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Tunable plasmon-enhanced broadband light harvesting for perovskite solar cells

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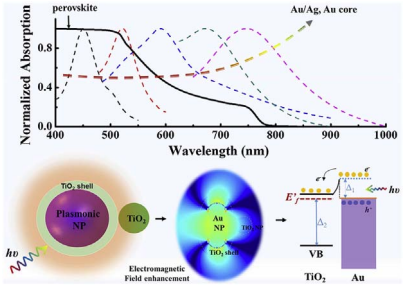
HIGHLIGHTS

- We introduce plasmonic NPs with λ_{LSPR} in visible range into PSCs for enhancing PCE.
- A series of approaches are implemented for obtaining plamonic NPs with different λ_{LSPR}.
- The PCEs of PSCs based on plasmonic NP are significantly improved from 15.04% to 17.85%.
- FDTD simulations are performed to verify the experimental results.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, we report a reliable method for synthesizing (Au, Au/Ag core)/(TiO_2 shell) nanostructures with their plasmonic wavelengths covering the visible light region for perovskite solar cells. The mono- and bi-metallic core-shell nanoparticles exhibit tunable localized surface plasmon resonance wavelength and function as "light tentacle" to improve the photo-electricity conversion efficiency. Plasmonic nanoparticles with different sizes and shapes, different thicknesses of TiO_2 shell and Ag interlayer are found to have a strong influence on the localized surface plasmon resonance enhancement effect. The experimental photovoltaic performance of perovskite solar cells is significantly enhanced when the plasmonic nanoparticles are embedded inmesoporous TiO_2 scaffolds. A champion photo-electricity conversion efficiency of 17.85% is achieved with nanoparticles (Au/Ag, λ_{LSPR} = 650 nm), giving a 18.7% enhancement over that of the pristine device (15.04%). Finite-difference time-domain simulations show that nanorod Au in mesoporus TiO_2 scaffold induces the most intense electromagnetic coupling, and provides a novel emitter for photon flux in mesoporous perovskite solar cells. These theoretical results are consistent with the corresponding experimental those. Thus, enhancing the incident light intensities around 650 nm will be most favorable to the improvement of the photo-electricity conversion efficiency of perovskite solar cells.

1. Introduction

With a low exciton binding energy, long charge diffusion lengths and high carrier mobility, organic-inorganic metallic halide perovskite materials [1–3], typically with $CH_3NH_3PbX_3$ (X = Cl, I or Br), are well-suited as absorbers in solar cells. Considerable research efforts have

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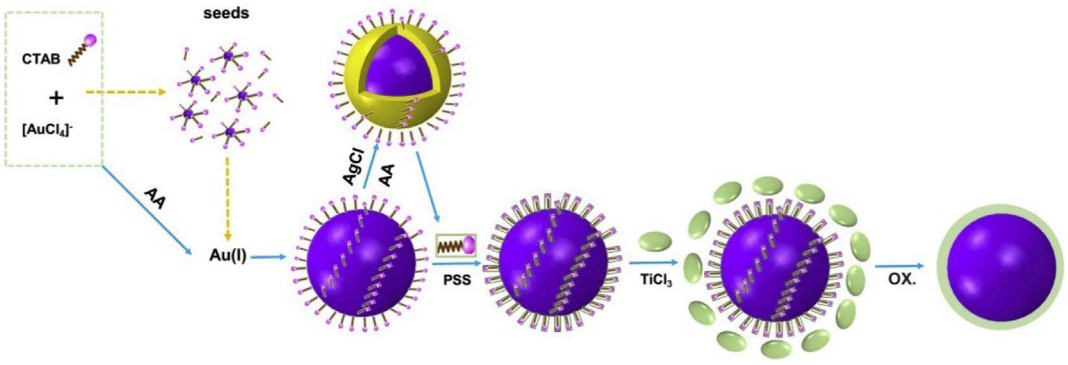

Fig. 1. Schematic diagram for the synthesis of (Au, Au/Ag cores)/(TiO_2 shell) NPs. Purple presents the Au core and Orange presents the Ag shell. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

