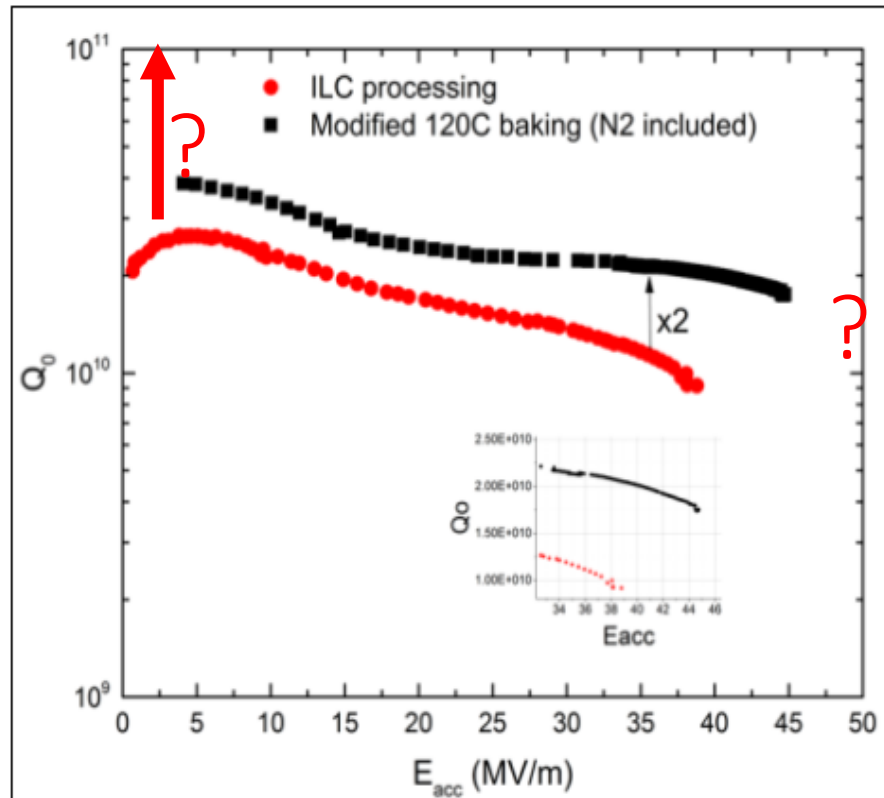


MATERIALS FOR SUPERCONDUCTING ACCELERATORS

BEYOND BULK Nb

A-M VALENTE-FELICIANO

Why looking beyond bulk Nb?



A. Grassellino and S. Aderhold, TTC meeting, Saclay, France (2016)

So far $Q(H)$ and E_{acc} of Nb cavities have been rising ...

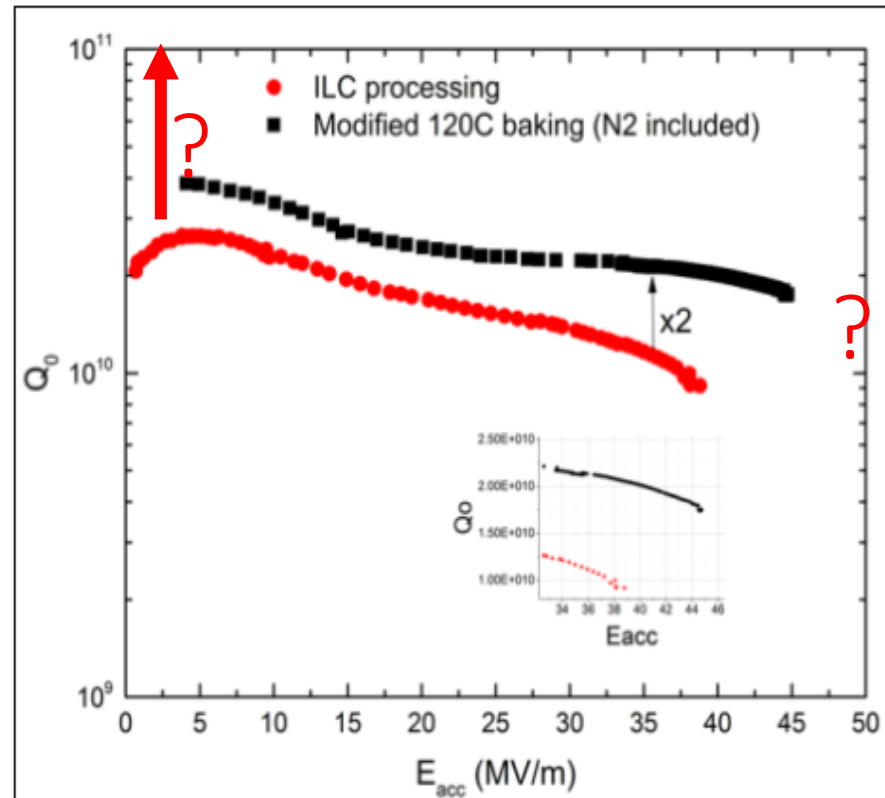
Fundamental limits of $Q(H)$ and the breakdown field?

Decrease of $Q(H)$ is consistent with current pairbreaking effects

How close is Nb to the fundamental instability of the Meissner state?

Breakdown fields close to the de-pairing limit of 50 MV/m for Nb have been achieved
Best Nb cavities approaching their intrinsic limit at $H_{max} = H_c$

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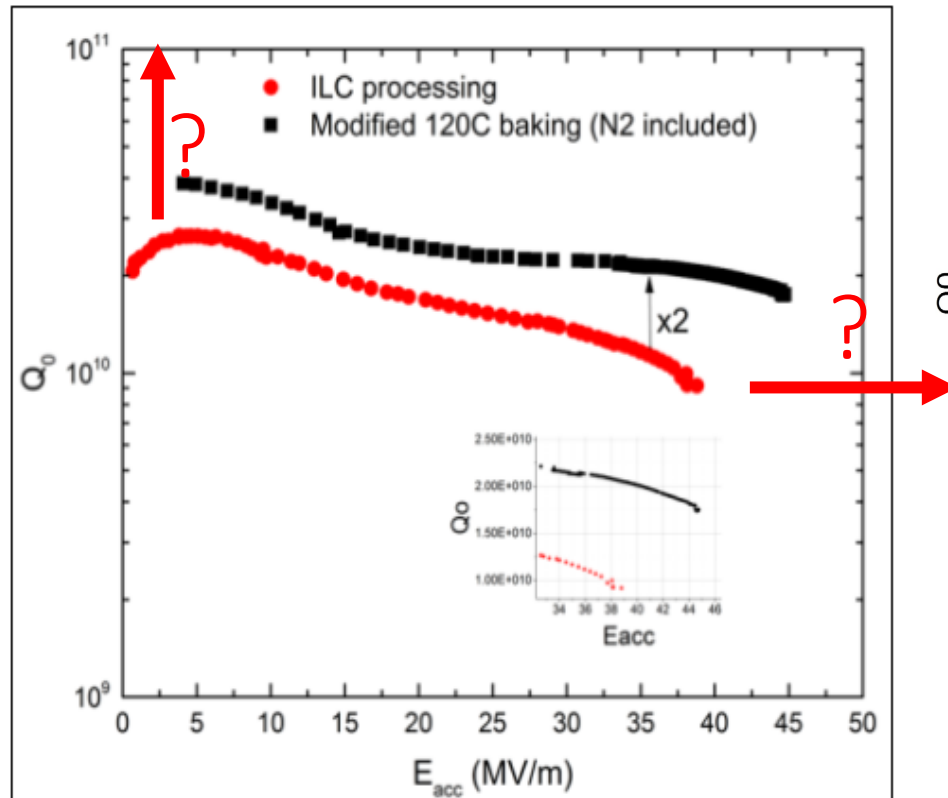
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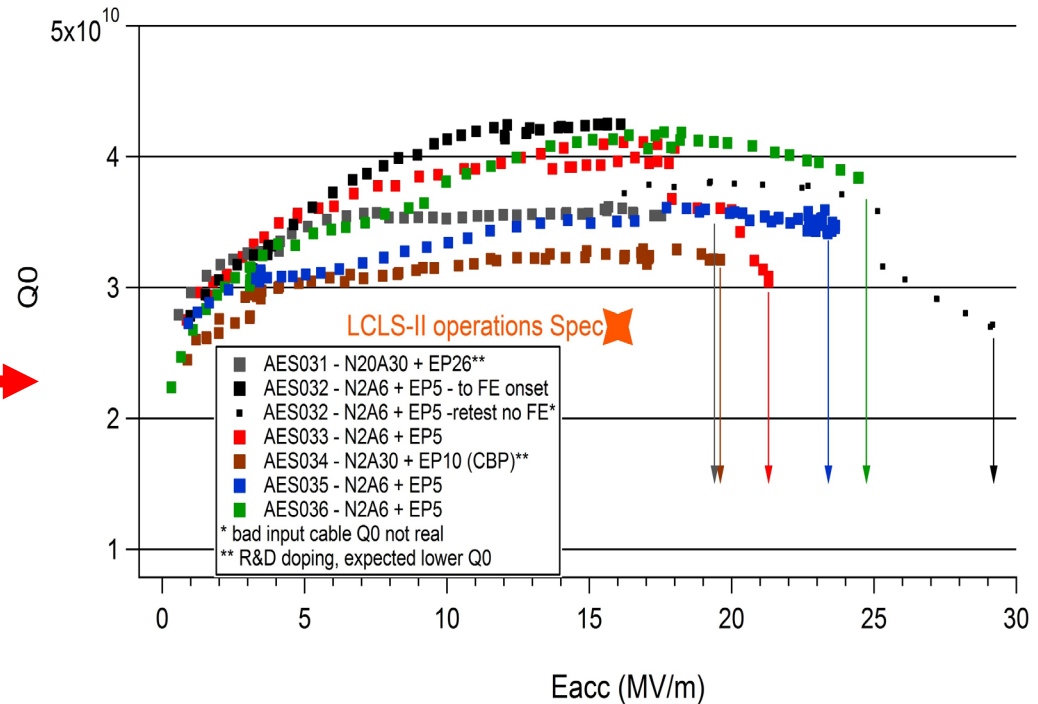
Best Nb cavities approaching their intrinsic limit at $H_{max} = H_C$

Recent improved cavity RF performance in Q with N doping/infusion

Why looking beyond bulk Nb?



A. Grassellino and S. Aderhold, TTC meeting, Saclay, France (2016)



All cavities were tested with Stainless steel flanges which add 1.4 nΩ. Residual resistance with 1.4 nΩ has been subtracted from the data. –

Breakdown fields close to the de-pairing limit of 50 MV/m for Nb have been achieved

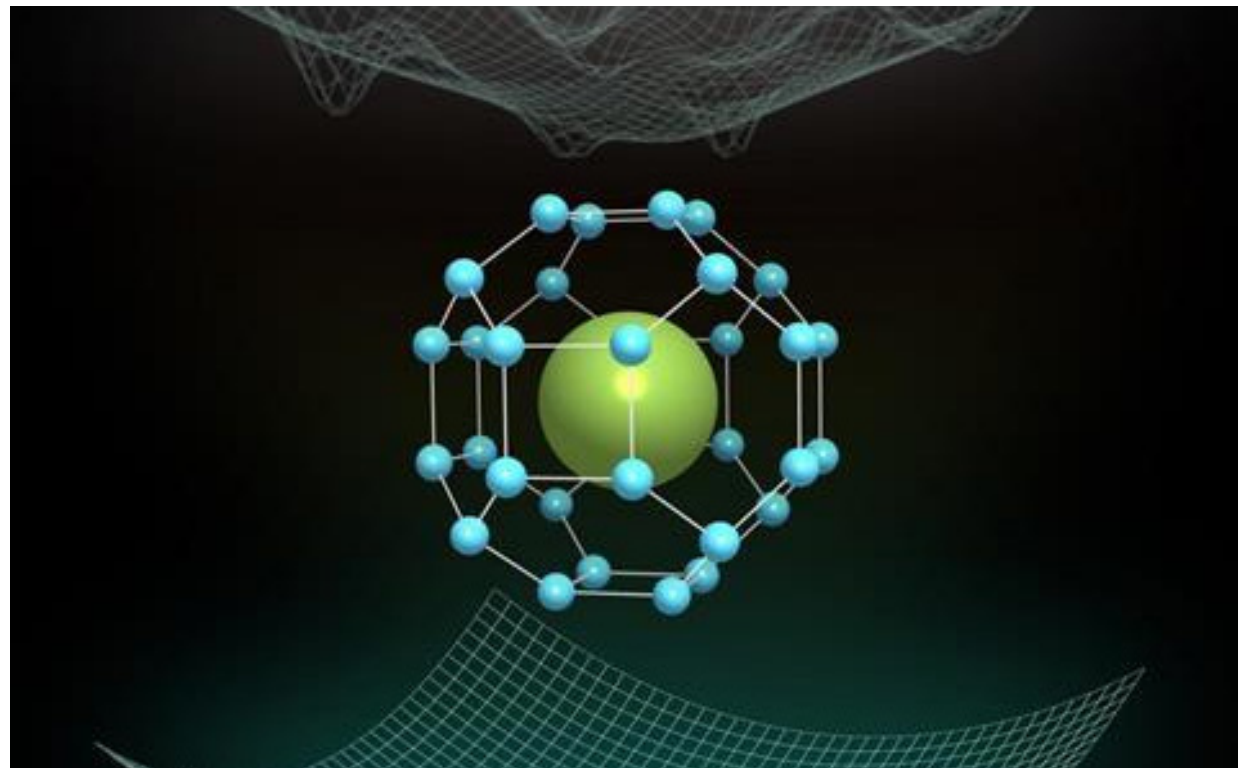
Best Nb cavities approaching their intrinsic limit at $H_{\max} = H_c$

Recent improved cavity RF performance in Q with N doping/infusion

For further improvement, innovation needed

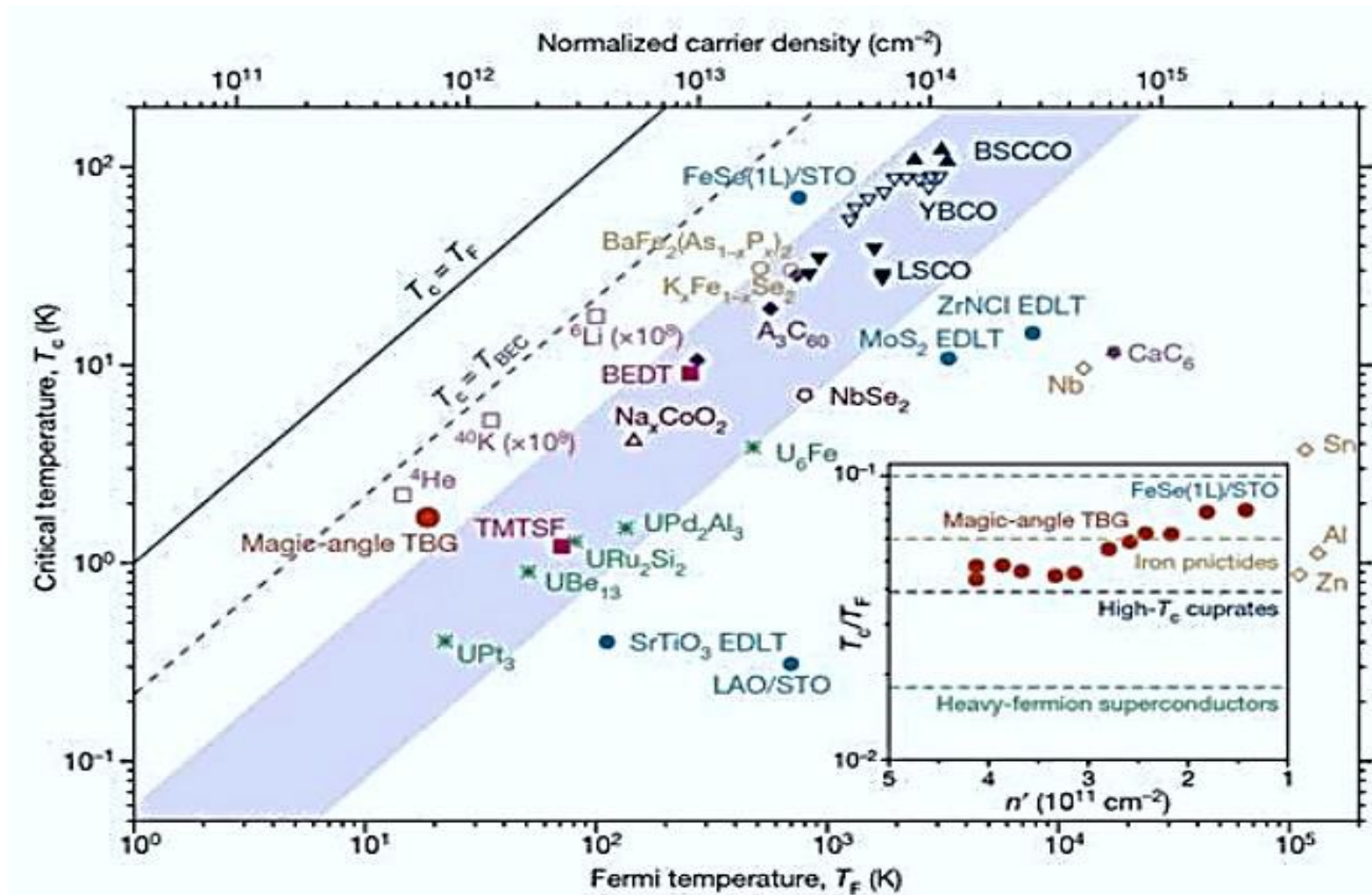
Possibilities to use higher performance superconductors other than bulk Nb?

RF SUPERCONDUCTOR: CHOICE CRITERIA ?



THOUSANDS OF SUPERCONDUCTORS ...

Thousands of SC exist, ~10 are currently used for applications, only bulk Nb works well for SRF !!!

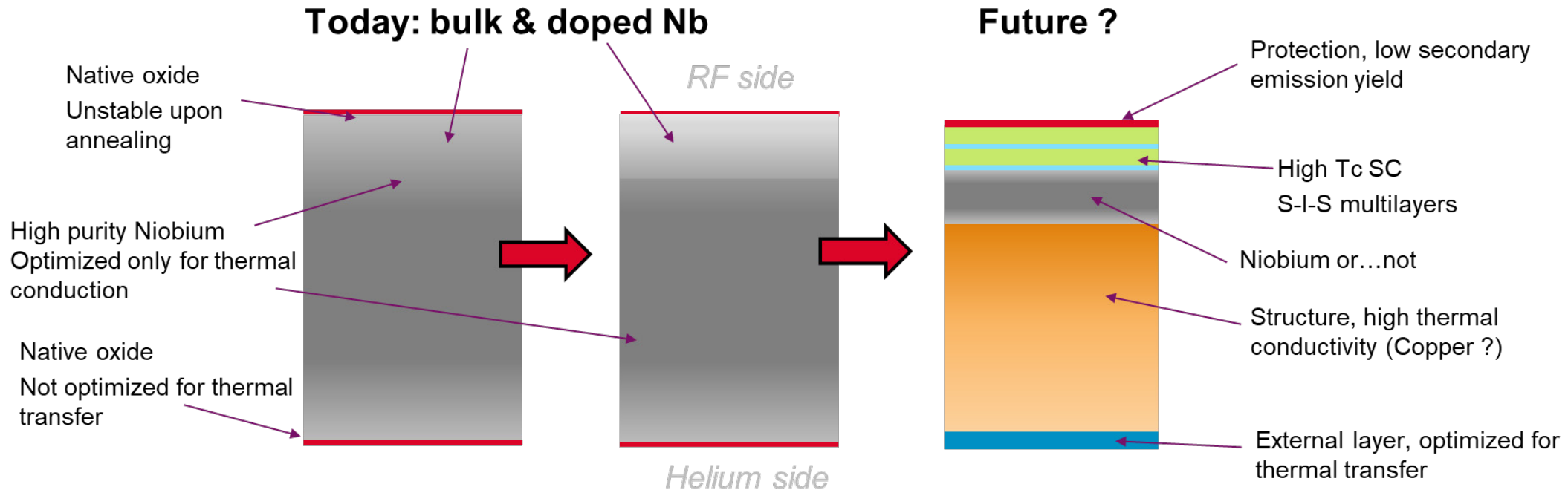


- ✱ Heavy-fermion superconductors
 - ▼ Cuprates
 - Iron pnictides
 - ◇ Conventional superconductors
 - BEC in atoms
 - Two-dimensional materials
 - Organic superconductors
 - ◆ A₃C₆₀
 - NbSe₂
 - △ Na_xCoO₂
 - CaC₆
 - Magic-angle TBG
- Bose Einstein Condensation*
- Twisted Bilayer Graphene*

Source: © 2018 Macmillan Publishers Limited, part of Springer Nature

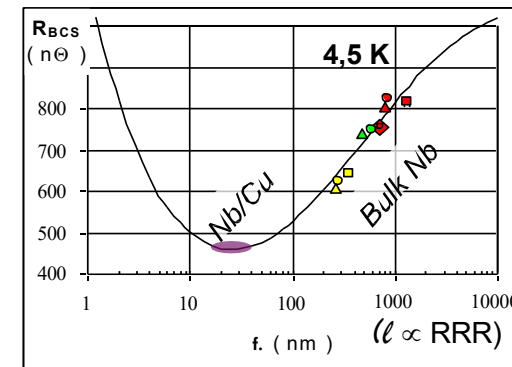
IDEAL SRF MATERIAL: TAILORED FOR APPLICATIONS

High RRR not required for superconductivity but for thermal stabilization in case of defects



“RF” layer optimized for superconductivity:
 Low surface resistance (medium m.f.p.)
and/or *High H_P (high H_{C1} or high H_{SH})

* *Depends on the application*



ULTIMATE LIMITS IN SRF

Niobium superconducting radiofrequency cavities

❑ Performances

$$E_{\text{acc}} \propto H_{\text{RF}}$$

$$Q_0 (\propto 1/R_S) \propto T_C \Rightarrow \text{Nb}_3\text{Sn, MgB}_2, \text{NbN... (but not YBCO)}$$

Limit = magnetic transition of the SC material @ H_{peak}

❑ Superconductivity only needed inside :

Thickness $\sim 10 \mu\text{m}$

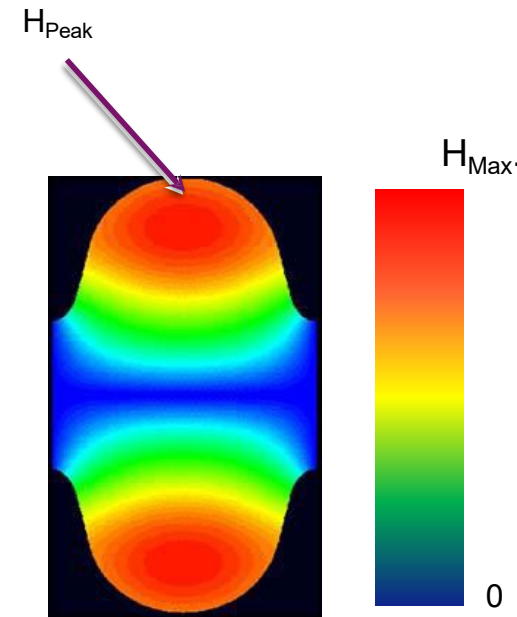
$\lambda \sim 1 \mu\text{m} \Rightarrow$ thin films (onto a thermally
conductive, mechanically resistant material, e.g. Cu)

❑ Today :

Thin films exhibit too many defects

Only Bulk Nb has high SRF performances (high Q_0 and high E_{acc})

❑ Issues : getting “defect free” superconductors



H field mapping in
an elliptical cavity

(Yes but not all defects are detrimental... See doping !)

HIGH Q_0 , E_{ACC} IN SRF => MEISSNER STATE !

SC phase diagram

- All SC applications except SRF: mixed state w. vortex

Vortices dissipate in RF !

- SRF => Meissner state mandatory !

Limit ?

- H_{C1} = limit Meissner/mixed state
 - Nb: highest H_{C1} (180 mT)

Or

- H_{SH} "Superheating field": Metastable state favored by $H //$ to surface

 **Difficult to get in real life !**

Surface resistance:

$$R_S = R_{BCS}(T) + R_{res}$$

$$R_{BCS} = A(\lambda_L^4, \xi_F, \ell, \sqrt{\rho_n}) \frac{\omega^2}{T} e^{-\Delta/kT}$$

Not one formula predicting R_{res} , at least $\propto \sqrt{\rho_n}$

Variety of phenomena involved

Intrinsic: Inhomogeneties, Metallic Inclusions within I, GB, Oxides

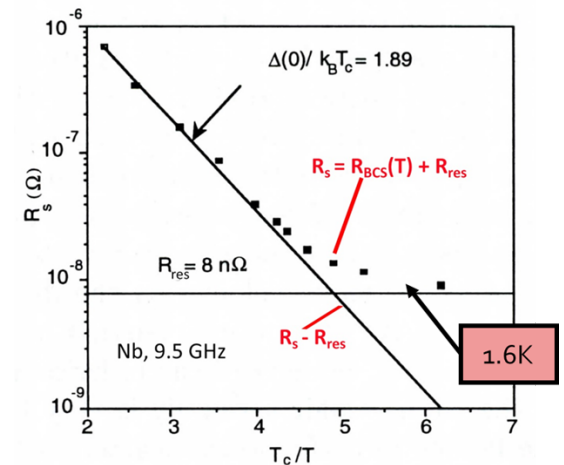
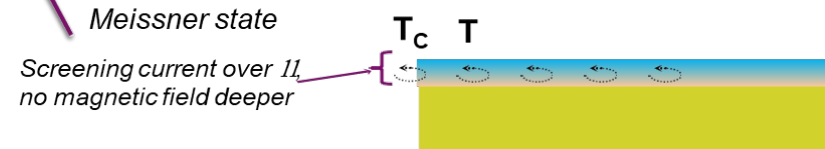
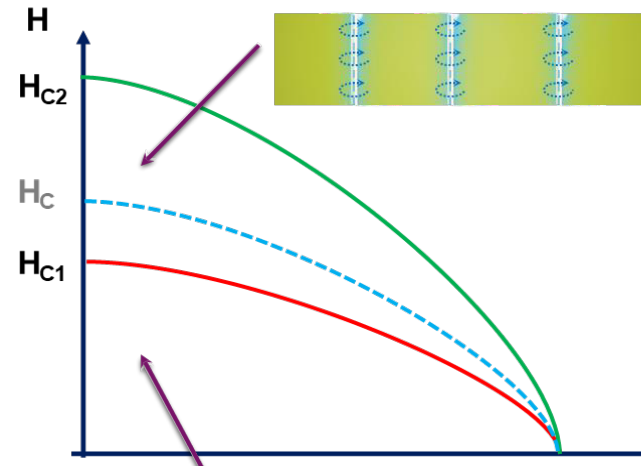
Extrinsic: Trapped Flux during cooling (can be avoided)

High T_c is better $T \ll T_c$ is better ($e^{-\Delta/kT}$)

Metallic character in NC state is better (ρ_n)

Dirty is better than high RRR (|) ? (e.g. doping, but more complex than that !)

