# Atomic layer deposition of tantalum oxide

## - a new material for coating cavities

TFSRF2024

19.09.2024

Marco Voige - on behalf of the SRF R&D Team Hamburg

# Reducing losses

## How can we improve cavities?

Improvement of SRF cavities

Idea: Getting rid of $Nb_2O_5$ (native oxide)

Removing $Nb_2O_5$ → Stop $Nb_2O_5$ from regrowing

Baking

Surface passivation

↓

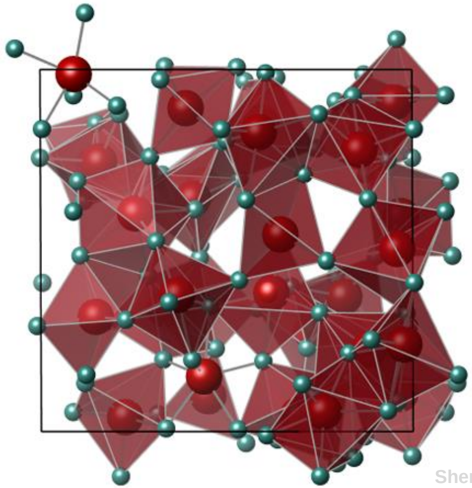
$Ta_2O_5$

# $Nb_2O_5$ as the potential cause of field losses

## How can we improve cavities? Surface passivation

Low (< 0.1 MV/m) — $E_{acc}$ (Number of photons) — High (> 10 MV/m)

**Cause of field losses**



Sheridan, E., et al., arXiv preprint arXiv:211111684, 2021

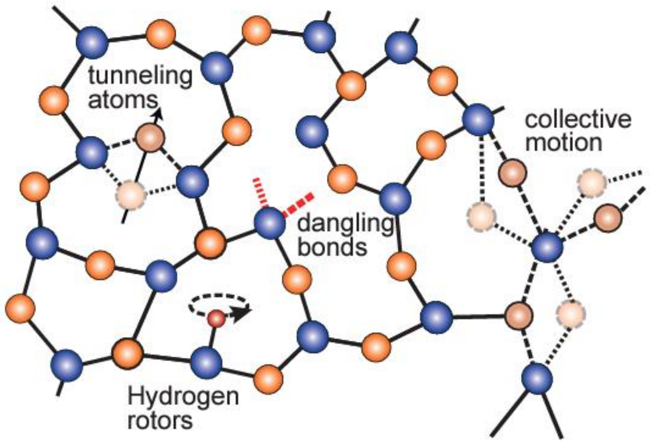
**Two-Level Systems**
Activation energy is taken from the rf field

**Magnetic impurities**
Spin B-Field breaking Cooper pairs

# Dangeling bonds result in Two-Level Systems (TLS)

## Why do amorphous materials lead to losses?

TLS-Variations




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

Schematic representation in an arbitrary amorphous material

**Local defects**

**Dangeling bonds**

**Unsatisfied valence electron bonds**

**Missing spin compensation**

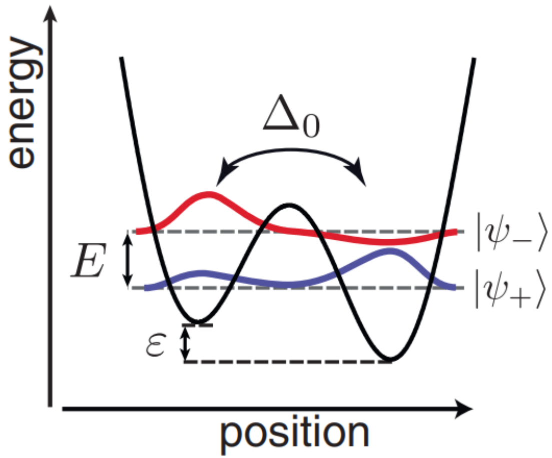
**Metastable system**

TLS

# Losses due to Two-Level Systems (TLS)

Quantum systems between two stable energy levels

Standard tunneling model




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

**Quantum system that can tunnel between two potential minima separated by a barrier**

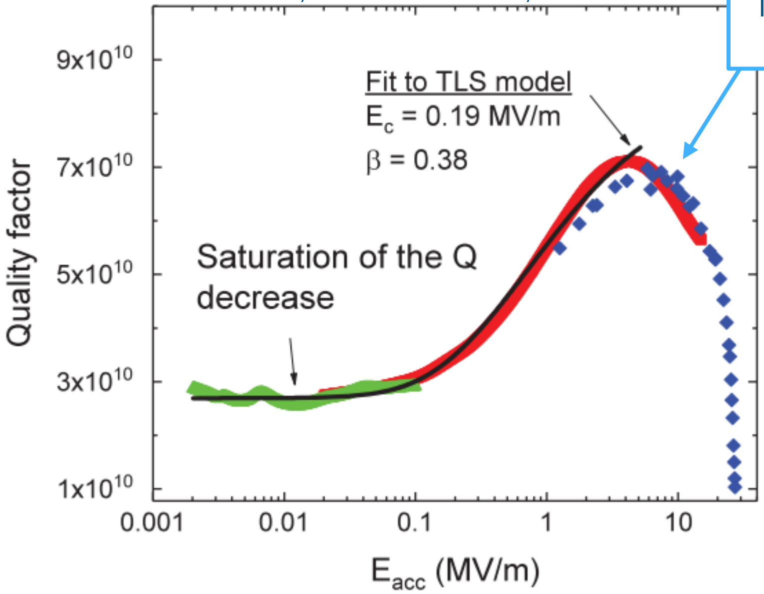
**Activation energy taken from rf field!**

