

Wiederholung Diffusion

- kT (Arrhenius)
→ Random Walk / Lattice Gas / 'on site'
- Fick's laws
- Diffusion affected by cooperativity
- Atomistic Mechanisms
- Elektromigration / Stark Effect

Repetition PEEM / LEEM



- Magnetic domains
- Slow electrons (surface/interface sensitivity)
- PEEM with soft X-rays as source (XMCD image)
- Polarized X-ray source
- PEEM with slow electrons as source
- Research example nanocrystals



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Repetition XAS, NEXAFS, XMCD



- Near Edge X-ray Absorption Fine Structure
- reflects density of unoccupied states
- Absorption processes and decay (soft X-rays)
- Sampling depths (total electron yield, secondary, Auger, Fluorescence)
- Multiplet structure (chemical, electronic sensitivity)
- XMCD (sum rules)
- XMLD
- Magnetism (spin and orbital moment, magnetocrystalline anisotropy)



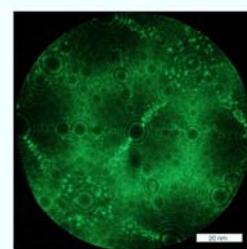
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Experimental Techniques

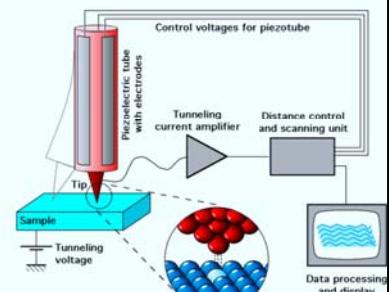
1) Direct observation:

- Field ion microscopy (FIM)
→ "image-anneal-image" technique
- Limited to refractory or noble metal surfaces



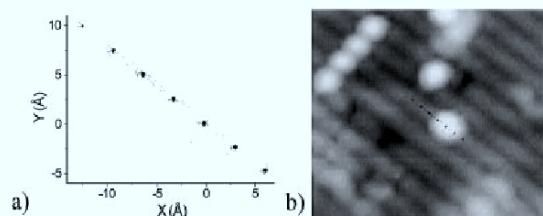
- Scanning tunneling microscopy (STM)
→ "image-while-hot" technique

- STM "movies" can be recorded
(at 0.01-1 frames per second)



Experimental Techniques

- Scanning tunneling microscopy (STM) → “atom-tracking” technique
 - STM tip locked onto an adparticle by 2D lateral feedback
 - Example: Si on Si(100) (Swartzentruber, PRL 1996)

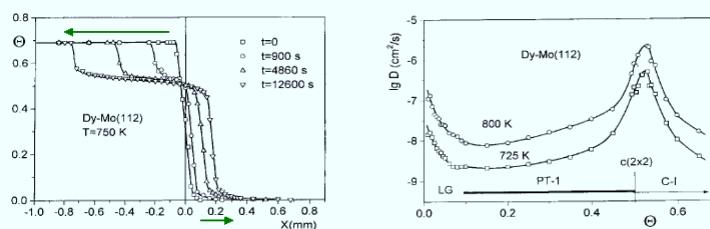


- Electric fields from the STM tip influence surface diffusion!

Experimental Techniques

2) Profile evolution method:

- Smearing of a sharp initial concentration profile is monitored
 - Initial profile deposited using a mask
 - AES, SIMS, SEM or local work-function...
 - $D(\Theta)$ can be evaluated
- Example: Dy on Mo(112) (Loburets et al., SS 1998)



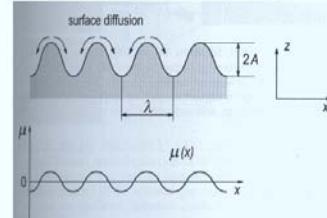
Experimental Techniques

3) "Capillarity" techniques:

- A surface is perturbed from its lowest energy configuration...
... and allowed to relax via diffusion
- Relaxation rate → Coefficient of diffusion

- For a sinusoidal profile (Mullins, JAP 1999):

$$A(t) = A_0 \exp \left[-\frac{\gamma D n_0 V^2}{k_B T} \left(\frac{2\pi}{\lambda} \right)^4 t \right]$$

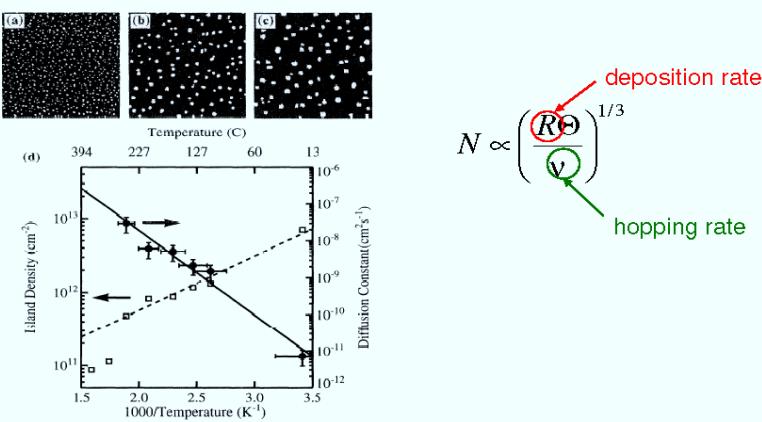


γ = surface tension; V = atomic volume; n_0 = surface density

Experimental Techniques

4) Island growth techniques:

- Number density of islands after submonolayer deposition is monitored
- Example: Fe on Fe(100) (Stroscio et al., PRL 1993)

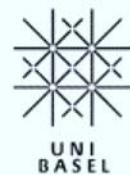


Further Reading

- K. Oura et al., Surface Science, Springer 2003, chapter 13
- A.G. Naumovets & Yu.S. Vedula, Surf. Sci. Rep. 4 (1985) 365
- R. Gomer, Rep. Prog. Phys. 53 (1990) 917
- G.L. Kellogg, Surf. Sci. Rep. 21 (1994) 1

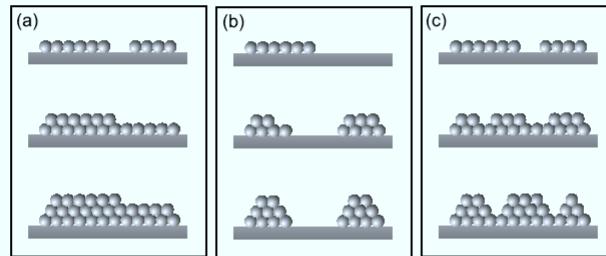
Surface Physics 2010

Growth of Thin Films



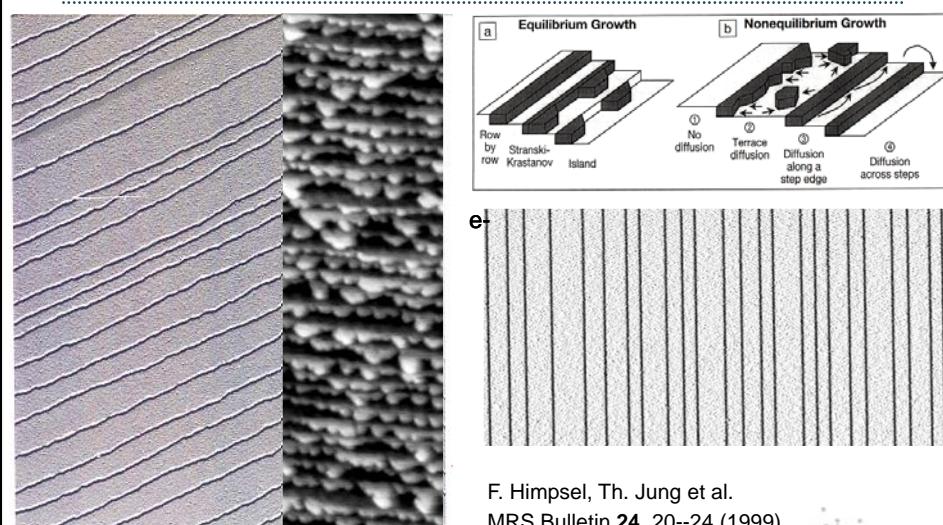
Lecturer: Dr. Enrico Gnecco
NCCR Nanoscale Science

Growth Modes



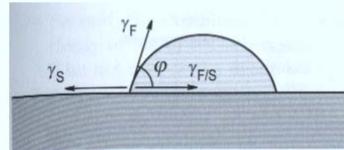
- (a) Layer-by-layer or **Frank-van der Merve** mode → 2D islands
- (b) Island or **Vollmer-Weber** mode → 3D islands
- (c) Layer plus Island or **Stranski-Krastanov** mode → 2D layer + 3D islands

'Physical' Self Assembly of e.g. Nanowires



Growth Modes

- Surface tension γ = work required to build a surface of unit area
(\equiv force per unit length)

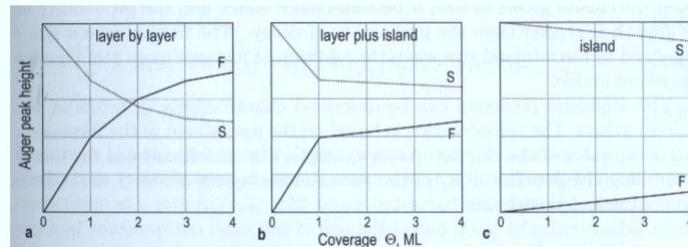


$$\gamma_S = \gamma_{SF} + \gamma_F \cos \varphi$$

- Island growth: $\varphi > 0 \rightarrow \gamma_S < \gamma_{SF} + \gamma_F$
- Layer-by-layer growth: $\varphi = 0 \rightarrow \gamma_S \geq \gamma_{SF} + \gamma_F$

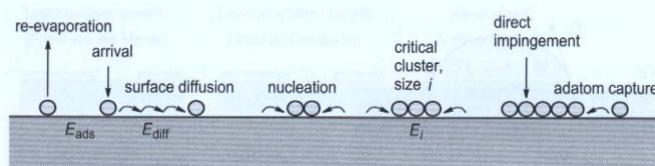
Growth Modes

- Exp: Monitor Auger signals from film and substrate while depositing...



