

Hybrid Silicon Nanocone–Polymer Solar Cells

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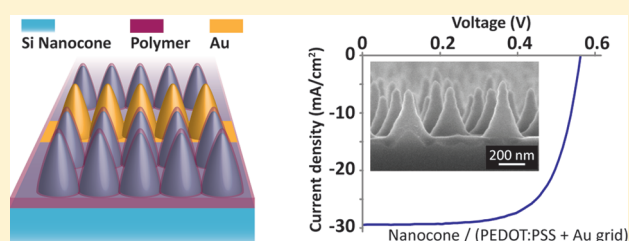
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Supporting Information

ABSTRACT: Recently, hybrid Si/organic solar cells have been studied for low-cost Si photovoltaic devices because the Schottky junction between the Si and organic material can be formed by solution processes at a low temperature. In this study, we demonstrate a hybrid solar cell composed of Si nanocones and conductive polymer. The optimal nanocone structure with an aspect ratio (height/diameter of a nanocone) less than two allowed for conformal polymer surface coverage via spin-coating while also providing both excellent antireflection and light trapping properties. The uniform heterojunction over the nanocones with enhanced light absorption resulted in a power conversion efficiency above 11%. Based on our simulation study, the optimal nanocone structures for a 10 μm thick Si solar cell can achieve a short-circuit current density, up to 39.1 mA/cm^2 , which is very close to the theoretical limit. With very thin material and inexpensive processing, hybrid Si nanocone/polymer solar cells are promising as an economically viable alternative energy solution.

KEYWORDS: Nanotexture, solar cell, heterojunction, conductive polymer, light trapping



The significant growth of the Si photovoltaic industry has been limited due to the high cost of the Si photovoltaic system. In order to make it an economically viable energy solution, its module cost, which was below \$1.1/watt in late 2011, needs to be reduced by more than 50%. According to the U.S. Department of Energy, about 32% of the module cost comes from the Si material.¹ Thus, there is great interest in reducing the thickness of the Si layer, which requires considerable improvement in light absorption. One promising approach for the light absorption enhancement is light trapping through nanoscale texturing, such as nanowire,^{2–5} nanocone,^{6–9} or nanodome¹⁰ structures. These structures have demonstrated significant light absorption improvement for various solar cells, and in particular, the effect of the nanoscale texturing is more important for thin-film solar cells compared to a traditional microscale texturing for a thick Si solar cell. Additionally, about 26% of the module cost comes from the fabrication processes of a Si solar cell.¹ Conventional Si solar cells have p–n junctions inside for an efficient extraction of light-generated charge carriers. However, the p–n junction is normally formed by ion implantation and annealing processes or dopant diffusion processes, both of which are very expensive and require very high temperatures (~ 1000 °C). Thus, there has been significant interest in forming a heterojunction at low temperatures. One method is to make a solar cell composed of a single-crystalline Si wafer surrounded by ultrathin amorphous Si layers, and it is called a HIT (heterojunction with intrinsic

thin layer) solar cell. The fabrication temperature is less than 200 °C, and its open-circuit voltage (V_{OC}) is 743 mV, which is the world record for a single-junction Si solar cell.¹¹ Another method for the low-temperature heterojunction solar cell is to combine an organic material with Si.^{12–14} The organic material deposited by a solution-processed method forms a Schottky junction with Si, which replaces the expensive Si p–n junction. The commonly used organic material for the hybrid device is a conjugated polymer, poly(3,4-ethylene dioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), which is transparent and conductive (< 1000 S/cm). Most of the incoming light is absorbed in Si; thus, the efficiency of the hybrid Si/organic solar cell may in principle be comparable to a conventional Si p–n junction solar cell. In order to reduce the absorber material thickness and fabricate a device with inexpensive methods at low temperatures, hybrid solar cells composed of Si nanowires and polymers have been developed recently with power conversion efficiencies up to 10%.^{15–20} They exploit the advantages of nanoscale texturing for enhancement of light absorption and a hybrid structure for low-cost processes. However, the space between Si nanowires is normally too small to be filled with the conductive polymer, PEDOT:PSS; thus,

