Trapping light with micro lenses in thin film organic photovoltaic cells

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Abstract: We demonstrate a novel light trapping configuration based on an array of micro lenses in conjunction with a self aligned array of micro apertures located in a highly reflecting mirror. When locating the light trapping element, that displays strong directional asymmetric transmission, in front of thin film organic photovoltaic cells, an increase in cell absorption is obtained. By recycling reflected photons that otherwise would be lost, thinner films with more beneficial electrical properties can effectively be deployed. The light trapping element enhances the absorption rate of the solar cell and increases the photocurrent by as much as 25%.

OCIS codes: (310.6845) Thin film devices and applications; (040.5350) Photovoltaic; (160.4890) Organic materials; (220.4000) Microstructure fabrication; (220.3630) Lenses

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A promising material class for manufacturing inexpensive photovoltaic cells is comprised of blends of conjugated polymers and fullerenes [1, 2]. These organic blends are sandwiched between transparent and reflective electrodes, and are typically 50-300 nm thick. The power conversion efficiency (PCE) of organic solar cells (OSC) is however still limited and they do not perform as well as their inorganic counterpart. One limitation of OSCs is the low charge carrier mobility which prevents generated charges from reaching the electrodes. Film thickness minimization is therefore a route to collect a larger fraction of the photo generated carriers, but a minimum thickness is always required for sufficient photon absorption. If the absorber is too thin, the device will simply reflect most impinging light. In this work we show how to construct and incorporate novel light trapping structures that enable higher photon absorption in a thin film by recycling the reflected photons.

Light trapping in thicker inorganic photovoltaic cells is a well known approach to increase the photocurrent [3 ,4]. In such cells the trapping of light is fulfilled by the introduction of a rough or structured active layer. As photons hit the structured features of the cell, they are forced to travel more in the plane of the absorber rather than perpendicular to it. As an efficient light scattering element should be somewhat larger than the wavelength of light to effectively alter the photon propagation direction, difficulties are inherent when incorporating such refractive or reflective structures inside a ~100 nm thick organic film. The electrical properties are extremely limiting when including light scattering elements in direct contact with such thin active layers. [5] Therefore, different engineering solutions may be required, where the light trapping element is separated from the thin active layer and thereby allow for beneficial scattering but avoid the possibility of electrical defects.

Light trapping techniques compatible with thin film polymer cells have previously been reported. Embossed gratings [6, 7] have demonstrated improved absorption via wave guide coupling. The utilization of small arrays of compound parabolic Winston collectors has also been suggested [8] but not realized. The recently reported folded solar cells have further shown considerable improvements in light collection and photocurrent [9, 10, 11]. We here demonstrate thin film organic solar cells, where the scattering and light trapping features are separated from the active layer in order to avoid the possibility of electrical defects. A light trapping element is placed in front of a solar cell with limited absorption. The trap is transparent to collimated light in one direction, and highly reflective in the opposite direction, to both collimated and directionally random light. Thereby an efficient trapping mechanism of the direct component of sunlight can be implemented. As light bounces back and forth between the reflective metal surface of the cell electrode and the back surface of the trap element, multiple transits through the active layer will occur, thus increasing the probability of photon absorption. Rules of reciprocity for collimated light propagation do not allow for the existence of such elements, but if one allows loosing the light collimation, it becomes feasible. The suggested element consist of an array of light focusing micro lenses on top of a reflecting mirror with small transmitting openings for the focused light (Fig. 1(a)). Ray tracing for a trap with realistic features on top of a 100% absorbing solar cell is displayed in Fig. 1(b) and for a 0% absorbing “solar cell” in Fig. 1(c). Real photovoltaic devices display a wavelength dependent absorption ranging in between these two extremes. For wavelengths where absorption is limited, photons will be recycled and get additional chances for absorption. For thin films, with an overall limited absorption, this type of trapping is thus expected to be even more beneficial. As thin active organic films often are desired due to electrical limitations such as poor mobilities and low built in electric fields, the light trap can be a tool to remedy the mismatch of photon absorption length and free carrier diffusion or drift length.

