Basic Research on the Science and Technology of High-efficiency Quantum Dot Superlattice Solar Cells

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I. Introduction

Recently, semiconductor quantum dots (QDs) incorporated in the active region of a *p-i-n* junction solar cell (SC) have attracted intense research as a possible means of exploiting the below bandgap infra-red photons to generate additional photocurrents beyond that corresponding to the band-to-band transition in a conventional single-junction SC[1,2]. In QDSCs, QDs must be uniform in size and periodically spaced in all 3 dimensions, which lead to the formation of an intermediate band to achieve the predicted high energy conversion efficiencies.

The experimental demonstration of QDSC requires fabrication of a three-dimensional QD superlattice placed in the active region. In our previous work[3], we reported on InP-based QDSC with 30 stacked InAs/AlGaInAs QD layers on InP (311)B substrate fabricated by *strain compensation technique*. Here the compressive strain due to QD layer was compensated by a tensile strain induced by the spacer layer. Consequently, the size homogeneity of QDs has improved to 12 ~ 13%. As our continuing effort, we applied this technique fabricate InAs/GaNAs QDSCs by the strain compensation technique[4-6].

II. Experiments and Results

We fabricated 4 series of solar cells S1, S2, S3, and S4 by molecular beam epitaxy (MBE) with a radio frequency (RF) plasma as the nitrogen source on n^+-GaAs (001) substrates[7,8]. The 4 samples were identical in the structure except for the *i*-layer region as shown in Fig. 1. The sample S1 and S2 were grown with 10 and 20 stacked pairs of an InAs QD layer with 2.0 monolayer (ML) thickness and a 20 nm-thick GaNAs strain compensation layer (SCL), respectively. The sample S3 comprised of 20 pairs of 2.0ML InAs QDs and 20 nm-thick GaAs spacer layers and hence the QD structure was strained. Lastly, the sample S4 was a GaAs control cell with a 400 nm-thick *i*-GaAs layer used as a reference. For electrical characterization, AuZn alloy was used for the top electrode and indium for the bottom, and a standard anti-reflection coating (ARC) was employed in $3 \times 3mm^2$ sized SCs.

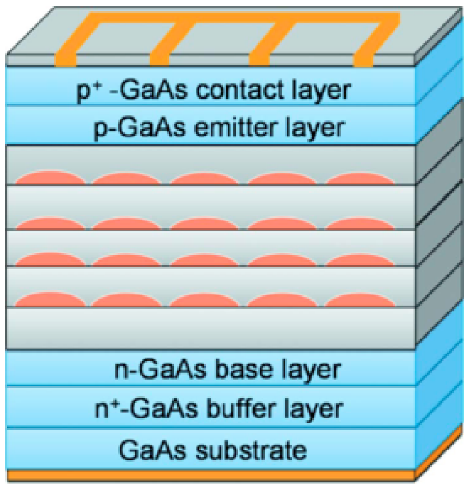

Fig. 1 Schematic layer structure of QDSC with multi-stacks of InAs QDs embedded by GaNAs strain compensation layers (SCLs), or conventional GaAs spacers.

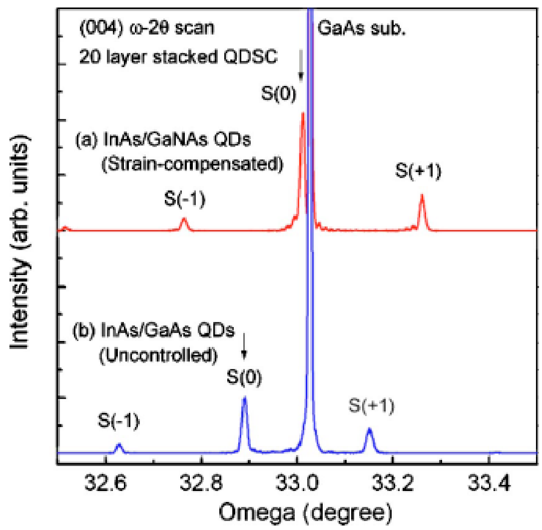

Fig. 2 Symmetric (004) *x*-ray diffraction patterns for samples with 20 stacked layers of InAs QDs structure embedded in (a) 20 nm-thick GaNAs SCLs (S2), and (b) 20 nm-thick GaAs spacer layers (S3), respectively.

