Stochastic Field Equation for Amorphous Surface Growth

Stefan J. Linz, Martin Raible, and Peter Hänggi

Theoretische Physik I, Institut für Physik, Universität Augsburg,
86135 Augsburg, Germany

Abstract. A minimal stochastic field equation aimed at modeling the amorphous surface growth generated by physical vapor deposition is derived, analyzed, and related to the underlying microscopic mechanisms.

1 Introduction and Basics

Since the formation and growth of interfaces are ubiquitous in nature and in a variety of technological applications, the theoretical understanding of their kinetics on a microscopic as well as on a mesoscopic level constitutes one of the major challenges in physics and material science [1–3]. In particular, the growth of solid amorphous films generated by physical vapor deposition that is important e.g. in the context of coating and the manufacturing of thin glassy ZrAlCu films has recently attracted interest [4–8] in material science. The focus of this contribution is to develop and to analyze a minimal model in form of a stochastic field equation that is appropriate to describe the spatio-temporal evolution of amorphous surface growth processes.

In its most elementary version, the generic setup of such a surface growth process, cf. also Fig. 1, consists of an initially almost flat substrate and a vapor particle beam being determined by the deposition flux and directed perpendicular to the substrate. In vapor deposition [4–6], the deposition flux is (i) typically low-energetic implying that no kick-off of surface particles occurs and (ii) basically constant in space and time with some weak super-imposed spatio-temporal fluctuations originating from the particle source. The particles from the beam are deposited at the surface and undergo various surface diffusion processes until they arrive at their final position. With time, this creates a growing layer build up by the deposited particles. The surface of the layer is characterized by its *height* or *morphology* $H(x, t)$ at time t and at the substrate location $x = (x, y)$. Microscopically speaking, the evolution of the surface morphology results from the only partly explored interaction of particles to be deposited at the surface and the already condensed particles. As experiments reveal [4–6], however, the spatio-temporal evolution of the surface morphology on a mesoscopic scale can be interpreted as the interplay of three different mechanisms: roughening, smoothening and pattern formation.
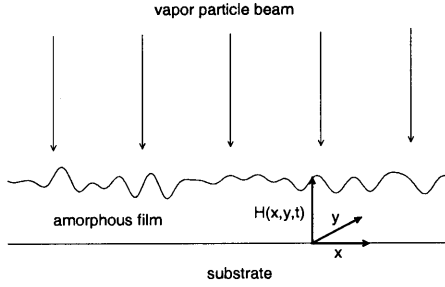
Fig. 1. Sketch of a vapor deposition experiment for amorphous thin film growth

Modern experimental investigation tools such as scanning tunneling microscopy combined with image processing allow for a detailed resolution of the surface morphology and its spatio-temporal evolution [4–6]. The obtained data set, however, is too immense and the data also contain some degree of stochasticity due to the small deposition noise resulting from the particle source. Therefore, an appropriate quantitative statistical measure of the information on height variations and lateral correlations is given by the *height-height-correlation* function

$$C(r,t) = \left\langle \left\langle [H(x+r,t) - \langle H \rangle_x][H(x,t) - \langle H \rangle_x] \right\rangle_\eta \right\rangle_{x,|r|=r} . \qquad (1)$$

Here, $\langle ... \rangle_\eta$ denotes an average over different samples (ensemble average), $\langle ... \rangle_x = L^{-2} \int_0^L d^2x...$ the spatial average over a sample area of size L^2, and $\langle H \rangle_x = \langle H \rangle_x(t) = \langle H(x,t) \rangle_x$ the spatially averaged surface profile at time t. From $C(r,t)$ the two most important global quantities that characterize the surface morphology can be determined: (i) The *correlation length* $R_c(t)$ that is given by the first maximum of $C(r,t)$ for non-zero r, i.e.

$$R_c(t) = \min\{r > 0 \mid \partial_r C(r,t) = 0, \ \partial_r^2 C(r,t) < 0\}, \qquad (2)$$

and, therefore, determines the typical length scale over which height fluctuations are correlated, and (ii) the *surface roughness* or root mean square deviation of the relative height fluctuations $w(t)$ that is determined by the $r = 0$ limit of $C(r,t)$,

$$w^2(t) = C(0,t). \qquad (3)$$

Any successful modeling attempt of the spatio-temporal evolution of $H(x,t)$ requires at least the validation of the temporal evolution of $R_c(t)$ and $w(t)$ in comparison to the experimental data.

