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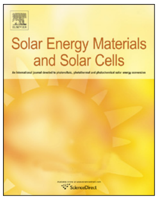

Pulsed laser deposition of refractive-index-graded broadband antireflection coatings for silicon solar cells

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ABSTRACT

This paper reports the design, fabrication and characterization of broadband antireflection coatings for silicon-based solar cells, using a graded-index technique and a novel dual-beam pulsed laser deposition method. A graded refractive index was realized by mixing two different optical materials, silicon and soda-lime glass, in a way that the composition of the mixture changes gradually from silicon (that of the solar cell) to glass (that of the outside protective glass) along the coating thickness direction following pre-designed refractive-index profiles. In this study, two different refractive-index profiles were fabricated, linear and Southwell profiles, on silicon substrates. In order to emulate a protective cover glass for solar-cells, coatings with an additional 400 nm-thick outer glass layer were also fabricated. The antireflection coatings showed reflectance of 2.2–5.5% (mostly around 4%) for a wavelength range of 400–1000 nm, and for the Southwell profile with a top glass layer, the reflectance was found to be between 2.2% and 4%, which is believed to be close to the theoretically best antireflection performance of a solar cell with a cover glass. Finite-difference time-domain simulations were also performed and the numerical results were in line with the measurement results. Because these coatings are based on silicon and glass and can be fabricated by the one-step pulsed laser deposition procedure, it can simplify the solar-cell structure considerably. The fabricated antireflection coatings may be also used for many other optical applications, such as thin-film optical attenuator and some optical sensors, and the presented pulsed laser deposition method is capable of fabricating a variety of high-performance optical coatings, which cannot be fabricated by other methods.

1. Introduction

It is a well-known fact that reflections will inevitably occur when light propagates across an interface between two materials with different refractive indices. In many fields, especially in photovoltaics, however, efficient light harvesting is very important [1], so the use of an antireflection (AR) coating is highly desirable. With an AR coating, the amount of light reflection can be considerably reduced, which improves the device performance [2,3]. For many years, the reduction of light reflection from the solar cell surface has always been one of the primary focuses of solar cell research [4], and a variety of low refractive index materials and fabrication methods have been developed to build high performance AR coatings.

One of the mostly employed approaches is the use of a graded index, which is generally realized using multilayer coatings [2,5] or special nanostructures [6–15] on the surface. Walheim *et al.* [16] used the phase separation of a macromolecular liquid to generate nanoporous polymer films with refractive index as low as 1.05 for multilayer films and reflectance was reduced to around 1% at some wavelengths. Leem et al. [5] demonstrated a single-material ZnS bi-layer AR coating based on porous/dense film structure using glancing angle deposition (GLAD), and the reflectance was reported to be ~5.8% in the wavelength range of 350–900 nm. Xi et al. [2] fabricated five layered graded-index films with TiO_2/SiO_2 nanorods by oblique-angle deposition. The minimum index was reported to be ~1.05 and lowest reflectance was only 0.1% at an incident angle of 30° for transverse-electric (TE) polarization at a certain wavelength. Many other nanostructures, such as nanocone, nano-sphere, nano-wire, porous structures, moth eyes structures and nano-imprinted textures, have been also used to achieve the reduction in reflectance instead of dense optical coatings [6–13,17]. Such a nanostructured surface layer leads to a density gradient and a refractive index gradient. Graded silicon surface layer, or black silicon (b-Si) [10,14,15], suppresses reflectance exponentially with the increase of the index-graded layer thickness. These nano-structures are generally fabricated by electron-beam lithography, interference lithography, etching,

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oblique-angle electron beam deposition and nano-imprint technique, which are complicated processes [17–20].

A majority of previously reported AR coatings were based on multilayer and/or nanostructure, and a single layer AR coating with a continuously-varying index gradient has not been reported in the literature. In this paper, we report the single-layer index-graded AR coating and its fabrication method, where the graded index is realized by controlling the volumetric composition of two different optical materials (silicon and soda-lime glass) along the coating thickness direction. AR coatings were deposited using a dual-beam pulsed laser deposition (PLD) method based on a 355 nm picosecond laser, with which a pre-designed composition profile of the coating is built by controlling laser pulses. In this deposition method, the picosecond laser beam is split into two beams and each beam irradiates on a different target material (silicon or soda-lime glass) and generates dissimilar plasmas, which are mixed uniformly in space and then deposited onto a silicon substrate. With this approach, the refractive index of the coating can be considered graded and varying gradually, and theoretically high-quality AR coatings can be fabricated.

