Experimental and theoretical study of the evolution of surface roughness in amorphous silicon films grown by low-temperature plasma-enhanced chemical vapor deposition

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Using real-time spectroscopic ellipsometry the evolution of the surface roughness of amorphous silicon thin films grown by low-temperature (200 °C) plasma-enhanced chemical vapor deposition (PECVD) at high process gas pressure (3 mTorr) has been studied as a function of the hydrogen dilution gas-flow ratio $R_H = [H_2]/[SiH_4]$ with $15 \leq R_H \leq 60$. To describe the roughness evolution, we have used a 3D linearized continuum equation which includes a negative surface-tension term to take into account the destabilizing effects of short-range attraction and/or shadowing, as well as a smoothing term corresponding to surface diffusion. Using this model we have obtained very good agreement with experimental results for the evolution of the surface roughness in the case of large dilution ratio. However, our results indicate that for small dilution ratio the surface slopes are significantly larger and as a result additional nonlinear terms need to be included at large thicknesses. Our results also indicate that surface diffusion plays an important role during PECVD film growth while the diffusion rate increases with increasing hydrogen dilution ratio. We also find that the early stages of island nucleation play an important role in determining the subsequent roughness evolution. In particular, the assumption of a large wetting angle ($\theta_W \simeq 90°$) for the 3D islands formed in the initial stages leads to significantly better agreement with experiments than a smaller wetting angle ($\theta_W \simeq 45°$). This is consistent with recent experiments on liquid Si droplets on SiO_2 [H. Kanai *et al.*, J. Mater. Sci. **42**, 9529 (2007)] substrates in which a wetting angle of 90° was observed.

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I. INTRODUCTION

Amorphous silicon (a-Si:H) thin films grown via chemical vapor deposition (CVD) methods are used in a variety of technologically important areas ranging from large-area microelectronic devices to solar cells. Since the device properties depend on thin-film quality, i.e., near-surface and bulk microstructure and H bonding,[1] there has been significant interest in developing a greater understanding of the growth process, as well as of the dependence of the properties on growth parameters.

Features in the evolution of the surface roughness of a-Si:H films have been correlated with material and device properties. In particular, it has been found that in a-Si:H based thin-film solar cells, the optimum device performances are achieved by incorporating intrinsic a-Si:H layers fabricated under conditions that produce enhanced surface smoothing. Therefore, in addition to modeling the thin-film microstructure and its dependence on hydrogen dilution ratio,[2,3] there is also interest in understanding the dependence of the surface roughness evolution on deposition conditions.

Recently, simplified kinetic Monte Carlo simulations[2,3] of atomistic models of a-Si:H grown by plasma-enhanced CVD (PECVD), based on *ab initio* calculations for the relevant activation barriers, have been used to model the dependence of the degree of crystallinity on the hydrogen dilution ratio $R_H = [H_2]/[SiH_4]$. However, due to the complexity of the PECVD process,[4–6] these simulations were not able to simulate the evolution of the surface morphology.[7] Therefore, it is of interest to consider somewhat simplified models in order to try to understand the roughness over larger length scales.

Here we present the results of simulations of a simplified coarse-grained continuum model of a-Si:H growth carried out in order to understand the evolution of the surface roughness and its dependence on the hydrogen dilution ratio. Our model takes into account two competing effects—the effect of roughening due to shadowing and/or attraction of depositing species to the film and a smoothing effect due to surface diffusion. In order to model the initial configuration, we have also used a simplified model of nucleation. For correlation with experiments, we have compared our results with real-time spectroscopic ellipsometry (RTSE) results for low-temperature (200 °C) PECVD at high process gas pressure (3 mTorr). By varying the parameters in our model, we find good agreement between our simulations and experiments for the case of high dilution ratio. However, our results also indicate that for the case of low dilution ratio, for which the surface is significantly rougher, additional effects (such as nonlinearities) may be important. A comparison of our results for different dilution ratios also indicates that the effects of diffusion are greatly enhanced with increasing R_H. Our results also indicate that the early stage of nucleation of 3D islands plays a very important role in determining the subsequent roughness evolution.

This paper is organized as follows. In Sec. II we first describe the experimental setup and data taken in our RTSE measurement of the surface roughness while in Sec. III we discuss in detail the model used in our simulations. Comparisons between our simulation results and experiments are presented in Sec. IV. Finally, in Sec. V we discuss our results as well as possible future work.

