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Modelling of organic triple-junction solar cells

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ABSTRACT

In order for organic bulk heterojunction solar cells to compete with the traditional inorganic cells, higher power conversion efficiencies are desirable. A characteristic of organic solar cells is their narrow absorption window, compared to the absorption band of inorganic semiconductors. A possible way to capture a wider band of the solar spectrum - and thus increasing the power conversion efficiency - is using two or more solar cells with different bandgaps in a row, referred to as a multi-junction solar cell. In this article, we study the theoretical efficiency potential of three organic cells in a row, i.e. a triple-junction. We study the influence of the energy levels of donor and acceptor, as well as different absorption windows of the subcells. We not only study the light harvesting potential of the usual monolithic configuration, but also consider a stacked set-up. Ideal material characteristics are obtained from these calculations, giving an idea of how the ideal organic triple-junction cell should look like. An interesting result is that it is not necessary to develop photovoltaic organic materials with an absorption window broader than 300 nm for triple-junctions, because hardly any efficiency gain can be achieved by a broader absorption window.

Keywords: photovoltaic energy, organic PV, triple-junction, multi-junction, absorption window

1. INTRODUCTION

Photovoltaic solar cells based on organic compounds are promising candidates for solar energy conversion. They have the potential for cost effectiveness, mechanical flexibility and easy processing. Nowadays, a record efficiency of 10 % is reached[1] for single junction cells of 1 cm² and 4.2 % for a submodule (10 series cells)[2,3]. In order to compete with the traditional inorganic cells, higher power conversion efficiencies, certainly for larger cells, are desirable.

A characteristic of organic solar cells is their narrow absorption window, compared to the absorption band of inorganic semiconductors[4]. A possible way to capture a wider band of the solar spectrum - and thus increasing the power conversion efficiency - is using more solar cells with different bandgaps in a row, referred to as a tandem or multi-junction solar cell. In this article, we will focus on triple-junction solar cells, i.e. three cells in a row. The absorber of the first single solar cell in such a triple-junction cell has a large bandgap E_{g1}. High-energy photons with an energy $h\nu > E_{g1}$ are absorbed by the first cell. The second cell, with a lower bandgap E_{g2}, absorbs the middle-energy photons with an energy between E_{g1} and E_{g2}. The third cell absorbs the low-energy photons between E_{g2} and E_{g3} (Figure 1). In this configuration, the photon energy is used more efficiently: the voltage at which electrical charge is collected in each subcell is closer to the energy of the photons absorbed in that subcell.

In the ideal configuration, the subcells are electrically separated. This is called the stacked or 6-terminal configuration (Figure 1a). However, experimental and commercial multi-junction solar cells are usually of the monolithic (integrated or 2-terminal) type (Figure 1b). This means that they are not only optically in series, but also electrically in series. This configuration will never reach an efficiency that is higher than that of a stacked (6-terminal) triple-junction cell, because all single cells cannot be operating at their optimal working point at the same time (unless they have an equal maximum-power current).

Organic tandem solar cells, where both single cells are of the organic solar cell type, have already been fabricated by several research institutes[5-8], as well as fully organic multi-junction cells[9-11]. The efficiency of these cells hardly achieves the maximal efficiency of a single organic cell. Nowadays, efficiencies of more than 6 % are reached for organic tandem cells[12].

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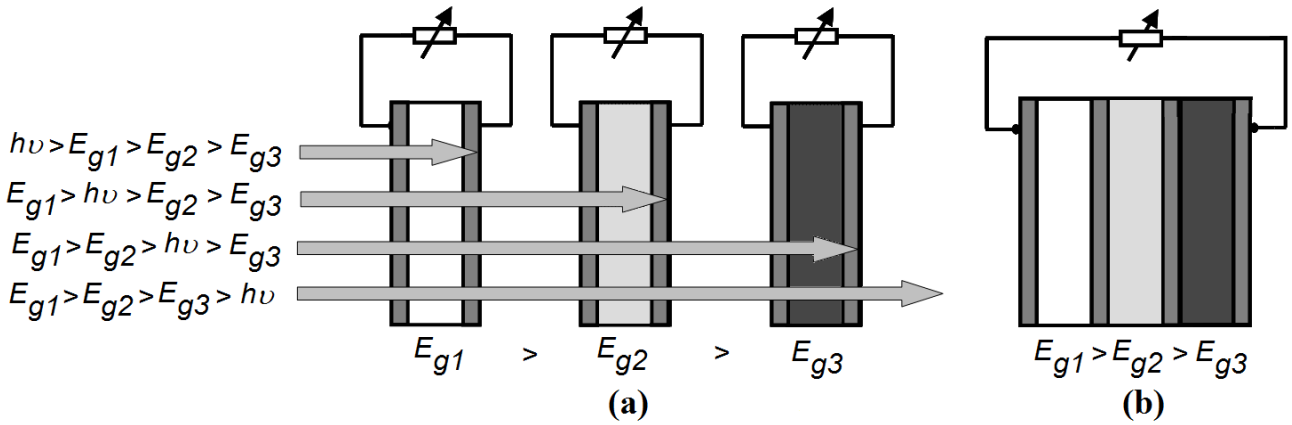

