Thin Solid Films 545 (2013) 245–250

Contents lists available at ScienceDirect

Thin Solid Films

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

Radiofrequency power effects on the optical and structural properties of hydrogenated silicon films prepared by radiofrequency magnetron sputtering

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ARTICLE INFO

Article history:
Received 14 April 2013
Received in revised form 4 August 2013
Accepted 9 August 2013
Available online 17 August 2013

Keywords:
Amorphous material
Non-crystalline materials
Nanostructures
Thin films

ABSTRACT

The optical and structural properties of hydrogenated silicon films, deposited by radiofrequency (rf) magnetron sputtering at low temperature (Ts = 100 °C), were carefully investigated by means of optical transmission measurements (OT), Fourier transform infrared (IR) spectroscopy and spectroscopic ellipsometry (SE) technique. By varying the rf-power from 100 W to 350 W, and keeping all other parameters of the plasma constant, the growth rate increases up to 1.02 nm/s. A remarkable change in the hydrogen-bonding configurations for both wagging and stretching vibration modes was observed. The observed changes demonstrate definite structural transformation of the films from a completely amorphous phase to another one with crystalline Si when the rf-power is increased from 180 W to 200 W. The difference between the two phases is well revealed by the OT and the IR absorption results, and strongly confirmed by the SE ones. The effect of hydrogen on the band gap and on the microstructure of the films is discussed.

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1. Introduction

In the last decade, hydrogenated nanocrystalline silicon (nc-Si:H) has attracted considerable attention for its promising application to high efficiency and stable solar cells [1–3] in comparison with hydrogenated amorphous silicon (a-Si:H) [4,5]. This material exhibits different complex morphology consisting of a multilayered structure along the growth direction. This structure is heterogeneous and is formed of an amorphous matrix (a-Si:H) in which nano-sized crystallites are embedded [6,7]. In addition, there are voids and amorphous silicon formed with high density islands separated by low density tissues.

It is well known that the performance of optoelectronic devices depends on the mixed "amorphous/crystalline" phase and on the content of the hydrogen and how it is bonded. The microstructure of the material is directly related to the growth mechanism and to the deposition techniques. In fact, many efforts have been directed towards developing various approaches for crystallization of a-Si:H films as by excimer laser annealing [8,9] or furnace annealing using high annealing temperatures which reach 600 °C [10,11]. However, the high temperatures affect the content of hydrogen and are not recommended and not compatible with the use of desired substrates such as glass or plastic. Several deposition techniques have been used to prepare nc-Si:H thin films, including plasma-enhanced chemical vapor deposition, and hot wire chemical vapor deposition [12,13]. However, the radiofrequency (rf) magnetron sputtering proves for the deposition as another choice, and very little is known about the microstructure and the growth processes.

In our previous studies [14,15], we have reported on the effects of different plasma parameters on the optical and structural properties of nc-Si:H films deposited by this last technique. In particular, we have demonstrated that it is possible to obtain homogeneous films, well crystallized at relatively low growth temperature (Ts = 100 °C), for a gas mixture of 30% of argon and 70% of hydrogen, when the total pressure in the deposition chamber is equal to 3 Pa. The results presented in this paper extend these works. We report on the effects of the rf-power on the deposition rate and on the optical and the microstructure of the films.

2. Experimental details

The samples studied were thin films deposited by rf sputtering of high-purity crystalline silicon target of 7 cm in diameter, and using an argon (30%) and hydrogen (70%) gas mixture. The target–substrate holder distance was fixed at 7 cm for all the films. The substrates were ultrasonically cleaned in three successive baths of trichloroethylene, acetone and propanol before loading into the deposition chamber. The chamber pressure was regulated at 3 Pa and the substrate temperature was fixed at Ts = 100 °C. The applied rf-power was varied from 100 W to 180 W for the series A samples, and from 200 W to 350 W for the series B samples (see Table 1).

Infrared (IR) transmission measurements were performed on nc-Si:H films deposited on roughened crystalline silicon substrates in the

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http://dx.doi.org/10.1016/j.tsf.2013.08.053

Table 1
Hydrogen content, C_H (at.%), film thickness, d, static refractive index, n_0, average energy, E_m, optical, E_T, gaps, and dispersion energy, E_d, for samples prepared at rf powers varying from 100 to 350 W. The errors are estimated, and only the maximum value of the error is indicated for each particular parameter.

Samples	rf (W)	C_H (at.%)	d (nm)	n_0	E_m (eV)	E_T (eV)	E_d (eV)
Series A							
E1	100	11.0	725 ± 3	3.56 ± 0.02	3.43 ± 0.02	1.75 ± 0.02	40 ± 2
E2	150	12.4	842 ± 3	3.47 ± 0.02	3.52 ± 0.02	1.81 ± 0.02	39 ± 2
E3	180	13.5	637 ± 3	3.38 ± 0.02	3.55 ± 0.02	1.85 ± 0.02	37 ± 2
Series B							
E4	200	15.5	624 ± 3	3.29 ± 0.02	3.66 ± 0.02	1.92 ± 0.02	36 ± 2
E5	250	16.8	732 ± 3	3.41 ± 0.02	3.67 ± 0.02	1.93 ± 0.02	39 ± 2
E6	280	16.4	571 ± 3	3.54 ± 0.02	3.65 ± 0.02	1.91 ± 0.02	42 ± 2
E7	300	17.5	662 ± 3	3.06 ± 0.02	3.68 ± 0.02	1.93 ± 0.02	31 ± 2
E8	350	18.2	720 ± 3	2.93 ± 0.02	3.67 ± 0.02	1.90 ± 0.02	28 ± 2

frequency range 400 cm^{-1}–2400 cm^{-1}, to determine the hydrogen bonding configurations and the total bonded hydrogen content CH (at.%).

