Epitaxial growth of quantum rods with high aspect ratio and compositional contrast

Citation for published version (APA):

Li, L., Patriarche, G., & Fiore, A. (2008). Epitaxial growth of quantum rods with high aspect ratio and compositional contrast. *Journal of Applied Physics*, 104(11), 113522-1/4. [113522].
<https://doi.org/10.1063/1.3032544>

DOI:

[10.1063/1.3032544](https://doi.org/10.1063/1.3032544)

Document status and date:

Published: 01/01/2008

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.

Epitaxial growth of quantum rods with high aspect ratio and compositional contrast

L. H. Li,^{1,a)} G. Patriarche,² and A. Fiore³

¹*Ecole Polytechnique Fédérale de Lausanne, Institute of Photonics and Quantum Electronics, Station 3, CH-1015 Lausanne, Switzerland*

²*LPN/CNRS, Route de Nozay, 91460 Marcoussis, France*

³*COBRA Research Institute, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands*

(Received 22 August 2008; accepted 10 October 2008; published online 9 December 2008)

The epitaxial growth of quantum rods (QRs) on GaAs was investigated. It was found that GaAs thickness in the GaAs/InAs superlattice used for QR formation plays a key role in improving the QR structural properties. Increasing the GaAs thickness results in both an increased In compositional contrast between the QRs and surrounding layer, and an increased QR length. QRs with an aspect ratio of up to 10 were obtained, representing quasiquantum wires in a GaAs matrix. Due to modified confinement and strain potential, such nanostructure is promising for controlling gain polarization.

© 2008 American Institute of Physics. [DOI: [10.1063/1.3032544](https://doi.org/10.1063/1.3032544)]

I. INTRODUCTION

Due to unique quantum confinement properties, semiconductor quantum dots (QDs) have attracted considerable interest in the past two decades. They have been extensively studied and successfully utilized in electronic and optoelectronic devices. For example, development of QD semiconductor optical amplifier (SOA) brings promising characteristics such as broadband amplification, high saturation output power, and ultrafast response.¹ However, the optical gain in the QD SOAs is polarization sensitive, which limits their applicability in fiber communication. Recently, it was suggested that this polarization sensitivity could be substantially reduced by employing columnar QDs.^{2–9} Columnar QDs with aspect ratio of as large as ≈ 4 were previously obtained by this technique.⁹ In these elongated nanostructures, which may be more appropriately termed quantum rods (QRs), the main quantization axis of the QRs is changed as compared to conventional QDs. Due to modified confinement and strain potential, the QRs can be used to control the gain polarization. Improved polarization properties were evidenced by photoluminescence (PL) and electroluminescence measurements.^{2,5,6} In electroluminescence measurements on the QRs with an aspect ratio of 3.5, below threshold, a transverse magnetic (TM)/transverse electric (TE) ratio of 0.8 has been achieved, far larger than that of the conventional QDs (about 0.1–0.2).⁶ However, above threshold, lasing is still TE-polarized, indicating a still dominantly heavy-hole character of the valence-band ground state. Recent calculations show that the strain from the two-dimensional (2D) InGaAs layer surrounding the QRs is responsible for this.¹⁰ Increasing the In compositional contrast between the QR (x_{QR}) and the 2D layer ($x_{2\text{D}}$), while keeping or further increasing the aspect ratio, is needed to favor TM gain and lasing. In our

previous experiments,⁶ $x_{\text{QR}}=0.35$ and $x_{2\text{D}}=0.16$, which are insufficient to achieve dominant TM gain. Lowering the $x_{2\text{D}}$ would be particularly beneficial for enhancing the TM gain. Indeed, for obvious symmetry reasons, QRs embedded in GaAs would have a TM-polarized dipole.⁴ However, decreasing the $x_{2\text{D}}$ and simultaneously keeping the formation of the QRs is very difficult. A compromise must be found. In this paper, we report the growth of QRs with improved structural properties. QRs with an increased In compositional contrast ($x_{\text{QR}}/x_{2\text{D}} > 3$), along with a large aspect ratio of up to 10, were obtained, which enable the realization of TM-polarized lasers.

II. EXPERIMENTAL PROCEDURE

The samples were grown by molecular beam epitaxy (MBE) on (001)-oriented GaAs substrate. The QRs were formed at the growth temperature of 500 °C by depositing a 1.8 ML InAs QD seed layer and a short period GaAs/InAs superlattice (SL).^{2,7} After growth of each InAs layer, a growth interruption of 5 s was applied in order to make the QR size distribution more uniform. The growth rates of GaAs and InAs were 0.7 and 0.1 ML/s, respectively. The samples were completed by a 100-nm-thick GaAs cap. During the growth, the QR evolution was monitored *in situ* by reflection high-energy electron diffraction (RHEED). For high-quality QR growth, alternative appearance of streaky diffraction rods and chevrons related with GaAs and InAs layer must be observed.^{3,7,8} After growth, the structural and optical properties of the samples were characterized by transmission electron microscopy (TEM), high resolution x-ray diffraction (HRXRD), and room temperature PL measurements.

