Growth and characterization of single quantum dots emitting at 1300 nm

Citation for published version (APA):
Alloing, B., Zinoni, C., Zwiller, V., Li, L., Monat, C., Gobet, M., Buchs, G., Fiore, A., Pelucchi, E., & Kapon, E. (2005). Growth and characterization of single quantum dots emitting at 1300 nm. *Applied Physics Letters*, *86*(10), 101908-1/3. Article 101908. https://doi.org/10.1063/1.1872213

DOI:
10.1063/1.1872213

Document status and date:
Published: 01/01/2005

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
• The final author version and the galley proof are versions of the publication after peer review.
• The final published version features the final layout of the paper including the volume, issue and page numbers.

Link to publication

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the “Taverne” license above, please follow below link for the End User Agreement:

www.tue.nl/taverne

Take down policy
If you believe that this document breaches copyright please contact us at:

openaccess@tue.nl

providing details and we will investigate your claim.

Download date: 07. Jun. 2026

APPLIED PHYSICS LETTERS **86**, 101908 (2005)

Growth and characterization of single quantum dots emitting at 1300 nm

B. Alloing,[a] C. Zinoni, V. Zwiller, L. H. Li, C. Monat, M. Gobet, G. Buchs, and A. Fiore[b]
Quantum Devices Group, Institute of Quantum Electronics and Photonics, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

E. Pelucchi and E. Kapon
Laboratory of Nanostructures, Institute of Quantum Electronics and Photonics, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

(Received 10 November 2004; accepted 25 January 2005; published online 3 March 2005)

We have optimized the molecular-beam epitaxy growth conditions of self-organized InAs/GaAs quantum dots (QDs) to achieve a low density of dots emitting at 1300 nm at low temperature. We used an ultralow InAs growth rate, lower than 0.002 ML/s, to reduce the density to 2 dots/μm^2 and an InGaAs capping layer to achieve longer emission wavelength. Microphotoluminescence spectroscopy at low-temperature reveals emission lines characteristic of exciton-biexciton behavior. We also study the temperature dependence of the photoluminescence, showing clear single QD emission up to 90 K. With these results, InAs/GaAs QDs appear as a very promising system for future applications of single photon sources in fiber-based quantum cryptography. *© 2005 American Institute of Physics*. [DOI: 10.1063/1.1872213]

Growth of self-assembled InAs/GaAs quantum dots (QDs) has been studied extensively over the past few years partly since these structures offer the prospect of temperature-independent ultralow-threshold lasers. The emergence of the field of quantum information and the development of quantum protocols for optical transmission of cryptographic keys has given a strong impetus to develop reliable sources of single photons. Single QDs obtained by strain-driven nucleation in the Stranski–Krastanov growth mode have been shown to emit single photons,[1–3] and thus represent a promising candidate for a solid-state, electrically pumped single-photon source.[1] However, most studies so far have concentrated on the $\lambda < 1000$ nm wavelength range, while fiber-based quantum cryptography requires an emission wavelength matching the 1300 or 1550 nm transmission window of optical fibers. Progress in this direction has been slow, due to the difficulty of growing sufficiently large and In-rich InAs/GaAs QDs for emission in the infrared, and because of the lower signal-to-noise performance of detectors in this wavelength range. Clear single QD emission (as proved by exciton-biexciton dynamics) was only demonstrated up to 1150 nm,[4] while discrete lines with unclear pump power dependence were reported for QDs emitting at 1300 and 1550 nm.[5,6]

In this letter, we describe a growth method which provides, at the same time, a low QD density in the 2–3 μm^{-2} range and an emission wavelength of 1300 nm *at low temperature* [i.e., ~1400 nm at room temperature (RT)]. This allows us to show the emission of single QDs at 1300 nm through a clear exciton-biexciton behavior. Moreover, we show a high-temperature stability of the luminescence of single QDs with well-isolated emission lines up to $T \sim 90$ K. These QDs may be thus suitable for single-photon sources at 1300 nm operating at liquid-nitrogen temperature.

