Physical Measurements in the 21st Century

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June 7, 2004

Evolution in physical science

macro $\rightarrow$ micro $\rightarrow$ nano

describe $\rightarrow$ understand $\rightarrow$ design

How do we get experimental information from a nanostructure?

↓

by interaction with particles

Ideal experiment:

Strongest interacting particles

Single particle detection

Noise only due to counting statistics

What is understanding?

Fitting a physical model to the experimental data

What is scientific progress?

Improving the physical model

Evolution in theoretical materials science

- Ultimate physical model: atom positions and types
- Prediction of properties from “first principles”
- Required precision: 0.01 Å

experiment → ← theory

Ideal particles: electrons

- strong interaction → nanostructures
- sub surface information
- easy to detect
- use of lenses (real space ↔ Fourier space)
- bright sources “A synchrotron in the electron microscope” [1]
- less radiation damage than X-rays [2]
- sensitive to ionization of atoms [3]

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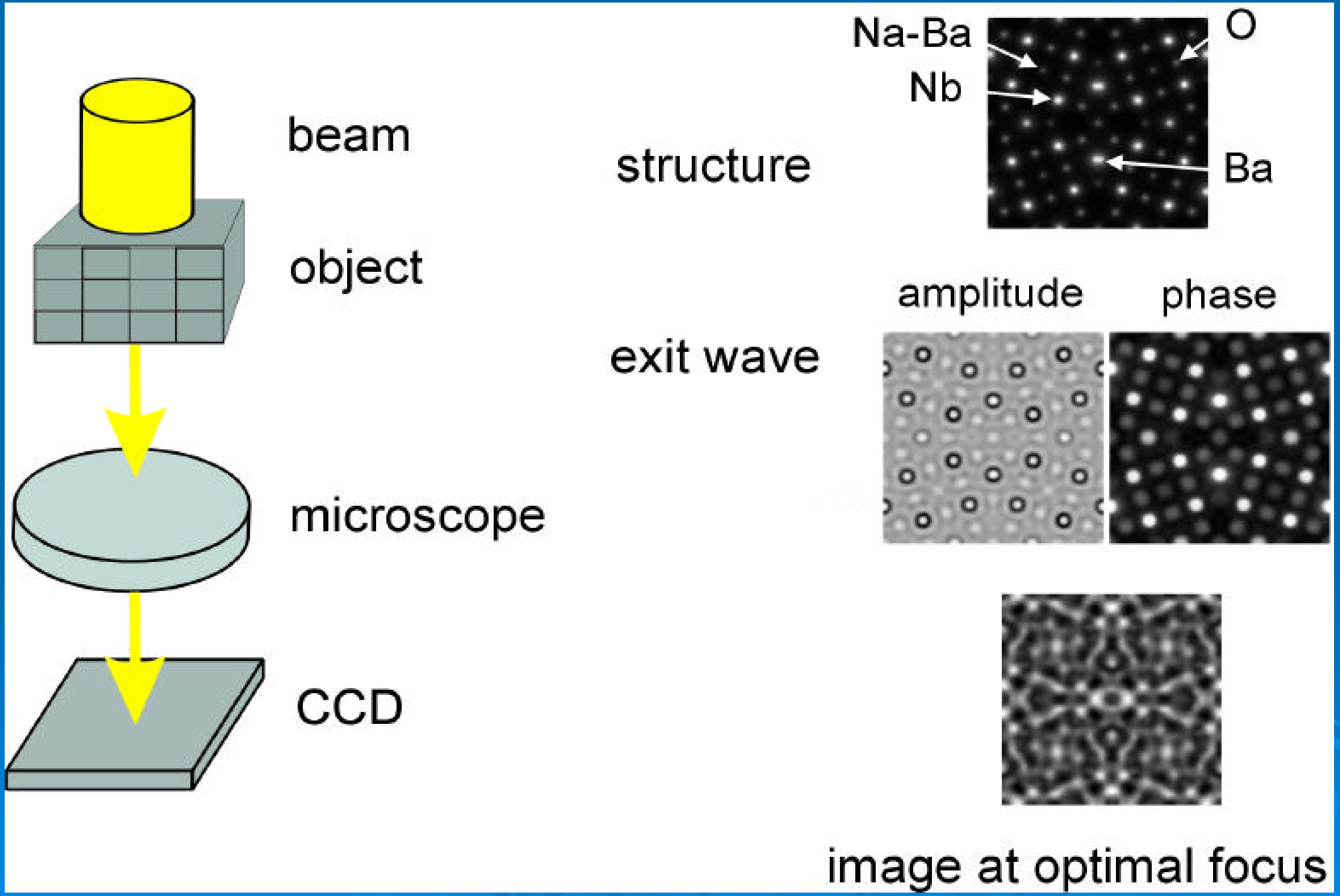
[1] M. Brown
[2] R. Henderson
[3] J. Spence