- Thermal activation is suppressed at low temperatures / governed by quantum tunneling
- Random atomic arrangements
  → wide distribution of pot. barrier heights
  → large range of switching rates / eigenenergies

# TLS as the cause of *Low Field Q-Slope* (*LFQS*)

Temperature **fixed**, Gradient **varied**

1,3 GHz T=1,5K

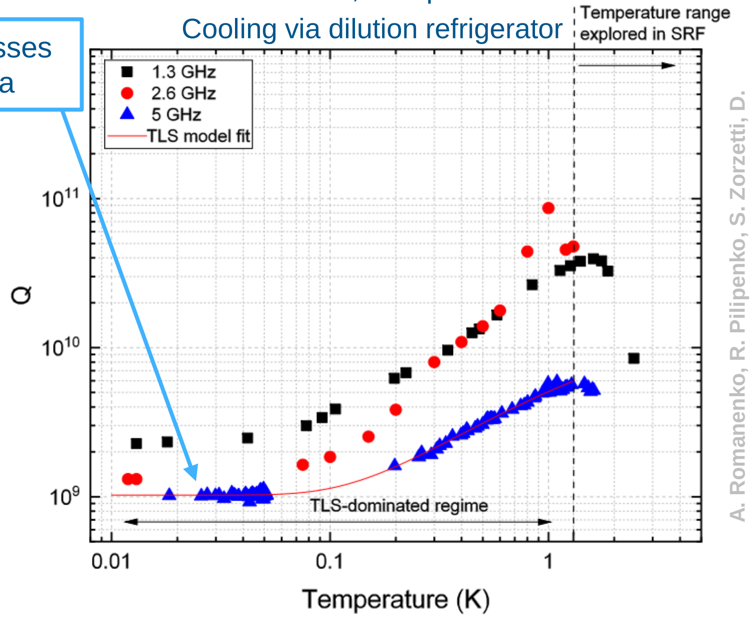



A. Romanenko and D. I. Schuster, Phys . Rev. Lett. 119, 264801 (2017)

Model of TLS losses describes data

Gradient **fixed**, Temperature **varied**

Cooling via dilution refrigerator




A. Romanenko, R. Pilipenko, S. Zorzetti, D. Frolov, M. Awida, S. Posen, A. Grassellino, arXiv:1810.03703

**Surface resistance only residual resistance, no BCS losses**

**Qubits: absorption of energy by TLS when tunneling between states results in noise / decoherence**

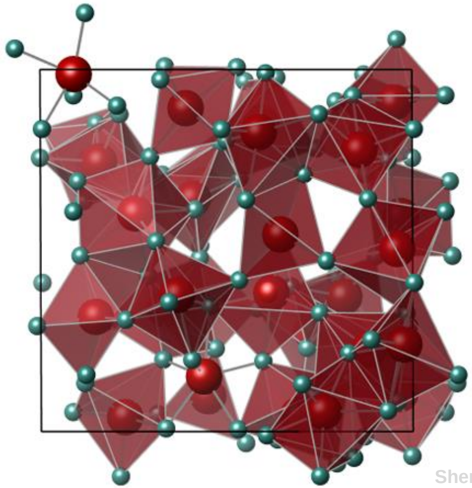
# $Nb_2O_5$ as the potential cause of field losses

## How can we improve cavities? Surface passivation

Low (< 0.1 MV/m) — $E_{acc}$ (Number of photons) — High (> 10 MV/m)

