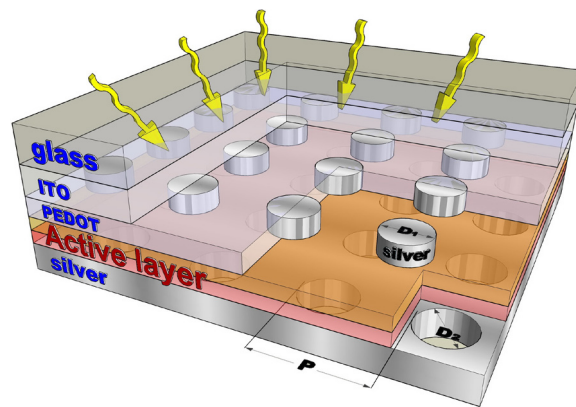


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Research Highlights on Organic Photovoltaics and Plasmonics

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Abstract: Incorporation of plasmonic nanostructures for light trapping is an attractive solution to enhance the optical absorption of the active light-harvesting layer(s) in thin-film photovoltaic cells. The latest research highlights on plasmonic-enhanced organic photovoltaics (OPVs), including metallic nanoparticles and periodic nanopatterned structures, are presented in this paper.

Index Terms: Organic photovoltaics, plasmonics, nanophotonics.

1. Introduction

As a promising clean and sustainable source of energy, photovoltaic (PV) technologies are expected to play a major role in meeting the global renewable energy challenge [1]. Considerable progress has been made recently in organic photovoltaics (OPVs). Compared with their inorganic counterparts, OPVs based on solution processable materials can be fabricated at low cost, over large areas, on rigid or flexible substrates. However, the low charge-carrier mobility and small exciton diffusion length of most organics limit the thickness of the active light-harvesting layers (30–150 nm) [2], [3] in OPVs and lead to relatively poor absorption, which results in insufficient carrier generation and low power conversion efficiency (PCE). To date the best reported PCE for solid-state OPVs is 6.7% [4] and ~8% [5] for molecular and polymer-based single and bulk heterojunctions, respectively. It is believed that the PCE has to reach at least 10% to make OPVs attractive to the marketplace. Effective light trapping schemes for thin-film PVs offer an attractive approach and are critical to ensure full absorption of the incident sunlight. Novel plasmonic nanostructures using surface plasmon polaritons (SPPs) are exciting alternatives that offer effective light trapping inside the active layer of a thin-film solar cell [6]. SPPs are collective oscillations of electromagnetic (EM) waves and free electrons at the interface of a metal and a dielectric or semiconductor material. They are strongly confined at the metal/dielectric interface, with their intensity decaying exponentially with the distance from the interface. The decay lengths of SPP modes in the visible are usually from tens to hundreds of nanometers, depending on the refractive index of the dielectric material at the interface. Incorporation of plasmonic nanostructures for light trapping using 1) random metallic nanoparticles and 2) periodic metallic nanopatterns offers a very attractive solution to enhance the optical absorption and current density in

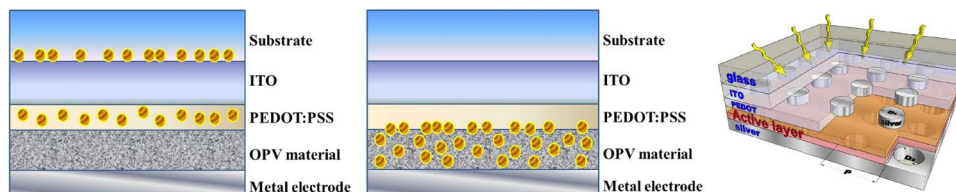


Fig. 1. Conceptual illustration of plasmonic-assisted OPV devices with random distributed metallic nanoparticles (a) outside and (b) embedded inside the active layers. (c) Conceptual illustration of plasmonic OPVs with periodic metallic nanoparticles on top and a nanopatterned metal film on the bottom interfaces of the active light-harvesting layer.

OPVs without increasing the thickness of the active layer(s). In this paper, we give a brief overview of research highlights that describe the latest progress on plasmonic-enhanced OPV devices.

