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Efficiency limit of InAs/GaAs quantum dot solar cells attributed to quantum dot size effects

Im Sik Han [a], Ryan P. Smith [b], Jong Su Kim [a,*], Sam Kyu Noh [c], Sang Jun Lee [c,*], Chang-Lyoul Lee [d], Jae-Young Leem [e]

[a] Department of Physics, Yeungnam University, Gyeongsan 712-749, Korea
[b] Department of Physics, California State University - East Bay, Hayward, CA 94542, USA
[c] Division of Convergence Technology, Korea Research Institute of Standards and Science, Daejeon 305-340, Korea
[d] Advanced Photonics Research Institute, Gwangju Institute of Science and Technology, Gwangju 500-712, Korea
[e] School of Nano Engineering, Center for Nano Manufacturing, Inje University, Gimhae 621-749, Korea

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ABSTRACT

The effects of quantum dot (QD) size on the optical and electrical properties of InAs/GaAs QD solar cells (QDSCs) were investigated. QDSCs with varying InAs QD size were fabricated by controlling the total InAs deposition thickness (θ) from 0 to 3.0 mono-layers (ML). The optical and electrical properties of the QDSCs were investigated using photoluminescence (PL), time-resolved PL (TRPL), photoreflectance (PR) spectroscopy, capacitance-voltage (*C-V*), and current-voltage (*J-V*) measurements. The QD size effects on the p-n junction electric fields ($\boldsymbol{F_{pn}}$) and the efficiencies (η) of the QDSCs were revealed. The QDSCs had a maximum η of 21.17% for θ=2.0 ML (the efficiency is enhanced by 17.4% over the reference GaAs-SC) and minimized $\boldsymbol{F_{pn}}$ (113 kV/cm) by an enhanced photovoltaic effect caused by improved carrier generation. We find that these optimal properties result from a balance between carrier generation and exhaustion processes through trapping and re-capturing by defects and relatively large QDs.

1. Introduction

Quantum dot solar cells (QDSCs) based on compound semiconductors have attracted much attention over the past several years [1–7]. One approach to improving the efficiency (η) of QDSCs is introducing strain-based InAs/GaAs QDs to extend the absorption wavelength into the near-infrared (NIR) region of the solar spectrum [6]. Many methods have been proposed to achieve highly efficient QDSCs, such as the enhancement of the photocarrier generation and collection and suppression of carrier losses by re-capturing and trapping carriers in larger QDs and defect states [1–7].

To realize high-efficiency QDSCs, strain compensation layers (SCLs) were introduced to reduce strain-induced defects; examples of such layers include GaP and GaNAs thin layers for InAs/GaAs-based QDs, and incorporation of group-III ions with different sizes into II-VI host materials for InGaN/ZnO QDs [2–5]. High-density QD growth techniques have been proposed to improve carrier generation in the NIR region [6]. Moreover, to extend the absorption wavelength of an InAs/GaAs QDSC to the NIR region, the QD size distribution should be controlled since the absorption wavelength is related to the size of the InAs QDs. In general, increasing the QD size, the absorption wavelength is shifted to a longer wavelength. However, the stain-related defects caused by interface strain between InAs and GaAs cannot be avoided. Furthermore, the photo-generated carriers can be re-captured easily by large QDs during carrier transport. Photo-generated carrier loss could be increased by trapping and re-capturing carriers at the strain-related defects and QD states, respectively [7]. The trapped and re-captured carriers are exhausted by radiative and non-radiative recombination processes.

The carrier re-capturing probability should increase when increasing the QD size while the absorption wavelength is shifted to a longer wavelength. The defect generation is related to the QD size due to the stain near the QD interface and the capping materials. Therefore, to understand the contribution of the QD size effect to the $\boldsymbol{\eta}$ of QDSCs, it is necessary to consider the carrier trapping and re-capturing by the defects and QD states based on the QD size.

The present work revealed the QD size effects on η and p-n junction electric fields ($\boldsymbol{F_{pn}}$). To study this, various QD sizes were embedded in GaAs p-n junction QDSC structures. The effects of QD size on the optical and electrical properties were investigated by photoluminescence (PL), time-resolved PL (TRPL), photoreflectance

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* Corresponding authors.
E-mail addresses: jongsukim@ynu.ac.kr (J.S. Kim), sjlee@kriss.re.kr (S.J. Lee).

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(PR), capacitance-voltage (*C-V*), current-voltage (*J-V*), and solar simulator measurements.

2. Experiment

Five solar cell samples were fabricated with various QD sizes using molecular beam epitaxy (MBE). The QD sizes were controlled by supplying various deposition thicknesses of InAs (θ=0, 1.7, 2.0, 2.5, and 3.0 mono-layers (ML). Fig. 1 shows a schematic of the InAs/GaAs QDSCs and an atomic force microscopy (AFM) image of 2 ML InAs QDs embedded in the p-n junction. Each QDSC contains eight InAs QD layers located in the *n*-GaAs (2.8×10^{17}/cm^3) absorption layer region of a p$^+$-n-n$^+$ junction grown on n$^+$-GaAs (100) substrates. Each QD layer is separated by a 40 nm-thick n-GaAs space layer, repeated eight times for a total thickness of 320 nm for the QD structure. The doping concentration of n-GaAs absorption layer (2.8×10^{17}/cm^3) was selected to optimize SC efficiency (the absorption layer doping levels were varied from 2×10^{15}/cm^3 to 6×10^{17}/cm^3) in solar simulator measurements.

