Energetic Condensation Growth of Nb and other Thin Films for SRF Accelerators

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presented at
SRF-13
September 25, 2013

Collaborators:

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Los Alamos National Laboratory	T. Tajima, L. Civale
Argonne National Lab	T. Proslier
Helmholtz-Zentrum Berlin für Materialien und Energie GmbH	O. Kugeler, S. Aull*, Raphael Kleindienst * At CERN

- **This research is supported at AASC by DOE SBIR Grants DE-SC0004994 and DE-FG02-08ER85162**

Outline

- Motivation for thin films in SRF cavities
- History of thin film development
- Energetic Condensation
- Coupon study of thin films produced by energetic condensation gave insights to motivate cavity coating
- Thin films on Cu and Nb cavities
- Other SRF related films grown using EC
- Future Plans

Motivation for AASC's SRF program

- The accelerator technology R&D research area of DOE (NP & HEP) develops the next generation of particle accelerators and related technologies for discovery science; and also for possible applications in industry, medicine and other fields
- SRF accelerators are a key part of this future development, as they can significantly reduce costs
- SRF technology also has many commercial applications:
  - More than 1000 particle accelerators worldwide; most use normal cavities
  - Superconducting RF (SRF) cavities offer a ~10x improvement in energy efficiency over normal cavities, even accounting for cryogenic costs at 2K
  - Operating at higher temperatures (~10K) would further improve accelerator energy efficiency as the cryogenic cooling becomes less demanding and moves away from liquid He and towards off the shelf cryo-coolers such as those used in cryo-pumps
  - Replacing bulk Nb with Nb coated Cu cavities would also reduce costs
  - The ultimate payoff would be from cast Al SRF cavities coated with higher temperature superconductors (Nb_3Sn, MoRe, MgB_2, Oxipnictides)

Our thin film superconductor development is aimed at these broad goals

Motivation for AASC's SRF program

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- SRF accelerators are a [illegible] future de[illegible], as they can [illegible] reduce costs
- SRF tec[illegible] has man[illegible] applications
  - More [illegible] 1000 n[illegible]ators worldwid[illegible]ormal cavities
  - Supercon[illegible]F) cavities [illegible]provement in energy efficiency over normal ca[illegible]ccounting for [illegible] at 2K
  - [illegible]ng at higher t[illegible] 10K) would further improve accelerator energy efficiency as the cryogenic [illegible]es less demanding and moves away from liquid He and towards off the sh[illegible] such as those used in cryo-pumps
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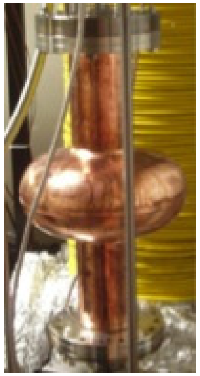
SRF uses less power than normal

SRF with cheaper cavity materials (Cu) would be better

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

Challenges for thin film SRF: path to success

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



Amorphous

- How do we grow low-defect Nb films on such substrates?
- Study adhesion, thickness, smoothness, RRR, stability
- Understand these issues at the coupon level
- Validate these films at low field levels on realistic geometries
- Proceed to RF cavity level and measure Q at high fields

Where we are

- Install 9-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

Where we are headed

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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. Arnolds, 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

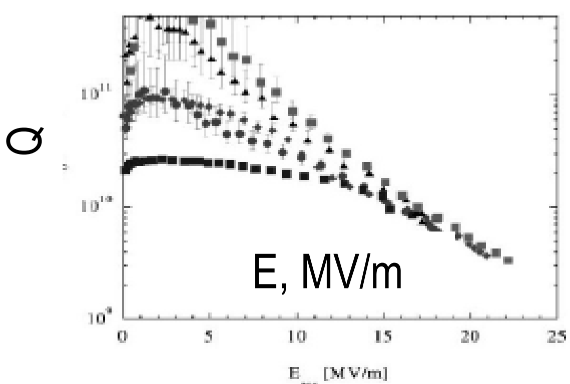
LEP2 at CERN tested Cu cavities Magnetron sputter coated with ~1.5μm Nb thin films

Thin Film Cavities & Q-Drop

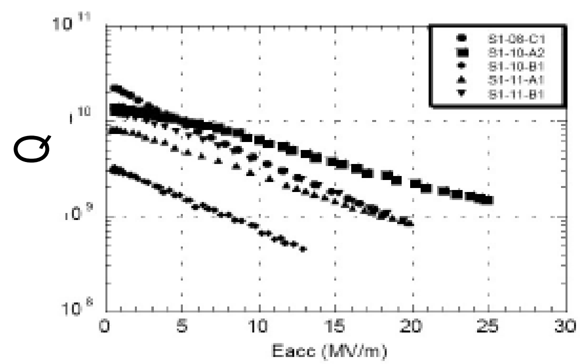
dapnia
cea
saclay

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

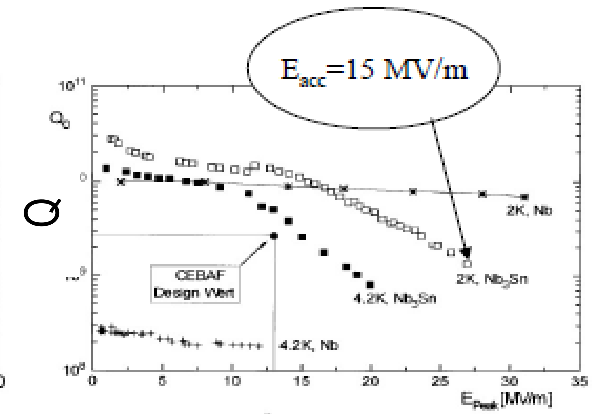
severe Q-drop limits High Gradient Performances E_{acc} < 25 MV/m



CERN – Nb / Cu – 1.5 GHz
Magnetron Sputtering in Kr
V. Arbet-Engels et al.- NIMA (2001)



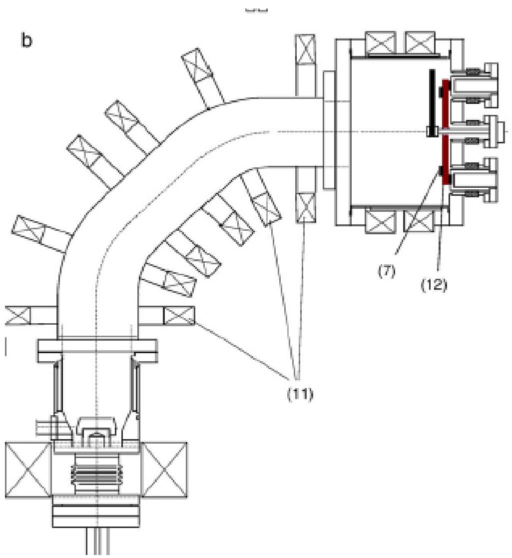
Saclay – Nb / Cu – 1.5 GHz
Magnetron Sputtering in Ar
P. Bosland et al.- ASC (1998)

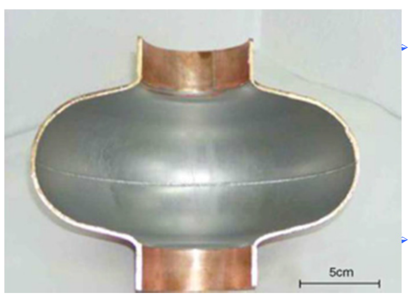


Wuppertal – Nb_3Sn / Nb – 1.5 GHz
Vapor Deposition Technique
G. Müller et al.- EPAC (1996)

(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 1–2×10^{-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°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

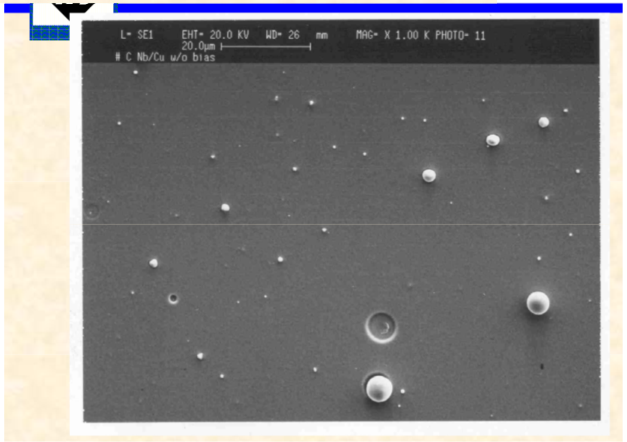
The INFN/Poland CARE program made good progress

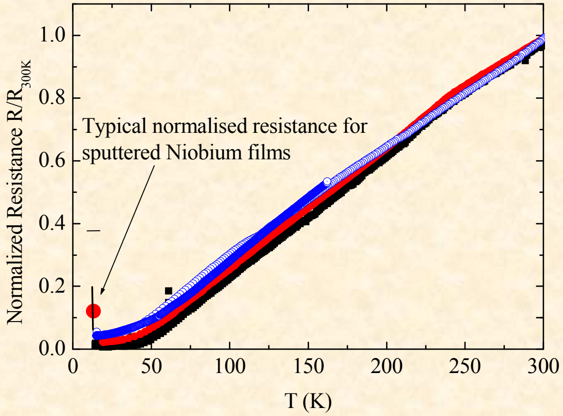
financed and supported by INFN and FP6 (European Program) "CARE"

High quality Niobium films produced by Ultra High Vacuum Cathodic Arc

Roberto Russo INFN-Na

- L. Catani, A. Cianchi, J. Lorkiewicz, S. Tazzari, - INFN Roma 2 –
- Dr. J.Langner , Prof. S. Kulinski and their group - SINS –





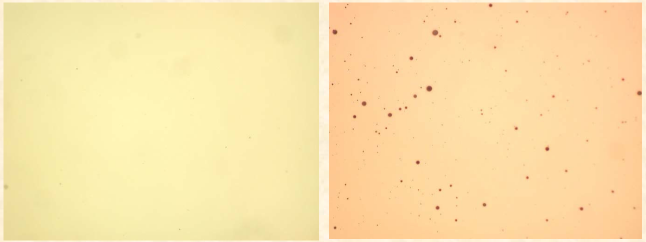
RRR values from 20 to 100 were obtained

Deposition temperature below 100°C

Thickness 100-2000nm

Roberto Russo - INFN

12 JLab 17-18/7/2005



Less than 100 particles / mm²

About 6000 particles / mm²

RRR values up to 100 (within the range of bulk Nb) were measured;

Nb macro-droplets were studied; a magnetic filter was developed to reduce macros

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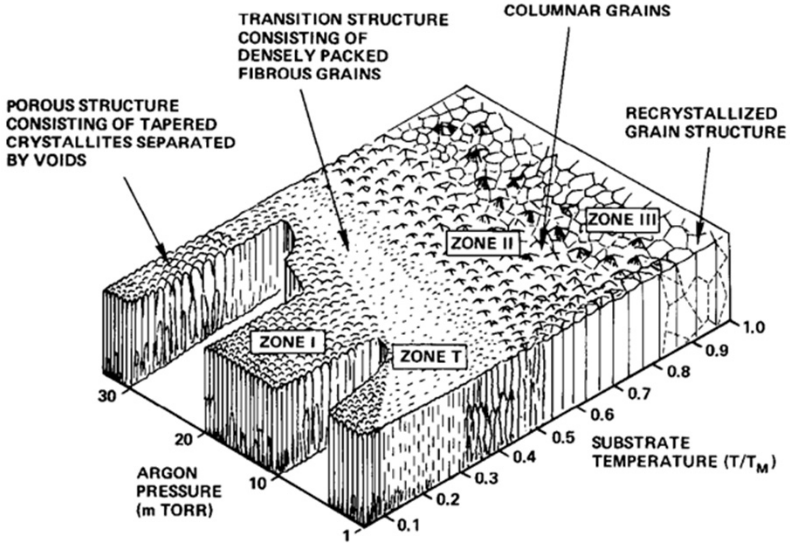
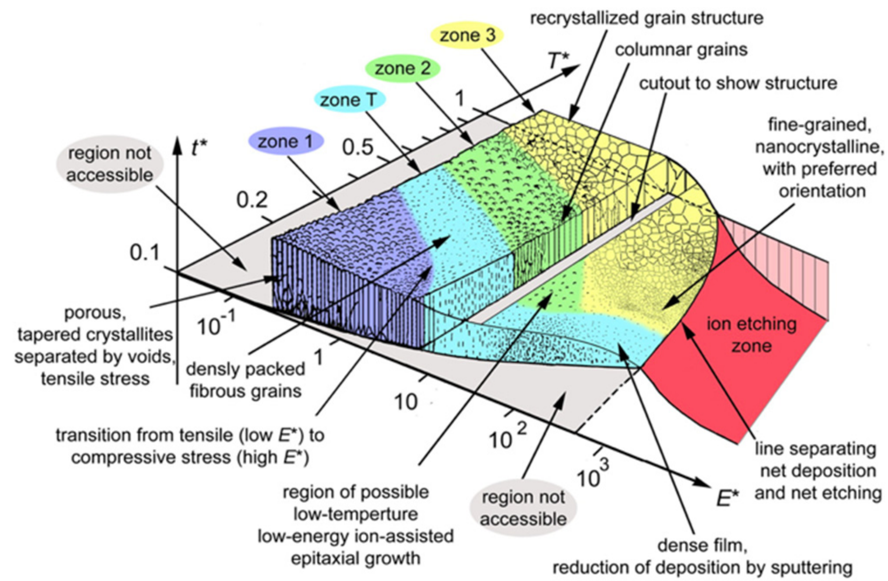
Energetic Condensation



