On the surface roughness development of hydrogenated amorphous silicon deposited at low growth rates

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The surface roughness evolution of hydrogenated amorphous silicon (*a*-Si:H) films has been studied using *in situ* spectroscopic ellipsometry for a temperature range of 150–400 °C. The effect of external rf substrate biasing on the coalescence phase is discussed and a removal/densification of a hydrogen-rich layer is suggested to explain the observed roughness development in this phase. After coalescence we observe two distinct phases in the roughness evolution and highlight trends which are incompatible with the idea of dominant surface diffusion. Alternative, nonlocal mechanisms such as the re-emission effect are discussed, which can partly explain the observed incompatibilities. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3179151]

The kinetic roughening of thin film growth follows from a competition between roughening and smoothening mechanisms. Thus from a study of the surface roughness evolution versus deposition time, insight into growth mechanisms, and their influence on structural properties of films can be gained. Consequently, there is a strong technological motivation to understand the origin of surface roughness and morphology.

Hydrogenated amorphous silicon (*a*-Si:H) develops extraordinarily smooth surfaces under optimum growth conditions, which is an indication for the presence of strong smoothening mechanisms during film growth. Surface smoothening is usually attributed to surface diffusion of growth precursor molecules[1] and is assumed to be a dominant surface mechanism for mass transport in different *a*-Si:H deposition techniques such as plasma-enhanced chemical vapor deposition (PE-CVD)[1] or hot-wire CVD (HW-CVD).[2] The driving force for surface diffusion of radicals toward surface valleys is described by a chemical potential proportional to the curvature of surface features. This leads to a diffusion away from surface hills (negative curvature) and toward surface valleys (positive curvature).[3] Diffusion lengths around 50–100 Å are typically assumed to explain the surface morphology obtained for *a*-Si:H thin films.[1]

However, regularly experimental and modeling results are published, which conflict with the idea of dominant surface diffusion of physisorbed SiH$_3$ radicals. Ceriotti and Bernasconi[4] have utilized *ab initio* calculations to investigate surface diffusion of SiH$_3$ radicals on fully hydrogenated H:Si (100) surfaces and obtained maximum diffusion lengths in the order of only a few lattice spacings at temperatures ranging from 300 to 1000 K for a fully hydrogenated surface, due to quick desorption of physisorbed SiH$_3$ radicals. Consequently, SiH$_3$ surface diffusion would not be able to explain the development of surface features in the nanometer range, as is observed for *a*-Si:H thin film growth with atomic force microscopy (AFM) (e.g., Sperling and Abelson[5]). Cheng *et al.*[6] concluded from experimental low-pressure CVD (LPCVD) studies utilizing a special cavity that surface diffusion does not play a significant role for step coverage. Smets *et al.*[7] obtained a rather high activation energy for surface smoothening of around 1 eV for *a*-Si:H film growth from solid-on-solid modeling, which conflicts with the low activation energy of SiH$_3$ radicals on a hydrogenated surface.

In this letter, we will present *in situ* real-time spectroscopic ellipsometry (RTSE) results of *a*-Si:H thin films grown with the ETP-CVD technique at growth rates of about 1 Å/s for substrate temperatures ranging from 150 to 400 °C. We will address the evolution of the surface roughness to gain insight into mass transport mechanisms that underlie our experimental observations and show that our results cannot be explained by a simple surface-diffusion dominated growth model of physisorbed SiH$_3$ radicals. Additionally, we study the effect of ion bombardment via external rf substrate biasing.

The *a*-Si:H thin films have been deposited on *c*-Si wafers (prime wafer, 500–550 μm) with ~2 nm of native oxide, as determined by spectroscopic ellipsometry (SE). Our RTSE measurements were performed using a J. A. Woollam Co., Inc. M-2000F rotating compensator spectroscopic ellipsometer. In our RTSE data analysis, we follow a standard procedure for RTSE data analysis is described in more detail by Van den Oever *et al.*[8] Koh *et al.*[9] have shown that the roughness obtained from SE measurements is linearly related to the rms roughness obtained from AFM measurements over a range of bulk film thicknesses up to at least 6500 Å, demonstrating that SE is a viable method for surface roughness analysis for the film thickness range utilized in this study. rf substrate biasing was generated with a rf generator (Coaxial RFGS 100 SE); the applied rf power was 60 W, leading to an average dc substrate voltage of 21 V. There was a delay between start of the deposition and activation of the substrate biasing of ~5 s, equivalent to approximately 5 Å of film growth.

