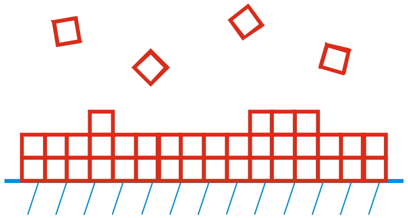
Self-organization on surfaces: an overview

O.Fruchart

Macroscopic concept: surface energies

2D

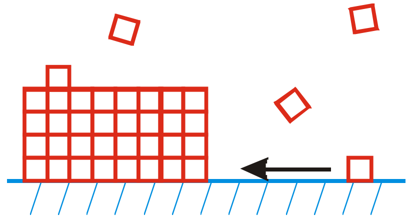
Franck van der Merwe



? substr. > ? film + ? interf.

3D

Volmer-Weber



? substr. < ? film + ? interf.

E.Bauer, Z.Kristallogr.110, 372 (1958)

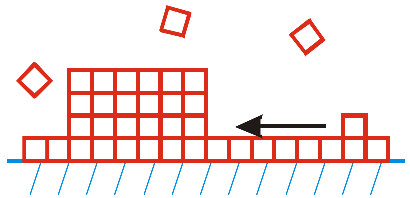
E.Bauer, Phys.Rev.B 33, 3657 (1986)

- Flat thin films: **high** substrate energy
- Islands (dots): **low** substrate energy

Another growth mode is frequently observed

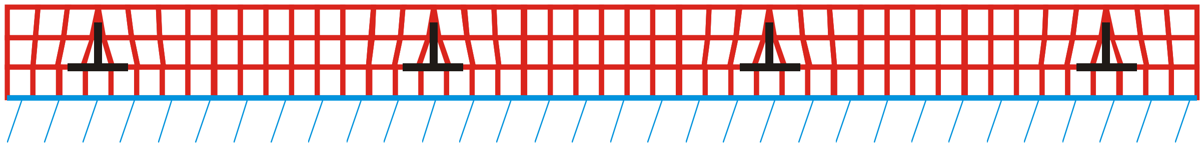
2D → 3D

Stranski-Krastanov

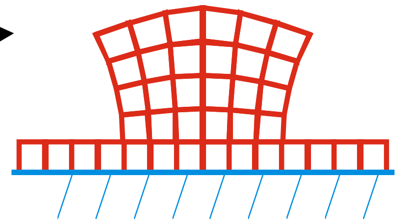


Microscopic concept: misfit accomodation

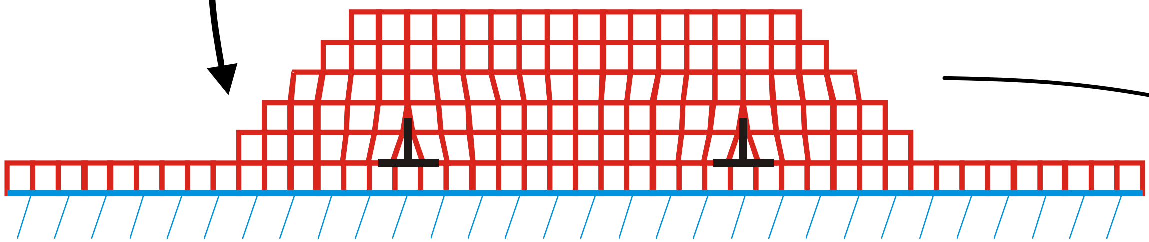
- High energy of dislocations



Misfit accomodation by elastic 3D relaxation

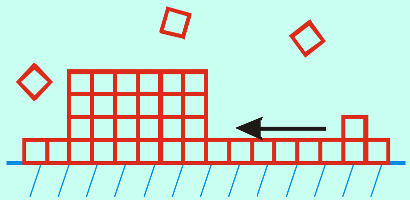


Additional effective interface energy



2D → 3D

Stranski-Krastanov



P.-O. Jubert *et al.*, Phys. Rev. B 64, 115419 (2001)

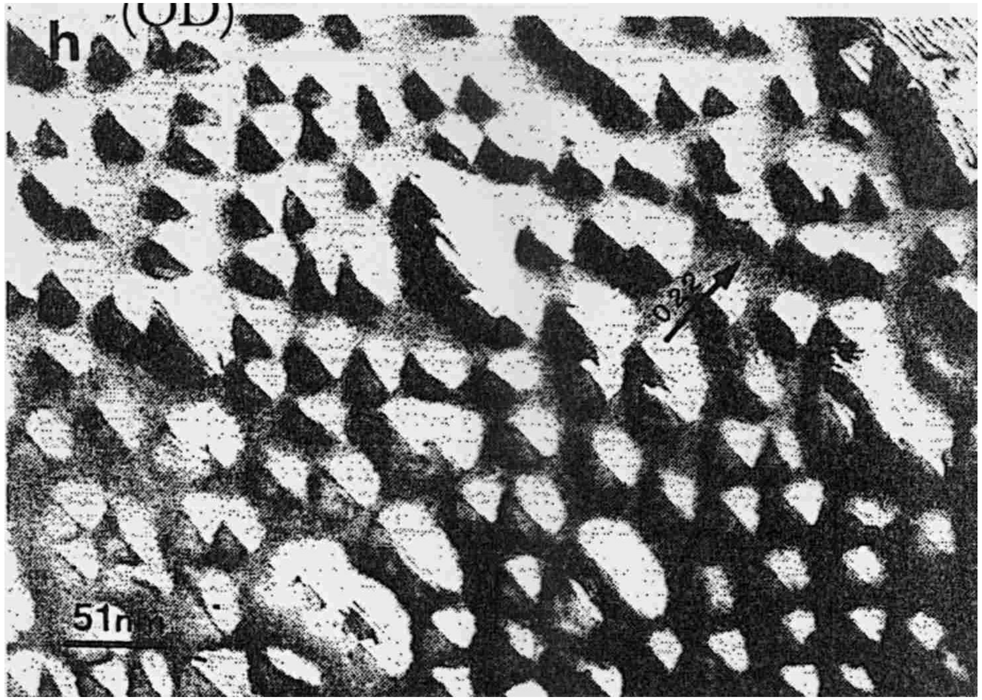
F. Tinjod *et al.*, submitted to J. Alloy Comp. (2002)

- Thickness-dependant surface stress

H.Ibach, Surf.Sci.Rep.29, 193 (1997)

> Many parameters for spontaneous island growth: Self-assembly

➢ $In_xGa_{1-x}As/GaAs$ quantum dots (OD)



S.Z.Chang *et al.*,J.Appl.Phys.73,4916(1993)

Laser diodes

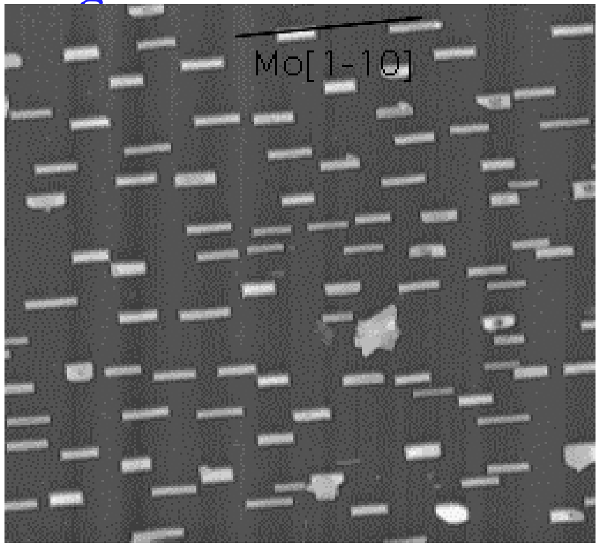
Improvements : $\frac{?\,L}{L} \approx 4\%$

Nishi et al., Appl.Phys.Lett.74, 1111(1999)

➢ Fe/Mo(110) elongated islands

6µm

SEM



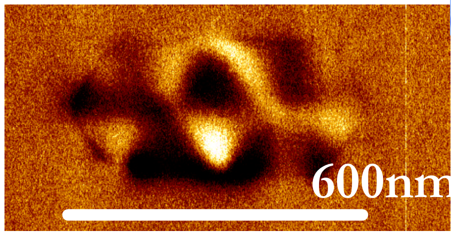
Target : $SmFe_2$

AFM

MFM.

Coll. Y. Samson

(CEA/France)



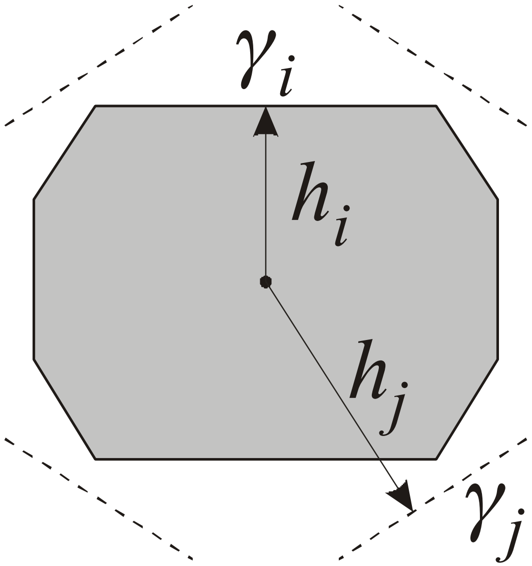
Target : Fe

P.-O.Jubert *et al.*, LLN.

Wulff's theorem

Free crystal

$$\frac{\gamma_i}{h_i} = \text{Constant}$$

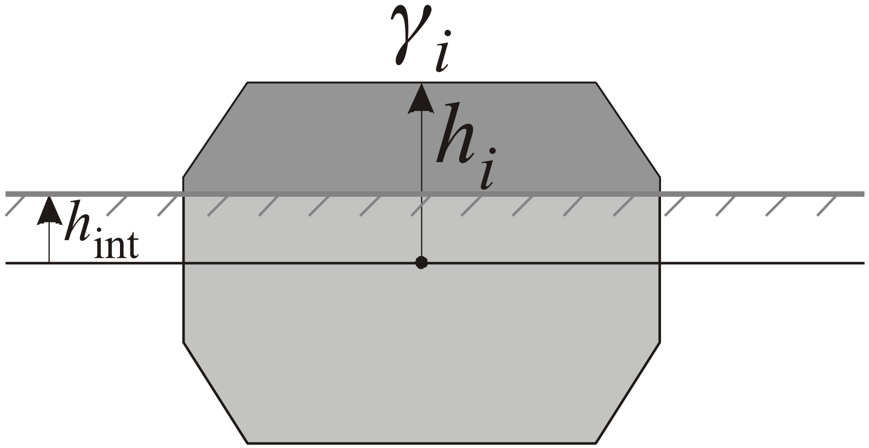


No facets with high surface energy

Wulff Kaishev's theorem

Supported crystal (growth on surfaces)

$$\frac{\gamma_i}{h_i} = \frac{\gamma_S - \gamma_{int}}{h_{int}} = \text{Constant}$$

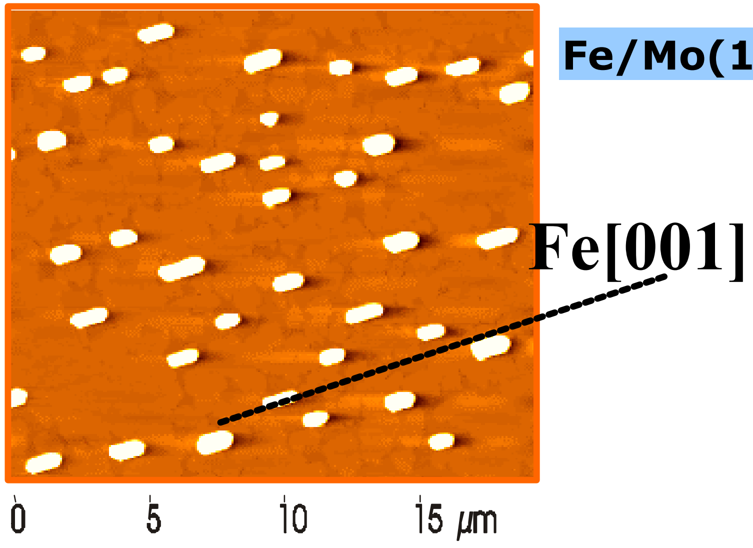


Truncated crystal

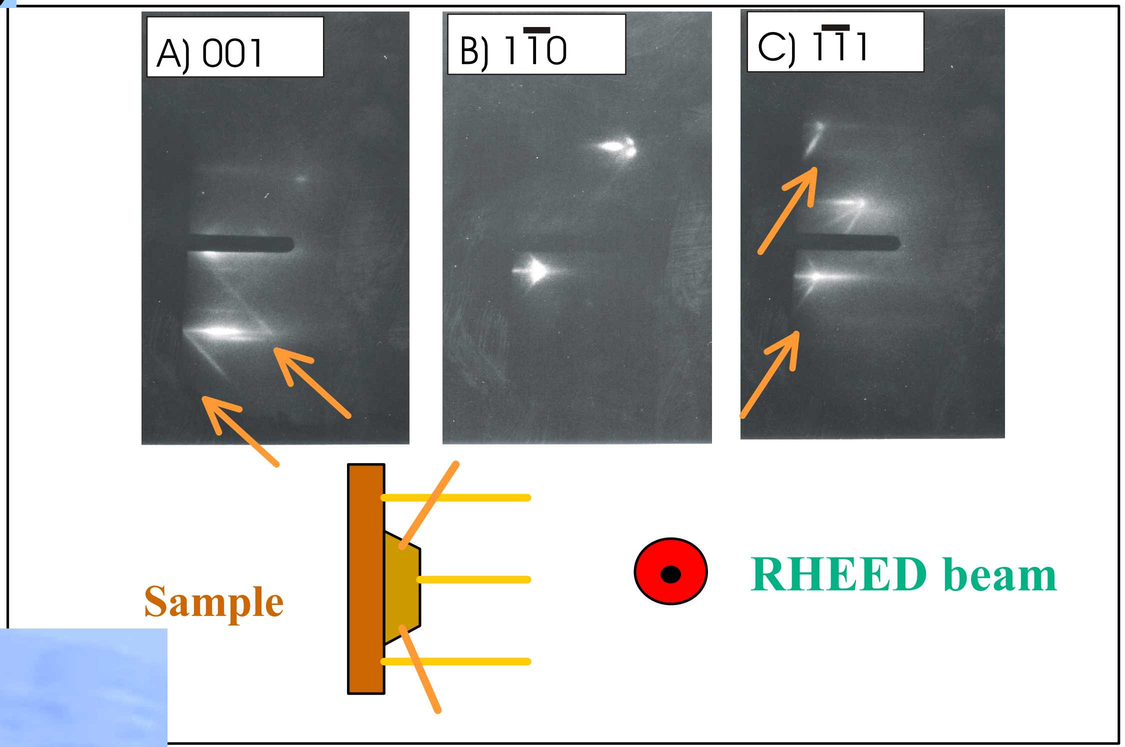
With strain:
Simulations or more complex models (e.g. small dots)

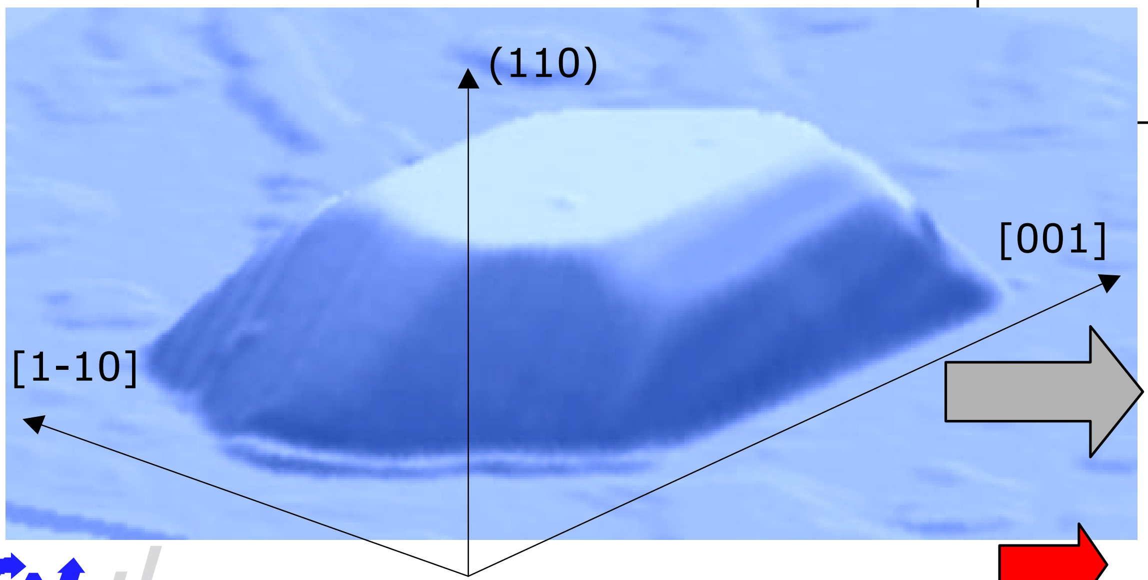
P. Müller and R. Kern, Surf. Sci. 457, 229 (2000)

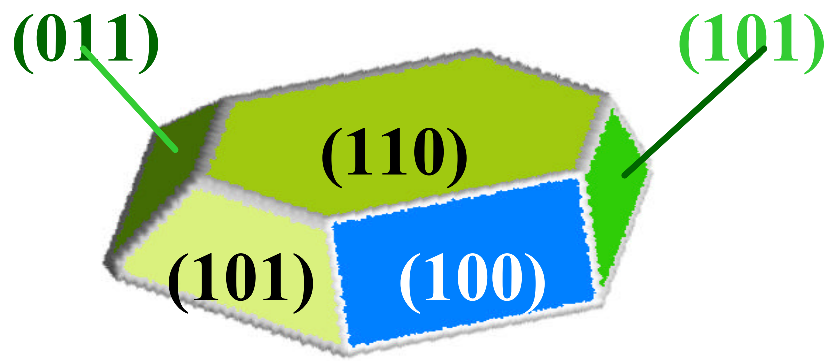
Fe/Mo(110)



AFM, 18mmx18mm





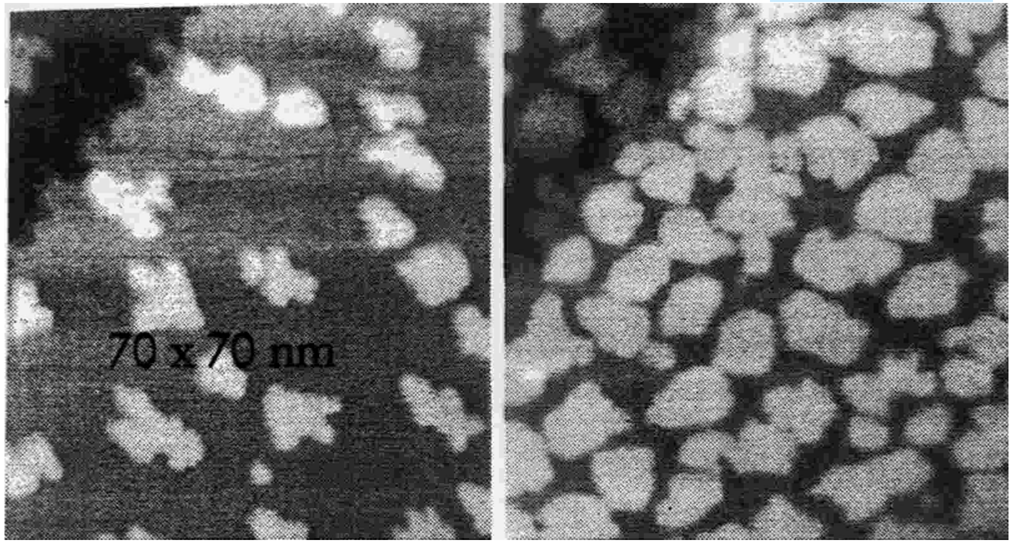


Model magnetic system

Very general phenomenon : many systems are suitable.

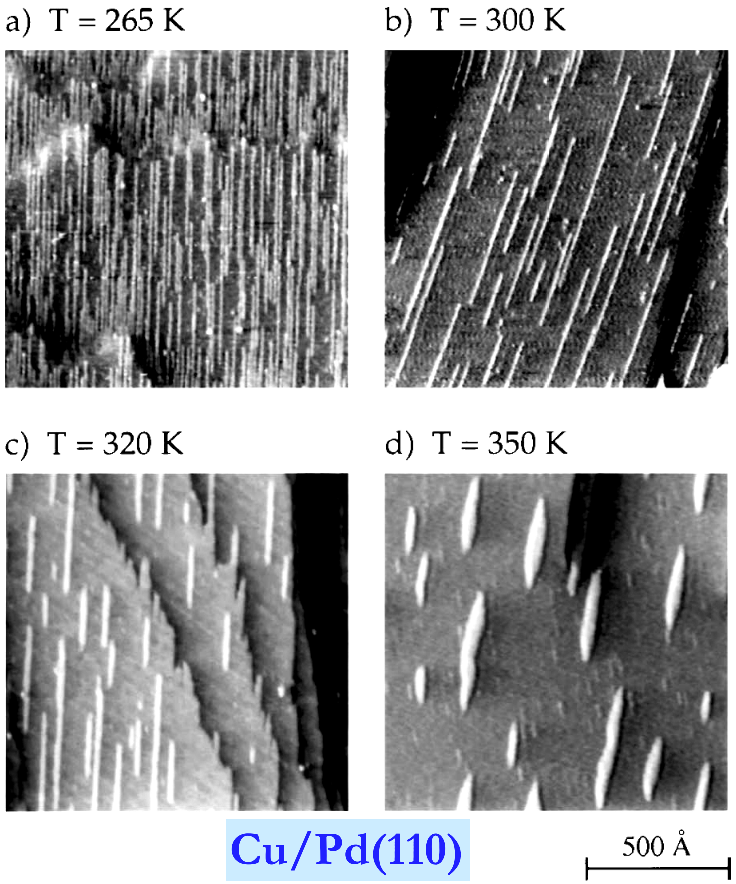
- Parameter : **coverage** $0 < x < 1$

Fe/W(110)



H.J.Elmers *et al.*, Phys.Rev.Lett.73, 898(94)

- Parameters: substrate **symmetry** and **temperature**



Cu/Pd(110)

J.P.Bucher *et al.*, Europhys.Lett.**27**, 473(93)

↳ « ***Test of scaling theory at a two-dimensional Ising-like transition*** »

Ch. Würsch et al., J. Magn. Magn. Mater 177-181, 617 (1998).

↳ Size and density of the dots can be tuned nearly independently

(1. Introduction)

2. Self-assembled epitaxial growth

3. Self-organized epitaxial growth

4. 3D self-organization via multilayer stacking

5. Perspectives of self-organization

6. X-ray investigation of SO systems

References

Self-assembly + 'long-range' positional order between dots.

