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Can up- and down-conversion and multi-exciton generation improve photovoltaics?

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ABSTRACT

Photon up-conversion (UC) and photon-induced multiple-exciton generation (MEG) are proposed directions that are of increasing interest for improving photovoltaic (PV) conversion efficiencies via "photon (or light) management". Straightforward analysis of these approaches for non-concentrated single-junction cells in the detailed balance limit yields a theoretical PV conversion limit of 49%, instead of 31% without UC and MEG. With what we estimate to be optimistic, maximal realistic efficiencies (25% for UC; 70% for MEG) this limit becomes $<$40%, i.e., ~1.25 times the theoretical efficiency of conventional single-band gap cells. While this result does not detract from the fascinating fundamental scientific challenge to make UC and MEG simple and cheap ways to improve PV, such reality checks should be considered when evaluating the short-term promises of these and other options, such as spectral splitting and tandem arrangements.

1. Introduction

The renewed interest in renewable energy sources has brought solar energy options and expectations back into the limelight. However, many obstacles remain for truly widespread exploitation, especially if we consider direct conversion of sunlight into electricity. There, the (cost)/(efficiency $\times$ lifetime $\times$ production yield) ratio for present photovoltaic (PV) devices is still several times too high for practical very large-scale use of the type that can compete in an open market with conventional power stations. There is, therefore, an enormous interest in finding ways that can radically decrease the above-mentioned ratio.

The various mechanisms that lead to the conversion losses in a PV cell and that limit its efficiency can be seen in Fig. 1, which is adapted from an original analysis by the late Martin Wolf for crystalline Si cells [1]. The most noticeable loss mechanism in non-thermal solar energy conversion relates to the fact that the basic electronic excitation process in PV (and also in photochemical processes, as well as in photobiological ones such as photosynthesis) is a threshold one. All photons with energy below the threshold energy for optical absorption (in the case of most PV cells, the semiconductor band gap) cannot be converted into electrical energy. Another portion of the solar energy that is lost is the fraction of the energy of photons with energies higher than the threshold that is beyond the threshold energy. Hitherto, attempts to find ways to transform also this excess energy into electrical, rather than thermal energy (through the creation of phonons) has not really been crowned with success, and this area is, therefore, one of great current interest.

In the early 1960s Shockley and Queisser [2] introduced the detailed balance limit of conversion efficiency for conventional semiconductor single-band gap solar cells. They calculated, in addition to the above-mentioned photon energy loss mechanisms, the thermodynamically unavoidable losses. For the physics that leads to the latter losses we refer to the original paper [2], as well as to some more recent literature [3]. Here we note that it is general to all devices that convert sunlight directly into electricity. While the highest ultimate efficiency was found to be ~44%, the detailed balance limit yields a maximum of ~31% efficiency for a ~1.4 eV threshold/band gap. The ubiquitous Si solar cell received thorough analysis by others [4] and has a maximum efficiency of nearly 29%. The loss mechanisms preventing current state-of-the-art Si solar cells from reaching their theoretical limit have been studied thoroughly and can be found in, for example, Ref. [5].

To be able to go beyond these theoretical limits several approaches have been suggested [6–8]. Perhaps the best-known ones are those that forego the single-band gap approach and use an arrangement of several materials with different band gaps. In the tandem cell several solar cells with decreasing band gaps are stacked on each other [9]. Earlier and also recent (e.g., DARPA-sponsored) efforts focus on spatially separated cells with different band gaps, which are illuminated by different parts of the solar spectrum, obtained from the incident sunlight by spectral splitting [10]. Careful choice of the band gaps can

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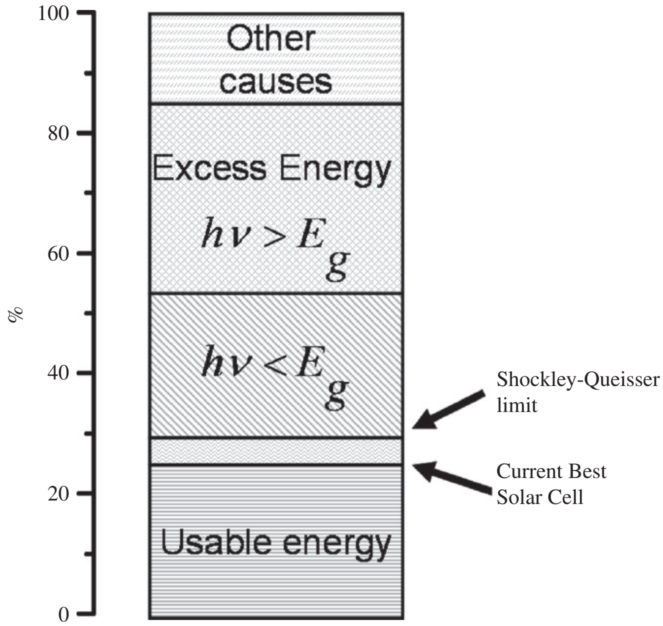

