Thin Solid Films 545 (2013) 451–457

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Accurate characterization of thin films on rough surfaces by spectroscopic ellipsometry

S.C. Siah [a], B. Hoex [b,*], A.G. Aberle [b,c]

[a] Massachusetts Institute of Technology, Cambridge, MA 02139, USA
[b] Solar Energy Research Institute of Singapore, National University of Singapore, 117574, Singapore
[c] Department of Electrical and Computer Engineering, National University of Singapore, 117576, Singapore

ARTICLE INFO

Article history:
Received 14 September 2012
Received in revised form 20 July 2013
Accepted 24 July 2013
Available online 1 August 2013

Keywords:
Spectroscopic ellipsometry
Thin film properties
Rough surfaces
Silicon nitride
Antireflection coating
Depolarization

ABSTRACT

Spectroscopic ellipsometry (SE) is the technique of choice to determine material properties of thin films in a fast and non-destructive way. However, its ability to accurately extract the material properties of thin films deposited on rough substrate surfaces still remains a challenge, due to depolarization of specularly reflected light. In this paper we present a method based on SE to determine the properties of thin films on rough surfaces. It is shown that, by analyzing SE data only at discrete photon energies, information such as the film thickness and the refractive index at these photon energies can be extracted. The discrete photon energies selected in this method can be related to destructive interference of light at the thin-film surfaces, which significantly reduces the depolarization problem. The method is demonstrated using thin amorphous hydrogenated silicon nitride (a-SiN_x:H) films deposited on both polished and rough silicon wafer substrates. By varying the a-SiN_x:H film thickness, it is shown that the depolarization effect of light is strongly dependent on the incident photon energies due to light interference at the surface of the a-SiN_x:H film. Based on this observation, energy selective ellipsometry (ESE) is proposed as a technique for analyzing thin films on random rough surfaces. It is shown that a-SiN_x:H films with a wide range of thickness and optical properties on rough silicon surfaces can be studied accurately by means of ESE. This technique is expected to have useful practical applications, for example in photovoltaics where films are typically deposited onto textured substrates as a means of enhancing light trapping.

1. Introduction

Spectroscopic ellipsometry (SE) is a state-of-the-art characterization technique used to determine the optical properties and thickness of thin dielectric films in a simple, non-destructive and accurate manner. Most commonly the film-of-interest is deposited onto a polished substrate surface, to avoid measurement artifacts related to the substrate's surface roughness. In practice, however, thin films are often deposited onto rough substrate surfaces, for example crystalline silicon wafer solar cells which feature a textured surface to reduce optical reflection losses (and to improve light trapping in the wafer) [1]. Unfortunately, ellipsometry on rough substrates has generally proven to be very difficult. One specific case where SE on rough surfaces has been shown to work are amorphous hydrogenated silicon nitride (a-SiN_x:H) films deposited on monocrystalline silicon (mono-Si) wafers which are textured using an anisotropic etch resulting a pyramid textured surface. Due to the very regular surface orientation of this texture, it is possible to measure these samples accurately under an angle of 54.7° [2,3]. This is, however, not possible for more randomly textured silicon wafers such as anisotropically etched multicrystalline silicon (multi-Si) or as-cut wafers. The ability to obtain accurate film information on these rough surfaces would be beneficial for the photovoltaic community.

Various authors have reported about the challenges involving SE analysis of thin films on rough silicon surfaces. For example, Williams determined the degree of depolarization of reflected light waves by determining the Mueller matrix of rough surfaces [4]. However, the complete determination of the Mueller matrix is relatively time consuming and not suitable for industrial application. More recently, Saenger et al. have shown that accurate results can be obtained from *alkaline textured* silicon surfaces [2]. This can be achieved by modelling the effect of the roughened surface using the effective medium approach, whereby the film's optical properties are a mixture of the optical properties obtained from the polished reference wafer and a virtual fraction which depends on the wafer's surface morphology. While this approach is capable of describing the depolarization effect of the textured surface, it is purely empirical as the virtual void fraction is used as a fitting variable. It was reported that a-SiN_x:H film deposited onto an acid textured multi-Si wafer would have a refractive index of 1.4 (at 2 eV photon energy), which is significantly lower than the typical value of 2.0 achieved on a polished silicon substrate using the same

* Corresponding author. Tel.: +65 6516 6571; fax: +65 6775 1943.
E-mail address: bram.hoex@nus.edu.sg (B. Hoex).

0040-6090/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.tsf.2013.07.067

film deposition recipe. The lower refractive index was accounted for by mixing the optical properties of the reference film with air (voids) using the Bruggeman type effective medium approximation (EMA) approach during the model-based analysis [5]. However, the EMA approach is only applicable if the feature size of the surface roughness is significantly smaller than the wavelength of the light used in the measurements [6]. Hence, this could explain the unrealistically low refractive index values reported by Saenger et al. The EMA approach can effectively be used to model the surface roughness of a-SiN_x:H films on polished silicon substrates, which typically lies in the 1–5 nm range [7]. It is our understanding that the optical properties of a-SiN_x:H films deposited by plasma-enhanced chemical vapor deposition (PECVD) are not significantly influenced by the substrate's surface morphology, but instead are primarily determined by the PECVD process conditions.

