Thin films developments for RF cavities at CEA

Yasmine Kalboussi on behalf of the CEA Saclay Team

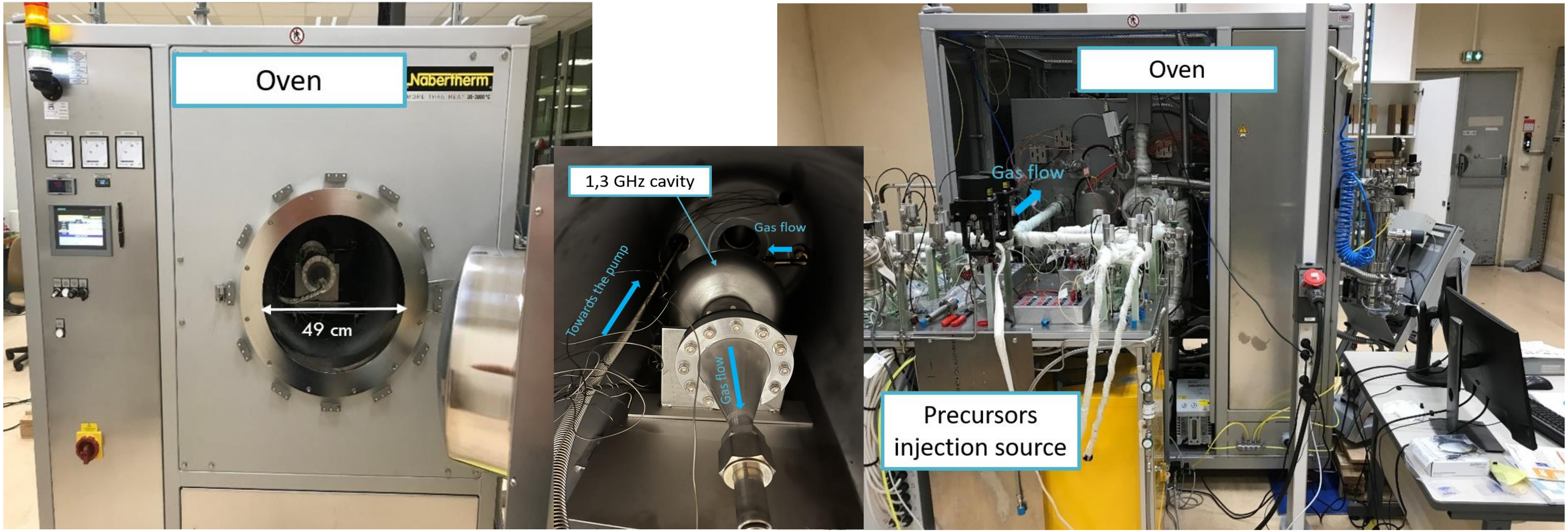
FCC week 2025 –Vienna 19-23 May

CEA Saclay Team

- Yasmine Kalboussi permanent staff – ALD / Surface characterization
- Ivana Curci – Thesis (3nd year) – Tunneling spectroscopy / ALD - supervisor T. Proslier
- Mathieu Lafarie – Thesis (3rd year) – Multipacting mitigation/ALD – supervisor T. Proslier/M. Belhaj (ONERA)
- Théo Dejob – Postdoc (1st year/2) – ALD/HIPIMS
- Fritz Moschtmann – CDD Engineer (1 st year/2) – Additive manufacturing
- Mathieu Benko, Contrat d'apprentissage – supervisor Y. Kalboussi
- **Thomas Proslier – HDR (June 2024) - Group leader**

ALD system for SRF cavity coating at CEA

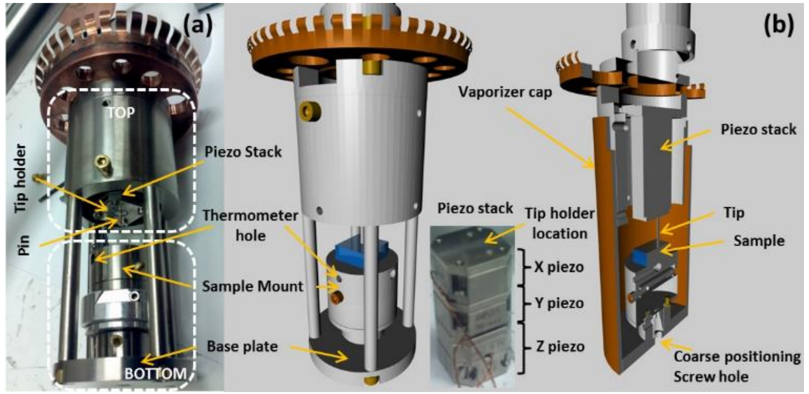
Front **Back**

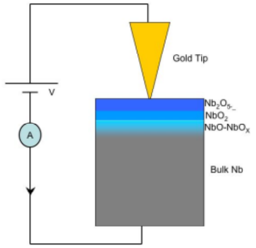


- **<u>High vacuum oven</u>:**
  - 650°C – 10^{-6} mbar / 900°C 1bar N_2
  - Volume retort: Φ = 49 cm, L= 110 cm (1.3, 0.7 GHz cavities)
- **<u>ALD system</u>:**
  - 9 precursor lines (2 gases, 2 liquids, 4 solids, 1 Ultra high temp).
  - RGA synthesis monitoring.
- **<u>MACHAFILM platform</u>**: EuroLabs.

- **<u>Interface and control:</u>**
  - Labview program of ALD system and Oven.
  - Automatic synthesis parameter control (overnight deposition) and monitoring.
- **<u>Cavities:</u>**
  - Can fit 0,65 to 1.3 GHz monocells
  - Multicells (2-3 cells 1.3 GHz)

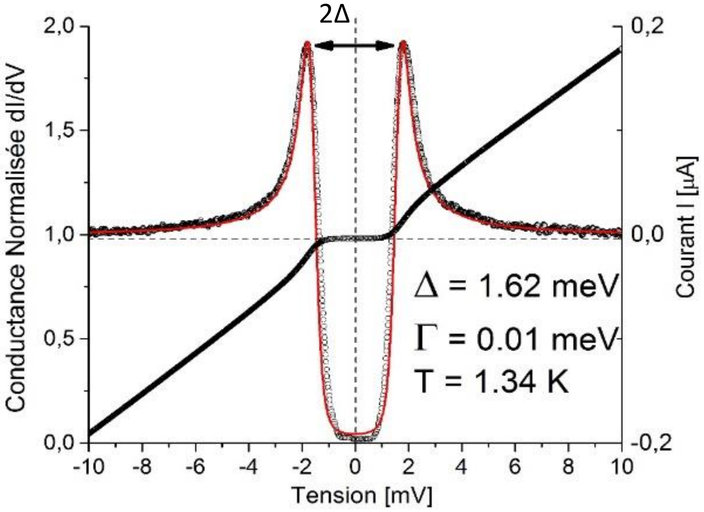
Tunneling spectroscopy system at CEA

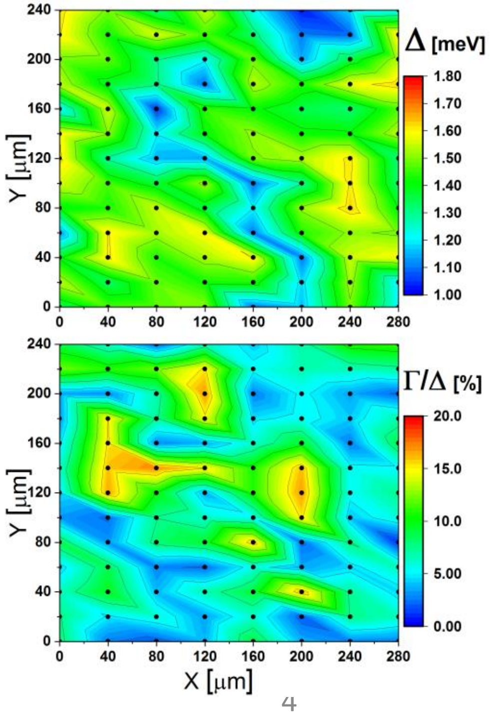




- Characterization of surface electronic properties:
- Measurement of SC fundamental properties: Δ, Tc, Hc
- ✓ Temperature from 1.4 to 300 K.
- ✓ **Cartography**: lateral size 10 µm– 1 mm.
- ✓ Materials under « real » conditions (oxide).
- ✓ Sample size about ~ 10x10 mm
- Research fields:
- ✓ *Accelerator: superconducting cavities*
- ✓ Qubits: dissipation phenomena
- ✓ superconducting thin films (ALD, HiPIMS, Sputtering, ECR, CVD…)
- Collaborations: CERN, INFN, STFC, JLab, ENS, Néel Institut, CEA
- MACHAFILM Platform: Eurolabs

Nb sample tunneling spectrum

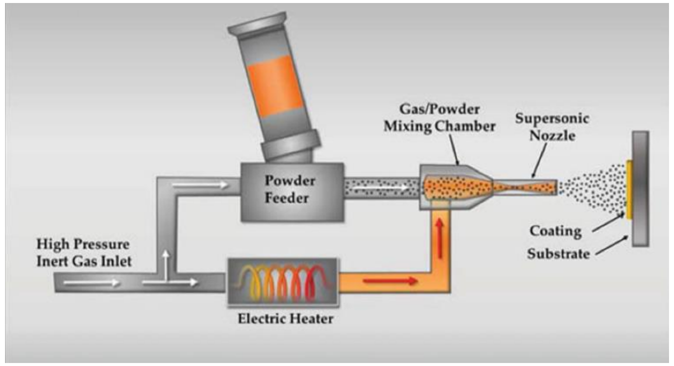




Additive manufacturing of copper SRF cavities

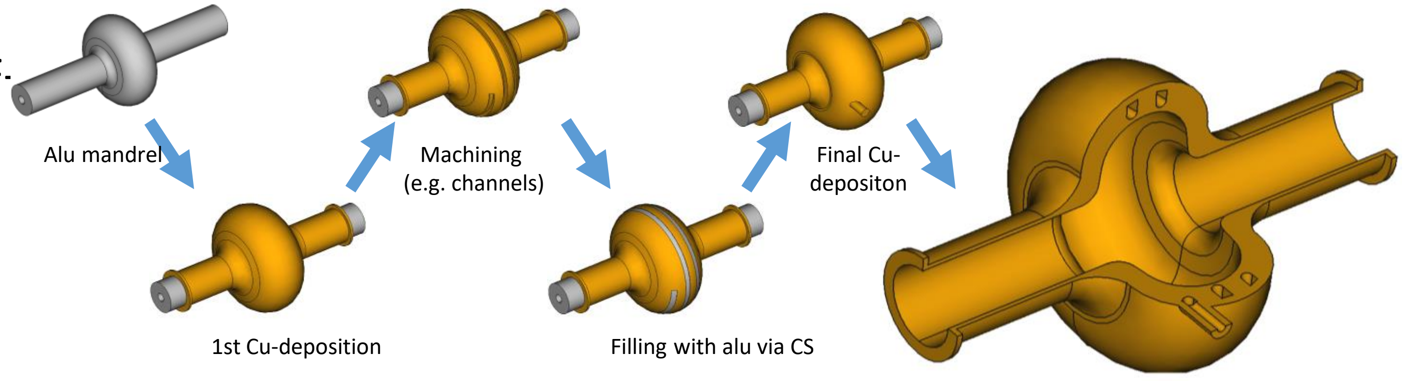
Additive Manufacturing of Copper SRF-Cavities

