Characterization of Blistering and Delamination in Depleted Uranium Hohlraums

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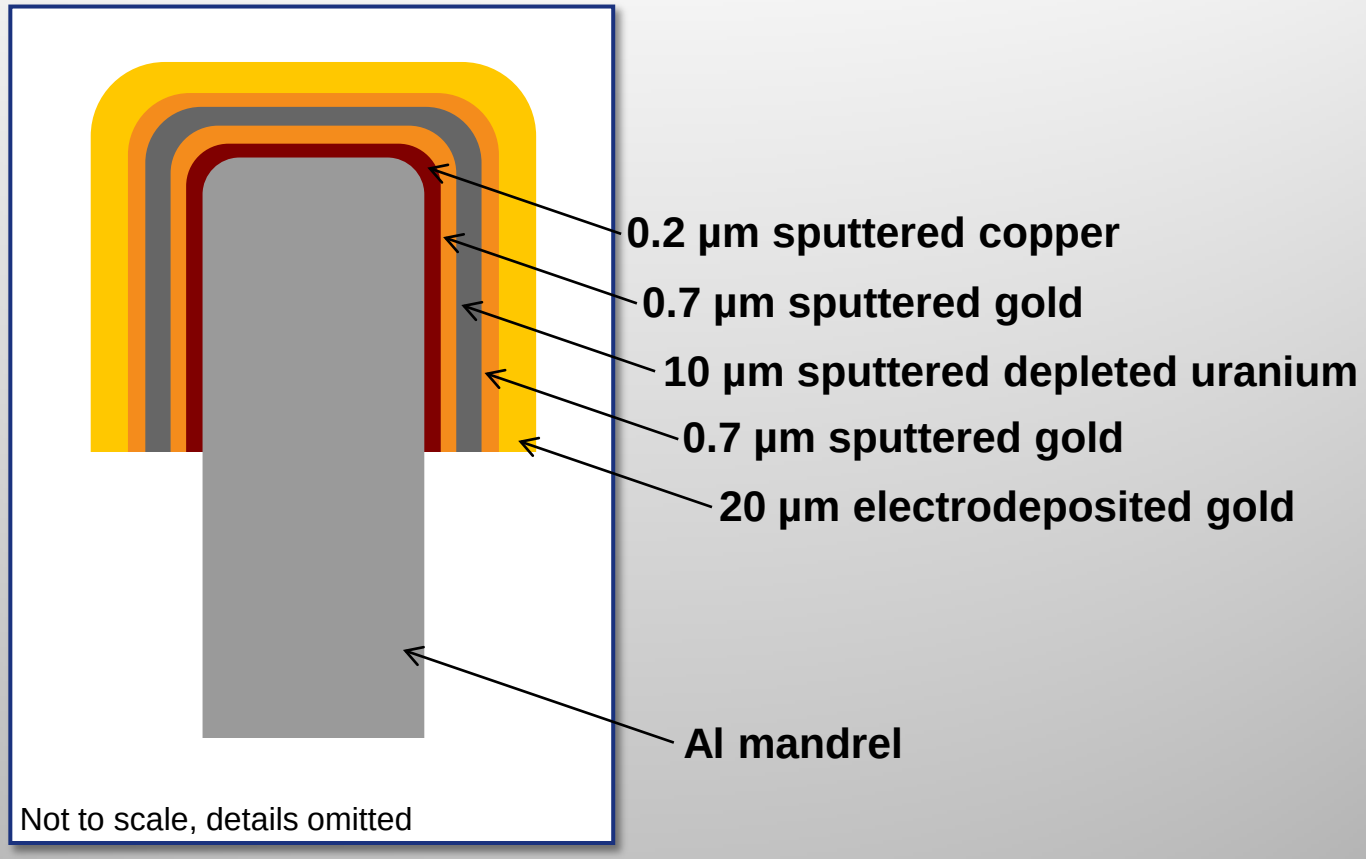
Microscopy techniques identified chemical attack during mandrel etching as a likely cause of DU hohlraum blisters

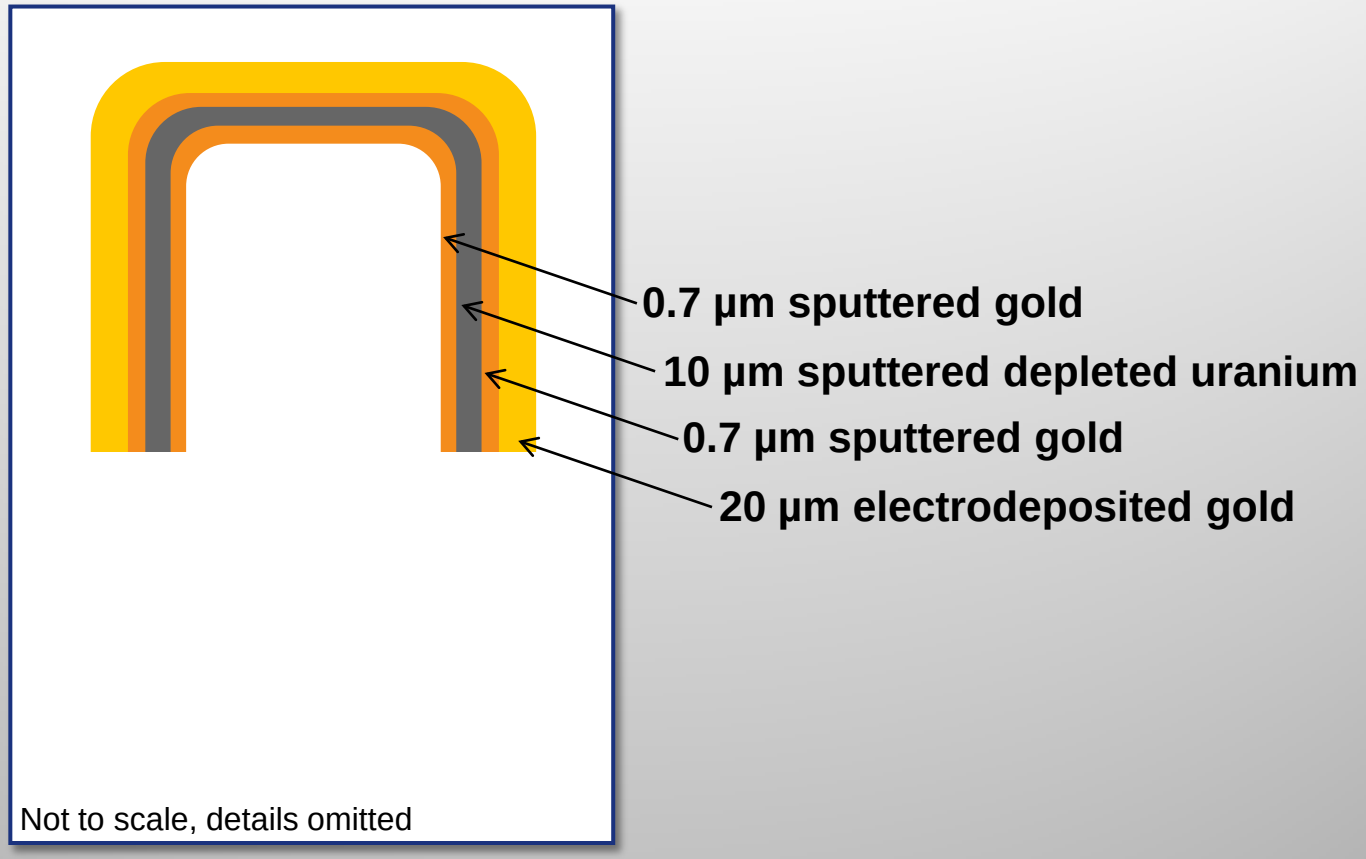
- Delamination occurs between Au liner and DU
- Rough Al mandrel surfaces are replicated in the Au and Cu layers
  - Pinhole formation is possible
- Our efforts are focused on better protection of the DU layer during chemical etching
  - Additional protective layers
  - Different Al mandrel alloy, which results in a smoother machined surface finish

DU hohlraums are multilayered structures deposited on an aluminum mandrel

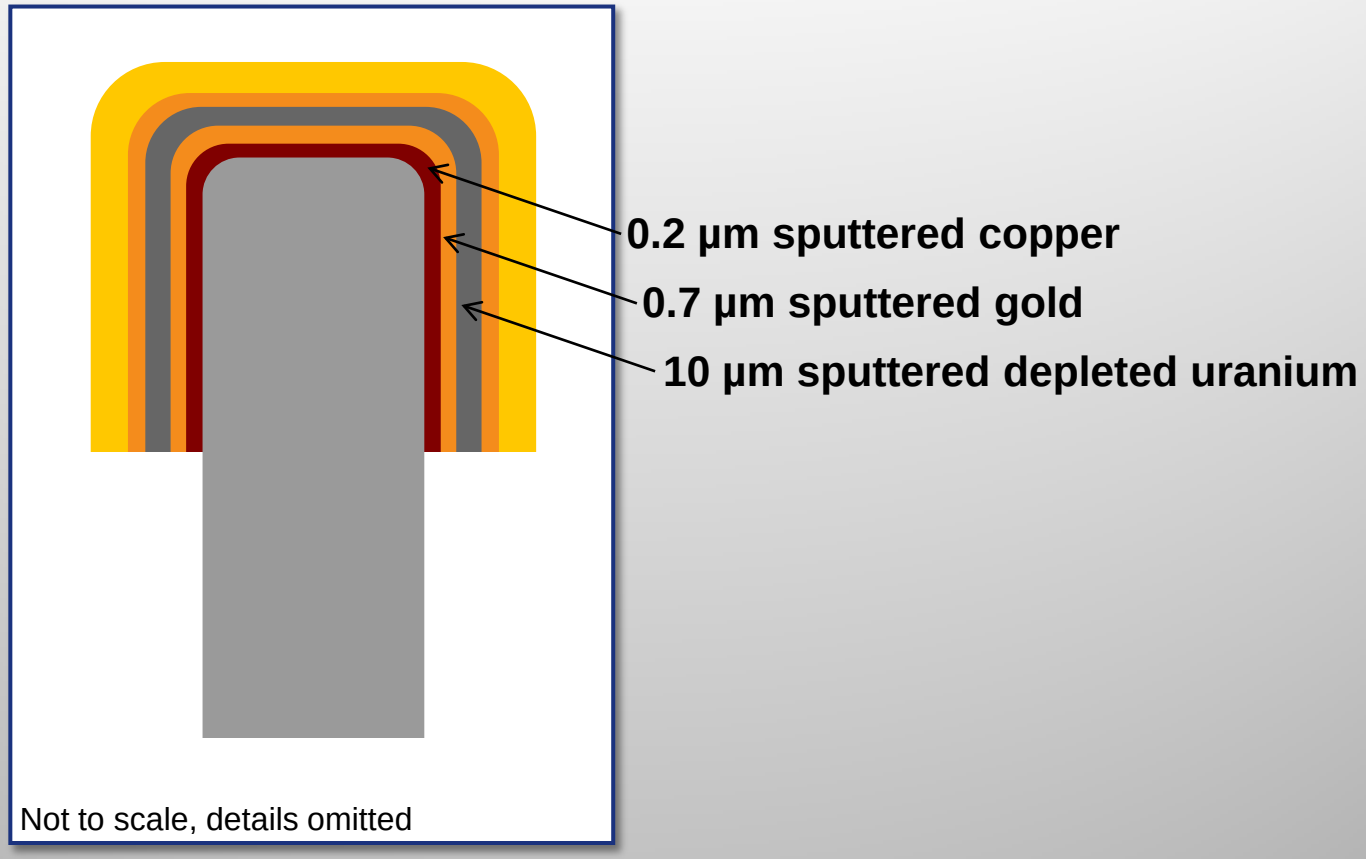


Hohlraums are deposited as “halfraums” and the 2 halves are assembled later

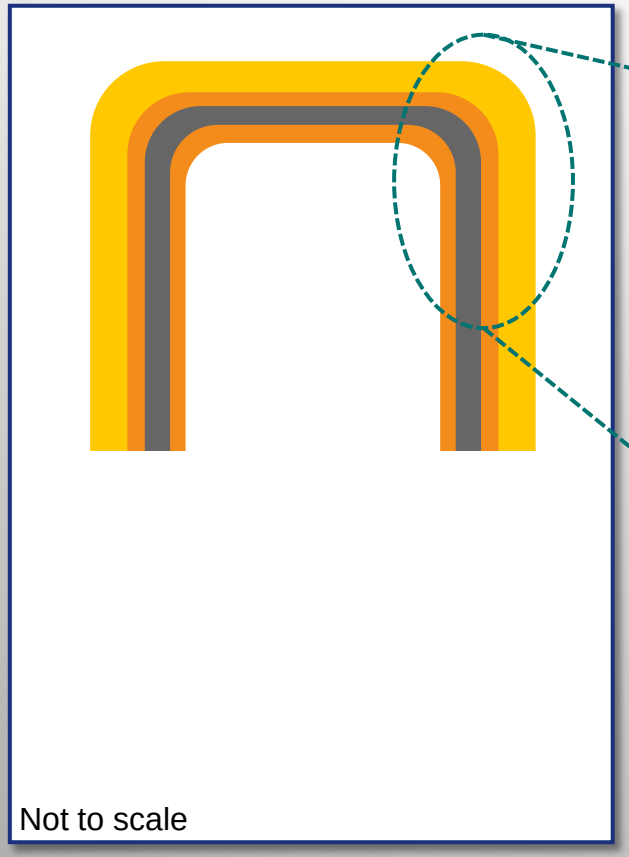
The Al and Cu are chemically etched, leaving a freestanding Au/Du/Au structure

