New ultrahigh vacuum setup and advanced diagnostic techniques for studying a-Si:H film growth by radical beams

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New ultrahigh vacuum setup and advanced diagnostic techniques for studying a-Si:H film growth by radical beams

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ABSTRACT

A new ultrahigh vacuum setup is presented which is designed for studying the surface science aspects of a-Si:H film growth using various advanced optical diagnostic techniques. The setup is equipped with plasma and radical sources which produce well-defined radicals beams such that the a-Si:H deposition process can be mimicked. In this paper the initial experiments with respect to deposition of a-Si:H using a hot wire source and etching of a-Si:H by atomic hydrogen are presented. These processes are monitored by real time spectroscopic ellipsometry and the etch yield of Si by atomic hydrogen is quantified to be 0.005±0.002 Si atoms per incoming H atom.

INTRODUCTION

Over the last two decades, the growth mechanism of a-Si:H has widely been studied, even to such an extent that it has become a model system for understanding thin film growth [1,2]. Meanwhile, the investigations have become more advanced and sophisticated although most of the experiments have been carried out under regular, but sometimes rather well-defined operating conditions for a-Si:H deposition [3]. However, to reach the next level of understanding, *i.e.* insight into the surface reactions at the atomic scale, surface science-like studies under very well-defined and idealized conditions will be required. Such studies can for example be carried out by mimicking a-Si:H film growth using several well-defined radical beams as, *e.g.*, done for Si plasma etching [4] and deposition of hydrogenated amorphous carbon [5]. In this paper, we present a new experimental setup for beam studies of a-Si:H film growth. In the design of this setup, three basic criteria have been taken into account:

1) The substrate, onto which the film is deposited, is the key factor within the setup and should therefore be easily accessible by (optical) diagnostics and the use of different substrate types, materials, and sizes should be possible. The substrate should also remain clean for a sufficiently long time setting strict demands on the level of vacuum reached in the setup.

2) With well-defined radical (and ion) beams it should be possible to mimic film growth under regular deposition conditions and the radical beams should preferentially be mono-radical and accurately quantified in terms of flux. For a-Si:H, radical beams of SiH_3 and H are most appropriate.

3) The growth process needs to be investigated by advanced *in situ* diagnostics with a clear preference for "all-optical" techniques which are able to monitor film growth in real time. Furthermore, the techniques need to be innovative such that key questions of a-Si:H film growth (*e.g.*, "What is exact role of surface Si dangling bonds?") can be addressed.

In this paper, a setup named GALAPAGOS, which meets these requirements, is presented and some initial experiments to characterize the setup are reported.

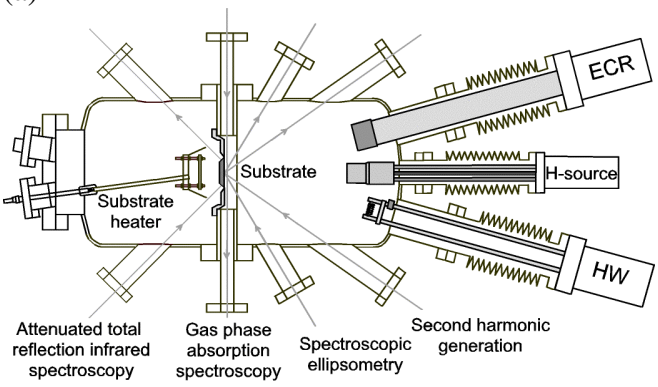
EXPERIMENTAL SETUP

The setup, schematically illustrated in Fig. 1(a), consists of two chambers with in between a flange which holds the substrate. Both chambers are separately pumped by turbo pumps and the setup is completely ultrahigh vacuum (UHV) compatible. Using only infrared heating lamps, a pressure of 10^{-8}–10^{-9} Torr is reached however a pressure of 10^{-10} Torr should be reachable after extensive backing. The substrate can be heated radiatively from the backside by a Boralectric^{TM} heater (from RT to 800 ºC) and the temperature is actively controlled.

For the moment, the setup is equipped with three radical sources. A thermal atomic H source, based on a heated capillary, is present to produce a well-defined H flux [6]. An electron-cyclotron-resonance (ECR) plasma source (TECTRA GmbH, Germany) can be operated in "atom-mode", "low-energy ion mode", and in "hybrid mode" [7] but is currently under development to produce a SiH_3 radical beam. Finally, a hot wire of tungsten is present to deposit a-Si:H films at reasonable deposition rate (compared to SiH_3 beam) while, when operated on H_2, it can also be used for *in situ* chamber cleaning. All sources are positioned on a linear shift such that the source-substrate distance can be varied. The gases H_2, D_2, SiH_4, SiD_4, Si_2H_6, and Ar can be introduced into the sources or chamber after passing a gas purifier.

