# 4) Growth

Sources:

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A. Pimpinelli and J. Villain, Physics of Crystal Growth (Cambridge University Press 1999)
J. A. Venables, Introduction to Surface and Thin Film Processes, (Cambridge University Press 2000)
J. Krug, Adv. Phys. 46, 139 (1997)
J. Krug, "Four lectures on the physics of crystal growth"
H. Lüth, Surfaces and Interfaces of Solids (Springer 1993)

# 4.1) General Considerations and Classification of Growth Modes

### 4.1.1) Three categories

We consider the adsorption of some film material on a solid substrate. Generally, three categories of growth scenarios are distinguished (Bauer 1958), which at first glance resemble the situation for wetting and spreading:

Frank-van-der-Merwe (FM) growth (layer-by-layer) ... corresponding to complete wetting;

Stranski-Krastanov (SK) growth (layer-by-layer followed by islanding) ... corresponding to the intermediate case of the "spreading scenarios" (see Sec.3.6);

Vollmer-Weber (VW) growth (islanding)

... corresponding to dewetting (finite wetting angle).



The specific conditions, under which these growth modes occur, can be related to the surface or interface tension  $\gamma$ , i.e. the characteristic free energy (per unit area) to create an additional surface or interface.



Since  $\gamma$  can also be interpreted as a force per unit length of boundary, force equilibrium at a point where substrate and 3D island of the deposited film touch requires

 $\gamma_{\rm S} = \gamma_{\rm F} + \gamma_{\rm S/F} \cos \Phi$ 

where  $\gamma_S$  is the surface tension of the substrate/vacuum interface,  $\gamma_F$  that of the film/vacuum, and  $\gamma_{S/F}$  that of the substrate/film interface. Using this equation the two limiting growth modes (layer-by-layer, FM) and (islanding, VW), can be distinguished by the angle wetting angle  $\Phi$ .

•	FM:	$\Phi = 0,$	$\gamma_{S} \geq \gamma_{F} + \gamma_{S/F}$
•	VW:	$\Phi > 0$ ,	$\gamma_{\rm S}$ < $\gamma_{\rm F}$ + $\gamma_{\rm S/F}$

The mixed mode (SK) can easily be explained in this picture by assuming that there is a lattice mismatch between the deposited film and the substrate. The lattice of the film tries to adjust to the substrate, but at the expense of elastic deformation energy. The transition from layer to island growth occurs when the spatial extent of the elastic strain field exceeds the range of the adhesion forces within the deposited material.

Importantly, however, these considerations are the only a "kinetic analogon" to the wetting scenarios; in fact, since growth is a non-equilibrium process, the surface energies and wetting scenarios can only serve as a limiting case for comparison, but they cannot suffice to properly describe the growth.

### 4.1.2) How to describe growth ?

As pointed out above, surface energies enter the "equilibrium structure", but actually growth is a non-equilibrium situation, a dynamical problem.

Therefore, dynamical parameters have to enter,

- → the time scale of incoming molecules (the flux F, which equals the growth rate on the surface only if all incoming molecules are adsorbed and if there is no re-desorption)
- → the time scale of dynamics on the growing surface (i.e. diffusion D), which "competes" with the time scale set by the flux, so that we can expect that in certain situations the ratio (F/D) is relevant

This already suggests that time dependent equations such as, e.g., rate equations with different kinetic processes are a possible approach to the describe growth.



Of course, the situation is more complex than suggested by only two parameters (or even only one, if exclusively the ratio (F/D) was relevant).

In fact, there are several processes contributing to the dynamics on the (growing) surface, and realistically they cannot all captured in one parameter D.



For a system with well-defined layers (e.g., a crystalline adsorbate), the diffusion within one layer (intralayer diffusion) has to be distinguished from the diffusion between different layers (interlayer diffusion). Also, nucleation processes of islands are important. Generally, of course, all these parameters depend on the temperature *T*. Overall, growth is a rich and still expanding area of science, involving rather advanced (non-equilibrium) statistical mechanics. Here we shall indicate only some basic concepts.

Note that we shall ignore in the following issues such as a possible lattice misfit between substrate and adsorbate for epitaxial systems, although the resulting elastic stress / strain can of course we very important.