II. EXPERIMENT

The experimental results presented here have been previously discussed in Ref. 8. However, for completeness here

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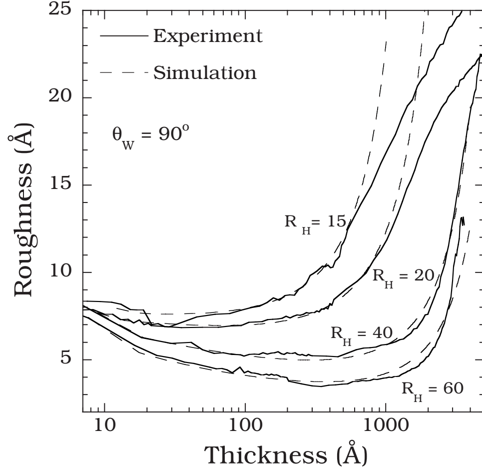

FIG. 1. Experimental roughness (solid lines) and theoretical fits (dashed lines) using Eq. (2) as function of average film thickness for H_2 dilution ratios R_H=15,20,40,60. Fits correspond to initial surface profile with wetting angle θ_W=90°.

we briefly summarize some of the experimental details. Real-time spectroscopic ellipsometry has been performed in order to study the evolution of the surface roughness thickness for Si:H films fabricated by PECVD on native-oxide-covered [(SiO_2)-covered] crystalline silicon (c-Si) wafer substrates. In our experiments, a rotating-compensator multichannel ellipsometer[9] was affixed to a PECVD chamber at an angle of incidence of 70° from the normal of the substrate, with a spectral range from 1.5 to 5.0 eV and a minimum spectral acquisition time of 80 ms.[9] The substrate temperature T, rf (13.56 MHz) plasma power P and total pressure p were fixed at the following values: T=200 °C, P=0.72 W/cm^2, and p=3 mTorr. The H_2 dilution ratio $R_H=[H_2]/[SiH_4]$, was set at four different values 15, 20, 40, and 60, leading to deposition rates of 18, 14, 9.5, and 6.6 Å/s, respectively. The motivation of this work was to apply a combination of high gas pressure and high hydrogen dilution—the latter up to but not exceeding the amorphous-to-(mixed phase nanocrystalline) transition $[a\rightarrow(a+nc)]$—in order to increase the deposition rate while retaining high-quality a-Si:H films for photovoltaic applications. The elevated process gas pressure of approximately 3 mTorr over the standard value (0.2 mTorr) leads to an almost 15× increase in deposition rate, from about 0.5 to 6.6 Å/s for the highest value of R_H (R_H=60) just before the $a\rightarrow(a+nc)$ transition.

As shown in Fig. 1, the surface roughness evolution depends strongly on R_H and exhibits three distinct regimes:[1] (i) smoothing (or coalescence) of initial nucleation-generated clusters, (ii) stable surface until a critical thickness, and (iii) roughening of the film beyond the critical thickness. For example, for R_H=60, the smoothing regime runs from a film thickness of approximately 8 to 200 Å while the stable regime runs from 200 to approximately 1200 Å and finally roughening begins at 1200 Å. The RTSE spectra collected during PECVD of these films were analyzed using a two-layer $\Sigma-\sigma$-minimization procedure.[10] In this approach, an unweighted error-function minimization method combining numerical inversion and least-squares regression analysis is used to simultaneously obtain the film-specific complex dielectric function spectra and the time evolution of the surface roughness and bulk film thickness.[11] Accurate characterization of the microstructural and optical properties of the substrate is obtained from spectra collected prior to a-Si:H deposition so that the two layers used in the $\Sigma-\sigma$—minimization procedure consist of only the bulk film of the material and a surface roughness layer represented by a Bruggeman effective-medium approximation of 50% volume fraction bulk and 50% volume fraction void.[12] In previous work, the surface roughness W_{RTSE} obtained from RTSE measurements has been compared to the root-mean-square surface roughness W_{AFM} obtained from antiferromagnetic measurements.[12,13] A linear relationship of the form $W_{AFM}=0.667\ W_{RTSE}-2$ Å was observed for W_{RTSE} values up to 100 Å. Therefore, we have used this expression to convert the surface roughness obtained from RTSE to the results presented here.

III. MODEL

In order to model the roughness evolution in low-temperature PECVD, we have assumed that the growth takes place in two stages. In particular, we assume that in the first stage 3D islands of a-Si:H nucleate on the native-oxide-covered c-Si substrate and then coalesce to form a bulk layer while in the second stage the film continues to grow but no new islands are nucleated. While it may eventually be possible to use atomistic models to simulate the nucleation and growth of 3D islands, for simplicity here we have assumed that at the end of the nucleation stage, the film consists of a random distribution of spherical islands or "caps" with wetting angle θ_W which have a Gaussian distribution of radii. In equilibrium, one expects that the wetting angle of a "droplet" satisfies Young's equation, e.g., $\cos(\theta_W)=(\gamma_{12}-\gamma_{13})/\gamma_{23}$, where γ_{12}, γ_{13}, and γ_{23} correspond to the free energies of the substrate/a-Si:H interface, a-Si:H surface, and the substrate surface, respectively. However, since these free energies are not known and may also depend on the dilution ratio $R_H=[H_2]/[SiH_4]$, for simplicity we have carried out simulations for two different values, e.g., θ_W=45° and θ_W=90°.

