Atomic Layer Deposition of Tantalum Oxide

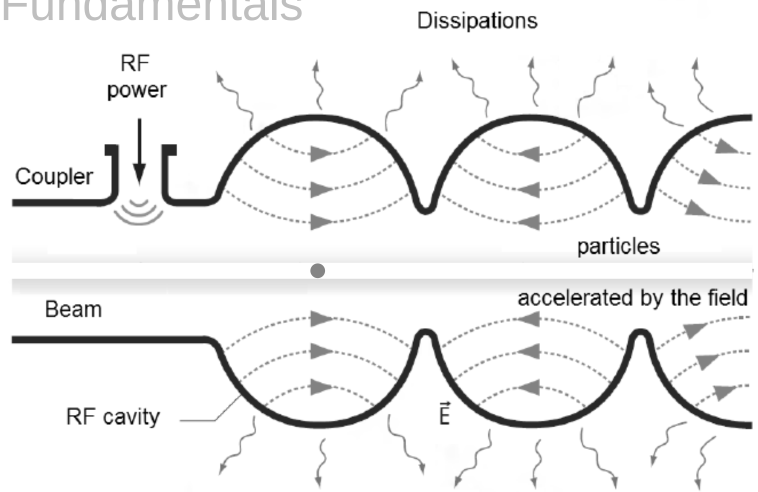
A New Material For Coating Cavities

Master Colloquium – Marco Voige

06.03.2025

Surface Losses Limit SRF Cavity Performance

Fundamentals



$$Q_0 = \frac{\omega_0 \cdot U}{P_c} = \frac{G}{R_s}$$

$Q_{0,max} \sim 1 \times 10^{11}$

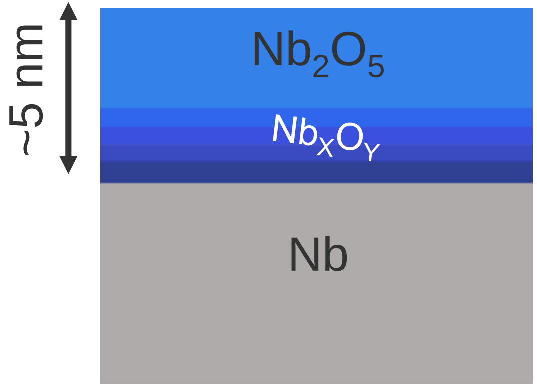
$$R_S = \frac{A \cdot f^2}{T} \cdot exp\left(\frac{\Delta(T)}{k_B \cdot T}\right) + R_0$$

~8 nΩ @ 2K, 1.3 GHz, **Nb**

~5 nΩ

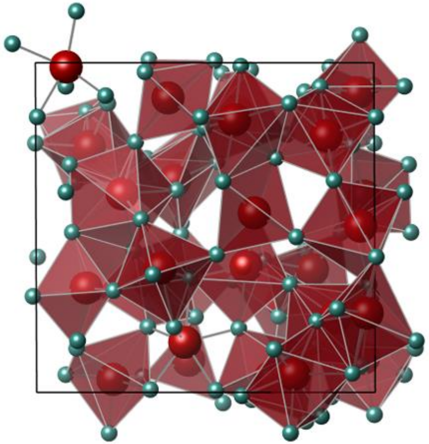
Oxide layers influence material properties (high SEY (Secondary Electron Yield)), which can trigger multipacting

