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Flexible solar cells based on PEDOT:PSS and vertically aligned silicon structures

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Abstract. Photovoltaic properties of hybrid solar cells based on poly-(3,4 ethylenedioxythiophene): polystyrene sulfonate (PEDOT:PSS) and Si nanowires (SiNWs) are studied. High values of the open circuit voltage (V_{OC}) and external quantum efficiency (EQE) at short wavelength region obtained for planar solar cells indicate sufficient passivation properties of *n*-Si/PEDOT:PSS interface. A technology for filling SiNWs (6 μm in height and 1.7 μm in diameter) with PEDOT:PSS has been developed using G-coating. Compared with planar hybrid cell, SiNWs/PEDOT:PSS cell exhibit lower total reflectance (~12%) and higher EQE in the long wavelength region. It should be stressed that an increase in the PEDOT:PSS layer thickness by the combination of the G-coating and spin coating methods does not affect the short wavelength region of EQE. This fact is important for development of flexible solar cells based on SiNWs.

Keywords: solar cell, silicon nanowires, PEDOT:PSS, flexibility

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Гибкие солнечные элементы на основе PEDOT:PSS и вертикально-ориентированных кремниевых структур

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Аннотация. Изучены фотоэлектрические свойства гибридных солнечных элементов на основе кремниевых нановолокон (SiNWs) и поли(3,4-этилендиокситиофен)-полистиролсульфоната (PEDOT:PSS). Высокие значения напряжения холостого хода (V_{xx}) и внешней квантовой эффективности в коротковолновой области, полученные для планарных солнечных элементов, указывают на достаточные пассивирующие свойства интерфейса *n*-Si/PEDOT:PSS. Технология заполнения SiNWs (6 мкм в высоту и 1.7 мкм в диаметре) слоем PEDOT:PSS была разработана с использованием метода G-центрифугирования. По сравнению с подобным кремниевым планарным элементом, солнечный элемент с радиальным p—n-переходом демонстрирует гораздо более низкий общий коэффициент отражения (~ 12%) и более высокую квантовую эффективность в диапазоне длин волн 430—1200 нм. Следует подчеркнуть, что увеличение толщины слоя PEDOT:PSS за счет комбинации G- и горизонтального центрифугирования не влияет на коротковолновую область EQE. Этот факт важен для разработки гибких солнечных элементов на основе вертикально-ориентированных структур.

Ключевые слова: солнечный элемент, кремниевые нановолокна, PEDOT:PSS, гибкость

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Introduction

Presently, a considerable interest is drawn to the development of renewable energy sources due to the gradual depletion of traditional energy sources and strong requirements to reduce carbon footprint. One of the most promising branches of renewable energy is solar power harvested by photovoltaic devices [1]. Crystalline silicon solar cells dominate the photovoltaic industry. However, these solar cells are rigid and bulky.

Therefore, the transition to the technology of flexible solar cells is promising [2—4]. First, such solar cells have trade-off efficiency and robustness. Besides, expensive glass substrates can be replaced with polymer ones. Secondly, flexible panels are lighter than rigid crystal ones, which makes them easier to transport and install. Third, it is possible to integrate such devices into various surfaces, such as smartphone screens, clothing, the electric car, etc. However, modern flexible solar cells may suffer from several problems. This is due to the use of unstable, expensive or toxic materials. Flexible solar cells based on perovskites are attractive for their increased optical absorption, but they degrade quickly. Flexible III-V tandem solar cells allow expanding the optical absorption spectrum. However, their wide use is limited by the complexity of the technology and the high cost. Promising flexible Copper Indium Gallium Selenide (CIGS) solar cells are includes rare (In) and expensive (Ga) elements may limiting its large-scale usage. Besides, indium-tin-oxide (ITO) is most often used as a transparent electrode of solar cell. ITO can be applied onto flexible plastic substrates [5, 6], but very thin layer and under numerous bending cycles ITO is cracking [7, 8] which significantly reduces of the performance cells. Besides, rare element In may prevent wide implementation of ITO. Hence, new concepts need to be exploited to address these concerns.

