Temperature dependence of the surface roughness evolution during hydrogenated amorphous silicon film growth

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The scaling behavior of the surface morphology of hydrogenated amorphous silicon deposited from a SiH3 dominated plasma has been studied using atomic force microscopy and in situ ellipsometry. The observed substrate temperature dependence of growth exponent \( \beta \) reflects a crossover behavior from random deposition at 100 °C to a surface diffusion controlled smoothening around 250 °C to full surface relaxation around 500 °C. This crossover behavior has been reproduced by Monte Carlo simulations assuming a site dependent surface diffusion process, revealing an activation energy of \( ~1.0 \text{ eV} \) for the ruling surface smoothening mechanism. The implications for \( \alpha \)-Si:H growth are discussed. © 2003 American Institute of Physics. [DOI: 10.1063/1.1543237]

The evolution of the surface morphology of thin films can show scaling characteristics that are directly related to the growth processes occurring during film growth. In this letter we address the evolution of the surface morphology of hydrogenated amorphous silicon (\( \alpha \)-Si:H) to obtain insight into surface diffusion processes that are generally assumed to be crucial for \( \alpha \)-Si:H growth. More specifically, we will provide more evidence for the fact that surface diffusion of physiosorbed SiH3 radicals is not responsible for the \( \alpha \)-Si:H surface smoothening mechanism, as has often been suggested in the literature.

Family and Viscek introduced a general scaling law that describes the evolution of the root-mean-square roughness \( W(L,t) \) of a growing film. Adopting the notation of Barabasi and Stanley, the \( W(L,t) \) values of a so-called self-affine surface obey the scaling laws \( W(L,t) \propto L^a F(u) \propto t^\beta \) for \( u \ll 1 \) and \( W(L,t) \propto L^a F(u) \propto L^\alpha \) for \( u \gg 1 \), with \( L \) the system size of the surface, \( t \) the mean film thickness, \( u = t/t_x \), a normalized scaling parameter, \( t_x \propto L^{1/\beta} \) the crossover thickness, \( \alpha \) the roughness scaling exponent, and \( \beta \) the growth scaling exponent. Every self-affine growth process can be classified according to its scaling exponents \( \alpha \) and \( \beta \). For example, a pure diffusion class with \( \alpha = 0.96 \) and \( \beta = 0.26 \) has been reported for \( \alpha \)-Si growth by thermal evaporation. However, for \( \alpha \)-Si:H growth by plasmas the reported \( \alpha \) and \( \beta \) values are in the range 0.5–1.0 and 0.1–0.5 for \( \alpha \) and \( \beta \), respectively. In this letter, we report the scaling behavior of the \( \alpha \)-Si:H roughness evolution for a broad range of growth rates \( R_d \) and substrate temperatures \( T_{sub} \). The films have been deposited by the remote expanding thermal plasma (ETP) under conditions in which film growth is dominated by SiH3 and in which ion bombardment is absent. The surface reactions are, therefore, purely chemical.

The scaling exponents for the \( \alpha \)-Si:H films have first been studied using atomic force microscopy (AFM). Figure 1 shows typical AFM data in which the normalized values \( W(l,t)/l^\alpha \) and \( W(l,t)/l^\beta \) have been plotted versus the scaling parameter \( u = t/t_x \). For these data both the AFM scan size \( l \) (10 nm–10 \( \mu \)m) and the film thickness \( t \) (10 nm–3 \( \mu \)m) have been varied. The slopes of the dotted lines in Figs. 1(a) and 1(b) correspond to a roughness and growth scaling exponent of \( \alpha = 0.9 \pm 0.2 \) and \( \beta = 0.28 \pm 0.03 \), respectively. The fact that \( W(l,t)/l^\alpha \) is proportional to \( u^\beta \) and \( W(l,t)/l^\beta \) is constant for \( u \ll 1 \) implies that \( \beta \) is well defined when \( u \ll 1 \). For \( u \gg 1 \) on the other hand, it is not evident that \( W(l,t)/l^\alpha \) is constant and that \( W(l,t)/l^\beta \) scales with \( u^{-\beta} \). This is caused by the limited sensitivity of AFM for small scan sizes. [Note that \( l \neq L \), however, \( W(l,t) \) scales with \( l^\alpha \) when \( t < t_x \) (Ref. 1)]. The \( u \gg 1 \) region is, therefore, experimentally hard to access, because accurate determination of \( \alpha \) requires large \( u \) values, and thus the measurement of very thick \( \alpha \)-Si:H films on small scan sizes \( l \). As a consequence, the rescaling of the data as done in Fig. 1 yields no reliable information on \( \alpha \).

