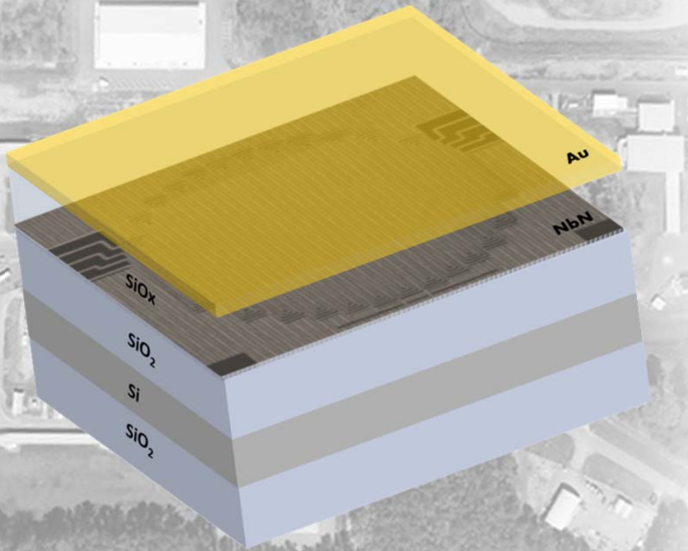
A-M Valente-Feliciano



Superconducting Thin Films @ JLab

Thomas Jefferson National Accelerator Facility is managed by
Jefferson Science Associates, LLC, for the U.S. Department of Energy's Office of Science

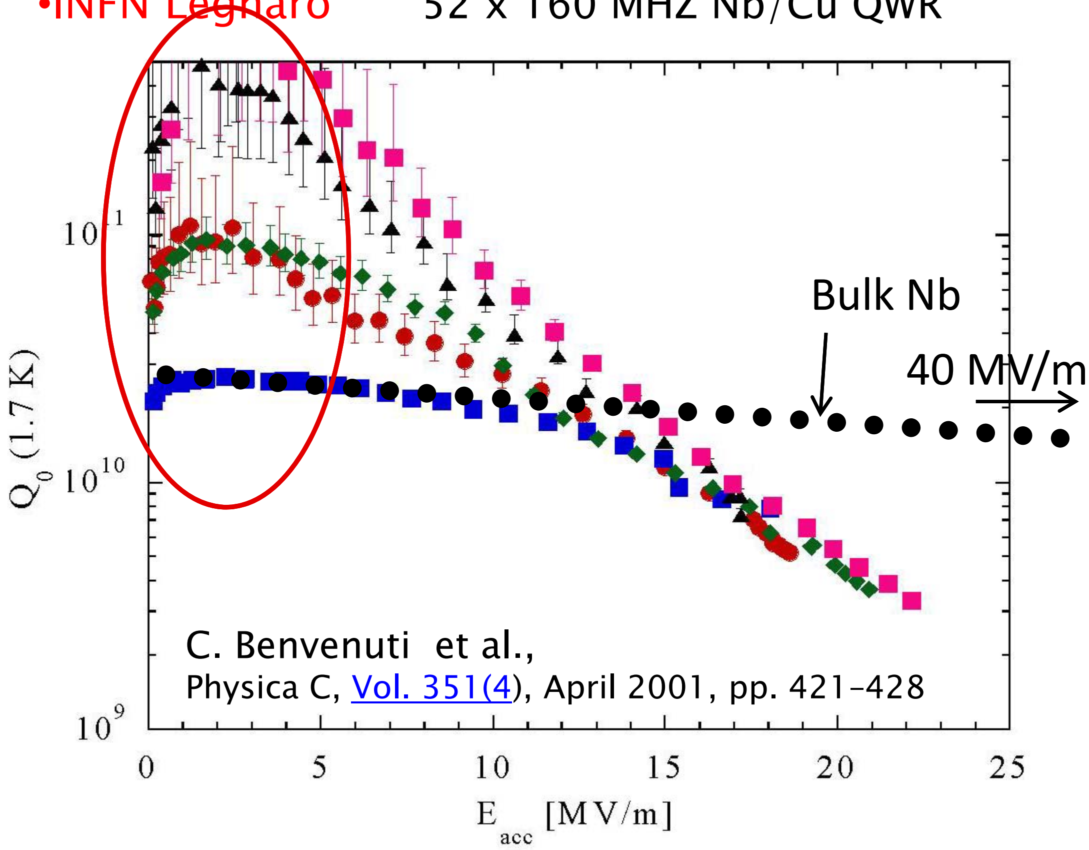
Outline

- ☐ Introduction
- ☐ Energetic Condensation of Nb films on Cu
- ☐ Development of SIS structures based on NbTiN
- ☐ Concluding Remarks

Thin Films for SRF - State of the Art

Thickness of interest for SRF applications = RF penetration depth, i.e. the very top 40 nm of the Nb surface.

•CERN LEP 2	272 x 353MHz Nb/Cu 4-cell cavities
•LHC	16 x 400 MHz Nb/Cu 1-cell cavities
•INFN Legnaro	52 x 160 MHZ Nb/Cu QWR



Bulk Nb

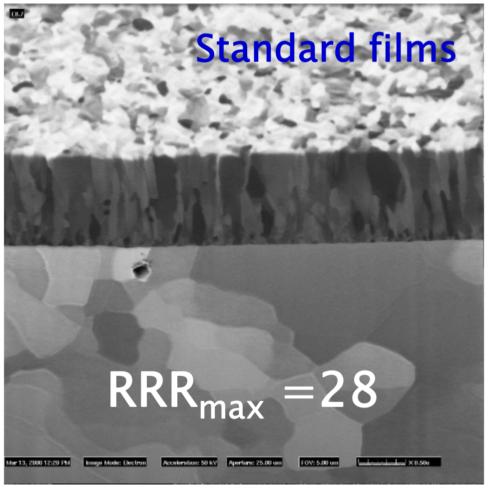
40 MV/m +

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

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

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

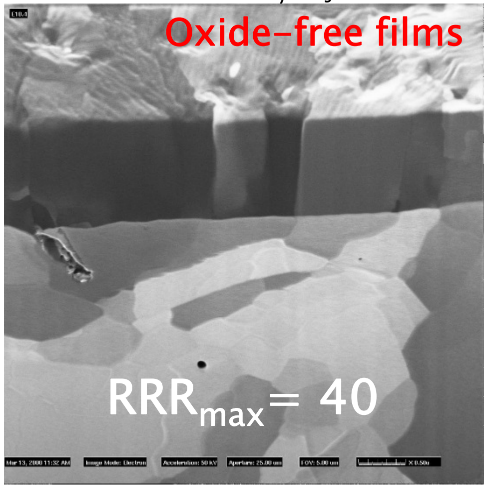


Standard films

RRR_{max} =28

Courtesy: P. Jacob - EMPA

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)



Oxide-free films

RRR_{max}= 40

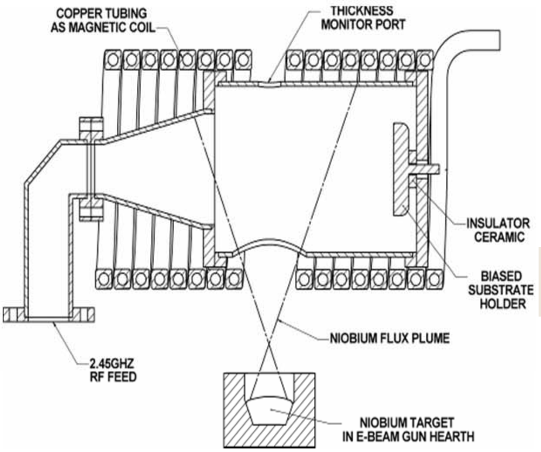
$$RRR_{Nb} = R_{300K}/R_{10K}$$

Gauge for mean free path & material quality, affected by scattering centers such as structural defects, impurities …

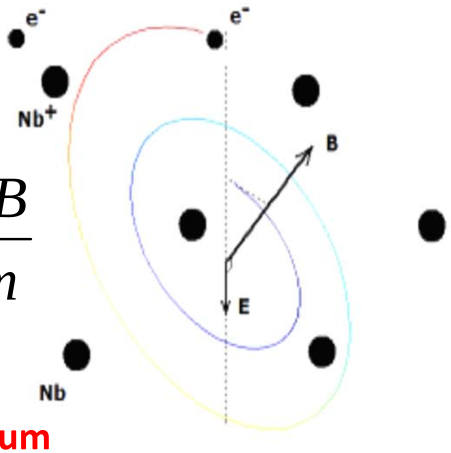
Energetic condensation with ECR

Generation of plasma - 3 essential components:

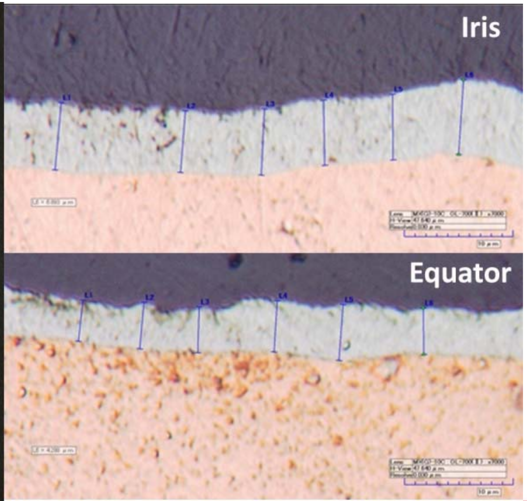
Neutral Nb vapor
RF power (@ 2.45GHz)
Static B $\perp$ E_{RF} with ECR condition



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



No working gas
Ions produced in vacuum
Singly charged ions **64eV**
Controllable deposition energy with Bias voltage
Excellent bonding
No macro particles
Good conformality

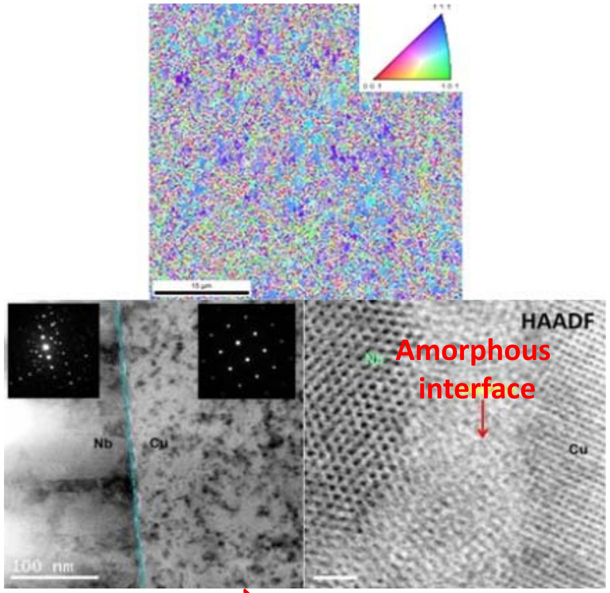


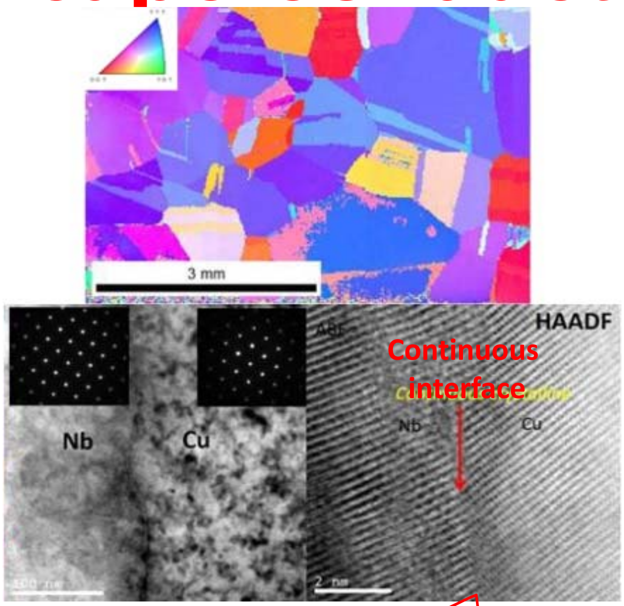
Conformality of the ECR process:

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

Note: the substrate is very rough, was only grossly mechanically polished

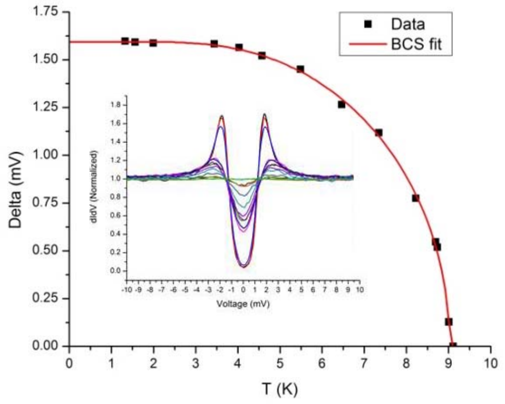
Structure, interface and superconducting gap

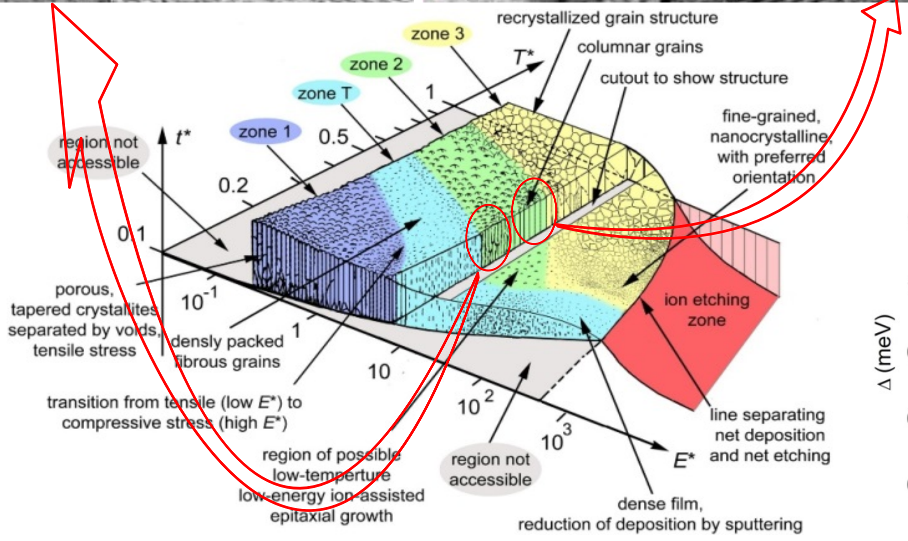




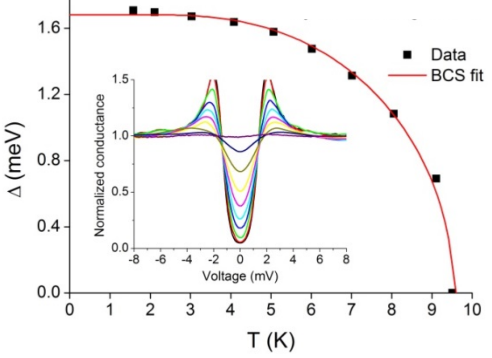
Gap measurements performed by PCT (point contact tunneling spectroscopy - T. Proslier)

