Surface engineering by ALD and heat treatment for 3D Niobium resonators for applications in superconducting qubits

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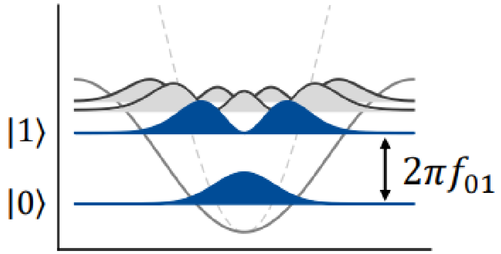
Context: Superconducting Qubits

Strongly anharmonic oscillator (qubit)

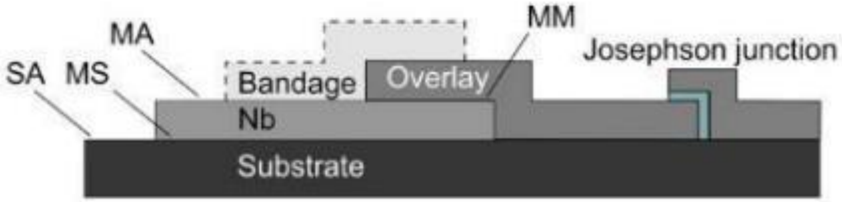


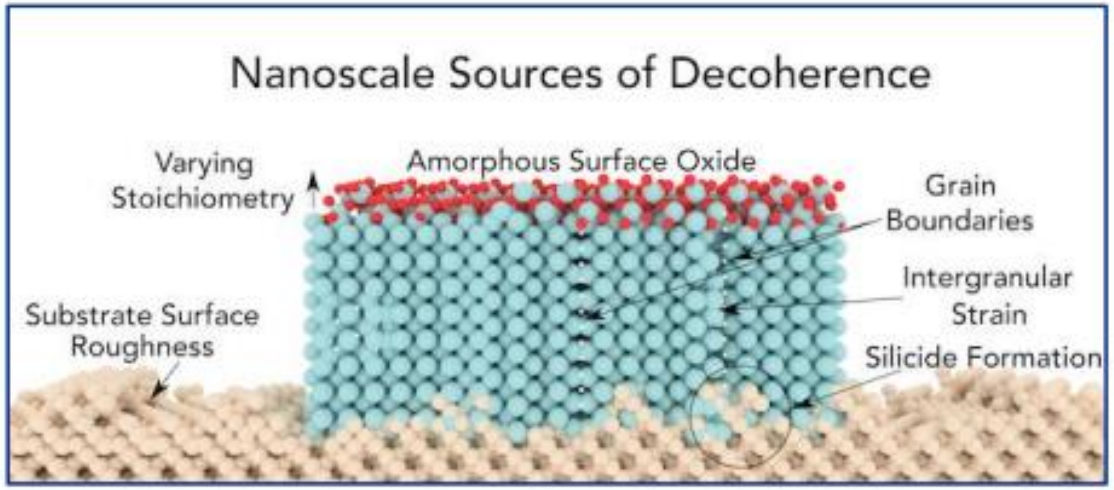
$$|\boldsymbol{\psi}\rangle = \cos(\theta)\,|\mathbf{0}\rangle + e^{i\varphi}\sin(\theta)|\mathbf{1}\rangle$$

- 2D superconducting qubits suffer from limited coherence times (at best $T_1 \sim 200\mu s$.)

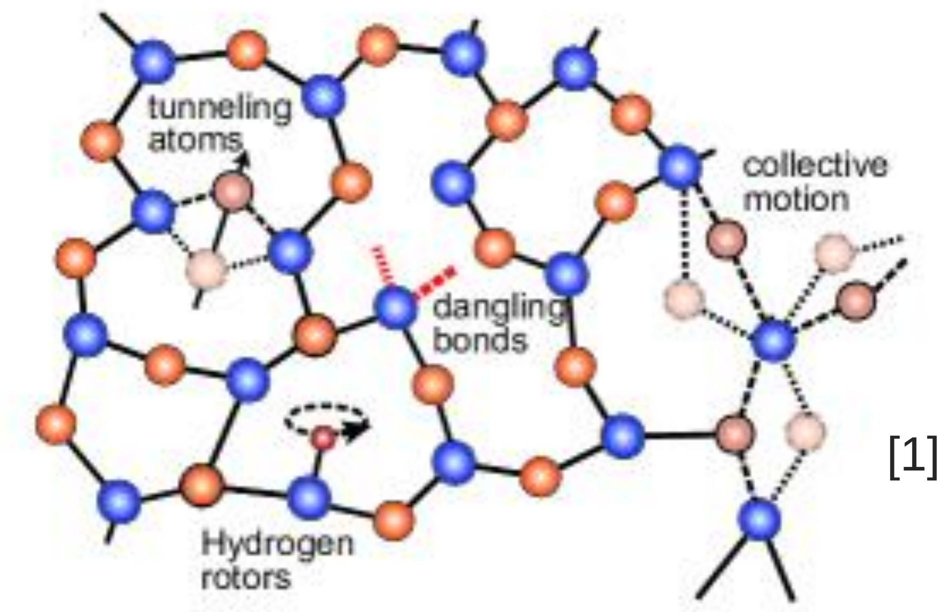
- Defects in the oxides layers are the major source of decoherence.

MS: Metal-Substrate
SA: Substrate-Air
MA: Metal-Air
MM: Metal-Metal





Two-level system defects in amorphous materials



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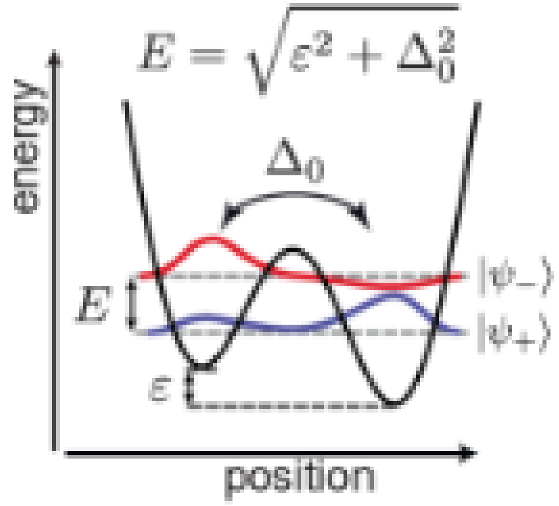
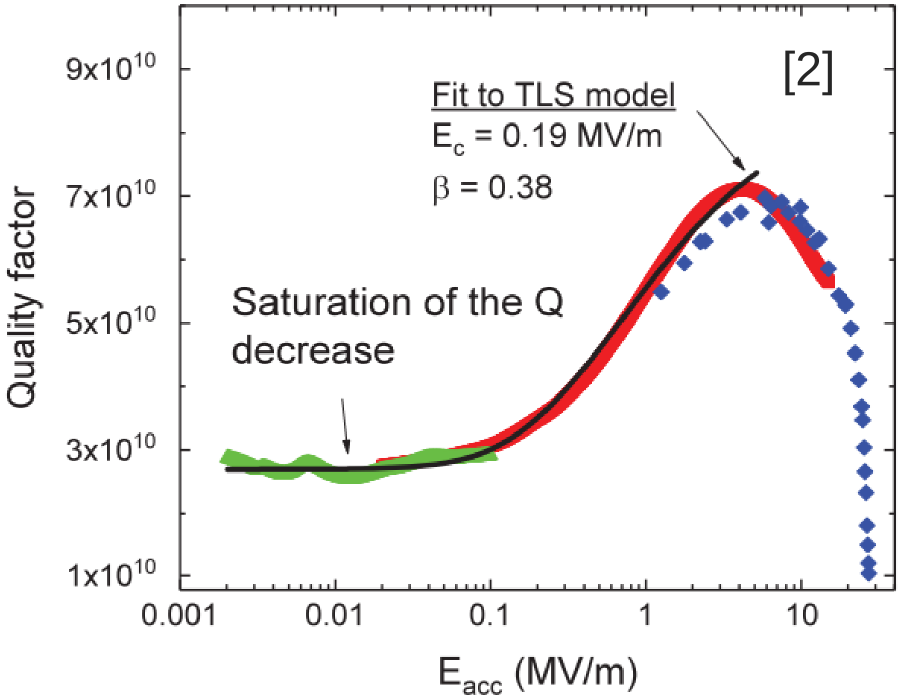


- Their microscopic nature is still ellusive:
- It can be tunneling atoms, tunneling electrons or spins and magnetic impurities.
- The TLS can exist in one of two energetically similar configurations
- At High temperature and high RF intensities, these defects are saturated.
- At low temperature, TLS couples to the electric field present in the qubit and causes decoherence.

SRF cavities are excellent tools to advance quantum computing

1. As a tool to study materials : SRF cavities exhibit the same dielectric losses at low fields.
2. As multi-levels quantum bits : Tesla cavities are being integrated in quantum processing units.



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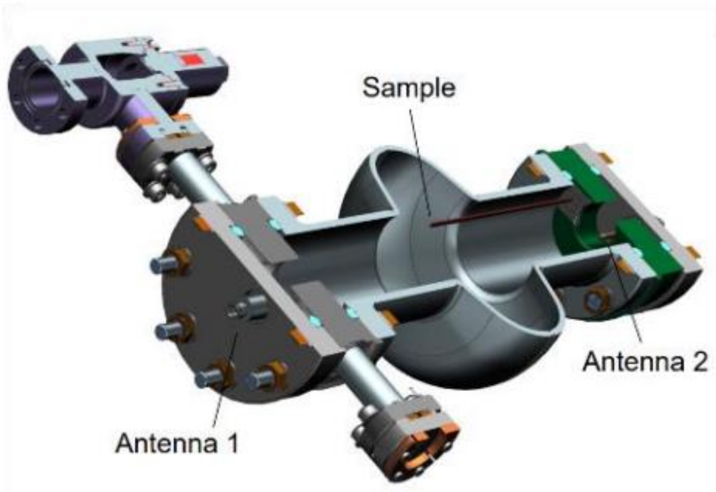