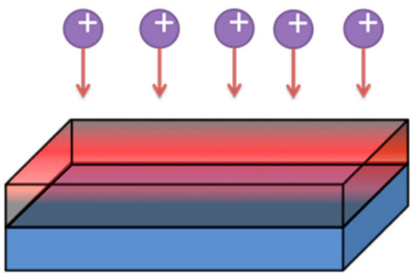


J.A. Thornton, "Influence of substrate temperature and deposition rate on the structure of thick sputtered Cu coatings", J. Vac. Sci. Technol. Vol. 12, 4 Jul/Aug 1975

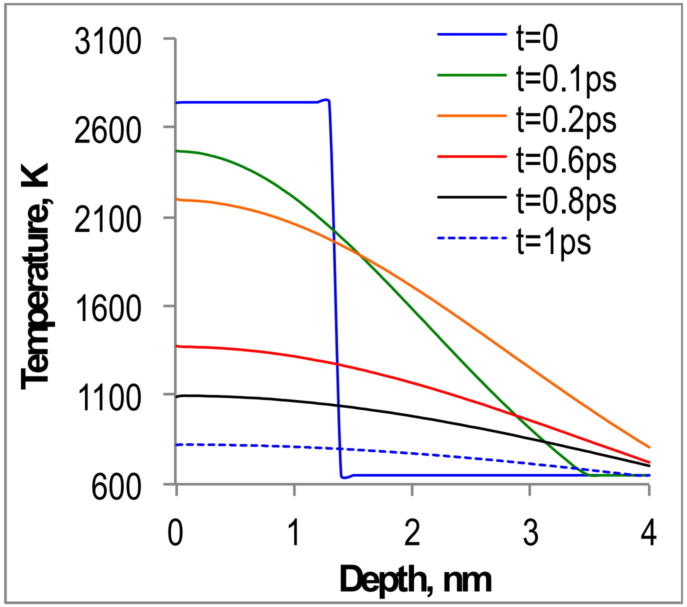
Andre Anders, A structure zone diagram including plasma-based deposition and ion etching, Thin Solid Films 518 (2010) 4087–4090

- In Energetic Condensation, the ions deposit energy in a sub-surface layer (≈3-5 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

Energetic Condensation: some book-keeping



$$\nabla^2 T = \frac{c\rho}{\kappa}\frac{\partial T}{\partial t}$$

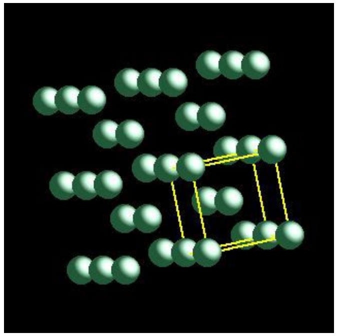


N.A. Marks, Phys. Rev. B 56(5) 1997 (thermal spikes paper)

$$E_{ion} = 0.26n \cdot \frac{2}{3}\pi r_0^{\ 3}$$

100eV ion into 8.45×10^{22} /cc solid

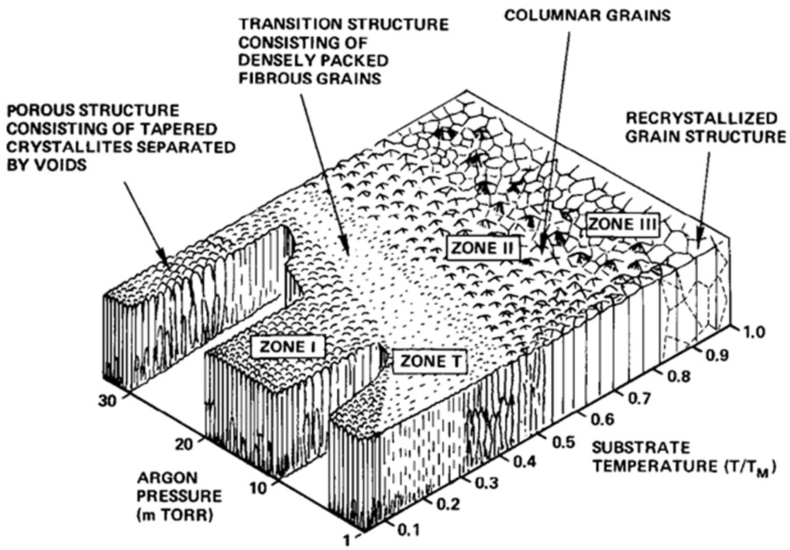
r_o = 1.5nm

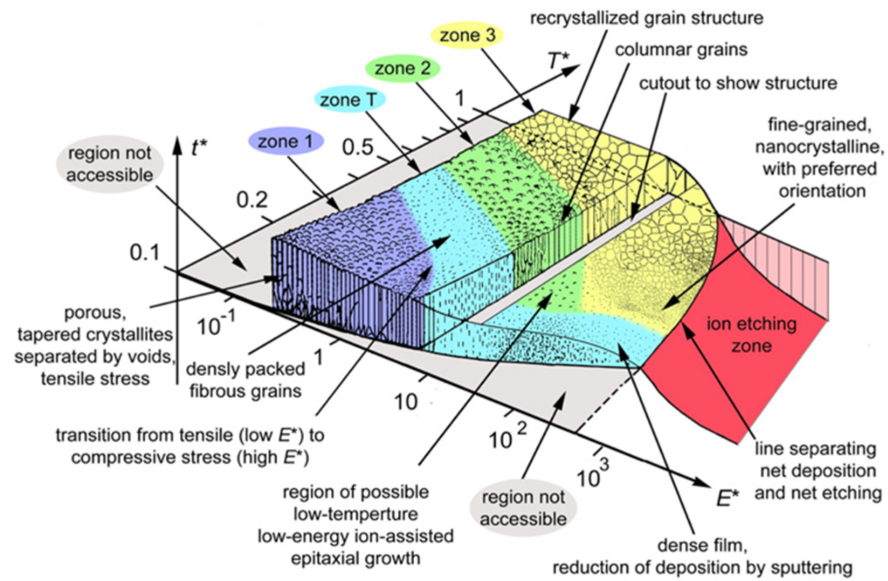


thermal spike energy=	0.24	eV
thermal speed=	705	m/s
Nb lattice spacing=	0.33	nm
time to move one spacing=	0.47	ps

- Observe that spike "quenching time" of ~0.8ps is comparable to the time for atoms to move and rearrange themselves; energetic condensation provides mobility for lattice rearrangement

Energetic Condensation





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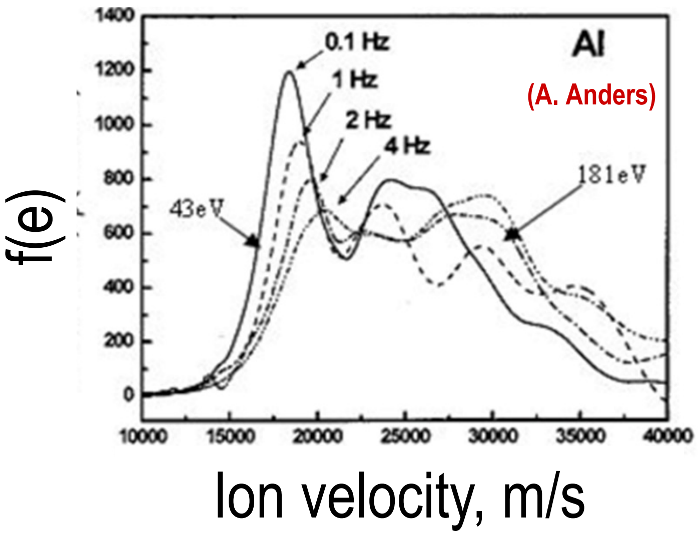
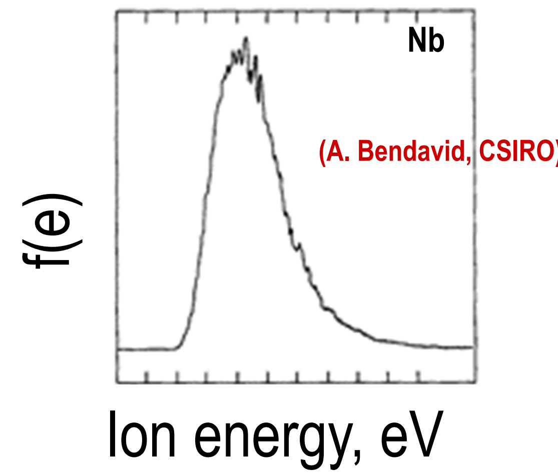
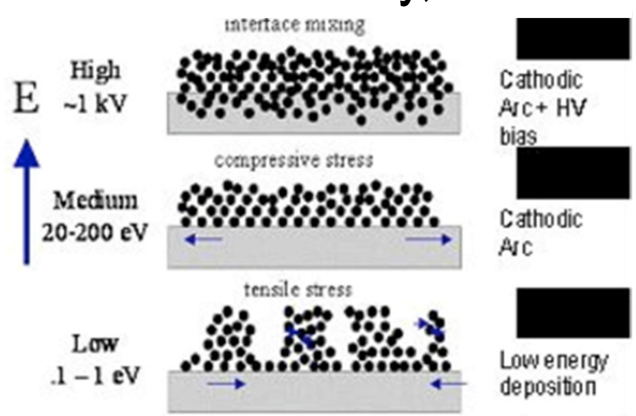
Energetic Condensation vs. sputtering and PVD

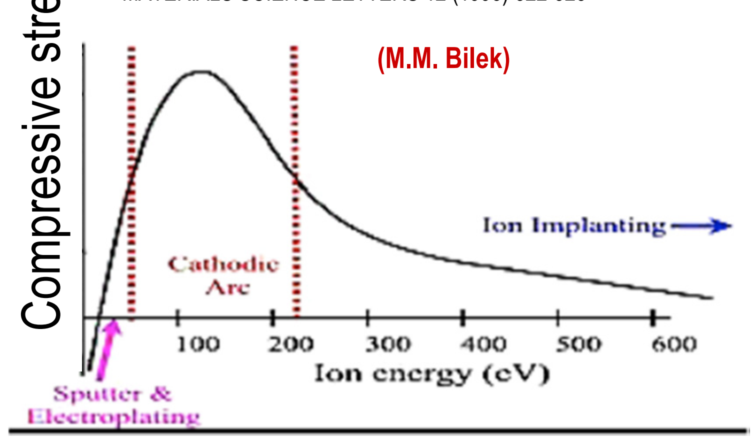




* A. BENDAVID, P. J. MARTIN, R. P. NETTERFIELD, G. J. SLOGGETT, T. J. KINDER, C., ANDRIKIDIS, JOURNAL OF MATERIALS SCIENCE LETTERS 12 (1993) 322-323



