



Improved Solar Cell Efficiency Through the Use of an Additive Nanostructure-Based Optical Downshifter

Final Subcontract Report
January 28, 2010 — February 28, 2011

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SpectraWatt, Inc.
Hillsboro, Oregon

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Technology Summary

Spectrawatt's downshifting technology is based on a nanostructured material system which absorbs high energy (short wavelength) light and reemits it at a lower energy (long wavelength) with high efficiency. This system has shown unprecedented performance parameters including near unity quantum yield and high thermal stability.

The downshifter technology consists of a luminescent layer composed of chromophores embedded in a transparent matrix that is optically coupled to the solar cell. A fraction of the blue light incident on the luminescent material is absorbed by the chromophores (the fraction absorbed depends on chromophore loading) and re-emitted into the solar cell as red light (figure 1). All other photons are absorbed by the solar cell directly as indicated in the figure. The chromophore emits isotropically into the transparent matrix. Photons that are re-emitted at angles less than the critical angle escape through the top of the solar cell (~12.8% for a material with an index 1.5). A fraction of the re-emitted photons are lost out of the edge and some are re-absorbed and go through these processes again. [See SI section III for further information.] All other photons end up in the solar cell either directly or through reflectance off of the top surface. The optical properties of the downshifter layer must be controlled to maximize the fraction of photons that end up in the solar cell and get converted to electricity.

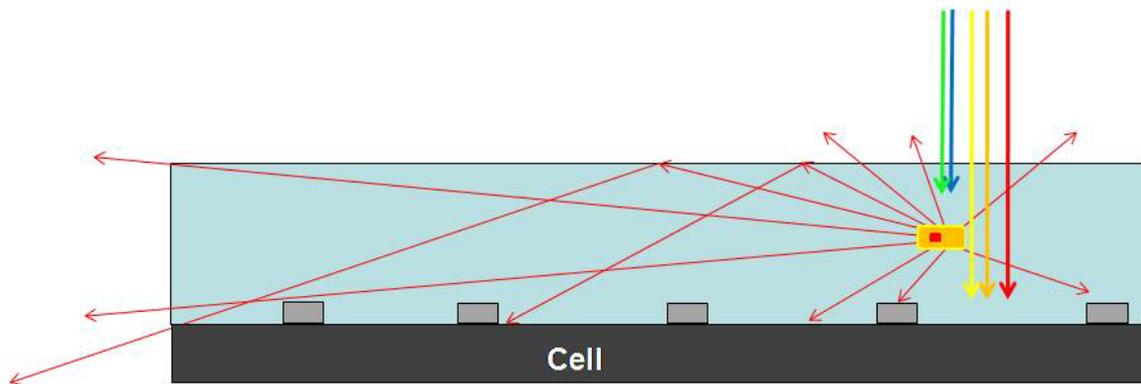


Figure 1: Operating principle of the luminescent downshifter

The goal of our materials system is to absorb the majority of the light below 500 nm where the solar cell is inefficient, and emit in the visible near 600 nm where the cell is the most efficient. The requirements for the downshifter are:

- High PLQY – this ensures that every photon absorbed is re-emitted as a redder photon
- Low self-absorption – since the chromophores emit isotropically, the photons can travel a large distance before entering the solar cell
- Long-term reliability – solar cells must survive 20+ years under the sun

- High temperature stability – solar panels in the field can reach temperatures as hot as 40 C above ambient
- Good compatibility between the chromophore and the host matrix – need to be able to avoid self-quenching through aggregation and need haze/clarity control.
- Optical compatibility between the downshifter and the PV module being modified.
- Good compatibility with the host matrix and the solar module.

A downshifter that meets our design rules can be seen in figure 2. The bulk of the absorption comes below 500nm while the emission is at ~600nm, a wavelength at which most solar cells are very efficient at converting light into energy (figure 2a). Such a system serves two purposes: first we are capturing photons that would otherwise be poorly converted, and secondly, by reducing overlap between absorption and emission spectra, we have very few self absorption events. The downshifter in figure 2 displays a PLQY of 90% in a film, which includes the losses due to the few self-absorption events that do occur. A comparison of our downshifter coupled to a CIGS cell and a standard encapsulated CIGS module is shown in figure 3b with a photograph of the two devices shown in the inset. The EQE of the two CIGS modules are shown. As can be seen in the figure, the response for the cell with the downshifter is much better in the blue. This difference accounts for ~1.5mA/cm² (or a 5% relative boost). The nearly flat EQE below 500 nm at 65% agrees with our accounting of all the photons.

