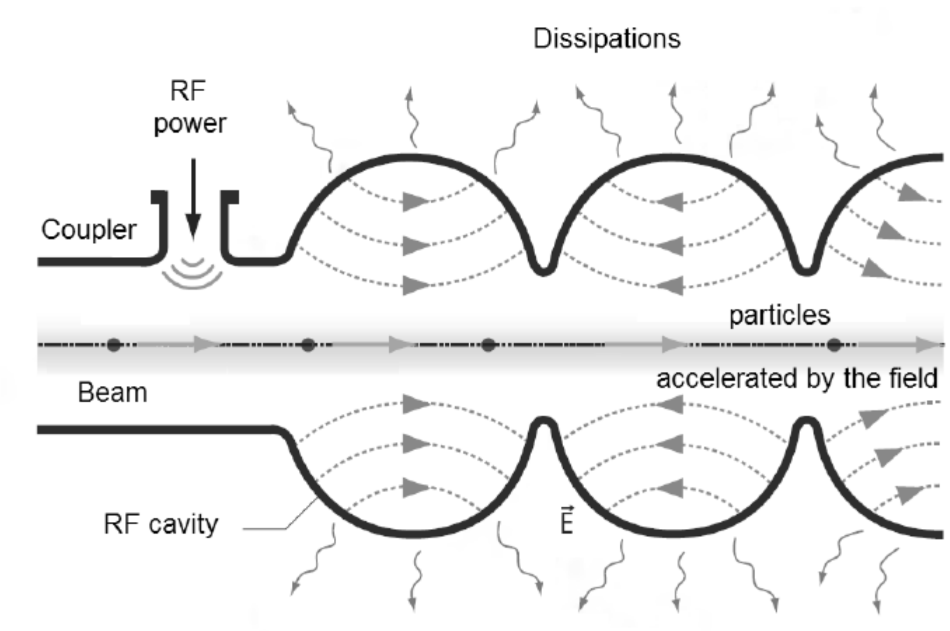
Beyond Niobium research and challenging the BCS Theory

What's next in SRF?

Marc Wenskat – on behalf of our joint SRF R&D Team
01.10.2021

Reminder and motivation

Superconducting Radiofrequency Cavities



$$E_{acc} = \frac{1}{l} \int_0^l V_{acc}\, dz$$

Higher gradient means shorter accelerator (less construction cost) or higher collision energy (for same cost)

Higher Q means reduced operational cost due to lower cooling power needed

$$Q_0 \propto \frac{1}{P_{diss}} \propto \frac{1}{R_{BCS} + R_{res}}$$

- Quality factor Q_0 as figure of merit for the loss
- Non-vanishing RF resistance

  R_{BCS} ~ 8 nΩ at 2K and 1.3 GHz, Nb

  R_{res} ~ 4-6 nΩ

→ $Q_{0,max}$ ~ 1×10^{11}

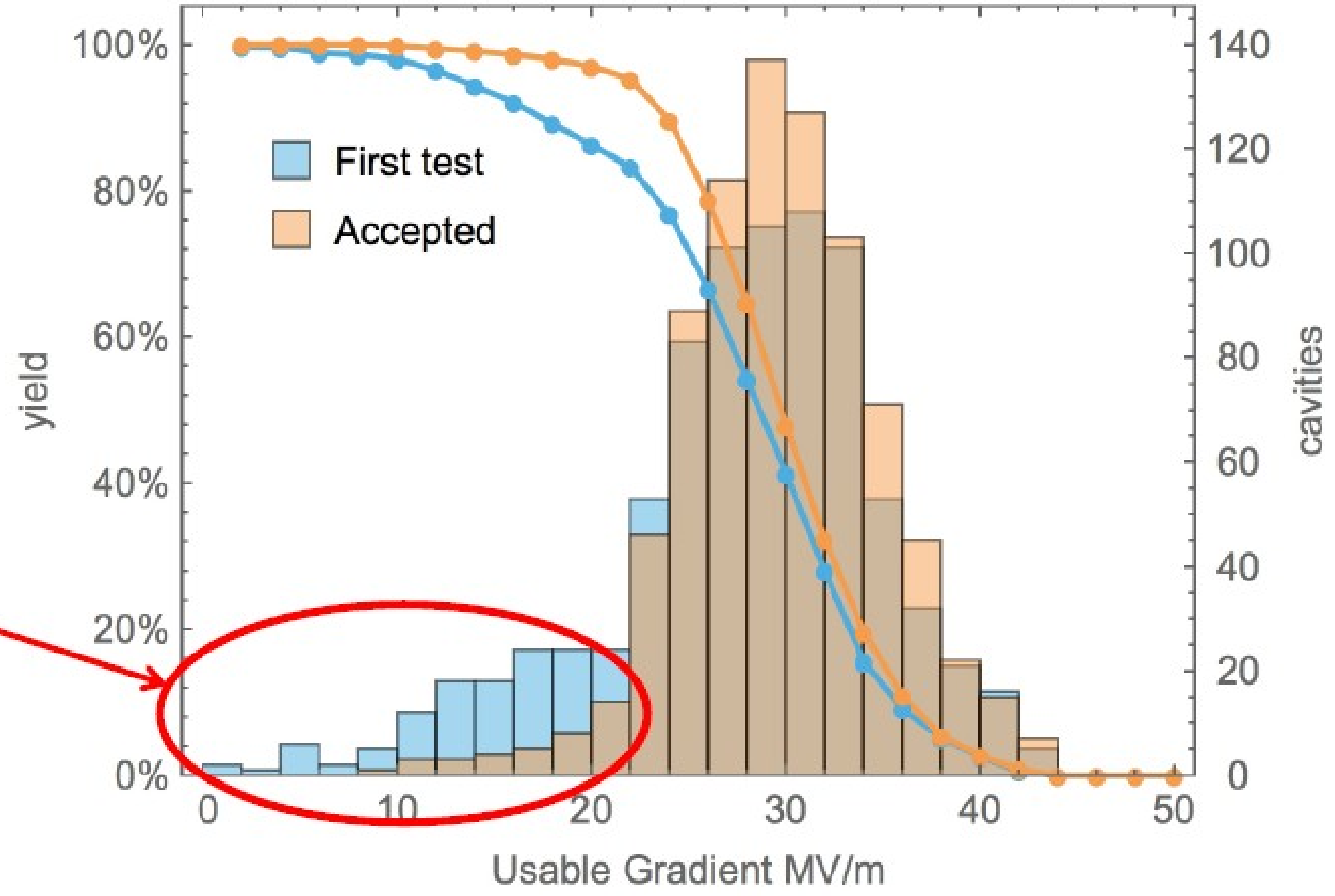
- critical magnetic field H_c: 230 mT → phase transition

→ $E_{acc,max}$ = 55 MV/m

European XFEL Fabrication Results

Final Performance (sent for module assembly)

- $\langle E_{\mathrm{usable}} \rangle = 29.8 \pm 5.1$ MV/m



impact of retreatment

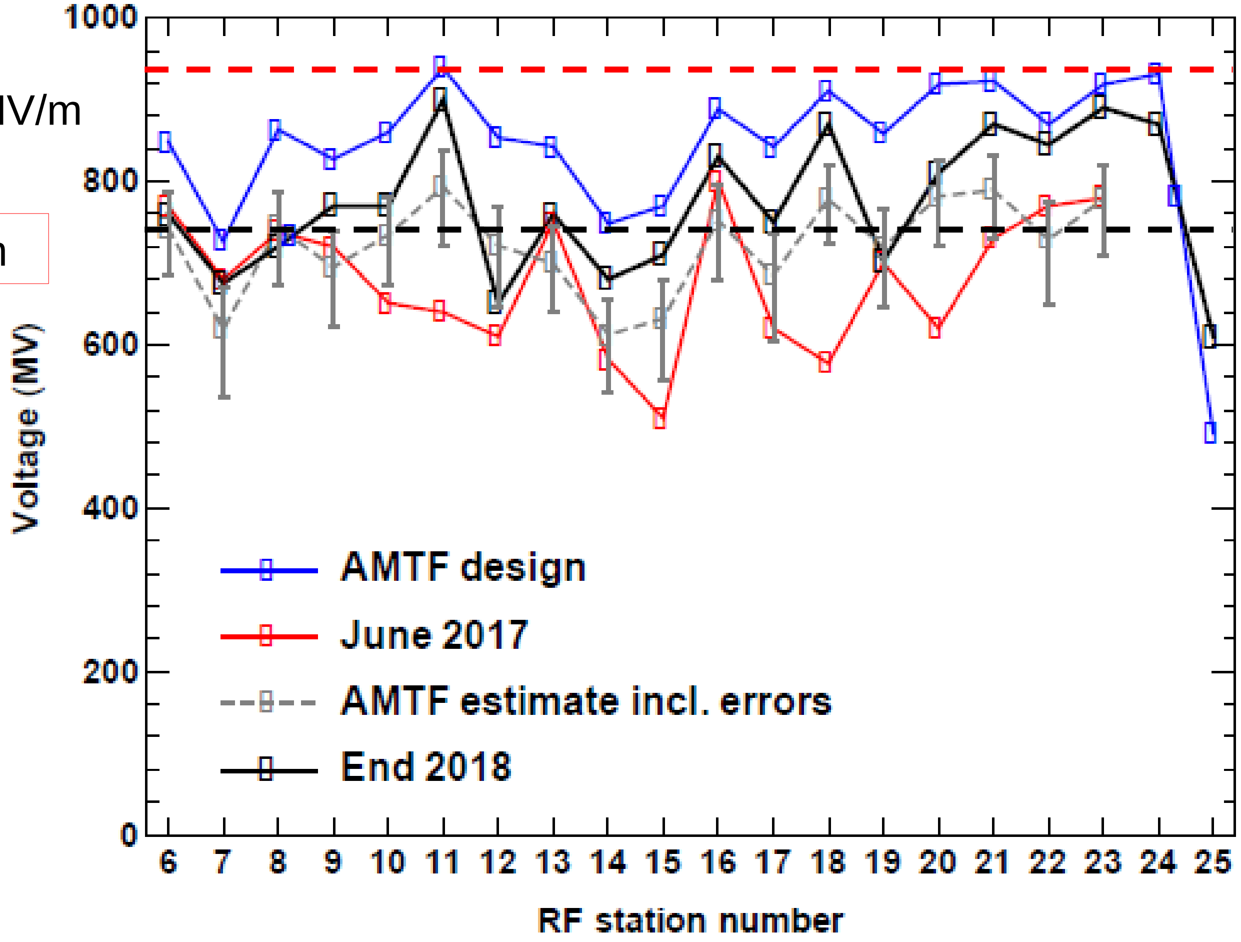
Operation of European XFEL

Incl. Installation in module, impact of waveguide system

RF Performance as of End of 2018

If all cavities in the RF station operate at 29 MV/m

Avg. Gradient is 23 MV/m



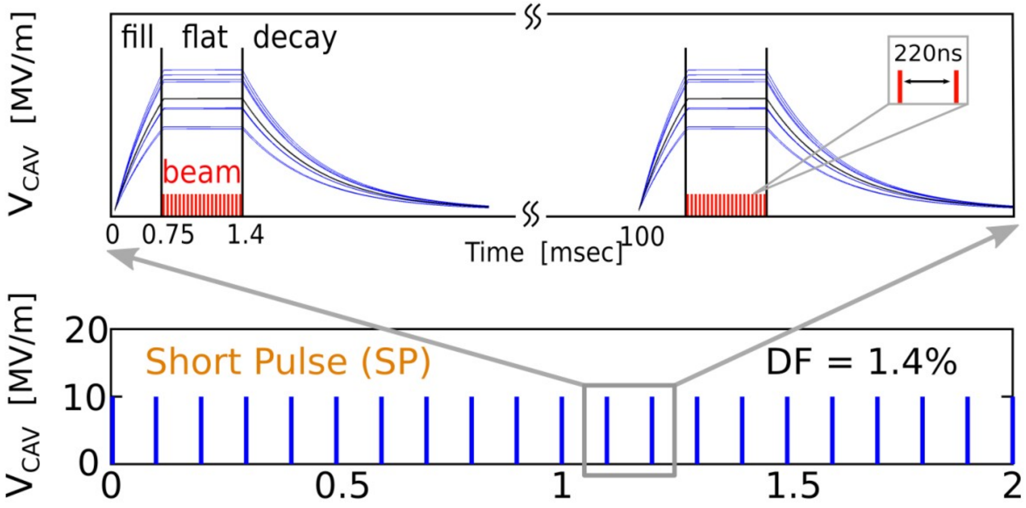
🟧 Reached an average of 93.6% of AMTF performance

European XFEL CW upgrade

Motivation

Benefits of Continuous Wave (CW) operation

- Flexible beam patterns for detectors
  Almost any macro pulse structure can be offered
- Slower repetition rate lasers
- Fill-transients no longer an issue



E = 17.5 GeV

N_{bunch}/s = 27k

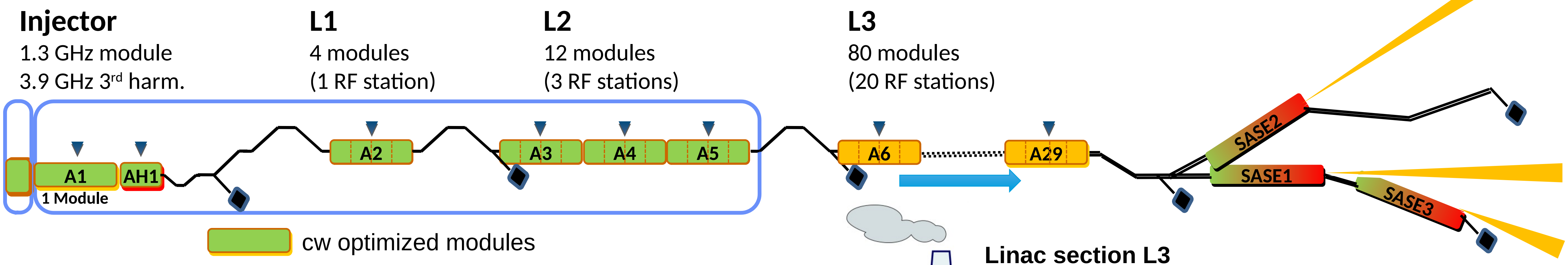
Benefits of Long Pulse (LP) operation

- Still high duty factor (DF = 10-50%)
- Higher gradients than CW with same heat load

Why?

- Users: less complex detectors
- *“Better 100 kHz bunches all the time rather than 4.5 MHz bursts with 100 msec gaps”*

European XFEL CW upgrade



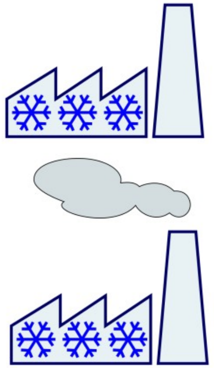
Injector
1.3 GHz module
3.9 GHz 3^{rd} harm.

L1
4 modules
(1 RF station)

L2
12 modules
(3 RF stations)

L3
80 modules
(20 RF stations)

🟩 cw optimized modules



1 – Replace the front-end cryomodules (17x)

- Larger cooling capability
- CW cavities

2 – Install CW capable RF sources

- 1x IOT per RF station

3 – Double the cryo plant (cost driver)

- 2.5 → 5kW

4 – Install CW capable gun:

- RF gun upgrade

Linac section L3

- operated at moderate CW gradients
- lengthened by former A2 … A5
  80 + 12 modules
  24 RF stations

Expected energy 8 GeV !

