

## Efficiency enhancement of polymer solar cells by incorporating a self-assembled layer of silver nanodisks

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### ARTICLE INFO

#### Article history:

Received 23 March 2011

Received in revised form

5 July 2011

Accepted 11 July 2011

Available online 15 August 2011

#### Keywords:

Polymer solar cell

Silver nanodisks

Localized surface plasmon resonance

### ABSTRACT

We report the efficiency enhancement of polymer solar cells by incorporating a silver nanodisks' self-assembled layer, which was grown on the indium tin oxide (ITO) surface by the electrostatic interaction between the silver particles and modified ITO. Polymer solar cells with a structure of ITO (with silver nanodisks)/poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) (Clevious P VP AI 4083)/poly(3-hexylthiophene):[6,6]-phenyl-C61 butyric acid methyl ester (P3HT:PC<sub>61</sub>BM)/LiF/Al exhibited an open circuit voltage ( $V_{OC}$ ) of  $0.61 \pm 0.01$  V, short-circuit current density ( $J_{SC}$ ) of  $9.24 \pm 0.09$  mA/cm<sup>2</sup>, a fill factor (FF) of  $0.60 \pm 0.01$ , and power conversion efficiency (PCE) of  $3.46 \pm 0.07\%$  under one sun of simulated air mass 1.5 global (AM1.5G) irradiation (100 mW/cm<sup>2</sup>). The PCE was increased from  $2.72 \pm 0.08\%$  of the devices without silver nanodisks to  $3.46 \pm 0.07\%$ , mainly from the improved photocurrent density as a result of the excited localized surface plasmon resonance (LSPR) induced by the silver nanodisks.

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### 1. Introduction

Organic solar cells have great potential as the future energy source due to the advantages of low cost, being mechanically flexible and suitable for large-area fabrication [1–7]. So far organic solar cells with power conversion efficiency (PCE) of 6% to 8% have been reported by incorporating novel low-band-gap polymers [8–10].

Because the photocurrent of a solar cell is directly related to the light harvesting, enhancing the light absorption is an efficient way to improve the short-current density ( $J_{SC}$ ), accordingly the PCE. In order to increase the absorption of the photoactive layer, one promising method is to introduce localized surface plasmon resonant (LSPR) metallic nanostructures into the bulk heterojunction (BHJ) solar cells [11–18]. It has been shown that the excitation of LSPR through the resonant interaction between the electromagnetic field of incident light and the surface charge of metallic nanostructures causes an electric field enhancement that can be coupled to the photoactive absorption region, and increases the absorption of photoactive conjugate polymer [19–24]. So far, several LSPR metallic nanostructures have been utilized for improving the performance of solar cells [25–27]. For instance, recently, Kim et al. [25] fabricated poly(3-hexylthiophene) (P3HT) and phenyl-C61-butyl-acid methyl ester (PC<sub>61</sub>BM) BHJ solar cell with surface plasmon-active silver nanoparticle layers on indium-tin oxide (ITO)

as anode, and the overall PCE was increased to 3.69% from 3.05%. Yoon et al. [26] fabricated polythiophene-fullerene BHJ solar cells using a plasmonic monolayer of silver nanospheres (diameter of ~4 nm) on ITO substrate as anode,  $J_{SC}$  increased to 7.0 mA/cm<sup>2</sup> from 6.2 mA/cm<sup>2</sup> due to the enhancement of the photogeneration of excitons near the plasmon resonance of the silver nanospheres. Kim et al. [27] obtained improved poly-3-octylthiophene (P3OT)/C<sub>60</sub> BHJ photovoltaic devices by doping with stable and highly electrical silver nanospheres. The doped devices showed 50–70% enhancement of efficiency than the undoped one.