2   Functional Form of the Growth Equation

As a first step towards a theoretical modeling, we use the machinery of stochastic field equations as a tool for the understanding and interpretation of the growth dynamics. The basic philosophy behind this approach is

to disregard the microscopic details of the particle interaction and to consider the growth process on a slighty larger length scale where the surface morphology can be treated in form of a continuum variable. Primary focus is the functional form of an appropriate evolution equation for $H(x,t)$. Therefore, the general starting point is given by an ansatz of the form

$$\partial_t H = G[H, \nabla H, x, t] + I(x,t) \tag{4}$$

where $I(x,t)$ represents the deposition flux and the functional $G[H, \nabla H, x, t]$ comprises all physical mechanisms leading to growth and relaxational processes on the surface. Such an approach, however, is implicitly limited by the fact that the surface morphology $H(x,t)$ is uniquely defined (single-valued in x), i.e. that overhangs in the surface morphology do not appear. In vapor deposition experiments [4–6], the deposition flux is basically constant with some small superimposed stochasticity resulting from the particle source. Therefore, the deposition flux can be split into a spatio-temporally constant mean deposition flux F and a fluctuating part

$$I(x,t) = F + \eta(x,t). \tag{5}$$

Here $\eta(x,t)$ represents spatio-temporal Gaussian white noise determined by

$$\langle \eta(x,t) \rangle_\eta = 0 \tag{6}$$
$$\langle \eta(x,t)\, \eta(x',t') \rangle_\eta = 2\, D\, \delta(x - x')\, \delta(t - t') \tag{7}$$

where $\langle ... \rangle_\eta$ denotes the ensemble average. Further simplification of (4) can be obtained by invoking standard symmetry requirements [2]: (i) *invariance under translation in time, $t \to t + \tau$*, since the form of (4) should be independent of the choice of the origin of time and (ii) *invariance under translation in the direction perpendicular to the growth direction, $x \to x + l$*, to exclude dependences on the choice of the origin of the coordinate system at the substrate. These two invariances exclude the appearance of an explicit dependence on the time t and the spatial position x in G, respectively.

Moreover, *invariance under translation in growth direction, $H \to H + z$*, must also be considered since (4) should be independent on the choice of the origin of the H-axis. This symmetry directly excludes the appearance of $H(x,t)$ in G. Therefore, only gradients and higher order spatial derivatives of $H(x,t)$ can enter in the functional G. By the way, the presence of the latter symmetry also distinguishes surface growth equations from other pattern forming equations such as the Ginzburg-Landau equation in the context of convection.

Since the mean deposition flux F is constant it proofs useful to introduce the height profile $h(x,t) = H(x,t) - Ft$ in the frame comoving with the velocity F. Then, (4) simplifies to

$$\partial_t h = G[\nabla h] + \eta(x,t). \tag{8}$$

Due to the isotropy of the amorphous phase, *invariance under rotation and reflection* in the plane perpendicular to the growth direction must also be regarded. This rules out any odd derivatives of h in G and implies that ∇-operators entering the various contributions in G must be multiplied in couples by scalar multiplication.

