Solar Energy Materials & Solar Cells 130 (2014) 225–233

Contents lists available at ScienceDirect

Solar Energy Materials & Solar Cells

journal homepage: www.elsevier.com/locate/solmat

The feasibility of high-efficiency InAs/GaAs quantum dot intermediate band solar cells

A. Mellor [a,*], A. Luque [a], I. Tobías [a], A. Martí [a]

[a] Instituto de Energía Solar, Universidad Politécnica de Madrid, Avda. Complutense 30, 28040 Madrid, Spain

ARTICLE INFO

Article history:
Received 17 March 2014
Received in revised form
3 July 2014
Accepted 4 July 2014

Keywords:
Quantum dot
Intermediate band
High efficiency
Light trapping

ABSTRACT

In recent years, all the operating principles of intermediate band behaviour have been demonstrated in InAs/GaAs quantum dot (QD) solar cells. Having passed this hurdle, a new stage of research is underway, whose goal is to deliver QD solar cells with efficiencies above those of state-of-the-art single-gap devices. In this work, we demonstrate that this is possible, using the present InAs/GaAs QD system, if the QDs are made to be radiatively dominated, and if absorption enhancements are achieved by a combination of increasing the number of QDs and light trapping. A quantitative prediction is also made of the absorption enhancements required, suggesting that a 30 fold increase in the number of QDs and a light trapping enhancement of 10 are sufficient. Finally, insight is given into the relative merits of absorption enhancement via increasing QD numbers and via light trapping.

1. Introduction

The intermediate band solar cell (IBSC) is a high efficiency solar cell concept whose detailed balance efficiency limit has been calculated as 63% [1] to be compared to the Shockley–Queisser limit of 41% [2] for conventional single-bandgap solar cells. IBSCs have been realized using semiconductor quantum dots (QDs) [3–9], highly mismatched alloys [10–12] and semiconductor bulk materials containing a high density of deep-level impurities [13–15], amongst others. This work deals exclusively the first of these (QD-IBSCs), paying specific attention to the InAs/GaAs based prototypes, which have received particular attention.

Research into IBSCs has largely been conducted in two phases; the earliest works were theoretical studies of the thermodynamic consistency [16] and absolute limiting efficiency [1] of the IBSC concept, and theoretical studies of possible implementations and design issues [3,17,18]. In the second phase, extensive work was undertaken to fabricate and develop QD-IBSC prototypes [4–8] and demonstrate that they fulfill the fundamental operating principles of the IBSC: most importantly the generation of subbandgap photocurrent due to sequential two-photon absorption via the intermediate band/levels [19], and a V_{oc} that is above the subbandgap absorption thresholds [20]. A recent review of which technologies have demonstrated these principles can be found in Ref. [21]; in the context of QD-IBSCs, they have been demonstrated on the InAs/GaAs material system, though the voltage preservation has only been demonstrated at low temperatures [20].

Following the demonstration of the operating principles, QD-IBSC research is now entering a third phase, whose ultimate goal must be to deliver solar cells with conversion efficiencies surpassing those of conventional single-gap devices. For this to be achieved, two broad technological advances are needed. First, QD-IBSCs must demonstrate high V_{oc}s at room temperature. This requires that thermal carrier escape from the QDs be suppressed [22] and that non-radiative processes in the cells be minimised. Recent results of reduced thermal escape due to a higher bandgap (AlGaAs) host material [23], and high QD-IBSC V_{oc}s due to improved material quality [24] are promising in this regard. Second, the subbandgap photocurrent must be improved significantly by increasing subbandgap photon absorption in the QD array. This may be achieved by increasing the number of QDs, by optical absorption enhancement, or a combination of both. QD arrays are being fabricated with an increasing number of QDs both in the context of QD-IBSCs [25] and elsewhere [26]. Optical absorption enhancement is standard in wafer-based crystalline silicon solar cells [27,28] and thin-film crystalline silicon solar cells [29], and we expect and encourage researchers to bring this technology into the context of QD-IBSCs.

This paper is aimed at informing this third phase of research. The experimental achievements described in the previous paragraphs have been accompanied by theoretical studies of the fabricated prototypes [7,30–34], giving us a much deeper understanding of their operation and highlighting pathways to improvement. Based on this understanding, it is now possible to make a realistic assessment of the feasibility of achieving high efficiencies

* Corresponding author. Tel.: +44 207 594 1030; fax: +44 207 594 2077.
E-mail address: a.mellor@imperial.ac.uk (A. Mellor).

http://dx.doi.org/10.1016/j.solmat.2014.07.006
0927-0248/

using InAs/GaAs QD arrays, and to quantify the advances that are required to do so. In this paper, such an assessment is made using our recently developed realistic detailed balance model, which has been shown to reproduce well the measured quantum efficiency of an InAs/GaAs QD-IBSC prototype and its temperature dependence [34].

This paper has three objectives:

1. to reassess the feasibility of achieving high efficiencies in InAs/GaAs QD-IBSCs, based on the understanding that has been acquired over the previous years.
2. to quantify the degree of absorption enhancement that is required for present QD-IBSCs to supersede the efficiency of an equivalent single gap reference.
3. to compare the relative merits of absorption enhancement via increasing the number of QDs and employing optical absorption enhancement techniques.

