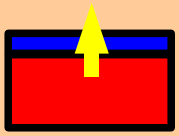
Controlling magnetism by stacking individual atomic monolayers of magnetic- and non-magnetic materials

Juan de la Figuera

http://hobbes.fmc.uam.es/loma

The easy axis of magnetization depends on the shape

- In a needle, the magnetization points along the needle axis
- In a thin film, the same effect tries to drive the magnetization IN PLANE
  - But in some special cases, very thin films present an out-of-plane magnetization
- Recording media traditionally uses in-plane magnetization in thin films
  - The discovery of **Perpendicular Magnetic Anisotropy (PMA)** opened the possibility of higher recording densities

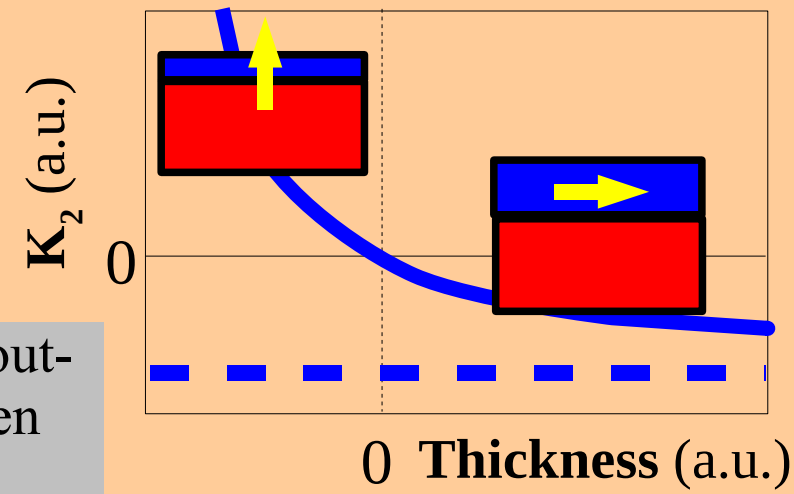


Easy axis of magnetization in a thin film ferromagnetic sample

- The Perpendicular Magnetic Anisotropy (PMA) can be due to different reasons: surface effects, magnetoelastic contributions, etc.

- In systems with PMA, often we observe:

Spin-reorientation transitions (SRT)



Usually: transition from out-of-plane to in-plane when increasing thickness

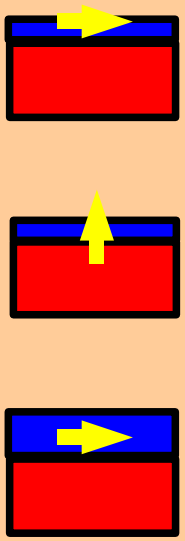
Magnetic Anisotropy Energy (MAE): Volume + Surface contrib.

$$\frac{E_a}{V} = -K_{2\mathrm{Eff}} \cos^2\theta = -\left(-\frac{1}{2}\mu_0 M_s^2 + K_{b,2} + K_{s,2}/d\right)\cos^2\theta$$

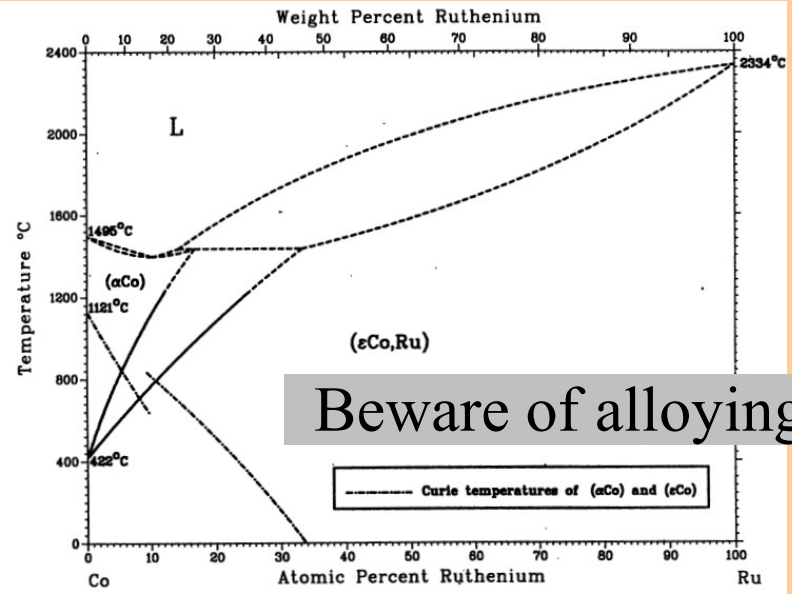
M. T. Johnson et al, *Rep. Prog. Phys.* **59** (1996) 1409

Some systems present several SRTs

- Ex. Ni/Cu(001):
  - in-plane to out-of-plane at ~12 ML
  - out-of-plane to in-plane at ~50 ML
- These transitions are driven by a combination of magnetoelastic effects associated with structural transformations and interface effects

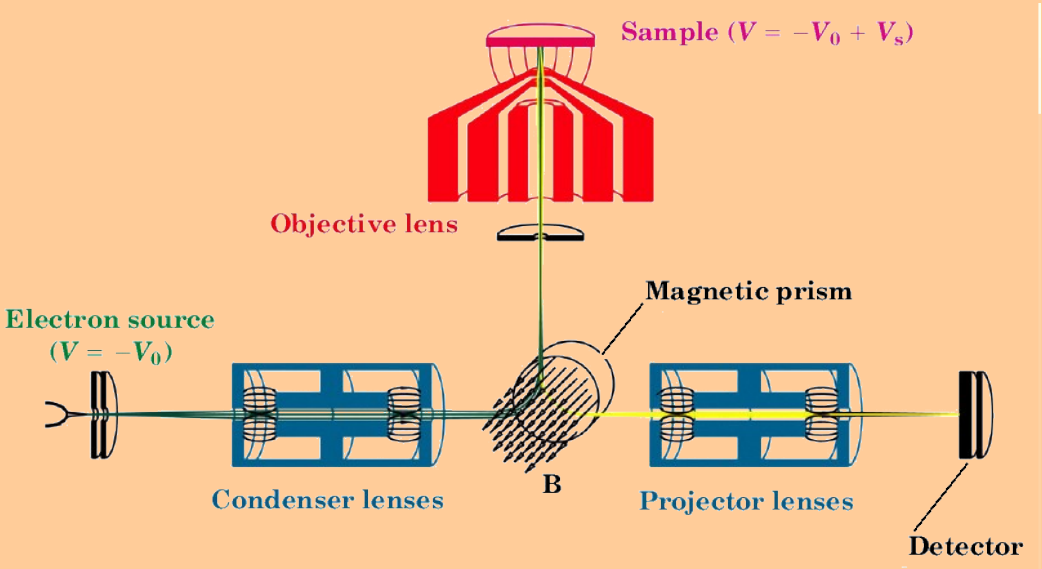


- Many studies of Co thin films
  - Expect PMA for small thickness
  - Co/Ru(0001):
    - Bader's group, Ounadjela's group
    - in-plane for < 4 ML



Beware of alloying!!

Low Energy Electron Microscope



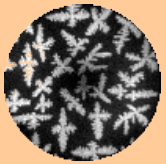
Collimated electron beam (or monochromatic light source) used for illumination.

Reflected (or photoemitted) electrons used for imaging

Complications: magnetic prism, immersion objective lens

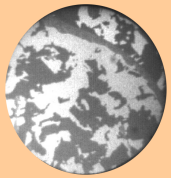
Imaging modes

Phase Contrast



PEEM

LEED

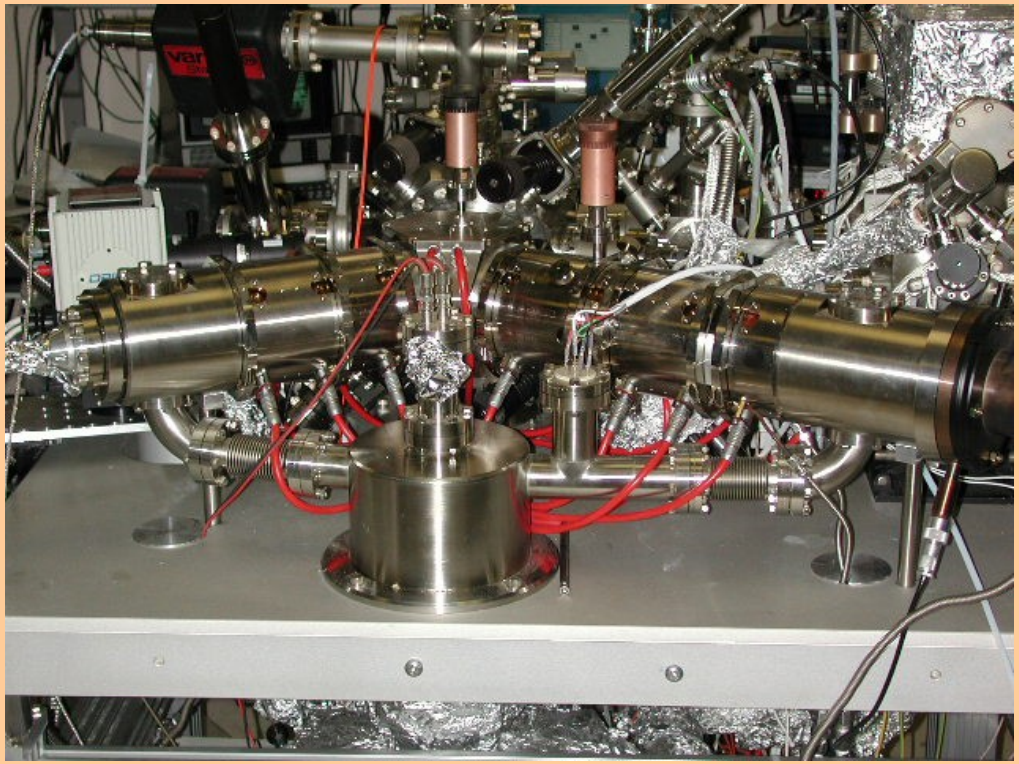


Dark/Bright field imaging

E. Bauer, *Low energy electron microscopy*, Rep. Prog. Phys. **57** (1994) 895
R.M. Tromp, *Low Energy electron microscopy*, IBM J. of Res. Dev. **44** (2000) 503

LEEM:Collaboration with Sandia National Laboratories

Commercial LEEM system: ELMITEC III



Resolution: 8 nm
30 frames/second

Main (joint) research lines:

- Stacking faults and grain evolution

F. El Gabaly, Wai Li W. Ling, K.F. McCarty, J. de la Figuera, *Science* **308** (2005) 1303

- Melting and condensing in 2D
- Dislocation networks in thin films
- Surface alloys

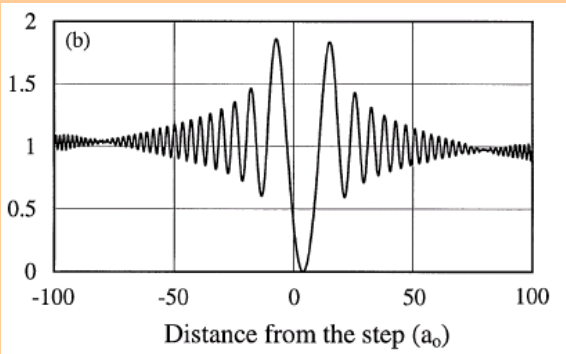
Sandia National Laboratories

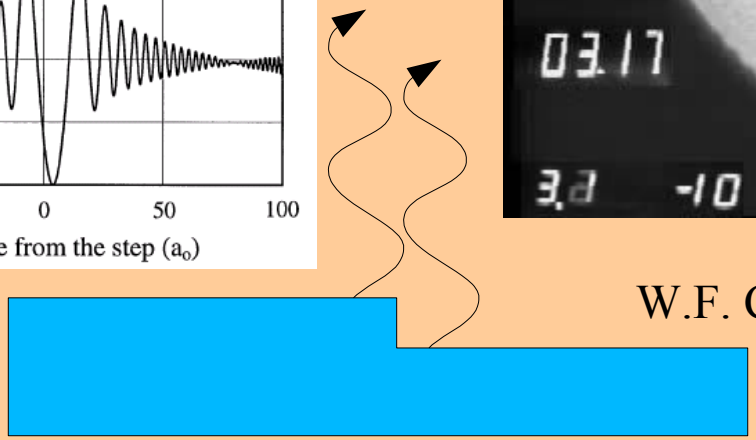
LEEM:
Real time observation of growth

2ML Co/Ru(0001)

10 μm FOV
200 ºC

Phase contrast allows for viewing atomic steps





W.F. Chung, M.S. Altman, *Ultramicroscopy* **74** (1998) 237

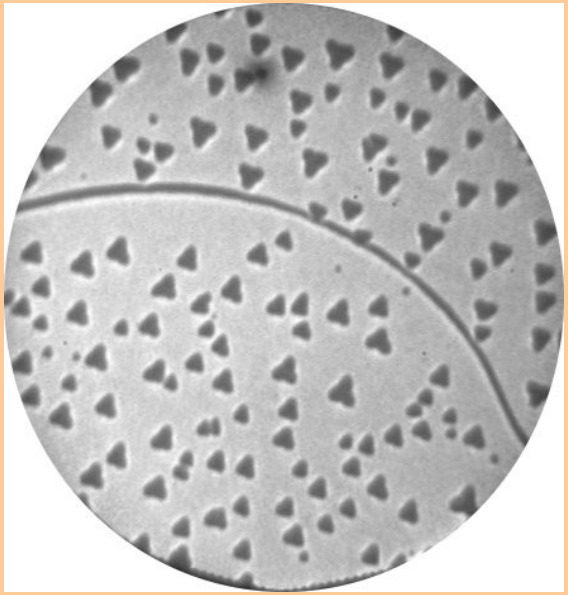
LEEM:
Real time observation of growth

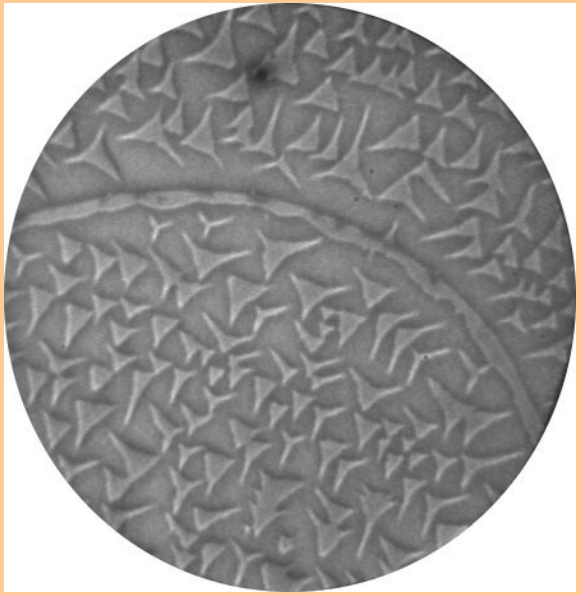
2ML Co/Ru(0001)

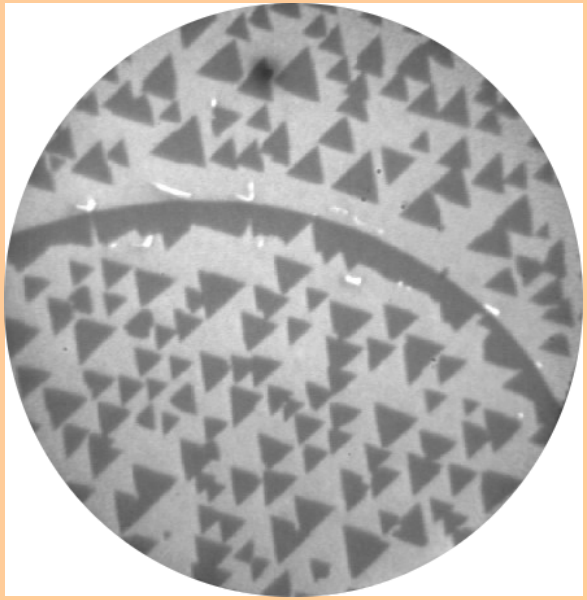
10 μm FOV
200 ºC

Surface can be observed while growth takes place!
(or while dosing gases, heating to 1900 K/cooling to 100K)

Growth of Co on “flat” Ru(0001)





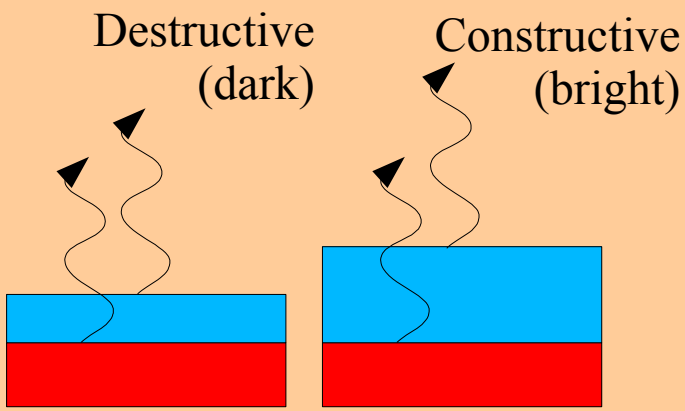


1 ML islands on Ru(0001)

200 ºC

2 ML islands on 1 ML Co/Ru

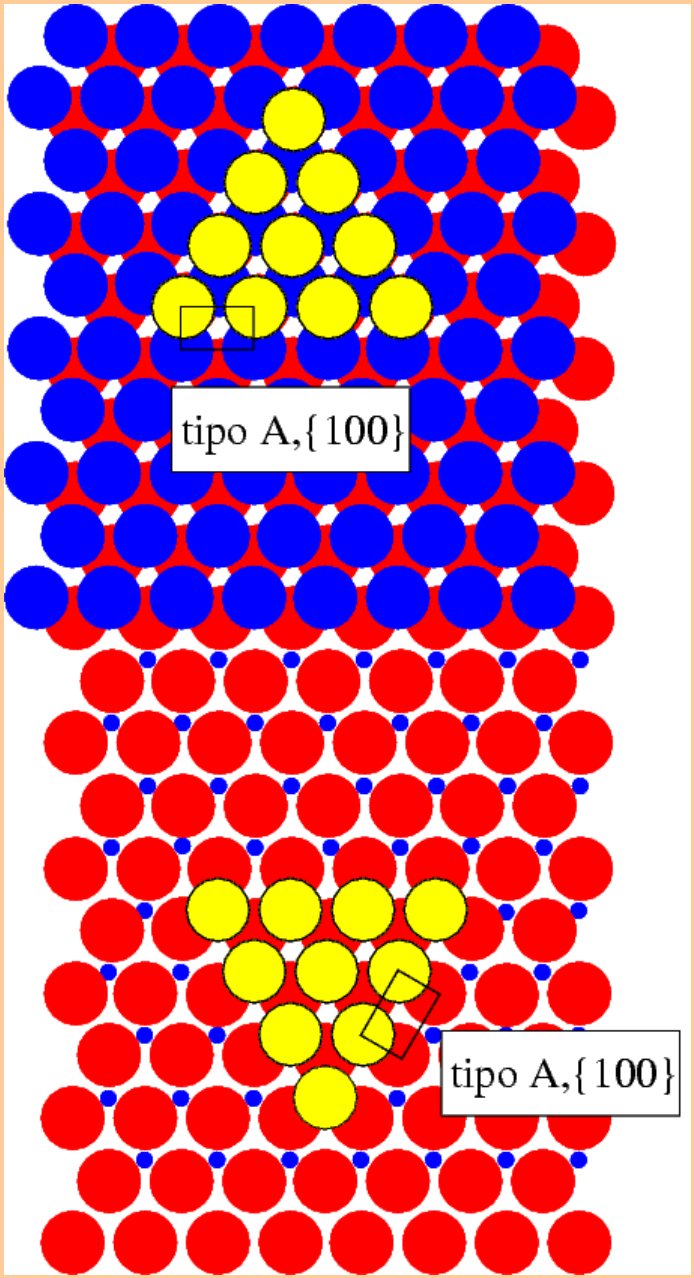
3 ML islands on 2 ML Co/Ru



Different contrast:
Quantum size effect in reflectivity

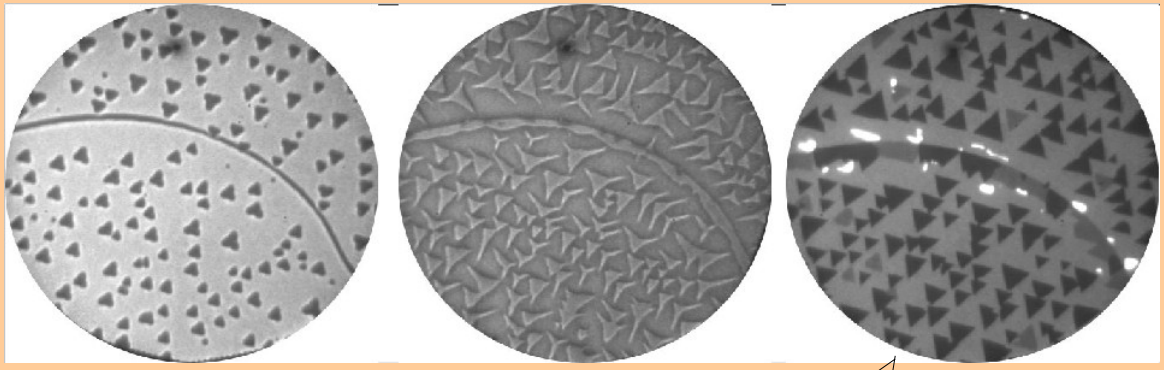
M.S. Altman, et al., *App. Surf. Sci.* **169-170** (2001) 82

Triangles indicate stacking sequence

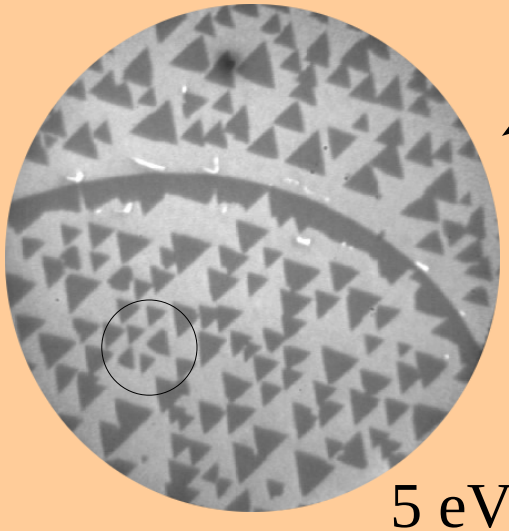


1 ML is hcp

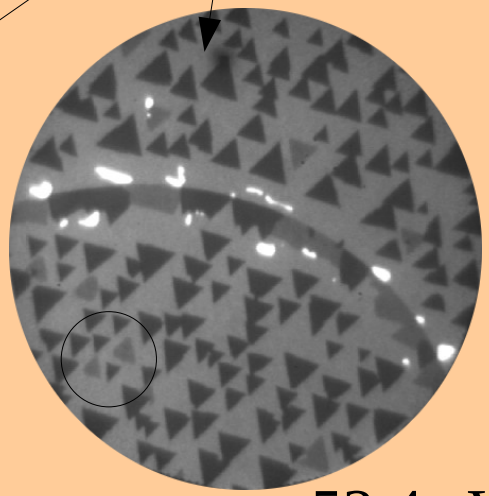
3 ML is fcc mostly!