Island Number Density

- Nucleation and growth on surfaces:



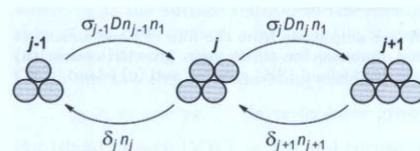
- Diffusion coefficient:
$$D = \frac{v}{4n_0} \exp\left(-\frac{E_{diff}}{k_B T}\right)$$

- Residence time:
$$\tau_{ads} = \frac{1}{v} \exp\left(\frac{E_{ads}}{k_B T}\right)$$

- Critical island size i : minimal size when the addition of one atom makes the island stable

Island Number Density

- Capture and decay processes → cluster size



- Rate equations:

$$\frac{dn_1}{dt} = R - \frac{n_1}{\tau_{ads}} + \left(2\delta_2 n_2 + \sum_{j=3}^i \delta_j n_j - 2\sigma_1 D n_1^2 - n_1 \sum_{j=2}^i \sigma_j D n_j \right) - n_1 \sigma_x D n_x$$

stable clusters

adatom density

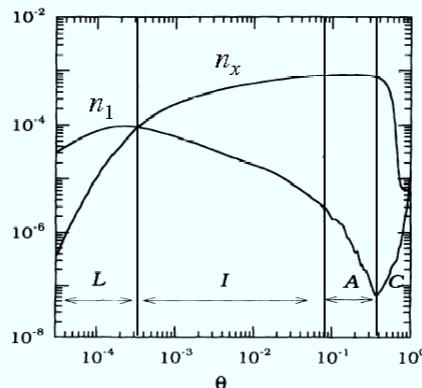
deposition rate

$$\frac{dn_j}{dt} = n_1 \sigma_{j-1} D n_{j-1} - \delta_j n_j + \delta_{j+1} n_{j+1} - n_1 \sigma_j D n_j \longrightarrow \text{metastable clusters}$$

$$\frac{dn_x}{dt} = n_1 \sigma_i D n_i \longrightarrow \text{stable clusters}$$

Island Number Density

- Numerical solution for $i = 1$ without re-evaporation (Amar, Family and Lam, PRB 1994):



- Four coverage regimes are found

Island Number Density

(1) Low-coverage nucleation regime: $n_1 \gg n_x$

- In such case:

$$n_1 \propto \Theta \quad n_x \propto \Theta^3$$

When $n_x \sim n_1 \rightarrow$ **(2) Intermediate-coverage regime**

- In such case:

$$n_1 \propto \Theta^{-1/3} \quad n_x \propto \Theta^{1/3}$$

When mean island separation \sim mean free path of adatoms

→(3) Aggregation regime ($\Theta \sim 0.1\text{-}0.4$ ML)

When the island join together

→(4) Coalescence and percolation regime

Island Number Density

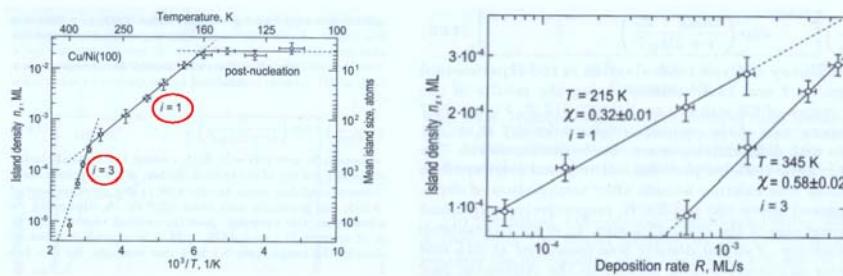
- Saturation density (Venables et al., 1984):

$$n_x = n_0 \eta(\Theta, i) \left(\frac{4R}{v_0 n_0} \right)^{\frac{i}{i+2}} \exp\left(\frac{iE_{diff} + E_i}{(i+2)k_B T} \right)$$

binding energy
of critical cluster

$\eta(\Theta, i) \sim 0.1 - 1$

- Example: Cu on Ni(100) (Müller et al., PRB 1996)



Island Shape

- At low T (slow edge diffusion): **ramified islands**
- **Diffusion-limited-aggregation (DLA) model** (Witten & Sander, PRL 1981):

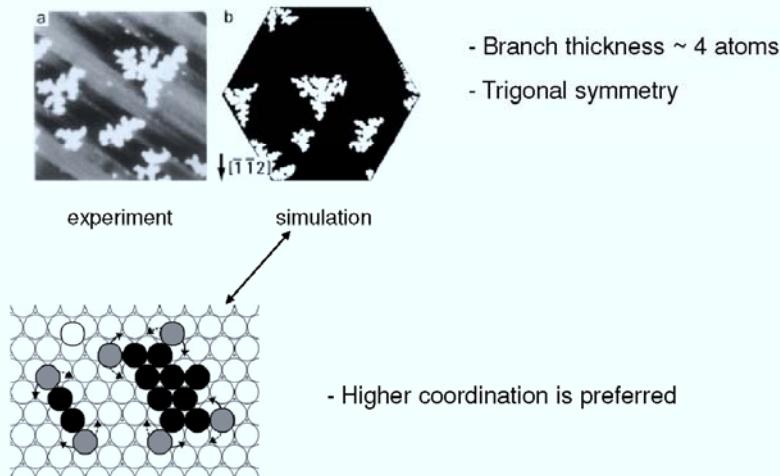


- Adatoms stick at islands
- Fractal shape
- Branch thickness ~ 1 atom
- No influence of lattice geometry

- In real growth (STM experiments):
- Fractal shape
- Branch thickness > 1 atom
- Influence of lattice geometry

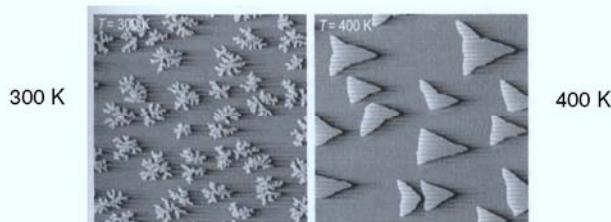
Island Shape

- Example: Pt/Pt(111) (Hohage et al., PRL 1996)

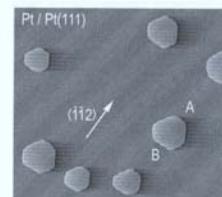


Island Shape

- At higher T: **compact islands**
- Example: Pt/Pt(111) (Bott et al., PRL 1992)

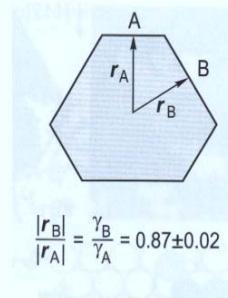


- The equilibrium shape is hexagonal:
(deposition at 425 K + annealing at 700 K)



Island Shape

- **2D Wulff theorem:** In a 2D crystal at equilibrium, the distances of the borders from the crystal center are proportional to their free energy per unit length



- For Pt(111): B/A ~ 0.87

Island Size Distribution

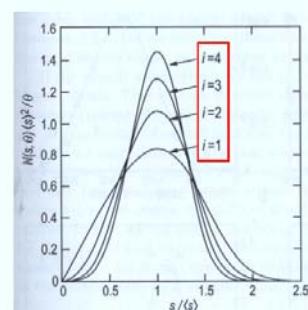
- **Island size distribution** depends on:

- Critical island size
- Coverage
- Substrate structure
- “Coarsening” (at high Θ)

- From **scaling theory** (Amar et al., PRB 1994):

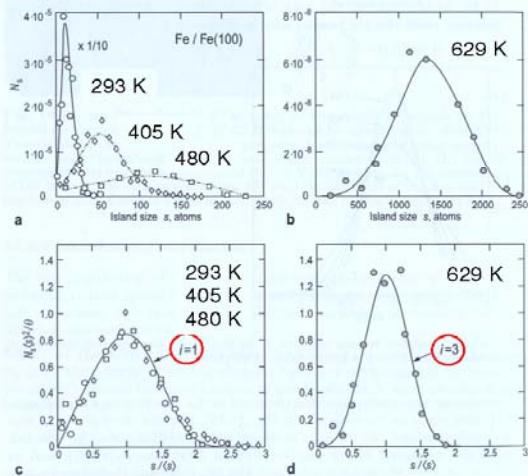
$$N_s = \frac{\Theta}{\langle s \rangle^2} f_i \left(\frac{s}{\langle s \rangle} \right)$$

- Comparing with experimental results
→ critical island size



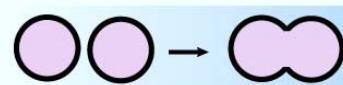
Island Size Distribution

- Example: Fe on Fe(100) (Stroscio et al., PRL 1993, Amar et al., PRB 1994)



Coarsening Phenomena

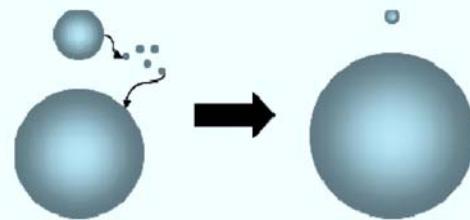
1) Coalescence:



- At 0.1 ML coverage: **dynamic coalescence (Smoluchowski ripening)**
- At 0.4-0.5 ML: **static coalescence**
- Higher coverages → **percolation growth** (→ change of physical properties!)

Coarsening Phenomena

2) (Ostwald) Ripening:



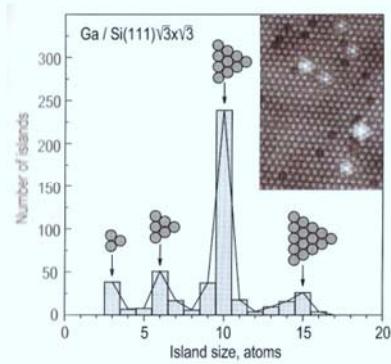
- Chemical potential of a circular island:

$$\mu(r) \propto \frac{\gamma}{r}$$

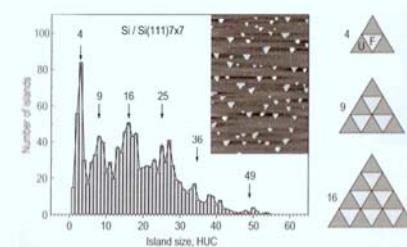
Reducing free energy \rightarrow net flow from smaller to larger islands!

Magic Islands

- Ga on Si(111) $\sqrt{3}\times\sqrt{3}$ (Lai & Wang, PRL 1998):



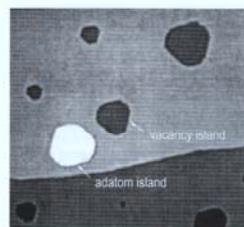
- Si on Si(111)7x7 (Voigtländer et al., PRL 1999):



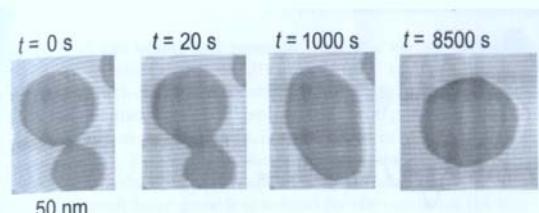
Formation of a new row \rightarrow high energy cost!

Vacancy Islands

- Ion bombardment → formation of **vacancy islands**

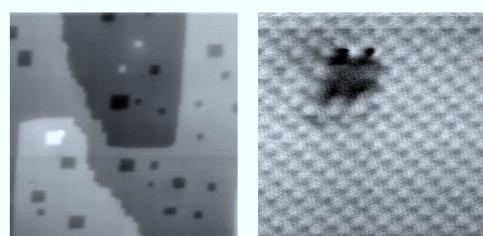


- Analogies with adatom islands



Vacancy Islands

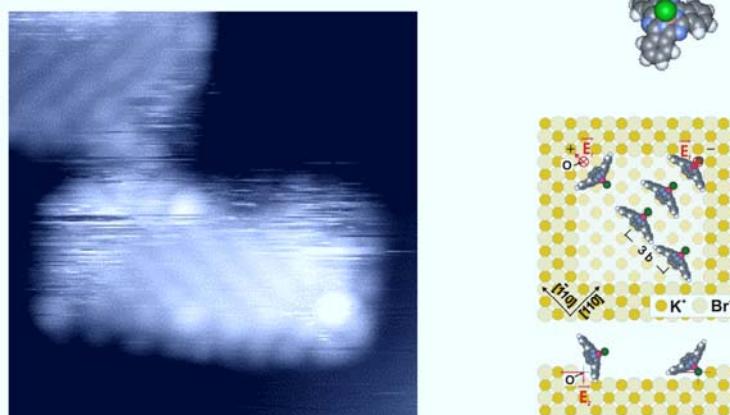
- Electron bombardment on insulating surfaces (Bennewitz et al., SS 2001):



- Irradiation with 1 keV electrons at 130 °C
- Rectangular pits with area of 1x1 nm² up to 30x30 nm²
- 1 ML deep (0.33 nm)

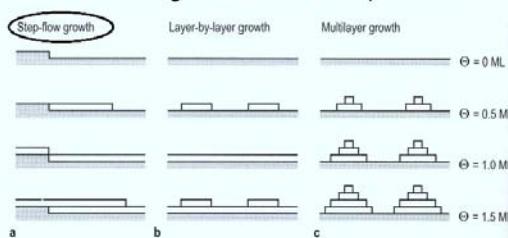
Vacancy Islands

- Vacancy islands can be used as molecular traps (Nony et al., NL 2004)

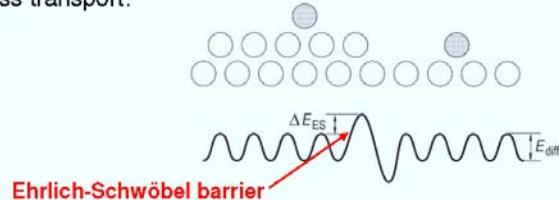


Kinetic Effects in Homoepitaxy

- Thermodynamics → Layer-by-layer growth but...
- Kinetic processes → Other growth modes are possible!

