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Why does a thick film dewet?

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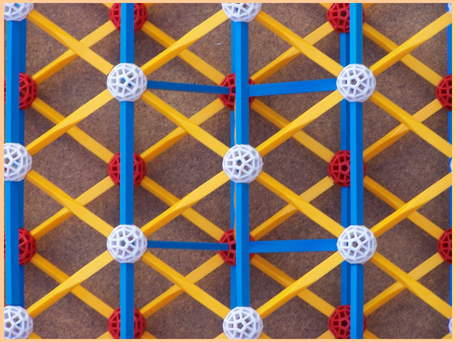
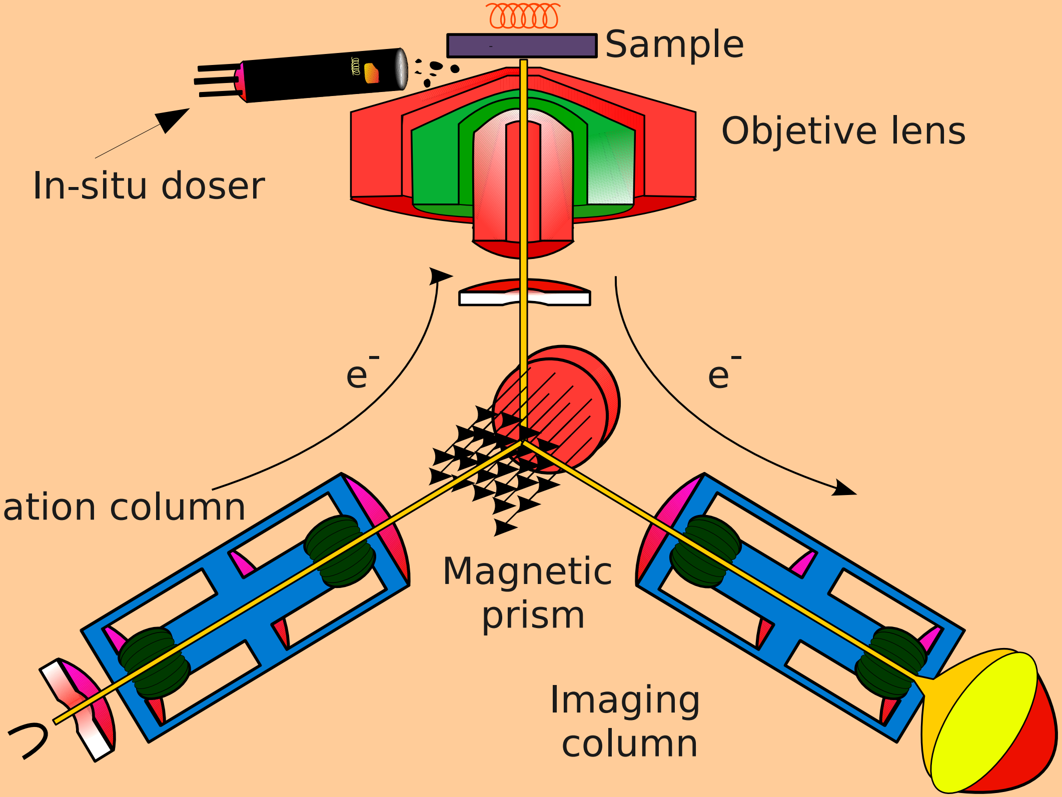
The start: the art of growing a flat film

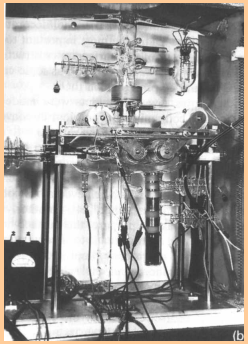
- Perfect flat films are difficult to grow by molecular beam epitaxy
  - Easiest way (if possible): grow layer-by-layer in step-flow mode
    - Step flow requires elevated temperature
    - Layer-by-layer breaks up in most films to give 3D+wetting layer growth
- Alternative way to grow a flat film:
  - Dose a thick layer at RT and anneal to improve crystallinity
- We choose Cr/W(110): Cr(110) is a nice compensated surface, spin density wave close to a c(2x2)

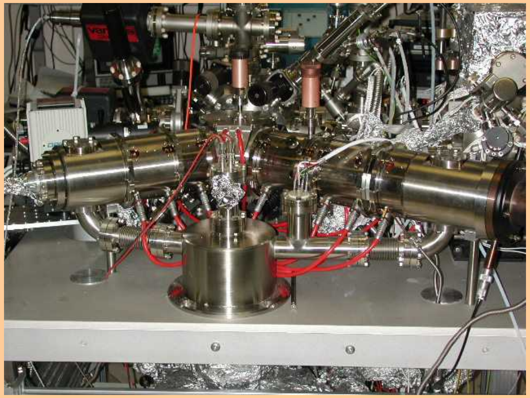
Cr : 2.91 Å
W : 3.16 Å



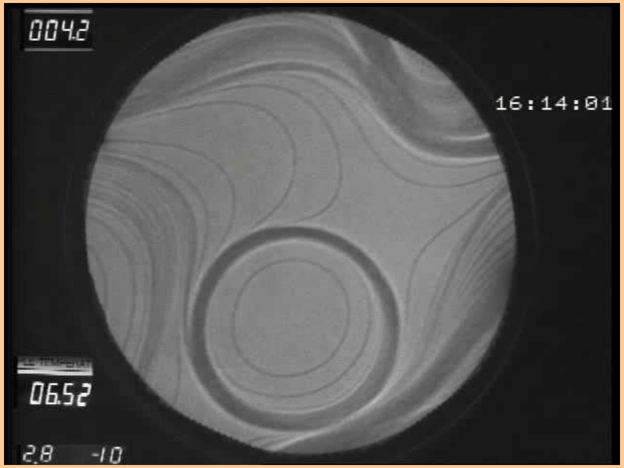
Real time imaging of growth processes: LEEM



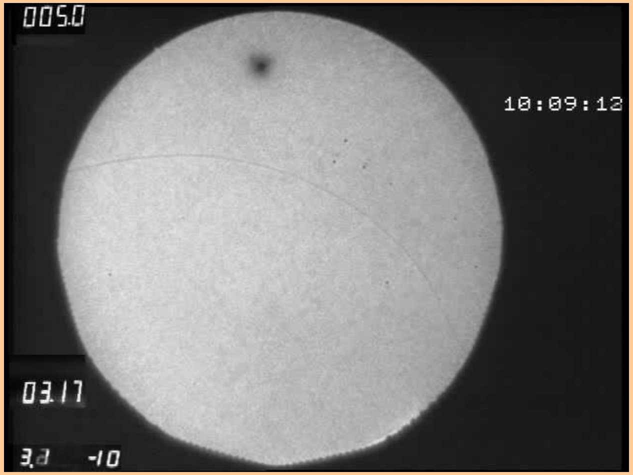




Step-flow.
Cr/W(110) FOV 7μm



Island nucleation.
Co/Ru(0001) FOV 10μm



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

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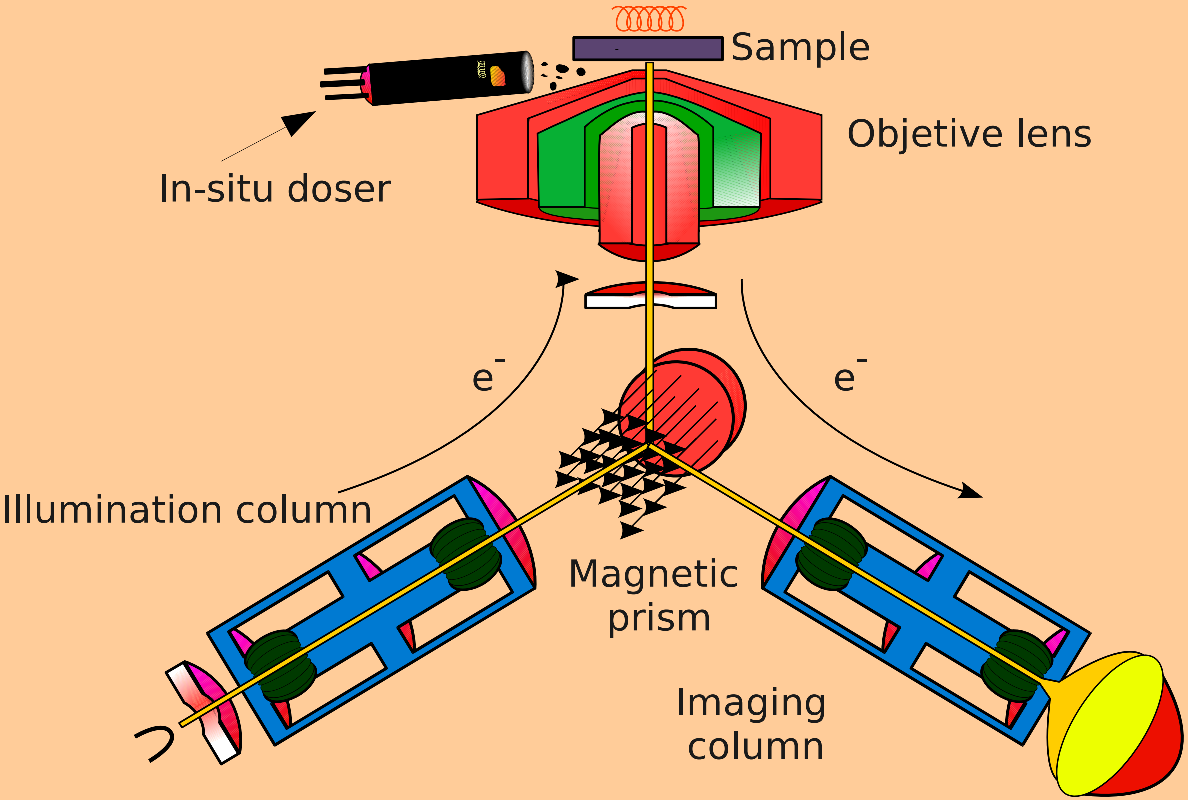

