Atomic Layer Deposition of Thin Superconducting Films and Multilayers: Coupons and Cavity Tests

Thomas Proslier

This work was supported by the U.S. Department of Energy, Office of Science under contract No. DE-AC02-06CH11357 and by the American Recovery and Reinvestment Act (ARRA) through the US Department of Energy, Office of High Energy Physics Department of Science.

Team:

Jeffrey Klug Fermi Scholar, ANL -> ALD/Characterization
Nick Groll Postdoc, ANL -> Point Contact
Chaoyue Cao Student, IIT -> Point Contact, Raman
Nick Becker Student, IIT -> ALD/Characterization
Mike Pellin Distinguished Fellow, ANL -> Boss

Collaborations:

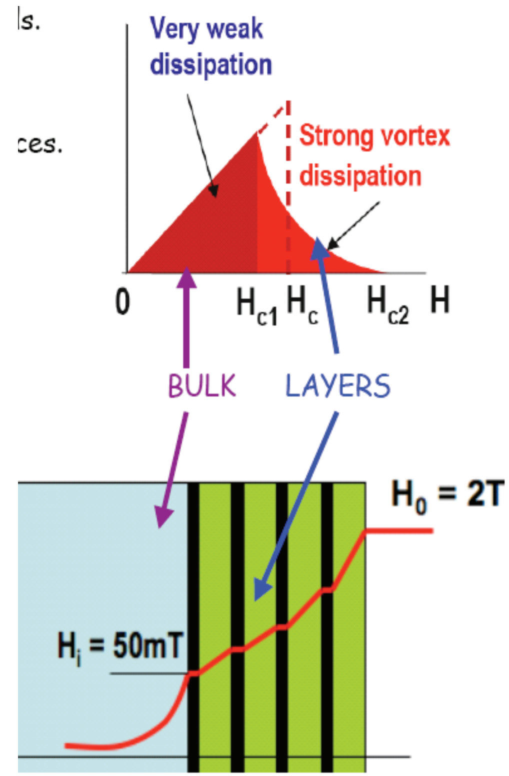
J. Zasadzinski, C. Segre, IIT
G. Ciovati, P. Kneisel, C. Reece, A.M. Valente JLAB
M. Khrishan, Irfan, Alameda
A. Romanenko, A. Grasselino, FNAL
T. Tajima, LosAlamos
A. Anders, LBNL
C. Antoine, CEA
V. Vinokur, ANL
T. Baturina, A. Mironov, ISP/UofC
A. Nassiri, R. Kustom, APS, ANL
M. Kharitonov, Rudgers
A. Glatz, I. Aronson, A. Snezhko, ANL
R. McDermott, U. of Madison

Decrease Cost I: Multilayers

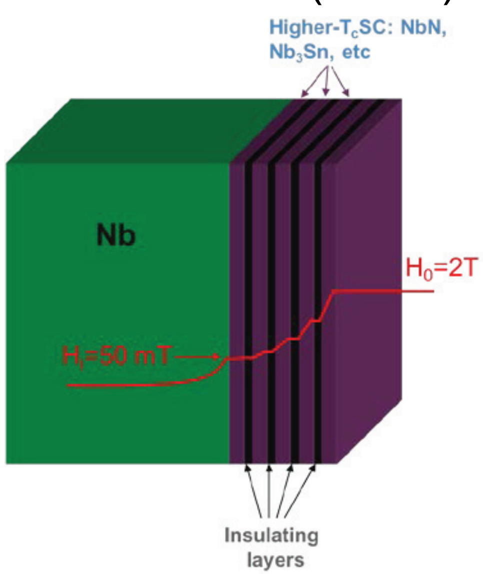
Fields in bulk Nb cavities approaching dc depairing limit for Nb, $H_c(0) \approx 200$ mT

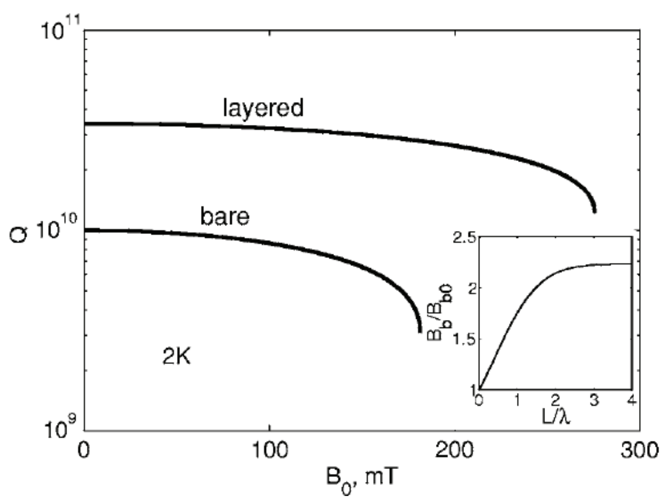
Superconductor-Insulator multilayer [Gurevich, *Appl. Phys. Lett.* **88**, 012511 (2006)]

- Increase performance
  - Move beyond limits of Nb
- Decrease cost (early career award 2011)
  - Higher operating temperature (reduce cryogen costs)
  - Replace bulk Nb with cheaper material (Cu/Al)



A. Gurevich, APL 88, 012511





$$B_{c1} = \frac{2\phi_0}{\pi d^2}\ln\frac{d}{\tilde{\xi}}, \quad d < \lambda,$$

- Coat inside Nb SRF cavity with precise, layered structure → ALD

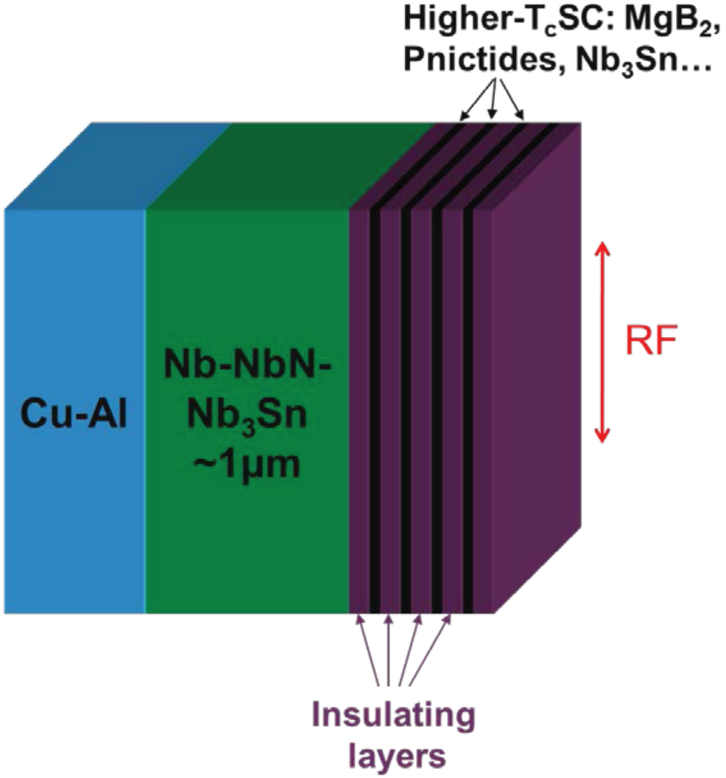
Decrease Cost II: Nb films on Cu

SRF cavities Goals E_{max} ~ 70 MV/m and Q > 10^{10} at T > 4.2 K

- All thin films
-> reduce surface preparation.

- High Tc materials:
Nb_3Sn, MgB_2, Pnictides, $BaKBiO_3$ etc…

- But:
Higher Tc -> Lower Hc1 -> Lower Gradient



Collaboration: Synergy of expertise

PHYS-APS
Processing/Testing

M. Kelly, R. Murphy

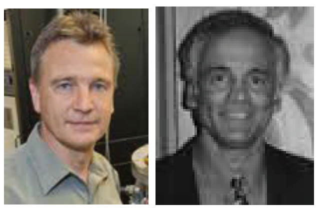
HEP
support-funding

Th. Proslier

MSD
ALD-PCT-characterization

J. Klug, N. Groll

Collab
Berkeley/Alameda
HIPIMS



A. Anders, M. Krishnan

IIT
Expertise/Raman

J. Zasadzinski
N. Becker, C. Cao

Collab
JLAB
test coupons/cavities
ECR

A-M. Valente, P. Kneisel, G. Ciovati

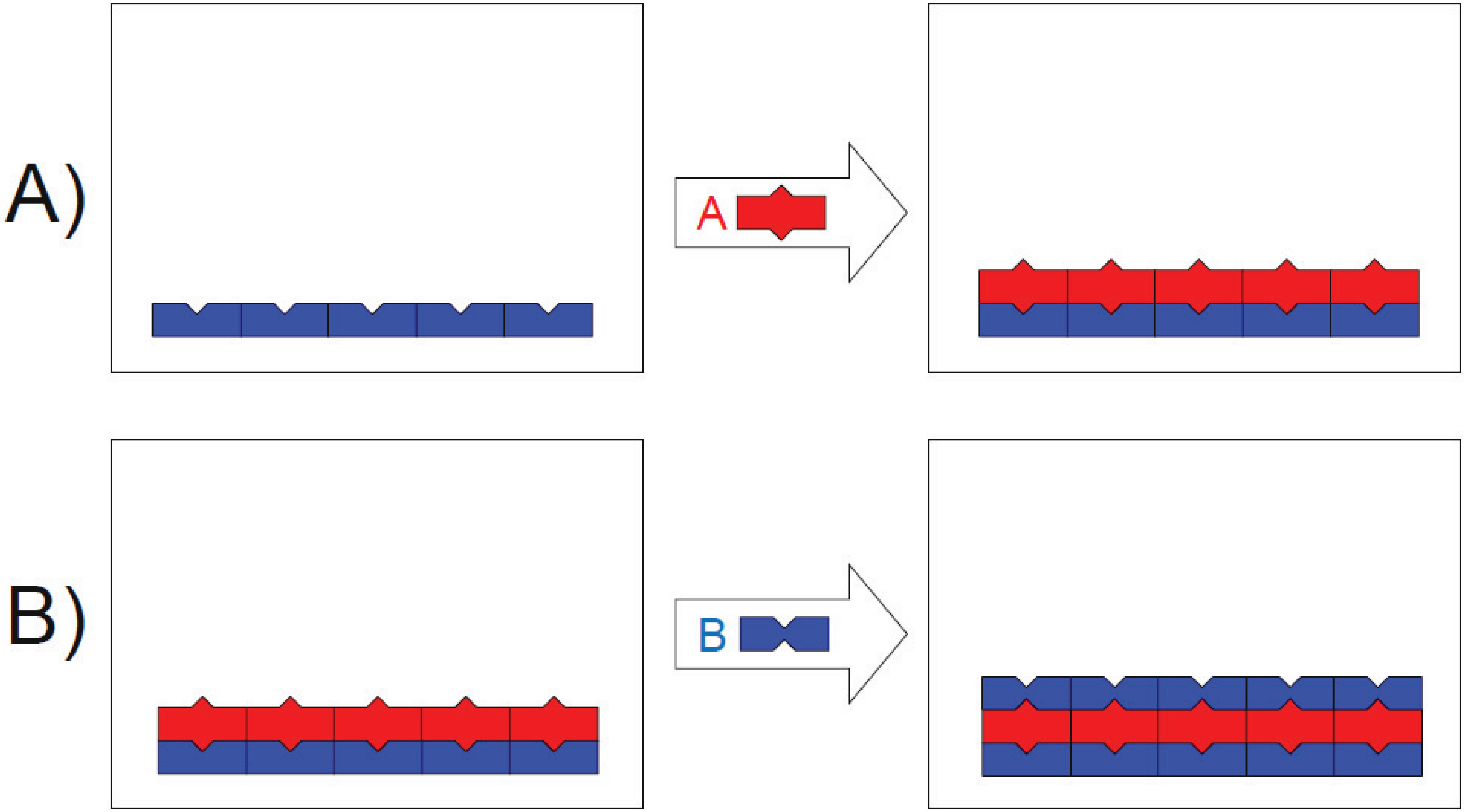
Collab
FNAL
Processing/testing

A. Grasselino, A. Romanenko,
L. Cooley, A. Rowe, B. Stone,
V. Yakovlev, C. Ginsburg…

- Multilayers:
  Atomic Layer Deposition of superconductors.
  Coupons & SRF cavities.

- Niobium on Copper
  Coupons and RF test.

Atomic Layer Deposition



Sequential pulsing of gas phase precursors (A and B) …(A, B, C…)

Superconductors by ALD

Goal for SRF is a material with a T_c higher than bulk Nb (9.2 K)

- Niobium Carbide: NbC 1.7 K
  - $NbF_5 + Al(CH_3)_3$
  - $NbCl_5 + Al(CH_3)_3$
- Niobium Carbo-Nitride: $NbC_{1-x}N_x$ 3.8 K
  - $Al(CH_3)_3 + NbF_5 + NH_3$
  - $Al(CH_3)_3 + NbCl_5 + NH_3$
- Niobium Silicide: NbSi 3.1 K
  - $NbF_5 + Si_2H_6$
  - $NbF_5 + SiH_4$
- Titanium Nitride: TiN 3.8 K
  - $TiCl_4 + NH_3$
- Molybdenum Nitride: MoN 11.8 K
  - $MoCl_5 + NH_3$
  - $MoCl_5 + Zn + NH_3$
- Niobium Titanium Nitride: $Nb_{1-x}Ti_xN$ 14 K
  - $(NbF_5, TiCl_4) + NH_3$
  - $(NbCl_5, TiCl_4) + Zn + NH_3$

J. Klug et al. J. Phys.Chem C 115, 25063

Th. Proslier et al. J. Phys.Chem C 115, 9477

Th. Proslier et al. Accepted to PRB

M. Mironov et al. submitted to Nature Materials
N. Groll et al. Accepted to APL.

ALD NbN with T_c = 10 K reported
Hiltunen *et al.*, *Thin Solid Films* **166**, 149 (1988)

Superconductors by ALD

Goal for SRF is a material with a T_c higher than bulk Nb (9.2 K)

- Niobium Carbide: NbC 1.7 K
  - $NbF_5 + Al(CH_3)_3$
  - $NbCl_5 + Al(CH_3)_3$
- Niobium Carbo-Nitride: $NbC_{1-x}N_x$ 3.8 K
  - $Al(CH_3)_3 + NbF_5 + NH_3$
  - $Al(CH_3)_3 + NbCl_5 + NH_3$
- Niobium Silicide: NbSi 3.1 K
  - $NbF_5 + Si_2H_6$
  - $NbF_5 + SiH_4$
- Titanium Nitride: TiN 3.8 K
  - $TiCl_4 + NH_3$
- Molybdenum Nitride: MoN 11.8 K
  - $MoCl_5 + NH_3$
  - $MoCl_5 + Zn + NH_3$
- Niobium Titanium Nitride: $Nb_{1-x}Ti_xN$ 14 K
  - $(NbF_5, TiCl_4) + NH_3$
  - $(NbCl_5, TiCl_4) + Zn + NH_3$

Growth characterization – NbSi

In situ monitoring:

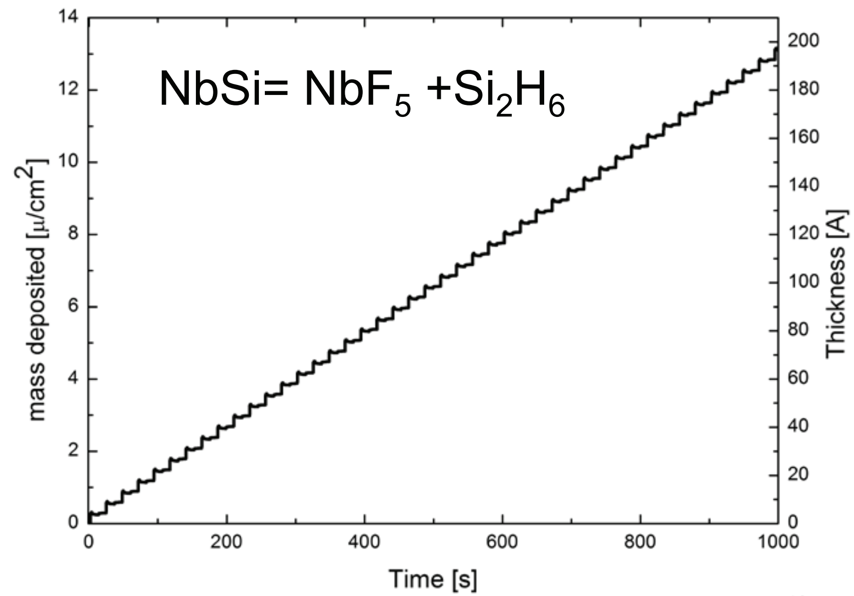
- Quartz crystal Microbalance (QCM)

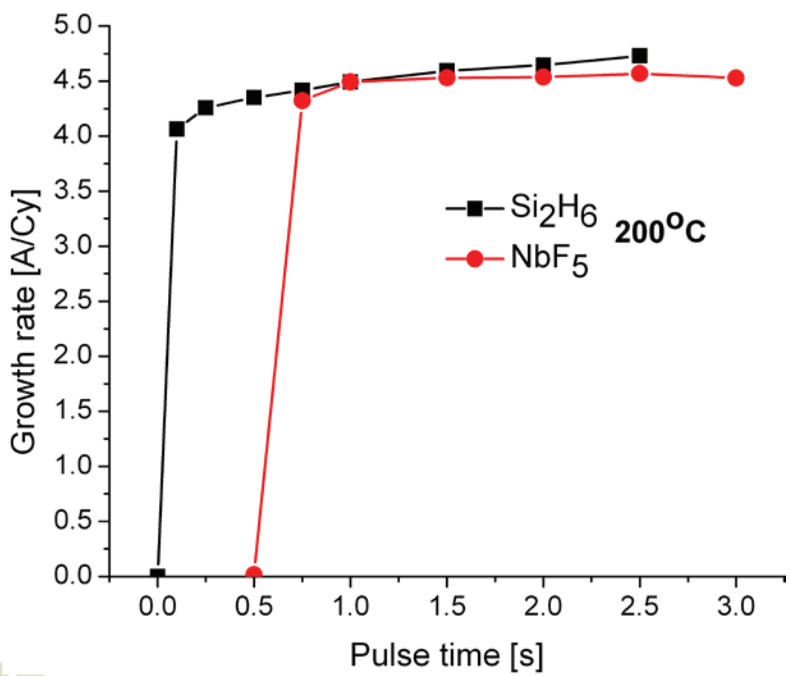
<300 ^{0}C: Mass gain (ng/cm^2)

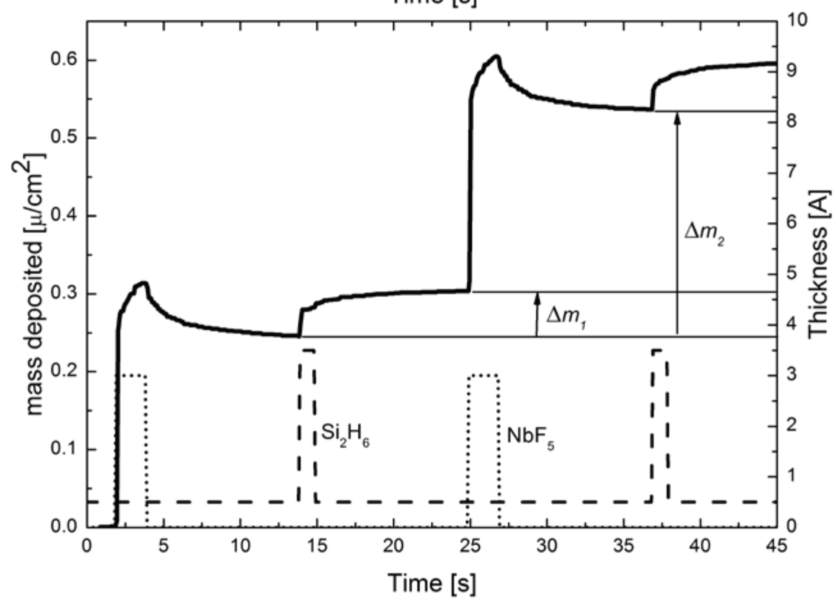
- Residual Gas Analyzer (RGA):

reaction products

-> Growth mechanisms



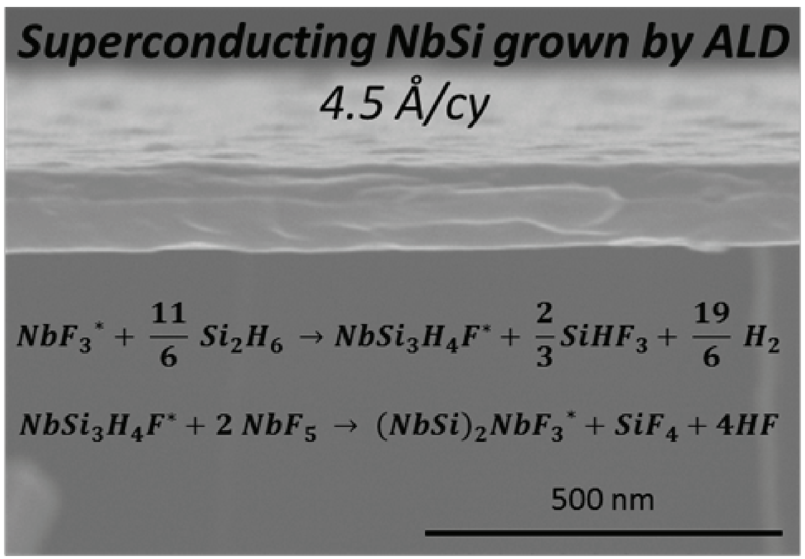


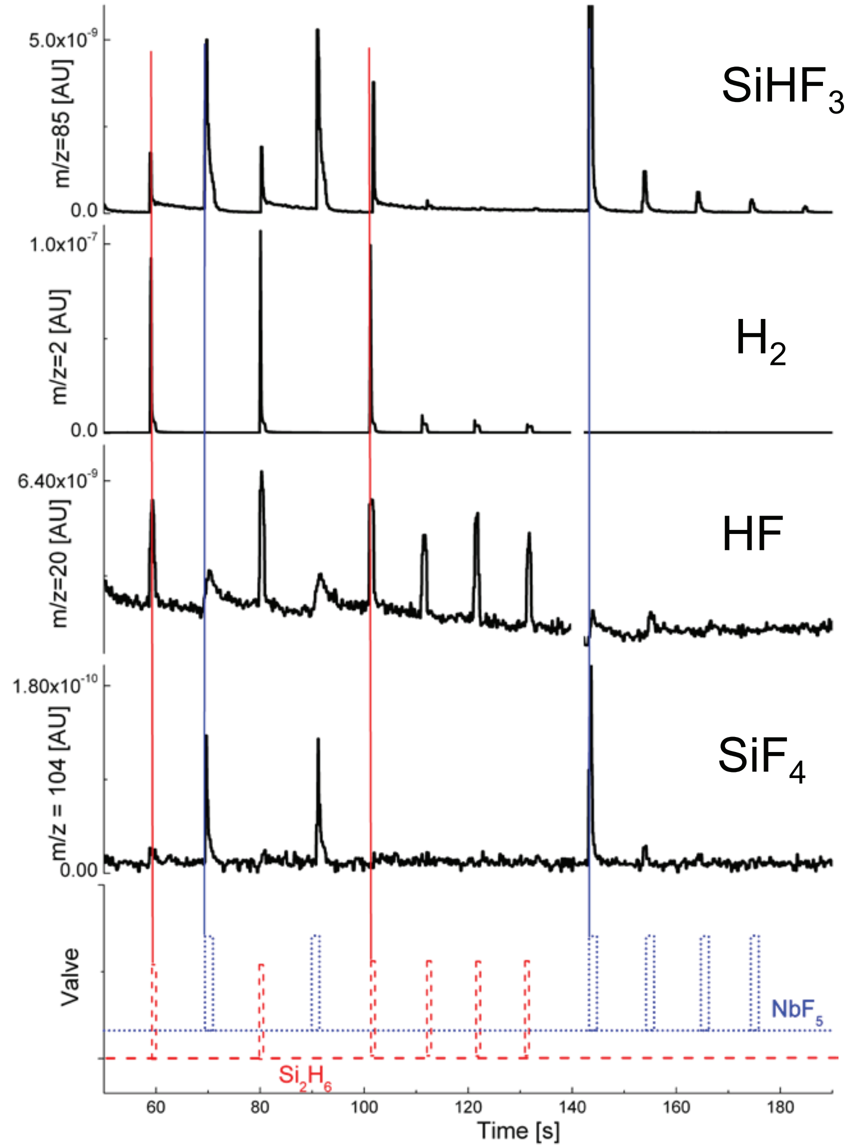


Growth characterization – NbSi

In situ monitoring:

- Quartz crystal Microbalance (QCM)
<300 ^{0}C: Mass gain (ng/cm^2)
- Residual Gas Analyzer (RGA):
reaction products
-> Growth mechanisms





Niobium silicide: NbSi

X-ray reflectivity (XRR)

- Constant GR between 150-275°C
  - CVD above 275°C
- Low roughness, increases with T
  - Amorphous-crystalline transition?
  - No peaks in XRD
- Density constant up to 350°C

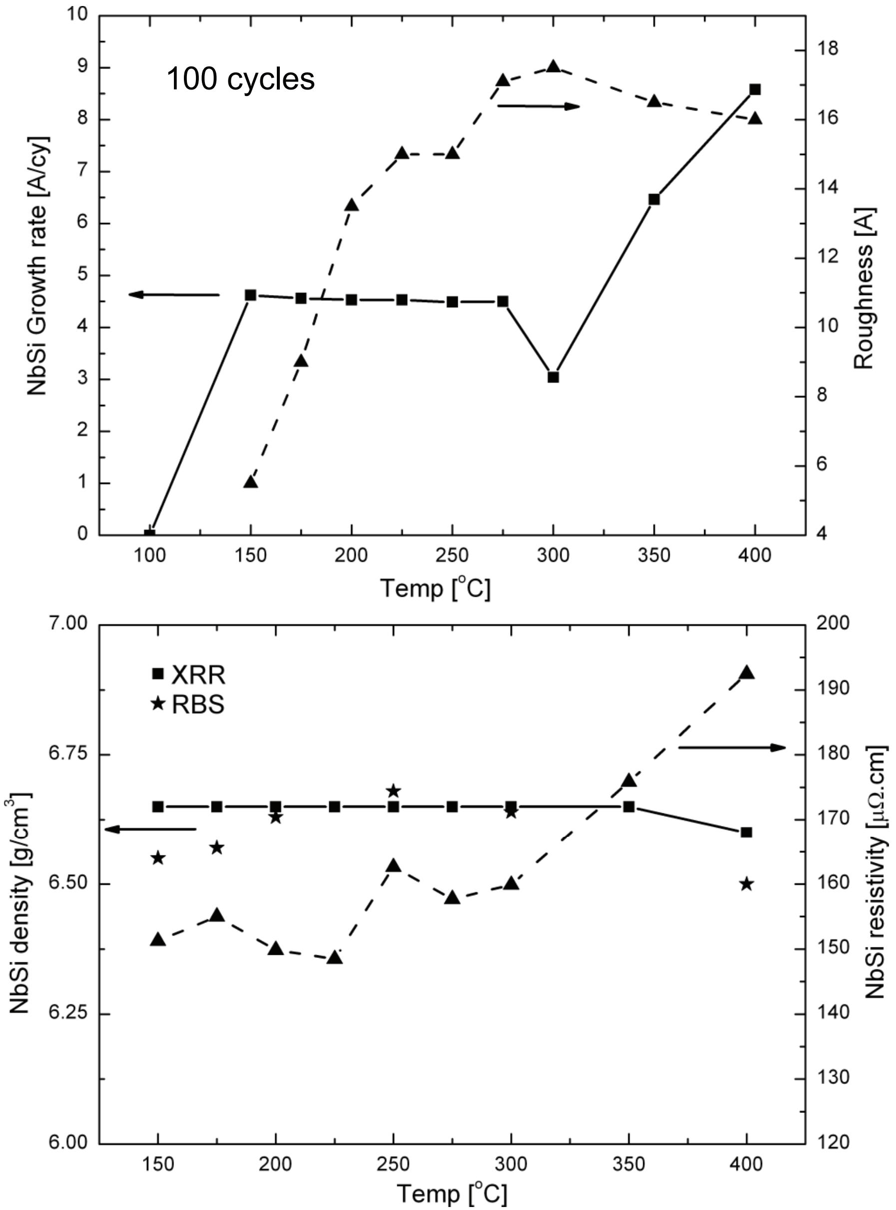
Composition

- No fluorine detected
- RBS: Nb:Si ratio 1:1
- XPS: Nb, Si chemically bound

Resistivity

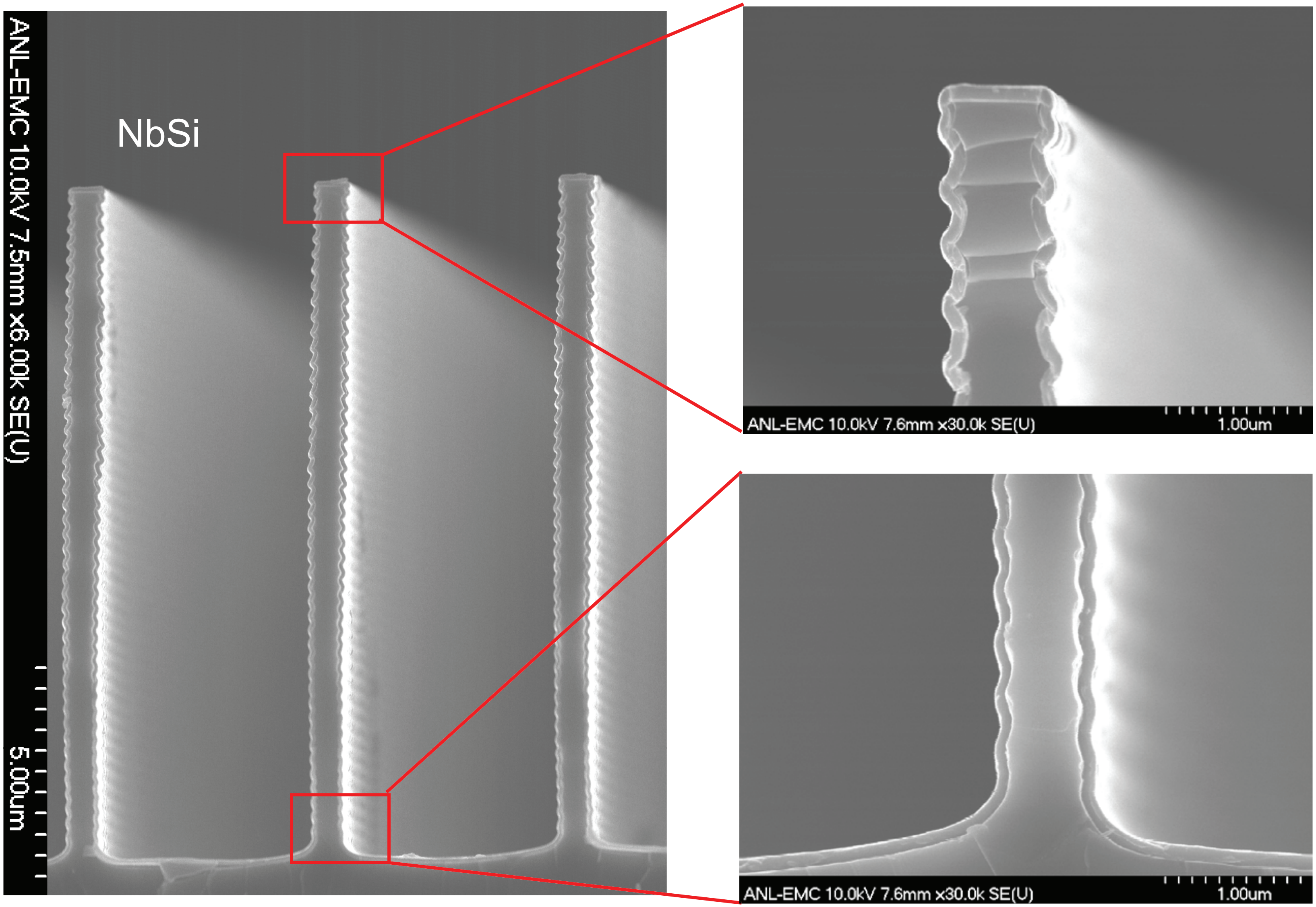
- 145-165 μΩ-cm in ALD regime

Do not grow on oxides!



