Substrate temperature dependence of the roughness evolution of HWCVD a-Si:H studied by real-time spectroscopic ellipsometry

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Abstract

The surface roughness of HWCVD deposited a-Si:H films has been monitored as a function of the film thickness for substrate temperatures between 70 and 450 °C using real-time spectroscopic ellipsometry. Information on the microstructural evolution of the a-Si:H films has been deduced from the data and different growth phases in this microstructural evolution are discussed in terms of the underlying surface processes such as nucleation and initial growth, surface smoothening and roughening processes, and surface diffusion. From the data, it is concluded that, for the specific conditions studied, the best material properties are obtained at ~250–350 °C.

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1. Introduction

Fundamental and technologically relevant information on the growth process of a-Si:H by hot wire chemical vapor deposition (HWCVD) can be obtained from real-time spectroscopic ellipsometry. This information can, for example, be obtained by studying the surface roughness as a function of bulk thickness, as done by Collins et al. for rf plasma enhanced chemical vapor deposition (PECVD) [1,2] and Levi et al. for HWCVD [3]. Here we present a first extensive investigation of the roughness evolution of HWCVD deposited a-Si:H including a detailed study of the substrate temperature dependence. The data has been analyzed by the same procedure as in the aforementioned work [1–3] giving information about the microstructural evolution of the a-Si:H and the corresponding surface processes. These include nucleation processes and initial growth, surface roughening and smoothening, and surface diffusion mechanisms [4,5]. In these processes, the substrate temperature plays generally a major role.

The HWCVD growth conditions have been chosen within the region of “optimum growth conditions” as reported in an extensive study by Molenbroek et al. [6] such that Si radicals produced at the filament react with SiH4 before reaching the substrate while the abundant formation of reactive higher silane radicals is avoided. Film growth is therefore expected to be dominated by SiH3 radicals produced by reactions between SiH4 and H atoms generated at the filament. It can therefore be assumed that the data have a generic validity for HWCVD growth under similar conditions [10].

2. Experiment

The a-Si:H films have been deposited in an ultrahigh vacuum setup (Fig. 1) dedicated to fundamental studies on a-Si:H using radical beams and advanced in situ and real-time film diagnostics [7]. The main features of the setup are a low-contaminant background, an accurately controlled substrate temperature, and a good accessibility of the substrate for “all-optical” diagnostic techniques.

For the present study, the hot-wire source has been operated at a filament current of 11 A (filament temperature is 2050 ± 200 °C) in undiluted SiH4. The SiH4 flow rate has been set at 3 standard cm3/min and the pressure has been controlled at 8 mTorr within the deposition chamber. With a hot-wire to substrate distance of 6.8 cm, the conditions are...
within the region of “optimum operating conditions” [6] as mentioned earlier. The resulting a-Si:H deposition rates range from 78 to 102 Å/min. The depositions have been carried out on crystalline Si (c-Si) substrates covered with native oxide while varying the substrate temperature $T_s$ between 70 and 450 °C. Before deposition, the substrates have been baked overnight at a temperature of 450 °C.

The deposition processes have been monitored by real-time spectroscopic ellipsometry using a Woollam M2000U ellipsometer (wavelength range 250–1700 nm). The optical properties of the films have been deduced from the ellipsometry data obtained at the end of the deposition (film thickness $\sim 2000$ Å) using an optical model and applying the Tauc–Lorentz formalism [8,9]. The optical model used is a simple two-layer model (bulk a-Si:H layer/surface roughness layer) with the common Bruggeman effective medium approximation of 50% voids and 50% bulk a-Si:H for the surface roughness layer. The optical film properties (i.e., dielectric function) have been assumed constant during film growth as corroborated by the fact that similar optical properties are obtained at different thicknesses.

3. Results and discussion

3.1. Surface roughness and a-Si:H microstructural evolution

Fig. 2 shows the variation of the surface roughness layer thickness $d_s$ as a function of the bulk layer thickness $d_b$ for several a-Si:H films deposited at 250 °C. The films have all been deposited under equal deposition conditions and Fig. 2 reveals therefore that the data reproduce well. The roughness evolution observed is in good agreement with the data reported by Collins and co-workers for a-Si:H deposited by rf PECVD at a similar deposition rate [1,2]. Using the interpretation of the data by Collins and co-workers as a starting point, several growth phases can be distinguished in the microstructural evolution of the a-Si:H:

I. The initial roughness ($\sim 13$ Å) at the onset of bulk growth reflects the final height of the nuclei that form on the substrate just before coalescence (see also II.) occurs. These nuclei result from the heterogeneous nucleation of a-Si:H on the native-oxide-covered c-Si which causes Volmer–Weber type of growth reflected by a relatively high surface roughness. The nuclei are generally represented by hemisphere-like structures and at coalescence, when the nuclei contact, their maximum radius is in the order of the maximum height reached. A nucleation density of $\sim 15 \times 10^{12}$ cm$^{-2}$ can therefore be calculated from the data in Fig. 2.

II. The decrease of $d_s$ in the first $\sim 150$ Å of bulk growth reflects the reduction of the heterogeneous-nucleation-
induced surface roughness after the initial nuclei have coalesced and when the surface is fully covered by a-Si:H. At this point surface diffusion can smoothen out the roughness existing at a relatively short lateral length scale provided that the surface diffusion process is active with a sufficiently large surface diffusion length. Following the analysis used by Collins and Yang [11], a fit of the data results in a surface diffusion length of $\sim 26$ Å, which is about equal to the distance between the initial nuclei. The smoothening effect leads to a minimum roughness of $d_s = 2$ Å at a thickness $d_b = 150$ Å.

