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A Study of Down-Conversion in Lanthanide Ions Doped Glasses for the Applications in Solar Cells

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Abstract – Solar energy is a renewable source of energy with low negative impact on the environment. For the enhancement of conversion of energy various techniques and material are studied by researchers. Upconversion and down conversion are two most important methods for conversion of energy from low to high and high to low frequency. Lanthanide ions are highly promising candidates for getting up and down conversion.

Keywords – Down Conversion; Lanthanides.

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INTRODUCTION

Solar energy is most readily available source of energy. Solar energy is a renewable source of energy with low negative impact on the environment. Its growing market is driven by the technological improvements and the policies supporting renewable energy sources [1]. Recently, there has been a strong interest in sustainable energy production to minimize the negative impact of global energy consumption on the environment. One alternative is to use the energy of the solar spectrum through solar cells.

Two approaches are usually explored to increase the efficiency: one is based on the search for new materials or techniques that can explore efficiently the photovoltaic effect, and the second one is related to the solar spectrum modification in order to match it with the solar cell spectra efficiency.

A reduction in price, however, may be achieved by either lowering the production cost or increasing the conversion efficiency Single junction solar cell efficiency is limited to 30% (the Schockley–Queisser theoretical limit) mainly because of losses caused by spectral mismatch, in which the largest part of the 70% energy loss is related to the thermalization electron–hole pairs and silicon transparency for the sub-band-gap incident photons [2]. Alternative techniques such as the texturization of silicon solar cells, the use of scattering particles, and the improvement of the fabrication process are also exploited to optimize the efficiency of the solar cells.

Down conversion Investigation

In addition to losses from sub-band gap photons it is estimated an even greater proportion of the solar spectral energy (Fig.1) lost from photons possessing too much energy. In silicon solar cells, a photon with energy greater than the band-gap will create an electron-hole pair but the once in the conduction band the charge carrier has excess energy which will mostly be lost as dissipated heat. Cells also have an optimal external quality efficiency (EQE) that varies with wavelength so in theory getting as much of the spectrum as possible at that energy would be an excellent way to improve efficiency. Although contrary to upconversion, lower band gap cells or systems with a poor UV response stand to gain the most from an efficient down conversion layer.

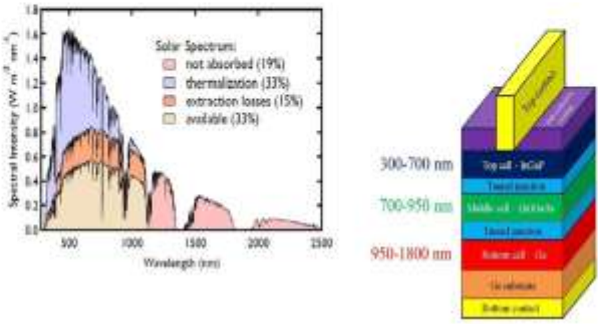

Fig. 1. The percentages of the solar spectrum represented as the energy losses that occur in a silicon cell. [3]

Working Principles of Down conversion

Owing again to their diverse energy structure, rare earth materials have considerable potential for down conversion devices. Down conversion mechanism is explained in Fig. 2.

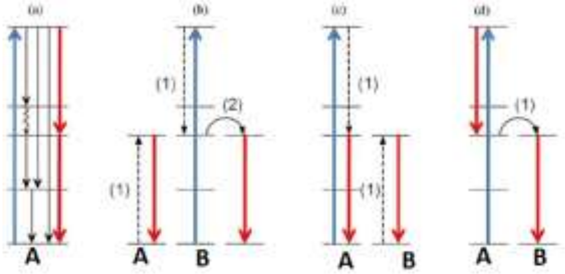

Fig.2 Down conversion mechanism

Firstly, a mechanism equivalent to ground state absorption (GSA) or excited state absorption (ESA) in reverse, a U-V photon is absorbed in a single ion, promoting an electron from the ground state to a secondary excited state before decaying sequentially, emitting two NIR photons. There also exist three possible routes for Down conversion via resonant energy transfer between two distinct ions as shown in the figure. Finally, a reverse co-operative energy transfer takes place when an ion absorbs a photon and transfers the energy to two additional ions promoting them to an excited state. These states then simultaneously decay emitting two low energy photons. Despite these similarities it has been shown unlike upconversion, Down conversion tends to be a linear process independent of light intensity [4]

Down conversion materials

The Down conversion process has been observed in several lanthanide compounds and from observing the energy transfer efficiency in lanthanide ions. Despite this and Trupke's earlier promising analysis, in 2007 Strümpel et al. [5] concluded Down conversion to have little practical promise in terrestrial PV due to the high excitation energies required falling within UV light which is mostly blocked by the atmosphere. A further problem arises from having to place the Down conversion material on top of the solar cell which leads to parasitic absorption and scattering losses.