Electron microscope



Electron microscope = coherent imaging

Image wave = object wave * impuls response

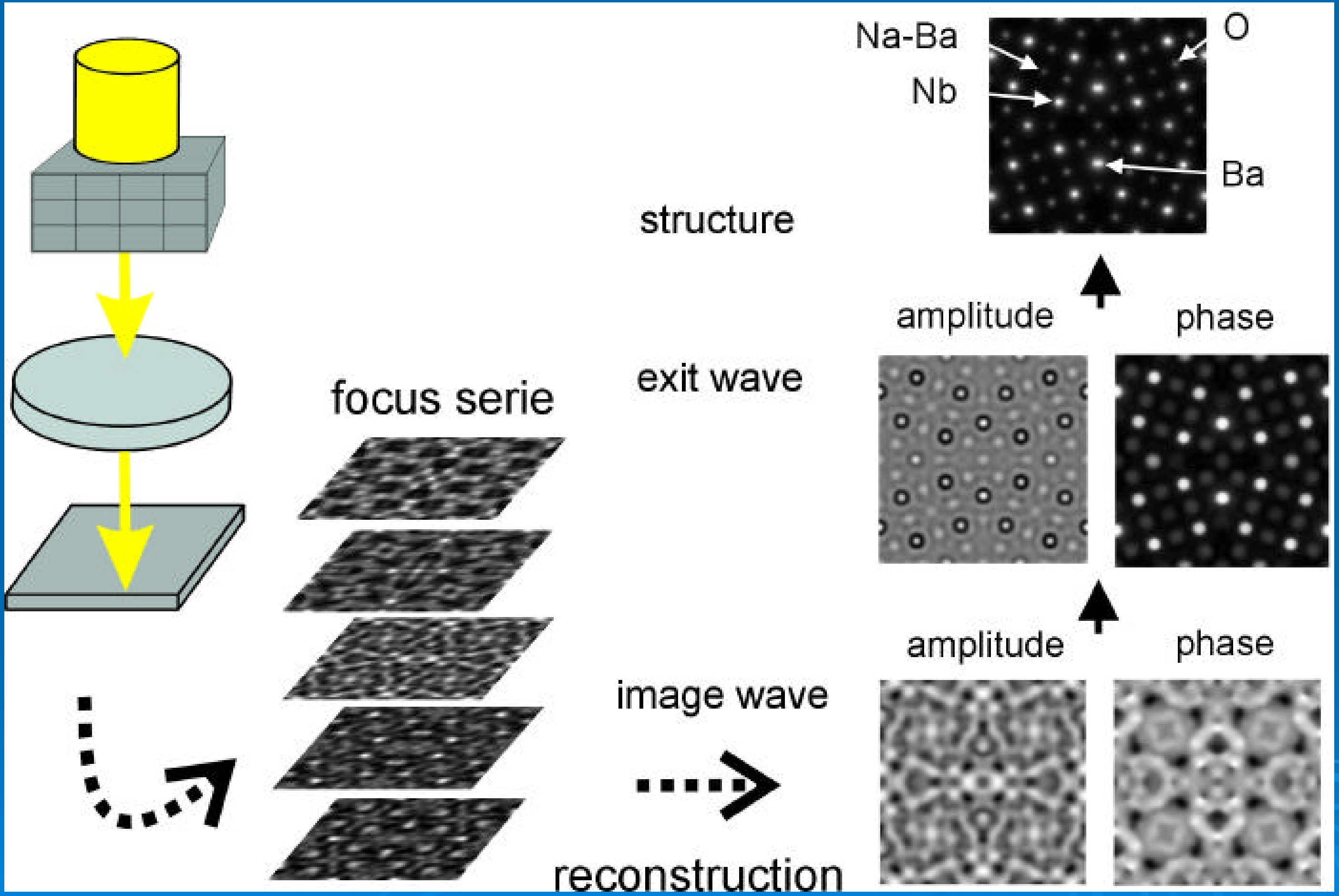
$$\Psi_{IM} = \Psi_{OB} * P$$

$$I_{IM} = |\Psi_{IM}|^2$$

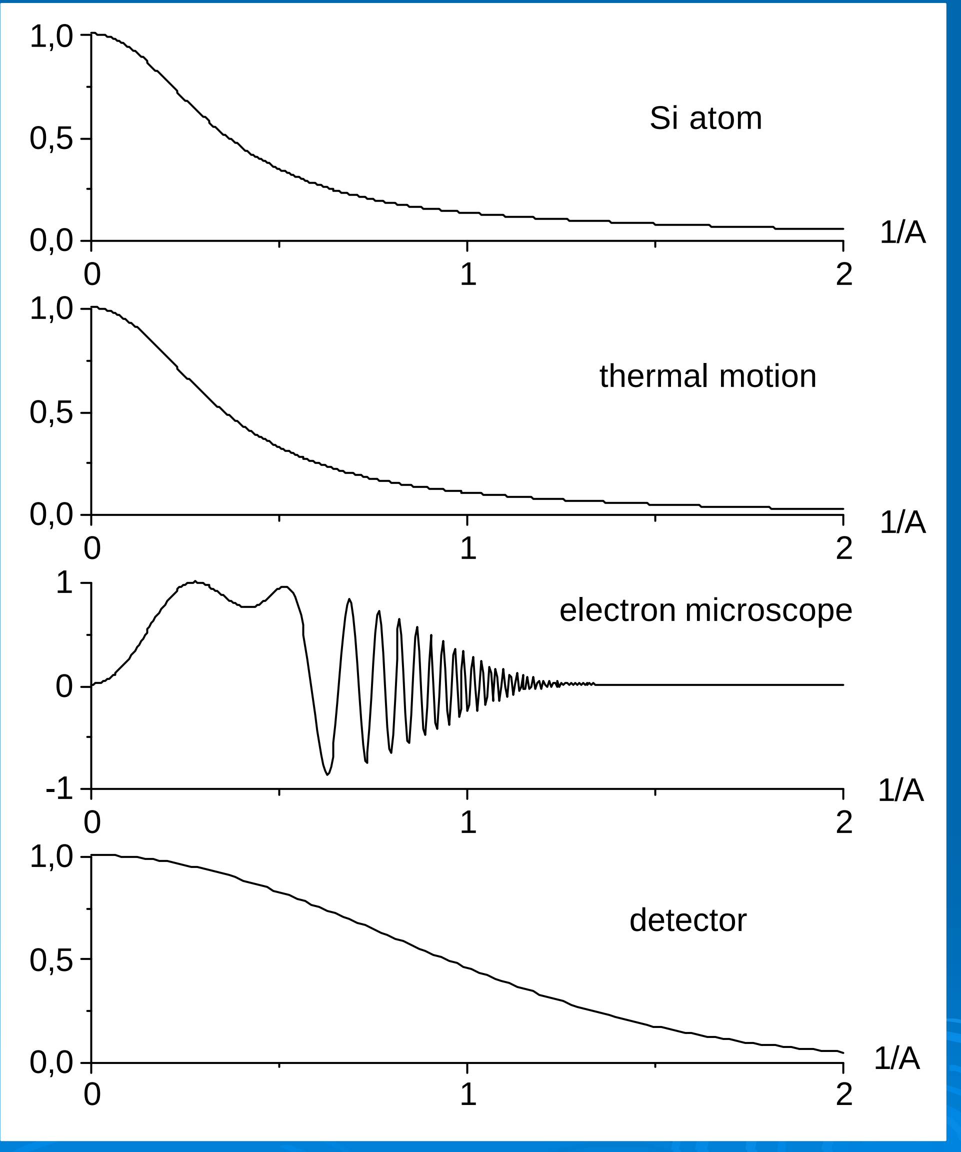
Deblurring (deconvolution) of the electron microscope

1) retrieve image phase: holography
2) deconvolute the impulse response function
3) reconstruct exit (object) wave

Focus variation method



Transfer functions



Ultimate resolution = atom

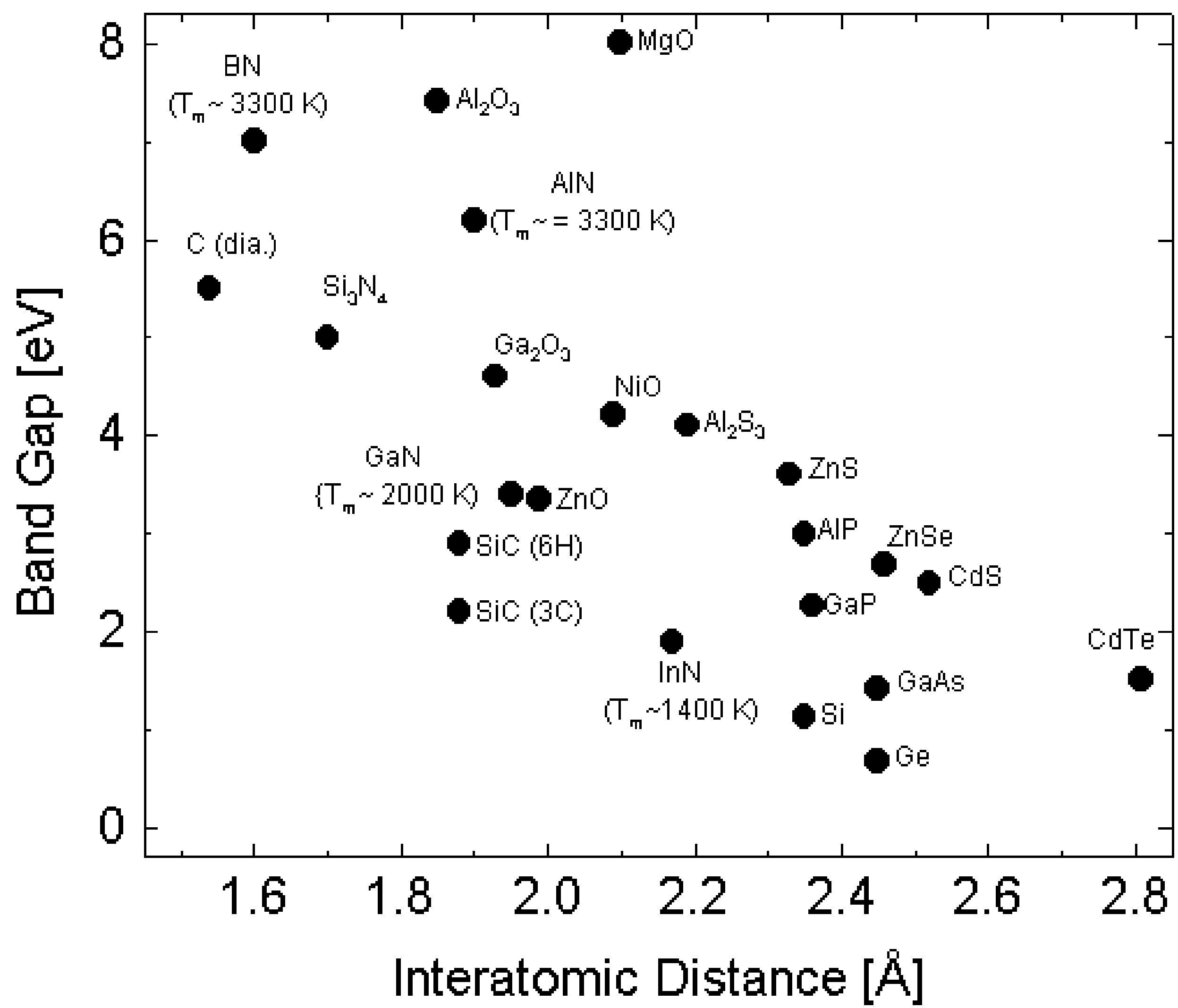
Resolving atoms = new situation

↓

Model based fitting (quantitative)

