Bi-Polar HiPIMS deposited Nb3Sn

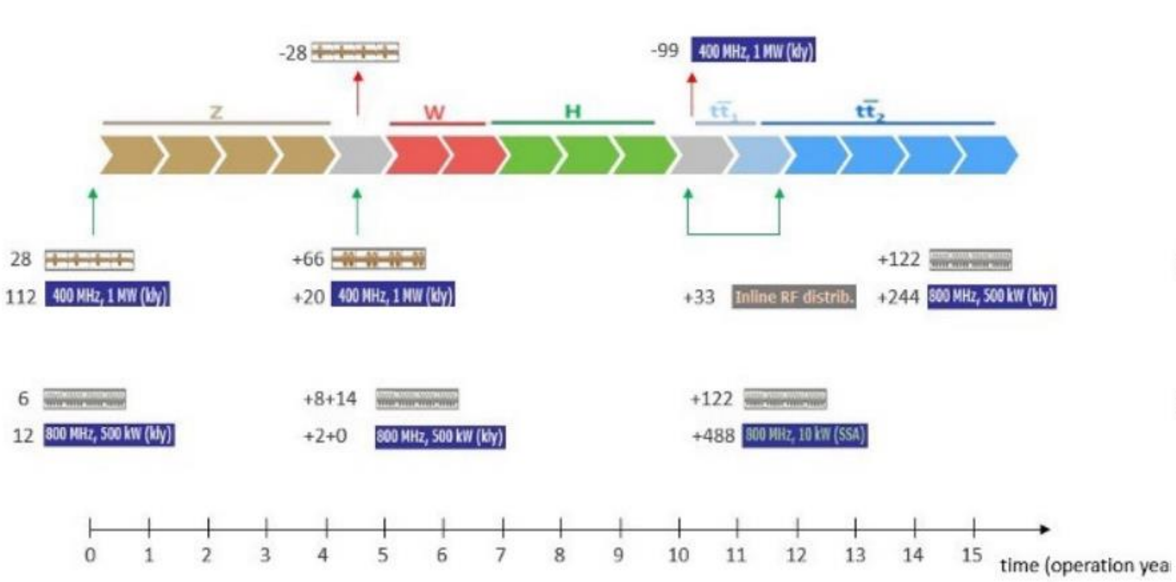
What we know so far – a practical overview

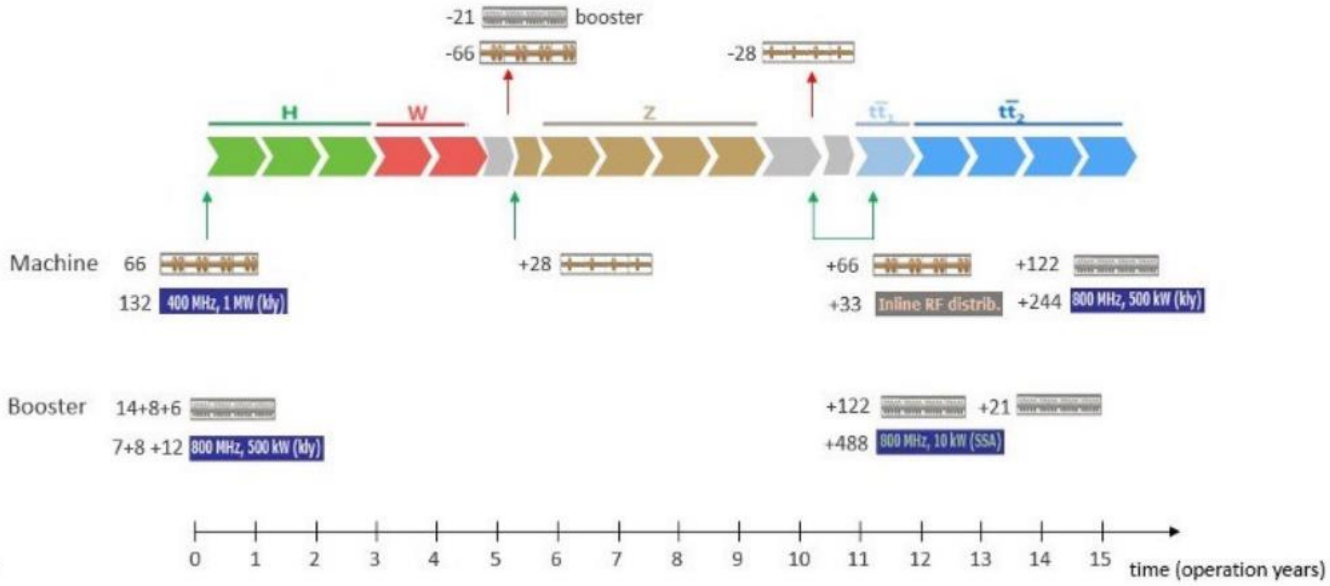
S. Leith, V. Giglia, G. Rosaz, C. P. A. Carlos, S, Pfeiffer, A. Moros, A.-T. Perez-Fontenla, L. Vega-Cid, K. Brunner, W. Venturini-Delsolaro, J. Bernardi, A. Steiger-Thirsfeld, J. Gruber, M. Himmerlich, V. Petit

Outline

1. Context
2. European overview
3. Experimental setup
4. Feasibility
5. Target Management
6. Some process parameters' effects
7. Macroscopic defects library
8. QPR samples

Context





FCC needs 100's of bulk Nb cavities for booster operation

Currently foreseen to operate at 2K

Could it be operated at 4.5K? Could we switch to Cu substrates?

Nb_3Sn / Cu appears as the most promising alternative

European overview - Actors

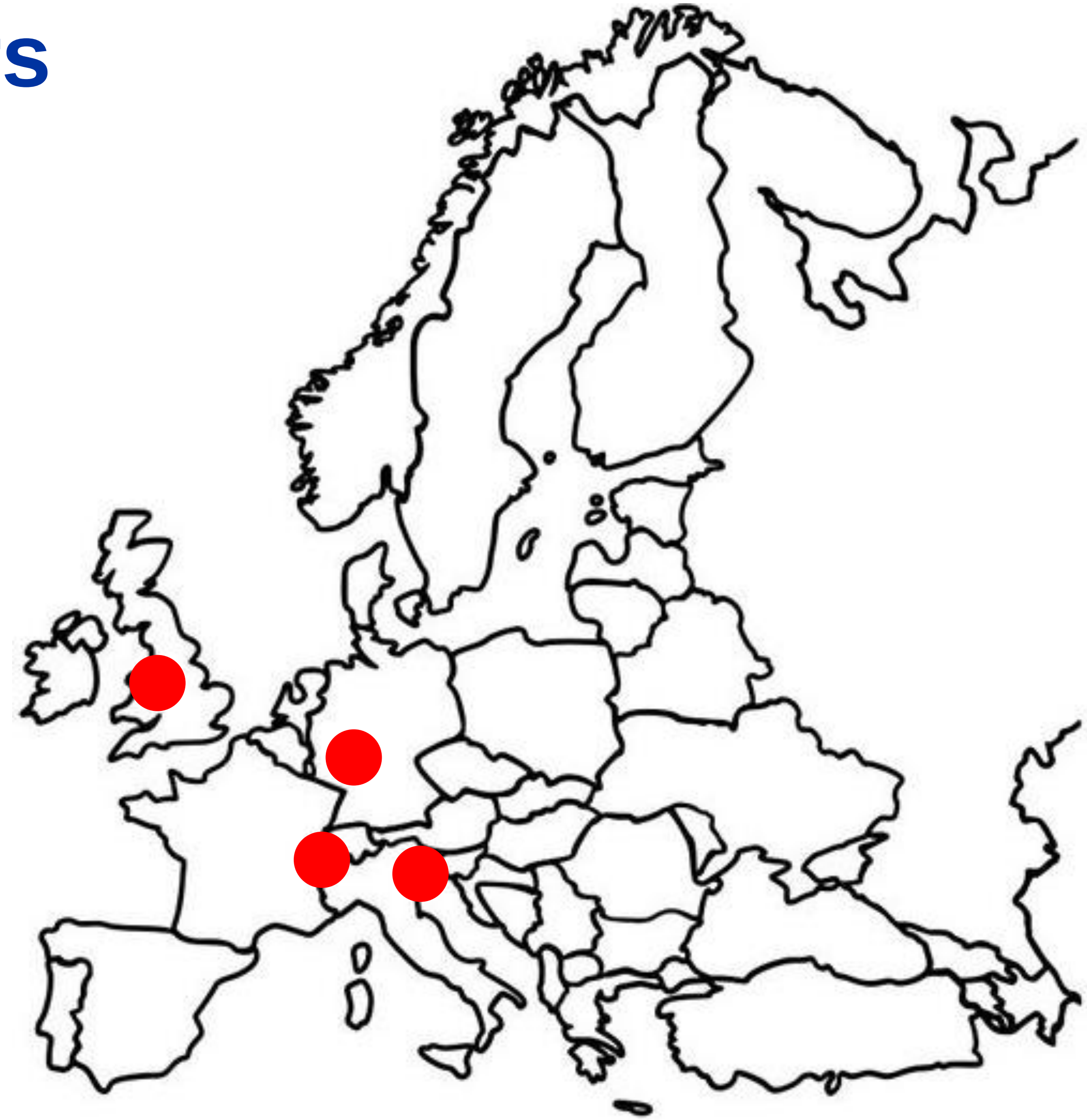
A15/Cu

STFC Daresbury Laboratory

TU Darmstadt

INFN Legnaro

CERN Geneva



European overview - Techniques

STFC Daresbury Laboratory

HiPIMS

TU Darmstadt

Dual cathode DCMS

INFN Legnaro

DCMS, dipped target

CERN Geneva

Bipolar HiPIMS

STFC Daresbury Laboratory

Samples / QPR

TU Darmstadt

Samples

INFN Legnaro

Samples, QPRs … soon cavities

CERN Geneva

Samples, QPRs, RADES, Magnetic flux lens discs

Experimental Setup

Our point of view:

Experimental Setup

Our point of view: HiPIMS is a MUST (as discussed yesterday)

Experimental Setup

Our point of view: HiPIMS is a MUST (as discussed yesterday)

However:

- Nb_3Sn forms at rather low energy level (~750°C) vs HiPIMS bias (10's eV)

- Biasing is prompt to damage the lattice (see yesterday talk)

- Bias is needed for smoothness, density control

- Heat treatment is required to form proper A15 phase AND to recover defects

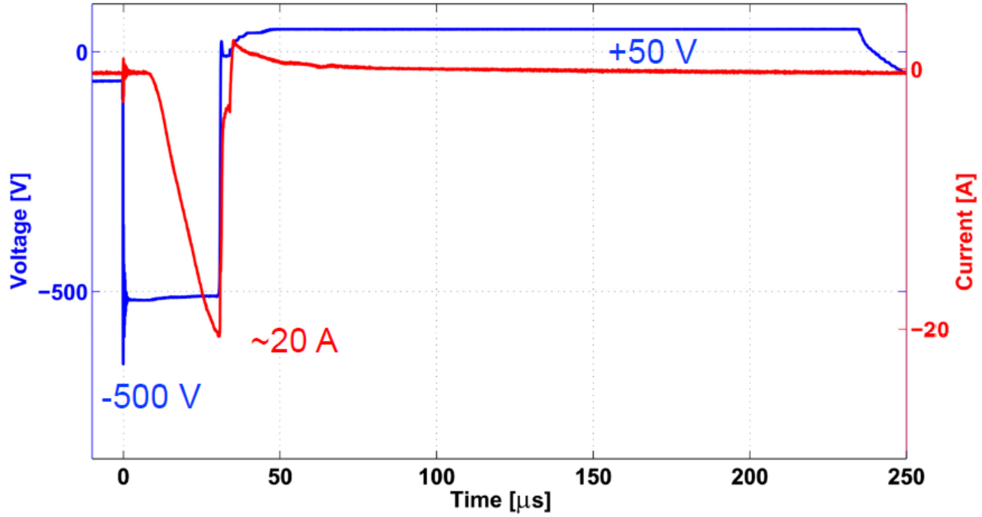
Experimental Setup

Bi-Polar HiPIMS (Energy Pulse Systems)

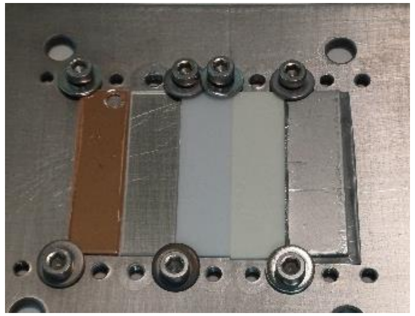
- Two magnetrons 150mm diameter
- Working distance (100mm)
- Base pressure ~10^{-10} mbar after BO
- Target: alloyed Nb_3Sn
- Heater: resistive, home-made

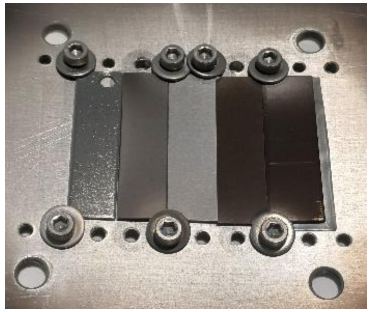
Coating parameters:

- *Gas:* Kr
- *T_s:* 500 … 750°C
- *P:* $7 \cdot 10^{-4}$ … $5 \cdot 10^{-2}$ *mbar*
- *PP:* 35 … 100 V
- *Post anneal:* 0 … 72 hrs



Feasibility - T_c

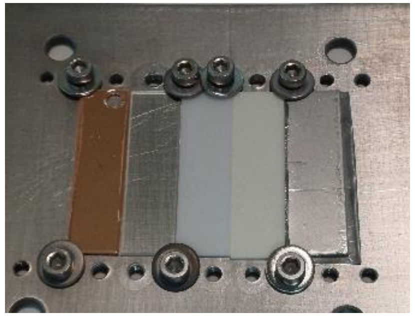


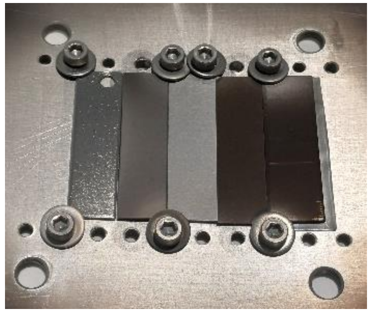


Can we elaborate high quality Nb_3Sn films?