**Cause of field losses**

**Two-Level Systems**
Activation energy is taken from the rf field

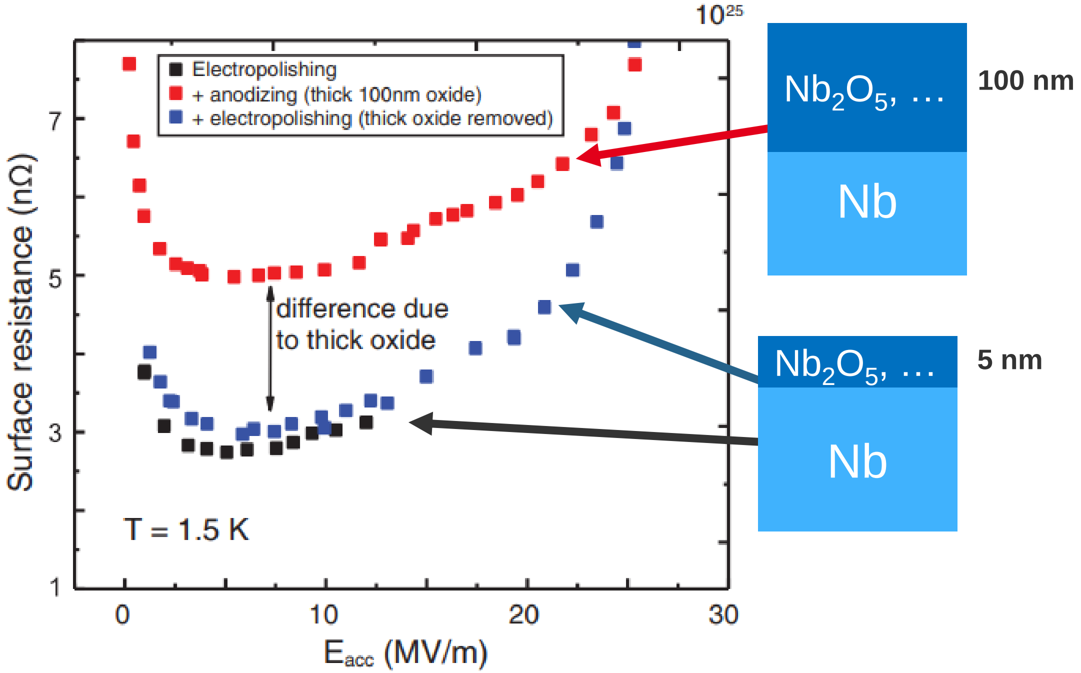


**Magnetic impurities**
Spin B-Field breaking Cooper pairs

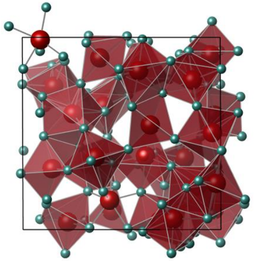
Sheridan, E., et al., arXiv preprint arXiv:211111684, 2021

# Magnetic impurities affect $R_s$

## How can we improve cavities? Surface passivation




A. Romanenko and D. I. Schuster, Phys . Rev. Lett. 119, 264801 (2017)



Sheridan, E., et al., arXiv preprint arXiv:2111111684, 2021

$Nb_2O_5$ is amorphous

Local non-stoichiometric

Unpaired Valence-electrons

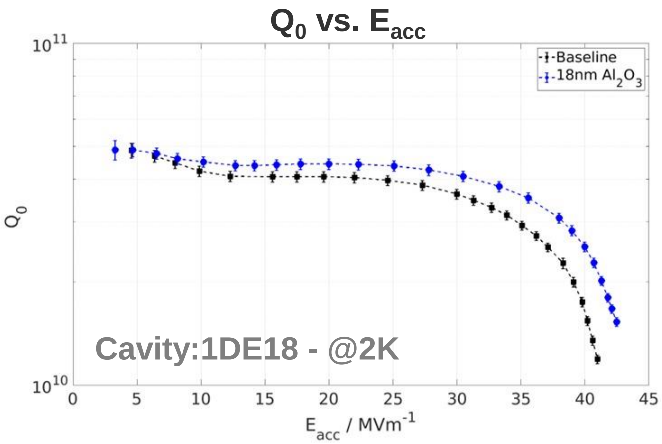
Magnetic Impurities

**$R_S$ affected**

# Coating / Passivation of the cavity surface

How do we prevent $Nb_2O_5$ from growing?

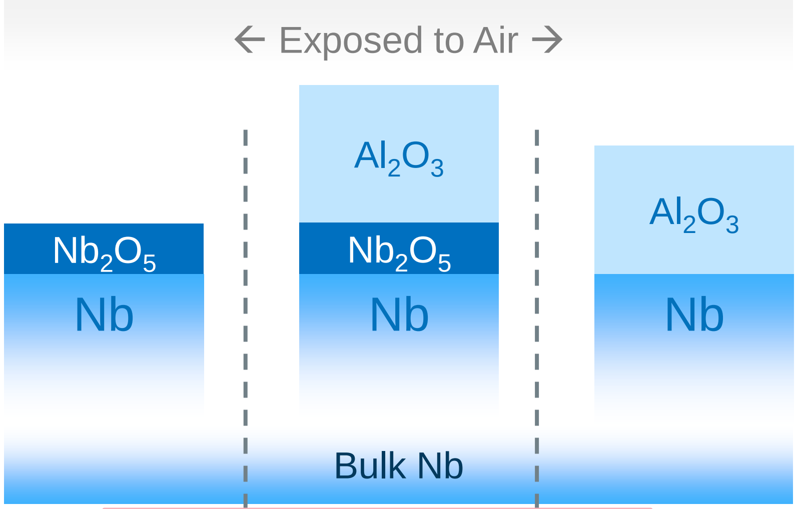
ALD on cavity already performed with $Al_2O_3$




**Insulator alone does not affect quality**

**+**

**Insulator prevents new $Nb_2O_5$ from forming**




**Baking removes the $Nb_2O_5$-layer!**

| *ex-situ* | Baseline / Å | After Coating / Å | After baking / Å |
|---|---|---|---|
| $Nb_2O_5$ | 20 | 18 | - |
| $Al_2O_3$ | - | 172 | 170 |

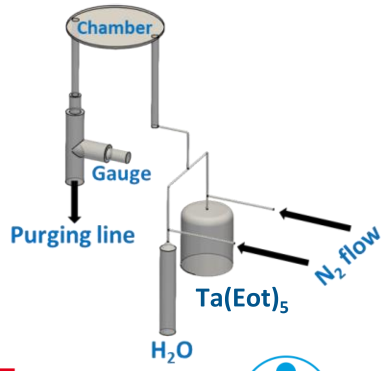
Wenskat, Marc, et al. Superconductor Science and Technology 36.1 (2022): 01501