Assuming that all surface relaxation processes are local, we finally expand the functional G in a power series in all possible spatial derivatives of h and keep only the terms that are linear or quadratic in h and only possess a maximum of four ∇-operators. As a result of the afore-mentioned symmetries, the deterministic part of (8) can only consist of the terms $\nabla^2 h$, $(\nabla h)^2$, $\nabla^4 h$, $\nabla^2 (\nabla h)^2$, $(\nabla^2 h)^2$, and $\nabla \cdot [(\nabla h)(\nabla^2 h)]$. The last term can be slightly rearranged in the form

$$\nabla \cdot [(\nabla h)(\nabla^2 h)] = \frac{1}{2}\nabla^2 (\nabla h)^2 + 2M \tag{9}$$

with

$$M = \det \begin{pmatrix} \partial_x^2 h & \partial_y \partial_x h \\ \partial_x \partial_y h & \partial_y^2 h \end{pmatrix}, \tag{10}$$

so that effectively only the functionally different terms $\nabla^2 h$, $(\nabla h)^2$, $\nabla^4 h$, $\nabla^2 (\nabla h)^2$, $(\nabla^2 h)^2$, and M appear in G. Consequently, the functional form of the growth equation for amorphous films is given explicitly by

$$\partial_t h = a_1 \nabla^2 h + a_2 \nabla^4 h + a_3 \nabla^2 (\nabla h)^2 + a_4 (\nabla h)^2 + a_5 (\nabla^2 h)^2 + a_6 M + \eta. \tag{11}$$

The approach of determining the leading order functional form of the growth equation does not reveal the sign of the coefficients a_i, $i = 1,..,6$, and the physical significance of the corresponding terms in (11). Focussing on the physics of amorphous growth, the coefficients a_i will be connected to the underlying microscopic processes in the next section.

Several remarks are in order.

(i) Equation (11) constitutes a *systematic* expansion of (4) derived by use of the afore-mentioned symmetry arguments and by taking into account all possible combinations up to $O(\nabla^4)$ and $O(h^2)$. It consists of two linear terms and four nonlinear terms in h. The term being proportional to a_6 becomes zero in the one-dimensional limit. This shows the principal problem that one-dimensionally motivated surface growth equations cannot be carried over to the two-dimensional case by simply replacing $\partial_x \to \nabla$. Moreover, it is interesting to note that Lai and Das Sarma [9] have also attempted to derive the leading order functional form of a growth equation using isotropy and the fact that the functional G in (8) is a scalar. Their result, however, significantly differs from (11) since the terms $a_5 (\nabla^2 h)^2$ and $a_6 M$ are missing. Therefore, we conclude that Lai and Das Sarma's growth equation [9] represents an inconsistent systematic expansion since the terms $(\partial_x^2 h)(\partial_y^2 h) - (\partial_x \partial_y h)^2$ and $(\partial_x^2 h)^2 + (\partial_y^2 h)^2 + 2(\partial_x^2 h)(\partial_y^2 h)$ are not properly taken into account.

(ii) The growth equation (11) contains several known limiting cases. The limit $a_i = 0$ for $i = 1, .., 6$, $\partial_t h = \eta$, is considered as an appropriate model for random deposition [2]. Setting $a_i = 0$ for $i = 2, .., 6$, $\partial_t h = a_1 \nabla^2 h + \eta$, yields the Edwards-Wilkinson (EW) equation originally motivated in the context of granular systems [10]. The limit $a_i = 0$ for $i = 2, 3, 5, 6$, determines the Kardar-Parisi-Zhang (KPZ) equation, $\partial_t h = a_1 \nabla^2 h + a_4 (\nabla h)^2 + \eta$, being the paradigm for a stochastic roughening process [11]. Finally, the limit $a_i = 0$ for $i = 3, 5, 6$, $\partial_t h = a_1 \nabla^2 h + a_2 \nabla^4 h + a_4 (\nabla h)^2 + \eta$, leads to the stochastic version of the Kuramoto-Sivashinsky equation.

(iii) Besides the invariances already invoked for its derivation, the growth equation (11) possesses an interesting additional symmetry: It remains invariant under the combined transformation

$$\{h, a_3, a_4, a_5, a_6\} \rightarrow \{-h, -a_3, -a_4, -a_5, -a_6\}. \tag{12}$$

As a consequence, one has to expect that a simultaneous change of the sign of the coefficients a_3, a_4, a_5, a_6 only leads to an inversion of the surface profile $h(x, t)$ about $h = 0$. Note, however, that (11) does *not* possess mirror symmetry about $h = 0$, i.e. it does not fulfil the up/down invariance $h \rightarrow -h$ (without inversion of the signs of the nonlinear coefficients). This already implies some degree of asymmetry of the resulting surface profile $h(x, t)$.

