Digital alloy interface grading of an InAlAs/InGaAs quantum cascade laser structure studied by cross-sectional scanning tunneling microscopy

Citation for published version (APA):
Offermans, P., Koenraad, P. M., Wolter, J. H., Beck, M., Aellen, T., & Faist, J. (2003). Digital alloy interface grading of an InAlAs/InGaAs quantum cascade laser structure studied by cross-sectional scanning tunneling microscopy. *Applied Physics Letters*, *83*(20), 4131-4133. https://doi.org/10.1063/1.1627942

DOI:
10.1063/1.1627942

Document status and date:
Published: 01/01/2003

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: 16. Nov. 2023

APPLIED PHYSICS LETTERS VOLUME 83, NUMBER 20 17 NOVEMBER 2003

Digital alloy interface grading of an InAlAs/InGaAs quantum cascade laser structure studied by cross-sectional scanning tunneling microscopy

P. Offermans,[a] P. M. Koenraad, and J. H. Wolter
Department of Semiconductor Physics, Eindhoven University of Technology, P.O. Box 513, NL-5600MB Eindhoven, The Netherlands

M. Beck, T. Aellen, and J. Faist
Institute of Physics, University of Neuchâtel, 1 Rue A.-L. Breguet, Neuchâtel, CH 2000 Switzerland

(Received 21 July 2003; accepted 26 September 2003)

We have studied an InGaAs/InAlAs quantum cascade laser structure with cross-sectional scanning tunneling microscopy. In the quantum cascade laser structure digital alloy grading was used to soften the barriers of the active region. We show that due to alloy fluctuations, softening of the barriers occurs even without the digital grading. © *2003 American Institute of Physics.*
[DOI: 10.1063/1.1627942]

Unlike quantum well semiconductor lasers where the light originates from recombination of electrons and holes across the energy gap that exists between the conduction band and valence band of the crystal, in quantum cascade laser (QCL) structures, the wavelength is essentially determined by quantum confinement, i.e., by the thicknesses of the layers of the active region rather than by the band gap of the material. The injection and extraction efficiency to and from the active regions has been significantly improved by means of wave function engineering, which involves control over the barrier/well thickness and interface quality. Interface roughness scattering is one of the important nonradiative scattering mechanisms of QCL structures. To reduce interface roughness scattering, digital alloy grading was used to soften the barriers of the active region of a four-quantum-well (4QW) midinfrared quantum cascade laser structure. This structure showed a reduced threshold current density and a narrower luminescence linewidth in pulsed mode than a device with abrupt interfaces.[1] However, this result could not be reproduced in a second growth run. We report cross-sectional scanning-tunneling microscopy (X-STM) measurements of 4QW QCL structures with and without graded interfaces. The aim of this letter is to show that interface roughness and grading occur due to fluctuations in the alloy concentration which are caused by indium segregation during growth.

The QCL structures were grown by molecular beam epitaxy on InP substrates using lattice-matched InAlAs/InGaAs active layers and an InP top cladding. Except for the interfaces, both types of devices are identical and based on a 4QW active region designed for a vertical lasing transition at 9.3 mm with a double phonon resonance.[2] The layer sequence of the graded structure is as follows: 31/**19**/30/**23**/29/**25**/26/**1**/2/**40**/5/**4**/6/**4**/8/(**3**/52/**1**/2)/(**2**/1/**3**/1/**2**)/(2/**1**/51/**1**/2)/(**2**/1/**3**/1/**2**)/(2/**1**/44/**1**/2)/(**2**/1/**19**)/34/**14**/33/**13**/32/**15** Å. The structure with the abrupt interfaces has the following layer sequence: 31/**19**/30/**23**/29/**25**/29/**40**/19/**7**/58/**9**/57/**9**/50/**22**/34/**14**/33/**13**/32/**15** Å. InGaAs wells are in roman, and InAlAs barriers in bold. Doped layers (Si, 3×10^{17} cm^{-3}) are underlined. The main differences between both structures are the design of injection barrier and the barrier/well interfaces of the active region which are graded by a digital InGaAs/InAlAs alloy of 1- and 2-Å-thick layers as indicated by parentheses.