An alternative way is to create SiNWs/PEDOT:PSS hybrid solar cells. The SiNWs provides broadband antireflection and have a good light-trapping effect, which can increase the optical absorption of solar radiation in active layers [9—12]. The SiNWs solar cells embedded in polymer matrix have enhanced mechanical stability compared to conventional planar tandem solar cells deposited on flexible substrate. Polymer PEDOT:PSS, in turn, is promising as a transparent electrode material [13]. This layer possess is more flexibility than ITO and can exhibit values of conductivity and transmittance close to those of ITO [7, 14]. Moreover, polymer PEDOT:PSS is a more attractive emitter material for SiNWs compared to *a*-Si:H layer because it is difficult to deposit (*p*)*a*-Si:H/(*i*)*a*-Si:H layer stack with precise thickness control on vertically aligned silicon structures. Besides, using PEDOT:PSS layer is low cost due to low-temperature processing such as spin coating. Thus, combine both of SiNWs and PEDOT:PSS advantages provide a possible simplify production processes and its low costs.

In this work, hybrid solar cells based on PEDOT:PSS with SiNWs structures are reported.

Experimental section

The *n*-type Si (100) wafers with a resistivity of 0.2 Ω·cm and 0.06 Ω·cm were used for solar cells fabrication. The layer of *n*-type doped μc-Si was deposited on the backside of Si substrates by PECVD to form ohmic contact. In order to verify the quality of the hybrid heterojunction, first, the planar Si/PEDOT:PSS solar cell structures were fabricated.

i) Planar Si/PEDOT:PSS solar cell

Previously the Si substrates were dipped in a 10 vol% hydrofluoric (HF) acid solution for 30 s to remove the native silicon oxide at the surface. Two types of PEDOT:PSS based solutions were tested to form emitter layer: 1) PEDOT:PSS with a dispersion content of 1wt% in water mixed with 5 wt% dimethyl sulfoxide (DMSO) to increase conductivity [15]; 2) PEDOT:PSS (DMSO) mixed with 0.25 wt% of Neonol AF 9-12, which was used as a surfactant to improve the adhesion to Si surface. The PEDOT:PSS based solutions were spin coated on Si substrates at a rate of 1000 rpm during 1 min. Then samples were annealed on a hot plate at 120 °C for 2 min in air. The Ag electrodes were applied on the organic PEDOT:PSS layer by vacuum evaporation.

Next, hybrid Si/PEDOT:PSS solar cell were fabricated based on array of Si nanowires.

ii) SiNWs/PEDOT:PSS solar cell

The array of SiNWs was obtained by deep cryogenic etching of Si in a SF_6/O_2 plasma using Oxford PlasmaLab System100 ICP380 [16–17]. The SiNWs samples were dipped in a 10 vol% hydrofluoric (HF) acid solution for 30 s to remove the native oxide on the SiNWs surface immediately prior PEDOT:PSS deposition. The both types of PEDOT:PSS based solutions described above for planar structures were used. The PEDOT:PSS layer initially was spin coated on the samples using EZ4 Spin Coater at a rate of 1000 rpm. During spin coating in spinner the PEDOT:PSS layer covered only the tops of the SiNWs (Fig. 1, *a*). It should be noted that reducing the viscosity of the PEDOT:PSS solution (Neonol addition) just slightly improve the process of its penetration between the wires.

Next, we used the G-coating method (Fig. 1, *b*) [18] to fill the area between SiNWs array with the use of swinging bucket centrifuge. It can be noted that during G-coating the PEDOT:PSS layer covers the nanowires along entire length. This is due to the high pressure (G-force) applied to the polymer PEDOT:PSS. Then samples were annealed at 120 °C and Ag electrodes were formed.

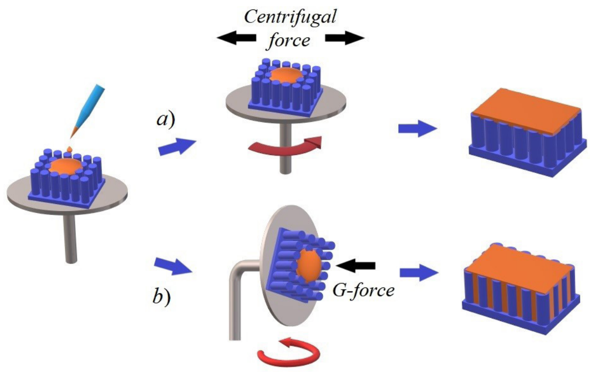

Fig. 1. Schematic of the spin coating (*a*) and G-coating (*b*) of the PEDOT:PSS layer

Besides, we created a structure by a combination of G-coating and spin coating methods. Initially the PEDOT:PSS layer on samples was G-coated on the samples at a rate of 5000 rpm for 5 min. This made it possible to fill the area between SiNWs. Then the PEDOT:PSS layer was spin coated at a rate of 1000 rpm for 1 min. This made it possible to cover the tops of the SiNWs as well.