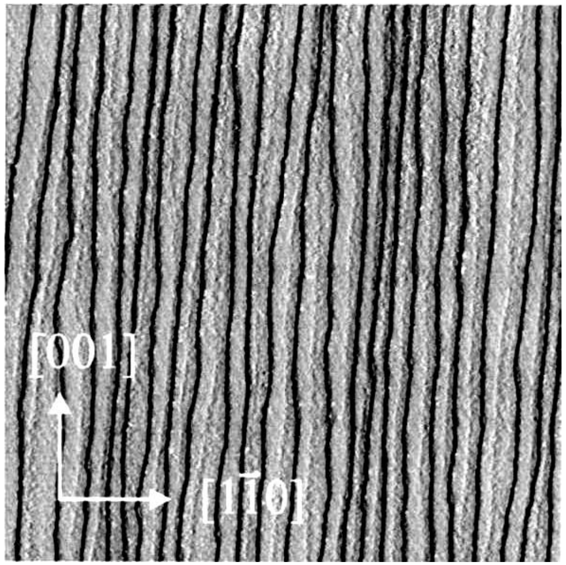
Dot-dot interactions << dot-substrate : the organization must be supplied by the substrate

STEP DECORATION

- Array of **atomic steps**. Ex: stripes (1D)

Vicinal Fe(110)/W(110)

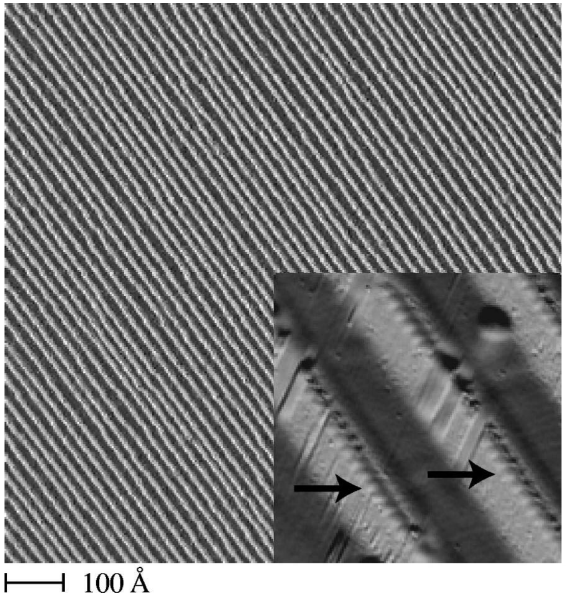
250 x 250 nm

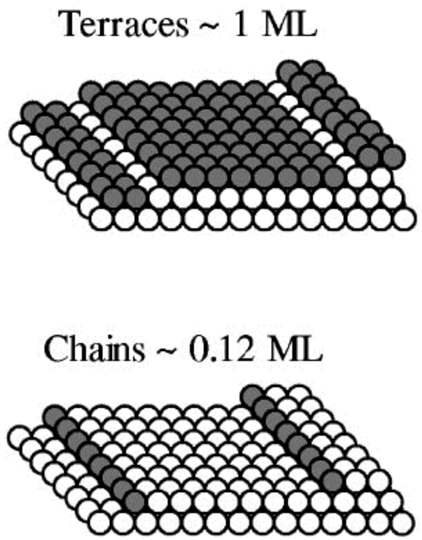


J.Hauschild *et al.*, Phys.Rev.B57, R677(1998)

Magnetic order in 1D (stabilized by dipolar interactions)

- **Ex:** Stripes and wires (1D)





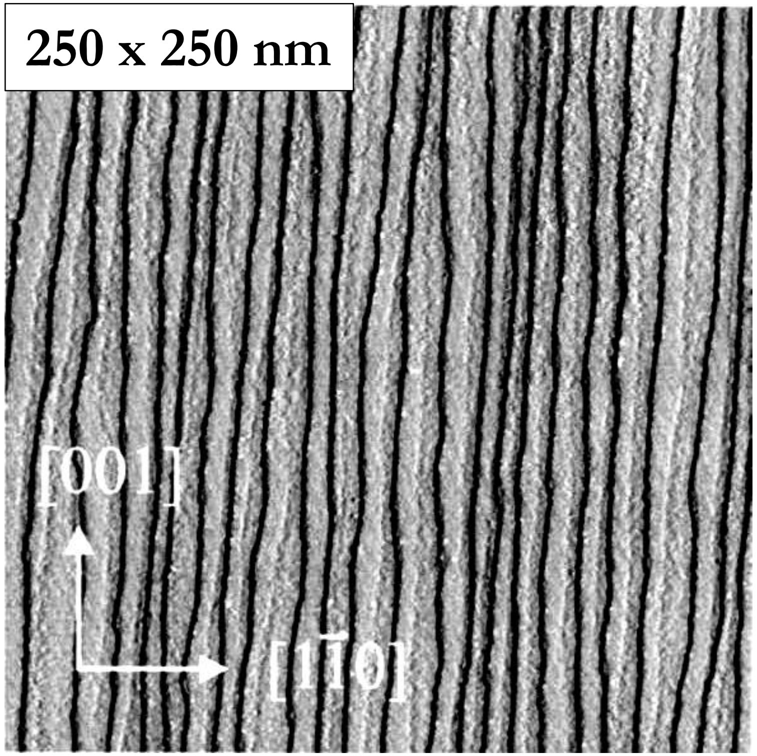
A. Dallmeyer *et al.*, Phys.Rev.B 61(8), R5153 (2000).

Co(250K - 0.12ML)/**Pt(997)**

Magnetic order and anisotropy in 1D

Example: Vicinal Fe(110)/W(110)

Thickness = 0.5AL

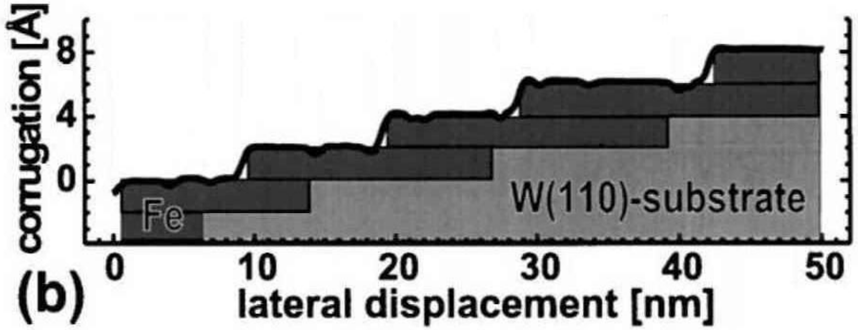


Tc=179K

Magnetic ordering in 1D (stabilized by dipolar interactions)

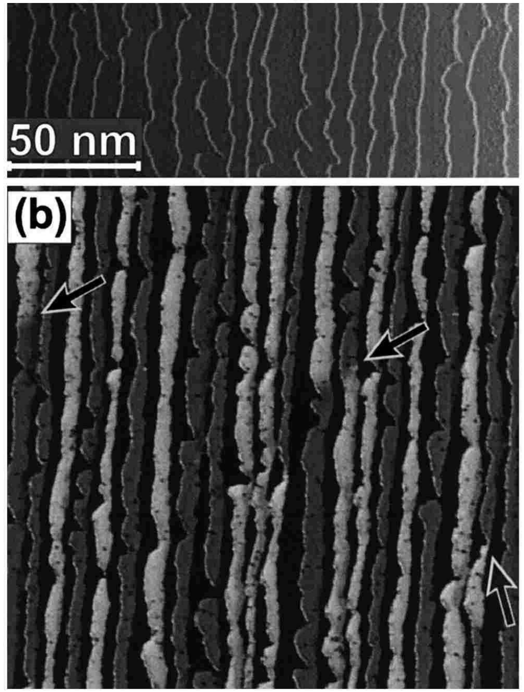
J.Hauschild *et al.*, Phys.Rev.B57, R677(1998)

Thickness = 1.5AL



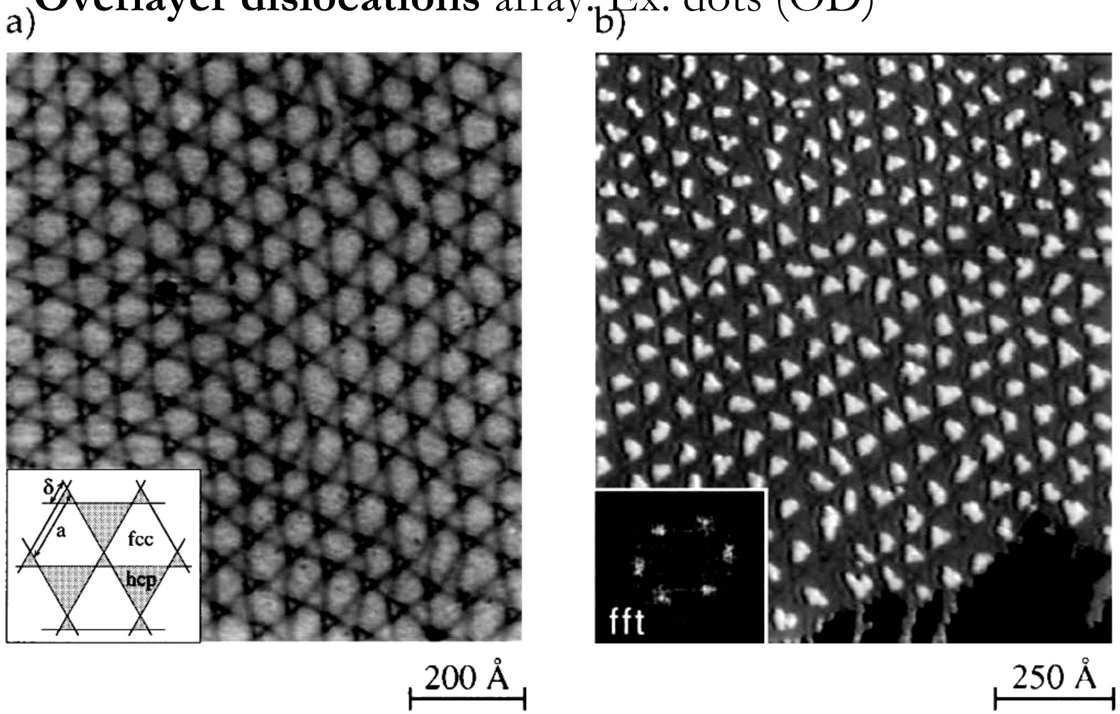
Spin-Polarized Scanning Tunneling Spectroscopy

(low temp.)



M. Bode et al, J. Electr. Spectr. Rel. Phenom. 114– 116, 1055 (2001)

- **Overlayer dislocations** array. Ex: dots (OD)

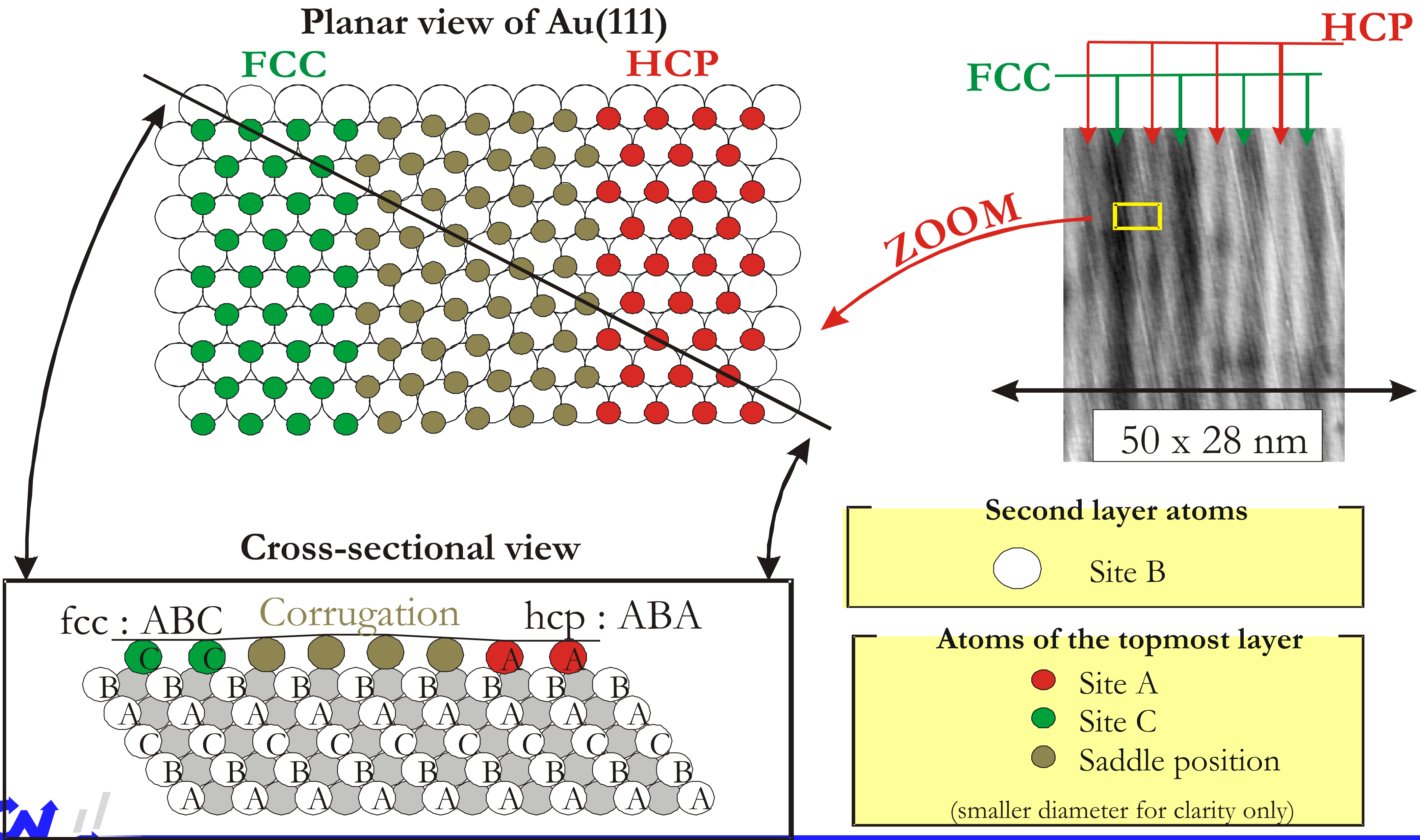


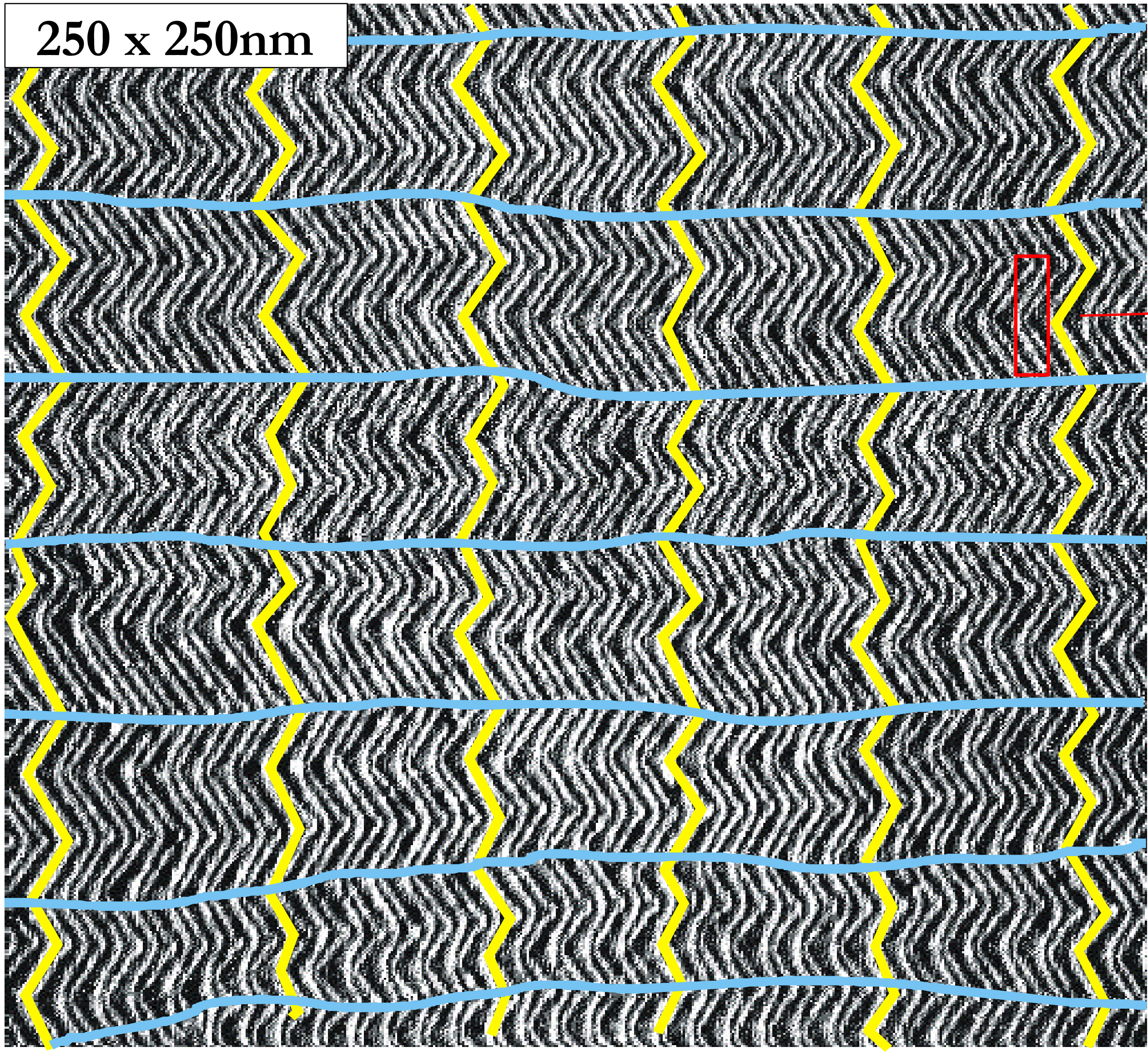
H.Brune *et al.*, Nature **394**, 451 (1998)

Ag(110K)/Ag(2ML, 400K annealed at 800K)/Pt(111)

Also : **improvement of size uniformity of the dots**

D.Y.Petrovykh, Surf.Sci. 407, 189 (1998)


Planar view of Au(111)
FCC
HCP
HCP
FCC
ZOOM
50 x 28 nm
Cross-sectional view
fcc : ABC
Corrugation
hcp : ABA
C C A A
B B B B B B B B B
A A A A A A A A A
C C C C C C C C C
B B B B B B B B B
A A A A A A A A A
Second layer atoms
Site B
Atoms of the topmost layer
Site A
Site C
Saddle position
(smaller diameter for clarity only)



Corrugation ~ 0.2 Å

Unit cell size :

~ 7.5 x 25nm

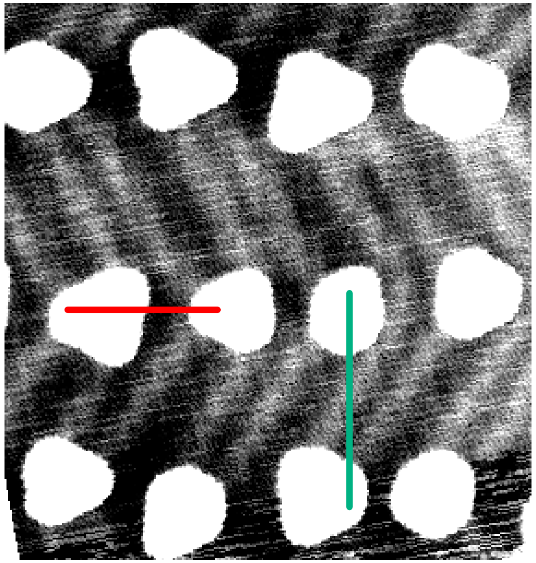
↳ Isotropic surface relaxation

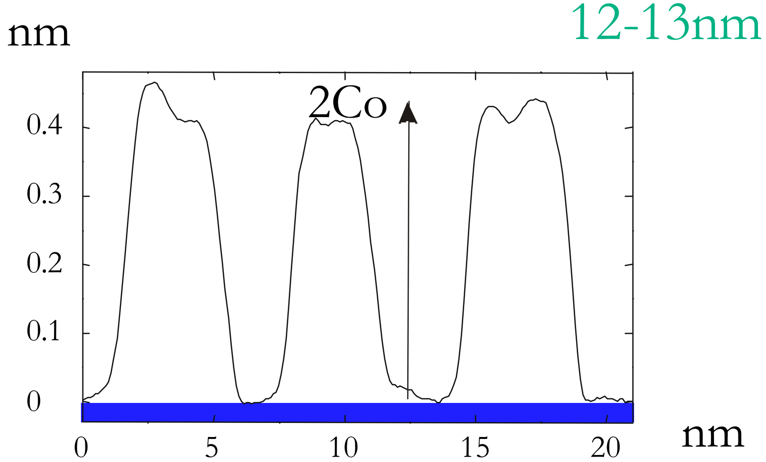
J. V. Barth et al., Phys. Rev. B 42 (15), 9307 (1990)

A.R. Sandy et al., Phys. Rev. B 43 (6), 4667 (1991)

- Co, Ni, Fe :
  Nucleation at the
  elbows of the chevrons

(0.20AL Co@300K)

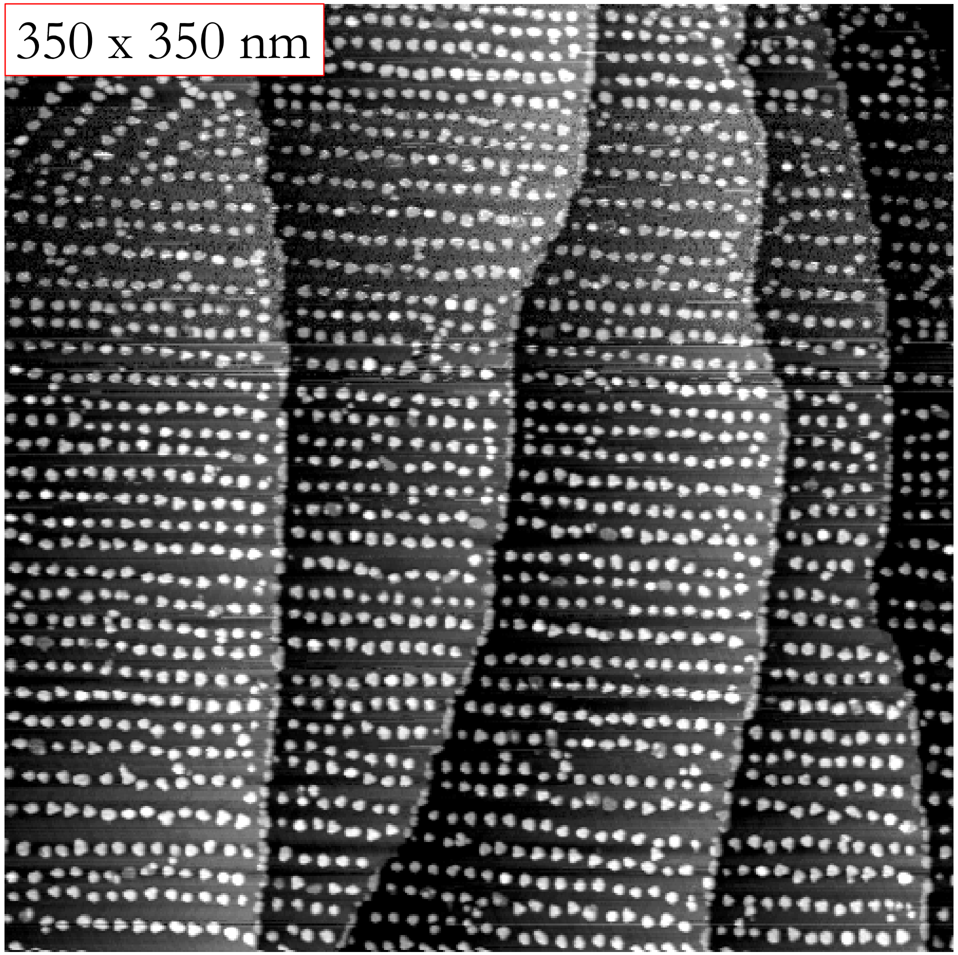




Fe, Ni : 1 AL-high dots

D.D. Chambliss et al., PRL **66**, 1721 (1991)