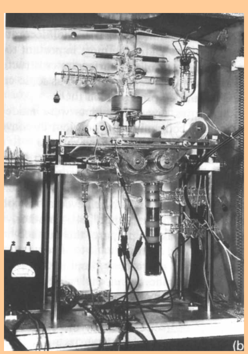

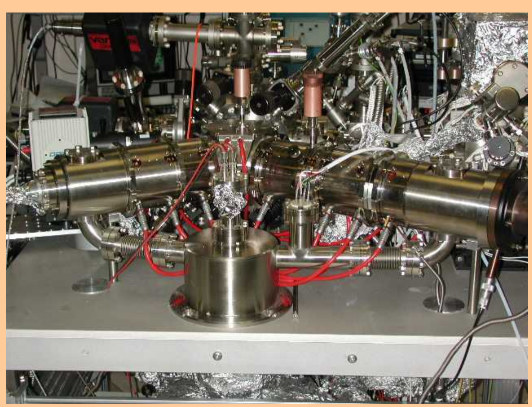

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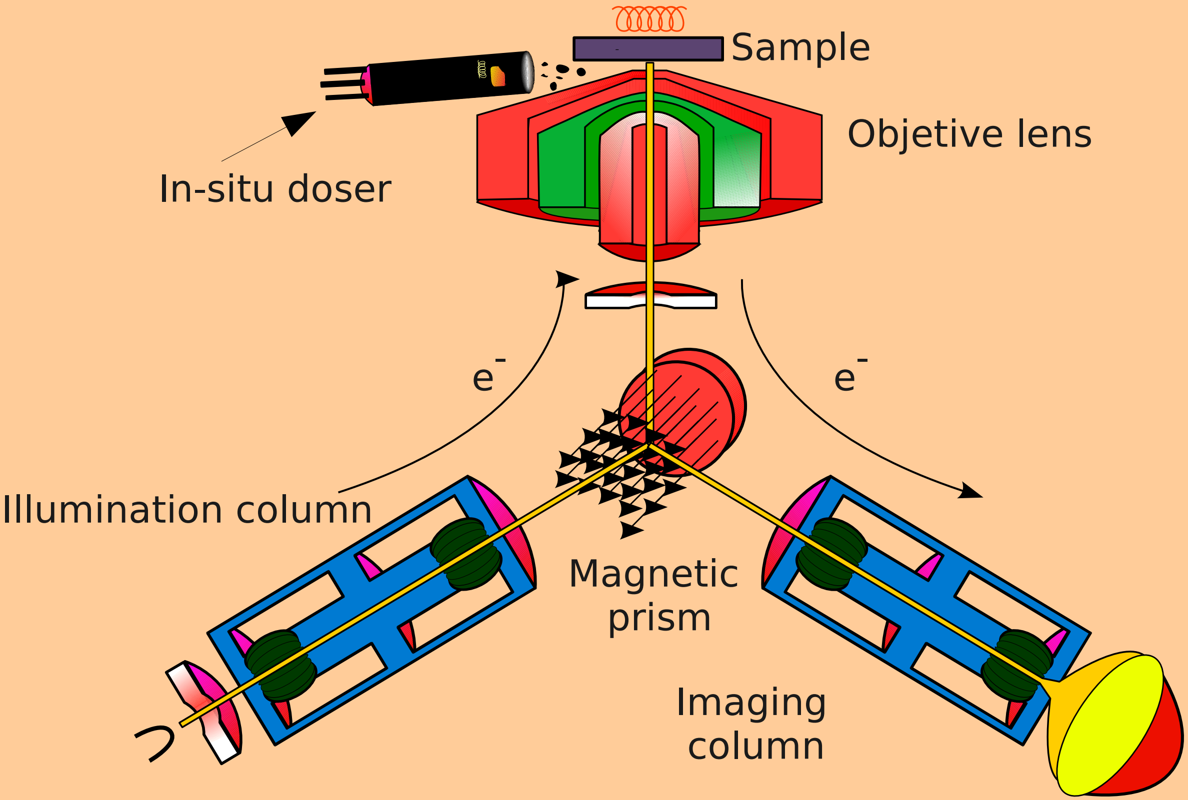

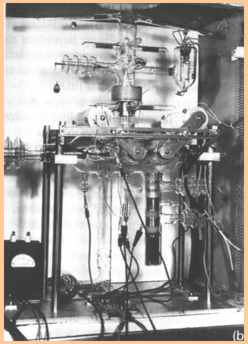

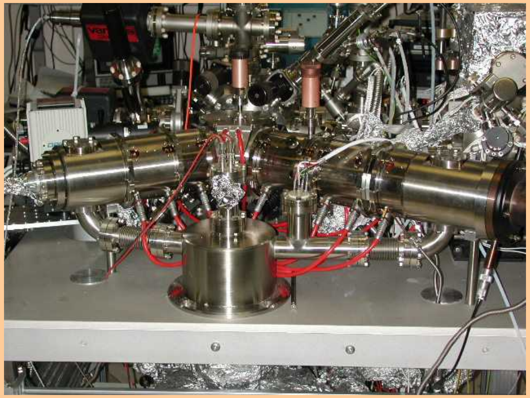

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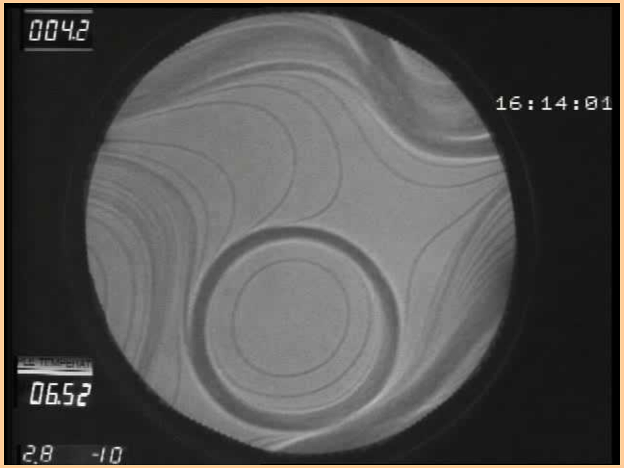

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Growing at RT+heating

- Depositing Cr at RT: very disordered
  - continuous decrease of reflected electrons
  - no LEED pattern
- Annealing
  - Increase of reflected intensity until W(110) features can be detected ("conformal" film)

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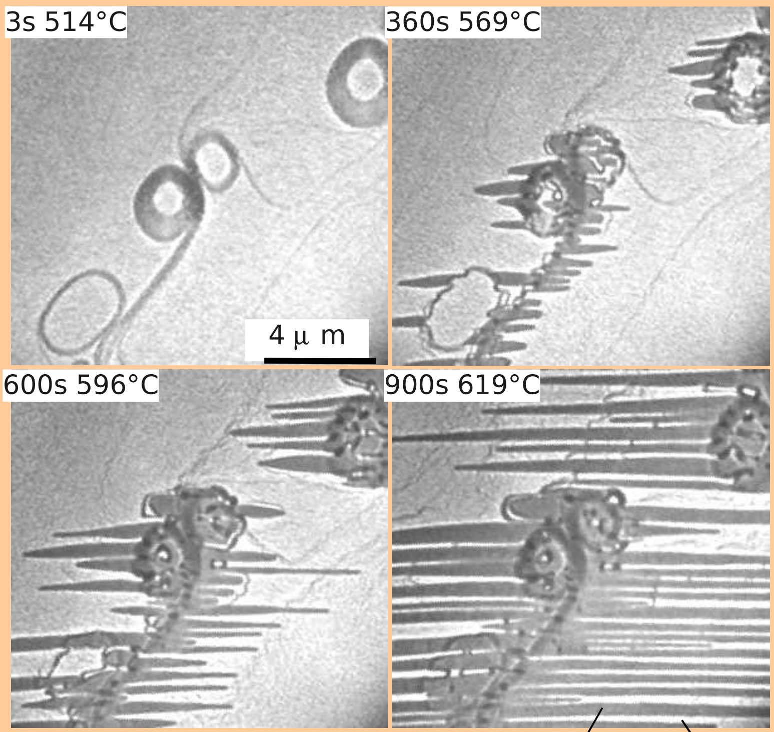
Trenches that expose the wetting layer nucleate at bunches of *substrate* steps



Trench that exposes the wetting layer

Cr stripe

100906ashort.ppt
300x300 pixel 20 um FOV
Frames 1, 120 200 300 3s/f 40ML
8.93 mV 514C, 10.0mv 569C, 10.53 mv 596C, 10.98mv 619C

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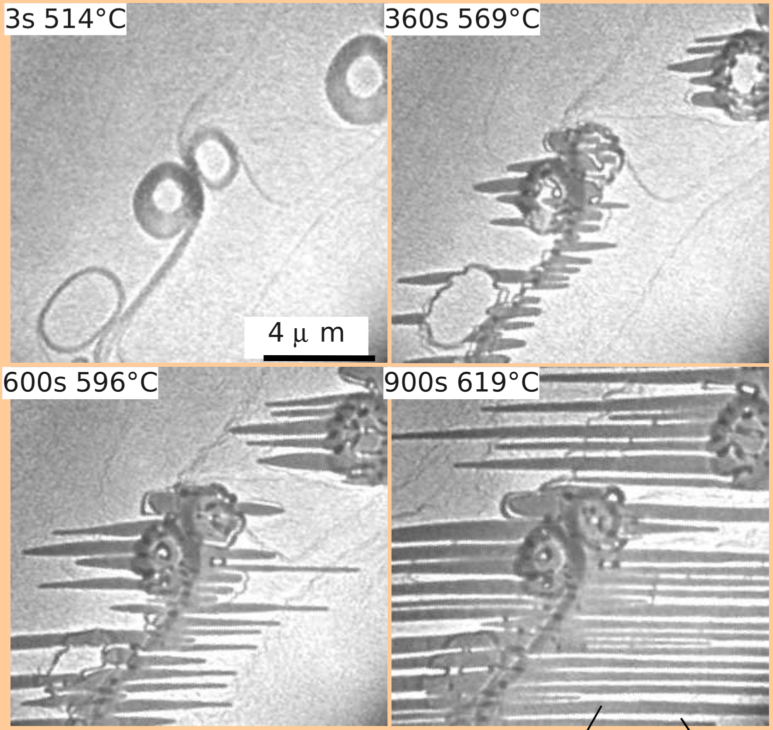