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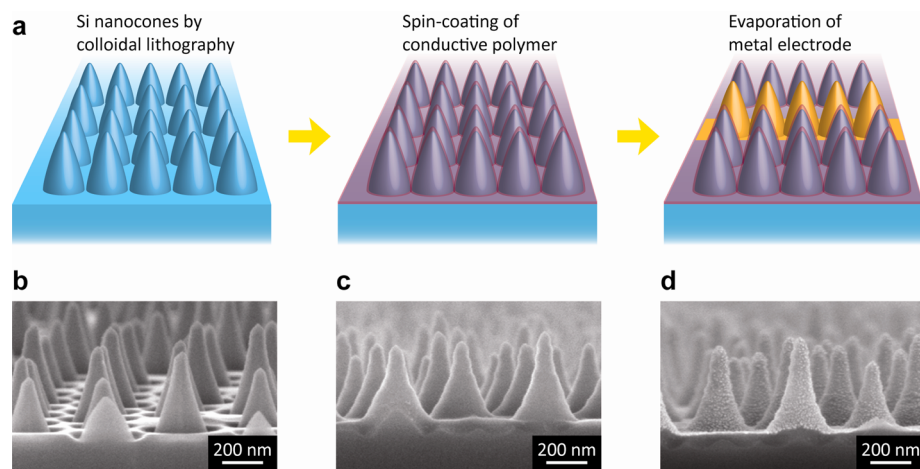


Figure 1. (a) Schematic illustration of the fabrication process for a Si nanocone/polymer solar cell. SEM images of cross-sectional view of Si nanocones (b) after a nanosphere lithography and RIE, (c) after a spin-coating of PEDOT:PSS with a spin speed of 4000 rpm, and (d) after evaporation of a metal electrode, Au, with a thickness of 15 nm.

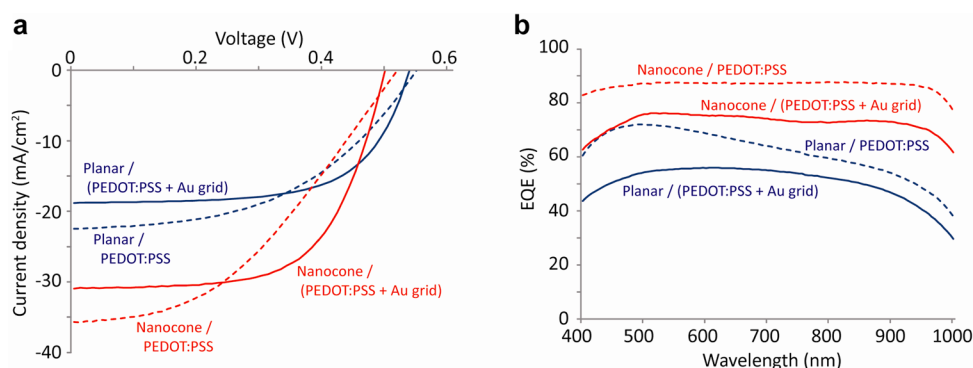


Figure 2. Characteristics of hybrid Si/polymer solar cells. (a) Current density–voltage (J – V) characteristics of four devices: planar Si coated with PEDOT:PSS, planar Si coated with PEDOT:PSS and Au grid, Si nanocones coated with PEDOT:PSS, and Si nanocones coated with PEDOT:PSS and Au grid. (b) EQE spectra of the four devices in (a).

another organic layer of small conjugated molecules was needed to provide complete surface coverage.

