EASISchool 3

Superconductivity and its applications

Dates: 28/9/2020-9/10/2020
Genoa, Italy

Cristian Pira

Nb thin films

INFN

Outline

- Motivation for thin films in SRF cavities
- How to realize a thin film coating?
- State of the art in Nb thin films (accelerators using thin film technology)
- Characteristics of Nb thin films
- R&D on Nb thin films

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Why thin films for SRF?

1. Reduce material cost
2. Change the surface properties (bulk properties ≠ surface properties)
3. Use materials with poor mechanical properties (but excellent SRF properties)
4. Realize complex structures

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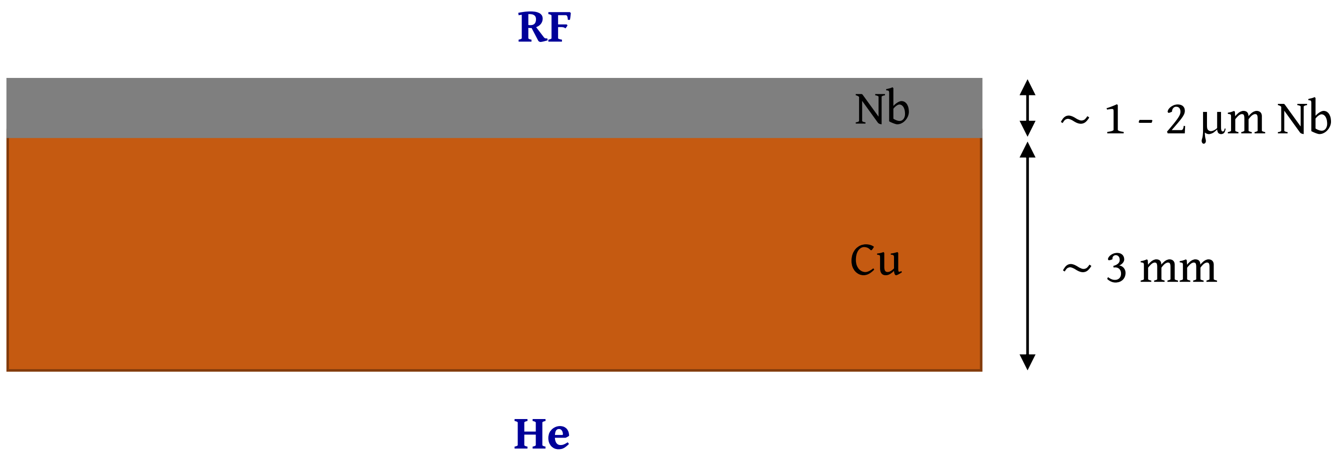
Reduce material cost

RF penetration in Nb is **limited by λ_L** (less than 100 nm)

Not necessary more than 1 micron of Nb at the surface



Cu is **more than 50 times cheaper** than high pure Nb

It is possible to **increase the mechanical stability** of the cavities increasing wall thickness

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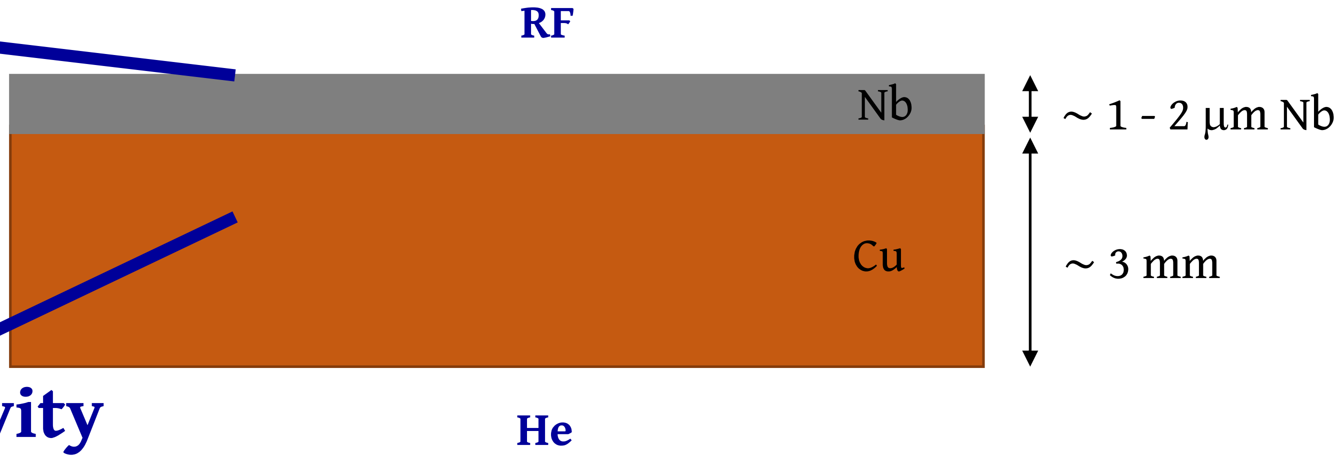
Change the surface properties

Surface: **Low Rs**

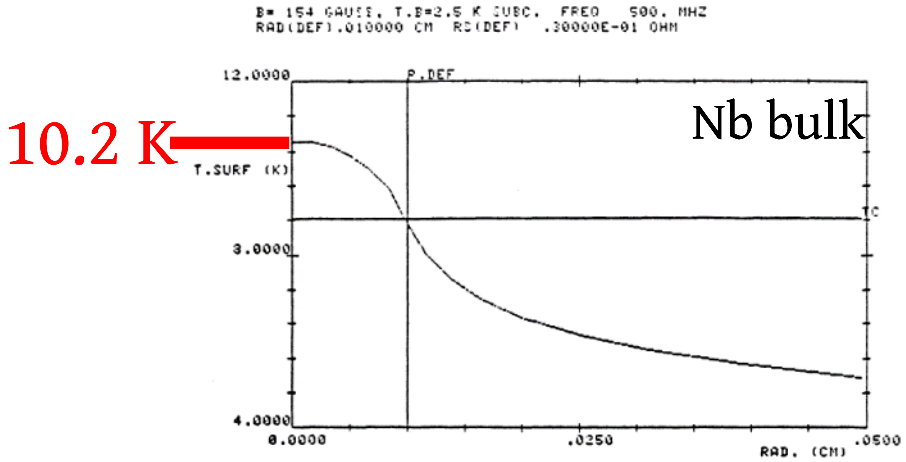


Bulk: **High thermal conductivity**

Cu presents high thermal conductivity → **resistance to quench**

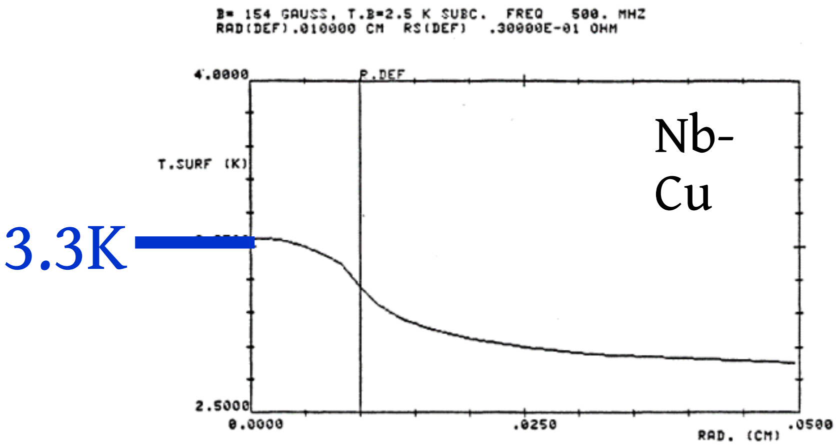
Temperature distribution in Nb-Cu

Temperature distribution simulation for an iron based defect imbedded in Nb or Cu



Copper prevents Quench

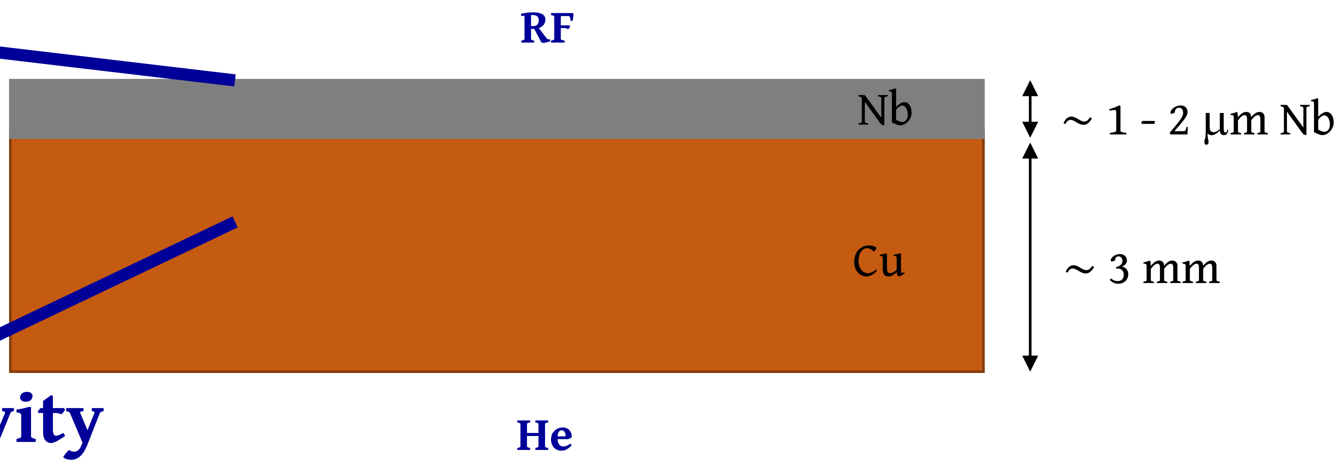
due to thermo-magnetic breakdown



Joachim Tuckmantel *Thermal effects in superconducting RF cavities: some new results from an improved program* CERN-EF-RF-84-6. - 1984

Change the surface properties

Surface: **Low Rs**

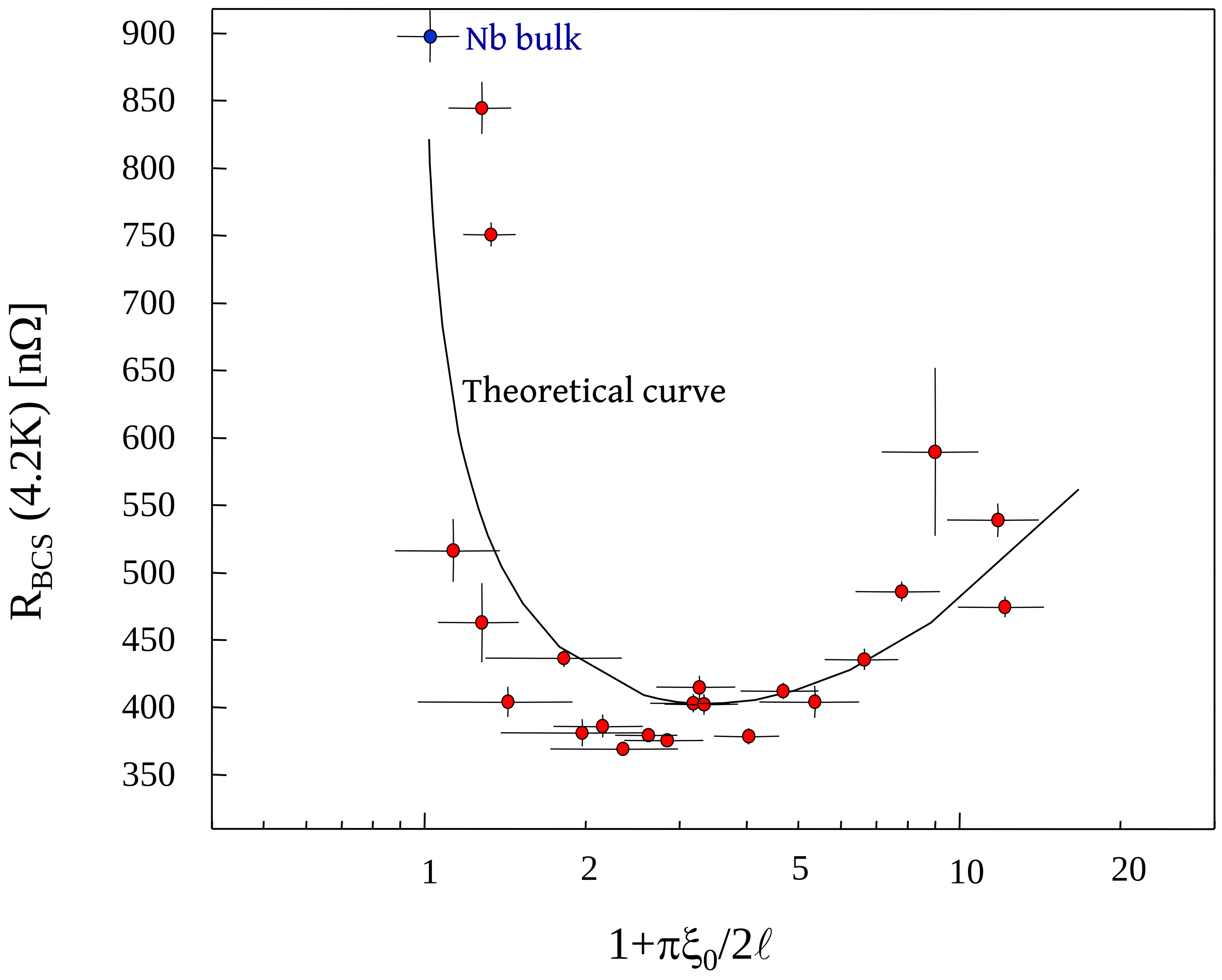


Bulk: **High thermal conductivity**

Cu presents high thermal conductivity → resistance to quench

Nb surface resistance could be modulated

BCS resistance depends on mean free path



R_{BCS} @ 4.2 K

Nb bulk: ~900 nΩ

Nb films: ~400 nΩ

R_{BCS} @ 1.7 K

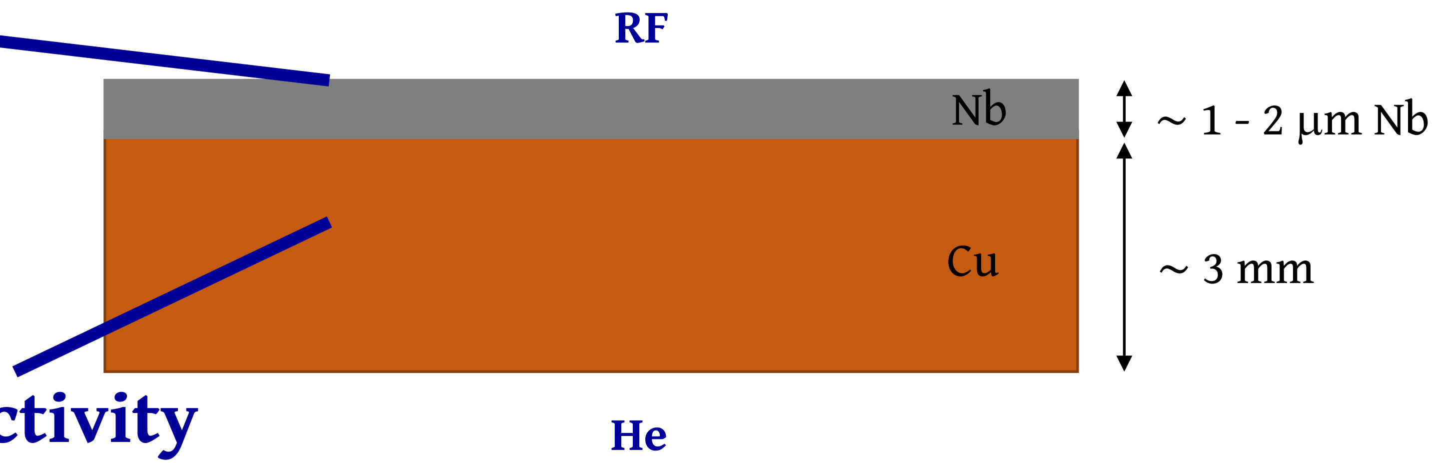
Nb bulk: ~2.5 nΩ

Nb films: ~1.5 nΩ

Benvenuti C et al 1999 Physica C 316 153

Change the surface properties

Surface: **Low Rs**



Bulk: **High thermal conductivity**

Cu presents high thermal conductivity → resistance to quench

Nb surface resistance could be modulated

Safer handling for the chemical surface treatments

Cu polishing VS Nb polishing

Nb Chemical Polishing

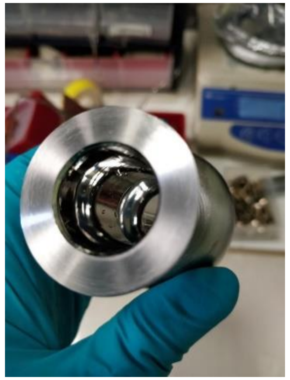
BCP composition (1:1:1 or 1:1:2)

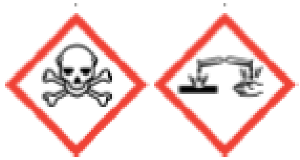
- **HF - Hydrofluoric acid (49%)**
- HNO_3 - Nitric acid (70%)
- H_3PO_4 - Phosphoric acid (85%)

Nb Electrochemical Polishing

EP bath composition (1:9)

- **HF - Hydrofluoric acid (49%)**
- H_2SO_4 - Sulphoric acid (96%)





Cu Chemical Polishing

SUBU5 composition

- sulfamic acid (5g/l)
- hydrogen peroxide 32% (50ml/l)
- n-butanol 99% (50ml/l)
- ammonium citrate (1g/l)

Cu Electrochemical Polishing

EP bath composition (3:2)

- H_3PO_4 - Phosphoric acid (85%)
- N-buthanol (99%)

No HF for Cu polishing

No chemical post treatment on Nb film necessary

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Use materials with poor mechanical properties (but excellent SRF properties)

A15 materials (Nb_3Sn, V_3Si, Nb_3Ge, etc.) present high T_c and High H_{c1} but are very brittle: can not be used as bulk materials for SRF cavities

(WAIT THE NEXT LECTURE)

Material	T_C (K)	ρ_n (μΩcm)	$\mu_0 H_{C1}$ (mT)*	$\mu_0 H_{C2}$ (mT)*	$\mu_0 H_C$ (mT)*	$\mu_0 H_{SH}$ (mT)*	λ (nm)*	ξ (nm)*	Δ (meV)	Type
Pb	7,1		n.a.	n.a.	80		48			I
Nb	**9,22**	**2**	**170**	**400**	**200**	**219**	**40**	**28**	**1.5**	**II**
NbN	**17,1**	**70**	**20**	**15 000**	**230**	**214**	**200-350**	**<5**	**2.6**	**II**
NbTi			4-13	>11 000	100-200	80-160	210-420	5,4		
NbTiN	17,3	35	30				150-200	<5	2.8	II
Nb_3Sn	**18,3**	**20**	**50**	**30 000**	**540**	**425**	**80-100**	**<5**	**<5**	**II**
Mo_3Re	15	10-30	30	3 500	430	170	140			II
MgB_2	**39**	**0.1-10**	**30**	**3 500**	**430**	**170**	**140**	**5**	**2.3/7.2**	**II- 2gaps****
2H-$NbSe_2$	7,1	68	13	2680-15000	120	95	100-160	8-10		II- 2gaps**
YBCO/Cuprates	**93**		**10**	**100 000**	**1400**	**1050**	**150**	**0,03/2**		**d-wave****
Pnictides $Ba_{0.6}K_{0.4}Fe_2As_2$	**38**		**30**	**>50000**	**900**	**756**	**200**	**2**	**10-20**	**s/d wave****

* @ 0K
** 2D => orientation problems ?

C. Antoine (CEA Saclay), SRF Tutorials 2019

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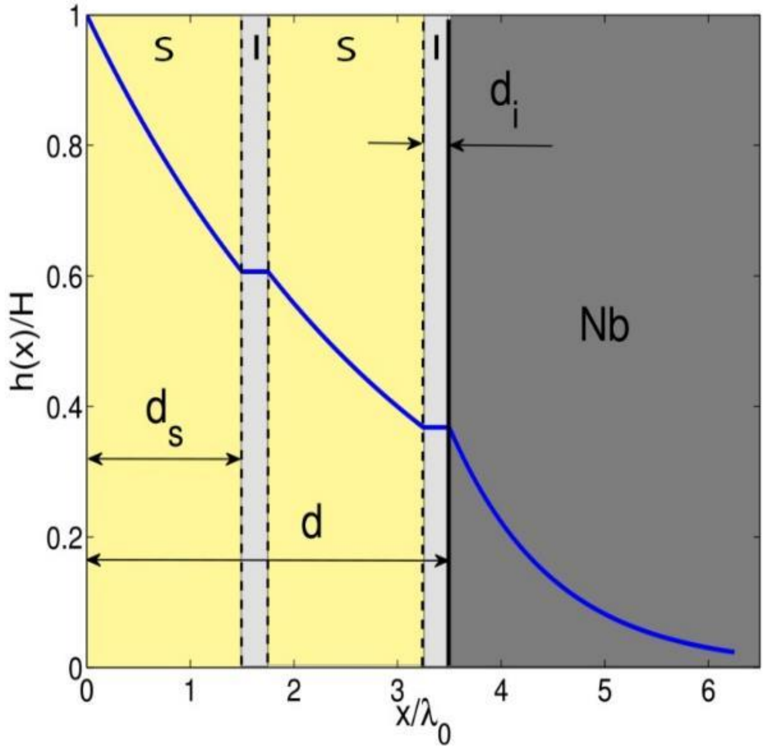
Realize complex structures

SIS Multilayer

(WAIT THE NEXT LECTURE)



Alex Gurevich, Appl. Phys. Lett. 88, 012511 (2006)

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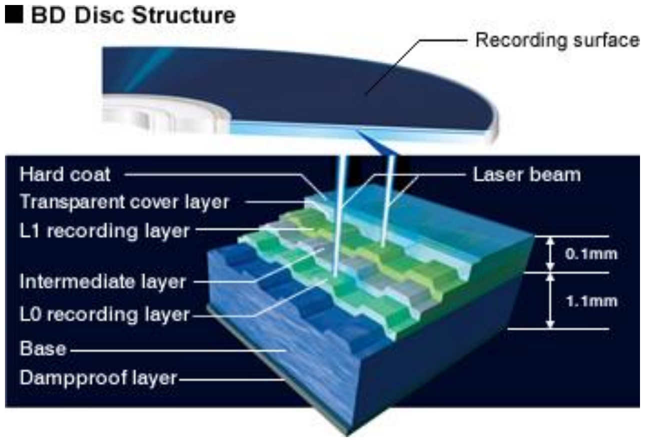
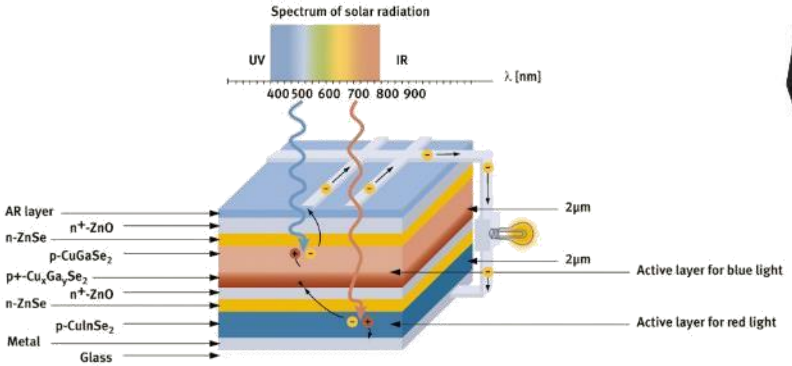
Thin film applications

Some of the most utilized applications of thin film deposition processes include:

- Single and multilayer films and coatings
- Nanolayered materials
- Optical films for transmission and reflection
- Decorative films
- Decorative and wear-resistant (decorative/functional) coatings
- Permeation barriers for moisture and gases
- Corrosion-resistant films
- Electrically insulating layers for microelectronics
- Coating of engine turbine blades
- Coating of high strength steels to avoid hydrogen embrittlement
- Diffusion barrier layers for semiconductor metallization
- Magnetic films for recording media
- Transparent electrical conductors and antistatic coatings
- Wear and erosion-resistant (hard) coatings (tool coatings)
- Dry film lubricants
- Composite and phase-dispersed films and coatings
- Nanocomposite materials
- Thin-walled freestanding structures and foils

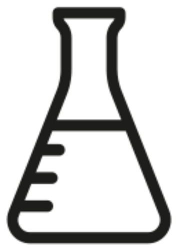




Thin film deposition techniques

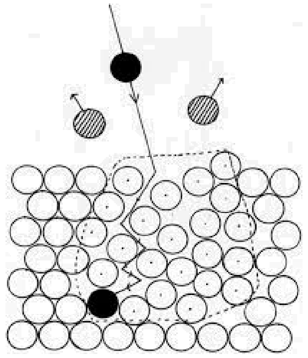
Chemical Deposition

- **Plating/Electroplating**
- **Dip/Spin coating**
- **Chemical Vapour Deposition**
  - Thermal CVD
  - Plasma Enhanced CVD
  - Atomic Layer Deposition (ALD)



Phisical Deposition

- **Physical Vapour Deposition**
  - Evaporation
  - Laser Ablation
  - Plasma Spray
  - Sputtering
  - Cathodic Arc



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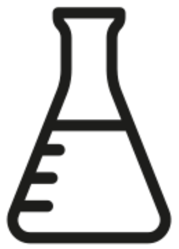

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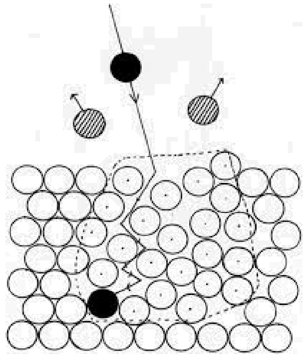


PVD deposition techniques

Physical vapor deposition processes (often just called thin film processes) are atomistic deposition processes in which material is vaporized from a solid or liquid source in the form of atoms or molecules and transported in the form of a vapor through a vacuum or low pressure gaseous (or plasma) environment to the substrate, where it condenses

Donald M. Mattox,
Handbook of Physical Vapor Deposition PVD) Processing

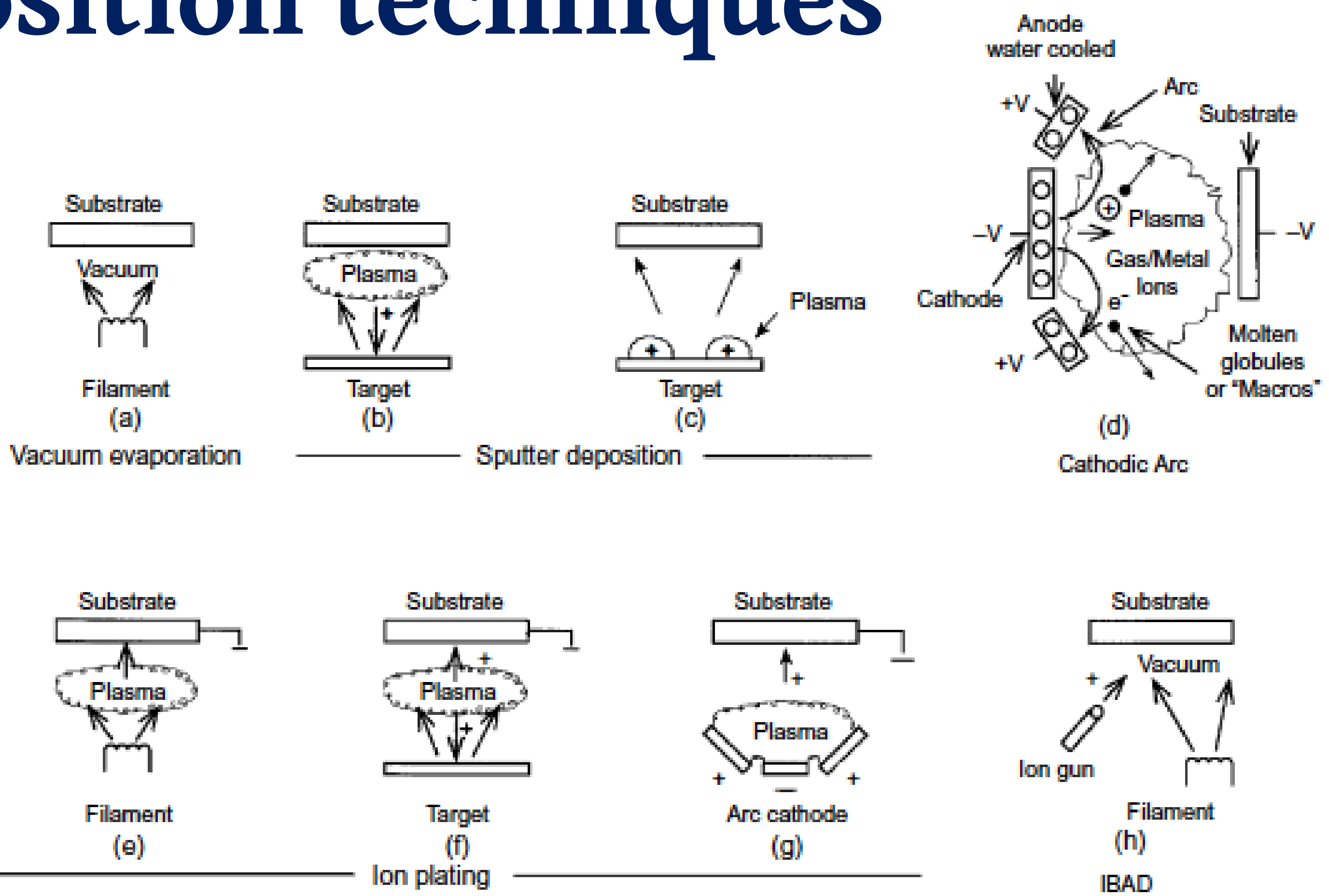
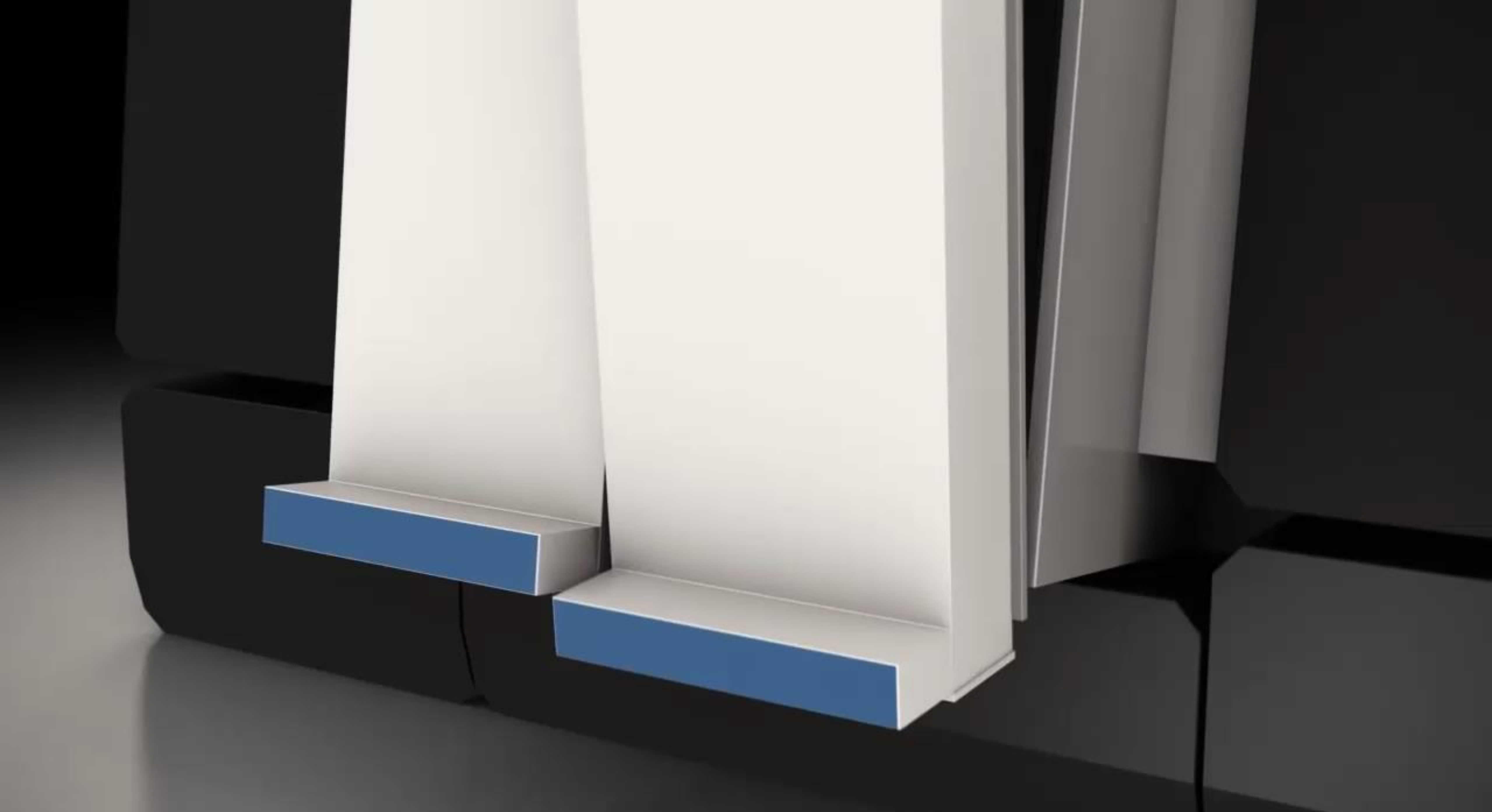


Figure 1.1: PVD Processing Techniques: (a) Vacuum Evaporation, (b) and (c) Sputter Deposition in a Plasma Environment, (d) Sputter Deposition in a Vacuum, (e) Ion Plating in a Plasma Environment with a Thermal Evaporation Source, (f) Ion Plating with a Sputtering Source, (g) Ion Plating with an Arc Vaporization Source, and (h) Ion Beam-Assisted Deposition (IBAD) with a Thermal Evaporation Source and Ion Bombardment from an Ion Gun



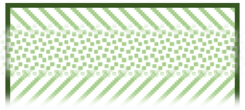
Plansee High Performance Materials, Youtube

Interface

The depositing film material may diffuse and react with the substrate to form a "interfacial region"

Nb-Cu case

Abrupt



Graded

Weak chemical reaction between atoms and substrate

Low deposition temperature

Surface contamination

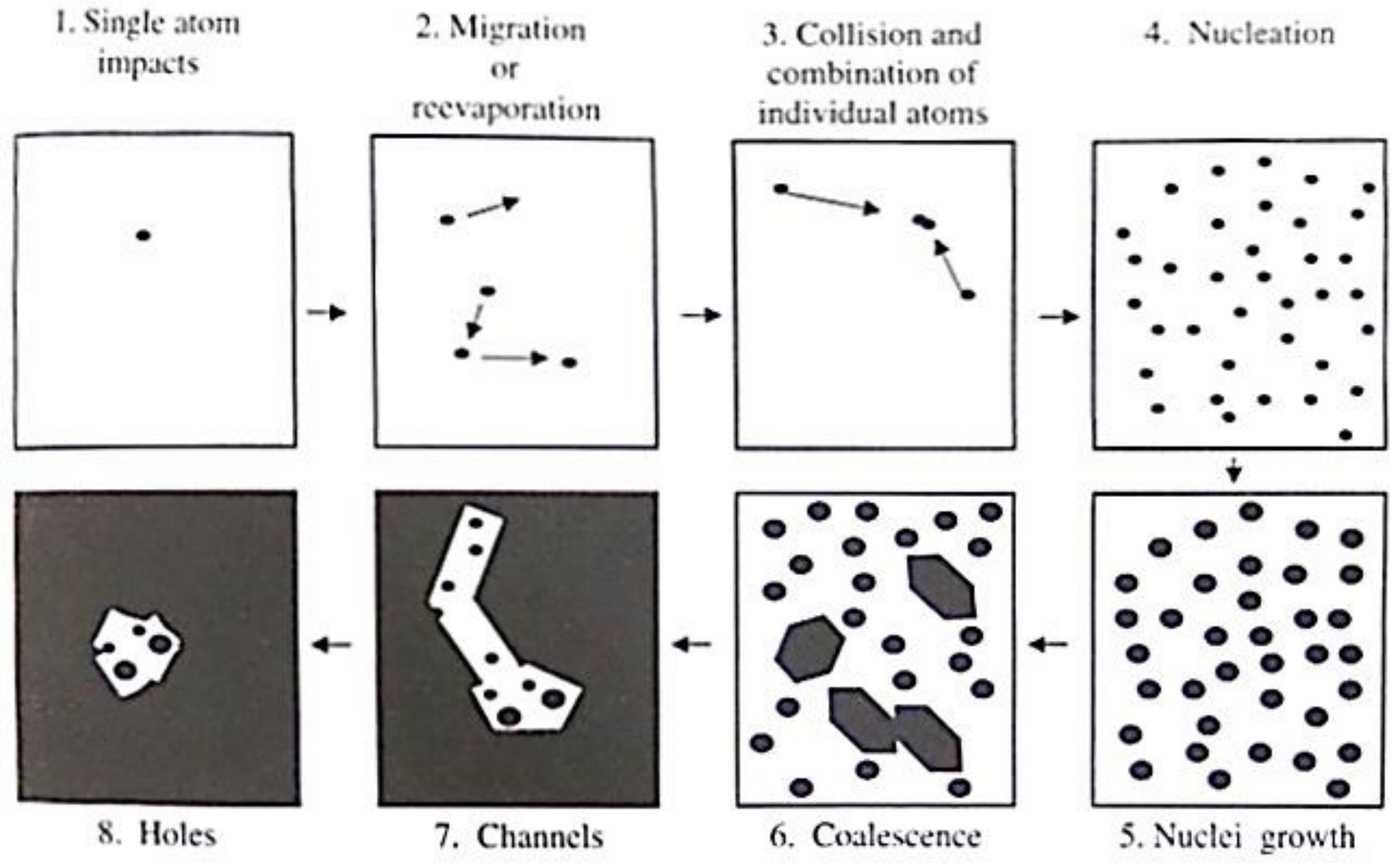
Low nucleation density

By diffusion (solubility, temperature, time, contaminations)

Chemical reaction (oxygen-active metals on oxide substrates)

By co-deposition or implantation of energetic ions of the material

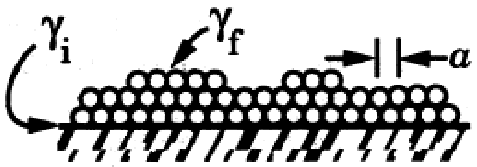
Nucleation stages



Phases of film growth: Nuclei growth

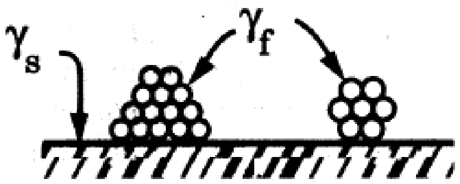
Nuclei grow by collecting adatoms which either impinge on the nuclei

or migrate over the surface



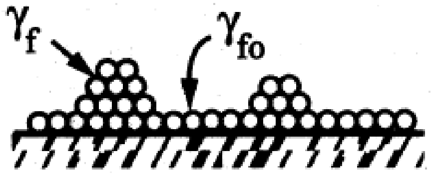
binding energy atom-atom **<** binding energy atom-surface

Layer by layer growth *(Frank-van der Merwe)*



binding energy atom-atom **>** binding energy atom-surface

Island growth *(Volmer-Weber)*



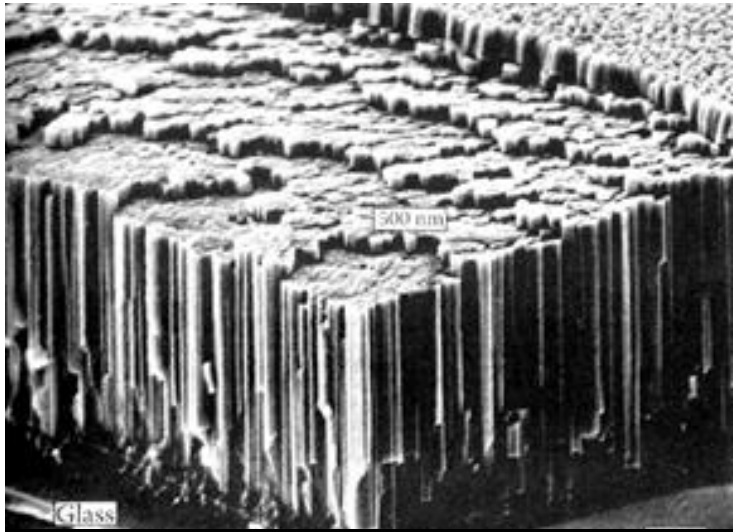
binding energy atom-atom **=** binding energy atom-surface