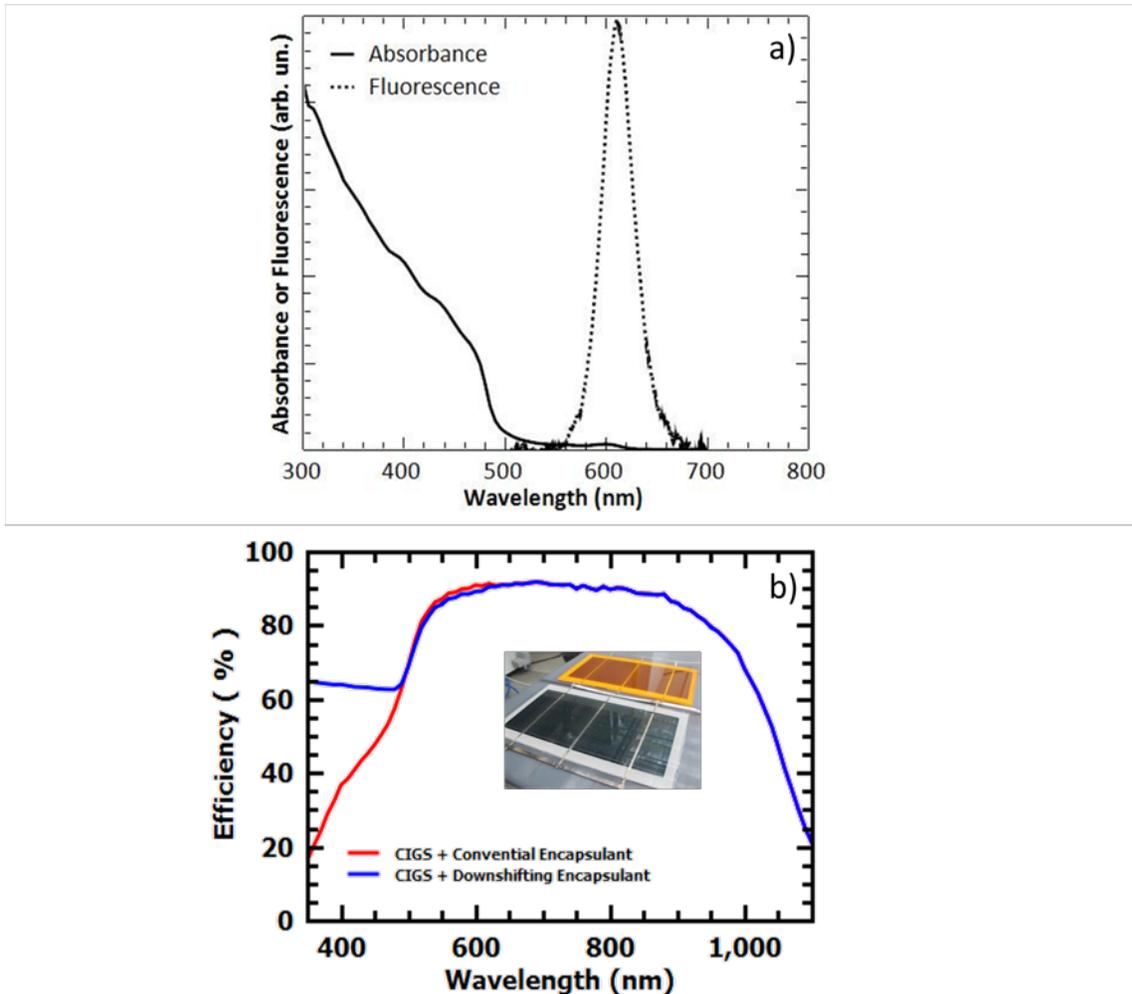


Figure 2: Optical and device properties of the luminescent downshifter. The absorbance and emission show minimal absorption by the chromophore at the emission wavelength – reducing self-absorption events. (a) Laminated CIGS cell with a standard encapsulant and with our downshifter (b).