B.Voigtlander et al., PRB **44**, 10354 (1991)

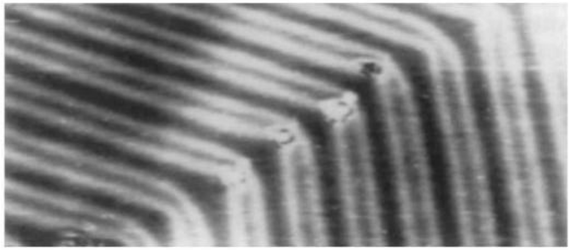


- Medium-ranged organization
- Steps do not necessarily disturb the order
  [see also:
  V.Repain, Europhys. Lett., **47** (4), 435 (1999)]

Fe, Co, Ni (etc.) nucleation: atomic place exchange mechnism with Au atoms

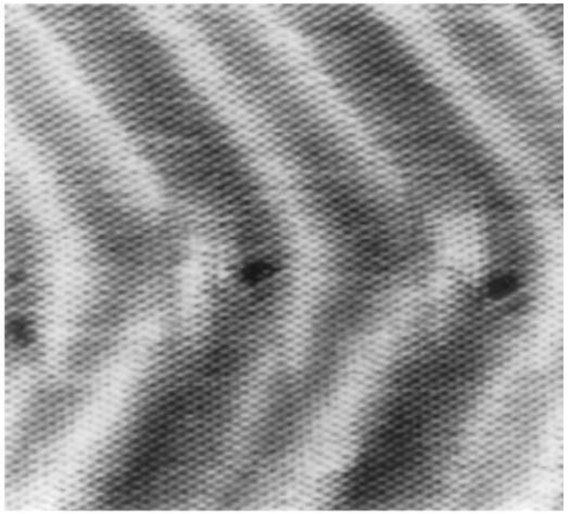
Example: 0.002ML Ni@300K

a)



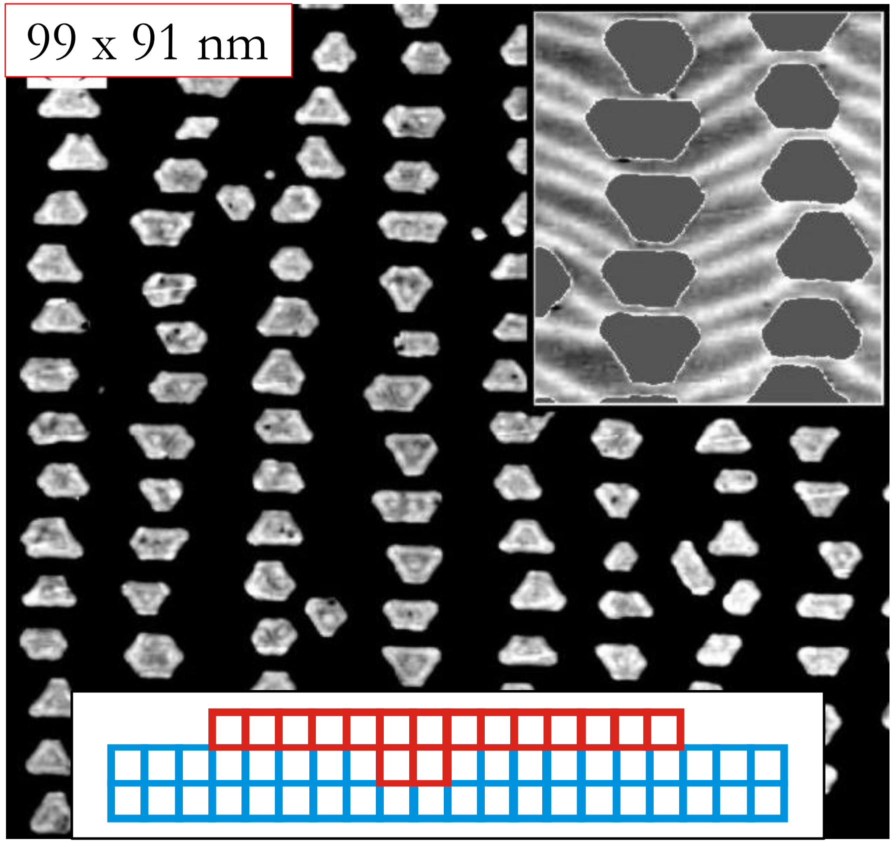
Element	Surface free energy (eV)	Heat of sublimation (eV)
Ag	0.50	2.95
Al	0.56	3.39
Cu	0.69	3.51
Au	0.72	3.79
Ni	0.90	4.45
Co	0.94	4.40
Fe	0.96	4.32

b)



J.A.Meyer et al., Surf.Sci.**365**, L647 (1996)

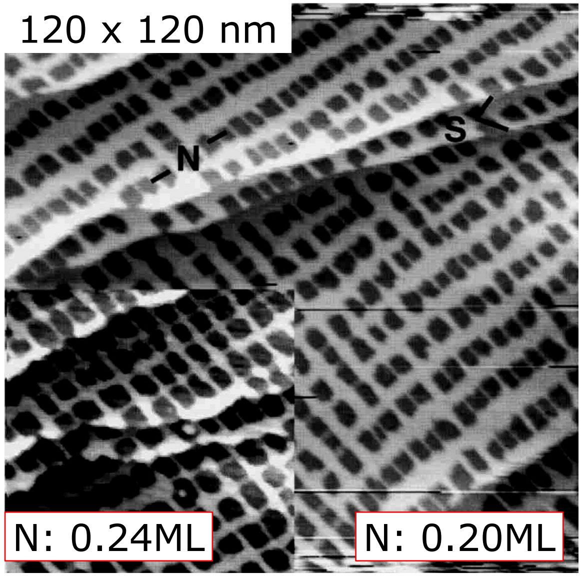
0.25ML Ni@300K : 1ML-high dots



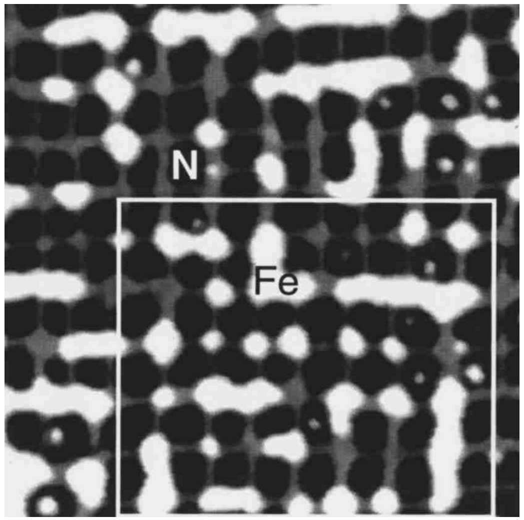
W.G.Cullen *et al.*, Surf.Sci.**420**, 53 (1999)

Leading parameter : deposit has a higher surface energy.
(and Au atoms stress near chevrons)

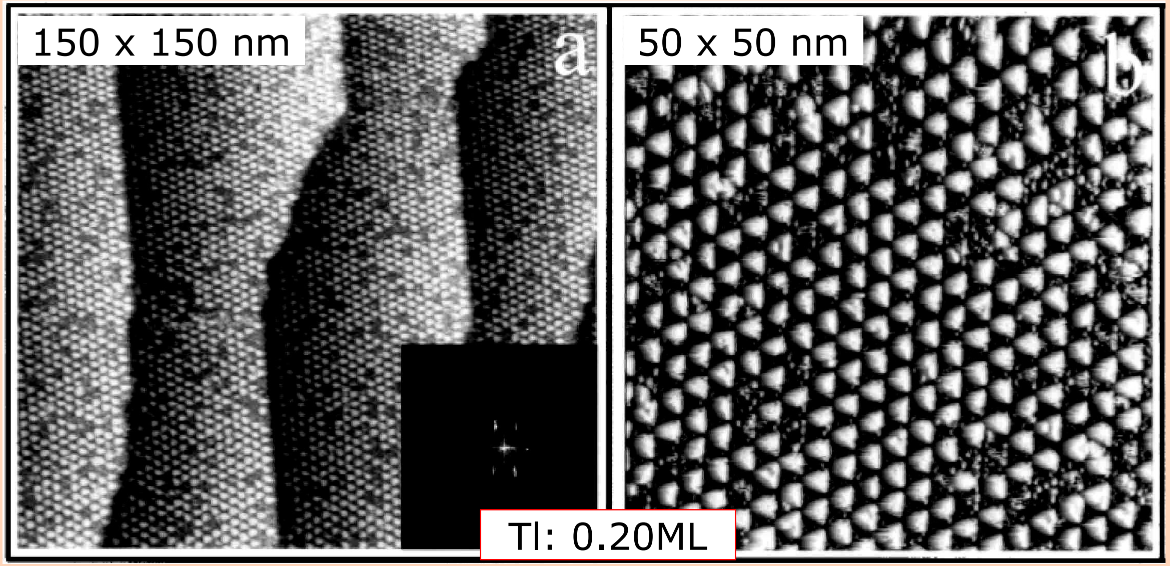
- Self-organized **adsorbtion**: N on Cu(100)



T.M.Parker, Phys.Rev.**B56**, 6458(1997)

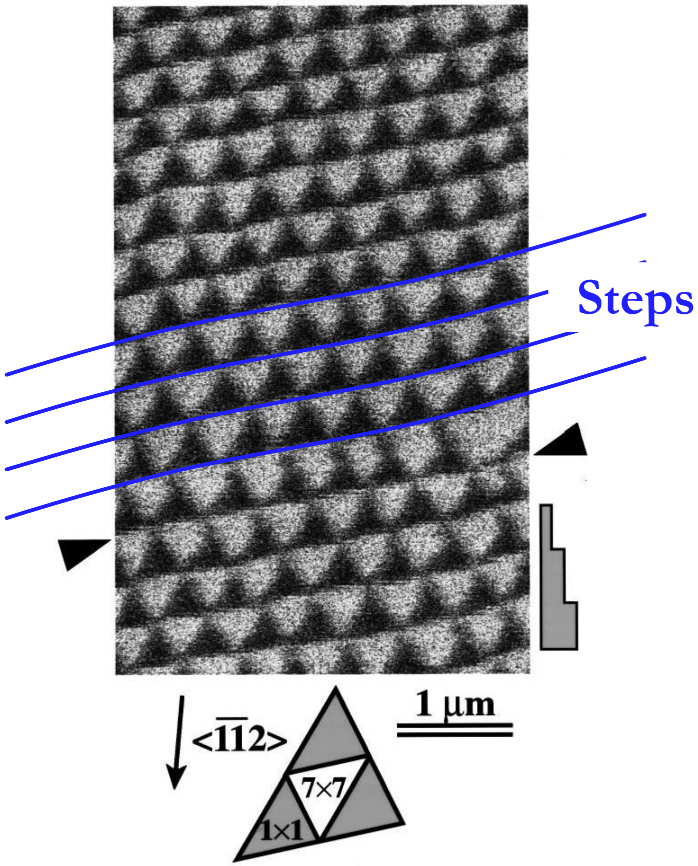


- Self-organized growth on **reconstructions**
  Tl on Si(111) 7x7



L.Vitali, Phys.Rev.Lett.**83**(2), 316 (1999)

- Superstructure of reconstructions: **7x7 and 1x1 triangles** on Si(111), misoriented along <11-2>



H.Hibino, Phys.Rev.**B58**(12), R7500 (1998)

- **Co on vicinal Au(111)**

Steps and reconstructions

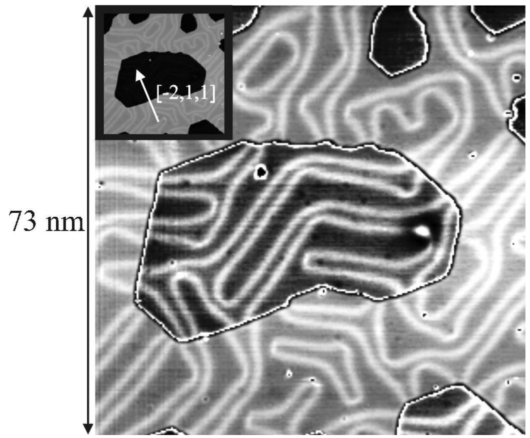


FIG. 4. STM image of a few vacancies islands. T=300 K. In inset, the raw STM image. The same image is shown where the corrugation due to the terrace levels has been subtracted in order to enhanced the reconstruction lines.

Reconstructions lie perpendicular to steps

1ML Co

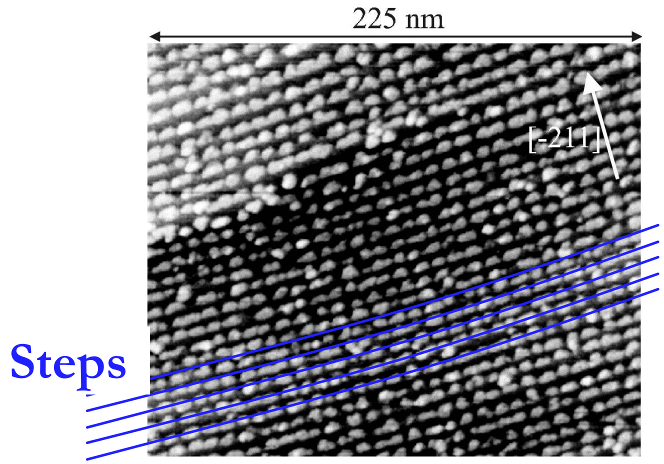


FIG. 6. Rectangular lattice (7 nm x 8 nm) of cobalt clusters on a vicinal surface misoriented from 1.7° with respect to Au(111) towards the [-211] azimuth. Cobalt coverage is 1 ML. Arrow indicates the descending steps direction.

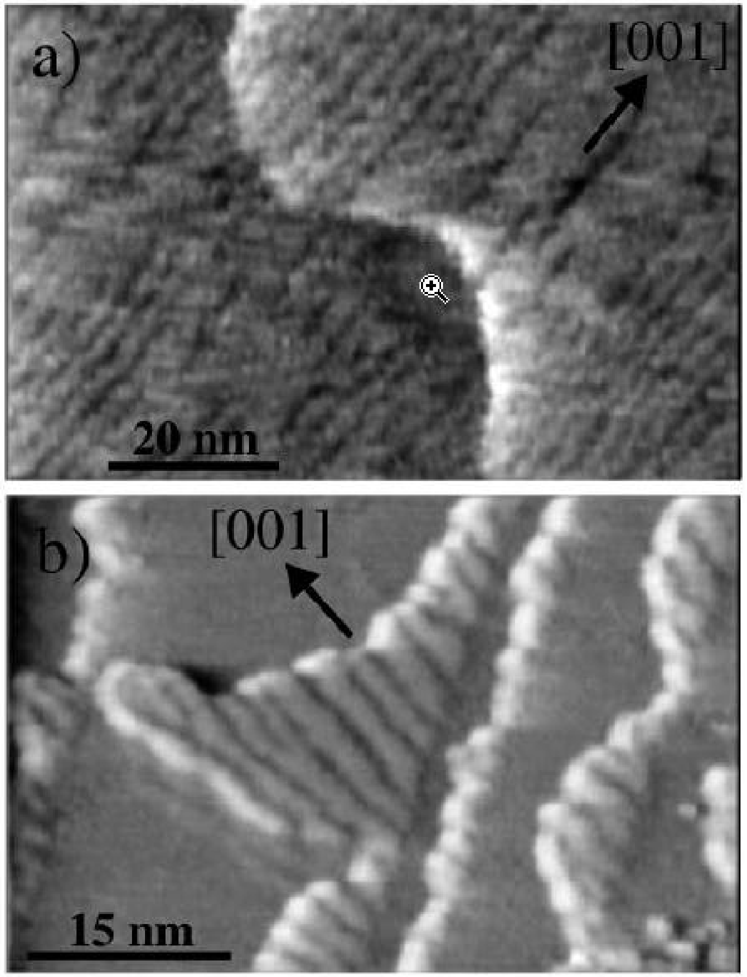
Nucleation both at steps and at reconstructions

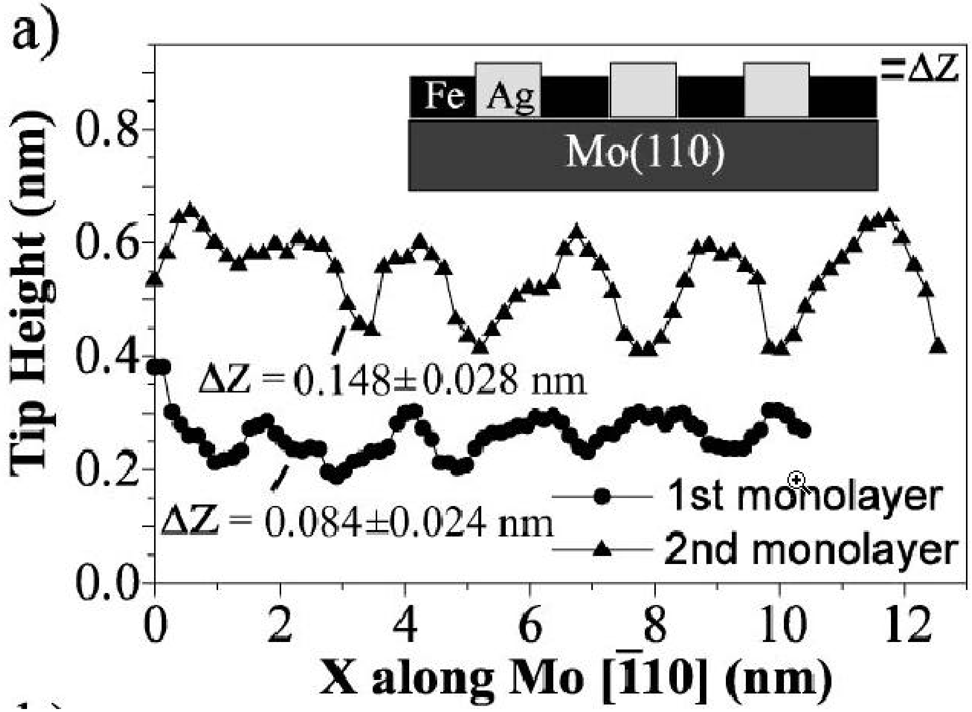
V.Repain, Europhys. Lett., **47** (4), 435 (1999)
V.Repain, ACSIN5 proceedings

Lateral modulation in ultrathin films

FeAg/Mo(110)
1AL

CoAg/Mo(110)
0.3AL





E. D. Tober et al., PRL81, 1897 (1998)

Stéphane ANDRIEU: possible link with Fe/V(110) ?