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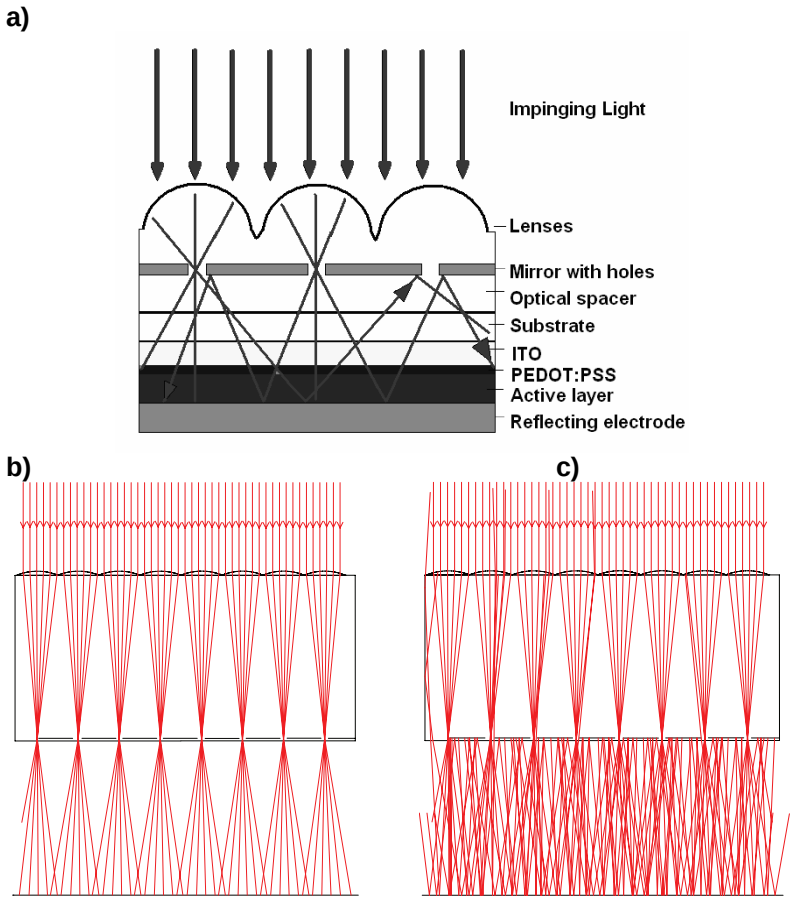

Fig. 1. Principle of the light trap. a) Operational principle of the light trap and its constituents. b) Ray tracing light with a 100% absorbing solar cell. c) Ray tracing light with a 100% reflecting solar cell.

Manufacturing of the light trap comprising the high optical quality array of micro lenses is depicted in Fig. 2. A detailed manufacturing description can be found in the method section below. Concave lenses are initially produced by controlled isotropic etching into a patterned Cr covered quartz substrate. The radius of curvature of the micro structures has been calculated and tuned to provide the required focal length of the desired final polymer micro lenses. As the micro lenses are dioptres, the focal length f is given by;

$$f=R[n_2/(n_2-n_{1)}] \quad (1)$$

where R is the radii of the lens, n_2 the refraction index of the material of the lenses and n_1 the refraction index of the surrounding medium (n_1=1 for air). The obtained array of lenses is subsequently imprinted in a malleable optical polymer resin on a glass or plastic substrate. The replication method, using imprint lithography, is a facile and easily up-scalable process, not very different to methods used for cheap production of post cards with lenses, so called lenticular arrays [12]. In the next step, apertures in a metal layer deposited on the opposite side of the substrate are generated in a self aligned process. This is a simple metal lift-off process, with the pattern defined by the UV exposure of a double layer resist through the micro lenses themselves. It therefore produces features which are self aligned to the array of the micro lenses. (Fig 2.2-2.7). This method could therefore be referred to as a “device self assembly process” as the holes generated by light are created in locations where they are also

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most useful for light trapping during solar cell operation. The final structures obtained are displayed in Fig. 3.

