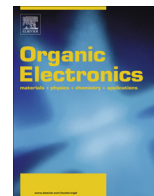




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## Plasmonic nanoparticle incorporation into inverted hybrid organic–inorganic solar cells

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### ABSTRACT

To enhance solar harvesting in hybrid organic–inorganic photovoltaic devices we positioned plasmonic gold nanostructures in titania-conjugated polymer devices. Two plasmonic structures were judiciously designed and studied. In the first, agglomerates of gold nanoparticles, ~150 nm in diameter, were positioned as scattering clusters at the active layer/ITO electrode interface. In the second approach, 5 nm isolated Au particles were suspended inside the hybrid active layer to induce localized plasmonic field enhancement. The location of the particles, size and distribution in the film was confirmed using scanning and transmission electron microscopy and X-ray scattering. The optical absorption measurements confirmed that both plasmonic structures enhance light harvesting, and photovoltaic devices utilizing each of the approaches showed improved photocurrent generation. Finite difference time domain (FDTD) simulations have corroborated that scattering-based and near-field absorption enhancements occur in the active layer, in good agreement with the measured absorption. Finally, combining both approaches to fabricate a hybrid photovoltaic device with both plasmonic nanostructures, i.e. Au NPs both in and on the active layer, resulted in a twofold increase in photocurrent generation.

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## 1. Introduction

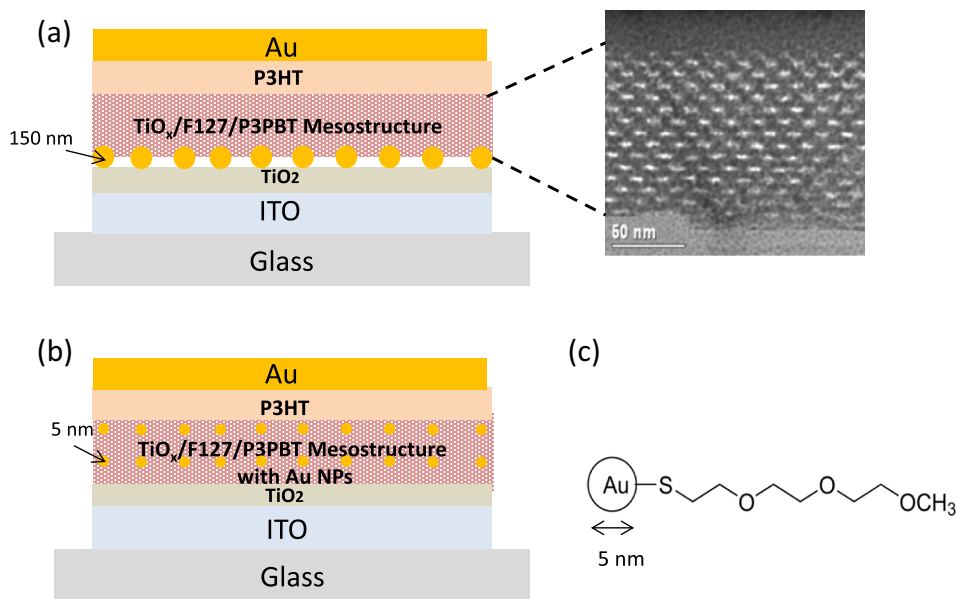
Photovoltaic cells based on hybrid organic–inorganic materials combine cost effective fabrication methods with high chemical and environmental stability, and have therefore been extensively studied in the last few years [1–4]. Like organic solar cells, the design of hybrid solar cells is dictated by the trade-off between light absorption and charge extraction. Accordingly, active layer thickness is typically 150–200 nm, resulting in an absorption-limited device performance. Recently, metallic nanoparticles (NPs) have been utilized to enhance light absorption in organic solar cells composed of a conjugated polymer donor and fullerene derivative acceptor in a bulk heterojunction (BHJ) morphology [5–11]. Absorption enhancement due to the presence of metallic NPs is typically induced by two physical mechanisms. In the first, metallic NPs serve as efficient scatters trapping additional photons in the active layer [12,13]. To apply this mechanism effectively, metallic nano-clusters or periodic nanostructures are placed at the front electrode–active layer interface prior to the deposition of the active layer [6,14–20]. Under such conditions, the NPs exhibit surface plasmons that strongly scatter into the overlaying absorbing films

and enhance the light absorption by the active layer. In the second mechanism NPs are embedded inside the active layer so that the highly localized surface plasmon (LSP) induces a near-field that enhances the absorption in the particles' vicinity [12,13]. To apply the second mechanism in organic solar cells the metallic NPs are usually blended with the active layer solution and suspended in the BHJ active layer film during its deposition [7,8,21,22].

Most plasmonic-enhanced OPVs reported to date were demonstrated using conjugated polymer–fullerene derivative BHJ since their efficiencies are usually higher than hybrid PVs. Nevertheless, hybrid PVs are often limited by their absorption, and therefore the potential of increasing the absorption and device performance by plasmonic nanoparticles is promising. In this report gold nanoparticles are used, both as scattering elements at the active layer/bottom electrode interface, Fig. 1a, and as embedded NPs inside the BHJ for LSP absorption, Fig. 1b, in inverted hybrid titania-conjugated polymer PV devices. We show that placing the Au NPs at the active layer/electrode interface results in an enhanced absorption in the conjugated polymer and photocurrent, while embedding the Au NPs in the BHJ results in an enhanced absorption in the conjugated polymer but only a moderate increase in the photocurrent. Finally, combining the two design strategies resulted in an over twofold increase of the photocurrent.