More accurate is the determination of \( \alpha \) from the power

![FIG. 1. Normalized root-mean-square roughness \( W(l,t)/l^\alpha \) (a) and \( W(l,t)/l^\beta \) (b), obtained from AFM analyses of \( \alpha \)-Si:H deposited at 70 \( \text{Å/s} \) and 250 °C, plotted vs scaling parameter \( u = t/t_x \).](image-url)
Fig. 2. Growth exponent $\beta$ of ETP $a$-Si:H vs substrate temperature for $R_d$=2 Å/s$^{-1}$ (●, RCE), 5 Å/s$^{-1}$ (□, RCE), 22 Å/s$^{-1}$ (△, RCE), 22 Å/s, and 70 Å/s (△, AFM). $\beta$ values obtained from AFM and scanning tunneling microscopy measurements as reported in the literature [●, 2 Å/s$^{-1}$ (Ref. 3); □ (Ref. 4); △, 0.3 Å/s$^{-1}$ (Ref. 5); ▲, 0.7 Å/s$^{-1}$ (Ref. 6)] are included as well. Simulation results using the SOS model are given by the solid line ($E_a=0.7$ eV, $E_1=0.3$ eV); and dashed line ($E_a=1.0$ eV, $E_1=0.3$ eV). The RD, RDD, and RDR universality classes are indicated by the horizontal dotted lines. In the inset the $W(l,t)$ values obtained by AFM (●) and RCE (●) are given vs deposition time $t$ ($R_d=22$ Å/s, $T_{sub}=250$ °C).

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spectrum density of the AFM scans.$^1$ This has revealed $\alpha$ values ranging from 0.4 up to 0.8 for the data described in this letter.

The AFM data in Fig. 1 show that reliable values of the growth exponents $\beta$ can be obtained from $W(l,t)$ as a function of sample thickness $t$ in the range $u \ll 1$. The $\beta$ values can be determined more accurately than $\alpha$ and, therefore, we will use the $\beta$ values to determine the scaling universality of $a$-Si:H growth. Furthermore, $\beta$ values have been obtained by monitoring the $a$-Si:H roughness evolution by real time ellipsometry,$^2,15$ which automatically yields data in the $u \ll 1$ range of Fig. 1 due to the macroscopic surface area probed by the ellipsometer. We used a rotating compensator ellipsimeter (RCE) operating at single wavelength (632.8 nm).$^15$ The inset in Fig. 2 shows a comparison between $W(l,t)$ values obtained from RCE and AFM for the same deposition condition. The good agreement between the $\beta$ values of RCE ($\beta=0.27 \pm 0.03$) and AFM ($\beta=0.29 \pm 0.03$) validate that RCE is a “fast” method to accurately determine $\beta$.$^15$

The values of $\beta$ obtained by AFM and RCE for various growth rates is plotted versus substrate temperature $T_{sub}$ in Fig. 2. It is evident that $\beta$ is always smaller than 0.5 and that $\beta$ decreases with increasing $T_{sub}$. This decrease suggests a temperature dependent surface smoothening mechanism. Moreover, this dependence of $\beta$ on $T_{sub}$ resembles a general trend as results reported in the literature (included in Fig. 2) for a wide variety of $a$-Si:H deposition conditions show good agreement. Furthermore, within the accuracy of $\beta$ no or a weak growth rate dependence has been found, although $R_d$ varies more than one order of magnitude.

In the literature, a limited number of universality classes has been reported. For a two-dimensional surface the simplest universality class is “random deposition” (RD) in which the randomly arriving SiH$_3$ radicals stick at impact on the surface without any subsequent surface smoothening mechanism. For the RD universality class $\beta=\frac{1}{2}$ and $\alpha$ is undefined.$^1$ The universality class in which the randomly arriving radicals diffuse instantaneously to the nearest step-like surface site is called “random deposition with surface diffusion” (RDD). For RDD, simulations have revealed that $\alpha \approx 1$ and $\beta \approx 0.25$. The “random deposition with relaxation” (RDR) universality class allows the radicals to stick at local height minima. RDR, which corresponds to $\alpha=\beta=0$, leads therefore to even more surface smoothening.$^1$ When we consider these three unique universality classes, Fig. 2 reflects the crossover from RD at 100°C to RDD around 250°C and to RDR for the highest substrate temperatures.$^7$

Figure 2 suggests a gradual transition between the different universality classes. This can be explained by the fact that surface smoothening mechanisms become activated when increasing $T_{sub}$, whereas RD, RDD, and RDR each describe growth with only one specific smoothening mechanism. This smoothening mechanism is assumed to be instantaneous and does, therefore, not reveal information about thermal activation. During actual $a$-Si:H growth, adsorption of the SiH$_3$ radical can take place in multiple ways and, consequently, also more than one activation energy for surface diffusion can be involved. We invoke, therefore, that the crossover in universality classes for $a$-Si:H growth under the purely chemical deposition conditions can be interpreted in a similar way as for film growth under molecular beam epitaxy (MBE) as done in the work by Das Sarma and co-workers.$^{16}$