To describe the subsequent thin-film evolution, we consider the simplest possible continuum equation which takes into account the effects of roughening due to shadowing and/or or the attraction of depositing species to the film, and smoothing due to surface diffusion and/or evaporation. In particular, we consider the following nonlinear equation for the surface height $h(\vec{r},t)$ at position $\vec{r}$ and time t,

$$\frac{\partial h(\vec{r},t)}{\partial t} = -A_1 M + A_2\sqrt{g}M - A_3\sqrt{g}\Delta_s M + F + \eta(\vec{r},t), \quad (1)$$

where $g=1+|\nabla h|^2$, $M=g^{-3/2}\Sigma_{i,j=x,y}(g\delta_{ij}-h_i h_j)h_{ij}$ is the mean curvature (where $h_x=\partial h/\partial x$, $h_{xy}=\partial^2 h/\partial x\partial y$, etc.), Δ_s is the surface Laplacian, F is the deposition flux, and $\eta(\vec{r},t)$ is a noise term which takes into account the effects of both deposition and diffusion noise. Assuming that $A_1=\rho F$, where ρ is the radius of the depositing species, the first term corresponds to "atomic shadowing" as first proposed by Mazor *et al.*[14] in the context of columnar growth. However, a simi-

lar term has also been derived by Raible *et al.*[15,16] due to attractive forces between the surface atoms and the incident particles. Similarly, the second term[17] corresponds to a (positive) "surface tension" due to evaporation/condensation or re-emission ("antishadowing") while the term with A_3 corresponds to surface diffusion.[18] We note that in the context of a single-species growth model, e.g., only SiH_3, one may write $A_3 = D_s \sigma_s \Omega^2 \epsilon / kT$, where D_s, Ω, and ϵ are the diffusion constant, atomic volume, and surface density of the diffusing species, respectively, σ_s is the surface tension, k is Boltzmann constant, and T is the film temperature. For amorphous growth we expect that the diffusion constant D_s is independent of the local slope and thus it is reasonable to assume that A_3 is a constant.

Linearizing this equation and dividing by the flux F—and also ignoring the noise term since it is expected to be small beyond the nucleation regime—we obtain,

$$\frac{\partial h(\vec{r},\bar{h})}{\partial \bar{h}} = 1 - \nu_2 \nabla^2 h(\vec{r},\bar{h}) - \nu_4 \nabla^4 h(\vec{r},\bar{h}), \qquad (2)$$

where $h(\vec{r},\bar{h})$ is a function of the transverse coordinate $\vec{r}$ and average height $\bar{h} = Ft$. Here $\nu_2 = (A_1 - A_2)/F$ and $\nu_4 = A_3/F$. As can be seen, this equation contains two competing terms—a second-order instability term due to shadowing and/or attraction modified by the effects of evaporation/condensation and a fourth-order smoothing term due to surface diffusion. As long as the local slope is less than 1, the solution of the linearized equation should give a reasonable approximation to the full nonlinear equation in the absence of deposition noise. In addition, it has the advantage that it can be efficiently integrated using a fast Fourier transform starting from the initial surface profile $h_0(\vec{r},\bar{h})$. In particular, changing from $\vec{r}$ to $\vec{k}$ space $h(\vec{r},\bar{h}) \to \tilde{h}(\vec{k},\bar{h})$ leads to the solution

$$\tilde{h}(\vec{k},\bar{h}) = \tilde{h}(\vec{k},\bar{h}_0)\exp[(\nu_2 - \nu_4 k^2)k^2 \bar{h}]. \qquad (3)$$

As can be seen from Eq. (3) there is a critical wave vector $k_c = \sqrt{\nu_2/\nu_4}$ such that short-wavelength surface modulations with $k > k_c$ disappear exponentially due to surface diffusion while those with $k < k_c$ (i.e., long wavelengths) grow exponentially with film thickness. The most unstable mode corresponds to $k_m = k_c/\sqrt{2}$. The surface profile $h(\vec{r},\bar{h})$ is then obtained from $\tilde{h}(\vec{k},\bar{h})$ by carrying out an inverse Fourier transform.

As already noted, in order to model the roughness evolution in low-temperature PECVD, we have assumed that the growth takes place in two stages. In particular, we have assumed that at the end of the initial nucleation stage, the film consists of a random distribution of semi-spherical islands or caps with wetting angle θ_W which have a Gaussian distribution of radii with mean value $\bar{R}$ and standard deviation σ. Using the relationship $W_{RTSE} = 3$ Å $+ 1.5$ W_{AFM} (see Sec. II) the initial roughness values in Fig. 1 may be converted to a surface roughness layer thickness $d_s = W_{RTSE}$ with a value of approximately 15 Å. Using a geometrical picture, this suggests that the average radius $\bar{R}$ at the end of the nucleation stage is on the order of 15 Å. Accordingly, in our simulations we have used values of $\bar{R}$ close to this value in order to fit the experimental results. In addition, since the initial roughness depends relatively weakly on the dilution ratio R_H, in our simulations we have assumed an initial island density ($N_0 = 2.9 \times 10^{-3}$ Å^{-2}) which is the same for all values of R_H. Thus, for a given choice of experimental dilution ratio and assumed wetting angle θ_W, values of the average island radius $\bar{R}$ and standard deviation σ were varied in order to obtain an initial configuration for which there is good agreement with the experimental roughness at an average film thickness in the range $\bar{h} \simeq 10-20$ Å. We note that since the initial island distribution was random, this resulted in a significant overlap between different islands—the resulting initial profile was then determined by the maximum height at each point.

