EXPERIMENTAL ADVANCES IN THE NEXT GENERATION OF SOLAR CELLS

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ABSTRACT: We consider next generation solar cells concepts those that have the potential to exceed the limiting efficiency calculated by Shockley and Queisser for single gap solar cells (40.7 %) and still have not been commercialized. Among these concepts, this paper deals with the multiple exciton generation (or impact ionization or multiple carrier generation) solar cell, the intermediate band solar cell and the hot carrier solar cell. These concepts were proposed theoretically more than ten years ago. In the last years, the number of experiments supporting the theories behind and paving the way towards their practical implementation has leaped forward. This work reviews these experimental advances.

Keywords: multiple exciton generation, intermediate band, hot carrier.

1 INTRODUCTION

In photovoltaics, by often refer as "next generation solar cells" to those solar cell concepts that have the potential for exceeding what is known as the Shockley and Queisser (S&Q) efficiency limit [1] and are not in the market. This limit, known as detailed balance limit, was calculated by S&Q for single gap solar cells (a review can be found in [2]) resulting in 40.7 % for maximum light concentration when the sun is assumed as black-body at 6000 K. The fundamental aspect behind this theory (that S&Q realized first) is that a solar cell can absorb photons through band to band transitions at the price of emitting a minimum number of photons caused by radiative recombinations. In fact, given a band to band absorption coefficient α, for a semiconductor material, the radiative recombination constant cannot take any value but is linked to α through the van–Roosbroeck-Shockley relation [3]. Furthermore, S&Q detailed balance is so nicely tuned, that implicitly it also takes into account photon recycling, that is, the possibility that an emitted photon can be reabsorbed again [4].

Multijunction solar cells have the potential to exceed this limit [5] and are already in the market. Therefore, they will be out of this review accordingly to definition given above. We will focus then, in particular, in the multiple exciton generation solar cell, the intermediate band solar cell and the hot carrier solar cell.

2 MULTIPLE EXCITON GENERATION SOLAR CELLS (MEGSC)

The basic concept behind the MEGSC relies on considering the possibility that the absorption of a photon with an energy at least twice the semiconductor bandgap can generate two, or more, electron hole pairs depending on how many times the bandgap the energy of the photon is (Fig.1).

Hence, while in a conventional single gap solar cell, (at least) the energy in excess the bandgap is lost as useful work, in the MEGSC, this energy has a chance of being more productively used by creating more than one electron hole pair.

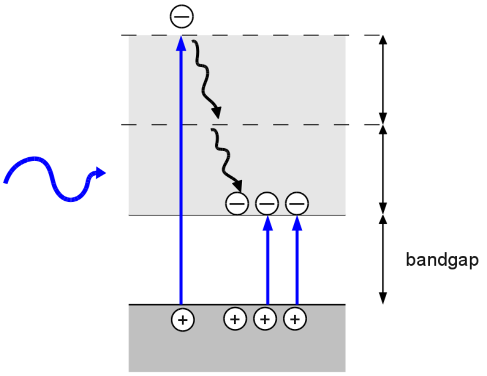

Figure 1: Illustration showing the fundamental operation of an impact ionization (or MEG) solar cell.

As [6] points out, the first reference to the possibility of one photon generating more than one electron-hole pair in solar cells can be traced back to footnote 12 of the original S&Q paper [1]. The context referred then to impact ionization or Auger generation taking place in bulk semiconductors. In this mechanism, a high energy photon produces, for example, a high energy electron that relaxes to the bottom of the conduction band by releasing its energy to an electron in the valence band (Fig. 2). Impact ionization was already experimentally known to exist in the 50's, not only in silicon and germanium diodes when operated in reverse, but also under X-ray illumination [7, 8]. A few more other materials such as InSb, CdS and CdSe [9] and, in particular PbS [10] (which will be relevant later for the discussions) had been experimentally studied using metal semiconductor junctions (PbS) under illumination and photoconductivity measurements (InSb, CdS and CdTe) finding in all of them evidence of the existence of impact ionization processes.

