Progress Towards Reliable EELS Quantification of GaSb-GaAs Heterostructures

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Compound III-V semiconductors have received much attention in recent years for photonic applications. One such III-V semiconductor system of current interest is GaSb in heterostructures with GaAs and other III-V compound semiconductors: this has potential applications in infrared detectors [1]. Epitaxial GaSb films were grown on single crystal GaAs substrates using Molecular Beam Epitaxy (MBE). Details of the growth are described elsewhere [2].

Samples were prepared for electron microscopy by a conventional cross-section method using sandwiching, mechanical polishing, and Ar ion beam thinning in a Gatan PIPS down to 500V for final polishing. Scanning transmission electron microscopy (STEM) was carried out using a probe-corrected JEOL ARM200F STEM equipped with a cold field emission gun operated at 200 kV, equipped with a Gatan GIF Quantum ER spectrometer. Spectrum images were acquired in DualEELS mode with a 300 V offset between the low loss and high loss regions and with a probe convergence semiangle of 29 mrad and a spectrometer acceptance semiangle of 36 mrad.

Figure 1a is a high-resolution annular dark field STEM image of the GaSb-GaAs interface, showing high quality single crystal GaSb film; detailed examination of such images shows periodic misfit dislocations at the interface. EELS spectrum images were then recorded across this interface and were then quantified by different methods to determine the elemental profile across it.

Figure 1b shows the chemical composition calculated using the Gatan Digital Micrograph EELS model-based Elemental Quantification plugin. This was performed using power law background subtraction before the Ga and As L₂,₃ edges and the Sb M₄,5 edge, together with the provided Hartree-Slater cross-sections for each element within the energy window chosen for quantification. This shows an apparent change in stoichiometry across the interface from 50:50 in the GaAs to 55:45 in the GaSb. This is not reasonable considering the high crystal quality of the material. It is more likely due to two problems with Sb. Firstly, Hartree-Slater cross sections for M-edges are probably overestimated [3]. Secondly, there are large background perturbations from EXELFS before this edge, suggesting that background subtraction will be problematic.

Figure 1c shows the result of simply renormalizing these results by assuming that Ga:As and Ga:Sb must be 50:50 away from the interface. This still leaves a bump in Gallium concentration at the interface, and it is unclear if this is real.

Figure 1d shows the result of an entirely manual calculation of elemental composition. This was done by a method of background subtraction, Fourier-ratio deconvolution to remove the effects of plural scattering, normalisation by zero loss peak intensity, and edge integration (using a 140 eV window), similar to recent work [4]. An effective cross section for Sb was calculated assuming a 50:50 concentration away from the interface. Additionally, the As concentration quantification was improved by using the GaSb area to determine the shape of the Ga L-edge and its extended structure where no As
is present, and then scaling this shape by the intensity in the 140 eV edge integration window and subtracting this shape from the whole spectrum using scripts developed previously [4]. This then left a pure As shape in the GaAs, without any disturbance from the Ga L₁ edge immediately before it, which especially improves the quantification when As is at low concentrations. The combined effect of correct Sb quantification, improved background subtraction for As and normalisation by the zero loss intensity is the graph of 1d, which shows that the apparent bump in Ga concentration has totally disappeared, and was an artefact in the previous quantification methodologies. It is therefore clear that quantitative nanoscale analysis of III-V heterostructures with EELS in the STEM requires careful determination of experimental cross sections and background subtraction in order to produce reliable results.

References:

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Figure 1. a) High resolution scanning transmission electron micrograph (HR-STEM) of the GaSb-GaAs interface. EELS quantification of GaSb-GaAs interface using b) Gatan Digital Micrograph model-based quantification plug-ins, c) similar model-based quantification but holding GaSb stoichiometrically balanced. And d) EELS quantification of GaSb-GaAs interface holding GaSb stoichiometrically balanced and correcting for variations in Zero Loss intensity at the interface.