2. The detailed balance model applied to the QD-IBSC prototype

The subject of our study is the sample labelled SB in Ref. [22] and S3 in Ref. [35]. This sample has been chosen since it has demonstrated all of the operating principles of the IBSC concept [21]. A simplified level/band diagram of a single QD in this exemplary QD-IBSC is shown in Fig. 1. The grey lines represent the VB and CB band edges, the offsets being due to the InAs QD. The confining potential in the CB supports three discrete bound state levels in the energy range of the host forbidden band. These are denoted IB levels, and are labelled IB1, IB2 and IB3, with energies E_{IB1}, E_{IB2} and E_{IB3}, respectively (these correspond to the states labelled (1 1 1), (2 1 1)/(1 2 1), (2 2 1) in Ref. [31]). The VB potential pedestal supports a quasi-continuum of confined heavy-hole states which act to displace the energy onset of the VB to $E_{VB,\ eff}$ (shown), thus reducing the effective overall bandgap from that of the GaAs host (1.42 eV) to the value $E_{g,\ eff}$= 1.24 eV. The energies of the principal transitions between the VB pedestal and the IB levels, E_{t1}, E_{t2}, E_{t3}, are given in the figure.

The system is treated as five distinct electron populations: that of the CB, that of the VB (whose upper energy onset is considered

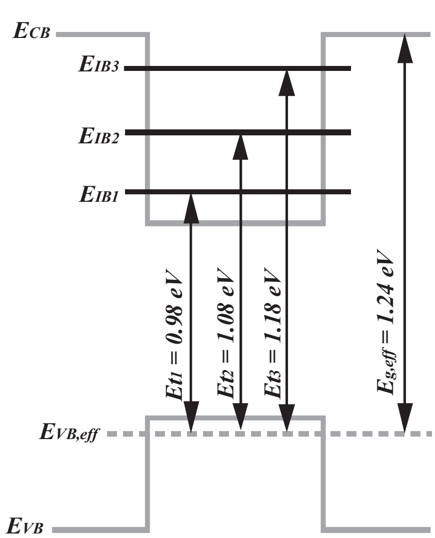

Fig. 1. A simplified band diagram of a single QD in the exemplary QD-IBSC. Upper grey line: conduction band edge. Lower grey line: valence band edge. Black lines: confined state energy levels whose energy is within the host forbidden band. Dashed grey line: effective valence band edge.

to be at $E_{VB,\ eff}$), and those of the three discrete IB energy levels. The electron populations are each described by a Fermi–Dirac function with a distinct quasi Femi level (QFL): $E_{F,IB1}$, $E_{F,IB2}$ and $E_{F,IB3}$, $E_{F,CB}$ and $E_{F,VB}$.

The model is based on solving five simultaneous equations to yield the five QFLs defined above. Three equations emerge from the condition that the electron population in each IB level is time constant, implying that the sum of transitions received by any IB level per second is equal to the sum of transitions proceeding from that level. One equation emerges from assuming that the net charge is zero in most of the QD layer stack. The final equation sets the difference between the VB and CB QFLs equal to the externally applied bias voltage, which is equivalent to assuming infinite carrier mobility throughout the device. This is approximately achieved in GaAs solar cells. Present QD-IBSC prototypes exhibit higher series resistance [36]. However, this is likely due to unwanted recombination and non-optimal contacting, both of which can be improved. Indeed, IV curves of more recent QD-IBSC prototypes suggest series resistances close to GaAs references have been achieved [24].

Once calculated, the QFLs are then used to calculate the strengths of the subbandgap transition currents. Adding these to the above-bandgap photocurrent, which is calculated using the standard single-gap Shockley–Queisser model with the bandgap of the GaAs host (1.42 eV), yields the total current produced at the terminals of the QD-IBSC for a given applied voltage.

The realistic detailed balance model differs from previous detailed balance models used to calculate the upper limits to IBSC efficiency [1] in two important aspects. First, whereas previous detailed balance models assume that the subbandgap transitions absorb all incident photons in the corresponding energy range, the present model calculates the subbandgap transition currents using the absorption coefficients that have been calculated for the investigated QD-IBSC using a four-band $k \times p$ model [32,37]. This $k \times p$ model has been shown to have good agreement with the measured absorption [32]. Second, the model considers the multiple intermediate levels, and not just the ground state. This is important since the thermal escape of electrons from the QD has been shown to occur via the ladder of excited states between the ground state and the CB [33,34]. Correspondingly, we expect the V_{oc} to be limited by recombination that occurs via the excited levels.

It must be emphasised that the model only considers radiative processes (i.e. photogeneration and radiative recombination). In our previous work, the model was shown to reproduce well the experimental quantum efficiency, and its temperature dependence, at short circuit: a condition under which the prototype is believed to be radiatively dominated. Closer to open circuit, the present prototypes are believed to be dominated by non-radiative processes, such as Shockley–Read–Hall recombination. Therefore, we expect the model to overestimate the V_{oc} of the present prototypes (a simple comparison of the results presented here with experimental V_{oc}s in Ref. [20] verifies this). Nonetheless, the use of this radiative model is justified for the present study, as is described in the following.

QD-IBSCs must be made to be radiatively dominated under operating conditions if they are to supersede the efficiency of single-gap devices. The purpose of this paper is to determine whether, using the present QD and host materials and QD dimensions, radiative operation combined with absorption enhancement is sufficient for these efficiencies to be achieved. It is therefore appropriate to use a radiative model. Notice that the recombination involved in the radiative model is unavoidable and therefore it represents its lower limit; non-radiative recombination is deemed to be avoidable.

A detailed description and derivation of the model can be found in Ref. [34]. A brief mathematical description is given in