2 ML is hcp also



5 eV



52.4 eV

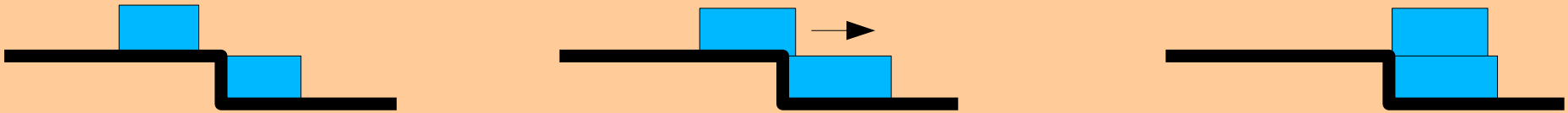
Change electron energy to see stacking faults!

Why layer by layer growth without steps?

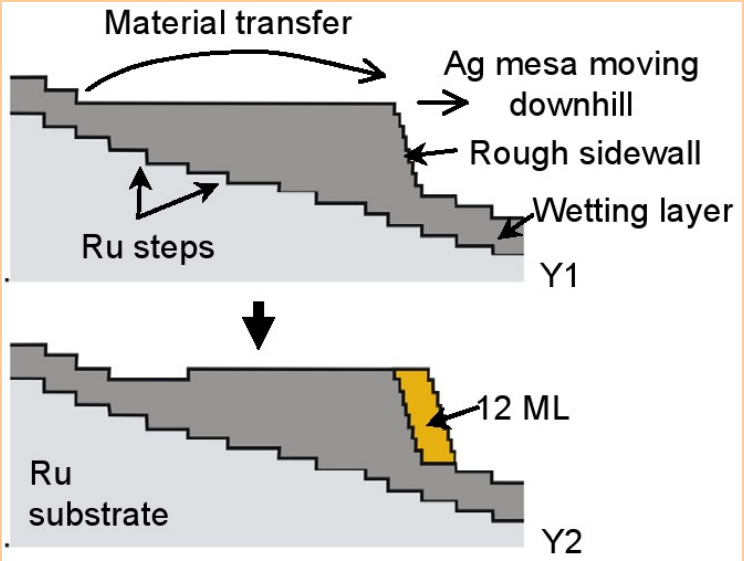
Thus, we conclude that Co on Ru(0001) has island growth after a non-ideal growth of the first layer.

C. Liu, S.D. Bader, *J. Mag. Mag. Mat.* **119** (1993) 81

Crucial role of substrate steps in de-wetting of crystalline thin films

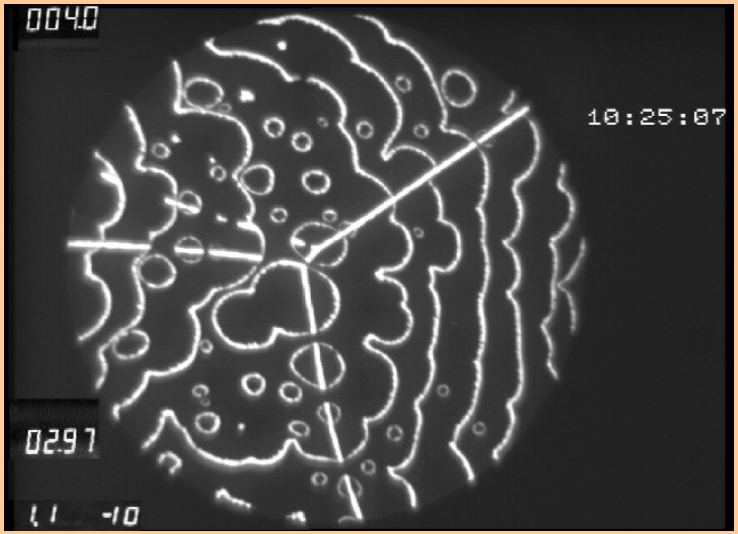


- Different samples:
  - Steps ARE important
  - Steps provide for the growth of higher layers islands *with no nucleation!*



W. L. Ling, T. Giessel , K. Thürmer, R. Q. Hwang, N. C. Bartelt, K. F. McCarty *Surf. Sci.* **570** (2004) L297

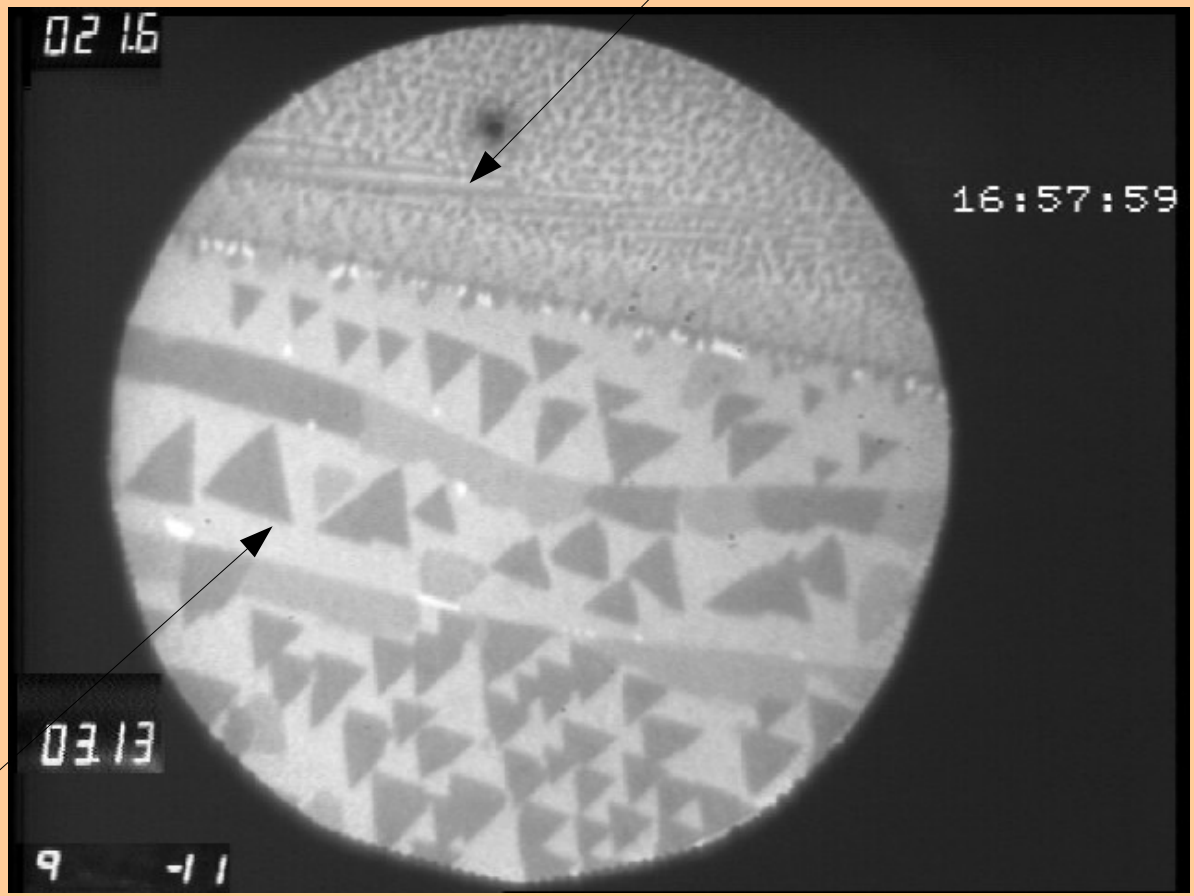
Growth mode depends on morphology



100 μm FOV in PEEM mode, some Ag decorating the Ru(0001) steps and grain boundaries

- Growth of 2.5 ML

3D growth on stepped areas

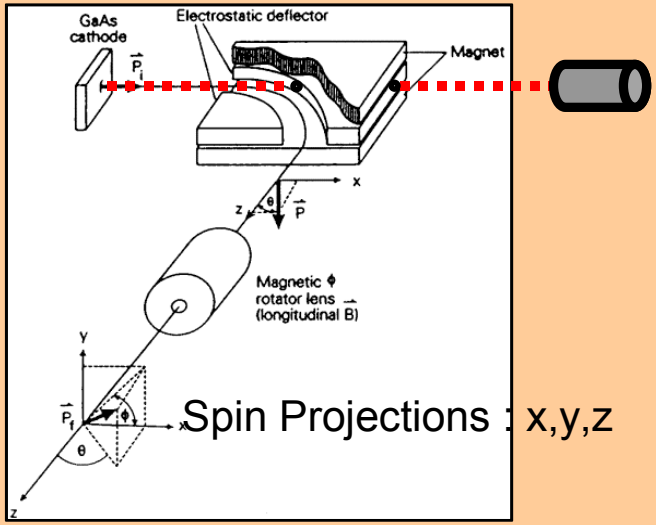


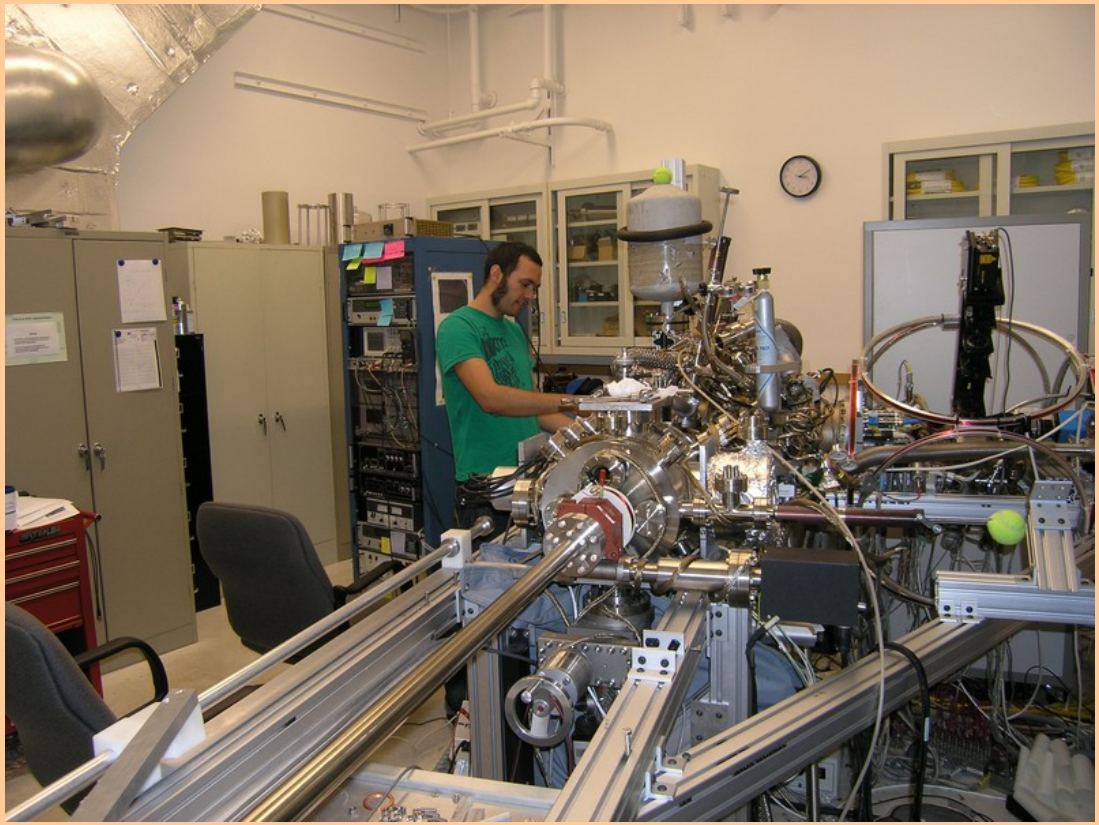
Layer-by-layer in flat areas

10 μm FOV

SPLEEM:Collaboration with Berkeley National Laboratory

Duden,T. Bauer,E.*Rev. Sci. Instrum.* **66** (1998) 2861





Only three systems in the world

Main (joint) research lines:

- Interaction between defects and magnetic films
- Surface Magnetic SRTs
- Surface Alloys

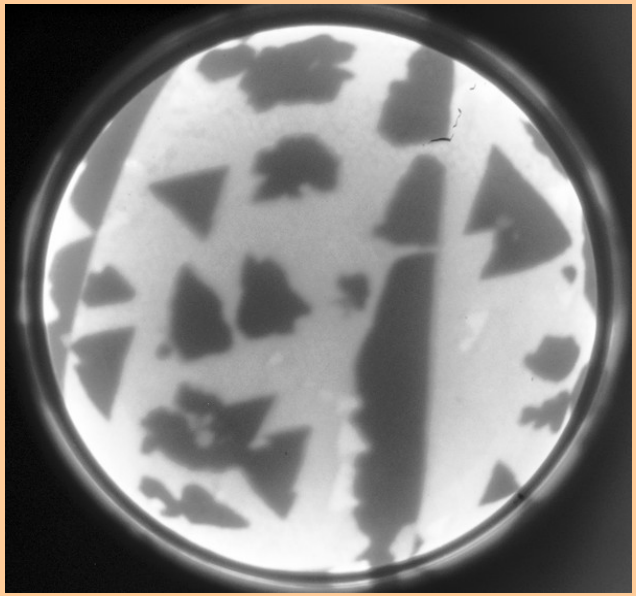
Christof Klein, Andreas K Schmid
National Center for Electron Microscopy

Observing magnetization with **SPLEEM**

- Contrast is due to spin-dependent exchange scattering:
  - **M:** orientation of the magnetization in the film
  - **P:** orientation in the beam polarization

Contrast∝**P·M**

Spin "up" direction
LEEM image



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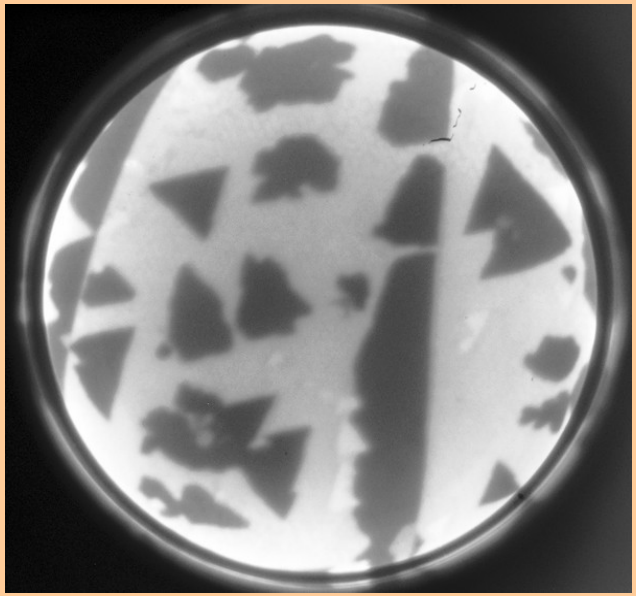
Spin "down" direction
LEEM image



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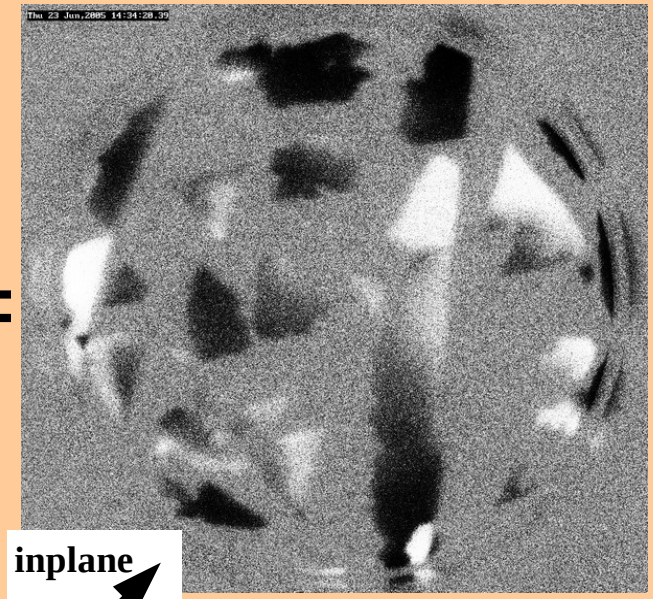
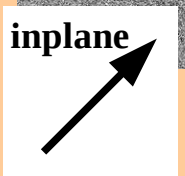
SPLEEM image
in-plane -64



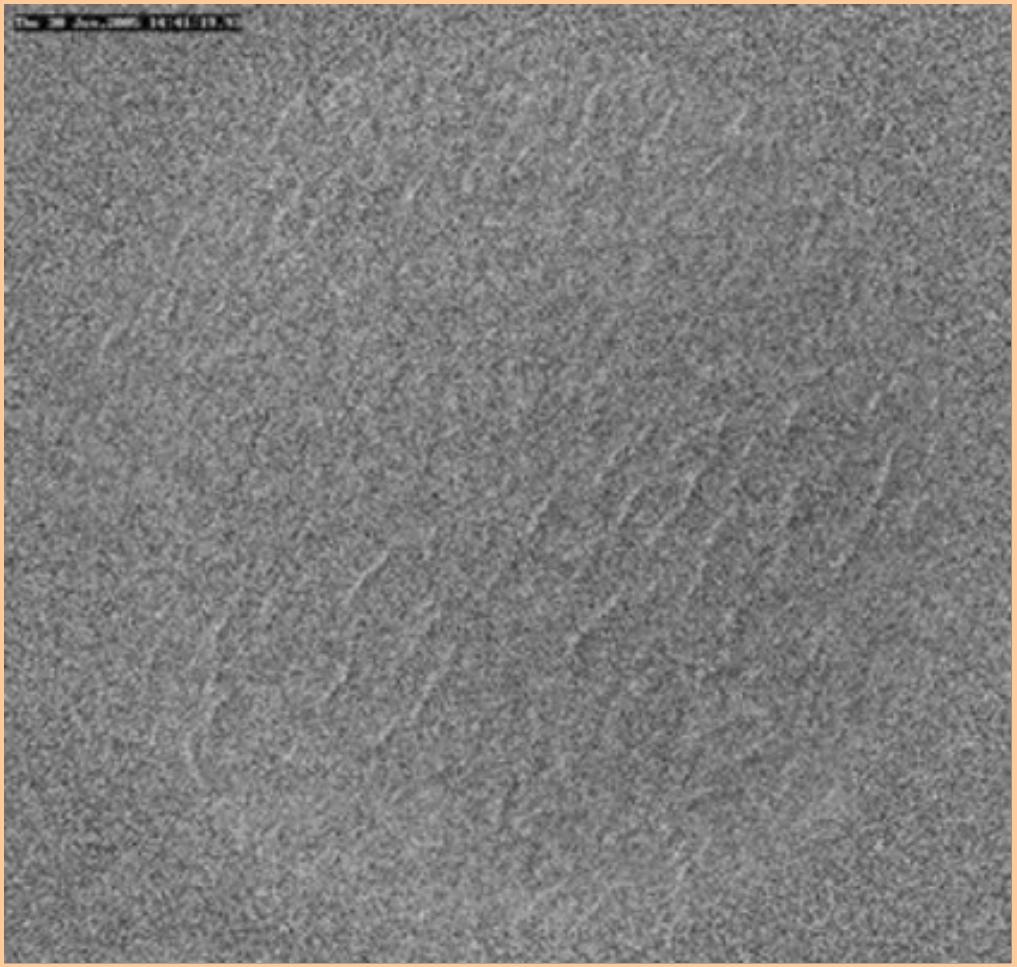


normalized

Growth of >3ML Co/Ru(0001)
(looking in-plane at RT)