Layer by layer + island growth *(Stranski-Krastanov)*

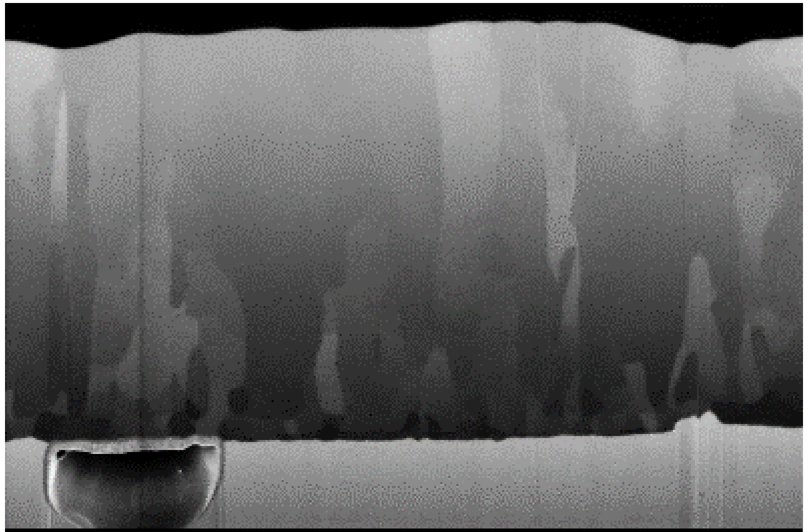
Phases of film growth: Film growth

Is the evolution of the nucleation, where arriving atoms are deposited on the previously deposited material

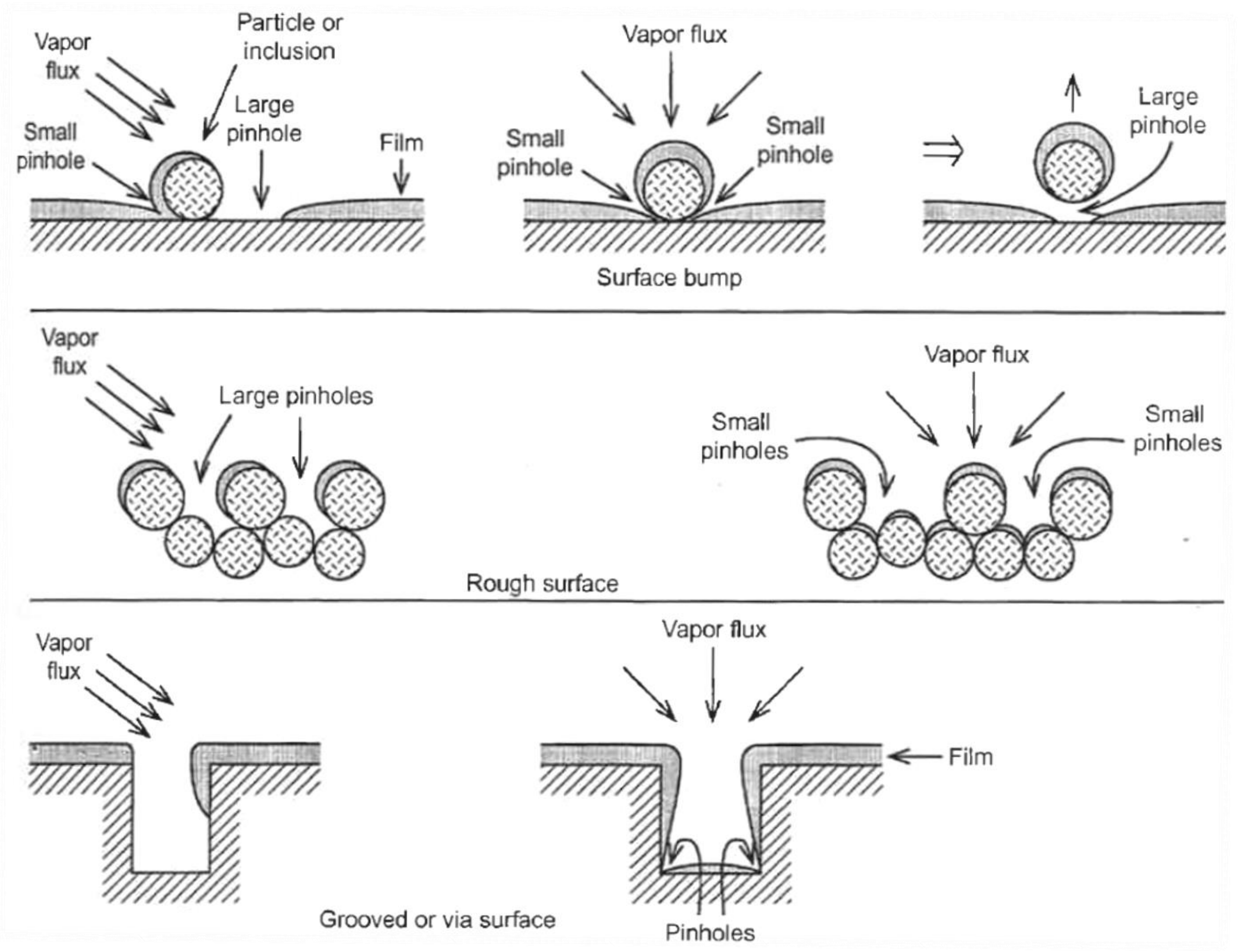
Usually exhibits a columnar morphology



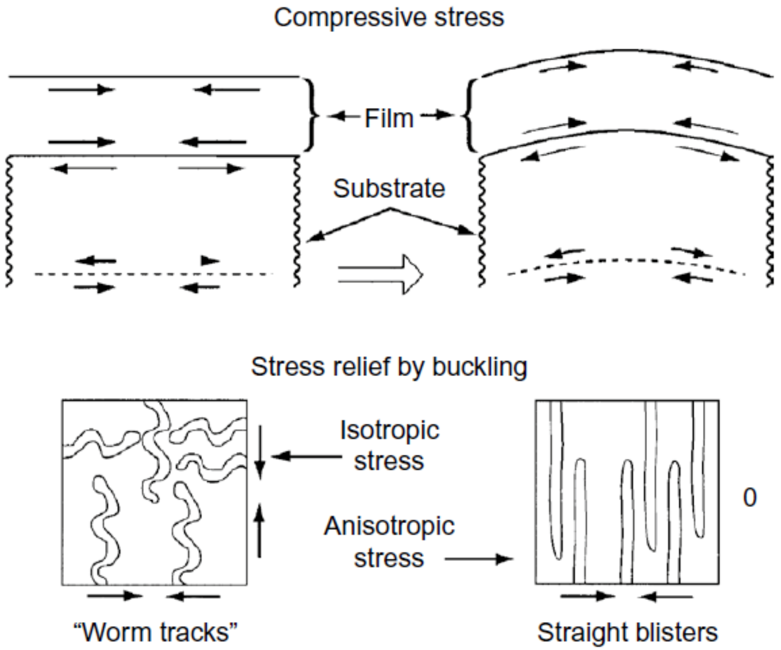
TiN on glass from Macleod's book

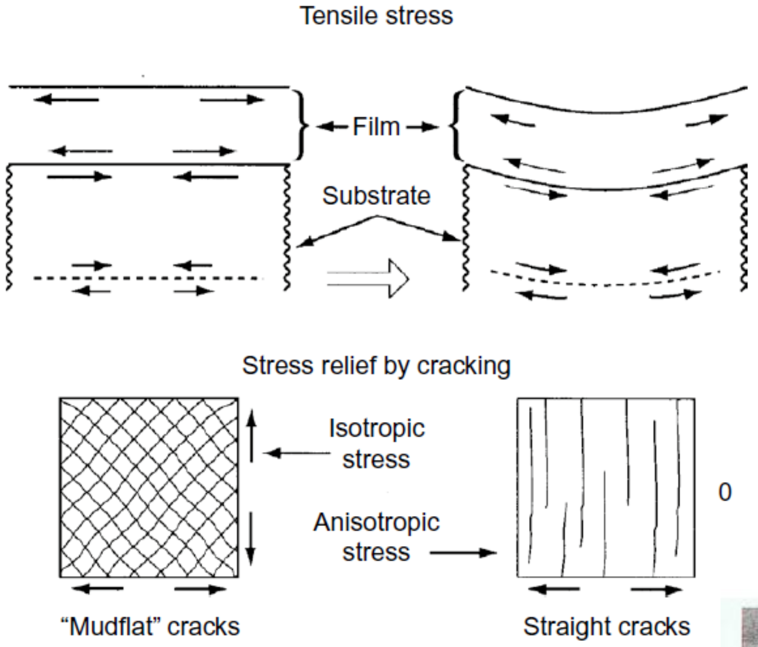

Nb on Cu (A. Sublet, CERN)

On growth and adhesion



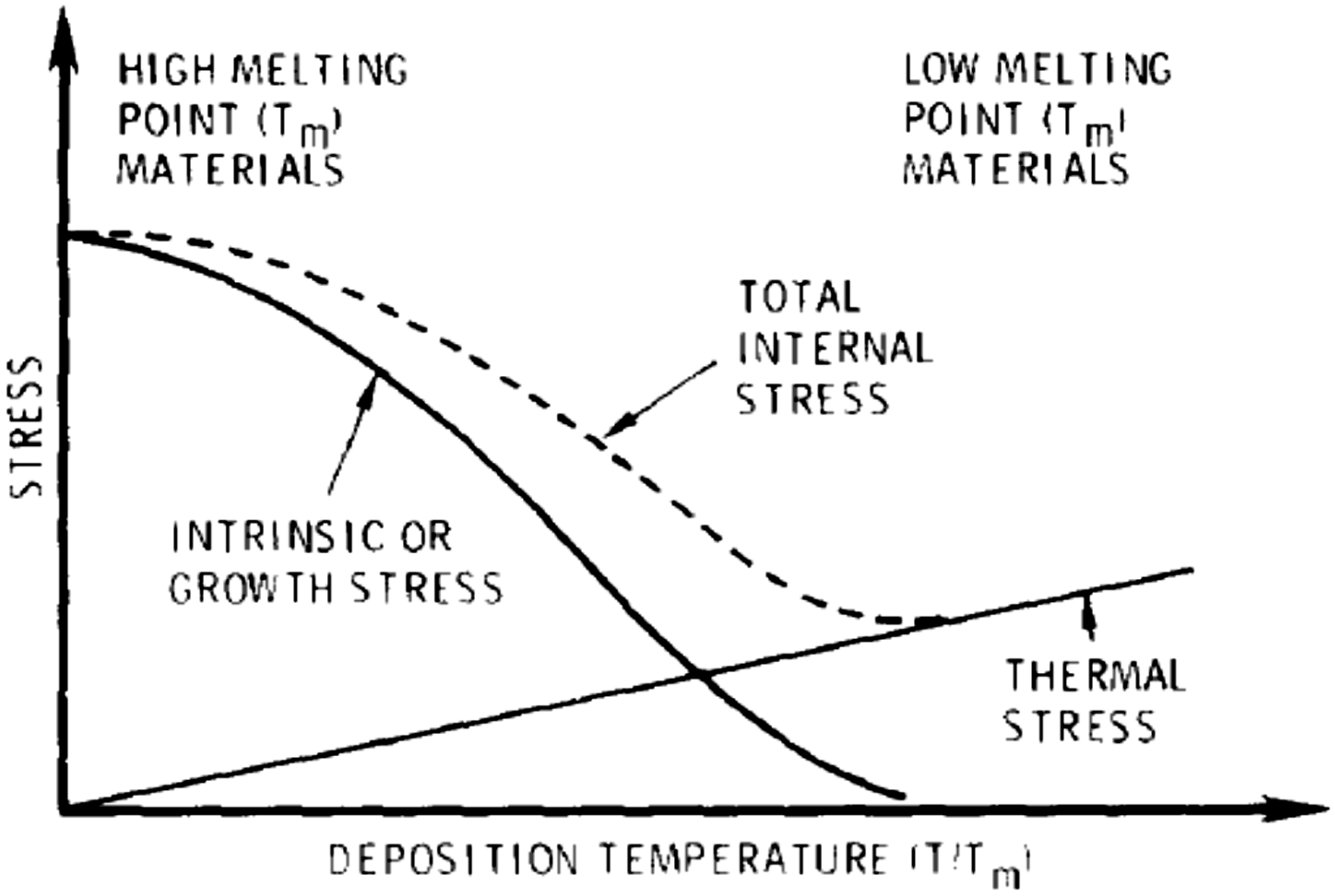
Stress on thin films





Donald M. Mattox, Handbook of Physical Vapor Deposition PVD) Processing

How to reduce film stress? Temperature effect



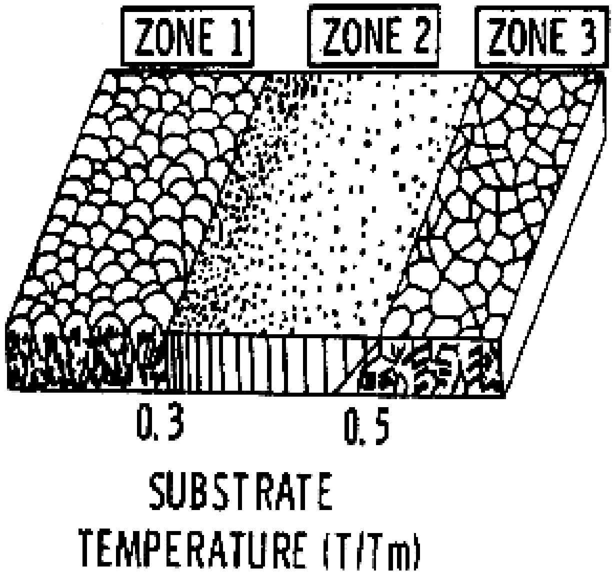
J. A. Thornton and D. W. Hoffman, "Stress-related effects in thin films," *Thin Solid Films*, 1989

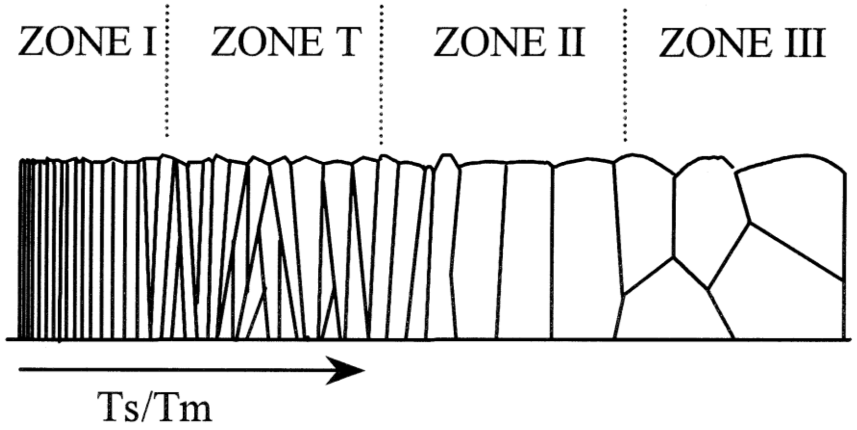
Temperature effect and Structure Zone Diagram (SZD)

- Based on the compilation of the experimental results, it is a guideline for "predicting" the structure of deposited thin films

- 1st proposed in 1969 by Movchan & Demchishin for films deposited by thermal evaporation.

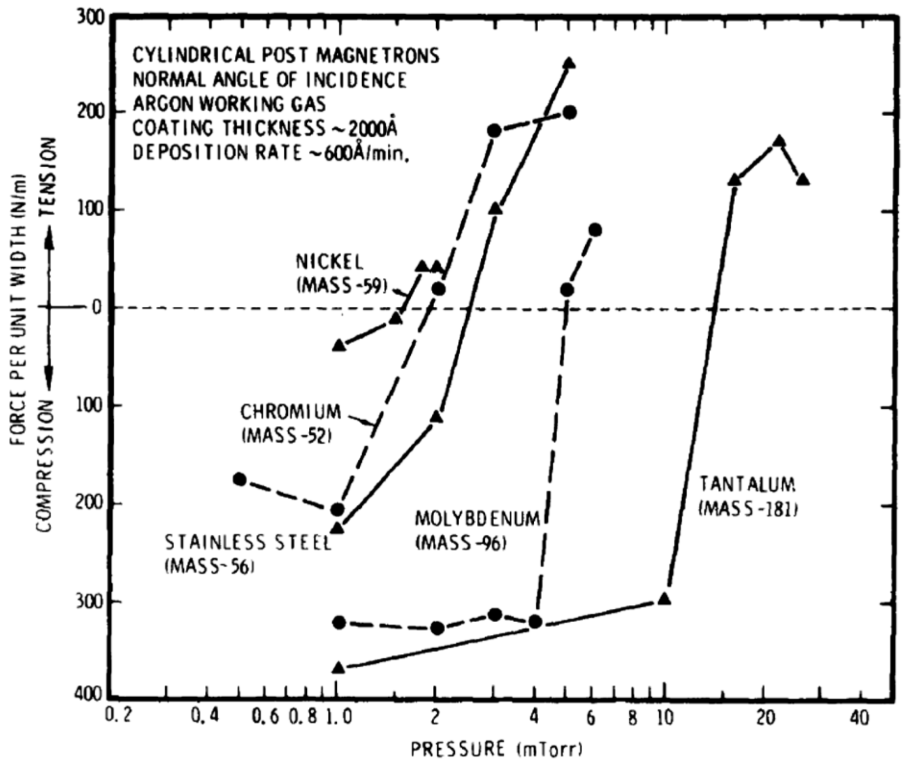
$$T_h = \frac{T}{T_m}$$ Homologous temperature





How to reduce film stress? Pressure effect

ZERO STRESS PRESSURE



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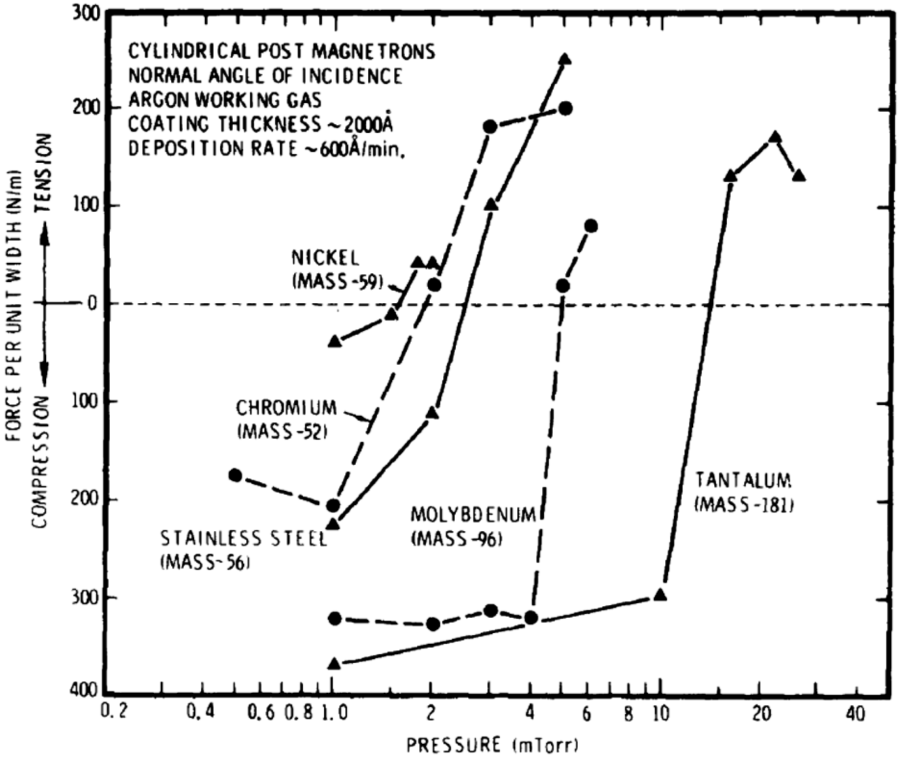

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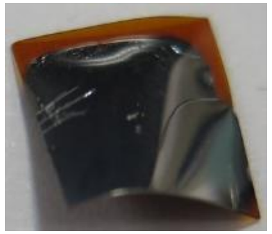
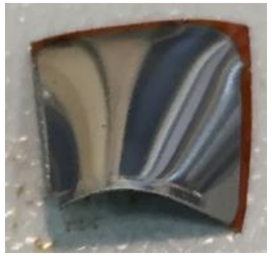
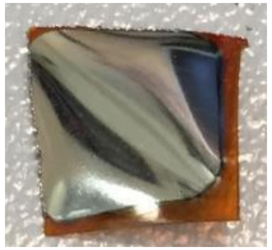


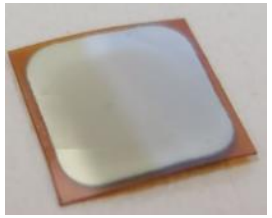
$7 \cdot 10^{-3}$ mbar



$9 \cdot 10^{-3}$ mbar



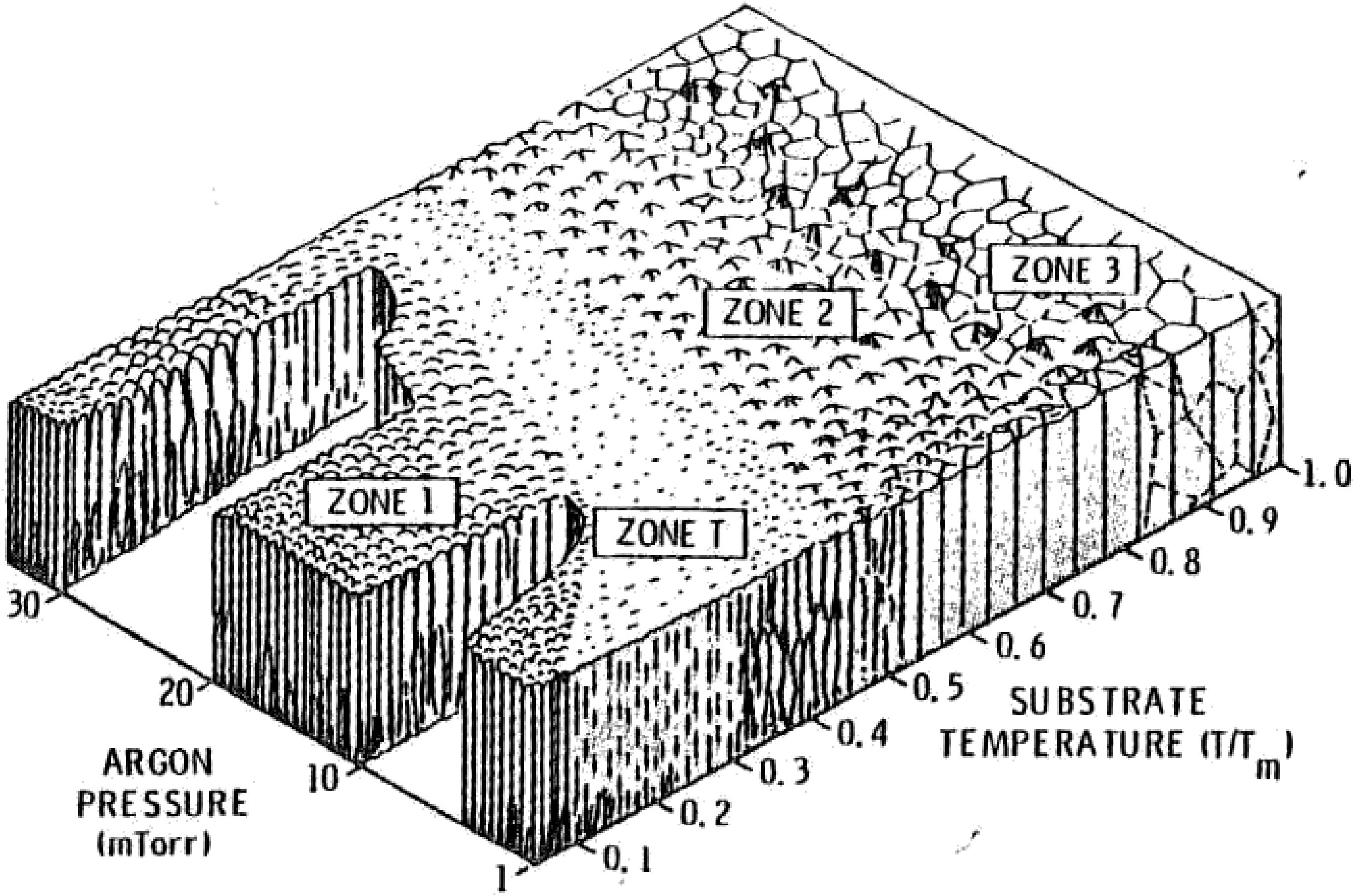
$2 \cdot 10^{-2}$ mbar



$5 \cdot 10^{-2}$ mbar

Thorton Structure Zone Diagram

- ZONE 1: characterized by a fine-grained structure of textured and fibrous grains, pointing in the direction of the arriving vapor flux. The morphology is caused by the low mobility of the adatoms that produce a continued nucleation of grain.
- ZONE T: a dense fibrous structure with a smooth, highly reflective surface. Diffusion is “remarkable” but grain boundary diffusion is strongly limited. Ionic bombardment of the growing film can move the morphology from zone 1 to zone T.
- ZONE 2: surface diffusion sets in, leading to uniform columnar grains.
- ZONE 3: dense films with large grains, drive by bulk diffusion and recrystallization.



J.A. Thornton and D.W. Hoffman, Thin Solid Films, vol. 171, no. 1, pp. 5–31, 1989

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Nb film is a well known technology

- 288 Nb/Cu cavities installed in LEP @ CERN
- 56 Nb/Cu cavities installed in ALPI @ LNL INFN
- 16 Nb/Cu cavities installed in LHC @ CERN
- 20 Nb/Cu cavities installed in HIE-ISOLDE @ CERN

- R&D in many different labs: CERN, INFN, JLAB, STFC, Cornell, IMP, …


90’s LEP2: 350MHz 4-cells


1998-2004 ALPI: 160 MHz QWR


2000’s LHC: 400MHz 1-cell


2010’s HIE-ISOLDE: 100MHz QWR

LEP2 @CERN

The first prototype niobium–copper superconducting cavity, pictured in December 1983 with Nadia Circelli, **Cristoforo Benvenuti**, Jacques Genest and Max Hauer. Image credit: CERN

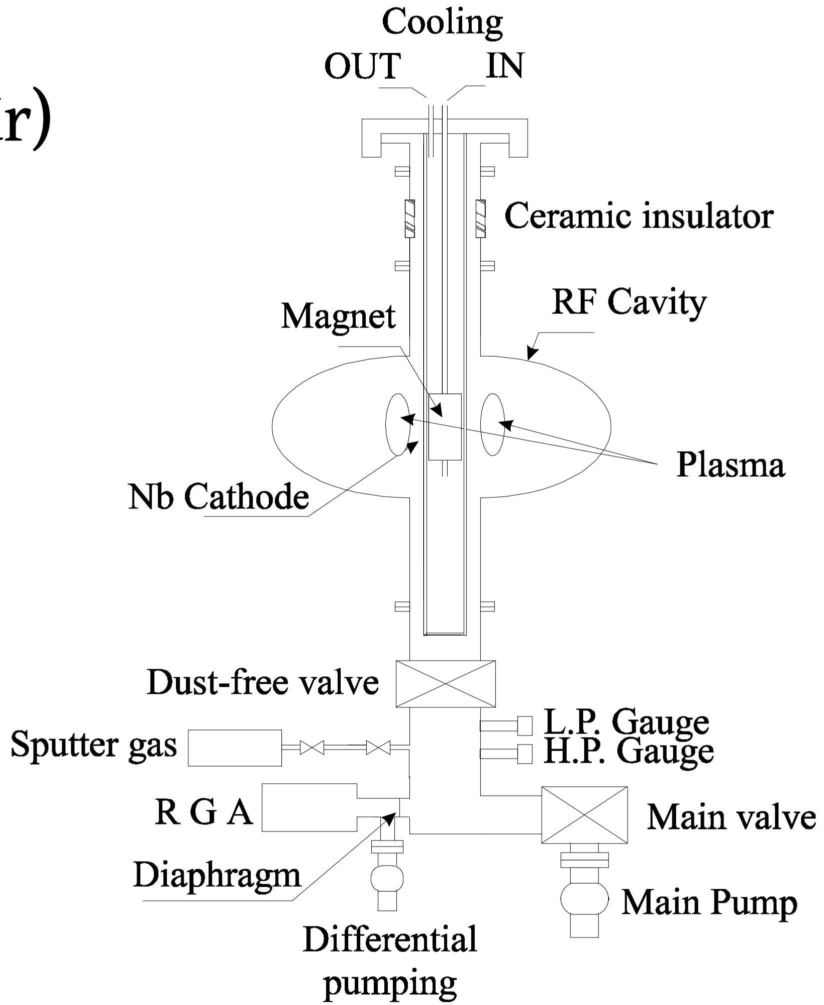
Elliptical cavities @CERN - LEP2, LHC and 1,5 GHz R&D

SPUTTERING PARAMETERS (1,5 GHz)

- Sputter gas pressure of 1.5×10^{-3} mbar (Ar or Kr)
- Plasma current stabilized at 3A – DC
- Sputter potential ~ -360 V
- **Coating temperature is 150 °C**
- Thickness: 1.5 µm

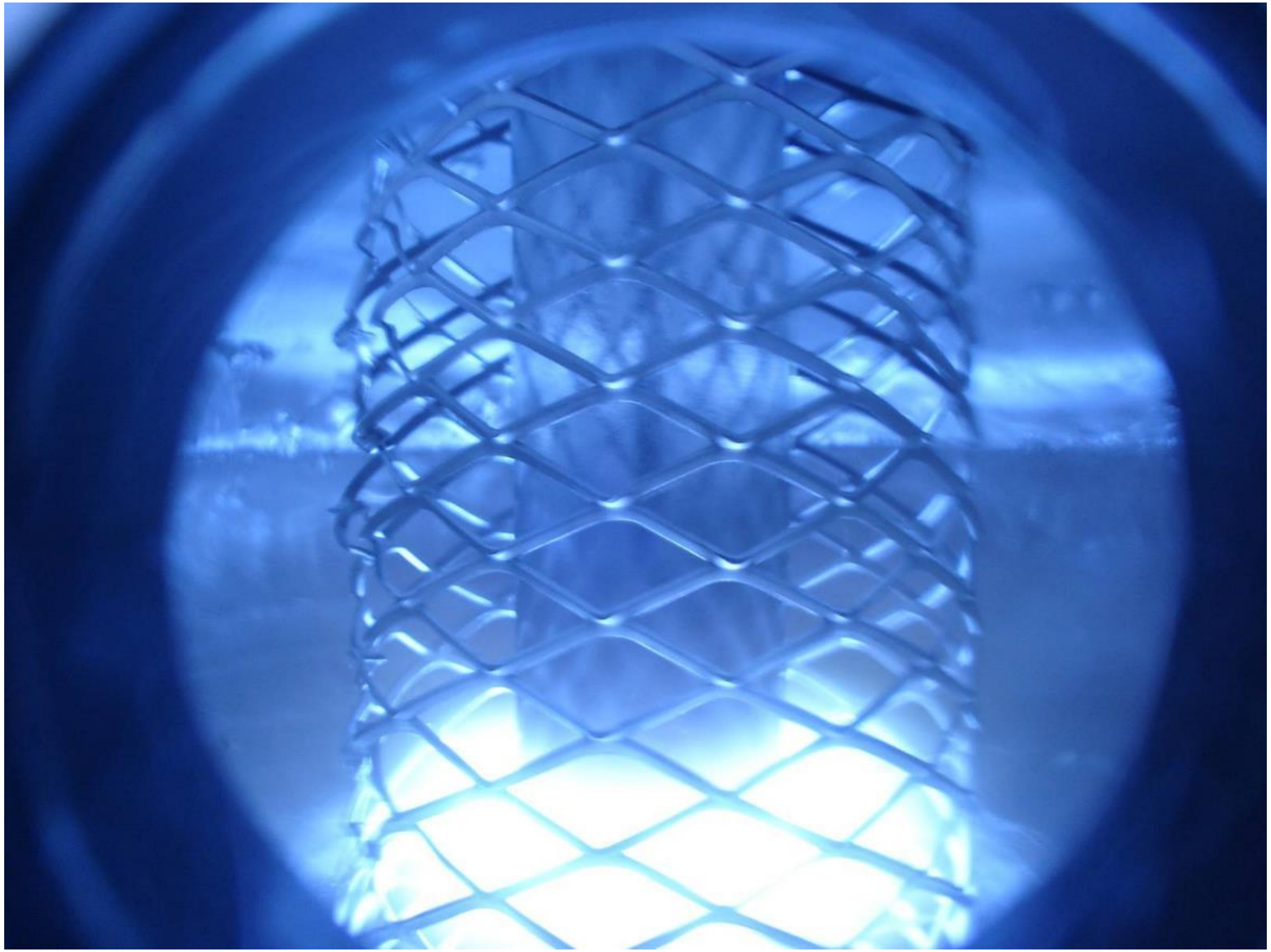
SPUTTERING PARAMETERS (1,5 GHz)

- RRR: 11.5 ± 0.1
- Argon content: 435 ± 70 ppm
- Grain size: 110 ± 20 nm
- **Tc: 9.51 ± 0.01 K**

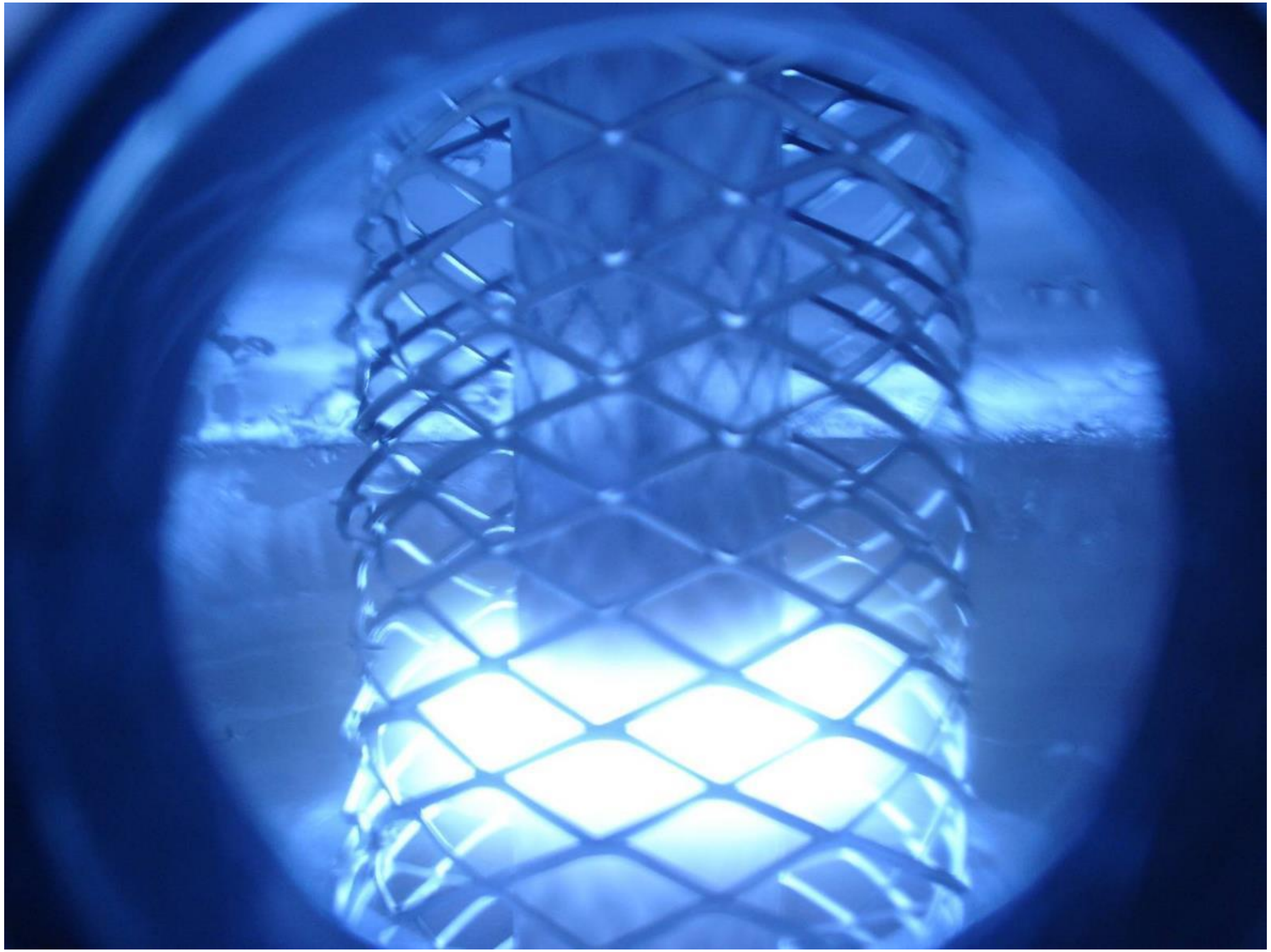


Courtesy of S. Calatroni (CERN)

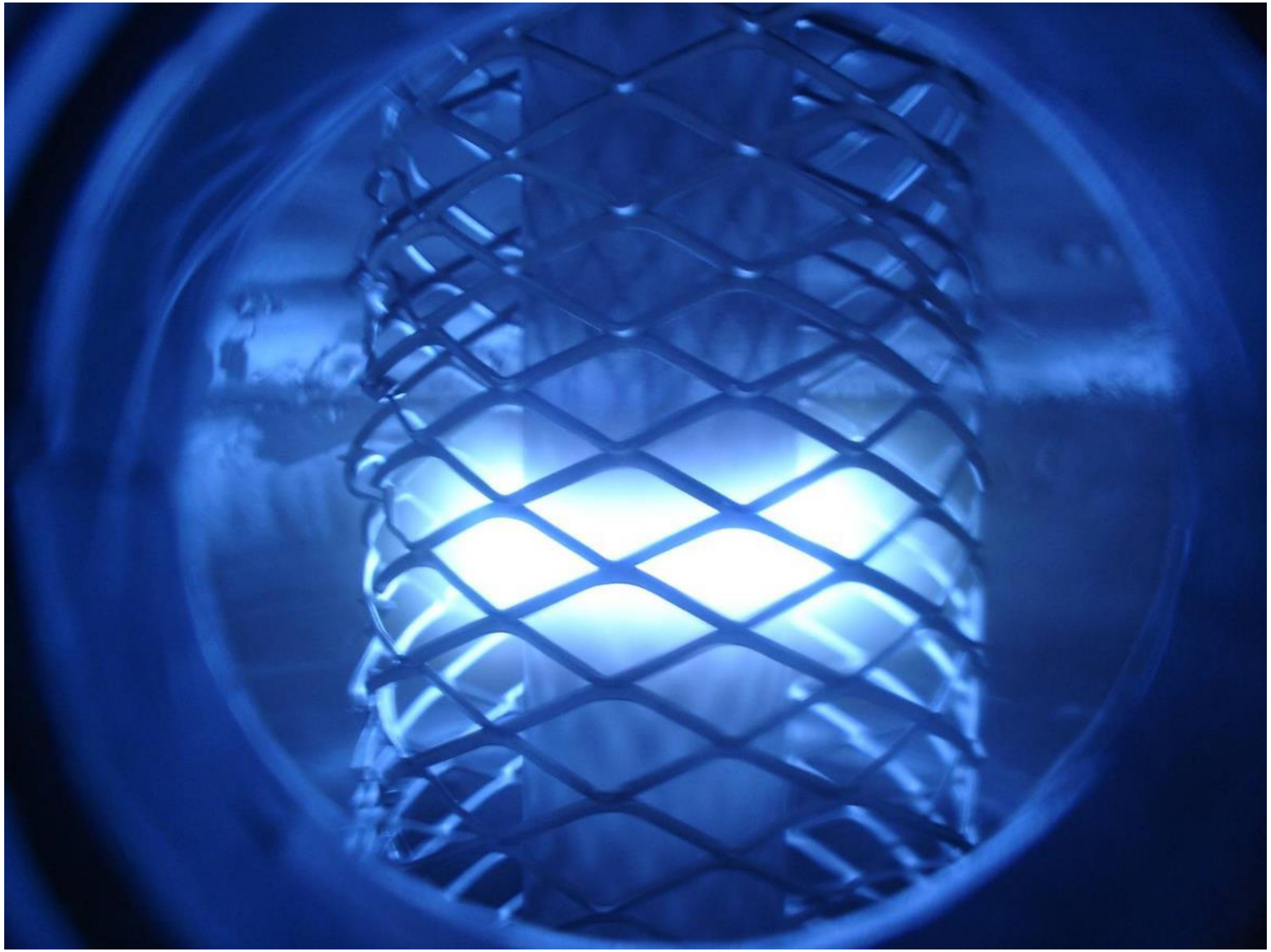
1.5 GHz Cavity Sputtering System @LNL (CERN like)