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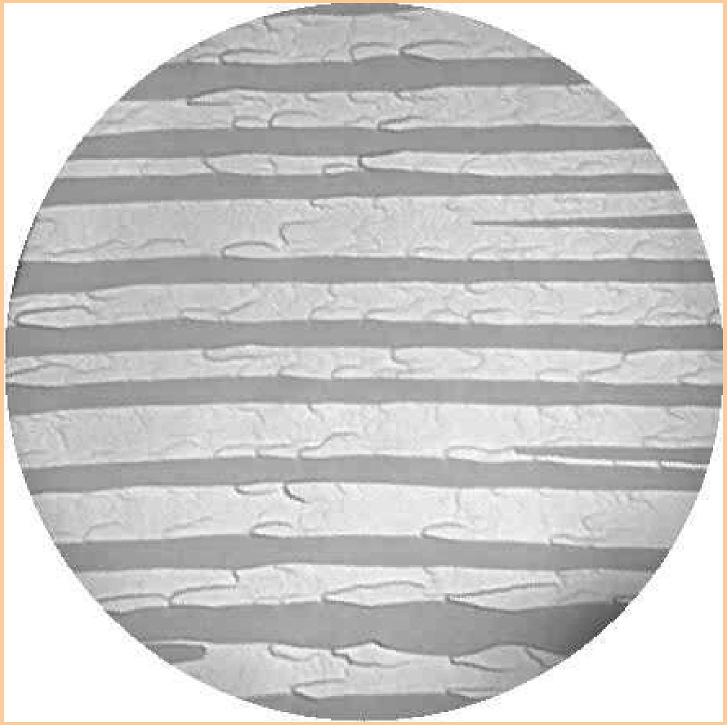
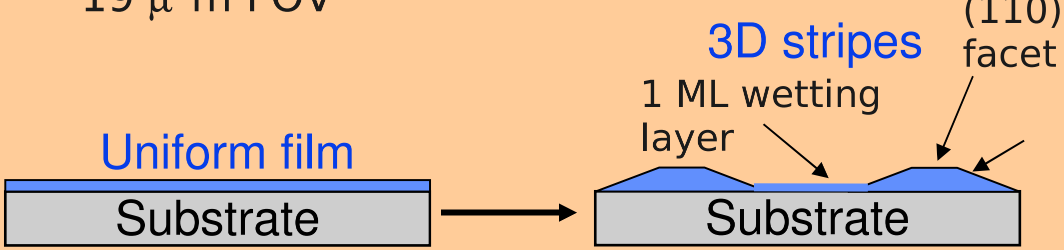
Cr stripes thicken and narrow with annealing

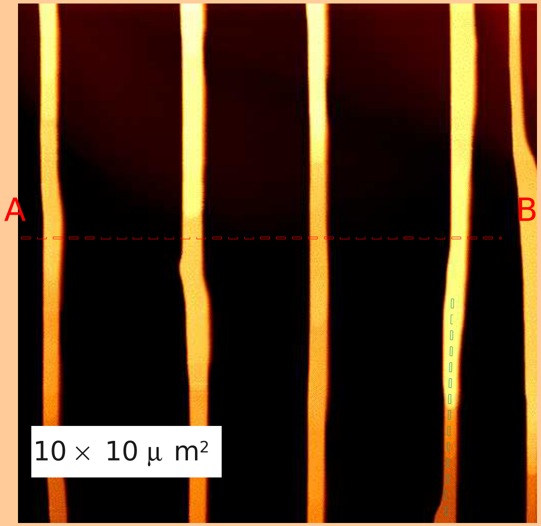


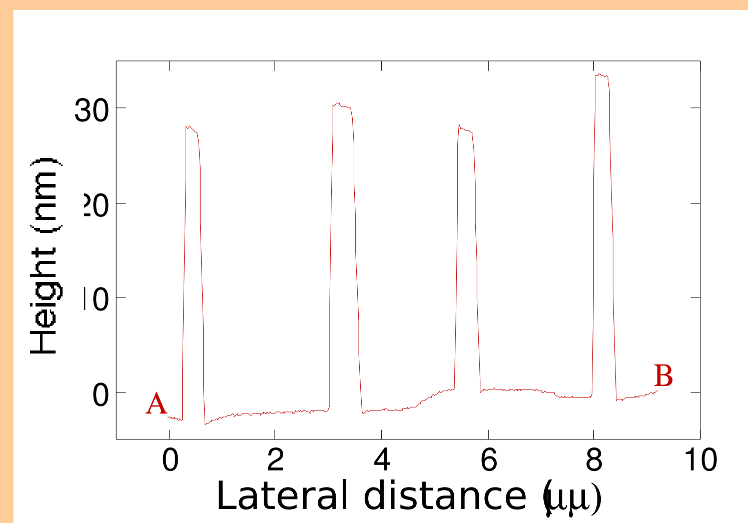
19 μ m FOV



120406n.ppt
440x440 pixel 20 um FOV
11.0 mV 620C
8 s/f
22 ML
Frame 1 & 339

30ML Cr on W(110)





AFM courtesy of Frank Jones, SNL

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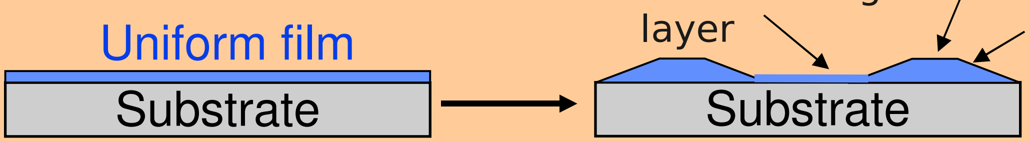

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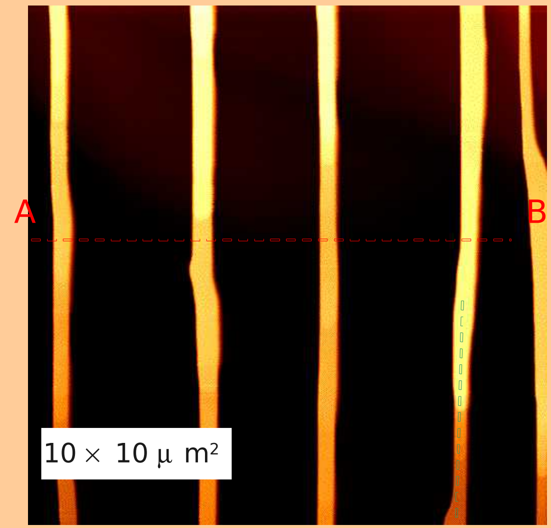
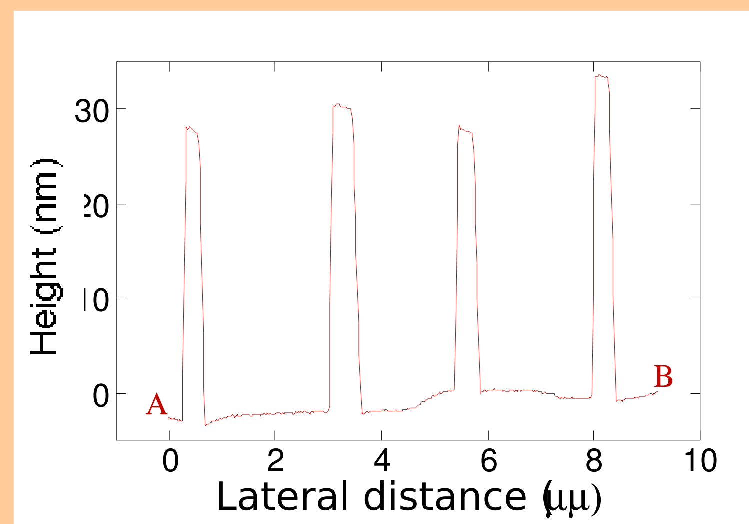
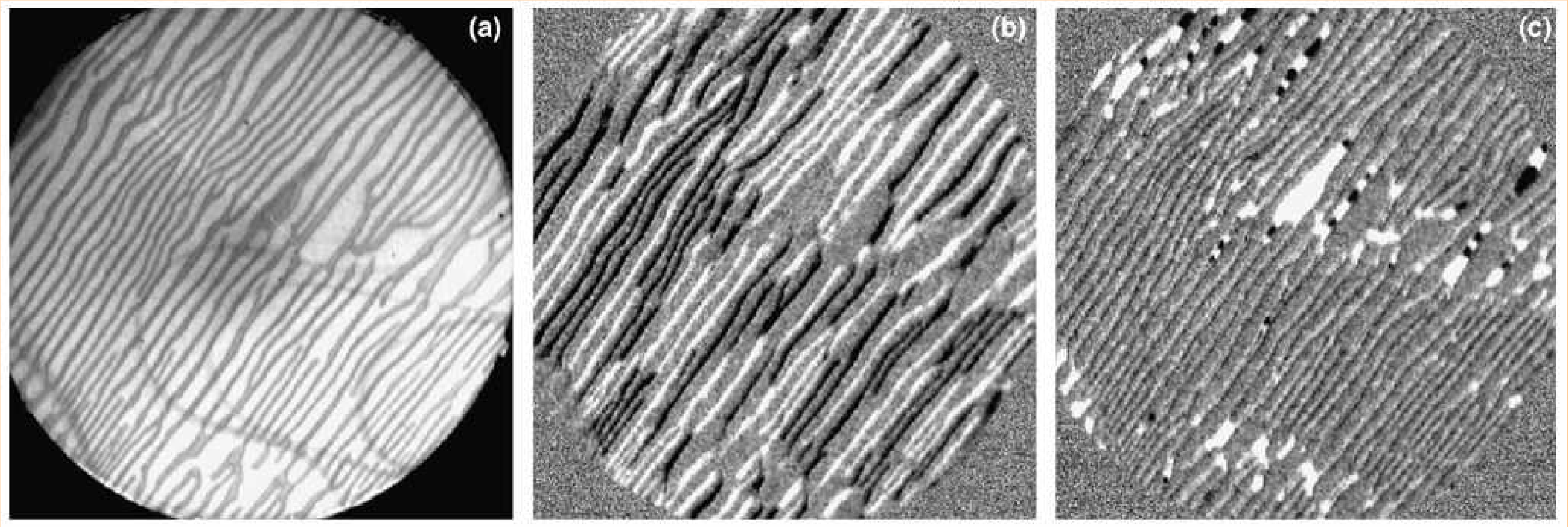