- **Exploration of Cold Spray Additive Manufacturing (CSAM) for copper cavities**
- Fast AM-method (deposition rate) ~ hrs for 1.3 GHz cavity.
- Easy upscaling with available fabrication equipment to larger cavities (0,8 to 0,4 GHz).
- Multiple metals (SS flanges, Cu cavity, Ti supports…) in one step process.
- Promising recent developments in quality of CSAM-copper (density > 99.5%, el. + th. Conductivity-RRR > 100…)
- **R&D project at CEA/Irfu/DACM**:
- Characterisation of Cu-deposits with respect to SRF-relevant properties
- Optimisation of process parameters
- Fabrication of a prototype cavity with integrated cooling circuit/volume
- Cryogenic tests of thermal transfer
- Collaborators: CEA/Irfu, UTBM/ICB



Source: *Practical Cold Spray*, Springer

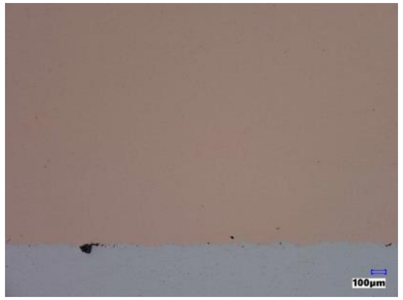
Cu-alloy combustion chamber with integral cooling channels made via CSAM
Source: https://impact-innovations.com/en/applications/



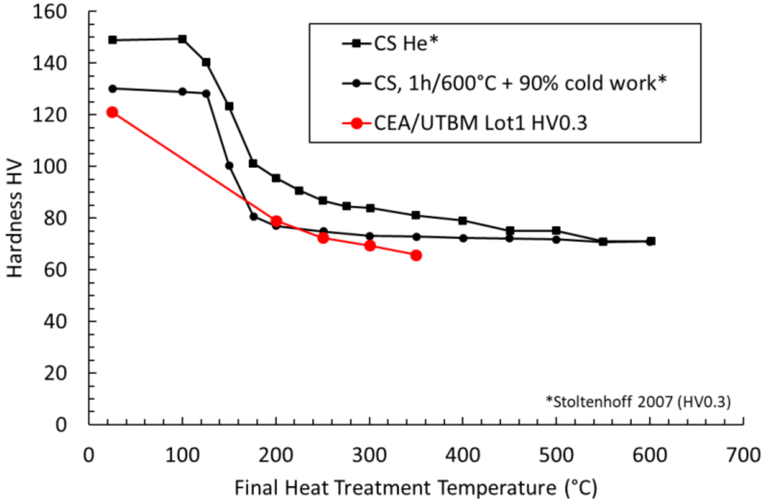
Final machining and chemical removal of aluminum

Additive Manufacturing of Copper SRF-Cavities

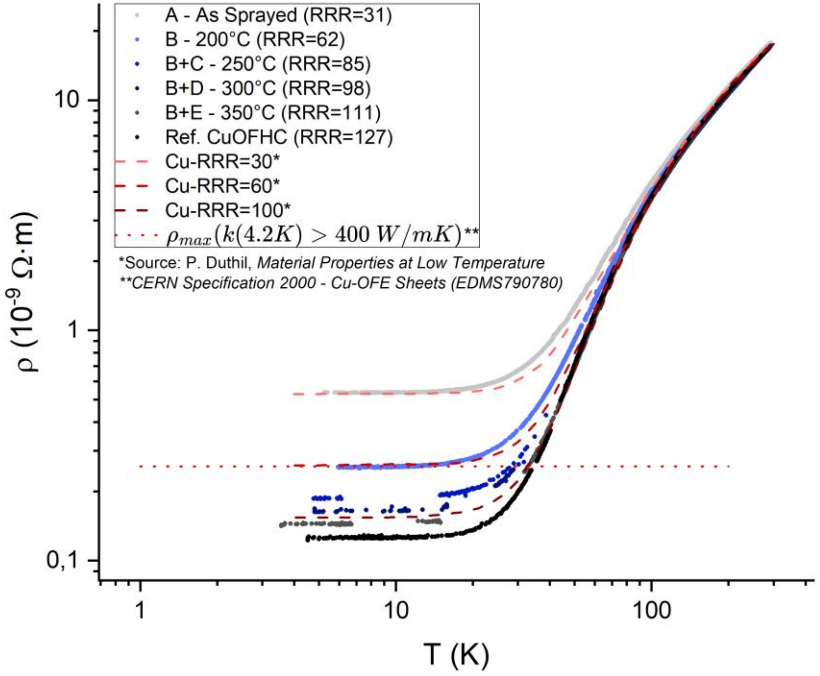
- **Exploration of Cold Spray Additive Manufacturing (CSAM) for copper cavities**

Top: Sample of pure Cu deposition (th. ≈10 mm) on aluminium
Right: Cross section of Cu on Alu-substrate

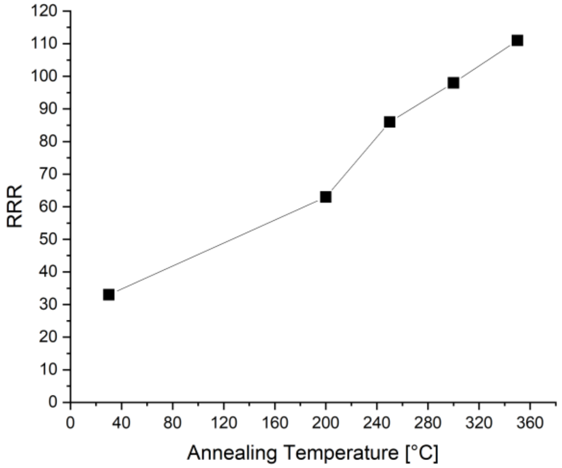


Hardness measurements of CSAM copper samples with different post-heat treatments compared to literature values with similar deposition parameters



Resisitivity curves (4-290 *K*) for CSAM copper samples with different post heat treatments and various references, RRR values in brackets

$$RRR = \frac{\sigma_{0\,K}}{\sigma_{273\,K}} = \frac{\rho_{273\,K}}{\rho_{0\,K}} \approx \frac{\rho_{273\,K}}{\rho_{4,2\,K}} \propto \frac{k_e}{T}$$



Preliminary results on state of the art CSAM copper:

- Dense deposition (>99.5% bulk), electrical conductivity >58 *MS* after heat treatments at 200°C or higher.
- Leak tight in UHV (< 10^{-11} mbar.l.s^{-1}).
- RRR values after post-heat treatments > 60, *k* values up to 600 *W/m.K* at 4.2 *K*.
- Slow decrease of hardness with increasing H.T. (compromise mech. properties-RRR)

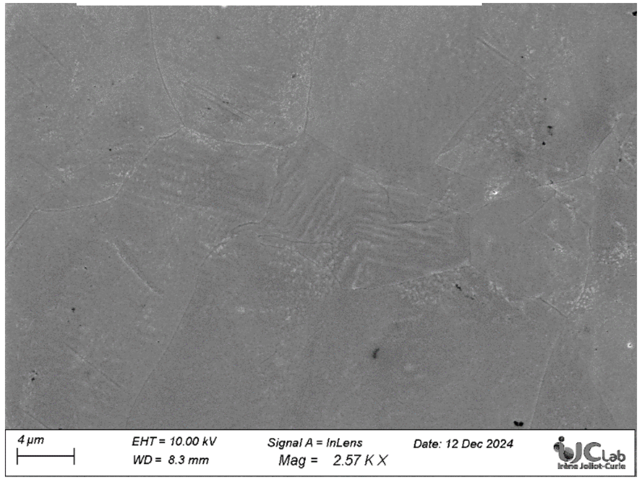
Diffusion barriers and adhesion layers on copper

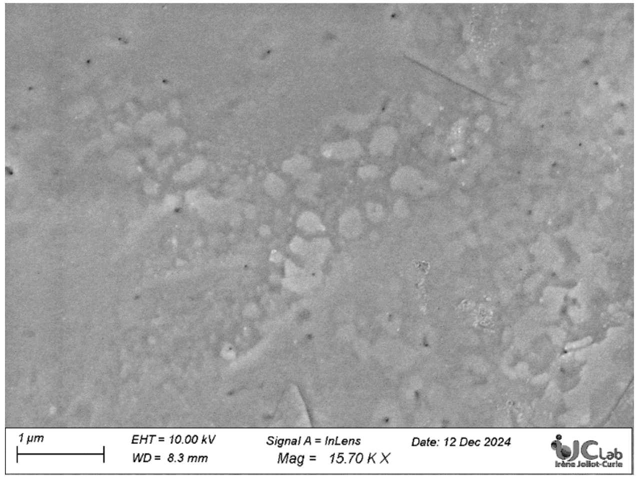
Context - purpose

- Insulating films on Cu: Mitigate thermo-currents effects in multi-metallic thin films cavities.
- Diffusion barriers in high temperature depositions (Nb_3Sn-Cu)
- Stabilize surface Cu chemistry.
- Adapt differences in thermal expansion coefficients (Nb_3Sn-Cu)/mecanical stress on layers.
- Projects: ISAS, IFAST, CERN

