Single particle dual plasmonic effect for efficient organic solar cells

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Abstract

Incorporating localized surface plasmon resonance (LSPR) into organic solar cells (OSCs) is a popular method for improving the power conversion efficiency (PCE) by introducing better light absorption. In this work, we designed a one-pot synthesis of Ag@SiO₂@AuNPs dual plasmons and observed an immense increase in light absorption over a wide range of wavelengths. Ag@SiO₂ plays the main role in enhancing light absorption near the ultraviolet band. The silica shell can also further enhance the LSP resonance effect and prevent recombination on the surface of AgNPs. The AuNPs on the Ag@SiO₂ shell exhibited strong broad visible-light absorption due to LSP resonance and decreased light reflectance. By utilizing Ag@SiO₂@AuNPs, we could enhance the light absorption and photoinduced charge generation, thereby increasing the device PCE to 8.57% and J_{sc} to 17.67 mA cm⁻², which can be attributed to the enhanced optical properties. Meanwhile, devices without LSPR nanoparticles and Ag@SiO₂ LSPR only showed PCEs of 7.36% and 8.18%, respectively.

Keywords Organic solar cells · Surface plasmon resonance · Light absorption · Photocurrent density

Introduction

Over the past few years, organic solar cells (OSCs) have become emerging energy harvesting devices owing to their easy fabrication, thin film active layer related to flexibility, and being semi-transparent, low-cost, room-temperature processability, and viability for large-area mass production by a roll-to-roll processing technique (Kaltenbrunner et al. 2012; Lu et al. 2015; Powell et al. 2009; Lim et al. 2018; Hoppe and Sariciftci 2004; Brabec et al. 2001; Scharber and Sariciftci 2013). A large amount of effort and importance have been devoted to improving the performance of OSCs, such as controlling the nanomorphology of the photoactive layer (Ma et al. 2014; Heeger 2014), designing new device architectures (Adam et al. 2016; He et al. 2015), and synthesizing new donor–acceptor materials (Cheng et al. 2009; Cui et al. 2019; Cao et al. 2017). Although the excellent results from these studies have greatly contributed to the development of OSCs, poor light absorption, relatively low carrier mobility, and short exciton diffusion length in the photoactive layer are still considerable issues to overcome (Heeger

(2014); Siegmund et al. 2017). These issues make the active layer become very thin (~ 100 nm) (Qi and Wang 2013). The limit of active layer thickness diminishes light-harvesting and photoinduced charge generation, which in turn leads to a low photocurrent density and device efficiency (Qi and Wang 2013; Gupta et al. 2010).

Plasmonic nanoparticles (NPs) have been widely used for the enhancement of light-harvesting in OSCs. In the field of LSPR-based solar cells, numerous efforts in areas such as controlling the particle size, shape, and geometrical arrangement have been conducted to enhance the capability of the LSPR (Ho et al. 2016; Shen et al. 2014; Choi et al. 2013; Yu et al. 2017; Kozanoglu et al. 2013; Liu et al. 2013; Jung et al. 2014). However, all these efforts have been limited to the use of metal nanoparticles (Ag or Au), which absorb a narrow wavelength. To achieve light absorption enhancement over a broad wavelength in the visible-light or infrared region, some cooperative plasmonic nanostructures from multiple metals have been reported (Lu et al. 2013; Kakavelakis et al. 2014; Gao et al. 2017). However, these results showed narrow light absorption enhancement at only different specific wavelengths and did not cover light absorption beyond 700 nm (infrared region) (Lu et al. 2013; Kakavelakis et al. 2014). Therefore, in this study, we developed a one-pot synthesis of Ag@SiO₂@AuNP dual plasmonic nanostructures. By introducing AuNPs on silica shells, we found that there

