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Linear growth of thin films under the influence of stress

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We have studied the growth of thin films in the presence of stress instability that enhances the roughness and roughening induced by conservative as well as nonconservative noise. It is clearly illustrated that nonconservative noise effects may enhance stress induced roughness. Nevertheless, the incorporation of conservative noise appears to also be substantial in growth processes driven by diffusion. For growth on a rough substrate the dependence of the amplitude of the surface roughness on the film thickness differs from that of a film growing on a flat substrate. The amplitude shows a minimum at a particular substrate thickness, which indicates that the growth up to this thickness is enforced by undulations of the substrate.
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In thin film technology the control of roughness induced by growth is of considerable importance because surface and interface roughness influences many physical properties, e.g., thermal, electrical and magnetic.[1–5] In many cases the growth of thin films occurs on substrates with different lattice parameters (heterogrowth), which imposes besides kinetic effects[6–11] additional constraints on the mode of film growth due to the development of stress.[10] In general, the morphology of the film surface will be the result of the competition between noise induced roughening, possibly step-edge barrier induced roughening, surface relaxation mechanisms, lateral growth nonlinearities,[7] as well as stress development at the film/substrate interface.

A lattice mismatch of 1% can easily lead, without plastic relaxation, to a stress level of the order of GPa (e.g., in InGaAs/GaAs). This effect becomes even more dramatic for nanometer scale system dimensions ($\sim$10 nm) where the contributions of surface tension are important.[10] The film may release stress by the creation of additional surface roughness to an extent that depends also on the possible surface relaxation mechanism. Indeed, linear stability analysis has shown that the nominally flat surface of an elastically stressed body is unstable with respect to growth of perturbations with a wavelength larger than a certain critical wavelength.[10] However, up to now there has been only scant research available on the properties of thin film growth in the presence of both stress and noise induced roughening effects.

In this work we concentrate on growth processes under the influence of stress for coherent film/substrate interfaces and materials that do not differ too much in elastic properties. Surface relaxation will be considered by surface diffusion which is a noisy process and thus contributes a noise term (so-called conservative noise) that obeys the fluctuation-dissipation theorem, in addition to the so-called nonconservative noise that is present in the beam of depositing adatoms.[11] The growth process will be described by linear Langevin dynamics that allow direct calculation of relevant roughness parameters.

If surface diffusion is the predominant mechanism of surface relaxation of the incoming adatoms on the surface, the growth front $h(r,t)$ ($\langle h(r,t)\rangle=0$) for weak roughness ($|\nabla h|\ll 1$) evolves according to[6,12]

$$\frac{\partial h(\mathbf{r},t)}{\partial t}=-C\gamma\nabla^4 h-(C/2M)\nabla^2\{[\sigma_{tt}(h)]^2-\sigma^2\}+\eta(\mathbf{r},t)+n_D(\mathbf{r},t). \quad (1)$$

The term $-C\gamma\nabla^4 h$ represents surface diffusion due to the curvature induced chemical potential gradient. $C=D_s\Omega^2\delta/k_BT$, with D_s the surface diffusion coefficient, T the substrate temperature, Ω the atomic volume, δ the number of atoms per unit area, γ the interfacial tension, and R the deposition rate. $\eta(r,t)$ represents a nonconservative Gaussian white noise of amplitude $D(<R)$ due to the deposition process with $\langle\eta(r,t)\rangle=0$ and $\langle\eta(\mathbf{r},t)\eta(\mathbf{r}',t')\rangle=2D\delta(\mathbf{r}-\mathbf{r}')\delta(t-t')$.[7,11] $\eta_D(\mathbf{r},t)$ is a conservative noise due to surface diffusion with $\langle\eta_D(\mathbf{r},t)\rangle=0$ and $\langle\eta_D(\mathbf{r},t)\eta_D(\mathbf{r}',t')\rangle=2k_s\nabla^2\delta(\mathbf{r}-\mathbf{r}')\delta(t-t')$.[7,11] The term $(C/2M)\times\nabla^2\{[\sigma_{tt}(h)]^2-\sigma^2\}$ (Ref. 10) is due to stress on the growing film because of film/substrate lattice mismatch. Subscript t indicates the tangential component to the surface of the stress field. M is the elastic modulus, and σ the mean stress of the growing film. A free surface is traction free along its normal direction with stress components $\sigma_{nn}=\sigma_{tn}=0$ with subscript n indicating the local direction normal to the surface. Perturbation analysis for a sinusoidal profile of wave vector $\mathbf{q}$ yields for weak roughness ($|\nabla h|\ll 1$) $\times(\Omega/2M)\{[\sigma_{tt}(h)]^2-\sigma^2\}=(2\Omega\sigma^2/M)q\sin(\mathbf{q}\cdot\mathbf{r})$.[12]

