Fabrication of 100 Layer-Stacked InAs/GaNAs Strain-Compensated Quantum Dots on GaAs (001) for Application to Intermediate Band Solar Cell

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ABSTRACT

In order to demonstrate the predicted high efficiency operation of a quantum dot intermediate band solar cell (QD-IBSC), high-density QD superlattice with good size homogeneity is required. Though multiple stacking is one promising way to increase the total QD density thereby increasing the optical absorption by QD-IB, it is difficult to maintain the size homogeneity and structural quality of QD superlattice. For this, we take advantage of strain-compensation growth technique, in which the compressive strain induced by each InAs QD layer is compensated, or balanced out, by embedding it with a tensile-strained GaNAs strain-compensating layer. In this work, we demonstrate a high quality growth of up to 100 layer-stacked InAs/GaNAs QD superlattice on GaAs (001) substrate. We have also characterized some basic solar cell characteristics.

INTRODUCTION

Recently, an intermediate band solar cell comprised of an ideal 3-dimensional semiconductor quantum dot (QD) superlattice incorporated in the active region of a *p-i-n* structure has attracted intense research [1]. For realization of high efficiency QD intermediate band solar cells (QD-IBSCs), QDs with good size homogeneity and high density are required. In general, multiple stacking of self-assembled QDs is one powerful way to increase the total QD density. However, the epitaxial growth maintaining the size uniformity and good crystalline quality of QD superlattice becomes a practical challenge, in the case of InAs/GaAs system. This is evidenced by accumulation of internal lattice strain with increasing number of stacks, which eventually leads to generation of dislocations. For this, we take advantage of strain-compensation, or strain-balanced growth technique [2,3], in which the compressive strain induced by each In(Ga)As QD layer is compensated by embedding it with a tensile-strained GaNAs strain-compensating layer (SCL). In this work, we demonstrate a high quality growth of up to 100 layer-stacked InAs/GaNAs QD superlattice on GaAs (001) substrate. We have also characterized some basic solar cell characteristics for cells with 25, 50, and 100 layer-stacked QD absorption layers.

EXPERIMENTAL

All growths were done by atomic hydrogen-assisted molecular beam epitaxy (H-MBE) with a RF plasma as the nitrogen source. An As_2 source was used for arsenic because we found previously [4] that overall crystalline quality of GaNAs material is better if As_2 were used instead of a more commonly used As_4 source. An As_2 flux was generated by cracking the As_4 beam at 900°C by using a commercially available valved-cracker effusion cell. For fabrication of QDSCs, the surface oxide was first removed with atomic H cleaning. Following deposition of a 250 nm-thick n^+-GaAs buffer layer and a 1000 nm-thick *n*-GaAs base layer at 580°C, a 20 nm-thick $GaN_{0.01}As_{0.99}$ SCL and a 1.82 monolayers of InAs QD layer were consecutively grown in pair up to 25, 50, and 100 multiple cycles at 480°C. Then, a 500 nm-thick *p*-GaAs emitter layer, a 30 nm-thick p^+-$Al_{0.4}Ga_{0.6}As$ window layer, and a 50 nm-thick p^+-GaAs contact layer were grown to constitute a *p-i-n* solar cell as schematically shown in Fig.1. The arsenic (As_2), hydrogen, and nitrogen beam pressures

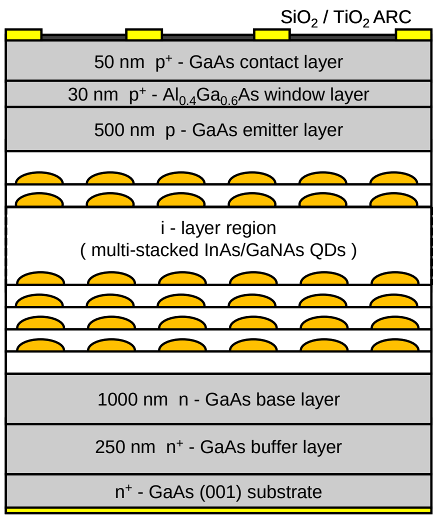

Figure 1 Schematic structure of multi-stacked InAs/GaNAs strain-compensated QDSCs fabricated on GaAs (001) substrate.