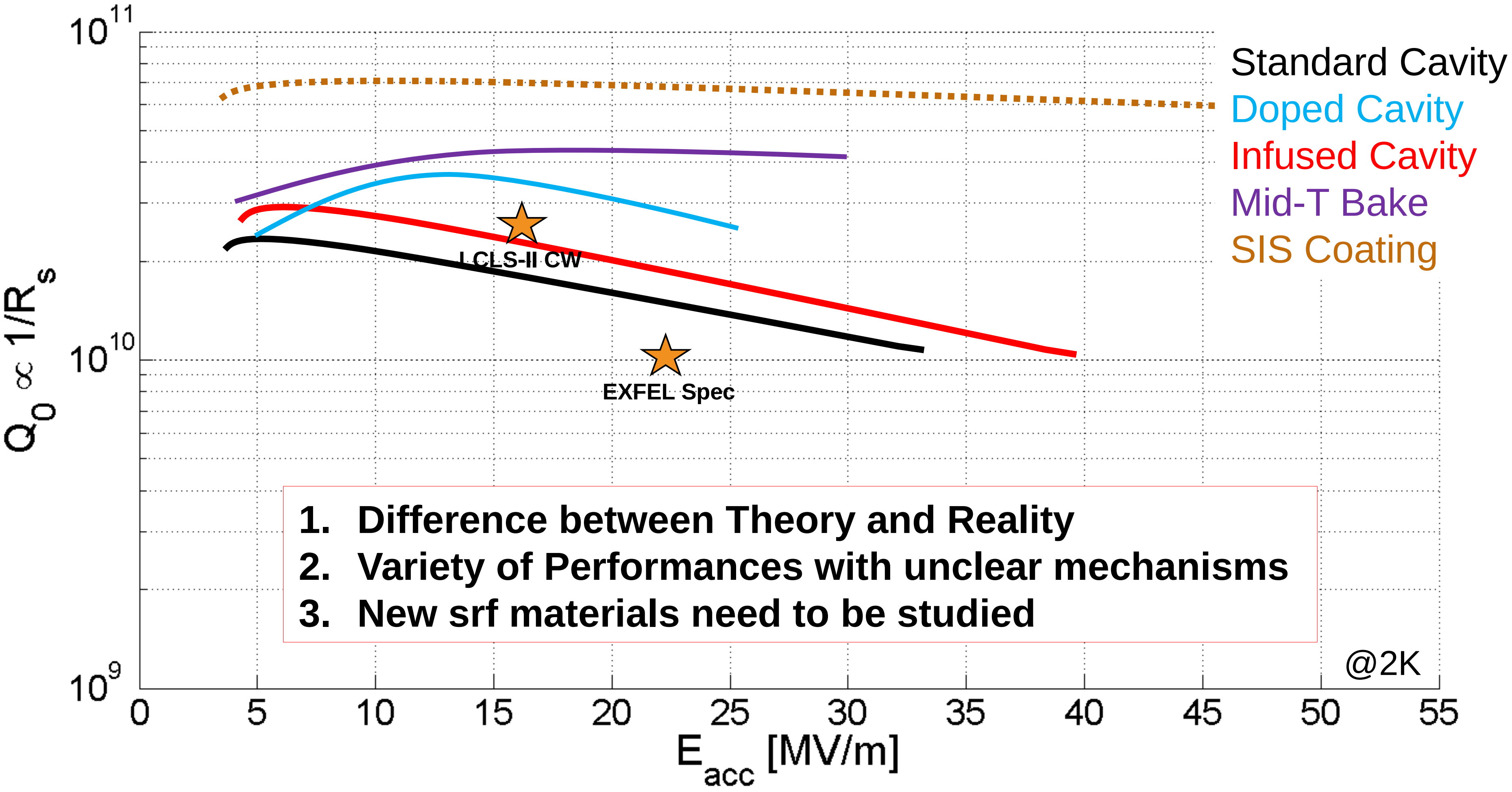
- **The former front-end cryomodules can be installed at the end of the linac to lengthen L3 (+4 RF stations)**

- **No further action required in L3 (>1km)**
- **The upgraded XFEL would be capable of short pulse long pulse AND continuous wave operation**

How to achieve CW operation from a cavity POV?

What could we do?

Project driven R&D and fundamental research



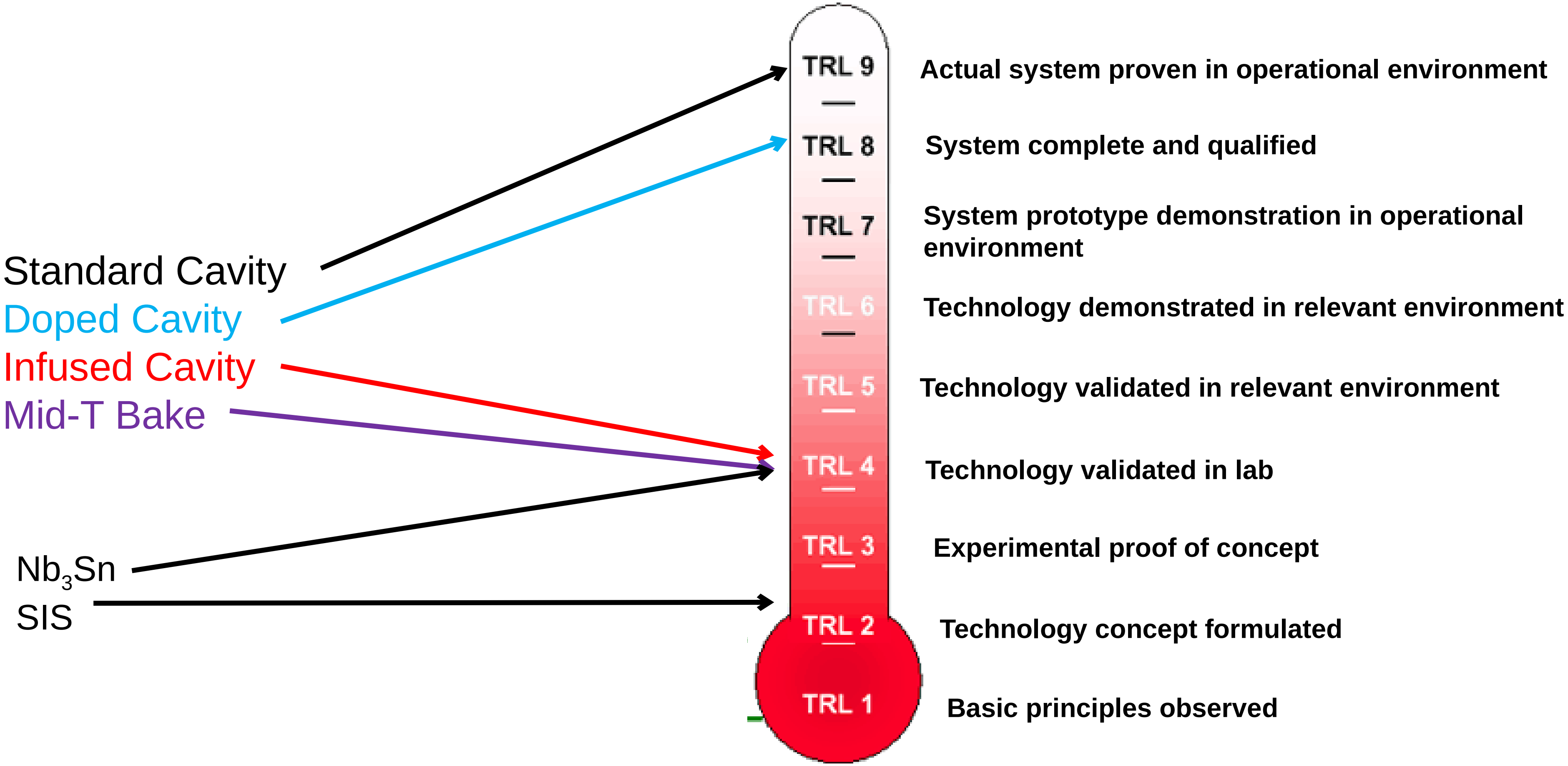
1. **Difference between Theory and Reality**
2. **Variety of Performances with unclear mechanisms**
3. **New srf materials need to be studied**

<u>Cavities limited by quench</u>

[Reschke et al., Phys. Rev. Accel. Beams, 20, 042004 (2017)]
[Grassellino et al., SUST, 26, 102001 (2013)]
[Grassellino et al., SUST, 30, 094004 (2017)]
[Posen et al., Phys. Rev. Applied 13, 014024]
[Gurevich, APL, 88, 012511 (2006)]

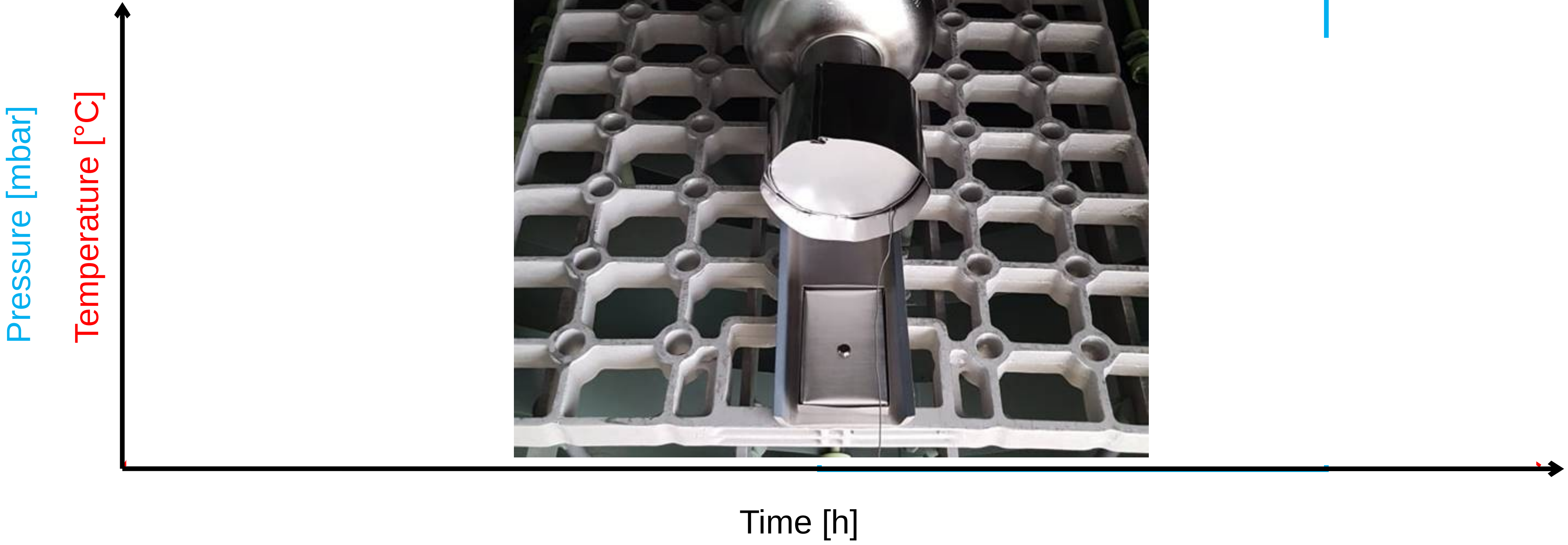
Technology Readiness Level

Developed by NASA in 1970



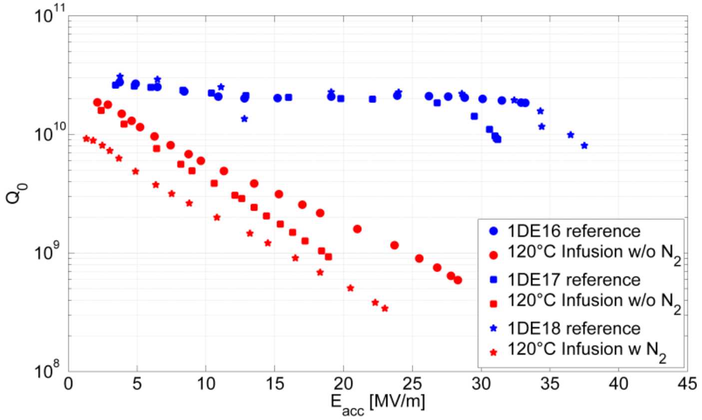
Nitrogen Infusion

The Recipe

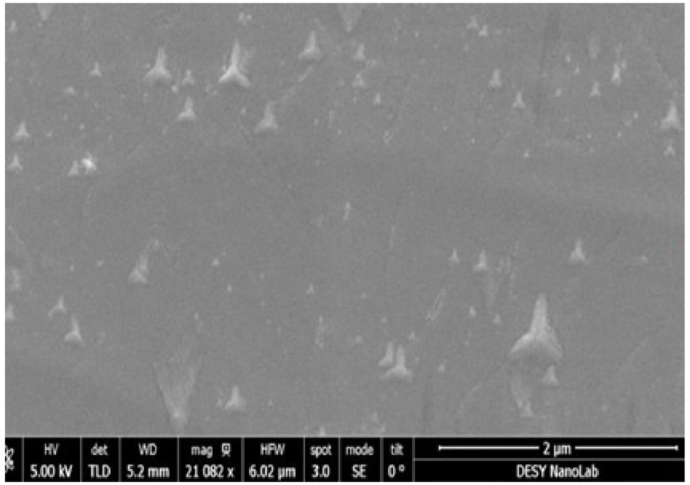


Problem: No one cooks like Grandma

First infusion runs failed drastically



	1DE18	1DE17	1DE16
Material	Ningxia fine grain	Ningxia fine grain	Plansee fine grain
Reference @ 2K			
$E_{acc,max}[\frac{MV}{m}]$	37.7 - BD	31.2 - BD	32.2 - BD
$Q_0(4\,\mathrm{MV/m})[\times 10^{10}]$	2.8	2.5	2.7
Baking Parameters			
p @ 800°C [mbar]	2×10^{-5}	1.1×10^{-5}	5.5×10^{-6}
P_{N_2} @ 120°C[mbar]	$7 - 300$ $\times 10^{-5}$	w/o	w/o
RF Test @ 2K			
$E_{acc,max}[\frac{MV}{m}]$	20.2 no FE	19.5 no FE	26.3 - BD no FE
$Q_0(4\,\mathrm{MV/m})[\times 10^{10}]$	0.5	1.2	3.2

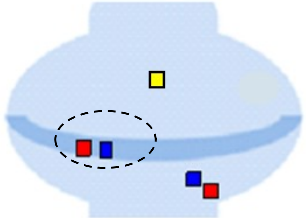


RGA during 800°C bake showed high mass contributions (Hydrocarbons)

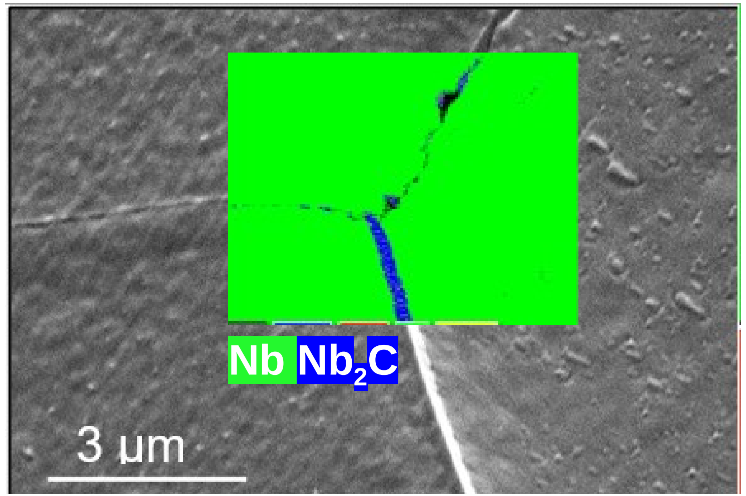
Samples within a standard 800°C bake showed precipitates as well

Grain boundary segregation: Nb grain decoupling

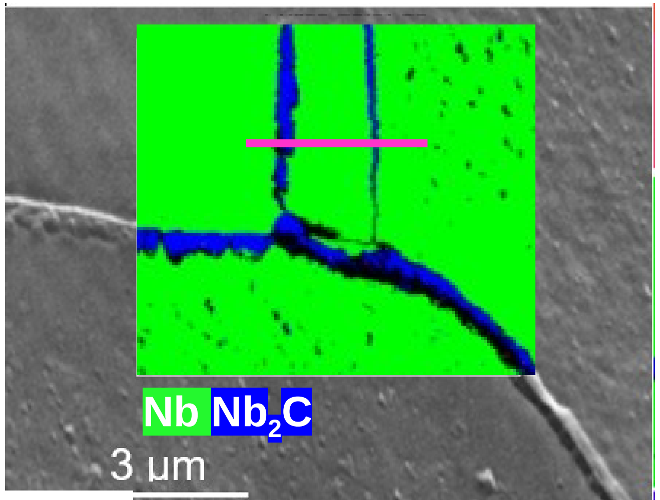
Cut a cavity for facts



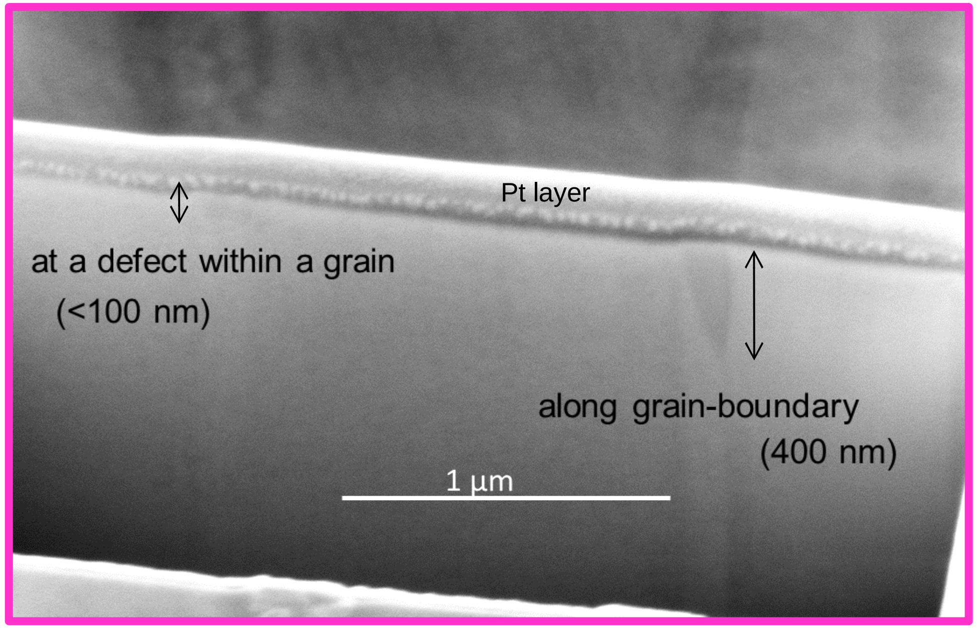
cold spots

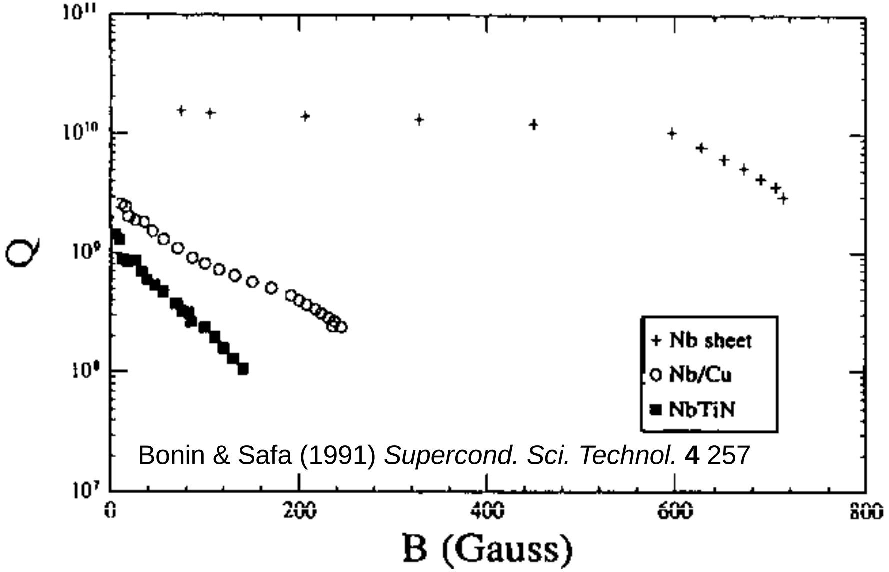


Quench-spot



FIB Cross-section view





Bonin & Safa (1991) *Supercond. Sci. Technol.* **4** 257

Depth of carbides into Nb >> RF penetration depth

⇒ **Decouples the Nb grains** within RF layer

⇒ **Enormous Q-degradation** as seen in granular thin films on cavities/ Ti contaminated Nb cavities

ANTOINE C, et al., Proceedings of the 1997 Workshop on RF Superconductivity, Abano Terme (Padova), Italy

A. Grassellino et al., arXiv (2013), arXiv:1305.2182

Correlation $p_{furnace}$ and performance

What about the caps?