In this study, we demonstrate a hybrid solar cell composed of periodic Si nanocones and PEDOT:PSS. The nanocone structure with an aspect ratio (height/diameter of a nanocone) less than two was an optimized shape for the light absorption enhancement because it provided both excellent antireflection (for short wavelengths of light) and light scattering (for long wavelengths of light) effects. Additionally, this tapered structure allowed for conformal polymer surface coverage via spin-coating. The improved optical properties resulted in a very high short-circuit current density, 35.6 mA/cm², which is only 17% lower than the world record for a monocrystalline silicon cell, 42.7 mA/cm².²¹ Figure 1 shows a schematic representation of the fabrication process and scanning electron microscope (SEM) images of the Si nanocone/PEDOT:PSS solar cell at various stages. The Si nanocones used in this study were fabricated by nanosphere lithography.²² First, SiO₂ nanoparticles synthesized by a modified Stober process²³ were deposited as a monolayer on a n-type Si substrate with a thickness of 500 μm. Various methods to make a monolayer of nanoparticles on a Si substrate have been developed,^{24–27} and we used Langmuir–Blodgett assembly in this study because it provides the most uniform coverage over large areas. Oxygen (O₂) and trifluoromethane (CHF₃) plasmas were used to reduce the SiO₂ nanoparticle diameter, followed by chlorine

(Cl₂) and hydrogen bromide (HBr) plasma etching for the pattern transfer into the Si substrate.²⁵ Figure 1b is a SEM image of Si nanocones fabricated with 400 nm diameter SiO₂ nanoparticles. After the nanocone fabrication, the Si substrate was cleaned with piranha solution for 20 min and hydrofluoric (HF) acid (2%) for 1 min. In particular, the HF acid cleaning step is critical because it removes any native SiO₂ layer on top of the Si and any residue of SiO₂ nanoparticles which might be left after etching. Immediately after the HF cleaning and drying steps, the polymer, PEDOT:PSS, was deposited by spin-coating (4000 rpm, 1 min) in air. As shown in the SEM image (Figure 1c), the polymer deposited by the spin-coating method formed a conformal film with a thickness of ~60 nm over the Si nanocone structure. This result demonstrates one big advantage of the hybrid Si nanocone/polymer solar cell. Because of the conformal coating of PEDOT:PSS over the Si nanocones, other intermediate organic materials are not needed for full coverage. Although the polymer, PEDOT:PSS, is conductive enough to make a Schottky junction to separate light-generated charge carriers from the Si substrate, it is not conductive enough to be used as a top electrode. We used a finger-grid thin film of gold (Au) as a top electrode (Figure 1d). The width of each finger was 80 μm, and the spacing between fingers was 450 μm.

Figure 2a shows the current density–voltage (J – V) characteristics of the four devices: Si planar/PEDOT:PSS, Si planar/PEDOT:PSS with a Au finger grid, Si nanocone/

PEDOT:PSS, and Si nanocone/PEDOT:PSS with a Au finger grid. They were measured under air mass (AM) 1.5 illumination with a power of 100 mW/cm², and the parameters are summarized in Table 1. The short-circuit current density

Table 1. Photovoltaic Properties of the Hybrid Si Planar and Nanocone Solar Cells

	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	PCE (%)
planar/PEDOT:PSS	22.5	0.55	48.0	5.92
planar/(PEDOT:PSS + Au grid)	18.8	0.54	64.1	6.47
nanocone/PEDOT:PSS	35.6	0.51	41.3	7.54
nanocone/(PEDOT:PSS + Au grid)	31.0	0.50	62.6	9.62

(J_{sc}) increased by 52.1% from the planar to nanocone solar cell, which is consistent with the improved external quantum efficiency (EQE) of these devices (Figure 2b). The EQE of the nanocone device is above 80% over a wide wavelength range between 400 and 950 nm, but the planar device has a maximum EQE of 72% at 500 nm and a severe decrease from 500 to 1000 nm. We attribute the large improvement at longer wavelengths to the antireflection effect from the nanocones, which is not sensitive to the wavelength of the incoming light.²⁸ The nanocone structure led to a 7.3% decrease in open-circuit voltage (V_{oc}), which can be explained by the increased interface area between the polymer and Si. Even though the conformal coating of the polymer can provide some surface passivation,^{17,29} increased interface area due to the nanoscale texturing will still lead to some increased recombination. The Au grid (30 nm thick) deposited by e-beam evaporation on the nanocone device decreased the light absorption due to shadowing losses, especially, in the range of 400–500 nm wavelengths, but it improved the fill factor (FF) by 51.6% due to a decrease in series resistance from 4.55 to 0.83 $\Omega\cdot\text{cm}^2$. The change of the shunt resistance was not significant (from 1652 to 1776 $\Omega\cdot\text{cm}^2$).