Mixed state w. Vortex (i.e. N. cond. flux line + screening currents)



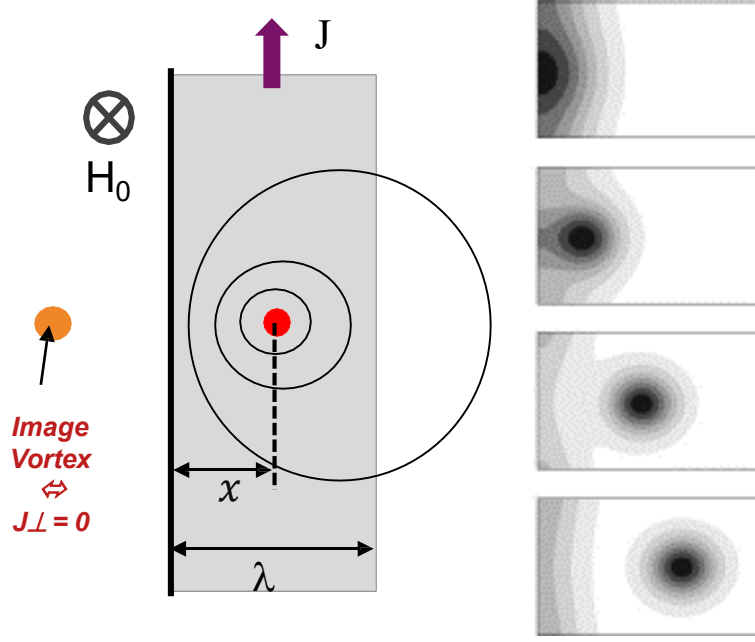
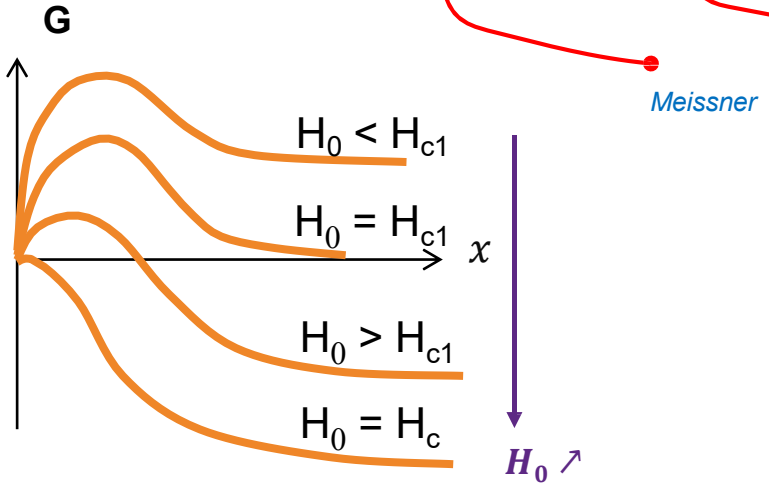
VORTEX PENETRATION WITH B //

Surface barrier

(Bean & Livingston, 1964)

- Boundary condition. ($J_{\perp} = 0$) \equiv “image” vortices
 - Supercurrent tends to push V_x inside
 - Image antivortex tends to pull it out
- Before entering the material V_x have to cross a surface barrier:
 - V_x thermodynamic Potential :

$$G(x) = \phi_0 \left[H_0 e^{-x/\lambda} - H_v(2x) + H_{c1} - H_0 \right]$$



“Ideal surface”

- Barrier disappears only at $H_{SH} \sim H_c > H_{c1}$
- Rationale used to predict SRF limits

BUT

- If \exists localized defect w.: $H_c^{local} \ll H_c^{bulk}$ (or $T_c^{local} \ll T_c^{bulk}$)
 \Rightarrow early penetration of 1 or several V_x there

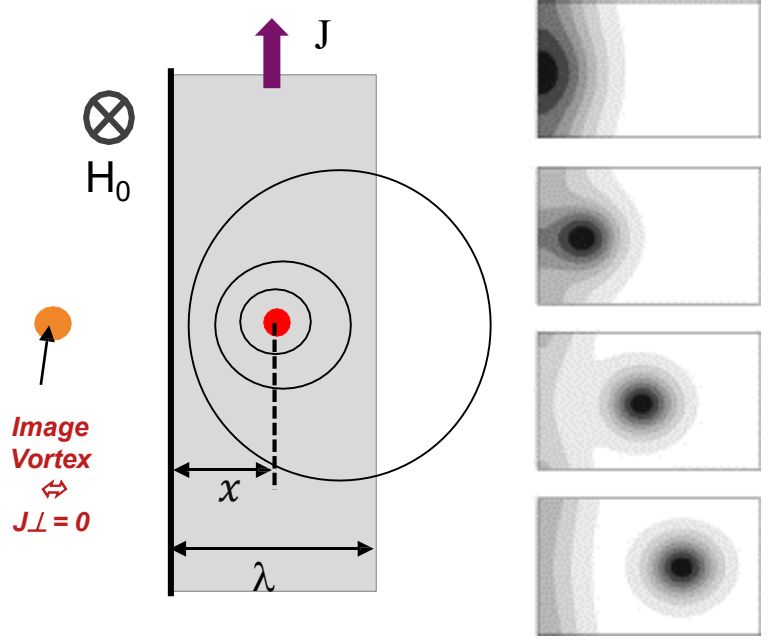
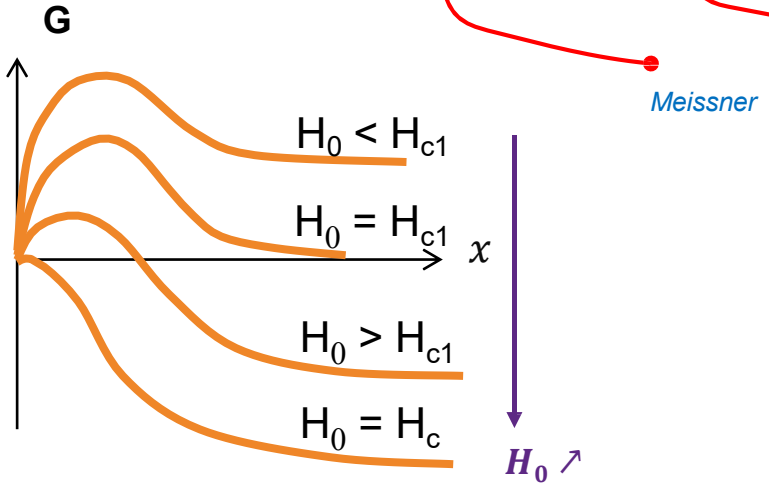
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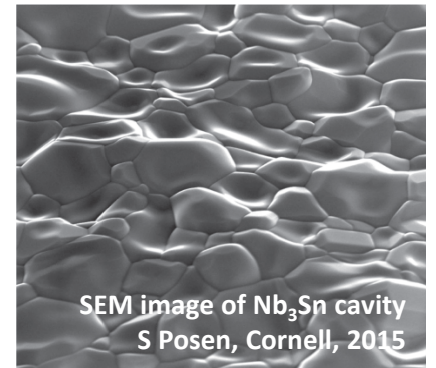
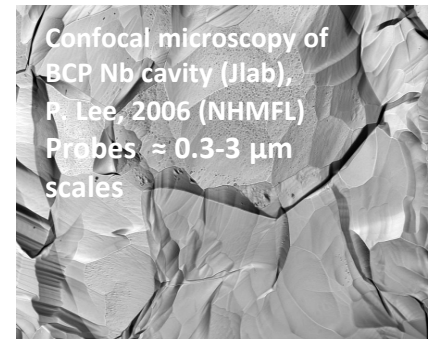
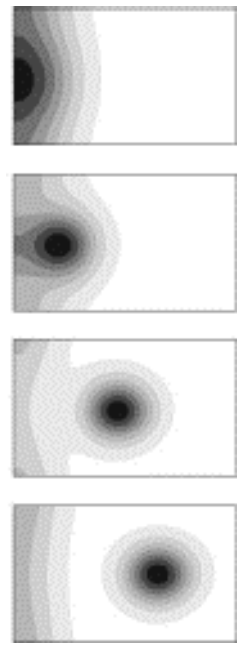
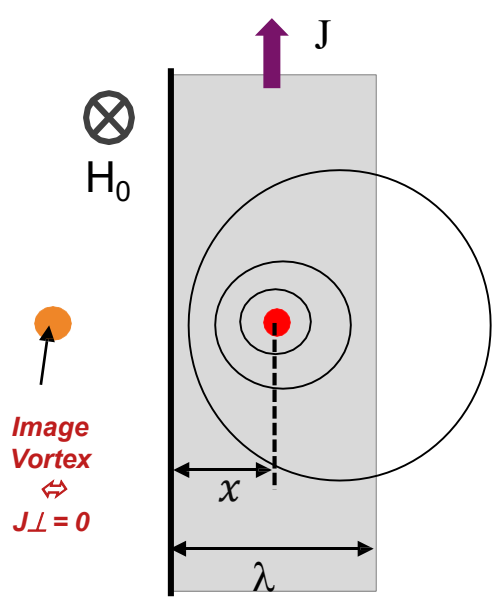
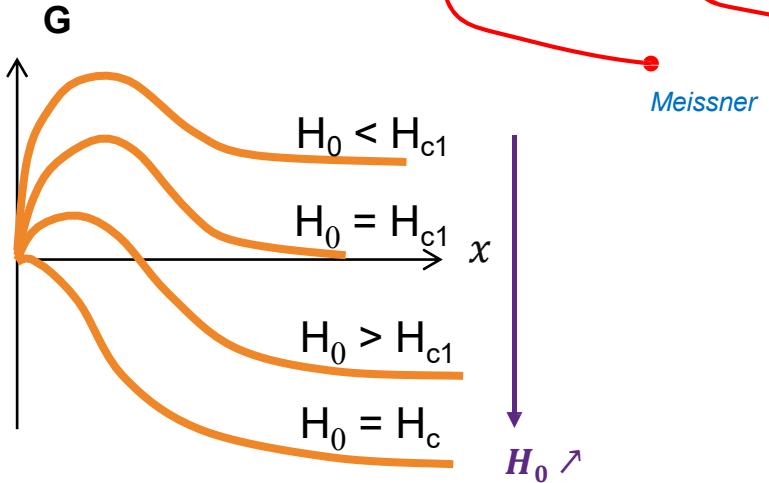
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SUPERCONDUCTORS FOR SRF ?

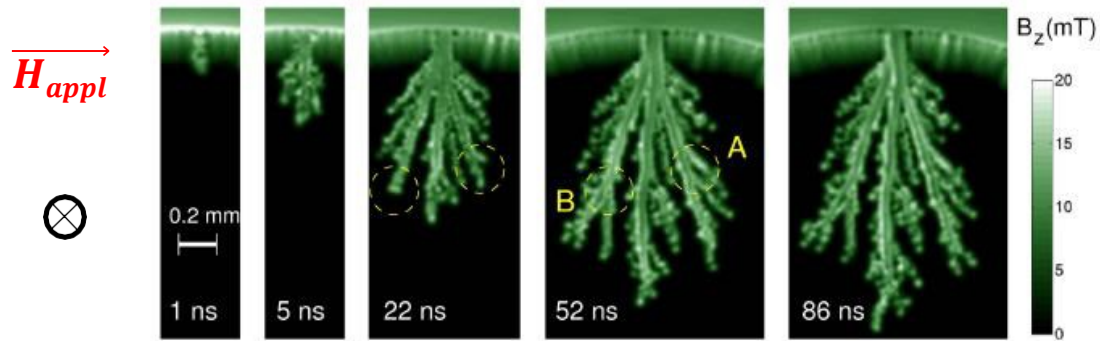
Material	T_c (K)	ρ_n (μOcm)	$\mu_0 H_{C1}$ (mT)*	$\mu_0 H_{C2}$ (mT)*	$\mu_0 H_c$ (mT)*	$\mu_0 H_{SH}$ (mT)*	λ (nm)*	ξ (nm)*	Δ (meV)	Type
Pb	7.1		n.a.	n.a.	80		48			I
Nb	9.23	2	170	400	200	219	40	28	1.5	II
NbN	17.1	70	20	15 000	230	214	200-350	<5	2.6	II
NbTi			4-13	>11 000	100-200	80-160	210-420	5.4		
NbTiN	17.3	35	30	15 000			150-200	<5	2.8	II
Nb₃Sn	18.3	20	50	30 000	540	425	80-100	<5	<5	II
Mo ₃ Re	15	10-30	30	3 500	430	170	140			II
MgB₂	39	0.1-10	30	3 500	430	170	140	5	2.3/7.2	II- 2gaps**
2H-NbSe ₂	7.1	68	13	2680- 15000	120	95	100-160	8-10		II- 2gaps**
YBCO/Cuprates	93		10	100 000	1400	1050	150	0.03/2		d-wave**
Pnictides Ba_{0.6}K_{0.4}Fe₂As₂	38		30	>50000	900	756	200	2	10-20	s/d wave**

* @ 0K

** 2D => orientation problems ?

WHAT IS THE ACTUAL LIMIT ($H_{fp}/H_{C1}/H_{SH}$) ?

❑ Avalanche penetration/flux jumps

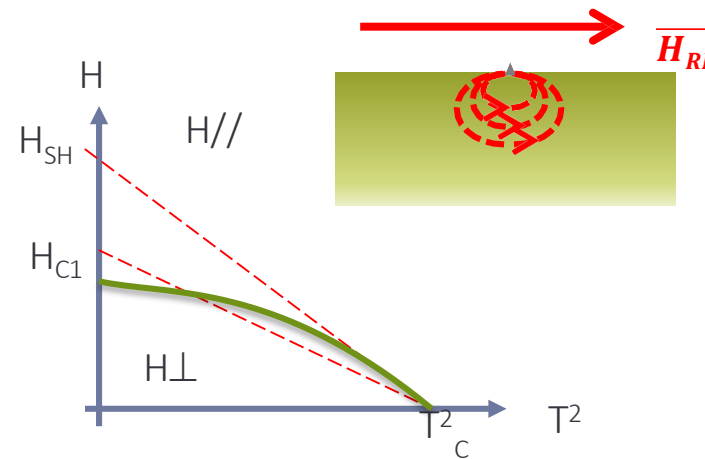
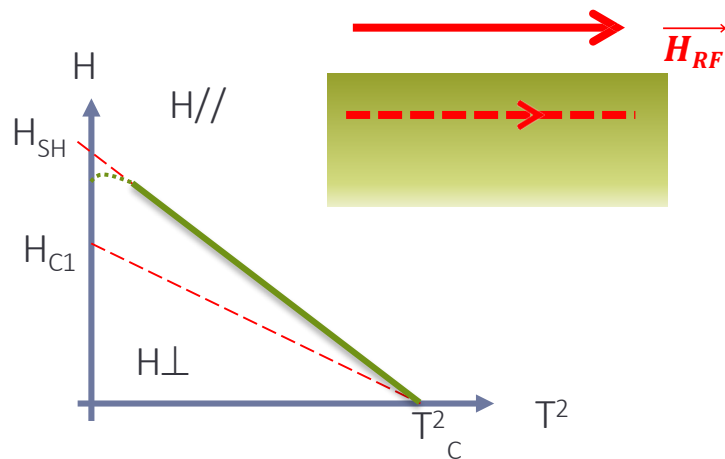


- ❑ $\sim 100 \mu\text{m}$ in 1 ns ($\sim RF$ period)
- ❑ Compare with λ field penetration depth
 - Nb : $\sim 40 \text{ nm}$
 - $\text{MgB}_2 \sim 200 \text{ nm}$
- ❑ Avalanche : high RF dissipation

MgB₂: http://www.nature.com/srep/2012/121126/srep00886/full/srep00886.html?message-global=remove&WT.ec_id=SREP-20121127

❑ In real world, cavities behavior is dominated by a few number of defects

It is very important to measure the penetration field of samples in realistic conditions



EFFECTS OF LOCAL DEFECTS

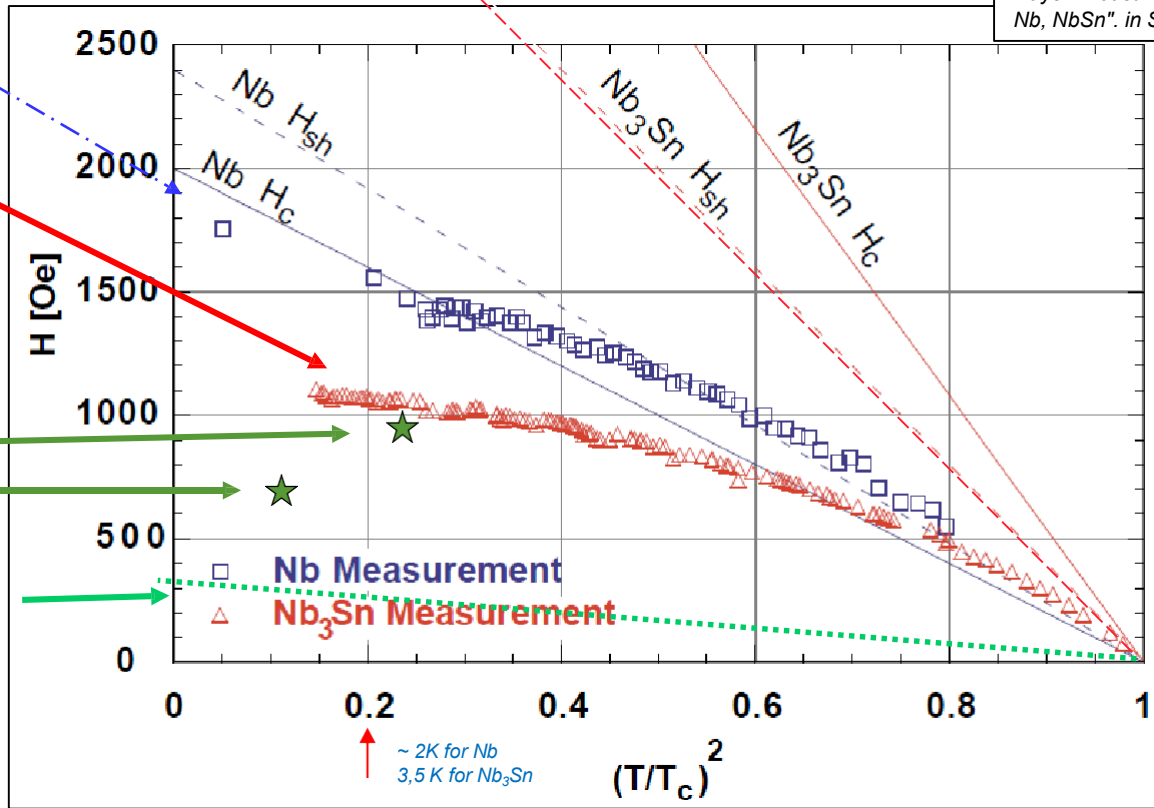
$H_{SH}^{Nb_3Sn}$
(~ 400 mT @ 0 K)

Nb H_{C1}
(170 to 200 mT)

Cornell, 1997
pulsed

Recent results from
Cornell & FNAL CW
Posen,
2019 (23 MV/m)
2015 (17 MV/m)

$H_{C1}^{Nb_3Sn}$ (~27mT)
(50 mT pour le bulk)



Hays. "Measuring the RF critical field of Pb, Nb, NbSn". in SRF 97. 1997.

Nb_3Sn
vs Nb

$$= H_i^0 \left[1 - \left(\frac{T}{T_c} \right)^2 \right]^2$$

Dissipations :

$$\frac{1}{2} \iint R_s H^2 dS$$

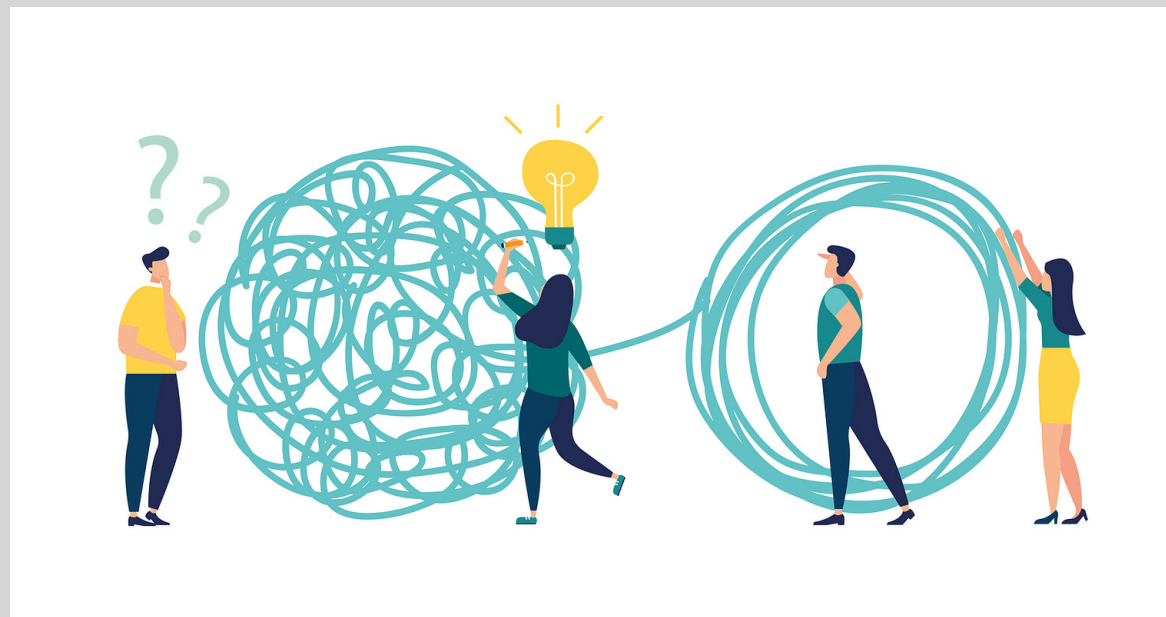
Vortices enter more easily at lower temperature (counter intuitive !)?

- @ $T \sim T_c$: H is low => low dissipations => easy to thermally stabilize
- @ $T \ll T_c$: H is high => even if small defect => high dissipations => Favors flux jumps

=> We have to reduce defect density

(yes but which ones?)

CHALLENGES TO FACE ON THE ROUTE TOWARDS OTHER SUPERCONDUCTORS: GENERALITIES



GENERAL ISSUES WITH SCs

Needed: high T_C , high H_{SH} (by default high H_{C1})

Advantages of niobium: pure metal.

- Highest T_C of metallic SC, H_{C1}
- Easy to form
- Uniform composition, *no phase transition in the domain of interest*
- Very large ξ : makes it less sensitive to small crystalline defects (e.g. GB)

Issues with alloyed, metallic SC compounds (e.g. NbTi)

- Higher T_C s, but smaller H_{C1} , ξ
- Still relatively easy to form (harder)
- Usually several phases, not all of them SC
- Risk of non homogeneity

Issues with non metallic SC compounds

- Higher T_C s, but smaller H_{C1} , ξ
- Brittle, no forming is possible, only films (*OK for SRF, but a more complex fabrication route is needed*)
- Usually several phases, not all of them SC
- Risk of non homogeneity

Sometimes local disorder =>

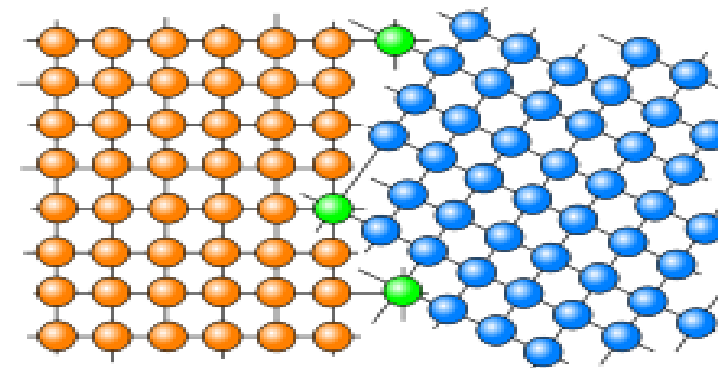
- **≠ local composition**, possibly non SC
- **Weak links** e.g. NC grain boundaries = main reason why HTC do not apply in SRF.

Metallurgy : started
~ 5000 Ys ago

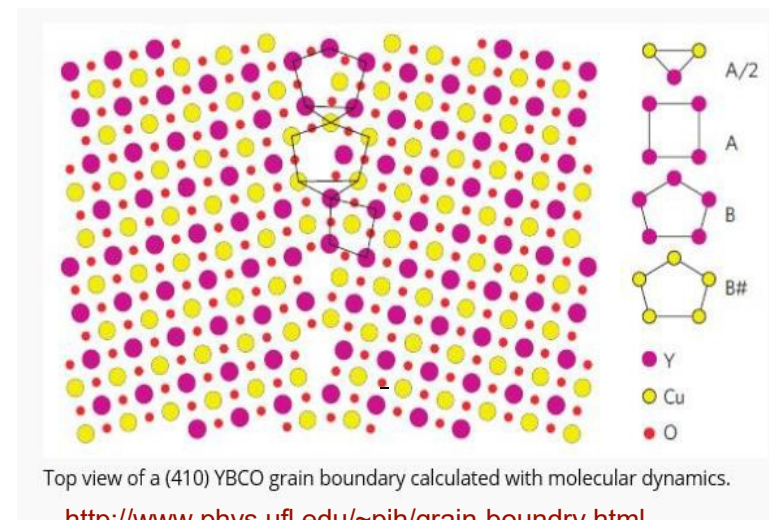
Chemistry : started
~ 200 Ys ago
Thin films techno.
< 100Y

If you are a theoretician you prefer to talk about the "existence of nodes in the gap of d-wave superconductors" : both are related to Brillouin structure

EX. : Grain boundaries



Some nm ← → Compare with ξ



Top view of a (410) YBCO grain boundary calculated with molecular dynamics.

<http://www.phys.ufl.edu/~pjh/grain-boundary.html>

THIN FILMS DEPOSITION



Nb : $\lambda \sim 50 \text{ nm}$ => **only a few 100s nm of SC necessary** (the remaining thickness= mechanical support) => **Make thin films !**

□ Advantages

- Thermal stability (*substrate cavity = copper, Aluminum, ... W*)
- Cost
- Opens route to innovative materials
- Optimization of R_{BCS} possible (*e.g. by playing with m.f.p*)

□ Disadvantages

- Fabrication and surface preparation of substrate (*at least*) as difficult as for bulk Nb
- Steep Q_0 decrease often observed by increase of RF field (*sputtered niobium films, improved lately*)
- Deposition of innovative materials is very difficult (*large parameters space to be explored*)
- Most of the known SC have been optimized for wire applications (*low H_{c1} , defects, pinning centers...*) => **most of the literature recipes are not fitted for SRF application** ☹ ☹ ☹

DEPOSITION TECHNIQUES: 3 MAJORS FAMILIES

❑ Physical deposition techniques (PVD, MS, DS...)

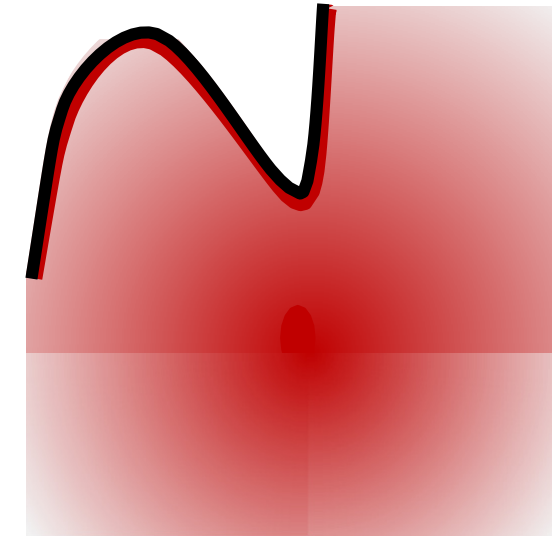
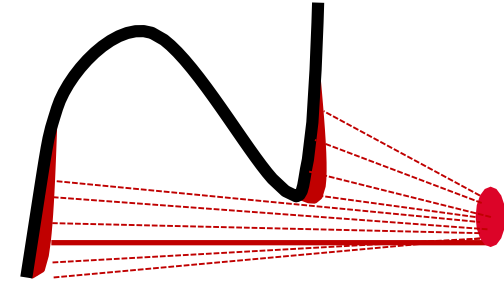
- line of sight techniques
- issues: getting uniform thickness/structure
internal stress and adhesion
- limited for complex geometry

❑ Thermal diffusion films

- limited compositions available
- non uniform composition issues (*S shaped diffusion front, differential diffusion rate with substrate grain orientation*)

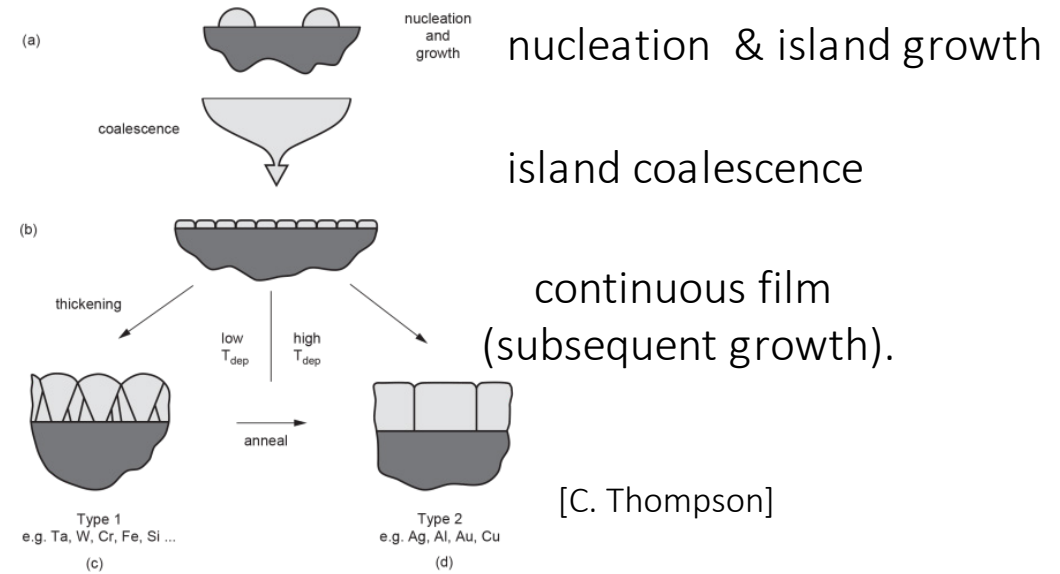
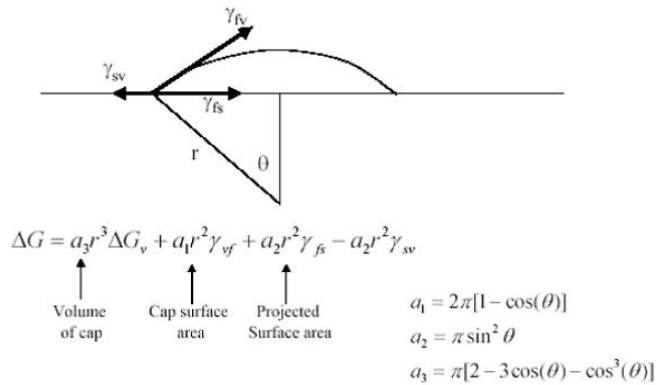
❑ Chemical techniques CVD, ALD

- conformational even in complex shape
- very quick for large surfaces
- issues: get the proper crystalline structure
- Required use of precursors introduces more impurities



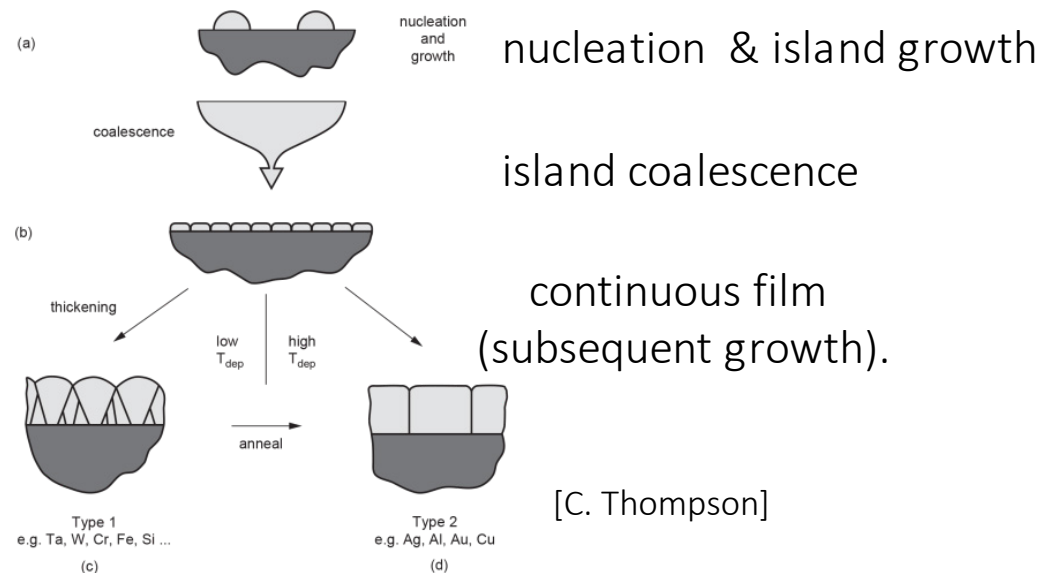
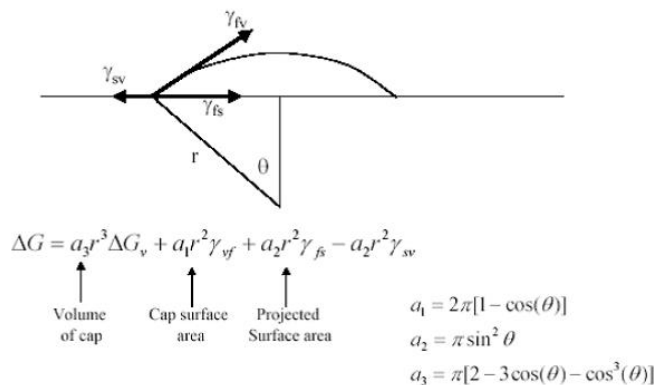
Thin Films Growth

Heterogeneous nucleation



Thin Films Growth

Heterogeneous nucleation



Control over the deposition process is exercised by only

3 first-order vapor parameters & 1 first-order substrate parameter

Vapor parameters

Absolute arrival rates of film atoms

Partial pressures of background gases in the chamber

Energies of the deposition fluxes.