A common approach to decrease the dot density is to grow a thin layer of InAs close to the critical thickness of the two- to three-dimensional growth mode transition.[7] But in this case, it is difficult to reach 1.3 μm wavelength emission since this requires large and thick QDs, i.e., a relatively large amount of InAs. Instead, we have used a combination of an ultralow InAs growth rate [<0.002 monolayer(ML)/s] and capping with an InGaAs layer to obtain, at the same time, low QD density and long-wavelength emission. The samples were grown by solid-source molecular-beam epitaxy on (001)-oriented undoped GaAs substrates. Two series of samples were grown for atomic force microscopy (AFM) and photoluminescence (PL) spectroscopy characterization. After oxide desorption, a 0.5 μm GaAs buffer layer was grown at 620 °C under a background As$_2$ pressure of 10^{-6} mbar. Then, 2.1 MLs of InAs were deposited at 505 °C to form QDs by self-assembly at different growth rates in the range 0.16–0.0012 ML/s. At the lowest growth rates, As$_2$ pressure was reduced to 5×10^{-7} mbar. In samples used for AFM, the substrate was immediately cooled down to RT just after InAs deposition and kept under an As overpressure. Figure 1

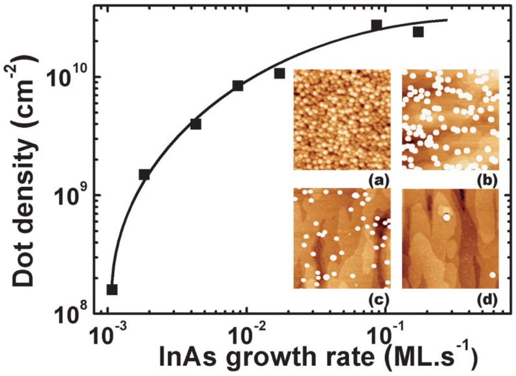

FIG. 1. (Color online) QD density, as measured from AFM images plotted as a function of InAs deposition rate. The solid line is a guide for the eyes. Inset: 1×1 μm^2 AFM images of 2.1 ML InAs deposited at the following growth rates: (a) 0.16 ML/s, (b) 0.08 ML/s, (c) 0.004 ML/s, (d) 0.0015 ML/s.

---

[a] Electronic mail: blandine.alloing@epfl.ch

[b] Permanent address: Institut of Photonics and Nanotechnology CNR-IFN, via del Cineto Romano 42, 00156 Roma, Italy.

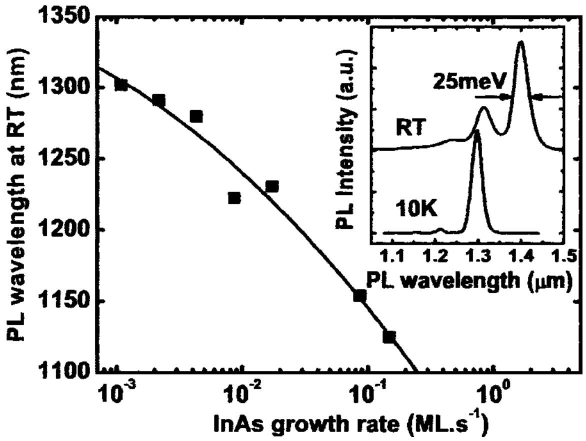

FIG. 2. Evolution of PL emission wavelength as a function of InAs growth rate. The solid line is a guide for the eyes. Inset: PL spectra at 10 K and RT of 2.1 ML InAs QDs grown at 0.002 ML/s with a 5 nm thick $In_{0.15}Ga_{0.85}As$ capping layer.

shows the QD density as a function of InAs growth rate: QD density decreases strongly from 3×10^{10} dots/cm^2 to approximately 2×10^{8} dots/cm^2 as InAs growth rate is reduced from 0.16 ML/s to 0.0012 ML/s. The values of QD density have been confirmed by transmission electronic microscopy investigation performed on InAs QDs capped by a GaAs layer. This trend was already reported for higher InAs growth rates.[8,9] According to these studies, the decrease of QD density is due to an increased migration length of In adatoms at low growth rates. In order to minimize strain and surface energy, In adatoms incorporate into existing dots instead of forming new dots.[9] This leads to a reduction in QD density and an increase in QD size.