As illustrated in Fig. 1(a), the setup has multiple ports for optical access. Spectroscopic ellipsometry can be applied from the front side (70º port) to monitor film growth in real time in terms of bulk thickness, surface roughness and optical film properties [8]. Infrared spectroscopy can be applied from the backside (45º port) using an attenuated total reflection (ATR) crystal for high sensitivity. This technique can be applied for H-depth profiling and to determine the H surface coverage of a-Si:H [3,9]. The same optical port will also be used in future studies in which dangling bonds will be probed at subgap energies by applying the evanescent-wave cavity ringdown technique [10,11]. Dangling bonds will also be studied from the front side of the substrate (35º port) using the surface-specific technique of second harmonic generation (SHG) [12]. Furthermore, gas phase absorption spectroscopy can be applied in front of the substrate and a residual gas analyzer can be used for thermal-programmed desorption studies.

(a)



(b)

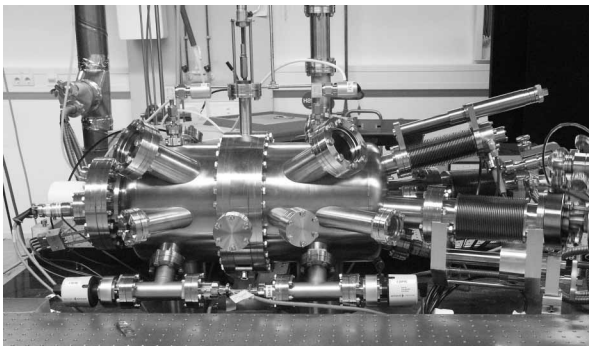


Figure 1. (a) Schematic top view of the UHV setup GALAPAGOS. On the right side the different plasma and radicals sources (ECR: electron cyclotron resonance plasma source; H-source: thermal atomic H source; HW: hot wire source) are shown. Some diagnostics that can be applied using the different optical ports are indicated. Not shown are: the residual gas analyzer the substrate shutter, and the infrared heating lamps. (b) Picture showing side view of the setup.

RESULTS

The first experiments that have been carried out with the GALAPAGOS setup are the deposition of a-Si:H films by the hot wire source and the etching of a-Si:H films by atomic H. The atomic H is generated by operating the three sources on H_2. These experiments have been carried out by using c-Si as a substrate and the processes have been monitored by real time spectroscopic ellipsometry RTSE (Woollam M2000U, 250-1700 nm). The deposition experiments give insight into film properties obtained as well as the deposition rate and the surface roughness evolution of the films. The etching experiments give insight into the source operation and can be used to compare the sources in terms of H flux produced. Using the calibration data available for the atomic H source, the etch yield of a-Si:H can be determined.

Deposition

The a-Si:H films have been deposited by the hot wire source from pure SiH_4 and at a substrate temperature of 250 ºC. The filament current has been set at 11 A (filament temperature is 2325±200 K), the SiH_4 flow at 3 sccm, and the pressure in the deposition chamber has been controlled at 8 mTorr. This has resulted in a deposition rate of 78 Å/min. The optical properties and the film thickness have been deduced from the RTSE data obtained at the end of the deposition (film thickness ~2000 Å) using an optical model and applying the Tauc-Lorentz formalism [13]. The optical model is a simple two layer model (bulk a-Si:H/surface roughness layer) with the common Bruggeman effective medium approximation of 50% voids and 50% bulk a-Si:H for the surface roughness. Following the work of Collins and co-workers [8], we have assumed the optical film properties constant during film growth (corroborated by the fact that similar optical properties are obtained at different thicknesses) and the variation of the bulk layer thickness d_b and surface layer thickness d_s has been deduced. Figure 2 shows the variation of d_s with d_b as well as the film's dielectric function and the Tauc-Lorentz parameters obtained.

