

An approach to downconversion solar cells

Mark B. Spitzer^{b,*}, Hans P. Jenssen^a, Arlete Cassanho^a

^a AC Materials, Inc., Tarpon Springs, FL 34689, United States

^b Photonic Glass Corporation, 661 Pleasant Street, Norwood, MA 02062, United States

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ABSTRACT

We describe an approach to solar downconversion in which a semiconductor is used for absorption, and substitutional rare earth ions are used for emission. The semiconductor would provide broad-band absorption, and the rare earth ions would provide emission of multiple photons by a cross relaxation process in a narrow wavelength band. The emitted photons are then absorbed by a solar cell formed from a low band gap semiconductor such as silicon. Er and Yb are suggested owing to strong emission at 980 nm which are useful when the down-converter is paired with silicon solar cells. The use of InGaN is proposed for the absorbing host semiconductor.

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1. Introduction

The last few years have seen renewed interest in methods to break the Shockley–Queisser limit [1] to the energy conversion efficiency of single-junction solar cells [2,3]. The Shockley–Queisser limit is a consequence of detailed balance which assumes that for a single absorber of energy band gap E_C , each photon with energy greater than E_C generates not more than one photon, and that photons with energy less than E_C generate no photocurrent.

Although current practice in space solar cells is to use multi-junction (tandem) solar cells to overcome this limit, there may still be advantages to single-junction cells if the efficiency can be raised beyond the Shockley–Queisser limit. Possible advantages may be reduced cost, reduced weight, elimination of current-matching issues and improved radiation tolerance. Since such cells have not yet been made and these advantages are speculative; it is nevertheless interesting to examine whether such a cell could be fabricated. In this paper we consider how such cell might be designed.

The keys to developing a single-junction cell with conversion efficiency greater than the Shockley–Queisser limit are: (i) to combine sub- E_C photons into one photon than can be absorbed (upconversion) or (ii) to split high energy photons into multiple photons that each have energy greater than E_C (downconversion). An excellent review may be found in Ref. 4.

Upconversion has been the subject of a number of investigations that have shown small gains in efficiency by placing an up-converter behind a bi-facial Si solar cell [2,3]. When placed behind

the cell, the up-converter does not interfere with absorption of photons with energy greater than E_C . A down-converter must be placed in front of the cell and for this reason, great care must be taken to maintain the cell's short circuit current (J_{sc}). We are not aware of an integrated solar cell and down-converter that has yielded improved J_{sc} but as we will discuss, we believe that such a cell is possible.

The approach discussed here is different than the intermediate band solar cell [5,6] which has shown promise for upconversion via intermediate states in the energy band gap of the absorbing semiconductor within the solar cell. In such cells, recombination via these intermediate states has the potential to interfere with minority carrier transport and may lead to increased dark saturation current. Our approach is similar to Shalav's ([2,3] because the up or downconversion process is separated from the photovoltaic process and consequently does not interfere with photogenerated carrier collection.

In this paper we will be concerned primarily with down-conversion. We make a distinction here between down-shifting and downconversion. By down-shifting, we mean a process in which a single photon is absorbed at a short wavelength followed by emission of a single photon at a longer wavelength. While down-shifting may improve cell efficiency if the solar cell quantum efficiency is weak at short wavelength, it cannot break the Shockley–Queisser limit because down-shifting does not change detailed balance. By downconversion we mean a process comprising the absorption of one photon followed by the emission of more than one photon, and therefore downconversion changes detailed balance. If both photons emitted in downconversion are to be absorbed by the solar cell, the original photon must have energy greater than twice the band gap of the absorbing semiconductor. For Si cells, the wavelength range of interest is 300 nm–560 nm.

* Corresponding author. Tel.: +1 781 492 9200; fax: +1 781 551 9801.
E-mail address: mbspitzer@photonicglass.com (M.B. Spitzer).

2. Er downconversion materials

Adding and splitting of photons can be attained by using cooperative nonradiative resonant energy transfer between nearby rare earth ions. These processes obtain owing to the unfilled states in the inner 4 f shell [6]. The lanthanides provide many inner shell states with transitions at optical wavelengths [7].

Er is a good candidate for downconversion. When Er substitutes for Y in fluoride crystals such as NaYF_4 , transitions can occur among inner shell states at energies in the desired optical range. Nonradiative recombination is low because such transitions require a phonon, and in the yttrium fluoride based crystals, the phonon spectra does not provide sufficient energy to support the larger transitions (such as $^4\text{I}_{13/2} \rightarrow ^4\text{I}_{15/2}$), therefore except for small transitions, the decay is largely radiative and the lifetime in the excited state is on the order of 1 ms.

In Er-doped materials, the absorption and re-emission process begin with absorption of a photon by the excitation of an electron from the $^4\text{I}_{15/2}$ ground state to an excited state such as for example the $^4\text{F}_{7/2}$ state. The absorption may also proceed by excitation to a higher energy state such as the $^4\text{F}_{5/2}$ followed by nonradiative decay to a nearby lower state, or in some cases excitation to a nearby higher state through phonon interactions, followed in some cases by radiative decay to the ground state.