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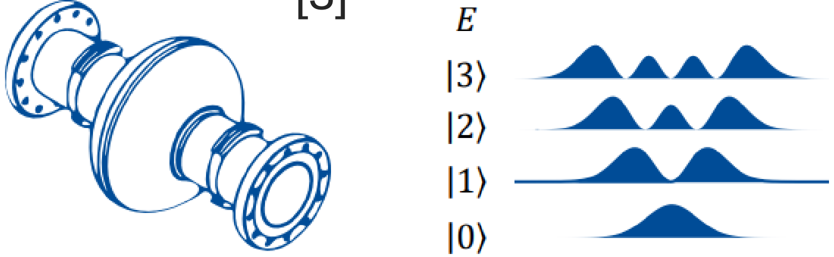
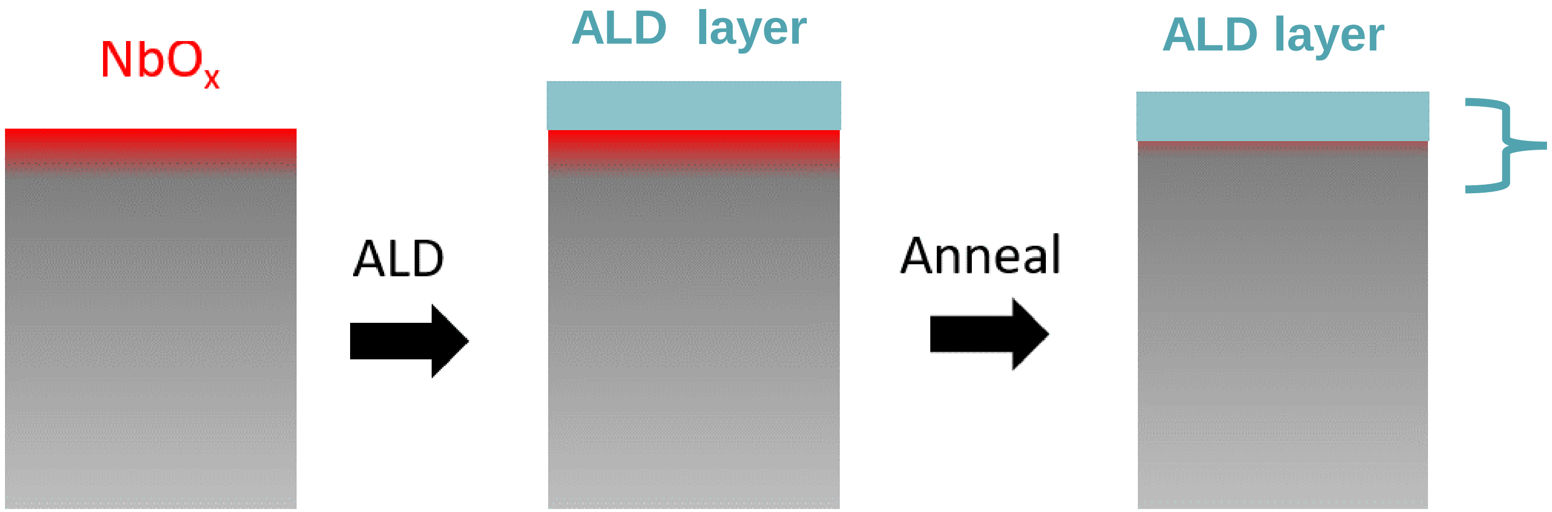


$$|\psi\rangle = c_1|\mathbf{0}\rangle + c_2|\mathbf{1}\rangle + \cdots + c_n|\boldsymbol{n}\rangle$$

The passivation approach at CEA:

To replace niobium native oxides with ALD-deposited protective layer [1]

1) Deposit ~few nm oxide layer by ALD (Al_2O_3 ,Ta_2O_5, ZrO_2…) onto Niobium.
2) Perfom a subsequent thermal treatement to dissolve niobium native oxide underneath (vacuum levels 10^{-6} mbar)



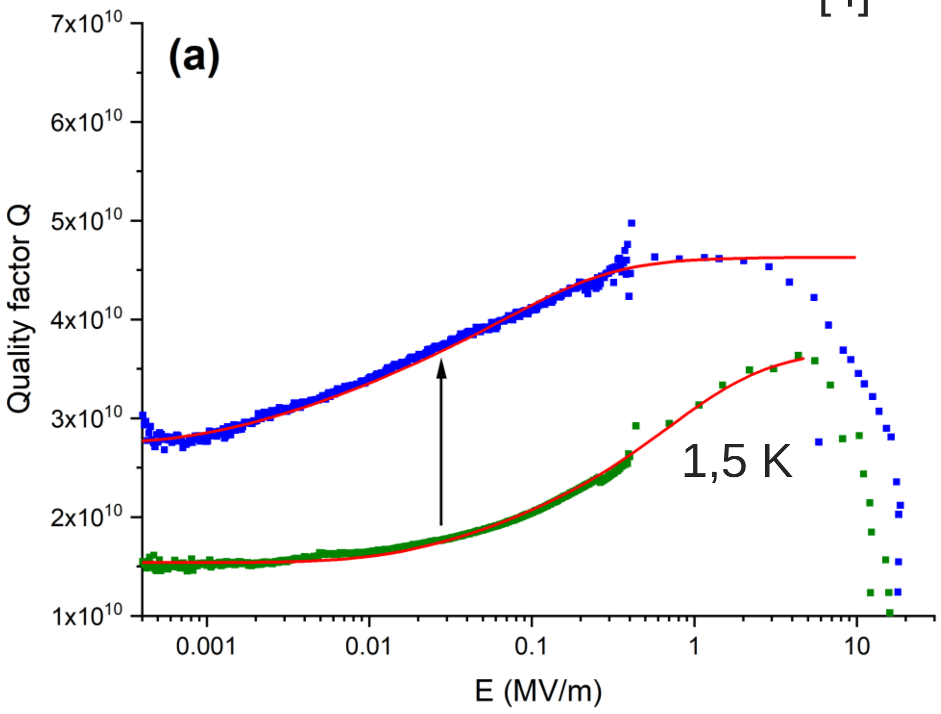
The oxide layer must be **thermally stable** and **have low dielectric** losses.

First results at CEA :

First experiment

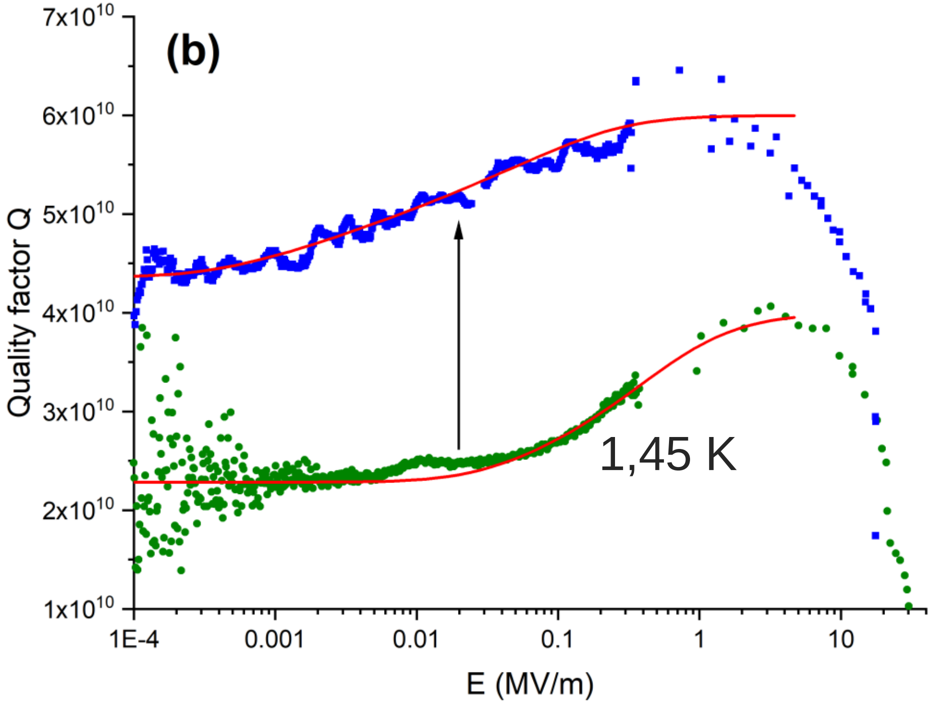
Al_2O_3-10nm- 650°C- 4hrs

[4]



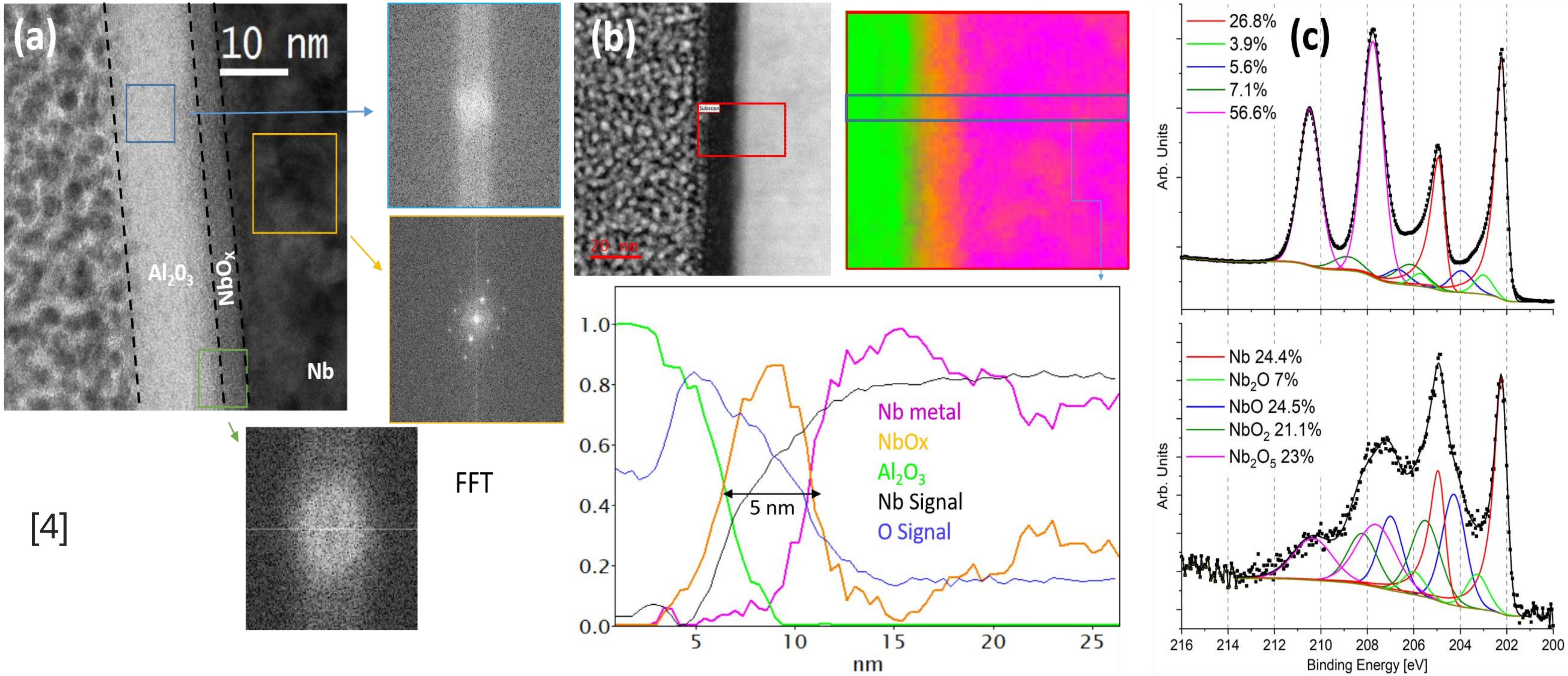
Second experiment

Al_2O_3-10nm- 650°C- 10hrs



- 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.