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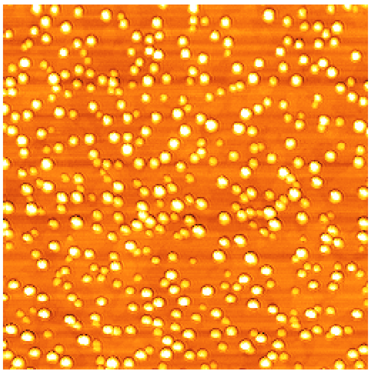

Figure 2 AFM image of the topmost surface for 100 layer-stacked InAs QDs with 20 nm-thick GaNAs SCL grown on GaAs(001). Scan size is 1μm × 1μm.

during growth were kept constant at 2.0×10^{-5}, 4.0×10^{-6}, 3.0×10^{-6} Torr, respectively.

For structural characterization of our samples, atomic force microscope (AFM), scanning transmission electron microscope (STEM), and reciprocal space mapping (RSM) around (224) reflection in high resolution X-ray diffraction (HR-XRD) were used. Further, photoluminescence (PL) measurements were performed using 532 nm diode-pumped solid-state laser at an output power of 1 W/cm^2 and an electronically cooled PbS photodetector. For solar cell characterization, Ti/Pt/Au alloy was used for the top contact and AuGeNi/Au for the bottom for 3×3 mm^2-sized solar cells, and SiO_2/TiO_2 anti-reflection coating (ARC) was used. The quantum efficiency (QE) measurements were done under a constant photon irradiation of 10^{16} /cm^2 under varying reverse bias in order to evaluate the effect of electric field on the carrier collection process.

RESULTS AND DISCDUSSION

Figure 2 shows the AFM image of the topmost QD surface after 100 layers of stacking. The sheet density, average height and diameter of the topmost QD layer are 4.2×10^{10} cm^{-2}, 5.4 nm, and 29.6 nm, respectively. The total QD density amounts to as high as 4.2×10^{12} cm^{-2}. Further, the size homogeneity in QD diameter is as good as 10.2%, and high-quality QDs without coalescence or defects are observed even after 100 layers of stacking as shown in the cross-sectional STEM images of Fig. 3.

Figure 4 shows the reciprocal space mapping (RSM) around (224) reflection in HR-XRD. Satellite peaks are clearly resolvable along the growth direction indicating a sharp repetition of multi-stacked QD structure. The 0th-order peak F_0 shows a near perfect lattice-match with the peak S for GaAs (001) substrate. Therefore, the average lattice constant of QD superlattice structure is nearly perfectly matched to GaAs, that is, the compressive strain in each QD layer is compensated by introducing a tensile strain by GaNAs SCL. In addition, the satellite peaks

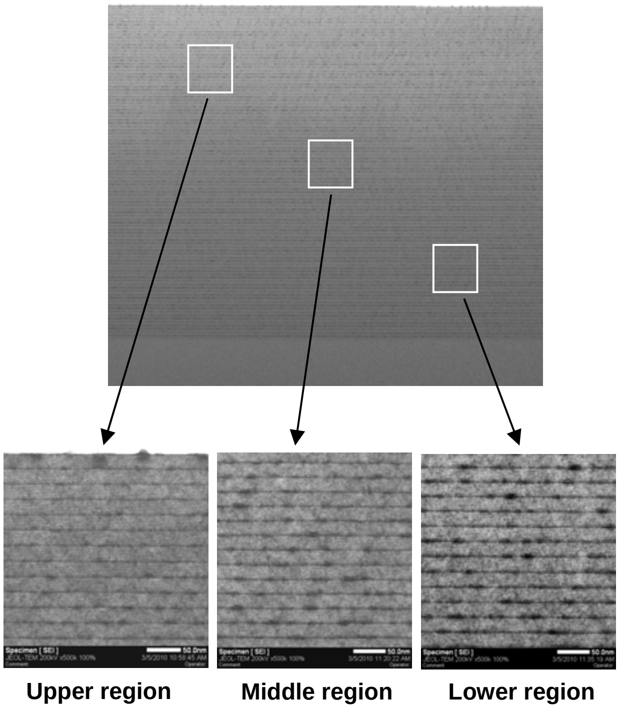

Figure 3 Cross-sectional STEM photographs for 100 layer-stacked InAs/GaNAs strain-compensated QD structure.

