Light-harvesting semiconductor quantum dot nanocrystals integrated on photovoltaic radial junction nanopillars

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In the last decades increased awareness for environmental problems, climate change, and limited energy sources has escalated research efforts around the globe to investigate photovoltaics as an alternative energy source. The sun offers an enormous potential for solar energy conversion, which makes photovoltaics quite attractive among green energy sources. However, there is still not an ultimate photovoltaic device which yet provides high efficiencies at low costs. Recently with the development of nanofabrication technologies, nanostructured solar cells have emerged at research labs (e.g., using nanowires, nanorods, nanotubes etc.) [1,2]. These nanostructured device architectures are intended to yield enhanced optical properties such as light trapping compared to existing planar devices [3,4]. In certain architectures, however, there are some nanofabrication bottlenecks that adversely affect the overall device performance of such nanostructured photovoltaics. These technical challenges can possibly be overcome with maturing fabrication methods in time. But, there also exist limitations stemming from intrinsic properties of the active materials that cannot be easily fixed unless hybrid approaches are exploited or materials are replaced. For example, silicon, which is the most commonly used material in photovoltaic industry [5], suffers too strong absorption at short wavelengths (i.e., UV-blue), undesirably leading to poor performance in this spectral range due to high front surface recombination rates [6]. However, UV-blue portion constitutes almost 10% of the sunlight, which is mostly unused by Si based photovoltaics [7]. To address this problem, we propose and demonstrate light-trapping radial *p-n* junction Si nanopillar solar cells that are furnished with CdSe quantum dot nanocrystals to harvest short-wavelength radiation in addition to long-wavelengths. The basic operating principle of these light-harvesting nanocrystals integrated on nanopillars relies on the wavelength up-conversion idea of incident photon absorption in these nanocrystals at short wavelengths and subsequent photon emission at longer wavelengths, which is in turn reabsorbed by the furnished nanopillar diodes. Although such light-harvesters were previously studied on planar devices or those with elliptical holes in them [8,9], nanocrystal-decorated nanopillars have not been investigated to date. Here we demonstrate that this nanopillar architecture provides additional advantage of trapping photon emission from the light-harvesting nanocrystals for wavelength up-conversion, compared to the planar case.

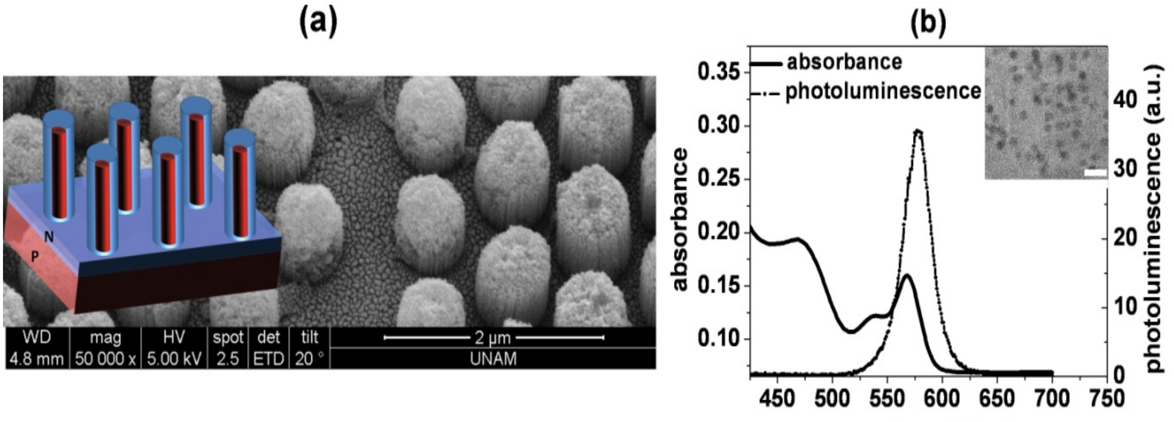

Figure 1. (a) Scanning electron microscopy (SEM) image of radial *p-n* doped silicon nanopillars, along with a schematic of device architecture (inset). (b) Photoluminescence emission and absorption spectra of light-harvesting CdSe quantum dot nanocrystals, together with a transmission electron microscopy (TEM) image of nanocrystal light-harvesters (with a scale bar of 10 nm) (inset).