Multipacting: Resonant electron avalanche leading to increased RF losses

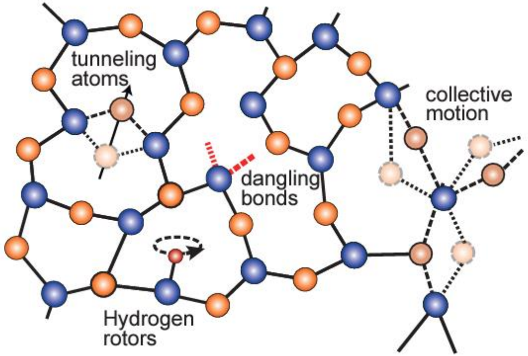


Nb_2O_5 Impacts SEY and Dielectric Losses

Fundamentals

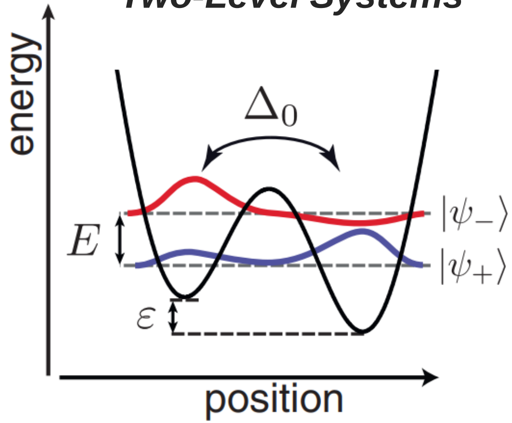


Sheridan, E., et al., arXiv preprint arXiv:2111.11684, 2021



C. Müller et al. Reports on Progress in Physics, 82(12), 124501

Two-Level Systems



C. Müller et al. Reports on Progress in Physics, 82(12), 124501

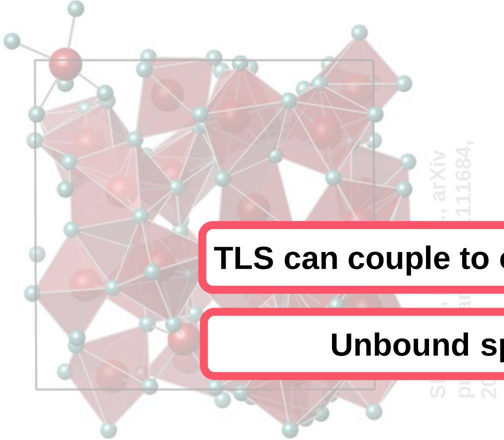
Amorphous Nb_2O_5 is energetically more favorable

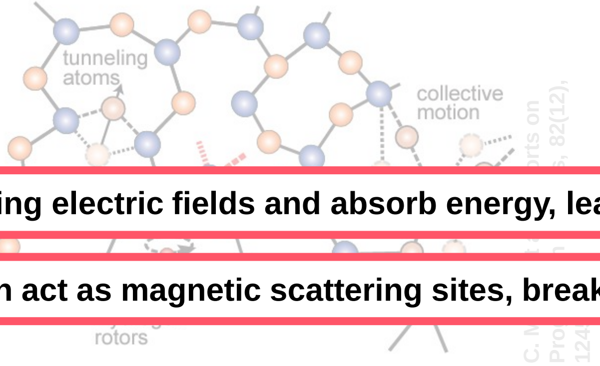
Non-stoichiometric phases lead to dangling bonds

Uncompensated spins create Two-Level Systems

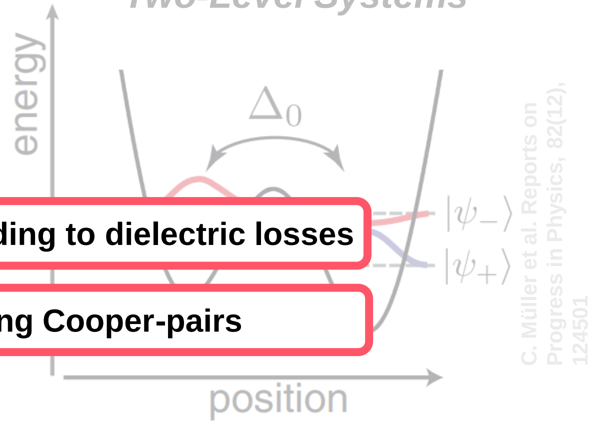
Nb_2O_5 Impacts SEY and Dielectric Losses

Fundamentals





Two-Level Systems



TLS can couple to oscillating electric fields and absorb energy, leading to dielectric losses

Unbound spins can act as magnetic scattering sites, breaking Cooper-pairs

Amorphous Nb_2O_5 is energetically more favorable

Non-stoichiometric phases lead to dangling bonds

Uncompensated spins create Two-Level Systems

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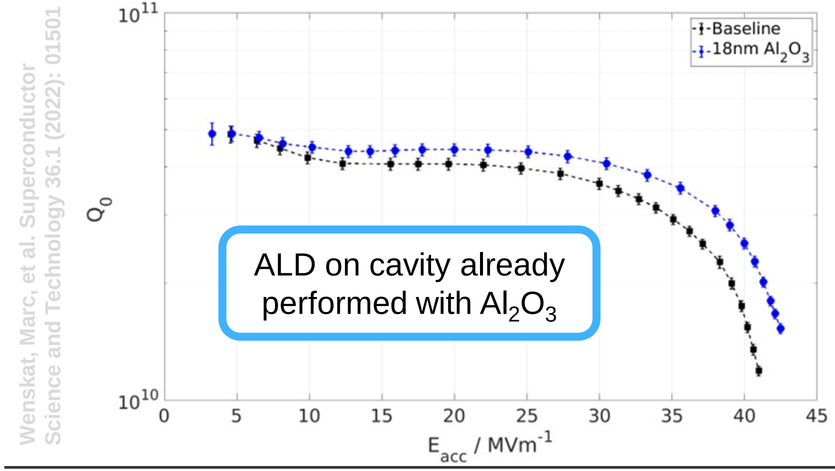
DESY.

