Final Report for DOE/EERE

Project Title: Solar upconversion with plasmon-enhanced bimolecular complexes

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Award Number: DE-EE0005331
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Working Partners: Bosch, RTC (through 10/01/2014)
Cost-Sharing Partners: Bosch, RTC (through 10/01/2014)

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Signature Date
Project Objective:
Upconversion of sub-bandgap photons is a promising approach to exceed the Shockley-Queisser limit in solar technologies. However, due to the low quantum efficiencies and narrow absorption bandwidths of upconverters, existing systems have only led to fractional percent improvements in photovoltaic devices (~0.01%). In this project, we aimed to develop an efficient upconverting material that could improve cell efficiencies by at least one absolute percent. To achieve this goal, we first used thermodynamic calculations to determine cell efficiencies with realistic upconverting materials. Then, we designed, synthesized, and characterized nanoantennas that promise >100x enhancement in both the upconverter absorption cross-section and emissive radiative rate. Concurrently, we optimized the upconverer by designing new ionic and molecular complexes that promise efficient solid-state upconversion. Lastly, with Bosch, we simulated record-efficiency semi-transparent cells that will allow for ready incorporation of our upconverting materials.

While we were not successful in designing record efficiency upconverters during our three years of funding, we gained significant insight into the existing limitations of upconverters and how to best address these challenges. Ongoing work is aimed at addressing these limitations, to make upconversion a cost-competitive solar technology in future years.

Significant Accomplishments:

1. We developed a thermodynamic model to accurately predict the photovoltaic efficiency enhancements that can be expected with upconverting materials. Our calculations show that upconverters can increase cell efficiencies by over one absolute percent (significant for commercial applications) with improved quantum yield. Our results highlight the interplay of absorption and quantum-yield in upconversion, indicate the need for significantly improved upconversion efficiencies compared to existing materials, and provide a platform for optimizing future solar upconverter designs. (lead: Stanford)

2. We developed optical models of several state-of-the-art commercial and research thin-film solar cells incorporating upconversion layers. Models were built for amorphous Si, CIGS, GaAs, and Cu2O cells, including the full cell layer structure and incorporating the interface texture. These features allowed us both accurately predict the cell efficiency improvements with an upconverter, and also optimize the cell design to achieve the maximum benefit from upconversion. Efficiency increases due to upconversion range from 1.5% to 5% (absolute percent), and are higher for higher-efficiency cells, such as GaAs. (lead: Bosch)
3. We designed, fabricated, and characterized a metal-dielectric "nanocrescent" that promises >100x enhancements in both the upconverter absorption cross-section and emissive radiative rate. The structures can be fabricated in high-yield at relatively low cost, and form the basis for future upconverting solids. *(lead: Stanford)*

4. We fabricated and characterized large-area (cm-scale) ordered crescent arrays. These arrays can be integrated with an upconverting material in several ways and stamped onto the back of semi-transparent cells; such work will be the subject of on-going research. Importantly for solar applications, the resonance of these arrays is easily tunable simply by changing the etch conditions of the original crescents. This will facilitate incorporation of these arrays with a variety of upconverting materials. *(lead: Stanford)*

5. We developed a new 3D characterization technique, which we term cathodoluminescence (CL) tomography. This technique enables the first three-dimensional visualization of structural, optical, and spectral properties of materials with nanometer-scale resolution. We anticipate that this new imaging technique will enable a deeper understanding of light-matter interactions in almost any inorganic or organic materials system, from probing luminescence centers in upconverting materials with atomic-scale resolution, to visualizing excitonic diffusion in organic photovoltaic devices. *(lead: Stanford)*

6. We used both mechanical strain and chemical dopants to tune the host lattice of lanthanide nanoparticles to modify their upconversion efficiency, demonstrating a 2x increase in upconversion for Yb,Er:NaYF4 nanoparticles. *(lead: Stanford)*

7. We designed a new scheme for broadband upconversion, based on hot-carrier extraction from plasmonic systems. This scheme promises solid-state upconversion efficiencies exceeding 25% (out of a maximum of 50% for a two-photon process). *(lead: Stanford)*

8. Using time-dependent density functional theory (TDDFT) code, we identified strategies to red-shift the absorption of molecular upconverters and to broaden the absorption peak to enable the capture of more photons for lower band gap cells. *(lead: Bosch)*

**Deliverables and Go/No Go Criteria:**
A go-no go milestone for year 3 was demonstrating at least a 3% absolute increase in quantum yield for upconverting cores with plasmonic nanocrescents over baseline quantum yield of at least 3%. Unfortunately, this milestone was not met, so it was agreed that funding should be used for other projects in year 4.
Patents:

1. J. Dionne, A. Atre, B. Brenny, T. Coenen, A. Polman, Nanoscale optical tomography with cathodoluminescence spectroscopy, The Board of Trustees of the Leland Stanford Junior University, U.S. Pat. Doc. 027,125

Publications/Presentations:

In Press:


Published:


Presentations:


17) A. Atre, A. Garcia-Etxarri, T. Coenen, B. Brenny, A. Polman, J. Dionne, “Nanoscale optical tomography with cathodoluminescence tomography” Next Generation Photovoltaics, Erlangen, Germany, December 2013


**Jobs Created:** Two funded students have graduated. One is now employed at Sunpower, the other is taking a year off before applying for postdoctoral/faculty positions.