Characterization of solar cells under AM1.5G (100mW/cm^2) illumination are undertaken before and after quantum dot hybridization, These characterization results showed that total power conversion efficiency enhancement is up to 13% by wavelength up-converting nanocrystal layer. Short circuit current (I_{sc}) and fill factor (FF) of the devices are increased from 6.56 mA to 6.90 mA and 50.4% to 54.5%, respectively. With the same amount of hybridization of quantum dots onto planar silicon solar cells, the efficiency enhancement is up to 11%. Fig.2 shows the I-V characteristics of the nanopillar and planar silicon solar cells before and after quantum dot incorporation.

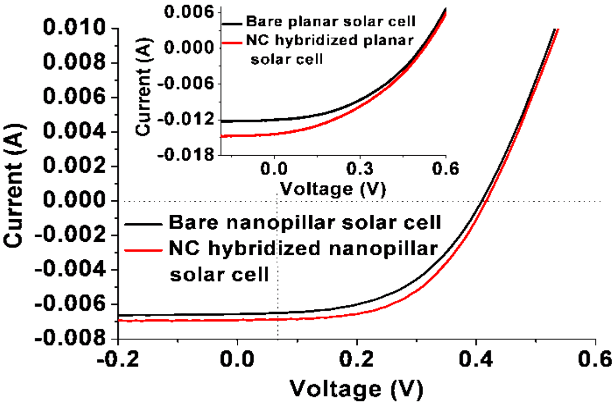

Figure 2. I-V measurements of radial p-n junction Si nanopillar solar cell under AM1.5G illumination, along with I-V of planar Si solar cell in the inset.

To understand the spectral enhancement effects of the wavelength up-converting nanocrystals, photocurrent and various spectroscopy measurements are performed (under zero external bias). Fig. 3 shows the responsivity enhancement as a function of incident wavelength for nanopillar and planar solar cells when the layer of wavelength up-converting quantum dots are introduced. The inset of Fig. 3 also depicts the external quantum efficiency (EQE) of both nanopillar and planar devices before and after the incorporation of quantum dots. In the nanopillar device, UV responsivity is enhanced by ca. 6 times with respect to the bare nanopillar device. Nanopillar architecture shows enhanced effect of nanocrystal layer because of better trapping of photons emitted by quantum dots, as also confirmed by our numerical simulations (using a finite-difference time-domain simulator developed by Lumerical Solutions, Inc., Canada).

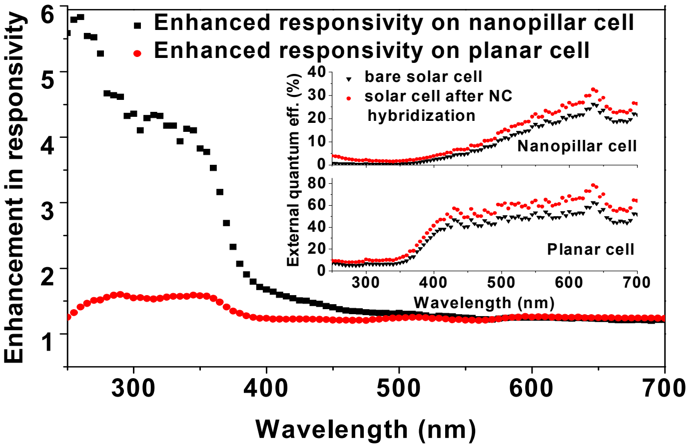

Figure 3. Enhancement of the responsivity of nanopillar and planar solar cell by nanocrystals layer. Inset shows the EQE of the both devices before and after the hybridization.

In conclusion, integrating wavelength up-converting layer consisting of luminescent CdSe quantum dots on top of radial junction nanopillar solar cells resulted in ca. 13% power conversion efficiency enhancement, with a responsivity enhancement as high as 6 folds in UV range. These experimental results indicate that nanopillar architecture is more advantageous than planar structures for light-harvesting of integrated quantum dots owing to improved light trapping properties in the nanopillars.

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