

Improved electrical properties of silicon quantum dot layers for photovoltaic applications

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ABSTRACT

The low density of silicon quantum dots (Si QDs) in a dielectric matrix is one of the plausible reasons for the low power conversion efficiency (PCE) of Si QD solar cells due to large spacing between QD and substrate. In this study, the electrical properties of the Si QD layers were improved by increasing the density of Si QDs and reducing the barrier width. In the previous studies, Si QD layers were fabricated by alternating deposition of SiO₂/SiO_x layers and subsequent high temperature (1100 °C) annealing, where the diffusion of Si species in the SiO_x layer into Si substrate was observed. In order to prevent the diffusion, we propose the substitution of the SiO_x layer with an amorphous Si (a-Si) layer. In [SiO₂ (2 nm)/SiO_x (2 nm)] × 34 multilayer structures, the first few SiO_x layers were substituted with a-Si layers. As a result, Si QDs could be well formed in the a-Si layer with high density and survived near the interfaces between the first Si QD layer and Si substrate. The distance of the nearest Si QDs from the Si substrates was also reduced from 7.2 nm to 3.6 nm, which results in the effective carrier transfer by tunneling between the Si QDs. The PCE of heterojunction B-doped Si QDs/crystalline Si solar cells was enhanced by using the Si QD structures with the improved electrical properties. Since the variations of open-circuit voltage and short-circuit current are not so large, the enhancement of PCE is mainly related with the improvement of fill factor due to the decrease of series resistance.

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1. Introduction

All silicon multiple junction solar cell has been proposed as a promising next-generation solar cell with high efficiency and low fabrication cost to overcome the efficiency limit of the crystalline Si solar cell. In a tandem solar cell with a top Si quantum dot (QD) layer with a band gap energy of 1.7–1.8 eV, the conversion efficiency has been expected to be increased to 42.5% [1]. It can be available by effectively utilizing the high energy photons in the solar light by a Si QD top cell with high band gap energy due to quantum confinement effect.

Various fabrication methods for self-assembled Si QD layer with Si QDs imbedded in SiO₂ matrix have been reported [2–5]. Heterojunction Si QD solar cells with the structure of Si QD layer/Si (100) substrate have been realized from the self-assembled Si QD layer [6–8]. Additional efforts in the optimization of QD density [9] and dielectric matrix [10–12] to improve photovoltaic properties of heterojunction Si QD solar cells as well as the realization of all Si tandem solar cell [13–15] were reported. In the study of single junction

Si QD solar cells on quartz substrates, although the open-circuit voltage of about 400 mV was obtained, the short circuit current was too low to realize the single junction Si QD solar cell [13,15–17].

An intrinsic drawback of the Si QD solar cells is the high series resistivity of the Si QD layer due to the insulating property of the SiO₂ matrix material [18]. The photon-generated carriers are transferred by tunneling between the Si QDs imbedded in SiO₂ matrix. The tunneling probability exponentially decreases as the distance increases [19]. For the conductivity improvement in the Si QD layer, Si QDs should be distributed closely and uniformly in the dielectric matrix.

In this paper, the distribution of Si QDs in SiO₂ matrix is investigated, and a new method to increase the density of Si QDs in the interface between the Si QD layer and the crystalline Si (100) substrate is suggested for the improvement of electrical conductivity of the Si QD layer. We also explore the effect of high density Si QDs near the interface on photovoltaic properties of heterojunction B-doped Si QDs/crystalline Si solar cells.