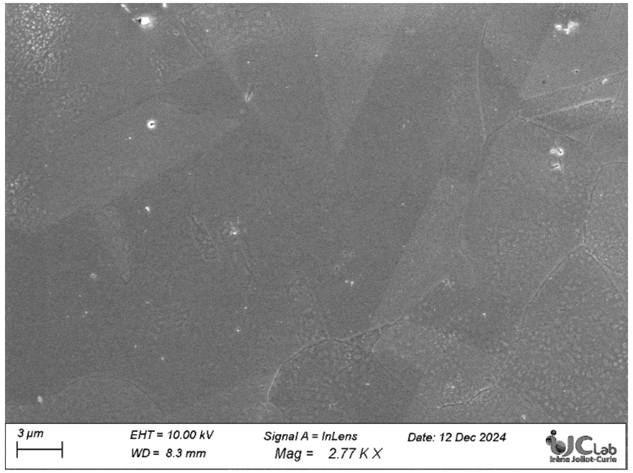
ALD Al_2O_3 on Cu

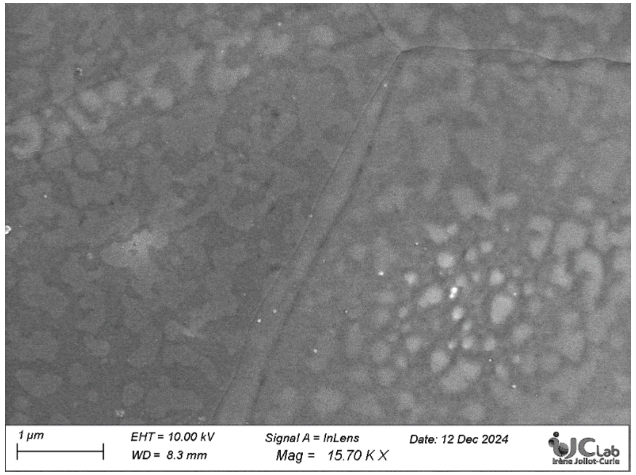
Before annealing



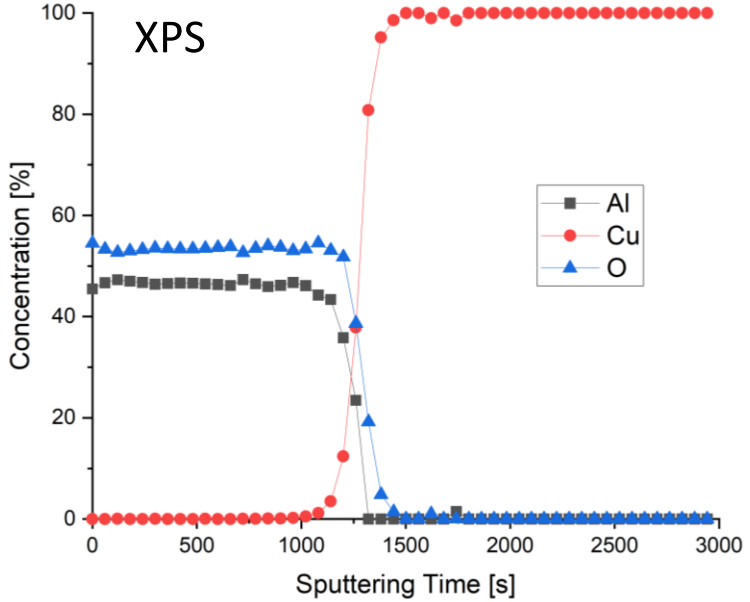


After annealing





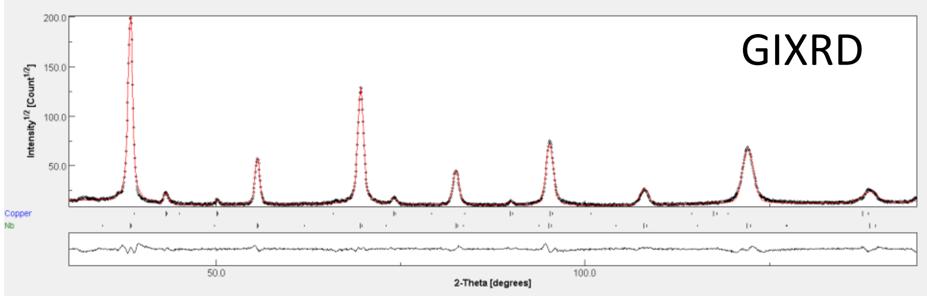
After annealing 700°C-2hrs + HPR



- No cracks, no holes
- Al_2O_3 layer (15 nm) stable after annealing up 700°C in high vacuum and HPR.
- No Cu diffusion through the Al_2O_3
- ***Amorphous*** before or after annealing. No structure observed by XRD or SEM.

Al_2O_3/Cu – Nb deposition

Baseline coating recipe:
1.2kW avg
100Hz, 200us HiPIMS pulses
-75V bias voltage
150C
$2.5.10^{-3}$ mbar Kr

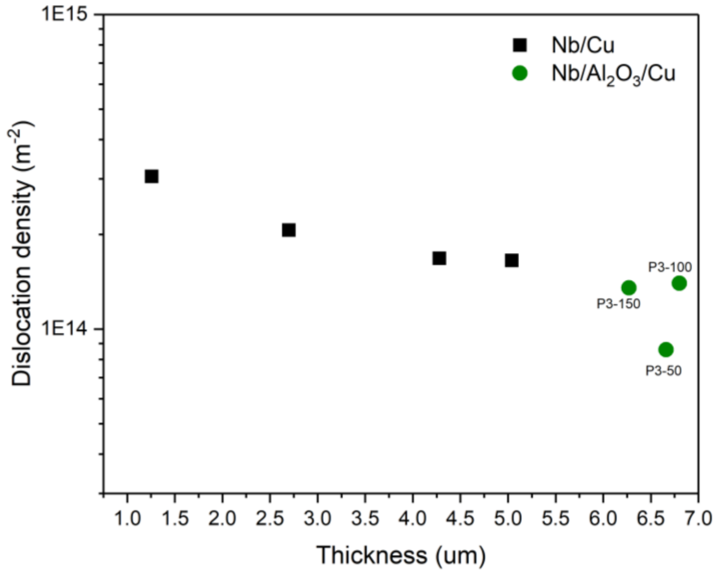


crystallite size, D
microstrain, ε
lattice parameter, a

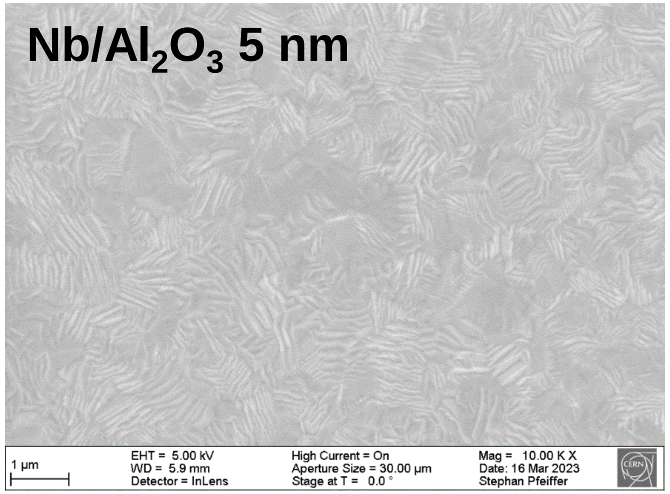
Dislocation density, ρ_D

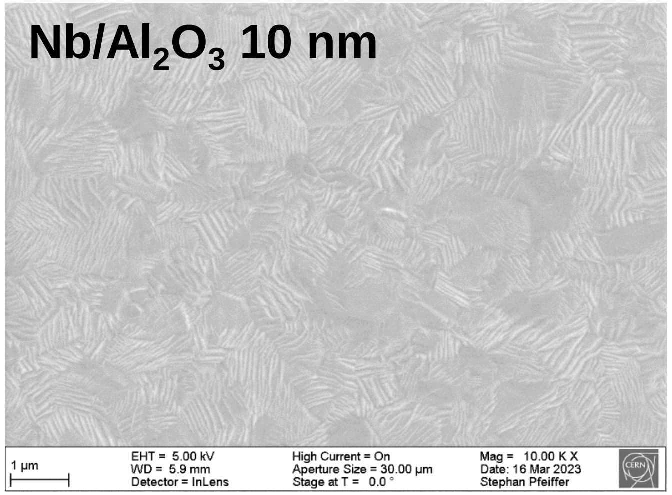
$$\rho_D = 2\sqrt{3}\,\frac{\langle \varepsilon^2 \rangle^{1/2}}{D \times b}$$

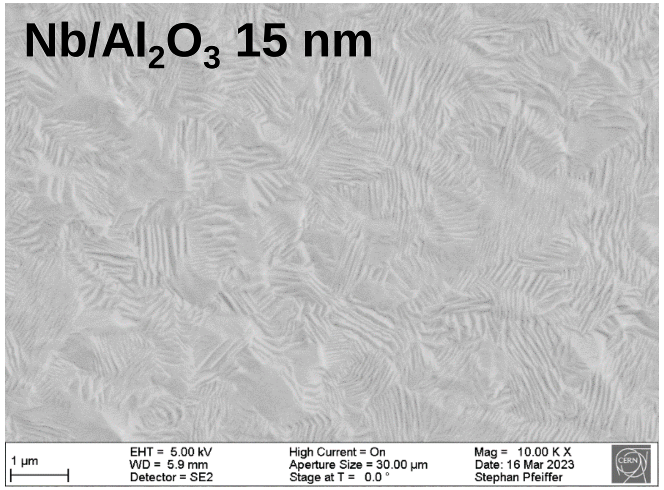
C. Pereira, S. Leith, G. Rosaz, S. Pfeiffer



- Nb/Cu and Nb/Al_2O_3/Cu do not show any difference from XRD.
- The lower dislocation density can be explained by the thicker layer.





