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Colloidal self-assembled nanosphere arrays for plasmon-enhanced light trapping in thin film silicon solar cells

Manuel J. Mendes[a,*], Seweryn Morawiec[a], Isodiana Crupi[a], Francesca Simone[a] and Francesco Priolo[a,b]

[a]*MATIS CNR-IMM and Dipartimento di Fisica e Astronomia, Università di Catania, via S. Sofia 64, 95123 Catania, Italy*
[b]*Scuola Superiore di Catania, Via Valdisavoia 9, 95123 Catania, Italy*

Abstract

To realize high-efficiency thin-film silicon solar cells it is crucial to develop light-trapping methods that can increase absorption of the near-bandgap light in the silicon material. That can be achieved using the far-field scattering properties of metal nanoparticles (MNP) sustaining surface plasmons. The MNPs should be inserted in the back of the cell, embedded in the transparent conductive oxide (TCO) layer which separates the rear mirror from the silicon layers. In this way, a plasmonic back reflector (PBR) is constructed that can redirect light at angles away from the incidence direction and thereby increase its path length in the cell material.

In this work, a novel technique is presented to fabricate PBRs (composed of Ag mirror/TCO/MNPs/TCO) containing colloidal gold MNPs patterned with a self-assembly wet-coating method. The method allows the construction of long-range ordered arrays of MNPs with monodisperse size and shape using fast, scalable, low-cost and low-temperature (<120^{o}C) procedures.

Colloidal MNPs are synthesized with spherical shapes, so their scattering properties are analytically modeled with Mie theory. Such formalism allowed the computation of the preferential MNP sizes that provide the best scattering performance for light-trapping in amorphous and microcrystalline thin-film silicon solar cells.

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Keywords: Thin film solar cells; Plasmonics; Light trapping; Colloidal Metal Nanoparticles; Mie theory

1. Introduction

Thin-film (TF) solar cells have stimulated enormous research interest as a low-cost alternative to bulk crystalline cells [1,2]. However, a limitation common to all thin-film photovoltaic (PV) technologies is that the absorption of the near-bandgap light is small, due to their reduced material thickness. The application of light trapping methods is a promising strategy to increase the optical density (and consequently the quantum efficiency) of TF cells at the wavelengths that are poorly absorbed by its PV material [3-6]. In addition, light trapping enables further reduction in the thickness of the PV material, which improves the conditions for charge carrier collection and lowers even more the material costs of the devices.

* Corresponding author. Tel.: +39-095-378-5239; fax: +39-095-378-5243.
E-mail address: manuel.mendes@ct.infn.it

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Nowadays, most commercial TF solar panels are composed of thin-film silicon (Si) layers grown on textured substrates. The texturing redirects the incident light to angles away from the incidence direction, thereby enhancing the path length of the photons inside the cell and, consequently, their absorption probability. However, the roughness induced in the Si layers causes dislocations and defects in the cell material, which have been shown to be detrimental to the open-circuit voltage and fill-factor of the devices [7].

The light trapping method developed in this work allows the enhancement of the light absorbed in the cell without producing roughness in the Si layers. The method makes use of the light scattering properties of metallic nanoparticles (MNPs) sustaining surface plasmons (SPs) [3,8,9]. Sub-wavelength MNPs can act as antennas at optical frequencies, gathering the light from their surroundings and scattering it to their far-field over a broad angular range. This makes the light follow more horizontal paths, in a similar way as occurred with texturing, increasing its absorption in the PV material. The SP resonance can be particularly pronounced in MNPs made of noble metals such as silver (Ag) or gold (Au), due to their low imaginary permittivity. Nevertheless, MNPs can also exhibit significant light absorption, so they should be preferentially located in the rear side of the cell, acting as a plasmonic back reflector (PBR), in order for them to interact only with light that is not absorbed in the first pass through the cell material [4,10,11]. The proposed PBR configuration is depicted in Fig. 1.