1.5 GHz Cavity Sputtering System @LNL (CERN like)



1.5 GHz Cavity Sputtering System @LNL (CERN like)



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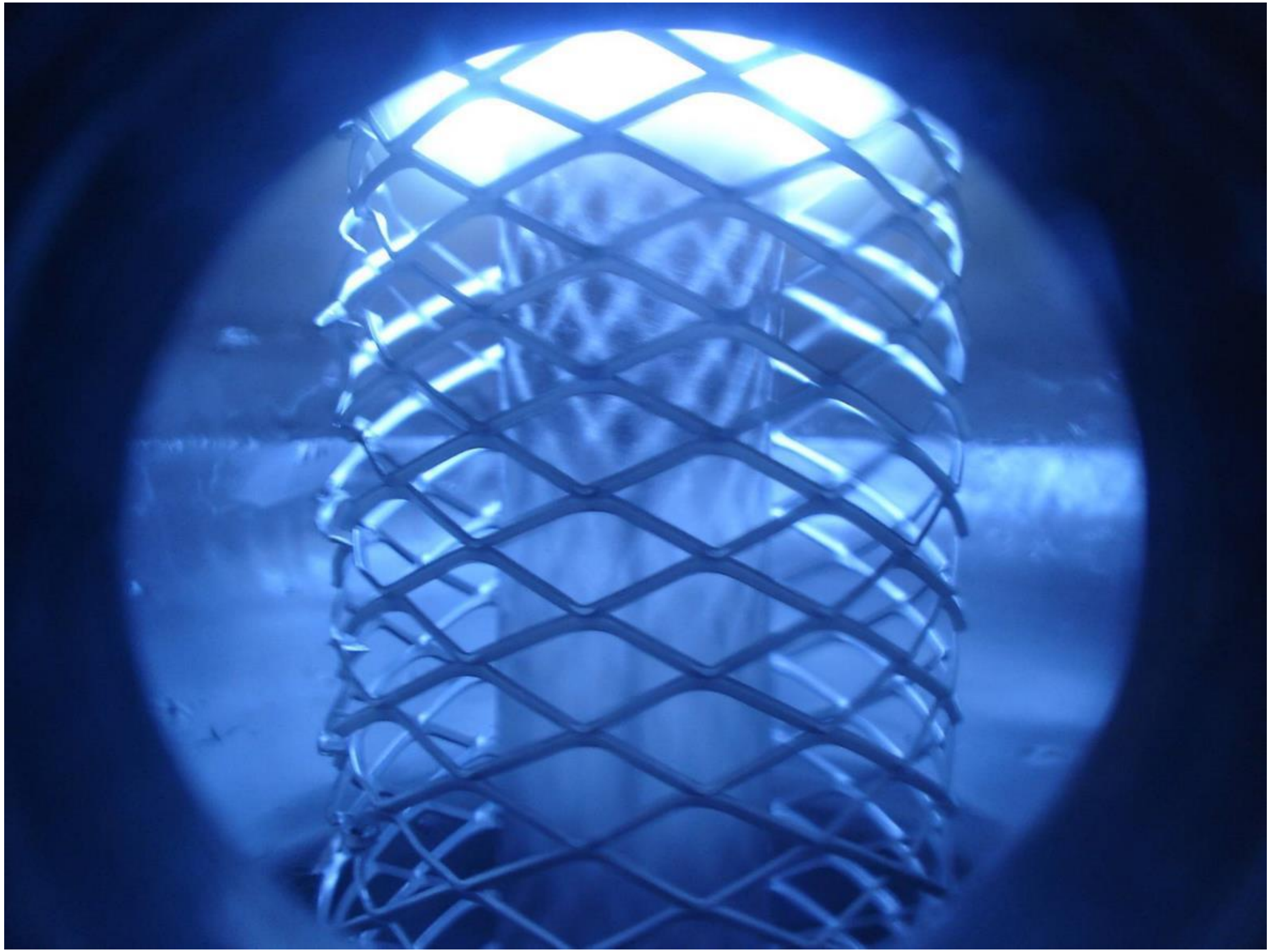
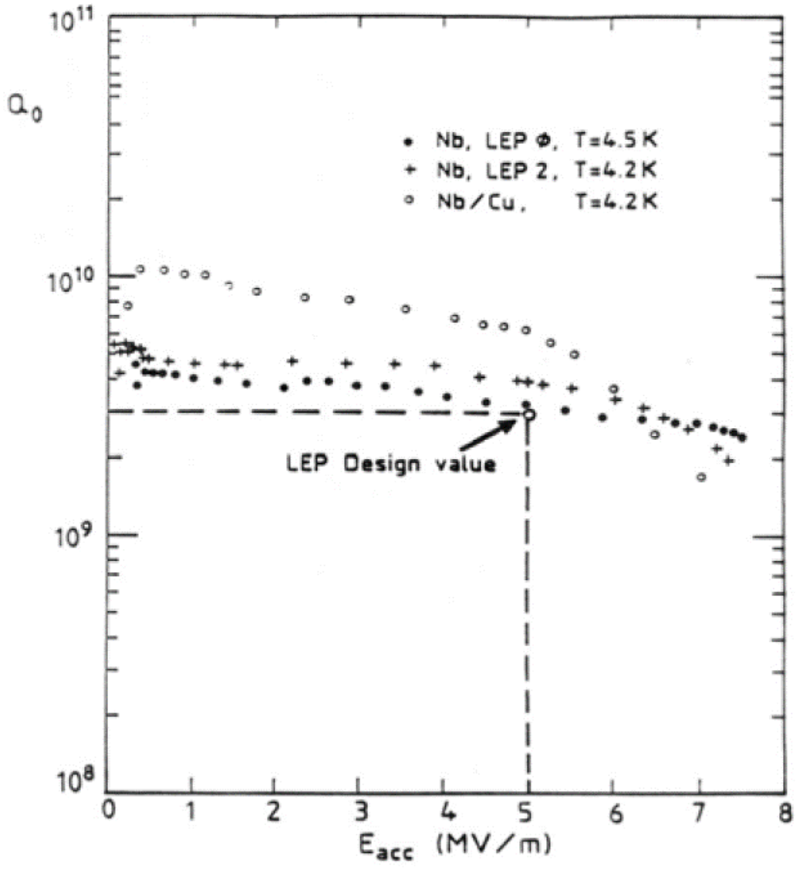


LEP2 Performances



G. Arnolds-Mayer et al. 1988 Proc. of the 3rd Workshop on RF Superconductivity

Eight pre-series 4-cell cavities for LEP were built at CERN, the remaining 264 were **made by three European industrial suppliers**

No thermal quench (contrary to bulk Nb)

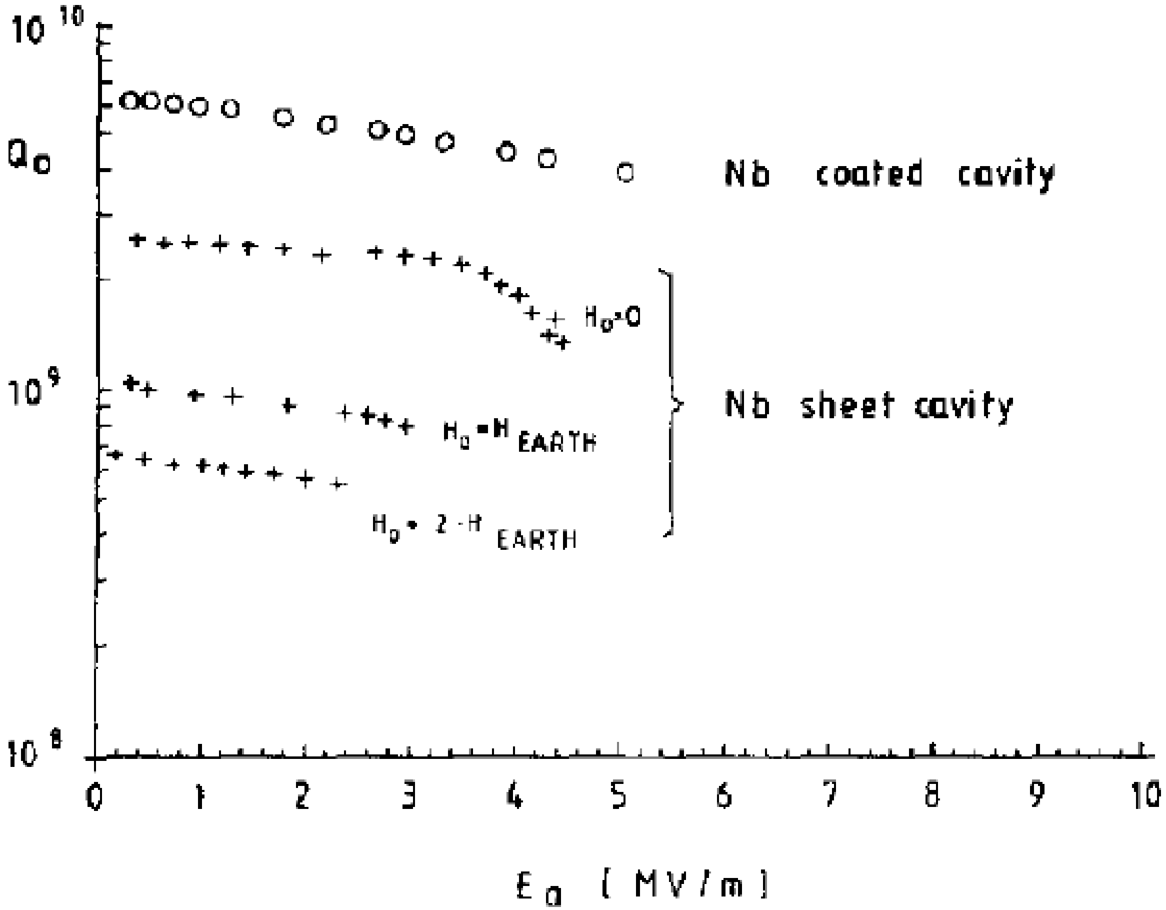
Higher performances compare to bulk Nb

Nb bulk cavities performance in the eighties were limited by the poor Nb thermal conductivity (RRR of 40)

Unexpected advantage

Almost insensitive to the Earth's magnetic field

- Bulk Nb: 100 nΩ/Gauss
- Nb films: 1 nΩ/Gauss



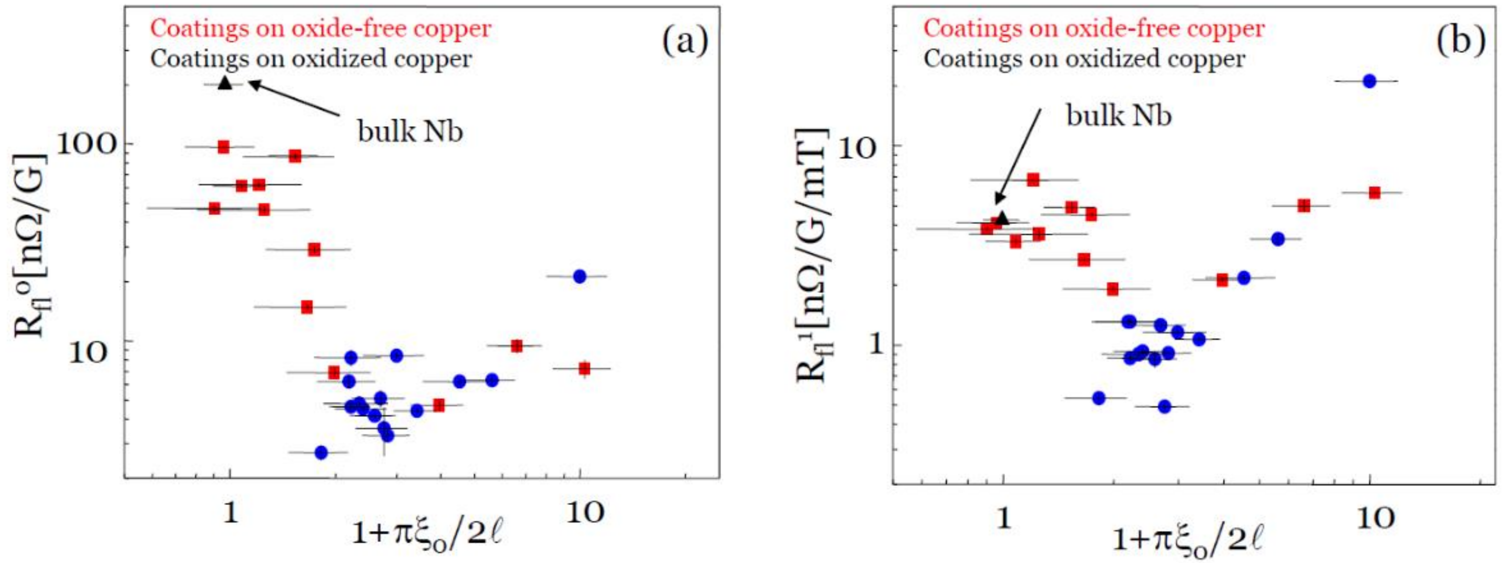
cheaper cryostats

Not necessary complex magnetic shielding of the cavities

C. Benvenuti et al. I Physica B 197 (1994)

Almost insensitive to the Earth's magnetic field

- Bulk Nb: 100 nΩ/Gauss
- Nb films: 1 nΩ/Gauss



cheaper cryostats

Not necessary complex magnetic shielding of the cavities

$$R_{fl} = (R_{fl}^{0} + R_{fl}^{1} H_{RF})\, H_{ext}$$

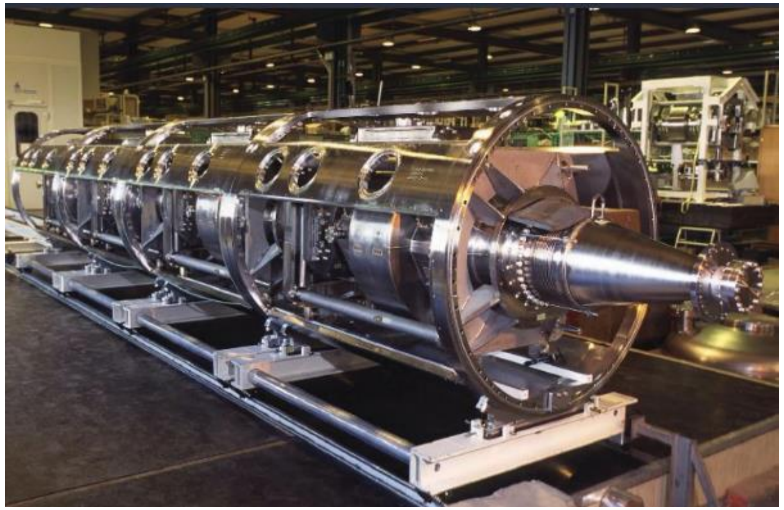
C. Benvenuti et al., Physica C 316 (1999) 153–188

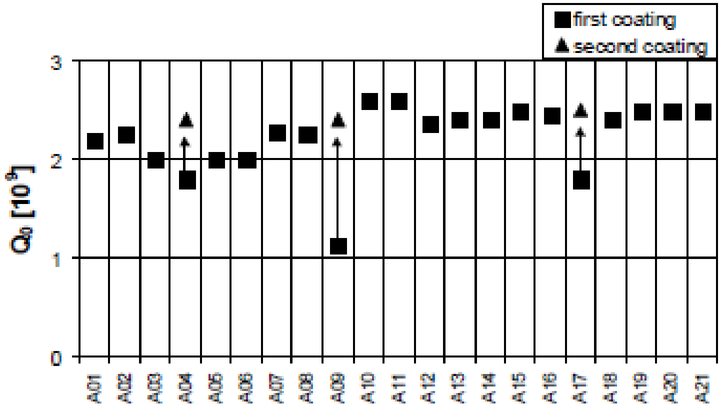
LHC cavities: from substrate fabrication to installation in the tunnel

LHC Point 4, ~ 100 m deep shaft → LHC tunnel

Courtesy of A. Sublet (CERN)

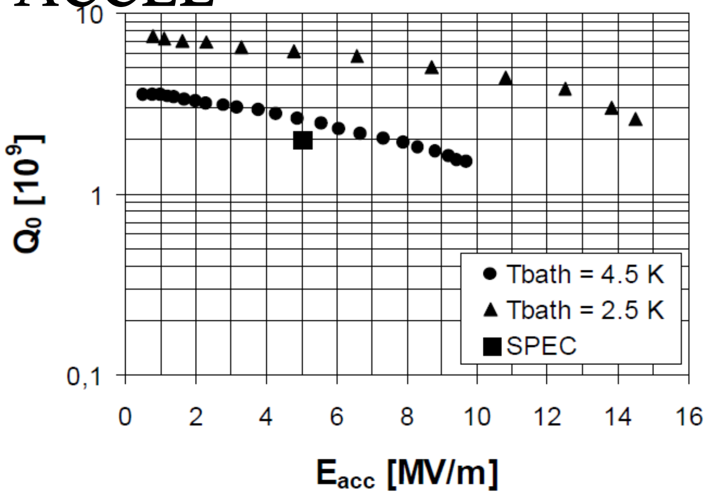
LHC

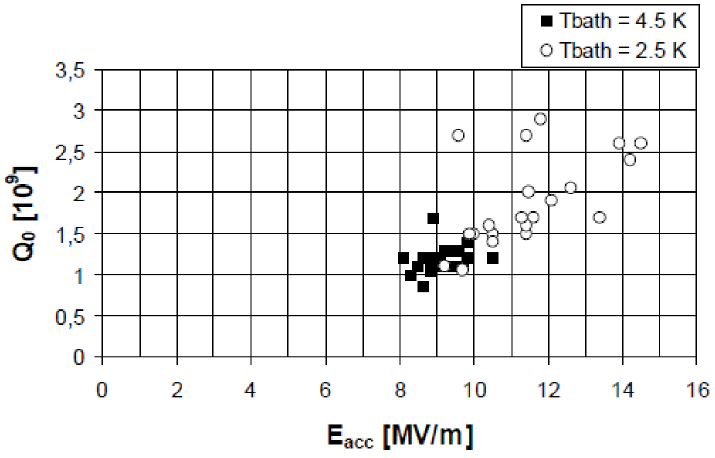




Distribution of cavity quality Q0 at a gradient of Eacc = 5 MV/m and a bath temperature of 4.5 K

21 cavities 400 MHz cavities produced at ACCEL

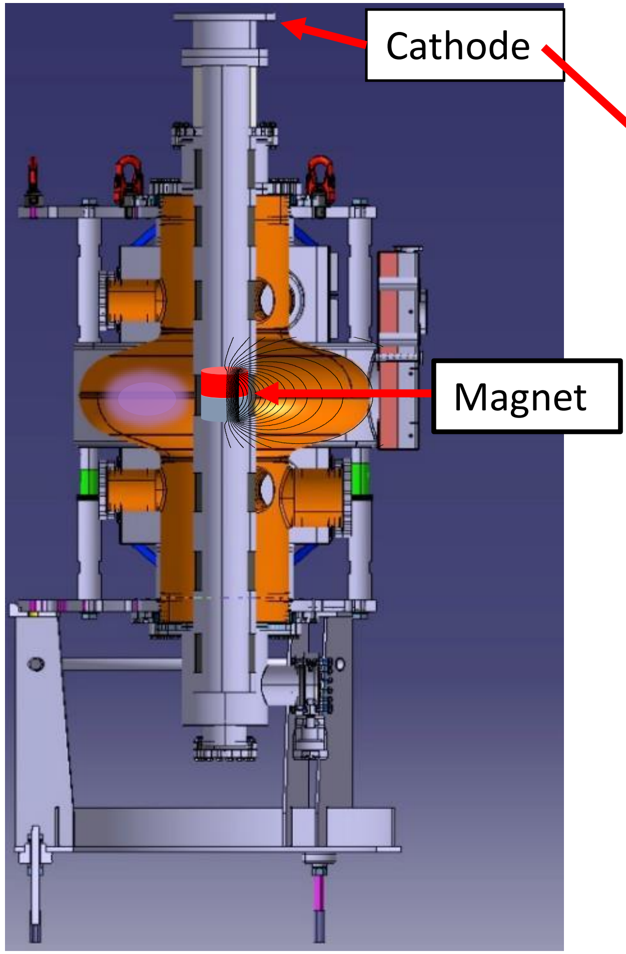




Highest gradients Eacc and quality factors Q0 at the highest gradients achieved at bath temperatures of 4.5 K and 2.5 K

S. Bauer *et al.*, "Production of Nb/Cu sputtered superconducting cavities for LHC," 1999

LHC cavities coating setup



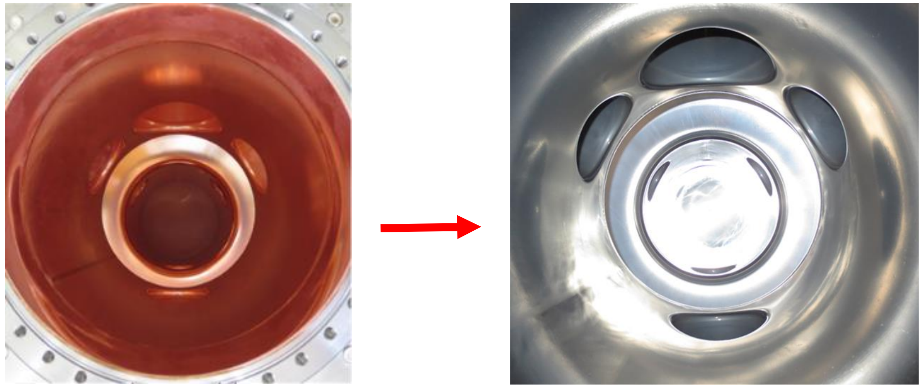
LHC cavity coating setup

- **Cavity as UHV chamber** (10^{-10} mbar base vacuum)
- Cavity = anode, grounded
- Nb cylindrical cathodes tubes
- **movable electromagnet inside**, liquid cooled

→ **DC-magnetron sputtering, 6 kW, 1.10^{-3} mbar Kr**

→ Cavity bake-out (bake-out tent) to 180°C
→ Coating 7 steps for the 7 different electromagnet positions
→ Duration = 1h 20' at low temperature (150°C)
→ Nb layer thickness ~ 2 mm

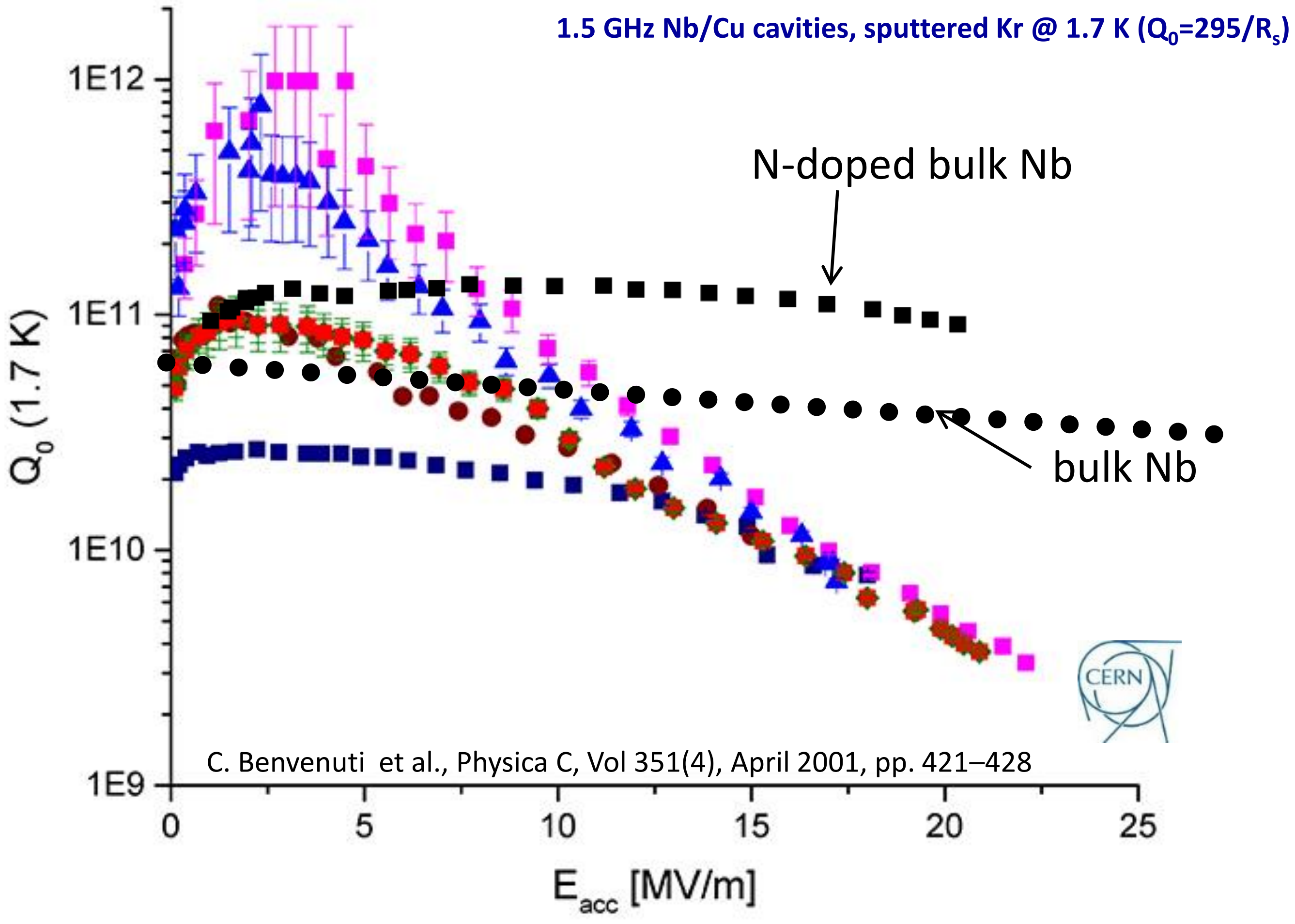
8 spare cavities produce

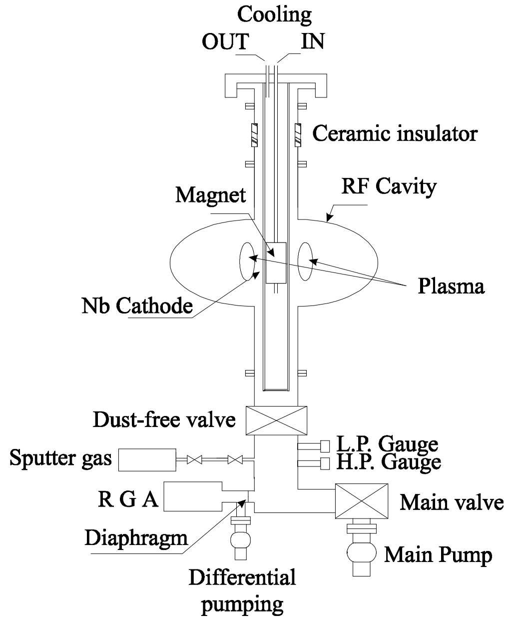


Before and after coating views from cavity main aperture

A. Sublet, Thinfilms Workshop 2018

R&D on 1,5 GHz - State of the art for Nb-Cu cavities





High Q at low field

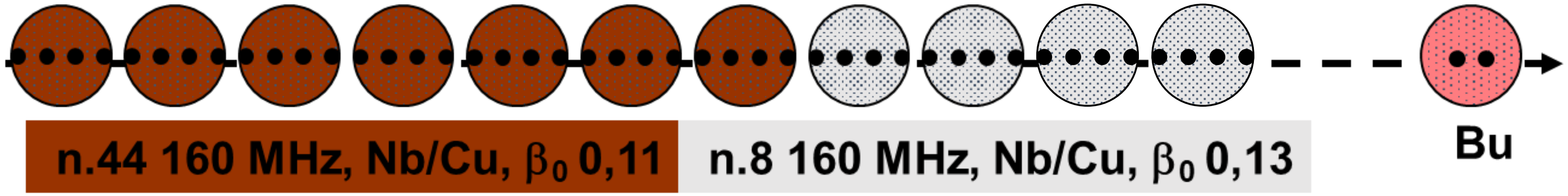
Strong Q-slope still present

Increasing complexity: Quarter Wave Resonator

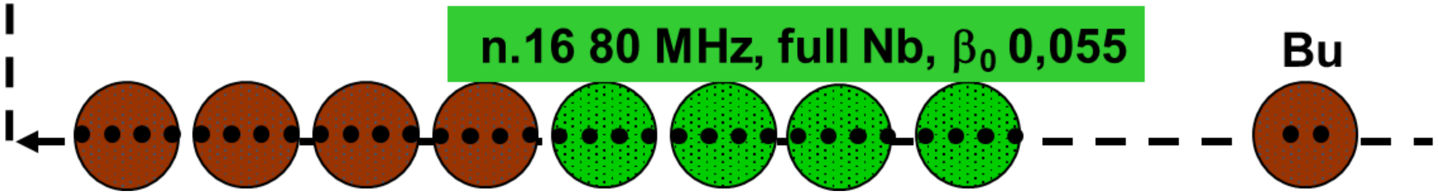
ALPI @ LNL – 160 MHz QWR

A more complex geometry than elliptical cavities

QWR in ALPI @ LNL INFN



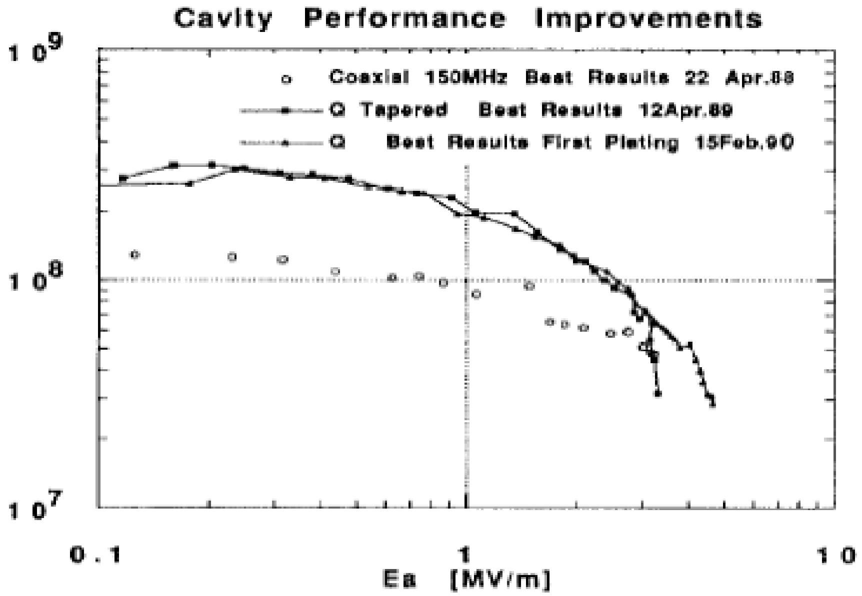
- **1988**: development of Pb/Cu QWRs (established technology at the time)
- **1994**: a U-shaped linac with Pb/Cu mid-β QWRs; R&D of low-β full Nb and high-β Nb/Cu
- **1998**: first high-β and 3 lower-β cryostats installed
- **1998-2004**: mid-β QWRs with Nb/Cu technology (same Cu-base); 2^{nd} high-β QWR, 4^{th} full-Nb QWR; development and construction of PIAVE full Nb QWRs and SRFQs;
- 2006-2008: liquid-N cooled coupler improve stability of lower-β full Nb QWRs; Nb/Cu on new Cu-bases in test cryostat CR15
- 2016-2018: upgrades of PIAVE SRFQs to improve field and phase stability
- **2019-2021**: 2 additional high-β Nb/Cu resonators



G. Bisoffi – Long-term SRF Experience at INFN-Legnaro, TTC Meeting – Vancouver 2019

The early ALPI with mid-β Pb/Cu QWRs

1992: ALPI during first assembly



E_a = 2.3÷2.7 MV/m

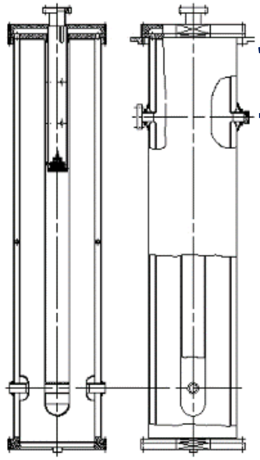
β_0=0,11

Reliable operation

Cheaper than full Nb, **mechanically stable, not** susceptible to **quench**, ideal for complicated geometries

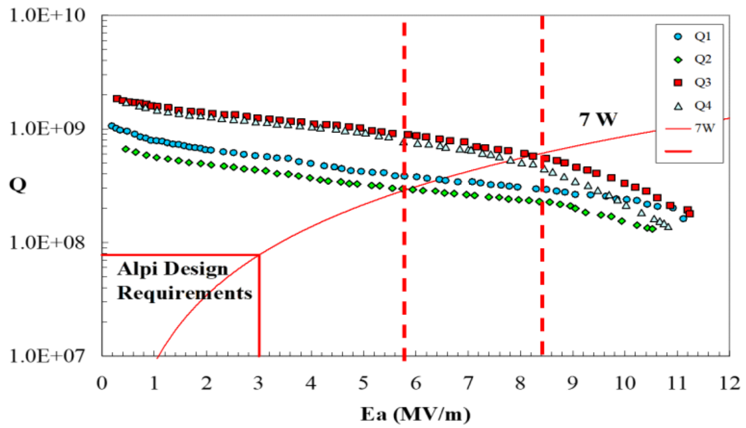
Limited performance, and some **degradation of E_a**

R&D on full-Nb and Nb/Cu QWRs

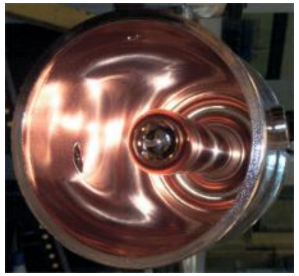


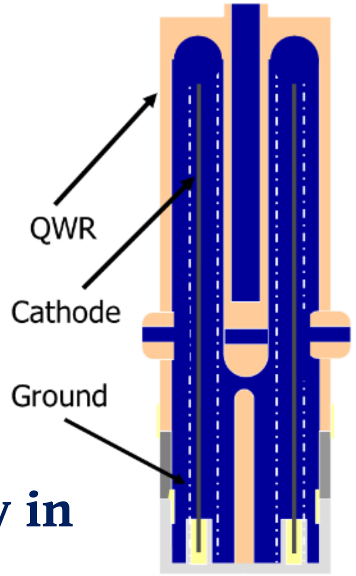
First 80 MHz low-β full Nb cavity in 1993

(double wall thin Nb outer conductor, equipped with original mechanical dampers)



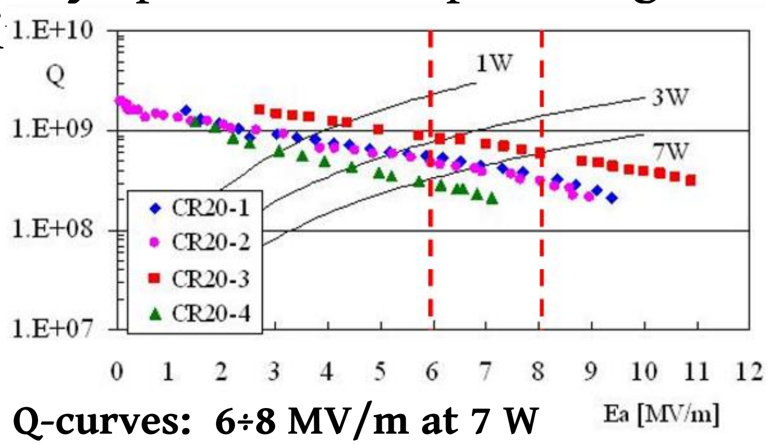
Off-line Q-curves: 6÷8 MV/m at 7 W





First 160 MHz high-b Nb/Cu cavity in 1993

(geometry optimized for sputtering, capaci plate)



Off-line Q-curves: 6÷8 MV/m at 7 W

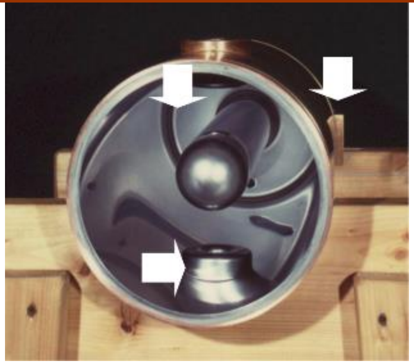
Higher mechanical stability
Less sensitive to microphonic

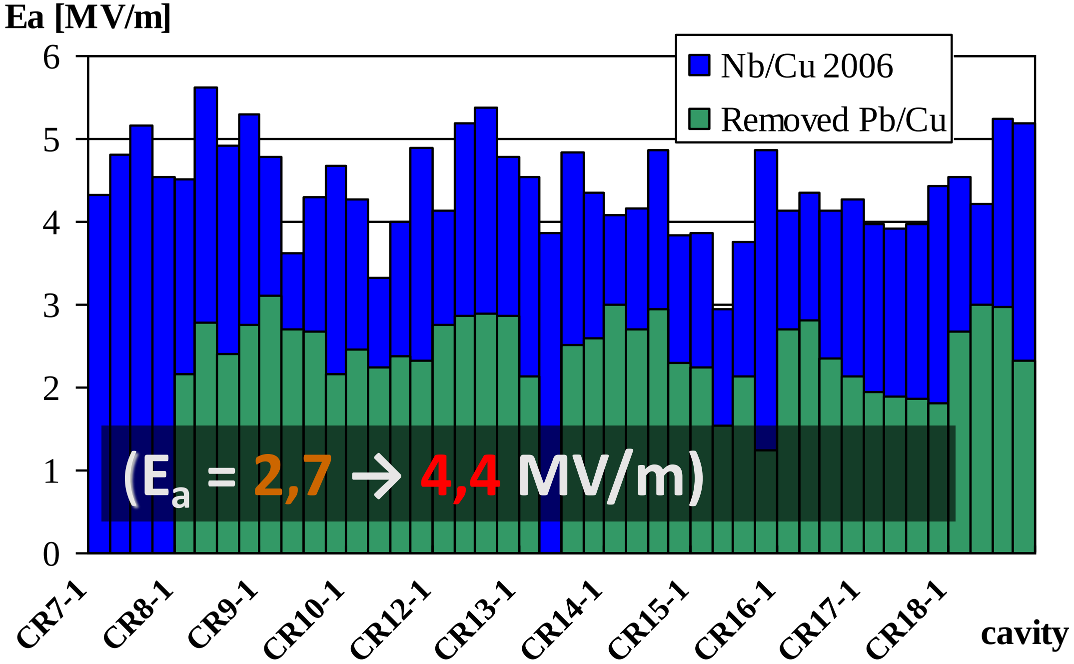
G. Bisoffi – Long-term SRF Experience at INFN-Legnaro, TTC Meeting – Vancouver 2019