2. Experimental

[SiO₂ (2 nm)/B-doped SiO_x (2 nm)] × 34 multilayer films were grown on 6" Si (100) substrates (n-type, 1–3 Ω cm) by ion beam

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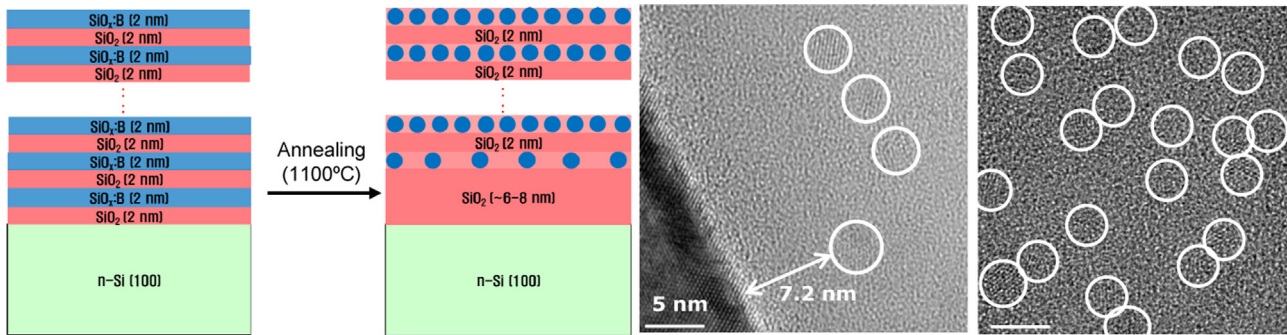


Fig. 1. Formation of Si QD multilayers by alternate deposition of SiO_x (2 nm) and SiO_2 (2 nm) layers and subsequent annealing at 1100 °C for 100 min. Cross-section HRTEM images were obtained (left) near the interface between a few Si QD layers and the Si substrate, and (right) in the center of whole Si QD layers.

sputtering deposition method. In order to form highly homogeneous films over the whole wafer, the substrate was rotated. The stoichiometric SiO_2 layers were deposited by reactive sputtering of a Si wafer using an Ar^+ beam with 800 eV under a high oxygen partial pressure of 1.7×10^{-4} Torr. The B-doped SiO_x ($\text{SiO}_x\text{:B}$) layers were deposited under low oxygen partial pressures by co-sputtering of Si and B using a composite target, in which a small boron chip of $1 \text{ cm} \times 1 \text{ cm}$ size is mounted on a Si wafer. The stoichiometry of the SiO_x layers was precisely controlled by variation of oxygen partial pressure and confirmed by in situ x-ray photoelectron spectroscopy (XPS) analysis. Details of the XPS system are described elsewhere [20]. In this experiment, the stoichiometry of the SiO_x layers was fixed to $\text{SiO}_{1.2}$. The doping concentration ($4.75 \times 10^{20} \text{ cm}^{-3}$) was measured by secondary ion mass spectrometer (CAMECA-IMS-7f system).

In this study, a number of the B-doped SiO_x layers near the substrate were substituted with a-Si layers to increase the density of Si QDs near the interfaces. The B-doped a-Si layers were simply grown by the same method as for the B-doped SiO_x layers without flow of oxygen gases. In addition, the thickness of the B-doped a-Si layer was reduced to 1.5 nm to get the same average atomic fraction of the Si QD layers. The multilayer films were annealed at 1100 °C for 100 min in nitrogen atmosphere to form Si QDs. The formation of Si QDs was investigated by high resolution transmission electron microscopy (HRTEM) and photoluminescence (PL) measurements. TEM images were obtained by a Cs corrected TEM, JEM-ARM200F (JEOL, Japan). PL spectra were measured using 404 nm laser as an excitation source and a streak camera detector at room temperature.

For the fabrication of heterojunction B-doped Si QDs/crystalline Si solar cell, the Si QDs films grown on 6 in. Si (100) wafers were mechanically diced to small chips of $1 \text{ cm} \times 1 \text{ cm}$ size. The surface oxide layer formed during the annealing process was etched by dipping the chip in buffered oxide etchant (BOE) to reduce contact resistivity. Aluminum films of about 500 nm thickness were deposited as the front and the back side electrodes by thermal evaporation. After the deposition of electrodes, the cells were heated at 425 °C for 40 min in an electrical furnace in air to enhance the contact properties. Photovoltaic properties of the Si QD solar cells were investigated with illuminated I - V measurements and external quantum efficiency (EQE) measurements. The effective area covered by finger-shape metal contact was 0.19 cm^2 . The I - V curves and EQE properties were obtained using a McScience K201 solar simulator and an Oriol IQE-200 instrument, respectively.