- Furnace pressure ≠ Cavity Pressure
- Simulations showed an increase inside by a factor of 8-13
- Assume the pollution is coming from outside the cavity, the caps protect the inside
- Hence $p_{Cavity} = p_{furnace} / R_{caps}$
- Assumption: Time does not play a role. It does in reality

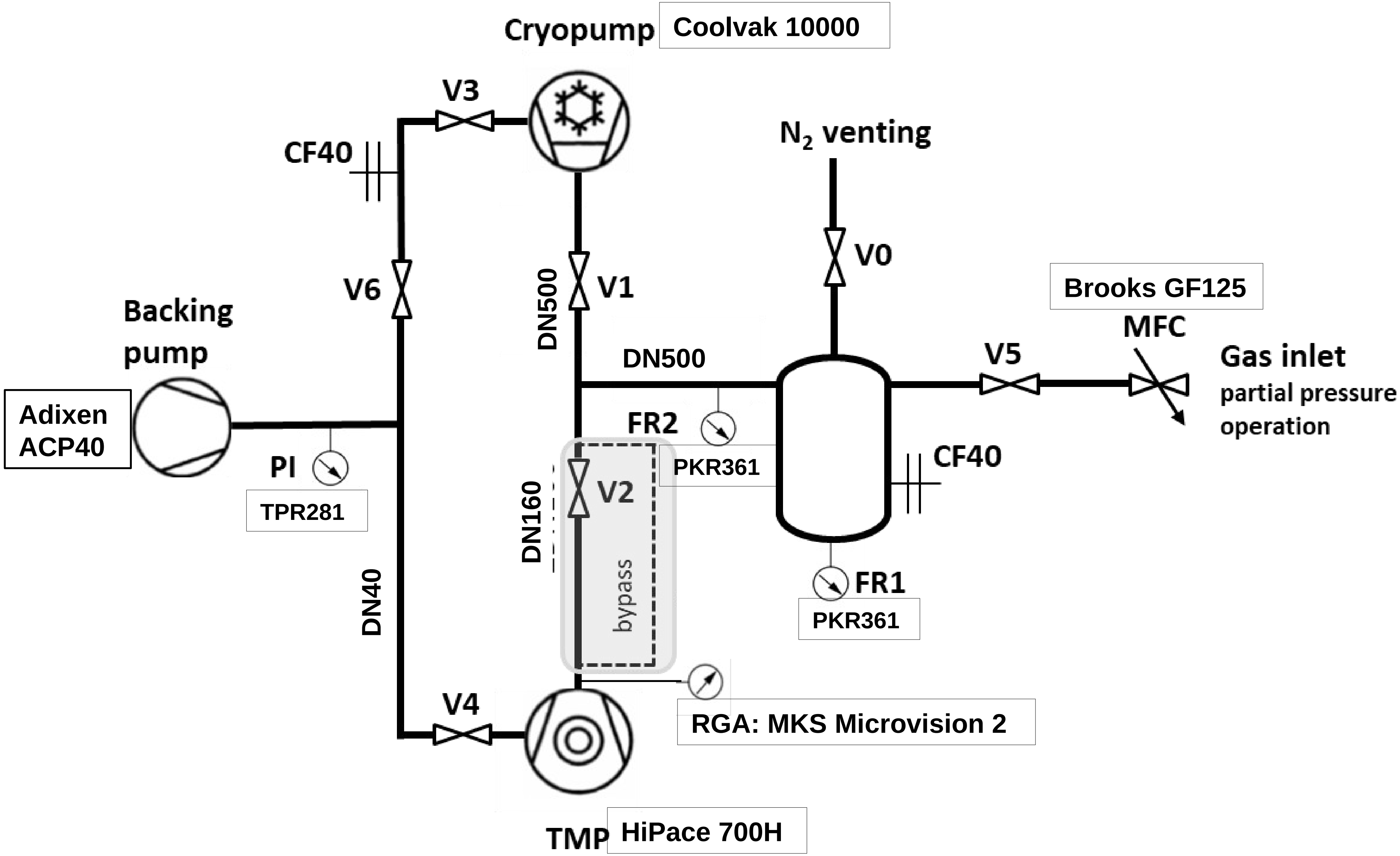
Correlation (Caps + CO) to Performance

New (U)HV Single-Cell Furnace

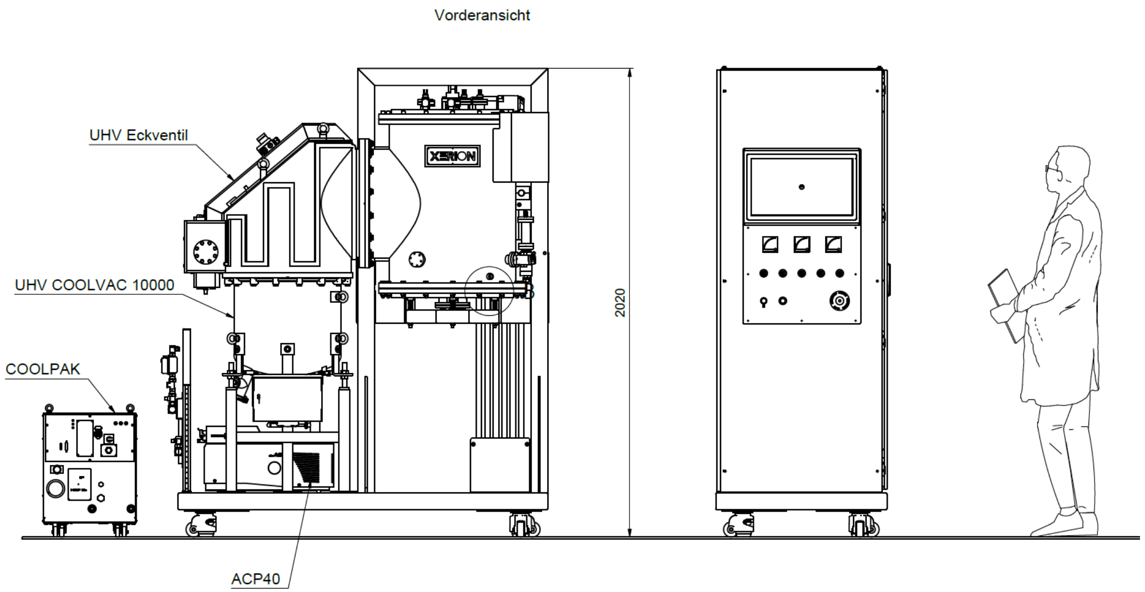
Starting Assumptions and Decisions

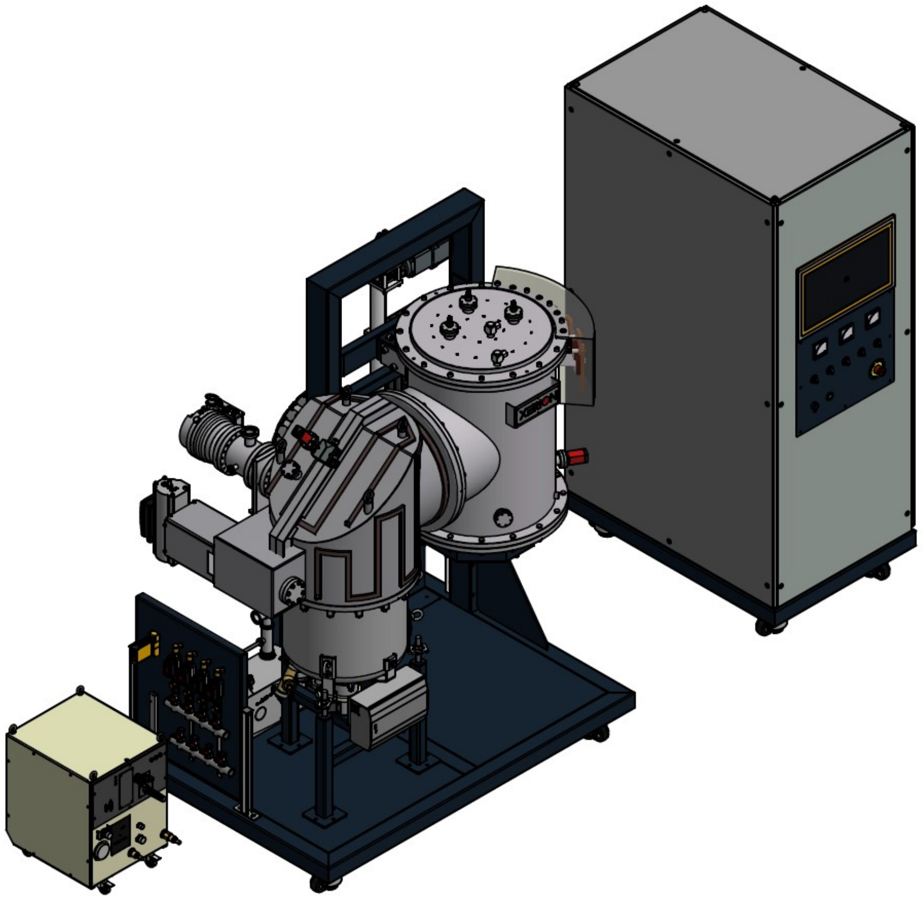
- Carbon and hydrocarbon-free (choice of pumps)
- High pumping speed for hydrogen, base pressure 10^{-8} mbar or better
- Bottled nitrogen to guarantee 6.0 (choice of vents/MFC/connections/lines)
- Stabilized pressure operation by programmable mass-flow controller and flow equilibrium
- RGA during partial pressure operation possible
- No backflow from roughing pumps / vacuum connection when switching
- Moly-Heater & all-metal sealed (except door)
- $T_{max} = 1100°C$
  - T-Stability: ±1K in time and ±5K at dwell time
  - >3 thermocouples to measure T of load
  - >3 thermocouples to measure T of furnace (used for control)
- Only single-cell cavities & QPR samples

Simplified Layout



Mechanical Layout





Placed in ISO5 cleanroom

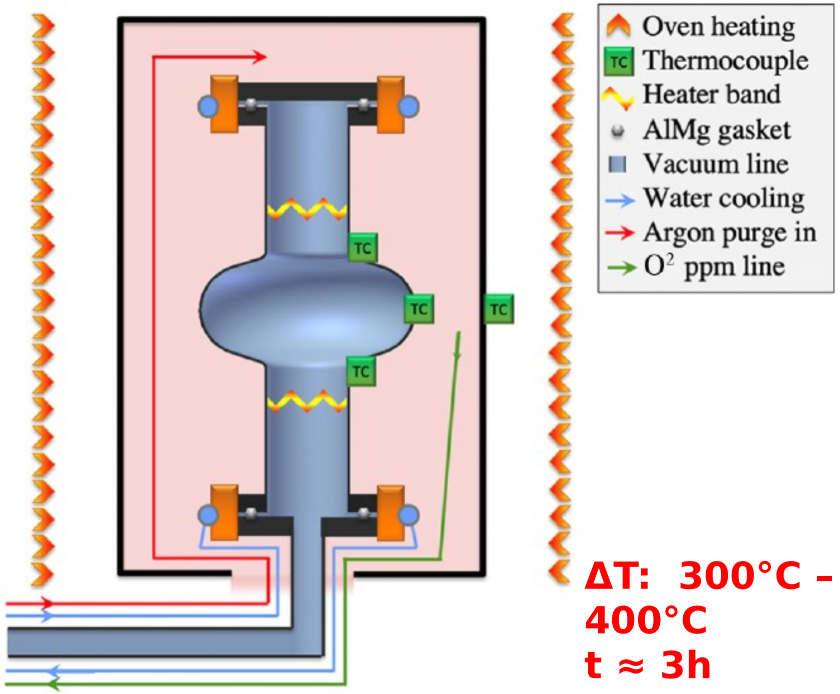
Delivery: 6.-10.12.2021

Mid-T Bake

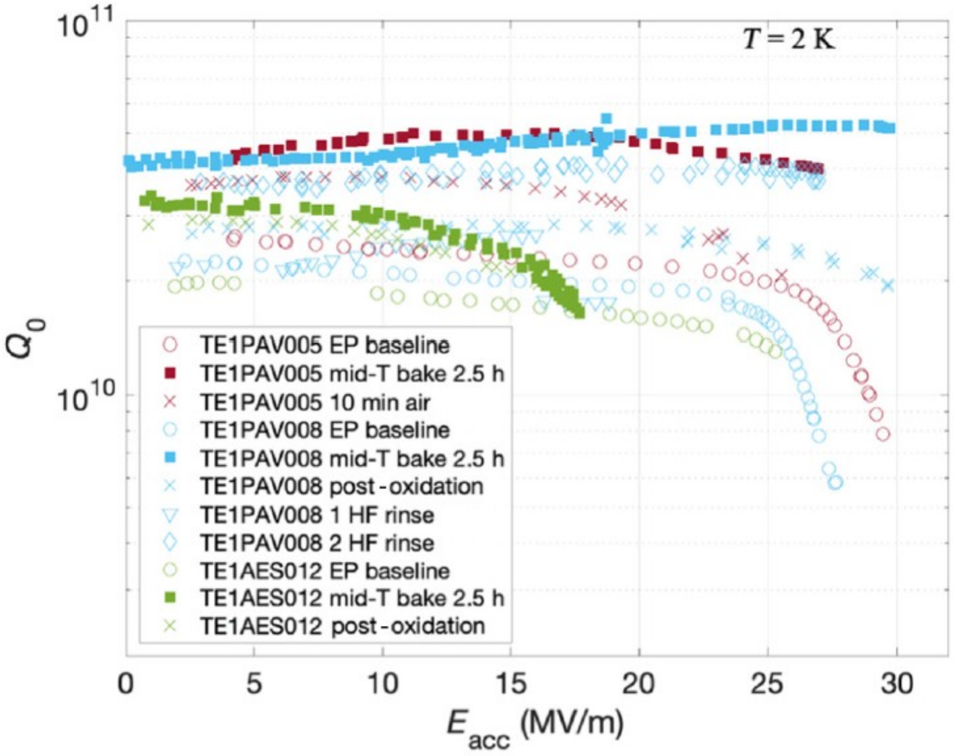
Mid-T bake

A new recipe which leads to high Q at high gradient

- In-situ studies by FNAL **[Posen, S., et al. *Phys. Rev. Appl.* 13.1 (2020): 014024.]**



ΔT: 300°C – 400°C
t ≈ 3h
p ≤ 10^{-6} mbar



$R_{BCS} \leq 6$ nΩ
$dQ_0/dE_{acc} > 0$
$R_{res} \leq 2$ nΩ

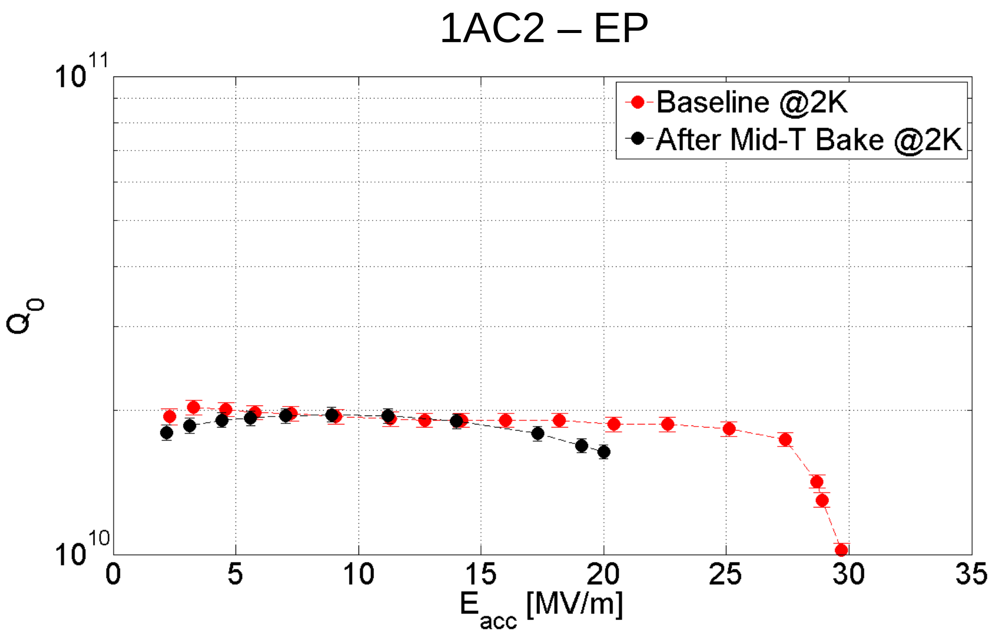
- IHEP and KEK reproduced results with modified recipes (in furnaces & with caps)

[Zhou, Q., et al. *Radiat. Detect. Technol. Methods* 4.4 (2020): 507-512.]