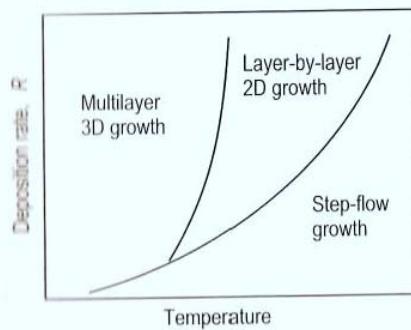


- Interlayer mass transport:



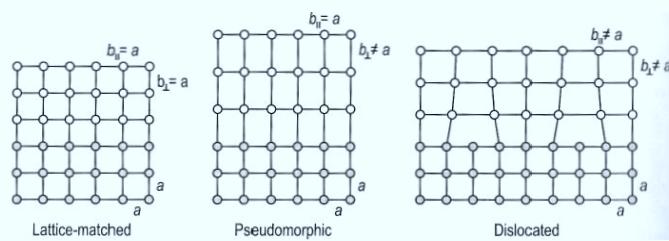
Kinetic Effects in Homoepitaxy

- Deposition rate and Temperature are important!
- Growth mode diagram (Rosenfeld et al., 1997):



Strain Effects in Heteroepitaxy

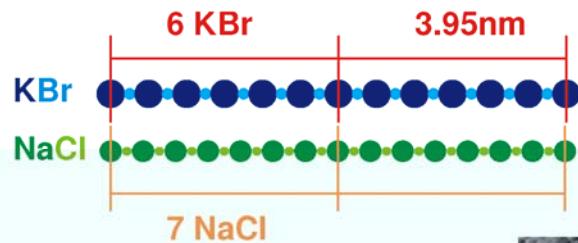
- Heteroepitaxy growth modes:



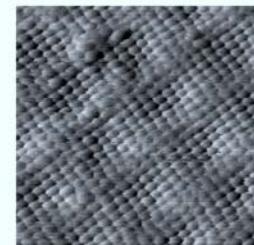
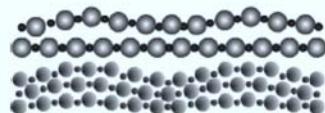
- Lattice **misfit** → elastic strain and dislocations
- Pseudomorphic growth below critical misfit and film thickness

Strain Effects in Heteroepitaxy

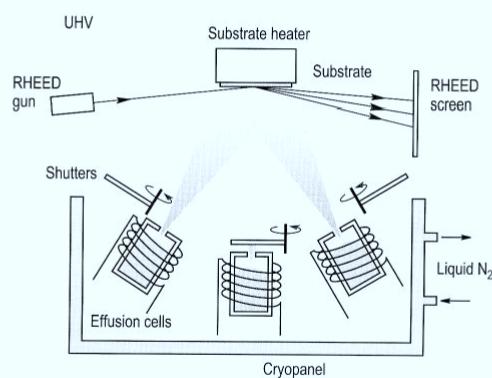
- Heteropitaxy on insulating surfaces (Maier et al., PRB 2007):



- A “Moiré pattern” appears (also in the substrate?)



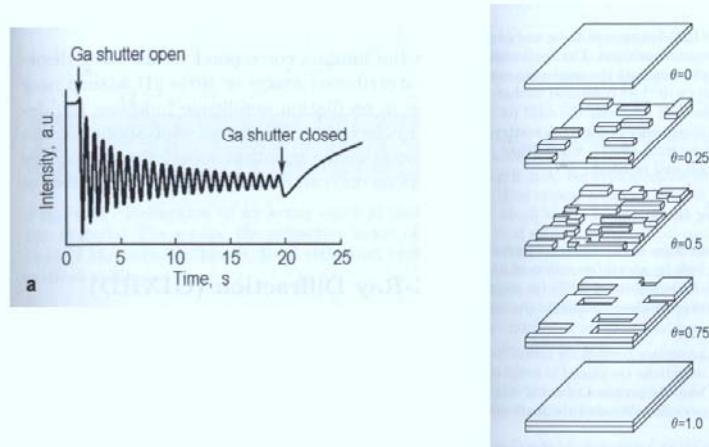
Molecular Beam Epitaxy



- Used both in research and in semiconductor device fabrications

Molecular Beam Epitaxy

- The growth process can be monitored by RHEED:



Solid Phase Epitaxy

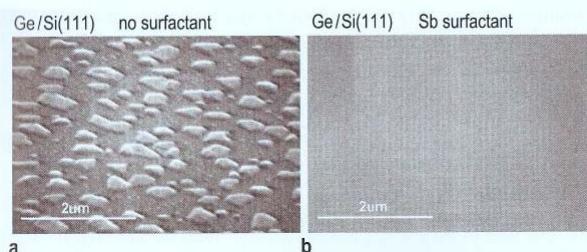
- Amorphous film deposited at low T and then crystallized upon heating at high T
- Lower crystallinity than MBE
- Used in semiconductor industry

Chemical Beam Epitaxy

- Growth by surface chemical reactions
- High temperature required
- High grow rate and crystallinity

Surfactant-Mediated Growth

- Impurity (**surfactant**) → different growth mode!
- Surfactant can be either segregated or trapped
- Example: Ge on Si(111) (Zahl et al., APA 1999)
 - Bare surface → Stranski-Krastanov mode
 - With surfactant (Sb) → Layer-by-layer mode



Further Reading

- K. Oura et al., Surface Science, Springer 2003, chapter 14
- J.A. Venables et al., Rep. Prog. Phys. 47 (1984) 399
- H. Brune, Surf. Sci. Rep. 31 (1998) 121

Oberflächenphysik



27/04/2010

Local Probes and Experiments I: Scanning Tunneling Microscopy (STM) Inelastic Tunneling and Scanning Tunneling Spectroscopy (STS)

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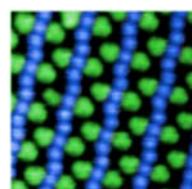


STM – local probe for surface science



surface analysis @ nanoscale

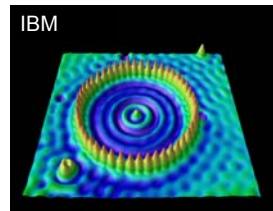
- scanning probe methods (STM/AFM)
 - working principle of STM
 - tunnel current
 - examples and image interpretation
- scanning tunneling spectroscopy (STS)
 - local electronic structure



STM image: self-assembled
molecular layer
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surface modifications @ nanoscale

- manipulation of atoms or adsorbates
- nanoindentation



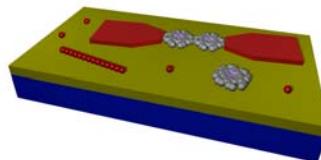
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Nanotools for surface analysis and modifications

Motivation

- Miniaturization / Technology
- Study of material properties at small dimensions
 - => **research on ultrathin insulator layers & organic molecules (building blocks for devices)**
- applications
 - OLED displays (Organic Light Emitting Diode)
 - future molecular electronics
 - catalysts
 - ...
- fundamental interest
 - electronic structure
 - molecular interactions
 - chemical reactivities
 - ...

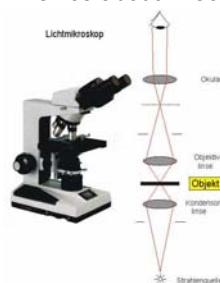


Microscopes

For the visualisation of millimeter to nanometer structures

Light Microscope

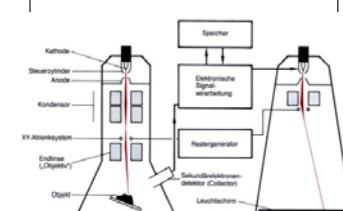
since about 1750



geometric optics
resolution about 500 nm
Light-Intensity contrast

Electron Microscope (SEM)

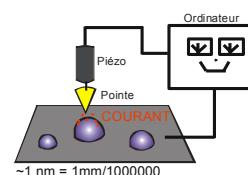
since about 1955



e-beam raster-scan
resolution 5 nm
secondary electron counting
projection image

Scanning Probe Microscope (SPM)

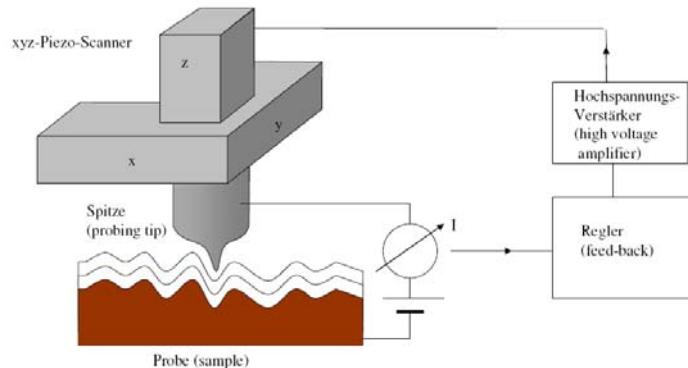
since about 1981



Local probe piezo-scan
resolution 0.1 nm
3D map of surface
~1 nm = 1mm/1000000



Rastertunnelmikroskop (STM)



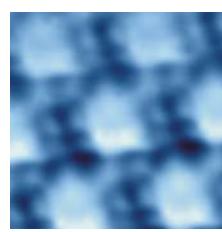
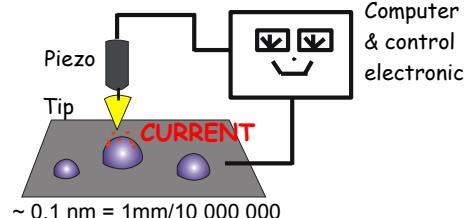
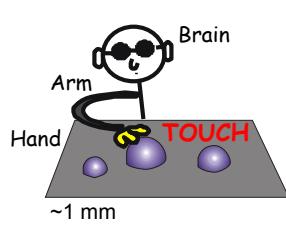
Ein Regler hält den Tunnelstrom (\sim pA-nA) zwischen Spitze und Probe konstant. Es werden Konturen konstanter Tunnelstroms abgerastert.