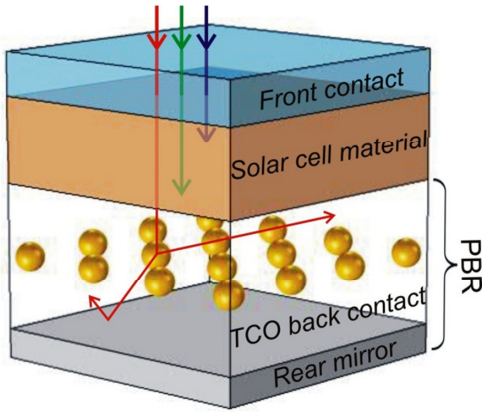

Fig. 1: A typical back reflector (also back contact) of a TF cell is composed of a metallic mirror (made of silver or aluminum) separated from the p-n junction(s) (referred in the image as solar cell material) by a TCO layer. The MNP structure should be preferentially located inside such TCO layer, since the TCO acts as a barrier for the diffusion of metallic impurities to the cell material [12] and prevents current degradation through carrier trapping and recombination at the MNPs surface.

Most studies performed so far, aimed at developing PBRs, construct the MNP structures by annealing a thin layer of a precursor metal [10,13]; a technique known as thin-film annealing (TFA). It has already been demonstrated that Ag MNPs, fabricated by TFA and embedded in a back reflector configuration, can provide light trapping performance comparable to state-of-the-art random textures in hydrogenated amorphous silicon (a-Si:H) solar cells [4,11]. However, there is a high non-uniformity in the geometry of MNPs fabricated with TFA and their material is usually not purely crystalline [10], which results in an attenuation of the resonant optical response of the overall nanostructures. Besides, TFA requires high-temperature (>300^{o}C) annealing to produce nanoparticles with appropriate sizes, so the MNP structure has to be fabricated prior to the deposition of the Si cell layers, otherwise the high temperature would damage the hydrogen bonds of the hydrogenated TF Si material.

The approach presented here is promising to overcome the aforementioned issues. A self-assembly colloidal deposition technique was developed to construct long-range ordered arrays of crystalline Au nanoparticles with monodisperse size and shape [14,15]. Using such wet-coating method, MNP arrays have been patterned throughout indefinitely large surfaces employing low-cost and low-temperature (<120^{o}C) procedures, which are compatible with large-scale production and allow the fabrication of the MNP structures as a post-process after the deposition of the Si cell layers.

Colloidal MNPs are synthesized in solution with approximately spherical shapes, so their scattering properties can be analytically modeled using Mie theory. This is an electromagnetic separation-of-variables method that can compute scattering by spherical particles of any size immersed in a homogeneous medium. In this work, a modified Mie theory formalism is used to account for absorption in the medium surrounding the particles [16]. Such formalism allowed the determination of the preferential particle diameters that can provide the best scattering performance, for distinct types of TF Si solar cells.

2. Mie theory studies

An important advantage of colloidal Au nanoparticle structures is that their optical response can closely match Mie theory computations, since they can be synthesized with highly mono-dispersed spherical shapes and with pure crystalline materials. In

the calculations performed here, the MNPs were taken to be immersed in a homogeneous Aluminum Zinc Oxide (AZO) medium, to model the incorporation of the particles in the TCO layer of the cell back contact (see Fig. 1).