Nevertheless, research in this field continued with Huang et al. cataloguing many studies from their 2013 review of both single ion and paired ion lattices [6]. The addition of other species can sensitize the Down conversion process by increasing the absorption cross section. These have often taken the form of divalent rare earth ions such as Eu^{2+} and Yb^{2+} or transition metal ions such as Mn^{2+}. Tai et al. [7] demonstrated Eu^{2+} acted as an efficient energy sensitizer for Yb^{3+} in a $SrAl_2O_4$ lattice, reaching a quantum efficiency of 147.36% under illumination by a 450 W xenon lamp. The broadband 250 to 450 nm light was shifted to 980 nm where it could be effectively utilized by a silicon solar cell. Most studies have implied use for c-Si but it's clear other technologies could benefit too, such as Ge cells with their very low band gap of 0.67eV (1850 nm). As with upconversion, it is important to carefully select the dopant concentration and layer thickness to minimize self-absorption losses. Boccolini et al. have presented an optical model to determine the optimal thickness of a Down conversion layer which was validated by experimental analysis of a Ce^{3+} - Yb^{3+} co-doped borate glass [8]. Li et al. [9] have also demonstrated broadband Down conversion in Yb^{3+} doped Na_2YMg_2 $(VO_4)_3$ which under UV illumination 240 to 400 nm showed intense NIR emission at 974 nm, ideal for silicon cells. By analyzing decay curves a mechanism based on combined energy transfer was proposed. For a recent review focusing exclusively on the available Down conversion materials and their application to PV the reader is referred to [10].

Down conversion Applications for Silicon Photovoltaic cell

Until recently there had been few physical DC-PV systems fabricated and their efficiency enhancements recorded. However there have been an increasing number of studies since 2015. That year, Dumont et al. [11] prepared Tb3+ - Yb3+ co-doped Silicon Nitride matrix down conversion layers via magnetron co-sputtering. By using novel optimization techniques and material characterization, its yield was improved from prior studies and was deemed to be suitable for application to a silicon solar cell, due to efficient of emission of 980 nm light following 325 nm illumination. It also had anti-reflective coating properties, a further reason for this being an excellent candidate for being placed atop an actual silicon cell. Florencio et al. [12] reported a 7% enhancement in the efficiency of a commercial silicon cell when Tb3+ - Yb^{3+} co-doped tellurite glass was placed on top as compared to un-doped glass.

Down conversion Applications for Emerging Photovoltaic Cell (PV)

Down conversion particles have also been applied to emerging PV cells. In a novel investigation Yao et al. [13] enhanced the conversion efficiency in a Dye Sensitized Solar Cells to 4.8% (an improvement of 245% from pure TiO_2) using Eu^{3+} -Dy^{3+} co-doped ZnO applied to the TiO_2 photo anodes alongside graphene loading to reduce recombination and interfacial resistance. Similarly in 2016, Hou's group have reported a Eu^{3+} doped $ZnGa_2O_4$ Nano phosphor in the porous TiO_2 of a perovskite solar cells [14]. This led to an enhanced short-circuit photocurrent by a maximum of 4.08 mA/cm2 and relative power conversion efficiency increase of 34.4% due to improved light harvesting.

Luminescent Downshifting Investigations

Rare earth doped phosphors [15], organic dyes [16] and quantum dots [17] have all been suggested to be utilized for achieving luminescent downshifting

(LDS) structures for PV cells. The responsible mechanisms will vary by material and do not appear to be as widely discussed in the literature as per the rare earth ion energy level diagrams described for upconversion/Down conversion.

However, Huang et al. [18] outline five key characteristics for luminescent downshifting materials:

1. Broadband absorption, particularly in regions of poor spectral response for the relevant solar cell
2. High absorption coefficient and quantum efficiency
3. High transmittance and narrowband emission in region of high device response
4. Large Stokes shift to minimise self-absorption losses
5. Long term stability

Rothemund et al. presented a simple, analytical optical model to calculate the potential EQE gains for various solar cell technologies following the addition of an LDS layer. This could be useful in future studies for evaluating experimental data, optimizing LDS layers and screening potential LDS materials [19]. In another investigation Lipovsek et al. [20] used 3-D ray tracing and Mie scattering theory, to produce an experimentally verified model which predicted Organic Solar Cells (OSCs) could gain 6% short-circuit current from an LDS layer. The model accounted for several parameters such as layer thickness, phosphor size distribution and volume concentration. Furthermore, Lesyuk et al. [21] have explored cadmium-free QDs as a non-toxic alternative to the often used CdSe/ZnS QD.

Luminescent Downshifting Applications for Silicon PV

Hung and Chen [22] prepared a Eu^{3+} doped gadolinium oxysulfide layer for a mc-Si solar cell. Under ambient solar illumination conditions an enhancement in short-circuit current density (JSC) of 6.47mA/cm2 and power conversion efficiency of 2.67% was observed. This material was simple and low cost to produce and also acted as an anti-reflective coating, reducing losses from Fresnel reflection. Similarly, Ho et al. [23] used two Eu^{2+} doped phosphor species mixed with SiO_2 and spin coated the solution on a single silicon solar cell. The cell's JSC increased 19.85% and relative power conversion efficiency by 15.97% due to a combination of broadband (512 to 610 nm) emission and forward scattering. Tm^{3+} doped fluoride glasses have also been investigated for their LDS properties and combined with a c-Si cell. Maalej et al. [24] reported a modest 1.4% relative power conversion efficiency enhancement at a 1% Tm^{3+} doping concentration when compared to un-doped glass. They concluded a greater absorption and collection of converted photons was needed to scale up these glasses for commercial application.

CONCLUSION

We have reviewed spectral modification to get better energy conversion through solar cells. Down conversion and luminescent downshifting were studied also have been presented that successfully attempt to apply these processes to harnessing greater energy from the solar spectrum. From the investigations using rare earth doped compounds, it seems the cells most likely to benefit from down conversion and luminescent downshifting. However, these studies may yield exciting prospects for expansion of solar cells.

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