The power conversion efficiency of 9.62% of the device fabricated by the simple and low-temperature process was quite high among hybrid Si/polymer solar cells, but the V_{oc} of the device was still low; that of the planar Si/PEDOT:PSS device was 0.55 V, which is 7% lower than that of the early study.²⁹ Based on the current–voltage measurement between Al pads deposited by e-beam evaporation at the back side of the Si, we found that the Si substrate did not have an ohmic contact with Al (Supporting Information Figure 1). Nonohmic contacts cause high contact resistance between the Si and the back electrode, which can deteriorate the device performance. There are various methods to make an ohmic contact, among which forming a highly doped Si layer is commonly used for high-efficiency Si solar cells. The highly doped layer not only decreases the contact resistance but also forms a built-in electric field on the back side that deflects minority carriers and reduces the recombination rate at the back surface, which can significantly lowers the saturation current density (J_0) and improve the V_{oc} .³⁰ In this report, we made the back surface of the Si substrate as a very highly doped n-type layer through a diffusion of phosphoryl chloride (POCl₃) gas. It is a high-temperature process (950 °C) which should be avoided for the cost reduction. However, the main point of this report is to demonstrate the Schottky junction between the polymer and Si to extract photon-generated charge carriers effectively. There can be different methods for the ohmic contact at the back side

of the Si, but in this study, we used this easily accessible method. After a thin, highly doped ($8 \times 10^{20} \text{ cm}^{-3}$) n-type layer was formed by a diffusion process at the back surface of the Si substrate for the nanocone device, the V_{oc} increased from 0.50 to 0.55 V (Figure 3a and Table 2). This enhancement of the

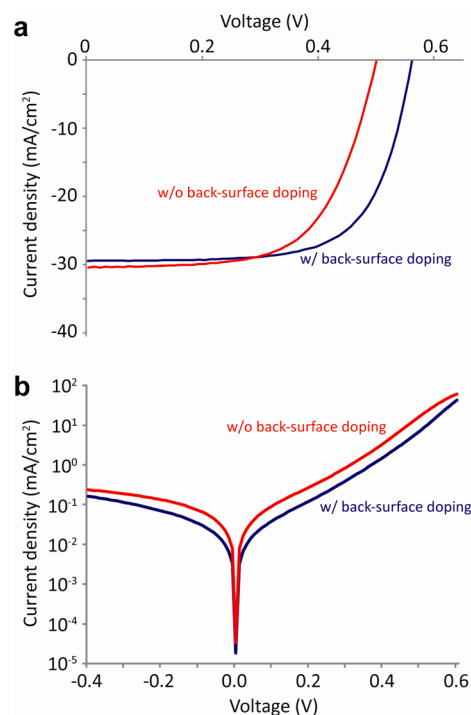


Figure 3. Effect of a back-side metal electrode contact. (a) Current density–voltage (J – V) characteristics of the Si nanocone/PEDOT:PSS solar cells, under AM 1.5 illumination, with and without a back-surface doping. The highly doped back surface helps to make an ohmic contact between the Si substrate and its metal electrode. (b) Dark current density–voltage (J – V) characteristics of the devices in (a).