Figure 1. (a) A stacked or 6-terminal triple-junction solar cell: the first single cell absorbs photons with an energy higher than E_{g1}. The second and third cell absorb photons with an energy between E_{g1} and E_{g2} ,respectively, E_{g2} and E_{g3}. Photons with an energy below E_{g3} are not absorbed. The three subcells are electrically separated. (b) A monolithic or 2-terminal triple-junction solar cell: the single cells are electrically connected in series.

In this article, we calculate the theoretical upper-limit for the efficiency of organic triple-junction solar cells. Although this maximum efficiency itself is only interesting from a theoretical point of view, the ideal material characteristics obtained from these calculations can give us an idea of how the ideal organic triple-junction solar cell should look like. Previous work on predicting the efficiency of organic (tandem) solar cells has been done by multiple authors[13-21]. In this work, we include the influence of among others different absorption windows for each subcell. Moreover, our calculations are not only presented for a stacked organic triple-junction cell, but also for a monolithic organic cell. The results presented in this paper are meant to increase the fundamental understanding of the relation between on the one hand the energy levels of donor and acceptor and the absorption window of the subcells, and on the other hand the light harvesting potential of the configurations.

2. ASSUMPTIONS

The active material in a single organic bulk heterojunction solar cell consists of an interpenetrating network of an *n*-type (electron acceptor, e.g. fullerene derivatives) and a *p*-type (semi)conductor (electron donor, e.g. conjugated polymer), sandwiched between two electrodes with different work functions. The optical bandgap E_g is defined as the difference between the lowest unoccupied molecular orbital (LUMO) and the highest occupied molecular orbital (HOMO) of the absorber material.

We consider a 6-terminal triple-junction solar cell, consisting of three single organic photovoltaic cells (see Figure 2a for the schematic energy band diagram of a single cell). We assume that in each single cell, only one material absorbs light. Usually, most of the light is absorbed by the *p*-type component; this is the case we will consider here onwards. In the other case, when the *n*-type material absorbs all light, the results remain the same[13] by permutation of *n* and *p*. Because we assume full absorption in each subcell, we neglect interference and optical coupling of the subcells, thus overestimating the efficiency potential. The organic cell with the widest absorber bandgap is at top (at the side of the sun), thus $E_{g1} > E_{g2} > E_{g3}$. The distance between the HOMO of the *p*-type (donor) and the LUMO of the *n*-type (acceptor) is considered as the thermodynamic limitation of the useful energy[22]. We call this value the interface bandgap E_i. For an organic solar cell with ohmic contacts, the open circuit voltage V_{oc} is linearly dependent on the interface bandgap E_i. This linear relationship was proven for the variation of the HOMO level of the donor[14,23,24] and of the LUMO-level of the acceptor[25-27]. For a cell with non-ohmic contacts, the V_{oc} is dependent on the work function difference of the electrodes[28]. In these calculations, we assume a cell with ohmic contacts.

For our simulation, the following fundamental assumptions are made about the stacked triple-junction cell (Figure 1a): (i) every photon with an energy $h\nu$ higher than the bandgap E_{g1} is absorbed by the first cell and leads to a useful energy E_{i1}. This assumption implies that each absorbed photon eventually leads to a free electron and a free hole, with an energy difference of E_{i1} between them. (ii) every photon with an energy $h\nu$ between E_{g1} and E_{g2} is absorbed by the second cell and leads to a useful energy E_{i2}. (iii) every photon with an energy $h\nu$ between E_{g2} and E_{g3} is absorbed by the third cell

and leads to a useful energy E_{i3}. (iii) photons with an energy $h\nu$ lower than E_{g3} are fully transmitted. The maximum efficiency η_{max} is therefore given by:

$$\eta_{\max} = \frac{E_{i1}\int_{E_{g1}}^{\infty} N(E)\,dE + E_{i2}\int_{E_{g2}}^{E_{g1}} N(E)\,dE + E_{i3}\int_{E_{g3}}^{E_{g2}} N(E)\,dE}{\int_{0}^{\infty} E\,N(E)\,dE}, \quad with \;\; E_{g1} > E_{g2} > E_{g3} \qquad (1)$$

with $N(E)$ the incident photon flux. For all our simulations, we use the AM 1.5 experimentally measured solar spectrum[29]. Note that the denominator is the total incident photon power density of the solar spectrum and does not depend on any bandgap. In this ideal scenario, the open circuit voltage V_{oc} of the subcells will be given by E_{ij}/q (j=1,2,3) with q the electric charge. The fill factor FF of the subcells is assumed to equal unity, as well as the external quantum efficiency EQE of the first cell for wavelengths below the cut-off wavelength λ_{g1} (corresponding with E_{g1}, see Figure 2b). The EQE of the second cell equals unity for wavelengths between cut-off wavelength λ_{g1} and λ_{g2} (corresponding with E_{g2}) and similar conditions apply to the third cell. In real materials, however, the optical absorption and hence the EQE are confined to a more or less narrow wavelength range, usually about 200 to 300 nm wide. We idealize this behaviour by introducing the concept of absorption windows[13], which are defined in Figure 2b, and will be treated further as a parameter.

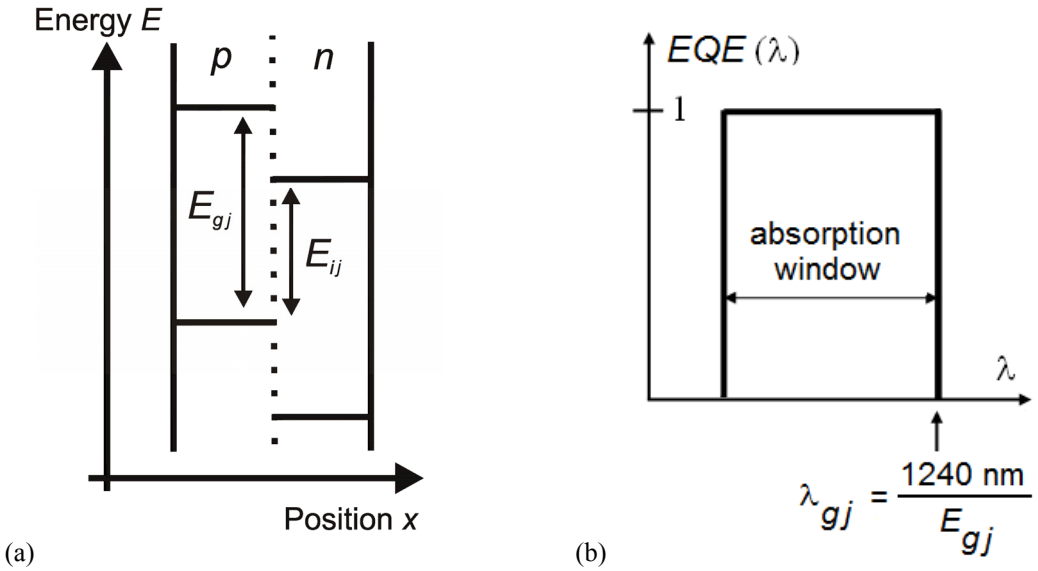

Figure 2. (a) The schematic energy band diagram of one subcell of a triple-junction organic solar cell (j=1,2,3). Only the p-material is the absorber. The mutual position of the single cells does not matter, because the cells are only optically and not electrically connected in series. The absorber bandgap E_{gj} and the interface bandgap E_{ij} of each subcell are indicated. (b) External quantum efficiency (EQE) as a function of the wavelength λ. Definition of the absorption window and the cut-off wavelengths λ_{gj} of one subcell (j=1,2,3). The absorption window of the second and third subcell maximally extends to the cut-off wavelength λ_{g1} respectively λ_{g2} of the other subcell. Notice that the order of the first and second subcell could be changed if there is no overlap between both absorption windows.