T= 237ºC, FOV= 7µm

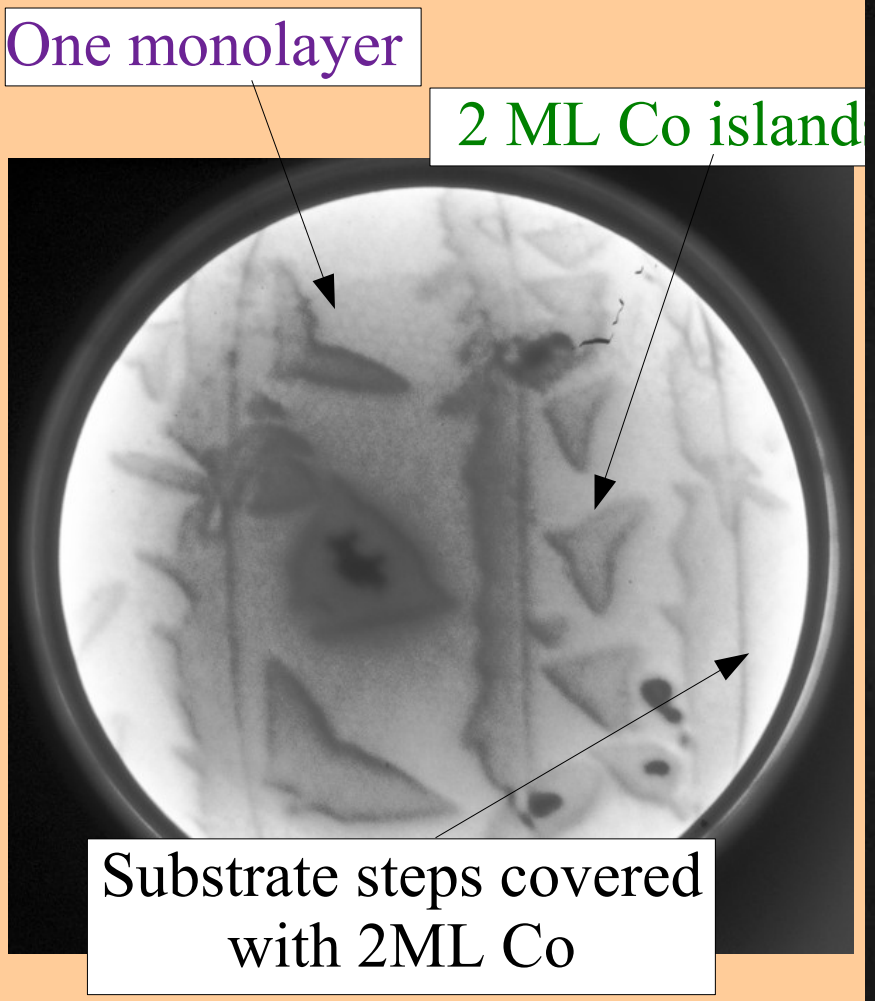
SPLEEM **in-plane**



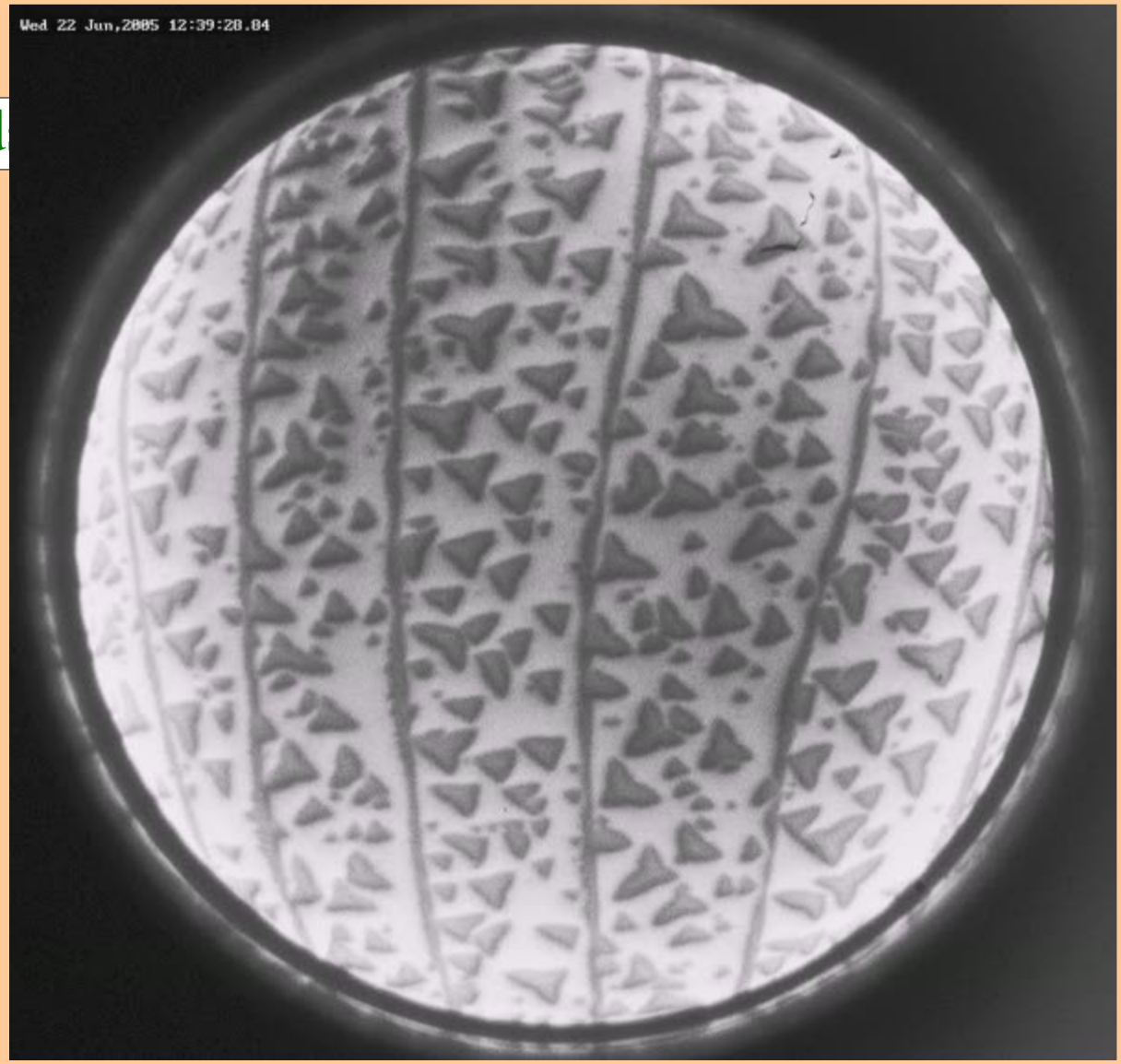
inplane

We put Co until magnetic contrast (in-plane) appears at RT

Magnetization of *2 ML islands on 1 ML Co/Ru(0001)*

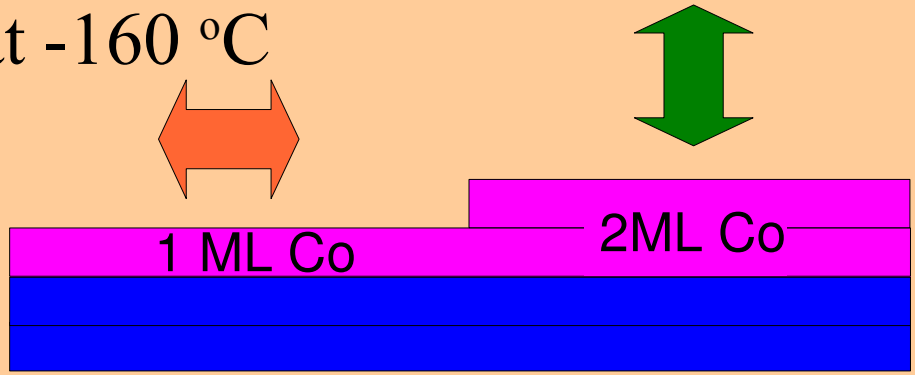


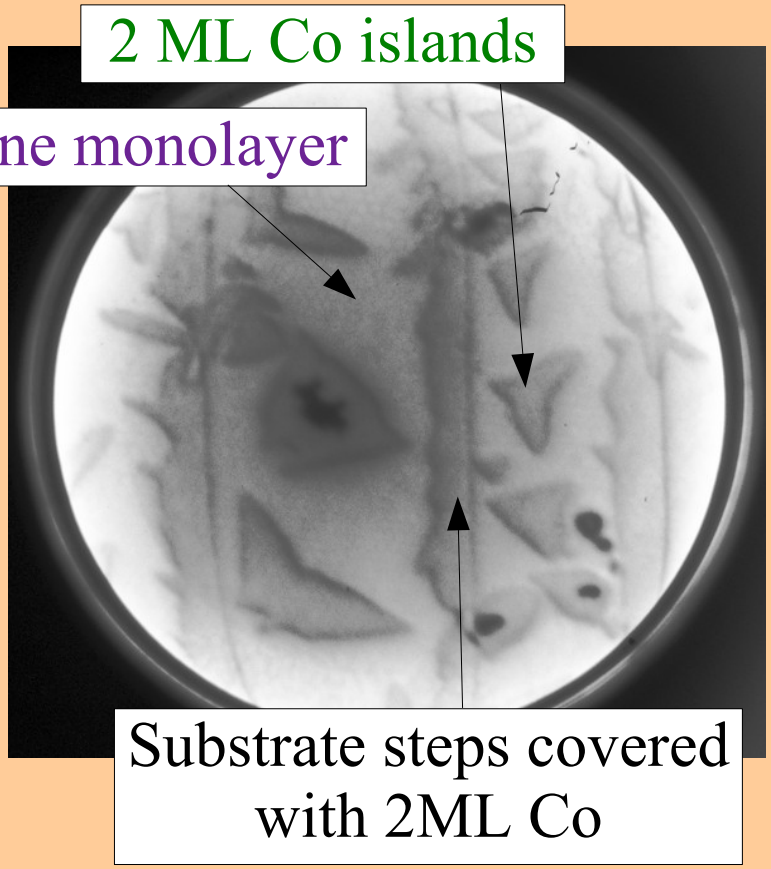
2.8 μm FOV
grown at 215 °C

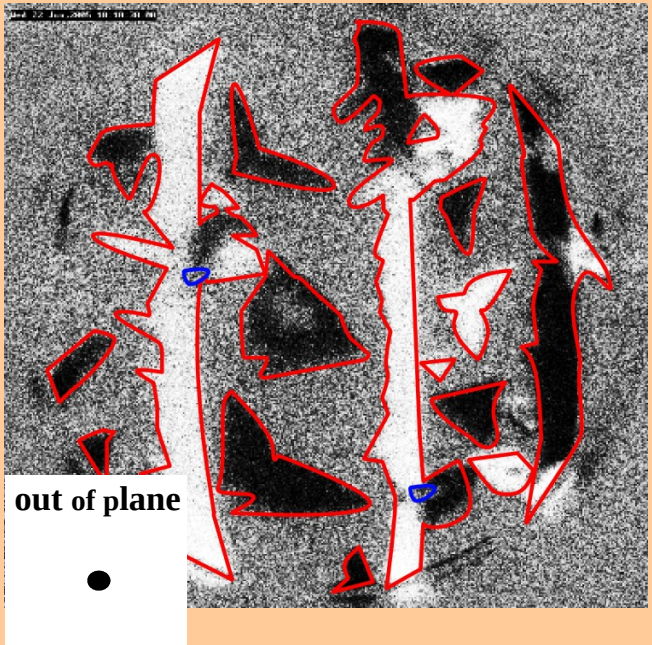


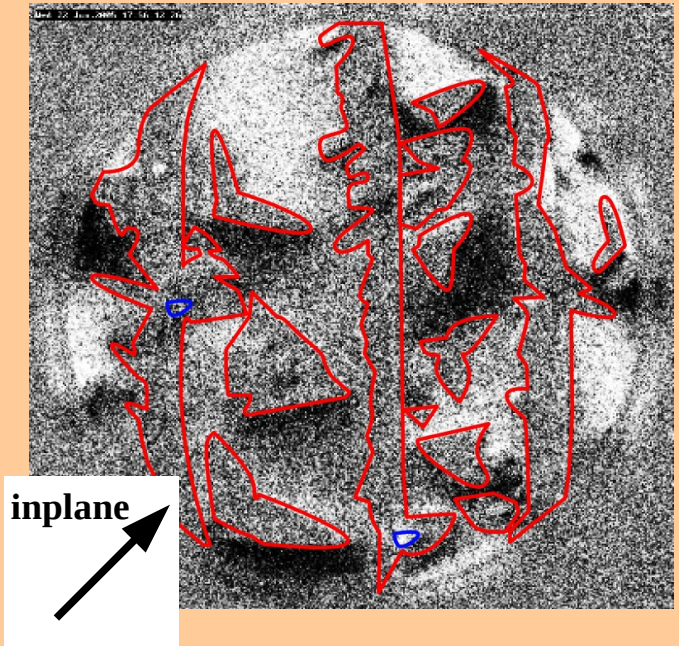
Magnetization of
2ML islands on 1ML Co/Ru(0001)

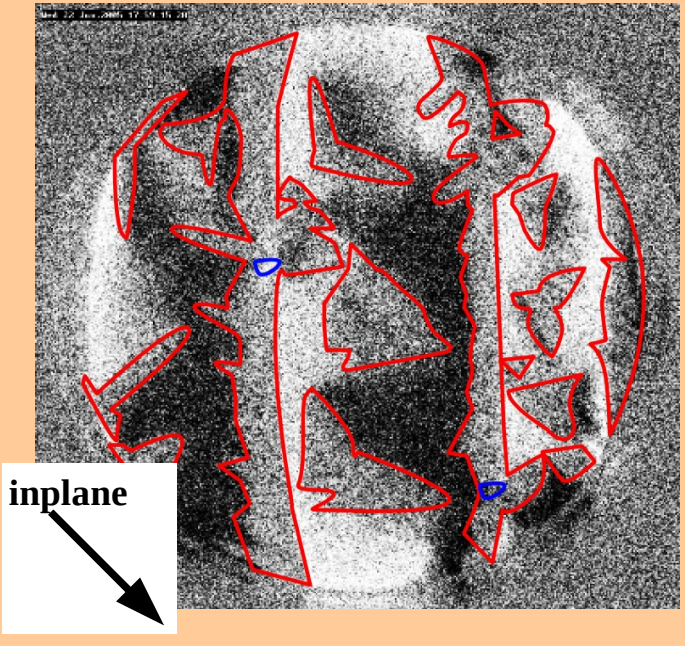
2.8 μm FOV
measured at -160 °C



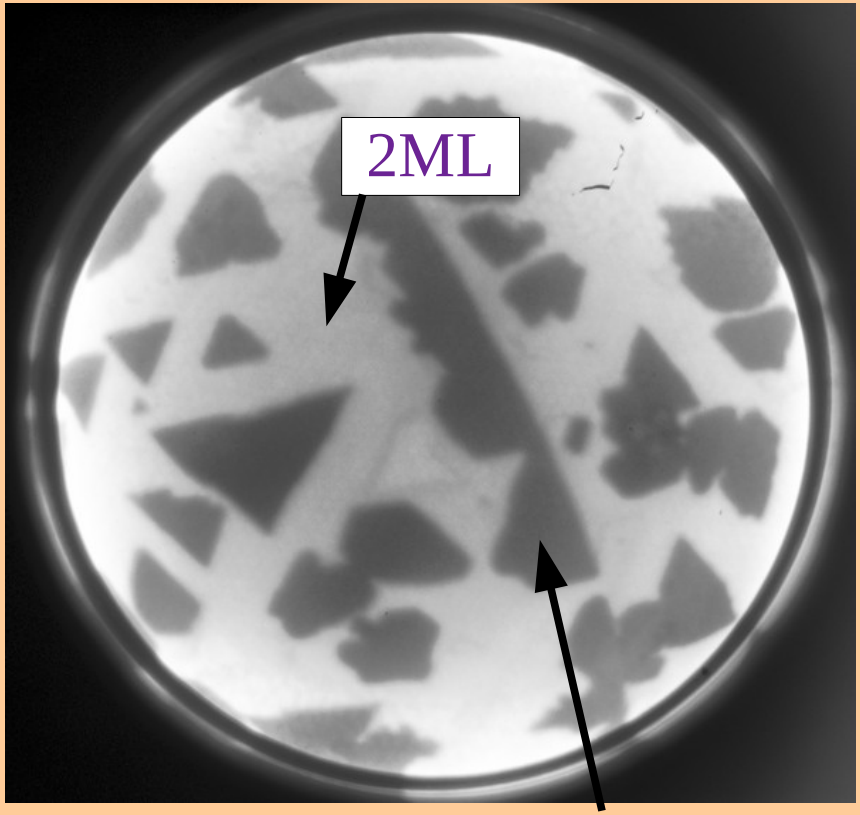


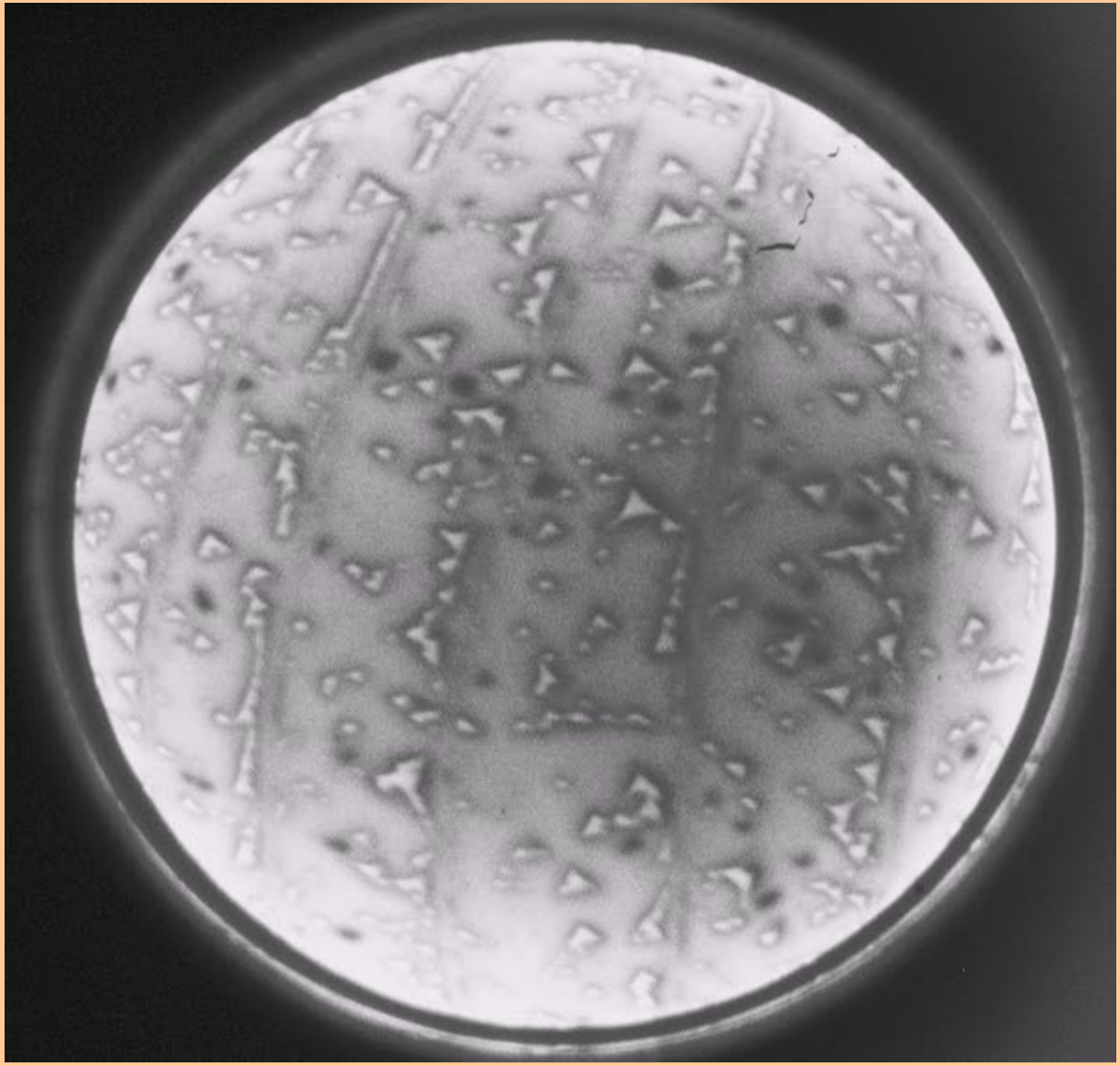






Magnetization of
3ML islands on 2 ML Co/Ru(0001)

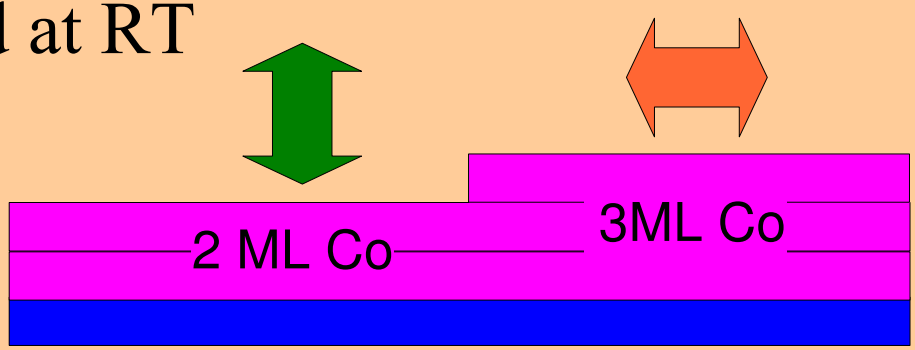


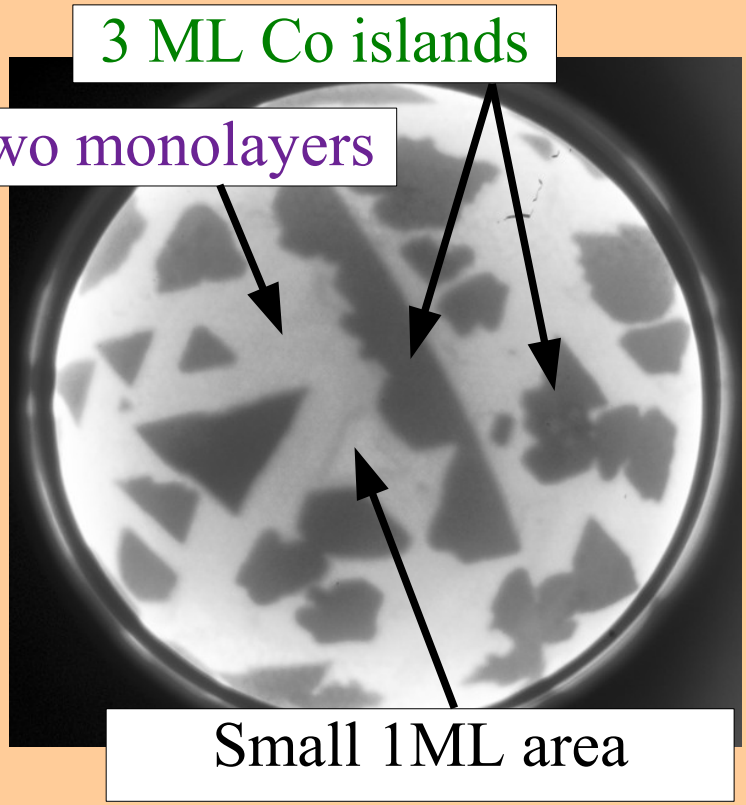


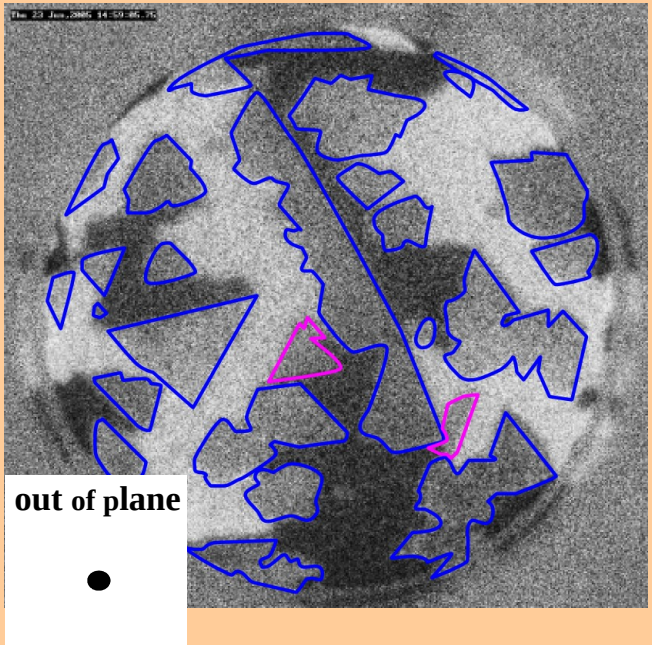
2.8 μm FOV
Grown at RT

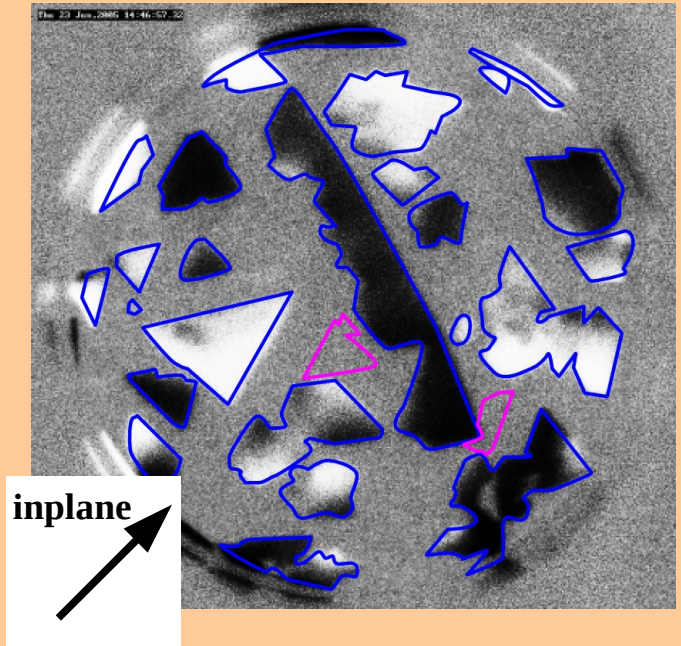
Magnetization of
3 ML islands on 2 ML Co/Ru(0001)