Table 2. Photovoltaic Properties of the Hybrid Si Nanocone Solar Cells with and without a Back-Surface Doping

	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	PCE (%)
w/o back-surface doping	31.0	0.50	62.6	9.62
w/back-surface doping	29.6	0.55	67.7	11.1
improvement (%)	–4.5	+10.0	+8.1	+15.3

V_{oc} can be analyzed with the dark currents of the devices (Figure 3b). The saturation current density (J_0) of the device after the back-surface doping was 0.13 $\mu\text{A}/\text{cm}^2$, which was much lower than that of the device without the doping, 1.07 $\mu\text{A}/\text{cm}^2$. In order to achieve the high V_{oc} , the J_0 needs to be very low; a well-designed heterojunction Si solar cell with a V_{oc} higher than 0.7 V has J_0 less than 1 nA/cm².³¹ The back-surface doping layer in the nanocone device improved the V_{oc} from 0.50 to 0.55 V, but for better performance, there needs to be further investigation to improve the V_{oc} .

In addition to the V_{oc} , we improved the FF up to 67.7% with a thicker Au finger grid. The nanocone device without the back-surface doping layer had a 30 nm thick Au grid, but the one with the doping layer had 80 nm of Au, which decreased the series resistance of the device from 0.83 to 0.44 $\Omega\cdot\text{cm}^2$. However, the thicker Au resulted in a 4.5% decrease in J_{sc} , from 31.0 to 29.6 mA/cm², stemming from the difference in