In a monolithic or integrated triple-junction solar cell (Figure 1b), the individual cells are electrically connected in series. This means that the total voltage over the cell is the sum of the voltages over each individual cell, and thus equals the sum of the interface bandgaps of the single cells. Furthermore, the same current flows through all single cells. Hence, the maximum efficiency η_{max} for a monolithic organic triple-junction cell is given by

$$\eta_{\max} = \frac{(E_{i1}+E_{i2}+E_{i3})\cdot \min\left(\int_{E_{g1}}^{\infty} N(E)\,dE,\ \int_{E_{g2}}^{E_{g1}} N(E)\,dE,\ \int_{E_{g3}}^{E_{g2}} N(E)\,dE\right)}{\int_{0}^{\infty} E\,N(E)\,dE}, \quad with \ \ E_{g1} > E_{g2} > E_{g3} \qquad (2)$$

with min(x,y,z) the minimum of x, y and z. The open circuit voltage V_{oc} of the whole monolithic cell will be given by $(E_{i1}+E_{i2}+E_{i3})/q$, the fill factor FF equals unity, as does the external quantum efficiency EQE for wavelengths below the cut-off wavelength λ_{g3}.

In organic bulk heterojunction solar cells, light absorption does not immediately lead to free charge carriers. Instead, an exciton is created. In an ideal scenario, the highest efficiency is reached when the LUMO of the *p*-material is as close as possible to the LUMO of the *n*-material[13]. However, a necessary condition for efficient dissociation of the created excitons is that the difference between the LUMOs of donor and acceptor (ΔLUMO) is higher than the exciton binding energy[30]. Thus, without a sufficient energy difference between the LUMOs of both materials, the solar cell cannot work. The value of the exciton binding energy (and the minimal ΔLUMO) in different materials is a subject of discussion, and values in a large range from 0.1 eV to 2 eV have been published[25,27,31,32]. The excess of this necessary minimum of the LUMO-difference corresponds with an energy loss. In earlier work[19], we showed how the optimum efficiency decreases for increasing ΔLUMO for different absorption windows. With a full absorption window, each additional difference of 0.1 eV between the LUMOs results in approximately an additional 10 % relative efficiency loss in the maximum attainable efficiency. In the following calculations, we assume a difference of 0.2 eV between the LUMOs of our organic solar cell. This value was put forward by us as an empirical threshold necessary for exciton dissociation[33]. Just because of this necessary energy difference between the LUMOs, the attainable efficiency for the organic bulk heterojunction solar cell drops by 16-17 % in comparison with their inorganic counterpart, purely because of the difficulties in exciton dissociation[19]. Choosing another value for ΔLUMO would lead to qualitative similar results.

In the next section, first we discuss the results for the case where both subcells of the triple-junction cell have a maximum absorption window. Second, we consider the case where both subcells have the same, narrow absorption window. Finally, a more realistic scenario is discussed.

3. RESULTS

3.1 Subcells with a full absorption window

Figure 3 shows the maximum efficiency in the ideal scenario for a stacked and monolithic organic triple-junction cell with bandgaps E_{g1}, E_{g2} and E_{g3}, a full absorption window for the subcells and a LUMO difference of 0.2 eV between *n*- and *p*-type. A maximum efficiency of 62.2 % and 61.0 % is reached for a stacked and monolithic cell respectively. As mentioned already, the efficiency of a monolithic configuration will never be higher than that of a stacked configuration. In comparison with a single junction organic cell, adding two subcells results in a relative gain of more than 50 % in power conversion efficiency[13]. If we compare the triple-junction with the tandem solar cell, adding a third subcell results in a relative gain of about 15 % in power conversion efficiency[19].

For higher bandgaps, less photons are being absorbed from the solar spectrum, but the useful output energy of each absorbed photon is higher. Hence, there is an optimum for each bandgap. For a single junction cell, the optimal bandgap is 1.1 eV. For a tandem cell, this maximum occurs for the stacked and monolithic tandem cell at a configuration (E_{g1}, E_{g2}) of (1.7 eV, 0.9 eV) and (1.6 eV, 0.9 eV) respectively[19]. Figure 3 reveals the optimum (E_{g1}, E_{g2}, E_{g3}) configuration for the triple-junction: (1.9 eV, 1.2 eV, 0.7 eV) and (1.8 eV, 1.2 eV, 0.7 eV) for the stacked and monolithic cell respectively.