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was an absorption enhancement over the scale of wavelength from visible light to the infrared region, and simultaneously, there was an increased photocurrent density in the organic solar cells. The effects of AuNPs on silica shells in the ZnO buffer layers in the OSCs were investigated by fabricating poly[[4,8-bis[(2-ethylhexyl)oxy] benzo[1,2-b:4,5-b'] dithiophene-2, 6-diyl] [3-fluoro-2-[(2-ethylhexy)carbonyl] thieno[3,4-b]thiophenediyl]] (PTB7): (Brabec et al. 2001; Brabec et al. 2001)-phenyl C71-butyric acid methyl ester (PC₇₁BM)-based devices. The photovoltaic performance of Ag@SiO₂@AuNP-3R-based NPs showed a significant improvement in the PCE up to 8.57% (J_{sc} 17.67 mA mA. cm^{-2}), whereas the reference device without metal nanoparticles showed a poor PCE of 7.36% (J_{sc} 15.62 mA mA. cm^{-2}).

Experimental details

Preparation of Ag core NPs

A 10-mL aqueous solution of 0.1 M silver nitrate (AgNO₃, Sigma-Aldrich, USA) was stirred with a magnetic bar at elevated temperatures and incorporated into a solution containing 100 mL of absolute ethanol (Sigma-Aldrich, USA) and 50 mL of deionized (DI) water. Within 5 min, 20 mL of a 1 mM polyvinylpyrrolidone (Sigma-Aldrich, USA) aqueous solution was added to the resulting mixture, which was stirred for 20 min. Next, 5.0 mL of 0.1 M sodium hydroxide (Junsei, Japan) was added to the solution, which was then cooled to room temperature (24.0 ± 1.0 °C). Ag NPs were obtained after 2 h of reaction.

Preparation of the Ag@SiO₂ NPs

The Ag@SiO₂ NP solution was synthesized as follows. A mixture containing 50 mL of absolute ethanol (Sigma-Aldrich, USA), 5.0 mL of ammonia (Junsei, Japan), and 1.0 mL of TEOS (Tetraethyl orthosilicate) (Sigma-Aldrich, USA) was slowly added to the silver colloids. The resulting solution was stirred at ambient temperature (24.0 ± 1.0 °C) for 12 h. The as-prepared product was washed with ethanol three times and then dispersed in ethanol for further use.

Preparation of the Ag@SiO₂@Au NPs

The photovoltaic devices were fabricated with the structure glass/ITO/ZnO/PTB7:PC₇₁BM/MoO_x/Ag. The ITO glass substrates were cleaned in deionized water, acetone, and isopropyl alcohol under ultrasonication for 1000 s each and subsequently dried in an oven. A thin film of the ZnO sol–gel was spin-coated at 2000 rpm for 40 s onto pre-cleaned ITO and then annealed at 180 °C for 10 min. Then, the substrates were transferred into an Ar-filled glove box. The Ag, Ag@

SiO₂, and Ag@SiO₂@AuNPs were spin-coated onto the active layer at 4000 rpm for 40 s. For the active layer, the PTB7:PC₇₁BM blend solution was prepared by dissolving 8 mg of PTB7 and 12 mg of PC₇₁BM in 1 mL CB/DIO (0.97:0.03 v/v) and stirred for 6 h at 60 °C. The active solution was spin-coated at 1000 rpm for 40 s and dried at room temperature prior to metal deposition. A thin layer of PEDOT: PSS was spin-coated onto the active layer. Finally, 100 nm of Ag was deposited under high vacuum conditions ($< 10^{-6}$ Torr). The active area of the devices was 0.38 cm², defined by a metal mask.

The photocurrent–density (J – sV) curves of the photovoltaic devices were measured using a Keithley 2400A source unit under the calibrated AM 1.5G simulated illumination of 100 mW cm^{−2} in ambient air conditions. The light intensity was calibrated using a standard Si photodiode detector with a KG-5 filter. The external quantum efficiency (EQE) spectra were measured under monochromatic light with a wavelength of 300–800 nm using the EQE measurement system. A calibrated silicon photodiode was used as a reference. The optical transmittance spectra were obtained using a UV–Vis spectrometer. The surface morphology of the cathode buffer layer films was observed by atomic force microscopy (AFM).