In order to determine the subsequent roughness evolution, the resulting initial surface configurations were then numerically integrated using Eq. (3) for different values of the parameters ν_2 and ν_4 to find the best fit. It is worth mentioning that for a given dilution ratio R_H, good fits were only possible for specific values of $\bar{R}$ and σ. For determination of these values an iterative process was used, e.g., values of $\bar{R}$ and σ matching the initial experimental roughness were first determined, and then ν_2 and ν_4 were varied to obtain the best match with experiment. If the resulting best fit was not good, then new values of $\bar{R}$ and σ were selected. This process was continued until reasonable results were obtained up to a film thickness of 500–2000 Å depending on the value of R_H. Thus, for a given choice of initial island density the resulting process puts significant constraints not only on the parameters ν_2 and ν_4 but also on the parameters $\bar{R}$ and σ describing the initial conditions.

IV. RESULTS

We first consider our simulation results for the case of a large assumed wetting angle $\theta_W = 90°$. Figure 1 shows the corresponding best-fit simulation results for the surface roughness along with the corresponding experimental results. As can be seen there is very good agreement with experiment for the two largest dilution ratios $R_H = 40$ and $R_H = 60$. However, for the two smaller values of R_H ($R_H = 15$ and 20) the simulated roughness is significantly higher than the experimentally obtained roughness for thicknesses larger than $\bar{h} \simeq 500-1000$ Å. We note that in this case the experimental roughness curve obtained via RTSE also exhibits a bend or inflection point. One possible explanation for this behavior is that a new physical mechanism comes into play for small dilution ratio which suppresses the continued rapid increase in the surface roughness at large thicknesses. Another possibility is that, as pointed out in Ref. 19, for large thicknesses the RTSE measurement tends to underestimate the surface roughness since it does not include the long-wavelength contribution. However, one would expect such an effect to occur only for thicknesses significantly larger than 1000 Å. In addition, it is not clear why such an effect should occur for small dilution ratios but not for larger dilution ratios.

TABLE I. Simulation parameters as a function of hydrogen dilution ratio R_H for different wetting angles. For all cases initial island density $N_0=2.9\times10^{-3}$ Å^{-2}.

$R_H(\theta=90°)$	ν_2 (Å)	ν_4 (Å^3)	σ (Å)	$\bar{R}$ (Å)
15	0.2	6	6	13.5
20	0.2	10.5	6	11
40	0.4	80	6	11
60	0.6	190	6	10
$R_H(\theta=45°)$	ν_2 (Å)	ν_4 (Å^3)	σ (Å)	$\bar{R}$ (Å)
15	1	110	36.5	10
20	1	250	31	10
40	1	570	26	10
60	1.6	1370	22	10

Table I shows the corresponding values for the average initial island radius $\bar{R}$ and standard deviation σ as well as of the parameters ν_2, and ν_4 as a function of the dilution ratio R_H for wetting angles $\theta_W=90°$ and $\theta_W=45°$. As can be seen, with our assumption of a dilution-ratio-independent initial island density N_0, the parameters $\bar{R}$ and σ also depend relatively weakly on the dilution ratio. In contrast the surface tension ν_2 increases with increasing R_H while the surface diffusion or smoothing parameter ν_4 increases even more dramatically with the H_2 dilution ratio. These results suggest that increasing the hydrogen dilution leads to increased surface diffusion of mobile species on the surface of the growing a-Si:H film. We note that this is reasonable since we expect that increasing the H_2 dilution leads to a decrease in the number of unsaturated bonds and/or defect sites at the surface, thus increasing the distance that mobile species can diffuse before being trapped or incorporated in the film.

Similar results are shown in Fig. 2 and Table I for the case of smaller wetting angle ($\theta_W=45°$). In this case, there is again reasonable agreement up to the largest film thicknesses for the two largest dilution ratios. However, the fit is not quite as good as for $\theta_W=90°$. Similarly, while there is good agreement for $\bar{h}<500-1000$ Å, for $R_H=15$ and 20, there is poor agreement for larger thicknesses. As for the case of $\theta_W=90°$, we find that the surface tension ν_2 increases weakly with increasing dilution ratio while the surface-diffusion parameter ν_4 increases much more rapidly with R_H. Also, the values of the parameter ν_2 and ν_4 are significantly larger for the case of small wetting angle. We note that while the average initial island radius $\bar{R}\simeq10$ Å is approximately the same as for higher wetting angle ($\theta_W=90°$), the width σ of the island-radius distribution is several times larger than the average radius. Figure 3 shows a summary of our results for the dependence of ν_2 and ν_4 on hydrogen dilution ratio R_H for both wetting angles. As can be seen, for both values of θ_W, the parameter ν_4 exhibits a power-law dependence on the dilution ratio, e.g., $\nu_4\sim(R_H)^\alpha$, where $\alpha\simeq2.6(1.7)$ for $\theta_W=90°(45°)$, respectively.