↓

resolution	precision
↕	↕
resolving	refining
↓	↓
1 Å	0.01 Å
resolution	precision



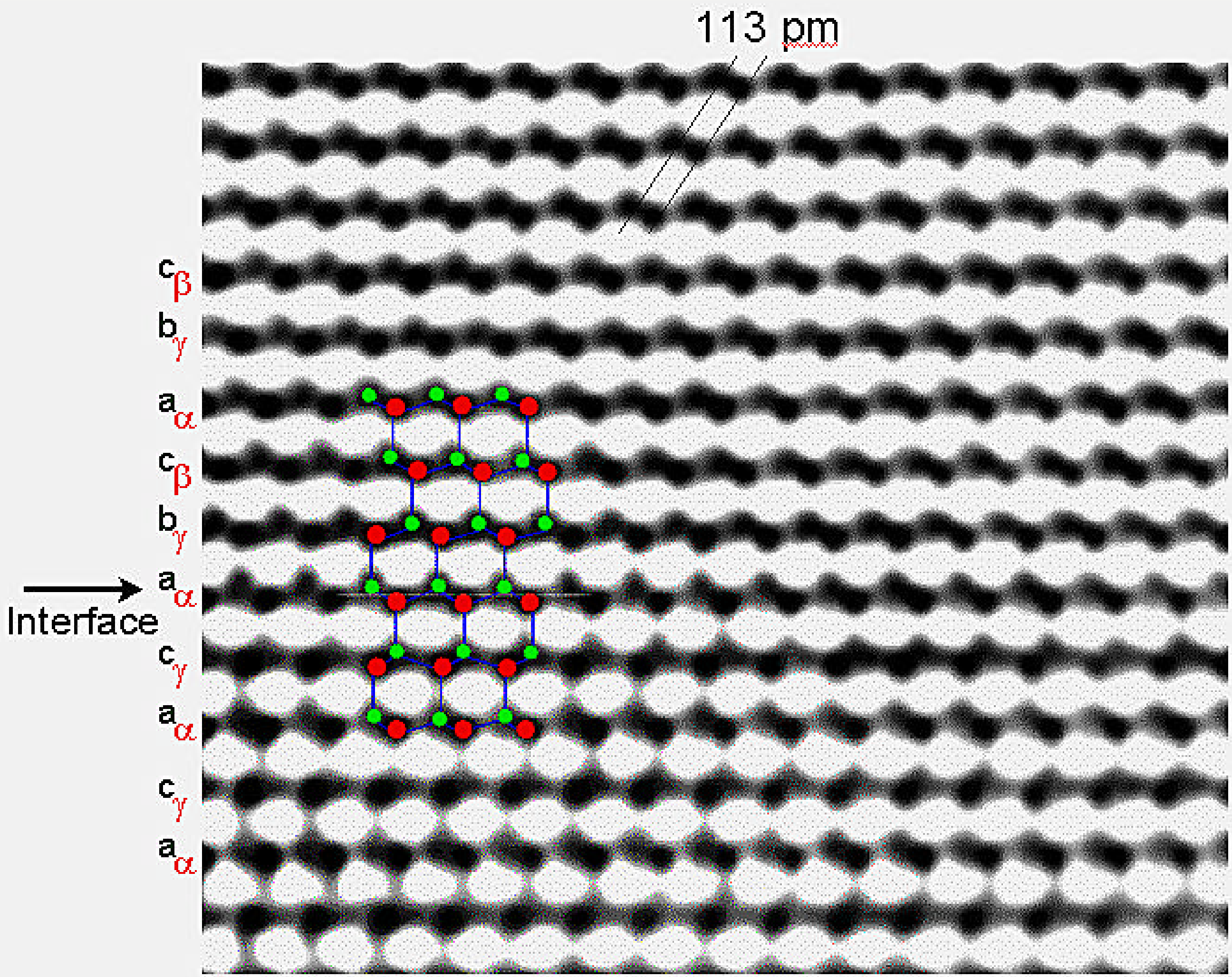
8
6
4
2
0
Band Gap [eV]
1.6
1.8
2.0
2.2
2.4
2.6
2.8
Interatomic Distance [Å]
MgO
BN
(T~m~ ~ 3300 K)
Al_2O_3
AlN
(T~m~ ~ = 3300 K)
C (dia.)
Si_3N_4
Ga_2O_3
NiO
Al_2S_3
GaN
(T~m~ ~ 2000 K)
ZnO
ZnS
AlP
ZnSe
SiC (6H)
CdS
SiC (3C)
GaP
CdTe
InN
(T~m~ ~1400 K)
GaAs
Si
Ge

Berkeley Lab
Materials Sciences Division

Exit Wave Function Reconstruction

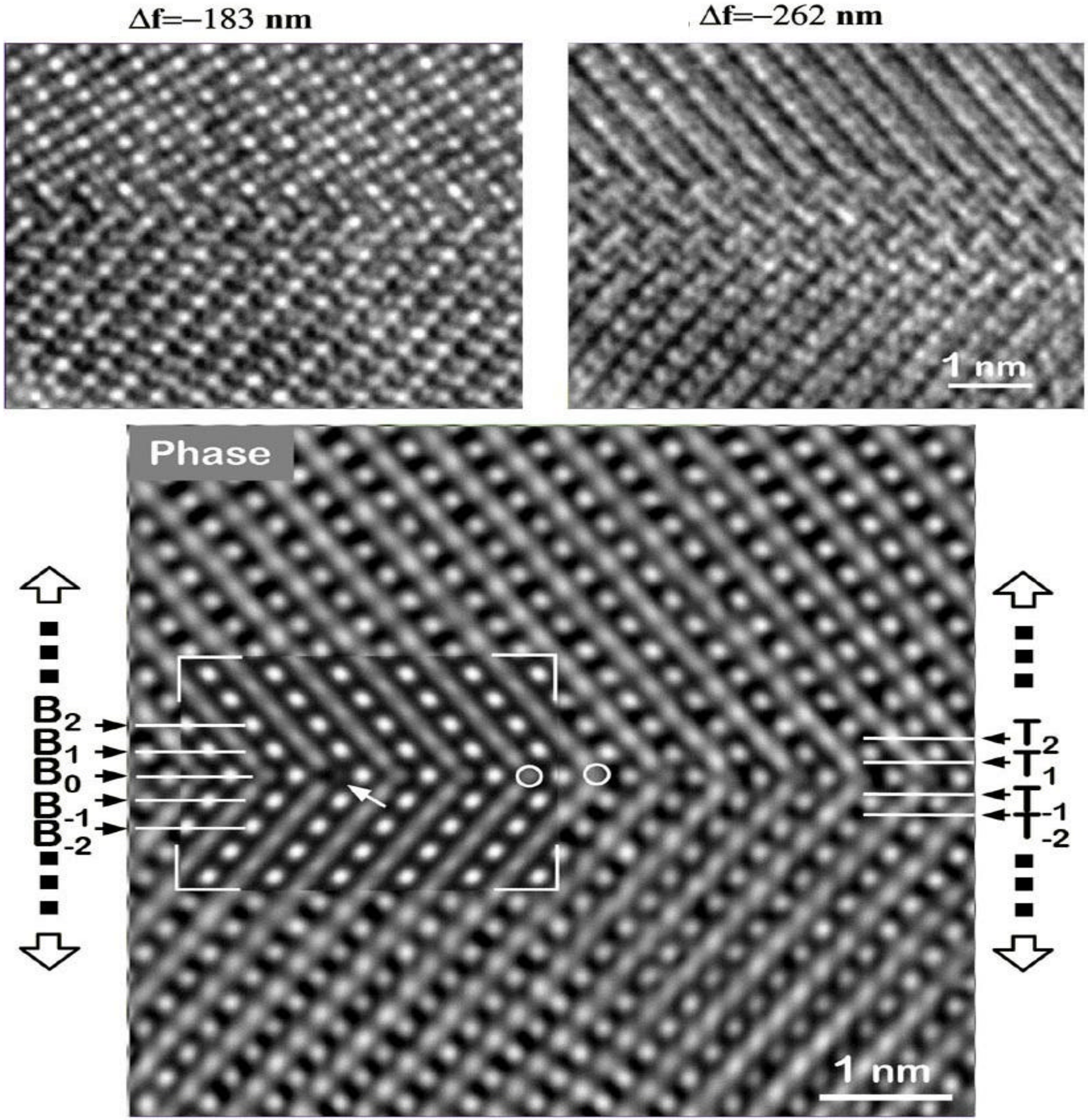
Interface: 3C-GaN / 2H-GaN

NCEM



- Reconstruction of electron exit waves with ultra-high resolution
  Information limit < 90 pm
- Separation of amplitude and phase of electron wave
  Here: Phase image
- Resolution of heavy (Ga) and light atoms (N) separated by 113 pm
- Detection of mono-atomic columns of light elements
  Here: Nitrogen
- Unambiguous determination of interface structure