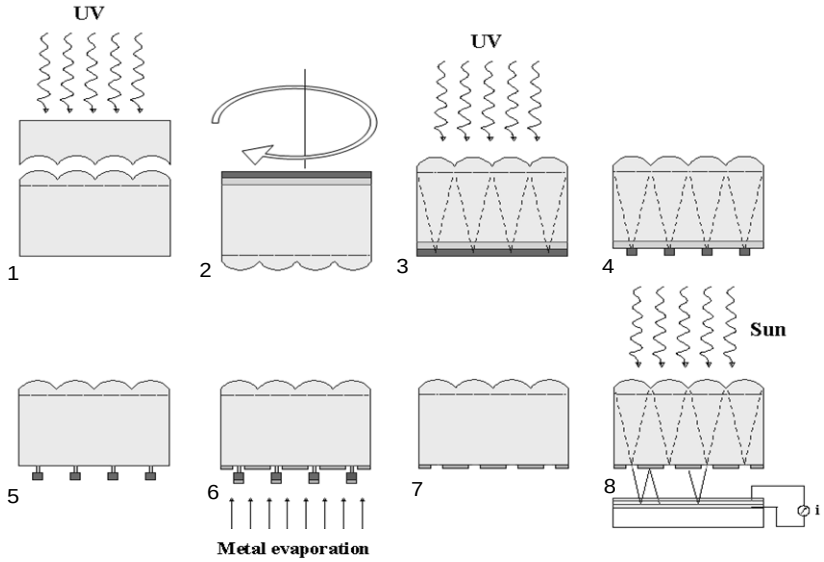

Fig. 2. Manufacturing route of the light trapping element. 1. Imprinting, 2. Resist deposition, 3. UV exposure through the lenses, 4-5. Resist development, 6. Metallization through evaporation, 7. Residual resist removal. 8.The light trap in operation with a solar cell.

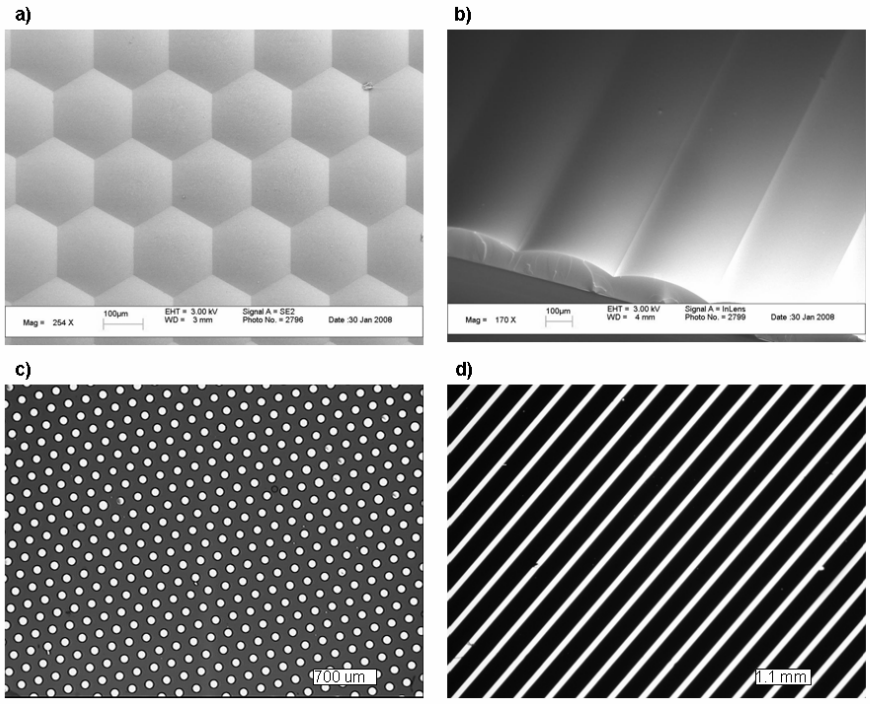

Fig. 3. Pictures of the obtained structures. SEM scans of the spherical (a) and cylindrical (b) polymer lenses. The periodicity is 200 and 400 μm respectively. Optical transmission microscope picture of the backside of the samples, comprising the perforated mirrors, is displayed in (c) and (d).

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The light trap is initially placed in front of an integrating sphere and the transmission in the forward and backward directions is characterized. Fig 4(a) shows the trap transmission as a function of wavelength. A strong asymmetry between the forward and backward transmission was confirmed, with transmission $\leq$ 90% in the forward direction (first through lenses and then mirror) and <15% measured with the sample illuminated from the side of the mirror. We observe that high forward transmission, obtained without the use of antireflection coatings or moth-eye structures, could be achieved only with the full surface coverage with micro lenses (Fig. 3), made possible by the present fabrication process. The dip in transmission at shorter wavelength is attributed to absorption in the lens material as well as in the metal mirror.