Substrate parameter

Substrate temperature T.

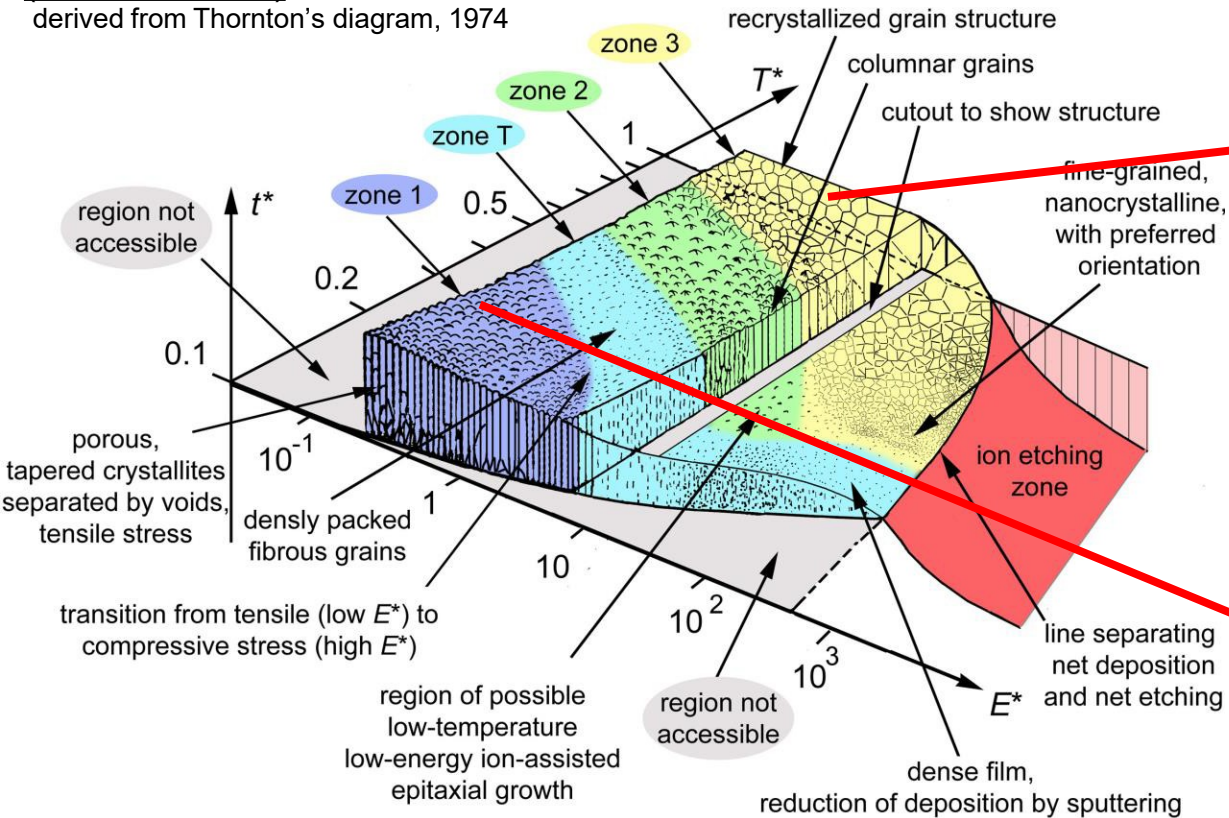
Without energetic atoms, only the substrate temperature influences the processes of physi- and chemisorption, thermal desorption, nucleation, nuclei dissociation, surface diffusion, and formation of specific nucleation sites.

SEARCH FOR BETTER STRUCTURE - Energetic Condensation

Condensing (film-forming) species : hyper-thermal & low energies (> 10 eV).

Generalized structure zone model (from A. Anders)

derived from Thornton's diagram, 1974



Energetic deposition

(HPIMS, CED, VAD...) Additional energy provided by fast particles arriving at a surface
=> Bulk like films

=> number of surface & sub-surface processes => changes in the film growth process:

- residual gases desorbed from the substrate surface
- chemical bonds may be broken and defects created thus affecting nucleation processes & film adhesion
- enhanced mobility of surface atoms
- stopping of arriving ions under the surface

Magnetron sputtering

=> A lot of defects
Cu limits annealing temperature
recrystallization

=> Changes & control in

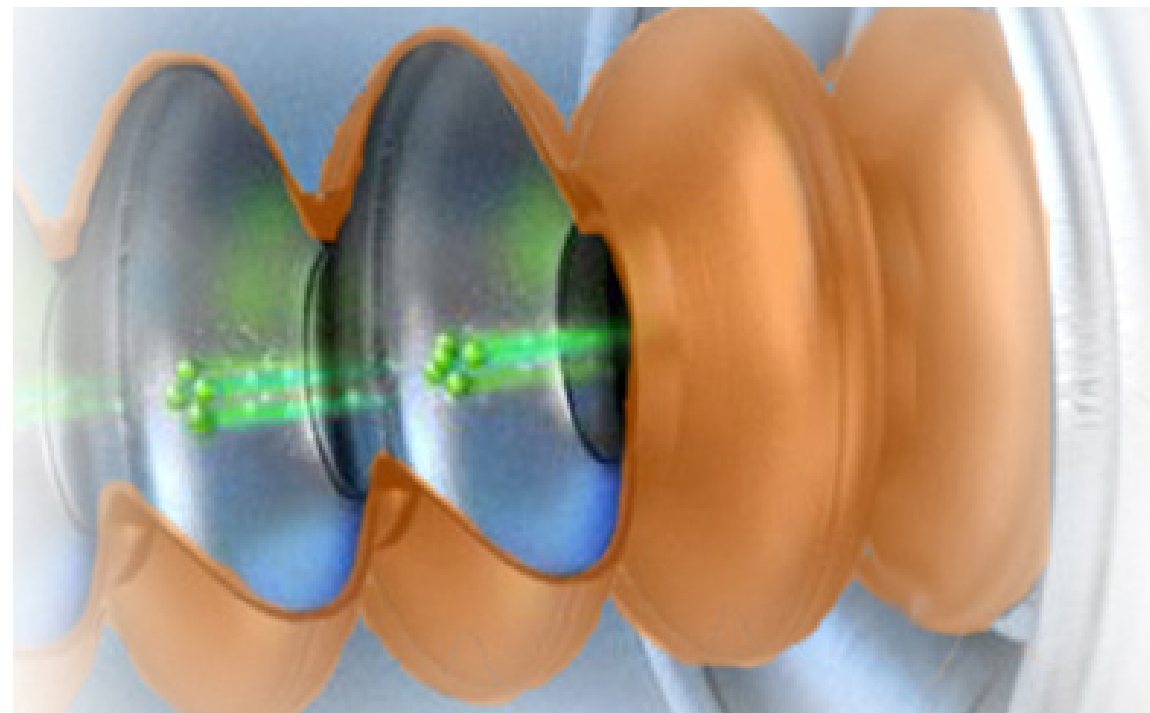
- Film density
- morphology
- microstructure
- Stress
- low-temperature epitaxy

A. Anders, Thin Solid Films 518, 4087 (2010).

11

Nb/Cu

The Jefferson Lab logo features a stylized grey graphic of a particle detector or accelerator structure. It consists of several interconnected circular nodes of varying sizes, connected by straight lines, all enclosed within a larger, faint circular outline.
Jefferson Lab



U.S. DEPARTMENT OF
ENERGY

Office of
Science

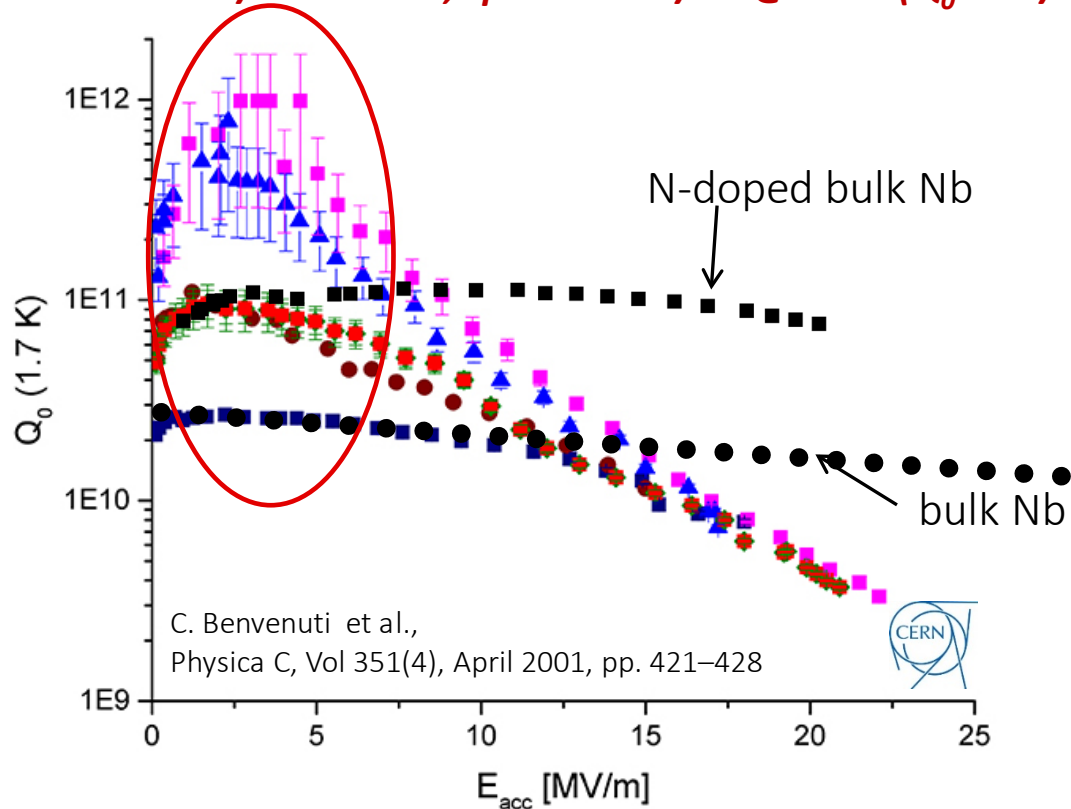


SPUTTERED Nb FILMS

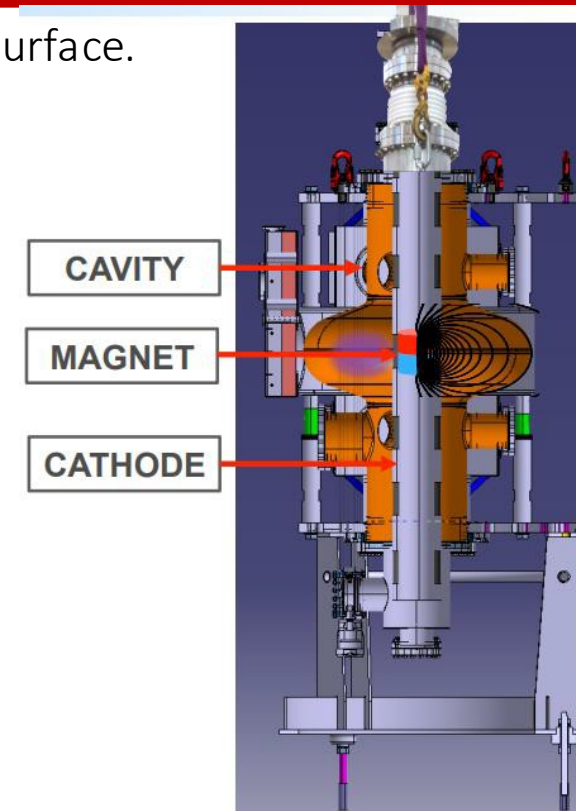
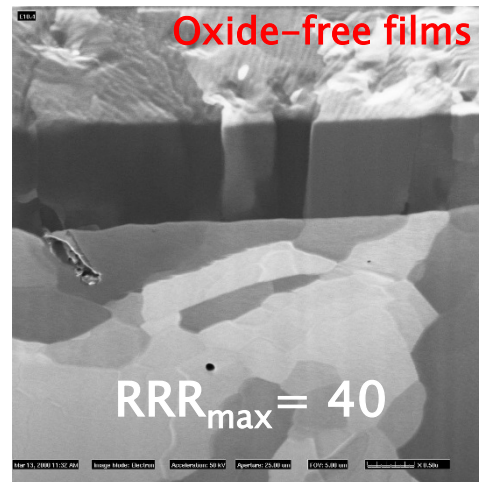
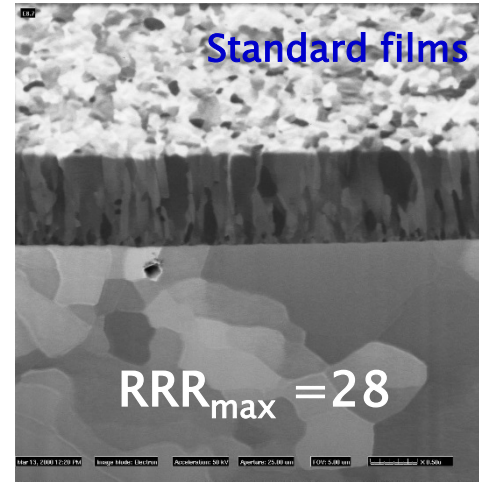
Thickness of interest for SRF applications = RF penetration depth, i.e. the very top 40 nm of the Nb surface.

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

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



High Q at low field BUT strong Q-slope



Possible origin of the slope

Depinning of trapped flux

Low H_{C1}

Early vortex penetration due to roughness

Current concentration due to porosities

(generating local electrical field)

Next-Generation Nb Films

Bulk-like performance Nb film

- ❑ Minimize R_{res} , maximize Q
- ❑ Potential major system simplifications
- ❑ Highest level of quality assurance and reliable performance.
- ❑ Use of substrates with higher thermal conductivity (Cu, Al)

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Bulk-like performance Nb film

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(not limited to Nb films alone, higher T_c films & multi layers also affected, especially with such issues as the entry of Josephson vortices driven by the RF field.)
focused on the entire problem and all possible causes in order to understand, identify, and eliminate the causes.

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What are the differences between bulk and thin-film Nb in reference to the RF surface?

Higher density of grain boundaries

Different spectrum of grain boundary energies than for bulk Nb surfaces

Thermal diffusivity of the RF surface (from the thermal properties of the film itself in addition to the thermal impedance of the Nb/ substrate interface).

Surface chemistry

Presence of defects (dislocations, porosities, inclusions)

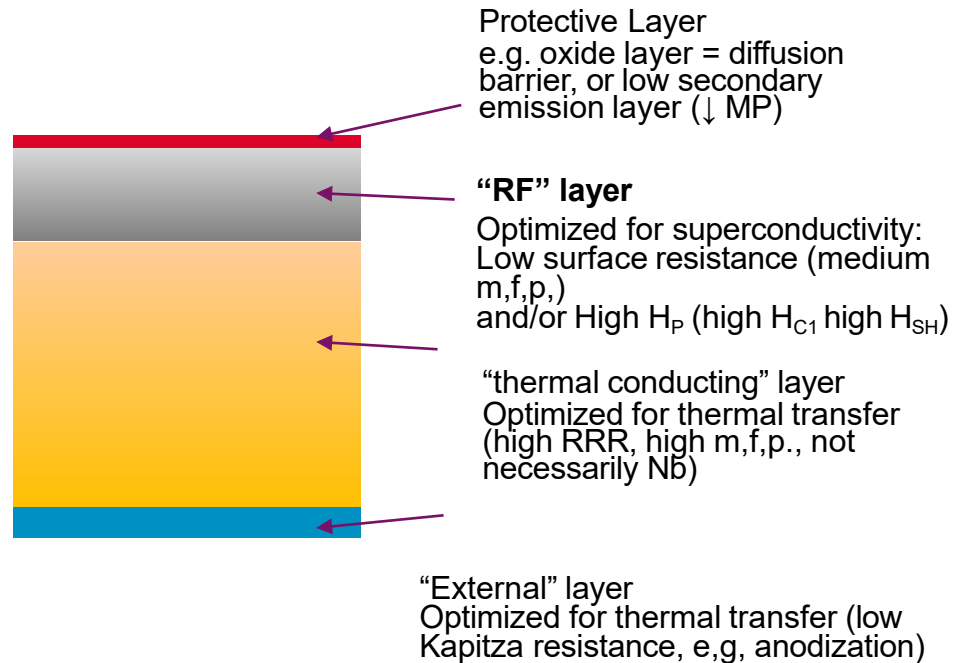
Surface morphology.

Magnetic field losses- several sources: field enhancing surface features, external stray fields captured in form of vortices or arising from thermoelectric currents as Nb transitions from normal to superconducting state.

Crystallographic structure (orientation, grain size)

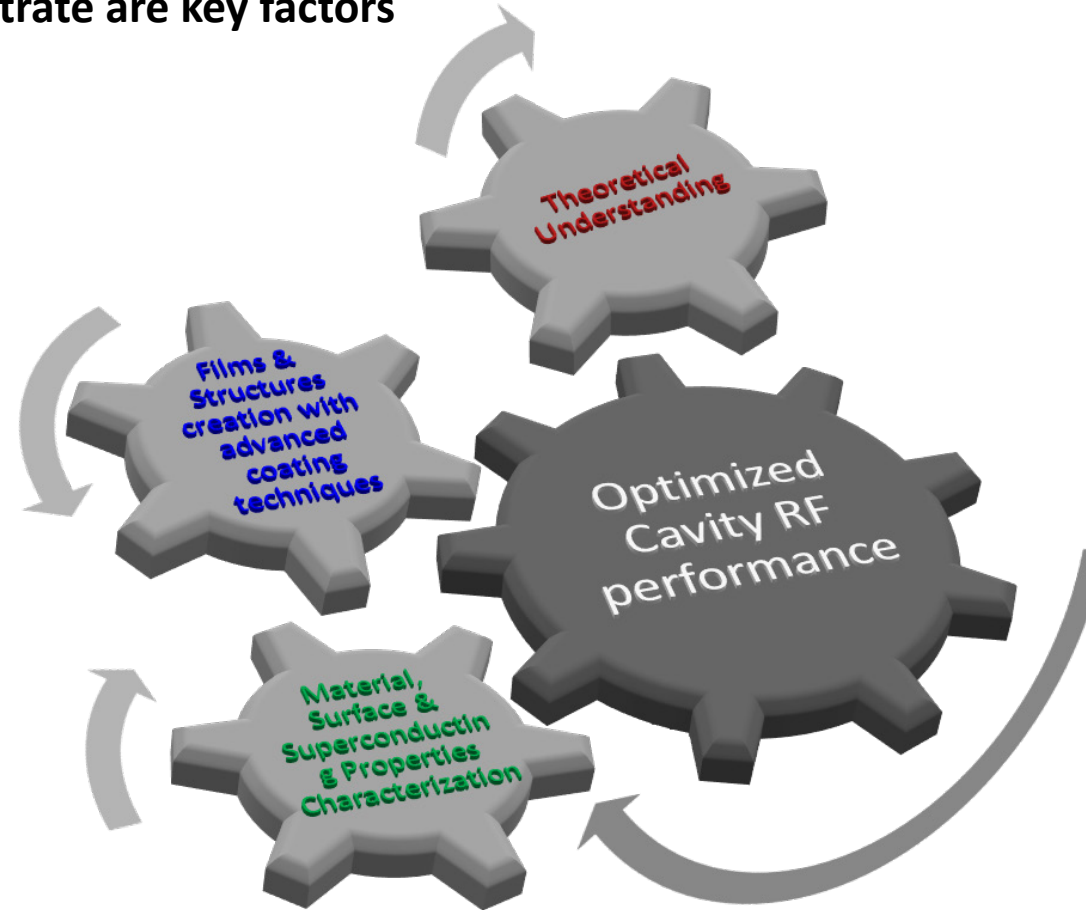
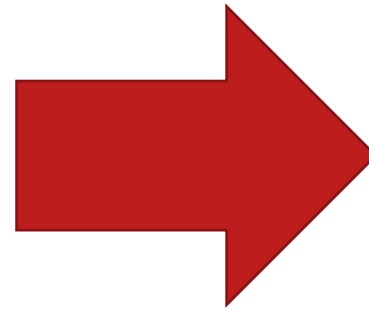
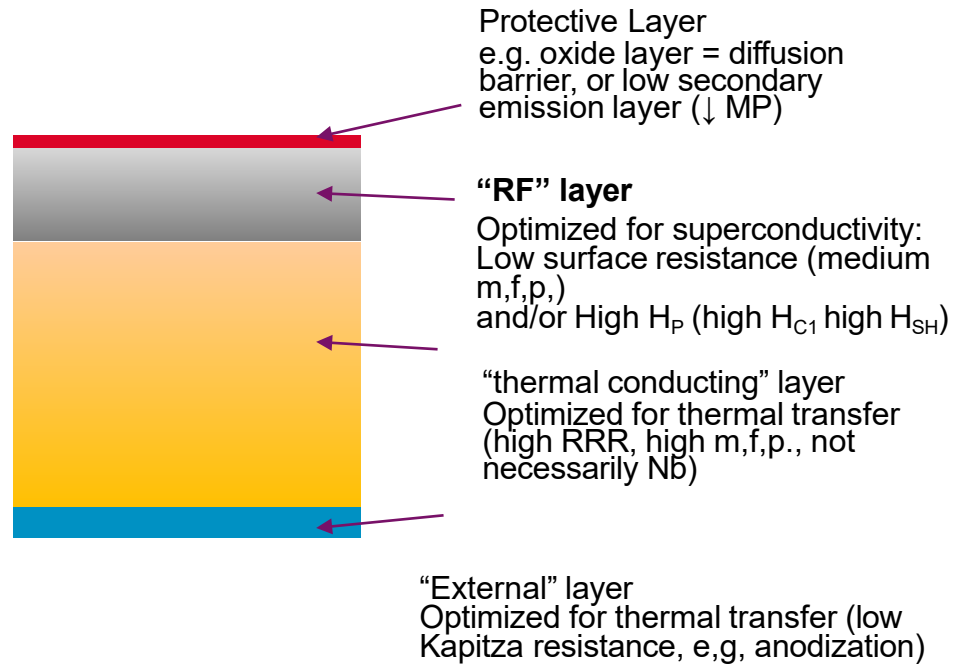
SRF Thin Films for Next-Generation Cavities

ALL film properties are a direct consequence of the film structure, defect/impurity content...
thus the technique, environment, substrate are key factors



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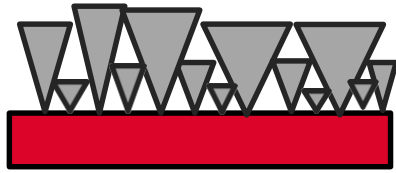
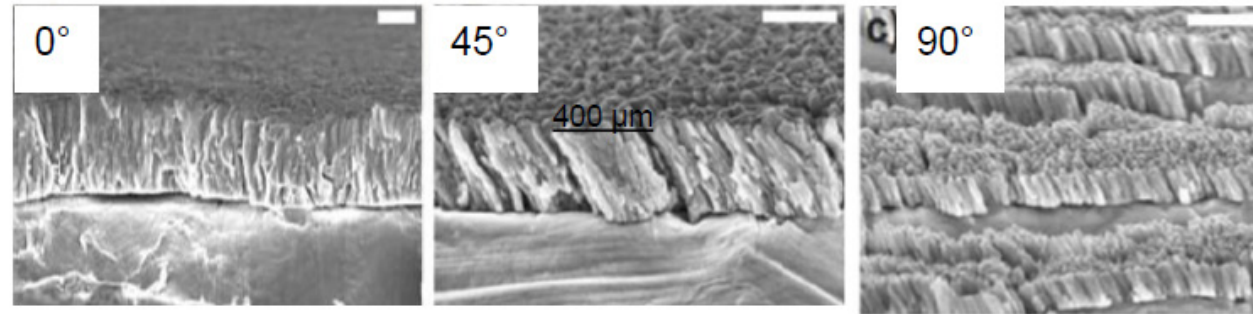


EXAMPLE OF QUALITY ISSUES OF FILMS

Magn. Sput. Nb

- Line of sight issues
=> porosities

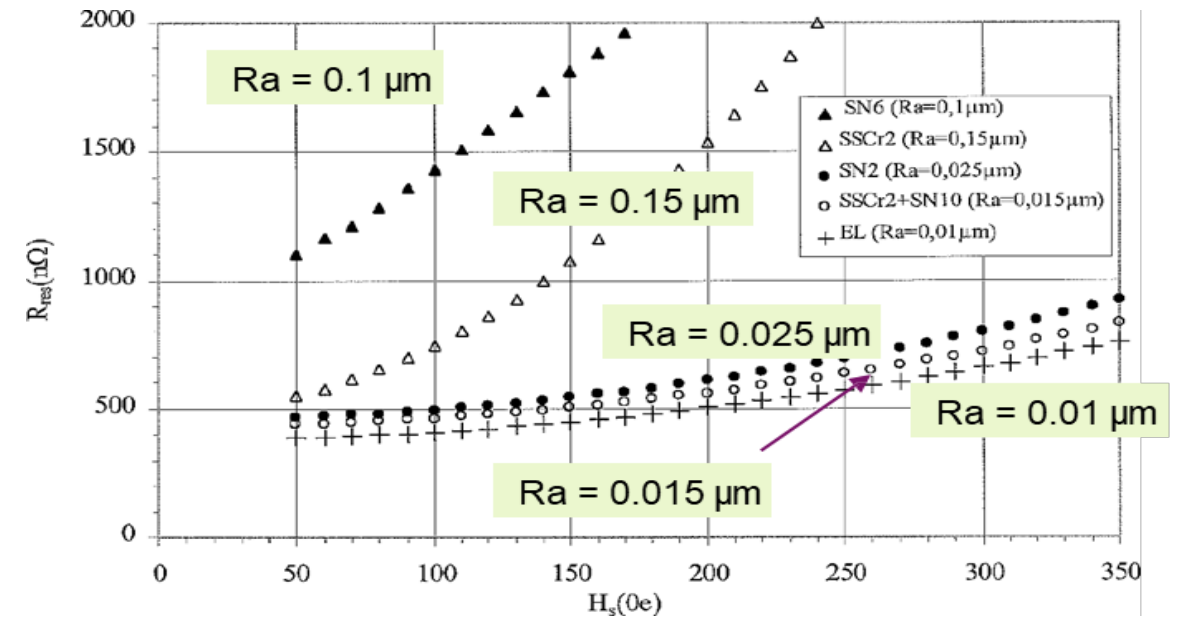
[G. Rosaz]