STM measurements have been performed in an UHV chamber with base pressure $<2\times10^{-11}$ Torr on the UHV-cleaved (110) cross-sectional surface. Tips are prepared by electrochemical etching of polycrystalline tungsten wires and are treated in the vacuum with a self-sputtering technique.[3]

In Fig. 1 we show a STM image of the QCL structure with graded interfaces. Figure 1 was taken at a negative sample bias of -1.6 V, showing the filled states associated with the As sublattice. The bright regions are the InGaAs well layers and the dark regions are the InAlAs barrier layers. The observed constrast between the barrier and the well layers is due to the band offset between the well and the barrier layers which causes an electronic contribution to the tunnel current at low sample bias ($V_{\text{sample}}<-2$ V). The

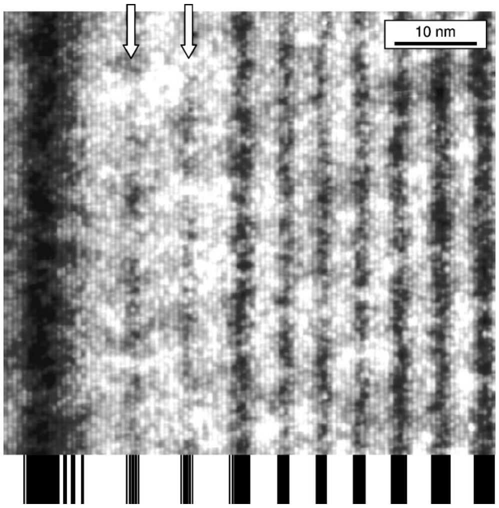

FIG. 1. 60×60 nm^2 filled states STM image of the graded structure with layer sequence indicated at the bottom of the image, $V_{\text{sample}}=-1.6$ V. The black layers are the InAlAs barriers and the white layers are the InGaAs wells.

---

[a] Electronic mail: p.offermans@tue.nl

0003-6951/2003/83(20)/4131/3/$20.00

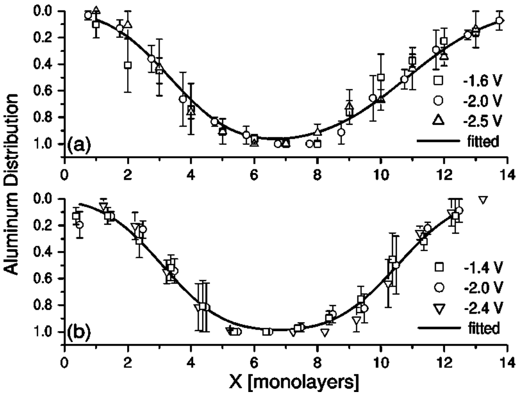

FIG. 2. Aluminum concentration of a 2.5 nm barrier of the structure with (a) graded interfaces and (b) abrupt interfaces. The aluminum concentration was derived from averaged line profiles taken at three sample voltages.

graded interfaces are indicated by arrows. The brighter and darker regions within and across the layers indicate the strong presence of alloy fluctuations which are caused, as we will show, by indium segregation. It is clear that the alloy fluctuations are causing strong interface roughness, which will influence the actual effect of the intentional grading of the interfaces.

In order to quantify the sharpness of the ungraded interfaces of both structures, we derive the distribution of the gallium and aluminum atoms across both interfaces of a 2.5 nm wide barrier by using averaged line profiles of the filled states image. Since we are imaging the filled states of the sample surface, we are not directly probing the aluminum and gallium atoms themselves but rather the electronic effects these two atomic species have on the surface arsenic

