Deterministic chaos, fractals and diffusion: From simple models towards experiments

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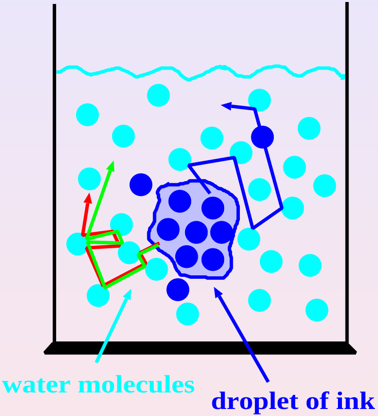
Outline

1. **Motivation:** random walks, diffusion and deterministic chaos
2. A simple model for **deterministic diffusion** with a **fractal diffusion coefficient**
3. From simple models towards experiments: **particle billiards** and **nanopores**

Microscopic chaos in a glass of water?

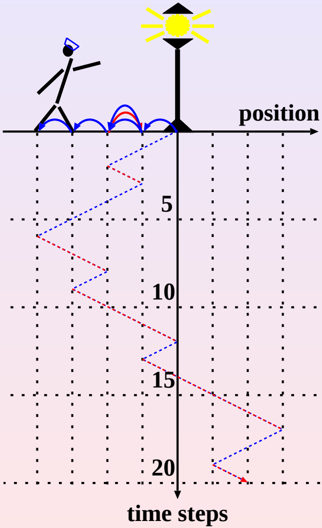


- dispersion of a droplet of ink by *diffusion*
- assumption: *chaotic collisions* between billiard balls

microscopic chaos
⇕
macroscopic transport

J.Ingenhousz (1785), R.Brown (1827), L.Boltzmann (1872), P.Gaspard et al. (1998)

The drunken sailor at a lamppost



simplification:
random walk in one dimension:

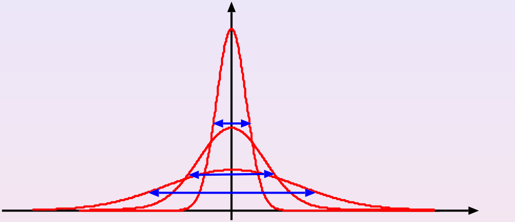
• steps of *length s* to the left/right

• sailor is **completely drunk**, i.e., the steps are *uncorrelated* (cp. to coin tossing)

K. Pearson (1905)

The diffusion coefficient

consider a large number (ensemble) of sailors starting from the same lamppost:



define the **diffusion coefficient** by the width of the distribution: it is a quantitative measure of how quickly a droplet spreads out

$$D := \lim_{n\to\infty} \frac{< x^2 >}{2n} \text{ with } < x^2 > := \int dx\; x^2 \rho_n(x)$$

as the *second moment* of the particle density ρ at time step n

A. Einstein (1905)

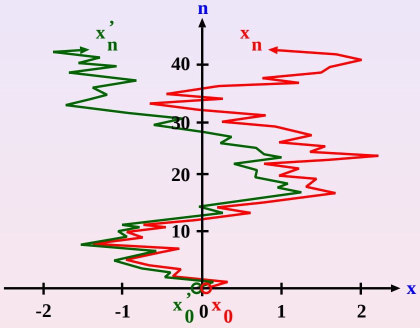
Basic idea of deterministic chaos

drunken sailor with memory? modeling by **deterministic chaos**

simple equation of motion

$$x_{n+1} = M(x_n)$$

for position $x \in \mathbb{R}$
at discrete time $n \in \mathbb{N}_0$
with chaotic map $M(x)$



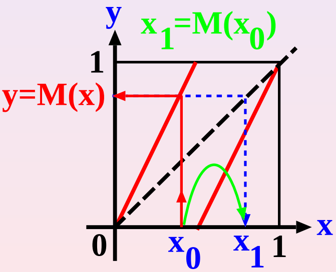
- the starting point **determines** where the sailor will move
- **sensitive dependence** on initial conditions

Dynamics of a deterministic map

goal: study **diffusion** on the basis of **deterministic chaos**

key idea: replace **stochasticity** of drunken sailor by **chaos**
why? determinism preserves all **dynamical correlations**!