Proslier, *et al.*, *J. Phys. Chem. C* **115**, 9477 (2011)

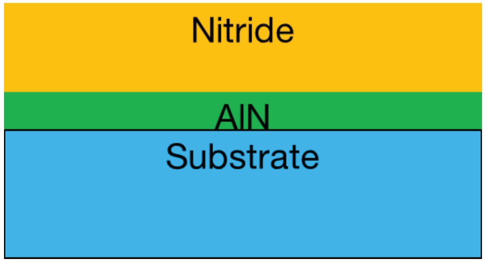
NbSi



ANL-EMC 10.0kV 7.5mm x6.00k SE(U)
NbSi
5.00um
ANL-EMC 10.0kV 7.6mm x30.0k SE(U)
1.00um
ANL-EMC 10.0kV 7.6mm x30.0k SE(U)
1.00um

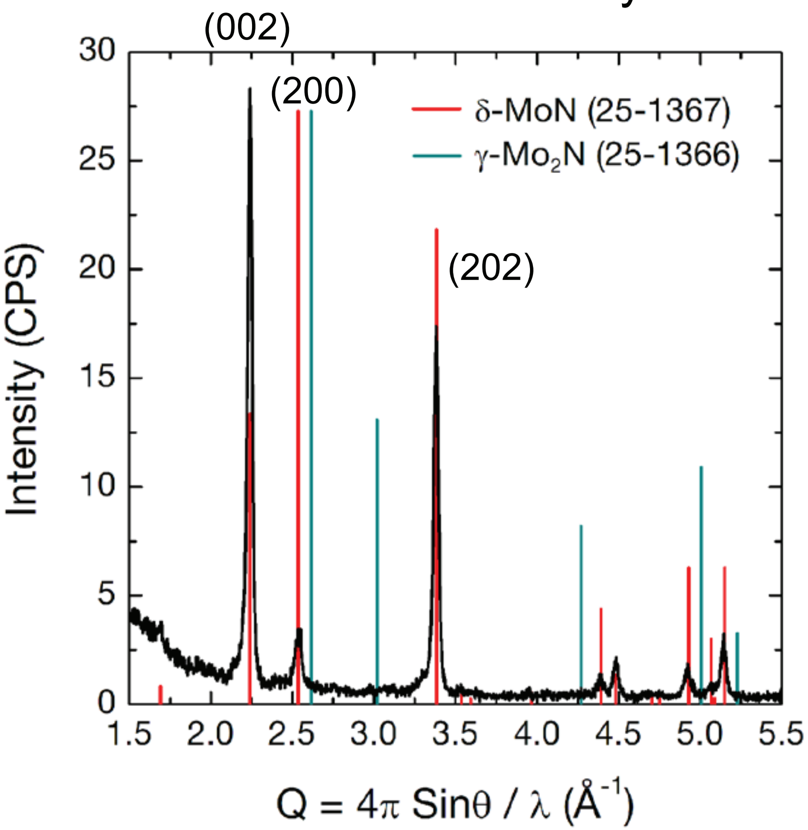
Molybdenum Nitride (MoN)

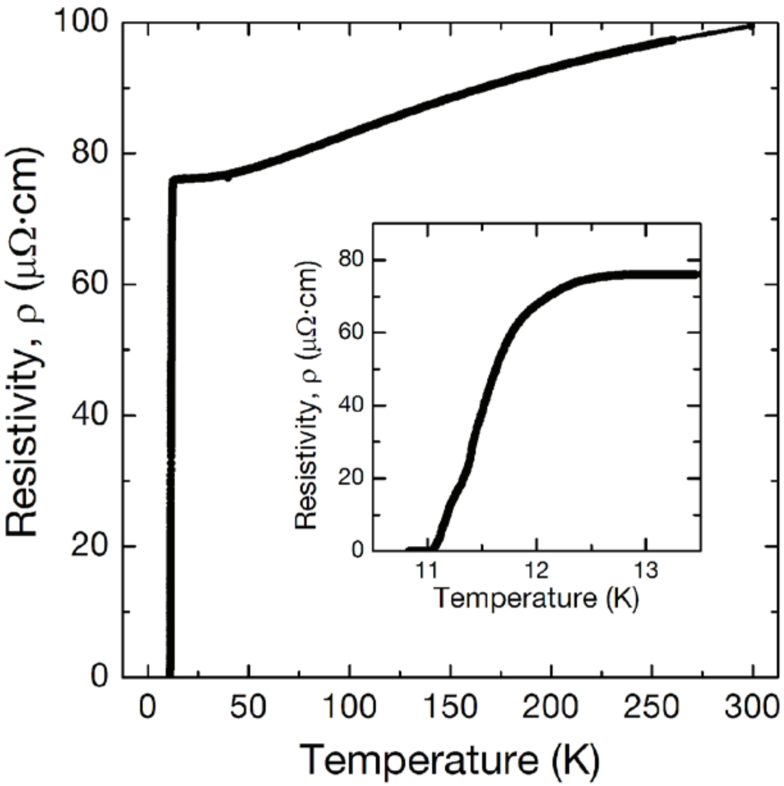
Growth temperature: 450 ^{0}C
$MoCl_5 + NH_3$



AlN : $AlCl_3 + NH_3$

1600 cy MoN/ 200Cy AlN/ Quartz

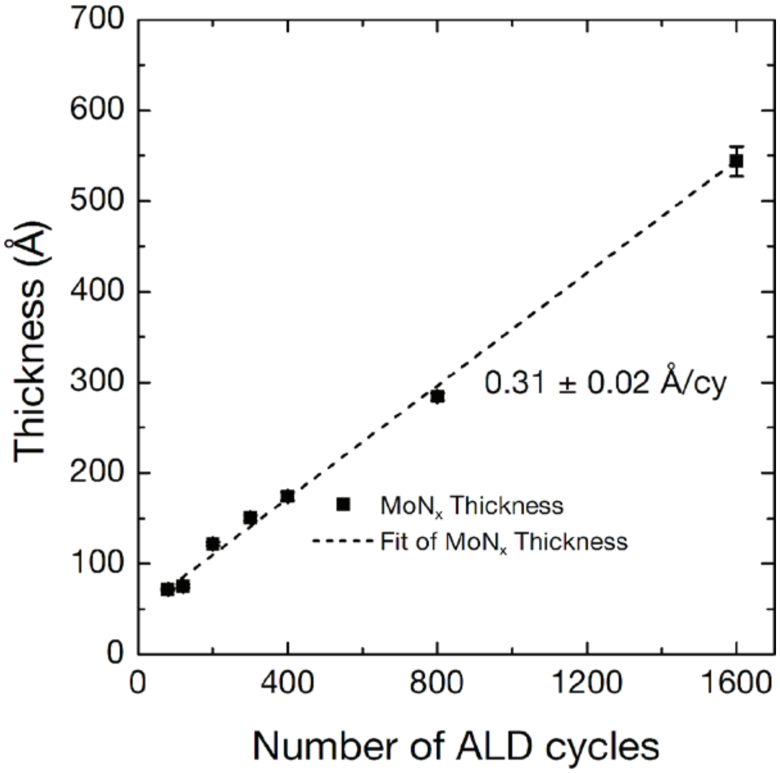


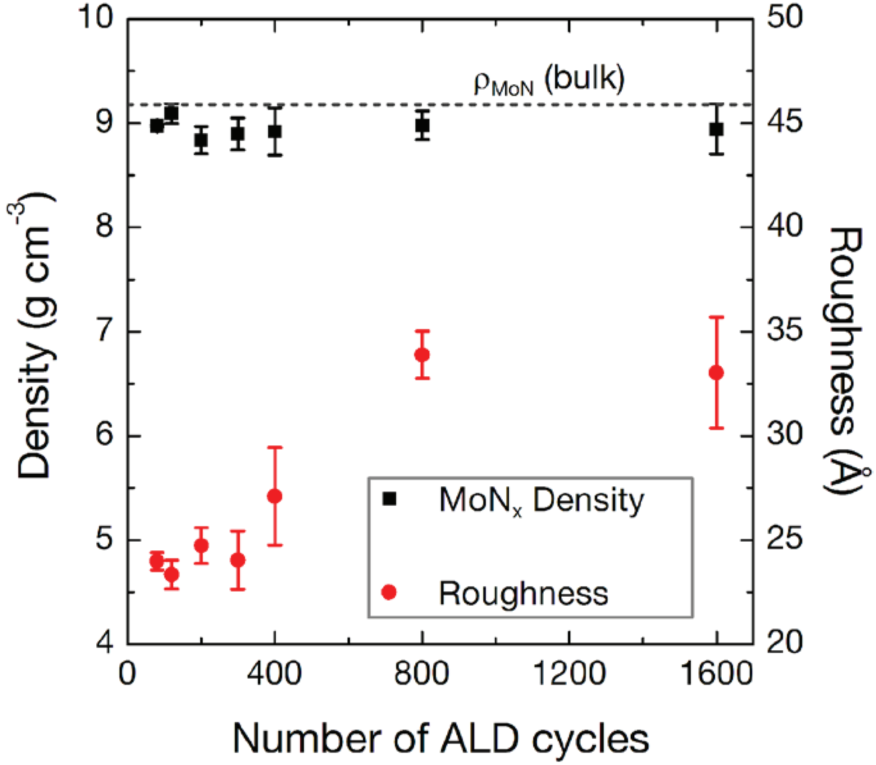


Preferred orientation: (002) / T_C = 11.5 K

XRR: Thickness study

450 ^{0}C

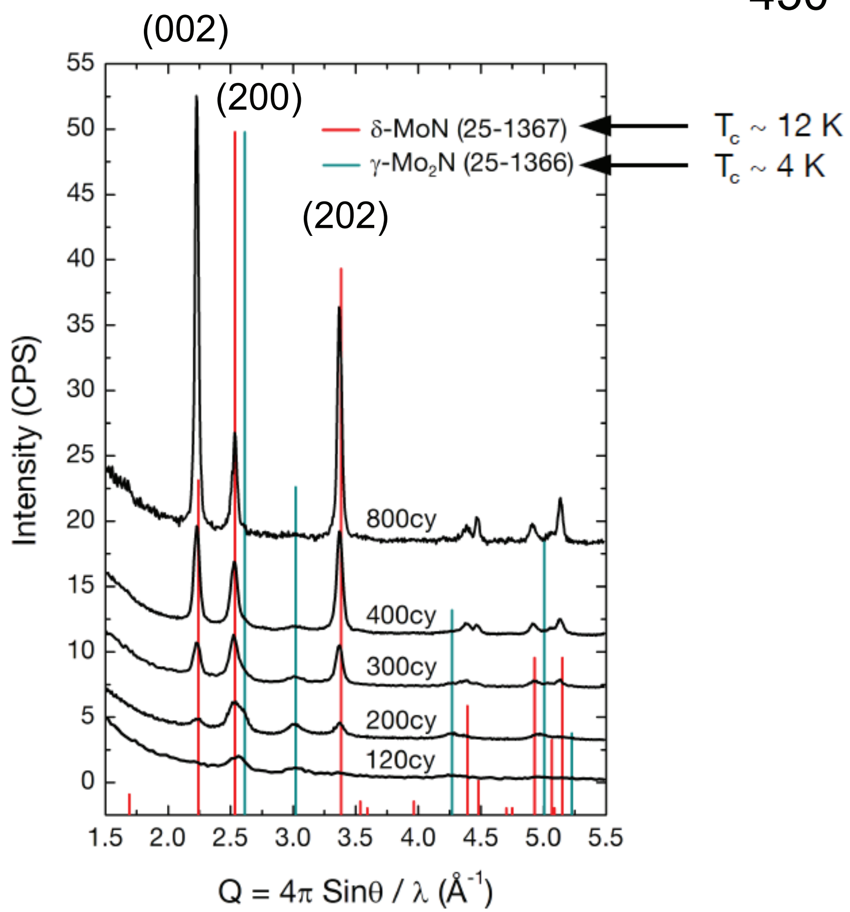


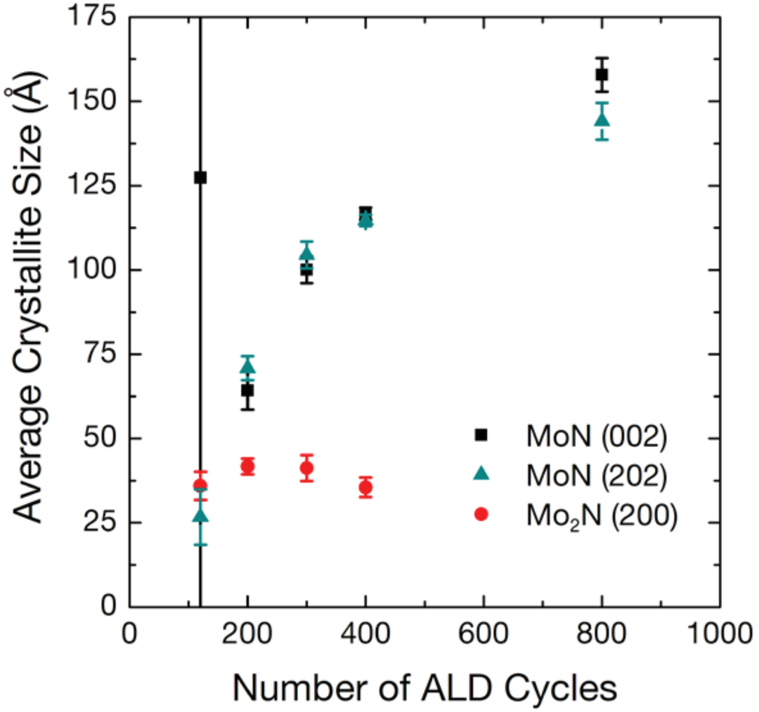


Linear, well controlled growth with fine thickness control
Bulk-like density, roughness ~ 25-35 Å

GIXRD: Evolution of structure with thickness

450 ^{0}C





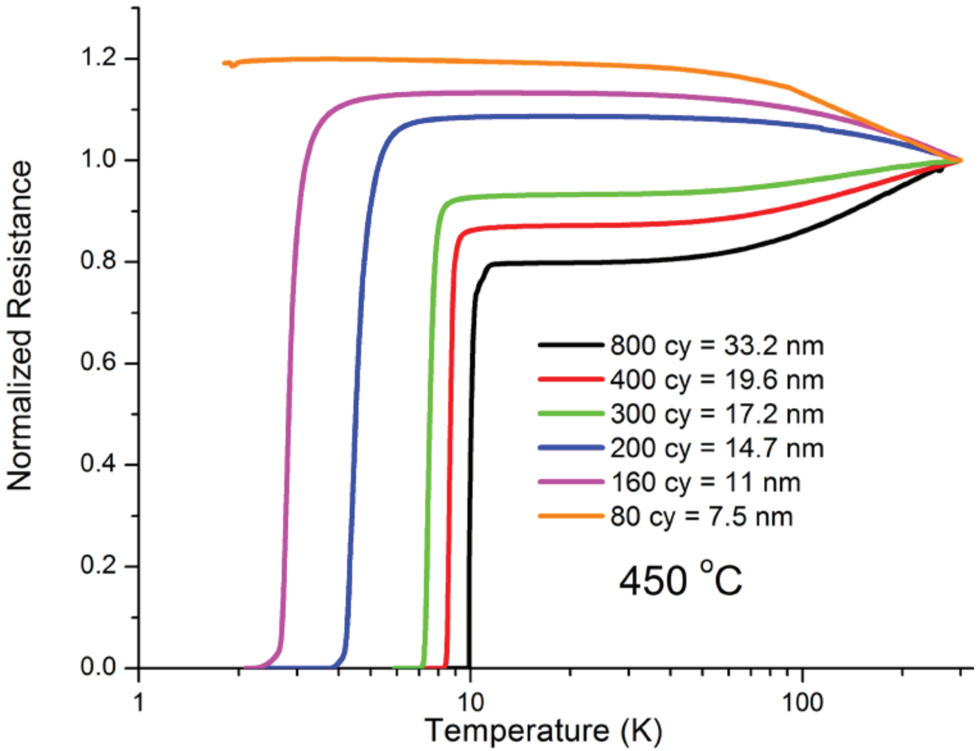
Mo_2N as a nucleation confirmed by XPS

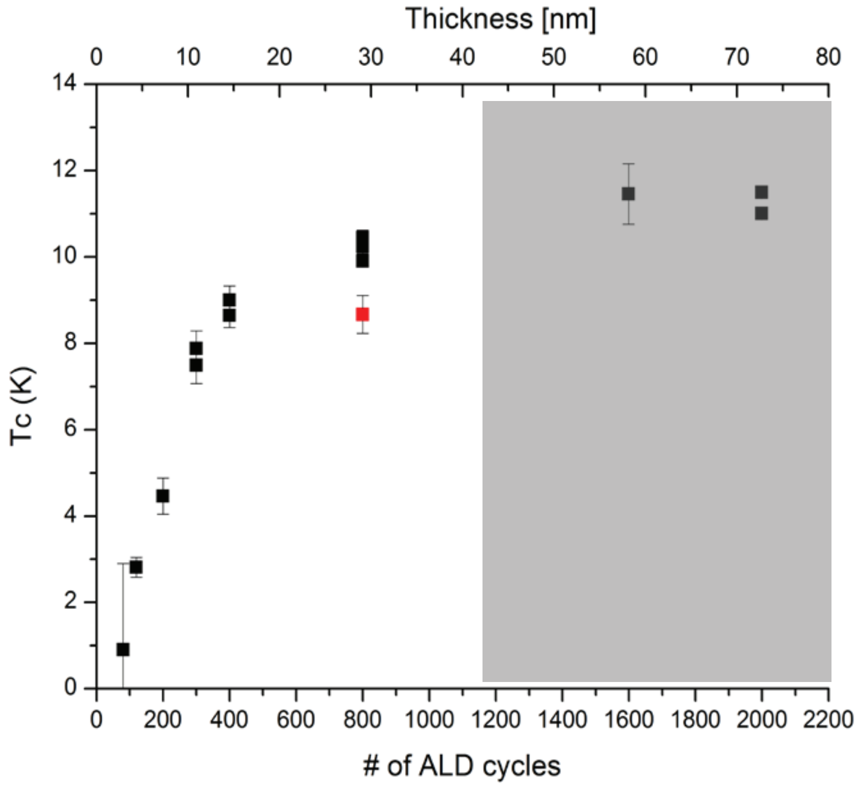
MoN phase grain size increase with thickness
Mo_2N does not

Confirmation of Mo rich nucleation

Transport: Evolution with thickness

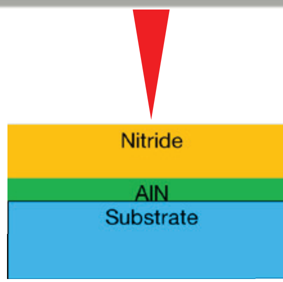
450 ^{0}C / H=0T

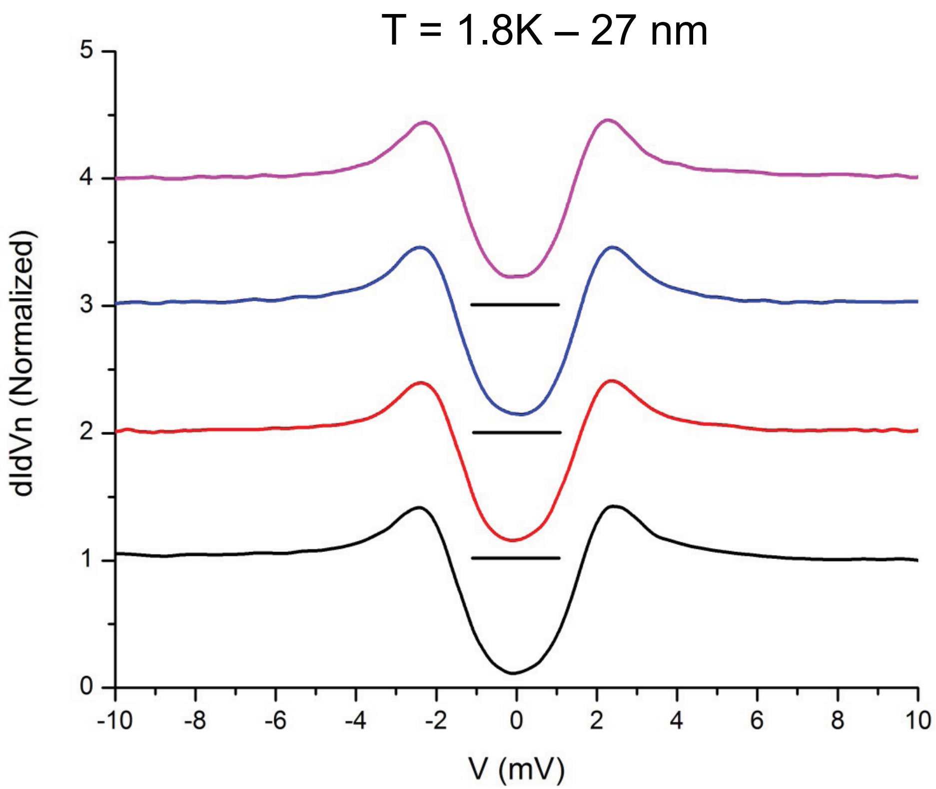


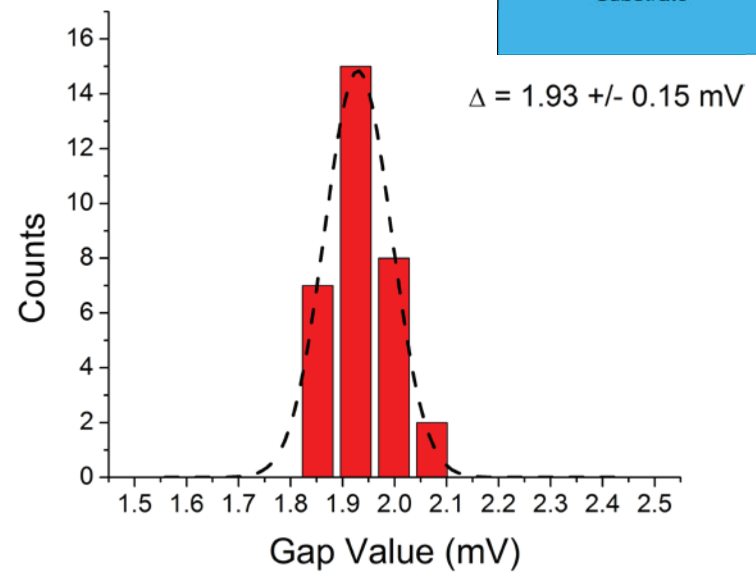


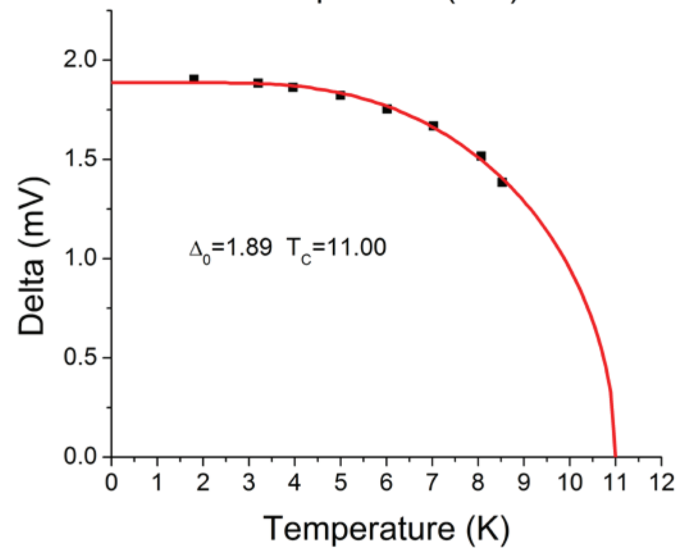
Resistivity bulk ~ 100 μΩ.cm
Tc bulk ~ 11.5 K
d>30 nm

Tunneling spectroscopy



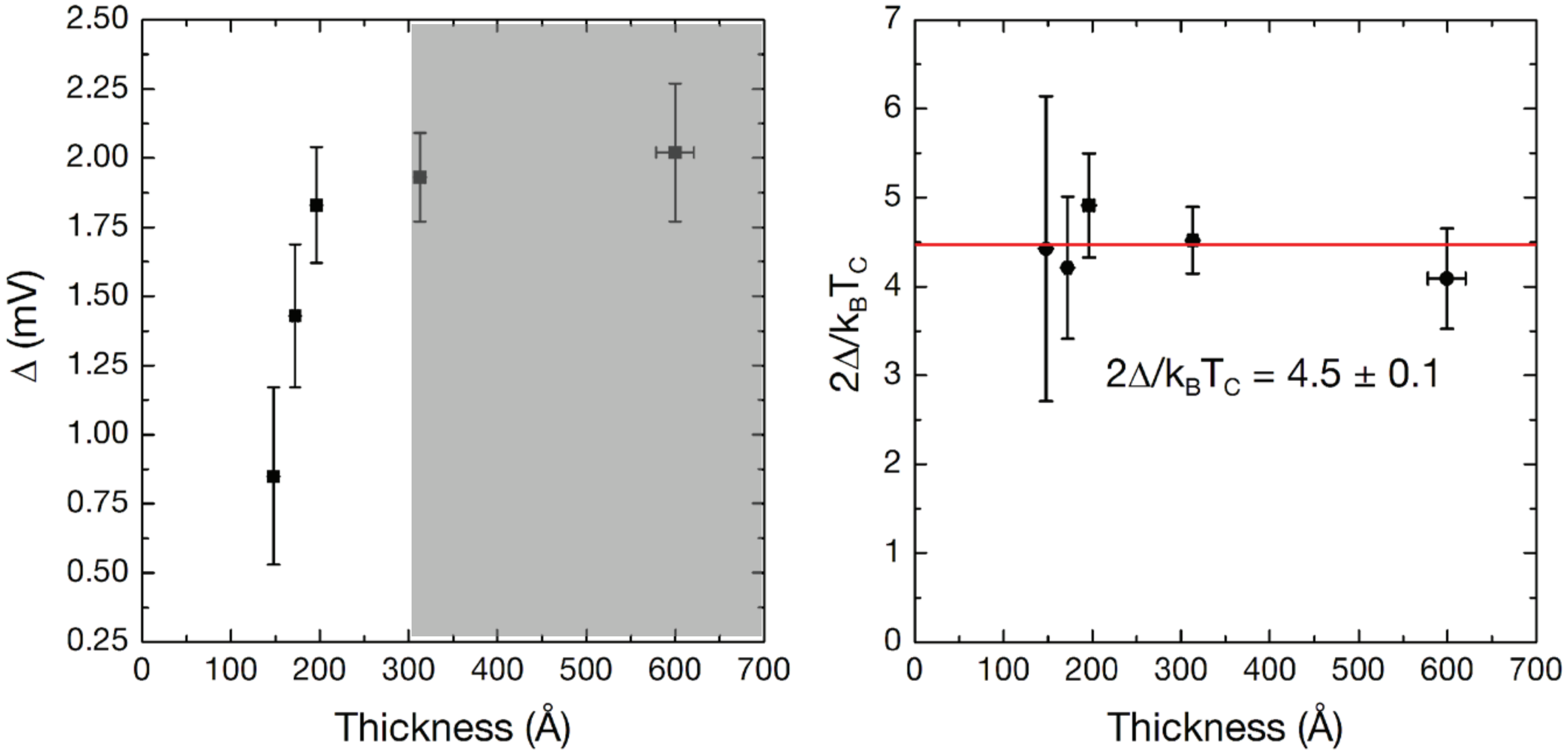






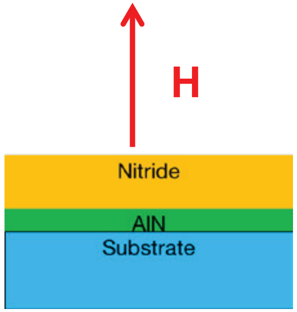
Tc tunneling match Transport
Homogeneous gap distribution

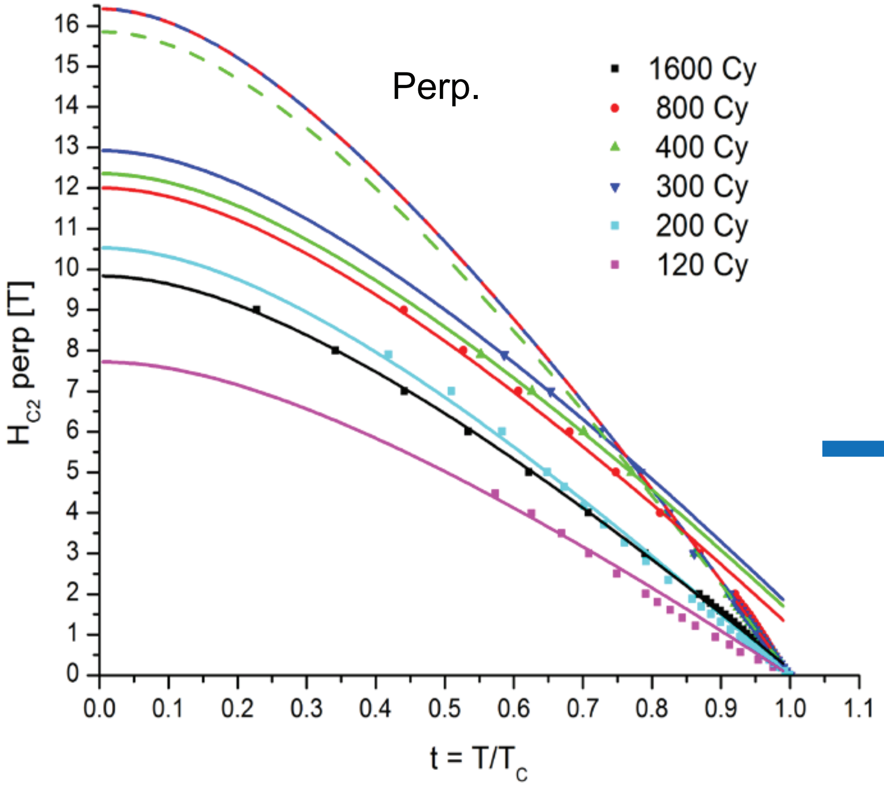
MoN: Δ, Tc vs thickness

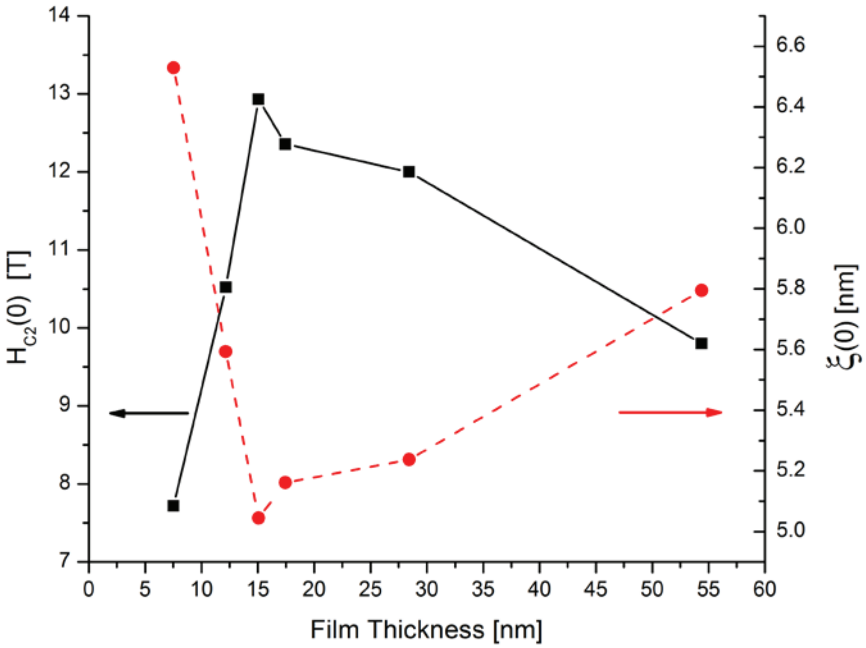


Bulk Limit for T_C and Δ > 30 nm
$2\Delta/k_BT_C$ = 4.5, strong coupling.

Magnetic field dependence: Transport

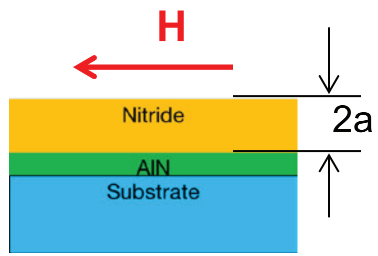


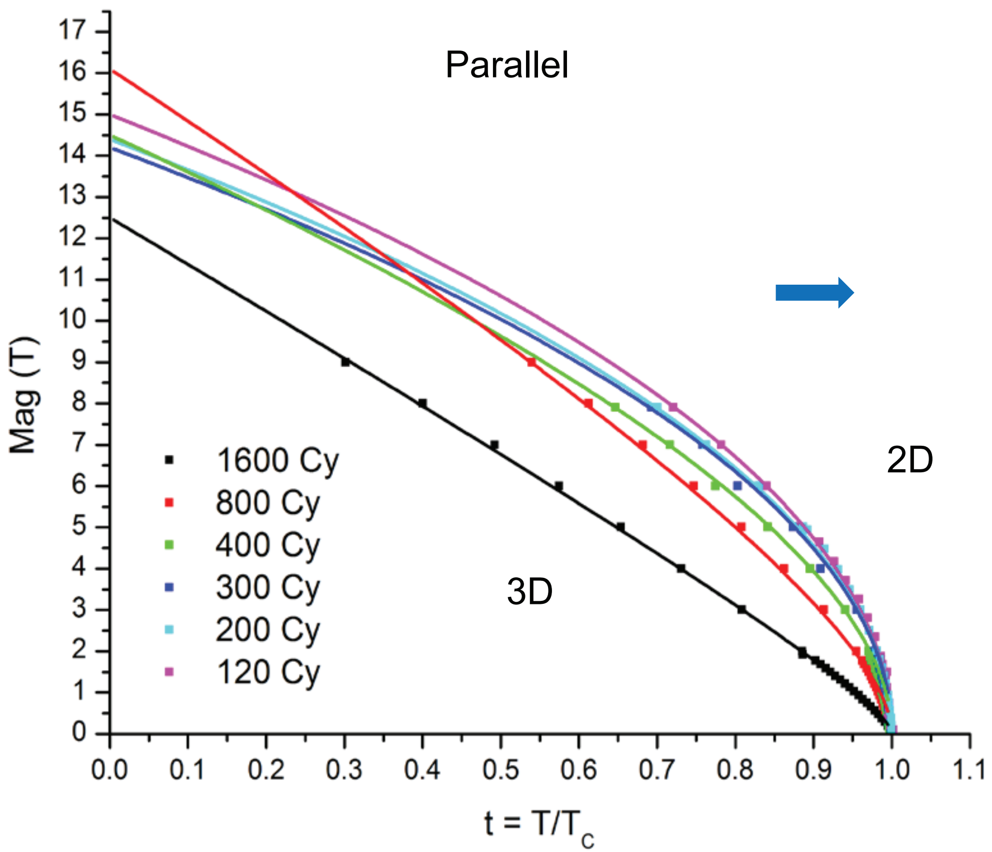


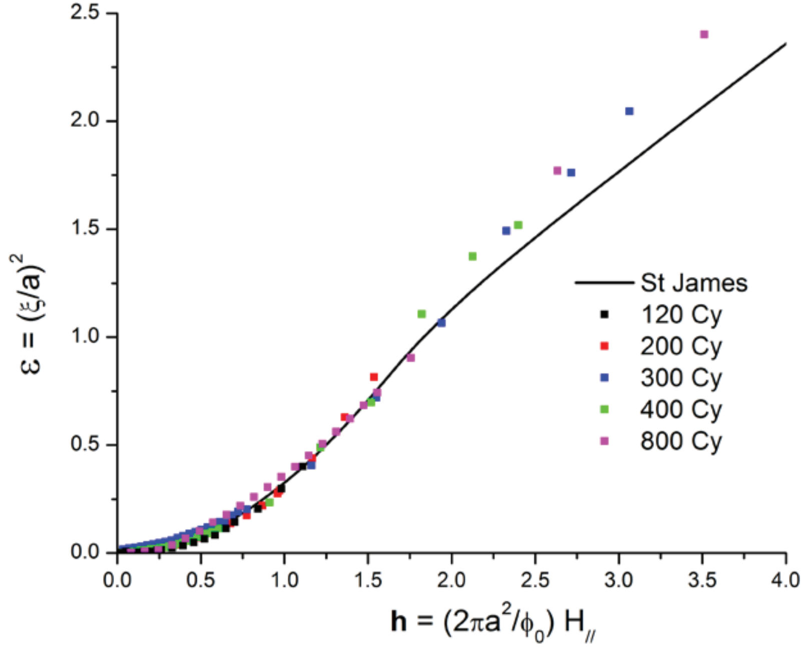


Coherence length, ξ_{GL} ~ 5.5 nm

Magnetic field dependence: Transport



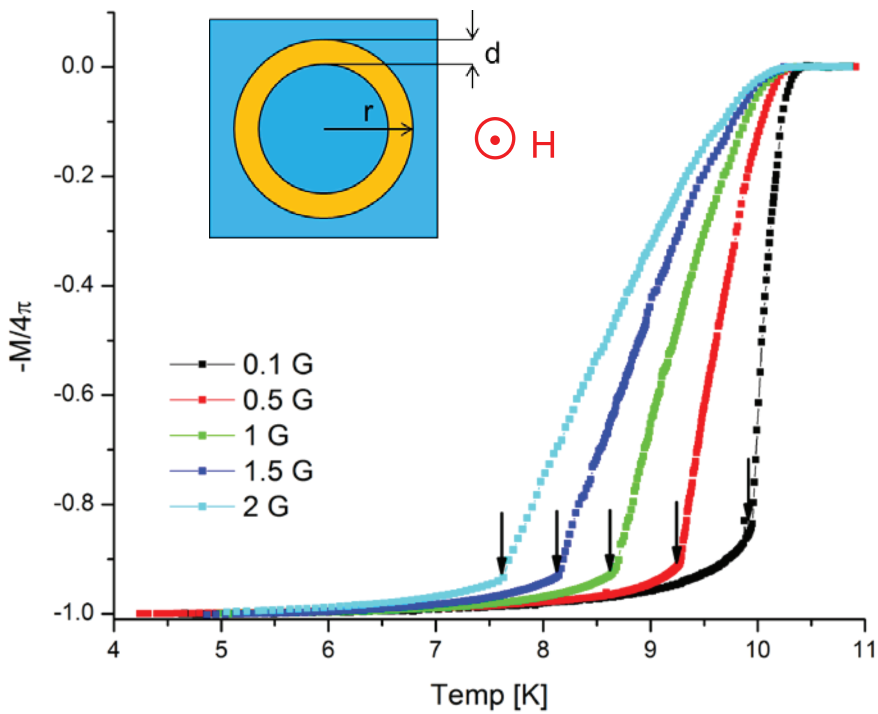


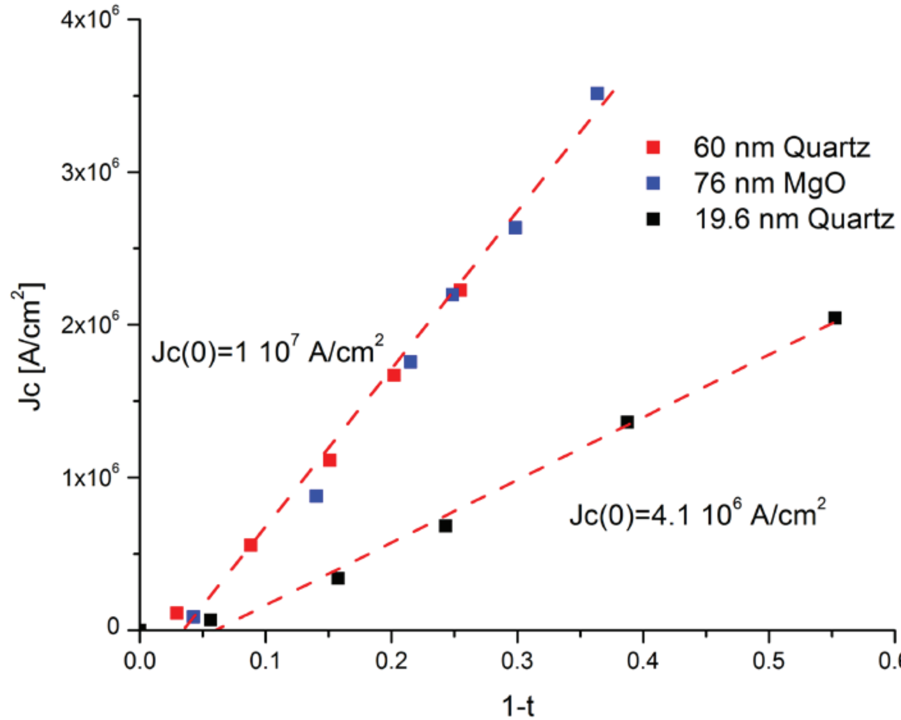


Thickness control -> 3D to 2D transition

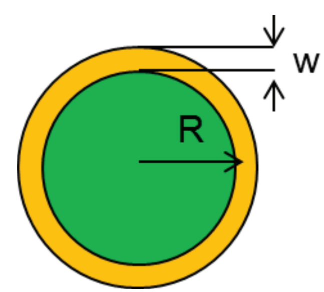
$\lambda_{MoN} \geq 150$ nm

Critical current





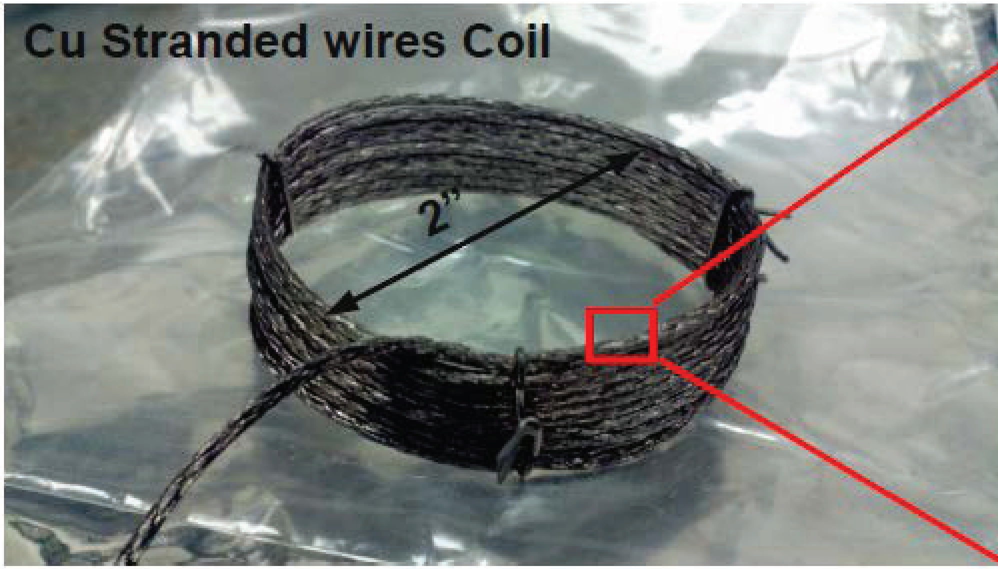
$$j_c = H \frac{\pi r}{Ln(8r/d - 0.5)} / w.d$$

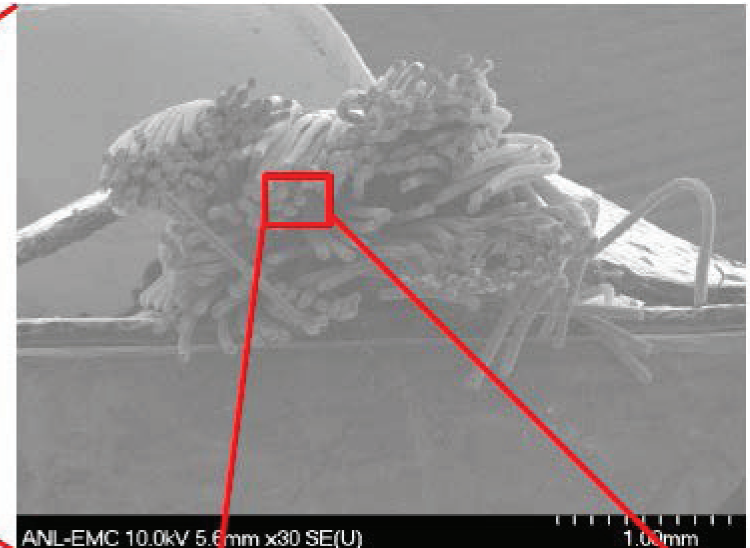


50 μm Cu wire / 60 nm MoN @ 2K -> I ~ 1 Amp

Scalability by ALD (magnets?)