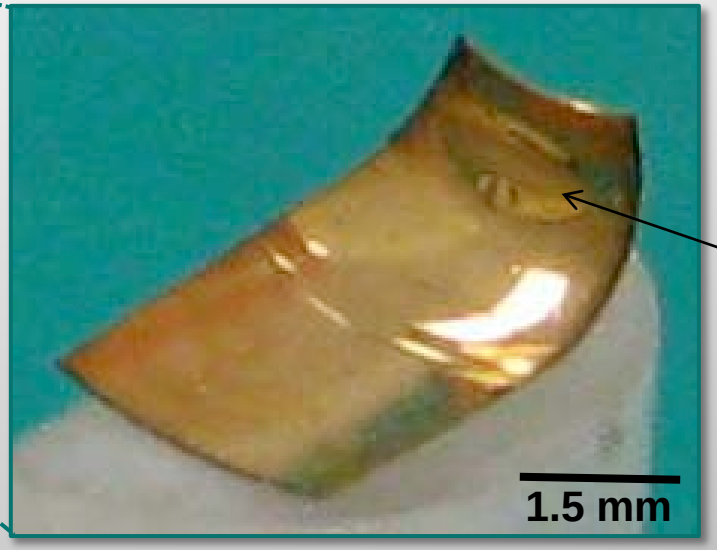


The Cu and inner Au layers are deposited to protect the DU from corrosion during etching of the mandrel



After etching, blisters are sometimes found in the interior radius area

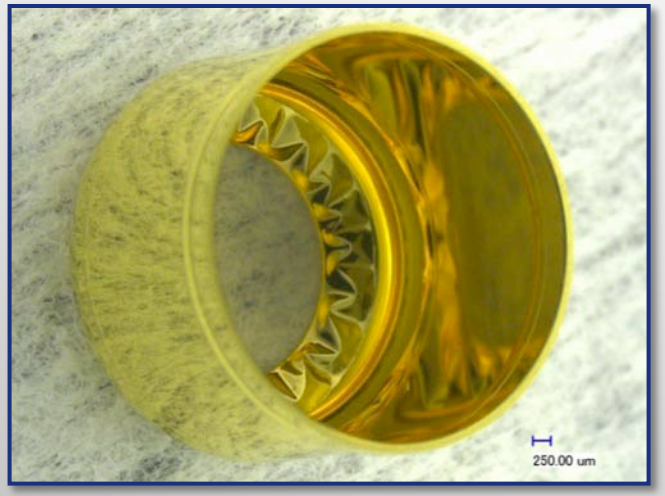


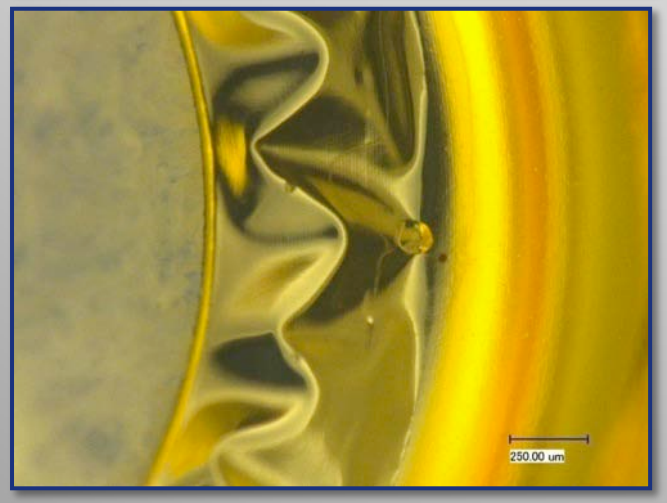


Blister at radius

Hohlraums with blisters generally cannot be used for experiments

We use microscopy techniques to characterize blisters, with the goal of modifying processing conditions to improve hohlraum yield

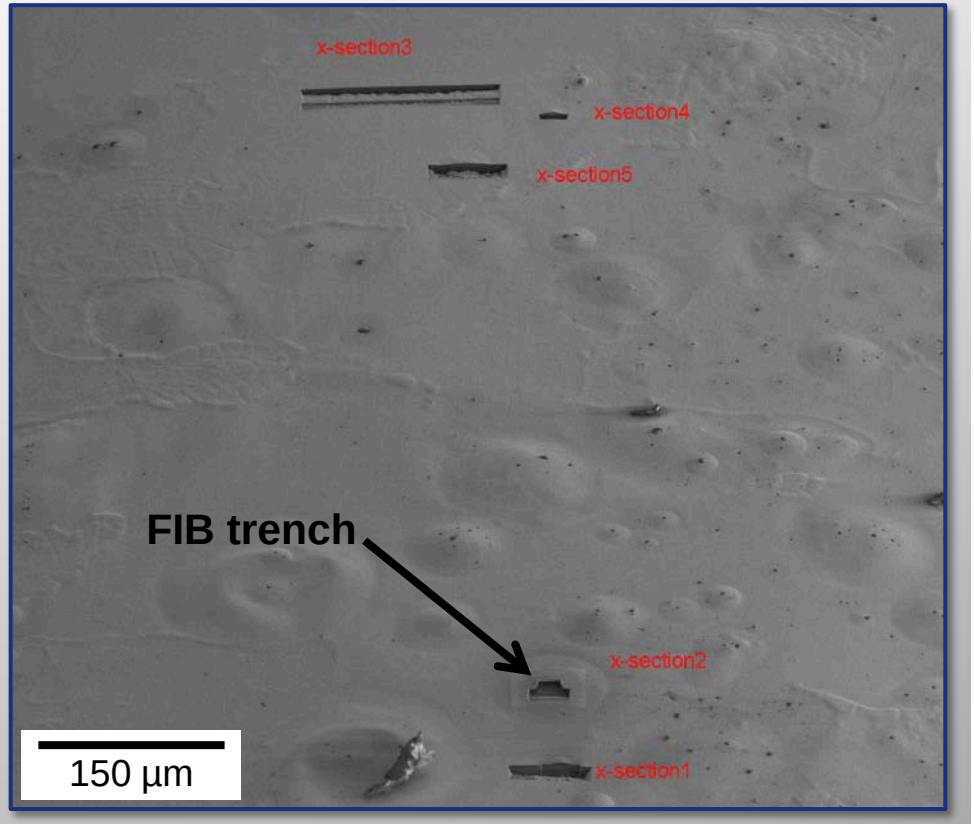




- Where is the delamination occurring?
  - Between layers? Which layers?
- What causes delamination?
  - Residual stress in films?
  - Poor adhesion during sputtering?
  - DU oxide formation during deposition?
  - Corrosion during Al mandrel leaching?

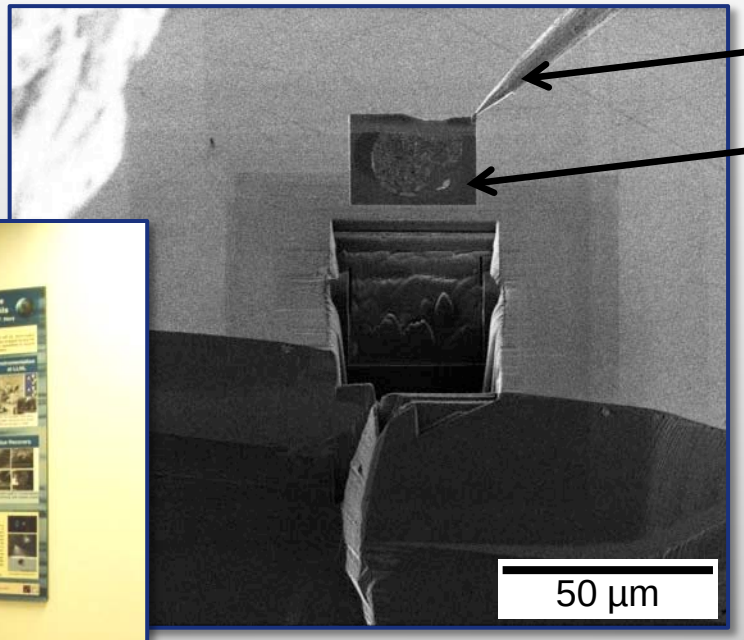
We utilize multiple characterization techniques to analyze blister failure modes

- Optical microscopy
- Scanning electron microscopy (SEM)
  - Imaging (microstructure)
- Electron dispersive spectroscopy (EDS)
  - Chemical composition
- Transmission electron microscopy (TEM)
  - Crystallographic structure
  - Microstructure
  - Chemical composition
  - Samples can be extracted with the focused ion beam (FIB) system



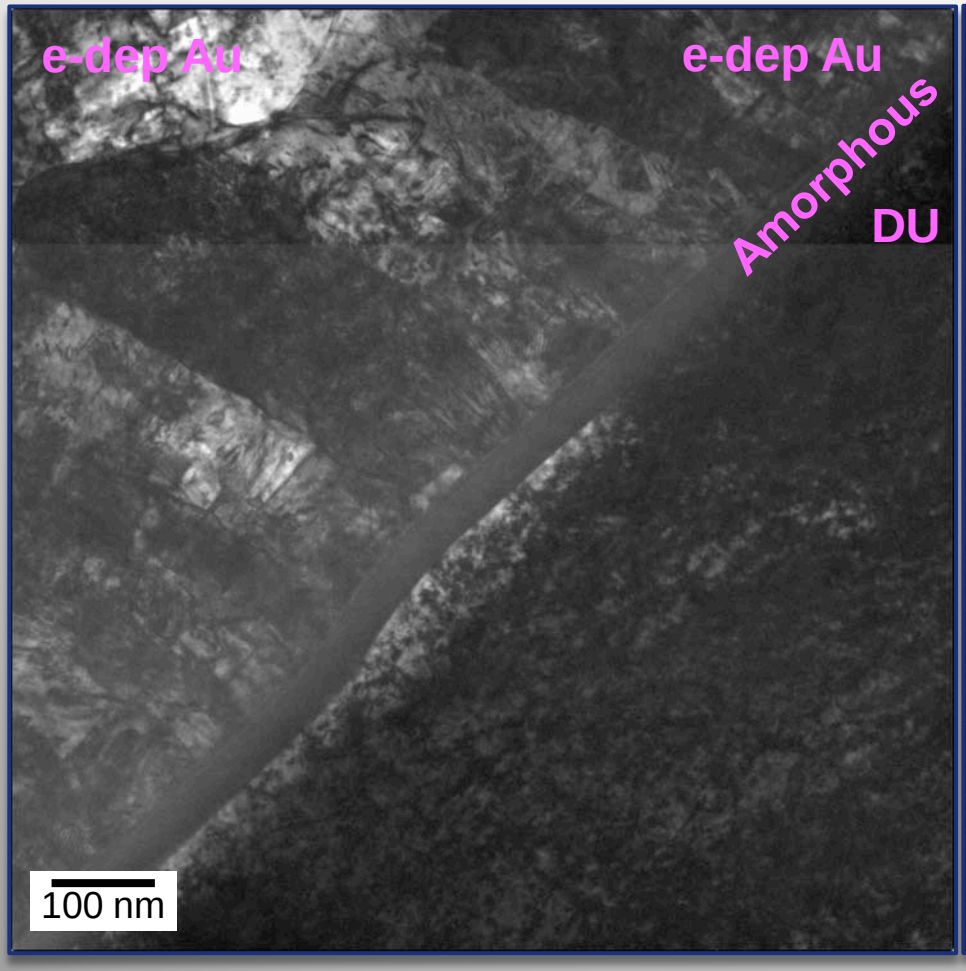
SEM image of a blistered gold layer showing areas investigated with FIB trenching