However, it was not until 1993 that the relevance of the issue for solar cell applications was retaken when Kolodinski, Werner, Wittchen and Queisser [11] actually measured quantum efficiencies higher than one in the ultraviolet region of high quality silicon solar cells. Armored with the power of the detailed balance theory, the limiting efficiency of the concept was found to be 85.4 % at maximum light concentration [12]. This result stimulated deeper research on the idea. Kolodinski, Werner and Queisser soon after emphasised [13] that, when taking place in bulk semiconductors, together with energy conservation, crystal momentum should also be conserved what reduced the probability of the mechanism taking place and therefore, reduced its practical application to solar cells.

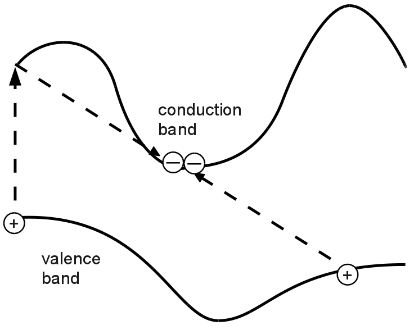

Figure 2: Illustration and impact ionization process in bulk semiconductors (adapted from [13]).

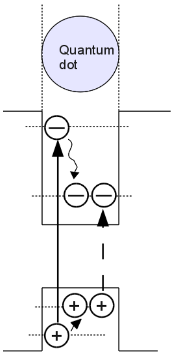

Figure 3: Illustration of the multiple generation of excitons mechanism in a quantum dot (adapted from [14]).

Probably, because of this conclusion, the idea slept again for some years until in 2002 Nozik [14] proposed that, if quantum dots were used instead, the likelihood for the impact ionization process taking place was probablely higher. The main reason for this enhancement would be that, in quantum dots, crystal momentum would not a good quantum number any more for determining which transitions were allowed or not. Other benefits when using quantum dots would come from the modified cooling rates or the enhanced coulomb interaction between electrons and holes in quantum dots. In addition, quantum dots offered the possibility of tailoring the bandgap of the system by controlling the size of the QDs. Instead of “high energy electrons”, the incidence of a high energy electron would create a high energy exciton in the QD that, when relaxing, would create multiple excitons. Because of the physics was so different from bulk semiconductors, the concept was preferred to be referred to as “multiple exciton generation” solar cell.

Schaller and Klimov (Los Alamos National Laboratory, LANL) were the first in measuring experimentally the MEG mechanism in PbSe colloidal quantum dots [15]. They reported 200 % efficient carrier multiplication (meaning the generation of two excitons from a single photon) using photons with an energy 3 times the bandgap. The experimental technique used is known as transient absorption (TA). Ellingson et al. [16] (National Renewable Energy Laboratory, NREL) reported shortly after efficiencies of 300 %, also in PbSe colloidal QDs for photon energies 4 times the bandgap. In 2005, LANL group reported an exceptional 700 % efficiency for photons with energy 7.8 times the bandgap. This result triggered the interest of other researchers in reproducing the result. In this respect, Nair, Geyer, Chang and Bawendi [17], from the Massachusetts Institute of Technology (MIT) reported efficiencies of only 125 % using photon energies 5 times the bandgap and claimed that it had not possible for them to reproduce LANL’s results by using transient photoluminescence as experimental technique. This generated some controversy in the scientific community, from which science resulted benefited by triggering studies that allowed a better understanding of the processes involved and that illustrated how variations in efficiency from 100 to 240 % could be obtained depending on the chemical treatment [18] of the quantum dots or how photocharging of the quantum dots [19] could have exaggerated the efficiency of previous results. Some other valuable studies contributed to provide additional data for the PbSe QD system [20-22].

Not only PbSe quantum dots, as referred above, have been studied, but also PbS[16, 17], PbTe [23], CdSe[24-28], CdTe [27, 28], Si [29] and InAs [22, 30, 31] with, sometimes, also discrepancy in the results.