(iv) In the context of the related molecular beam epitaxy, a frequently invoked further requirement on surface growth equations [2, 3] is that the functional G should be represented by the divergence of a surface current, $G = -\nabla \cdot j_G(\nabla h)$ since no desorption of particles occurs. Such an assumption directly rules out the appearance of a KPZ term $(\nabla h)^2$ and the term $(\nabla^2 h)^2$ in G. It also implies that the spatial and ensemble averaged height is related to the deposition flux by $\langle \langle H(x, t) \rangle_\eta \rangle_x = Ft$ or, equivalently, that $\langle \langle h(x, t) \rangle_\eta \rangle_x = 0$ holds. The afore-mentioned assumption, however, also implicitly implies that no coarse-grained *density* variations can occur. In the presence of local density variations, a discussion starting from the condition that no incoming particles are lost (cf. the following section) can lead to a KPZ term $(\nabla h)^2$ and a term $(\nabla^2 h)^2$ and, therefore, also to a nonzero *excess velocity* $v = \langle \langle \partial_t h \rangle_\eta \rangle_x = \langle \langle a_4 (\nabla h)^2 + a_5 (\nabla^2 h)^2 \rangle_\eta \rangle_x$ of the surface profile $h(x, t)$.

3   Physical Interpretation of the Growth Equation

In this section, we relate the various terms appearing in the growth equation (11) to the underlying microscopic mechanisms that dominate physical vapor deposition. This also leads to insights into the signs and the order of magnitude of the appearing coefficents in (11).

The two terms in (11) that are proportional to a_1 and a_6 can be interpreted as follows. The arriving particles from the beam experience close to the growing surface a deflection due to the interatomic attractive interaction
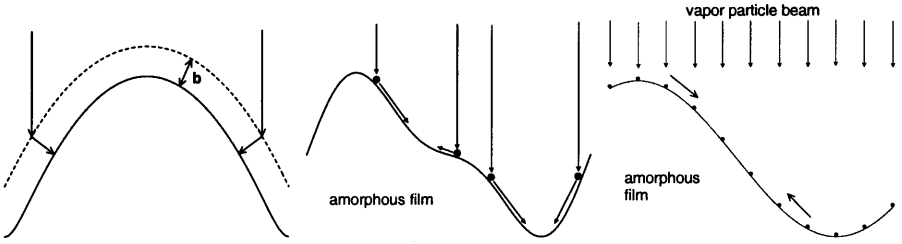
Fig. 2. Microscopic effects of amorphous surface growth. Left part: Inflection of particles due to interatomic interaction. Middle part: Surface diffusion of deposited particles due to surface relaxation. Right part: Equilibration of the inhomogeneous particle concentration due to the geometry of the surface

with the already condensed surface particles. Therefore, the particles do not arrive perpendicular to the substrate orientation, but perpendicular to the surface. This directly implies that more particles arrive at positions at the surface with negative curvature, $\nabla^2 h < 0$, than at positions with positive curvature $\nabla^2 h > 0$. We refer to Ref. [12] for experimental indications of the relevance of this effect. To model this scenario in a dynamical way, we use the idealization that (i) the particles experience a change of direction only if they reach a critical distance b (the effective range of the interaction) from the surface and (ii) are then attracted such that they arrive perpendicular to the surface, cf. left part of Fig. 2. A detailed mathematical derivation [8] using a reparametrization in the coordinates of the imaginary surface where the interaction becomes effective (cf. dotted line in the left part of Fig. 2) and a small gradient expansion in h in fact shows that this scenario gives simultaneously rise to the two contributions $a_1 \nabla^2 h$ and $a_6 M$ in (11). Moreover, the coefficients a_1 and a_6 can be related to the mean deposition flux F and the effective range b of the interatomic interaction yielding $a_1 = -Fb$ and $a_6 = Fb^2$ [8]. Although b cannot be directly measured its magnitude will be typically of the order of one atomic diameter and, therefore, much smaller than the radius of the surface curvature. This implies that the term proportional to a_6 is of minor relevance in comparison to the a_1-term and can be neglected. Moreover, the sign of a_1 is negative.