Inverted pyramid crystalline growth

- Internal stress
 - Advantage: higher T_c (up to a certain impurity concentration)
 - Disadvantage: adhesion issues (peel-off, delamination)
- High impurities content
 - Nb = getter material (nearly as good as Ti => high interstitial content)
 - Carrier gas incorporation (Ar)

- Sensitivity to Cu roughness (the smoother, the better)



[M. Ribeau, PhD]

SUBSTRATE ISSUES

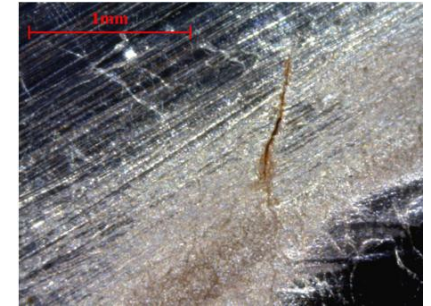
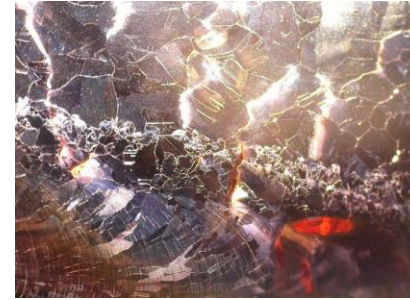
Cu and Nb not miscible (especially in presence of O)

Advantage: low interdiffusion

Disadvantage: adhesion issues (delamination)

Issues on Cu welding arrears

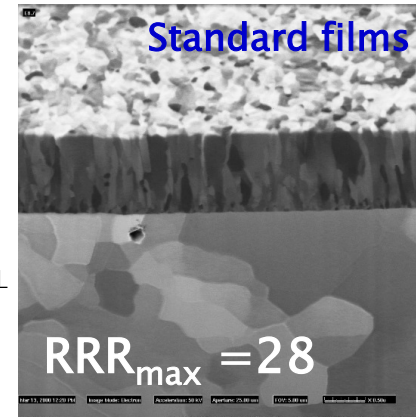
where



Best results are not always expected:

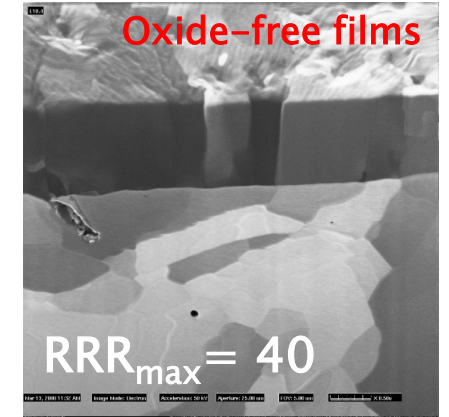
	Standard	Oxide-free
RRR	~10	~30
T_c (K)	9.51 ± 0.01	9.36 ± 0.04
Ar cont. (ppm)	435 ± 70	286 ± 43
Texture	(110)	(110), (211), (200)
	Fiber texture	Hetero-epitaxy
Grain size (μm)	0.1–0.2	1–5
$\lambda/\lambda_{\text{clean}}$	1.51 ± 0.04	1.04 ± 0.09
H_{c2} (T)	1.15 ± 0.025	0.77 ± 0.01
a_0 (Å)	3.3240(10)	3.3184(6)
Stress (Mpa)	-706 ± 56	-565 ± 78
Strain $\Delta a_{\perp}/a_{\perp}$ (%)	0.636 ± 0.096	0.466 ± 0.093

Columnar grains,
size ~ 100 nm
In plane diffraction
pattern: powder
diagram
(110) fiber texture \perp
substrate plane



Courtesy: P. Jacob – EMPA

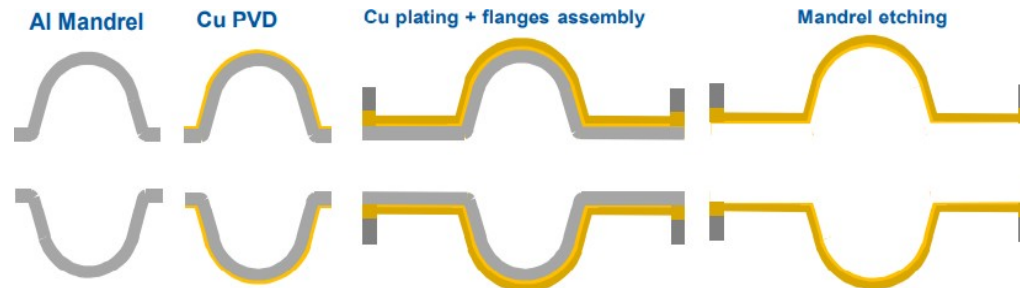
Equi-axed grains,
size ~ 1–5 μm
In plane diffraction
pattern: zone axis
[110]
Heteroepitaxy
Nb (110) //Cu(010)
, Nb (110)
//Cu(111), Nb (100)
//Cu(110)



❑ Bulk like films did not perform better initially ! (but recent changes !!!)

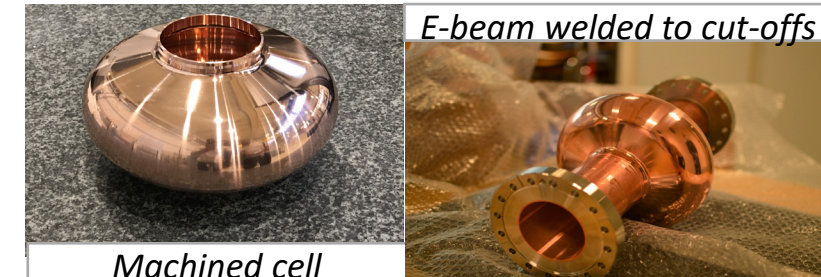
❑ New approaches:

- Bulk machining
- Electroplating
- Laser surfacing ...

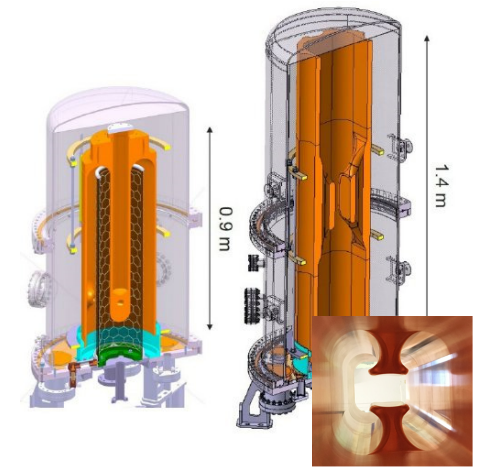
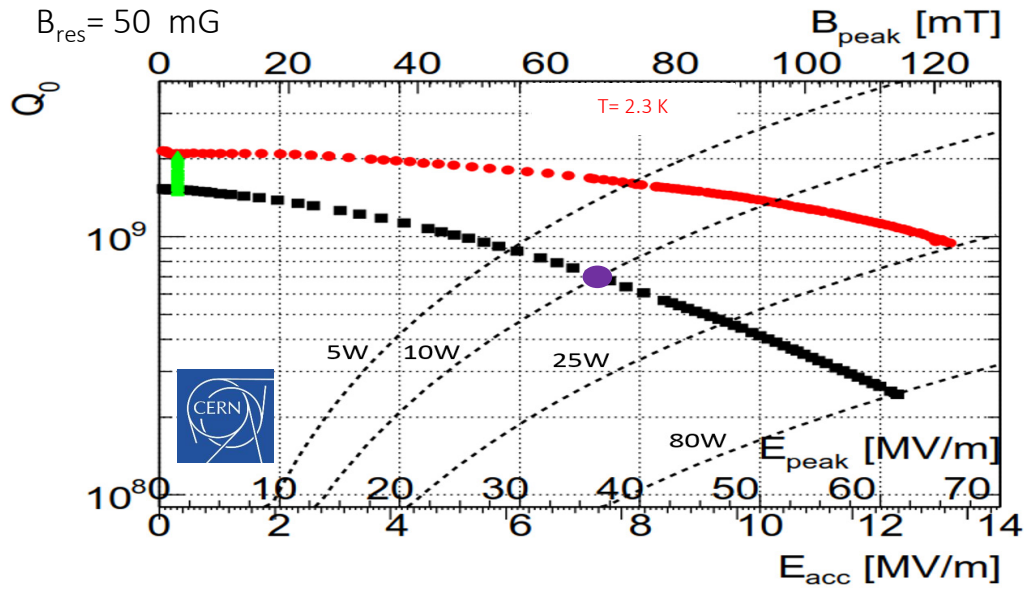
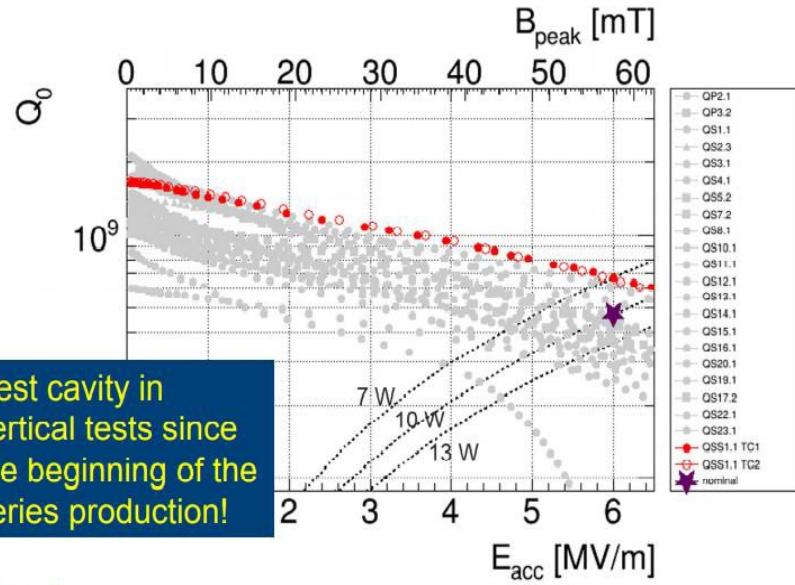


Srf Tutorials 2023 - Beyond Bulk Nb

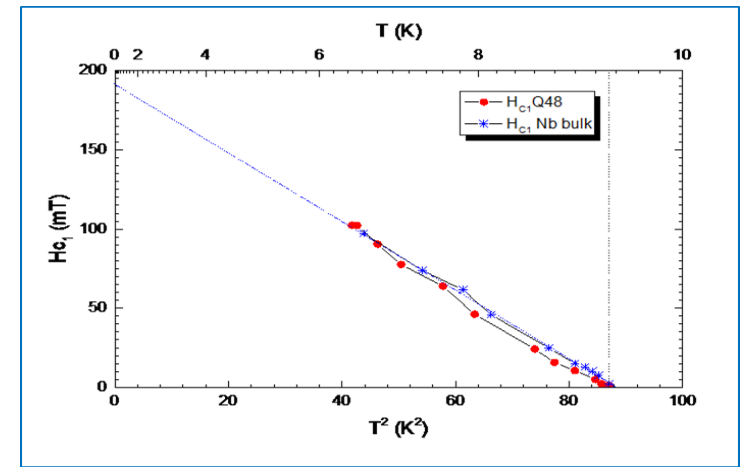
Courtesy: G. Rosaz, K. Scibor - CERN



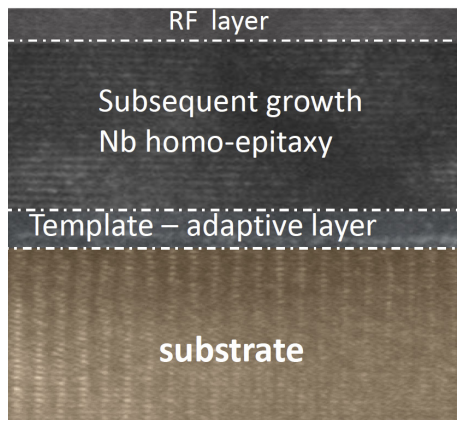
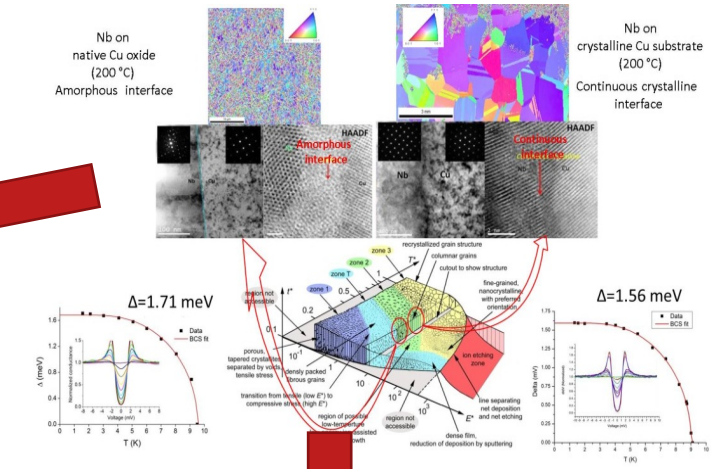
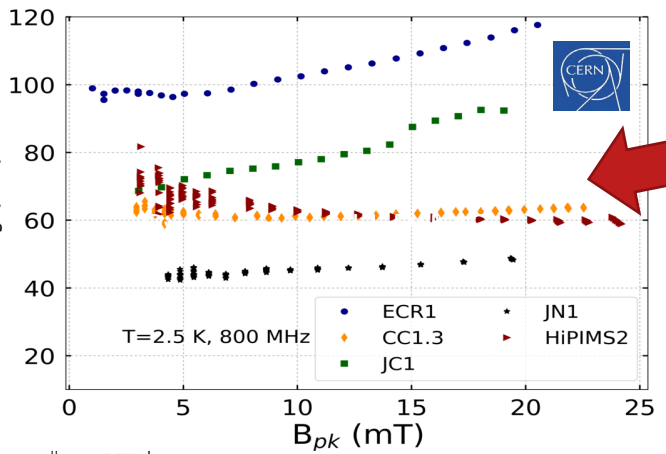
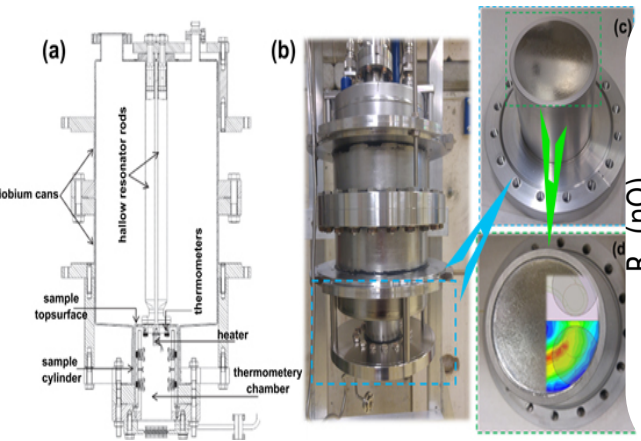
SEAMLESS HIE-ISOLDE CAVITY



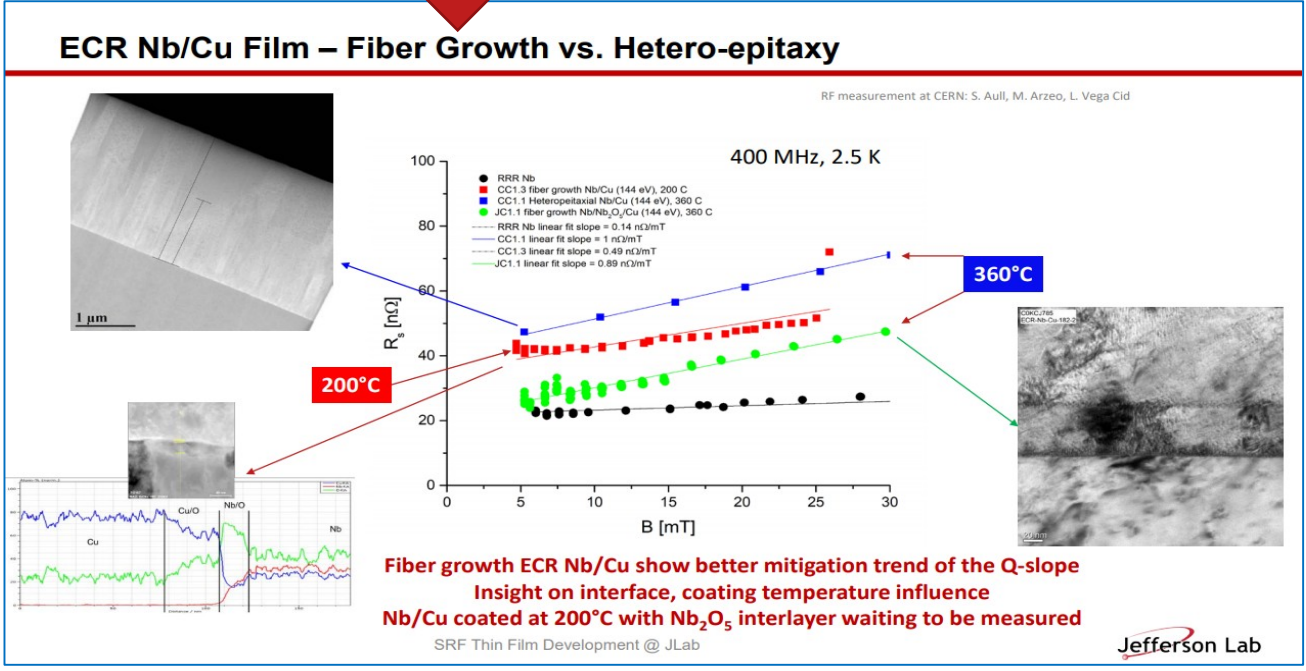
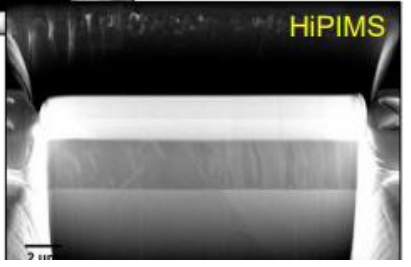
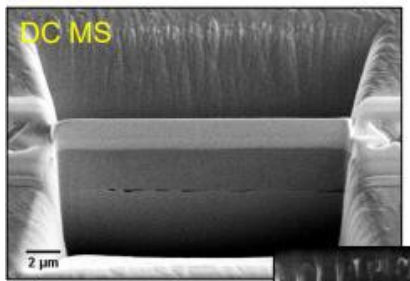
- Local magnetometry on a sample deposited in the same condition
- First evidence of a bulk-like behavior for a thin film !!!!



FILM DENSIFICATION /MITIGATION OF THE Q-SLOPE

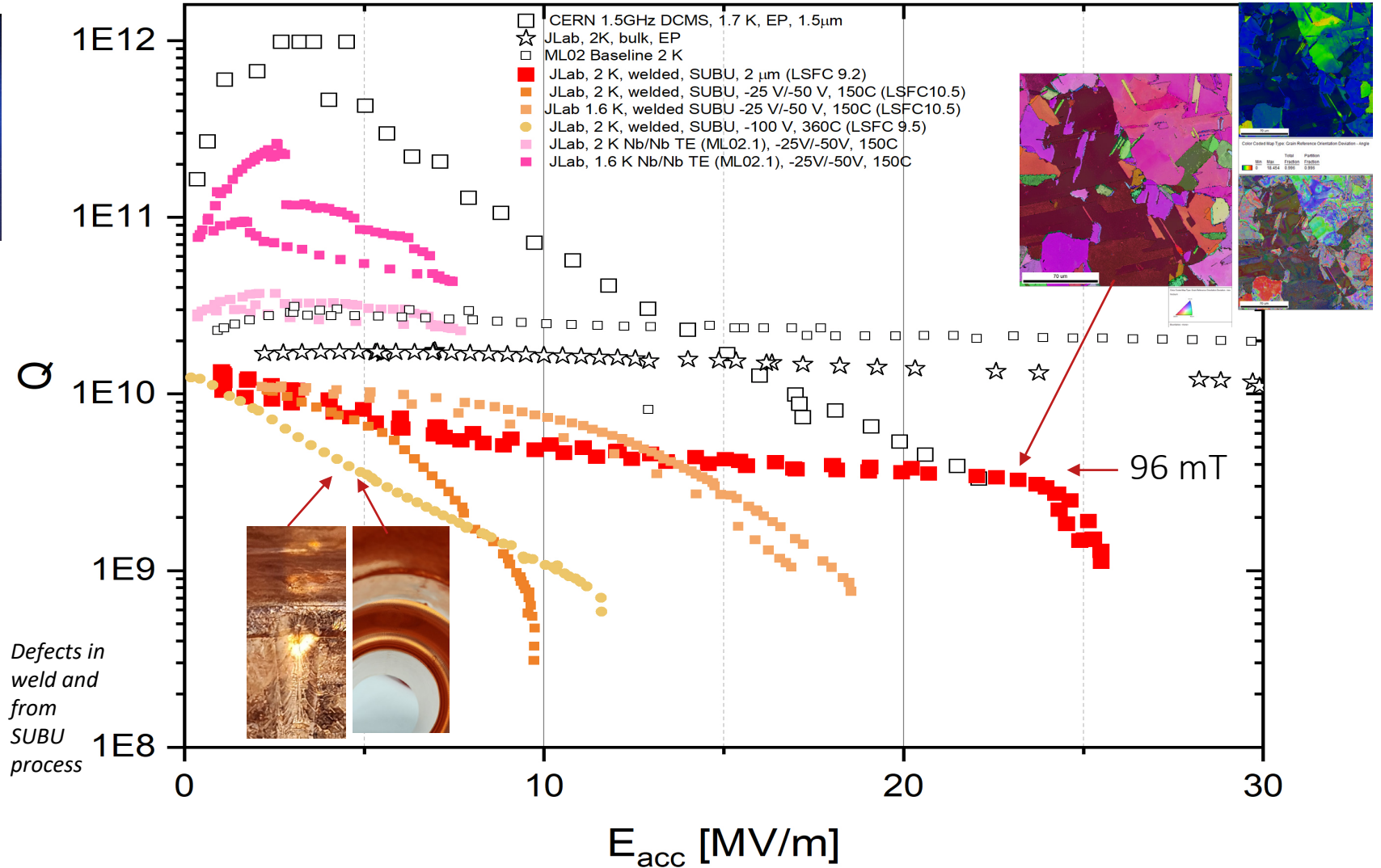
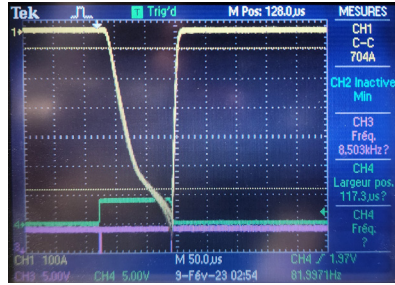
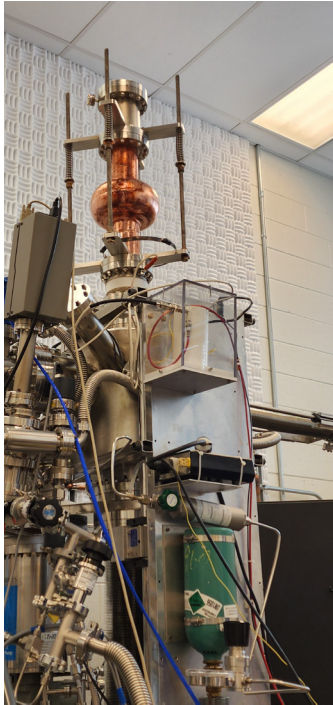


Sample	400 MHz		800 MHz		1200 MHz	
	R _{s0} (nΩ)	R _{s1} (nΩ/mT)	R _{s0} (nΩ)	R _{s1} (nΩ/mT)	R _{s0} (nΩ)	R _{s1} (nΩ/mT)
ECR	19.7	0.84	65.8	1.14	126.2	1.35
HiPIMS	19.8	0.11	n/a*	n/a*	100.9	0.3
bulk Nb	21.2	0.13	42.1	0.32	120.1	0.69



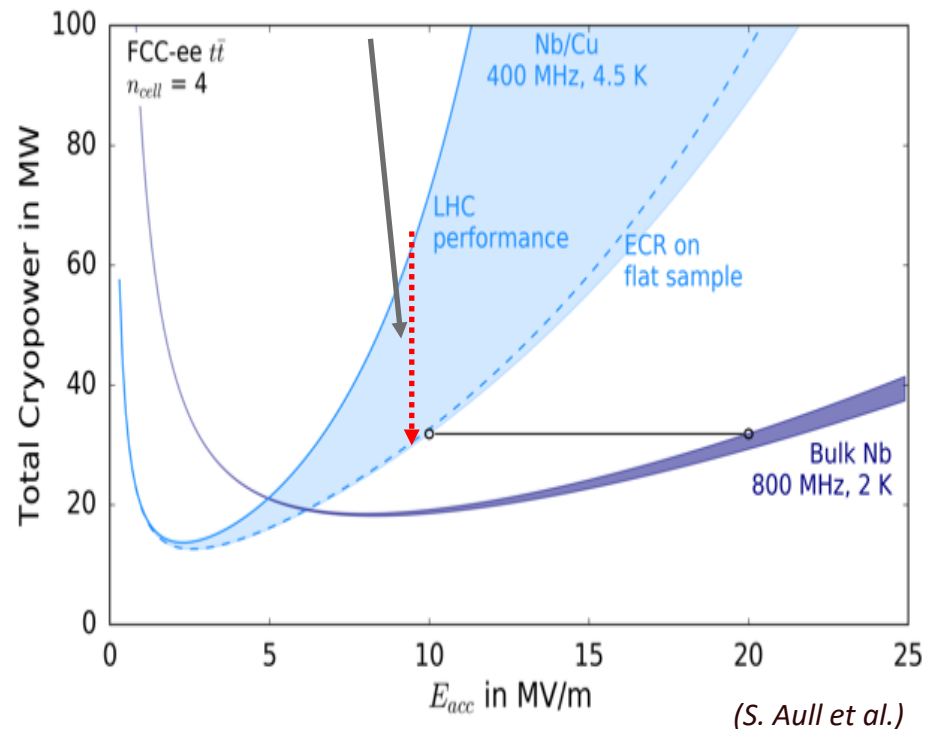
Development of Nb/Cu Cavities by Energetic Condensation

Development of Nb/Cu SRF Surfaces Development (HiPIMS & ECR)



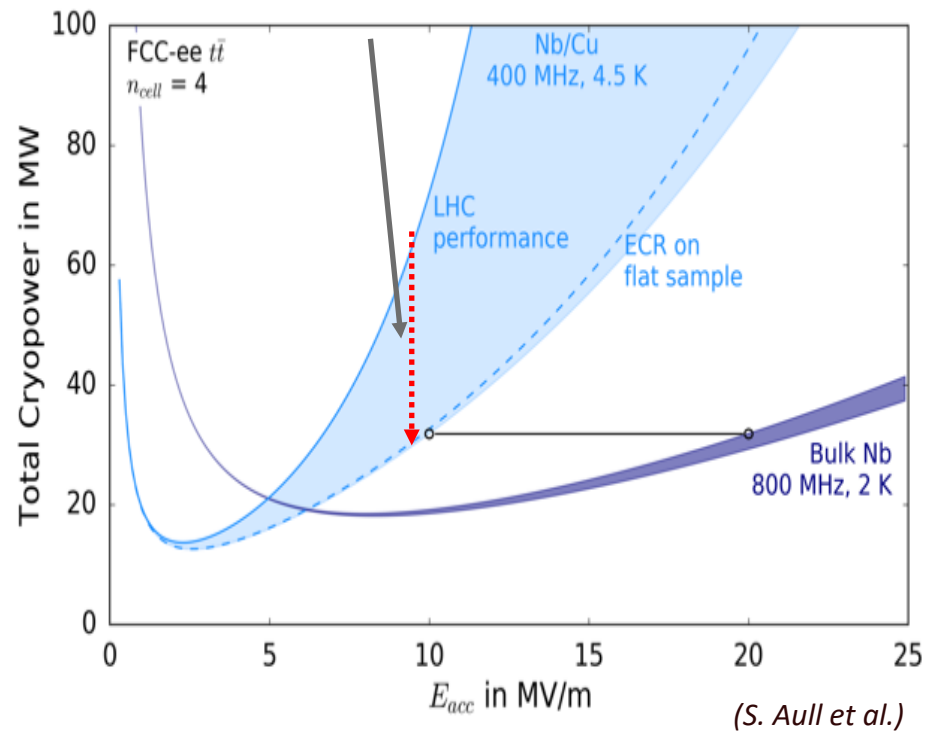
Tailored Nb films via energetic condensation

- ❑ **Tune thin film structure and quality** with ion energy and substrate temperature on a variety of substrates (amorphous, polycrystalline and single crystal)
- ❑ **Achieve film structures and properties only achievable at higher temperature** with classic coating methods
- ❑ **Tune RRR values** from single digits to **bulk Nb values** → No intrinsic limitations
- ❑ **Lower impurity (H) content** than bulk Nb
- ❑ **Good adhesion to the substrate** (delamination threshold : function of ion energy and temperature)
- ❑ **Tailoring interface** with high energy and subsequent growth at energy minimizing defect creation can **contribute to lower R_s and mitigate Q-slope.**



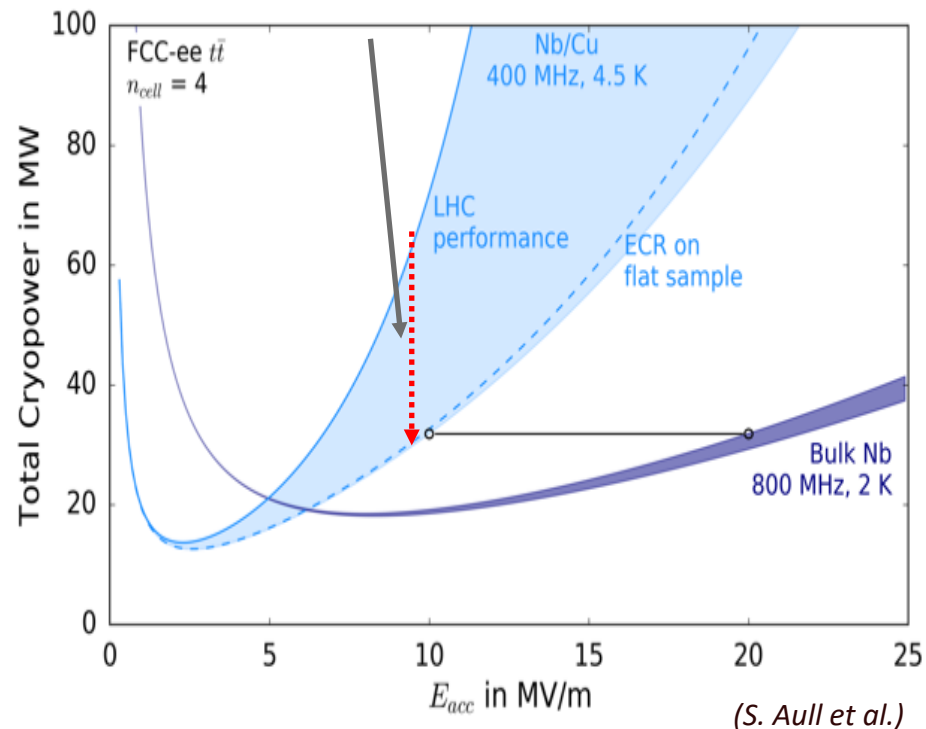
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- **Basic material processes exist**
 - proof-of-principle demonstrations for modest field applications
- **Principal challenges at present**
 - **Establish adequate process controls**
 - **Address technical challenges with scale-up**
(Though not fundamental, these require serious resource investment to establish “industrial” capability).
 - Resources drive the timeline.
- **Develop cavity coating & refine process parameters in parallel on smaller scales.**
- Done right, reasonable stepping stone to truly “engineered surface”, with all the benefits of high Q, high field, low cost, high reliability systems.