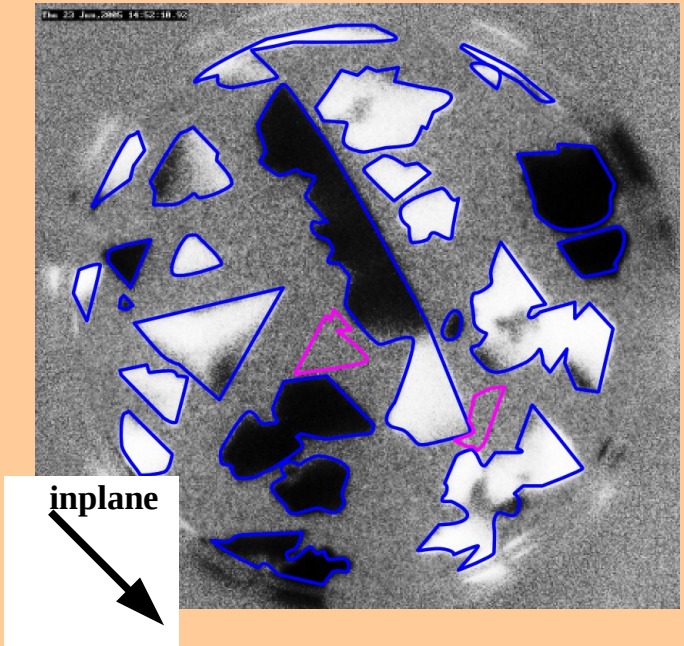
2.8 μm FOV
measured at RT



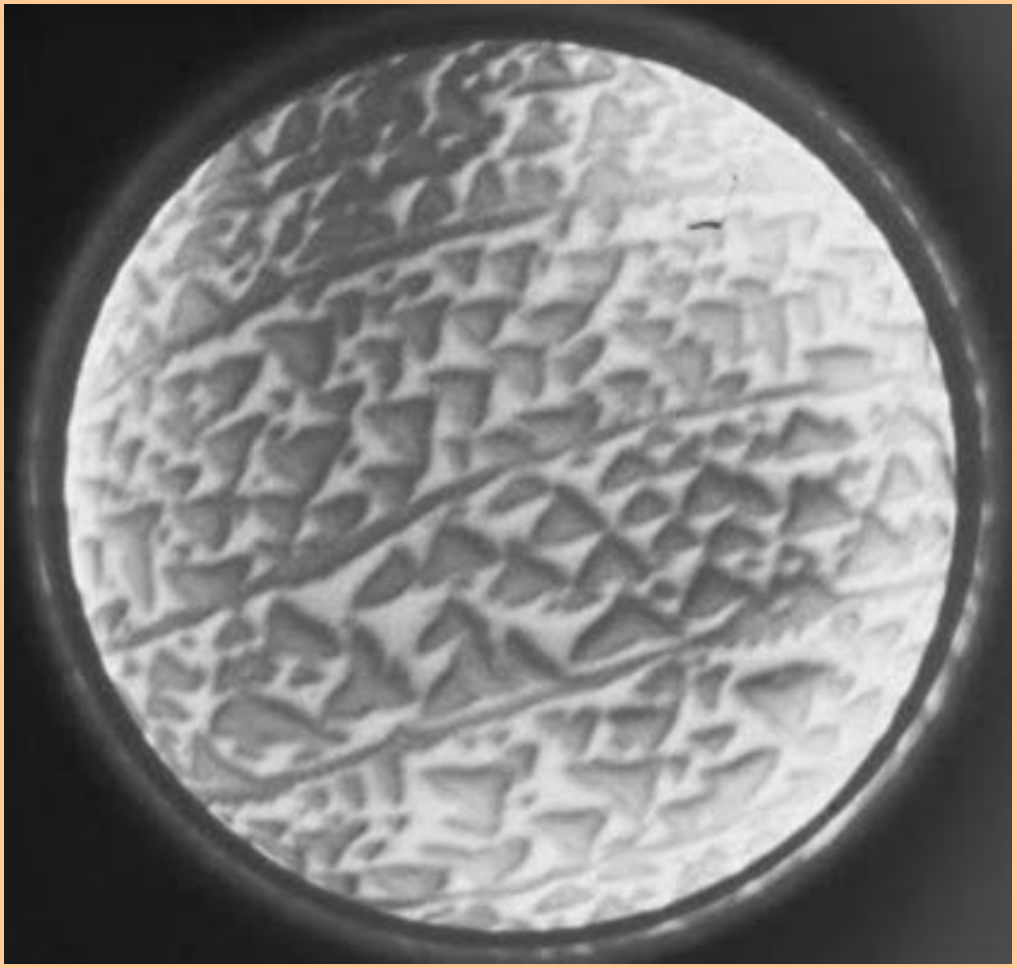


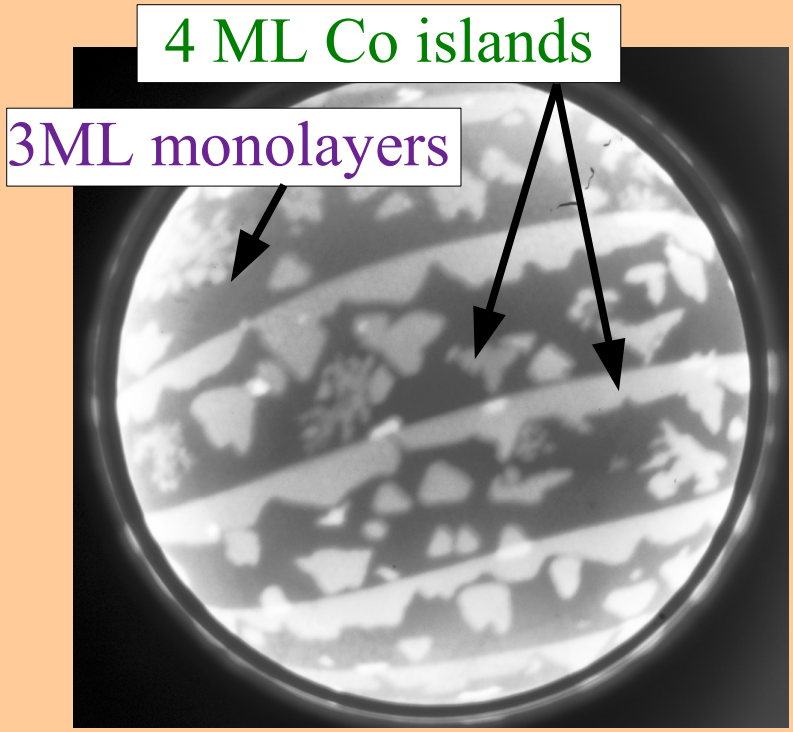






Magnetization of
4ML islands on 3ML Co/Ru(0001)

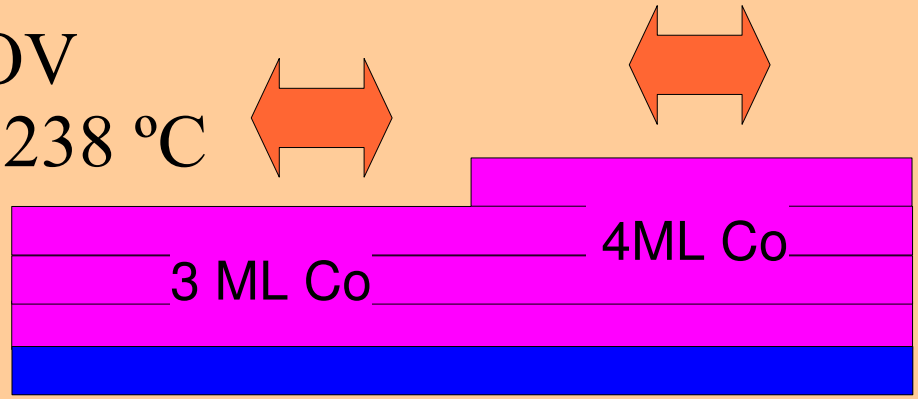


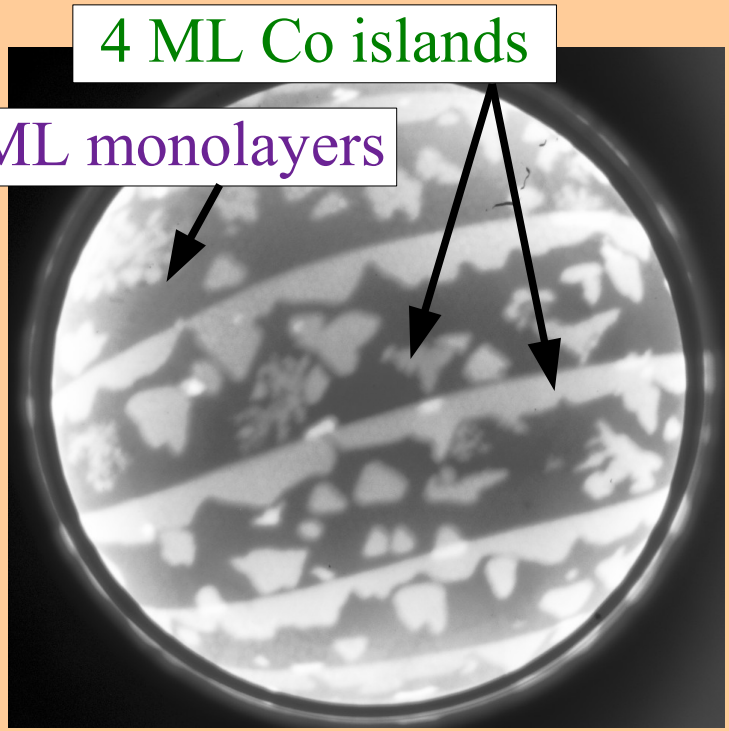


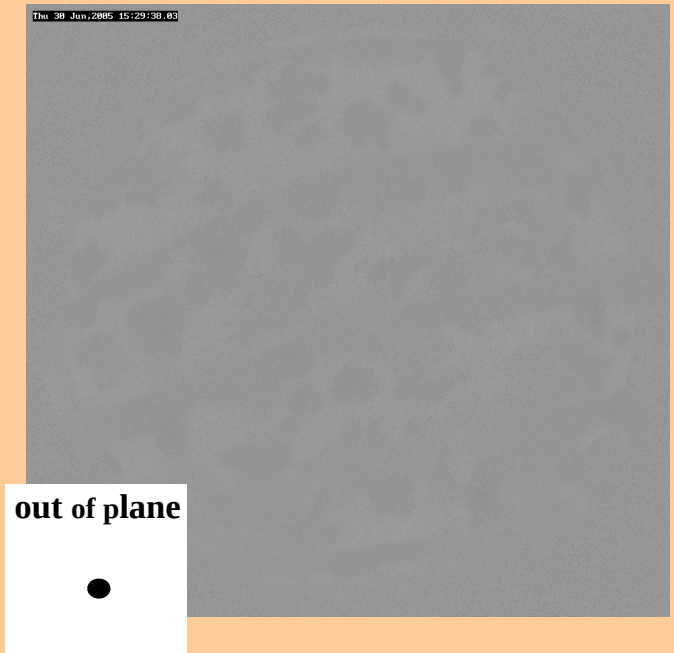
T= 237ºC, FOV= 7μm

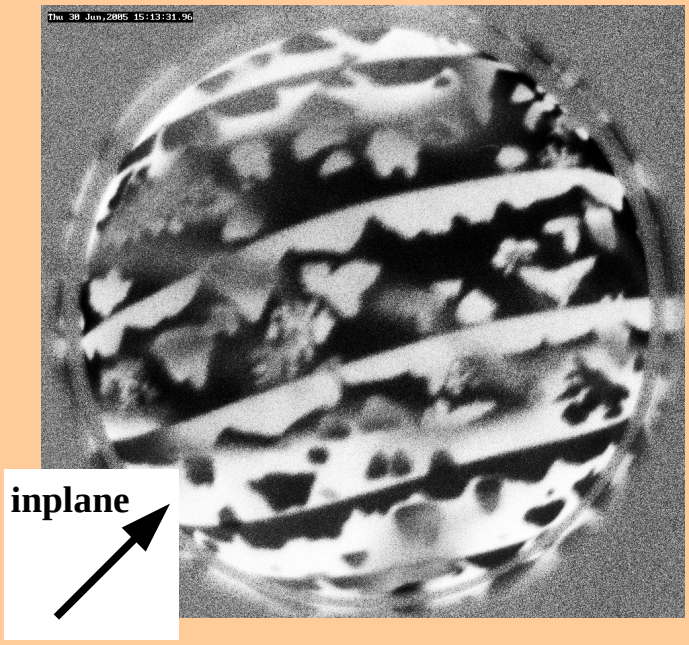
Magnetization of
4 ML islands on 3 ML Co/Ru(0001)

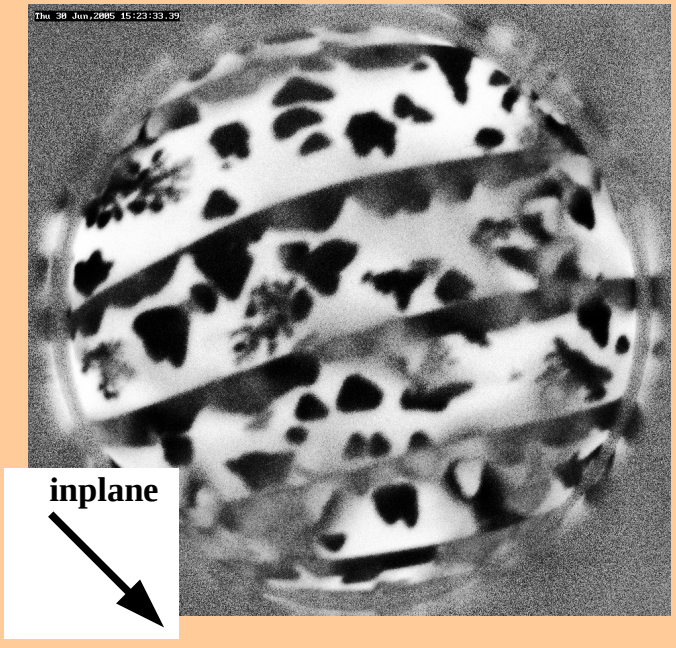
7 μm FOV
measured at 238 ºC



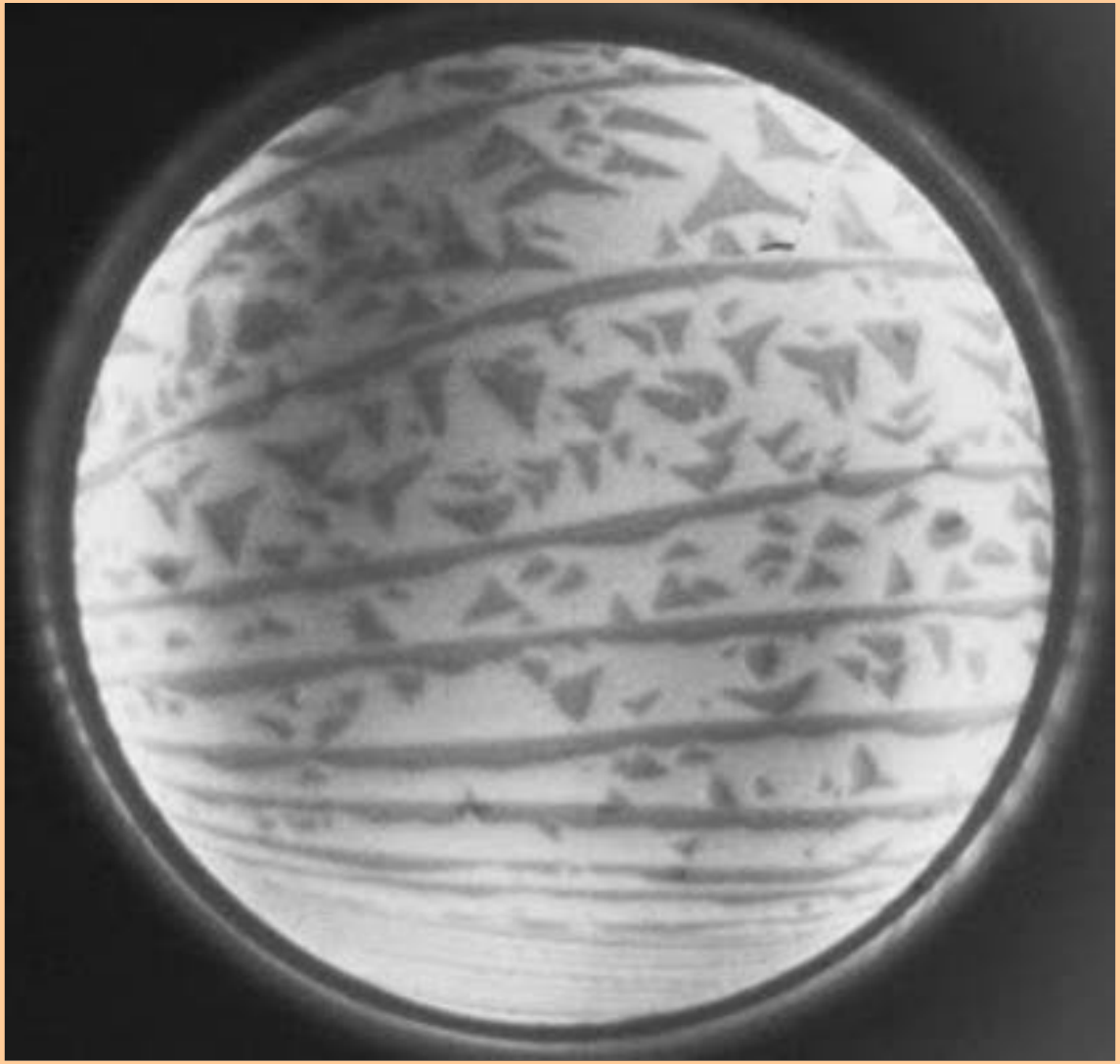




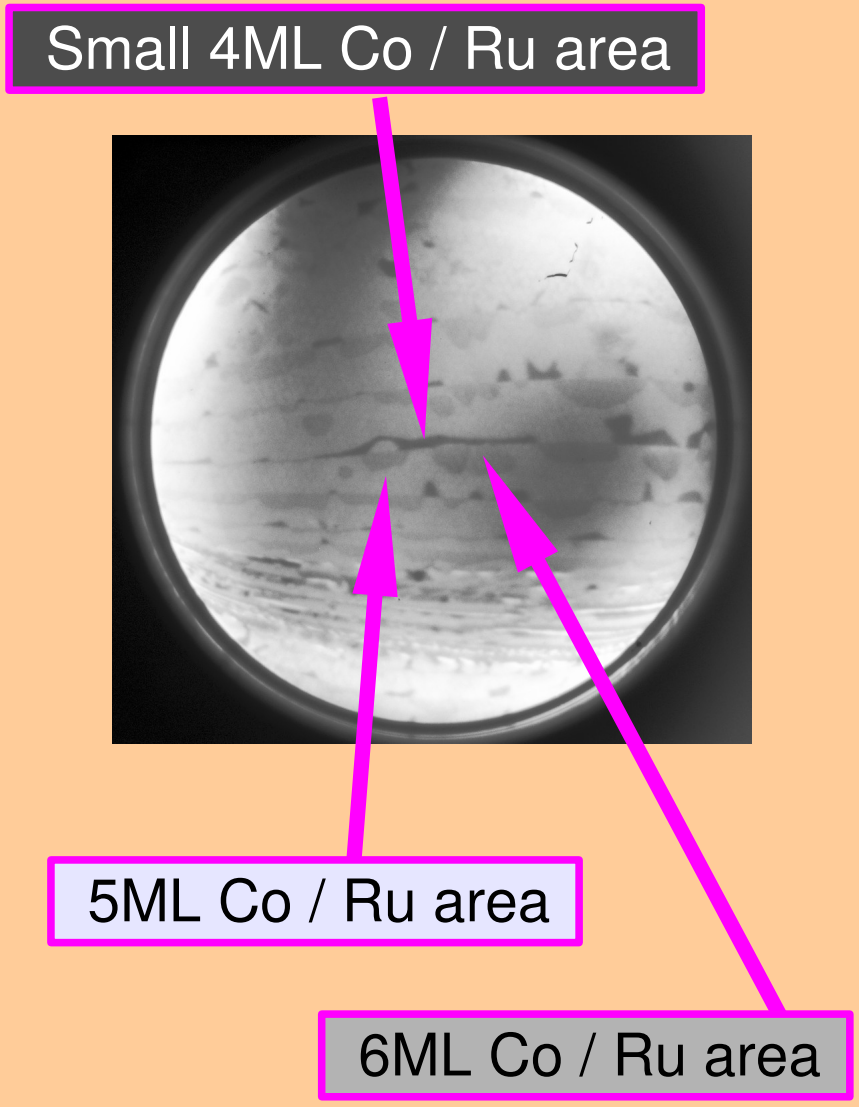




Magnetization of
6ML islands on 5ML Co/Ru(0001)



T= 240ºC, FOV= 7µm E=7eV

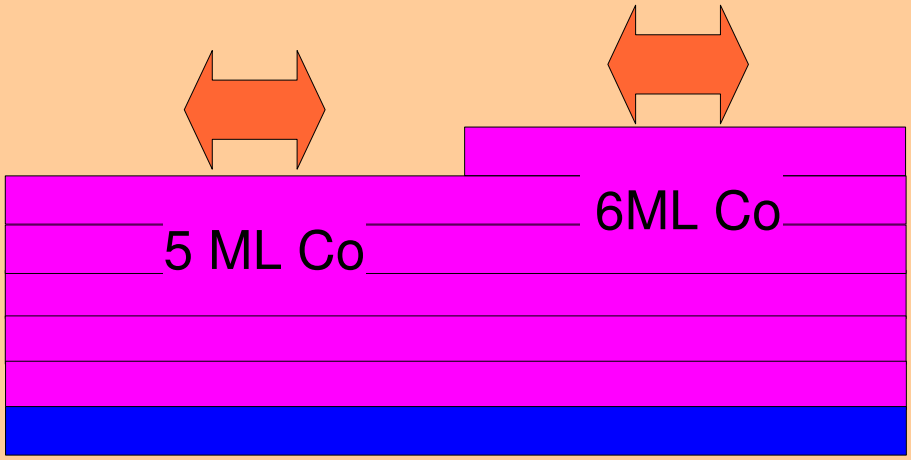


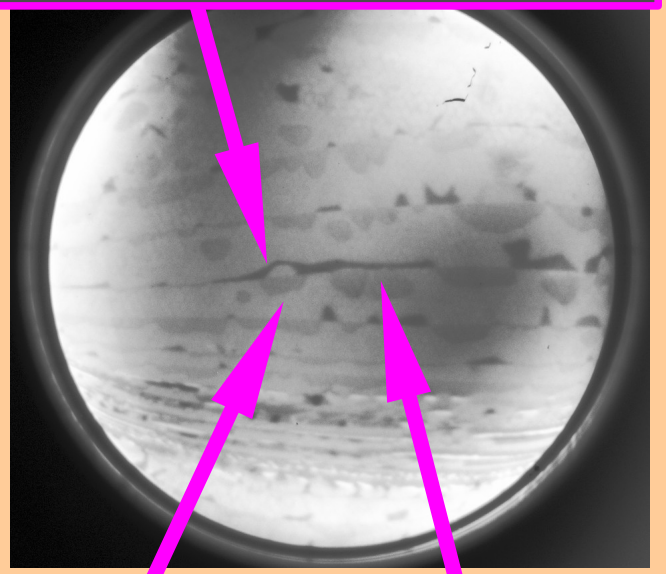
Magnetization of
6 ML islands on 5 ML Co/Ru(0001)