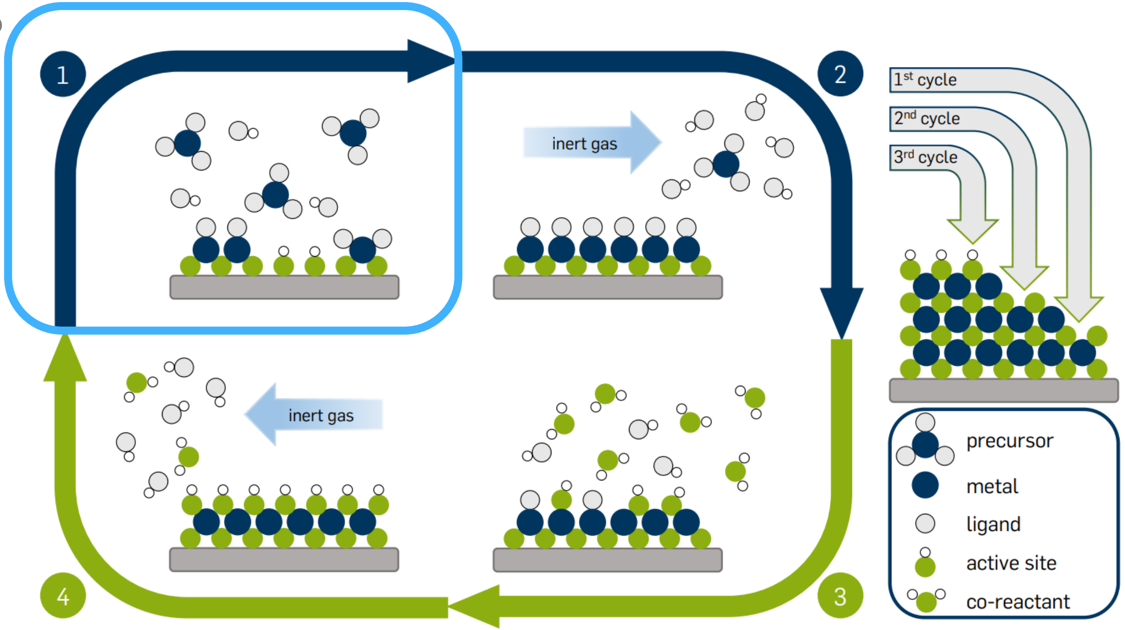
SRF 2023 - Zaidman - WEPWB072

# Thermal ALD basics

## How does one coat a cavity?

Schematic layout of the system




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

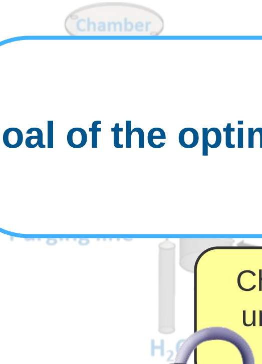
→ **Thermal ALD requires two chemicals, one precursor: Ta(Eot)$_5$, one co-reactant: $H_2O$**

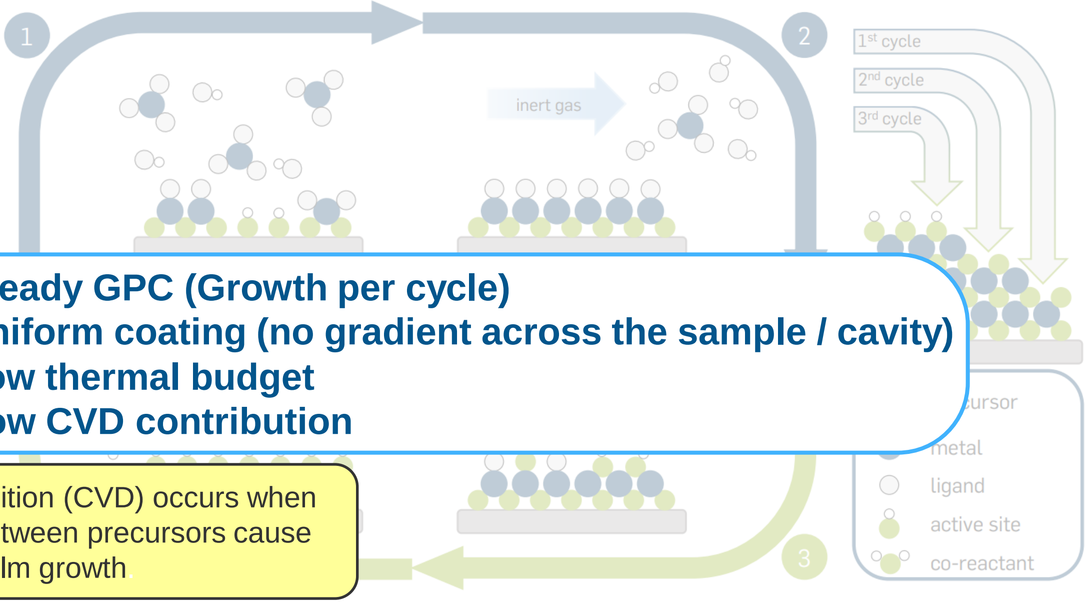
**ALD enables precise and uniform coatings on high aspect ratio substrates**

# Thermal ALD basics

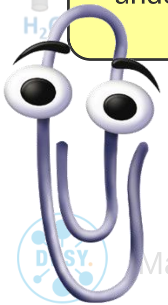
## How does one coat a cavity?

Schematic layout of the system




**Goal of the optimisation** {
- **Steady GPC (Growth per cycle)**
- **Uniform coating (no gradient across the sample / cavity)**
- **Low thermal budget**
- **Low CVD contribution**




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

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

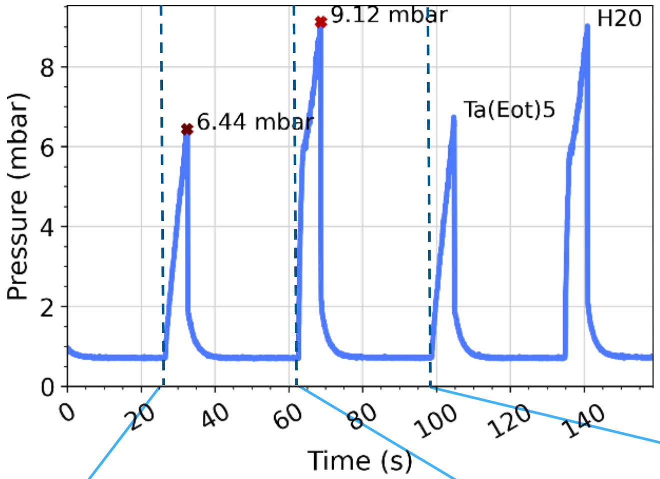


Source:https://www.derstandard.de/consent/tcf/story/2000121096252/clippy-microsofts-virtuelle-bueroklammer-die-nutzer-in-den-wahnsinn-trieb