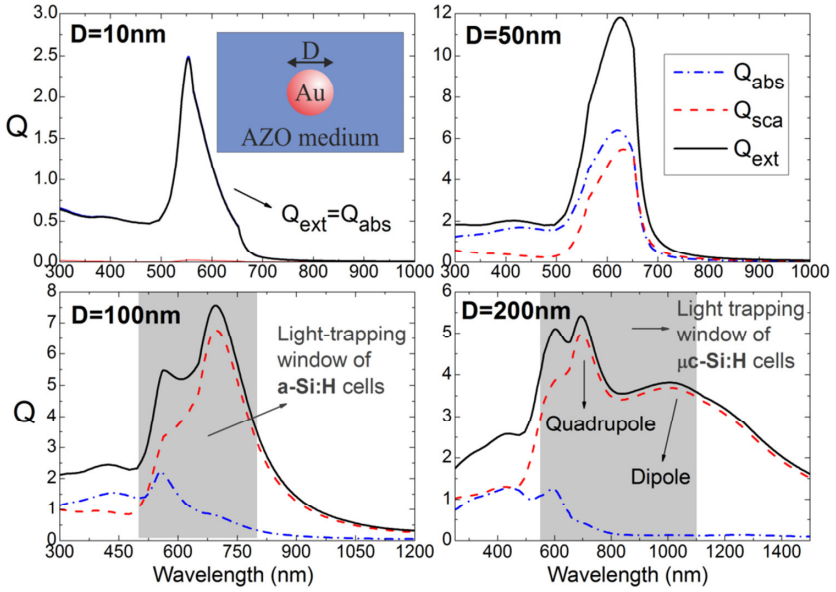

Fig. 2: Mie theory computations of the spectra of the extinction (Q_{ext}), scattering (Q_{sca}) and absorption (Q_{abs}) efficiencies of individual Au MNPs taken to be spherical with diameter D and immersed in an infinite AZO medium. The dielectric functions of the particle and medium materials were taken from experimental data [17]. The grey areas cover the preferential spectral window for light trapping in a-Si:H cells (in the plot of D=100nm) and μc-Si:H cells (in D=200nm).

The amount of light scattered by a MNP is determined by its scattering cross-section ($C_{sca}=W_{sca}/I_i$), a quantity with dimensions of area given by the ratio between the energy scattering rate (W_{sca}) and the incident irradiance (I_i) [9]. In the literature of single particle scattering, it is more usual to represent the dimensionless scattering efficiency, Q_{sca}, which is the ratio between C_{sca} and the particle cross-sectional area, A. Figure 2 compares Q_{sca} with the absorption ($Q_{abs}=W_{abs}/I_iA$, being W_{abs} the absorption rate) and extinction ($Q_{ext}=Q_{abs}+Q_{sca}$) efficiencies, for four different Au MNP diameters (D).

The results of Fig. 2 reveal that the smaller is the MNP the sharper is its plasmonic response. However, the smaller particles (diameters around 10nm or less) radiate little light to their far-field; the light is mainly absorbed inside the particles or in their near-field surroundings. The opposite occurs for bigger particles (D>100nm) where $Q_{sca}>>Q_{abs}$. Nevertheless, the optical response of the particle broadens with size; so, if the size is too big (D>200nm) the resonant character of the particle begins to disappear since several higher-order scattering modes (quadrupolar, octopolar, etc.) appear in the scattering spectrum.

A typical TF a-Si:H solar cell contains a ~300nm thick *p-i-n* junction which absorbs practically all the light with wavelength below 500nm in a first pass through the cell (illustrated by the green and blue rays in Fig. 1). On the other hand, photons with wavelength above 800nm cannot be absorbed by the cell material since their energy is below the a-Si:H bandgap [1]. Therefore, in a-Si:H cells the preferential spectral window for photocurrent enhancement via light trapping corresponds to the wavelength range between 500 and 800nm (the red rays in Fig. 1). According to the Mie theory results of Fig. 2, for the case of a-Si:H cells the preferential MNP diameter would be D=100nm, considering Au particles immersed in a homogeneous AZO medium. Such particles exhibit a pronounced Q_{sca} peak and small Q_{abs} in the desired spectral range (indicated by the grey area in the figure).

Hydrogenated microcrystalline silicon (μc-Si:H) is another widely used material in thin-film photovoltaics, mostly employed in double-junction TF Si solar cells composed of a μc-Si:H bottom cell and an a-Si:H top cell [7]. Cells made of μc-Si:H exhibit quantum efficiency response for wavelengths up to ~1050m, so their photocurrent can be enhanced by light trapping in a wider spectral range (550-1050nm) than that of a-Si:H cells. As such, for the case of μc-Si:H cells, it is preferable to have a wider dipolar Q_{sca} peak that can extend throughout the spectral window for light trapping in μc-Si:H cells. It is therefore advantageous to use a bigger particle size, such as the D=200nm MNP case shown in Fig. 2, that sustains broader and more red-shifted SP resonances relative to smaller particles.