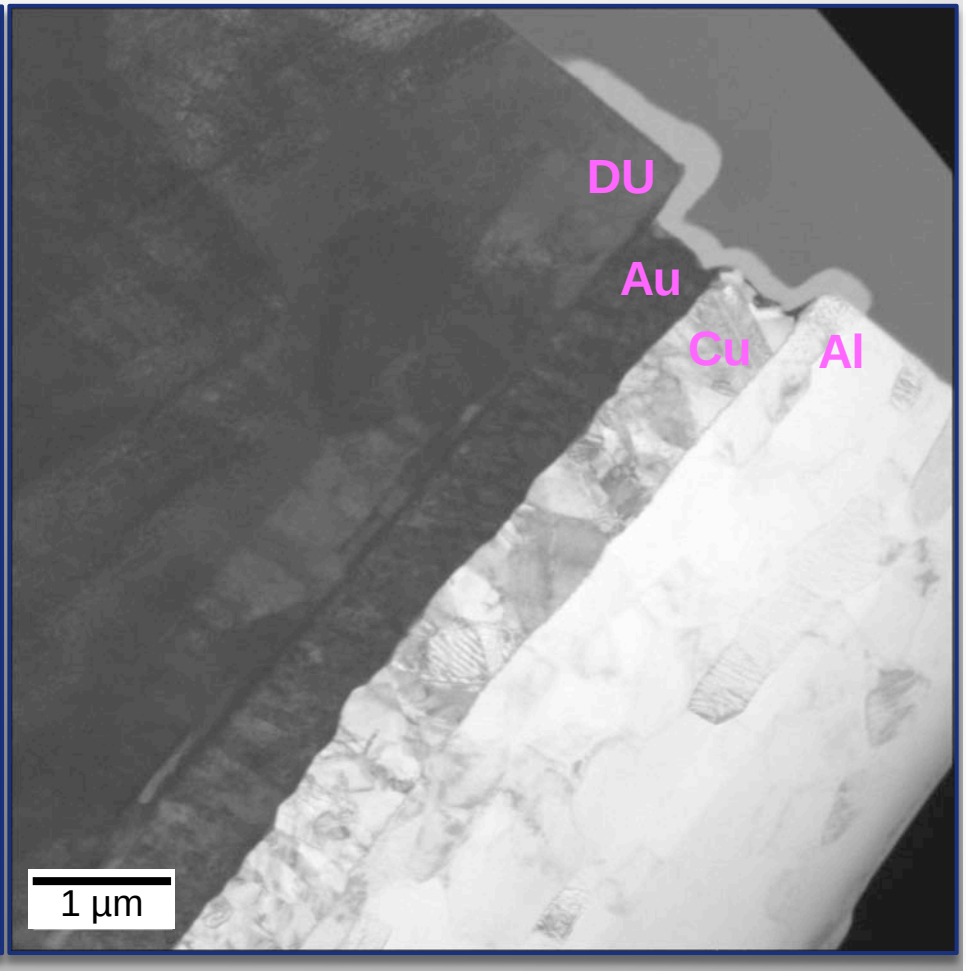
The Focused Ion Beam system provides cut-away views of samples



- Removes material by ion milling to reveal cross-sections
- Samples can be extracted for TEM analysis

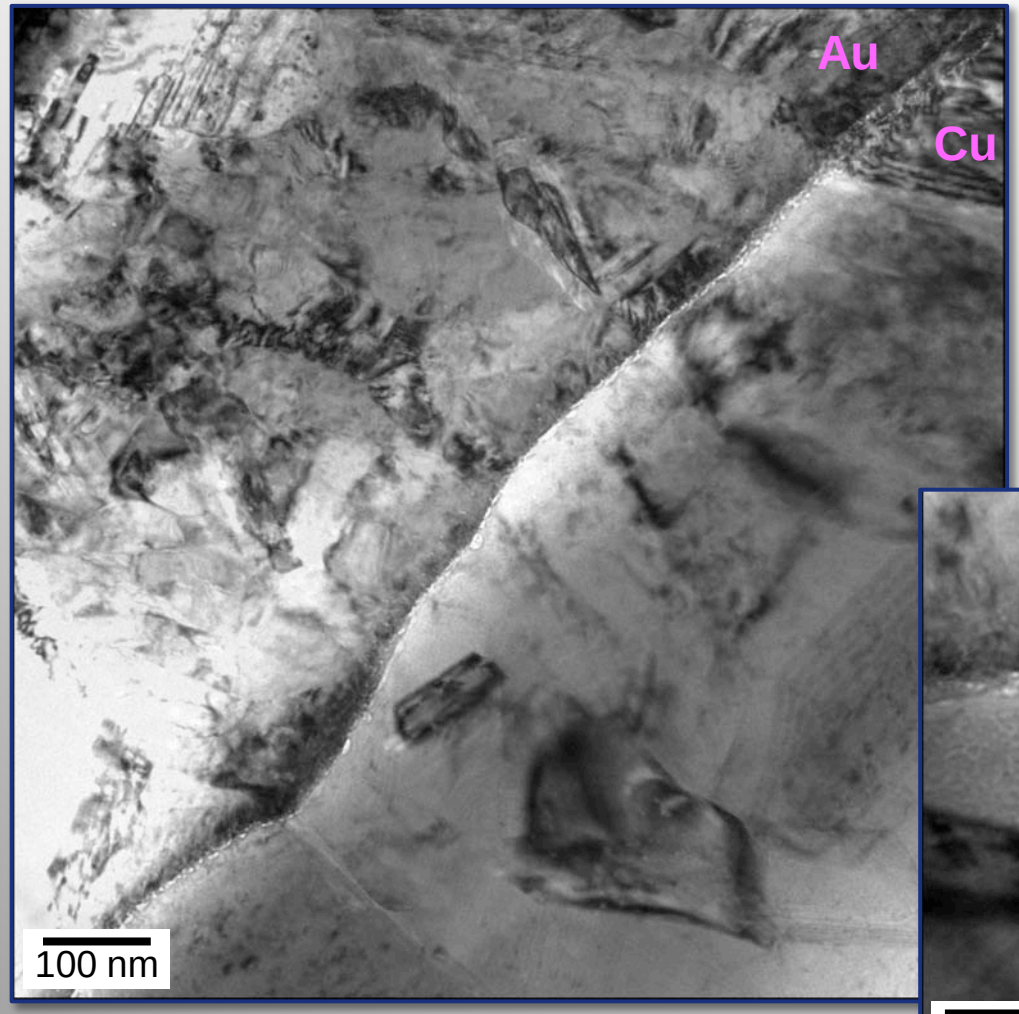
As-deposited, there is an amorphous layer at both Au/DU interfaces, and it is not an oxide