Ta_2O_5 as a Potential Coating Material for SRF Cavities

Hypothesis & Motivation



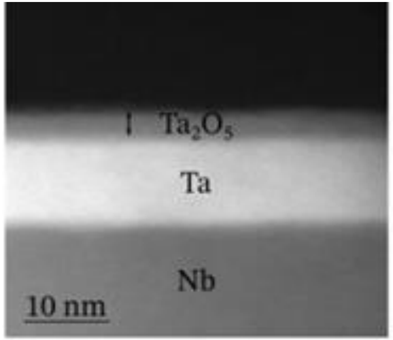
ALD on cavity already performed with Al_2O_3

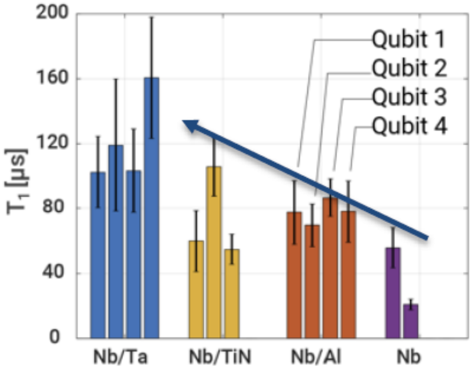
ex-situ	Baseline /Å	After Coating /Å	After Baking /Å
Nb_2O_5	20	18	-
Al_2O_3	-	172	170

SRF 2023 - Zaidman - WEPWB072

Baking removes the Nb_2O_5-layer!

Bal, Mustafa, et al., *arXiv preprint arXiv:2304.13257* (2023)

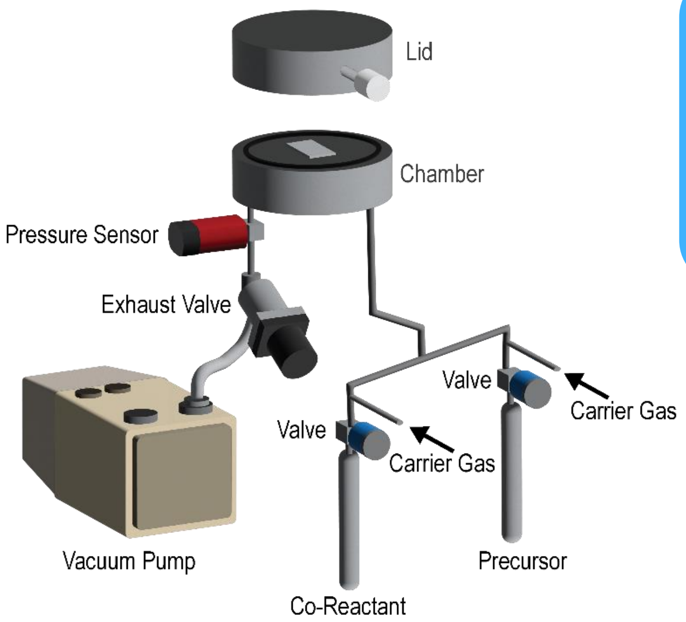




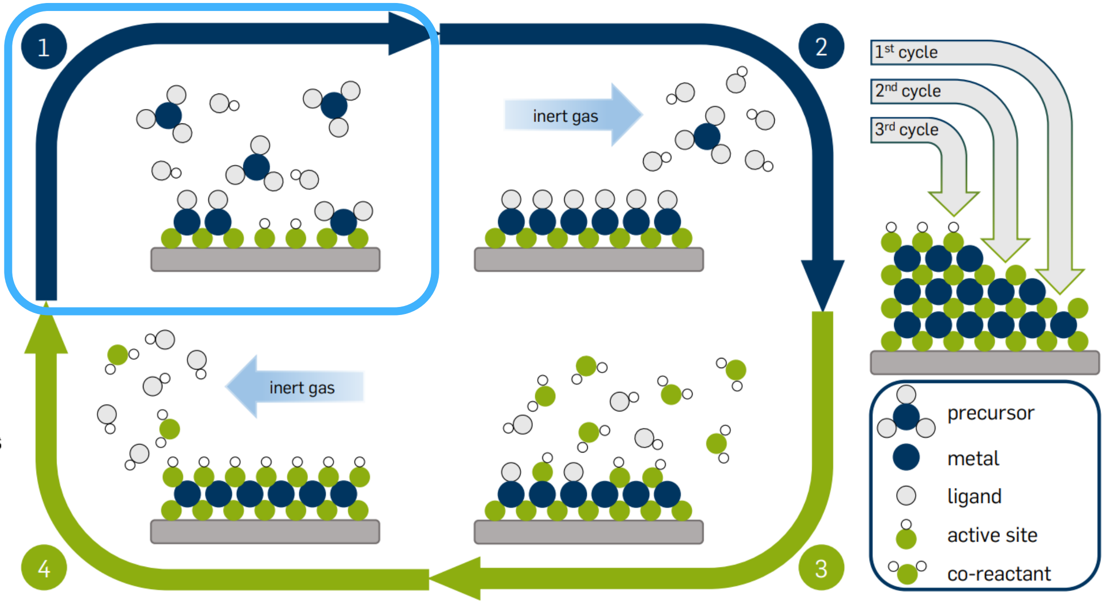
Ta_2O_5 exhibit lower TLS density than Nb oxides, which has been shown to improve qubit lifetimes

