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Quantitative investigation of the onset of islanding in strained layer epitaxy of InAs/GaAs by X-ray mapping in STEM

T Walther and M Hopkinson
Department of Electronic & Electrical Engineering, University of Sheffield,
Sir Frederick Mappin Building, Mappin Street, Sheffield S1 3JD, UK
E-mail: t.walther@sheffield.ac.uk

Abstract. The Stranski-Krastanow transition from two- to three-dimensional growth describes the onset of roughening and the formation of islands. For InAs/GaAs, islanding occurs abruptly for a deposition of about one unit cell of InAs. We combine scanning transmission electron microscopy and X-ray elemental mapping to measure the layer thicknesses and chemical compositions of thin layers of InAs deposited on GaAs(001) for nominal thicknesses of 1.6, 1.8 and 2.0 monolayers of InAs. Different methods to analyse ratios of such elemental maps have been compared to measure quantitatively the indium content within these ultra-thin layers. The most accurate and reliable of the three methods investigated is shown to be the analysis of the inverse slope of plots of As/In ratio maps versus scan window size perpendicular to the layers.

1. Introduction
The Stranski-Krastanov transition describes the sudden onset of islanding during the growth of a previously perfectly flat film. For the InAs/GaAs(001) system this has been reported from surface analytical techniques to occur at a deposition of 1.8 monolayers of indium, i.e. just before the completion of a single unit cell of InAs [1, 2]. We investigate this transition by X-ray mapping combined with scanning transmission electron microscopy (STEM), with the aim to quantify the amount of indium in layers grown at nominal InAs thicknesses just around that value and to correlate this directly with the onset of islanding observed.

2. Experimental
Three layers of InAs of nominal thicknesses of 1.6, 1.8 and 2.0 monolayers indium (defined as 1ML=d_{002}=a/2=0.283nm for GaAs) were deposited upon GaAs(001) with 70nm GaAs barrier layers. The samples were grown by molecular beam epitaxy at 505ºC, using elemental gallium, indium and arsenic sources. The InAs growth rate was set as 0.10 ML/s, a value derived from calibrations based on indium induced reflection high-energy electron diffraction (RHEED) oscillations on InAs substrates, taking into account the differences in lattice constant. The nominal thicknesses are subject to errors of ± 0.1ML due to the nature of the RHEED measurement and the possibility of growth rate drift. A <100> cross-sectional sample was prepared using standard cutting, grinding, polishing and Ar⁺ ion milling techniques and studied in a JEOL2010F field-emission transmission electron microscope operated at 197kV in STEM mode and equipped with a Gatan Imaging Filter, an Oxford Instruments Si:Li X-ray detector with ultrathin window and the Oxford Instruments ISIS300 software for quantification. Figs. 1 and 2 depict bright-field and annular dark-field images recorded with the
specimen oriented edge-on, close to a <100> zone axis. 128×100 pixel X-ray maps at a sampling of 2.2nm/pixel were recorded for all X-ray lines (K and L) of all elements (In, Ga and As) using signal integration from successive scans during a long (~2 hrs) time where drift was corrected to 1-2 pixels precision. The sample was tilted 19º towards the detector, with a 4º in-plane component that blurred the layers in projection to apparent widths of ~6nm (3 pixels). The sample thickness was estimated to be ~80nm. The maps are shown in Figure 3.

Figure 1. Edge-on bright-field (BF) STEM image of InAs/GaAs layer structure. Growth towards the top right corner.

Figure 2. Annular dark-field (ADF) STEM image of the same area. Quantum dots are only visible in the top of the three InAs layers.

Figure 3. (a) ADF image and X-ray elemental maps of (b) C_K (26-184 counts), (c) background of S_K (0-29 counts), (d) As_L (30-205 counts), (e) As_K (29-240 counts), (f) In_Lα (1-33 counts), (g) In_Lβ (0-29 counts), (h) In_Kα (0-20 counts), (i) Ga_L (50-336 counts) and (j) Ga_K (32-246 counts).

We then compared three different techniques of evaluating the indium content of the layers quantitatively, namely by measuring (and for 1. and 2., integrating the signals along growth direction):
1. the relative decrease of gallium concentration from line profiles of the Ga/As ratio (Fig. 4);
2. the relative increase of indium concentration from line profiles of the In/(Ga+In) ratio (Fig. 5); and
3. the inverse slope of plots of the As/In ratio as a function of the length L of the scan window perpendicular to the layers (Fig. 6), as indicated schematically in Figs. 3e and 3h for K-lines.