[Ito, H., et al. *Prog. Theor. Exp. Phys. 2015* (2021).]

Vertical test results 1st run

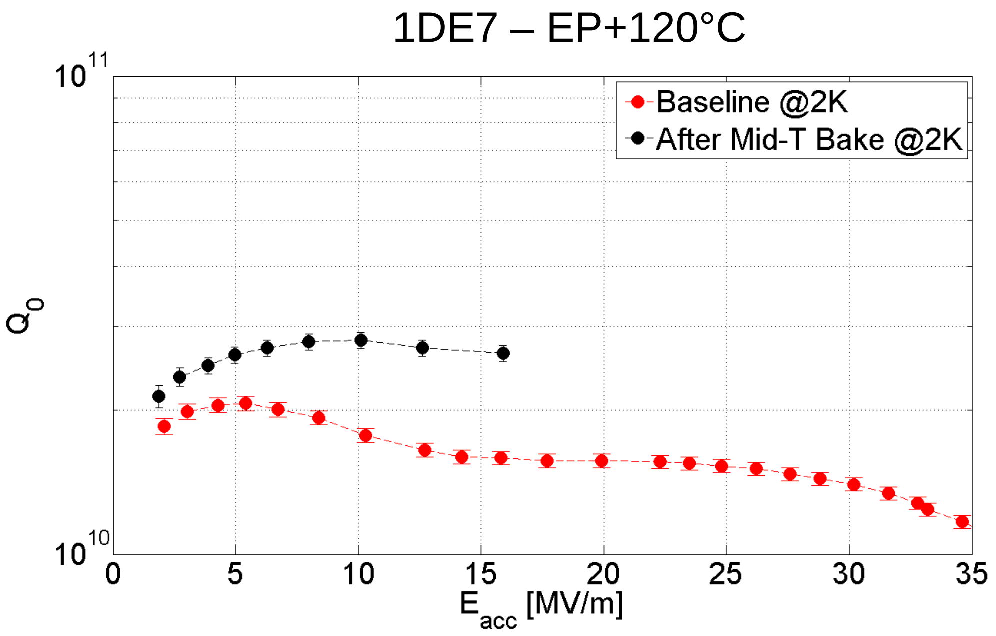
Mid-T Bake done in industry!


1AC2 – EP

From Q vs. T:

R_{res} before Mid T = 1.9 nΩ

R_{res} after Mid T = 8.5 nΩ


1DE7 – EP+120°C

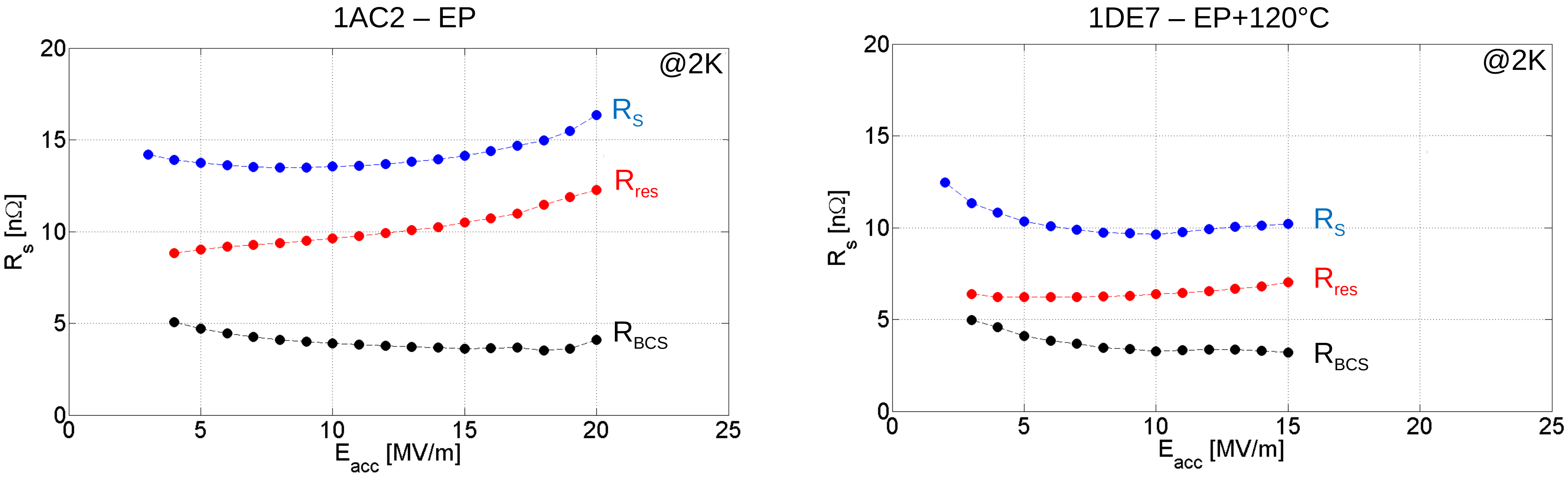
From Q vs. T:

R_{res} before Mid T = 5.5 nΩ

R_{res} after Mid T = 5.1 nΩ

Decomposition of the surface resistance

Glass is half full!



R_{BCS} behaves as expected:

- $R_{BCS} \leq 6$ nΩ
- $dQ_0/dE_{acc} > 0$

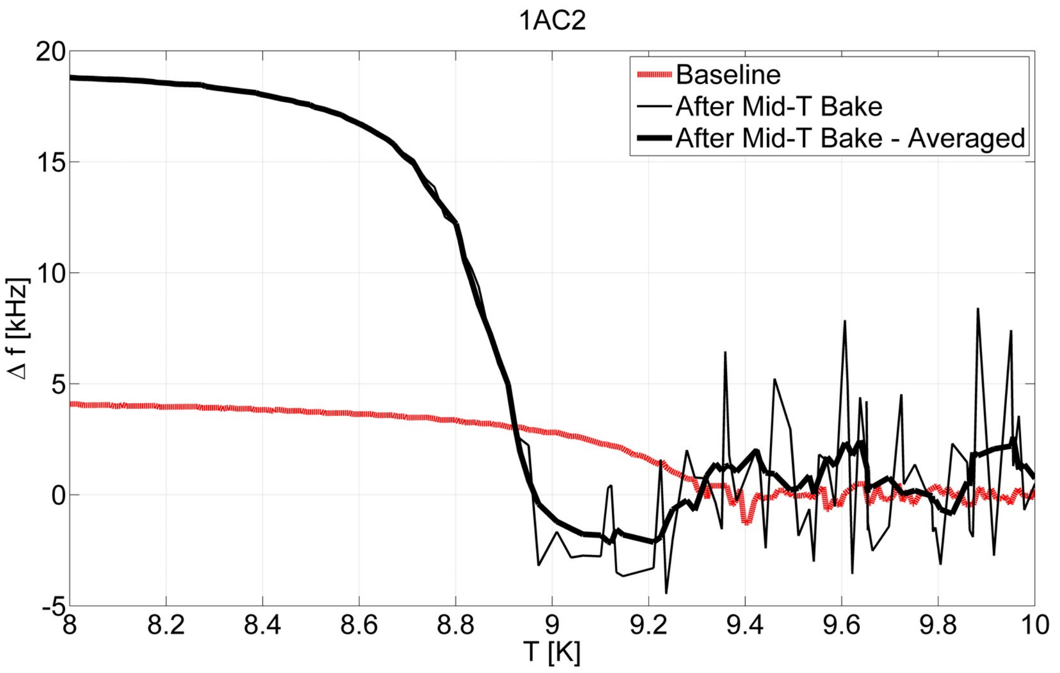
but R_{res} doesn‘t

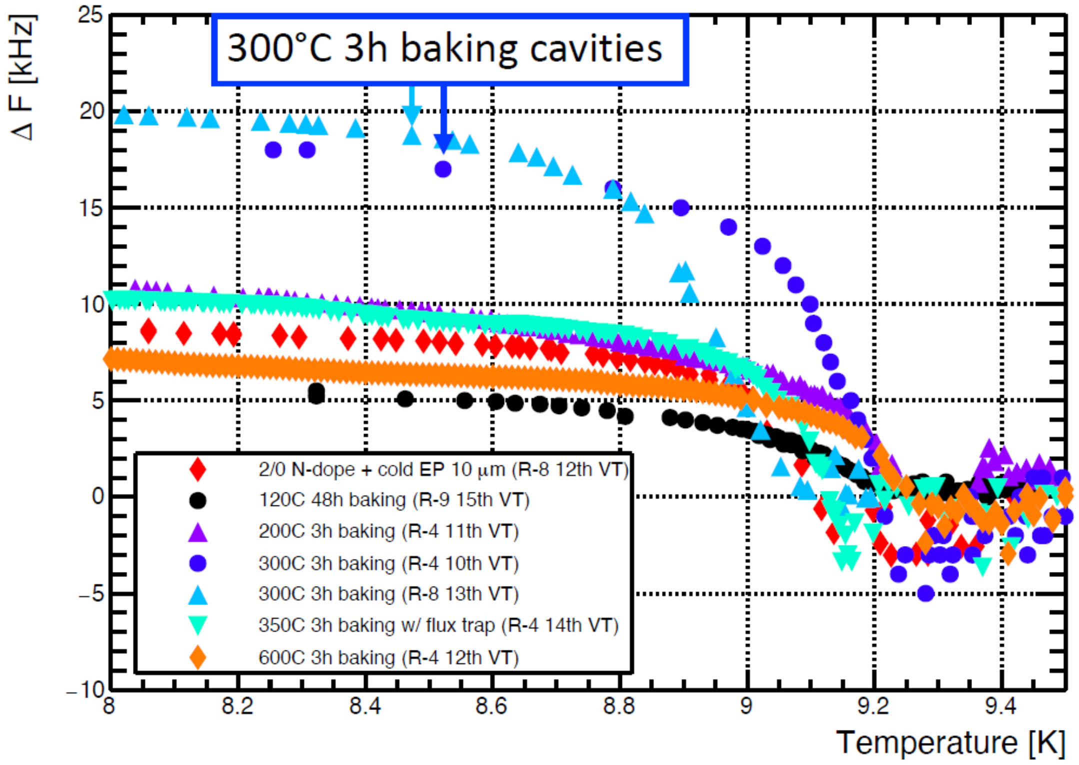
Decomposition of the surface resistance – 2nd run

Same recipe – but with caps

Frequency measurements

A local minium below T_c appears!



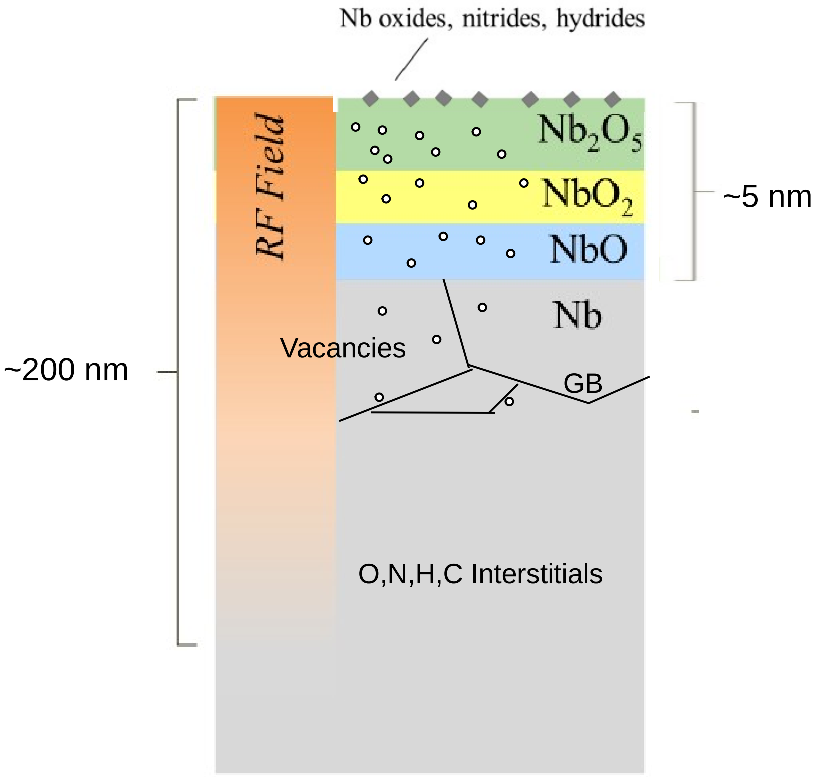


[Ito, H., et al. *Prog. Theor. Exp. Phys.* 2015 (2021).]

This dip is a feature of the Mid-T bake!
Was discussed only in context of doping before!
[Bafia, D., et al. *arXiv preprint arXiv*:2103.10601 (2021).]

Understanding SRF performance needs material science

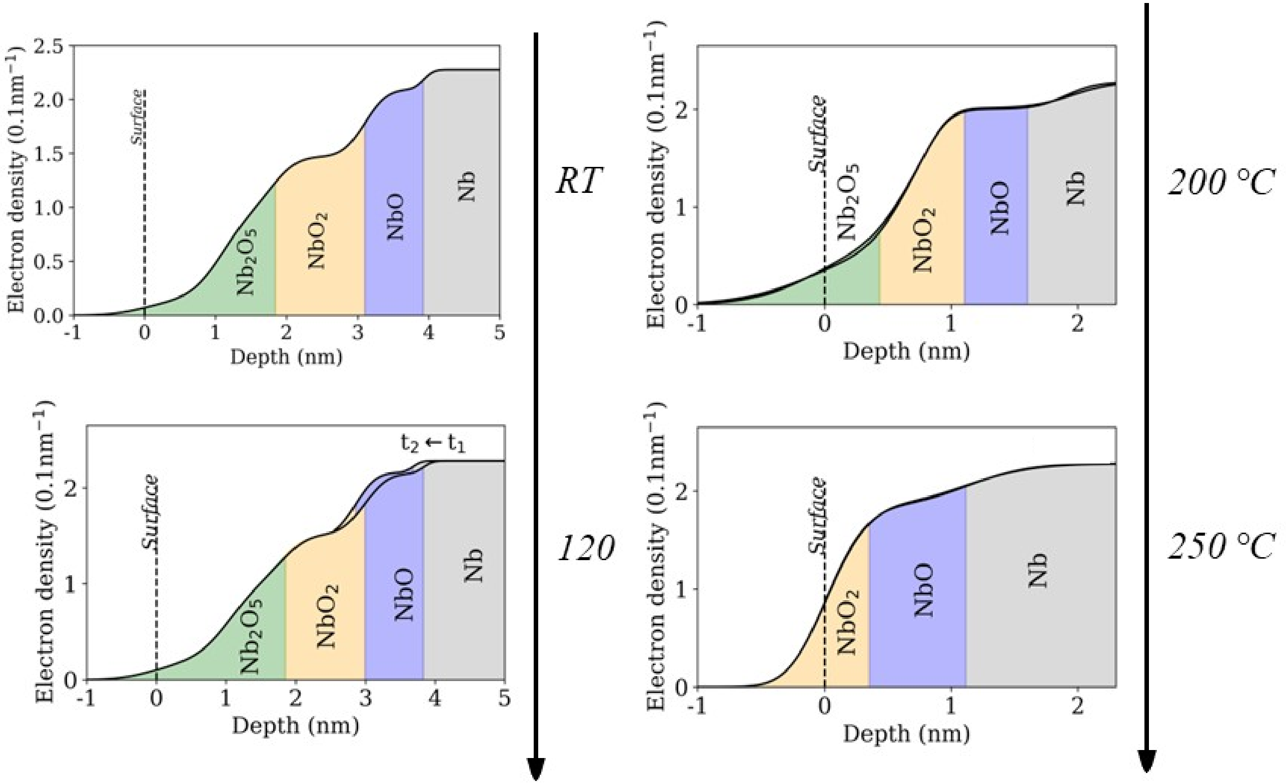
- RF performance is affected by multiple surface properties
  - Recipes to achieve good rf performance exists (European XFEL, LCLS-II,…)
  - Don’t always know why recipe works – and why sometimes it doesn’t
- Opportunity to push our understanding of SRF by challenging observations
  - Doping vs. Mid-T Bake and Infusion vs. 120°C anneal
- Understanding of underlying surface dynamics
  - is mandatory to identify key parameters for SRF performance
  - helps to develop stable recipes for industry / large scale applications

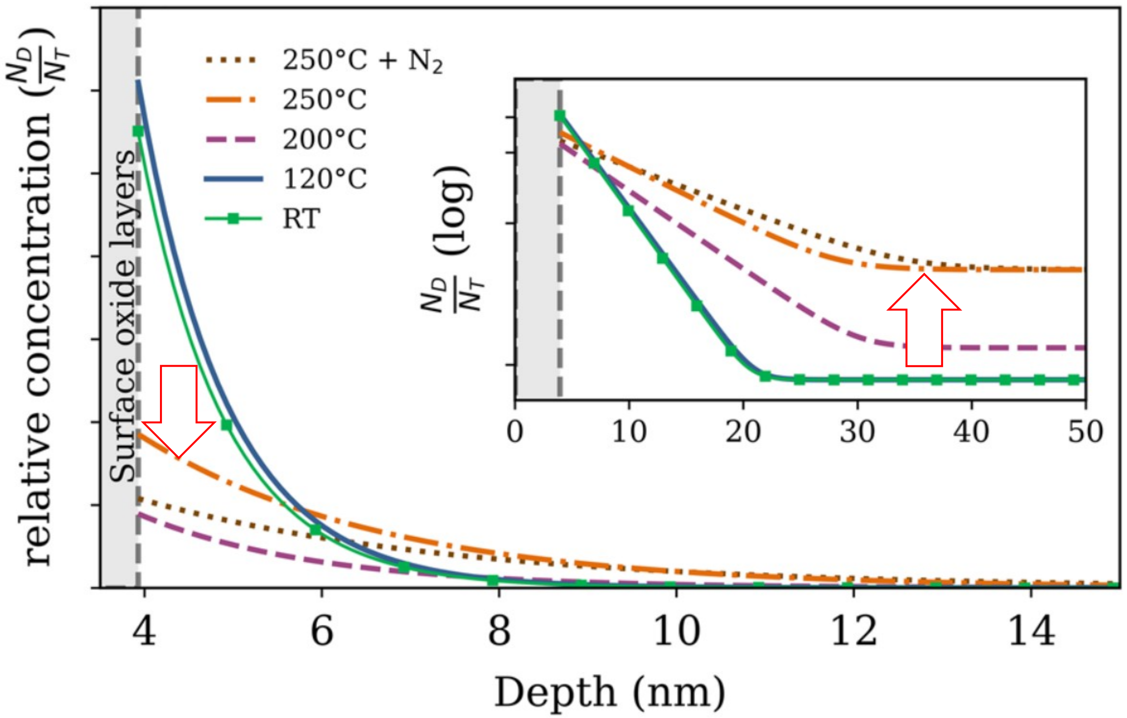


Chemical composition of the oxide-layer

From this point of view nothing new

[Semione, G. D. L., et al. *PRAB* 22.10 (2019): 103102.]