The term proportional to a_2 can be related to the surface relaxation originally suggested by Mullins [13,14]. The basic idea behind this effect (cf. also the middle part of Fig. 2) is that the deposited particles favor places at the surface with positive curvature $\nabla^2 h > 0$ since there, the already condensed surface particles create a local environment with higher binding energy. Therefore, one expects a diffusion current $j_m \propto \nabla(\nabla^2 h)$ leading to the term $-\nabla \cdot j_m = a_2 \nabla^4 h$ in (11) with a_2 being necessarily negative. This term can be interpreted as the result of a surface tension that attempts to minimize the area of the surface and, therefore, to smooth the surface morphology.

The nonlinear term proportional to a_3 arises as a consequence of the effect of equilibration of the non-homogeneous concentration c of the deposited particles just after arriving at the surface. This effect has originally been suggested by Villain [15, 16] (cf. also [17]). The deposition flux is basically homogeneous implying that more particles per surface area arrive at places with a small or zero modulus of slope ∇h than at places being strongly inclined with respect to the particle beam, cf. also the right part of Fig. 2. Therefore, the concentration is weighted by the slope, $c \propto 1/\sqrt{1 + (\nabla h)^2}$ leading to $c \propto 1 - \frac{1}{2}(\nabla h)^2$ in a small gradient expansion. The tendency to equilibrate the concentration gives rise to a diffusion current $\boldsymbol{j}_c \propto -\nabla c \propto \nabla(\nabla h)^2$, or equivalently, to the term $-\nabla \cdot \boldsymbol{j}_c = a_3 \nabla^2 (\nabla h)^2$ appearing in (11). It also directly follows that the coefficent a_3 is necessarily negative. The following simple dimensional argument leads to an estimate for a_3. Equation (11) implies that the coefficient a_3 has the dimension of length^3/time. The magnitude of a_3 necessarily depends on the deposition flux F that possesses the dimension of length/time and the mean diffusion length l under concentration equilibration which is the only relevant length scale determining this process. The simplest combination of F and l leading to the correct dimension of a_3 is Fl^2. Therefore, one expects $a_3 \propto -Fl^2$. A detailed discussion of the concentration equilibration [8] supports this argument and yields the explicit relation $a_3 = -\frac{1}{8}Fl^2$. Moreover, the typical magnitude of l is of the order of several atomic diameters.

As already mentioned in section 2, the physical origin of the nonlinear terms proportional to a_4 and a_5 is determined by the potential variations of the coarse-grained density [7,8]. These terms cannot result from particle desorption since the substrate is held at room temperature and the particle energy in the vapor beam is rather low (typically of the order 0.1eV). Therefore, *all* arriving particles finally contribute to the surface growth. As a consequence, any term that cannot be recast in form of the divergence of a current in (11) arises from changes of the coarse-grained density. Assuming for the moment that the deposition noise is zero ($\eta = 0$), particle conservation implies that the rate of change of the number of particles per substrate area at a given substrate location, C, is determined by a continuity equation $\partial_t C = -\nabla \cdot \boldsymbol{j}_C + \rho_0 F$. Here the divergence of the current $\boldsymbol{j}_C$ is given by the combination of all surface relaxation processes (cf. the afore-mentioned arguments), i.e. by $-\nabla \cdot \boldsymbol{j}_C = \rho_0[a_1 \nabla^2 H + a_2 \nabla^4 H + a_3 \nabla^2 (\nabla H)^2 + a_5 M]$. Allowing for density variations at the growing surface, the rate of change of C is related to the rate of change of the height H by $\partial_t C = \rho(\nabla H)\partial_t H$. Here $\rho(\nabla H)$ denotes the density at the surface. Without the incorporation of density changes ($\rho = \rho_0 = const.$), there is a direct proportionality $\partial_t C = \rho_0 \partial_t H$. If small density variations are taken into account, $\rho(\nabla H)$ can be expanded in the derivatives of H yielding $\rho(\nabla H) = \rho_0[1 + q_1(\nabla H)^2 + q_2\nabla^2 H]$ in lowest order approximation. Therefore, $\partial_t H = \rho_0^{-1}[1 - q_1(\nabla H)^2 - q_2\nabla^2 H]\partial_t C$ holds. Inserting this in the continuity equation from above, explains the presence