Task 2: Quantum Dot Heterostructure Synthesis

The chromophore that generates the optical properties of figure 2 is a Quantum Dot Heterostructure (QDH). The QDH is based on a core/shell nanocrystal structure, in which a cadmium selenide (CdSe) core acts as the emitter, and a cadmium sulfide (CdS) shell is the absorber. [4] These two materials form a type I, or nested band structure as shown in figure 3a. This system is ideal for our purposes as it provides the downshift in photon energy, coupled with the ability to attain near unity photoluminescence quantum yields (PLQY). The quantum confinement of the CdSe core (figure 3b) allows us to tune the emission wavelength of the QDH simply by tuning its size. [5] The CdS, on the other hand, is a large rod shaped shell (figure 3c). The idea behind this architecture is to maximize the absorption ratio of CdS to CdSe as described above. The bigger this ratio, the more the absorption is due to CdS, reducing any self absorption. The shell is grown as a high aspect ratio rod to reduce the effects of strain when growing a thick

epitaxial shell. If too much strain builds up, as often happens in a large spherical shell, cracking and grain boundaries can form, which will lead to traps and reduced PLQY. [6]

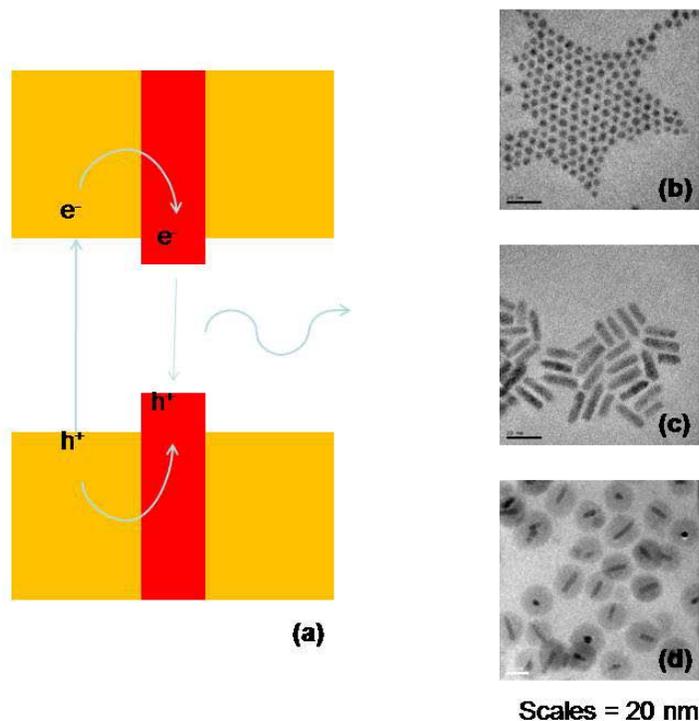


Figure 3: The quantum dot heterostructures. In the type I bandgap structure, an exciton excited in the wide bandgap material will recombine in the lower bandgap material, emitting a photon of lower energy (a). Transmission electron micrographs of the CdSe core (b), the CdSe/CdS QDH (c), and the encapsulated QDH (d).

Previous attempts to achieve stable PLQY >95% of high aspect ratio QDH have been less successful. [4] The QDH in these reports have suffered from incomplete surface passivation of the nanocrystal surface by the organic ligands. Because surface atoms make up a large portion of a nanocrystal, they are particularly sensitive to defects and external interactions on their surface [7]. These challenges are exacerbated in solid films – aggregation driven by ligand-ligand interactions leads to concentration quenching of the PLQY.

