

# Influence of gold-silica nanoparticles on the performance of small-molecule bulk heterojunction solar cells



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## ABSTRACT

Light trapping by gold (Au)-silica nanospheres and nanorods embedded in the active layer of small-molecule (SM) organic solar cell has been systematically compared. Nanorod significantly outperforms nanosphere because of more light scattering and higher quality factor for localized surface plasmon resonance (LSPR) triggered by nanorods. The optimum concentration of nanorod was characterized by charge carrier transport and morphology of the active layers. At optimum nanorod concentration, almost no change in the morphology of the active layer reveals that LSPR and scattering effects rather than the morphology are mainly responsible for the enhanced power conversion efficiency. In addition, the preliminary lifetime studies of the SM solar cells with and without Au-silica nanorods were conducted by measuring the current density–voltage characteristics over 20 days. The results show that plasmonic device with nanorods has no adverse impact on the device stability.

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## 1. Introduction

Solution-processed small-molecule (SM) donors have stimulated much interest as a viable alternative to the widely studied conjugated polymers for organic bulk heterojunction (BHJ) solar cells [1–4]. Solution-processed SM donors have the advantage over conjugated polymer counterparts since the SM donor molecules are monodisperse and can be purified using reproducible fabrication protocols and therefore provide more uniform performance in SM-based devices [5,6].

Similar to polymer solar cells (PSCs), SM solar cells have major limitation in insufficient sunlight absorption because the active layer of SM solar cells should be very thin (~100 nm) due to low carrier mobility and short exciton diffusion length [7–9]. Hence, there is a need to capture more light without increasing the thickness of the BHJ layer. As discussed in a recent review [10], the light trapping techniques for thin film organic solar cells include the cells with V-shaped (folded) geometry, incoupler at the front surface, structured back reflector and substrates. In addition, in recent

years, there has been intense interest in applying the plasmonic properties of metallic nanoparticles (NPs) to enhance the performance of PSCs by doping these NPs directly into the active layer [11–14]. Typically, most existing plasmonic materials for improved light absorption in PSCs are focused on the spherical NPs with a diameter less than 50 nm [15–18]. Here, a near field effect attributed to the enhancement of local electric field around the NP is thought to enhance the optical absorption in the BHJ. However, the localized surface plasmon resonance (LSPR) spectrum of these NPs has only one LSPR peak in the shorter wavelength region. In addition, the small size and spherical shape in these NPs greatly weakens the light scattering of NPs inside of PSCs [19]. Recently, the incorporation of large-sized nanorods into the low bandgap PSCs was reported to result in 26% relative improvement in power conversion efficiency (PCE) [20]. This enhancement was due to far field scattering by the incorporated nanorods increasing the optical path length within the cell. Despite this enhancement in PSCs, the influence of the nanorods on PSCs has not been explored in sufficient detail. In a recent paper by Wadam et al. [21], it was shown that the PCE enhancement mechanism in poly(3-hexylthiophene) (P3HT): fullerene BHJ cells incorporated with Au nanorods was not due to plasmonic effects but rather by a favorable modification of the BHJ nano-morphology by the nanorods.

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In this paper, we incorporated core-shell structured Au-silica NPs into SM solar cells based on 7,7'-(4,4-bis(2-ethylhexyl)-4H-silolo[3,2-b:4,5-b']-dithiophene-2,6-diyl)bis(6-fluoro-4-(5'-hexyl-[2,2'-bithiophen]-5-yl)benzo[c][1,2,5]thiadiazole):[6,6]-phenyl-C<sub>71</sub>-butyric acid methyl ester (p-DTS(FBTTh<sub>2</sub>)<sub>2</sub>:PC<sub>70</sub>BM). The two different geometries of NPs, namely nanorod and nanosphere were examined and the geometry effect on SM devices were studied by comparing total light scattering and the quality factor for LSPR. In addition, the X-ray diffraction (XRD) and surface morphology measurements at the optimized Au-silica nanorod concentration were conducted to verify that the SM BHJ structure does not change with the Au-silica nanorods unlike P3HT system [21]. The experimental results confirm that for the p-DTS(FBTTh<sub>2</sub>)<sub>2</sub>:PC<sub>70</sub>BM system, light scattering and LSPR effect rather than morphology of the BHJ layer contribute to the enhancement of PCE in plasmonic SM device. Moreover, the charge carrier transport characterization and atomic force microscopy (AFM) were used to identify the factors that determine the optimum concentration of nanorods in plasmonic devices. Finally, the impact of nanorods on the stability of SM device was also examined.

## 2. Experimental

### 2.1. Synthesis of nanorods and nanospheres

We synthesized Au-silica core-shell nanorods/nanospheres, using the seed-mediated method [20,22]. The procedure of Au-silica nanorods consists of two parts: (i) seed-mediated synthesis of gold nanorods using a seed solution and the reagent hexadecyltrimethylammonium bromide and (ii) solution synthesis of the silica shell using tetraethyl orthosilicate [23]. In order to have a more rigorous comparison with core-shell gold nanorods, bare gold nanorods were also synthesized by the seed-mediated method only.