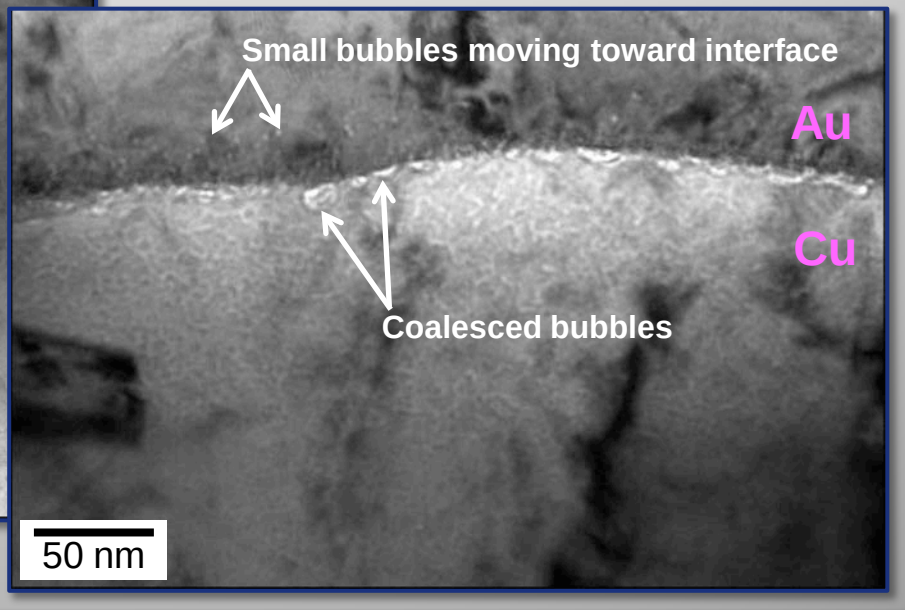


TEM images from a FIB liftout

The as-deposited Cu/Au interface is decorated with bubbles

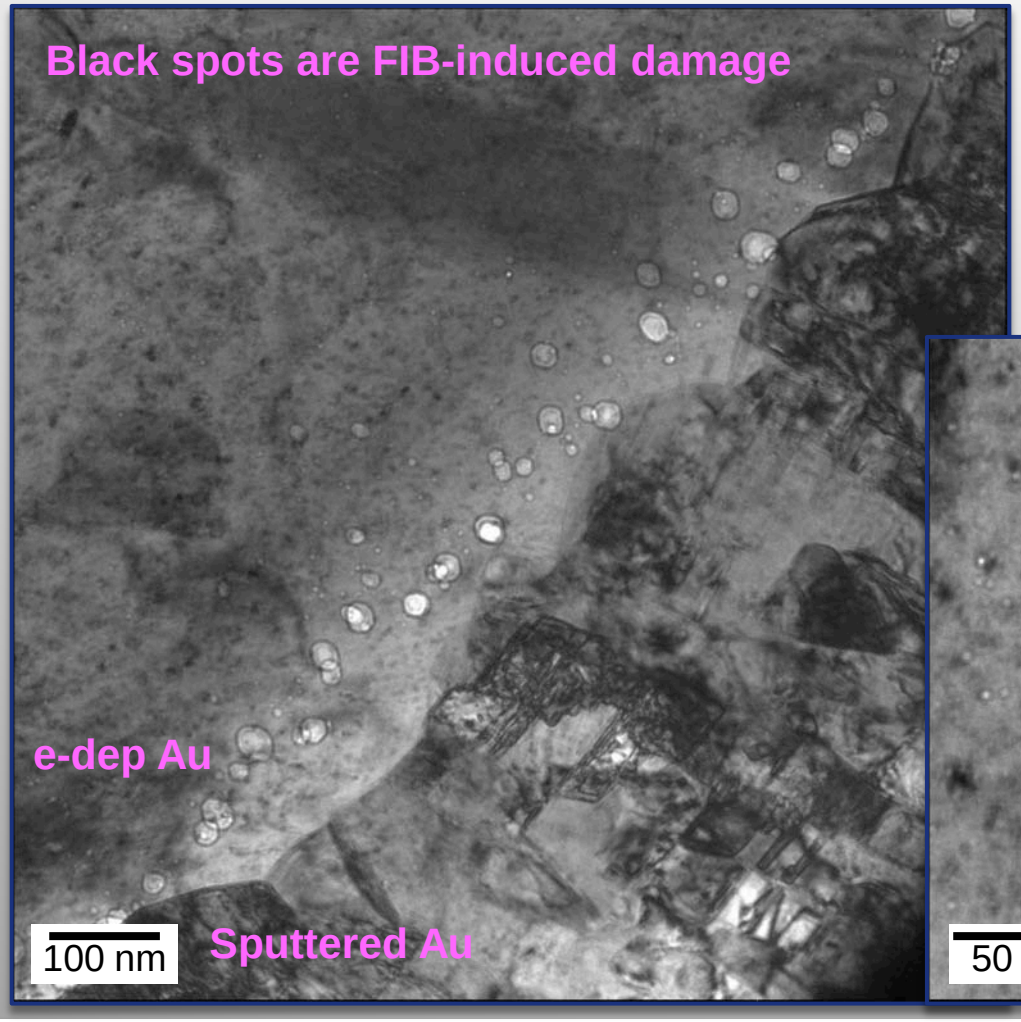


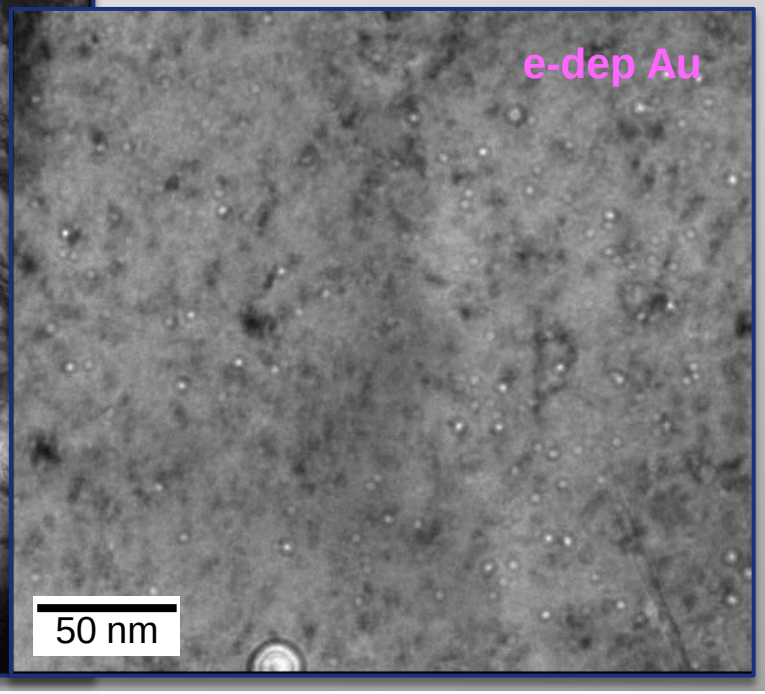
- Bubbles may contain incorporated sputter gas
- Heat during deposition may cause these bubbles to migrate from the "bulk" to the lower-energy interface where they coalesce



The electrodeposited Au layer is populated with bubbles (voids), which appear to migrate to the interface and coalesce

- Bubbles may be H_2 or O_2 incorporated during electrodeposition
- This Au layer is only structural, so bubbles are unlikely to affect hohlraum performance

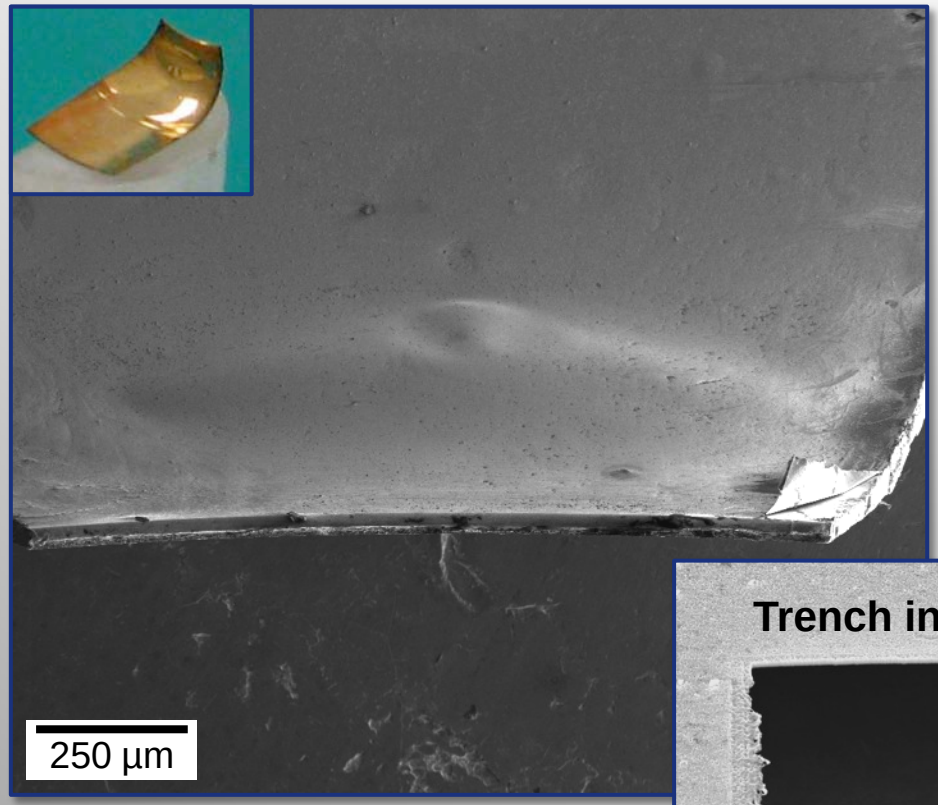




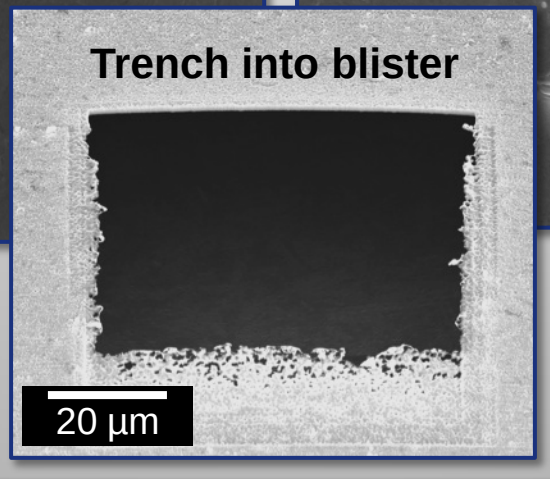
Our initial look at the hohlraums indicates no significant adhesion or delamination issues prior to chemical etching

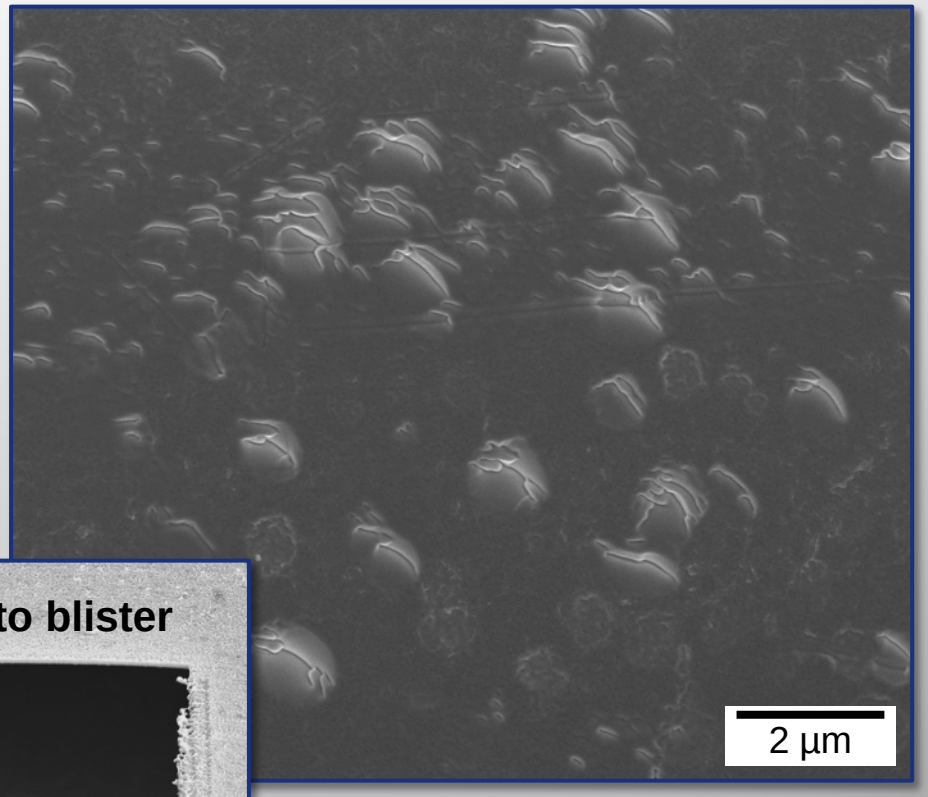
- We are still investigating other areas on an as-deposited hohlraum
  - FIB/SEM analysis showed some anomolies, which we are currently studying
- Chemical etching appears to be a prime suspect for causing blister formation
- Our efforts are focused on characterizing and strengthing the Cu and Au liner layers

Using the FIB, we milled into a blister on an etched part and determined that delamination was at the Au liner/DU interface



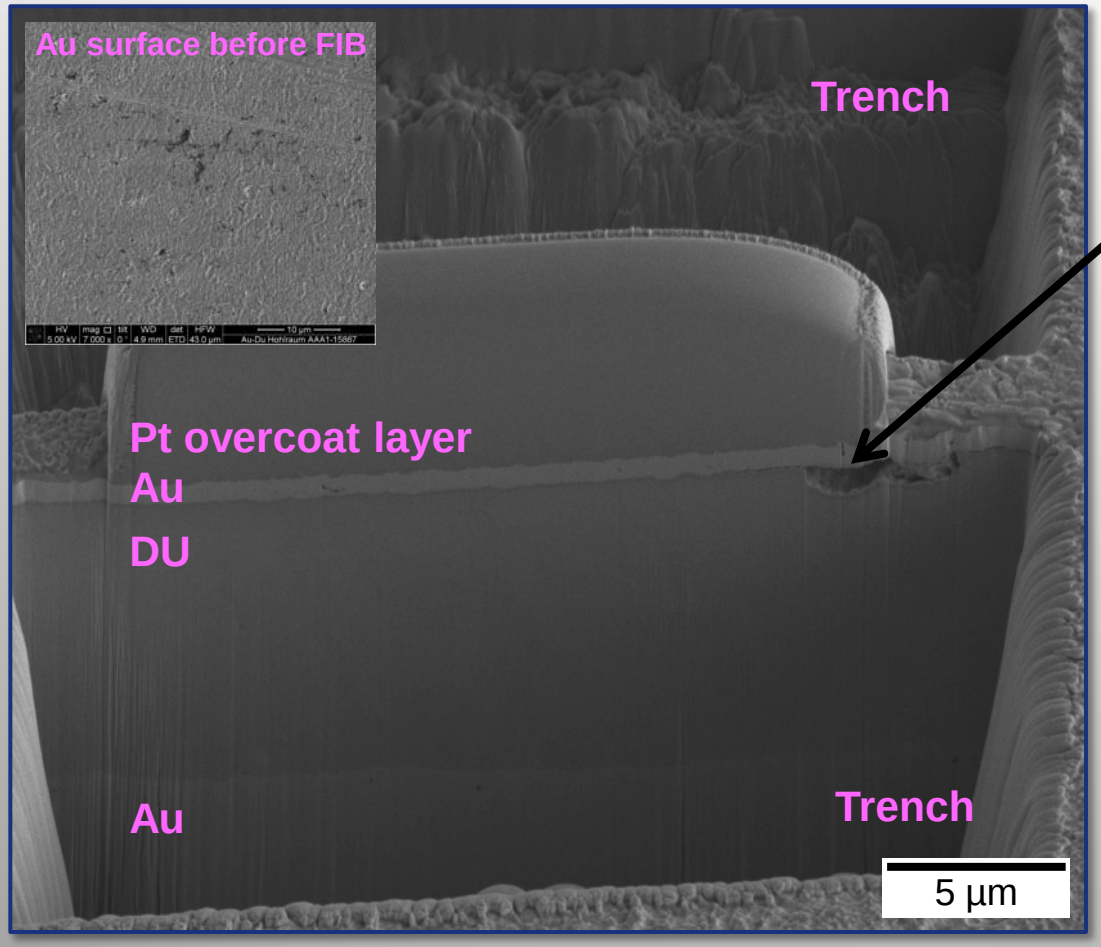
Blister before trenching





Cracked and corroded DU at the bottom of the trench

A FIB liftout in an unblistered region uncovered subsurface pitting and oxidation