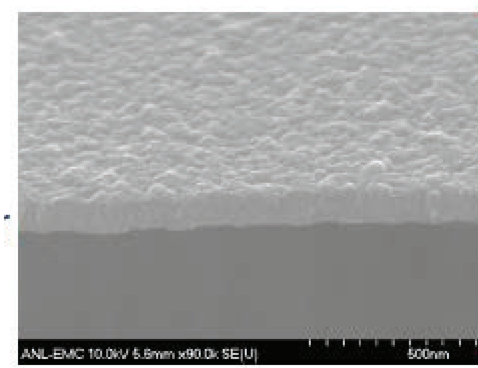
- MoN(1000cy)/AlN(300cy) @ 450°C on Cu coils: **266 wires/ 50 μm diameter**

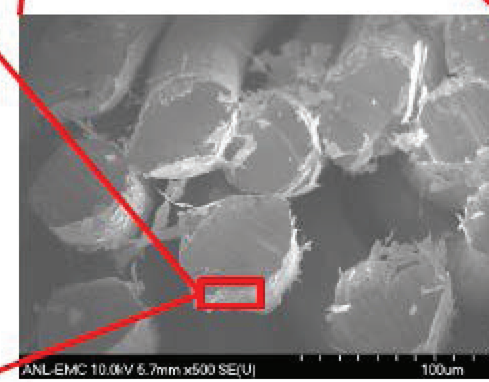




Conformal deposition.

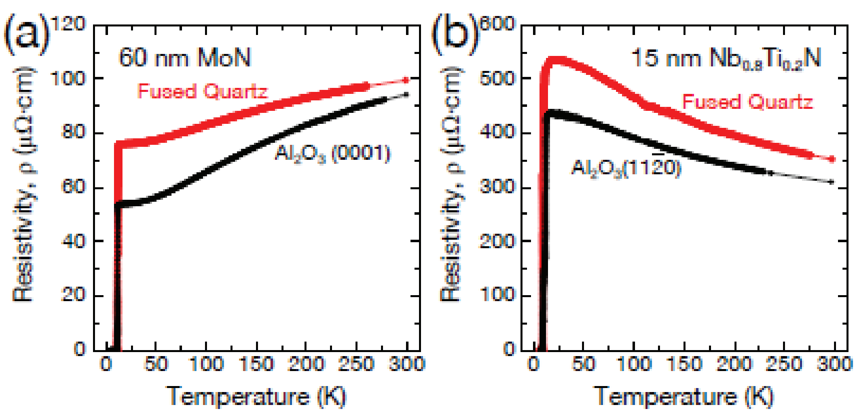
~ 250 Amps per layer of MoN
4 layers AlN/MoN -> 1kAmps!

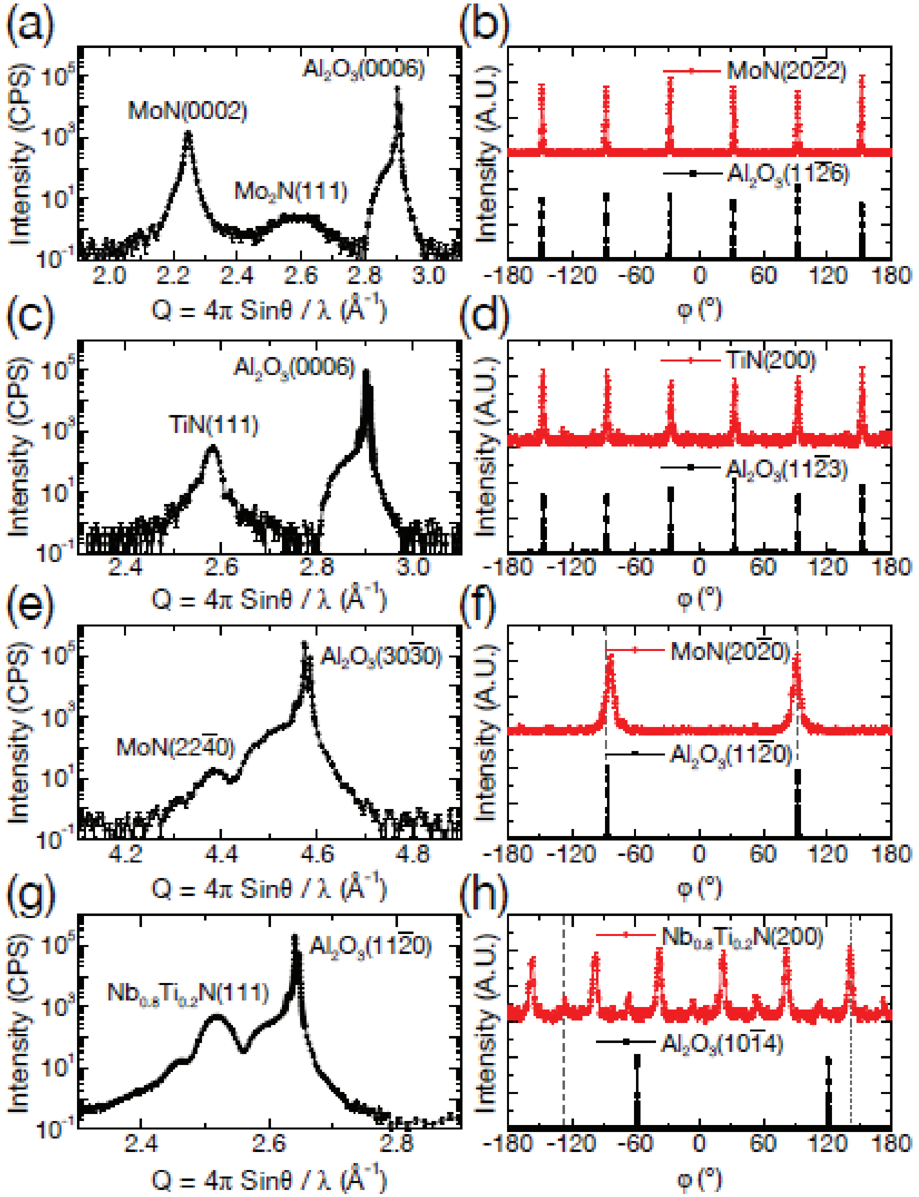




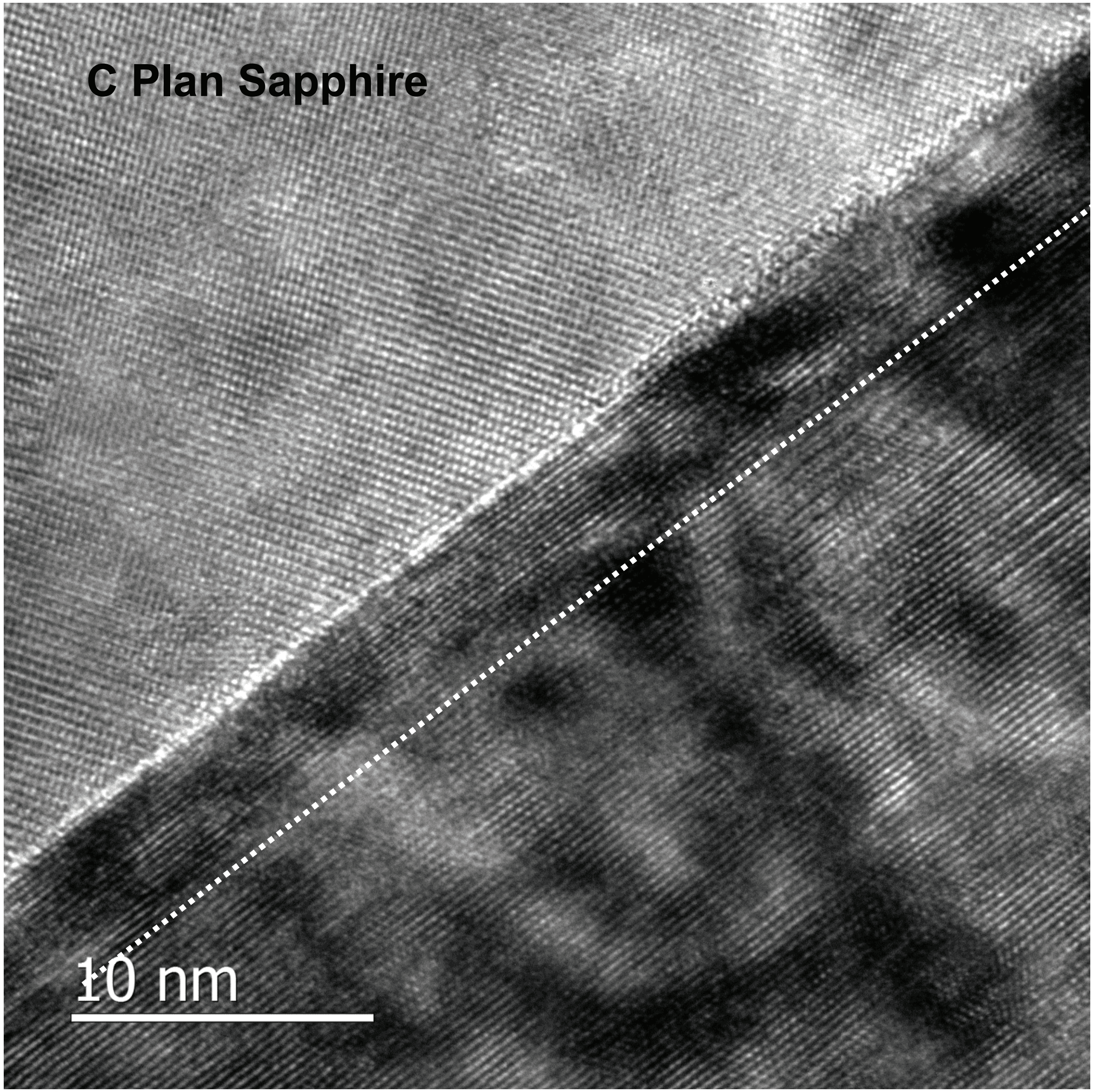
Epitaxy

Film	Substrate	Epitaxial relationship
MoN	c-Al_2O_3	$(0001)\langle 10\bar{1}0\rangle MoN \| (0001)\langle 11\bar{2}0\rangle Al_2O_3$
NbN	c-Al_2O_3	$(111)[1\bar{1}0]NbN \| (0001)[10\bar{1}0]Al_2O_3$ $(111)[\bar{1}10]NbN \| (0001)[10\bar{1}0]Al_2O_3$
TiN	c-Al_2O_3	$(111)[1\bar{1}0]TiN \| (0001)[10\bar{1}0]Al_2O_3$ $(111)[\bar{1}10]TiN \| (0001)[10\bar{1}0]Al_2O_3$
MoN	m-Al_2O_3	$(11\bar{2}0)[1\bar{1}00]MoN \| (10\bar{1}0)[1\bar{2}10]Al_2O_3$
$Nb_{0.8}Ti_{0.2}N$	a-Al_2O_3	$(111)[1\bar{1}0]Nb_{0.8}Ti_{0.2}N \| (11\bar{2}0)[1\bar{1}00]Al_2O_3$ $(111)[\bar{1}10]Nb_{0.8}Ti_{0.2}N \| (11\bar{2}0)[1\bar{1}00]Al_2O_3$ $(111)[1\bar{1}0]Nb_{0.8}Ti_{0.2}N \| (11\bar{2}0)[0001]Al_2O_3$ $(111)[\bar{1}10]Nb_{0.8}Ti_{0.2}N \| (11\bar{2}0)[0001]Al_2O_3$
MoN	r-Al_2O_3	$[1\bar{2}12][10\bar{1}0]MoN \| (1\bar{1}02)[\bar{1}101]Al_2O_3$ $[1\bar{2}12][\bar{1}010]MoN \| (1\bar{1}02)[\bar{1}101]Al_2O_3$
TiN	r-Al_2O_3	$(135)[1\bar{2}1]TiN \| (1\bar{1}02)[\bar{1}101]Al_2O_3$ $(\bar{1}35)[\bar{1}2\bar{1}]TiN \| (1\bar{1}02)[\bar{1}101]Al_2O_3$





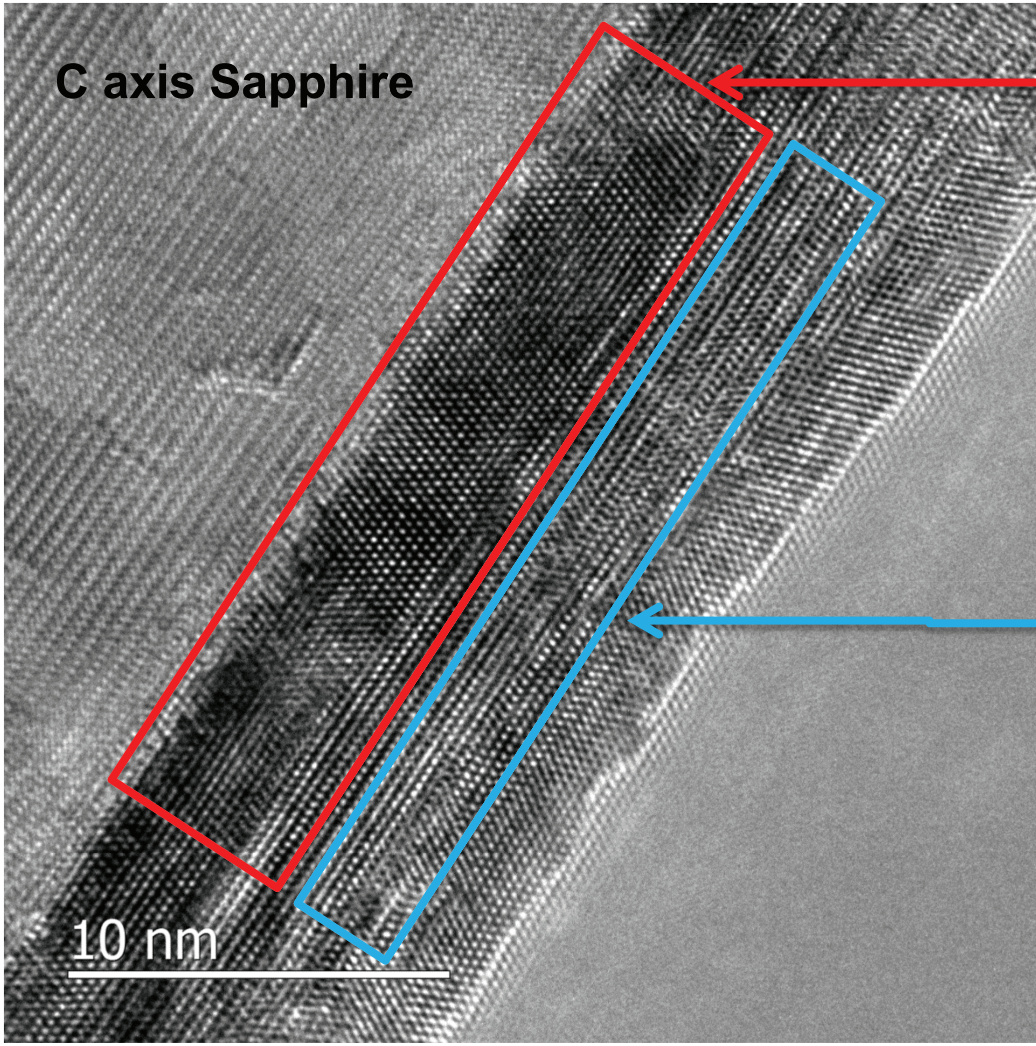
MoN: High Res. Cross section TEM

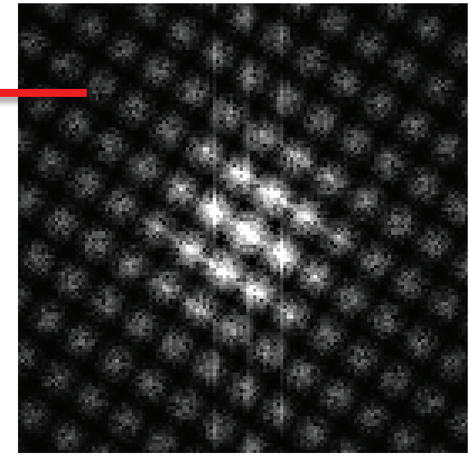


From this direction with the film very thick we can confirm that we have the hexagonal phase through-out the majority of the film

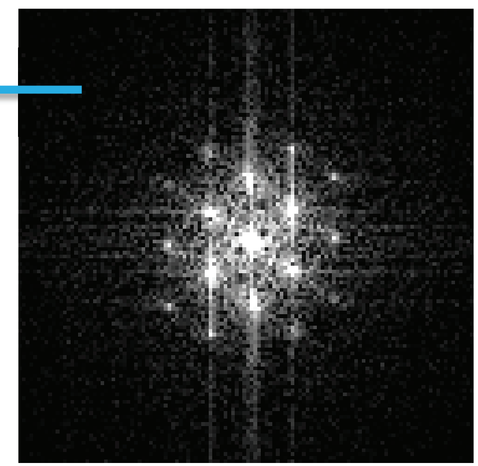
By addition gentle ion milling we can thin the sample and focus on the interface

Nucleation of MoN





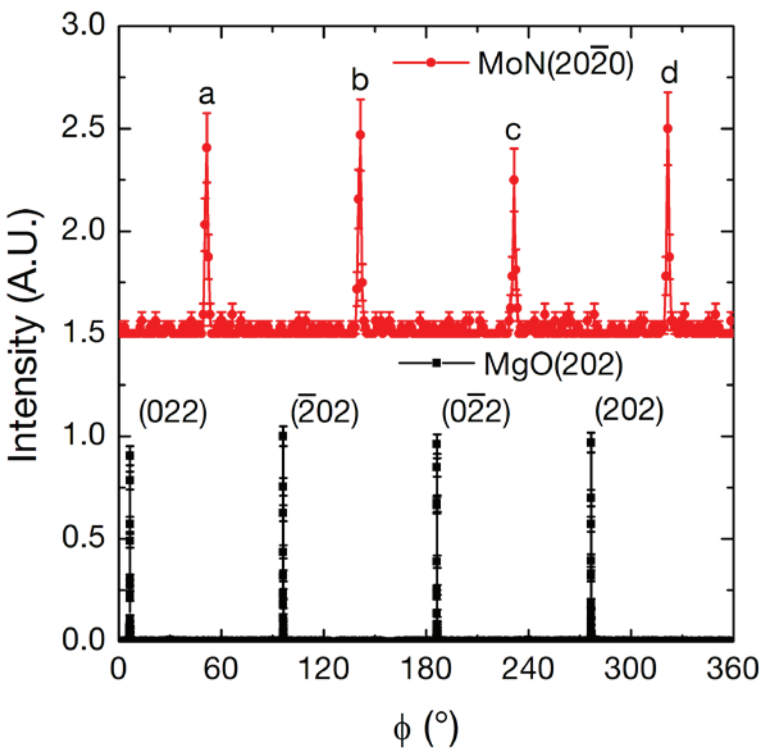
[110] of Mo_2N

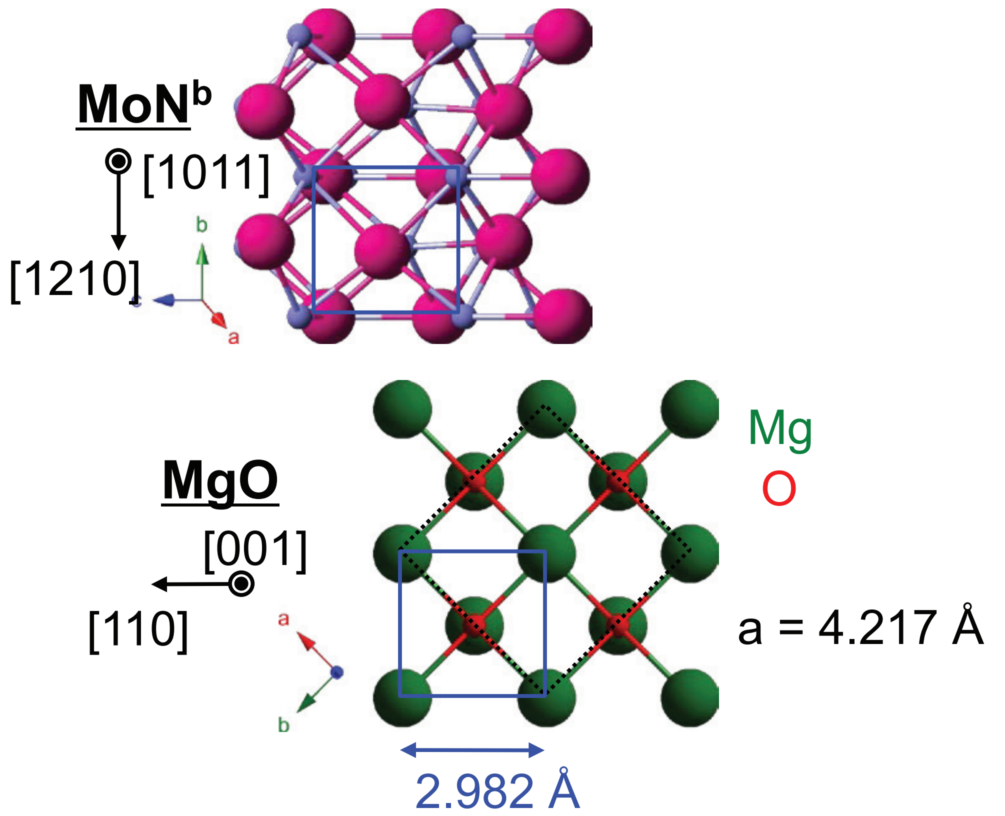


[110] of MoN

MgO(001)-MoN (211)

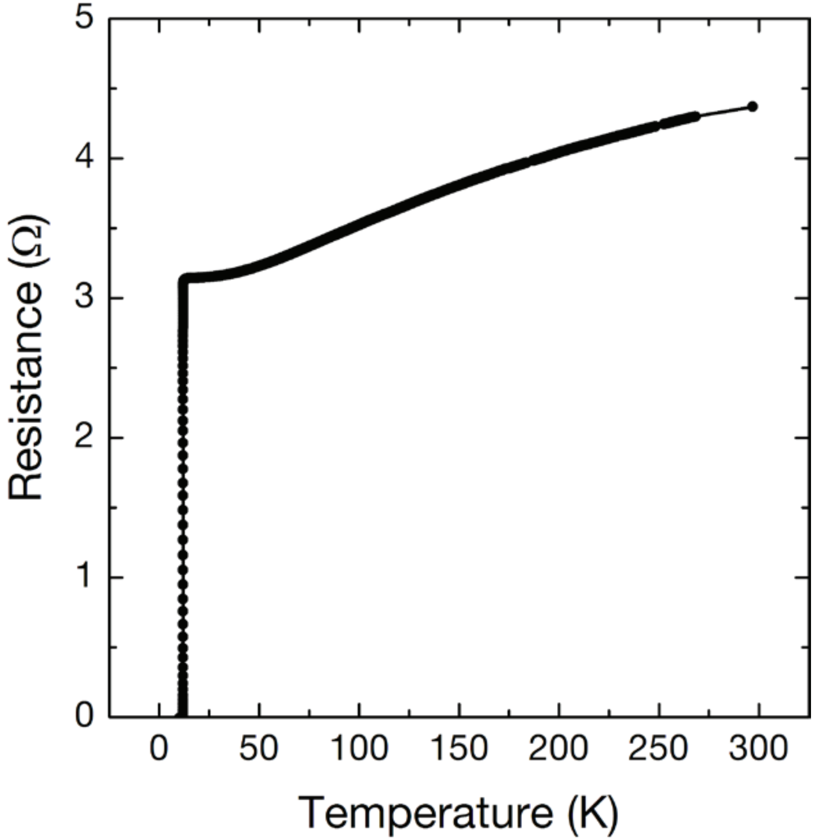
Grown at 450 °C

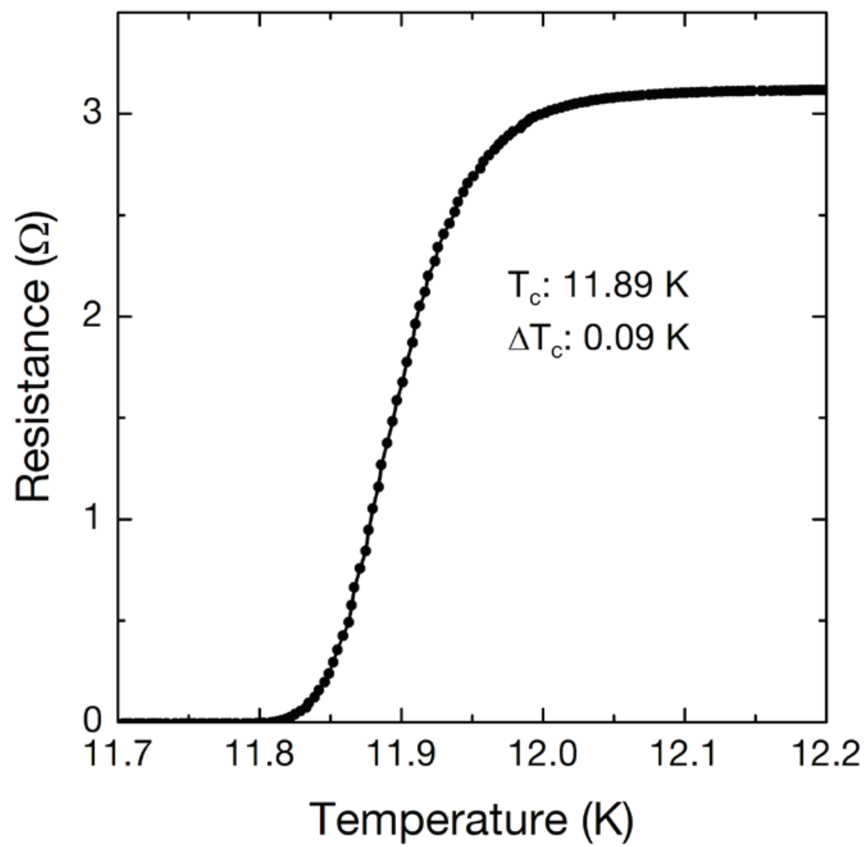




MgO-MoN

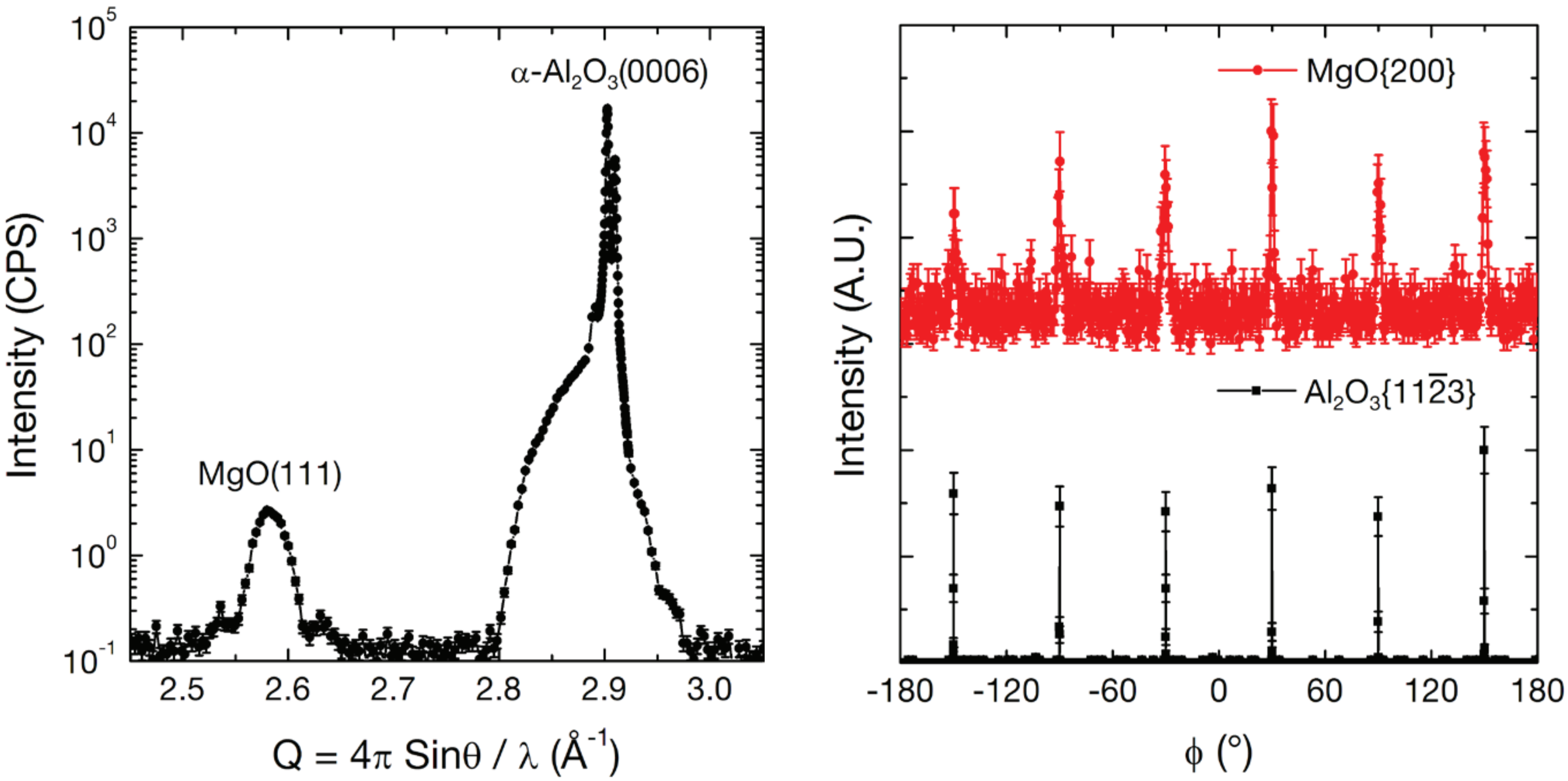
- MgO is a good tunnel barrier (for Josephson junctions etc)
- Good lattice match with Nb : attractive for SRF cavity
- We have high, sharp T_c for MoN on (001)MgO



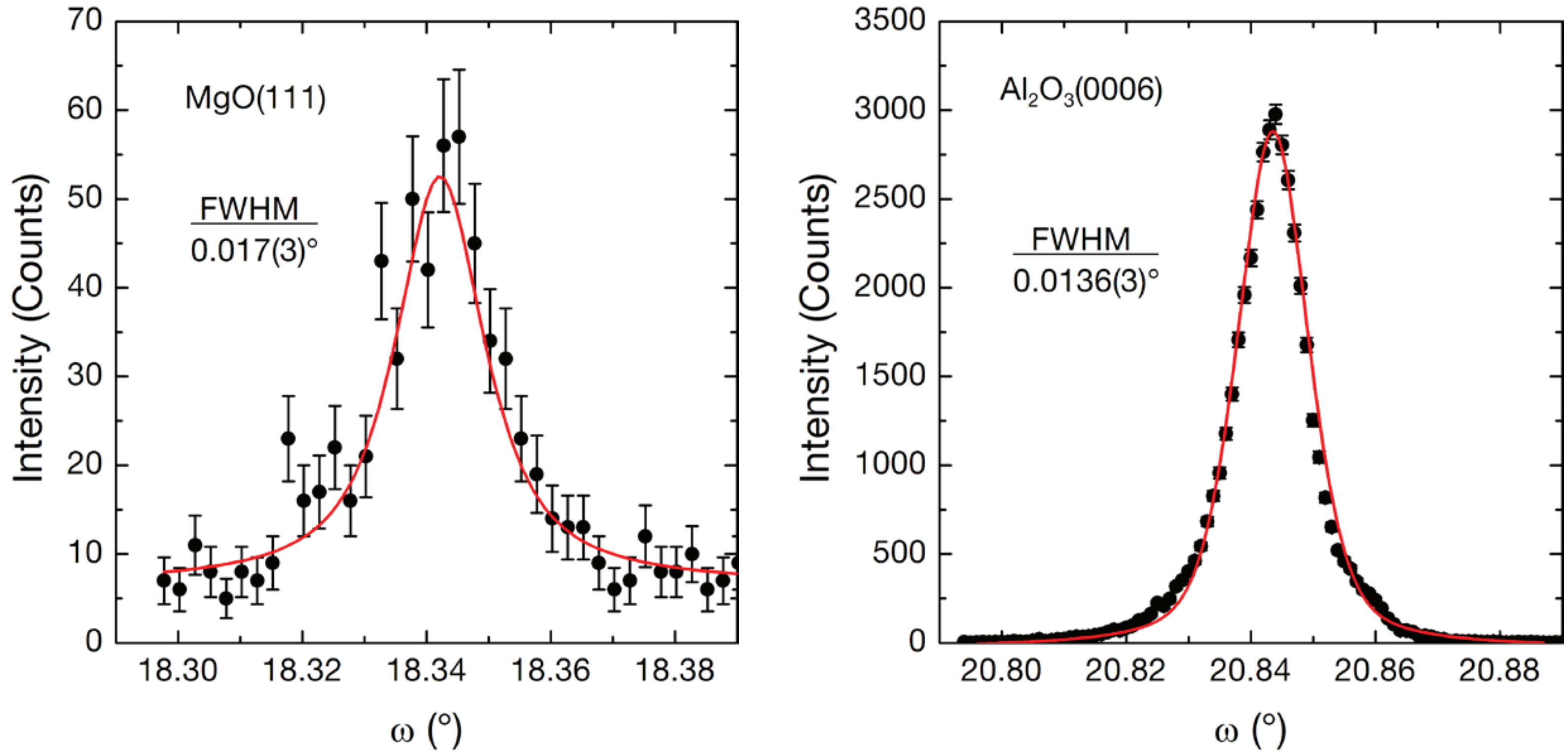


Can we grow MgO by ALD? OUI!

- 200 cycles $Mg(Cp)_2/H_2O$ at 300 °C -- 10nm thick

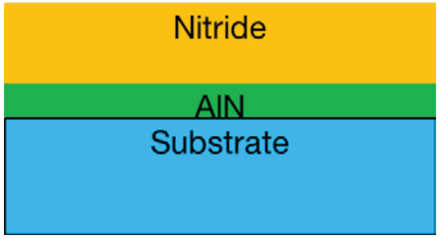


MgO/(0001)Al_2O_3



- OP/IP epitaxy: (111)[110]MgO || (0001)[1010]Al_2O_3
  (111)[110]MgO || (0001)[1010]Al_2O_3
- OP/IP alignment (FWHM): $\Delta\omega$: 0.017 ± 0.003°, $\Delta\phi$: 2.1 ± 0.4°
- Same epitaxy as observed for NbN and TiN *at much lower temperature*
- Many materials haven't been deposited epitaxially by ALD simply because no one has tried.