- Self-organization generally from substrate, not from deposit !
- Relies on surface science fundamental investigations

combine

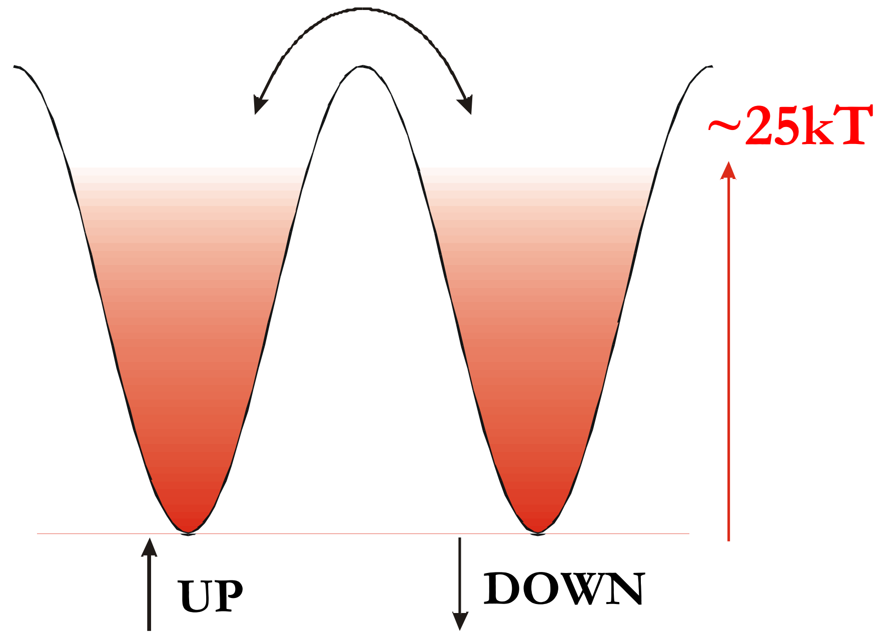
- Surface reconstructions
- Overlayer array of dislocations
- Adsorbates patterns
- 1D or 2D array of steps

Better size control

- Steps (or bonding ?) and reconstructions

- Good pattern does not necessarily imply good overgrowth

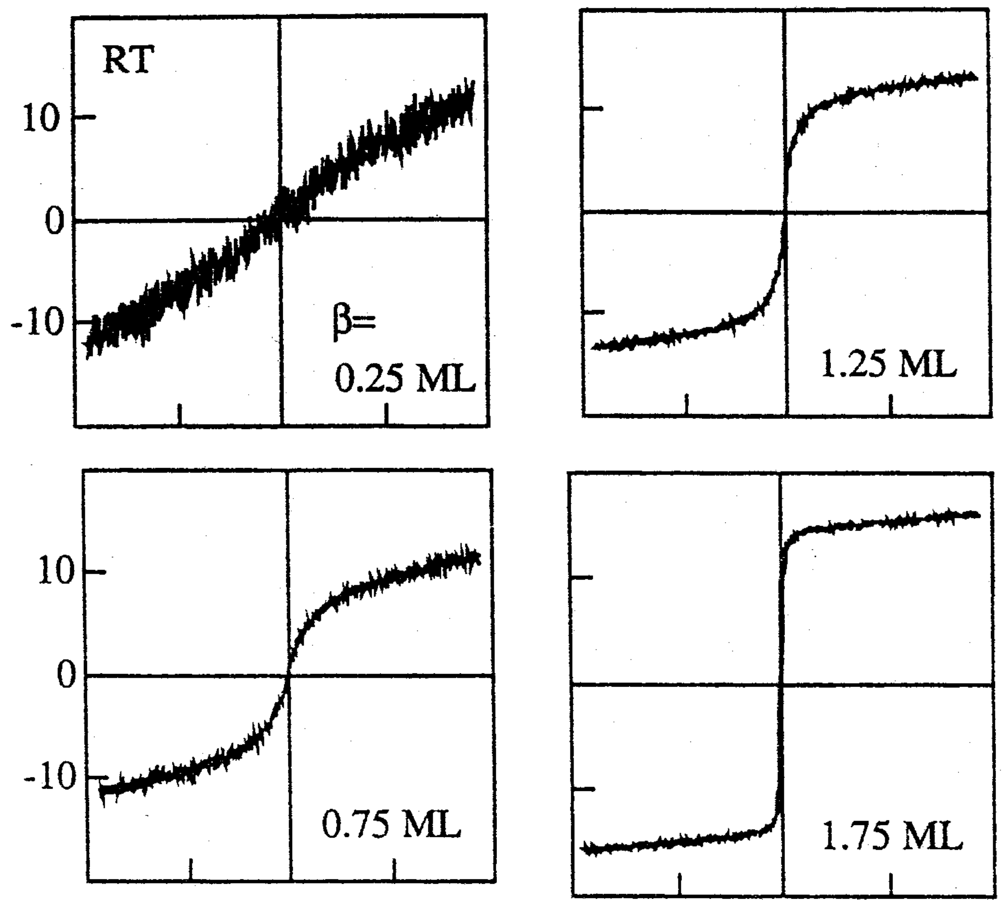
Anisotropy barrier ~KV



Blocking temperature Tb ~ 20K

H.Dürr et al., PRB**59**, R701 (1999)

Example: Co/Au



H.Takeshita et al., JMMM**165**, 38 (1997)

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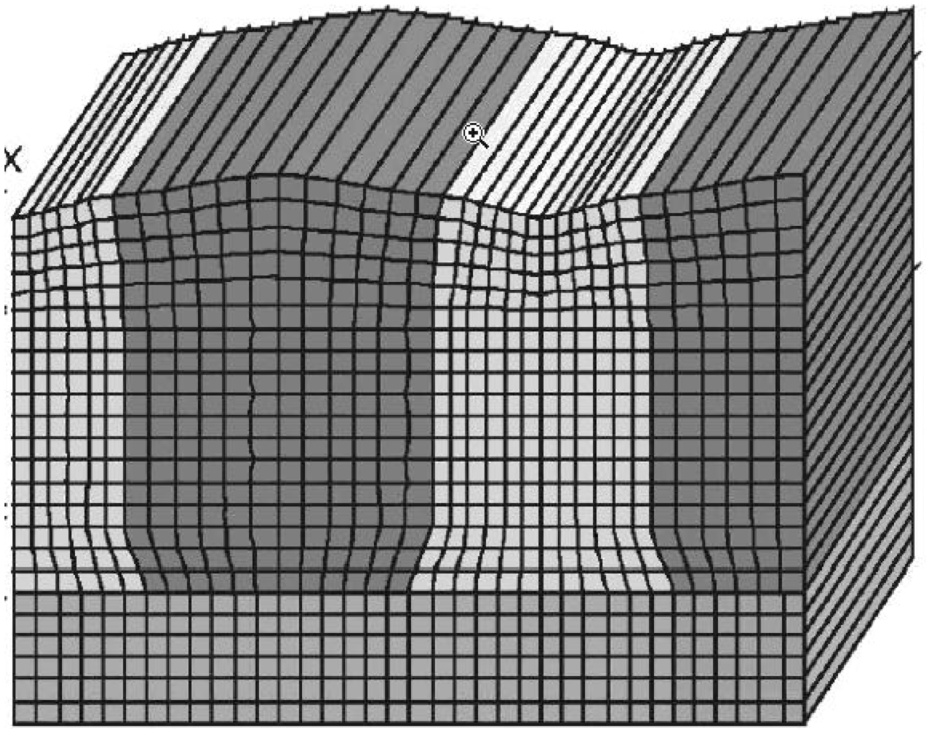
References

Principle

Strain release
Here: accumulation during growth

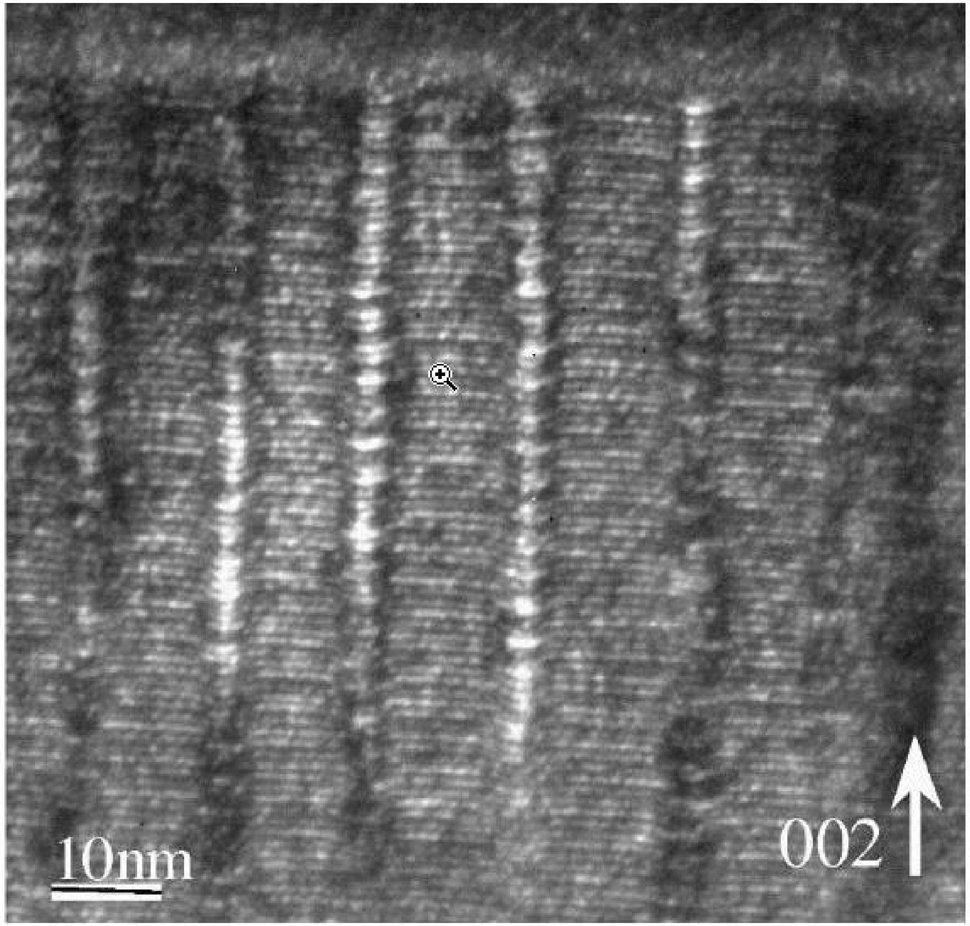


R. D. Twesten et al, PRB60, 13619 (1999)

[Cf Grinfeld instability: M. A. Grinfeld, Dok. Akad. Nauk SSSR 290, 1358 (1986)]

Experiments

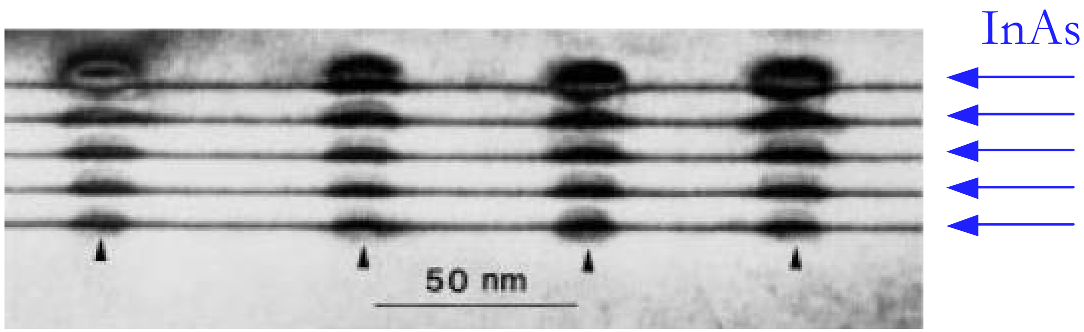
InAs/AlAs short-period multilayers



R. D. Twesten et al, PRB60, 13619 (1999)

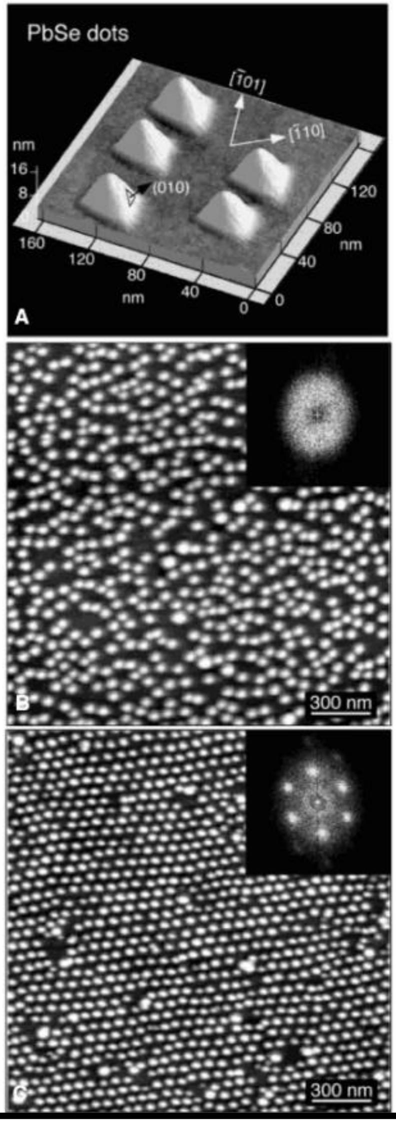
InAs / GaAs(100)

- Strain field in spacer layer :
  - vertical stacking of dots



Q.Xie, Phys.Rev.Lett.75(13), 2542 (1995)

- From 2D assembly to 3D- organization



Zoom : facetted dots

Single layer : Self-assembly, no organization.

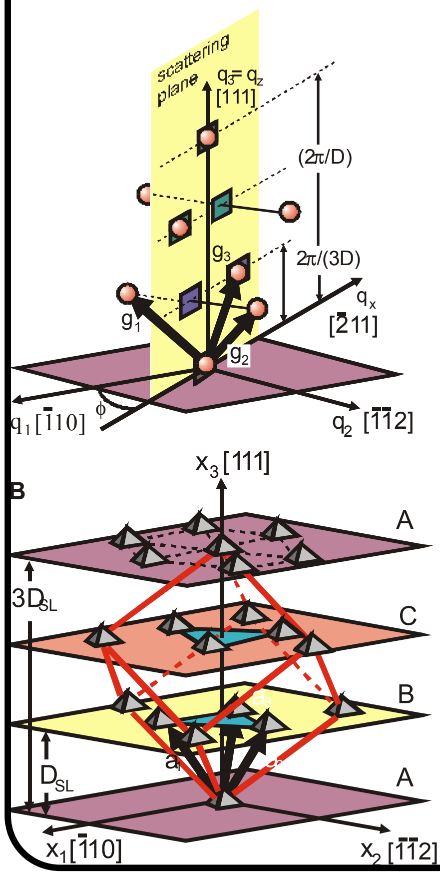
Multilayers : improvement of the organization.

(After 60 layers)

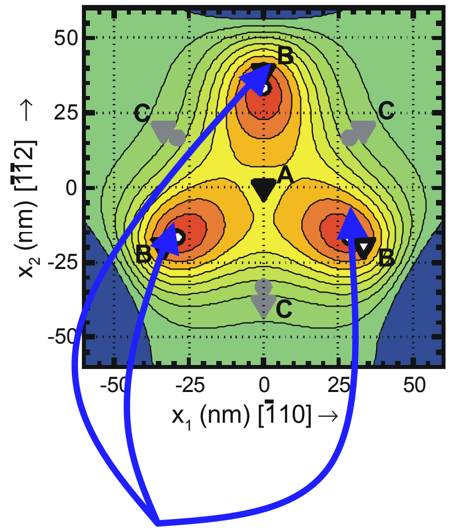
PbSe / PbTe(111)

- Anisotropic elastic media
  - f.c.c. superstacking, not vertical.

X-Ray diffraction



Anisotropic in-plane distribution of elastic energy density



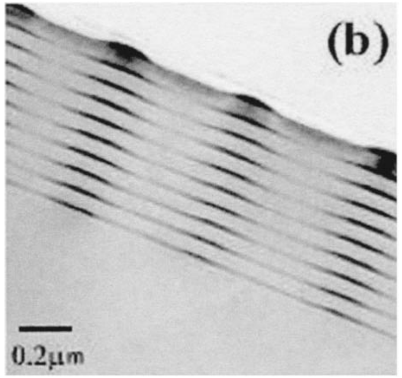
Misfit reduced by 2%

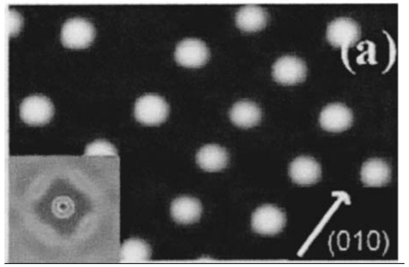
- preferential nucleation

G.Springholz, Science **282**, 734 (1998)

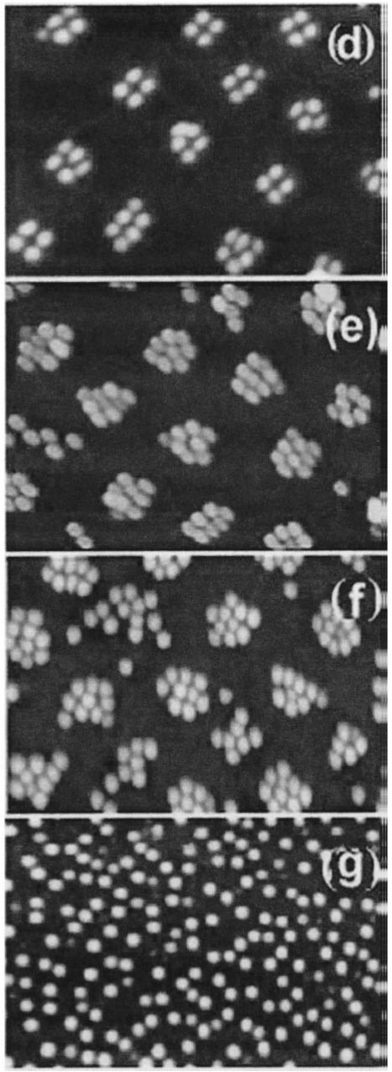
Step 1
multilayers for organization

Ge islands/Si(001)





Step 2
use as a template



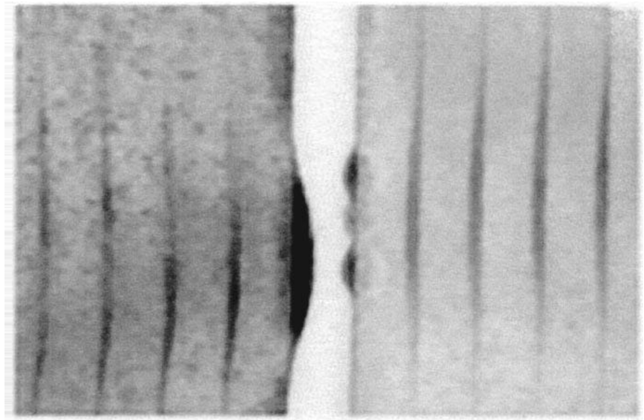
35nm

45nm

65nm

200nm

HREM view



Template

Overgrowth

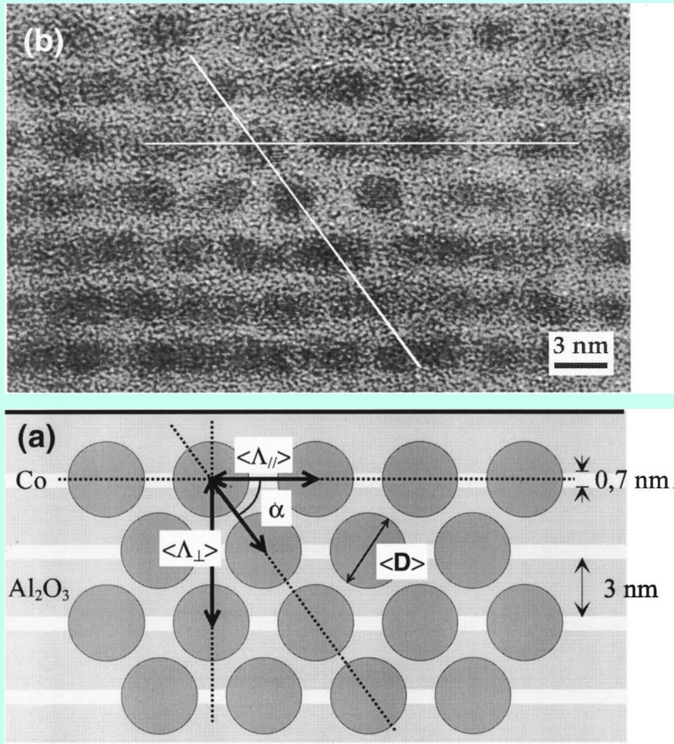
G. Capellini *et al.*, APL82, 1772 (2001)

Co/Al_2O_3 granular system (sputtering)

(Co: 0.7 nm/Al_2O_3: 3 nm)$_{14}$

REAL SPACE

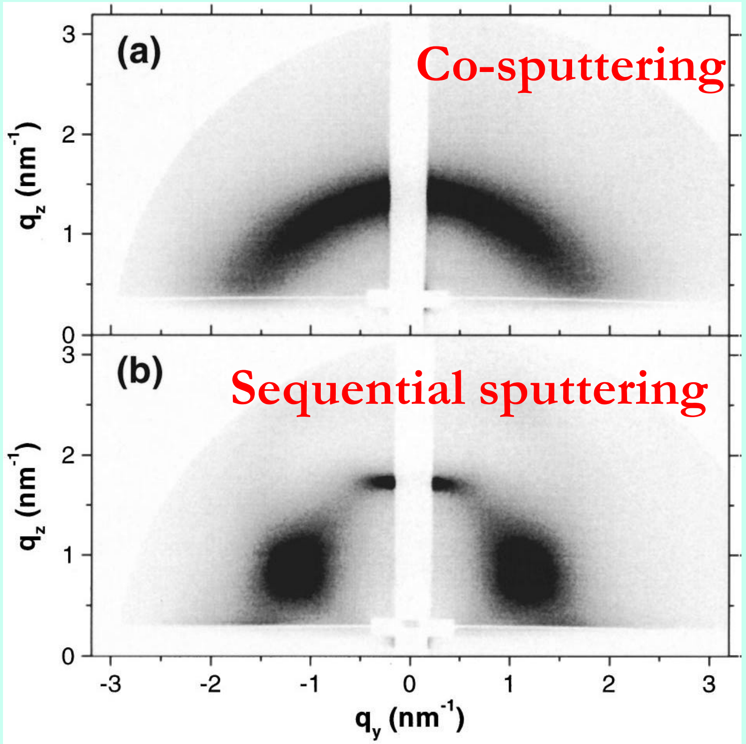
TEM cross-section (Sequential sputtering)



RECIPROCAL SPACE

GISAXS:

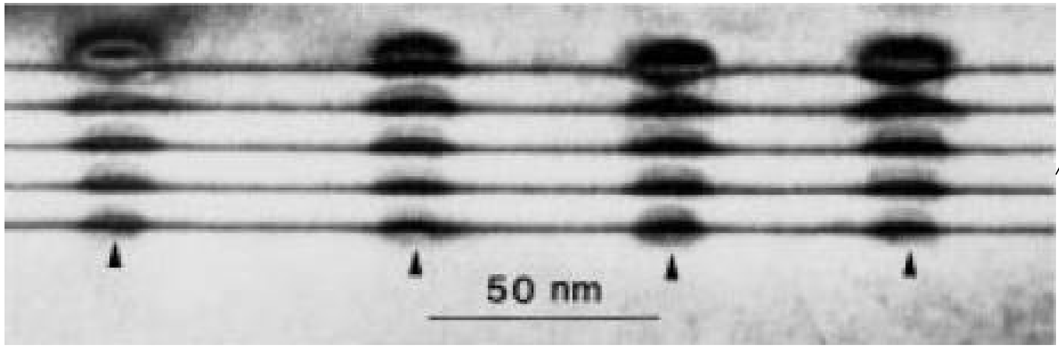
Grazing Incidence Small Angle X-Ray Scattering



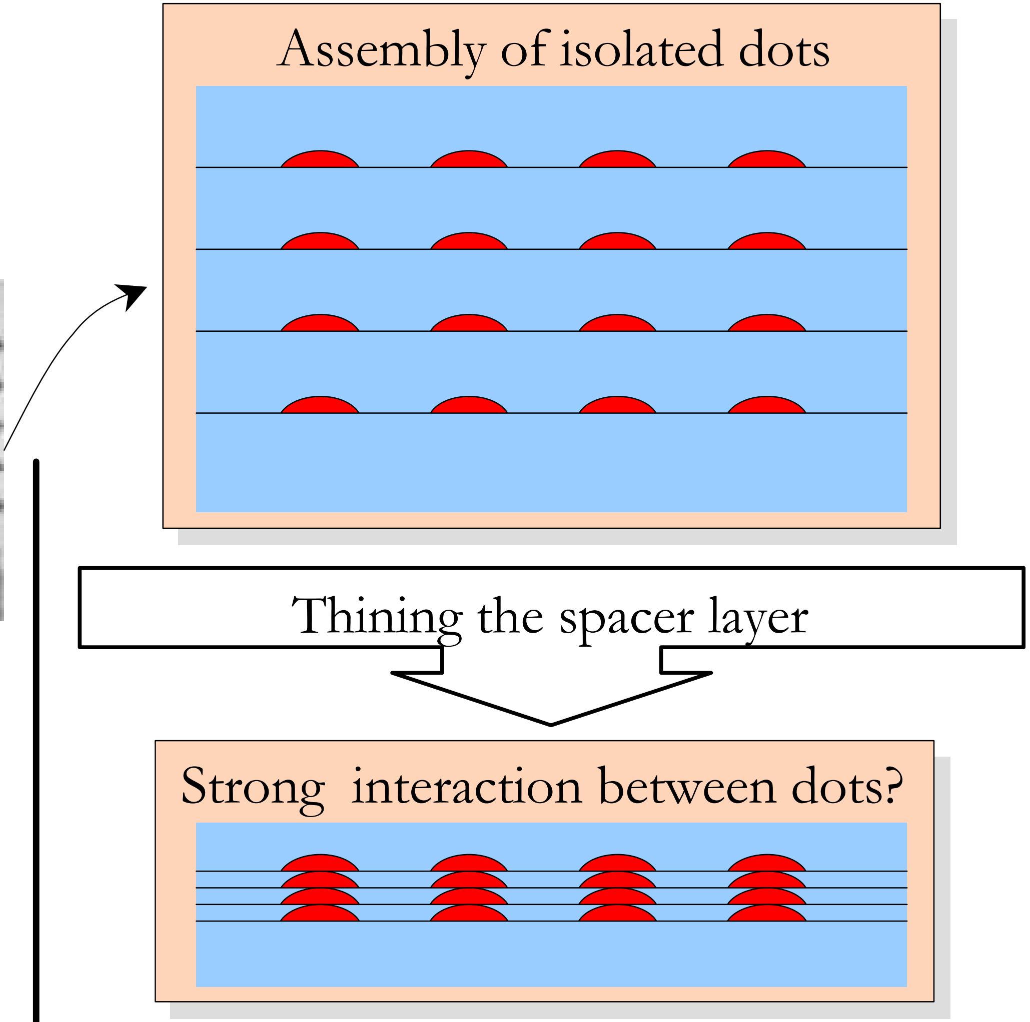
➢ FCC vertical stacking (not epitaxial !)