III. RESULTS AND DISCUSSION

In order to decrease the $x_{2\text{D}}$ of the 2D InGaAs layer surrounding the QRs, we recall that this layer is made from GaAs/InAs SL.^{2,7,9} The $x_{2\text{D}}$ can be roughly estimated by

^{a)}Present address: School of Electronic and Electrical Engineering, The University of Leeds, Woodhouse Lane, Leeds LS2 9JT, UK. Electronic mail: l.h.li@leeds.ac.uk.

FIG. 1. (Color online) Dependence of the x_{2D} on the GaAs thickness, for different InAs thicknesses. The data (circles) in the figure are experimentally deduced from HRXRD measurements.

$d_{\text{InAs}}/(d_{\text{InAs}}+d_{\text{GaAs}})$, where d_{InAs} and d_{GaAs} are the InAs and GaAs thicknesses in the SL, respectively. By using this relationship, the expected dependence of the x_{2D} on the GaAs thickness is plotted as lines in Fig. 1 for different InAs thicknesses. The x_{2D} strongly depends on the GaAs and InAs thicknesses. Decreasing the InAs thickness or increasing the GaAs thickness decreases the x_{2D} . However, varying only one thickness in a wide range is impractical, as the InAs and GaAs thicknesses required for high-quality QRs are strongly related. This is based on the fact that the critical thickness of a subsequent InAs layer grown on an embedded InAs QD layer strongly depends on GaAs thickness.^{11,12} Due to the weakened effects of the strain field created by the seed QD layer,¹³ the critical thickness of the InAs layer increases as the GaAs thickness increases. As mentioned, for high-quality QR growth, alternative appearance of streaky diffraction rods and chevrons should be observed during the growth.^{3,7,8} To satisfy such conditions, the InAs thickness for a given GaAs thickness must be appropriately chosen. We determined the optimized InAs thicknesses by growing a large number of the QR samples with $N=10$ SL periods and under optimized growth conditions.⁷ It was found that a small thickness deviation of about $\pm 8\%$ results in QR size dispersion or 2D growth of the SL. Figure 2 shows the optimized parameter space (area within error bars) for high-quality QR growth. The circles represent $(d_{\text{GaAs}}, d_{\text{InAs}})$ couples for which alternative appearance of streaky diffraction rods and chevrons can be observed during the growth. From these samples, well-defined and intense PL spectra were obtained (see, for example, b in inset). Points outside the shadow correspond to thickness combinations for which either no QR formation (triangle) was observed or large QR size dispersion (square) results. The PL spectra measured from these samples are shown in the inset (a and c). The inferior optical properties of the sample with larger InAs thickness (spectrum a) are attributed to the generation of dislocations due to plastic relaxation. The solid line in the figure is a guide to the eyes. Therefore, the x_{2D} decrease can only be realized by simultaneously changing both the InAs and the GaAs thicknesses. As the optimum InAs thickness increases sublinearly with GaAs thickness, we chose to decrease the $d_{\text{InAs}}/(d_{\text{InAs}}+d_{\text{GaAs}})$ by increasing the GaAs thickness, while adjusting the InAs thickness to remain in the optimized parameter win-

FIG. 2. (Color online) Optimized $(d_{\text{GaAs}}, d_{\text{InAs}})$ parameter space for QR growth. The area within the error bars represents $(d_{\text{GaAs}}, d_{\text{InAs}})$ couples for which alternative appearance of streaky diffraction rods and chevrons can be observed during the growth. From these samples, well-defined PL spectra can be obtained (experimental points are indicated as circles, PL spectrum b in inset is an example). The line is a guide to the eyes. Below the shadowed region, no QR formation (triangle) is obtained, as confirmed by RHEED. Above the shadowed region, QR size dispersion (square) results. The PL spectra measured from these samples are shown in inset (a and c).

dow shown in Fig. 2. The function of increasing the GaAs thickness is twofold. It can help not only to decrease the x_{2D} but also to increase the maximum QR length, due to the lower average strain in the system. However, for large GaAs thicknesses, a short-range composition modulation related to the cycled deposition mode might appear. The best example is the growth of closely stacked InAs QDs, where distinct InAs islands are observed^{13–15} and the electronic wave functions become localized in the islands. In order to decrease the x_{2D} and simultaneously keep uniform QRs and delocalized wave functions, an optimum thickness combination of GaAs/InAs SL has to be found.