Titanium Nitride (TiN) and NbTiN



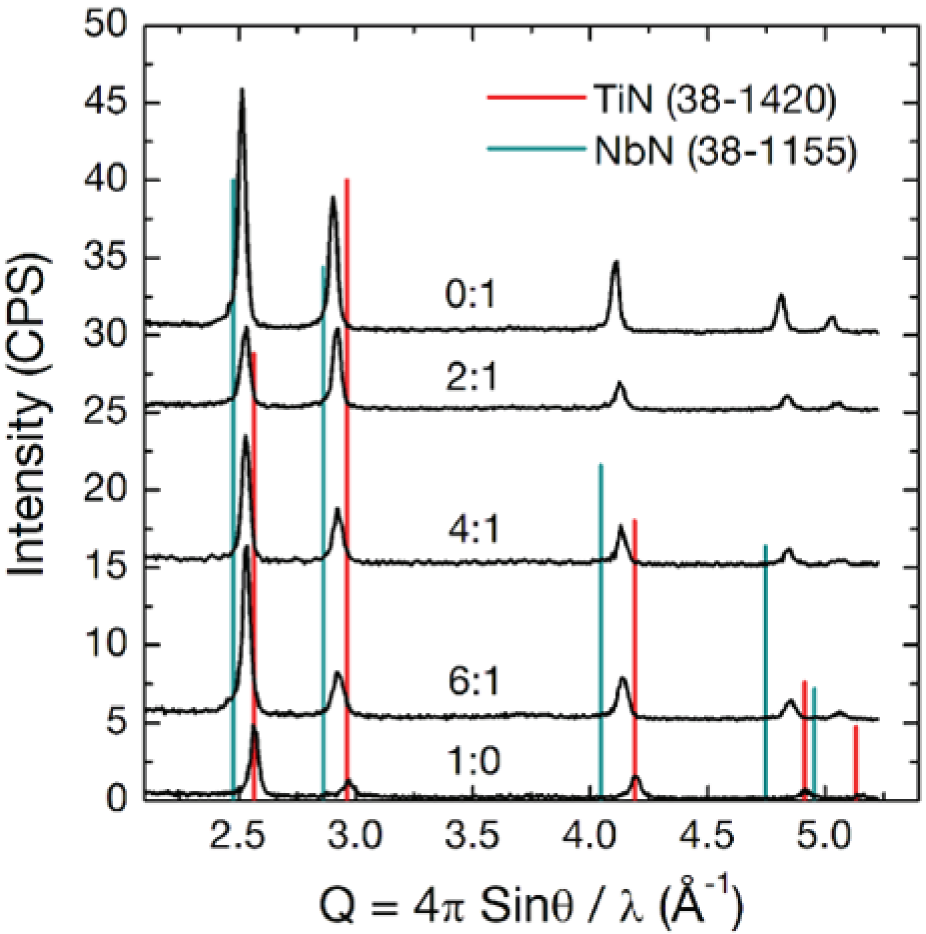
- Chemistry: ($NbCl_5$:$TiCl_4$) + NH_3 at 350°C and 450°C
  - TiN and NbN GR < 1 unit cell ~ 0.3 and 0.2 Å/cy respectively
- Can vary Ti content with $NbCl_5$:$TiCl_4$ ratio (1:2 ~ 20% TiN)
  - Cubic δ phase in all films

With increasing TiN

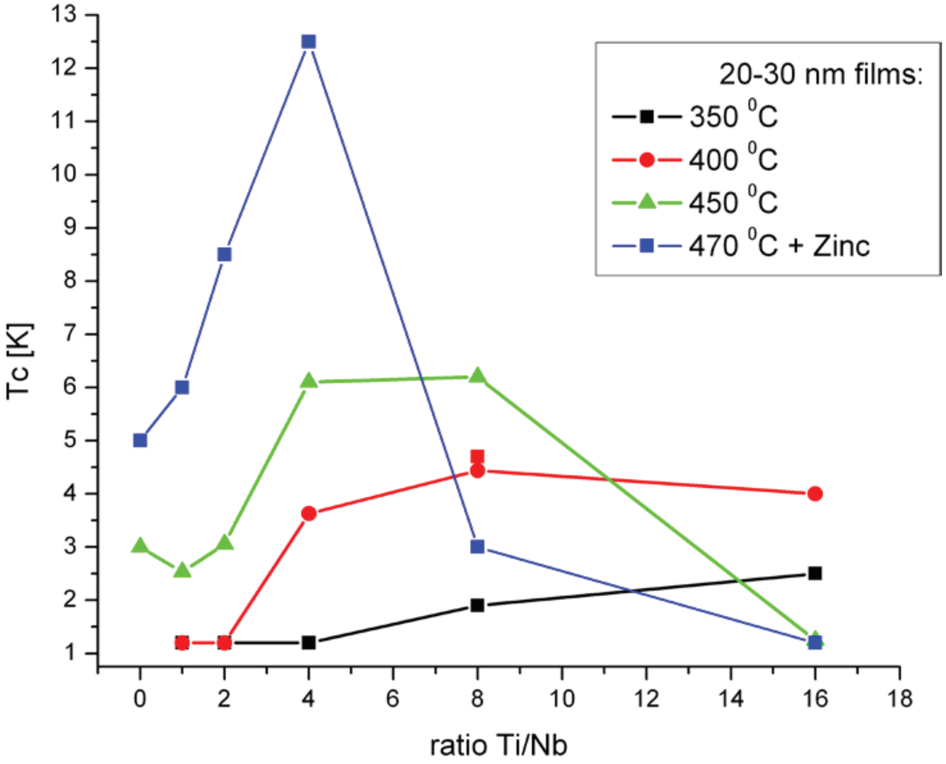
- Peaks shift to higher angle
- Density decreases
  - 7.2 g/cm^3 (1:0)
  - 5.7 g/cm^3 (1:4)
- RT resistivity decreases
  - 380 μΩ-cm (1:0)
  - 130 μΩ-cm (1:4)

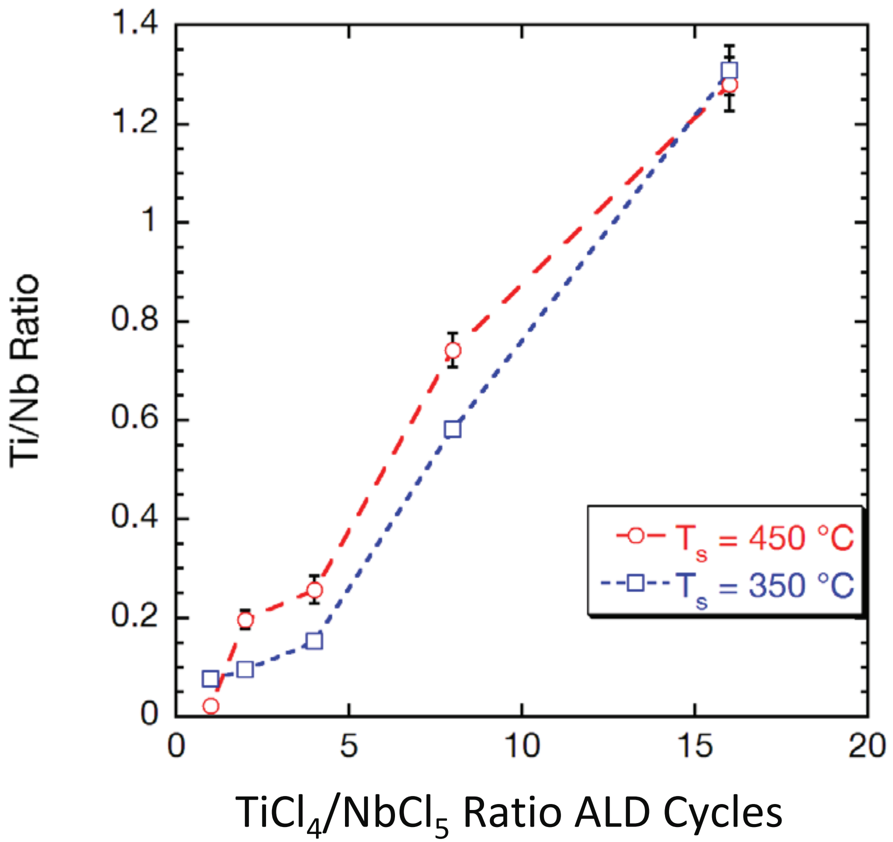
Chlorine content: 0.05 atom %

Are they good superconductors?



Controlled composition

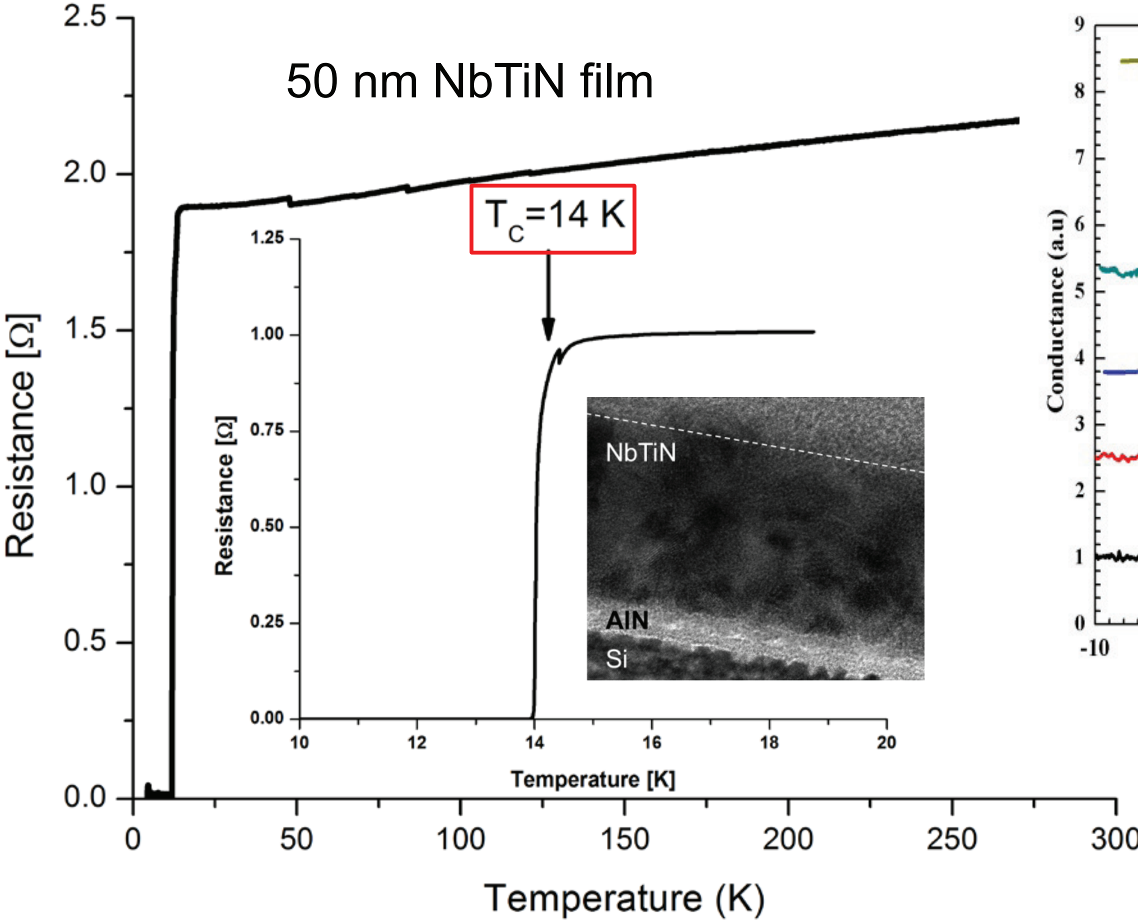




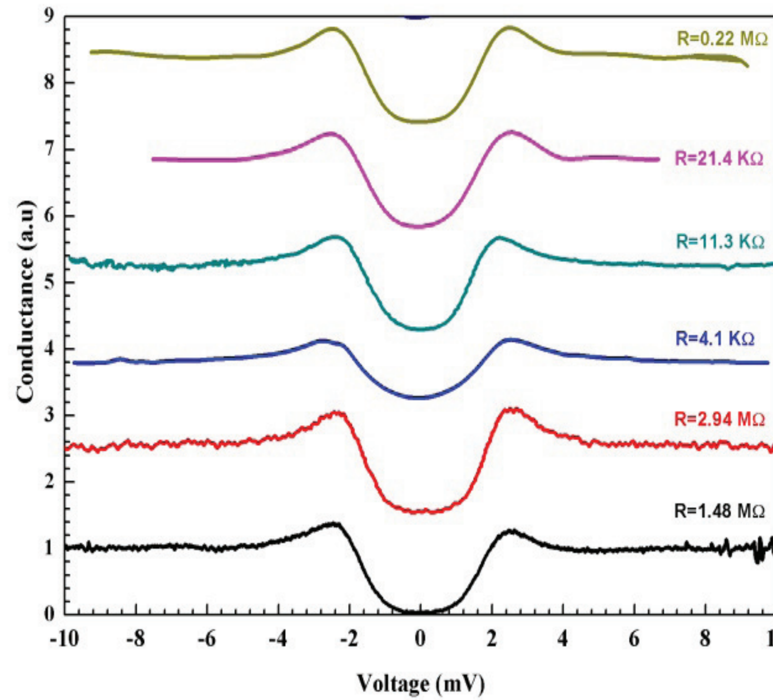
Atomic control of Composition -> Tune Tc

$Nb_{1-x}Ti_xN$: Superconducting T_c

- Achieved superconducting T_c=14 K 4:1
  - 40% higher than previously reported value for ALD NbN [Hiltunen *et al.*, *Thin Solid Films* **166**, 149 (1988)]



T= 1.8K



Δ = 2.3 meV +- 0.1

Synergy

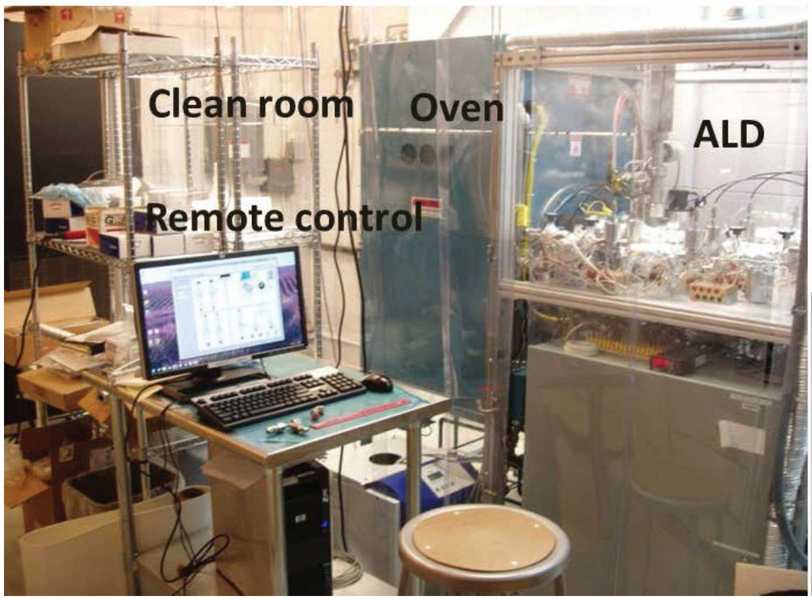
Jlab

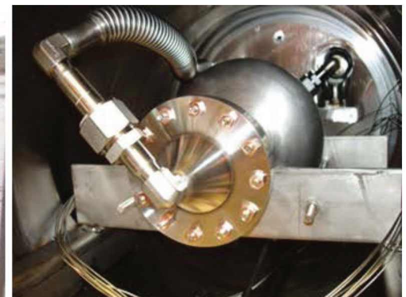
Single Cell Nb Cavities Testing

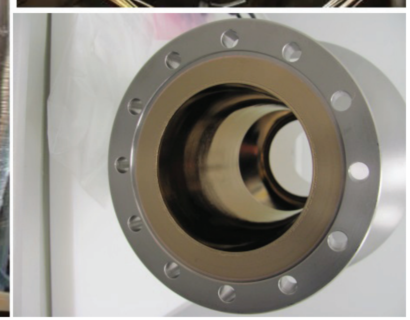
FNAL

Single Cell Nb Cavities

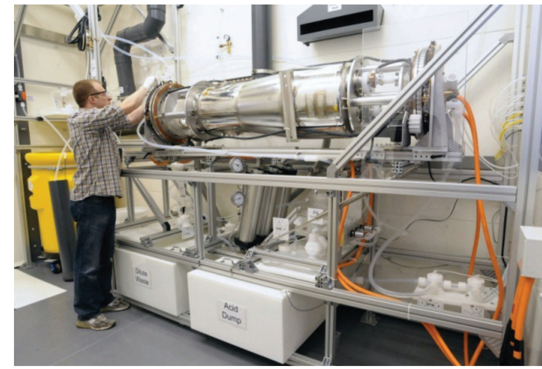
MSD/HEP







PHYS/APS/HEP

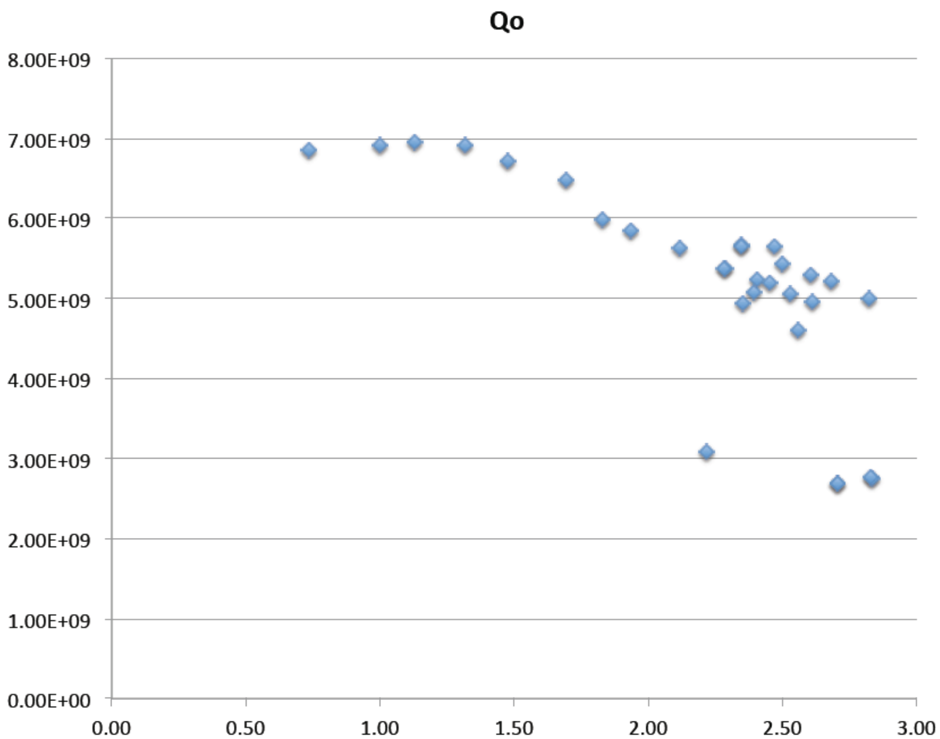


Material science fundamentals

Surface treatments
RF testing

Cavity test Results: NbTiN/AlN coating

PAV002 After coating by ALD (15 nm AlN/ 60 nm NbTiN)



Reprocessing:
- Tumbling
- Chemical etching
- Baking
- Baseline testing

Thermal Q switch of the cavity ~ 3 MV/m
-> does the multilayer quench?
-> Is the Tc of NbTiN what we anticipated (14K)?

Other Materials to try: MoN/AlN

Temperature mapping will give us answers !!

- Multilayers:
  Atomic Layer Deposition of superconductors.
  Coupons & SRF cavities.

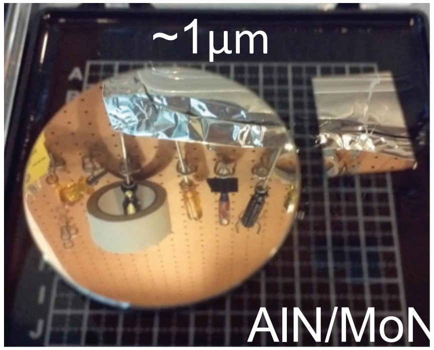
- Niobium on Copper
  Coupons and RF test.

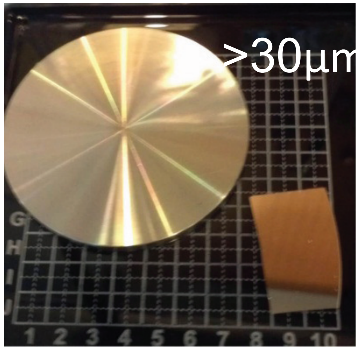
Nb onto Cu Initial surface roughness?

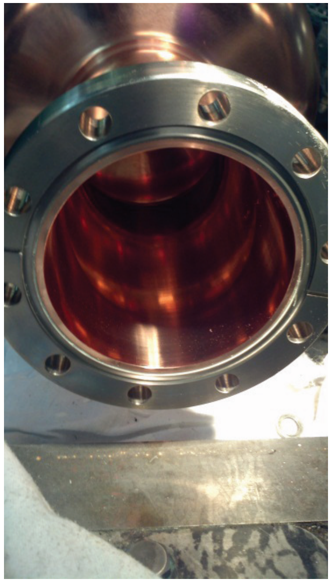
- **Cu surface: Surface roughness, Sticking onto Cu**

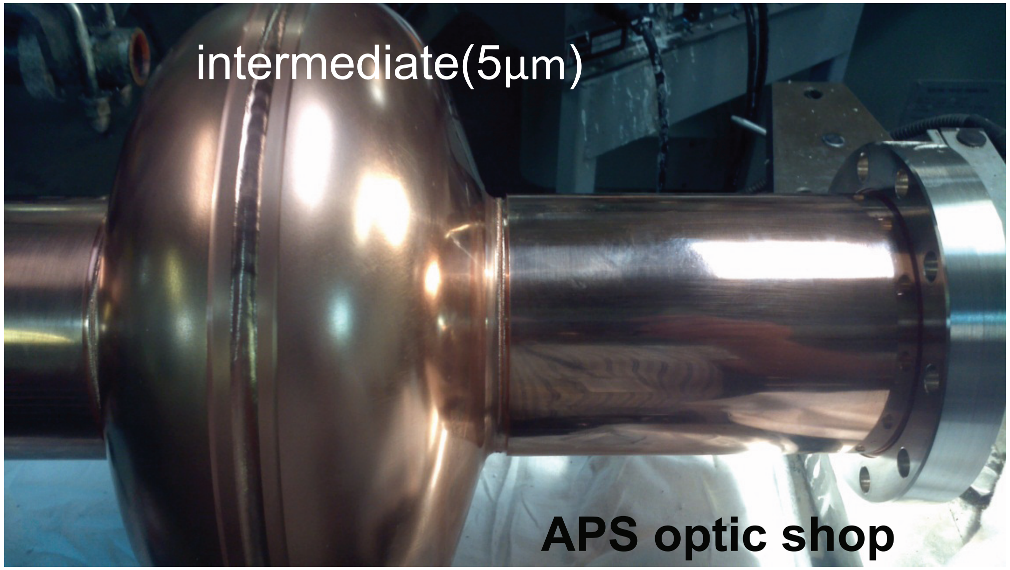
µm Sup films <-> roughness
4/HPR, All
3/EP (Able Electro)
2/Mechanical polish
(mirror-intermediate(5µm))
1/Machining

10 coupons









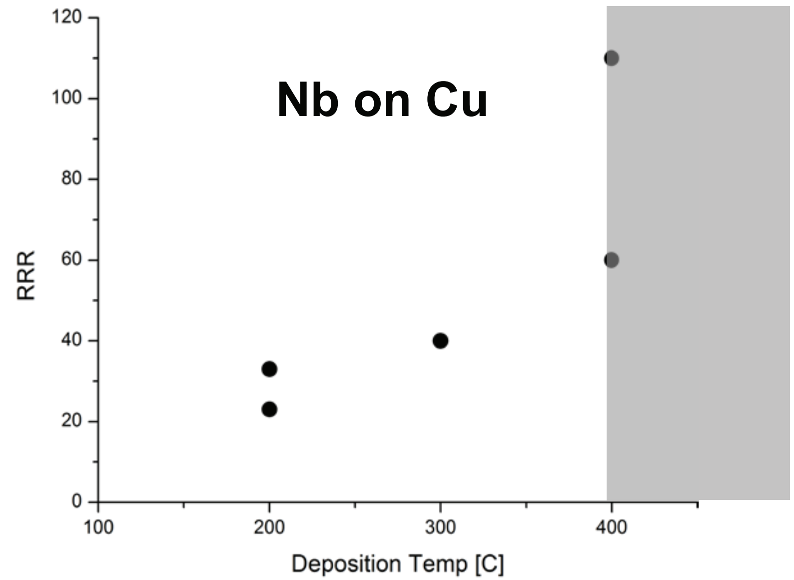
1 Cu cavity EP
1 Cu cavity Pol + EP

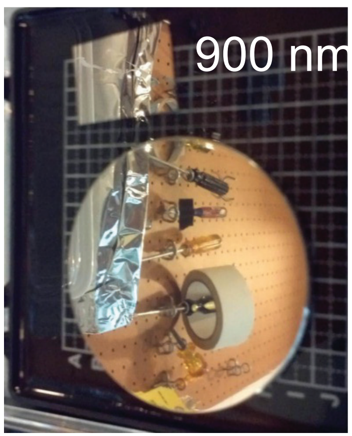
Nb/ Cu cavity
Alameda/Jlab

Sticking/diffusion barrier

High RRR for T >~ 400 C
BUT! delamination occurs

Solution: seed layer/diffusion barrier
ALD can Help:
Nitrides

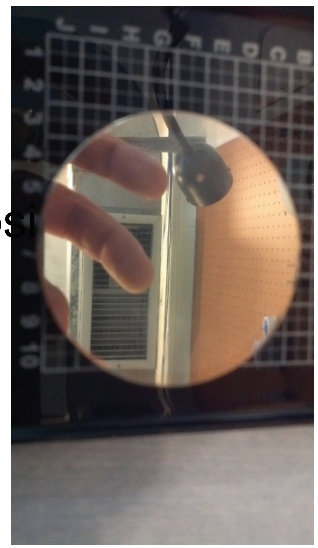




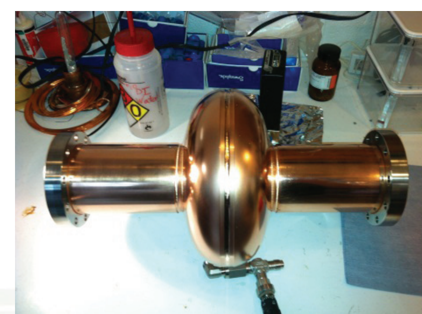
900 nm

AlN/MoN/AlN

HPR 2000 psi



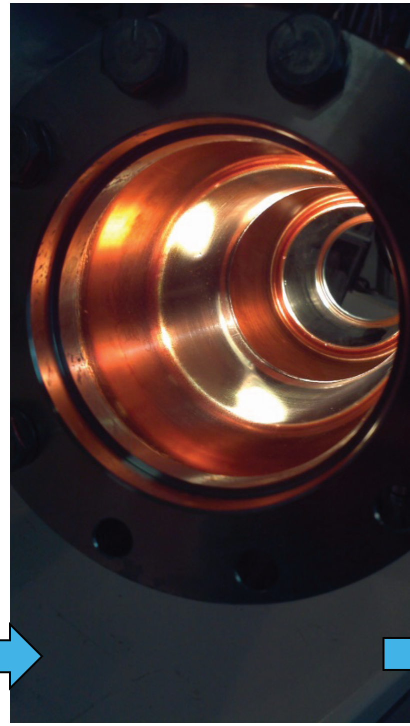
- Send coupons to HIPIMS/Alameda/Jlab
  RF tested @ Jlab (end of March)
- Send EP Cu Cavity to Alameda
  Dep of Nb @ 450C and above.

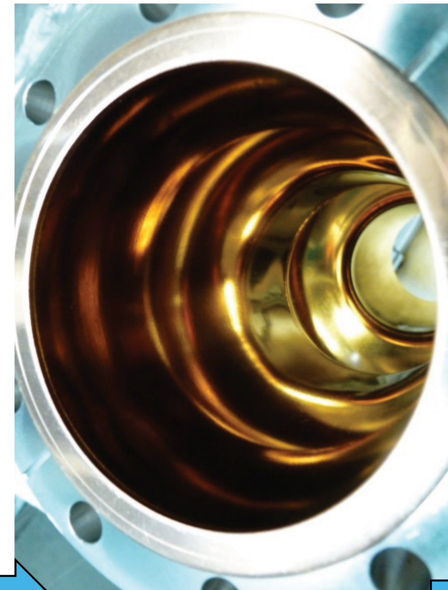


Nb sputt @ 450 ^{0}C

TiN, MoN/TiN coating works

Cavity Coating test





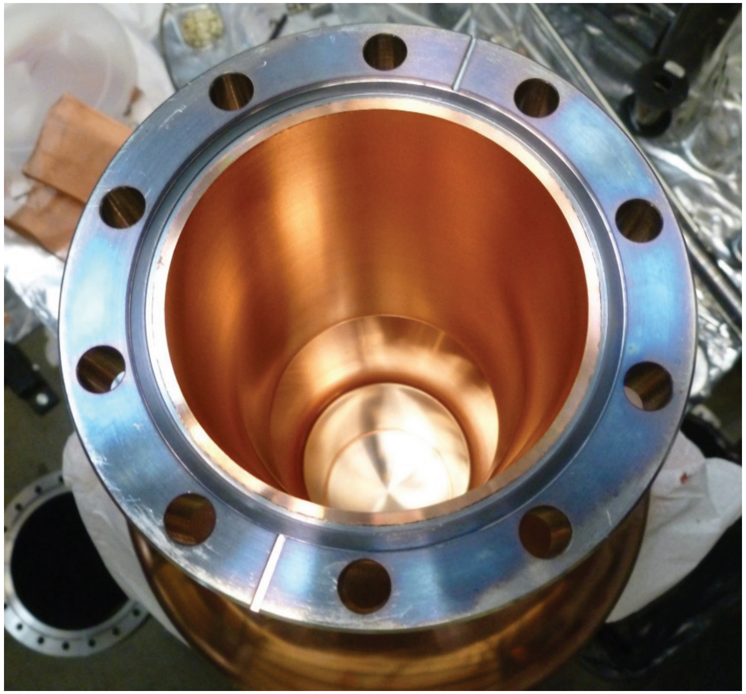
Cu Cavity AES

Mechanical/Chemical Polishing

ALD coating TiN 20 nm @ 450 ^{0}C

Nb coating - 5μm Iris ~ 25 μm Beam Tube 375 ^{0}C

Cavity Coating test



AES cavity / HPR ANL

Nb Coating 2 µm Iris / 5 µm beamtubes
350 ^{0}C

Q ~ 1-2 10^7 4K, no improvement upon cooling

Summary

- Tune Tc and transport properties by controlling film thickness
- Tune Tc and transport properties by controlling composition: $Nb_{1-x}Ti_xN$, TiN, MoN, NbSi, NbC, NbCN … and thickness.
- Growth temperature : lower Temp for High quality nitrides
- Fundamental interest for Quasi 2D limits + applications: Bolometers, High energy physics accelerators, magnets…
- Future work: MgB_2 (40K), FeSeTe (15K), $K(FeSe)_2$ (31K) etc…

  More cavity coating/Testing

  Nb onto cu cavities

Goal

Our research is directed towards depositing MgB_2 films onto cooper or other high thermal conductivity metals which would allow future superconducting rf cavities to be fabricated as film-coated structures.

Challenges

- Material synthesis: atomic layer deposition
  - Low-temperature growth (<500 C) of high-quality MgB_2
  - Create new chemistries and precursors (more reactive precursor for magnesium is needed).
  - ALD systems for cavity scale up and coupons research available and working
- Materials synthesis: hybrid physical chemical vapor deposition (HPCVD)
  - High-temperature growth (> 600 C)
  - Growth on copper challenging: Sticking and diffusion can be an issue.
  - Thickness control and conformality limited -> bulk films (> 10 λ).

Synergy

- Nucleation/diffusion layer by ALD can mitigate contamination @ high temp. in HPCVD
- Bulk HPCVD + Multilayer ALD of MgB_2 can further increase performance @ 10° K and increase $E_{max.}$.

Cavity and cryomodule concept

Typically superconducting Nb CW cavity designs require about 20 to 60 W at 4 K

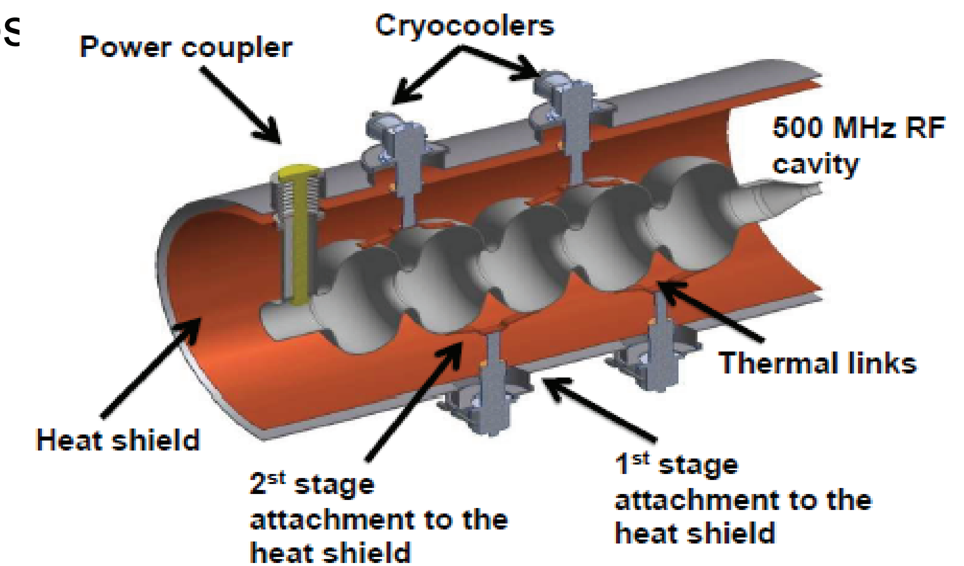
MgB2 films can achieve similar surface resistance at 8 -12 K.

It makes is possible to use cryocooolers for heat removal.

The cryomodule without liquid cryogens is considerably simplified. No liquid filling ports or internal piping, liquid reservoirs, or internal gas piping is required. Cryocoolers.

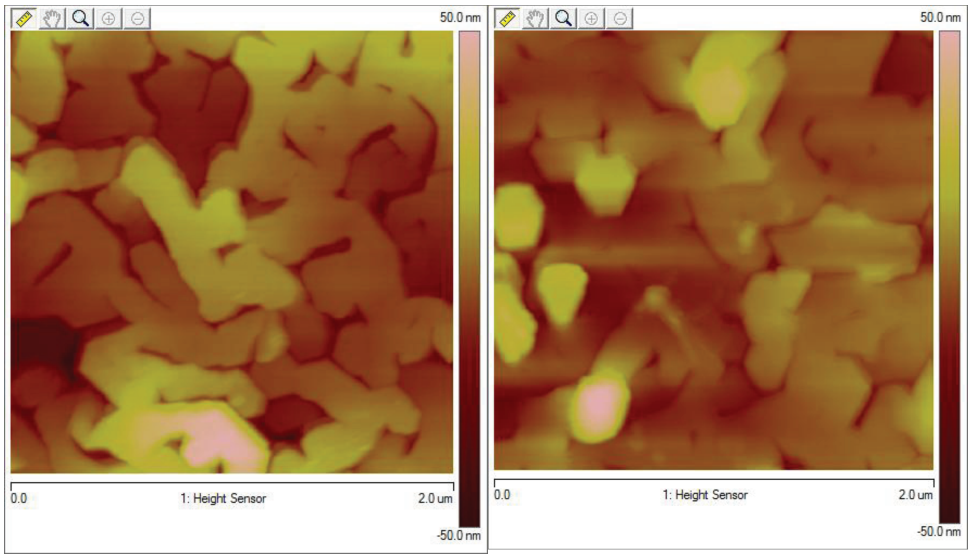
Ideally, a high thermal conductivity material like copper would be best for the cavity.

The thermal conductivity of Nb is about 200 W/mK, so even Nb would be usable as a subs ... per.