The bandgap requirements for a close to optimal configuration of the stacked triplje-junction cell are quite broad, permitting that the values of the bandgaps for optimal cells are not that strict (Figure 3). This is not the case for the monolithic configuration; especially the value of the bandgap E_{g1} of the first subcell, and in a lesser extent E_{g2} of the second subcell are more critical than for a stacked cell. This can also be seen in Figure 4, where we plot the maximum efficiency for a fixed given bandgap; the other two bandgaps are treated as parameter and are chosen so that the efficiency is maximized. We see very clearly that for a stacked solar cell (Figure 4a), the requirements for E_{g1} and E_{g2} are

quite broad. However, for E_{g3}, this is not the case. A low bandgap E_{g3} is necessary for a good device. For a monolithic configuration (Figure 4b), the requirements are much stricter: all three bandgaps have only a small region wherein the power conversion efficiency of the solar cell is high.

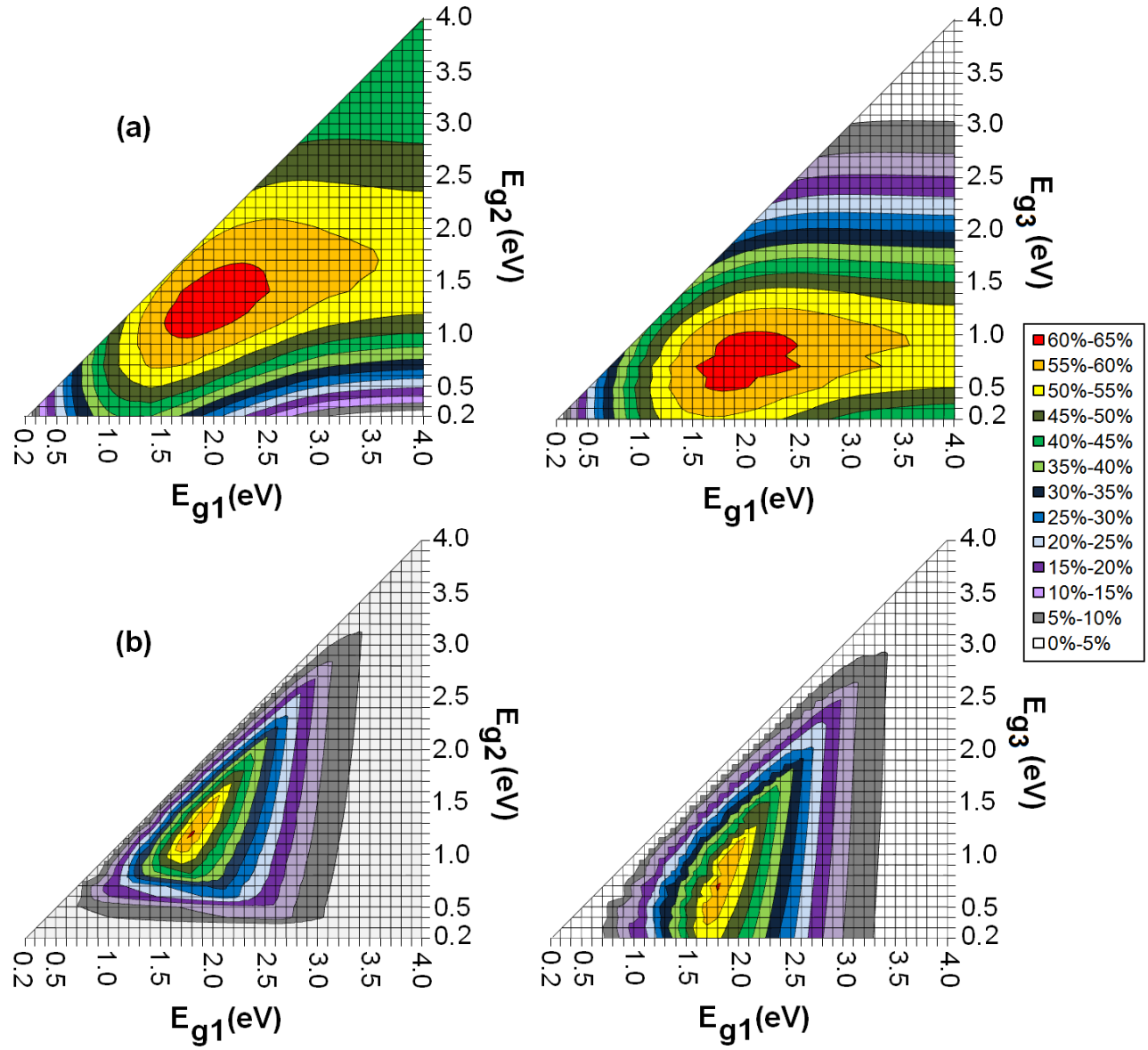

Figure 3. The maximum efficiency in the ideal scenario for a stacked (a) and monolithic (b) organic triple-junction solar cell with bandgaps E_{g1}, E_{g2} and E_{g3}, a full absorption window and a LUMO difference of 0.2 eV between *n*- and *p*-type.

