



# Ultra-fast plasmonic back reflectors production for light trapping in thin Si solar cells

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## ABSTRACT

A fast method is presented to fabricate plasmonic light trapping structures in just ten minutes ( $> 5 \times$  faster than the present state of art), with excellent light scattering properties. The structures are composed of silver nanoparticles (Ag NPs) deposited by thermal evaporation and self-assembled using a rapid thermal annealing (RTA) system. The effect of the RTA heating rate on the NPs production reveals to be crucial to the decrease of the annealing process. The Ag NPs are integrated in thin film silicon solar cells to form a plasmonic back reflector (PBR) that causes a diffused light reflectivity in the near-infrared (600–1100 nm wavelength region). In this configuration the thicknesses of the AZO spacer/passivating layers between NPs and rear mirror, and between NPs and silicon layer, play critical roles in the near-field coupling of the reflected light towards the solar cell absorber, which is investigated in this work. The best spacer thicknesses were found to be 100 and 60 nm, respectively, for Ag NPs with preferential sizes of about 200 nm. The microcrystalline silicon ( $\mu\text{-Si:H}$ ) solar cells deposited on such improved PBR demonstrate an overall 11% improvement on device efficiency, corresponding to a photocurrent of 24.4 mA/cm<sup>2</sup> and an efficiency of 6.78%, against 21.79 mA/cm<sup>2</sup> and 6.12%, respectively, obtained on flat structures without NPs.

## 1. Introduction

Thin-film (TF) photovoltaic (PV) technology is one of the candidates to take over from the current commercial-dominant Si wafer-based technology in the long-term, as it has potential for higher performance/cost ratio due to the use of less amount of silicon (Si) and by employing Chemical Vapour Deposition (CVD) methods which are deemed to be more cost-effective than the traditional crystalline silicon (c-Si) ingot cutting (Tan et al., 2013a, 2013b, 2012). Moreover, solar cells with film thicknesses in the 1–2  $\mu\text{m}$  range can be deposited on cheap module-sized substrates which can be rigid, such as glass, or mechanically flexible, such as plastic (Wilken and Smirnov, 2014), stainless steel foils (Atwater and Polman, 2010) or even paper-based (Vicente et al., 2015; Águas et al., 2015). A common limitation to many TF PV technologies, relays with a low light absorbance for energies near the bandgap (the absorption coefficient decreases significantly with increasing illumination wavelength) (Rech et al., 2004), due to their reduced absorber thicknesses, in particular for the indirect-bandgap semiconductor Si. For example, although the Si bandgap lies at  $\sim 1120$  nm, in nanocrystalline (nc-Si) TFs, the absorption coefficient drops quickly below

1000 cm<sup>-1</sup> for wavelengths above 800 nm (Meillaud et al., 2015; Yan et al., 2012).

Enhancement of optical absorption by light trapping (LT) is crucial to obtain high efficient TF Si solar cells. LT structures makes the long-wavelength photons travel in the solar cell distances much longer than the device thickness, increasing the probability of their absorption. This can be achieved by the scattering of light at the cell's interfaces, either by transmission at the top interfaces or by reflection at the rear ones. In conventional TF Si solar cells, LT is typically achieved using random texturing of either the front transparent contact, in superstrate cell configurations (Rech et al., 2004; Yan and Venkataraj, 2013; Ju et al., 2009), or of the rear contact/mirror in substrate configurations (Palanchoke et al., 2013; Rath, 2014; Haug et al., 2013). However, the texturing approach produces high surface roughness which contributes to the creation of bulk and surface defects (charge carrier traps) and, as a consequence, increases recombination in the silicon layers which degrades the electrical performance of the device (Karin et al., 2010). A recent alternative solution consists in the use of metal nanoparticles (MNPs), providing low surface roughness, to plasmonically scatter light (Tan et al., 2013a, 2012; Mendes et al., 2014a, 2014b). Ag NPs have