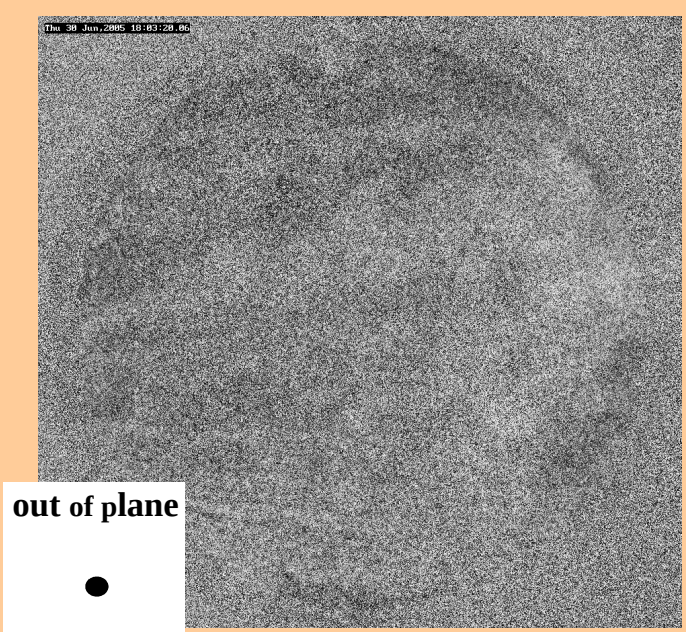
T= 245ºC

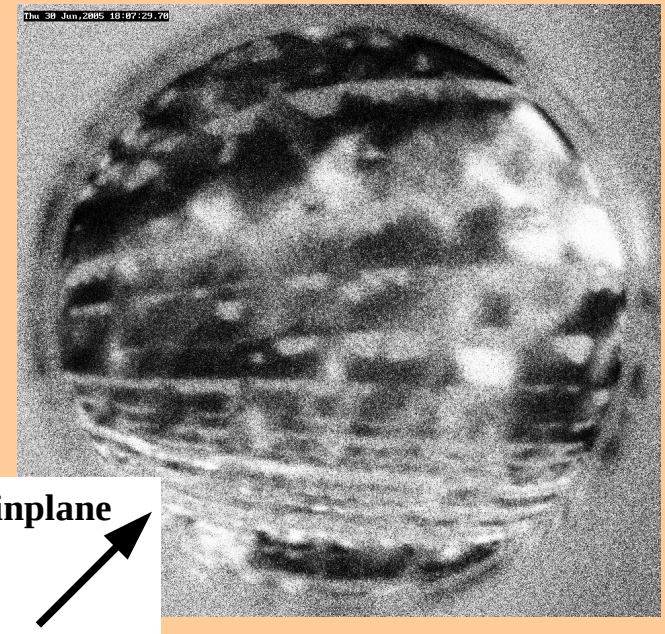
FOV= 7µm

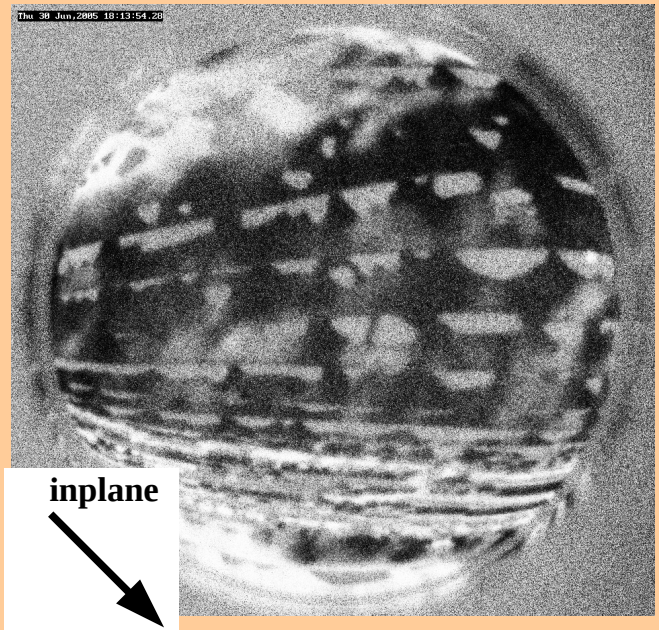
E= 7eV







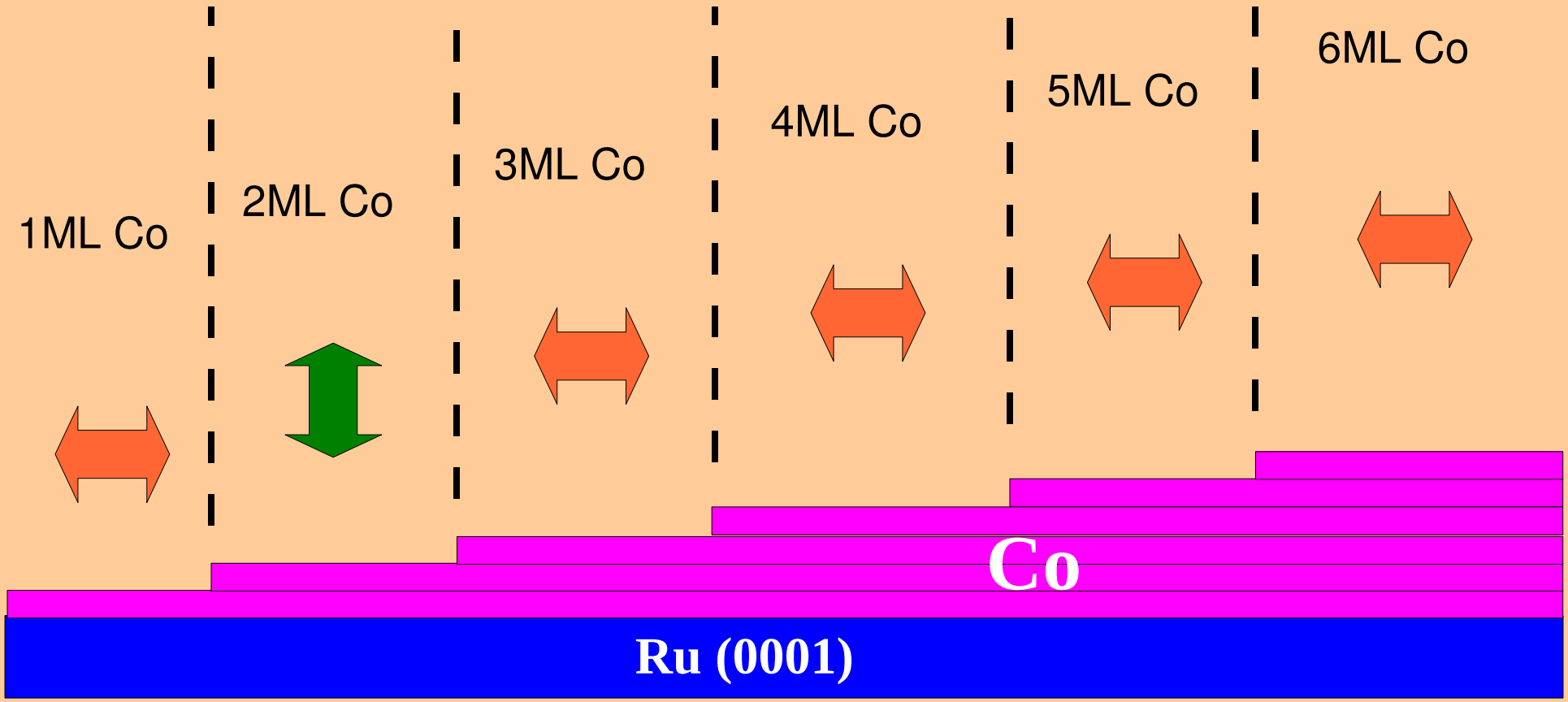




SPLEEM summary
x Co / Ru

- 1ML in-plane
- 2ML out-of-plane
- 3ML in-plane

- 4ML in-plane
- 5ML in-plane
- 6ML in-plane



Real time observation of domain evolution!

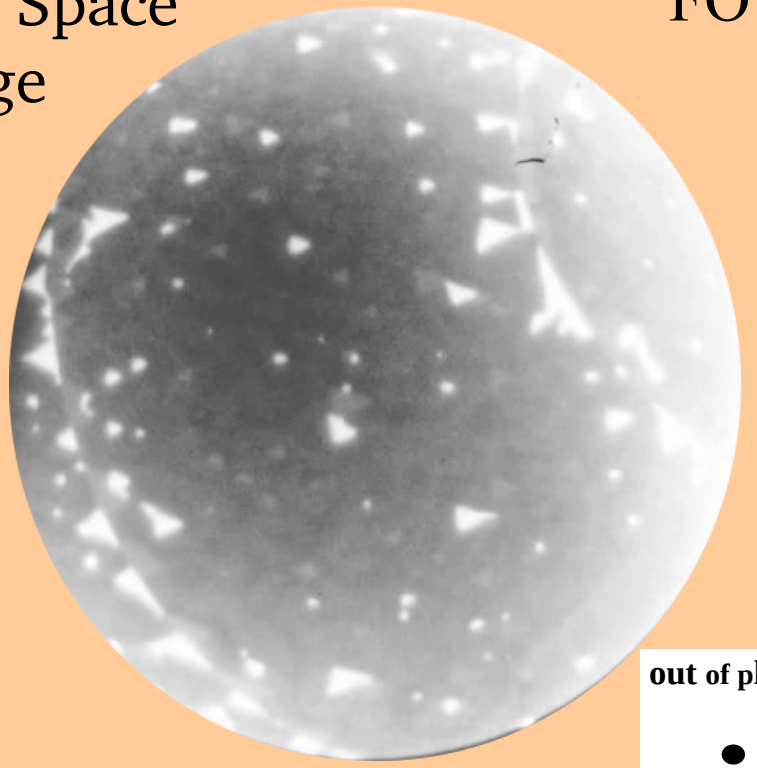
- The SPLEEM allows to observe in REAL TIME the change in magnetic domains on the surface!

2ML Co/Ru (0001)

When cooling domains coarsen

Real Space Image

FOV 7μm



out of plane

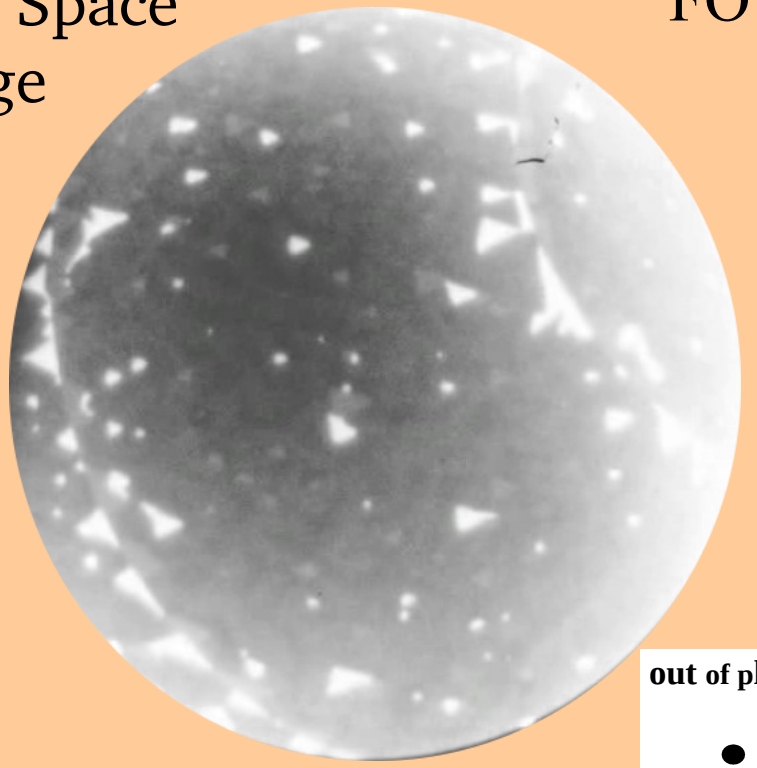
Determining the Curie temperature!

- The SPLEEM allows to observe in REAL TIME the change in magnetic domains on the surface!

2ML Co/Ru (0001)

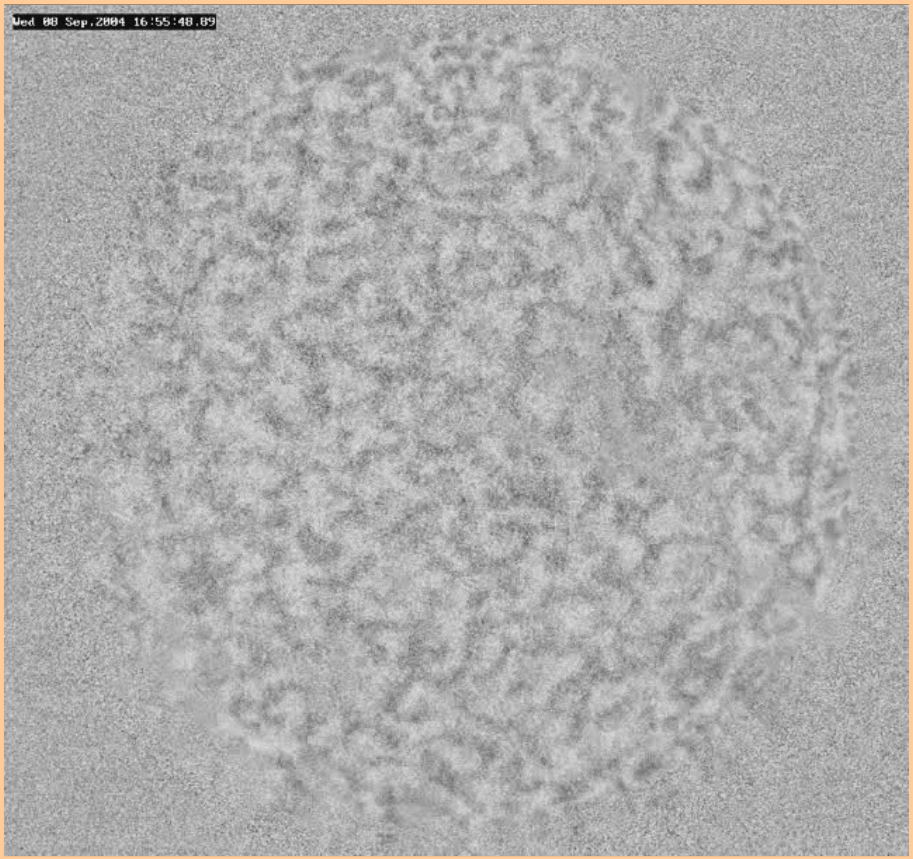
Real Space Image

FOV 7μm



out of plane

Heating until contrast is lost



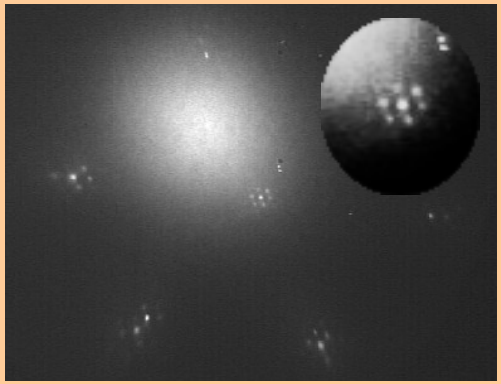
Magnetization and structure of Co/Ru below 4 ML

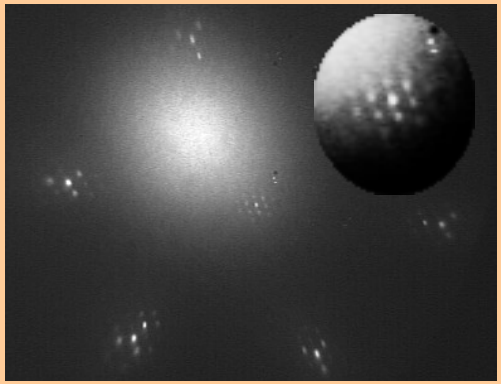
- 1 ML Co/Ru(0001):
  - in-plane magnetization, T_c = -100 ^oC
  -
- 2 ML Co/Ru(0001):
  - out-of-plane magnetization, T_c ~ 200 ^oC
  -
- 3 ML Co/Ru(0001)
  - in-plane magnetization, T_c>200 ^oC
  -

Magnetization and structure of Co/Ru below 4 ML

- 1 ML Co/Ru(0001):
  - in-plane magnetization, T_c = -100 °C
  - Pseudomorphic 1x1 structure
- 2 ML Co/Ru(0001):
  - out-of-plane magnetization, T_c ~ 200 °C
  - Relaxed Moiré structure
- 3 ML Co/Ru(0001)
  - in-plane magnetization, T_c>200 °C
  - Relaxed Moiré structure

70 eV





What drives the spin reorientation transitions?

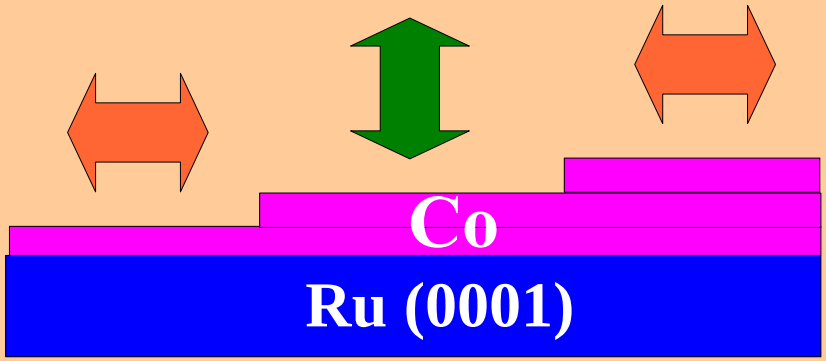
Study of LEED spots gives a relaxation for 2ML and 3ML of 5±1% (7.9% is Co/Ru misfit)

$$K_2 = K_{shape} + K_{b,2} + K_{s,2}/d$$

- Anisotropy Energy ($K_2 < 0$ → inplane mag):
  - Bulk terms:
    - Shape anisotropy
    - Magnetocristalline term (hcp)
    - Magnetoelastic term (expanded lattice)
  - Surface anisotropy ?

D. Sander, *Rep. Prog. Phys.* **62** (1999) 809

2 ML → 3 ML:
Shape anisotropy wins
1 ML → 2 ML:
Maybe magnetoelastic effects?
NEED ab-initio calculations



Ab-initio calculations: Screened Korringa-Kohn-Rostoker (KKR) method

L.Szunyogh, P. Weinberger, *Comp.Mat. Sci.* **17**, 414 (2000)

Fully relativistic spin polarized version: It is NOT a perturbation method

System with bidimensional periodicity and infinite in $k_{\parallel}$

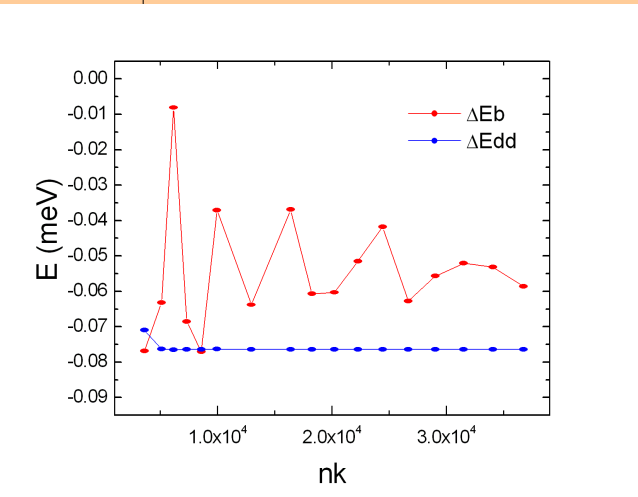
Two terms in Magnetic Anisotropy Energy (MAE):

$$MAE = \delta E_b + \delta E_{dd} \qquad \delta E = E(\perp) - E(=)$$

δE_b $\Rightarrow$ band energy, calculated by the SKKR method

δE_{dd} $\Rightarrow$ classic interaction between mag. dipoles

positive MAE $\mathbf{M}_{\perp}$ negative MAE $\mathbf{M}_{=}$



Calculations by

Silvia Gallego and Carmen Muñoz (ICMM),

L. Szunyog and P. Weinberger (TU-Wien)

ICMM

TU WIEN Technische Universität Wien Vienna University of Technology

Results from the calculations: 1 ML

Not linear! (out of magnetoelasticity)