of the two terms $-q_1 F(\nabla H)^2 = a_4(\nabla h)^2$ and $-q_2 a_1(\nabla^2 H)^2 = a_5(\nabla^2 h)^2$ appearing in (11). From the physical point of view, however, density changes are primarily connected to the gradients of the surface profile reflecting the local arrangement of the particles at the surface and not so much to the surface curvature. Therefore, it is pausible to disregard the term $a_5(\nabla^2 h)^2$ in a minimal description of the growth evolution. Since the density variations result from a widening of the mean inter-particle distances at the surface one has to expect that they locally decrease the density implying that $a_4 > 0$ holds.

Taking into account the afore-mentioned physical arguments, the terms $a_5(\nabla^2 h)^2$ and $a_6 M$ can be neglected and we finally end up with *two minimal model equations for amorphous film growth*, namely

$$\partial_t h = a_1 \nabla^2 h + a_2 \nabla^4 h + a_3 \nabla^2 (\nabla h)^2 + \eta \qquad (13)$$

($a_i < 0$ for $i = 1, 2, 3$) that applies to growth processes with a basically homogeneous coarse-grained density and its more general version [7, 8]

$$\partial_t h = a_1 \nabla^2 h + a_2 \nabla^4 h + a_3 \nabla^2 (\nabla h)^2 + a_4 (\nabla h)^2 + \eta \qquad (14)$$

($a_4 > 0$) for the case that significant density variations occur during the growth process. Equations (13) and (14) constitute minimal models for amorphous surface growth in the following sense: They are based on the systematic minimal nonlinear functional form (11) of the growth equation, further reduced by taking into account the physical relevance of the various terms in (11) and contain the leading order nonlinearities that prevent the surface morphology from an exponentially rapid growth in time.

As a phenomenological ansatz, a growth equation similar to (14) has also been discussed in the context of molecular beam epitactic growth of crystalline structures (cf. [2]), albeit with different signs of the coefficients. Most significantly, the sign of the coefficient a_1 (being attributed to desorption effects) is then assumed to be positive. This in turn leads to a significantly different spatio-temporal evolution of the surface morphology since, in comparison to amorphous surface growth, a growth instability [8] is missing.

Whether (13) or (14) is the most appropriate minimal growth equation for specific amorphous films can also depend on the type of materials used. We note, however, that a recent study [7] considering the case of amorphous $Zr_{65}Al_{7.5}Cu_{27.5}$ films indicates the necessity of incorporating density variations – at least for that material.

4   Some Properties of the Growth Equation

In the remainder of this contribution, we address one specific point of the spatio-temporal evolution of the surface morphology, namely the differences of the temporal evolution of the correlation length R_c and the surface roughness w in (13) and (14). Using stochastic numerical simulations of the surface
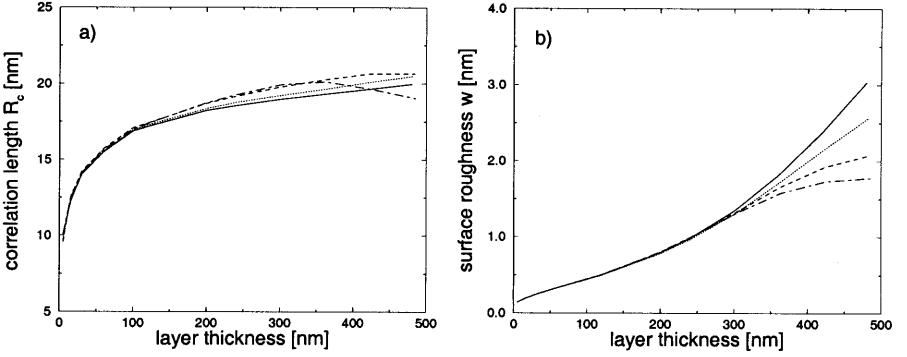
Fig. 3. Correlation length R_c and surface roughness w calculated from the nonlinear growth equation (14) using the parameters $a_1 = -0.0826\text{nm}^2/\text{s}$, $a_2 = -0.319\text{nm}^4/\text{s}$, $a_3 = -0.10\text{nm}^3/\text{s}$, $D = 0.0174\text{nm}^4/\text{s}$ and various values of a_4: $a_4 = 0\text{nm}/\text{s}$ (solid lines), $a_4 = 0.016\text{nm}/\text{s}$ (dotted lines), $a_4 = 0.040\text{nm}/\text{s}$ (dashed lines), and $a_4 = 0.055\text{nm}/\text{s}$ (dash-dotted lines).