The next step: apply Nb/Cu to mid-β QWRs…

Bu

n.44 160 MHz, Nb/Cu, β_0 0,11 | n.8 160 MHz, Nb/Cu, β_0 0,13





Bu

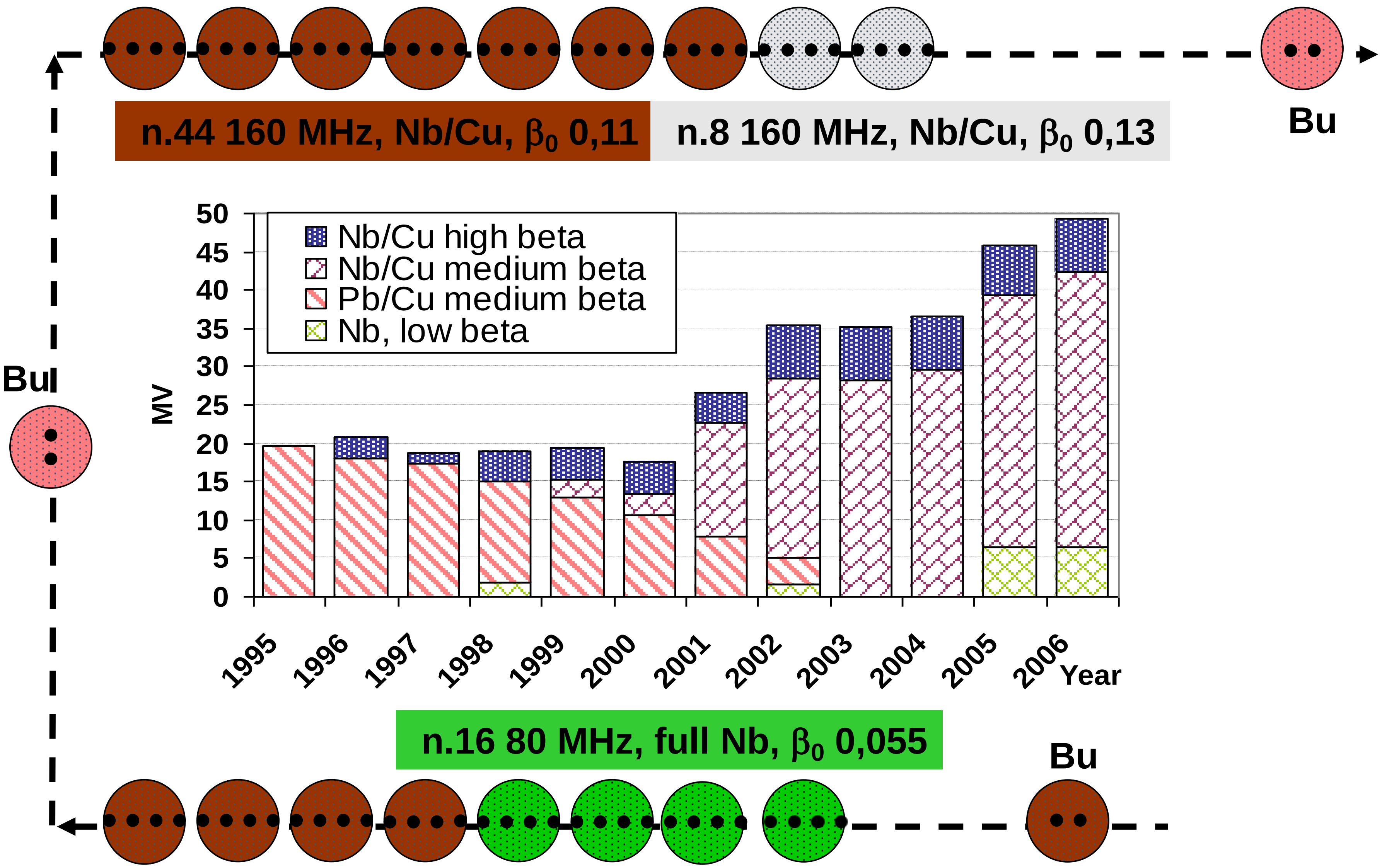
… on a non-Nb/Cu-optimized geometry and Cu-brazed joints (smaller radii on end pkate and beam-ports, beam coupler hole in high-H region)

n.16 80 MHz, full Nb, β_0 0,055

Bu

G. Bisoffi – Long-term SRF Experience at INFN-Legnaro, TTC Meeting – Vancouver 2019

ALPI V_{eq} from 20 to 48 MV



G. Bisoffi – Long-term SRF Experience at INFN-Legnaro, TTC Meeting – Vancouver 2019

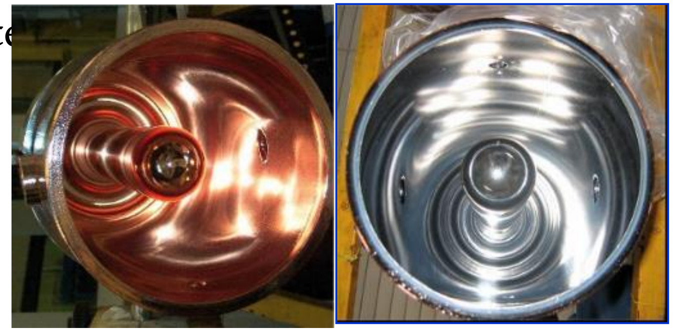
ALPI QWR - Effect of the substrate

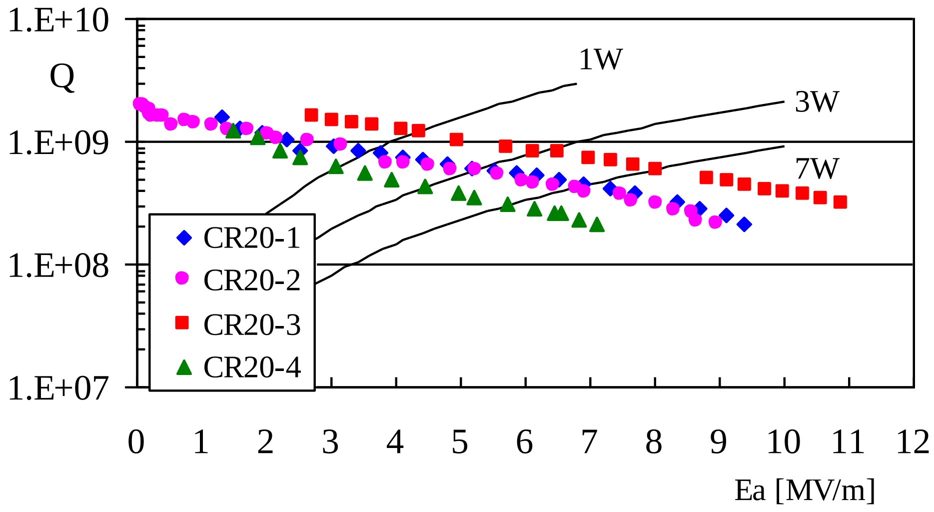
High β QWRs (β=0.13, 160 MHz)

Drilled by a billet of OFHC Cu, 99.95% grade

No brazed joints, beam ports jointed by indium gaskets

Rounded shorting plate



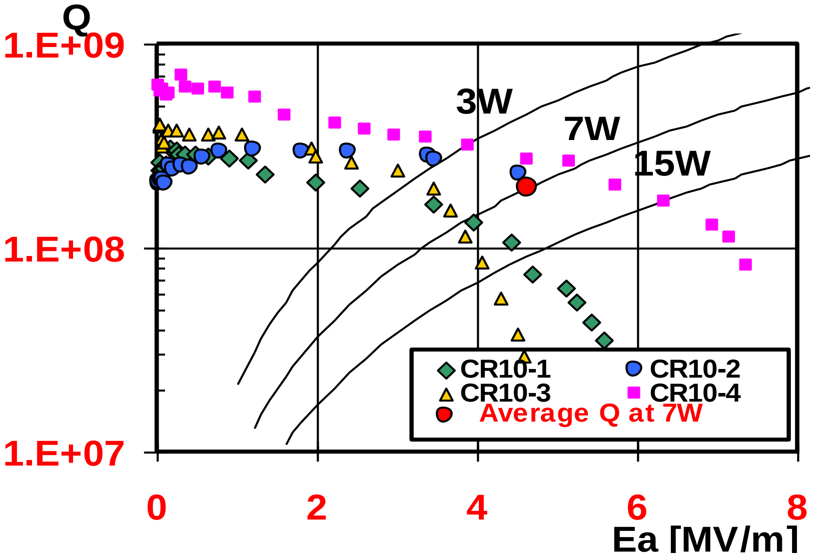


Medium β QWRs (β=0.13, 160 MHz)

Brazed joints (especially the ones in the outer resonator surface)

Flat shorting plate

Beam ports shape



A. Porcellato, Nb Sputtered Cu QWR, Thin Films Workshop LNL 2006

Nb/Cu sputtering advantages

Mechanical stability (mechanical vibrations are not an issue)

Frequency not affected by changes He bath Dp (<0.01Hz/mbar)

Reduced over-coupling (smaller amplifier, coupler do not need cooling, rf lines have reduced size and limited rf dissipation)

High thermal stability (less prone to hot spots, conditioning easier)

Stiffness (in case of loss of isolation vacuum leak)

Absence of Q-disease (less demand on cryogenic system cooling velocity and reliability)

Insensitivity to small magnetic fields (no magnetic shielding)

High Q of the N.C. cavity (easier coupling in N.C state)

Absence of In vacuum joints (vacuum leaks less probable)

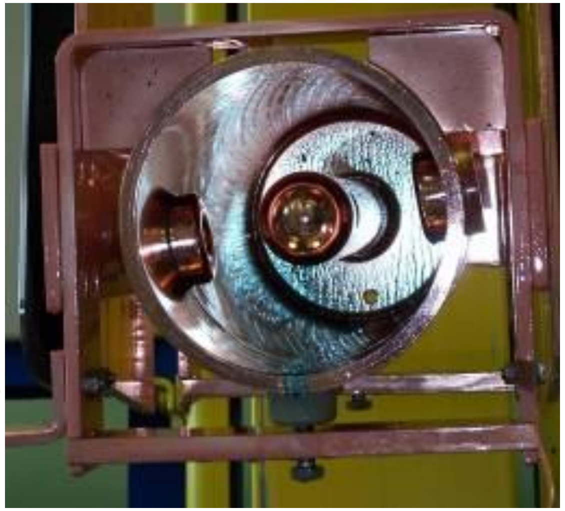
Price (both material and construction)

The lower performance of Nb/Cu cavities at high fields, due to the more pronounced Q-slope of Nb/Cu resonators, is not an issue in QWRs as it is in b>0.5 cavities, because beam dynamic constraints require to limit the accelerating gradient in the low b section of linacs to values well reachable by Nb sputtered resonators

A. Porcellato, Nb Sputtered Cu QWR, Thin Films Workshop LNL 2006

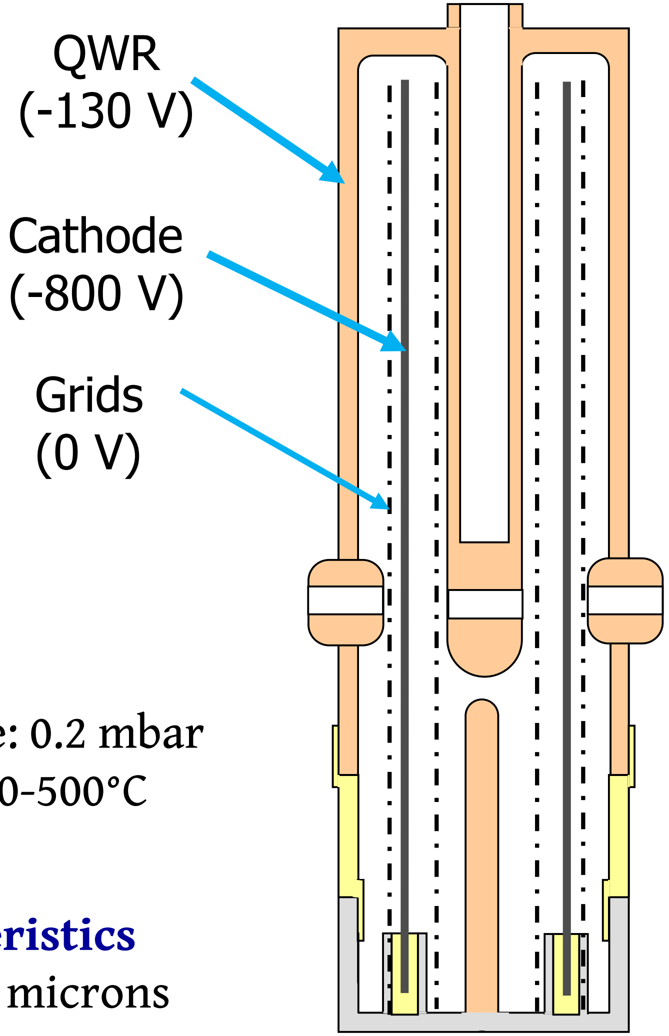
ALPI QWR - Substrate preparation

- Electropolishing (20μm, 2 hours)
- Rinsing (water, ultrasonic water, HPR)
- Chemical polishing (10μm, 4 min, SUBU5)
- Passivation (sulphamic acid)
- Rinsing (water, ultrasonic water, HPR)
- Drying (ethanol, nitrogen)



A. Porcellato, Nb Sputtered Cu QWR, Thin Films Workshop LNL 2006

ALPI QWR - Sputtering process



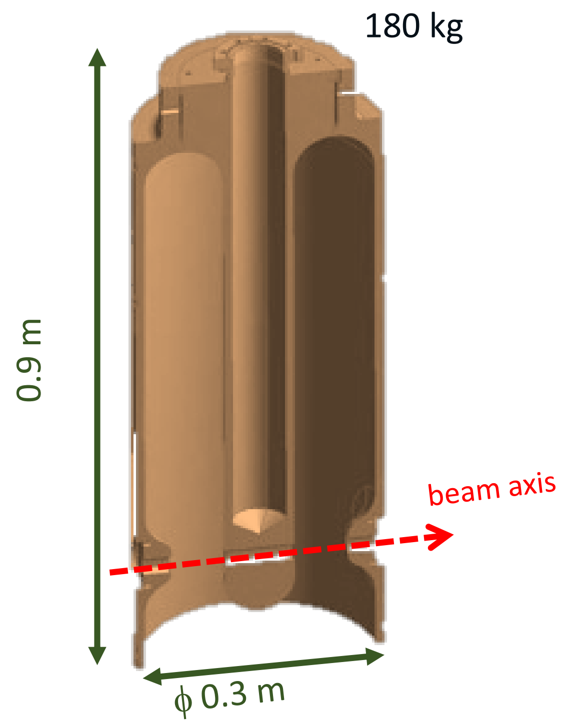
Parameters
Argon pressure: 0.2 mbar
Substrate T: 300-500°C

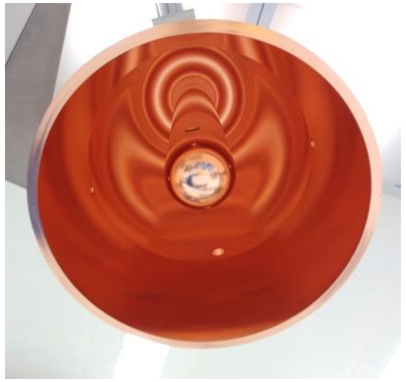
Film characteristics
Thickness: 1-2 microns
RRR: 9-20

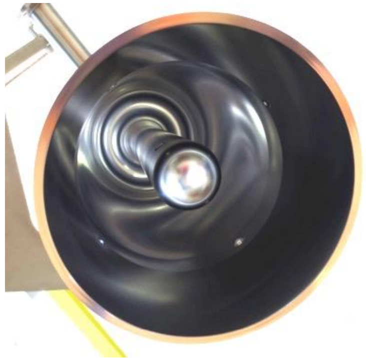
A. Porcellato, Nb Sputtered Cu QWR, Thin Films Workshop LNL 2006

HIE-ISOLDE @ CERN

Superconducting linear accelerator for energy upgrade of ISOLDE radioactive ion beam facility







Cryomodule clean room assembly

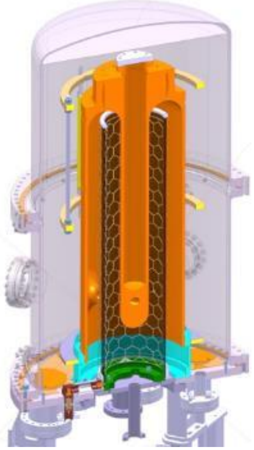
4 cryomodules in HIE-ISODLE Linac

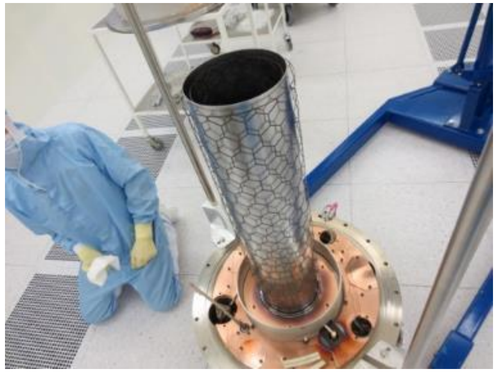
Courtesy of A. Sublet (CERN)

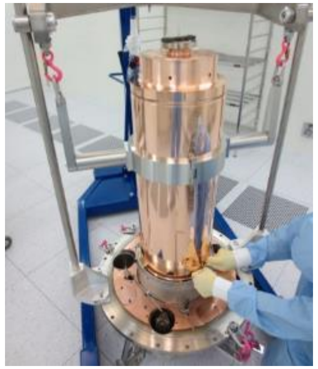
HIE-ISOLDE @ CERN

Same sputtering **configuration** as in **ALPI**

Clean room assembly improvement







Cavity in UHV chamber (10^{-8} mbar base vacuum)

3D-forged Cu cavity substrate, biased at -80 V

Nb cylindrical cathode used on both sides, not cooled

DC-bias diode sputtering, 8 kW, Ar 0.2 mbar

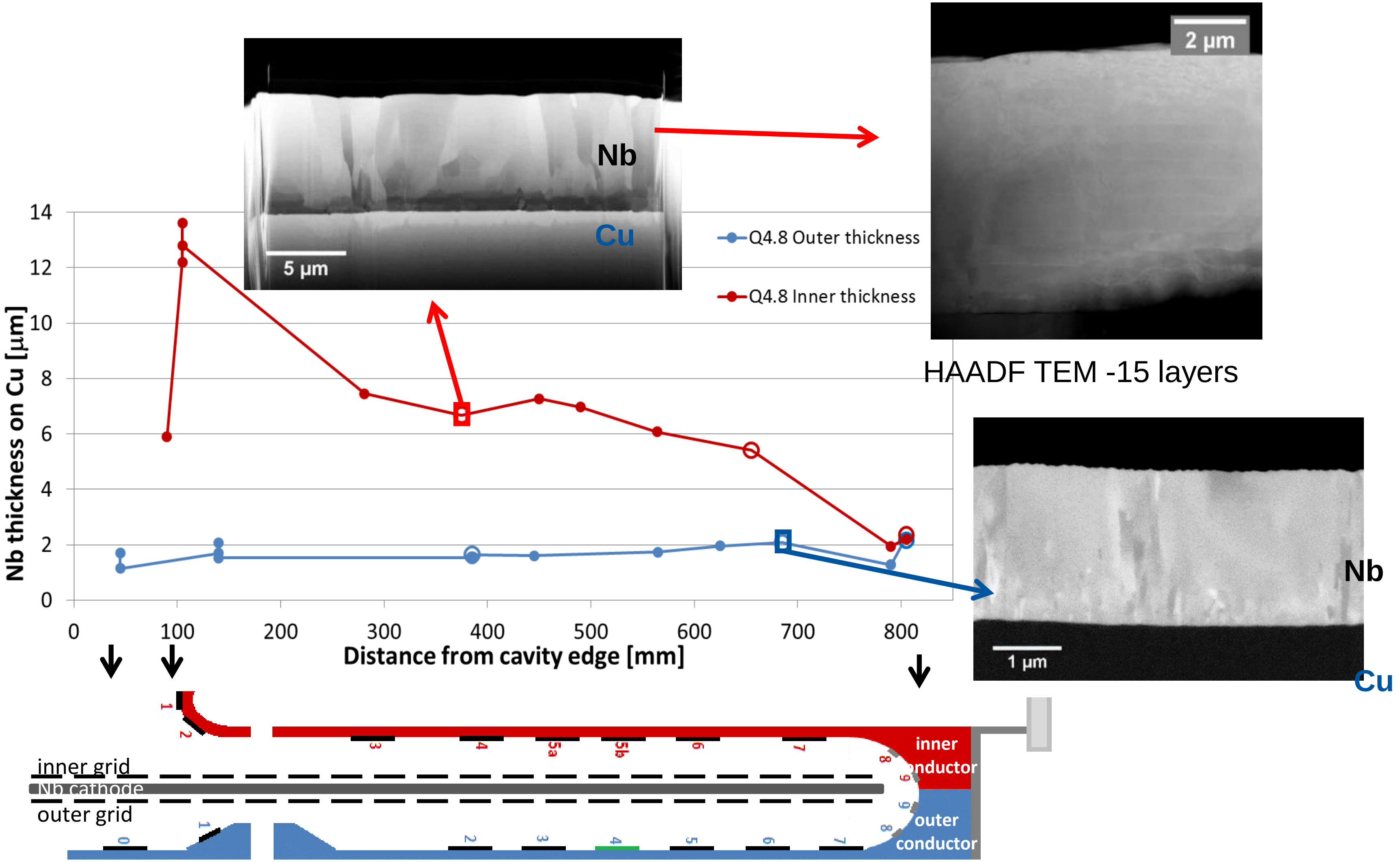
Coating at high temperature (300 → 620°C)

Done in 15 run/cool-down cycles (4 days)

Nb layer thickness ranging from 1.5 mm to 12 mm

Courtesy of A. Sublet (CERN)

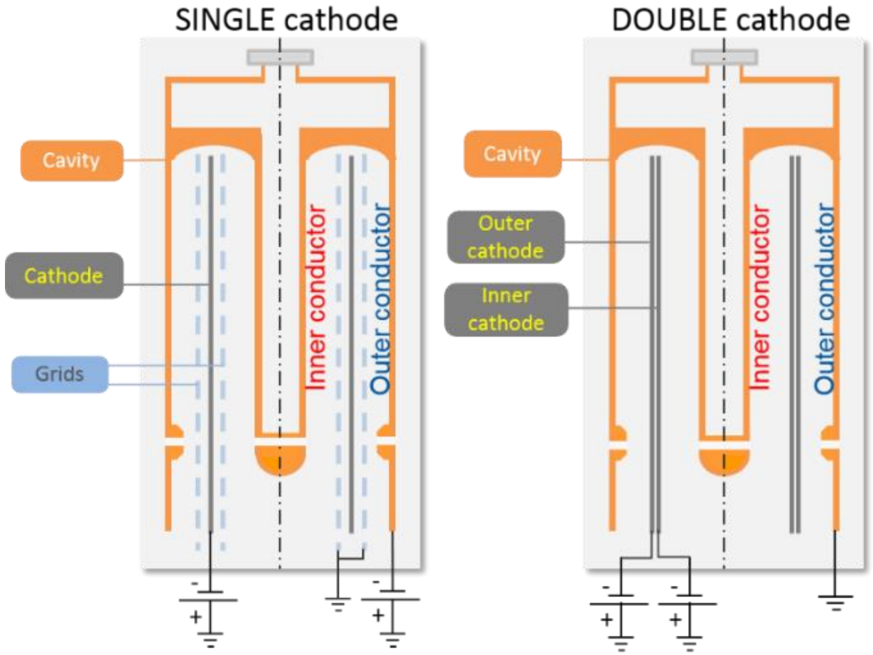
HIE-ISOLDE film characteristics

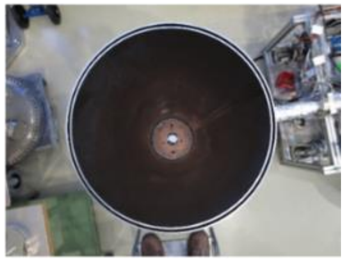


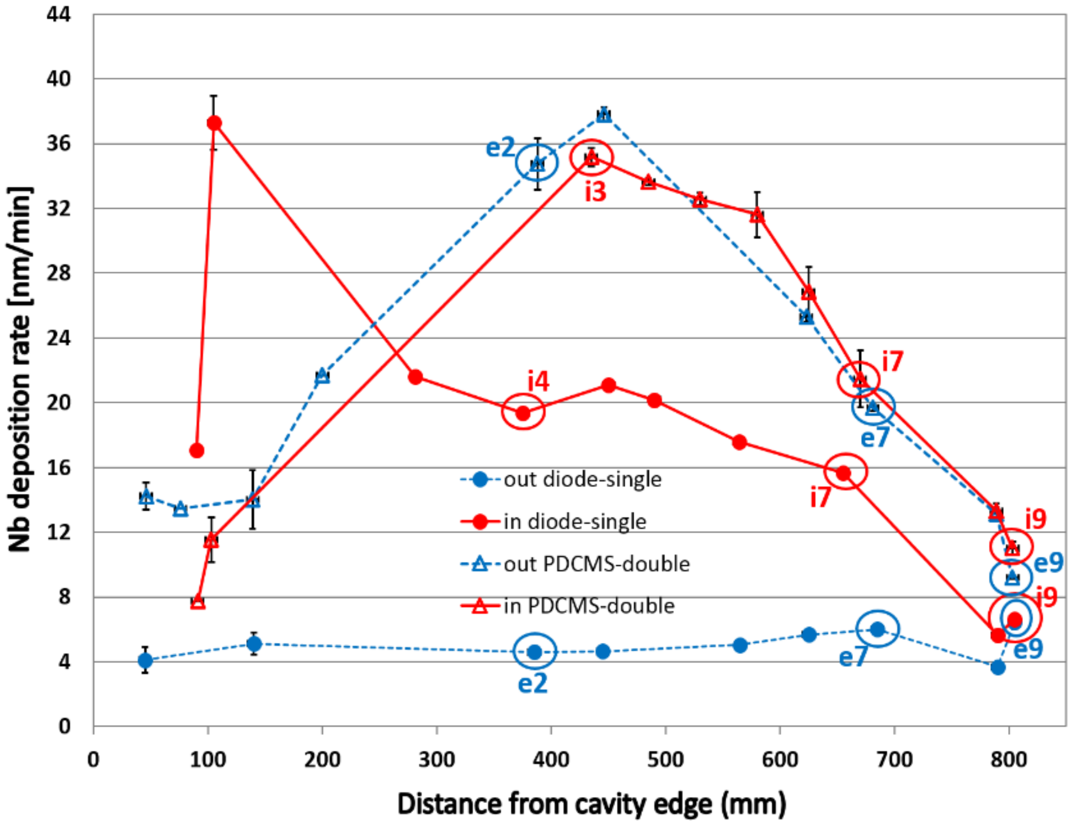
→ Influence of "multilayered" Nb film/dislocations/morphology on the RF performances?

A. Sublet (CERN), Thinfilms Workshop, LNL 2018

HIE-ISOLDE: double cathode scheme DCMS





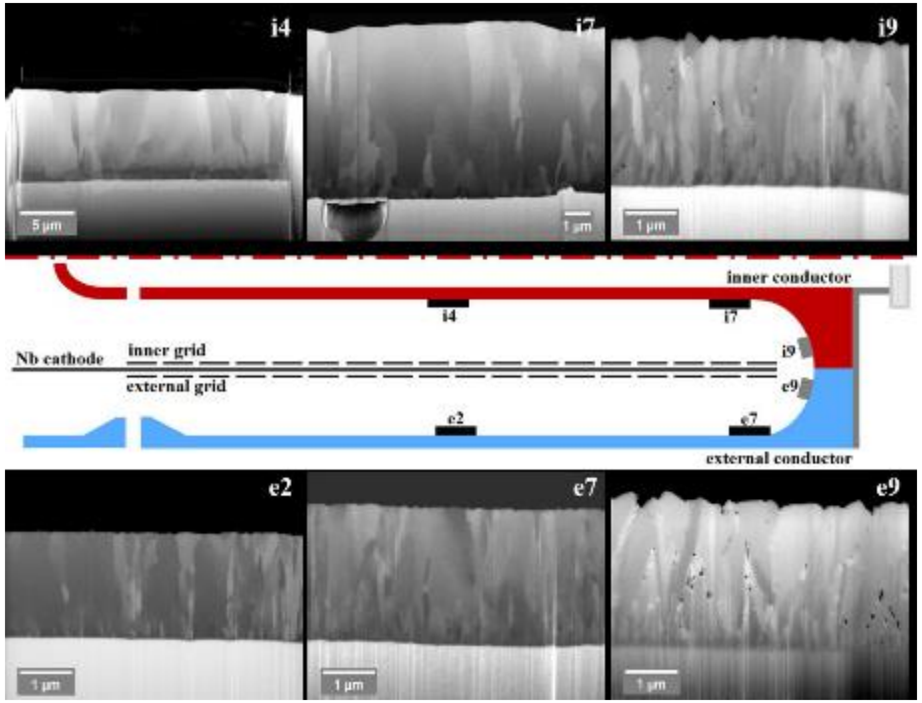


→ **Decouple inner/outer plasma**, independent power control: tune layer uniformity

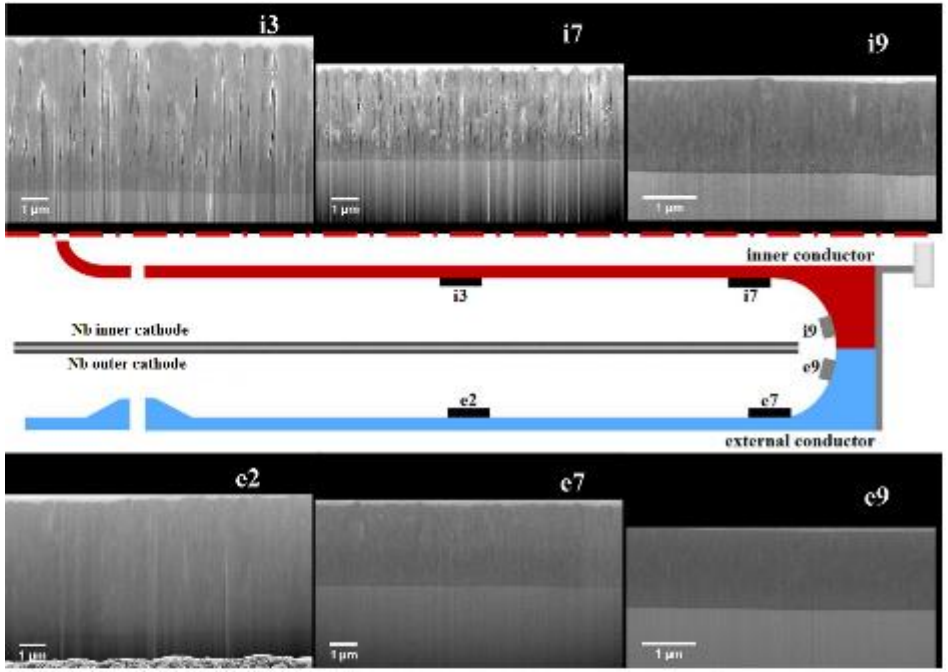
A. Sublet (CERN), Thinfilms Workshop, LNL 2018

HIE-ISOLDE layer morphology

diode



p-DCMS



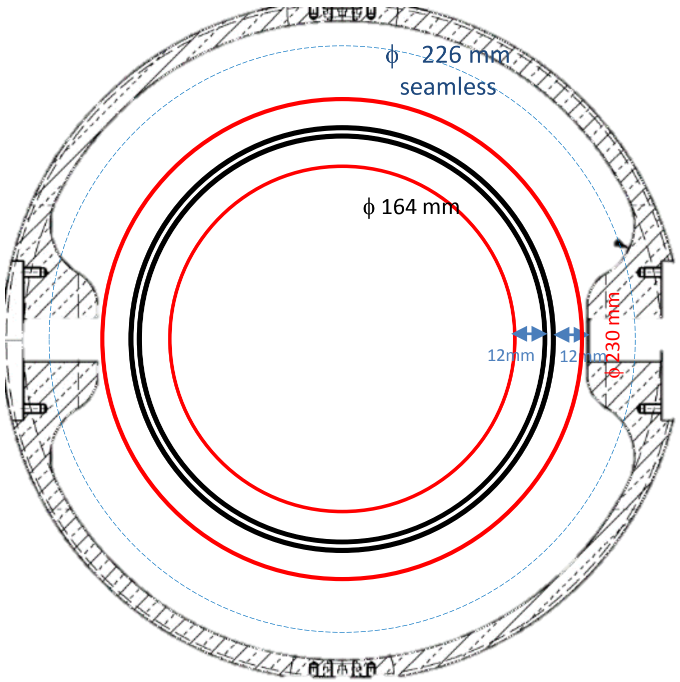
Technique	Cathode scheme	Pressure [mbar]	Magnetic field[G]	Cathode(s) power [kW]	Duration [min]	Energy [kWh]
Diode	Single	$2x10^{-1}$	none	8 (5.4 in, 2.6 out)	345	43 (29 in, 14 out)
Pulsed DCMS	Double	1.6×10^{-2}	118	2 in, 4.5 out	145	4.4 in, 9.8 out

→ Denser film outer and at the top in pulsed DCMS, but rough and porous on the inner

→ Geometry and transport at low pressure in DCMS may explain these observations

A. Sublet (CERN), Thinfilms Workshop, LNL 2018

HIE-ISOLDE double cathode + grids

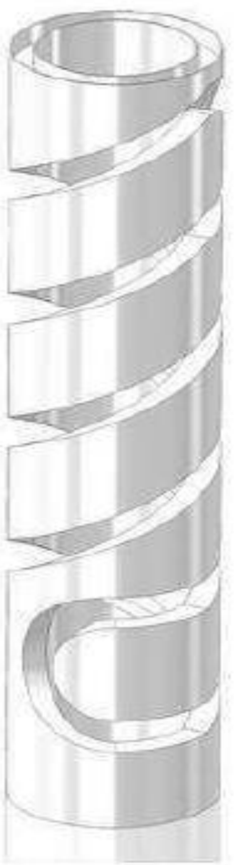


→ Bias on cavity (as for single cathode diode baseline) with diode or (p-)DCMS

→ Densify film by ion bombardment assistance

HIE ISOLDE R&D @LNL

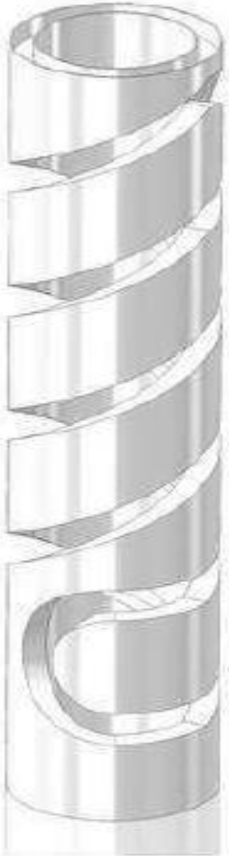
Helicoidal magnetic configuration

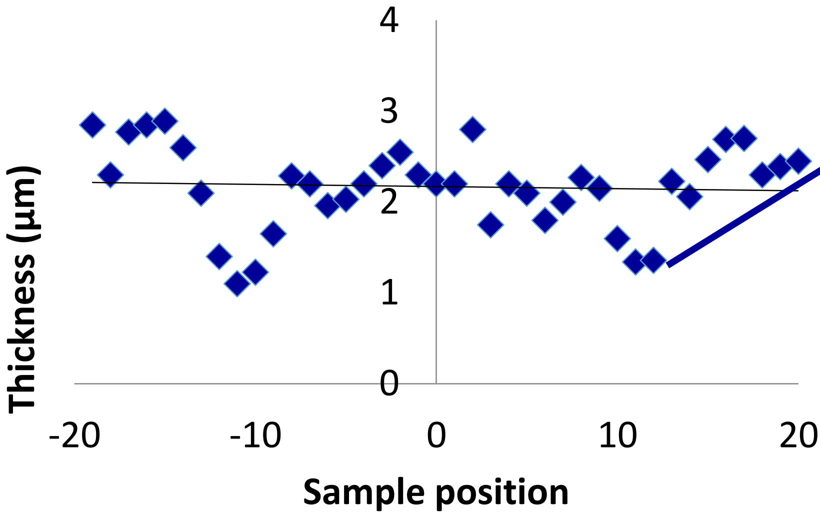


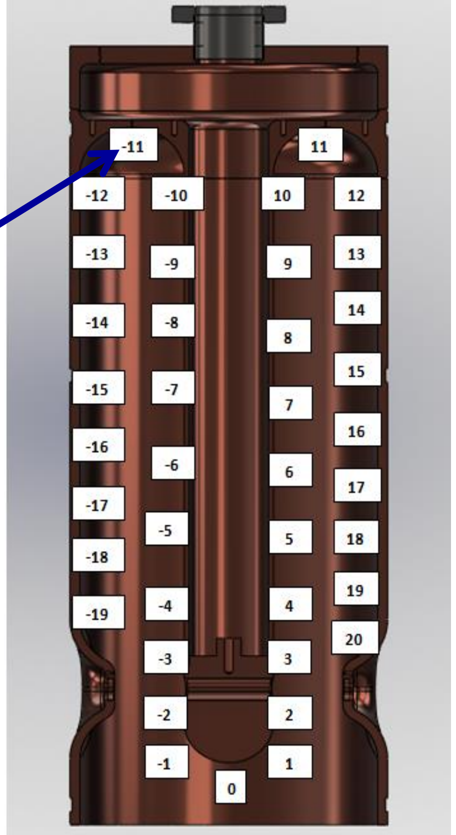
D. Franco, PhD Thesis

HIE ISOLDE R&D @LNL

Helicoidal magnetic configuration







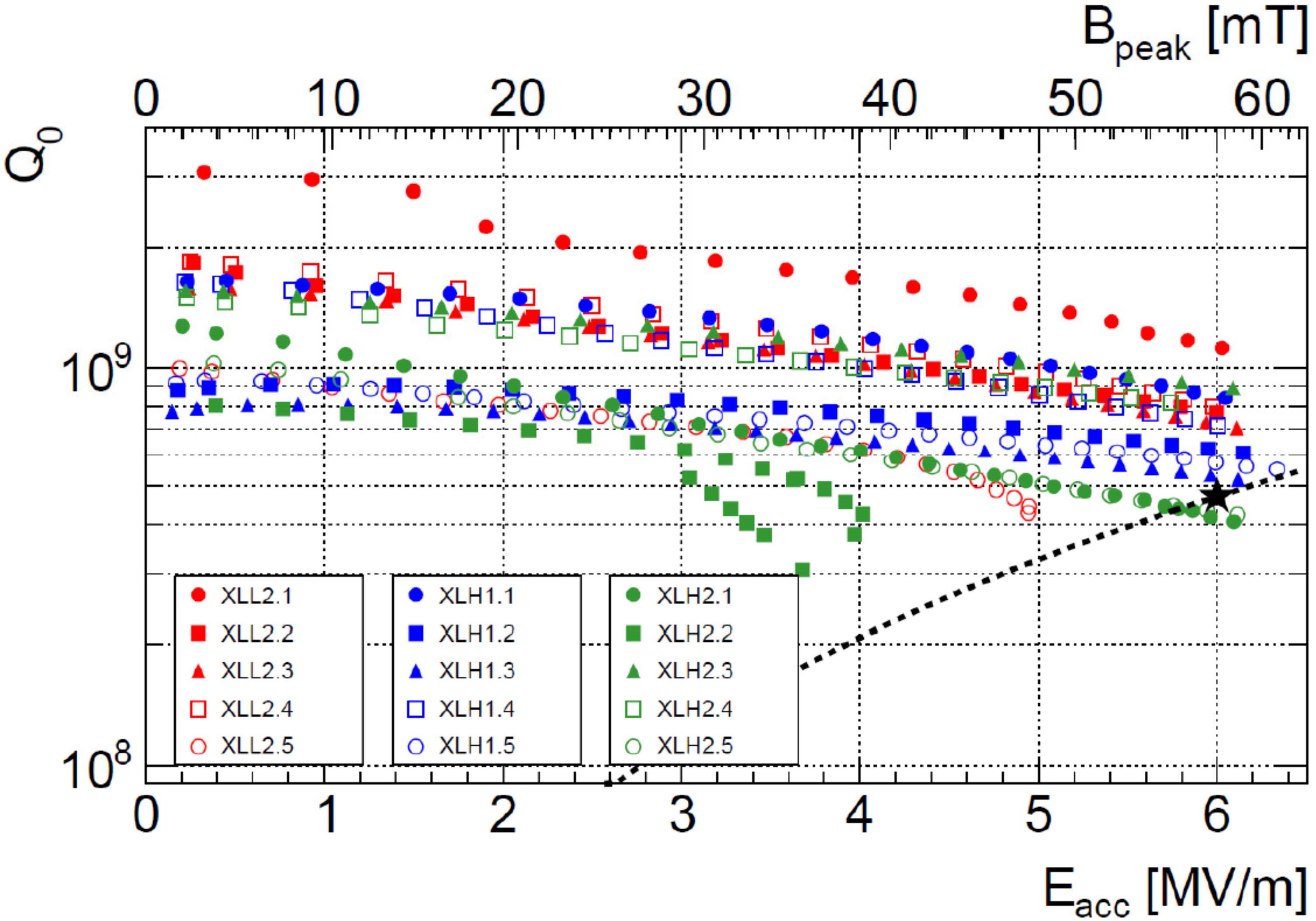
Good thickness uniformity

R&D stopped due to difficulties in the handling of the chemical polishing

(HIE ISOLDE QWR larger than ALPI QWR)