The active bulk heterojunction [13] consists of a 1:3 ratio blend of the low band gap alternating polyfluorene APFO Green-9 and the soluble fullerene derivative PCBM[70] whose molecular structures are inset in Fig. 4(b). The active layer in this device is spin coated from a 7 mg/ml chloroform solution at a speed of 3000 rpm. The layer thickness is only 30 nm, and has therefore a limited absorption. The reflection (R) from an operational solar cell with an area of 1.3 cm^2 is measured and the corresponding device absorptance (1-R) together with the active layer absorption is presented in Fig 4(b).

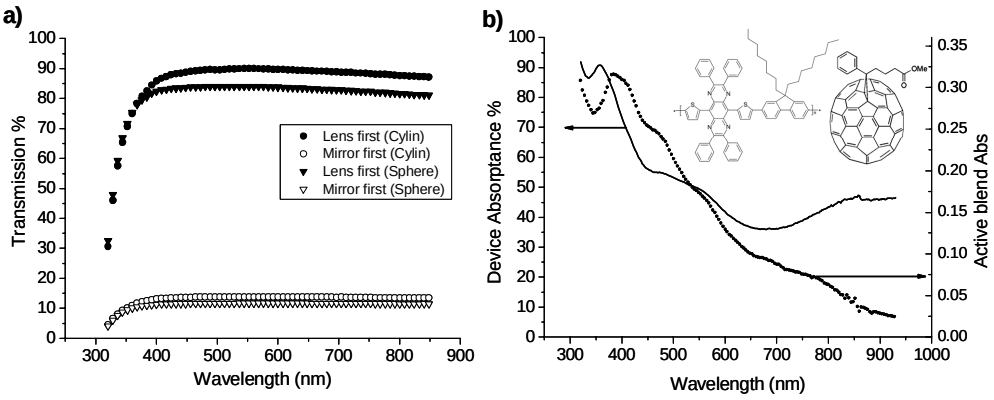

Fig. 4. Optical characteristics of the trap and the cell. a) Trap element transmission in forward and backward direction. b) Device absorptance as measured from reflectance and blend material absorption as measured from transmission. Inset: chemical structures of the utilized donor (APFO Green 9) and acceptor (PCBM[70]) materials in the blend.

The photoresponsivity (PR) from the thin solar cells, measured with and without the light trap laminated to the device, is presented in Fig. 5(a). The irradiance of the AM 1.5 solar illumination spectra is inset as a comparison. As can be seen from the PR curve, the light trap works very effectively over a broad spectral range. For this material and thickness, light trapping is beneficial for all wavelengths longer than 450 nm. The loss of the high energy photons is no major concern as their abundance is rather limited simultaneously as their energy may actually be harmful for the stability of the active polymer. The current/voltage curve (Fig. 5(b)) under AM 1.5 simulated solar illumination also demonstrate an increase in short circuit current, as high as 25%, upon the inclusion of the light trap.

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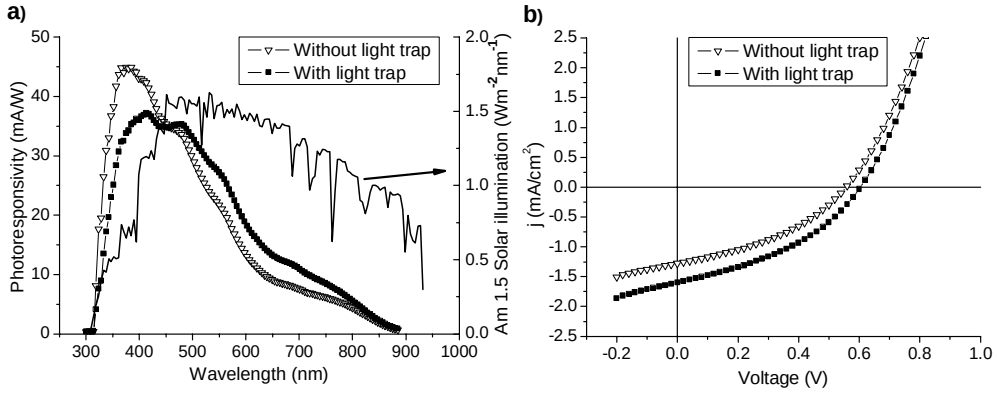