ALD Enables Precise Ta_2O_5 Thin Film Growth

Methodology – ALD



Schematic layout of the ALD system



L. Mai, (2020), DOI:10.13154/294-7658.

→ **thermal ALD requires two chemicals, one precursor: $Ta(OEt)_5$, one co-reactant: H_2O**

The deposited

ALD enables precise and uniform coatings through self-limiting reactions

Optimizing ALD for High-Quality Ta_2O_5 Coatings

Methodology – Process Adjustments

Systematic ALD recipe improvement

Run process → Measure thickness via spec. ellipsometry → Adjust process parameters

- -Process Times
- -Temperatures
- -Carrier gas flow

-P
-T
-C

Goal of the optimization

Deposited films should be:

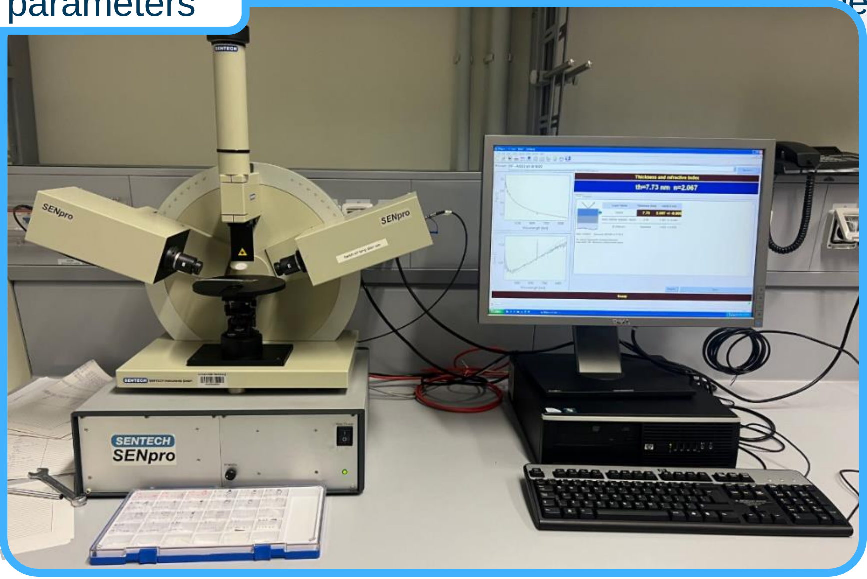
le

Points of referenc

Literature values for the deposition of Ta_2O_5 with $Ta(OEt)_5$:

Deposition temperature: **150 – 325°C**

Growth Per Cycle: **0.03 – 0.08 nm/Cycle**



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Optimizing ALD for High-Quality Ta_2O_5 Coatings

Methodology – Process Adjustments

Final recipe for planar samples (GPC ~ 0.07 nm/Cycle)

Phase		Time /s
Internal Boost	Purge	5
	Prec. Puls	30
	Pulse	2
	Purge	10
Precursor ($Ta(OEt)_5$)	Pulse	0.5
	Exposure	10
	Purge	60
Co-Reactant (H_2O)	Pulse	0.5
	Exposure	10
	Purge	60

Position	Temperature /°C
Precursor	190
Co-Reactant	20
Chamber	200
Lid	200
Exhaust	120

