

PROOF OF CONCEPT THIN FILMS AND MULTILAYERS TOWARD ENHANCED FIELD GRADIENTS IN SRF CAVITIES

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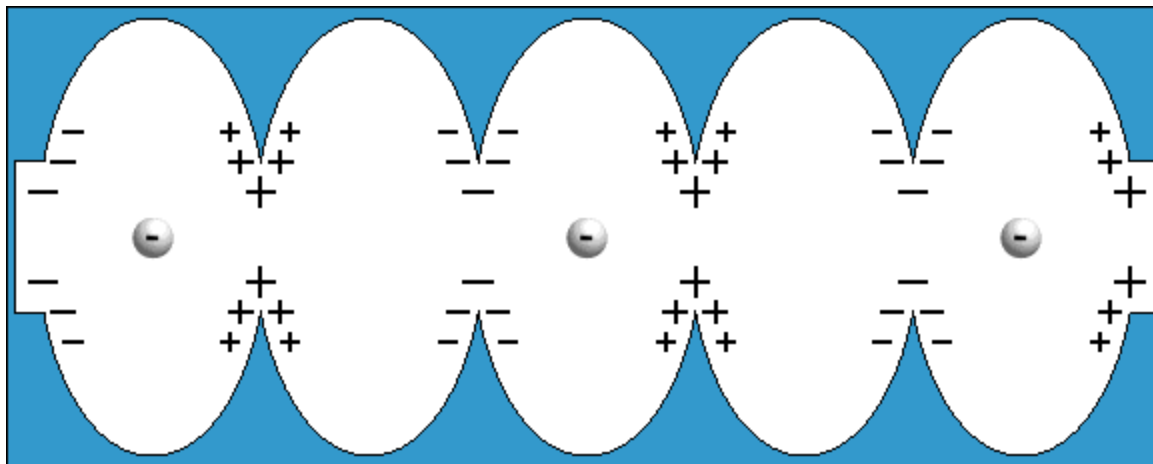
Temple University, Philadelphia, Pennsylvania, USA



**WILLIAM
& MARY**

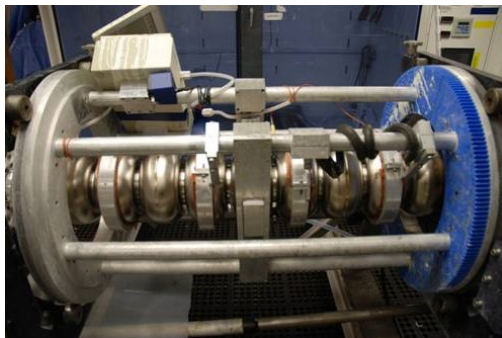
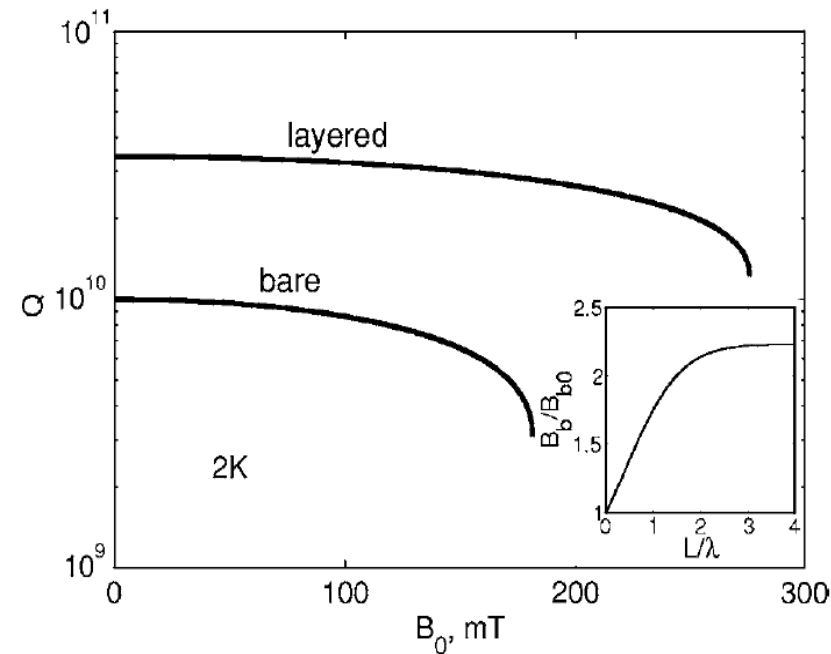
Nb SRF cavities

- The choice of Nb for superconducting cavities has been dictated by the requirement of having a material with a high lower critical field B_{c1} and a large energy gap Δ to prevent vortex dissipation and provide a low surface resistance R_s caused by thermally-activated quasiparticles at $T \ll T_c$ and $\omega \ll \Delta$,
- $R_s = (A\omega^2/T) \exp(-\Delta/T) + R_i$ where R_i is a small temperature independent residual resistance and A depends on SC parameters and ω and T



The Gurevich model

A. Gurevich, *Appl. Phys. Lett.* **88**, 012511 (2006).



- Significant improvement could be achieved if a Nb cavity is coated with a multilayer consisting of alternating superconducting S layers with higher B_c and dielectric I layers
- The S layer has thickness $d < \lambda$, and therefore can remain in the Meissner state at fields much higher than B_{c1} bulk due to the increase of the parallel B_{c1} in a thin film, while the insulating layer (~ 15 nm) is needed to prevent Josephson coupling between the SC layers.
- *Such structure would be particularly efficient in the case of elliptical cavities where the magnetic field is concentrated well inside the cavity and is parallel to the surface.*

Thin film geometry $\rightarrow B_{c1}$ enhancement \rightarrow Multilayer shielding

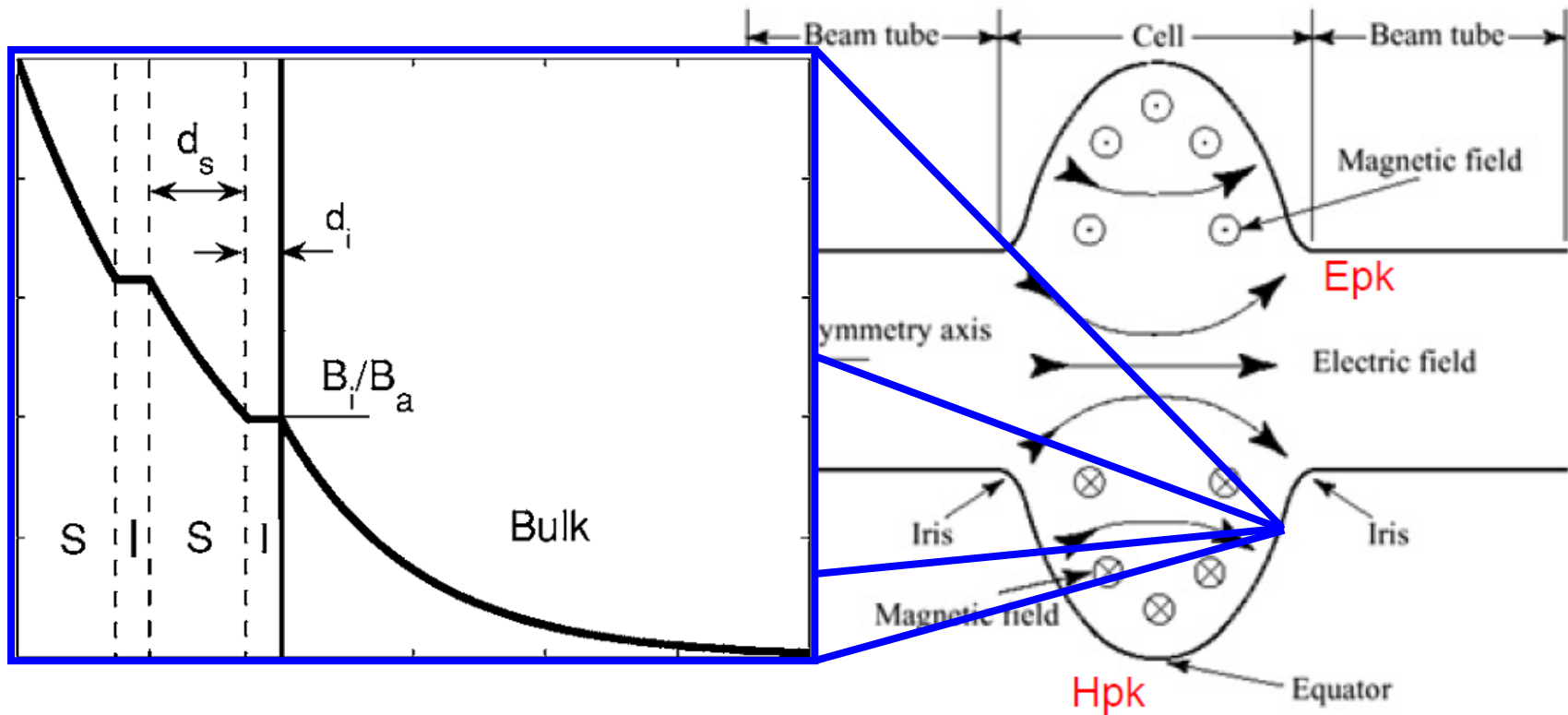
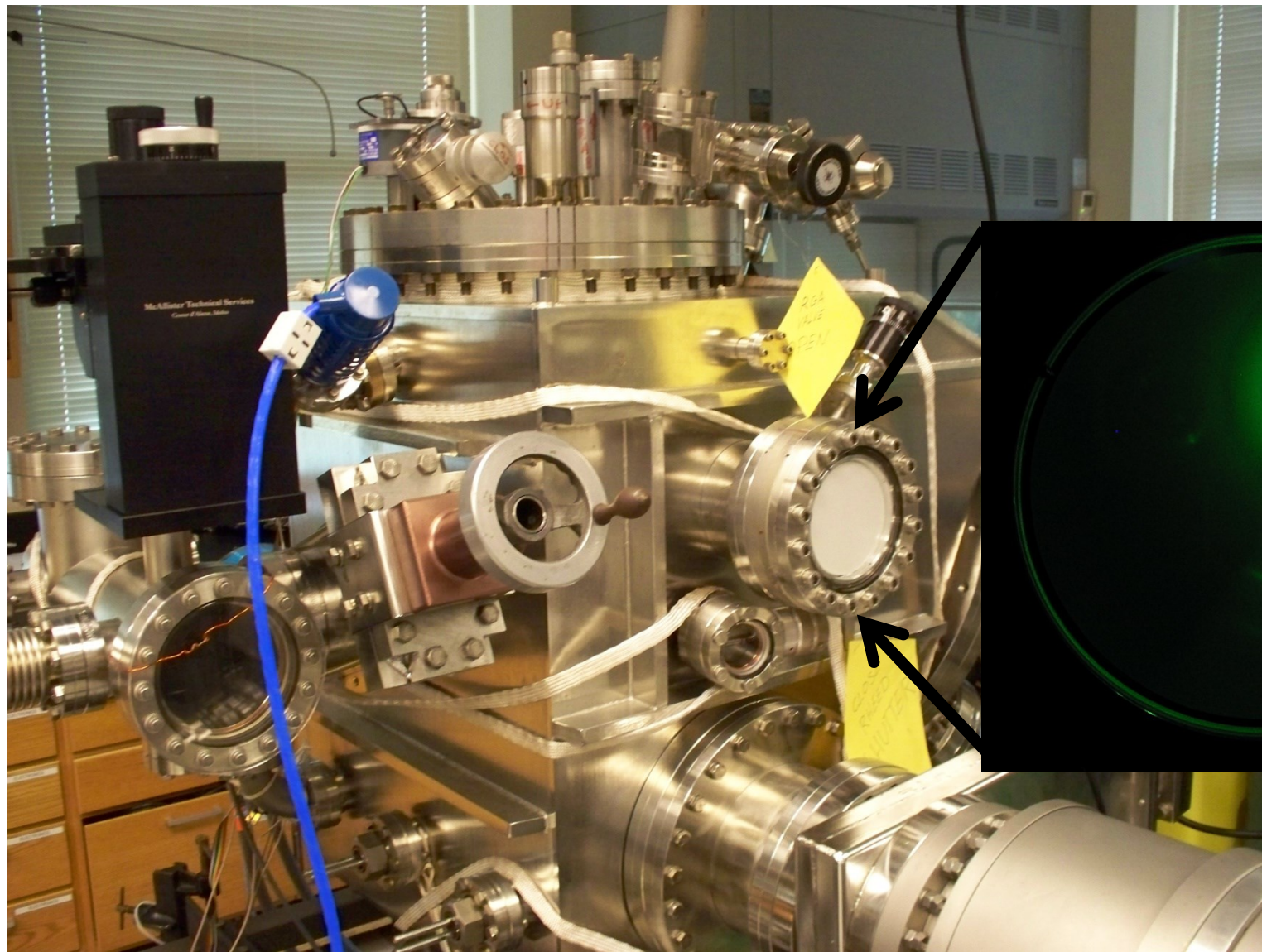


Image: CERN Accelerator School

Our experimental approach and methods

- In order to test the Gurevich model we have investigated the effect of microstructure and morphology on the superconducting properties of Nb thin films deposited onto different ceramic surfaces. In particular we studied a-plane sapphire and (001) MgO.
- We have also investigated Nb, NbN, NbTiN and MgB₂ based S/I/S trilayers.
- We monitored the microstructure of the films, the morphology of the surface and the superconducting properties as well as the DC and RF transport properties.
- We explored several aspects in the thin film deposition parameters-space, such as growth rate, substrate temperature during growth, annealing treatments, etc.

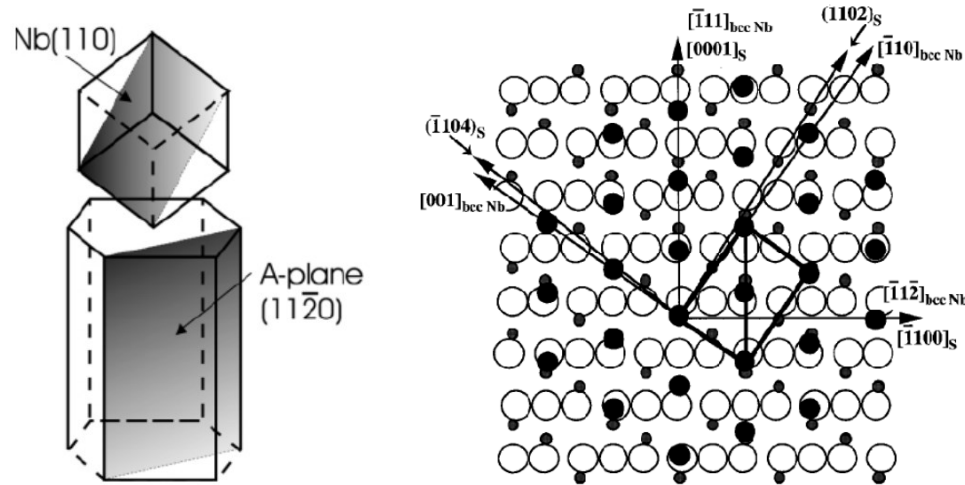


Thin film growth onto various different surfaces

- Growth on sapphire, magnesium oxide and copper surfaces

Nb growth on a-plane sapphire

- Nb can grow epitaxially on a-plane sapphire, with Nb(110)//Al₂O₃(11-20)



Comparison of RRR values obtained by different groups:

Group	Nb film thickness (nm)	RRR
Lukaszew	600	97
S. A. Wolf [1]	600	82
G. Wu [2]	235	50.2*

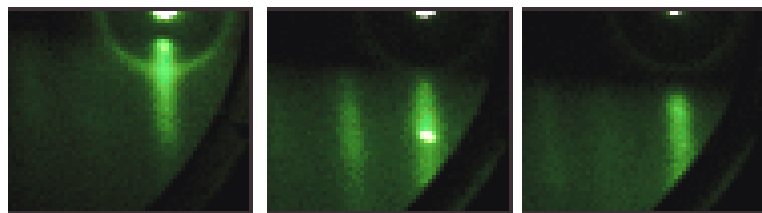
** RRR values for niobium thin films is highly dependent on thickness*

[1]. S. A. Wolf *et al.*, J. Vac. Sci. Technol. A 4 (3), May/June 1986

[2] G. Wu *et al.*, Thin Solid Films, 489 (2005) 56-62

Early stages of growth

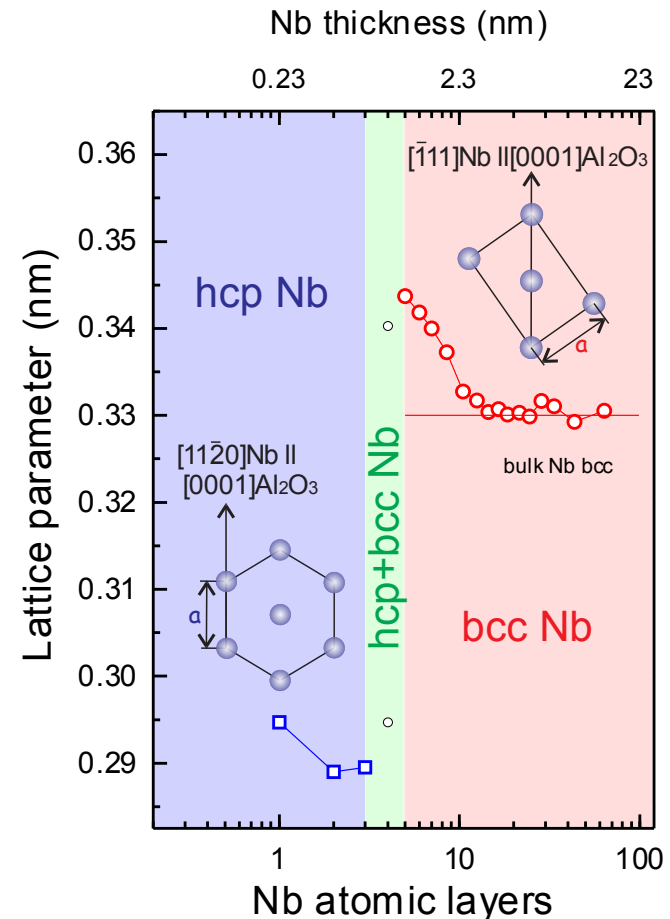
- Using Reflection high energy electron diffraction (RHEED), we observed a **hexagonal Nb surface structure** for the first 3 atomic layers followed by a strained *bcc* Nb(110) structure and the lattice parameter relaxes after 3 nm.
- RHEED images for the hexagonal phase at the third atomic layer. Patterns repeat every 60 deg.



