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)

\[
\begin{align*}
\text{Cr} & : 2.91 \text{ Å} \\
\text{W} & : 3.16 \text{ Å}
\end{align*}
\]
Real time imaging of growth processes: LEEM

Step-flow. Cr/W(110) FOV 7µm

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


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Real time imaging of growth process: LEEM


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Real time imaging of growth process: LEEM


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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.
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Cr stripes thicken and narrow with annealing

30ML Cr on W(110)

19 µm FOV

Uniform film

Substrate

3D stripes

1 ML wetting layer

(110) facet

AFM courtesy of Frank Jones, SNL

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Cr stripes thicken and narrow with annealing

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Uniform film

1 ML wetting layer

(110) facet

19 µm FOV

10 × 10 µm²

Frame 1 & 339

AFM courtesy of Frank Jones, SNL
Magnetism in wires? Fe/W

Fe also forms stripes on W(110)

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
Trenches nucleate by film steps retracting uphill

Adjacent film steps move in opposite directions

Advancing steps

Retracting steps

Wetting layer

4.5 x 4.5 µm²
Trenches nucleate by film steps retracting uphill

Adjacent film steps move in opposite directions

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

Analogous to “downhill migration” mechanism of island thickening

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Analogous to “downhill migration” mechanism of island thickening

Trenches move faster uphill than downhill

Uphill moving trenches - no pit nucleation needed
Trenches moving downhill get stuck at descending substrate steps.

Trenches moving downhill get stuck at descending substrate steps.
Trenches moving downhill get stuck at descending substrate steps.

Substrate step ➔ Trench ➔ Trench end

Distance (µm)

Times (s)

184 s
500 nm
638 s

[001] ➔ [110] ➔ [001]
Why does the film break up?

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.

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.

Energy ↓

For the observed mass flow from step $s_1$ to $s_2$:

$$E(h) = \text{film energy/area}$$

$$\frac{d^2E}{dh^2} < 0$$
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 $E_{n+1} + E_{n-1} - 2E_n < 0$
The people!

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