# Optimisation of the ALD recipe – insufficient deposition

## What have we done so far to use $Ta_2O_5$?

Examplary vacuum profile - 2 Cycles




Process as started

**Not enough deposition / unsteady GPC!**

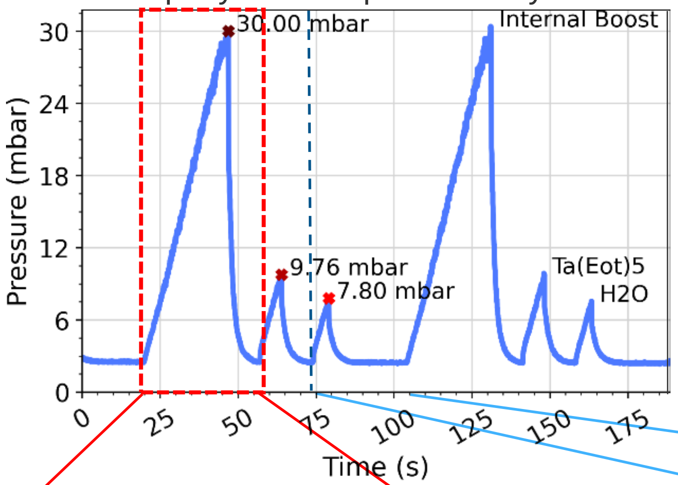
**Problem: vapor pressure of the precursor is too low**

| Ta(Eot)5 | | | | H2O | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| **Pulse (s)** | **Exposure (s)** | **Purge (s)** | **Temp (°C)** | **Pulse(s)** | **Exposure (s)** | **Purge (s)** | **Temp (°C)** | **Process Temp. (°C)** | **N2-Flow (SCCM)** |
| 0.5 | 5 | 20 | 190 | 0.5 | 5 | 20 | 20 | 220 | 20 |

# Optimisation of the ALD recipe – initial boost

What have we done so far to use $Ta_2O_5$?

Examplary vacuum profile - 2 Cycles




Process with initial boost

**Enough deposition / steady GPC!**

**Internal boost corrects low vapor pressure of the precursor**

| Internal Boost | | | | Ta(Eot)5 | | | | H2O | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Purge (s) | Exposure (s) | Pulse (s) | Purge(s) | Pulse (s) | Exposure (s) | Purge (s) | Temp (°C) | Pulse(s) | Exposure (s) | Purge (s) | Temp (°C) | Process Temp. (°C) | N2-Flow (SCCM) |
| 5 | 25 | 2 | 10 | 2 | 5 | 10 | 190 | 0.1 | 5 | 20 | 20 | 220 | 20 |

# Optimisation of the ALD recipe – parameter variation

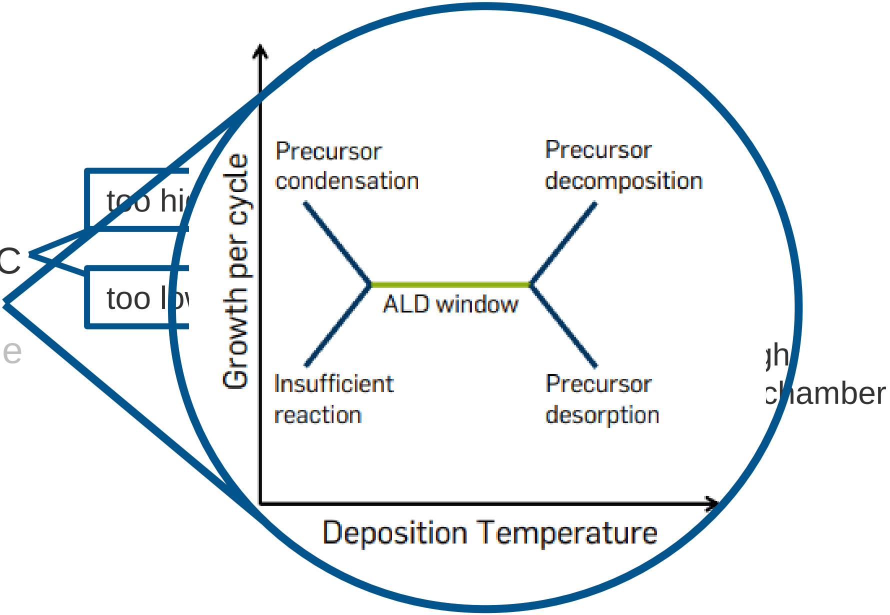
What are we currently working on to use $Ta_2O_5$?