0 deg

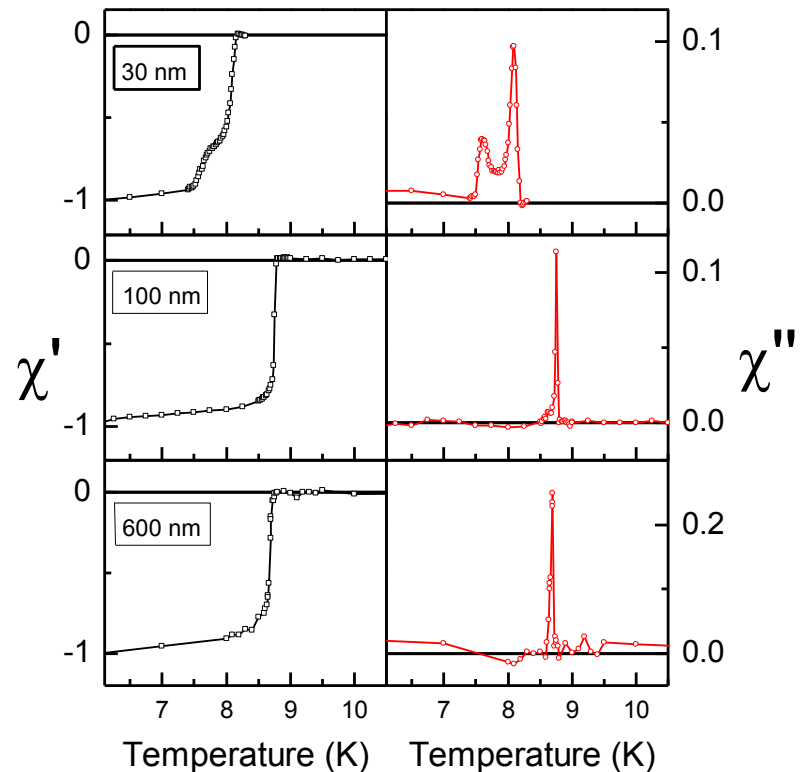
30 deg

60 deg



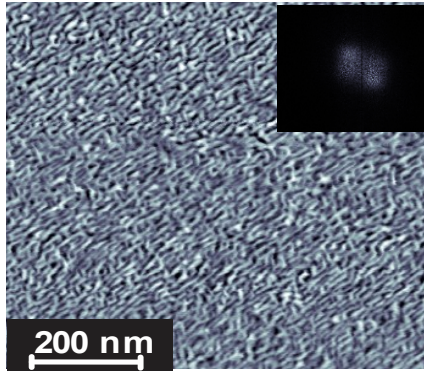
Susceptibility AC measurements

- The thinner Nb film exhibits two steps in the χ' susceptibility transition accompanied by two peaks in the χ'' susceptibility due to strained Nb layers at the interface.
- Growth on a-plane sapphire initially follows a hexagonal surface structure to relax the strain and to stabilize the subsequent growth of *bcc* Nb(110) phase.
- Such initial layers affect the superconducting properties of the films and these effects must be taken into account in the design of multilayers.

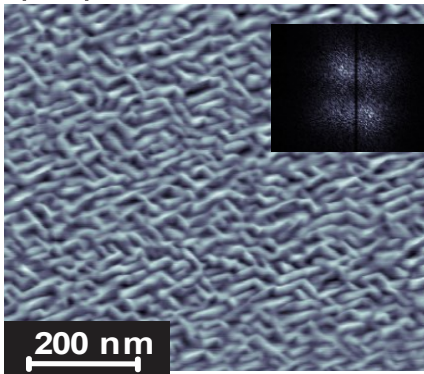


$$\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$$

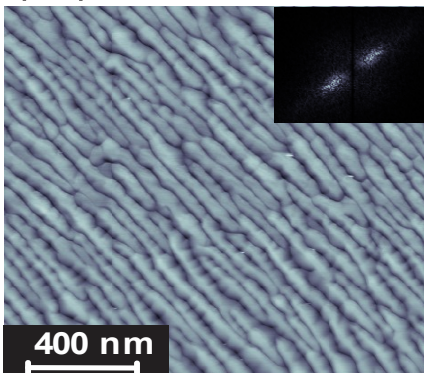
(a) 30 nm Nb



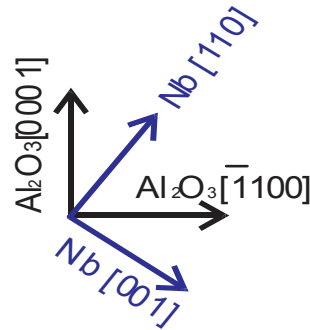
(b) 100 nm Nb



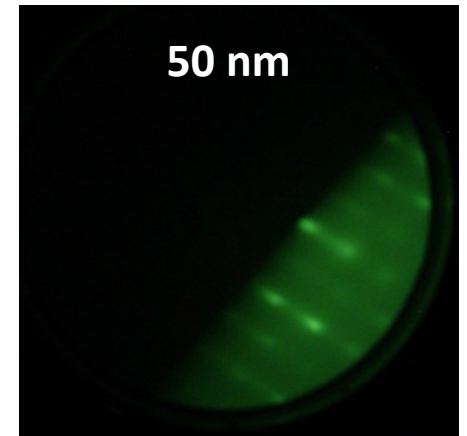
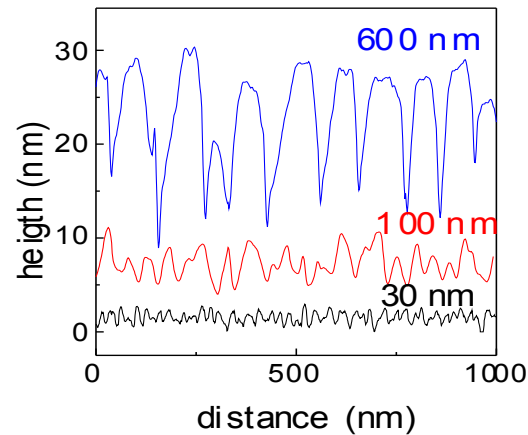
(c) 600 nm Nb



Subsequent growth



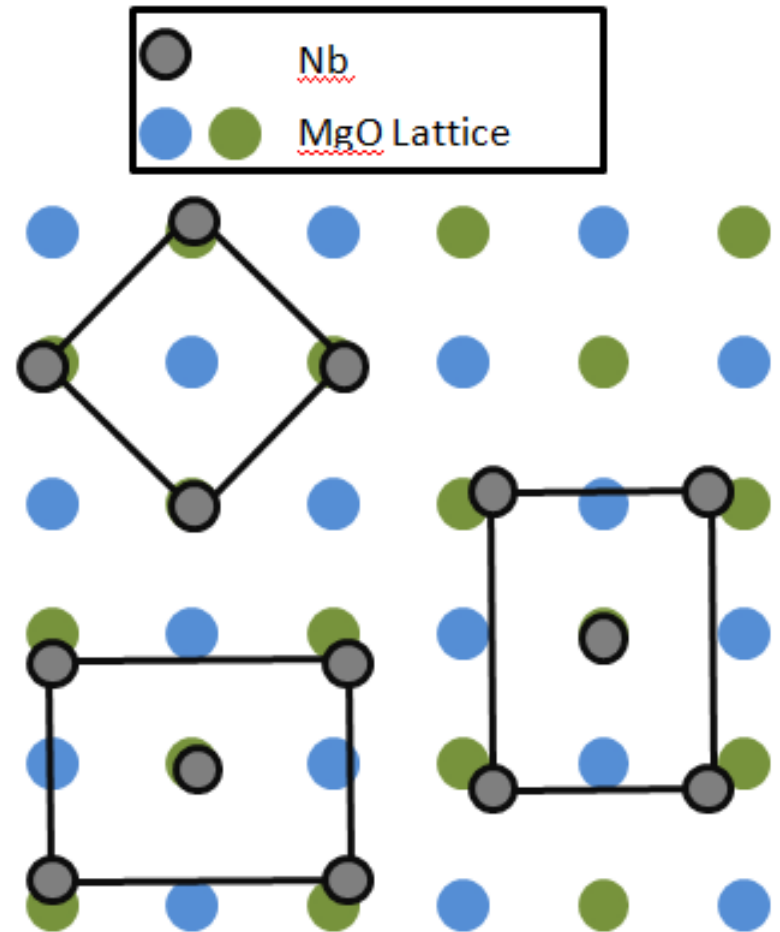
(d)



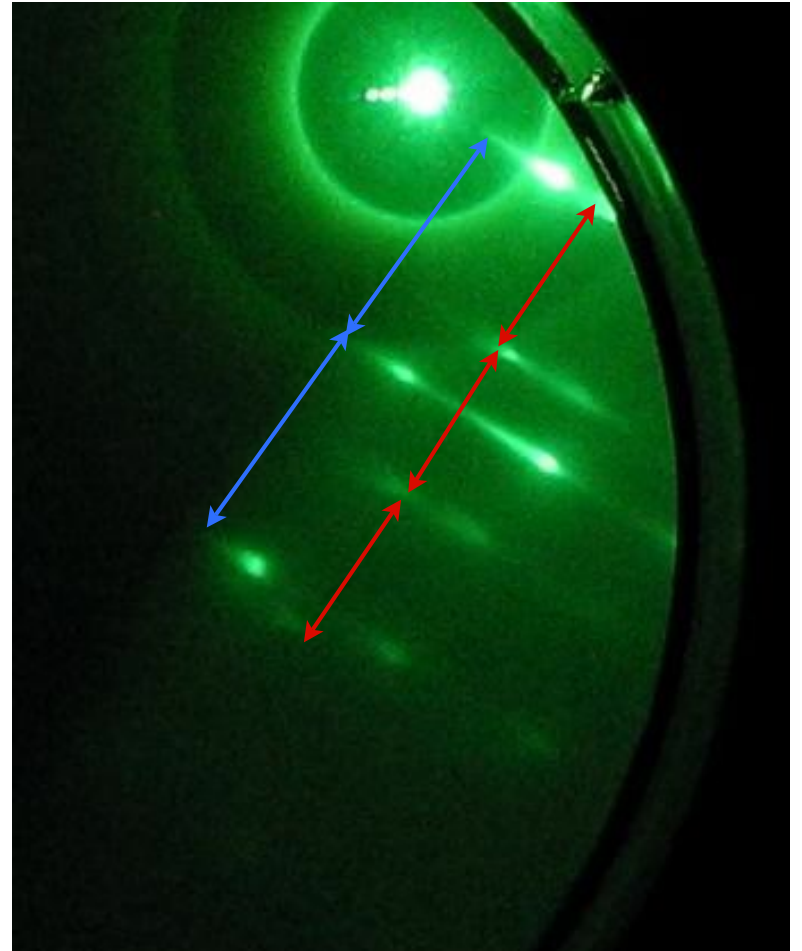
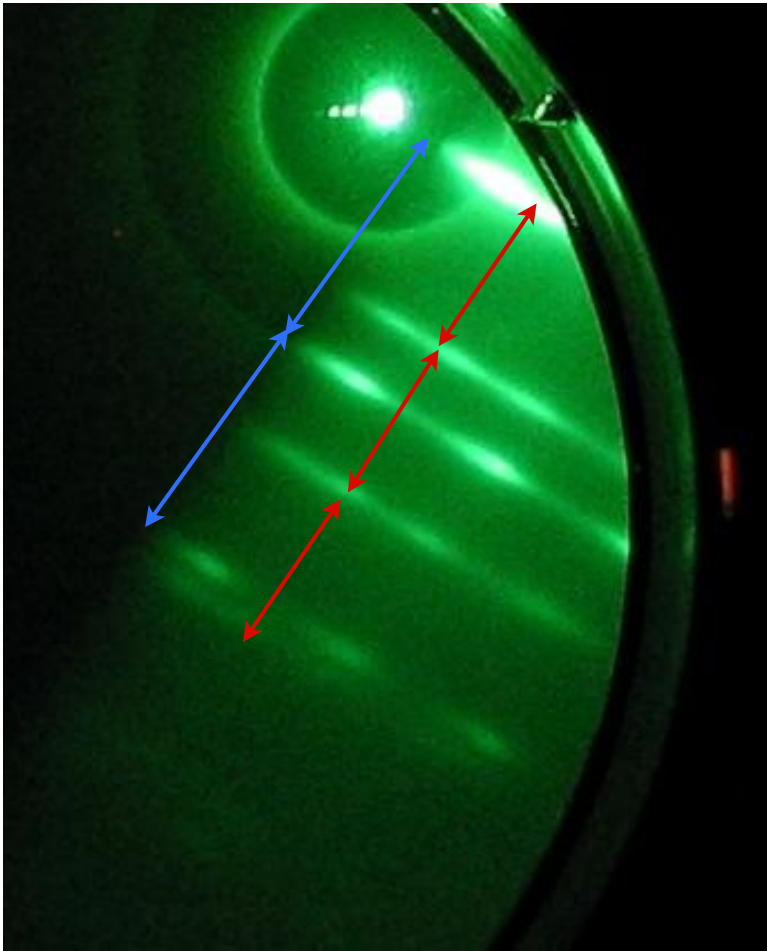
Biaxial anisotropy is observed for thicknesses up to 100 nm while **uniaxial anisotropy** is observed. For thicker films

Nb growth on (001) MgO

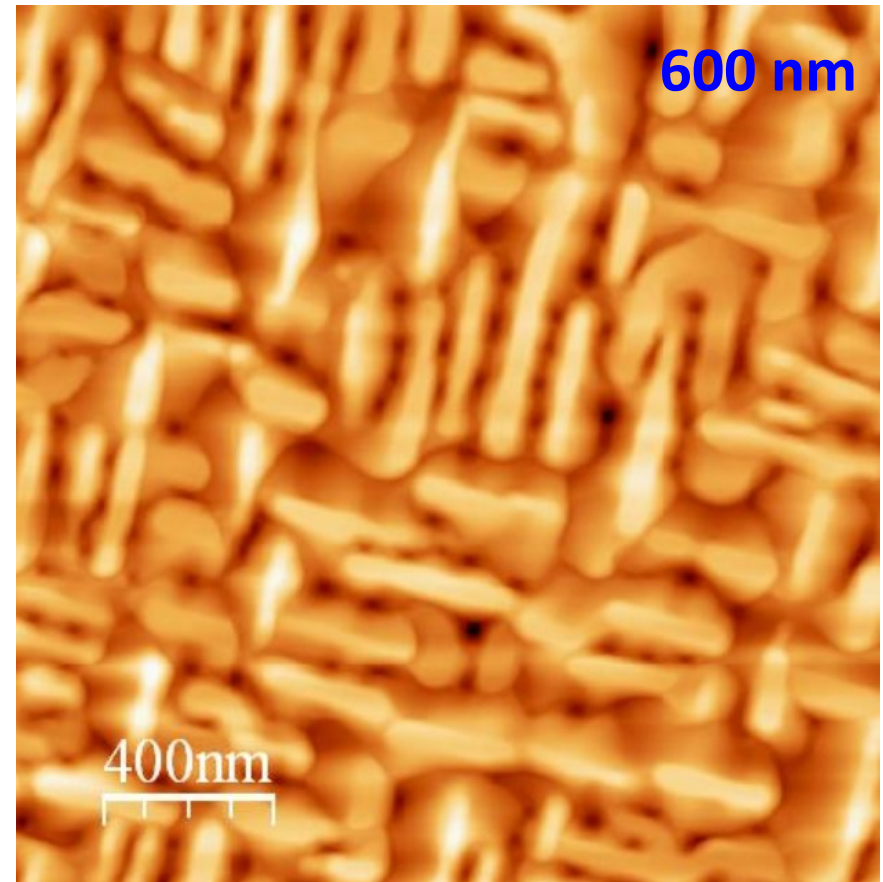
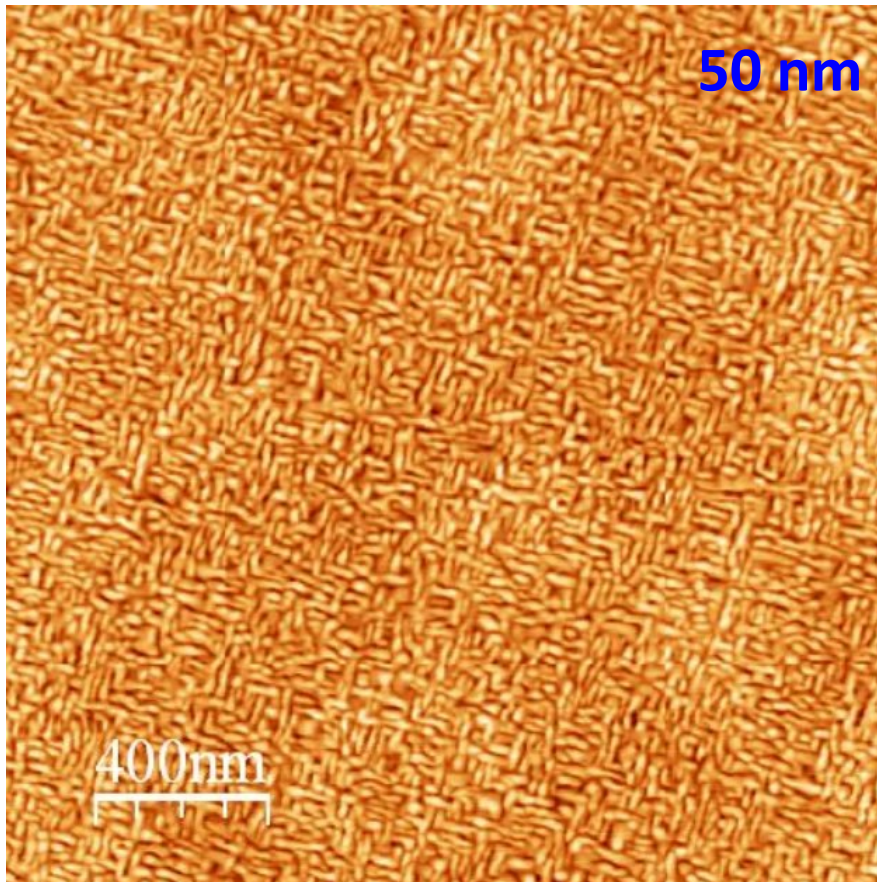
- Nb can also be epitaxially grown on (001) MgO surfaces.
- *Unexpected findings:*
We have found that depending on the deposition conditions it is possible to tailor different epitaxial possibilities.