Well known behavior:

- Dissociation of Nb_2O_5 at 200-250°C (in vacuum)
- Increase of oxygen concentration in the lattice deep into the bulk
- In agreement (and not!): SIMS of Mid-T samples @TJNAF

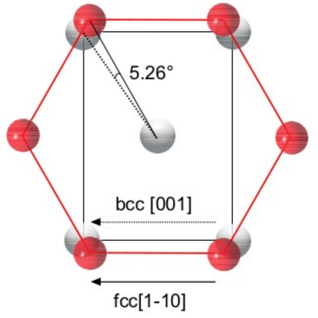
[Lechner, E., et al. *arXiv preprint arXiv* 2106.06647 (2021).]

XRD: Annealing at 300°C reduces lattice mismatch

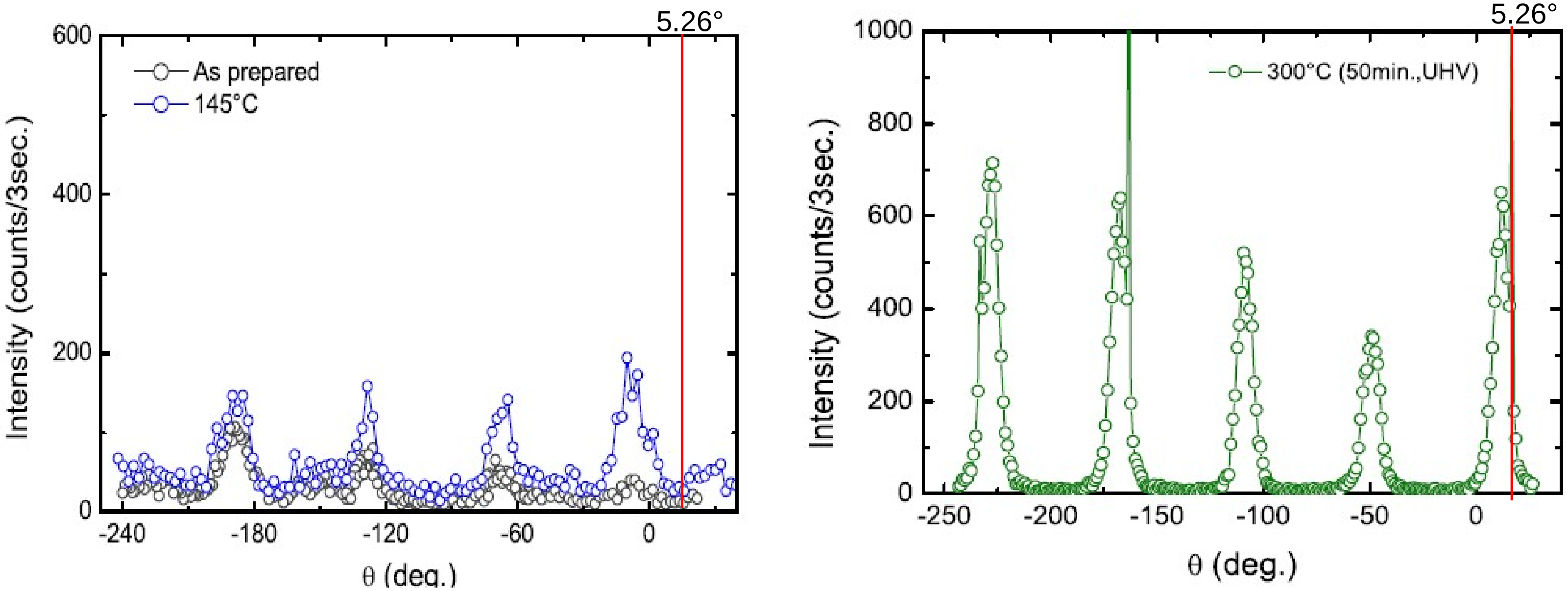
And creates new ordered interstitial oxygen layer

- Nb – NbO have a lattice mismatch (bcc – fcc) when stacking
- For energetic optimum, e.g. for Nb(100), a tilt between cells of 5.26° is expected
- In reality, depends on: layer thickness, lattice orientation, growth temperature, interstitial concentration & type

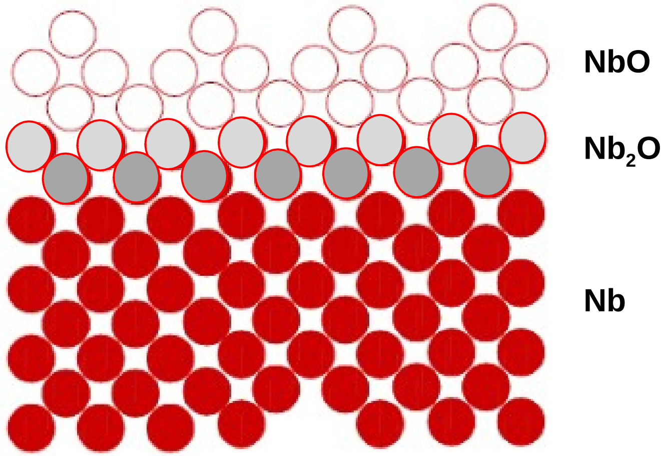
Only Nb-atoms shown



Nishiyama-Wasserman (NW)



[Delheusy, M., "X-ray investigation of Nb/O interfaces." Thesis, (2008).]



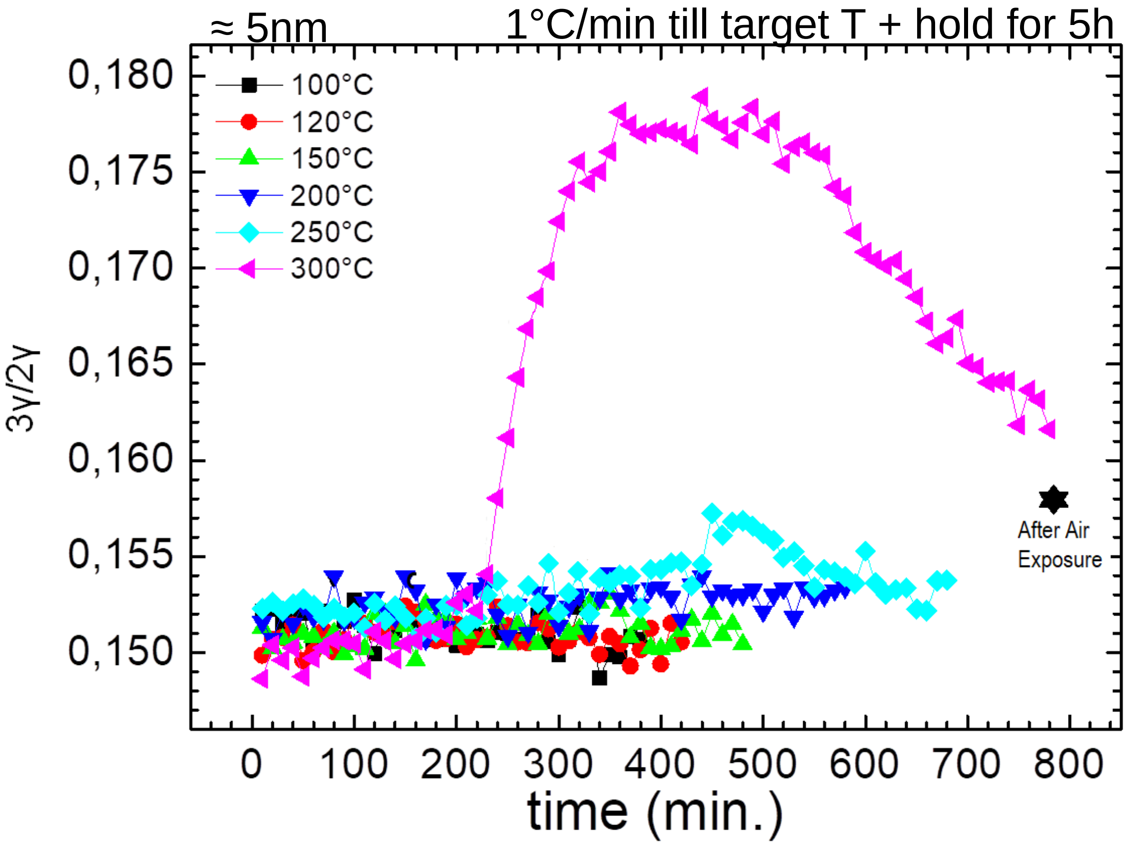
Interstitial oxygen ordering causing smooth transition

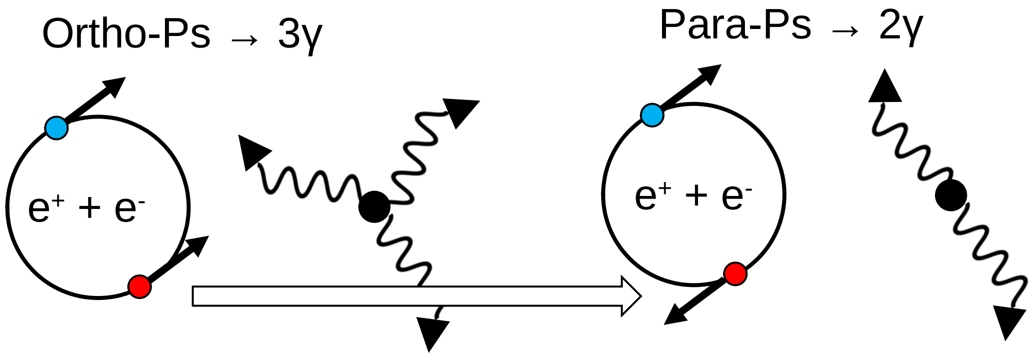
[Zhussupbekov, K., et al. *SciRep* 10.1 (2020): 1-9.]

[Todorova, M., et al. *PRL* 89.9 (2002): 096103.]

Baking at 300°C reduces magnetic impurities

When the lattice mismatch is reduced, vacancy concentration decreases





- At 300°C
  - 3γ/2γ increases
  - Remains elevated at RT & even after air exposure!
  - Nb_2O_5 already gone, hence not the origin!

- Increase of 3γ/2γ means decrease of magnetic impurities
- Origin: localized magnetic moments at oxide-vacancies which can lead to surface ferromagnetism

[Weissmann, M., et al., *Physica B: Condens* 398.2 (2007): 179-183.]
[Venkatesan, M., et al. *Nature* 430.7000 (2004): 630-630.]
[Hong, N.H., et al. *Phys. Rev. B* 73.13 (2006): 132404.]

- Magnetic impurities are not new in SRF and correlation with RF behavior was shown already

[Proslier, Thomas, et al. *IEEE Trans. Appl. Supercond* 21.3 (2011): 2619-2622.]

Looking for magnetic impurities

Kerr-Microscopy determines magnetization at room temperature

Ferromagnetic behavior

⇩

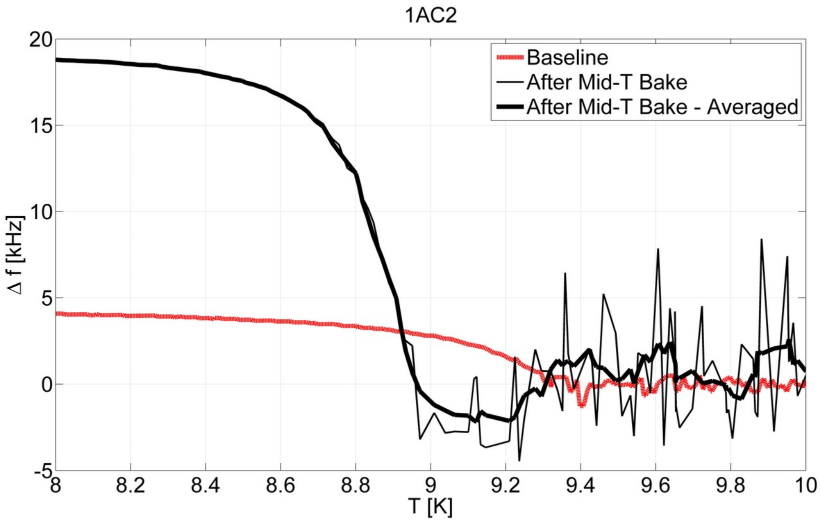
Paramagnetic behavior

Magnetic coupling becomes weaker when hydrogen-hydrogen distance increases

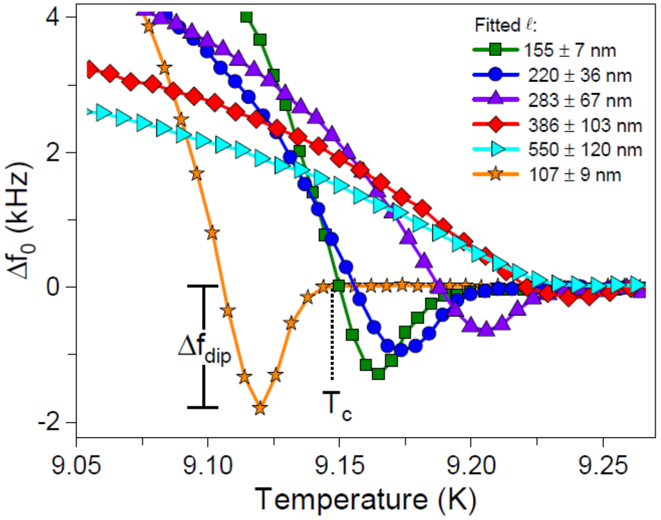
[Esquinazi, Pablo, et al. *IEEE Trans. Mag.* 49.8 (2013): 4668-4674.]

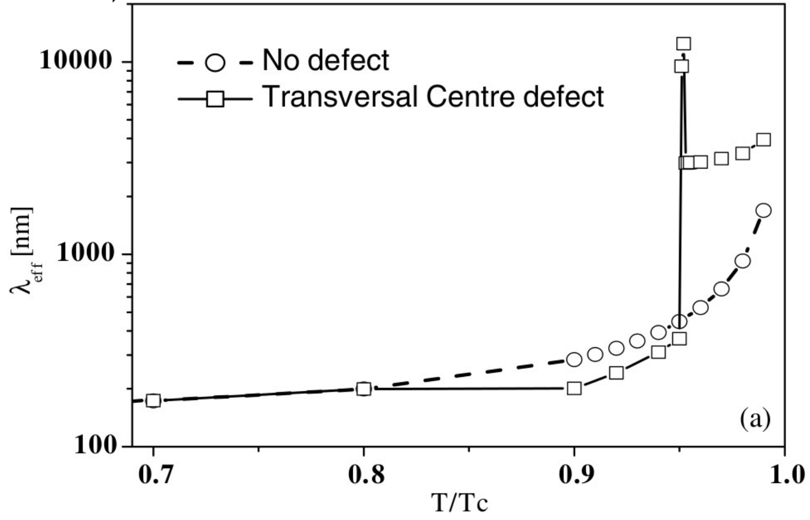
- Oxide layer reduces lattice mismatch at 300°
  1. Reduction of oxygen vacancies (acting as magnetic impurities)
  2. Formation of Nb_2O layer
- Nb_2O_5 layer dissolves and oxygen diffuses into the lattice

Dip in f vs. T caused by current redistribution



Bafia, D., et al. *arXiv preprint arXiv*:2103.10601 (2021).] ⇨ **[Barra, M., et al. *SUST* 18.3 (2005): 271.]**





Current redistribution due to regions with $T_c < T_{c,Nb}$

Origin for Doping and Mid-T bake:

- β-Nb_2N has been found on the surface of doped cavities after 5µm EP **[Spradlin, J. K., et al., SRF2019 MOP030.]**
- An increased oxygen concentration reduces T_c of Nb (≈ 1K per at.%)

This would also explain the higher sensitivity nΩ/mGg towards trapped flux

How can the Mid-T bake affect the surface resistance?