Fig. 1. Illustration of the various loss mechanisms that limit the efficiency for state of the art Si solar cells [41] (adapted from Ref. [1]).

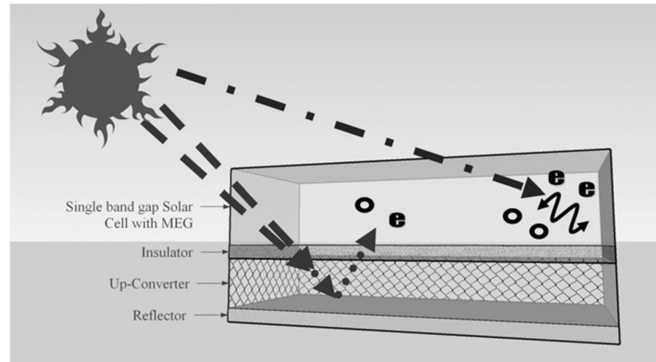

Fig. 2. Illustration of a proposed scheme for combining up-conversion (UC) and multi-exciton generation (MEG). One photon with energy $h\nu \geqslant 2E_g$ (dash dotted beam), yields two (or more) electron–hole pairs with energy E_g via MEG. Excess energy is expressed as phonons (spring-like entities). For UC, two photons with energy $\frac{1}{2}E_g \leqslant h\nu < E_g$ (two dashed beams) create one photon with $h\nu \geqslant E_g$ (dotted beam). This photon will be reflected back to the semiconductor, where it can now create an electron–hole pair.

improve significantly the efficiency that can be obtained for a given solar spectrum. In addition to these, we note among other methods those that aim at minimizing thermodynamic losses using light concentrators and those that help minimize shading by contacts, spurious absorption and reflection (the loss box at the top of Fig. 1).

Often-proposed directions to improve photon management are up-conversion (UC) and down-conversion of photon energy. In UC [11], two photons with energy $\frac{1}{2}E_g \leqslant h\nu < E_g$ (where E_g is the semiconductor band gap) create one photon with $h\nu \geqslant E_g$, while in down-conversion [12] one photon with energy $h\nu \geqslant 2E_g$, yields two photons with energy $h\nu \geqslant E_g$.

A different conceptual way to make more efficient use of high-energy photons is the use of multi-exciton generation [13,14] (MEG) where one photon with energy $h\nu \geqslant nE_g$ yields up to n electron–hole pairs with energy E_g. MEG has clear advantages over down-conversion since MEG can create numerous electronic excitations while down-conversion can create only two photons. For this reason in the following we will, instead of considering both down-conversion and MEG, consider only MEG, even though, strictly speaking it is not a "photon management" process.

While UC and MEG are two approaches that are usually referred to separately, one can relatively easily combine these two methods, as is explained below.

In Fig. 2, we show schematically how in UC two photons with energy $\frac{1}{2}E_g \leqslant h\nu < E_g$ (illustrated by two dashed beams) that are not absorbed in the semiconductor (because their energy is below the band gap) pass to the up-converter. There, through a process such as non-linear sum-frequency generation (but see also below), the two photons create one photon with $h\nu \geqslant E_g$ (dotted beam). This photon will be reflected back to the semiconductor, where it can now create an electron–hole pair.

In MEG one photon with energy $h\nu \geqslant 2E_g$ (dash dotted beam), yields two (or more) electron–hole pairs with energy E_g. The excess energy above $2E_g$ is expressed as phonons (spring-like in the figure). MEG is not limited to yielding two electron–hole pairs, and there is evidence of up to 7 excitations from a single photon with sufficiently high energy [15]. Thus, in theory, for a single-junction cell with $E_g = 1$ eV, all photons with $0.5\,\text{eV} \leqslant h\nu < 1$ eV can participate in UC and all photons with $h\nu \geqslant 2$ eV can create multiple-electron–hole pairs via MEG.