Δ=1.56 meV



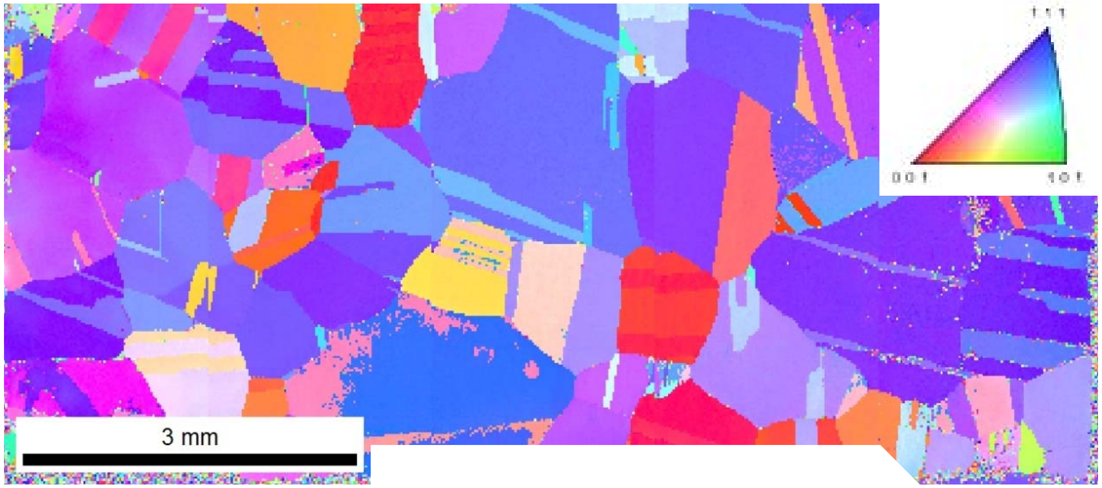


Δ=1.71 meV

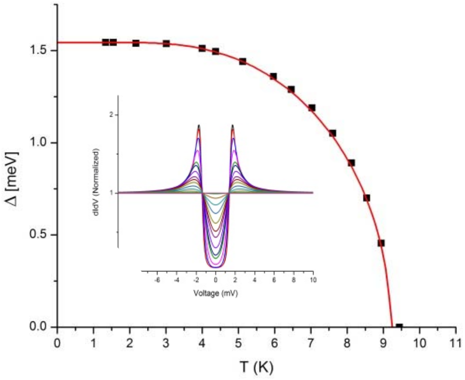


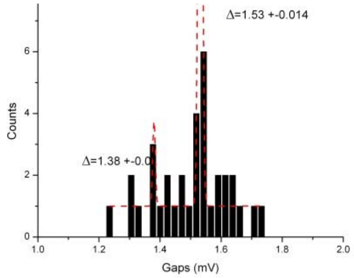
Superconducting gap (1.56-1.62meV) **similar to bulk Nb** ($\Delta_{\text{Nb bulk}}$ =1.55meV measured on the same setup) for hetero-epitaxial ECR Nb films on polycrystalline Cu.

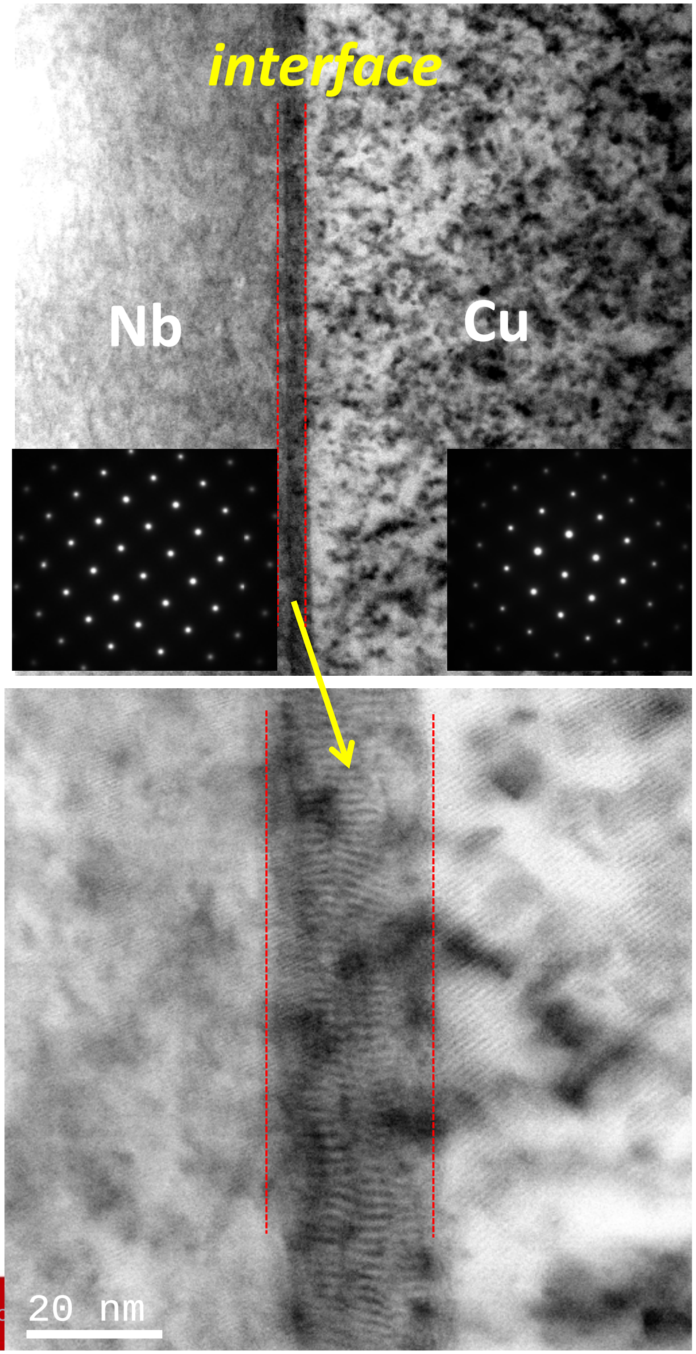
Hetero-epitaxy, High $T_{coating}$



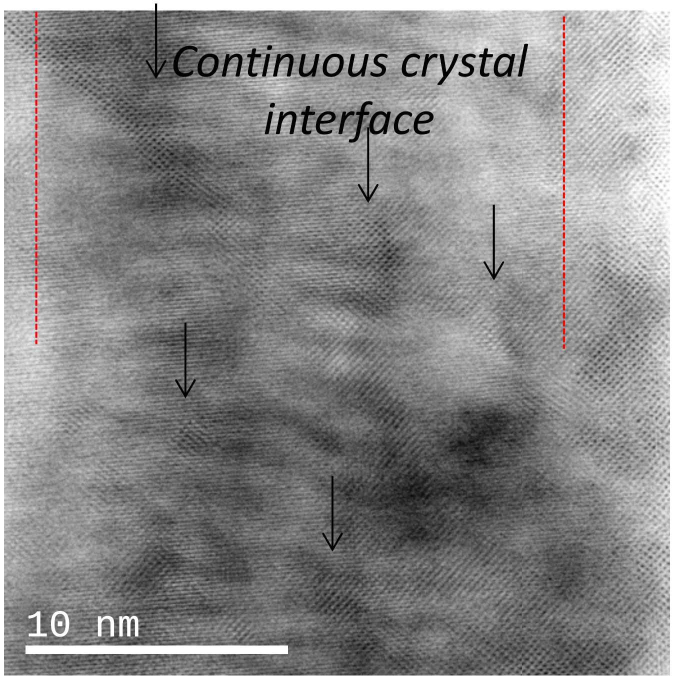
T_{bake} = 500 °C
$T_{coating}$ = 360 °C
$E_{Nb\ ions}$ = 184 eV
then 64 eV
Very thick film
Thickness = 4.5 µm
RRR = 305
T_c= 9.37 ± 0.12 K
Δ=1.53 meV (1.38 meV?)

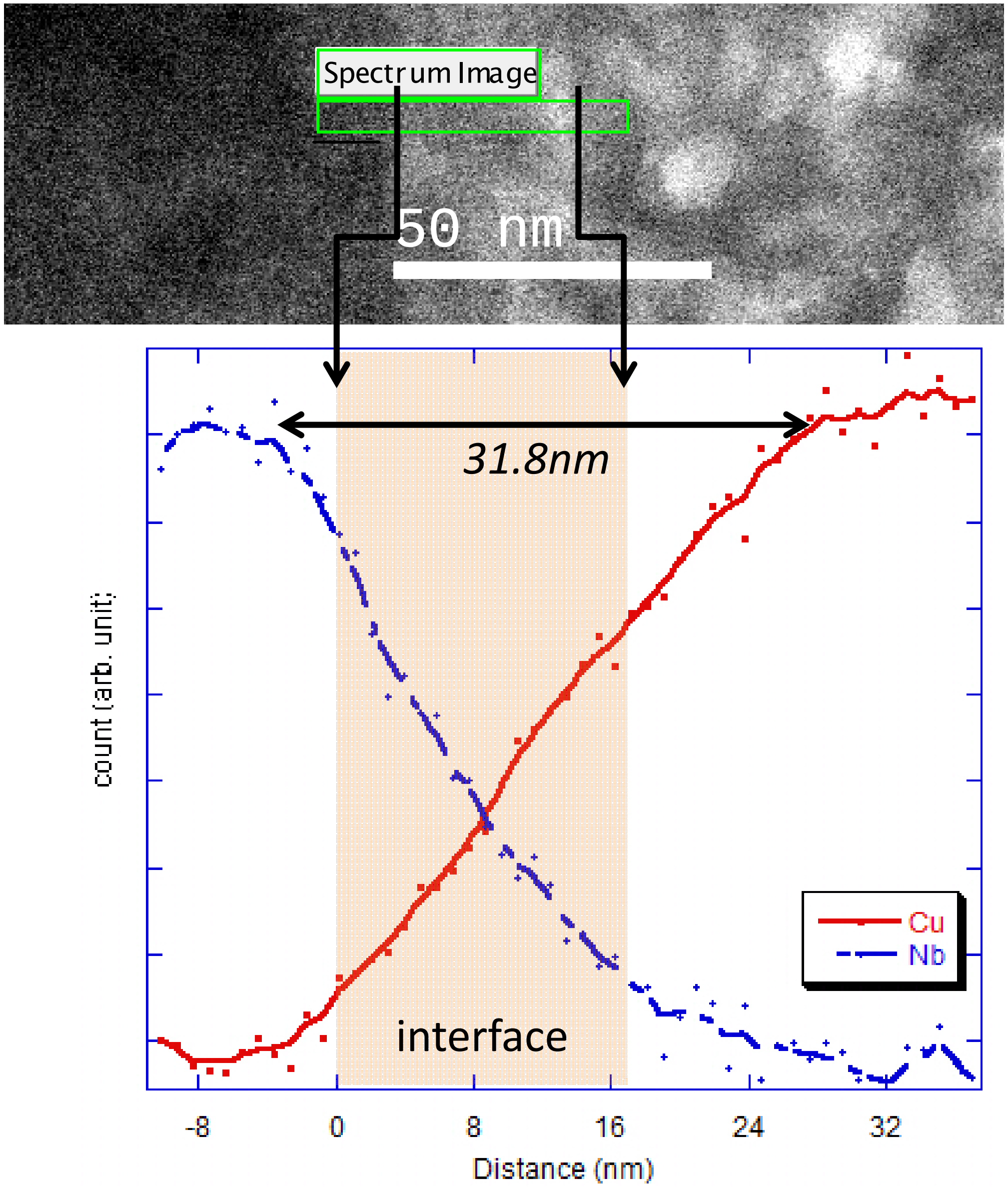






EELS plot for Cu/Nb signal across interface

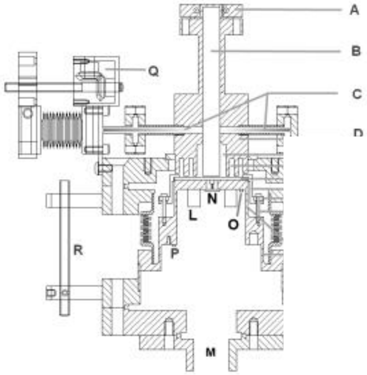




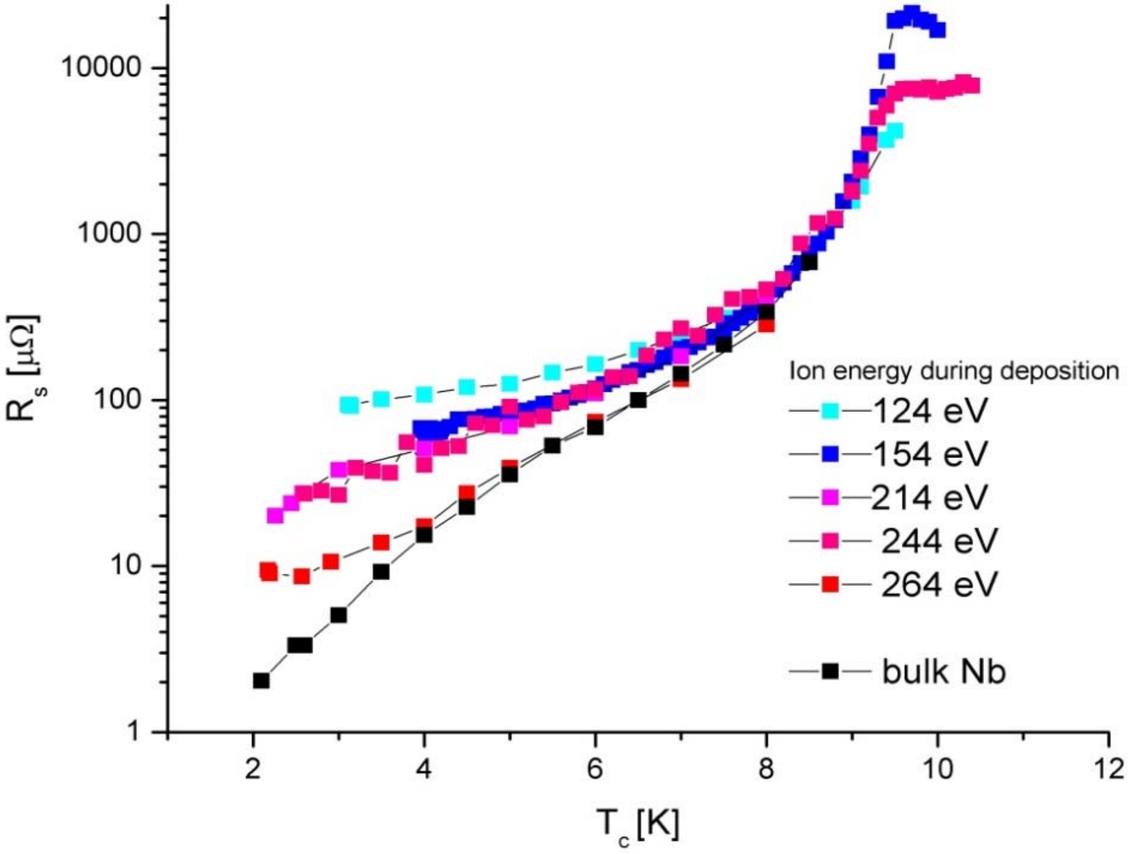
Interface thickness
(e^{-1} of highest density)

