Energetic Condensation Growth of Nb films for SRF Accelerators *

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SRF-2011, Tuesday July 26, 2011

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Outline

- Motivation for thin films in SRF cavities
- History of thin film development
- Energetic Condensation
- Morphology of thin films produced by energetic condensation
- Thin films on Cu
- Future Plans

Motivation

- More than 10000 particle accelerators worldwide; most use *normal* cavities
- Facility for Rare Isotope Beams (FRIB), ILC … large facilities
  - *NSAC report states that as a res… …al advances, a … rare isotope fa… can be built at ≈ half the c… …iginally planned R… …pe Accelerator (R… employing* ***a supe… …g linac***
- SRF at 2K is g… …t operating at … …d further redu… …osts as the cryogenic cooling moves tow… …shelf cryo-co…
- Replacing bulk Nb w… …ated Cu caviti… …also reduce costs
- The ultimat… …would be fro… …ust Al SRF cavities coated with higher temperature supercondu… …, Mo_3Re, Nb_3Sn, MgB_2, oxypnictides)

SRF uses less power than normal

SRF with cheaper cavity materials (Cu) would be better

SRF with cheaper cavity materials and high-T_c thinfilms would be better still

AASC's thin film superconductor development is aimed at these goals

Two examples of early SRF thin film development

LEP at CERN tested Cu cavities Magnetron sputter coated with ~1.5µm Nb thin films (1984-)

- C. Benvenuti, N. Circelli, M. Hauer, Appl. Phys. Lett. 45, 1984. 583; C. Benvenuti, Part. Accel. 40 1992. 43; C. Benvenuti, S. Calatroni, G. Orlandi in: 20th International conference on Low Temperature Physics, Eugene 1993, to be published in Physica B; C. Benvenuti, S. Calatroni, I.E. Campisi, P. Darriulat , M.A. Peck,R. Russo, A.-M. Valente, Physica C316, 1999, 153–188, (Elsevier): *Study of the surface resistance of superconducting niobium films at 1.5 GHz*
  - *RRR=11.5 on oxide coated Cu, 29 on oxide-free Cu coated at 150^o C*

- G.Amolds, Doktorarbeit, University of Wuppertal, WUB 79-14 (1979)
  - Wuppertal studied 8, 3 and 1GHz Nb cells vapor deposition coated with Nb_3Sn

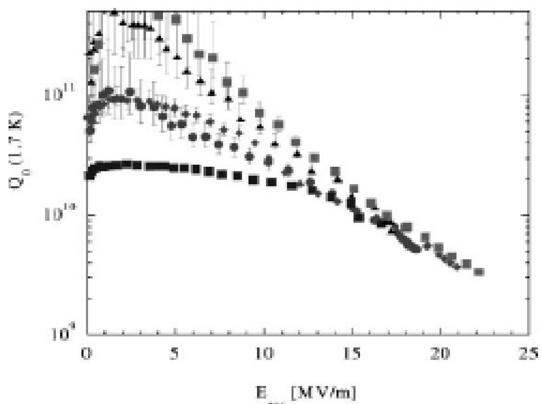
Limitations of Magnetron sputtering and Vapor Deposition

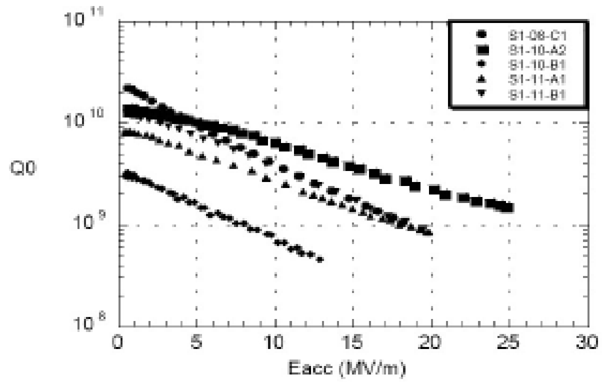
Thin Film Cavities & Q-Drop

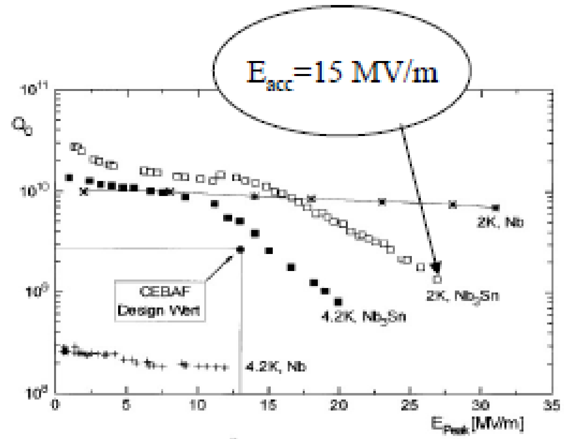
dapnia
cea
saclay

Advantages to use Thin Film Technology for SRF Cavities :
Reduced Cost - New Superconducting Material (higher T_c & H_{sh})

severe Q-drop limits High Gradient Performances E_{acc} < 25 MV/m



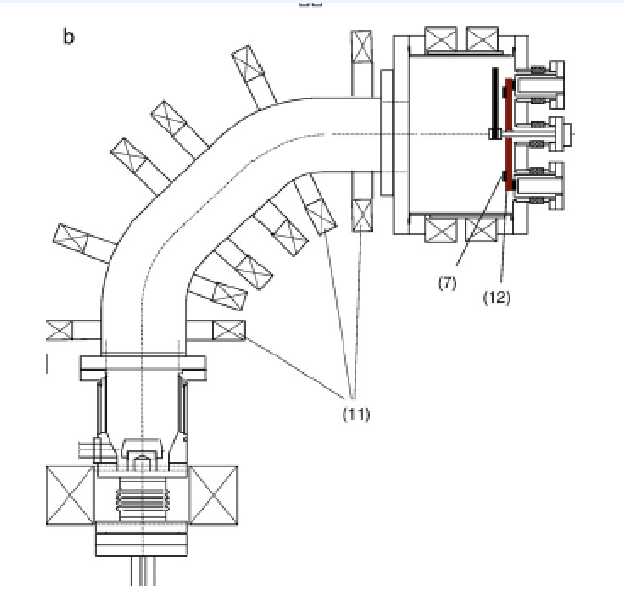




CERN – Nb / Cu – 1.5 GHz	Saclay – Nb / Cu – 1.5 GHz	Wuppertal – Nb_3Sn / Nb – 1.5 GHz
Magnetron Sputtering in Kr	Magnetron Sputtering in Ar	Vapor Deposition Technique
V. Arbet-Engels et al.- NIMA (2001)	P. Bosland et al.- ASC (1998)	G. Müller et al.- EPAC (1996)

(no field emission, no quench only RF power limitation)

Prior UHV Arc Nb thin films: Tor Vergata, Rome/IPJ, Poland



- R. Russo, A. Cianchi, Y.H. Akhmadeev, L. Catani, J. Langner, J. Lorkiewicz, R. Polini, B. Ruggiero, M.J. Sadowski, S. Tazzari, N.N. Koval, Surface & Coatings Technology 201 (2006) 3987–3992
  - Base pressure of 1–2$\times$10^{-10} mbar in the system is reached after one night bake at 150 °C, but pressure increased to 10^{-7} mbar during run
  - Laser trigger to minimize impurities in Nb film
  - Magnetic sector filter to reduce macro-particles in film
  - Lattice parameter (from XRD spectra) showed much lower stress than observed in niobium deposited by magnetron sputtering
  - Surface roughness was ~few nm on sapphire and on Cu, was comparable to that of the Cu substrate itself
  - RRR of 20-50 was measured on ~1µm Nb films deposited on Quartz at room temperature (higher than typical sputtering values)

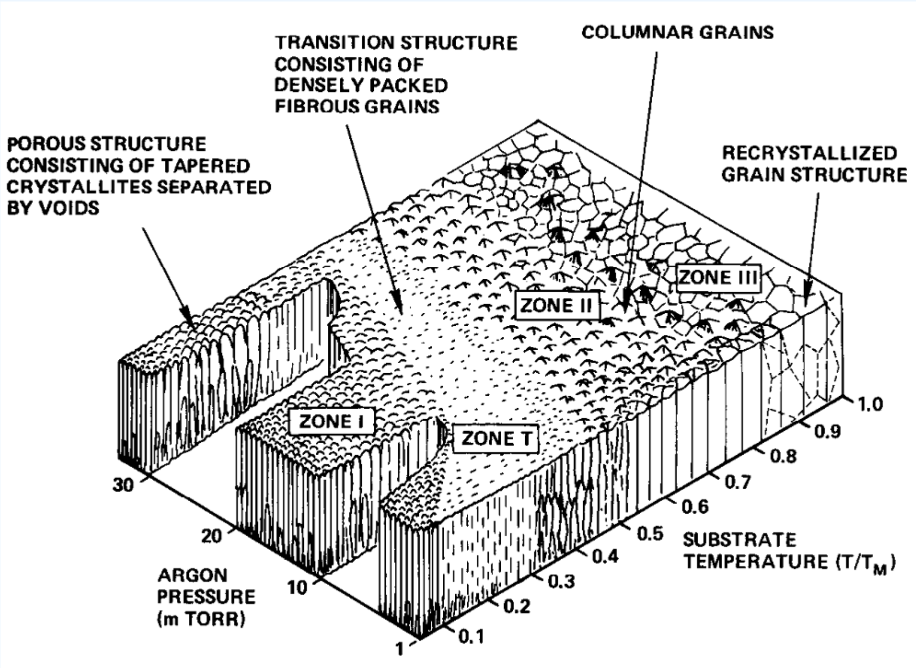
RRR up to 80 was reported with substrate heated to 200^{o}C

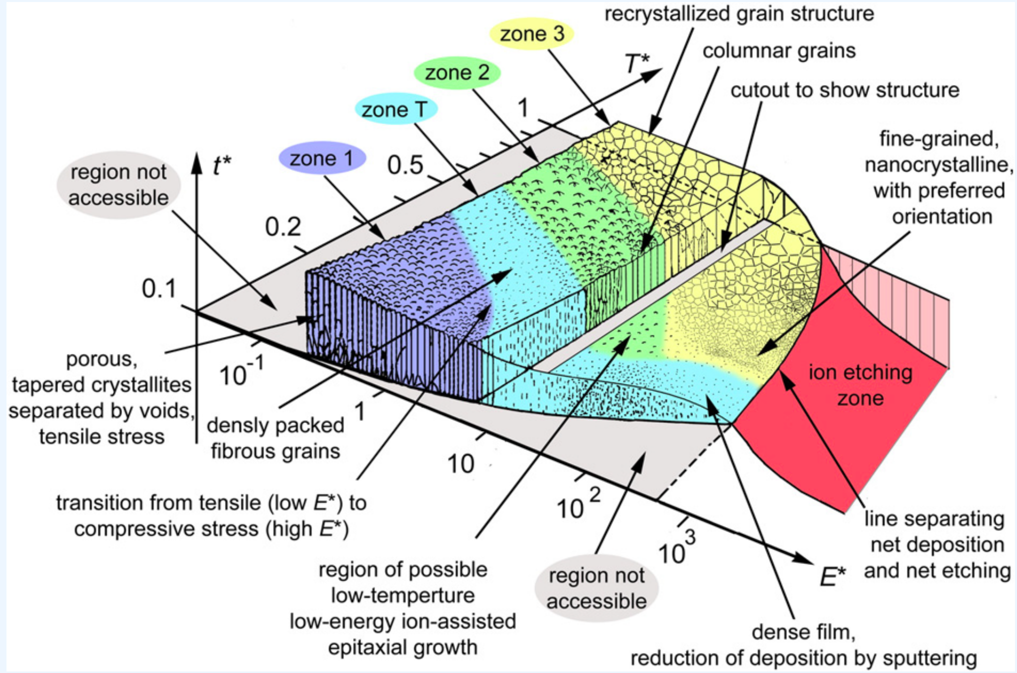
  - Dominant impurity was H atoms outgassed from Nb cathode (needed careful bakeout of cathode to minimize this problem
- J. Langner, R. Mirowski, M.J. Sadowski, P. Strzyzewski, J. Witkowski, S. Tazzari, L. Catani, A. Cianchi, J. Lorkiewicz, R. Russo, Vacuum 80 (2006) 1288–1293

Outline

- Motivation for thin films in SRF cavities
- History of thin film development for SRF cavities
- Energetic Condensation (a very brief primer)
- Morphology of thin films produced by energetic condensation
- Thin films on Cu
- Future Plans

Energetic Condensation





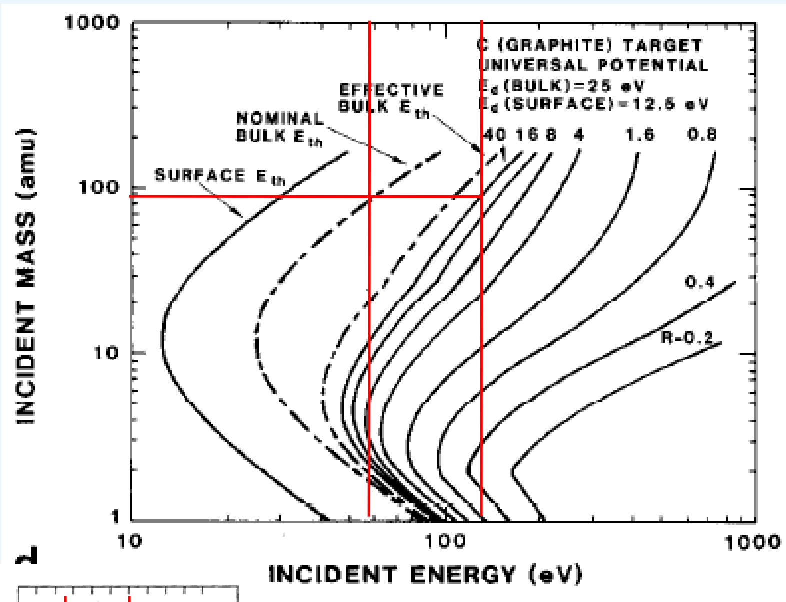
J.A. Thornton, "Influence of substrate temperature and deposition rate on the structure of thick sputtered Cu coatings", J. Vac. Sci. Technol. Vol. 12, 4 Jul/Aug 1975

Andre Anders, A structure zone diagram including plasma-based deposition and ion etching, Thin Solid Films 518 (2010) 4087–4090

- In Energetic Condensation, the ions deposit energy in a sub-surface layer (≈3 atomic layers deep for ~100eV Nb ions), shaking up the lattice, causing adatom mobility and promoting epitaxial crystal growth
- Energetic Condensation when combined with substrate heating promotes lower-defect crystal growth

