A-M Valente-Feliciano

SOTTO L'ALTO PATRONATO DEL PRESIDENTE DELLA REPUBBLICA
UNDER THE HIGH PATRONAGE OF THE PRESIDENT OF THE ITALIAN REPUBLIC

Beyond bulk Nb

Acknowledgements

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A. Sutter

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Beyond bulk Nb

Bulk-like Nb film

high κ film
on Cu, Al
substrate

- Minimize Rres, maximize Q
  critical for CW RF
- Potential major system simplifications
- Highest level of quality assurance and reliable performance.
- Use of substrates with higher thermal conductivity (Cu, Al)

Higher-T_c Materials

Thick film
Nb_3Sn
…
Nb, Cu
substrate

SIS structures

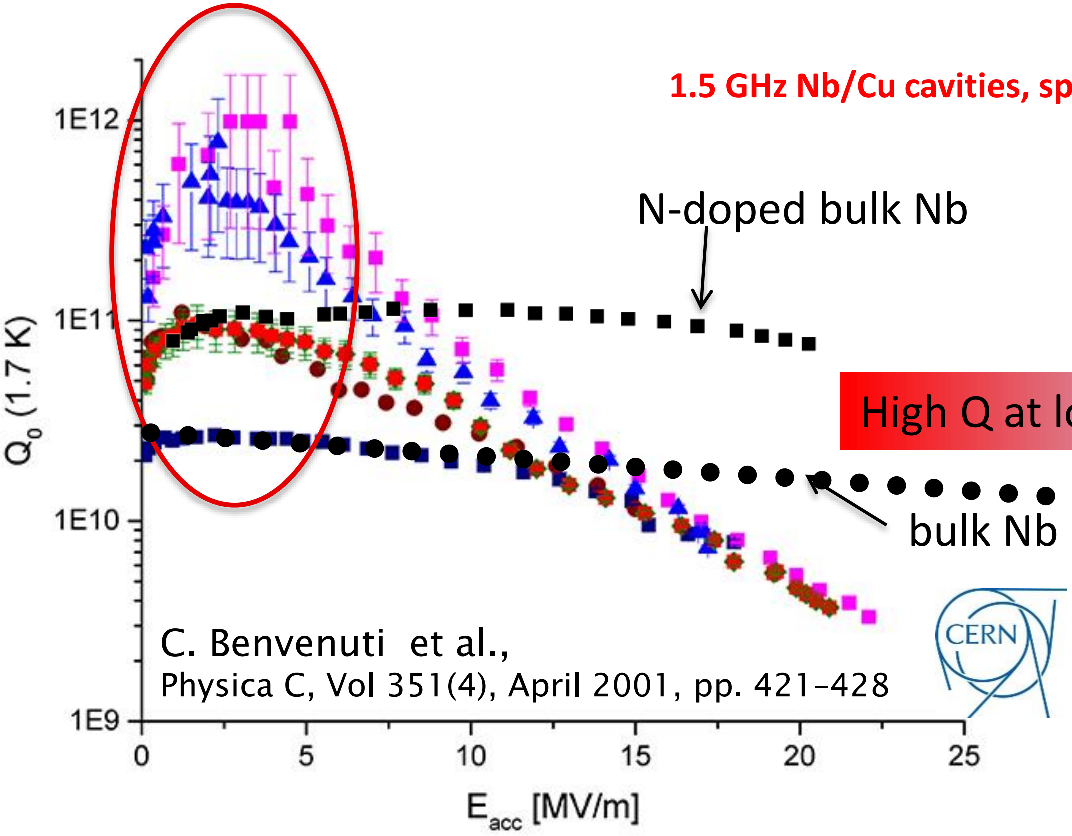
on Nb
(bulk, thick film)
substrate

- function at **higher temperatures** or **higher fields**
- Delay vortex entry in multilayer structures

Accessible almost only via film route: deposition, synthetization, diffusion

Nb Thin Films for SRF - State of the Art

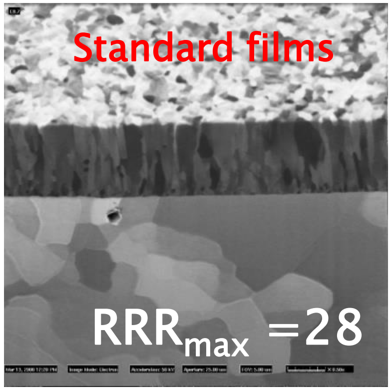
1.5 GHz Nb/Cu cavities, sputtered w/ Kr @ 1.7 K ($Q_0=295/R_s$)



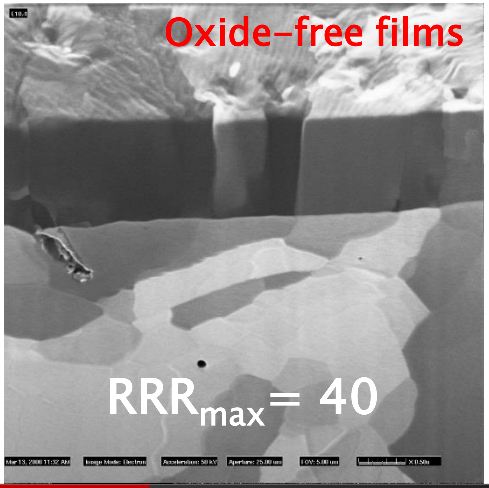
C. Benvenuti et al.,
Physica C, Vol 351(4), April 2001, pp. 421-428

High Q at low field BUT strong Q-slope

Columnar grains,
size ~ 100 nm
In plane diffraction
pattern: powder
diagram
(110) fiber texture ⊥
substrate plane



Equi-axed grains,
size ~ 1-5μm
In plane diffraction
pattern: zone axis
[110]
Heteroepitaxy
Nb (110) //Cu(010)
, Nb (110)
//Cu(111),Nb (100)
//Cu(110)

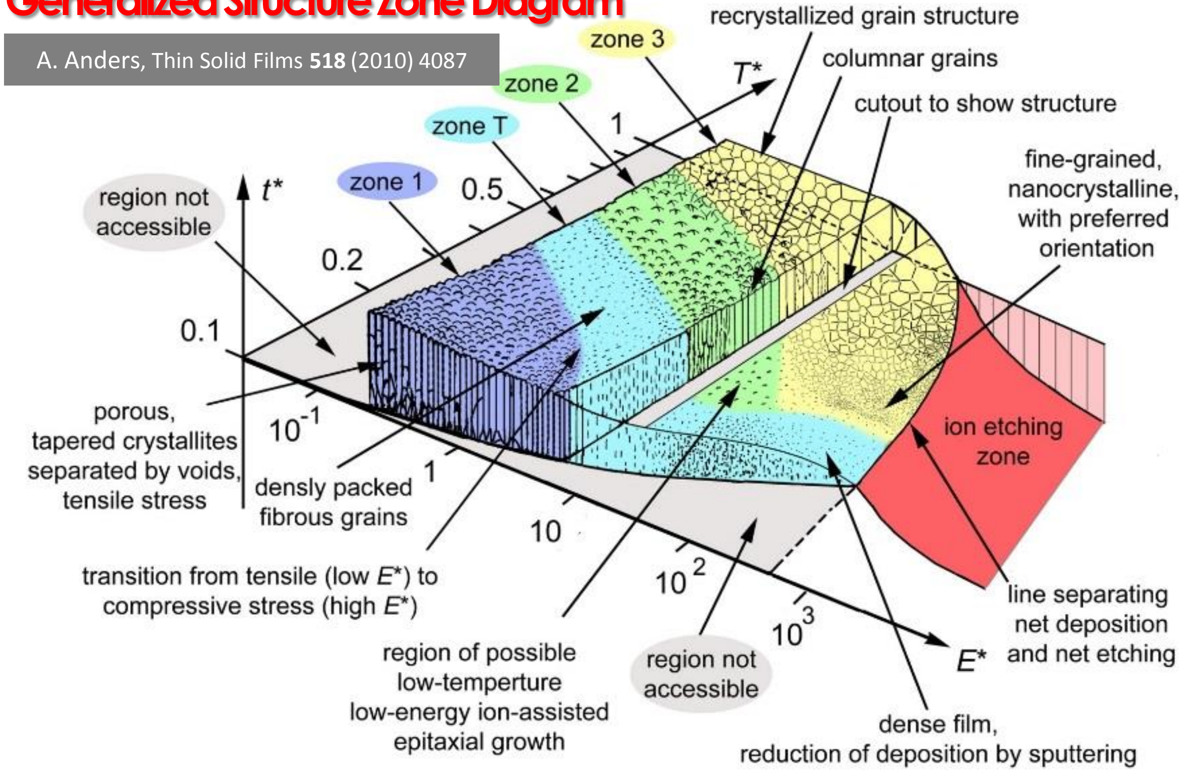


Courtesy: P. Jacob - EMPA

Energetic Condensation

Condensing (film-forming) species : hyper-thermal & low energies (>10 eV).

