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Voltage dependence of two-step photocurrent generation in quantum dot intermediate band solar cells

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ABSTRACT

We studied in detail the voltage dependence of two-step photocurrent generation through a two-step process of absorbing sub-band gap photons of different photon energies in a GaAs/AlGaAs quantum dot Intermediate Band Solar Cell. Our experiments revealed that two-step photocurrent generation is largely dependent on voltage, and exhibits a maximum at −0.3 V. A notable feature is a monotonic decrease in two-step photocurrent in the forward bias region, where the operating point of the solar cell lies. Using a model of rate equations, we extracted the voltage dependence of the individual escape and recombination rates, and found that the decrease in two-step photocurrent in the forward bias region is related to a monotonic increase in recombination rate in the quantum dots with increasing bias.

1. Introduction

Intermediate Band Solar Cells (IBSCs), known as one of Third Generation Photovoltaics concept [1], have attracted considerable research attention due to their potential to overcome the Shockley-Queisser conversion efficiency limit [2–7]. The IBSC model suggests that the total efficiency of a solar cell can be increased by generating an additional photocurrent by means of a two-step photon absorption process through an Intermediate Band, that is, intermediate energy states located between the conduction and valance band of a semiconductor, while maintaining the open-circuit voltage of the host material. The advantage of IBSCs lies in the fact that maximum theoretical conversion efficiencies reach >60% under full concentration of sunlight and their device structure which requires only a single p-n junction is simple. Several ways to create IB energy states have been proposed such as highly mismatched semiconductor alloys [8], impurities in the semiconductor band gap [9], and quantum structures [4]. Among them, quantum dots (QDs) are practically important because of a large freedom in material combinations and controllability of IB energy levels by their size and composition. QDs ideally exhibit a zero density of states between their confined energy states and conduction/valance bands which is of particular importance to isolate the IB from conduction and valance band [10]. Several groups have observed two-step photocurrent generation using QDs [5–7]. However, the results have hitherto been limited to mostly short-circuit condition, far from the operating point of a practical solar cell, or the signal was observed only at low temperatures (<70 K) because of their shallow confinement energies.

From the viewpoint of device performance, it is important to study the voltage dependence of the two-step photocurrent signal at room temperature. Comparison of the two-step signal with current-voltage characteristics of the solar cell in turn provides an understanding of IBSCs, since the signal is directly related to the carrier density, n, stored in the QD. n is given by the balance of escape and recombination of carriers in the QD when carriers are generated only in the confined states, which is modeled by rate equations [11–13].

Here we report the voltage dependence of the two-step photocurrent generation in a QD-IBSC at room temperature and explain it in relation to its current-voltage characteristics under QD excitation using rate equations. In this study, to observe a signal at room temperature, we employ a 2-nm-thick GaAs artificial wetting layer [14,15] underneath the GaAs QDs, which lowers the QD ground state energy.

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2. Experimental

2.1. Sample fabrication

We grew AlGaAs p-i-n solar cells with GaAs QDs using standard solid-source molecular beam epitaxy on n-type GaAs (0 0 1) substrates. The sample structure is shown schematically in Fig. 1a). Five QD layers were embedded in the middle of a 600-nm-thick undoped AlGaAs layer, sandwiched by n- and p-AlGaAs layers ($Al_xGa_{1-x}As$, $x=0.28$). An AlGaAs reference solar cell was grown using an identical growth recipe, except that the 600-nm-thick AlGaAs i-layer was continuously grown at 600 °C without QD layers.

Each QD layer consists of a 2-nm-thick GaAs quantum well, hereinafter termed an ‘artificial wetting layer’, with droplet epitaxy QDs grown on top. The artificial wetting layer increases the effective height of the QDs and lowers their ground state energy [15]. The artificial wetting layer is doped by Si with a doping density of 1×10^{18} cm^{-3} to supply electrons as the majority carries inside the quantum structure [16]. The thickness of the AlGaAs barrier between each QD layer is 20 nm.

The GaAs QDs were grown using standard droplet epitaxy. First, Ga droplets were formed at 200 °C by supplying a Ga flux equivalent to five-monolayer deposition of GaAs. These Ga droplets were then crystallized using an As flux of 6×10^{-5} Torr beam equivalent pressure for ten seconds, after which the QDs were annealed at 400 °C for 10 min. As shown in Fig. 1b), the QDs exhibited a well-defined shape with an average height of 3.5 nm and average diameters of 34.8 and 43.0 nm in the [1 1 0] and [1$\bar{1}$0] direction, respectively. The effective dot height, including artificial wetting layer, was 5 - 6 nm. We achieved a high areal density of 4.6×10^{10} cm^{-2} to ensure strong optical absorption by the QD layers. It should be noted that droplet epitaxy can be used to grow extremely high densities of QDs [17].