Comparison of Stress build-up in low energy deposition, energetic condensation, and energetic condensation plus high voltage bias

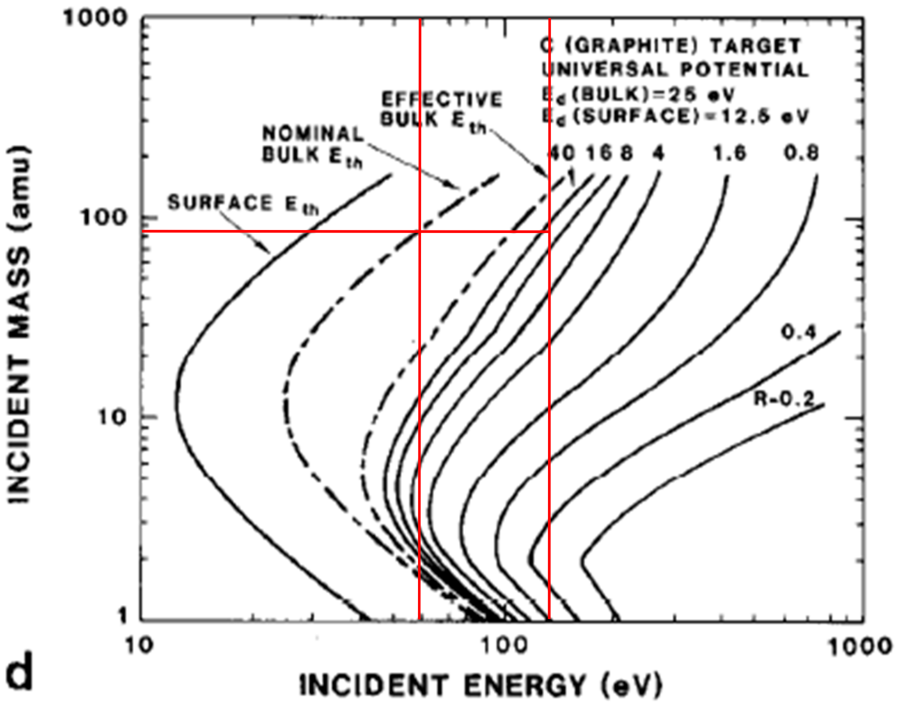


Relief of compressive stress by pulsed ion impact [*]

[*] M. M. Bilek, R N. Tarrant, D. R. McKenzie, S H. N. Lim, and D G. McCulloch "Control of Stress and Microstructure in Cathodic Arc Deposited Films" *IEEE TRANSACTIONS ON PLASMA SCIENCE*, VOL. 31, NO. 5, OCTOBER 2003

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; see also W. D. Wilson, Radiat. Eff. 78, 11 (1983)



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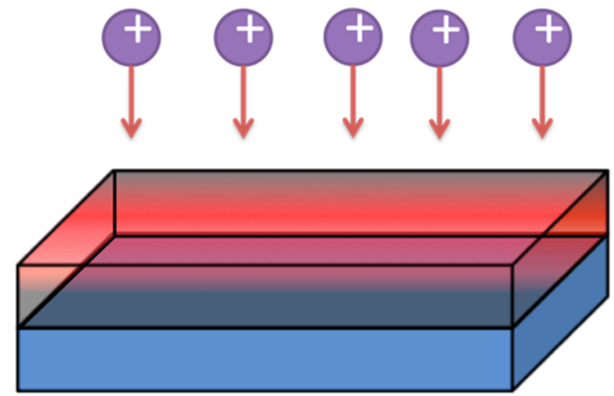
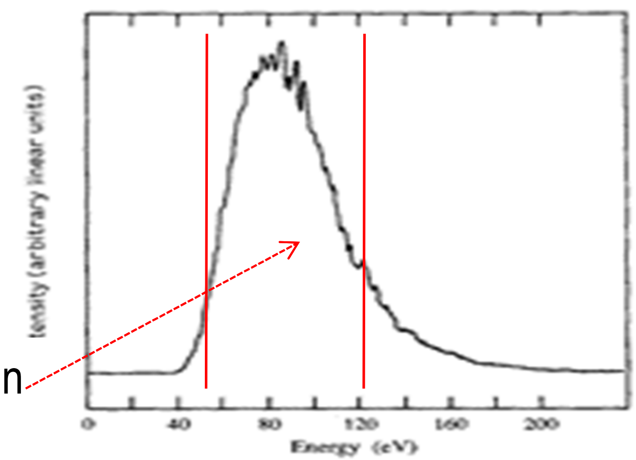


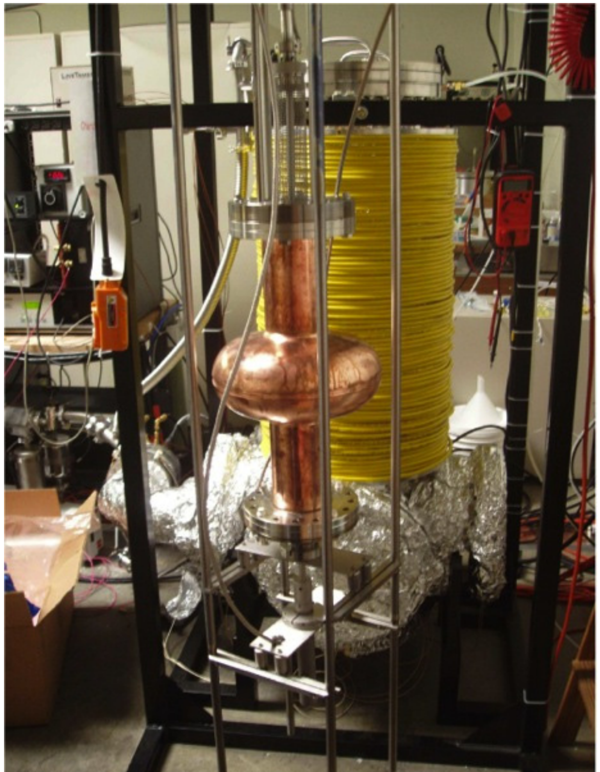


50-120eV energy spread in Nb ions from cathodic arc

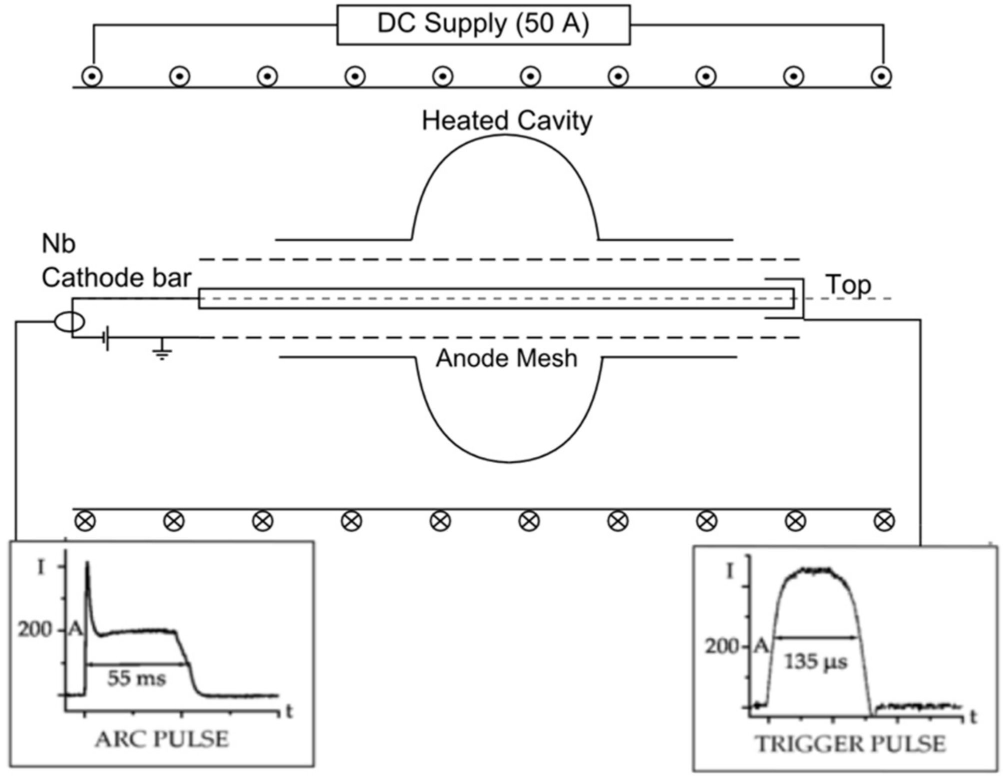
* A. BENDAVID, P. J. MARTIN, R. P. NETTERFIELD, G. J. SLOGGETT, T. J. KINDER, C., ANDRIKIDIS, JOURNAL OF MATERIALS SCIENCE LETTERS 12 (1993) 322-323

Coating Facilities at AASC

Coaxial Energetic Deposition (CED)

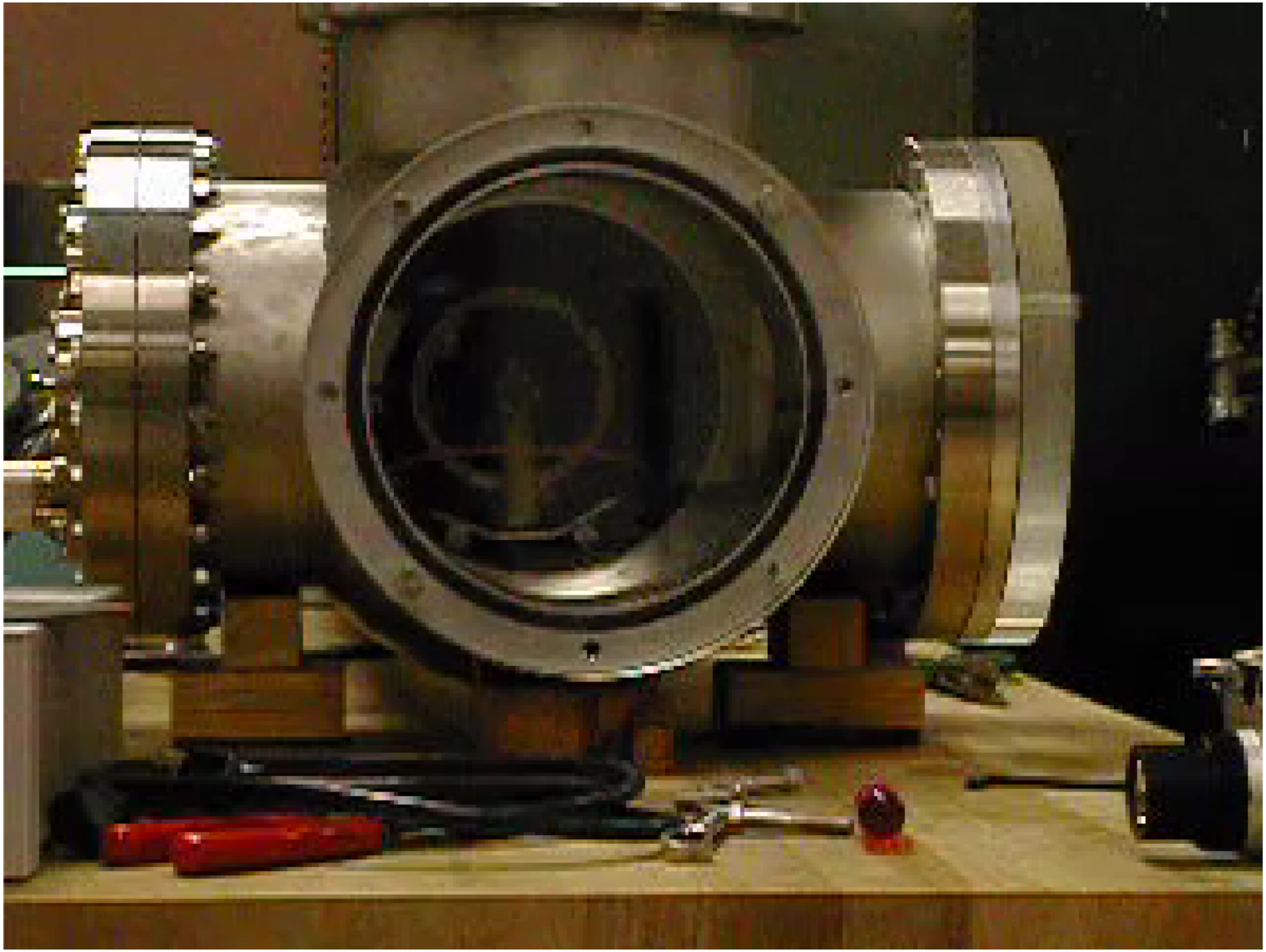


Coaxial Energetic Deposition (CED™)