Analogously to the work on MBE, we have assumed that the crossover phenomena in Fig. 2 are caused by a multiple diffusion processes of species adsorbed on the film’s surface. In a solid-on-solid (SOS) model, similar to the one used by Das Sarma and co-workers,$^{16}$ we have included multiple hopping rates for the growing species.$^7$ Although film growth is assumed to take place by adsorption of SiH$_3$ radicals, there is in the SOS model no need to actually specify the diffusing species that result in the silicon atom transport over the surface. We also want to point out that the $a$-Si:H surface is covered by hydrogen, such that the diffusion process is probably more complex than in the case of MBE. The activation energies in the model are assumed to be described by $E_0 + nE_1$, in which $n=1$ corresponds to diffusing species on terrace sites, $n=2$ corresponds to diffusion of stronger bonded species at step sites, etc. Note that $E_0$ is the lattice potential for diffusion whereas $E_1$ reflects a binding energy and, therefore, only the sum of $E_0 + nE_1$ has the physical meaning of a hopping activation energy.

Figure 2 shows the $\beta$ values obtained from the SOS model by Monte Carlo simulations. Comparing the computed $\beta$ values with the experimental ones, reveals that relatively high values for $E_0$ ($\sim 0.7$ eV) and $E_1$ ($\sim 0.3$ eV) yield the best agreement. This implies that the lowest activated diffusion process has a relatively high activation energy of about $\sim 1.0$ eV. This high activation energy also causes that only a slight increase in $T_{sub}$ is needed to enhance the diffusion rates, and thus to compensate for the increased arrival rate of growth species. This is in agreement with the weak $R_d$ dependence observed in Fig. 2. The model also predicts that $\beta \approx 0$ for high substrate temperatures ($>500$ °C), in agreement with both the experiments and the RDR regime.

Although the model and simulations are considerably
simplified, the SOS model shows that the crossover in universality classes for increasing $T_{\text{sub}}$ can be explained by surface diffusion processes with rather high activation energies. The mechanism that is ruling the smoothening of the surface is activated by $\sim 1.0$ eV and this value is incompatible with surface smoothening by diffusion of physisorbed SiH$_3$ radicals, with assumed activation energies of $\sim 0.2$--$0.3$ eV, as often proposed in the literature. This provides evidence that physisorbed SiH$_3$ is not responsible for the decreasing surface roughness of a-Si:H with increasing $T_{\text{sub}}$, in accordance with the earlier conclusion that not physisorbed SiH$_3$ but the position of the dangling bonds on the surface determines the surface roughness. The dangling bonds act as growth sites for the SiH$_3$ radicals and ultimately the SiH$_3$ radicals will stick at these dangling bonds, even in the case that the physisorbed SiH$_3$ radicals have diffused over the surface. Note, that from our data we do not exclude the presence of physisorbed SiH$_3$ radicals. Physisorbed SiH$_3$ radicals might still be necessary to explain the low dangling bond (DB) density by a Kisliuk type of adsorption process.

A mechanism that could be compatible with the high activation energy for surface diffusion is the diffusion of chemisorbed hydrogen. This process is observed on hydrogenated crystalline silicon and might have a reduced activation energy on the relaxed amorphous surface of a-Si:H. Hydrogen diffusion can cause the dangling bonds to end up at preferential surface sites such as steps or in valleys. However, when we hold on to the existence of physisorbed SiH$_3$ radicals, the lifetime $\tau_{DB}$ of a dangling bond is given by $\tau_{DB} = (\theta_{DB} \nu_3)^{-1}$, with $\theta_{DB}$ the thermally activated attempt frequency for hopping of the SiH$_3$ radical. If we assume that $\theta_{DB} = 10^{-4}$ and $\nu_3 = 10^{10}$ Hz, we find $\tau_{DB} = 10^{-6}$ s a value too short for the dangling bonds to reach the surface steps or valleys within the growth time of a monolayer (typically, $> 10$ ms). In this case, the diffusion of chemisorbed surface silicon atoms might cause the surface smoothening. The diffusion of this kind of species would also be compatible with the high activation energy of $\sim 1.0$ eV, although exact activation energies for such species are difficult to predict due to the presence of hydrogen on the a-Si:H surface.

In conclusion, we have investigated the substrate temperature dependence of the growth exponent $\beta$ for a-Si:H growth dominated by SiH$_3$ radicals under purely chemical conditions. A general trend in which $\beta$ decreases with increasing substrate temperature has been observed reflecting the crossover from universality classes RD $\rightarrow$ RDD $\rightarrow$ RDR with increasing temperature. Comparing this trend with Monte Carlo simulations based on a simplified SOS model we have concluded that the diffusion process ruling surface smoothening is activated with $\sim 1.0$ eV. This high activation energy provides additional evidence that physisorbed SiH$_3$ does not explain the decreasing surface roughness of a-Si:H with increasing substrate temperature.

18. Cases in which activation energies of $\sim 0.2$ eV were reported from studies of the roughness evolution (see Ref. 8) rely on the identification of the correlation length of the surface roughness with the diffusion length of the diffusion species. This identification is not substantiated and probably erroneous (see Refs. 1 and 3).