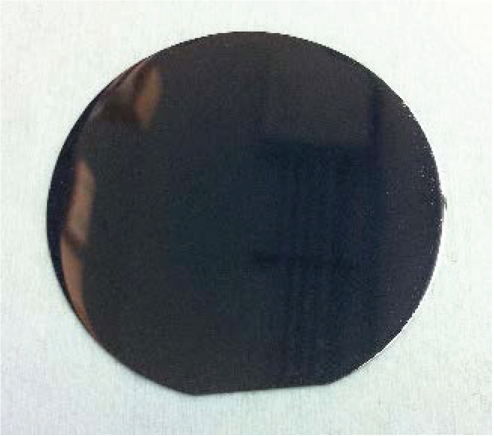
MgB_2 Film on 2" Sapphire Substrate

Surface Morphology: AFM

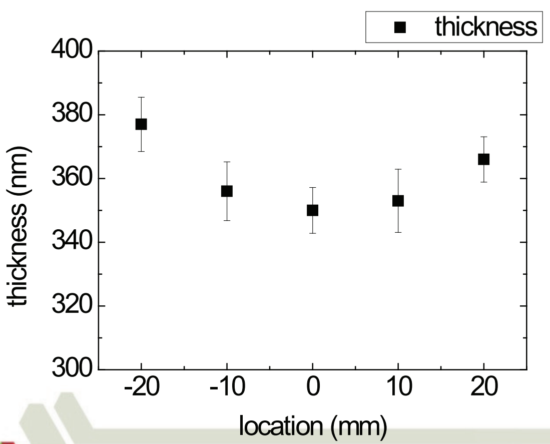


Center: Rq=10.4 nm **Edge: Rq=10.1 nm**

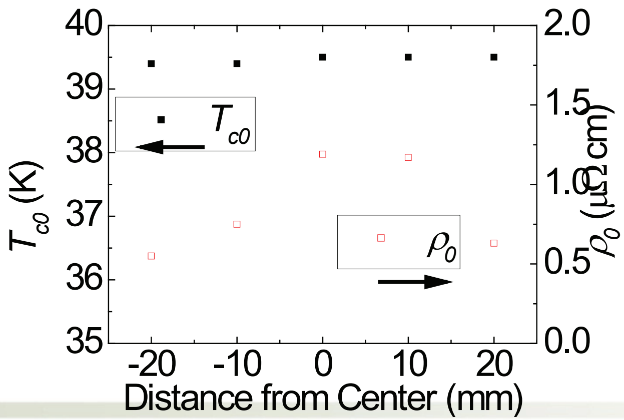
Appearance



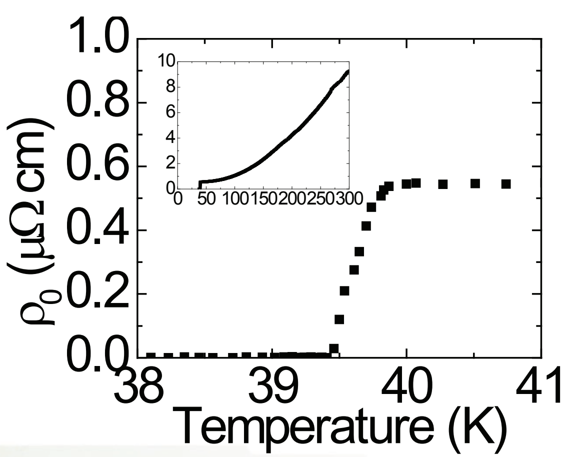
Film thickness vs location



T_c, ρ_0 distribution



Typical ρ-T curve

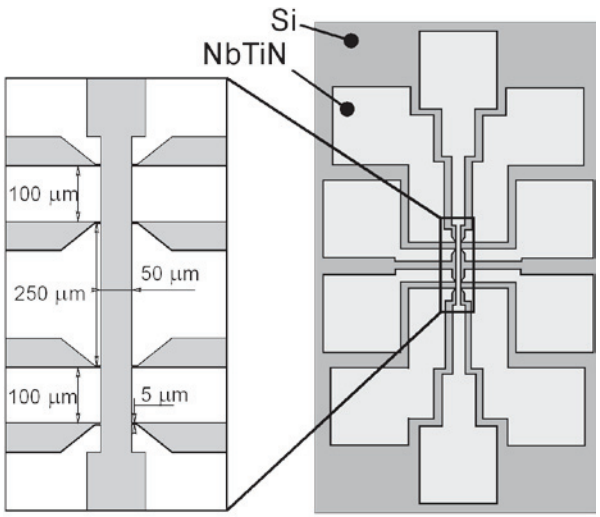


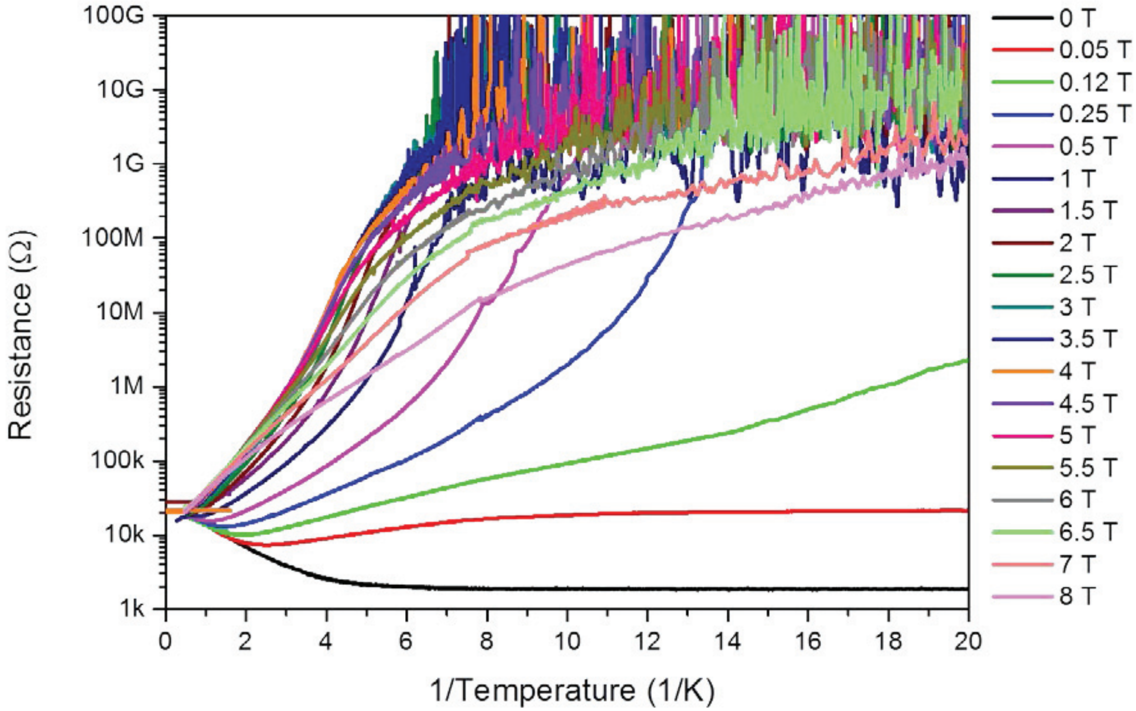
End

Super-insulating transition

UofC-ANL collaboration

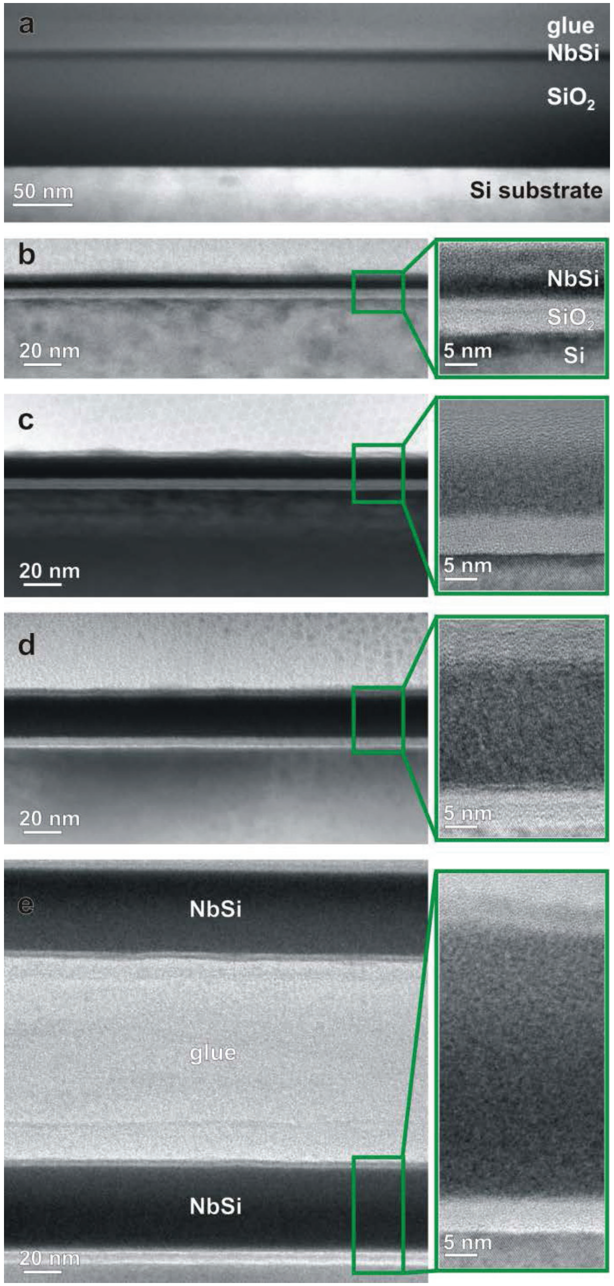
NbTiN is: 2x($TiCl_4$ + NH_3) and 1 ($NbCl_5$ + NH_3).
Grown @ 350C
$Nb_{0.8}Ti_{0.2}N$ – 2% Cl. ~ 5 nm

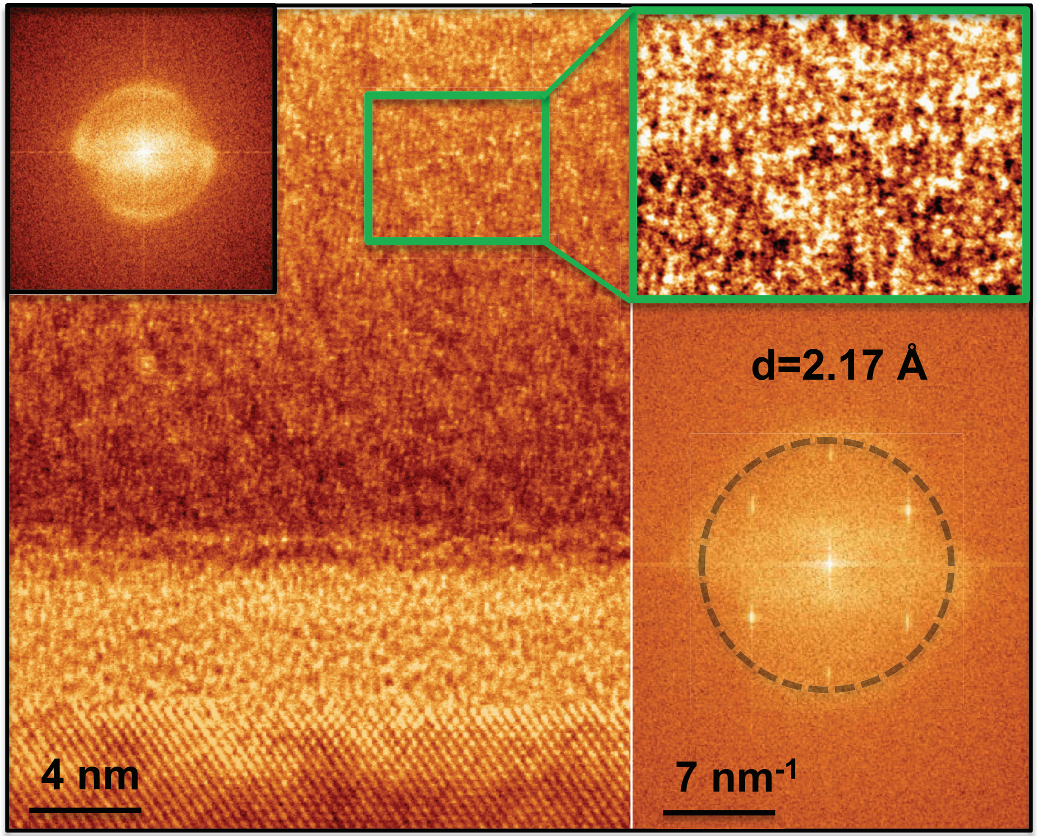




Possible super-insulating
new compound
higher T*

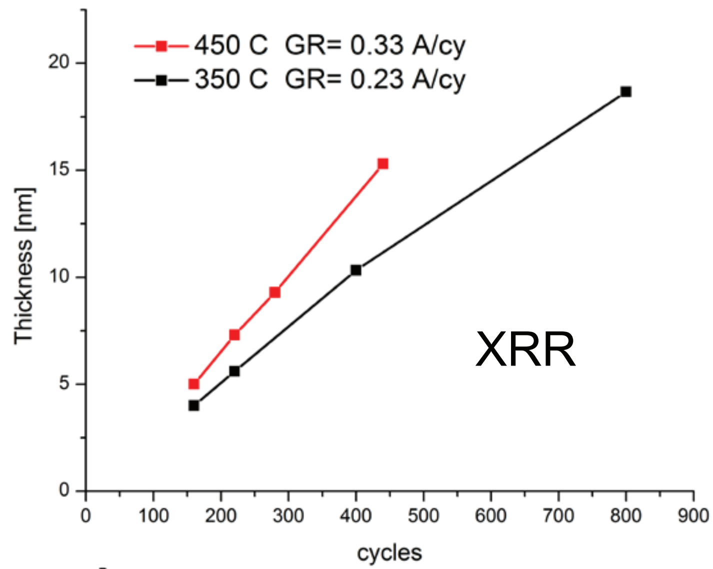
NbSi-TEM

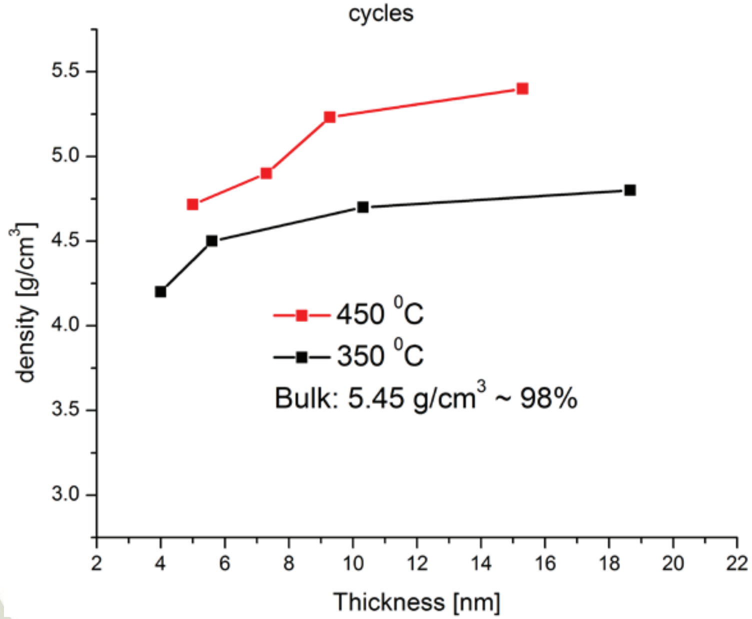


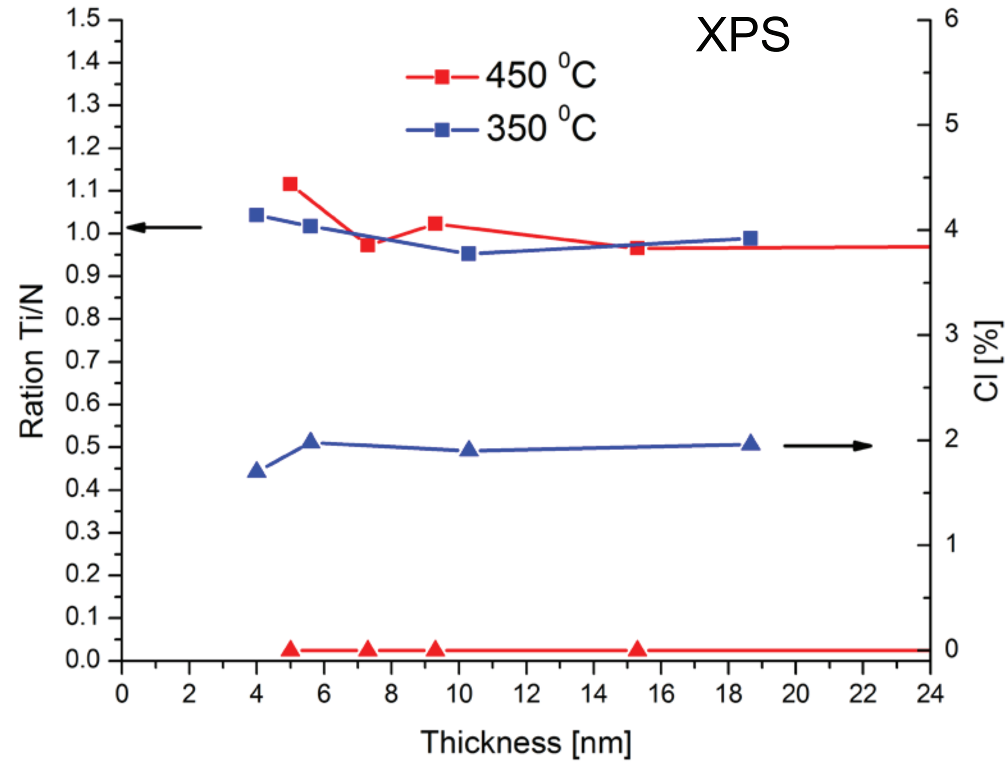


Polycrystalline- d= 2.174 A+- 0.03A

Titanium Nitride (TiN) (1:0)

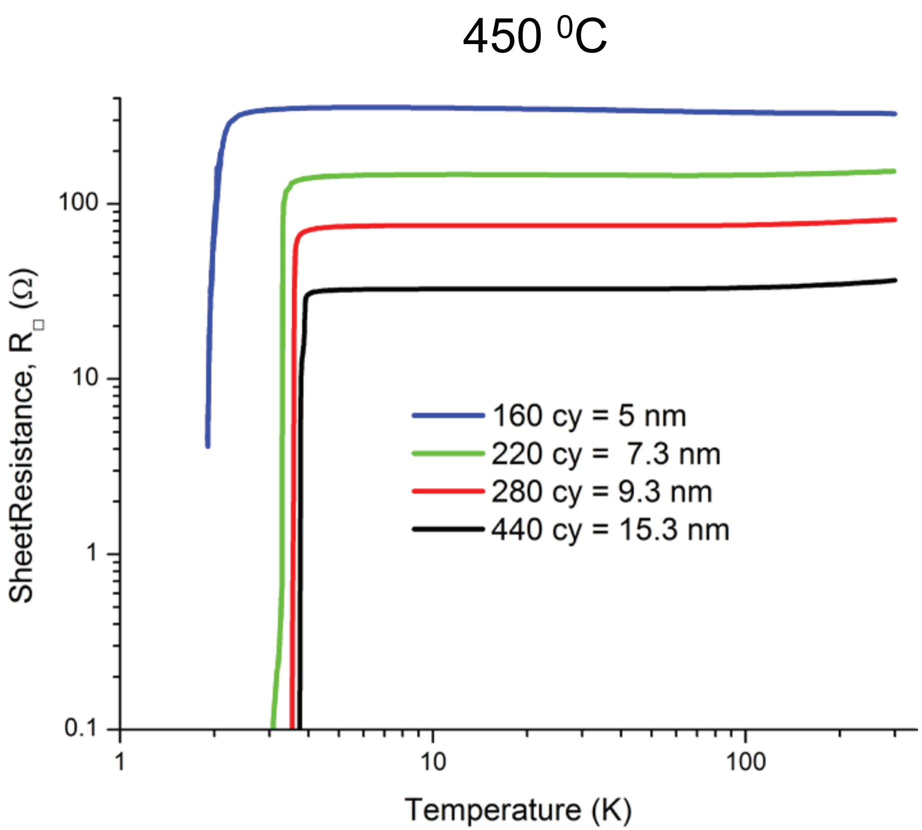


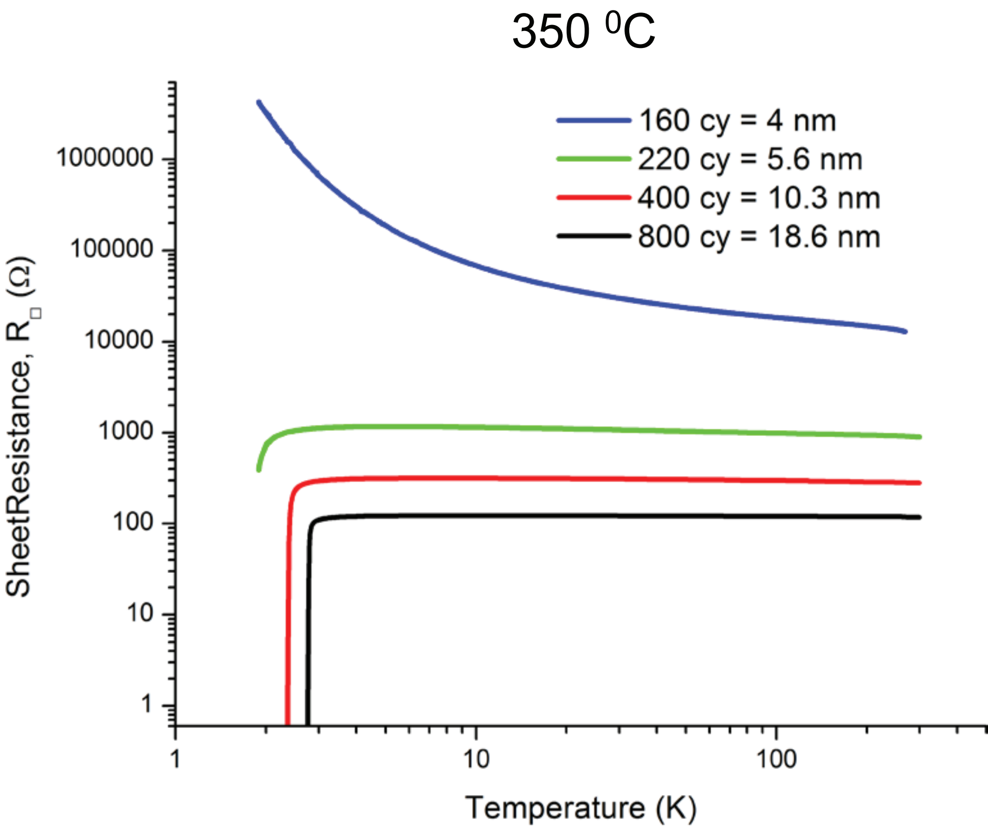




2 % Cl Impurities @ 350 ^{0}C
Lower densities (87%)

Titanium Nitride (TiN) (1:0)-transport

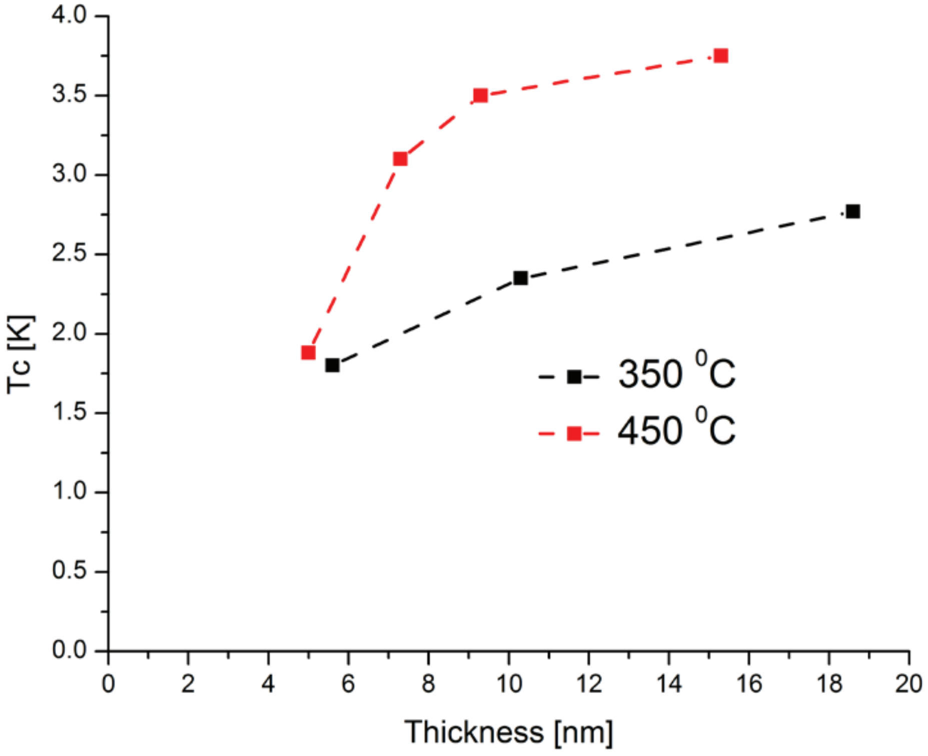


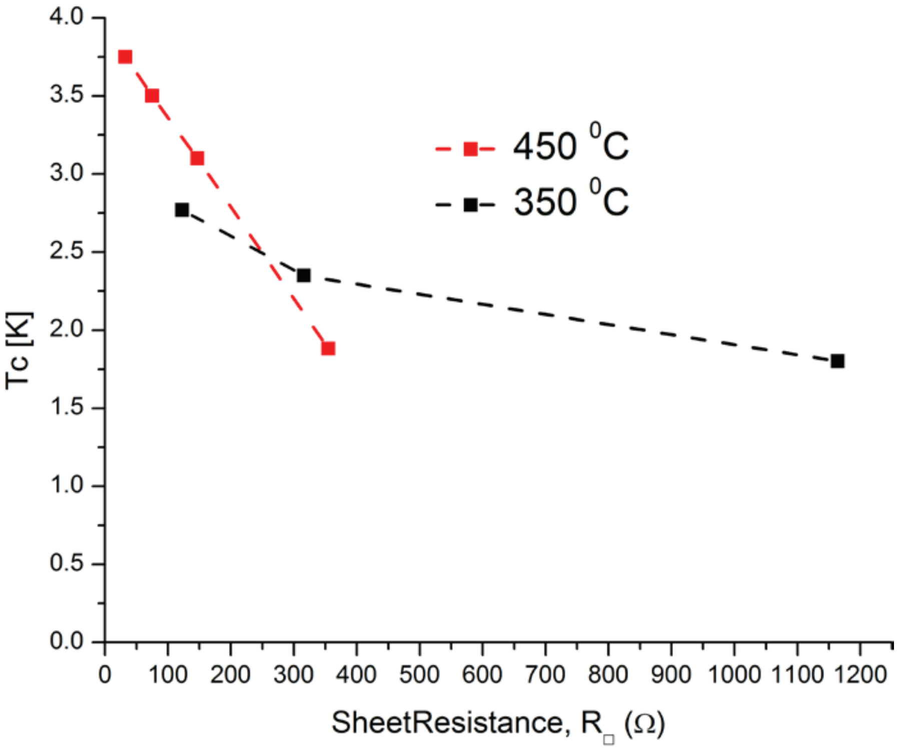


Resistivity bulk ~ 70 μΩ.cm
Tc bulk ~ 3.9 K

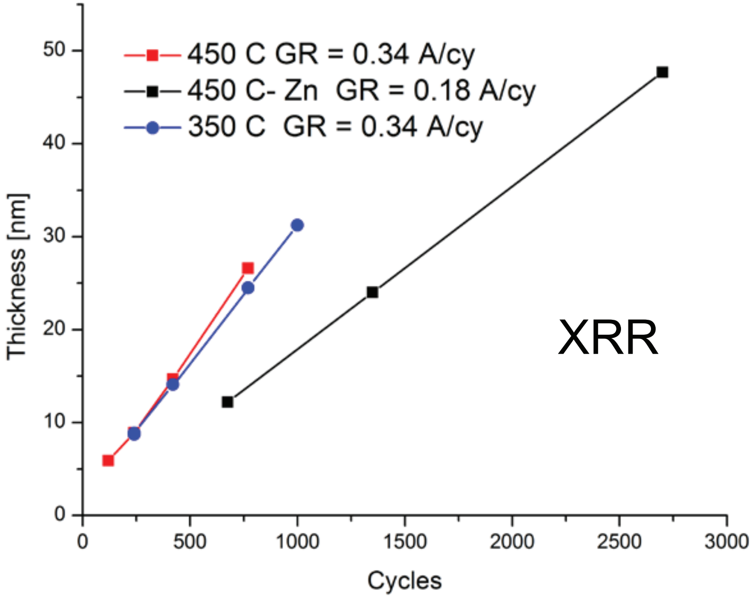
Resistivity bulk ~ 200 μΩ.cm
Tc bulk ~ 2.8 K

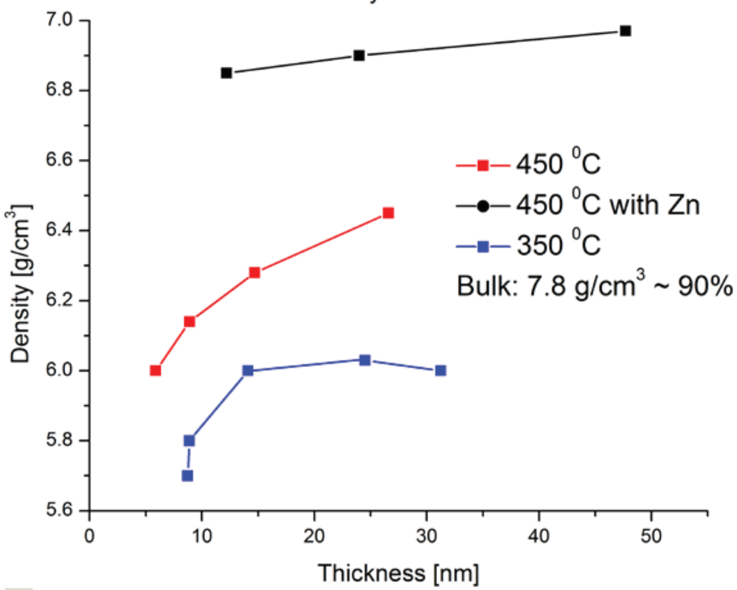
Titanium Nitride (TiN) (1:0)-transport





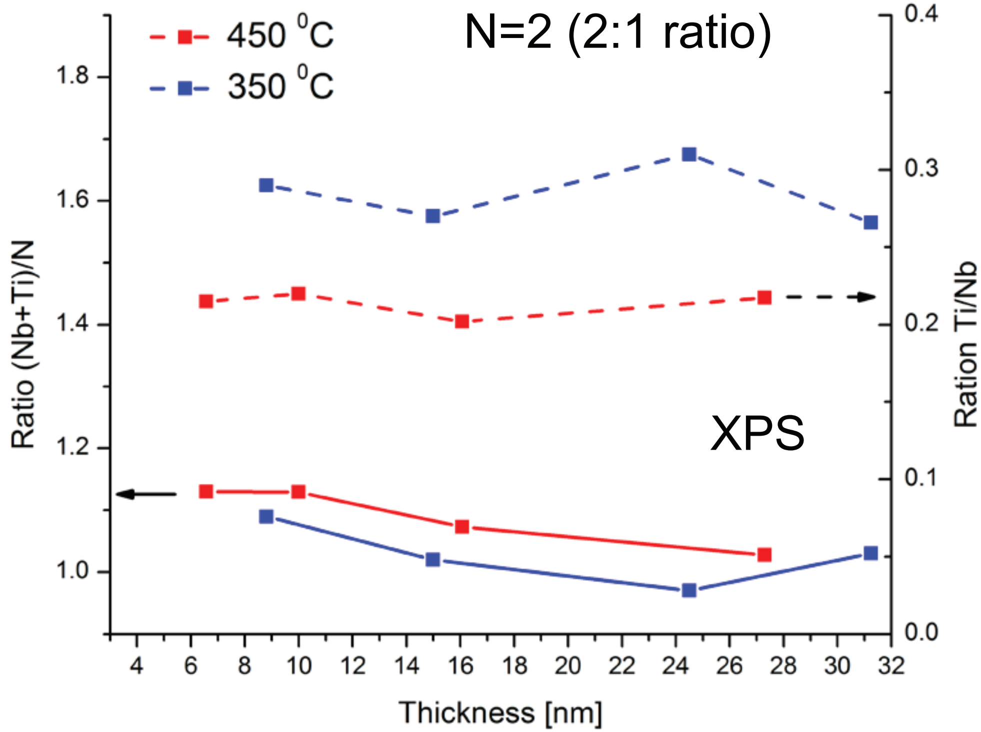
Niobium Titanium ($Nb_{1-x}Ti_xN$) 2:1





$(NbCl_5 + Zn + NH_3)_N + (TiCl_4 + Zn + NH_3)$

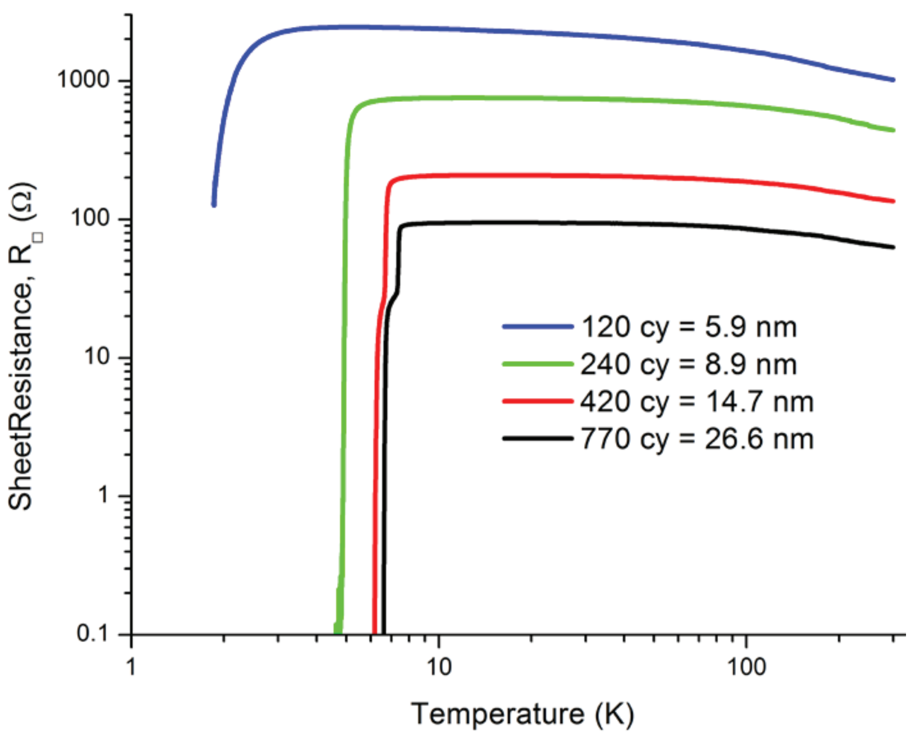
$\rightarrow ZnCl_2$ (> 410 ^{0}C)



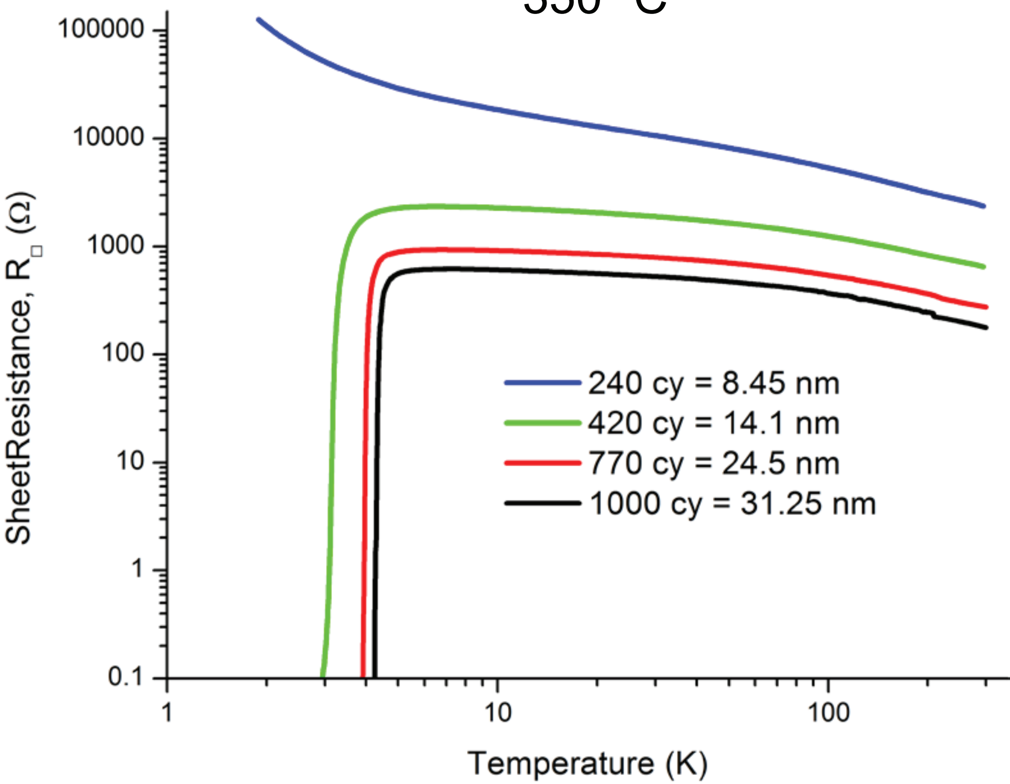
Decrease temp:
lower density
more impurities ~ 2% Cl @ 350 ^{0}C