Can we reach 18.3K Tc? First good sign of longe range order within the material lattice

Feasibility - T_c





Can we elaborate high quality Nb_3Sn films?

Can we reach 18.3K Tc? First good sign of longe range order within the material lattice

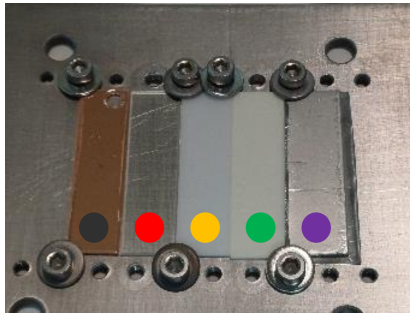
DCMS

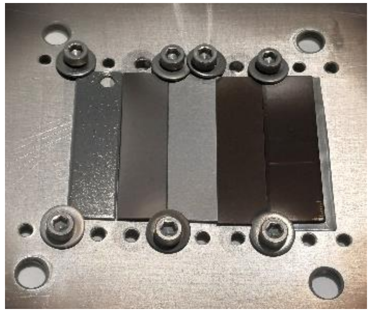
Table 1. Influence of the substrate on critical temperature of the Nb_3Sn films.

	Critical temperature (K)	
Process pressure	Copper substrate	Ceramic substrate
$P = 1 \times 10^{-3}$ mbar	14.7 K	17.5 K
$P = 5 \times 10^{-2}$ mbar	15.5 K	17.4 K

https://doi.org/10.1088/1361-6668/aaf61f

Feasibility - T_c





Can we elaborate high quality Nb_3Sn films?

Can we reach 18.3K Tc? First good sign of longe range order within the material lattice

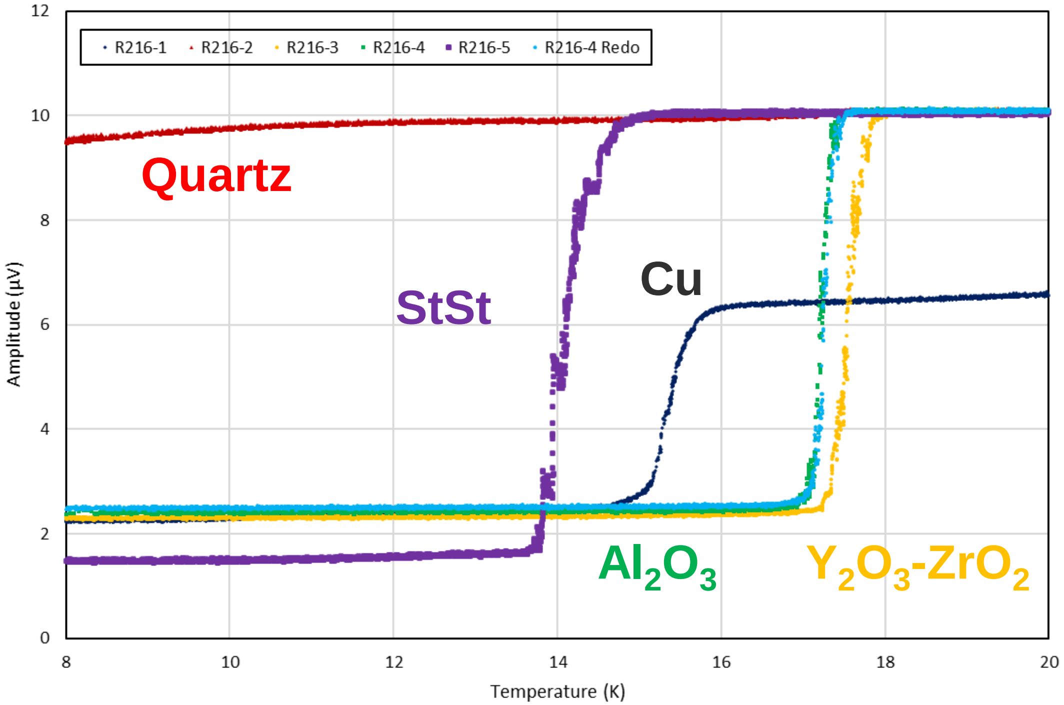
DCMS

Table 1. Influence of the substrate on critical temperature of the Nb_3Sn films.

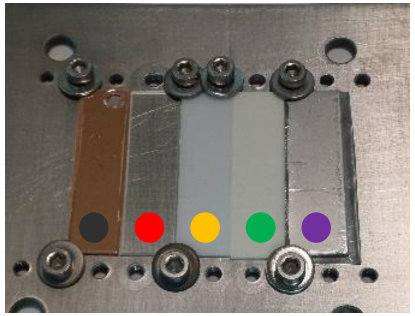
Process pressure	Critical temperature (K) Copper substrate	Critical temperature (K) Ceramic substrate
$P = 1 \times 10^{-3}$ mbar	14.7 K	17.5 K
$P = 5 \times 10^{-2}$ mbar	15.5 K	17.4 K

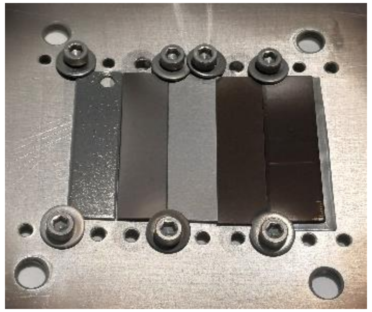
https://doi.org/10.1088/1361-6668/aaf61f

HiPIMS



Feasibility - T_c





Can we elaborate high quality Nb_3Sn films?

Can we reach 18.3K Tc? First good sign of longe range order within the material lattice

DCMS

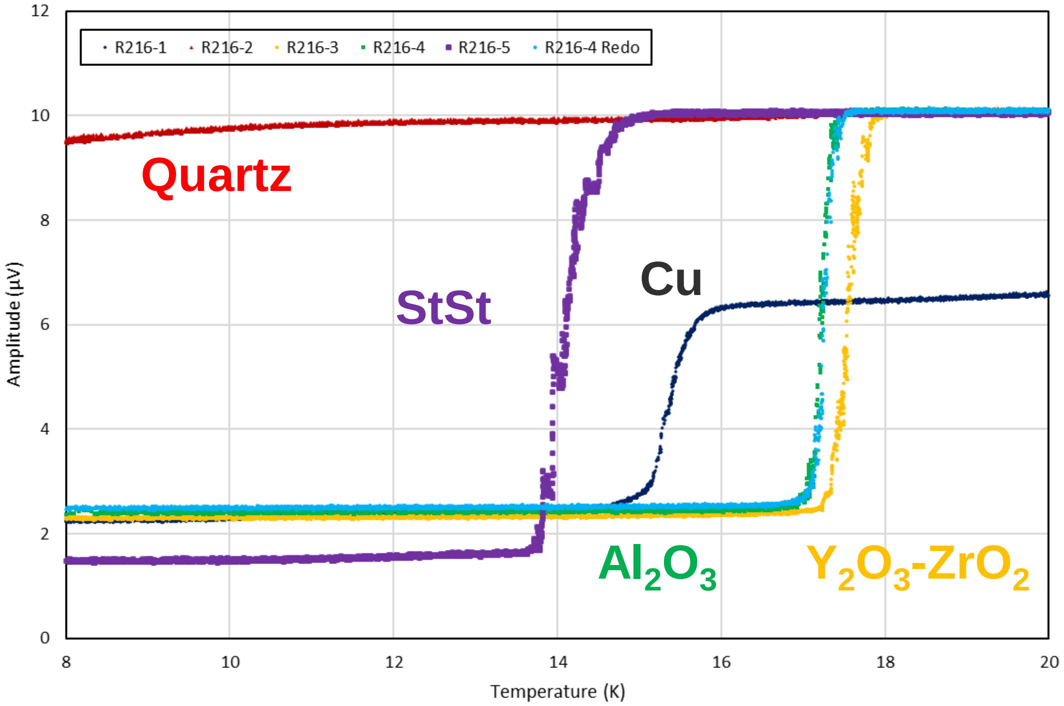
Table 1. Influence of the substrate on critical temperature of the Nb_3Sn films.

	Critical temperature (K)	
Process pressure	Copper substrate	Ceramic substrate
$P = 1 \times 10^{-3}$ mbar	14.7 K	17.5 K
$P = 5 \times 10^{-2}$ mbar	15.5 K	17.4 K

https://doi.org/10.1088/1361-6668/aaf61f

SUBSTRATE'S THERMAL EXPENSION COEFFICIENT DRIVES THE RESIDULA STRESS → LRO → Tc

HiPIMS

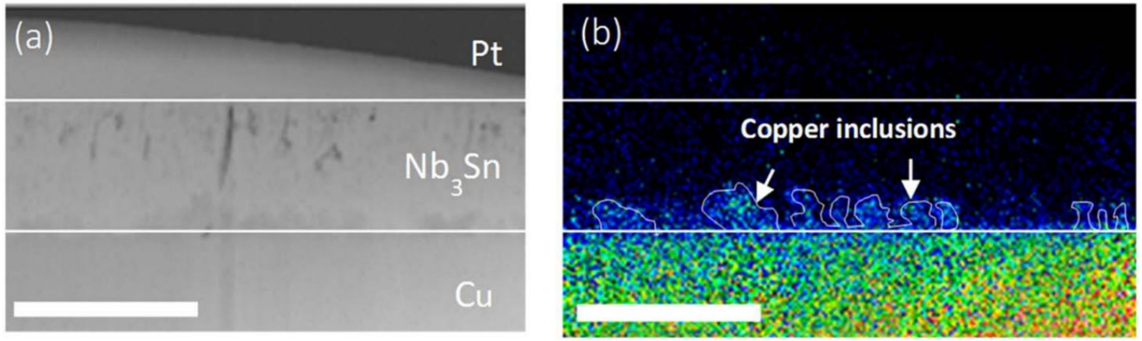


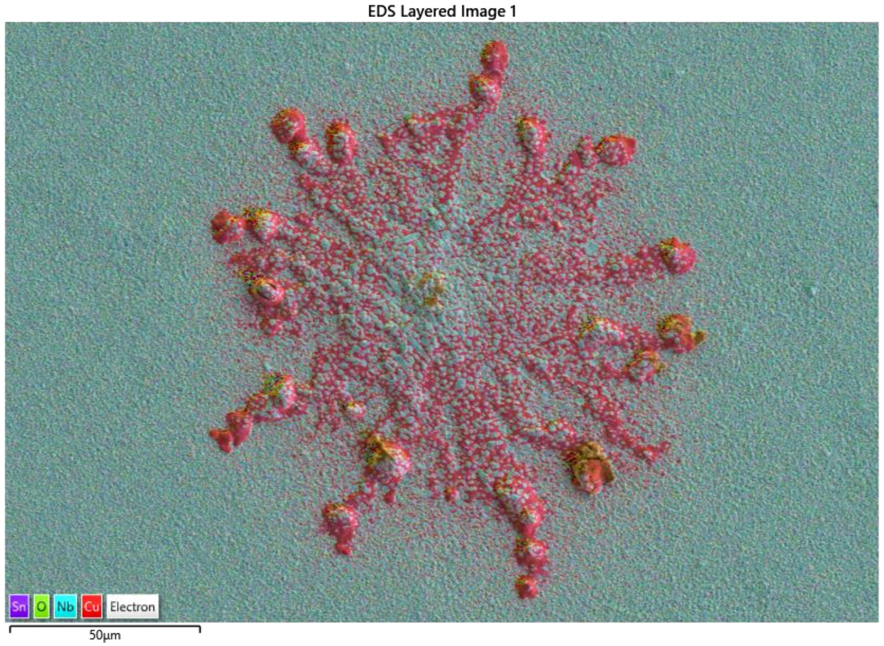
Feasibility – Cu

How problematic is Cu?