Results and discussion

Figure 1a schematically shows the synthesis process of the core–shell–seed nanoassemblies. The AgNPs (colloid silver) were prepared by mixing silver nitrate in a mixture of ethanol and deionized water under stirring at ~80 °C. Polyvinylpyrrolidone (PVP) and sodium hydroxide were added to the mixture solution under vigorous stirring and then cooled to room temperature. The Ag@SiO₂ was prepared by mixing ammonia, ethanol, and TEOS with colloidal silver. The AgNP core with a diameter of approximately 10 nm was well covered with a thickness of approximately 6 nm, as shown in Fig. 1b, c (high-resolution transmission electron microscopy (HR-TEM) images). The clear boundary between the Ag core and silica shell is caused by the large difference in electron density. The transparent silica shell on the AgNPs can prevent oxidation on Ag and suppress quenching close to Ag due to charge tunneling (Yu et al. 2017; Yip et al. 2016). The Au seeds (Fig. 1d) were deposited on the amino-functionalized silica surface and were grown into AuNPs (Fig. 1e) via an electroless Au-plating method. The diameter of the AuNPs on the silica shell grew after reduction. The TEM image reveals that the mean distance between the Au NPs on the silica shell was approximately 2 nm, which is much smaller than the wavelength of the incident light. To obtain the optimum LSPR effect, NPs well dispersed in ethanol were deposited on ZnO by spin-coating.

Fig. 1 **a** Schematic diagram for the fabrication procedure of Ag@SiO₂@AuNPs. TEM images of the plasmonic core-shell nanoparticles prepared from each synthesis step; **b** as-prepared Ag NPs, **c** Ag@SiO₂ NPs, **d** Ag@SiO₂@Au seeds, and **e** Ag@SiO₂@AuNPs after Au reduction

Figure 2a shows the UV–Vis absorption spectrum of the AgNPs, Ag@SiO₂ NPs, Ag@SiO₂@Au seeds, and the three different Ag@SiO₂@AuNPs (Ag@SiO₂@Au NP-1R, -2R, and -3R) solutions prepared by changing Au-plating conditions. An LSPR absorption centered at 400 nm was obtained for AgNPs, while a broader and a slight red-shift of the absorption to 420 nm for Ag@SiO₂ NPs was likely caused by the silica shell (Yu et al. 2017; Zhang et al. 2016). A red-shifted LSPR peak at 430 nm and an increase in absorption along visible light were observed, which confirms the presence of Au seeds on the surface of the silica shell. The red-shifted and strong peaks which occurred at longer wavelengths corresponded to Ag@SiO₂@AuNPs. The peaks at longer wavelengths correspond to AuNPs, which have a higher permittivity than AgNPs (Gao et al. 2017). It is expected that the strong peaks are affected by the strong interactions between the AuNPs on the silica surface. These peaks increase as the plating rate increases, corresponding to the larger size of the AuNPs. As revealed in the TEM images, the average sizes of the Au NPs were approximately 4.5, 6.7, and 10.0 nm for Ag@SiO₂@Au NP-1R, -2R, and -3R, respectively. At this size, the Au NPs act as sub-wavelength antennas for LSPR excitation. Thus, the LSPR effect dominates the absorption enhancement rather than the scattering effect (Feng et al. 2018; Morfa et al. 2008). The increasing near-electric field intensity in the active layer enhances light absorption. Moreover, the LSPR for the AuNPs smaller than ~ 10 nm will be altered by quantum size effects. For instance, the AuNPs on silica shells with sizes