OTHER SUPERCONDUCTORS

 Jefferson Lab



U.S. DEPARTMENT OF
ENERGY

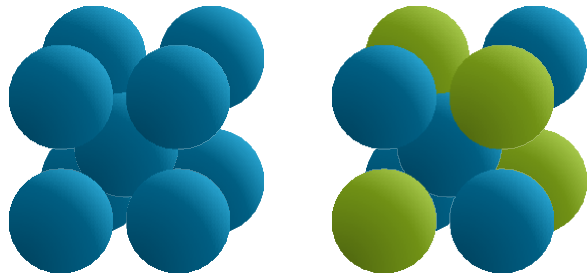
Office of
Science



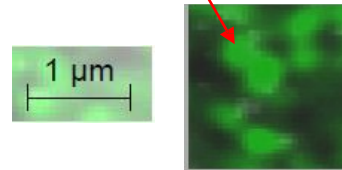
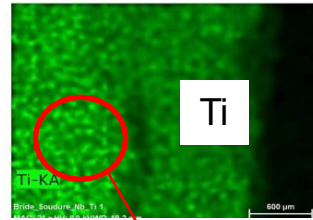
MOST COMMON SUPERCONDUCTORS

- **A2** (e.g. NbTi, Transition metal alloys, BCC structures)
- **B1** (e.g. NbN, NbTiN, Transition metal carbide or nitride, NaCl structures)
- **A15** (e.g. Nb₃Sn, Compounds, NaCl structures)
- **2-D SC** (Compounds, anisotropic)
 - MgB₂
 - Cuprates, Pnictides
 - (others TaS₂, organic...)
- **SPECIAL SRF: METAMATERIALS** (Multilayers)

A2 SC ALLOYS: e.g. NbTi



BCC pure metal and solid solution alloy

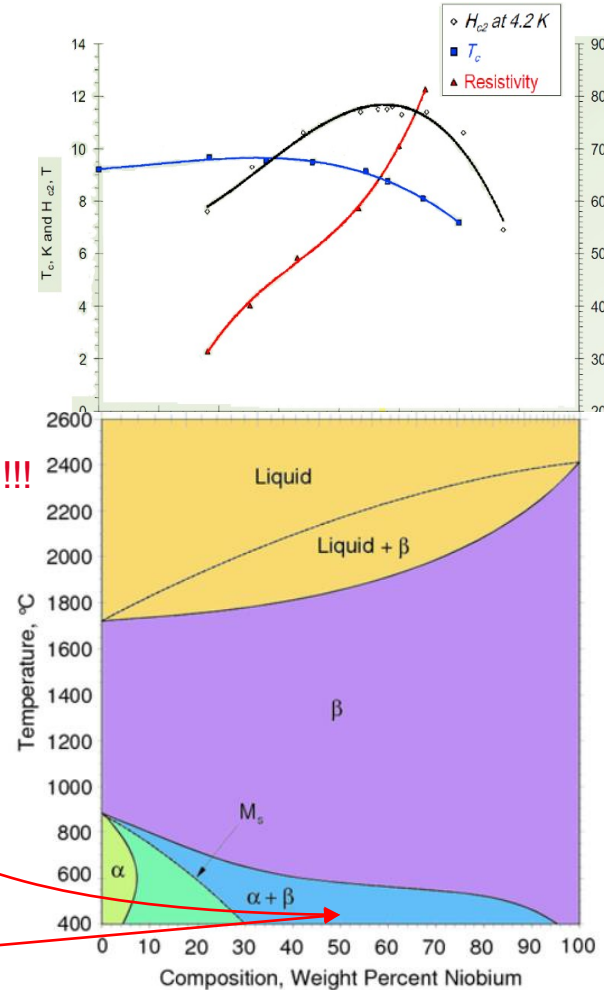


Ti precipitates ($\varnothing \sim 0,4 \mu\text{m}$)
NC Metal => RF dissipation !!!!

- NbTi widely used in coils
- Available alloys range around 45-55 % Ti
- Ti is not fully miscible inside Nb (Ti precipitates at low T when [Ti] > 5 W%)

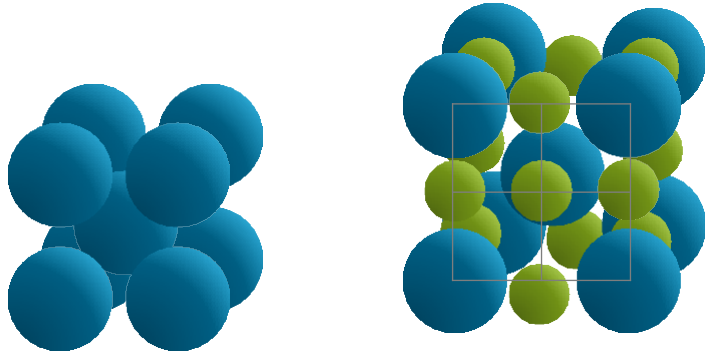
=> no RF !!!

$Ti \varnothing = Nb \varnothing$
Ti precipitates in a niobium matrix (with a few Nb replaced by substitutional Ti) => ~ same T_C , same H_C as Nb, but not same ℓ => high K



<http://www.dierk-raabe.com/titanium-alloys/biomedical-titanium-alloys/>

B1 SC COMPOUNDS: e.g. NbN, NbTiN



BCC pure metal + smaller atoms (N, C) in interstitial location => NaCl structure

NbN cubic phase : $T_C \sim 17-18$ K

NbTiN stabilization of cubic (SC) phase NbN

not too sensitive to local variation
of composition !

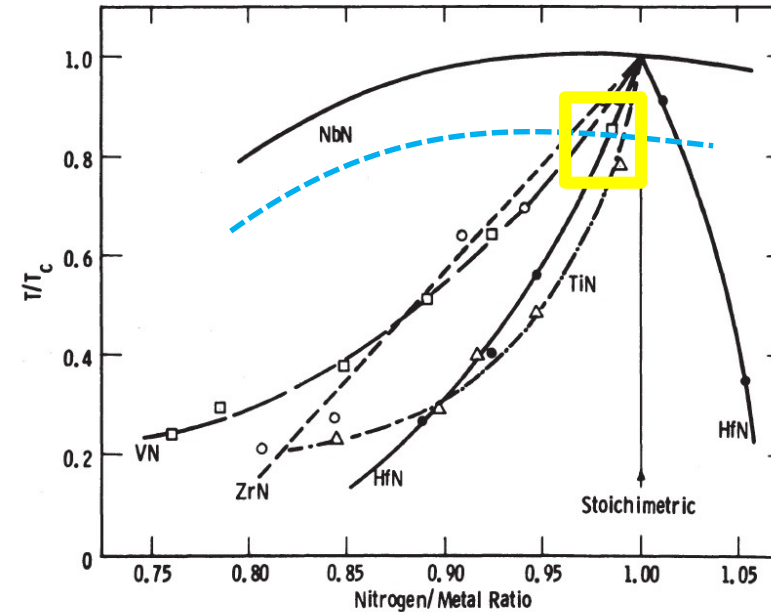
Solid solution => relatively easy fabrication (*thermal diffusion*, . 20
reactive sputtering...)

Good model SC

Widely used for JJ and SC electronics

Also a material of choice for the
development of multilayers (see below)

<https://link.springer.com/content/pdf/10.1007%2F978-1-4757-0037-4.pdf>



Critical temperature versus nitrogen-to-metal ratio for various B1-structure nitrides of the transition metals (data assembled by Hulm and Blaugher).

NbTiN RF Results

INFN : reactive sputtering with Ar/N₂ in DC Triode Magnetron Sputtering @ 600°C and 200°C

(Nb_{1-x}Ti_x)N films with 1-x < 0.5 present a lower calculated surface impedance, lower critical fields and better surface properties than NbN, especially when deposited at low temperatures.

R. Di Leo et al. J. of Low Temp. Phys, vol 78, n1/2, pp41-50, 1990

Reactive Magnetron Sputtering:

CEA Saclay :

NbTiN films deposited on 12 cm copper disks by magnetron sputtering and tested in a cylindrical TE₀₁₁ cavity reached RF field levels of 35 mT
 low residual surface resistance (< 100 nΩ at 4 GHz) with a very small BCS resistance
 4 cavities deposited but no RF measurement due to film blistering on large area of the cavity.
 Rs slope significantly decreased when coating with bias ranging from -50V to -100V

CERN:

Samples and six 1.5 GHz Cu cavities coated by reactive cylindrical magnetron sputtering

Best cavity result for thicker film (4.3μm) and lower deposition temperature (265°C)

Rs = 330nΩ @ 4.2K

M. Marino, Proceedings of the 8th Workshop on RF Superconductivity, October 1997, Abano Terme (Padua), (Rep) 133/98, vol.IV, p.1076

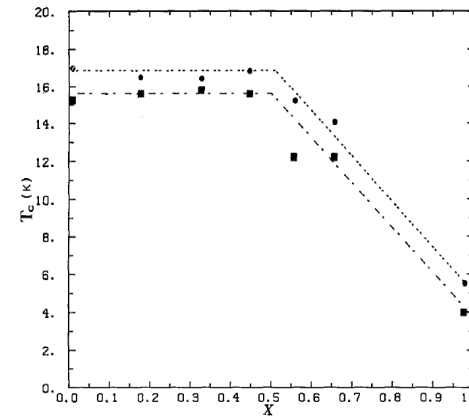


Fig. 1. Superconducting critical temperature T_c as a function of the titanium composition (x) for the (Nb_{1-x}Ti_x)N films deposited at $T_d = 600^\circ\text{C}$ (circles) and at $T_d = 200^\circ\text{C}$ (squares).

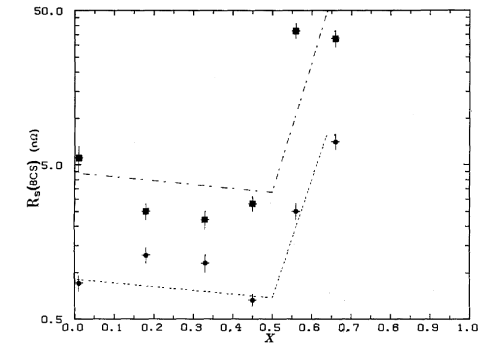


Fig. 3. Calculated BCS surface impedance $R_s(\text{BCS})$ as a function of the titanium composition (x) for the (Nb_{1-x}Ti_x)N films deposited at $T_d = 600^\circ\text{C}$ (circles) and at $T_d = 200^\circ\text{C}$ (squares). The continuous lines correspond to the values of the lines through the data in Figs. 1 and 2.

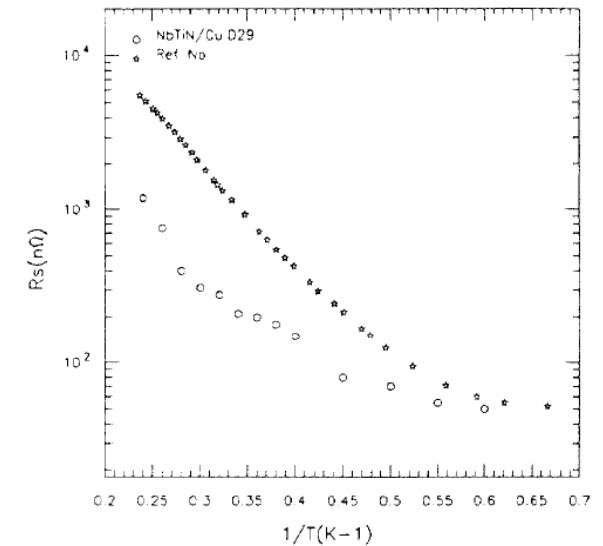
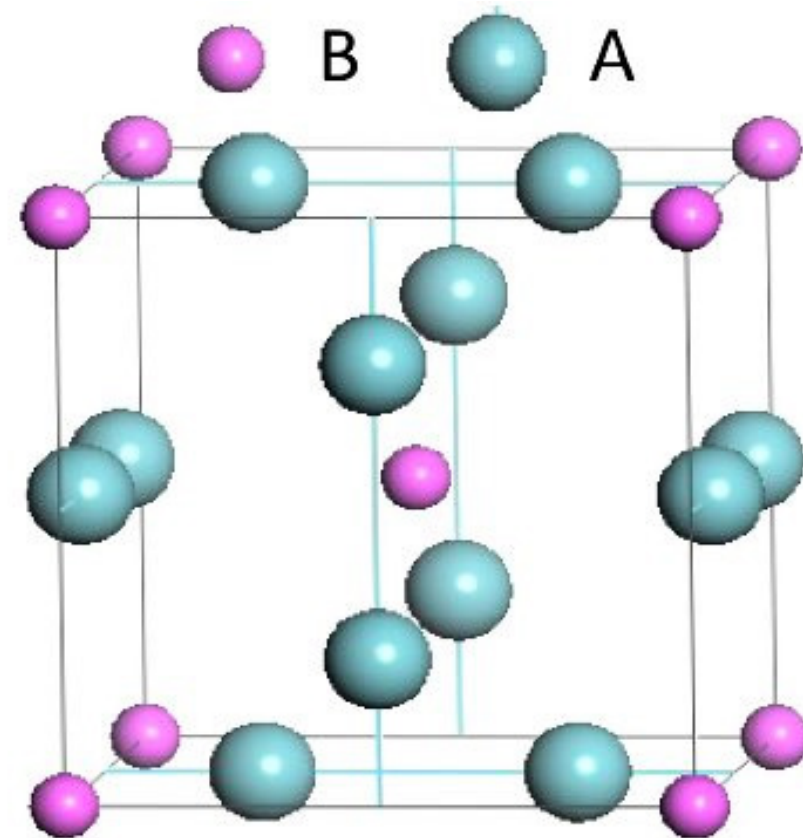


Figure 3 Surface resistance vs temperature for a NbTiN sample, at 4 GHz.

A15 COMPOUNDS



A15 COMPOUNDS : HIGH T_c

compound	T_c (K)	compound	T_c (K)	compound	T_c (K)	compound	T_c (K)
Ti ₃ Ir	4.6	V ₃ Os	5.15	Nb ₃ Os	0.94	Cr ₃ Ru	3.43
Ti ₃ Pt	0.49	V ₃ Rh	0.38	Nb ₃ Rh	2.5	Cr ₃ Os	4.03
Ti ₃ Sb	5.8	V ₃ Ir	1.39	Nb ₃ Ir	1.76	Cr ₃ Rh	0.07
		V ₃ Ni	0.57	Nb ₃ Pt	10	Cr ₃ Ir	0.17
Zr ₃ Au	0.92	V ₃ Pd	0.08	Nb ₃ Au	11		
Zr ₃ Pb	0.76	V ₃ Pb	3.7	Nb ₃ Al	20.3	Mo ₃ Re	15
		V ₃ Au	3.2	Nb ₃ Ga	18.9	Mo ₃ Os	11.68
		V ₃ Al	9.6	Nb ₃ In	8	Mo ₃ Ir	8.1
		V ₃ Ga	15.4	Nb ₃ Ge	23	Mo ₃ Pt	4.56
		V ₃ In	13.9	Nb ₃ Sn	18.3	Mo ₃ Al	0.58
		V ₃ Si	17.1	Nb ₃ Bi	2.25	Mo ₃ Ga	0.76
		V ₃ Ge	/			Mo ₃ Si	1.3
		V ₃ Sn	4.3	Ta ₃ Ge	8	Mo ₃ Ge	1.4
		V ₃ Sb	0.8	Ta ₃ Sn	6.4		
				Ta ₃ Sb	0.72		

[after Due-Hugues]

Phases with proper stoichiometry
(A₃B) not stable in normal condition
(RT to Cryogenic temp)
=> Quenching necessary

- Among the Nb and V based high T_c (15 – 20 K)
 - Nb₃Ga and Nb₃Ge do not exist as stable bulk materials at 3:1 stoichiometry
 - Nb₃Al exists only at high temperature causing excessive atomic disorder
- Production of above materials need non equilibrium processes*
- V₃Ga, V₃Si & Nb₃Sn are stable bulk material and have high T_c
 - Nb₃Sn =Special interest for SRF since the 1980's
 - Another A-15 compound holding promise is Mo₃Re (T_c =15K)

A15 COMPOUNDS : HIGH T_c

compound	T_c (K)	compound	T_c (K)	compound	T_c (K)	compound	T_c (K)
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		V ₃ Ni	0.57	Nb ₃ Pt	10	Cr ₃ Ir	0.17
Zr ₃ Au	0.92	V ₃ Pd	0.08	Nb ₃ Au	11		
Zr ₃ Pb	0.76	V ₃ Pb	3.7	Nb ₃ Al	20.3	Mo ₃ Re	15
		V ₃ Au	3.2	Nb ₃ Ga	18.9	Mo ₃ Os	11.68
		V ₃ Al	9.6	Nb ₃ In	8	Mo ₃ Ir	8.1
		V ₃ Ga	15.4	Nb ₃ Ge	23	Mo ₃ Pt	4.56
		V ₃ In	13.9	Nb ₃ Sn	18.3	Mo ₃ Al	0.58
		V ₃ Si	17.1	Nb ₃ Bi	2.25	Mo ₃ Ga	0.76
		V ₃ Ge	/			Mo ₃ Si	1.3
		V ₃ Sn	4.3	Ta ₃ Ge	8	Mo ₃ Ge	1.4
		V ₃ Sb	0.8	Ta ₃ Sn	6.4		
				Ta ₃ Sb	0.72		

[after Due-Hugues]

- Among the Nb and V based high T_c (15 – 20 K)
 - Nb₃Ga and Nb₃Ge do not exist as stable bulk materials at 3:1 stoichiometry
 - Nb₃Al exists only at high temperature causing excessive atomic disorder
- Production of above materials need non equilibrium processes*
- V₃Ga, V₃Si & Nb₃Sn are stable bulk material and have high T_c
 - Nb₃Sn =Special interest for SRF since the 1980's
 - Another A-15 compound holding promise is Mo₃Re (T_c =15K)

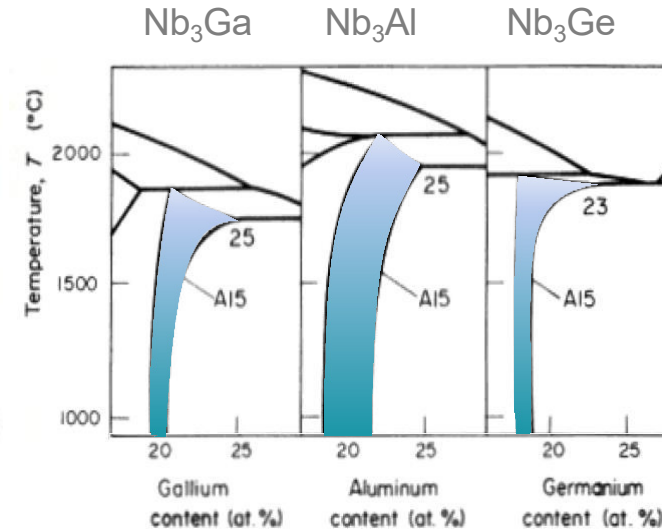
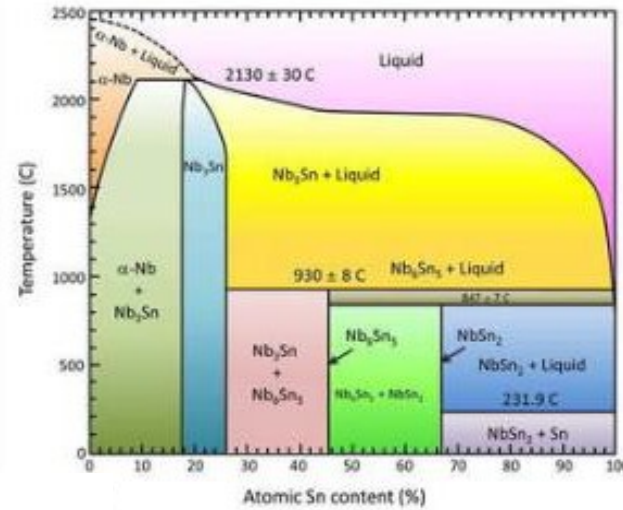
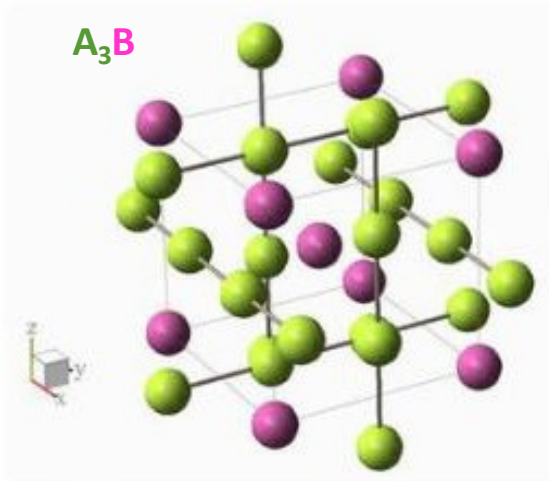
❑ Extreme brittleness !!!

- ❖ cannot be plastically formed
- ❖ thin/thick film route only !

nm ↙ ↘ μm

Phases with proper stoichiometry (A₃B) not stable in normal condition (RT to Cryogenic temp) => Quenching necessary

A15 COMPOUNDS : NARROW DOMAIN OF SC



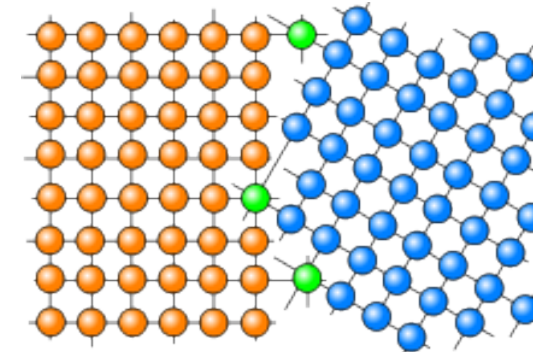
B atoms occupy corners and center of BCC structure

A atoms form orthogonal chains bisecting the faces of the BCC unit cell.

Linear Chain Integrity is crucial for T_c (long-range order required)

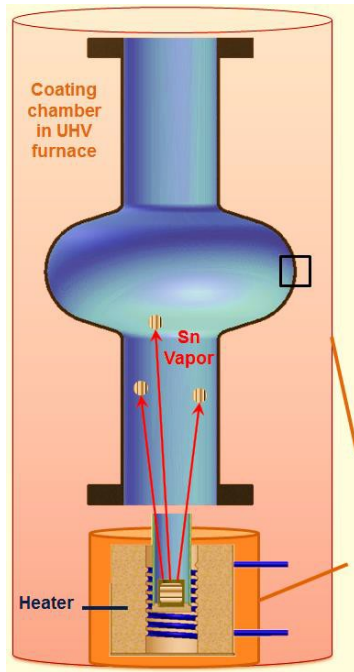
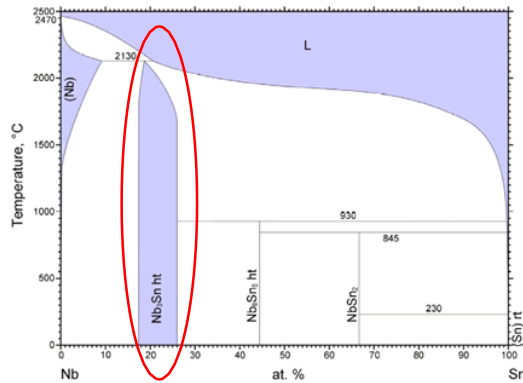
■ Narrow range of concentration for the SC phase:

- Highest T_c area is even narrower
- Difficult to get uniform SC phase everywhere*
- Special issues at grain boundaries: “intrinsic” local deviation of stoichiometry*
- In Nb_3Sn wires : GB exhibit degraded SC => weak links, pinning centers

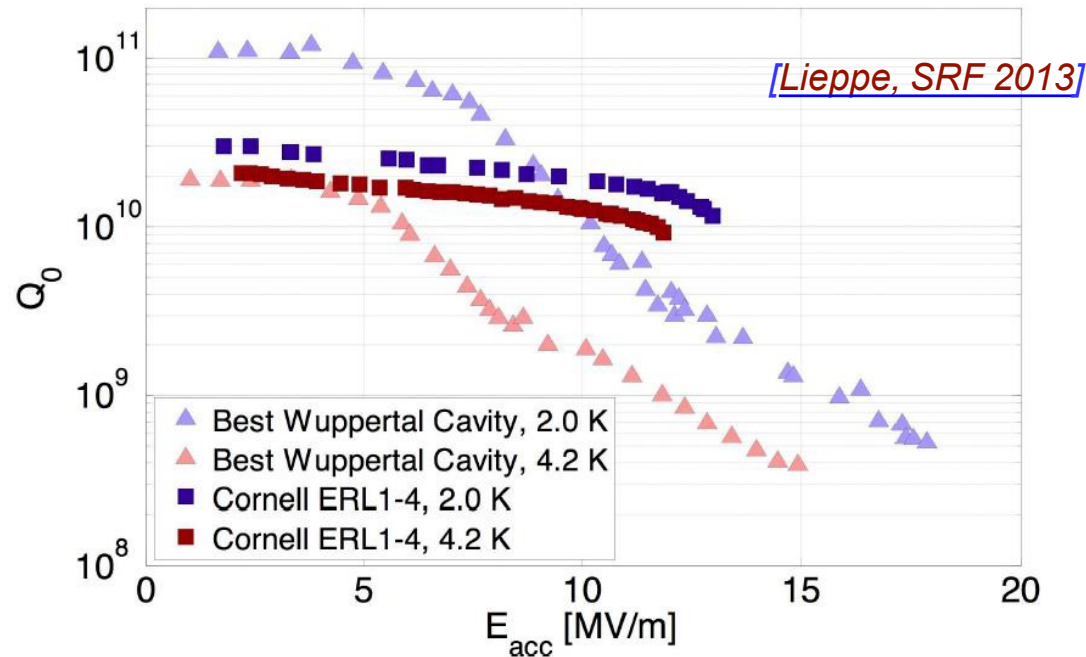


↔
Compare with ξ

NB₃SN ON NB (thermal way)

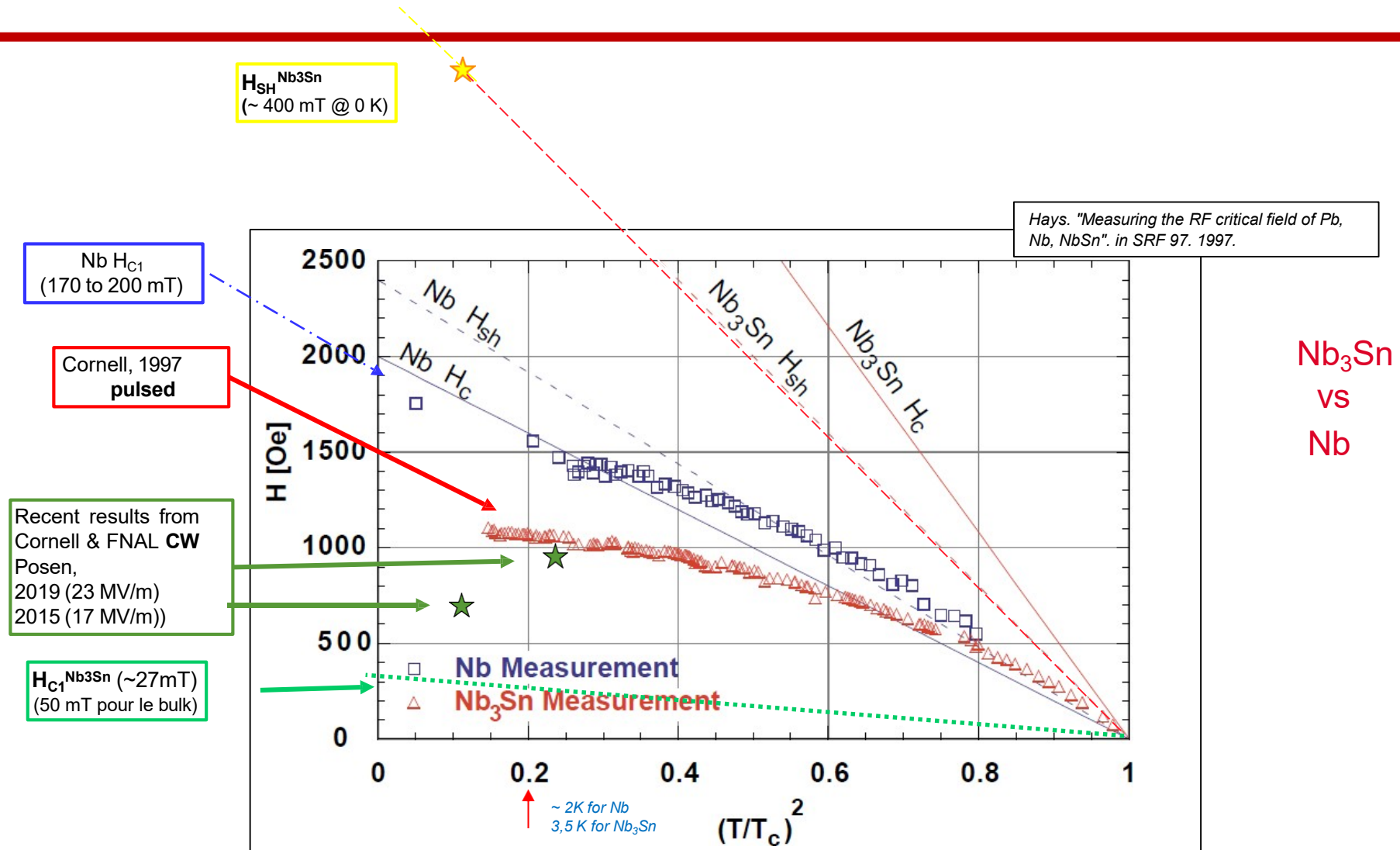


Pioneer work: Wuppertal, Cornell



- @ 4,2 K: $Q_0 \times 20$ compare to Nb, @ 2K ~ the same
- Limited in E_{acc} , best results today ~20 MV/m
- Important developments: FNAL, JLAB, CERN, PKU....