C. Kisielowski, M.A. O'Keefe, C. Nelson, C. Song, R. Kilaas and A. Thust, 1999



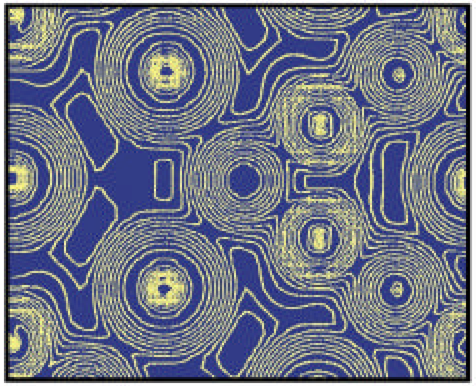
IFF/IMF C.L. Jia and A. Thust, Phys. Rev. Lett. 82 (1999) 5052.

Comparison: Theory & Experiment

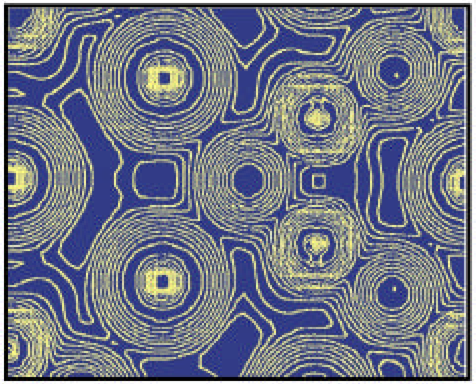
Plane Distances at Σ3(111) Twin Boundary

	Ti - Ti [pm]	Ba - Ba [pm]
Geometric	232	232
Experiment	270	216
Theory	267	214

Charge Density from First Principles Calculation:



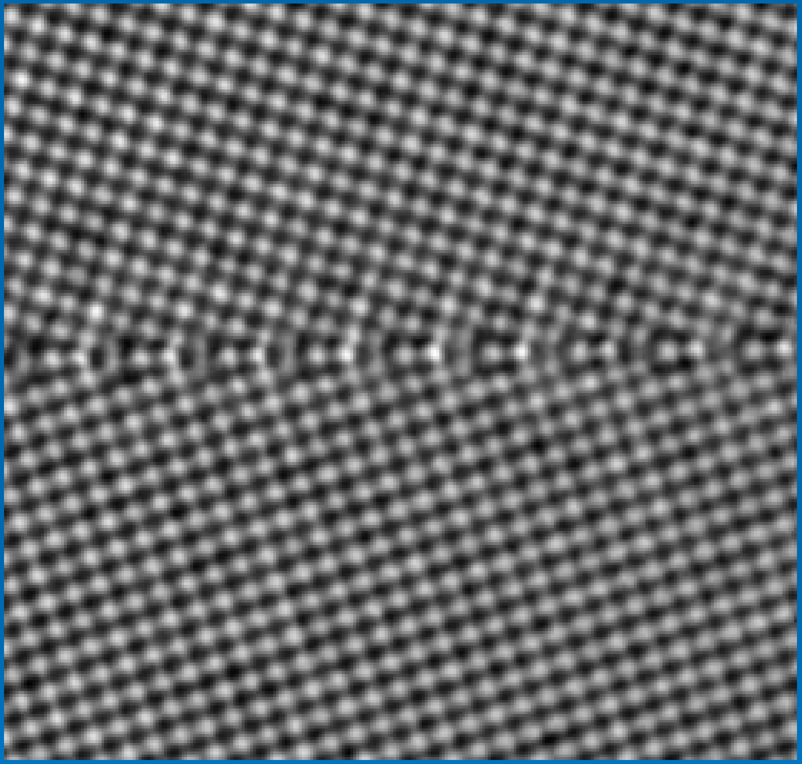
Ideal Geometrical Model



Optimized Structure

	Ideal Geometrical Model	Optimized Structure
Ti-Ti:	$4.5 \times 10^{-2}\ e\,/\mathrm{a.u.}^3$	$3 \times 10^{-2}\ e\,/\mathrm{a.u.}^3$
Ba-Ba:	$5 \times 10^{-3}\ e\,/\mathrm{a.u.}^3$	$9 \times 10^{-3}\ e\,/\mathrm{a.u.}^3$

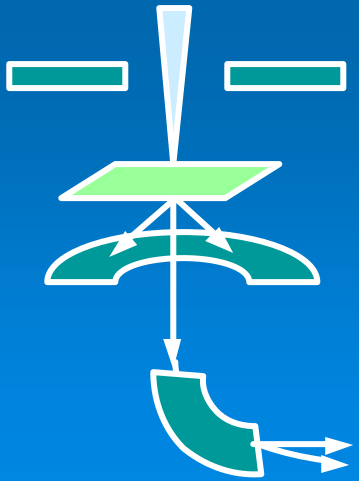
Phase of total exit wave
Σ 5 Al: Cu



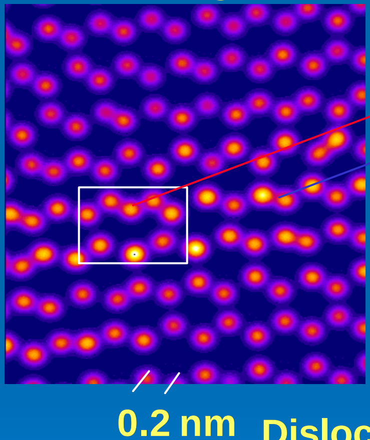
Courtesy C. Kisielowski (NCEM,Berkeley)

Spectroscopy in the electron microscope

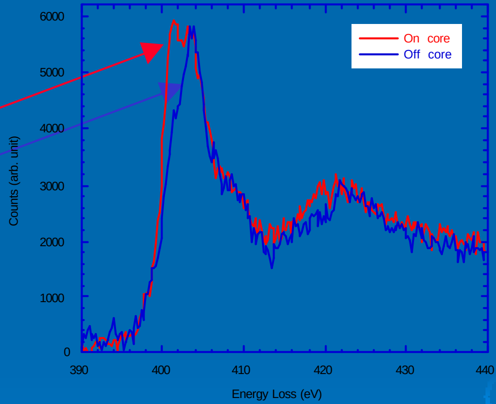
A STEM / HRTEM :