Nb: 12.5 nm
Cu: 20.1 nm

Influence of ion energy on surface resistance



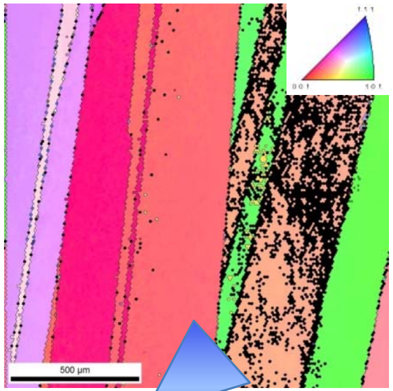
Growth at continuous energy (360 °C)

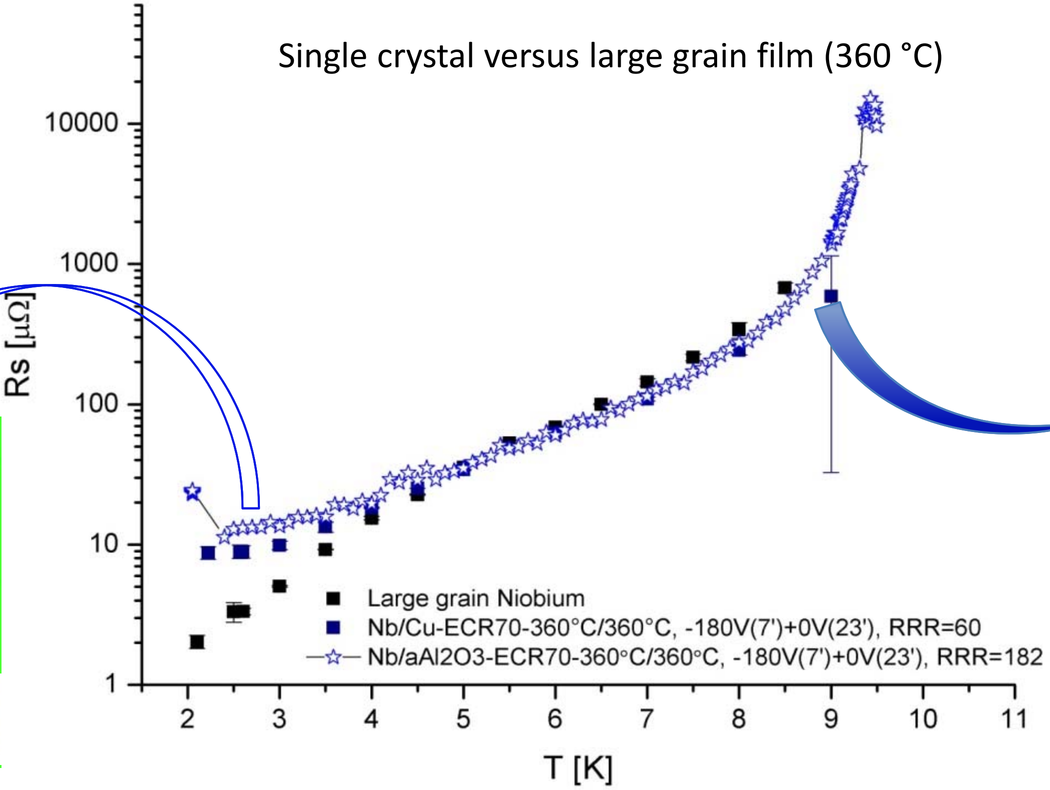


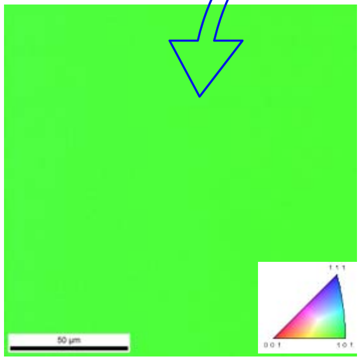
Decrease of R_s with increasing incident ion energy

ECR Nb/Cu – surface resistance

Polycrystalline Nb







Nb single crystal (110)

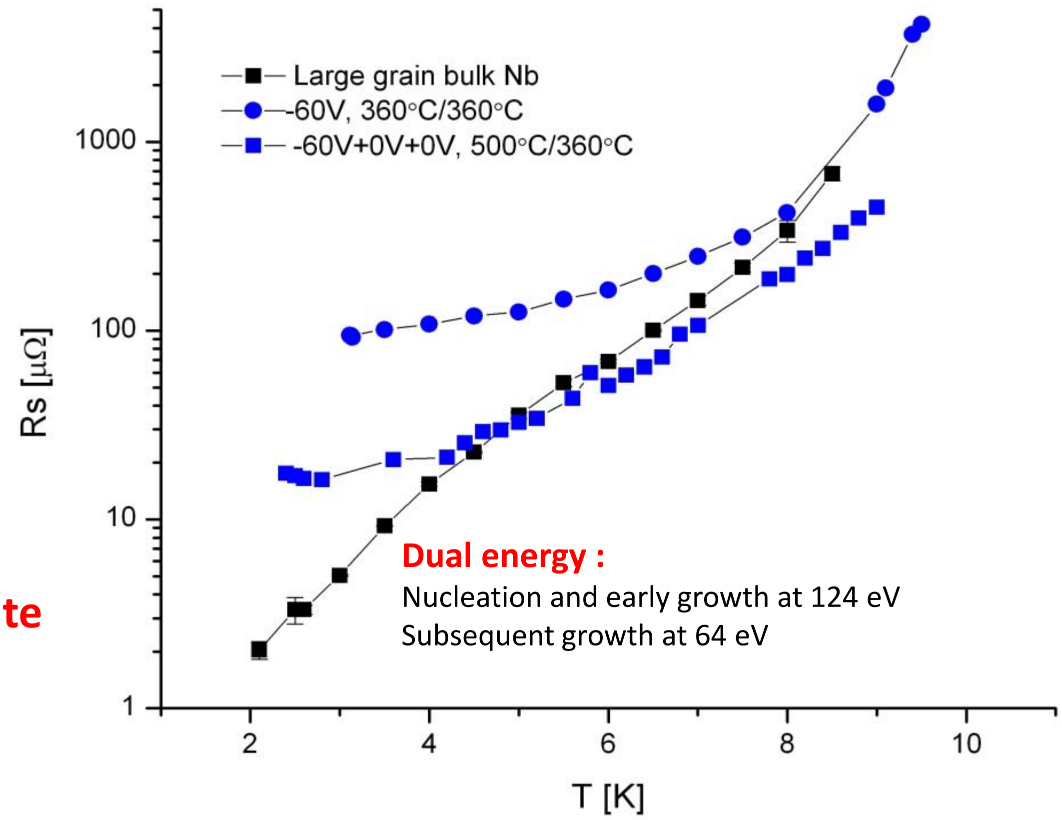
If dense, grain boundaries not necessarily detrimental to RF performance

ECR Nb/Cu – Surface resistance

Approach: 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.

- ☐ **Film nucleation on the substrate**
- ☐ **Subsequent Growth**

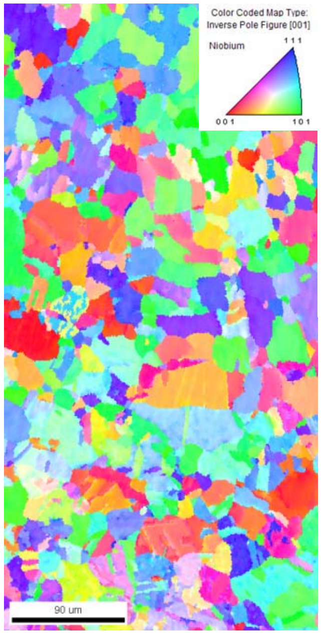


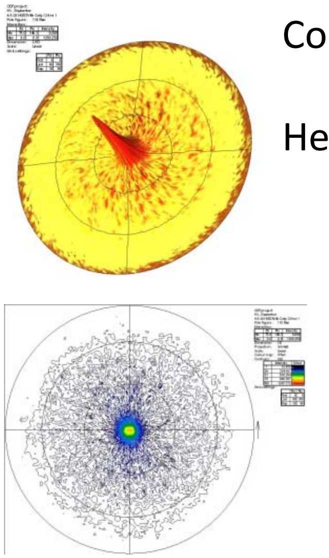
Dual energy :
Nucleation and early growth at 124 eV
Subsequent growth at 64 eV

Addressing the film deposition in 2 phases (nucleation @ high energy, subsequent growth @ 64 eV) shows some improvement in R_s

ECR Nb/Cu– R_S measured on QPR

A-M Valente-Feliciano, JLab
S. Aull, CERN





Bake & coating temperature: 360 °C

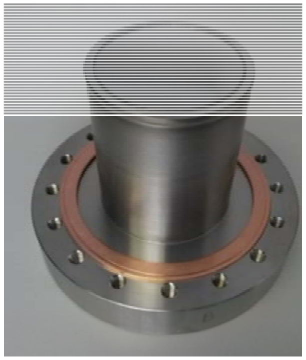
Coating with dual ion energy: 184 eV for nucleation/early growth
64 eV for subsequent growth

Hetero-epitaxial film Nb on OFHC Cu

T_c= 9.36 ± 0.12 K

RRR = 179 (Nb/a-Al_2O_3, witness sample)

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

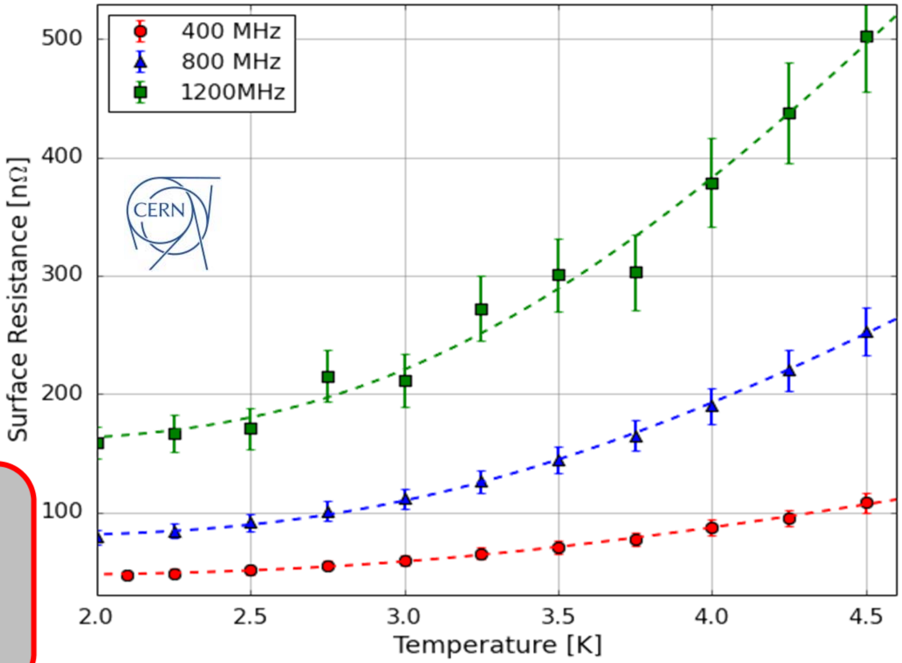


	R_{res} [nΩ]	λ(0K) [nm]
400 MHz	46.6 ± 0.8	40 ± 2
800 MHz	79 ± 2	38 ± 1
1200 MHz	156 ± 11	38 ± 1