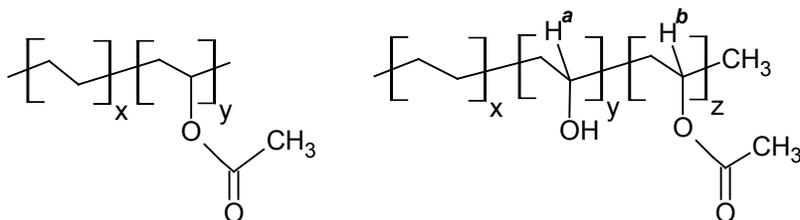
One way to overcome this problem is by the addition of an additional inorganic shell. [8] By tuning synthetic conditions, we incorporated a single QDH into silica spheres as shown in figure 3d. This shelling leaves the QDH surface well passivated, allowing PLQY values close to 100% in both solution and solid films.

Task 3: Thin films of QDH in a polymer matrix

Incorporation of the silica-encased QDH into PV encapsulants must be minimally disrupted to existing fabrication sequences of solar cell modules and, preferably, at no cost beyond that of the requisite materials. On one hand, the matrix must satisfy specific requirements set by the PV industry standards such as high adhesion to the substrate, high transparency, structural strength and excellent outdoor performance. On the other hand, in order to maximize the benefits of using the luminescent downshifter, the host material must provide an environment in which the PLQY of the incorporated QDH is stable. To date, the PV industry has had the most success with transparent polymers. Despite a wide range of options, only few meet all conditions.

In most of the cases, creating organic/inorganic hybrid materials represents a challenging problem. The systems are characterized by poor compatibility, leading to phase separation and QDH aggregation which leads to a drastic drop in PLQY. Consequently, device performance is compromised. Poly(ethylene-*co*-vinyl acetate) (EVA) is the most commonly used polymer as an encapsulant in the PV industry. By modifying the side chains, we can alter the macromolecules to the point that the QDH are well dispersed in concentrated polymer solutions with a long shelf-life. The hydrophilic-hydrophobic balance was controlled by mixing potassium hydroxide into EVA until the degree of hydrolysis was between 1 and 2%.

Scheme 1: Chemical structures of Ethylene-Vinyl Acetate Copolymer (EVA) and partly hydrolyzed EVA (HEVA)



The combination of the silica-encased QDH with the hydrolyzed ethylene-vinyl acetate copolymer (HEVA) yields a film that displays a PLQY >90%. This combination leads to colloidal stability and preservation of optical properties for both host and luminescent materials. To make the films compatible with standard solar panel manufacturing conditions, several cross-linking additives were added to the film.

The samples that yielded the 5% relative boost to CIGS cells were tested for their temperature, light and humidity stability to ensure their durability and performance maintenance over the extended lifetime expected of solar products, as well as the wide range of temperatures and environmental conditions experienced by an outdoor installation. Details of characterization methods are available in the experimental section.

Task 2 & 3 Experimental

All manipulations were carried out under non-inert atmosphere except the QDH synthesis. The materials (all reagent grade) were purchased from Aldrich and used as is. Polymers were characterized by NMR, FTIR and gel permeation chromatography (GPC). NMR spectra were

recorded on a Bruker 500 spectrometer. ¹H resonances were referenced internally to the deuterated solvent resonances. Polymers molecular weights were determined by GPC using a Viscotek GPC MAX liquid chromatography equipped with a triple detector array. GPC columns were calibrated versus polystyrene standards (American Polymer Standards). A flow rate of 1.0 mL/min was used with ACS grade THF as the eluent.

QDH synthesis and shelling

The CdSe seeds and CdS shells were synthesized with a modified version of the recipe reported in reference 7. The silica shells were formed with modification of the method described in reference 11.