III. After reaching a bulk thickness of $\sim 160$ Å, $d_s$ increases again. At this so-called “amorphous-to-amorphous (a-Si→a-Si) roughening transition” [1, 2], the surface roughness becomes most likely dominated by roughness features stretching out over relatively large lateral length scales [12]. These roughness features evolve since initial film growth and cannot be smoothened out by the aforementioned surface diffusion process which is only effective at relatively short lateral length scales. Analyzing the data in terms of $d_s \propto (d_b - d_{b,0})^\beta$ with $\beta$ the growth exponent [4], results in a $\beta > 0.5$ indicating unstable growth with non-self-affine scaling. When starting from the very smooth a-Si:H surface ($d_s = \sim 2$ Å) a $\beta$ of $\sim 0.5$ can be explained by “random growth” but this growth mode holds only for the first new monolayer [4]. The evolution of roughness in this growth phase is therefore due to another physical mechanism such as, e.g., addressed by Raible et al. for amorphous film growth [13].

IV. At a certain film thickness ($\sim 500$ Å) the strong increase in surface roughness levels off. Although we do not have a conclusive explanation for this transition, it appears that a mechanism sets in when a certain $d_s$ or $d_b$ value has been reached such that the strong roughening behavior is attenuated. This could be a “smoothening” mechanism which is active on long length-scales and manifesting itself similar to the surface relaxation mechanism as proposed by Edwards and Wilkinson [4, 10]. Also the $\beta$ values of $0.10–0.12$ that can be deduced for this post-initial ($d_b > 1000$ Å) growth regime correspond to the onset of such a surface relaxation mechanism as addressed by Smets et al. [5].

3.2. Substrate temperature dependence of the a-Si:H microstructural evolution

In Fig. 3 the variation of $d_s$ as a function of $d_b$ is given for the a-Si:H films deposited at the different substrate temperatures $T_s$ while Fig. 4 shows the real and imaginary part of the corresponding dielectric functions. The global trend in the data is roughly similar to one for 250 °C and several conclusions adding to the insight into the microstructural evolution of HWCVD deposited a-Si:H films can be drawn:

- The initial roughness at the onset of bulk growth is very similar for the different $T_s$ values. This suggests that the nucleation process is controlled by the substrate itself and not by the substrate temperature. Most likely is that nucleation takes place at defect sites at the native-oxide-covered c-Si with the nucleation density ruled by the density of the defect sites.
- The initial decrease of $d_s$ is less pronounced or even absent for $T_s$ values of 150 and 70 °C. This can be explained by the fact that the surface diffusion process, which is thermally activated, is not sufficiently active at these temperatures to smoothen out the nucleation-induced surface roughness. For $T_s \geq 250$ °C the decrease in $d_s$ is initially very similar suggesting that the surface diffusion length is sufficiently large to smoothen out the roughness at these temperatures. On the other hand, the
minimum roughness reached for 350 and 450 °C is not as low as for \( T_s = 250 \) °C while the minimum roughness value is also reached at a lower value of \( d_b \). This effect is certainly significant for \( T_s = 450 \) °C for which the smoothening is less effective as for 250 and 350 °C.

• The a-Si\( \rightarrow \)a-Si roughening transition is observed for all \( T_s \) while for the a-Si:H films deposited at 350 and 450 °C it takes place earlier than for the film deposited at 250 °C. This suggests that for \( T_s > 250 \) °C the surface roughness becomes earlier dominated by the roughness features stretching out over relatively large lateral length scales \[14\] which cannot be smoothened out by the surface diffusion process that acts only at short lateral length scales. The films deposited at \( T_s \geq 250 \) °C all show a severe roughening at the a-Si\( \rightarrow \)a-Si roughening transition with relatively similar \( \beta \) values.

• The strong increase in surface roughness levels off for all \( T_s \) values when a bulk thickness value of \( d_b = 500 \) Å is reached. The “smoothening” mechanism that slows the roughening down is therefore eventually present at all substrate temperatures although it is more pronounced at higher \( T_s \). This is also reflected by the \( \beta \) values as is evident for the post-initial growth regime in Fig. 3: \( \beta \) decreases from \( \sim 0.18 \) at 70 °C to \( \sim 0.05 \) at 350 and 450 °C. Furthermore, the surface roughness for \( d_b = 2000 \) Å is very similar for \( T_s \geq 250 \) °C while the roughness is much higher for 150 and 70 °C. AFM scans (with scan areas up to \( 3 \times 3 \) μm\(^2\)) have revealed similar rms roughness values with a same trend when going to \( T_s \) values of 70 and 150 °C \[10\].

Finally, we want to note that Collins and co-workers have found a correlation between a high electronic performance and stability of the a-Si:H and a large initial decrease in \( d_b \). This effect is certainly significant for \( T_s = 450 \) °C for which the smoothening is less effective as for 250 and 350 °C.

4. Conclusion

Insight into the microstructural evolution of HWCVD deposited a-Si:H has been obtained by monitoring the surface roughness as a function of the bulk layer thickness and substrate temperature by real time spectroscopic ellipsometry. The underlying surface processes and in particular the presence of thermally activated surface diffusion processes has been discussed.

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References

[9] Also the Cody–Lorentz formalism has been applied in the optical model but this did not reveal significant differences in the fit or parameters deduced compared to the Tauc–Lorentz formalism.