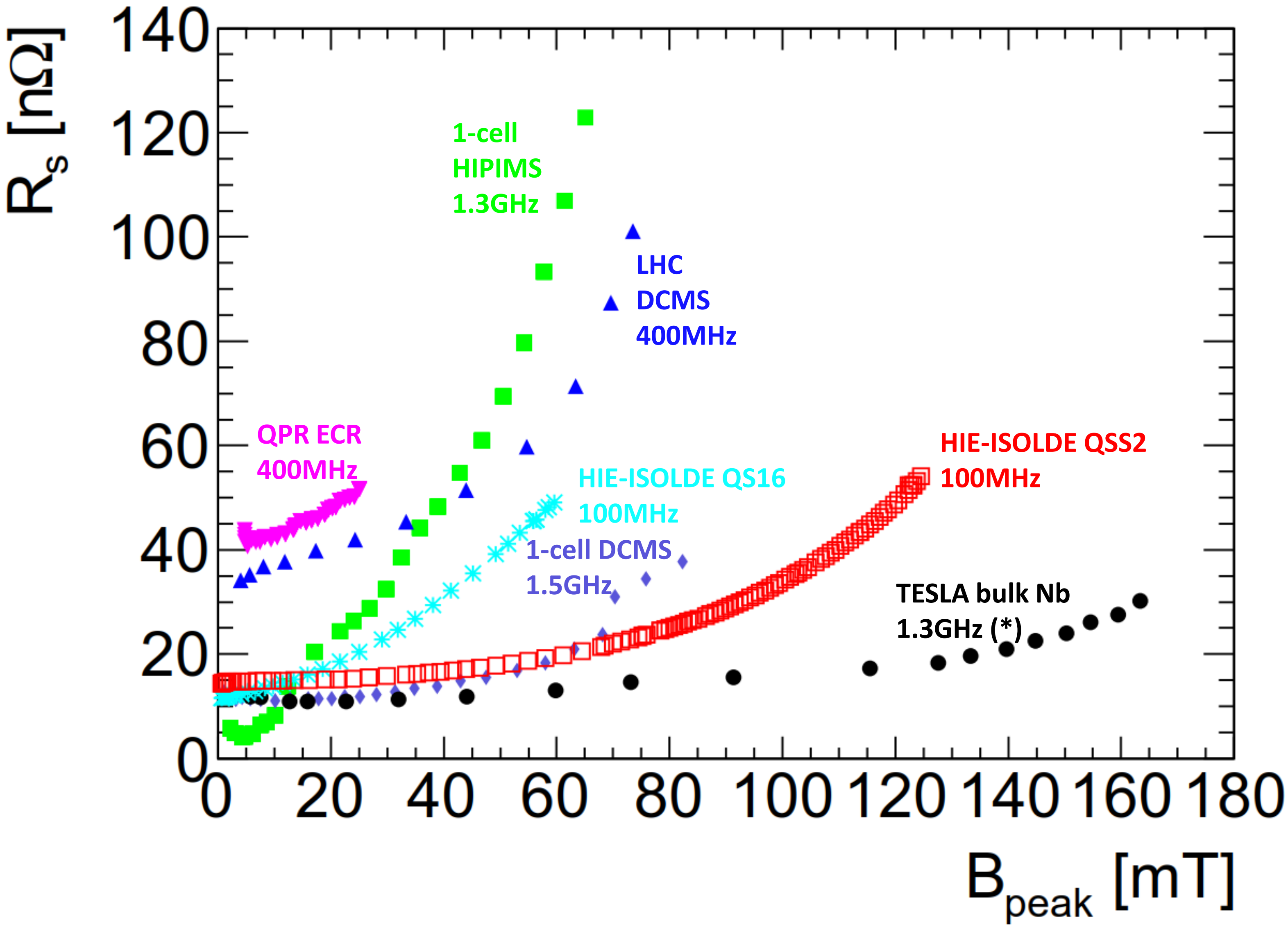
D. Franco, PhD Thesis

HIE-ISOLDE QWR Performances



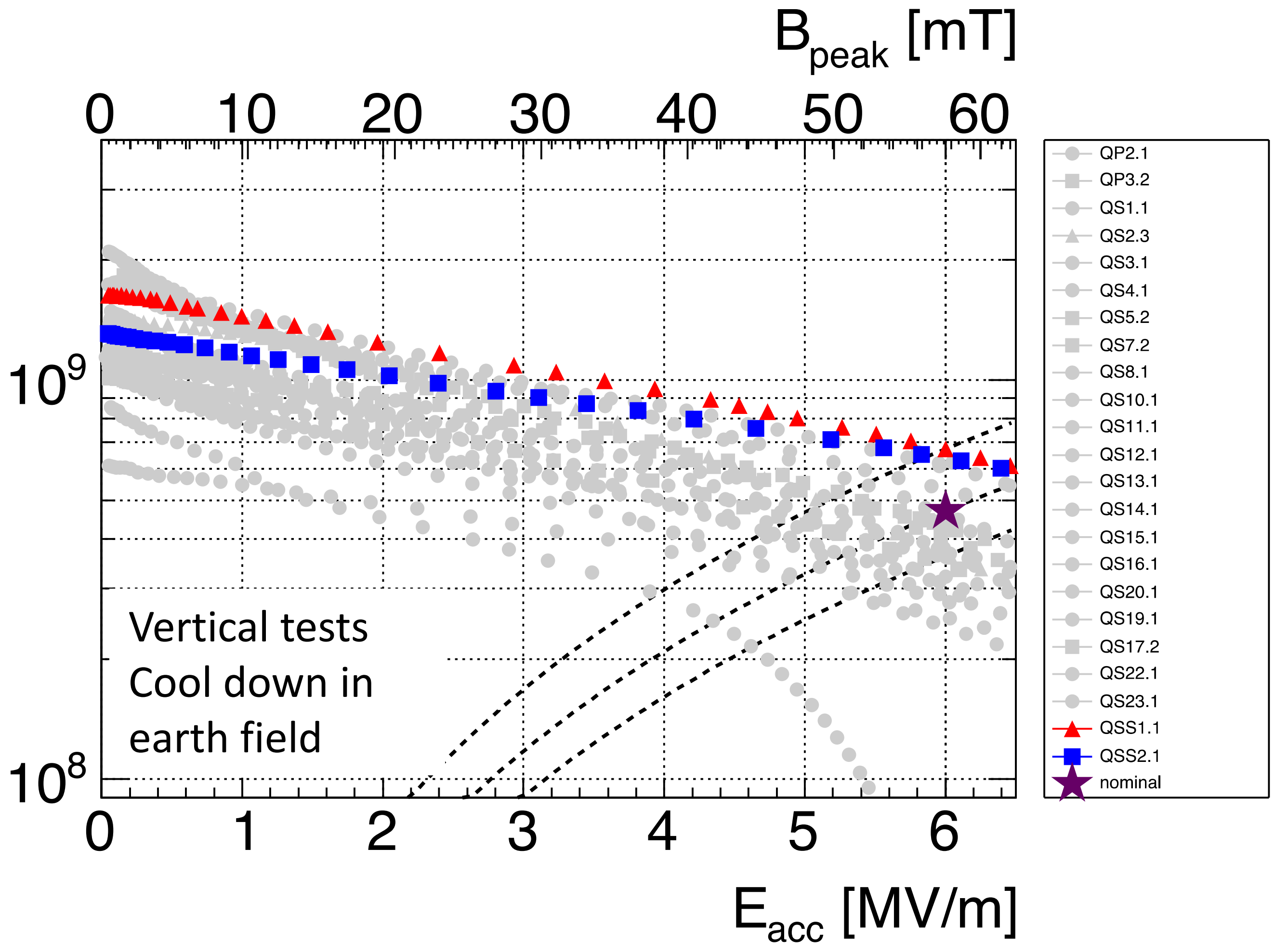
Courtesy of W. Venturini (CERN)

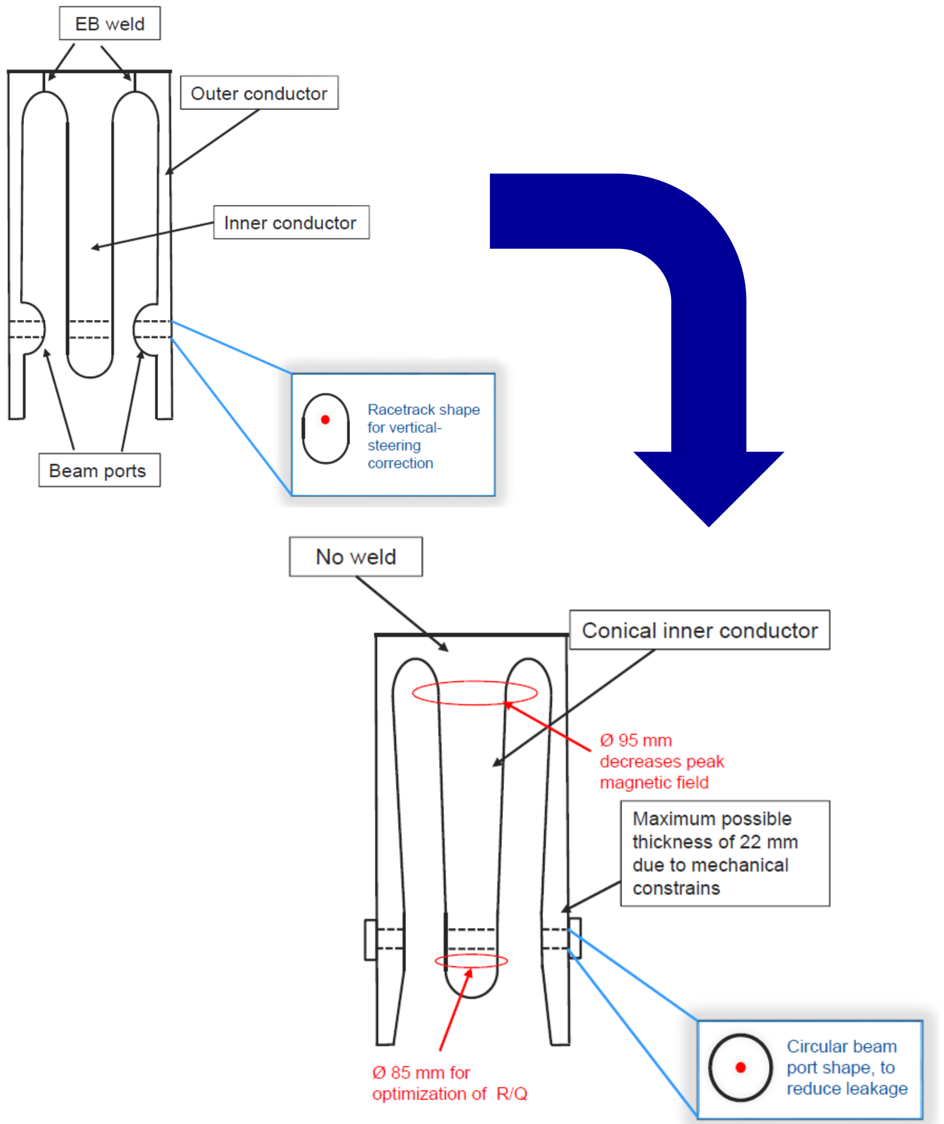
State of the art of Nb-Cu films around 2 K



Courtesy of W. Venturini (CERN)

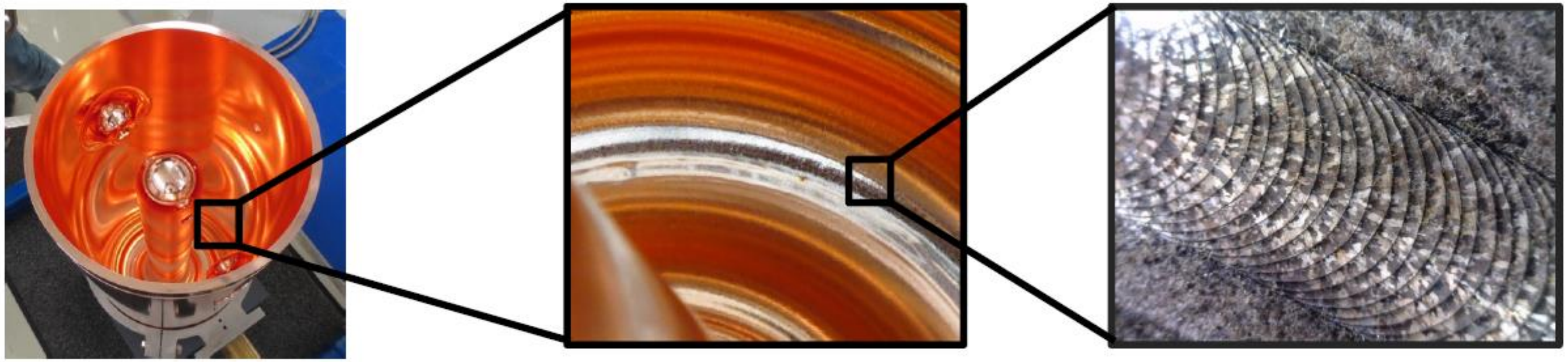
HIE-ISOLDE QWR Seamless Design

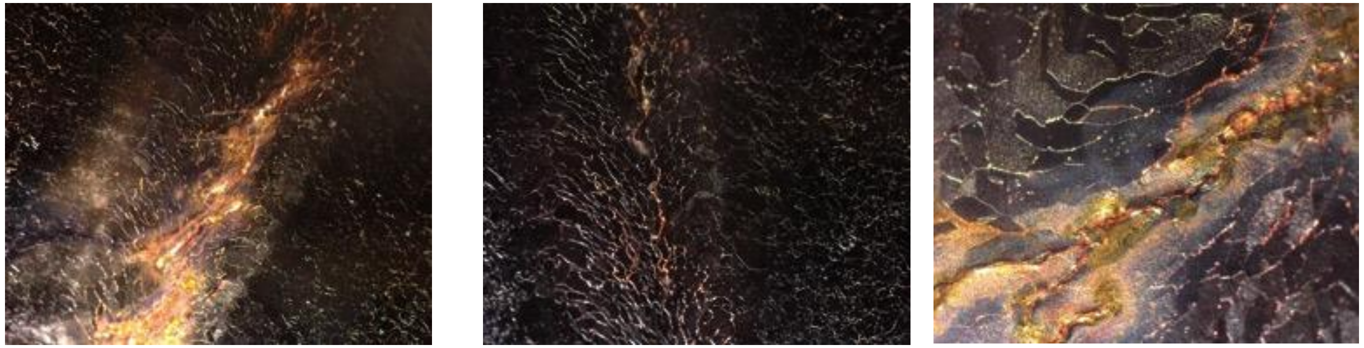




Courtesy of W. Venturini (CERN)

Motivation through the seamless design





→ **longitudinal fracture** along the weld

→ Source of chemicals trap/release

→ oxidation/contamination → peel-off

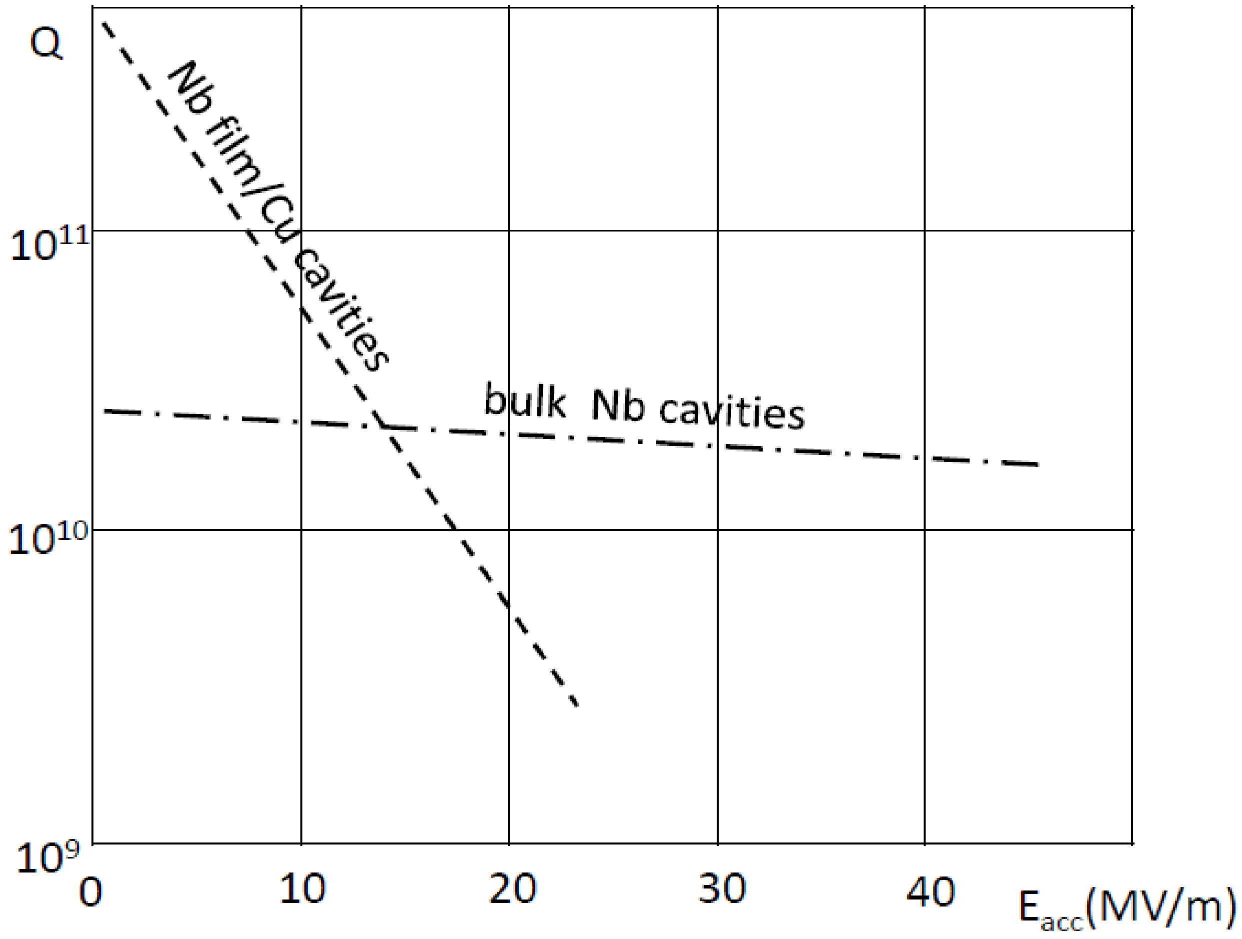
Courtesy of A. Sublet (CERN)

QWR Main Lesson learned: the substrate is important!

Outline

- Motivation for thin films in SRF cavities
- How to realize a thin film coating?
- State of the art in Nb thin films (accelerators using thin film technology)
- **Characteristics of Nb films**
- R&D on Nb films

Q-slope problem



Unsolved problem since 1990s

Several theory proposed

Depinning of trapped flux

Low HC1

Early vortex penetration due to roughness

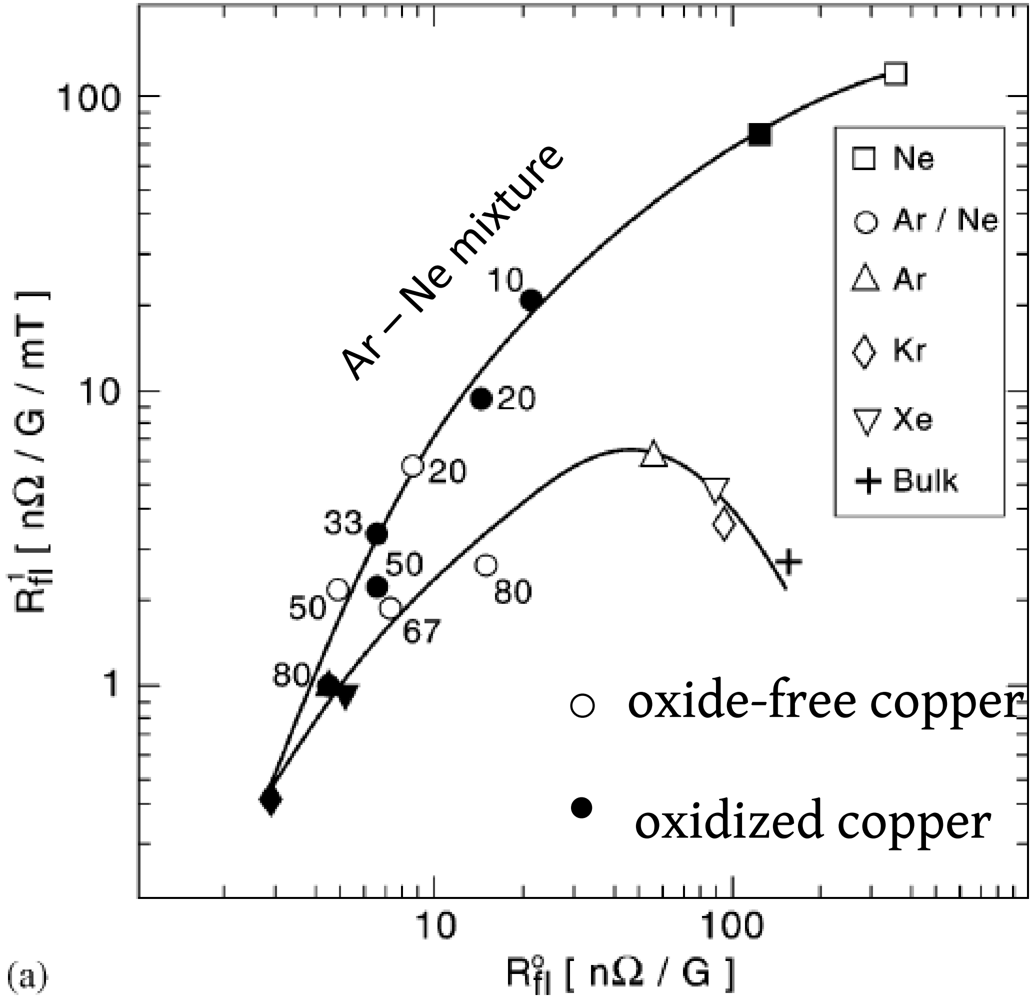
Grain boundaries

Bad thermal contact at the interface

Not intrinsic problem of the films

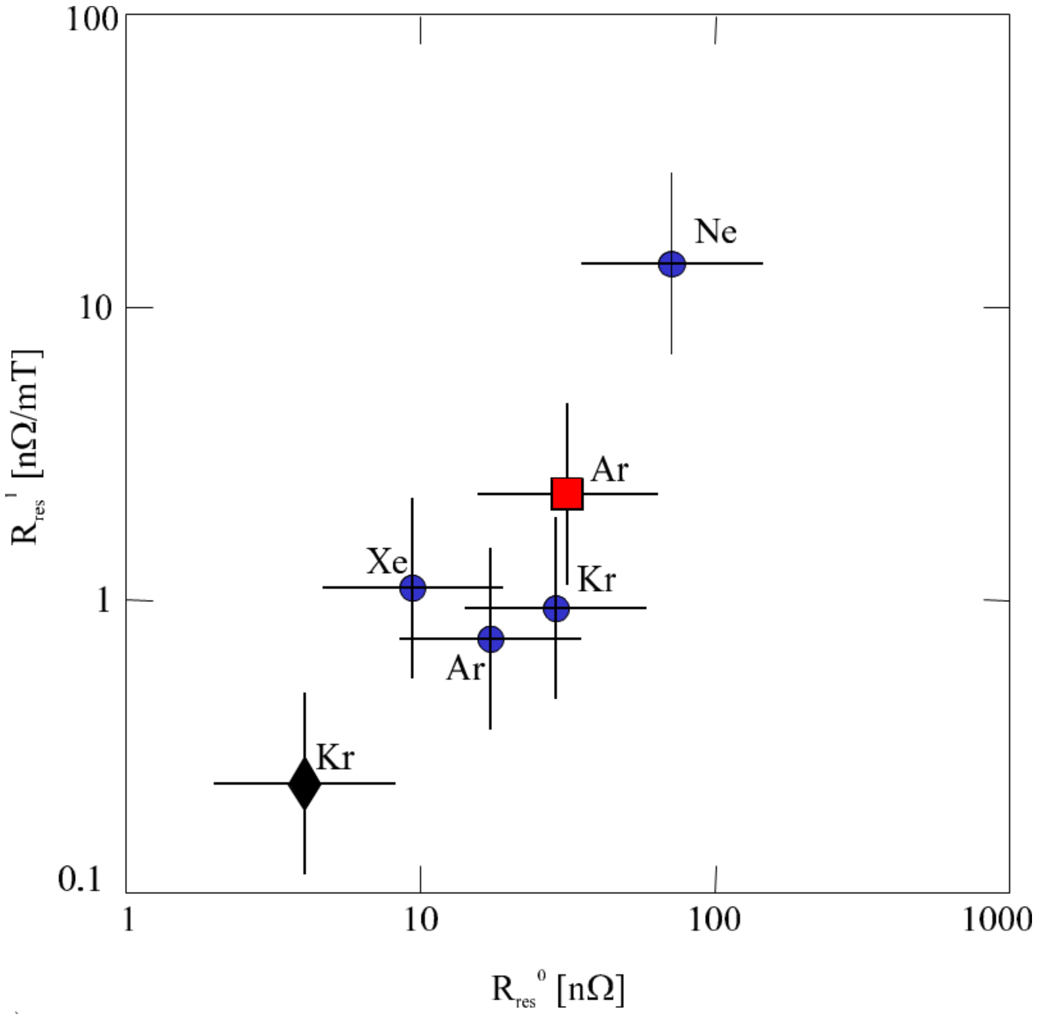
Substrate is important

Effect of the gas



S. Calatroni (CERN), SRF 2001

Effect of Polishing



Courtesy of S. Calatroni

Average roughness of chemically polished spun cavities: 0.2µm

Average roughness of chemically polished hydroformed cavities: 0.8µm

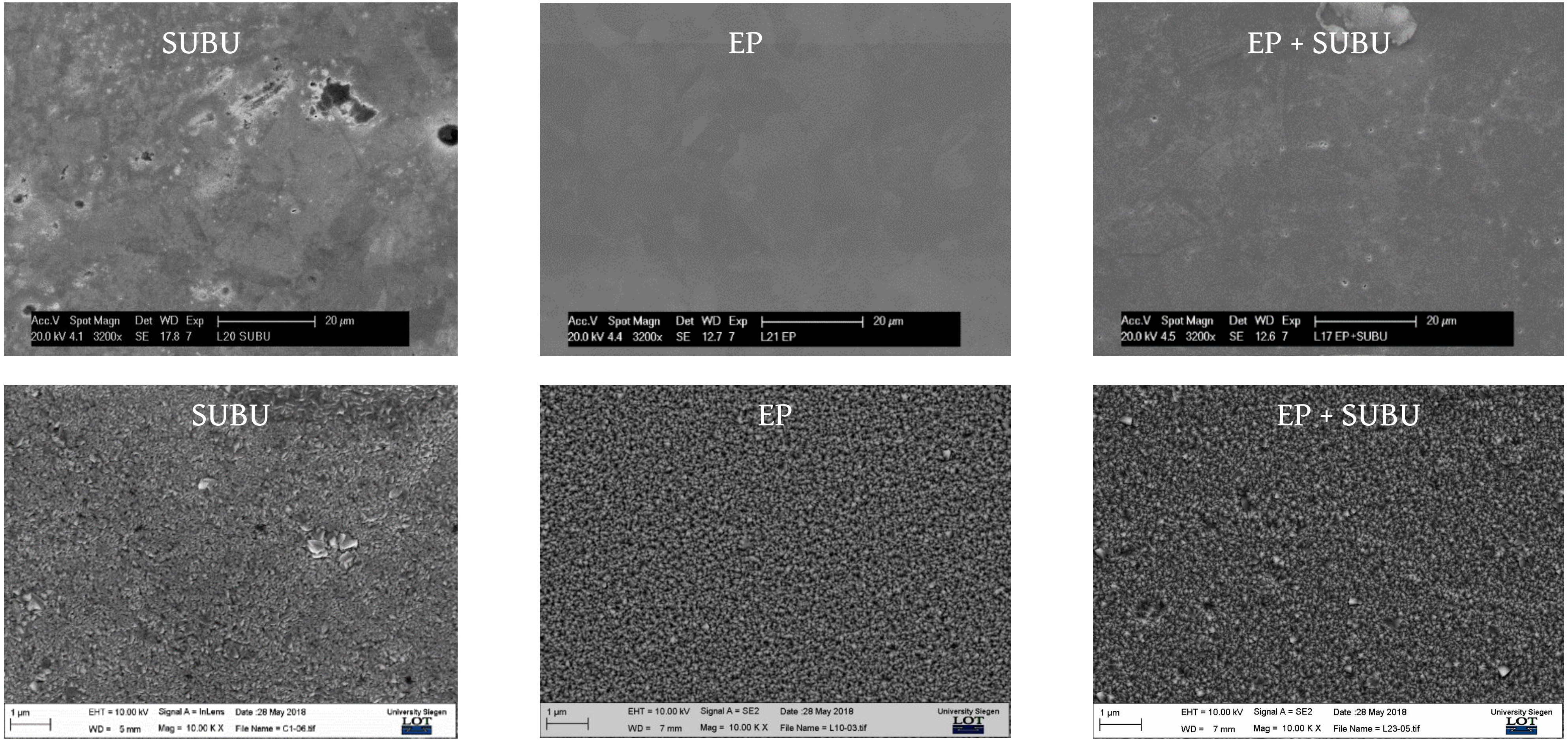
Average roughness of electropolished spun cavities: 0.04µm
Absence of defects (etching pits)

EP seems better than SUBU5

Effect of Polishing

Cu

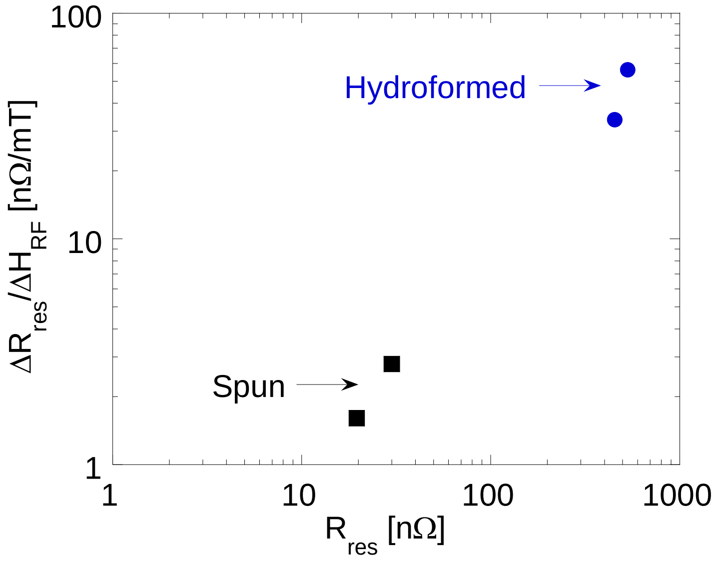
Nb film



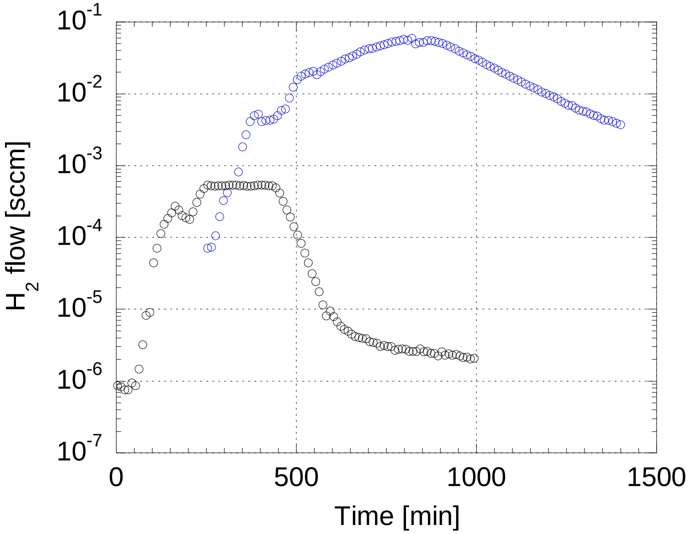
PVD film mimate the surface morphology

ARIES Collaboration (2018)

Effect of the Cu substrate forming process



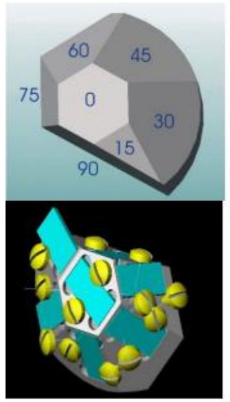
Coatings on oxide-free **hydroformed** cavities consistently **worse than** for **spun cavities**? Why?

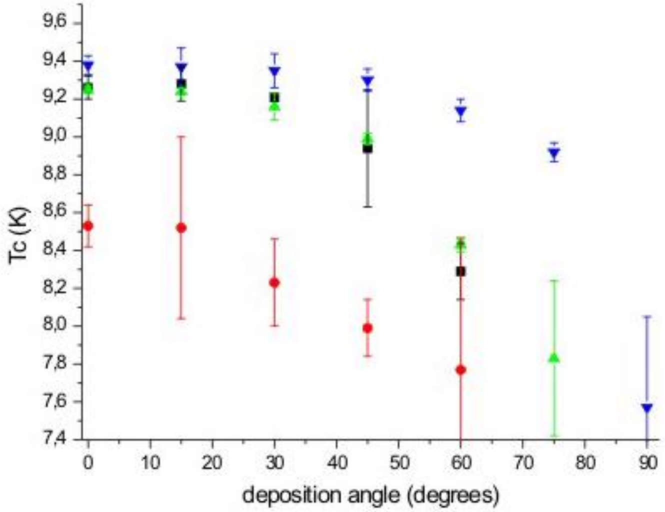


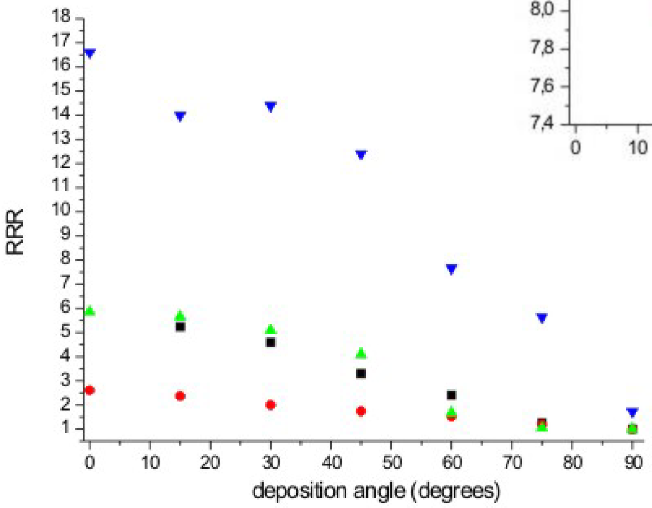
Possible answer: a larger quantity of **hydrogen** was migrating into the film from the hydroformed cavity

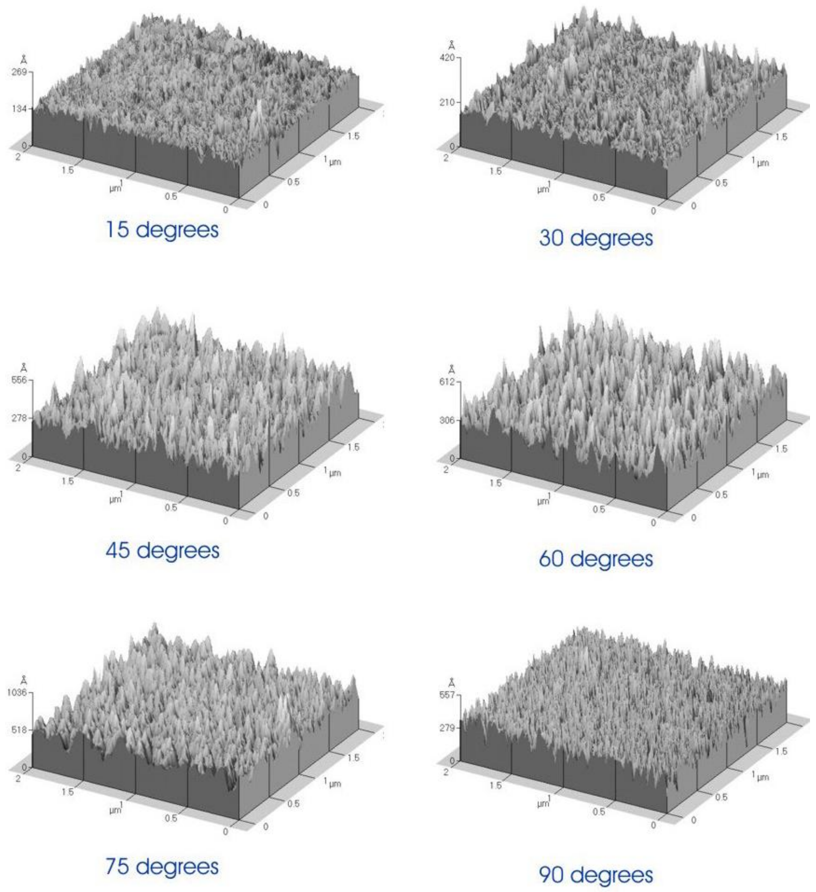
S. Calatroni (CERN), SRF 2001

Angle of incidence of coating









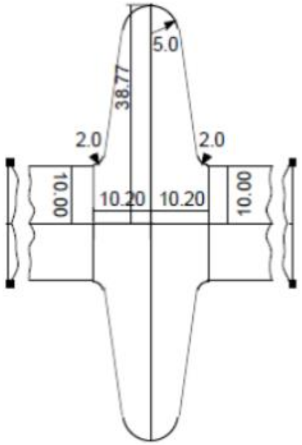
Superconducting properties of niobium films depends on deposition angle between target and substrate

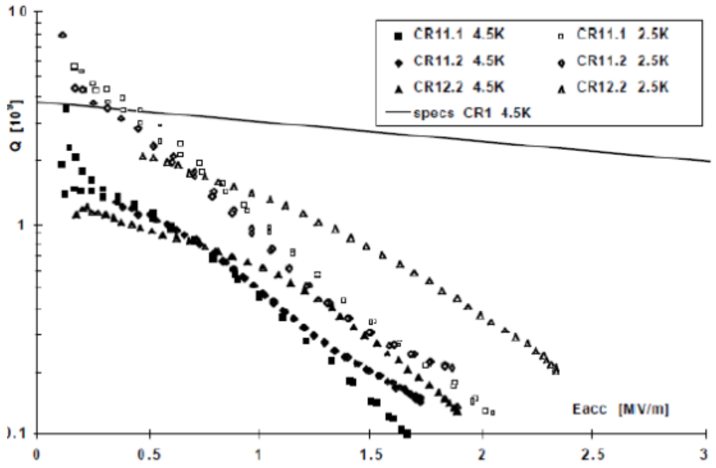
The effect is related to change in the coating morphology

D. Tonini et al, Morphology of niobium films sputtered at different target-substrate angle, SRF99, THP11

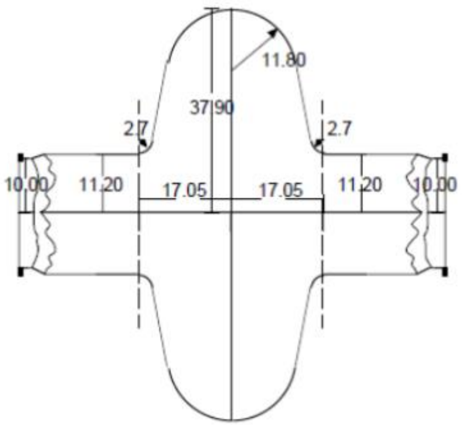
Angle of incidence of coating

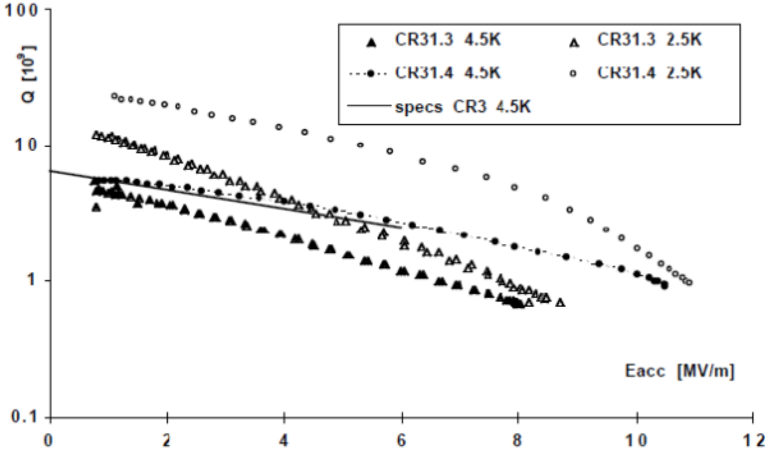
β = 0.48





β = 0.8





C. Benvenuti et al, Production and test of 352 MHz Niobium Sputtered Reduced Beta cavities, 1997, SRF97D25

Outline

- Motivation for thin films in SRF cavities
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- State of the art in Nb thin films (accelerators using thin film technology)
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- **R&D on Nb films**

Nowadays the research is mainly focused on film morphology improvement

Next generation Nb films

ALL film properties are a direct consequence of the film structure, defect/impurity content… thus the technique, environment, substrate are key factors

Full control of the deposition process & tailored SRF performance

UNDERSTANDING OF

- The **chemistry** of the involved species
  - **Reactivity**
  - **Stoichiometric sensitivity**
  - Reaction process **temperatures**
- stal structure **dependence on substrate structure**
- **uence of deposition energy** on resulting structure
- **sitivity to** the presence of **contaminating species, defects**
- **ilization** of desired film against subsequent **degradation**

Careful **characterization of the attained composition and microstructure** (RHEED, STM, XRD, EBSD, AFM, optical profilometry, XPS, SIMS,TEM, FIB).