The current–voltage (*I–V*) measurements were performed using an Abet Technologies solar simulator (AM 1.5G, 100 mW/cm^2) and a Keithley 2400 electrometer. The external quantum efficiency (EQE) spectra were obtained using an SLS M266 monochromator, a halogen lamp and a Si reference cell. The total reflectance spectra of the cells were measured using an integrating sphere and spectrometer AvaSpec SensLine.

Results and Discussion

i) Planar Si/PEDOT:PSS solar cell

Figure 2 compares the total reflectance spectra of the planar Si and SiNWs structures with and without PEDOT:PSS layer. The bare planar Si structure exhibits the total reflectance of more than 23% from 400 to 800 nm. The bare SiNWs structure exhibits the total reflectance at around ~ 15%. Thus, using SiNWs structures can help to improve the light absorption. It should be noted that for the samples with PEDOT:PSS the reflectance is decreased. The SiNWs structure with PEDOT:PSS layer exhibits the reflectance at around ~ 12%. This means that the PEDOT:PSS film exhibits antireflection properties.

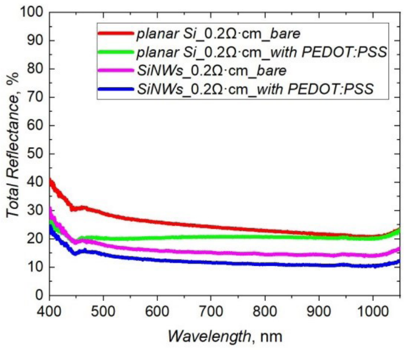

Fig. 2. Measured total reflectance spectra of the Si structures

The current density-voltage (*J*—*V*) curves and photovoltaic parameters of the planar Si/PEDOT:PSS hybrid cells are summarized in Fig. 3, *a* and Table 1, respectively. The photovoltaic parameters such as the open circuit voltage (V_{OC}) and the fill factor (FF) were calculated from the illuminated *I*—*V* curves. The short circuit current density (J_{SC}) was extracted from the quantum efficiency spectra.

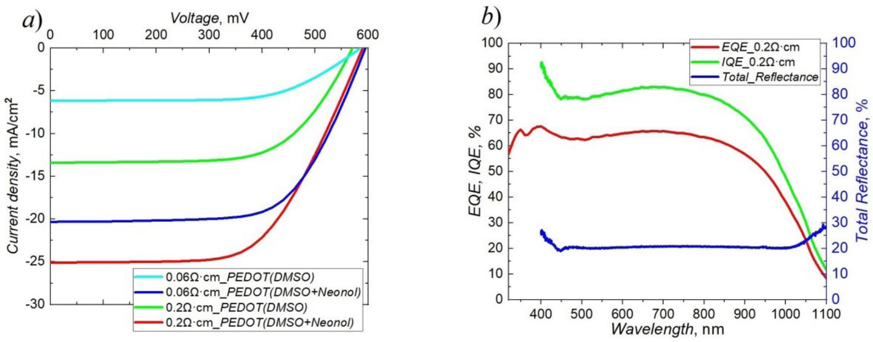

Fig. 3. Measured *J*—*V* curves (*a*) and quantum efficiency, and total reflectance spectra (*b*) of the planar Si/PEDOT:PSS hybrid cells

Table 1

Photovoltaic parameters of the planar Si/PEDOT:PSS cell

Sample	Wafer resistivity	Co-solvents of PEDOT:PSS	V_{OC}, mV	J_{SC}, mA/cm^2	FF, %
Planar Si	0.2 Ω·cm	DMSO	571	13.35	64.68
	0.2 Ω·cm	DMSO + Neonol	589	24.96	60
	0.06 Ω·cm	DMSO	584	6.16	63.15
	0.06 Ω·cm	DMSO + Neonol	595	20.22	65.03

The structure with a DMSO addition exhibits the J_{SC} of 13.35 mA/cm², the V_{OC} of 571 mV and the FF of 64.68 %. Compared with this structure, the cell with DMSO and Neonol AF 9-12 addition has improved in all photovoltaic parameters. It inferred that the Neonol AF 9-12 can improve the wettability and conductivity of the PEDOT:PSS layer and thus enhance the photovoltaic parameters of the planar structure.