Solar cell devices were fabricated by patterning the sample by photolithography and sputtering a Ti/Pt/Au front side contact. The solar cell devices had a total area of 0.66 mm^2 with a total area for illumination of 0.34 mm^2.

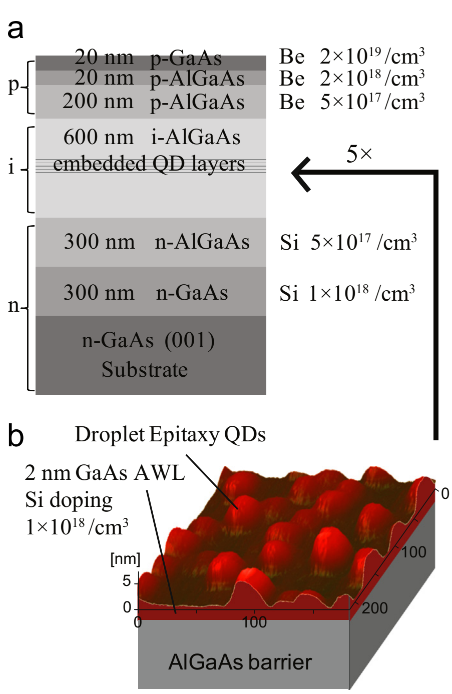

Fig. 1. Sample structure. a) Sample structure of the p-i-n AlGaAs solar cell with GaAs QDs. b) Three-dimensional AFM image of droplet epitaxy-grown QDs with underlying artificial wetting layer (AWL) and applied Si doping.

2.2. Characterization methods

Photoluminescence (PL) measurement was performed by exciting the grown sample with a 532 nm laser (15 W/cm^2) and detecting the radiative recombination emitted from the sample fed through a spectrometer mounted with a charge-coupled device camera. The photocurrent measurement was performed by illuminating the sample with monochromized light from a halogen lamp and measuring the generated photocurrent under short-circuit condition.

To measure the two-step photocurrent generation, ΔJ_{ph}, we first measured the voltage dependence of the photocurrent density J_{ph} under first light illumination of 780 nm (5.8 mW/cm^2) and then repeated the measurement with an additional 1.55 μm laser (1.4 W/cm^2) to obtain J_{ph}'. The two-step photocurrent signal is defined as the difference in photocurrent generation under one and two light illumination, $\Delta J_{ph}=J_{ph}' - J_{ph}$. The photon energy of the 1.55 μm laser of 800 meV can only induce the intra-band transition of carriers out of quantum confinement to the conduction band but does not have sufficient photon energy to induce the inter-band transition of carriers in the QDs or bulk material: this was confirmed by noting zero photocurrent under illumination by the laser alone. Any heating effects caused by the strong second light illumination were prevented by choosing appropriate light intensity and providing good thermal contact and air ventilation.

3. Results and discussion

3.1. Photoluminescence and photocurrent spectra

Fig. 2a shows the PL spectra of the QD solar cell. Clear PL emission is observed at room temperature. The two peaks at 820 nm (1.513 eV) and 695 nm (1.785 eV) are the emissions from the QDs and AlGaAs, respectively. The shoulder at 759 nm

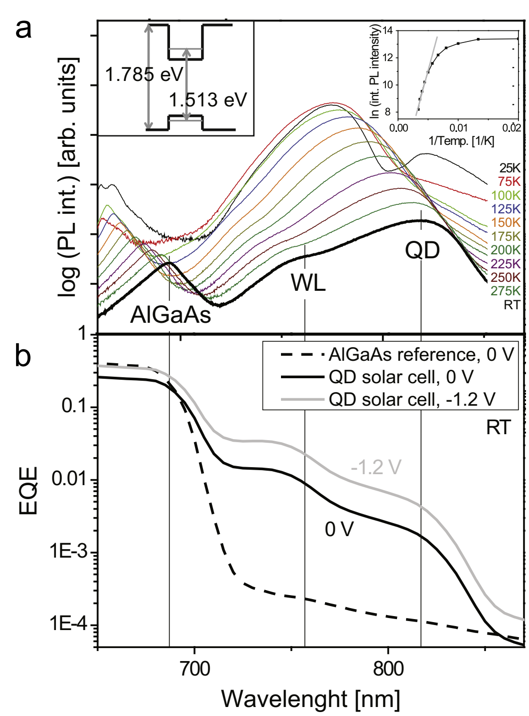

Fig. 2. Photoluminescence and external quantum efficiency. a) Photoluminescence emissions of the QD embedded solar cell. The insets show a schematic illustration of the band diagram of the structure (left) and the Arrhenius plot of the integrated PL intensity of QD peak (right). b) External quantum efficiency of the QD solar cell and AlGaAs reference at room temperature under short-circuit condition and $V=-1.2$ V.