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Enhanced light-scattering properties of sputtered Ag/Al:Si bilayers deposited at a low substrate temperature – A potential back reflector for highly efficient flexible thin-film photovoltaic devices

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ABSTRACT

Nanotextured Ag/Al:Si back reflectors with enhanced light-scattering efficiency for flexible thin-film solar cells were developed at a low substrate temperature (T_s) by sophisticated control of the microstructural evolution of metal films. Development was based on a structure zone model and a bilayer concept involving a high reflectivity Ag top layer deposited onto a surface-textured Al:Si bottom layer. The texture evolution with a root-mean-square (σ_{rms}) surface roughness of 60.1 nm in the Al:Si bottom layer was successfully achieved at a low T_s of 100 °C by abnormal grain growth. The growth was induced by the low melting point of Al and the impurity drag effect on the grain boundaries caused by the Si dopant, whereas the σ_{rms} of the pure Ag film deposited at T_s=200 °C was 46.9 nm. After room temperature deposition of highly reflective Ag films onto the nanotextured Al:Si bottom layers, significant improvement in the light-scattering properties of the Ag/Al:Si bilayers was obtained compared to the Al:Si films without Ag films, for which the reflection significantly decreased due to the interband absorption and low reflectivity of Al. Under optimum conditions, the light-scattering efficiency of nanotextured Ag/Al:Si bilayers deposited at T_s=75 °C is comparable to that of reference Ag films at T_s=200 °C. When the nanotextured Ag/Al:Si bilayers deposited at T_s=75 °C as back reflectors were applied to flexible nc-Si:H thin-film solar cells, a high conversion efficiency of 8.6% was successfully achieved, and no detrimental reduction in cell performance was observed in comparison with the solar cell using the reference Ag back reflectors deposited at 200 °C.

1. Introduction

Hydrogenated silicon-based thin-film solar cells have been considered as alternatives to crystalline silicon solar cells to enable the low-cost generation of solar electricity over the past few decades because of the reduced consumption of raw materials and large-volume production capacity [1–3]. In particular, flexible Si thin-film solar cells using light-weight, unbreakable and inexpensive substrates have additional advantages, such as higher throughput using roll-to-roll processing, ease of handling and transportation, and robustness [4]. These thin-film solar cells also enable the esthetic and innovative design of solar products with various shapes and sizes [5]. Despite the strong possibility of becoming a next-generation photovoltaic technology, a challenge to the development of flexible Si thin-film solar cells is their relatively low conversion efficiency because of low light absorption. To overcome this limitation, a light trapping strategy to enhance the light absorption has been intensively studied [6–8]. When the interfaces at the front and the back of the thin absorber layers are ideally roughened to randomly scatter the incident light, the optical path length in the thin-film photovoltaic devices can be increased by a factor of $4n^2$ compared to the path length of devices with flat interfaces at the limit of weak absorption, where n is the refractive index of the absorber [9]. This $4n^2$ enhancement is known as the Lambertian limit. In flexible Si thin-film solar cells in the n-i-p configuration, the rough interfaces of the Si absorber layers are generally induced by the textured metal back reflectors; therefore, the light absorption in the solar cells is strongly dependent on the surface features of the back reflectors [10]. In addition, the excellent reflectance of the back reflectors should be used to effectively reflect the light that is unabsorbed by the device back into the Si active layers without the undesirable internal absorption by multiple reflections on the rough back reflector surfaces [11]. Although new schemes, including plasmonic scattering, diffraction gratings and nanorod arrays, have been

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proposed to enhance the light absorption [12–14], optimized randomly textured back reflectors are still being considered for use in commercial solar products because they can be manufactured in large sizes and they have sufficient light-trapping properties. Among these back reflectors, the textured Ag back reflector called 'hot silver' has been widely applied to flexible Si thin-film solar cells. As the name implies, the surface texturing of the Ag back reflectors can be achieved by abnormal grain growth of polycrystalline Ag films deposited at elevated temperatures [15,16]. However, for the roll-to-roll process, where the flexible substrate is quickly and continuously moving, a high temperature process is not suitable because the substrate cannot be heated to a sufficiently high temperature to induce the microstructural evolution of metal films deposited. The resultant heating zone would be long, adding complexity to the manufacturing equipment and high cost to the production. This high temperature requirement also limits the use of polymeric substrates with low heat resistance. Although a simple way to fabricate highly textured back reflectors at a low substrate temperature is obviously attractive for mass production, there are few reports on the low temperature processing of highly textured metal back reflectors until now.

In this paper, we developed an alternative metal back reflector with a highly textured surface and improved light-scattering properties. We produced the reflector at a low substrate temperature using a two-step method of direct-current (dc) magnetron sputtering. In this process, the highly textured Al:Si films were deposited onto the flexible substrates at substrate temperatures of 75 °C, followed by the room temperature deposition of highly reflective Ag film. The nanotextured Ag/Al:Si bilayers were applied to flexible Si thin-film solar cells, and the light-scattering efficiency improvement of the metal back reflector was evaluated.

