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Analysis of voltage dependence of two-step photocurrent generation in GaAs/AlGaAs quantum dot solar cells using rate equations

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The Intermediate Band Solar Cell (IBSC) has attracted considerable research attention since the concept was theoretically proposed in 1997 with the promise of overcoming the Shockley-Queisser conversion efficiency limit. In an IBSC, an additional photocurrent is generated by a two-step photon absorption process which excites carriers via intermediate energy states from the valance band to the conduction band while the open-circuit voltage of the high band gap material is maintained. While some progress has been achieved in fabricating structures where intermediate energy states are realized by using quantum dots (QDs) in which a two-step photocurrent generation process can be observed, a detailed understanding of this key IBSC process is still insufficient. To gain a physical understanding of the experimental results of IBSCs, the dependence of carrier population in the intermediate energy states, as well as the influence of recombination and escape rates on the two-step photocurrent generation needs to be investigated. Using the droplet epitaxy technique to fabricate solar cells with embedded lattice-matched GaAs/AlGaAs QDs, we have achieved a clear two-step photocurrent generation at room temperature [1, 2]. Here, we analyze the voltage dependence of the two-step photocurrent generation using rate equations and extract the individual recombination and escape rates of QD carriers.

A p-i-n SC structure is grown on GaAs (100) substrate by molecular beam epitaxy. Five QD layers separated by 20 nm barrier layers are embedded in the middle of a 600-nm-thick i-region. To lower the ground state energy level of the QDs, we introduced a 2-nm-thick GaAs quantum well layer underneath each QD layer which increases the effective QD volume [3]. Si-doping is applied to the wetting layer underneath the QDs with a doping density of 2×10^{11}cm^{-2}, which is about four times the areal density of QDs (4.6×10^{10}cm^{-2}) and is designed to supply electrons as majority carriers in the QD energy states.

To measure the two-step photocurrent generation, we first measure the voltage dependence of the photocurrent under 780 nm illumination (one light illumination), which excites carrier in the QD energy states, and then repeated the measurement with an additional 1.55 μm laser (two light illumination). Note that the 1.55 μm laser can only excite the intraband transition of carriers from the intermediate QD energy states to the conduction band/valance band and does not have sufficient photon energy to excite the QD or AlGaAs interband transition. Compared to one light illumination, under additional 1.55 μm laser light, a clear increase in photocurrent generation is observed which originates from a second photon absorption process, i.e. the two-step photocurrent generation. Its voltage dependence reveals a maximum of the two-step photocurrent generation at -0.3 V. Importantly, a monotonic decrease in two-step photocurrent with increasing voltage is present in the forward bias region, where the operating point of the solar cell lies. We use a model of rate equations and extract the voltage dependence of the individual escape and recombination rates of QD carriers from the experimentally measured photocurrent generation under one and two light illumination. The escape rate is high under high reverse voltage and decreases with increasing voltage. This dependence is explained by the bias dependence of the thermally-assisted tunneling escape of carriers from the QD energy states, which allows easy tunneling through the barrier under high reverse bias, and is dominated by thermal escape towards flat band condition, which is only weakly dependent on bias. The recombination rate exhibits an opposing trend and increases with increasing voltage, which is explained by the increasing overlap of wave functions, and increasing hole population towards forward bias condition. Therefore, we can directly link the two-step photocurrent generation to the carrier population in the QD energy states and explain the decrease towards forward bias by the increasing recombination rate. The presented analysis of the voltage dependence is important to understand the two-step photocurrent generation under operating condition of an IBSC device.

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