- CED coater uses "welding torch" technology
- Arc source is scalable to high throughputs for large scale cavity coatings
  - Present version deposits ~1 monolayer/pulse ~0.2 ms
- UHV and clean walls are important

Movie of the arc spoke moving down the tube



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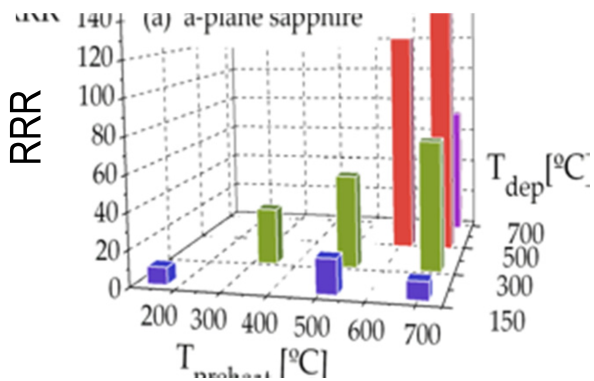
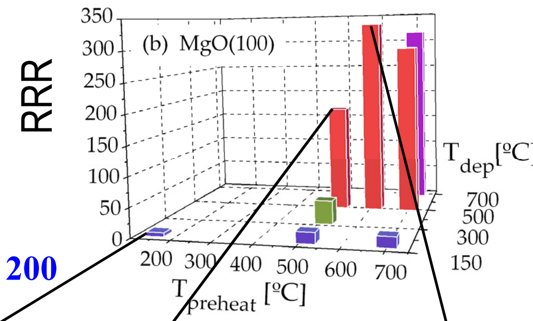
High RRR on coupons motivates coating accelerator structures

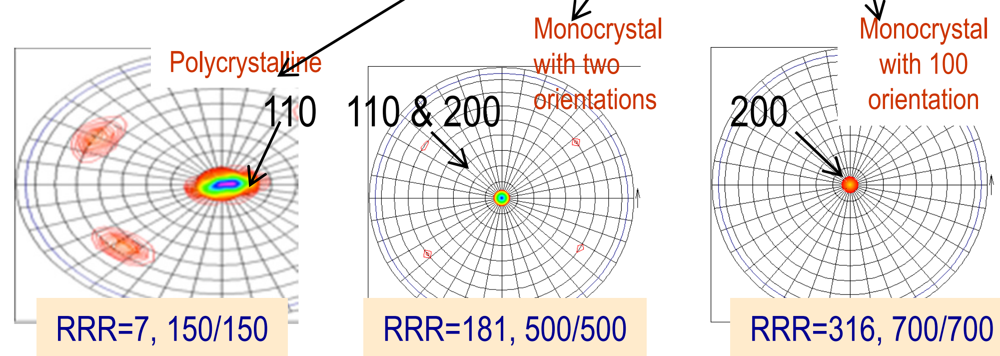


RRR-330 measured on a-sapphire

RRR-585 measured on 5µm film on MgO

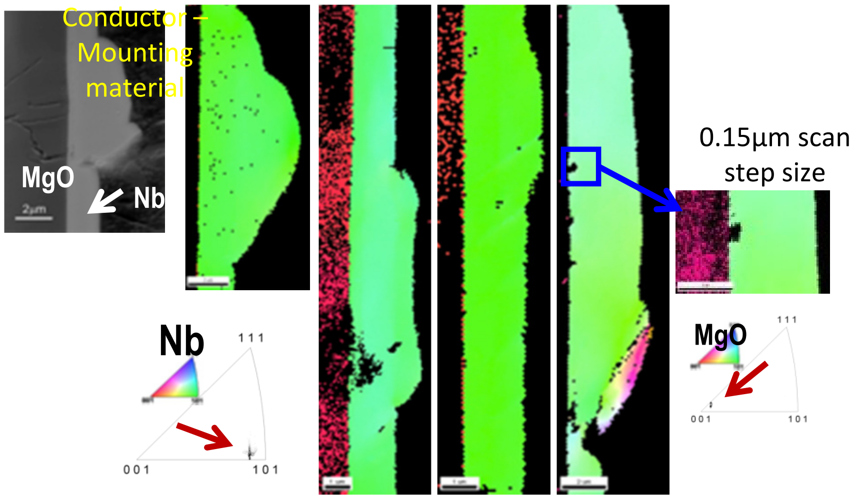
- **Change in crystal orientation from 110 to 200 at higher temperature**





- M Krishnan, E Valderrama, B Bures, K Wilson-Elliott, X Zhao, L Phillips, A-M Valente-Feliciano, J Spradlin, C Reece and K Seo, "Very high residual-resistivity ratios of heteroepitaxial superconducting niobium films on MgO substrates," Superconductor Science and Technology , vol. 24, p. 115002, November 2011
- M. Krishnan, E. Valderrama, C. James, X. Zhao, J. Spradlin, A-M Valente Feliciano, L. Phillips, and C. E. Reece, K. Seo, Z. H. Sung, "Energetic condensation growth of Nb thin films", PHYSICAL REVIEW SPECIAL TOPICS - ACCELERATORS AND BEAMS 15, 032001 (2012)
- X. Zhao, L. Philips, C. E. Reece, Kang Seo, M. Krishnan, E. Valderrama, "Twin symmetry texture of energetically condensed niobium thin films on a-plane sapphire substrate", Journal of Applied Physics, Vol 115, Issue 2, 2011

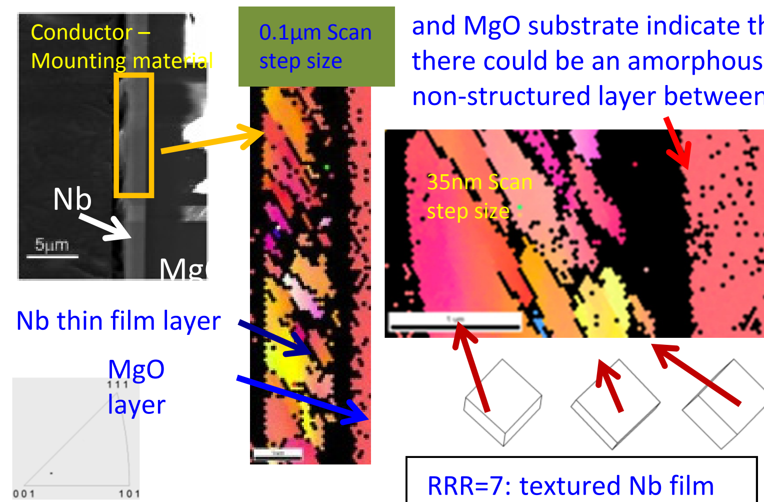
RRR-316 & 7: Cross sectional EBSD and macroparticles



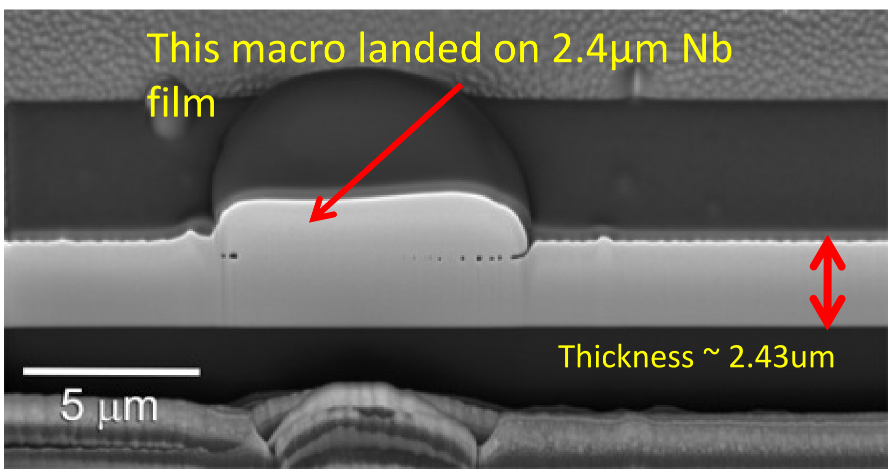
RRR=316: single crystal growth of Nb

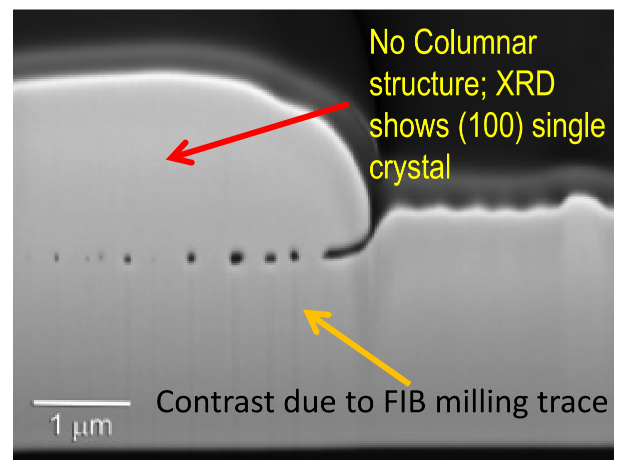
BSE image – X-section view

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



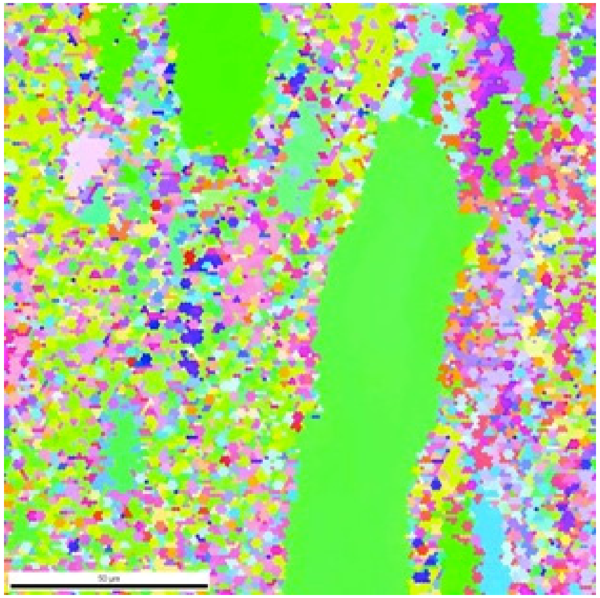
RRR=7: textured Nb film



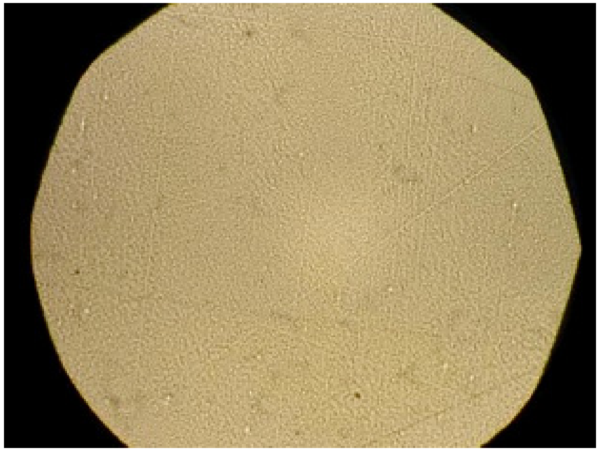


RRR=316: Macro-particle condenses onto Nb film and acquires film's (100) crystalline structure