2. Experimental details

Metal films were prepared on flexible stainless steel (SS) substrates (size: $50 \times 50\ mm^2$, thickness: 100 μm) using a dc-magnetron sputtering system equipped with 4-inch-diameter high-purity Al–1 wt% Si (Al:Si), Ag and Cr metal targets. The 30-nm-thick Cr film was applied as an adhesive layer between the SS substrate and the metal film. The base pressure was 6.7×10^{-5} Pa, and the flow rate of the argon was maintained by a mass flow controller (MFC) at 10 sccm, which corresponds to a working pressure of 1.3×10^{-1} Pa. The target to substrate distance and plasma power density were held constant at 100 mm and 1.3 W/cm^2, respectively. The film thickness was controlled by adjusting the sputtering period. The samples were heated in situ by a halogen heater, and the substrate temperature was measured using a thermocouple directly on the SS surface. Details of the experimental set-up and deposition parameters are listed in Table 1.

The morphology of the films was investigated by plan-view and cross-sectional scanning electron microscopy (SEM) using a HITACHI S-4800 electron microscope. The root-mean-square (σ_{rms}) surface roughness was measured by atomic force microscopy (AFM) using a PARK SYSTEM XE-100. The microstructure and elemental distribution of the films were examined by transmission electron microscopy (TEM) and energy-dispersive X-ray spectroscopy (EDX) using a FEI Tecnai G2 F30 S-Twin. The total and diffuse reflectance spectra (R_{total} and $R_{diffuse}$, respectively) of the samples were measured using a UV–vis–NIR spectrophotometer (SHIMAZU UV-3101PC, Japan) equipped with an integrated sphere. The resolved angle distribution of the scattered light on the film surface was evaluated using an angular distribution function measurement. A He–Ne laser (λ=633 nm) was used as a source of incident light. The intensity of scattered light was recorded by a detector, which moved around the samples at discrete angles in 1° increments.

Hydrogenated nanocrystalline silicon thin-film (nc-Si:H) solar cells in the n-i-p configuration were deposited onto the back reflectors using the plasma-enhanced chemical vapor deposition (PECVD) technique. The structure of the nc-Si:H thin-film solar cells was as follows: SS substrate/Cr (20 nm)/metal back reflector/ZnO:Al (100 nm)/n-nc-Si:H (30 nm)/i-nc-Si:H (1.0 μm)/p-nc-SiC:H (20 nm)/In_2O_3:Sn (80 nm)/Al grid. In the deposition of the intrinsic nc-Si:H layers, a mixture of silane (SiH_4) and hydrogen (H_2) gases was used. The intrinsic nc-Si:H layers were deposited using a 60 MHz very high frequency (VHF) glow discharge at a power density of 0.1 W/cm^2. The substrate temperature and chamber pressure for the deposition of the i layers were fixed at 150 °C and 40.0 Pa, respectively. The silane concentration ratio SC=[SiH_4]/([SiH_4]+[H_2]) was maintained at 6.5%. The p- and n-type silicon layers were deposited using a 13.5 MHz radio frequency (RF) discharge with the gases of SiH_4, H_2 and CH_4 (50% in H_2) along with B_2H_6 (1% in H_2) and PH_3 (1% in H_2), respectively. The 100-nm-thick ZnO:Al (2 wt% Al_2O_3) films acting as barriers to metal diffusion were deposited onto the metal back reflectors at room temperature using dc-magnetron sputtering. The 80-nm-thick ITO films and Al metal grids were deposited using rf-magnetron sputtering and thermal evaporation, respectively. A double source (class A) solar simulator (WACOM WXS-155S-L2, Japan) was used to measure the conversion efficiency of the solar cells under standard test conditions (25 °C, AM1.5G, 100 mW/cm^2), as well as the open circuit voltage (V_{oc}), the short-circuit current density (J_{sc}) and the fill factor (FF). The spectral response of the solar cells was measured with an EQE measuring system (SOMA S-9200, Japan) under a 0 V bias voltage and no light bias.

3. Results and discussion

Fig. 1 shows the plan-view SEM images of the 300-nm-thick Al:Si films deposited onto the SS substrates with varying substrate temperature (T_s) from 25 °C to 200 °C. The insets in Fig. 1 represent the cross-sectional microstructure of the corresponding films. The Al:Si film deposited without substrate heating (T_s=25 °C)

Table 1
Experimental set-up and deposition parameters of Al:Si and Ag films.

Parameter		Value			
Target size and composition		4 in., Al:Si (Al 99 wt%:Si 1 %wt) 4 in., Ag			
Target–substrate distance (mm)		100			
Base pressure (Pa)		$< 6.7 \times 10^{-5}$			
Working pressure (Pa)		1.3×10^{-1}			
Plasma power density (W/cm^2)		1.3			
Sputtering gas and flow rate (sccm)		Ar, 10			
Temperature	Heater temperature (°C)	25 ± 1	150 ± 5	200 ± 5	300 ± 5
	Substrate temperature (°C)	25 ± 1	75 ± 5	100 ± 5	200 ± 5