin and non-radiative relaxations. Therefore, the anisotropy of the decay times reflects the anisotropy of radiative lifetimes. As a result, we can estimate radiative lifetime for each of x-and y-polarized exciton ground state precisely, i.e., ${T_1}^y = 1.0 \pm 0.1$ ns for y-polarized state and ${T_1}^x = 1.7 \pm 0.2$ ns for x-polarized state.

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