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Time-Resolved characterization of InAsP/InP quantum dots emitting in the C-band telecommunication window

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The dynamic response of InAsP quantum dots grown on InP(001) substrates by low-pressure Metalorganic Vapor Phase Epitaxy emitting around $1.55\text{ }\mu\text{m}$, is investigated by means of time-resolved microphotoluminescence as a function of temperature. Exciton lifetime steadily increases from 1 ns at low temperature to reach 4 ns at 300K while the integrated photoluminescence intensity decreases only by a factor of 2/3. These characteristics give evidence that such InAsP/InP quantum dots provide a strong carrier confinement even at room temperature and that their dynamic response is not affected by thermally activated non-radiative recombination up to room temperature.

In recent years, self-assembled semiconductor quantum dots (QDs) have been considered for utilization in quantum information processing (such as quantum communication [1, 2, 3]), in addition to their conventional applications in optoelectronics, for example. However, optoelectronics and the implementation of practical quantum computation networks require mostly the use of QDs emitting in the telecommunications C-band ($1.53 - 1.56\text{ }\mu\text{m}$). Emission at wavelengths around $1.55\text{ }\mu\text{m}$ has already been achieved from QDs grown on a GaAs substrate by means of different techniques, for instance by unusually low temperature growth [4] or epitaxial growth on metamorphic buffer layers [5, 6]. Yet, one of the most attractive material combinations for fabricating QDs emitting in the C-band is InAs on InP substrate. Most of the studies, involving high-vacuum growth techniques such as molecular beam epitaxy, [7, 8] of InAs QDs on (001)-oriented InP substrates, demonstrated the formation of strongly elongated nanostructures (namely, “quantum dashes” or “quantum sticks”) rather than three-dimensional QDs. To circumvent this difficulty, a few groups have used (113)B-oriented InP substrates [9], which are however not compatible with the standard processes used in the fabrication of optoelectronic devices. Metalorganic vapor phase epitaxy (MOVPE) has also been used for the growth of InAs/InP(001) QDs. Various studies have shown that MOVPE allows one to obtain QDs rather than quantum dashes [10, 11]. Recently, microphotoluminescence signals around $1.5\text{ }\mu\text{m}$ evidenced sharp spectral features corresponding to radiative transitions from trapped single electron-hole pairs in such InAs/InP QDs grown by MOVPE [13, 14, 15]. Conversely, not much work has been done on the dynamic response of such MOVPE-grown dots and its dependence as a function of the temperature [19]. Yet, measurements of this dynamic response and the impact of thermally activated non-radiative mechanisms are quite important to assess the potential of such QDs devices for example for high-speed direct modulation or efficient generation of quantum states of light at high temperature (more than 77 K). This study forms the basis of this letter. The de-

pendence of the large measured radiative lifetime and the small decrease of the integrated intensity as a function of temperature demonstrate the high structural quality of these QDs, which offer strong carrier confinement: non radiative recombination inside the QDs and carrier evaporation to the wetting layer are not critical to the operation of such long-wavelength QD-based devices up to 300 K.

Samples were grown in a vertical-reactor low-pressure MOVPE system using hydrogen as the carrier gas and standard precursors [arsine, phosphine, and trimethylindium] [11]. The quantum dots are grown on a thick InP buffer layer deposited on an exactly (001)-oriented InP substrate. The QDs are formed at a lowered temperature of $510\text{ }^{\circ}\text{C}$ by depositing 6.3 monolayers (ML) of InAsP at growth rate of 0.36 ML.s^{-1} and under a phosphine/arsine flow ratio of 30. Finally, a 63 nm thick InP capping layer is grown over the QDs at a rate of 0.2 ML.s^{-1} . Such a growth sequence leads to the formation of InAsP QDs with an average height of 3.8 nm and a density of $15 \times 10^9\text{ cm}^{-2}$, measured by Transmission Electron Microscopy (TEM) experiments [17]. Time-resolved microphotoluminescence characterization of samples was obtained under pulsed excitation with a 5 ps pulse width at 80 MHz delivered by a Ti:Sa laser emitting at 840 nm. The excitation pulses were focused on the samples by a microscope objective (Numerical Aperture (NA) = 0.4). The excitation spot spreads over roughly $5\text{ }\mu\text{m}$ on the sample, corresponding to the excitation of approximately 2.9×10^3 quantum dots. The QDs luminescence is collected by the same microscope objective and separated from the pumping laser by means of a dichroic mirror and an antireflection coated silicon filter. The spontaneous emission is spectrally dispersed by a 0.5 m spectrometer and detected either by a cooled InGaAs photodiode array (Roper Scientific) or a time-resolved single photon counter (SCONTEL) with a time resolution of 50 ps, a measured quantum efficiency of 3% at $1.55\text{ }\mu\text{m}$ and dark count rates lower than 30 counts per second. The histogram of the time interval between the photon detection and the subsequent laser pulse is