Figure 3 shows HRXRD curves, recorded near the (004) GaAs, for a set of samples grown with the optimized $(d_{\text{GaAs}}, d_{\text{InAs}})$ combinations shown as circles in Fig. 2. The peaks on the high-angle side ($\sim 33^\circ$) correspond to the diffraction from the GaAs substrate and the peaks on the low-angle side ($\sim 32.2^\circ$) correspond to the composite diffraction from the QRs and the InGaAs layer.⁷ With increasing GaAs thickness, the low-angle peak systematically shifts to higher

FIG. 3. (Color online) HRXRD curves recorded near the (004) GaAs from the samples with different GaAs/InAs SLs ($N=10$).

FIG. 4. (Color online) Normalized room temperature PL spectra from the samples with different GaAs/InAs SLs. Inset: dependences of the PL peak wavelengths of the QRs and InGaAs layer on $d_{\text{GaAs}}/d_{\text{InAs}}$ ratio. The calculated data (hollow circles) were obtained by solving the Schrödinger equation for a square potential well, where the In composition and InGaAs layer thickness deduced experimentally from the HRXRD measurements were used.

angles, indicating the decrease in the x_{2D} . The In composition has been estimated by using dynamical XRD theory and corresponds well to the simple estimation based on the InAs and GaAs thicknesses in the SL (see Fig. 1). The slightly lower In composition deduced experimentally is probably due to In atom migration from the InAs layer to the QRs.¹⁶

The decrease in the x_{2D} was also confirmed by PL measurements. Figure 4 shows normalized PL spectra of the abovementioned samples. The dependences of the PL peak wavelengths of the QRs and the 2D InGaAs layer on $d_{\text{GaAs}}/d_{\text{InAs}}$ ratio are summarized in the inset. With increasing GaAs thickness, the short-wavelength PL peak related with the InGaAs layer blueshifts, indicating a reduced In composition,⁷ as expected (see the calculated data). In contrast, the PL peak related with the QRs first redshifts and then blueshifts with increasing GaAs thickness. The redshift may result from increased x_{QR} and/or increased QR length.⁹ According to theoretical calculations, any of the cases will be highly beneficial for achieving TM-polarized gain.¹⁰ The blueshift for the largest GaAs spacing values may originate from the localization of the wave functions in a fraction of the QR length due to short-range composition modulation.^{9,15} The strongest PL emission and the longest PL peak wavelength were observed from the sample with GaAs (6 ML)/InAs (0.95 ML) SL. Such a thickness combination yields $x_{2D} \approx 12\%$, which is $\sim 3\%$ lower than that of the sample with GaAs (3 ML)/InAs (0.62 ML) SL. Increasing GaAs thickness up to 12 ML can even bring the x_{2D} down to about 9%. However, the inferior optical properties and possible In composition modulation across the center of the QRs make it less attractive.

To validate the feasibility of obtaining QRs with an increased GaAs thickness, we focused on the growth of the QR samples with GaAs (6 ML)/InAs (0.95 ML) SL. A set of samples with different number of periods were grown. Fig-

FIG. 5. (Color online) Room temperature PL spectra from the samples with different number of periods in the SL. Inset: $g=(002)$ dark-field cross-sectional TEM image taken from the QR sample with a number of periods of 30.

ure 5 shows the PL spectra from these samples. As expected, a PL redshift is observed for increasing period number, due to wave function delocalization.^{7,9} For $N > 40$, a strong decrease in the PL intensity occurs, which is attributed to plastic relaxation as confirmed by HRXRD measurements. It was found that a period number around 30 maximizes the PL radiative efficiency. In order to confirm the QR formation, TEM measurement on the sample with number of periods of 30 was performed. The inset shows the $g=(002)$ dark-field cross-sectional TEM image. The QR formation is clearly evidenced. However, a slight In composition modulation across the center of the QRs is observed due to the increased GaAs thickness. The in-plane diameter of the QR is about 5–7 nm and the height of the QR is about 70 nm. These lead to an aspect ratio of up to 10, far larger than those of the conventional InAs QDs (about 0.5) and of the QRs with GaAs (3 ML)/InAs (0.62 ML) SL (about 4.1).⁹ Actually, the structures represent InGaAs quasiquantum wires embedded in a GaAs matrix. The estimated x_{2D} and x_{QR} by TEM measurement¹⁷ are about 12% and 40%, respectively, confirming the results from the HRXRD and PL measurements.