Niobium Titanium ($Nb_{1-x}Ti_xN$) 2:1

450 ^{0}C



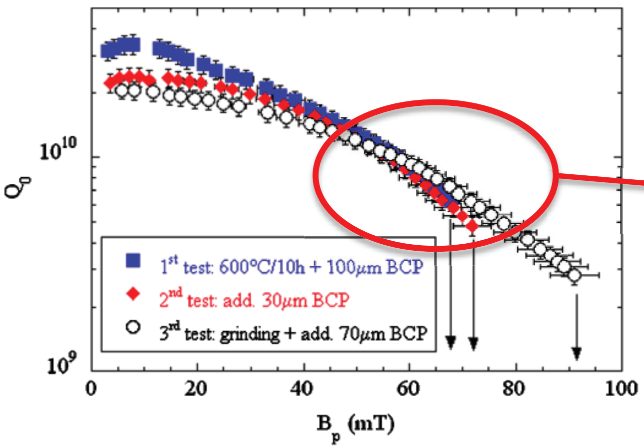
350 ^{0}C

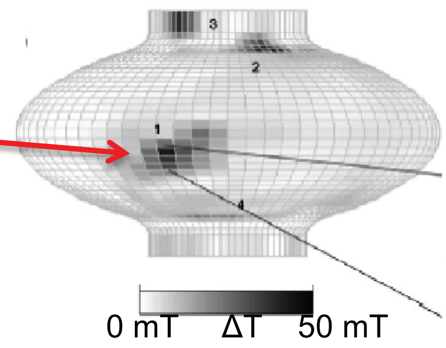


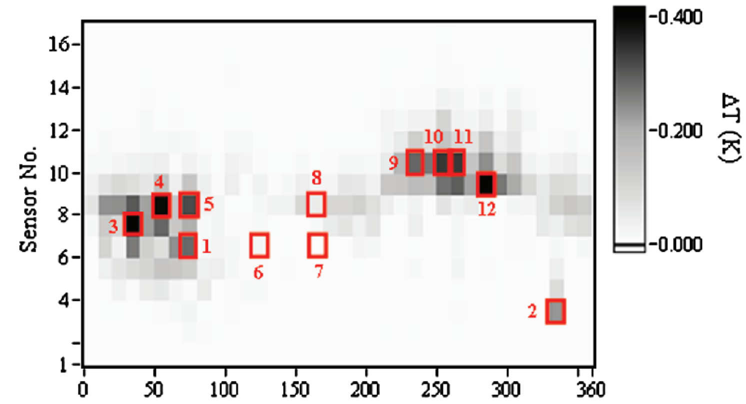
Resistivity bulk ~ 250 μΩ.cm
Tc bulk ~ 7.8 K

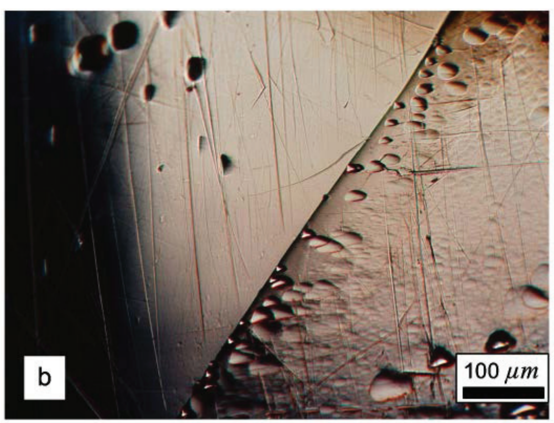
Resistivity bulk ~ 630 μΩ.cm
Tc bulk ~ 4.5 K

Point contact tunneling on hot spots from Nb SRF cavities

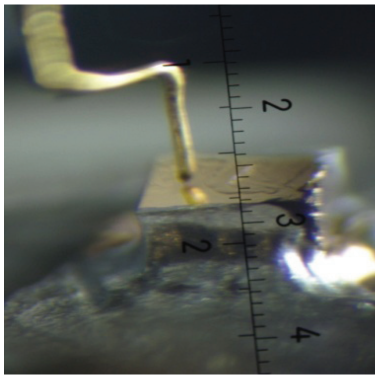


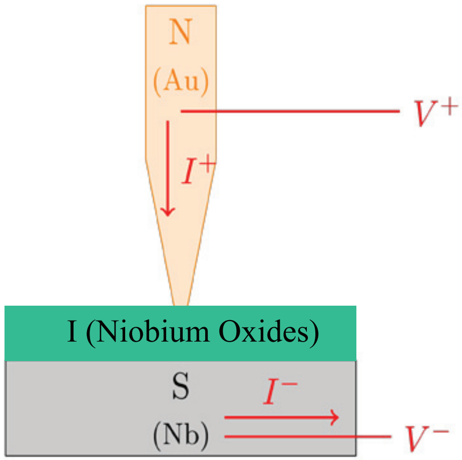






X. Zhao et al. PRSTAB 13, 124701 (2010)

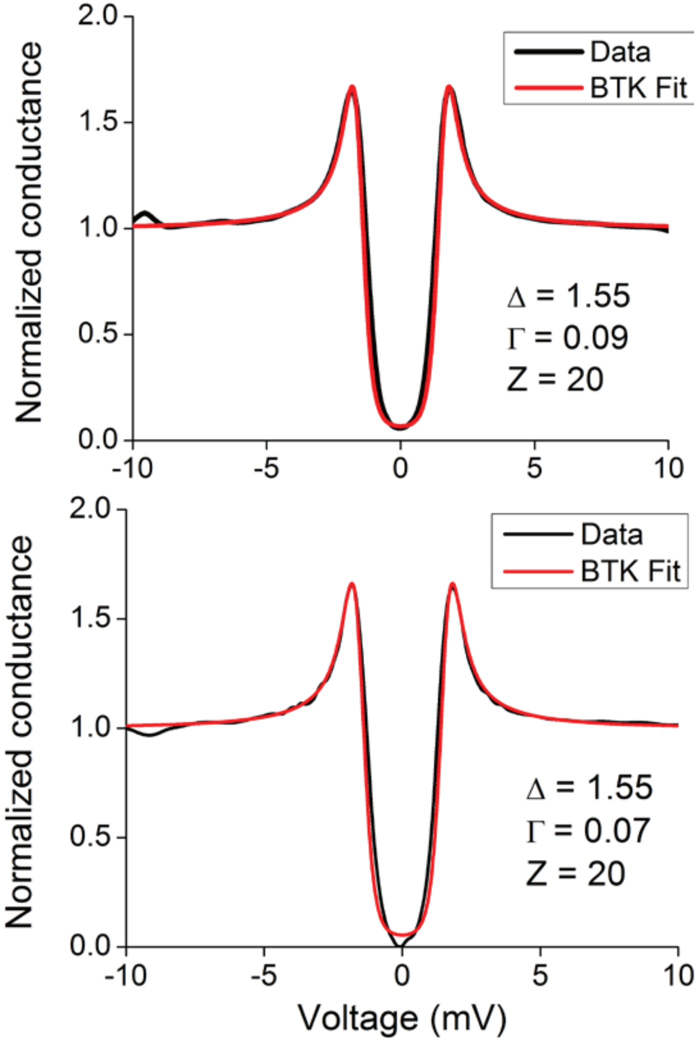


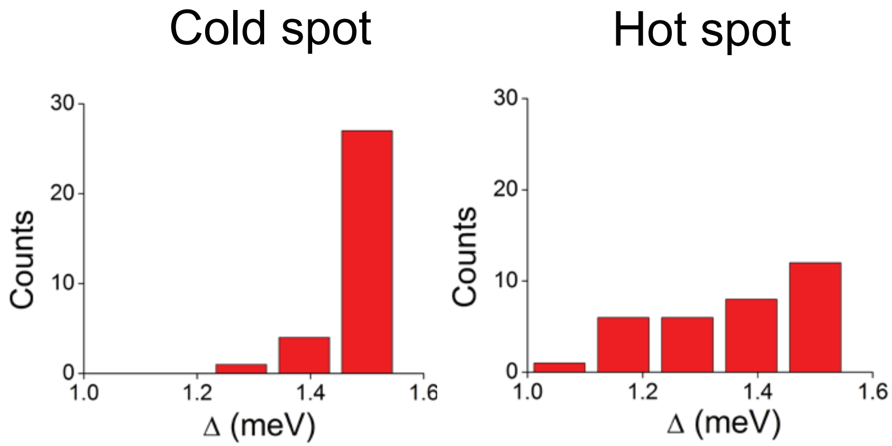


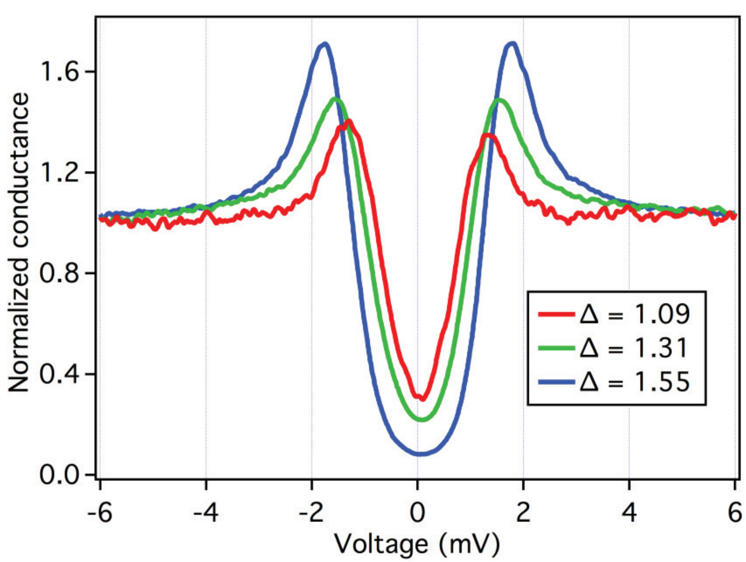
Cold spots vs Hot spots Medium Field Q slope

BTK Fit (Δ, Γ, Z)

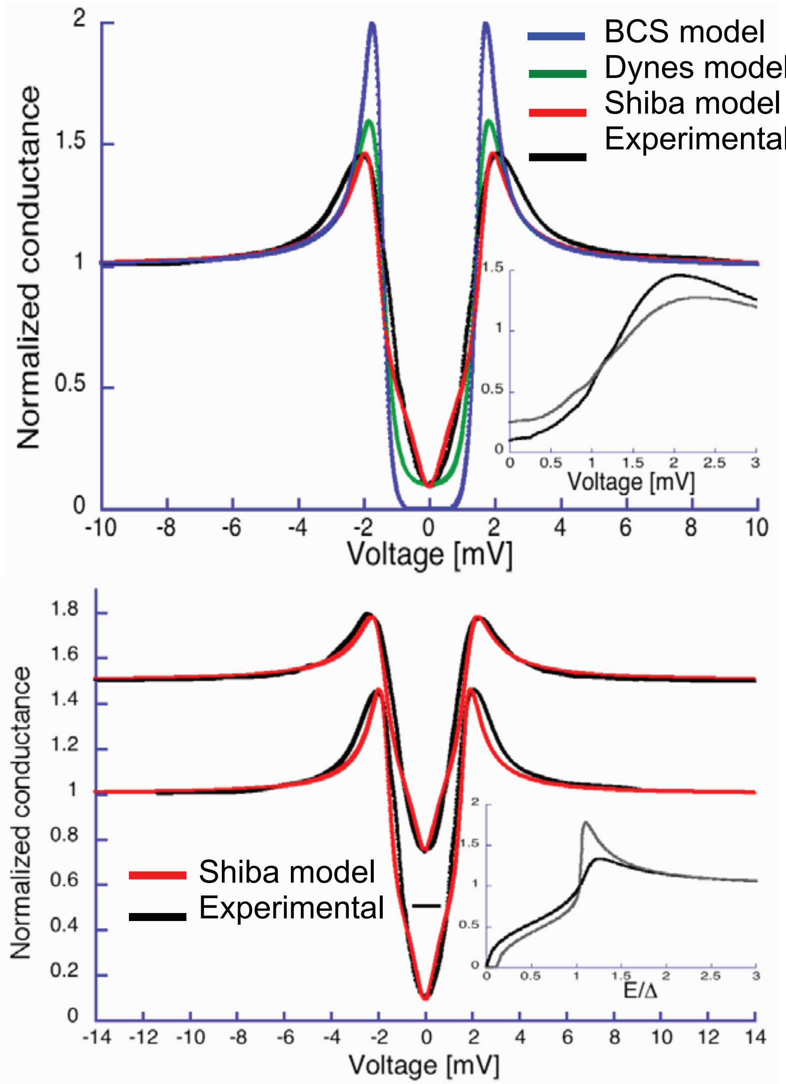
High quality spectra, Γ/Δ < 6%







surface paramagnetism on Nb

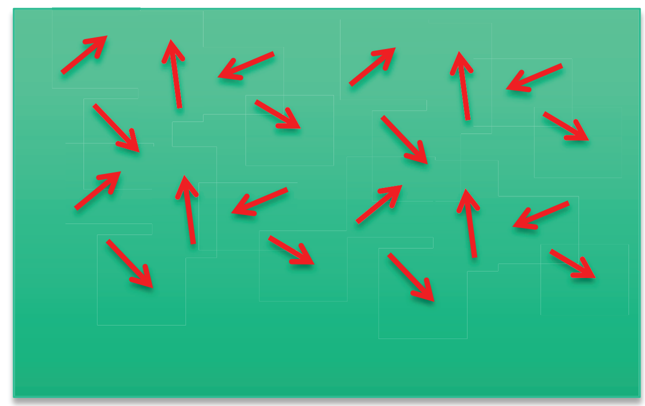


T. Proslier et al. Appl. Phys. Lett. 92, 212505 (2008)

Pair breaking parameter

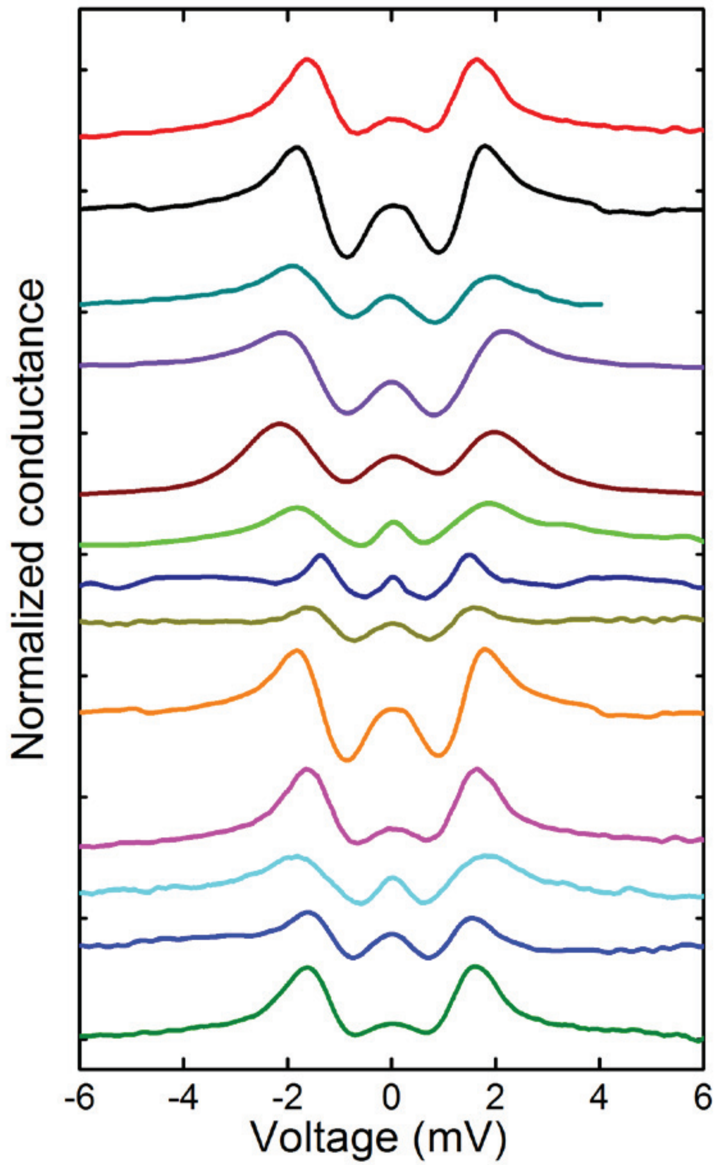
$$\alpha \sim \frac{c}{4} N(0) S(S+1) J^2 = \frac{\Gamma}{\Delta}$$

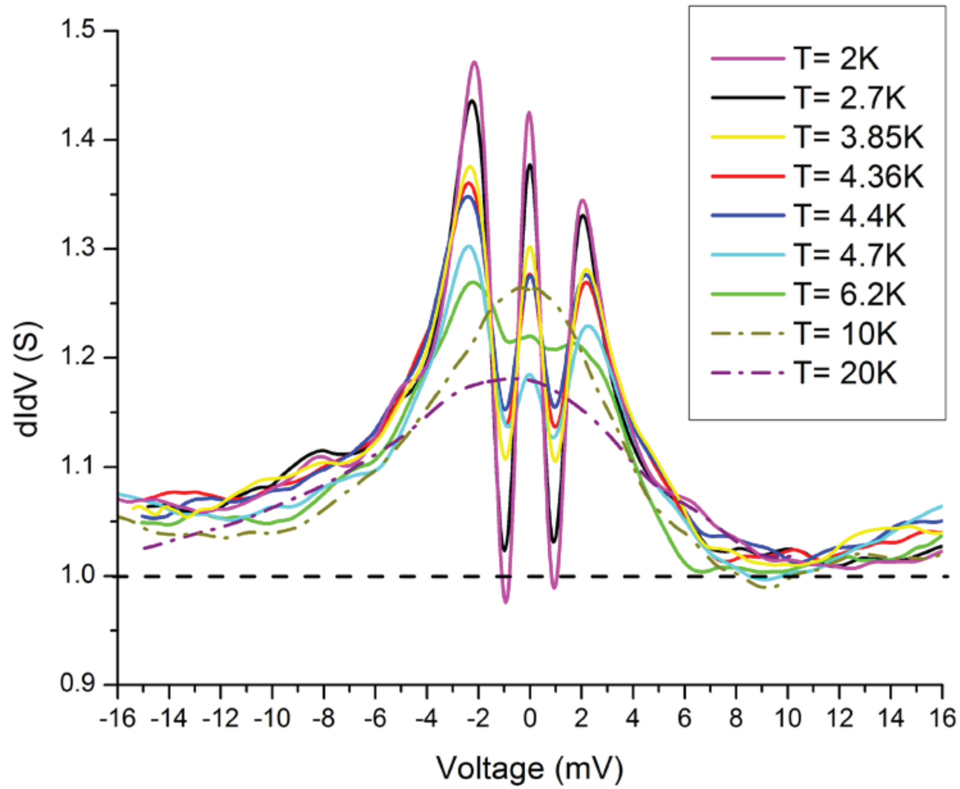
Homogenous Mag
Moments
concentration on ξ



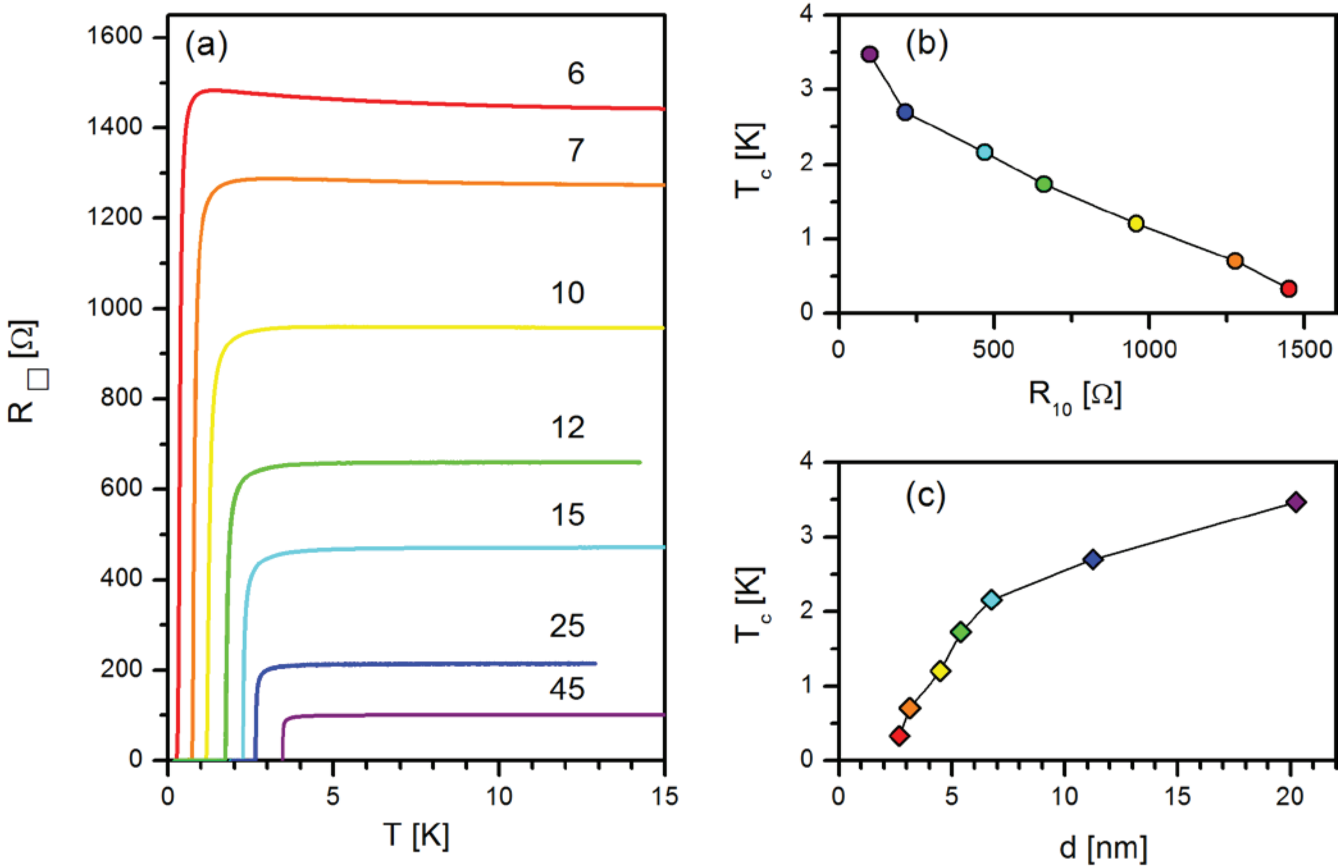
Γ/Δ ~ 0.1 to 0.3
Concentration ~ 800 ppm on ξ
$2\ 10^{17}$ Spin/m^2
SQUID -> $5\ 10^{17}$

Hot Spots
Zero bias peaks





- T_c tunable with film thickness (0.25-3.1 K)
  - ALD NbSi suitable for ultra-low temperature (down to 10 mK) transport/ 2D superconductivity studies (in progress)

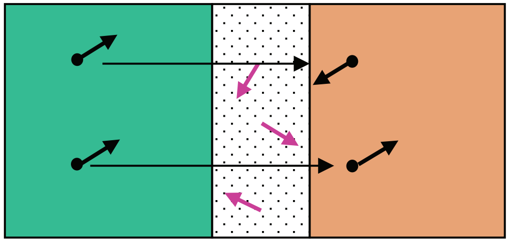


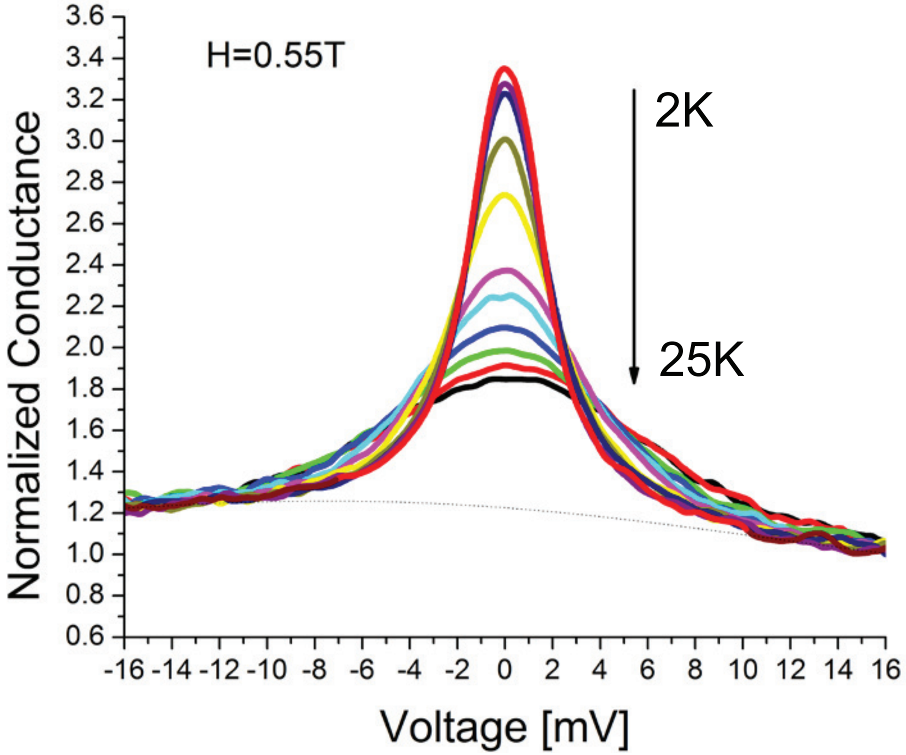
2D quantum correction start appearing for ≤ 7 cycles

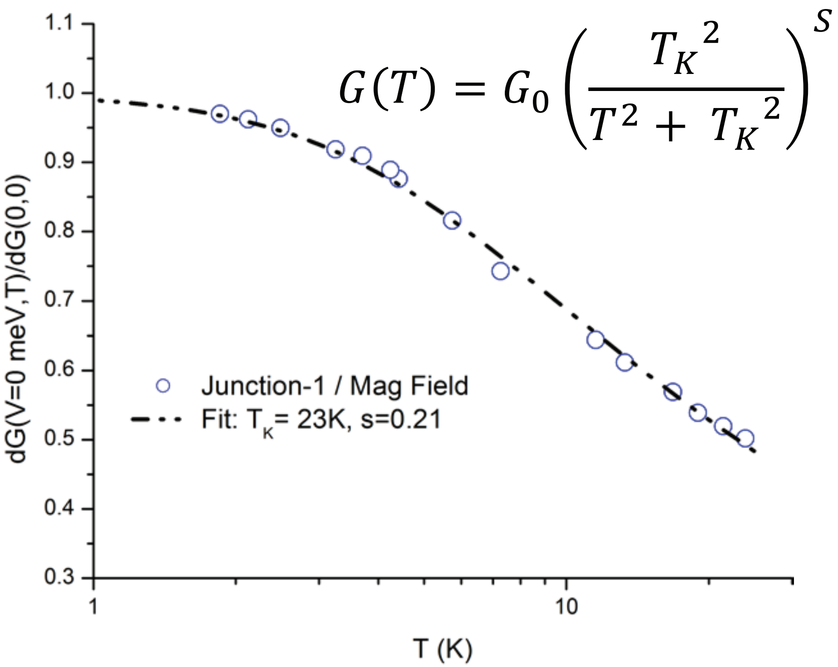
Th. Proslier, T. Baturina et al. To be published

Temperature dependence

Spin Flip (Kondo) Tunneling Channel



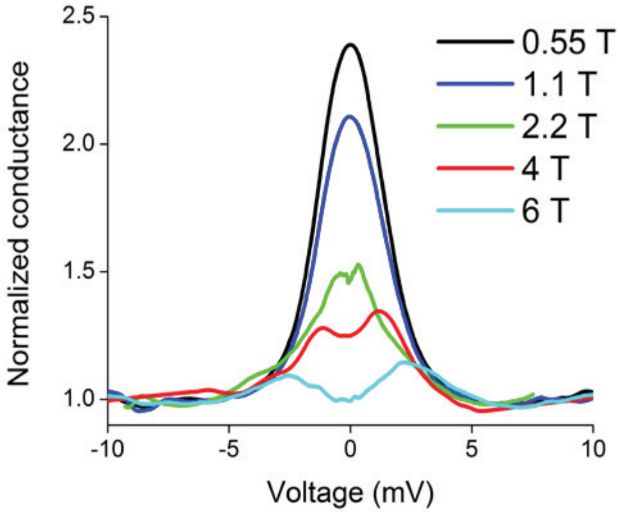


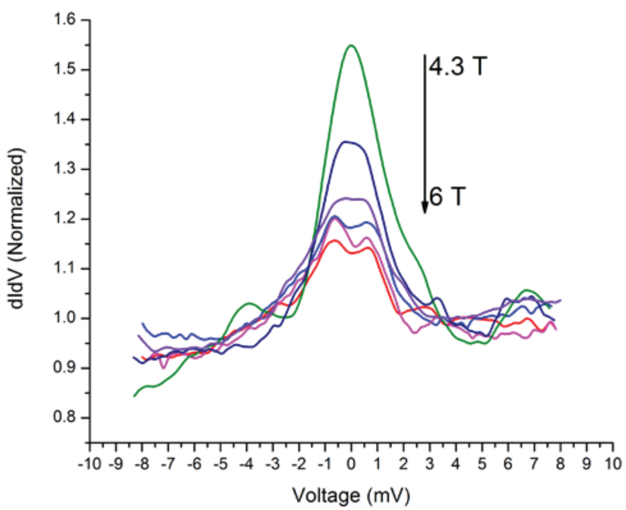


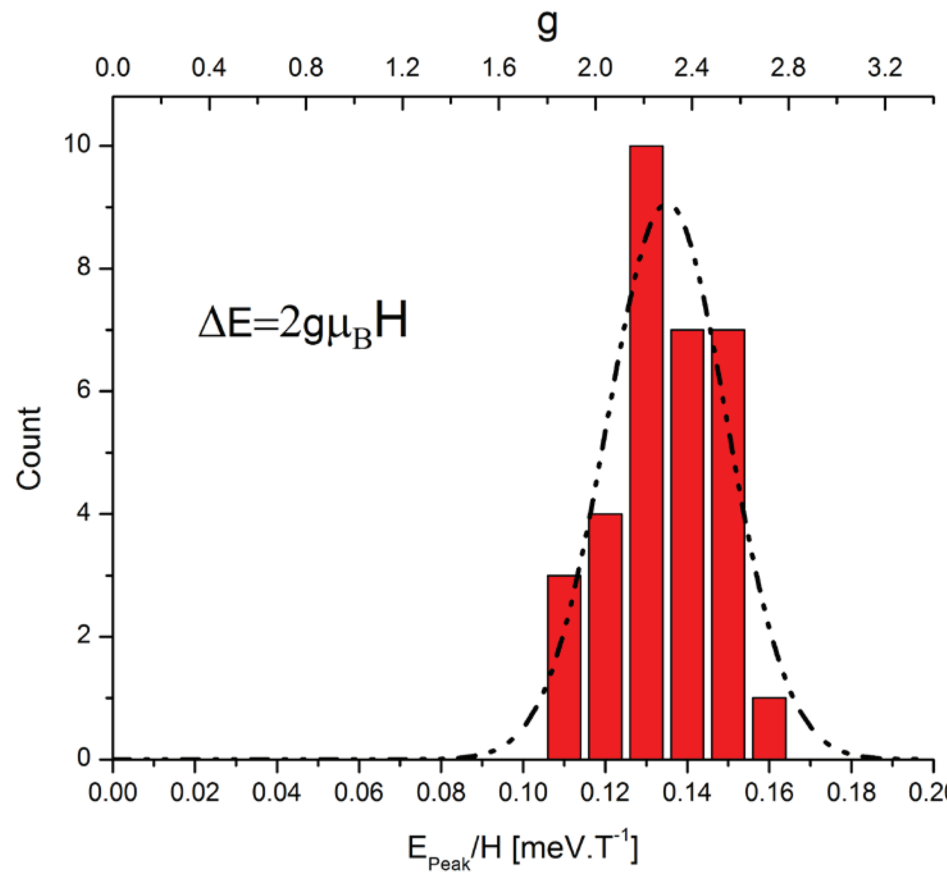
S=0.21 -> Spin ½ -> g ~ 2

T_K > T_C -> depairing superconducting state

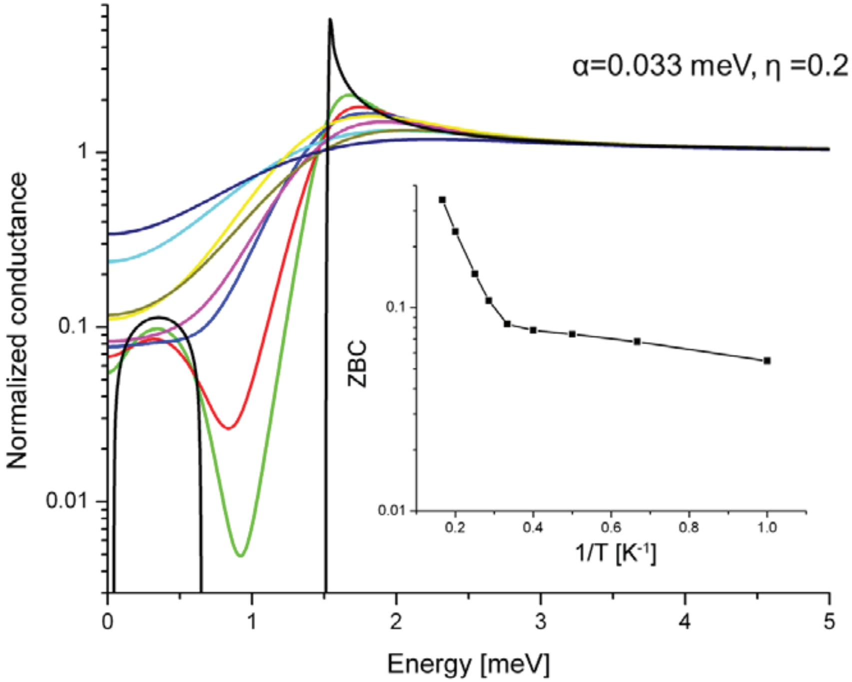
Field dependence statistic

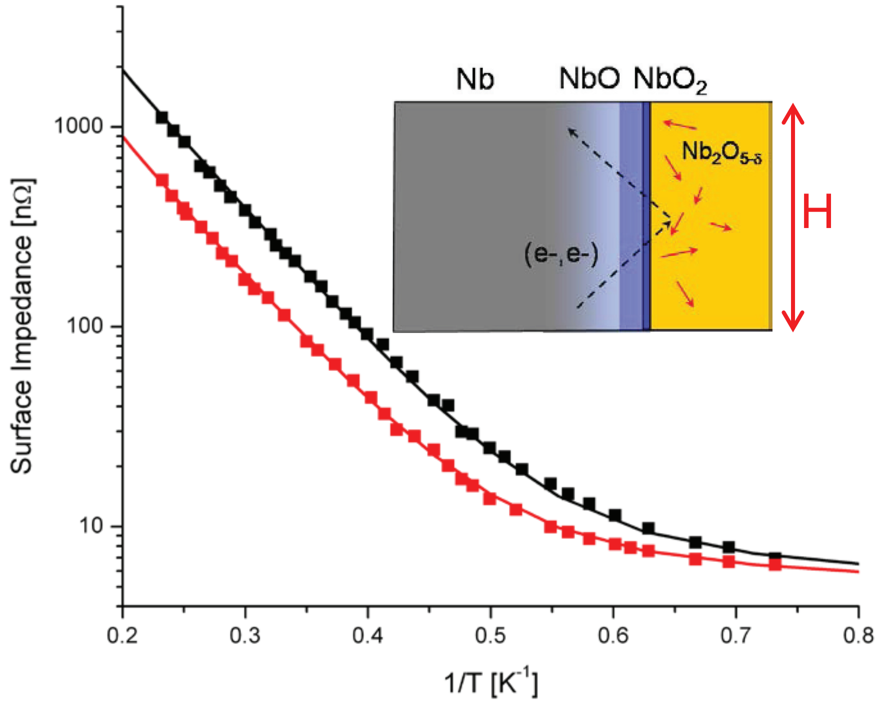






Magnetic impurities on Surface resistance





Concentration of Mag. Imp: 300 ppm -> 6.10^{16} spins/m^2

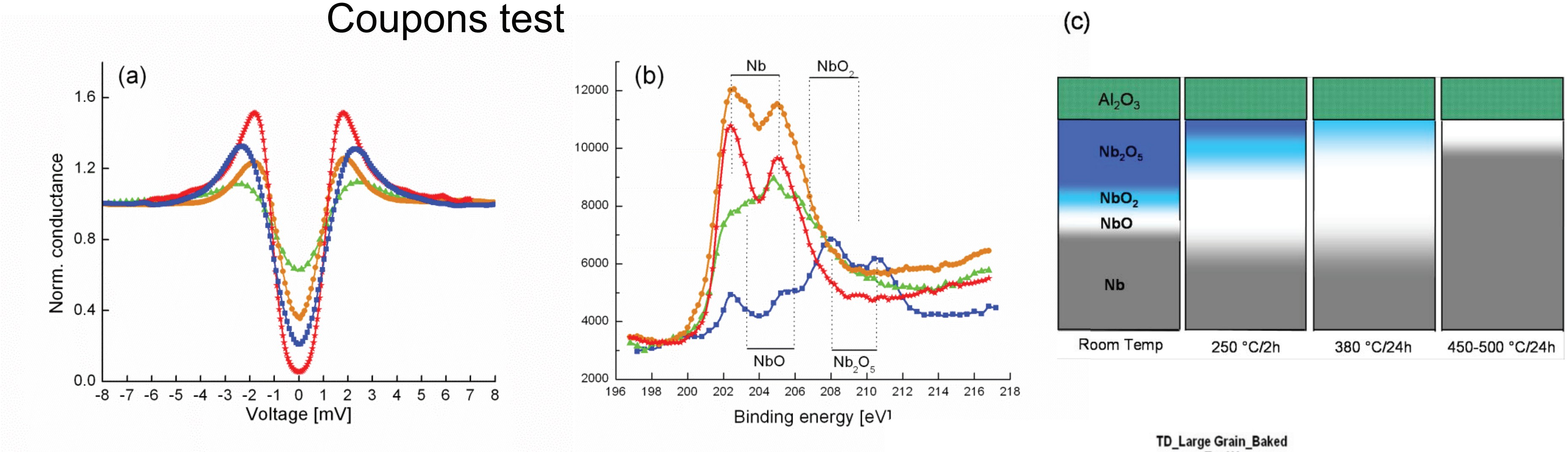
RF probing few λ ~ 40 nm -> 100 nm

Tunneling probing Few k_f or ξ from the insulating interface ?