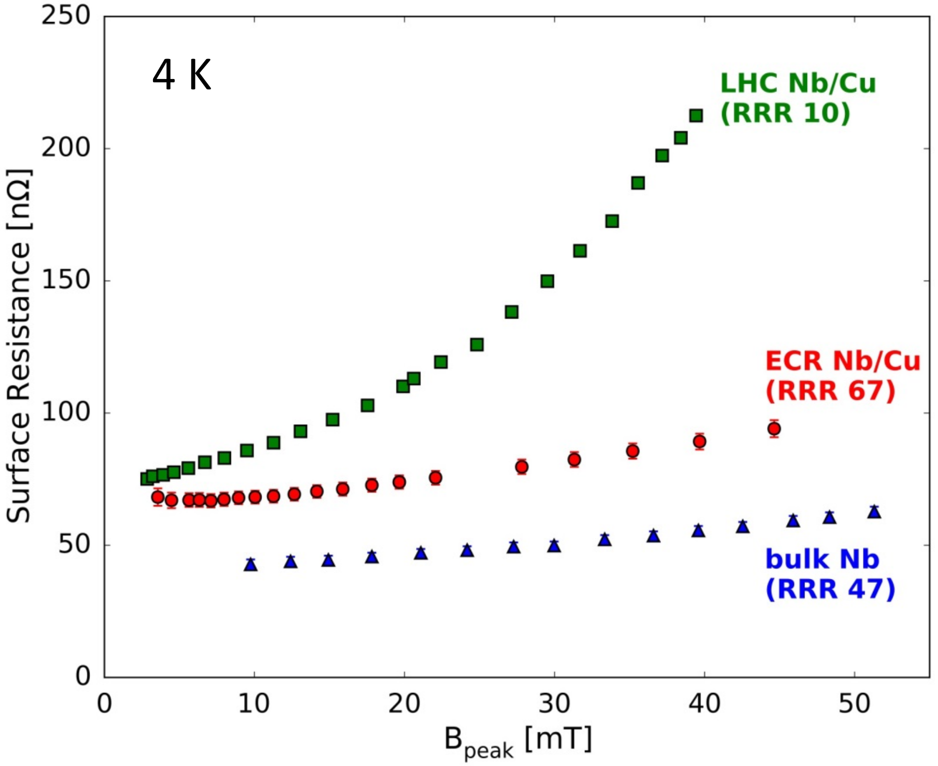
ℓ* [nm]	RRR
144 ± 20	53 ± 7

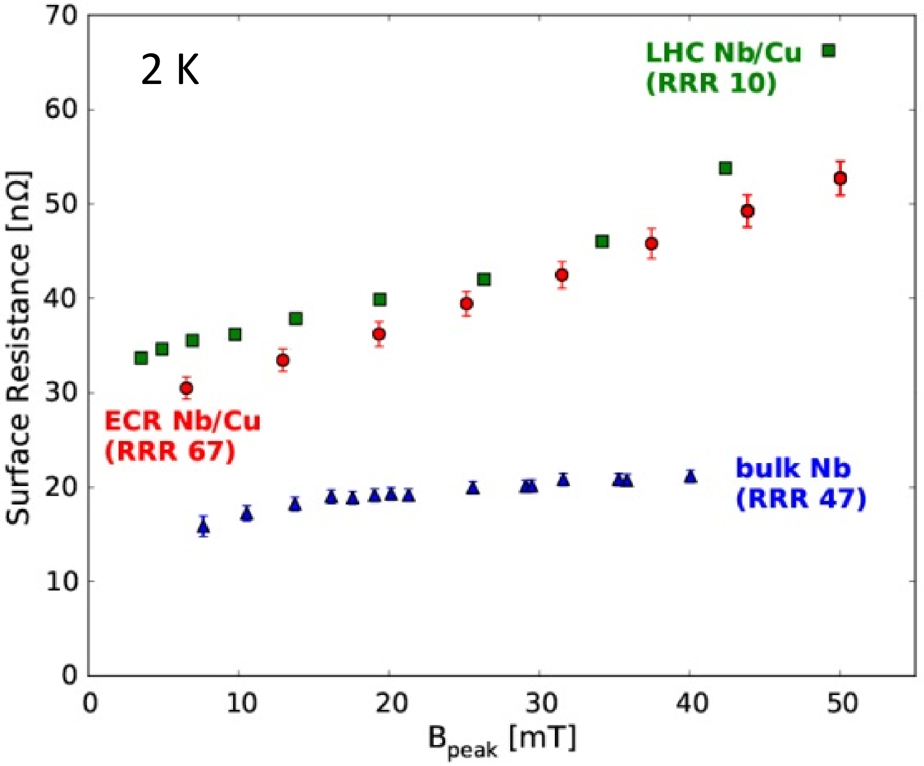
* with λ_L = 32 nm and ξ_0 = 39 nm

Nb film in the clean limit



QPR RF Measurements (400 MHz)
Comparison with Sputtered Nb/Cu and bulk Nb





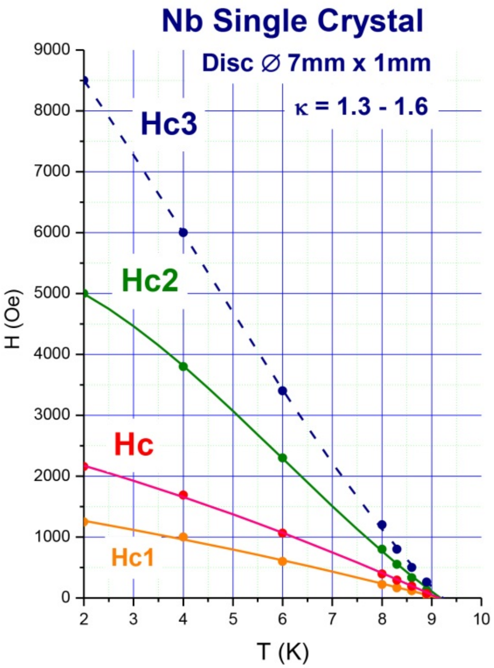
S. Aull

Significantly reduced Q-slope at 4 K

Flux Penetration Measurements on ECR Nb films

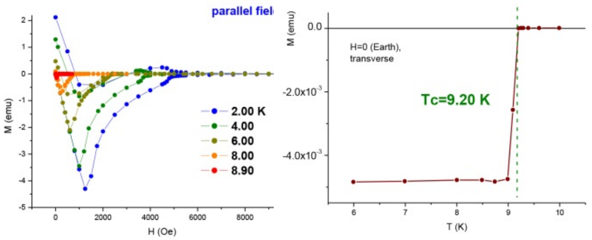
Magnetometry (@ Leuven, V. Kozhevnikov)

Magnetic structure of superconductors in the intermediate (type-I) and in the mixed (type-II) states


Nb Single Crystal

Disc ∅ 7mm x 1mm

κ = 1.3 - 1.6

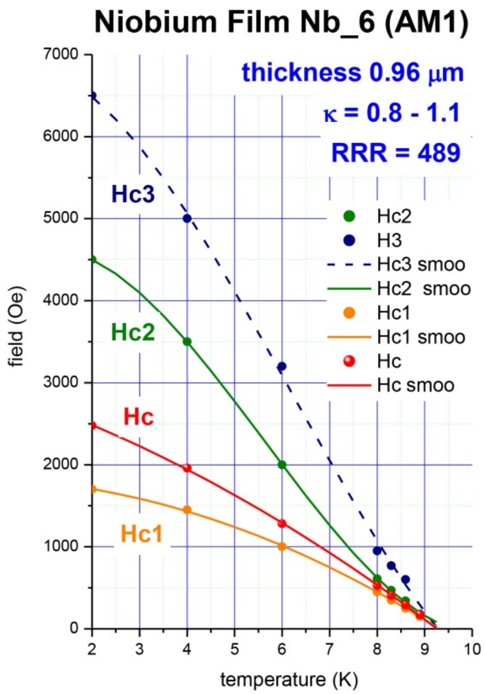


H_{c1}=125 mT

H_c=216 mT

Nb/a-Al_2O_3

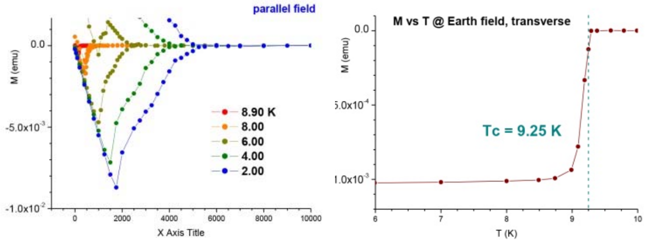
184 eV, 500 °C


Niobium Film Nb_6 (AM1)

thickness 0.96 μm

κ = 0.8 - 1.1

RRR = 489

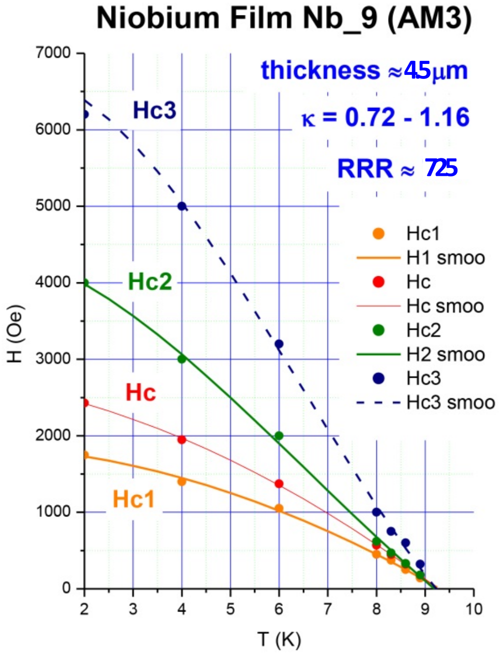


H_{c1}=**170** mT

H_c=248 mT

Nb/r-Al_2O_3

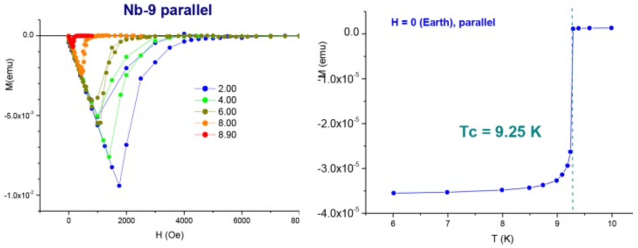
124 eV/64 eV, 360 °C


Niobium Film Nb_9 (AM3)

thickness ≈45μm

κ = 0.72 - 1.16

RRR ≈ 725

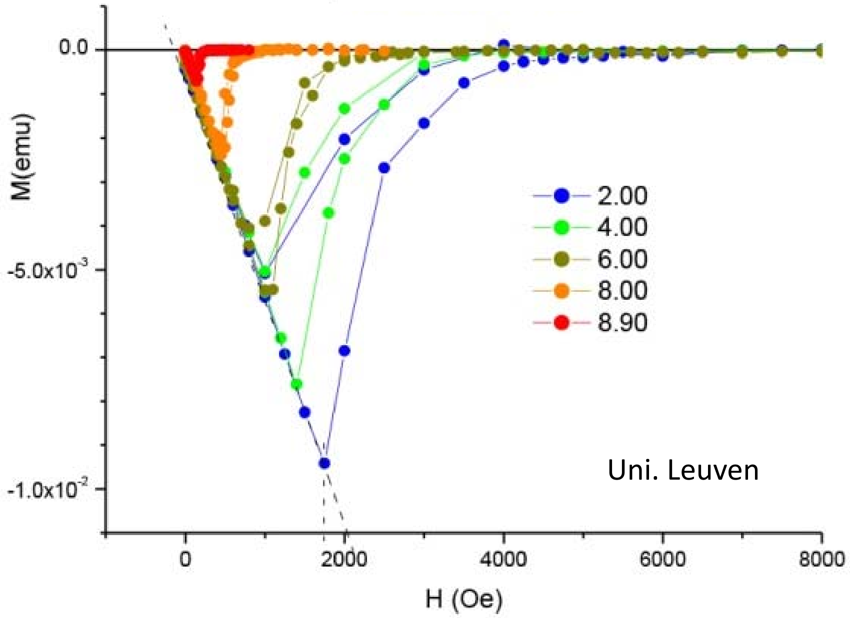


H_{c1}=**175** mT

H_c=243 mT

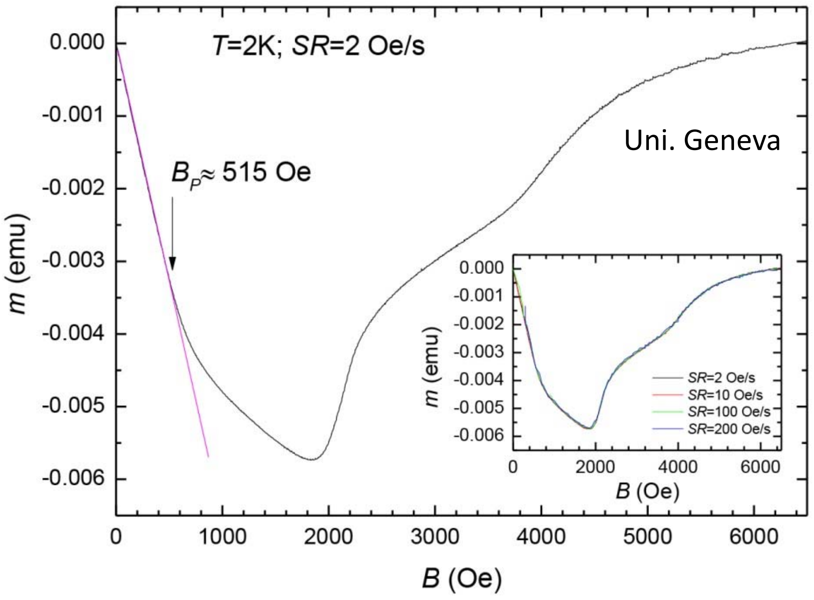
Flux Penetration Measurements on ECR Nb Films