Note also that the kinetic description of growth is also relevant for the case that the adsorbate and the substrate are of the same material (which for (single-)crystalline materials would be called homoepitaxy). While in equilibrium we would naively expect the surface of this system to be simply flat, due to the kinetic (non-equilibrium) nature of growth it will not. See, for example the following pictures of Pt islands grown on a Pt(111) surface. In fact although seemingly not very exciting, homoepitaxial systems as these have been used as very useful model systems for understanding growth



In order to get a first idea of the concepts involved, we will discuss two simple growth models. After that, we shall discuss possible refinements of these and other approaches.

### **4.2) Simple Growth Models**

### 4.2.1) Langmuir Growth for a Monolayer

Consider a system for which the adsorbate grows only in the first monolayer, and further growth (i.e. occupancy of the second layer etc.) is impossible. This approximates relatively well the situation for some surfactant systems and selfassembled monolayers (SAMs) of the thiol-on-gold type, but it has nothing to do with, e.g., the evaporation of, say Au on Si.

Of course, in this case the comparison with wetting / dewetting scenarios is not very fruitful, but it is nevertheless useful to get a feeling for kinetic equations.

In the simplest case we assume that there is no lateral interaction between different sites, so that the occupancy of a given adsorption site (again, all in the first monolayer) has no impact on the adsorption rate in the neighbouring sites. The adsorption sites in the monolayer are thus considered all independent. The rate of chance of the coverage  $\theta$  (normalised to one) is thus simply proportional to the incoming flux times *F* some probability of adsorption  $\Omega$  (which we combine in a growth rate (1/ $\tau$ )) and to the fraction (1- $\theta$ ) of unoccupied adsorption sites

$$\frac{d}{dt}\theta = \frac{1}{\tau}(1-\theta)$$

This differential equation is easily solved (with the boundary condition that for the starting time the coverage is zero, i.e.  $\theta$  (*t*=0)=0)

$$\theta(t) = 1 - \exp(-t/\tau)$$

A typical example (taken from thiols on gold) is shown in the Figure.



### 4.2.2) Multilayer Growth without Interlayer Diffusion ("Statistical Growth")

We consider a system, in which not only a monolayer, but all layers with their respective coverage  $\theta_n$  can be occupied and grow. As the key ingredient in this model we assume that there is no interlayer diffusion, i.e. a given adsorbate molecule will stay in the layer in which it was initially accommodated and can only diffuse laterally *within* this layer. This model is referred to as "statistical growth" or also "random deposition".

The growth rate of the *n*th layer is then given by

$$\frac{d}{dt}\theta_n = \Omega F(\theta_{n-1} - \theta_n)$$

with the boundary condition  $\theta_{n=0} = 1$ .

The *exposed* layer coverages (i.e., the fraction of a given layer which is *not* covered by the one above it) is

$$\varphi_n = \theta_n - \theta_{n+1}$$

*n*+1

n

*n*-1

The solution of this system of differential equations is (Michely/Krug, p.130)

 $\varphi_n$ 

$$\theta_n = 1 - e^{-\theta} \sum_{k=0}^{n-1} \frac{\theta^k}{k!}$$

where the total coverage is

$$\theta = \sum_{n=1}^{\infty} \theta_n = \Omega Ft$$
 with  $0 \le \theta_n \le 1$ 

The *exposed* layer coverages (i.e., the fraction of a given layer which is *not* covered by the one above it) thus follow a Poisson distribution

$$\varphi_n = \theta_n - \theta_{n+1} = \frac{e^{-\theta}\theta^n}{n!}$$

This describes the "terraced structure", i.e. the fractional size of a given layer, the "envelope" of which has a variance

$$W = \sqrt{\theta}$$

characteristic for a Poisson distribution.

The main result is the evolution of the roughness according to

roughness ~  $\sqrt{\theta}$ 

Importantly, the interlayer diffusion does *not* enter this result. For experimental studies (to be discussed later) we note that the scattering at the anti-Bragg point for statistical growth behaves as

$$I \sim e^{-4\theta}$$



# **4.3) Intralayer and Interlayer Diffusion and the Ehrlich-Schwoebel** Barrier

Generally, of course, there will be some form of transport (diffusion) of the adsorbates. We distinguish

- --- intralayer diffusion D (diffusion within one given layer) and
- --- interlayer diffusion D' (diffusion between different layers)

Intralayer diffusion D (in combination with the incoming flux F) determines to a large extent the typical island sizes and distances in the plane. We will come back to this in the sections on nucleation and on island sizes and shapes. D will typically depend on the temperature via

$$D = D_0 \exp(-E_D/k_BT)$$



Interlayer diffusion D' usually encounters an additional energy barrier  $\Delta E_s$  which adsorbates have to overcome for diffusing over an edge into the next layer as shown schematically in the figure.