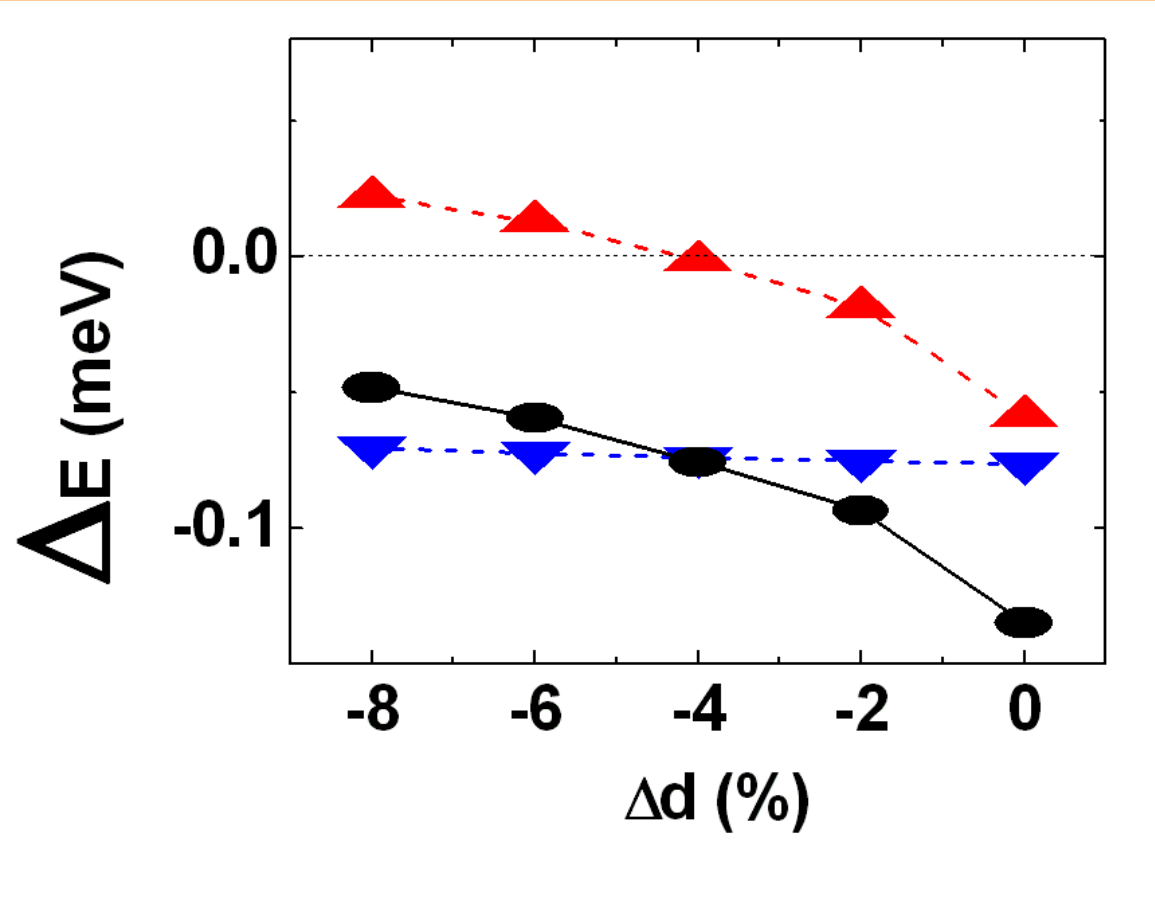
E_b band term

E_{dd} dipolar term

Total MAE

Ru in-plane lattice parameter

Changing out of plane distances



For 1 ML all the calculated structures present an in-plane magnetization

Results from the calculations: 3 ML

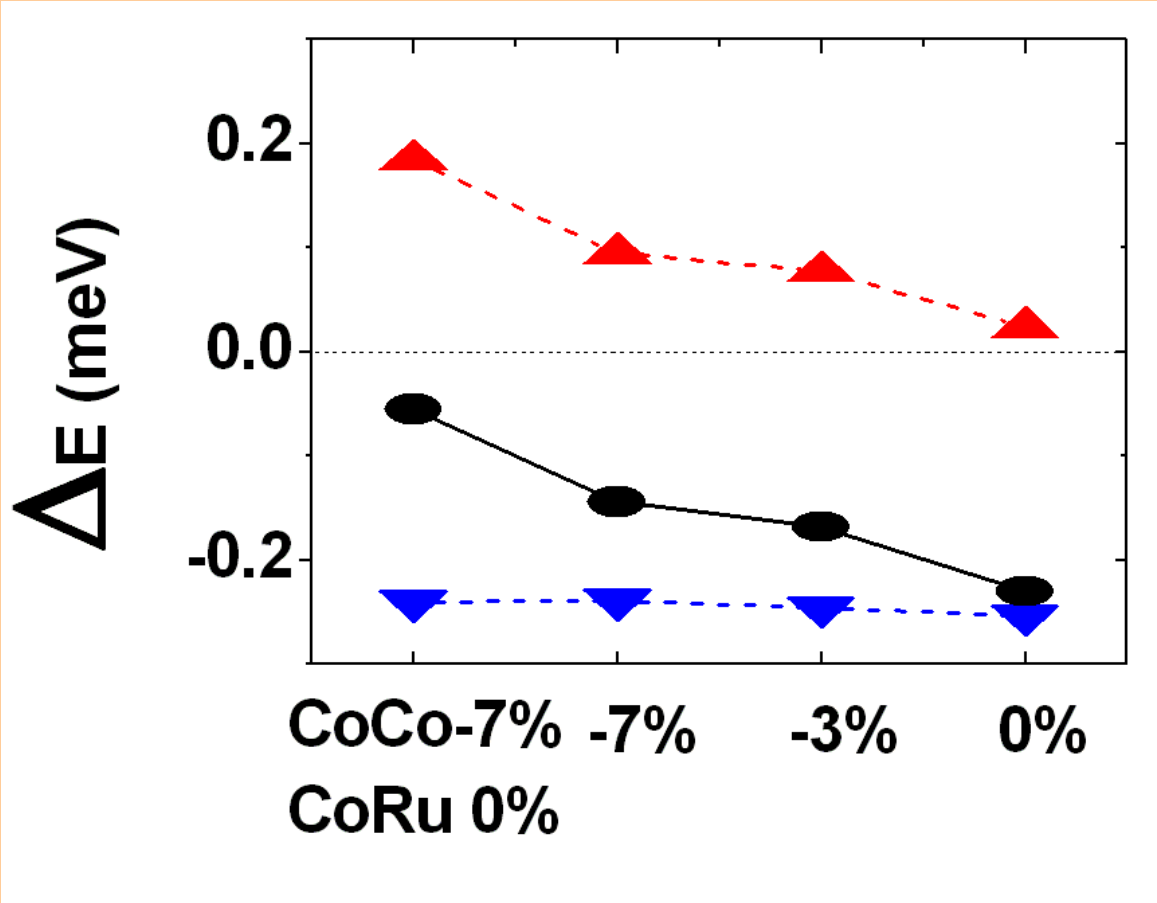
E_b band term

E_{dd} dipolar term

Total MAE

Experimental in-plane lattice parameter

Changing out of plane distances



For 3 ML all the calculated structures present an in-plane magnetization

Results from the calculations: 2 ML

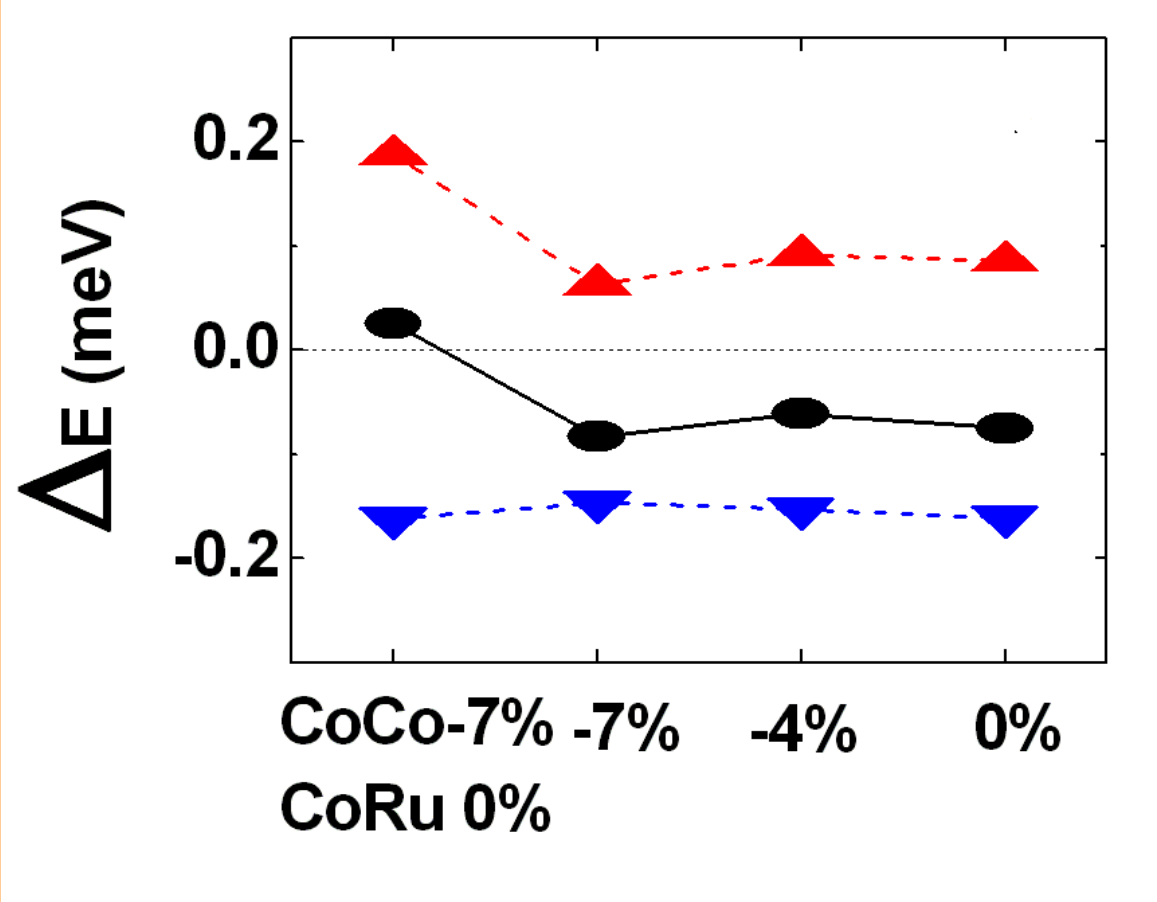
E_b band term

E_{dd} dipolar term

Total MAE

Experimental in-plane lattice parameter

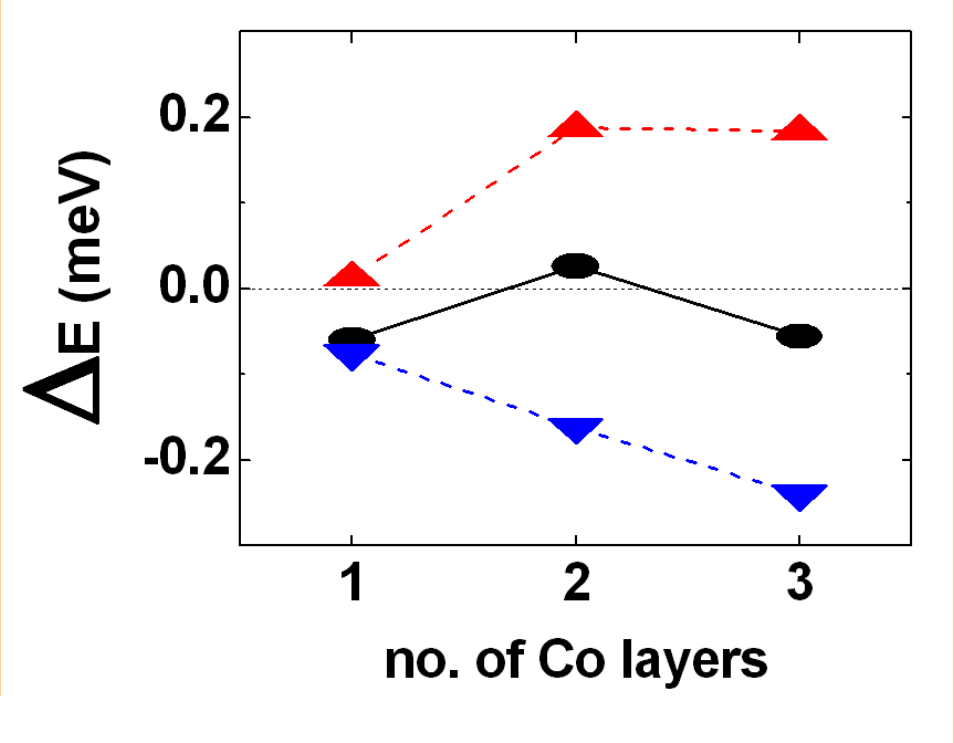
Changing out of plane distances

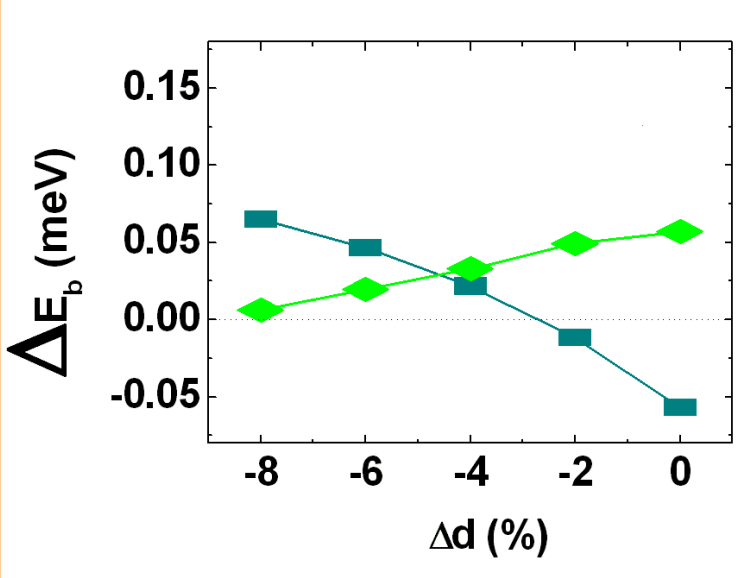


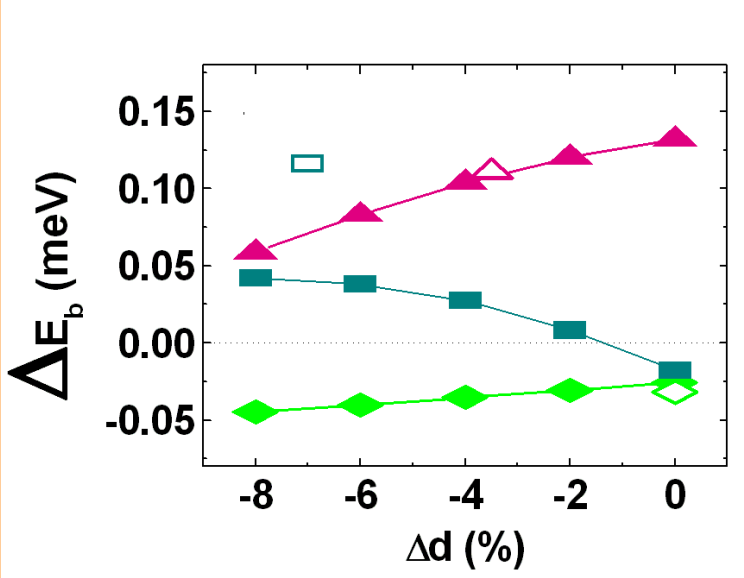
For 2 ML an appropiate combination of interlayer and in-plane lattice parameters provides out-plane magnetization

Calculations summary

Only for 2 ML an appropiate combination of vertical lattices spacings give out-of-plane magnetization: surface and interface effects *plus* strain







E_b band term

E_{dd} dipolar term

Total MAE

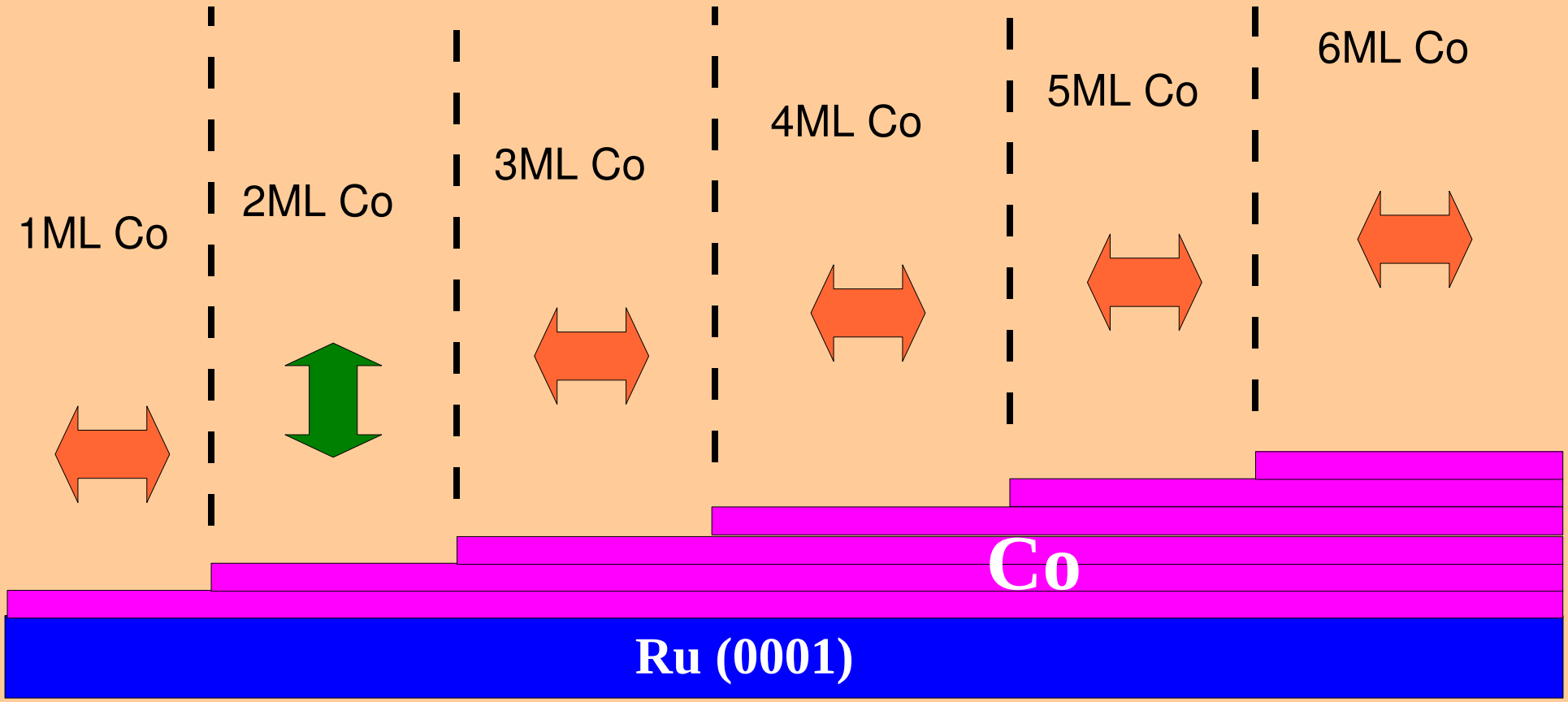
Interface Ru

Surface Co

Interface Co

SPLEEM summary
x Co / Ru

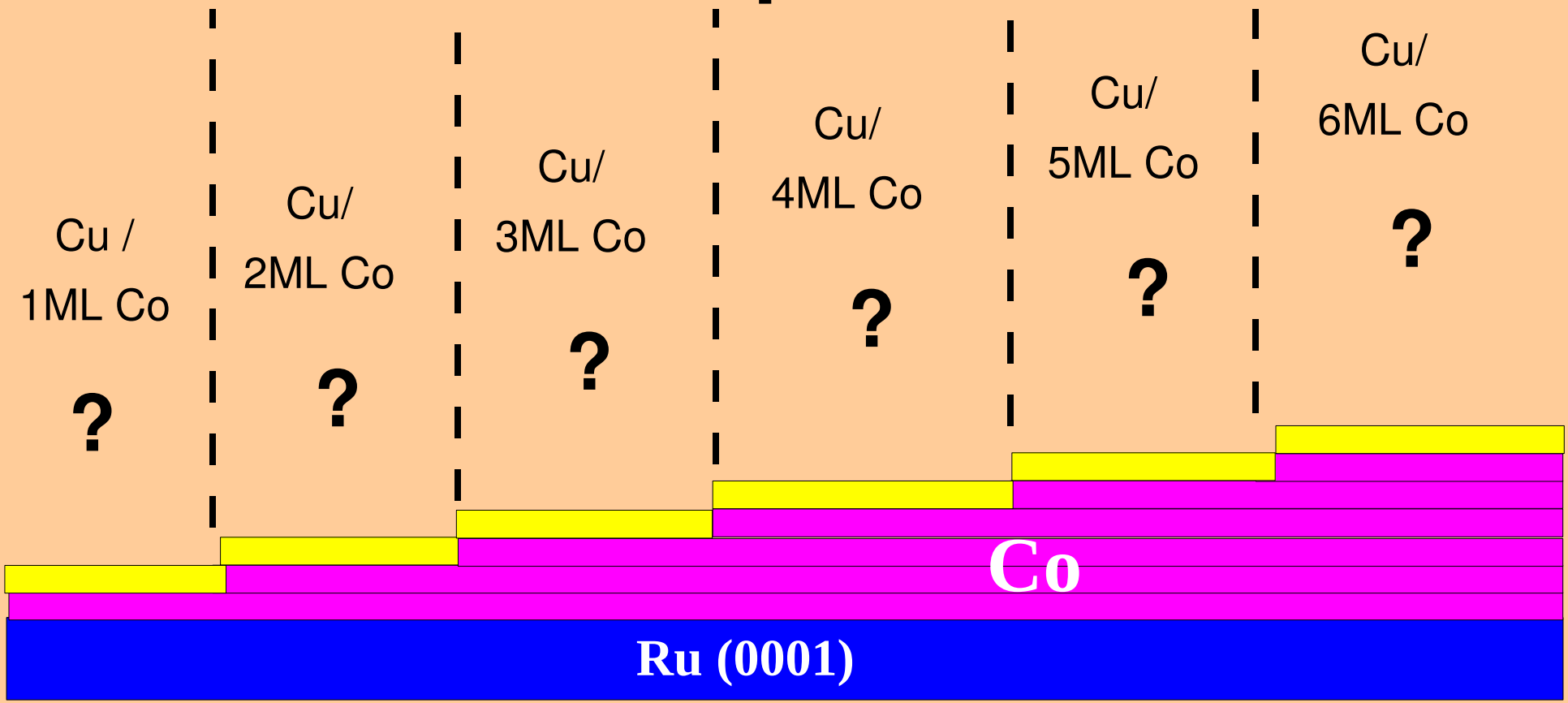
- 1ML in-plane (Curie T=170K)
- 2ML out-of-plane
- 3ML in-plane
- 4ML in-plane
- 5ML in-plane
- 6ML in-plane



How do things change with Cu?
Cu / x Co / Ru

- 1ML ?
- 2ML ?
- 3ML ?

- 4ML ?
- 5ML ?
- 6ML ?