N_2 Flow /SCCM	20

Main system modifications during optimization

- **Software Correction**
- **Fully Heated System**
- **Internal Boost**

Problem: Low material flow of the precursor due to the low vapor pressure

Problem: Material decomposition if precursor temperature is too high

Solution: Increasing the pressure in the precursor container

Optimizing ALD for High-Quality Ta_2O_5 Coatings

Methodology – Process Adjustments

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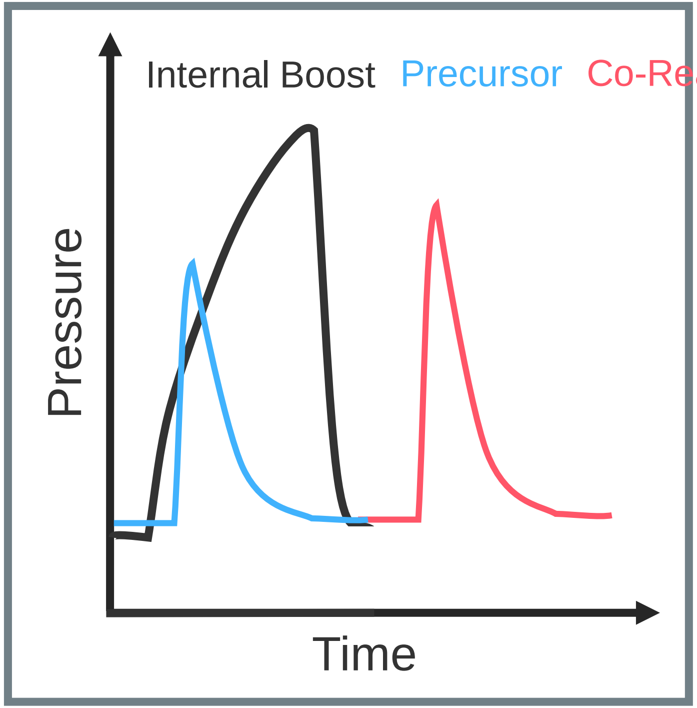
Examplary ALD Process **with internal boost**



No depo

GPC ~ 0.07 nm/Cycle

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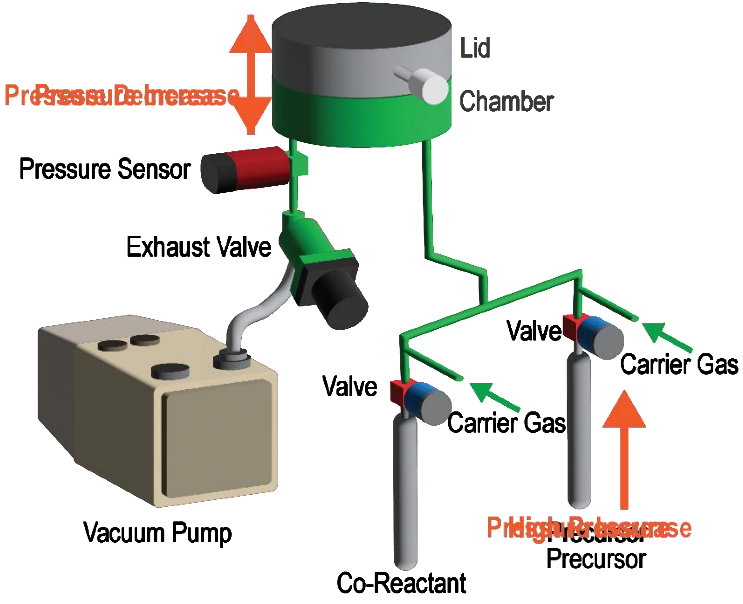
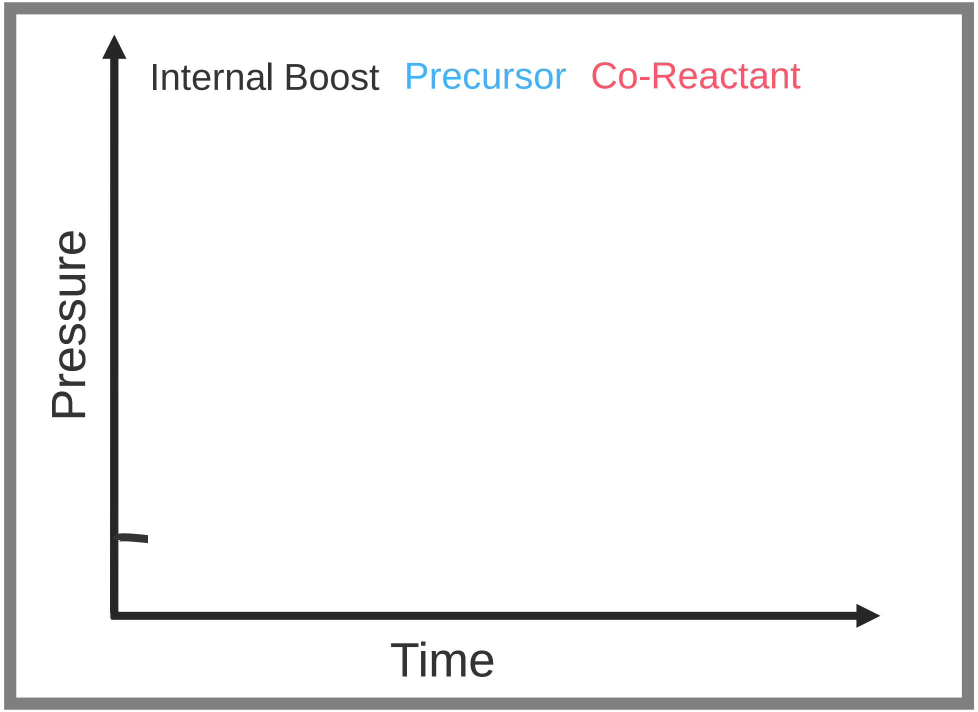