Interdiffusion → NC spots at the RF surface

Also promoting the A15 phase formation





Feasibility – Cu

How problematic is Cu?

Interdiffusion → NC spots at the RF surface

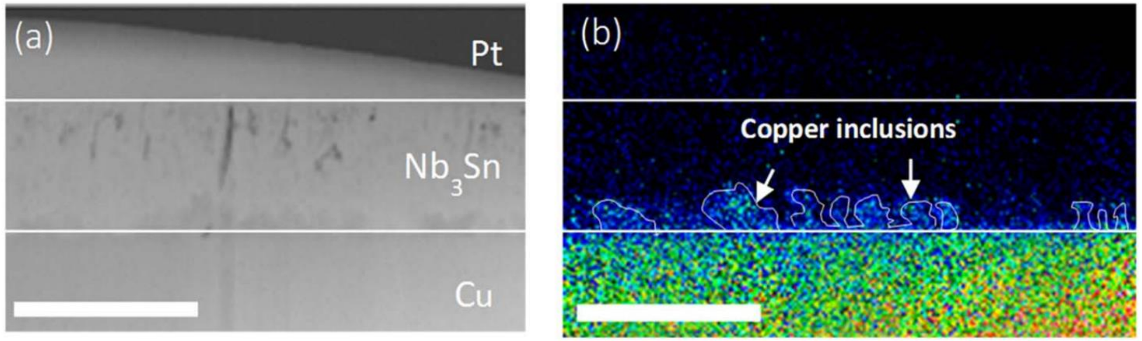
Also promoting the A15 phase formation

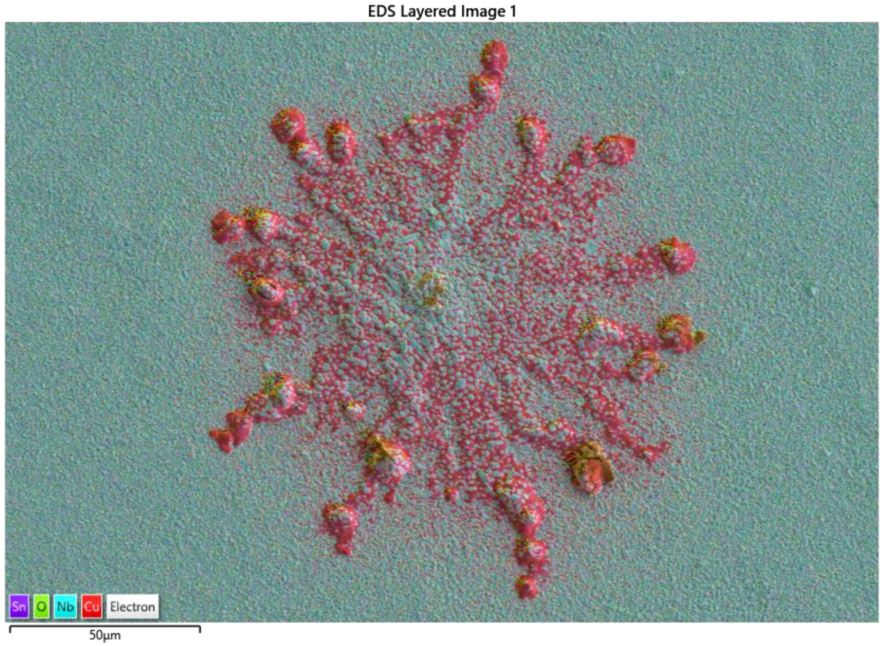
How to block it?

Diffusion barrier layer (Ta, HEA …)

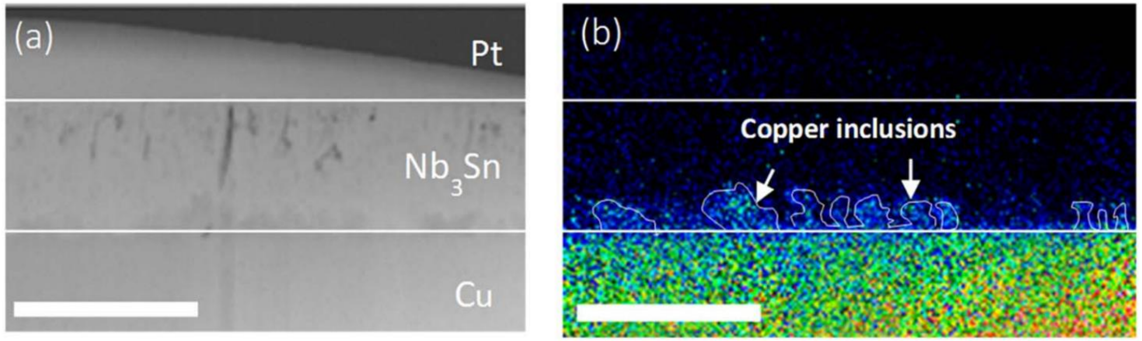
Ta: requires a very specific crystalline phase (α)

HEA: promising, needs more investigations, amorphous even at high T





Feasibility – Cu



How problematic is Cu?

Interdiffusion → NC spots at the RF surface

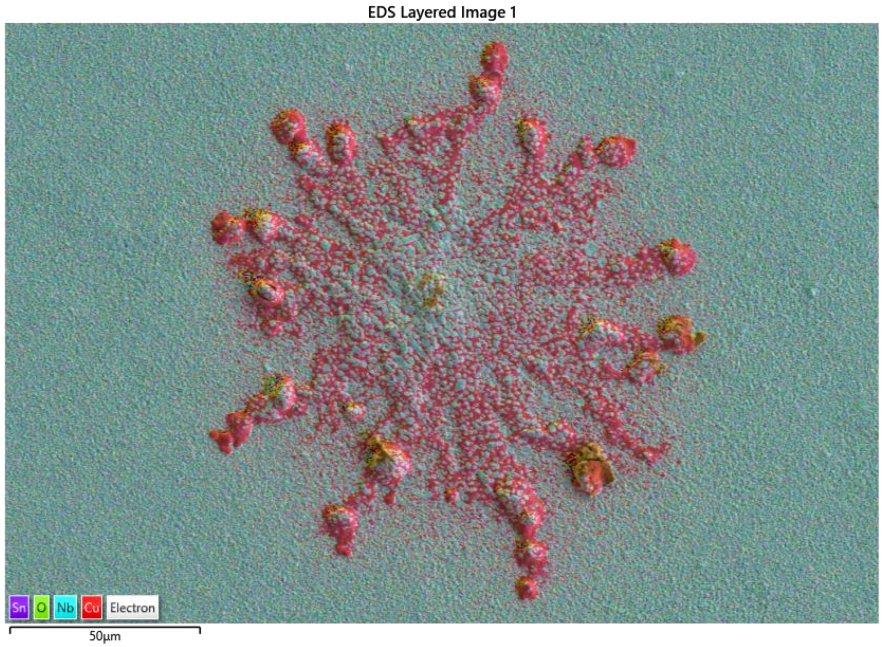
Also promoting the A15 phase formation

How to block it?

Diffusion barrier layer (Ta, HEA …)

Ta: requires a very specific crystalline phase (α)

HEA: promising, needs more investigations, amorphous even at high T



Then… the surface is absolutely free of Cu?

Feasibility – Cu

How problematic is Cu?

Interdiffusion → NC spots at the RF surface

Also promoting the A15 phase formation

How to block it?

Diffusion barrier layer (Ta, HEA …)

Ta: requires a very specific crystalline phase (α)

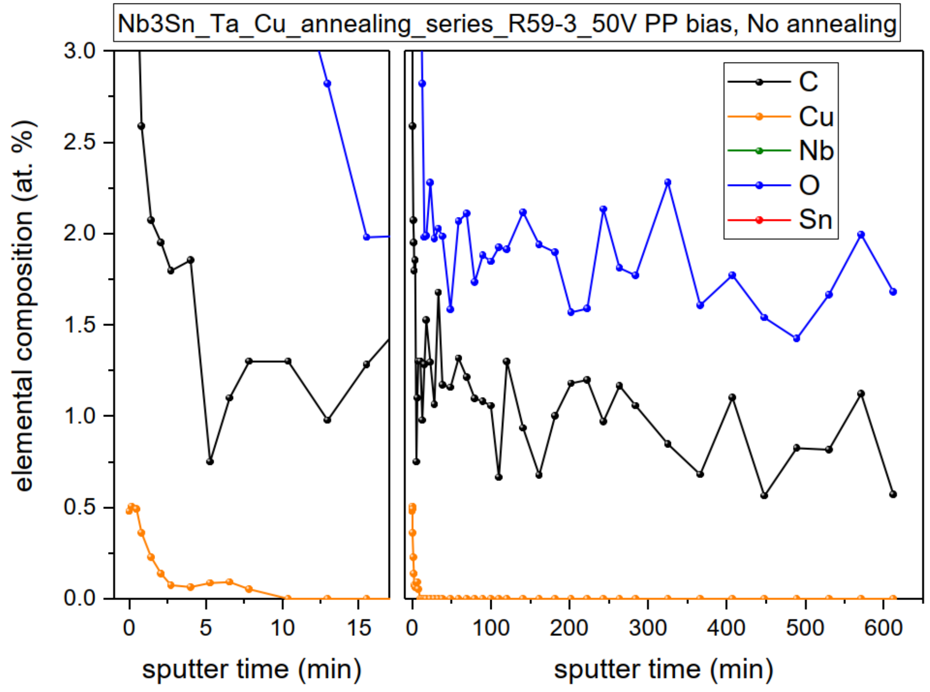
HEA: promising, needs more investigations, amorphous even at high T

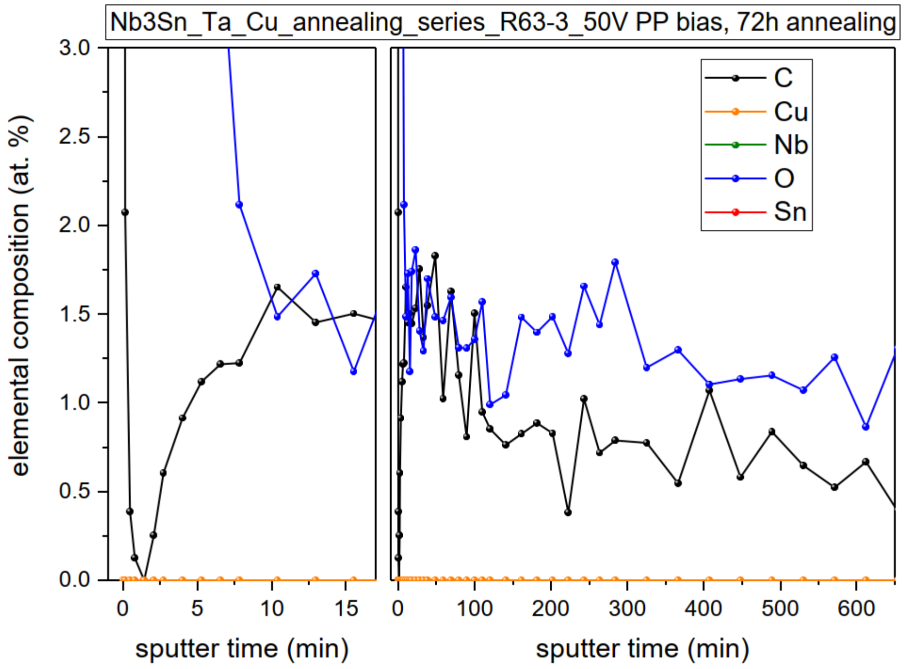
Then… the surface is absolutely free of Cu?

No

Surface contamination.

Disappears after HT.