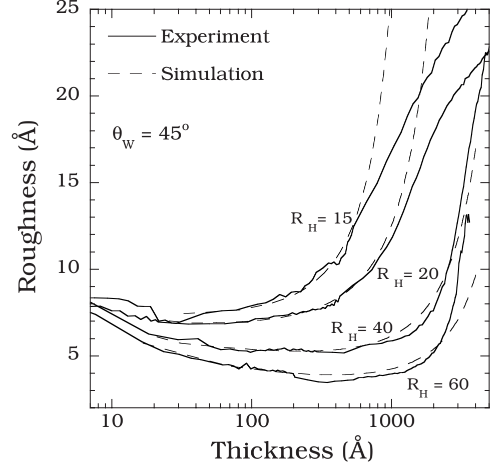

FIG. 2. Experimental roughness (solid lines) and theoretical fits using Eq. (2) (dashed lines) as function of average film thickness for H_2 dilution ratios $R_H=15,20,40,60$. Fits correspond to initial profile with wetting angle $\theta_W=45°$.

Figure 4 shows typical plots of the morphology obtained in our simulations at different thicknesses for the case $R_H=40$ and both wetting angles, e.g., $\theta_W=90°$ [(a)–(c)] and $\theta=45°$ [(d)–(f)]. As can be seen, the lateral length-scale tends to increase with increasing film thickness due to the elimination of small wavelength fluctuations. However, it appears to saturate at large thicknesses $\bar{h}>1000$ Å.

To study this behavior more quantitatively, we have fit the circularly averaged height-difference correlation function $G_2(r)=\langle[h(r)-h(0)]^2\rangle$ to the form $G_2(r)=w^2(1-e^{-r^2/\xi^2})$, where w is the surface roughness and ξ is the lateral correlation length. Figure 5 shows the corresponding results for the dependence of ξ on film thickness $\bar{h}$ for all four values of the dilution ratio for $\theta_W=90°$. Similar results (not shown) have been obtained for $\theta_W=45°$. As can be seen, while the correlation length ξ tends to increase with thickness at small thicknesses, it tends to saturate to a value proportional to the maximally unstable wavelength $\lambda_m\simeq\sqrt{2\nu_4/\nu_2}$ at larger thick-

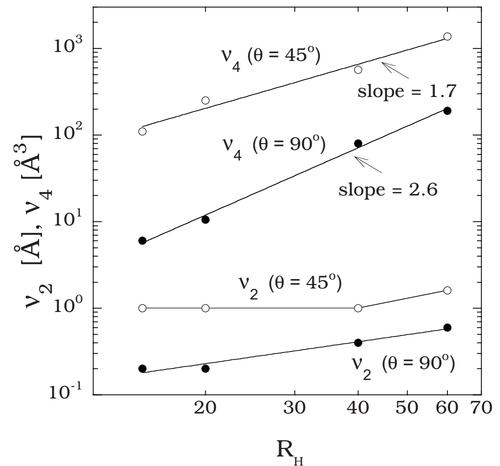

FIG. 3. Dependence of parameters ν_2 and ν_4 on hydrogen dilution ratio R_H for $\theta_W=90°$ and $\theta_W=45°$

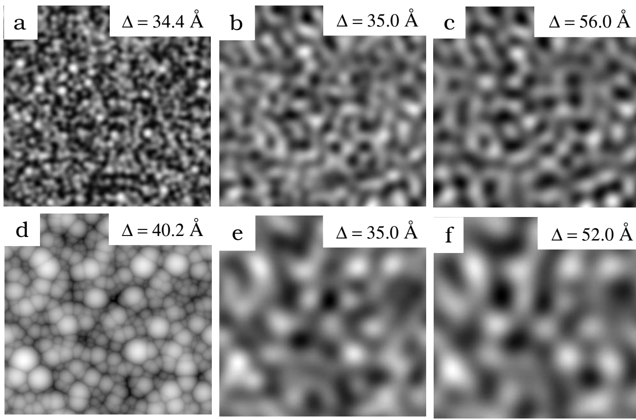

FIG. 4. Gray-scale pictures (1024 Å × 1024 Å) of simulated a-Si:H film surface for R_H=40 for thicknesses (going from left to right) $\bar{h}$=20 Å, $\bar{h}$=500 Å, and $\bar{h}$=2000 Å. (a)–(c) correspond to θ_W=90° while (d)–(f) correspond to θ_W=45°. Value of Δ indicates total height variation in each picture.

nesses. We also note that, except for the case R_H=15 for which the initial lateral length scale is *larger* than the selected wavelength λ_m, the film thickness at which the correlation length saturates corresponds approximately to that at which the surface roughness begins to increase rapidly as shown in Fig. 1.

V. DISCUSSION

Using RTSE we have measured the evolution of the surface roughness in low-temperature (200 °C), high-pressure (3 mTorr) a-Si:H growth via PECVD for values of the hydrogen dilution ratio ranging from R_H=15 to R_H=60. In general, we find that the surface roughness decreases with increasing R_H for a given bulk-layer thickness. In addition, the RTSE results indicate that the growth process may be divided into four distinct regimes: (i) an initial island nucleation regime, (ii) the formation of a bulk layer via island coalescence, (iii) growth at approximately constant roughness, and (iv) a rapid increase in roughness for large thicknesses.

