Light Trapping on Plasmonic-Photonic Nanostructured Fluorine-Doped Tin Oxide

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ABSTRACT: Plasmonic Au nanoparticles of ~50–200 nm in diameter were generated via thermally assisted self-assembly from Au films evaporated on fluorine-doped tin oxide (FTO). A comparative study has been made on the light trapping effects of the plasmonic Au nanoparticles on original FTO and FTO with photonic nanopatterns fabricated using nanoimprint lithography. While strong localized surface plasmon resonance (LSPR) in the visible spectrum has been confirmed in both



cases, quantitative differences exist and may be attributed to the Au nanoparticle morphology and their interface with FTO. In particular, the LSPR frequency depends on the Au nanoparticle structure and size, while the LSPR peak width is affected by FTO surface morphology (original or nanopatterned). It has been found that the combined plasmonic-photonic nanostructured FTO has the best light trapping, which agrees well with the finite difference time domain simulations and provides a promising transparent electrode for high-efficiency thin film solar cells and other optoelectronic devices.

INTRODUCTION

Light management has been an important approach in the development of high-performance and low-cost thin-film photovoltaics for clean and sustainable solar energy that plays a major role in meeting world energy need.¹ One approach is to incorporate photonic nanostructures for improved light scattering and thus light absorption in photovoltaics. Such structures have been successfully applied to photovoltaics in photoactive layers^{2–4} and back reflectors.^{5–7} Our recent work in generating photonic nanostructures directly on the transparent electrodes, such as fluorine-doped tin oxide (FTO), has led to enhanced light scattering and improved photocurrent in thin-film dye-sensitized solar cells.⁸ The unique advantage of this approach is in minimal interruptions to the photovoltaic devices grown atop.

Plasmonic light trapping is regarded as an attractive approach to improve light management in solar cells^{9,10} and has prompted extensive studies recently in design consideration^{11,12} as well as performance characterization of plasmonic photo-voltaics^{13,14} that generally involve incorporation of metal nanoparticles. Plasmon is light-induced collective oscillations of electrons on the metal surfaces, which becomes localized on metal nanoparticles due to the dimension restriction. When the light frequency is resonant with collective oscillation of electrons, both strong absorption and strong scattering of incident light may occur, which is known as localized surface plasmon resonance (LSPR). The frequency of the LSPR is determined primarily by the physical parameters of metal nanoparticle (complex dielectric constant, dimensions, and shapes), as well as the refractive index of the surrounding material. LSPR is expected to enhance light-harvesting in photovoltaics through scattering and local (typically in the range of tens to hundreds of nanometers) enhancement in electromagnetic fields.

This work explores the combination of plasmonic and photonic nanostructures on FTO transparent electrodes, and the goal is to understand the benefits of this combination that could be effectively integrated for photovoltaic applications. With conducting both experiments and computer simulation and modeling, we evaluate merits and shortcomings of the combined plasmonic-photonic nanostructures as front electrode and back reflectors for improved light management in solar cells.^{15,16} A unique advantage of implementing plasmonicphotonic nanostructures on electrodes instead of inside the semiconductors is to minimize generating additional defects as charge traps that reduce charge mobility and increase charge recombination. Specifically, plasmonic Au particles were generated on photonic nanostructures as well as original FTO electrodes using a thermally self-assembly process. Measurements of light scattering and plasmonic effects on these samples in comparison with that on the photonic nanostructured FTO allow comparison of the combined effects of plasmonic-photonic nanostructures on FTO with separated effects of plasmonic and photonic counterparts.

EXPERIMENTAL SECTION

Commercial FTO glass TEC 8 (Hartford Glass) with thickness of approximately 550-600 nm FTO layer coated on ~3 mm thick glass was utilized in this experiment. Before processing, FTO glass was cleaned in an ultrasonic bath with detergent, DI water, acetone, and isopropanol in sequence. Au films were deposited using e-beam evaporation. The Au thickness of 6 nm Au was found to be optimal for obtaining Au nanoparticles of

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lateral dimension around 100 nm in the low-temperature range of 300–550 °C. The results presented in this work employed post-deposition annealing that was carried out in an Ar atmosphere at two different conditions of 300 °C for 10 h and 550 °C for 30 min, respectively. The same conditions for Au nanoparticle generation were applied to original and nanopatterned FTO in this work.