recorded by a LeCroy 8620A oscilloscope.

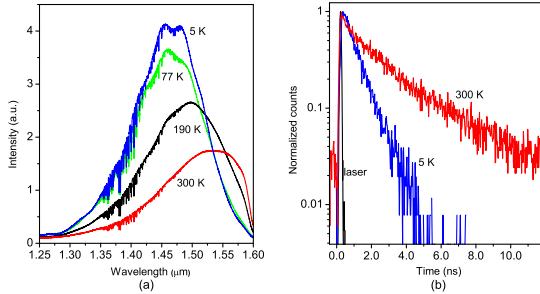


FIG. 1: (a) Emission spectra of the ground states of InAsP/InP QDs under pulsed excitation as a function of temperature. (b) Typical lifetime decay curves for QDs at 5 K (blue) and room temperature (red).

All the following experiments are performed under low excitation power (of the order of 200 W/cm^2), so that only the fundamental transition of the QDs (one trapped electron-hole pair per dot at most) contributes to the optical emitted spectrum at low temperature (5 K). For each temperature a photoluminescence spectrum is recorded (fig. 1(a)) and emission lifetime is measured (fig. 1(b)). Emission spectra show excess noise in the 1350-1410 nm region arising from water absorption in air. Due to the QD's composition (InAs_{0.82}P_{0.18}) [17], leading to a smaller energy difference between subsequent monolayers, it is difficult to distinguish in such spectra different QD families; yet, it is possible to fit the photoluminescence spectra with a good fidelity by considering an 8-modal distribution of dot height. The low-temperature large inhomogeneous linewidth of the spectra is mainly due to height dispersion of the dots [12]. It broadens with temperature due to the thermally activated population of the first excited states in the QDs, while the photoluminescence peak intensity decreases. The abrupt decrease of the photoluminescence around 1600 nm (775 meV) arises from the cutoff of the InGaAs detector array. Due to the redshift of the fundamental transition with temperature, lifetime measurements were performed at the maximum of the distribution probing the same quantum dot family (same height in monolayers) [16].

The measured rise time of the order of 100 ps is limited by the timing resolution of the set-up and does not correspond to the carriers capture time in the QDs. At temperatures below 200 K, the experimental data can be fitted by a monoexponential decay, with a constant decay time of the order of 1 ns for $T < 77 \text{ K}$ (see Figure 2). Above 77 K and up to 200 K, the measured lifetime increases from 1 ns to 1.8 ns. At higher temperatures ($\geq 200 \text{ K}$), the data are well fitted by a bi-exponential decay (see fig. 1(b)) with a short decay time of the order of 800 ps and a long decay time of the order of few ns, plotted in fig. 2. We only observe a small contribution of the short decay photoluminescence, leading to large error

bars, and it seems to display a flat dependence as a function of temperature, while the long decay time increases from 1.8 ns to 4.2 ns. We attribute the short decay to the emission of thermally excited states (such as an electron-hole pair on a p shell) of lower energy QDs and the long decay time to the emission of the ground states of the QDs at the maximum of the dots distribution. This hypothesis is corroborated by lifetime measurements under higher excitation power (not shown here).