For UC no complex layering is needed (unlike in multiple-band gap tandem cells), because there is no need for electrical transport in the up-converter. In fact, to prevent any unwanted electrical transport between the semiconductor and the up-converter, an insulator can be inserted to block any such possibility. This configuration allows optimizing the optical properties of the up-converter separately from any requirements of the cell itself, a very useful basic engineering approach.

While these approaches of "photon management" appear at the forefront of efforts to arrive at radical changes in PV efficiencies [8,16], we could not find an analysis of what are the maximum possible gains that we can hope for from the combination of UC with MEG. Some years back we analyzed the combination of up- and down-conversion [17] as was also done more recently by Conibeer et al. [18], but as noted above MEG is the more general and flexible approach. It is, therefore, intrinsically a more attractive process than down-conversion, except for the above-mentioned optimization problem as a down-converter can be optimized separately from a cell, which is not possible for MEG. We performed such analysis and will now assess in a realistic and optimistic way the promise of such systems to future PVs.

Our analysis can serve as a basis for the maximum expected efficiency from the "photon management" approach, namely manipulating the photon energy before the PV conversion process. Such analysis can then be compared, in terms of short- and long-term promises with the "photon splitting" and tandem approaches, where photons of different energy ranges reach different solar cells. Such a comparison should help the quest for making very large-scale use of PV systems a reality.

2. Calculations

We calculate the ultimate efficiency (Fig. 3) and the detailed balance limit (Fig. 4) for a single-band gap semiconductor PV system (without UC or MEG), for one with MEG alone (no UC), for one with UC alone (without MEG) and for UC and MEG combined. All calculations (except calculating the number of photons and electron–hole pairs for UC and MEG) follow Shockley and Queisser [2] and here we show merely the outline of their calculations for clarity.

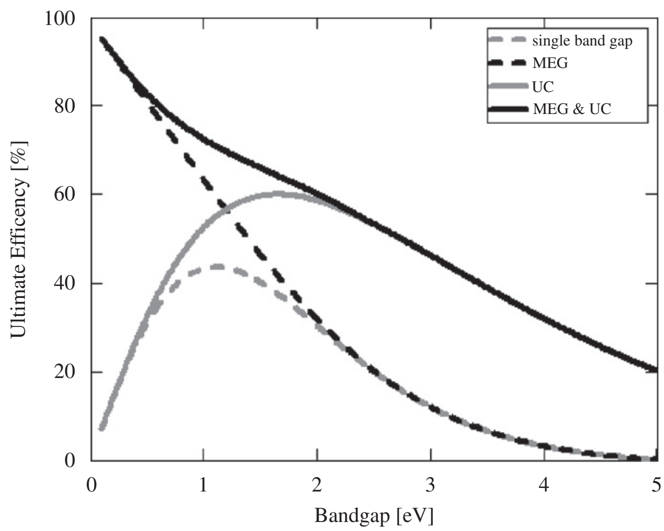

Fig. 3. Ultimate efficiency (ignoring detailed balance considerations) as a function of semiconductor band gap, assuming 100% MEG and UC efficiencies.

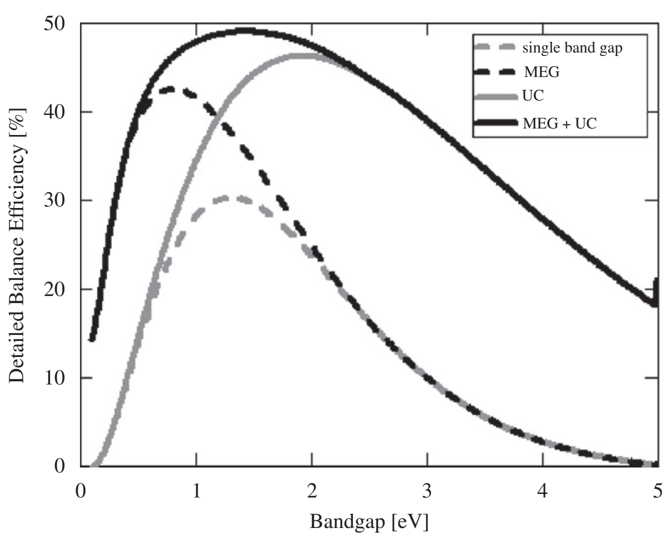

Fig. 4. Detailed balance efficiency as a function of semiconductor band gap assuming 100% MEC and UC efficiencies.