The development of the surface roughness layer, d_s, versus the bulk film thickness, d_b, for depositions *without* substrate biasing can be seen in [Fig. 1(a)], and for depositions *with* substrate biasing in [Fig. 1(b)], in both cases for a tem-

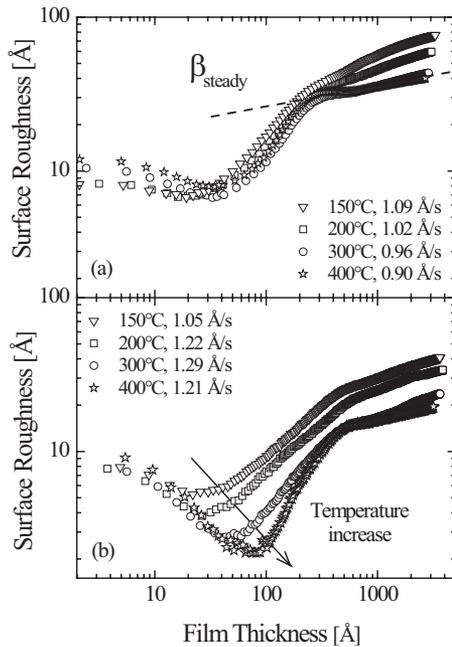[a]Electronic mail: m.a.wank@dimes.tudelft.nl.

FIG. 1. Surface roughness layer thickness development as a function of bulk film thickness for depositions (a) without substrate biasing and (b) with substrate biasing.

FIG. 2. Growth exponent β_{steady} determined from the roughness development in the steady growth phase shown in Fig. 1.

perature range of 150–400 °C. Substrate biasing leads to an increase in deposition rate of about 15%–20% [see legends in Figs. 1(a) and 1(b)]. It was concluded by Hoefnagels *et al.*[10] that this increase is caused by the production of additional SiH_3 radicals.

For all depositions, with and without biasing, we can identify several phases. Phase I: A phase in which the roughness decreases until a minimal roughness is reached—commonly this initial roughness is attributed to nucleation on the oxidized silicon wafer surface.[11] At the onset of film growth, coalescence of neighboring nuclei occurs and smoothening mechanisms result in considerable mass transport into the surface valleys between adjacent nuclei, leading to the roughness reduction. For unbiased deposition in [Fig. 1(a)], the lack of temperature dependence of the negative slope suggests that surface processes are virtually temperature independent or have extremely low activation energy. Also for biased depositions in [Fig. 1(b)] the negative slope appears to be temperature independent, but the duration of phase I increases with increasing temperature and a lower roughness in the minimum at higher bulk film thicknesses is reached. Phase II: A subsequent phase with a strong roughness increase—the onset of this phase is in between 20 and 100 Å, depending on the bulk thickness at which the minimum occurs. This onset might be related to the a-Si$\rightarrow a$-Si roughening transition introduced by Collins *et al.*[11] Also this phase shows no strong temperature dependence for unbiased depositions. For biased depositions the roughening is stronger for higher substrate temperatures. When analyzing the surface roughness development in this phase according to the dynamic scaling theory, we can determine the growth exponent β.[3] A growth exponent of 0.5 is obtained under complete absence of any smoothening or roughening conditions, simply due to the random nature of the growth flux distribution. Consequently, a β-value below 0.5 is a direct result of the presence of smoothening mechanisms. The peculiarly strong roughening in this phase with $\beta > 0.8$ (not shown)
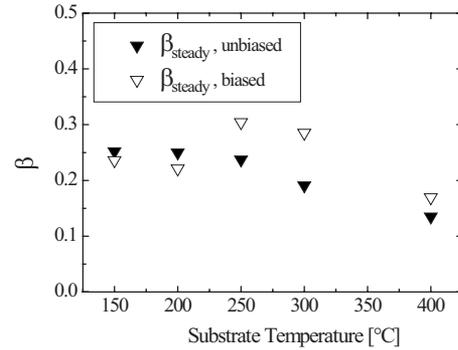
indicates the dominance of roughening mechanisms. A comparable strong roughening early in the deposition was previously reported e. g. for HW-CVD depositions,[2] as well as a strikingly similar roughness development throughout the deposition in general. In other literature, strong roughening, either throughout or in certain stages of film growth, has been reported for amorphous thin-film deposition in recent years, ranging from $\beta = 0.7-1.5$, both in experiment and in simulation.[2,5,12–21] Typical mechanisms for strong roughening include shadowing,[13,14,20,21] columnar growth,[17] or diffusion barrier steps.[18] Phase III: The steady growth phase with the β-value labeled β_{steady}—at a film thickness between 300 and 400 Å the strong roughening levels off and the film enters the steady growth phase for the rest of the deposition. When analyzing the surface roughness development in this phase according to the dynamic scaling theory (see, e.g., Ref. 16), we can determine the growth exponent β from the relation $d_s \sim t^\beta$ with t the deposition time. A β-value below 0.5 as we observe in our experiment ($\beta_{steady} \sim 0.25-0.15$, Fig. 2) is a direct result of the presence of a smoothening mechanisms. We can observe a weak temperature dependence for β.