Polymer modification

Synthesis of hydrolyzed ethylene-vinyl acetate copolymer (HEVA) was obtained from a copolymer with a vinyl acetate content of 40% by mole. The degree of hydrolysis of the vinyl acetate component was controlled by the amount of KOH and the reaction time. A typical procedure is described here. 25 g EVA was dissolved in 250 mL tetrahydrofuran. Separately, a 0.4 M solution of KOH in methanol was prepared. 2 mL of KOH solution was added to the stirring solution of polymer according to the desired degree of hydrolysis. The reaction was carried out for 30 min for a degree of hydrolysis of 2% and 2 h for a degree of hydrolysis of 5%. The hydrolyzed polymer was isolated from solution by precipitation in methanol or isopropanol. The organic solvents were decanted from precipitated polymer and the product washed with isopropanol or water to remove remaining salt and base, potassium acetate (KOAc) and KOH respectively. After filtration, the polymer was dried for 24 hours in a constant temperature vacuum oven at 40°C under vacuum. The total yield was 97%. FTIR was used to quickly identify the presence of hydroxyl groups in the polymeric product. In addition, NMR was used to determine the degree of hydrolyzation on the basis of chemical shifts integration of the protons in vinyl acetate and vinyl alcohol groups. When compared with starting material (EVA40), the GPC analysis of a HEVA with 5% degree of hydrolysis indicated only a slightly drop in molecular weight (from ca. 28 kg/mol to 24 kg/mol) but a significant narrowing in polydisperse index (1.83 to 1.35).

Film preparation

The QDH-polymer nanocomposite films were prepared by drop-casting from QDH-polymer dispersion in toluene prepared as follows. A known volume of QDH dispersion in toluene is added to a 10 % wt. toluene solution of HEVA and stirred/hold on the shaker overnight. The amount of QDH dispersion added to polymer solution depends on the type of particle (R_{abs}), the concentration of the QDH dispersion and the targeted O.D. Prior to dropcasting we add initiator (2% by wt) and accelerator (0.2% wt). Typically, films were prepared by dropcasting from a 650 μ L of QDH-polymer dispersion on the 25 mm glass rounds. Larger films can be prepared simply by scaling the casting volume to the surface area. Slow evaporation of the toluene in ventilated fume hood yielded highly transparent homogeneous luminescent films readily available for the PLQY and UV-vis measurements, lamination, reliability tests and device integration.

PLQY measurement

The PLQY was quantitatively measured by optically exciting samples and measuring the number of photons absorbed and emitted by the sample. This was accomplished by using a spectrally calibrated CCD detector incorporating an integrating sphere. The sphere ensures photons

emitted in all directions are collected, the CCD allows separation of spectral components due to excitation and emission, and spectral calibration allows the calibrated response of the system to be spectrally flat. Dividing the integrated emission counts by the integrated absorbed photon counts gives the PLQY.

Task 5: Achieve > 80% Light coupling efficiency

I. Photon Accounting

Early on in the project, our team devised a method to rapidly prototype the downshifter concept and to characterize the realistic efficiency ceiling and losses. The quantum dot heterostructures were not yet optimized to achieve high quantum yields and solid-solution films of well-dispersed QDHs had not yet been developed. The method involved fabricating test structures comprised of a glass well, or corral, coupled to an underlying solar cell which was filled with a liquid solution containing luminescent material. Because the optics of liquid downshifters are identical to solid solution downshifters, we could rapidly screen materials, and prototype concepts. For the loss-analysis, a laser dye, Coumarin 6, in toluene was chosen to fill a well on a well understood mc-Si cell. Coumarin 6 was chosen because its PLQY in toluene is near unity. Figure S3 shows representative test structures.

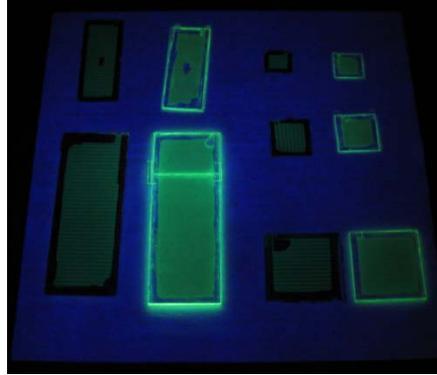


Figure S1: Pictures of the Coumarin 6 liquid wells.