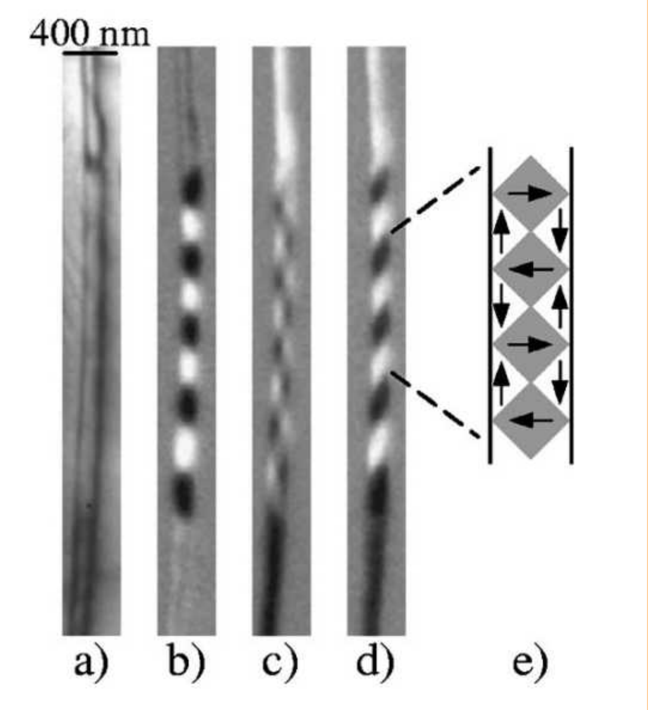
AFM courtesy of Frank Jones, SNL

Magnetism in wires? Fe/W



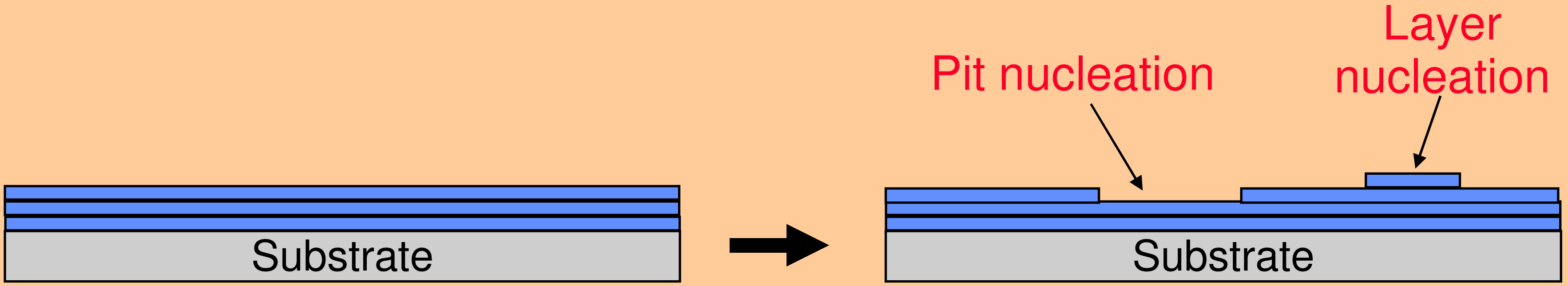
Fe also forms stripes on W(110)

- *Self-organized Fe nanostructures on W(110)*, R. Zdyb, A. Pavlovska, M. Jałochowski, E. Bauer, *Surf. Sci.* **600** (2006) 1586–1591
- *Self-organization and magnetic domain microstructure of Fe nanowire arrays*, N. Rougemaille and A. K. Schmid, *J. App. Phys.* **99** (2006) 08S502



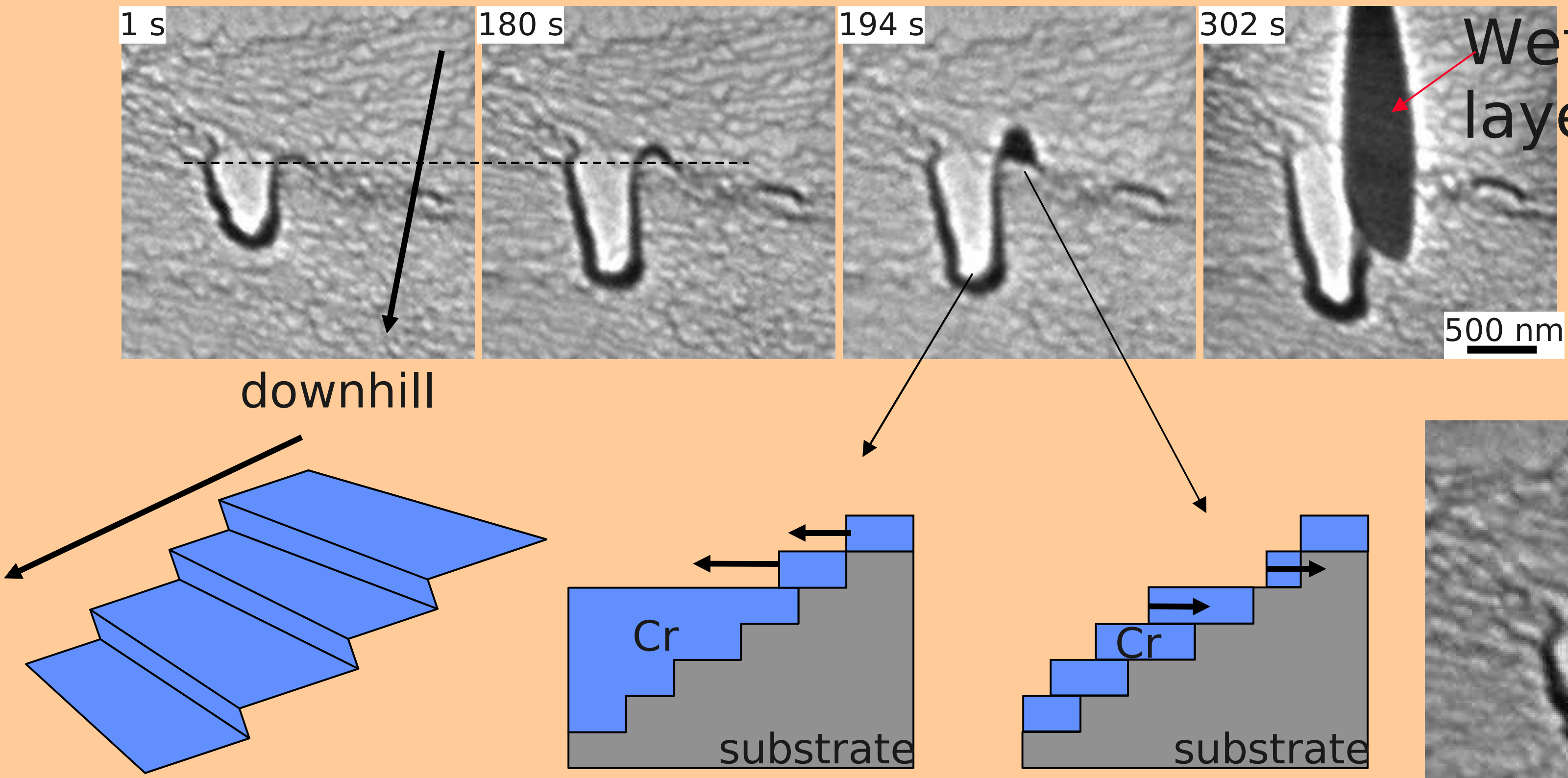
I- How do flat films evolve into 3D islands? II- Why do they evolve?

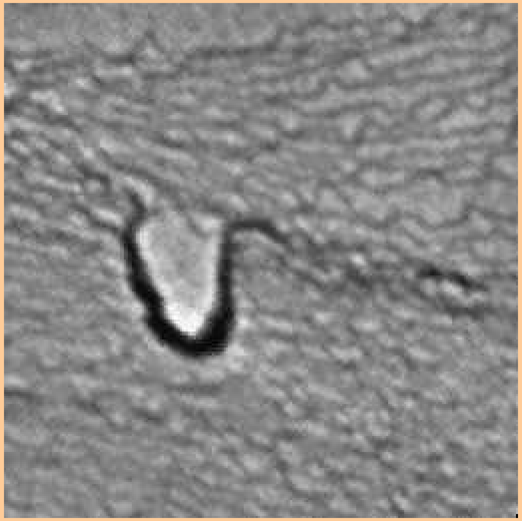
- Flat films frequently are unstable relative to 3D islands
  - Initially flat film de-wets, making 3D islands
- How does a uniform, defect-free film evolve into 3D islands?
  - Thinning & thickening processes both seem to require costly nucleation events
    - Mullins & Rohrer, J. Am. Ceram. Soc. 83 (2000) 214.



Trenches nucleate by film steps retracting uphill

Adjacent film steps move in opposite directions





$4.5 \times 4.5\ \mu m^2$

121506l.ppt
256x256 pixel 4.5 um FOV
9.1 mV = 523C
Frames 1, 90, 97, 151
2 s/f

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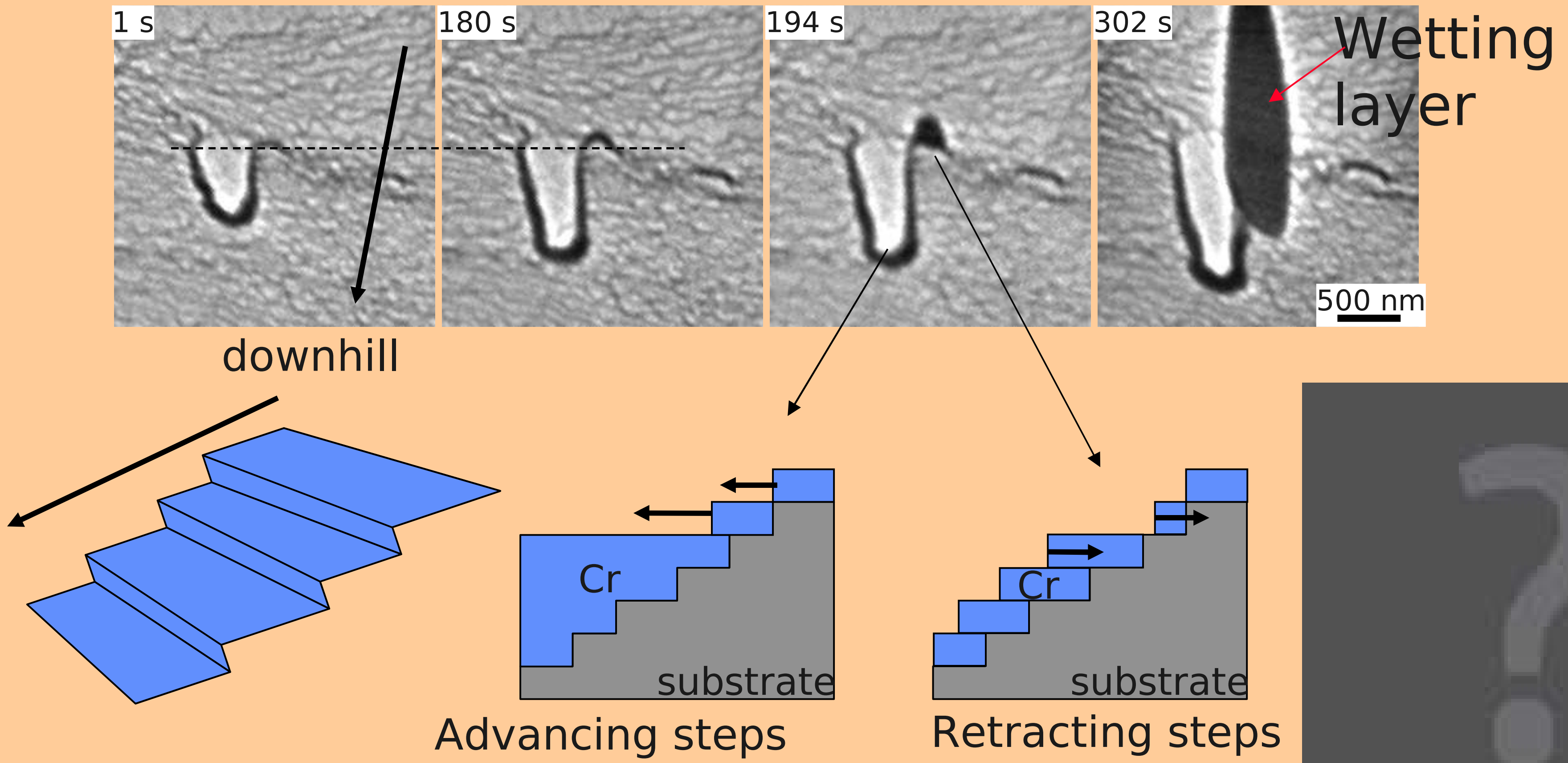