D. Babonneau, Appl. Phys. Lett. 76, 2892 (2000)

- Vertical 3D self-organization of $In_xGa_{1-x}As/GaAs$:



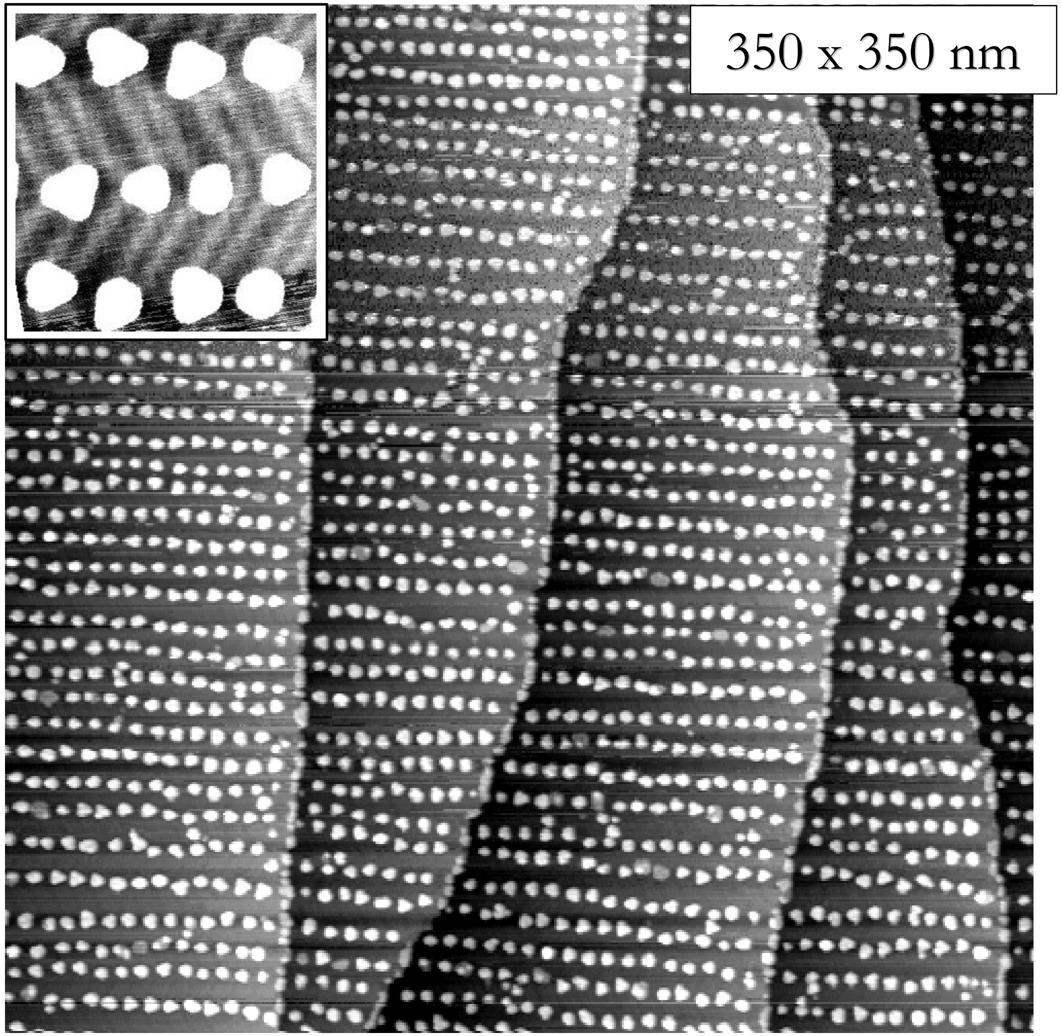
Q.Xie et al., Phys.Rev.Lett.75(13), 2542 (1995)



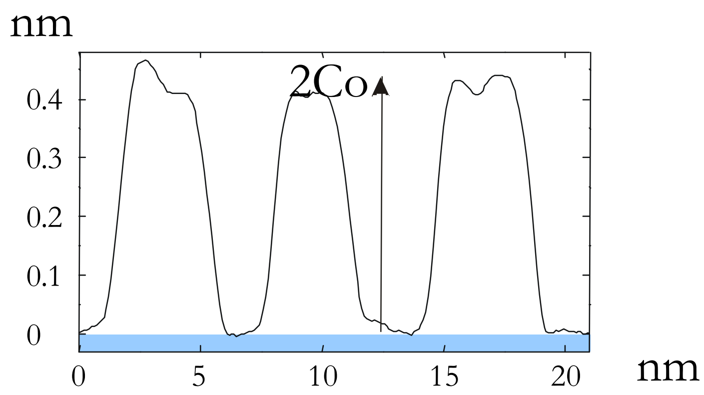
↪ superparamagnetism overcome ?

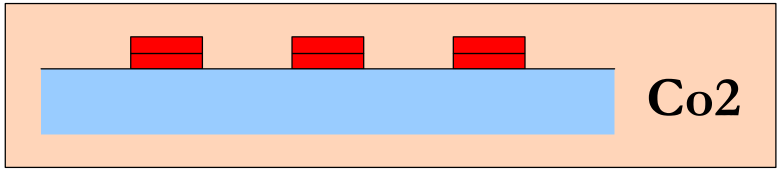
↪ Enhanced magnetic signal

➤ **Step 1** : 0.2ML Co @300K

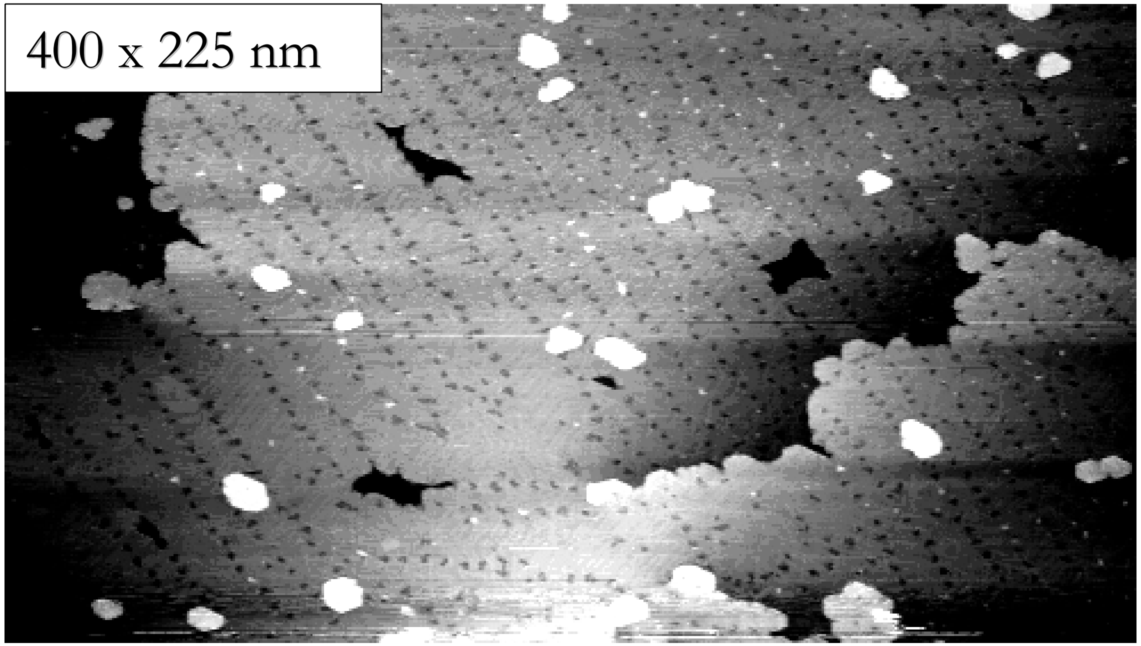


Typical cross-section :





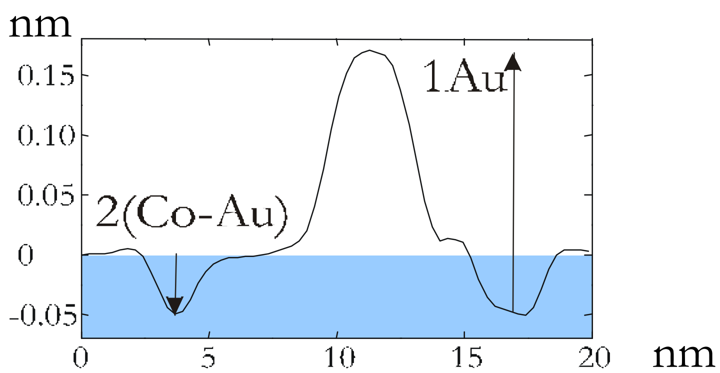
➤ **Step 2 :** 3.8ML Au @375↗410K

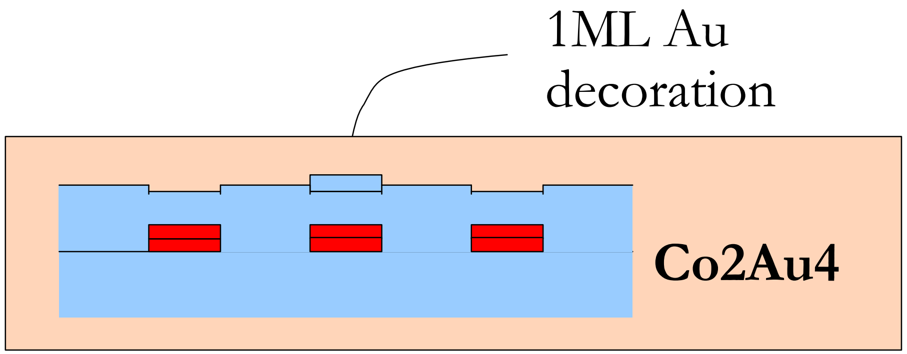


See also : Wollschläger *et al.*, Surf.Sci.**277**, 1 (1992)

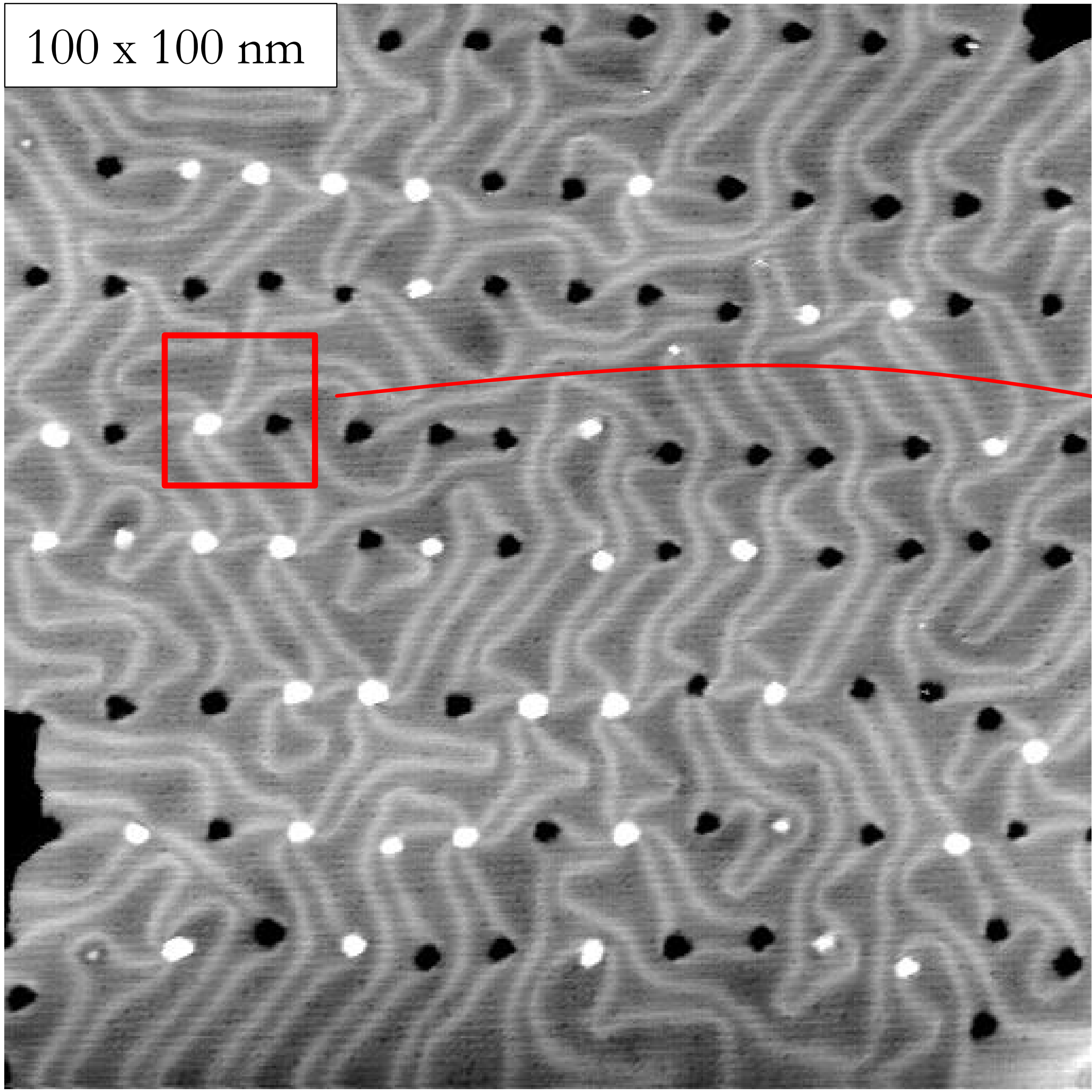
1ML Co hcp ≈ 0.205nm
1ML Au fcc ≈ 0.235nm
} Array of hollows

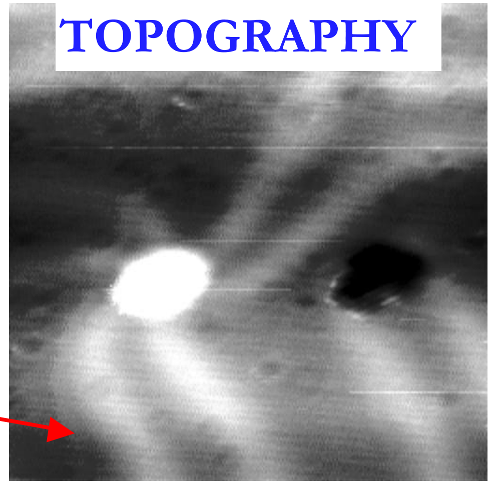
Typical cross-section :

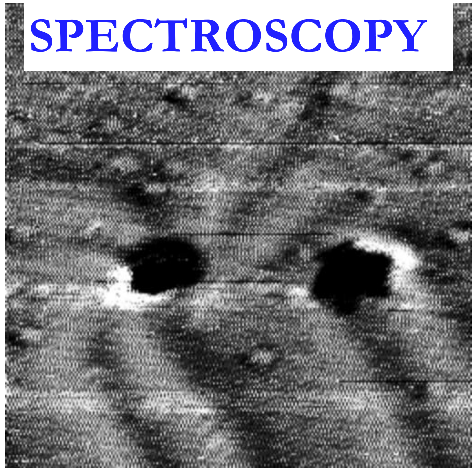




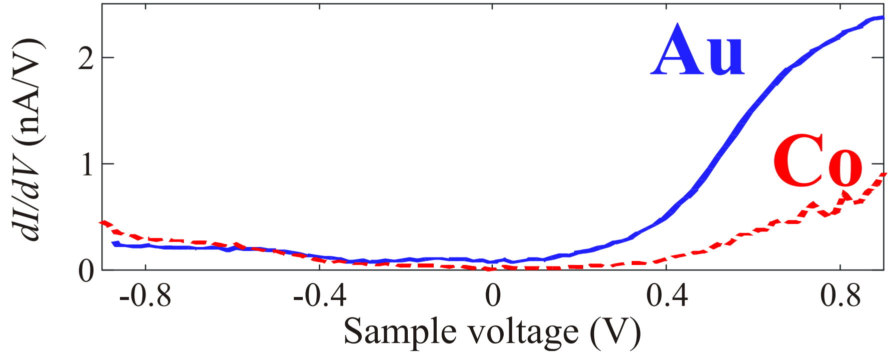
➤ Surface smoothing
➤ Unaffected Co dots





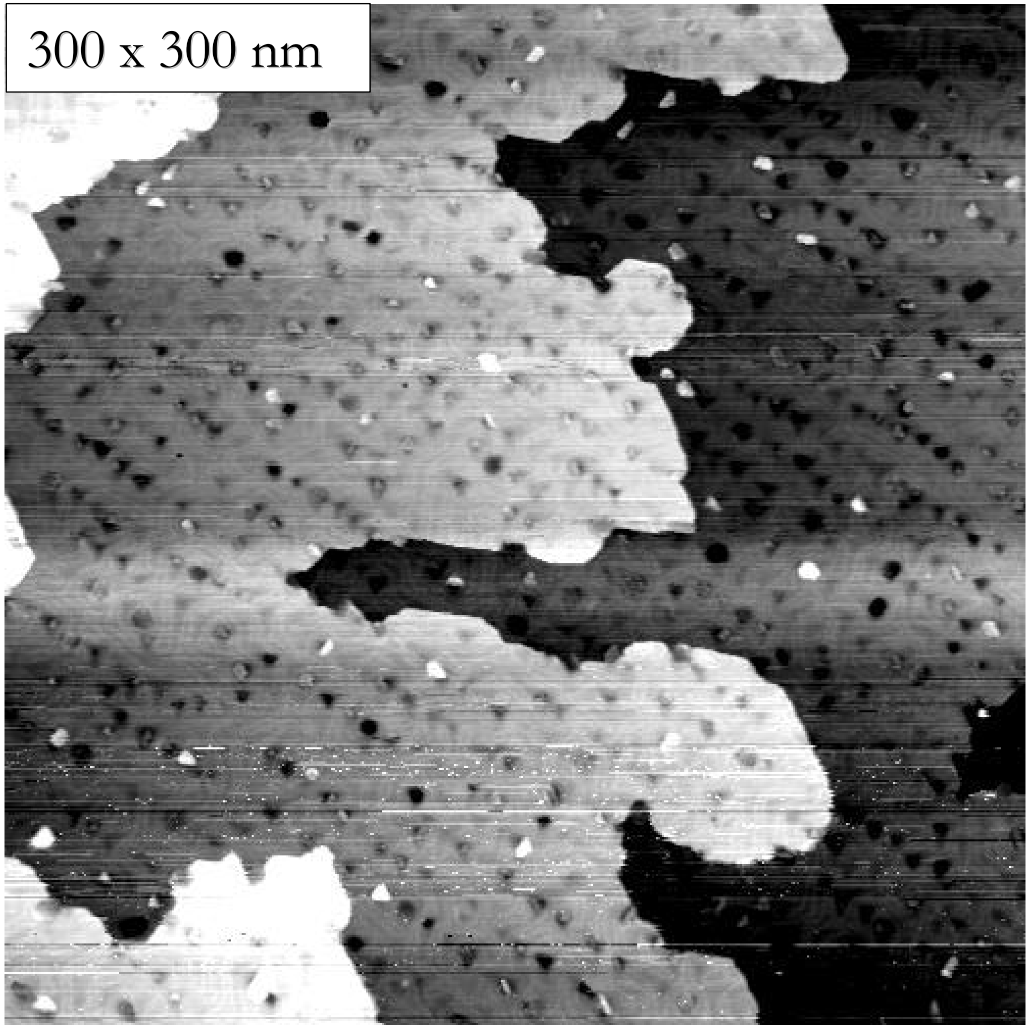


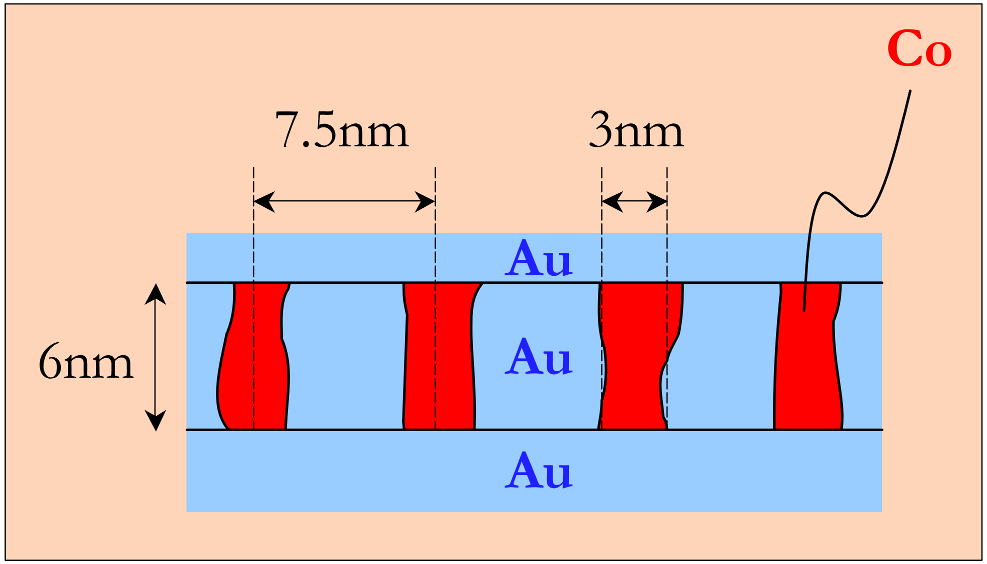
SAME AREA



Co atoms grow only on existing dots

⇘ **VERTICAL SELF-ORGANIZATION**

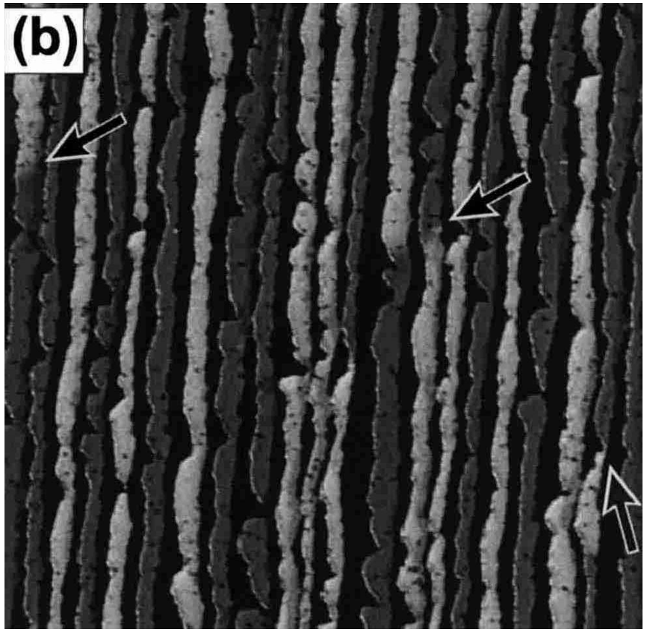




- Self-organization nearly undisturbed
- Pillars with 2:1 vertical aspect ratio
- Unclear to this point :
  - Exchange mechanism
  - Limitating factors ?
  - Composition, microstructure ?