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**Fig. 1.** The plasmon-enhanced hybrid device structures used in this study: (a) Au NPs are deposited as a scattering layer between the transparent electrode and the hybrid active layer. A TEM image of a typical hybrid active layer, F127-directed  $\text{TiO}_x$ /P3PBT mesostructure (with no NPs), is shown in the inset. (b) Au NPs are incorporated inside the hybrid F127-directed  $\text{TiO}_x$ /P3PBT mesostructure during its formation. (c) Chemical structure of 1-Mercapto-(triethylene glycol) methyl ether functionalized gold nanoparticles used in this study.

## 2. Experimental

Functionalized gold NPs 1-Mercapto-(triethylene glycol) methyl ether with diameter of  $4.5 \pm 1$  nm (Fig. 1c) were purchased from Sigma-Aldrich, and used as received. These NPs show localized surface plasmon resonance absorption centered at 514 nm (see Supplementary Information section Fig. S1), which is very close to the maximum absorption of the active layer used in this study and hence could further enhance the absorption.

The hybrid active layer used in this study was a self-organized BHJ of conjugated polymer donor and  $\text{TiO}_x$  acceptor. The highly ordered BHJ structure was obtained by the co-assembly of the titania precursor and conjugated polymer with a structure directing agent (SDA) surfactant, as previously reported [23,24]. In a typical synthesis, titanium tetra-ethoxide (TEOT, Aldrich) was hydrolyzed by adding 0.18 ml of 12 M HCl to 0.5 ml of liquid TEOT, after which the mixture was vigorously stirred for 60 min or until the solution was clear. Simultaneously, 0.095 g of the SDA Pluronic™ F127 ((ethylene oxide)<sub>106</sub>-(propylene oxide)<sub>70</sub>-(ethylene oxide)<sub>106</sub>,  $M_n = 12,600$ , BASF) was dissolved in 2 ml of methanol. After dissolution, the F127/alcohol solution was added to the hydrolyzed titanium ethoxide, and the mixture was left for further stirring for at least 2 h. P3PBT (Poly[3-(potassium-4-butanoate)thiophene-2,5-diyl]), a water-soluble polythiophene, was used as the conjugated polymer absorber because it could be co-assembled with the  $\text{TiO}_x$  precursor to form a highly ordered hybrid film. P3PBT was dissolved in deionized water at concentration of 7 mg/ml. The solution was heated to 60 °C and stirred for an hour to fully dissolve the polymer and then cooled to room temperature. The final precursor solution was prepared by adding the polymer solution to the  $\text{TiO}_x$ /SDA solution in 1:1 volume ratio.

PV devices with inverted architecture were fabricated on  $12 \times 12$  mm<sup>2</sup> glass plates covered with patterned tin-doped indium oxide (ITO), designed as inverted PV cells. The substrates were thoroughly cleaned by sonication in acetone, methanol, and isopropanol. Then, a  $7 \pm 0.5$  nm dense  $\text{TiO}_2$  electron transporting layer (ETL) was deposited by ALD using tetrakis-dimethyl amino titanium and  $\text{H}_2\text{O}$  precursors. Two approaches for the incorporation

of Au NPs into the hybrid devices were considered in this study. In the first approach, the Au NPs suspension in ethanol (0.8 w/v%) was spin casted onto the  $\text{TiO}_2$ /ITO electrode at 1500 rpm prior to the deposition of the hybrid active layer by dip coating (configuration A, Fig. 1a). In the second approach, the Au NPs suspension (50  $\mu\text{l}$  of 2 w/v% in ethanol) was added to the active layer's precursor solution prior to dip coating. During dip coating the NPs were incorporated into the self-organizing mesostructure (configuration B, Fig. 1b). Dip coating of the active layer, with or without the Au particles, was performed at 5 mm/s in an environmentally controlled chamber with a relative humidity of 50%. Films were also dip coated on glass, silicon and polyimide for structure and chemical analysis. The film thickness, 200 nm, was constant for films with and without gold nanoparticles. To complete the device fabrication the films were then dried in a glove box and a  $\sim 30$  nm layer of neat P3HT hole transporting layer (HTL) was deposited on top from a 20 mg/ml DCB solution by spin-coating at 1500 rpm in a nitrogen atmosphere. All films were then heat treated at 90 °C under vacuum for 3 h for final drying. The top contact, 100 nm-thick gold, was deposited by thermal evaporation of through an eight-pixel shadow mask, defining eight 2.4 mm<sup>2</sup> devices. Device fabrication was completed by post-evaporation annealing for 5 min at 90 °C in a nitrogen atmosphere.

Small-angle X-ray scattering (SAXS) patterns of the active layers with and without gold NPs were acquired using a Rigaku SMAX-300 system. Transmission electron microscopies (TEM) imaging were acquired using a FEI Titan 80–300 keV S/TEM operating at 200 keV. Scanning electron microscopy (SEM) images were acquired using Zeiss Ultra-Plus FEG-SEM. The absorption spectra were measured using a Cary 5000 agilent UV–vis–NIR spectrophotometer equipped with an integrating sphere over a wavelength range of 300–800 nm. The absorption of the active layers, with and without the Au NP scattering layers, were calculated by subtracting the absorption of the bare ITO/ $\text{TiO}_2$  electrode (see Supplementary Information, Fig. S2) from the spectra of the active layers on the electrode [25]. This was done to ensure that the measurements reflect the morphology of the films as incorporated into the devices. Device current–density–voltage ( $J$ – $V$ ) curves were

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