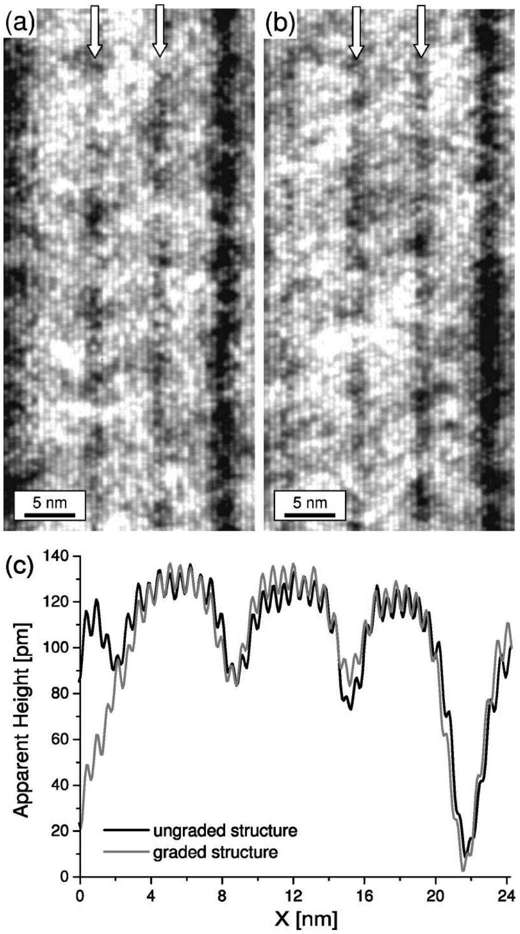

FIG. 3. 25×55 nm^2 filled state STM image of the active region of (a) the structure with graded interfaces and (b) the structure with abrupt interfaces. (c) shows the averaged line profiles of the active region.

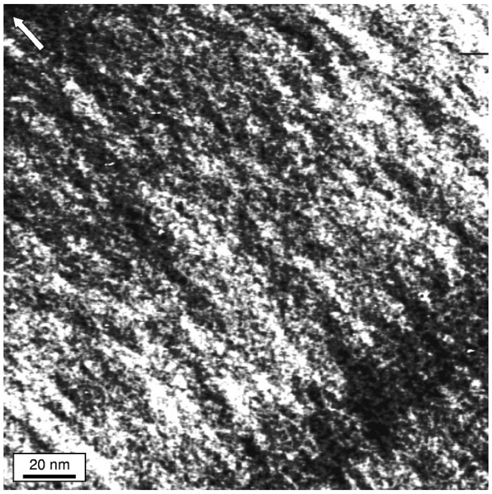

FIG. 4. 185×185 nm^2 empty states image showing the outward relaxation of inidum rich regions. In the bottom right corner, part of the active layers can be seen. The growth direction is indicated by the arrow.

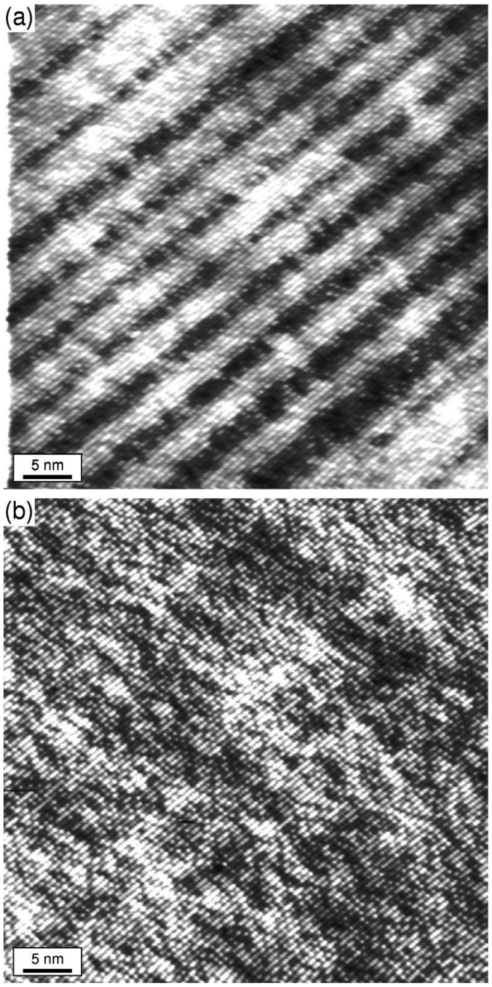

FIG. 5. 50×50 nm^2 filled states (a) and empty states (b) image of the injector region. In both images brighter and darker regions correspond to indium rich and indium poor regions.