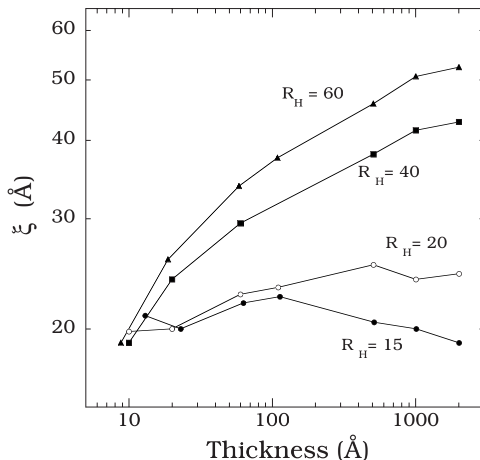

FIG. 5. Lateral length-scale parameter ξ as a function of average film thickness for R_H=15,20,40, and 60 and θ_W=90°. Saturation value of ξ corresponds to most unstable mode $k_m=k_c/\sqrt{2}=\sqrt{\nu_2/2\nu_4}$.

To get a better understanding of the processes determining the evolution of the surface roughness after the initial stages of island nucleation and coalescence, we have carried out simulations using a simplified linear equation which includes two competing terms describing the local growth rate—a negative surface-tension term ($\nu_2\nabla^2 h$) which takes into account the combined effects of atomic shadowing[20] and evaporation/condensation, and a surface-diffusion term[18] ($\nu_4\nabla^4 h$) which tends to smooth the surface on short length scales. Since it is difficult to accurately model the process of island nucleation and coalescence, as an initial configuration in our simulations we have assumed a random distribution of 3D spherical caps, with a dilution-ratio-independent initial island density while the average island-radius and standard deviation were chosen to match the initial experimental roughness at small film thickness ($\bar{h}\simeq 10-20$ Å). Since the experimental wetting angle θ_W was unknown, we have carried out simulations with two different possible values, e.g., θ_W=45° and 90°. The values of the parameters ν_2 and ν_4 were then varied to obtain the best possible agreement with experiment.

For the case of large dilution ratio (R_H=40 and 60), we find very good agreement between our simulation results and experimental results for the surface roughness for *all* thicknesses up to 3000 Å. We note that better agreement is obtained for the case of a large wetting angle (θ_W=90°) than for a small wetting angle (θ_W=45°). It is worth noting that this is consistent with experiments on liquid Si droplets on SiO_2 substrates[21] in which a wetting angle of 90° was observed.

Our results also suggest that, at least for large dilution ratios, the dominant processes determining the evolution of the surface roughness may be described by two main effects—an instability due to atomic shadowing (modified by attachment/detachment) and smoothing due to surface diffusion. In this connection, we note that the value of the parameter ν_4 found in our simulations increases rapidly with increasing R_H. This is consistent with the expectation that increasing the dilution ratio will lead to a decreased density of trapping sites and an increased diffusion length. Similarly, we find that ν_2 depends relatively weakly on the dilution ratio. In addition, at least for the case of large wetting angle, the magnitude of ν_2, e.g., 0.2–0.6 Å, is significantly less than expected using the Mazor-Srolovitz prediction $\nu_2=-\delta$, where δ is the radius of the SiH_3 adsorbate. This is consistent with our expectation that the destabilizing effects of atomic shadowing are compensated by the effects of detachment, evaporation, or re-emission[22] thus reducing the magnitude of ν_2.

It is also worth noting that the short-range attraction of depositing particles to the substrate may also play a role in enhancing this instability,[15,23] although we would expect the effects of attraction to be relatively weak, given the relatively high kinetic energies of depositing particles in PECVD. In this connection, we note that the slope on a log-log plot of

the experimental surface roughness as a function of film thickness (not shown) is significantly larger than 1 for large film thickness and high dilution ratio (R_H=40 and 60). While this is consistent with our model (since the negative surface-tension term is expected to lead to exponential growth of the surface roughness at large thicknesses) it is not consistent with a model which only includes global shadowing since this leads to a roughening exponent $\beta=1$ at late times.[22]

As already noted, the good agreement between our simulation results and experiments for high dilution ratio indicates that surface diffusion plays an important role in the evolution of the surface roughness in low-temperature PECVD. This behavior is consistent with the results of Sperling and Abelson[19] who measured the structure factor $S(q)$ for a-Si:H films grown via hot-wire CVD and found a q^{-4} dependence which is typical of surface diffusion. However, this raises the question of what is diffusing or perhaps more precisely what is the dominant diffusing species? In this connection, we note that recent atomistic kinetic Monte Carlo models based on first-principles calculations[2,3] which have been able to account for the dependence of the degree of crystallinity on temperature and dilution ratio do not include the effects of diffusion. This suggests that while diffusion may play an important role in determining the surface roughness, it may be less important for the nucleation and growth of nanocrystals.