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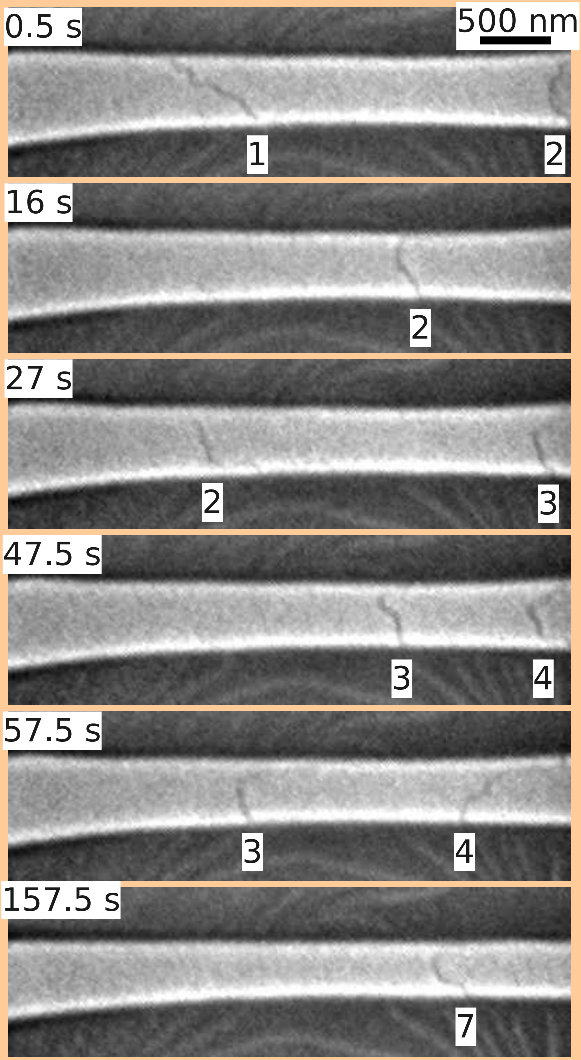
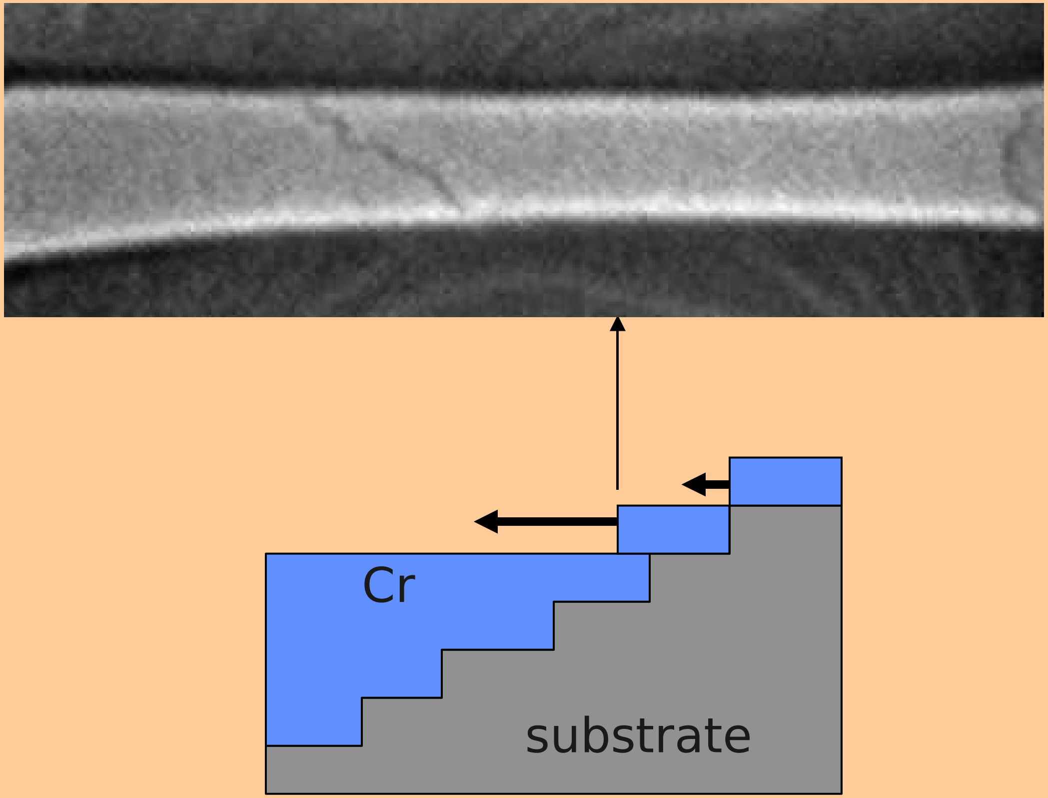
Cr stripes thicken by steps flowing down the staircase of substrate steps



121506g.ppt
400x120 pixel 4.5 um FOV
11.2 mV = 630
Frames 1, 32, 54, 95, 115, 315
0.5 s/f



Analogous to "downhill migration" mechanism of island thickening

- W. L. Ling, T. Giessel, K. Thürmer, R. Q. Hwang, N.C. Bartelt and K. F. McCarty, *Crucial role of substrate steps in de-wetting of crystalline thin films*, Surface Science **570** (2004) L297.

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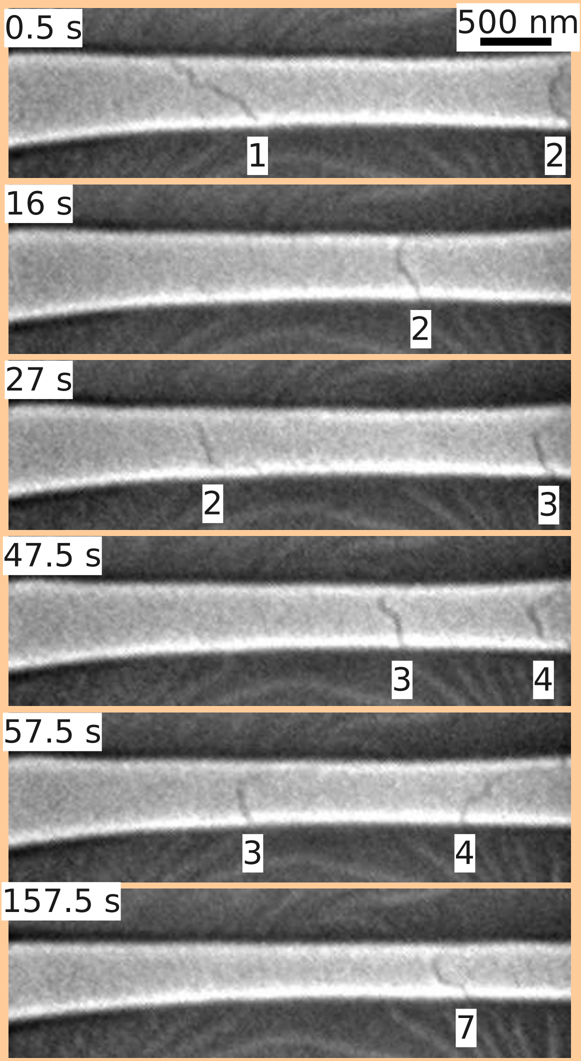

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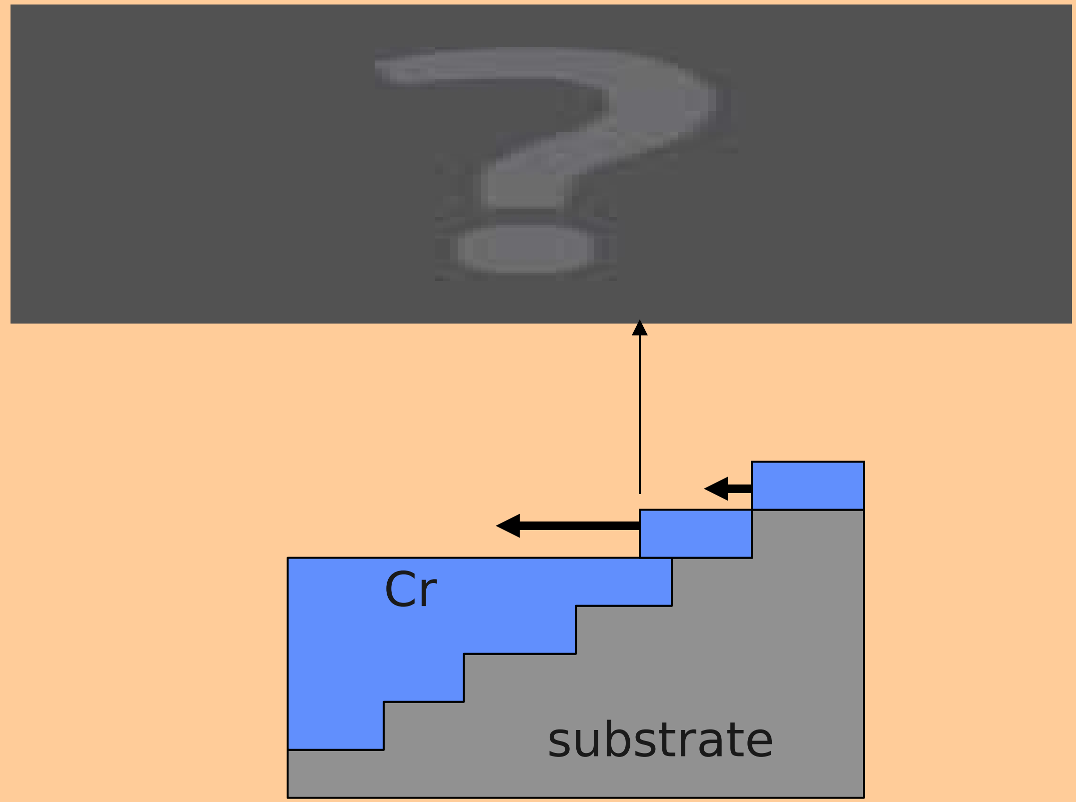