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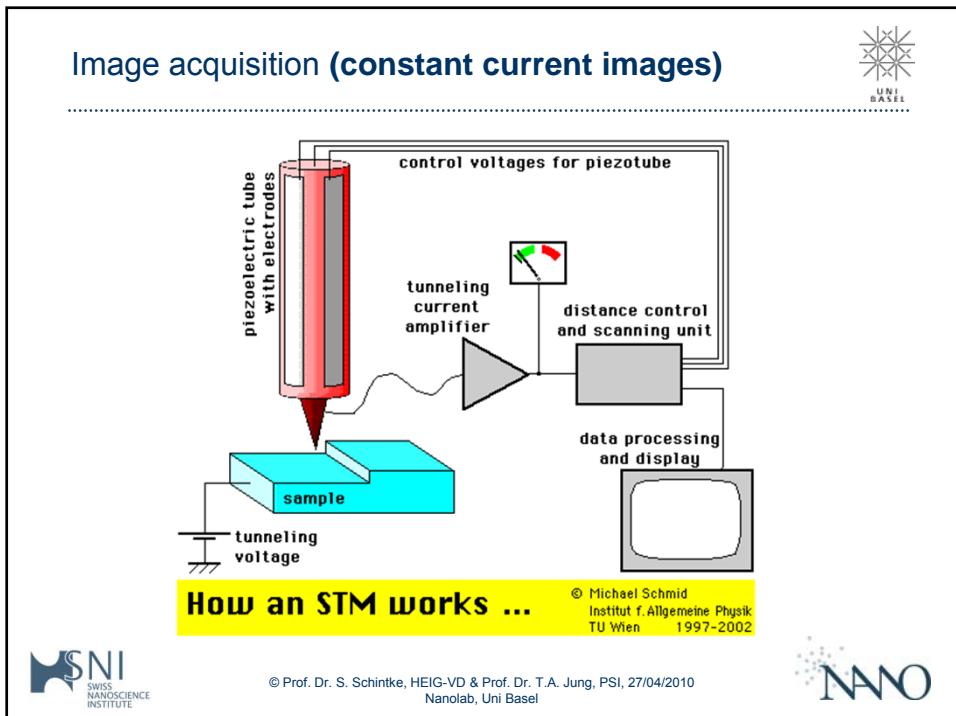
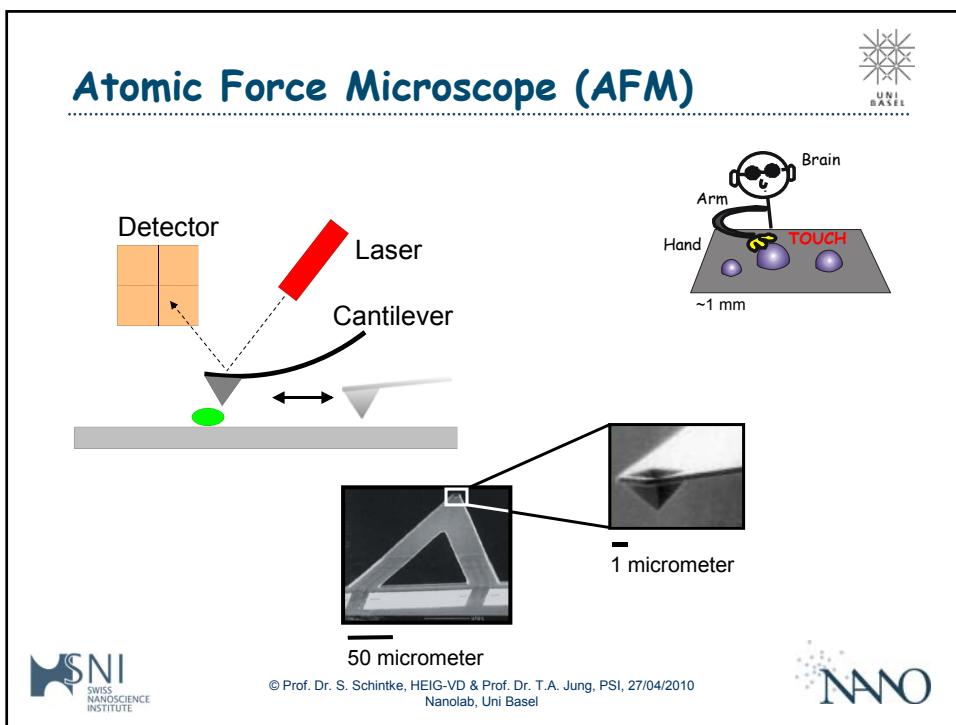


Scanning Tunneling Microscopy

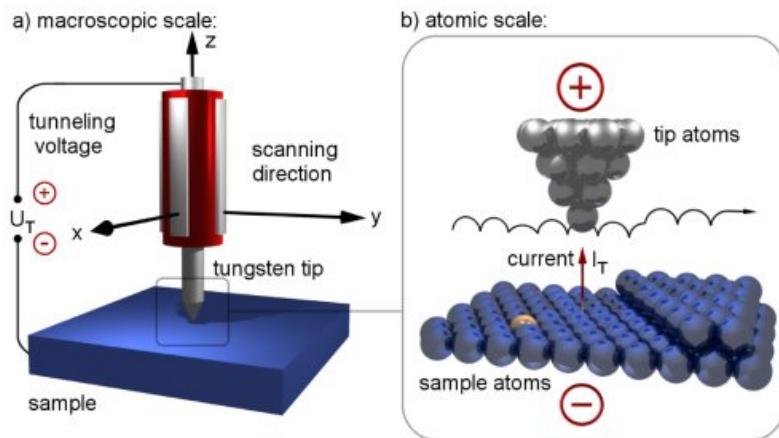


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Piezo unit and tip displacement



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History of Scanning Tunneling Microscopy

- **1979-1981 Development at IBM Zürich**
G. Binnig, H. Rohrer, Ch. Gerber, E. Weibel,
Appl. Phys. Lett. 40 (1982), *Phys. Rev. Lett.* 49, 57 (1982)
- **1986 Nobel prize of Physics** for Gerd Binnig and Heinrich Rohrer



The soccer ball team of the IBM research lab; 15 October 1986
C. Julian Chen: *Introduction to Scanning Tunneling Microscopy*;
Original photo: Blick



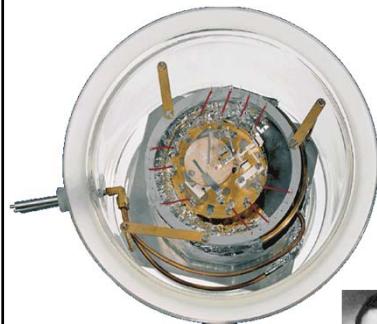
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The first Scanning Tunneling Microscope



1981 development at
IBM Rüschlikon, Switzerland



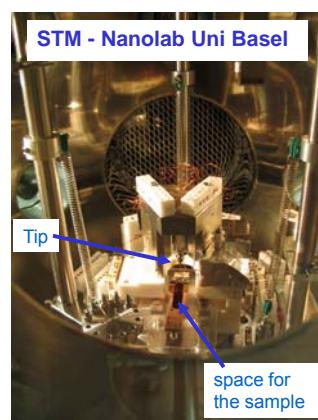
G. Binnig and H. Rohrer
Nobelprize for physics 1986



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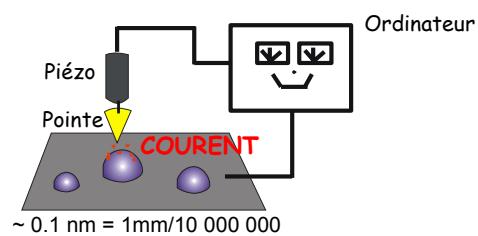
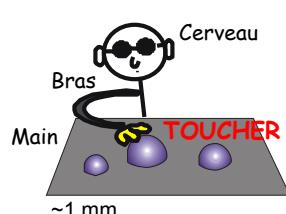
The « Nanolab » at the University of Basel



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Les composants principaux d'un STM



Composants principaux:

Main: sonde locale => pointe métallique, fine

Bras: balayage => actuateur piézoelectrique

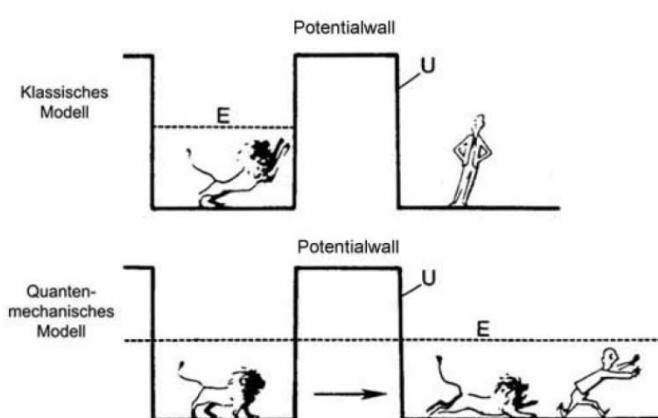
Cerveau: régulation & enrégistrement => électronique de régulation & ordinateur



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Quantum Mechanic Tunneling



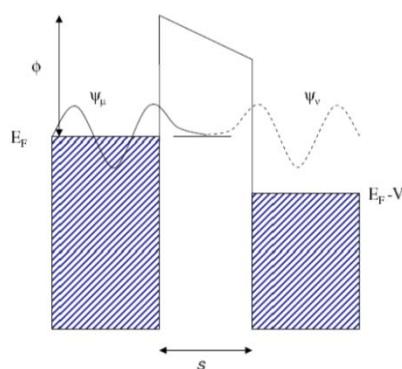
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Tunneleffekt

Schon zu Beginn der Quantenmechanik wurde der Tunneleffekt vorausgesagt. Der Überlapp der Wellenfunktionen führt zu einer Transmission von Elektronen durch ein klassisch verbotenes Gebiet. Zwischen zwei Metallen, die durch Vakuum oder ein Oxid getrennt sind, fließt ein Tunnelstrom.



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Tunneleffekt



$$I = f(U) \exp(-A\sqrt{\phi} s)$$

J. Frenkel, *Phys. Rev. B* **36**, 1604 (1930)

I: Tunnelstrom

U: Extern angelegte Spannung

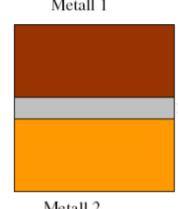
s: Distanz zwischen Probe und Spitze

ϕ : Barriereförde

$$\phi \approx \frac{\phi_1 + \phi_2}{2} \quad \phi_1, \phi_2 \text{ Austritsarbeiten von Metall 1 und Metall 2}$$

$$A = 2\sqrt{\frac{2m}{\hbar^2}} = 1.025 \text{ Å}^{-1}\text{eV}^{-1/2}$$

f(*U*): Funktion der elektronischen Struktur von Probe und Spitze
Für freie Elektronen $f(U) \sim U$



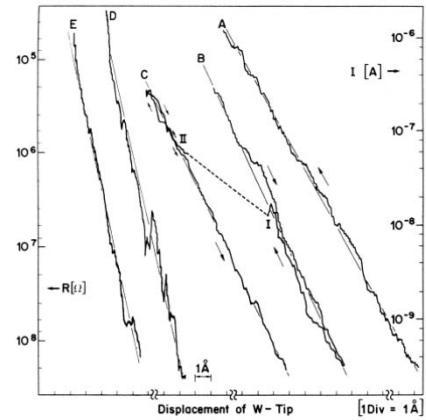
Der Tunnelstrom hängt exponentiell vom Abstand *s* ab. Für typische Austritsarbeiten von $\phi=4.5\text{eV}$ ändert sich der Strom etwa um eine Größenordnung, wenn die Distanz um 1\AA verändert wird. Historisch wurde zuerst das Oxit tunneln realisiert. Erst mit dem STM konnte Vakuumtunneln beobachtet werden.