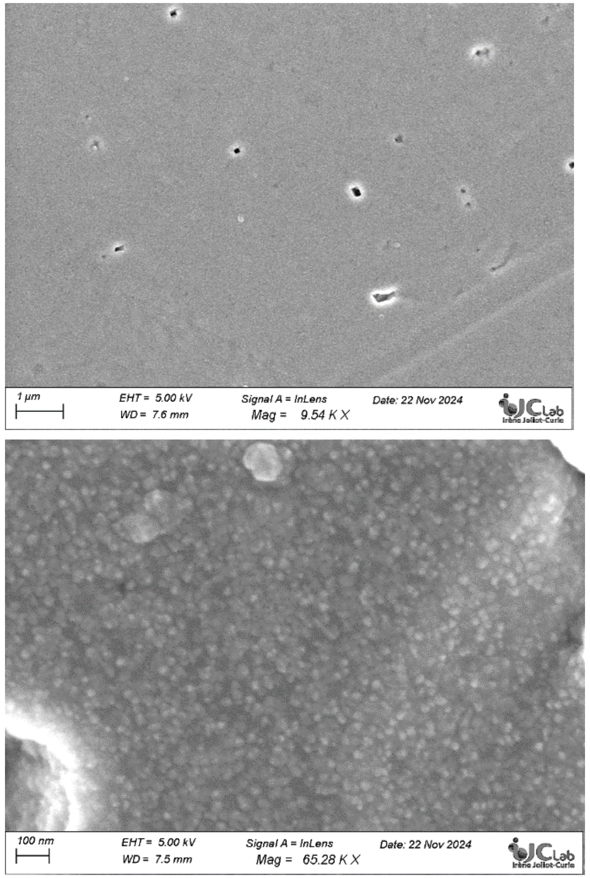


$Nb/Al_2O_3/Cu$ - Cavities

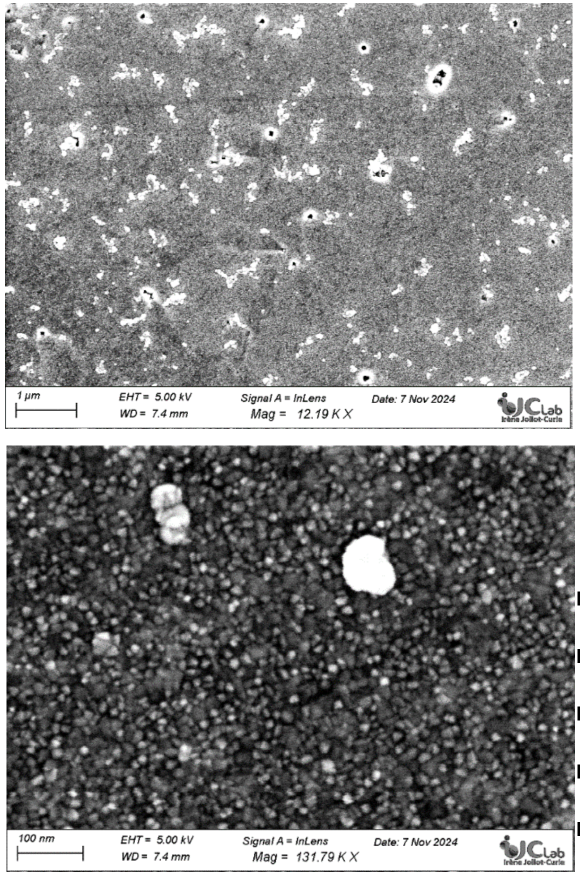
- No difference observed by XPS before or after HPR of 5, 10 or 15 nm thick Al_2O_3 layers on Cu.
- Stable up to at least 700°C and HPR.
- $Nb/Al_2O_3/Cu$ films have very similar structural and superconducting properties as Nb/Cu.
- ***Coating of two 1,3 GHz cavity EP from CERN with 15 nm of Al_2O_3. Send back to CERN for HPR and Nb deposition + HPR.***
- Open the way to coatings at higher deposition temperature (>150°C) for Nb_3Sn.

ZrO_2 (20 nm) /Cu – Crystalline vs Amorphous

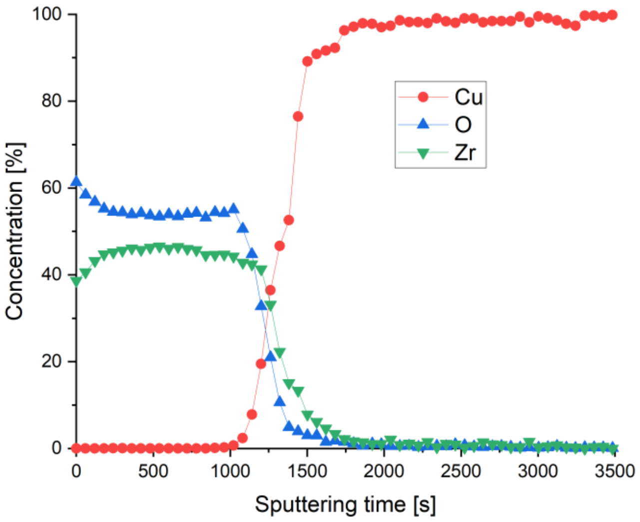
Before annealing



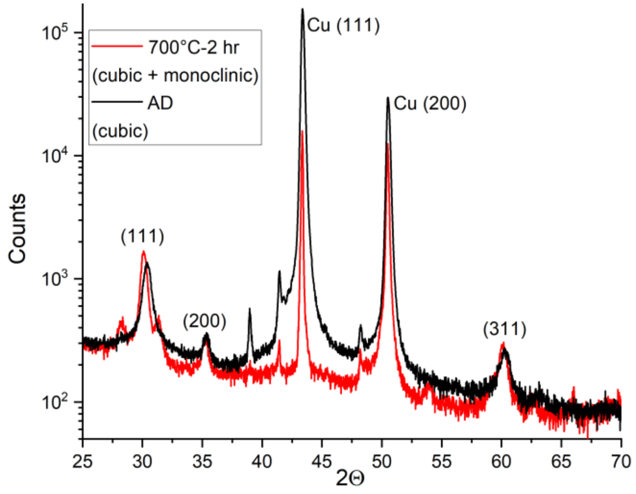
After annealing



XPS



XRD

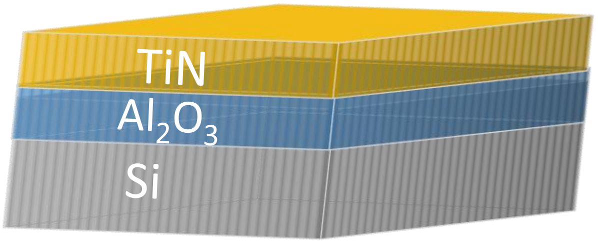


- ***Crystalline*** ZrO_2 grains 5-10 nm as grown or after annealing.
- No cracks for ZrO_2 films ≤ 20 nm on Cu.
- Cubic as deposited and mixture tetragonal after annealing.
- No copper diffusion after 700°C-2hrs.
- ZrO_2/Metallic Cu interface.
- Verified for 10 nm ZrO_2 films.

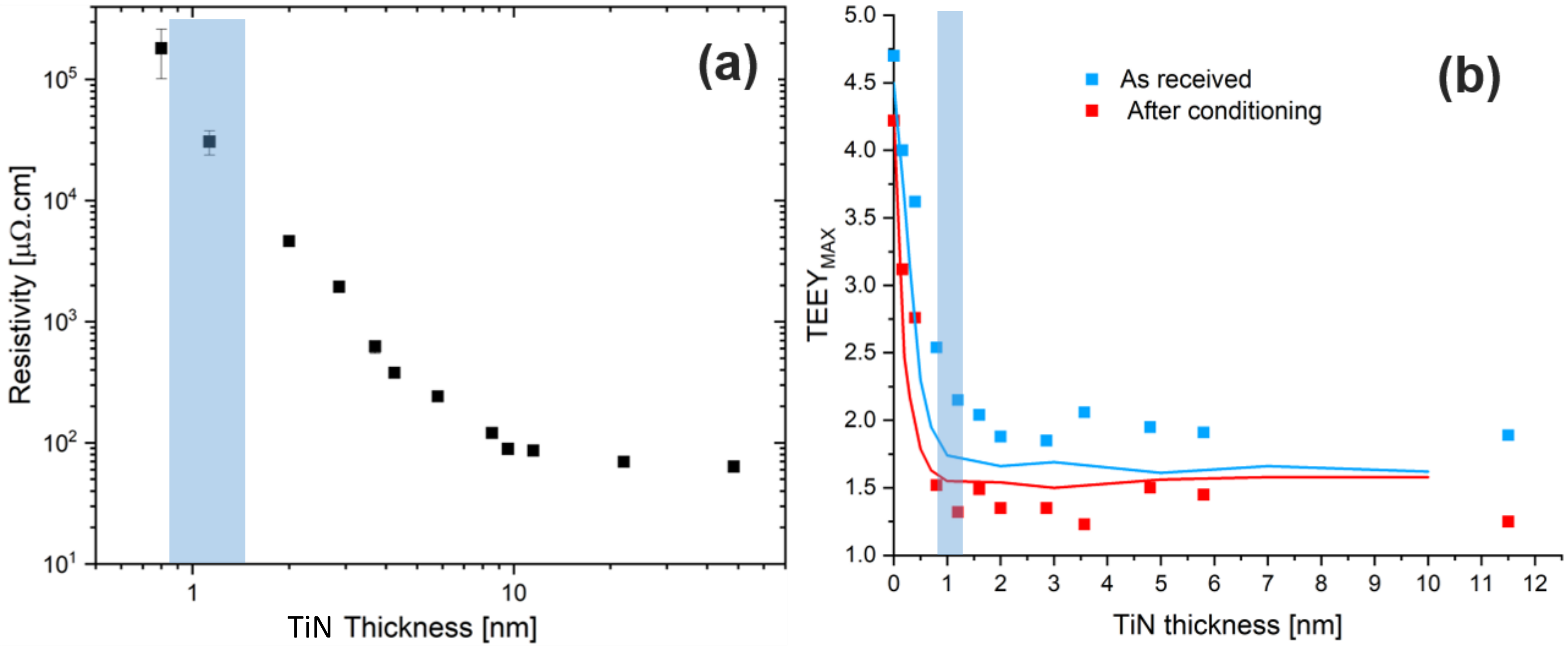
➢ Several interlayers identified and stable at High Temperature and HPR -> sent for Nb_3Sn deposition

Multipacting mitigation in SRF cavities

TEEY measurements on the TiN- Al_2O_3 samples



- TEEY (Total electron emission yield) is a critical surface parameter in multipacting.
- Electrical resistivity (ρ) is a critical parameter that affect SRF cavity performances.
- ALD -> atomic scale control of the thickness -> tune TEEY and ρ.



