Recent Progress in Design of Plasmonic Thin-Film Solar Cells with Enhanced Efficiency

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Abstract: Plasmonics is concerned with the interaction between electromagnetic radiation and conduction electrons at metallic/dielectric interfaces and has been a hot research area since 2000. Plasmonics is now attracting more research activities because a plasmonic nanostructure is one of the most promising candidates to serve as the light scattering and trapping agent to reduce the physical thickness of current solar cells while maintaining the optical thickness. In this patents review, we describe the basic mechanisms of plasmonic thin-film solar cells, summarize the common design and fabrication techniques, and discuss the future prospects.

Keywords: Efficiency, fabrication, light trapping, plasmonic nanostructures, thin-film solar cells.

INTRODUCTION

Solar cells can provide virtually unlimited amounts of energy by effectively converting sunlight into clean electrical power. The cost of current solar cells still needs to be significantly reduced and the efficiency must increase substantially to enable wider implementation. Thin-film solar cells (TFSCs) may provide a viable pathway towards this goal because of the low materials and processing costs [1-7]. A TFSC is made by depositing one or more thin layers (thin film) of photovoltaic materials such as copper indium diselenide [8], Gallium arsenide [9], organic polymer [10], titanium dioxide [11], zinc oxide [12], as well as amorphous [13] and polycrystalline silicon [14] on a substrate coated glass, metal, or plastic. The thickness of the layer can vary from a few nanometers to tens of micrometers. With the exception of planar cells, TFSCs can be introduced to non-planar cells because of their physical flexibility [15]. Therefore, TFSCs have large potential in terrestrial and space photovoltaics and offer a variety of choices in terms of the device design and fabrication. In this review, we describe the basic mechanisms of plasmonic thin-film solar cells, summarize the common design and fabrication techniques, and discuss the future prospect.

PRINCIPLE

The performance of TFSCs is improving steadily due to increasingly better understanding of the unique and wide range of structural, chemical, and optoelectronic characteristics of thin-film materials. Ideally, the light absorbing materials of a TFSC should be a direct bandgap semiconductor with a bandgap of ~1.5eV and efficient solar power absorption. However, there are no suitable elemental semiconductor materials available with a direct bandgap close to 1.5eV and a major limitation in TFSC technologies is that the absorbance of near-band gap light is ineffective [4]. Silicon being an indirect bandgap material with a gap ~1.1eV is by no means an ideal material. For effective solar absorption, Si wafers have to be at least 50mm thick unless optical enhancement techniques are used to improve light absorption. Structuring the TFSCs so that light is trapped inside and absorption is enhanced is an alternative approach. It is possible to achieve light trapping by forming wavelength-scale textures on the substrate prior to deposition of the TFSC, and large increases in the photocurrent have been achieved in this way [16-18]. However, a rough semiconductor surface leads to more surface recombination, and semiconductors deposited on rough surfaces typically have low materials quality. The unique optical properties of metallic structures have been recently used to boost the efficiency of solar cells. These metallic nanostructures exhibit easily accessible collective electron oscillations known as surface plasmons [19-21].

Plasmonics, first described more than a century ago, deals with the interactions between electromagnetic (EM) radiation and conduction electrons at the metallic/dielectric interfaces and has been extensively researched since 2000. Mainly designated as surface plasmons and localized surface plasmons, plasmonics spurs many intriguing applications such as ultrafast chips, surface-enhanced spectroscopy, and photovoltaics. Surface plasmons are surface-bound EM waves coupled to electron density oscillations and confined to the metallic interface well below the diffraction limit λ_0 / n (λ_0 is wavelength in vacuum and n is the refractive index of the surrounding environment), thereby overcoming the size incompatibility between optical devices and current micro-fabrication techniques. A plasmonic nanostructure is