Examplary ALD Process **with internal boost**



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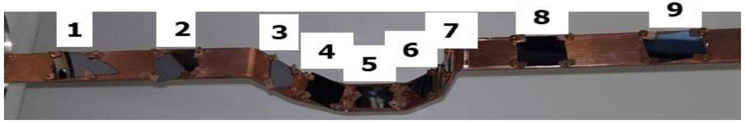
Cavity Coating Challanges – Full Coverage & Uniformity

Discussion – Challenges in Application

Goal: Adapt ALD recipe to cavity geometries

Sample holder enables measurements of the thickness gradient in the cavity

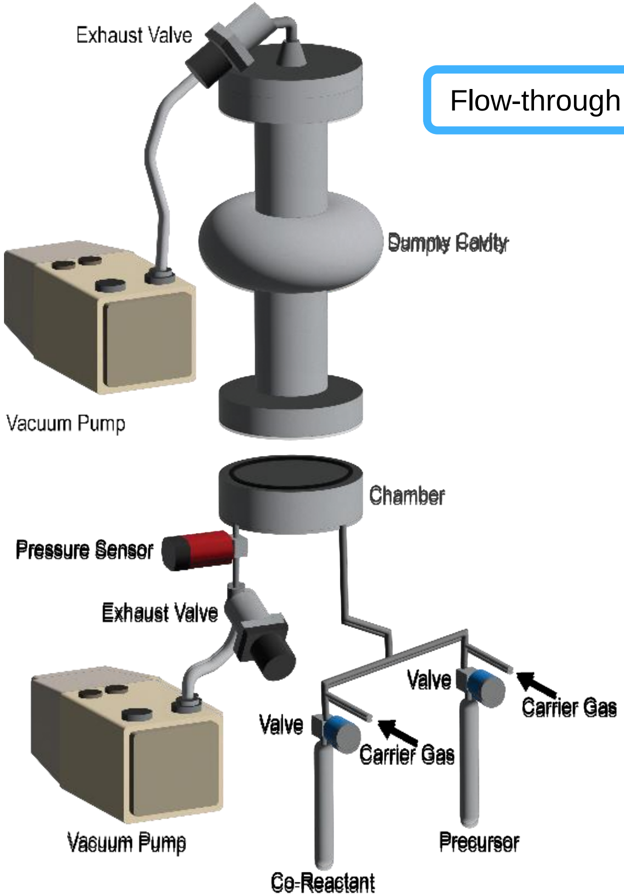


Dummy cavity sample holder for planar Si samples

Problem: Recipes show high thickness gradients across the cavity volume

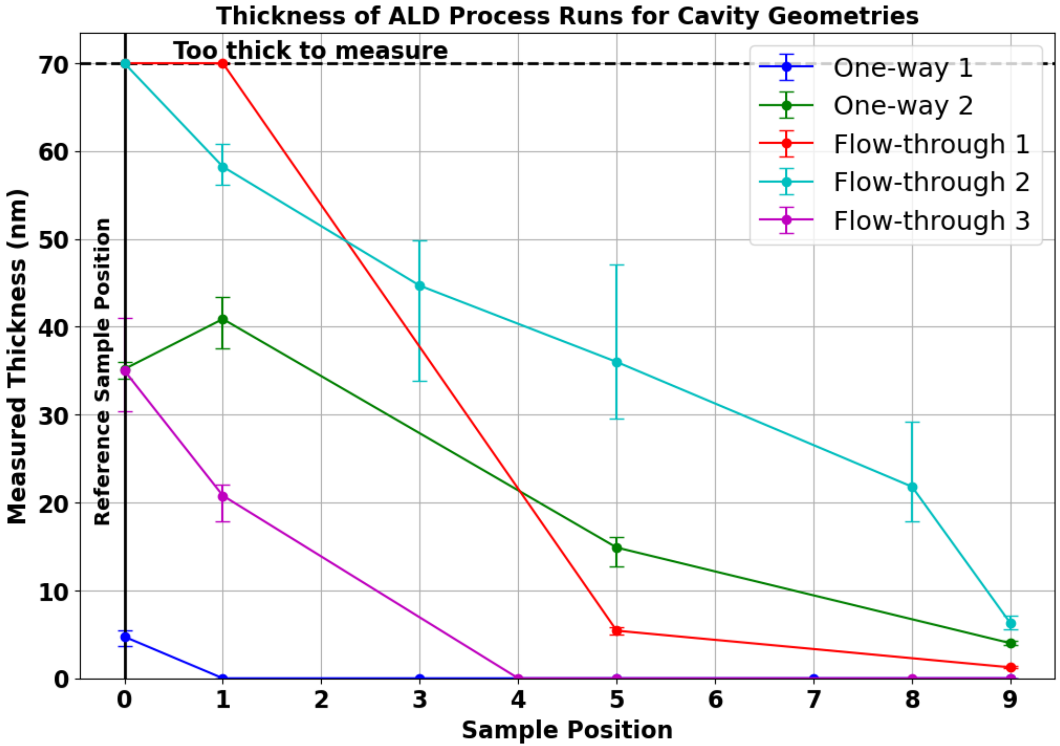