This barrier  $\Delta E_s$  is known as Ehrlich-Schwoebel barrier and usually leads to D' < D, i.e.

$$D'/D = \exp(-\Delta E_s/k_BT) < 1$$

A typical order of magnitude is, e.g.,  $\Delta E_s \approx 0.14 \text{ eV}$  for Pt growth on Pt(111).

### **4.4) Nucleation Theory**

### 4.4.1) Classical (Continuum) Nucleation Theory

### 3D nucleation

Nucleation is a classical process for the formation of a new phase. After formation of a nucleus, new material from the environment can diffuse and attach to the nucleus. The key question concerns the stability of the nucleus.

Associated with the formation of a spherical nucleus of a new phase is a change in Free Energy (here in three dimensions)

$$\Delta F = \Delta F_{Volume} + \Delta F_{Surface} = \frac{4\pi}{3} r^3 \Delta g_V + 4\pi r^2 \gamma$$

The interface energy  $\gamma$  of the nucleus is in any event positive. The volume part  $\Delta g_V$  can be negative (i.e. favourable), i.e.  $\Delta g_V < 0$ . Then the nucleus *can* be stable above a size  $r^*$ . For  $r > r^*$  (above the local maximum of  $\Delta F$ ) with

$$r^* = \frac{2\gamma}{\Delta g_V}$$

the nucleus is stabilised by further growth (growing *r*).

### 2D nucleation

The same picture can be used to discuss the mechanism of the formation of (2D) islands on the surface of a solid. The (possibly stable) nucleus is now twodimensional, and the boundary (the interface, i.e. the rim) is now one-dimensional. The formation of the interface will also here cost energy ( $\sim r$ ), but this can be (over)compensated by the energy gain proportional to the area of the island ( $\sim r^2$ ), if the island is sufficiently large ( $r > r^*$ ').

### **4.4.2)** Atomistic Nucleation Theory

If the nucleus is very small, we can no longer use a continuum picture. We thus employ an atomistic picture.

### 2D Nukleation

- Ziel ist es, die Dichte der Nukleationskeime bei gegebener Rate F und Diffusionskonstante D auf der Oberfläche mit Hilfe von Ratengleichungen zu berechnen.
- Dazu wird die Bildung eines "kritischen Keims", dessen Größe auch atomare Dimensionen annehmen kann, wesentlich sein.
- Allgemein kann dieses Problem durch Computersimulationen behandelt werden

1. Schritt:

Aufstellung der Ratengleichungen für die über große Flächen gemittelte Clusterdichten  $n_s$  (d.h.  $n_1$  = Adatomdichte,  $n_2$  = Dimerdichte, usw.)

Da die Beweglichkeit von einzelnen Atomen ist im allgemeinen viel größer als die Beweglichkeit von Clustern, kann man sich auf die Anlagerung bzw. Abspaltung einzelner Atome an die bestehenden Cluster beschränken.

Rate  $\Gamma_s$  beschreibe die Bildung von Clustern mit s+1 Atomen. Dann gilt

 $\begin{array}{ll} \displaystyle \frac{dn_{\, {\tt s}}}{dt} & = \Gamma_{{\tt s}-1} \, \cdot \, \Gamma_{\tt s} & ({\tt s} \geq 2) & (*) \\ \displaystyle \Gamma_{\tt s} & = \sigma_{\tt s} D \, \, n_1 \, \, n_{\tt s} \, \cdot \, \gamma_{\tt s+1} \, \, n_{\tt s+1} & (*) \end{array}$ 

Es werden die Raten der zugehörigen Elementarprozesse verwendet:

 $\gamma_{s}$  : Abspaltung eines Atoms vom Cluster mit s Atomen

 $\sigma_{\text{s}}$  : Anlagerung eines Atoms an Cluster mit s Atomen

für Adatome lautet die Ratengleichung

$$\frac{\mathrm{d}n_1}{\mathrm{d}t} = \mathbf{F} - 2\Gamma_1 - \sum_{\mathfrak{a} \ge 2} \Gamma_\mathfrak{a} \qquad (*)$$

mit

- F: Depositionsrate
- Γ<sub>1</sub>: Bildung von 2er Clustern
- Γ<sub>s</sub>: Bildung von Mehrfachclustern

2. Schritt:

Vereinfachung des Gleichungssystems (\*):

Einführung einer kritischen Clustergröße i\*

 $\begin{cases} \gamma_{\epsilon} = 0 & \text{für } \epsilon \geq i^* + 1 \quad (\text{Cluster stabil}) \\ \gamma_{\epsilon} \text{ groß} & \text{für } \epsilon \leq i^* \quad (\text{sehr schnelle Abspaltung}) \end{cases}$ 

i\* ist ein kritischer Parameter, der auf der typischen Zeitskala des Wachstums gültig ist

• Dichte der stabilen Cluster ist definiert als  $N = \sum_{a=i^{*}+1}^{\infty} n_{a}$ 

mit  $\Gamma_s = 0$  für  $1 \le s \le i^*-1$  folgt aus (\*)

$$\frac{dN}{dt} = \sum_{\mathfrak{s}=i^*+1}^{\infty} \frac{dn_{\mathfrak{s}}}{dt} = \sigma_{i^*} Dn_1 n_{i^*} \qquad (\#)$$

mit

$$\begin{split} n_{\mathfrak{s}} = n_{\mathfrak{s}}^{1} e^{-E_{\mathfrak{s}}/k_{B}T} & (\text{mit } 2 \leq \mathfrak{s} \leq i^{*} + 1) \quad (\#) \\ \overline{\sigma} = \frac{1}{N} \sum_{\mathfrak{s} \geq i^{*} + 1}^{\infty} n_{\mathfrak{s}} \sigma_{\mathfrak{s}} & \text{mittlere Einfangrate stabiler Cluster} \end{split}$$

$$\frac{dn_1}{dt} = F - \sigma_{i*} Dn_1 n_{i*} - \overline{\sigma} D_1 n_1 N \qquad (\#)$$

- Lösung des 2D-Nukleationsproblems: 2 Regime für das Gleichungssystem (#)
  - $\begin{array}{ll} \label{eq:alpha} \alpha \mbox{) frühe transiente Phase:} & \Theta = Ft, & N = \Theta^{i^*+2} \\ \beta \mbox{) steady state} & n_1 \approx F \, / \, D \overline{\sigma} N \end{array}$ 
    - einsetzen in (#) und Integration

$$\begin{split} \mathrm{N} &= \Theta^{\frac{1}{i^*+2}} \left(\frac{\mathrm{F}}{\mathrm{D}}\right)^{\frac{i^*}{i^*+2}} \mathrm{e}^{\mathbf{E}_{i^*}/(i^*+2)k_{\mathrm{B}}\mathrm{T}} \\ \mathrm{N} &\propto \left(\frac{\mathrm{F}}{\mathrm{D}}\right)^{\chi} \quad \mathrm{mit} \quad \chi = \frac{i^*}{i^*+2} \end{split}$$

Man findet also, dass N einem universellen Potenzgesetz für (F/D) folgt!

beschränkt auf kleine Bedeckungen 0

# 4.5) Island Shapes

(see Michely / Krug book, Chapter 3)

The last section was concerned with nucleation, which determines the number density of islands.

This section is concerned with their *shape*. The shape is determined by two additional factors, namely the atomic or molecular processes occurring at the island edge, and the effects of the local environment of the island on the diffusion field of the adatoms. Consider Fig.3.1 (fromMichaly/Krug) for an overview of the variety of scenarios of island shapes even for one and the same system (Pt/Pt(111)) when only the temperature is varied, i.e. when different kinetic processes are "sequentially" switched on.





600 K

700 K

Fig.3.1 (from Michely/Krug, p.62) Pt/Pt(111) at different deposition temperatures (image size  $(156 \text{ nm})^2$ ). We should learn how to read the shapes ("A snowflake is a letter to us from the sk

We should learn how to read the shapes ("A snowflake is a letter to us from the sky." (p.62)).