3. Self-assembled arrays of colloidal gold nanoparticles

The colloidal Au MNP aqueous solutions used in this work were purchased from *Bristish BioCells* (BBI). Each solution contains particles with monodisperse diameter that remain stabilized in water by negatively-charged organic agents attached to their surface which cause the MNPs to repel each other and prevent their aggregation. A wet-coating method was developed to

self-assemble the colloids in long-range ordered arrays, on the surface of various materials [14,15,18]. That is achieved by chemically functionalizing the surface with molecular linkers that have positively charged end-groups which attract the negatively-charged nanoparticles. During the wet-coating process the MNPs are free to diffuse across the surface, so they settle in their lowest energy configuration which corresponds to an ordered lattice. The MNPs inter-particle distance is proportional to the MNPs surface charge density; so it can be tuned, for instance, by changing the capping agent, solvent liquid or pH of the solution. It was verified that such structure can extend throughout the entire area of arbitrarily large samples, and the wet-coating method can be adapted to practically any surface material, since most surfaces can be functionalized with positively-charged linkers.

So far this method has been mainly applied to deposit small MNPs (with sizes on the order of 10nm) on the surface of glass or Si. As discussed in the previous sections, for our application the MNPs should have bigger sizes (on the order of 100nm) and should be patterned on TCO films such as AZO. Therefore, the wet-coating method was adapted to pattern such big-sized MNPs onto AZO-coated glass substrates without affecting the electrical properties, i.e. the conductivity, of the AZO layers.

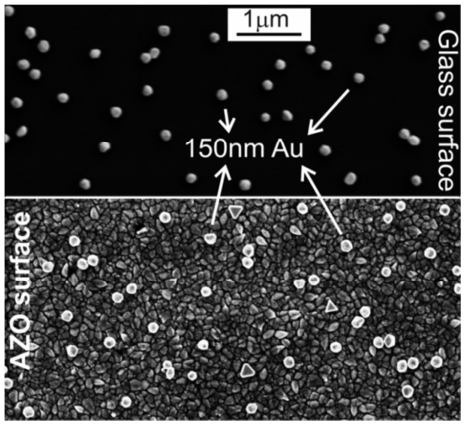

Fig. 3: SEM images of Au colloids with 150nm diameter patterned on glass (top) and on an AZO-coated glass (bottom) using the wet-coating method developed in this work. Most particles have a spherical shape, which was confirmed by cross-sectional SEM images (not shown).

Figures 3 and 4 are examples of the obtained MNP distributions. The images were acquired with a field-emission Zeiss Supra 25 scanning electron microscope (SEM). Fig. 4 shows large areas of the usual distributions obtained, which are similar throughout the entire surface of the samples. According to tests performed by the authors, the employed wet-coating method is able to pattern reproducible MNP distributions, similar to those of Fig. 4, with different MNP sizes between 60 and 150nm and throughout the surface of AZO films with distinct thicknesses in the 100-500nm range. The density of the patterned arrays is usually of 4-5 particles/μm^2, which corresponds to a surface coverage of ~5%. According to the maximum values of the scattering cross sections obtained with Mie theory calculations, it would be preferable a 3 times higher particle density in the array in order for the incoming light to completely interact with the MNPs.

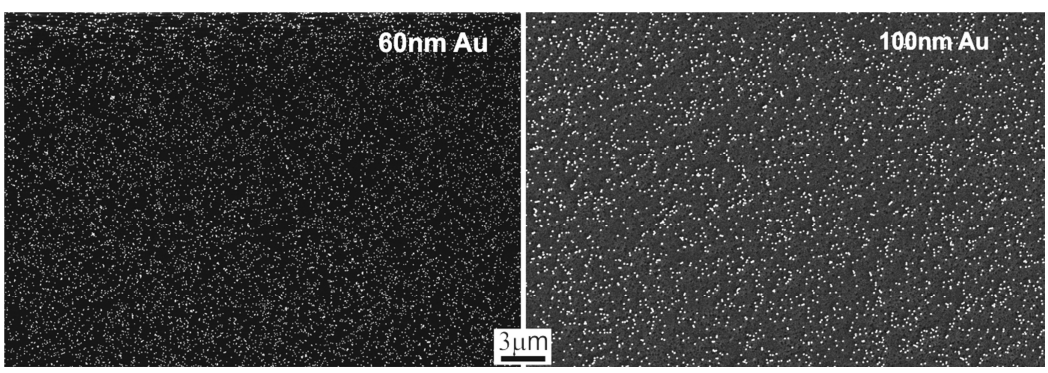

Fig. 4: Typical distributions obtained throughput the surface of 2.5x2.5cm^2 samples.
(left) 60nm diameter Au MNPs patterned on glass; (right) 100nm Au MNPs patterned on AZO-coated glass.