For all the calculations the cell temperature was set to 300 K and f_w (the fraction of the solid angle about the sphere subtended by the sun) was set to 2.18×10^{-5}. The incoming solar radiation is taken as that of a 6000 K black body and the number of photons with different energies is in accordance with the formulae of the Planck distribution.

The ultimate efficiency μ is defined as

$$\mu = \frac{h\nu_g Q_s}{P_s} \tag{1}$$

where $h\nu_g$ is the energy needed to produce one electron–hole pair, Q_s is the number of photons with energy higher than $h\nu_g$ and P_s is the total incident power.

The detailed balance limit efficiency η is defined as

$$\eta = t_s \mu \xi m \tag{2}$$

where t_s is the probability to produce an electron–hole pair from a photon with energy $h\nu_g$ (or higher) and is set to unity, ξ is the ratio between the open circuit voltage to the energy gap and m is the impedance matching factor.

For UC we assume

$$2\left(\tfrac{1}{2}E_g \leqslant h\nu < E_g\right) \Rightarrow 1(h\nu \geqslant E_g) \tag{3}$$

and, thus, the total number of photons with a minimum energy of $h\nu_g$ is

$$Q_{UC} = \tfrac{1}{2} Q_{(1/2)E_g \leqslant h\nu < E_g} + Q_{E_g \leqslant h\nu} \tag{4}$$

For MEG we assume

$$1(h\nu \geqslant nE_g) \Rightarrow n(e^- + h^+) \tag{5}$$

where e^-+h^+ refers to an electron–hole pair and n is an integer. The total number of electron–hole pairs produced is

$$Q_{MEG} = \sum_{n=1}^{\infty} nQ_{nE_g \leqslant h\nu < (n+1)E_g} \tag{6}$$

For UC and MEG occurring together, the total number of electron–hole pairs produced is

$$Q_{UC+MEG} = \frac{1}{2} Q_{(1/2)E_g \leqslant h\nu < E_g} + \sum_{n=1}^{\infty} nQ_{nE_g \leqslant h\nu < (n+1)E_g} \tag{7}$$

These calculations assume 100% efficiency plus a $2E_g$ onset for MEG, and 100% efficiency for UC.

3. Discussion

It is instructive to look at the ultimate PV conversion efficiency calculations in Fig. 3. This scheme, which does not include detailed balance considerations, is misleading in terms of possible efficiencies. A single-band gap solar cell shows a ~44% maximum PV conversion efficiency at ~1.1 eV band gap. With UC we find a PV conversion efficiency of roughly 60%. Including MEG leads to a sharp increase towards an unrealistic 100% PV conversion efficiency as we decrease the semiconductor band gap. This is because "carrier cooling" losses in the MEG process are assumed to be less than the band gap (if they were higher than the band gap they should have produced an additional electron–hole pair) and as the band gap approaches zero those losses approach zero as well.

When examining the detailed balance limit in Fig. 4 it is interesting to see where are the highest PV conversion efficiencies for the different schemes. While for simple single-band gap PV cells the maximum PV conversion efficiency is for semiconductors with a band gap around 1.4 eV, if we include UC, the band gap for maximum PV conversion efficiency shifts to ~1.8 eV. With MEG the highest conversion efficiency is with a band gap around 0.9 eV. We find that combining MEG with UC yields as band gap for most favorable performance again a value of ~1.4 eV. We note that much research and development has already been done in optimizing semiconductors with band gaps around this value, the optimal one in terms of single-band gap PV conversion efficiency at air mass 1, which may well be advantageous for meeting the materials challenges for efficient MEG.

The calculated efficiencies for MEG and UC separately are similar to those found previously [11,12,19]. Our original finding is that the maximum PV conversion efficiency for 100% efficient UC together with MEG processes is 49%, a maximal gain over the 31% conversion efficiency for a single-band gap solar cell of a factor of 1.58. It is important to note that if we use AM0, AM1 or AM1.5 solar spectra (instead of the 6000 K black body radiation spectrum), this does not significantly change the maximal gain achieved and might even slightly decrease it (for a silicon cell), because the portion of extra energy available for MEG (and for down-conversion) drops [20].

More interesting than what is the gain with unrealistic 100% efficient UC and MEG is what is the gain for realistic cases. When trying to gauge what is realistic we should keep in mind that making systems with efficient UC and/or MEG presents an enormous challenge for materials researchers for reasons we will discuss below.

