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Structural and Chemical Assessment of InAs/AlGaAs quantum Dot Structures for Enlarged Bandgap Intermediate Band Solar Cells

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An Intermediate Band (IB) solar cell (SC) consists of an IB material situated between the n- and p-type regions of a host semiconductor. The IB material has a band of states in the energy gap (E_g) of the host semiconductor, which allow an electron–hole pair generation by means of the absorption of two photons of energy lower than E_g. For this kind of SC, Luque and Martì [1] demonstrated a limiting efficiency of 63.2%, in comparison to the Shockley–Queisser limit of 40.7% [2] for a conventional single-gap solar cell under the same operating conditions.

Epitaxial quantum dots (QDs), such as InAs QDs embedded in the intrinsic region of a GaAs p-i-n single junction structure, have been proposed as IB material [3]. One of the major issues arising from the use of InAs QDs as intermediate band material in GaAs based IB solar cell is the decrease of the open circuit voltage, V_{OC}, with respect to the reference cell without QDs [4]. A suitable nanoscale engineering of the semiconductor band structure can be made by a proper choice of the materials to be used as intermediate band and host semiconductors, in alternative to the mostly used InAs/GaAs; this can result in a partial or complete recovery of the V_{OC}. InAs/AlGaAs QDs system represents an interesting alternative to the most common InAs/GaAs system, since the higher energy gap of the host barrier can better confine the IB QDs states; therefore an improved efficiency is expected as theoretically predicted for E_g values as large as 1.95 eV [1]. Nevertheless, the different mobility of In adatoms on Al-containing surfaces may result in a rough interface between AlGaAs and InAs, which may negatively affects the growth of uniform and pseudomorphic QDs.

In order to overcome this problem, a peculiar multistep growth approach was developed and followed in the present work for the formation of the QDs layer, which consisted in a gradual composition evolution from a quaternary (AlInGaAs) to ternary (InGaAs) and finally binary (InAs) compound. This growth procedure, together with the intrinsic mechanism of QDs formation, leads to a complex chemical and structural profile, which strongly affects the electro-optical properties of the system. A detailed scanning transmission electron microscopy (STEM) study was carried out in order to assess the structural and chemical properties of the QDs layers. In particular, high resolution STEM images and energy dispersive X-ray point spectra, scan profiles and maps allowed to define the compositional and strain distribution within these complex nanostructures and to correlate these results with the electro-optical performances of the related IBSC.

In order to optimize the growth process, a preliminary set of samples was grown by changing the chemistry of the surrounding matrix, from GaAs to AlGaAs, whose effect on the morphology of the dots, grown according the multistep process described above, was investigated. These samples consisted of a single QD layer, buried 50 nm below the sample surface, and of an uncovered QD layer, grown on the sample surface. Information about the QD surface reorganization deriving from the capping procedure with different barrier materials was also inferred from the analyses of these samples. In figure

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1, the High Angle Annular Dark Fields (HAADF) (a, c) and Bright Field (BF) (b, d) images of QDs embedded respectively in GaAs matrix (represented by a 5 nm thick layer grown before and after the QDs layer) (figure 1a and 1b) and AlGaAs matrix (figure 1c and 1d). Well-developed QDs are observed in the latter case, as also confirmed by the intense and narrow PhotoLuminescence (PL) peak obtained from this sample. On the basis of these results, three QDs layers were stacked and integrated in an IBSC device, according to the schemes reported in Figure 1. Figure 1f represents the low magnification BF STEM image of the dot stack: the micrograph evidences the formation of coherent islands laterally connected by a 2 nm thin wetting layer and having no vertical correlation; the image demonstrates the overall good structural quality of the sample. The high resolution HAADF image in figure 1e shows the morphology of a single dot of the vertical stack. No sharp chemical interfaces, resulting from the multistep growth process, are evident from the image contrast. The contrast profile, obtained from the rectangular region of the imaged dot and having an integration with of 7 nm, shows a bottommost interface more abrupt than the topmost one; this is consistent with the species mobility and temperature gradients applied during the growth. Compositional analyses were carried out by Energy Dispersive x-ray spectroscopy (EDS) to derive information about the Al content in the barrier layers and eventual chemical gradients inside and dots. Since, in the case of the dot, the EDS spectra integrate the signals coming from the quantum dot volume and from the surrounding AlGaAs matrix, above and below the QD in the TEM lamella, a proper calculation scheme was developed in order to deconvolute the signal coming from the dot from the one coming from the barrier. In segregation inside the dot as well as strong In depletion and Al enrichment in the WL nearby the dot were observed.

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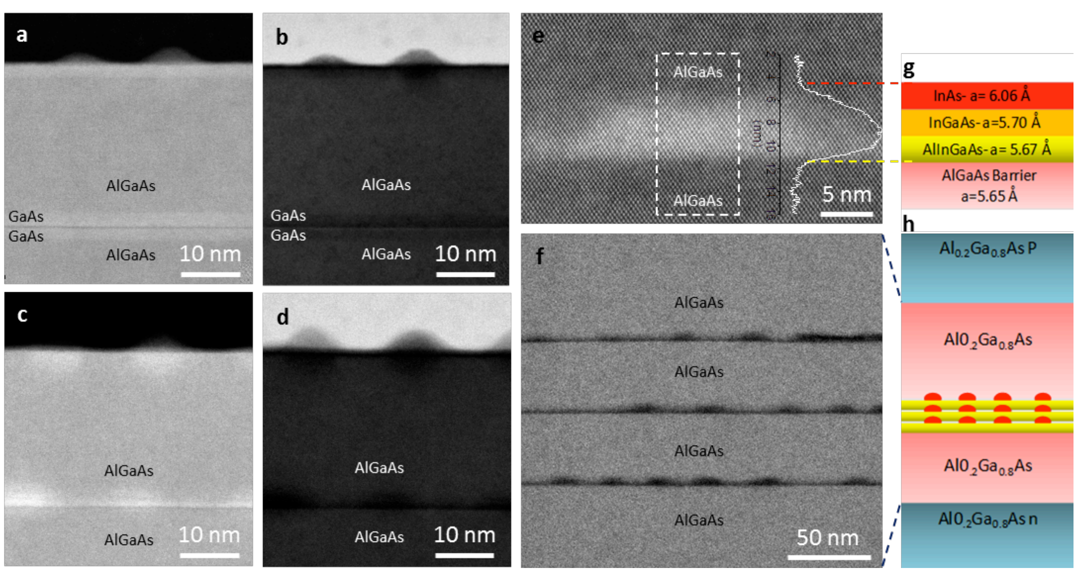

Figure 1. STEM images of (a-d) single InAs QDs layers embedded in different barrier materials and (e,f) stacked QDs grown according to the scheme in (g, h) and integrated in a IBSC.

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