The plasmon resonance of the deposited MNPs in a AZO medium can be measured, and compared with the Mie theory results of Fig.2, by collecting their scattered light when the particles are inserted in between thick (relative to the MNPs size) AZO

layers. This has been performed using a Varian Cary 500 double-beam UV-Vis-NIR spectrophotometer equipped with an integrating sphere setup able to measure the diffuse reflected (R_{DIF}) and transmitted (T_{DIF}) light coming from the samples. Figure 5 shows the results of a sample containing an array of 100nm MNPs, as that of Fig. 4, patterned on a 500nm AZO layer and then covered by another 250nm AZO capping layer. All the AZO depositions in this work were performed by RF magnetron sputtering. The measured peaks in Fig. 5 correspond to the light scattered at the dipolar SP resonance of the particles and match precisely the Q_{sca} peak computed with Mie theory in the bottom-left plot of Fig. 2.

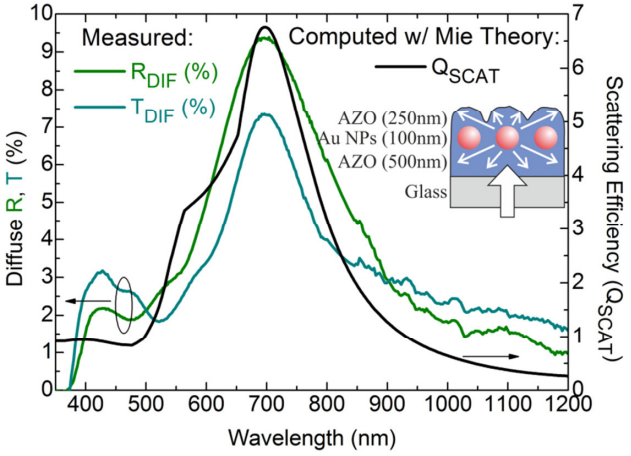

Fig. 5: Measured diffuse reflection and transmission (left axis) from an array of 100nm Au colloids deposited in between two AZO layers, as depicted in the sketch on the right. The thick white arrow in the sketch indicates the illumination direction in the measurement. The light diffusion peaks match the Q_{sca} peak (right axis) computed with Mie theory considering a single spherical MNP in an infinite homogenous AZO medium.

The results of Fig. 5 indicate that: 1) The physical properties (material, size, shape) of the MNPs are quite monodisperse in the array and correspond to the expected ones. 2) The distances between the deposited particles are long enough to prevent considerable inter-particle interactions. 3) The deposition of AZO material on top of the MNPs by sputtering is produced in a conformal way and does not affect the distribution of the particles. 4) The texturing in the top AZO surface induced by the MNPs shape should not have a significant contribution to the scattered light. Therefore, apart from the low surface coverage, the colloidal MNP distributions obtained present the preferential characteristics for their implementation in PBRs.

4. Colloidal Plasmonic Back Reflectors

Prototype PBR structures have been fabricated using the method described in section 3. The PBRs were characterized by measuring the total (R_{TOT}) and diffuse light reflected from the samples, using the previously referred spectrophotometry equipment. From these quantities the Haze in reflection (R_{DIF}/R_{TOT}) of the PBR is determined, which is directly related to the light scattered by the MNPs [4,9,10].

4.1. Substrate configuration

We start by analyzing the PBR structure shown in the inset of Fig. 6a. It contains an array of 100nm Au colloids deposited on a sample composed of a glass substrate coated with a layer of Ag (the back mirror) and AZO. After the deposition of the MNPs, the sample was covered with a top AZO layer that embeds the particles.