The reduction of the surface roughness minimum in the coalescence phase at the transition from phase I to phase II seems to be enhanced by substrate biasing at elevated temperatures, resulting in smoother surfaces at higher temperatures in the roughness dip, as can be seen in [Fig. 1(b)]. However, we suggest that surface smoothening in this phase is not due to a reduction of the actual surface roughness, but is in fact related to the presence of a hydrogen-rich layer in the early growth phase for unbiased depositions and its removal/densification under substrate biasing. The formation of a hydrogen-rich layer in the early growth phase has been reported, e.g., by Fujiwara *et al.*[22] It has a significantly lower density and dielectric function than the bulk a-Si:H network and can thus be misinterpreted as surface roughness by SE measurements. This was also demonstrated by Fujiwara *et al.*[23] and a hydrogen concentration of ~25% was estimated for the initially deposited monolayers. The removal or densification of this layer by ion bombardment can explain what is misinterpreted as a reduction in surface roughness in the coalescence phase by the SE. Due to the broad ion energy distribution obtained with rf substrate biasing both ion-surface atom interactions as well as ion-subsurface atom interactions, which require higher energetic ions, could lead to this densification. From further SE analysis we can see that indeed the surface roughness layer in this phase shows a void fraction of about 33% for unbiased depositions and around

42% for biased depositions at 400 °C (not shown), indicating that material with very low density might be interpreted as surface roughness for unbiased depositions and is removed by substrate biasing at elevated temperatures. The temperature dependence of this effect suggests that also thermally activated mechanisms are involved and in fact required in order to lead to the removal/densification of the hydrogen rich layer, e.g., by facilitating the abstraction of hydrogen atoms from surface or subsurface layers. Within this interpretation we must conclude that thermal energy alone, however, is not sufficient and ion bombardment is required, as can be deduced from [Fig. 1(a)] where a reduction in roughness at the roughness minimum cannot be observed even at 400 °C.

The presence of smoothening mechanisms in phase I is obvious, demonstrated by the strong decrease in roughness during that phase. Dominance of smoothening mechanisms in the steady growth phase, phase III, is less obvious, but can be deduced from dynamic scaling theory as discussed above. The presence of extraordinarily strong roughening in phase II, however, indicates a temporary reduction of smoothening processes. This observation is not compatible with surface diffusion-driven smoothening. If surface diffusion is the main smoothening mechanism in phase III, it would require rather long diffusion lengths comparable to the feature size on the surface. However, with such long diffusion lengths any kind of surface features present in phase II would be smoothened and strong roughening should not occur. Consequently, would surface diffusion be dominant in both phase I and III, the presence of phase II would require a strong temporary reduction of radical diffusion at the roughness minimum and its resumption at the beginning of phase III. Such change in radical diffusion is highly unlikely, as there is no change in radical flux arriving at the surface. Alternatively, we might have a very strong roughening mechanism that is only present in phase II, but such a short-term roughening mechanism has not been observed or suggested in literature yet. Therefore we anticipate the presence of strong roughening in phase II implies that surface diffusion cannot be the dominant smoothening mechanism in a-Si:H film growth.

Re-emission is an alternative nonlocal mass transport mechanisms that can explain smoothening on large lateral length scales. In the re-emission model a particle with a sticking coefficient <1 can be re-emitted from surface features upon impact and transported deeper into the surface valley, thus transporting mass into surface valleys. Re-emission is related to the shadowing effect where particles with high sticking coefficients lead to enhanced growth of surface protrusions over surface valleys by receiving more growth flux under a non-normal angular distribution. A balance between shadowing and re-emission as dominant roughening and smoothening mechanisms during film growth is able to explain β-values in a wide range from 0.1 to >1.[24] It can therefore explain both the dominant roughening in phase II and dominant smoothening in phase III by implying a change in the balance between re-emission as smoothening effect and shadowing as a roughening effect. Re-emission is a temperature independent process for growth precursors with temperature independent sticking coefficients as determined for SiH$_3$ precursors.[10]

However, also with re-emission as dominant mass transport mechanism, the origin of the strong roughening in phase II cannot easily be explained, as the nature of the shift in balance between re-emission and shadowing needs to be identified. It might be related to the fact that re-emission requires a certain inclination of the surface slopes before it can act as smoothening mechanism. Also the formation of cusps at surface feature edges, as suggested by Singh *et al.*,[25] might play a role here.

In conclusion, we have investigated the temperature dependence of surface roughness evolution for a-Si:H thin film deposition with and without external rf substrate biasing. The effect of external rf substrate biasing on the coalescence phase is discussed and a removal/densification of a hydrogen-rich layer is suggested to explain the observed roughness development in this phase. Following a discussion of two distinct phases in the roughness development of bulk film growth we suggest that alternative, nonlocal growth mechanisms, like the re-emission effect, could play an important role in a-Si:H film growth.

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