In this study, two graded index profiles were considered: Southwell's profile [21] and linear profile. Southwell's profile is known as an extremely efficient index profile for AR coatings, close to the ideal profile, and the linear profile approximates Southwell's profile reasonably well. We also investigated the effect of an additional pure glass layer deposited on top of the two profiles because it can serve as a protective cover glass for silicon-based solar cells. In this way, therefore, an AR coating and a protective glass can be deposited in a single PLD procedure and can be completely integrated into a single unit on a silicon-based solar cell (Fig. 1c). The fabricated AR coatings showed reflectance of 2.2–5.5% for a wavelength range of 400–1000 nm, and for the Southwell profile with a top glass layer, the reflectance was found to be between 2.2% and 4%. Note that with the protective glass layer, the theoretical minimum reflectance is ~4%, which is very close to our results.

2. Design of graded index AR coatings

An AR coating with a graded refractive index can be realized by changing the composition of two different optical materials along the coating thickness direction. In order to design an index gradient profile, the refractive index of material mixture needs to be calculated as a function of mixture composition. Several mixture rules for refractive index are available in the literature [22], and in this study, the volume fraction method [16,22] was adopted, which is described as

$$n_{\text{mix}} = \phi_1 n_1 + \phi_2 n_2 \tag{1}$$

where n_1, n_2 and n_{mix} are the refractive indices of material 1, material 2, and their mixture, respectively; ϕ_1 and ϕ_2 are the volume fractions of material 1 and material 2.

In this research, silicon and soda-lime glass were chosen as two optical materials, and a 1 μm thick AR coating was planned on top of a silicon substrate. Note that SiO_2 is one of the well-known passivation materials, so we think the auto-oxidation interface of Si–SiO_2 (from Si and glass) can effectively reduce the surface state density. The composition changes from 100% silicon at the substrate surface to 100% glass at the coating outer surface gradually, and two different refractive index profiles were considered, Southwell and linear profiles, which are mathematically expressed as

$$\begin{aligned} n_{\text{Southwell}}(x) = n_{\text{Si}} - (n_{\text{Si}} - n_{\text{glass}}) \Bigg[10 \times \left(\frac{x}{1.0}\right)^3 - 15 \\ \times \left(\frac{x}{1.0}\right)^4 + 6 \times \left(\frac{x}{1.0}\right)^5 \Bigg] \end{aligned} \tag{2}$$

$$n_{\text{linear}}(x) = n_{\text{glass}} x + n_{\text{Si}}(1 - x) \tag{3}$$

where x is the coordinate measured from the substrate surface to the given location inside the AR coating and $0 \le x \le 1$ μm.

Here, a coating thickness of 1 μm is quite thick compared to typical AR layers for solar cells (< 100 nm). In this graded coating approach, the coating thickness must be determined considering the wavelength range that is targeted because longer wavelength light requires a thicker coating for effective AR performance.

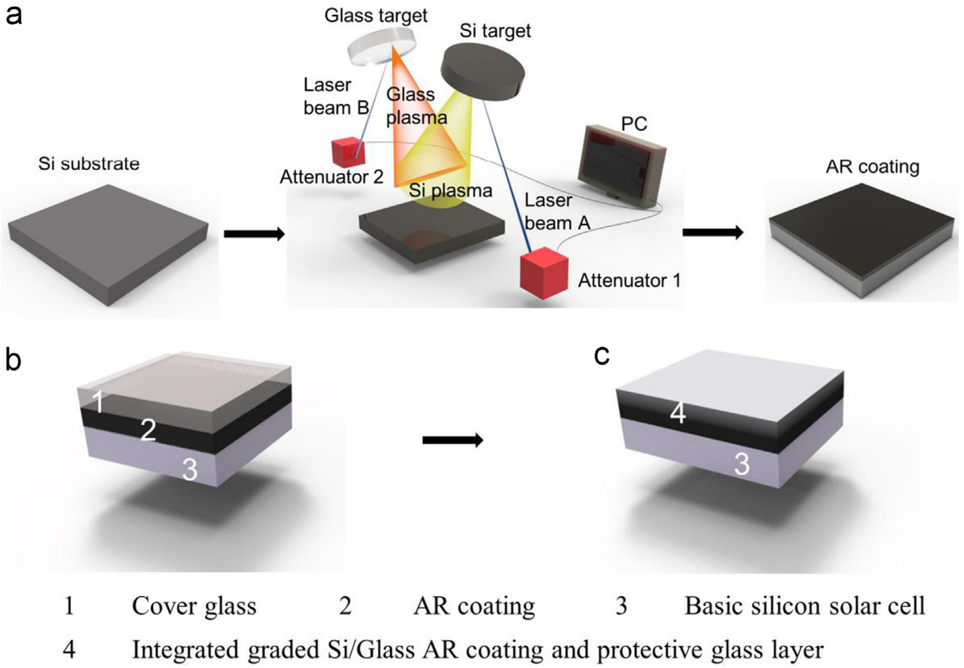

Fig. 1. Fabrication of a Si/glass graded AR coating by the dual-beam pulsed laser deposition. (a) Schematic drawing of the overall experimental procedure for fabricating a Si/glass graded AR coating. (b) A generic silicon-based solar cell structure. (c) Simplified silicon-based solar cell structure using the present graded AR coating.