Towards quantitative three-dimensional characterisation of buried InAs quantum dots

Kadkhodazadeh, Shima; Semenova, Elizaveta; Schubert, Martin; Thuvander, M.; Stiller, K. M.; Yvind, Kresten; Dunin-Borkowski, Rafal E.

Published in:
Journal of Physics: Conference Series

Link to article, DOI:
10.1088/1742-6596/326/1/012046

Publication date:
2011

Citation (APA):
Kadkhodazadeh, S., Semenova, E., Schubert, M., Thuvander, M., Stiller, K. M., Yvind, K., & Dunin-Borkowski, R. E. (2011). Towards quantitative three-dimensional characterisation of buried InAs quantum dots. Journal of Physics: Conference Series, 326(1), 012046. DOI: 10.1088/1742-6596/326/1/012046

Towards quantitative three-dimensional characterisation of buried InAs quantum dots

S. Kadkhodazadeh[1,2], E. S. Semenova[2], M. Schubert[2], M. Thuvander[3], K. M. Stiller[3], K. Yvind[2], R. E. Dunin-Borkowski[1]

[1]Center for Electron Nanoscopy, Technical University of Denmark, DK-2800, Kgs. Lyngby, Denmark

[2]Department of Photonic Engineering, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark

[3]Chalmers University of Technology, Applied Physics, 412 96 Gothenburg, Sweden

shka@cen.dtu.dk

Abstract. InAs quantum dots grown on InP or InGaAsP are used for optical communication applications operating in the 1.3 – 1.55 µm wavelength range. It is generally understood that the optical properties of such dots are highly dependent on their structural and chemical profiles. However, morphological and compositional measurements of quantum dots using transmission electron microscopy can be ambiguous because the recorded signal is usually a projection through the thickness of the specimen. Here, we discuss the application of scanning transmission electron microscopy tomography to the morphological and chemical characterisation of surface and buried quantum dots. We highlight some of the challenges involved and introduce a new specimen preparation method for creating needle-shaped specimens that each contain multiple dots and are suitable for both scanning transmission electron microscopy tomography and atom probe tomography.

1. Introduction

Self-assembled III-V semiconductor quantum dots (QDs) are of interest for applications that include optical amplifiers, temperature-stable low-noise lasers and mode-locked lasers [*e.g.* 1-3]. Important electronic properties such as density of states and internal carrier dynamics, which determine the optical characteristics of the QDs, are largely affected by the morphologies and chemical compositions of both the QDs and the material surrounding them. A thorough understanding of these properties is therefore essential for successful applications. The majority of structural characterisation carried out on InAs QDs has involved the study of either uncapped QDs using atomic force microscopy (AFM), or projections through the thickness of specimens in transmission electron microscopy (TEM). However, the spatial resolution of AFM is often insufficient to accurately determine the exact morphologies of QDs. Furthermore, most device applications require QDs to be overgrown with a material that has a different bandgap. This step, known as capping, can introduce significant changes to the structure and chemistry of the QDs. While conventional TEM techniques can be used to study capped QDs in plan-view or cross-sectional geometries, the resulting images can give ambiguous information about their three-dimensional properties. Here, we discuss the applicability of high angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) tomography to the study of InAs/InGaAsP QDs. We discuss the challenges of three-dimensional characterisation of buried QDs and present a new method of preparing specimens for combined experiments using both electron tomography and atom probe tomography (APT).

2. Results and discussion

InAs/InGaAsP samples comprising 1.5 ML of InAs deposited on (001) $In_{0.85}Ga_{0.15}As_{0.35}P_{0.65}$ (lattice matched to InP) were grown by metal organic vapour phase epitaxy (MOVPE). Buried QDs were overgrown with 100 nm of InGaAsP. Plan-view and cross-sectional TEM specimens were prepared using mechanical polishing, dimpling and argon ion milling and examined at 300 kV using an FEI Titan TEM equipped with a field emission electron source and a spherical aberration corrector on the condenser lens system. Inner and outer annular dark-field detector collection semi-angles of 48 and 240 mrad, respectively, were used with an electron probe convergence semi-angle of 17.5 mrad. Alignment and reconstruction of tomographic data was carried out using the simultaneous iterative reconstruction technique (SIRT) algorithm in Inspect3D software (typically 15 iterations were used). Visualisation of reconstructions was performed using Aviso v.6 software.

2.1. Morphological characterisation using electron tomography

HAADF STEM tomography was carried out on plan-view specimens to study the morphologies of surface QDs. Figure 1(a) shows a HAADF STEM image of a representative surface QD. Consistently, elongated hexagonal shapes were observed for the bases of the surface QDs. The elongation direction was determined to be [110] using selected area electron diffraction, high-resolution HAADF STEM and AFM. The HAADF STEM images also suggest a double-terraced geometry for the surface QDs, with steeper facets around their bases and shallower facets towards their tops. This observed geometry is consistent with a theoretical model of InAs QDs on an InGaAs substrate lattice matched to InP [4] shown in figure 1(b). A tomographic reconstruction of a QD produced using a series of 67 images obtained at 2° intervals over a tilt range of ± 66° is displayed in figure 1(c). The elongated hexagonal base is visible in the three-dimensional reconstruction.