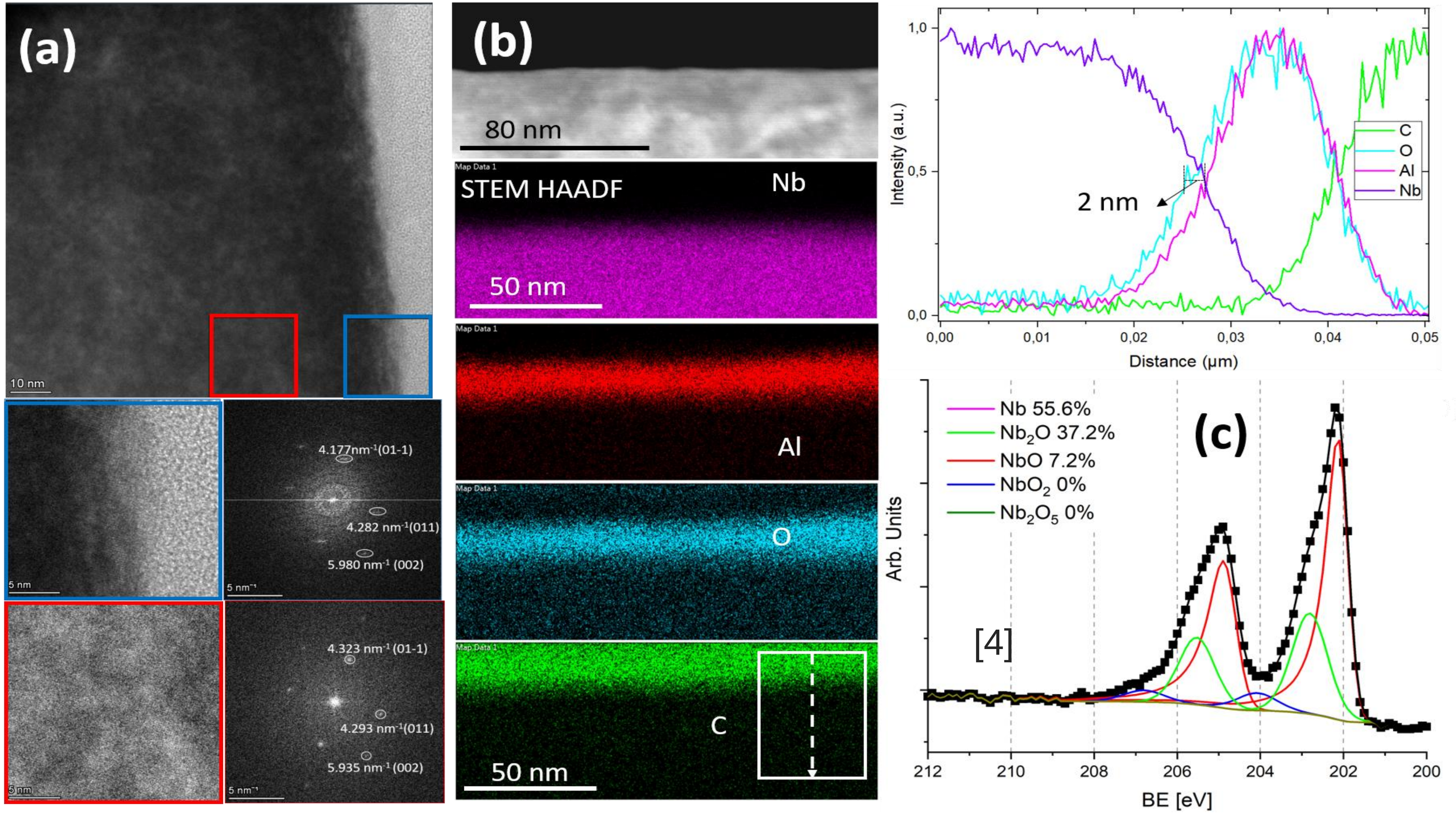
Before Annealing :



[4]

- Before annealing, we have 5 nm of Nb oxide between the Al_2O_3 and the Nb metal.

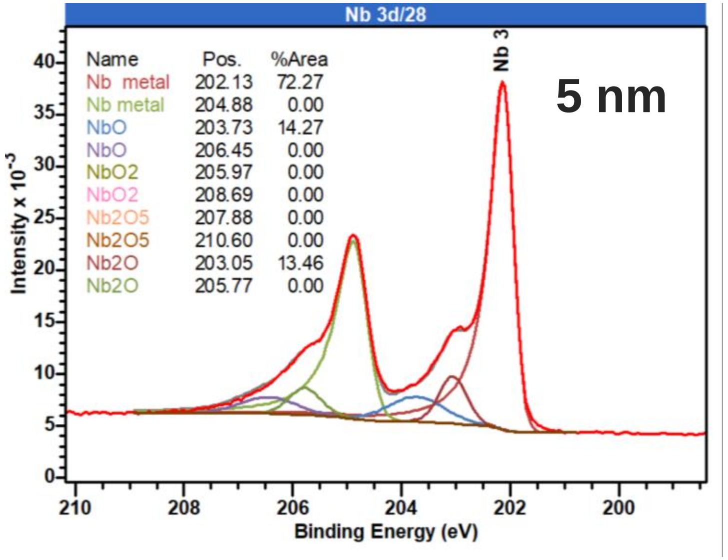
After Annealing



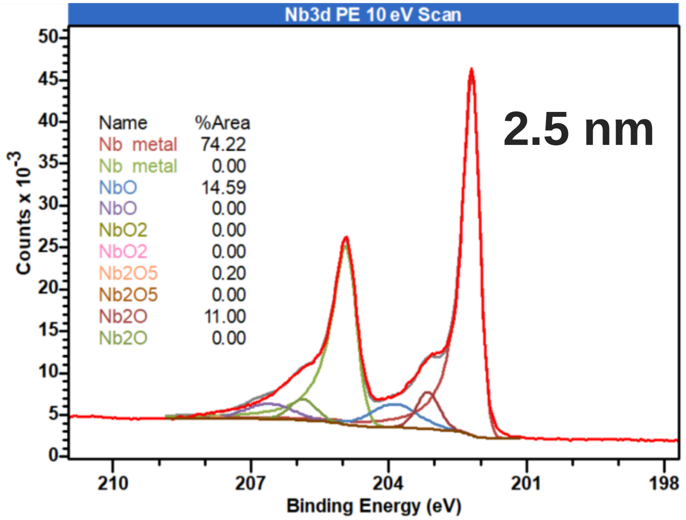
- After annealing, we have 2 nm of Nb oxide, mainly NbO and Nb_2O. No Nb_2O_5.
- The interface is crystalline with the bulk Nb.

Al_2O_3 hosts TLS too …

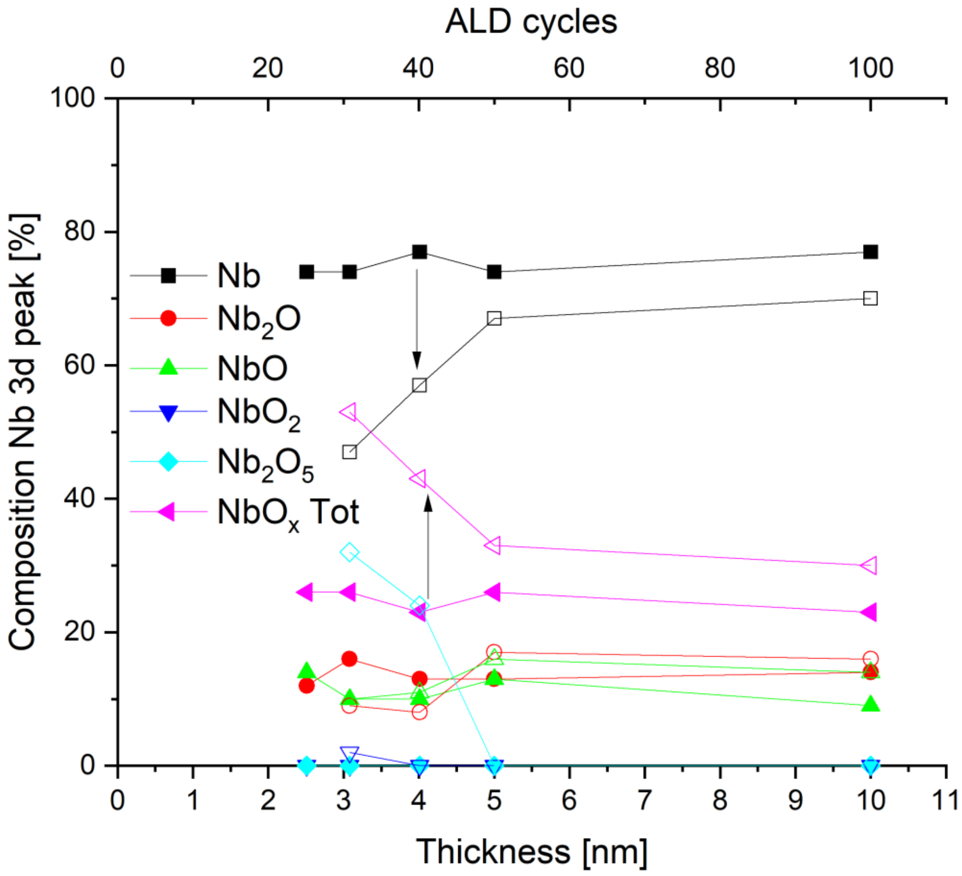
- We tested different thickness of Al_2O_3 + Annealing @650°C+10 hours



Name	Pos.	%Area
Nb metal	202.13	72.27
Nb metal	204.88	0.00
NbO	203.73	14.27
NbO	206.45	0.00
NbO2	205.97	0.00
NbO2	208.69	0.00
Nb2O5	207.88	0.00
Nb2O5	210.60	0.00
Nb2O	203.05	13.46
Nb2O	205.77	0.00



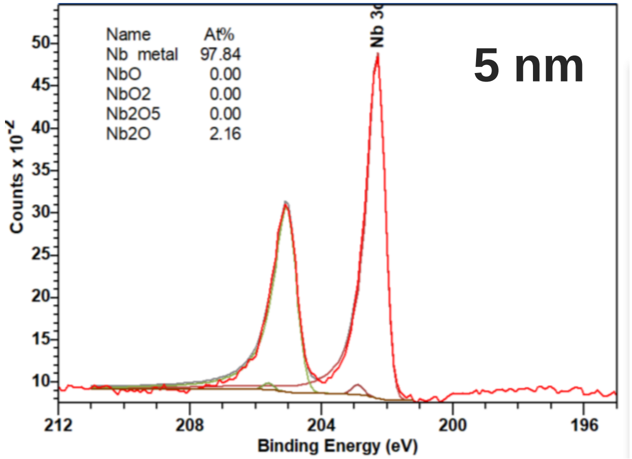
Name	%Area
Nb metal	74.22
Nb metal	0.00
NbO	14.59
NbO	0.00
NbO2	0.00
NbO2	0.00
Nb2O5	0.20
Nb2O5	0.00
Nb2O	11.00
Nb2O	0.00

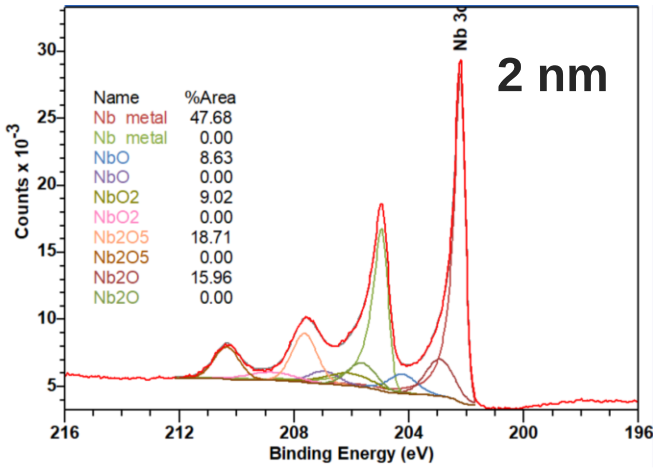


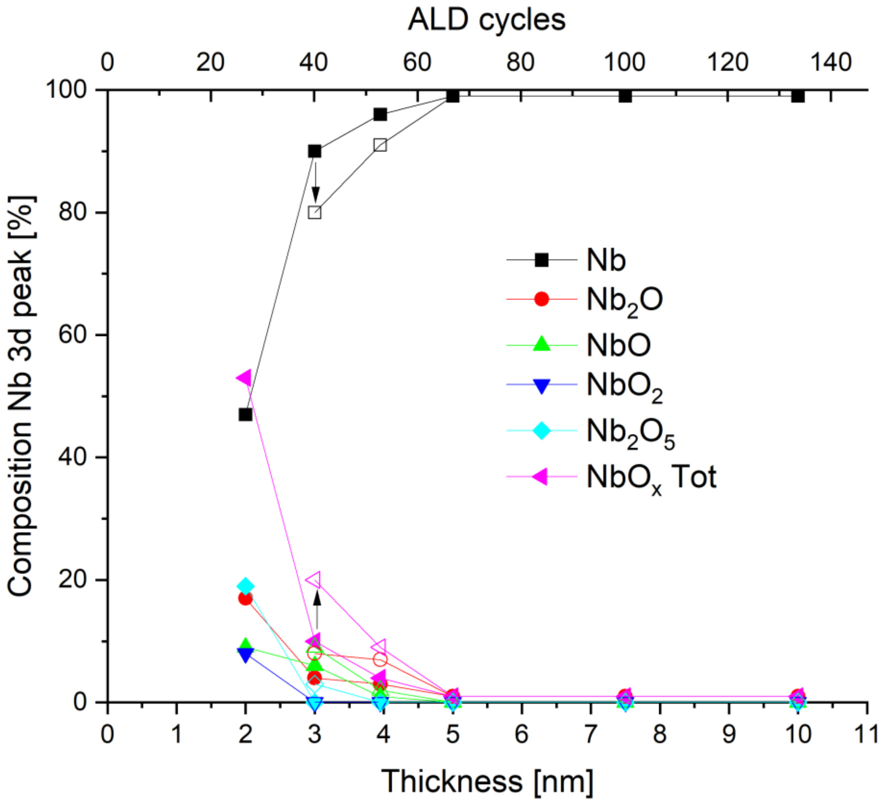
- The Nb remains passivated even with only 2,5 nm of Al_2O_3
- After HPR, we need at least 5 nm to protect the Nb surface.