Grow fo 1 ML Cu on 1MLCo/Ru(0001)

FOV: 7µm

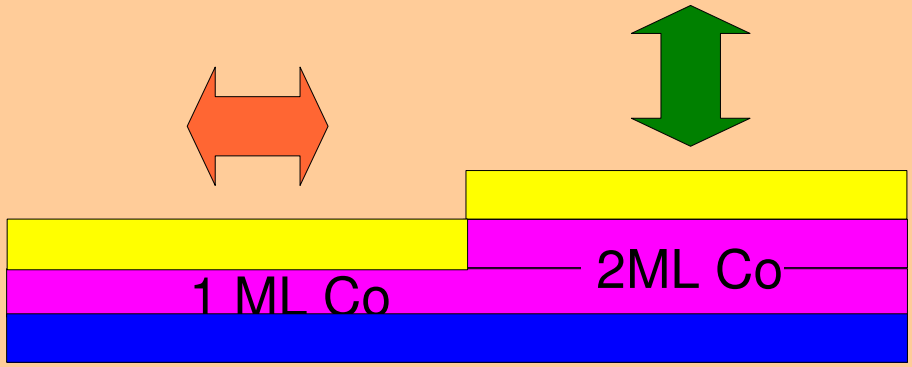
beam:7eV

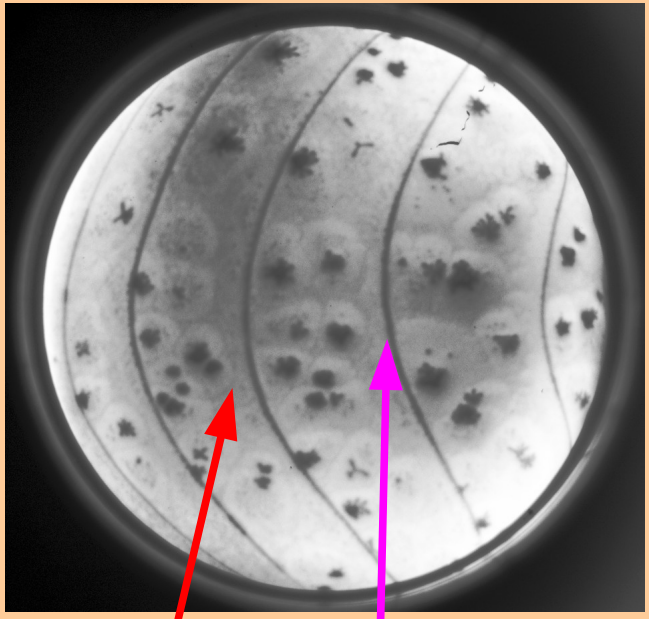
T:330ºC

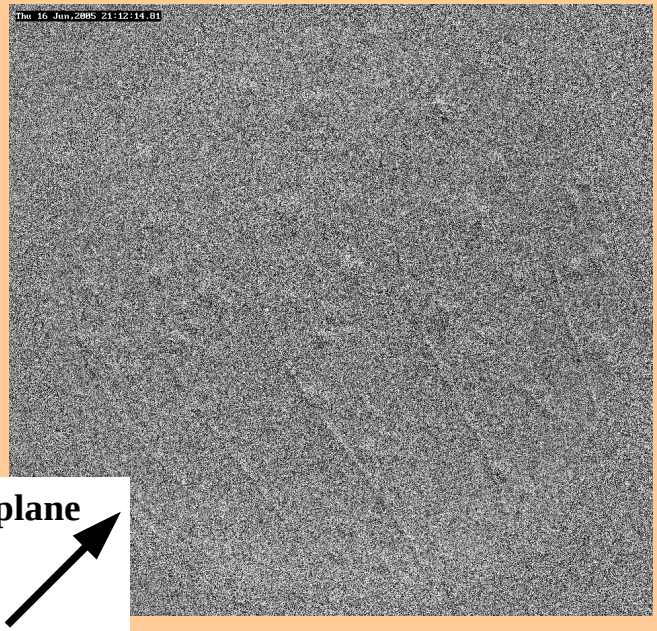
1ML Cu capping layer
on 1.2 ML Co/Ru(0001)

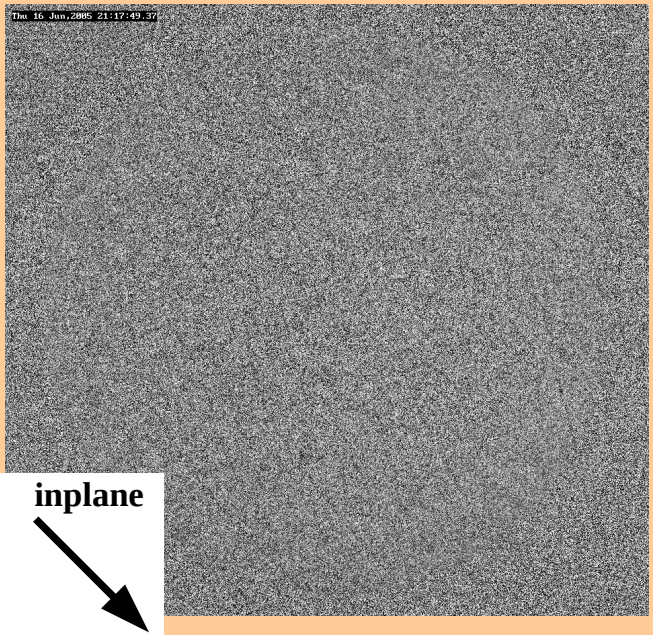
FOV: 7µm

beam:7eV







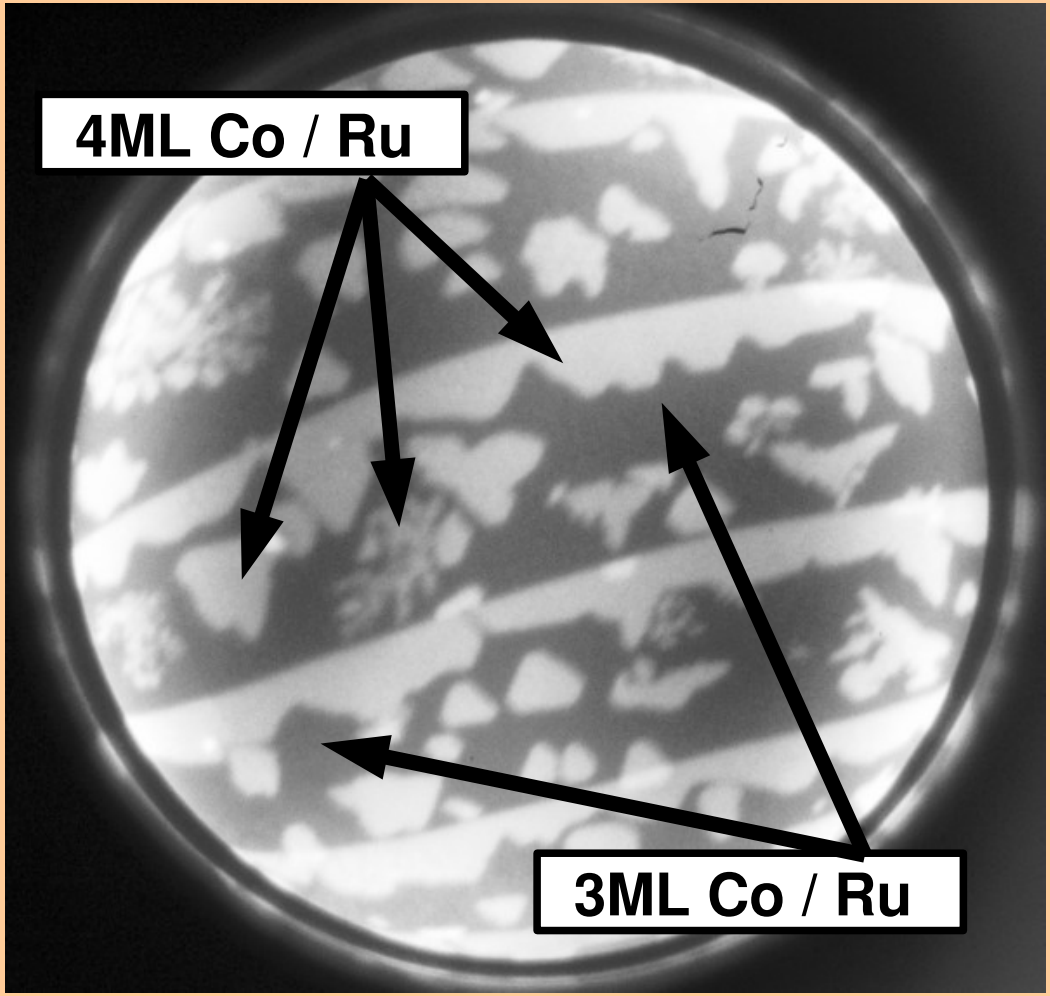


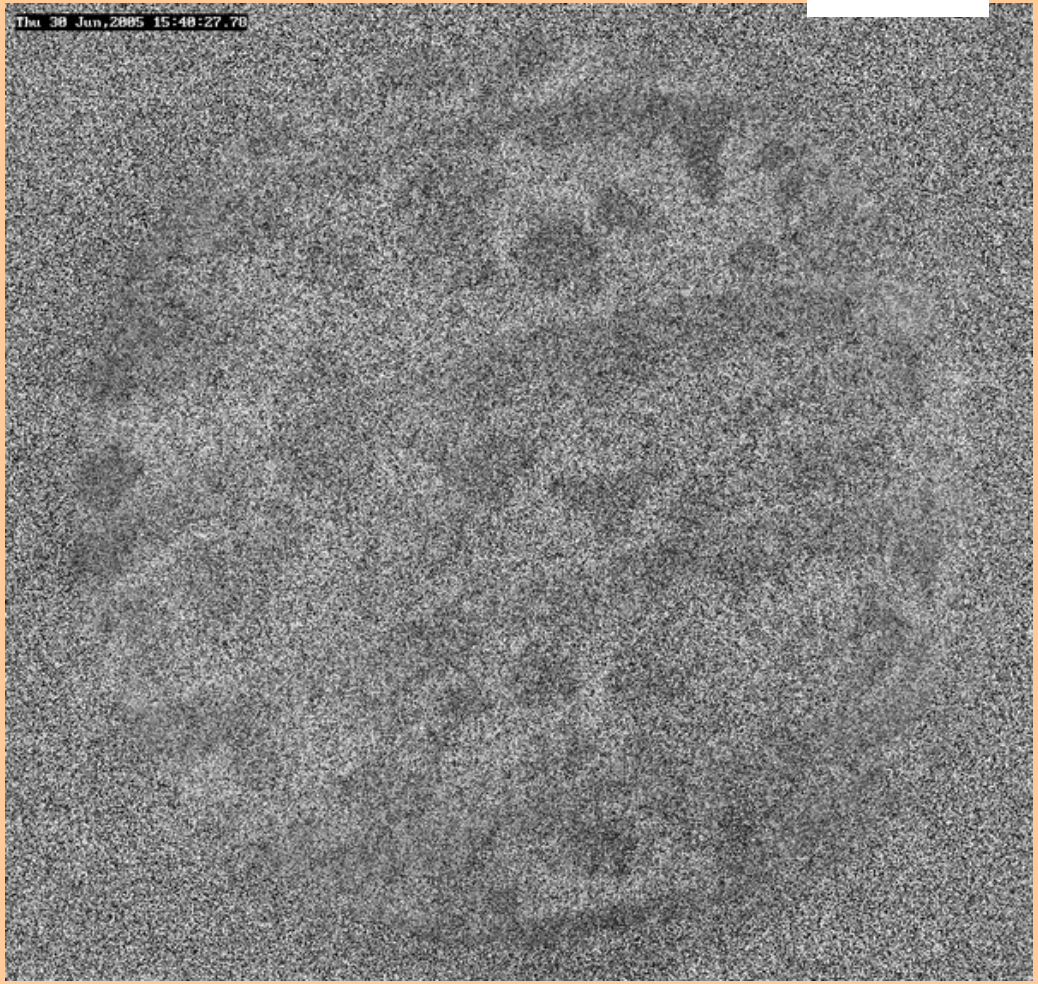
Start with 3.5 ML Co / Ru

LEEM

SPLEEM

out of plane





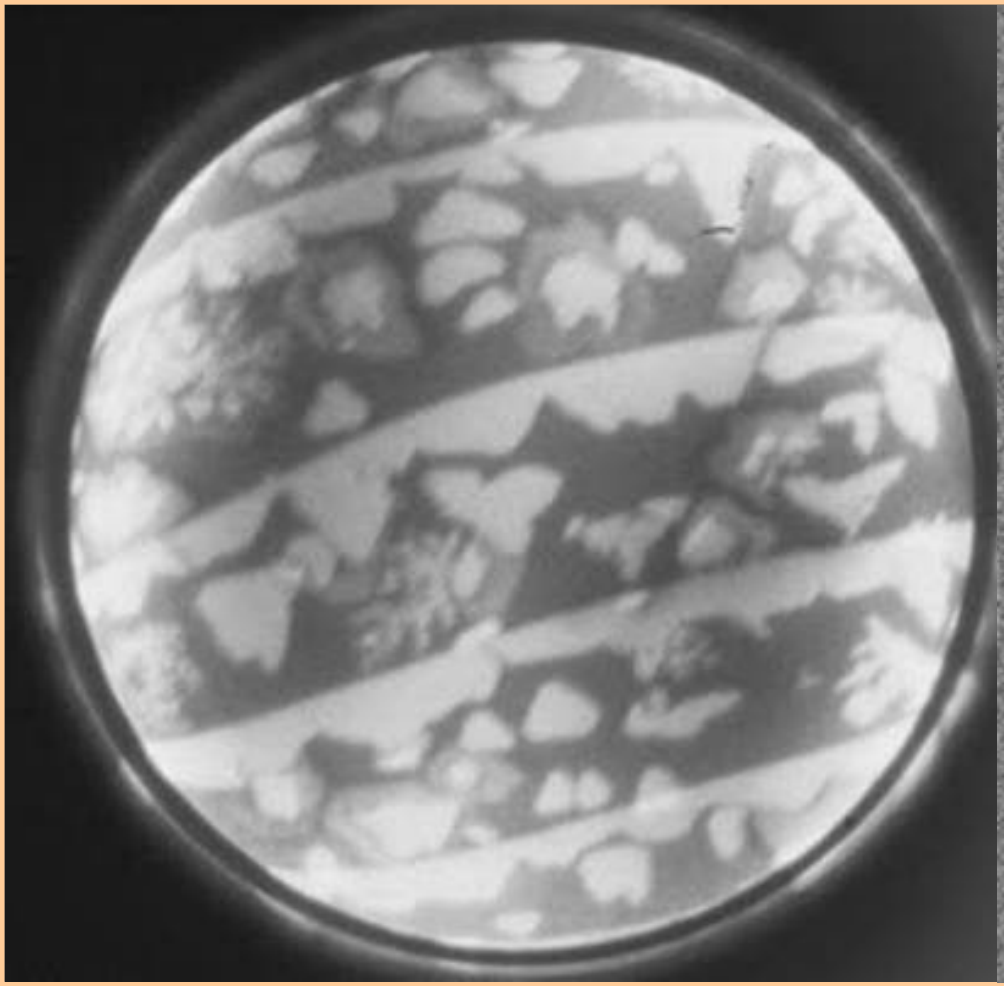
T= 240ºC, FOV= 7μm

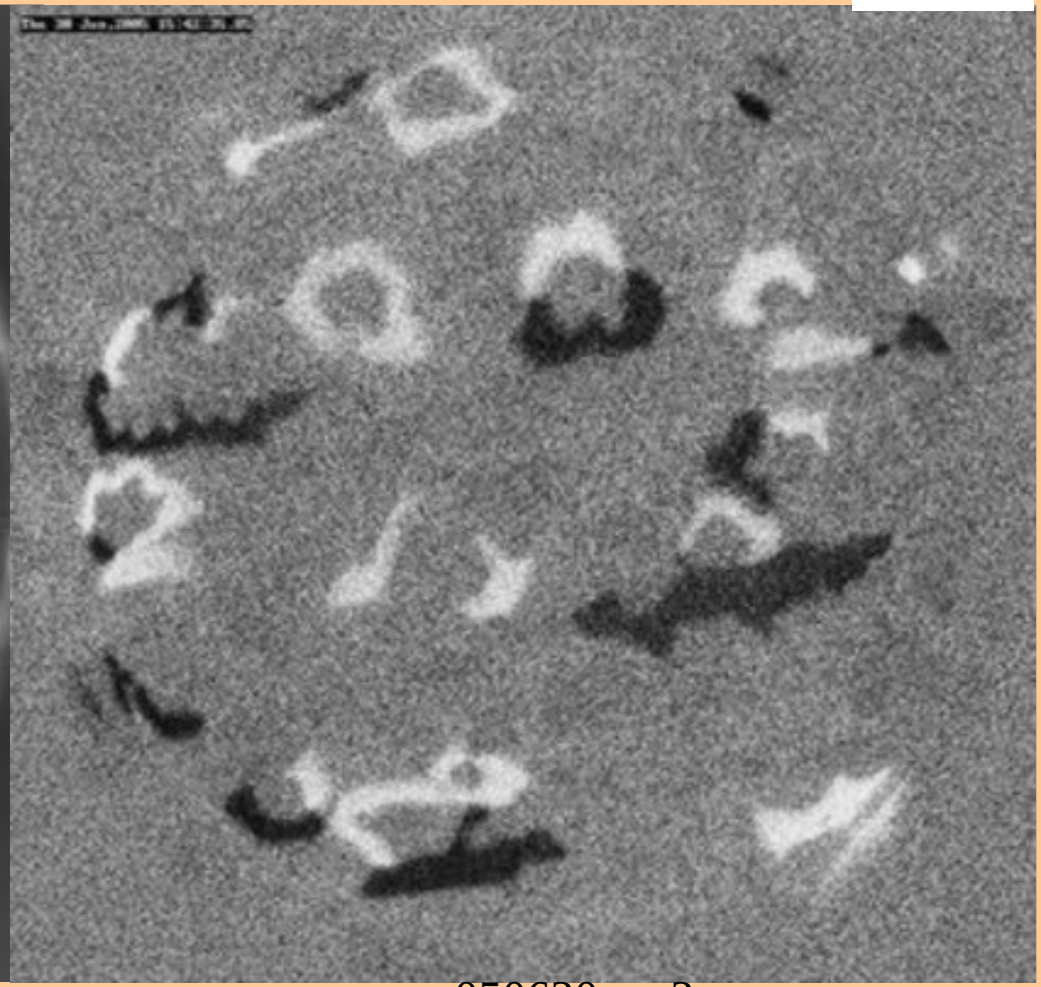
Grow 1ML Cu over 3.5 ML Co / Ru

LEEM

SPLEEM

out of plane



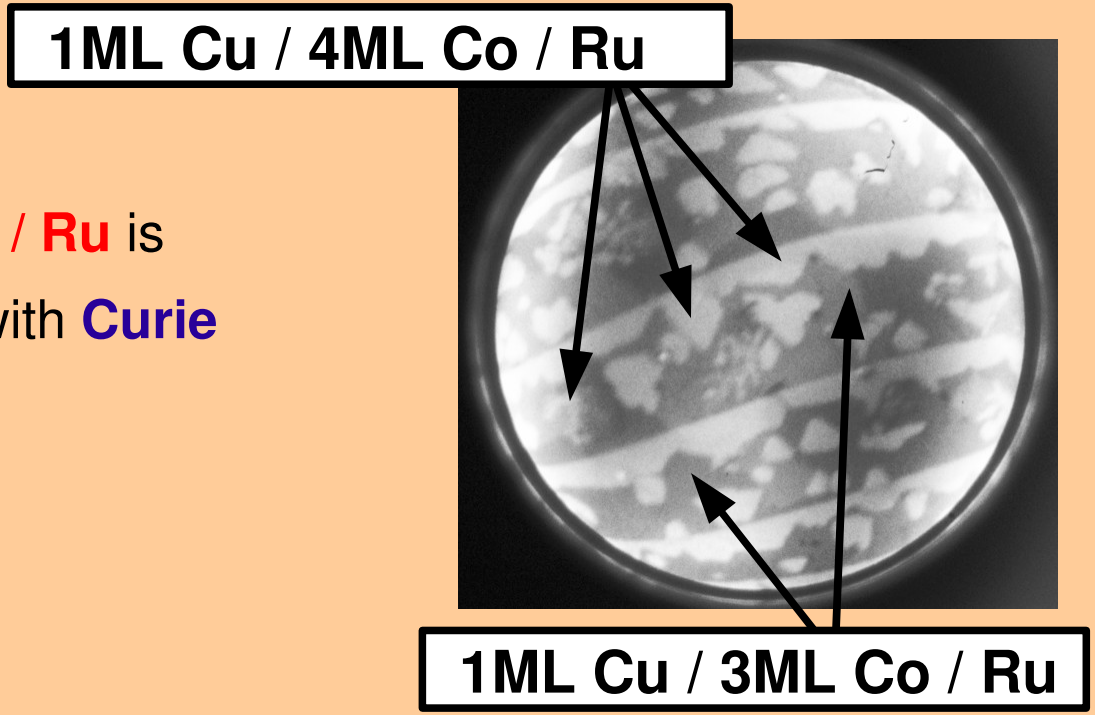


050630_no3

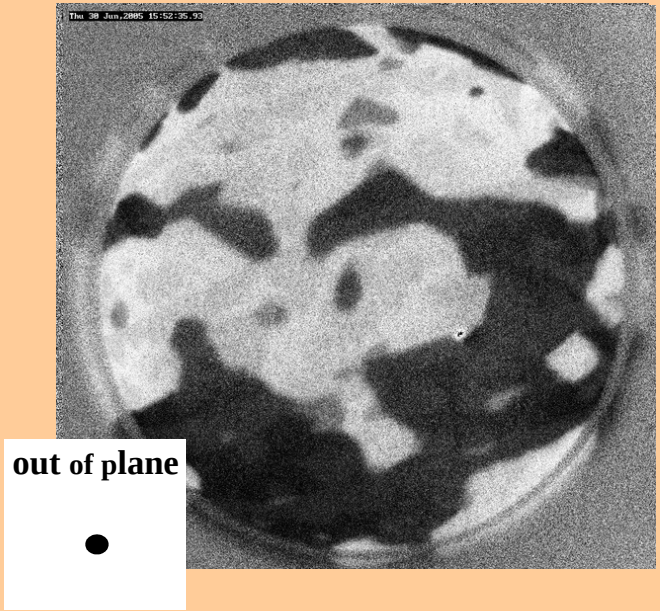
T= 240ºC, FOV= 7μm

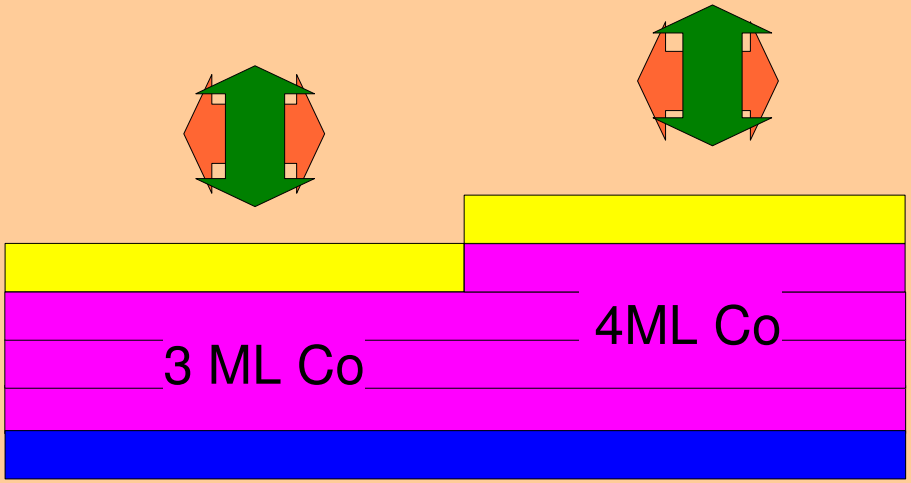
1ML Cu / 3 & 4 ML Co / Ru

T= 240ºC, FOV= 7µm E=7eV



- The system **1ML Cu / 3 and 4 ML Cu / Ru** is completely **out-of-plane** magnetized, with **Curie temperature > 540K**