Possible Solution: Flow-Through Systems (chemicals must pass through the entire cavity)

Flow-through System



Cavity Coating Challanges Impact Uniformity

Discussion – Challenges in Application



Recipe parameter set changed between the runs

Full coverage of the cavity accomplished

Uniform cavity coating not achieved

But: flow-through system enables thicker coatings in the top tube of the cavity

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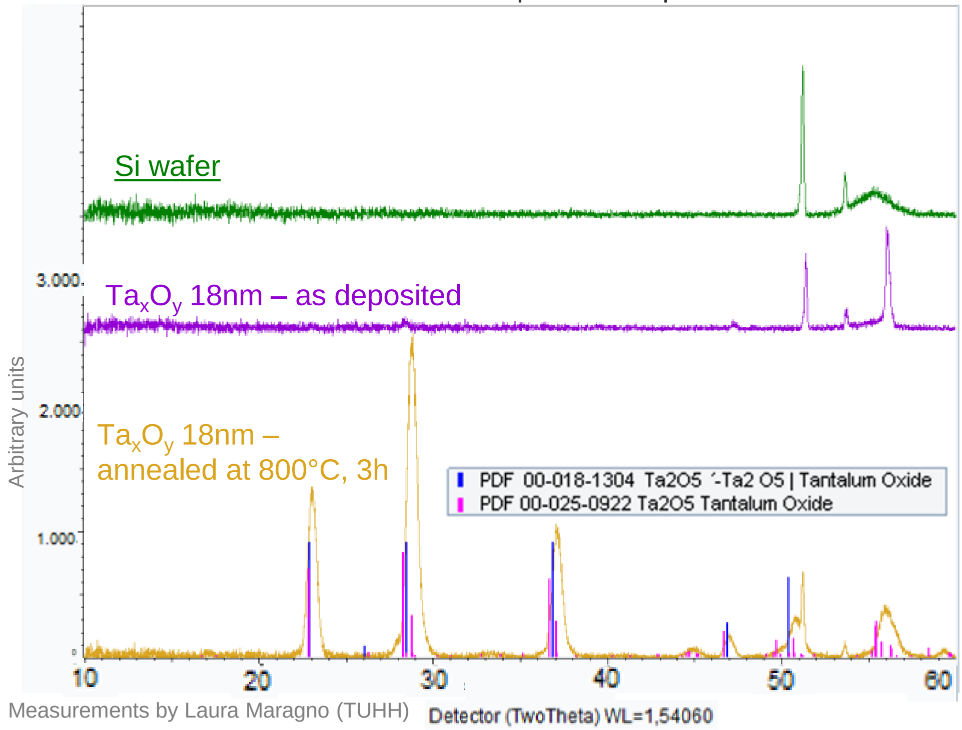
XRD Confirms Crystallization of Ta_2O_5

Results

Measurement of 3 planar samples



Measurements by Laura Maragno (TUHH)