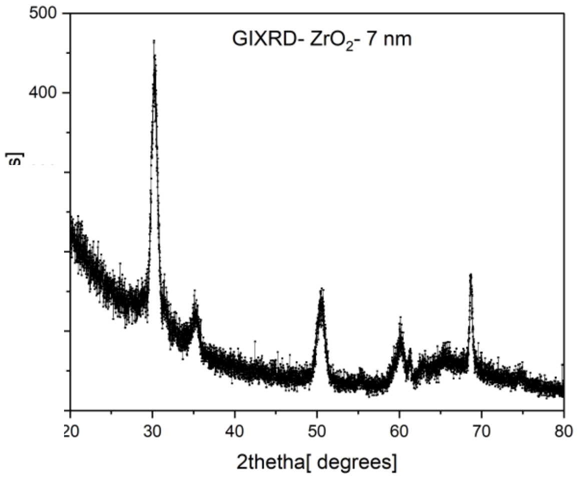
A crystalline oxyde: ZrO_2

- We tested different thickness of ZrO_2 on Nb + Annealing 800°C – 3hrs





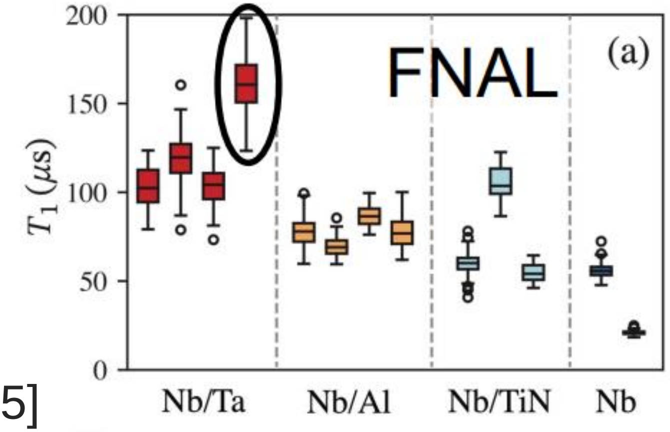




- We need at least 5 nm of ZrO_2 to passivate efficiently Nb.

Another condidate: Ta_2O_5

- Tantalum oxide results in lower losses than niobium oxide:
- Possibly due to consistency in Ta^{5+} valence state and low vacancy concentration.



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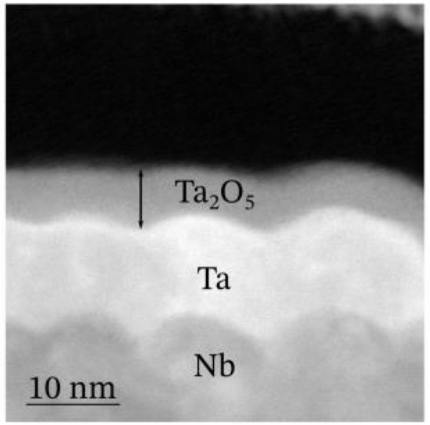
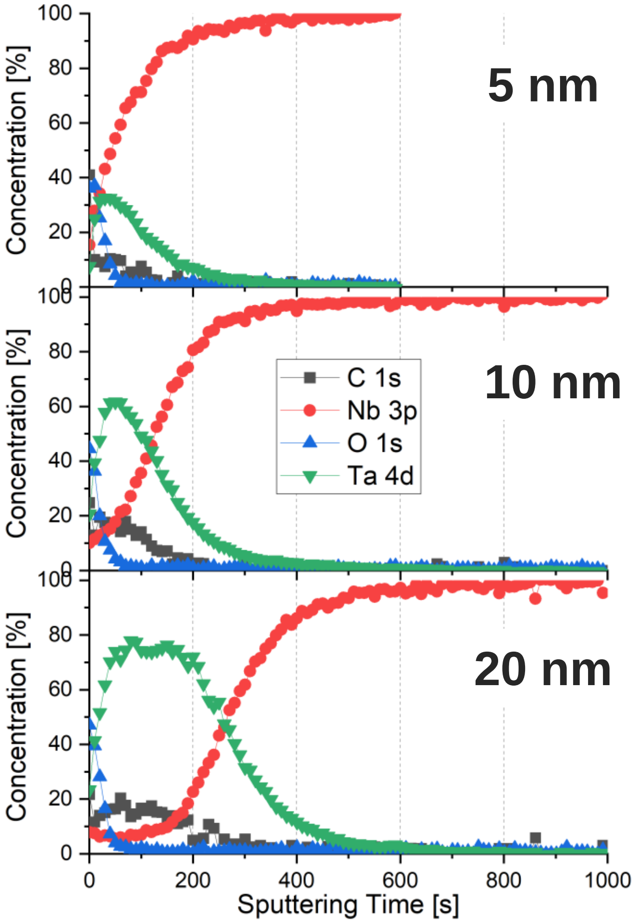
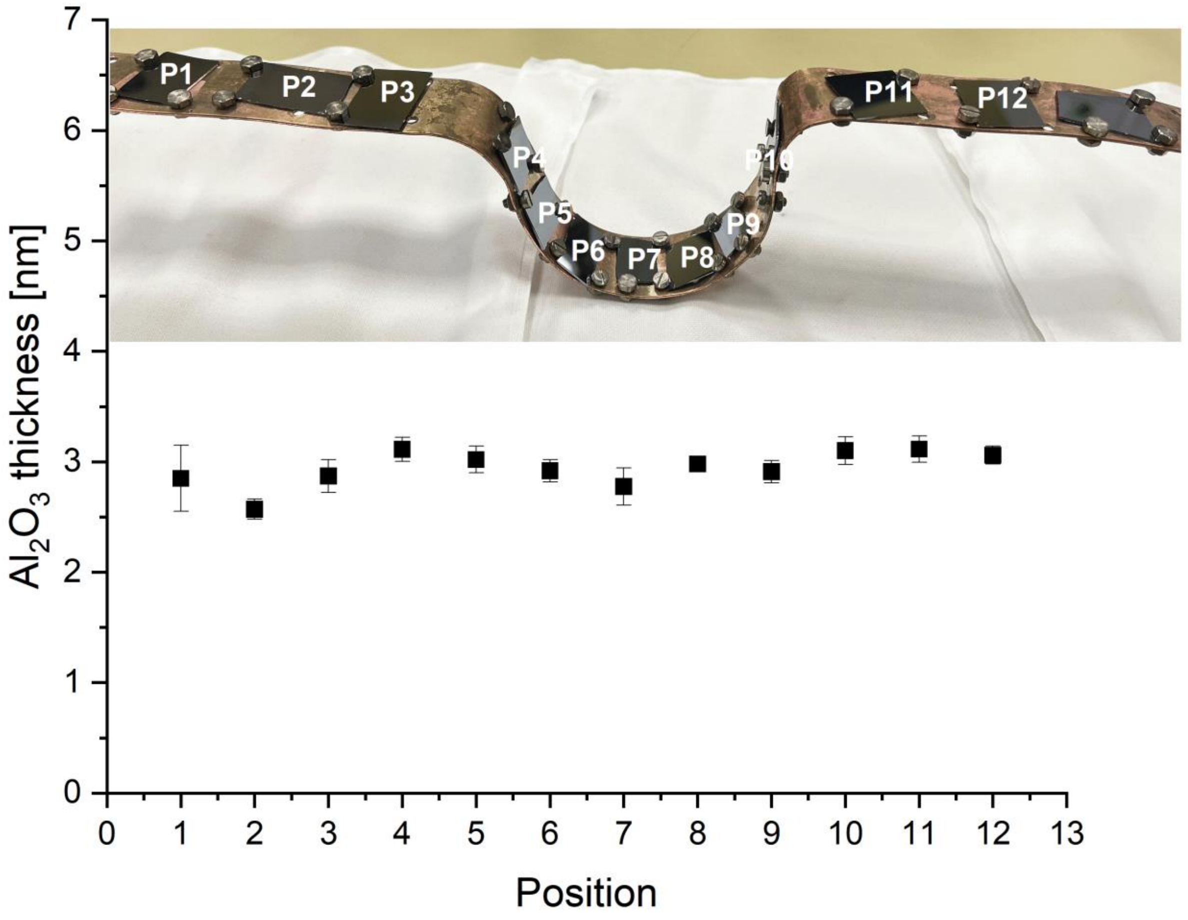


Annealed at 900°C-3hrs



- We observe intermixing between Tantalum and Niobium.
- Optimization is ongoing in collaboration with FNAL.