Therefore, the solution of Eq. (1) is straightforward through Fourier transformation, $h(\mathbf{r},t)=(1/2\pi)\int e^{i\mathbf{q}\cdot\mathbf{r}}d^2\mathbf{q}\times\int_0^t[\Theta(\mathbf{q},\tau)+\Theta_D(\mathbf{q},\tau)]e^{-[C\gamma q^4-(2C\sigma^2/M)q^3](t-\tau)}d\tau$,[13] which yields the roughness spectrum of the growing surface front,

$$\langle|h(\mathbf{q},t)|^2\rangle=\left(\frac{1}{2\pi}\right)\frac{4(D+q^2D_s)}{[C\gamma q^4-(2C\sigma^2/M)q^3]}\times(1-e^{-2[C\gamma q^4-(2C\sigma^2/M)q^3]t}), \quad (2)$$

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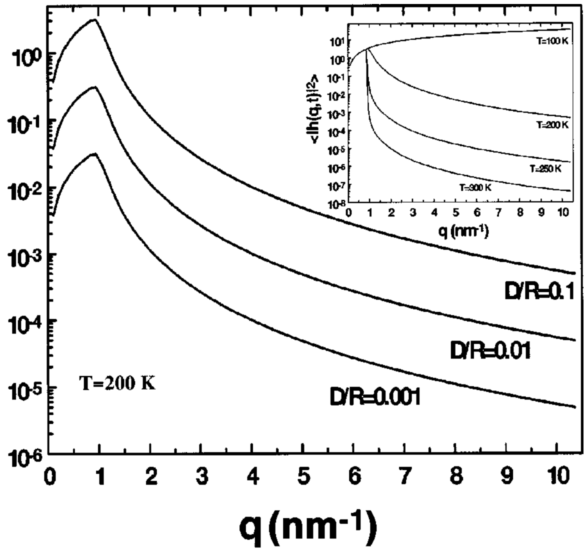

FIG. 1. Calculations of $\langle |h(\mathbf{q},t)|^2\rangle$ from Eq. (2) vs wave vector $\mathbf{q}$ for various nonconcervative noise ratios D/R and $E=0.5$ eV, $t=30$ s. The inset shows $\langle |h(\mathbf{q},t)|^2\rangle$ vs q for various substrate temperatures T, $D/R=0.1$, $t=30$ s, and $E=0.5$ eV.

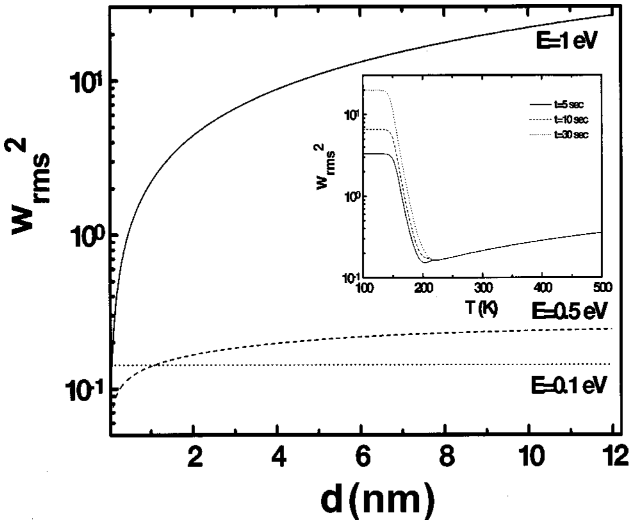

FIG. 2. $w_{\rm rms}$ vs film thickness $d(=Rt)$ for various activation energy barriers E, $D/R=0.1$, and $T=300$ K. The inset shows $w_{\rm rms}$ vs substrate temperature T for various growth times t, $D/R=0.1$, $E=0.5$ eV.