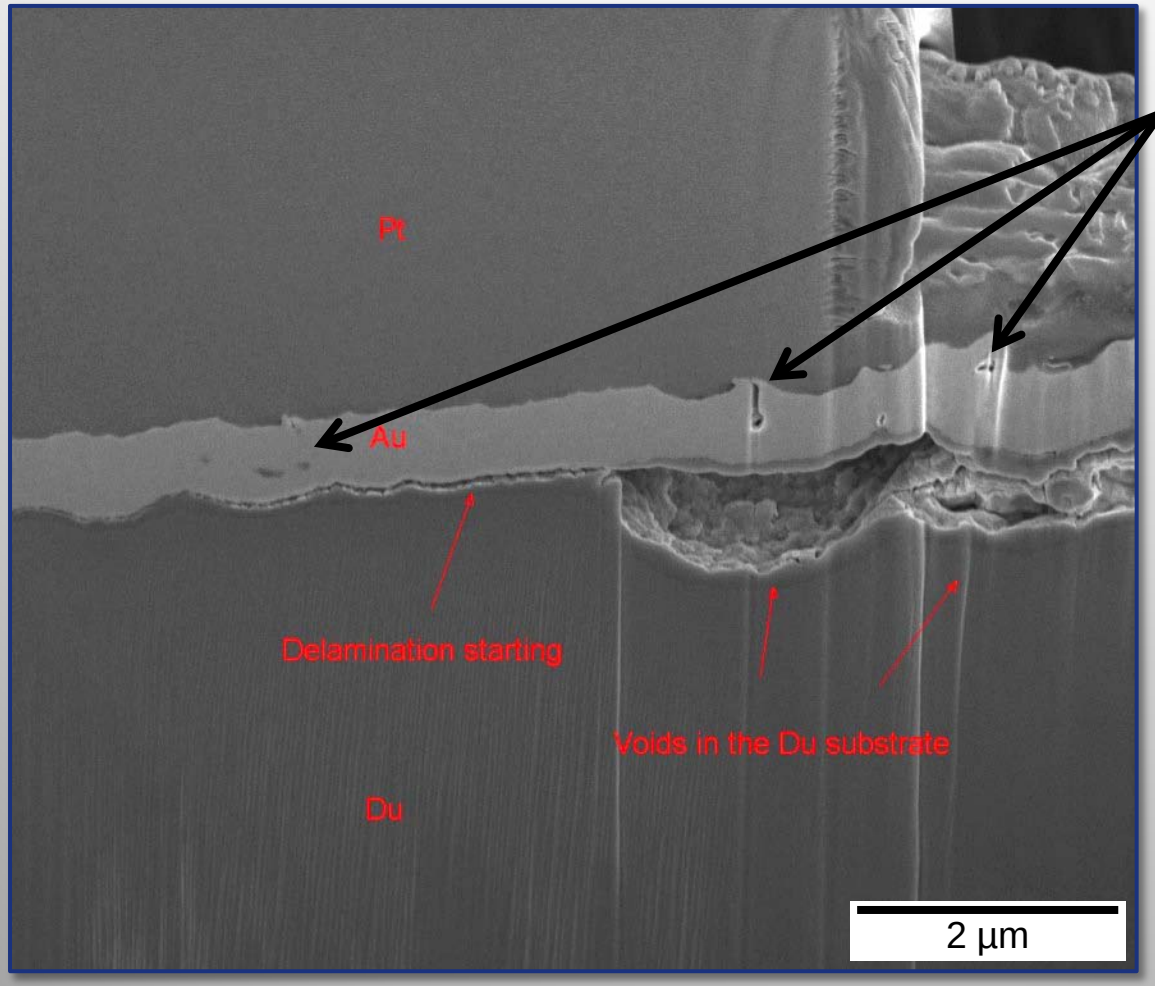


Subsurface pit in the DU

Au surface showed no evidence of pitting/blistering

Were we really lucky, or is pitting common?

The Au liner layer contains voids and possible pinholes and oxides

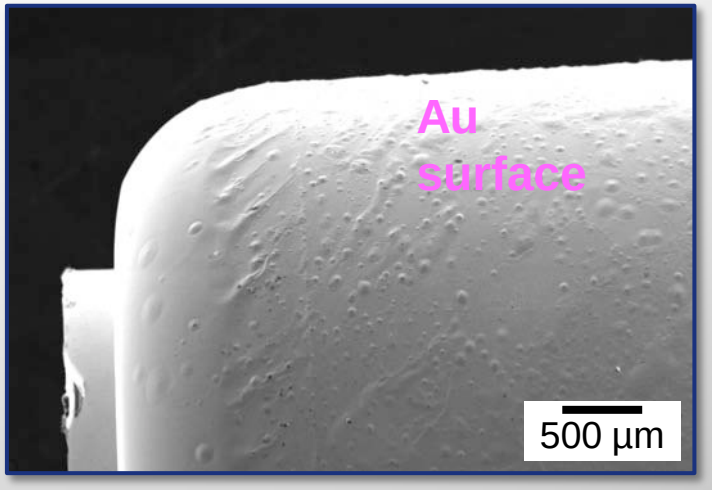


Voids and pinhole in Au layer

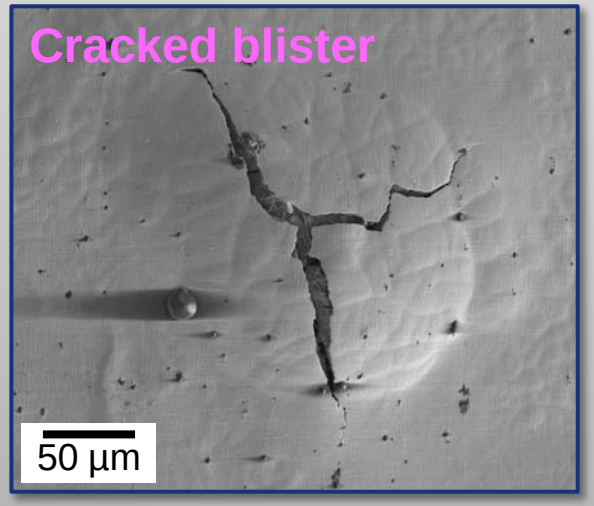
Thin oxide layers are not expected to affect hohlraum performance

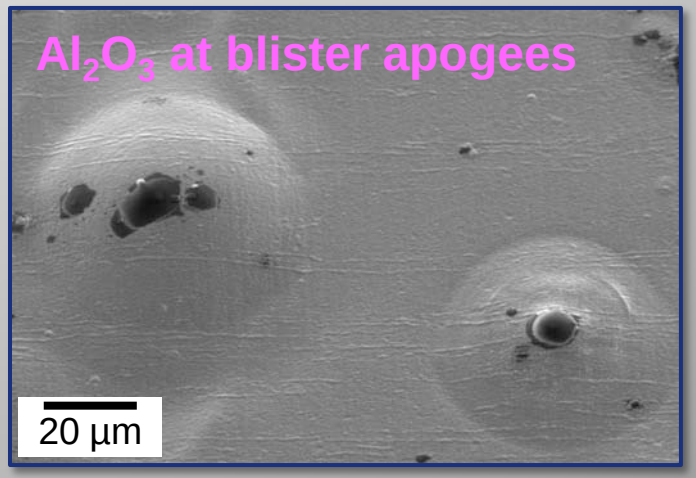
DU pitting and oxidation suggest that the Cu and Au layers do not fully protect the DU during etching

- Continuity of the Au layer was assessed with a test part
  - Al mandrel + 1 µm Cu + 0.7 µm Au
  - Layers deposited using the typical production process
  - Subjected to severe etch in nitric acid (longer etch than used on production parts)
- A pinhole-free Au layer would prevent blistering and degredation of the Cu layer under it

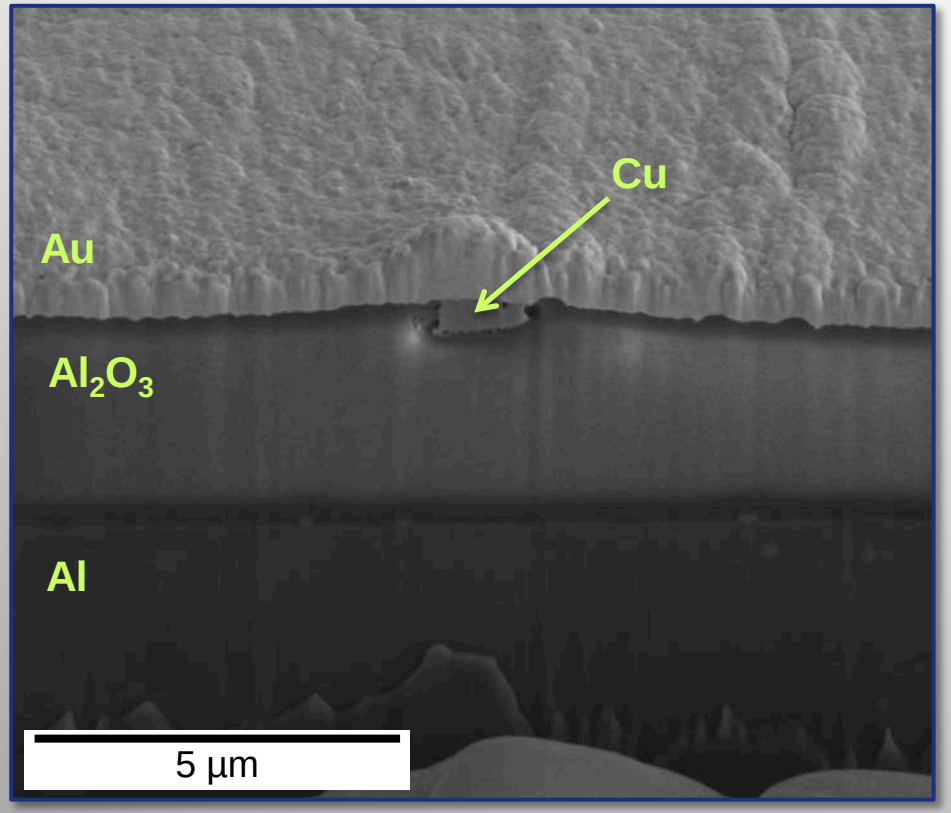


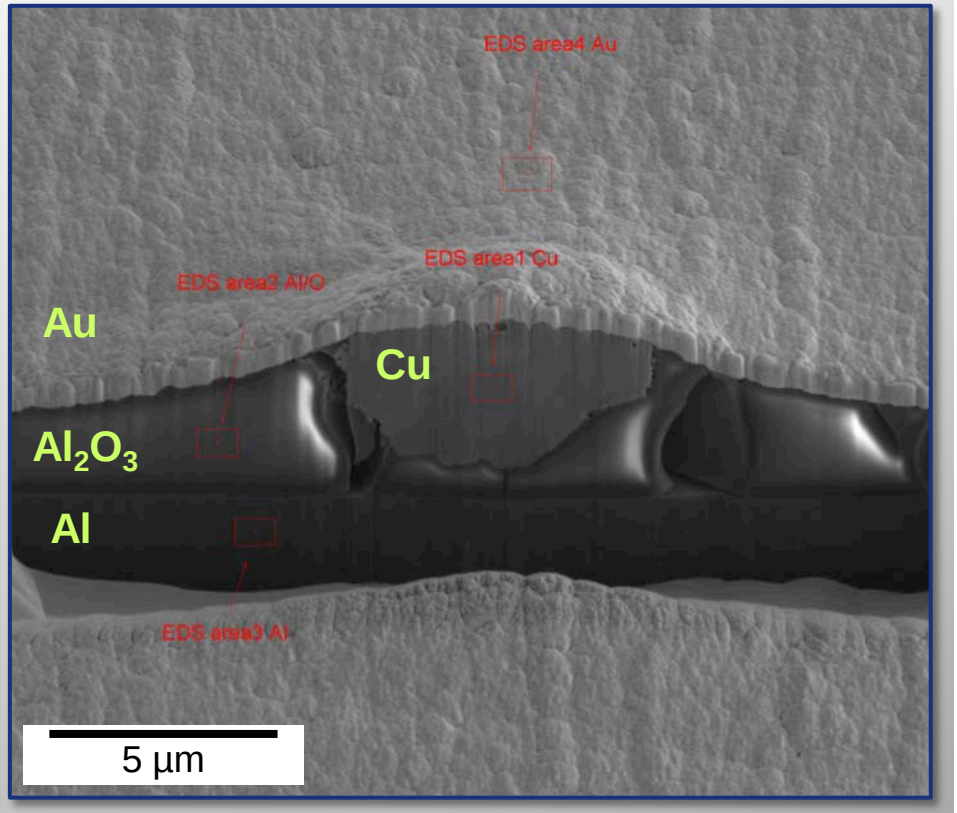
After the nitric acid etch, the part had a high density of blisters





