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Plasmonic degradation and the importance of over-coating metal nanoparticles for a plasmonic solar cell

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ABSTRACT

Plasmonics is an emerging area of study for photovoltaic applications. The last decade has seen an increasing interest in the application of metal nanoparticles (MNP) on the surface of solar cells to scatter light and subsequently achieve light trapping. In this study we investigate the optical effects of single and double layer rear light trapping reflectors – planar or scattering; direct or detached, which are fabricated on the 400 μm front-planar silicon solar cells. As a single layer reflecting scheme, we find that Ag metal nanoparticles (Ag MNP) as a scattering reflector outperforms all other single layer reflectors with a maximum current enhancement of 11% (calculated from 900 nm to 1200 nm) compared to metal back reflectors. However over time we notice that the scattering properties are dampened due to tarnishing of the silver nanoparticles. A double layer reflecting scheme using optimised Ag nanoparticles as the first layer and the second layer of evaporated Ag (E Ag) separated via MgF_2 is introduced to overcome this issue together with additional light trapping. We find that no *EQE* or current degradation is observed over time. More importantly, after optimising the over-coating MgF_2 thickness, the best-performing double layer reflector structure improves *EQE* by 4.5-fold at 1160 nm and enhances photocurrent by 25.6% (calculated from 900 nm to 1200 nm), compared to cells with metal back reflectors. We also conclude that the optimum thickness of the over-coating layer is dependent on the wavelength to be optimised, the angular distribution of the plasmonic nanoparticles scattering within the layer, and the type of adjacent metal reflector used. It performs best when condition for constructive interference or enhanced electric field at the rear Si interface is satisfied. An enhancement of effective optical path length factor *Z* of around 9-fold compared to an Al reflector is achieved by using the optimised double layer reflector, higher than the 6-fold enhancement reported previously.

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1. Introduction

Recent design studies of the highest performance silicon solar cells such as those conducted at UNSW have proposed that improvements in the rear optical properties are still warranted [1]. Possible issues include light absorption by the rear reflectors which accounts for around 5% of the light impacting on the rear of the cell and light escaping from the front surface after the first double pass across the cell. In the latter case, approximately 40% of the long wavelength reflected light from the back surface escapes after a double pass. One possible way to reduce rear parasitic absorption is by replacing the conventional Al metal by noble metals with better reflectivity (e.g. Cu and Ag [2,3]) if there is no compromise in the cell's rear electrical performance and can satisfy the work function requirements for a metal contact. Another method is by using detached reflectors with lower metal absorption. The losses due to light escaping the front can

be mitigated by implementing more efficient large angle scattering back reflectors. This is possible by maximising scattering beyond the escape cone of the semiconductor allowing total internal reflection and thereby increasing the optical pathlength in the solar cell. Commercial white paint, such as that containing rutile titanium dioxide dispersed in a polymeric binder, is one possible candidate for a scattering reflector [4,5]. Another scattering mechanism attracting attention is the use of localised surface plasmon excitations on metal nanoparticles which provides a tunable high scattering cross section to scatter light to larger angles without increasing the silicon surface area and associated losses [6].

2. Background

Last decade has seen a surge in the application of metal nanoparticles that support localised surface plasmons to achieve light trapping in solar cells [7]. The particles in this case could be embedded as in organic solar cells or deposited on the cells as in the case of solid state devices. The light trapping is achieved either

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due to near-field enhancement [8] or far-field effect like scattering [9,10]. Previous experimental and theoretical results on solar cells have verified the concepts of metal nanoparticles based light trapping in different types of solar cells by excitation of surface plasmons and the associated scattering effects. The theory and background is now well understood placing the field on a sound footing. In previous studies for thin film solar cells, the important designing factors for effective surface plasmon-enhanced light trapping have been reported, with a maximum current enhancement of 44% achieved by a “nanoparticle/magnesium fluoride/diffuse paint” back-surface reflector structure [11]. Research of metal nanoparticles incorporated in organic solar cells shows a 1.2% efficiency enhancement by using a dual metallic nanostructure [12]. In our previous studies for wafer based solar cells, we have experimentally validated different design considerations based on size/shape of the nanoparticles [13,14], the location of the nanoparticles – front and back of the solar cell [6,14]. These include effect of precursor metal mass thickness on the scattering property of nanoparticles and effect of dielectric spacer layer thickness on the optical and electrical properties of the cell. However, continued efforts are required to establish the practicality of this approach for wafer based solar cell application.

Gold (Au), copper (Cu) and silver (Ag) are well known plasmonic materials. The strong oxidation of Cu when exposed to air limits its application in plasmonic study. Gold is commonly used in microelectronic applications that require small area devices. However for PV applications which require large area processing, Au would be a very expensive option to be considered. Silver has been widely used as it is quite stable. Considering that only a very thin layer (tens of nm) is required, and the process we use for deposition followed by annealing is a solar cell production line compatible process, we do not expect a large increase in processing costs. Eventually the additional cost of incorporating plasmonics will be dictated by the fabrication method rather than the cost of silver.