Subplantation Models

- Y. Lifshitz, S. R. Kasi, J.Rabalais, W. Eckstein, Phy. Rev. B Vol. 41, #15, 15 May 1990-II: Subplantation model for film growth from hyperthermal species
- D.K. Brice, J.Y. Tsao and S.T. Picraux: PARTITIONING OF ION-INDUCED SURFACE AND BULK DISPLACEMENTS
- W. D. Wilson, Radiat. Eff. 78, 11 (1983)



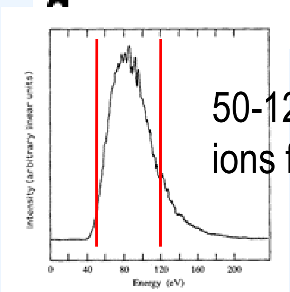
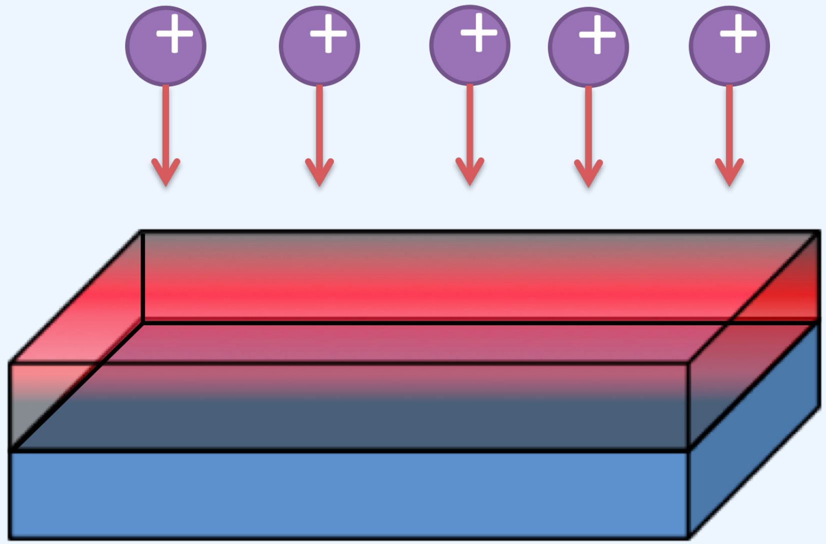


Figure 1 Energy distribution of Nb ions from a 95 A d.c. filtered arc.

50-120eV energy spread in Nb ions from cathodic arc

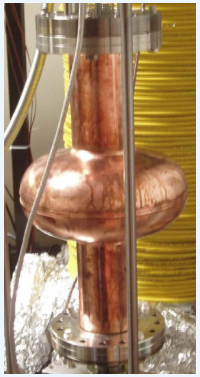
* A. BENDAVID, P. J. MARTIN, R. P. NETTERFIELD, G. J. SLOGGETT, T. J. KINDER, C., ANDRIKIDIS, JOURNAL OF MATERIALS SCIENCE LETTERS 12 (1993) 322-323



Thermal-Vacuum Stability of the Surface Oxide Complex on Cu
M. Bagge-Hansen, R.A. Outlaw, K. Seo, C. Reese, J. Spradlin, Anne Marie Valente-Feliciano and D.M. Manos, Journal of Vacuum Science and Technology (2011), in press

Challenges for thin film SRF: path to success

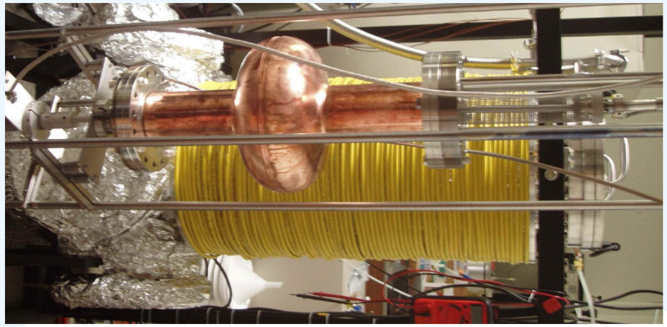
- Cu and/or Al cavity substrates might be of two different forms

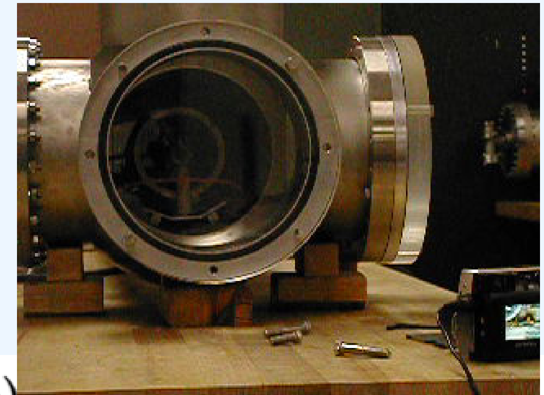


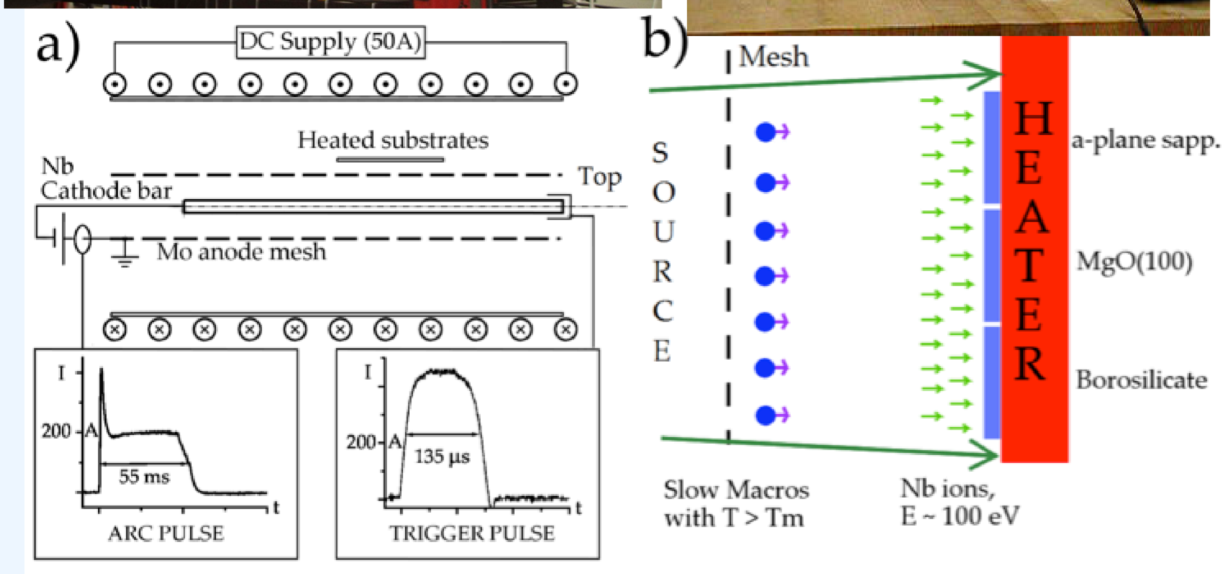
- How do we grow low-defect Nb films on such substrates?
- Study adhesion, thickness, smoothness, RRR, stability
- Understand these issues at the coupon level
- Proceed to RF cavity level and measure Q at high fields
- Install multi-cell Nb coated Cu modules in SRF accelerator and validate the thin film solution
  - Spur acceptance of thin film Nb by accelerator community
- Continue R&D towards higher T_c films and Al cavities

Coaxial Energetic Deposition (CED) Coater at AASC

Coaxial Energetic Deposition (CED™)





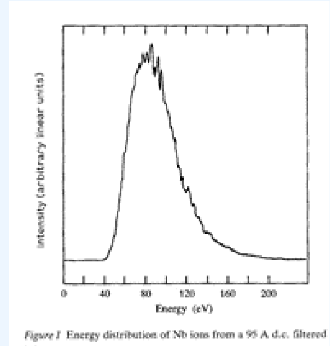


- CED coater uses "welding torch" technology
- Arc source is scalable to high throughputs for large scale cavity coatings
  - Present version deposits ≈1 monolayer/pulse in ≈1ms
- Russo's and Langner's emphasis on UHV and clean walls is important

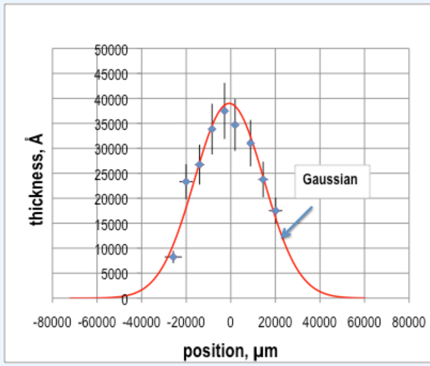
CAD: Pulsed Biased deposition; Dual Targets

Cathodic Arc Deposition (CAD)

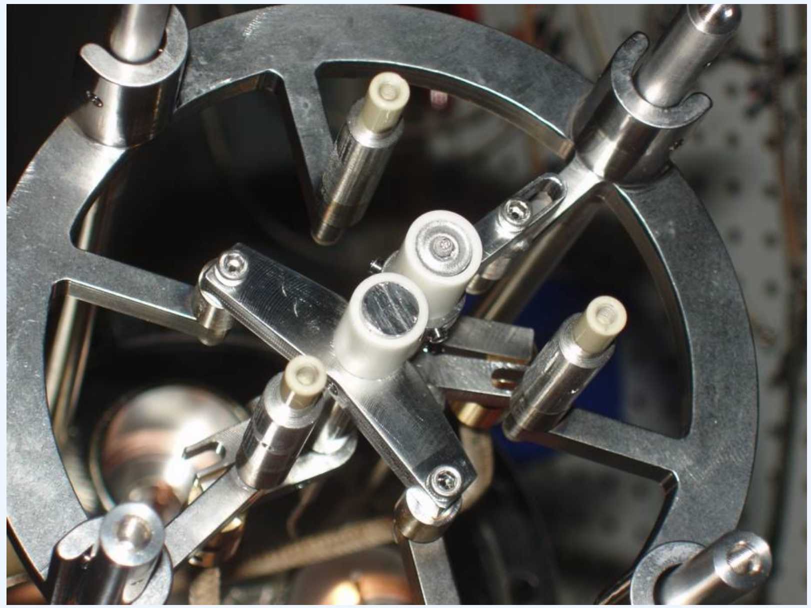
Pulsed Bias capability



* A. BENDAVID, P. J. MARTIN, R. P. NETTERFIELD, G. J. SLOGGETT, T. J. KINDER, C., ANDRIKIDIS, JOURNAL OF MATERIALS SCIENCE LETTERS 12 (1993) 322-323



vol under gaussian=	0.012	cc
mass under gaussian=	0.100	g
mass/shot=	14.3	μg
charge/shot=	0.5	C
anode transm.=	0.7	
erosion rate=	41	μg/C
peak thickness/pulse=	5.6	Å
instant. rate=	5600	Å/s



Dual Target source for Nb_3Sn, Mo_3Re, MgB_2 etc.

Please visit Poster THPO077 on Thursday: Mo-Re Films for SRF Applications", Dr. Enrique Valderrama

Collaborative progress made by AASC and JLab

- AASC's UHV Arc coater (CED/CAD) and JLab's ECR source are both Energetic Condensation Coaters
  - The CED/CAD plasma generates Nb^{2+} and Nb^{3+} ions with 40-120eV energy spread
  - The ECR plasma also produces ~100eV ions by biasing the substrate, with Nb^{1+} ions
- AASC and JLab collaborate on thin film development for SRF. Over the past year, we have achieved record RRR values in Nb films
  - RRR=585 in Nb on MgO using CED
  - RRR=488 in Nb on a-sapphire using ECR
  - RRR=289 in Nb on LG Cu using ECR

Please see A-M Valente Feliciano' s talk later in this session!

- We are learning more about the epitaxial growth modes on different substrates, building a knowledge base from which to proceed to Cu cavity coatings in the near future

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- History of thin film development
- Energetic Condensation
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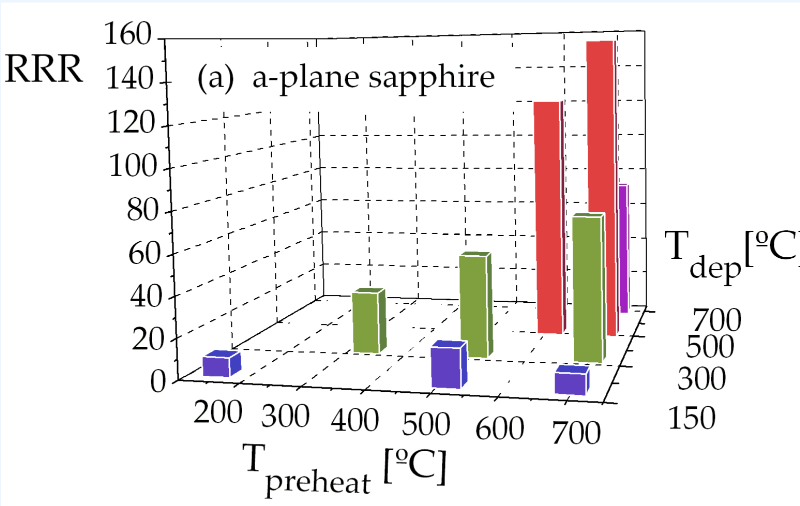
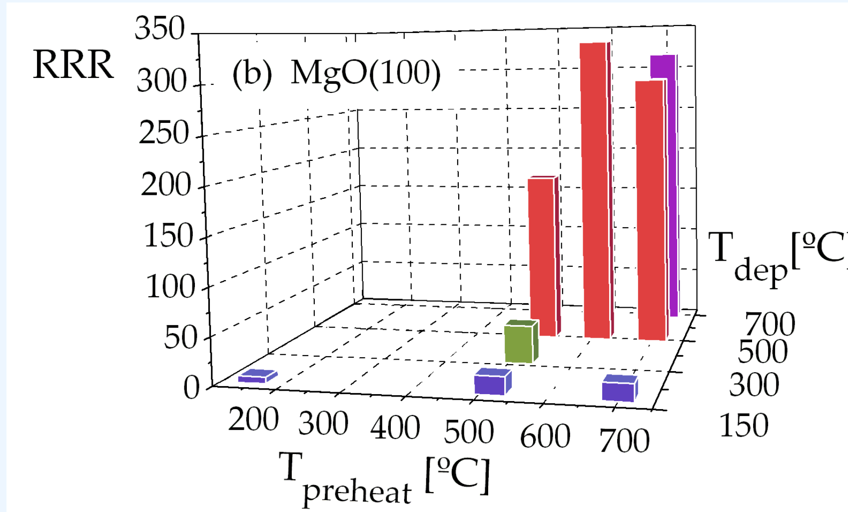
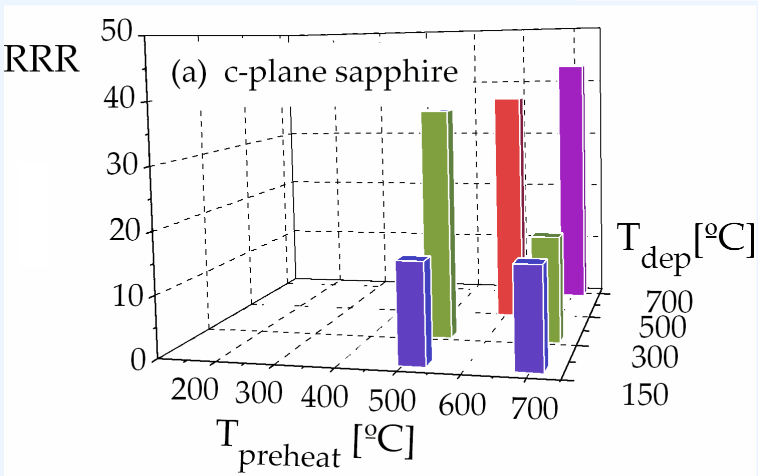
RRR of Nb thin films on sapphire and MgO substrates