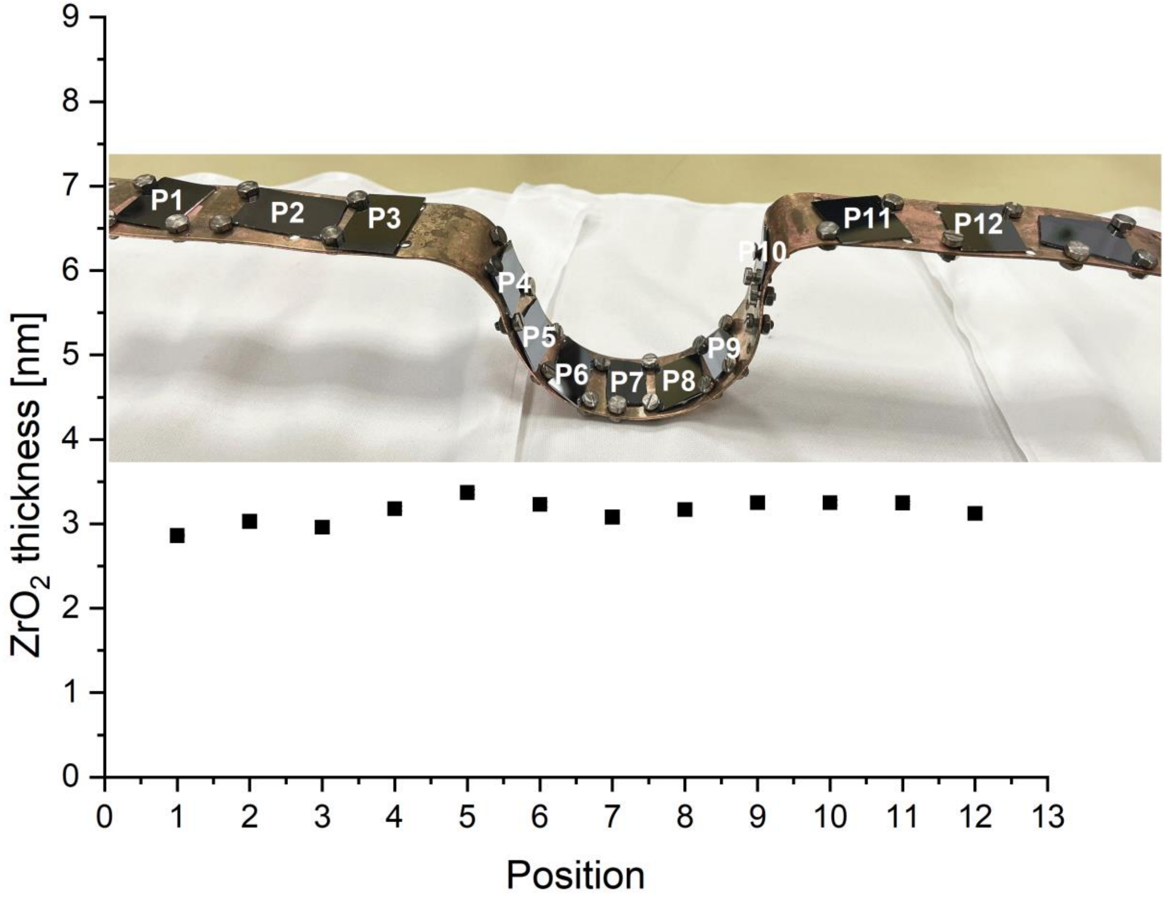
Thickness homogeneity of Al_2O_3 test on 1.3 GHz



- We performed homogeneity test of 2,5 nm of Al_2O_3 inside a 1,3 Ghz cavity.
- The precusors are TMA and H_2O at 250°C .

- We coated Two 1,3 Ghz Nb cavities and we are waiting for them to be tested one at CEA and one at FNAL.

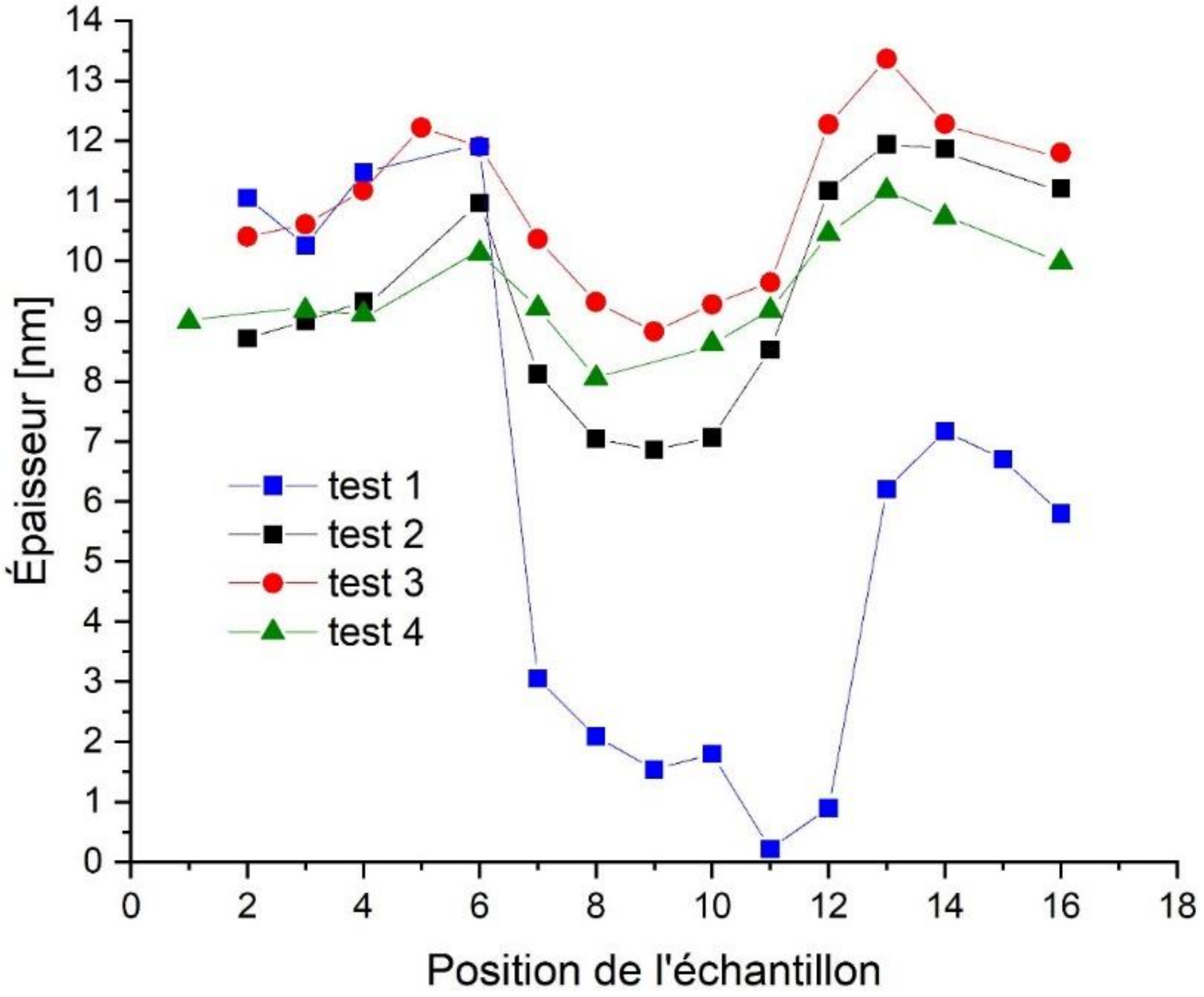
Thickness homogeneity of ZrO_2 test on 1.3 GHz



- We performed homogeneity test of 3 nm of ZrO_2 inside a 1,3 Ghz cavity.
- The precusors are $Zr(NMe_2)_4$ and H_2O at 200°C .

- We coated one 1,3 Ghz Nb cavity and we are waiting for the RF test at FNAL.

Thickness homogeneity of Ta_2O_5 test on 1.3 GHz



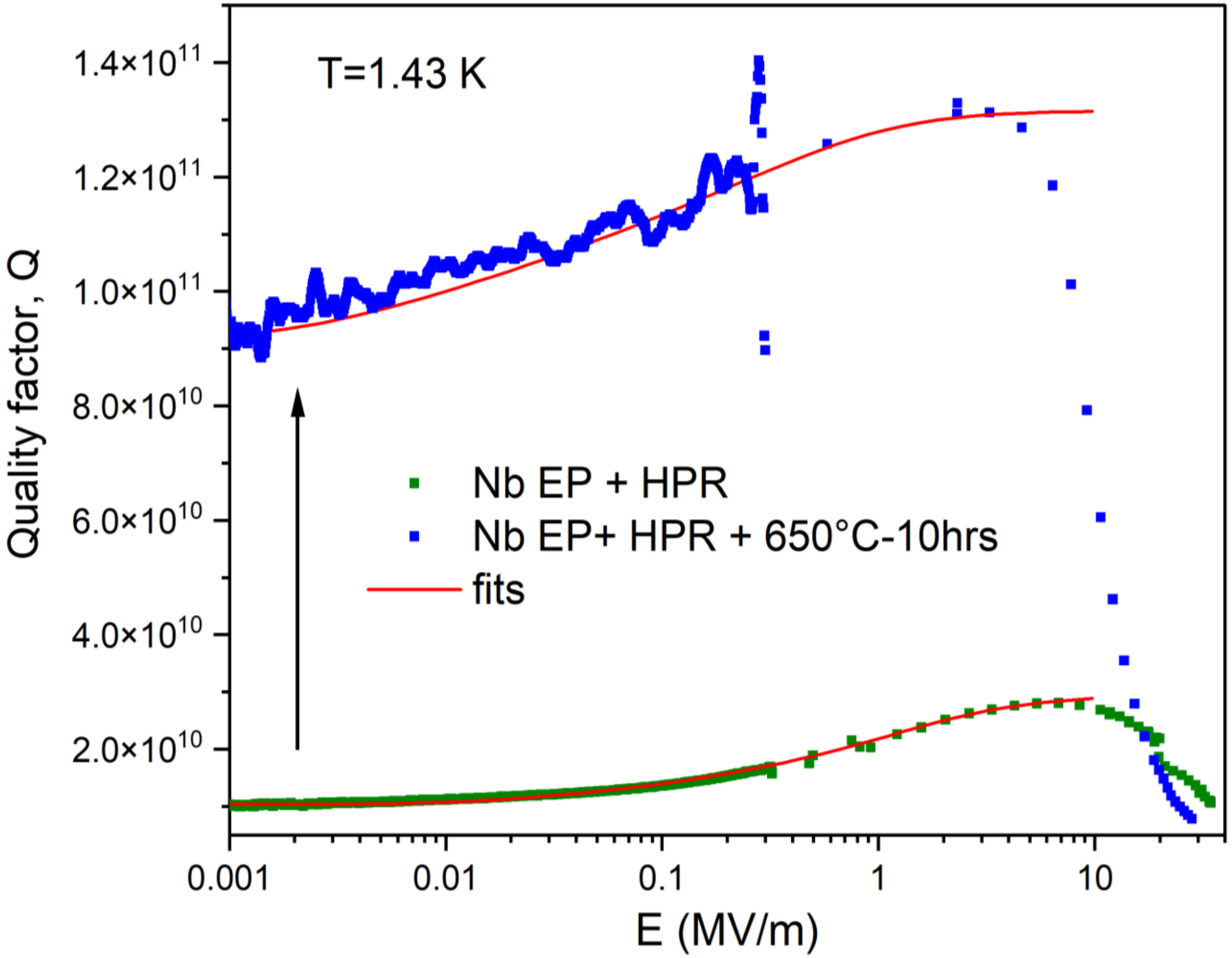
- ❑ ALD recepie:

$Ta(OC_2H_5)_5$ and H_2O at 250°C

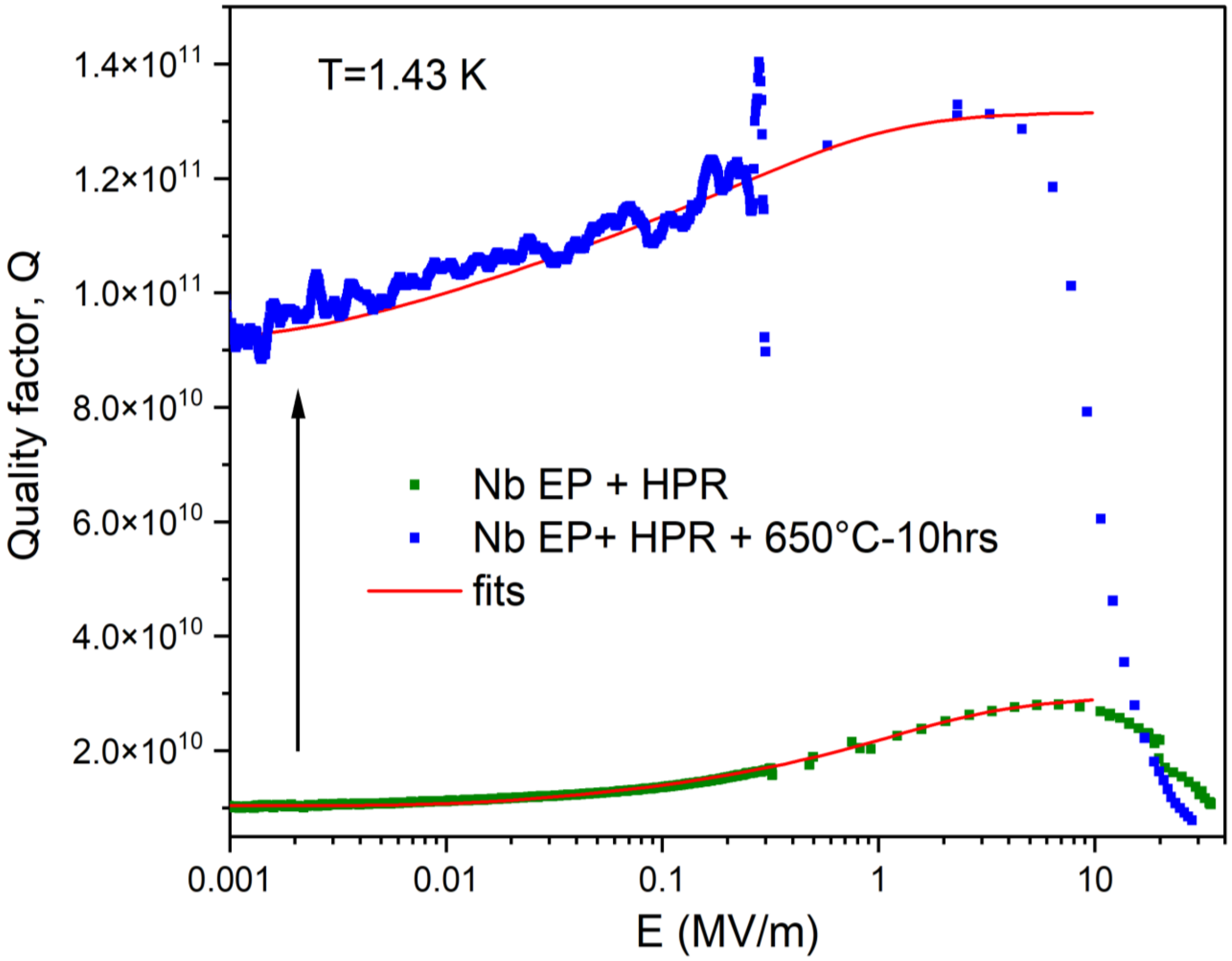
Test1: 1,5s / 20s / 1,5s / 15s
Test2: **2,5s** / 20s / 1,5s / 15s
Test3: **3s** / **60s** / 1,5s / 15s
Test4: 3s / 60s / 1,5s / **60s**

- ❑ We coated one 1,3 Ghz Nb cavity and we are waiting for the RF test at FNAL.

The highest coherence time ever measured on Air exposed Nb: T_1= 17 s at 1.4 K



The highest coherence time ever measured on Air exposed Nb: T_1= 17 s at 1.4 K



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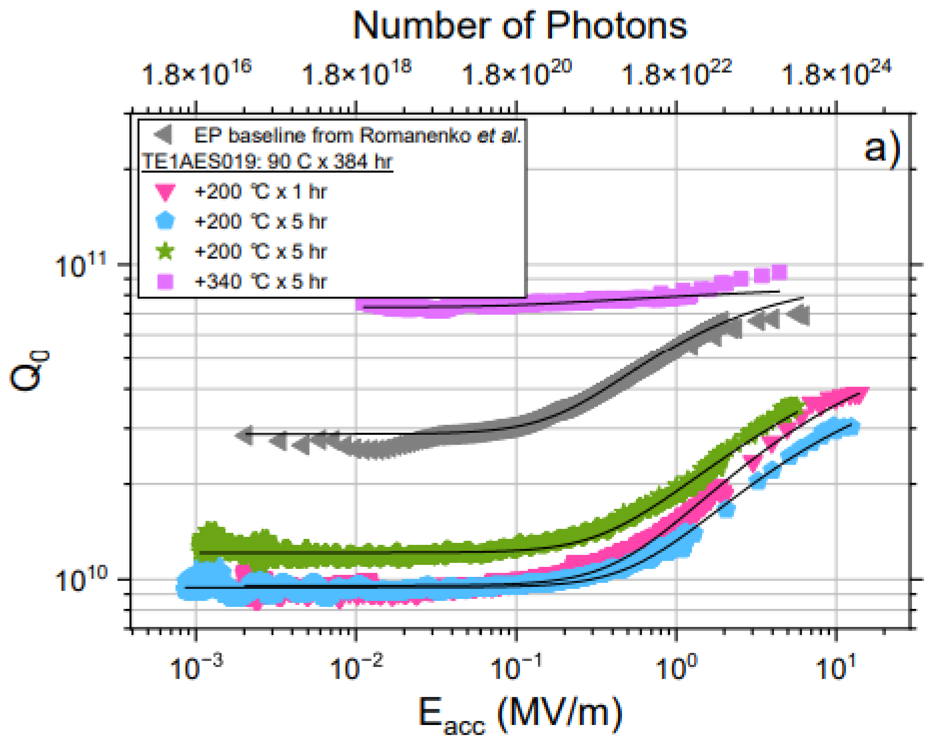
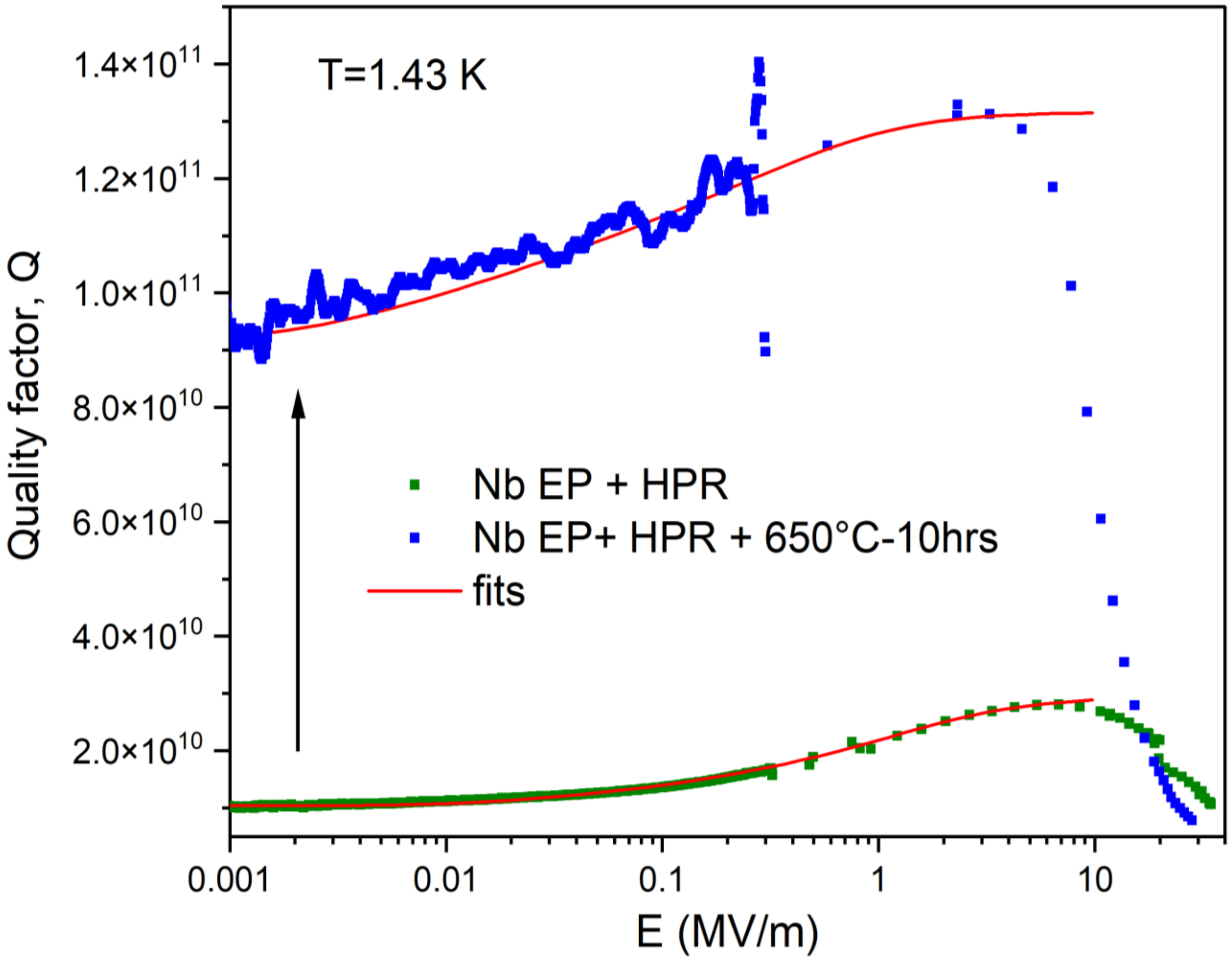


The highest coherence time ever measured on Air exposed Nb: T_1= 17 s at 1.4 K



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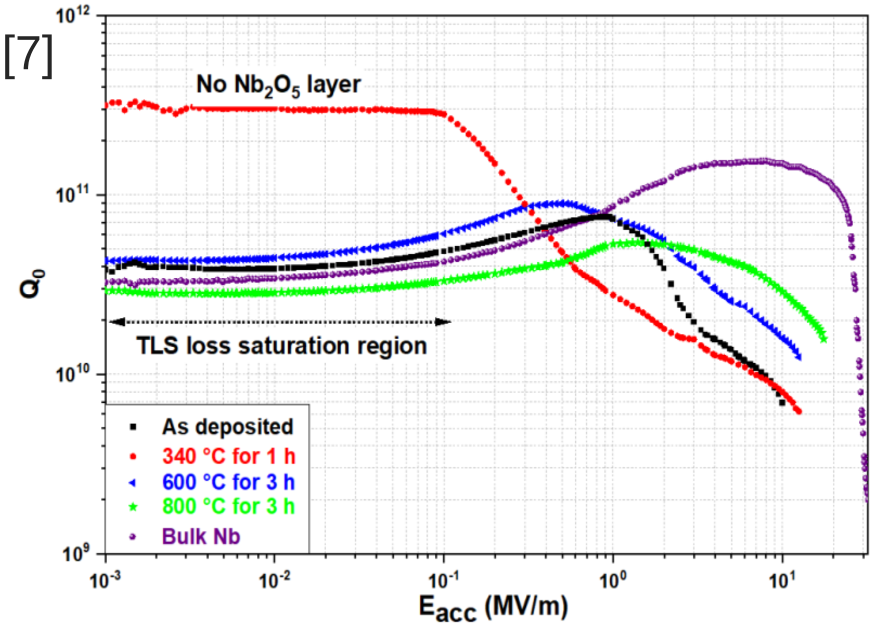