Feasibility – Cu

Also observed:

A careful setup cleaning reduces surface Cu contamination.
Surface diffusion from samples
A problematic already known with dramatic industrial consequences

For future applications and scale-up

How to manage this contamination?

Curative approach: post coating Cu wet etching : Ammonium persulfate

Cu at. %	Pre Rinse	Post Rinse
Sample 1	3.9	0.6
Sample 2	5.3	1.6

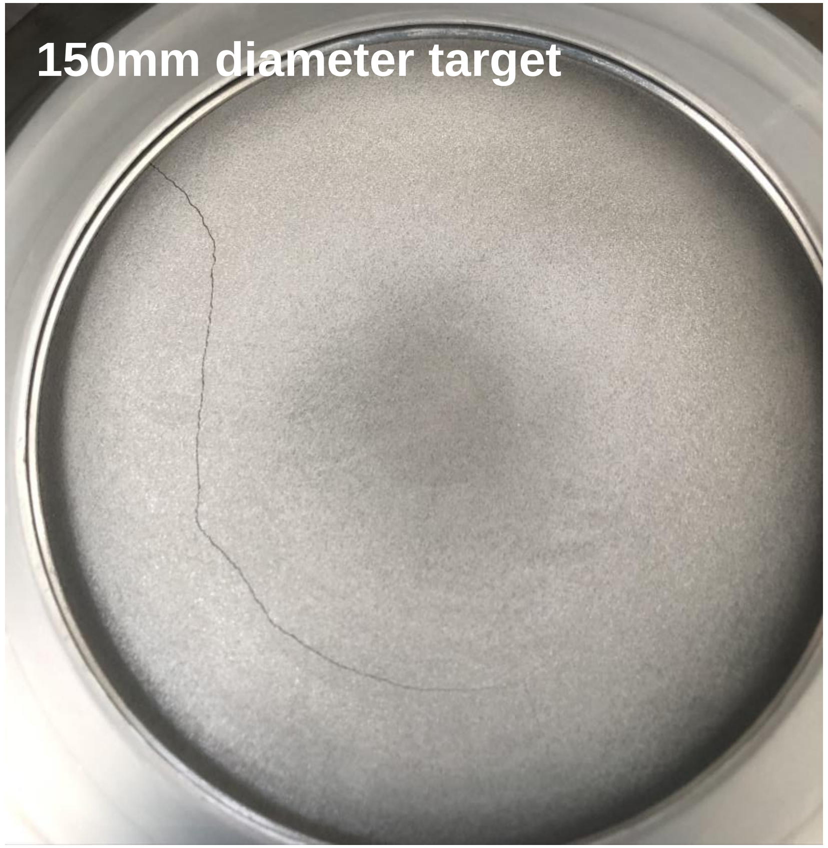
Target management

CERN's choice:

alloyed target : reduce the needs down to 2 targets: 1 for DBL and 1 for Nb3Sn

Problematic:

Fragile
Prompt to cracking
Dust creation
Impossibility to buy cylindrically shaped targets



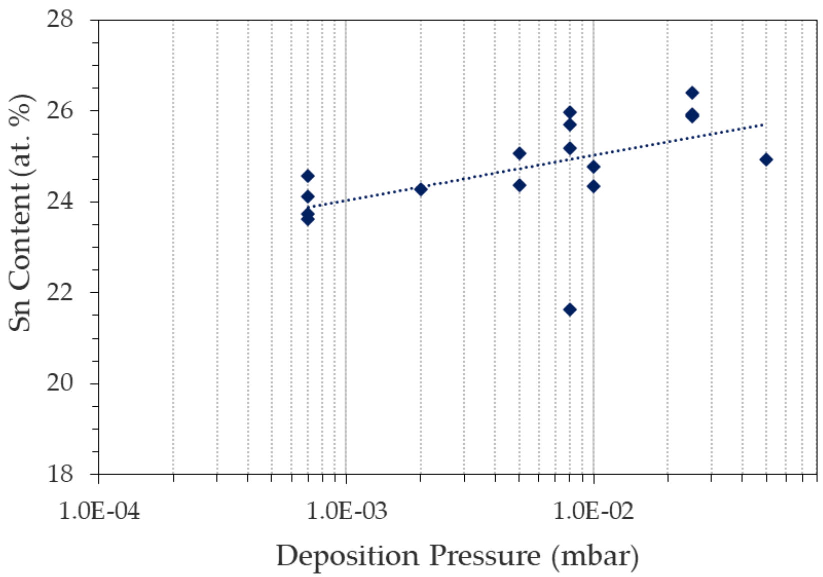
Study on going to adress this specific issue:

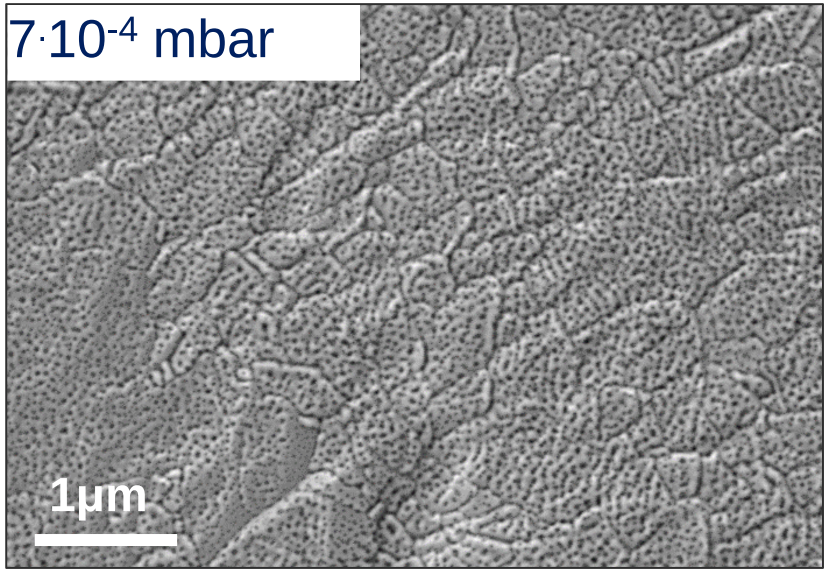
Thermal cycles
Power ramp-rate
Max power
Backing plate bonding strategy

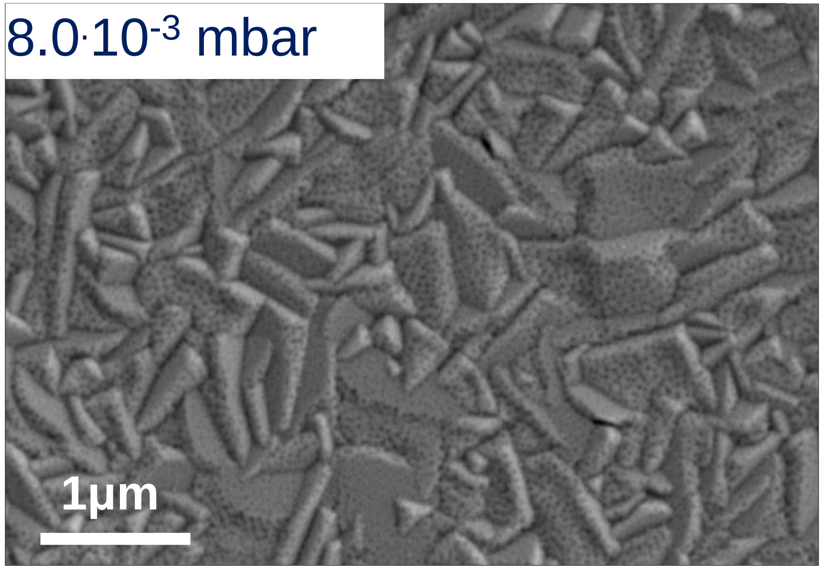
Some process parameters' effects

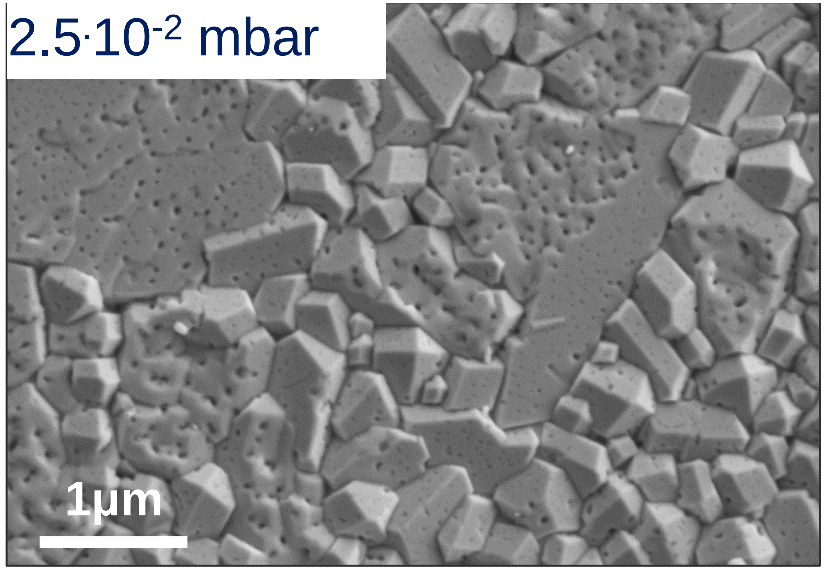
Coating pressure:

- Smoother at lower pressure (less gas phase collisions)
- Composition: Sn content increases with P
- An optimal has to be found between composition and roughness
- Films look as well more crystalline at high P (XRD quantitative analysis to be performed)





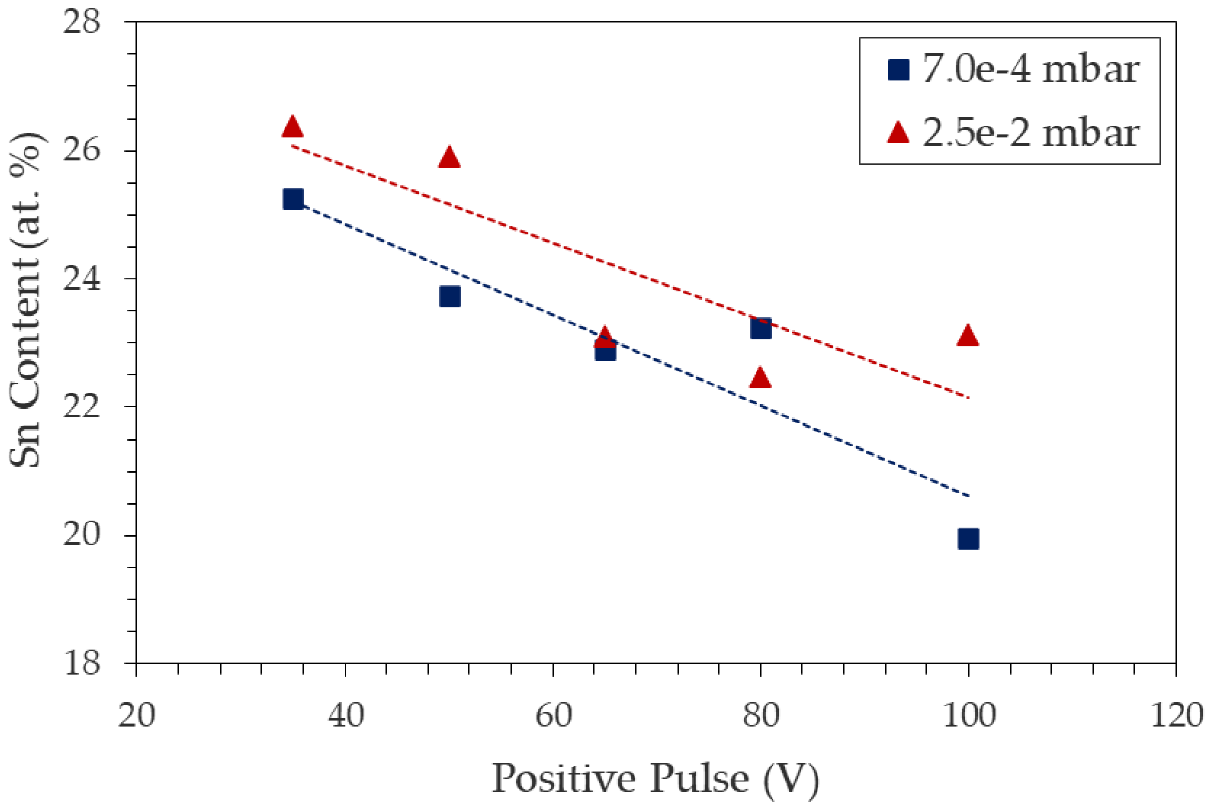




Some process parameters' effects

Positive Pulse:

- Sn very sensitive to re-sputtering
- Sn content decreases with positive pulse voltage
- Composition and density evolve on an opposite trend

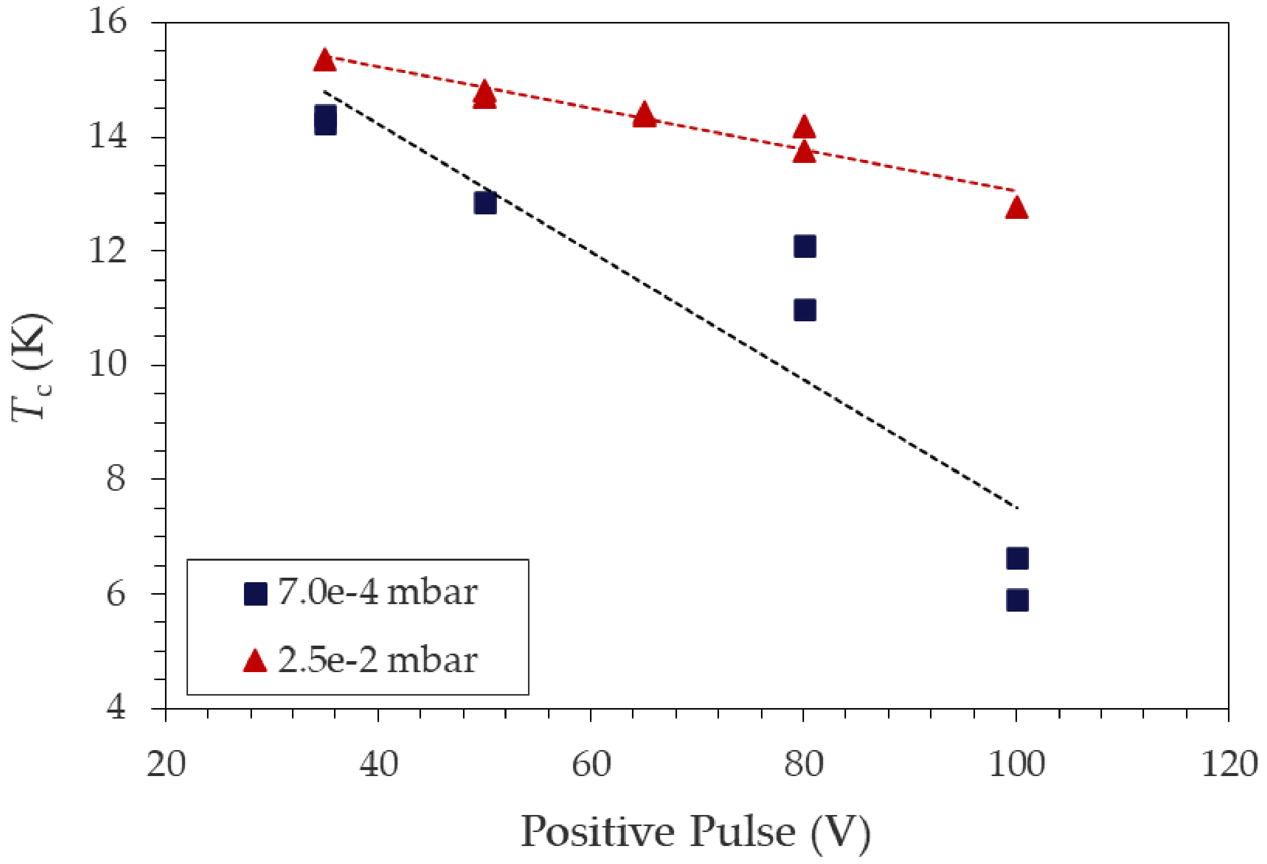


Some process parameters' effects

Effect on the Tc

- Increase in pressure → increase in Sn content → increase in T_c

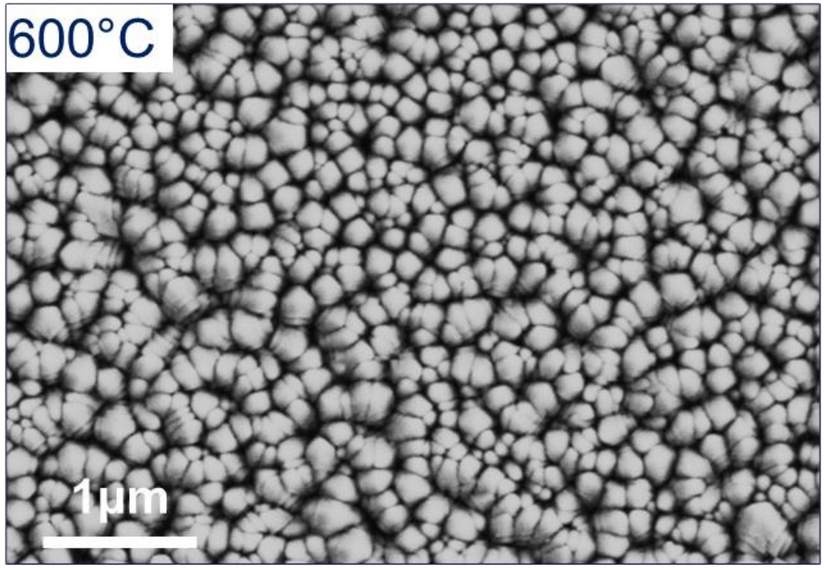
- Increase in PP → decrease in Sn content → decrease in T_c

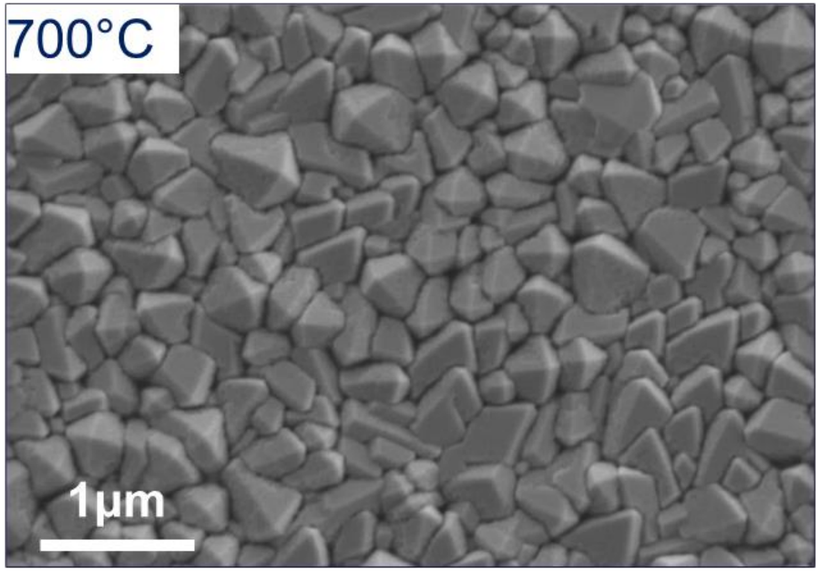


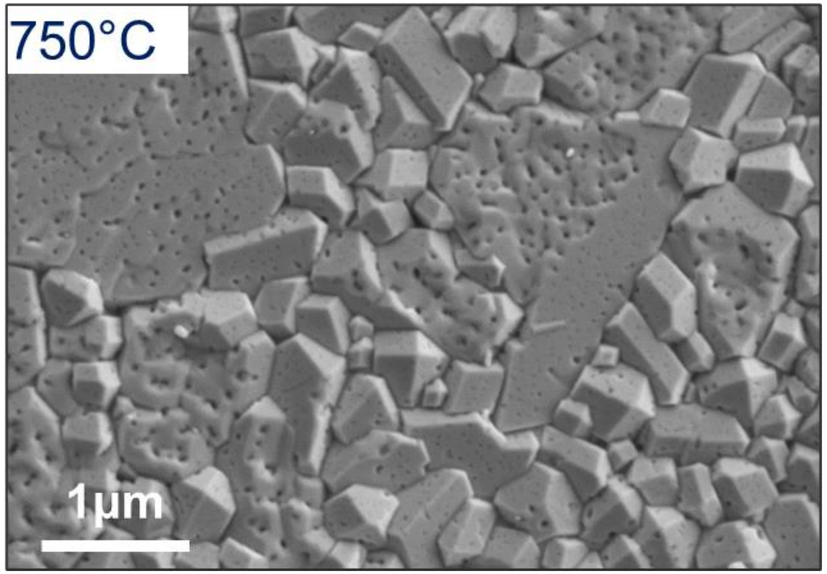
Some process parameters' effects

Coating temperature

- For a given coating recipe
- Lower temperature → lower adatoms mobility
  - Porous layer
  - Layer appears much less crystalline







Macroscopic defects library

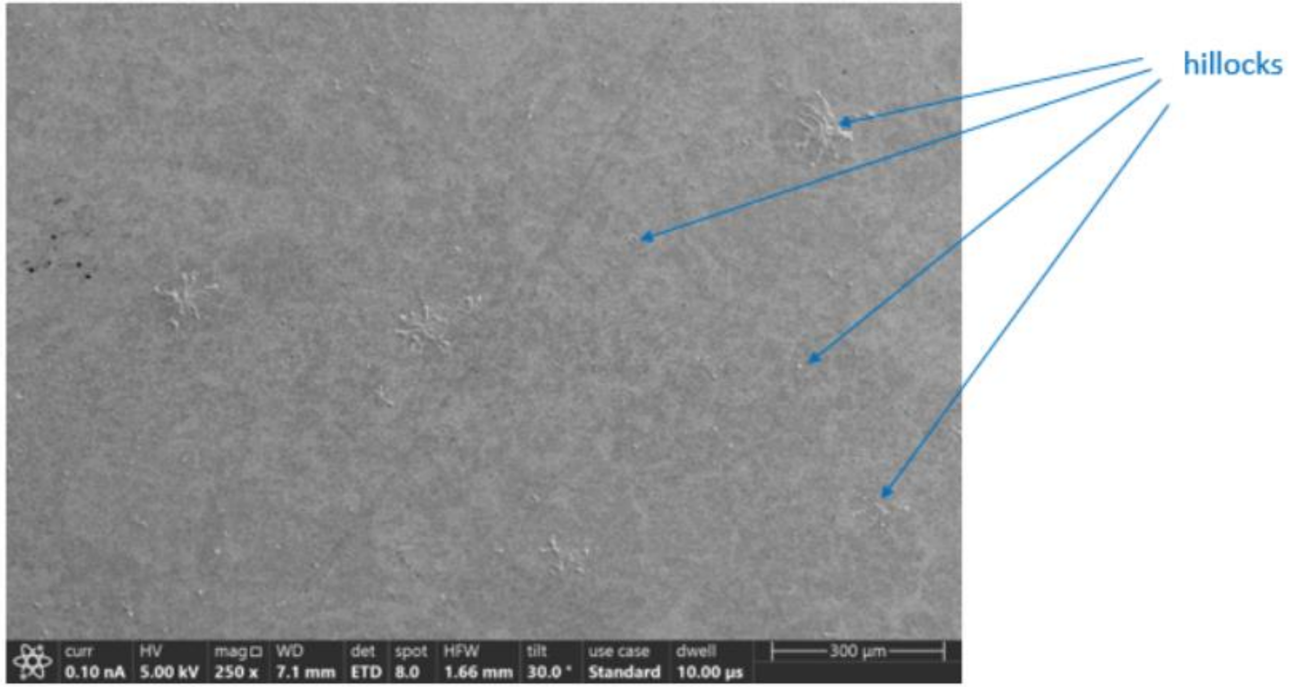
Problematic: very time consuming to perform defect analysis (FIB-SEM, cross sections, TEM …)

Solution: Establish a catalogue of visual defects that can be linked to microscopic feature in view of speeding process qualification → avoid unnecessary investigations

Macroscopic defects library

Problematic: very time consuming to perform defect analysis (FIB-SEM, cross sections, TEM …)

Solution: Establish a catalogue of visual defects that can be linked to microscopic feature in view of speeding process qualification → avoid unnecessary investigations