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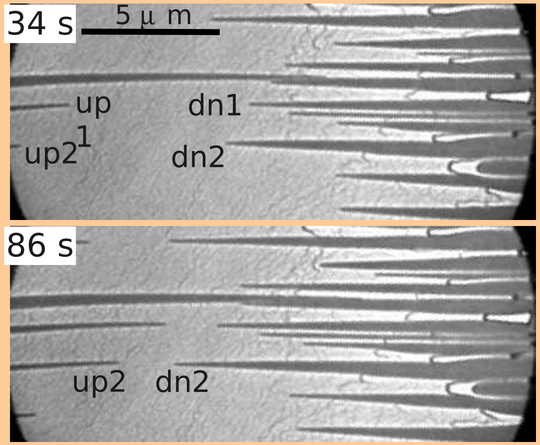
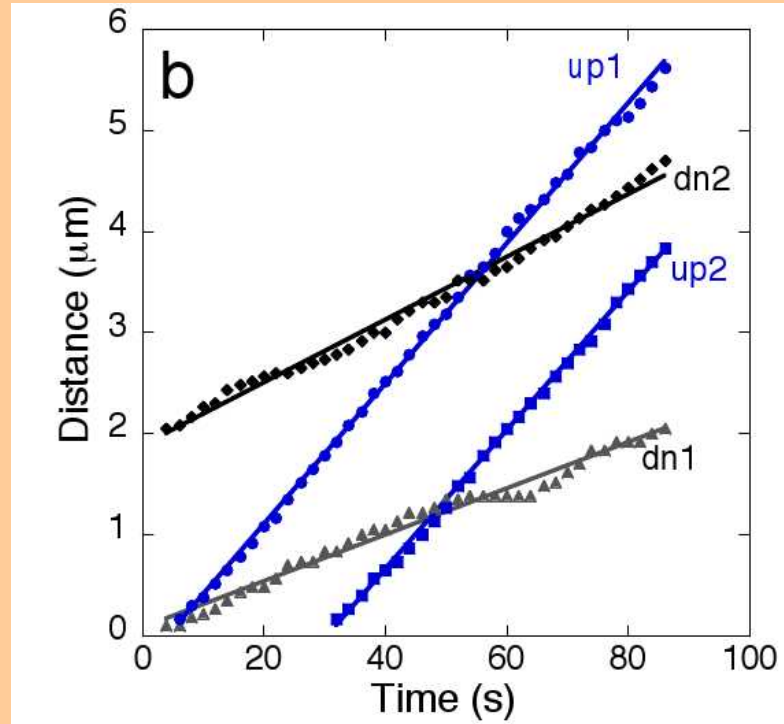
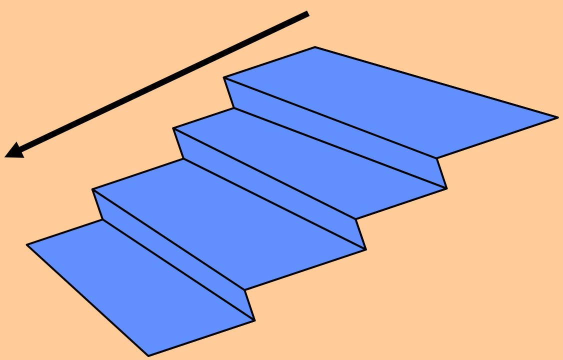
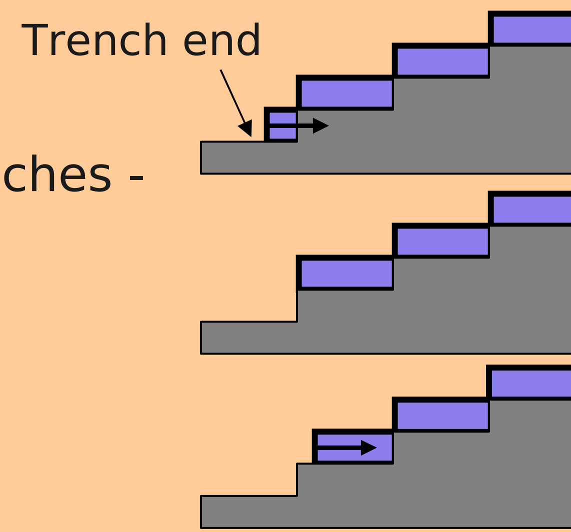
Trenches move faster uphill than downhill





downhill

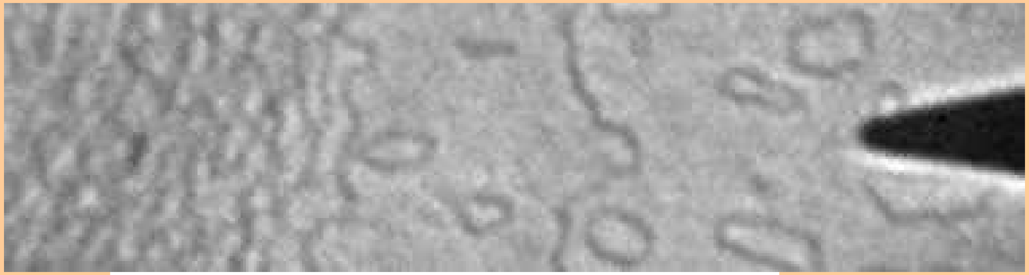


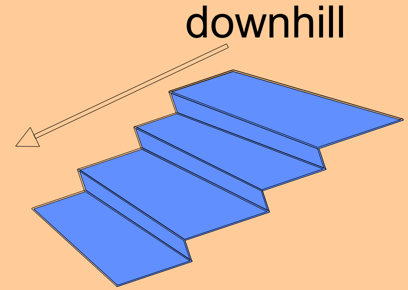


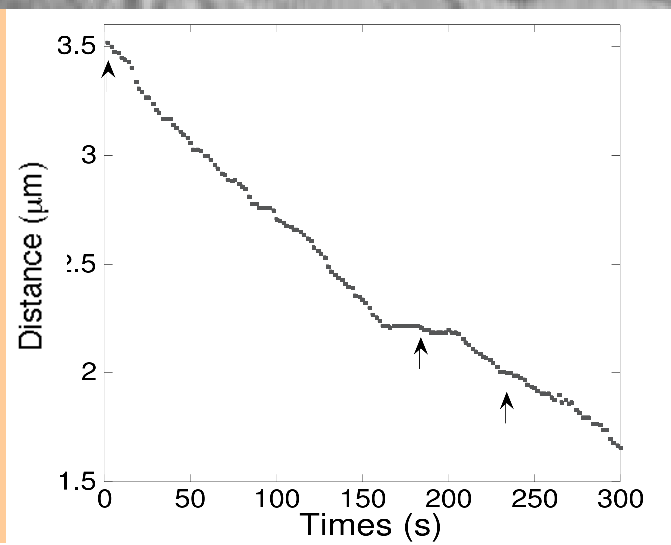
Uphill moving trenches -
no pit nucleation
needed

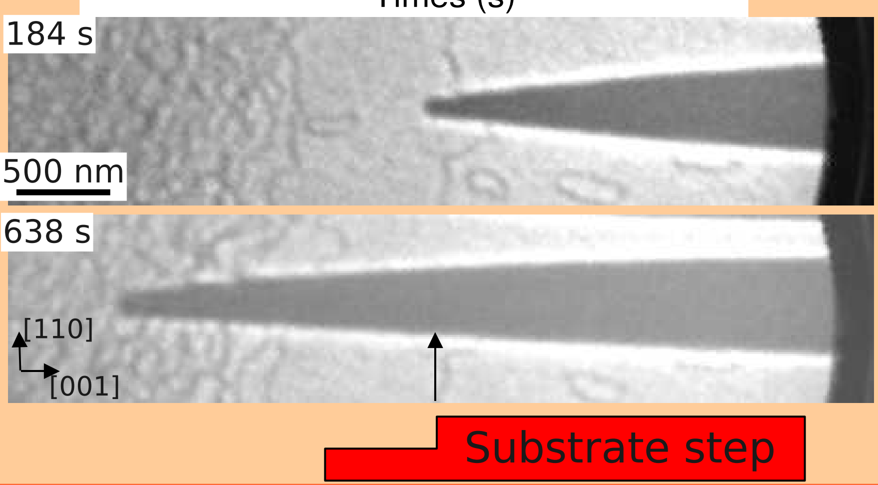
102506e.ppt Uphill vs. downhill trench velocity
2s/f 20um FOV 102506e_crop is frames 289-331 of 1024506e.mov
10.47 mv = 593C frames 2, 17, 30, 43 of crop, 440x180 pixels
22ml

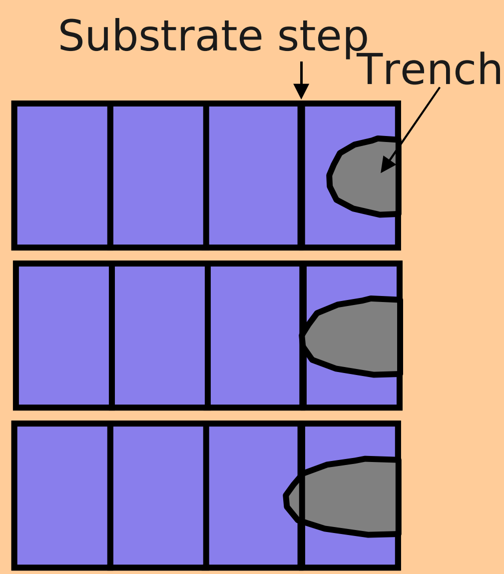
Trenches moving downhill get stuck at descending substrate steps