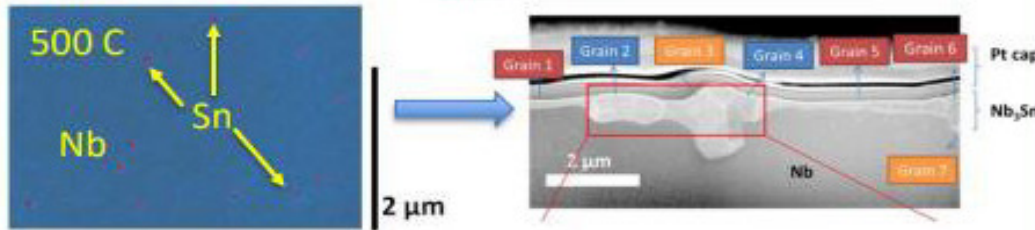
EFFECTS OF LOCAL DEFECTS



=> We have to reduce defect density (yes but which ones?)

Issues with Nb₃Sn by vapor diffusion

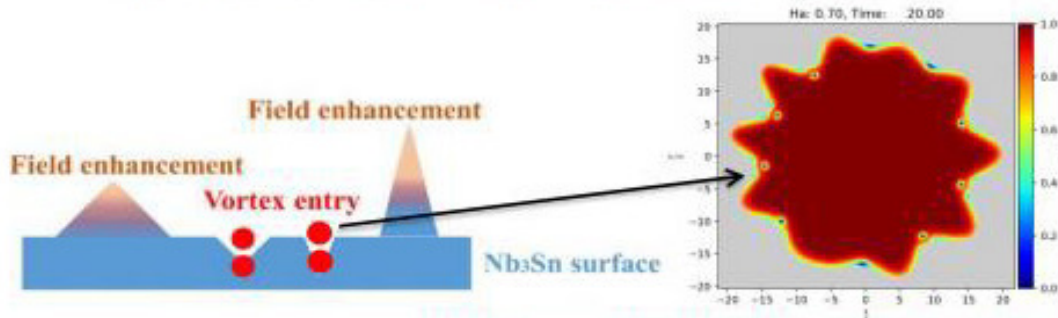
- Large surface roughness



Non-uniform nucleation events
(Ryan Porter, et al., LINAC'19)

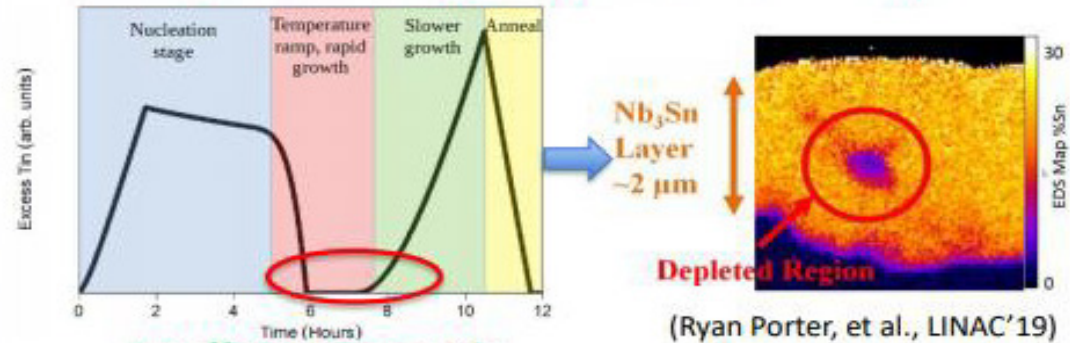
Source of surface roughness: large variation in grains
(Jaeyel Lee, et al., Supercond. Sci. Technol., 2018)

- Impact on the RF performance



Vortex nucleation for rough surface
(Alden Pack, et al., PRB, 2020)

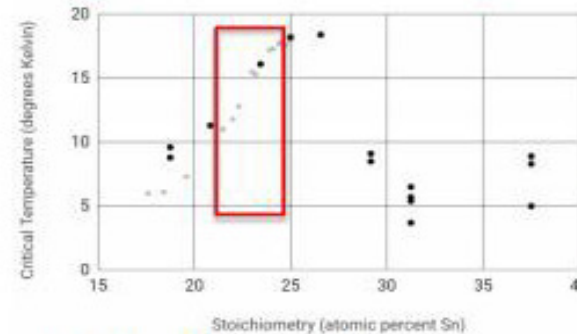
- Sn depletion region/stoichiometry



Insufficient Sn supply
(Nathan Sitaraman, et al.)

arXiv:1912.07576 [cond-mat.supr-con]

- Impact on the RF performance



(Nathan Sitaraman, et al.)

arXiv:1912.07576 [cond-mat.supr-con]



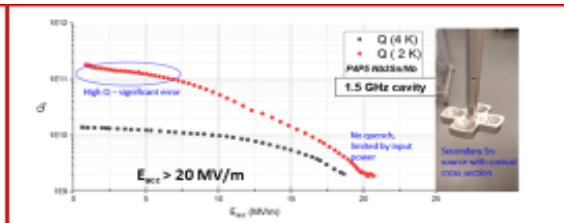
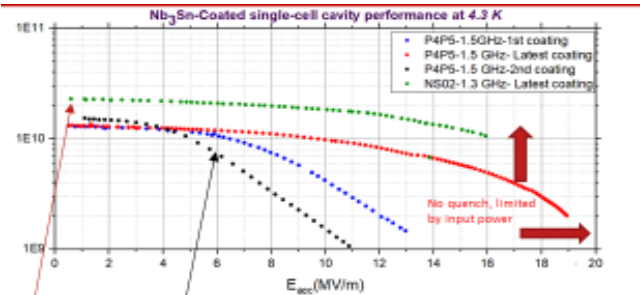
Conventional Sn vapor diffusion method for generating Nb₃Sn

https://indico.jlab.org/event/405/contributions/7865/attachments/6613/8979/Zeming%20Sun_Thin%20film%20workshop%20slide.pdf

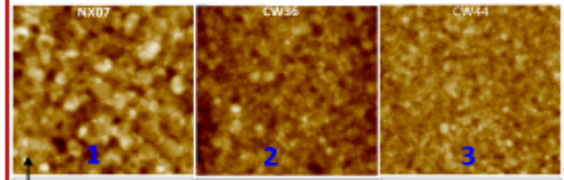
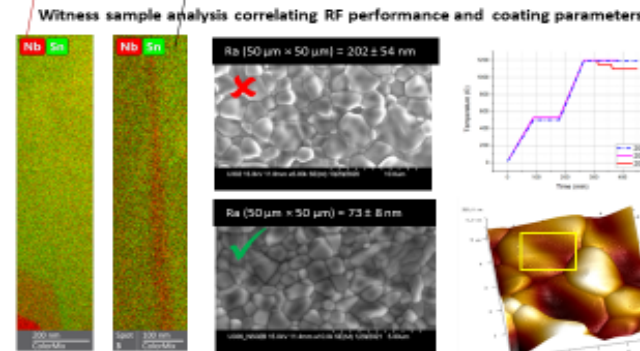
Developments around Nb₃Sn



Material studies and development of Nb₃Sn-coated cavities



Several cavities reached to ~ 20 MV/m, material studies and surface improvement is in progress to maintain high Q at high E_{acc}.

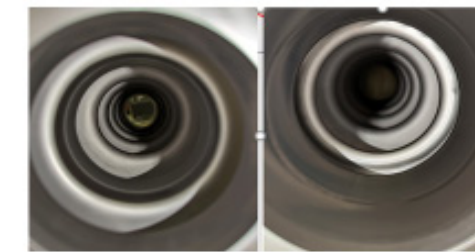
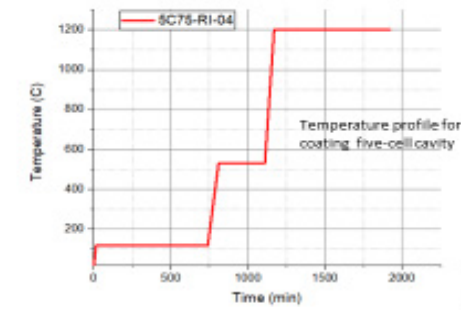


Expt	1200C	1150C	1100C	Ra (nm)	Grain size (μm)	Thickness (μm)
00	58 min	45 min	85 min	85.4 ± 3.1	1.13	-
01	10 min	45 min	85 min	69 ± 3	0.93	1.07*
02	10 min	45 min	-	40 ± 1	0.75	1.0 +/- 0.2
03	10 min	-	-	27 ± 2	0.55	0.75
04	10 min	-	85 min	43.5 ± 1.5	0.6	1.02

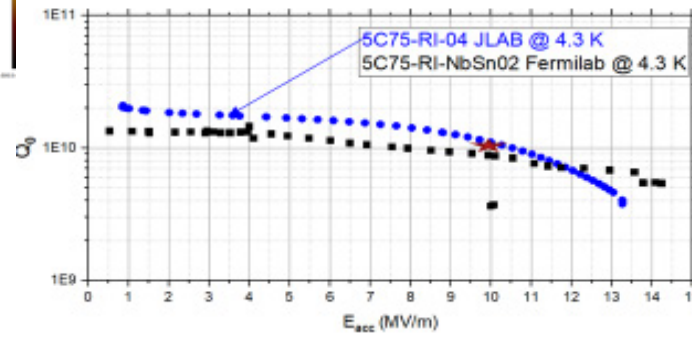
- Several temperature profiles are identified to control roughness and thickness.
- Cavity coatings with selected profiles are in progress for potential improvement in RF performance.

Jefferson Lab

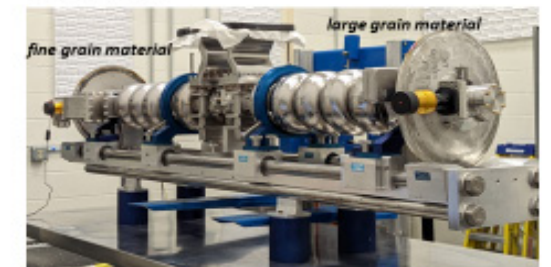
Multi-cell cavity coating and Nb₃Sn QCM



- Spec: 1E10 @ 10 MV/m
- One cavity was coated at Fermilab and another at Jlab.
- Cavity assembled in the pair and subjected to disassembly because of a leak, and assembled again with some degradation in the cavity performance
- Ready to be installed into a quarter module; test in the UITF later this year.



- Original C75 cavity made of *large grain material*
- Limited by multipacting (no quench)



Based on G. Ereemeev's ECA, Jlab cavity work supported by R&D fund.

Jefferson Lab

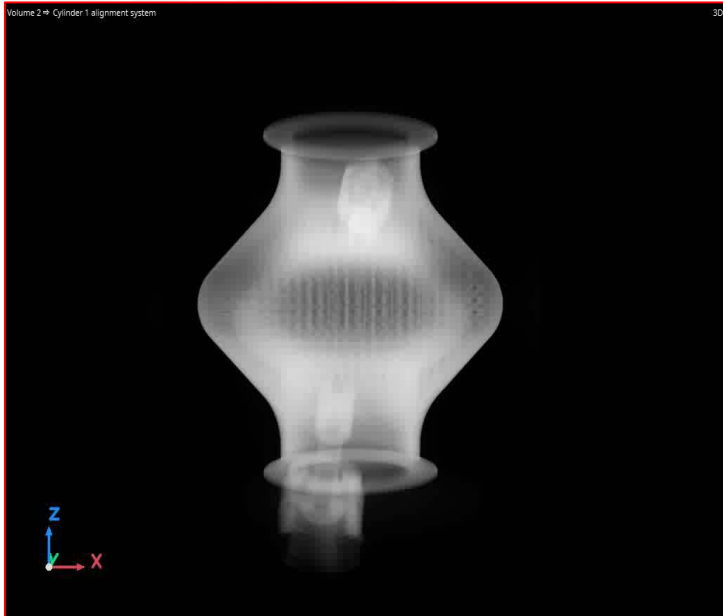
U. Pudasaini

Developments around Nb₃Sn



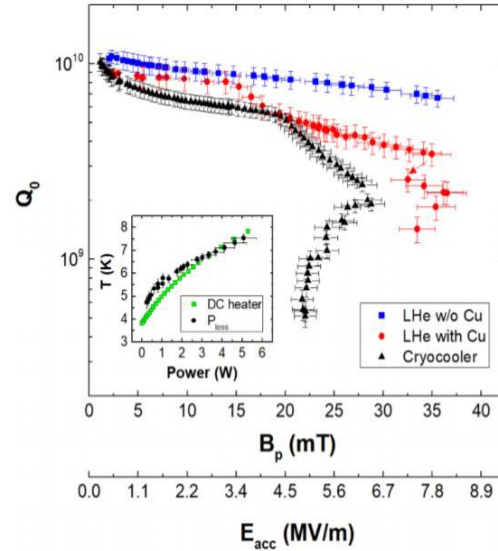
Compact Accelerators for Societal Needs

Courtesy G. Ciovati et al.



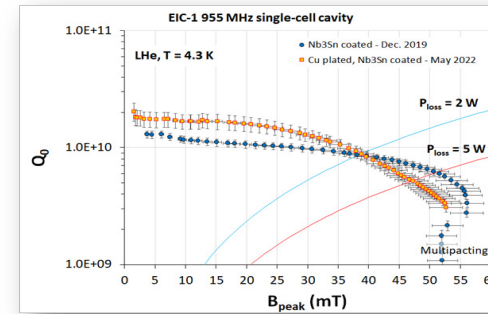
3D add. Fabrication with cooling circulating capillary integrated in the walls (thermosiphon approach)

- Reduced He volume
- Efficient cooling

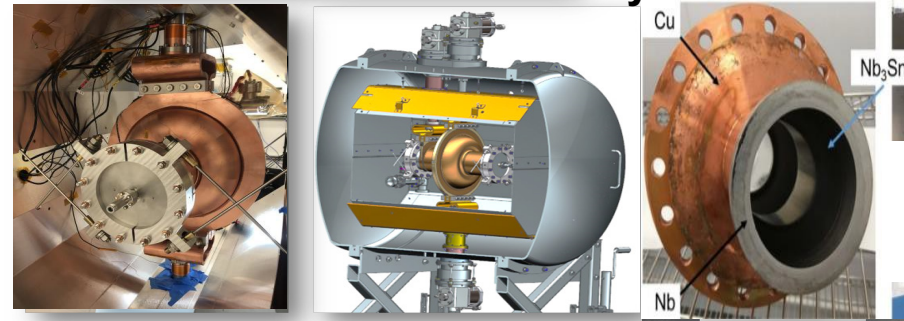


General Atomics

Development of conduction-cooled SRF cavities



Cryocooler



Horizontal Test Cryostat for conduction-cooled SRF cavities

G. Ciovati et al., "Development of a prototype superconducting radio-frequency cavity for conduction-cooled accelerators", arXiv:2302.07201 [physics.acc-ph], 2023

A15 Compounds – Preparation Methods

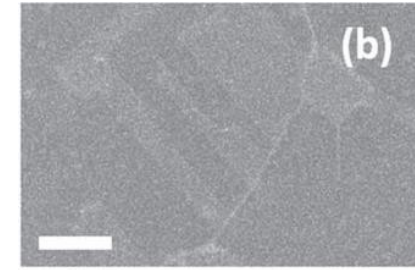
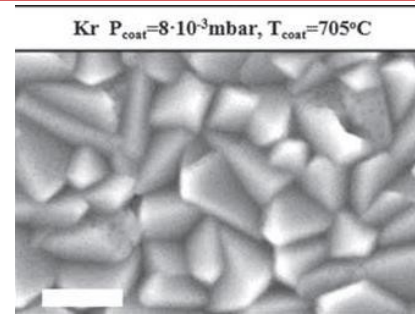
Nb₃Sn: Other Deposition Methods

❑ Sputtered Nb₃Sn films on copper

- Activities at [Cern](#), [STFC](#), [Jlab](#), [Cornell](#),,,
- RT deposited films : right composition but no A15 structure
- Heating of substrate (CERN)
- And/or post annealing

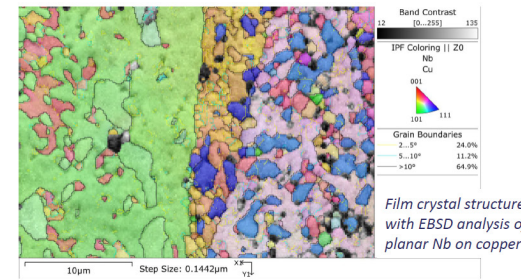
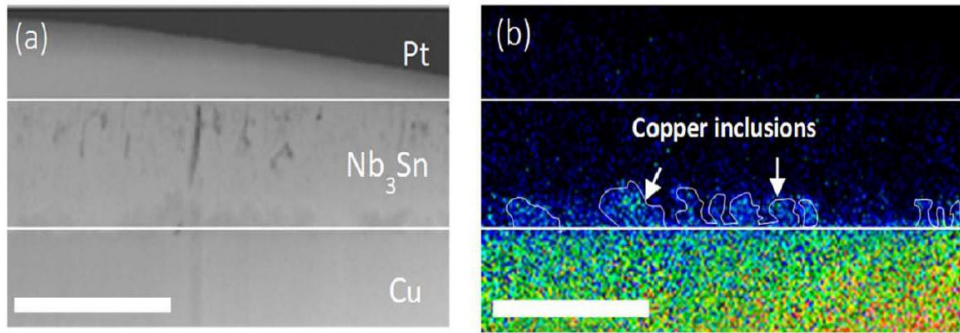
❑ Other issues

- Cracks on the layer (due to differential dilatation coef)
- Diffusion of copper in the layer
- Carrier gas incorporation (Ar, Kr)
- Sn evaporation at higher temperature (> 1000°C)

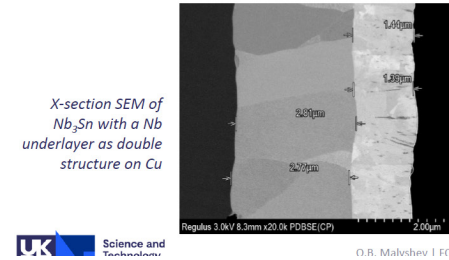


Supercond. Sci. Technol. 32 (2019) 035002

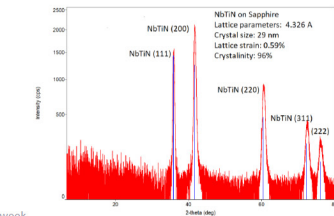
E A Ilyina et al



Film crystal structure with EBSD analysis of planar Nb on copper



X-section SEM of Nb₃Sn with a Nb underlayer as double structure on Cu



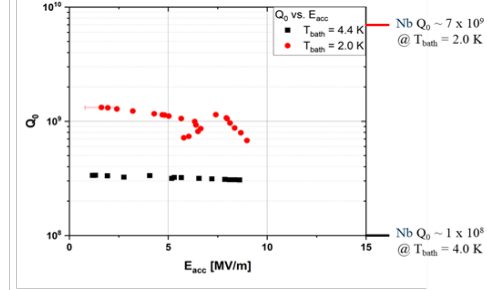
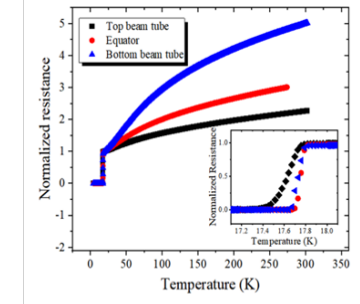
XRD analysis of NbTiN deposited on copper

Cylindrical Magnetron Sputtering of Nb₃Sn coating (2.6 GHz Nb SRF cavity)

HEP Stewardship

50 nm Nb
25 nm Sn
50 nm Nb
25 nm Sn
50 nm Nb
25 nm Sn
200 nm Nb
Nb substrate

Courtesy S. Sharifuzzaman et al.



Results

(Q₀) of 3.2 x 10⁸ at E_{acc} = 5 MV/m at T_{bath} = 4.4 K

(Q₀) of 1.1 x 10⁹ at E_{acc} = 5 MV/m at T_{bath} = 2.0 K

Science and Technology

Courtesy of R. Valizadeh (STFC)



Nb₃Sn: Other Deposition Methods

❑ Electrochemical deposition + diffusion through copper

- Proposed at [FNAL](#)
- Inspired from wire fabrication
- Not expensive !!!!

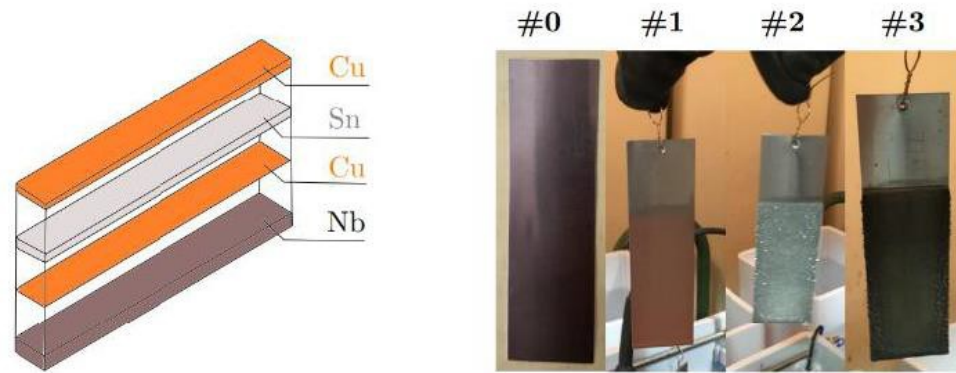
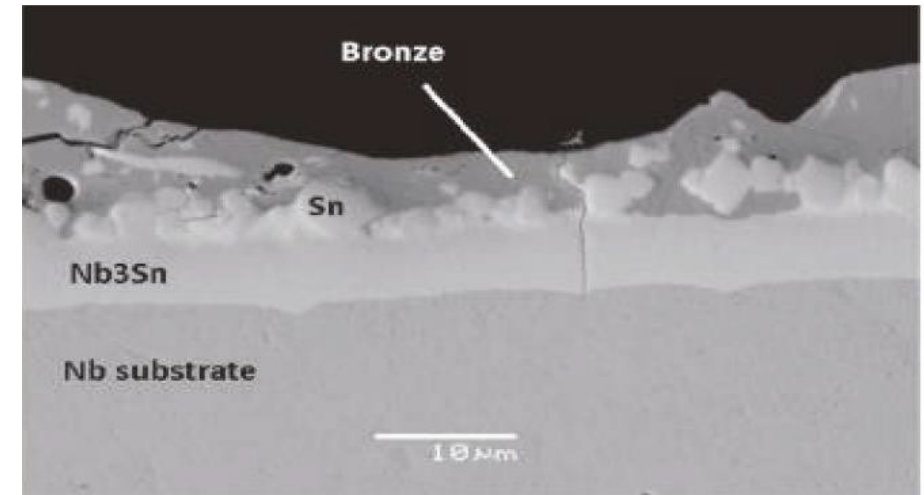


Fig. 3: Sequence of deposited layers (left), and pictures of sample at each deposition step (right).

- Multilayer is heated => solid state diffusion
- Cu lowers the formation T_p° of A15 phase and suppresses the unwanted NbSn₂ and Nb₆Sn₅ phases.

❑ Bronze Route

- Nb deposition on CuSn
- Heat treatment to diffuse Sn into Nb
- Not expensive !!!!



Current landscape for developments around Nb₃Sn

...non-exhaustive list

Method	Sn vapor/ liquid diffusion	Sputtering ! target	Co-sputtering	Sequential sputtering + annealing	Bronze routes (electrochemical deposition + annealing)
substrate	Nb only	Nb or Cu	Nb or Cu	Nb or Cu	Nb or Cu
Activities at	Cornell FNAL Jlab INFN KEK	Cornell Cern Jlab INFN STFC FSU	STFC* Peking U. Darmstadt Wisconsin U.*	IMP Jlab ODU FNAL	IMP Cornell FNAL FSU E2P

* In Multilayer structures

■ NB : work on V₃Si also in progress

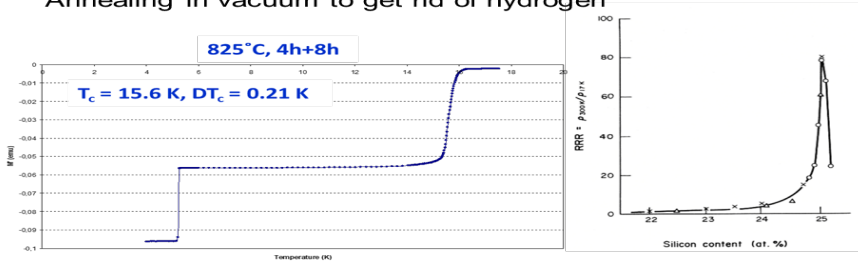
OTHER A15 COMPOUNDS

S. Deambrosi et al., Physica C 441 (2006) 108-113

Highly ordered compound, RRR~80 achievable, max T_c (17.1K) when stoichiometric composition (25at.% Si)

V₃Si layers by silanization of V substrate and Thermal Diffusion

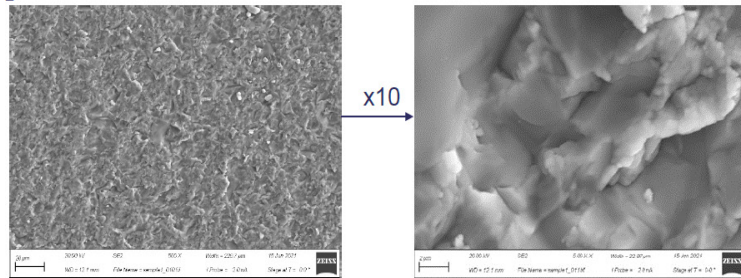
V substrate heated to get SiH₄ decomposition and Silicon diffusion
 Film grown by silanization with p (SiH₄) ~ 10⁻³-10⁻⁴ mbar
 Annealing in vacuum to get rid of hydrogen



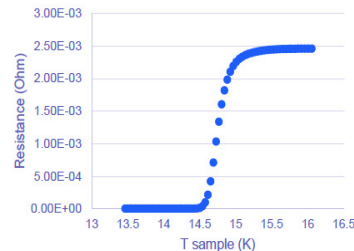
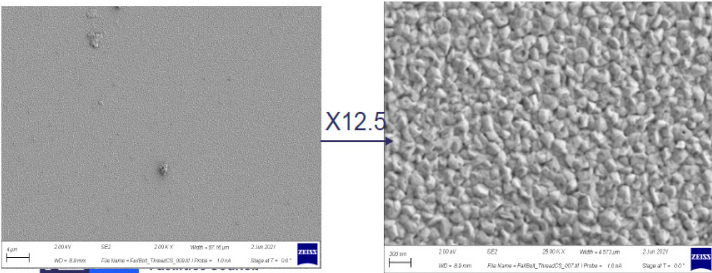
- * Diffusion parameters and silane flow rate have been optimized
- * T_c ~ 16 K is routinely obtained
- * RF measurement on 6 GHz V-cavities will be available soon

To improve film quality a new set-up capable of depositing up to 800 °C was developed. Sapphire samples were also used at this higher temperature.