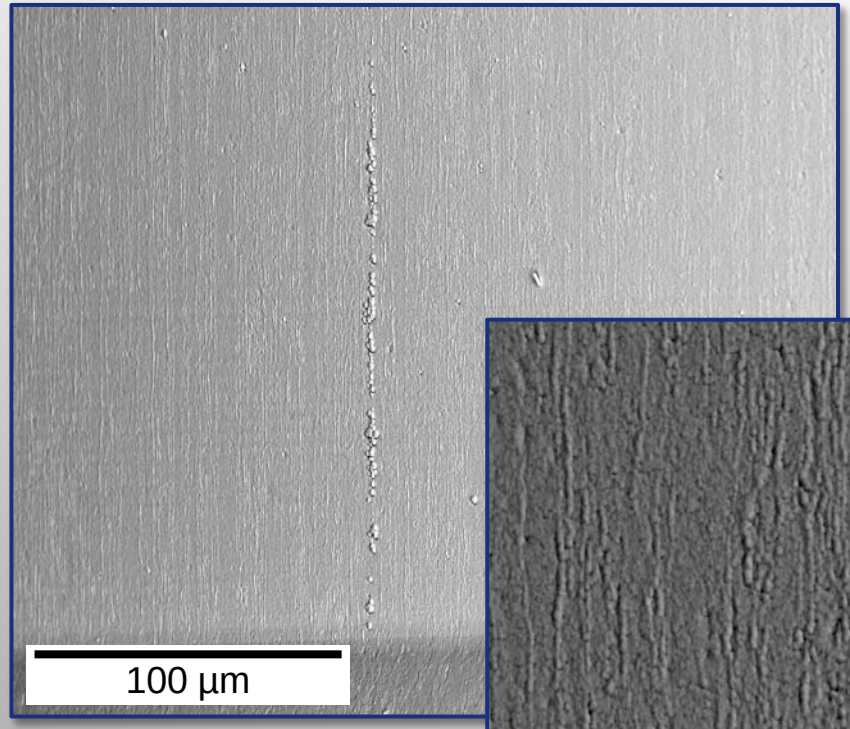
A cross-section through a blister shows that much of the Cu has been replaced by Al_2O_3

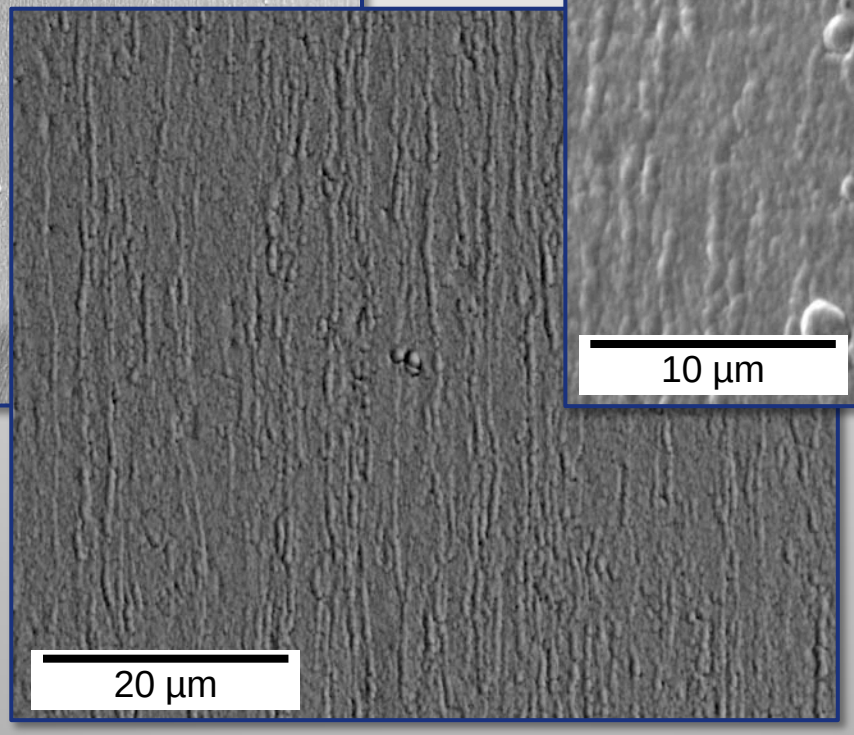


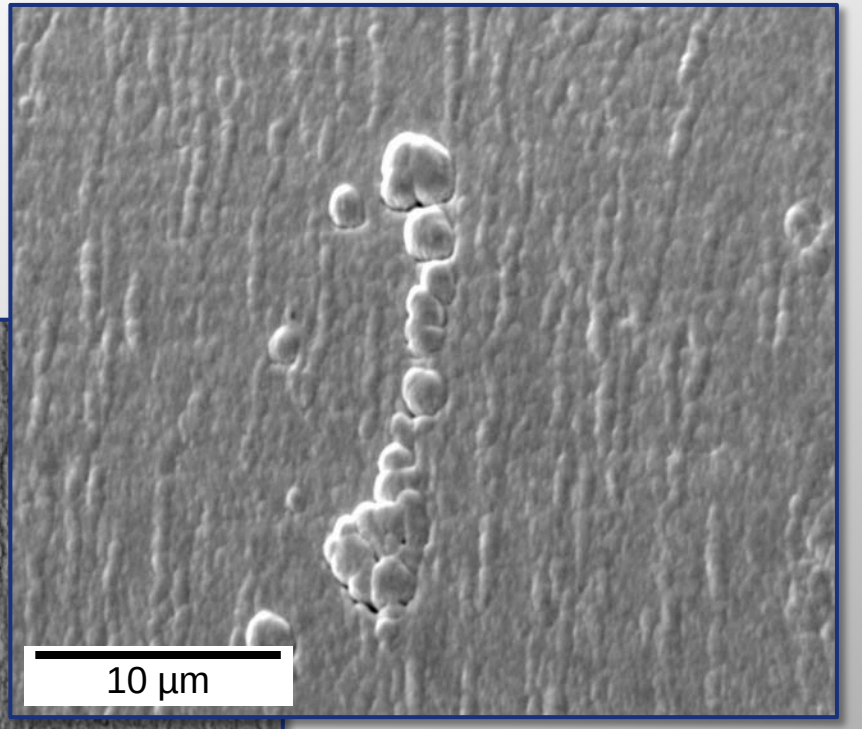


The Au layer, and likely the Cu layer, are permeable to the etchant

The as-deposited microstructure of the sputtered Au liner is columnar and contains strings of asperities