Various experiments were performed to characterize all optical and electronic losses. The equations below show how the product of all photophysical efficiencies yields the EQE in the region the downshifter absorbs.

$$EQE_{total} = EQE_{DS} + EQE_{Filtered\ Cell}$$

$$EQE_{DS} = (1 - R) \cdot F_{abs} \cdot PLQY \cdot (1 - F_{escape}) \cdot (1 - F_{SA}) \cdot (1 - F_{other}) \cdot IQE_{cell}$$

$$EQE_{Filtered\ Cell} = (1 - R) \cdot (1 - F_{abs}) \cdot IQE_{cell}$$

Here, R is the reflection off the cell+well structure, F_{abs} is the fraction of light absorbed by the downshifter material, F_{escape} is the amount of light that is emitted within the escape cone, PLQY is the efficiency of converting absorbed photons to emitted ones, F_{SA} is the fraction of photons that undergo self absorption loss (through reduced quantum yield and increased light outcoupling), F_{other} characterizes all other losses fundamental or specific to the measurement (edge loss, grid loss, etc.) and IQE_{cell} is the internal quantum efficiency of the underlying cell.

The table below summarizes the measured losses using a multicrystalline Si cell as the underlying solar cell.

Table 1. Measured losses.

	mc-Si
Reflection off Glass	5%
PLQY	99 +/- 2%
Top Escape Pred.	12.8%
Top Escape	12.5 +/- 2%
Primary Side Escape Pred.	3%
Total Side Escape	4.5 +/- 2%
2ndry Side Escape	Small ~0%
Grid Losses Pred.	0.8 – 4.7%
SA Losses	Small to <5%
Si IQE	96 +/- 2%
EQE Predicted	73 +/- 3%
EQE Measured	72 +/-1%

Primary losses come from reflection off the top glass, and photons that are emitted within the escape cone, which from a medium with refractive index $n=1.5$, equals 12.8%. Other losses are specific to the cell and measurement: side escape, due to the small area devices tested, grid losses, which depend on the grid and busbar geometry and IQE of the underlying cell. The degree of self absorption strongly depends on the overlap between absorption and emission spectra of the downshifter material. The close agreement between measured and predicted EQEs clearly demonstrates that all photons have been accounted for.

The above photon accounting has been verified through the use of a 3D Ray-Tracing program BLAM was coded in the open-source mathematics computing platform Octave [ref website]. The program was developed in response to the apparent complications of complex geometries, inclusion of materials with varying optical properties, and multiple self-absorption events. The program is designed to take simple inputs in the form of optical density, absorption ratio (ratio of absorption at peak to absorption at emission wavelength), material geometries and indices of refraction, and compute statistics regarding photon fates, i.e. top-loss, side-loss, absorption into underlying cell, etc., based on Snell's and Beers' Law. The program takes inputs for sinks, reflectivity (Lambertian, specular, distribution functions), and optionally displays an overlay of path-lengths. The space is sampled randomly for a specified number of photons by computation of a probability matrix based on Beer's law.

Task 6: Reliability

Light exposure and Damp Heat (DH) accelerated testing (85C, 85%RH) testing over thousands of hours was performed as a step in the refinement of both QDH optimization and film (QDH + HEVA + additives) formulation. A variety of atmospheric conditions, e.g. vacuum, nitrogen, desiccation, were applied to films in development to isolate primary degradants and establish a range of acceptable operating conditions for the downshifter. Interestingly, the films are in general insensitive to ambient or elevated humidity, and only demonstrate decreased performance under the most desiccated atmosphere over the course of weeks, a condition not realized in their packaged operating environment for solar applications. Shown below in figure 4

is electrical data for a QDH/HEVA film incorporated into a typical thin-film CIGS package as an EVA replacement (exact configuration is proprietary), compared with an identical cell with plain encapsulant. The data is normalized to facilitate direct comparison of the samples with and without downshifter, demonstrating no appreciable difference in performance over >1000 sun hours. This power maintenance far exceeds performance criteria of IEC standards and demonstrates apparent viability for products carrying a twenty year performance guarantee.