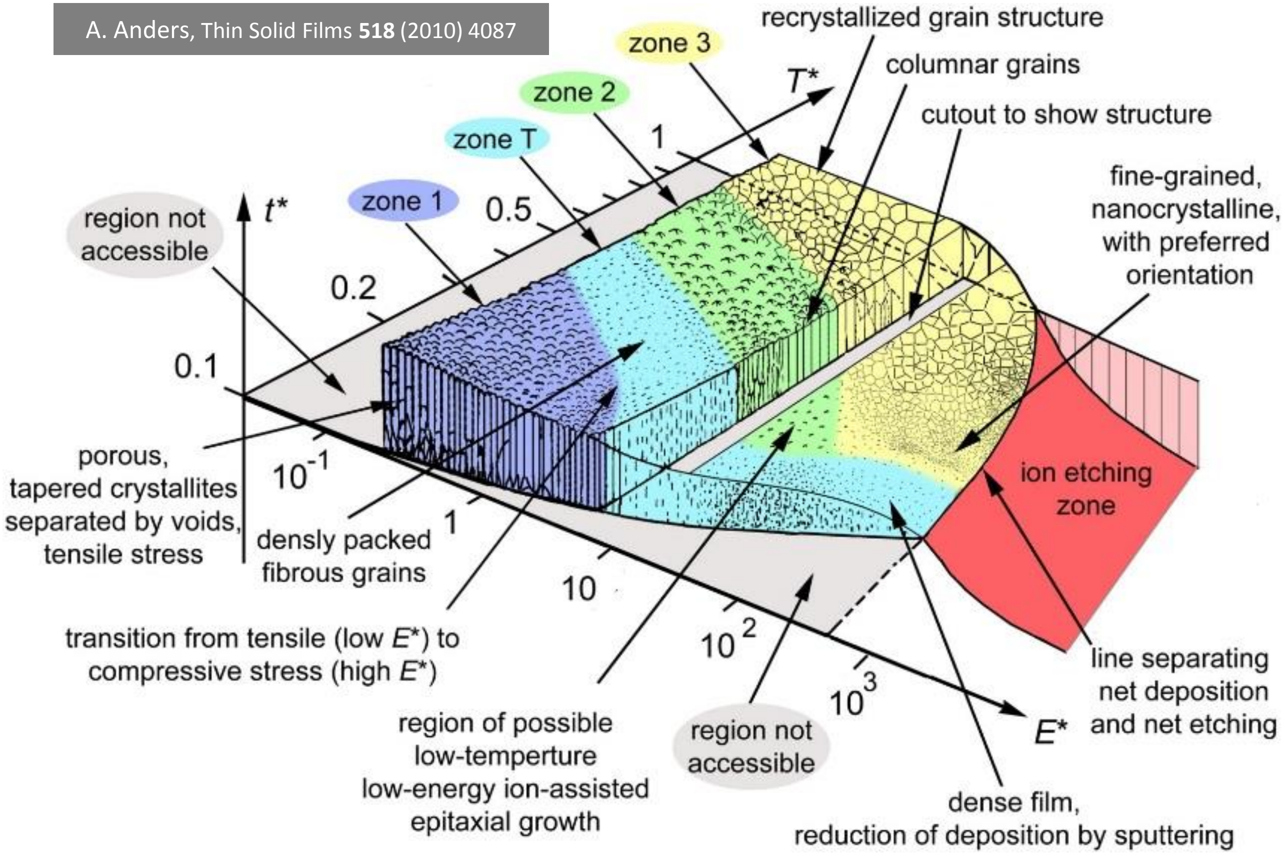
Close association with **resulting RF surface impedance & superconducting** properties (λ, Δ, T_c, H_c, RRR)

A-M Valente, SRF2017 Tutorials

Energetic Condensation

Generalized Structure Zone Diagram

A. Anders, Thin Solid Films **518** (2010) 4087



Additional energy provided by fast particles arriving at a surface:

- residual gases desorbed from the substrate surface
- chemical bonds may be broken and defects created thus affecting nucleation processes & film adhesion
- enhanced mobility of surface atoms

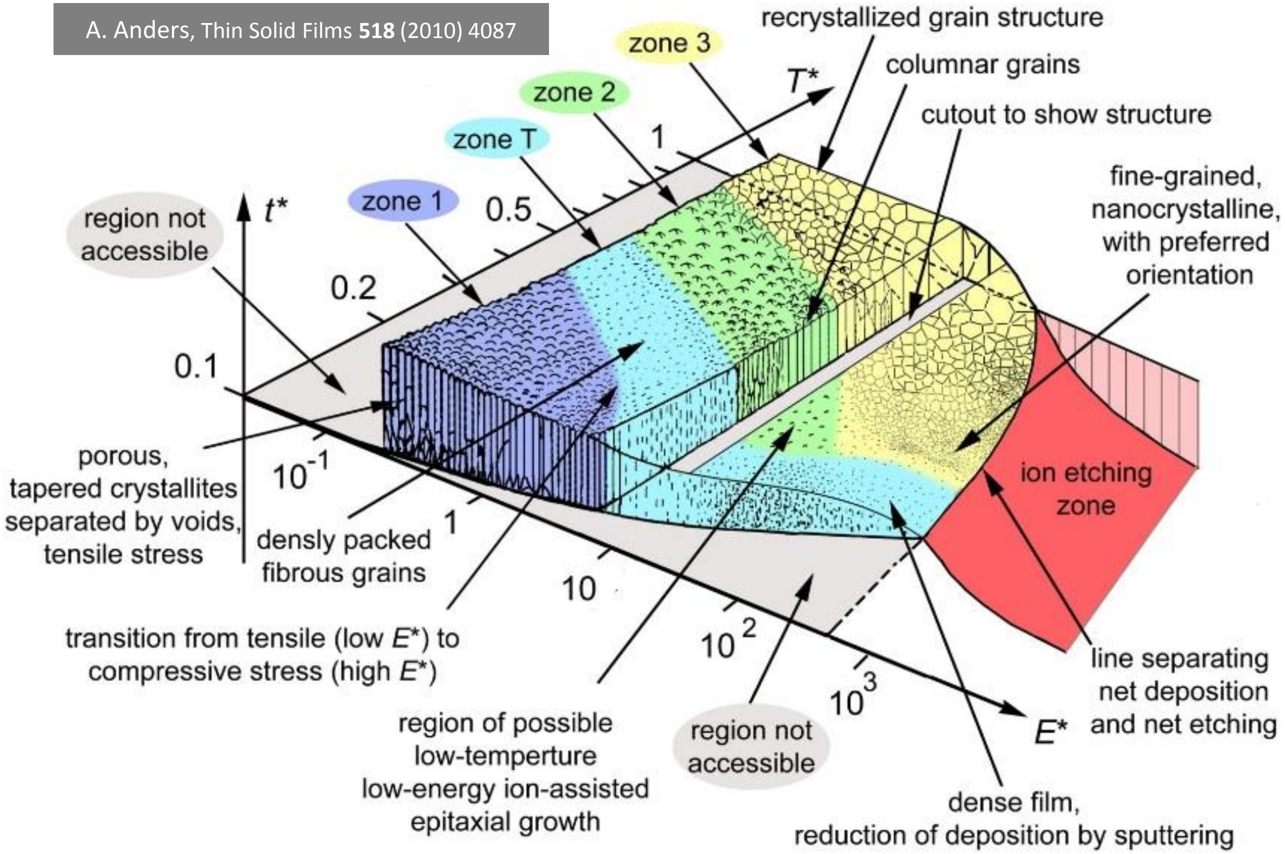
Changes & control in:

- Film density
- morphology
- microstructure
- Stress
- low-temperature epitaxy

Energetic Condensation

Generalized Structure Zone Diagram

A. Anders, Thin Solid Films **518** (2010) 4087



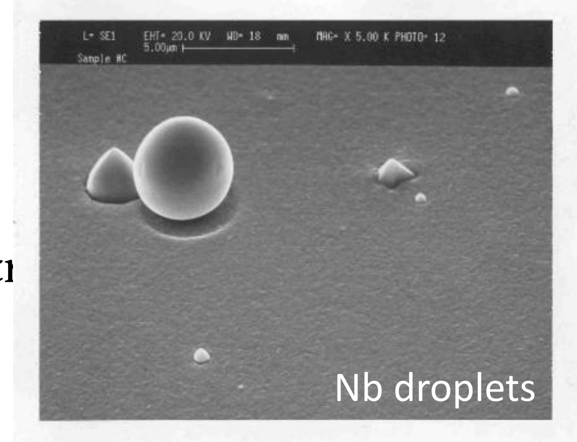
A variety of techniques with distinct technologies

- Vacuum Arc Plasma & Coaxial Energetic Deposition (CED)
- Electron cyclotron Resonance (ECR)
- High Impulse Power Magnetron sputtering (HiPIMS)

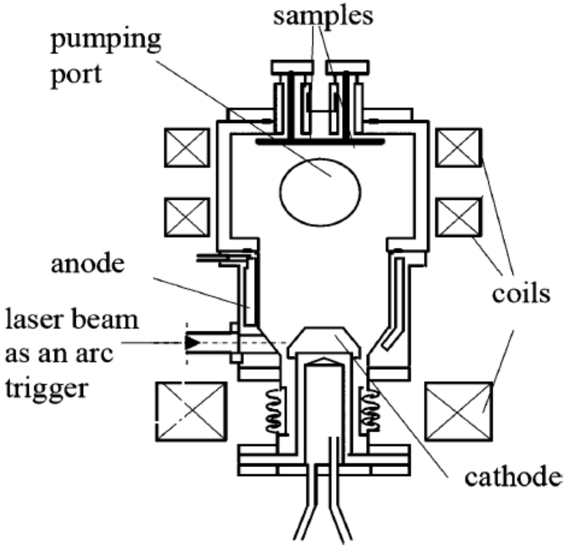
Cathodic Arc Deposition

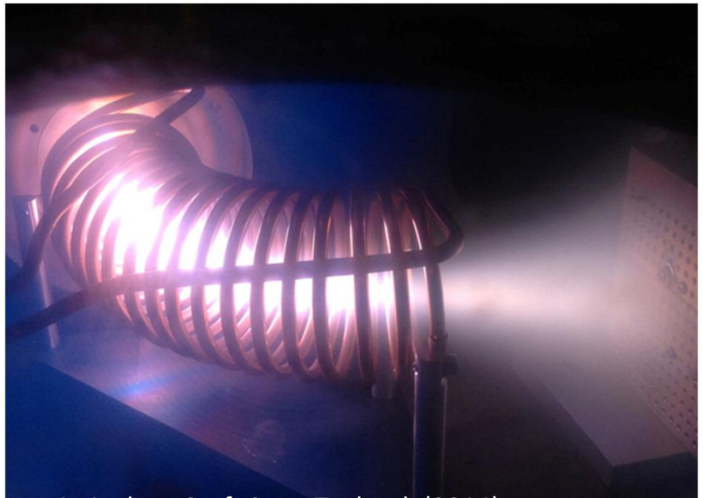
INFN/Poland CARE program (R. Russo et al.)
Alameda Applied Science Corporation (M. Krishnan et al.)

Cathodic Arc Deposition



- The Nb vapor is almost **fully ionized**
- In the plasma arc an electric discharge is established directly onto the Nb target, producing a plasma plume from which ions are extracted and guided onto the substr by a bias and/or magnetic guidance
- **Magnetic filtering** (and/or arc pulsing) is also necessary to remove **droplets**
- A **trigger** for the arc is necessary: either a third electrode, or a laser
- Arc spot moves on the Nb cathode at about 10 m/s
- Arc current is 100-200 A (~ 35 V)
- Voltage bias on samples 20-100 V

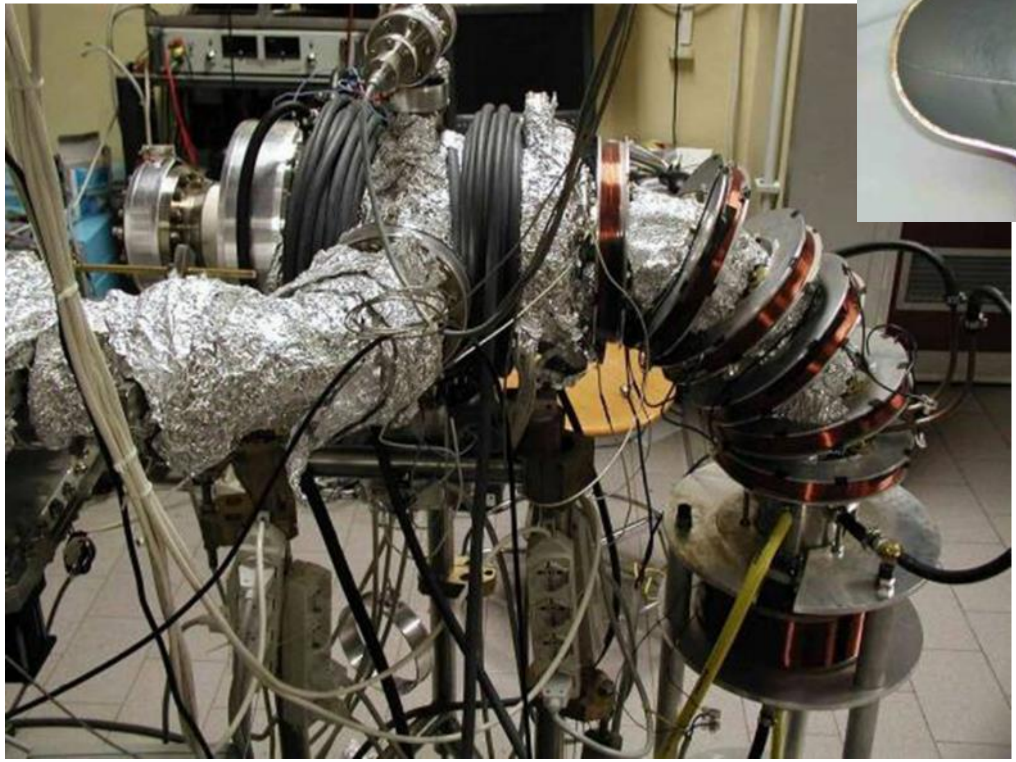


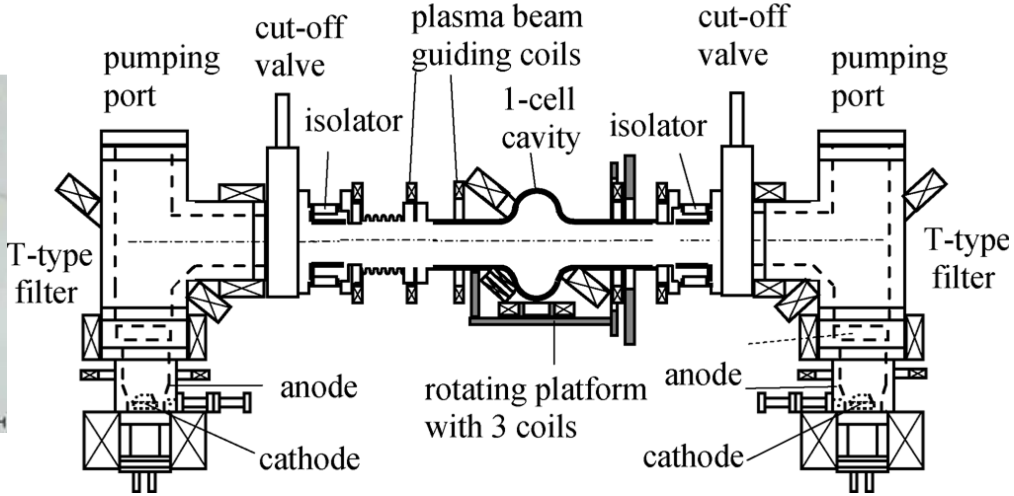

A. Anders, Surf. Coat. Technol. (2014),

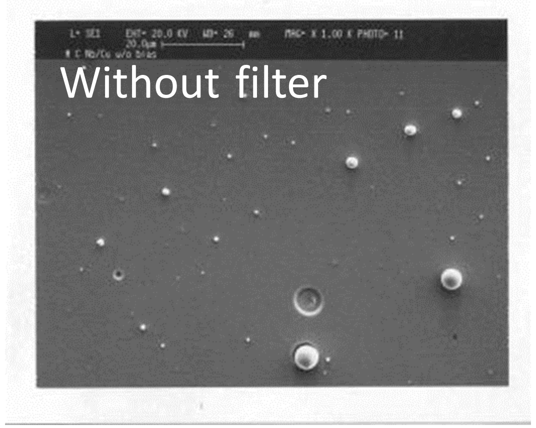
RUSSO *et al.*: NIOBIUM COATING OF CAVITIES USING CATHODIC ARC, IEEE TRANSACTIONS ON APPLIED SUPERCONDUCTIVITY, VOL. 19, NO. 3, JUNE 2009

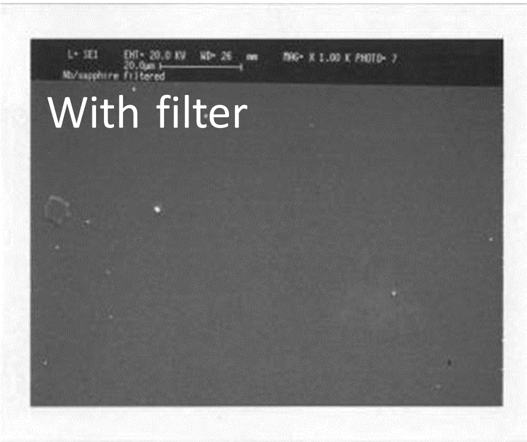
UHV Arc Nb thin films, INFN/Poland CARE project

- RRR up to 80 was reported with substrate heated to 200°C
- No cavity measured







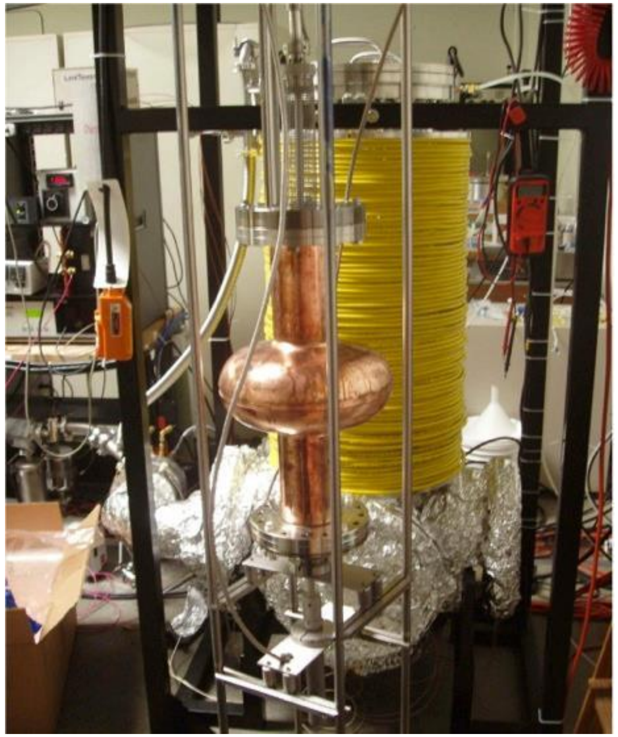


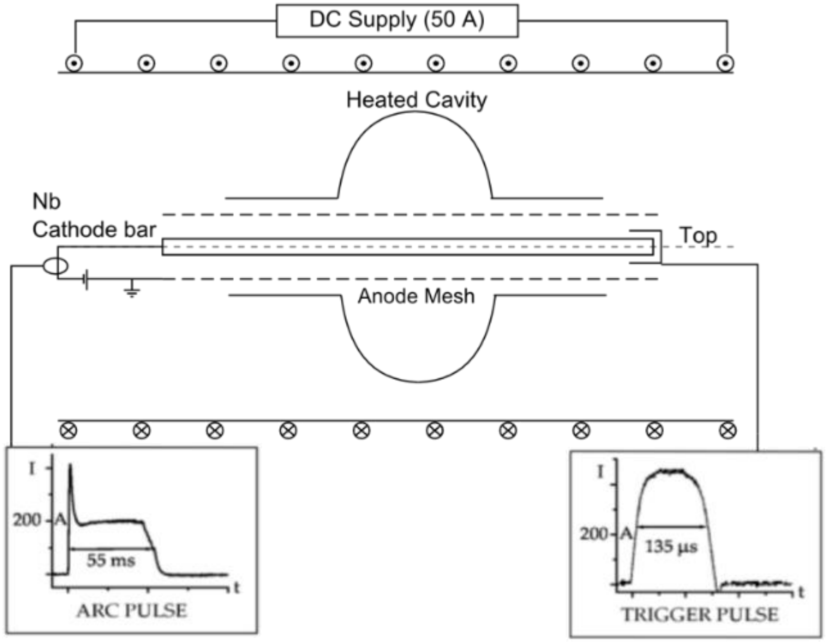
RUSSO *et al.*: NIOBIUM COATING OF CAVITIES USING CATHODIC ARC, IEEE TRANSACTIONS ON APPLIED SUPERCONDUCTIVITY, VOL. 19, NO. 3, JUNE 2009

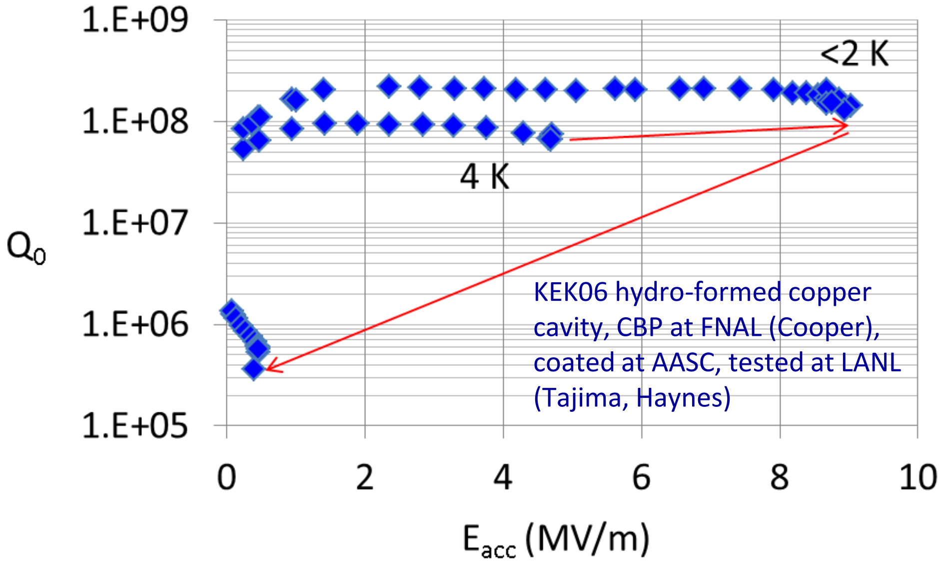
RUSSO, Thinfilms Workshop , LNL 2006

Coaxial Energetic Deposition (CED™) - AASC

- CED coater uses "welding torch" technology
- Arc source is scalable to high throughputs for large scale cavity coatings
- ~1 monolayer/pulse ~0.2 ms
- Good structure, RRR but presence of macro-particles







The difference between 4K and 2K is smaller than expected: BCS resistance should be about 40x less if all the surfaces are Nb.
Suggests presence of areas not well coated well and lossy presence of macro-particles

M. Krishnan, Thinfilms Workshop , LNL 2014

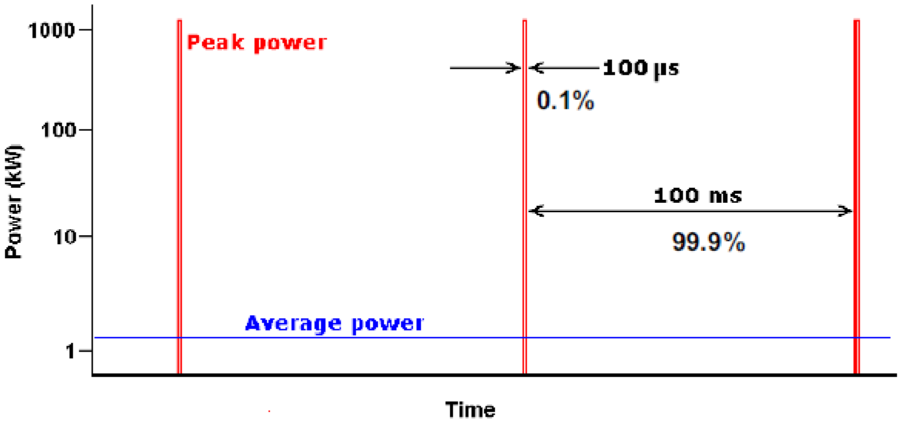
HiPIMS

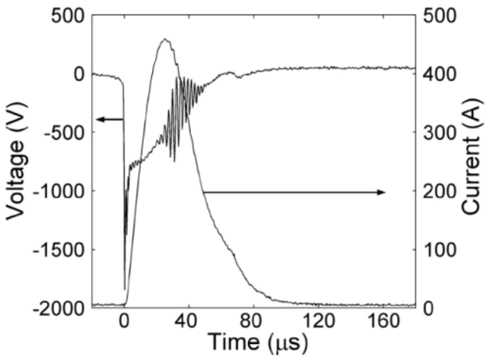
CERN (G. Rosaz et al.)
Jefferson Lab (A-.M. Valente et al.)
STFC ASTeC (R. Valizadeh et al.)
Siegen University (M. Vogel et al.)

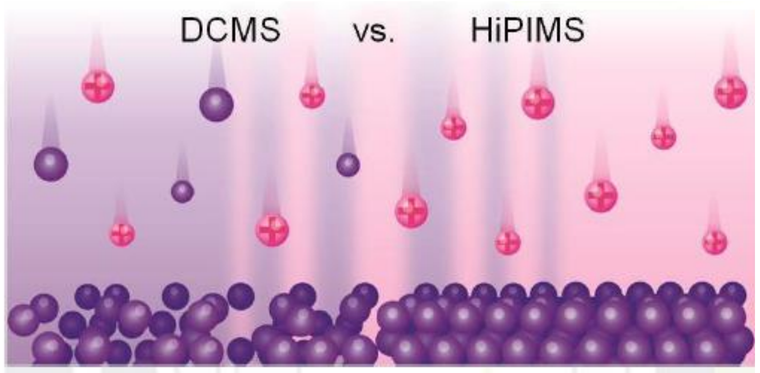
Lawrence Berkeley National Laboratories (A. Anders et al.)

HiPIMS

- **Pulsed sputtering** where the peak power exceeds the average power by typically two orders of magnitude
- The target material is **partially ionized**
- Large concentration of ions producing **high-quality homogeneous films**
- Possibility to self sustain discharge







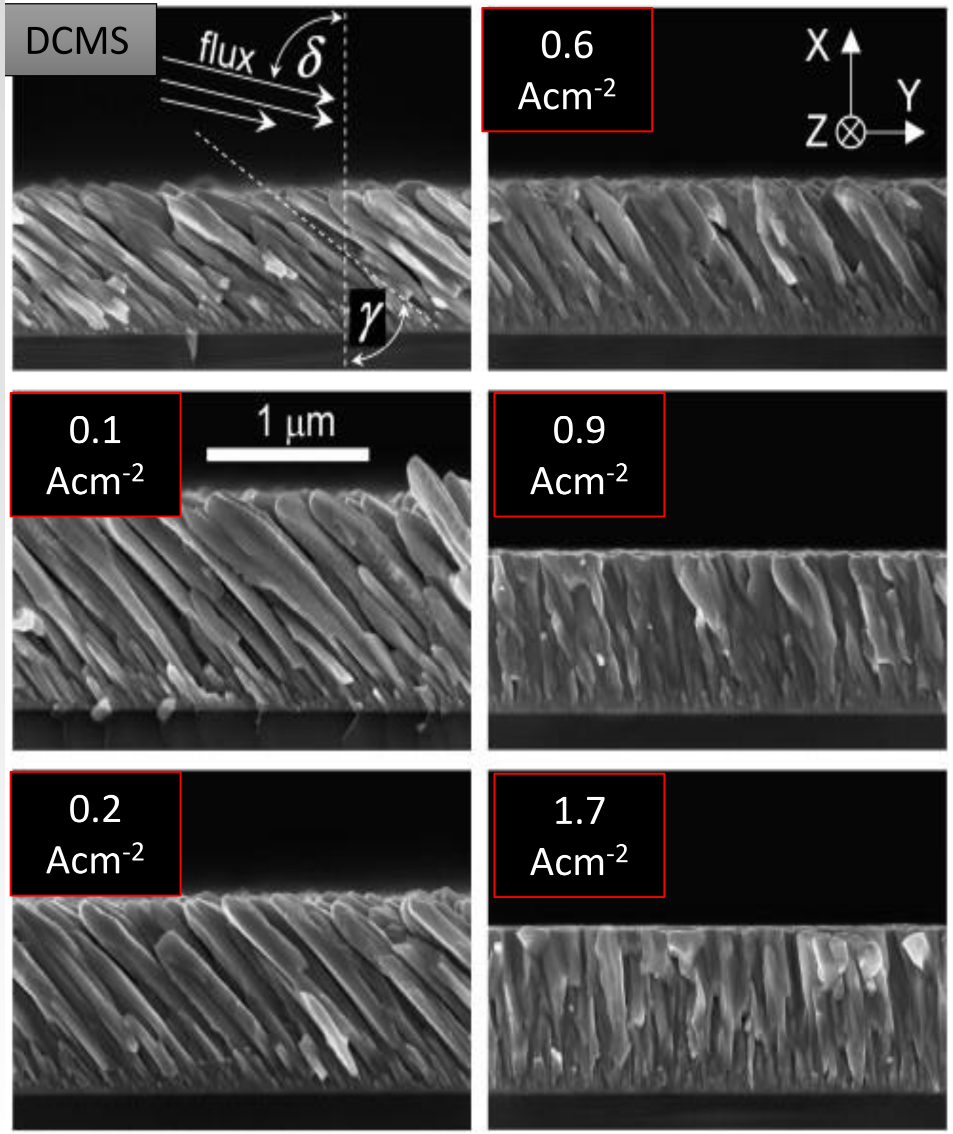
Very high purity
Excellent adhesion
better (normal) conductivity,
Large crystal grains, low defect density
Suppression of fiber structure
Superior density
Decreased roughness
Homogeneous coating even on complex-shaped surfaces
Phase composition tailoring
Interface engineering

Lower coating rate :
ions captured at the cathode

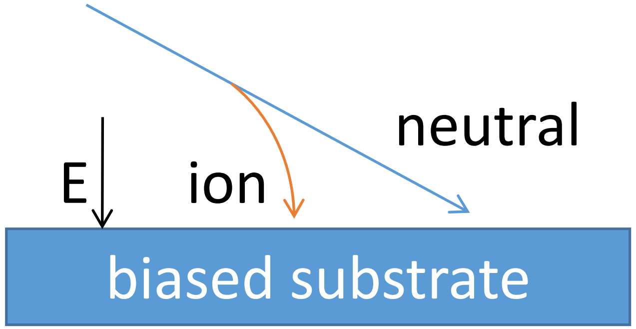
Very sensitive to cathode surface state (roughness), induced arcing

J. Böhlmark et al., J. Vac. Sci. Technol. A 23 (2005) 18

Conformal Coating



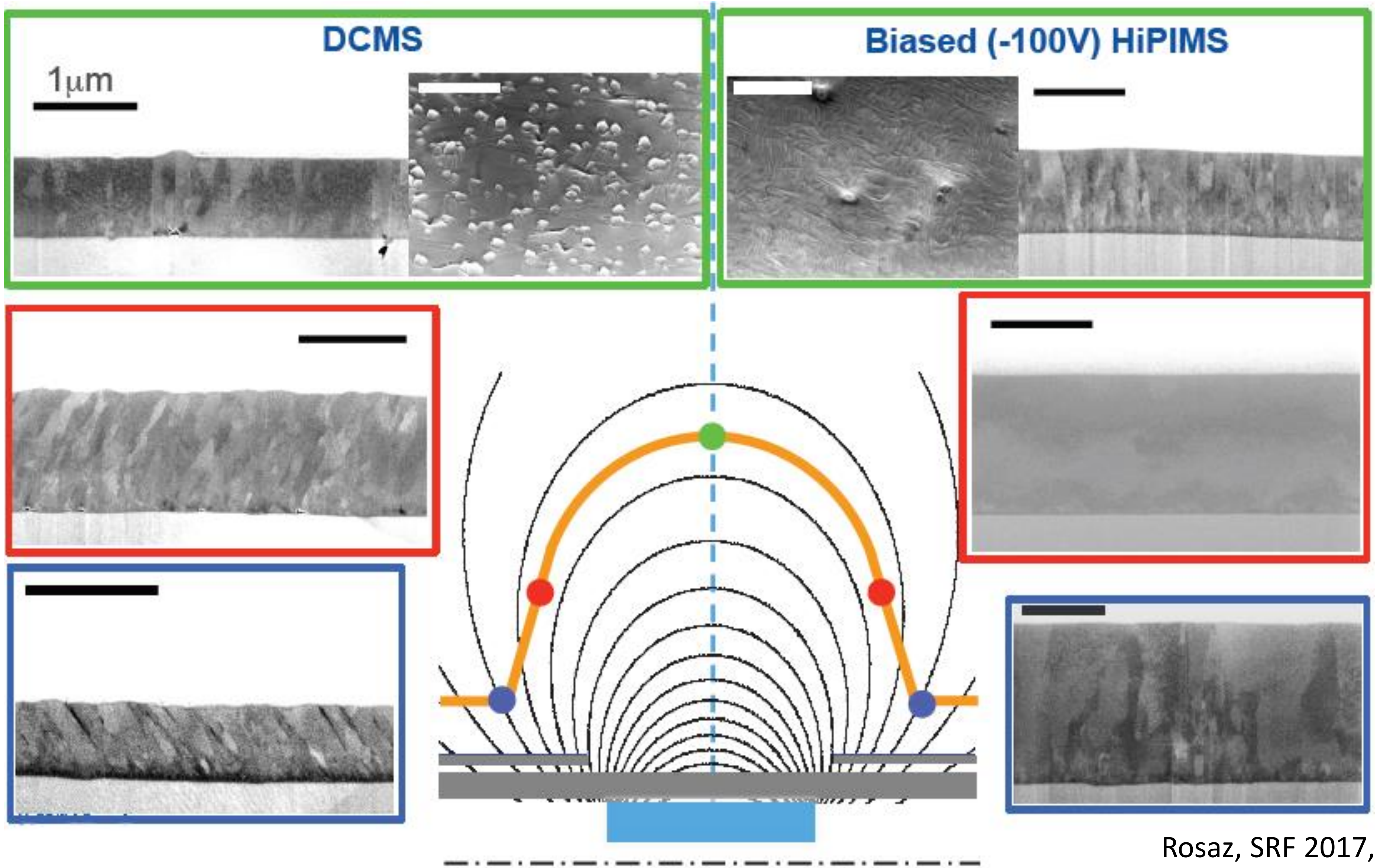
Inclination of columns is reduced at high target current densities due to high ion-to-neutral ratio



CrN Glancing Angle Deposition

G. Greczynski, *et al.*, Thin Solid Films 519 (2011) 6354.

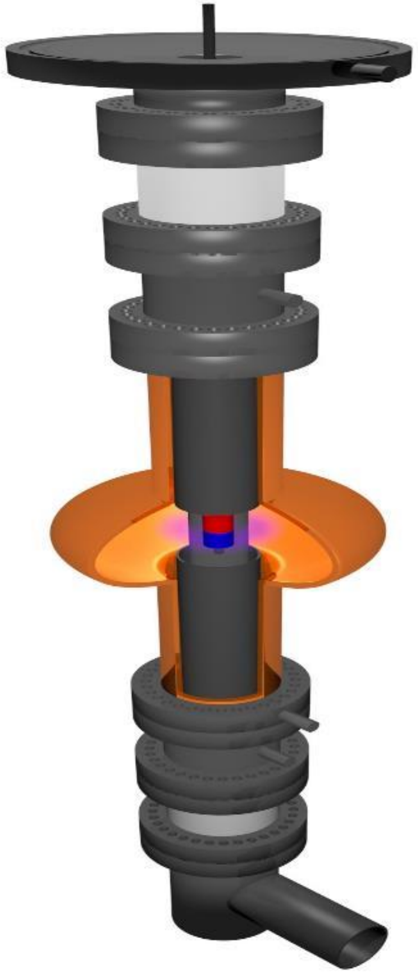
Conformal HiPIMS @CERN



Rosaz, SRF 2017, Lanzhou (China)

CERN HiPIMS Setup

1.3 GHz cavity coating setup



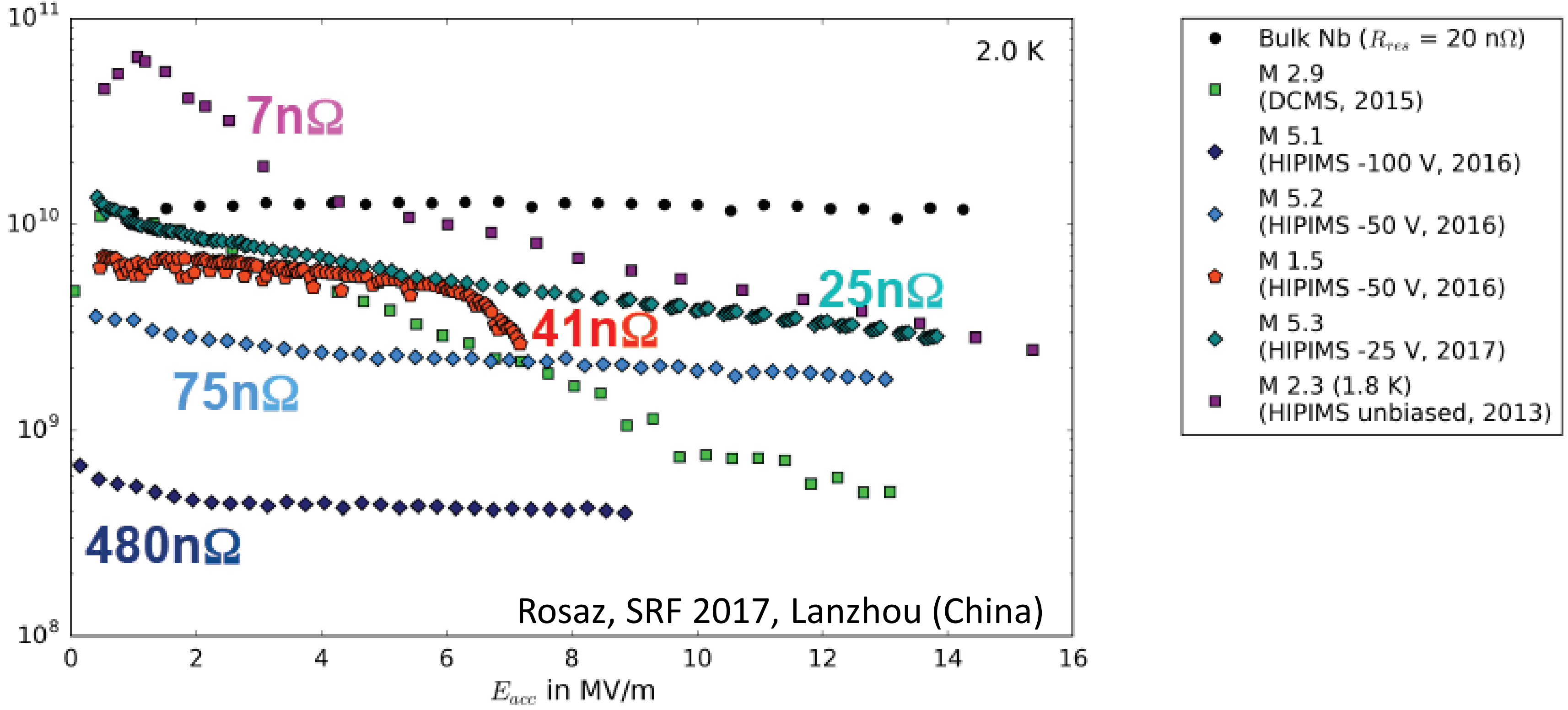
Nb cathode with permanent magnets inside and Nb anodes