Nb/ r-Al_2O_3



H_{c1} = 175 mT
RRR = 725

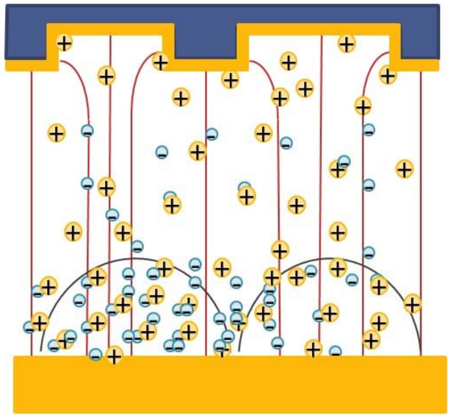
Nb/ Cu

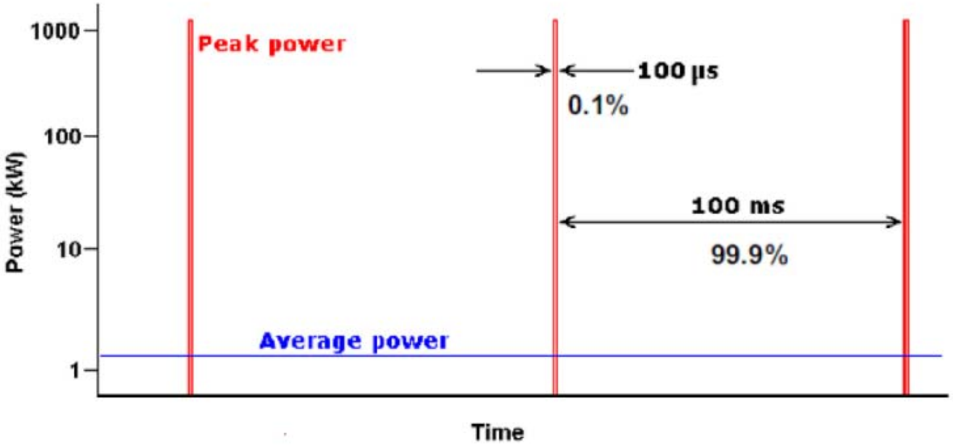


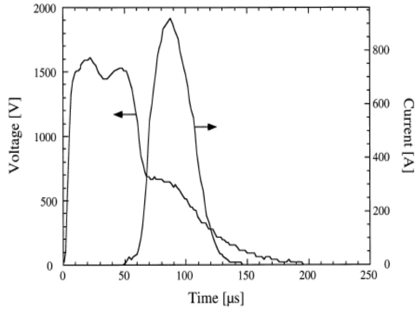
H_{c1} = 51.5 mT
RRR = 67

Verify measurement on the same setup at W&M for the same samples

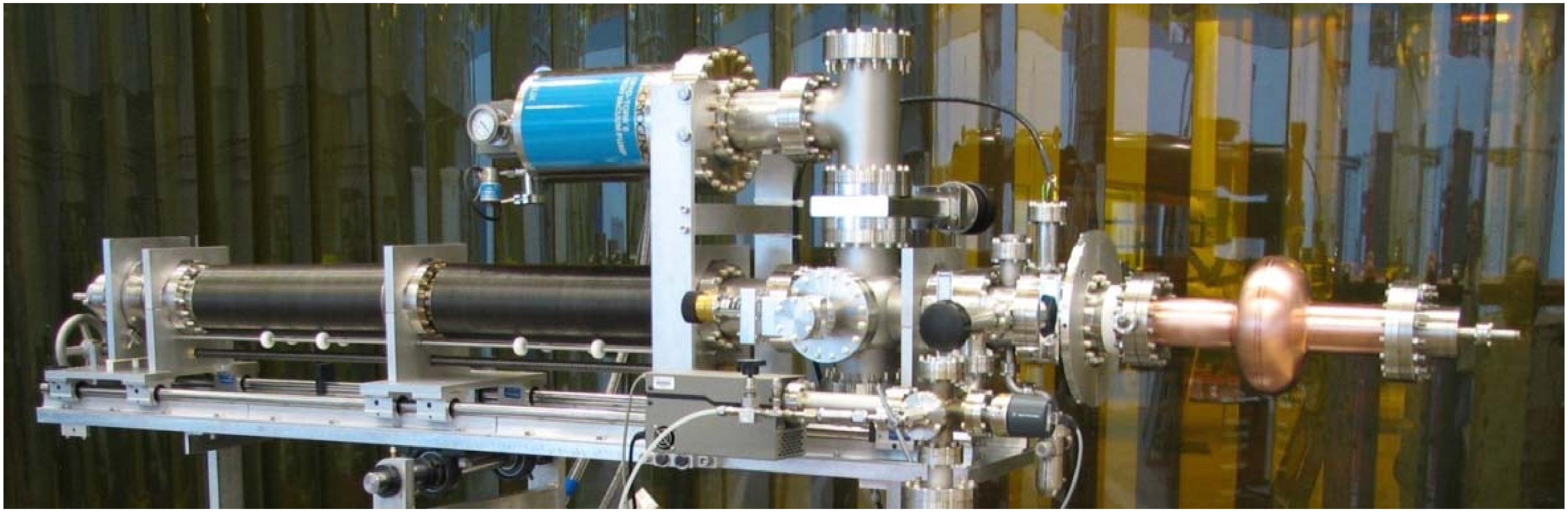
Energetic condensation with HiPIMS







V. Kouznetsov, *et al.*, Surf. Coat. Technol. **122** (1999) 290



❑ Development of a new HiPIMS power supply

❑ First single cells coated and RF tested

M. Burton, L. Phillips

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
- RF performance: indication of a reduced Q-slope

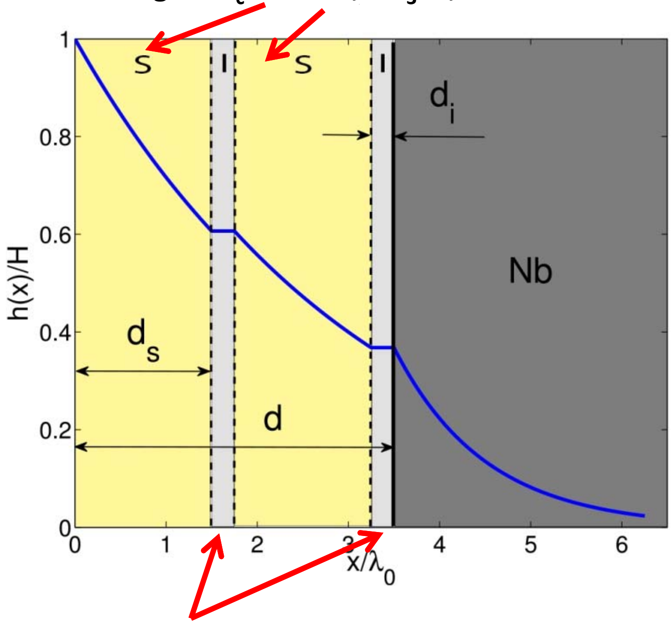
		Substrate	RRR_{max}
Insulating	Single crystal	a-Al_2O_3	488
		r-Al_2O_3	641
		c-Al_2O_3	247
		MgO (100)	188
		MgO (110)	424
		MgO (111)	270
	Polycrystalline amorphous	Al_2O_3 ceramic	135
		AlN ceramic	110
		Fused Silica	84
Metallic	Single crystal	Cu (100)	181
		Cu (110)	275
		Cu (111)	245
	Polycrystalline	Cu fine grains	193
		Cu large grains	305

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)

Higher-T_cSC: NbN, Nb_3Sn, etc



Insulating layers

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

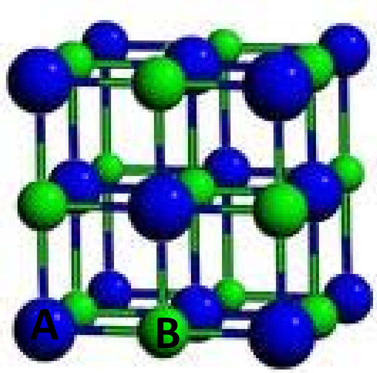
- Strong increase of H_{c1} in films allows using RF fields > H_c of Nb, but lower than those at which flux penetration in grain boundaries may become a problem=> no transition, no vortex in the layer
- High H_{C1} ,applied field is damped by each layer
- Insulating layer prevents Josephson coupling between layers
- Applied field, i.e. accelerating field can be increased without high field dissipation
- Strong reduction of BCS resistance (ie high Q_0) because of using SC layers with higher T_c, Δ (Nb_3Sn, NbN, etc)

Possibility to move operation from 2K to 4.2K

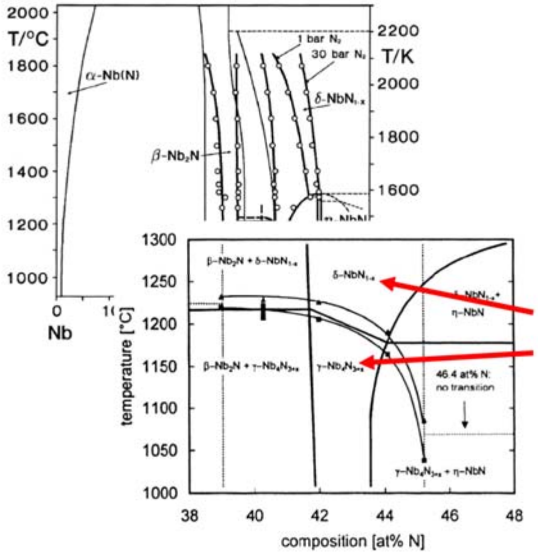
Choice of Materials for S-I-S structures

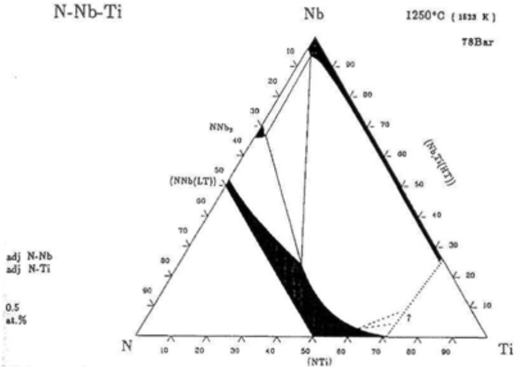
Ternary Nitride $(Nb_{1-x},Ti_x)N$ (T_c=17.3 K, a= 4.341 Å)

Presence of Ti found to reduce significantly the resistivity
And facilitate formation of a pure cubic structure.
The δ-phase remains thermodynamically stable even at RT.
T_c as high as for good quality NbN, for Nb fraction (1-x)>0.5



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





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

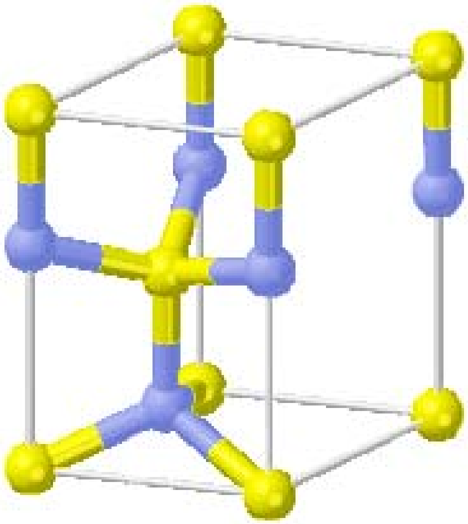
$\gamma - NbN \rightarrow T_c \sim 12 - 15\ K$

extreme hardness, excellent adherence on various substrates, very good corrosion and erosion resistance, high-sublimation temperature, and relative inertness

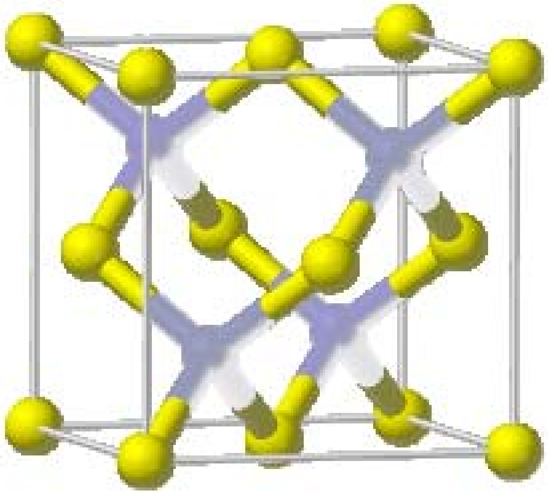
Choice of dielectric for S-I-S structures

AlN is an insulator that :

- can be grown with a wurtzite (hcp, a=3.11Å, c=4.98Å) or sphalerite (B1 cubic, a= 4.08 Å) structure.
- has been found to enhance the properties (T_c) of NbN and NbTiN, in particular for very thin films (THz mixers) .
- has a large thermal conductivity (3.19 W/cm.K at 300 K, comparable with Cu, 4.01 W/cm.K)



Wurtzite structure



Sphalerite structure

Experimental Setup

Base pressure range: 10^{-10} Torr

- ☐ 3 x 2” DC/RF Magnetrons
- ☐ Ion source

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

<u>Substrates:</u>

MgO
AlN ceramic
Bulk Nb
ECR Nb films

CHALLENGES

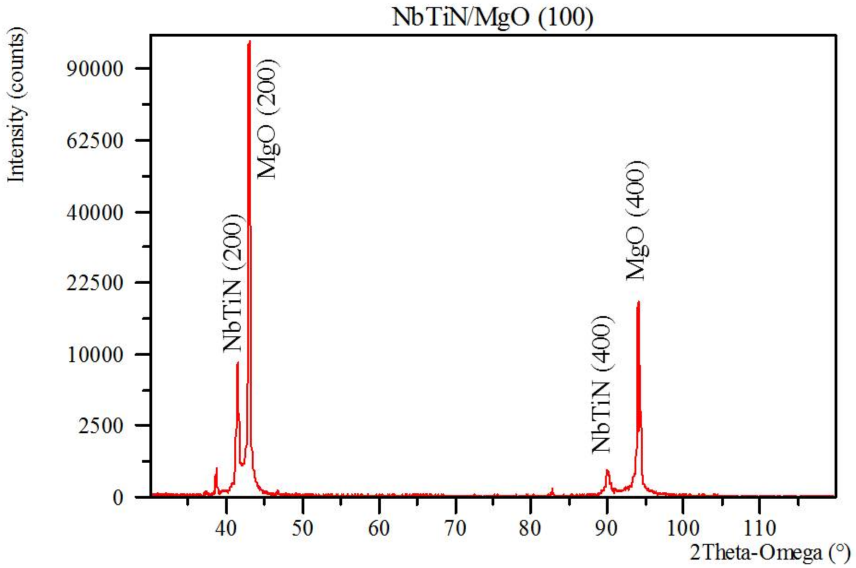
- ☐ Develop good quality and uniform thin layers
- ☐ Sharp interfaces
- ☐ Growth of equally performing S/I and I/S layers

NbTiN film

NbTiN are grown on various substrates at 600°C by reactive sputtering with targets of different Nb/Ti weight ratios.