1. Direct impact on density of states (DOS)
   - Nb_2O formation changes core-levels and valence-band distributions
   - Electric properties of Nb_2O_5 varies depending on substrate properties

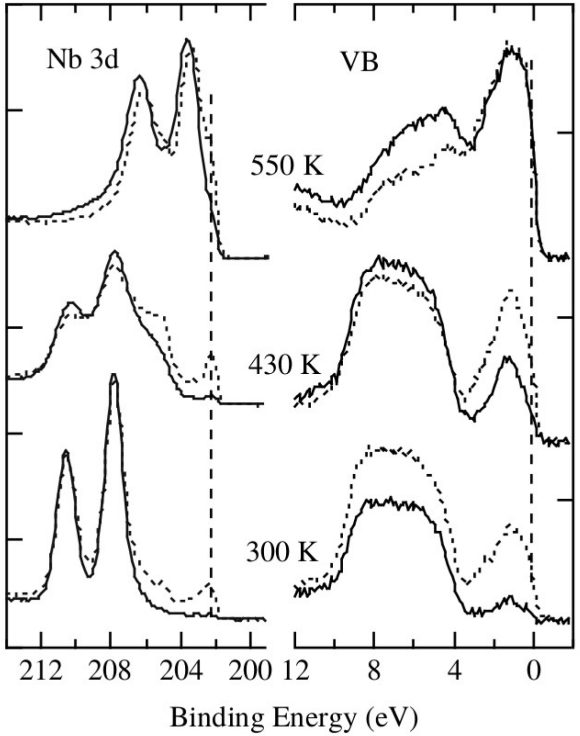
[Zhussupbekov, K., et al. *SciRep* 10.1 (2020): 1-9.]

2. Indirect impact on DOS by magnetic impurities

[Proslier, Thomas, et al. *IEEE Trans. Appl. Supercond* 21.3 (2011): 2619-2622.]
[Kharitonov, M., et al. *Physical Review B* 86.2 (2012): 024514.]

3. Oxygen (like nitrogen) has a high trapping potential for hydrogen, mitigating hydrides

[Zapp, P. E., and Birnbaum H.K. *Acta Metallurgica* 28.11 (1980): 1523-1526.]
[Romanenko, A., et al. *SUST* 26.3 (2013): 035003.]



[Ma, Qing, et al. *JApplPhys.* 96.12 (2004): 7675-7680]

Modifications of DOS and their impact on the residual and BCS resistance are being theoretically discussed

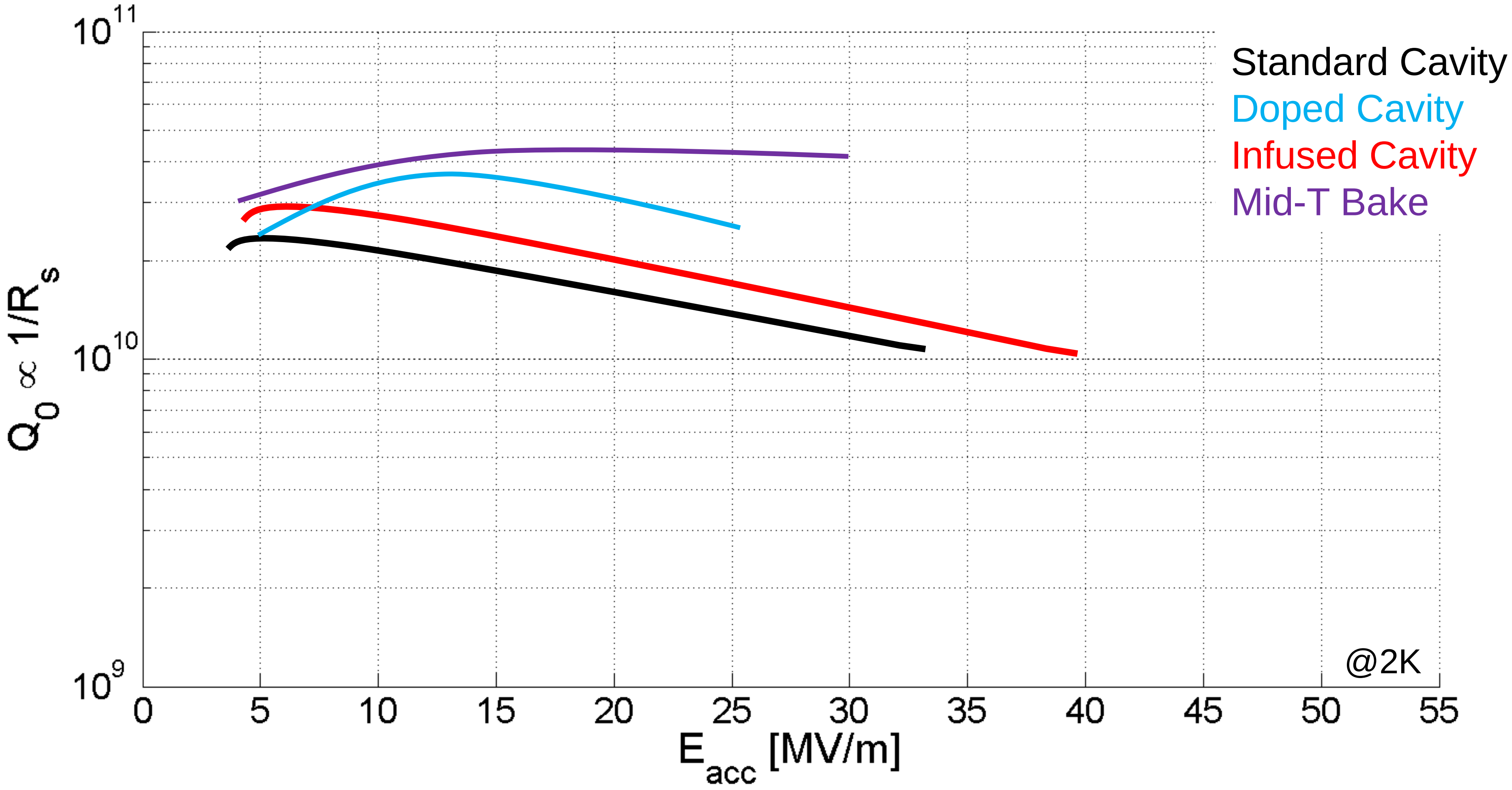
[Kubo, T., & Gurevich, A. *Phys Rev B* 100.6 (2019): 064522.]

But what is challenging BCS Theory?

BCS shouldn‘t be working in the first place!

But $dR_s/dE_{acc} > 0$ „makes sense“

[Kubo, T., & Gurevich, A. *Phys Rev B* 100.6 (2019): 064522.]



[Reschke et al., Phys. Rev. Accel. Beams, 20, 042004 (2017)]
[Grassellino et al., SUST, 26, 102001 (2013)]
[Grassellino et al., SUST, 30, 094004 (2017)]
[Posen et al., Phys. Rev. Applied 13, 014024]

Cavities limited by quench

WHAT IF...?

- Infusion @ 160°C looks like a doped cavity (both introduce N into Nb)
- Mid-T Bake has „anti-Q-slope“ like doped cavity and “the dip” (but no N into Nb)
- Infusion below 160°C (w./ N) looks like 120°C bake (w./o. N) but different offset

What if all these annealing procedures are related or do the same thing!

What might „the same thing“ be?

How does “this thing” influence the rf properties?

„Impurity Tailoring“

Mixture of several models, measurements and ideas

- Hydrogen is bad – tends to accumulate near the surface, form lossy (nano)hydrides
- Native Nb-Oxides have vacancies which act as TLS creating losses

→ Near-Surface Lattice is not in the perfect shape

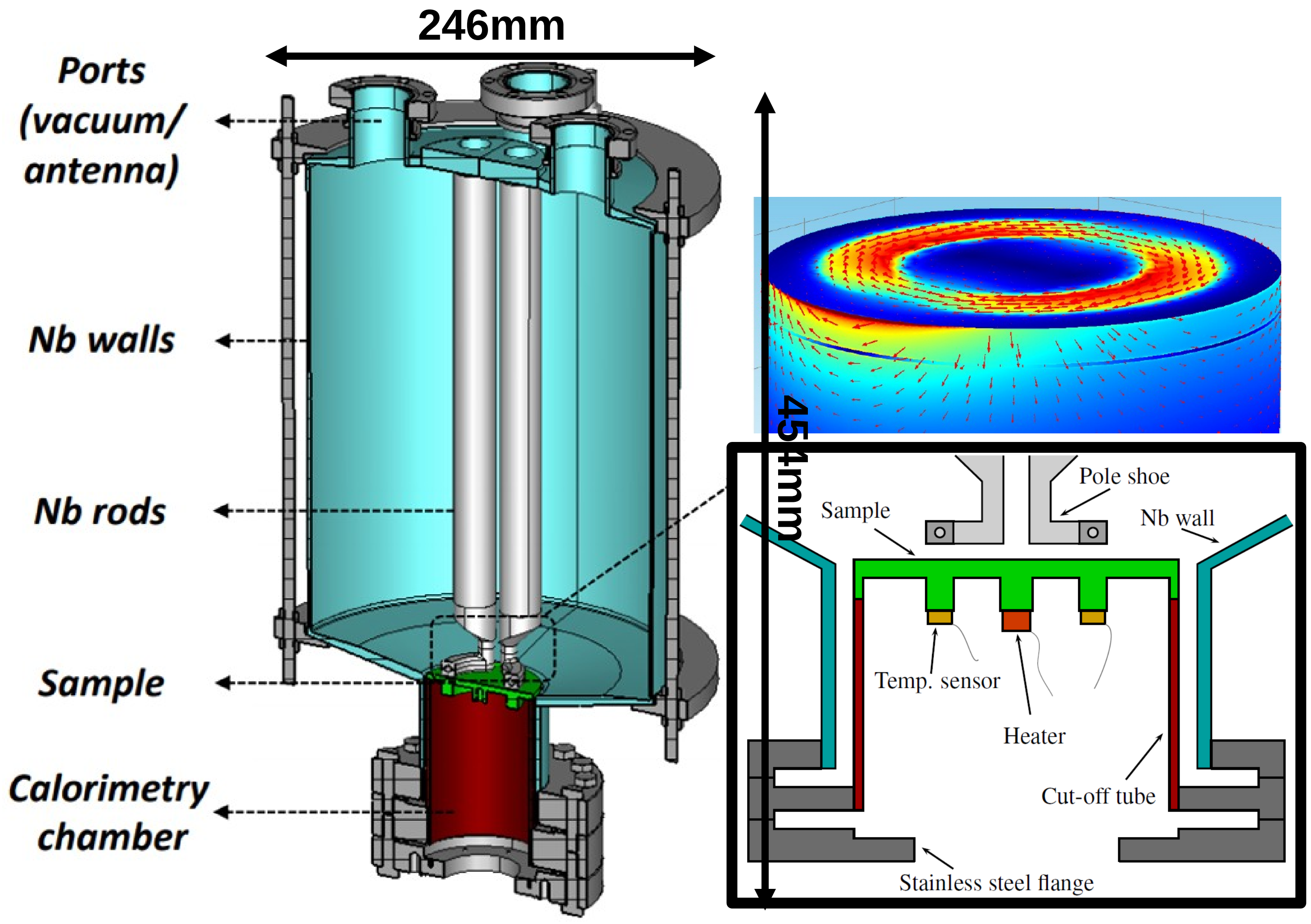
- Annealings do one thing: modify concentrations of C, H, N, O and vacancies
  - Vacancies and interstitial N or O can trap hydrogen / prevent hydride formation
  - Modify Nb-Oxides to form less defective phases
  - Shift induced currents away from the lossy surface region by manipulating λ_L
  - Spread currents over larger volume, effectively increase applicable gradient
  - Change DOS, electron-phonon coupling and qp relaxation times

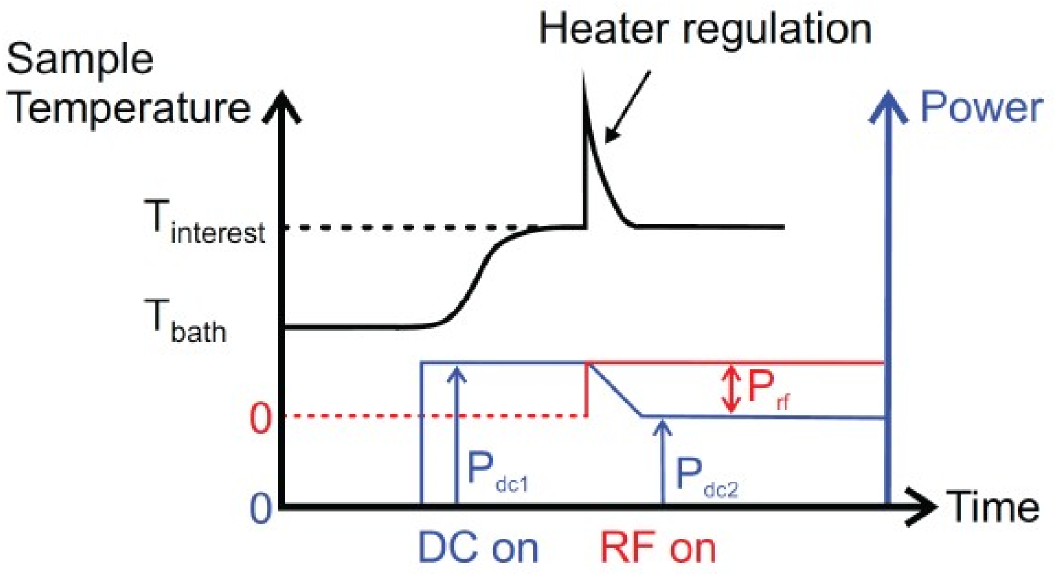
Fascinating new ideas – completely new approaches – fundamental new understanding

Extend Capabilities: RF Properties of Samples

Quadrupole Resonator:full rf characterization of samples

Opens both eyes: Easy accessible surface and „cavity-like“ rf test



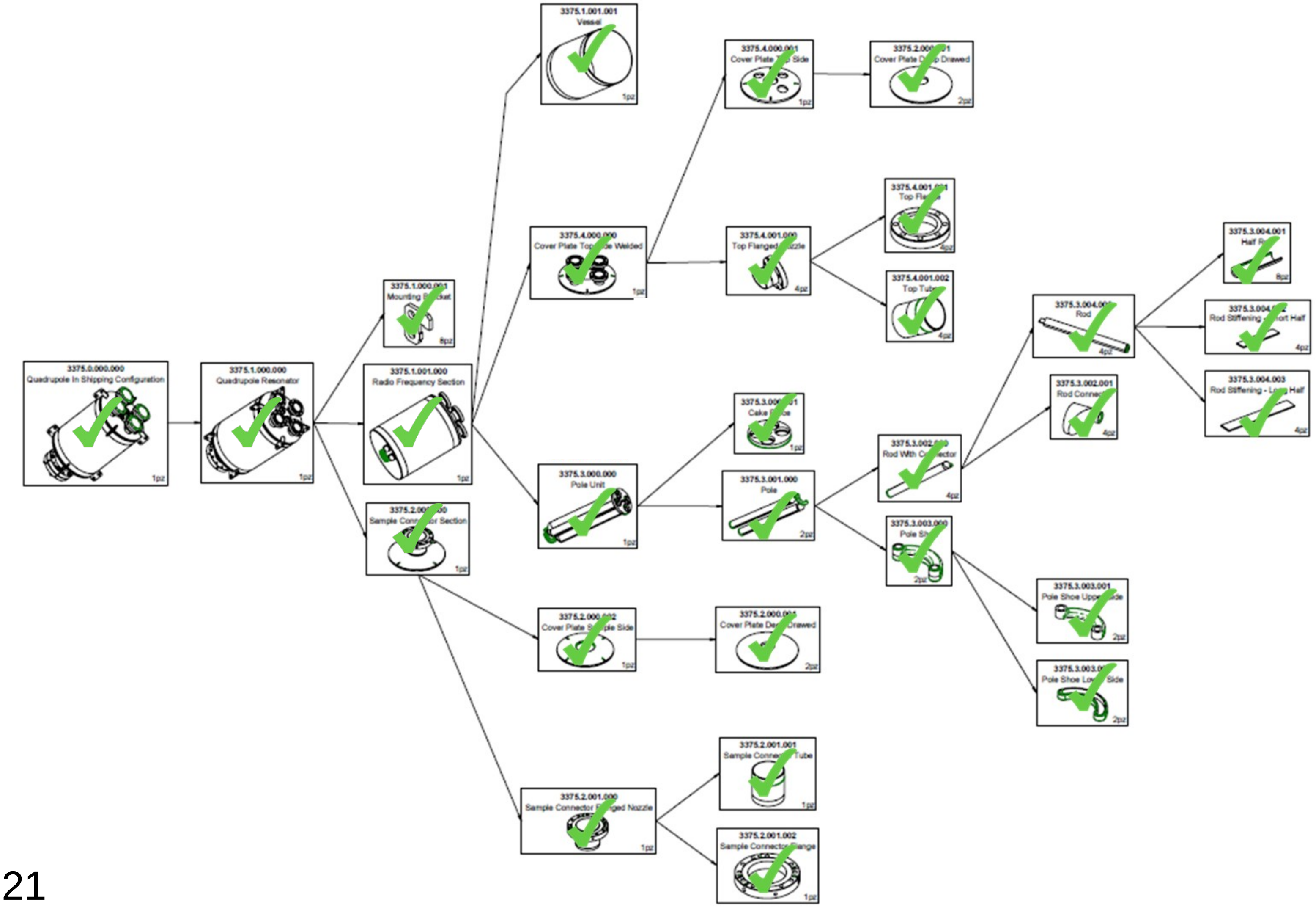


f = n x 433 MHz
T = 1.8-15 K
B ≤ 120 mT

- Surface Resistance R_s (T,B,f) and R_{res}(B,f)
- Critical magnetic field H_C(T)
- Thermodynamic limit $H_{C,max} = H_C(0K)$
- Penetration depth λ_L
- Mean free path ℓ / RRR