Nanopatterned FTO was fabricated using nanoimprint lithography (NIL) followed by reactive ion etching. The details of this process were previously reported.⁸ In this work, two types of patterned FTO electrodes (nanopillar and nanoline) were fabricated with corresponding stamps (Lightsmith). The nanopillar stamp has a 700 nm period in the hexagonal pillar arrangement of 290 nm pillar diameter. The nanoline stamp also has a 700 nm period with 350 nm feature width. After NIL and reactive ion etching (RIE), a thin Au film of 5-15 nm was evaporated onto the patterned FTO, followed by thermal annealing to form plasmonic-photonic nanostructures on FTO. The entire fabrication procedure is depicted schematically in Figure 1.



Figure 1. Schematic description of the fabrication procedure for plasmonic Au nanoparticles on patterned FTO electrodes.

RESULTS AND DISCUSSION

Scanning electron microscopy (JEOL JSM-6380) was applied to analyze surface morphology of the samples. Figure 2a,b compares the Au nanoparticle morphology formed on flat FTO under two different annealing conditions. The Au nanoparticles have irregular shapes with lateral dimension of 50–80 nm for the lower temperature annealing case and 50-100 nm in the higher annealing temperature case. This indicates that the mobility of gold during annealing also plays an important role in controlling the Au particle geometry and size is the initial Au film thickness. Above the Au/FTO eutectic point, the lower Au mobility at a lower annealing temperature can be partially compensated by longer annealing time. In particular, the annealing of 300 °C for 10 h and 550 °C for 30 min in Ar gas yielded Au nanoparticles on flat FTO with comparable geometry with slightly smaller lateral dimension.

Considering that larger Au nanoparticles with lateral dimension around 100 nm are more favorable in terms of minimizing Ohmic loss,⁹ the higher annealing temperature of 550 °C was employed for generating Au nanoparticles on nanopatterned photonic FTO. Another advantage of using higher annealing temperature is the shorter annealing time required. Figure 3 showed the SEM images of Au nanoparticles formed on patterned FTO nanolines and nanopillars, respectively. A dense layer of Au nanoparticles can be clearly seen in both cases. Interestingly, the average lateral dimension of the Au nanoparticles is considerably larger probably due to the reduced mobility of Au on the patterned FTO surfaces. Most of the Au particles are rounded with a diameter of approximately 50-200 nm. The ridged structure of nanoline FTO (Figure 3a) holds some Au particles on the wall, while most are at the bottom of the ridges. In contrast, the Au nanoparticles distribute uniformly on both the nanopillar top and bottom in nanopillar FTO (Figure 3b).

Figure 4 illustrates the simulated effect of electromagnetic field distributions when passing through different FTO/Si interfaces in three different cases: flat FTO/Si interface (Figure 4b), patterned FTO structure without Au particles (Figure 4c), and patterned FTO structure (Figure 4d) with Au nanoparticles, as shown in Figure 4a. In the simulation, a Gaussian beam at 580 nm wavelength was launched upward from FTO to Si across the interface. Here Si is chosen as an example for the case of silicon thin film photovoltaic devices on FTO transparent electrodes. For a flat FTO/Si interface, the power distribution of the Gaussian beam is largely undisturbed when it passes across the interface, as shown in Figure 4b. When the trapezoid structure, which is similar to nanopillars and nanolines, is patterned on the FTO interface with the size shown in Figure 4a, the Gaussian beam is scattered by the photonic nanostructure, resulting in scattered power distribution near and above the FTO/Si interface, as shown in Figure 4c. This light scattering is beneficial to increase the path length of the optical beam in the silicon layer and hence enhances light absorption at a given thickness of Si thin films. When Au



Figure 2. SEM images of surface morphology of Au nanoparticles formed on flat FTO after annealing the Au film of 6 nm in thickness on FTO at (a) 300 $^{\circ}$ C for 10 h and (b) 550 $^{\circ}$ C for 30 min, respectively, in Ar gas.



Figure 3. SEM images for Au nanoparticles on (a) FTO with nanoline pattern and (b) FTO with nanopillar pattern. The Au nanoparticles were formed by annealing the Au film of 12 nm in thickness coated on nanopatterned FTO at 550 C for 30 min.



Figure 4. (a) Surface structure of Si/FTO interface illustrating trapezoid FTO surface pattern and locations of Au particles. (b) Power distribution of a Gaussian beam passing across a flat Si/FTO interface. (c) Power distribution when the interface has patterned FTO structure but without Au particles. (d) Power distribution when the interface has both patterned FTO structure and Au particles as shown in panel a.