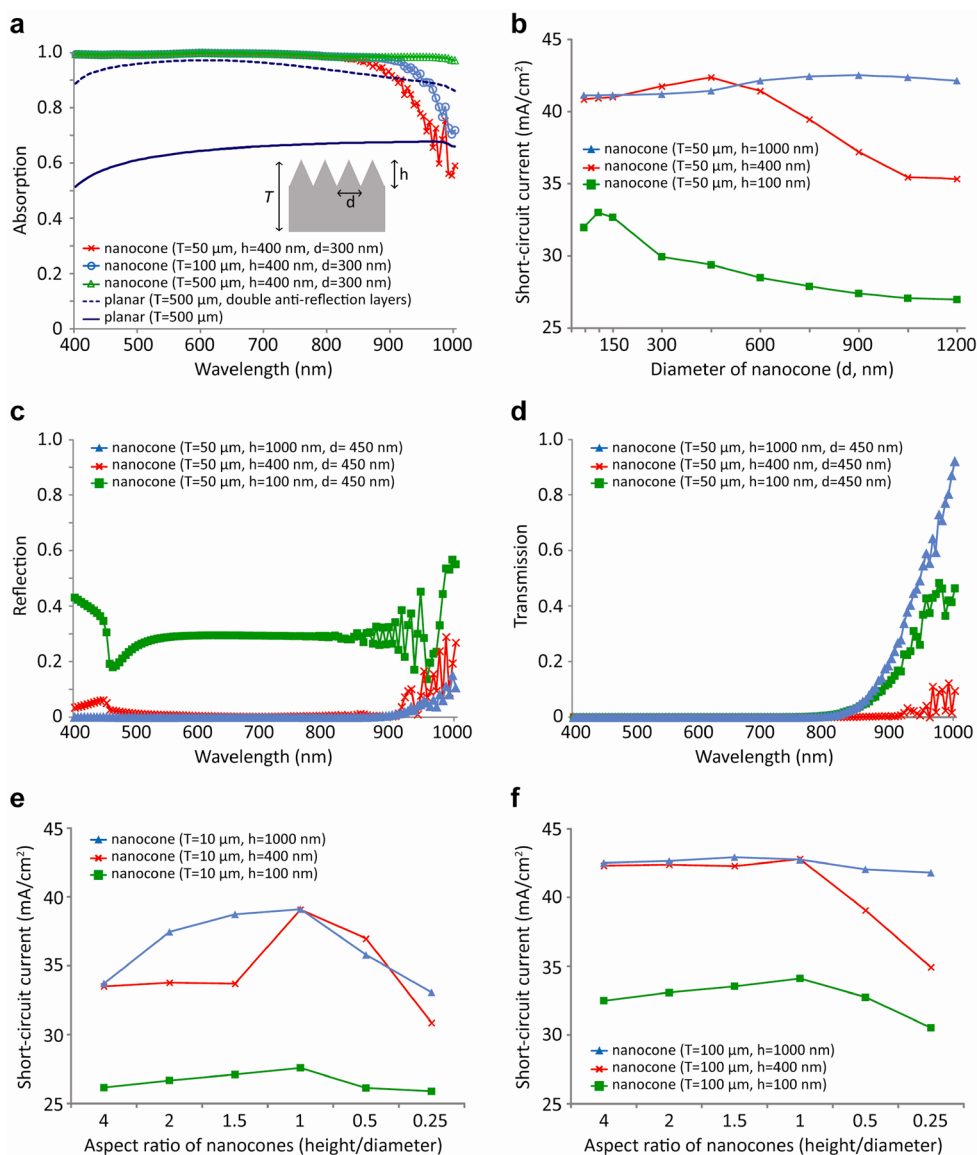


Figure 4. Simulation of optical properties for different nanocones and planar structures. (a) Absorption data of 50, 100, and 500 μm thick Si substrates with nanocone dimensions similar to those that we fabricated for the hybrid solar cells, a 500 μm thick, planar Si substrate with double antireflection layers (100 nm thick SiO_2 followed by 60 nm thick Si_3N_4), and a planar 500 μm thick Si substrate without any coating. (b) Calculated J_{SC} of nanocone structures with different sizes. The total thickness of the device is 50 μm . (c, d) Reflection and transmission data of the three nanocone structures in (b). (e, f) Calculated J_{SC} of nanocone structures plotted as a function of their aspect ratios. The total thickness of the device is (e) 10 μm and (f) 100 μm .

optical transmission through the Au fingers. With improvements of the V_{OC} and FF, we achieved an efficiency of 11.1% from the hybrid Si nanocone/polymer solar cell.

The substantial improvement in J_{SC} (>52% as indicated in Table 1) is mainly due to the significant antireflection and light-trapping effects caused by the nanocone structure. In order to study these optical effects in more detail, we performed simulations using a rigorous coupled wave analysis (RCWA) algorithm.^{32,33} The simulation data, shown in Figure 4a, compare the light absorption of five devices: 50, 100, and 500 μm thick Si substrates with nanocone dimensions similar to those that we fabricated in this study, a planar 500 μm thick Si substrate coated with a conventional antireflection double layer (100 nm thick SiO_2 film/60 nm thick Si_3N_4 film), and a planar 500 μm thick Si substrate without any coating. The 500 μm thick Si substrate with the nanocones absorbed the visible light

almost completely, which is better than the antireflection coated planar substrate with the same thickness. The superior light-trapping and antireflection properties of the nanocones compared to the conventional antireflection film were found even in thinner substrates. The light absorption improvements of the 50 and 100 μm thick Si substrates with the nanocones are 47% and 51%, respectively, compared to that of the planar 500 μm thick Si substrate without any coating. These improvements are even better than that of the planar 500 μm thick Si substrate with the antireflection double layer (45%). Previous work has demonstrated that nanocones or nanotips with high aspect ratios show remarkably low reflection due to the tapered structure.^{7,28} Here, we also confirm that the nanocone structure with a higher aspect ratio has superior antireflection properties to the one with a lower aspect ratio (Supporting Information Figure 2a,c). The calculated J_{SC} with