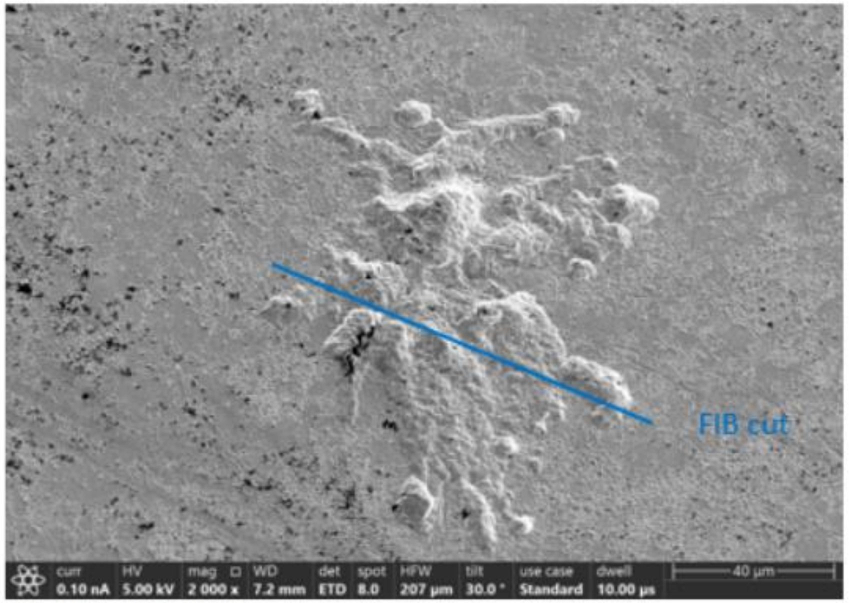


SEM image: surface with defects

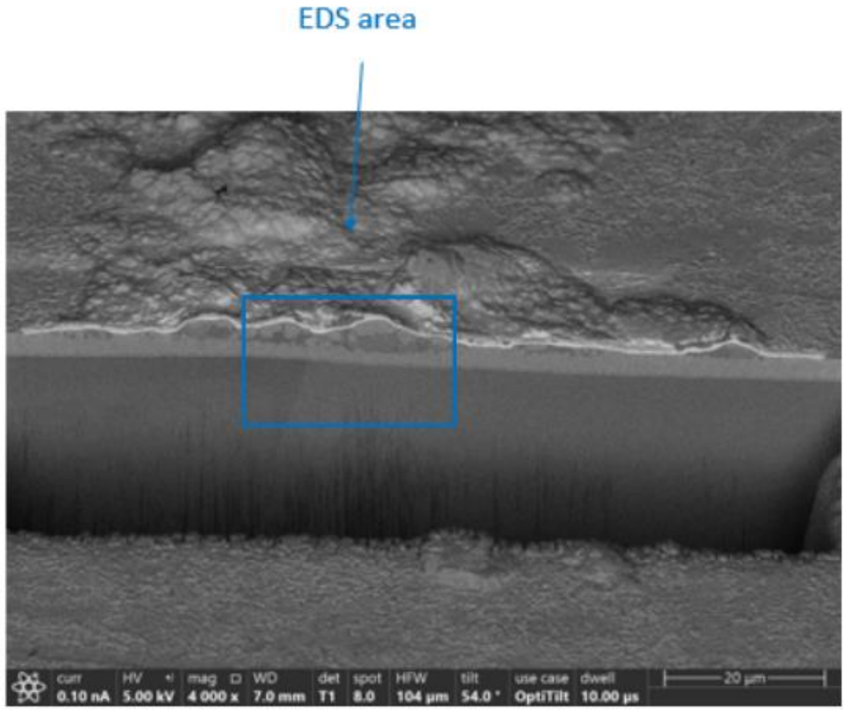
Macroscopic defects library

Problematic: very time consuming to perform defect analysis (FIB-SEM, cross sections, TEM …)

Solution: Establish a catalogue of visual defects that can be linked to microscopic feature in view of speeding process qualification → avoid unnecessary investigations



SEM image: hillock



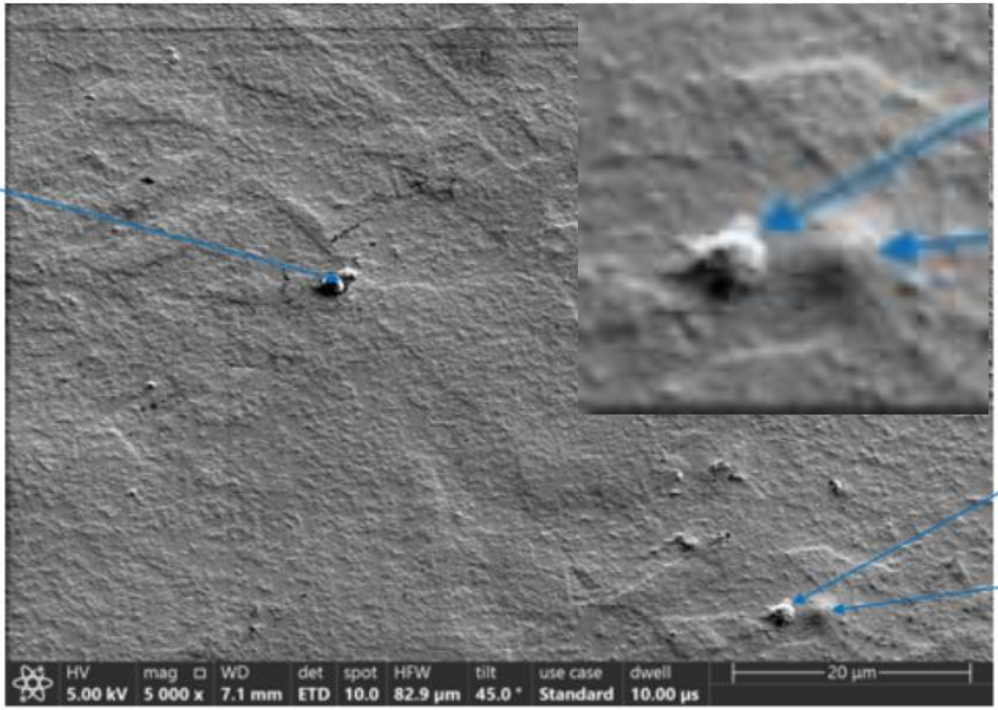
SEM image: FIB x-section through the hillockck

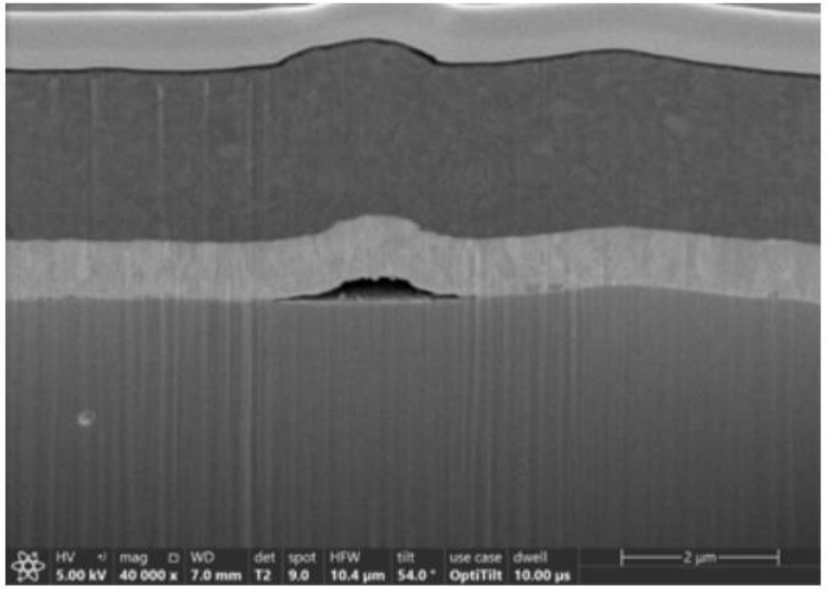
DBL failure
- Stress induced crack
- dust

Macroscopic defects library

Problematic: very time consuming to perform defect analysis (FIB-SEM, cross sections, TEM …)

Solution: Establish a catalogue of visual defects that can be linked to microscopic feature in view of speeding process qualification → avoid unnecessary investigations





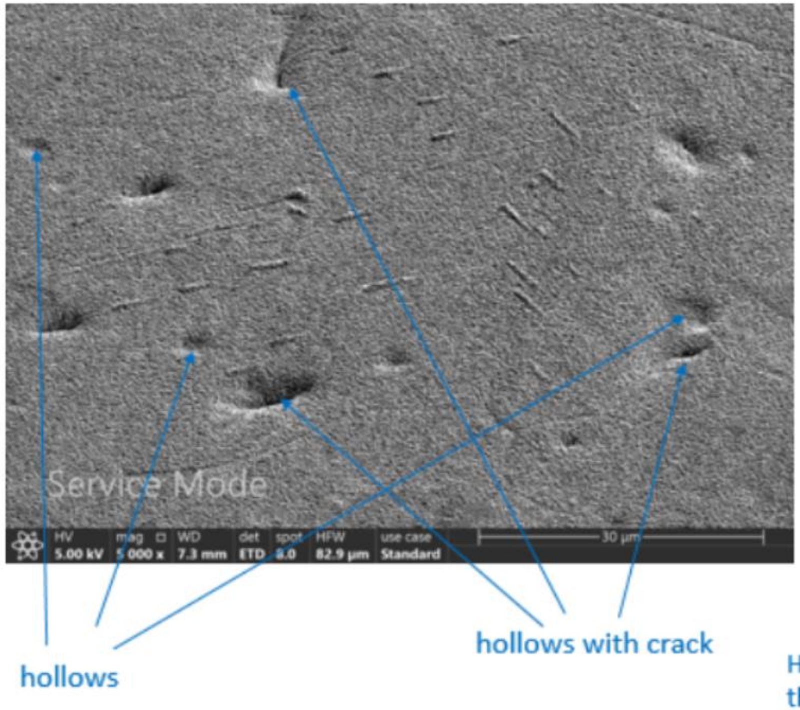
FIB cross section through the hillock and tapered hillock

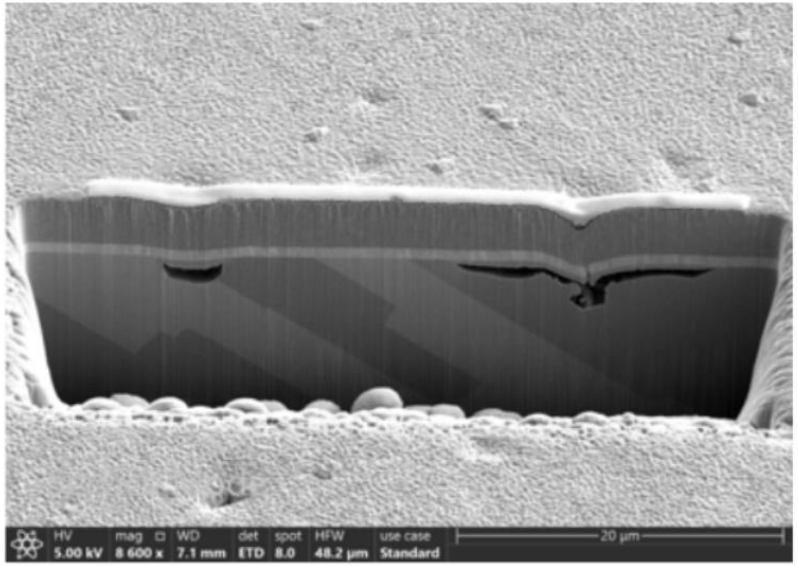
Surface particulate contamination
Stress accumulation

Macroscopic defects library

Problematic: very time consuming to perform defect analysis (FIB-SEM, cross sections, TEM …)

Solution: Establish a catalogue of visual defects that can be linked to microscopic feature in view of speeding process qualification → avoid unnecessary investigations





Cross-section through a hollow and hollow with crack

Hollows and hollows with crack are associated with voids or maybe blowholes in the Cu substrate.

Cu diffusion.
Already seen in HIE-ISOLDE

Typical of high temperature processes

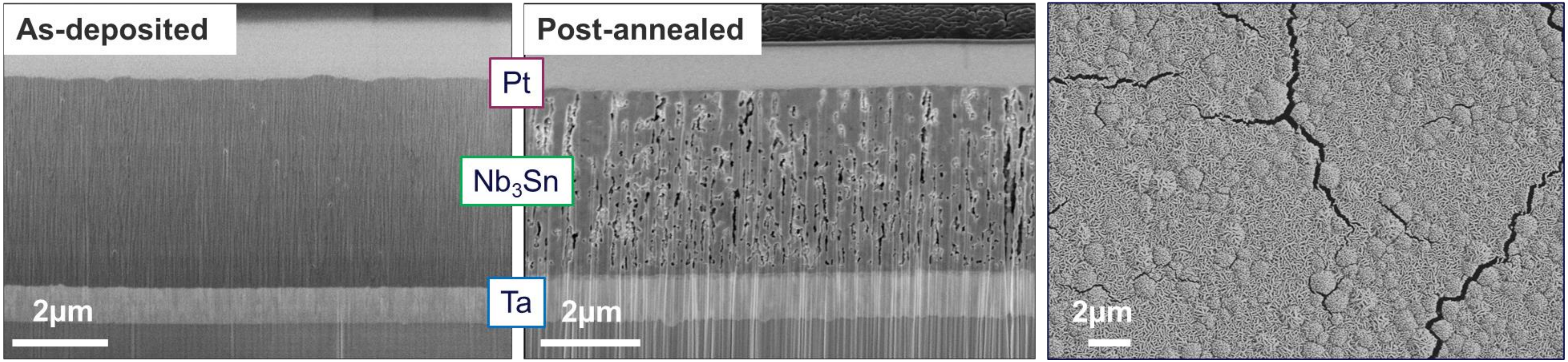
→ Layer fragility?