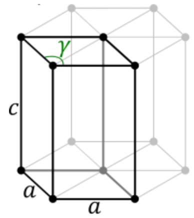
XRD Analysis was conducted to investigate the structural properties of Ta_2O_5

Increased crystallinity in Ta_2O_5 films can reduce the presence of TLS

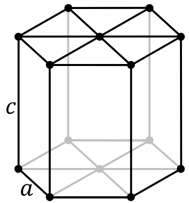
As-deposited Ta_2O_5 films are amorphous

Analysis of annealed sample indicates the formation of a crystalline phase

Hexagonal cell



Orthorhombic cell



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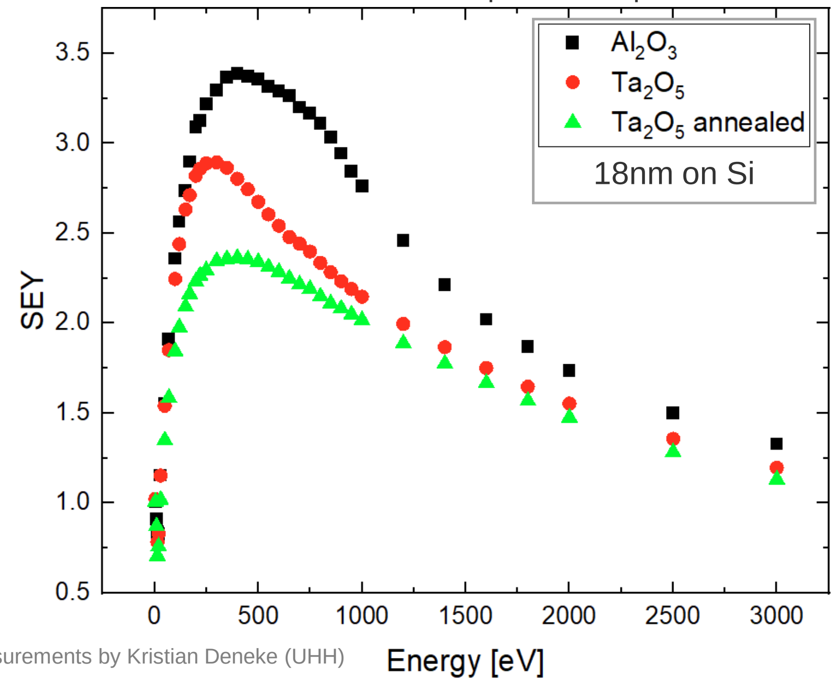
SEY Measurements Shows Lower Electron Emission in Ta_2O_5

Results

Measurement of 3 planar samples



Measurements by Kristian Deneke (UHH)

Air exposed niobium has an SEY of 2.2

R. Noer, S. Mitsunobu, Y. Kijima, and K. Saito. Secondary electron yield of nb rf cavity surfaces. 10th Workshop on RF Superconductivity, 2001.

A high SEY increases the likelihood of secondary electron avalanches, which can lead to multipacting

As-deposited Ta_2O_5 shows a significant decrease in SEY compared to as-deposited Al_2O_5

Post-deposition thermal annealing at 800°C for 3h decreased the SEY to ~2.4

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Ta_2O_5 Shows Potential but Needs Optimization

Conclusion

- **ALD → planar samples**
  - Optimization resulted a uniform and reproducible coating

- **ALD → cavity structures**
  - Full coating coverage achieved
  - Thickness gradient across the cavity observed (~70 to ~7nm)
    - Cause of gradient still under investigation

- **Material study**
  - XRD showed that crystalline Ta_2O_5 forms with annealing at 800°C for 3h
    - Crystalline structure may reduce the TLS density
    - Phase transformation changes dielectric properties
    - Al_2O_3 requires higher crystalizing temperatures >1000°C
  - Ta_2O_5 shows lower SEY value as Al_2O_3
    - Ta_2O_5 as-deposited shows reduction of ~14%, annealed of ~31% compared to Al_2O_3
    - Decreased SEY could reduce impact of multipacting in cavities

Further Research Can Improve Coating Quality

Outlook

- **Precursor Output Stability**

  → Unable to monitor precursor release dynamically. Installing a bubbler device could stabilize vapor pressure and improve reproducibility.

- **Potential Cavity Seal Leakage**

  → Seal operating near 200°C may cause leakage, affecting precursor distribution and coating uniformity

- **Cavity Recipe Requires Further Optimization**

  → Complex geometry demands further optimized process times and temperatures to ensure homogeneous deposition

The results demonstrate that Ta_2O_5 coatings offer promising properties for SRF cavities, with ALD proving to be a viable deposition method that can be further optimize to enhance its potential.

Acknowledgments

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