RHEED images for Nb(110) on MgO

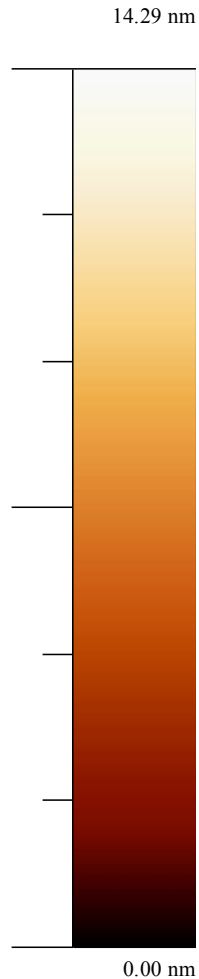
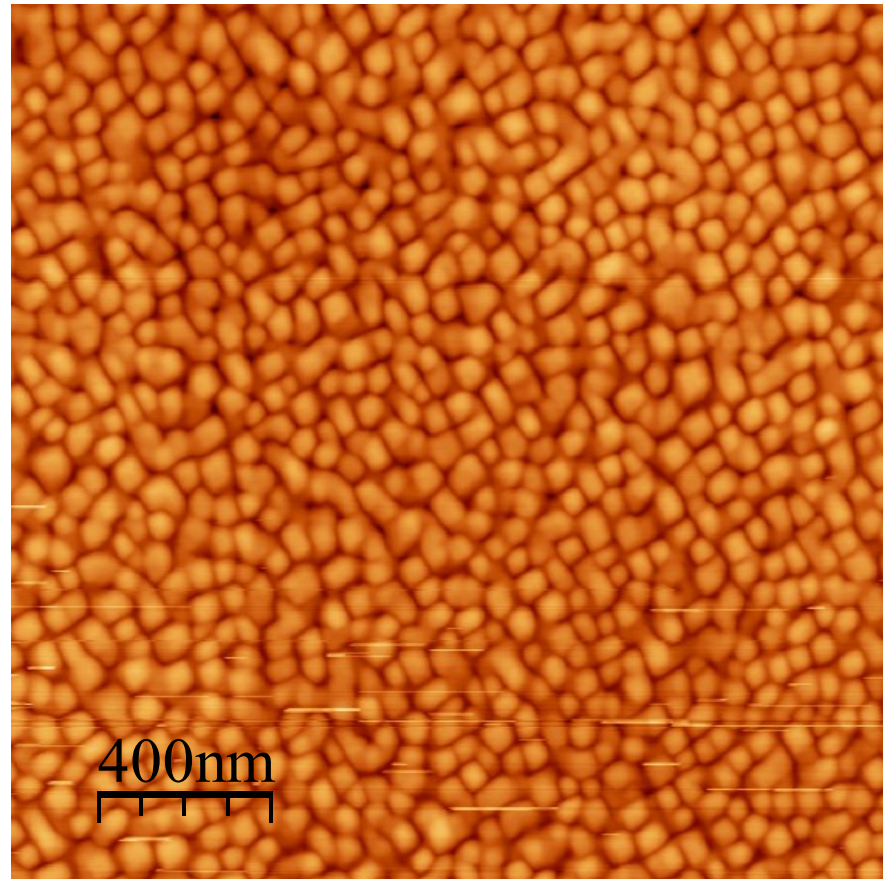
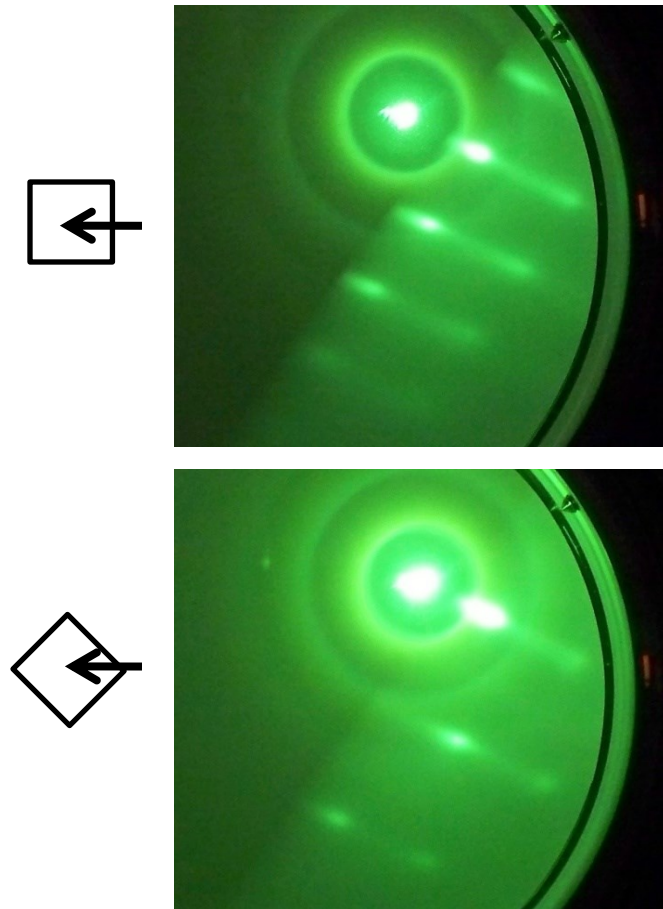


Scaling of surface features



RRR = 46.5
RMS = 6.51 nm

Nb (001) on MgO



RRR = 165 RMS = 4.06 nm

>200 RRR values!

Nb (001) on MgO

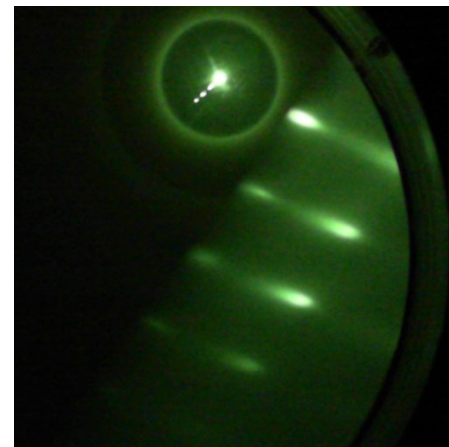
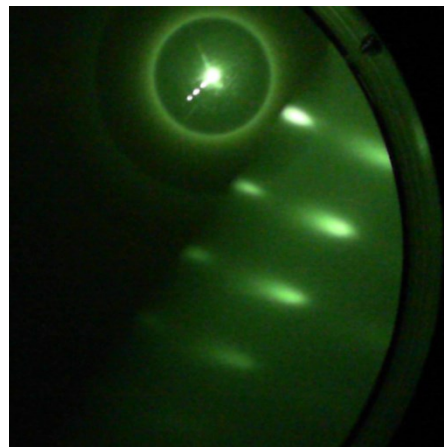
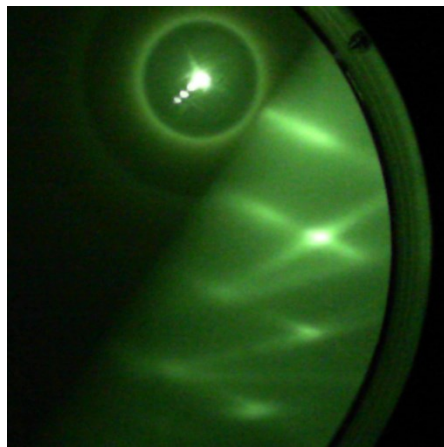
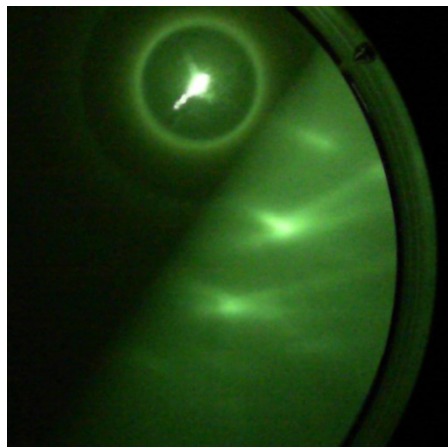
RHEED beam along MgO [100]  ←

MgO out of box

MgO annealed at 600 °C

30 nm Nb

100 nm Nb



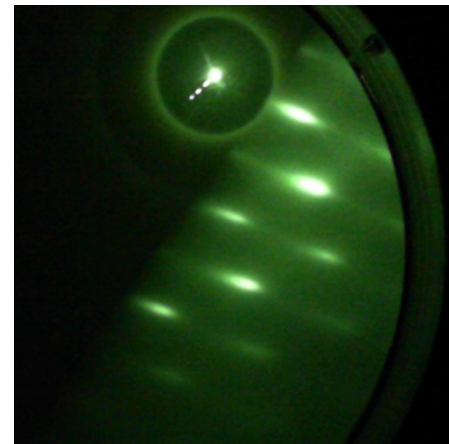
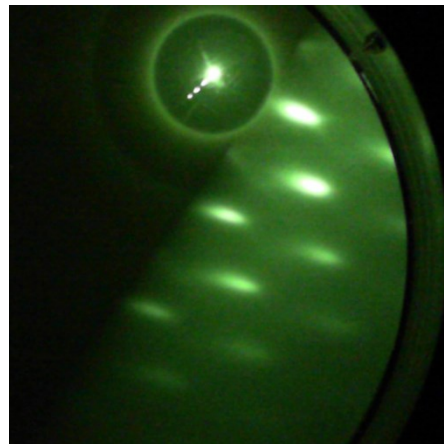
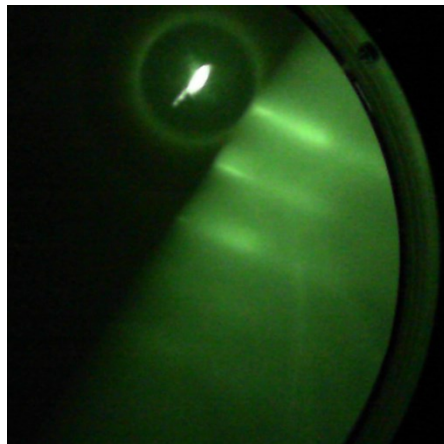
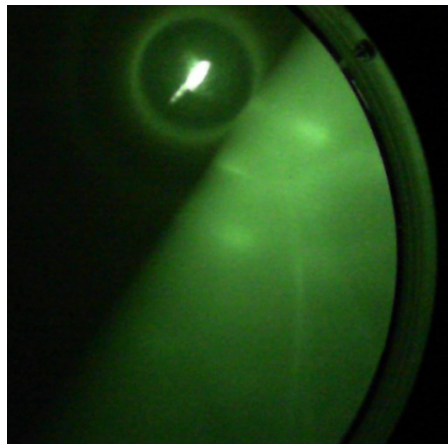
RHEED beam along MgO [110]  ←

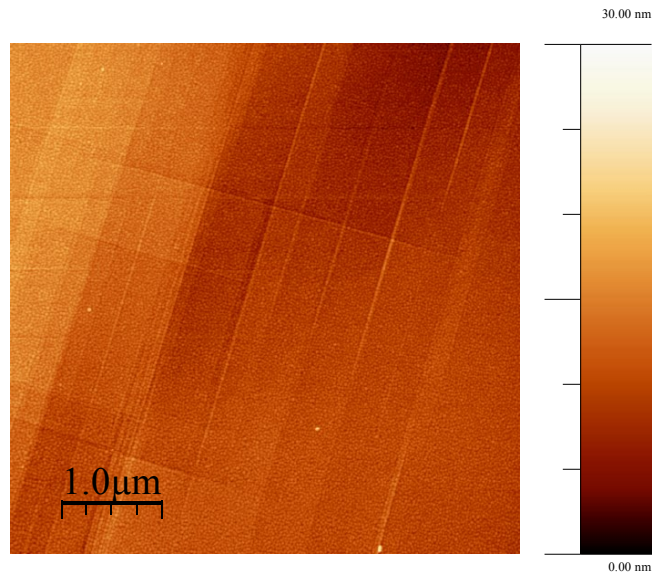
MgO out of box

MgO annealed at 600 °C

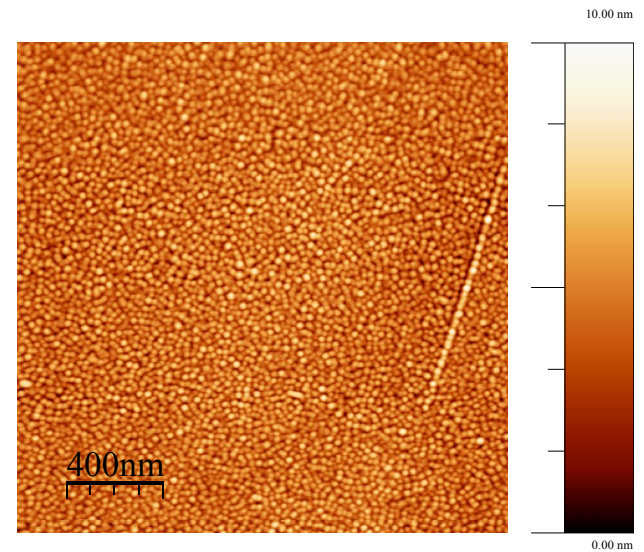
30 nm Nb

100 nm Nb

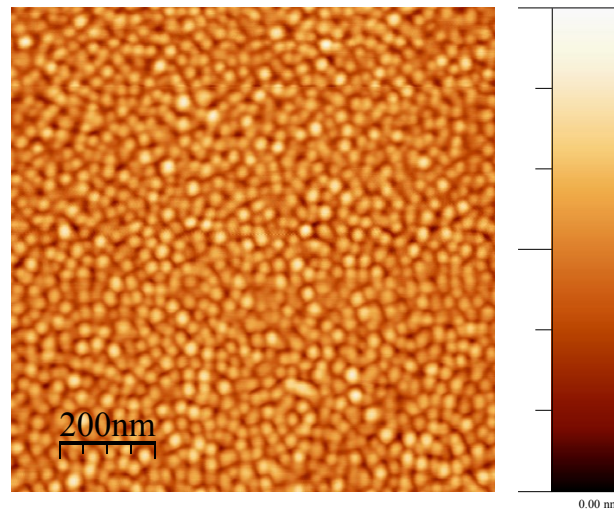




RMS = 2.90 nm



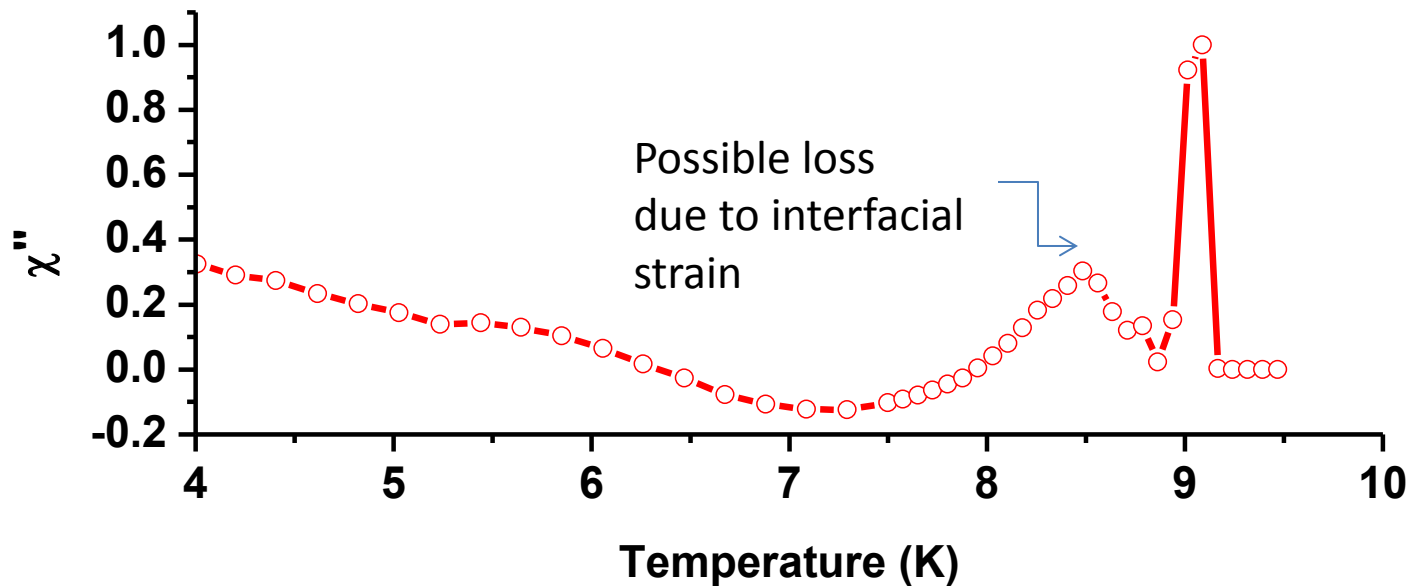
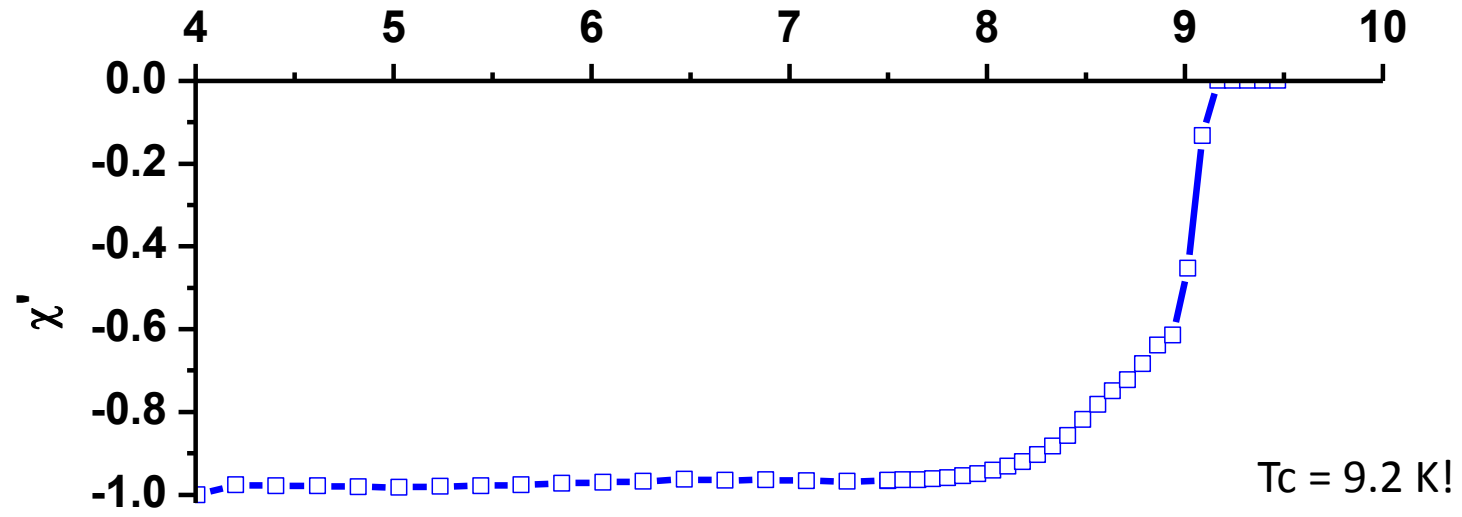
RMS = 1.21 nm



RMS = 1.08 nm

D. B. Beringer, W. M. Roach, C. Clavero, C. E. Reece, and **R. A. Lukaszew**, "Roughness analysis applied to niobium thin films grown on MgO(001) surfaces for superconducting radio frequency cavity applications," *Phys. Rev. ST Accel. Beams* **16**, 022001 (2013).