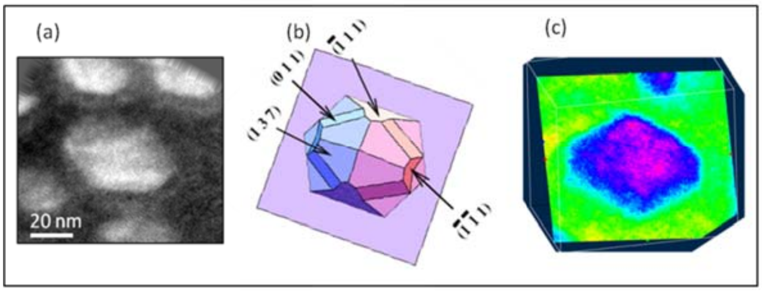

Figure 1: (a) HAADF STEM image of a surface InAs QD on an InGaAsP substrate that is lattice matched to InP. (b) Theoretical energetically favourable shape of InAs QDs on an InGaAs substrate lattice matched to InP, taken from [4]. (c) Tomographic reconstruction of a surface QD from 67 HAADF STEM images taken at 2° intervals over the tilt range ± 66° using the SIRT algorithm.

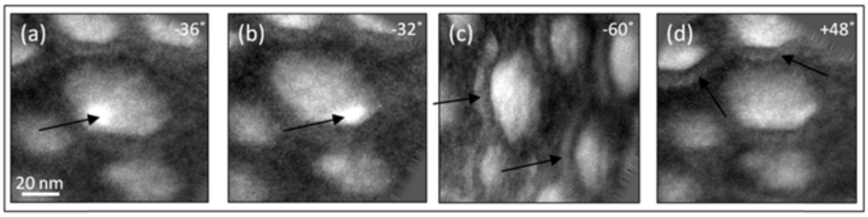

Figure 2: HAADF STEM images of a surface InAs/InGaAsP QD at different tilt angles, showing different diffraction effects appearing at different tilt angles.

Despite the large inner detector angle used, interesting diffraction effects appear in the tilt series of HAADF STEM images. Figures 2(a) and (b) show regions of localised bright contrast that change rapidly with specimen tilt and may be associated with surface strain. Furthermore, at large tilt angles bright bands appear around the bases of QDs, as shown in figures 2(c) and (d). These bands may also be associated with diffraction effects. These features are interesting in their own right. However, they are inconsistent with the projection requirement, which states that the recorded intensity should be a monotonic function of a property of the object [5], and may therefore lead to artefacts in the tomographic reconstruction.

HAADF STEM tomography of capped QDs was attempted in both cross-sectional and plan-view specimen geometries. In both cases, a limitation was found to be the available tilt range before the specimen became too thick for imaging. In the case of cross-sectional specimens, it is also possible that only a section of a QD may be contained within the specimen. The tilt axis chosen for tomography of cross-sectional specimens was found to be particularly important, as the use of tilt axes other than [001] (the growth direction) resulted in the signal from the wetting layer becoming very weak after only a few degrees of tilt. Figure 3 shows an attempt at three-dimensional reconstruction of a capped InAs/InGaAsP QD reconstructed from 37 HAADF STEM images acquired at 2° intervals over a tilt range of only ± 36°. Unfortunately, the images became too blurred and noisy beyond this tilt range. Elongation due to the large missing wedge in Fourier space resulted in artefacts in the reconstruction, which make complete morphological characterisation impossible. Moreover, the contrast arising from strong electron channelling at low order zone-axis orientations results in significant departures from the projection requirement for tomography. HAADF STEM tomography analysis of a buried InAs/InGaAsP prepared in plan-view geometry is shown in figure 4. A series of 61 images acquired at 2° intervals over a ± 60° tilt range was used for reconstruction. The QD appears to have an elongated shape along the <110> direction. Although evidence of faceting of the QD is present in both the original HAADF STEM images and the final reconstruction, the facets are not as clearly visible as those of the surface QD in figure 1. This difference may result from chemical intermixing between the QD and the capping material during overgrowth, as well as from artefacts in the tomographic reconstruction.

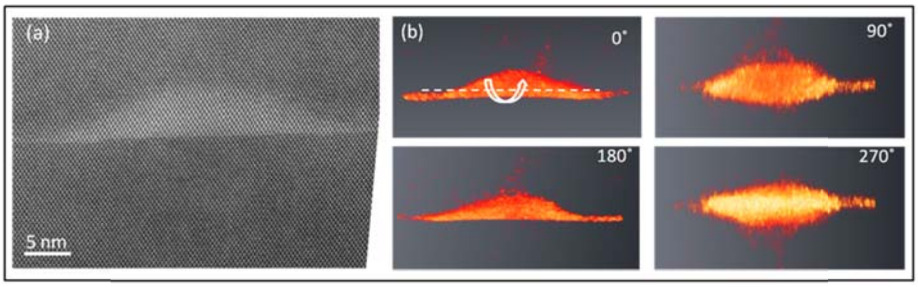

Figure 3: (a) HAADF STEM image of a buried InAs/InGaAsP QD viewed along the [110] direction in cross-sectional geometry. (b) Reconstructed image of the same QD from 37 images acquired at 2° intervals over a ± 36° tilt range. The reconstruction was carried out using the SIRT algorithm (15 iterations). The arrow indicates the direction of rotation.