ATOMIC LAYER RANGE : WETTING

↳ nanometer-world / surface physics



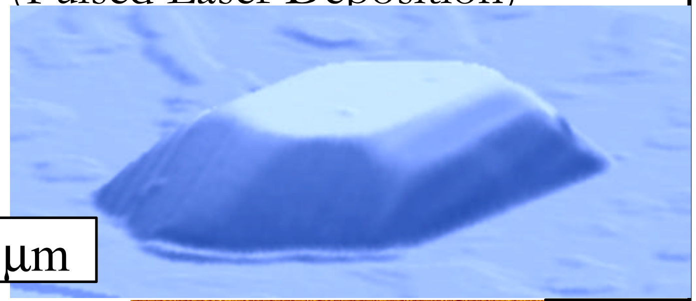
1.5AL on vicinal Fe/W(110)

M. Bode et al, J. Electr. Spectr. Rel. Phenom. 114– 116, 1055 (2001)

THICK DEPOSITS : NO WETTING

↳ micrometer-world / materials physics

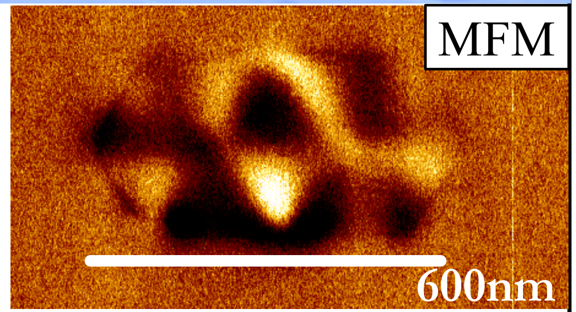
Fe/Mo(110) (Pulsed Laser Deposition)



MFM:

Y. Samson

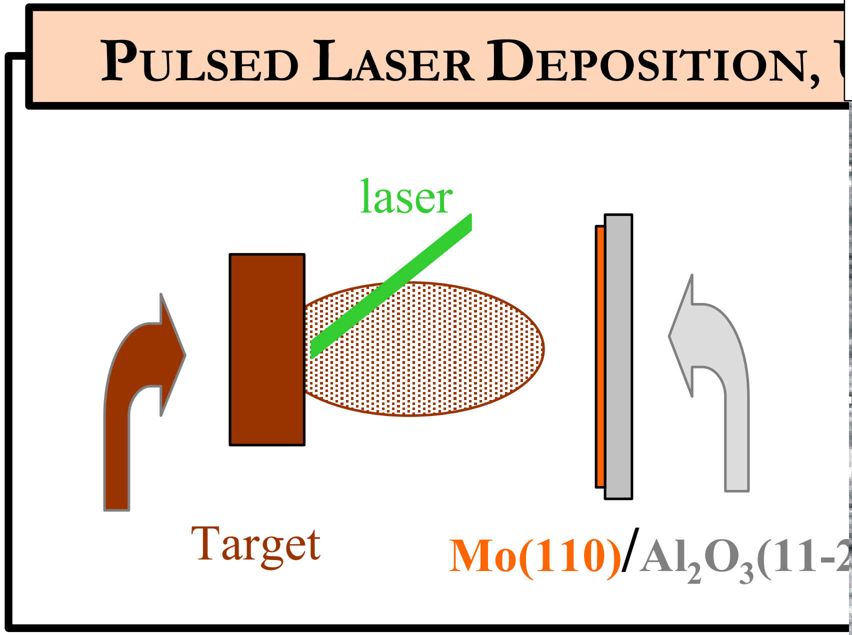
(CEA/France)

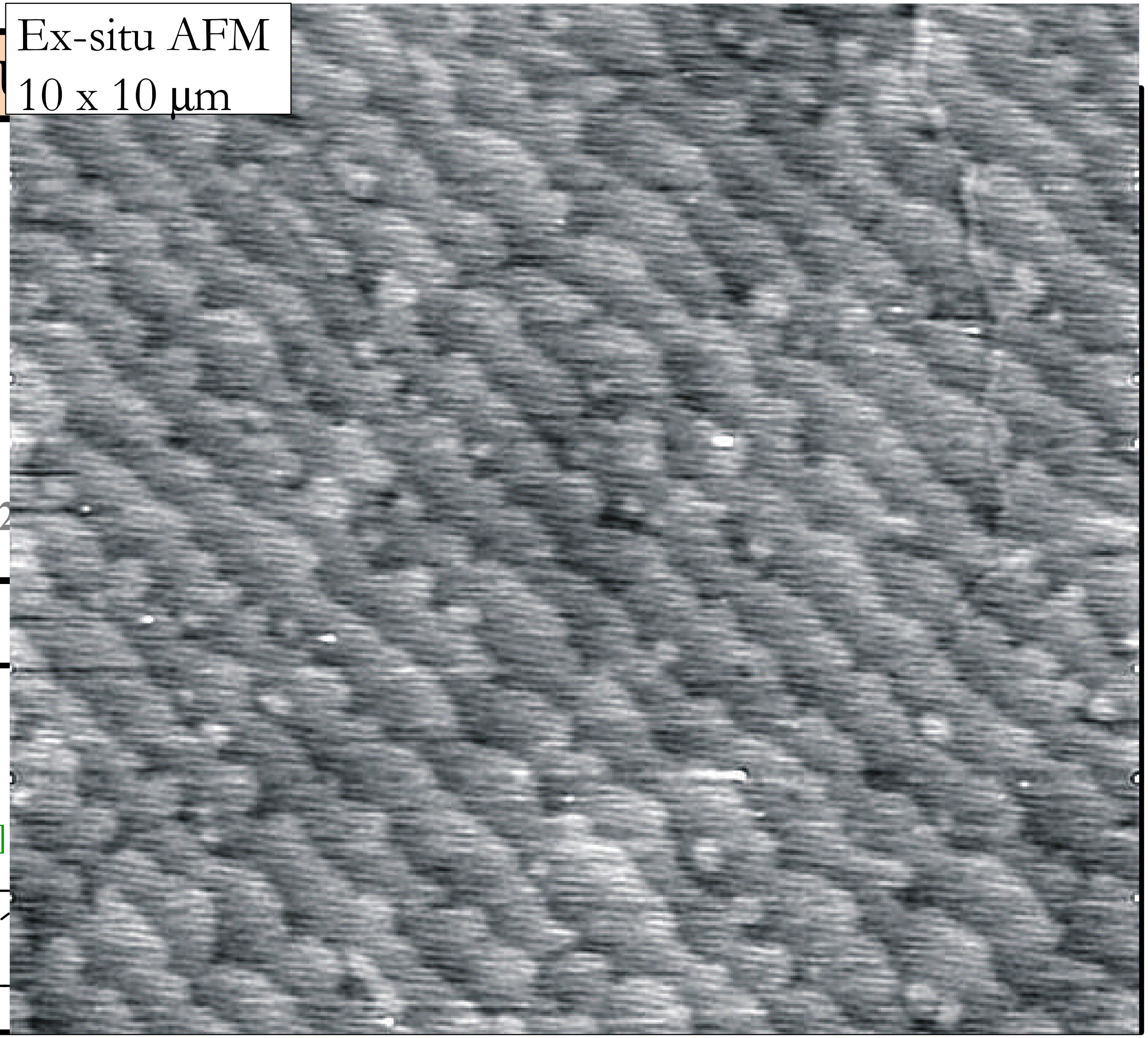


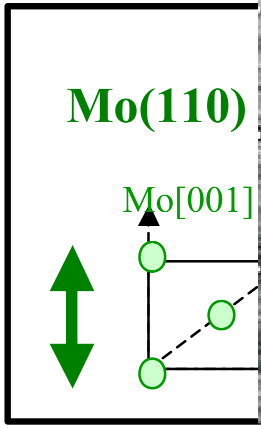
P.-O.Jubert *et al.*, JMMM 226, 1842 (2002)

P.-O.Jubert *et al.*, PRB64, 115419 (2002)

↳ **Is there an intermediate world ?**





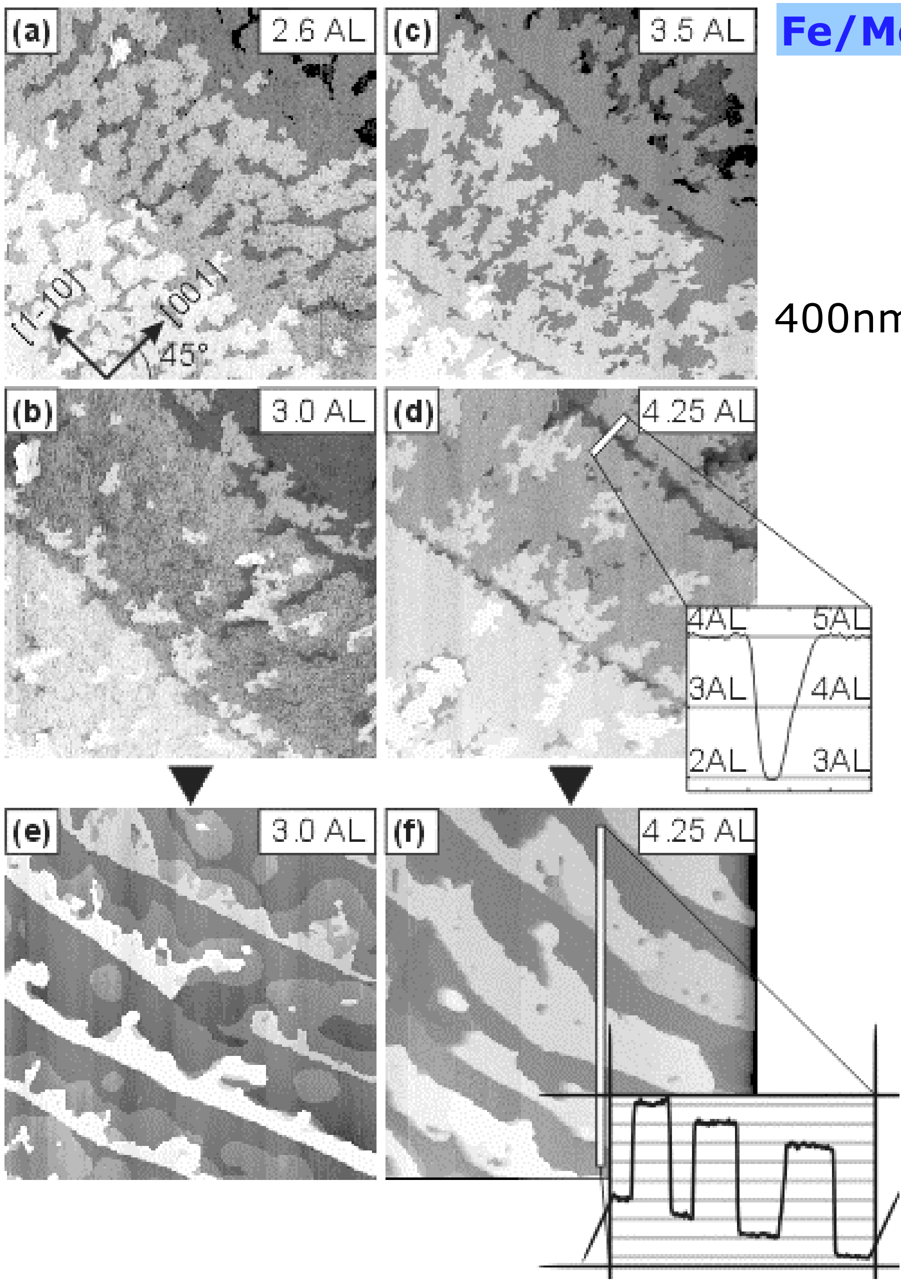


Buffer layer growth : **O. Fruchart, S. Jaren, J. Rothman, Appl. Surf. Sci. 135, 218 (1998)**

Fe/Mo(110)

150°C deposition

450°C annealing



400nm

400nm

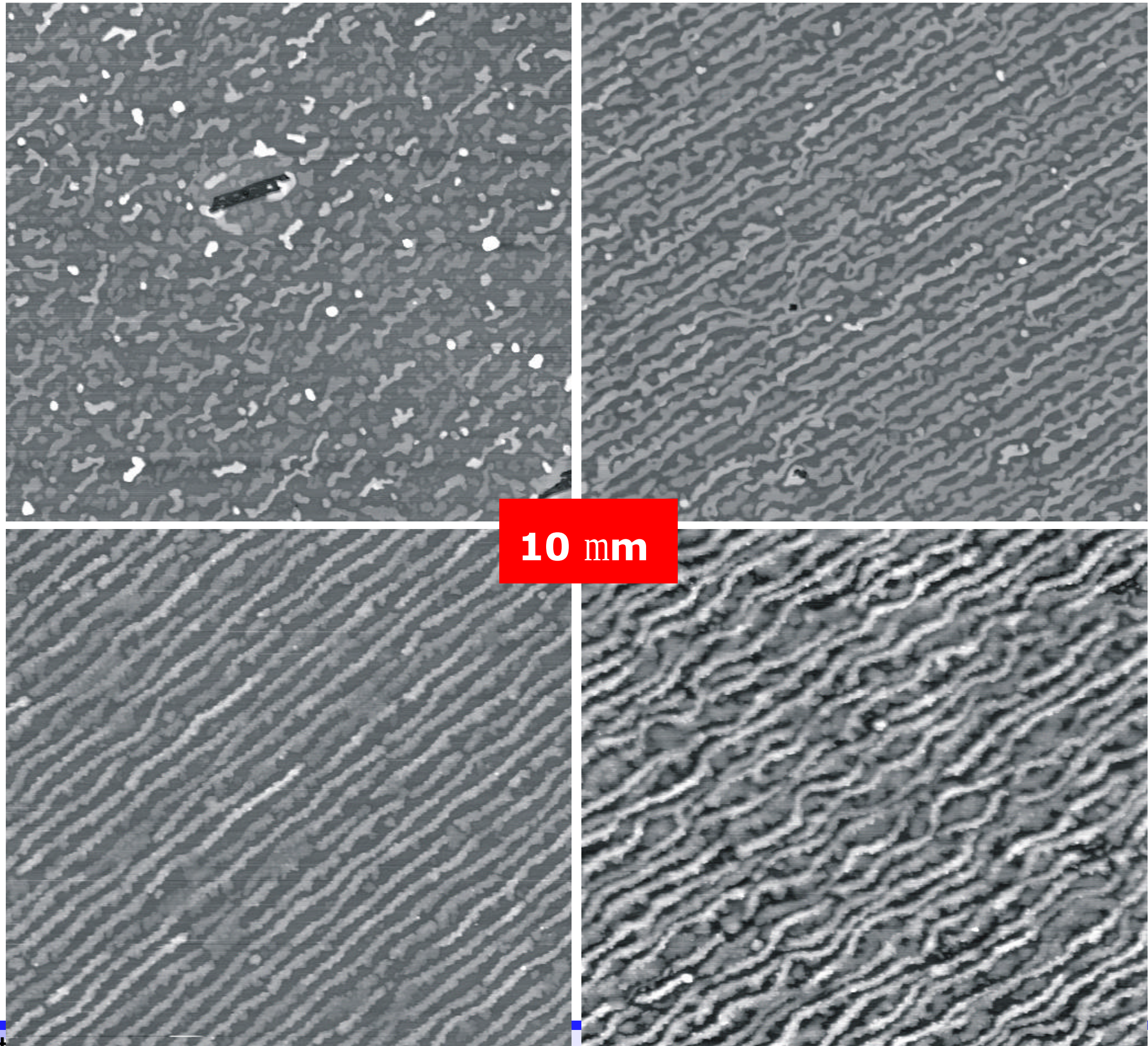
800nm

Height : 1.2nm

Sapphire\Mo(8nm)\interface\Fe(150°C, 450°C annealing)

Average thickness: 1nm | 2.5nm

Chemical interface: W(<1nm) | Mo(raw)

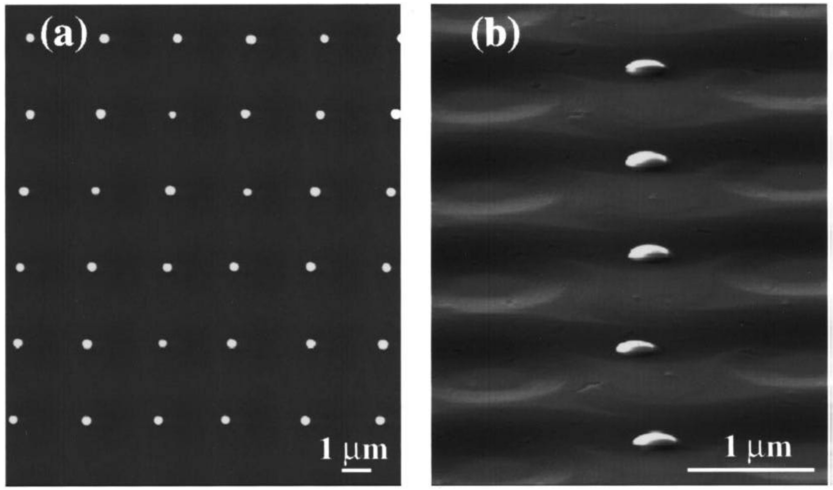


Height : 5nm

Height : bimodal 1.5/4nm

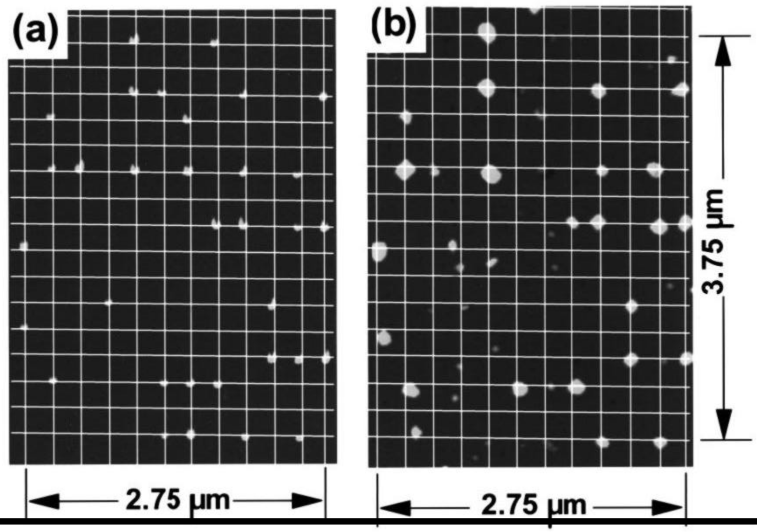
- Lithography + Etching + Annealing → holes

Au/Si



Y.Homma, Appl.Phys.Lett. **74**(6), 815 (1999)
T.Ogino, Surf.Sci.386, 137 (1997)

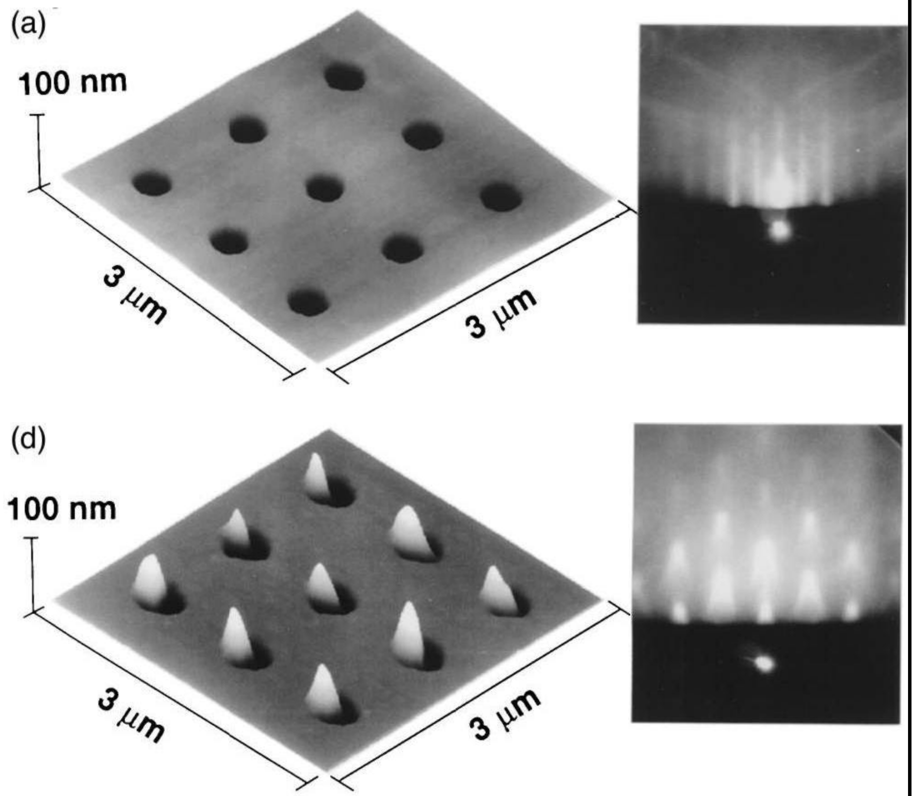
- Nanoimprint → Si mesas → Ge / Si



T.I.Kamins, Appl.Phys.Lett. **74**(12), 1773 (1999)