Most organic absorbers have a wide bandgap and the production of suitable organic absorbers for photovoltaic applications with a narrow bandgap is problematic[4]. If we consider an organic cell with bandgaps E_{g1} = 2.5 eV, E_{g2} = 2.0 eV and E_{g3} = 1.5 eV, the stacked cell still has a maximum efficiency of 48.2 %, whereas the monolithic cell only reaches 33.7 %. We may conclude that a monolithic cell is much less efficient than a stacked cell in a non-optimal bandgap configuration. For an optimal bandgap configuration, however, the difference is negligible. This means that for the production of multi-junction cells, the choice of good bandgap combinations (and thus material combinations) is much more limiting for a monolithic configuration than it is for a stacked configuration.

The current extracted from the monolithic configuration is almost equal to the photocurrent of the subcell that generates the lowest current. If one of the subcells generates much more current than an adjacent subcell, the excess of charge

carriers cannot recombine at the intermediate contact between the subcells. This will cause a charging at the intermediate contact and will partially compensate the built-in voltage across the other cell until the current of both subcells matches. This will lower the power conversion efficiency and explains the inferior performance of monolithic cells for non-optimal bandgap configurations. Current matching is therefore necessary in a monolithic configuration. We want to stress that this effect is not implemented in the model presented in this paper.

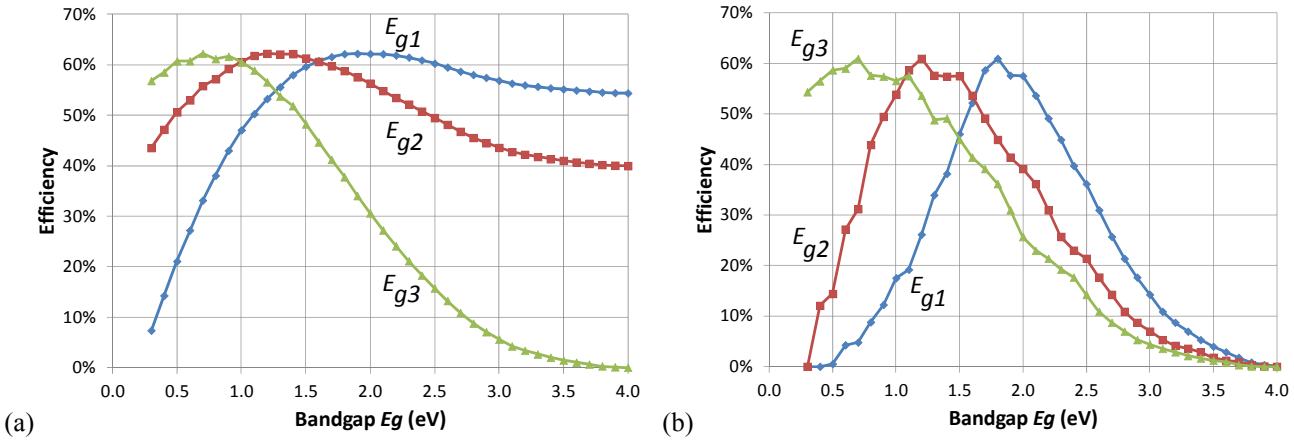

Figure 4. The maximum attainable efficiency for a fixed given bandgap in the ideal scenario for a stacked (a) and monolithic (b) organic triple-junction solar cell with bandgaps E_{g1}, E_{g2} and E_{g3}, a full absorption window and a LUMO difference of 0.2 eV between *n*- and *p*-type. If we keep one bandgap fixed, the other bandgaps are treated as parameter and are chosen so that the efficiency is maximized.

3.2 Subcells with the same (limited) absorption window

We now take into account the narrow absorption window which is characteristic for organic materials. For ease of presentation, we assume that all three subcells of the triple-junction structure have the same absorption window in nm.