Films exhibit good crystalline structure in general

Best results at 600 °C on MgO



NbTiN grown at 600°C on MgO

Thickness = 2µm

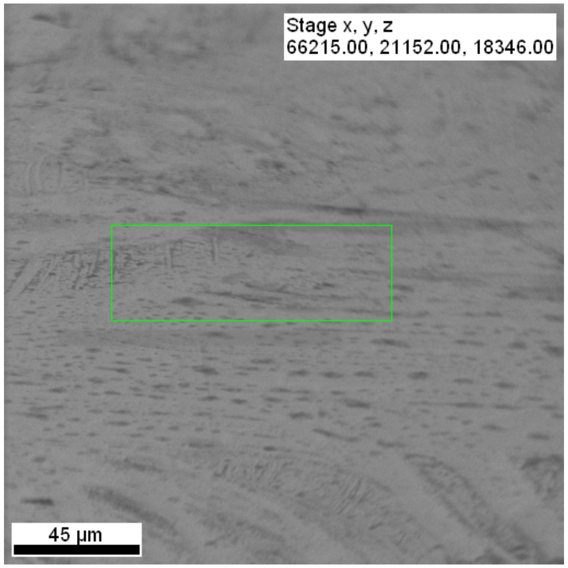
Lattice parameter = 4.34 Å

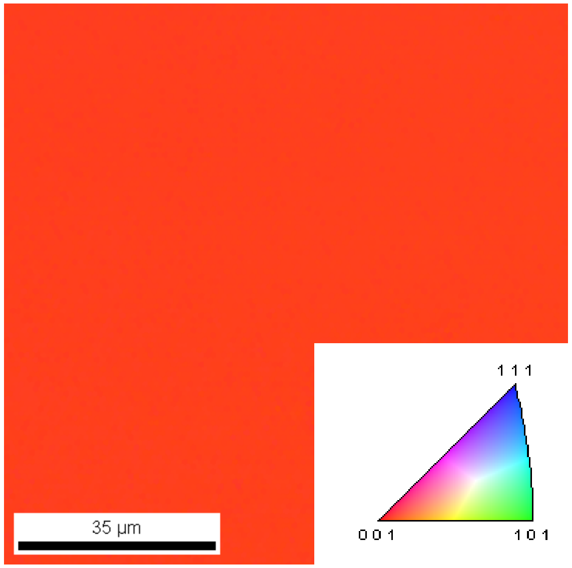
High quality single crystal

T_c= 17.25 K

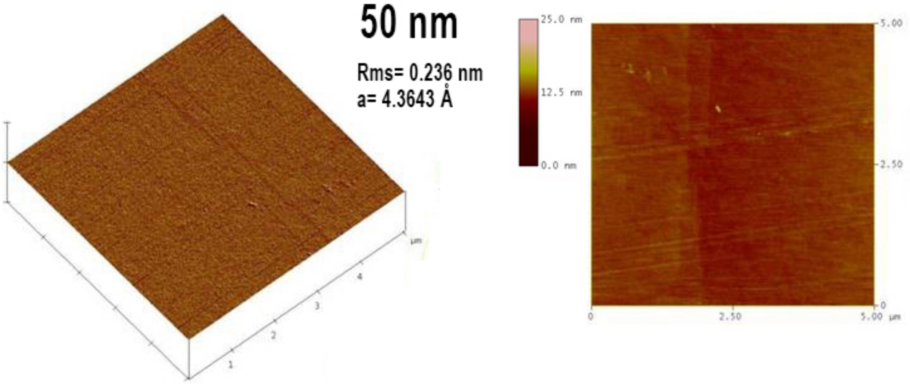
H_{c1} = 30 mT

Bulk-like film



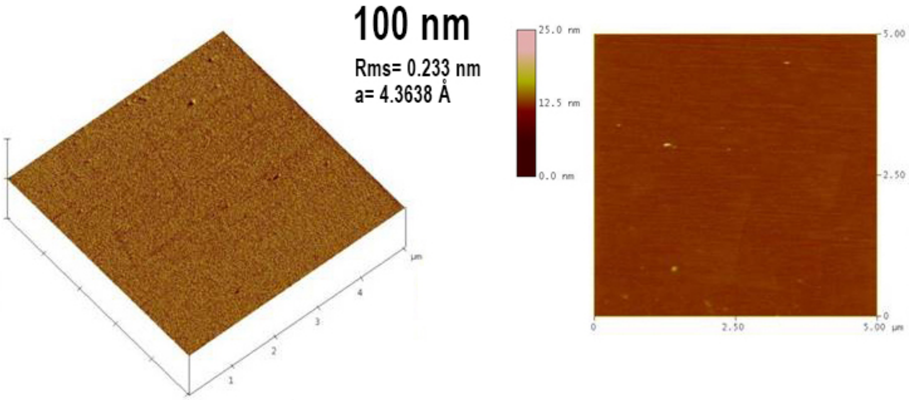


NbTiN Films – Influence of Thickness on T_c



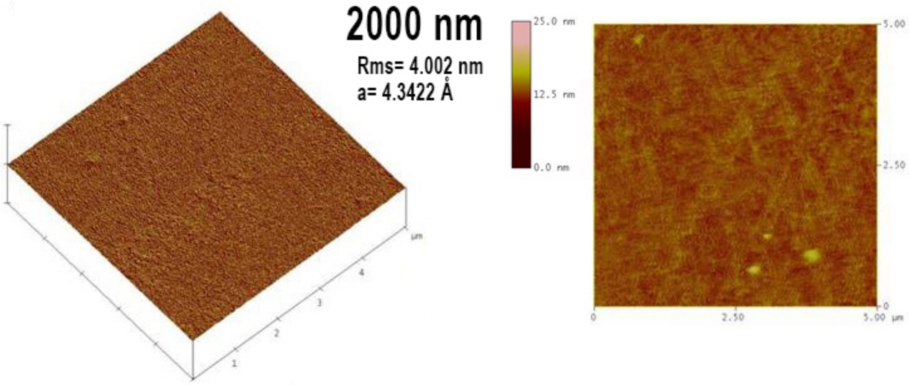
50 nm

Rms= 0.236 nm
a= 4.3643 Å



100 nm

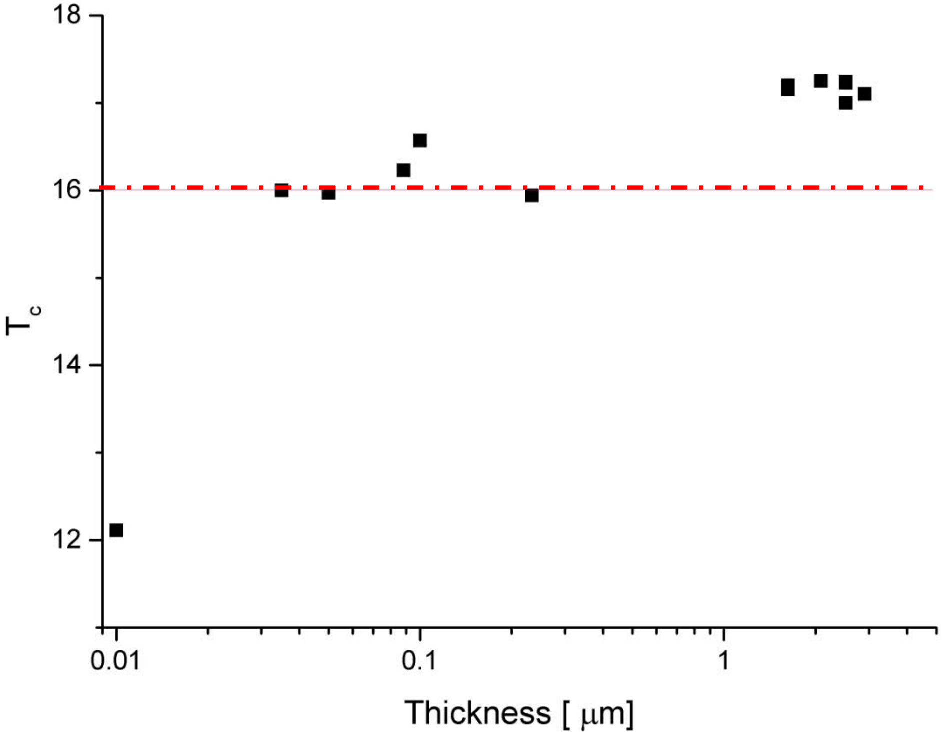
Rms= 0.233 nm
a= 4.3638 Å



2000 nm

Rms= 4.002 nm
a= 4.3422 Å

Single crystal NbTiN/MgO films (XRD/EBSD)
Very smooth films (~ substrate)

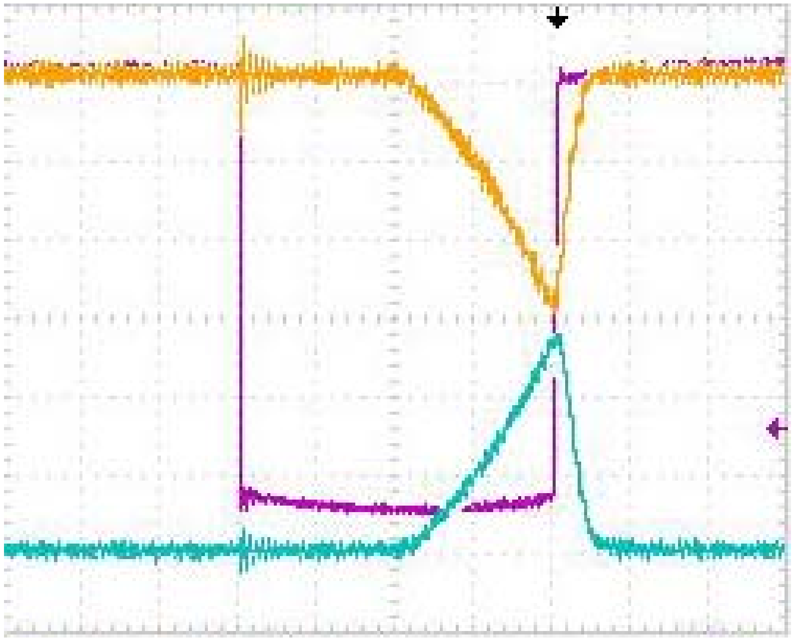


T_c ≥16 K for
film thickness > 35 nm

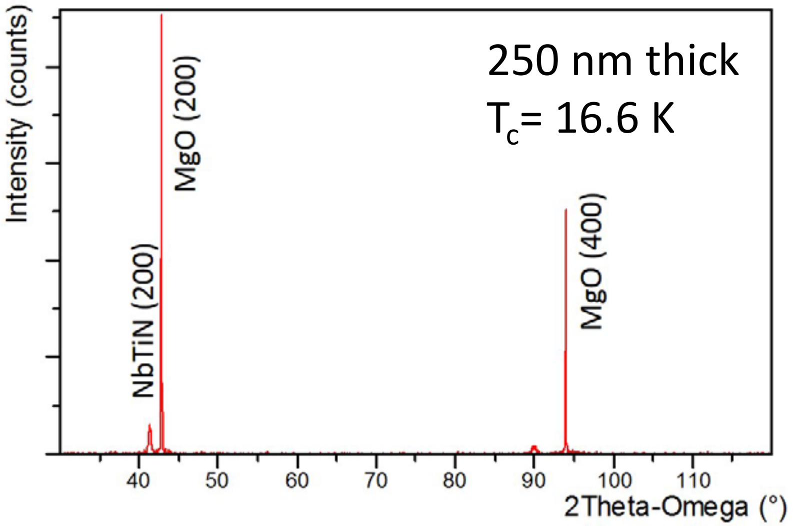
NbTiN Films with HiPIMS

Thickness [nm]	Average Power [W]	Peak current [A]	Pulse width [µs]	Repetition rate [Hz]	Coating time [min]
30	100	115	100	100	120
250	400	140	100	200	120
230	400	100	150	200	30
118	400	150	100	200	30
252	400	150	100	200	60
218	400	150	100	200	120

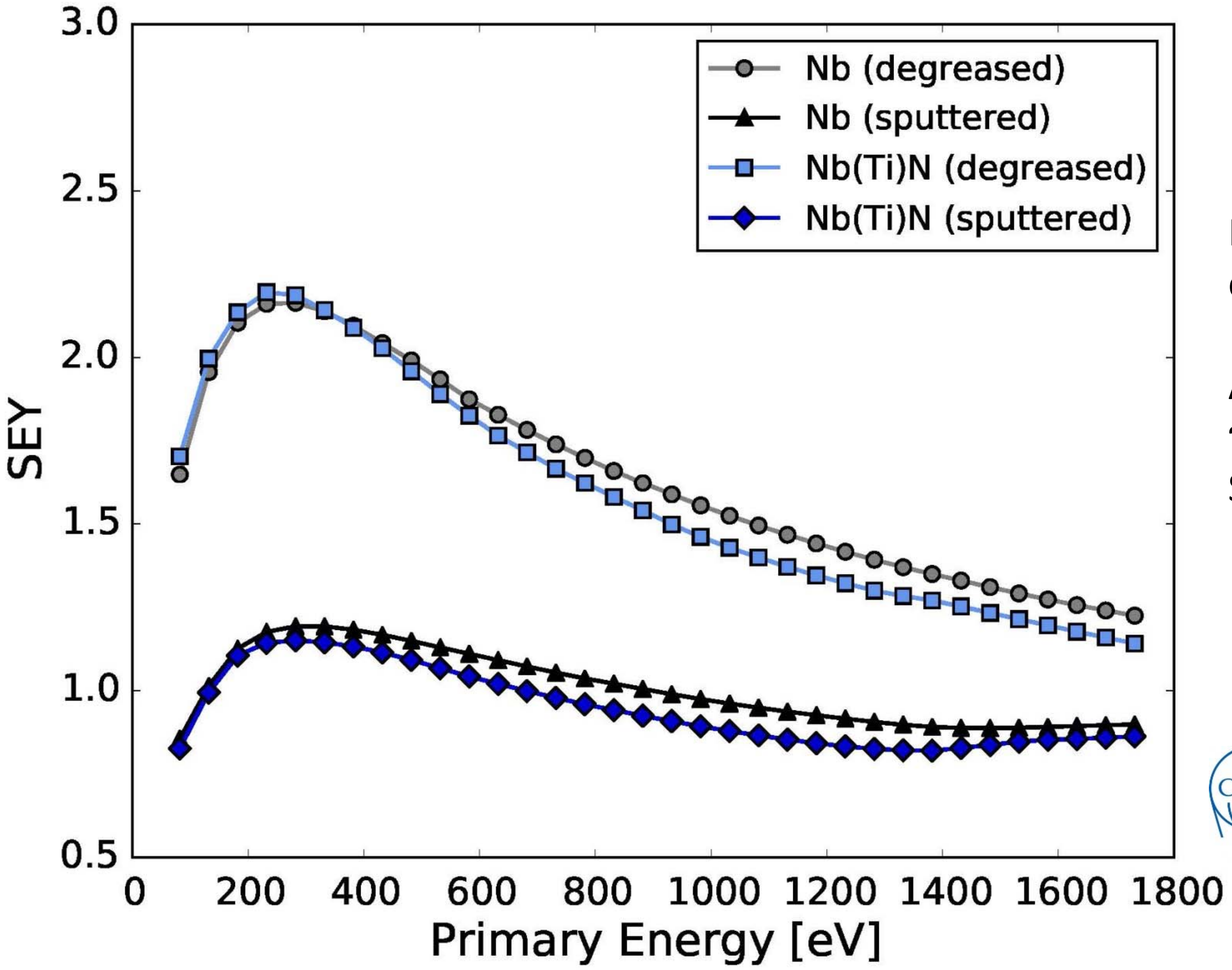
θ-2θ scans of the first films produced by HiPIMS reveal that only the films produced with an average power of 400 W and repetition rate of 200 Hz have the δ-phase. The measured T_c is 16.6 K for a 250 nm thick film.