an assumption of 100% internal quantum efficiency demonstrates a similar aspect ratio effect for various nanocone sizes: higher aspect ratio nanocones have higher J_{SC} (Supporting Information Figure 2b,e). However, this trend does not appear for nanocones on thin substrates: when the total substrate thickness is 50 μm , the J_{SC} from 400 nm height nanocones becomes larger as their diameter increases from 50 to 450 nm but becomes smaller as the diameter increases from 450 to 1200 nm (Figure 4b, red line). In other words, the light absorption is maximized with an aspect ratio equal to 0.9, and it decreases as the ratio decreases or increases from 0.9. Similar trends were found for different nanocone sizes (Figure 4b, blue and green lines). From these results, it can be concluded that the light absorption increases as the aspect ratio of the nanocones approaches one. This surprising result can be explained by the scattering effect of the nanocone, which increases the optical path length inside the material; because of the scattering, the light can propagate more laterally, which results in higher light absorption. This is confirmed by the slightly higher reflection but substantially lower transmission of light from the 400 nm height nanocone compared to the 1000 nm one (Figure 4c,d). As the substrate becomes thinner, the light trapping effect which increases the optical path length becomes more important. Thus, nanocones with an aspect ratio around one provide the best balance between the antireflection and light trapping effects for thin silicon substrates (Figure 4e). For thick Si substrates, light trapping is less since most of the light can be absorbed in a single pass (Supporting Information Figure 2d), and light absorption only depends on the antireflection effect (Figure 4f and Supporting Information Figure 2e). This information is critical for a thin, hybrid Si/polymer solar cell: for the hybrid device, a conformal coating of the polymer over Si forms a Schottky junction for charge-carrier extraction, but it has been demonstrated that the polymer has problems coating high aspect ratio structures, such as nanowires. Therefore, our nanocone structure is ideal for thin hybrid solar cells when considering antireflection, light trapping, and polymer coating properties.

In conclusion, we demonstrated a hybrid Si nanocone/polymer solar cell with a power conversion efficiency of 11.1%, which is the highest among hybrid Si/organic solar cells to our knowledge. The conductive polymer, PEDOT:PSS, made a Schottky junction with Si to extract light-generated charge carriers while passivating the Si surface. This junction was formed by a simple solution-processed method at a temperature as low as 120 $^{\circ}\text{C}$. Based on our simulation study about light absorption, the optimum nanocone structures for a thin-film Si solar cell need to have their aspect ratios around one. This structure is well-suited for the hybrid Si/polymer solar cell since it allows for conformal polymer coating.

Methods. *Fabrication of Si Nanocones by RIE Method.* After a monolayer of SiO_2 nanoparticles (400 nm diameter) was formed on a n-type Si substrate, the nanoparticles were etched isotropically to reduce the diameter to about 250 nm diameter by RIE. Oxygen (O_2) and trifluoromethane (CHF_3) were used with flow rates of 6 and 85 sccm, respectively. After the etching process, the Si substrate below the SiO_2 nanoparticles was etched to make an array of nanocones. The Si was etched by a mixture of chlorine (Cl_2) and hydrogen bromide (HBr) gas with flow rates of 40 and 100 sccm, respectively. The etching was conducted for 2 min with a radio-frequency power of 250 W.

Preparation of PEDOT:PSS Solution. PEDOT:PSS (PH1000 from Clevios) solution was mixed with 5 wt % dimethyl sulfoxide (DMSO from Fisher Scientific) and 1 wt % surfactant (Zonyl-FSH from Dupont).

Fabrication of a Thin, Highly Doped n-Type Layer at the Back Side of a Si Substrate. The polished surface of a n-type Si wafer with a doping concentration of $4 \times 10^{15} \text{ cm}^{-3}$ was covered with a thermally grown oxide layer, and the back surface was not covered to make a highly doped layer on it. Then, the wafer was put into a diffusion furnace with a flow of phosphoryl chloride (POCl_3) gas. The diffusion step was conducted at 950 $^{\circ}\text{C}$ for 30 min, which resulted in an ~ 220 nm depth of the doping layer. The concentration of the dopant at the surface matched the solid solubility of phosphorus (P) at the diffusion temperature ($8 \times 10^{20} \text{ cm}^{-3}$ at 950 $^{\circ}\text{C}$).

■ ASSOCIATED CONTENT

Supporting Information

Figures 1–3. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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Notes

The authors declare no competing financial interest.

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