- In-situ lithography on oxide layer

InAs / GaAs

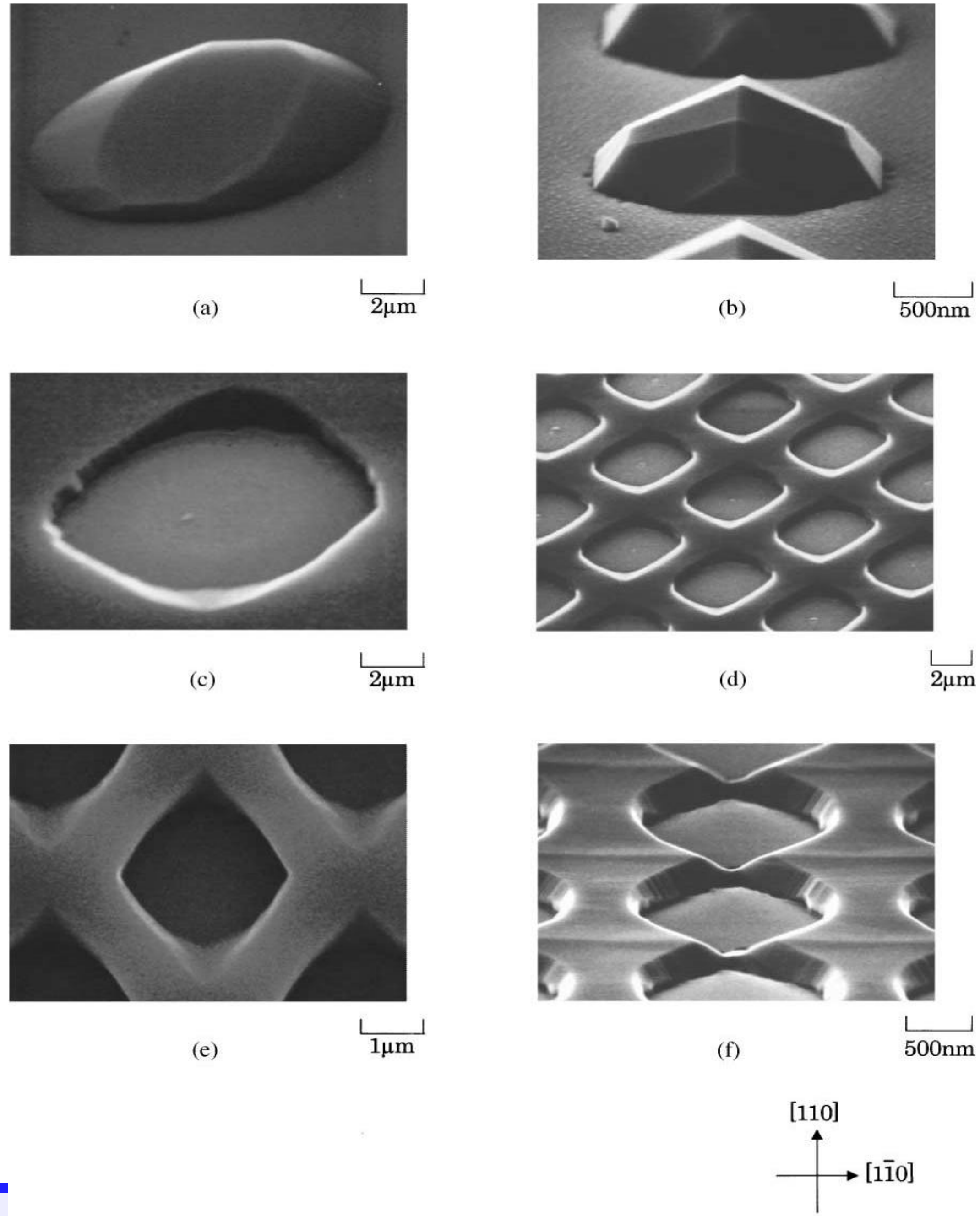


T.Ishikawa, Appl.Phys.Lett. **73**(12), 1712 (1998)

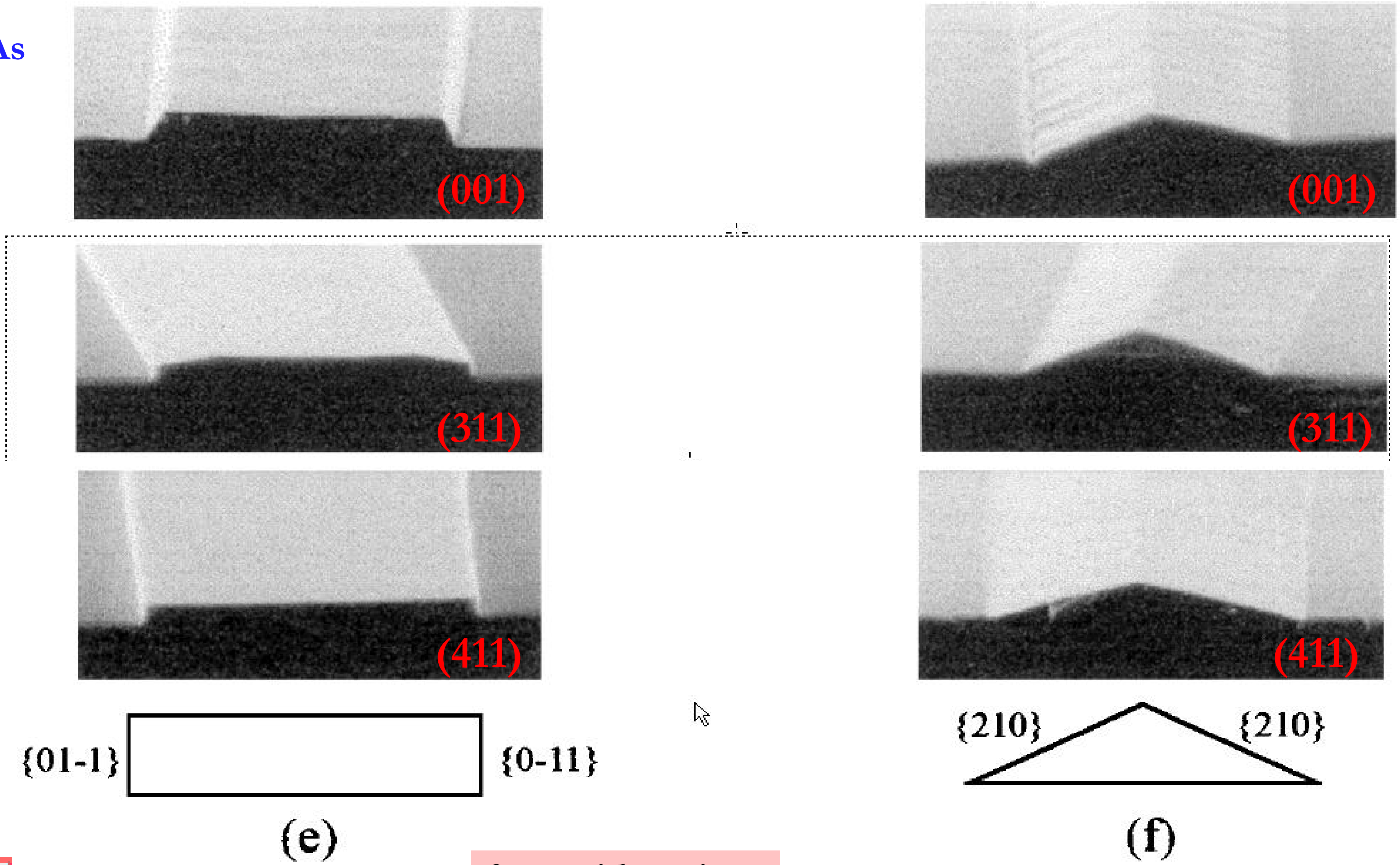
GaAs / GaAs

- **Step 1**: Deposition of SiO_2 layer
- **Step 2**: Ex-situ lithography on oxide layer, plus wet chemical etching
- **Step 3**: Growth of GaAs through windows, either in dot or antidot array.

H. Hasegawa, J. Cryst. Growth 227-228, 1078 (2001)



GaAs / GaAs



3mm wide stripes

H. Hasegawa, J. Cryst. Growth 227-228, 1078 (2001)

ELSEVIER

Surface Science 432 (1999) 37–53

surface science

www.elsevier.nl/locate/susc

How to control the self-organization of nanoparticles by bonded thin layers

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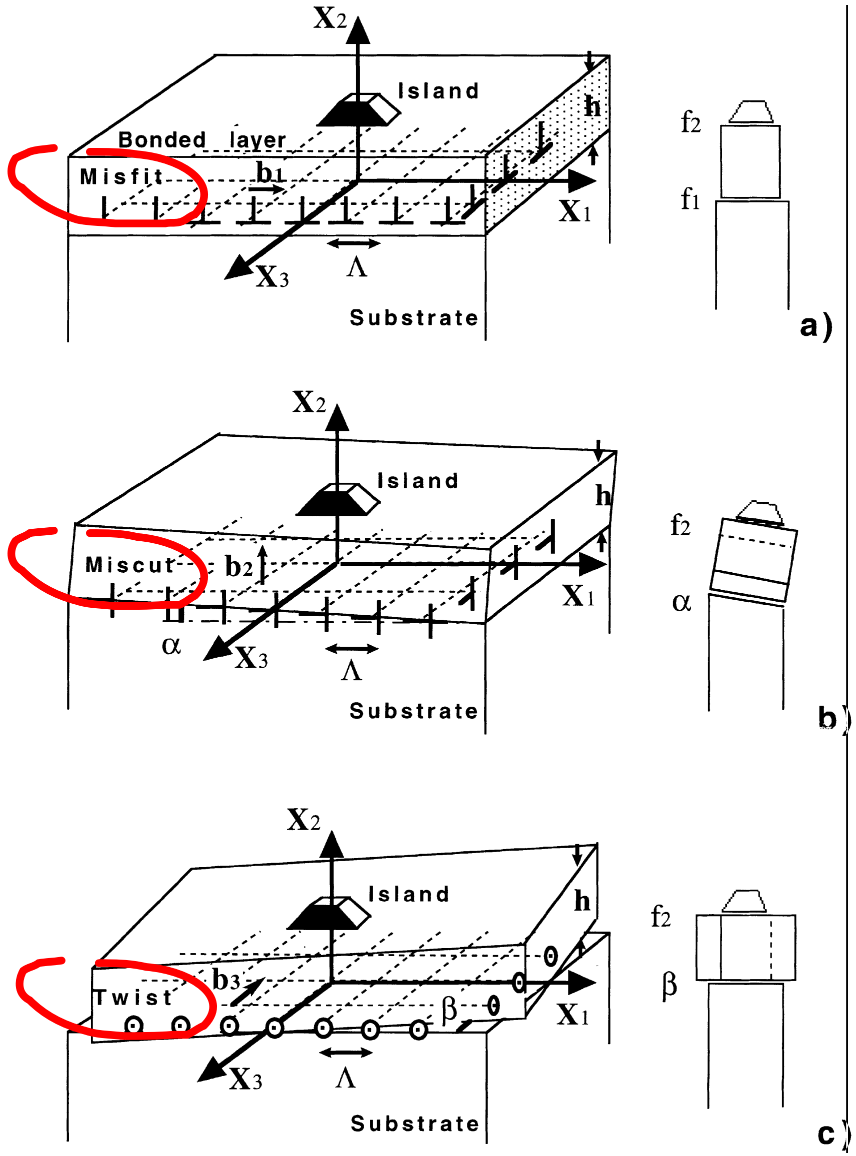
Département de Recherche Fondamentale sur la Matière Condensée, SP2M[1], CEA Grenoble, 17 rue des Martyrs 38054, Grenoble Cedex 9, France

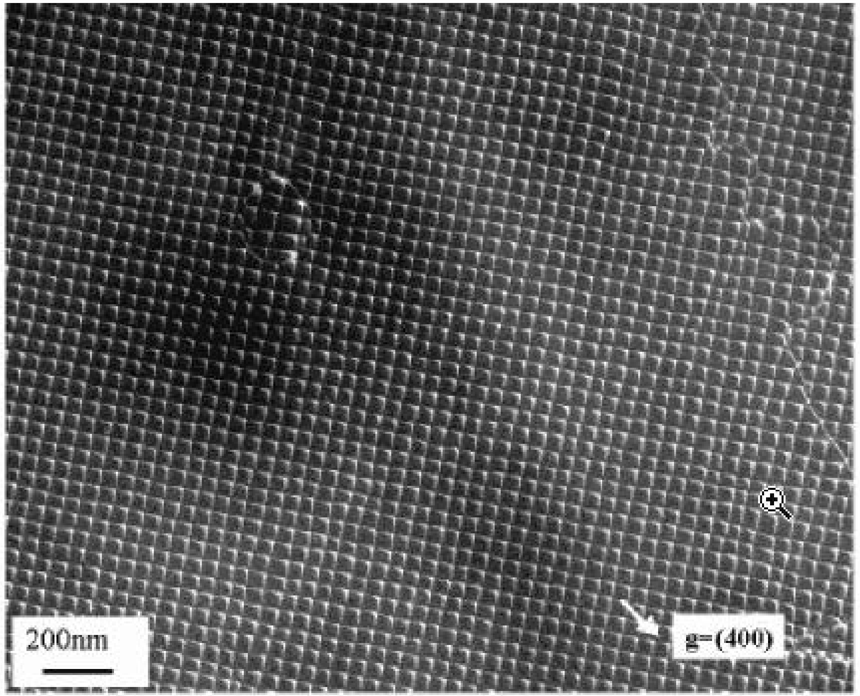
Received 21 December 1998; accepted for publication 6 April 1999



- Substrate bonding and smart cut process.
  - array of dislocations arise from either:
    - Misfit
    - Miscut
    - Twist

Step 1: wafer bonding

Screw dislocations

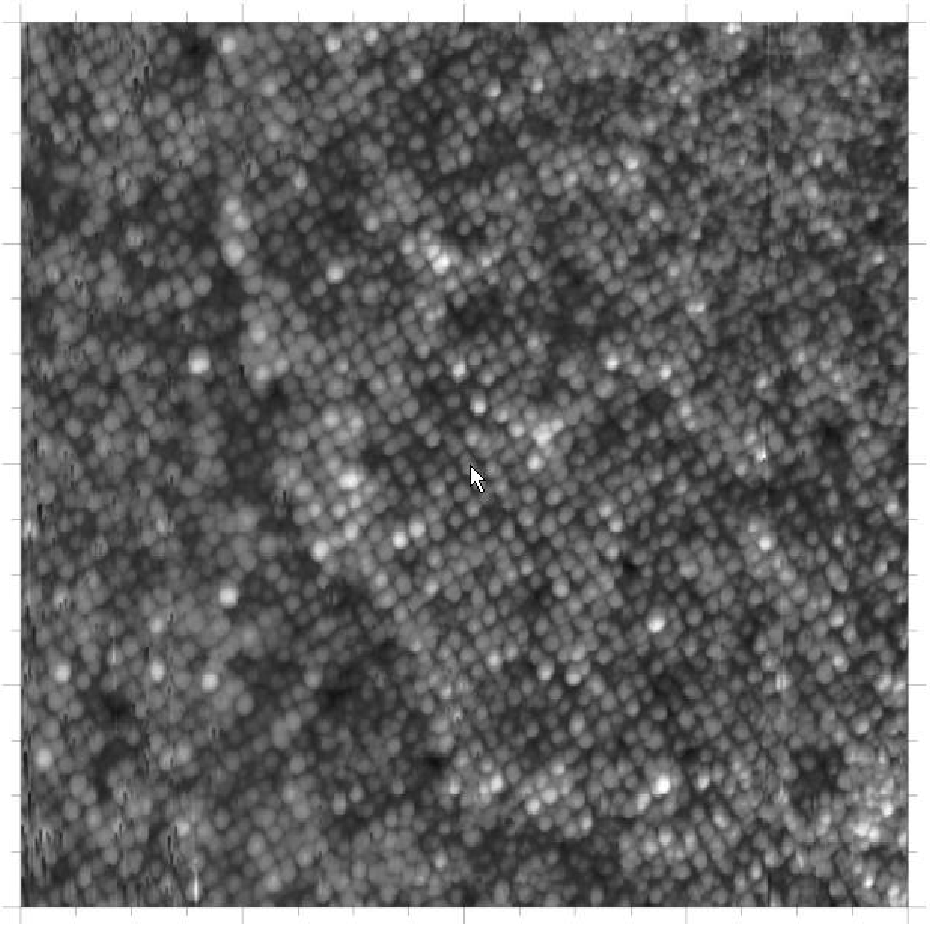


J. L. Rousseau *et al.*, APL80, 4121 (2002)

CEA-Grenoble

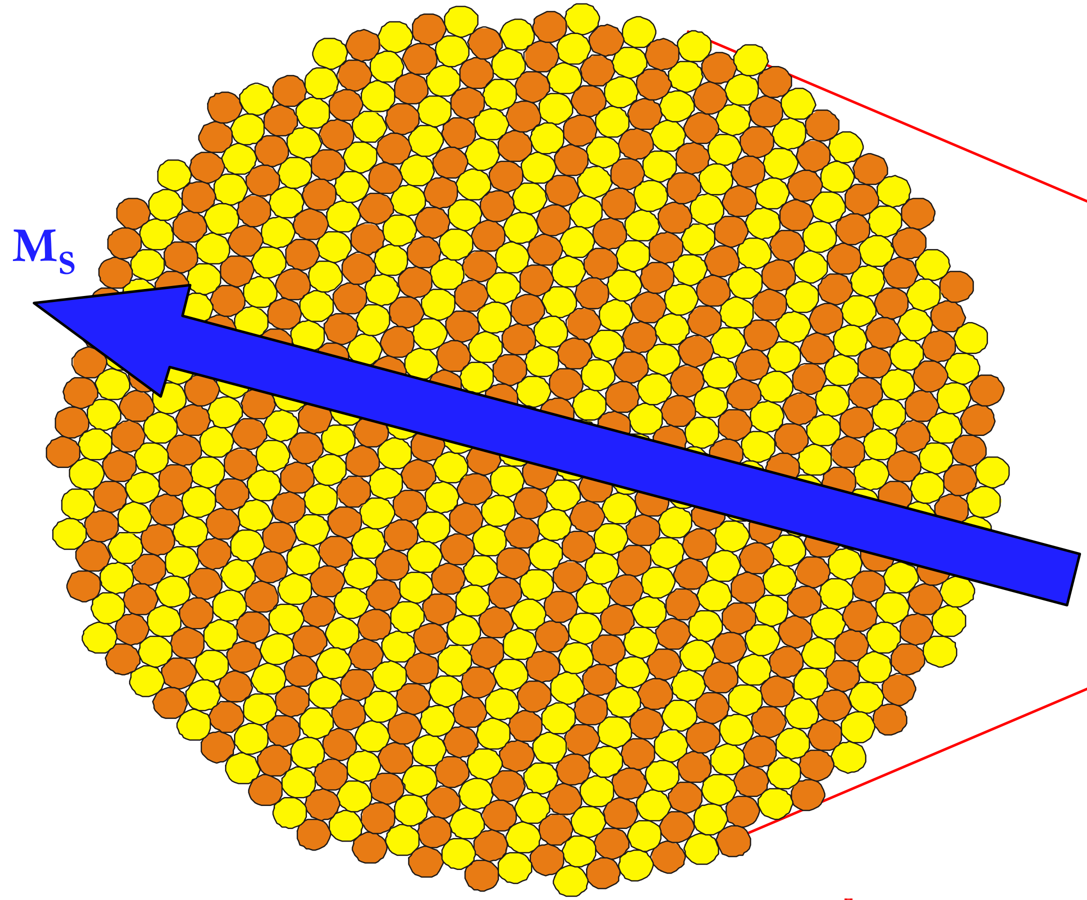
Step 2: template for growth

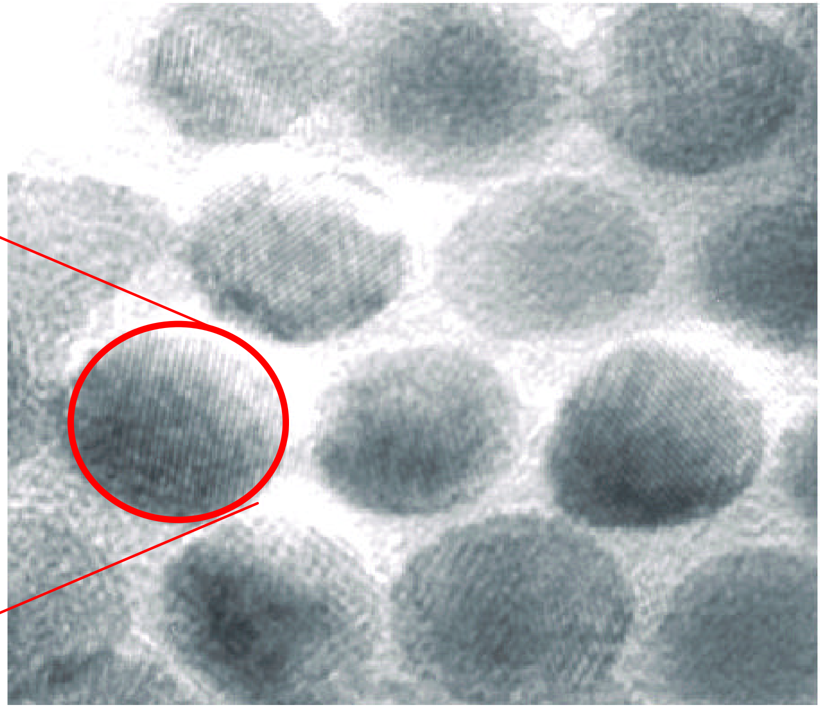
1. Chemical etching > corrugation enhanced
2. Growth (here: Si)



J. Eymery, Habilitation (2003)

$L1_0$ phase : alternation of Fe and Pt monoatomic planes
↪ extremely high magnetocristalline anisotropy K





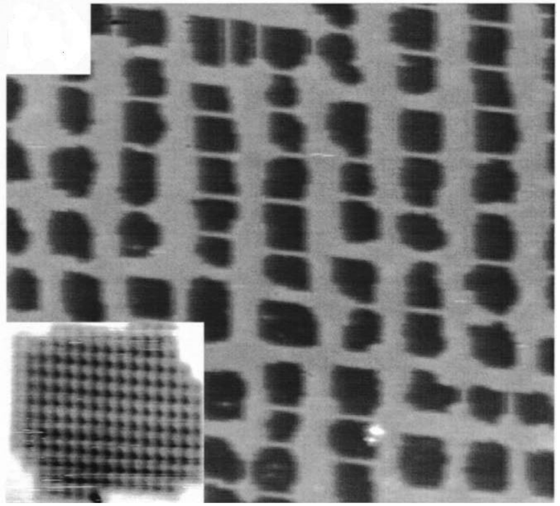
Random orientation of anisotropy axes

S. Sun, Science 287, 1989 (2000)

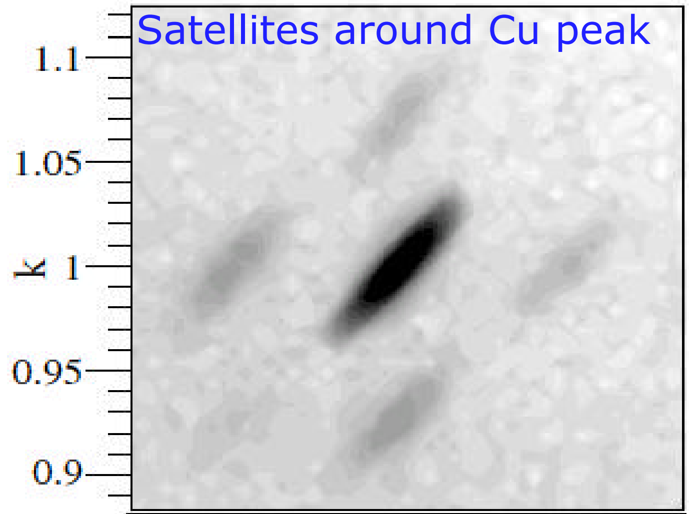
(Image: courtesy of D. Weller – Seagate via N. Dempsey – LLN/CNRS)