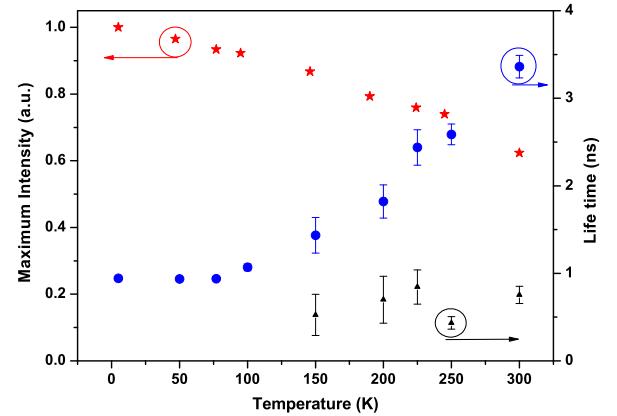


FIG. 2: Measured lifetime (circles) and photoluminescence peak intensity (stars) of the QDs as a function of temperature. At temperatures above 200 K, the decay curves are fitted by a biexponential decay with a fast (triangles) and a long decay time (circles).

The constant lifetime value at low temperature (here 1 ns up to 77 K) and the subsequent increase with temperature (here from 1 ns to 4.2 ns for temperatures ranging from 77 K to 300 K) has already been observed on InAs/GaAs quantum dots [16, 18, 19]. This increase is due to the thermal dissociation of the exciton and the excitation of the carriers to trapped excited states. Such states can be either a hole or an electron in the p-shell, while the electron (or hole) stays in the s-shell. These states are non-radiative and the electron (or the hole) has to fall back to the s-shell in order to emit a photon, which induces an increase of the measured lifetime with no change on the total number of emitted photons. These lifetime measurements suggest that the optical properties of these dots are little affected by thermally activated non-radiative mechanisms, which is corroborated by measurements of the integrated photoluminescence intensity as a function of temperature. Although the current experimental setup does not allow to deduce directly such value due to the cutoff of the InGaAs array, an approximate value can be inferred by fitting the spectra with a gaussian curve on the high energy part of the distribution. These inferred values are plotted in fig 2. A slight decrease of the integrated photoluminescence intensity can be observed as the temperature rises, together with an increase of the lifetime. Since the repetition rate of the

laser is 12 ns, one can not neglect the impact of the long QDs lifetime of the order of few ns, which will induce a decrease in the integrated intensity. Without correction, this decrease between 5 K and 300 K is of only a factor of one third at most.

The observed weak impact of non-radiative processes in such QDs are in good agreement with previous theoretical studies on InAs/InP quantum dots [20], indicating that the InAs/InP system provides a stronger hole confinement compared to its InAs/GaAs counterpart, with an hole energy spacing ranging from 20 to 40 meV compared to less than 20 meV for InAs/GaAs[20]. Electron energy spacing is higher than 50 meV for both quantum dots. This feature could explain the fact that no significant change in the exciton lifetime is observed below 100 K ($kT \simeq 8$ meV). Above 100K, one could expect a decrease in the integrated signal, since the p shell of the hole, whose wavefunction spreads more significantly in the barriers and thus is more sensitive to surrounding defects, is thermally populated. While such a behavior is significantly observed in InAs/GaAs quantum dots, we only observe a small decrease. The experimental result is in good agreement with the theoretical estimations. While for InAs/GaAs less than 85% of the hole wave-

function lies inside the quantum dot, it is estimated to be more than 97% for InAs/InP QDs for any trapped excited state[20]. We expect that the small phosphorous incorporation during growth should not change drastically the hole confinement. Consequently, such InAsP/InP QDs should be less sensitive to any non-radiative defects located in the surroundings.

In conclusion, we have measured the decay times and the integrated photoluminescence intensity as a function of temperature from the ground states of InAsP/InP quantum dots grown by MOVPE emitting at 1.55 μm . The increase of the lifetime and the slight decrease of the integrated intensity evidence the outstanding structural quality of the QDs and that their luminescence properties are not degraded by any non-radiative thermally activated mechanisms up to room temperature. Such high quality quantum dots could be used as gain medium for the implementation of semiconductor laser diodes or optical amplifier operating at room temperature as well as for the implementation of single photon source for quantum communications operating at 77 K.

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