Typical pulse for reactive HiPIMS of NbTiN



Secondary Electron Yield of NbTiN Films

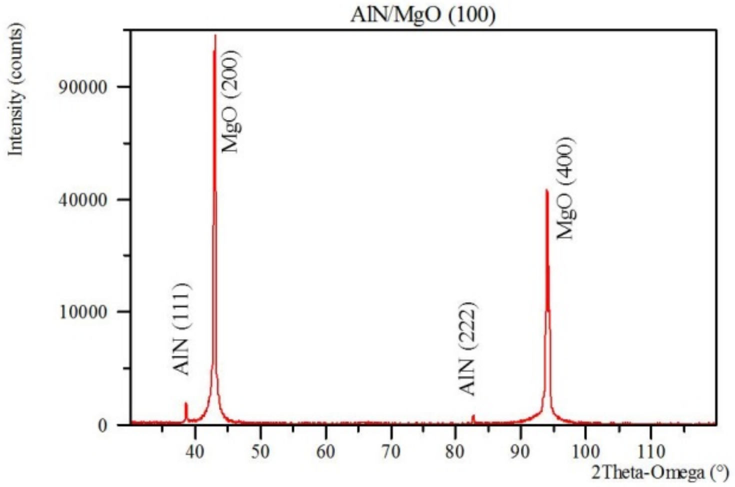


Measurements at
room temperature

Max. SEY =2.2 ± 0.1
comparable to EP Nb

After sputtering away
~ 3 nm,
SEY down to 1.15

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

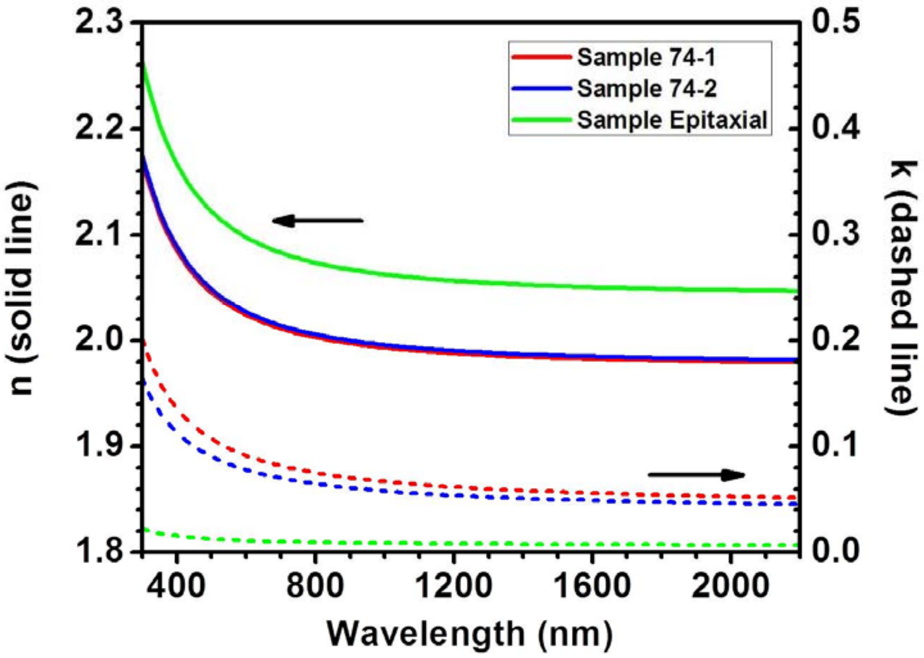
Roughness - EMA with 50% Void (XRR thickness used)

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



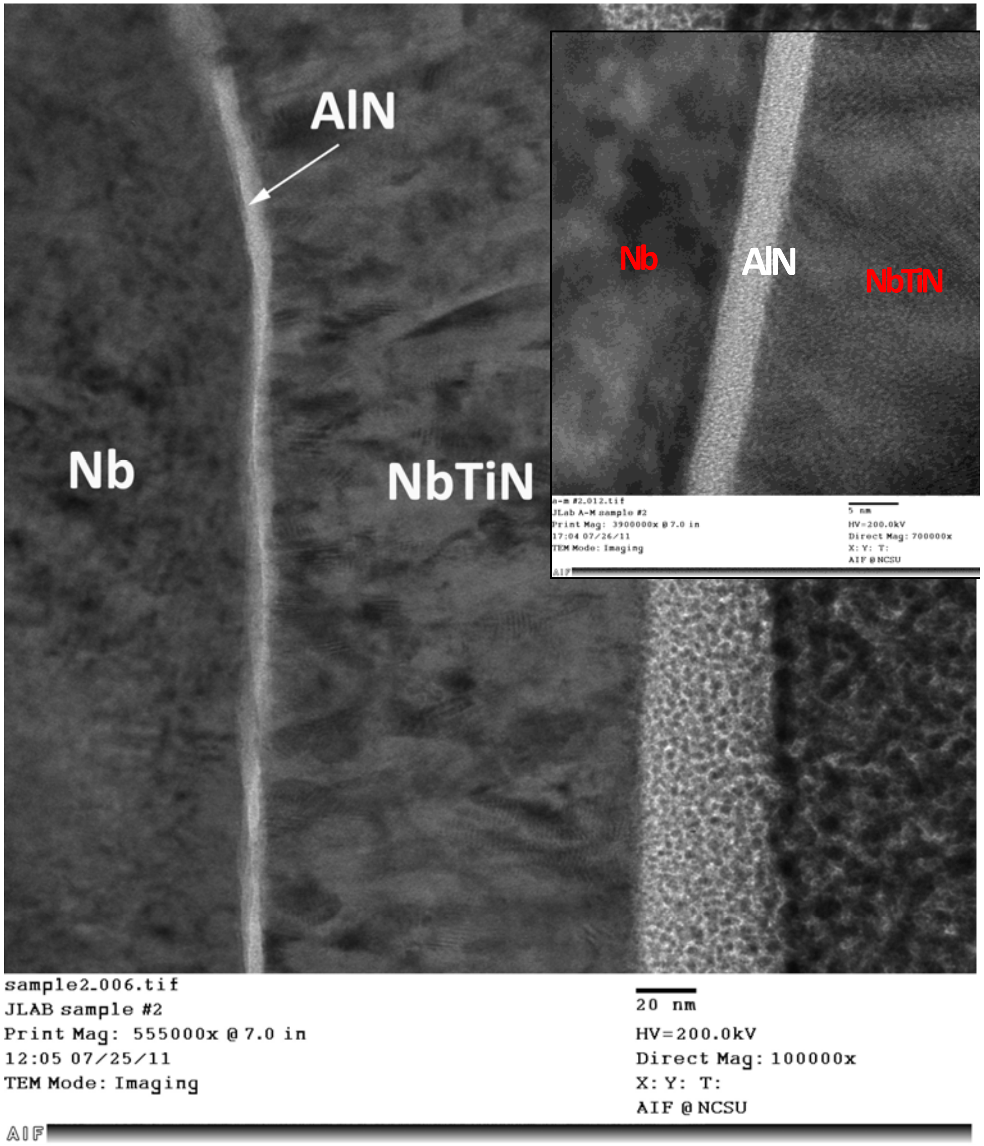
SRF Multilayer Structures Based on NbTiN
Influence of coating temperature

NbTiN/AlN/Nb film
at 600 °C

	AlN	NbTiN
N_2/Ar	0.33	0.23
Total pressure [Torr]	2x10^{-3}	2x10^{-3}
Sputtering Power [W]	100	300
Deposition rate [nm/min]	~ 2.5	~ 18
Thickness [nm]	5	100
T_c [K]	N/A	14

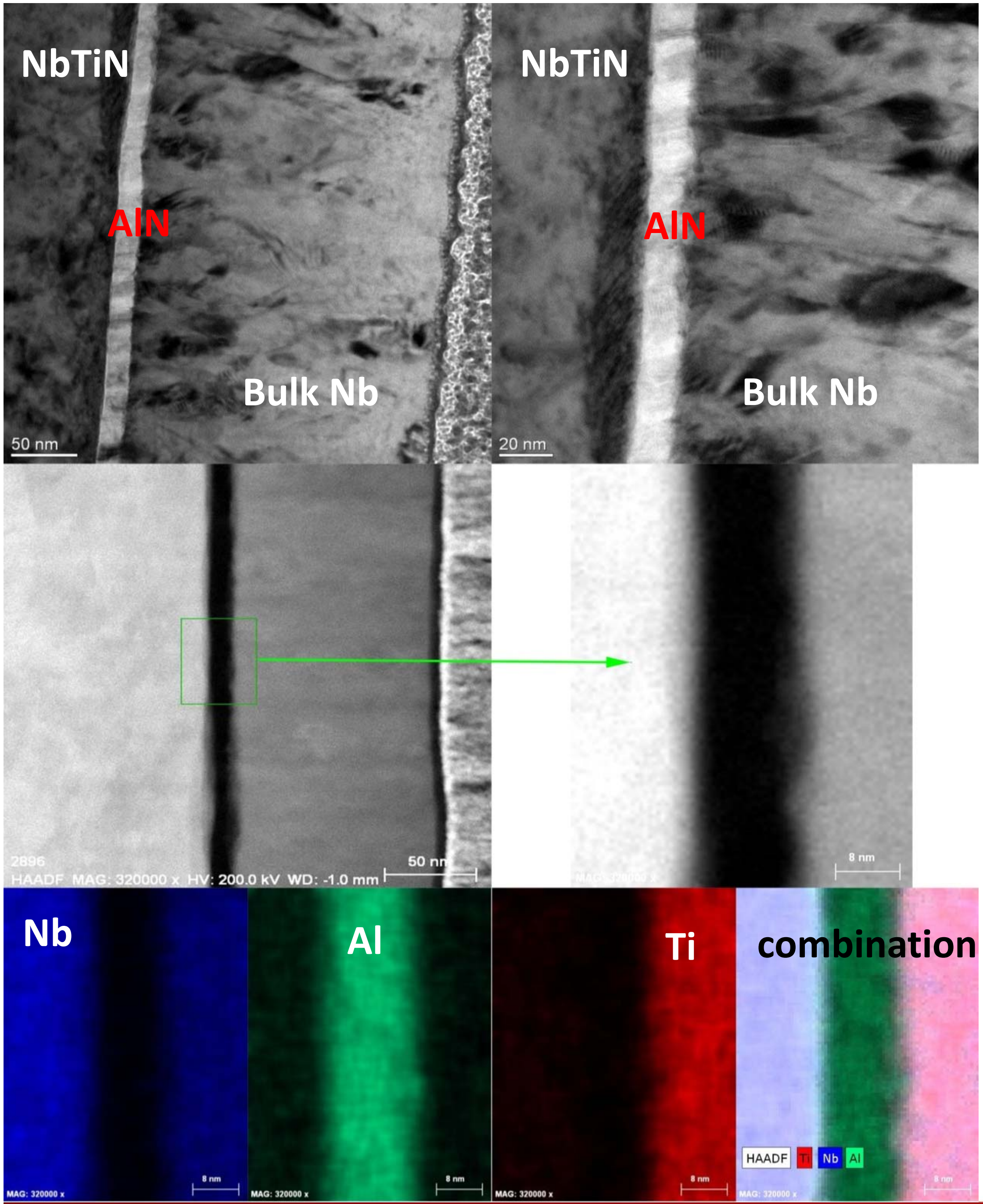
TEM cross-section (FIB cut)
of NbTiN/AlN/Nb/Cu
structure

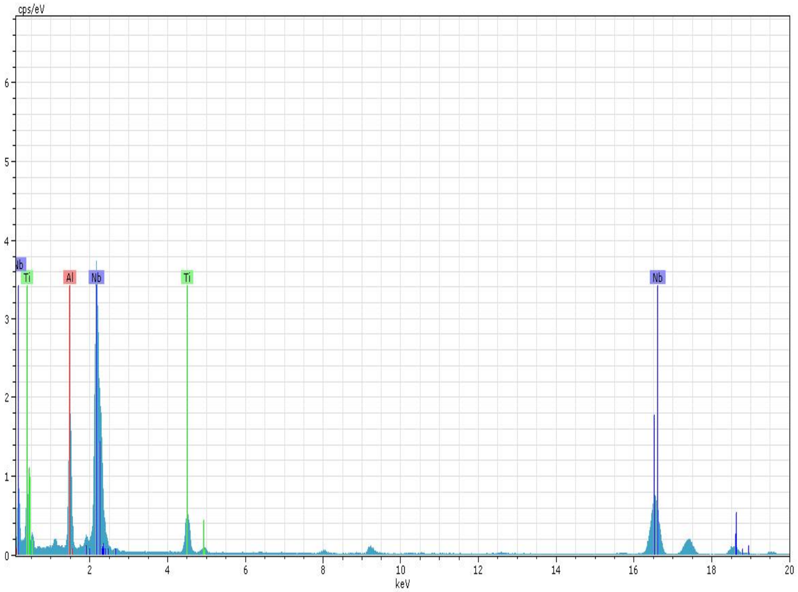
Miscibility of AlN into Nb and NbTiN at 600 °C



NbTiN/AlN on bulk Nb

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

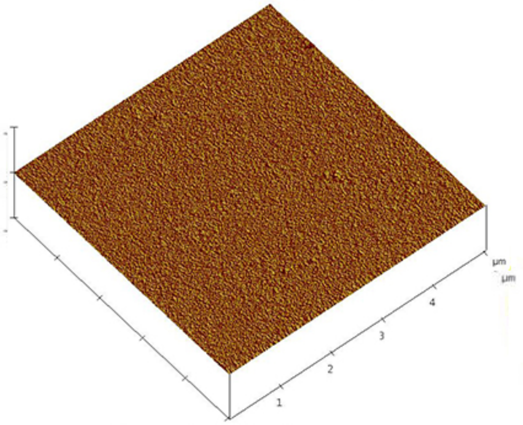




SRF Multilayer Structures Based on NbTiN

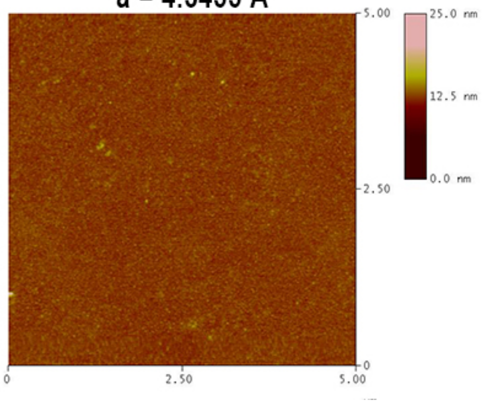
Influence of roughness & interlayer on T_c

NbTiN/AlN/MgO (100)

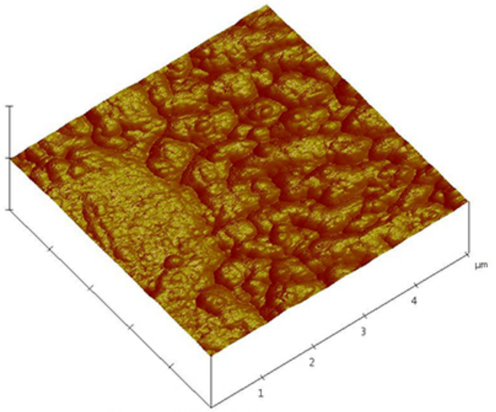


Rms=0.396 nm

a = 4.3455 Å

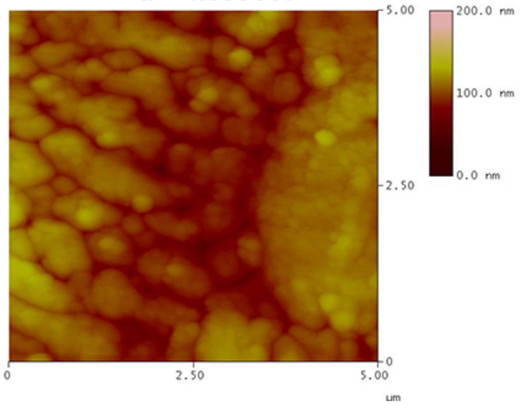


NbTiN/AlN/AN ceramic



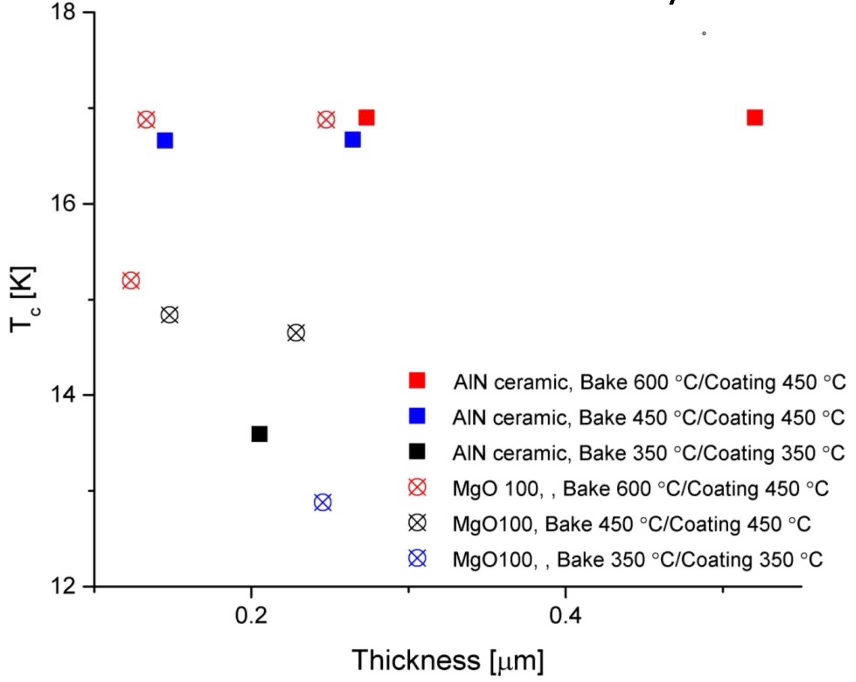
Rms=13.434 nm

a = 4.3584 Å



Quality of underlying AlN dictates
quality of the NbTiN film
Roughness of substrate not detrimental to Tc