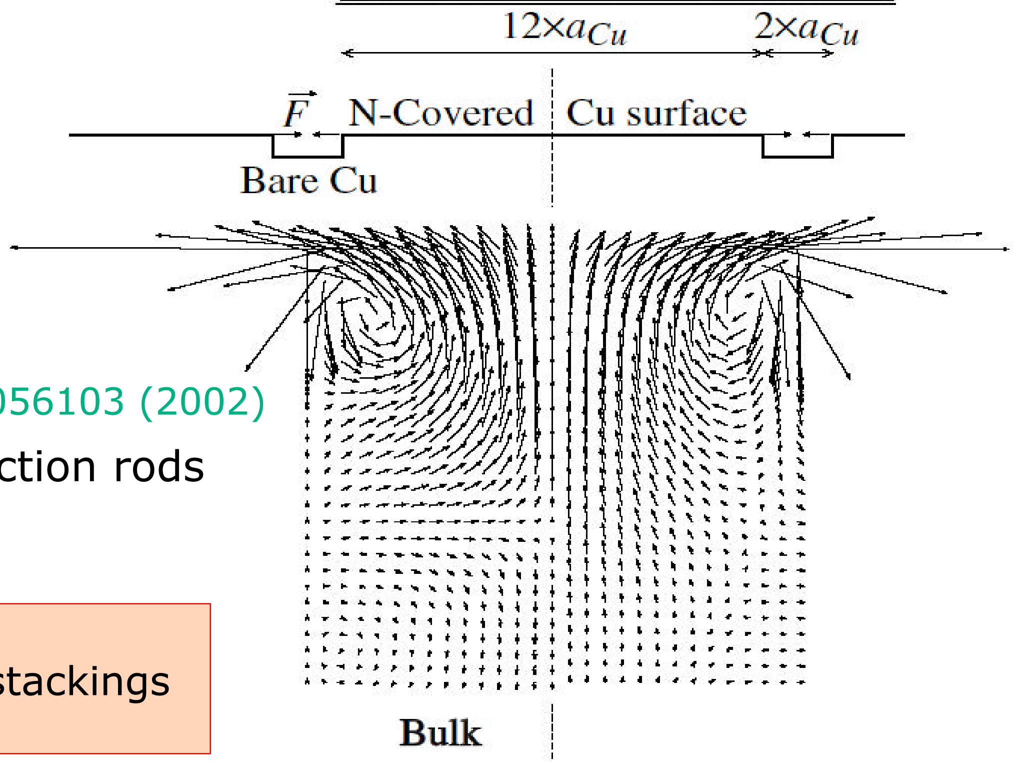
SXRD: self-organized N/Cu(001)

0.2AL N/Cu(001)



S.L. Silva, APL76, 1128 (2000)
F.M. Leibsle, SS317, 309 (1994)





B. Croset, PRL88, 056103 (2002)

- Periodic surface strain give rise to satellite diffraction rods

Other groups:
T. Metzger (ESRF, Grenoble): Quantum dots stackings
J. Eymery (CEA Grenoble): wafer bonding

Collaborators

G. Renaud, M. Noblet, O. Ulrich
DRFMC/SP2M/IRS (CEA), Grenoble

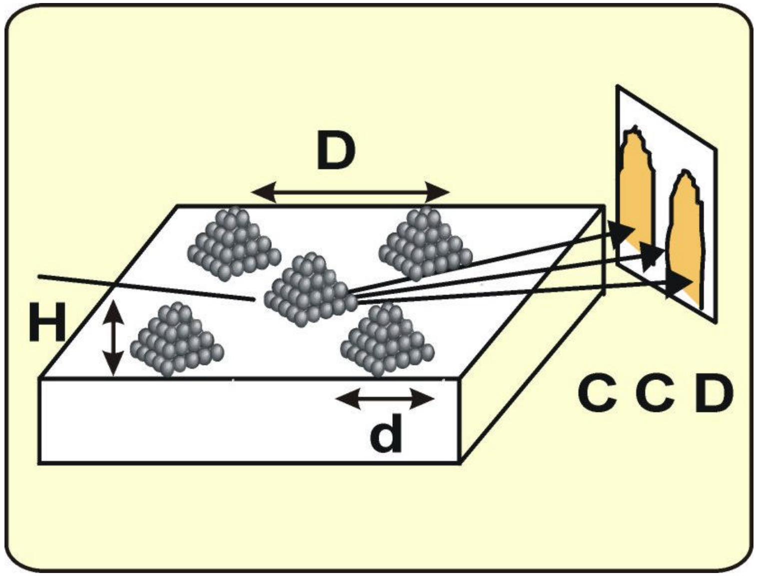
J.-P. Deville, A. Barbier, F. Scheurer, J. Mané-Mané
IPCMS (CNRS/ULP/ECPM), Strasbourg

V. Repain, G. Baudot, S. Rousset
GPS-Jussieu, Paris

GISAXS: Grazing Incidence Small Angle X-ray Scattering

- Small Angle → **Objects >> atoms**
- Grazing Incidence → **Surface sensitive**

PRINCIPLE



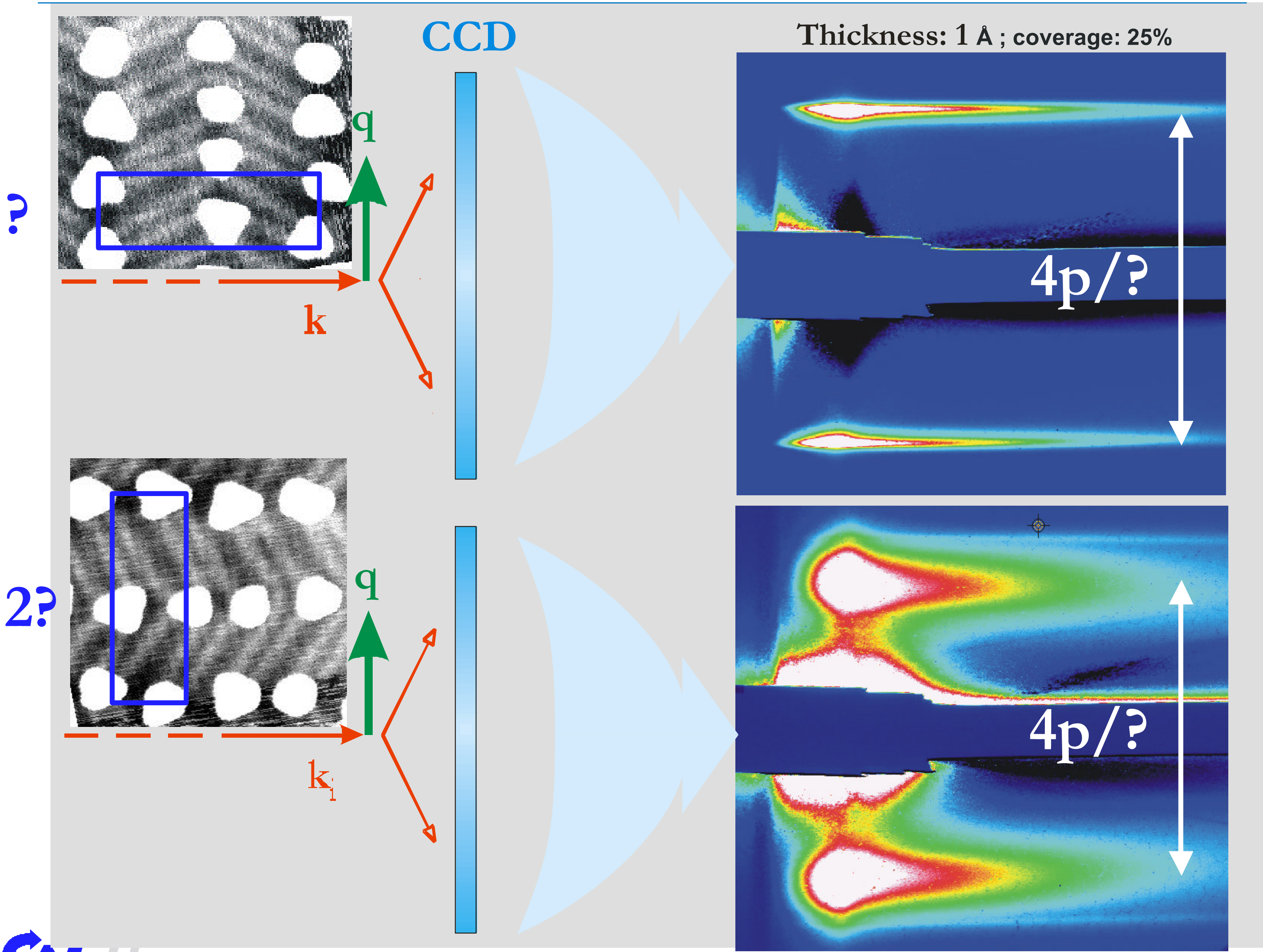
- In-plane / out-of-plane diffusion
- Probes size (H,d), shape, correlations

TECHNICAL ASPECTS

- ID32 beamline at ESRF
- In-situ under UHV:
  - real time
  - background substraction
- No window before sample : low background

G. Renaud et al., Science 300, 1416 (2003)

O. Fruchart et al., Europhys. Lett. 63, 275 (2003)

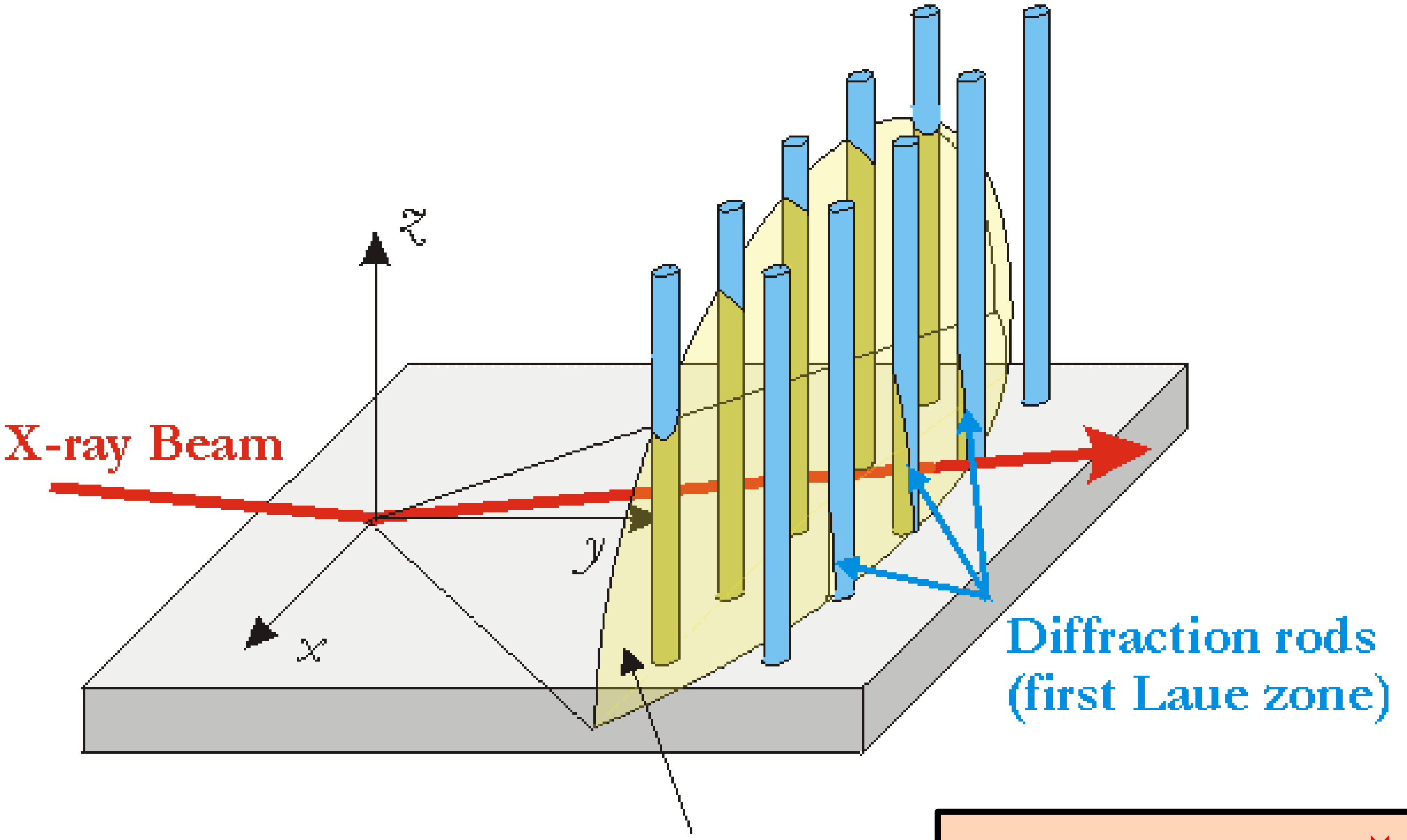


INTRA-ROW ORDER: SUPER-CRYSTAL

DQ/Q=3%

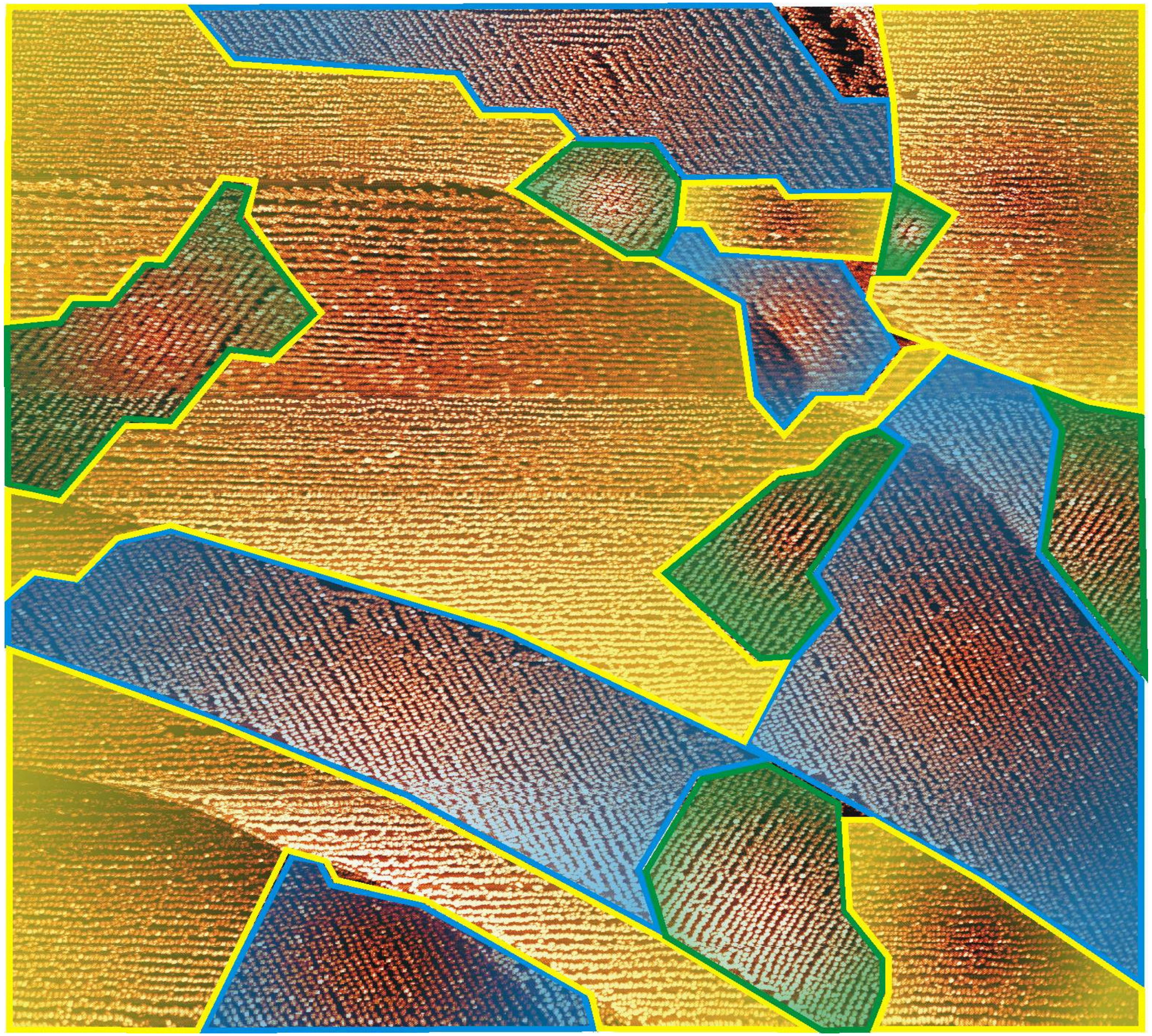
INTER-ROW ORDER: LIQUIDE-TYPE

K=8.5nm
s =2.1nm



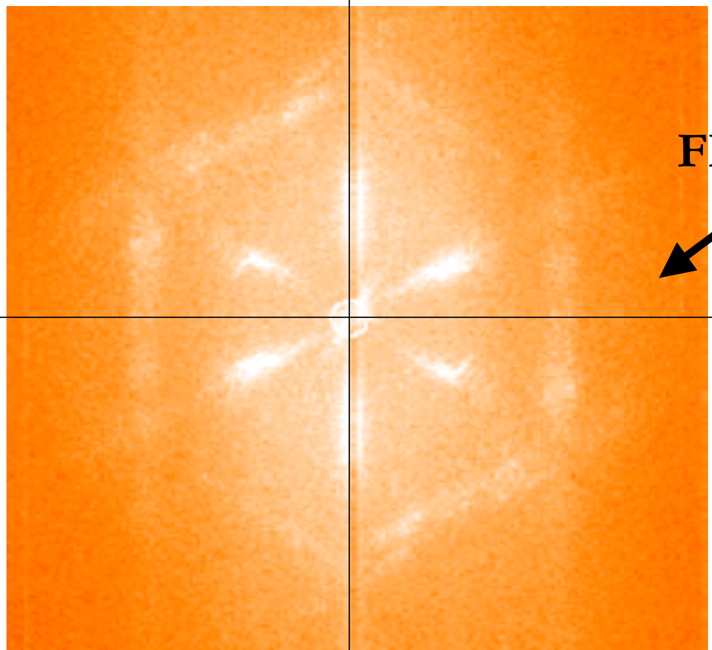
- Ordered sample ↳ 'super' reciprocal space ?
- Grazing incidence and 2D detector
  ↳ similarity with RHEED

1.5mm x 1.5mm STM image

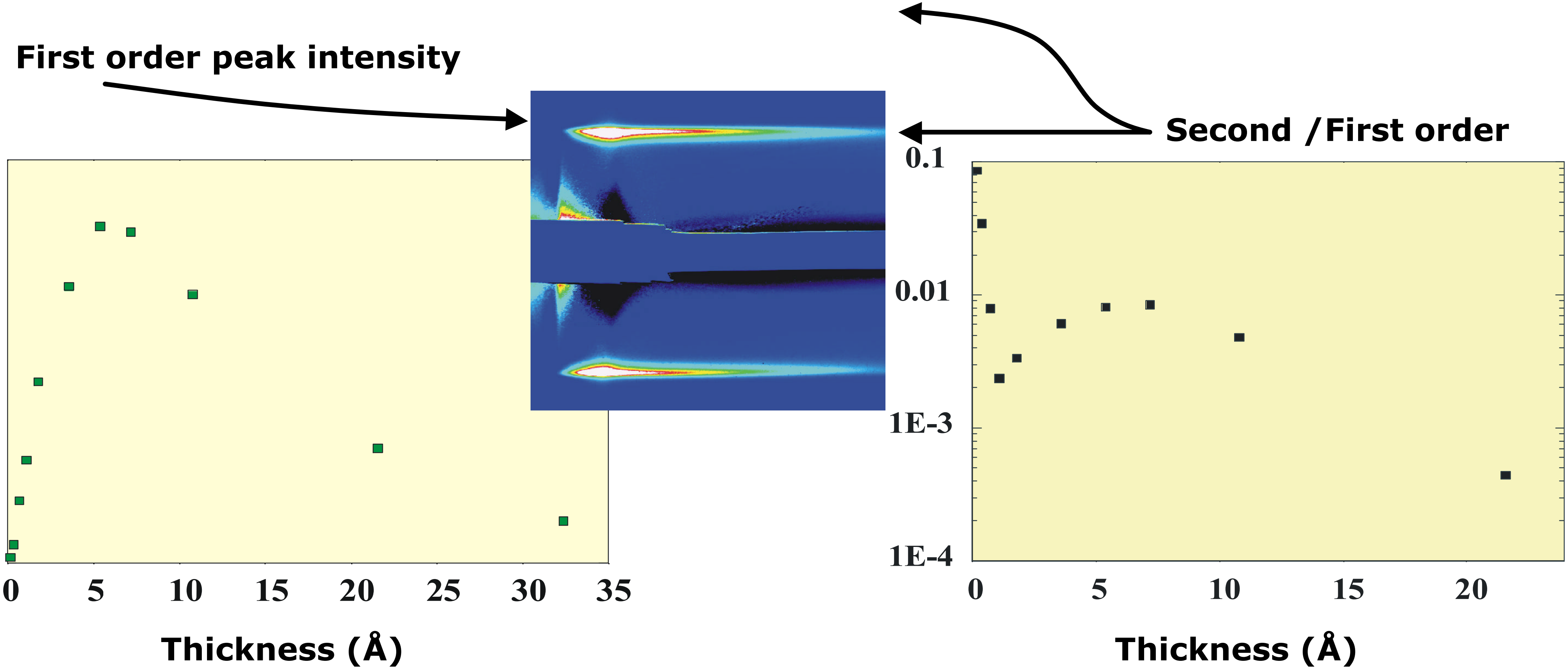


3-fold symmetry of Au(111) crystal

- 3 'equivalent domains'



FFT



Features to explain

- Maximum at 6Å = 3 atomic layers ?
- Significant intensity for continuous film ?
- Weak intensity ?
- Intermediate minimum ?

RATIO: Second /First order peak intensity

- Deposition rate calibration

Simple model of 1D coalescence