using the noise transforms $\Theta_D(q,t)=(1/2\pi)\times\int\eta_D(\mathbf{r},t)e^{-i\mathbf{q}\cdot\mathbf{r}}d\mathbf{r}$, $\langle\Theta(\mathbf{q},t)\Theta(\mathbf{q}',t')\rangle=2D\delta(\mathbf{q}+\mathbf{q}')\delta(t-t')$, $\langle\Theta_D(\mathbf{q},t)\Theta_D(\mathbf{q}',t')\rangle=2D_sq^2\delta(\mathbf{q}+\mathbf{q}')\delta(t-t')$, and $\langle\Theta(\mathbf{q},t)\rangle=\langle\Theta_D(\mathbf{q},t)\rangle=0$.[13,14]

Our calculations were performed for a film of modulus $M=147$ GPa, mean stress $\sigma=5.8$ GPa, interface tension $\gamma=0.5\ \mathrm{J/m^2}$,[10] atomic spacing $c=0.3$ nm, $\Omega=c^3$, $\delta=1/c^2$, and an average deposition rate $R=0.3$ nm/s (the film thickness is $d=Rt$) such that $R>D$.[13] Although under equilibrium conditions the noise amplitude D behaves as $D\propto\sqrt{R}$; for far from equilibrium growth the relationship between D and R is more complex.[13] For D_s we assumed $D_s=(10^{-6}\ \mathrm{m^2/s})\exp(-E/k_BT)$ with E a diffusion activation barrier. We omit any temperature dependence of the average stress, σ, because we consider relatively low substrate temperatures during the film growth. As Eq. (2) indicates, the system will experience unstable growth for roughness wavelengths larger than $L=\pi\gamma M/\sigma^2$ which yields for these parameters $L=6.86$ nm.

We now discuss growth on a flat substrate. Because $\langle |h(\mathbf{q},t)|^2\rangle\sim D$, the roughness amplitude will increase significantly with increasing noise amplitude D, indicating the importance of including noise effects in the growth process (Fig. 1). Clearly, noise effects enhance the formation of roughness due to stress instability. Moreover, at low temperatures where surface diffusion is minimal, the roughness spectrum $\langle |h(\mathbf{q},t)|^2\rangle$ increases monotonously with $\mathbf{q}$ over the natural range of wave vectors $0<\mathbf{q}<\mathbf{q}_c(=\pi/c)$ (inset of Fig. 1). It decreases for wave vectors $\mathbf{q}>\mathbf{q}_L(=2\pi/L)$ at an increasing rate with increasing substrate temperature.

Furthermore, from Eq. (2) we can calculate the root mean square (rms) roughness amplitude $w_{\rm rms}$, which is defined by $w^2_{\rm rms}=(2\pi)\int_{0<q<q_c}\langle |h(\mathbf{q},t)|^2\rangle\mathbf{q}d\mathbf{q}$. Figure 2 shows $w_{\rm rms}$ versus film thickness $d(=Rt)$ for various diffusion energy barriers E. As the energy barrier E increases and thus diffusion becomes less predominant the roughening induced by the presence of stress predominates the growth mode. In this case, $w_{\rm rms}$ increases with film thickness rather fast (solid line, Fig. 2). However, for low energy barriers E (fast diffusion), $w_{\rm rms}$ is small and dominated by thermal noise fluctuations due to the diffusion process (dotted line, $E=0.1$ eV). Similar is the situation with increasing substrate temperature T (inset of Fig. 2). Indeed, $w_{\rm rms}$ is larger with increasing deposition time at low temperatures, while at higher temperatures (for the parameters used) all the curves collapse and increase with increasing temperature (the thermal or diffusion noise effect).

In the absence of conservative noise the roughness amplitude will continuously decrease with increasing substrate temperature (Fig. 3). Moreover, with increasing amplitude D of the nonconservative noise (inset of Fig. 3), the roughness amplitude increases at low temperatures. The transition to a thermally dominated regime occurs with the presence of a minimum, which is more pronounced as D decreases. Actually, the transition shifts toward lower substrate tempera-