The V_{OC} at around 589 mV and 595 mV were obtained for Si wafers (PEDOT:PSS with DMSO and Neonol AF 9-12 addition) with a resistivity of 0.2 Ω·cm and 0.06 Ω·cm, respectively. With the decrease of resistivity Si wafers the J_{SC} also decreases to 20.22 mA/cm². It can be explained by a shorter lifetime of minority carriers in the Si substrate. These results agree well with the results in Sara Jäckle et.al [19].

Figure 3, *b* shows quantum efficiency and reflectance spectra of the planar Si/PEDOT:PSS cells with DMSO and Neonol AF 9-12 addition. Since the total reflectance spectra was measured in the wavelength range from 400 to 1200 nm, the values of the internal quantum efficiency are calculated in the same wavelength range.

Spectral response measurements (Fig. 3, *b*) demonstrate extremely high quantum efficiency in UV region spectrum below 400 nm meaning low absorption losses in PEDOT:PSS layer. Moreover, high values of the V_{OC} and quantum efficiency at short wavelength region indicate low recombination losses at the Si/PEDOT:PSS planar interface. Thus, PEDOT:PSS is a promising candidate to use as an emitter layer for SiNWs structure because of sufficient passivation properties.

ii) SiNWs/PEDOT:PSS solar cell

The *J—V* curves of the SiNWs/PEDOT:PSS hybrid cells are summarized in Fig. 4, *a*. The photovoltaic parameters such as V_{OC} and FF extracted from the illuminated *I—V* curves are collected in Table 2. The J_{SC} values were calculated from the quantum efficiency spectrum and are also presented in Table 2. It should be noted that the addition of Neonol AF 9-12 reduced the photovoltaic parameters of the SiNWs/PEDOT:PSS hybrid cells. Besides, the addition of Neonol AF 9-12 reduces the viscosity of the solution, and therefore, it is more difficult to control the process of coating the PEDOT:PSS layer on the SiNWs.

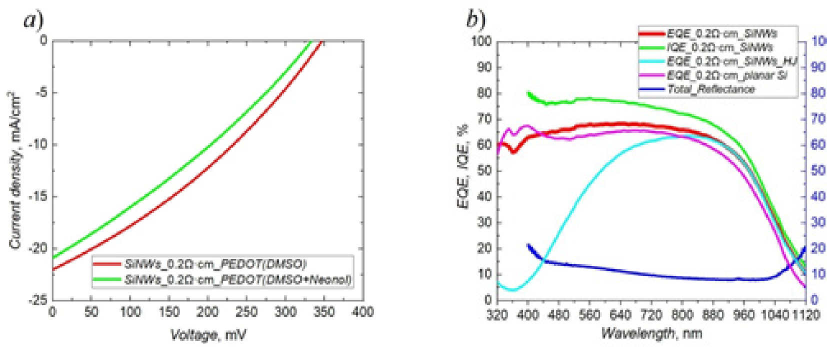

Fig. 4. Measured *J—V* curves (*a*) and quantum efficiency, and total reflectance spectra (*b*) of the SiNWs/PEDOT:PSS hybrid cells

Table 2

Summary of photovoltaic parameters of the SiNWs/PEDOT:PSS hybrid cells

Sample	Wafer resistivity	Co-solvents of PEDOT:PSS	V_{OC}, mV	J_{SC}, mA/cm²	FF, %
SiNWs	0.2 Ω·cm	DMSO	350	22.02	34
	0.2 Ω·cm	DMSO + Neonol	330	20.79	37.75
HJ (SiNWs)	0.2 Ω·cm	DMSO	440	20.41	–

The SiNWs structure with DMSO addition exhibits the J_{SC} of 22.02 mA/cm², the V_{OC} of 350 mV and the FF of 34%. The obtained values of V_{OC}, J_{SC} and FF are lower than for planar Si cells. It can be explained by the fact that high aspect ratios of SiNWs surface properties start to define the electrical characteristics. The SiNWs/PEDOT:PSS cell performance could be improved by a better SiNWs surface preparation.