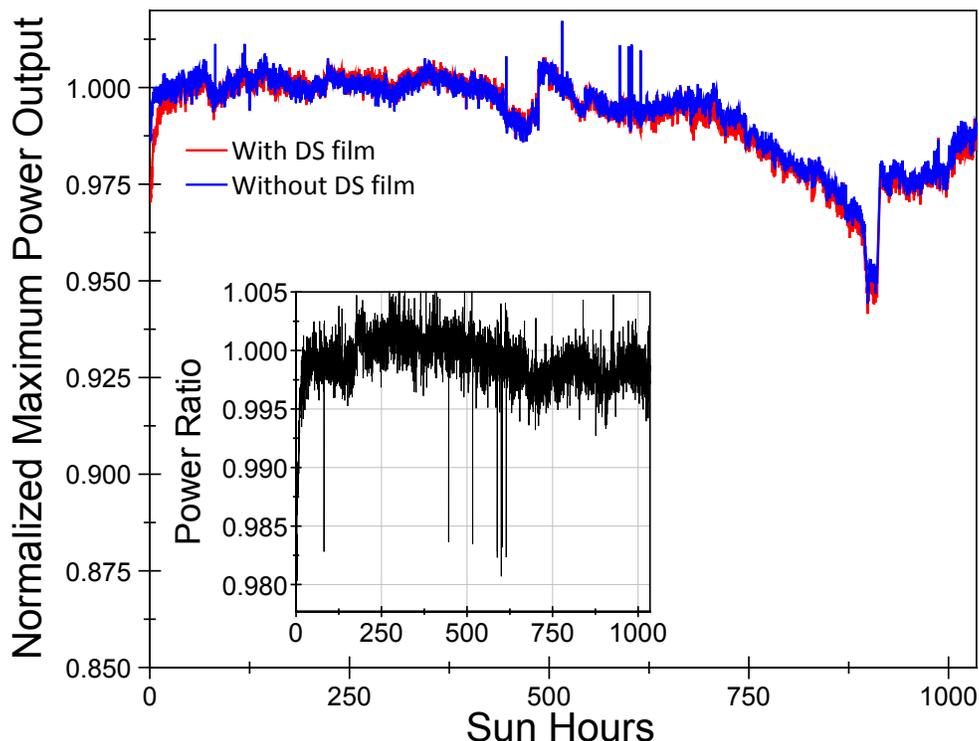


Figure 4: Pseudo-normalized power output vs. time of thin-film CIGS solar cell under one-sun illumination with and without QDH/HEVA film as an EVA topsheet replacement.

Fluctuations in the power output are due to lighting and electrical instabilities and fluctuations in operating temperature due to ambient temperature variation. This is demonstrated by the stability of the ratio of power output with and without the QDH/HEVA film to within 0.5% relative. Overall, the DS film does not result in a change in the reliability of the underlying device.

QDH films and devices were measured at a variety of temperatures to determine suitable operating temperatures. While this aspect of film performance presented the greatest challenge in process optimization, the apparent instability of comparable material systems at high temperature also provides the largest window for improvement.

In conclusion we have produced chromophores with nearly 100% PLQY specifically tailored for the downshifting application. These can be well dispersed in a variety of host materials common to the Solar industry. On CIGS solar cells, our downshifter adds about 1.5mA of current (5% relative increase on top of 30mA/cm²).

Reliability measurements

QDH solutions, films, and devices were tested at each stage of development for their ability to maintain quantum efficiency under the range of operating conditions relevant for solar applications. Furthermore, experimental conditions were employed to elucidate the origin of degradation, i.e. isolate the degradant and accelerating conditions.

Solar light-soaking: solution, film and active solar cell samples were exposed to one Sun conditions while temperatures of the sample were actively controlled. Furthermore, solid samples were often isolated with the use of cell containment units that allowed the samples to be in a carefully controlled atmosphere, most often vacuum, high-purity nitrogen, or constant-moisture air. Solution and film samples were monitored periodically for changes in both absorbance and luminescence with UV-Vis and Photoluminescence measurements, yielding a relative performance change against an absolute value measured in an integrating sphere. Electrical samples were continuously monitored by custom active loading electronics and maintained at their Maximum Power Point (MPP).

85/85 Humidity Testing: Films and devices were exposed to accelerated humidity and temperature testing, as is standard solar cells, e.g. as specified in UL1703 and IEC61646. Samples were placed inside a Hastest environmental test chamber and left at a temperature/humidity condition of 85C and 85% RH for extended periods of time (hundreds to thousands of hours). Samples were periodically removed, cooled, and tested: UV-Vis / PL measurements for optical films and IV characterization under one sun for devices.