model a single step by a **deterministic map**:



steps are iterated in discrete time according to the equation of motion

$$x_{n+1} = M(x_n)$$

with

$$M(x) = 2x \bmod 1$$

Bernoulli shift

Quantifying chaos: Ljapunov exponents

Bernoulli shift dynamics again: $x_n = 2x_{n-1} \bmod 1$

what happens to small perturbations $\Delta x_0 := x_0' - x_0 \ll 1$?

use equation of motion: $\Delta x_1 := x_1' - x_1 = 2(x_0' - x_0) = 2\Delta x_0$

iterate the map:

$$\Delta x_n = 2\Delta x_{n-1} = 2^2 \Delta x_{n-2} = \cdots = 2^n \Delta x_0 = e^{n \ln 2} \Delta x_0$$

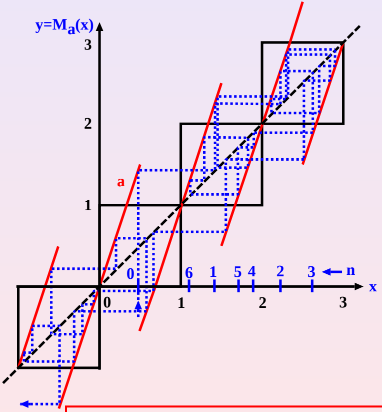
$\lambda := \ln 2$: **Ljapunov exponent**; A.M.Ljapunov (1892)

rate of **exponential growth** of an initial perturbation

here $\lambda > 0$: Bernoulli shift is **chaotic**

A deterministically diffusive model

continue the Bernoulli shift on a **periodic lattice** by *coupling* the single cells with each other; Grossmann, Geisel, Kapral (1982):



$$x_{n+1} = M_a(x_n)$$

equation of motion for **non-interacting point particles** moving through an array of identical scatterers

slope $a \geq 2$ is a **parameter** controlling the step length

challenge: calculate the **diffusion coefficient** $D(a)$

Computing deterministic diffusion coefficients

rewrite Einstein's formula for the diffusion coefficient as

$$D_n(a) = \frac{1}{2}\langle v_0^2 \rangle + \sum_{k=1}^{n} \langle v_0 v_k \rangle \to D(a) \quad (n \to \infty)$$

Taylor-Green-Kubo formula

with velocities $v_k := x_{k+1} - x_k$ at discrete time k and equilibrium density average $< \ldots > := \int_0^1 dx\, \varrho_a(x) \ldots\ ,\ x = x_0$

1. inter-cell dynamics: $T_a(x) := \int_0^x\, d\tilde{x} \sum_{k=0}^{\infty} v_k(\tilde{x})$ defines *fractal functions* $T_a(x)$ solving a (*de Rham*-) functional equation

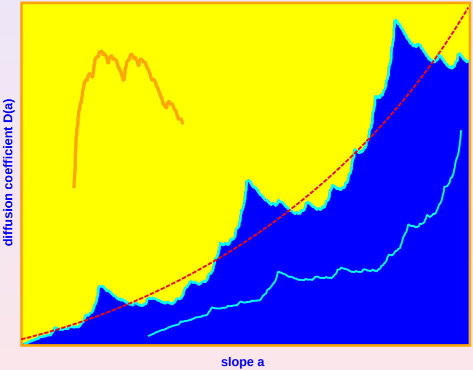
2. intra-cell dynamics: $\varrho_a(x)$ is obtained from the Liouville equation of the map on the unit interval

structure of formula:
first term yields **random walk**, others higher-order **correlations**

Parameter-dependent deterministic diffusion

exact analytical results for this model:

$D(a)$ exists and is a **fractal function of the control parameter**

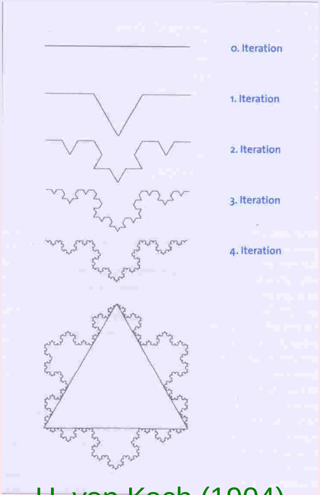


compare diffusion of drunken sailor with chaotic model:

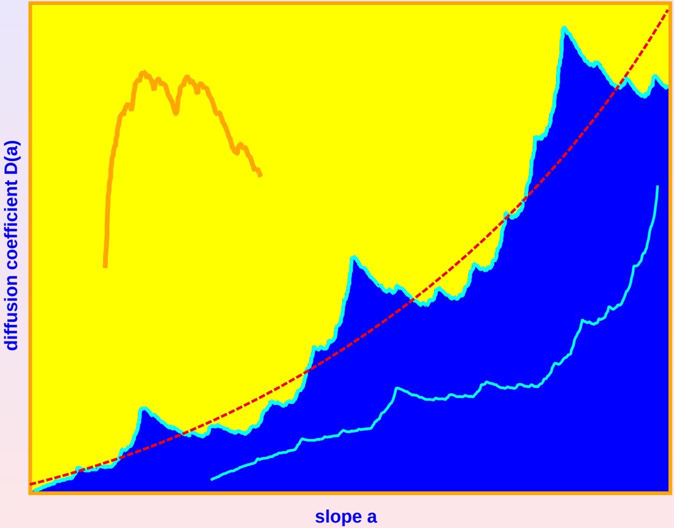
$\exists$ **fine structure beyond simple random walk solution**

R.K., Dorfman, PRL (1995)