Generalized Structure Zone Diagram


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

derived from Thornton's diagram for sputtering (1974)

Additional energy provided by fast particles arriving at a surface ⇒number of surface & sub-surface processes ⇒changes in the film growth process:

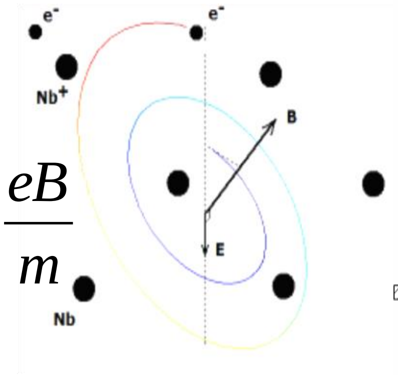
- residual gases desorbed from the substrate surface
- chemical bonds may be broken and defects created thus affecting nucleation processes & film adhesion
- enhanced mobility of surface atoms
- stopping of arriving ions under the surface

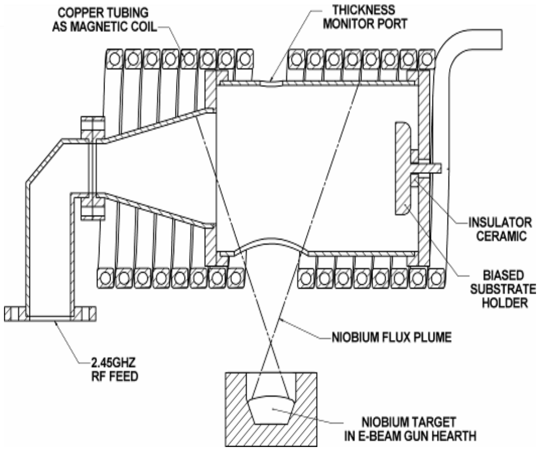
Possibility of controlling the film properties

- Morphology & microstructure
- Stress
- Density of the film
- Film composition
- Crystal orientation may be controlled to give the possibility of low-temperature epitaxy

Energetic condensation with ECR

$$\omega = \frac{eB}{m}$$





No working gas

Ions produced in vacuum

Singly charged ions **64eV**

Controllable deposition energy with Bias voltage

Excellent bonding

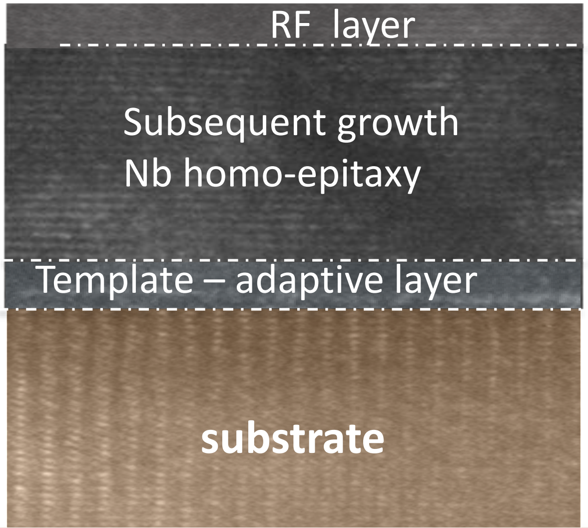
No macro particles

Good conformality

Generation of plasma 3 essential components:

Neutral Nb vapor, RF power (@ 2.45GHz), Static B $\perp$ E_{RF} with ECR condition

Engineering for optimum RF performance

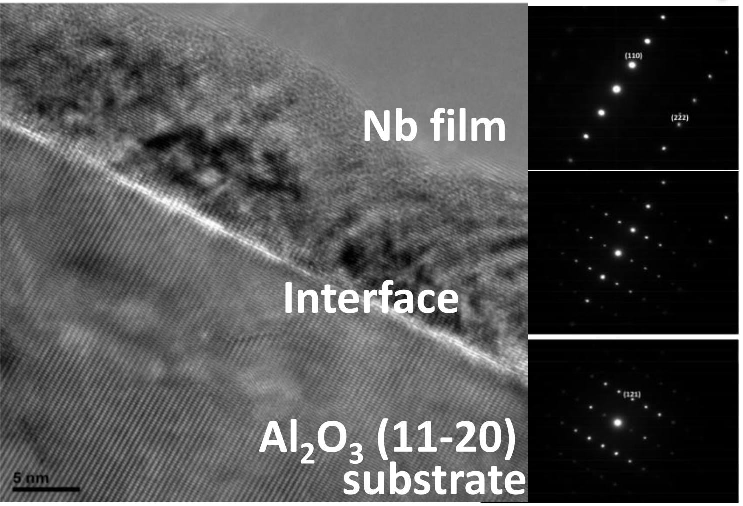


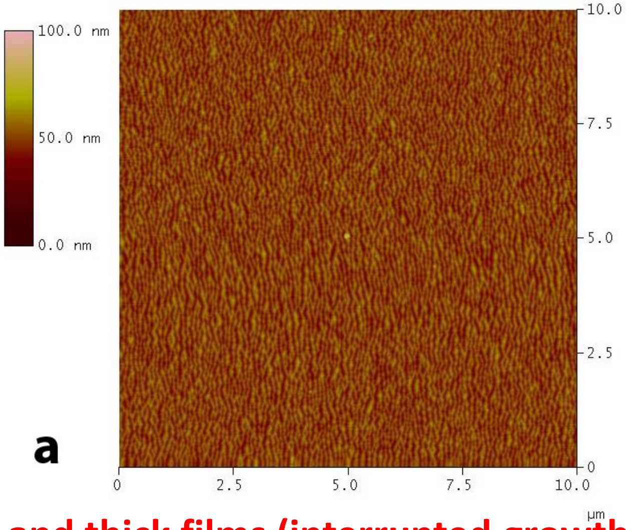
3 sequential phases for film growth

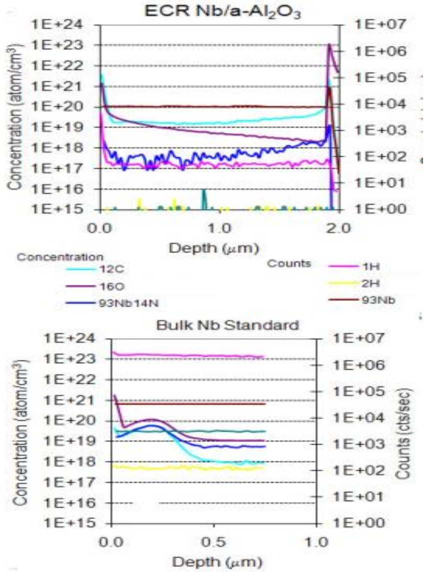
- ☐ Film nucleation on the substrate (Nb, Al_2O_3, Cu; single crystal, polycrystalline, amorphous)
- ☐ Growth of an appropriate template for subsequent deposition
- ☐ Deposition of the final surface optimized for minimum defect density.

ECR Nb films on ideal substrates

Crystalline Al_2O_3 (11-20) substrate → Heteroepitaxy with continuously crystalline interface

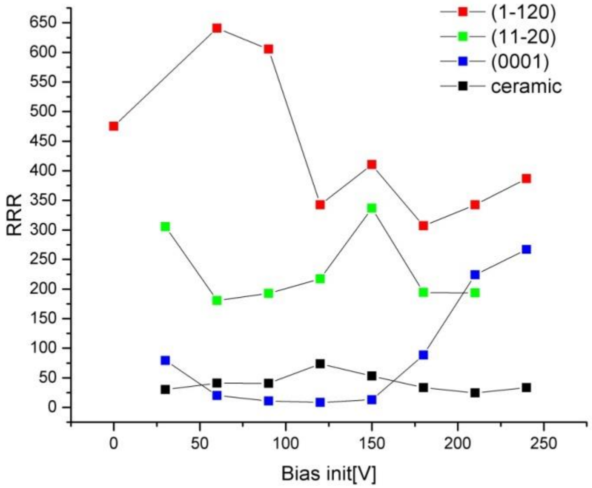




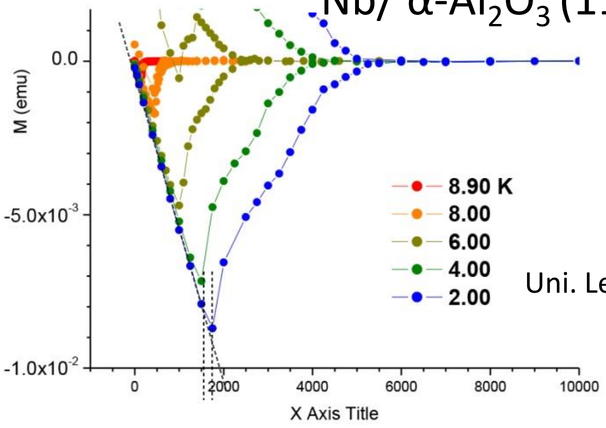


Bake @ 500 °C, coating @ 360 °C

dual energy and thick films (interrupted growth)

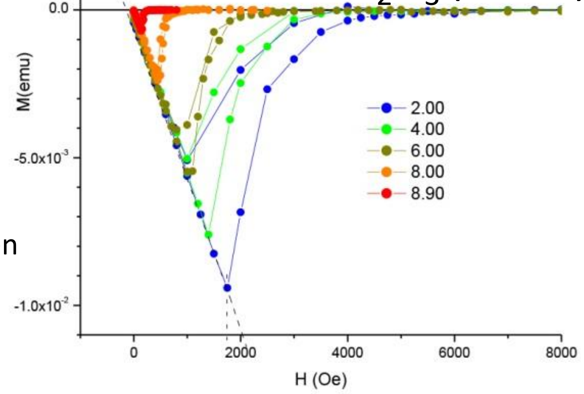


Nb/ α-Al_2O_3 (11-20)



Uni. Leuven

Nb/ α-Al_2O_3 (1-120)



Nb (100)/(1-120) Al_2O_3 always higher RRR and bulk-like T_c than Nb (110) &(111)

