

# High-density quantum dot superlattice for application to high-efficiency solar cells

current topics in solid state physics

Ryuji Oshima<sup>\*,1,2</sup>, Yoshitaka Okada<sup>1,2</sup>, Ayami Takata<sup>1,2,4</sup>, Shuhei Yagi<sup>1,3</sup>, Kouichi Akahane<sup>1,4</sup>, Ryo Tamaki<sup>1,2</sup>, and Kenjiro Miyano<sup>1,2</sup>

Received 14 June 2010, revised 23 August 2010, accepted 29 August 2010 Published online 3 November 2010

Keywords quantum dots, molecular beam epitaxy, solar cells

We characterized the optical absorption and solar cell characteristics of high-density InAs self-assembled quantum dots (QDs) grown on GaAs (001) substrates by atomic hydrogen-assisted molecular beam epitaxy. The GaNAs material can be used as strain-compensating layer (SCL) thereby minimizing the net strain, and thus is advantageous for multi-stacking of InAs QDs structures. As a result, dislocations and coalesced islands were not ob-

served in 100 layer-stacked QDs. For QD solar cell characterization, the short-circuit current density of QDSC increases with increasing number of QD stacks, and reaches as high as  $26.4~\text{mA/cm}^2$  for 50 layer-stacked sample under air-mass 1.5 condition. However, the light absorption by QD superlattice as determined by optical absorption measurements at is limited to  $\sim 10~\%$  even for 100~layer-stacked samples.

© 2010 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

## 1 Introduction

Recently, self-assembled quantum dots (QDs) have been widely applied to laser diodes [1] and semiconductor optical amplifiers [2] for optical fiber communication systems, as well as to high-efficiency intermediate-band solar cells (QDSCs) [3,4]. In particular, in QDSCs, the QDs are required to be homogeneous in size and periodically and closely spaced in all 3 dimensions, which would then lead to formation of an intermediate-band (IB) or a superlattice miniband, rather than a multiplicity of discrete quantized levels, so as to achieve the predicted high power conversion efficiencies.

However, Stranski-Krastanov (S-K) growth in InAs/GaAs system usually results in a degradation of stacked QD structure and generation of misfit dislocations, typically after 10 layers of stacking, as a result of accumulation of internal strain beyond the critical thickness [5,6]. For this reason, tensile-strained barriers such as GaP, GaAsP, and

GaNAs have been studied to balance out or compensate for the compressive strain induced in QD active regions [7-11]. In this work, we fabricated up to 100 stacked InAs/GaNAs QDs for application to IB-QDSCs. The dependence of the number of QD stacks on optical absorption and solar cell characteristics were characterized and analyzed.

## 2 Experiments

All growths were done by atomic hydrogen-assisted solid-source molecular beam epitaxy (H-MBE) with a radio frequency (RF) nitrogen plasma source [12,13]. We fabricated multi-stacked InAs/GaNAs QDSC on  $n^+$ -GaAs (001) substrate as shown in Fig. 1. An InAs QD layer with 2.0 monolayers (MLs) thickness and a 20 nm-thick GaN<sub>0.01</sub>As<sub>0.99</sub> strain-compensating layer (SCL) were consecutively grown in pair up to 50 layers and QD layer were incorporated in the center of the intrinsic region. The total thickness of intrinsic layer was set to 1.0  $\mu$ m for each cell

Research Center for Advanced Science and Technology (RCAST), The University of Tokyo, 4-6-1 Komaba, Meguro-ku, 153-8904 Tokyo, Japan

<sup>&</sup>lt;sup>2</sup> School of Engineering, The University of Tokyo, 4-6-1 Komaba, Meguro-ku, 153-8904 Tokyo, Japan

<sup>&</sup>lt;sup>3</sup> Saitama University, 225 Okubo, Sakura-ku, 338-8570 Saitama, Japan

<sup>&</sup>lt;sup>4</sup> National Institute of Information and Communications Technology, 4-2-1 Nukui-Kitamachi, Koganei, 184-8795 Tokyo, Japan