First, we measured the symmetric scans around (004) reflection in a ω-2θ geometry in high resolution x-ray diffraction (HR-XRD) in order to characterize the effect of strain compensation. Figure 2 show the XRD spectra for sample S2 and S3. The satellite peaks originating from the periodic superlattice structure can be observed in each sample, and hence abrupt heterointerfaces are maintained. For S2, the compressive strain induced by InAs QD layers is compensated almost perfectly by 20 nm-thick GaNAs SCLs as shown in Fig. 2 (a), where the zeroth-order satellite peak indicated by an arrow in the figure shows a near perfect lattice-match with GaAs substrate.

Figure 3 shows the cross-sectional scanning transmission electron microscope (STEM) image measured for S2. A vertical alignment of lens-shaped QDs along the growth direction is maintained without degrading the structure and size uniformity from one layer to the next. Further, we observe no strain-induced defects or dislocations from the image, which would otherwise be generated if the build-up of lattice strain exceeds the critical layer thickness during the stacking. The mean QD diameter, height, size dispersion in diameter, and in-plane density were 39.1 nm, 5.2 nm, 13.4%, and $3.6 \times 10^{10}/cm^2$,

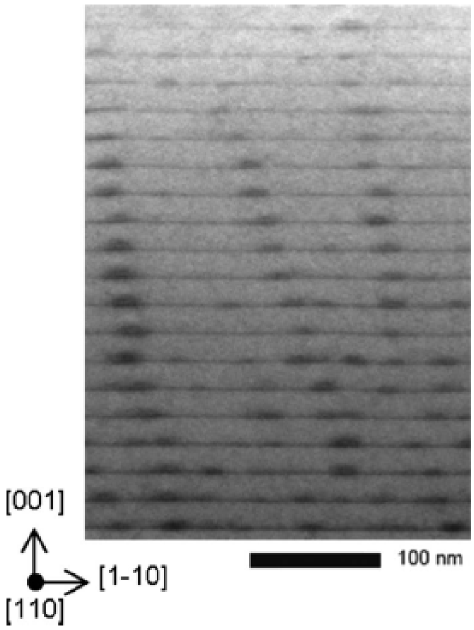

Fig. 3 Cross-sectional STEM image of 20 stacked layers of InAs QDs embedded in 20 nm-thick GaNAs SCLs (S2).

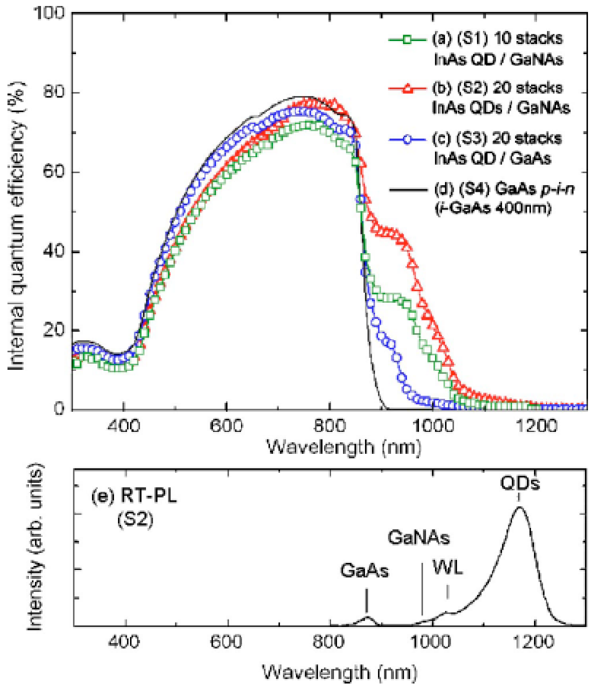

Fig.4 IQE spectra measured for;
(a) strain compensated InAs/GaNAs QDSC with 10 QD stacks (S1),
(b) strain compensated InAs/GaNAs QDSC with 20 QD stacks (S2),
(c) InAs/GaAs QDSC with 20 stacks (S3),
(d) GaAs *p-i-n* control cell (S4), and
(e) PL spectrum at room temperature for S2.