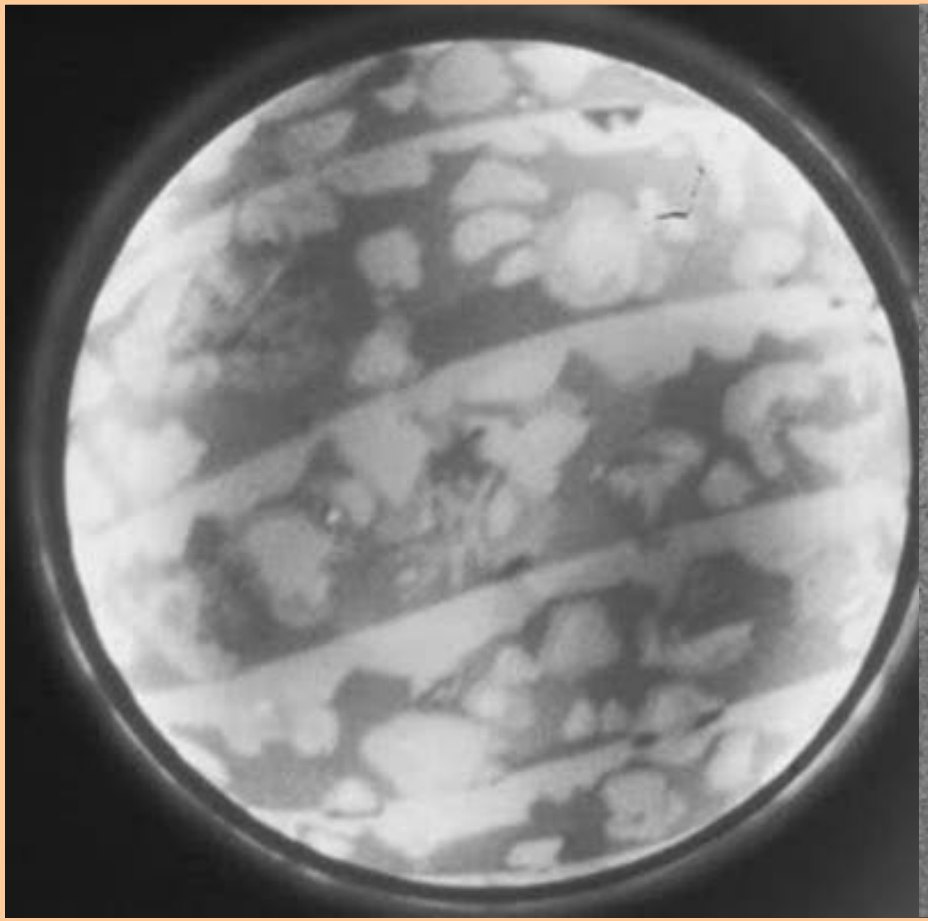


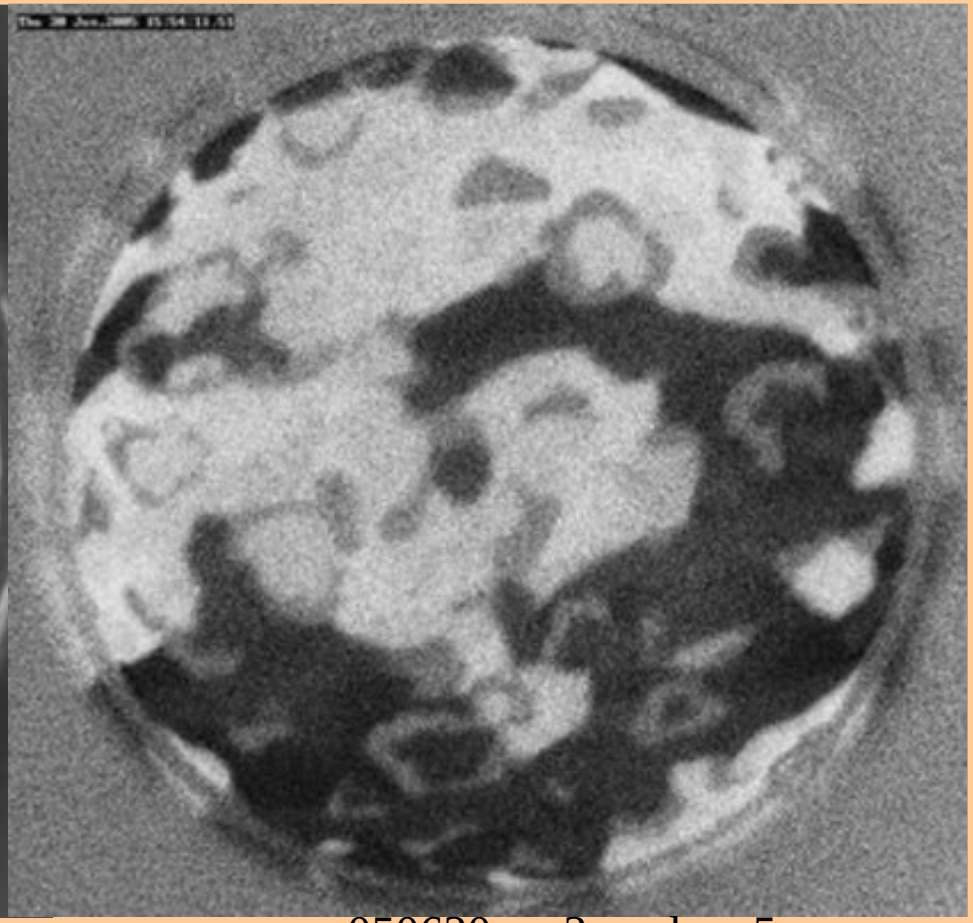


Grow another Cu monolayer:
2ML Cu capping layer on 3.5 ML Co / Ru

LEEM

SPLEEM **out-of-plane**





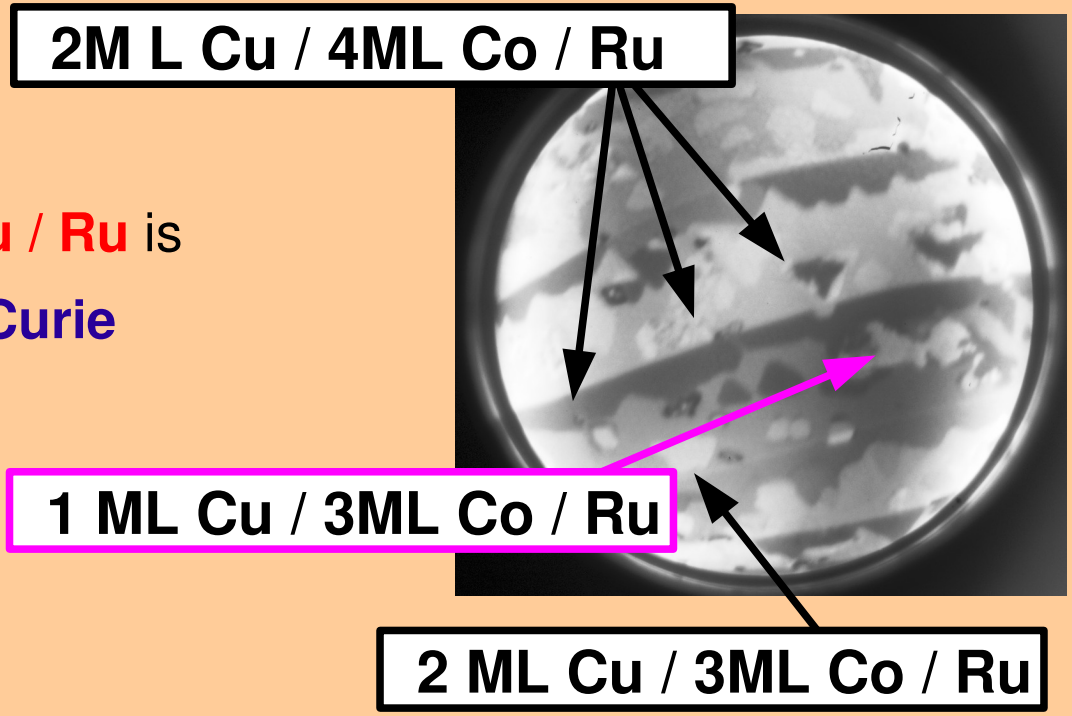
050630_no3_and_no5

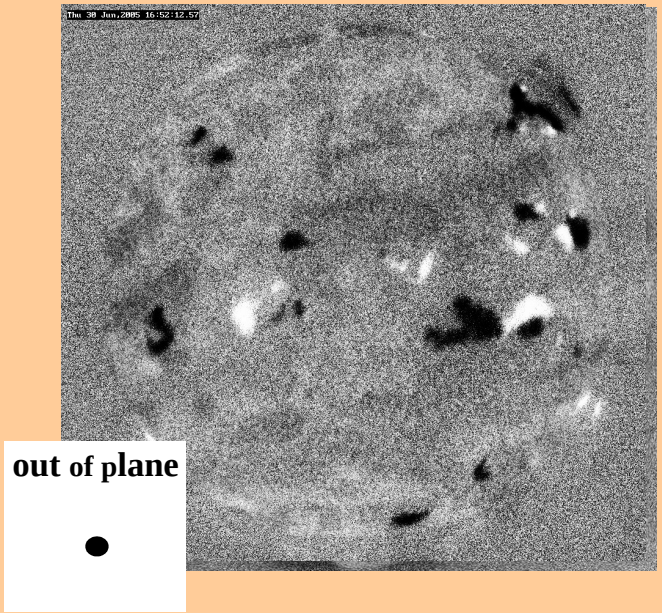
T= 240ºC, FOV= 7µm

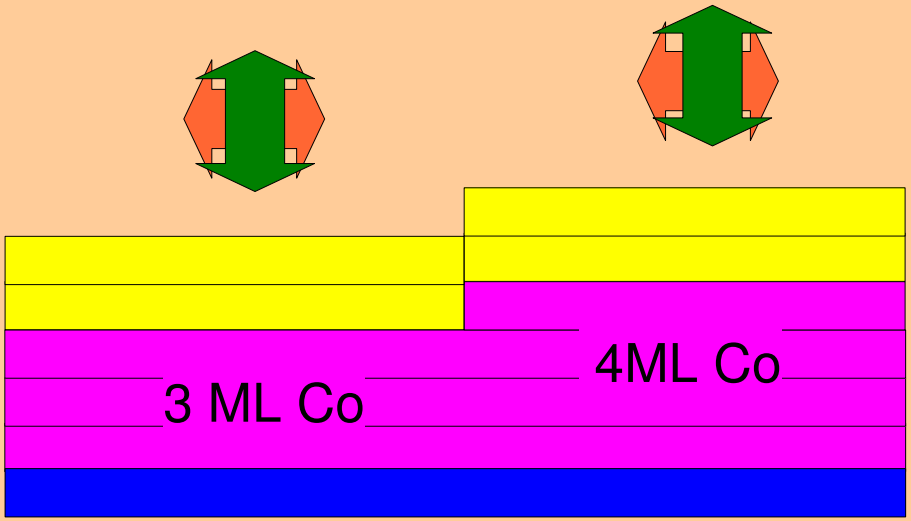
2 ML Cu / 3 & 4 ML Co / Ru

T= 240ºC, FOV= 7µm E=7eV

- The system **2 ML Cu / 3 and 4 ML Cu / Ru** is completely **in-plane** magnetized, with **Curie temperature > 540K**







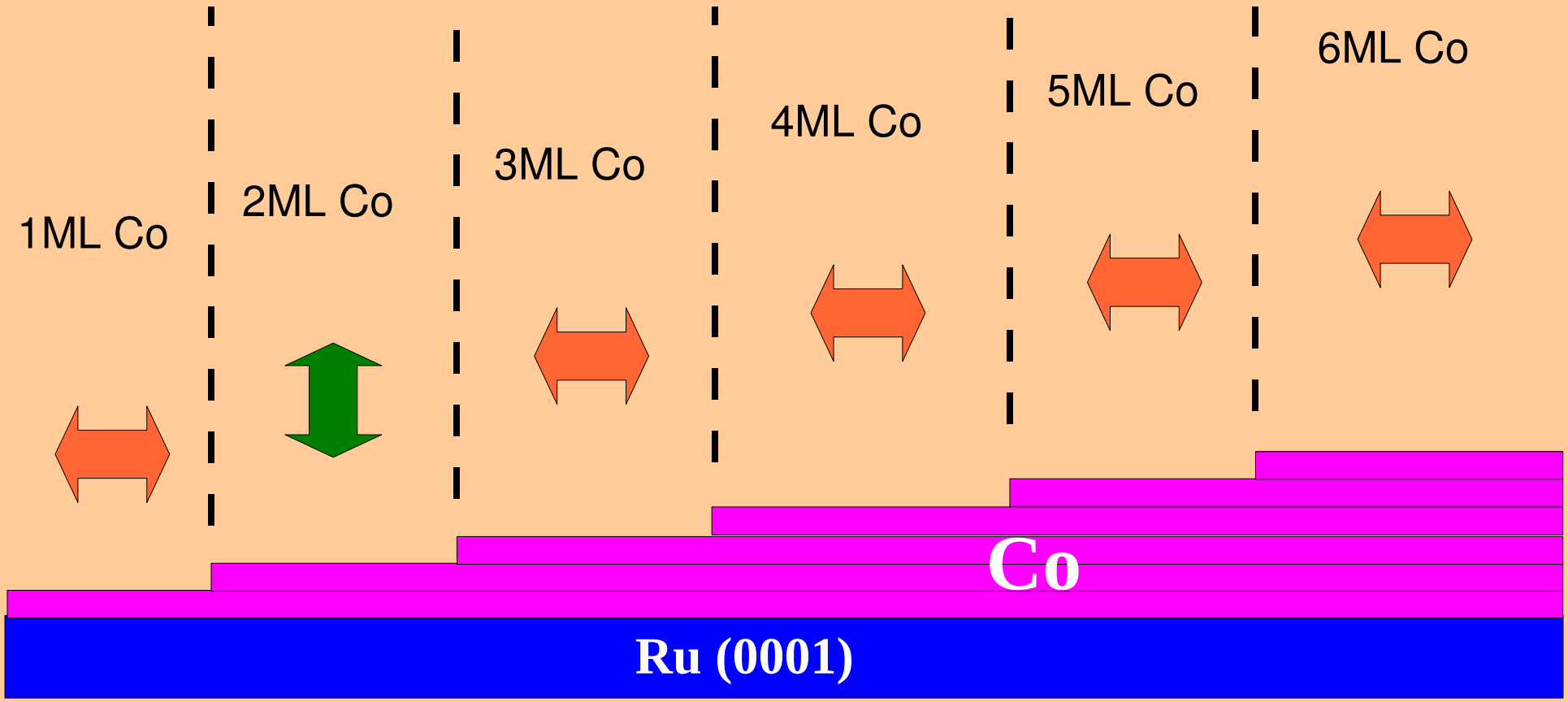
LEED 3ML Co / Ru

LEED 1ML Cu on 3ML Co / Ru

LEED 2ML Cu on 3ML Co / Ru

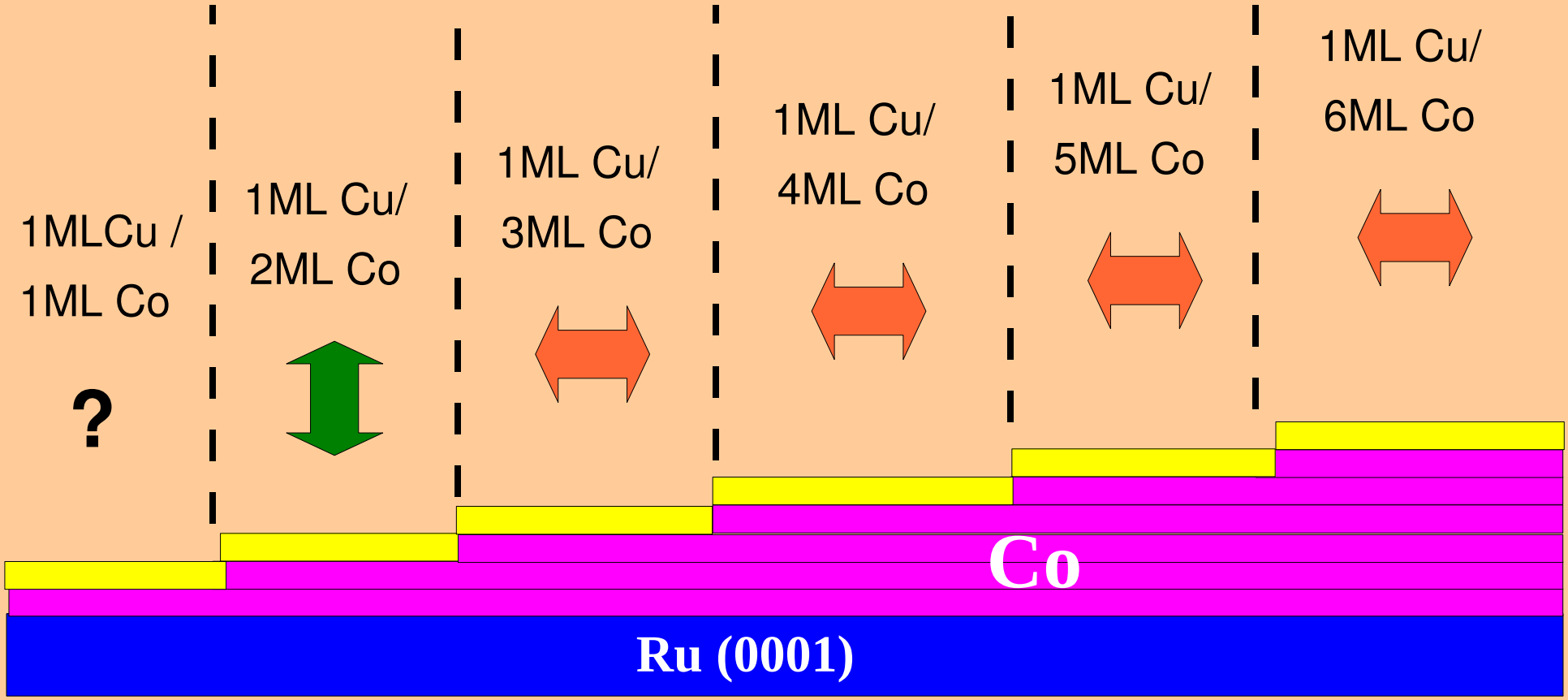
SPLEEM summary
x Co / Ru

- 1ML in-plane (Curie T=170K)
- 2ML out-of-plane
- 3ML in-plane
- 4ML in-plane
- 5ML in-plane
- 6ML in-plane



SPLEEM summary
1 ML Cu / x Co / Ru

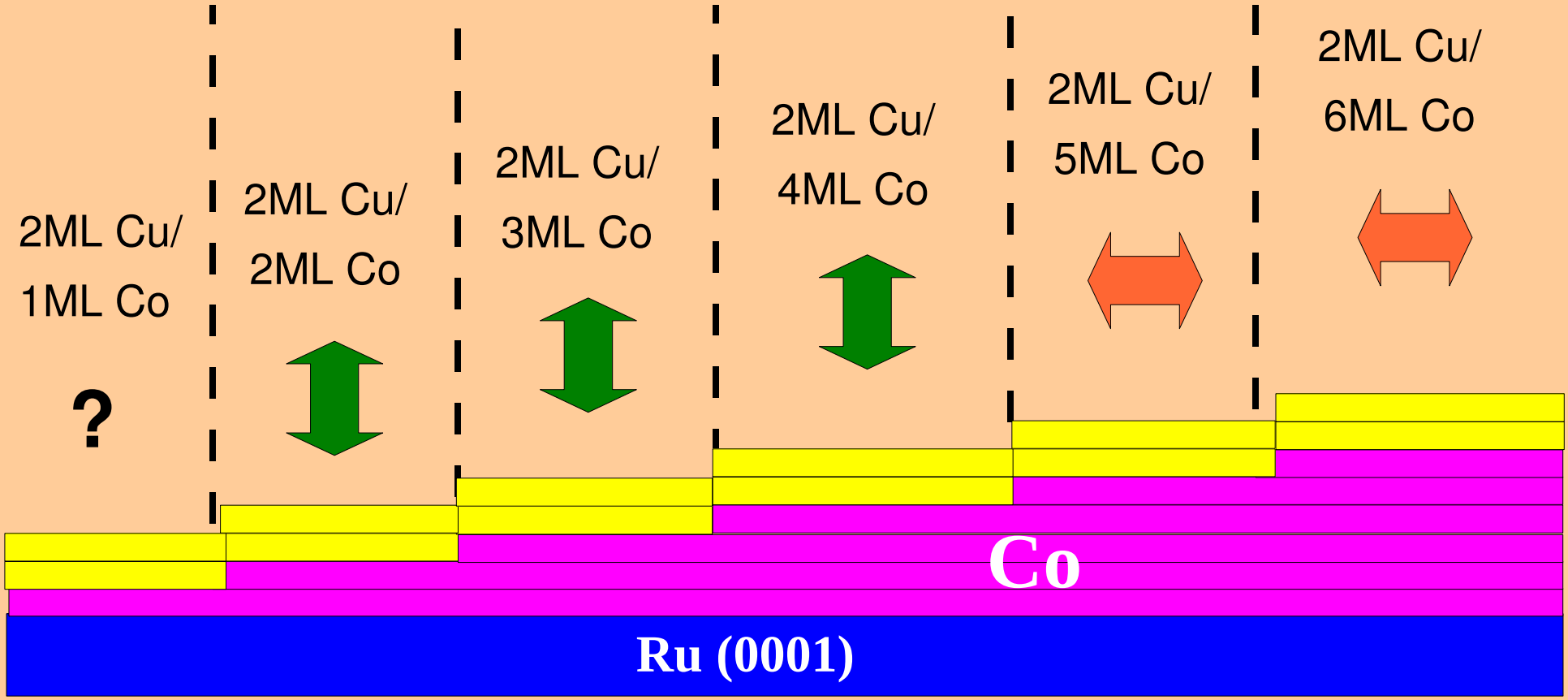
- 1ML ? (Curie T<110K)
- 2ML out-of-plane
- **3ML out-of-plane**
- **4ML out-of-plane**
- 5ML in-plane
- 6ML in-plane



SPLEEM summary
2 ML Cu / x Co / Ru

- 1ML ? (Curie T<110K)
- 2ML out-of-plane
- **3ML in-plane**

- **4ML in-plane**
- 5ML in-plane
- 6ML in-plane



Summary: Magnetization reorientations on CuCo films

- In bare ultra-thin Co films on Ru:
  - 1 ML and >2ML Co are magnetized in-plane
  - 2ML Co is magnetized out-of-plane with high Tc
- Calculations show that 1 and 3 ML are always in-plane
  - Only in 2 ML interface and surface effects added to strain can give rise to out-of-plane
- Depositing one Cu layer on top of 3 and 4 Co ML drives the magnetization out of plane (no change for the rest)
    - One more Cu layer drives it in-plane again

The most important part: the people

SPLEEM

Andreas K. Schmid

Christof Klein

LEEM

Kevin F. McCarty

Juan de la Figuera

Farid El Gabaly

THEORY

Silvia Gallego
Carmen Muñoz

Lazlo Szunyogh
Peter Weinberger

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Austria Ministry of Labour and Economy
Hungary Research Fund

http://hobbes.fmc.uam.es/loma