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EELS analysis of $\text{In}_x\text{Ga}_{1-x}\text{As}_y\text{Sb}_{1-y}$ nanostructures

AM Beltrán^{1,*}, AM Sánchez², F de la Peña³, T Ben¹, DL Sales¹, AG Taboada⁴, JM Ripalda⁴, M. Varela⁵, SJ Pennycook⁵ and SI Molina¹

1. Departamento de Ciencia de los Materiales e I.M. y Q.I., Facultad de Ciencias, Universidad de Cádiz, Campus Río San Pedro, s/n, 11510 Puerto Real, Cádiz, Spain
2. Physics Department, University of Warwick, Coventry CV4 7AL, U.K.
3. Department of Materials Science, University of Cambridge, Pembroke Street, CB2 3QZ, Cambridge, U.K.
4. Instituto de Microelectrónica de Madrid (CNM-CSIC) Isaac Newton 8, 28760 Tres Cantos, Madrid, Spain
5. Materials Science and Technology Division, Oak Ridge National Laboratory, 37831 Oak Ridge, Tennessee, USA

*Present address: CNRS, LAAS, 7 avenue du colonel Roche, F-31400 Toulouse, France- Univ de Toulouse, LAAS, F-31400 Toulouse, France and CEMES-CNRS, 29 Rue Jeanne Marvig, BP 94347 31055 Toulouse Cedex 4, France

beltran@cemes.fr

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Semiconductor nanostructures, such as quantum dots (QDs) are widely studied for a number of opto-electronic applications. Several approaches have been studied to achieve long wavelength emission values. Among them, GaSb incorporation to InAs/GaAs QDs is considered an important advance for improving the opto-electronic properties due to staggered band alignment of the GaSb/InAs systems [1]. In spite of the reported miscibility gap for quaternary alloys of GaSb and InAs in the absence of strain, the formation of ternary and quaternary alloys from the intermixing of the binary compounds is very likely when strain is present.

Since opto-electronic properties depend on the structural quality and compositional distribution, the characterization of the heterostructures is important in order to establish a relationship between real composition and structure and the achieved properties. The characterization techniques available in the electron microscope are very well suited to help to relating the opto-electronic properties of the QDs to their structure at the atomic level. In effect, the size, distribution, crystalline quality and composition of the nanostructures can be studied by high angle annular dark field imaging (HAADF), electron diffraction and electron energy loss spectroscopy (EELS), respectively.

Although different approaches to introduce Sb in InAs/GaAs QDs heterostructures have been analysed with the aim to identify the key parameters that enhance the opto-electronic properties of the nanostructures, this work is focused on the configuration which results in the best photoluminescence (PL) results.

All the heterostructures have been grown by molecular beam epitaxy (MBE), following similar conditions (details about growth conditions can be found in [2]). Over a GaAs (001) substrate, InAs has been grown to form a wetting layer (WL). The formed QDs were capped by a GaSb layer (sample S0), a GaAs layer of different thickness previous to the GaSb layer (S3, S6 and S12, of 3, 6 and 12 monolayers, ML, of thickness, respectively) or irradiated by a Sb flux before the deposition of 3 ML of GaAs and 2.2 ML GaSb (sample S). All of them were finally capped by a GaAs layer. EEL spectra were acquired at the SuperSTEM laboratory (UK) and ORNL (USA). For the compositional analysis, core loss and/or low EELS is frequently used. The studied systems are difficult to analyse conventionally in the core loss region because of the overlapping of the edges. For this purpose, *Hyperspy* software has been used over these edges. It is based on a combination of principal components analysis and blind source separations methods [3, 4].

Focusing on the sample which shows the better PL emission, sample S [2], concerning the structural characterization, a lower defect density is observed for this sample, and the height of the QDs is slightly larger than the other samples [5, 6]. Hence the size of the QDs and the crystalline quality contribute to improve the PL intensity value.

Compositional analysis by EELS in combination with the *Hyperspy* software of sample S, shows that In is mainly located inside the QD, although it is also detected outside due to the well-known

segregation phenomenon very common in InAs-GaAs systems. Whereas, Sb is also observed inside QD but seems to cover the core of the QDs (Figure 1). There is no evidence of the GaAs and GaSb layers which has been nominally grown over the QDs. Instead a continuous composition is observed as a consequence of the intermixing phenomena between elements involving GaAs-GaSb and InAs, as well as the Sb from the flux. Ternaries and quaternary alloys are formed from these elements. Ga and As are detected in all the analysed area, since they are mainly in the substrate and top-capped layer and they are also inside the QD due to the segregation process.

Due to the projection effects we do not exactly know if we have analysed a completed QD or just a portion but from the results we are able to estimate the concentration and confirm the presence of Sb inside the QD in very low proportion.

Comparing to the other studied approaches, in the case of sample S0, Sb has been also detected inside the QDs, although in a higher concentration [7]. Samples S3, S6 and S12 were grown with the aim to avoid the formation of the quaternary alloy by the growth of an intermediary GaAs layer. Nevertheless, EELS analyses have also revealed the formation of ternary and quaternary alloys as in the previous samples. We observe a different behaviour depending on the thickness of the GaAs intermediate layer. Up to 6 ML two InGaAs intermediate layer while for 12 ML of GaAs layer, all In is segregated and only the formation of a quaternary alloy $\text{In}_x\text{Ga}_{1-x}\text{As}_y\text{Sb}_{1-y}$ is observed. In both cases; the Sb content is lower than 5% [8].

In conclusion, Sb irradiation of InAs QDs grown by MBE on GaAs and further GaSb/GaAs capping results in a huge increase of the room temperature PL signal at about 1.3 μm , thanks to the good crystalline quality of the heterostructure and the compositional distribution even with the formation of an $\text{In}_x\text{Ga}_{1-x}\text{As}_y\text{Sb}_{1-y}$ alloy inside the QD.

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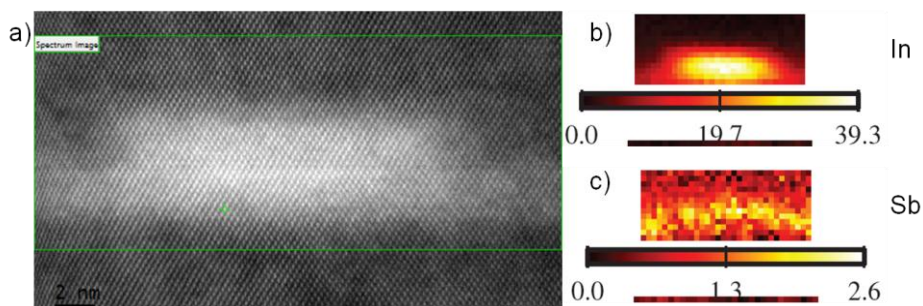


Figure 1. a) HAADF image of sample S and b) compositional map of elemental distribution obtained for In and Sb (C).