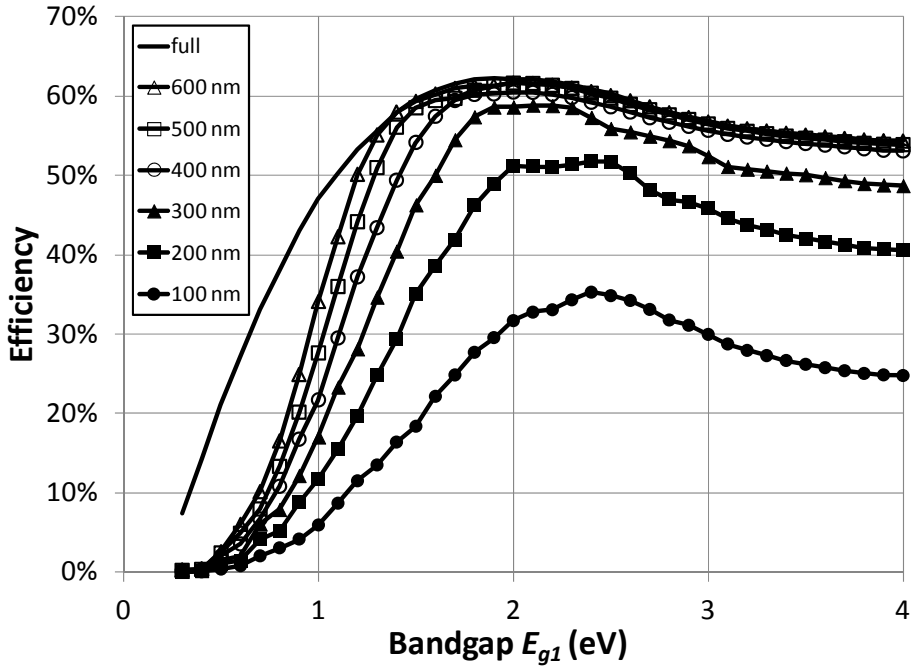

Figure 5. The maximum efficiency in the ideal scenario for a stacked organic triple-junction solar cell with varying bandgap E_{g1}, optimal chosen bandgaps E_{g2} and E_{g3}, and a LUMO difference of 0.2 eV between *n*- and *p*-type. The absorption window varies from full absorption width to 100 nm absorption width.

First, as example, we consider the maximum efficiency for a stacked organic triple-junction solar cell with varying bandgap E_{g1}, optimal chosen bandgaps E_{g2} and E_{g3} and a LUMO difference of 0.2 eV between *n*- and *p*-type (Figure 5). We vary the absorption window from full absorption width to 100 nm absorption width. Notice that the full absorption width can also be found in Figure 4a. We notice two important results. First, the efficiency remains quite high, until the absorption width decreases under 300 nm. Secondly, the smaller the absorption window, the higher the optimal bandgap E_{g1} of the first cell.