RRR = 489

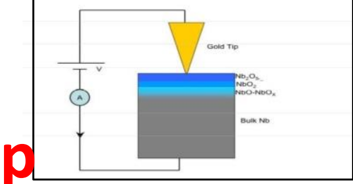
H_{fp} = 155 mT

RRR = 725

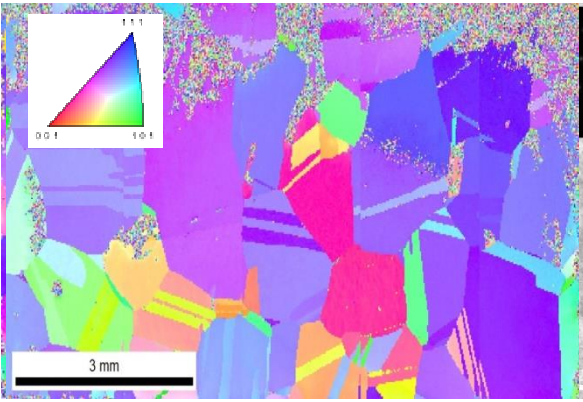
H_{fp} = 175 mT (~ H_{c1}^{Nb})

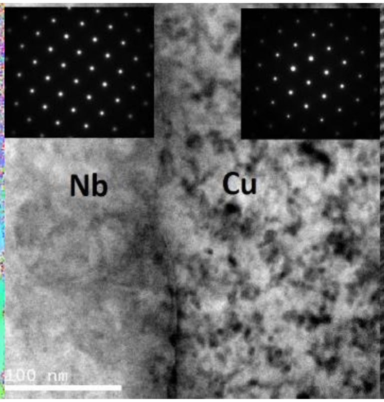
ECR Nb films on Cu substrate

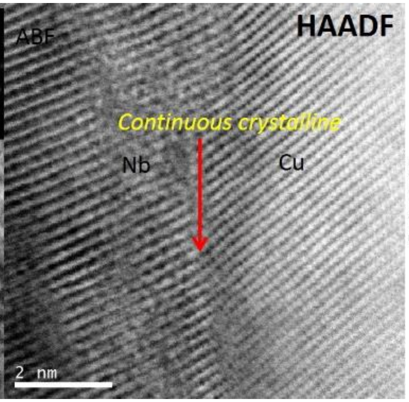
Structure, interface and superconducting gap

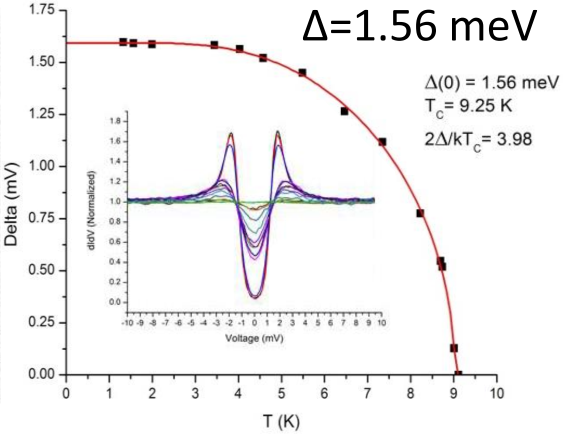


Nb on crystalline Cu substrate (360 °C)   Continuous crystalline interface

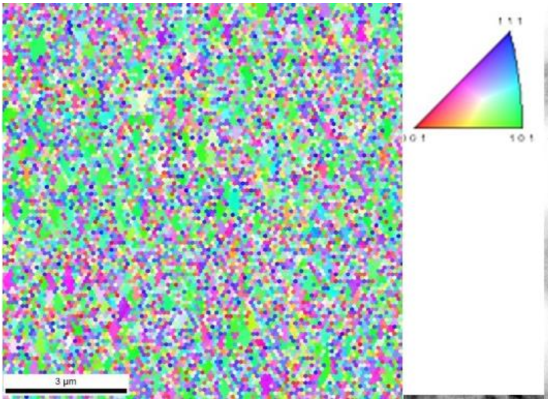


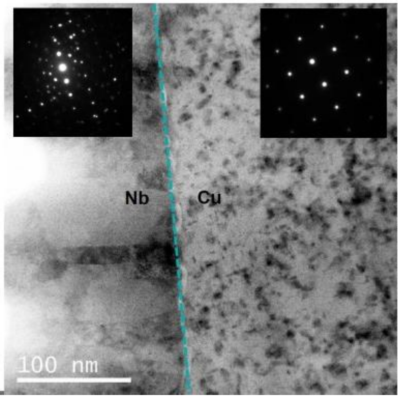


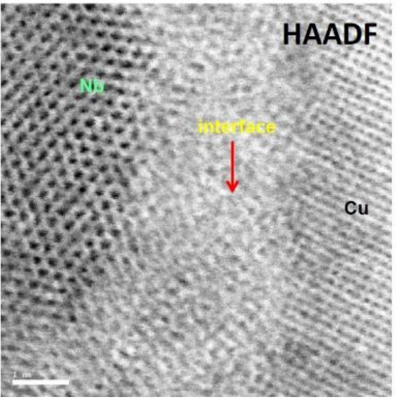


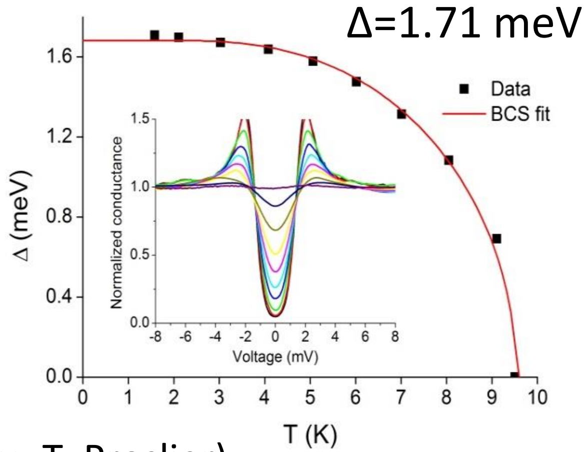

Δ=1.56 meV

Nb on Cu oxide (200 °C) Amorphous interface





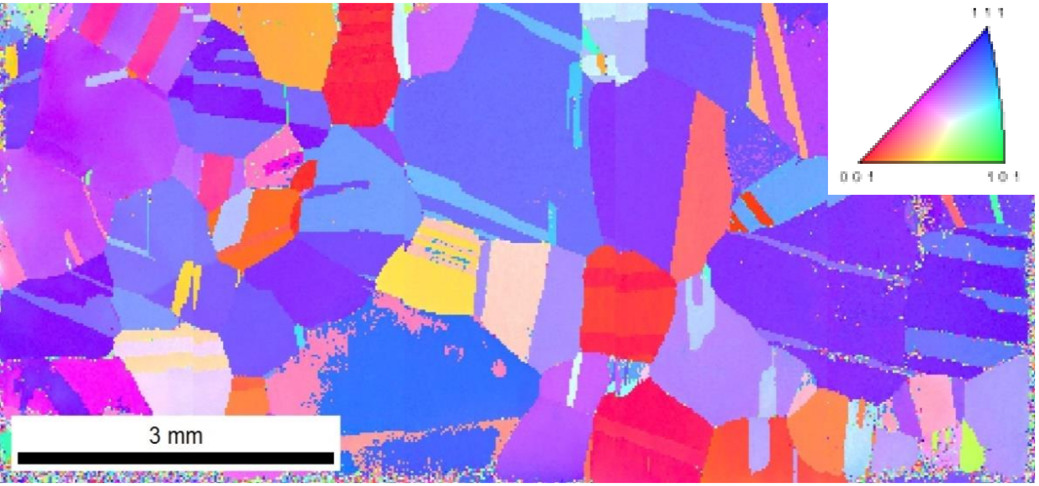



Δ=1.71 meV

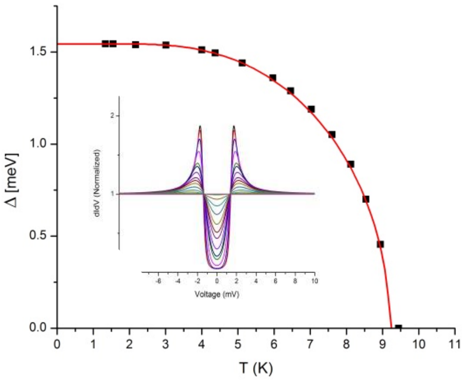
Gap measurements performed by PCT (point contact tunneling spectroscopy- T. Proslier)