SQUID characterization



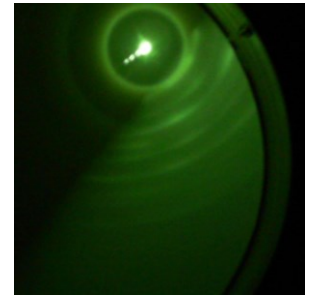
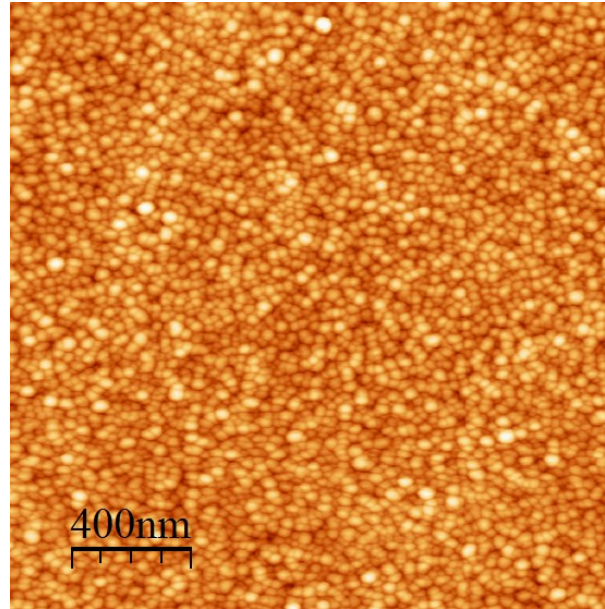
Nb-based trilayer

30 nm Nb

15 nm MgO

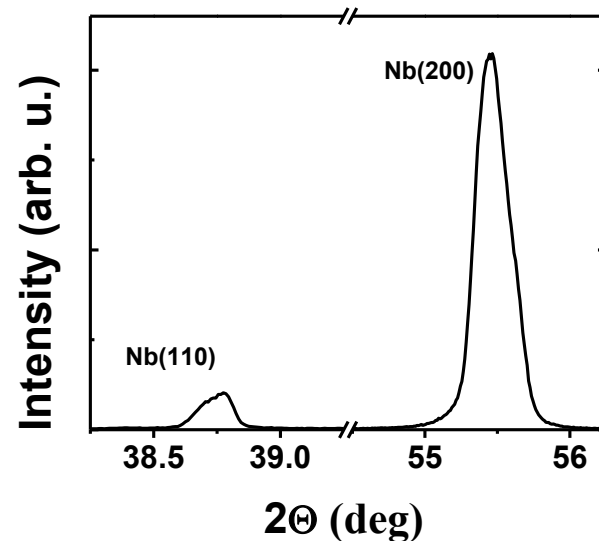
250 nm Nb

MgO (100)



RHEED indicates
film with high
degree of (001)
texture

XRD confirmed RHEED results:

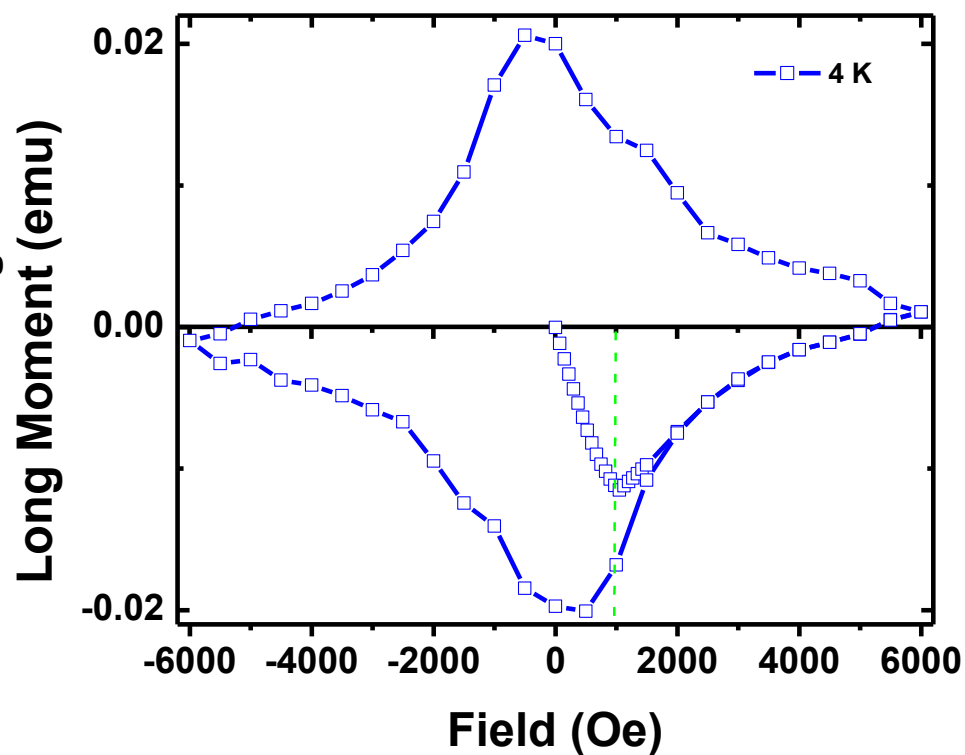
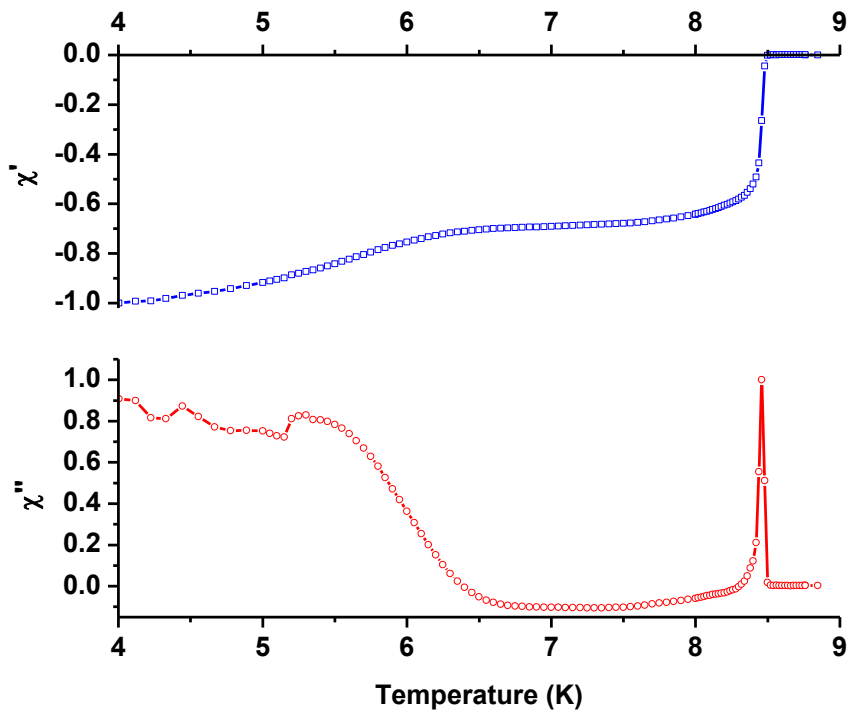


SQUID magnetometry

- Antoine et al [1] using SQUID magnetometry as well as third harmonic analysis to validate SQUID magnetometry measured the vortex penetration field on multilayered samples and demonstrated field enhancement.
- In our work, we measured hysteresis loops as well as trapped moments that appear after application and removal of the applied field, following the work of C. Bohmer et al. [2]

[1] C. Z. Antoine, S. Berry, S. Bouat, J.-F. Jacquot, J.-C. Villegier, G. Lamura, and A. Gurevich, *Phys. Rev. ST Accel. Beams*, vol. 13, p. 121 001, 2010; C. Z. Antoine, S. Berry, M. Aurino, J.-F. Jacquot, J.- C. Villegier, G. Lamura, and A. Andreone, *IEEE Trans. Appl. Supercond.*, vol. 3, p. 2601, 2011; [2] C. Böhmer, G. Brandstätter, and H. W. Weber, *Supercond. Sci. Technol.* 10 A1 (1997).

SQUID characterization



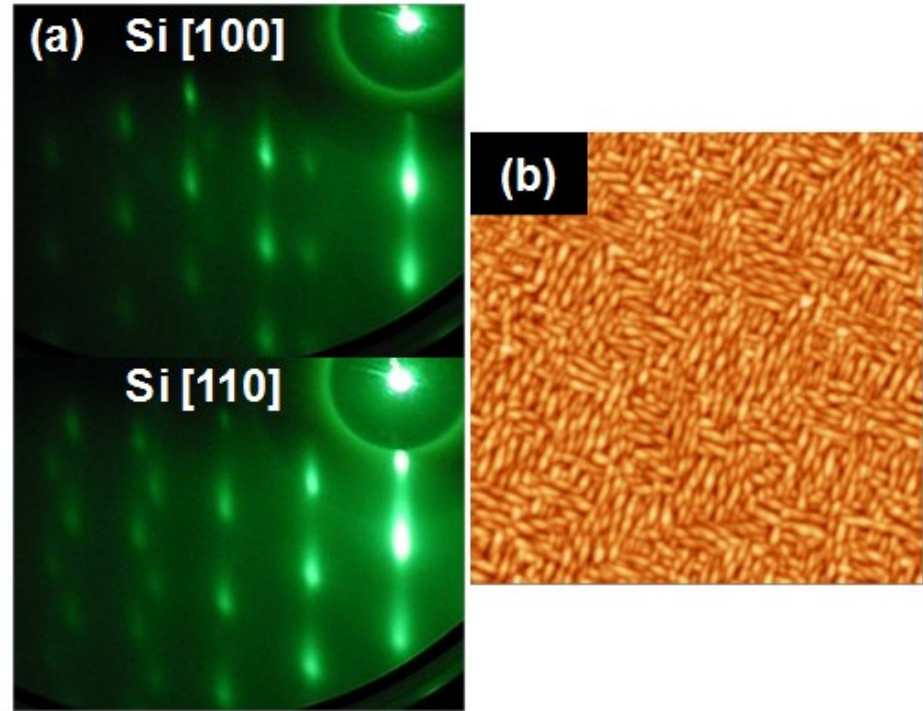
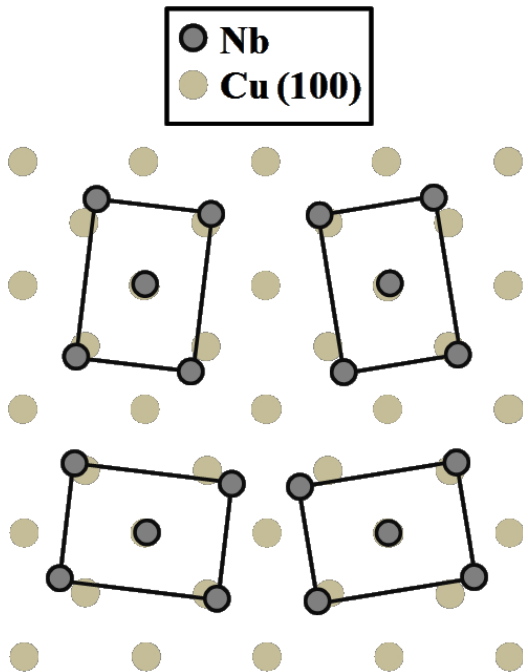
Nb on Cu (111)

- Growth at room temperature and annealing at 350 °C leads to the crystallization of Nb islands in a hexagonal surface structure, even though Nb is expected to grow tetragonal (110).



Nb films on Cu (001) surfaces

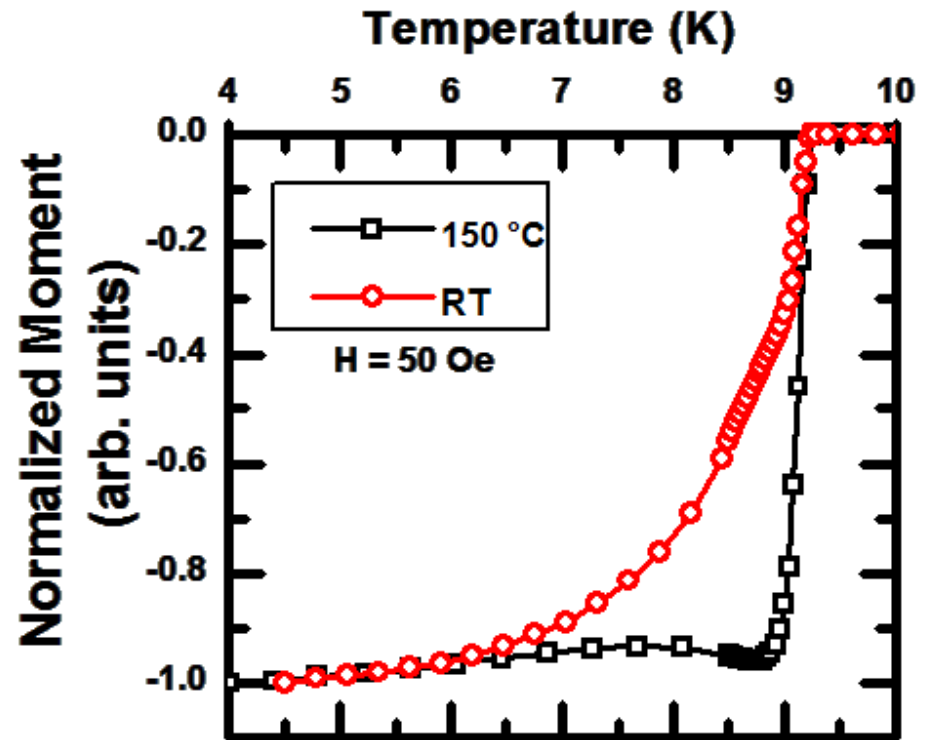
Possible Nb/Cu(100) epitaxy:



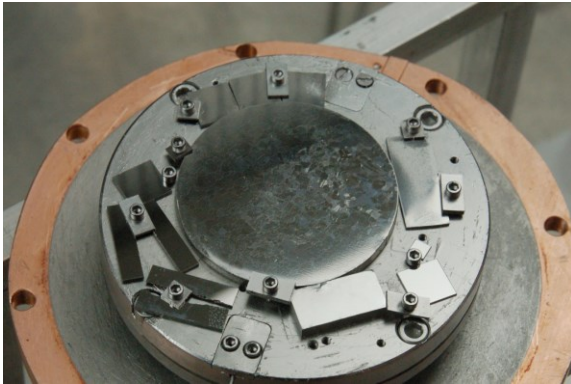
(a) RHEED pattern for Nb(110)/Cu(100)/Si(100) along the Si[100] and Si[110] azimuths. (b) A representative $2\ \mu\text{m} \times 2\ \mu\text{m}$ AFM scan for Nb films on the Cu template.

SC properties for different growth T

- The films grown at 150 °C have a very sharp transition from the superconducting state to the normal state that begins at ~9 K while films grown at RT have a much more gradual transition.
- Our results suggest that an increased deposition temperature of Nb onto Cu leads to films with higher crystalline quality (grain size) and thus improved superconducting properties (H_{C1}).

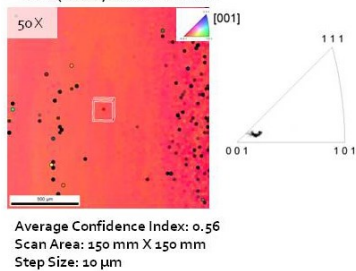


Nb films on Cu

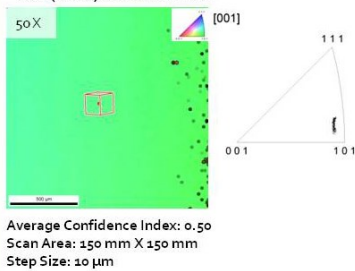


Nb films with quality comparable to high RRR bulk Nb (as used for SRF cavities) have been produced both on single crystal and polycrystalline Cu substrates with energetic condensation via ECR (electron cyclotron resonance) at Jlab. SRF measurements are in progress.