To perform PL measurements, a second series of samples was grown under the same conditions except that the dots were capped by 100 nm thick GaAs. Figure 2, reporting the RT PL measurements, shows a redshift of PL peak emission wavelength, attributed to the increased QD size,[8] as the growth rate is decreased. Wavelength reaches 1310 nm at RT at the lowest growth rate. In order to further shift the PL emission to 1400 nm at RT, we used an InGaAs capping layer to reduce the strain and the In segregation from the QDs.[10,11] InAs QDs were grown at 0.002 ML/s and capped by a 5 nm thick $In_{0.15}Ga_{0.85}As$ layer followed by 100 nm thick GaAs layer. PL spectra from this sample are reported in the inset of Fig. 2, showing strong PL emission peak at 1400 nm (RT) and 1300 nm (10 K). This result allows spectroscopy investigations on single InAs QDs at low temperature.

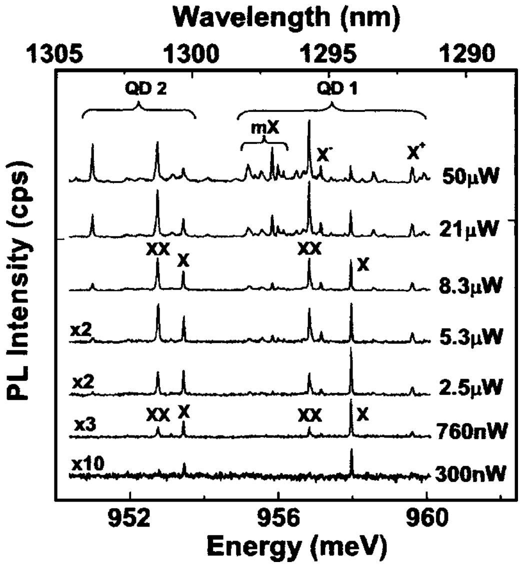

FIG. 3. PL spectra of QDs embedded in a microcavity (1 μm diameter mesa) at different excitation powers. Peaks X and XX are attributed to exciton and biexciton emission.

In order to increase the extraction efficiency and thus signal-to-noise ratio, we incorporated those QDs at the center of an epitaxied $1-\lambda$ microcavity with a 13.5 pair (one pair) $Al_{0.9}Ga_{0.1}As/GaAs$ bottom (top) Bragg mirror, respectively. This resulted in a 13-fold increase in the PL signal collected into a microscope objective of numerical aperture of 0.5. 1 μm^2 mesas were processed by photolithography and wet etching to isolate QDs. The emission from the QDs in the mesas was dispersed in a 1 m focal length spectrometer equipped with a linear array of InGaAs detectors (Jobin-Yvon). A Ti–Sa laser emitting at 835 nm was used as a pump source. PL spectra at 10 K obtained on one of these mesas are presented in Fig. 3 for different excitation powers. We selected a mesa showing two sets of lines, corresponding to the emission of two single QDs, to highlight the consistency of the observed behavior. Two sharp lines at 957.9 meV and 956.8 meV can be attributed to exciton (X) and biexciton (XX) recombinations, respectively, in a single QD (QD1), as evidenced by their excitation power dependence: At very low pump power only the X line is present, with a saturation and concomitant rise of the XX line at higher excitation levels. The same behavior is observed in QD2 for the two peaks at 953.4 meV and 952.7 meV. The integrated intensities of X and XX lines of QD1 are plotted in Fig. 4 as a function of excitation power. At low excitation power, the PL intensity dependence on the laser power P can be fitted (lines) by the relation $I_{X,XX}\propto P^n$, with $n=0.71$ and 1.4 for X and XX lines, respectively. The deviation from the ideal linear and quadratic dependencies has been observed before in QDs capped with InGaAs.[4] However the fact that $I_{XX}\propto I_X^2$ (see inset in Fig. 4) confirms that the XX line corresponds to biexciton emission. The biexciton binding energy of about 1 meV falls within the wide range reported for self-organized