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one of the most promising light scattering and trapping agents in the effort to reduce the physical thickness of current solar cells while maintaining the optical thickness. Three configurations of incorporating plasmonic structures to improve energy conversion are illustrated in Fig. (**1**) [6]. Metallic nanostructures can be used as the subwavelength scattering elements to couple and trap freely propagating plane waves from the sun penetrating the absorbing semiconductor thin film by folding the light into a thin absorber layer (see Fig. (**1a**)). In the second approach, metallic nanostructures can be used as the nanoantennae to couple the plasmonic near-field to the semiconductor thereby increasing the effective absorption cross section (see Fig. (**1b**)). Thirdly, a corrugated metallic film fabricated on the backside of a photovoltaic absorber layer can couple sunlight into the surface plasmon modes at the metal/semiconductor interface as well as guided modes in the semiconductor slab, whereupon light is converted to photocarriers in the semiconductor (see Fig. (**1c**)). Therefore, light absorption can be improved in ultra-thin layers of plasmonic nanostructures to lead to smaller recombination currents, larger open circuit voltages, and higher conversion efficiency [22].

DESIGN

The design of metallic nanostructures for plasmonic TFSCs has received renewed interest lately due to the availability of new nanofabrication tools and better understanding of their optical properties. In general, a planar plasmonic device includes a first layer having a surface configured to receive at least one photon of incident light [23] and an adjacent patterned plasmonic nanostructured layer is optically coupled to the first layer. The patterned plasmonic nanostructured layer includes (a) at least a portion of the surface of the patterned plasmonic nanostructured layer involving a textured surface and (b) at least one compound nanofeature including the first material adjacent to the second one within the nanofeature.

In different cell designs, both near-field light concentration close to the individual particle resonance and effective light trapping by nanometallics have been explored. Catchpole *et al.* developed fundamental design principles to help increase the efficiency of solar cells using light trapping by scattering from metal nanoparticles [24]. Cylindrical and hemispherical particles can increase the path length more than spherical particles because of enhanced near-field coupling. In the meantime, the path length enhancement achieved from an electric point dipole is even higher than the Lambertian value. Silver particles provide much larger path length enhancements than gold particles. The scattering cross section of the particles is very sensitive to the thickness of the spacer layer at the substrate, which provides additional tunability to the design of particle arrays. Davis *et al.* have developed a theory concerning the coupling of evanescent optical fields between metallic nanoparticles to provide the basis for designing plasmonic systems [25]. It should be noted here that the plasmon hybridization theory is an important analytical model to calculate the localized plasmon modes in the complex metallic nanostructures [26-29]. It is helpful to design metallic nanostructures with specific optical properties, especially when multiple metallic or dielectric surfaces are present [30].

There is a clear need for effective optimization strategies that lead to broadband absorption enhancements over the entire solar spectrum. This type of optimization for plasmonic TFSCs is now possible. Pala *et al.* have developed the basic design rules for the use of metallic nanostructures to realize broadband absorption enhancement in TFSCs. It is applied to a relevant and physically intuitive model system consisting of a two-dimensional, periodic array of Ag strips on a silica-coated Si film supported by a silica substrate, as shown in Fig. (**2**) [31]. Another group has also presented criteria on optimizing the light trapping efficiency of periodic arrays of metal nanoparticles in Si solar cell applications [32]. The scattering cross section of the nanoparticles and diffraction efficiency of the gratings should be maximized in the long wavelength range. The grating pitch should be chosen to allow higher order diffraction modes for long wavelengths while maintaining the highest possible fill factor. These conditions place stringent constraints on the optimal parameters (particle size of ~200nm and pitch of ~400nm) for periodic arrays of metal nanoparticles, in