Figure 6a shows the R_{TOT} and Haze of the fabricated PBR, together with the R_{TOT} of a reference BR composed of the same structure but with no MNPs deposited. The R_{DIF} of such reference BR has also been measured, and it was verified that it is zero for the analyzed spectral range.

The Haze curve in Fig. 6a presents a pronounced peak in the 600-700nm range, which is within the spectral range of interest for a-Si:H cells (the grey area in the D=100nm plot of Fig. 2). When the PBR is incorporated in the rear of an a-Si:H cell, as depicted in Fig. 6b, the Si material can influence the scattering properties of the MNPs causing, among other effects, a red-shift in their SP resonances. The interaction between the MNPs and the Si layers has not been considered in this work. However, previous studies of PBRs fabricated using the conventional TFA technique [4,11] have demonstrated photocurrent enhancement in a-Si:H cells due to the light scattered from PBR structures with maximum Haze around 600nm. That is close to the wavelength of the Haze peak in Fig. 6a, so such colloidal PBR should also be appropriate for enhancing the absorption of red light in a-Si:H cells.

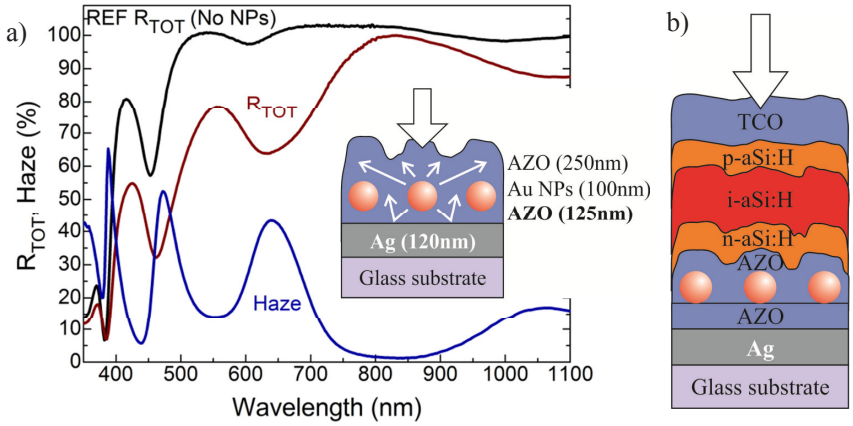

Fig. 6: (a) Total reflection (R_{TOT}) and Haze of a Plasmonic Back Reflector (PBR) composed of the structure depicted in the inset. For comparison, it is shown the R_{TOT} of a reference (REF) BR with the same structure but without MNPs. The PBR exhibits a pronounced Haze peak with maximum of 43.3% at 640nm, so it would be appropriate for implementation in an a-Si:H solar cell, as illustrated in (b).

The reflection and Haze of the PBR is particularly sensitive to the spacing between the MNPs and the Ag mirror; i.e. to the thickness of the AZO layer deposited below the particles [11,19]. Figure 7a shows the results of a PBR similar to that of Fig. 6a but with a bottom AZO layer having twice the thickness. In the a-Si:H spectral range of interest (500-800nm) the Haze is lower in this structure than in that of Fig. 6a, but it presents a higher and broader peak at longer wavelengths (750-950nm) which reaches a maximum of 49%.

The PBR of Fig. 7a would therefore be more appropriate to increase the generated photocurrent of μc-Si:H solar cells, or of double-junction cells as depicted in Fig. 7b, since such devices can exhibit an extended photocurrent response for infrared wavelengths up to ~1050nm.