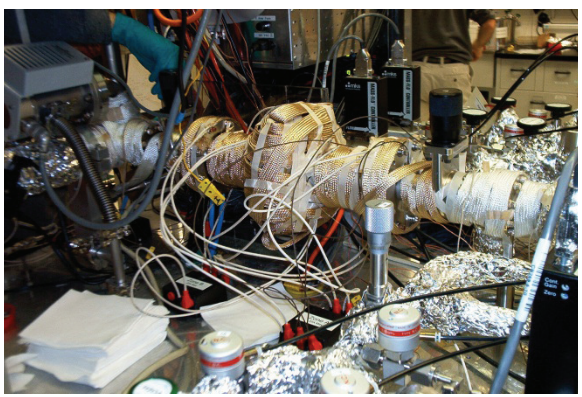
M. Kharitonov, Th. Proslier, A. Glatz, PRB 024514 (2012)

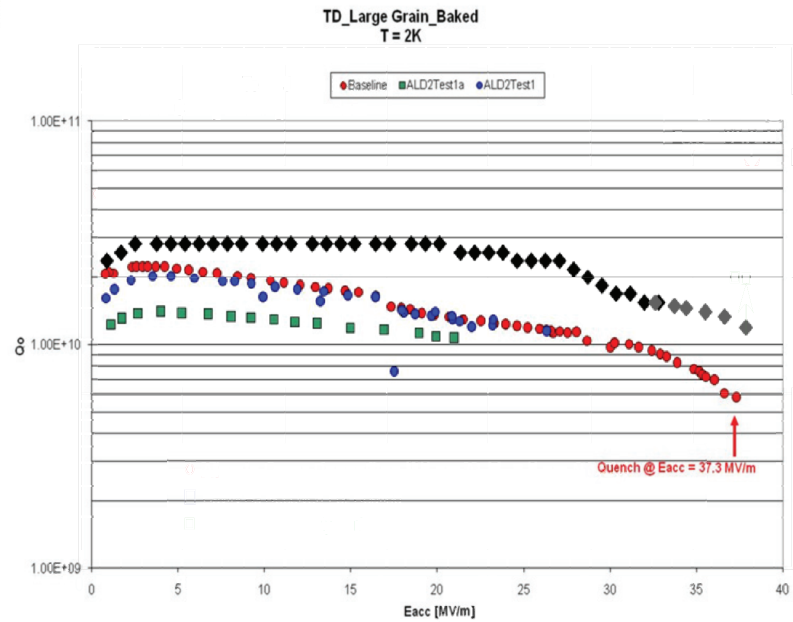
How to improve Nb?

Coupons test



Cavity test





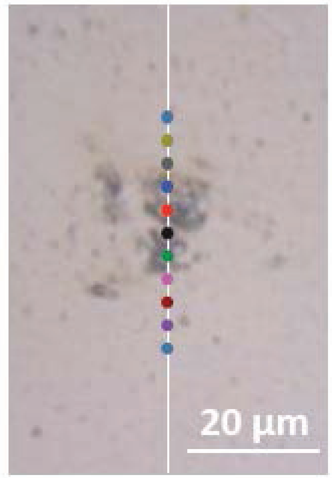
Higher Q reproducible

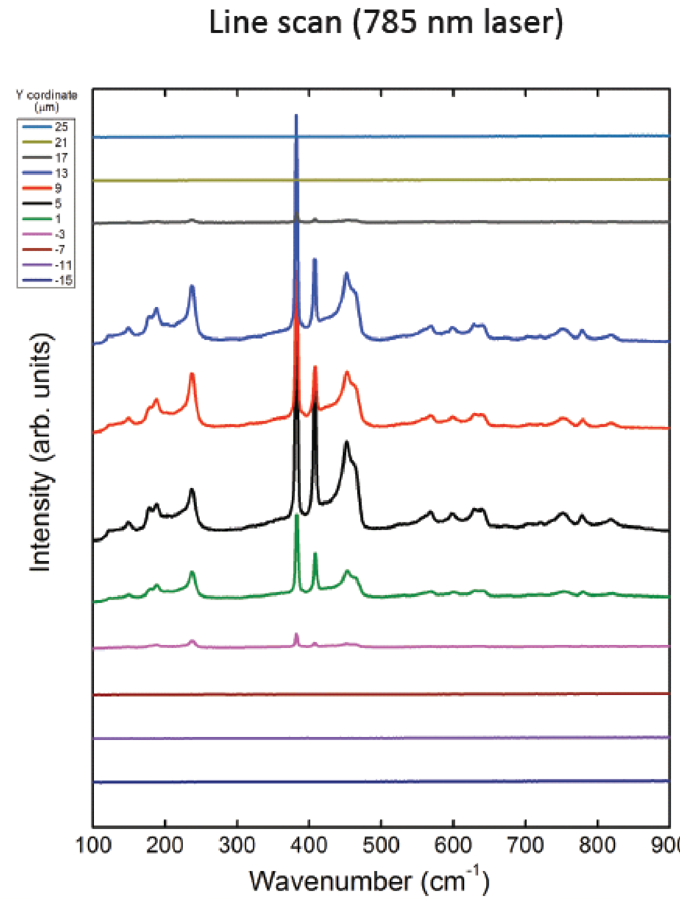
Origin of magnetism?

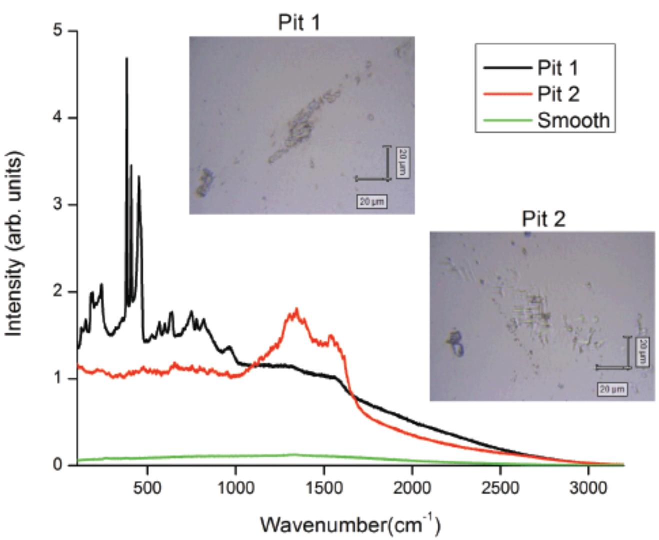
- Oxygen vacancies? Not enough
- Surface chemical composition:

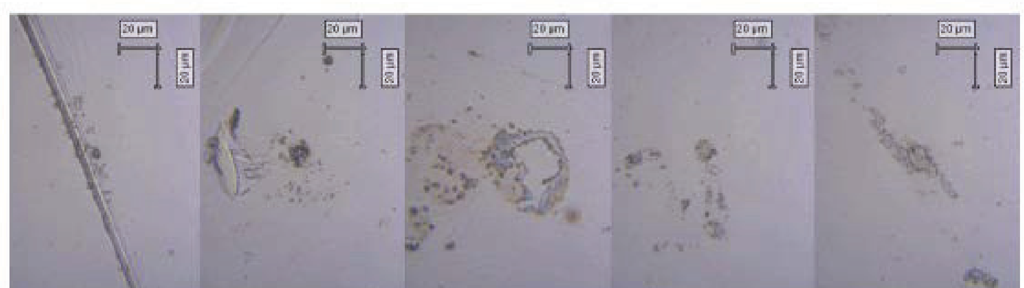
-SEM shows a lot of Carbon
-Raman shows NbC_X, Amorphous, Graphite…

- Still unclear.







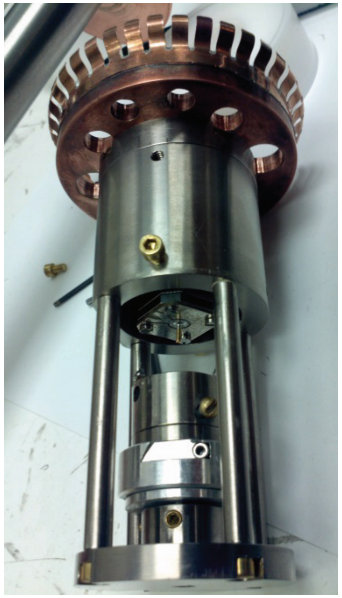


Conclusions

- Tunneling spectroscopy data correlate with RF cavities performances
  Predictive tool.
  ALD superconducting layers.

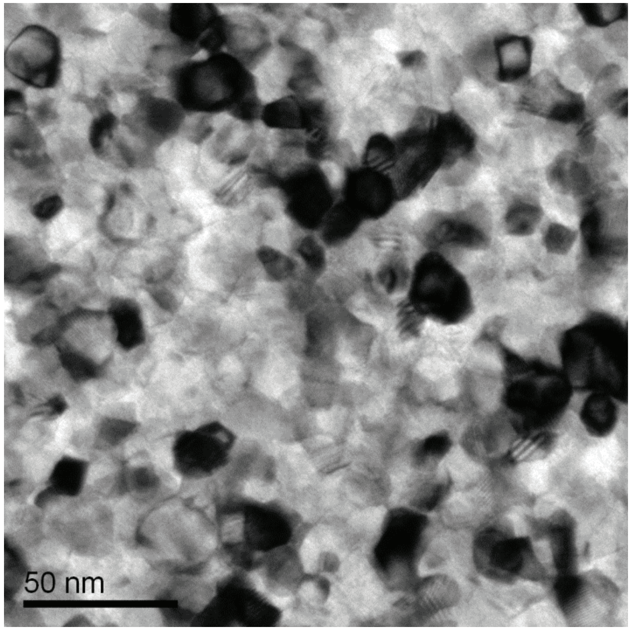
- Presence of Magnetic impurities -> Origin? (Q-bits, super-Q dots)
  Oxygen vacancies
  Raman shows C (NbC and Amorphous)

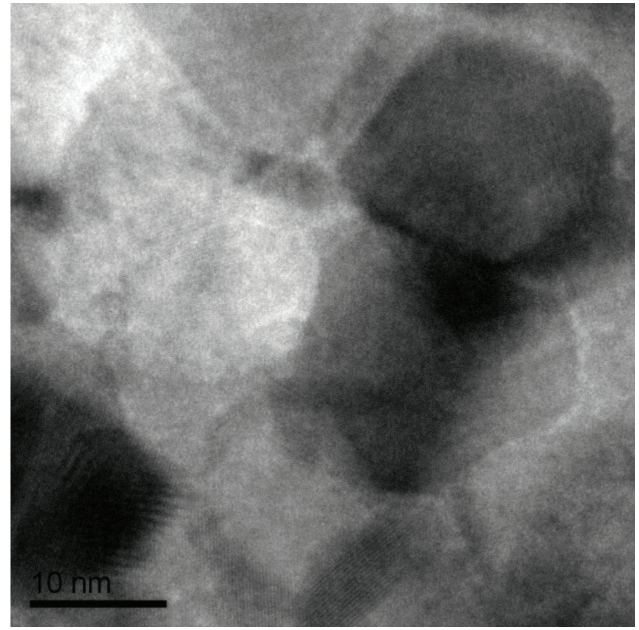
- Future:
  -Scanning x,y -> Piezo stage (tip)
  -new electronic -> Higher R_J
  -Other dielectric ALD capping

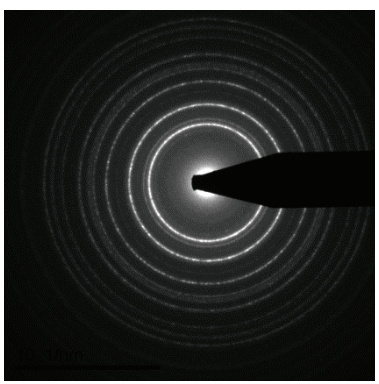


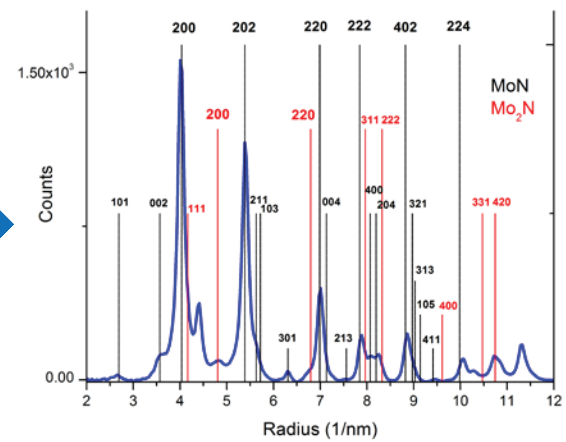
MoN: TEM

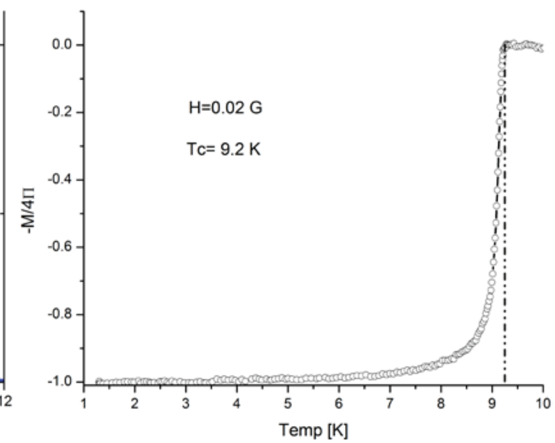
500 cy = 22 nm @ 450C





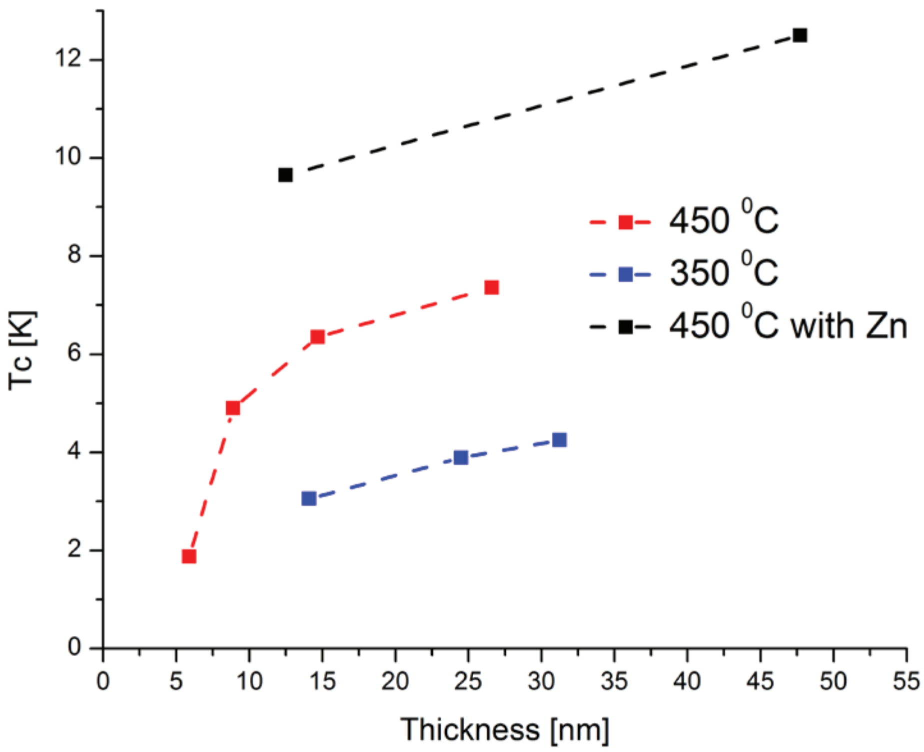


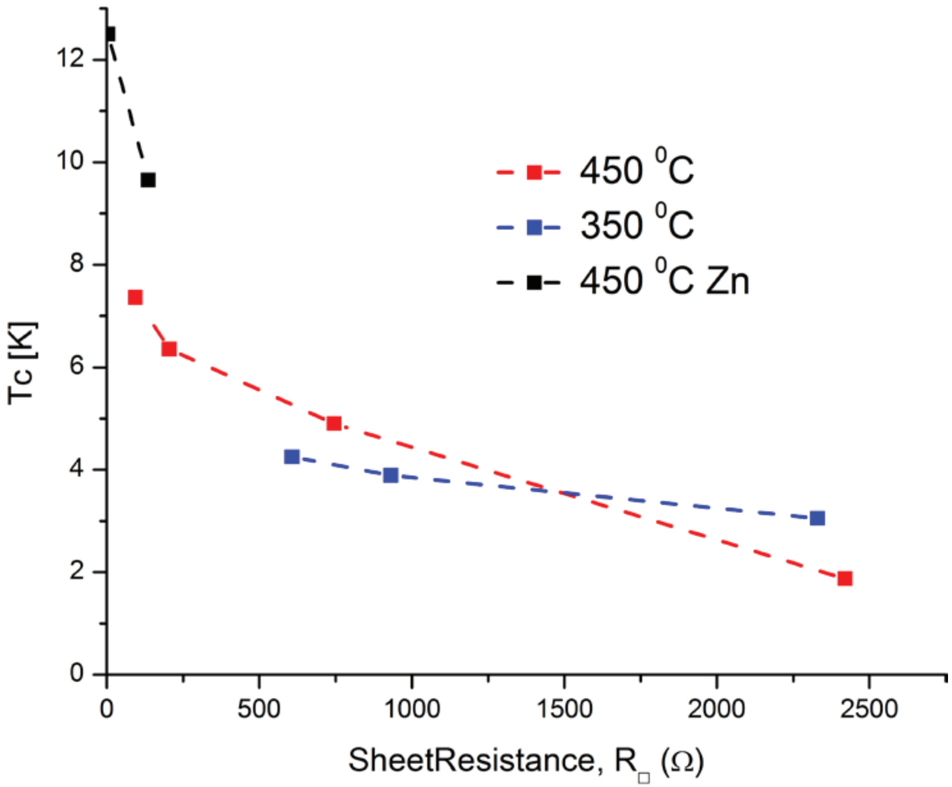




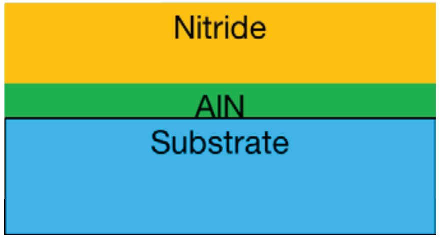
Grain size ~ 12 to 20 nm / Tc = 9.2 K

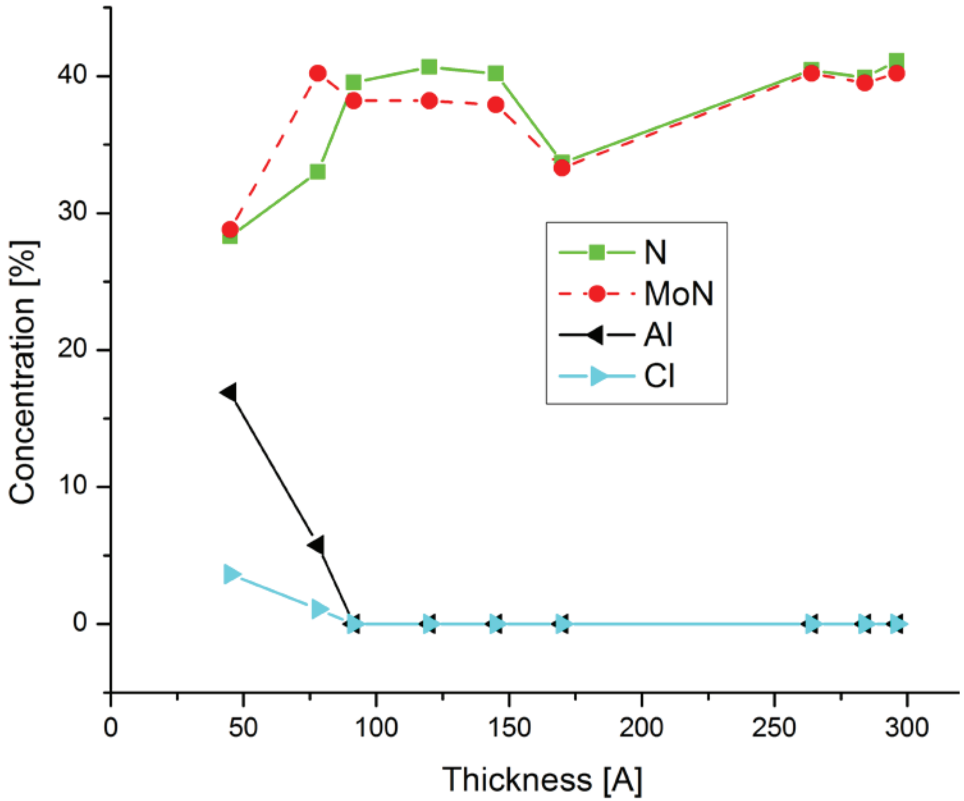
Niobium Titanium ($Nb_{1-x}Ti_xN$)

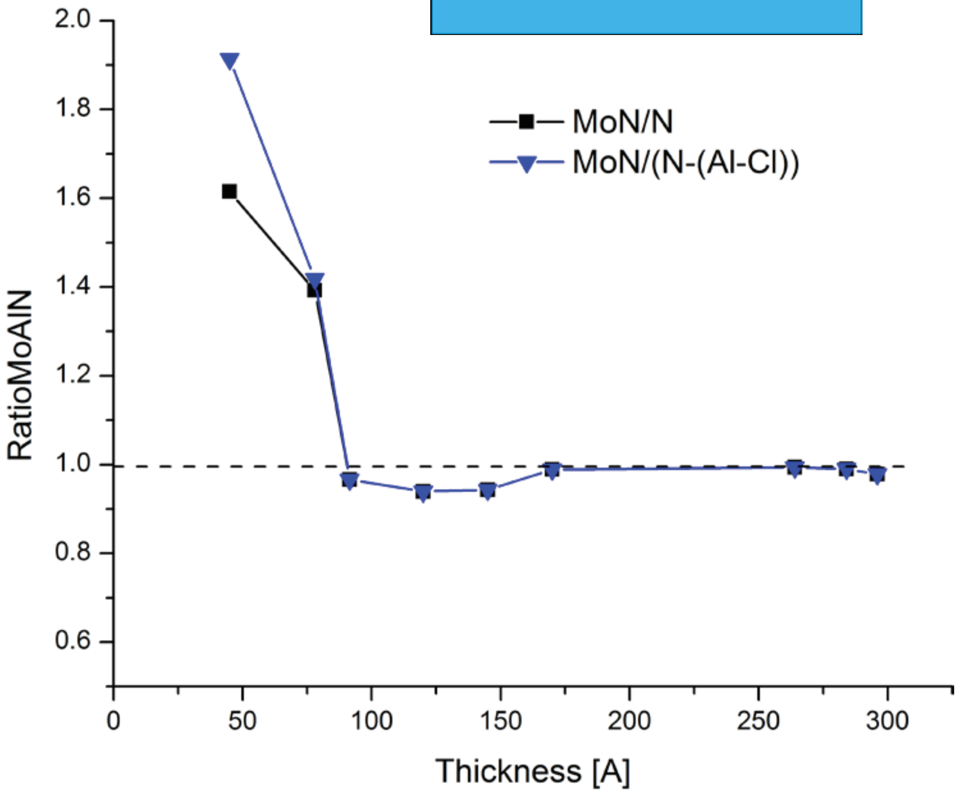




XPS: Composition Evolution with thickness



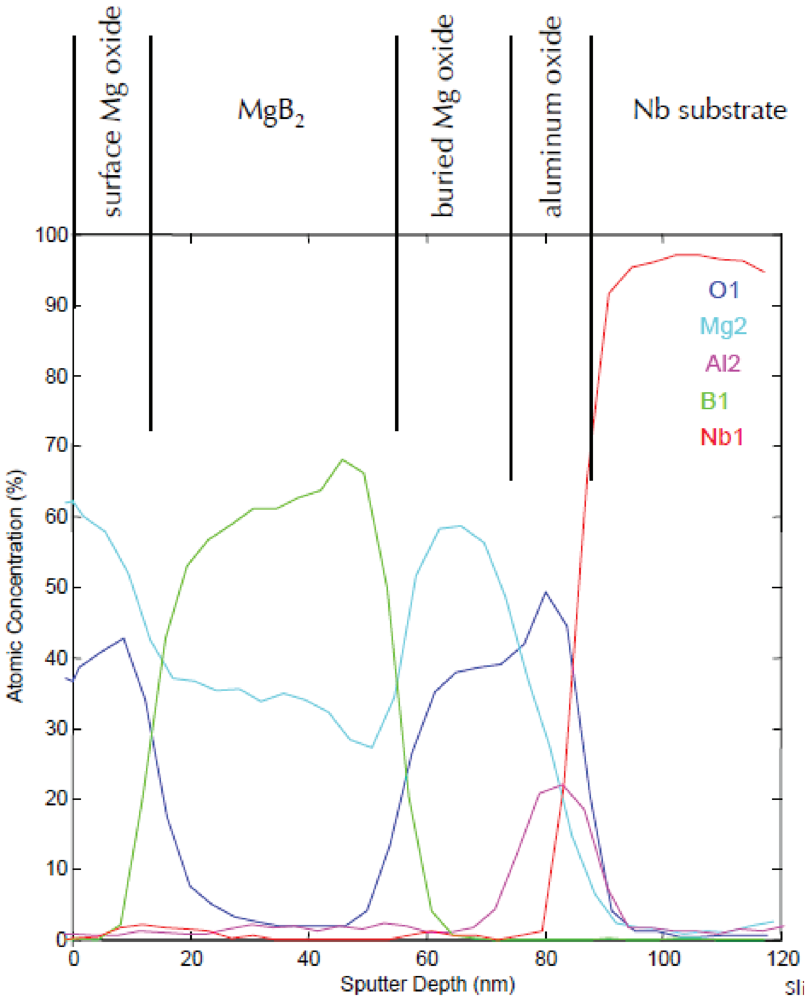




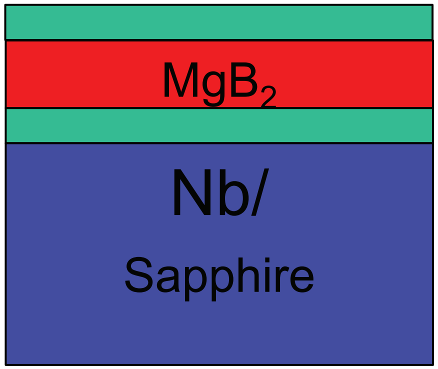
Composition: MoN 1:1 + Nucleation as Mo_2N

Sputtering change stoichiometry from MoN to Mo_2N

Multilayer structure: Which Dielectric?



Alumina coatings

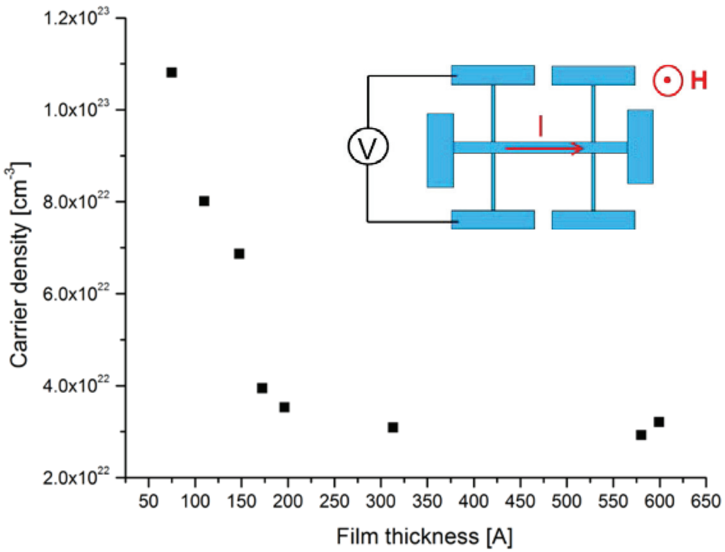


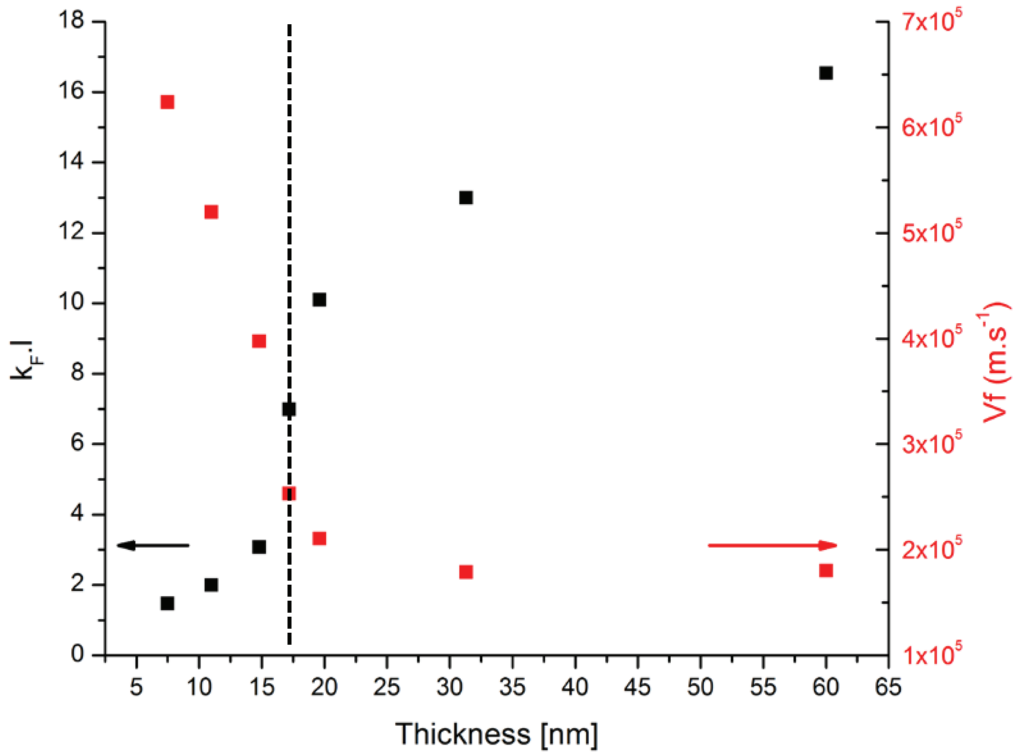
- MgB_2 dep at 600 ^oC ~ 80 nm
- ALD insulator at 300 ^oC ~ 10 nm
- Inter-diffusion !

T. Tsuyoshi and al. to be published

Hall measurement

Carrier concentration





$$k_F = (3n\pi^2)^{1/3}$$

$$V_F = \frac{l}{\tau}$$

$$l = \frac{k_F.\hbar}{(n\,e^2\,\rho)}$$

$$\tau = \frac{l^2}{3\,D}$$

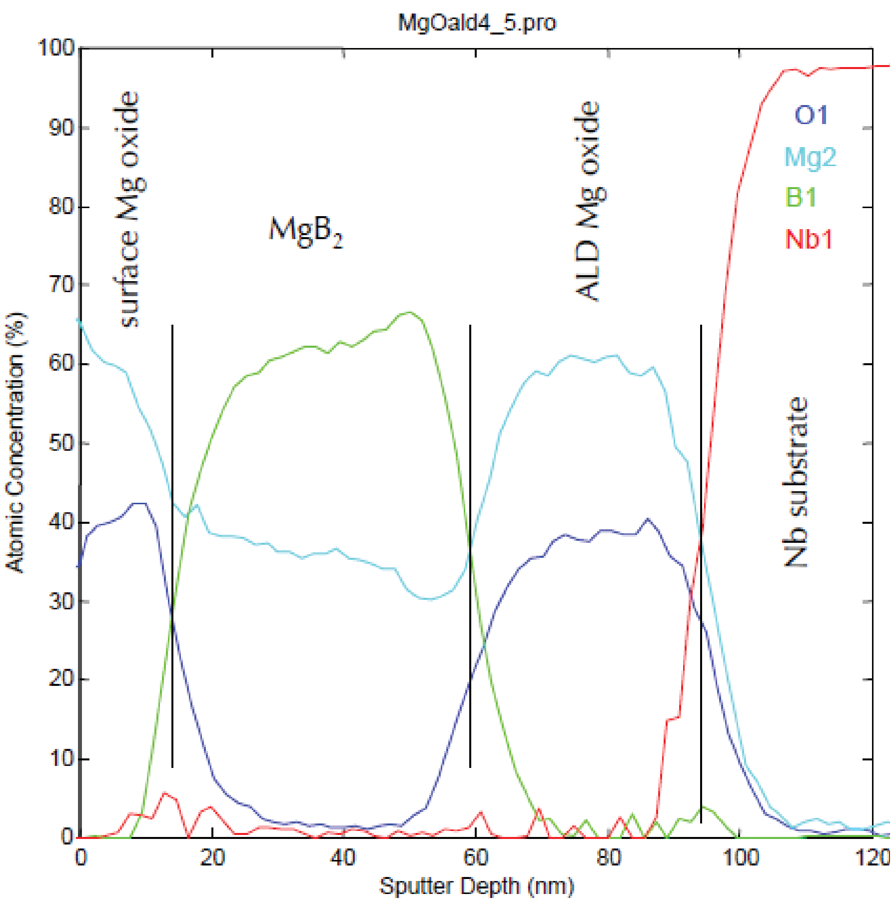
$$D = -4k_B \frac{dT_c/dH}{\pi\,e}$$

$$m_{eff} = m_e \frac{\hbar.k_F}{V_F}$$

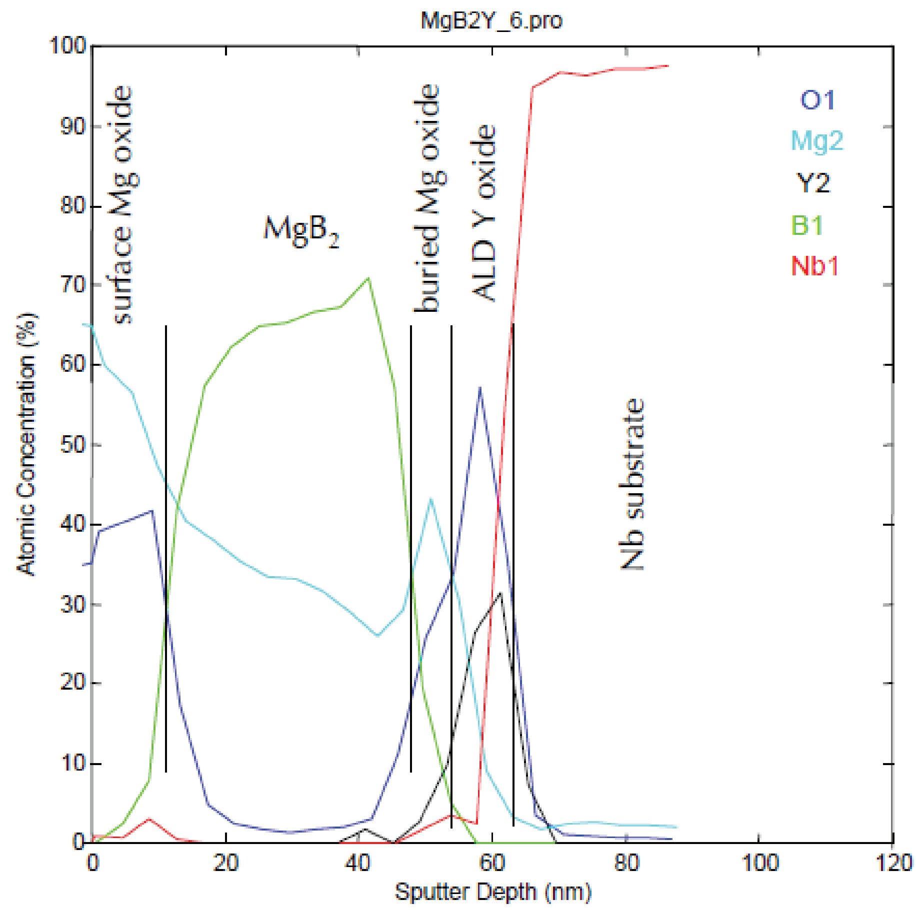
Dirty limit: ξ > l
Transition:
Strong disorder $k_F.l \sim 1$ to $k_F.l >> 1$
$m_{eff} = 6.4\ m_e$

Multilayer structure: Which Dielectric?