Downconversion occurs when energy is exchanged *non-radiatively* between nearby Er ions. Fig. 1a shows a simplified energy diagram in which some 4 f states have been omitted for clarity. One possible downconversion process begins by excitation of an electron to the Er $^4\text{F}_{7/2}$ state. The excited electron may then relax to the $^4\text{I}_{11/2}$ state via non-radiative energy exchange, by contributing energy to a nearby electron in an Er $^4\text{I}_{15/2}$ ground state, thereby exciting this nearby electron from the Er $^4\text{I}_{15/2}$ ground state to the Er $^4\text{I}_{11/2}$ state. In this cross relaxation process, nonradiative energy transfer results in conversion of the energy of one photon to the energy of two excited electrons in $^4\text{I}_{11/2}$ states. These electrons return to the ground state by emitting two photons with wavelength of approximately 980 nm.

The density of states associated with the 980 nm emission can be increased by adding Yb, which has only one level ($^2\text{F}_{5/2}$) and the energy of this level is approximately the same as the Er $^4\text{I}_{11/2}$ state [7]. Cross relaxation between Yb and other lanthanides is well-known [6]; the addition of Yb enhances the probability of a transition that emits at 980 nm. Fig. 2b shows how Er and Yb may cooperate in downconversion. In this case the excited electron

cross relaxes to the Er $^4\text{I}_{11/2}$ state by nonradiative energy transfer to an electron in the Yb ground state, thus exciting the electron to the Yb $^2\text{F}_{5/2}$ level. The two electrons decay to their ground states by emitting two photons at 980 nm.

3. Downconversion results

Er-doped KY_3F_{10} and BaY_2F_8 samples were grown by the Czochralski method, and were sliced and polished. The sample thickness is 1 mm. The transmission of these samples was measured and data for KY_3F_{10} are shown in Fig. 2. The absorption lines can be associated with known Er 4 f levels [9]. The transmission data reveal several problems with the use of such crystals for downconversion. Note first that reasonably thick (> 1 mm) samples are needed to absorb fully the wavelengths associated with these levels, meaning that thin Er-doped films may not be useful for absorption.

A second problem with the use of Er-doped KY_3F_{10} or similar crystals for either up or downconversion that is evident in Fig. 2 is that Er-doped KY_3F_{10} will only absorb in specific narrow bands, whereas worthwhile downconversion requires that all photons in the desired band be absorbed and converted. A further problem with the use of Er-doped KY_3F_{10} , particularly on the front of a solar cell, is the parasitic absorption at 650 nm and 800 nm.

We have also examined the photoluminescence (Fig. 3) of KY_3F_{10} and BaY_2F_8 by pumping with an Ar laser (488 nm). The strong signature of the Er $^4\text{I}_{11/2}$ state is present suggesting downconversion. We also observe upconversion emission at about 550 nm when we pump with 980 nm [see Ref. 8, which also provides some initial lower bounds on quantum efficiency of the processes discussed here]. While it could be argued that we are observing down-shifting and not downconversion, the fact that we observe upconversion is a strong indicator of the presence of the two-electron cross relaxation processes we described in Fig. 1. Nevertheless, proof that we observe conversion of one absorbed photon into two emitted photons requires a definitive measurement of quantum efficiency. Quantitative measurements of photoluminescence after absorption of 980 nm photons by the Er $^4\text{I}_{11/2}$ state have only shown a quantum efficiency of approximately 0.5 [8], meaning that quantitative proof has not yet been attained by us.

We used a second PL system to probe below 1000 nm and the result is shown in Fig. 4. These data show the signature of the Er

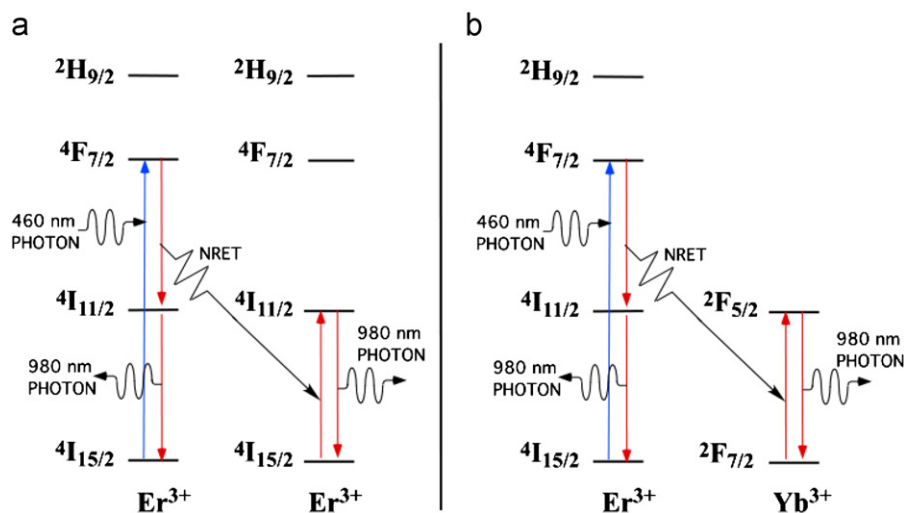


Fig. 1. Downconversion based on Er. (a) Two Er ions exchange energy non-radiatively. (b) Energy is exchanged between Er and Yb.

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