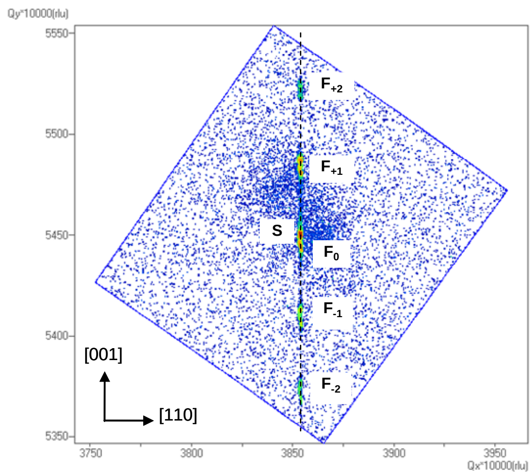

Figure 4 RSM of diffracted X-ray intensity around (224) reflection measured for 100 layer-stacked InAs/GaNAs QDs sample.

aligned along [001] direction suggests that lattice relaxation has not occurred and hence dislocations were not generated after stacking.

Figure 5 show the PL spectra measured at 77K for 50 and 100 layer-stacked QD samples. In both samples, the PL peaks are observed at around 1065 nm with a narrow

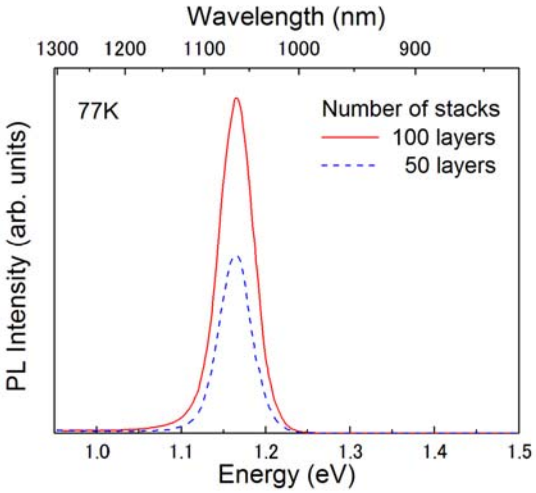

Figure 5 PL spectra measured at 77K for 50 and 100 layer-stacked InAs/GaNAs QDs sample.

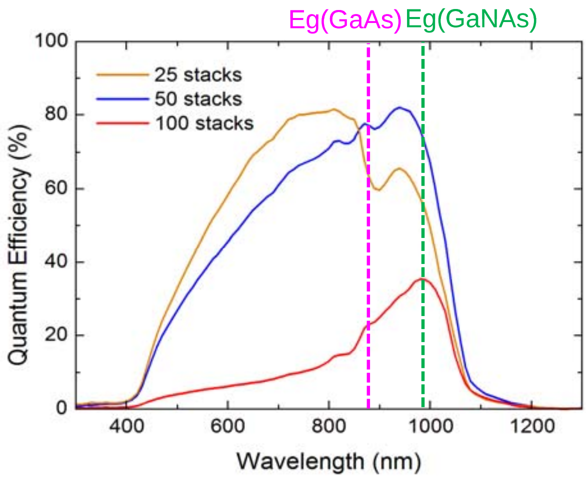

Figure 6 EQE spectra for 25, 50, and 100 layer-stacked InAs/GaNAs QDSC.

linewidth of ~ 40 meV indicating that the homogeneity of QD structure is unaffected during stacking. Furthermore, PL intensity is almost doubled by increasing the number of stacks from 50 to 100 layers and hence a good absorption and current can be expected if integrated into a solar cell structure.