<sup>\*</sup> Corresponding author: e-mail oshima@mbe.rcast.u-tokyo.ac.jp, Phone: +81 3 5452 6514, Fax: +81 3 5452 6504

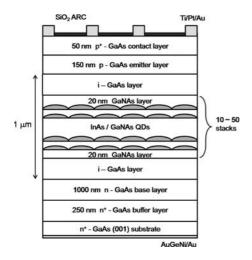


in order to make the built-in electric field to be constant at ~13.6 kV/cm. Other growth conditions have been reported elsewhere [11,14]. For solar cell characterization, Ti/Pt/Au alloy was used for the top contact and AuGeNi/Au for the bottom for 3×3 mm²-sized solar cells, and SiO<sub>2</sub> anti-reflection coating (ARC) was used. In order to study both structural and optical characterization, we fabricated up to 100 layers stacked InAs/GaNAs QDs on GaAs substrates without *p-i-n* structure.

### 3 Results and discussion

Figure 2(a) shows the AFM image of the topmost QD layer in 100 stacked layers of InAs/GaN<sub>0.01</sub>As<sub>0.99</sub> straincompensated QD sample. The mean QD diameter, height, size uniformity in diameter, and sheet density are 29.6 nm, 5.4 nm, 10.2 %, and  $4.2 \times 10^{10}$  cm<sup>-2</sup>, respectively. Figure 2 (b) shows the cross-sectional STEM image, and (c) shows the middle portion of 100 layer stacked sample, respectively. Compensating for the strain induced by QDs by using a spacer layer that exerts an opposite biaxial strain in strain-compensation growth works remarkably well to achieve good size uniformity as well as to avoid generation of defects and dislocations in multiple stacked QD structures. In Figs. 3(b) and (c), QD shape and size are homogeneous throughout the layers. As a result, the total density of QDs amounts to  $4.2 \times 10^{12}$  cm<sup>-2</sup>, which could not have been achieved by the conventional S-K growth without any strain control.

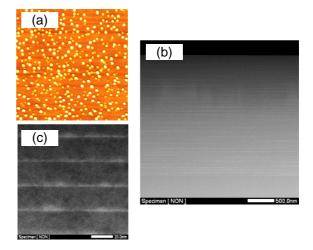
Figures 3(a)-(c) plot the projected current-voltage curves measured for each stacked QDSC and (d) shows that for GaAs solar cell as a reference, respectively. It can be seen that the short-circuit current density  $I_{\rm SC}$  increases from  $I_{\rm sc}=21.0$  to 26.4 mA/cm² as the number of stacks is increased from 20 to 50. This result was due to the enhancement in quantum efficiency above the band edge of GaAs (870 nm) by incorporating InAs/GaNAs QDs report-



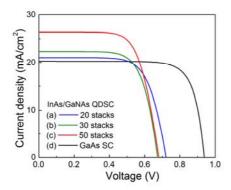
**Figure 1** Schematic structure of multilayer stacked InAs/GaNAs QDSC fabricated on GaAs (001) substrate.

ed in Ref. [14]. The filtered  $I_{\rm sc}$  above the GaAs band edge of 880 nm for 50 layers stacked QDSC is 6.29 mA/cm², and the contribution solely from InAs QDs, i.e, above the wavelength of 1000 nm, is 1.49 mA/cm², respectively. On the other hand, the open-circuit voltage ( $V_{\rm OC}$ ) drops by incorporating InAs/GaNAs QDs in the intrinsic region. However, the fill factor (FF) for each QDSC sample is almost the same of ~0.70 and increasing the number of stacks does not affect the diode factor of QDSC of ~1.6, which thus indicate that a good heterostructure quality is maintained even after 50 stacks.