With the revised efficiencies, a first question arising was whether there was an actual advantage in using the QDs over the bulk materials or not. When plotted against the absolute energy of the incident photons, bulk materials were able to produce more carriers per photon. However, QDs were able to produce them at a lower energy threshold (lower energy/bandgap ratio) [19]. It was realized, however, that there was actually more knowledge over the impact ionization efficiencies in QDs than in bulk materials where data still came from the research done in the 50’s (see paragraphs above) and had been done using different experimental techniques. In this framework, Pijpers et al. made another valuable contribution measuring the impact ionization rate in bulk PbSe and PbS using THz spectroscopy. Recently, with the available data, Beard et al. [32] have recalculated the limiting efficiencies of bulk PbSe QDs and compared with bulk PbSe finding the QD system more favourable. They also have provided convincing arguments in favour of analyzing the results as a function of the photon energy/bandgap ratio.

Another question arising is how to take to device level the colloidal QD systems described above. In this respect, Luther et al [33] have sandwiched PbS and CdS nanocrystal films in-between ITO and several metal electrodes obtaining 2.1 % efficiency devices (corrected to AM1.5 G 100mWcm-2) with unusual high shortcircuit current densities 21.4 mAcm-2). In spite of this high current density, the authors acknowledge they cannot obtain clear evidence of quantum efficiencies higher than one. Other works have followed with similar solar cell structures obtaining 3 % efficiencies with PbS [34] and 5% with CdTe colloidal nanocrystrals [35]. Remarkablely, Sukhovatkin et al. [36] have made PbS QD photodectors that, although not operating in the photovoltaic mode, have shown the production of more than one electron-hole pair per incident photon leading to the first photoconductive photodetector exploting this phenomena with QDs. On the other hand, recently, in a different system (single-walled carbon nanotubes) close to the limit high carrier multiplications efficiencies have been measured [37].

3 INTERMEDIATE BAND SOLAR CELLS (IBSC)

Fig. 4 illustrates the intermediate band solar cell concept. The fundamental principle behind it is the absorption of below bandgap energy photons thanks to the existence of an intermediate band within the semiconductor bandgap and the subsequent production of an increased photocurrent with an output voltage still limited by the total semiconductor bangap E_G [38, 39].

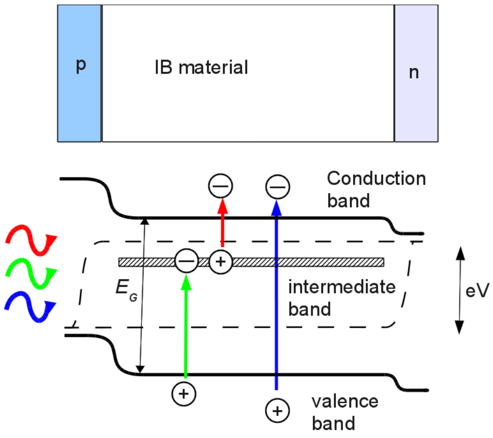

Figure 4: Illustration of the intermediate band solar cell concept.

First references to the possibility of using energy levels inside the bandgap to increase the solar cell efficiency appeared in 1960 in a paper by Wolf [40] but, again, it slept for more than 30 years, perhaps because it was believed that energy levels inside the bandgap would introduce more losses due to non-radiative recombination than benefits. In 1994, Keevers and Green [41], with a revised theory of the Shockley-Read-Hall recombination that included photogeneration by the impurity and light trapping, applied it to In doped silicon with the In acting as deep centre and predicted a small efficiency improvement (1-2 % in absolute terms).

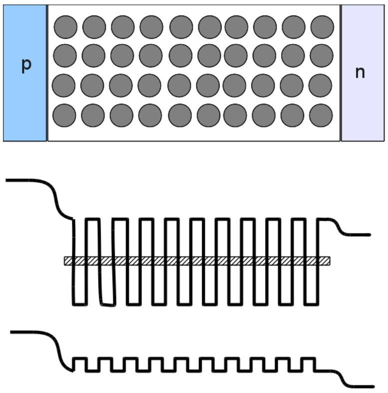

Figure 4: Illustration of the intermediate band solar cell concept implemented with QDs.