Sapphire at 650 °C showed a rough surface finish. However, increasing the temperature closer to 700 °C leads to a smoother film with T_c 15.2 °K



SEM images of V₃Si deposited on Sapphire at 650 °C



T_c measurement of V₃Si deposited on Sapphire at 700 °C

Mo₃Re thin films by DC magnetron deposition: Mo₇₅Re₂₅, Mo₆₀Re₄₀

Solid solution, free of bulk and surface inhomogeneities, low interstitials solubility compared to Nb, low κ, high H_{c1} (500G)

Bulk in σ phase, tetragonal low T_c (6K)

but T_c up to 18K reported in literature with bcc structure

S.M. Deambrosi et al., Physica C 441(2006) 108-113

- * Deposition on Sapphire, Cu and Nb substrates
- * Substrate temperature up to 950° C
- * Post-annealing to increase crystallinity and transition sharpness
- * T_c = 12K obtained for composition Mo₆₀Re₄₀

Higher deposition temperature, longer annealing time

➔ Higher T_c

SRf Tutorials 2023 - Beyond Bulk Nb

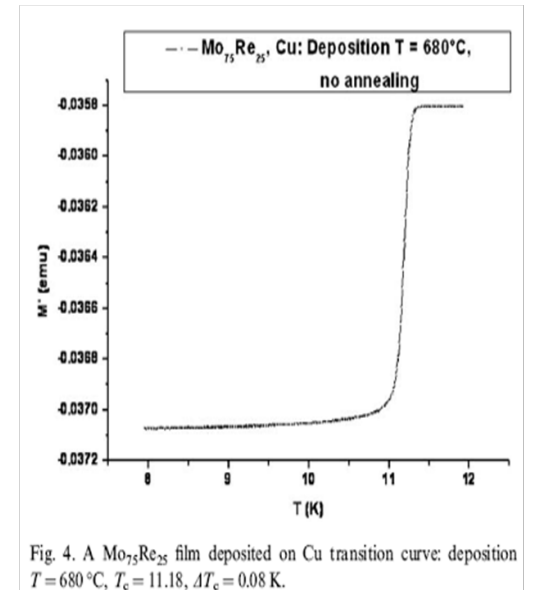


Fig. 4. A Mo₇₅Re₂₅ film deposited on Cu transition curve: deposition T = 680 °C, T_c = 11.18, ΔT_c = 0.08 K.

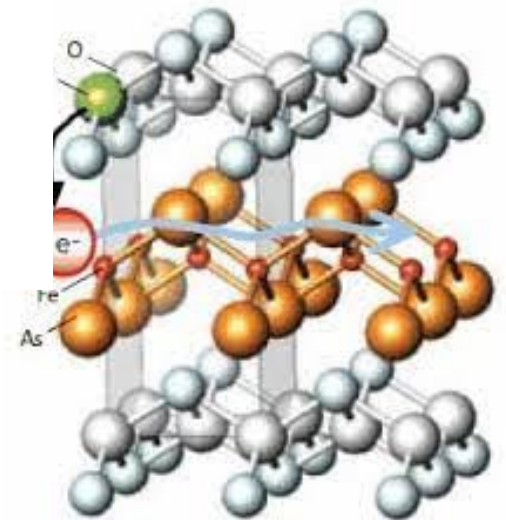
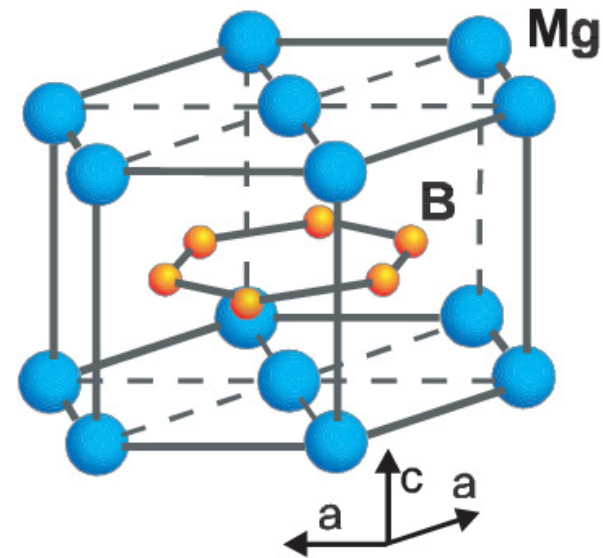
Courtesy R. Valizadeh

2-D SC (Compounds, anisotropic)

-MgB₂

-Cuprates, Pnictides

-Multilayers



MAGNESIUM DIBORIDE (MgB₂)

BCS type superconductor

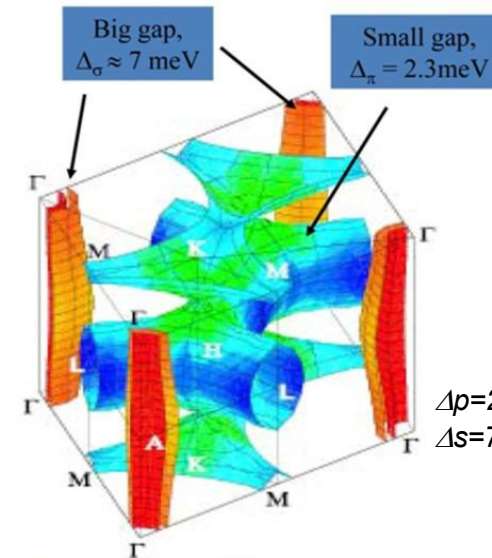
- ❑ T_C ~40 K, two-gap nature
- ❑ Advantages:
 - Very high T_C (higher temp operation)
 - Semimetal, cheap (fertilizer !)
 - ξ , 1l of high quality* MgB₂ similar to Nb (~50 nm) (transparency of GB to current flow)
 - LOW ρ_n (lower R_s)
- ❑ Disadvantages:
 - Orientation issues (in polycrystalline materials !)
 - RF dominated by lower gap ☹ !
 - Still better than Nb :

$$\Delta_{\text{Nb}} = 1.5 \text{ meV} < \Delta_{\text{MgB}_2} = 2.3 \text{ meV} < \Delta_{\text{Nb}_3\text{Sn}} = 3.1 \text{ meV}$$

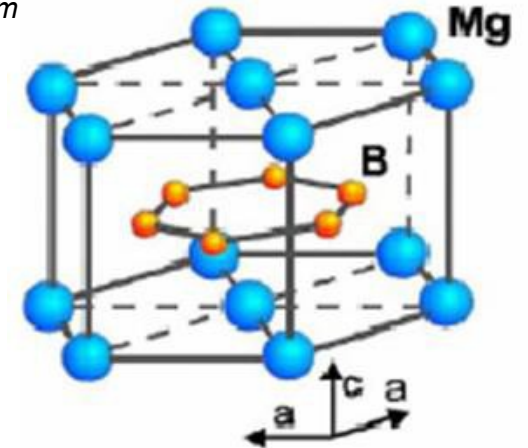
$$\Delta_{\text{MgB}_2} < \Delta_{\sigma} = 7.1 \text{ meV}$$

- Sensitive to H₂O (capping necessary ?)
- Thin film routes difficult to achieve

Graphite-type boron layers separated by hexagonal close-packed layers of magnesium



Liu, Mazin and Kortus (2002);
Choi et al, (2002)



$\Delta_p = 2.3 \text{ meV}$, 2D, in-plane s-orbital
 $\Delta_s = 7.1 \text{ meV}$ 3D, out-of-plane p-orbitals

A. Floris et al., cond- mat/0408688v1 31 Aug 2004

MAGNESIUM DIBORIDE (MgB₂)

Phase diagram: at low Mg pressure only extremely low deposition temperatures can be used

□ Optimal T for epitaxial growth $\sim T_{\text{melt}}/2$

- For MgB₂ $T_{\text{melt}}/2 = 540^\circ\text{C} \Rightarrow P^{\text{Mg}} \sim 11$ Torr
- Too high for UHV deposition techniques (PLD, MBE...)

□ At $P^{\text{Mg}} = 10^{-4}$ - 10^{-6} Torr, and $T_{\text{sub}} \sim 400^\circ\text{C}$

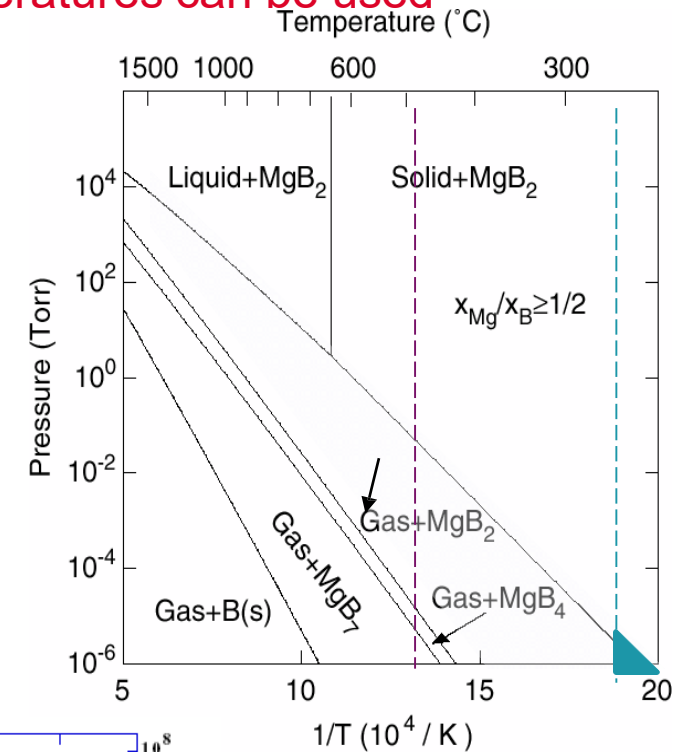
□ Compatible with MBE, and other deposition techniques MgB₂ is stable, but

no MgB₂ formation:

- Mg atoms re-evaporate before reacting with B

□ At $P^{\text{Mg}} = 10^{-4}$ - 10^{-6} Torr, and lower T

- MgB₂ is stable,
- If $T_{\text{sub}} > 250^\circ\text{C}$, free Mg is lost because the re-evaporation rate is higher than the impinging rate. If $T_{\text{sub}} < 250^\circ\text{C}$

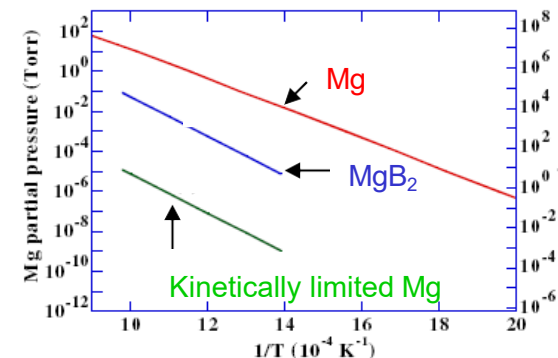


Z.-K. Liu et al., APL 78(2001) 3678.

M. Naito and K. Ueda, SUST 17 (2004) R1

.Growth rate is very slow,
(kinetically limited by available Mg)

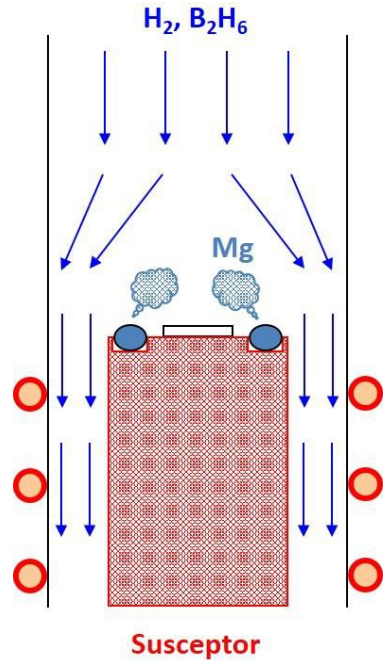
evaporation pressure of Mg from MgB₂ < decomposition curve of MgB₂ < Mg vapor pressure



MgB₂ – HPCVD ON METAL SUBSTRATES

[X. Xi- TempleUniversity]

HYBRID PHYSICAL CHEMICAL VAPOR DEPOSITION



Polycrystalline MgB₂ films deposited: On stainless steel, Nb, TiN, and other substrates.

Flat samples and tubes (*conformational*)

Fitted for SRF apps:

RRR > 80

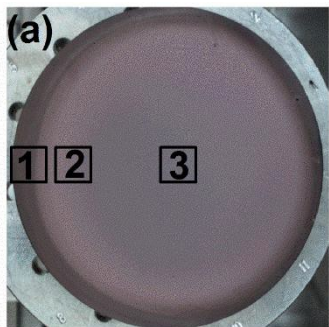
low resistivity (< 0.1 μΩ) and long mean free path high T_c ~ 42 K (due to tensile strain),

low surface resistance, short penetration depth

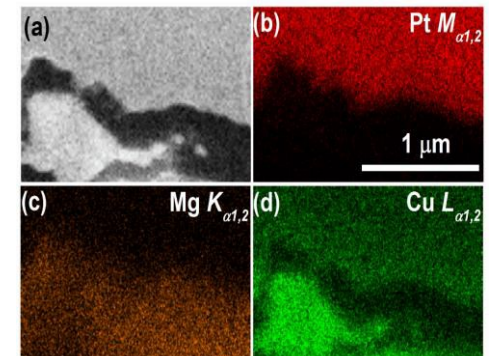
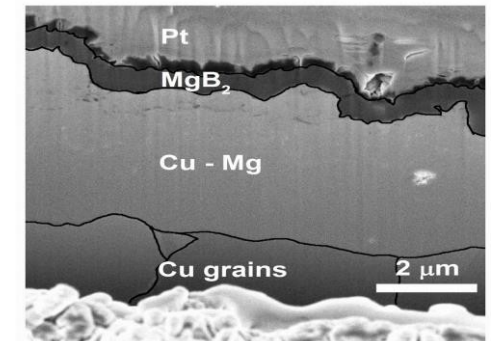
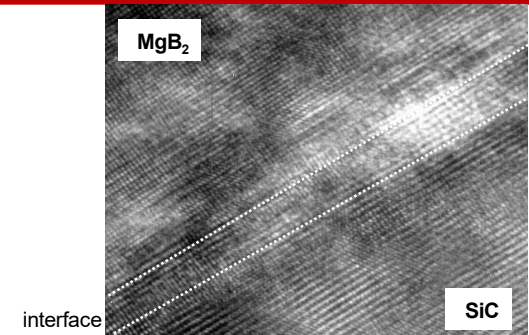
smooth surface (RMS roughness < 10 Å with N₂ addition) good thermal conductivity (free from dendritic magnetic instability)

Keys to high quality MgB₂ thin films: High Mg pressure for thermodynamic stability of MgB₂

- Oxygen-free or reducing environment Clean Mg and B sources
- Preventing formation of spurious phase (e.g.
- Mg-Cu alloy islands on a Cu substrate



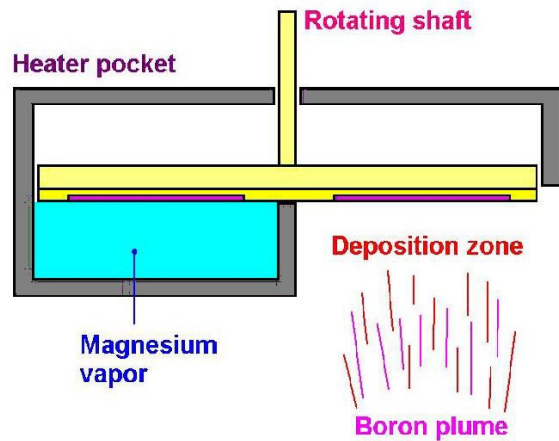
Reactor/reaction designs require complex calculation in thermodynamics and hydrodynamics



MgB₂ – OTHER ROUTES

□ In-situ reactive evaporation @ 550°C

- High quality flat samples
- Difficult to apply to complex geometries



[T. Tajima, LANL]

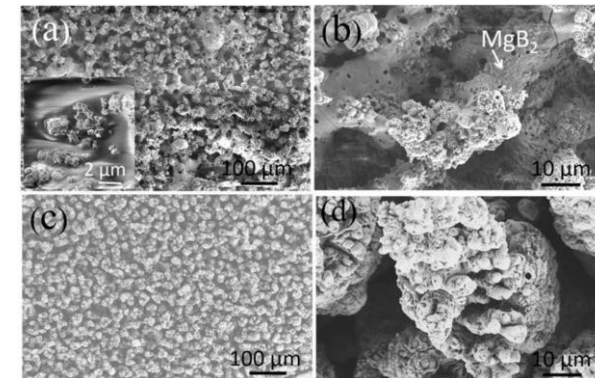
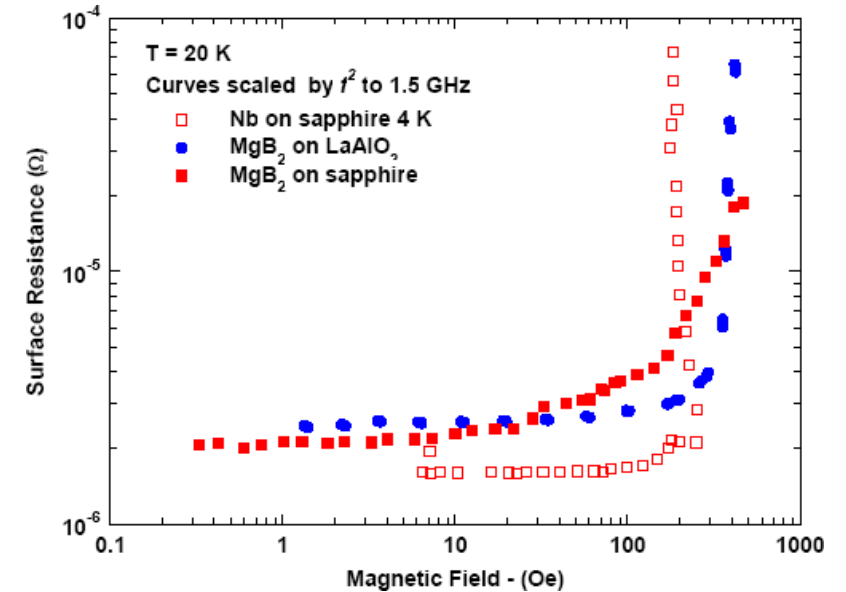
Superconducting
Technologies Inc.

□ Plasma electrolytic oxidation (PEO)

- MgB₂ particles in suspension in an electrolyte MgB₂ Islands deposited on the surface
- Issues : homogeneity, purity

To be further explored

RF measurement @ MIT/Lincoln Lab



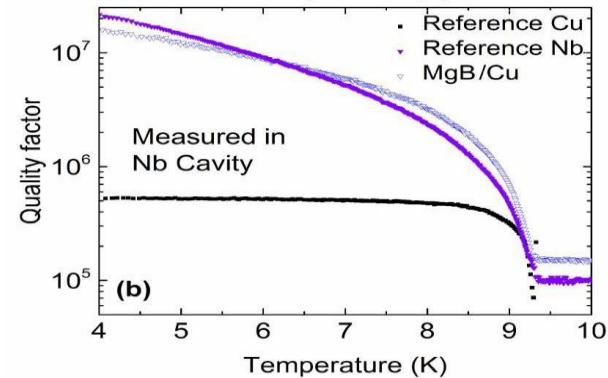
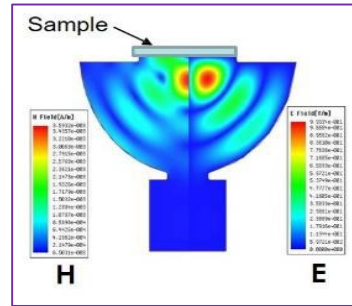
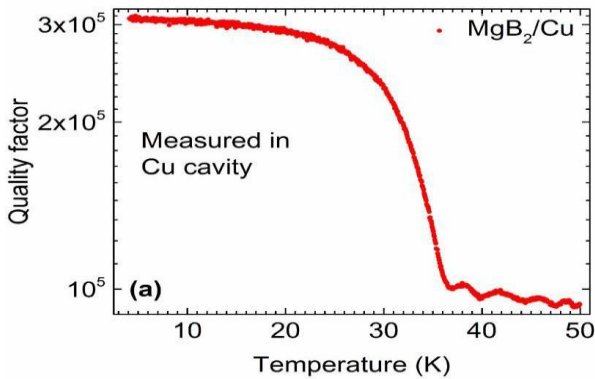
[R. Valizadeh, STFC]

HPCVD MgB₂ – RF MEASUREMENTS

11.4 GHz TE013 cavity @ SLAC

- The MgB₂ coatings were also characterized at 11.4GHz at SLAC using a cryogenic RF system.
- The samples showed a Q factor comparable to a Nb reference sample and higher than the Cu reference sample.
- The films showed a T_c of 37 K.

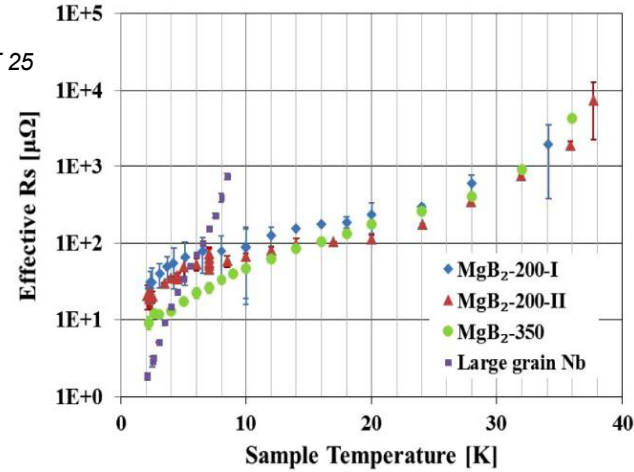
B.P.Xiao et al., SUST 25 (2012) 095006.



[P. Welander, SLAC]

Oates et al., SUST 23, 034011 (2010)

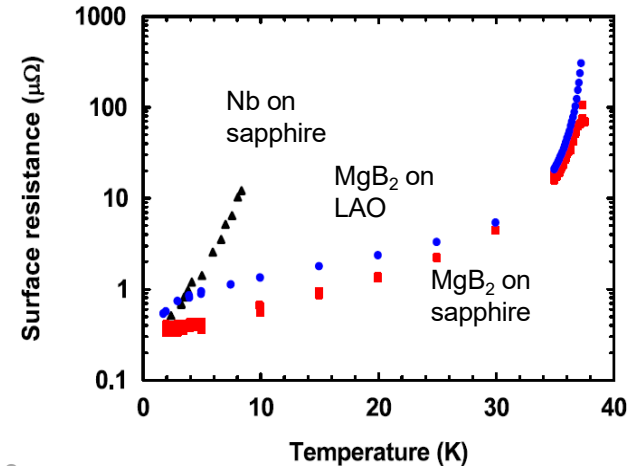
7.5 GHz sapphire-loaded TE011 cavity at JLab



Stripline resonator

Scaled to 1,5 Ghz

Lower surface resistance comparable to Nb film.



THE IMPORTANCE OF MEAN FREE PATH, MgB₂ EXAMPLE

- Most developments of HT_c are done in view of magnet applications : small ξ high ℓ by playing with ℓ
- Small x makes the superconductors very sensitive to (usual) crystalline defects (wanted for magnets, not for SRF)
- Typical values found in literature for MgB₂ : $\lambda \sim 100-150$ nm $\xi \sim 5-10$ nm

■ if $\ell \uparrow$ then:

$\xi \nearrow$
 $\lambda \searrow$
 $K \searrow \searrow$

ℓ mean free path

$$\frac{1}{\xi} = \frac{1}{\xi_0} + \frac{1}{\ell}$$

$$\lambda = \lambda_L \cdot \left(\frac{\xi_0}{\xi} \right)^{\frac{1}{2}} = \lambda_L \cdot \left(1 + \frac{\xi_0}{\ell} \right)^{\frac{1}{2}}$$

TABLE I. Penetration depth of MgB₂ calculated from 12 square-shaped junctions of different length and top electrode materials.

Top electrode material	Junction length (μm)	Calculated penetration depth (nm)
Pb	250	67.1
Pb	250	40.2
Pb	250	38.0
Pb	250	52.2
MgB ₂	8	46.3
MgB ₂	12	37.2
MgB ₂	15	50.2
MgB ₂	20	39.9
Nb	20	42.0
Nb	12	37.0
Nb	12	38.1
Nb	10	35.3

Handwritten notes in red:
 11-50 nm
 $\xi \sim$
 some 10 nm ?

APPLIED PHYSICS LETTERS **102**, 072603 (2013)

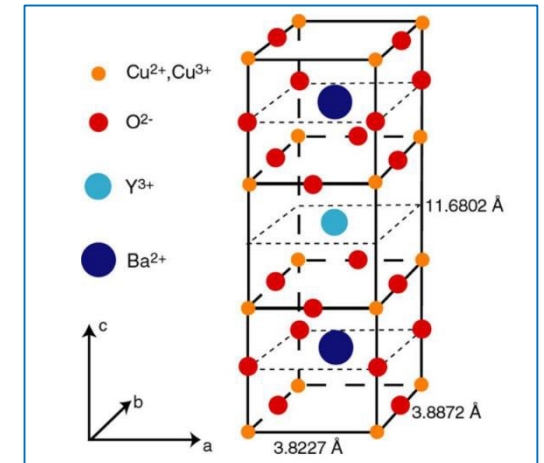
Penetration depth of MgB₂ measured using Josephson junctions and SQUIDS

Daniel Cunnane,¹ Chenggang Zhuang,¹ Ke Chen,¹ X. X. Xi,¹ Jie Yong,² and T. R. Lemberger²

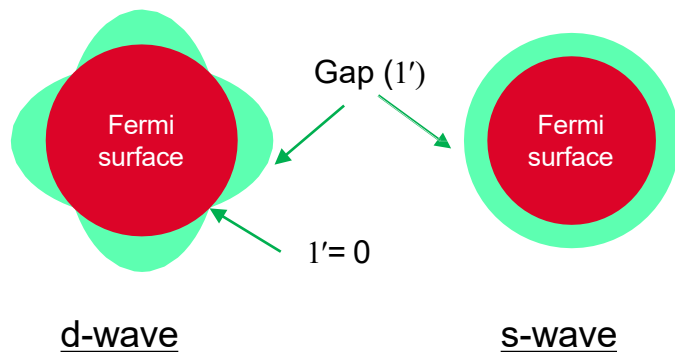
¹Department of Physics, Temple University, Philadelphia, Pennsylvania 19122, USA
²Department of Physics, The Ohio State University, Columbus, Ohio 43210, USA

YBCO FAMILY... NOT FOR SRF !

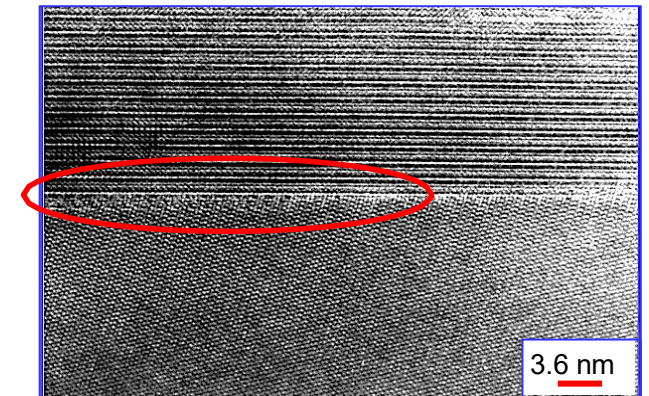
- Mono crystal: J_c maximum for (a,b) planes and minimum when // c axis
 - ξ_c (~0,03 nm) $\ll \xi_a, \xi_b$ (~1-2 nm) => "layered material"
- Realistic material : polycrystalline, ceramic, fragile...
 - $\xi_c \ll$ disordered area at G.B => grains are decoupled (weak links)
 - => try to introduce preferential orientation (epitaxy): difficult to get on a cavity (but is applied to fabricate tapes for magnets)
- D-symmetry of the gap
 - superconducting gap is also anisotropic
 - = zero at 4 line nodes located at the diagonals of the Brillouin zone
 - $l'=0 \Rightarrow$ power law for R_S : $R_S \propto T^2-T^3$
 - For the recall: gaps of conventional SC have s symmetry: isotropic and $R_S \propto e^{-1/T}$ (BCS resistance)



<http://mason.gmu.edu/~grobert1/2014syl641.htm>



Crystal structure is also related to Brillouin zones. So the relative orientation of the grains can influence the way Cooper pairs are scattered by defects



Twin boundary in YBCO

<https://areeweb.polito.it/ricerca/superconductivity/melt.htm>

PNICTIDE FAMILY... MAYBE YES ?