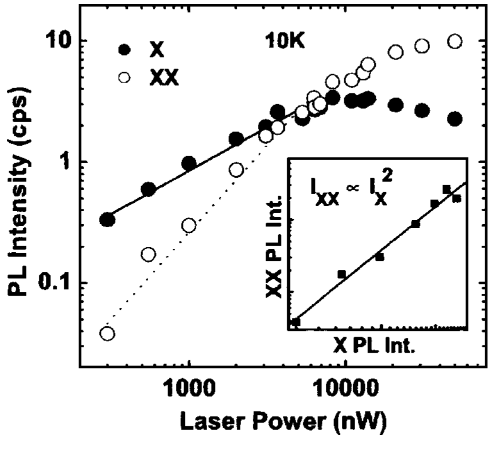

FIG. 4. Integrated intensities of X and XX peaks as a function of the laser excitation power, under continuous-wave excitation. Solid (dashed) line shows the fitted power dependence of X (XX) intensity and the inset reports the dependence of the XX-PL intensity versus the X-PL intensity in log-log scale.

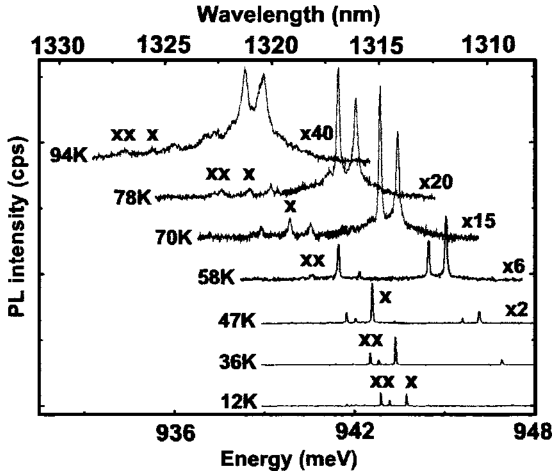

FIG. 5. PL spectra of a single QD in a microcavity (1 μm diameter mesa) at different temperatures (excitation power =1 μW, pump wavelength =850 nm).

In(Ga)As/GaAs QDs (from negatives values up to about 5 meV)[12] as determined by the balance of Coulomb interaction, correlation, and exchange. Other sharp lines appear at higher excitation power. According to their energy position[13,14] two of them can be attributed to negative (X^-) and positive (X^+) charged excitons. Moreover, we observed on several mesas other lines (mX) which appear concomitantly with the excited state emission and can therefore be attributed to multiexciton emission.[15] The ensemble of these observations confirms that the emission of single QDs in the 1300 nm region is clearly identified.

We further investigated the temperature stability of the single QD emission. Figure 5 presents PL spectra obtained on an other mesa at different temperatures: Sharp lines arising from discrete electronic states are seen at temperatures well above 77 K. As the temperature is raised, the X linewidth increases from 36 μeV (12 K, resolution limited) to 160 μeV (70 K), due to phonon-induced homogeneous broadening.[16] Moreover, the charged exciton lines increase at the expense of the X line, as previously reported.[17] We note that charged exciton line can also be used for single-photon emission.

In conclusion, we have shown that it is possible to reach low density dots with a wavelength emission at 1300 nm by optimizing growth conditions, i.e., drastically reducing InAs growth rate and using a InGaAs capping layer. Single InAs QD spectroscopy was performed and exciton/biexciton emission at 1300 nm was clearly observed at low temperature. The temperature stability of the emission suggests that these QDs may be used for single-photon sources operating at liquid-nitrogen temperatures. The low density should also allow the realization of a single-photon microcavity light-emitting diode.