- Nb thin films grown on sapphire and MgO crystals have demonstrated higher levels of RRR than were reported by the pioneers, and XRD spectra reveal crucial features







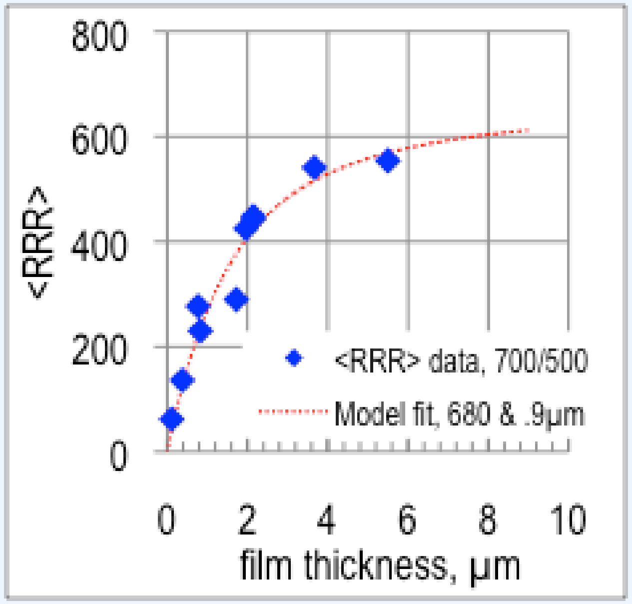
RRR-585 (±1% error) measured on 5µm film on

RRR-330 measured on a-sapphire

- M. Krishnan, E. Valderrama, B. Bures, K. Wilson-Elliott, X. Zhao, L. Phillips, Anne-Marie Valente-Feliciano, Joshua Spradlin, C. Reece, K. Seo, submitted to Superconducting Science and Technology

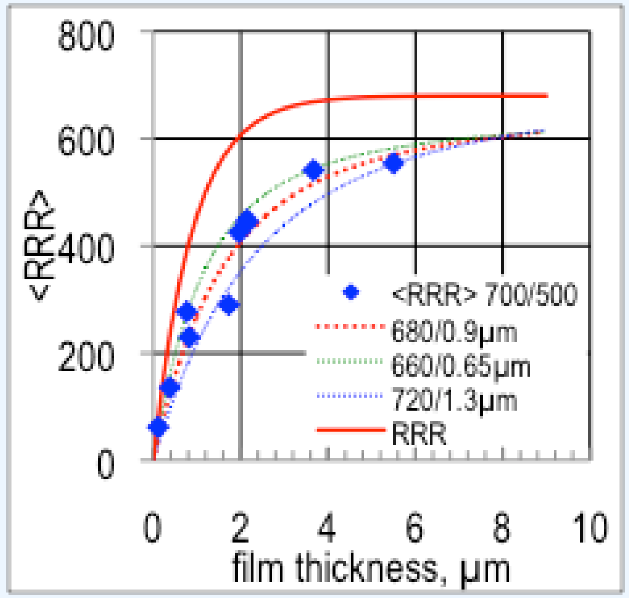
Nb on MgO: Local RRR vs. averaged <RRR>

- <RRR> vs. film thickness



$$< RRR > = \frac{1}{x_o}\int_0^{x_o} R_o * \left(1 - e^{-\frac{x}{x^*}}\right) dx = R_o\left[1 - \frac{x^*}{x}\left\{1 - e^{-\frac{x}{x^*}}\right\}\right]$$

- Ad hoc model fit to <RRR> data

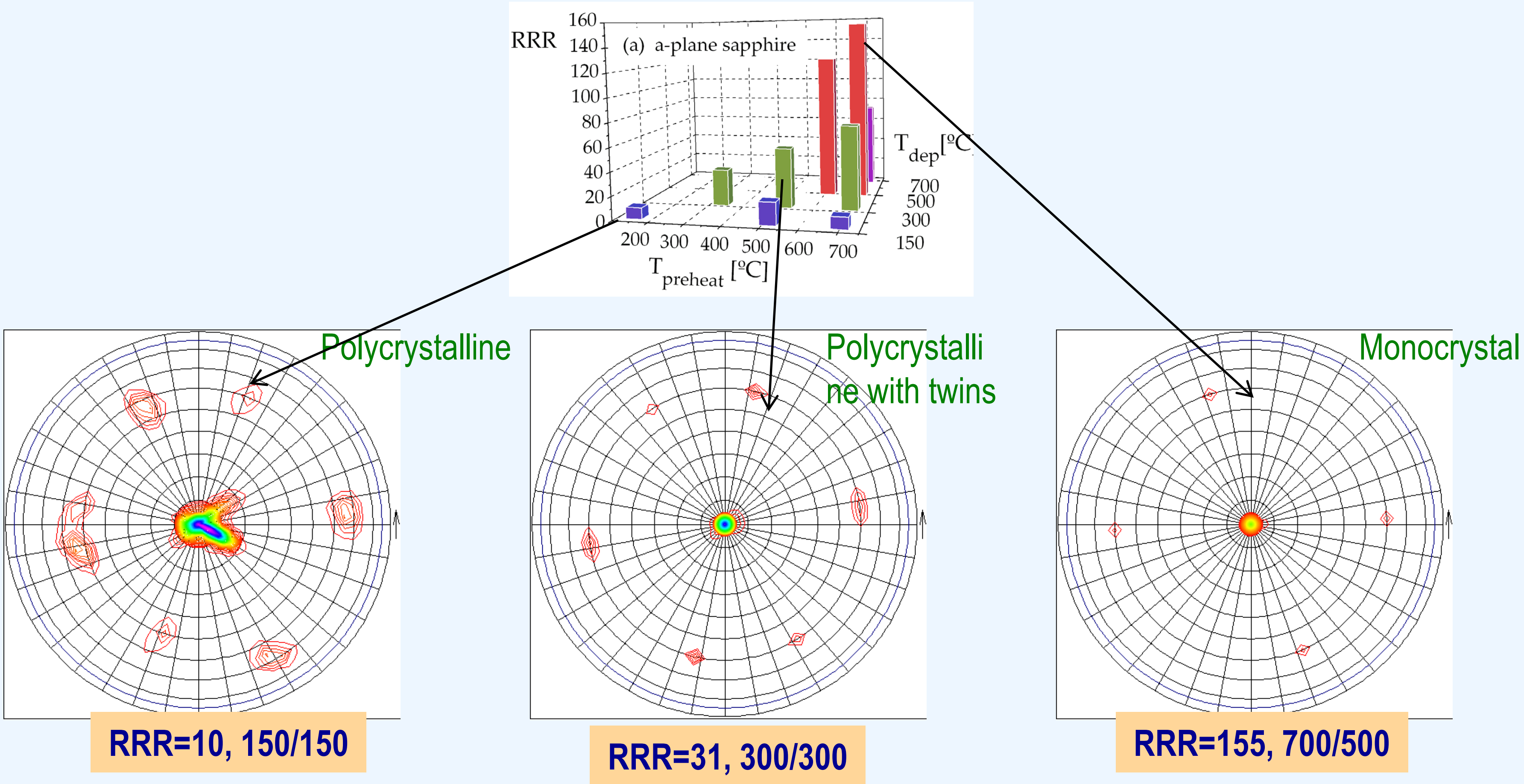


$$RRR = R_o * \left[1 - e^{-x/x^*}\right]$$

G. Orlandi, C. Benvenuti, S. Calatroni, F. Scalambrin, *Expected dependence of Nb-coated RF cavity performance on the characteristics of niobium, Proc. Of 6th workshop on RF Superconductivity, CEBAF, (1993)*

RRR of Nb thin films on a-sapphire substrates

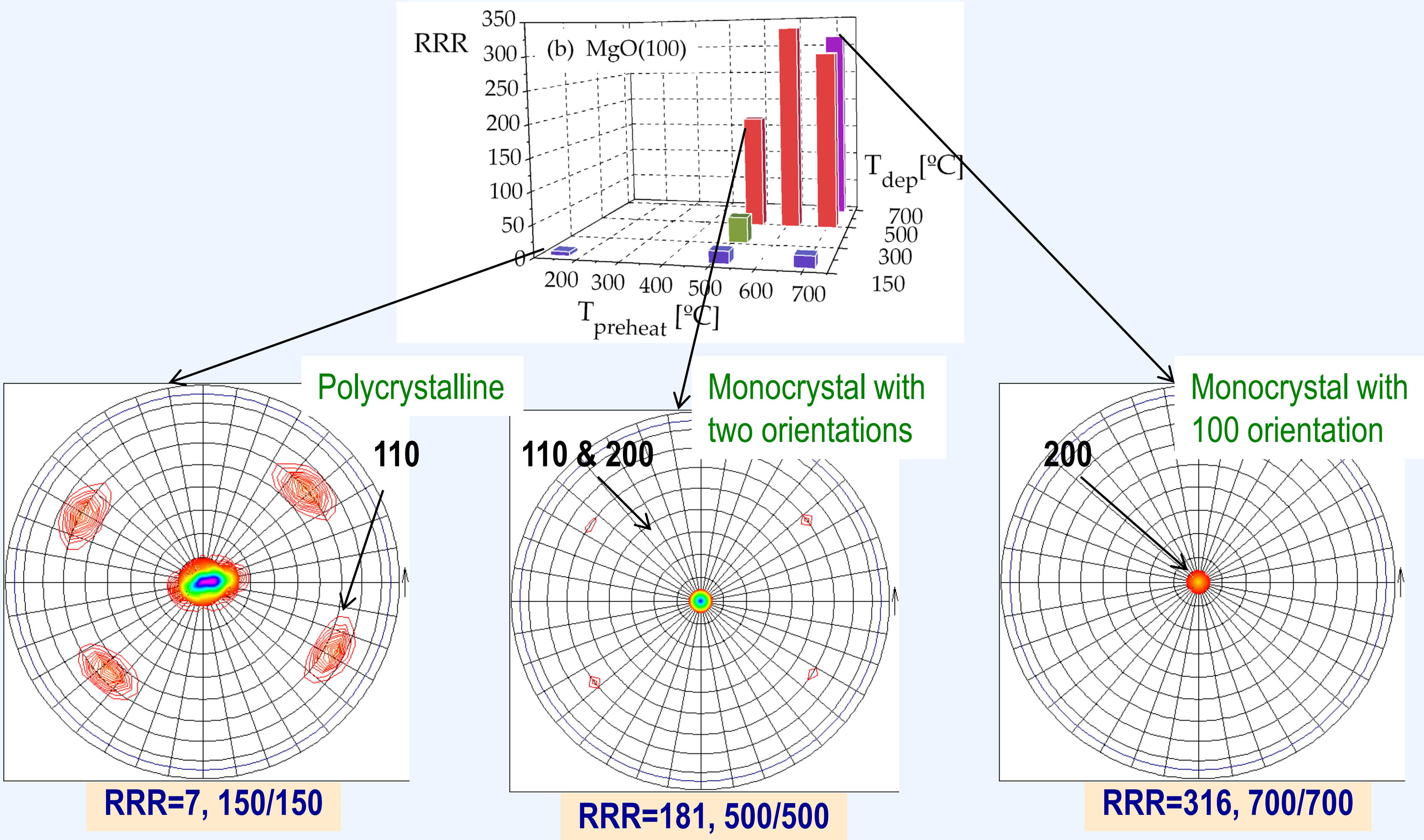
- XRD Pole Figures show that higher RRR correlates with higher crystalline order



- "Twin symmetry texture of energetically condensed niobium thin films on sapphire substrate (a-plane Al2O3)", X. Zhao, L. Philips, C. E. Reece, Kang Seo, M. Krishnan, E. Valderrama, Journal of Applied Physics, Vol 115, Issue 2, 2011, [in-press],

RRR of Nb thin films on MgO substrates

- Pole Figures show change in crystal orientation from 110 to 200 at higher temperature