Side Note

Coating at low T and reacted after coating

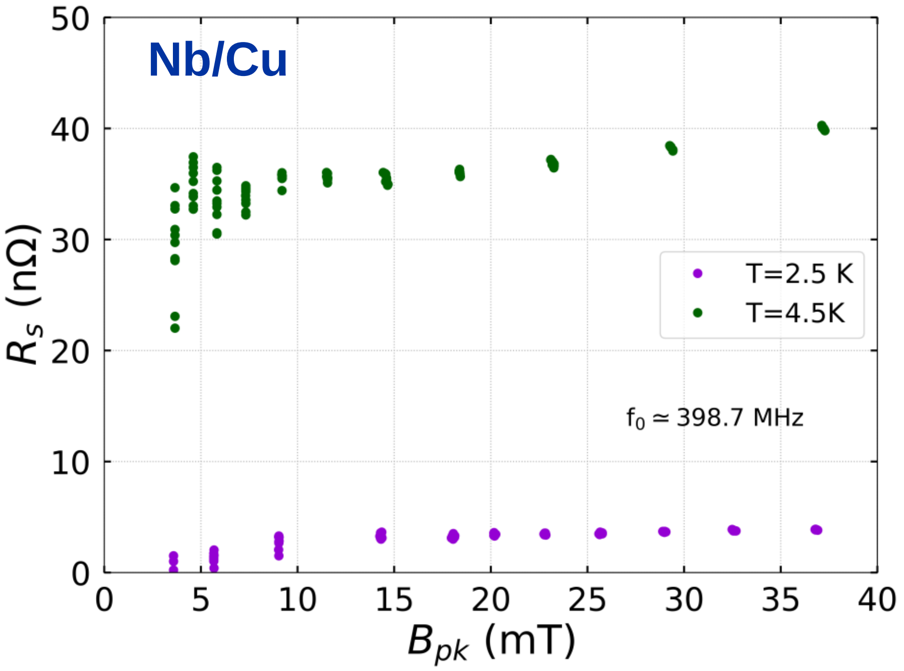
SYSTEMATICALLY LEADS TO FILM CRACKING

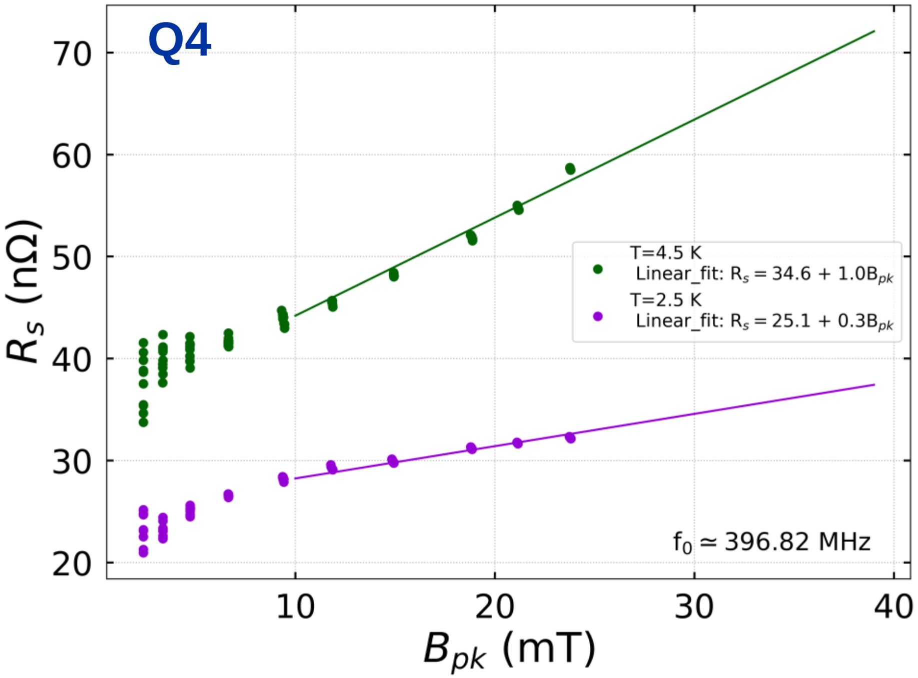
This option is for the moment put aside as a potential process route

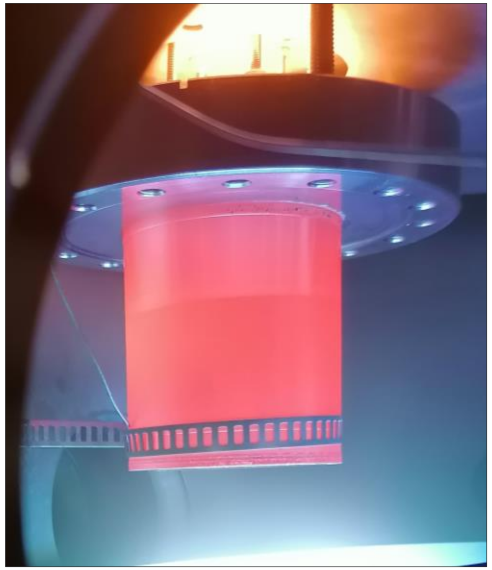


QPR samples tests – RF data

Technical remark: heating up a QPR to a suitable temperature is an actual challenge.
Right now: only home-made heater has shown satisfactory behavior.

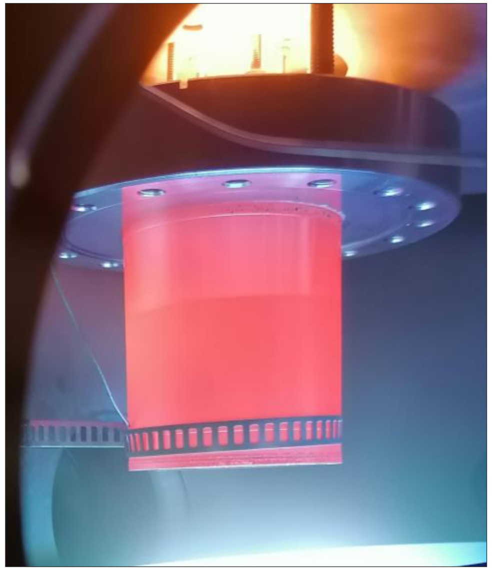


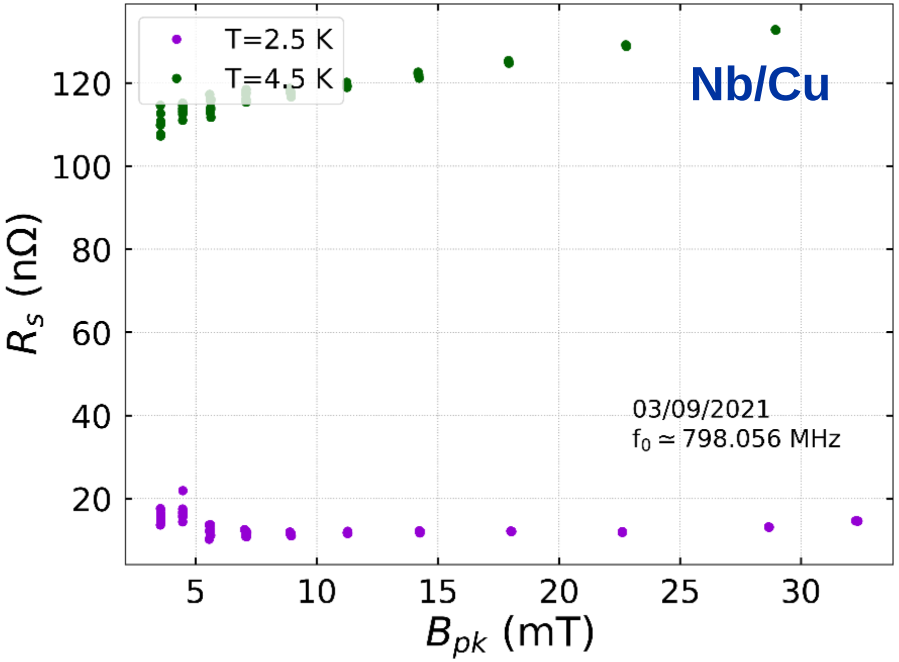


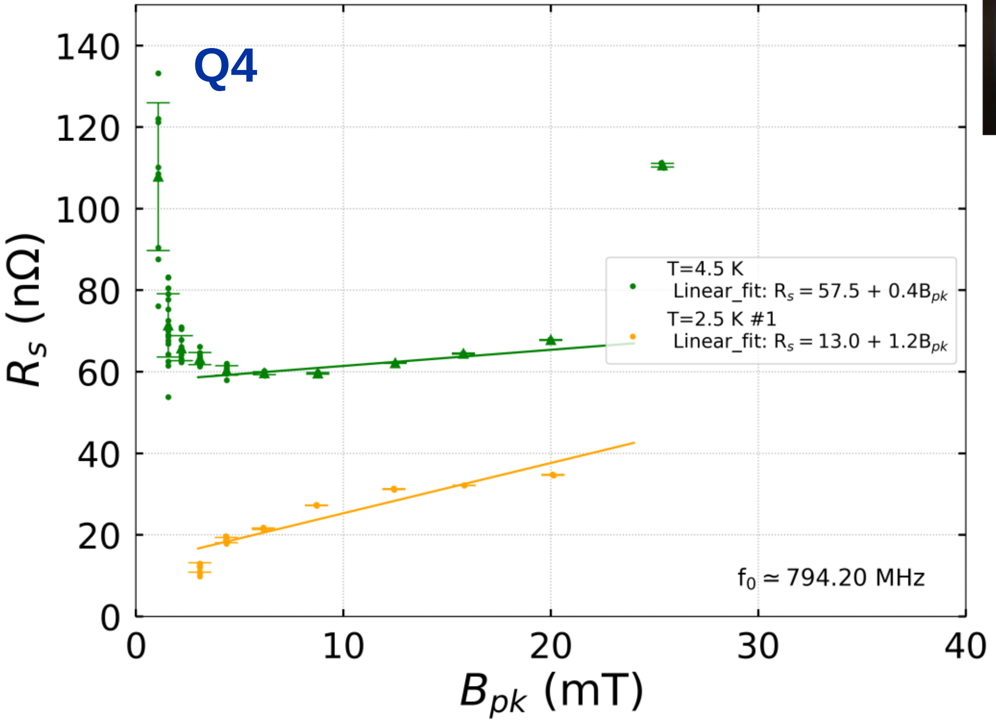


QPR samples tests – RF data

Technical remark: heating up a QPR to a suitable temperature is an actual challenge.
Right now: only home-made heater has shown satisfactory behavior.







RADES– RF data

Technical remark: heating up a QPR to a suitable temperature is an actual challenge.
Right now: only home-made heater has shown satisfactory behavior.