Cu (100) Substrate



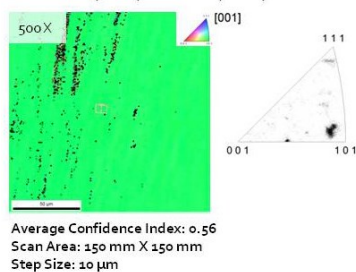
Cu (110) Substrate



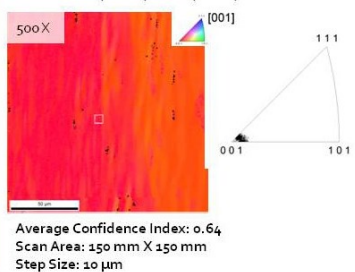
Cu (111) Substrate



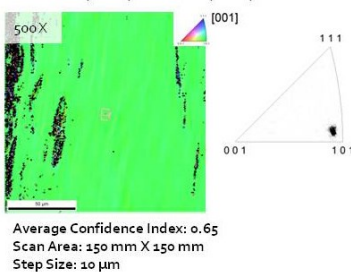
ECR Nb (110) on Cu (100)



ECR Nb (100) Cu (110)



ECR Nb (110) on Cu (111)



Substrate	RRR
Single crystal	
Cu (100)	129
Cu (110)	275
Cu (111)	242
Polycrystalline	
Cu fine grains	150
Cu large grains	289

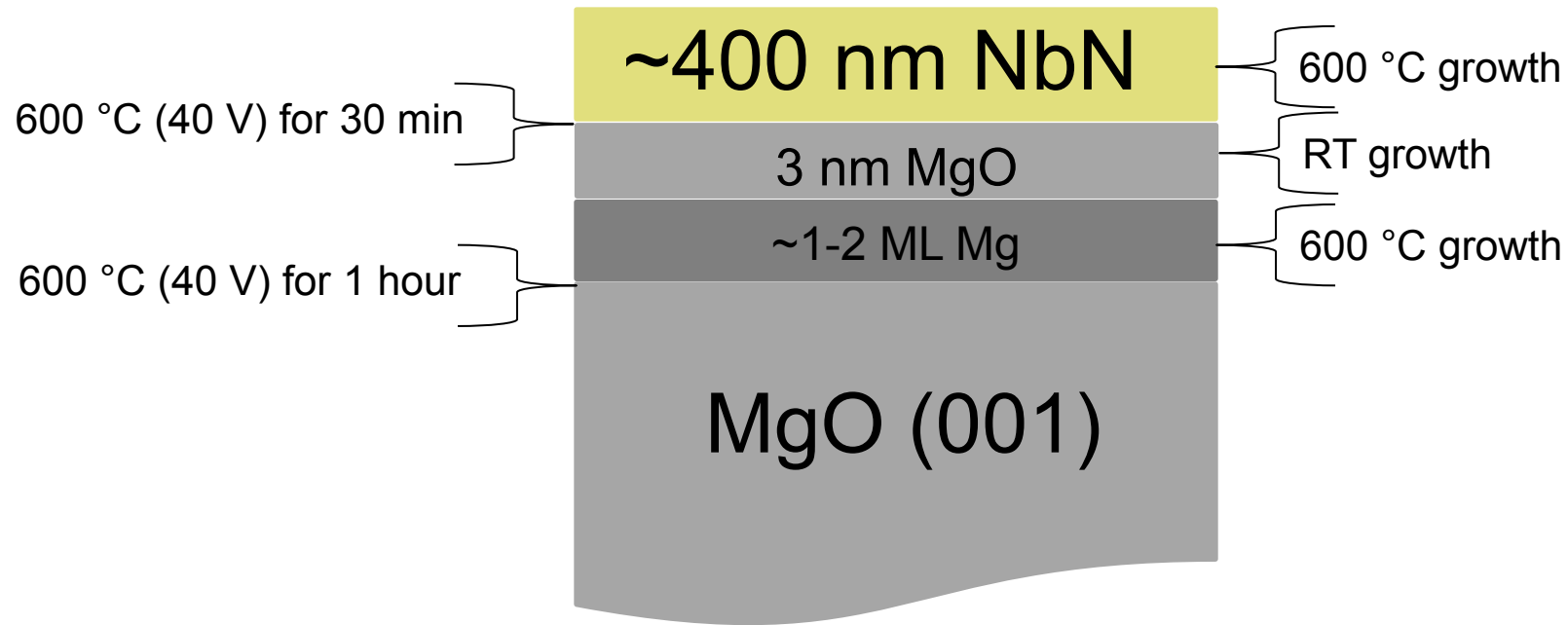
Hetero-epitaxial relationships between Nb and Cu verified.

Other possible SC thin films for the SIS model

NbN, MgB₂, etc.

Growth Procedure for NbN Films

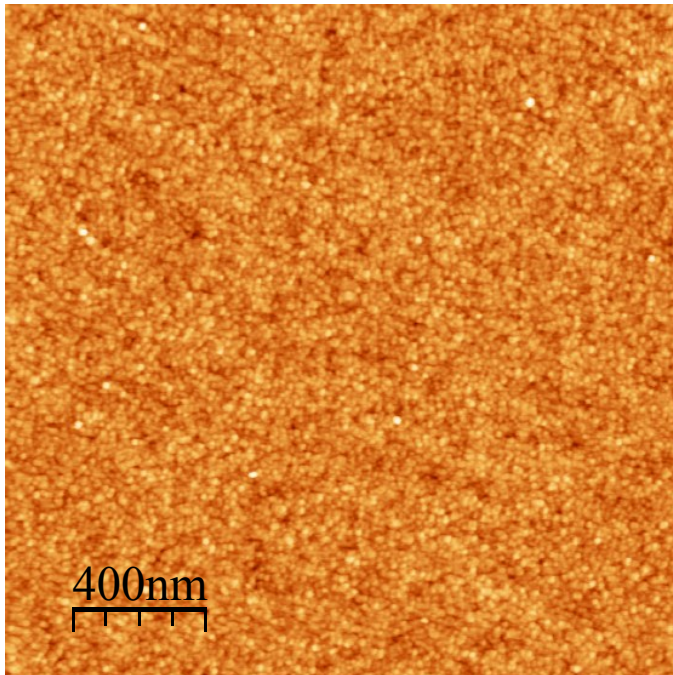
Partial Pressure Series



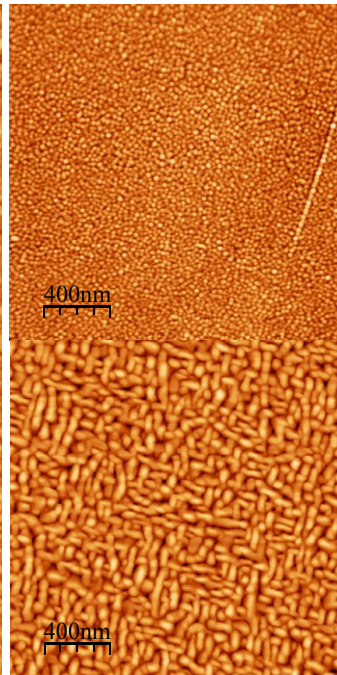
All NbN films are ~200 nm thick based on XRR/Profilometry

Compare Surface Morphology of Nb and NbN similar films

NbN



Nb(100)



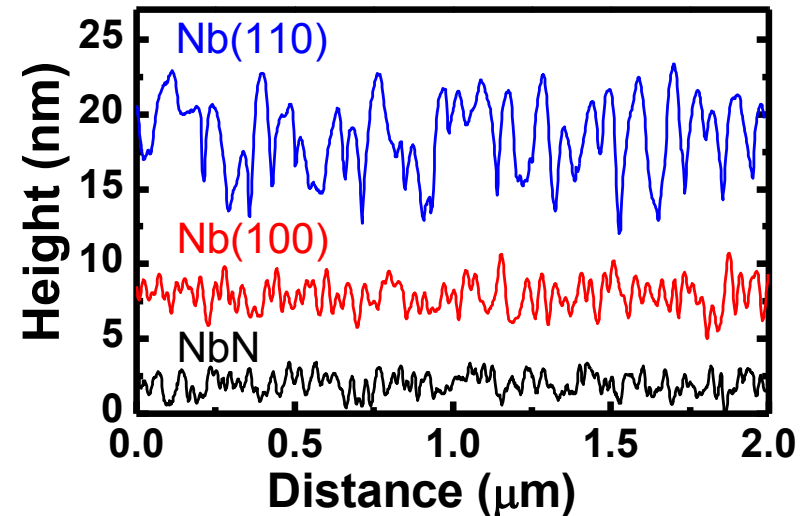
Nb(110)

RMS Roughness for comparable film thickness:

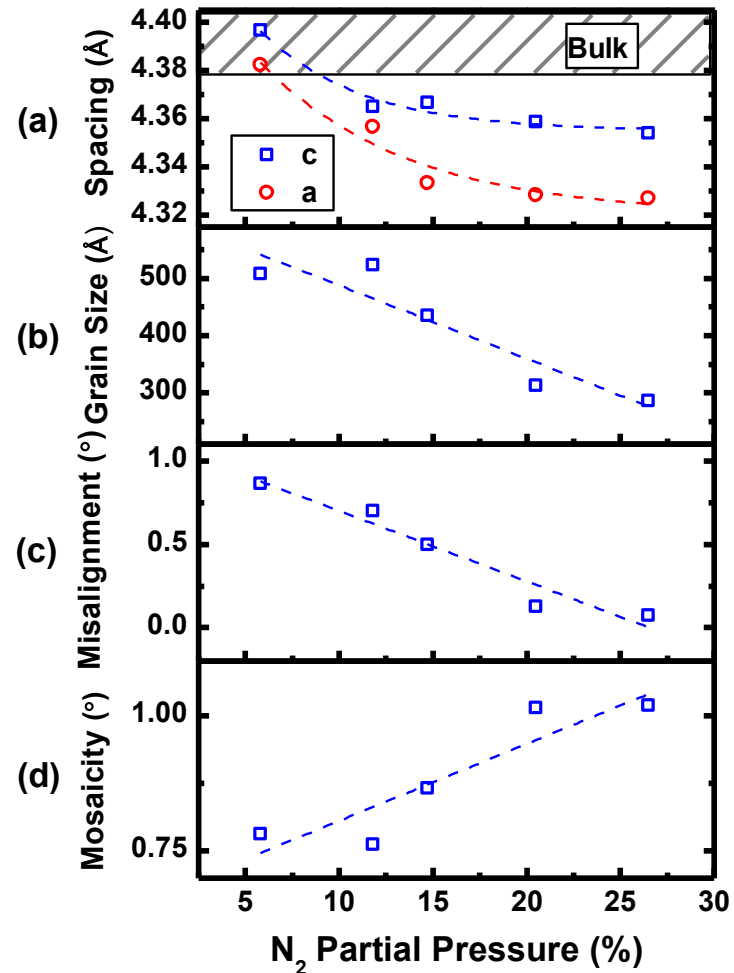
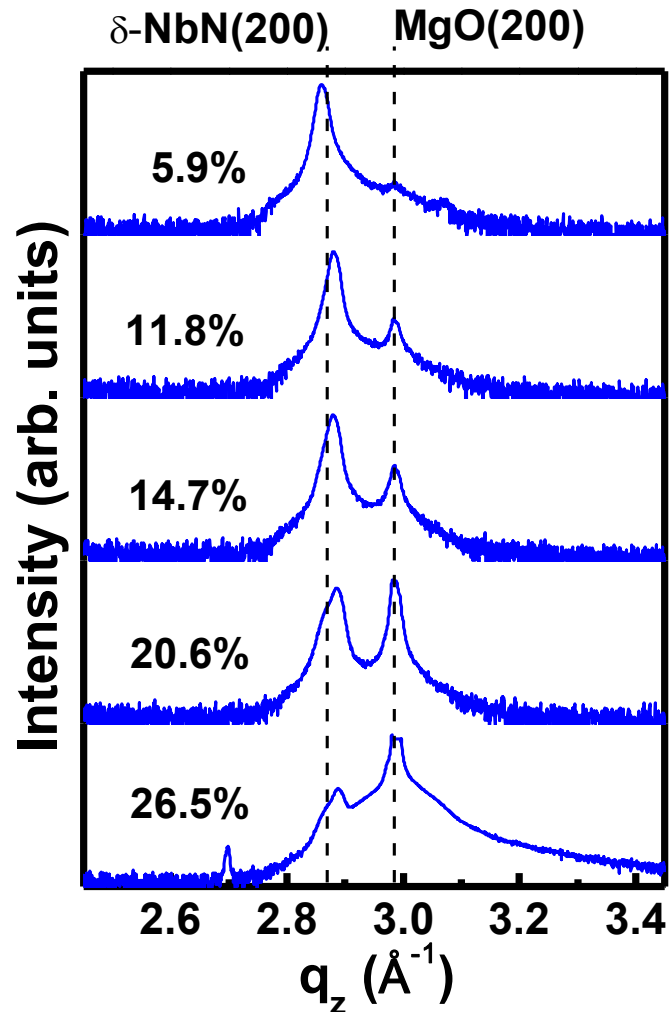
NbN <1 nm

Nb(100) 1.21 nm

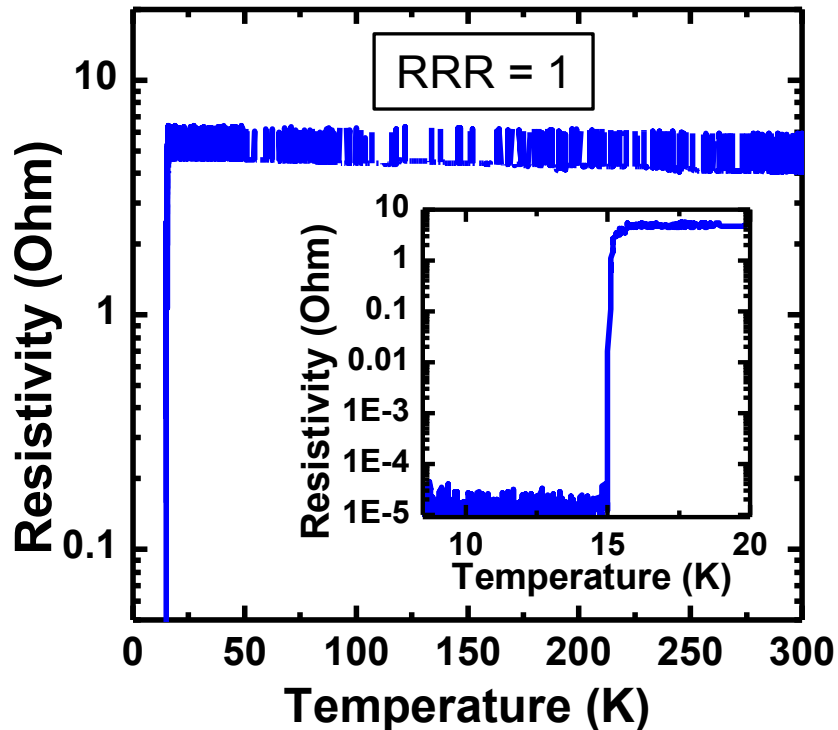
Nb(110) 2.45 nm



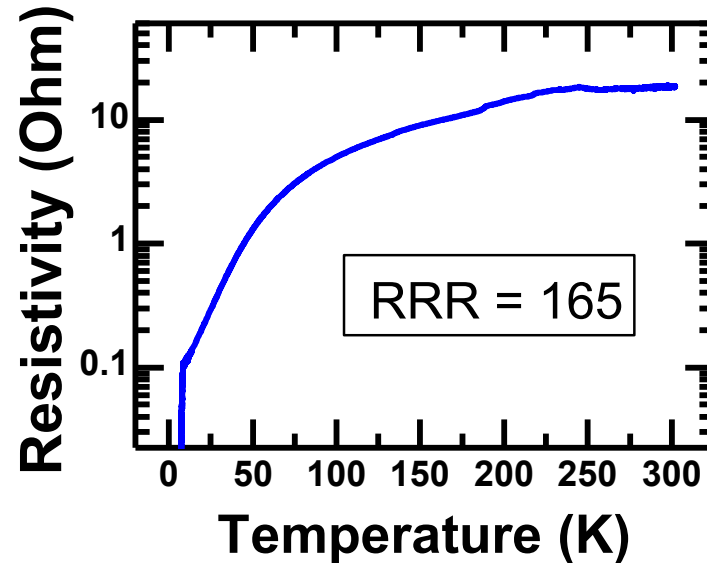
NbN films microstructure



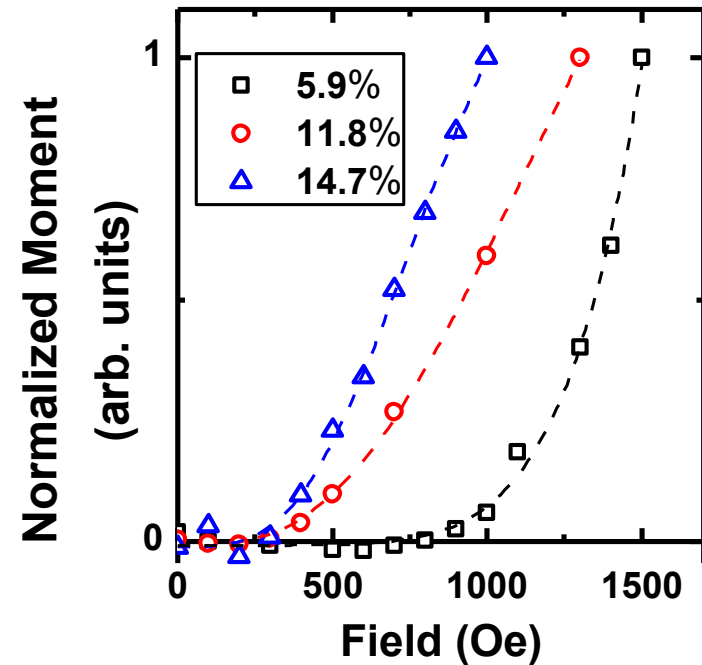
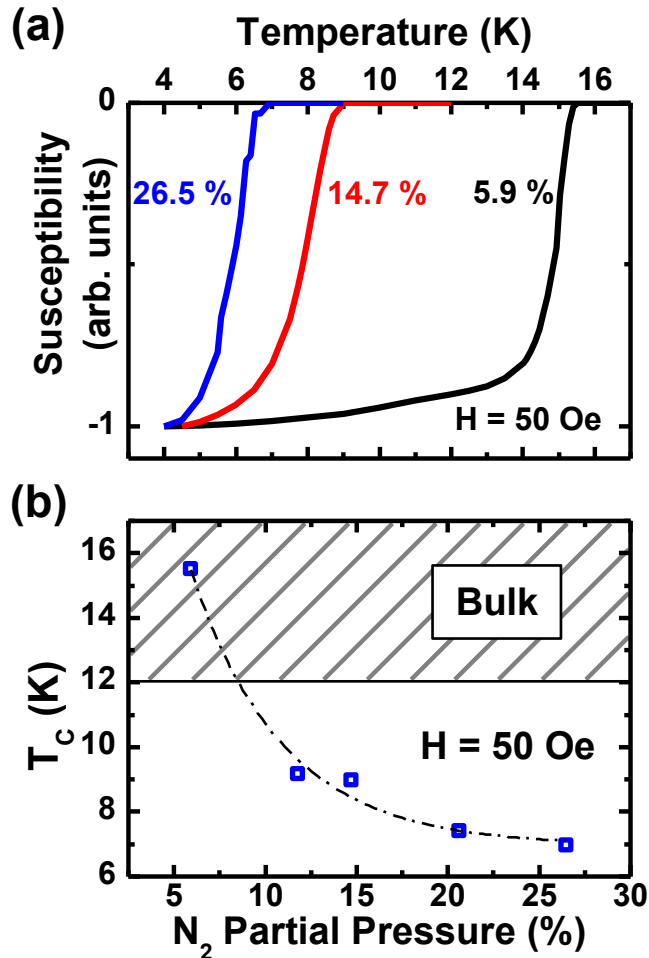
Residual Resistance Ratio



Resistive behavior for NbN differs from that of metals such as Nb. RRR=1 is indication of very good quality film!



