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Increasing the optical absorption in a-Si thin films by embedding gold nanoparticles

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ABSTRACT

The light conversion efficiency of traditional a-Si thin-film solar cells is limited by their low optical thicknesses, especially in the NIR. A possible approach to increase the light-trapping efficiency over the entire solar spectral range is to design solar-cell architectures which rely on the optical properties of plasmonic nanocomposite materials. We demonstrate that it is possible to have a controlled Gold nanoparticle optical absorption by varying the thickness of a covering a-Si thin-film. For thick a-Si films the Gold nanoparticle plasmon resonance vanishes likely due to the formation of a silicide. Optical absorption measurements as well as finite difference time-domain (FDTD) simulations were employed to determine the a-Si thickness-dependent optical absorption properties, which demonstrated a significantly increased optical absorption in a-Si.

1. Introduction

The incorporation of plasmonic nanostructures in the design of solar cells has shown potential for the development of high-efficiency, low-cost thin-film solar cells [1,2]. Light trapping techniques using plasmonic nanostructures have been the subject of considerable interest and, specifically, in solar cell architectures employing metallic nanoparticles as scattering elements [3–5] or in textured plasmonic-back reflectors [6–9]. The metallic nanoparticles enhance the optical absorption by increasing the optical path of the incident light inside the thin semiconductor absorber layer with the theoretical possibility of exceeding the Yablonovitch $4n^2$-thermodynamic limit on the light absorption [10].

Another possible approach to enhance the optical absorption consists in embedding the metal nanoparticles inside the semiconductor absorbing layer making use of their plasmonic near field properties. The metal nanoparticles act as antennas, capturing the energy of the incident light into localized surface plasmon resonance (LSPR) modes. The local electric field near the metallic surface can become orders of magnitude higher than that of the incident field. The plasmonic near field enhancement has been exploited both in organic and dye-sensitized solar cells [11–15] and in inorganic solar cells, such as CdSe/Si [16] and amorphous Silicon (a-Si) thin-film hetero-structures [6,17–19].

In silicon based solar cells, the inclusion of metallic nanoparticles inside the semiconductor photoactive layer theoretically favors the direct absorption of the radiation through scattering and the near-field enhancement effect [20]. However, it may also be responsible for an efficiency drop of the device performance since the introduction of additional metallic structures into the semiconductor photoactive layer causes further ohmic losses and may introduce significant structure defects which are quite detrimental to the photocurrent by increasing the recombination rate [21,22]. It is therefore of paramount importance to tailor the size, density, shape and location of the nanoparticles in the design of an appropriate solar cell architecture in order to provide both an efficient light trapping scheme and a reduction of the energy losses allowing an enhancement over the entire spectrum of the photovoltaic performance [21,23]. Although Ohmic losses in the metallic nanoparticles are considered an obstacle for thick solar cells [24], for very thin film solar cells the benefits may outweigh these parasitic losses since their optical absorption can benefit from the smallest boost [25]. In particular in thin a-Si photoactive layers, the light trapping scheme should be able to achieve a large absorption cross section near the band-edge regions (600–800 nm) of the absorption spectrum where the absorption coefficients typically become

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small and the efficiency is low. Therefore a better understanding of the optical properties of a-Si nanocomposite containing metallic nanoparticles is important and may lead the further optimization of light-trapping schemes being effective across the whole useful solar spectrum. This is particularly of importance to thin film solar cells which have a considerable market share and offer interesting prospects [26,27].

In the present work, we report the possibility to tune the light absorption inside thin a-Si films deposited on glass substrates and incorporating a random array of Gold nanoparticles from the visible toward the NIR as a function of the film-thickness. The nanoparticles were fabricated by a gas phase aggregation clusters source which produces ultra clean nanoparticles of any composition at a fast rate [28]. Recently such nanocluster sources have been used to explore light management for photovoltaics [29–31], providing a novel and economically interesting approach. UV–Vis spectroscopy measurements showed thickness-dependent absorption properties in the Gold/a-Si nanocomposite material. The effective dielectric environment causes a redshift of the Gold nanoparticles plasmonic resonance inside a-Si with increasing thickness. These experimental findings are explained with finite-difference time domain (FDTD) simulations, by which also the role played by the near field coupling between Gold nanoparticles is elucidated. The intensity and the broadening of the plasmonic resonance as a function of the a-Si layer thickness and Gold nanoparticles deposition time is discussed as well.