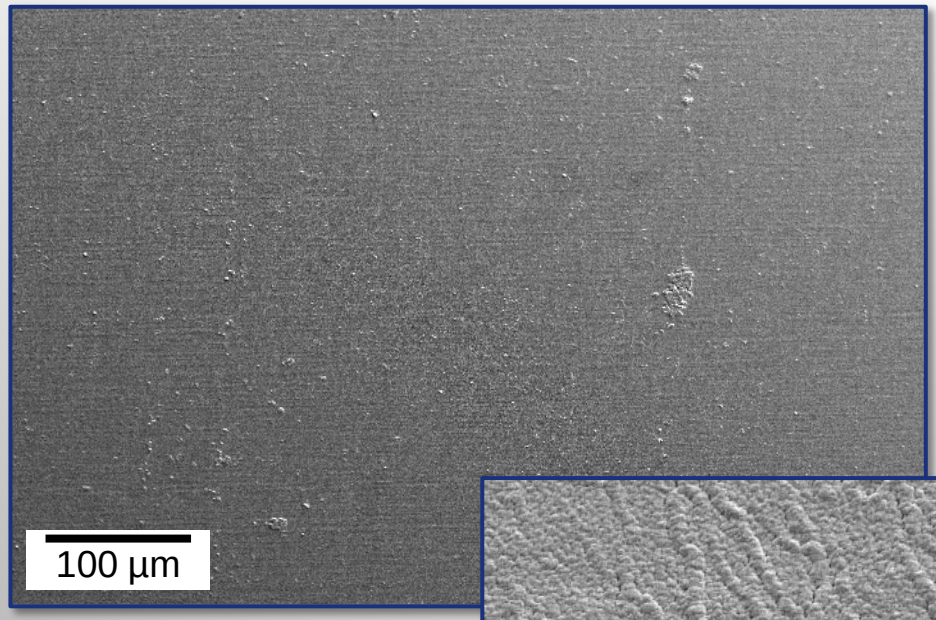


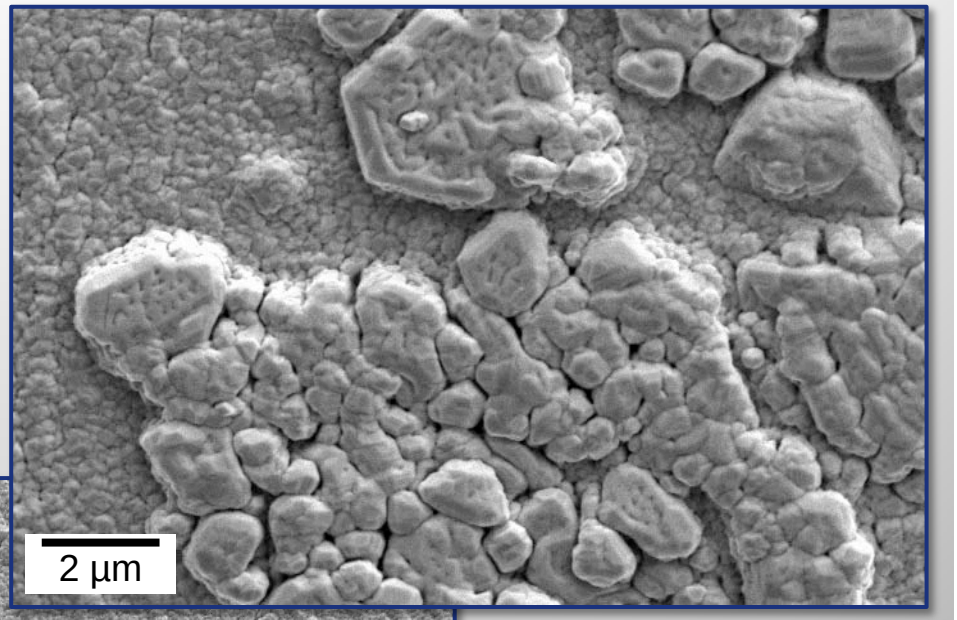


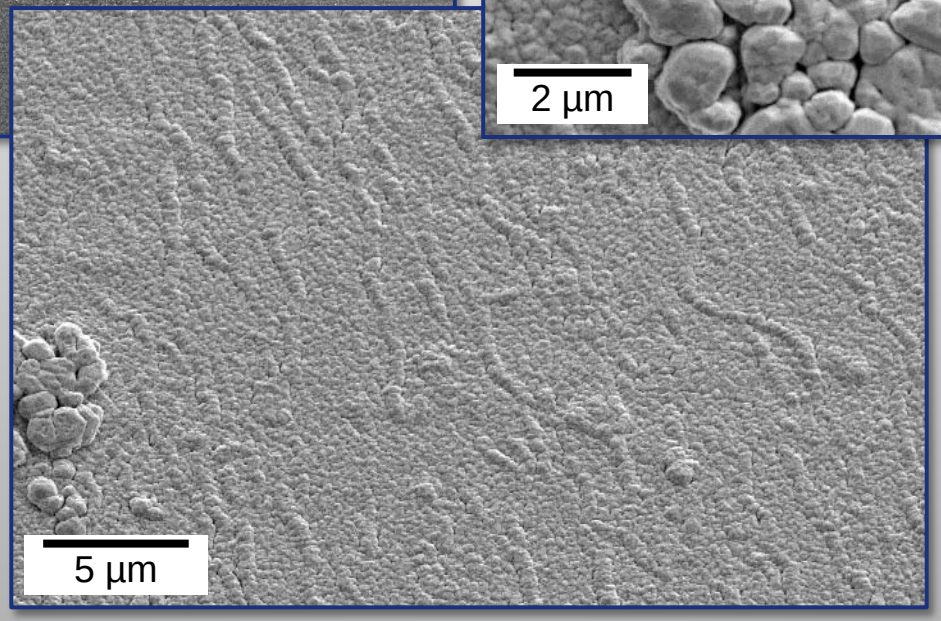


SEM images of as-deposited Au

The gold layer replicates asperities from the sputtered copper layer below it

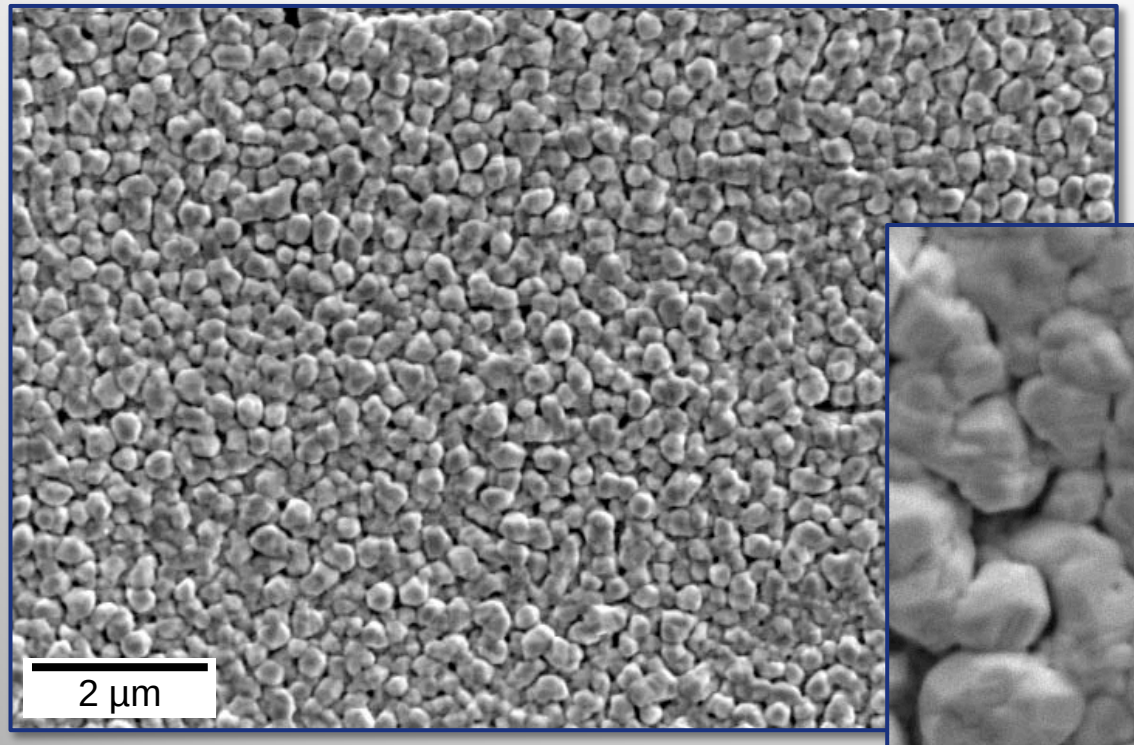




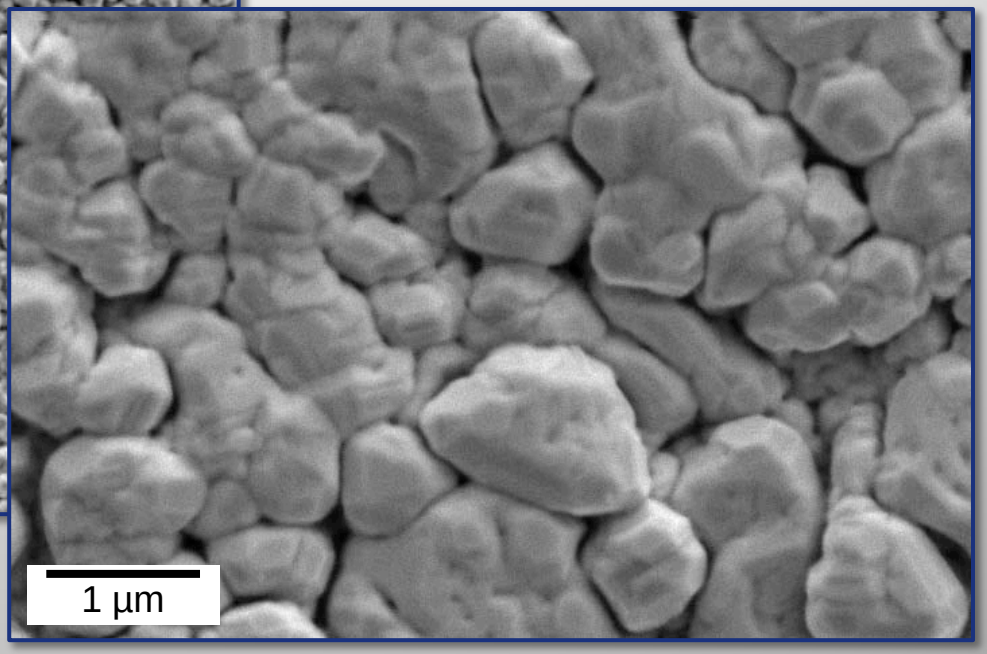


SEM images of as-deposited Cu

The Cu surface appears to be porous with a columnar grain structure

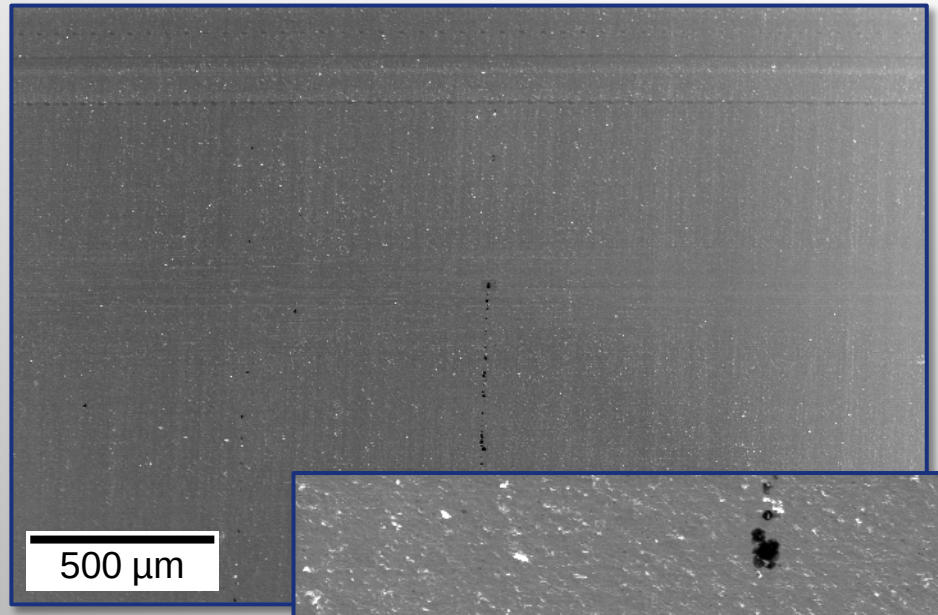


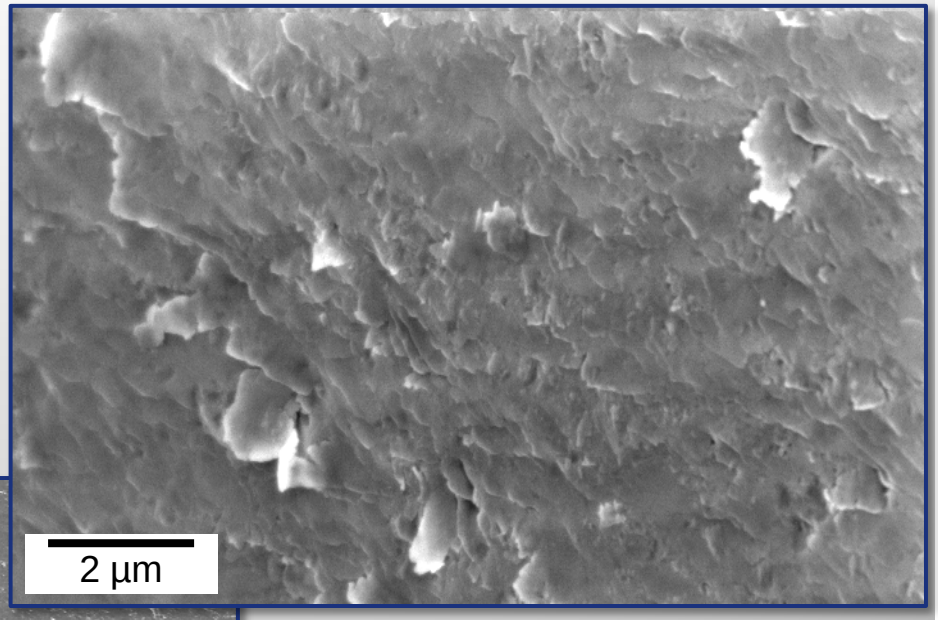
Columns likely extend through the film thickness

