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Nanostructured multilayer graded-index antireflection coating for Si solar cells with broadband and omnidirectional characteristics

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Design, fabrication, and characterization of a broadband, omnidirectional, graded-index antireflection (AR) coating made using nanostructured low-refractive-index ($n=1.05-1.40$) silica deposited by oblique-angle deposition are reported. Averaged over wavelength range from 400 to 1100 nm and $0°-90°$ angle of incidence, polished Si reflects ~37% of incident radiation. The reflection losses are reduced to only 5.9% by applying a three-layer graded-index AR coating to Si. Our experimental results are in excellent agreement with theoretical calculations. The AR coatings reported here can be optimized for photovoltaic cells made of any type of material. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3050463]

Despite the early discovery of the photovoltaic (PV) effect by Becquerel in 1839 and almost 125 years since the first solar cell was built in 1883 by Fritts, PV cells have seen limited commercial success until recently. The primary reason for this is the low efficiency and corresponding high cost per kW h of energy produced by PV cells. Even today, Si solar cells have efficiency at best just over 20% (Ref. 1) and it has not improved much over the past decade.[2] Reflection of incident light from the surface of the solar cell is one of the major optical loss mechanisms seriously affecting the solar cell efficiency. Figure 1 shows the broadband nature of solar irradiance spectrum. Nearly 90% of commercial solar cells are made of crystalline Si.[3,4] A polished Si surface reflects as much as ~37% light when averaged over all angles of incidence ($0°-90°$) and range of wavelengths of the solar spectrum that can be absorbed by Si (400–1100 nm).

For several years, the reduction in reflection from the surface of the solar cell has been one of the primary focuses of solar cell research.[5–10] Conventionally, a single layer antireflection (AR) coating with an optical thickness equal to one-quarter of the wavelength of interest is used. Ideally such single layer $\lambda/4$ AR coating should have refractive index (RI), $n_{\lambda/4}$, given by[11]

$$n_{\lambda/4}=\sqrt{n_{\text{semiconductor}}\times n_{\text{air}}}. \quad (1)$$

Often due to the unavailability of materials desired, exact value of the RI, the performance of such $\lambda/4$ AR coatings deviates from the optimum. For example, Si_3N_4, which has a RI value between that of Si and air, is used for Si solar cells. However, fundamentally these single layer AR coatings can minimize reflection only for one specific wavelength and for one specific angle of incidence (AOI), typically for normal incidence. The conventional Si_3N_4 single layer AR coating is inherently unable to cover the broad range of wavelengths present in a solar spectrum and the broad range of AOIs. These coatings reduce the reflection to ~18%. In order to reduce the reflection further, surface texturing is often used, which has shown to reduce the reflection to ~13%.[12,13] However, unlike any other method, the approach described in this paper offers unique advantages such as tunability of RI of the single material, flexibility in the choice of the material, simplicity of the physical vapor deposition process, and freedom of optimization for any substrate-ambient material system, all at once. As a proof of concept demonstration we report the fabrication of an optimized three-layer AR coating for Si solar cell.

Rayleigh, in 1880, mathematically demonstrated that graded-RI layers have broadband AR properties. However, until recently, due to the unavailability of optical materials with very-low RIs ($n<1.4$), such near-perfect graded-index AR coatings could not be realized. We have recently demonstrated a RI as low as 1.05 for SiO_2.[14,15] Other groups have also reported the theory and application of this technique to produce porous broadband AR coatings.[16] It is now possible to tune the RIs of an optical material to virtually any value between its bulk value and 1. Next, a systematic study of multilayer AR coatings is performed. For multilayer AR coatings, the RI of the layers is graded, i.e., gradually decreased, from the semiconductor to air. This can be done either in a continuous fashion or in discrete steps. Since the broadband solar spectrum is incident on the solar cell over a

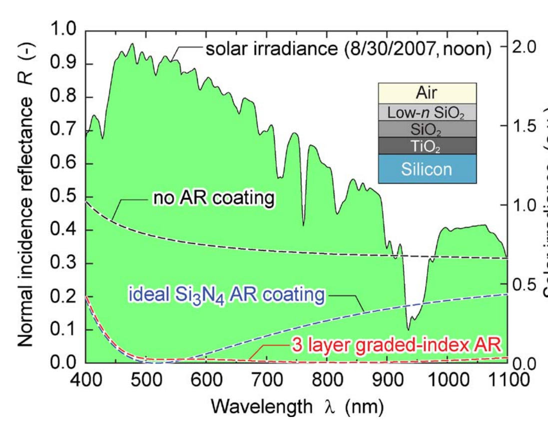

FIG. 1. (Color online) Solar spectrum and reflectance of Si substrate with (a) no AR, (b) $\lambda/4$ AR, and (c) three-layer AR coating.