IV. CONCLUSION

In conclusion, we reported MBE growth of the QRs with improved structural properties on GaAs. The GaAs thickness in the GaAs/InAs SL used for QR formation plays a key role in controlling the In compositional contrast between the QRs and the 2D InGaAs layer. Increasing the GaAs thickness results in not only an increased In compositional contrast but also an increased QR length, thus favoring the TM gain. QRs with a length of about 70 nm and an aspect ratio of up to 10 were obtained. In fact, quasiquantum wires have been formed. Such nanostructure is promising for controlling gain polarization and is expected to open new opportunities for novel devices.

ACKNOWLEDGMENTS

We are grateful to P. Ridha (EPFL, Switzerland), E. O'Reilly (Tyndall National Institute, Ireland), J. Andrzejewski, and G. Sęk (WUT, Poland) for useful discussions. We acknowledge financial support from the Swiss Commission for Technology and Innovation (CTI-TOPNANO21 program), the OFES (COST program), EU-FP6 project ZODIAC (Contract No. FP6/017140), and the Swiss National Science Foundation.

- ¹M. Sugawara, H. Ebe, N. Hatori, M. Ishida, Y. Arakawa, T. Akiyama, K. Otsubo, and Y. Nakata, *Phys. Rev. B* **69**, 235332 (2004).
- ²T. Kita, O. Wada, H. Ebe, Y. Nakata, and M. Sugawara *Jpn. J. Appl. Phys., Part 2* **41**, L1143 (2002).
- ³J. He, R. Notzel, P. Offermans, P. M. Koenraad, Q. Gong, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, *Appl. Phys. Lett.* **85**, 2771 (2004).
- ⁴T. Saito, T. Nakaoka, T. Kakitsuka, Y. Yoshikuni, and Y. Arakawa, *Physica E* **26**, 217 (2005).
- ⁵T. Kita, N. Tamura, O. Wada, M. Sugawara, Y. Nakata, H. Ebe, and Y. Arakawa, *Appl. Phys. Lett.* **88**, 211106 (2006).
- ⁶P. Ridha, L. H. Li, A. Fiore, G. Patriarche, M. Mexis, and P. M. Smowton, *Appl. Phys. Lett.* **91**, 191123 (2007).
- ⁷L. H. Li, G. Patriarche, M. Rossetti, and A. Fiore, *J. Appl. Phys.* **102**,

- 033502 (2007).
- ⁸J. He, H. J. Krenner, C. Pryor, J. P. Zhang, Y. Wu, D. G. Allen, C. M. Morris, M. S. Sherwin, and P. M. Petroff, *Nano Lett.* **7**, 802 (2007).
- ⁹L. H. Li, P. Ridha, G. Patriarche, N. Chauvin, and A. Fiore, *Appl. Phys. Lett.* **92**, 121102 (2008).
- ¹⁰J. Andrzejewski, G. Sęk, J. Misiewicz, and E. P. O'Reilly, personal communication (2007).
- ¹¹P. B. Joyce, T. J. Krzyzewski, P. H. Steans, G. R. Bell, J. H. Neave, and T. S. Jones, *J. Cryst. Growth* **244**, 39 (2002).
- ¹²D. M. Schaadt, S. Krauss, R. Koch, and K. H. Ploog *Appl. Phys. A: Mater. Sci. Process.* **83**, 267 (2006).
- ¹³Q. Xie, A. Madhukar, P. Chen, and N. P. Kobayashi, *Phys. Rev. Lett.* **75**, 2542 (1995).
- ¹⁴H. Ishikawa, H. Shoji, Y. Nakata, K. Mukai, M. Sugawara, M. Egawa, N. Otsuka, Y. Sugiyama, T. Futatsugi, and N. Yokoyama, *J. Vac. Sci. Technol. A* **16**, 794 (1998).
- ¹⁵N. N. Ledentsov, V. A. Shchukin, M. Grundmann, N. Kirstaedter, J. Bohrer, O. Schmidt, D. Bimberg, V. M. Ustinov, A. Y. Egorov, A. E. Zhukov, P. S. Kopev, S. V. Zaitsev, N. Y. Gordeev, Z. I. Alferov, A. I. Borovkov, A. O. Kosogov, S. S. Ruvimov, P. Werner, U. Gosele, and J. Heydenreich, *Phys. Rev. B* **54**, 8743 (1996).
- ¹⁶P. B. Joyce, T. J. Krzyzewski, G. R. Bell, T. S. Jones, S. Malik, D. Childs, and R. Murray, *Phys. Rev. B* **62**, 10891 (2000).
- ¹⁷G. Patriarche, L. Largeau, J. C. Harmand, and D. Gollub, *Appl. Phys. Lett.* **84**, 203 (2004).