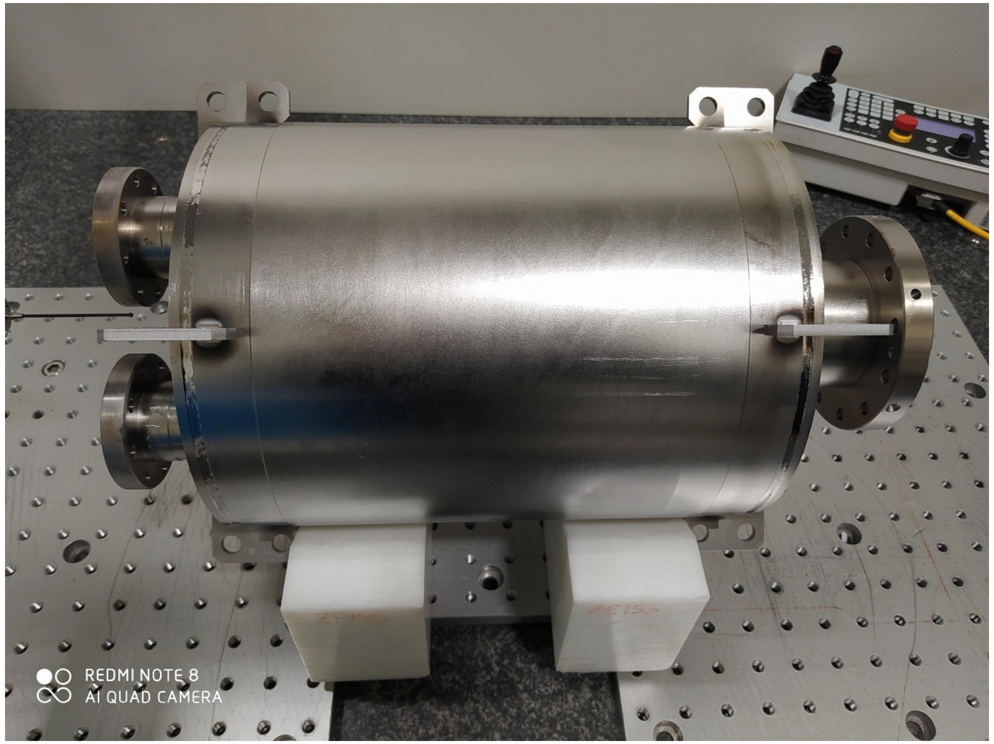
Project Timeline

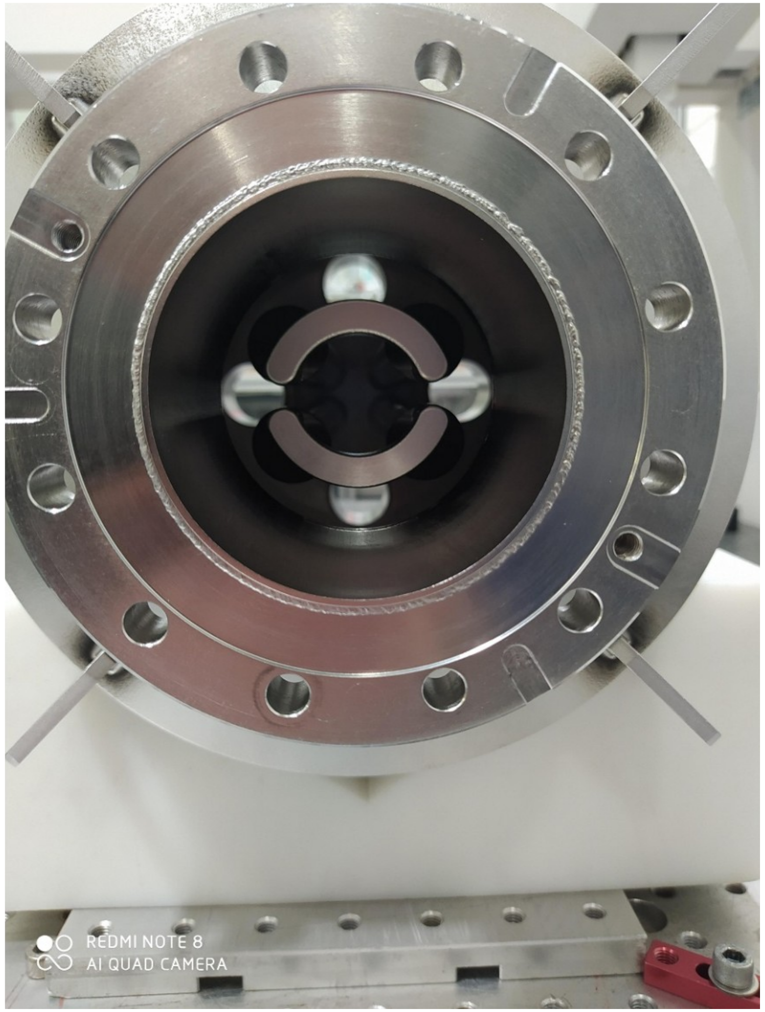
- ~1y order preparation
- Order placed @ Zanon Nov. 2019
- First Projectmeeting Dec. 2019
- Material delivered Week 1 2020
- SARS-COV-2 (+7 months)
- Fabrication started 24. July 2020
- Final Weld: 24. February 2021
- QPR delivered @DESY 7. May 2021
- First BCP 31. August 2021
- First commissioning cooldown Dec. 2021



Final QPR

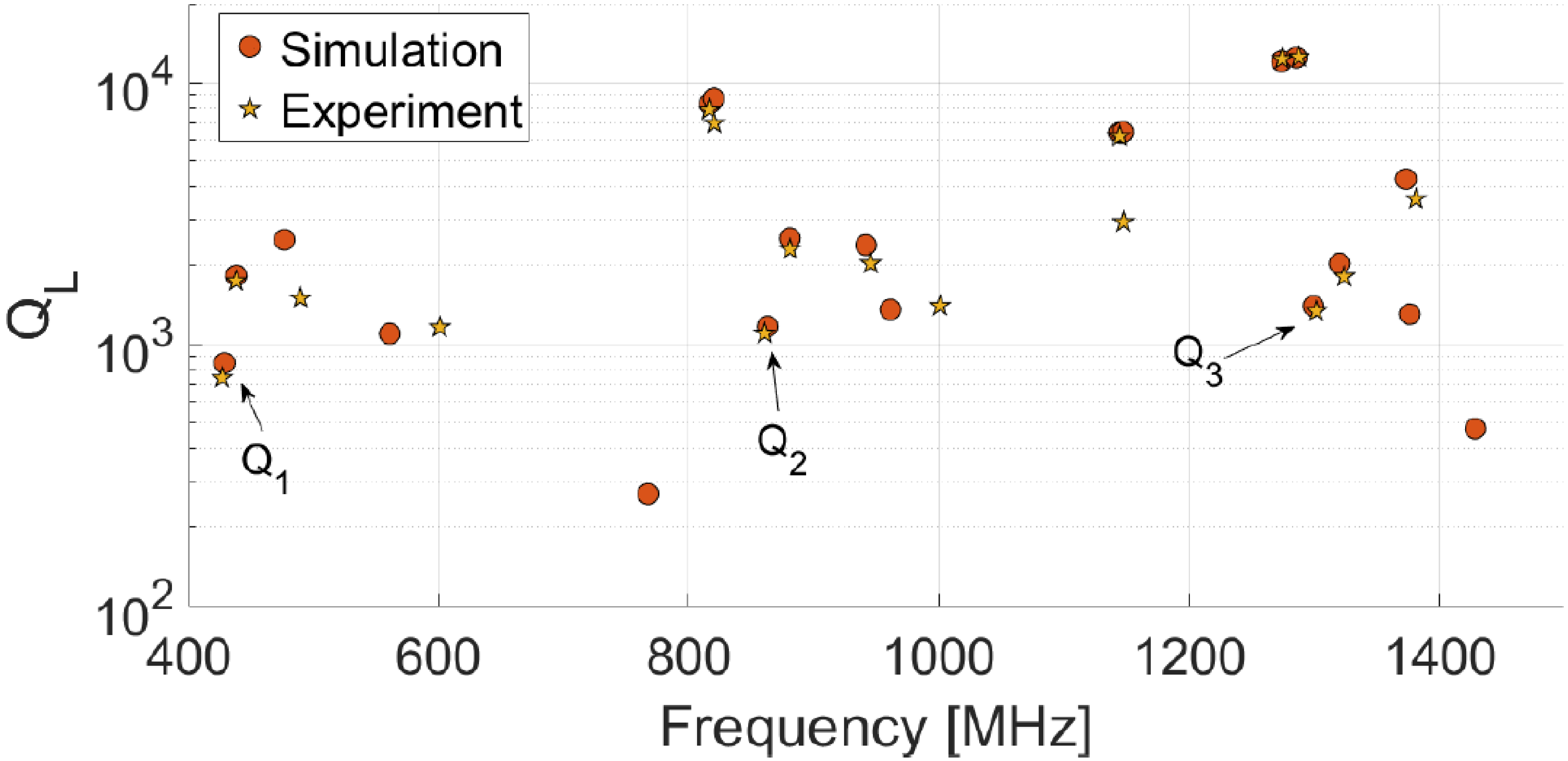
Before Treatment





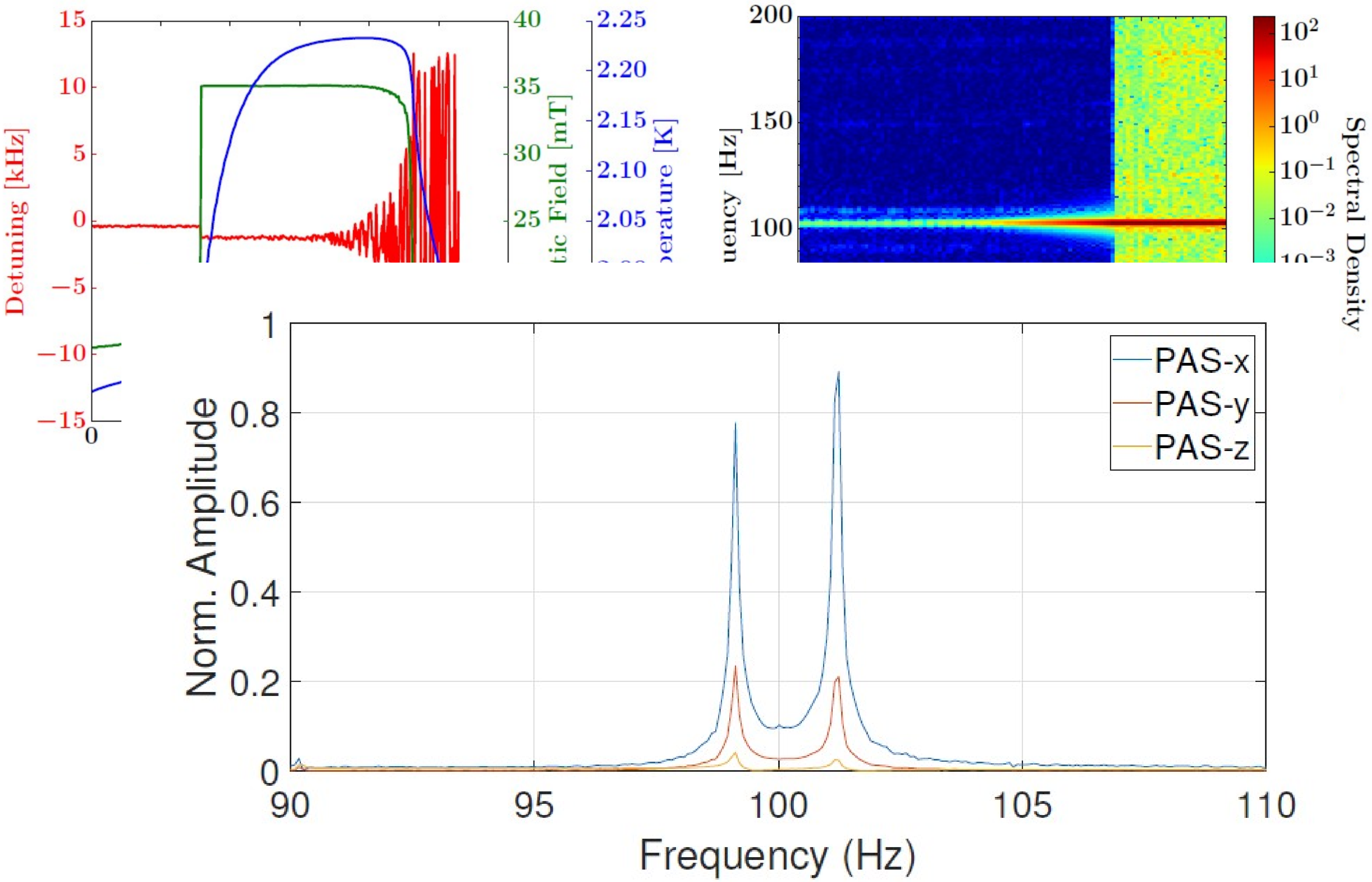
First commissioning

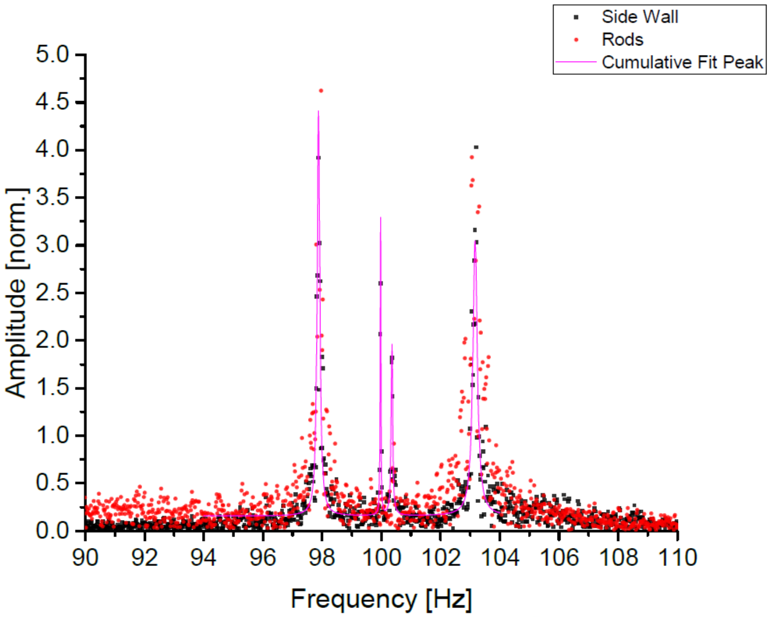
RF Spectra @ RT



First commissioning

Microphonics





Beyond Niobium

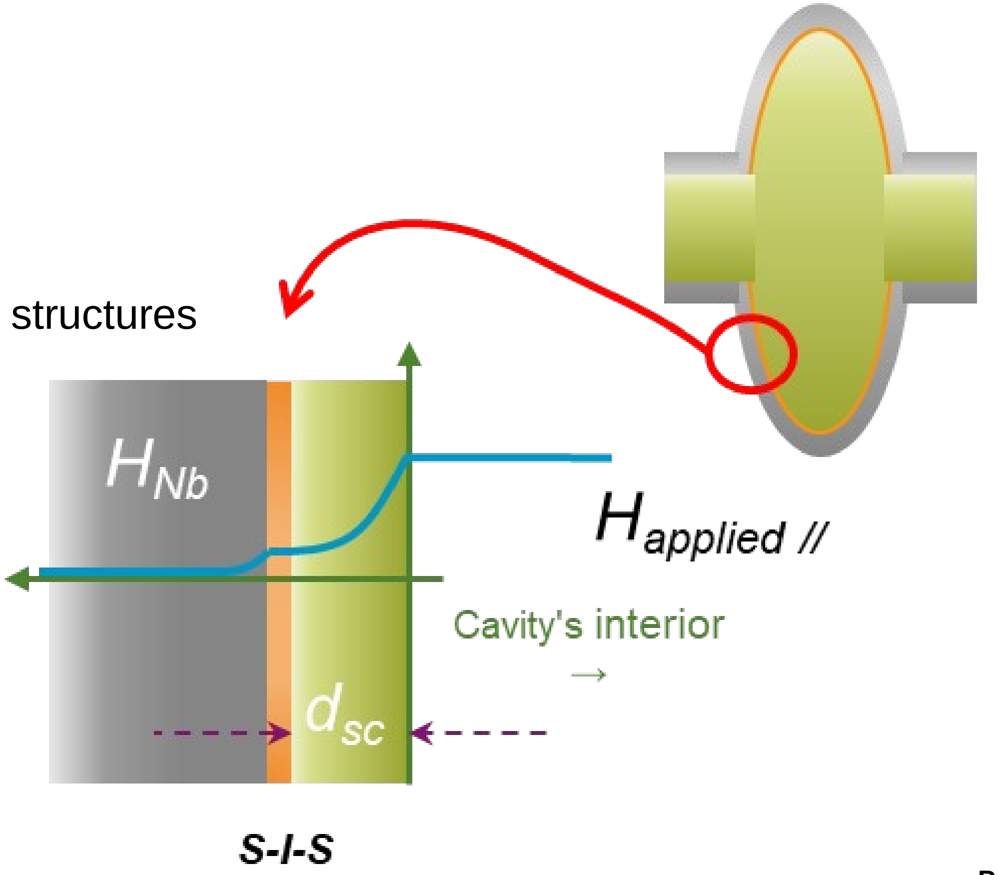
Why do we have to go there?