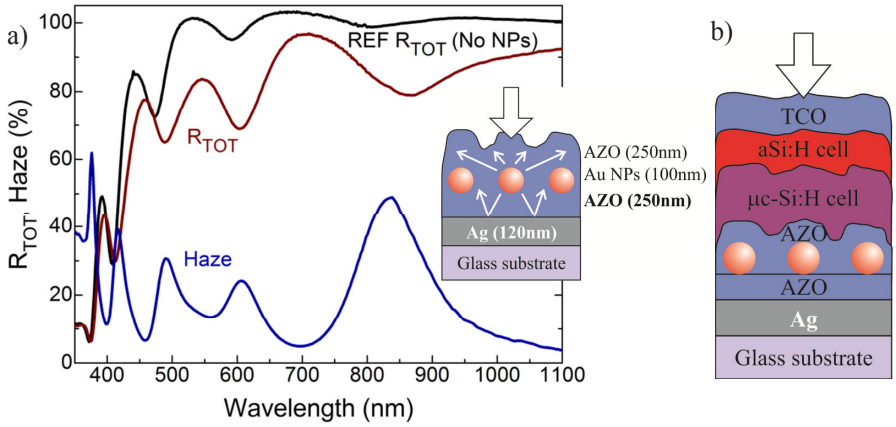

Fig. 7: (a) R_{TOT} and Haze of a PBR composed of the structure depicted in the inset. For comparison, it is shown the R_{TOT} of a reference (REF) BR with the same structure but without MNPs. The PBR exhibits a pronounced Haze peak with maximum of 49% at 835nm, so it would be appropriate for implementation in a μc-Si:H solar cell, or in a double-junction solar cell as shown in (b).

4.2. Superstrate configuration

The wet-coating process employed to pattern the colloidal MNPs is mostly performed at room temperature, apart from a step where the sample is heated at 120°C to bake the positively-charged molecular linkers that functionalize the surface. Since the process involves only low-temperature steps, it allows patterning the colloids onto previously-fabricated solar cells without damaging the quality of their PV materials. For instance, thin film hydrogenated Si cells can only withstand temperatures up to ~200°C, since higher temperatures damage the Si-H bonds. As such, it would not be possible to fabricate MNP structures as a post-process on TF Si cells using the conventional thin-film annealing method. As referred in section 1, TFA requires temperatures above 200°C to dewet the film and allow particle formation [4,10,13]. So, PBRs formed by TFA can only be implemented in cells having a substrate configuration (as those in section 4.1) where the cell material is grown on top of the

previously-fabricated PBR. However, commercial thin film solar cells are usually fabricated on glass substrates with a superstrate configuration, since the glass helps to protect the cell material from the external environment [1,7]. Therefore, the fabrication process of the TF cells that are currently in the market is not compatible with the conventional TFA technique, but it is compatible with the colloidal approach presented here.

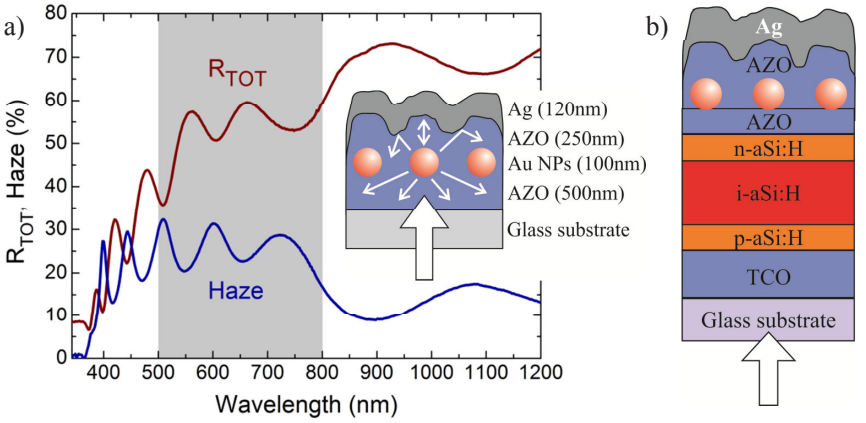

Fig. 8: (a) R_{TOT} and Haze of a PBR composed of the structure depicted in the inset. The PBR exhibits a Haze of 20-30% within the light trapping window of a-Si:H solar cells (marked by the grey area), so it would be appropriate for implementation in an a-Si:H cell having a superstrate architecture as shown in (b). In this configuration the Ag mirror is not flat as in the previous PBRs, it has a certain roughness induced by the MNPs shape.