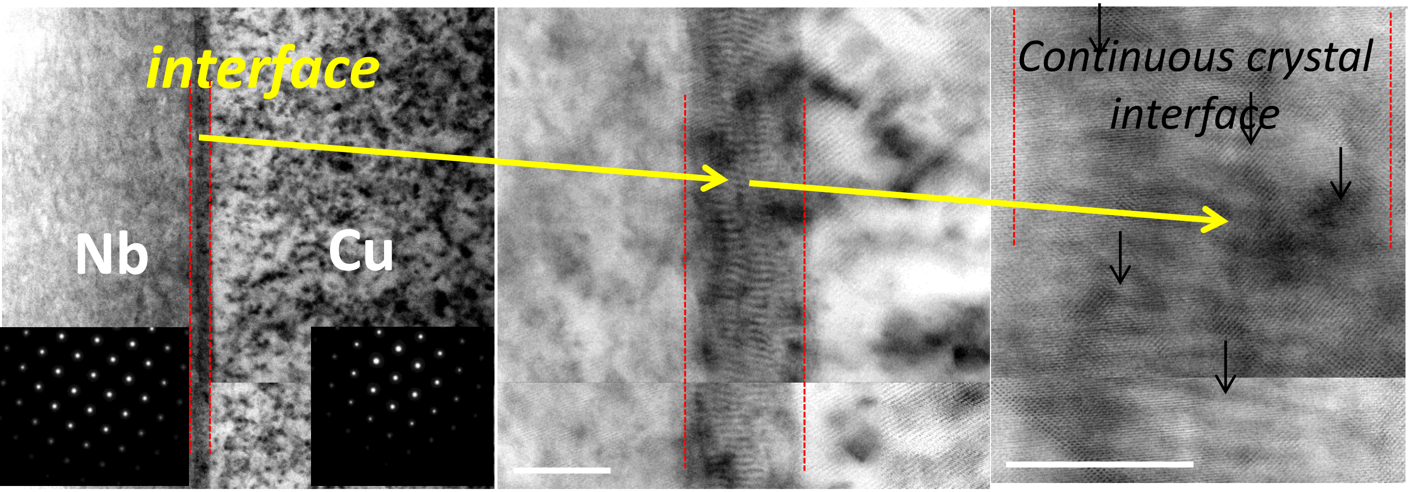
Superconducting gap (1.56-1.62meV) **similar to bulk Nb** ($\Delta_{Nb\ bulk}$ =1.55meV measured on the same setup) for hetero-epitaxial ECR Nb films on polycrystalline Cu.

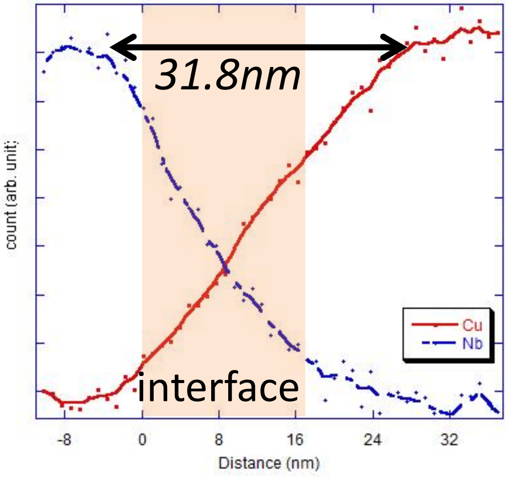
Hetero-epitaxy, High $T_{coating}$



T_{bake} = 500 °C
$T_{coating}$ = 360 °C
$E_{Nb\ ions}$ = 184 eV/64 eV
Thickness = 4.5 µm
RRR = 305
T_c= 9.37 ± 0.12 K
Δ=1.53 meV



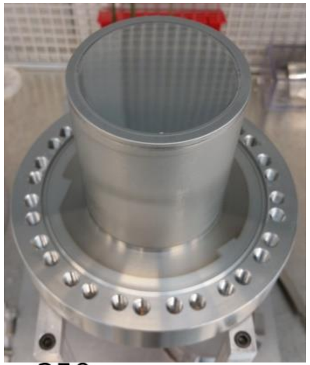


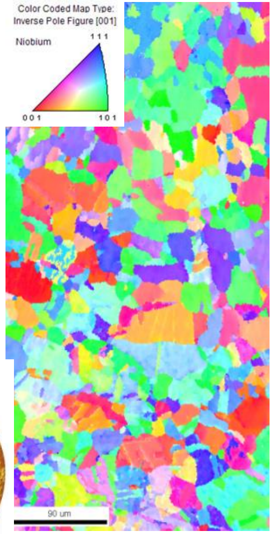


EELS plot for Cu/Nb signal across interface

Interface thickness (e^{-1} of highest density)
Nb: 12.5 nm
Cu: 20.1 nm

ECR Nb/Cu– Surface resistance





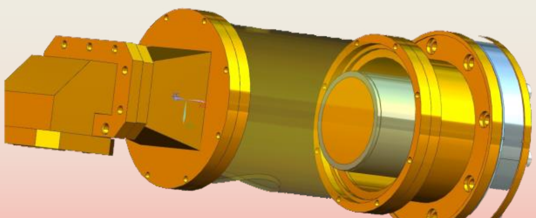
Hetero-epitaxial film Nb on OFHC Cu, 360 °C bake & coating

184 eV for nucleation/early growth + 64 eV for subsequent growth

T_c= 9.36 ± 0.12 K

RRR = 179 (Nb/α-Al_2O_3 (11-20), witness sample)

EBSD IPF map and XRD pole figure show very good crystallinity and grain sizes in the range of the typical Cu substrate



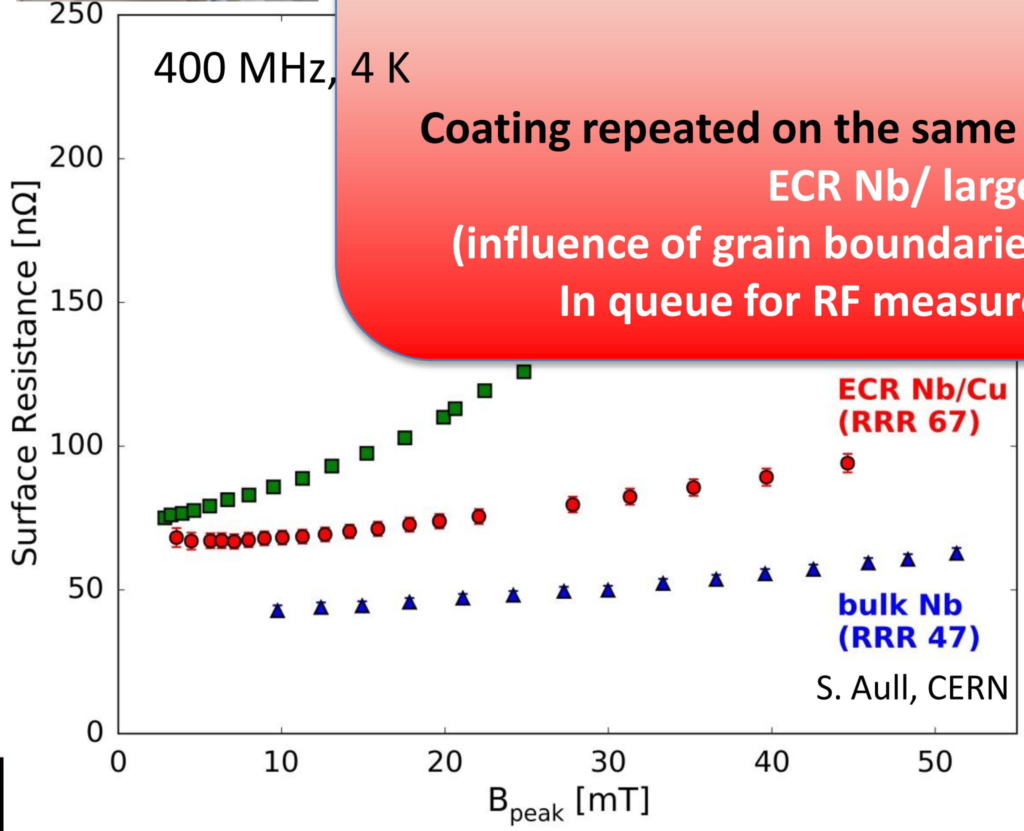
Coating repeated on the same substrate, same conditions

ECR Nb/ large grain Cu

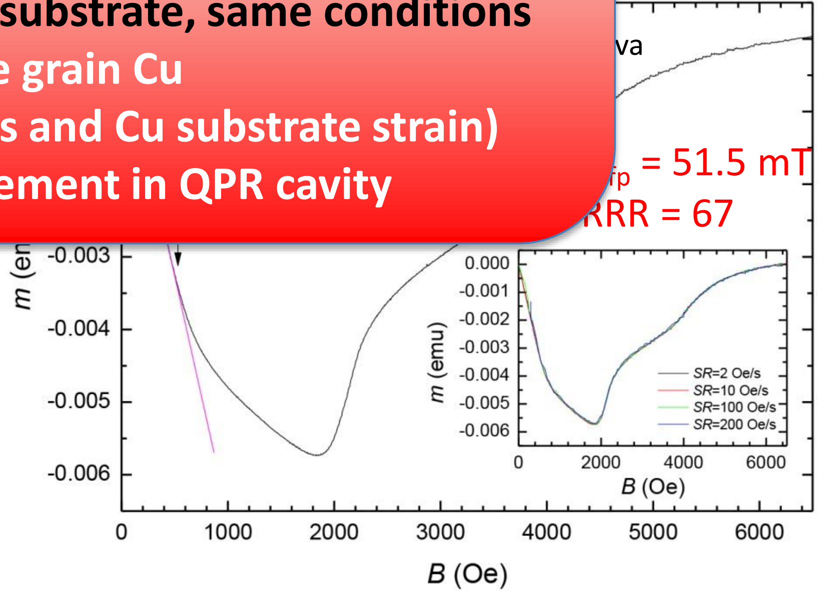
(influence of grain boundaries and Cu substrate strain)