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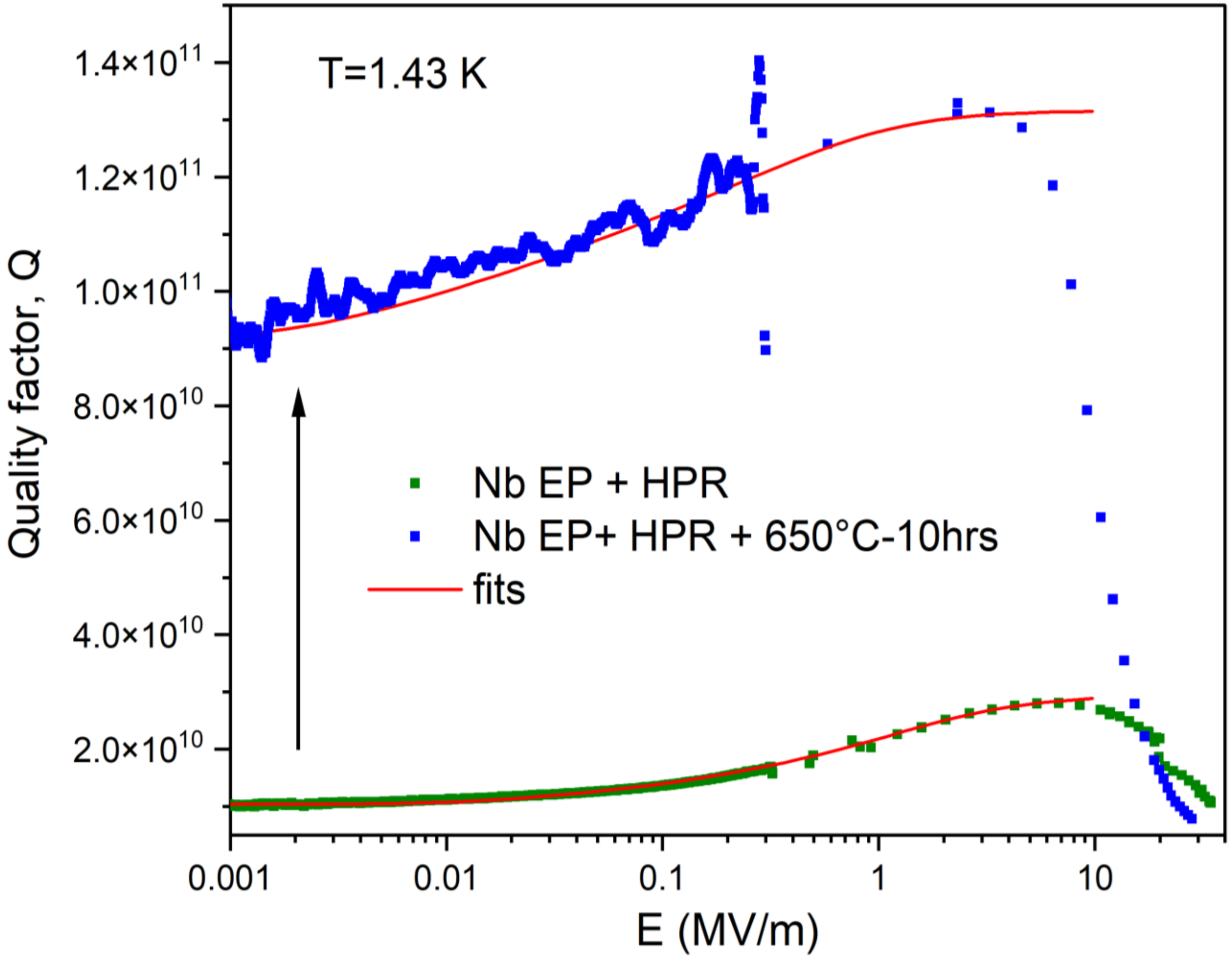
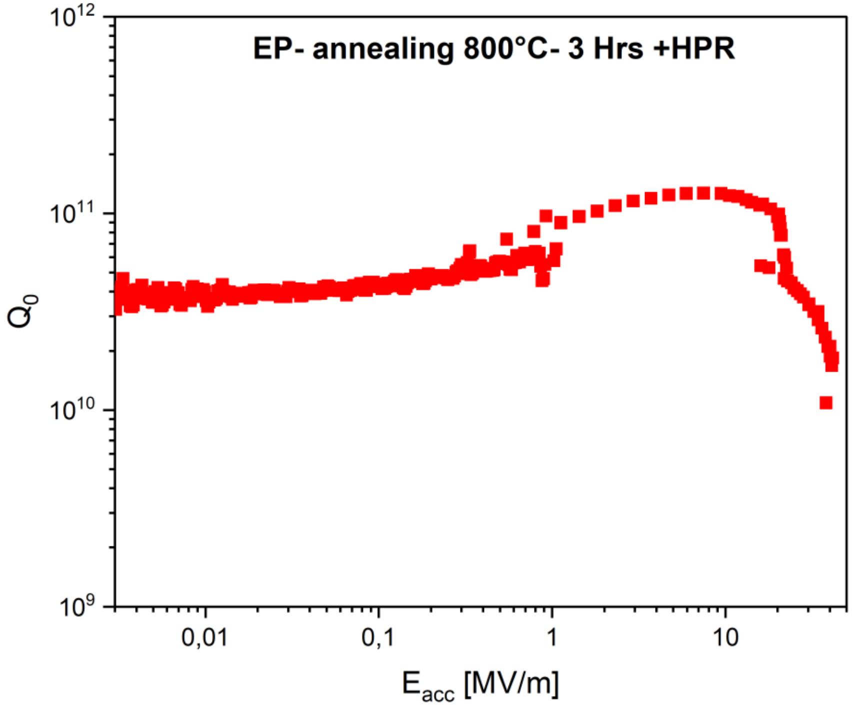
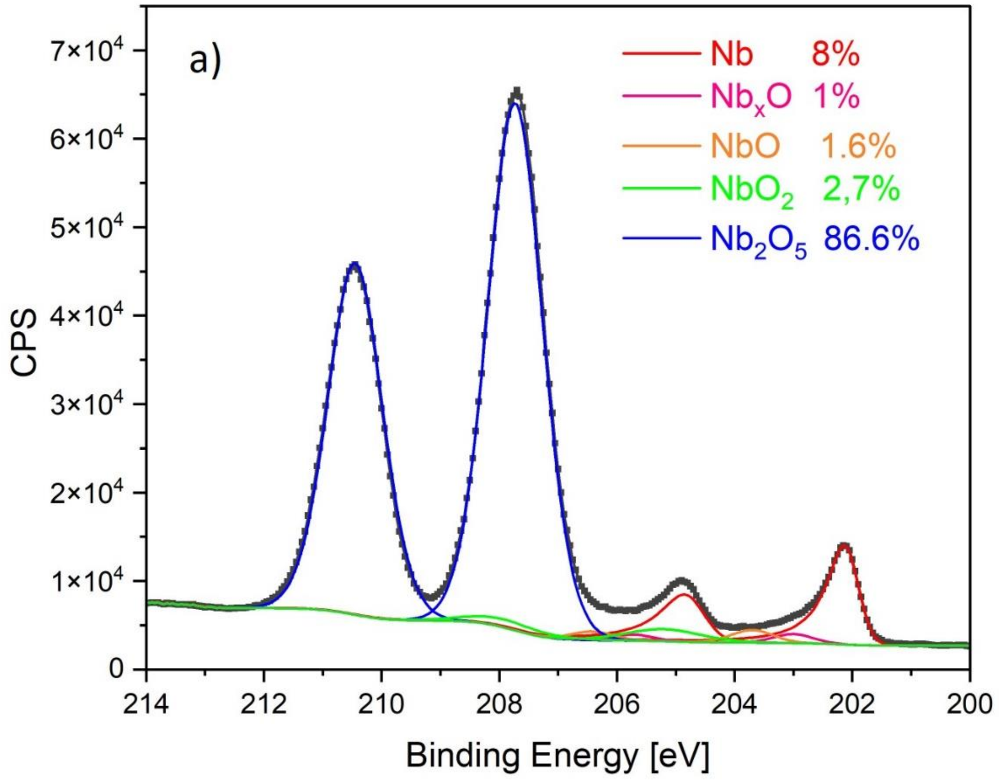
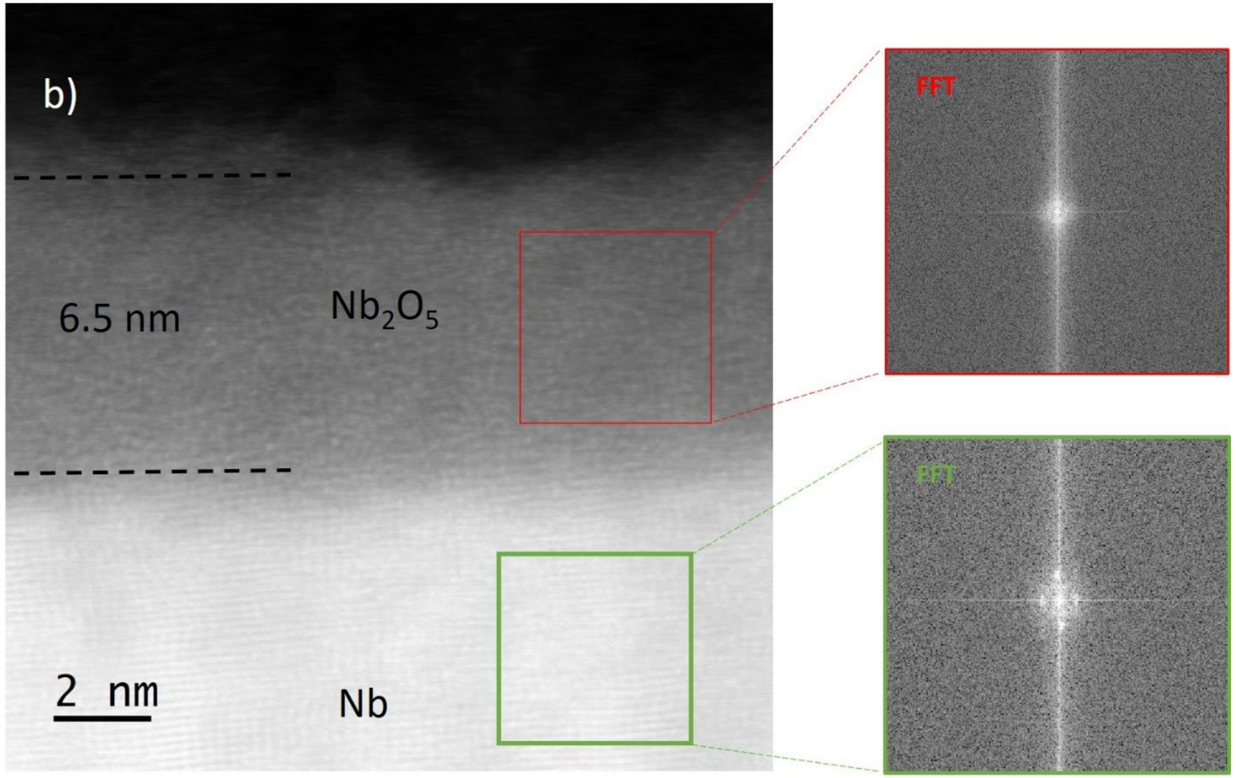