Figure 8a shows the reflective properties of a PBR with the structure depicted in the inset, where the Ag mirror was inserted on top of the AZO layers with the embedded MNPs array. Such PBR is intended for TF cells with a superstrate configuration (see Fig. 8b), as those currently being commercialized. The Haze of the PBR oscillates between about 20 and 30%, in most of the wavelength range of interest for a-Si:H cells (marked by the grey area in Fig. 8a), and the R_{TOT} has an average value of ~55% in the same range. These values are not as high as those of the PBRs of Figs. 6 and 7, since the PBRs perform better with a flat Ag mirror layer instead of a rough mirror as that deposited over the MNPs+AZO layers. Nevertheless, the obtained Haze and R_{TOT} of this PBR should still allow considerable light trapping and, consequently, an improvement in the photocurrent generated by a-Si:H cells.

5. Conclusions and advantages of colloidal approach for PBRs

Solution-based self-assembly provides a simple, scalable and low-cost method for producing ensembles of nanoparticles in a controllable manner, in order to exploit their collective properties in functional devices. The fabrication and optical results obtained in this work open the way for a new research line in PBRs, using solution-processed colloidal MNPs. Colloidal-based PBRs are promising due to the following technological advantages:

1) The MNP deposition uses **low temperature** (<120ºC) methods, so the PBR can be incorporated as a post-process on top of already-fabricated solar cells, without degrading the cells' PV material.
2) Are compatible with **large-scale** processing, since the colloidal MNPs can be self-assembled in arbitrary large areas. So, the MNPs array can be patterned in an entire panel at once simply by immersing the panel in the necessary solutions.
3) It is a **low-cost** technique, since both the colloidal synthesis and patterning can be performed with standard chemical procedures using inexpensive compounds.
4) There is a high **control of the physical properties** (geometry, material) of the MNPs. Colloidal MNPs can be fabricated with remarkably monodisperse sizes and spherical shapes, and their material is purely crystalline [14]. In contrast, particles formed by the conventional TFA method usually have a broad range of sizes and shapes and their material can be polycrystalline [10].
5) It allows the **control of the inter-particle spacing**, since the distances between the deposited MNPs are determined by the particles' surface charge which can be tuned in several ways, as mentioned in section 3.
6) The colloidal deposition is **substrate-independent**; it only requires the functionalization of the surface with positively-charged molecules. As such, the MNPs can be patterned in any surface material, which enables the implementation of colloidal PBRs in virtually all type of thin film solar cell.

The analysis of the optical properties of the colloidal PBR structures presented here allows a preliminary assessment of their performance for implementation in solar cells. However, in an actual cell the absorbing photovoltaic material attached to the

PBR would influence its scattering properties, as commented in section 4.1. Such effect has not been taken into account in the present study, since it is only intended to provide a first-order evaluation of the produced PBRs for integration in substrate and superstrate solar cell architectures. The PBRs constructed for substrate cells show promising light scattering properties, with high Haze and total reflection in the wavelength ranges of interest for a-Si:H and μc-Si:H or double-junction TF Si cells. The PBR for superstrate cells exhibits worse performance mainly due to the roughness induced by the MNPs in the Ag mirror, but can still allow significant light absorption enhancement in a-Si:H devices. However, since this PBR is to be deposited on top of previously-fabricated cells, it enables important technological advantages. Mainly, it does not generate roughness in the cell PV layers, so it can provide effective light trapping on perfectly flat cells. It is well-known that roughness (as that formed by texturing the substrate) improves the light absorption of thin film cells, but it is detrimental to their electrical properties since it causes dislocations and defects in the cell material which decrease the open-circuit voltage and fill-factor relative to flat devices [7]. The novel colloidal approach presented here offers the key possibility of having an optically dense thin film cell with flat PV layers, which can exhibit enhanced optical and electrical performance relative to the conventional textured TF devices.

The reflective properties of the colloidal PBRs can be further improved by increasing the concentration of deposited MNPs. As mentioned in section 3, the surface coverage used in the analyzed PBRs (~5%) is about 3 times lower than the preferential one for full interaction between the incoming light and the particles' array. The authors are currently engaged in obtaining PBRs with such preferential MNP concentrations, which should provide pronouncedly higher light diffusion.

Acknowledgements

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