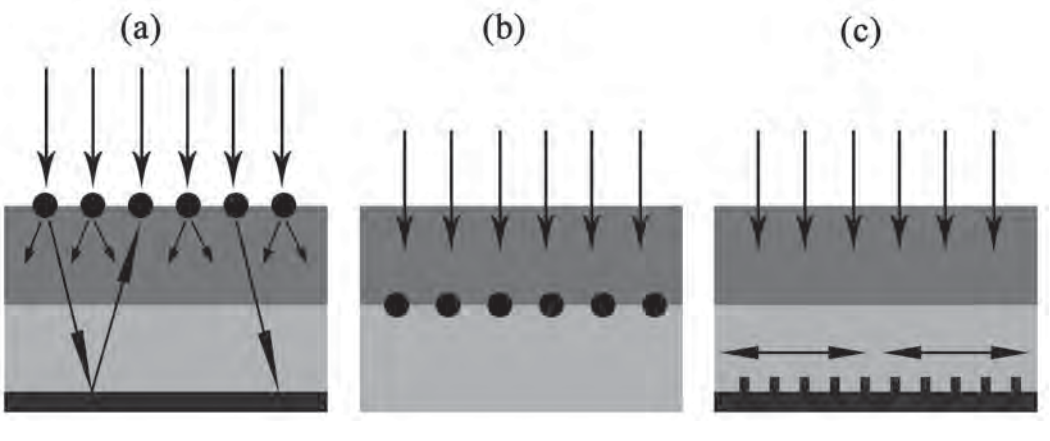

Fig. (1). Plasmonic light-trapping geometries in TFSCs: (**a**) Light trapping by scattering from metal nanoparticles at the surface of the solar cell. (**b**) Light trapping by the excitation of localized surface plasmons in metal nanoparticles embedded in the semiconductor. (**c**) Light trapping by excitation of surface plasmon polaritons at the metal/semiconductor interface.

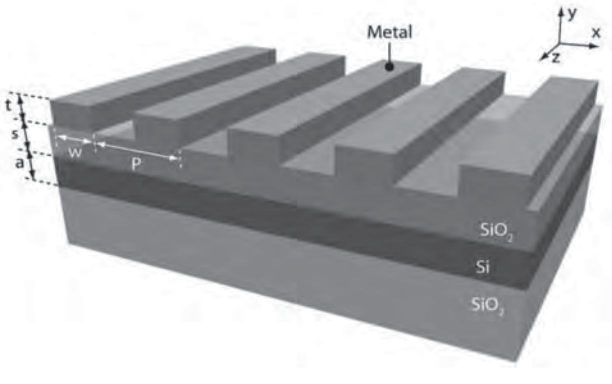

Fig. (2). A schematic of the proposed plasmon-enhanced cell structure [31].

contrast to dielectric gratings, where a relatively wide range of periods and feature sizes can be used for efficient light trapping.

Ji *et al.* [33] have recently developed a planar plasmonic device as the back reflector in light-collecting optics for solar cells. This device is composed of a silicon wafer with the surface to receive photons and an adjacent patterned plasmonic nanostructured layer is optically coupled to the layer. The nanostructured layer comprises a patterned metal film with a textured surface and some of the nanofeatures are coated on the materials to support the plasmon waves. The device layers can be used in conjunction with a set of solar cell layered absorbers in order to efficiently absorb photons and increase efficiency. The solar cell layers are more efficient energy converters over different wavelength ranges. The device can be used in conjunction with an absorbing layer to guide the light into the layer, reflect light back to the absorbing layer effectively after the light passes through the solar cell without absorption, and trap light with the absorbing layer.

FABRICATION

Recent experimental studies on both organic [34-37] and inorganic [38-41] plasmonic TFSCs have shown that the optical properties can be significantly improved by metallic nanoparticles deposited on top of the active layer. By incorporating plasmonic nanoparticles on surface modified transparent electrodes, the overall power conversion efficiency can be boosted. It mainly results from the improved photocurrent density as a result of enhanced absorption due to the high EM field strength in the vicinity of the excited surface plasmons [42-44]. Pioneering work in the area of plasmonic enhancement of light-sensitive devices was done by Stuart and Hall, who showed that the resonance associated with metal island films could be used to enhance the sensitivity of very thin semiconductor photodetectors [45]. More recently, Margaret introduced a process to synthesize silver nanoplates [46]. Brennan demonstrated that the shape of the silver nanoplates could be one of the following: circular shaped plate, elliptical shaped plate, triangular shaped plate, hexagonal shaped plate, and other flat polygonal shaped plate [47] and different shapes led to different properties. Dusan has introduced an approach to fabricate an array of metal particles or metal compound particles dispersed on a substrate [48]. The process includes providing a solid substrate having a metal of metal compound film on the surface and heating the solid substrate for a temperature and time sufficient to convert the film into an array of particles of metal or metal compound on the surface. Martin has introduced a plasmonic TFSC structure comprising a photoelectric conversion structure, a grating coupler for coupling incident light into the photoelectric conversion structure, and a resonator structure to provide light trapping in the photoelectric conversion structure [49].