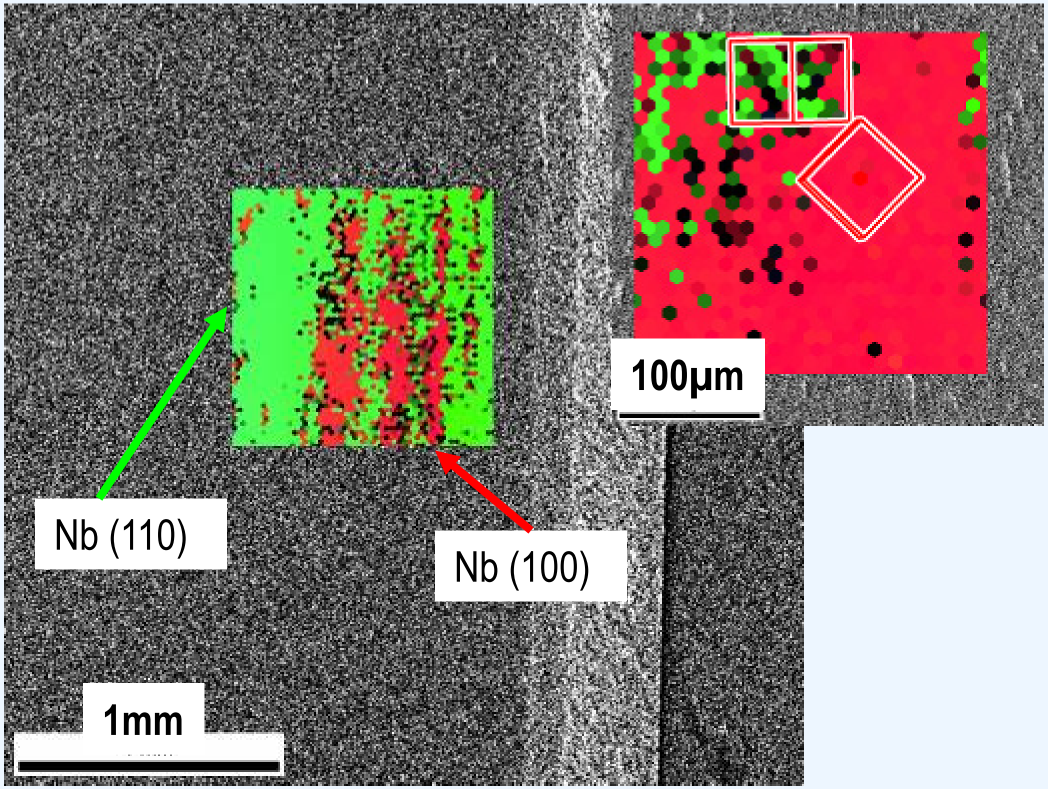


- Please visit poster THP0042 (given by Xin Zhao of JLab) for more details

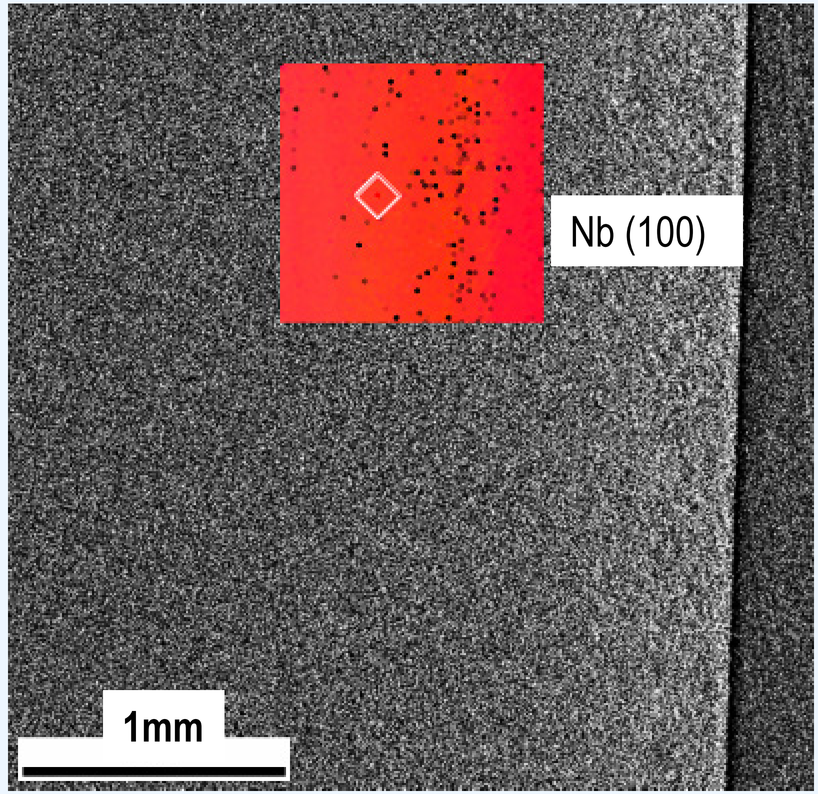
Nb on MgO: temp. driven transition in crystal orientation

- EBSD data shows intermixed Nb (110) and Nb (100) for lower RRR samples
- High RRR samples show pure Nb (100) for the entire surface

RRR = 196, 500/500



RRR = 333, 600/500



EBSD shows 110 to 100 transition (Pole figure captures the 200 plane):

Observations about bulk single crystal Nb orientation

PERFORMANCE OF SINGLE CRYSTAL NIOBIUM CAVITIES, **P. Kneisel**, G. Ciovati, TJNAF, Newport News, VA 23606, U.S.A. W. Singer, X. Singer, D. Reschke, A. Brinkmann, Proceedings of EPAC08, Genoa, Italy MOPP136

PROGRESS ON LARGE GRAIN AND SINGLE GRAIN NIOBIUM – INGOTS AND SHEET AND REVIEW OF PROGRESS ON LARGE GRAIN AND SINGLE GRAIN NIOBIUM CAVITIES, **P. Kneisel**, Proceedings of SRF2007, Peking Univ., Beijing, China

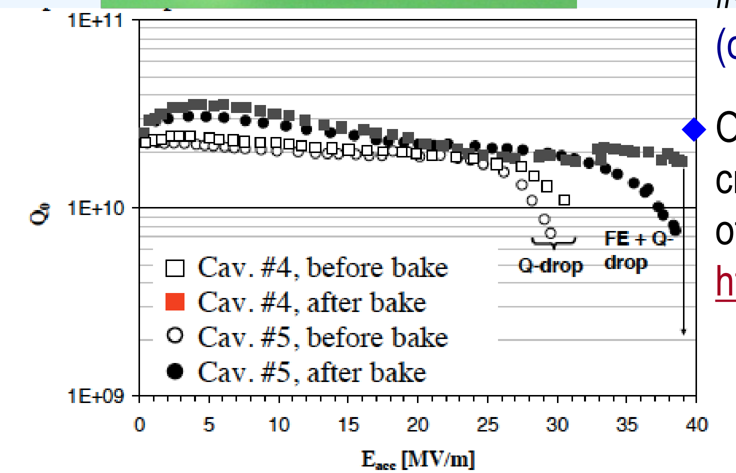


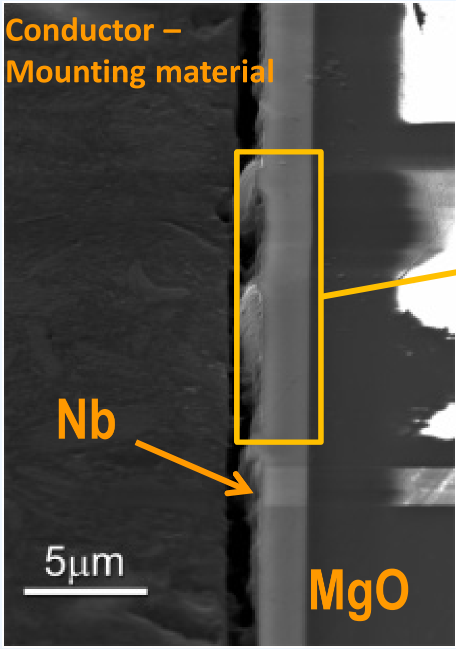
Figure 4: Q_0 vs. E_{acc} at 2 K for single crystal cavities #4 and #5 (best performance).

- Cavity #5 showed a different behavior than all other single crystal/large grain cavities after baking: the Q-drop did not disappear after 12 hours. The crystal orientation of the single crystals of this cavity was (110) with a tilt against the surface. For cavity #4 the crystal orientation was (100).
- The surface of both cavities appeared quite different after BCP: whereas cavity #4 exhibited a very smooth, shiny surface, the surface of cavity #5 was “rough” (orange peel/fish scale appearance) and less shiny.
- Obviously, there is a difference in the reaction of the BCP chemicals at different crystal orientations {D. Baars et al., ”Crystal orientation effects during fabrication of single or multi-crystal Nb SRF cavities”, SRF07, Beijing, Oct. 2007, TH102; http://www.pku.edu.cn/academic/srf2007/proceeding}

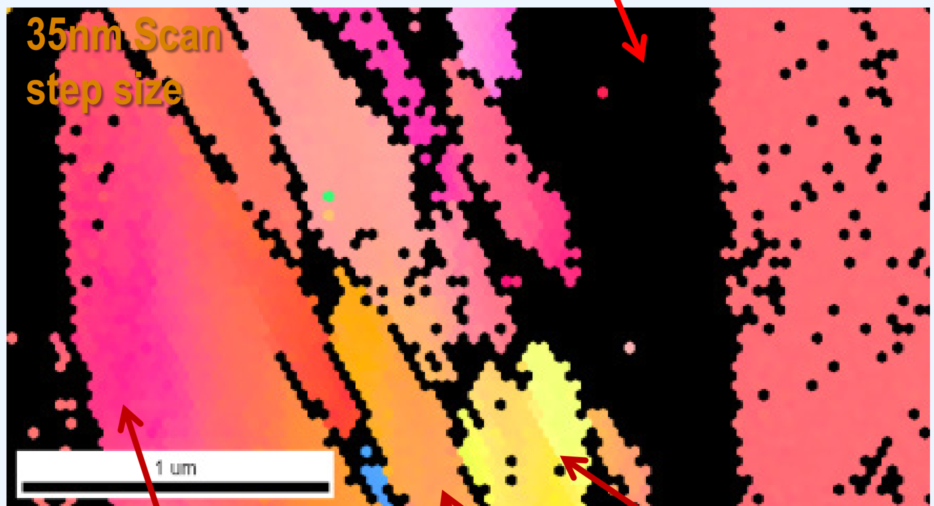
Could CED (Energetic Condensation) be used to grow (100) Nb films on existing Nb cavities to help improve performance?

RRR-7: Cross sectional EBSD (OIM)

BSE image – Cross sectional view



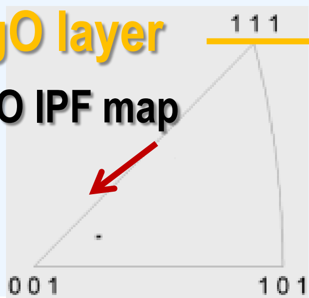
Lower CI values between Nb matrix and MgO substrate indicate that there could be an amorphous or non-structured layer between them

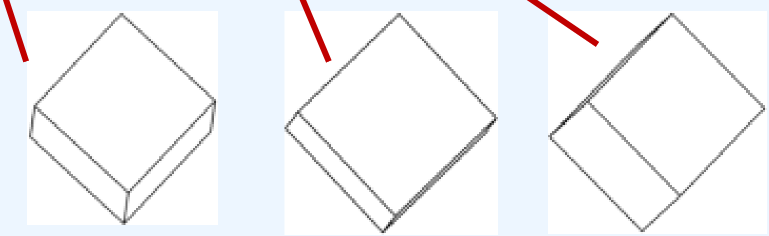


Nb thin film layer

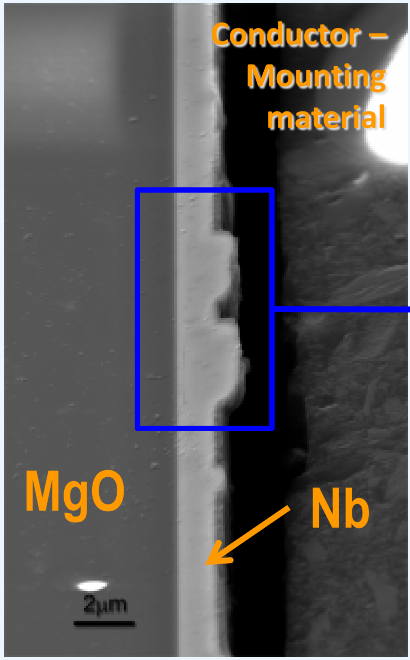
MgO layer

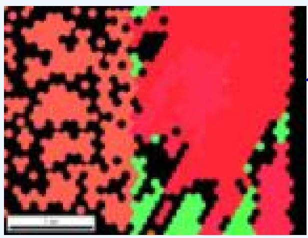
MgO IPF map

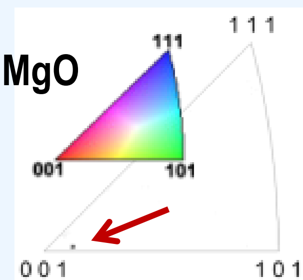




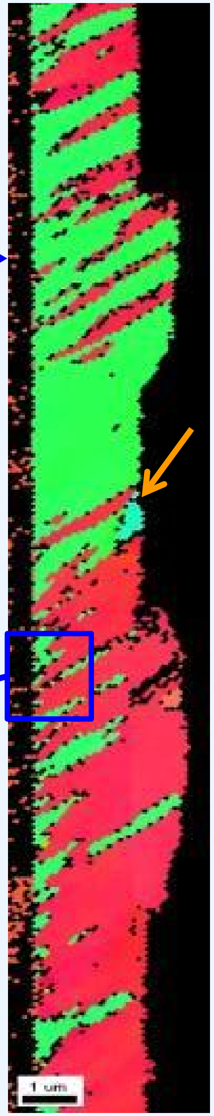
RRR-196: Cross sectional EBSD: shows mix of (110) and (100)



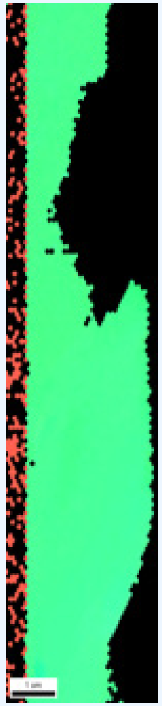




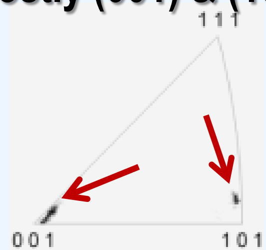
Area A



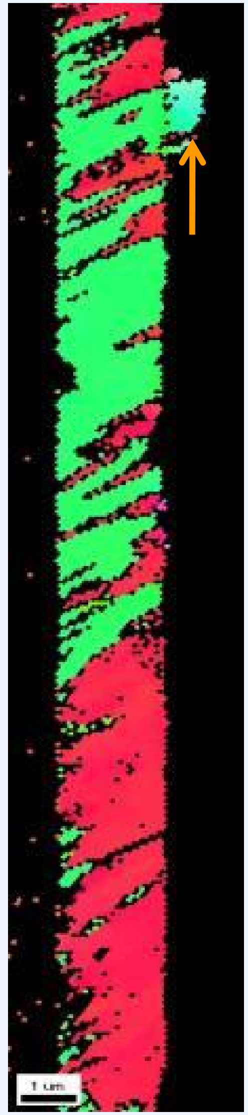
Area B



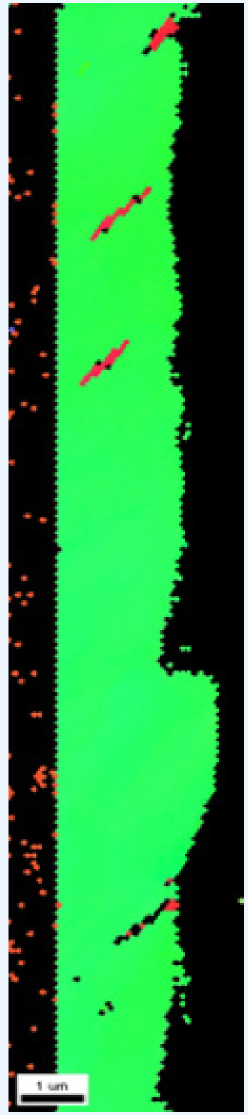
Nb structures are mostly (001) & (101)