ONERA
THE FRENCH AEROSPACE LAB

➢ There is a window of 30 - 50 cycles where the TEEY is low and the TiN film resistance is high.

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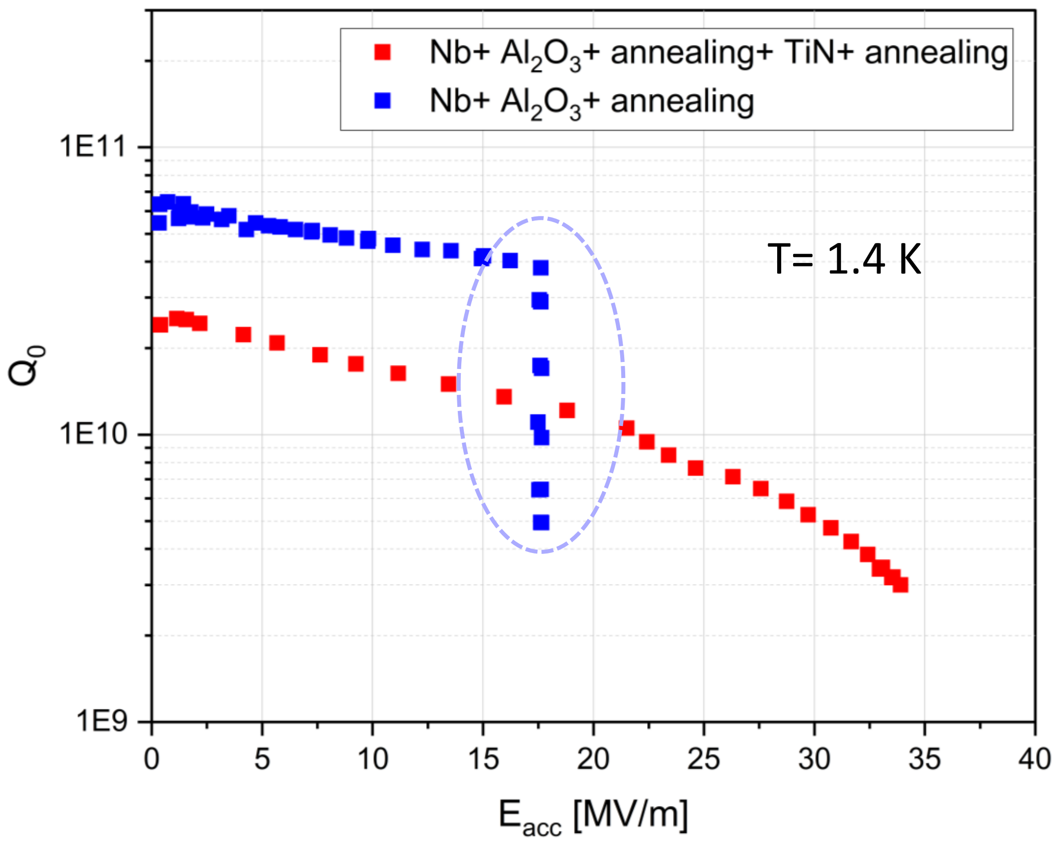
RF test of the TiN thin film on 1.3 GHz Niobium cavitiy

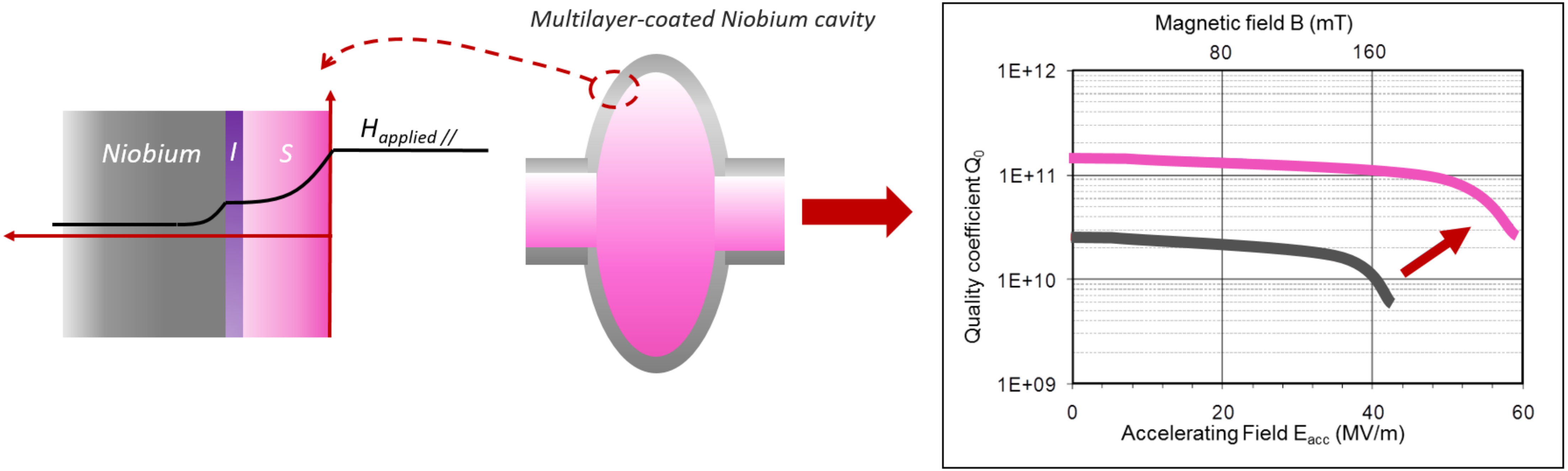


- The quality factor is slightly lower but the 1.5 nm thick TiN film is effective as a multipacting mitigation layer.
- Apply this approach to RF couplers, embarked systems in spatial applications (RF filters, solar panels…)

[*]Kalboussi, Y., Dadouch, S., Delatte, B., Miserque, F., Dragoe, D., Eozenou, F., ... & Proslier, T. (2024). Multipacting mitigation by atomic layer deposition: The case study of titanium nitride. *Journal of Applied Physics*, *136*(8). https://doi.org/10.1063/5.0221943