HiPIMS discharge

- **Same hardware as for DCMS**
- Pulsed Power supply
  - 1% duty cycle
  - Short pulses: 200 μs
  - High peak current (200 A vs 3 A for DCMS)
  - High peak power (80 kW peak for 1kW avg)
- Ionization of sputtered species
- Lower coating rate than DCMS

Courtesy of G. Rosaz (CERN)

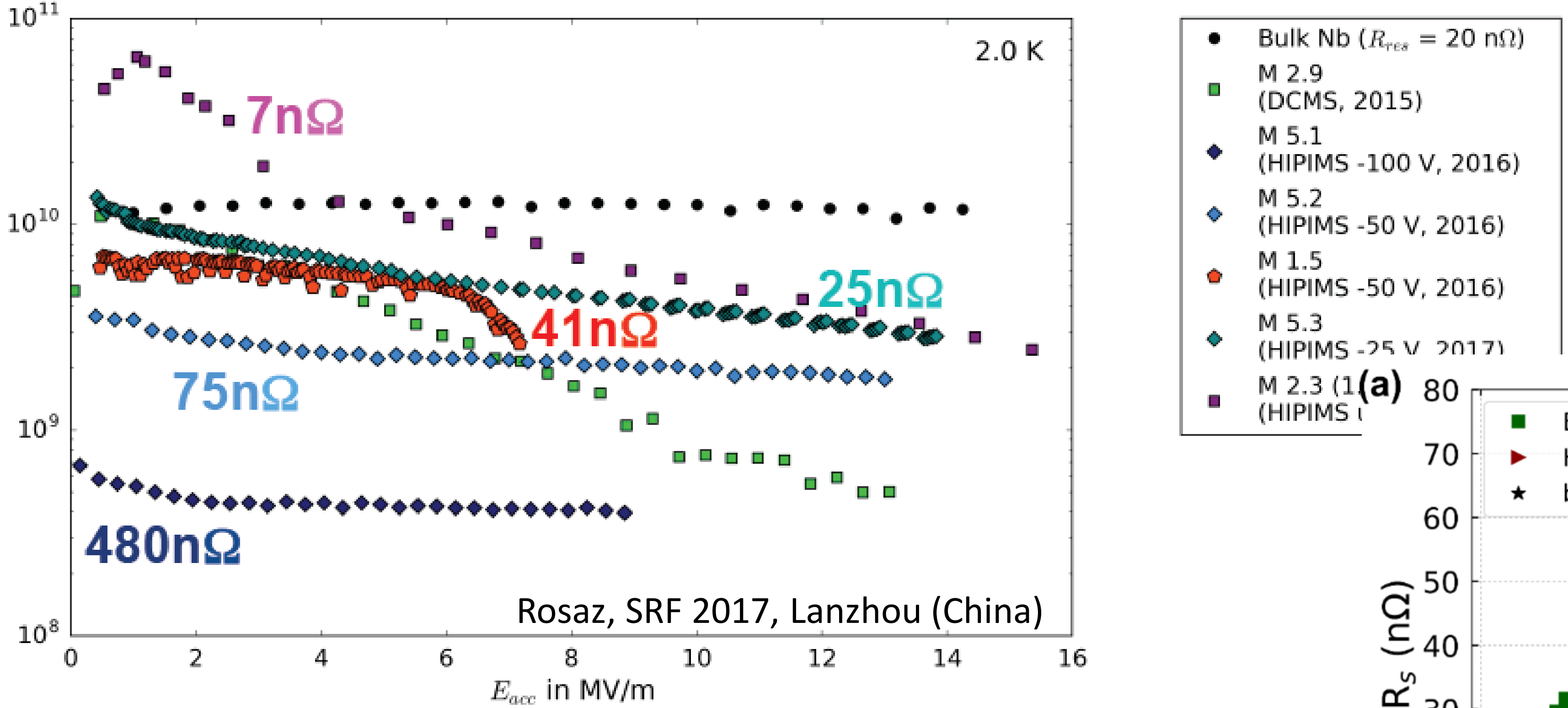
HiPIMS Results @ CERN

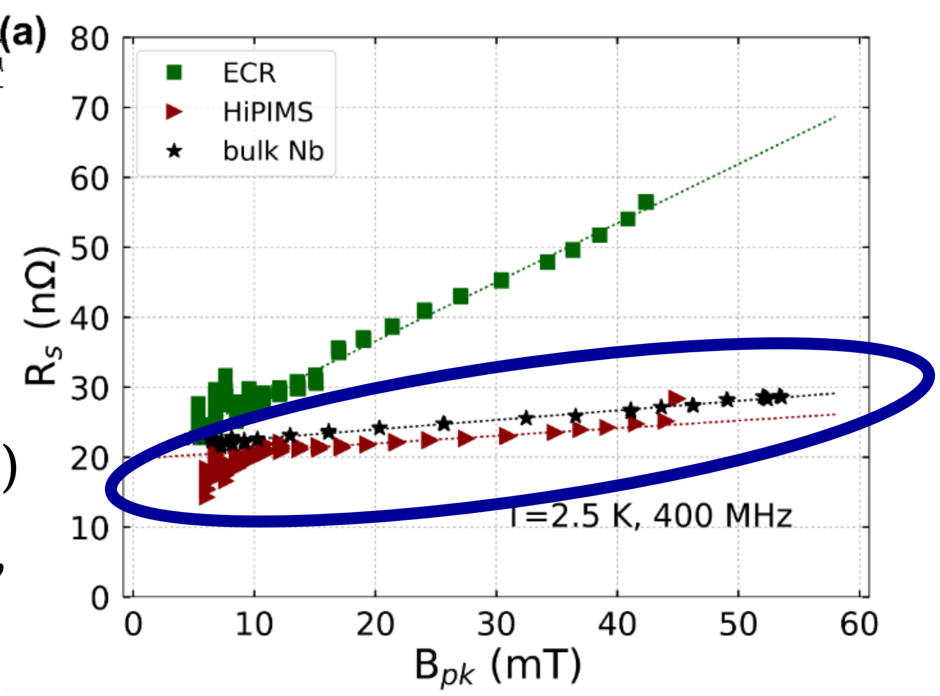


- High Bias does not give good results (gas implantation , stress)
- Lower pressure tends to better performances (contamination, stress)
- **Q-slope looks mitigated vs DCMS coating**

Courtesy of G. Rosaz (CERN)

HiPIMS Results @ CERN





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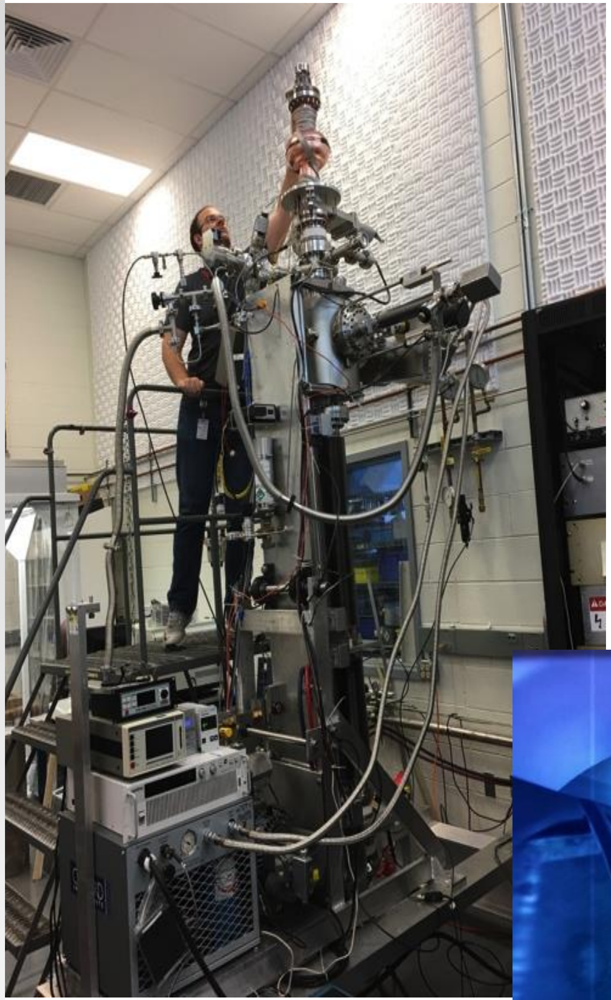
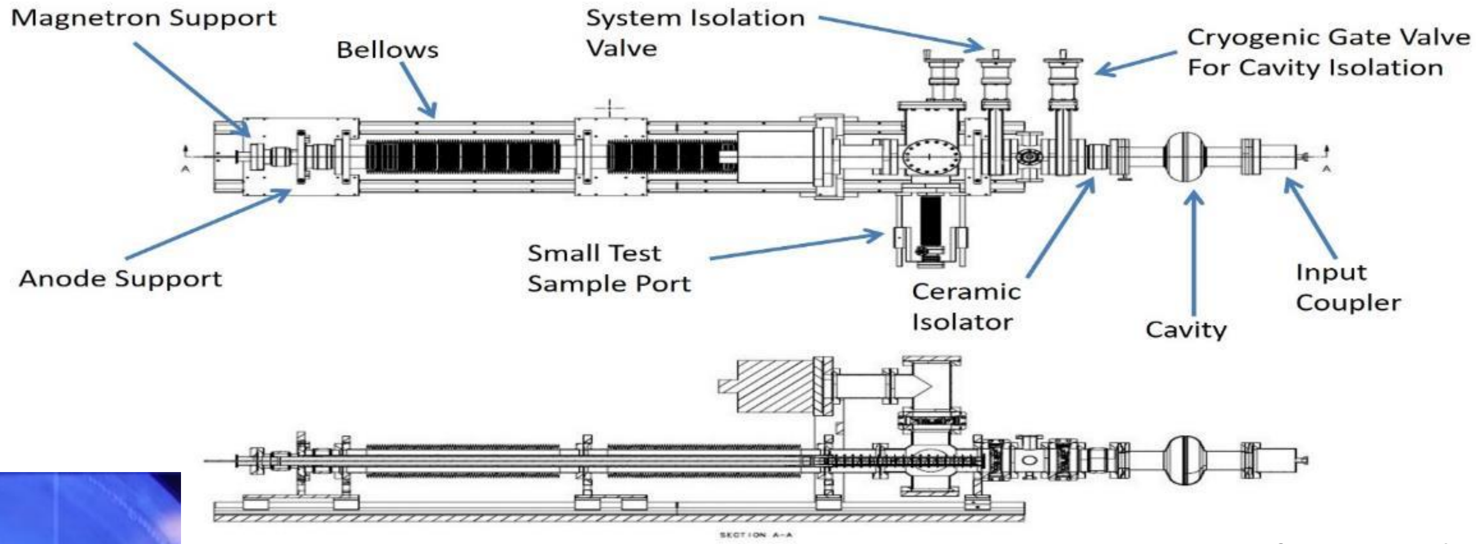
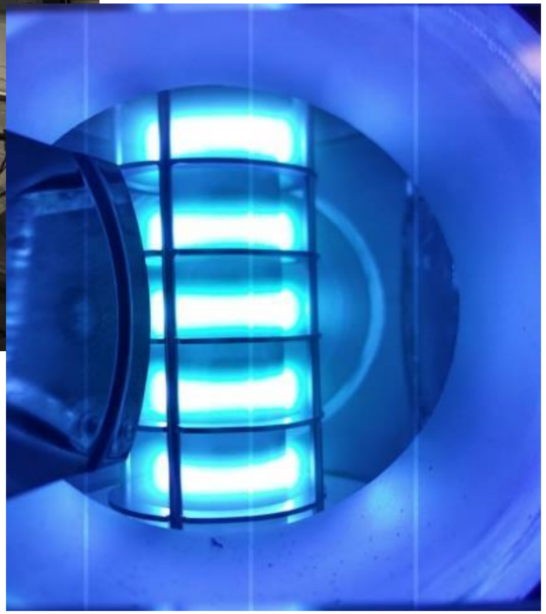
HiPIMS @ JLAB



System re-commissioned
June 2018



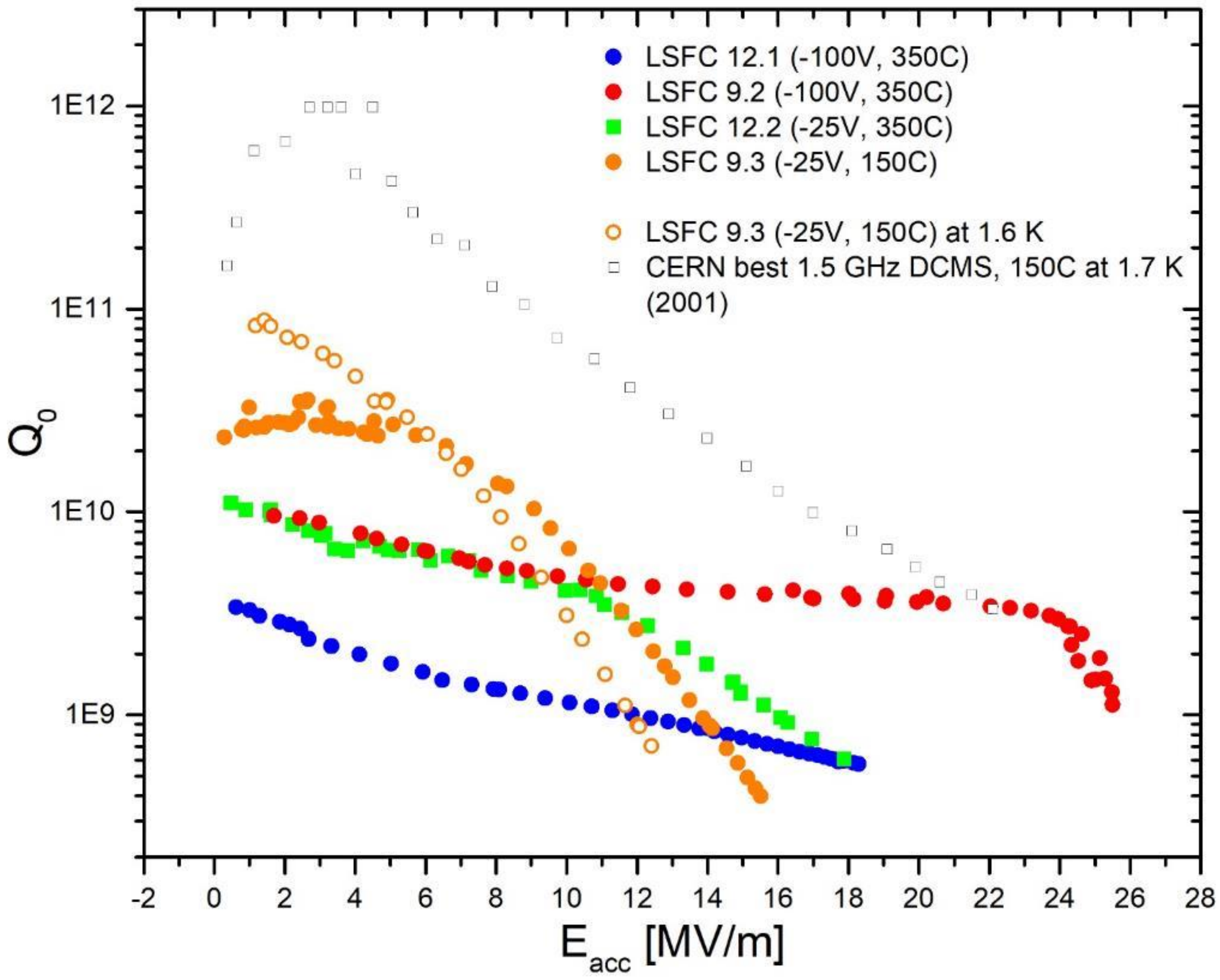
Courtesy of A.-M. Valente



- **Movable** cylindrical Nb **cathode**
- Background pressure in 10^{-9}-10^{-10} Torr
- Coating **temperatures up top 400 °C** under external nitrogen flow
- Kr atmosphere

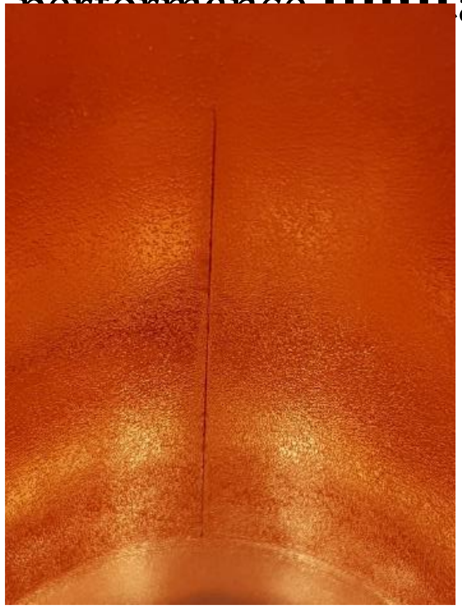
Courtesy of A-M. Valente (JLAB)

HiPIMS Results @JLAB



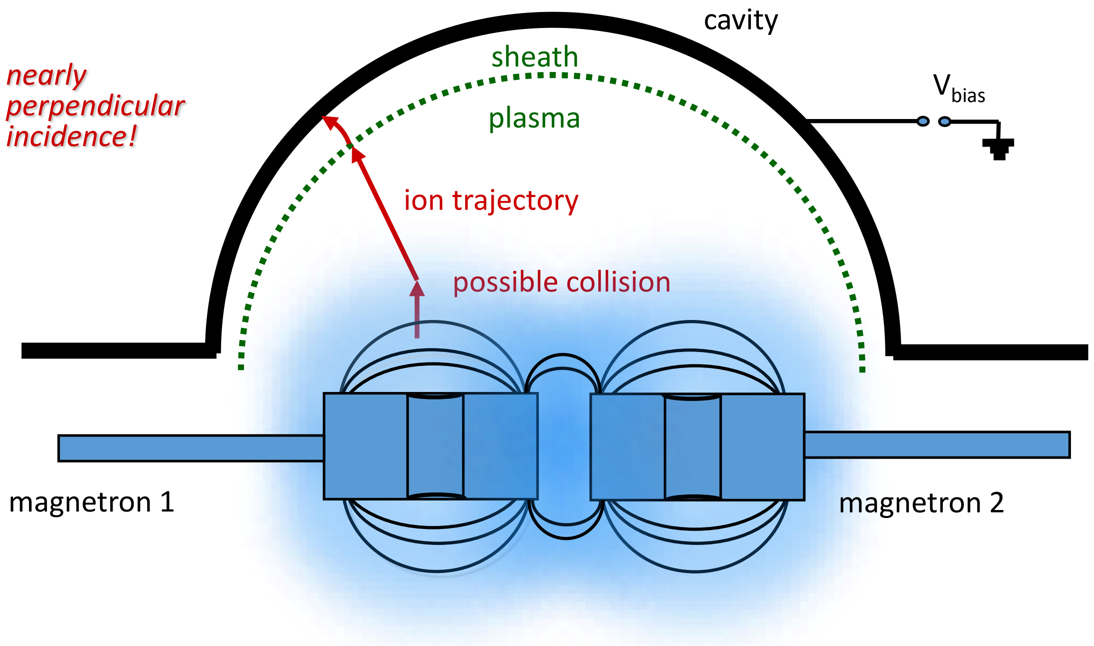
Some HiPIMS Nb/Cu cavities show **mitigation** of the characteristic **Q-slope**

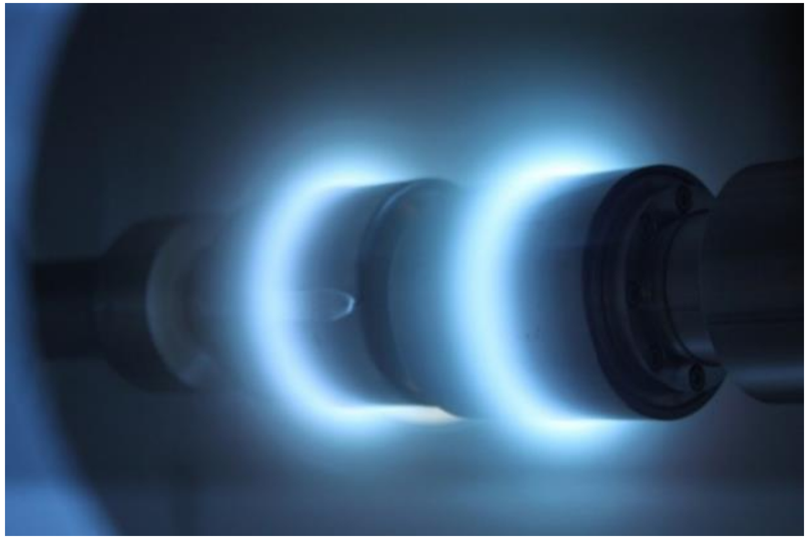
Substrates are a possible **cause** of performance **limitation**



Courtesy of A-M. Valente (JLAB)

HiPIMS configuration @ LBNL



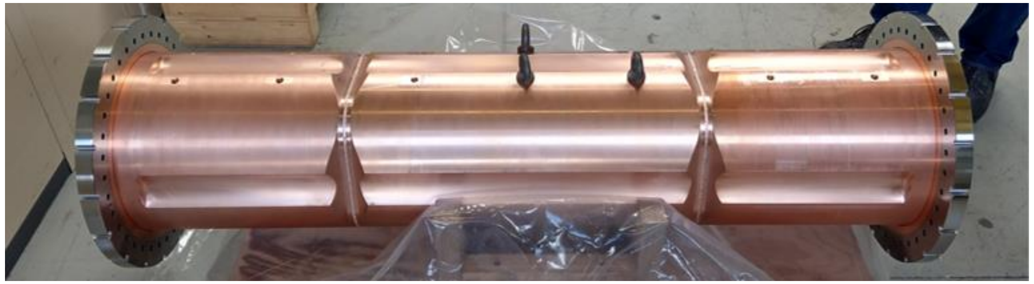


- HiPIMS **Dual Magnetron Configuration**
- Most effective for Biasing & influencing Ion Energies & Trajectories
- High power mode (above runaway threshold)
- Dominated by Nb emission
- No cavity RF tested

A. Anders, Thinfilms Workshop 2016, JLab

The WOW cavity coating challenge

Wide-Open Waveguide (WOW) crab cavity (Nb/Cu), 1st prototype completed in 2018



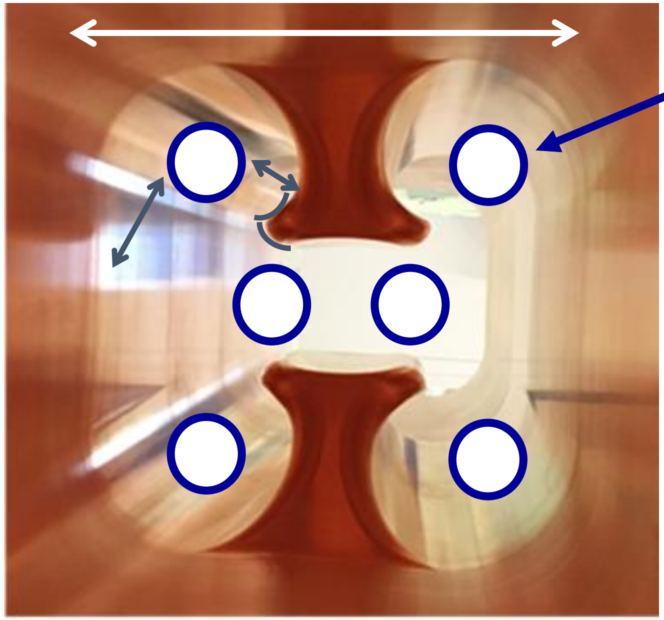
1.4m / 290kg

HiPIMS +
- **negatively biased substrate**
- **positive pulse**

More metal ions

High energy metal ions to densify the film

252mm



x6 Cylindrical magnetrons

- Distances (20 – 80 mm)
- Angles of incidence (0 – 90°)

F. Avino (CERN), TTC Meeting, CERN 2020

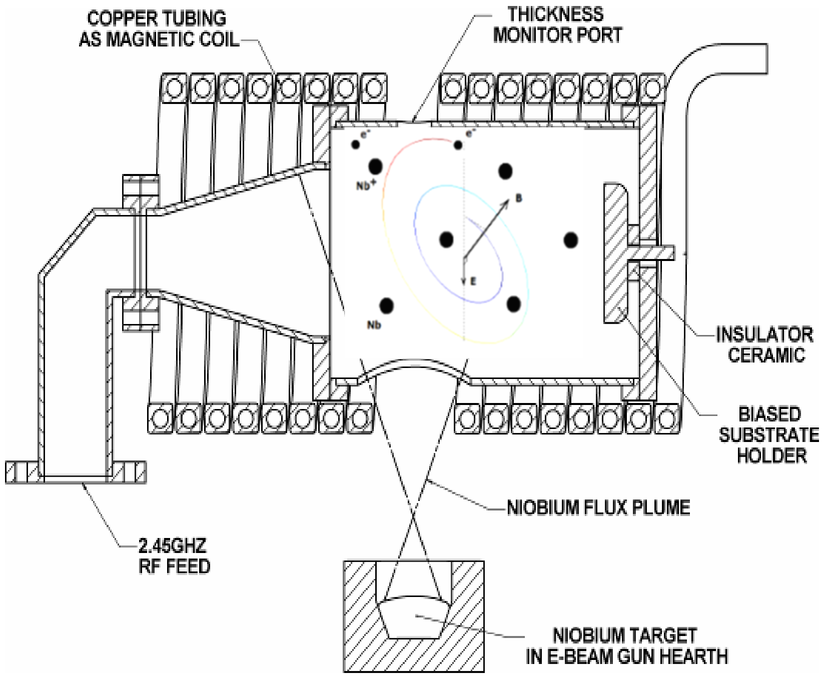
ECR

Jefferson Lab (A-.M. Valente et al.)

Energetic Condensation with ECR @JLAB

ECR DEPOSITION PROCESS

1. Nb is evaporated by e-beam in a separate vacuum chamber
2. Nb vapours are ionized by an ECR process
   - RF power (@ 2.45GHz)
   - Static B ⊥ E_{RF} with ECR condition
3. Nb ion are accelerate to the substrate (cavity) by a bias voltage



No working gas

Singly charged ions (64eV) produced in vacuum

Controllable deposition energy with Bias voltage

Excellent bonding , **No macro particles**

Good conformality

Scalability?

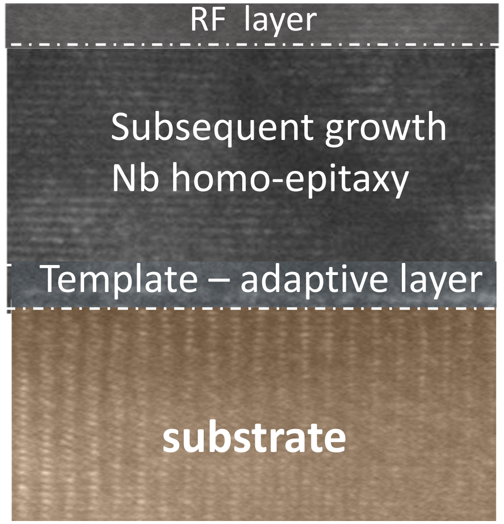
ECR film properties

	Substrate	RRR max
Insulating	a-Al_2O_3	591
	r-Al_2O_3	725
	c-Al_2O_3	247
	MgO (100)	188
	MgO (110)	424
	MgO (111)	270
	Al_2O_3 ceramic	135
	AlN ceramic	110
	Fused Silica	84
Metallic	Cu (100)	181
	Cu (110)	275
	Cu (111)	245
	Cu fine grains	193
	Cu large grains	305

Courtesy of A-M. Valente (JLAB)

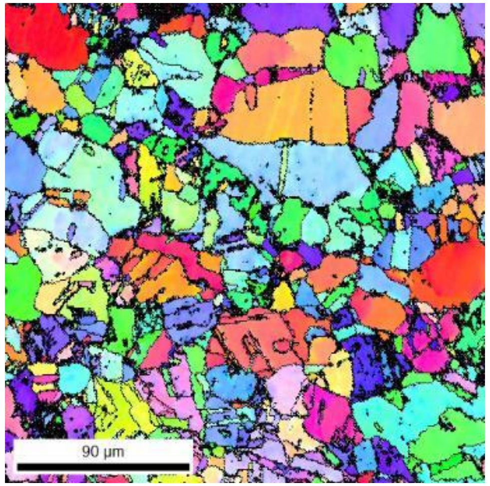
SEQUENTIAL PHASE FOR FILM GROWTH

- Interface
- Film nucleation (184 eV)
- Growth of appropriate template for subsequent deposition (64 eV)
- Deposition of final surface optimized for minimum defect density

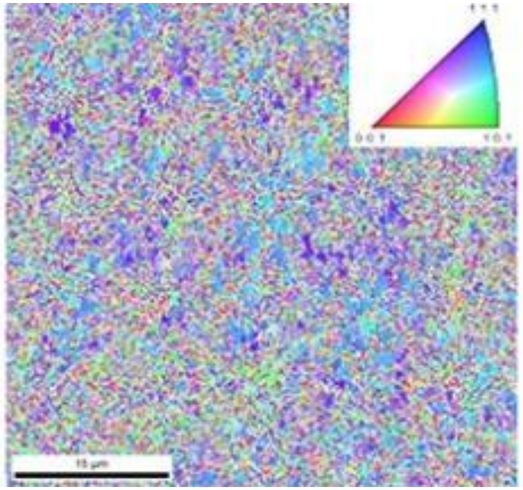


Bulk like properties

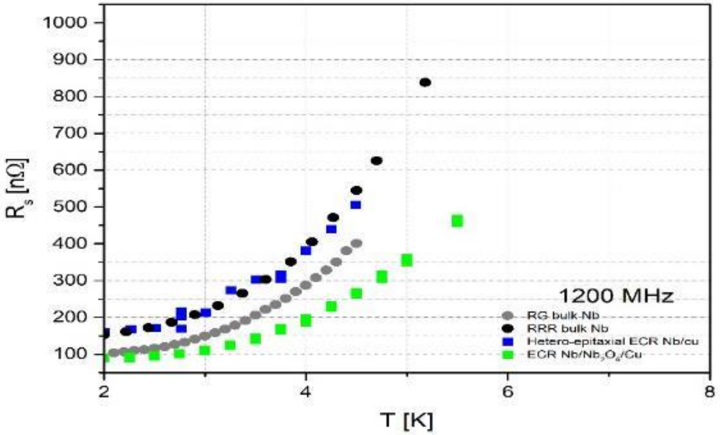
Opportunity for film engineering



Hetero-epitaxial growth



Growth on amorphous interface

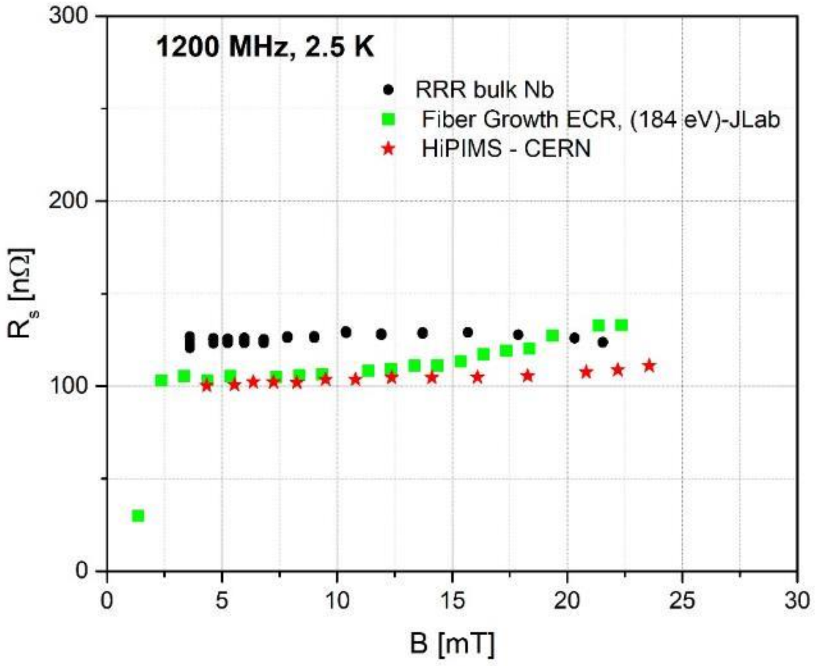


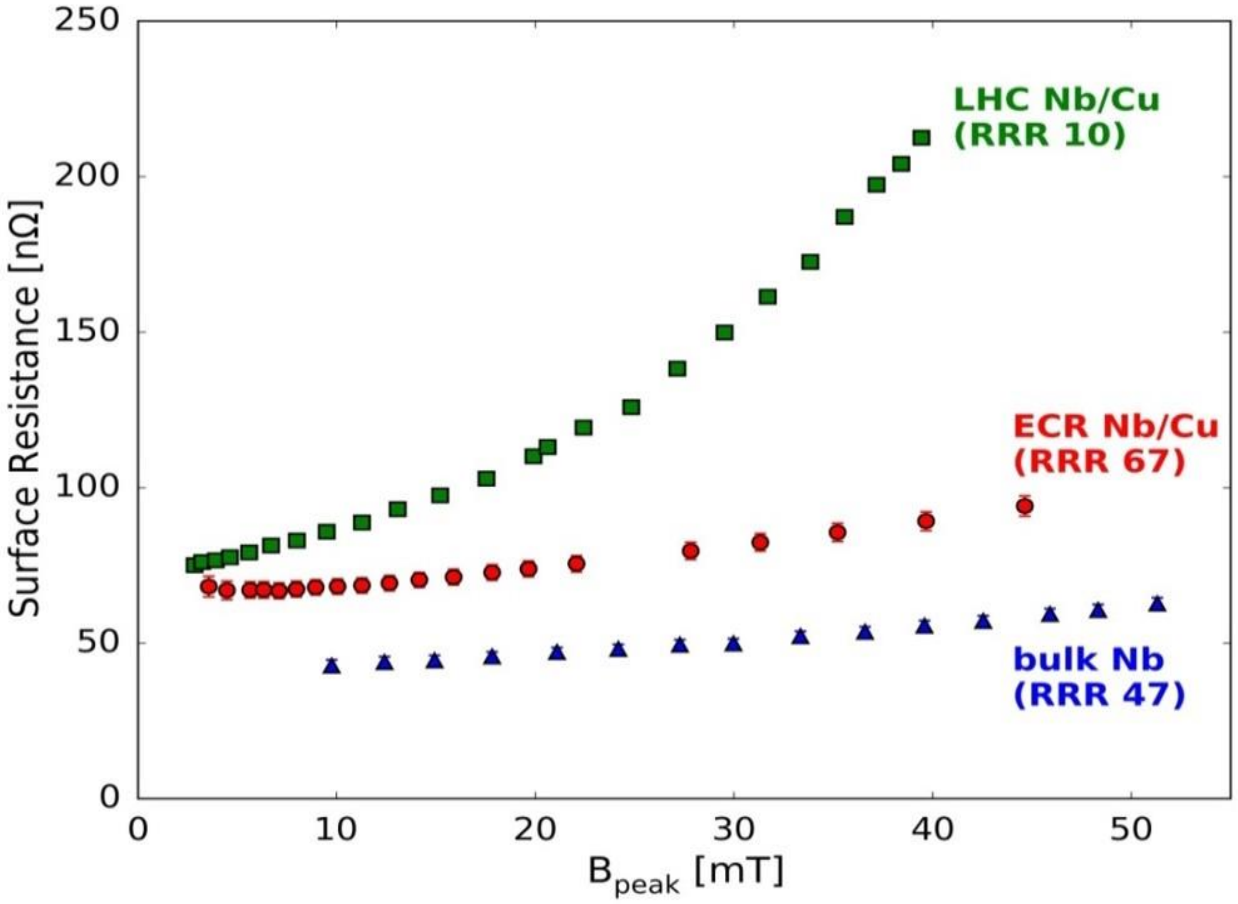
Fiber growth ECR Nb/Cu films perform better than hetero-epitaxial ones

ECR Results

Mitigation of Rs slope possible

Energetic Condensation Nb/Cu films show **similar RF behavior compare to bulk Nb** in QPR measurements



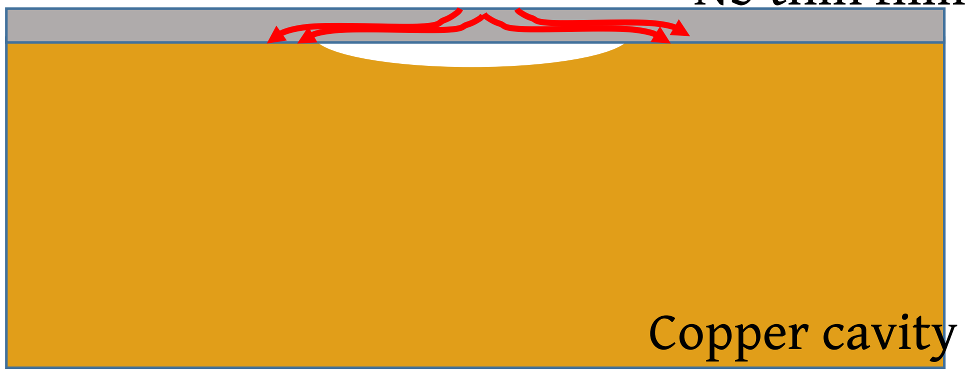


Courtesy of A-M. Valente (JLAB)

THICK FILMS

DCMS @INFN LNL (C. Pira, V. Garcia et al.)
CVD @CORNELL and ULTRAMET
CVD @STFC (R. Valizadeh et al.)