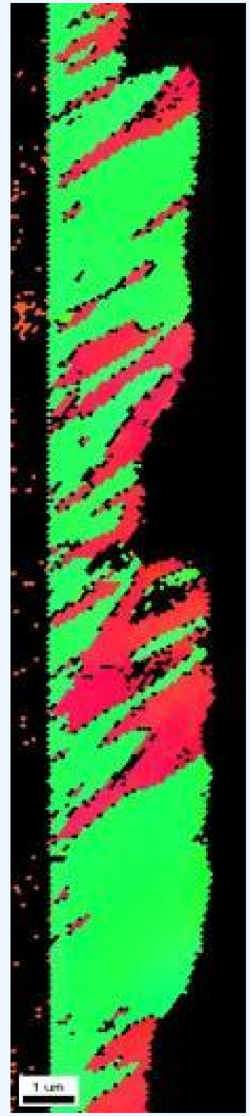
Area C



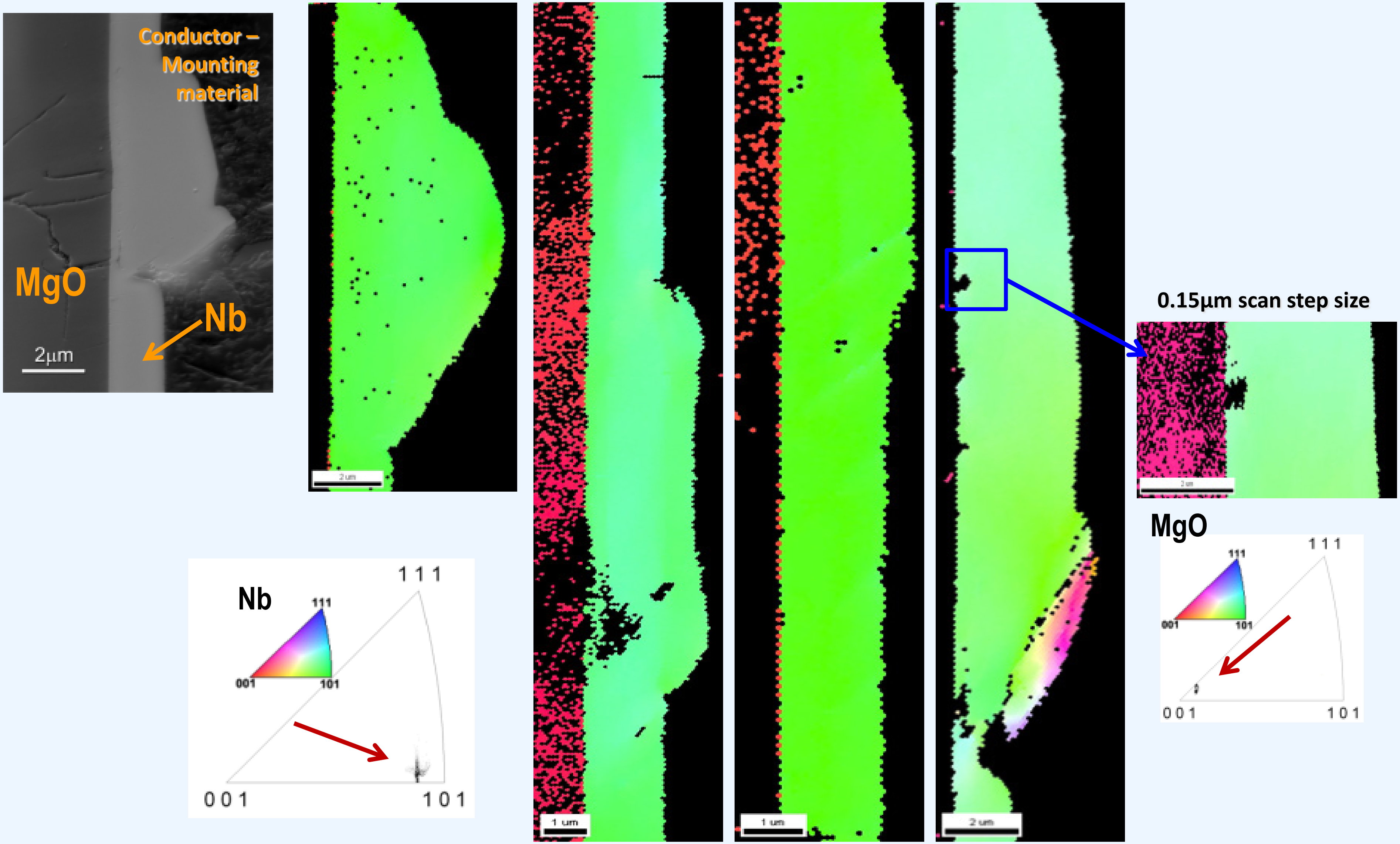
Area D



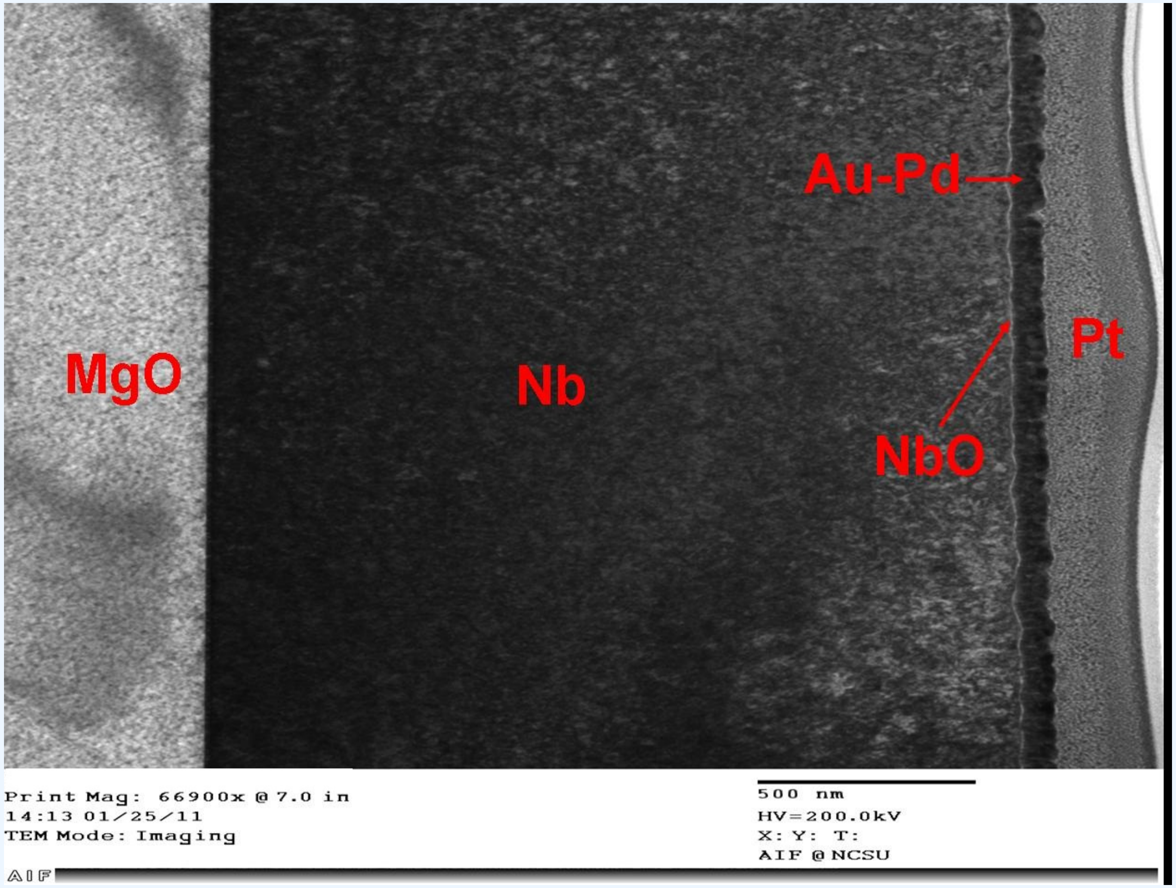
Area E

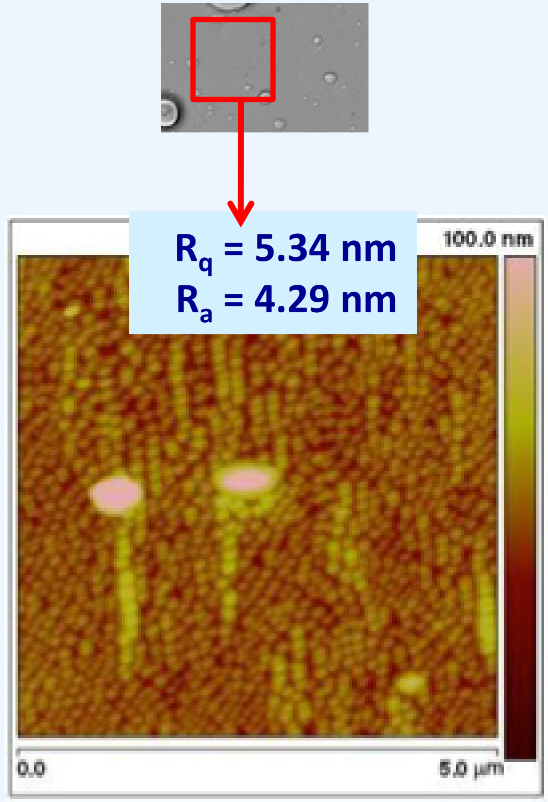


RRR-316: Cross sectional EBSD



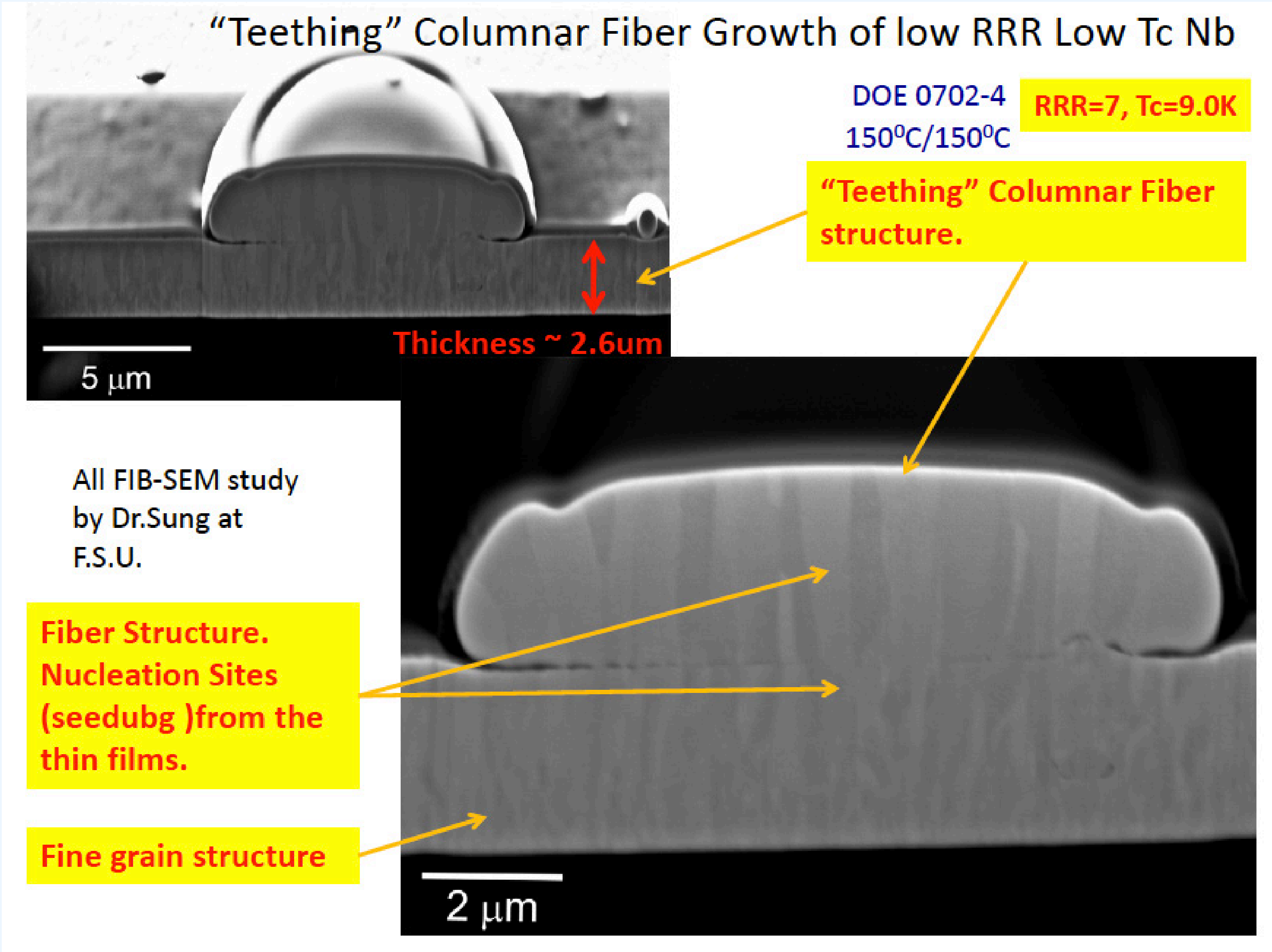
TEM/AFM of high RRR samples





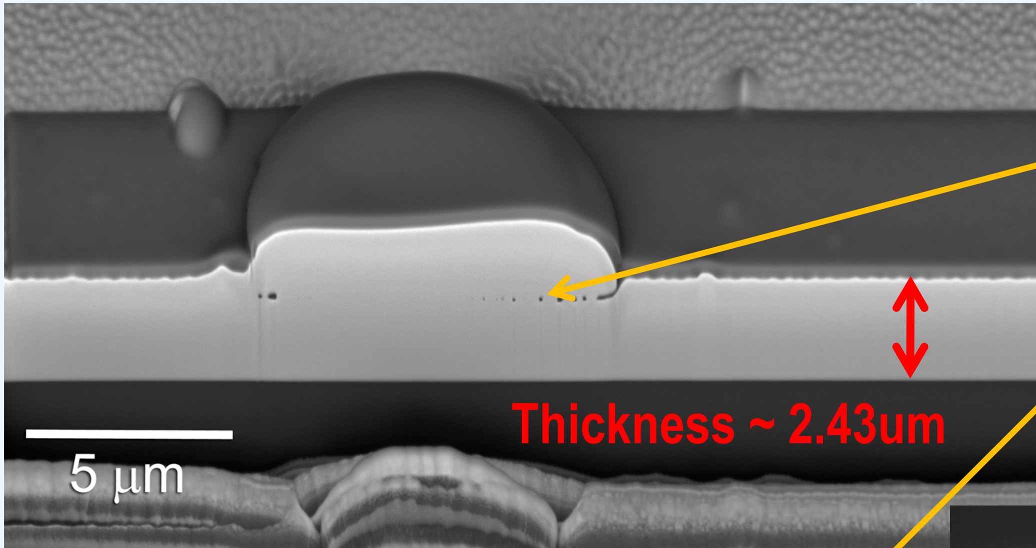
- ◆ Energetic Condensation (subplantation) physics drives an adhesive, non-porous, dense interface between substrate and Nb film
- ◆Nb surface is smooth (~5nm roughness)

“Mold” effect of subplanted Nb films: low RRR film



The macroparticle acquires the same crystallite structure as does the underlying Nb thin film that preceded it . This is for a low RRR film.