Metal, however, when exposed to air can get tarnished easily causing the loss of its plasmonic properties. This could cause plasmonic degradation which can be a serious issue as the scattering of the metal nanoparticles weakens and therefore the light trapping properties are completely dampened as is evidenced in our results. This necessitates an improved double layer scattering reflector, with the dielectric over-coating film between the two reflecting-layer protecting metal nanoparticles and the second reflecting-layer inhibiting losses due to out-coupling of plasmons or light otherwise transmitted from the cell. The thickness of the over-coating film becomes critical considering the interference effect for light reflected/scattered from the two reflecting-layers. The over-coating film is also required to be thick enough to avoid degradation of nanoparticles over time as will be discussed in the study. Similar to the first reflecting-layer, the second reflector can either be planar or scattering; detached or direct, as will be thoroughly investigated and compared in this work.

In this paper we investigate the effect of metal nanoparticle scattering due to plasmonic effect as in a single reflector and in a double reflector configuration. The term reflector used in this study stands for either a scattering reflector (MNP or paint) or a specular reflector (metal mirror). It needs to be highlighted that this work involves 400 μm thick FZ Si solar cells that are not conventionally used in industry. However, they are used here due to the ease of fabrication and modification of the rear as is required for the experimental study. Thinner solar cells are likely to give a much better response due the light trapping effects; hence the results presented here show a lower limit to photocurrent enhancements. The motivation for this work is to check on the effectiveness and stability of the approach in comparison to other light trapping structures.

3. Experimental method

The cell structures investigated in this study were fabricated on the rear of planar-front 1.5 Ωcm 400 μm thick *p*-type FZ Si Passivated Emitter Rear Totally diffused (PERT) cells. The solar cell fabrication process is similar to that reported elsewhere [15,16]. The rear contact schemes of the solar cells in this study were modified to aid the different rear reflector depositions. The rear Al metal from the original structure was removed and replaced by Al grids with a surface coverage less than 15%. These Al grids act as rear electrodes for the cells and also allow subsequent reflector depositions. Thermally-grown SiO_2 is used to passivate the cells with modified rear reflectors. Solar cells used in each of the experiments have similar initial performance before reflector depositions with a current error range of 1–2 mA. The geometries of the different reflectors used in this study are shown in Fig. 1 (single layer reflectors) and Fig. 2 (double layer reflectors).

The single reflectors as shown in Fig. 1 are categorised into three general types: (1) planar reflectors are the conventional evaporated metal reflectors using an Al or Ag layer as a mirror (specular reflection) whereby the light is reflected at the same angle as incident light; (2) scattering or diffuse reflectors consist of Ag nanoparticles or commercial white paint. The Ag nanoparticles are fabricated by self-assembly technique of thermal evaporation of a thin Ag film followed by a N_2 atmosphere anneal at 200 $^\circ\text{C}$. A mass thickness of 28 nm is used as the precursor layer for the Ag nanoparticles. This mass thickness has been found to give the best results as reported in previous study [13]. The metal nanoparticles fabricated this way have broad extinction peaks due to a large distribution of different sizes and shape particles which favors broadband scattering. In the case of a white paint reflector, rutile titanium dioxide pigments (white knight) are directly brushed onto the rear of solar cells [4]. (3) The third type of single reflector uses a detached reflecting layer that is deposited on another substrate and placed on the cell so that it is optically coupled to the cell with air as buffer layer. It can either be a metal film or a layer having scattering properties. In this study with single reflectors two different thicknesses for the passivating SiO_2 are used: 55 nm and 96 nm. A bare cell comprising of Si/ SiO_2 is used as the reference for the single reflector case for performance comparison of the different structures shown in Fig. 1.

To further capitalise on the benefits brought about by the Ag MNP reflector, improved double layer reflectors are investigated as shown in Fig. 2. As mentioned in Section 1, the additional reflector is introduced to eliminate the plasmon-induced out coupling losses and light transmitted, such that light can be redirected back into Si. The second reflector is either detached or separated by a 500 nm MgF_2 film (this thickness is further optimised as discussed in Section 4.4) which acts as a caulking layer to eliminate shunting. The second reflector separated by MgF_2 film also prevents Ag MNP from plasmonic degradation which will be discussed in Section 4.3. The rear passivating SiO_2 thickness in this part of the study is 34 nm. The reference used for comparing different double layer reflectors is Si/ SiO_2 /Ag MNP. A conventional mirror reflector is not used as a reference here as it is intended to study the contribution from the second added layer. Moreover, both the rear passivating SiO_2 thickness and the capping MgF_2 layer thickness used in this study are not the optimised values. The MgF_2 thickness is optimised in Section 4.4. All enhancements in Section 4.4 are compared to the reference metal reflector case. A final rear structure with every element optimised is also reported.

For the optical characterisation, reflection and transmission spectra were measured using a Perkin Elmer spectrophotometer with an integrating sphere. Light absorption was then calculated by subtracting measured reflection and transmission from unity. For electrical characterisation, the external quantum efficiency

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