2. Experimental procedure

Gold nanoparticles were deposited with a gas aggregation nanocluster source based on magnetron sputtering (NC200U-B Oxford Applied Research Ltd.) [32,33] on glass substrates. For the deposition of nanoparticles, argon gas was used as both sputter and carrier gas inside the gas aggregation chamber. The Au target had a purity of 99.99%, the argon flow rate in the cluster source was 15 sccm, the magnetron DC power 30 Watts and the aggregation length 60 mm for all samples. The deposited nanoparticles were subsequently covered with a conformally grown a-Si thin film, also by magnetron-sputtering from a polycrystalline silicon target (99.9%). All thin film depositions were performed by fixing the value of the forwarded power to 124 W and an argon flow of 40 sccm. The thickness of the deposited a-Si films was controlled by varying the thin film deposition time: 100, 200, 300 and 400 s. The corresponding estimated film thicknesses, determined by a profilometer (KLA-Tencor stylus profiler), are respectively: 6, 12, 17 and 23 nm with an accuracy of ±2 nm. A schematic cross-sectional representation of the samples in the present work is given in Fig. 1. An experimental scanning electron microscopy (SEM) cross section of a similar sample confirming the conformal growth can be found in Ref. [34]. The reference samples consisted of only an a-Si film deposited on glass with the same thicknesses as the corresponding a-Si films deposited on the Gold nanoparticles.

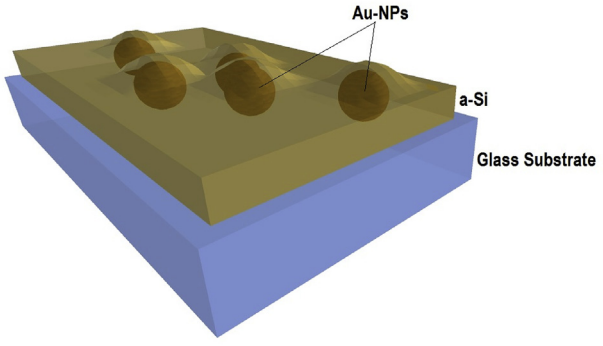

Fig. 1. Schematic depiction of a cross-section of a sample characterized in the present work.

The Gold nanoparticles were investigated with a Transmission Electron Microscope (TEM) Philipps CM10 (FEI) electron-microscope operated at 80 kV. TEM micrographs were taken of Gold nanoparticles deposited for 20s on holey a-C TEM grids keeping the deposition parameters identical to the parameters used for the nanocomposite samples which were deposited on glass. The morphological characteristics of the sample-surfaces were determined by atomic force microscopy (AFM) using a Bruker-Catalyst Atomic Force Microscope in Peak- Force Tapping® mode in air, equipped with ScanAsyst-AIR tips. UV–visible absorbance spectra of the samples were recorded with a commercial double beam Agilent Cary-100 spectrophotometer with a wavelength range from 190 to 900 nm and a wavelength resolution of 0.50 nm.

To study the effect of a-Si on the Gold plasmon resonance energy, the experimental optical absorbance spectra of the Gold nanoparticle samples were subtracted with the absorbance of the reference a-Si film or the reference glass substrate in the case of the “bare” Gold nanoparticles. The measured absorbance spectra were compared to the total optical absorption, defined as the ratio of absorbed light with respect to the incident light power, obtained from finite difference time-domain simulations (FDTD).

The FDTD simulations were performed with a commercial software (Lumerical Solutions Inc.) on systems consisting of two spherical Gold (Palik [35]) nanoparticles supported on a glass substrate and embedded in a-Si films with different thicknesses (Fig. 4b). The optical data for a-Si (Inset Fig. 6a) were obtained from spectroscopic transmission and reflection measurements.

The mean size of the Gold nanoparticles (24 nm) determined from the TEM micrographs was used to model the diameter of the simulated spherical nanoparticles whereas the mean inter particle distance obtained from a statistical analysis performed on the AFM images was used as the distance between the two simulated spherical Gold particles. The simulation of only two interacting gold particles was chosen to encompass all the present properties and interactions to describe the system. A periodic array would generate unwanted interference effects. A convergence test was performed which confirmed sufficient accuracy with a mesh size of $dx = dy = dz = 1.5$ nm on the Au-nanoparticles. The simulation box volume was $920 \times 920 \times 920$ nm^3 with 12 PML layers on each side.

3. Results and discussion

3.1. Morphology

The optical properties of a nanocomposite depend both on the material and morphological properties of its structural units (cluster or nanoparticles) and of its surfaces [37].

The geometrical structure as well as the mean size of the deposited Gold nanoparticles are determined primarily by the parameters chosen during the deposition. Different studies on the fabrication of nanoparticles by the gas aggregation process have shown that the cluster size distribution can be accurately described by a lognormal distribution [38,39]. This is confirmed by our statistical analysis of the TEM micrographs of the isolated Gold nanoparticles (Fig. 2a and b), which have a mean diameter of 24 ± 1.5 nm. The deposition of nanoparticles with a gas aggregation techniques produces both small Au-clusters and large nanoparticles having typically icosahedral or dodecahedral geometries [40] in agreement with the here observed hexagonal shapes.

Since the typical energy of the particles ejected from a gas-