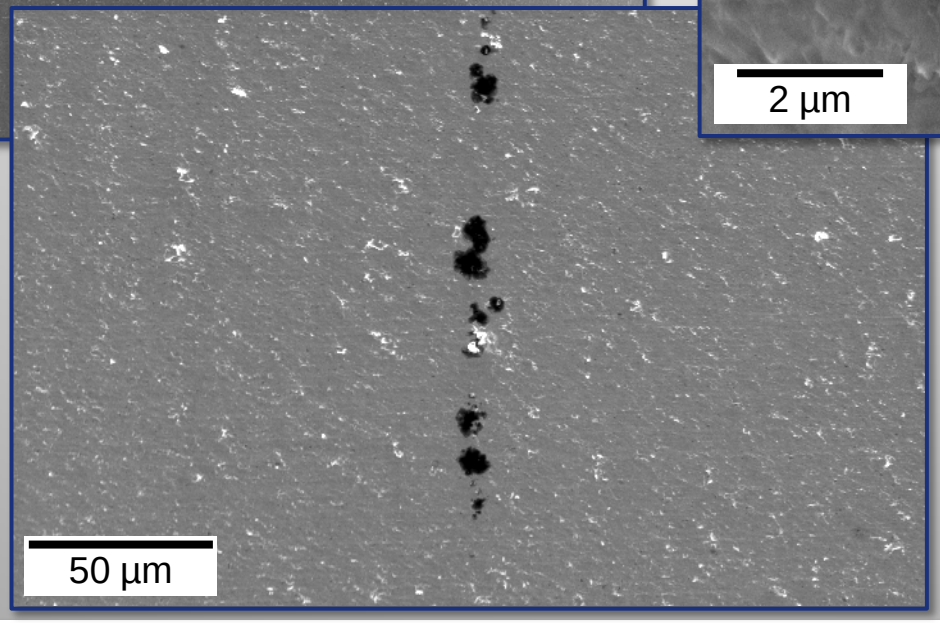


Cu is not fully dense, which can result in pinholes

Asperities in the Cu layer originate on the machined Al mandrel

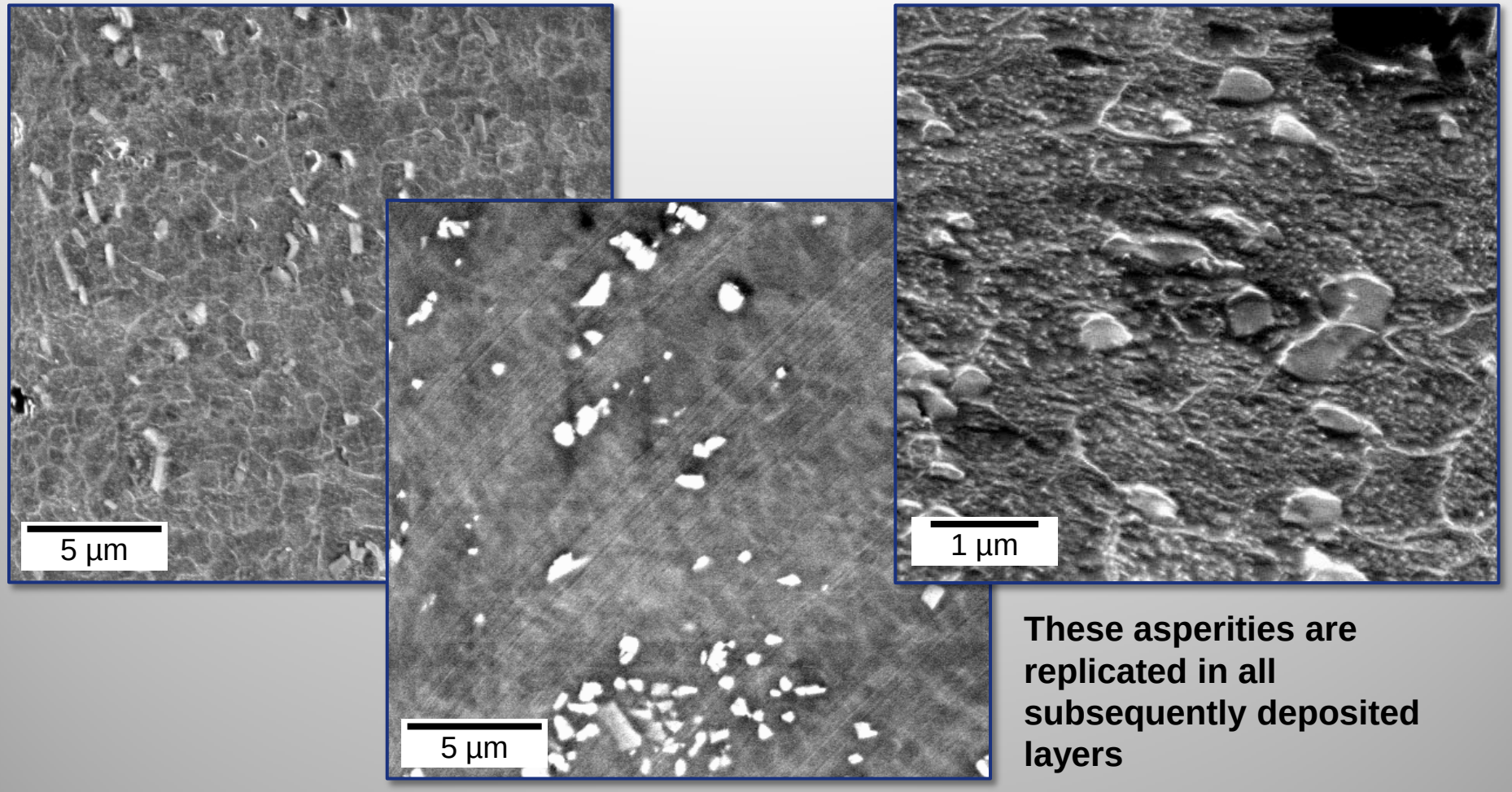






Hard Si and Fe inclusions in the Al 1100 alloy can dull/chip diamond tools resulting in a degradation of suface finish

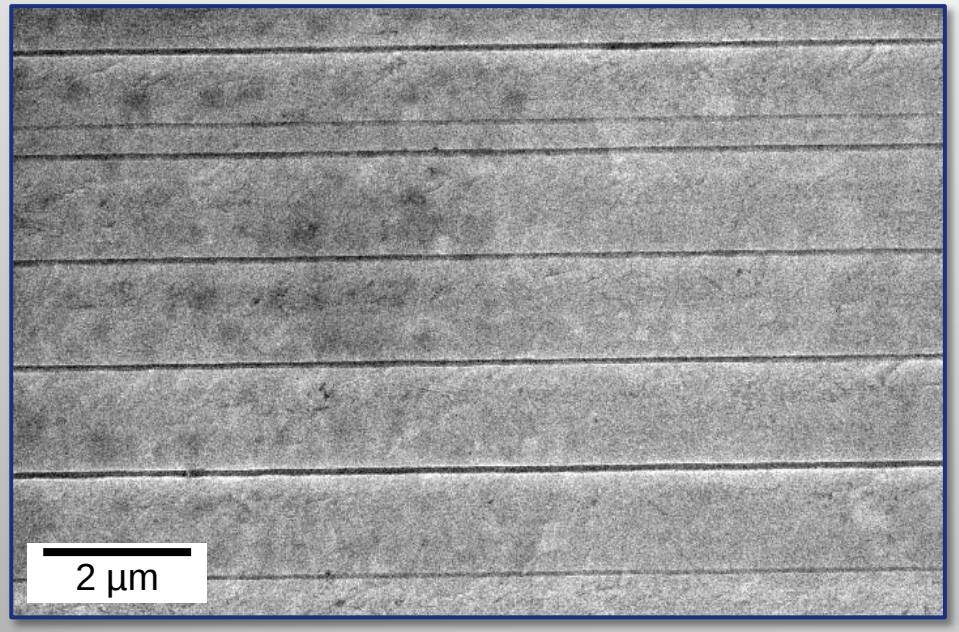
Ion etching of the Al mandrel preferentially removes Al, leaving precipitates protruding from the surface

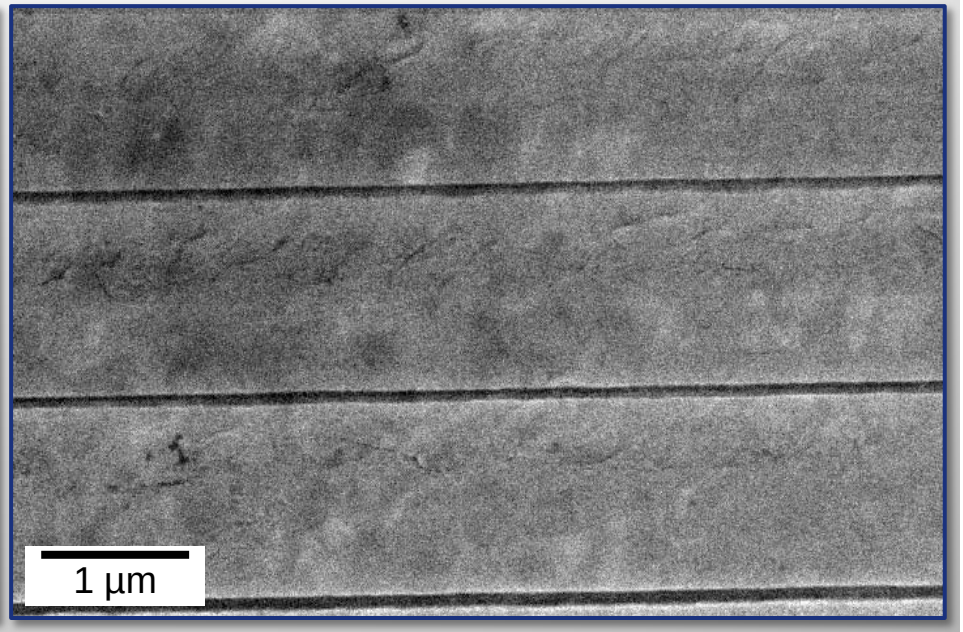


These asperities are replicated in all subsequently deposited layers

GA recently demonstrated an improvement in hohlraum quality by decreasing ion etch time

We are performing trial depositions on 99.99% pure Al mandrels





Parallel lines are machining marks

The asperities and smearing observed on the 1100 Al mandrels are not present in high-purity Al mandrels

Rougher substrate substrates result in more pinholes

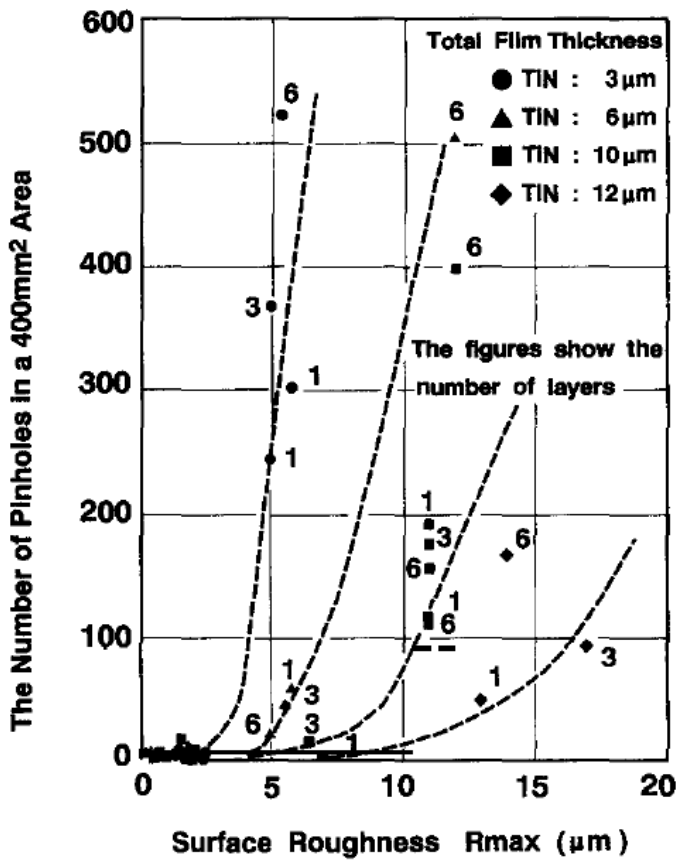


Fig. 2. Variation in the number of pinholes with surface roughness R_{max} of substrates for TiN films. The total film thickness was varied from 3 to 6 to 10 to 12 μm, and the number of layers was varied from 1 to 3 or 6.

The number of pinholes can be reduced by increasing the numbers of sputtered layers

Munemasa & Kumakiri,
Surface and Coatings Technology, 1991

Based on data obtained from characterization of mandrels and deposited films, we are focusing our efforts on improving the Cu and Au liner layers

- Higher quality Cu and Au layers will protect the DU during chemical etching
  - Shorter ion etch time (has already shown improvement)
  - Different mandrel alloy
  - Change in sputter parameters to modify microstructure
- Additional protective layers can decrease pinhole coincidence
  - Electrodeposited Cu layer
  - Al layer deposited on Al mandrel
- Decrease in chemical etch time
  - Modify mandrel geometry by drilling out some of the Al mandrel

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