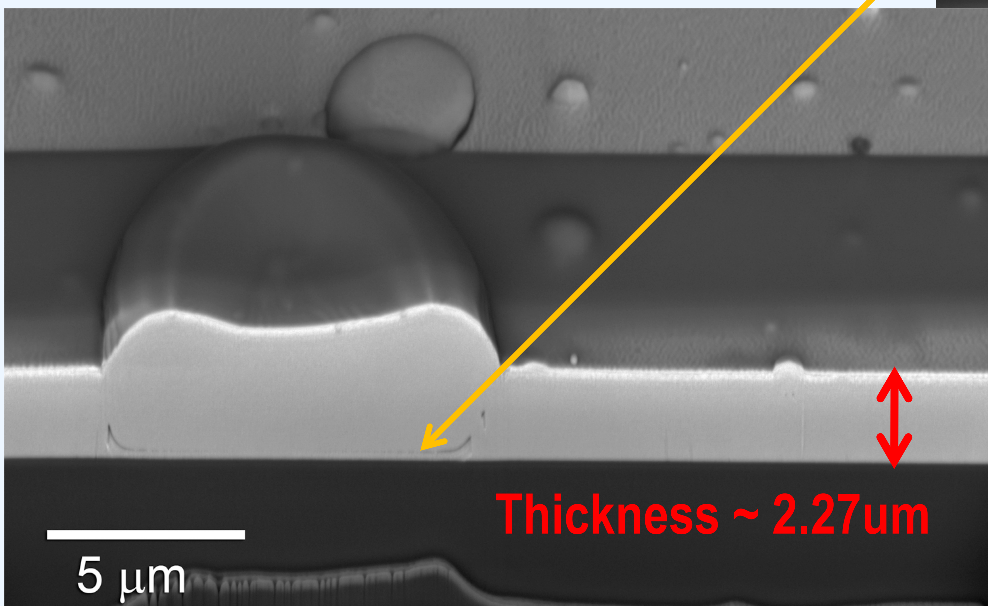
“Mold” effect of subplanted Nb films: high RRR film

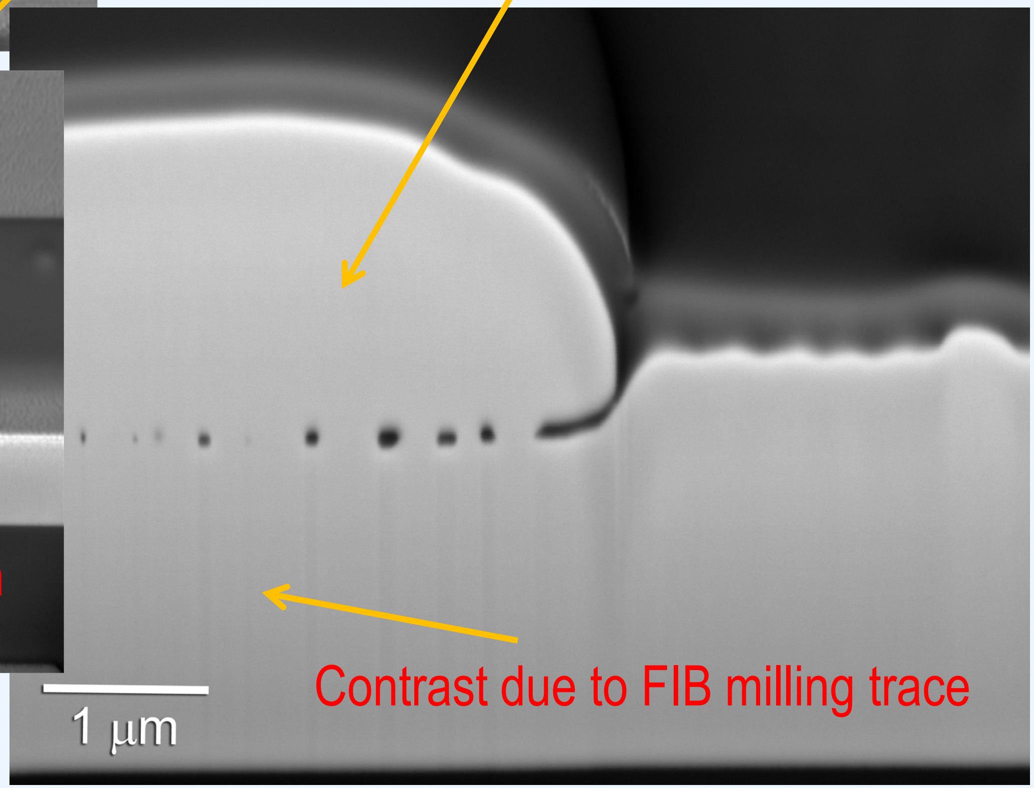


This macro landed on 2.4µm Nb film

This macro landed *near* MgO surface

No Columnar structure; XRD shows (100) single crystal





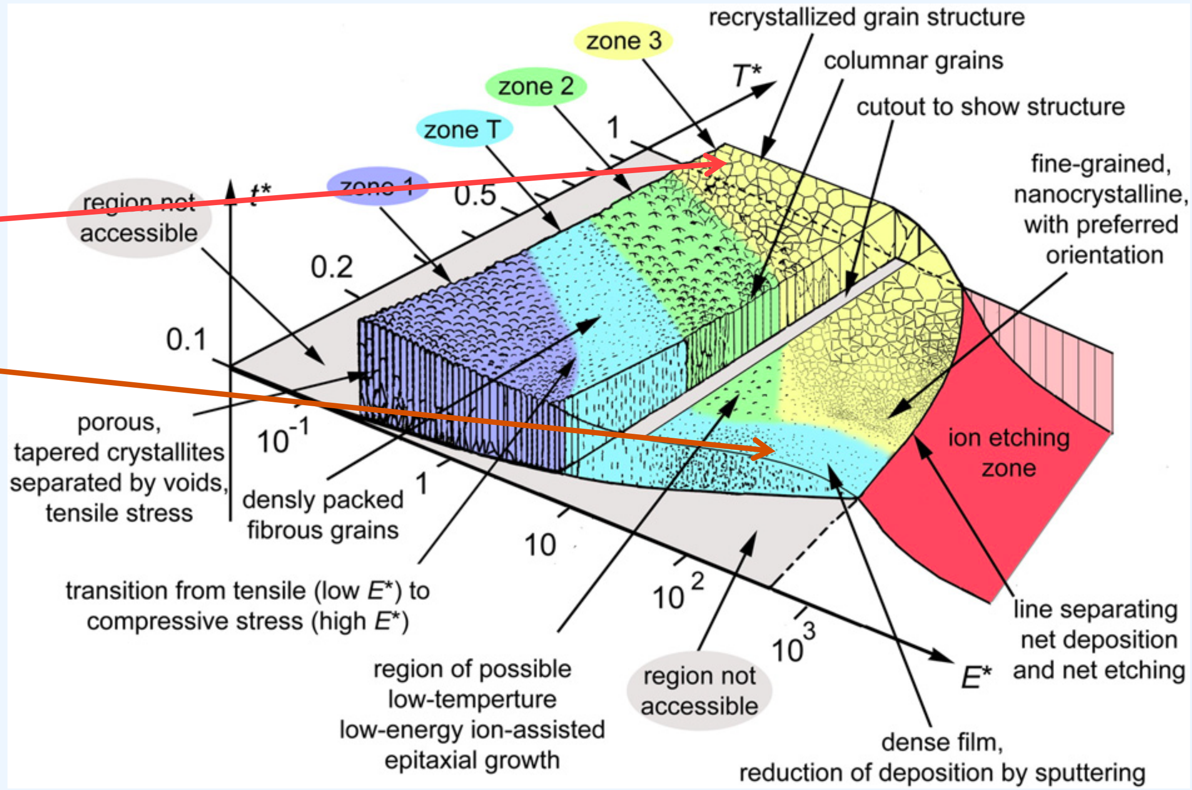
Contrast due to FIB milling trace

Revisit Andre Anders' SZD

- Macro-particles appear to have the same crystal structure as subplantation grown films
- Macroparticles arrive at surface with very low kinetic energy but $T/T_m \sim 1$

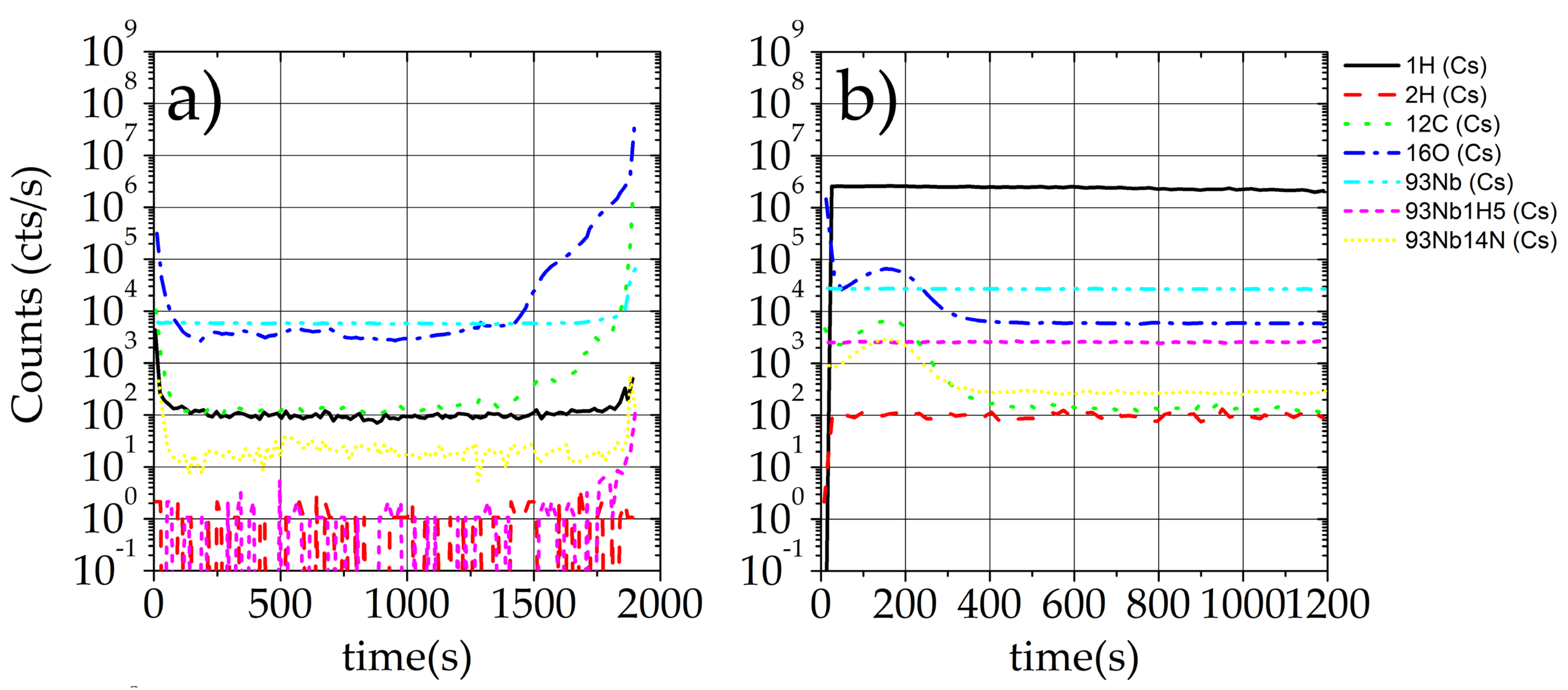
Macro-particles

Nb ions



Can we develop a "hybrid" growth concept in which Energetic Condensation plants the seed and then pure molten metal is poured on this seed, to develop a highly crystalline order via the "mold" effect?

SIMS with Cs beam of high RRR Nb films:



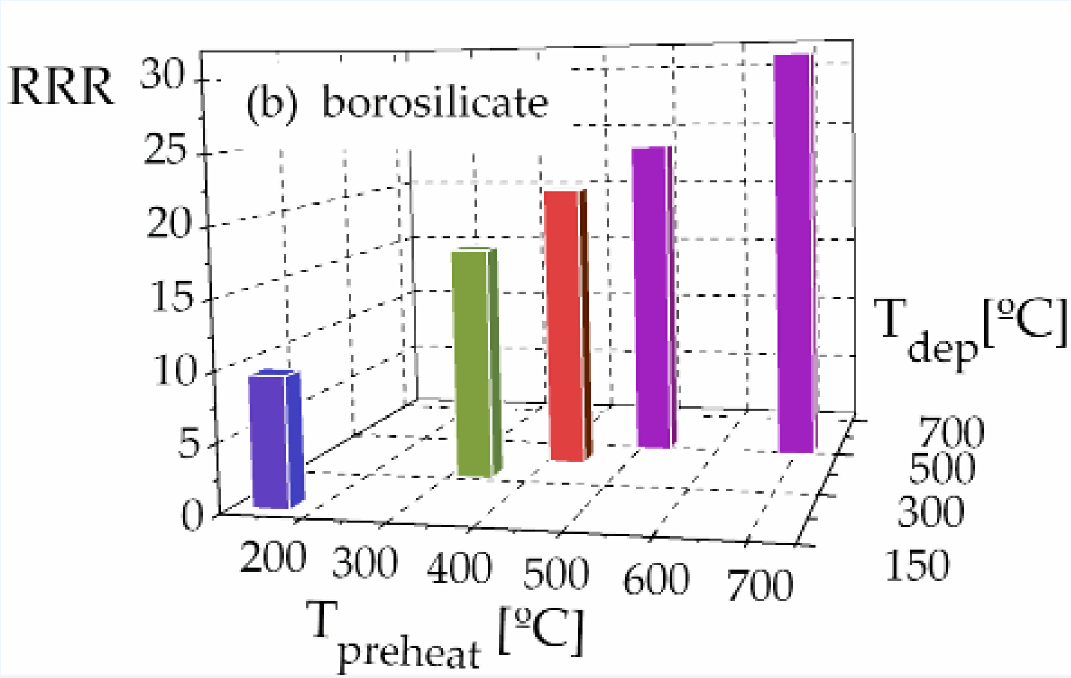
Nb (100) on MgO: RRR=442

Bulk Nb sample: RRR~300

H count rate (normalized) in thin films is $7x10^3$ times lower than in bulk Nb!