Figure 6 show the external quantum efficiency (EQE) curves measured for 25, 50, and 100 layer-stacked InAs/GaNAs strain-compensated QDSCs. The thickness of GaNAs SCL is 20 nm in each cell. First, by embedding QD structure into the intrinsic layer, EQE clearly extends into the long wavelength range beyond the bandgap of host GaAs material of 880 nm in all samples. The EQE response from 880nm up to ~1100 nm is attributed to contribution mainly from GaNAs SCLs, and further response up to ~ 1200 nm is solely from InAs QDs, respectively. However, we found in a separate experiment that absorption by QD superlattice as determined by optical absorption measurements is small, on the order of 10 % even for 100 layer-stacked sample [5], and total light absorption by InAs QDs from the valence band to quantized states in QD, or IB needs to be further increased.

Second, it is noted that QE in the short wavelength region becomes largely degraded in 100 layer-stacked QDSC. Figure 7 plot QE spectra measured under short circuit condition for 100 layer-stacked InAs/GaNAs QDSC taken under varying applied reverse bias V_{appl} = 0 ~ -3.4 V. It is evident that applied reverse bias strongly affects the QE spectral shape as well as its magnitude over the entire spectrum. The QE response is significantly improved by applying a stronger reverse bias voltage. This suggests that the carriers generated in *p*-GaAs emitter region can not travel across the thick *i*-region (of > 2 um in 100 layer-stacked QDSC) and hence trapped and recombine in InAs QDs, as non-doped InAs QDs act as efficient recombination centers. Figure 8 plot the photocurrents as

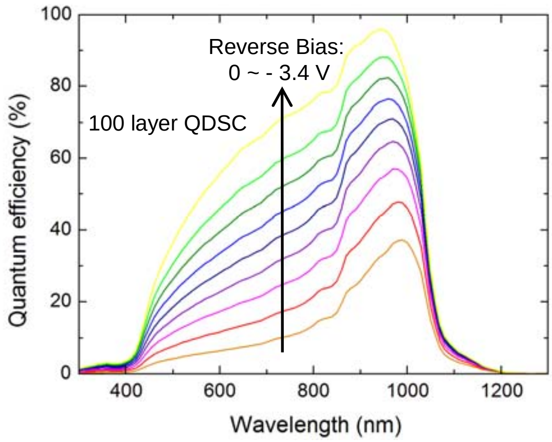

Figure 7 EQE spectra for 100 layer-stacked InAs/GaNAs QDSC as a function of applied reverse bias.

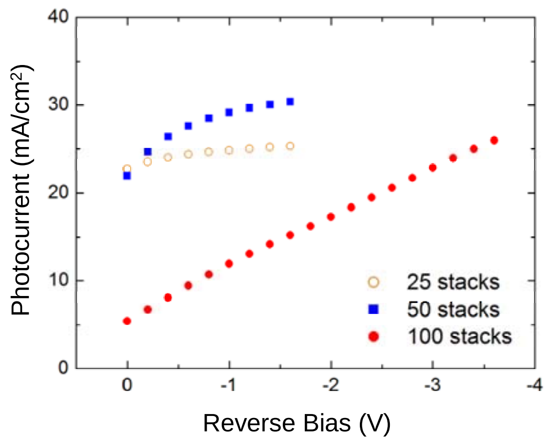

Figure 8 Photocurrents of QDSCs as a function of applied reverse bias.

a function of applied reverse bias voltage for InAs/GaNAs QDSC with 25, 50, and 100 QD multi-stacks. The photocurrent was calculated by taking the product of QE and AM 1.5 solar spectrum. The degree of increase in the current with reverse bias is largest for 100 layer-stacked QDSC. This again shows that photogenerated carriers are extracted effectively if there is a sufficient field across the QD structure.

SUMMARY

We have fabricated and characterized 25, 50 and 100 layer-stacked InAs/GaNAs IB-QDSCs on GaAs substrate using strain compensation technique. The short-circuit current density of QDSC increases with increasing number of QD stacks. However, QE in the short wavelength region was largely degraded in 100 layer-stacked QDSC. For this, reverse bias is necessary to extract photocarriers from the 100-layer stacked QDSC. From the absorption point of view, there is still a large mismatch between the rates of excitation of carriers from the valence band into IB, and from IB into the conduction band in our current SC structure.

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