growth equations starting from a flat substrate, we investigate the evolution of R_c and w as a function of the experimentally measurable layer thickness $\overline{H}$. This quantity is determined by $\overline{H} = \langle\langle H(x,t)\rangle_\eta\rangle_x = Ft + \langle\langle h(x,t)\rangle_\eta\rangle_x$ and, is, in general, implicitly connected to the time t via the solution of (14). For the special case (13), however, $\overline{H}$ is directly given by the product of the mean deposition flux and the time, i.e. Ft, since there is no excess velocity.

For the specific example of the growth of $\text{Zr}_{65}\text{Al}_{7.5}\text{Cu}_{27.5}$ films [4–7], a parameter estimation procedure discussed in detail in [7] yields for the coefficients in (14) $a_1 = -0.0826\text{nm}^2/\text{s}$, $a_2 = -0.319\text{nm}^4/\text{s}$, $a_3 = -0.10\text{nm}^3/\text{s}$, and $a_4 = 0.055\text{nm}/\text{s}$ and for the strength of the deposition noise $D = 0.0174\text{nm}^4/\text{s}$. For these values, a basically perfect agreement between experimental and simulation results has been obtained [7]. The dash-dotted lines in Fig. 3 correspond to this choice of the coefficents. Lowering the coefficient a_4 that determines the strength of the density variations during the growth process to zero reveals the following:

(i) Setting a_4 equals zero (solid line), the correlation length R_c initially increases proportional to a $t^{1/4}$ power law. This reflects the growth behavior in the time range where the linear terms in (13) are dominant and the nonlinear term is not yet excited due to the smallness of h. In contrast to the linearized form of (13) that leads to a constant value $R_c \propto \sqrt{2a_2/a_1}$ for large times, the nonlinear term in (13) significantly contributes to the spatio-temporal evolution of $h(x,t)$ and yields a further increase of R_c with the time t. For nonzero a_4 in (14), the dependence of R_c on the layer thickness agrees with the case $a_4 = 0$ up to $\overline{H} = 120\text{nm}$. Beyond this layer thickness, the correlation length R_c first increases slightly stronger than in the case $a_4 = 0$. For even larger layer thicknesses, a maximum of R_c is reached with a subsequent decrease of

R_c. This behavior can already be observed for $a_4 = 0.040$nm/s (dashed line) and is even more pronounced for $a_4 = 0.055$nm/s (dash-dotted line). As an aside, we note that R_c levels off again for layer thicknesses much larger than 500nm. As a consequence, the inclusion of density variations in the growth equation (14) significantly changes the typical length scale over which height fluctuations are correlated. Including density variations, *the correlation length R_c varies non-monotonically with layer thickness.*