respectively. As a result, the total density of QDs amounts to on the order of $10^{12}/cm^2$, which would be difficult to achieve by the conventional strained heteroepitaxy.

Figure 4 (a) ~ (d) show the internal quantum efficiencies (IQEs) of QDSCs, S1 through S4, respectively. The IQE was determined from a direct measurement of the external QE under a constant photon irradiation of $10^{16}/cm^2$, then discounting for the shadowing loss and residual reflection loss arising from ARC. We also measured photoluminescence (PL) for each sample at room temperature by using a standard lock-in technique with a cw 532 nm second harmonic generation (SHG) Nd:YVO4 laser as an excitation source and InGaAs photodetector. The PL spectrum obtained for S2 is shown in Fig. 4 (e), which is used to identify the photoabsorption edge for each layer. Firstly, a single PL emission peak from the ground-state of InAs QDs is observed at around 1170 nm with a full-width half-maximum (FWHM) of 65.4 meV. In addition, a shoulder peak originating from GaNAs spacer layer and InAs wetting layer (WL) is seen at around 980 nm for S1 and S2, and 880 nm for S3, respectively. Secondly, the absorption edge redshifts from 1150 nm for S3 up to 1200 nm for S2 and this is because the quantum confinement is lower at the conduction band interface when GaNAs is used as the barrier instead of GaAs.

The filtered short-circuit current density (Isc) of sample S2 (strain compensated, 20 stacks) above the GaAs bandedge of ~ 880 nm, which gives the contribution from QD layer was calculated to be 2.47 mA/cm^2 by taking the product of IQE and 1.5 air-mass solar spectrum. This value is twice that of S1 (strain compensated, 10 stacks), and 4 times larger than that of S3 (strained, 20 stacks), respectively. Though the obtained value is notably higher than the reported values for QDSCs, the maximum IQE from the QD layers even for S2 is still limited to ~ 5 %, and thus a larger stack of QD layers is required to obtain higher photoabsorption. Furthermore, the fact that degradation of QEs in the shorter wavelength region is not observed after increasing the QD layer from 10 to 20 stacks suggests that the crystalline quality is maintained in our strain compensated QDSCs as the contribution to the photocurrent generated by the *p*-GaAs emitter is not affected. The dark I-V measurements show an improved diode factor of n = 1.42 for S2 compared to n = 1.62 for S3 due to the reduction of recombination loss as result of strain compensation. To this end, I_{sc} improved to 21.1 mA/cm^2 for S2 compared to 19.0 mA/cm^2 for S1, and 19.6 mA/cm^2 for S3, respectively, and I_{sc} of control cell S4 with 400 nm-thick *i*-GaAs layer was 19.7 mA/cm^2 as a reference.

III. Summary

In summary, we have successfully fabricated and characterized InAs/GaNAs QDSCs by strain compensation technique on GaAs(001) substrates. Compensating for the strain induced by QDs with a spacer layer that produces the opposite strain works well to achieve (1) improved size uniformity, (2) redshift of absorption edge, and (3) avoid generation of defects and dislocations. The total QD density of our QDSC with 20 InAs QD stacks is on the order of 10^{12}/cm^2, and I_{sc} = 21.1 mA/cm^2. The filtered I_{sc} above GaAs bandedge of 2.47 mA/cm^2 is 4 times higher than that for a strained QDSC with the identical cell structure.

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