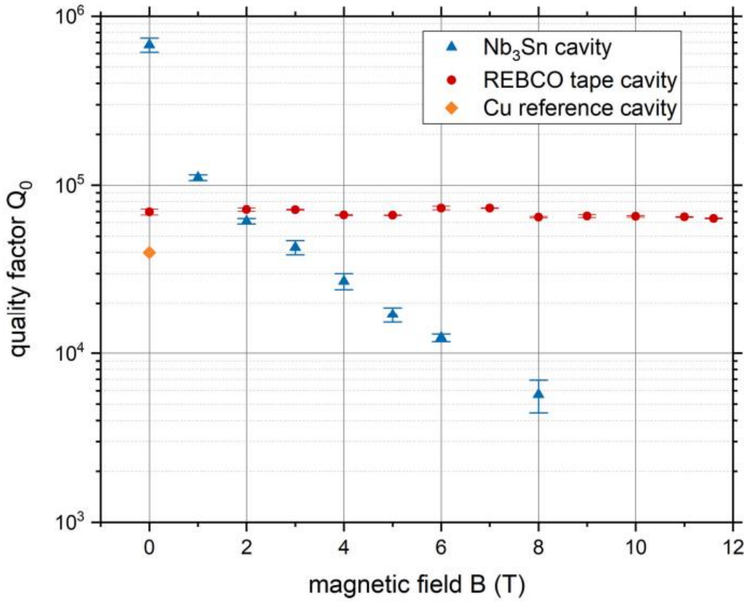
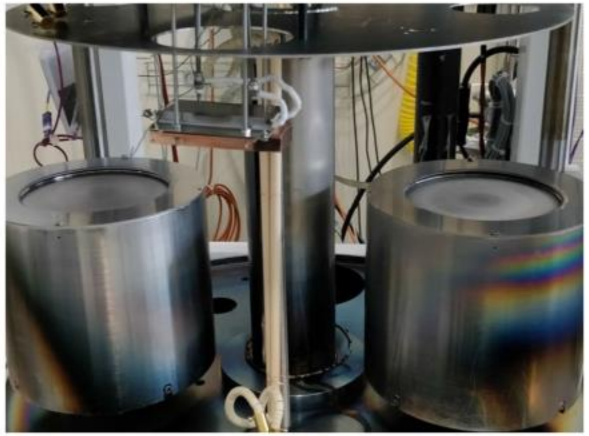


Fig. 6. Results of quality factor measurements with the cavity immersed in liquid helium. The resonance frequency of the cavities is f = (8.8 ± 0.1) GHz at 4.5 K.

TABLE I
COATING PARAMETERS FOR THE Ta AND Nb_3Sn LAYERS

	t (min)	p (mbar)	Pw (W)	T (°C)	Main pulse t (μs)	Main pulse ν (kHz)	Positive Pulse t (μs)	Positive Pulse Δt (μs)
Ta	40	1×10^{-3}	350	750	50	1	200	4
Nb_3Sn	75	7×10^{-4}	350	750	-	-	-	-



(a)

(b)

Fig. 3. Photographs of the (a) coating setup and (b) a cavity half in coating position.

J. Golm *et al.*, "Thin Film (High Temperature) Superconducting Radiofrequency Cavities for the Search of Axion Dark Matter," in *IEEE Transactions on Applied Superconductivity*, vol. 32, no. 4, pp. 1-5, June 2022, Art no. 1500605, doi: 10.1109/TASC.2022.3147741.

QPR samples tests – RF data

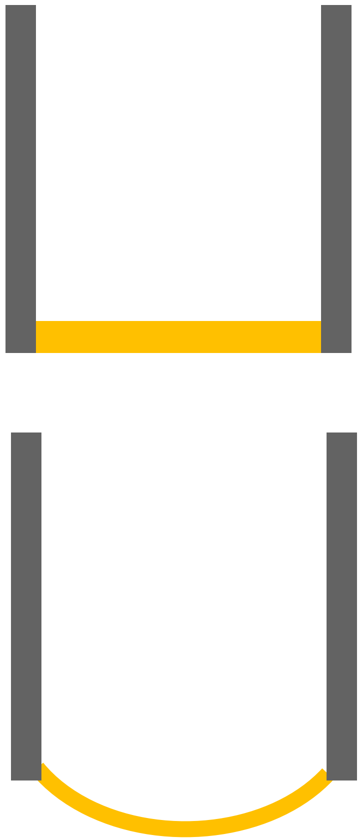
BE CAREFUL:

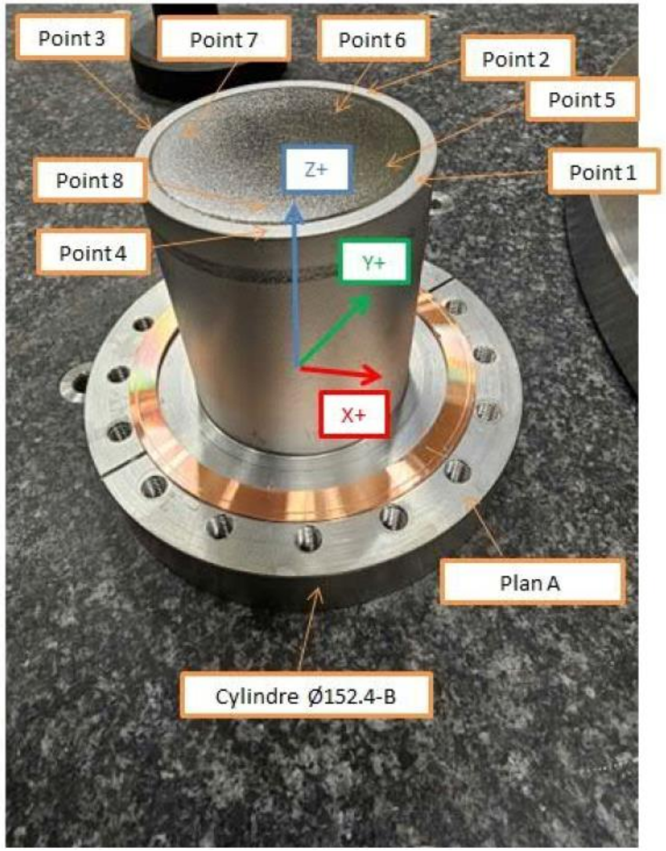
High temperature during coating can lead to the mechanical deformation of the QPR surface → bulging

Serious impact onto the RF surface resistance estimation

Full surface metrology is **MANDATORY** after RF test to feedback the actual profile to RF simulation.

Depending on how you mount the sample: surface can either collapse, bulge or even the Nb tube could buckle (horizontal position)

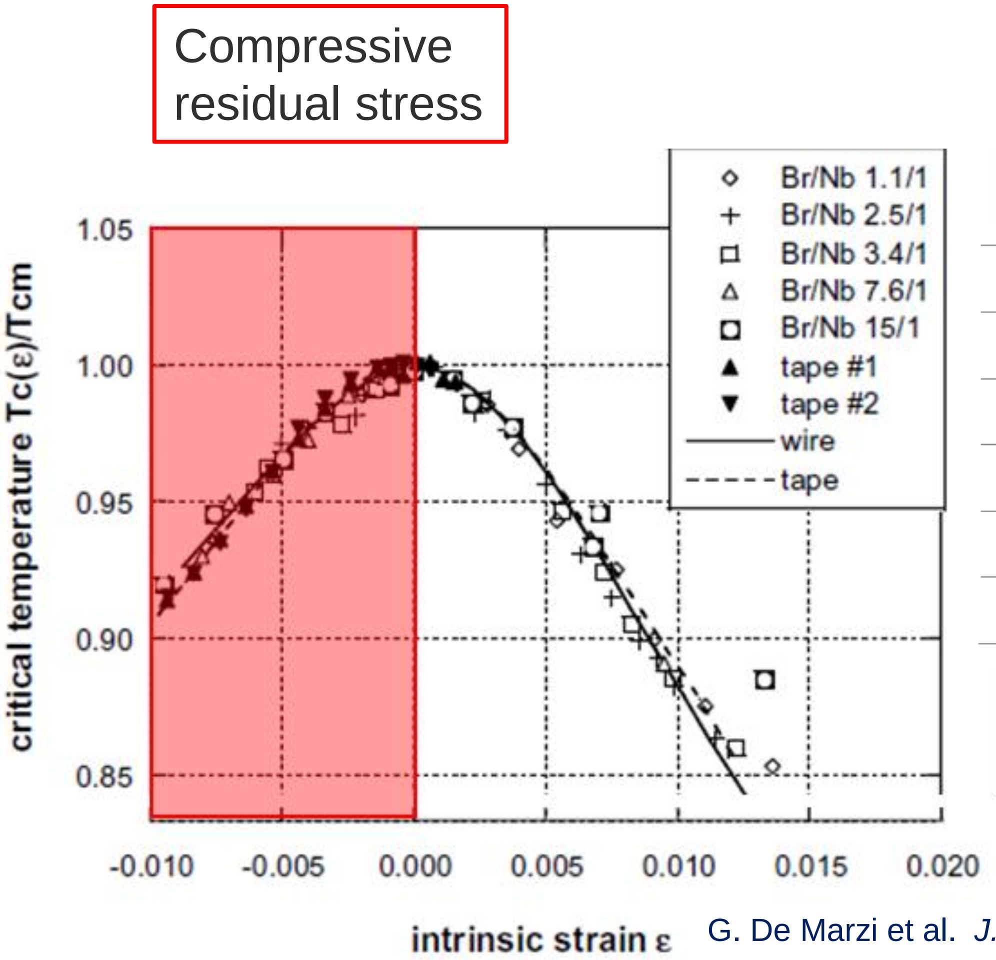




Regular surface re-machining might be a standard to apply

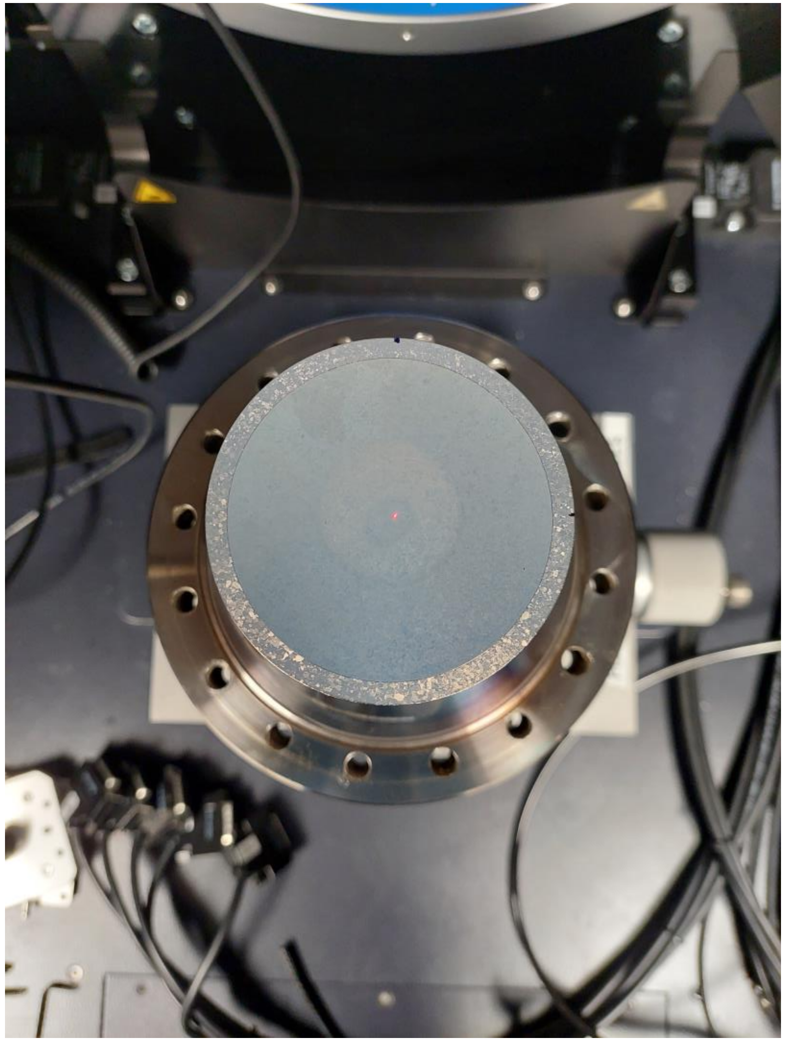
QPR samples tests – Material analysis

Residual stress quantification by XRD analysis



G. De Marzi et al. *J. Phys.: Condens. Matter* **25** (2013)

Residual stress by XRD (MPa)
Centre #1: -888 ± 20
Centre #2: -760 ± 20
Outer #1: -975 ± 19
Outer #2: -949 ± 13
Outer #3: -1236 ± 17



Scale-up strategy at CERN

1. One have to confirm the competitiveness of Nb_3Sn/Cu wrt Nb/Cu

2. Continue with QPR testing and process optimization

3. Objects to be coated: last SRF CERN workshop consensus on moving on straight to 800MHz cavities. (potentialy compatible with 1.3GHz)

4. The cost of such a coating hardware will be > 500kCHF : one should target for success

5. Sub-components (target) shall be comissioned and validate prior full integration

6. New CERN SRF building to be «furnished» in 2029

Outlook

- Nb_3Sn/Cu is an attractive system to compete with Nb/Cu
- 800MHz performance appear promising
- Linear thermal dilatation coefficient difference between substrate and film remains an issue : look for another substrate?
- Cu interdiffusion can be controlled/mitigated. Does not appear yet as a showstopper.
- High temperature is a pre-requisite DURING coating
- Target fragility has to be handled for a proper scale-up: could be a showtopper
- RF tests on QPR are showing performance close to Nb/Cu: there is room for hope