In 1997 we freed [38] the theory from the constrains of assuming that an energy level inside the bandgap had to perform non-radiatively and assumed they could perform radiatively. This allowed us to calculate a limiting efficiency of 63.2 %, at maximum concentration, for the concept (Strandberg and Reenaas [42] have recently reviewed the limiting efficiencies at lower concentration finding them higher than previously calculated when selective optical filters were included). We also clarified the conditions for preserving the output voltage at the same time the intermediate band allowed to increase the photocurrent. This voltage preservation would be achieved by sandwiching the intermediate band material between two single gap semiconductors what would indeed allow the electron and hole quasi-Fermi levels, whose split determines the output voltage, to be limited by the total bandgap E_G (Figure 4).

Nevertheless, something had to be done about this level or levels to behave radiatively and, in particular, we hypothesized that the collection of energy levels inside the bandgap, when implemented in bulk material, should constitute a band [39] (therefore the name, “intermediate band”). In 2000 [43], together with Cuadra, we proposed the implementation of the concept with quantum dots (Fig. 5) and in [44], together with Antolín and Tablero, gave form to the theory that allowed the formation of a band from deep centers.

The first QD intermediate band solar cell, based on InAs/GaAs, was manufactured in 2004 [45], together with Prof. Stanley’s group at the University of Glasgow and Compound Semiconductor Technologies, and demonstrated the production of photocurrent for below bandgap energy photons. It must be said, however, that an ideal IBSC, when illuminated with monochromatic below bandgap energy photons should not produce any current [46] and that the existence of such current reveals the existence of some non-ideal phenomena, such as, for example, the existence of thermal escape from the IB to the CB [47].

In spite of InAs/GaAs QDs does not lead to an optimum IBSC system, its mature technology recommended continuing using it to demonstrate the principles of operation of the IBSC. Hence, our group, together with the University of Glasgow that manufactured the devices, focused research on demonstrating that it was possible to generate one net electron-hole pair from two below bandgap energy photons and that voltage was not limited by any of the two sub-bandgaps the IB divides the total bandgap E_G into. The two-photon mechanism was experimentally demonstrated in 2006 [48] and an output voltage larger than the subbangaps, was demonstrated recently [49] after some partial proof related to the existence of three separated quasi-Fermi levels (associated each to one band) was found in 2005 [50].

In this framework, our knowledge about QD-IBSCs has benefited from the empirical research done by many groups worldwide. Hence, for example, researchers at Rochester Institute of Technology and NASA [51] investigated strain compensation in order to improve material quality and prevent the degradation of the emitters of the cells [52]; at NREL, Popescu et al. [53] have also investigated strain compensation and carried out a detailed analysis of the potential of the (In,Ga)As/Ga(As,P) system at fundamental level confirming it is not an optimum material system to achieve high efficiencies (see [54] in this Conference for a re-evaluation of the InAs/GaAs system considering tandem structures); Oshima et al., from the University of Tokyo and the University of Tsukuba [55] have investigated strain compensation with GaNAs achieving one of the largest contributions from subbandgap photocurrent; Guimard et al., from the University of Tokyo, have recently shown no degradation of the opencircuit voltage in comparison with the voltage due to the wetting layer [56]; Ban et al. [57], from the University of Delawere, the Ariona State University and NREL have investigated the InAs/GaAsSb system which has the potential advantage of being a QD system with zero valence band offset. Blockin et al.,[58] from the St. Petersburg Physics and Technology Centre for Research and Education, the Ioffe Physicotechnical Institute and Innolume GmbH, with a 18 % efficient device, have obtained so far the device with the highest efficiency.

Wang et al. [59], from the University of Michigan have implemented the first intermediate band solar cell based on bulk semiconductors. Their system is based on the incorporation of oxygen into ZnTe.