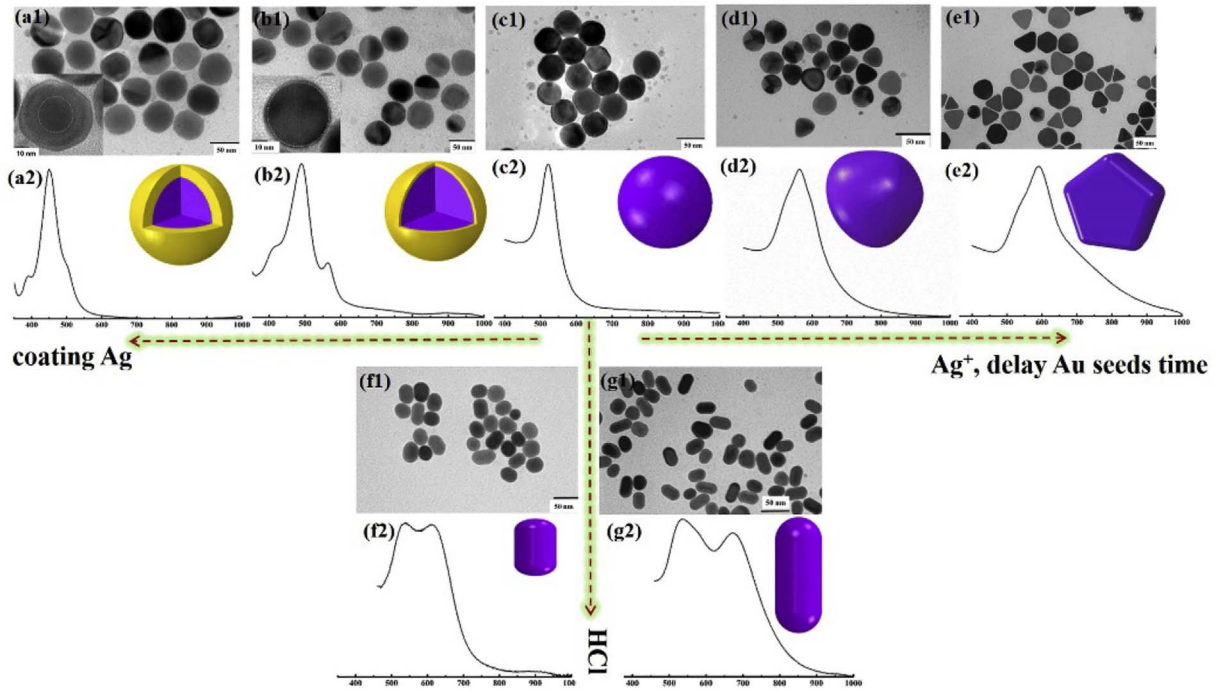

Fig. 2. TEM images, absorption spectra and structures of Au and Au(core)/Ag(shell) NPs.

been put into boosting the efficiency and stability of perovskite solar cells (PSCs) and the device efficiencies have exceeded 22.7% through material design and optimization of processing techniques [4–8]. The perovskite film thickness of about 300–500 nm plays a crucial role in device performance [9]. Maximizing the film thickness can lead to an increase in the absorption of the incident light, but at the same time the charge carriers may not be able to travel through the thick film. Only when charge carriers' migration distance is less than the thickness of the active layer, the charge carriers can be collected by the electrodes [10,11]. Therefore, further improvement is more likely to be obtained through enhancing the electronic properties of perovskite absorber and the interfaces of the n- and p-type charge transporting layers. The optical response of a perovskite film drops significantly beyond $\lambda = 550$ nm [12,13], particularly, longer wavelengths between 600 and 800 nm were not efficiently utilized ever.

When plasmonic nanostructures are illuminated at their resonance frequencies, the incident electromagnetic (EM) fields would be significantly enhanced at their surfaces, boosting the nonlinear absorption properties of PSC at a lower excitation energy [14]. These nonlinear optical responses of the dielectric material are generally several orders of magnitude higher than that at low field [15,16]. A plasmonic nanostructure can be considered as a secondary luminous source to increase photon flux at its resonant wavelength resulting in more light being absorbed by the solar cells [17–21]. Noble metal nanostructures can reinforce optical absorption at particular spectral bands [12–24]. Gold nanoparticles (Au NPs) display local surface plasmon resonance (LSPR), which can be tuned to resonate at specific spectral band [25]. Because $\varepsilon(\mathrm{Ag}) \approx 13\,000\ (\mathrm{Mcm})^{-1}$ and $\varepsilon(\mathrm{Au}) \approx 3300\ (\mathrm{Mcm})^{-1}$ ($(\mathrm{Mcm})^{-1}$ presents molar absorptivity), their intrinsic extinction spectral peaks occur at 390 nm and 520 nm, respectively. The LSPR wavelengths of Au(core)/Ag(shell) NPs are tunable by adjusting their composition [26]. Fermi levels of Au (−5.1 eV) and Ag (−4.7 eV) are close to the lowest energy of the TiO_2 conduction band of −4.3 eV [23]. Consequently, when plasmonic NPs are incorporated intoTiO_2 mesoporous matrixes, the electrons from the Au or Ag cores can be transferred to the TiO_2, thus enhancing absorption can be tunable in the visible light region [27–31]. Many previously work devoted to research plasmonic metal NPs incorporating into mesoporous solar cells for improving light harvesting, *i. e.* dye-sensitized solar cells (DSSCs) and in PSCs. Kamat et al. employed SiO_2- and TiO_2- capped Au NPs to enhance the short-circuit current (J_{sc}) and open circuit voltage (V_{oc}) in DSSCs [32]. Tang et al. studied the effects of Au@TiO_2 hollow