3. Results and discussion

Boron-doped Si quantum dots (B-doped Si QDs) on a n-type Si (100) wafer was fabricated by multiple alternate deposition of SiO_x (with an O/Si atomic ratio of 1.2) and stoichiometric SiO_2 layers. Films comprising thirty four periods of B-doped $\text{SiO}_x/\text{SiO}_2$ bi-layers were

deposited, and the thickness of each SiO_x and SiO_2 layer was controlled to 2 nm for the effective carrier tunneling between the Si QDs through the dielectric SiO_2 layer. When a thinner SiO_2 layer, which is preferable for carrier transport, is used as the barrier, the Si QDs tend to form larger sizes of QDs due to the coalescence across the porous sputtered SiO_2 layers [11,13,21,22]. Fig. 1 shows a general method for the formation of Si QD multilayers by precipitation of Si QDs from the $\text{SiO}_{1.2}$ layers during subsequent post-annealing at 1100 °C for 100 min. As shown in the high resolution TEM (HRTEM) images (Fig. 1), the density of Si QDs near the interface between the Si substrate and the Si QD layer is very low compared with that in the central area of the whole Si QD layer. It may be caused by the fact that the metallic Si species in the SiO_x layers near the Si substrate are diffused and precipitated into the Si substrate across the thin SiO_2 layers during the annealing process [11,13,21,22]. In addition, we found the separation between the nearest Si QD layer and the Si substrate is about 7.2 nm, which is too long for effective carrier transport.

In order to increase the density of Si QDs near the interface and reduce the separation distance, we substituted the first SiO_x layer (2 nm) with 1.5 nm of amorphous Si (a-Si) layer. Fig. 2 shows the film structures for the three different Si QD solar cells and HRTEM images of the individual cells near the interface region. Cell 1 is a typical $\text{SiO}_x[2 \text{ nm}]/\text{SiO}_2[2 \text{ nm}]$ multilayer structure same as the structure shown in Fig. 1. To prevent the metallic Si in the SiO_x layers near the Si substrate from precipitating into the substrate, we fabricated the Cell 2, where the first SiO_x layer was substituted with 1.5 nm of a-Si layer. A HRTEM image of Cell 2 (Fig. 2) exhibits that the separation between the first Si QD layer and the Si substrate is 4.2 nm. Furthermore, the average distance measured from different sample areas and samples is 4.4 nm. In order to quantify the enhanced Si QD density near the interface, the density in TEM images was optically counted in the first 25 nm (height) from the substrate by 40 nm (width) rectangular region. The optical analysis suggested that the average Si QD density for Cell 1 and Cell 2 measured from five different TEM images is 0.015 and 0.02 per nm^2 . The TEM results indicate that the density of Si QDs near the interface of Cell 2 is higher than that of Cell 1, resulting in the reduced separation between the first Si QD layer and the Si substrate from 7.2 nm to 4.2 nm.

We also substituted the first three SiO_x layers from the substrate with a-Si layers to further increase the Si QD density near the interface and reduce the separation. Even though the separation (3.2 nm) was further reduced compared with the Cell 2, we found a large size and laterally elongated shape of Si QDs near the interface caused by aggregation of precipitated Si QDs (Supporting information, Fig. S1). Since the large size of Si QDs reduce the quantum confinement effects, this structure was excluded from further evaluations. To further reduce the separation and prevent the aggregation, we substituted only the first and third SiO_x layers from the substrate with 1.5 nm of a-Si layers. The TEM results of Cell 3 (Fig. 2) show that the separation and the average Si QD density of Cell 3 is 3.6 nm and 0.027 per nm^2 ,

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