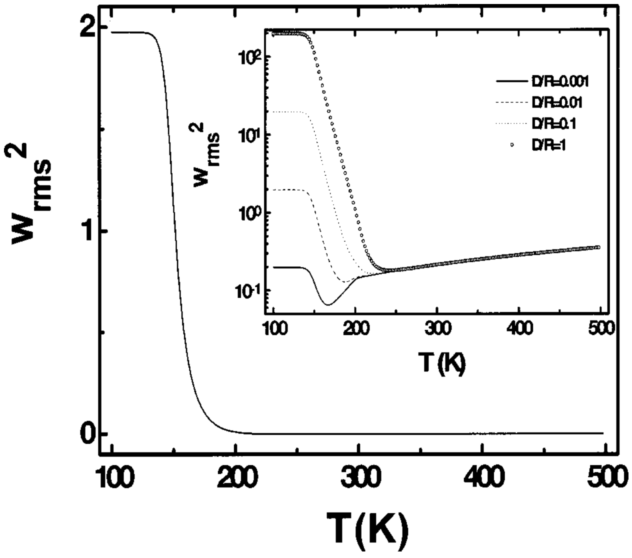

FIG. 3. $w_{\rm rms}$ vs substrate temperature without conservative diffusional noise, $D/R=0.01$, $t=30$ s, $E=0.5$ eV. The inset shows $w_{\rm rms}$ vs substrate temperature T for various nonconservative noise amplitudes D, $t=30$ s, $E=0.5$ eV.

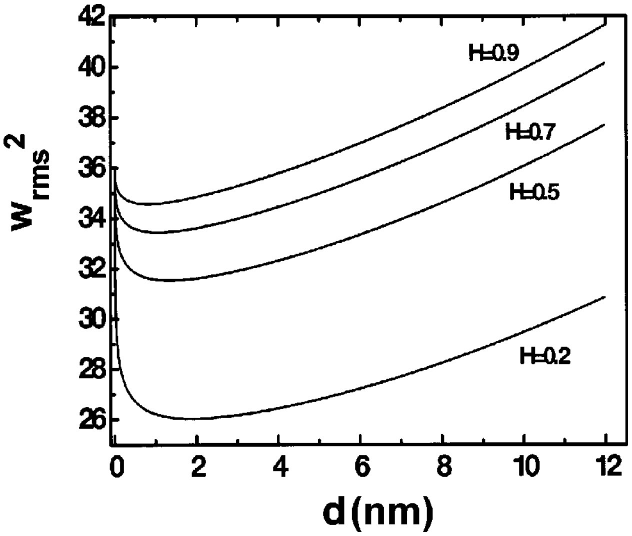

FIG. 4. $w_{\rm rms}$ vs film thickness d for various substrate roughness exponents H, substrate correlation length $\xi = 50$ nm, and substrate rms amplitude $w = 0.5$ nm. The other parameters are $E = 0.5$ eV, $D/R = 0.1$, and $T = 200$ K.

tures. Therefore, the effect of conservative noise is rather distinct in the roughening growth front, and its inclusion appears to be substantial in a diffusive growth process.

Next we discuss growth on rough substrates. If growth commences on a rough substrate with roughness spectrum $\langle |h_s(\mathbf{q},t)|^2 \rangle$, the term $\langle |h_s(\mathbf{q},t)|^2 \rangle e^{-2[Cyq^4-(2C\sigma^2/M)q^3]t}$ in Eq. (2) should be considered. For the sake of simplicity we shall consider the case of a self-affine substrate roughness to model substrate deviations from flatness. This type of rough morphology is described by a rms roughness amplitude w, an in-plane correlation length ξ, and a roughness exponent $H(0<H<1)$. These parameters quantify the details of the roughness at short wavelengths $(<\xi)$ such that as H becomes smaller the surface becomes more irregular. $\langle |h_s(\mathbf{q})|^2 \rangle$ is modeled by a simple form,[15] $\langle |h_s(\mathbf{q})|^2 \rangle = (1/2\pi)[w^2\xi^2/(1+aq^2\xi^2)^{1+H}]$ with $a = 1/2H[1-(1+aq_c^2\xi^2)^{-H}]$. As Fig. 4 shows the dependence of the surface roughness amplitude on film thickness differs from that of a film growing on a flat substrate (Fig. 2). The amplitude shows a minimum at a particular substrate thickness, which indicates that the growth up to this thickness is enforced by undulations of the substrate. The initial decrease of the roughness is governed by conservative noise roughening.[16,17] The behavior is also similar for increasing correlation lengths ξ.

In conclusion, we studied the growth of a thin film in the presence of stress instability and noise induced roughening. It is illustrated that nonconservative noise can enhance stress induced roughness. Conservative noise appears to have a substantial effect in the growth process driven by surface diffusion. A precise understanding of stress influences on film growth requires the inclusion of nonlinear growth aspects and stress release by dislocation formation at the film/substrate interface.[18]

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