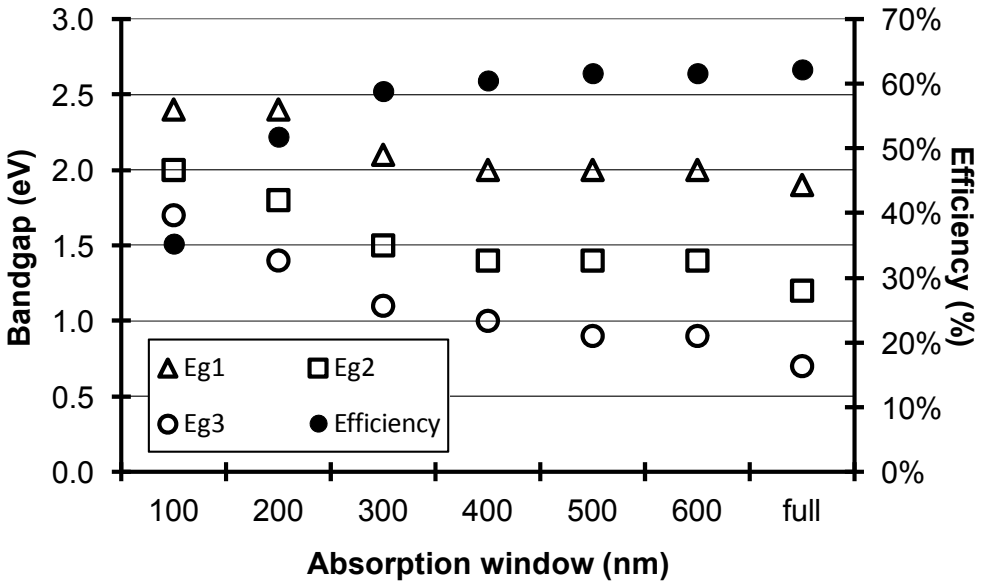

Figure 6. The maximum efficiency is plotted in the ideal scenario (with ΔLUMO = 0.2 eV) for an organic stacked triple-junction cell as a function of the absorption window (from 100 nm absorption width to full absorption width). Also the optimum bandgaps E_{g1}, E_{g2} and E_{g3} for the stacked cell are plotted as a function of the absorption window.

A more general view can be found in Figure 6 which shows the maximum efficiency η_{max} and the optimum bandgaps E_{g1}, E_{g2} and E_{g3} in the ideal scenario for a stacked configuration as a function of the absorption window width. The broader the absorption window, the higher the efficiency. The power conversion efficiency of the monolithic configuration is only a negligible bit lower than the stacked structure, because we consider here the optimal bandgap configuration. As explained above, the efficiency for non-optimal bandgap configurations of the monolithic cell will be much lower than for the stacked cell. In the case of non-optimal bandgaps, the advantage of the stacked solar cell over the monolithic cell decreases for increasingly smaller absorption windows.

Figure 6 shows that the optimum bandgap of the cells shifts towards higher energies for lower absorption windows. For example, the optimum bandgap shifts from E_{g1} = 1.9 eV, E_{g2} = 1.2 eV and E_{g3} = 0.7 eV for a full absorption band to E_{g1} = 2.4 eV, E_{g2} = 1.8 eV and E_{g3} = 1.4 eV for a cell with an absorption window of only 200 nm. This is a satisfying result, because, as we already mentioned, the production of suitable low bandgap organic materials is difficult. For an absorption window of 300 and 400 nm respectively, already 95 and 97 % of the maximum attainable efficiency (for a full absorption band) is reached. Hence, it would not pay off to try to develop organic materials for triple-junction solar cells with an absorption window broader than 300 to 400 nm, because hardly any efficiency gain can be achieved by widening the absorption window further. The optimum bandgaps with a sufficient absorption window of 300 nm are E_{g1} = 2.1 eV, E_{g2} = 1.5 eV and E_{g3} = 1.1 eV.

3.3 The maximum obtainable efficiency in a more realistical situation

To estimate the maximum obtainable efficiency in a less ideal situation, we assume a more realistic scenario. We consider an absorption window of 300 nm wide for both subcells, an *EQE* of 65 %, a fill factor *FF* of 60 %, and a voltage factor *f* of 60 %, with *f* defined by:

$$f = \frac{q.V_{oc}}{E_{gi}} \tag{3}$$

We consider that all subcells of the triple-junction structure have the same *EQE*, *FF* and *f*. This results in a maximum attainable efficiency of 14 % and 13 %, respectively, for stacked and monolithic organic solar cells, both at an optimal configuration (E_{g1}, E_{g2}, E_{g3}) of (2.1 eV, 1.5 eV, 1.1 eV).

4. CONCLUSIONS

A maximum attainable efficiency of 62.2 % and 61.0 % , respectively, is reached in our ideal model for an organic stacked and monolithic triple-junction cell at a bandgap configuration of (1.9 eV, 1.2 eV, 0.7 eV) and (1.8 eV, 1.2 eV, 0.7 eV), taken into account the necessary ΔLUMO difference for exciton dissociation. This is a relative gain of more than 50 % when compared with a single junction organic cell, and a 15 % relative gain compared with a tandem configuration. The requirements for a close to optimal configuration of the stacked cell are quite broad. This is not the case for the monolithic configuration. We found and quantified that a monolithic cell is much less efficient than a stacked cell for a non-optimal bandgap configuration. We also studied the influence of the narrow absorption window on the efficiency. The optimum bandgap of the cells shifts towards higher energies for lower absorption windows. Ideal material characteristics are obtained from these calculations, giving an idea of how the ideal organic triple-junction cell should look like. An interesting result is that it would not pay off to try to develop organic materials with an absorption window broader than 300 nm because hardly any efficiency gains can be achieved by further broadening the absorption window. The optimal bandgaps with a sufficient absorption window of 300 nm are E_{g1} = 2.1 eV, E_{g2} = 1.5 eV and E_{g3} = 1.1 eV.

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