### **4.5.1) Equilibrium Island Shapes**

(see Michely / Krug book, Chapter 3)

For equilibrium, we expect the island shape to be determined mechanisms such as the minimization of the surface (i.e. boundary / perimeter) energy. This would correspond to the concept of the Wulff construction.

For an isotropic system this would be a circular shape; for a crystalline system, the equilibrium island shape would reflect the crystal symmetry (see Fig.3.1(f) with the hexagonal island for growth at high temperature, as expected for an fcc(111) surface). Note that for heteroepitaxy (i.e. two different materials on top of each other), strain due to the lattice mismatch will enter the considerations.

If the island shape is *not only* determined by equilibrium energy considerations, but also or even predominantly by kinetic factors, it will be different. This is already demonstrated by Fig.3.1(a)-(e), where the island shape obviously deviates from the equilibrium hexagonal (compact) shape.

### 4.5.2) Fractal-Dendritic Islands

Michely/Krug (their Sec.3.3, p.81ff; see also Sec.3.6). http://en.wikipedia.org/wiki/Dendrite\_(crystal)

The term "dendrite" is derived from the Greek word *dendron* ( $\delta \epsilon \nu \delta \rho o \nu$ ) for "tree".

This is a rather commonly found morphology for a number of growth phenomena, from snowflakes to islands on metal surfaces and even high-voltage breakdowns (see below).

The formation of fractal-dendritic islands is a common feature in low-temperature homoepitaxial growth on fcc(111) metal surfaces (Michely/Krug, p.81). Typically, from the onset of island formation up to ~ 15 % of the melting temperature of the material.



Fig.3.9 (from Michely/Krug, p.81) Fractal-dendritic islands created by homoepitaxial growth of (a) Pt(111); (b) Al(111); (c) Ag(111), in each case at deposition temperatures around 10 % of T\_melt. (image size  $(120 \text{ nm})^2$ ).

Michely/Krug discuss different types of mechanisms for the formation of fractaldendritic islands (Sec.3.3 and Sec.3.6).

We will not go into the details of this rather involved discussion, but refer to the the diffusion-limited aggregation model (DLA) below, which helps to rationalize important features of denrite formation in the limit of low coverage ( $\theta$ <<1) and low island density ( $\Omega$ N<<1), although even in these limits there are important differences to the experiments (Michely/Krug, p.82).

### 4.5.3 Diffusion-Limited Aggregation (DLA) --- A Simple Model

Witten & Sander, PRL 47 (1981) 1400 Witten & Sander, PRB 27 (1983) 5686 Michely/Krug, p.63; p.81/82; p.100

A simple model related to, but not identical to dendritic growth discussed above is that of DLA.

A few key points in the context of DLA:

The Eden model is a lattice model, in which particles are added on at a time at random to sites adjacent to occupied sites. This produces relatively compact clusters.

DLA is a variant of the Eden model.

After placing a seed particle at the origin, a second particle is added at some random site at large distance from the origin.

This particle walks randomly (diffuses) until it visits a site adjacent to the seed.

There the walking particle then becomes part of the existing cluster

Then the next particle is added, diffuses, and attaches to the existing "cluster" and so on.

The exposed ends of the clusters tend to grow more rapidly than other perimeter sites because perimeter sites near the center are "shadowed".

DLA produces aggregates less compact than the Eden model.

The correlation function  $C(r) \sim \Sigma_r \rho(r')\rho(r'+r)$  decays as  $C(r) \sim r^{-1/3}$ 

The clusters formed in DLA are referred to as Brownian trees. (Wikipedia) Of critical importance is that the number of particles undergoing Brownian motion in the system is kept very low so that only the diffusive nature of the system is present. (Wikipedia)



Fig.XXX (from Witten & Sander, PRB 27 (1983) 5686) Computer simulation of 3000 particles on a square lattice growing according to DLA. The "screening effect" is illustrated by showing the first 1500 particles to attach to the aggregate as open circles, the rest (latter 1500 particles) as dots.

### Related Systems in this Context

Snowflakes are an obvious related system, but the implications are broader. Witten/Sander already pointed out that the features predicted for DLA may appear in many processes, including gelation, condensation polymerisation, and agglutination of biological molecules.



Fig.XXX Snowflake (www.chris-j.co.uk/images/dendritessnow.jpg). Note that according to Pimpinelli/Villain (p.177) the formation of snowflakes is still not quantitatively understood.