In queue for RF measurement in QPR cavity

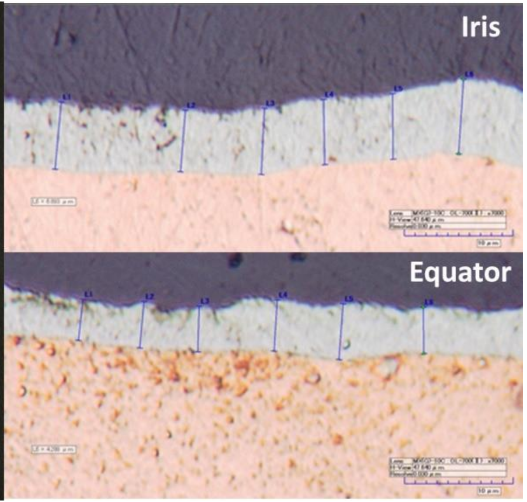
	RRR
	67 ± 7



S. Aull, CERN



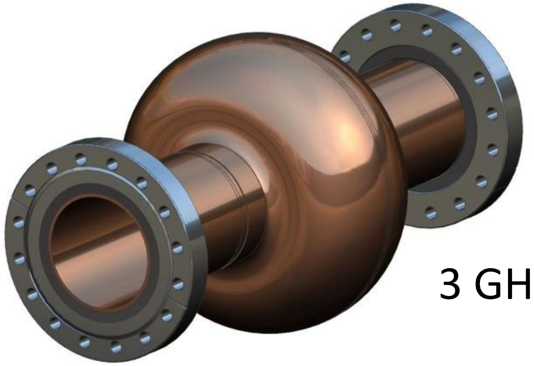
Deposition on cavities



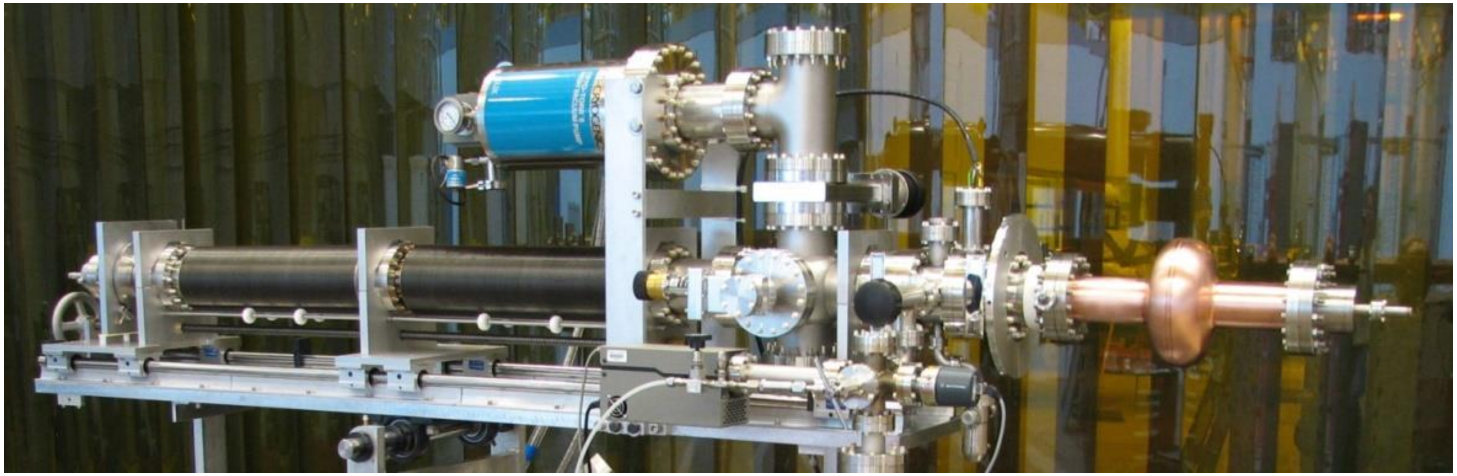
Conformality of the ECR process:

Film thickness along a 3GHz half-cell profile varies from 4μm (equator) to 6μm (iris)

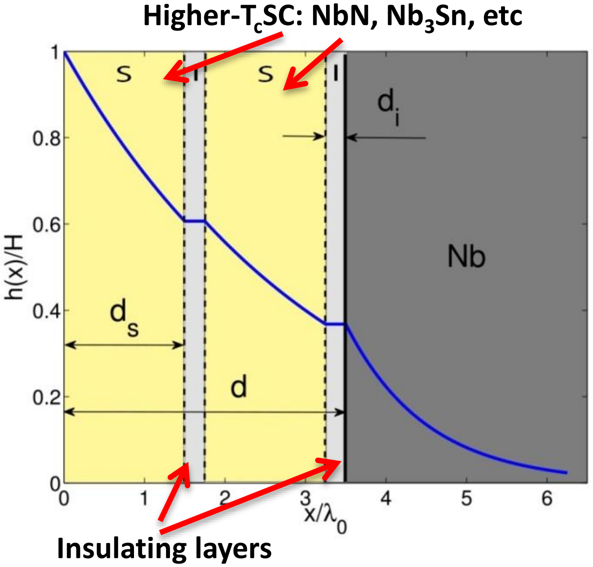
Note: very rough substrate , only grossly mechanically polished



HiPIMS cylindrical coating system for single cell commissioned



Beyond Nb: SIS Multilayers

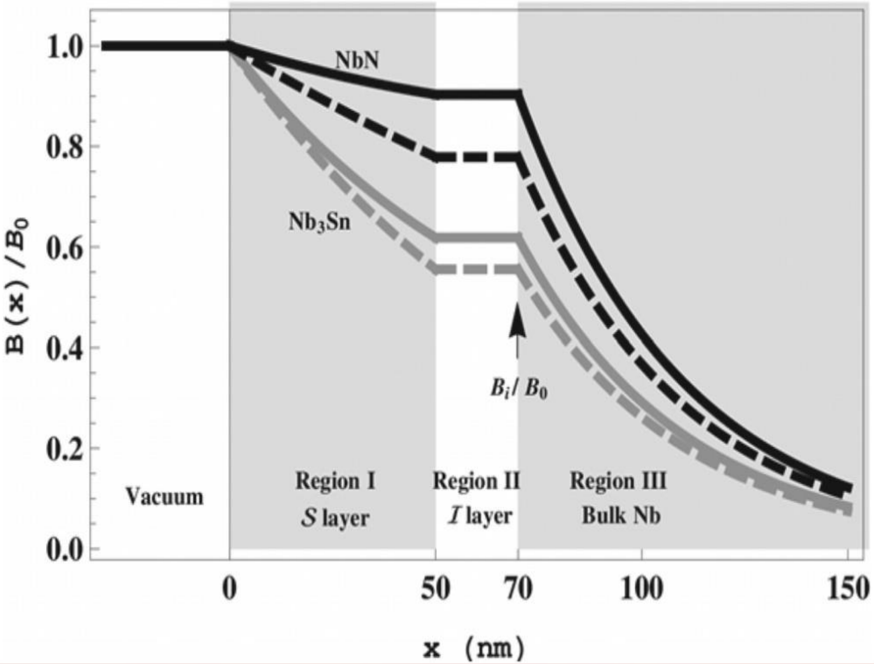


Taking advantage of the high $-T_c$ superconductors with much higher H_c without being penalized by their lower H_{c1}…

Alex Gurevich, Appl. Phys. Lett. 88, 012511 (2006)
Alex Gurevich, AIP ADVANCES 5, 017112 (2015)
T. Kubo, Applied Physics Letters 104, 032603 (2014)

Multilayer coating of SC cavities:
alternating SC and insulating layers with $d < \lambda$

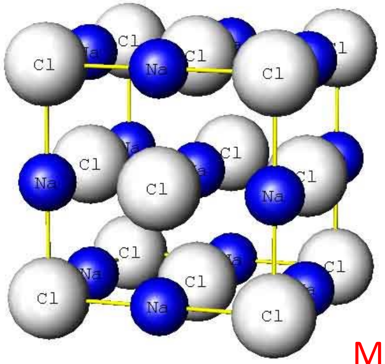
Higher T_c thin layers provide magnetic screening of the Nb SC cavity (bulk or thick film) without vortex penetration



- Strong increase of H_{fp} in films allows using RF fields > H_c of Nb, but lower than those at which flux penetration in grain boundaries may become a problem=> no transition, no vortex in the layer
- High H_{fp} ,applied field is damped by each layer
- Insulating layer prevents Josephson coupling between layers
- Applied field, i.e. accelerating field can be increased without high field dissipation
- SC layers with higher T_c, Δ (Nb_3Sn, NbN, etc.) => Strong reduction of R_{BCS} (ie high Q_0)

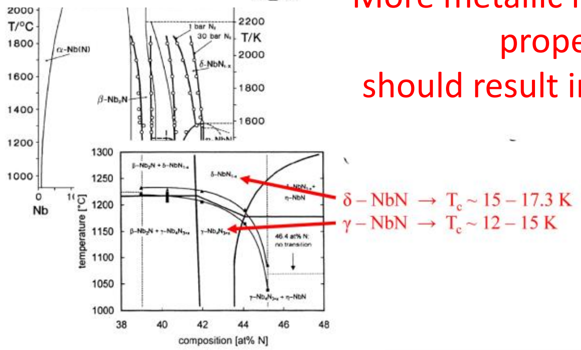
Possibility to move operation from 2K to 4.2K