From the point of view of processing and manufacturing, elemental materials are the simplest. Therefore, Si TFSCs are rather general in the photovoltaic field and have been used in many ways. Silver nanoparticles have been fabricated on Si TFSCs by thermal annealing [50], vapor phase deposition [35], electrochemical corrosion [51] and so on. A typical thermal annealing method to fabricate plasmonic Si TFSCs is described in the following [52]. Metal nanoparticles are deposited by thermal evaporation of thin layers of silver followed by annealing. Owing to surface tension, the particles coalesce together to form islands and the surface plasmons can increase the spectral response of TFSCs over almost the entire solar spectrum. At wavelengths close to the band gap of Si, significant absorption enhancement has been observed from both thin-film and wafer-based structures.

A recent addition to the list of fabrication methods is interference lithography which has been investigated for photovoltaic applications with promising results [53]. Interference lithography is commonly used to generate patterns on substrates and profile-controlled sinusoidal plasmonic crystals. It also offers the most economical and efficient way to produce 1D and 2D structures over a large area in a short and simple cycle [54]. The typical fabrication steps start with cleaning the Si substrate, depositing a photoresist film, writing of the grating profile with an interference pattern, and finally evaporating silver and gold.

In 2010, Blackwood *et al.* reported the influence of silver and gold nanoparticles deposited by electroless plating on the performance of silicon photodiodes [55]. The advantage of electroless plating is the ease of implementation as it involves a simple displacement reaction, i.e., the noble Ag or Au plates at the expense of the corrosion of the more base silicon [56]. The overall surface plasmon resonance from the metallic nanoparticles can improve the efficiency of silicon based TFSCs. Similar work has been reported concerning photocells consisting of several spherical particles with the inner part having one conduction type while the outer part having the opposite conduction type [57]. The advancement associated with this device is the reduced thickness of the photosensitive layer due to larger absorption of the entire EM radiation spectrum and concentration of EM radiation in the region of the photosensitive layer via the surface plasmon resonance effect. Nanowire based TFSC has also been reported [58]. The nanowire has excellent electroconductivity and optical characteristics and can be manufactured easily. Moreover, the nanowire consists of core containing crystalline or non-crystalline silicon-rich oxide which enables light emission in the visible region.

An interesting alternative to Si TFSCs are dye sensitized solar cells (DSSCs), which combine interesting properties such as low cost, simple fabrication process, and excellent mechanical and processing properties of organic materials [59]. Despite intensive research, the highest reported efficiency of traditional DSSCs is only slightly higher than that achieved in the mid-1990s [60]. While improvements in the photocurrent and fill factor can be envisioned, the greatest possibilities for efficiency gains are likely to stem from increase in the photovoltage. One way is to increase the dye absorption cross sections [61] and another approach is to couple dyes to plasmonic silver or gold nanoparticles [62-64]. The enhanced EM field near the particle is capable of increasing the effective molecular absorptivity greatly [65]. It should be noted that I^-/I_3^- is extremely corrosive toward silver and gold nanoparticles. Thus, plasmonic particles must be protected when designing plasmonic DSSCs.

The sol-gel process is the most common preparation method for DSSCs. Typical precursors are metal alkoxides and metal salts (such as chlorides, nitrates and acetates), which undergo various forms of hydrolysis and polycondensation reactions. Protective and decorative coatings, and electro-optic components can be applied to glass, metal and other types of substrates with these methods. Cast into a mold, and with further drying and heat-treatment, dense plasmonics metallic particles can be formed that cannot be created by any other method. For instance, DSSCs based on ZnO nanorods are fabricated and modified by the addition of Au nanoparticles [66]. ZnO nanorods can be synthesized from a precursor of zinc acetate dihydrate in an alcoholic solution [67-69]. Electrons generated by photo-absorption through thick aggregated Au nanoparticles layer may have a lower injection rate to ZnO nanorods compared to those absorbed by the dye. TiO_2 is another typical metallic oxide material used in DSSCs. In addition, DSSCs can be fabricated using graphene-TiO_2 composite photoanodes [70, 71]. It has been reported that the incorporated graphene exhibits both increased dye adsorption and significantly longer electron lifetime.