What is the limit?

- Intrinsic material properties of Nb
  - Still open questions – but a “saturation” in performance is obvious
- Go to alternative materials
  - Nb_3Sn
  - …
- Go to new structures
  - alternating Superconducting - Insulator - Superconductor (SIS) structures

[A. Gurevich, APL 88 (2006)]

[T. Kubo, SUST 30 (2017)]



Taken from: C. Antoine, TTC Meeting 2020

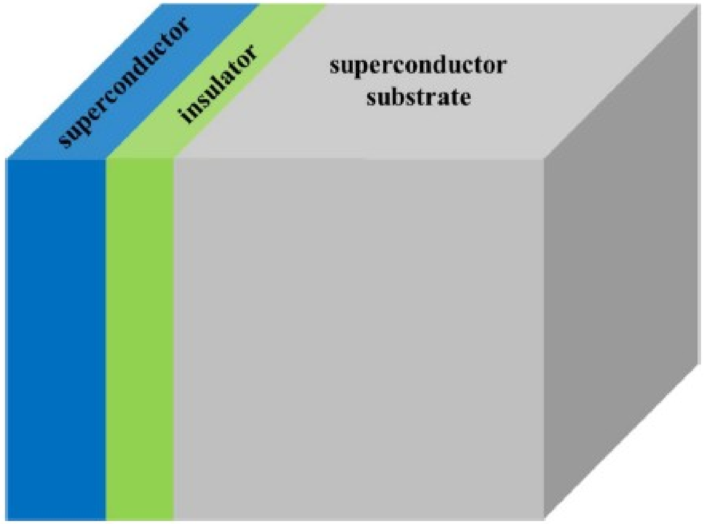
Why SIS and not simply bulk?

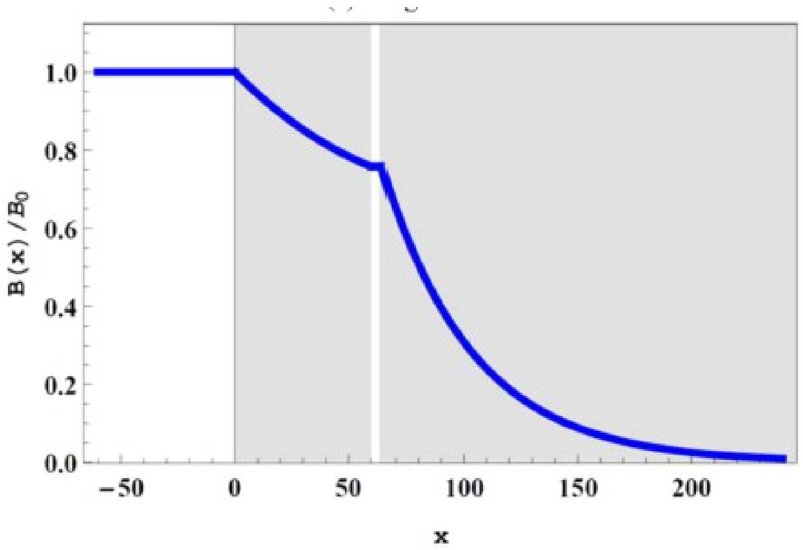
- Use higher T_c superconductor as top-layer
  - Fewer losses at 2K comared to Nb due to $R_{BCS} \sim \exp(-T_c)$
  - Distribute current due to higher λ_L
- Max. $E_{acc} \sim H_{sh}$ (not H_{c1})
- Why H_{sh}? Meta-stable against flux penetration
- Bean-Livingston Barrier prevent vortex penetration
  - Need mirror surfaces

Insulator plays crucial role

Not just from rf pov

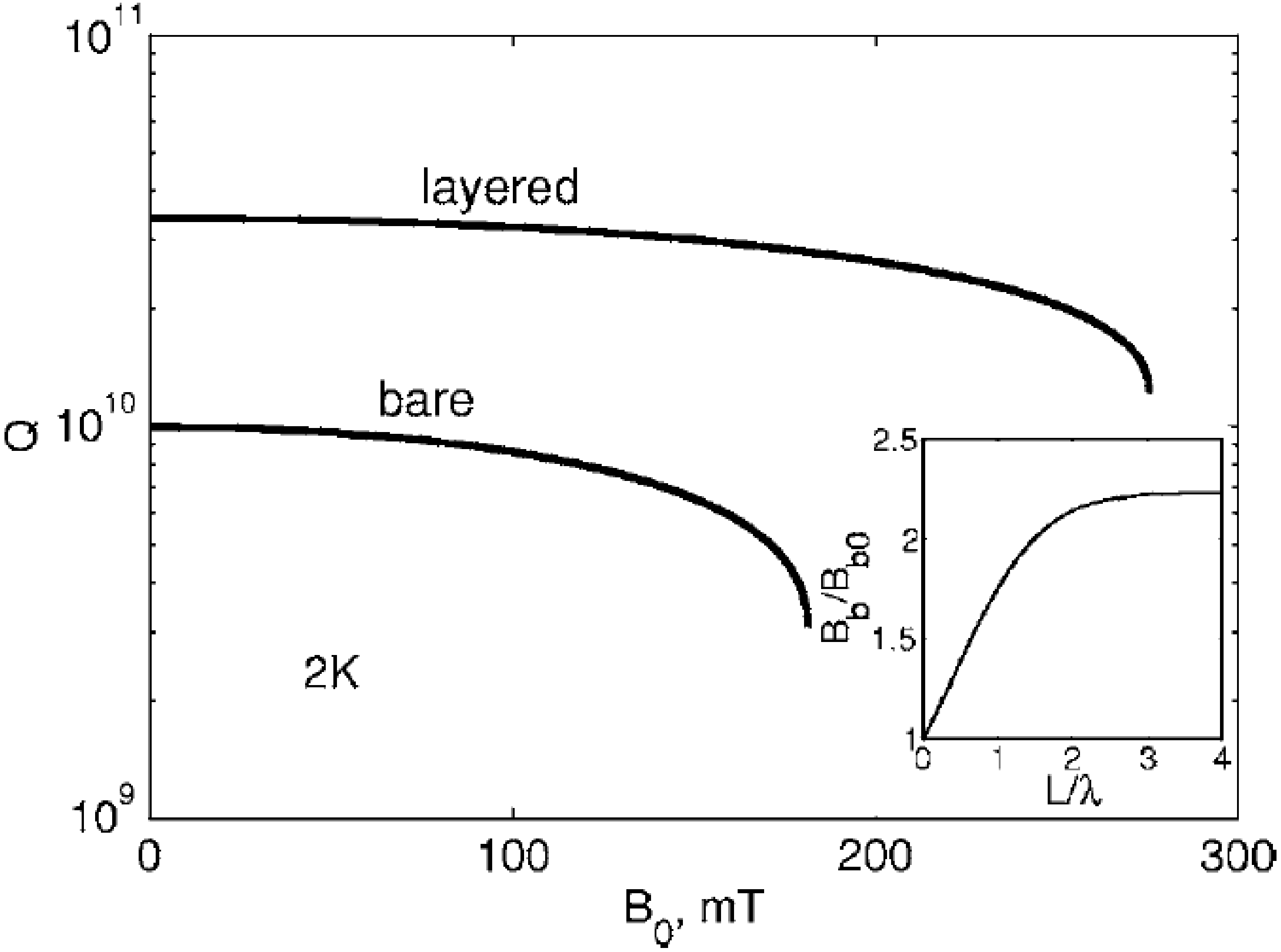
1. Majority of losses in the S layer
2. More “mirrors” create more screening currents means less flux!
3. If flux enters – trap it
   - No avalanche leading to a quench
4. Isolater thickness plays a role to prevent Josephson Junction





What is the result?

[A. Gurevich, APL 88 (2006)]



Is there an optimal layer thickness?

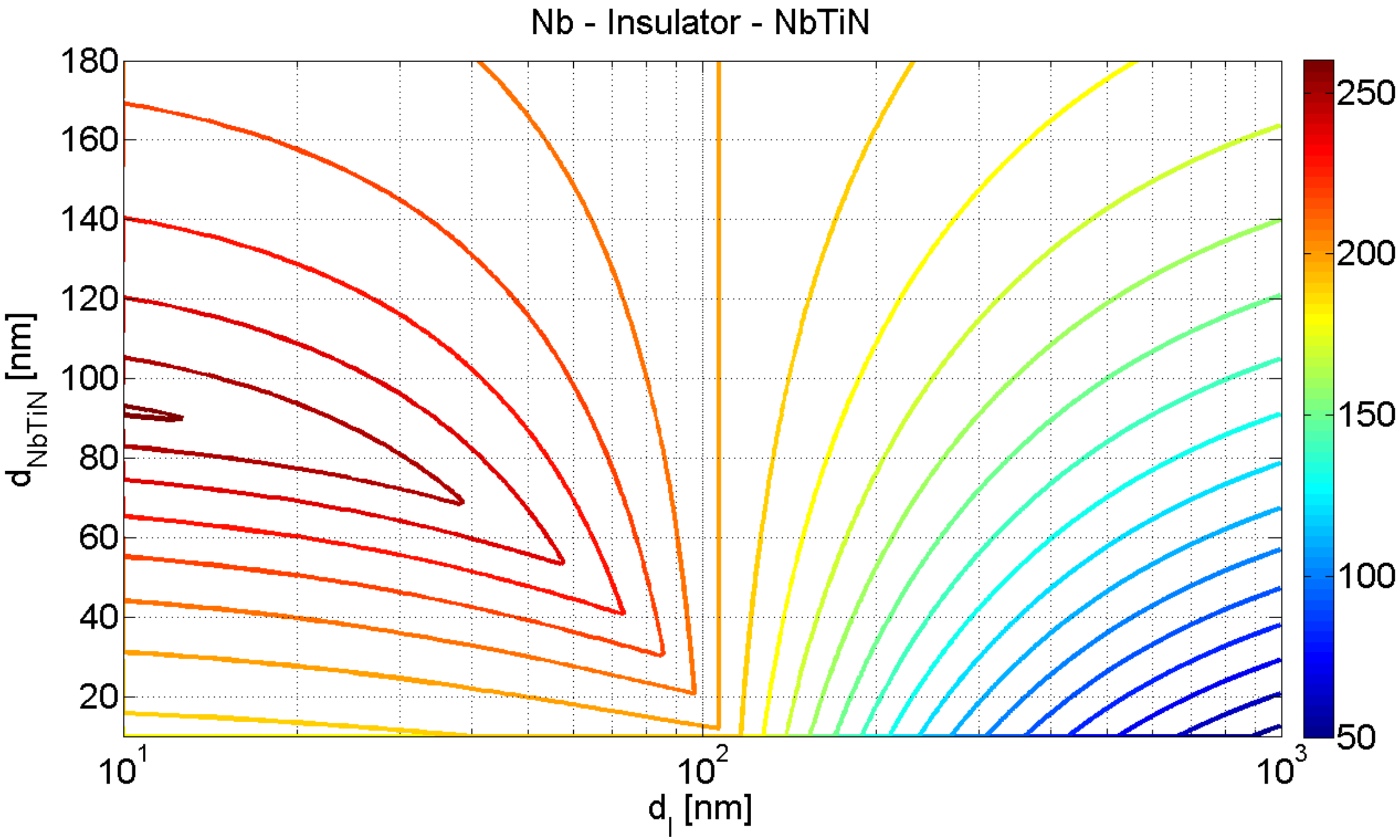
[T. Kubo, SUST 30 (2017)]

- Depends on $H_{c,1}$ and λ_L of both S
  - Here NbTiN – I – Nb
    - $H_{c,1}$ Nb 180 mT
    - $H_{c,1}$ NbTiN 200 mT
- Weapon of choice: Atomic Layer Deposition

Pros:
  - Achieve required film homogeneity
  - Cavity geometry not a problem

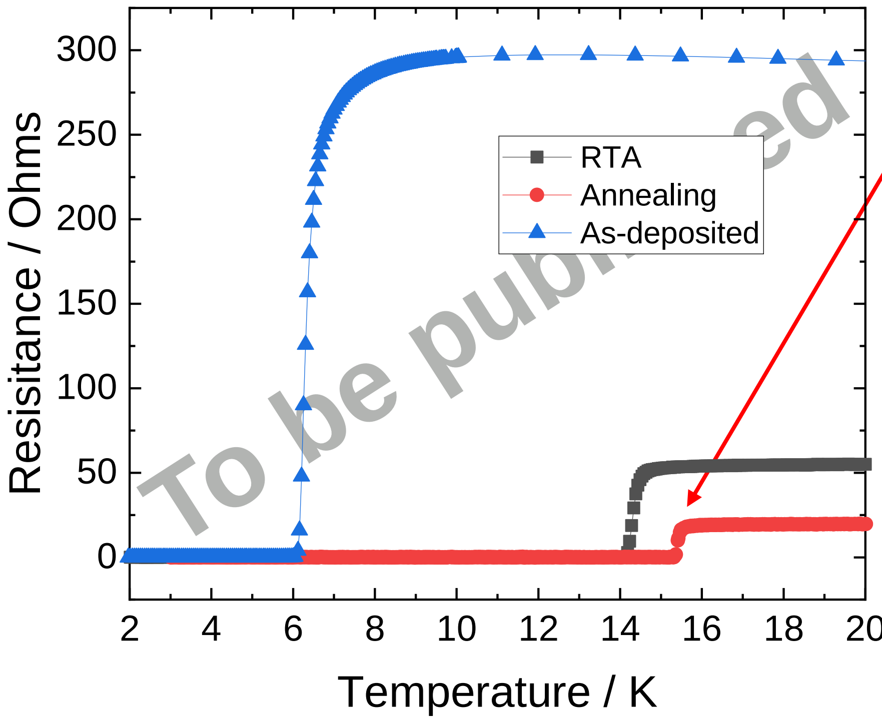
Cons:
  - Not all stoichiometries possible
  - Elevated temperatures

To be decided:
  - Films are amorphous
  - Native Nb-oxides vs. Insulator



Where we have gotten so far

- Successful R&D pursued in current **SMART** project shall be continued in **NOVALIS**
- The goal of the project is to coat single-cell cavities with layered structures and investigate new rf materials
- Developed **SMART recipe leads to a T_c of 15.4 K** on Nb-samples coated with 15 / 25 nm of AlN / NbTiN



NOVALIS will apply SMART recipe to real cavities

Proof-of-Principle experiment to coat a DESY single-cell cavity with Al_2O_3 using Thermal ALD currently prepared

Summary

- Project driven (DESY) and fundamental research (UHH) ongoing – beneficial for both
- Our annealing studies had their ups and downs
  - „Forensic“ material analysis key to improvement
  - Major invest in new furnaces
  - New recipes will be studied will material characterization and cavity tests
- A sweet spot in „SRF history“
  - Challenge settled knowledge
  - New recipes provide new insights we should leverage
- Gold standard of rf measurements of samples needed
  - QPR delivers just that
- Make the jump for “the next big thing” – and see if we stick the landing
  - Many promising and fun ideas – we moved to SIS with ALD

Thanks to

Martin Aeschlimann (TUKL), Silke Baltruweit (DESY), Christopher Bate (UHH/DESY), Maik Butterling (HZDR), Jakub Cizek (U Prague), Arti Dangwal-Pandey (DESY), Alexey Ermakov (DESY), Erik Hirschmann (HZDR), Wolfgang Hillert (UHH), Bernward Krause (DESY), Arno Jeromin (DESY), Thomas Keller (UHH), Nicolay Krupka (DESY), Oskar Liedke (HZDR), Lutz Lilje (DESY), Giso Marquardt (DESY), Detlef Reschke (DESY), Jörn Schaffran (DESY), Marco Schalwat (DESY), G.D.L. Semione (U Bremen), Sven Sievers (DESY), Lea Steder (DESY), Martin Stiehl (TUKL), Andreas Stierle (DESY), Antonio Speyer (DESY), Birte van der Horst (DESY), Andreas Wagner (HZDR), Hans Weise (DESY), Zanon R.I.

Questions?

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