Current technology:
HAADF-image



Local energy spectrum

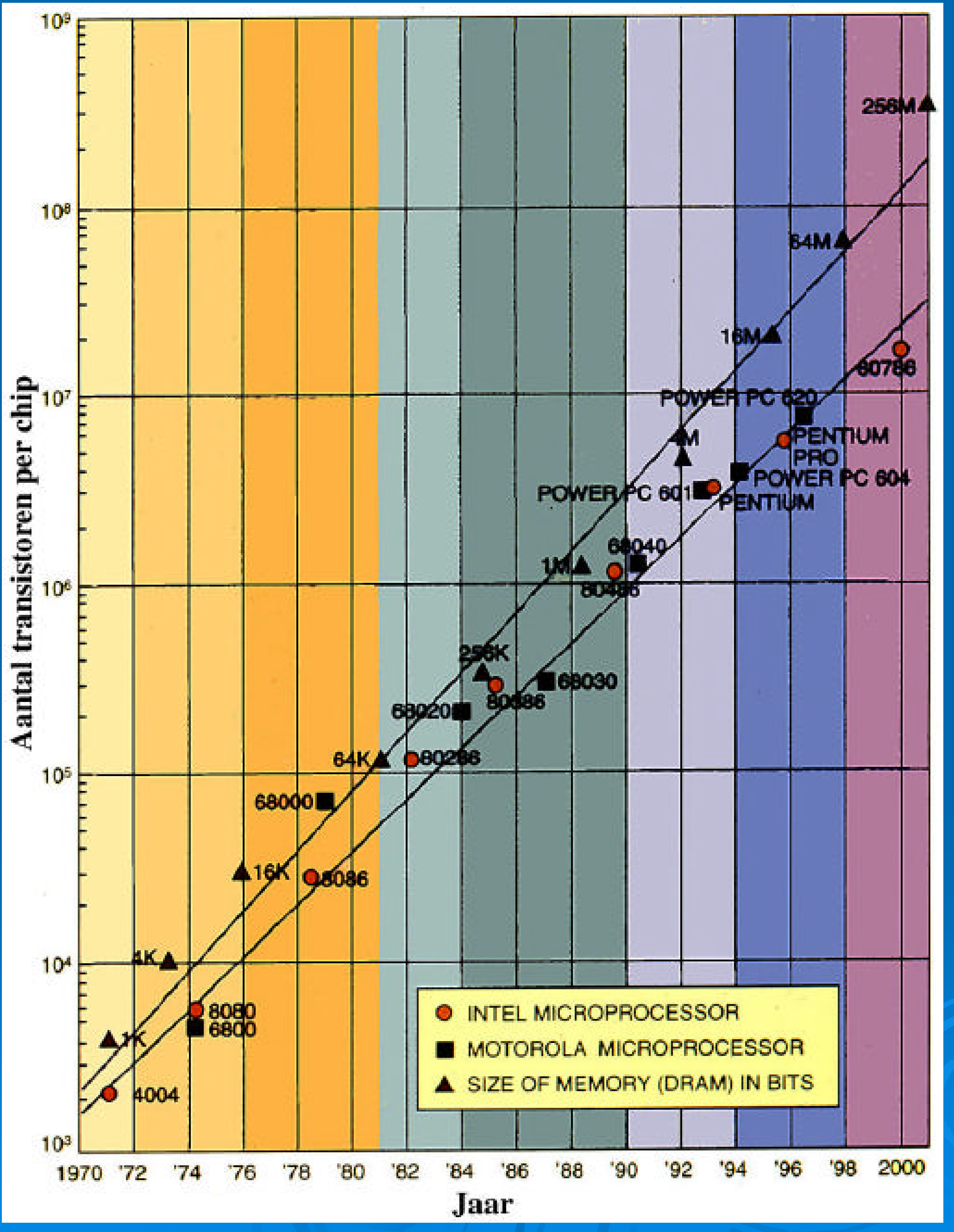


Dislocation core in GaN [0001]

N. Browning, C. Kisielowski, LDRD, 2002-2003

Spectral information:

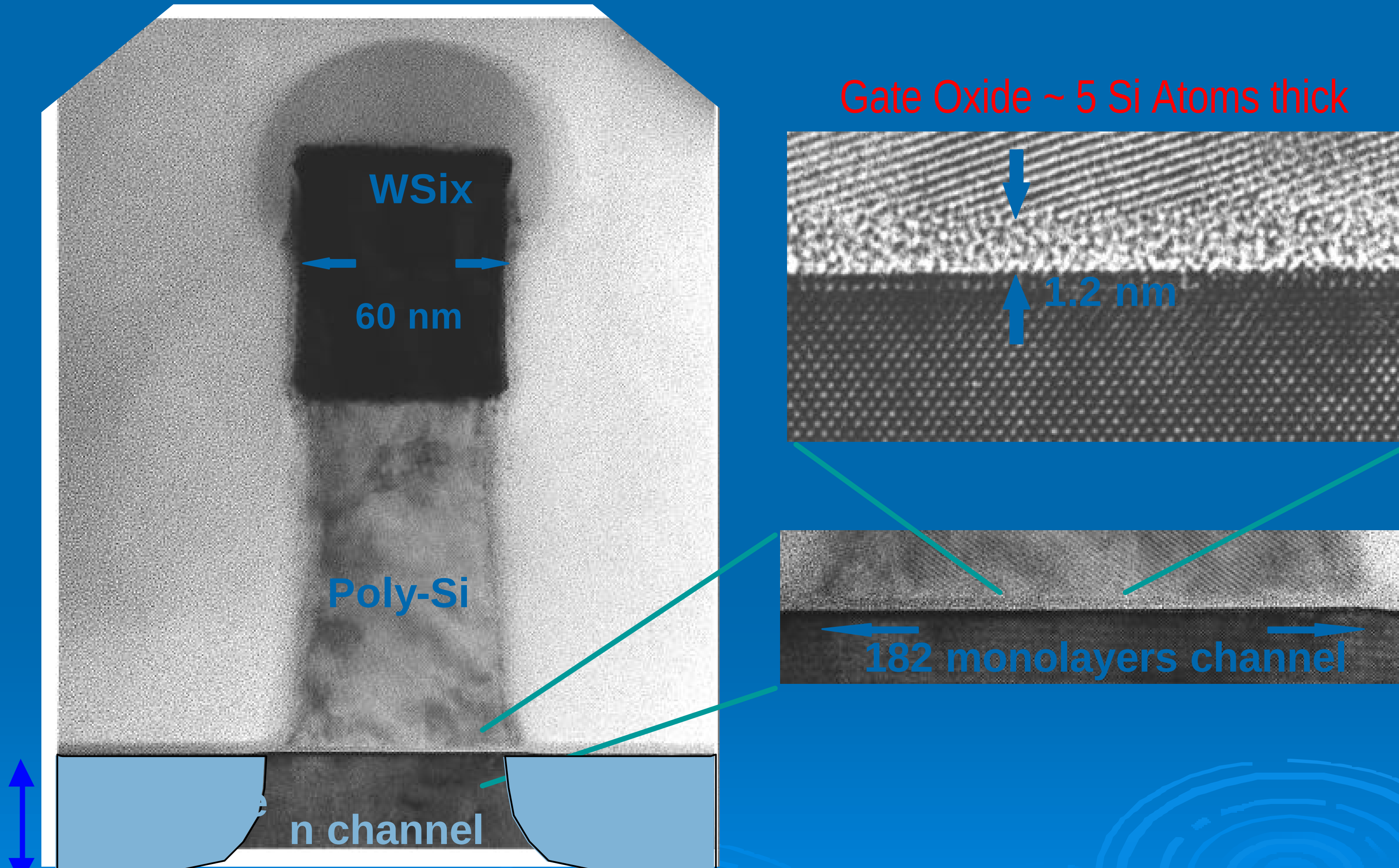
- Composition
- Bonding
- Electronic
- Local configuration



Aantal transistoren per chip
Jaar
1970
'72
'74
'76
'78
'80
'82
'84
'86
'88
'90
'92
'94
'96
'98
2000
4004
1K
4K
8080
6800
16K
8086
68000
64K
80286
68020
256K
80386
68030
1M
80486
68040
POWER PC 601
PENTIUM
POWER PC 604
PENTIUM PRO
POWER PC 620
16M
64M
256M
80786
INTEL MICROPROCESSOR
MOTOROLA MICROPROCESSOR
SIZE OF MEMORY (DRAM) IN BITS

Scaling Limits to CMOS Technology

(For proper operation, all vertical and lateral dimensions scale simultaneously)



