Solar Energy Materials & Solar Cells 144 (2016) 128–135

Contents lists available at ScienceDirect

Solar Energy Materials & Solar Cells

journal homepage: www.elsevier.com/locate/solmat

Impact of alloyed capping layers on the performance of InAs quantum dot solar cells

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ARTICLE INFO

Article history:
Received 6 March 2015
Received in revised form
28 July 2015
Accepted 11 August 2015

Keywords:
Solar cells
Quantum dots
Quantum wells
Type-II
Carrier transport
Electron trapping

ABSTRACT

The impact of using thin GaAs(Sb)(N) capping layers (CLs) on InAs/GaAs quantum dots (QDs) is investigated for their application in solar cell devices. We demonstrate the ability to combine strain-balancing techniques with band engineering approaches through the application of such CLs. Extended photoresponse is attainable by means of an independent tunability of the electron and hole confinements in the QD. Moreover, the CL acts itself as a quantum well (QW), providing an additional photoresponse, so that the devices work as hybrid QD–QW solar cells. The use of a GaAsSb CL is particularly beneficial, providing devices with efficiencies under AM1.5 conditions 20% higher than standard GaAs-capped QDs. This is mainly due to a significant increase in photocurrent beyond the GaAs bandgap, leading to an enhanced short-circuit current density (J_{sc}). The addition of N to the CLs, however, produces a strong reduction in J_{sc}. This is found to be related to carrier collection problems, namely, hindered electron extraction and retrapping in the CLs. Nevertheless, the application of reverse biases induces a release of the trapped carriers assisted by a sequential tunneling mechanism. In the case of GaAsN CLs, this leads to a complete carrier collection and reveals an even higher QD–QW-related photocurrent than when using a GaAsSb CL. The hindered carrier collection is stronger in the case of the quaternary CLs, likely due to the faster recombination rates in the type-I GaAsSbN/GaAs QW structure as compared to the type-II ternary counterparts. Nevertheless, alternative approaches, such as the use of a thinner CL or a short-period superlattice CL, lead to significant improvements, demonstrating a great potential for the quaternary CLs under a proper device design.

1. Introduction

Recently, quantum dots (QDs) have been considered as a feasible opportunity to exceed the Shockley–Queisser efficiency limit of single-gap materials [1]. On one side, electron–hole pair multiplication, predicted to provide higher conversion efficiency limits [2], is expected to become a competitive mechanism in zero-dimensional structures [3]. Indeed, efficiencies in QD-based solar cells exhibiting multiple-exciton generation have been theorized to undergo significant increases as compared to bulk material and quantum efficiencies higher than 1 have already been demonstrated [4,5]. On the other side, the use of QDs has also been proposed as a possibility to realize the intermediate-band solar cell concept [6]. Particularly, the use of the well-known InAs/GaAs system has been the focus of increasing efforts to take advantage of these approaches and implement QD-based enhanced solar cells. The introduction of absorption levels by the QDs leads to an extended photoresponse beyond the GaAs band edge, allowing the achievement of higher short-circuit currents [7–10]. Further control of the absorption QD energy levels can be obtained through the use of a modified thin capping layer (CL) on top of the QDs, which could allow a precise control and extension of the absorption energy. Moreover, the CL could also act itself as a quantum well (QW), providing an additional photoresponse at energies below the GaAs band edge. However, although improved solar cell characteristics could be expected, this approach has been all but unexplored in this field [9,11].

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http://dx.doi.org/10.1016/j.solmat.2015.08.009
0927-0248/

Particularly, the application of a thin GaAsSbN CL could be of great interest on this point, allowing engineering the band structure as well as the strain of the QDs. On one side, the beneficial impact of thin GaAsSb CLs on InAs QDs properties is already well known [12], where the presence of Sb extends the QD ground state wavelength by reducing the valence band offset [13]. The use of such a CL has been found to significantly enhance the efficiency of both light emitting devices [14,15] and photodetectors [16], while improved solar cell characteristics have been also recently reported in stacked-QD devices [9]. However, a limitation may be found regarding the number of layers that can be stacked due to strain accumulation, since GaAsSb CLs increase the overall compressive strain in the stack. On the other side, GaAsN CLs have also been found to improve the InAs QD luminescence for low N contents [17,18]. With this CL, the emission wavelength is extended because of the QD–CL conduction band offset reduction induced by the presence of N [19]. Furthermore, the presence of N reduces the accumulated strain, allowing the stacking of a large number of QD layers [20]. Thus, the combination of both elements, Sb and N, could take advantage of both approaches. The use of GaAs(Sb)(N) CLs would allow an individual control of carrier escape times through the possibility to tune both the electron and hole confinements [21]. In addition, a suitable ratio of Sb to N in the CL would compensate the accumulated strain, allowing a higher number of stacked QD layers before relaxation via the origination of extended defects. This would also be expected to mitigate the loss in open-circuit voltage (V_{oc}) induced by the presence of InAs QDs, as found through different strain-balancing techniques [22–24].