Nb on copper coupons



EBSD image of Nb on mechanically polished Cu showing large (~75 um) grains

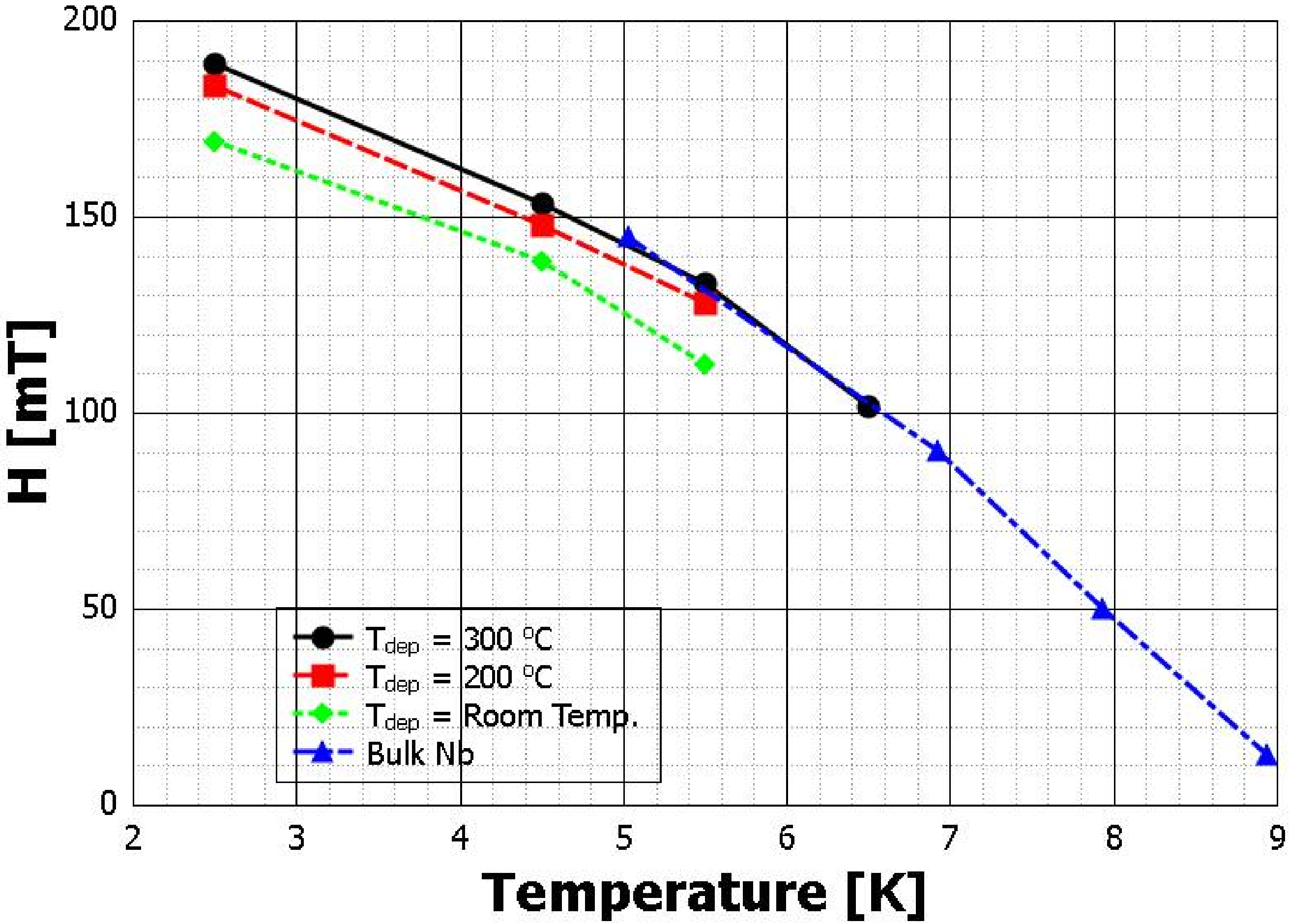


Fine grain Cu coupons polished at AASC

Sample	Substrate	RRR	Tc	Delta Tc	Thickness	Temperature
CED-083	Cu	23	9.37	0.09	3um	200
CED-085	Cu	33	9.37	0.05	10um	200
CED-087	Cu	40	9.41	0.09	3um	300
CED-089	Cu	12	9.21	0.08	10um	300
CED-090	Cu	60	9.55	0.2	3um	400
CED-091	Cu	110	9.44	0.11	10um	400

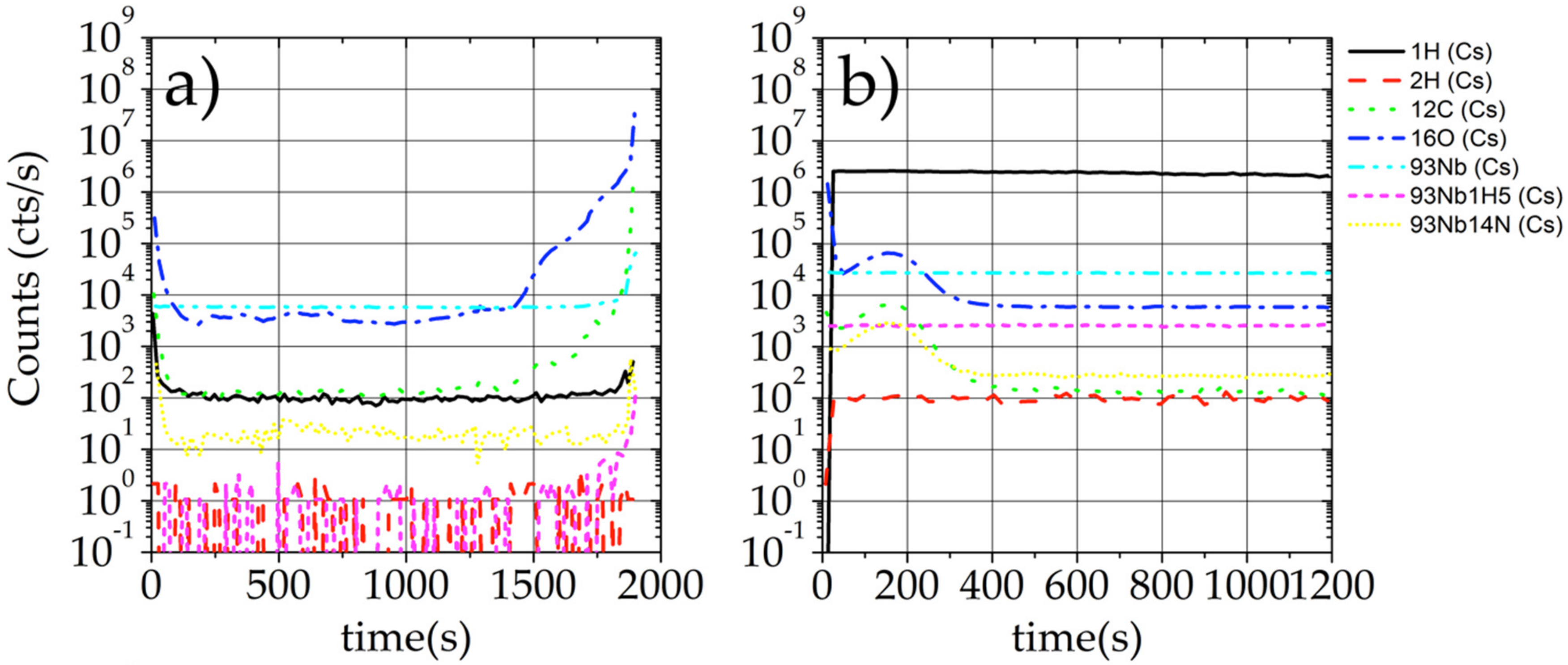
- Nb/Cu results confirm that Nb film quality improves with both deposition temperature and thickness

Magnetic vortex penetration in Nb/Cu films:
Data provided by T. Tajima and L. Civale of LANL



Colt James, Mahadevan Krishnan, Brian Bures, Tsuyoshi Tajima, Leonardo Civale, Nestor Haberkorn, Randy Edwards, Josh Spradlin, Hitoshi Inoue, IEEE Trans. Appl. Supercond. 23

SIMS with Cs beam of high RRR Nb films:

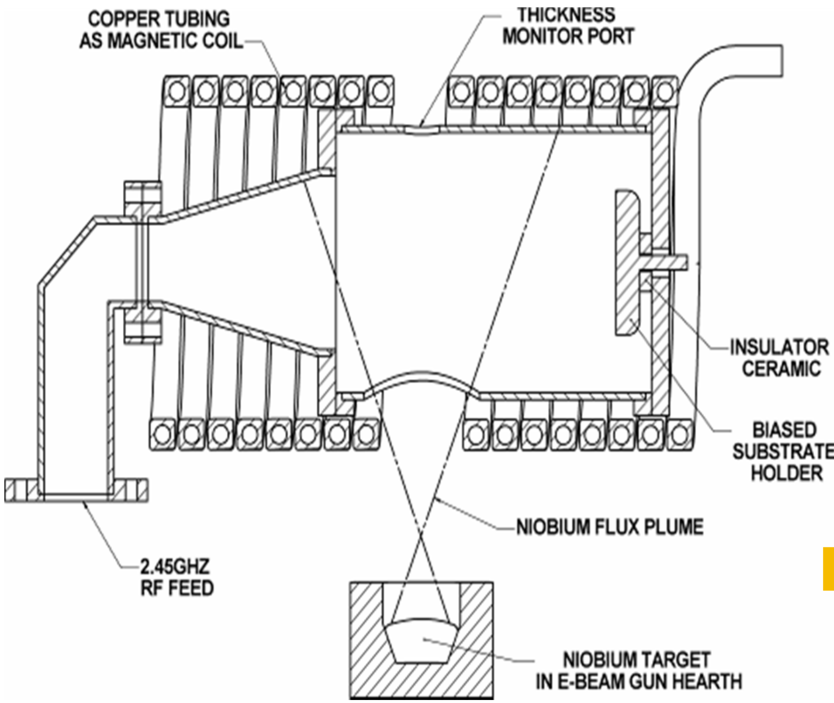


Nb (100) on MgO:
RRR=442

Bulk Nb sample: RRR~300

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

Electron Cyclotron Resonance



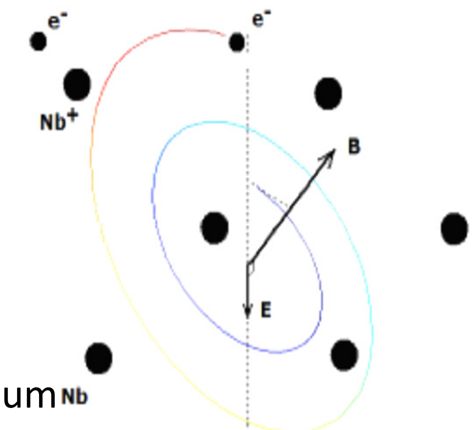
Generation of plasma

3 essential components:

Neutral Nb vapor

ower (@ 2.45GHz)

ic B ⊥ E_{RF} with ECR condition

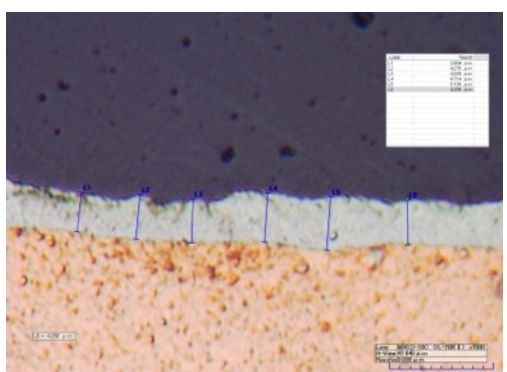


- No working gas
- Ions produced in vacuum
- Singly charged ions 64eV
- Controllable deposition energy with Bias voltage
- Excellent bonding
- No macro particles
- Good conformality