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Tunnelstrom



Exponentieller Abfall des Tunnelstromes mit dem Abstand



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STM



Nanosurf AG, Liestal



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Tunnel Current - simple approximation

Sample Tip

Tunnel Current

Energy

E_{vac}

Φ

e^-

E_{vac}

E_F

d

ρ Sample

ρ Tip

$$I_t \approx c_1 \cdot U_{bias} \cdot e^{-c_2 \sqrt{\Phi} \cdot d}$$

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Tip

space for the sample

NANO

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Measurement modes: constant current vs constant height

z

e^-

at each position (x,y)
record signal:
tip displacement in z direction

e^-

at each position (x,y)
record signal:
tunnel current I_t

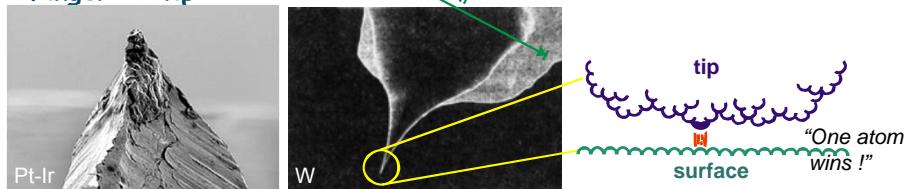
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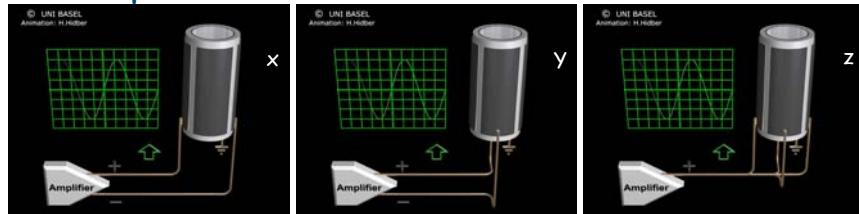
The « finger » and the « arm »



“Finger” = tip



“Arm” = piezoelectric actuator



Animations: H. Hidber, University of Basel



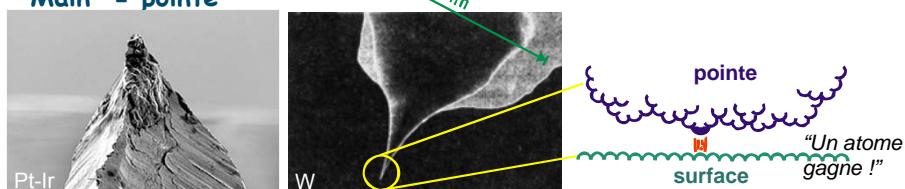
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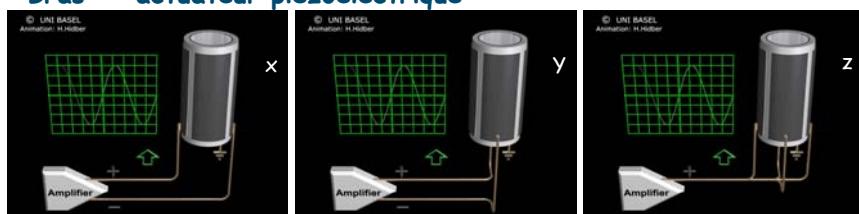
La « main » et le « bras »



“Main” = pointe



“Bras” = actuateur piézoélectrique



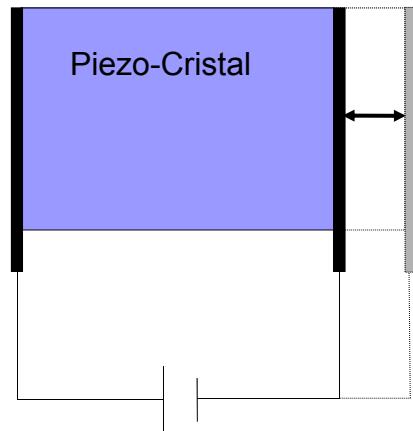
Animations: H. Hidber, Université de Bâle



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The Piezo Scanner



E-field modifies
X-tal structure
and elongation

DC voltage



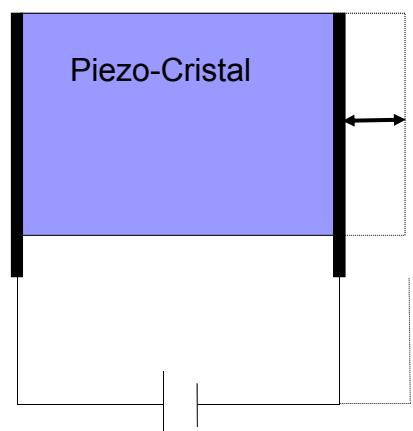
-220 to +220 V

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The Piezo Scanner



contracted !

DC voltage



-220 to +220 V

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The Piezo Scanner



Piezo-Cristal

extended !

DC voltage



+220 to +220 Volt

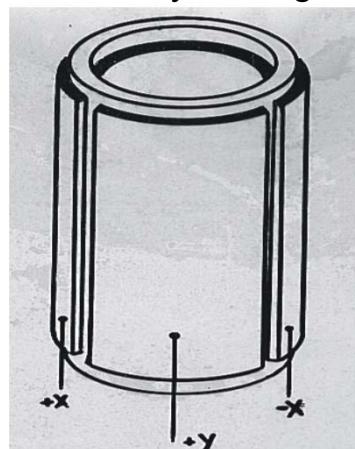
G-VD & Prof. Dr. T.A. Jung, PSI, 27/04/2010
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XYZ – scan unit



'precision at your fingertips'

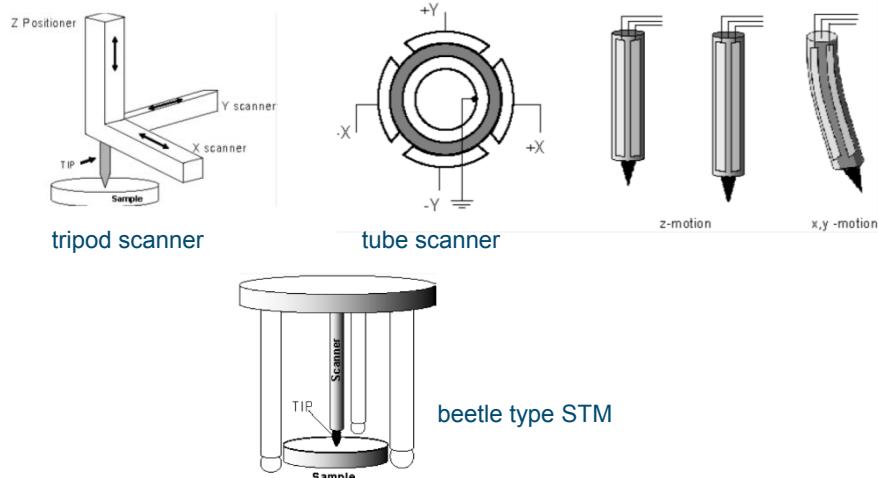


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Precision: 1 pm

Piezo scan unit



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Principe d'un microscope à effet tunnel



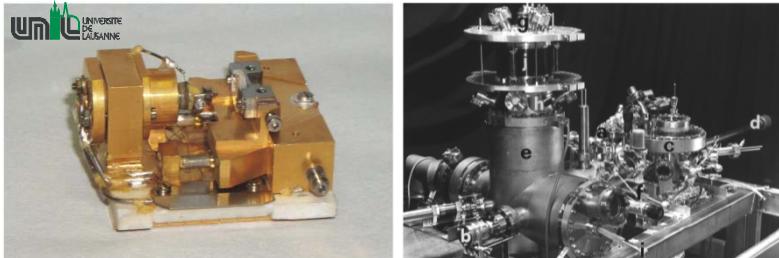
Animation: IBM



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Advantages of UHV and low temperature



Reinheit der Probenoberflächen
Präparation der Kristalloberfläche und Messung im Ultrahochvakuum (UHV) ($p < 10^{-10}$ mbar)

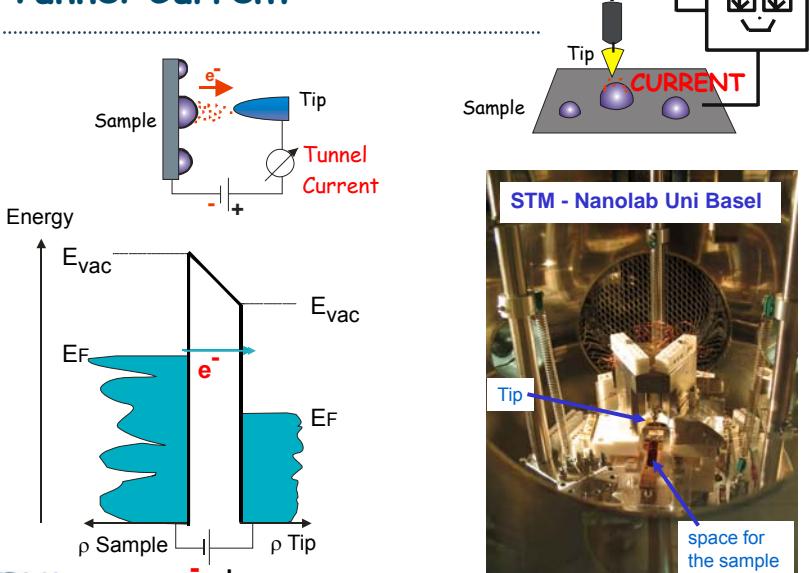
Schwingungsdämpfung
- STM auf pneumatisch gelagertem massiven Rahmen
- STM Mess-Station mit Faltenbalg schwingungsisoliert
- Schallschutzkammer um Gesamtanlage während der Messungen

Stabilität atomarer Strukturen
Messung bei Tieftemperatur
=> reduzierte Beweglichkeit von Atomen und Molekülen auf der Oberfläche
 $T=50\text{K}$ (fester Stickstoff)
 $T=5\text{K}$ (flüssiges Helium)

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Tunnel Current



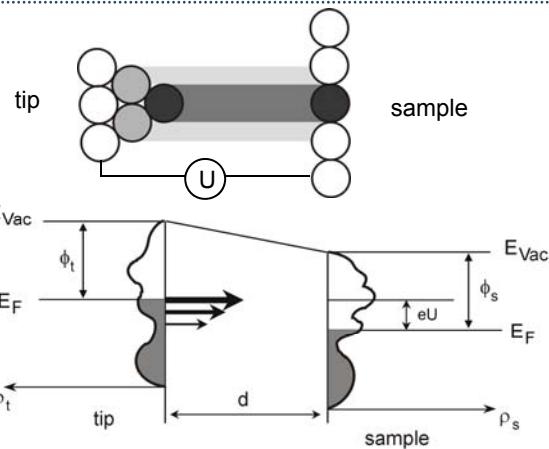
STM - Nanolab Uni Basel

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Tunnel Current in STM



Tunneling current: $I_{\text{tunnel}} \sim U \rho_t \rho_s(x,y) e^{-\text{const } d}$ (Tersoff and Hamann)