APPROACH



Superconductor

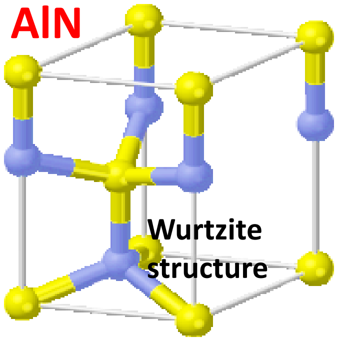
Ternary Nitride **$(Nb_{1-x},Ti_x)N$**
T_c= 17.3 K, a= 4.341 Å for δ-phase
Presence of Ti found to reduce significantly the resistivity

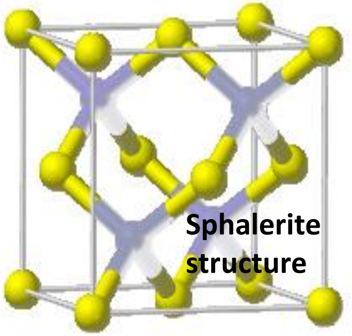


$\delta - NbN \rightarrow T_c \sim 15 - 17.3$ K
$\gamma - NbN \rightarrow T_c \sim 12 - 15$ K

More metallic nature and better surface properties than NbN
should result in better RF performance

Insulator: AlN


Wurtzite structure


Sphalerite structure

- ☐ Grown with a wurtzite (hcp, a=3.11Å, c=4.98Å) or sphalerite (B1 cubic, a= 4.08 Å) structure.
- ☐ Found to enhance the properties (T_c) for very thin NbN and NbTiN films .
- ☐ Large thermal conductivity (3.19W/cm.K at 300K, comparable with Cu, 4.01W/cm.K)

CHALLENGES

- ☐ Develop good quality and uniform thin layers
- ☐ Sharp interfaces
- ☐ Growth of equally performing S/I and I/S layers

Base pressure range: 10^{-10} Torr

- ☐ dc-Magnetron Sputtering (reactive mode)
- ☐ HiPIMS (Huettinger 2000 V, 3000 A)

Substrates:
MgO (ideal)
AlN ceramic (worst case)
Bulk Nb (real)
ECR Nb films (real)

- ☐ **Good quality standalone NbTiN deposited by reactive DC magnetron sputtering**.

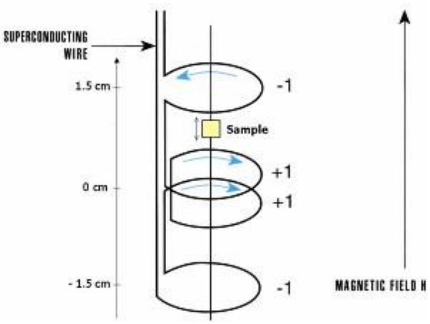
Bulk (2µm) NbTiN films with a T_c =17.3 K and H_{c1} =30 mT.
Cubic δ-phase and T_c > 16 K for thicknesses > 30-50 nm and coating temperatures of 450 ˚C or higher.

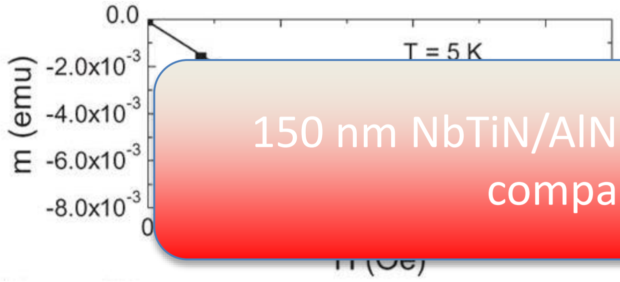
- ☐ **AlN dielectric films with good dielectric properties - n in the range of 1.98- 2.15.**

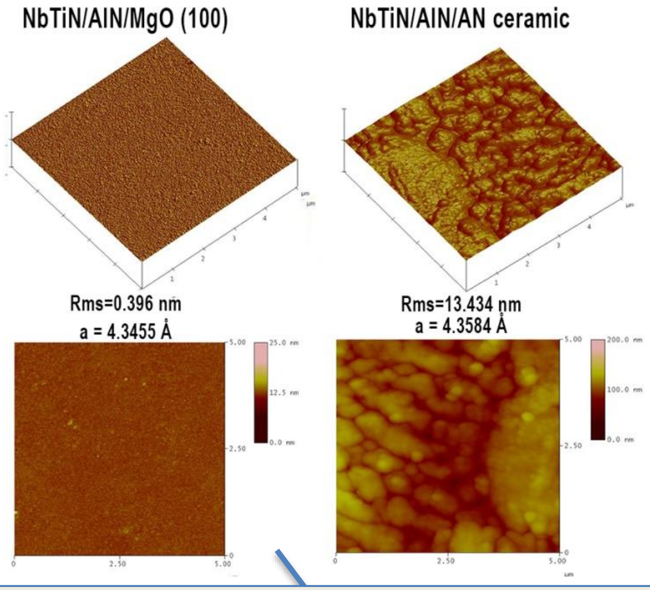
NbTiN/AlN Films (SI) – Flux penetration

SQUID Magnetometry

(Prof. A. Lukaszew group, College William & Mary)

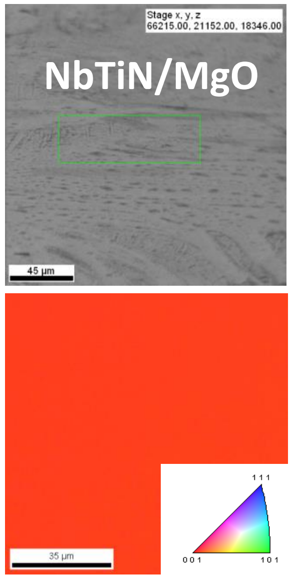




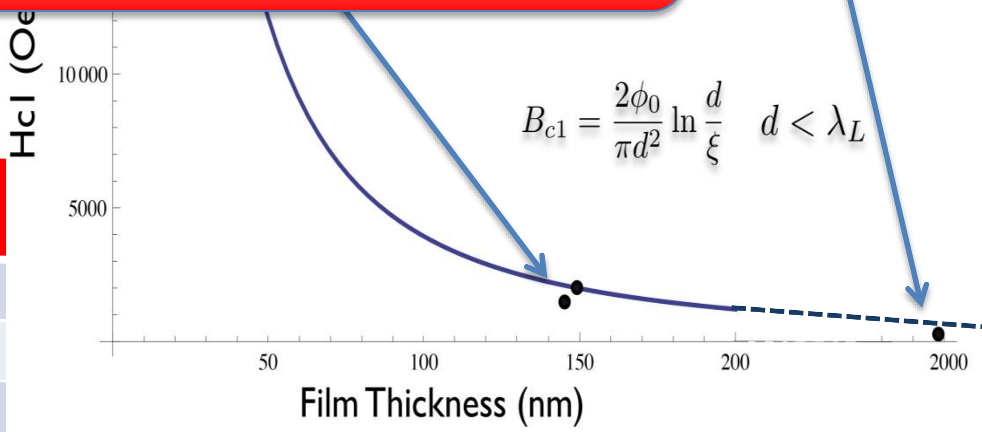


Rms=0.396 nm
a = 4.3455 Å

Rms=13.434 nm
a = 4.3584 Å



150 nm NbTiN/AlN SI structures exhibit **H_{fp} enhancement** compared to bulk-like NbTiN film



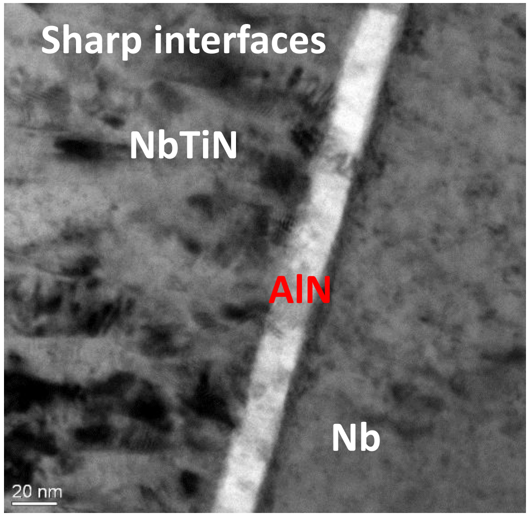
$$B_{c1} = \frac{2\phi_0}{\pi d^2}\ln\frac{d}{\xi} \qquad d < \lambda_L$$

	Thickness [nm]	H_{c1} [mT]	T_c [K]
NbTiN/MgO	2000	30	17.3
NbTiN/AlN/AlN ceramic	145	135	14.8
NbTiN/AlN/MgO	148	**200**	16.7