Figure 4, *b* shows quantum efficiency and total reflectance spectra of the SiNWs/PEDOT:PSS cells with DMSO. Compared to planar Si/PEDOT:PSS cell, SiNWs/PEDOT:PSS cell exhibit much higher external quantum efficiency in the wavelength range from 430 to 1200 nm due to enhanced light trapping.

Then, we considered the PEDOT:PSS layer as a transparent electrode material. Silicon heterojunction (HJ) structures based on SiNWs and stack of thin *i*- and *p*- layers amorphous hydrogenated silicon (*a*-Si:H) thin film were fabricated. A strong drop of the EQE in the wavelength range from 350 to 800 nm is observed for heterojunction solar cells (see Fig. 4, *b*). It can be explained by significant absorption, and therefore, recombination losses in the *a*-Si:H layer. However, compared with hybrid SiNWs structure, the heterojunction cell has higher V_{OC} of 440 mV. On the one hand, it can be explained by better passivation properties of *a*-Si:H, which could provide excellent surface passivation [20]. On the other hand, the obtained value of V_{OC} is extremely low compared to that (above 0.7 V) of high-performance heterojunction solar cells. It should be stressed that the planar *a*-Si:H/c-Si solar cells with V_{OC} near to 0.7 V were fabricated using the same deposition conditions. It means that an additional treatment of the SiNWs surface is required to reduce concentration of defects or impurities created during cryogenic etching. Thus, there is a still a room for improvement for hybrid solar cell based on SiNWs. Besides, using thin *a*-Si:H passivation layer improves the wettability of PEDOT:PSS (DMSO). This fact opens a way to combine the advantages of heterojuction and hybrid technology. Deposition of ultra-thin intrinsic *a*-Si:H layer could provide excellent passivation and wettability of SiNWs surface, while PEDOT:PSS provides enhanced quantum efficiency in short wavelength region due to its transparency.

To provide the full filling of the SiNWs array by PEDOT:PSS a combination of the coating techniques was used. For one of the samples after the G-coating, the PEDOT:PSS (DMSO) layer was re-coated on the sample using the spin coating. Then the sample was annealed at 120 °C and Ag electrodes were formed. Figure 5 shows quantum efficiency spectra of the PEDOT:PSS/SiNWs hybrid cells with different methods of coating the PEDOT:PSS layer.

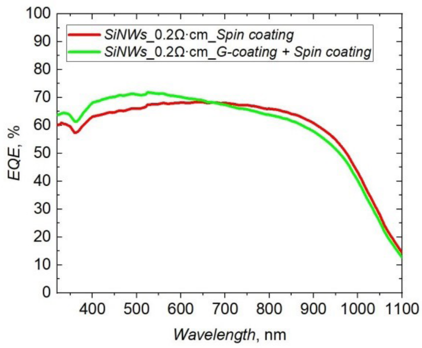

Fig. 5. Measured quantum efficiency of the PEDOT:PSS/SiNWs hybrid cells with different methods of coating the PEDOT:PSS layer

It should be noted that the EQE measurement results indicate that compared with the spin coating the PEDOT:PSS layer, the combination of the G-coating and spin coating methods exhibited a pronounced EQE enhancement in the spectral region of 320–660 nm. An increase in the PEDOT:PSS layer thickness does not affect the short wavelength region of EQE. This fact is important for development of flexible solar cells based on SiNWs.

Next, we examined the structure in relation to flexibility. For mechanical strength, nanowires were filled with Su-8 photoresist. Then the substrate was sunk from the backside first mechanically, and then by etching in plasma. The resulting devices are flexible with a bending radius of 25 mm. The resulting thickness of the substrate was 40 μm.

Conclusion

In summary, SiNWs/PEDOT:PSS hybrid solar cells where the PEDOT:PSS layer considered both as an emitter material and a transparent electrode were fabricated. It was succeeded to fill the area between SiNWs with the PEDOT:PSS by using G-coating for a SiNWs solar cells. Compared planar Si/PEDOT:PSS cell, SiNWs/PEDOT:PSS cell exhibit much lower total reflectance and higher EQE in the wavelength range of 430–1200 nm. Besides, SiNWs/PEDOT:PSS flexible structures were successfully fabricated.

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