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been applied mostly due to their strongest plasmon resonances in the visible to near-infrared (NIR) spectral region (Bohren and Huffman, 1983; Temple et al., 2009; Hartland, 2011), and due to the fact that they can be fabricated with high control of size and shape by various methods (Atwater and Polman, 2010; Morawiec et al., 2013; Araújo et al., 2016). The optimized solution should provide strongest LT, via light scattering, keeping the PV film as flat as possible, to maximize both the photo-generated current and the operating voltage and fill factor. In our work our corrugations are smaller and less sharp (more flat) than the conventional textures because the NPs are also small (100–200 nm) and round (not pointy). This contributes to increase the photocurrent of the solar cell maintaining the electrical properties.

The resonant optical properties of MNPs depend strongly on the geometry and size of the particles, as well as on their material and surrounding media (Temple et al., 2009; Gaspar et al., 2013; Kelly et al., 2003; Araujo et al., 2016). Thus, such fabrication parameters must be well designed to maximize scattering and minimize the parasitic losses (light absorption within the MNPs) across the wavelength range of interest, which for TF Si solar cells corresponds to the NIR (600–1100 nm range) (Tan et al., 2013). Solid-state dewetting (SSD) is a well-known industrially-scalable process capable of morphologically transforming (by self-assembly) a continuous thin film in islands or droplets due to a thermal treatment. This technique is commonly used for the fabrication of metallic nanostructures (Tan et al., 2013; Mendes et al., 2014; Thompson, 2012), with advantages of low-cost, simplicity, direct scalability and compatibility with industrial manufacturing processes. However, most of the typically employed annealing processes (e.g. using a tube furnace) require annealing times of one or more hours (Morawiec et al., 2013; Nasser et al., 2013; Bai et al., 2014) to form the preferential morphologies composed of individual 100–300 nm semi-ellipsoidal MNPs. The RTA (Bai et al., 2014) process optimized in this work is an advantageous method to yield such plasmonic surfaces with high reproducibility and large-area uniformity within just a few minutes of annealing time (Araújo et al., 2016), bringing industrial benefits in terms of time and energy consumption. Yiming Bai *et al.* observed that the rapid annealing treatment allows a more uniform size distribution of the nanoparticles, compared with the in-situ heating process. The agglomeration of Ag films annealed under in-situ (IS) mode derived from the fractal branched growth of void that resulted in small-sized particles occurring, while Ag films annealed under RTA mode agglomerated following the grain boundary grooving (GBG) mechanism to favor the formation of NPs with uniform size.

However, more work is needed due to the lack of robust experimental proofs about the influence of the rapid annealing treatment, namely the ramp heating rate on the nanoparticles sizes and distribution and also in the time of the annealing to obtain this type of NPs. The first part of this study concerns the investigation of the effect of the RTA heating rate on the morphological properties of Ag NPs, which revealed to be the key parameter that enabled the dramatic decrease in the annealing process time. By increasing the heating rate, PBRs with pronounced light diffusion and reflection properties, comparable to the best in the state-of-the-art, have been formed in only 10 min of RTA.

In solar cell applications, the Ag MNP arrays are usually embedded in a thin transparent conductive oxide (TCO) layer (Tan et al., 2012, 2013b; Thompson, 2012; Santbergen et al., 2012), located between the silicon absorbing material and a flat Ag mirror (rear contact), in a configuration known as PBR (Tan et al., 2013a; Atwater and Polman, 2010; Nasser et al., 2013; Araújo et al., 2013). In a previous work, the authors have shown that the physical properties of such TCO layer play a determinant role on the morphology of the nanostructures formed on it (Araujo et al., 2016). Besides, the position of the MNPs array within the TCO is also of crucial importance to optimize the distance-dependent near-field optical interaction (parasitic absorption and scattering) with both the silicon layer and the mirror (Tan et al., 2013a; Mendes et al., 2014b), as has been studied theoretically (Sesuraj et al., 2013).