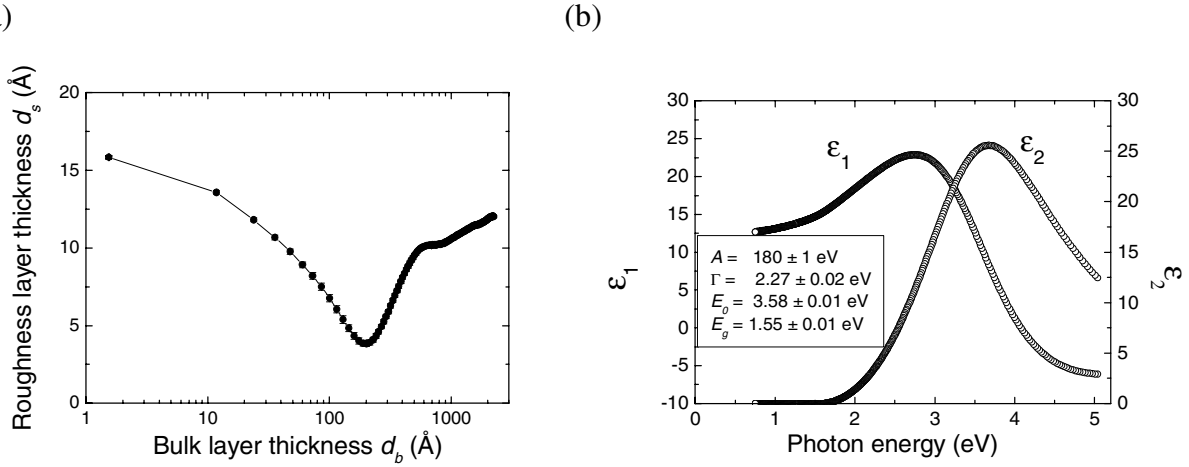


Figure 2. (a) Surface roughness layer thickness vs. bulk layer thickness from RTSE collected during the deposition of an a-Si:H film at a substrate temperature of 250 ºC. (b) Corresponding dielectric functions obtained *in situ* at 250 ºC after deposition of the a-Si:H film. The parameters of the Tauc-Lorentz fit are indicated in the figure.

Table I: Properties of the a-Si:H films deposited at 250 ºC. The analysis techniques are presented in the third column (FTIR = Fourier transform infrared spectroscopy; ERD = elastic recoil detection; RBS = Rutherford Backscattering; AFM = atomic force microscopy)

Refractive index n (2 eV)	4.3	RTSE
Extinction coefficient k (2 eV)	0.177	RTSE
Tauc band gap E_{Tauc}	1.67 eV	RTSE
Microstructure parameter R*	0.06	FTIR
H-content [H]	10.7 at. %	FTIR
H-content [H]	11.3 at. %	ERD
Si atomic density n_{Si}	5.1×10^{22} cm^{-3}	ERD/RBS
O atomic density n_O	2.2×10^{16} cm^{-3}	ERD
Surface roughness w_{rms}	11 Å	AFM

The dielectric function and the Tauc-Lorentz parameters are in good agreement with those reported for a-Si:H in the literature [8,13]. Detailed comparison of the data with those reported in Ref. [14] shows that the broadening parameter Γ matches best with those for very thin a-Si:H films obtained by H_2 diluted SiH_4 in an rf plasma. Therefore we have verified that our films are purely amorphous using Raman spectroscopy. The evolution of the surface roughness d_s with d_b shows also good agreement with the data reported by Collins and co-workers for a-Si:H deposited by an rf plasma at a similar deposition rate [8,14]. In the first 200 Å, d_s decreases due to coalescence of the nucleation-induced microstructure and this smoothening effect leads to a minimum roughness d_s = 4 Å at a thickness d_b = ~200 Å. Subsequently, d_s increases again due to the amorphous-to-amorphous (a → a) roughening transition [8]. The position of this a → a transition is in good agreement with the data reported for the rf plasma while our initial decrease in roughness Δd_s is bigger. The final roughness at a film thickness of 2000 Å is somewhat smaller and shows good agreement with the *rms* surface roughness deduced from AFM data (see Table I). Furthermore, for a film thickness d_b > 700 Å, the roughness can be analyzed in terms of the dynamic scaling coefficient β (i.e., $d_s \sim d_b^{\beta}$). This analysis leads to a β = 0.16 [15].

Table I summarizes the film properties obtained from the ellipsometry data as well as information on the H content and bonding, film atomic densities, and the surface roughness. The H content is typical for a film deposited at 250 ºC and the two values obtained by the two diagnostics show good agreement. The Si atomic density of the film is fairly high and the oxygen contamination is very low [16]. The latter can probably be attributed to the relatively low base pressure (10^{-9} Torr) during the deposition experiments and the use of gas purifiers.

It can therefore be concluded that a-Si:H with good (structural) film properties can be obtained by the hot wire source present in the UHV setup and that the film properties and growth process is similar to those reported in the literature.

Etching

Films of a-Si:H deposited under the aforementioned conditions, have been etched at a substrate temperature of 85 ºC by operating the three sources on pure H_2. Under this condition, the sources produce atomic H. From the etch rates obtained, the three sources can be compared to each other in terms of H flux produced.