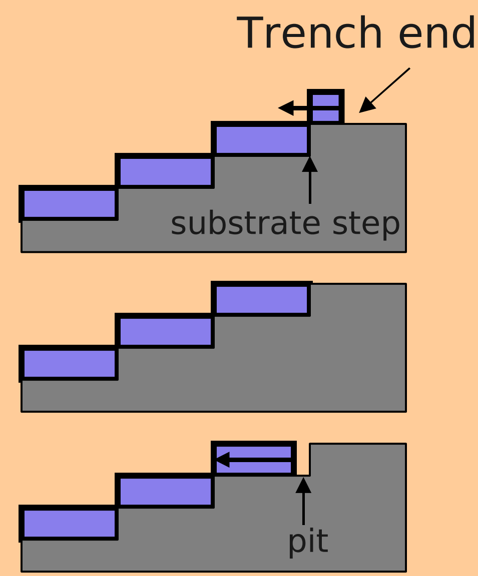












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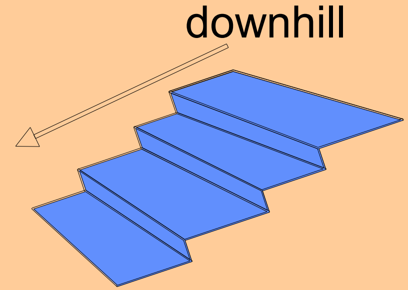
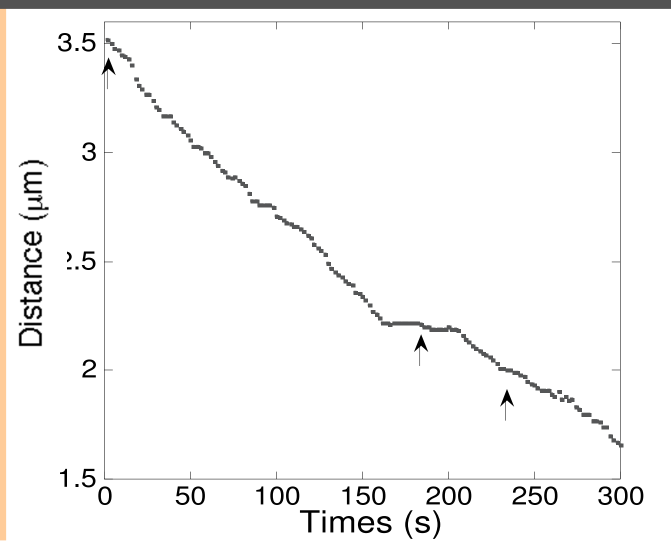
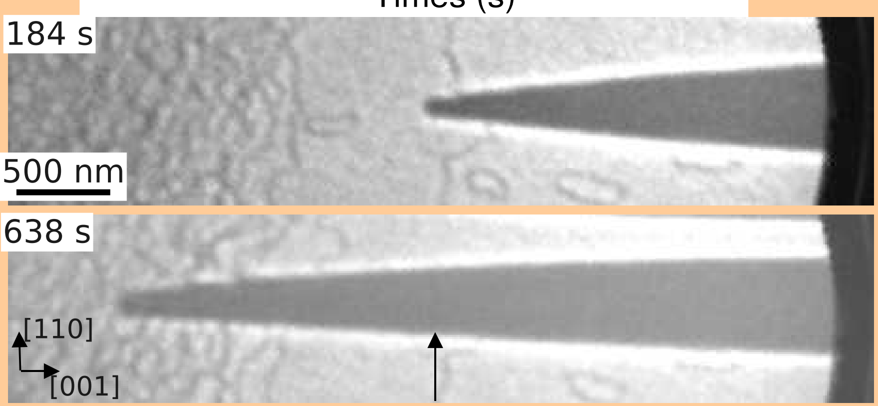
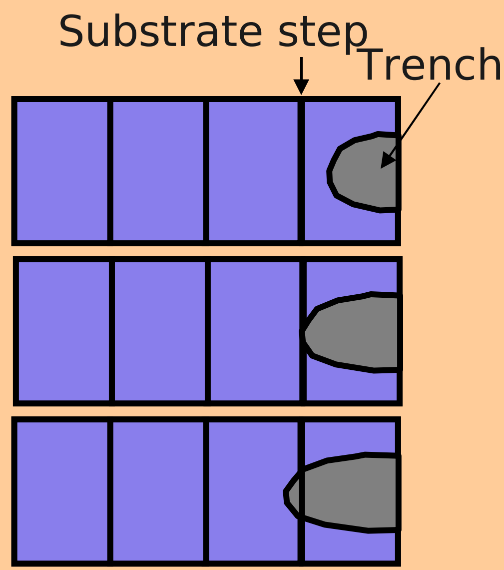
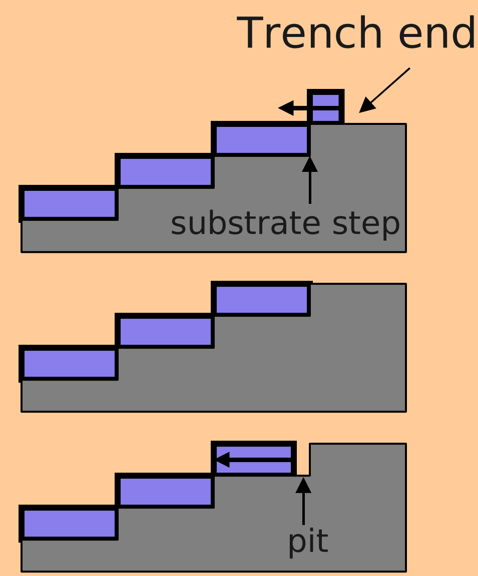


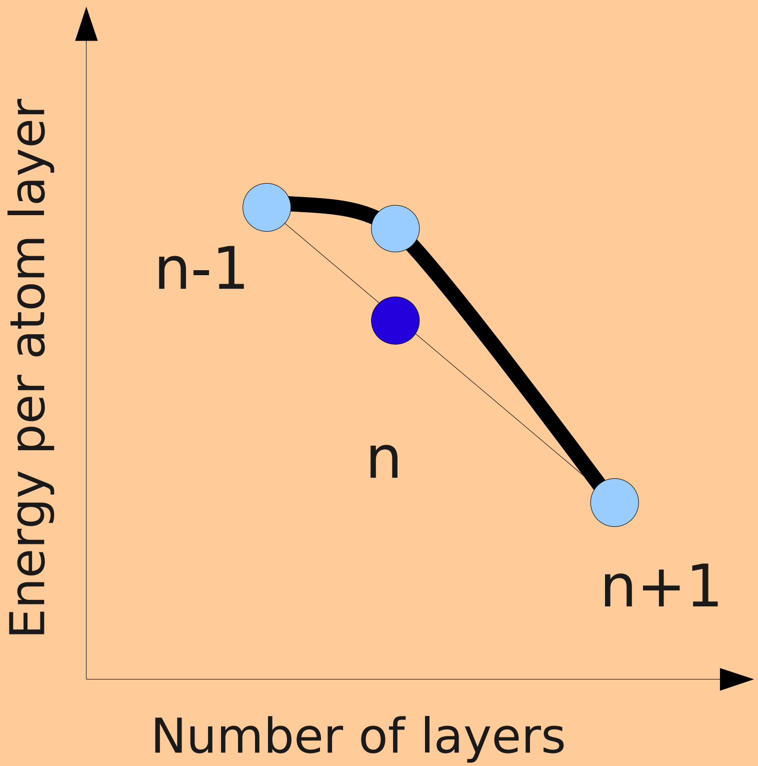






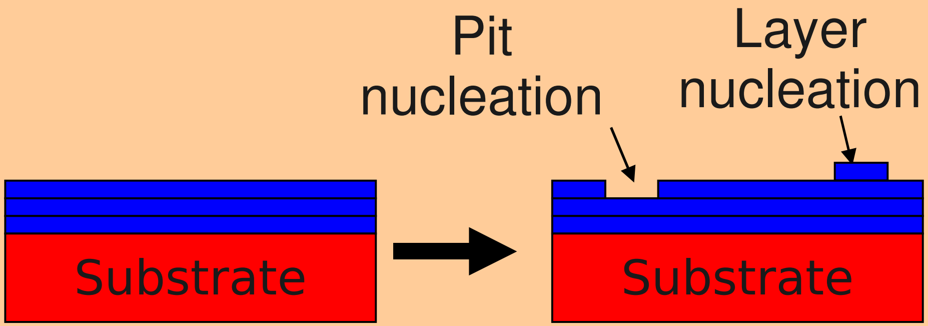


Why does the film break up?



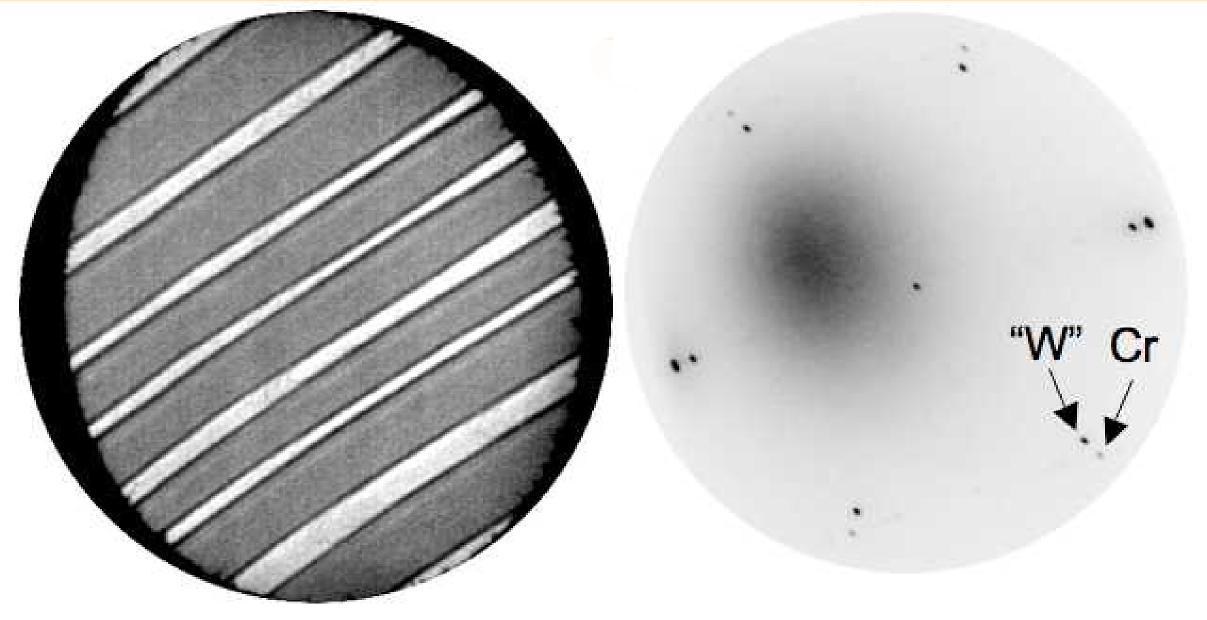
$$E_{n-1} + E_{n+1} - 2E_n < 0$$

$$\frac{d^2 E_n}{dn^2} < 0$$

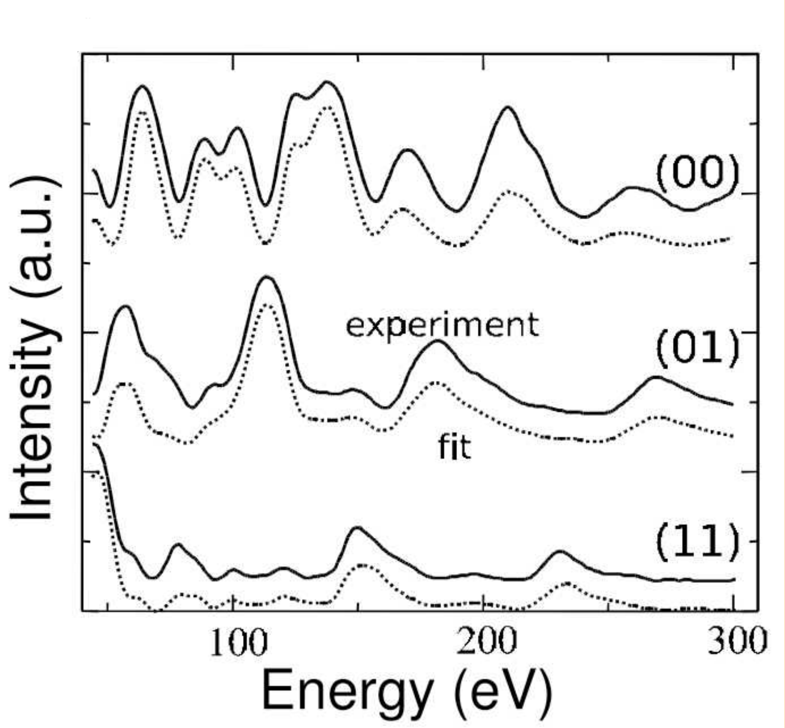


- Leaving aside kinetic limitations (which moving material downhill avoids), why the film wants to roughen in the first place?
- The energy per atom curve must have a convex shape
- In the ultra thin film case, it can be understood by comparing the wetting layer with a thicker film, but in a reasonably uniform thick film?
  - Strain?