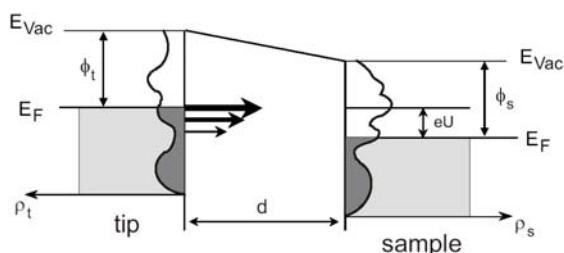
=> sensitivity to local electronic structure of the sample



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Scanning Tunneling Current



Bardeen approximation PRL 6, 57 (1961)

$$I \propto \int_0^{eU} \rho_s(E) \rho_t(\pm eU \mp E) T(E, eU) dE$$

$$T(E, eU) = \exp \left(-\frac{2z\sqrt{2m}}{\hbar} \sqrt{\frac{\Phi_s + \Phi_t}{2} + \frac{eU}{2} - E} \right)$$



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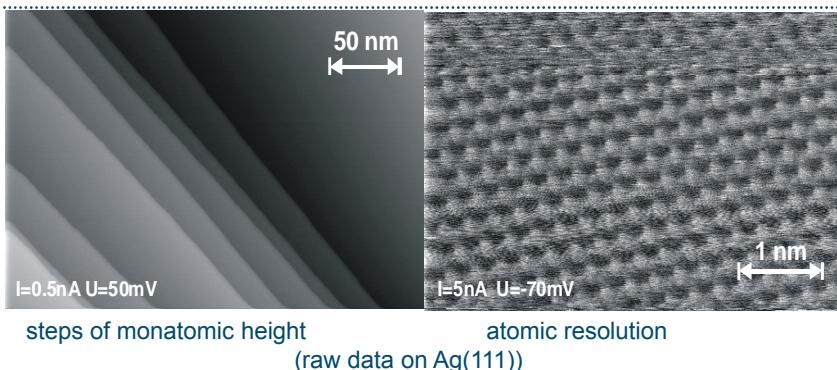
STM Application for Surface Analysis and Surface Material Science & Engineering



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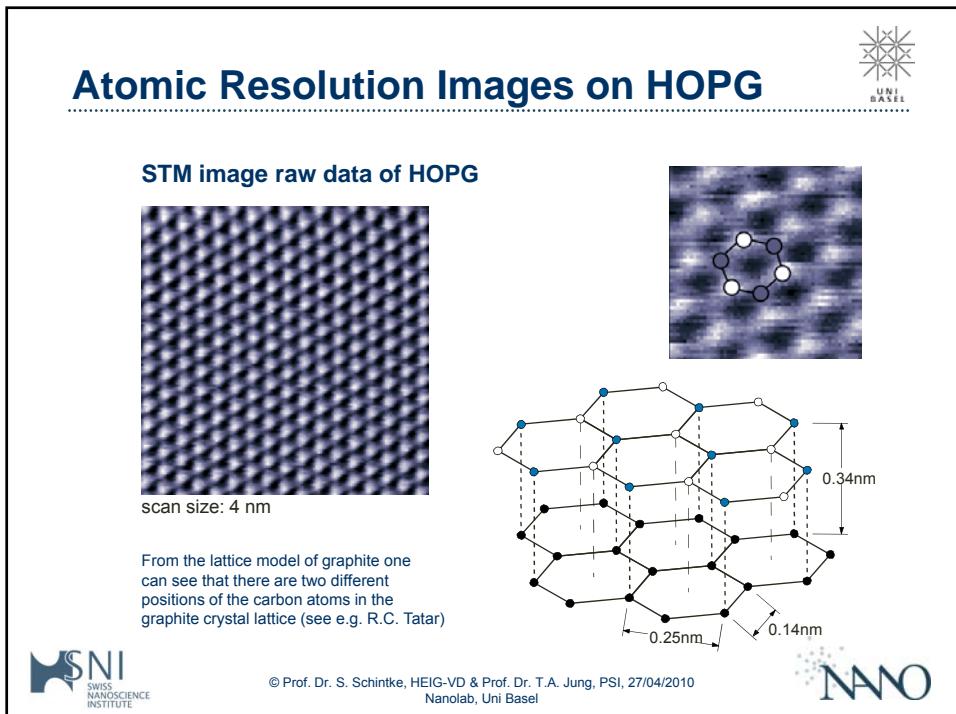
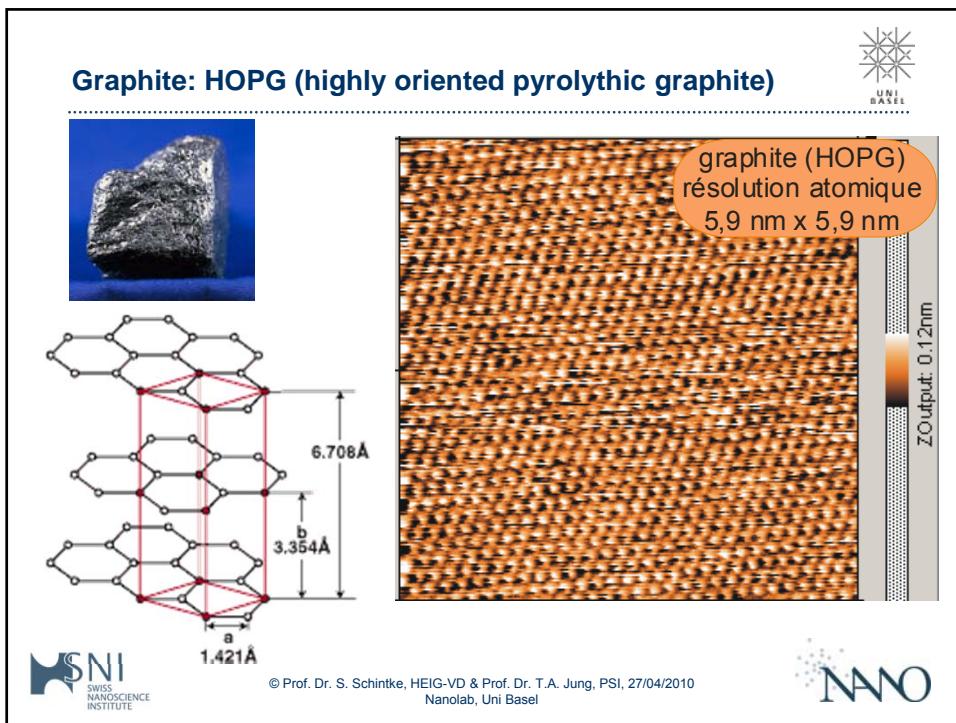


Atomic resolution Metals

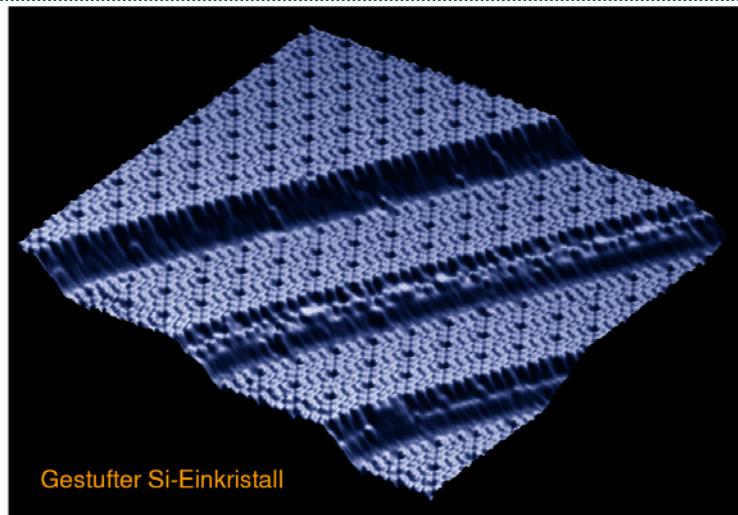


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Semiconductors: Silicon



Gestufter Si-Einkristall

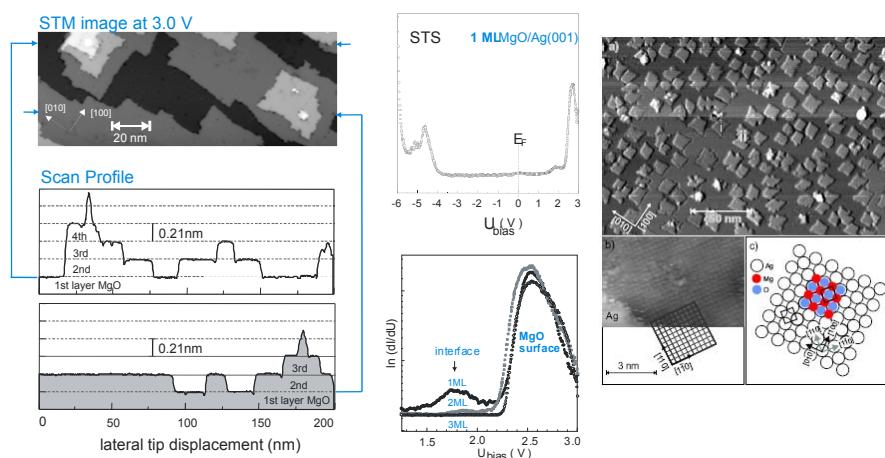


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Ultrathin insulators $\text{MgO}/\text{Ag}(001)$: Insulator at the limit

nature physics portal
research highlights
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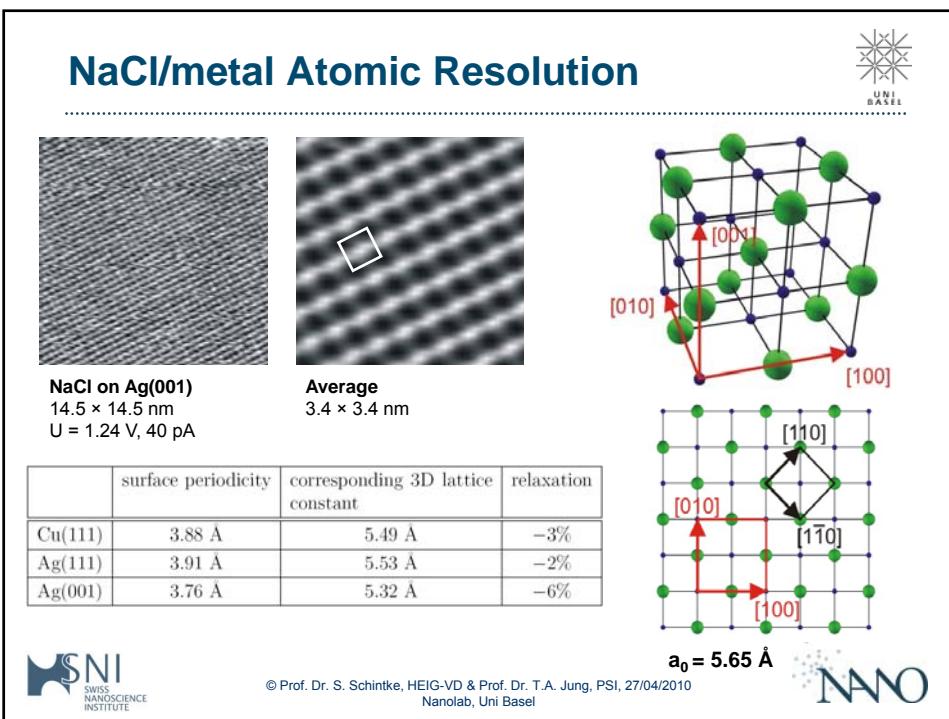
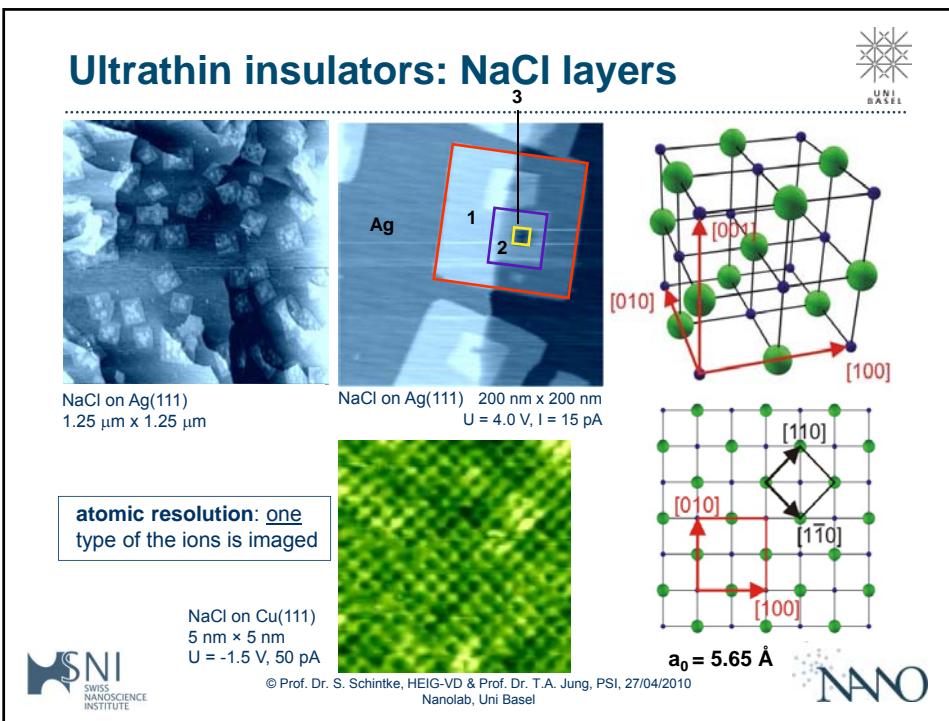
S. Schintke et al., *Insulator at the ultrathin limit: MgO on $\text{Ag}(001)$* , Phys. Rev. Lett. **87**, 276801 (2001)