2. Metallic Nanoparticles for Plasmonic-Enhanced OPVs

Metallic nanoparticles are probably the most popular nanostructures used to enhance the performance of PV devices, due primarily to the relatively ease of fabrication. General design considerations have been reviewed extensively for inorganic PVs [6]. Metallic nanoparticles using different materials, concentrations, shapes, sizes to list a few have been introduced in different layers and interfaces at various positions in OPV devices. Fig. 1(a) shows metallic nanoparticles embedded in layers outside the active light-harvesting layer(s). Low cost and simple techniques such as vapor-phase deposition [7], pulse-current electrodeposition [8], and thermal annealing [9] have been used to deposit metallic nanoparticles on indium tin oxide (ITO) electrodes or poly(3,4-ethylenedioxythiophene): poly(styrene sulfonate) (PEDOT:PSS) buffer layers. The dependence of the localized surface plasmon resonance (LSPR) on the size and composition of the nanoparticles have been studied, with reported enhancement in the PCE [8]–[12]. For instance, the PCE of OPVs using poly(3-hexylthiophene)/[6]-phenyl C₆₁ butyric acid methyl ester (P3HT:PCBM) as the active light-harvesting layer were increased by ~18.8% (PCE from 3.48% to 4.19% [10]) to ~69% (PCE from 1.3% to 2.2% [7]) by blending Ag or Au nanoparticles (10 ~ 15 nm in diameter [7], [8], [10] and 45 nm ± 5 nm in diameter [11]) within the PEDOT:PSS buffer layers, respectively. Au nanoparticles (15 nm) have also been introduced in the PEDOT:PSS layer of an OPV device using poly(2-methoxy-5(20-ethylhexyloxy)-1,4-phenylenevinylene (MEH-PPV) as the active layer and led to an enhancement in PCE from 1.99% to 2.36% [12]. More recently, Au nanoparticles (~72 nm) were deposited in the interconnecting layer of an inverted tandem polymer solar cell consisting of a cell based on a P3HT: indene-C₆₀ bis-adduct (IC₆₀BA) and another based on poly[(4,4'-bis(2-ethylhexyl)dithieno[3,2-b:2',3'-d]silole)-2,6-diyl-alt-(2,1,3-benzothiadiazole)-4,7-diyl] (PSBTBT): [6,6]-phenyl-C₇₁-butyric acid methyl ester (PC₇₀BM) cell, and resulted in a 20% increase in PCE from 5.22% to 6.24% [13]. However, all these enhanced PCEs are still lower than the experimental records recently achieved [4], [5]. In most these reports, the nanoparticles are located relatively far from the active organic layers, and the absorption enhancement arises from light concentration/scattering from nanoparticles rather than any near-field enhanced LSPR modes.

Embedding metallic nanoparticles into the active layers of OPVs as shown in Fig. 1(b) will make full use of the strong field of the LSPR confined within the surfaces and interfaces between consecutive layers. Metallic nanoparticles are easier to disperse into the active organic layers of an OPV compared to an inorganic PV due to the ease of processability of the organic materials. By tuning the geometric parameters of the metallic nanoparticles, the interaction between the longitudinal and transverse modes supported in the nanoparticle array/chain/cluster embedded in the OPV active layer can lead to impressive absorption enhancement [14], [15]. However, it is recognized that nanoparticles embedded inside the OPV active layer may be experimentally challenging because of the fine tuning that is needed to minimize loss mechanisms such as nonradiative decay and charge-carrier recombination. For instance, it was reported that Ag nanoparticles tend to phase segregate from P3HT:PCBM polymer blends at high concentrations and when incorporated in OPVs lead to a

decrease of carrier extraction [16]. Although an enhancement of PCE from 1% to 2.4% was observed by placing Au nanoparticles at the interface between P3HT:PCBM and PEDOT:PSS [17], no study was reported on PCE enhancement when metallic nanoparticles are dispersed inside the active layer. Most studies focused on optical absorption enhancement of OPV materials containing nanoparticles of various metals (e.g., Au, Ag, Cu, Al, etc.) [18]–[20]. Cu nanoparticles (20 nm) embedded inside P3HT layers were shown to enhance the dissociation of excitons without increasing the P3HT optical absorption [21]. In summary, the roles of metallic nanoparticles dispersed inside the active layer of an OPV need to be further investigated and better understood. It remains to be seen whether the plasmon-enhanced absorption of the active light-harvesting layer will lead to an enhancement in the current density and PCE of an OPV device.