Superconducting multilayers to increase SRF cavities performances

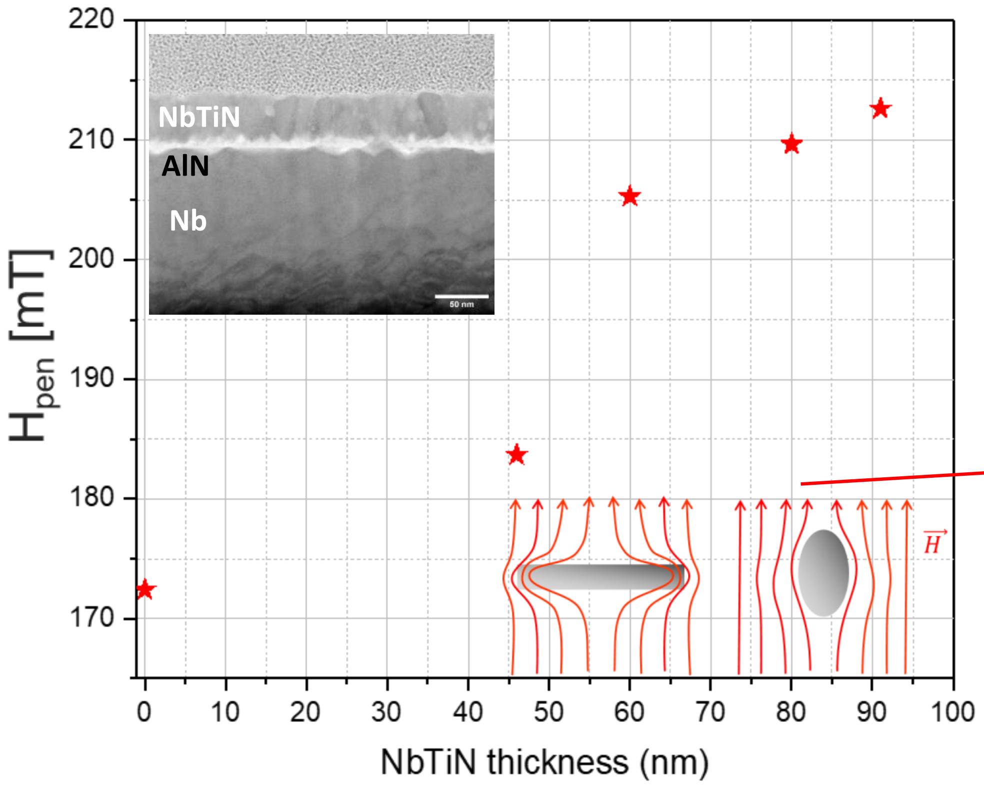
Multilayers

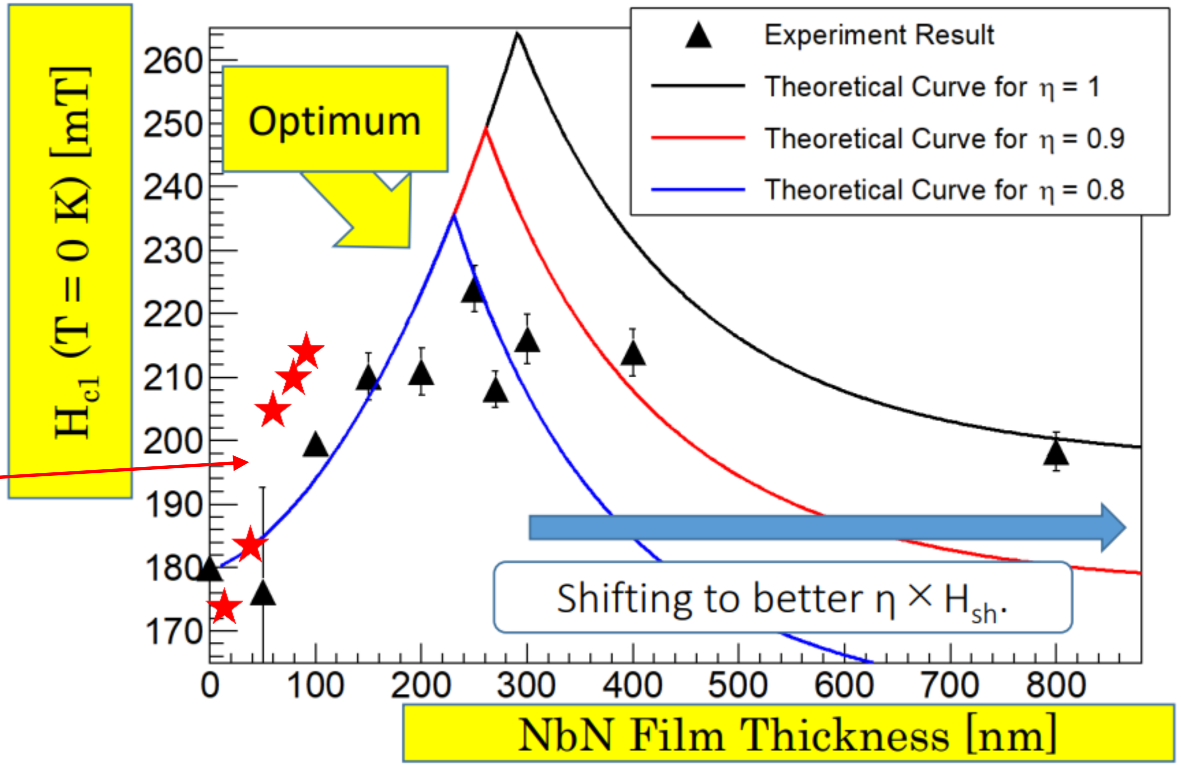


- A theoretical approach proposed by A. Gurevich (2006) to improve RF cavities through depositing a superconducting multilayer to screen the magnetic field.
- The thickness of the superconductor must be lower than its penetration depth.
- The superconducting layer must have higher T_c than Nb.

AlN-NbTiN multilayers

➢ The Niobium ellipsoid was coated and annealed with the optimized NbTiN-AlN bilayer recipe.

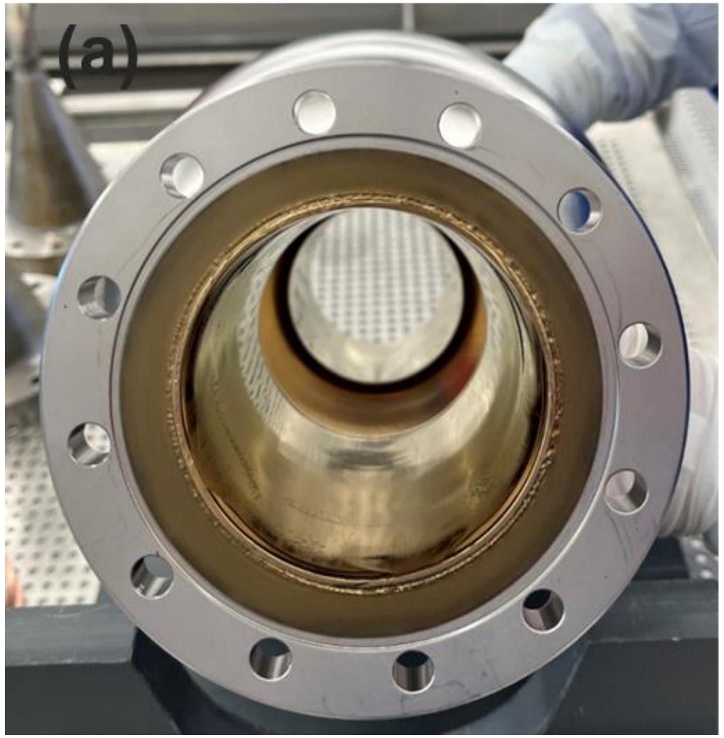




T. SAEKI, TFSRF21: Overview of thin-film studies at KEK and Kyoto University, (2021).

- Enhancement of first penetration field demonstrated.
- Thicker layer (~ 200 nm) to determine ξ and the predicted optimal thickness

NbTiN-AlN Multilayer on 1,3 GHz cavity



- Coating had a bright golden and uniform colour.
- The cavity was annealed @ 900°C.
- Vacuum degradation during the annealing step. ($P>10^{-5}$ mbar)

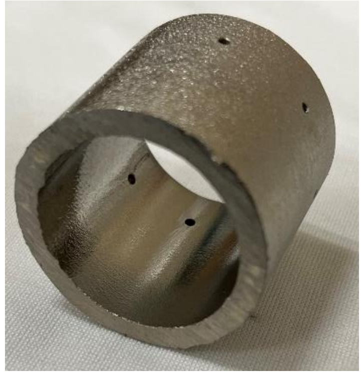
- Observed delamination in the beam tubes after annealing.

Delamination studies

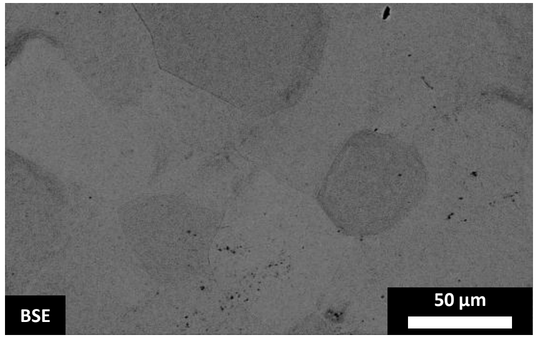
- Leak detected and fixed.
- Upscaling of the samples with tubes and curve plate

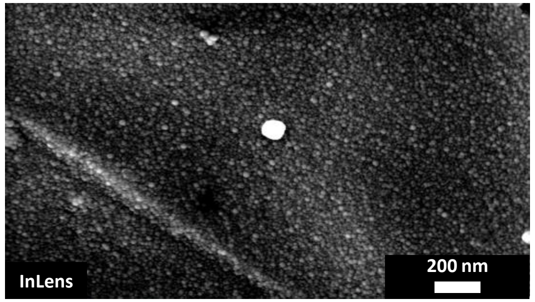
Height : 30 mm
Diameter : 37 mm

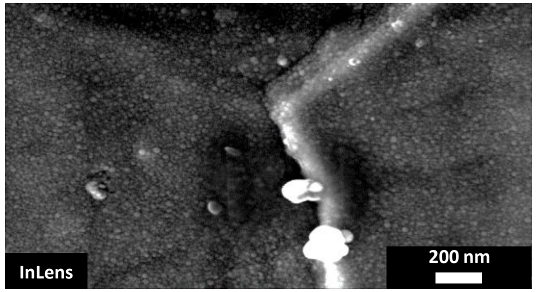
Annealed at 900°C



55 by 22 mm





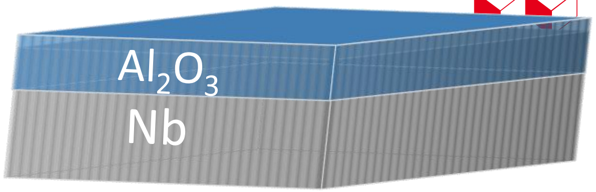


- Presence of film confirmed by XRD, EDS, MEB
- No delamination observed
- ➢ Test again on Nb cavities.

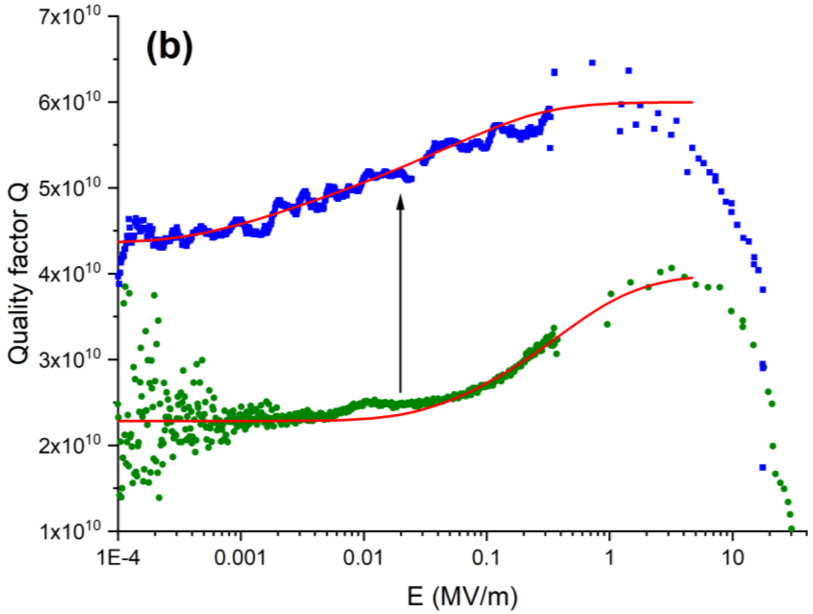
- Tc ~ 14.5 - 15 K (42 nm)
- New multilayer diffusion barrier.