Figure 5. (a) Measured LSPR of Au-coated flat FTO including as-deposited Au film (black); Au films followed by annealing at 550 °C for 30 min (red); and Au films followed by annealing at 300 °C for 10 h (green). (b) Measured LSPR of Au nanoparticles on patterned FTO including on nanolines (blue) and nanopillars (purple). The Au nanoparticles were formed from annealing of coated Au film at 550 °C for 30 min.

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nanoparticles with 100 nm diameter and 50 nm thickness are added to the trapezoid FTO nanostructures, the effect of light scattering is further enhanced, as shown in Figure 4d. Although these patterns help elucidate the path length enhancement in the silicon layer due to the surface nanostructure, a more quantitative analysis of wavelength-dependent resonance absorption due to surface plasmonic effect is necessary, which will be discussed later.

The optical transmittance measurement was carried out using a Horiba iHR550 imaging spectrometer in transmission mode with an integrating sphere. Figure 5a compared the spectra taken on three Au (6 nm thick)-coated flat FTO samples. Two of them were made under the same conditions as in Figure 2, and the third sample did not go through thermal annealing. LSPR can be clearly seen in the first two samples (red and blue), and the LSPR wavelength is at ~580 nm, comparable for these two samples. This is in contrast with the much longer LSPR wavelength of 750 nm for the Au-filmcoated FTO without annealing (black). The similar LSPR wavelengths for the two plasmonic Au nanoparticles on flat FTO samples are expected because they depend primarily on Au nanoparticle sizes that are comparable in the two cases. It is interesting to note that the sample annealed at 550 °C exhibited a stronger LSPR resonance than that annealed at 300 °C, which may be ascribed to the Au nanoparticle shape difference. At the higher annealing temperature, the Au nanoparticles are more rounded and collapse onto the substrate, which results in better contact with the substrate with a hemisphere shape and slightly increased particle size. Consequently, an enhanced LSPR amplitude was observed for Au nanoparticles formed at higher annealing temperature of 550 °C, as shown in Figure 5a, which agrees well with the prediction based on simulations of light scattering and trapping of metal particles with different shape and size.⁹ A more indepth discussion will be given in the discussion of Figure 6 in simulation of the shape effect of the Au nanoparticles.



Figure 6. Simulated LSPR for Au particles on FTO surface with perfect contact (squares), Au particles on FTO surface with imperfect contact (circles), and Au particles inlaid into FTO (stars).

Figure Sb shows the transmittance spectra of plasmonic Au nanoparticles on patterned FTO nanolines and nanopillars processed under the same conditions in Figure 3. For comparison, the spectrum of plasmonic Au nanoparticles on flat FTO also annealed at 550 $^{\circ}$ C for 30 min is included in the same Figure. The LSPR peak position of the Au nanoparticles on patterned FTO is ~610 nm irrespective of the patterns,

which is red-shifted as well as broadened as compared with that of the Au nanoparticles on flat FTO (580 nm). The red shift and broadening of the LSPR absorption spectra may be attributed to the size nonuniformity of particles on patterned FTO surfaces, as evidenced from Figure 3. In addition, the transmittance is considerably reduced by replacing the flat FTO with the patterned one, which may be ascribed to the difference of reflectance, absorption, particles structure and size. Such a red shift and broadening of resonance is expected to be advantageous for thin-film photovoltaics due to enhanced light trapping. Interestingly, the combination of the plasmonic Au nanoparticles with nanoline patterned FTO has the lowest transmittance and therefore the strongest light trapping.

To obtain some insights into the origin of the LSPR features on the observed transmission curves shown in Figure 5, we carried out an FDTD simulation based on a FULLWAVE simulation package¹⁷ (R-Soft). A Au nanoparticle in the disk shape was considered in the simulation with 50 nm in diameter and 15 nm in thickness. The Au nanoparticle was placed at the interface between FTO and air, and a plane lightwave was normally launched with a single polarized electrical field E_x . The simulation area was 100 nm \times 100 nm in the horizontal dimension and 30 nm into the substrate in the direction normal to the interface. Perfectly matched layer (PML) condition was assumed for the boundary of the simulation window, which absorbs the energy without inducing reflections. Only one Au disk was included in the simulation area to minimize the simulation time. This means the effect of interference between different Au nanoparticles was not included. The transmittance was evaluated by placing a power meter 30 nm away from the Au/FTO interface inside the FTO film to measure the optical power that passed through the interface. The measured power was then normalized for the cases of with and without Au nanoparticle on the interface. The Drude model¹⁸ was applied for complex dielectric function of Au.