RF characterization of NbTiN/AlN/Nb structures

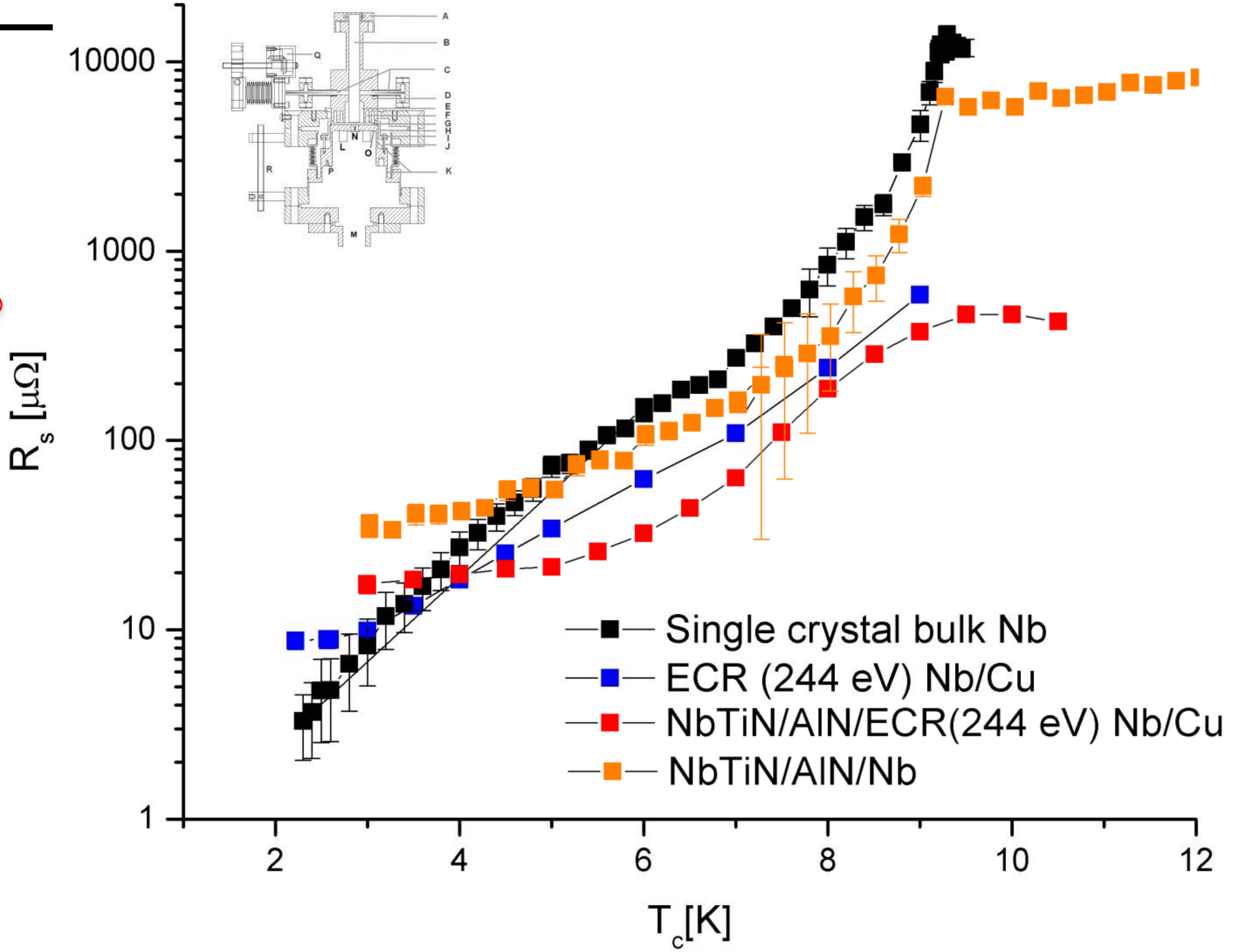
SIS structures coated on ECR Nb/Cu film: 24h-bake, coating and annealing for 4 h at 450°C.

	AlN	NbTiN
N_2/Ar	**0.33**	**0.23**
Total pressure [Torr]	**2×10^{-3}**	**2×10^{-3}**
Sputtering Power [W]	**100**	**300**
Deposition rate [nm/min]	**~ 2.5**	**~ 18**
Thickness [nm]	**20**	**150**
Tc [K]	**N/A**	**16.9**



TEM cross-section (FIB cut) of NbTiN/AlN/Nb/Cu structure

RF Measurement in 7.5 GHz sapphire-loaded TE_{011} cavity

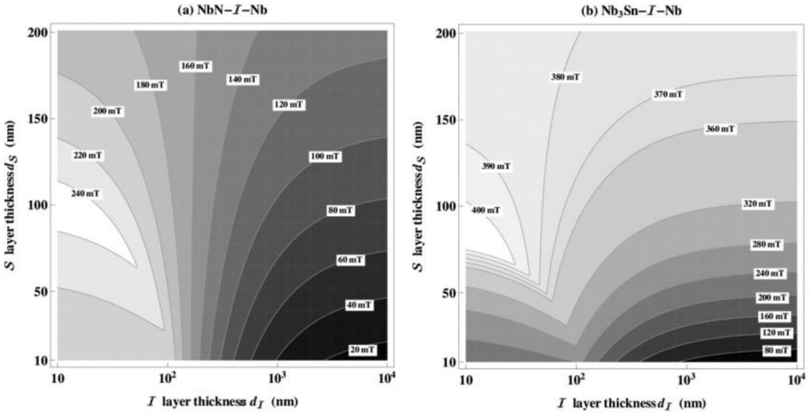


Lower BCS resistance beyond 4 K for SIS coated surfaces compared to standalone ECR film & bulk SC Nb.

NbTiN based SIS Optimization

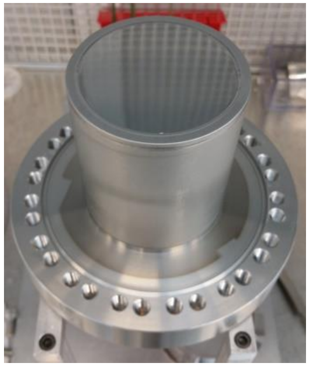
- ☐ Thickness series to **determine/verify optimum layer thicknesses** with H_{fp} measurements
- ☐ **Implementing energetic condensation via HiPIMS (High power impulse magnetron sputtering) to lower the coating temperature while maintaining a good quality δ-phase for NbTiN.**

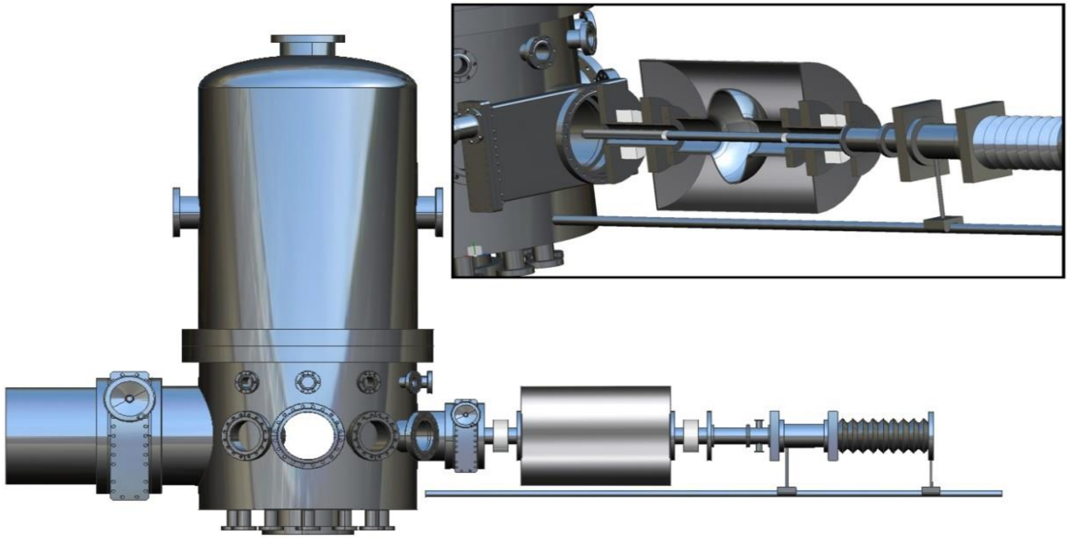
HiPIMS NbTiN films with reasonable results (T_c ~ 16.5-16.9 K).



T. Kubo, SRF 2015

- ☐ **RF measurement** for SIS NbTiN/AlN structures on previously characterized bulk Nb **QPR samples**.



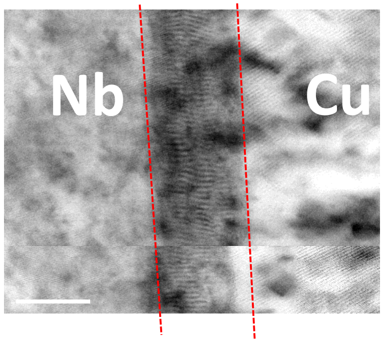


SIS structure coating on Nb & Nb/Cu cavities concept

Conclusions

☐ **Nb films deposited by energetic condensation** (ECR)

- ✓ Know how to coat high quality Nb films and tune properties: crystallinity, impurity content, RRR, superconducting gap
- ✓ RF characterization (QPR) of Nb/Cu surfaces deposited with various energetic condensation techniques – under way
- ✓ Tailor the interface to optimize structure & manage thermal impedance @ interface for maximum SRF performance (ion stitching, interlayer…)
- ✓ 3rd phase coating to study effect of top SRF surface doping/alloying
- ✓ Coating on 3 GHz and 1.5 GHz cavities



☐ **NbTiN based SIS structures**

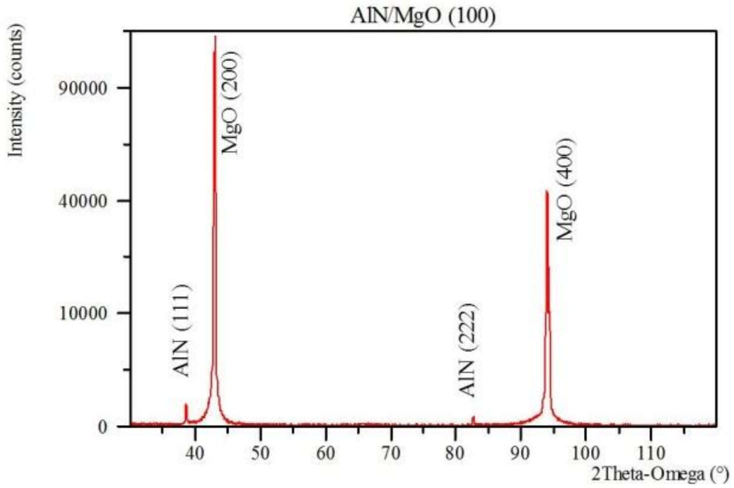
- ✓ Good quality standalone NbTiN & AlN layers
- ✓ SIS NbTiN/AlN layers with a $T_{c,\ NbTiN}$ between 16.6 and 16.9 K.
    - ○ **Growth conditions for SIS structures** need to be a compromise between optimum conditions for standalone films and minimizing interaction between layers .
- ✓ **H_{fp} enhancement** (SQUID magnetometry) observed for 150 nm NbTiN films. Further studies under way to determine /verify optimum layer thickness.
- ✓ **RF characterization of NbTiN/AlN structures coated on Nb surfaces reveal a promise of delaying flux penetration and lower RF losses for SIS coated Nb surfaces, both bulk and thick film**.