Fig.XXX Lichtenberg figure (from Wikipedia on dendrites). High-voltage dielectric breakdown within a block of plexiglass creates a fractal pattern.

# **4.6) Some aspects and pictures on growth beyond the monolayer** (see work by Krug and others)

# Wedding cakes



Wedding cakes (from Krug et al., PRB 61 (2000) 14 037); here Pt/Pt(111) with CO partial pressure.



# <u>Kinetic growth modes (according to Krug)</u> (an alternative scheme to FvdM / SK / VW discussed earlier)



Key factors: Intra-layer and inter-layer mobility

### **4.7 Statistical Description of Surfaces by Correlation Functions**

We consider a film on a substrate or an A-on-A depositon (homoepitaxy) of some thickness well beyond the monolayer.

The surface profile, i.e. the height *h* as a function of lateral coordinate (x,y) is described by h(x,y).

The surface width, i.e. the root-mean-squared roughness

$$\sigma = \langle [h(x',y') - h(x,y)]^2 \rangle^{1/2} (x'-x), (y'-y) \to \text{infinity} \rangle$$

where  $\langle ... \rangle$  means the average over all lateral positions, will then have some significant value typically well above a monolayer thickness.

If we want to compare this to theoretical predictions or to other surfaces, it is obviously not practical to compare the complete function z(x,y), but to extract certain features which are *statistically significant*.

How do we describe the surface appropriately in a statistically significant way ? A typical approach is the height difference correlation function

$$G(r) = \langle [h(x+r) - h(x)]^2 \rangle$$

which is related to the height-height correlation function

by

$$G(r) = 2\sigma^2 - 2C(r)$$

 $C(r) = \langle [h(x+r) h(x)] \rangle$ 

Typical examples are illustrated next.

(see also correlation function in the condensed matter lecture in the fifth term)





Fig.xxx shows the typical shape of the correlation function, which can be intuitively interpreted (see also Pimpinelli/Villain, p.207).

Initially, the height difference is small, since the probability to experience a significant height difference relative to the starting point for small lateral distances r is small.

At some lateral distance  $\xi$  typically the correlations are "lost", but the differences also to not increase anymore and the "maximum peak to valley height difference is reached". Beyond  $\xi$ , the correlation function levels off to a constant value, which corresponds to  $2 \sigma^2$ .

This type of correlation function is thus described by three parameters

- 1) Initial slope, given by the "Hurst exponent", usually called h or  $\alpha$ .
- 2) Lateral correlation length,  $\xi$ .
- 3) Value of the correlation function for infinite lateral distances, i.e. roughness,  $\sigma$

In order to connect the small r and the large r limit, often an approximate form of C(r) is used, namely

$$C(r) = \sigma^2 \exp[(-r/\xi)^{2\alpha}]$$

Below we discuss typical examples for G(r) or 2C(r). Scaling arguments (why the initial slope follows a power law etc.) are given later.

### **Examples (for constant roughness in all cases)**

Note that the figures (courtesy of Universität Kiel) show C(r) instead of G(r), but this essentially only inverts the sign.



# Variation of the correlation length ξ

## Variation of the Hurst parameter

# **<u>4.8 Notes on the Background of Scaling Laws in Growth Physics and</u> <u>Self-Affinity</u>**

Pimpinelli / Villain, p.208 (good explanation even with calculation for example) Michely / Krug, p. 197 (we follow their introduction here)

The concepts of fractal geometry have been used extensively to characterize structures formed in growth far from equilibrium.

For our purposes, fractals can be defined as objects that show statistical scale invariance, which means that they look the same (in a statistical sense, i.e. exhibit the same parameters characterizing their morphology) under arbitrary levels of magnification (i.e. in images of any scale).

This self-affinity is then expressed in a transformation

$$h(x) \implies b^{-\alpha} h(bx)$$

where *b* is a scale factor and the exponent  $\alpha$  the Hurst parameter ( $\alpha$ =1 for self-similarity and  $\alpha$ <1 for self-affinity).

This has direct implications for the height difference correlation function, namely

$$G(r) = b^{-2\alpha} G(br)$$

Setting b = 1/r directly leads to

$$G(r) \sim r^{2\alpha}$$

which is the hallmark of self-affine scaling.

This is indeed found in many systems in a good approximation for small lateral distances r.