First the atomic H source has been used to etch a-Si:H. The source has been operated at a H_2 flow of 0.43 sccm (pressure is 7×10^{-3} mTorr) and the temperature of the hot capillary has

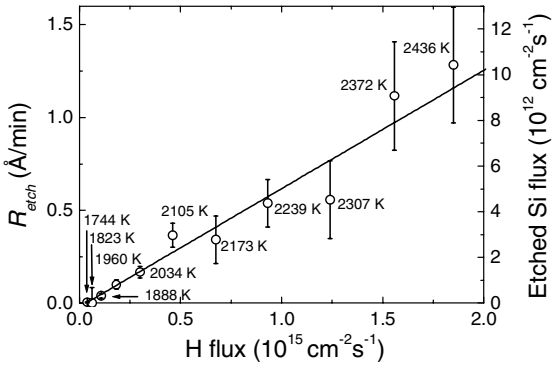


Figure 3. Etch rate R_{etch} as a function of the H flux produced by the atomic H source. The H flux, varied by varying the capillary temperature, has been calibrated by the manufacturer of the source. On the right axis the etched Si flux is displayed.

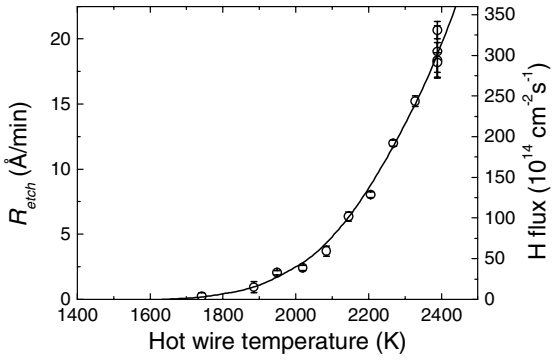


Figure 4. Etch rate R_{etch} as a function of the hot wire temperature (pressure is 120 mTorr, distance hot wire–substrate is 6.8 cm). On the right axis the H flux is displayed as deduced from the atomic H source data shown in Fig. 3.

been varied (see Fig. 3). The distance between source and substrate was set at 15.4 cm. Because the atomic H flux at the substrate position is known from a calibration of the source by the manufacturer [6], the etch rate has been plotted in Fig. 3 as a function of the H flux. For the analysis of the RTSE data of the etching experiments, a three-layer model had to be assumed. Compared to the deposition experiment, an additional layer below the surface roughness layer is present to account for H in-diffusion into the a-Si:H film [17]. This leads to an H interaction layer which saturates at a thickness of 115 Å (at 85 ºC) when reaching steady state etching conditions. When applying this model, a linear relation between etch rate and atomic H flux is observed in Fig. 3. This relationship can be used to quantify the atomic H flux from the other two sources, but it can also be used to determine the etch yield of H. The etch rate can be converted into "etched Si flux" when using the information on the Si atomic density of the film (Fig. 3). Subsequently, from the slope in Fig. 3 we can deduce an etch yield of 0.005±0.002 Si atom per incoming H atom. The accuracy of this etch yield is mainly determined by the systematic uncertainty in the calibration data of the source [6].

For the ECR plasma source and hot wire source, similar etching experiments have been carried out. A typical result is given in Fig. 4 for the hot wire source. Using the relation between etch rate and atomic H flux as shown in Fig. 3 (for other data sets we have evidence that R_{etch} is linear in H flux for a H flux up to ~10^{16} cm^{-2}s^{-1}), we can deduce the atomic H flux produced by the hot wire source at the position of the substrate. In Table II, the range of atomic H flux that can be obtained by the three sources (by varying the operating conditions and the distance between source and substrate, etc.) is summarized.

Table II. Ranges of H fluxes that can be obtained by the three sources when operated on H_2.

	Range H flux (cm^{-2} s^{-1})	**Variable operating conditions**
Atomic H source	$10^{12} – 10^{16}$	H_2 flow, capillary temperature, pressure, distance
Hot wire radical source	$10^{13} – 3\times10^{16}$	H_2 flow, hot wire temperature, pressure, distance
ECR plasma source	$10^{13} – 10^{15}$	H_2 flow, microwave power, pressure, distance

CONCLUSION

A new setup has been presented for well-defined beam studies to reveal the surface science aspects of a-Si:H film growth. Initial experiments with respect to deposition of a-Si:H and H-induced etching of a-Si:H have been carried out to characterize the setup. Next, experiments will be carried out by advanced diagnostics such as second harmonic generation, attenuated total reflection infrared spectroscopy, and evanescent-wave cavity ringdown spectroscopy, etc.

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