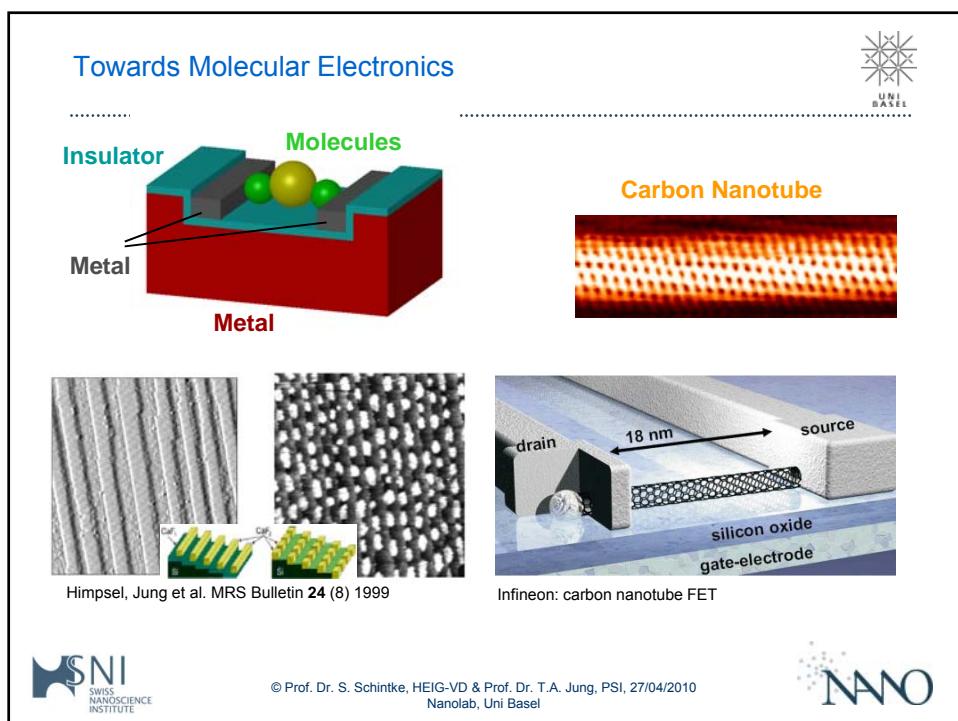
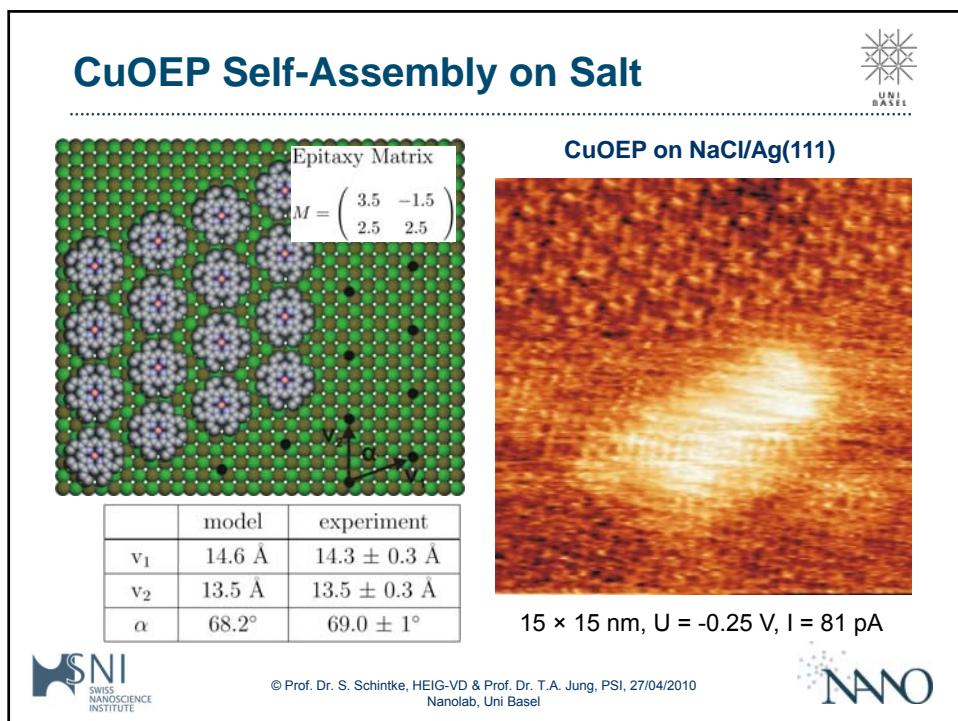
S. Schintke and W.-D. Schneider, *Insulators at the Ultrathin Limit: Electronic Structure studied by Scanning Tunneling Microscopy and Scanning Tunneling Spectroscopy*, J. Phys.: Condens. Matter **16**, R49-R81 (2004)



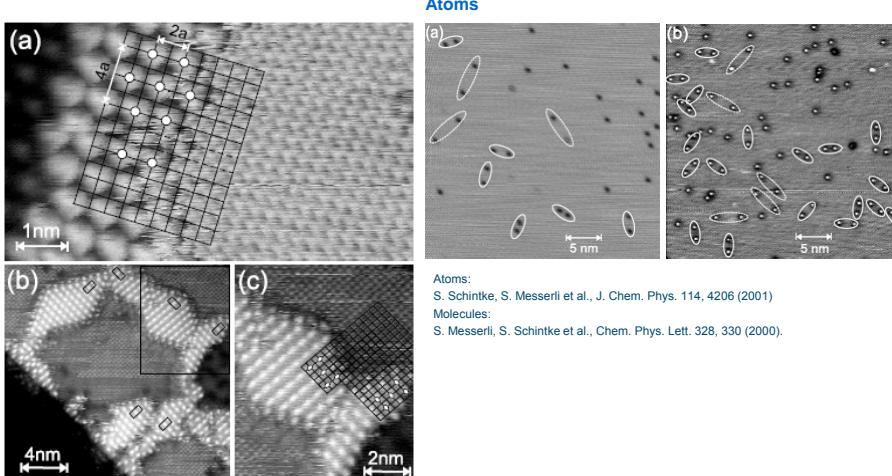
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Oxygen on Ag(001): molecular superstructures; far- ranged dissociation



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Oxygen/Ag(001)

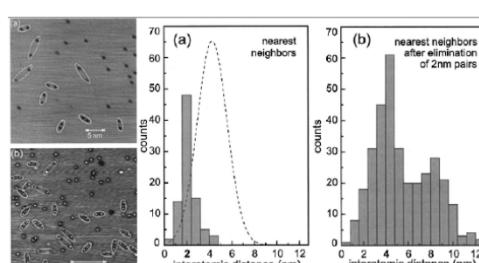


Fig. 3.1 Left: Pairing of oxygen atoms on Ag(001) at different coverages observed in STM images (50 K).
Right: Distribution of interatomic distances analysed from STM data.

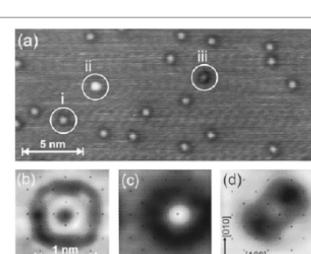


Fig. 3.2: Binding sites of oxygen atoms.
Top: experimental STM data (50 K). Bottom: Calculated STM images a) hollow site, b) on-top site, c) bridge site.

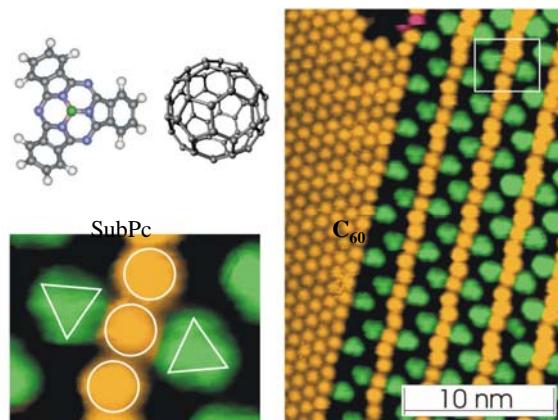
S. Schintke, S. Messerli, K. Morgenstern, J. Nieminen, and W.-D. Schneider, Journal of Chemical Physics 114, 4206-4209 (2001)



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Self-intermixed monolayer



Scan range 4.3nm x 3.2nm. V_{bias}=1.3V, I_t=20pA. Scan range 17nm x 25nm. V_{bias}=1.3V, I_t=20pA.