To obtain the final parameters, the individual parameters must be varied in measurement series

→ **Find the ALD window**

**Which parameters are to be varied:**

- Ta precursor temperature ~150-190°C
- Deposition temperature ~ 170-220°C
- Water pulse, exposure and purge time
- Ta pulse, exposure and purge time
- Initial boost exposure time
- N2 flow rate

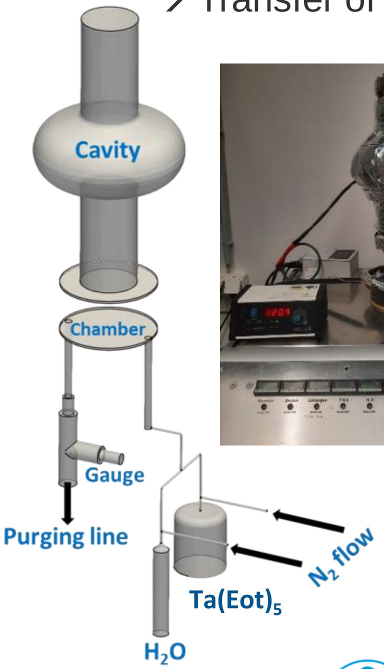


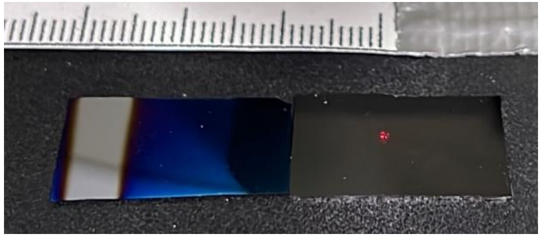

L. Mai, (2020), DOI:10.13154/294-7658.

# Optimisation of the ALD recipe – GPC study

## What are we currently working on to use $Ta_2O_5$?

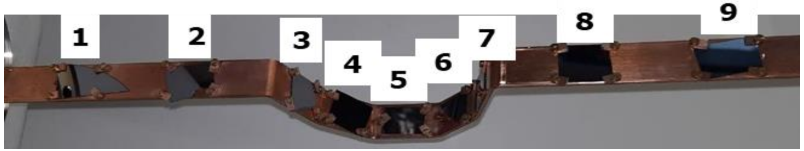
→ Transfer of the recipe from planar samples to 2D/ 3D cavity structure



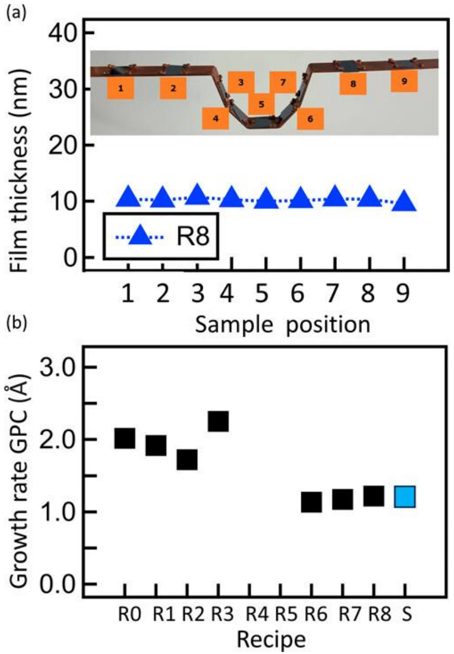



**Silicon sample after coating with $Ta_2O_5$**

**Silicon sample before coating**




Deyu, Getnet, et. al, Chem. Mater. 2024, 36, 6 , 2846-2856




GPC for various coatings already done with $Al_2O_3$ in the dummy cavity

# Optimisation of the ALD recipe – current recipe

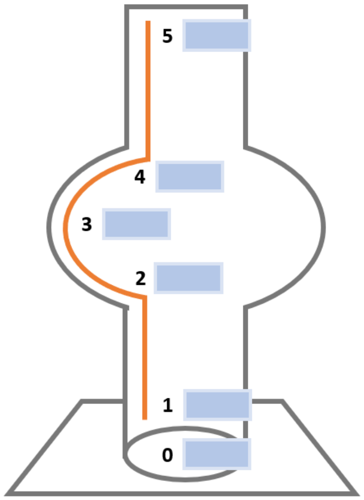
## What are we currently working on to use $Ta_2O_5$?

| Internal Boost | | | | $Ta(Eot)_5$ | | | H2O | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| Purge (s) | Exposure (s) | Pulse (s) | Purge(s) | Pulse (s) | Exposure (s) | Purge (s) | Pulse(s) | Exposure (s) | Purge (s) | N2-Flow (SCCM) |
| 5 | 30 | 2 | 10 | 2 | 5 | 10 | 0.1 | 5 | 20 | 20 |

| Temperatures (°C) | | | | | |
|---|---|---|---|---|---|
| $Ta(Eot)_5$ | $H_2O$ | Chamber | Lid/Cavity | Valves | Exhaust |
| 190 | 20 | 200 | 200 | 160 | 120 |