Superconducting Properties



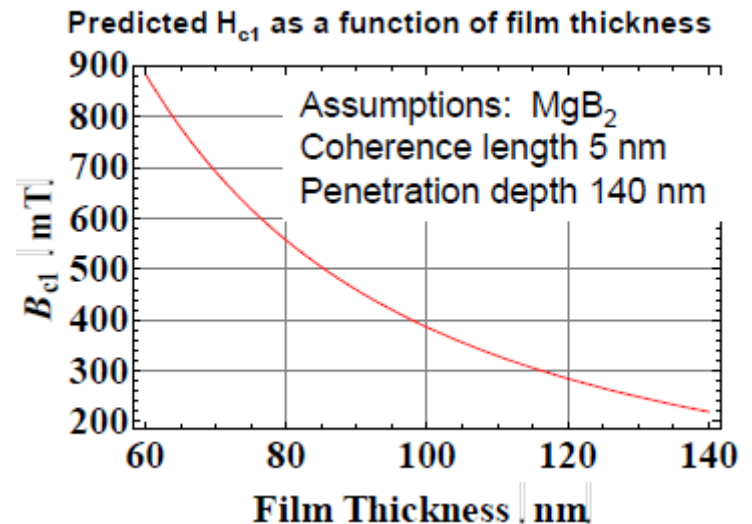
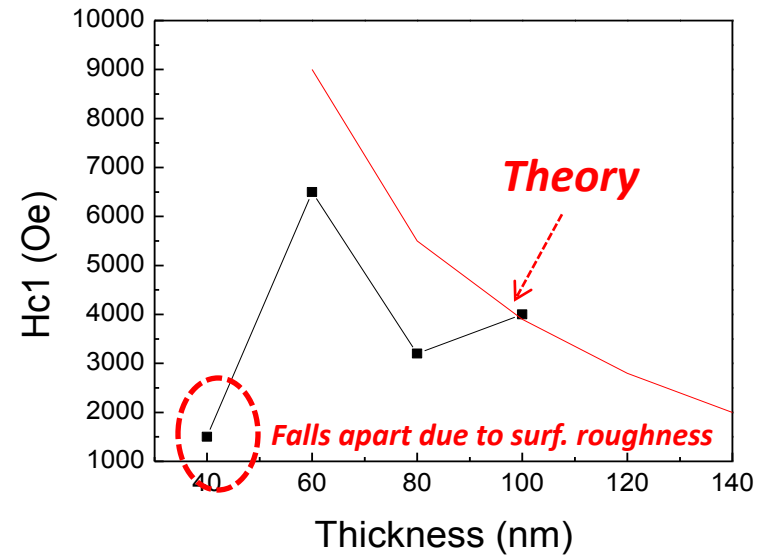
W. M. Roach, J. R. Skuza, D. B. Beringer, Z. Li, C. Clavero, and **R. A. Lukaszew**, "NbN thin films for superconducting radio frequency cavities". *Supercond. Sci. Technol.* **25**, 125016 (2012).

MgB₂ thin films

We have initiated investigations on MgB₂ thin films.

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

[Gurevich, APL 88 (2006) 012511]

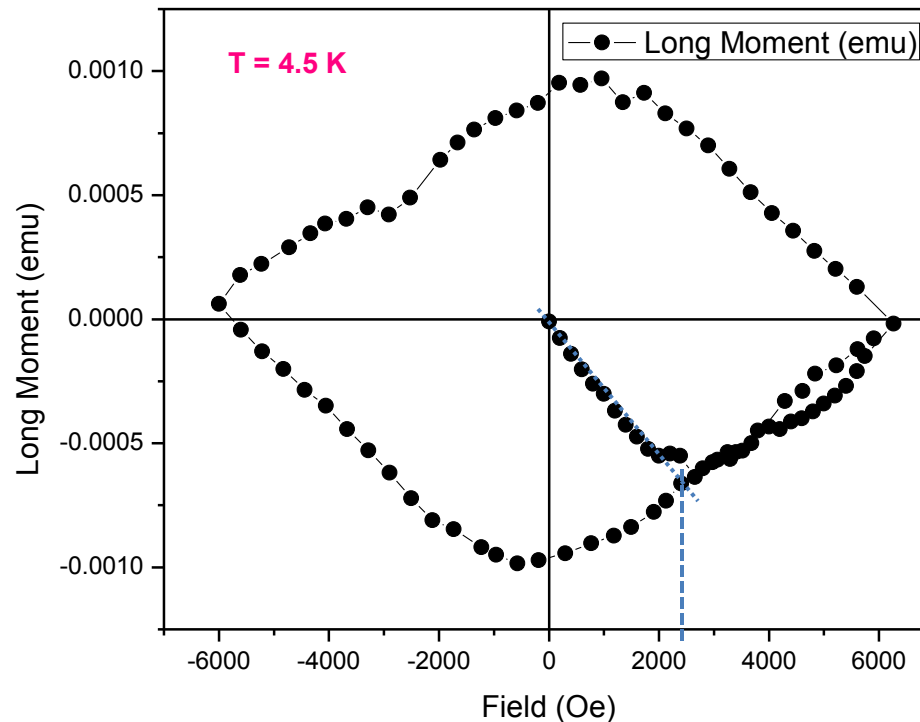
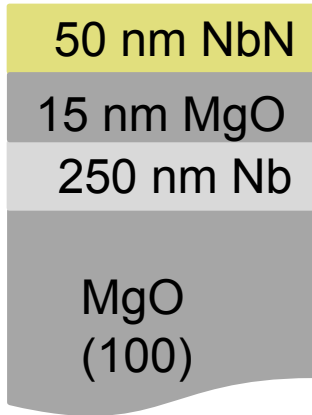


D. B. Beringer, C. Clavero, T. Tan, X. X. Xi, W. M. Roach, and R. A. Lukaszew, "Thickness Dependence and Enhancement of H_{c1} in Epitaxial MgB₂ Thin Films," *IEEE Trans. Appl. Supercond.* **23**, 7500604 (2013).

SIS layers

- NbN-based, MgB₂ based and NbTiN-based trilayers

NbN-based multilayer

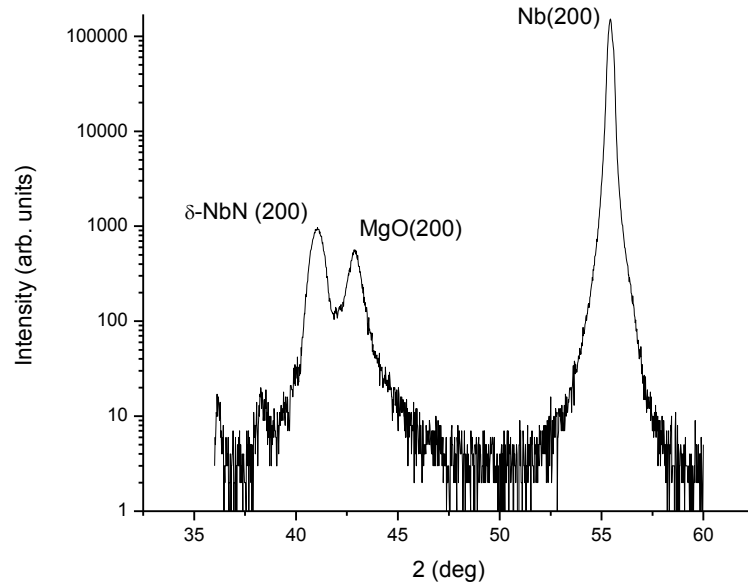


$H_{c1\text{-NbN-based-Multilayer}} \sim 220 \text{ mT!}$

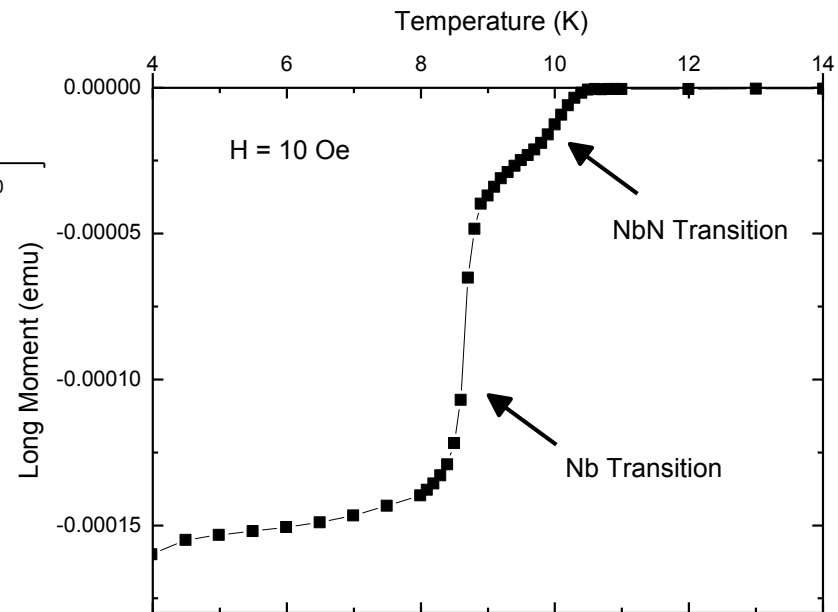
$H_{c1\text{-bulk Nb}} = 170 \text{ mT}$

“Magnetic Shielding Larger than the Lower Critical Field of Niobium in Multilayers” W. M. Roach, D. B. Beringer, Z. Li, C. Clavero, and R. A. Lukaszew, *IEEE Trans. Appl. Supercond.* **23**, 8600203 (2013).

XRD



SQUID



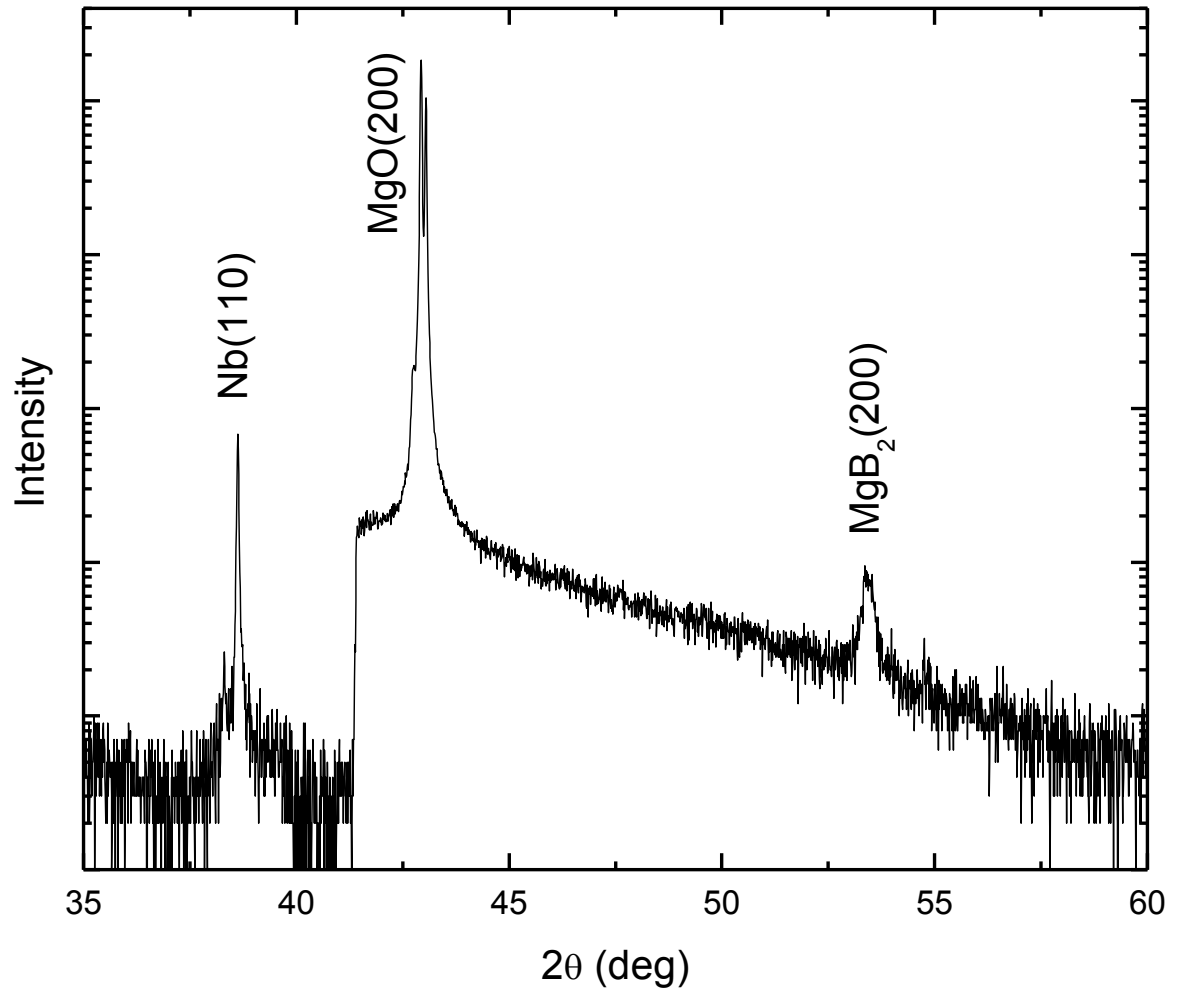
XRD characterization MgB₂-based ML sample

50 nm NbN

15 nm MgO

250 nm Nb

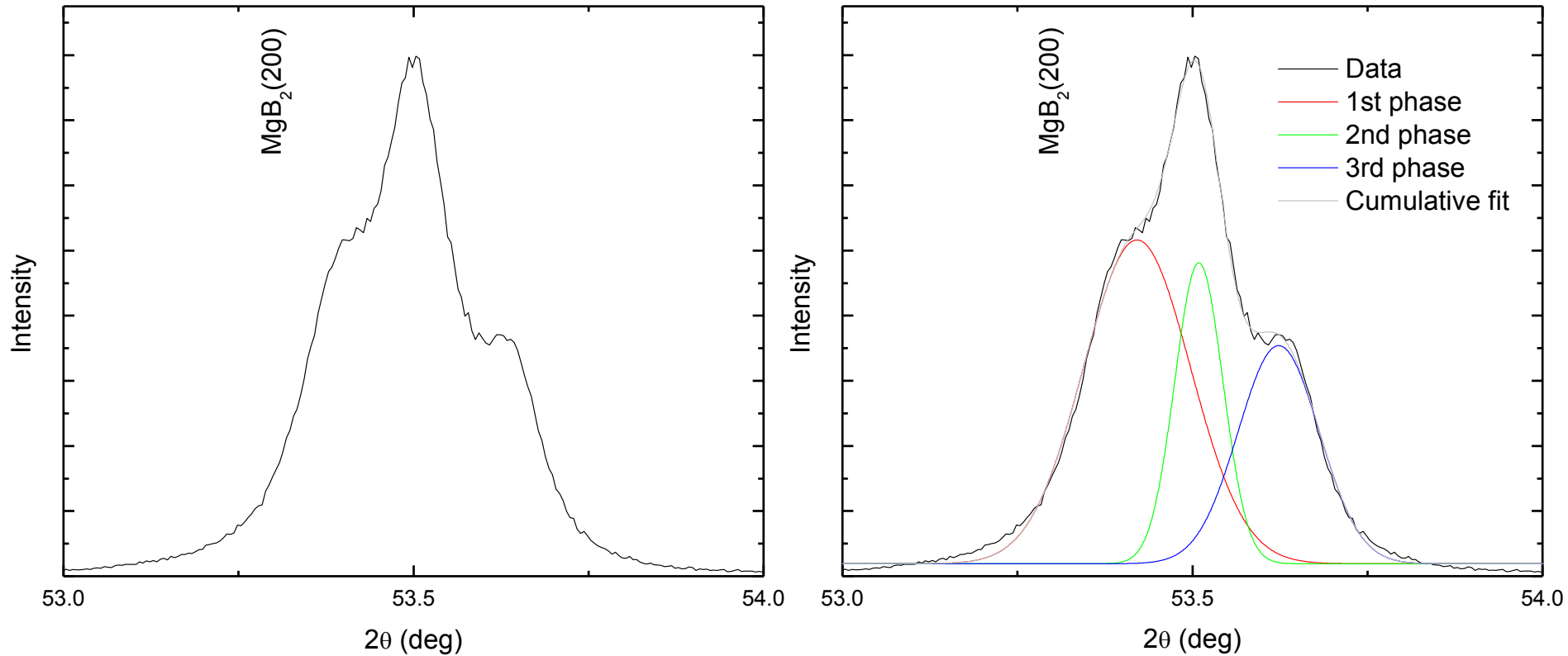
MgO
(100)



XRD detail

XRD scan optimized on the $\text{MgB}_2(200)$ peak.