However, the smaller (< 20 nm diameter) spherical silver particles they used lead to the weak surface plasmon resonances, due to the weak scattering, which could not couple and trap propagating plane waves efficiently from the Sun into the photoactive layer [16]. Furthermore, the plasmonic spherical silver particles, to some extent, could improve the harvest of photons from the solar spectrum, but the spectral mismatch between their plasmon resonances (typically below 450 nm) and the absorption region of P3HT leads to the weak absorption enhancement in photoactive conjugate polymer, which limited the further improved PCE in the BHJ solar cell with plasmonic spherical silver particles as interfacial layer. In addition, most of the spherical silver particles were made using electrodeposition technique, thermal evaporation, and doping method, which was not beneficial for the control over the particle's size/shape and consequently their plasmon resonances.

As usual, LSPR effect is dependent on several features of the nanoparticles, such as size, shape, composition, and crystallinity,

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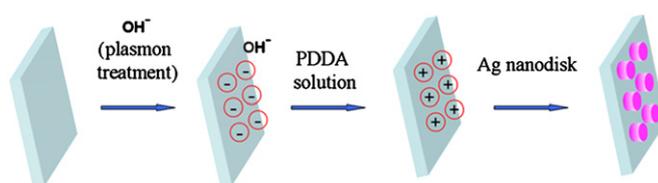
of which size and shape are more significant for optimizing the optical properties of nanoparticles for photovoltaic applications [28–30]. Solution-processable approaches are efficient for fabricating well-dispersed metallic nanostructures with control over the size, shape, and density of particles [28,31–34]. Colloidal nanoparticles synthesized by solution-processable approaches, such as silver nanodisks, could be processed from a variety of aqueous and organic solutions [29]. They could be synthesized in a variety of controlled sizes affording control over their optical properties. Furthermore, the LSPR peak of silver nanodisks could be tuned across the nearly entire visible (600–440 nm) region, which leads to the improved photocurrent density in BHJ solar cells as a result of enhanced absorption of the photoactive conjugate polymer, due to the better spectral overlap between the active layer absorption and the plasmon resonances of the nanoparticles, which cannot be easily done with spherical particles [29].

In this study, we report a simple method to grow silver nanodisks on ITO substrates by the electrostatic assembly. The silver nanodisks enhance the absorption of P3HT:PC<sub>61</sub>BM layer due to the LSPR effect. Solar cells with the self-assembled layer of silver nanodisks [ITO/silver nanodisks/poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS)/P3HT:PC<sub>61</sub>BM/LiF/Al] exhibited an open circuit voltage ( $V_{oc}$ ) of  $0.61 \pm 0.01$  V,  $J_{sc}$  of  $9.24 \pm 0.09$  mA/cm<sup>2</sup>, a fill factor (FF) of  $0.60 \pm 0.01$ , and a PCE of  $3.46 \pm 0.07\%$  under one sun of simulated air mass 1.5 global (AM1.5 G) irradiation (100 mW/cm<sup>2</sup>). The PCE was increased from  $2.72 \pm 0.08\%$  to  $3.46 \pm 0.07\%$  as compared to the devices without silver nanodisks self-assembled layer, mainly resulting from the improved photocurrent density as a result of the excited localized surface plasmon resonance induced by the silver nanodisks.

## 2. Experimental section

Silver nanodisks were synthesized using the photoinduced seed growth method [29]. The detailed synthesis procedure can be divided into three steps: first of all, silver seeds were prepared by dropwise addition of NaBH<sub>4</sub> solution (8.0 mM, 1.0 mL) to an aqueous solution of AgNO<sub>3</sub> (0.1 mM, 100 mL) in the presence of trisodium citrate (0.1 mM) under vigorous stirring. Then, the yellow silver seeds (100 mL, in glass conical flask with a stopper) were irradiated under a sodium lamp (NAV-T 70 model from Osram China Lighting Co., Ltd.). The color of silver colloids changed from yellow to green to blue during the irradiation process. The blue silver colloid transformed into purple silver nanodisks after 5 h irradiation. More trisodium citrate (0.1 M, 0.4 mL) was added into the as-prepared silver nanodisks (100 mL) to stabilize the nanoparticles. Finally, the synthesized nanodisks were capped with a layer of negatively charged trisodium citrate [35].