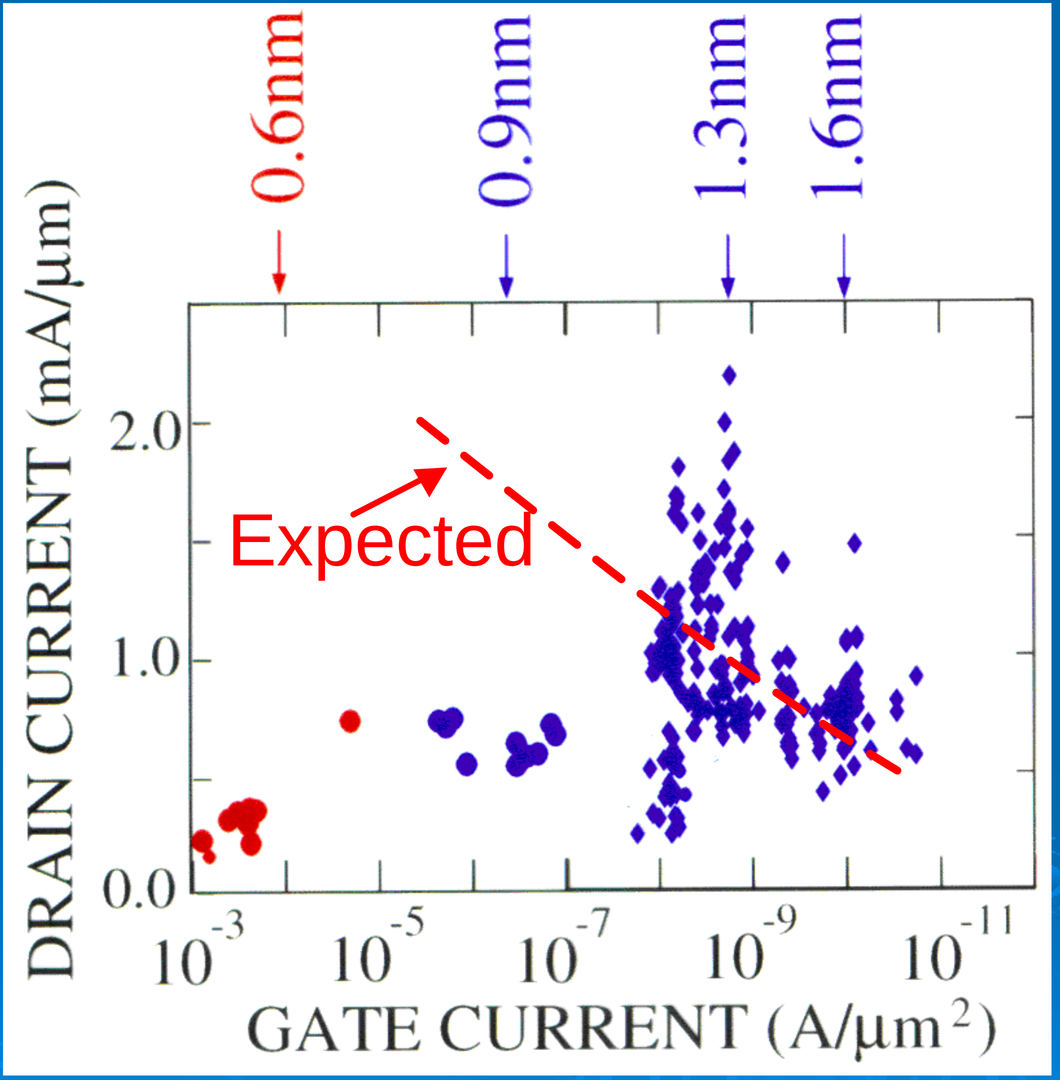
Shrinking the junction depth ➡ increasing the carrier concentration

TEM. F. H. Baumann

Lucent Technologies
Bell Labs Innovations

Gate Oxide Characterization

- What happens at 12Å?
  - Roughness?
  - Interlayers?
  - Interface Scattering?



- Is there a fundamental limit to the oxide thickness?

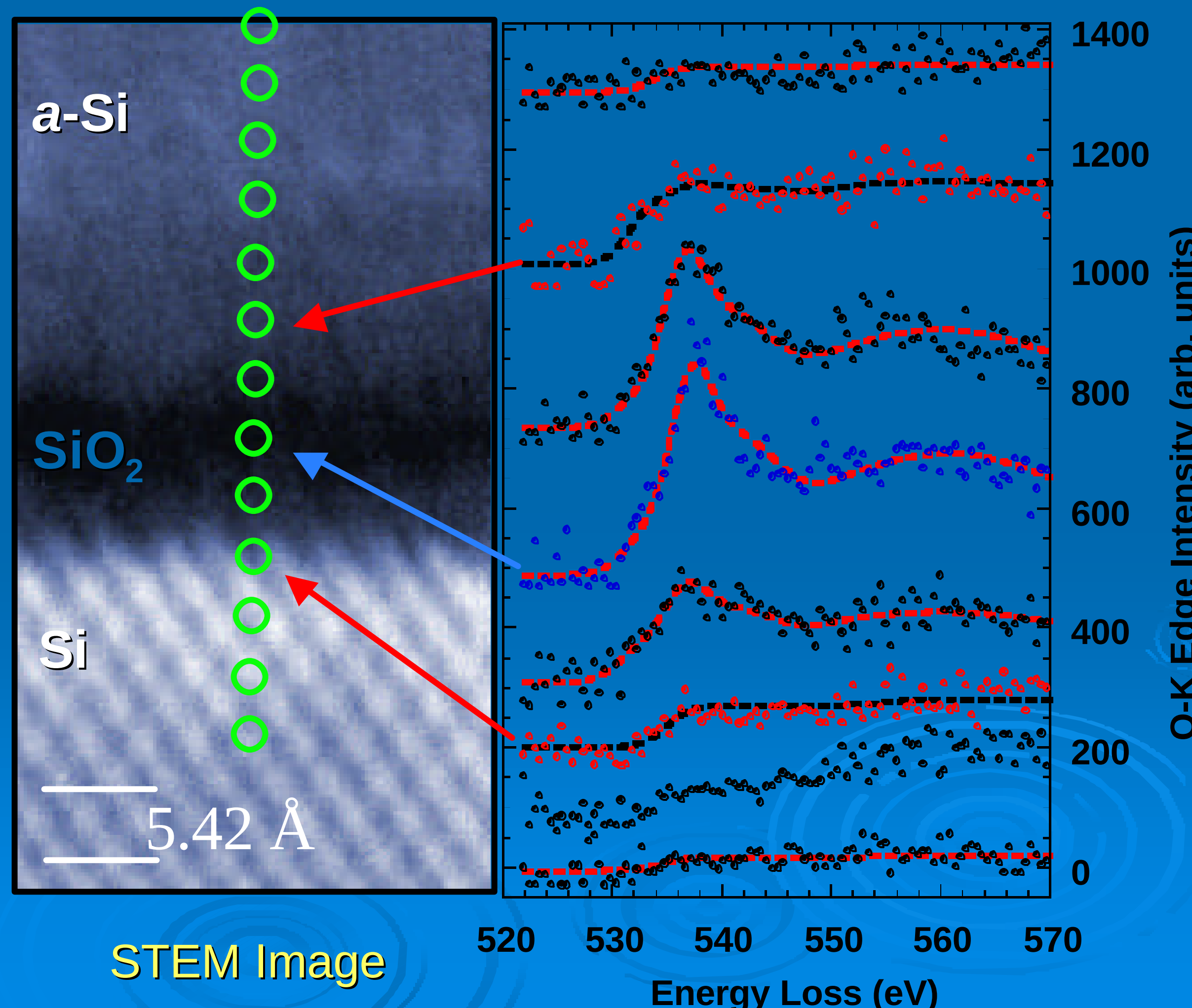
Oxygen Bonding from EELS (Chemistry on an Atomic Scale)

Nominal 1.1 nm SiO_2:

0.8 - 1 nm Bulk SiO_2

1.6 nm wide oxygen profile

Interface States



STEM Image

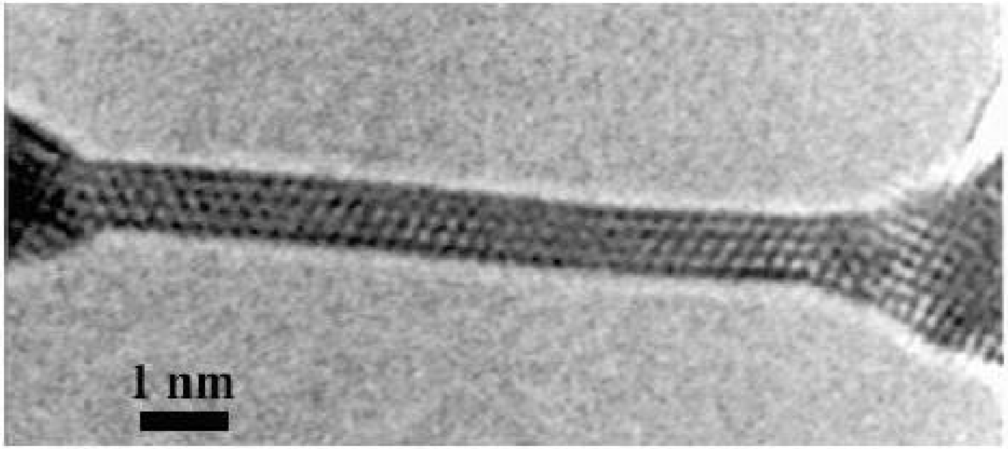
D. Muller

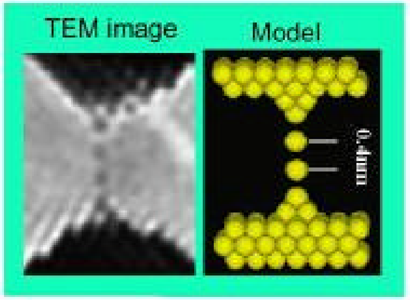
A nano lab in the microscope

In basic physics..

More space !

Quantization of conductance in nanowires.

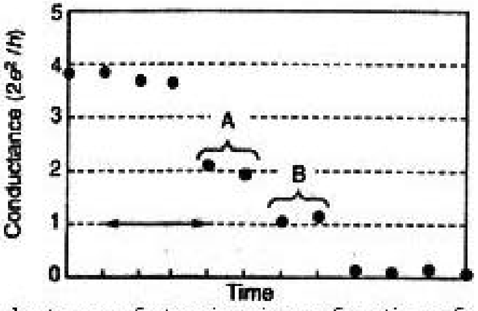




Single chain of gold atoms between tips.

TEM observation of quantized resistance in gold nanowire while imaging at atomic resolution, room temp. (Takayanagi, Nature 1998). This establishes equipartition for electron transport in the quantum regime.

Conductance is measured to be $2e^2/h = 13\ k\Omega$ per string of atoms. Stretching chain with piezo changes number of atomic strings, down to single-atom string (right).
Number of atomic strings in chain is counted from image.
Chains seen differ from "neck" assumption of STM work.



Conductance of atomic wire as function of time, during which number of atom strings in wire changes, showing quantization.

Ballistic, Transverse Fermi wavelength, energies, quantized, Landauer, Kubo.

Statistical experimental design

What is the optimal experiment?

↓

The experiment that yields the most precise model parameters for a given particle (electron) dose

Theory

1. parametric statistical model of the observations (single particle detection)

$$P\left(\omega;\theta\right)=\prod_{m=1}^{M}\frac{\lambda_m^{\omega_m}}{\omega_m!}\exp\left(-\lambda_m\right)$$

observations $\omega=\left(\omega_1 \ldots \omega_M\right)^T$

expectation model $E\left[w_m\right]=\lambda_m=\phi_m\left(\theta\right)$

parameters $\theta=\left(\theta_1 \ldots \theta_T\right)^T$

2. Fisher information matrix

$$F = -E\left[\frac{\partial^2 \ln P(w;\theta)}{\partial\theta\,\partial\theta^T}\right]$$

$$F_{rs} = \sum_{m=1}^{M} \frac{1}{\lambda_m}\frac{\partial\lambda_m}{\partial\theta_r}\frac{\partial\lambda_m}{\partial\theta_s}$$

3. Cramér-Rao Lower Bound (CRLB) F^{-1}

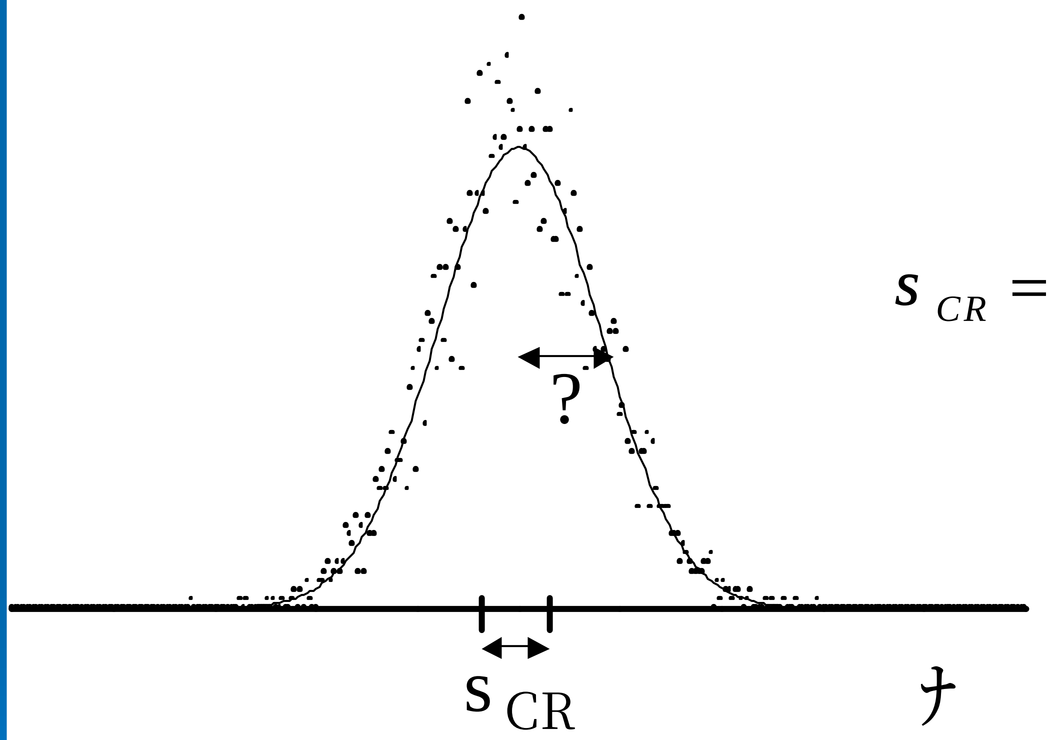
theoretical lower bound on the variance of the parameter estimates for any unbiased estimator $\widehat{\theta}$

$$\boxed{cov(\widehat{\theta}) \geq F^{-1}}$$

Optimal experimental or instrumental design

1. CRLB is computed
2. CRLB is minimized with respect to the microscope settings (for a fixed electron dose)
3. These settings are suggested as optimal