increasing from ~ 4.5 to ~ 10.0 nm showed red-shifted peaks and an increase in the light absorption. It was observed that the Ag@SiO₂@AuNP-3R solution showed a higher absorption than other devices in the range of 500–800 nm, showing a noteworthy LSPR peak at 539 nm. This peak theoretically corresponds to the light absorption of Au due to LSPR (Kakavelakis et al. 2014). The light absorption near-infrared region can harvest more photons converted into photocurrent (Dai et al. 2007; Xia et al. 2010; Vasseur et al. 2013; Li et al. 2015). Meanwhile, for the Au seeds, the LSPR is strongly quenched owing to the surface damping effect. These results agree with the previous findings that the absorption efficiency decreases as the particle size decreases (Scholl et al. 2012). Thus, the light absorption enhancement of this Au seed was hardly observed.

To further investigate the plasmonic characteristics of the AgNPs, Ag@SiO₂ NPs, and Ag@SiO₂@AuNPs, we simulated the field distribution using the finite-difference time-domain (FDTD) method. It can be used to easily determine the electromagnetic field distribution around nanoparticles, nanorods, and other nanostructures by adjusting the size, number, and material as well as the environmental properties (Dhawan et al. 2009; Yang et al. 2010). Figure 2b shows a field snapshot image for Ag@SiO₂ with a 413 nm monochromatic wave as the input source (amplitude: 1 Vm⁻¹) and a plot for the near-electric field intensity (Vm⁻¹) vs. time (ns) obtained from the marked position (i). A field snapshot image of Ag@SiO₂@AuNP-3R with a 539 nm monochromatic wave as input is shown in Fig. 2c. The plot is for the

Fig. 2 **a** Absorption spectra of Ag NPs, Ag@SiO₂ NPs, Ag@SiO₂@Au seeds and three different Ag@SiO₂@AuNPs (Ag@SiO₂@AuNP-1R, -2R, and -3R) prepared by changing Au-plating conditions. FDTD simulation results for **b** Ag@SiO₂ and **c** Ag@SiO₂@AuNP-3R

near-electric field intensity vs. time at marked position (ii). Localized SPs were generated for wavelengths of 413 nm for Ag/SiO₂ and 539 nm for Ag@SiO₂@AuNP-3R. By incorporating AuNPs on the silica shell, the number of near-electric field intensity sites increases. Moreover, the near-electric field intensity with respect to time at the marked position was also investigated. The spectra show that, upon incorporation of the AuNPs on the silica shell, the field intensity is four times higher than that without the AuNPs.

We fabricated PTB7:PC₇₁BM-based OSCs to investigate the superior light collection capability of the Ag@SiO₂@AuNPs on device performance. The photovoltaic devices were fabricated with the structure glass/ITO/ZnO/PTB7:PC₇₁BM/PEDOT:PSS/Ag, as shown in Fig. 3a. The Ag@SiO₂@AuNPs are positioned in the ZnO nanoripple layer, as shown in the AFM image in Fig. 3b. We also fabricated a device with Ag NPs as a reference. The current