In this paper, we introduce a method to analyze SE data obtained from rough surfaces. In this so-called energy selective ellipsometry (ESE) technique, we exploit the fact that the measurement artifacts of SE measurements on rough surfaces are strongly wavelength dependent. By analyzing the raw ellipsometric spectra only for selected energy intervals that correspond to destructive interference of reflected light, the optical properties and thickness of the thin film on the rough substrate can be extracted accurately. Using PECVD a-SiN_x:H films deposited on as-cut and acid-textured multicrystalline silicon (multi-Si) wafers as examples, we show that accurate information on the measured films can be obtained by understanding the interaction of light with the textured substrates.

2. Experimental details

The a-SiN_x:H films in this study were deposited using a commercial inline remote PECVD reactor (SiNA®-XS from Roth & Rau AG). The precursor gases for a-SiN_x:H depositions were ammonia (NH_3) and silane (SiH_4) and the depositions were performed at a substrate temperature of 450 °C, chamber pressure of 22 Pa and plasma power of 2200 W. The total gas flow rate was maintained at 345 sccm and the other deposition conditions for the samples are summarized in Table 1. The substrates used for this work were (i) double side polished float-zone grown *n*-type monocrystalline silicon (mono-Si) wafers and (ii) as-cut as well as acid textured multi-Si wafers. The as-cut wafers were mechanically textured by wire sawing and the textured wafers were etched anisotropically by acid texturing in a $HF/HNO_3/H_2SO_4$ solution for 2–3 min at a bath temperature of 8 °C. The thickness and refractive index non-uniformity of the silicon nitride films on as-cut wafers over 3 separate runs were within 2.34% and 0.55%, respectively.

For samples 1 to 7, the NH_3/SiH_4 ratio was varied in order to obtain a-SiN_x:H films with an increasing Si content. Samples 8 and 9 were prepared using identical process conditions, apart from the carrier speed. The thickness of the resulting a-SiN_x:H film scales inversely with the carrier speed, hence the a-SiN_x:H film of sample 9 is expected to be twice as thick as that of sample 8. Similarly, samples 9 and 10 were prepared on acid textured multi-Si wafers using identical deposition conditions, except for the carrier speed. In addition, reference samples were also prepared simultaneously for samples 8 to 11 on polished mono-Si substrates so that the results obtained for the rough samples can be compared.

All samples were studied using SE. In SE, the two spectroscopic angles psi (ψ) and delta (Δ), which express the change in the polarization state of the light after interaction with a sample, are measured. A common representation of ψ and Δ is

$$\tan(\psi)e^{i\Delta} = \frac{r_p}{r_s}, \quad (1)$$

where r_p and r_s are the complex reflection coefficients for light waves polarized parallel and perpendicular to the plane of incidence, respectively [6,8].

In our experiment, the (ψ,Δ) spectra were recorded over a spectral range of 1.5 to 5.0 eV at an angle of incidence of 75° using a rotating polarizer-fixed analyzer ellipsometer (Sopra, model Ges5 SE). As the measured SE (ψ,Δ) spectra do not directly represent the film's optical properties, these were obtained from a model based analysis in which the (ψ,Δ) spectra were calculated from an optical model and the mismatch between the modeled and measured (ψ,Δ) spectra was subsequently minimized. The ellipsometric data were fitted to a three layer model consisting of air, an a-SiN_x:H film, and a c-Si substrate. The dielectric functions of the a-SiN_x:H film are parameterized using the Tauc–Lorentz formalism after Jellison et al. [7]. A surface roughness layer between air and a-SiN_x:H has only been modeled for the reference samples using EMA [7]. In addition, the SE measurements were performed using 3 different illumination spot sizes, as summarized in Table 2.

For comparison, the as-cut and textured multi-Si wafers were also measured by single-wavelength ellipsometry (SWE, Sentech SE400) at an angle of incidence of 75° at 632.8 nm and with an illumination spot size diameter of 1 mm. SWE can determine the optical properties of thin films at one single wavelength, typically at 632.8 nm (~2 eV) using a HeNe laser [9]. While SWE provides a fast and easy way of characterizing thin films, it only provides information on the material properties at a single wavelength. Secondary electron microscopy (SEM, Carl Zeiss, model Auriga) was used to characterize the surface morphology of the as-cut and acid textured silicon wafers and two samples were investigated by transmission electron microscopy (TEM, JEOL, model 2010 F) to confirm the film thickness obtained from ellipsometry. The accelerating voltages used for SEM and TEM imaging are 5 kV and 200 kV respectively.

Table 1
Summary of deposition conditions.

Sample	Substrate type	Surface condition	NH_3/SIH_4 gas flow ratio	Carrier transport speed [cm/s]	Remarks
1	Mono-Si	Polished	6.0	70	
2	Mono-Si	Polished	5.0	70	
3	Mono-Si	Polished	4.0	70	Standard recipe
4	Mono-Si	Polished	3.0	70	
5	Mono-Si	Polished	2.0	70	
6	Mono-Si	Polished	1.5	70	
7	Mono-Si	Polished	1.25	70	
8	Multi-Si	As-cut	4.0	70	
12	Mono-Si	Polished	4.0	70	Reference for s8
9	Multi-Si	As-cut	4.0	35	
13	Mono-Si	Polished	4.0	35	Reference for s9
10	Multi-Si	Textured	4.0	80	
14	Mono-Si	Polished	4.0	80	Reference for s10
11	Multi-Si	Textured	4.0	40	
15	Mono-Si	Polished	4.0	40	Reference for s11

Table 2
Details of the three different illumination modes.

Illumination mode	Dimensions
Parallel beam	3 mm × 12 mm
Micro-spot	365 μm × 470 μm
Ultra micro-spot	120 μm × 60 μm