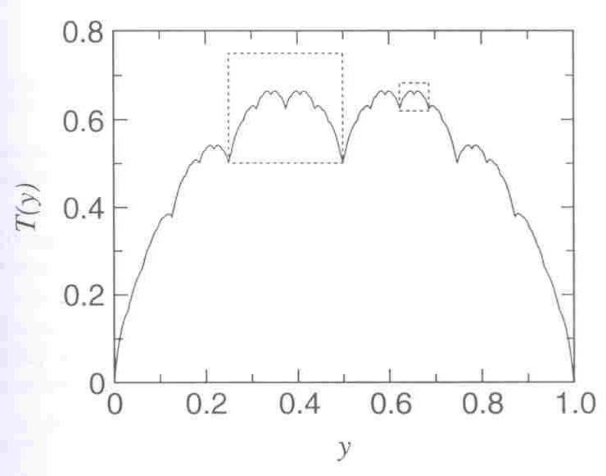
Fractals 1: von Koch's snowflake



H. von Koch (1904)

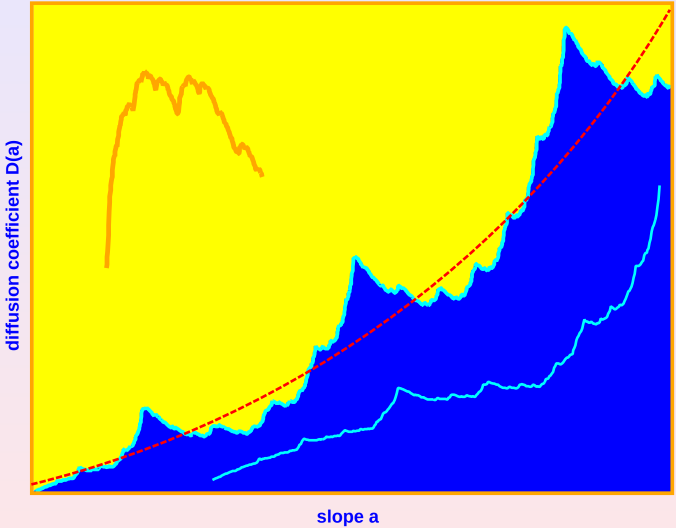

diffusion coefficient D(a)
slope a

Fractals 2: the Takagi function

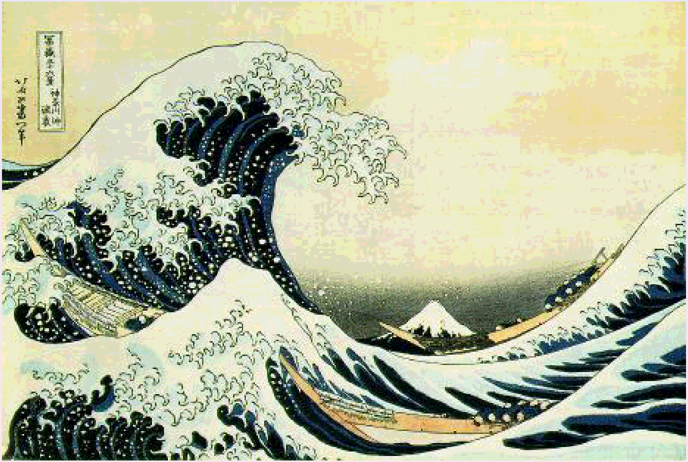


T.Takagi (1903)

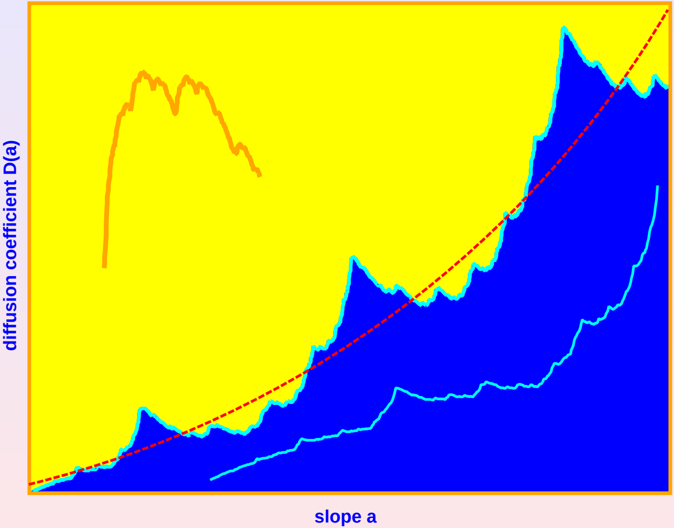
example of a continuous but nowhere differentiable function


diffusion coefficient D(a)
slope a

‘Fractals 3’: art meets science

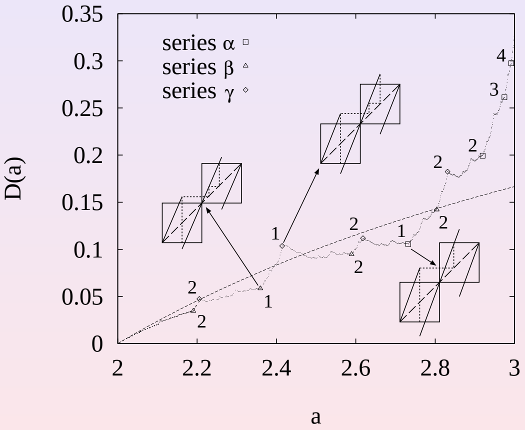


K.Hokusai (1760-1849)
The great wave of Kanagawa; woodcut


diffusion coefficient D(a)
slope a

Physical explanation of the fractal structure

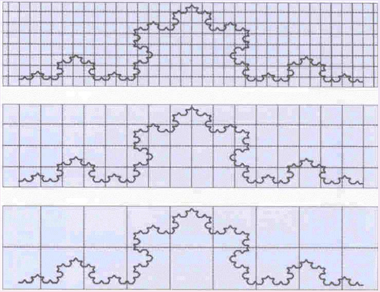
blowup of the initial region of $D(a)$:



local extrema are related to specific sequences of **correlated microscopic scattering processes**

Quantify fractals: fractal dimension

example: von Koch's curve; define a 'grid of boxes'



- count the number of boxes N *covering* the curve
- reduce the box size ϵ
- assumption: $N \sim \epsilon^{-\mathbf{d}}$

$$d = -\ln N / \ln \epsilon \; (\epsilon \to 0)$$

box counting dimension

- can be integer:
point: $d = 0$; line: $d = 1$; ...
- can be fractal:
von Koch's curve: $d \simeq 1.26$
Takagi function: $d = 1$ **!**
diffusion coefficient: $d = 1$ but

$$N(\epsilon) = C_1 \epsilon^{-1} (1 + C_2 \ln \epsilon)^{\alpha}$$

with $0 \leq \alpha \leq 1.2$ locally varying

Keller, Howard, R.K. (2008)