The authors are grateful to Roger Rochat for his invaluable technical contribution and the Swiss National Science Foundation (NCCR Quantum Photonics and ”Professeur boursier” program) for financial support.

[1] P. Michler, A. Kiraz, C. Becher, W. V. Schoenfeld, P. M. Petroff, L. Zhang, E. Hu, and A. Imamoglu, Science **290**, 2282 (2000).
[2] C. Santori, M. Pelton, G. Salomon, Y. Dale, and Y. Yamamoto, Phys. Rev. Lett. **86**, 1502 (2001).
[3] V. Zwiller, H. Blom, P. Jonsson, N. Panev, S. Jeppesen, T. Tsegaye, E. Goobar, M. E. Pistol, L. Samuelson, and G. Björk, Appl. Phys. Lett. **78**, 2476 (2001).
[4] S. Kaiser, T. Mensing, L. Worschech, F. Klopf, J. P. Reithmaier, and A. Forchel, Appl. Phys. Lett. **81**, 4898 (2002).
[5] K. Takemoto, Y. Sakuma, S. Hirose, T. Usuki, and N. Yokoyama, Jpn. J. Appl. Phys., Part 2 **43**, L349 (2004).
[6] M. B. Ward, D. C. Unitt, Z. Yuan, P. See, R. M. Stevenson, K. Cooper, P. Atkinson, I. Farrer, D. A. Ritchie, and A. J. Shields, Physica E (Amsterdam) **21**, 390 (2004).
[7] D. Leonard, K. Pond, and P. M. Petroff, Phys. Rev. B **50**, 11 687 (1994).
[8] P. B. Joyce, T. J. Krzyzewski, G. R. Bell, T. S. Jones, S. Malik, D. Childs, and R. Murray, J. Cryst. Growth **227**, 1000 (2001).
[9] Y. Nakata, K. Mukai, M. Sugawara, K. Ohtsubo, H. Ishikawa, and N. Yokoyama, J. Cryst. Growth **208**, 93 (2000).
[10] M. V. Maximov, A. F. Tsatsul’nikov, B. Volovik, D. A. Bedarev, A. E. Zhukov, A. R. Kovsh, N. A. Maleev, V. M. Ustinov, P. S. Kop’ev, Zh. I. Alferov, R. Heitz, N. N. Ledentsov, and D. Bimberg, Physica E (Amsterdam) **7**, 326 (2000).
[11] N.-T. Yeh, T.-E. Nee, J.-I. Chyi, T. M. Hsu, and C. C. Huang, Appl. Phys. Lett. **76**, 1567 (2000).
[12] S. Rodt, R. Heitz, A. Schliwa, R. L. Sellin, F. Guffarth, and D. Bimberg, Phys. Rev. B **68**, 035331 (2003).
[13] L. Landin, M.-E. Pistol, C. Pryor, M. Persson, L. Samuelson, and M. Miller, Phys. Rev. B **60**, 16640 (1999).
[14] A. Kiraz, S. Fälth, C. Becher, B. Gayral, W. V. Schoenfeld, P. M. Petroff, L. Zhang, E. Hu, and A. Imamoglu, Phys. Rev. B **65**, 161303 (2002).
[15] J. J. Finley, A. D. Ashmore, A. Lemaître, D. J. Mowbray, M. S. Skolnick, I. E. Itskevich, P. A. Maksym, M. Hopkinson, and T. F. Krauss, Phys. Rev. B **63**, 073307 (2001).
[16] M. Bayer and A. Forchel, Phys. Rev. B **65**, 041308 (2002).
[17] K. F. Karlsson, E. S. Moskalenko, P. O. Holtz, B. Monemar, W. V. Schoenfeld, J. M. Garcia, and P. M. Petroff, Appl. Phys. Lett. **78**, 2952 (2002).