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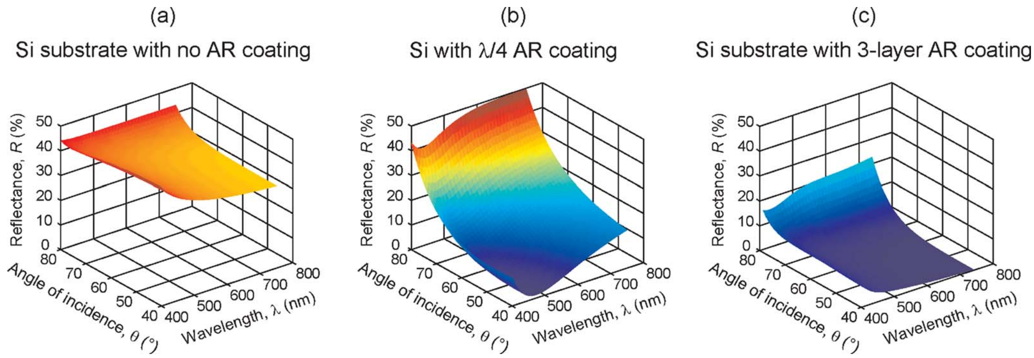

FIG. 2. (Color online) Calculated wavelength and angle resolved absolute reflectance of Si substrate with (a) no AR, (b) λ/4 AR, and (c) three-layer graded-index AR coating.

wide range of angles during the course of the day, it is important to use a figure of merit which gives a fair comparison of the performance of various AR coatings. We use R_{av} which is defined as follows:

$$R_{av} = \frac{1}{\Delta\lambda}\frac{1}{\Delta\theta}\int_{\lambda_{min}}^{\lambda_{max}}\int_{\theta_{min}}^{\theta_{max}} R\, d\theta\, d\lambda, \qquad (2)$$

where θ is the zenith angle.

All coatings are optimized in the wavelength range of 400–1100 nm and AOI range of 0°–90°. The details of the MATLAB optimization algorithm are discussed elsewhere.[17]

A three-layer AR coating, optimized for polished Si solar cells, is fabricated and characterized, as described in the next section. For simplicity of design and fabrication, only the thickness t of each layer in the three-layer graded index is varied during optimization while keeping the RI n fixed for each. Bulk TiO_2 is chosen as material for the first layer due to its high RI value, bulk SiO_2 for the second layer, and 80% porous SiO_2 for the third layer. This choice is made only for simplicity and is not due to the limitation of any kind. Figure 2 shows the surface plots of calculated reflectance for Si substrate (a) with no AR coating, (b) with a conventional λ/4 Si_3N_4 AR coating, and (c) with three-layer graded-index AR coating. The significant advantages of the three-layer graded-index AR coating in terms of drastically reduced reflectance over a wide range of wavelengths and AOI are elucidated by the plots.

The λ/4 Si_3N_4 AR coating in sample (b) is deposited using a reactive rf sputtering and is optimized for lowest normal incidence reflection at 550 nm wavelength and has a RI, $n=2.2$, measured at 550 nm and thickness, $t=62.5$ nm. The three-layer AR coating in sample (c) is deposited over a polished crystalline Si substrate using rf sputtering for the first two layers and oblique-angle e-beam evaporation for the third layer. The schematic of the AR coating is shown in Fig. 3(a). It is composed of the first layer of TiO_2 ($n=2.66$ at 550 nm), the second layer of SiO_2 ($n=1.47$ at 550 nm), and the third layer of low-n SiO_2 ($n=1.07$ at 550 nm). The thicknesses of each layer are 45, 120, and 200 nm, respectively. The first layer of TiO_2 is deposited by reactive sputtering using 200 W of rf power to the 2 in. TiO_2 target, 5.0 SCCM (SCCM denotes cubic centimeter per minute at STP) of Ar, and 0.5 SCCM of O_2 for 57 min at an operating pressure of 2 mTorr. A substrate bias of 5 W and a substrate temperature of 500 °C are used. The second layer of SiO_2 is also deposited by reactive sputtering using 200 W of rf power to the 2 in. SiO_2 target, 5.0 SCCM of Ar, and 0.5 SCCM of O_2 for 60 min at an operating pressure of 2 mTorr. A substrate bias of 5 W is used without any external heating. The third layer of porous SiO_2 is deposited using oblique-angle e-beam evaporation technique. The desired low RI is achieved by mounting the sample such that the substrate normal is at 85° to the incoming flux. Details of the oblique-angle evaporation technique have been discussed elsewhere.[18–20] The thickness and RI values are measured using ellipsometry. The thickness is confirmed using scanning electron microscopy (SEM). Figure 3(b) shows the SEM image of the three-layer AR coating.