**Simple chamber for plain Si-Samples**
1000 Cycles
→ GPC 0.3 Å/Cycle
→ CVD share <25% (0.07 Å/Cycle)

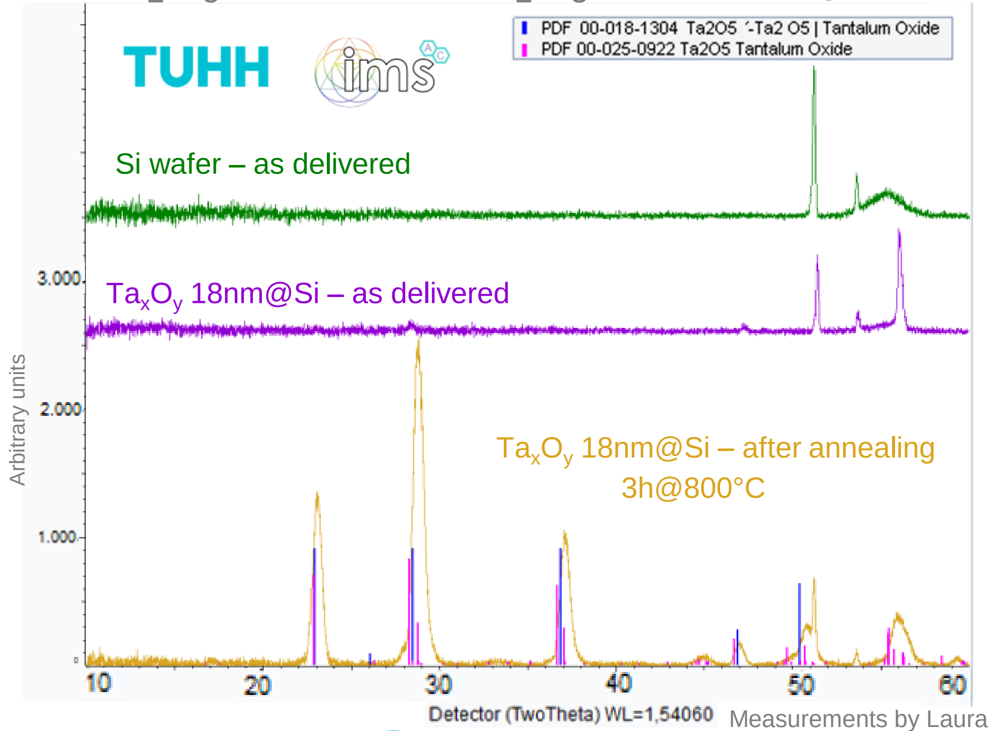
**Dummy Cavity for Si-Samples**
→ GPC
→ CVD share



Measured thicknesses across the dummy cavity

# $Ta_2O_5$ instead of $Al_2O_3$ – crystallisation

$Ta_2O_5$ better than $Al_2O_3$ the industry standard?




Measurements by Laura Maragno (TUHH)

$Ta_2O_5$ crystallizes at 800°C – $Al_2O_3$ needs >1000°C

→ Nb annealing at 800°C is a standard recipe

→ better order means less TLS

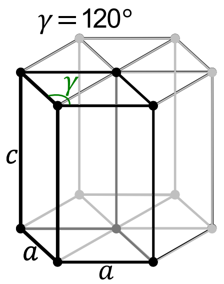
$Nb_2O_5$ dissolves above 250°C

→ $Ta_2O_5$ passivates Nb after annealing

→ $Ta_2O_5$ less prone to TLS then $Nb_2O_5$

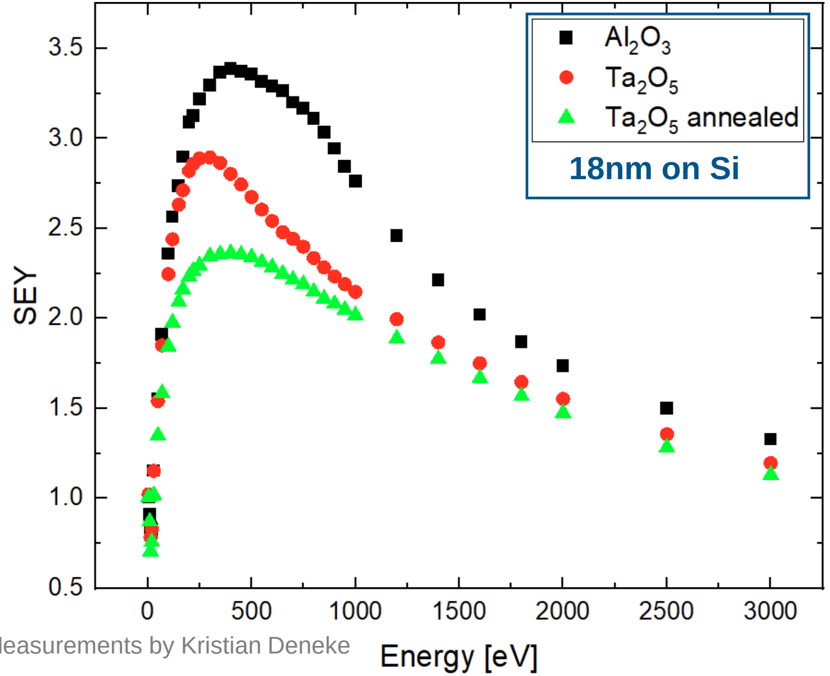
After annealing 3 main peaks
→ 23°, 29°, 37° that match with $Ta_2O_5$

Patterns:
00-018-1304 – hexagonal
00-025-0922 – orthorhombic



# $Ta_2O_5$ instead of $Al_2O_3$ – field emission

## Is $Ta_2O_5$ better than the industry standard?




Measurements by Kristian Deneke

$Al_2O_3$ prone to field emission due to higher secondary electron yield (SEY)

→ Measurements of our layers show a significant reduction of SEY (Nb = 2.2)

# Future activites

What needs to be done in order to use $Ta_2O_5$ to coat cavities?

**-Coat a cavity**

## -HPR Test (High pressure rinsing)

→ Can the coatings withstand the HPR?