atoms. The apparent height of an arsenic site should be dependent on the number of aluminum atoms in the nearest neighbor positions at the cleaved (110) surface.[4] There are three nearest neighbor positions where an Al atom can reside. Two neighbor positions are in the surface layer and one neighbor position is in the subsurface layer. In our analysis of the apparent height profile, the average number of aluminum atoms of a single bilayer can be assigned to both the surface and subsurface layers, which will give a spread in the possible number of aluminum atoms in each monolayer. The final aluminum distribution is determined by fitting with a concentration profile in the form of: $c(x)=1/2\{\mathrm{erf}[(L/2+x)/2\sigma_x]+\mathrm{erf}[(L/2-x)/2\sigma_y]\}$, where L is the width of the barrier and σ_x and σ_y indicate the sharpness of the interfaces of the barrier. Figures 2(a) and 2(b) show for both QCL structures the aluminum distribution of the 2.5 nm wide InAlAs barrier with ungraded interfaces. The analysis was done for profiles taken at sample voltages of -1.6, -2.0, and -2.4 V. Although the observed contrast in the STM image depends on the applied voltage, the counted aluminum distribution does not. We find that for both structures the full width at half maximum is in agreement with the growth menu, however, in both cases the interfaces have a gradient of 4 ML. We propose that this grading is caused by large scale alloy fluctations due to indium segregation during growth. This means that any additional digital grading of the interfaces of the active region will not have much effect.

In Figs. 3(a) and 3(b) we show detailed images the active region of the graded structure and the structure with the abrupt interfaces. The barriers of the active region are indicated by arrows. The injection barriers of both structures are clearly different, however, the thin barriers in the active region look the same. This can also be seen in Fig. 3(c) which shows the averaged lineprofiles of the active region of both structures.

Figure 4 shows the InGaAs region which was grown on top of the active layers. In the lower right corner part of the active layers can be seen. The image was taken at high positive voltage $(+2.5$ V) where electronic contrast is minimized.[5,6] The topographical contrast is due to the outward relaxation of indium rich regions. The lateral scale variation of alloy fluctations is about 10 nm, which is large enough to influence the device characteristics.[7]

Figures 5(a) and 5(b) show images of exactly the same active region at low negative voltage (filled states) and high positive voltage (empty states), respectively. The filled states image clearly shows the electronic contrast between the barriers and wells. The empty states image shows the topographical contrast due to outward relaxation of indium rich regions. In both the barriers and the wells of the filled states image brighter and darker regions can be seen, which correspond to the indium rich and indium poor regions of the empty states image. This shows to our surprise that the indium clusters extend across the barrier and wells, which results, on average, in graded interfaces.

We have compared QCL structures with and without digitally alloyed interfaces on the atomic level with X-STM. We showed that due to indium segregation across the barrier and well layers, fluctuations in the width of the layers occur, which on average lead to graded interfaces of about 4 ML. We find that, because of the unintentional grading, the digital alloy graded interfaces are not significantly smoother than the ungraded interfaces. We propose that in order to reduce interface roughness scattering, fluctuations in the layer thicknesses due to indium segregation should be taken into account.

[1] T. Aellen, M. Beck, D. Hofstetter, J. Faist, U. Oesterle, M. Ilegems, E. Gini, and H. Melchior, *Proc. of the 29th International Symposium on Compound Semiconductors*, 7–10 October 2002, Lausanne, Switzerland.

[2] M. Beck, D. Hofstetter, T. Aellen, J. Faist, U. Oesterle, M. Ilegems, E. Gini, and H. Melchior, Science **295**, 301 (2002).

[3] G. J. de Raad, P. M. Koenraad, and J. H. Wolter, J. Vac. Sci. Technol. B **17**, 1946 (1999).

[4] A. R. Smith, K.-J. Chao, C. K. Shih, Y. C. Shih, K. A. Anselm, and B. G. Streetman, J. Vac. Sci. Technol. B **13**, 1824 (1995).

[5] D. M. Bruls, P. M. Koenraad, M. Hopkinson, J. H. Wolter, and H. W. M. Salemink, Appl. Surf. Sci. **190**, 258 (2002).

[6] R. Feenstra, Physica B **273**, 796 (1999).

[7] J. H. Marsh, Appl. Phys. Lett. **41**, 732 (1982).