The curve marked with open squares in Figure 6 shows the transmittance simulated using a regular Au disk with the perfect contact on the FTO interface as illustrated by the inset at the bottom left corner of the Figure. A minimum transmittance was found at \sim 605 nm, which is \sim 25 nm longer than the measured resonance wavelength in Figure 5a. The curve marked with open circles in Figure 6 was obtained using a Au disk with nonideal contact to the FTO interface. In particular, only a 36 nm diameter area of this 50 nm Au disk is in contact with the FTO film, as illustrated by the inset near the top of the Figure. This nonideal contact between Au nanoparticle and FTO reduces the resonance absorption wavelength to ~590 nm, which agrees well with the measured LSPR wavelength of 580 nm, as shown in Figure 5a. In fact it is possible for the surface tension to cause the slight undercut of Au particles and introduce imperfect contact between Au particles and FTO substrate.

Another consequence of imperfect contact between Au particle and the FTO surface is the reduced amplitude of resonance absorption. This explains the measured result shown in Figure 5a, in which annealing at 550 °C for 30 min produces a stronger LSPR resonance absorption compared with annealing at 300 °C for 10 h due to the more collapsed shape of Au nanoparticles on FTO and thus better surface contact. In fact, if the Au particle is further inlaid into the FTO, then the resonance absorption will be even stronger, as shown by stars in Figure 6. At the same time, the resonance wavelength will be red-shifted. This explains the measured



Figure 7. Simulated transmittance of FTO/Si interface with (solid line) and without (dashed line) Au disk at the interface. (a) Silicon layer is considered semi-infinite so that there is no reflection from the top boundary and (b) the silicon layer is 300 nm thick, which forms a slab waveguide layer.

results shown in Figure 5b where some of the Au particles fall into holes on the patterned substrate, as can be seen in Figure 3a,b. In fact, on the basis of the Mie scattering theory, plasmonic resonance wavelength is determined by both the material property and the size of the metal particle as well as the dielectric constant of the material surrounding the metal particle. Assuming that the particle size is much smaller than the wavelength, the classical formula for Rayleigh scattering can be used, and the scattering cross section is proportional to:^{19,20}

$$Q_{\rm sca} \propto \left(\frac{a}{\lambda}\right)^4 \left|\frac{\varepsilon_{\rm m} - \varepsilon_{\rm s}}{\varepsilon_{\rm m} + 2\varepsilon_{\rm s}}\right|^2$$

where λ is the wavelength, *a* is the radius of the particle, and ε_m and ε_s are dielectric constants of the particle and the surrounding material, respectively. Because the real part of the dielectric constant of Au is negative and its magnitude increases with the wavelength, the resonance of scattering cross-section in the vicinity of $|\varepsilon_m| = 2\varepsilon_s$ would move to longer wavelength with the increase in ε_s . The inlaid Au nanoparticles have a better contact with the substrate and hence stronger dielectric effect of the substrate, which contributes to the increased resonance absorption as well as the red shift of the resonance wavelength.

Our simulation also indicated that a larger particle size would generally increase the scattering cross-section and also increase the resonance wavelength,⁹ which can be understood as due to the increased cavity size. Specifically for the structure under study, increasing nanoparticle diameters from 50 to 70 nm would increase resonance wavelength from 610 to 720 nm. Experimentally, with patterned FTO surface, the size of the Au nanoparticles becomes less uniform and the surface contact condition also becomes increasingly nonregular, as shown in Figure 3. These are the major reasons for the broader wavelength window of plasmonic resonance shown in Figure Sb.