In this work we perform a detailed comparative study concerning the impact of thin GaAs(Sb)(N) CLs on the overall performance of InAs/GaAs QD-based p–i–n junction solar cells. We will show how the presence of a CL plays a major role in the photoresponse, providing an additional contribution to the photocurrent (PC). However, a problem arises regarding carrier collection, intrinsically related to the introduction of N, so that N-containing CLs act as potential wells deep enough to hinder carrier transport and extraction and degrade the final characteristics of the device. This drawback is systematically addressed, from both experimental and theoretical points of view, and new possible approaches to circumvent this undesired effect are proposed.

2. Experimental details

All of the p–i–n junction solar cells under study were grown by solid-source molecular beam epitaxy on n^+-GaAs (001) substrates under As_4-stabilized conditions. Due to the comparative nature of this study, a simple device structure was chosen so no window layer or antireflection coating was used (parameters such as the doping levels or the emitter and intrinsic region thicknesses are not either optimized). A schematic diagram of the structure is shown in Fig. 1. A 500 nm-thick n-GaAs base buffer was grown at 580 °C, while the emitter consists of 350 nm-thick p-GaAs followed by a 150 nm-thick p^+-GaAs. The active region always consists of 10 QD layers embedded in the middle of 400 nm-thick intrinsic GaAs. QDs were grown by depositing 2.8 monolayers (MLs) of InAs at 450 °C and 0.04 ML s^{-1} resulting in a density, estimated through atomic force microscopy scans of similar uncapped QDs, near $3\cdot10^{10}$ cm^{-2}. The different CL materials, described below, were always deposited at 470 °C, immediately followed by a 2.5 nm-thick low-temperature (LT) GaAs layer to avoid Sb/N desorption. Subsequently, 30 nm-thick GaAs spacers were grown at high temperature (580 °C), found to be a critical factor for solar cell characteristics [25].

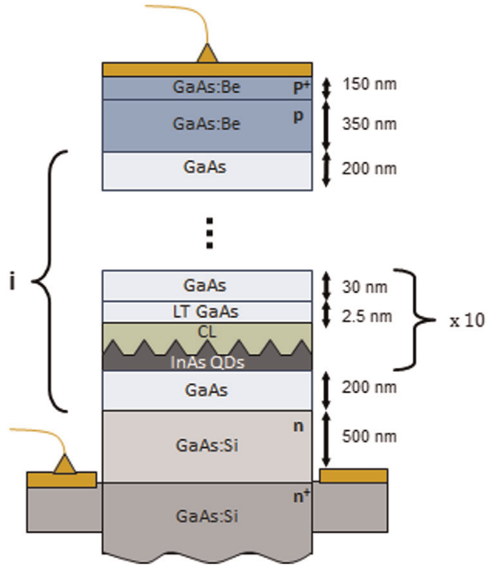

Fig. 1. Schematic sketch of the p–i–n devices containing 10 stacked InAs QD layers capped with a thin GaAs(Sb)(N) CL.

To get a detailed insight into the structural properties, conventional transmission electron microscopy (TEM) analyses were performed using a JEOL JEM-2100 LaB6 operating at 200 kV. The structural analysis was complemented with X-ray diffraction (XRD). Unlike TEM, the XRD is a rapid and a non-destructive technique which retrieves structural information averaged over extended sample area (>1 mm^2). The measurements were performed with Cu-$K\alpha1$ line ($\lambda_X=1.5406$ Å), in a commercial Panalytical X'Pert Pro diffractometer equipped with a Ge(220) hybrid monocromator.

Photoluminescence (PL) measurements were performed using a He–Ne laser as excitation source. The emitted light was collected, dispersed through a 1 m-spectrometer, and then detected with a nitrogen-cooled Ge detector.

The test devices consisted of 200 µm-diameter cylindrical mesas processed by standard fabrication techniques, with AuGe/Au and Au/AuZn/Au for the n- and p-type contacts, respectively. Photocurrent density–voltage (*J–V*) measurements were conducted using a Keithley 2400 sourcemeter. The light source is a solar simulator equipped with a 150 W Xenon Lamp and an AM 1.5D filter combination (Newport). The total light intensity was calibrated to 900 mW/cm^2 using a GaAs reference cell. PC and *I–V* measurements under monochromatic illumination were carried out using a Keithley 230 voltage source and a Keithley 617 electrometer, utilizing light from a quartz halogen lamp dispersed through a 0.34 m-monochromator.

3. Results and discussions

The comparative analysis was carried out using 5 nm-thick GaAsSbN, GaAsSb, and GaAsN CLs (samples S-SbN, S-Sb, and S-N, respectively). A 2 ML s^{-1} growth rate, particularly beneficial for N-containing samples [26], was used for the growth of the CL in all these samples. The Sb and N nominal contents were always set to 10% and 2.0%, respectively. In addition, a sample containing GaAs-capped QDs (sample S-0) and a control GaAs sample, with no QDs inside, were grown for comparison.