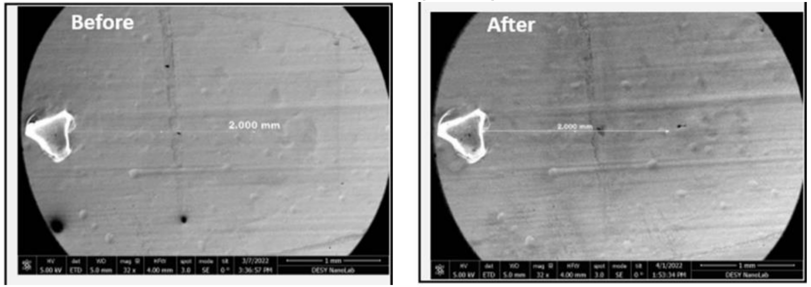
## -Morphology Test

How does the crystallinity of $Ta_2O_5$ change after annealing at different temperatures?

## -Passivation Test

How well does $Ta_2O_5$ passivate the inner surface of a cavity?

Wenskat, Marc, et al. (2022), DOI:10.1088




Examplary SEM images of a $Al_2O_3$-coated Nb-Sample before and after HPR

# Summary

- $Ta_2O_5$ as potential material for improving SRF cavity performance beyond the niobium limit.
- $Ta_2O_5$ provides effective passivation, reducing Two-Level Systems (TLS)
- Ongoing process optimization to achieve uniform coating with optimal Layer Growth
- Further testing to confirm $Ta_2O_5$'s suitability for coating a single-cell cavity

***$Ta_2O_5$ shows strong potential as a coating material to enhance SRF cavity performance by reducing losses and enabling superior surface passivation through optimized ALD processes.***

Contact: marco.voige@desy.de

# *Backup-Slides*

## Atomic layer deposition of tantalum oxide

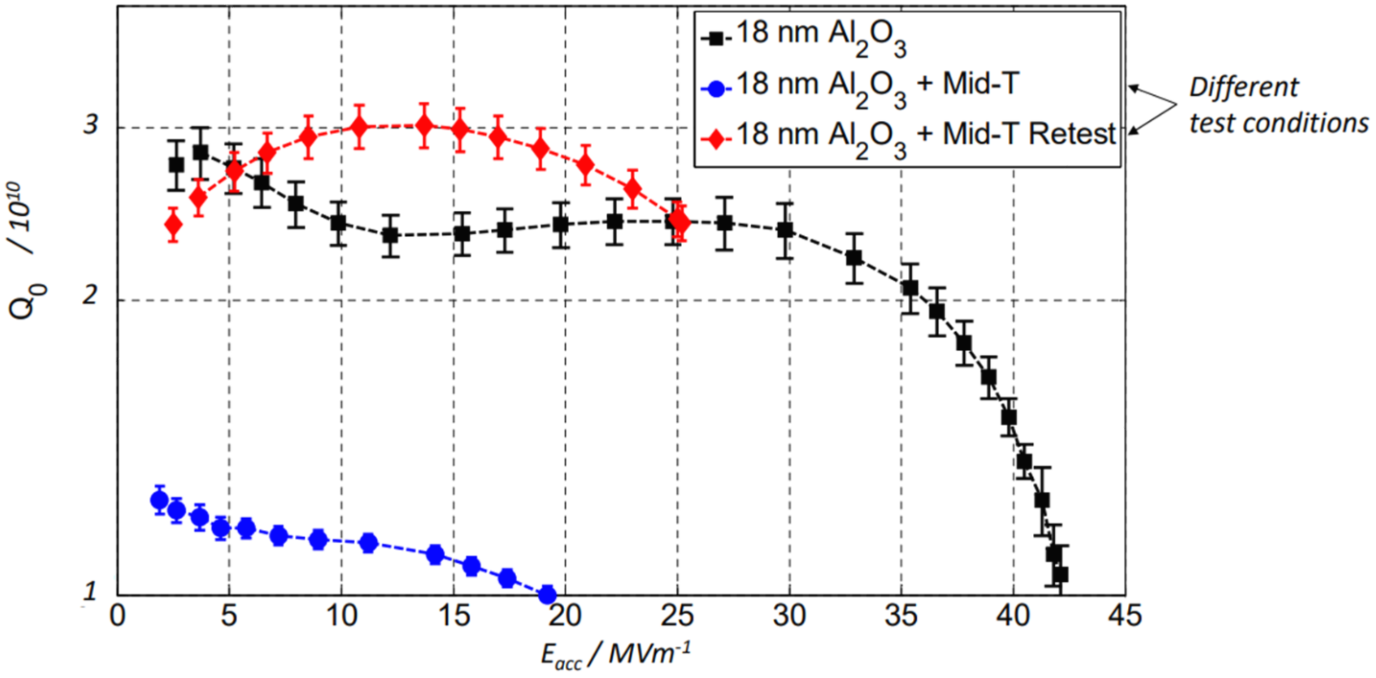
### a new material for coating cavities

19.09.2024

Marco Voige - on behalf of the SRF R&D Team Hamburg

# Mid-T heat treatment of a coated cavity

Wenskat - TUIBA02



# Reduced $R_{BCS}$ beyond regular mid-T cavities

Wenskat - TUIBA02
Bate - MOPMB022
Ghanbari - MOPMB021

- $R_{BCS}(2K) \approx R_S(2K) - \underbrace{R_S(1.5K)}_{\approx R_{res}}$
- *$R_{BCS}$ smaller than for other mid-T cavities*

  **3nΩ @ 2K**
- *$R_{res}$ is higher due to mid-T*

  *before: 3nΩ*
  *after: 6nΩ*
- *Why is $R_{res}$ higher?*
  - *sensitivity higher for mid-T cavities → increased $R_{flux}$ ?*
  - *agrees with low $Q_0$ in first test after mid-T*
  - *120°C before mid-T may affect $R_{res}$ negatively*