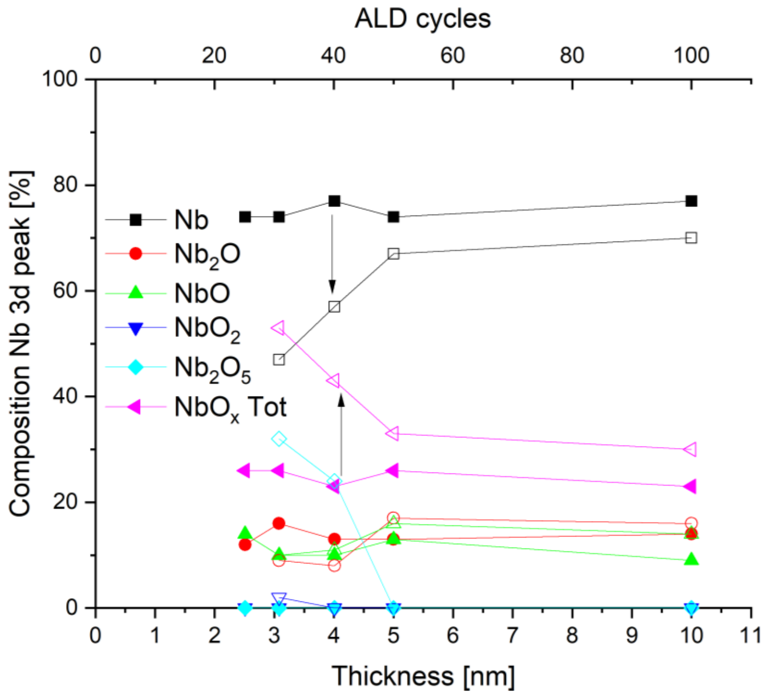
ALD on bulk Nb cavities: Control of surface for SRF cavities and Qubits

Al_2O_3 Thin films

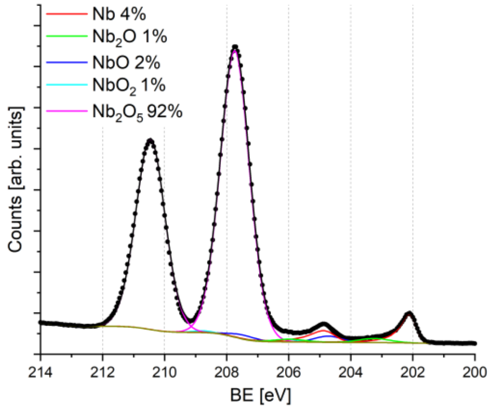


Al_2O_3-10nm- 650°C- 10hrs

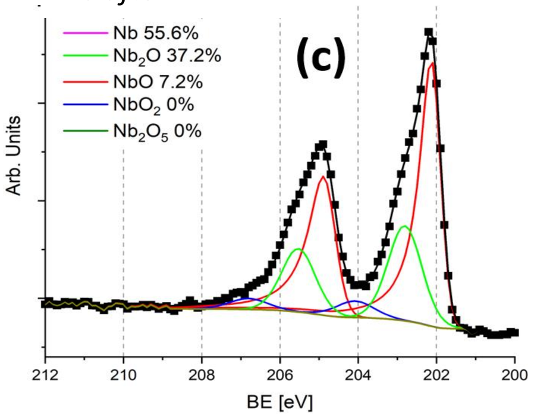




Nb after EP + HPR



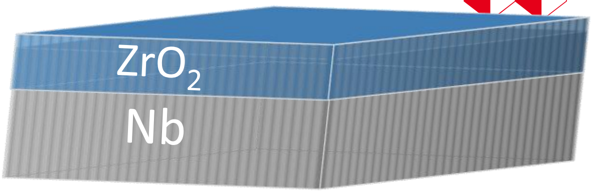
Nb after ALD + TT

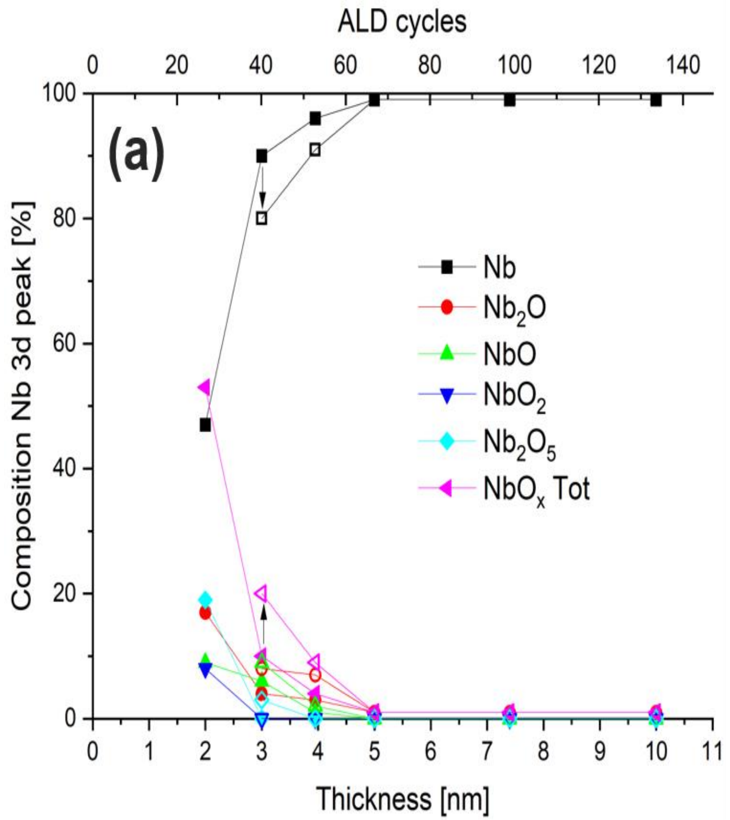


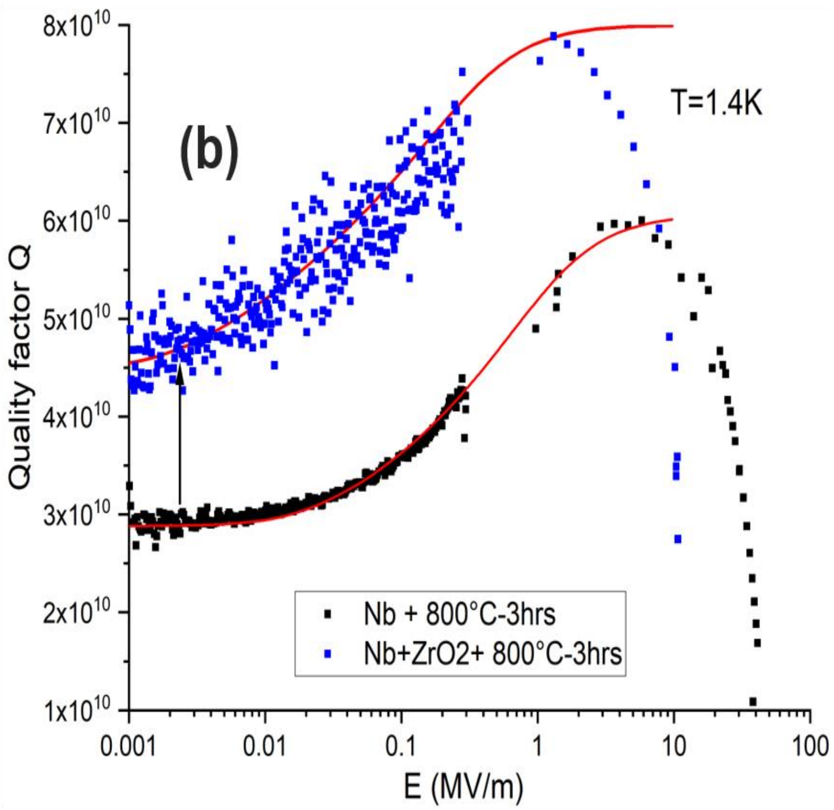
- The 10 nm Al_2O_3 film + annealing at 650°C for few hours significantly improves the quality factors of the Nb cavity in the low field regime.
- Optimization of the Al_2O_3 thickness -> Ok down to 2.5 nm.

[1] Y. Kalboussi et al, "Reducing two-level system dissipations in 3D superconducting niobium resonators by atomic layer deposition and high-temperature heat treatment," Applied Physics Letters, 2024, 124 (13), pp. 134001. ⟨10.1063/5.0202214⟩. ⟨hal-04470953⟩.
https://doi.org/10.1063/5.0202214

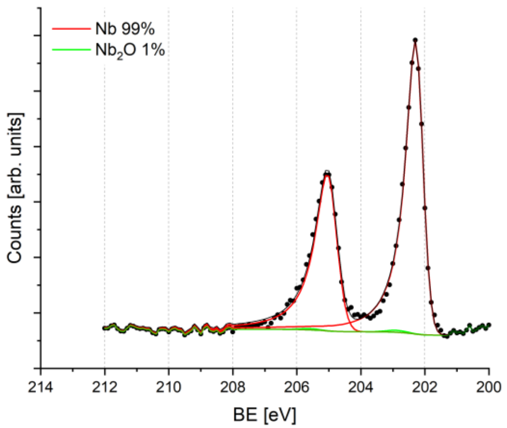
ZrO_2 thin films

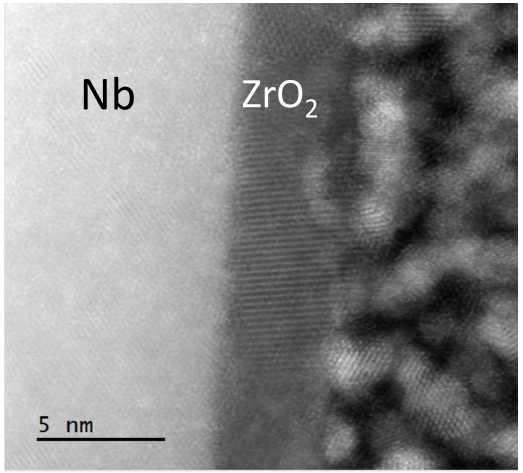






800°C – 3hrs / 5 nm ZrO_2

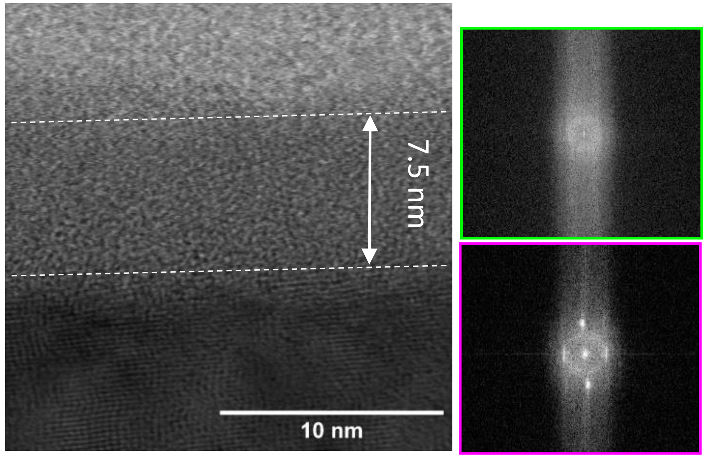


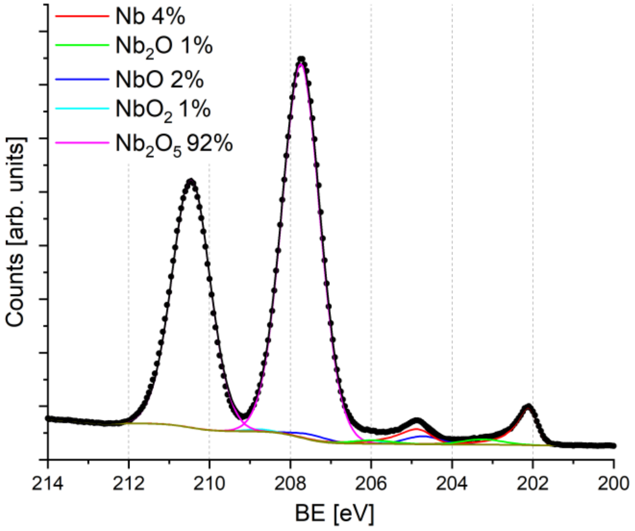


- ZrO_2 growth cubic phase
- Stable up to 900°C on Nb and sharp Nb metal/Oxide layer after air exposure and HPR.