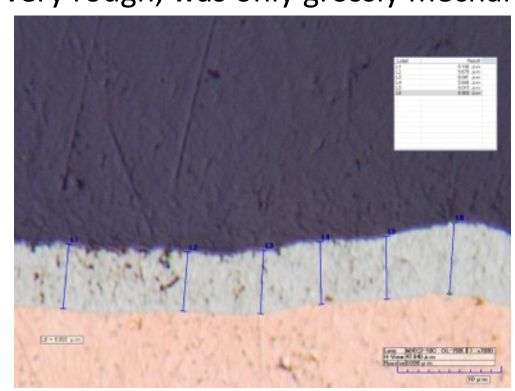
A 3Gz half-cell was coated in the ECR chamber: The film thickness along the cell profile varies from 4µm (equator) to 6µm (iris)

→conformality of the ECR process

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

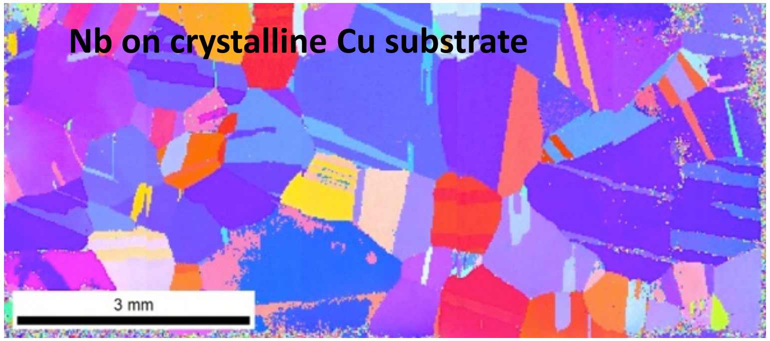


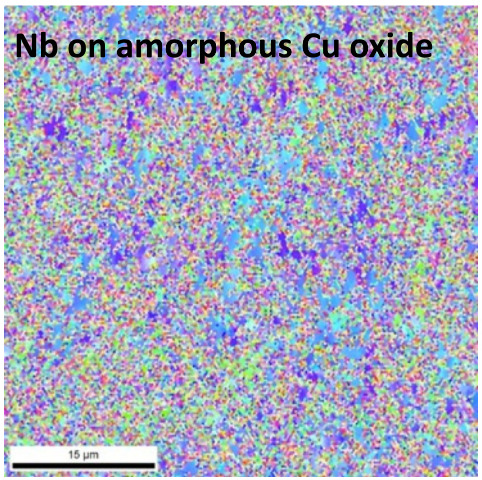
Location	Thickness [µm]
L1	3.804
L2	4.275
L3	4.200
L4	4.714
L5	5.136
L6	4.288

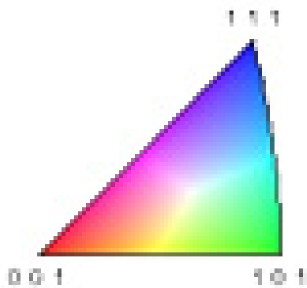


Location	Thickness [µm]
L1	6.189
L2	5.675
L3	6.081
L4	5.986
L5	6.015
L6	6.893

EBSD IPF maps





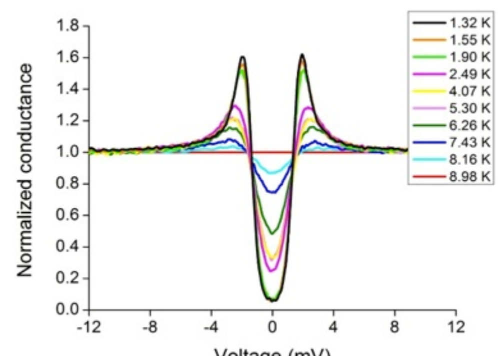


Substrate	RRR
Single crystal	
Cu (100)	**129**
Cu (110)	**275**
Cu (111)	**245**
Polycrystalline	
Cu fine grains	**150**
Cu large grains	**289**

Substrate	RRR
Single crystal insulator	
MgO (100)	**176**
MgO (110)	**424**
MgO (111)	**197**
a-Al_2O_3	**488**
c-Al_2O_3	**247**
Amorphous	
Al_2O_3 ceramic	**89**
AlN ceramic	**74**
Fused Silica	**43**

Tune thin film structure and quality with ion energy and substrate temperature : RRR values from single digits to bulk Nb values.

Gap measurements performed by PCT (point contact tunneling spectroscopy- ANL) show a superconducting gap (1.56-1.62meV) similar to bulk Nb ($\Delta_{Nb\ bulk}$ =1.55meV measured on the same setup) for hetero-epitaxial ECR Nb films on polycrystalline Cu.

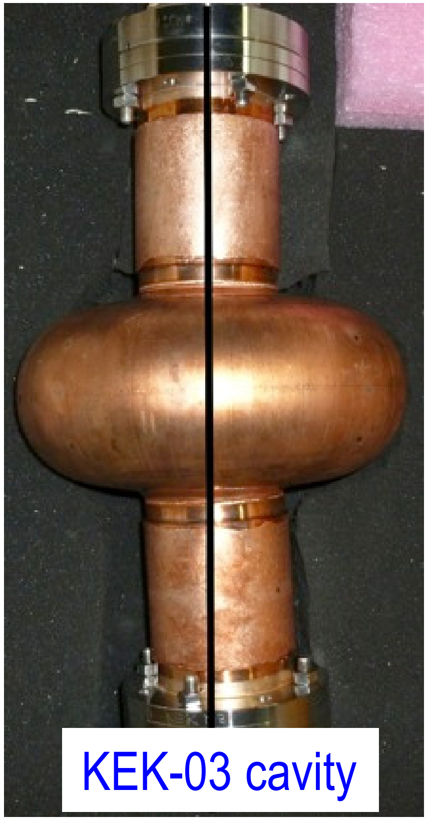


Outline

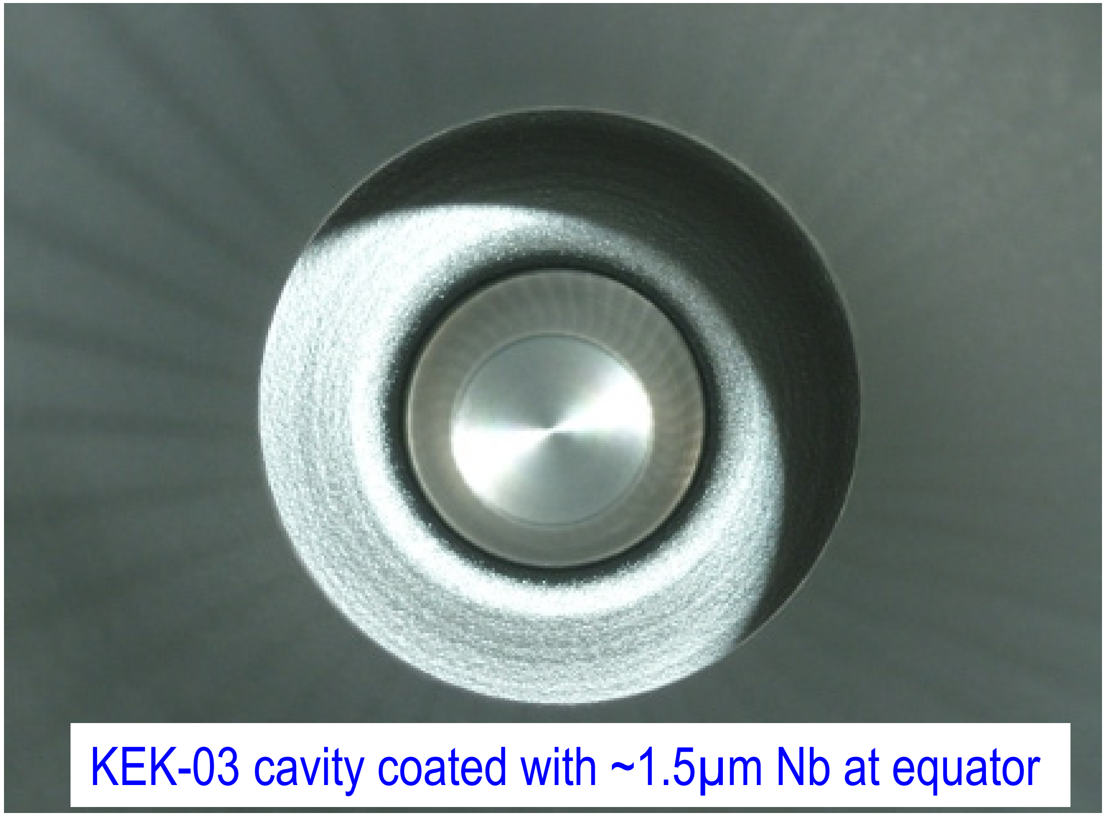
- Motivation for thin films in SRF cavities
- History of thin film development
- Energetic Condensation
- Morphology of thin films produced by energetic condensation
- Thin films on Cu and Nb cavities
- Future Plans

KEK03 hydro-formed Cu cavity cell

- 13,500 pulses at a cavity temperature of 330C (measured half way down the beam tube); ~5μm Nb film in the beam tube and ~1.5μm at the equator.