The flower-shaped billiard

deterministic diffusion in physically more realistic models:

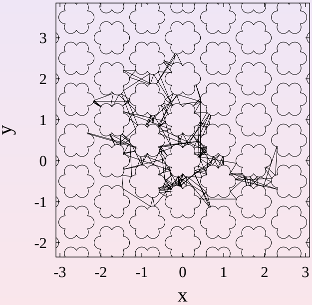
Hamiltonian particle billiards

example:

flower-shaped hard disks on a two-dimensional periodic lattice

moving point particles collide elastically with the disks *only*:

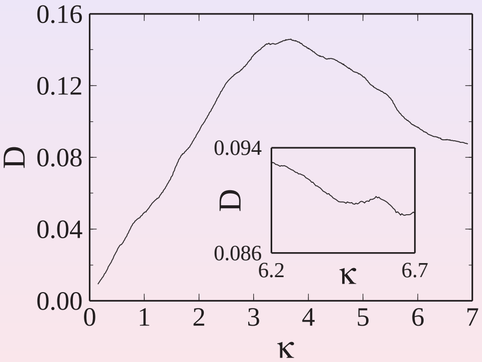
Knudsen diffusion (1909)



similar settings for electrons in semiconductor antidot lattices and for diffusion in porous media

Diffusion in the flower-shaped billiard

diffusion coefficient as a function of the curvature $\kappa = 1/R$ of the petals from simulations:



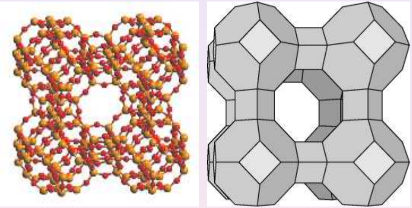
again a non-monotonic function of the control parameter with irregular structure on fine scales (fractal material?)

Harayama, R.K., Gaspard (2002)

Molecular diffusion in zeolites

zeolites: nanoporous crystalline solids serving as molecular sieves, adsorbants; used in detergents and as catalysts for oil cracking

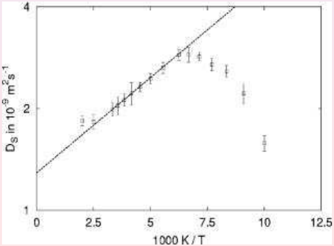
example: unit cell of Linde type A zeolite; strictly periodic structure built by a "cage" of silica and oxygen



Schüring et al. (2002): MD simulations with ethane yield non-monotonic temperature dependence of diffusion coefficient

$$D(T) = \lim_{t\to\infty} \frac{< [\mathbf{x}(t) - \mathbf{x}(0)]^2 >}{6t}$$

due to dynamical correlations



Summary

- **central theme:**

  relevance of **deterministic chaos**
  for **diffusion in periodic lattices**

- **main theoretical finding:**
  existence of diffusion coefficients that are irregular (fractal) functions under parameter variation, due to *memory effects*

  expected to be typical for classical transport in low-dimensional, spatially periodic systems

- **some generalizations:**
  - analogous theoretical results for other transport coefficients like conductivities and chemical reaction-diffusion
  - same phenomena in anomalous diffusion
  - irregularities are quite robust against random perturbations
- clearcut verification in **experiments?** good candidates are
  - nanopores
  - vibratory conveyors
  - antidot lattices
  - Josephson junctions

approach should be particularly interesting for *small systems*

Acknowledgements and literature

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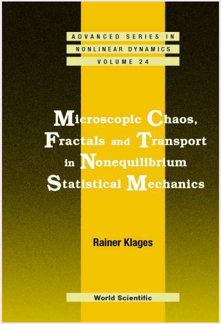
literature:



details and taster sections on www.maths.qmul.ac.uk/˜klages