MgO



Y_2O_3



MgO and Al_2O_3 are amorphous
Y_2O_3 is crystalline and more stable

T. Tsuyoshi and al. to be published

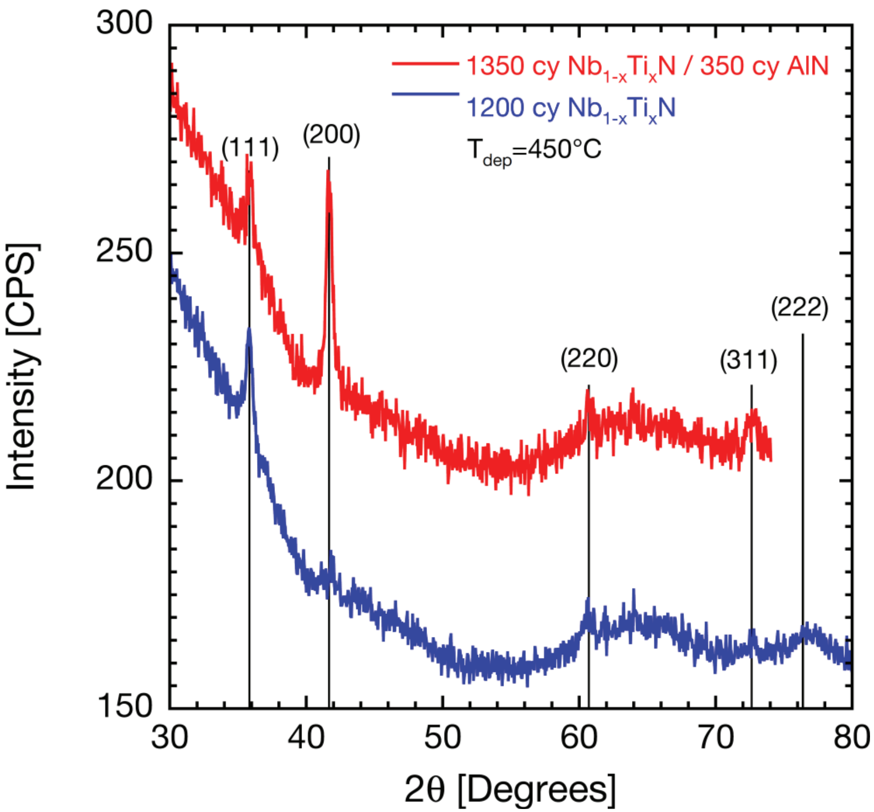
$Nb_{1-x}Ti_xN$-based superconductor/insulator heterostructures

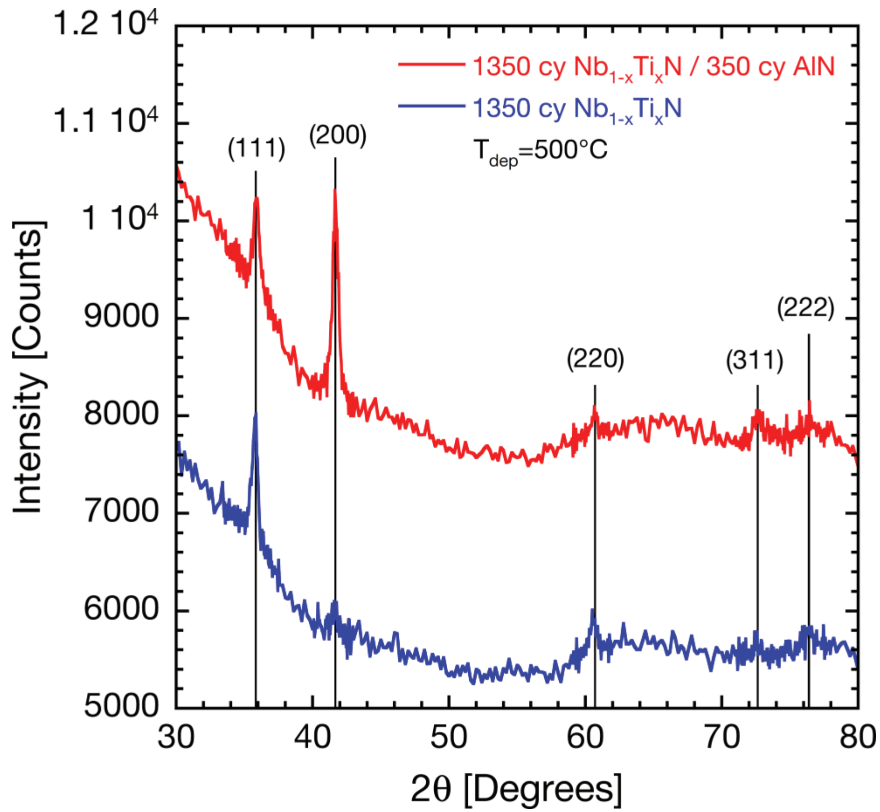
Aluminum nitride: AlN

- Oxygen-free insulator, stable interface with Nb(Ti)N
- Similar structure to Nb(Ti)N
  - 0.27% mismatch between in-plane spacing of (0001)-oriented AlN and (111)-oriented NbN
- Can be grown with $AlCl_3$ and NH_3 at same temperature as Nb(Ti)N
  - No thermal cycling between deposition steps
  - ALD previously demostrated [K.-E. Elers, et al. *J. de Phys. IV* **5** (1995)]
- NbN/AlN multilayers grown previously by sputtering
  - Enhanced J_c at high fields [J.M. Murduck, et al. *Appl. Phys. Lett.* **62** (1988)]
  - Model system for vortex matter in HTS [E.S. Sadki, et al. *Phys. Rev. Lett.* **85** (2000)]

$Nb_{1-x}Ti_xN$ / AlN: X-ray diffraction

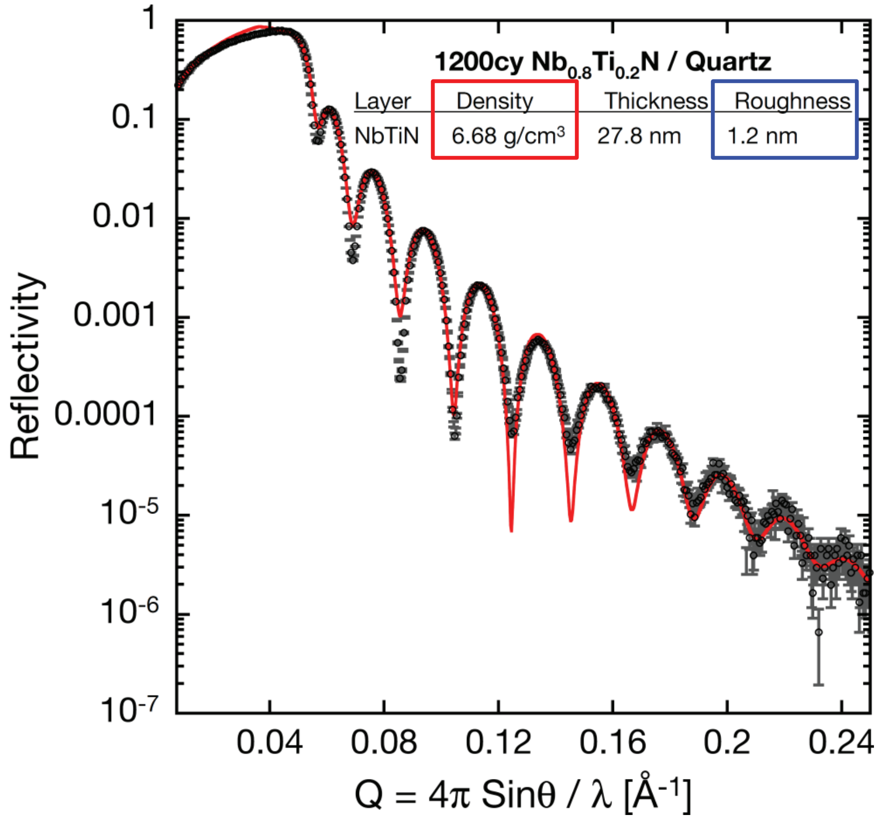
- AlN changes $Nb_{1-x}Ti_xN$ orientation

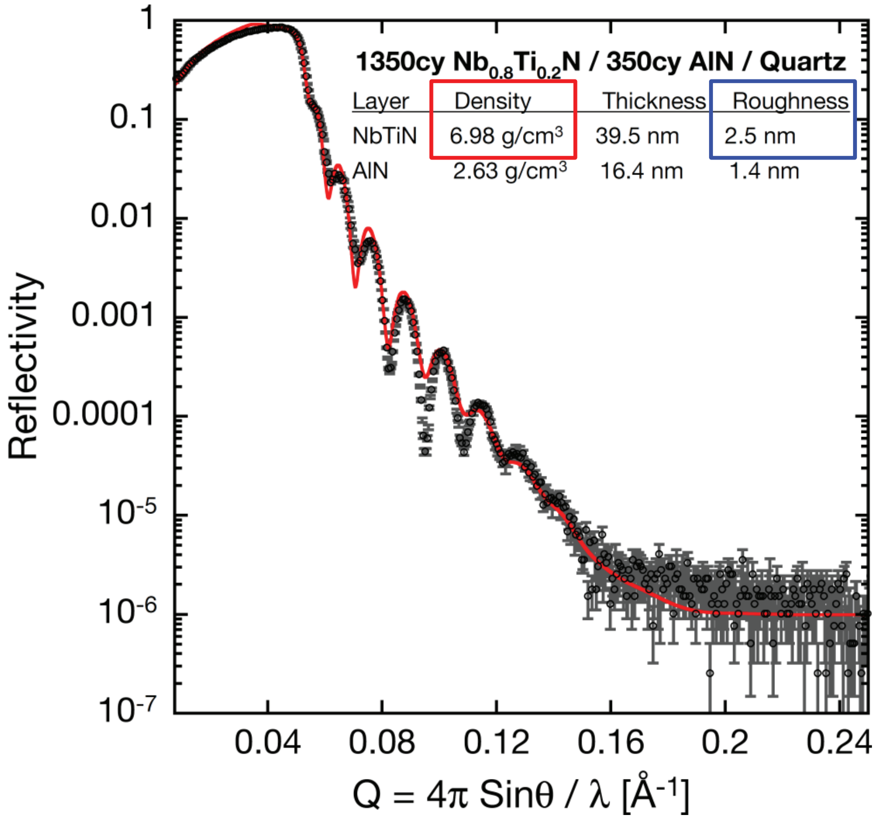




- With AlN, total integrated intensity increases by ~2x
- Film thickness/growth rate?

$Nb_{1-x}Ti_xN$ / AlN: X-ray reflectivity

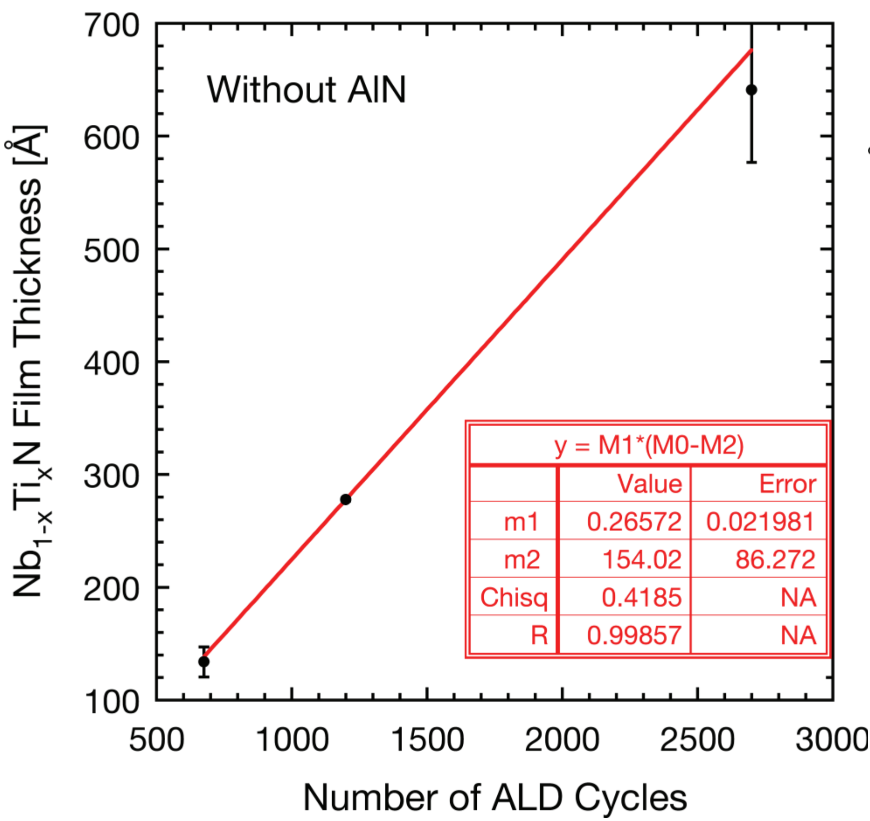


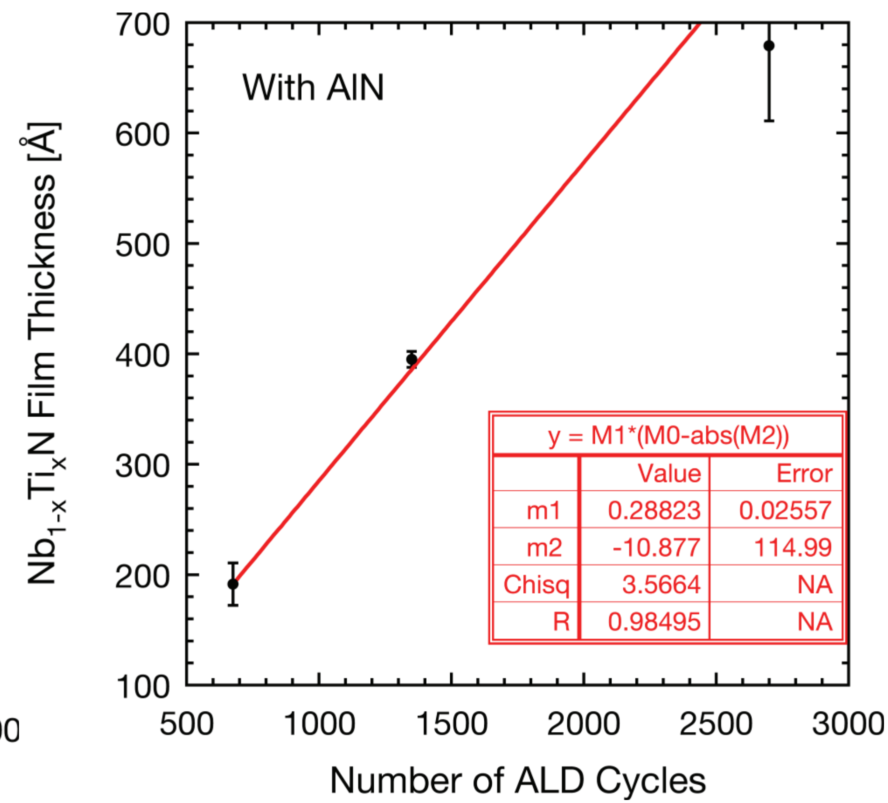


- Density ~5% higher with AlN
- Roughness ~2x higher with AlN
- Change in thickness/cycles

$Nb_{1-x}Ti_xN$ / AlN: Growth rate

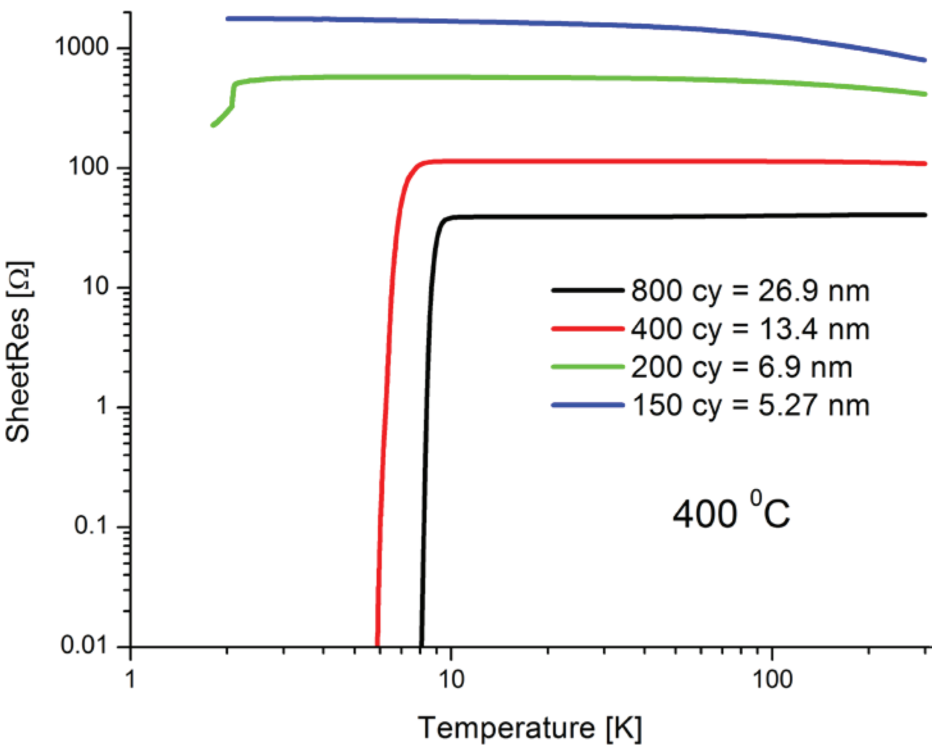
- Thickness/cy change → difference in nucleation delay
  - G.R. ~0.27 Å/cy in both cases
  - Delay 100-200 cycles on bare quartz

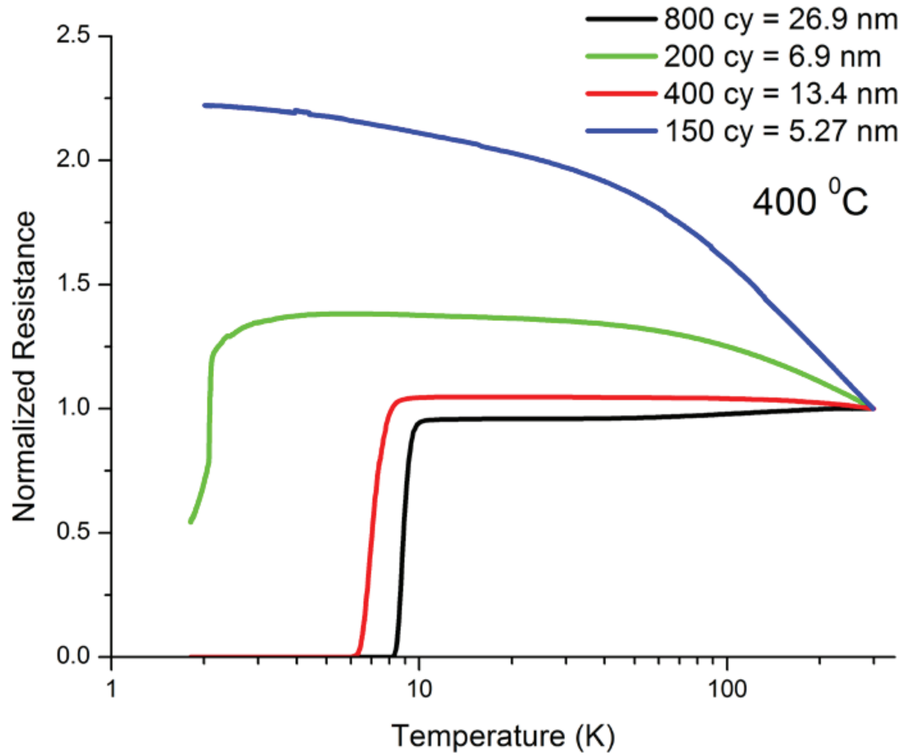




Transport: Evolution with thickness

400 ^{0}C / H=0T

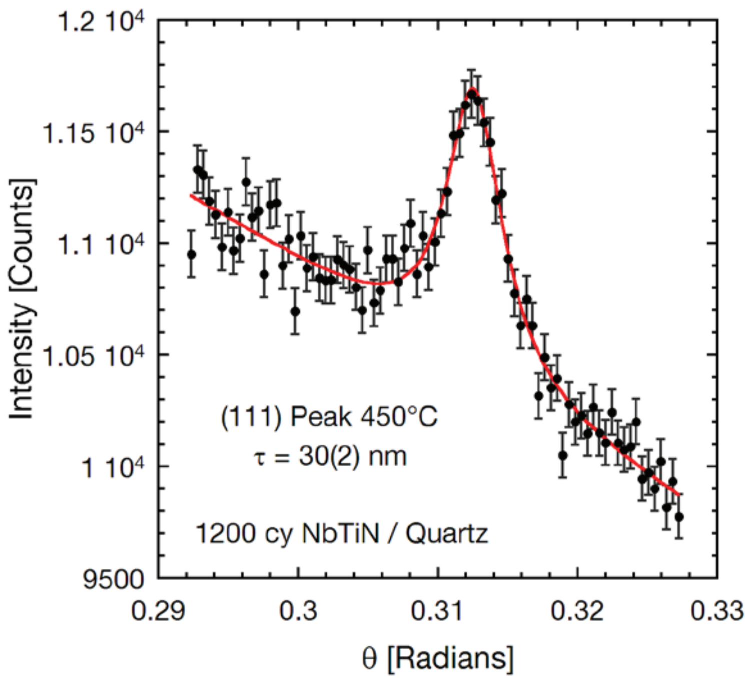




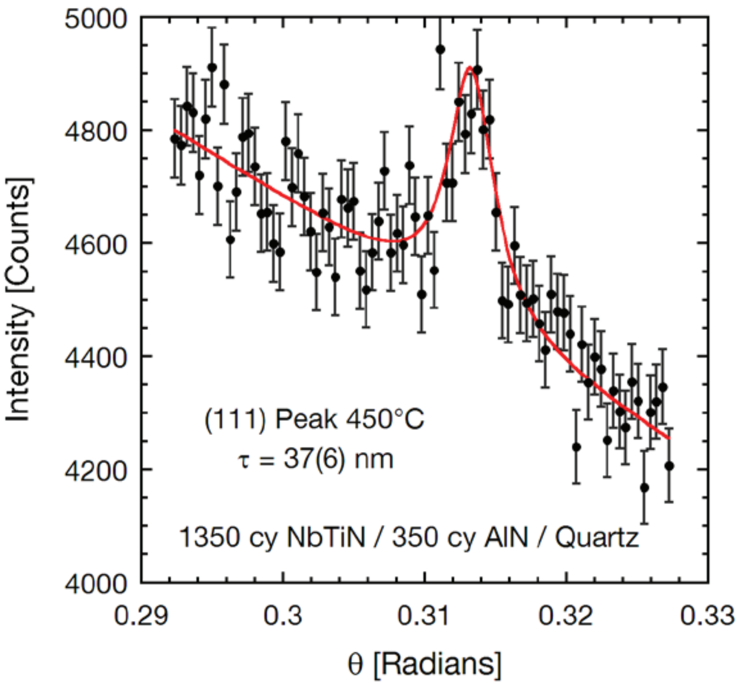
Resistivity bulk ~ 110 μΩ.cm
Tc bulk ~ 8.6 K

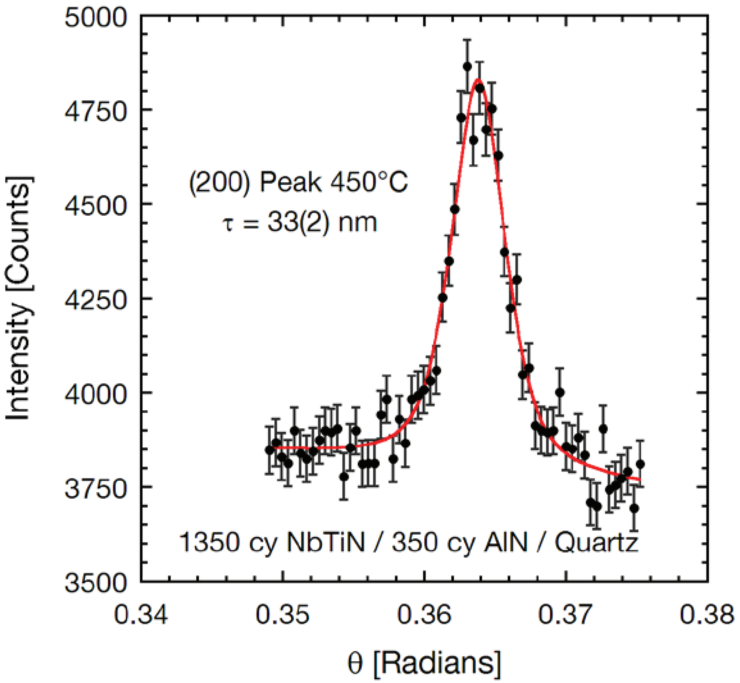
$Nb_{1-x}Ti_xN$ / AlN: Grain size

- No significant difference in XRD grain size (Scherrer)

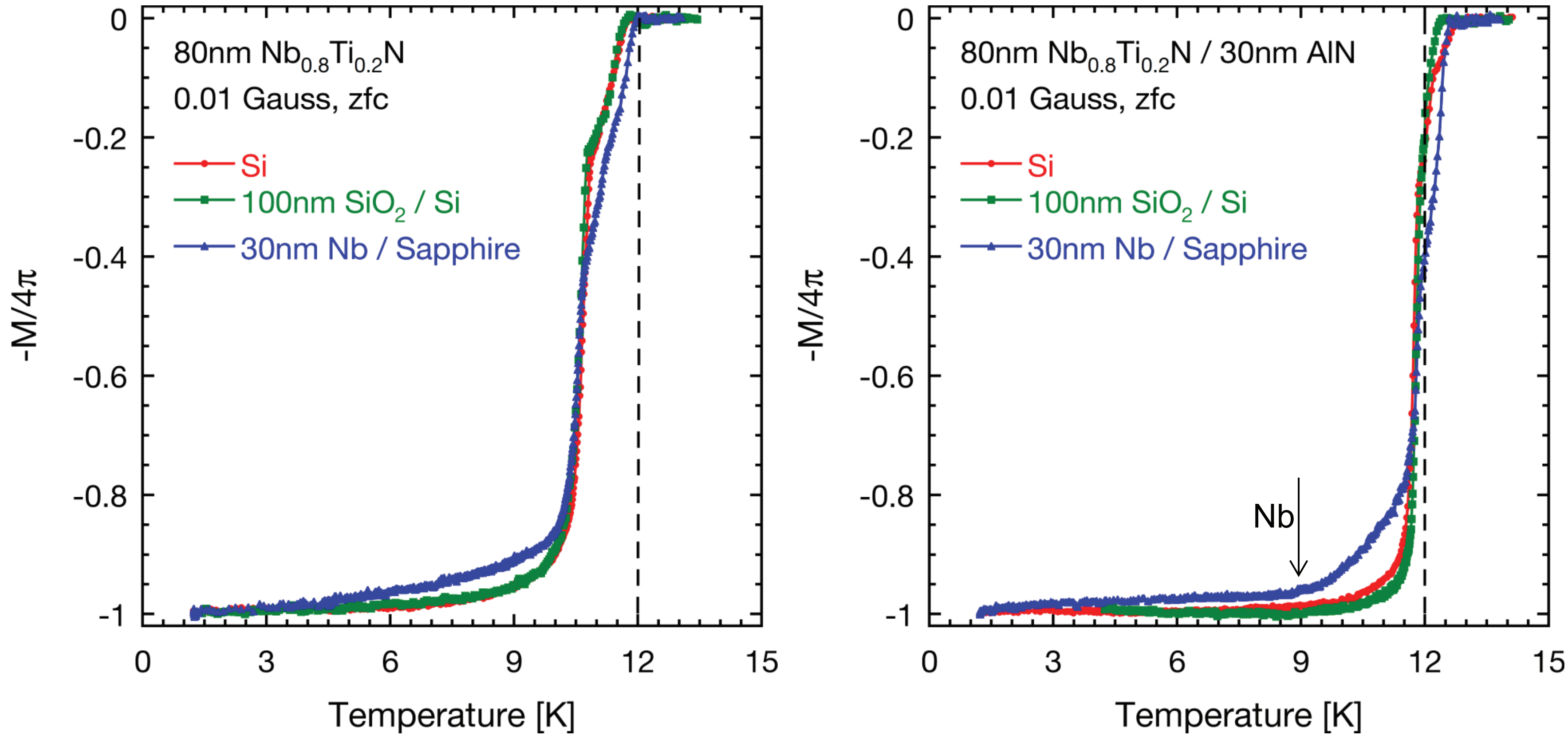


- TEM studies are underway





$Nb_{1-x}Ti_xN$ / AlN: Superconducting T_c



J. Klug, T. Proslier et al. to be published

$Nb_{1-x}Ti_xN$: Ongoing work

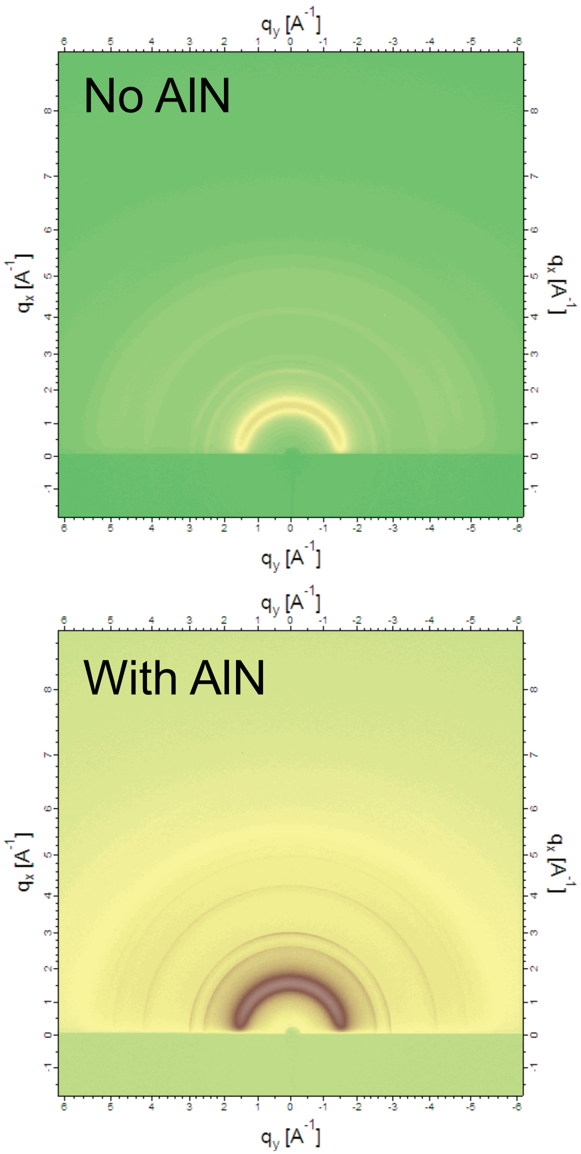
- Synchrotron grazing incidence XRD measurements
  - Depth dependence of structure, texture, and strain.
- Comprehensive compositional analysis
  - RBS, XPS
  - Study effects of deposition temperature, etc.
- High resolution TEM studies of S-I multilayers
  - Grain structure
  - Interface morphology

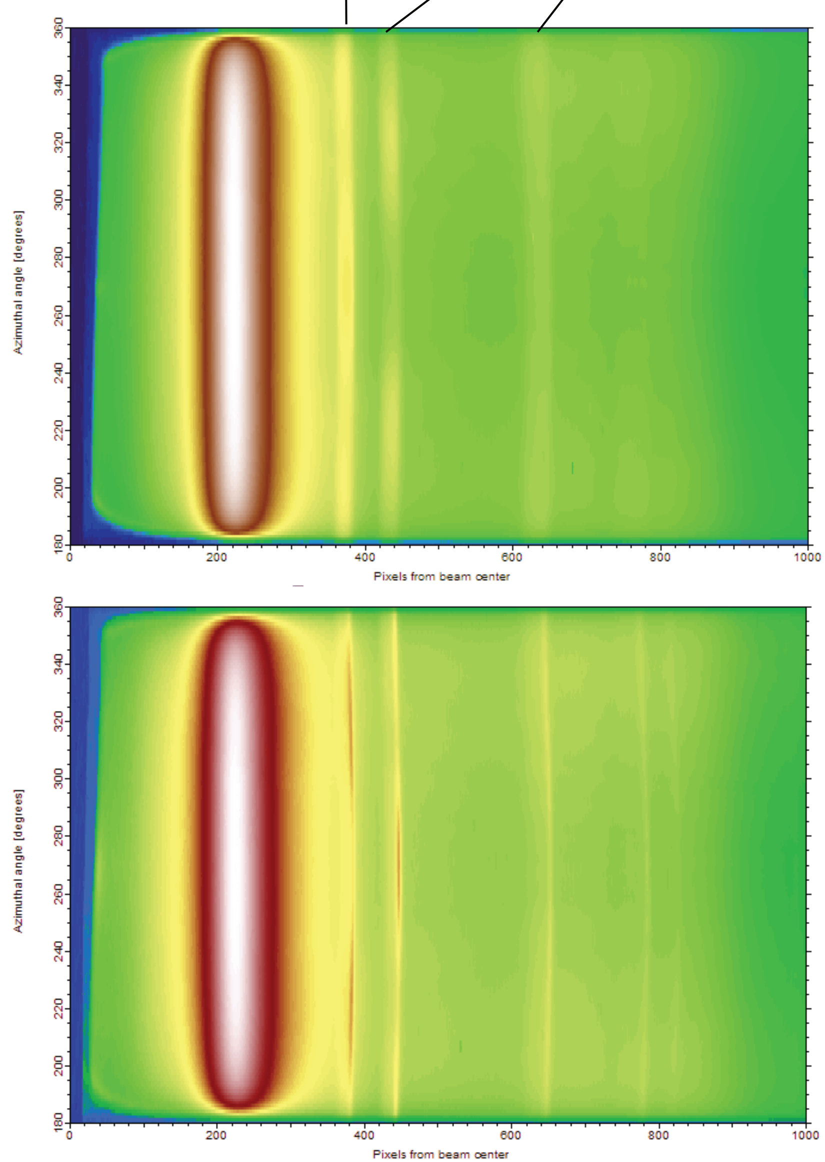
J. Klug, T. Proslier et al. to be published

- Construction of UHV ALD chamber for coating single-cell Nb SRF cavities

- Plasma ALD system.

Extra slide 1: $Nb_{1-x}Ti_xN$ GIXRD

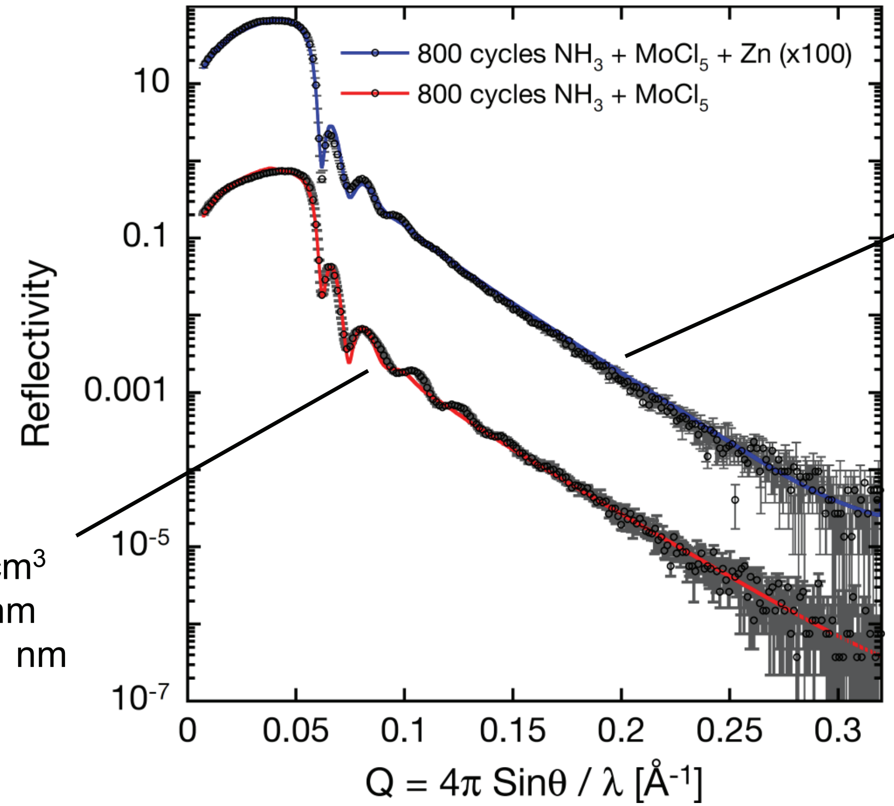




Extra slide 2: Molybdenum nitride

Effects of intermittent Zn pulse at 450°C

- 6% higher density with Zn
- No significant change in growth rate
- Larger roughness with Zn



With Zn
Density: 8.6 g/cm^3
Thickness: 25 nm
Roughness: 3.5 nm

Without Zn
Density: 8.1 g/cm^3
Thickness: 26 nm
Roughness: 3.1 nm

Extra slide 3: MoN XRD

Effects of intermittent Zn pulse at 450°C

- Hexagonal δ-MoN in both cases (small trace of cubic γ-Mo_2N)
  - Zn leads to change in texture, (200) → (202), (002)
  - Similar to 500°C results of Alen, *et al.*, *J. Electrochem. Soc.* **152**, G361 (2005)