Fig. 5. Photovoltaic characteristics with the light trap. a) Photoresponsivity with and without the light trapping element in front of the cell, together with the solar irradiance spectra. b. JV characteristics from simulated AM 1.5 solar illuminated APFO Green 9/PCBM[70] solar cell with and without the trap. An increase in short circuit current of 25% can be confirmed upon the addition of the light trap.

For devices with significantly higher absorption, less or no improvement from the light trap can however be observed. If solar cell device reflectance (R_S), trap forward transmission (T_T) and trap backward reflection (R_T) is measured, it is possible to make a series expansion of the multiple absorption events in the cell, and compare it to the absorption of a device without a trap element and determine when trapping will be beneficial. Fig. 6(a) describes how the intensity is decreasing at the different interfaces after transmission and reflection at the involved interfaces.

Light trapping will therefore be beneficial if the following holds:

$$(1-R_s) \le T_T(1-R_s) + T_T R_S R_T (1-R_s) + T_T R_S^2 R_T^2 (1-R_s) + \ldots \qquad (2)$$

(Abs no trap) ≤ (First order Abs + second order Abs + third order Abs…)

For infinite bounces, the series in the right hand side of the equation will converge to:

$$T_T(1-R_S)\frac{1}{(1-R_S R_T)} \qquad (3)$$

Hence only if

$$R_S(\lambda) > \frac{1-T_T(\lambda)}{R_T(\lambda)} \qquad (4)$$

will light trapping be favorable. From equation (4) it is clear that for thicker samples with high absorption, the transmission and reflection properties of the light trap must be very good to enable any significant effect of trapping. For the 30 nm thin APFO Green 9/PCBM[70] device used, with a rather limited absorption, this is fulfilled for all wavelengths above 400 nm. The absorptance from a cell with no lenses compared to a cell exploiting the light trap is displayed in Fig. 6(b). Different number of reflections is also considered in the graph via eqn. (2). A strong enhancement of the solar photocurrent is possible when the reflectance from the thin film solar cell is sufficiently high, compared to the trapping efficiency of the light trap.

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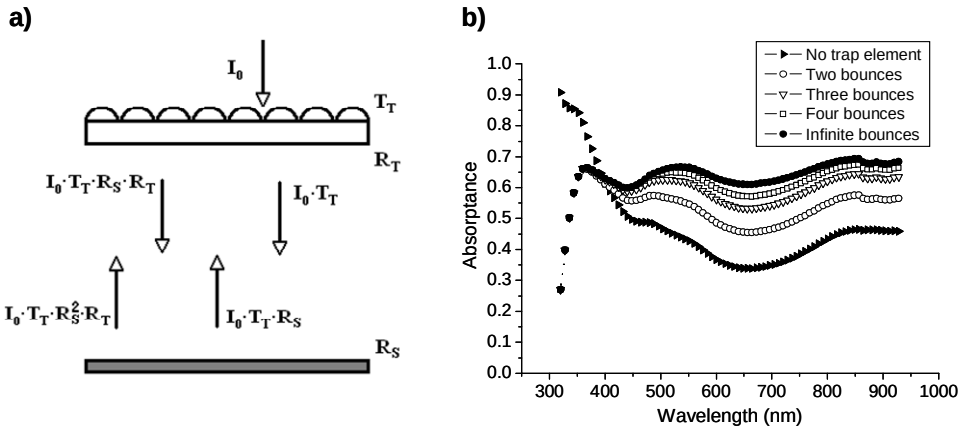

Fig. 6. Effect of multiple absorption events. a) Schematic of the multiple bounces and the according Fresnel intensity coefficients. b) Calculated total cell absorptance for the APFO Green 9/PCBM[70] system with and without the trap system. For different number of absorption events included, a noticeable improvement above 400 nm can be found.