3.1. Up-conversion

State-of-the-art, in-cavity, laser frequency doublers can show efficiencies of up to ~70%. This efficiency is achieved for monochromatic sources that are uni-directional (with one phase-matched direction for the incident and frequency doubled beam) and with the frequency-doubling crystal placed *inside* the laser cavity. Best extra-cavity converters reach efficiencies of 20–25%. A broad-band up-converter (for, say $0.5E_g \leqslant h\nu < E_g$) with polychromatic, non-directional light source direction (the sun) cannot possibly show efficiencies even approaching 70% because manufacturing an optical active material that does not show significant dispersion over a wide spectral range contradicts the Kramers–Kronig relationship [21]. The reason is that the changes in the real and imaginary parts of the dielectric constant are interconnected (they must be an analytical function of any physical parameter). The total dielectric constant must be a smooth function, meaning that a strong change in the real part will lead to a concomitant change in the imaginary part, which implies dielectric loss, i.e., optical absorption.

An additional physical limitation is posed by the fact that practical UC as used today [22], relies on a non-linear process. Generally, a system with high non-linearity over a wide spectral range and light intensity range must show large loss, and correcting this problem in one part of the spectrum will make the situation worse in other parts of the spectrum. Therefore, since UC efficiency is a strong function of intensity, optimizing the material for a certain light intensity makes it inefficient for others. In addition, efficient UC is usually (though not always [23]) achieved at very high light intensities, which raises the question of long-term material stability against radiation damage.

The need to provide phase matching for multi-directional broad-band illumination adds additional constraints to the up-converting medium. In light of these restricting factors we suggest that a 25% efficient broad-band converter is a very high upper limit for total efficiency of broad-band up-converters, one that we actually do not know how to reach today, but that may be achieved by some new approaches, such as the one described in Refs. [23,24]. The "non-coherent up-conversion" approach that is described there is *not* based on a non-linear mechanism. Therefore, it does not require phase matching, which is its greatest advantage. However, one can anticipate that the efficiency of this mechanism must strongly depend on the light intensity because the occupancy of the intermediate levels and the energy loss due to non-radiative transitions depend on the intensity, because the radiation absorption and emission coefficients change with the occupancy. Therefore, 50% efficiency (maximum occupancy of the intermediate levels under excitation) seems to be an absolute theoretical limit of this approach. Because this fundamental limit does not include other losses that will be part of the process, such as competitive de-excitation channels and some stabilization energy of the intermediate molecular electronic level, our optimistic estimate of 25% is quite generous for this process, too.

3.2. Multiple-exciton generation

Creating more than one electron–hole pair from one photon, as in MEG, carries an energy penalty [25,26], in addition to that paid in the normal PV process. Multi-(electron–hole) pair generation from a single photon is, naturally, well-known in avalanche photodetectors, for, e.g., X- and γ-ray detectors. Auger recombination in quantum-confined systems suitable for MEG [27–29], and hot carrier relaxation must also be taken into account. While inhibiting or at least decreasing carrier relaxation is possible by surface modification of quantum-confined systems to trap the holes, such modification must be made in such a way that collection of the carriers, will not be impeded.

Assuming efficient carrier multiplication via MEG has been achieved [30], we need, to make MEG useful for PVs, to

(a) separate the electron–hole pairs and
(b) to collect them efficiently.

While there are some remarkably encouraging recent results [31], there is little question that these are difficult issues, especially for quantum-confined systems. The issues are well-known in organic solar cells and, indeed, present major obstacles to improving them so as to have them somewhat approach theoretical efficiencies [32].

Last but not least is the issue of the threshold for impact ionization. Theory teaches that this depends on the ratio between the effective mass of the electrons and the holes. Theoretical analyses show that for known semiconductors the minimum energy needed a quantum yield >1 is $\sim 2.2E_g$ [33,34]. In practice, no cases are known as yet that show carrier *doubling* (more than two electron–hole pairs/photon) at less than $3E_g$ photon energy. For the extensively studied electron beam-induced electron–hole pair generation processes (e.g., for EBIC) the empirical relation, admittedly only in bulk materials, is $E_e(\text{incident}) = 2.1E_g + 1.3\,\text{eV}$ [35].

Therefore, MEG is expected to be constrained by the following long-investigated but so far unresolved issues:

(a) competing (de)excitation processes;
(b) recombination of excitons without carrier separation;
(c) high-impact ionization threshold.