Figures 4(a)-(d) show the differential optical density (OD) spectra. Because OD signals from GaAs and GaNAs spacer layer were subtracted from the measured spectra, (a)-(d) show the actual contribution from QDs. The OD intensity below 1.25 eV increases with increasing number of stacks. Further, OD peak around 1.2 eV is contribution from the wetting layer (WL), while sholder structure at 1.1 eV is contribution from the ground state of QDs, and these are in good agreement with PL result as shown in Fig. 4(e). However, we found that the absorption by QD superlattice as determined by optical absorption measurements at RT is small, on the order of 10 %, even for 100 layer-stacked samples. These results indicate that light absorption from InAs QDs is not enough and thicker QD layer as well as some optical management may be required for sufficient light absorption from the valence band to quantized states in QD, or IB. There is 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. Thus the recombination loss within QDs is the dominant factor limiting the open-circuit voltage as seen from Fig. 3.



**Figure 2** (a) AFM image of the topmost QD layer, (b) cross-sectional STEM image, and (c) magnified view of 100 layer stacked QD sample, respectively.



**Figure 3** (a)-(c) Plot of the projected current-voltage curves measured for each stacked QDSC and (d) shows that for GaAs solar cell as a reference.

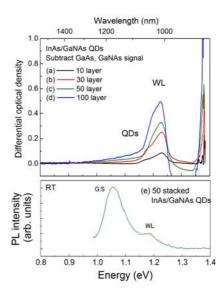


We successfully fabricated up to 100 layers of stacked InAs/GaNAs QDs using strain compensation technique and IB-QDSCs were characterized. The short-circuit current density of QDSC increases with increasing number of QD stacks, and reaches as high as 26.4 mA/cm² for 50 layer-stacked sample under air-mass 1.5 condition. However, the absorption by QD superlattice as determined by optical absorption measurements is on the order of 10 % even for 100 layer-stacked samples. These results indicate that InAs QD absorption is not enough and thick QD layer as well as optical management may be required for sufficient light absorption.

**Acknowledgements** This work is supported by the Incorporated Administrative Agency New Energy and Industrial Technology Development Organization (NEDO), and Ministry of Economy, Trade and Industry (METI), Japan.

# References

- [1] Y. Arakawa and H. Sakaki, Appl. Phys Lett. 40, 939 (1982).
- [2] M. Sugawara, H. Ebe, N. Hatori, M. Ishida, Y. Arakawa, T. Akiyama, K. Otsubo, and Y. Nakata, Phys. Rev. B 69, 235332 (2004).
- [3] A. Luque and A. Marti, Phy. Rev. Lett. 78, 5014 (1997).
- [4] A. J. Nozik, Physica E 14, 115(2002).
- [5] Z. R. Wasilewski, S. Fafard, and J.P. McCaffrey, J. Cryst. Growth 201/202, 1131 (1999).
- [6] A. Marti, N. Lopez, E. Antolin, E. Canovas, and A. Luque, Appl. Phys. Lett. 90, 233510 (2007).
- [7] S. M. Hubbard, C. D. Cress, C. G. Bailey, R. P. Raffaelle, S. G. Bailey, and D. M. Wilt, Appl. Phys. Lett. 92, 123512 (2008).
- [8] V. Popescu, G. Bester, M. C. Hanna, A. G. Norman, and A. Zunger, Phys. Rev. B 78, 205321 (2008).
- [9] N. Nuntawong, S. Birudavolu, C. P. Hains, S. Huang, H. Xu, and D. L. Huffaker, Appl. Phys. Lett. 85, 3050 (2004).



**Figure 4** (a)-(d) Differential optical density (OD) spectra at RT. (e) PL spectrum measured for 50 layer stacked QDs at RT.

- [10] Y. Okada, R. Oshima and A. Takata, J. Appl. Phys. 106, 024306 (2009).
- [11] R. Oshima, A. Takata, and Y. Okada, Appl. Phys. Lett. 93, 083111 (2008).
- [12] A. Takata, R. Oshima, H. Shigekawa, and Y. Okada J. Cryst. Growth 310, 3710 (2008).
- [13] Y. Shimizu, N. Kobayashi, A. Uedono, and Y. Okada, J. Cryst. Growth 278, 553 (2005).
- [14] R. Oshima, A. Takata, Y. Shoji K. Akahane, and Y. Okada, Physica E, in press.