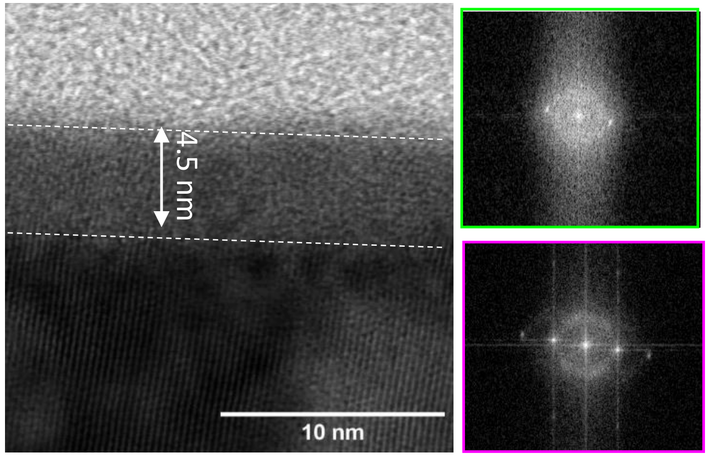
Changing NbO_x structure

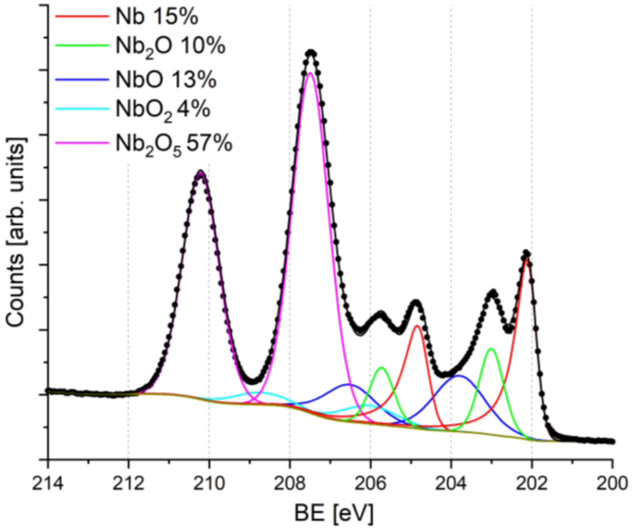
- Nb EP + HPR

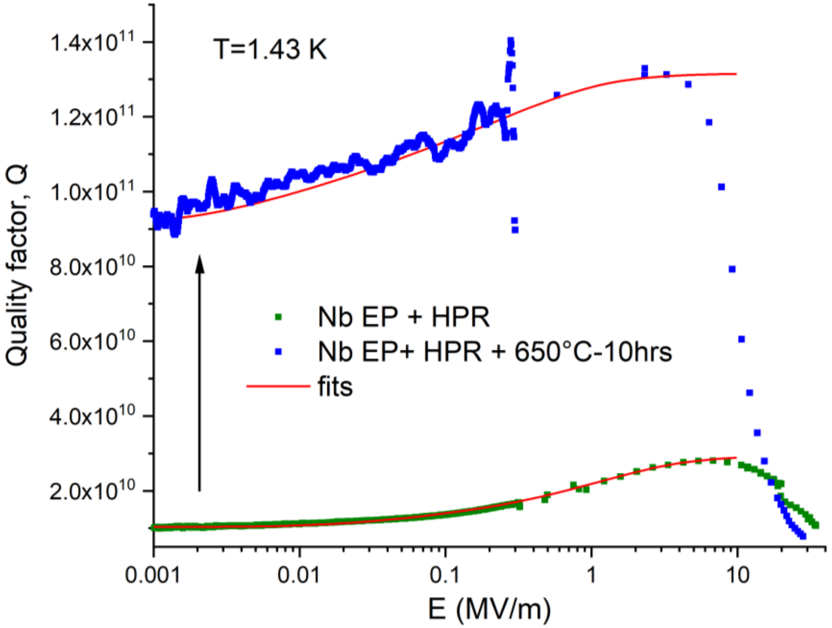




- Nb EP + HPR + 650°C-10hrs + HPR







- World record Q values at low fields (T ≈ 15 s) after air exposure and high pressure water rinsing (!)
- σ_{TLS} and $\tan(\delta_{TLS})$ decreased by a factor 10.
- Change in chemical / structure of the native NbO_X with indication of NbO_X (?) and Nb_2O_5 crystals.
- Under investigation.

[2] Kalboussi, Y., Curci, I., Miserque, F., Troadec, D., Brun, N., Walls, M., ... & Proslier, T. (2025). Crystallinity in niobium oxides: A pathway to mitigate two-level-system defects in niobium three-dimensional resonators for quantum applications. *Physical Review Applied*, *23*(4), 044023. https://doi.org/10.1103/PhysRevApplied.23.044023

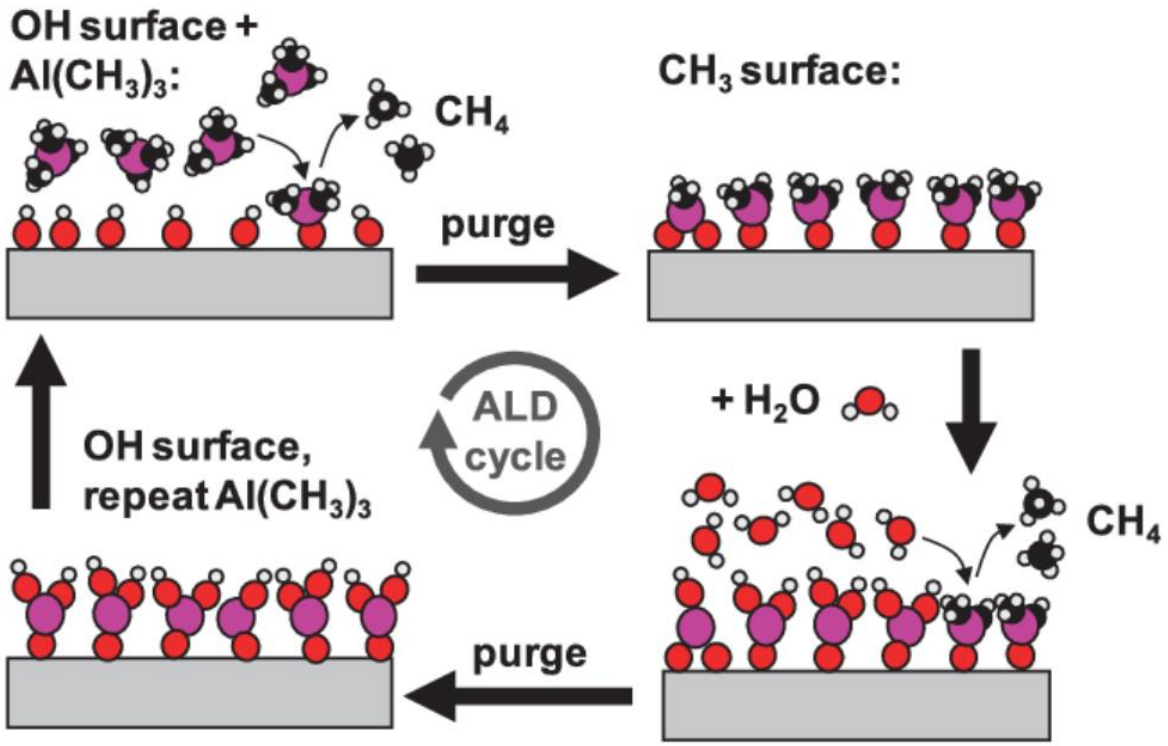
Future directions

- **Scarcity of ressources: higher operation temperature / reduce He volume/ reduce raw materials.**
- Development of new facilities owing to ANR Equipex+ PACIFICS funding.
  - New coating capability: HIPIMS for higher T_C material deposition – 1.3 to 0.8 GHz. **4.2 K operation.**
  - New cryostat with cryocooler cool down of SRF cavities – 6 to 1.3 GHz. **Cryocooled closed loop He.**
  - New RF testing facility (metrology, multi single cell testing, higher frequency range) - 6 to 0.65 GHz.
  - Upgrade and new chemical facility (Cu and Al alloys electropolishing, vertical and horizontal bench) – 6 to 0.4 GHz. **Beyond bulk Nb.**
- Developement of ceramic (Nb_3Sn…) targets for HiPIMS in cavities.
- Additive manufacturing to optimize He ressources and cooling process.
  - Cold Spray as scalable technique with multi metal manufacturing capability (Cu-SS/Cu-Ti). **Less raw materials.**
  - in-wall integrated LHe cooling channels. **Reduce by a factor 10 the volume of LHe.**
- Qubits: mitigate the TLS defects at the surface of superconductors -> from 3D to 2D.
- Characterization facilities:
  - Tunneling spectroscopy (in house). XPS, SEM, XRD, TEM…(sub contracts). **Predict RF performances from coupons.**
- Other applications: Multilayers, diffusion barriers / adhesion layer on Cu, for detectors (ANR clearmind), catalysis dilute Uranium filters (DES), Multipacting (ICONE)…

Spare

Atomic layer deposition – ALD

- ALD is a chemical gas phase deposition technique based on **sequential**, **self-saturating** gas-surface reactions.



Applications:
- Microelectronics
- Photovoltaics
- Nano-patterning for biomedical applications (nano-particles, nanotubes)

Advantages:
- Precise thickness control up to sub-nanometer scale
- Excellent conformality.
- Large choice of possible materials (oxides, nitrides and some metals …)

Limitations:
- Low growth rates (~ 0.1 nm per minute)
- New alloys require new chemistries (new precursors)