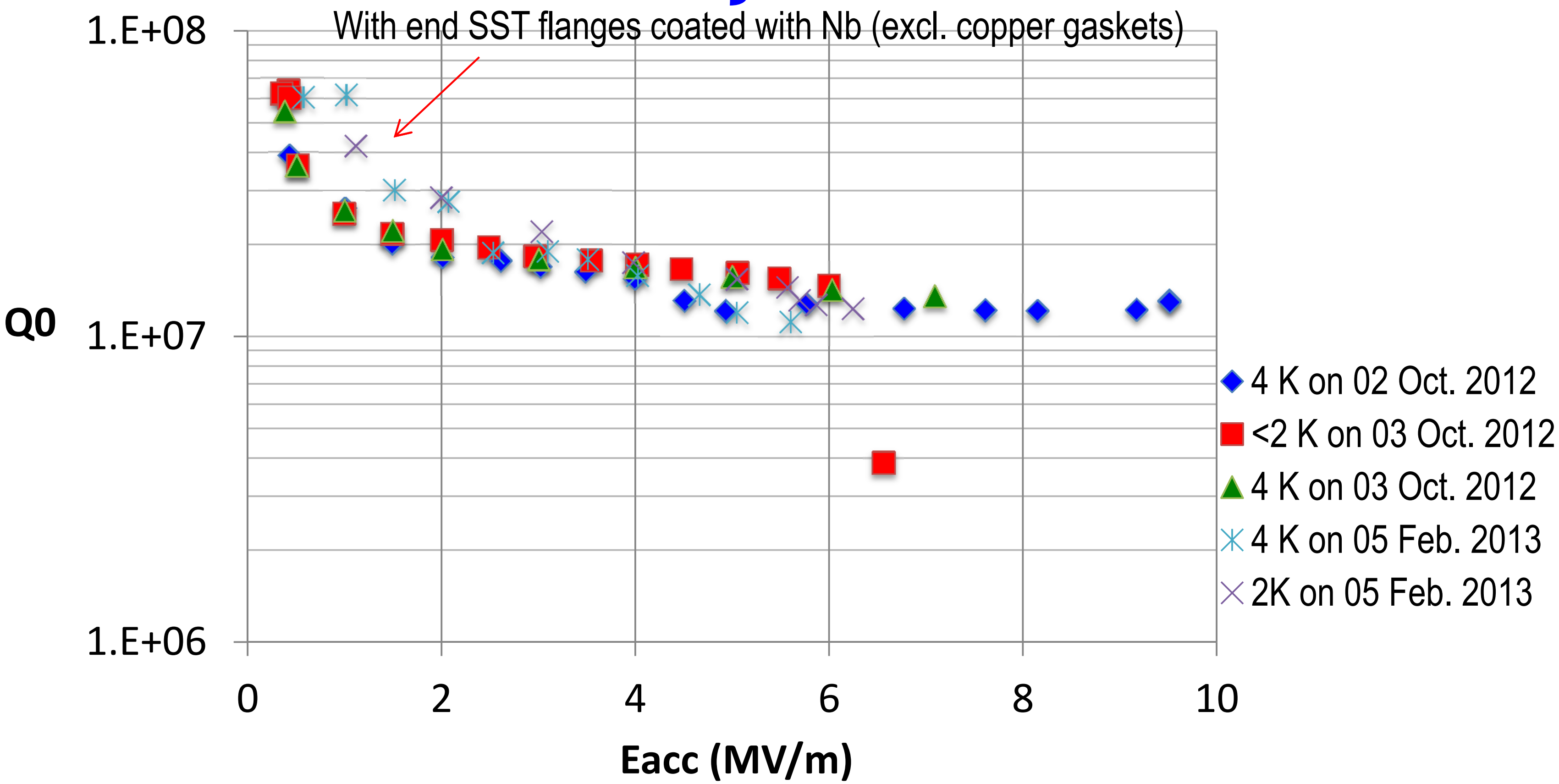
KEK-03 cavity



KEK-03 cavity coated with ~1.5μm Nb at equator

- Cavity provided by T. Tajima/LANL and tested at LANL

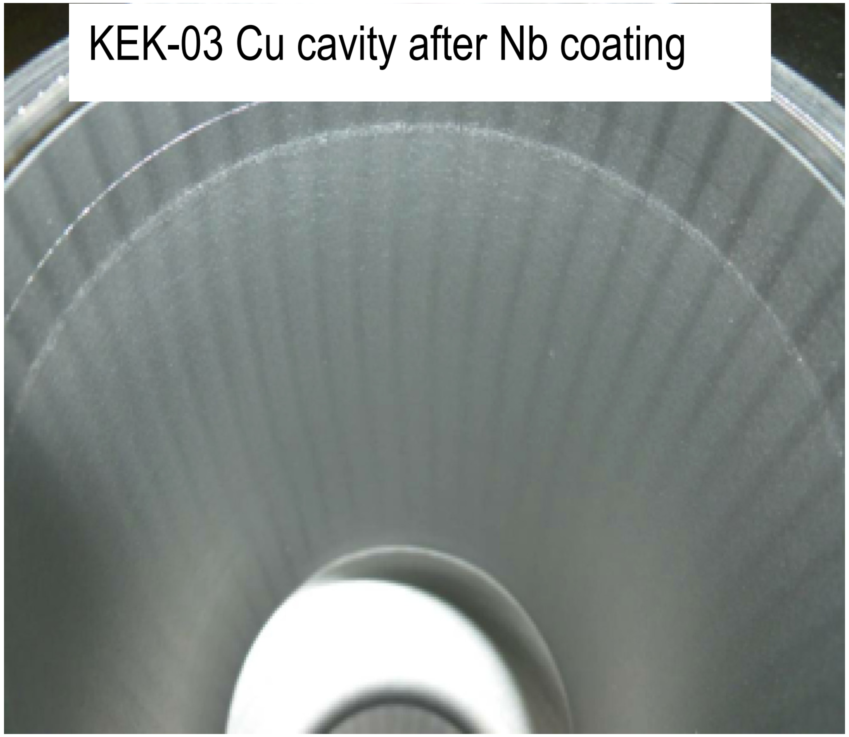
KEK03 Cu cavity cell RF tested @LANL by T. Tajima

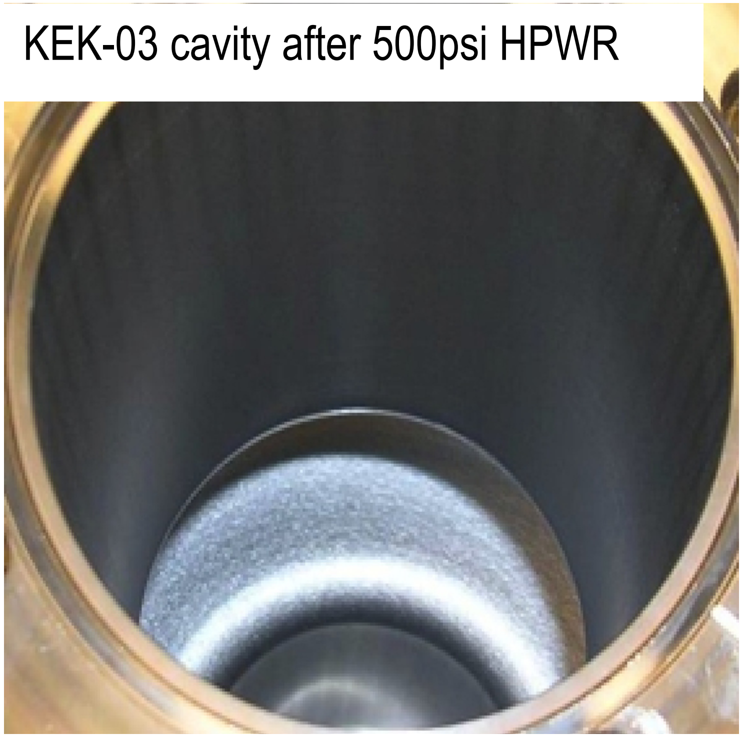


- No difference between 4 K and 2 K results suggests that losses are due to beam-pipe flanges; Cavities with 50 mm longer beam pipes were next coated.

KEK03 Cu cell coating survived HPWR @ LANL

- Cavity provided by T. Tajima/LANL (via KEK) and tested at LANL

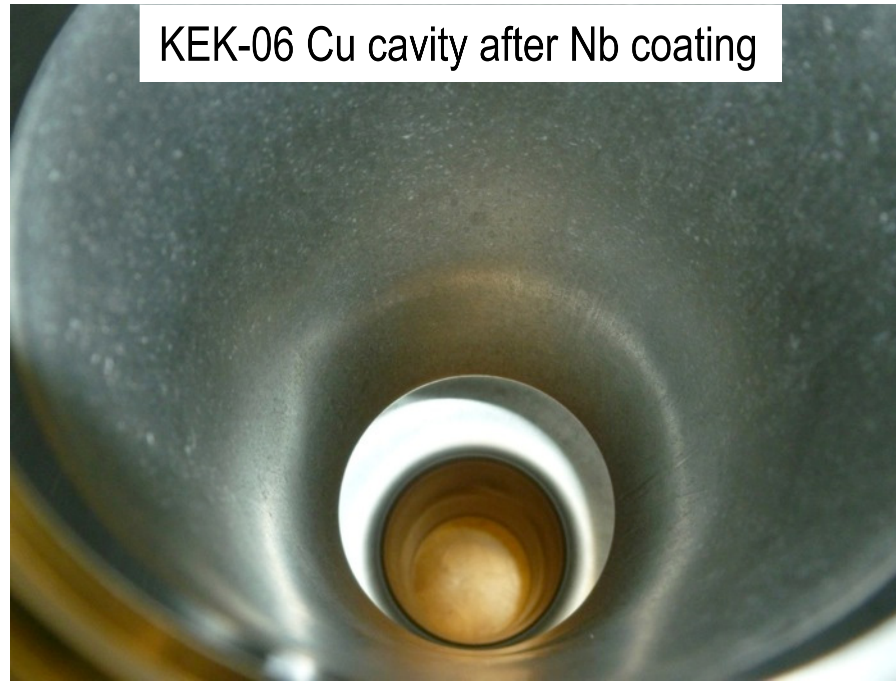




- The film appears to adhere well to copper

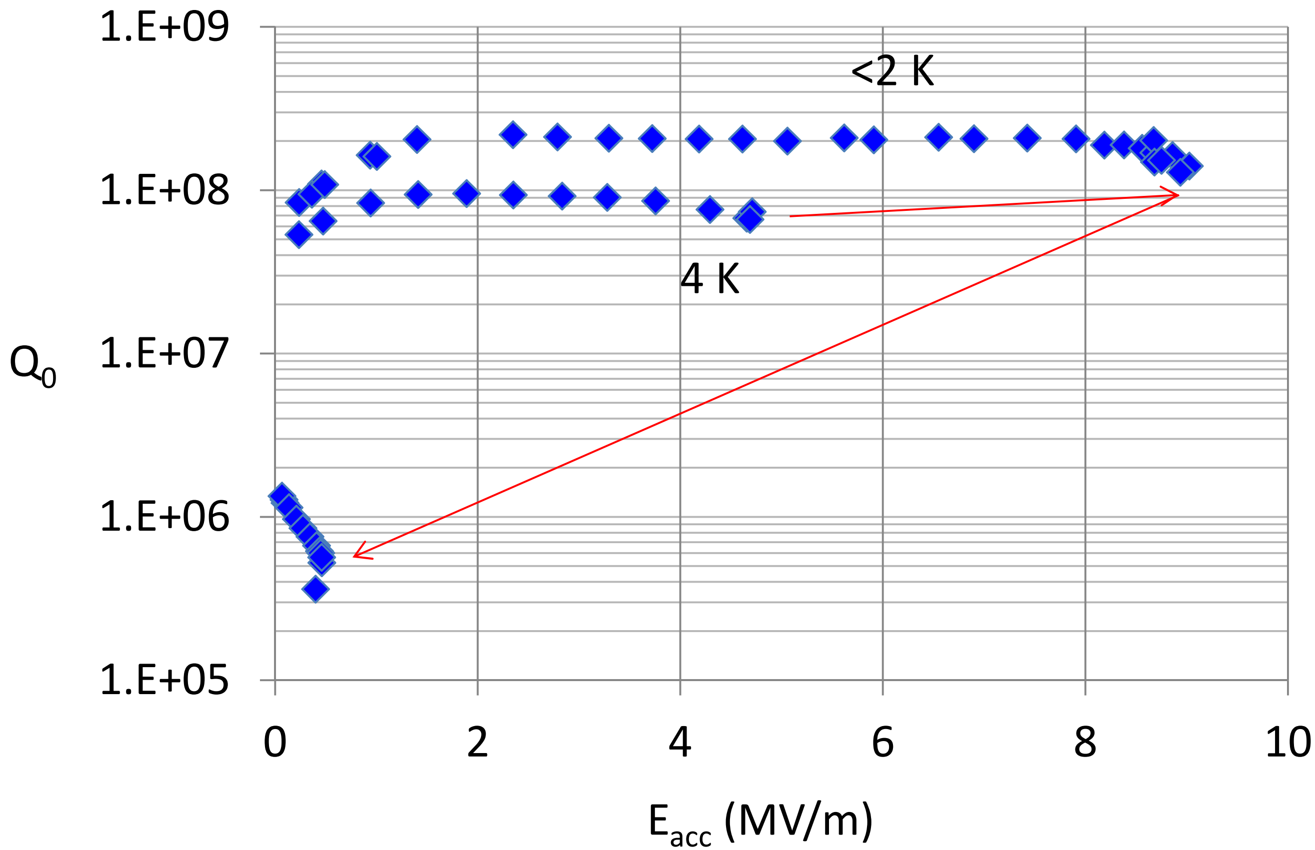
KEK06 hydro-formed Cu cavity cell from T. Tajima

- This cavity used 12500 shots at 350-360C to produce a fully covered coating



- KEK-06 was tested at LANL

KEK06 hydro-formed copper cavity, CBP at FNAL (Cooper), coated at AASC, tested at LANL (Tajima, Haynes)

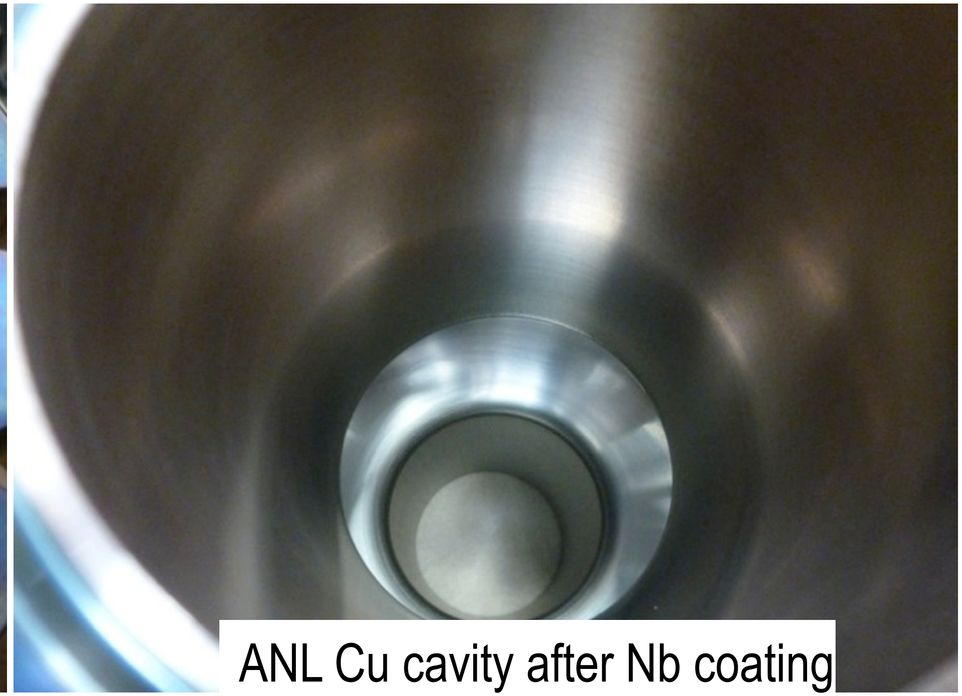


- The difference between 4K and 2K is smaller than expected: BCS resistance should be about 40x less if all the surfaces are Nb.
- This suggests that there are areas that are not coated well and lossy, which is causing the lower than expected Q_0

Cu cavity cell from ANL (T. Proslier) coated with Nb

- 12,500 pulses at a cavity temperature of 350C (measured half way down the beam tube); ~5μm Nb film in the beam tube and ~1.5μm at the equator.