This also implies further scaling laws (here without proof; see Michely/Krug, p.199)

$$\begin{array}{lll} \sigma \left( t \right) & \sim & t^{ \alpha / z} \\ \xi \left( t \right) & \sim & t^{ 1 / z} \end{array}$$

We note that more generally than the above presentation one writes

where

$$\sigma(t) \sim t^{\beta}$$

$$\beta = \alpha / z + \lambda$$

 $\lambda = 0$  corresponds to the "no steepening" case.

 $\lambda > 0$  is referred to as *anomalous*.

In other words, the equations on the previous page correspond to the no steepening.

The typical values for these exponents are "of the order of 1", but of course depend on the specific model, i.e. mechanism of growth, which applies to a given system. In practice, the exponents are determined experimentally and then compared to different models.

# 4.9) Addendum: Notes on Self-Similarity and Fractals

The concepts to describe growth morphologies have substantial overlap with those of fractals. We have seen this already in Sec.4.5 with some of the examples of island formation at low temperature as well as with snowflakes. Therefore, we include an excursion to fractals.

### Please note, though

1) The concepts of fractals can be used for some purely 2D (in-plane) morphologies as discussed in the context of island shapes (of the first (sub)monolayer) in growth; They can ALSO be used for some "3D" growth morphologies of a more complex height profile h(x,y) well beyond the monolayer regime, i.e. with roughnesses also well above the monolayer.

2) Of course, there are many more scenarios, and not all growth morphologies are fractal.

3) If a real growth system exhibits elements of fractal nature, this can only apply to a limited length scale, since obviously the smallest length scale is the size of the atoms or molecules of which the material is built us. Below this, the concept of fractals cannot work.

### From Wikipedia

(see also live animations on Wikipedia)

A **fractal** is "a rough or fragmented <u>geometric shape</u> that can be split into parts, each of which is (at least approximately) a reduced-size copy of the whole,"<sup>[11]</sup> a property called <u>self-similarity</u>. Roots of the idea of fractals go back to the 17th century, while mathematically rigorous treatment of fractals can be traced back to functions studied by <u>Karl Weierstrass</u>, <u>Georg Cantor</u> and <u>Felix Hausdorff</u> a century later in studying functions that were <u>continuous</u> but not <u>differentiable</u>; however, the term <u>fractal</u> was coined by <u>Benoît Mandelbrot</u> in 1975 and was derived from the Latin <u>fractus</u> meaning "broken" or "fractured." A mathematical fractal is based on an <u>equation</u> that undergoes <u>iteration</u>, a form of <u>feedback</u> based on <u>recursion</u>.<sup>[2]</sup> There are several examples of fractals, which are defined as portraying exact self-similarity, quasi self-similarity, or statistical self-similarity. While fractals are a mathematical construct, they are found in nature, which has led to their inclusion in <u>artwork</u>. They are useful in medicine, soil mechanics, <u>seismology</u>, and <u>technical analysis</u>.



Fig.4.9.1 (From Wikipedia) Even 2000 times magnification of the Mandelbrot set uncovers fine detail resembling the full set.

### **Related Issues in this Context**

Bunde/Havlin, chapter by Stanley, p.2/3 Illustration what (nonrandom) fractals based on the Sierpinski gasket fractal

Key point:

The dependence of mass M (normalized to one unit as 1) as well as density  $\rho$  on length L is not related to an integer power law. We show this after defining density as

 $\rho$  (L) = M(L) / L<sup>2</sup>

in 2-dimensional space as in this example. We rather find

$$\rho = L^{\alpha}$$



Fig.4.9.2



Fig.4.9.3

With the exponent  $\alpha$  given by the slope in the corresponding figure as

$$\alpha = \text{slope} = [\log 3/\log 2] - 2$$

although it would usually be expected to be an integer number, corresponding to the dimension of the respective space the object "lives" in, i.e. 1 for a line, 2 for an area, and 3 for a volume.

The fractal dimension df is then defined through

$$M(L) = A L^{df}$$

or after re-substitution

$$\rho(L) = A L^{df-2}$$

For the Sierpinski gasket the dimension is indeed "fractal", i.e. non-integer. The fractal dimension for the Sierpinski gasket is

$$df = [\log 3/\log 2] = 1.58...$$

### Fractal dimension of a snowflake



Fig.4.9.4 (a) Typical snow crystal. (b) A DLA simulation. (c) Comparison between the fractal dimensions of (a) and (b) obtained by plotting (log-log) the number of pixels inside an LxL sandbox vs. L. The same slope is found for both: df = 1.85 + 0.06.