To grow silver nanodisks on ITO substrates, the ITO-coated glass substrates (20 Ω/sq) were cleaned by detergent, chloroform and acetone, and then dried under a N<sub>2</sub> stream. The substrates were then treated with oxygen plasma for 10 min and converted to negative. ITO-coated glass substrates were converted to positively charged surfaces by immersing them into 1 mM poly(diallyldimethylammonium chloride) (PDDA) polyelectrolyte solution. Then positively charged ITO substrates were immersed into silver nanodisks solution. Because the silver nanodisks were already negatively charged [35], they can self-assemble on the ITO substrate through the electrostatic interaction. A schematic of the steps involved in the fabrication of the electrostatic self-assembled layer of silver nanodisks is shown in Scheme 1. The coverage density of the silver nanodisks on the ITO substrates was controlled by varying the reaction time (15 and 30 min).



**Scheme 1.** Schematic of the steps involved in the fabrication of the electrostatic self-assembled layer of silver nanodisks.

Films of P3HT (Lumin Tech Inc.) were prepared by spin coating from a 6 mg/ml chlorobenzene solution on ITO substrates with and without silver nanodisks. Their thicknesses are ~20 nm. The thickness was measured using a Veeco DEKTAK 150 surface profilometer. UV–vis absorption spectra were recorded using Ocean Optics USB4000 spectrometer. Photoluminescence (PL) spectra were recorded using the front face geometry of P3HT films by a spectrofluorometer (Jasco FP-6500) with excitation at 405 nm. The size and the shape of the silver nanodisks were measured using a field-emission scanning electron microscope (FE-SEM, JEOL, JSM-6700 F) and a transmission electron microscope (TEM, Hitachi H-800) operating at 200 kV. Samples for TEM analysis were prepared by dripping a drop of silver nanodisks colloids onto the carbon-coated copper grids and drying them in air at room temperature.

To fabricate the organic solar cells, a 50 nm hole-transport layer of PEDOT:PSS (Clevious P VP Al 4083, H. C. Starck) was spin-coated onto the ITO glass substrates with and without silver nanodisks from its aqueous solution followed by annealing the film at 140 °C for 10 min in the air. The substrates were then transferred into a nitrogen-filled glove-box. An active layer of P3HT:PC<sub>61</sub>BM (Lumin Tech Inc.) (1:0.6) was spin-coated onto the PEDOT:PSS layer from a chlorobenzene solution with total concentration of 40 mg/ml. The thickness of the active layer is 150 nm. Finally, a cathode of LiF (0.6 nm)/Al (100 nm) was thermally deposited in a vacuum system to finish the device fabrication. Afterwards, the devices were annealed on a hot plate inside the glove box at 150 °C for 10 min. The effective area of the devices is about 5 mm<sup>2</sup>.

Current–voltage ( $J$ – $V$ ) characteristics were recorded using a Keithley 2400 Source Meter in the dark and under AM1.5G 100 mW/cm<sup>2</sup> irradiation (Sciencetech SS-0.5K Solar Simulator). Spectral response was recorded by an SR830 lock-in amplifier under short circuit condition when the devices were irradiated by a monochromatic light from a Xeon lamp through a spectrometer. All characterizations were performed under ambient atmosphere at room temperature.

## 3. Results and discussion

The UV–vis absorption spectra of the silver nanodisks in aqueous solution (concentration  $1 \times 10^{-4}$  M) are shown in Fig. 1. The average diameter of the silver nanodisks was 46.7 nm with a very good uniformly-sized distribution as determined by TEM (Fig. 1). The UV–vis spectrum of the silver nanodisks solution and self-assembled silver nanodisk film shows a peak at 540 nm, indicating the presence of a plasmon absorption band (Fig. 1). The spectral overlap between the absorption spectrum of P3HT (400–650 nm) and the plasmon region of the silver nanodisks (540 nm) could be achieved, and accordingly the efficient plasmonic near field excitation enhancement is expected.

The difference in the particle density of the silver nanodisks films between the cases of the low density (15 min) silver nanodisks film and high density (30 min) silver nanodisks film is shown in Fig. 2. Most of the silver nanodisks are present in

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