ANL Cu cavity before Nb coating

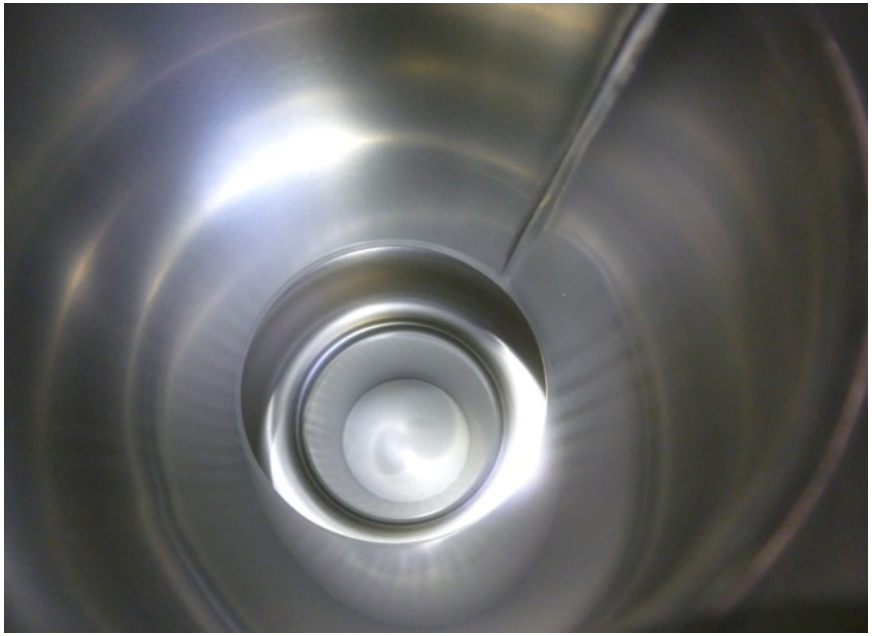


ANL Cu cavity after Nb coating

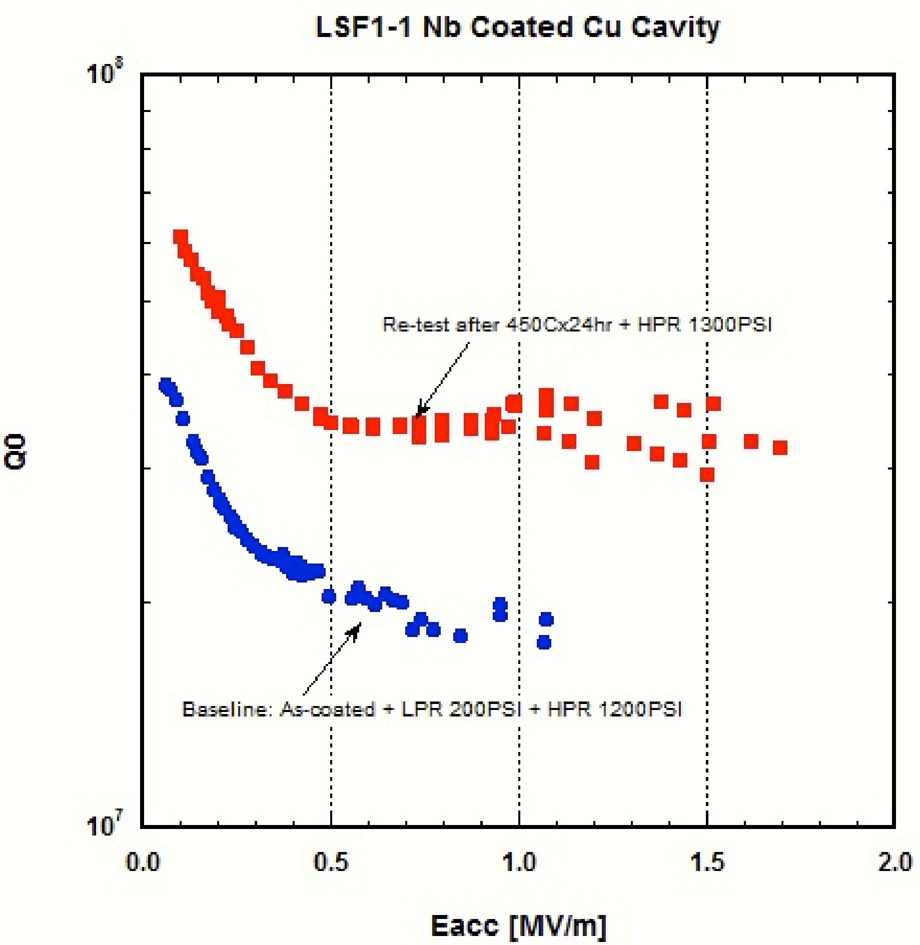
- Cavity provided by T. Proslier/ANL and tested at JLab on Monday Sep. 23 by P. Kneisel
- Result was disappointing; Q_0~ 10^7 at 4K

Cu cavity cell from JLab (Rong-Li Geng) coated with Nb

- ~5μm Nb film in the beam tube and ~1.5μm at the equator.



JLab Cu cavity after Nb coating



- Result was consistent with KEK/LANL and ANL cavities and consistently disappointing; $Q_0 \sim 10^7 - 10^8$ at 4K

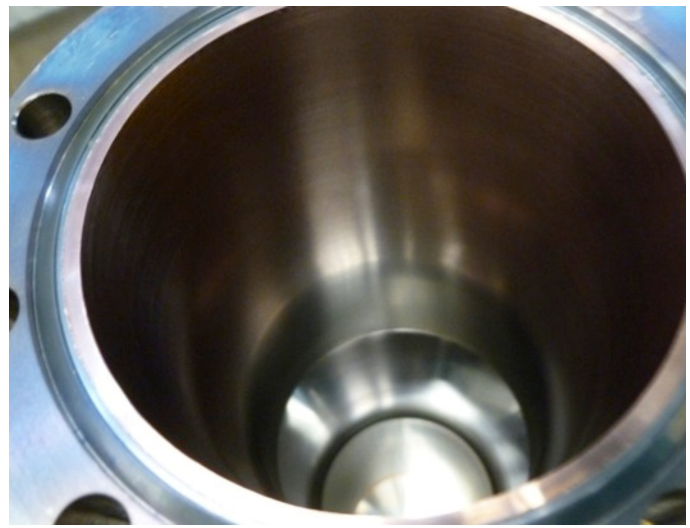
Trials and Tribulations of R&D

- The KEK-04 cavity suffered a melted heater!

- We learned how to melt Aluminum

Trials and Tribulations of R&D

- The first ANL cavity coating suffered from incomplete coverage due to a software glitch
- One beam tube was fully coated, whilst the other was partially coated



- RF tests (P. Kneisel/JLab showed poor performance; this cavity was recoated and tested again at JLab; but second result was also poor.

Trials and Tribulations of R&D

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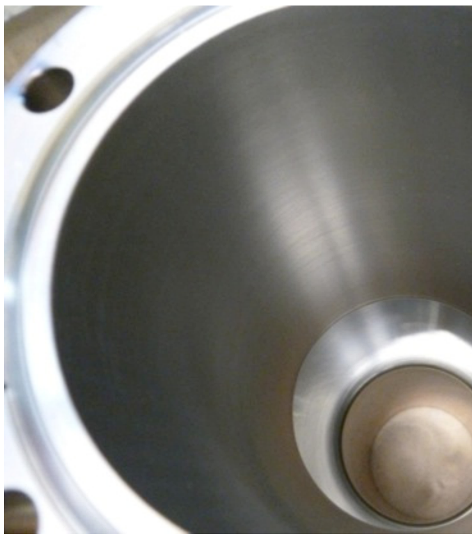

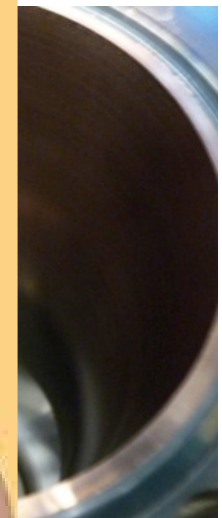

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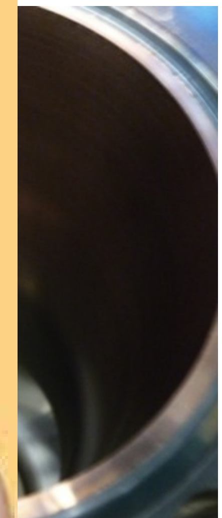
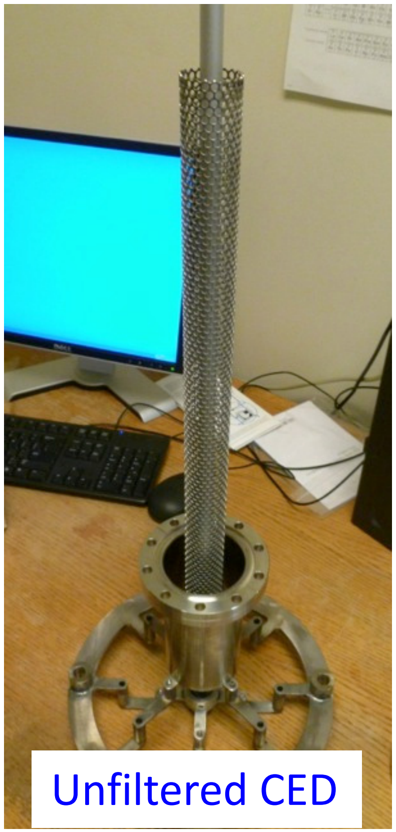


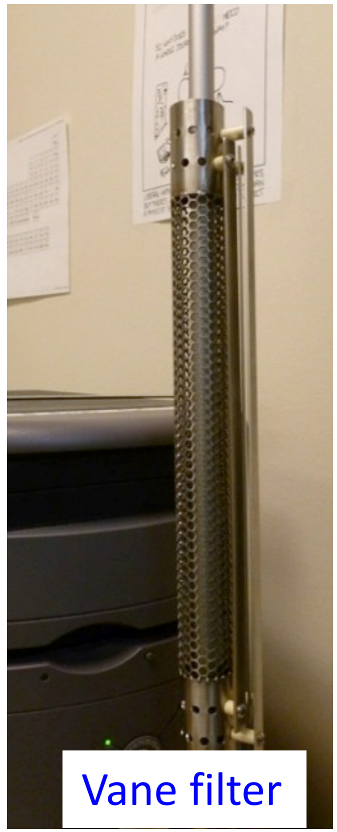
- RF tests (P. Kneisel/JLab showed poor performance; this cavity was recoated and tested again at JLab; but second result was also poor.

The Macroparticle Problem

- As Russo et al had pointed out, macroparticles must be filtered out of the plasma stream for higher quality SRF films



Unfiltered CED



Vane filter

Close-up of vane filter

- AASC has designs for “vane filters” [passive and active] under test