In this paper, we study the design routes of plasmonic PBR produced

by ultra-fast RTA process applied to hydrogenated  $\mu\text{-Si:H}$  solar cells, where both, thickness of spacer AZO layer between silver film and silver nanoparticles and spacer AZO layer between silver nanoparticles and n-doped  $\mu\text{-Si:H}$  layer are found to have a significant influence on the performance of a solar cell with PBR. We experimentally also demonstrate that the thickness of the bottom AZO layer is crucial on the morphological and optical performance of the PBR and consequently on the optical performance of the solar cell.

## 2. Experimental details

The plasmonic BRs were fabricated with the structure: glass/Ag mirror (120 nm)/AZO/Ag NPs/AZO (see Fig. 3). The planar Ag mirror layers on glass were deposited by electron gun-assisted thermal evaporation technique, and the Al-doped ZnO (AZO, ZnO:Al) (Lyubchik et al., 2016) layers were deposited by RF magnetron sputtering. The Ag NPs were formed from the deposition of thin Ag layers at room temperature, by electron gun-assisted thermal evaporation, on glass with sizes at list  $20 \times 20 \text{ cm}^2$  followed by annealing under nitrogen atmosphere using a RTA system. The evaporation of the Ag thin films was carried out with a base pressure of  $10^{-6}$  mbar, a working pressure of  $10^{-5}$  mbar and a deposition rate of 0.07 nm/s. A calibrated quartz crystal sensor inferred the thicknesses and growth rate of the films, with an error < 1%. A series of 6 nm Ag films were deposited on glass, previously coated with 60 nm of AZO, under the same conditions. The films were then annealed in RTA at 500°C under nitrogen atmosphere, using different temperature ramp-up rates (ranging from 16 to 150 °C/s) and for different times (2–40 min) of steady-state (close to 500 °C) temperature.

The thickness of ZnO:Al layers between planar Ag film and Ag NPs ( $d_{\text{Ag-NPs}}$ ) and the thickness between Ag NPs and n-doped a-Si:H layer ( $d_{\text{NPs-Si}}$ ) were varied and their influence in the PBR performance was evaluated.

The size of the Ag NPs was measured using a Carl Zeiss AURIGA CrossBeam FIB-SEM workstation Hitachi 4800 scanning electron microscopy (SEM). The surface coverage, NP size distribution, and circularity ( $4\pi \cdot \text{area}/\text{perimeter}$ ) were determined from the SEM images using the Image J image processing software (Abramoff et al., 2004). The surface morphology of Ag NPs was also characterized by an atomic force microscopy (AFM). The total and diffuse reflectance of the plasmonic BRs was measured with a double beam UV–Vis–NIR spectrometer (Lambda 950) equipped with an integrating sphere, in the wavelength range of 300–1100 nm.

Hydrogenated microcrystalline silicon ( $\mu\text{-Si:H}$ ) solar cells, with an n-i-p (substrate) configuration (shown in Fig. 5(b)) were deposited on the PBRs at a temperature of 160 °C using a multi-chamber plasma-enhanced chemical vapor deposition (PECVD) system (Lyubchik et al., 2015), elsewhere described. The thickness of the n-i-p layers were 54, 1500, and 20 nm, respectively. This was followed by the application of a Kapton mask with circular holes, 3 mm in diameter, on the cell's front surface and by the deposition of the IZO front electrode via RF magnetron sputtering. After removing the masks, reactive ion etching with SF<sub>6</sub> plasma using a Trion Phantom III RIE ATCH system was applied to remove the Si material left uncovered by the IZO layer, thereby defining circular devices with 3 mm in diameter. The solar cells were characterized by measuring the J–V curves under AM1.5 G ( $100 \text{ mW cm}^{-2}$ ) illumination in a Spire 240 A Sun Simulator, under standard conditions and by measuring the external quantum efficiency (EQE) in short-circuit condition in the 400–1100 nm wavelength range. The open-circuit voltage and fill factor of the cells were extracted from the J–V curves, and the short-circuit current density was determined from the integration of the EQE convoluted with the AM1.5 G photon flux. The reflective properties of the PBR and solar cells were measured with the aforementioned spectrophotometer.

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