Nb thin films grown on Borosilicate: a proxy for Cu/Al

- Nb thin films grown on amorphous borosilicate show a similar trend of higher RRR with higher substrate temperature



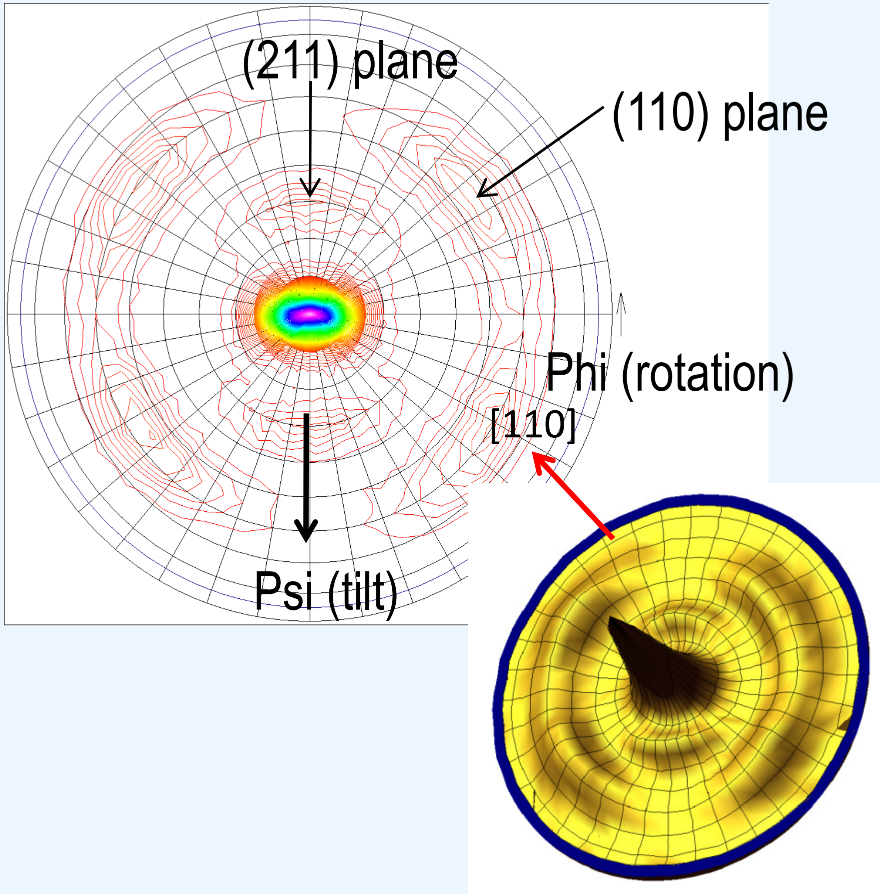
One option for Nb-on-Cu growth is to first "amorphize" the substrate by high energy "subplantation", then attempt to drive Nb crystal growth
Aluminum substrates would also use this approach. *Borosilicate is a proxy*

Please visit THPO069 on Thursday: Nb Film Growth on Crystalline and Amorphous Substrates" Dr. Enrique Valderrama

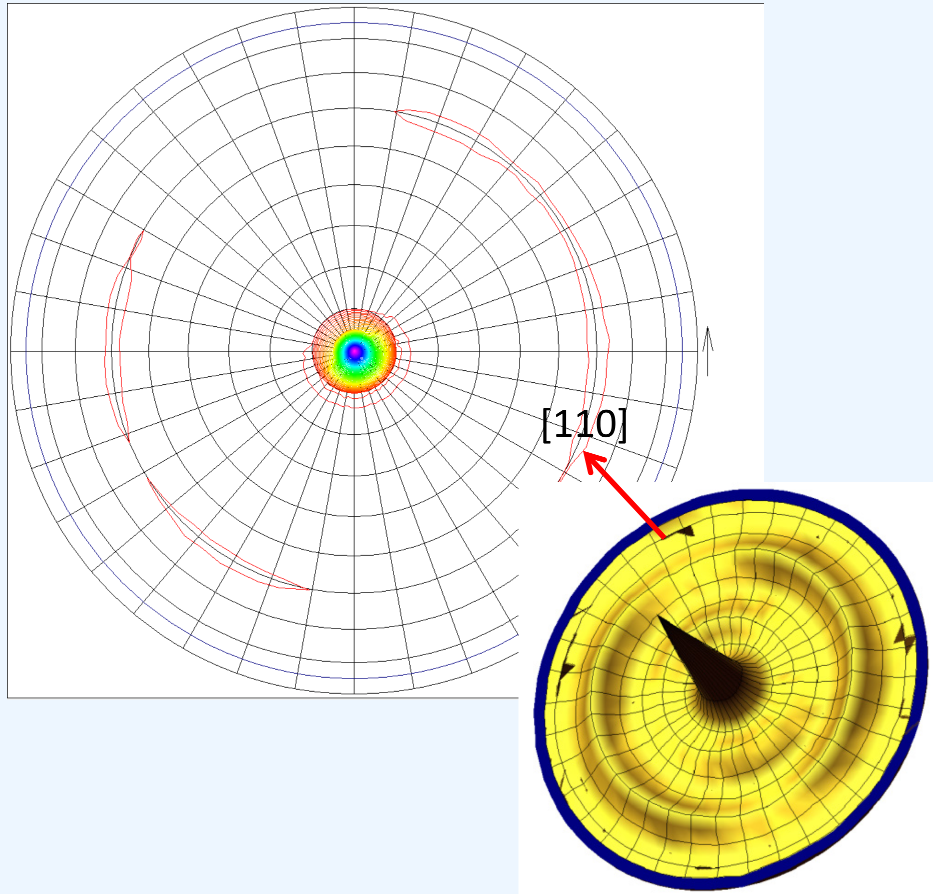
XRD Pole Figures for Nb thin films grown on Borosilicate

Subplantation physics of energetic condensation at work here

(110) Nb on Borosilicate at 150/150C



(110) Nb on Borosilicate at 400/400C



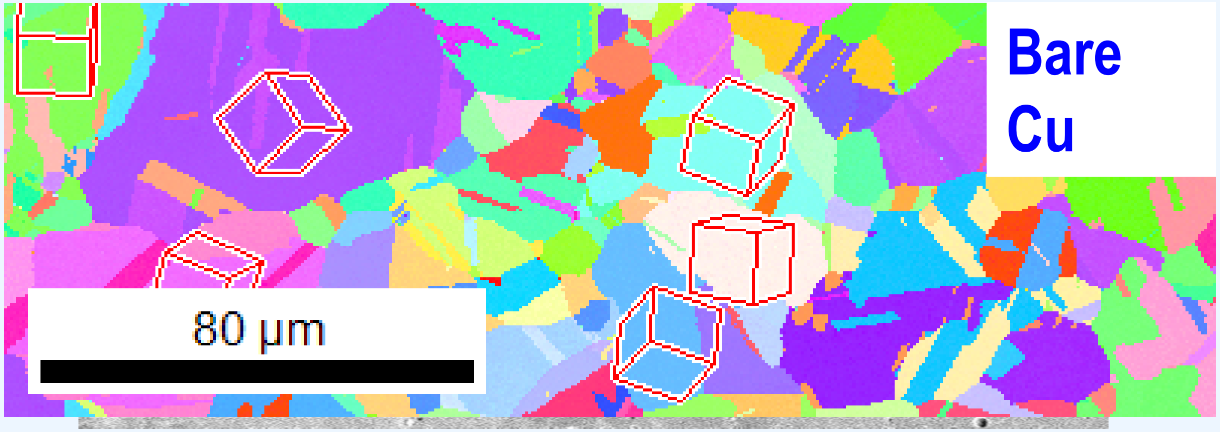
A strong [110] Nb fiber structure perpendicular to the substrate

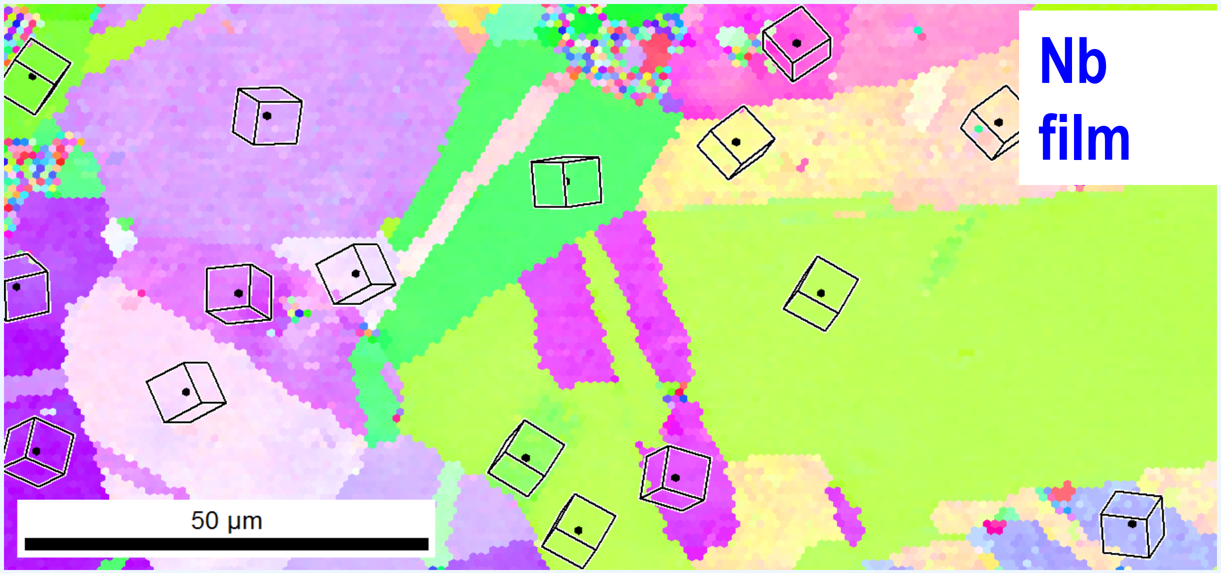
At higher coating temperature, in-plane texture shows that [110] fiber texture is highly oriented to the substrate

~50μm grains of Nb measured in 0.4μm thick films on Cu

"Large crystal grain niobium thin films deposited by energetic condensation in vacuum arc", X. Zhao, A.-M. Valente-Feliciano, C. Xu, R. L. P. Geng, L., C. E. Reece, K. Seo, R. Crooks, M. Krishnan, A. Gerhan, B. Bures, K. W. Elliott, and J. Wright, Journal of Vacuum Science & Technology A 27, 620-625 (2009).

CED™ Nb Film on polycrystalline Cu substrate: Hetero-epitaxy





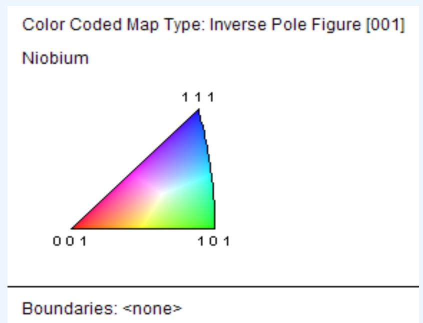
Working Distance: 15.000000

Number of points: 13094
Number of good points: 13093

150.00 microns x 74.48 microns
Step: 1.00 microns

Average Confidence Index: 0.42
Average Image Quality: 2633.56
Average Fit [degrees]: 1.35

EBSD Image of Sample TF-AASC-CED-Nb-Cu-103,CED™, Cu substrate T=400C



Energetic Condensation demonstrated growth of very large Nb grains on Cu.

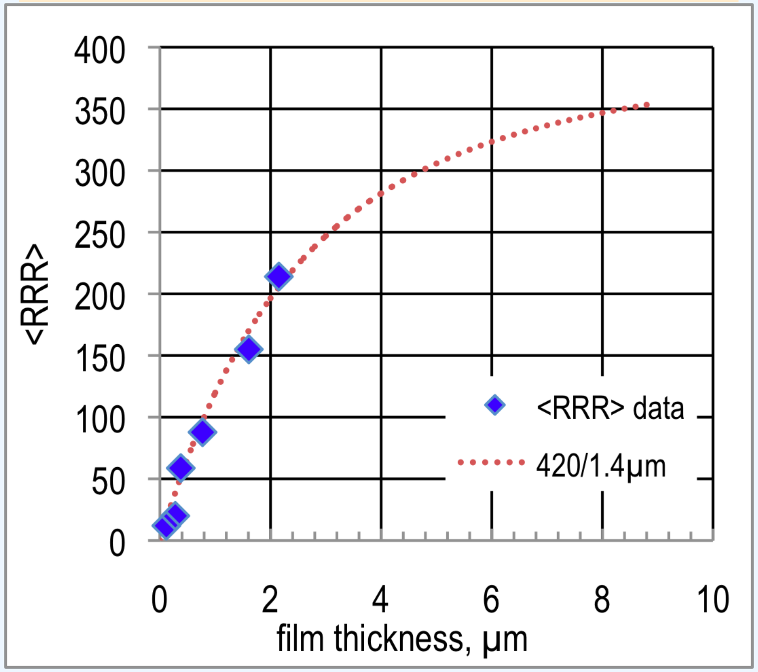
Please see Anne-Marie's talk later for recent RRR and other data from an ECR source

Vielen Dank

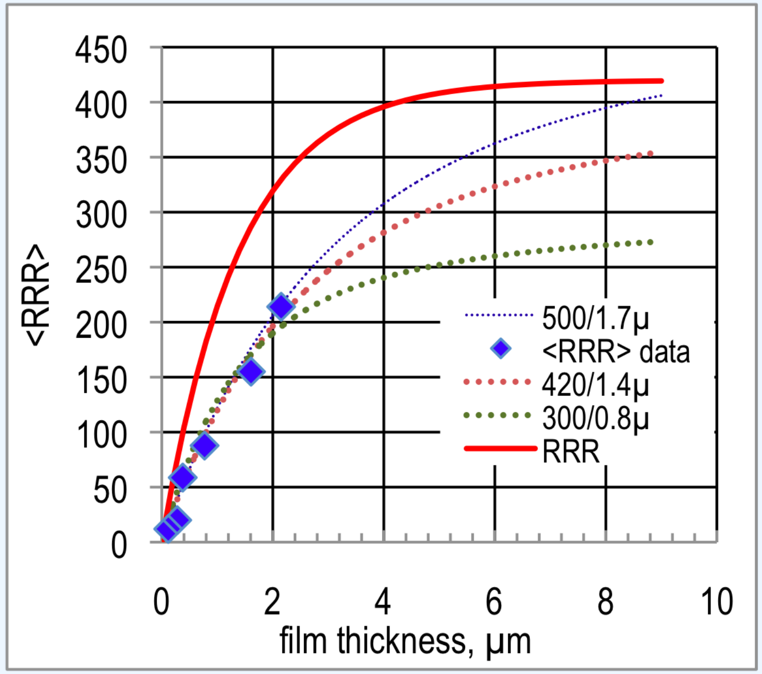
Fragen, bitte?