In photovoltaic applications, Au nanoparticles created on the FTO transparent electrode may provide a promising approach to increase resonance absorption through plasmonic effect. Figure 7 shows the simulated transmittance when Si is used as the photoactive material. The photonic structure used for simulation is shown in the inset of Figure 7, where a Au disk with 30 nm in diameter and 15 nm in thickness is incorporated between the Si/FTO interfaces. In Figure 7a, the power

monitor was placed inside the silicon layer 300 nm away from the Si/FTO interface, and we have assumed 30% optical attenuation across the 300 nm silicon layer. The width of the simulation area was ± 30 nm, and periodic boundary condition was used on the side walls, which is equivalent to taking into account periodically spaced Au particles with 60 nm horizontal spacing between each other. We have used PML boundary condition for the silicon/air interface behind the power monitor to eliminate optical reflection in the vertical direction. The solid and the dashed lines show the transmittance with and without Au particle at the Si/FTO interface. Without Au disk, the transmittance is primarily determined by the linear absorption of the silicon layer and the Fresnel reflection on the Si/FTO interface. With the Au disk, the plasmonic resonance wavelength is at ~1100 nm. In this configuration, because the majority of the surface area of the Au disk interfaces with silicon, which has a much higher refractive index than air, the red shift of plasmonic resonance wavelength is expected in comparison with Figure 6.

The results shown in Figure 7a are focused on the investigation of transmittance only due to Si/FTO interface so that the power monitor was placed inside the silicon layer and a PLM boundary conduction was used behind the power meter, and thus the silicon/air interface is perfectly absorbing without reflection. However, for a practical solar cell, the thickness of silicon layer is deterministic, which forms a slab optical waveguide. Considering the Fresnel reflections on the top (Si/air interface) and the bottom (Si/FTO interface) facets, the transmittance for optical power of normal incidence can be described by a Fabry–Perot equation:²¹

$$T(\lambda) = \frac{\eta (1 - R_1)(1 - R_2)}{(1 - \eta \sqrt{R_1 R_2})^2 + 4\eta \sqrt{R_1 R_2} \sin^2(2\pi nd/\lambda)}$$

where R_1 and R_2 are Fresnel reflectivity of the two facets, η is the optical absorption of the silicon layer, n is the refractive index, and d is the layer thickness. To take into account the slab waveguide effect of the silicon layer in the simulation, we moved the power monitor immediately outside the 300 nm thick silicon layer, as illustrated in the inset of Figure 7b.

The dashed line in Figure 7b shows the calculated transmittance in the 450–900 nm wavelength window without the Au disk at the Si/FTO interface, and the result exhibits a typical Fabry–Perot transfer function. When a gold disk with

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30 nm diameter and 15 nm thickness is added at the Si/FTO interface, the transmittance is significantly reduced in the long wavelength region due to the plasmonic effect. The Au nanodisk not only creates plasmonic resonance absorption of the near field on the metal surface but also scatters the normally incident lightwave as illustrated in Figure 4 and creates horizontally propagated slab waveguide mode in the silicon film. This significantly increases the propagation length of the optical signal inside the active layer of the solar cell and thus improves the conversion efficiency. Note that the effect of the plasmonic Au nanoparticles on patterned FTO is manifested in the light scattering in far field and light trapping in near field in proximity to the Au nanoparticles that act as an effective "antenna" to store the incident energy from light to an LSPR mode. In the study presented here, the absorption of the substrate is not considered. However, in practical photovoltaic applications, the absorption rate in the semiconductor around these antennas needs to be larger than the reciprocal of the plasmonic decay time of typically 10-50 fs to avoid the LSPR energy dissipation to Ohmic loss.⁹ This high absorption rate is achievable in many organic and direct bandgap inorganic semiconductors, which assures that the resonant near field will primarily contribute to the generation of photocurrent.

CONCLUSIONS

In conclusion, an experimental study integrated with theoretical modeling and simulation has been carried out to understand the light management effect of plasmonic Au nanoparticles on FTO transparent electrodes with or without photonic nanostructures. It has been found that the plasmonic Au nanoparticles can be obtained in a wide temperature range of 300-550 °C via thermally assisted self-assembly from coated Au films on both original (or flat) and nanopatterned FTO surfaces (nanopillars and nanolines) fabricated using nanoimprint lithography. Nevertheless, the geometry of the Au nanoparticles and their interface with FTO are affected by annealing temperature and microscopic morphology of the FTO surface; both affect the LSPR frequency and peak width. Importantly, this work has demonstrated that the LSPR by Au nanoparticles and the light scattering by the photonic nanostructures on FTO can be combined, generating complementary broadband light trapping in solar spectrum. This plasmonic-photonic nanostructured FTO can be scaled up for thin film solar cells with much enhanced light absorption and power conversion efficiency.

AUTHOR INFORMATION

Notes

The authors declare no competing financial interest.

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