### 2.2. Device fabrication

SM BHJ solar cells were fabricated by the following process. Indium tin oxide (ITO)-coated glass substrates were cleaned by detergent, deionized water, acetone and isopropyl alcohol sequentially in an ultrasonic bath. Each substrate was blow dried by nitrogen and treated with oxygen plasma for 10 min to reform the ITO surface. Poly(3,4-ethylene-dioxythiophene):poly(styrenesulphonate) (PEDOT:PSS) (Baytron P 4083) was spin-coated at 5000 rpm for 40 s on the ITO-coated substrate to obtain a thickness of 30 nm. The PEDOT:PSS film was then annealed at 150 °C for 20 min on a hotplate. The SM BHJ solutions were prepared by adding 35 mg of p-DTS(FBTTh<sub>2</sub>)<sub>2</sub>:PC<sub>70</sub>BM blend with a weight ratio of 3:2 and Au-silica (or Au) NPs into 1 ml of chlorobenzene (CB) with 0.4 v/v% 1,8 octanedithiol processing additive. The SM BHJ solutions were spin-casted at 2000 rpm for 45 s on the surface of PEDOT:PSS layer and then thermally annealed at 80 °C for 10 min to remove residual solvent in a glove box. All SM BHJ films have the thicknesses of ~100 nm measured by surface profiler. Finally, the cathode consisting of 20 nm Ca and 80 nm Ag was thermally deposited under vacuum condition of  $1 \times 10^{-4}$  Pa. All fabricated devices with an active area of 8 mm<sup>2</sup> were encapsulated using epoxy and cover glass before removal from the glove box.

For the charge transport studies, hole-only devices were fabricated with a structure of the ITO/Molybdenum trioxide (MoO<sub>3</sub>)/p-DTS(FBTTh<sub>2</sub>)<sub>2</sub>:PC<sub>70</sub>BM/MoO<sub>3</sub>/Ag. The second layer of MoO<sub>3</sub> blocks the injection of electrons from the Ag electrode. Electron-only devices have a structure of ITO/Al/p-DTS(FBTTh<sub>2</sub>)<sub>2</sub>:PC<sub>70</sub>BM/Ca/Al. The first layer of Al blocks the injection of holes from the ITO electrode.

### 2.3. Device characterization

The dimensions of the Au-silica NPs were characterized by a JEOL 1400 transmission electron microscope (TEM) at an accelerating voltage of 100 kV. The height and phase images of the p-DTS(FBTTh<sub>2</sub>)<sub>2</sub>:PC<sub>70</sub>BM films with and without nanorods were scanned in the tapping mode using a Cypher S AFM (Asylum Research). XRD measurements were conducted using D8 Advance Eco X-ray diffractometer at 40 keV and 25 mA using Cu K $\alpha$  ( $\lambda = 1.54 \text{ \AA}$ ).

Optical absorption spectra of the BHJ layers were measured using the Perkin Elmer Lambda 950 spectrophotometer which is fitted with a 150 mm diameter integrating sphere. For absorption measurement, the reflectance and specular exclusion ports of the integrating sphere are blocked by white standard plates. By unblocking these two ports consecutively, the ratio of diffuse scattering to total scattering or the haze factor could also be measured. Prior to absorption measurements, the film thickness of all layers was measured by a KLA-Tencor P-10 surface profiler to ensure the thicknesses are the same.

For solar cell characterization, current density-voltage (*J*-*V*) characteristics under dark and illuminated conditions were measured with a Keithley 2400 source meter unit. A solar simulator (Solar Light Inc.) provides simulated sunlight with AM1.5G spectrum and irradiance of 100 mW/cm<sup>2</sup>. The external quantum efficiency (EQE) spectra were conducted by a Bentham PVE 300 system consisting of a calibrated silicon photodiode, a xenon lamp, monochromator, lock-in amplifier and chopper wheel.

## 3. Results and discussion

### 3.1. Comparison of Au-silica nanospheres and nanorods

TEM images in the insets of Fig. 1 display that the average diameter of nanospheres core and the thickness of the silica shell are ~25 nm and ~5 nm respectively. The average diameter and length of the Au nanorods core are ~31 nm and ~86 nm respectively and the thickness of the silica shell is ~5 nm. The histograms of TEM image measurements used to determine these average NP size parameters can be found in the [Supplementary information](#). A 5 nm shell thickness is chosen because as reported recently [24], significant enhanced localized field should exist outside the shell

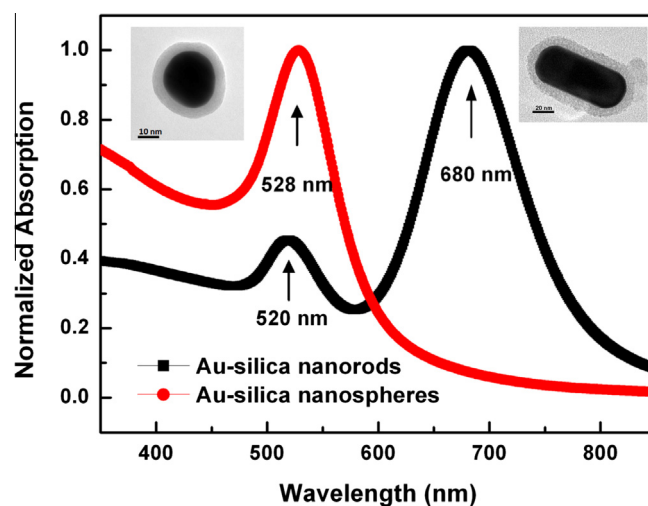


Fig. 1. Normalized absorption spectra of the Au-silica nanorods and nanospheres in CB. The inset is the TEM images of Au-silica nanosphere and nanorod. The scale bars represent 10 nm and 20 nm respectively.

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