Nb on a-sapphire: Local RRR vs. averaged <RRR>

- <RRR> vs. film thickness



- Ad hoc model fit to <RRR> data



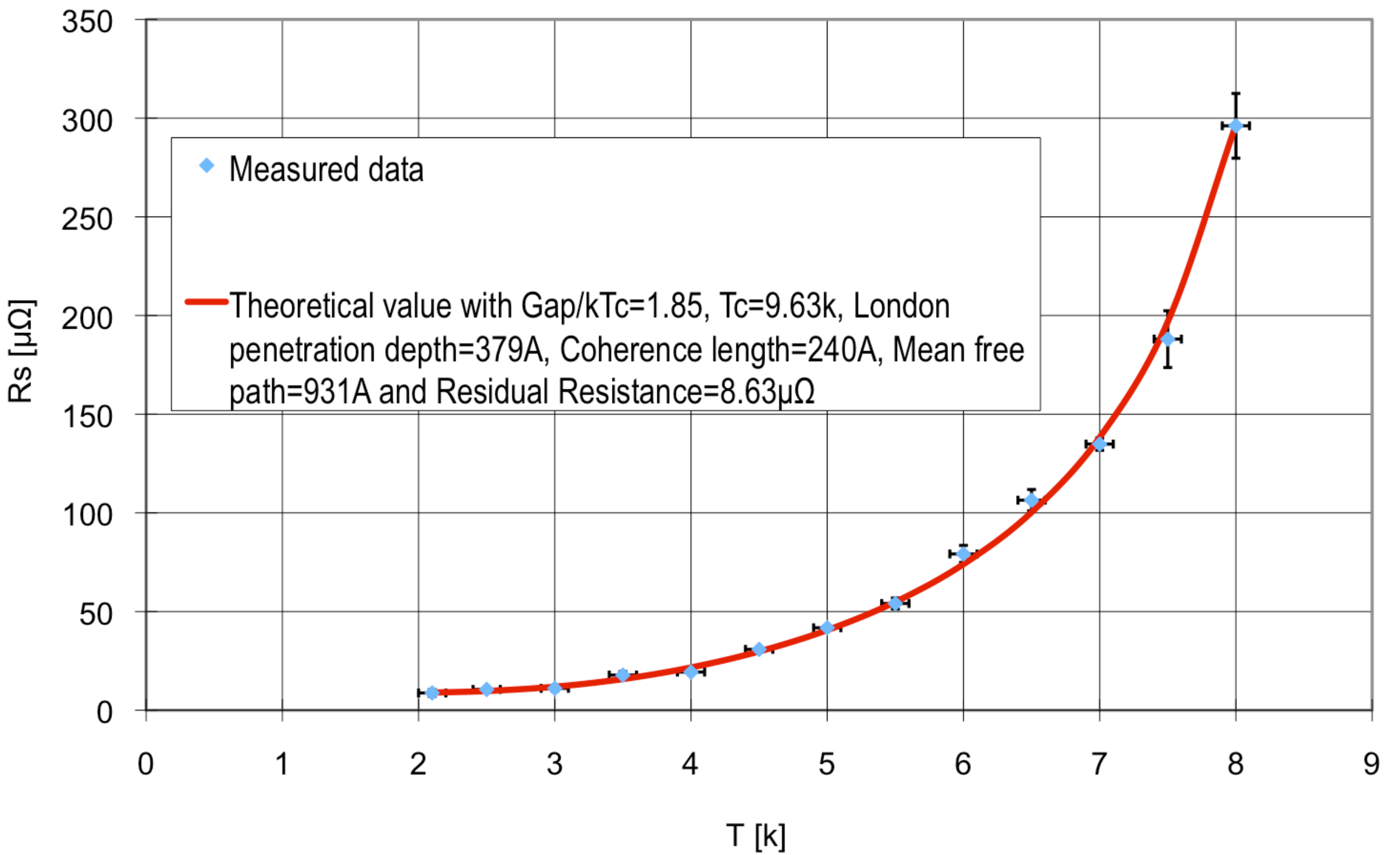
$$< RRR >= \frac{1}{x_o}\int_0^{x_o} R_o * \left(1 - e^{-\frac{x}{x^*}}\right)dx \;= R_o\left[1 - \frac{x^*}{x}\left\{1 - e^{-\frac{x}{x^*}}\right\}\right]$$

$$RRR = R_o * \left[1 - e^{-x/x^*}\right]$$

Thicker films must be analyzed before we can do a better model fit

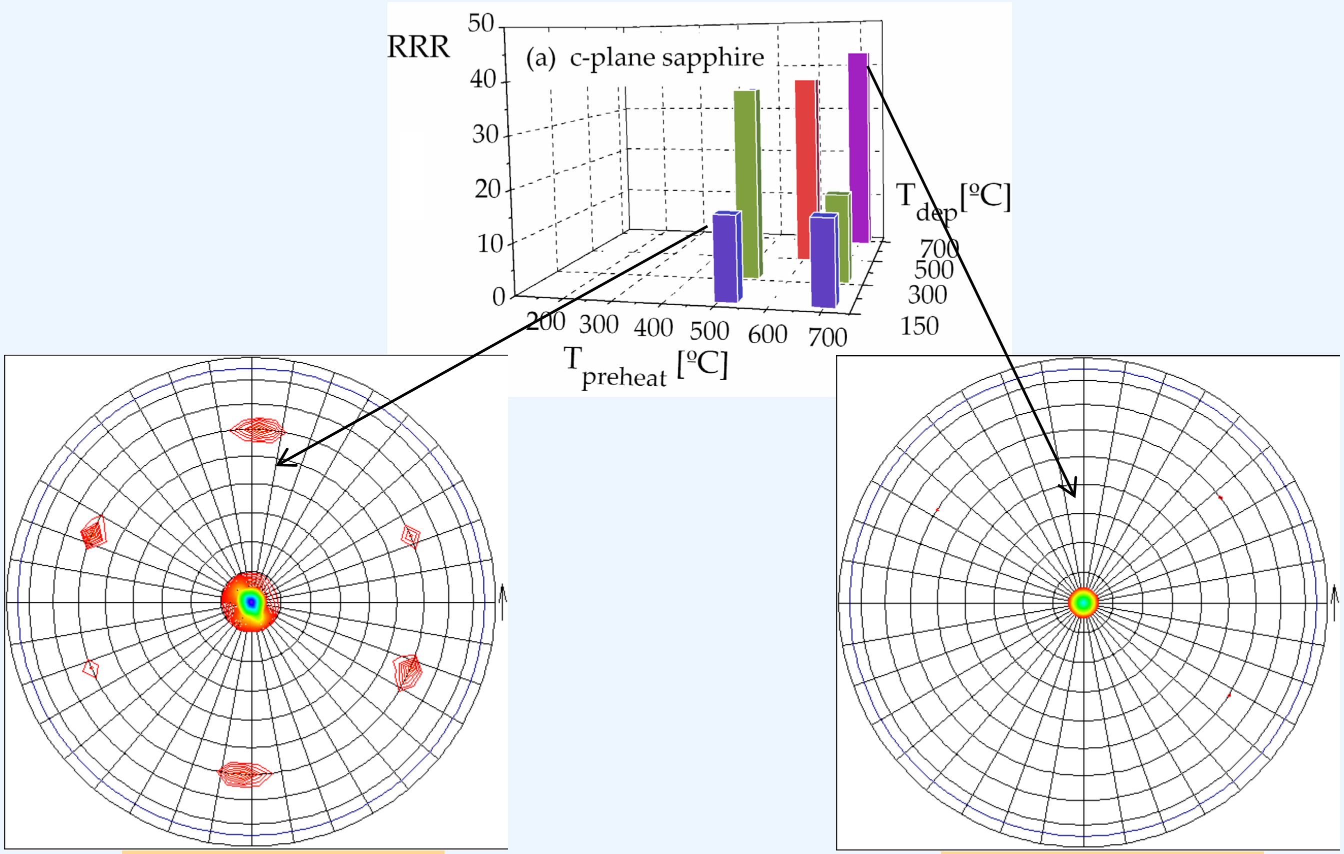
SIC measurements at JLab

R_s vs. T: thin film Nb on Cu sample TF-AASC-CED-103; EP, annealed at 750 C, then EP again



RRR of Nb thin films on c-sapphire substrates

- Pole Figures show better crystal structure at higher temperature

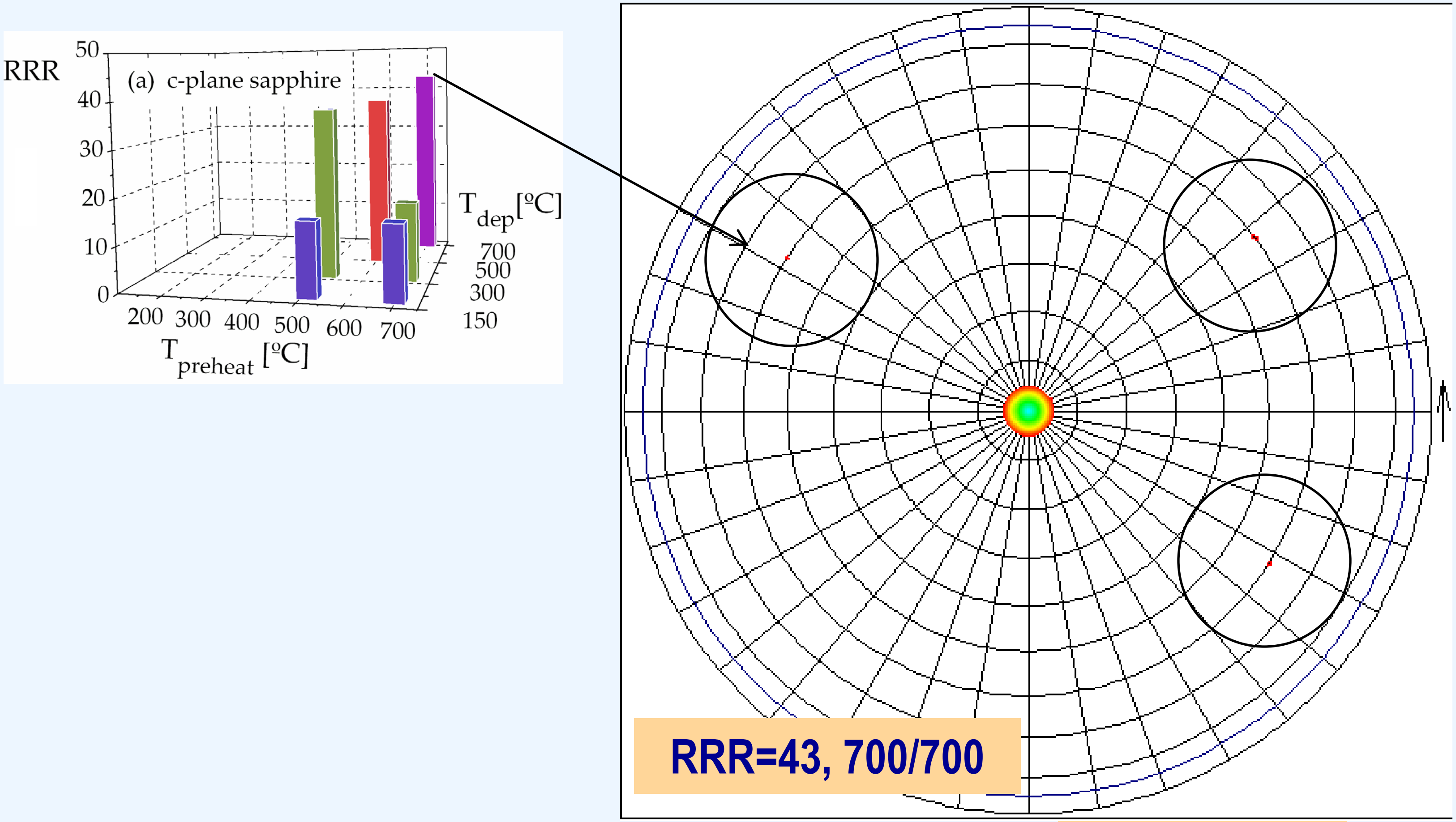


RRR=16, 700/150

RRR=43, 700/700

RRR of Nb thin films on c-sapphire substrates

- Pole Figures show better crystal structure at higher temperature



- Despite high crystalline order, RRR is only 43. High order is a necessary condition for high RRR, but film might have impurities that limit electron mean free path.

RRR-196: Cross sectional EBSD: shows mix of (110) and (100)

sample ID	RRR	error bar	T_c	# of shots	delta, µm	anneal temp	dep temp	substrate	base pressure
CED-113010-66	541	10%	9.30	20000	3.67	700	500	MgO(100), MTI, 2 x1 cm^2	2.60E-08
CED-120310-68	554	25%	9.30	30000	5.50	700	500	MgO(100), MTI, 1 x1 cm^2	2.70E-08
CED-120310-69	585	1%	9.34	30000	5.5	700	500	MgO(100), MTI, 1 x1 cm^2	2.70E-08