### Determination of the fractal dimension from experiments Image analysis: Ag grown on Pt(111) at 110 K Harald Brune et al., Nature 369 (1994) 469



Fig.4.9.6 (from Brune) similar plot as for snowflake; the resulting df is 1.78.

### Scattering data

(Bunde / Havlin, p.275/279)

From simple law for spatial correlations  $G(r) \sim r^{df-d}$ Then the structure factor is  $S(q) \sim q^{-df}$ It is as simple as that.

### Random walk

Also the pattern arising from a random walk can be analysed in terms of fractal geometry. (Bunde/Havlin, chapter by Stanley, p.4/5)

### How Long Is the Coast of Britain? Statistical Self-Similarity and Fractional Dimension From Wikipedia, the free encyclopedia



Unit = 200 km, length = 2400 km (approx.)



Unit = 50 km, length = 3400 km

**"How Long Is the Coast of Britain? Statistical Self-Similarity and Fractional Dimension"** is a paper by <u>mathematician Benoît Mandelbrot</u>, first published in <u>Science</u> in 1967. In this paper Mandelbrot discusses <u>self-similar</u> curves that have <u>Hausdorff dimension</u> between 1 and 2. These curves are examples of <u>fractals</u>, although Mandelbrot does not use this term in the paper, as he did not coin it until 1975.

The paper examines the <u>coastline paradox</u>: the property that the measured length of a stretch of coastline depends on the scale of measurement. Empirical evidence suggests that the smaller the increment of measurement, the longer the measured length becomes. If one were to measure a stretch of coastline with a <u>yardstick</u>, one would get a shorter result than if the same stretch were measured with a 30cm <u>ruler</u>. This is because one would be laying the ruler along a more curvilinear route than that followed by the yardstick. The empirical evidence suggests a rule which, if extrapolated, shows that the measured length increases without limit as the measurement scale decreases towards zero. This discussion implies that it is meaningless to talk about the length of a coastline; some other means of quantifying coastlines are needed. Mandelbrot discusses an empirical law discovered by Lewis Fry Richardson, who observed that the measured length L(G) of various geographic borders was a function of the measurement scale G. Collecting data from several different examples, Richardson conjectured that L(G) could be closely approximated by a function of the form

$$L(G) = MG^{1-D}$$

where M is a positive constant and D is a constant, called the dimension, greater than or equal to 1. Intuitively, if a coastline looks smooth it should have dimension close to 1; and the more irregular the coastline looks the closer its dimension should be to 2. The examples in Richardson's research have dimensions ranging from 1.02 for the coastline of <u>South Africa</u> to 1.25 for the West coast of <u>Britain</u>.

Mandelbrot then describes various mathematical curves, related to the Koch snowflake, which are defined in such a way that they are strictly self-similar. Mandelbrot shows how to calculate the Hausdorff dimension of each of these curves, each of which has a dimension D between 1 and 2 (he also mentions but does not give a construction for the space-filling Peano curve, which has a dimension exactly 2). He notes that the approximation of these curves with segments of length G have lengths of the form  $G^{1-D}$ . The resemblance with Richardson's law is striking. Note that the paper does not claim that any coastline or geographic border actually is a fractal. Instead, he notes that Richardson's empirical law is compatible with the idea that geographic curves, such as coastlines, can be modelled by random self-similar figures of fractional dimension.Near the end of the paper Mandelbrot briefly discusses how one might approach the study of fractal-like objects in nature that look random rather than regular. For this he defines statistically self-similar figures and says that these are encountered in nature. The paper is important because it shows Mandelbrot's early thinking on <u>fractals</u>, and is an example of the linking of mathematical objects with natural forms that was a theme of much of his later work.

- List of countries by length of coastline
- <u>Benoît Mandelbrot</u>, 1967, *How Long Is the Coast of Britain? Statistical Self-Similarity and Fractional Dimension. Science*, New Series, Vol. 156, No. 3775. (May 5, 1967), pp. 636-638. doi:<u>10.1126/science.156.3775.636</u>