At material level, Yu et al., from Lawrence Berkeley National Laboratory, University of California and MIT have demonstrated through photoreflectance measurements the creation of intermediate band materials in II-VI diluted dioxides [60]. Yu and his co-workers, from the Lawrence Berkeley National Laboratory and Applied Materials, have also experimentally identified GaNAsP quaternary alloys as intermediate band materials [61] using this technique (see [62] at this conference for a description and application of the photoreflectance technique to IBSC and other photovoltaic structures). Antolin et al.,[63] from our group and in collaboration with the Universidad Complutense de Madrid, have measured the effective lifetime of Ti implanted silicon wafers finding an improvement in its lifetime as the Ti dose increases in agreement with our predictions in [44]. Lucena et al.,[64] from the Consejo Superior de Investigaciones Científicas and our Institute at the Universidad Politécnica de Madrid, have measured the absorption spectra of V:In_2S_3, synthesized using solvothermal techniques and finding the signatures of the intermediate band that had been previously predicted theoretically using ab-initio methods [65].

4 HOT CARRIER SOLAR CELLS (HCSC)

Fig. 5 illustrates the basic operation of a hot carrier solar cell. In this cell, a high energy photon excites a high energy electron hole pair that has to be extracted as photocurrent before it recombines. This cell was proposed by Ross and Nozik, from the Ohio State University and the Solar Energy Research Institute in 1982 [66]. Würfel's emphasized [67, 68] that to be effective, this cell would require of special contacts capable of isoentropically cooling the hot carriers and increase their electro-chemical potential. Würfel also calculated a 85 % limiting efficiency for this device.

Perhaps, because of the difficulty of achieving both conditions (special contacts and inhibited carrier cooling), the concept was considered very difficult to take to practice and again was abandoned for several years. However, in 2003, Coniber et al. [69], from the University of New South Wales (UNSW), proposed the use of tunnel resonant structures implemented with QDs to manufacture the required contacts. First experimental work in this respect, by the same group, appeared published in 2008 [70]. We have calculated that if ideal selective contacts would be applied to Si and Ge cells, otherwise ideal but for the existence of inelastic interaction mechanism (multi-valley scattering) between electrons and phonons, their limiting efficiency is improved.

In order to prevent carrier cooling, Conibeer, Guillemoles and Green proposed the use of phononic bandgap structures [71] that could be given in materials, such as InN, characterized by a large difference in the atomic masses of its elements. Together with Koning, they have extended the idea to the use of quantum structures [72]. A recent review can be found in [73] and at this same Conference [74].

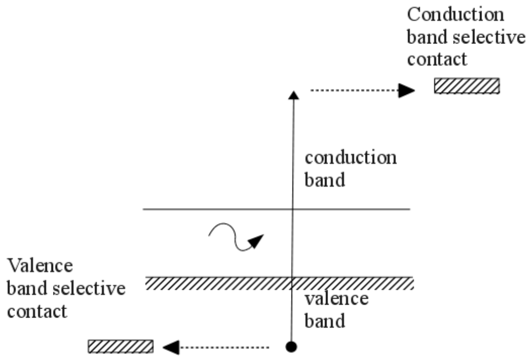

Figure 5: Illustration of the operation of the hot carrier solar cell, including the selective contacts (adapted from [67]).

5 SUMMARY

Next generation solar cells have moved from theory to experimental research. In the case of the MEGSC, several colloidal quantum dot systems (in particular, PbSe) have shown evidence of carrier multiplication. The challenge now is, perhaps, to produce a photovoltaic device, based on quantum dots, in which quantum efficiencies higher than one can be effectively measured [33].

In the case of the intermediate band solar cell, most of the experimental research has taken place at device level, in particular using devices integrating quantum dots that have allowed to demonstrate the principles of operation of the concept. Best QD-IBSC exhibits an efficiency of 18 % [58]. Recently, the first intermediate band solar cells made of bulk material (O:ZnTe) has been manufactured [59].

In the case of the hot carrier solar cell, experimental research focuses at material level by attempting to engineer both materials with reduced carrier cooling as suitable schemes for the selective contacts.

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