The thicknesses d, the optical gap E_T, the average gap E_m, the static refractive index n_0, as well as the dispersion energy E_d were determined from standard optical transmission (OT) measurements performed on the films deposited simultaneously on Corning 7059 glass substrates. These measurements were recorded on a Shimadzu (UV 3600) double beam spectrophotometer over the wavelength range 0.5 μm–2.5 μm.

In order to estimate imaginary part ε_2 of the complex dielectric function, a parameter that determines the structure of a film in terms of its ε_2 peak position and the maximum peak intensity, the samples were investigated using UV–visible spectroscopic ellipsometry (SE) measurements within the 1.5 eV–5 eV photon energy range.

3. Results

3.1. Deposition kinetics

The low deposition rate (r_d ~ 0.1 nm/s) constitutes a serious drawback in applications when thick films are desired. Therefore, an increase of r_d in nc-Si:H film preparation with a high crystalline volume fraction is required. In this study, the increase of r_d up to 1.02 nm/s has been reached by operating the discharge in the high rf-power regime. The values of r_d (obtained from the ratio of the film thickness to the deposition time) are presented in Fig. 1 as a function of the rf input power. One can notice that with the plasma conditions, r_d increases linearly from 0.16 nm/s to 0.94 nm/s with increasing rf-power from 100 W to 280 W. This behavior can be explained by the fact that in the high rf-power regime, the fluxes of the Ar^+ sputtering ions that reach the silicon target and the sputtered Si atoms increase. The discharge becomes resistive and the rf energy is more efficiently coupled to the plasma through the bulk electrons which gain energy from the electric field produced in the inter-electrode space. As a consequence, more radicals are formed in the discharge and reach the grounded electrode with a high stick coefficient.

In the highest rf-power range (>280 W), the r_d appears to increase more slowly up to 1.02 nm/s. This effect can be explained by the possibility that all species are excited at these powers, and so the addition of further rf-power cannot increase the gas utilization and the flux of the Ar^+ sputtering ions.

3.2. OT analysis

The envelope method, first applied by R. Swanepoel [16] to homogeneous films of a-Si:H, is used here to evaluate very accurately the refractive index n, the thicknesses d, and the absorption coefficient a of the films. This method is based only on the transmission spectra when the optical thicknesses are sufficient to generate several interference extremes. The refractive index n and the quantity $x = e^{-\alpha d}$ are determined from the envelopes and the thickness d from the interference fringes. The refractive index values obtained for the energies lower than the optical gap were then fitted by the Wemple–DiDomenico one-oscillator model [17]. According to this model, the dispersion relation of the refractive index can be described, to a very good approximation, by the following formula:

$$n^2(\hbar\omega) = 1 + \frac{E_d\ E_m}{E_m^2 - (\hbar\omega)^2} \tag{1}$$

where E_d denotes the dispersion energy related to the coordination number of the atoms, and E_m defines the average gap usually considered as the energy separation between the centers of both the conduction and the valence bands. Plotting $[n^2(\hbar\omega) - 1]^{-1}$ against $(\hbar\omega)^2$ allows one to determine these parameters by fitting a straight line to the points. The static refractive index values n_0 (which are usually related to the material density, to the optical gap or to both of them) are then deduced from the following relation:

$$n_0^2(0) = 1 + \frac{E_d}{E_m}. \tag{2}$$

The deduced values of these parameters are listed in Table 1.

The optical band gap E_T values have been determined according to the non-direct transition model proposed by Tauc [18], as the intersection of the straight line through the high energy points of a graph of $(\alpha\,\hbar\omega)^{1/2}$ versus $\hbar\omega$, with the energy axis:

$$(\alpha\hbar\omega)^{1/2} = K(\hbar\omega - E_T) \tag{3}$$

where K denotes the so-called Tauc slope. The E_T values deduced from this analysis are also indicated in Table 1 for completeness. It is clearly seen that E_T and E_m values follow the same trend for both the two series A and B. They increase for the series A, and remain constant, within experimental errors, for the series B while n_0 is varied. In order to correctly interpret these results, we use the information from IR hydrogen wagging and stretching vibration modes.

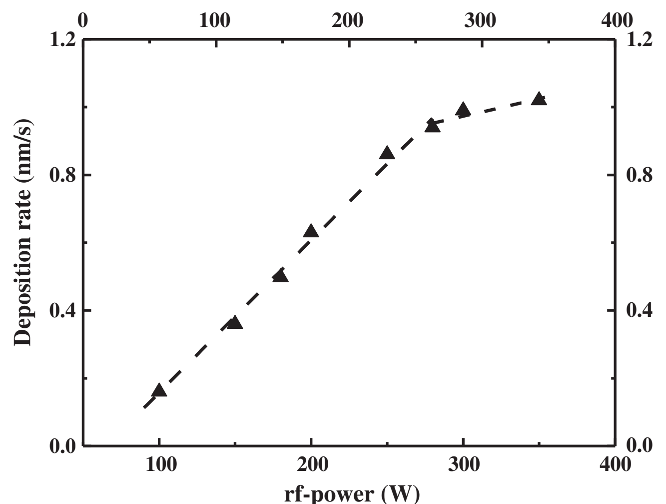

Fig. 1. Effect of the rf-power on the deposition rate r_d obtained from the ratio of the film thickness to the deposition time. The deposition rate increases linearly with increasing rf-power from 100 W to 280 W, and more slowly up to 1.02 nm/s in the highest rf-power range (>280 W).

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