Exploiting the properties of surface plasmons is an effective approach to improve the absorption efficiency of organic TFSCs. The evanescent nature of surface plasmons permits the manipulation and enhancement of optical fields below the diffraction limit, allowing the use of organic TFSC layers without sacrificing absorption potential. The integration of plasmonic structures with organic TFSCs has been introduced above and they have been used to increase optical absorption. Although promising, the field enhancement from metallic nanoparticles is relatively short range and their presence can quench excitons. Another approach utilizes surface plasmon mediated energy transfer across a Ag film to improve optical absorption in organic TFSCs. Lindquist *et al.* have demonstrated enhanced power conversion efficiency in organic photovoltaic cells incorporated into a plasmonic nanocavity array [72, 73]. The work improves the absorption efficiency through the use of a plasmonic nanocavity array that combines local field enhancement near a patterned metal anode with strong plasmonic nanocavity modes throughout the structure. The response of the nanocavity can be tuned and provide significant control over the internal optical field distribution. This offers a new approach for the integration of plasmonic structures in organic TFSCs and is broadly applicable to the enhancement of absorption and cell power conversion efficiency in devices. In 2010, another group used photoinduced absorption spectroscopy to measure long-lived photogenerated charge carriers in optically thin donor/acceptor conjugated polymer blend films near plasmon-resonant silver nanoprisms [74]. Figure **3** depicts the schematics of the polymer blend films with and without silver nanoprisms. The plasmon-resonant silver nanoprisms can significantly enhance the generation of long-lived charge carriers in optically thin, bulk heterojunction blend polymer films despite possible competing induced losses due to the presence of the metal surfaces. The results suggest that solution-processable metal nanoparticles such as plasmon-resonant silver nanoprisms can serve as viable optical antennae or scattering centers with tunable plasmon resonance in order to improve light harvesting in organic TFSCs.

EFFICIENCY ENHANCEMENT

There have been a number of reports describing the efficiency enhancement of TFSCs with plasmonic nanostructures [75-85]. Experimentally, high peak enhancement in the tens of percent range at specific wavelengths [52] and overall efficiency enhancements of 8.3% and 8% have been achieved with the use of plasmonic structures employing a-Si [75] and GaAs [9], respectively. However, no detailed comparison has yet been made regarding cells employing alternative light trapping technologies. The power conversion efficiency of the polymer photovoltaic devices incorporating the Au nanoparticles shown in Fig. (**4**) improved to 4.24% from a value of 3.57% for the device fabricated without Au nanoparticles [82]. Almost at the same time, the enhancement ratio as high as 20% was also reported in plasmonic polymer tandemsolar cell by simply incorporating Au nanoparticles [83]. Recent studies have led to effective optimization strategies for broadband absorption enhancement over the entire solar spectrum [31, 86-88]. For instance, Sefunc *et al.* propose and demonstrate a design concept of volumetric plasmonic resonators that relies on the idea of incorporating coupled layers of plasmonic structures embedded into a solar cell in enhanced optical absorption for surface-normal and off-axis angle configurations, beyond the enhancement limit of individual plasmonic layers [89]. Brongersma's group proposed a general design strategy for the realization and optimization of broadband absorption enhancement in TFSCs using 2D, periodic arrays of metallic nanostructures [31]. An array of Ag strips on a thin (50nm) Si film cell revealed photocurrent enhancement of up to 43%. Even more substantial enhancement (>50%) can be accomplished from thicker cells (100nm-1μm) in which more waveguide modes can contribute to the overall enhancement. The general design strategy can be readily extended to 3D arrays and aperiodic structures thereby opening up new hope for even higher efficiency.