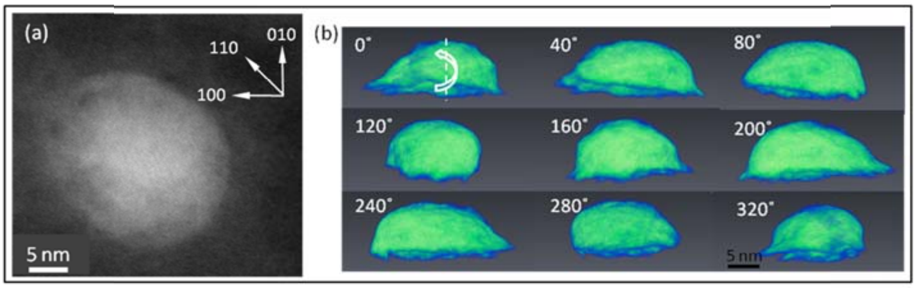

Figure 4: (a) HAADF STEM image of a buried InAs/InGaAsP QD in plan-view geometry viewed along the [001] orientation. (b) Tomographic reconstruction of the QD from 61 images acquired at 2° intervals over a ± 60° tilt range using the SIRT algorithm (15 iterations). The arrow indicates the direction of rotation.

2.2. Fabrication of needle-shaped specimens

The specimen thickness limitation in electron tomography can, in principle, be overcome by fabricating needle-shaped specimens using focused ion beam (FIB) milling, to allow tilting by up to 180° without significant increase in projected specimen thickness. However, the preparation of a needle-shaped specimen containing a single QD is highly challenging, as most QDs are too small to be visible during FIB preparation. Moreover, the FIB can introduce considerable damage in III-V semiconductors, including amorphisation and gallium ion implantation [6]. We have fabricated needle-shaped specimens that are 100 nm in diameter, using reactive ion etching, selective wet etching and critical point drying in plan-view geometry (figure 5). The choice of a plan-view geometry for the needles means that each specimen contains several QDs. The needles can be detached from the substrate by cleaving (see figure 5(b)) or lifted out and mounted onto suitable grids using a micro-manipulator in the FIB with minimal additional damage (figure 5(c)). Besides electron tomography, these needle-shaped specimens can be used for APT, for which needle-shaped specimens with sharp tips (narrower than 100 nm) are required. Our ongoing experiments involve the application of both HAADF STEM tomography and APT to the same QDs in order to better understand their morphologies and compositions. A comparison between the reconstructions obtained using these two techniques will also assist in the evaluation and mitigation of potential artefacts in each case.

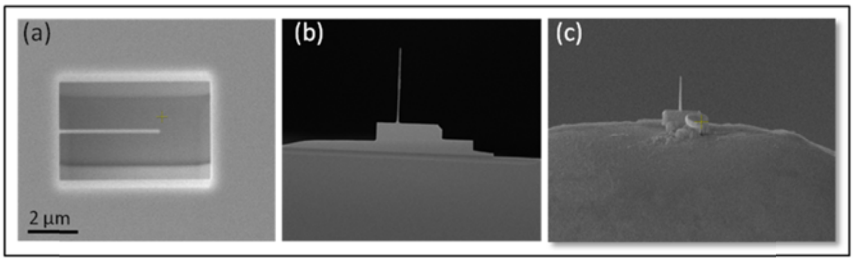

Figure 5: (a) Scanning electron microscopy images of needle-shaped specimens fabricated using reactive ion etching, selective wet etching and critical point drying. The needle specimens can be (b) detached from substrates by cleaving or (c) lifted out and mounted onto a suitable grid.

3. Conclusions

HAADF STEM tomography of surface and buried InAs/InGaAsP QDs has been presented and some of the challenges of the application this technique for the morphological and chemical characterisation of buried QDs have been discussed. A new method of specimen preparation suitable for both electron tomography and APT of buried QDs has been introduced.

References

[1] Akiyama T and Arakawa Y 2007 *Proc. IEEE* **95** 1757

[2] Sugwara M and Usami M 2009 *Nat. Photonics* **3** 30

[3] Thompson M G, Rae A R, Xia M, Penty R V and White I H 2009 *IEEE J. Sel. Top. Quantum Electron.* **15** 661

[4] Kratzer P, Chakrabarti A, Liu K K Q, Scheffler M 2009 *New J. Phys.***11** 73018

[5] Hawkes P W 1992 *The electron microscope as a structure projector: three-dimensional imaging with the transmission electron microscope* (New York: Plenum Press) pp 17-38

[6] Rubanov S, Munroe P R 2005 *Microsc. Microanal.* **11** 446