In light of these problems a 70% MEG efficiency is a very generous estimate, and it is not at all clear how we can reach it *with relevant materials*.

With these general upper limits for any scheme of MEG and UC, the maximum PV conversion efficiency for a combined multi-exciton generating and up-converting system reaches ~38% (Fig. 5). This is a gain of a factor of 1.23 over the optimal theoretical detailed balance efficiency for a conventional single-band solar cell. Taking into account the overhead we must pay for integrating MEG and UC in our PV conversion system a gain of less than 25% is food for thought as reality check.

It is also instructive to compare MEG to the tandem approach [36,37], and to that of spectral splitting for multiple-band gap solar cells [10]. The reason is that, if we optimize use of the solar spectrum, the multiple-band gaps need not and normally will not be integer multiples of each other, in contrast to what is the case with MEG, which, thus, will mostly be inferior to spectral splitting and tandem approaches. While the latter is indeed very expensive, it would appear that spectral splitting can be done at relatively low costs and, thus, is the approach to which MEG should be compared. If we recall that the "photon splitting" concept shows a 42% Shockley–Queisser limit for a (1.9+1.0 eV) two band gap solar cell [36,38] (and a higher one for more than 2 bandgaps), another factor should be considered. The problems that beset the "photon splitting" concept are rather well-known basic engineering and production ones [37]. At the same time the "photon management"

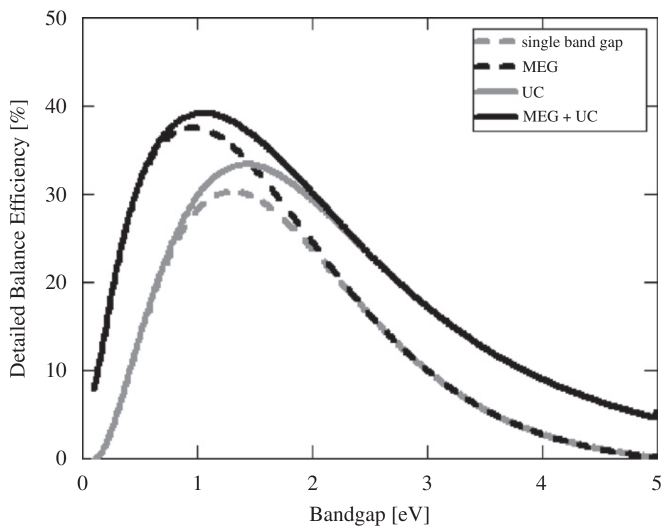

Fig. 5. Detailed balance efficiency as a function of semiconductor band gap assuming 70% efficient MEG and 25% efficient UC.

concepts that we discussed here still need significant basic scientific breakthroughs before they reach the phase of having to overcome "mere" engineering obstacles. It would appear that this distinction is not always made clear when future possibilities are reviewed and predictions of future developments are made.

None of the above should be construed as a reason to forego the fascinating fundamental research in these and related areas. Rather, we make a plea for making a clearer distinction between *long-range* basic research and work that can realistically be considered to lead to practical, tangible results in the *short-range* [39].

Making the public aware of the difference between much-needed long-term basic research and short-term solutions can help us avoid creating unrealistic levels of expectations that, often, backfire [40]. This is a message that life science at large has succeeded to convey to the public in its quest for finding cancer cures and there is no reason why the alternative energy research community in general and the solar cell one in particular, should not be able to do so.

4. Conclusions

We find a maximum possible PV conversion efficiency for 100% efficient UC plus MEG of 49%, a gain of 1.58, compared to a simple single-band gap solar cell. Taking into account what we estimate to be very optimistic upper limits to efficiencies for broad-band UC and MEG creation (25% and 70%, respectively), we find a maximum PV conversion efficiency of ~38%, a 1.23 gain over that for a simple single-band gap solar cell. Therefore, for UC, MEG (or down-conversion) or UC+MEG to yield the type of *radical* decrease in the (cost)/(efficiency × lifetime × production yield) ratio that is needed to allow large-scale economic introduction of solar cells presents a truly formidable challenge, the size of which appears not to be truly appreciated in many future scenarios for PV. Serious, long-term, continuing world-wide support for fundamental research is needed here and, as is often the case, may well bear fruit (also) in directions different from those described here.

Acknowledgments

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