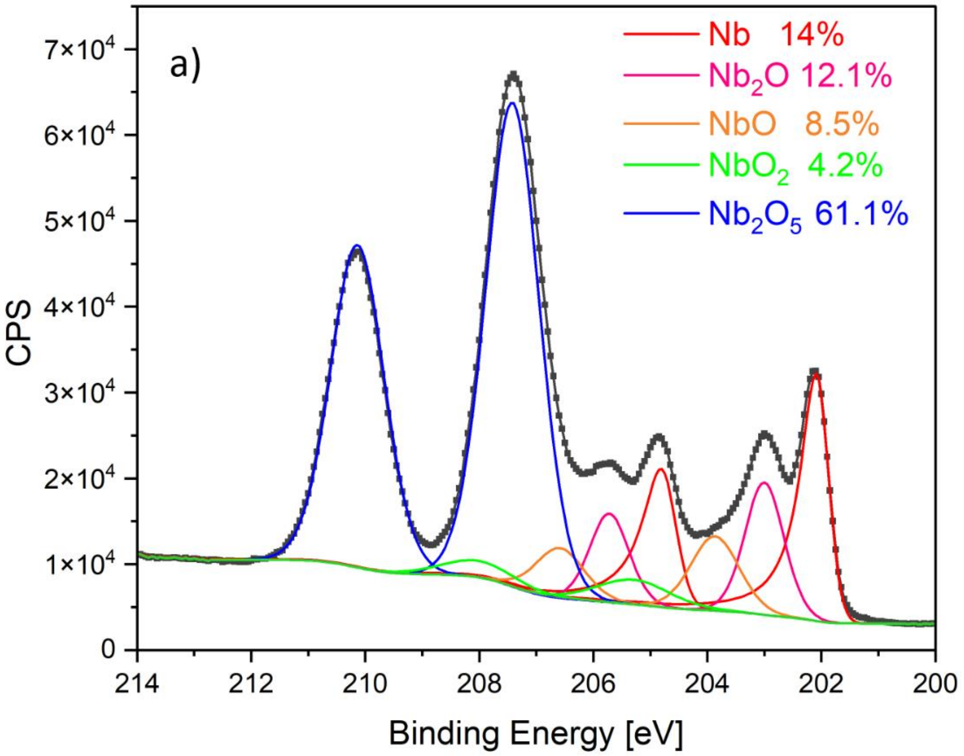
Before the annealing : EP + HPR

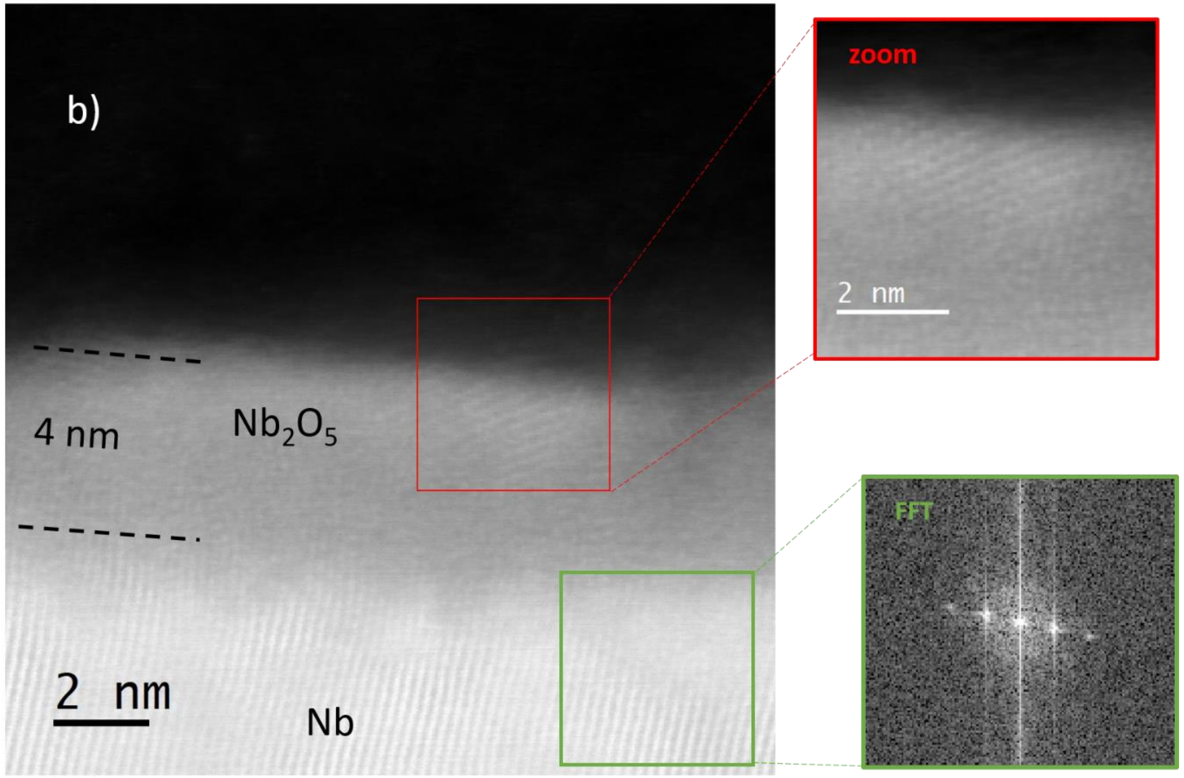




- We observe an amorphous Nb oxide of 6.5 nm thickness

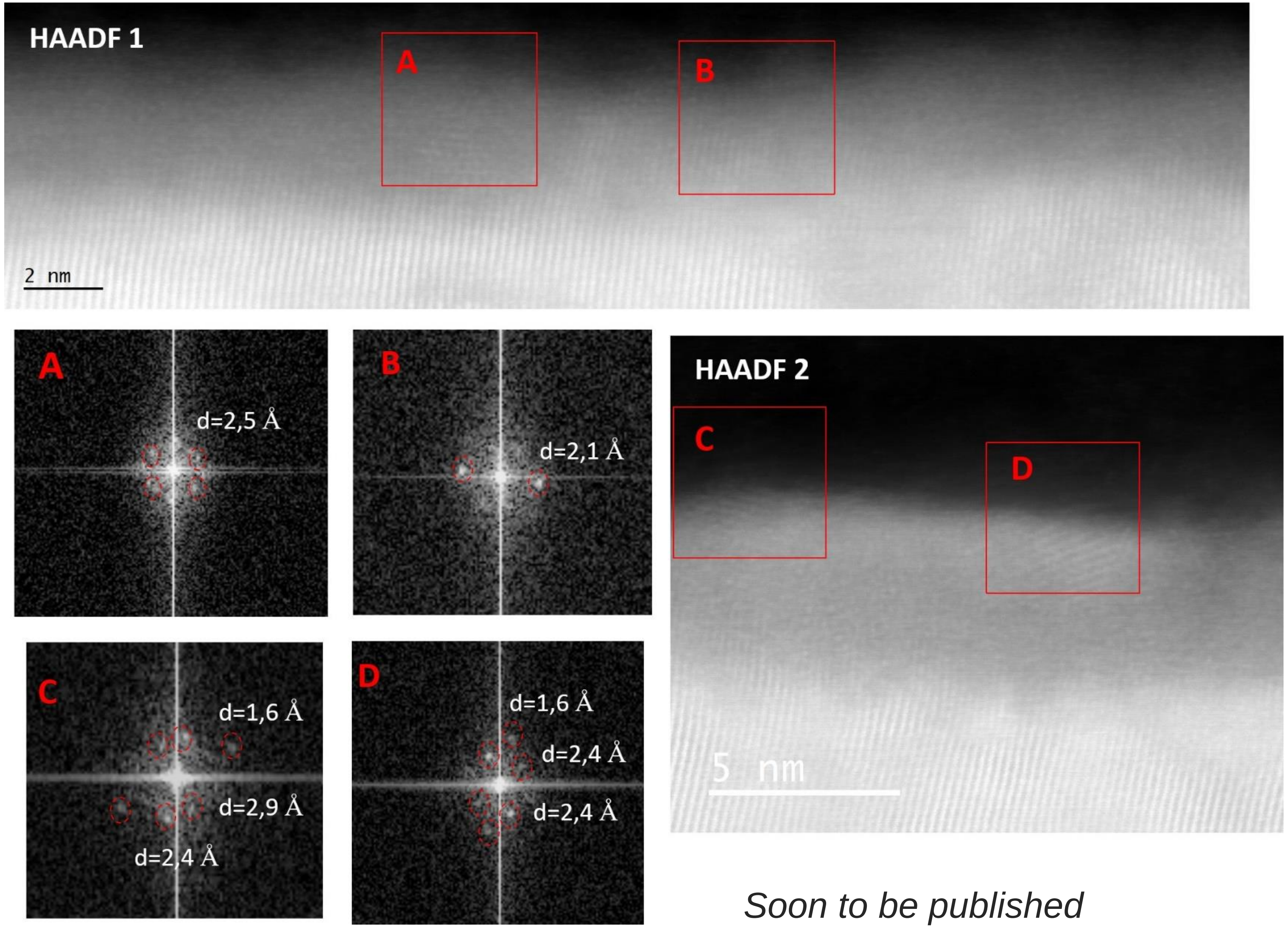
After Annealing: 650°C-10hrs + HPR





- After annealing, the oxide is significantly thinner and shows crystalline regions

After Annealing



Soon to be published

- FFT analysis shows that those crystalline regions are mainly NbO crystallites.
- We conclude that the annealing at 650°C- 10Hrs transfroms the Nb_2O_5 native oxide into partially crystalline NbO.
- When re-exposed to air, Nb_2O_5 regrows partially around these crystallites.
- In-situ synchrotron analysis can be done to confirm this mechanism.

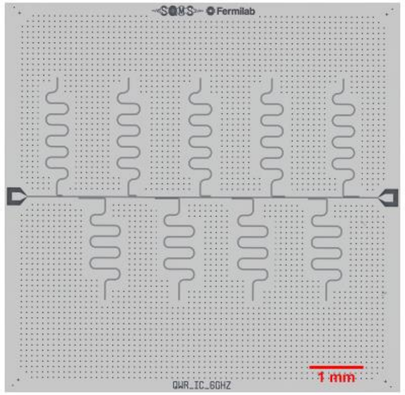
In the future ?

- We are waiting for RF tests on ZrO_2, Al_2O_3 and Ta_2O_5 cavities.
- We are collaborating with the qubits community in order to test this approach on 2D resonators and Qubits.

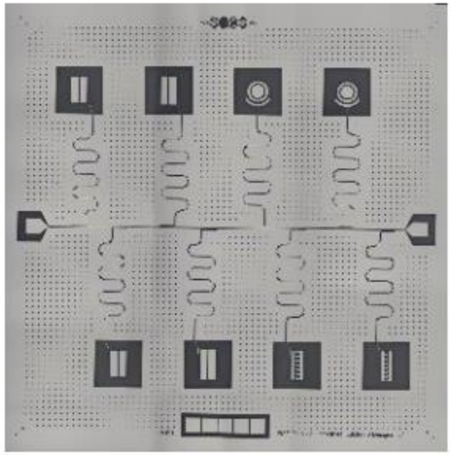
Thank you for your attention !

questions ?

2D resonators



2D resonators + Qubits



SRF cavities
(3D resonators)

References

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[2]] Romanenko, A., and D. I. Schuster. "Understanding quality factor degradation in superconducting niobium cavities at low microwave field amplitudes." *Physical Review Letters* 119.26 (2017): 264801.

[3] Roy, Tanay, et al. *Qudit-based quantum computing with SRF cavities at Fermilab*. No. FERMILAB-CONF-24-0026-SQMS. Fermi National Accelerator Laboratory (FNAL), Batavia, IL (United States), 2024

[4] Kalboussi, Yasmine, et al. "Reducing two-level systems dissipations in 3D superconducting niobium resonators by atomic layer deposition and high temperature heat treatment." *Applied Physics Letters* 124.13 (2024).

[5] Murthy, Akshay, et al. "Detailed Structural and Chemical Analysis of Amorphous Compounds in Superconducting Qubit Systems." (2024).

[6] Romanenko, A., et al. "Three-dimensional superconducting resonators at T< 20 mK with photon lifetimes up to τ= 2 s." *Physical Review Applied* 13.3 (2020): 034032.

[7] Abdisatarov, B., Bafia, D., Murthy, A., Eremeev, G., Elsayed-Ali, H. E., Lee, J., ... & Grassellino, A. (2024). Direct Measurement of Microwave Loss in Nb Films for Superconducting Qubits. *arXiv preprint arXiv:2407.08856*.