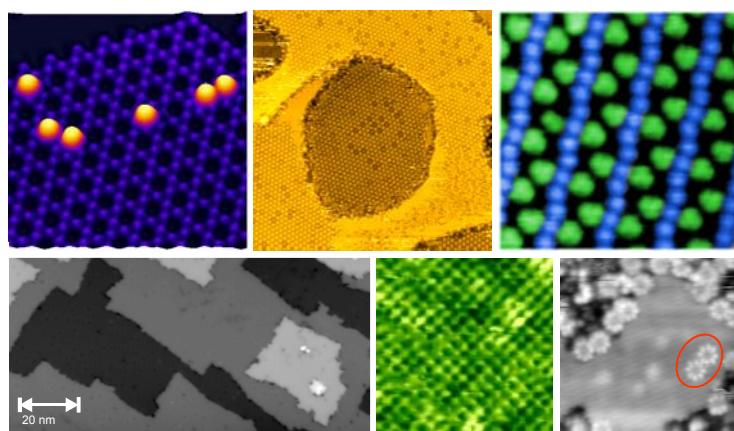
M. de Wild *et al.*, *ChemPhysChem* **10**, 181 (2002)
M. de Wild *et al.*, *Chimia* **10**, 56 (2002)



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Further STM images from research

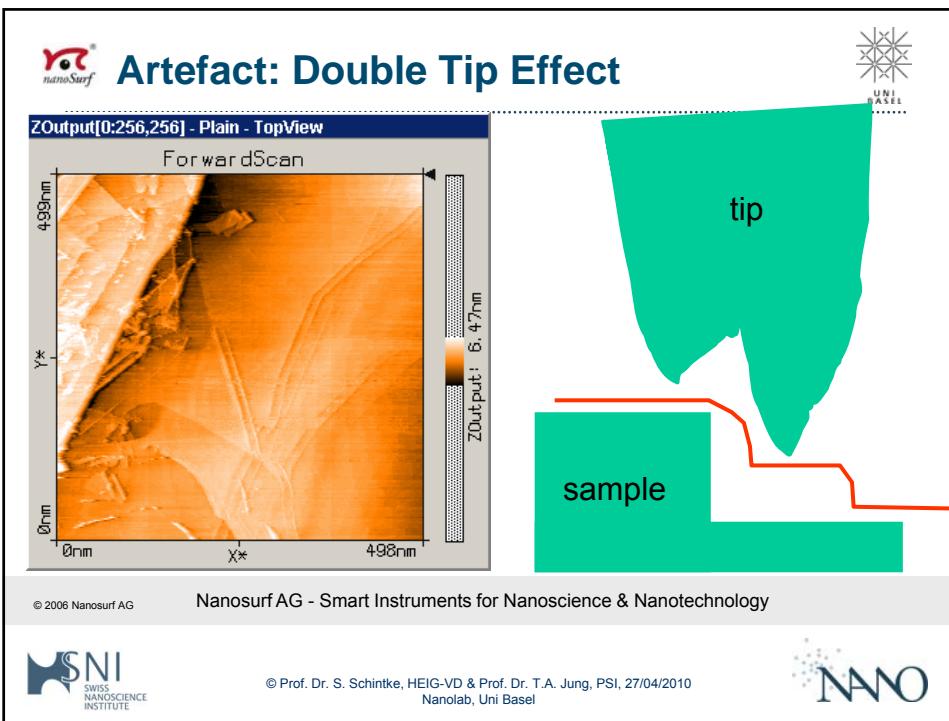


www.nccr-nano.ch (Uni Basel)



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2. STM beyond imaging

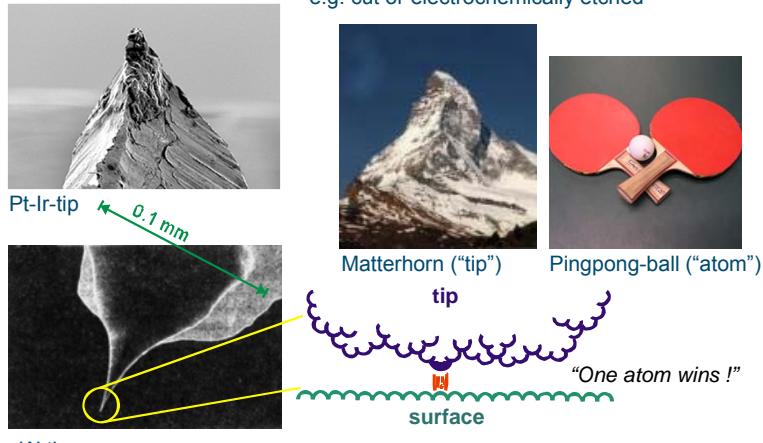
- STM a tool for nanofabrication:
Adsorbate manipulation
- STM a local spectroscopy tool:
Scanning Tunneling Spectroscopy (STS)
Inelastic Tunneling Spectroscopy (IETS)
Local Photoluminescence spectroscopy

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« Nanotool »: STM-Tip



Materials: Tungsten (W), Iridium (Ir), Platinum-Iridium (Pt-Ir), Gold (Au),...
e.g. cut or electrochemically etched

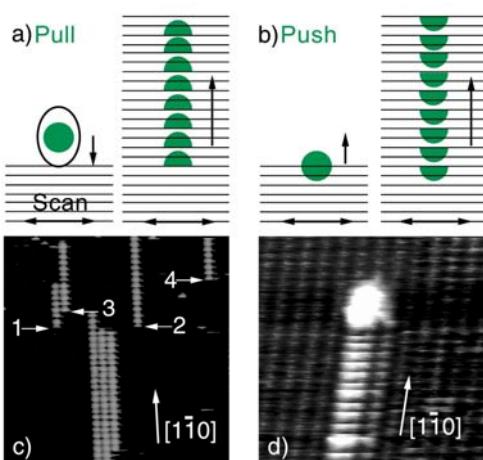


W-tip
Scanning electron micrograph of an STM tip
(Hamann, Hietschold, Rastertunnelmikroskopie)

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Tip-sample interaction



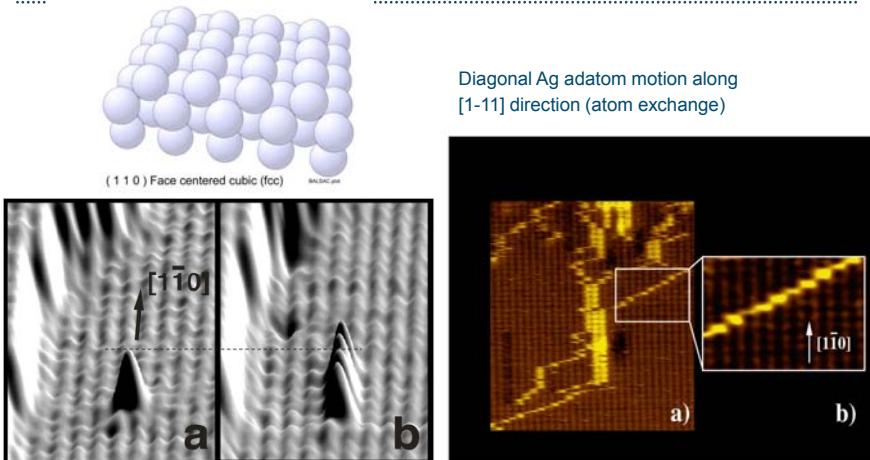
Low-temperature manipulation of Ag atoms and clusters on a Ag(110) surface,
J.T. Li, W.-D. Schneider, and R. Berndt, *Appl. Phys. A*, 66, 575 (1998).



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Tip-sample interaction



*Low-temperature manipulation of Ag atoms and clusters on a Ag(110) surface,
J.T. Li, W.-D.Schneider, and R. Berndt, Appl. Phys. A, 66, 575 (1998).*



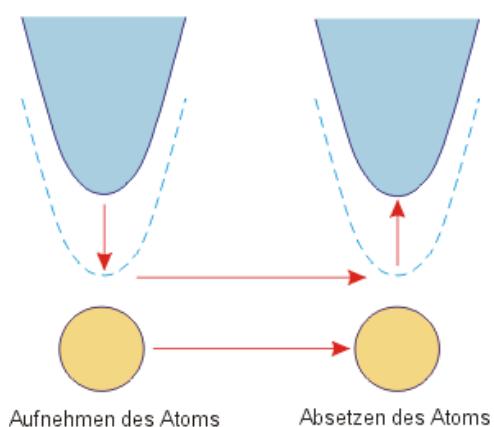
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Adsorbate manipulation by STM



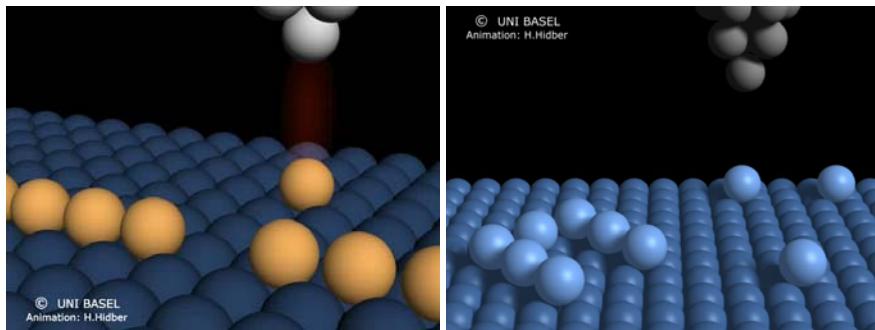
Bewegen eines Atoms



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Déplacer des atomes ou molécules



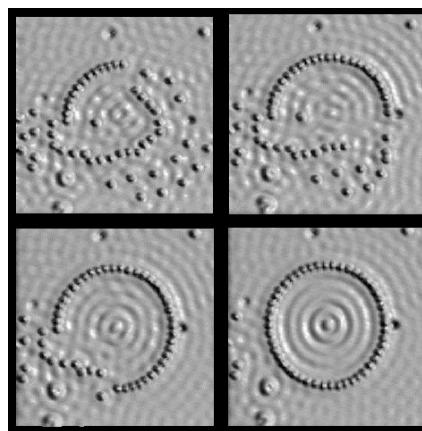
Animations: H. Hidber, Université de Bâle



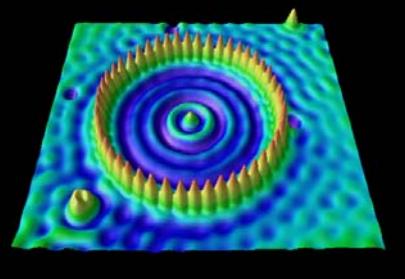
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Quantum Corral



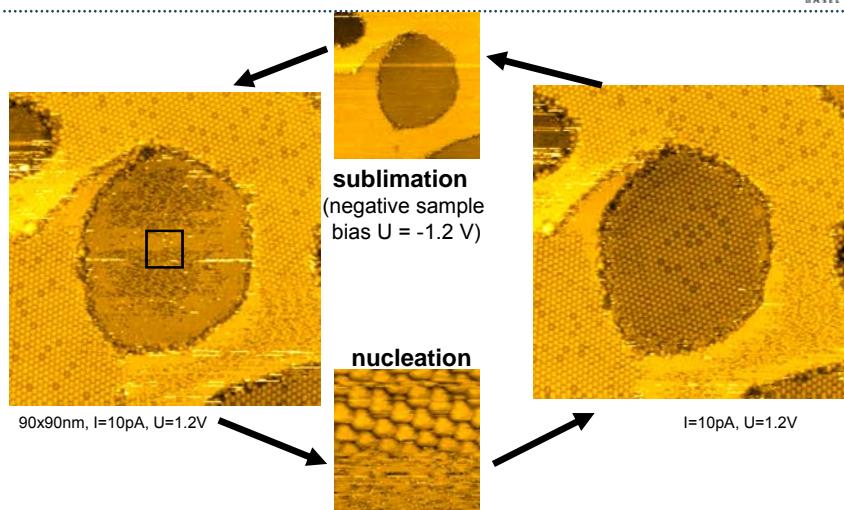
IBM



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Reversible 2D Phase Transition controlled by the STM tip



Controlled phase transition 2D fluid \leftrightarrow 2D solid



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