Thick Cr films are not strained by the substrate



LEED IV on the thick Cr areas indicate that the films have bulk Cr lattice parameter, with the expected surface layer relaxation



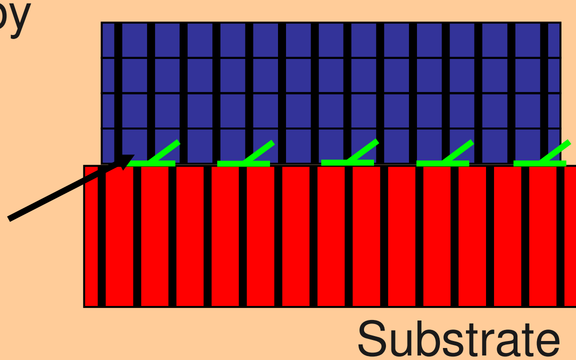
Benito Santos et al., *Structure and magnetism of ultra-thin chromium layers on W(110)*, New J. Phys. **10** (2008) 013005

Atomistic simulations confirm this idea

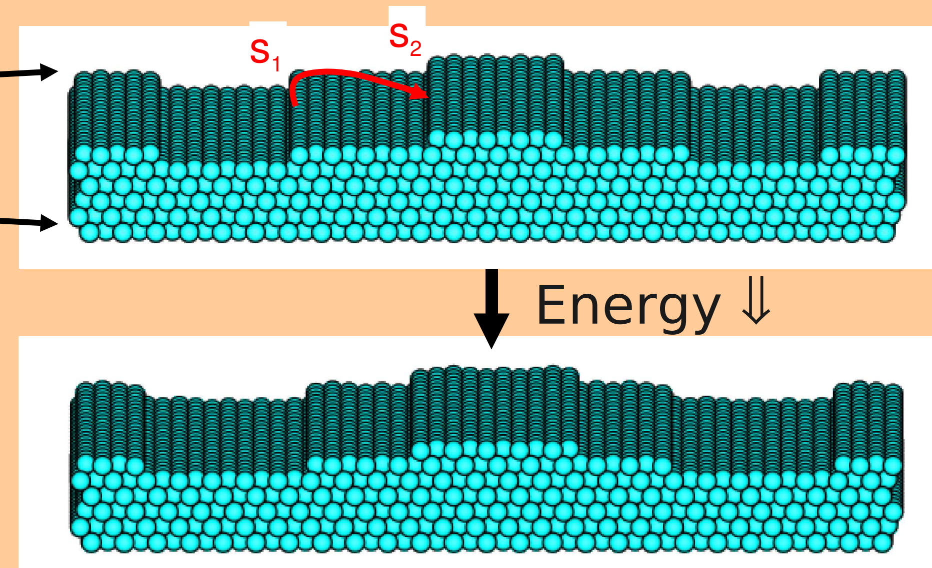
Even though they are not lattice matched to the substrate

Cr not strained by the substrate

Network of interfacial dislocations



Consider film as slab decoupled from substrate

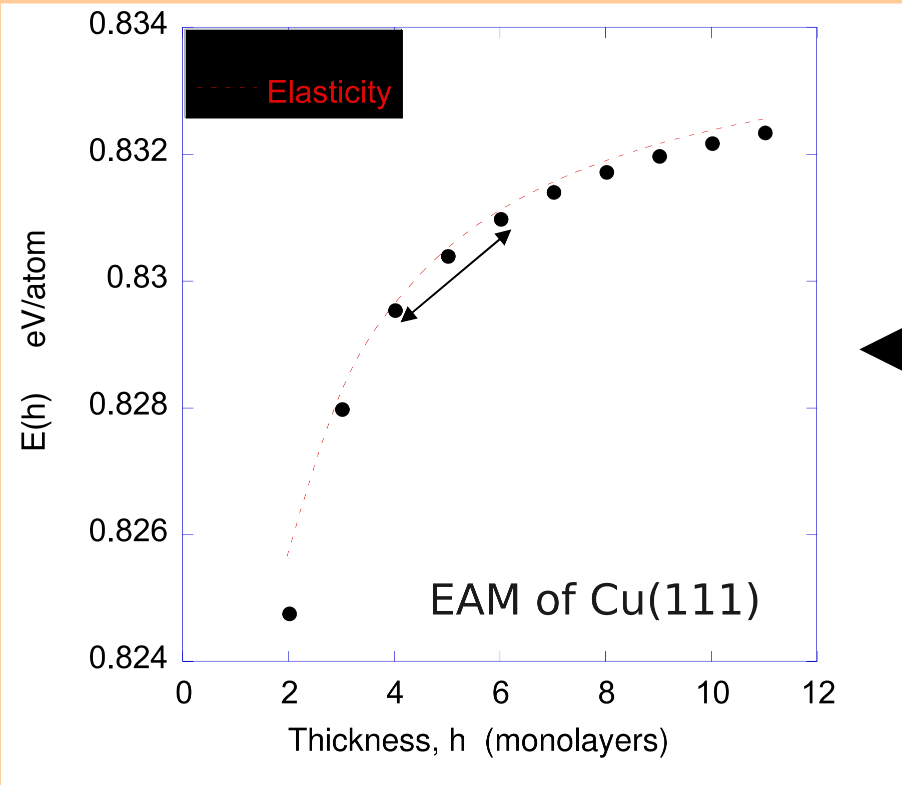


$E(h)$ = film energy/area

For the observed mass flow from step s_1 to s_2:

$$\frac{d^2E}{dh^2} < 0$$

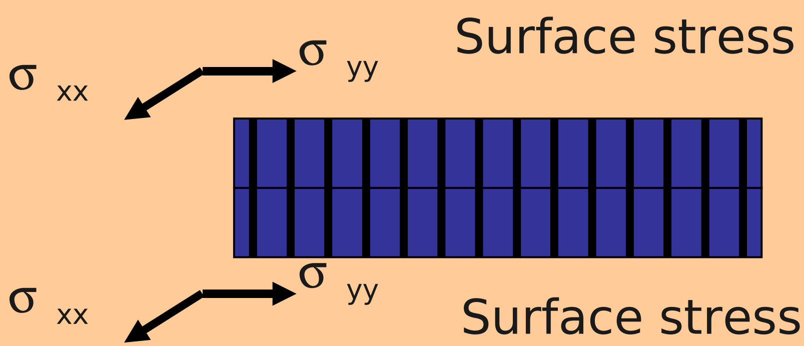
Surface stress provides a driving force to de-wet films not strained by substrates



- Energy/area *increases* with slab thickness

$$\frac{d^2E}{dh^2} < 0$$

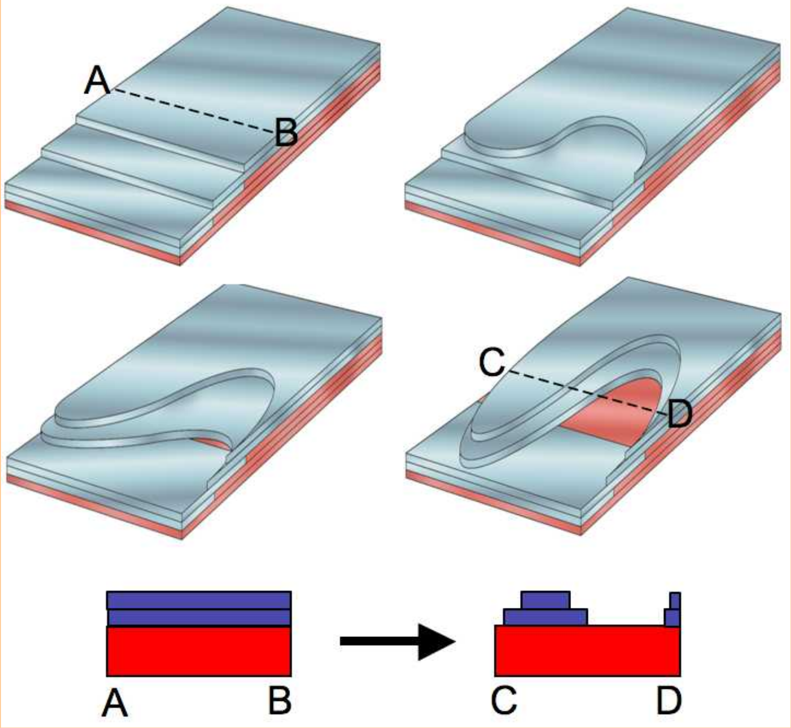
e.g., 5-layer film lowers energy by making 4- and 6-layer regions

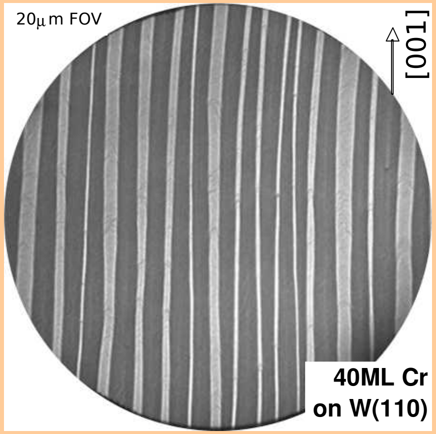


- Surface stress strains the entire slab in-plane
  - Thin slabs are strained the most

Summary of film de-wetting: Cr/W(110)

- How:
  - dewetting starts at step bunches
  - Along [100] directions
  - By downhill transport of Cr
    - avoid nucleating steps
  - De-wetting occurs by cooperative motion of film steps relative to substrate steps
- Why:
  - Wetting layer has lower energy that thicker films
  - Surface stress and a relaxed thin film make $E_{n+1}+E_{n-1}-2E_n<0$





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