The absolute reflectance of samples (a)–(c) is measured using the variable angle spectroscopic ellipsometry. For each sample, data are measured for wavelengths between 400 and 750 nm and for AOIs between 40° and 80° with 1° increments. This restricted choice of range is due to the limitation of the measurement equipment. However, it serves the purpose of comparison of measured and calculated values in the same range without any loss of accuracy. A large number of data points ensure accuracy in R_{av} obtained from the measurement. Figure 4 shows the surface plots of the measured reflectance data. Figure 4(c), in contrast with Fig. 4(b), shows very-low reflectance over a wide range of wavelengths and AOIs, clearly demonstrating the predicted broadband and omnidirectional characteristics of the three-layer graded-index AR coating.

The measured absolute reflectance results are in excellent agreement with the theoretically calculated values shown in Fig. 2. The resultant R_{av} for samples (a)–(c) are 37.0%, 17.3%, and 5.9%, respectively. Figure 5 shows the photograph of the three samples for side-by-side comparison. The λ/4 AR coating shows blue color due to its significantly high reflectance in the wavelength range below the zero-reflection wavelength of 550 nm. The superiority of the three-layer AR coating is clearly evident.

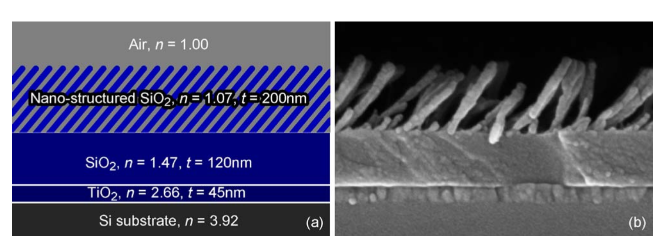

FIG. 3. (Color online) (a) Scanning electron micrograph and (b) schematic of three-layer graded-index AR coating. The RI values are measured at 550 nm.

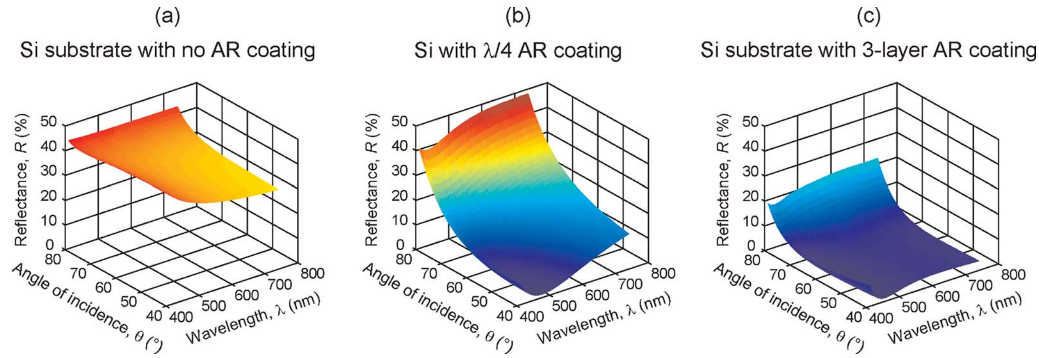

FIG. 4. (Color online) Measured wavelength and angle resolved absolute reflectance of Si substrate with (a) no AR, (b) λ/4 AR, and (c) three-layer graded-index AR coating.

In conclusion, we have demonstrated a broadband, omnidirectional, graded-index AR coating. The availability of the nanostructured low-*n* materials deposited by oblique-angle deposition technique has allowed the design of near-perfect AR coatings which can be used in a wide variety of applications. Generally porous films are mechanically not as stable as dense films. However, this was not a concern for our research related to optical properties of the thin film coatings. No disintegration, degradation of performance, or other adverse effect was observed even after several months. In specific cases, it may be practical to limit the maximum allowable porosity of the films to achieve desired mechanical properties. Additional means may be employed to protect the top layer, such as a thin pour closure or capping layer. Measurements show dramatic reduction in reflection over a wide range of AOI and a broad range of wavelengths in comparison with conventional λ/4 AR coatings. The average reflectance, R_{av}, of 5.9% was measured for the triple-layer graded-index AR coating as compared to 17.3% for the conventional Si_3N_4 λ/4 AR coating widely used for Si solar cells. These values are in excellent agreement with the theoretical calculations which predict R_{av} of 4.9% for the triple-layer graded-index AR coating and 18.2% for the λ/4 AR coating. This broadband and omnidirectional character of our AR coating is very well suited for application in solar cells.

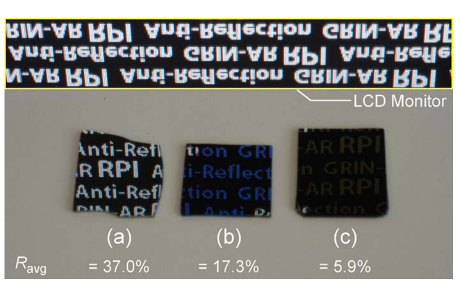

FIG. 5. (Color online) Photograph of Si substrate with (a) no, (b) λ/4, and (c) three-layer graded-index AR coating.

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