CURRENT & FUTURE DEVELOPMENTS

The emphasis on research and development of plasmonic TFSCs is to better understand the science and devices, enhance the efficiency, and develop economically viable manufacturing processes. The plasmonic efficiency enhancement

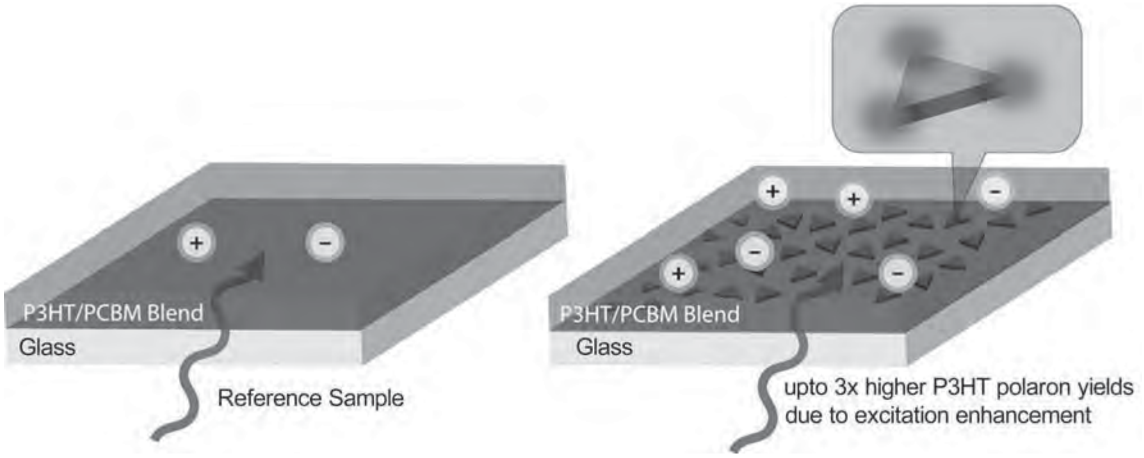

Fig. (3). Schematics of polymer blend films with and without silver nanoprisms [74].

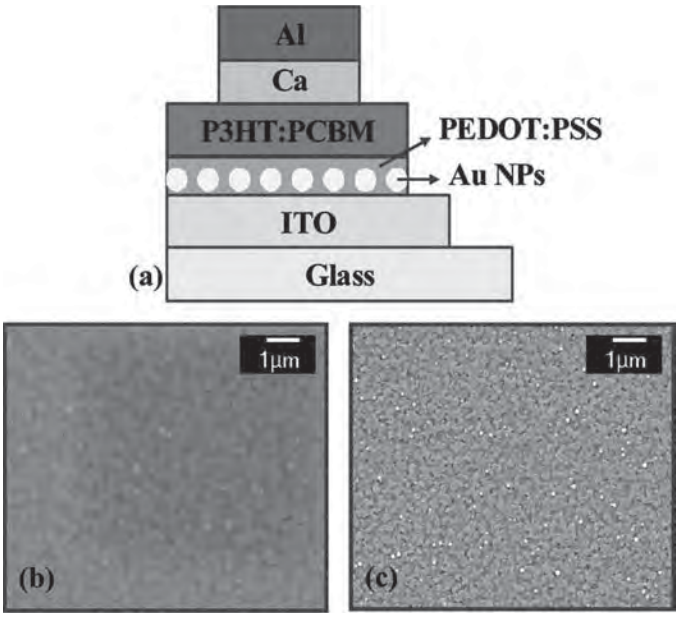

Fig. (4). (**a**) Device architecture of the polymer photovoltaic devices incorporating Au nanoparticles in the poly(3,4-ethylenedioxythiophene): poly(styrene sulfonate) (PEDOT:PSS) layer. (**b**,**c**) SEM images of anodic buffer layers prepared with (**b**) Pristine PEDOT: PSS and (**c**) PEDOT: PSS featuring embedded Au nanoparticles. The Au nanoparticles appear as white dots in (**c**) [82].

depends mainly on the geometrical configuration of the metal nanoparticles. That is, the enhancement is induced by the aggregation state which can generate the corresponding plasmon resonance. Therefore, there is a definite need for nano-technology that can better determine the optimal particle distribution in solar cell applications, take into account interactions between the particles, and include the interactions with the absorbing substrate in a realistic way. With regard to fabrication challenges, efforts are expected to extend existing methods proven useful for arraying ordered metallic arrays such as nanoimprint and chemical synthesis to device fabrication.

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CONFLICT OF INTEREST

The author declares no conflict of interest.

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