3. Periodic Nanopatterned Structures for Plasmonic-Enhanced OPVs

Although the synthesis of plasmonic metal nanoparticles seems relatively simple, it is quite challenging to control their size and shapes by thermal evaporation and electrodeposition techniques. Another approach that may lead to better tunability, employs periodic patterned metallic nanostructures to enhance the optical absorption of the organic active layers in OPVs. For instance, 1-D Ag grating nanostructures were used to design and fabricate OPVs using several different active organic layers such as P3HT:PCBM [22], PCBM [23], and copper phthalocyanine (CuPc): 3,4,9,10-perylenetetracarboxylic bis-benzimidazole (PTCBI) [24]. By tuning the geometric parameters of the 1-D metallic nanogratings for top transparent anodes [24] or back reflector cathodes [22], the resonant wavelength of the SPP modes could be tuned easily in the visible to near-IR spectral range. However, due to the polarization dependence of the 1-D grating nanostructures, the light trapping is very different for transverse-electric and transverse-magnetic polarized modes, and the simultaneous optimization of the optical absorption for both polarization modes is quite challenging. In addition, due to the intrinsic dispersive metal properties, the resonant wavelengths of SPP modes supported by optically thick metal films depend significantly on the incident light angle. Their bandwidths are also relatively narrow, which is not desirable for OPV applications. Consequently, it is critical to achieve broadband, polarization insensitive/independent, and angle-insensitive plasmonic nanostructures [see Fig. 1(c)] [25].

A straightforward solution to overcome the polarization dependence of 1-D nanostructures in the design of OPVs is to use 2-D nanopatterned structures with high-order symmetries. For instance, 2-D metallic nanovoids were proposed to enhance the coupling between localized SPP modes and J-aggregate excitons of organic semiconducting films [26]. More recently, these 2-D nanovoid plasmonic structures have been applied to an OPV device using P3HT:PCBM as the active layer and resulted in an enhancement factor of 3.5 and 4 in external quantum efficiency and in PCE (i.e., from 0.052% to 0.2%), respectively [27]. In another example, periodic and quasiperiodic 2-D nanoaperture arrays with high-order symmetries were employed as the back reflector or concentrator to enhance the optical absorption of P3HT:PCBM layers [28]. However, the enhanced absorption was demonstrated in a very narrow spectral region close to the excitation wavelength. Consequently, it is still important to achieve broadband absorption enhancement over the entire visible and part of the near-infrared solar spectrum to further improve the overall PV device efficiencies.

Although randomly distributed nanoparticles of varying size and shape received much attention [6], their resonant frequencies for periodic plasmonic nanostructures usually occur within a narrow spectral band. Physical mechanisms that lead to broadband absorption enhancement and/or a robust predictive capability for designing plasmonic nanostructures have recently been pursued [29]–[31]. Due to the dispersive nature of SPP modes, plasmonic nanostructures are usually sensitive to incident angle, which led to designs of plasmonic black bodies, perfect absorbers or omnidirectional absorbers based on 1-D grating nanostructures [32], 2-D complex metallic nanopatterned structures [33], and randomly distributed metamaterials [34]. These support angle-insensitive hybrid modes in the visible and near-IR spectral region. Incorporating these nanostructures into OPV devices offers a promising approach to address the angular tolerance limitation for conventional plasmonic nanostructures. Metallic triangular/pyramid nanogratings [35] and

nanostrips [36] were designed as the back reflector in order to realize angle insensitive absorption enhancement in the active light-harvesting layer (e.g., P3HT:PCBM [35] and the donor: acceptor CuPc:C₆₀ molecular heterojunction [36]).

In addition to their role in enhancing absorption in the active light-harvesting layer, the nanopatterned metal structures can also function as a device electrode. Nanopatterned metal films [29] and metallic nanowires [31] with a low fill factor have been employed as alternative electrodes to replace ITO films in organic optoelectronic devices. For example, one study introduced a 15-nm thick Au/AuO_x film to replace the ITO layer for high-performance flexible organic optoelectronics [37]. In another investigation, 40-nm thick 1-D metallic nanowires were used as anodes for organic solar cells [38], [39]. The sheet resistance of arrays of 40-nm thick nanopatterned metal electrodes (Au, Cu, Ag) varies depending on the metal and linewidth but does not exceed 20–30 Ω · sq⁻¹ [38]. These alternative nanomaterials for transparent electrodes are highly desirable in many optoelectronic applications due to the potential shortage in indium and other problems associated with the use of ITO, which are outside the scope of this paper. It is of interest to design metallic nanostructures for OPVs that serve concurrently for light trapping and as transparent electrodes [40]–[42].

In summary, we have reviewed the latest progress of plasmonic enhanced OPVs reported mainly in 2011. In general, most studies focused on the absorption enhancement of OPV materials, which need to be translated into improvement in current density and device efficiency. We hope that this brief overview of the research status can excite more creative research that will result in OPV devices with improved performance.

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