The last method is equivalent to the concept of concentric electron probes in TEM, originally introduced for X-ray analysis of atoms segregated to planar faults studied by convergent beam
illumination [3] and later extended to STEM illumination [4]. Its principle is that the number of matrix atoms (here: As) within an irradiated volume is proportional to the size of the area illuminated times the specimen thickness, while the number of segregated atoms (here: In) in the irradiated volume is proportional to the lateral extent of the illumination times the specimen thickness. Hence, the ratio of the first to the second should be proportional to the extension of the illuminated area and independent of the specimen thickness. If beam broadening were negligible, the slope would be inversely related to the effective chemical thickness of the planar fault or thin layer analysed. With beam broadening taken into account, deviations from linearity will occur which can be quantified by linear regression analysis and thus give a measure of the expected accuracy [3, 4]. Using Monte Carlo simulations of electron scattering and X-ray generation this method called conceptEM was previously shown to be more reliable and more accurate for the measurement of the chemistry of thin layers than standard profiling methods [5], given the same illumination conditions. In this experimental study, the biggest problem was to find appropriate $k$-factors because the ISIS300 software provides $k$-factors only for specific X-ray lines within individual spectra but not for X-ray maps generated from integrating spectral intensity over windows of certain widths that will depend on the individual energies of X-ray lines chosen. As a compromise, for the corresponding windows we determined self-consistent effective $k$-factors based on the assumption that the stoichiometry of the group III/V ratio is unity (i.e. 50at% of the InGaAs material is comprised of As atoms) and that In replaces Ga in InGaAs (i.e. the sum of In and Ga atoms must also account for 50at%). This implies that maps of the As signals (group-V sub-lattice) should be of the same height as the sum of maps ($k_{\text{In,As}} + k_{\text{Ga,As}}$) for the group-III sub-lattice.

Figure 4. Relative decrease of gallium signal, measured by profiles of the Ga$_K$/As$_K$ ratio (above) and the Ga$_L$/As$_L$ ratio (below) vs. distance along [001] in nm. The max. decrease of the Ga signal amounts to ~4, 7 and 9% of the Ga signal in GaAs (i.e. to be multiplied by 50at% for occupancy of group III sub-lattice).

Figure 5. Relative increase of the indium signal as measured by profiles of the ratio In$_K$/[In$_K$+$k_{\text{GaK,InK}}$ Ga$_K$] (above) or of the ratio In$_L$/[In$_L$+$k_{\text{GaL,InL}}$ Ga$_L$] (below) vs. distance in nm. The peak increase of In amounts to ~3, 4 and 5% of the group III sub-lattice.

Figures 4-6 show results from the three quantification methods, and Table 1 compares the numerical results obtained for the total amount of indium in the layers. It can be seen that the first method based on the relative decrease of the Ga signal is simple but suffers from the largest statistical errors (due to
the noisiness of the line scans in Figure 4) and the results from K-line quantification are systematically larger (by 0.38±0.12ML) than those from L-line quantification. The second method measuring the increase in the In signal from the profiles yields consistent values for K- and L-line quantification (the difference of −0.04±0.11ML is within statistics) but all values are still much smaller than the nominal values expected from the calibrated deposition rates. The third method quantifying the slopes of the plots of the As/In ratio vs. scan window length in Figure 6 yields generally larger indium concentration values closer to the nominal ones and smaller statistical error bars due to linear regression analysis. The data for K- and L-line quantification are, however, only consistent within these small error bars of ~±0.1ML for the measurement of the 3rd layer, while the values obtained for the 1st and 2nd layer from K- and L-lines disagree by more than this statistical error, which we attribute to these regions of the specimen being somewhat further from the specimen edge, hence a little thicker so that the softer X-rays are more strongly absorbed. This trend can, however, only be noticed when the error bars are sufficiently small. It should be noted that the correlation coefficients of the linear fitting were 0.994±0.005 for the K-lines and 0.998±0.001 for the L-lines, which from previous simulations [3] would indicate relative errors of a few %, in agreement with our observations.

![Figure 6. Plot of k-factor corrected As/In ratio as function of window size around In(Ga)As layers.](image)

### Table 1. List of amount of indium in the three layers measured by the various techniques.

| method      | lines | 1st layer | 2nd layer | 3rd layer |
|-------------|-------|-----------|-----------|-----------|
| RHEED       | -     | 1.6±0.1   | 1.8±0.1   | 2.0±0.1   |
| decrease of $k_{Ga,As}/Ga$ | K     | ±0.3      | ±0.3      | ±0.3      |
|             | L     | 0.47      | 0.71      | 1.01      |
| increase of $In/(In+k_{Ga,As})$ | K     | ±0.25     | ±0.25     | ±0.27     |
|             | L     | 1.25      | 1.39      | 1.63      |
| inverse slope of $k_{As,In}/As/In$ | K     | ±0.13     | ±0.11     | ±0.13     |
|             | L     | 1.46      | 1.48      | 1.76      |

Values in the table are given in equivalents of full InAs monolayers.

### 3. Summary

The most accurate and reliable method to quantify the indium content of InGaAs thin layers from X-ray microanalysis is to perform linear regression analysis to plots of As/In ratio measurements as a function of area size analysed around these layers. In thin regions quantification from K- and L-lines can yield consistent values to better than ±0.1ML.

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