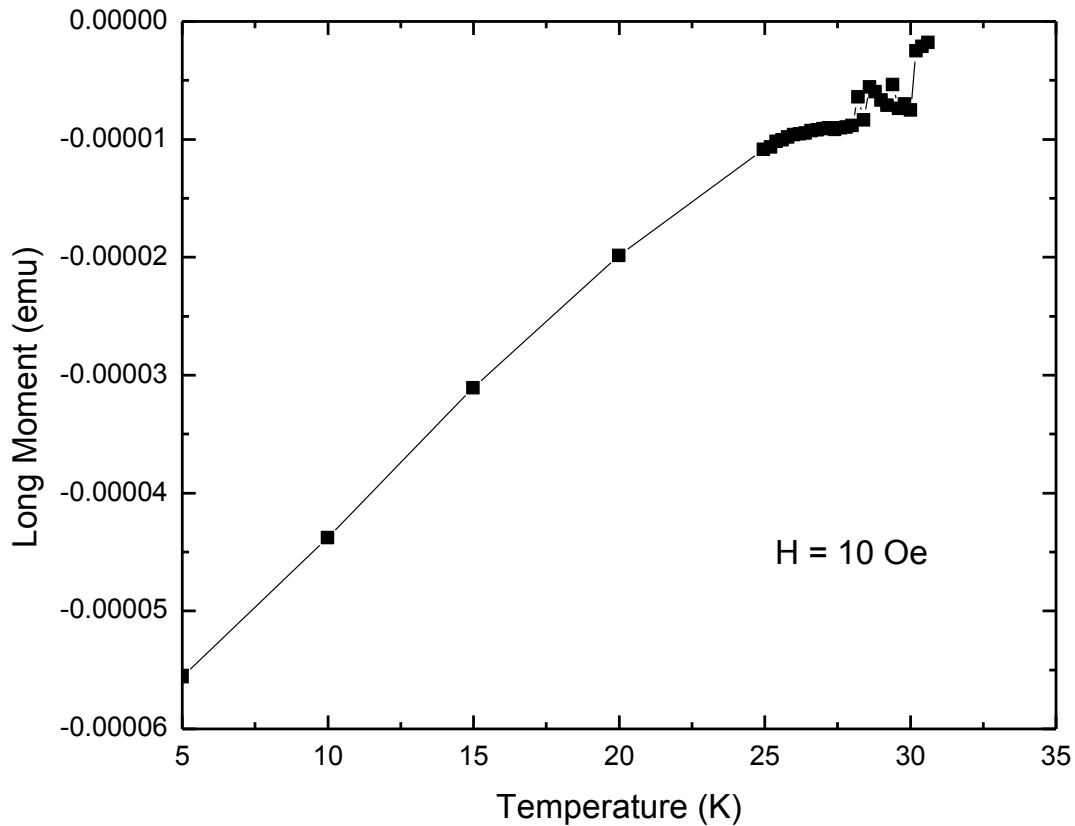
The scan indicates that there are multiple MgB_2 phases present, all strained



Bulk 3.523 Å
(2θ = 51.863°)

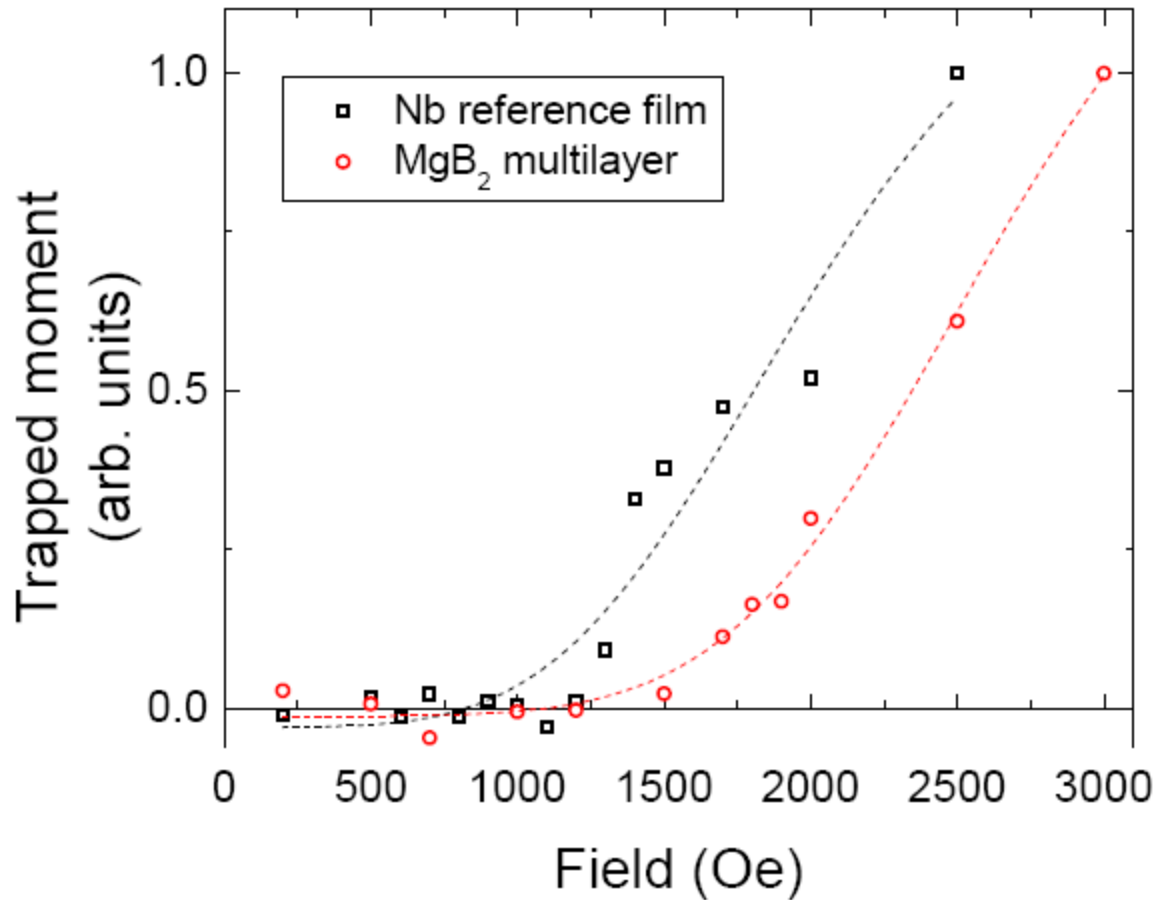
1 st phase	3.4275 Å	2.7108% strain
2 nd phase	3.4223 Å	2.8584% strain
3 rd phase	3.4155 Å	3.0514 % strain

SQUID characterization



$T_c \sim 30.2 \text{ K}$ (recall that bulk $T_c = 39\text{K}$)

SQUID measurements



Reference Nb penetration field ~ 1300 Oe; MgB₂-based ML penetration field ~ 1700 Oe

Surface impedance characterizations (SIC)

- The ultimate performance test of the films and multilayers for this application is a measurement of their surface impedance, R_s .
- We note that R_s can be written as:

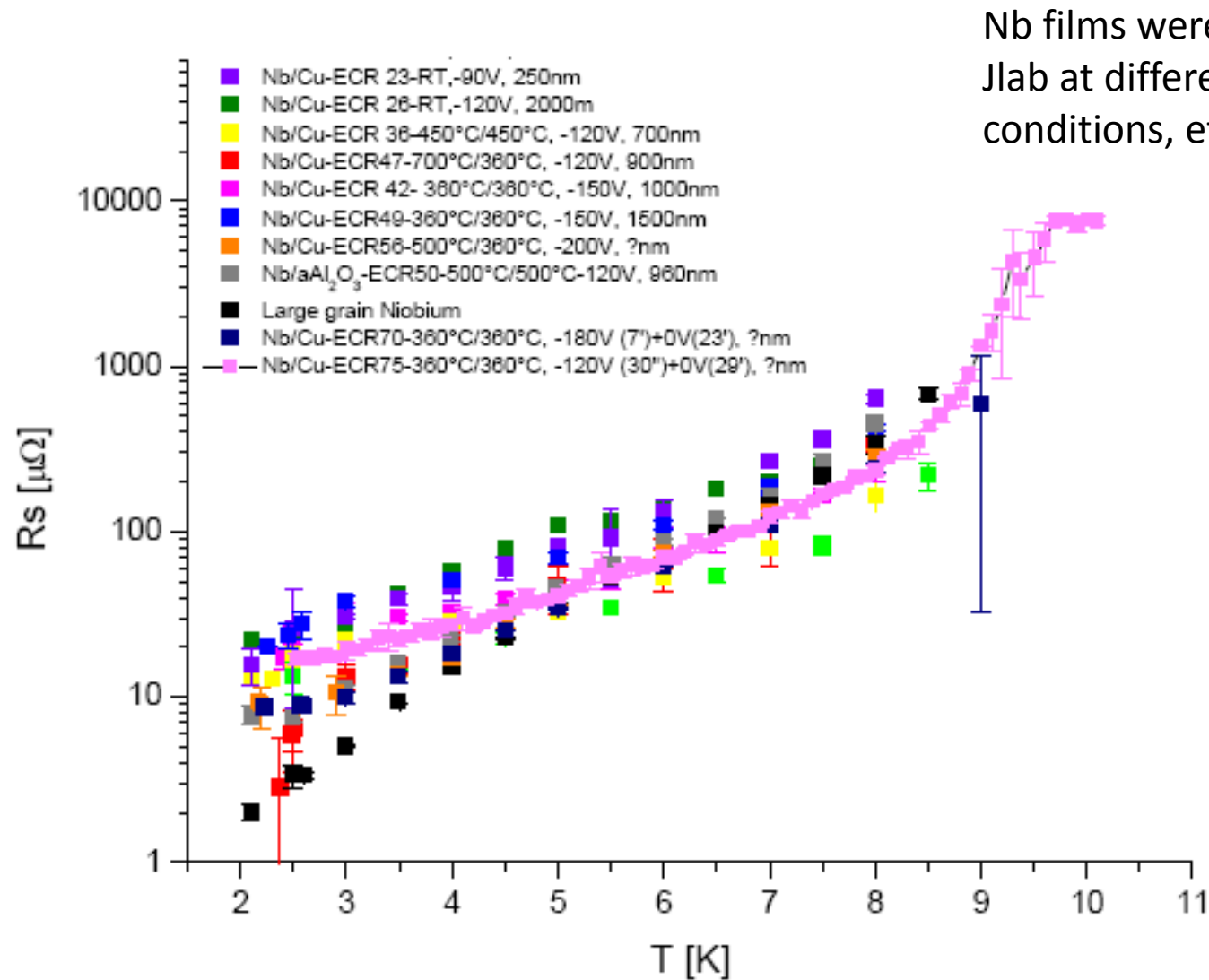
$$R_s = R_{\text{BCS}}(T) + R_i,$$

where $R_{\text{BCS}} = (A\omega^2/T) \exp[-\Delta/(k_B T)]$

Note: R_s decreases strongly for higher- T_c materials with larger superconducting gap $\Delta = 1.86T_c$, implying that materials with $T_c > 20\text{K}$, could have the R_{BCS} at **4.2 K** comparable to R_{BCS} of Nb at **2K**. However, small R_s also implies **small residual resistance R_i** and no nodes in the superconducting gap, which rules out the d-wave high- T_c cuprates for which $R_s \propto T^2$

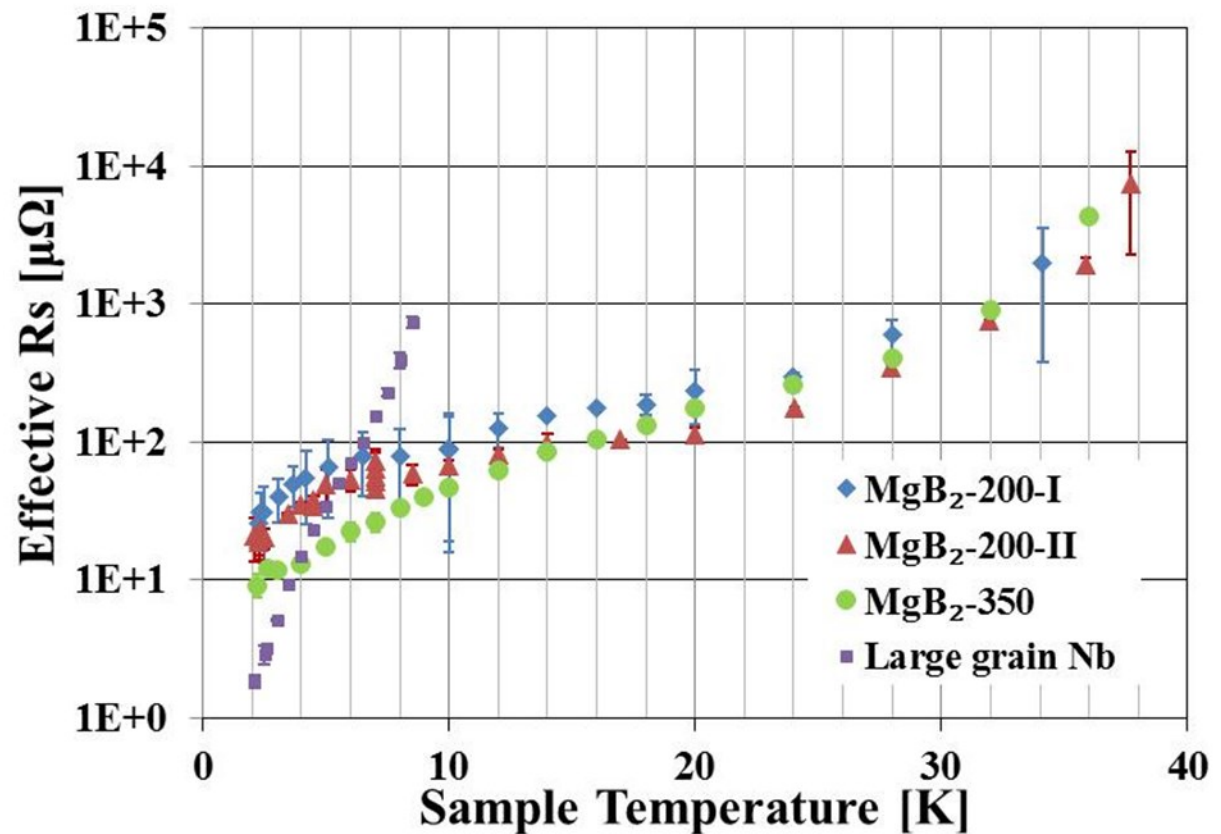
- JLab's SIC system is uniquely capable of making temperature-dependent RF surface impedance measurements on 2inch-sized thin film samples.
- Summary of 2 inch samples studied:
 - Nb thin films grown under various conditions
 - Epitaxial MgB_2 films
 - NbN-based trilayer on copper
 - NbTiN-based multilayers deposited on sapphire

Nb thin films grown under different conditions



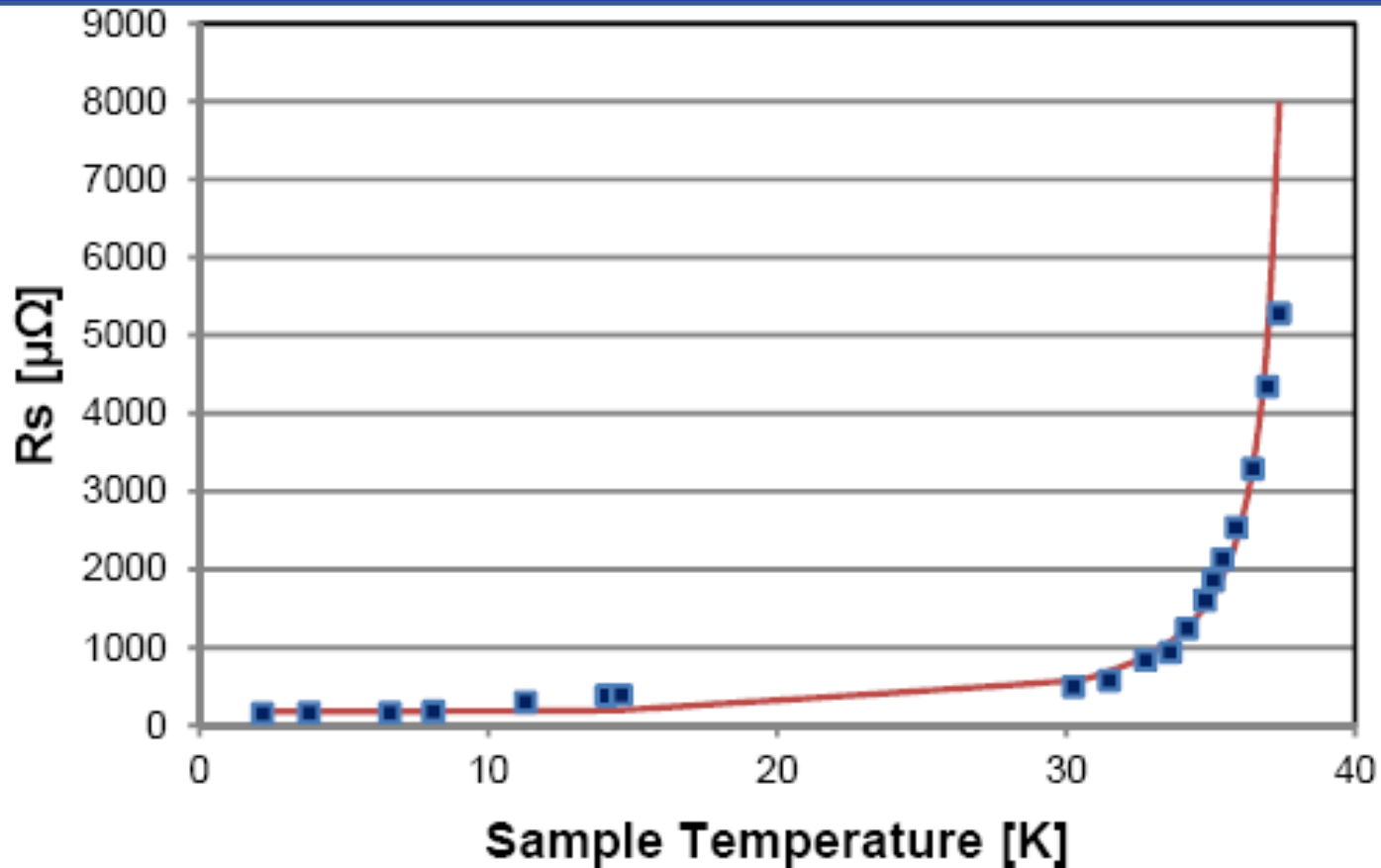
MgB₂ films epitaxially grown on sapphire