For real solar power conversion, one limitation of the system can however be found in that it is based on refraction and accurate focusing and that rather sensitive angle dependence is displayed. Since focusing may then come about at locations where no holes are located, light will simply not illuminate the solar cell. This setback is pronounced for the cells exploiting the 2D hexagonal array of semi spherical lenses, where perpendicular alignment in two dimensions is very crucial. However, for the 1D semi cylindrical array, it is sufficient to have alignment only in the plane parallel to the axis of the lenses. For linear lenses, such as the cylindrical ones, lens axis alignment close to parallel to the ecliptic plane of the sun is sufficient and daily solar tracking may therefore not be required in locations closer to the equator.

In conclusion, for thin film solar cells where the conflict between optical absorptance versus electrical transport and collection is the rule, the described light trap can be a significant advantage. The possibility to produce light traps over large areas using simple imprint methods further enables the use of these elements also for low cost organic photovoltaics. In principle, for any solar cell where some part of the light is reflected instead of being absorbed, the described light trapping element may improve the photovoltaic performance.

1. Methods

The fabrication of high quality micro lens arrays with 100% surface coverage is obtained according to the concepts and processes described previously [14]. Initially, a master comprising an array of inverted (concave) lenses is needed. Conventional proximity photolithography and wet etching was therefore deployed to pattern the chromium layer of a 3 mm thick fused silica photo mask. Shipley Microposit S1813 photo resist was spun to a thickness of 1.5 μm and the exposed structures were subsequently developed in MF-319, followed by an etching step to remove the now exposed small chromium parts. After resist removal, the samples were introduced in hydrofluoric acid (HF) 48% for the etching of the unprotected fused silica. The radius of curvature of the isotropically etched structures can be controlled by adjusting the etching time in the HF solution, as the acid etches the substrate at a known rate of ~1 μm/min. Before the imprinting of the lenses, the master sample was functionalized with an anti adhesive monolayer coating of dodecyltrichlorosilanes to allow for removal after imprinting. The polymer micro lenses were then imprinted in Norland Optical Adhesive 73 (NOA 73) on a glass or plastic substrates. Primarily, a 20 x 20 mm glass slide of 1 mm thickness cleaned in hot acetone and IPA was deployed as substrate. Liquid NOA 73

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was sandwiched between the surface of glass slide and that of the master. Special care had to be taken in order to avoid the formation of air bubbles in the polymer film. A UV light source (~350 nm) was employed to cure the NOA 73 adhesive. The process of hole generation in the mirror is implemented by first spinning a double layer of a LOR 10 B resist and a MR-L 6005 negative optical resist. The LOR 10B was used to produce an undercut structure below the negative resist and as a sacrificial layer for the lift-off of the top negative resist. After UV flood exposure through the array of micro lenses (resulting in a UV-crosslinked pattern corresponding to the foci of micro lenses) and development steps of the photo resist, the LOR was developed with MF 319 to obtain the under cut necessary for the lift off process. Subsequently, a metal tri-layer comprising 5 nm chromium, 150 nm silver and 5 nm gold was evaporated. The chromium layer is an adhesion promoter, the silver is the real reflective layer whereas the thin gold film prevents the oxidation of silver. After the evaporation, the resist structures were removed by MF 319.

In order to produce dots or lines of varying sizes, and to solve problem of edge effects, chromatic aberrations and divergence of the beam, the samples have been mounted on a rotating stage (60 rpm) during the UV exposure. The axis of rotation was now tilted with respect to the axis of the collimated UV light source, allowing a controlled enlargement of the features produced by the self-aligned exposure. The rotation of the sample also improved the uniformity of the structures and reduces the edge roughness in the negative photo resist, as well as the angle sensitivity of the final light trap.

The I-V characteristics under simulated solar light (AM 1.5 100 mW/cm^2) from a SS 50A Photo emission Tech. simulator was recorded with a Keithley 2400 source meter. The thickness of the deposited organic films was determined with a surface profilometer (Sloan Dektak). Reflection studies of the cells and trap elements were performed with a Perkin-Elmer Lambda 950 spectrophotometer equipped with a 15 cm diameter integrating sphere to collect light at all angles. The photocurrent action spectra under short circuit conditions were recorded with a Keithley 485 picoammeter as the devices were illuminated with monochromatic light from a halogen lamp. All device manufacturing and characterization was executed under ambient conditions without a protective atmosphere.

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