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Novel nanostructures for efficient photon upconversion and high-efficiency photovoltaics

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ABSTRACT

Upconversion of low-energy photons theoretically allows the creation of single-junction solar cells with efficiency far above the Shockley-Queisser (SQ) limit. However, the net efficiency gains that can be realized depend critically on details of the upconversion process employed. We define three important metrics of the performance of an upconversion material: upconversion quantum efficiency (UQE), photon energy sacrifice (PES), and absorption bandwidth (AB). We analyze the performance of existing upconversion materials relative to both these metrics and existing computational models of a single-junction photovoltaic (PV) cell backed by an upconverter. Guided by the results of this analysis, we develop a design for new solid state upconversion nanostructures that suppresses the dominant energy loss pathways and can enable substantial improvements in overall solar energy harvesting. We describe and model the performance of a specific realization of this design that uses an InAs quantum dot (QD) and a graded InAlBiAs layer to suppress both radiative and nonradiative loss pathways. We show that this design can be tailored to maximize upconversion efficiency and can enable a practical upconversion-backed PV system to exceed the SQ limit.

1. Introduction

Solar cells are a promising path towards the development of renewable energy technologies that can displace fossil fuels, but many challenges must be overcome. A key challenge is the reduction of the installed cost per watt, which can be lowered by reducing the installed module cost or increasing the power generated per module. Research on first and second generation solar cell technologies will lead to increases in practical efficiency and decreases in module costs, but the theoretical maximum solar energy conversion efficiency of 33% places a limit on the total reduction in balance of systems costs that can be realized with these approaches [1]. The primary origins of the 33% limit derived by Shockley and Queisser for a single-junction photovoltaic (PV) are: (1) photons with energy larger than the bandgap lose their excess energy to heat, (2) photons with energy below the bandgap cannot be absorbed, (3) the formation of p-n junctions sacrifices energy to guide carrier transport and allow photocurrent to be harvested, and (4) a p-n junction at equilibrium has equal optical absorption and recombination and the power output is zero. Third generation solar cells utilize the solar spectrum in ways that overcome the Shockley-Queisser (SQ) limit. For example, multiple exciton generation (MEG) PV devices convert photons with energies at least twice the bandgap into two electron-hole pairs, thus doubling the photocurrent generated by these high-energy photons. There has been extensive work to produce MEG solar cells, but simultaneously enhancing MEG and extracting photocurrent is extremely challenging and the efficiency of existing devices remains poor [2,3]. Multi-junction solar cells, which utilize multiple materials with different bandgaps to absorb sunlight over the whole spectrum, have demonstrated efficiency in excess of 40% using three-junctions based on III-V semiconductors [4–6]. However, the material growth cost and current matching requirements continue to be a significant challenge for cost-effective multi-junction solar cells. Additionally, the need for current matching makes multijunction devices extremely susceptible to changes in the solar spectrum (e.g. clouds), which can result in practical efficiencies below that of the single-junction cell. Intermediate band solar cells (IBSC) introduce energy levels in the middle of the bandgap to improve overall efficiency by enabling absorption of a wider portion of the solar spectrum [7,8]. Although intermediate bands within wide bandgap materials have been realized with quantum dots [9], band anticrossing [10], and impurity levels [11], the additional loss pathways introduced by the intermediate levels have outweighed any efficiency gains from utilizing low-energy photons [12].

Photon upconversion is another approach that utilizes more of the solar spectrum in order to improve overall solar energy

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conversion efficiency [13–15]. Upconversion-based PV is conceptually simple. High-energy photons are harvested by a host PV cell. Photons with energy below the host cell bandgap pass through the host cell to an upconversion material. Within the upconversion material sequential absorption of two or more low-energy photons promotes an electron through intermediate states to a high-energy state from which it relaxes by emitting a single high-energy photon. This high-energy photon is returned to the host solar cell where it is absorbed. This approach increases the effective photon flux on the host cell, and thus the net current, while preserving a large open circuit voltage. A schematic single-junction solar cell equipped with an upconverter is shown in Fig. 1(a) and a schematic upconversion process is shown in Fig. 1(b). A key advantage of upconversion-backed PV systems is that the upconverter is electrically isolated from the host solar cell by an insulating layer acting as a transparent back contact. The electrical isolation of the upconversion layer avoids both the need for current matching between the host cell and upconversion layer and the degradation of the performance of the host cell due to the presence of mid-bandgap recombination centers.