Tailored Nb films via energetic condensation

- Tune thin film structure and quality with ion energy and substrate temperature on a variety of substrates (amorphous, polycrystalline and single crystal)
- Achieve film structures and properties only achievable at higher temperature with classic coating methods
- Tune RRR values from single digits to bulk Nb values →No intrinsic limitations
- Lower impurity (H) content than bulk Nb
- Good adhesion to the substrate (delamination threshold determined as function of ion energy and temperature)
- Grain boundaries not necessarily detrimental (if dense) to R_s
- Tailoring interface with high energy and subsequent growth at energy minimizing defect creation can contribute to lower R_s

		Substrate	RRR_{max}
Insulating	Single crystal	a-Al_2O_3	488
		r-Al_2O_3	725
		c-Al_2O_3	247
		MgO (100)	188
		MgO (110)	424
		MgO (111)	270
	Polycrystalline amorphous	Al_2O_3 ceramic	135
		AlN ceramic	110
		Fused Silica	84
Metallic	Single crystal	Cu (100)	181
		Cu (110)	275
		Cu (111)	245
	Polycrystalline	Cu fine grains	193
		Cu large grains	305

AlN Films



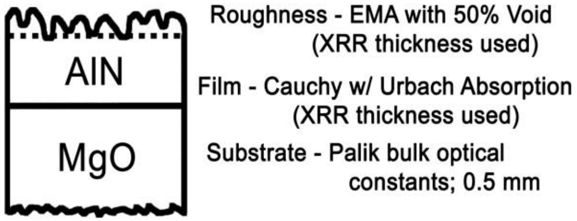
Structure

AlN films were coated by reactive sputtering with different parameters. They were found to become fully transparent for N_2/Ar ratios of ~33%.

Good quality AlN are readily produced at 600 and 450°C by dc-reactive magnetron sputtering.

The films exhibit the cubic structure (single crystal) at 600 °C and the hexagonal structure (polycrystalline) at 450 °C .

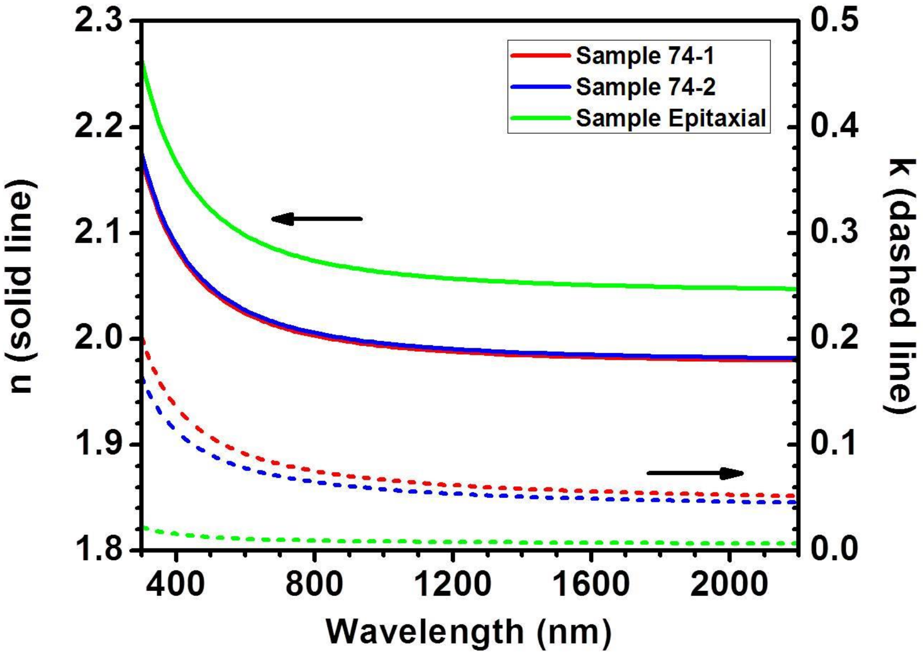
Dielectric Behavior



Roughness - EMA with 50% Void (XRR thickness used)

Film - Cauchy w/ Urbach Absorption (XRR thickness used)

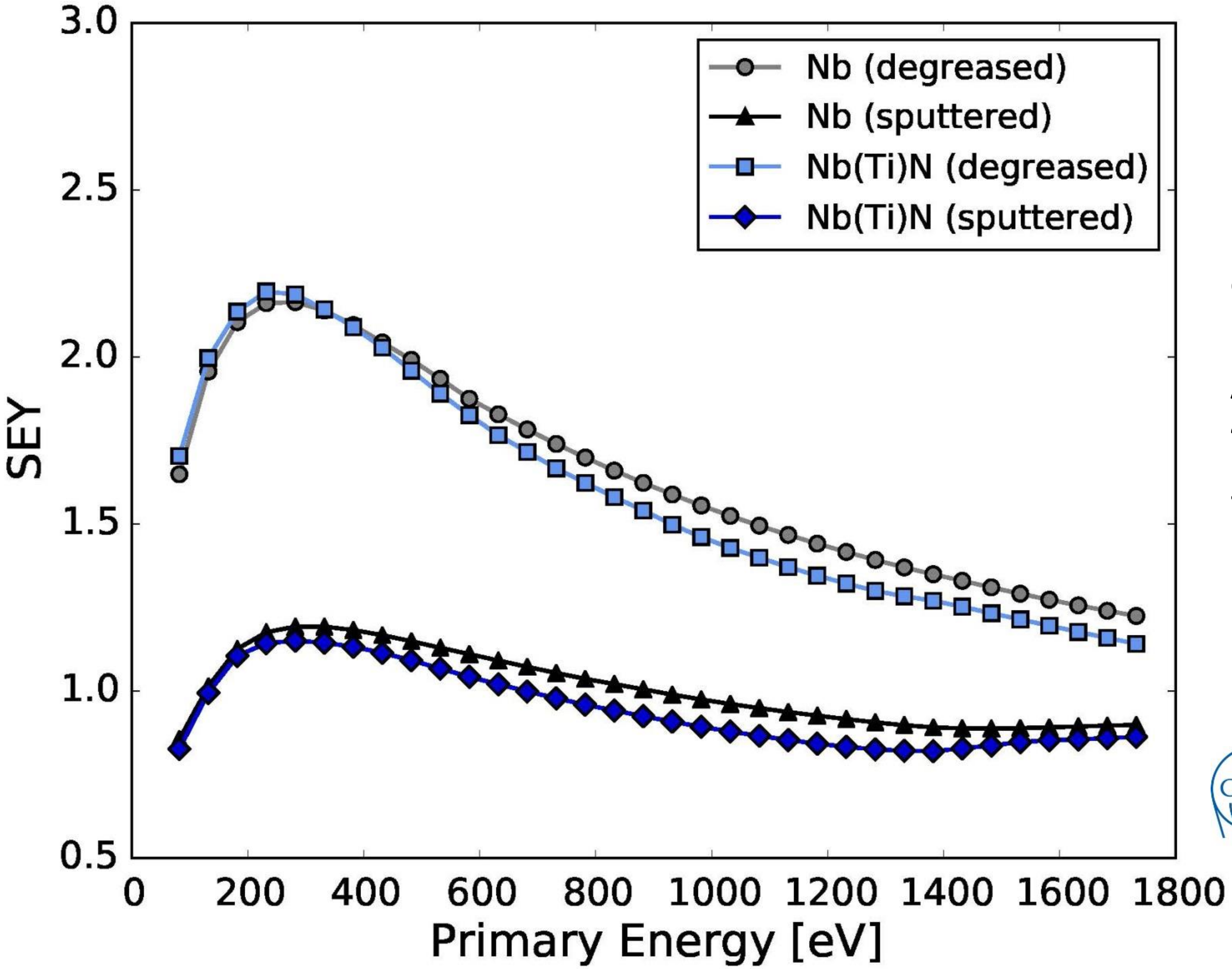
Substrate - Palik bulk optical constants; 0.5 mm



At 450 °C, 30 nm AlN films exhibit dielectric properties of polycrystalline AlN films

n in the range of 1.98- 2.15

Secondary Electron Yield of NbTiN Films



Measurements at
room temperature

Max. SEY =2.2 ± 0.1
comparable to EP Nb

After sputtering away
~ 3 nm,
SEY down to 1.15