The reference GaAs p$^+$-n-n$^+$ junction solar cell (as a reference SC; θ=0 ML) consists of a 300 nm n$^+$-GaAs layer (2×10^{18}/cm^3), a 1.5 μm n-GaAs absorption layer, and a 0.6 μm p$^+$-GaAs layer (2×10^{18}/cm^3), followed by a 50 nm p$^+$-Al$_{0.9}$Ga$_{0.1}$As window and 10 nm p$^+$-GaAs layer for Ohmic contact. During the growth of QDSCs, the growth temperature was kept at 580 °C for all structures except the InAs QDs and the n-GaAs space layer (470 °C). A 320 nm n-GaAs layer grown at low temperature (470 °C) was inserted in the reference GaAs-SC to investigate only the QD size effect without the growth temperature effect. To enhance the photon recycling effect, the position of the QD layers were placed bottom of the n-GaAs absorption layer from the p$^+$-n interface. With this structure, relatively higher energy photons ($>$1.42 eV) are absorbed by n-GaAs absorption layer closer to the surface while NIR photons are absorbed by the InAs QD layers near the n-n+interface. These additional NIR photo-carriers contribute the photo-current because of the n-n+interface electric field.

The optical and electrical properties of the QDSCs were investigated by low-temperature PL, TRPL, room-temperature PR, *C-V* and *J-V* measurements. *J-V* measurements of the QDSCs were performed in the dark and under simulated AM1.5 G white light illumination using a Hewlett-Packard 4155A semiconductor parameter analyzer and a solar simulator (Polaronix K201 Lab50, McScience). The AM1.5 G white light was produced by a solar simulator based on a filtered Xe lamp (Oriel, 91,193). Its intensity was adjusted with a Si reference cell (Fraunhofer ISE, certificate No. C-ISE269) with a one-sun light intensity of 100 mW/cm^2.

3. Results and discussion

The AFM results indicate that the average diameters of the 1.7, 2.0, 2.5, and 3.0 ML InAs QDs were 21.5, 24.0, 40.6, and 53.5 nm, respectively, while the heights were 1.5, 2.5, 5.5, and 6.7 nm. The InAs QD size increased with increasing θ on the GaAs surface [8,9]. The density for the samples changed from $\sim 10^{10}$ to $\sim 10^{9}$/cm^2. The QD densities of the 1.7, 2.0, 2.5, and 3.0 ML QDs were 4.3×10^{10}, 5.0×10^{10}, 3.6×10^{10}, and 5.2×10^{9}/cm^2, respectively.

To investigate the effects of the QD size on optical properties such as the emission wavelength, PL spectra were measured with various QD sizes, as shown in Fig. 2(a). With increasing θ, the emission wavelengths shifted to the longer wavelength due to the QD size effect. However, the integrated PL intensities decreased with increasing θ with more than 2.5 ML. The full width at half maximum (FWHM) also increased, as shown in Fig. 2(b). The increased FWHM is attributed to the change in the QD size distribution with increasing θ. The small increase of the integrated PL intensity of the QDs between 1.7 ML and 2.0 ML can be explained by the QD density effect. However, the reduced intensity beyond 2.5 ML is predominately caused by increasing the defect densities because of the strain-induced defect generation in the S-K growth mode [9,10].

TRPL measurement was performed to investigate the formation of defects with increasing QD size and their role in the optical properties. Fig. 3(a) shows the low-temperature TRPL spectra with various θ, and Fig. 3(b) summarizes the PL intensity decay time as a function of θ. With increasing θ, the radiative carrier lifetimes (τ_R) drastically decreased, as shown in Fig. 3(b). Generally, τ_R increases with the QD size due to the reduced oscillation strength [11]. However, the decreasing τ_R is caused by the introduction of the non-radiative recombination process through the defect states. Therefore, the reduction of τ_R can be explained by the formation of defects with increasing QD size. Based on the PL and TRPL results, we find that the optical properties of InAs QDs predominately depend on the formation of strain-induced defects with increasing QD size.

To study the exhaustion of photo-generated carriers through trapping and re-capturing, the photovoltaic effect was investigated by PR measurements [12]. The PR spectra are sensitive to the interface electric fields such as the p$^+$-n junction ($\boldsymbol{F_{pn}}$) and surface ($\boldsymbol{F_s}$) fields. In the high electric field regime, Franz-Keldysh

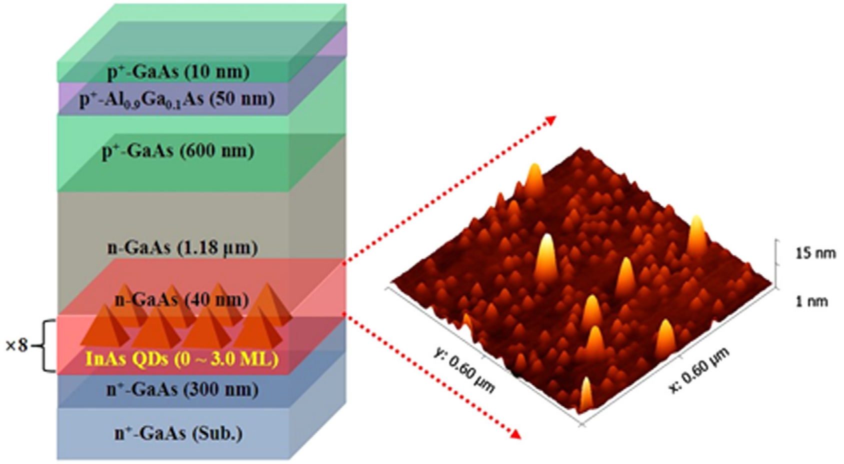

Fig. 1. Schematic of InAs/GaAs quantum dot solar cell (QDSC) and atomic force microscopy (AFM) image of 2 ML InAs QD embedded in the GaAs p-n junction.