(ii) Setting again a_4 equals zero (solid line), the surface roughness w initially increases exponentially. This again reflects the growth behavior in the time range where the linear terms in (13) are dominant and is therefore basically determined by the most unstable wave number of the linearized growth equation. For larger times, when the nonlinear term in (13) has a significant influence on the spatio-temporal evolution of $h(x, t)$, this increase crosses over to a power law, $w \propto t$ [8]. For nonzero a_4 in (14), the dependence of w on the layer thickness agrees with the case $a_4 = 0$ up to $\overline{H} = 300$nm. Beyond this layer thickness, the surface roughness w increases significantly weaker than in the case with $a_4 = 0$. More importantly, also the *curvature of w* changes with the layer thickness. This behavior can be clearly observed for $a_4 = 0.040$nm/s (dashed line) and is even more pronounced for $a_4 = 0.055$nm/s (dash-dotted line). As a consequence, the inclusion of density variations in the growth equation (14) *primarily changes the curvature of the evolution of the surface roughness* which leads to a strongly delayed increase, or even to a saturation of w.

(iii) Comparing the results for R_c and w, one infers that the impact of density variations shows evidence in the correlation length R_c at much earlier stages of the growth process than in the surface roughness w.

5   Conclusion

In this contribution, we have systematically derived the minimal functional form of a growth equation appropriate for the understanding of amorphous thin film growth, cf. equation (11). We have also related the terms occuring in the functional form (11) to underlying microscopic surface relaxation mechanisms and presented some results on the temporal evolution of the correlation length and the surface roughness of the corresponding surface morphology. Finally we showed that the incorporation of density variations of the growing material significantly reduces the surface roughness.

References

1. Tong W. M., Williams R. S. (1994) Kinetics of surface growth. Annu. Rev. Phys. Chem. **45**, 401-438

2. Barabasi A.-L., Stanley H. E. (1995) Fractal concepts in surface growth. Cambridge University Press

3. Marsili M., Maritan A., Toigo F., Banavar J. R. (1996) Stochastic growth equations and reparametrization invariance. Rev. Mod. Phys. **68**, 963-983

4. Reinker B., Moske M., Samwer K. (1997) Kinetic roughening of amorphous ZrAlCu film investigated in situ with scanning tunneling microscopy. Phys. Rev. **B 56**, 9887-9893

5. Mayr S. G., Moske M., Samwer K. (1998) Early stages in amorphous $Zr_{65}Al_{7.5}Cu_{27.5}$ film growth on HOPG. Europhys. Lett. **44**, 465-470

6. Mayr S. G., Moske M., Samwer K. (1999) Identification of key parameters by comparing experimental and simulated growth of vapor deposited amorphous $Zr_{65}Al_{7.5}Cu_{27.5}$ films. Phys. Rev. **B 60**, 16950-16955

7. Raible M., Mayr S. G., Linz S. J., Moske M., Hänggi P., Samwer K. (2000) Amorphous thin film growth: theory compared with experiment. Europhys. Lett. **50**, 61-67

8. Raible M., Linz S. J., Hänggi P. (2000) Amorphous thin film growth: minimal deposition equation. submitted for publication

9. Lai Z.-W., Das Sarma S. (1991) Kinetic growth with surface relaxation: continuum versus atomistic models. Phys. Rev. Lett. **66**, 2348-2351

10. Edwards S., Wilkinson D.R. (1982) The surface statistics of a granular aggregate. Proc. Roy. Soc. London **A 381**, 17-31

11. Kardar M., Parisi G., Zhang Y. C. (1986) Dynamic scaling of growing interfaces. Phys. Rev. Lett. **56**, 889-892

12. van Dijken S., Jorritsma L. C., Poelsema B. (1999) Steering-enhanced roughening during metal deposition at grazing incidence. Phys. Rev. Lett. **82**, 4038-4041

13. Mullins W. W. (1957) Theory of thermal grooving. J. Appl. Phys. **28**, 333-339

14. Mullins W. W. (1959) Flattening of a nearly plane solid surface due to capillarity. J. Appl. Phys. **30**, 77-83

15. Villain J. (1992) Continuum models of crystal growth from atomic beams with and without desorption. J. Physique **I 1**, 19-42

16. Pimpinelli A., Villain J. (1998) Physics of crystal growth. Cambridge University Press

17. Moske M. (1997) Mechanische Spannungen als Sonde für Schichtwachstum und Schichtreaktionen. Habilitation thesis, Universität Augsburg, unpublished