Simplified example



$$s_{CR} = \frac{r}{\sqrt{N}}$$

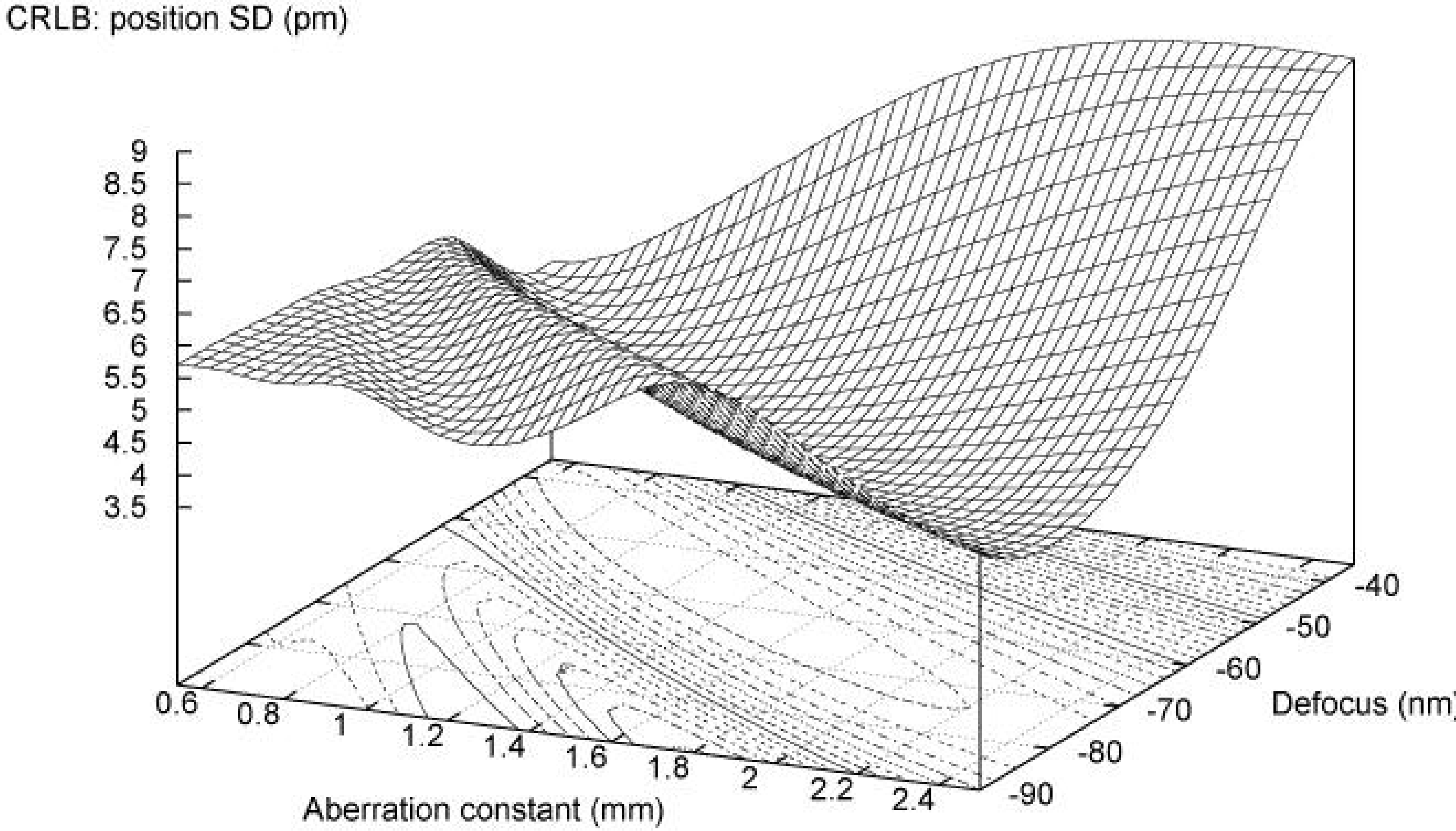
resolution

dose

? = 1 ナ

N= 10000

s_{CR}= 0.01 ナ



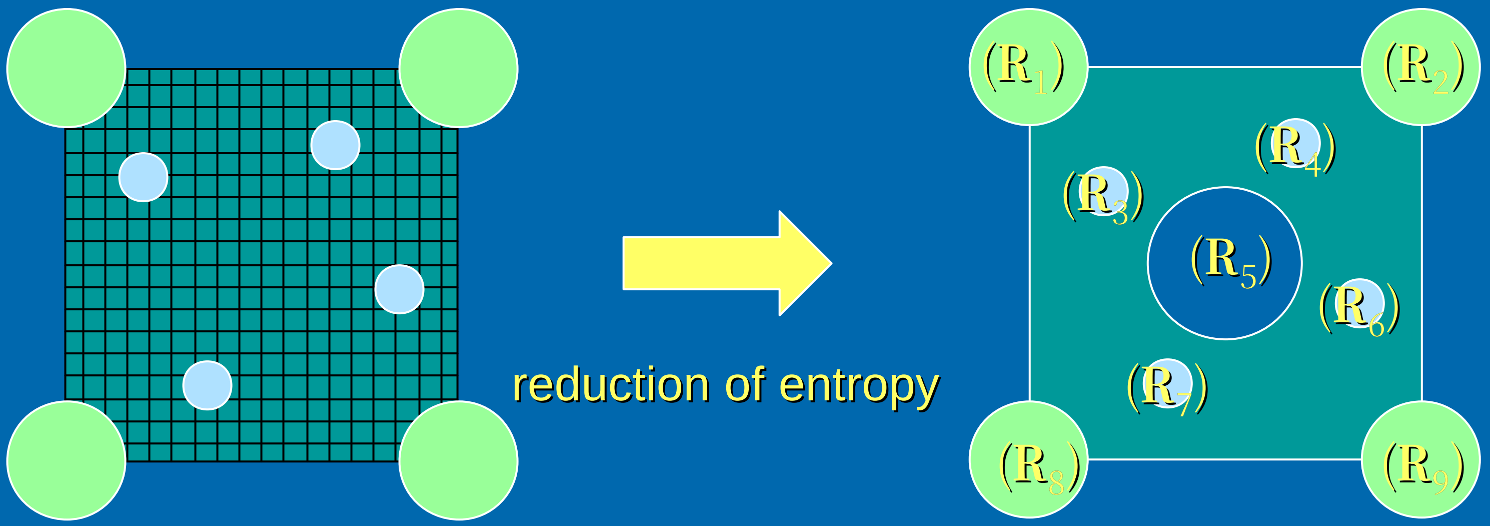
CRLB: position SD (pm)
9
8.5
8
7.5
7
6.5
6
5.5
5
4.5
4
3.5
0.6
0.8
1
1.2
1.4
1.6
1.8
2
2.2
2.4
Aberration constant (mm)
-40
-50
-60
-70
-80
-90
Defocus (nm)

Towards an automatic interpretation

Why do we do experiments?

- to understand
- to find underlying laws
- to find hidden order
- to reduce the apparent entropy in our experimental data set

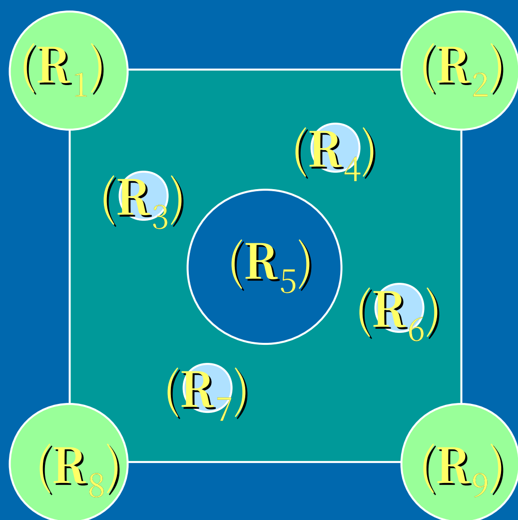
Example:crystal unit cell



sampled unit cell

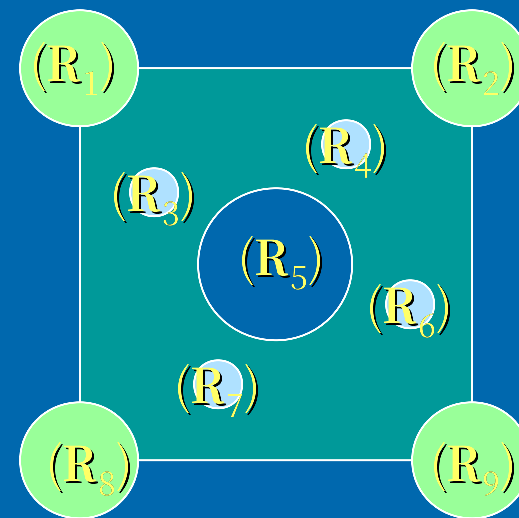
atom positions
and types

Example:crystal unit cell



atom positions
and types

reduction of entropy



chemical laws

Can science be automated?

- Finding laws
- Pattern recognition
- Artificial intelligence

Will it still be fun?

Acknowledgements

- A.J. den Dekker (DCSC, Delft University of Technology)
- A. van den Bos (Faculty of Applied Sciences, Delft University of Technology)