Temperature stability: luminescent films were heated through a range of temperatures in order to trace their performance through a range of realizable operating conditions for solar applications. Films were mounted in a combined UV-Vis Photoluminescence measurement system, between thermoelectric unit coupled with a heat-spreading aluminum plate and plastic insulator. Temperature was monitored via a digital temperature sensor, and samples were cycled between ambient (~20C) and 80-100C.

Conclusions

In conclusion, SpectraWatt successfully integrated several processes from existing literature to synthesize and QDH and protect them from the environment. Taken together, these procedures enabled the production of a highly efficient downconverting material.

Ultimately the gains realized from downshifting compete with gains achieved in the blue. In the case of Silicon downshifting is outperformed by technologies like selective emitters. In the case of CIGS thinning of CdS or selection of an alternate window layer frustrate the application of downshifting technology. For superstrate CdTe, the optical losses associated with the thick top glass, TCO and high temperature of the process pose significant challenges to the application of the technology.

Despite these challenges we achieved very respectable gains on CIGS (0.5% absolute) which for some players may well prove to be a decisive advantage.

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Supplementary Information

I. Synthesis Optimization

As mentioned in the body of the document, the re-absorption of the emitted photons can be minimized by increasing the ratio of the CdS to the CdSe. Increasing this ratio allows the downshifter to absorb all of the light below 500 nm without absorbing much in the region where the solar cell is already performing well. A good way to define the degree of self-absorption is by the absorbance at 400 nm as compared to the absorbance at the exciton peak. E.g. if the ratio is 50:1 and the downshifter requires an optical density of 1 at 400 nm, the optical density at the emission wavelength would be 0.02. Figure S1 shows how the ratio affects the absorbance. Note how for equivalent absorbance at 600 nm, the absorbance below 500 nm increases with increasing ratio

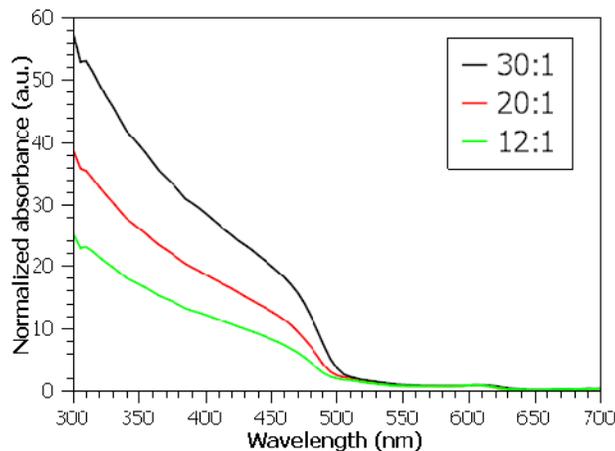


Figure S2: Absorbance of the QDH with increasing CdS to CdSe ratio.

II. Emission Tuning

One of the nice properties of the QDH is that the emission peak can be varied by varying the size of the CdSe core size. Tuning this emission is extremely useful for several of the applications of a luminescent downshifter. Figure S2 shows 3 of the particles we have made that span the colors green to red.

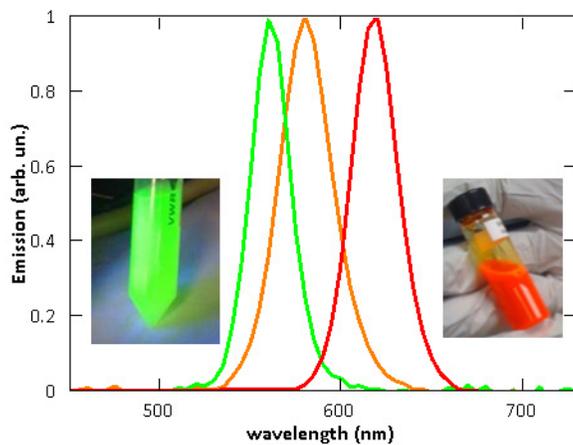


Figure S3: Emission tuning of the QDH with varying CdSe particle size

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