We also find relatively good agreement between our simulations and experiments for low hydrogen dilution ratio (R_H=15 and 20) for thicknesses which are not too large, e.g., up to the point (e.g., "amorphous-to-amorphous transition"[8]) where the roughness starts increasing rapidly. However, in this case the rate of increase of the experimentally measured roughness decreases at large thicknesses. One possibility is that some new physical mechanism comes into play at large thicknesses for small dilution ratios which suppresses the rapid increase in roughness. However, another possibility is that the development of large slopes at large film thicknesses implies that the linearized Eq. (2) cannot be used but instead the fully nonlinear Eq. (1) should be used.

In order to determine if this is the case, we have examined the distribution of surface slopes $F(m)$ (where m is slope magnitude) at large thicknesses for both small and large dilution ratios. As can be seen in Fig. 6, for large dilution ratio the slopes are typically significantly less than 1 even at large film thicknesses. In contrast, for small dilution ratio significantly larger slopes on the order of 1 or larger are observed in our simulations. This suggests that for small dilution ratios and large film thicknesses, it may be necessary to use the nonlinear form of Eq. (1). In this connection, we note that the strength of the instability term $(-A_1M)$ in Eq. (1) decreases with increasing slope $|\nabla h|$ and $g=1+(\nabla h)^2$, thus possibly explaining this effect. On the other hand, the nonlinear surface-diffusion term $-A_3 g\Delta_s M$ *also* decreases with increasing g while the stabilizing attachment/detachment term $A_2\sqrt{g}M$ is also reduced but less significantly. Therefore, numerical integration of the fully nonlinear Eq. (1) will be needed to determine if the differences for small dilution ratio and large film thicknesses can be explained by these nonlinear effects.

Besides the nonlinearities inherent in Eq. (1), which may be important for small dilution ratios and large film thicknesses, another important issue is the initial configuration. Since in our simulations it was found that the surface roughness evolution is quite sensitive to the initial configuration, it would be desirable to directly model the early stages of island nucleation and coalescence. While a full atomistic simulation of a-Si growth is difficult because of the complexity of the PECVD process, we expect that in the future, simulations of a simplified atomistic model may allow us to obtain a better understanding of the thin-film morphology during the nucleation regime.

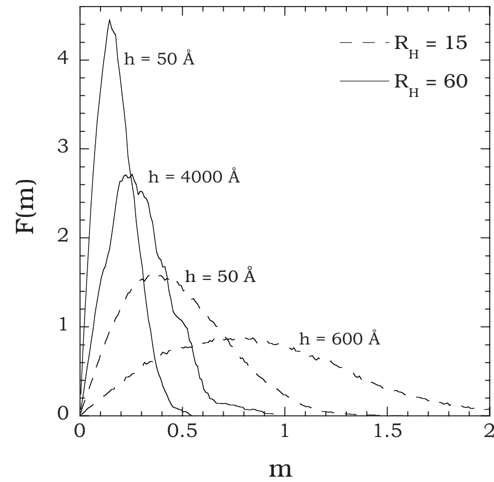

FIG. 6. Distribution $F(m)$ of surface slopes $m(\theta_W=90°)$ for R_H=60 (solid lines) and R_H=15 (dashed lines).

We now consider some possible extensions or modifications to the simplified continuum model presented here. In particular, we note that Raible *et al.*[16,24] have suggested a number of additional terms to describe amorphous thin-film growth. These include a Kardar-Parisi-Zhang (KPZ)-type term[25] proportional to $|\nabla h|^2$ which may be related to the dependence of the film density on slope, as well as a "conserved KPZ term" (proportional to $\nabla^2|\nabla h|^2$) due to the slope dependence of the local density of diffusing species.[26,27] By adding terms of this form and also including the effects of finite scanning-tunneling microscopy tip size, Raible *et al.*[15] were able to quantitatively explain the experimentally observed saturation of the surface roughness in amorphous ZrAlCu thin films grown by physical vapor deposition. In the future, it may be of interest to consider the addition of similar nonlinear terms to Eq. (1) since these may become important at large thicknesses and for low R_H.

Another effect which has not been directly considered in our simulations is the effect of *global* shadowing, which may occur when atoms are deposited at an angle with respect to the substrate. Such an effect may be expected in PECVD due to the fact that deposition takes place in a plasma in which the radicals are traveling in random directions before hitting the surface, thus leading to shadowing of valleys by hills. While this provides another possible explanation for the instability term $-\nu_2\nabla^2 h$ in Eq. (2), shadowing is a nonlocal effect and therefore cannot be fully described simply by including this term. Therefore, it may also be of interest to carry out simulations in which shadowing and/or re-emission are directly taken into account.