Thick film motivation

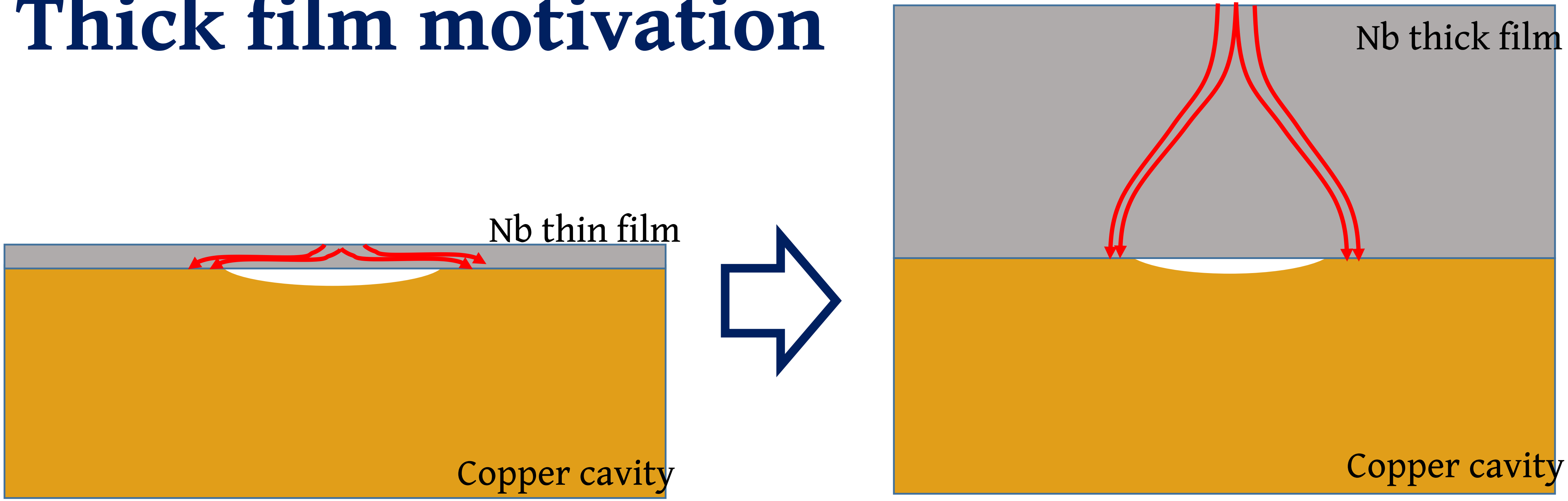


Q-slope is related to local enhancement of the **thermal boundary resistance at the Nb/Cu interface**, due to a poor thermal contact between film and substrate

Theoretical model from Vaglio and Palmieri

V. Palmieri and R. Vaglio, *Supercond. Sci. Technol.*, vol. 29, no. 1, p. 015004, Jan. 2016

Thick film motivation



Q-slope is related to local enhancement of the **thermal boundary resistance at the Nb/Cu interface**, due to poor thermal contact between film and substrate

Theoretical model from Vaglio and Palmieri

V. Palmieri and R. Vaglio, *Supercond. Sci. Technol.*, vol. 29, no. 1, p. 015004, Jan. 2016

Chemical Vapour Deposition

Fundamental sequential steps in every CVD process

1. Convective and diffusive transport of reactants from the gas inlets to the reaction zone
2. Chemical reactions in the gas phase to produce new reactive species and by-products
3. Transport of the initial reactants and their products to the substrate surface
4. Adsorption (chemical and physical) and diffusion of these species on the substrate surface
5. Heterogeneous reactions catalyzed by the surface leading to film formation
6. Desorption of the volatile by-products of surface reactions
7. Convective and diffusive transport of the reaction by-products away from the reaction zone

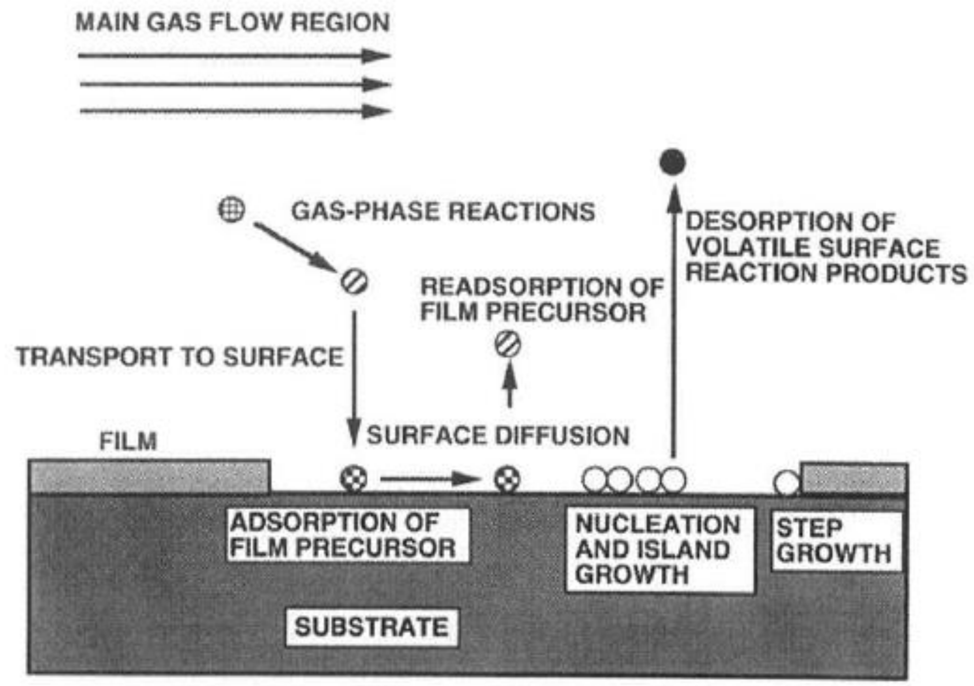


Figure 6-2 Sequence of gas transport and reaction processes contributing to CVD film growth. (From *Chemical Vapor Deposition*, edited by M. L. Hitchman and K. F. Jensen. Reprinted with the permission of Academic Press, Ltd., and Professor K. F. Jensen, MIT.)

CVD Thick film @ Cornell University and Ultramet

Fundamental sequential steps in every CVD process

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5. Heterogeneous reactions catalyzed by the surface leading to film formation
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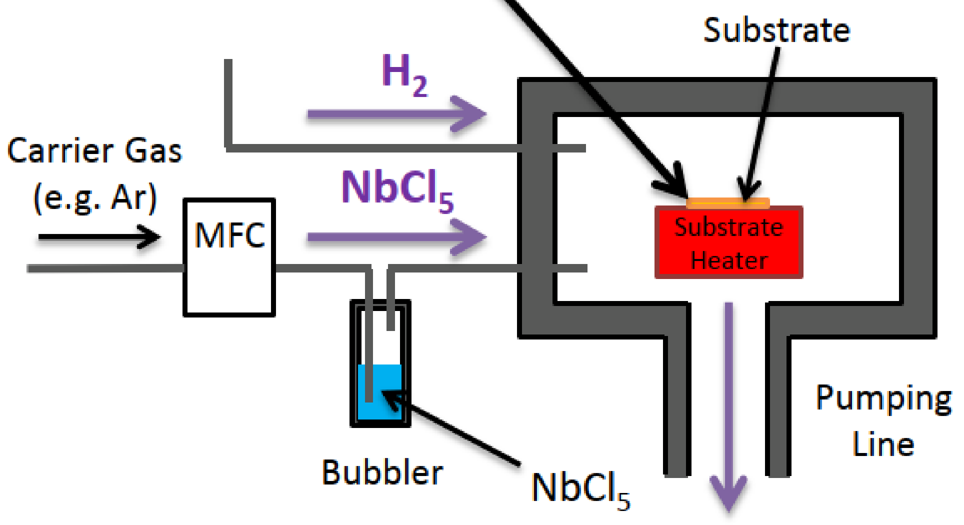
7. Convective and diffusive transport of the reaction by-products away from the reaction zone

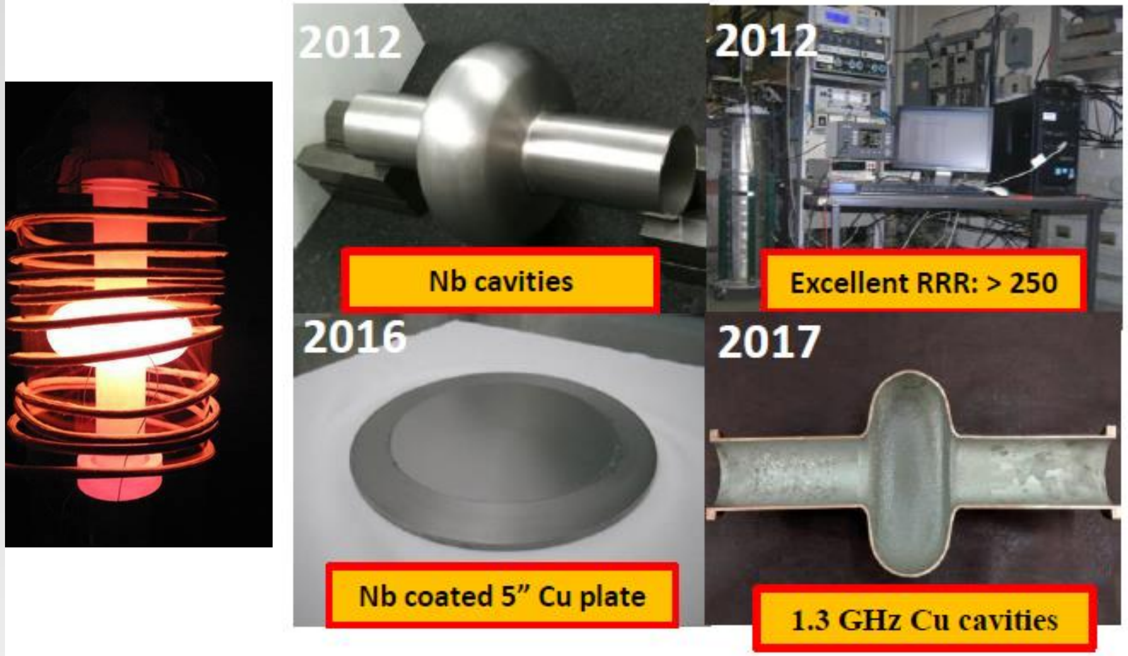
$$2\,NbCl5 + 5\,H2 \rightarrow 2\,Nb + 10\,HCl$$

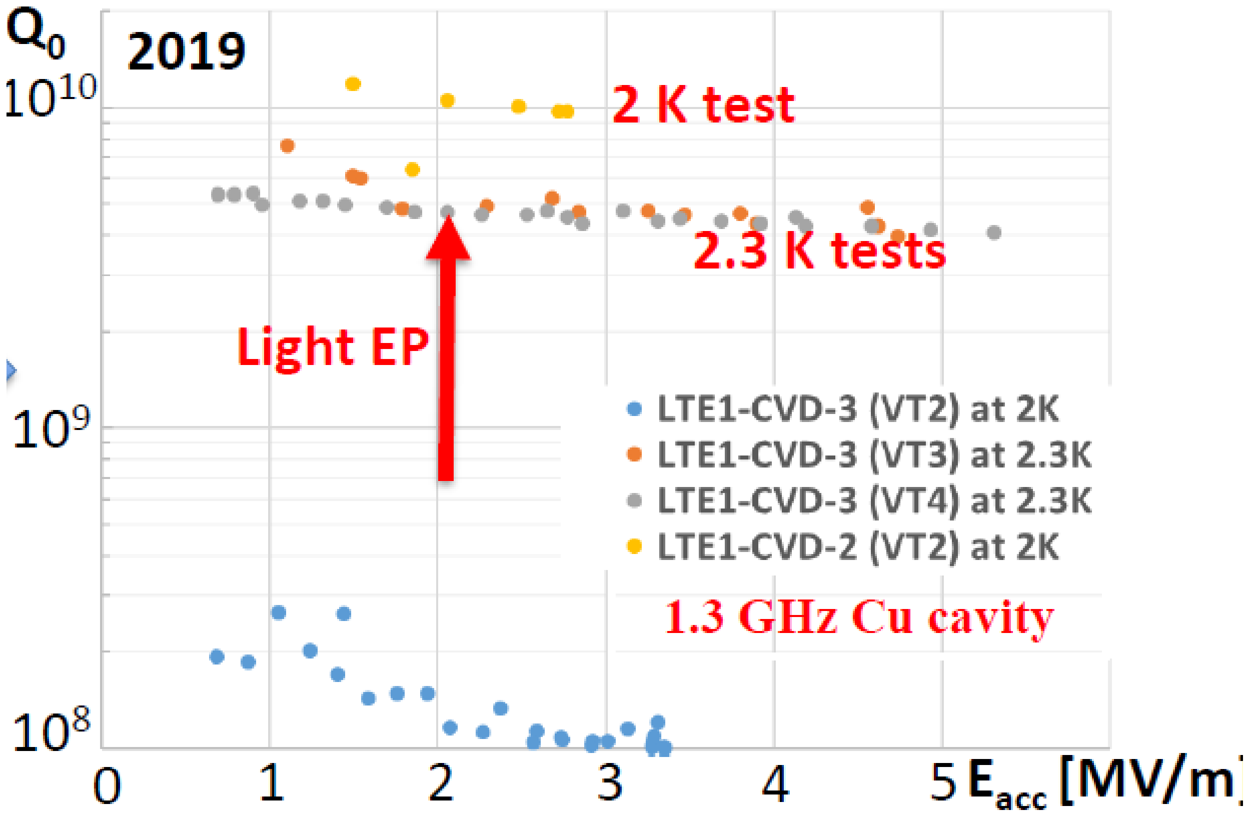


Reactor diagram showing use of $NbCl_5$ to produce CVD niobium

P. Pizzolet al., (STFC) IPAC (2016)

CVD Thick film @ Cornell University and Ultramet





Film optimization & process scale-up

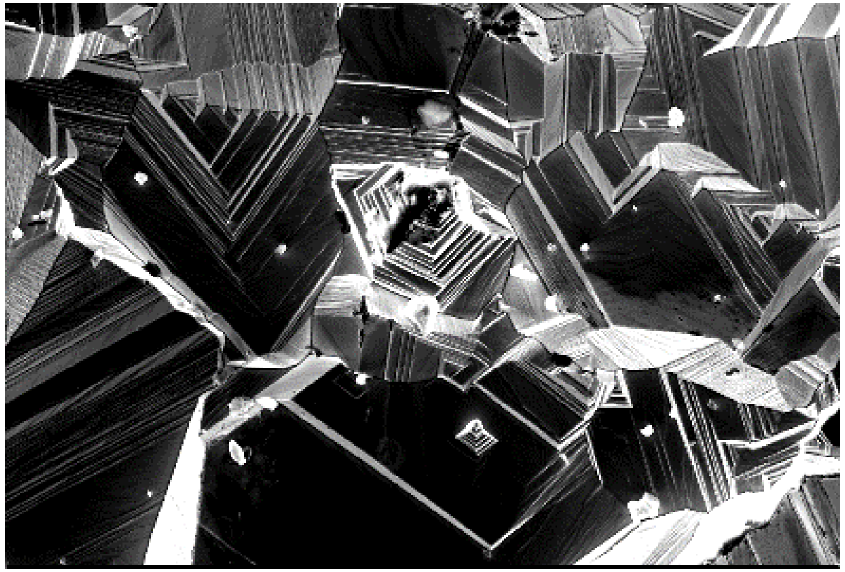
High purity (high RRR)

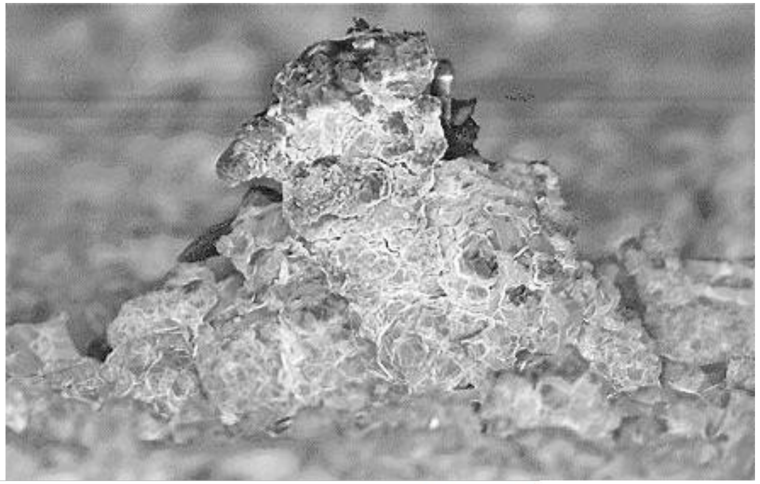
Excellent adhesion

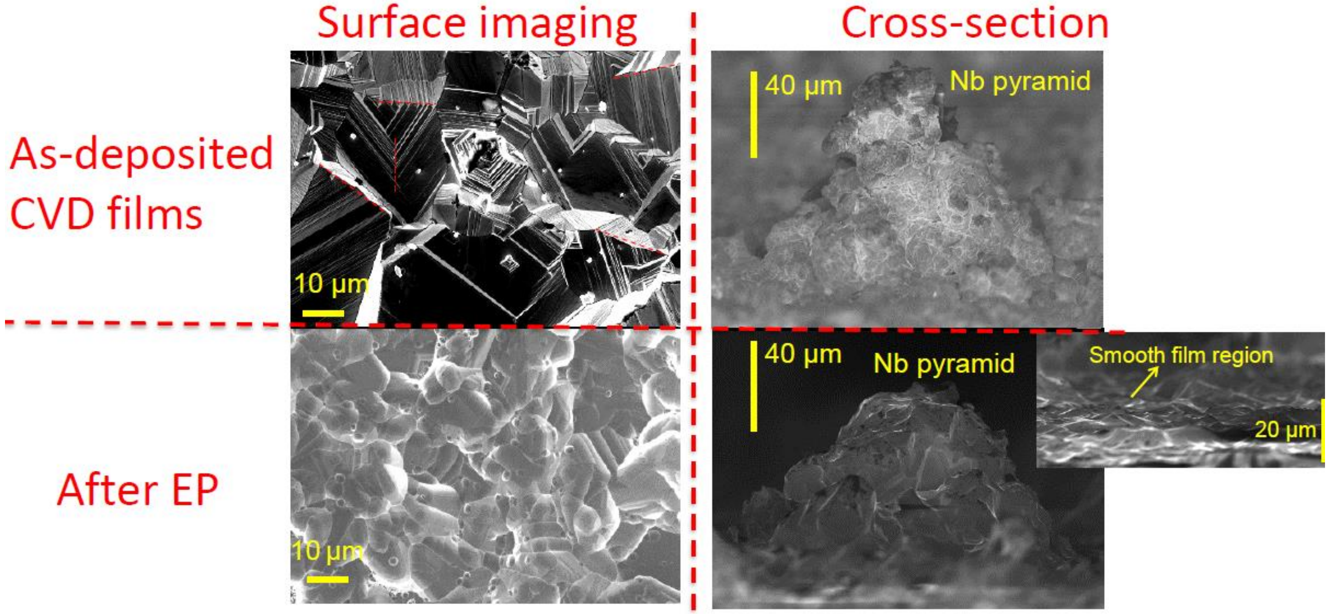
Full size cavity

Zeming Sun Mingqi (Cornell), TTC Meeting, CERN 2020

CVD Thick film @ Cornell University and Ultramet







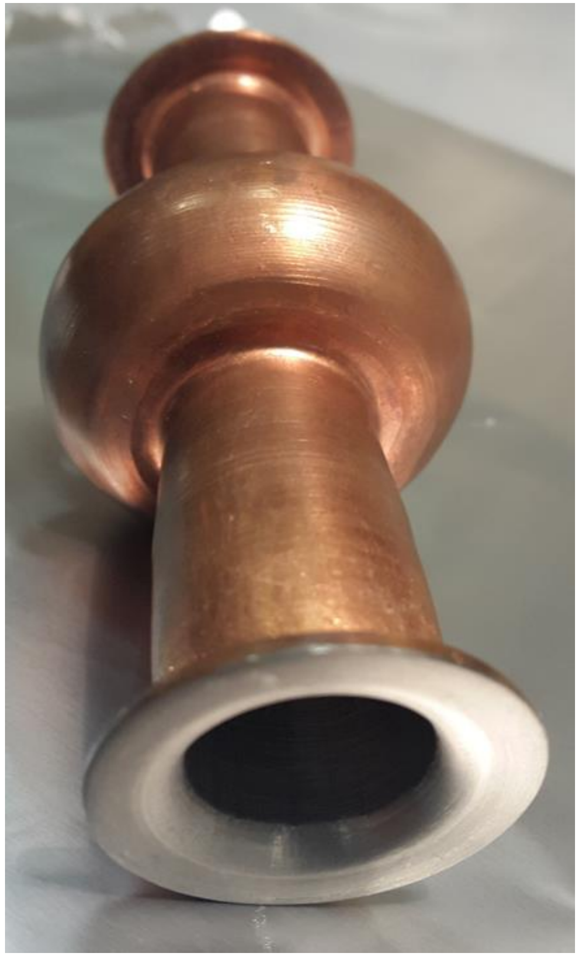
Very Rough Surface

EP smooth pyramids

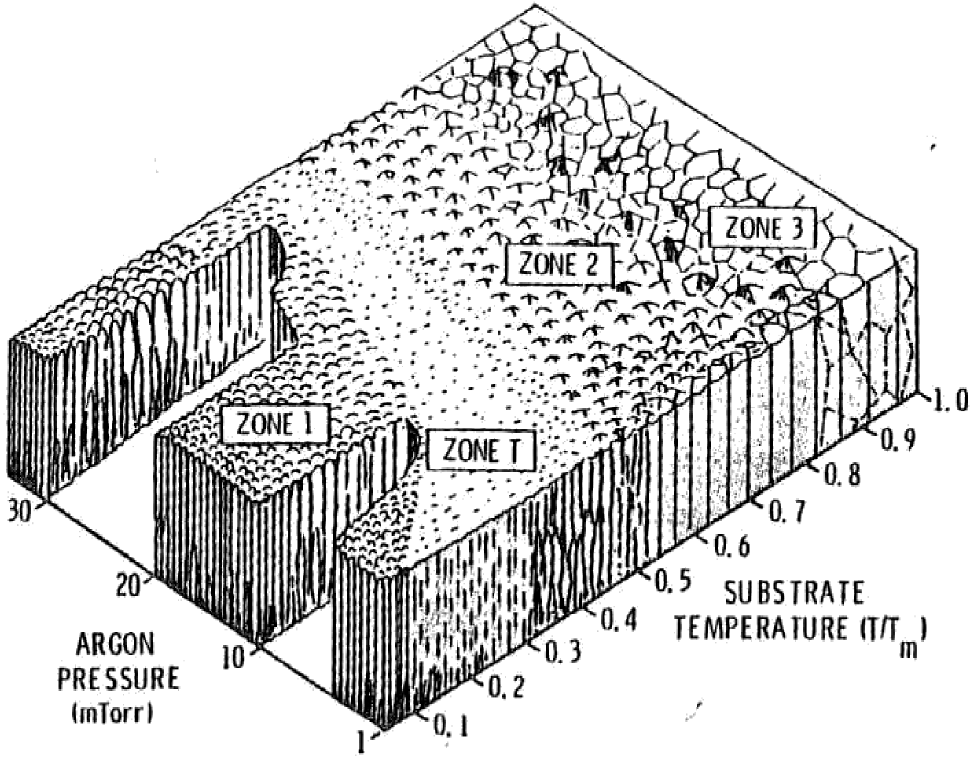
Zeming Sun Mingqi (Cornell), TTC Meeting, CERN 2020

LNL Approach

- Thick films
- High temperature



High Temperature Deposition Motivation



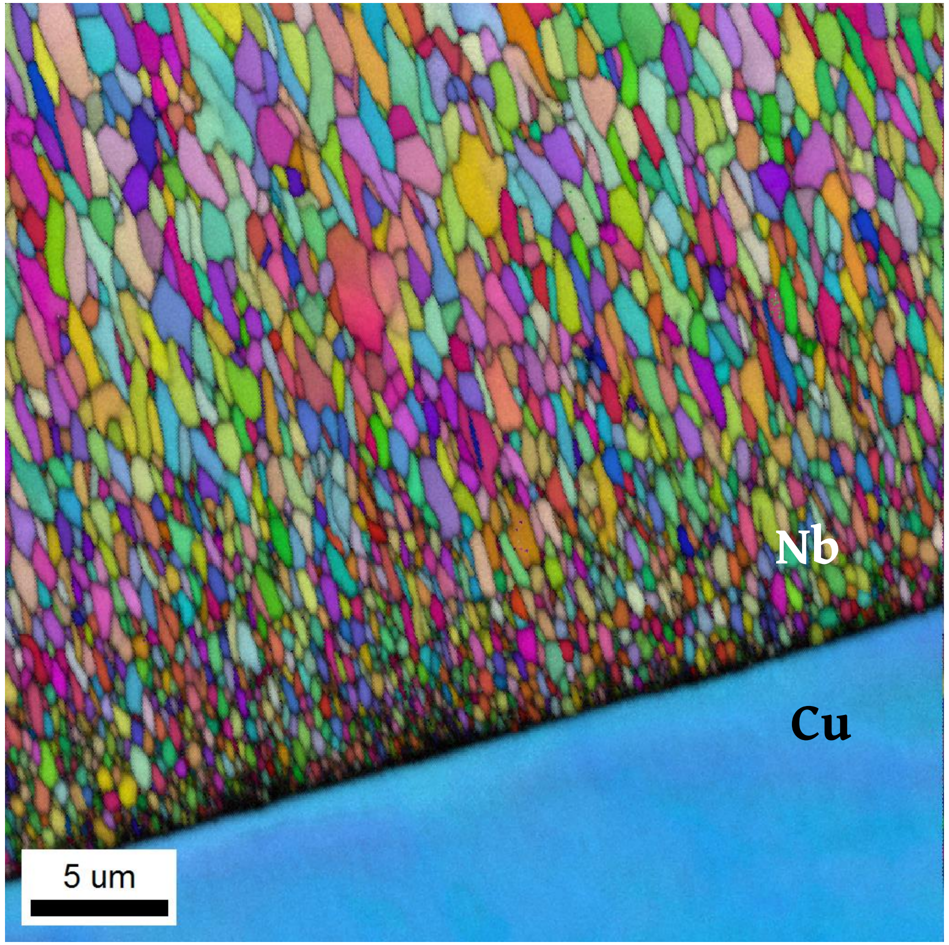
- Thorton SZ Diagram
  J.A. Thornton and D.W. Hoffman, Thin Solid Films, vol. 171, no. 1, pp. 5–31, 1989
- CERN **(550 °C)**
  C. Benvenuti et al., *Physica B 197 (1994) 72-83*
- LNL Alpi Linac **(300-500 °C)**
  Stark et al., Proceedings of SRF1997
- Hie-Isolde **(650 °C)**
  Sublet et al., Proceedings of SRF2013

Thick films increase grain dimensions and RRR

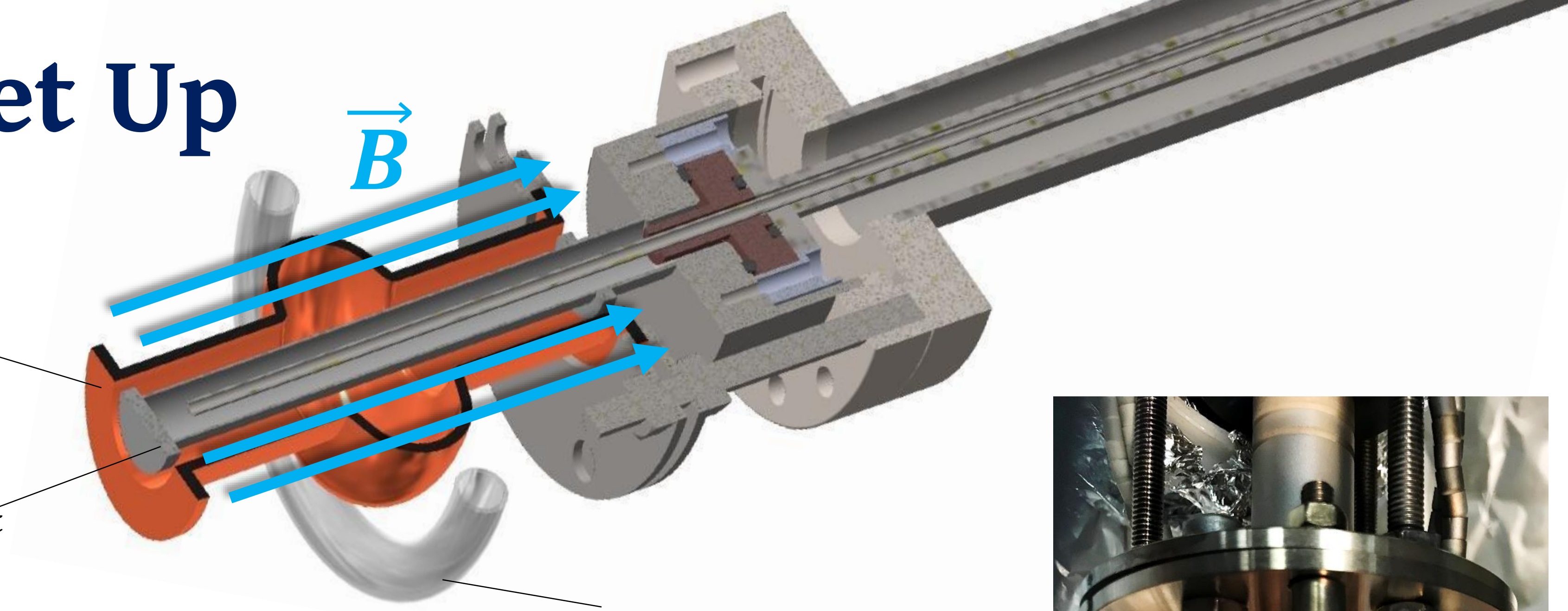
Grain dimension ≈**1-10 µm**

RRR > 60

Bulk like properties

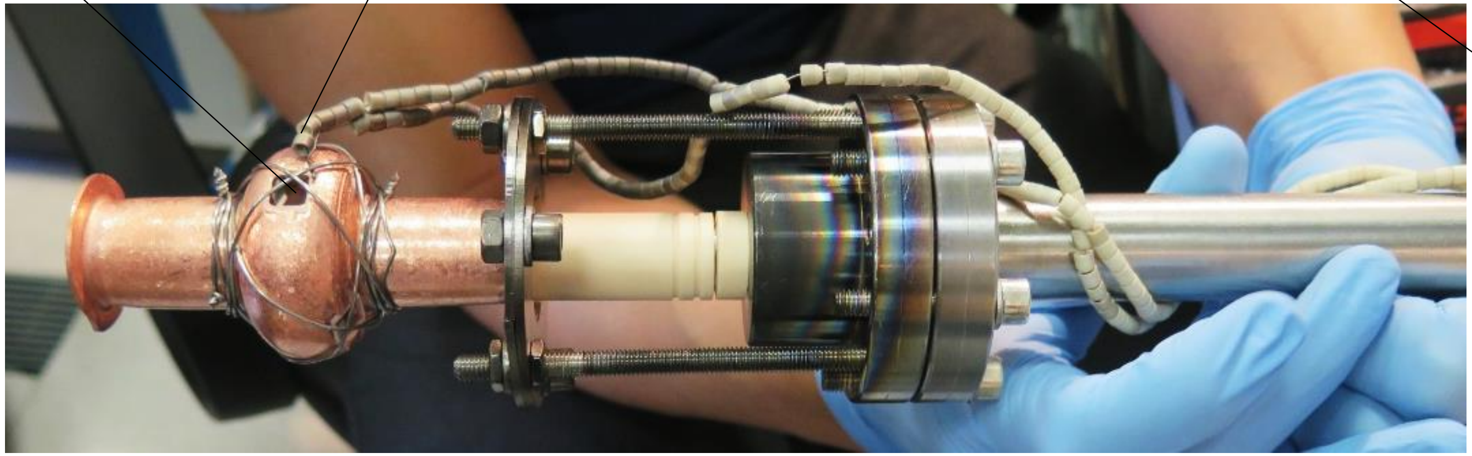


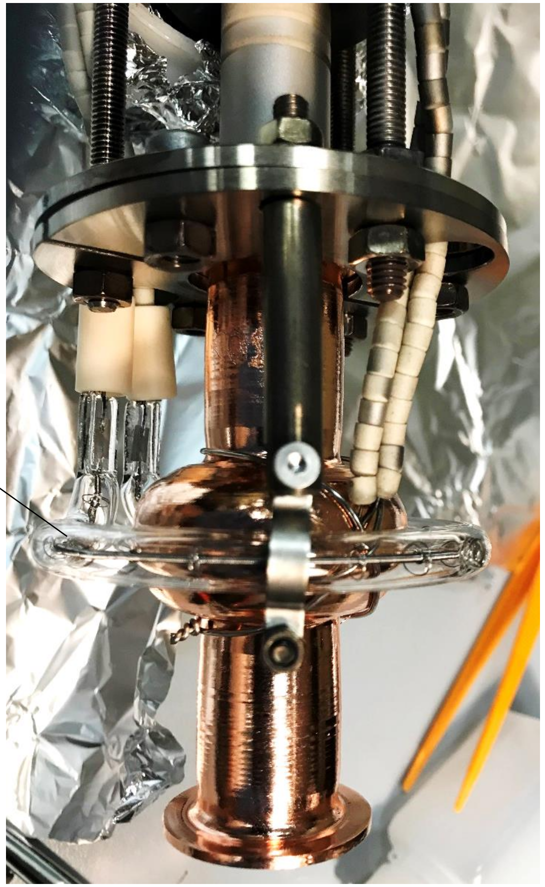
Deposition Set Up



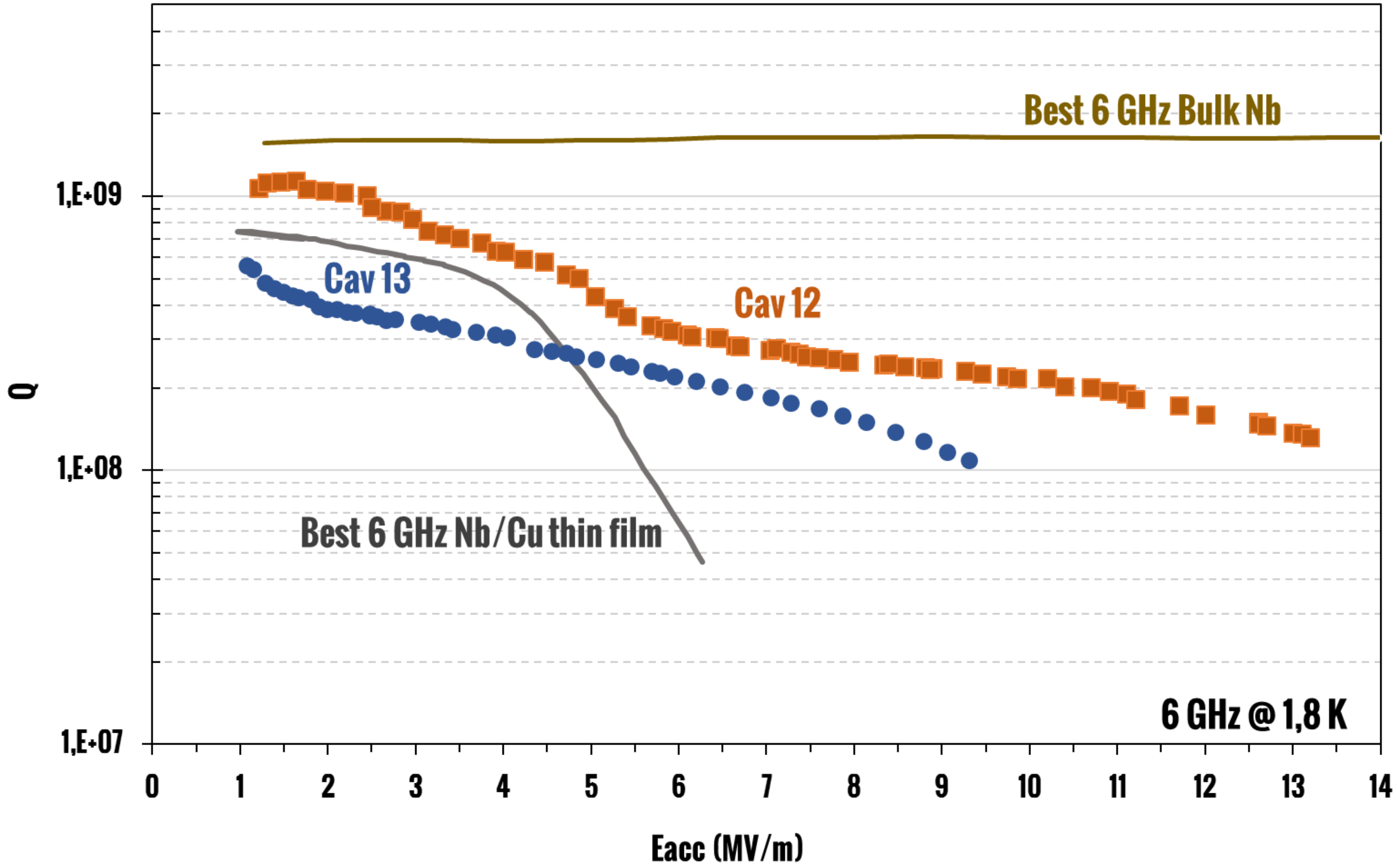
Quartz sample

Thermocouple

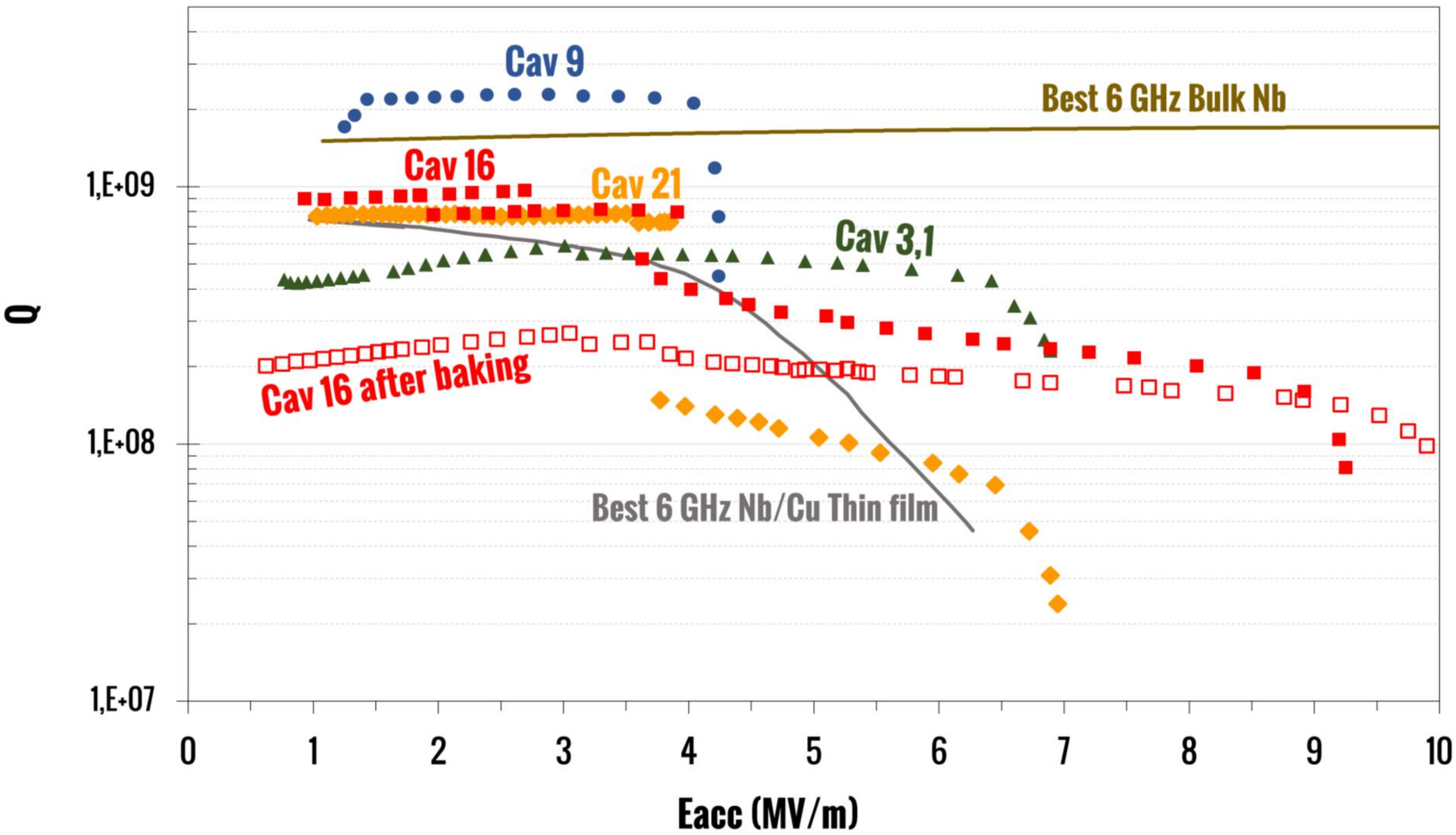




Q-slope remains in many cavities…



...but not all!


Cav 9
Best 6 GHz Bulk Nb
Cav 16
Cav 21
Cav 3,1
Cav 16 after baking
Best 6 GHz Nb/Cu Thin film
Q
1,E+09
1,E+08
1,E+07
0
1
2
3
4
5
6
7
8
9
10
Eacc (MV/m)

Conclusions

Nb thin films are a great choice for low gradient/4.2 K applications

- Cost reduction
- $R_{BCS\ film} < R_{BCS\ bulk}$ → $Q_{0\ film} > Q_{0\ bulk}$
- Thermal stability
- Mechanical stability
- Less sensitivity to magnetic field trapping
- Safer handling for the chemical surface treatments

Mitigation of Q-slope for high gradient applications seems possible

We need to understand the reason of the Q-slope

- Establish adequate process controls
- Mandatory have better substrates and chemical processes
- Need more RF measurements statistics