MgB₂ films produced by Temple Univ. were measured calorimetrically at 7.5 GHz



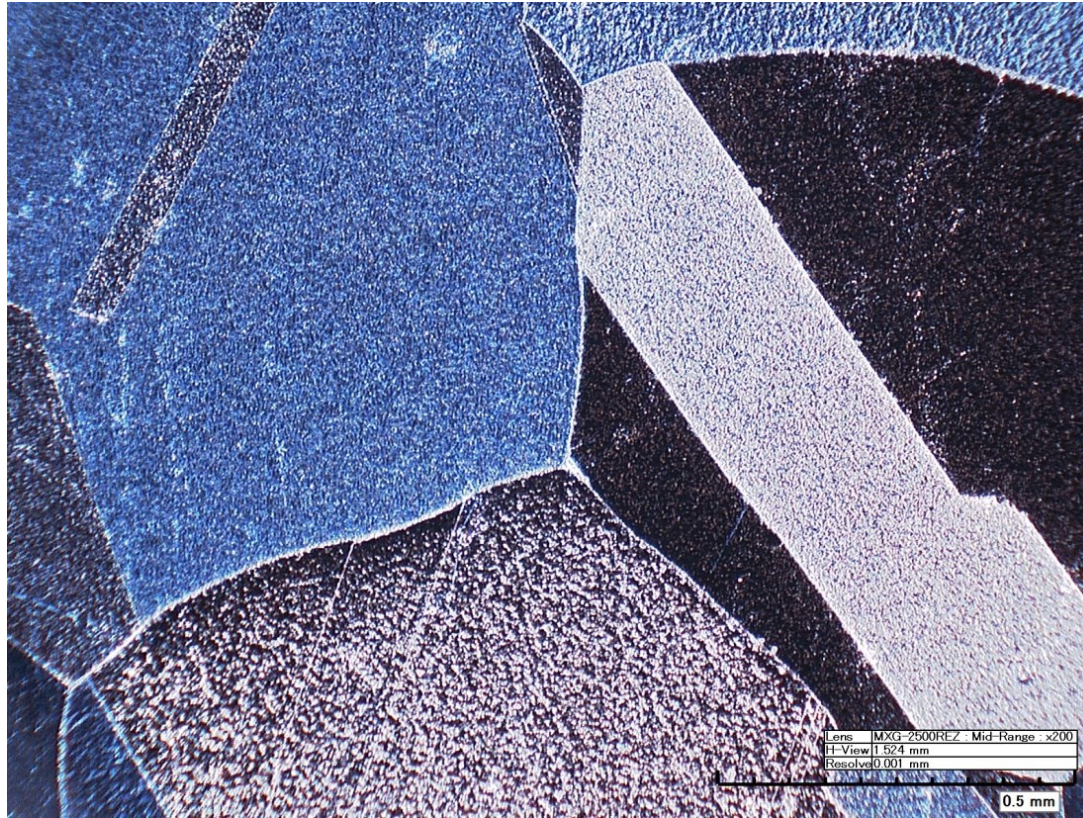
Effective surface resistance of 200 nm and 350 nm MgB₂ on sapphire substrates, together with surface resistance of large grain Nb sample. (7.5 GHz)

MgB₂ based SIS structure



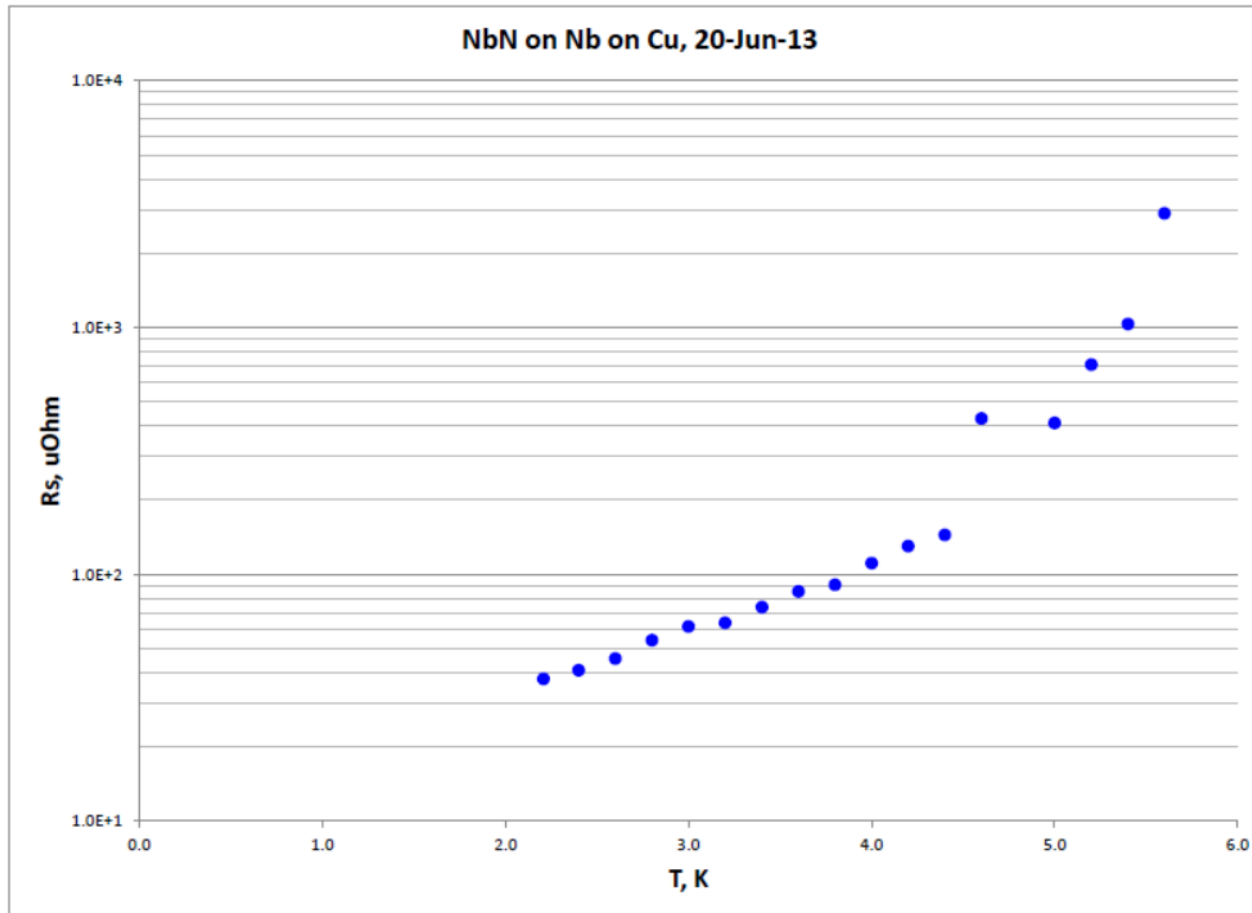
At the SRF 2011 the surface impedance of a MgB₂-based multilayer was reported, and the residual resistance was found $\sim 181 \mu\Omega$

NbN-based SIS samples deposited on Cu substrates



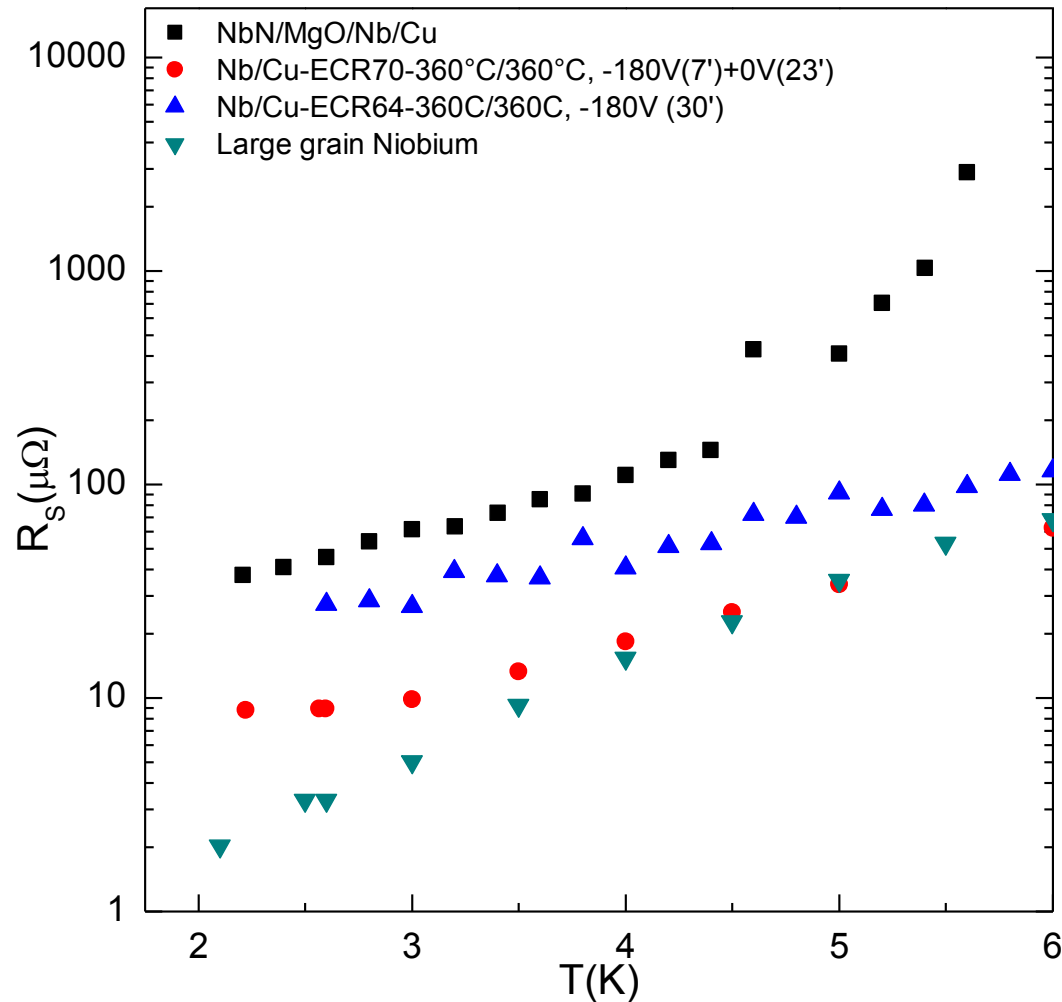
Samples grown on Cu substrates contained large grains (on the order of millimeters) that were visible to the naked eye. The surfaces of these samples are dominated by rough features as seen with SEM and optical microscopy. After annealing 10 nm of MgO and 60 nm of NbN were deposited (W&M).

SIC for NbN/MgO/Nb trilayer on Copper



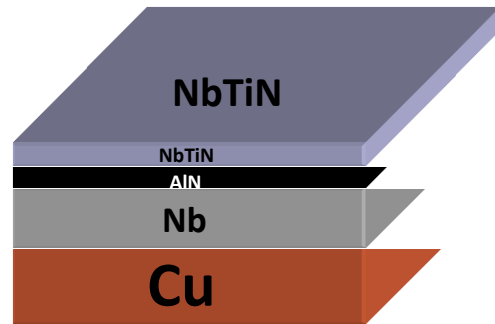
We note that the residual resistance (R_i) around 2K is approximately 35 $\mu\Omega$.

SIC comparison of different thin film and ML samples

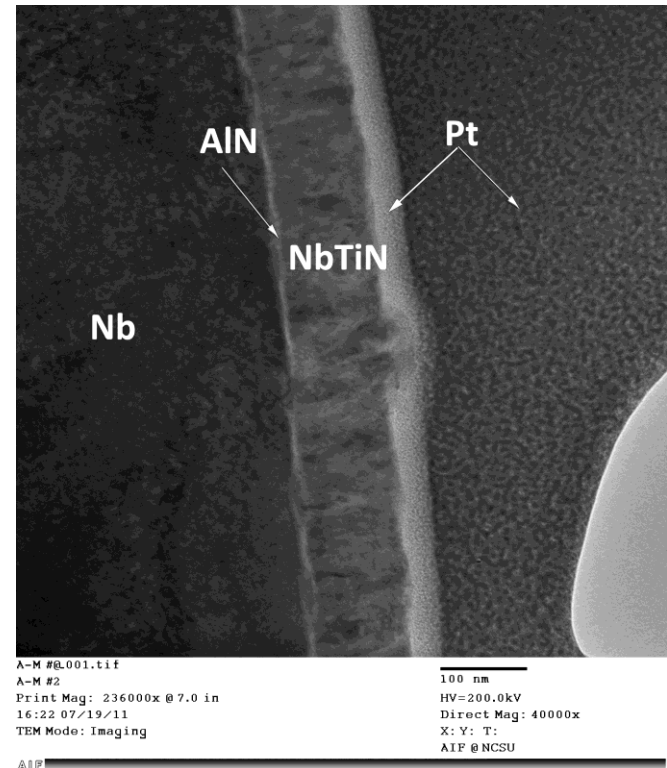


We notice that the residual resistance of these thin film samples is one order larger than that of bulk Nb around 2K but in general lower than that of MgB₂-based ML samples.

NbTiN-based SIS structures on bulk Nb and Nb/Cu substrates

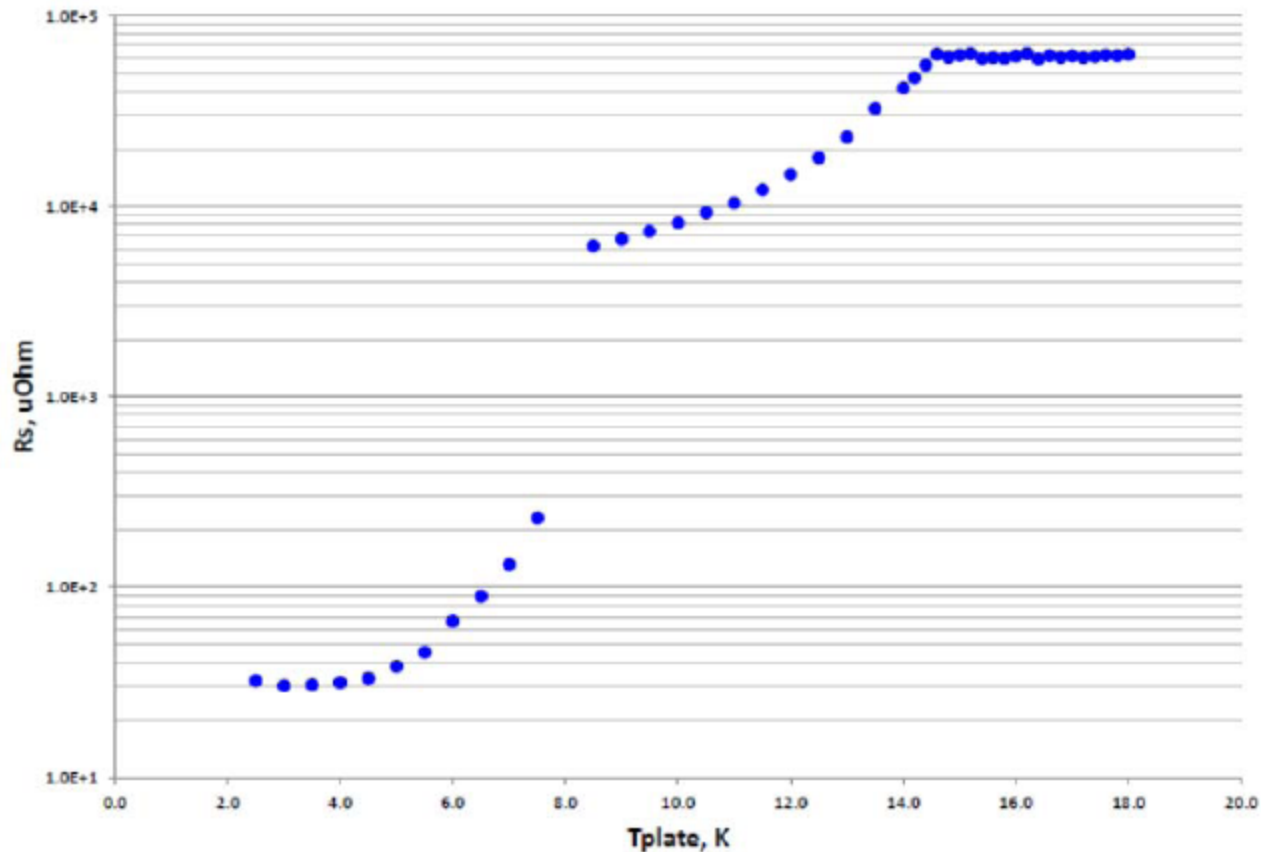


	AlN	NbTiN
N ₂ /Ar	0.33	0.23
Total pressure [Torr]	2x10 ⁻³	2x10 ⁻³
Sputtering Power [W]	100	300
Deposition rate [nm/min]	~ 5	~ 18
Thickness [nm]	10	100
T _c [K]	N/A	14



NbN-based multilayers have been very promising regarding magnetic shielding, but we note that this material suffers from a higher resistivity due to the presence of both metallic and gaseous vacancies randomly distributed while the ternary nitride NbTiN presents all the advantages of NbN and also exhibits increased metallic electrical conduction properties with higher titanium (Ti) percentage.

SIC for NbTiN/AlN/Nb trilayer on sapphire



We note that the residual resistance (R_i) around 2K is approximately 30 $\mu\Omega$. We also distinguish two temperature regimes with transitions around 8.5 K and 14.5K, related to Nb and NbTiN respectively.

Conclusions

- Trilayers incorporating NbN and following the “Gurevich model” were shown to shield niobium in the pioneer work by Antoine *et al.* using *SQUID* magnetometry as well as third harmonic analysis.
- By tailoring thin film growth parameters, and also using *SQUID* magnetometry we were able to demonstrate shielding beyond the critical field of Nb also using NbN-based trilayers.
- We have also demonstrated that other suitable superconductors show promise for SRF applications, but further studies to optimize thin film deposition conditions must be undertaken in this case.
- This work is supported by the Defense Threat Reduction Agency (DTRA) under grant # HDTRA1-20- 1-0072.