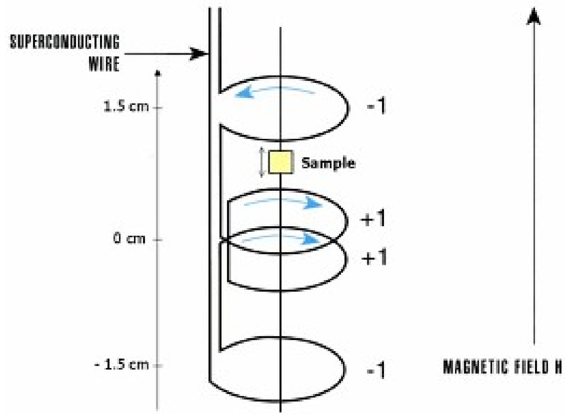
SI layers on MgO and AlN
coated simultaneously

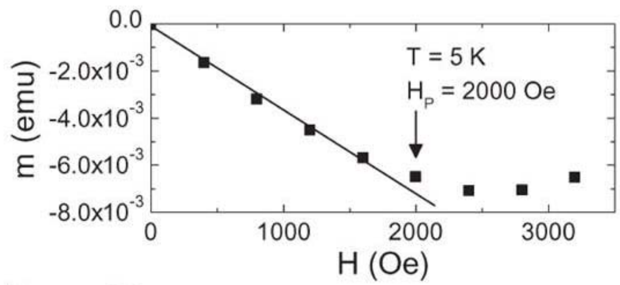


NbTiN Films (SI) – Flux Penetration Measurement

SQUID Magnetometry

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

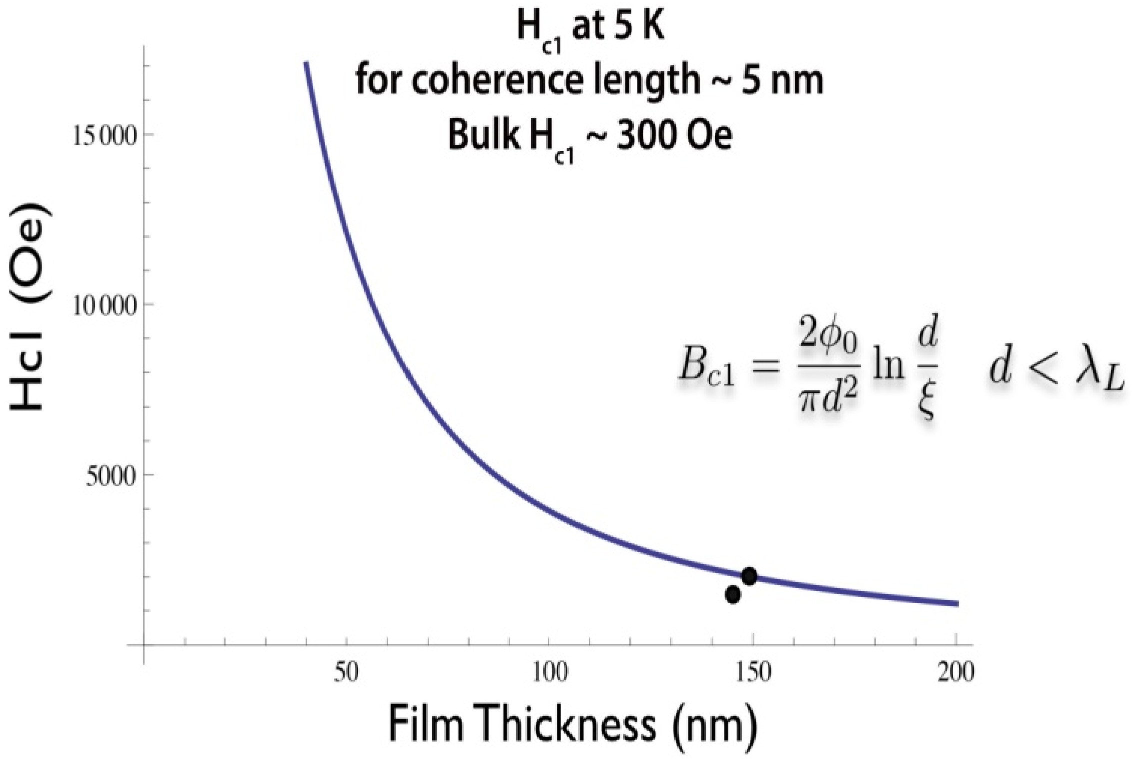




150 nm NbTiN/AlN films exhibit
H_{c1} enhancement
compared to
bulk-like NbTiN film

Thickness series study under progress

	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

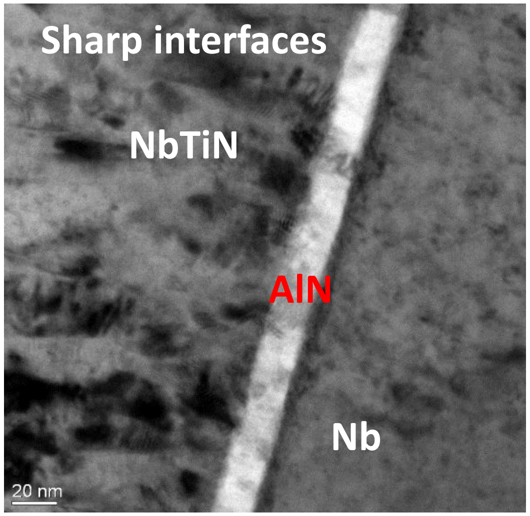


$$B_{c1} = \frac{2\phi_0}{\pi d^2} \ln \frac{d}{\xi} \qquad d < \lambda_L$$

R_s of NbTiN/AlN structures on Nb surfaces

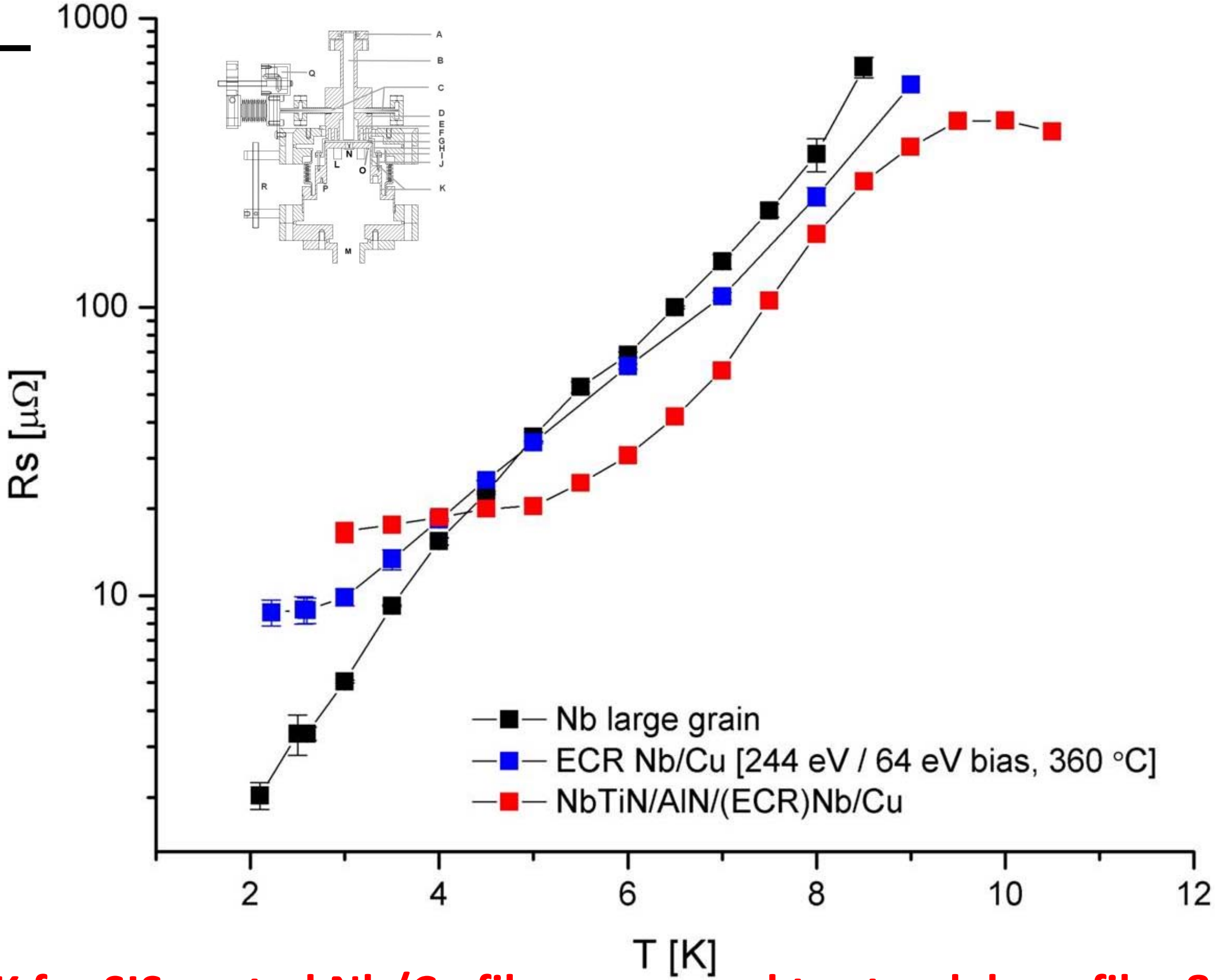
SIS structures coated on ECR Nb/Cu film: 24h-bake, coating and annealing for 4 h at 450°C.

	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

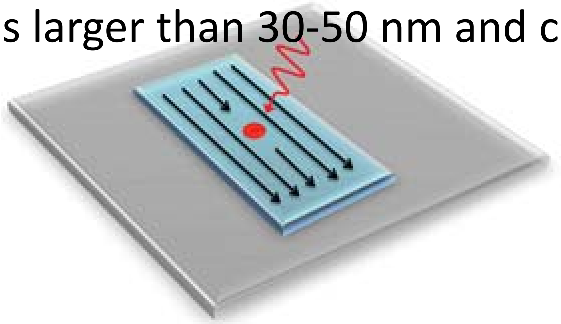
RF Measurement in 7.5 GHz sapphire-loaded TE_{011} cavity



Lower BCS resistance beyond 4 K for SIS coated Nb/Cu film compared to standalone film & bulk Nb. Similar effect observed for NbTiN/AlN/bulk Nb

SIS Multilayer Structures - Summary

- Good quality standalone NbTiN deposited by reactive DC magnetron sputtering.
  - Bulk, i.e. thicker than 1 micron, NbTiN films readily produced with a T_c of 17.3 K and H_{c1} of 30 mT.
  - Cubic δ-phase and T_c above 16 K for thicknesses larger than 30-50 nm and coating temperatures of 450 ˚C or higher.
  - 10 nm NbTiN layer with T_c ~ 12 K (SNSPD).



- AlN dielectric films with good dielectric properties.

- Good quality 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 .
  - If the dielectric can be grown as an adequate template, the substrate macro-roughness is not necessarily detrimental to the T_c of the superconducting film.

- **H_{c1} 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** (along with other experiments: cf Antoine C. –CEA, Lukaszew A. - W&M).

Conclusions

❑ Nb films deposited by energetic condensation (HiPIMS, ECR)

Significant improvement in film quality (crystallinity, impurity content, RRR, superconducting gap) leading to improved RF performance.

❑ Multilayer SIS for potential higher fields

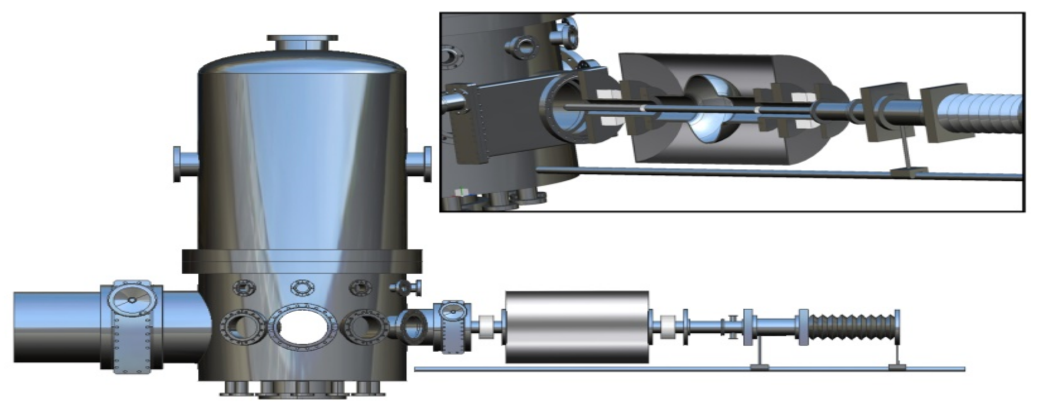
RF characterization of NbTiN/AlN coated on Nb surfaces reveal a promise of delaying flux penetration and lower RF losses for SIS coated Nb surfaces.

Further improvement of interfaces needed. HiPIMS may be useful for improving the structure and properties of both NbTiN and AlN and lower the deposition temperature.

❑ NbTiN & other material layers for SNSPD

Development of adequate NbTiN based structures

❑ Coating systems available or under development



Concept for SIS structure coating on Nb & Nb/Cu cavities