In conclusion, using RTSE we have measured the evolution of the surface roughness in low-temperature (200 °C) and high-pressure (3 mTorr) a-Si:H growth via PECVD for values of the hydrogen dilution ratio ranging from $R_H=15$ to $R_H=60$. In general, we find that the surface roughness decreases with increasing R_H. In addition, our results indicate that the growth process may be divided into several distinct regimes including a regime corresponding to growth at approximately constant roughness as well as a rapid increase in roughness for large thicknesses. To model the experimentally observed roughness evolution we have used a simplified linearized continuum equation which takes into account the destabilizing effects of atomic shadowing and/or attraction as well as the smoothing effects of surface diffusion. By varying the initial configuration as well as the parameters ν_2 and ν_4, we have found good agreement with experiment in the case of large dilution ratios ($R_H=40$ and $R_H=60$). The good agreement between our simulations and experiments at high dilution ratio indicates that surface diffusion plays an important role. However, there is disagreement at large thicknesses for small dilution ratios ($R_H=15$ and $R_H=20$) when surface slopes m become larger than 1. Further work will be needed to take into account a variety of nonlinear effects which may affect the surface roughness in this case, as well as to understand the atomistic mechanisms of a-Si:H surface growth.

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[1] R. W. Collins, A. S. Ferlauto, G. M. Ferreira, Chi Chen, Joohyun Koh, R. J. Koval, Yeeheng Lee, J. M. Pearce, and C. R. Wronski, Sol. Energy Mater. Sol. Cells **78**, 143 (2003).
[2] X. Tan and G. W. Yang, Appl. Phys. Lett. **93**, 061902 (2008).
[3] P. L. Novikov, A. Le Donne, S. Cereda, Leo Miglio, S. Pizzini, S. Binetti, M. Rondanini, C. Cavallotti, D. Chrastina, T. Moiseev, H. von Kanel, G. Isella, and F. Montalenti, Appl. Phys. Lett. **94**, 051904 (2009).
[4] K. Tanaka, *Glow-discharge Hydrogenated Amorphous Silicon* (Kluwer, Boston, 1989).
[5] G. Bruno, Pio Capezzuto, and Arun Madan, *Plasma Deposition Processes of Amorphous Silicon-Based Materials* (Academic, Boston, 1995).
[6] J. R. Abelson, Appl. Phys. A **56**, 493 (1993).
[7] We note that in the atomistic simulations of Refs. 2 and 3 only a minimal amount of roughness was observed.
[8] N. J. Podraza, C. R. Wronski, and R. W. Collins, J. Non-Cryst. Solids **352**, 950 (2006).
[9] I. An, J. A. Zapien, C. Chen, A. S. Ferlauto, A. S. Lawrence, and R. W. Collins, Thin Solid Films **455-456**, 132 (2004).
[10] I. An, H. V. Nguyen, N. V. Nguyen, and R. W. Collins, Phys. Rev. Lett. **65**, 2274 (1990).
[11] I. An, Y. M. Li, H. V. Nguyen, and R. W. Collins, Rev. Sci. Instrum. **63**, 3842 (1992).
[12] H. Fujiwara, J. Koh, P. I. Rovira, and R. W. Collins, Phys. Rev. B **61**, 10832 (2000).
[13] J. Koh, Y. Lu, C. R. Wronski, Y. Kuang, R. W. Collins, T. T. Tsong, and Y. E. Strausser, Appl. Phys. Lett. **69**, 1297 (1996).
[14] A. Mazor, D. J. Srolovitz, P. S. Hagan, and B. G. Bukiet, Phys. Rev. Lett. **60**, 424 (1988).
[15] M. Raible, S. G. Mayr, S. J. Linz, M. Moske, P. Hanggi, and K. Samwer, Europhys. Lett. **50**, 61 (2000).
[16] M. Raible, S. J. Linz, and P. Hanggi, Phys. Rev. E **62**, 1691 (2000).
[17] F. Family and J. G. Amar, Fractals **1**, 753 (1993).
[18] W. W. Mullins, J. Appl. Phys. **28**, 333 (1957).
[19] B. A. Sperling and J. R. Abelson, Appl. Phys. Lett. **85**, 3456 (2004).
[20] A. Mazor, B. G. Bukiet, and D. J. Srolovitz, J. Vac. Sci. Technol. A **7**, 1386 (1989).
[21] H. Kanai, S. Sugihara, H. Yamaguchi, T. Uchimaru, N. Obata, T. Kikuchi, F. Kimura, and M. Ichinokura, J. Mater. Sci. **42**, 9529 (2007).
[22] J. T. Drotar, Y. P. Zhao, T. M. Lu, and G. C. Wang, Phys. Rev. B **62**, 2118 (2000).
[23] Y. Shim, V. Borovikov, and J. G. Amar, Phys. Rev. B **77**, 235423 (2008).
[24] M. Raible, S. J. Linz, and P. Hanggi, Phys. Rev. E **64**, 031506 (2001).
[25] M. Kardar, G. Parisi, and Y. C. Zhang, Phys. Rev. Lett. **56**, 889 (1986).
[26] We note that an additional nonlinear term proportional to the Gaussian curvature $K_G=[(h_{xx}h_{yy}-h_{xy}^2)\ /[1+(\nabla h)^2]$ has been suggested in Ref. 15 due to the effects of attraction of incoming particles to the film and has also recently been suggested on more general grounds by Escudero (Ref. 27). However, the coefficient of this term is expected to be relatively small (Ref. 15).
[27] C. Escudero, Phys. Rev. Lett. **101**, 196102 (2008).