density–voltage (J–V) characteristics are shown in Fig. 3c. The Ag@SiO₂@AuNP-3R device, the best device, achieved a PCE of 8.57% (J_{sc} of 15.97 mA cm⁻², V_{oc} 0.77 V, FF of 0.62), whereas the OSC device with the Ag@SiO₂ NPs' device showed a PCE of 8.18% (J_{sc} of 16.25 mA cm⁻², V_{oc} of 0.73 V, FF of 0.69). The Ag reference device exhibited a PCE of only 7.36%, J_{sc} of 15.97 mA cm⁻², FF of 0.62, and V_{oc} of 0.74 V. The photovoltaic performance obtained from the J–V measurement is summarized in Table 1. We used external quantum efficiency (EQE) to analyze the optical and electrical responses upon the incorporation of NPs in OSCs. Figure 3d shows EQE spectra as a function of the wavelength of OSCs with and without NPs (reference device) in the ZnO layer. To ease the EQE analysis, we provide EQE enhancement as a function of the wavelength, as shown in Fig. 3e. The photocurrent generation over a wide range of ~400–700 nm generally increased after the incorporation of Au NPs on the silica shell. A significant EQE enhancement was observed in the range of 470–580 nm, which was closely aligned with the absorption spectra. Moreover, the IPCE exceeds 70% in the range 650 nm < λ < 680 nm, confirming a noticeable EQE enhancement in the range of 600–740 nm. These results strongly agree with the reflectance and absorption measurement results. The three broad and noteworthy EQE enhancements of the Ag@SiO₂@Au NPs are in good agreement with the calculated absorption enhancement ($\Delta\alpha$) from reflectance measurements. The absorption enhancement spectra are shown in Fig. 3f. This proves that the EQE enhancement is mostly improved by better light absorption. Moreover, generated hot electrons from the gold conduction band and the excited plasmons may induce more effective electron–hole separation.

A device with a glass/ITO/ZnO/PTB7:PC₇₁BM/PEDOT:PSS/Ag was used to investigate the absorption in the active layer. As shown in Fig. 4a, the reflectance of the Ag@SiO₂@AuNP-3R device decreases over a wide range of ~400 to ~700 nm, corresponding to enhanced absorption in the active layer. To clarify the LSPR effect of the NPs, the ZnO layer was characterized by Raman spectroscopy. As shown in the Raman spectra in Fig. 4b, no shift was observed after the incorporation of the AuNPs. However, the Raman intensity increased drastically. This noticeable increase is attributed to surface-enhanced Raman scattering (SERS). This enhancement proves the near-field enhancement of the NPs.

Conclusion

In conclusion, compared with the untreated and Ag@SiO₂ devices, incorporating the Au NPs on the Ag@SiO₂ shell effectively enhanced the light absorption and reduced the

Fig. 3 **a** Schematic representation of the Ag@SiO₂@AuNPs structured device. **b** AFM images of ZnO buffer layers including Ag@SiO₂@AuNPs. **c** *J*–*V* characteristics, **d** EQE spectra, **d** EQE enhancement (%) of inverted solar cells without or with the core–shell nanoparticles; Ag@SiO₂ and Ag@SiO₂@Au nanoparticles after Au reduction (Ag@SiO₂@AuNP -1R and 3R). **e** Calculated absorption enhancement (Δα)

Table 1 Photovoltaic performance of inverted OPVs with or without of the plasmonic core–shell metal nanoparticles incorporated into the ZnO layers

Type of NPs	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF	PCE (%)
Reference (without NPs)	0.74	15.97	0.62	7.36
Ag@SiO ₂	0.73	16.25	0.69	8.18
Ag@SiO ₂ @AuNP-1R	0.76	17.23	0.64	8.34
Ag@SiO ₂ @AuNP-2R	0.77	17.58	0.62	8.39
Ag@SiO ₂ @AuNP-3R	0.77	17.67	0.63	8.57

oparticles; Ag@SiO₂ and Ag@SiO₂@Au nanoparticles after Au reduction (Ag@SiO₂@AuNP -1R and 3R). **e** Calculated absorption enhancement (Δα)

reflectance in the active layer, resulting in superior photoinduced charge generation. Therefore, a high *J*_{sc} and PCE were achieved without sacrificing FF. The PCE increased from 7.36% for the device without the NPs to 8.57% for the device with Ag@SiO₂@AuNP-3R, and a significant enhancement of over 16% was obtained. This finding suggests that enhancing light absorption in the active layer with the incorporation of AuNPs on silica shells is an effective way to improve device performance.

Fig. 4 **a** Reflectance spectra and **b** Raman spectra collected from the OPVs with and without core-shell structured plasmonic nanoparticles in ZnO layer

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Compliance with ethical standards

Conflict of interest The authors declare no competing financial interests.

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