Despite the conceptual simplicity, upconversion is difficult to implement because it always competes against both radiative and nonradiative loss processes. Upconversion has been observed in lanthanide and transition-metal-ion systems [16–19], quantum structures [20,21], and sensitized triplet-triplet-annihilation (TTA) molecules [22,23]. Photovoltaic devices backed by upconversion materials have also been shown to achieve small peak efficiency enhancements under solar concentration [24]. However, all existing photon upconversion materials (e.g. [19,23]) have severe limitations: (1) only a narrow band of incident photon energies can be absorbed, (2) there is a low probability of upconversion and (3) much of the absorbed energy is lost as heat. We describe an approach that overcomes these limitations by using semiconductor materials with wide absorption bands and applying heterostructure engineering to make a small, controlled sacrifice of energy after each absorption step. This approach dramatically improves upconversion probability, minimizes the loss of energy to heat, and allows optical absorption and emission wavelengths to be tailored for PV or other applications.

2. Modeling upconversion materials and systems

The theoretical efficiency of a single junction solar cell backed by an upconverter has been modeled by Trupke [13] and Atre [14] using detailed balance methods. In their approach, the upconverter is modeled as a series of two small bandgap solar cells driving a light-emitting diode (LED). The two sub-cells have different bandgaps, enabling the upconverter to harvest two sub-bands of the solar spectrum in a manner analogous to both multi-junction solar cells and IBSCs. This model system allows the application of well-established detailed balance models for each of the upconverter sub-cells and standard diode equations for the LED. Using this approach, Trupke calculated a maximum theoretical device efficiency of 47.6% for nonconcentrated sunlight [14]. The results of Trupke and others demonstrate that significant gains in solar energy conversion efficiency could be obtained with upconversion-backed solar cells. However, detailed balance models consider only the bandgaps of hypothetical materials and do not provide metrics by which potential upconversion materials can be evaluated.

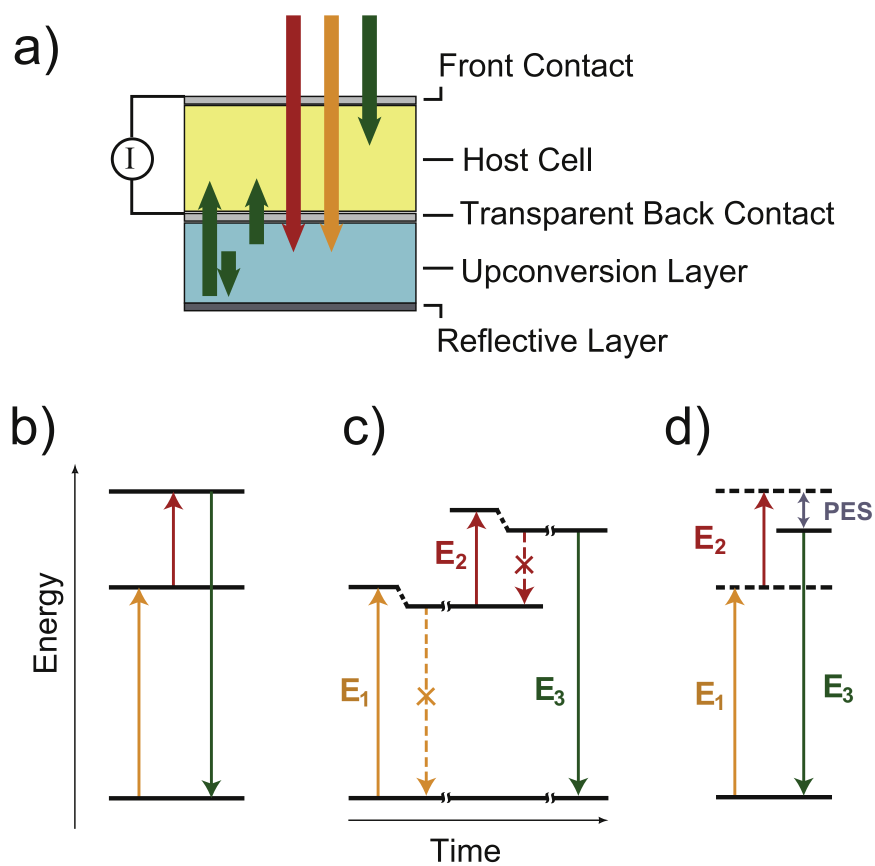

Fig. 1. (a) Schematic diagram of a single-junction solar cell backed by an upconverter. High-energy photons (green) will be absorbed by the host solar cell. Low-energy photons (orange and red arrows) will pass through to the upconversion layer and be converted to high-energy photons that are then returned to the host solar cell. (b) Schematic depiction of perfect upconversion with no energy loss. (c) Schematic depiction of intentional energy loss used to suppress radiative and nonradiative loss. (d) Depiction of PES. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).