❑ Oxypnictide base: ReOMPn

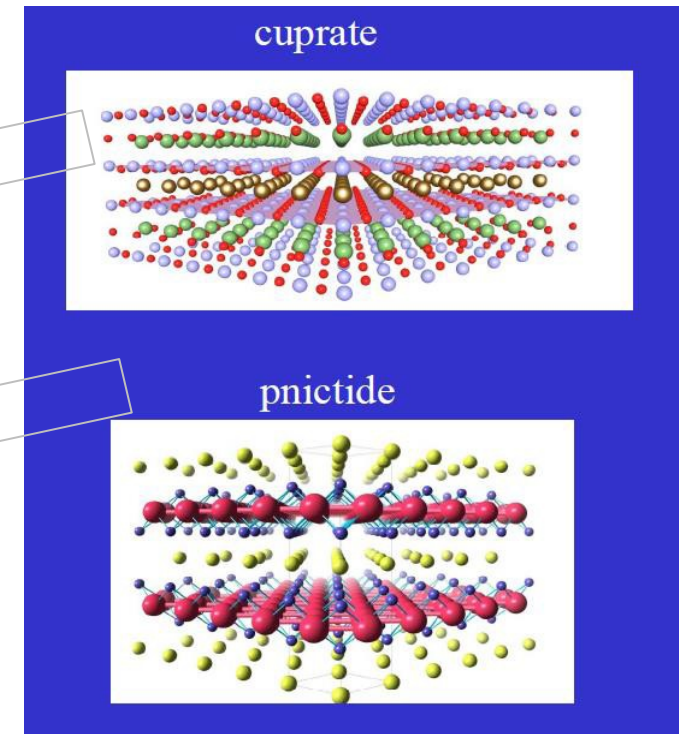
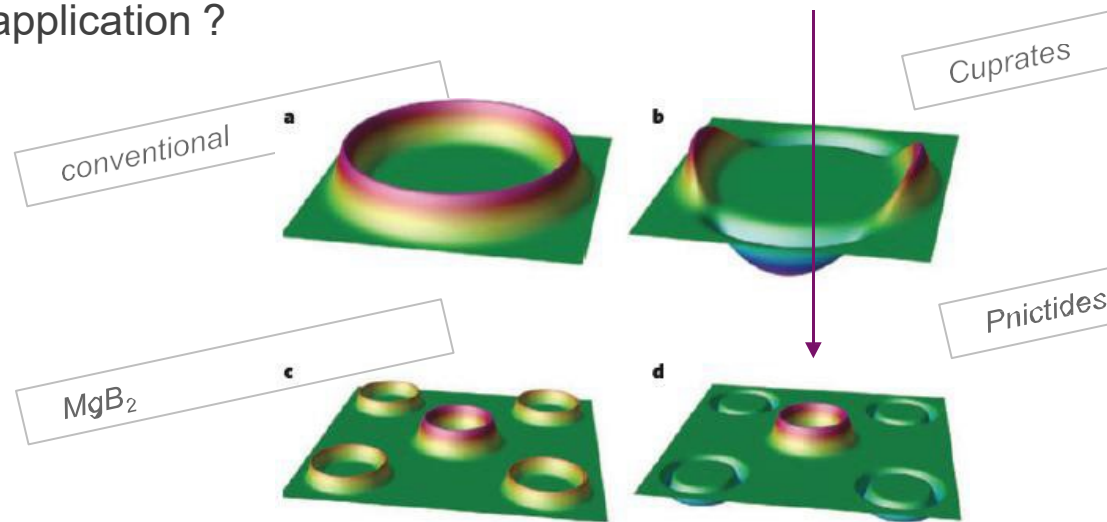
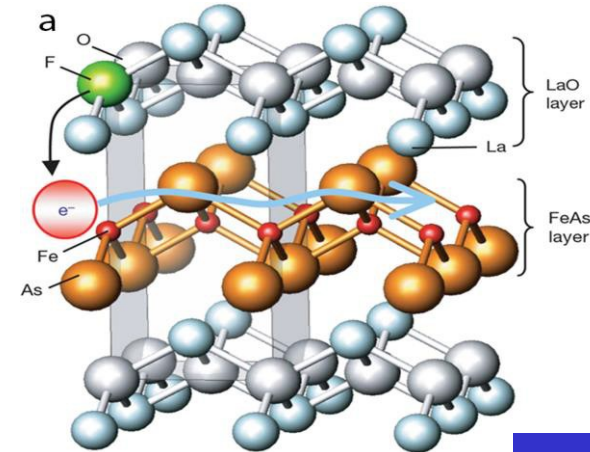
- $M = \text{Fe, Co, Ni, Pn} = \text{As or P}$
- $\text{Re} = \text{La, Nd, Sm, Pr}$

❑ A lot in common with YBCO

- High T_C (10-55 K up today) Layered structure
- Brittle material
- d-wave symmetry observed for some member of the family

❑ But most compounds exhibit s-wave gaps...?

- Opening for SRF application ?



PNICTIDE FAMILY... MAYBE YES ?

□ A lot in common with YBCO

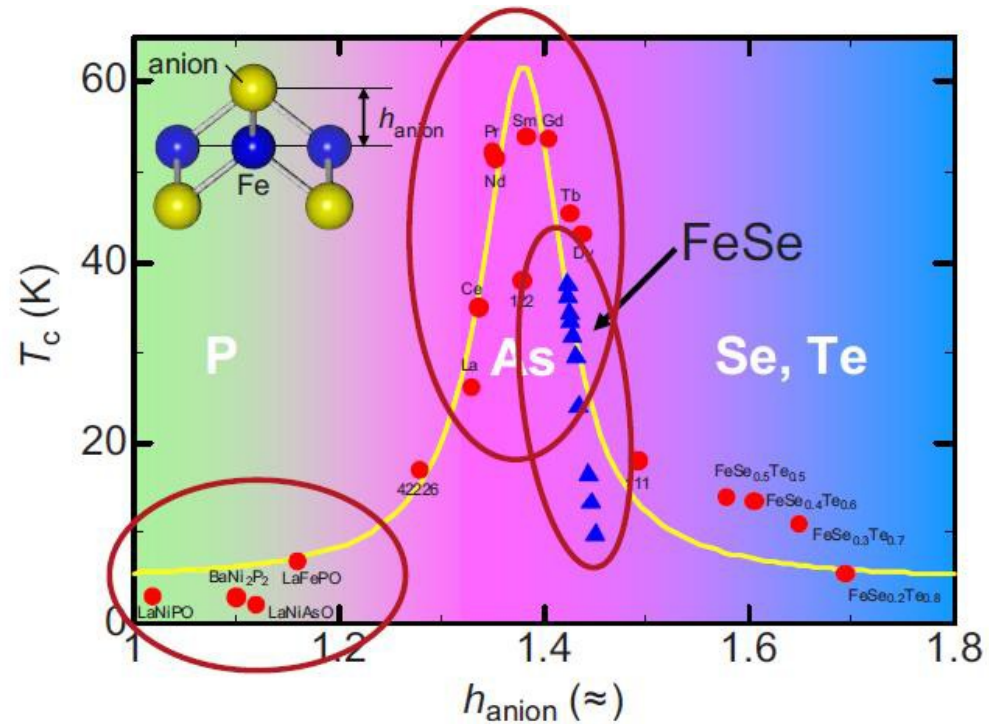
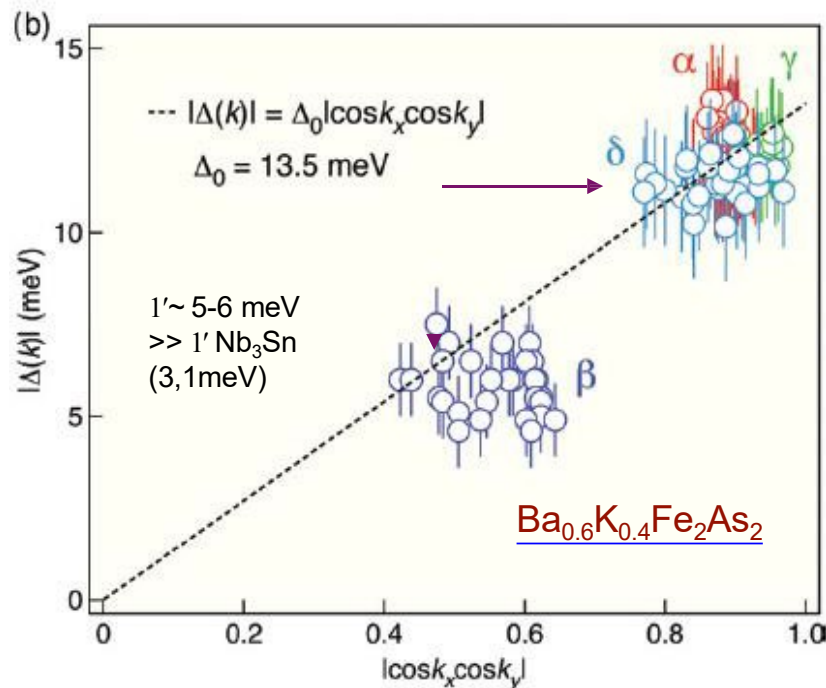
- ❖ High T_C (10-55 K up today)
- ❖ Layered structure
- ❖ Brittle material

but

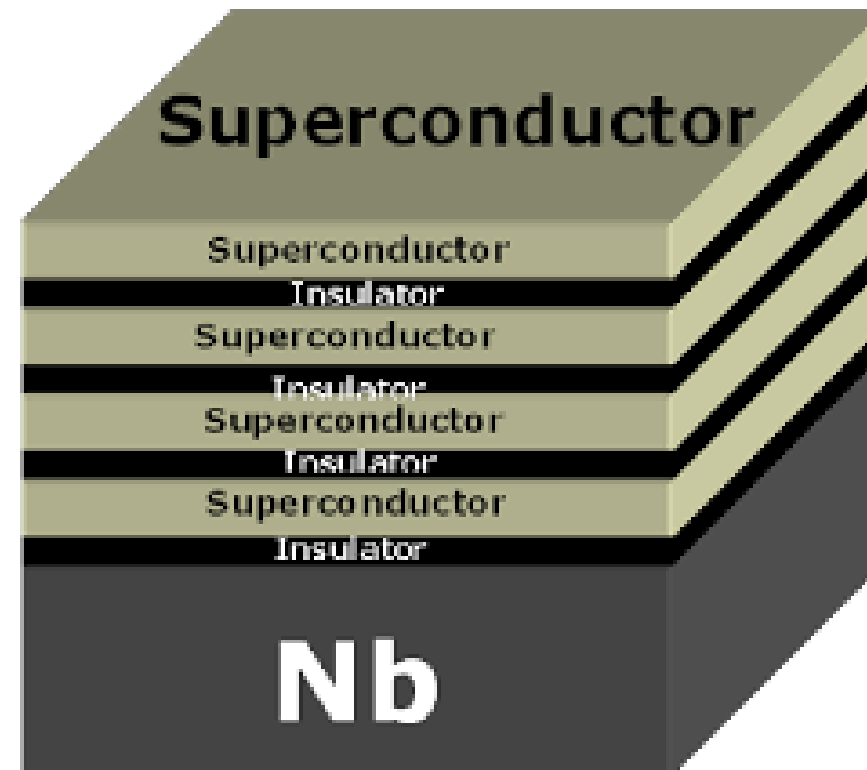
- ❖ Most compounds exhibit s-wave gaps
- ❖ Very sensitive to impurities content (either magnetic or not)

$\text{NaFe}_{1-x}\text{Co}_x\text{As}$ ($x = 0.0175$)
= ferromagnetic

$\text{NaFe}_{1-x}\text{Co}_x\text{As}$ ($x=0.045$)
= SC



MULTILAYERS



NANOCOMPOSITES MULTILAYERS

Taking advantage of the high T_c superconductors with much higher H_c without being penalized by their lower H_{c1} ...

Multilayer coating of SC cavities: alternating SC and insulating layers with $d_{SC} < \lambda$

Higher T_c thin layers provide magnetic screening of the Nb SC cavity (bulk or thick film) without vortex penetration



Structures proposed by A. Gurevich in 2006, SRF tailored

□ Dielectric layer

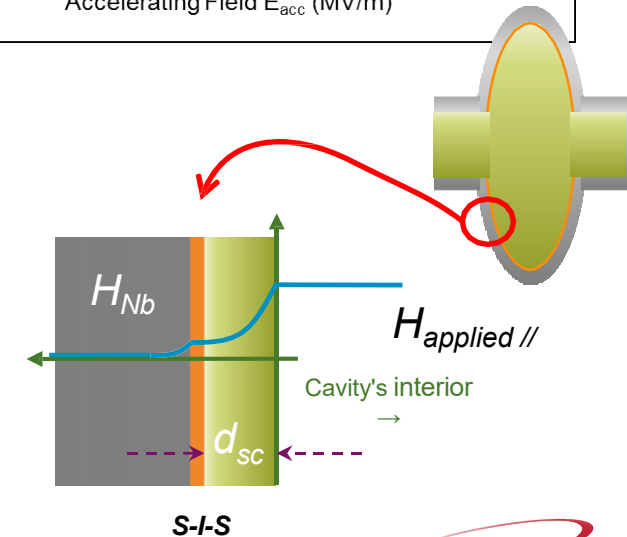
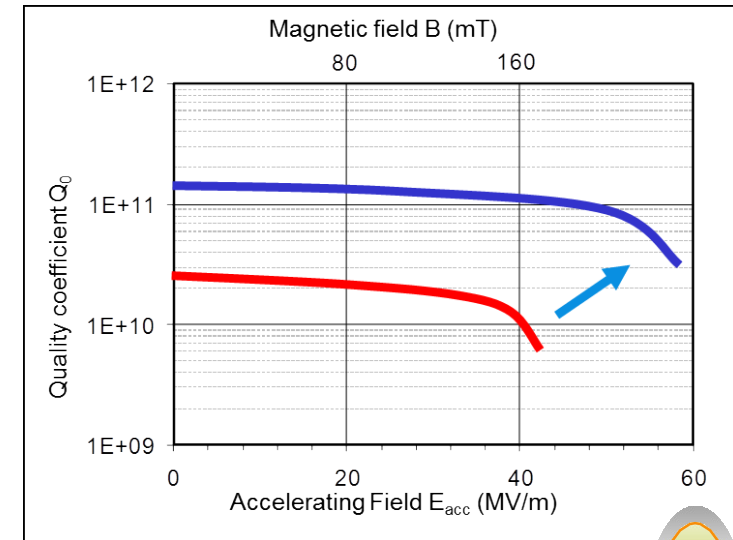
- Small \perp vortex (short \rightarrow low dissipation) Quickly coalesce (w. RF)
- Blocks avalanche penetration

\Rightarrow **Multilayer** concept for RF application

□ Nanometric S//S// layers deposited on Nb

- SC nanometric layers (≤ 100 nm) $\Rightarrow H_{c1} \uparrow \Rightarrow$ Vortex enter at higher field
- Nb surface screening \Rightarrow allows high magnetic field inside the cavity \Rightarrow higher E_{acc}

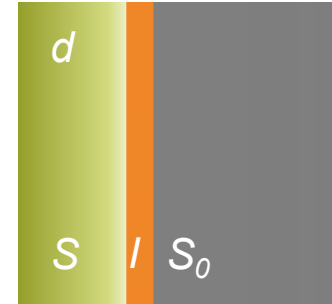
SC w. high T_c than Nb (e.g. NbN): $R_s^{NbN} \approx \frac{1}{10} R_s^{Nb}$
 $\Rightarrow Q_0^{multi} \gg Q_0^{Nb}$



FIRST APPROACH: TRILAYERS

■ Meissner state stable if:

- Screening current @ both SC surface is < depairing current
- $J(0) < J_d = H_s/\lambda$ and $J(d) < J_{d0} = H_{s0}/\lambda_0$
- If d is small, H_{SH}^S is high, but most of the field reach S_0
- If d is thicker, H_{SH}^S is lower, but screening is more effective



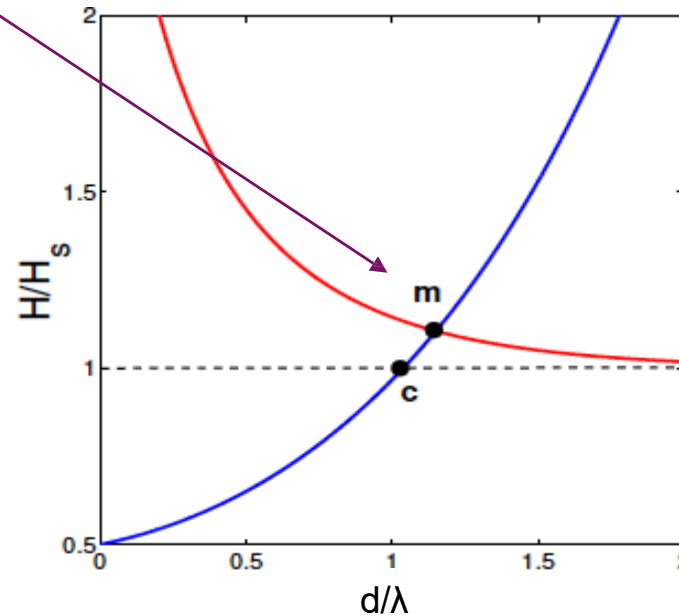
■ an optimum thickness and a maximum screening field!!!

$$\frac{(e^{2d/\lambda} - k)H}{e^{2d/\lambda} + k} \leq H_s, \quad \frac{H(1+k)e^{d/\lambda}}{e^{2d/\lambda} + k} \leq H_{s0}$$

$$d_m = \lambda \ln(\mu + \sqrt{\mu^2 + k}), \quad \mu = \frac{H_s \lambda}{(\lambda + \lambda_0) H_{s0}}$$

$$H_m = \left[H_s^2 + \left(1 - \frac{\lambda_0^2}{\lambda^2}\right) H_{s0}^2 \right]^{1/2}$$

Maximum screening field H_m at the optimum S thickness $d^S = d_m$

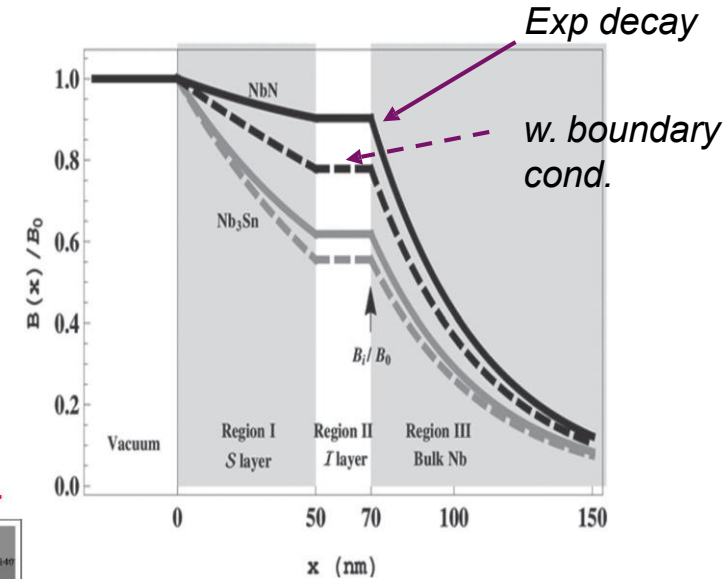


SIS OPTIMIZATION: IMPORTANCE OF MODELS

■ First approach: trilayers

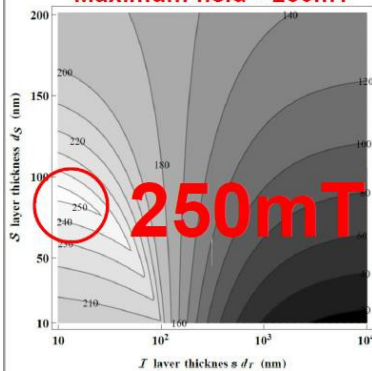
- Boundary conditions implemented (including effect of an insulating layer. finite thickness)
- H_{SH} determined initially in London approx., further improved w. quasiclassical theory (valid @ $T \ll T_C$)
- Initially assume “perfect conditions”
- (bulk values, field // to surface)

A. Gurevich, T. Kubo



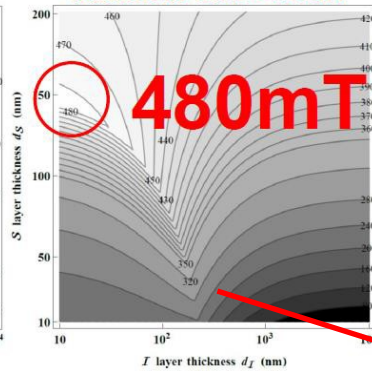
Dirty Nb // I / Nb

Optimum $d_s \sim 90\text{nm}$
Maximum field $\sim 250\text{mT}$



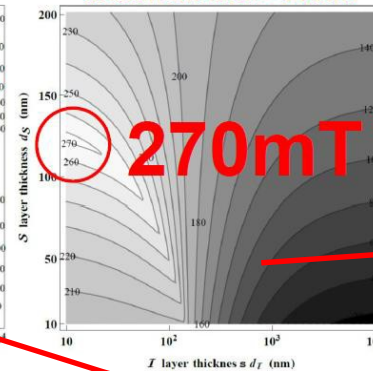
Nb_3Sn // I / Nb

Optimum $d_s \sim 150\text{nm}$
Maximum field $\sim 480\text{mT}$



NbN // I / Nb

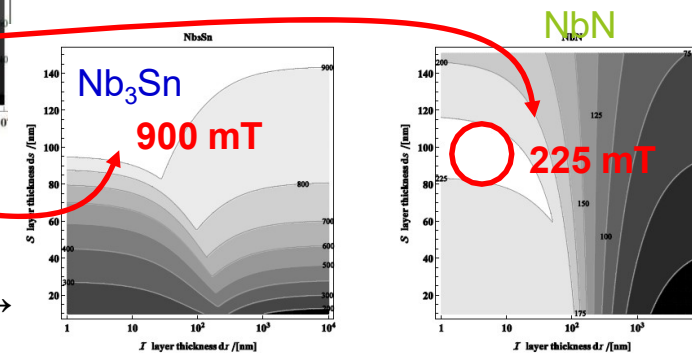
Optimum $d_s \sim 130\text{nm}$
Maximum field $\sim 270\text{mT}$



← Quasiclassical Approach,
Can be further improved...

[Kubo 2013-17]

Previous calculation w. London theory only: not realistic →



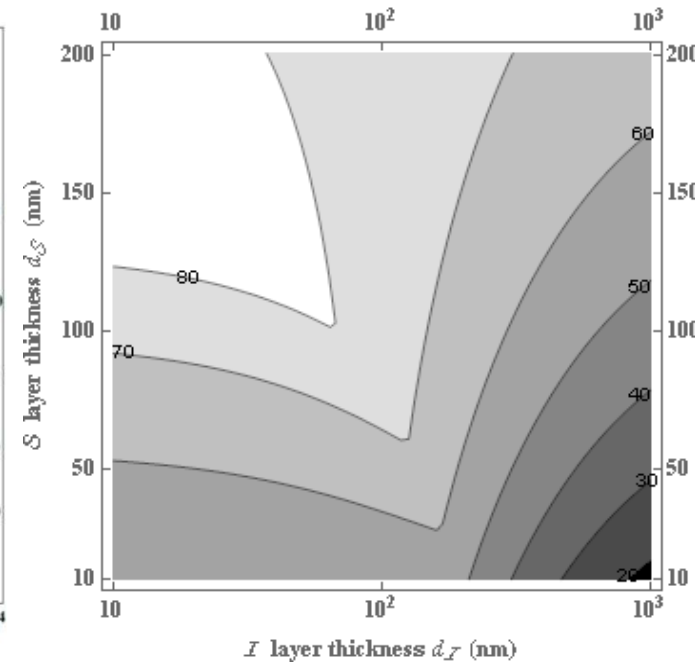
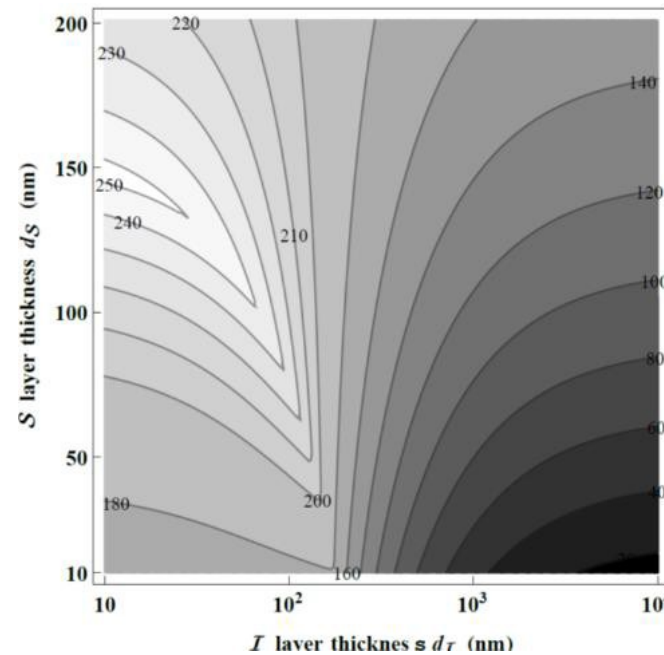
TRILAYER OPTIMIZATION (...)

[A. Gurevich, T. Kubo](#)

■ Go for realistic condition

- Layers present defects, non-negligible surface roughness, non-uniform thickness.
- $\rightarrow H_{SH}^S$ suppressed due to of the local screening current enhancement.
- Introducing material suppression factor $\eta = f(\text{defect size and aspect ratio, } \xi^S)$
- $\eta \sim 0.85$ for typical electropolished Nb surface)
- H_{SH}^{SIS} and optimal S layer thickness d_m^S can be determined w. surface topographical data

Ideal Nb substrate
with $B_{C1}=170$ mT



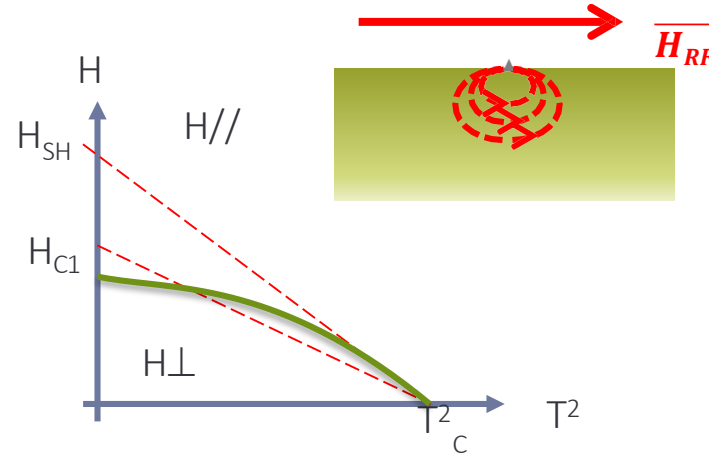
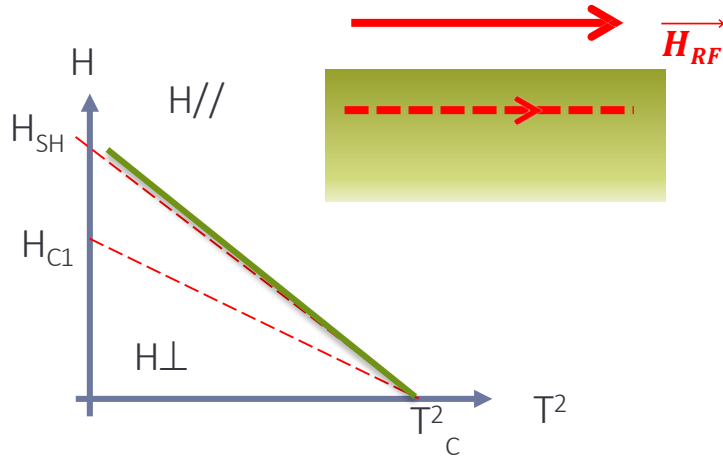
Nb with defects*,
with $B_{C1}=50$ mT

* e.g. morphologic
defects that allow earlier
vortex penetration

See exp proof later on

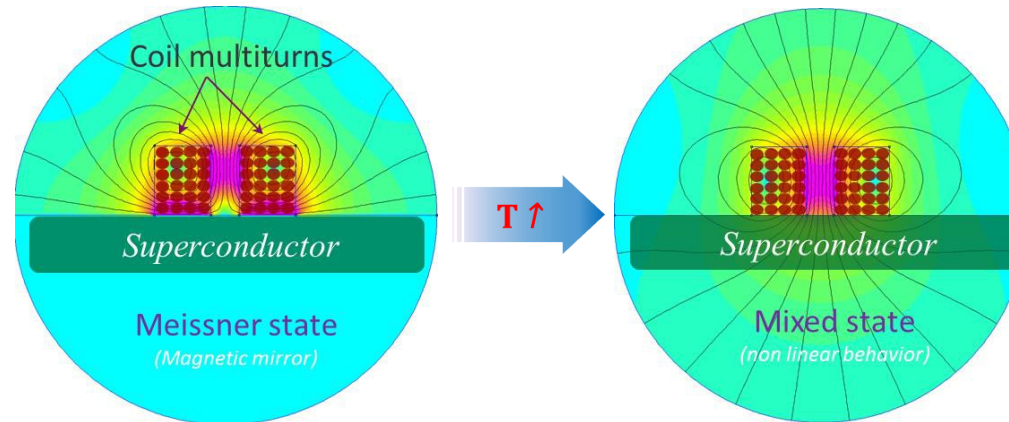
WHAT IS THE LIMIT ($H_{fp}/H_{C1}/H_{SH}$) ?

- Real world cavities behavior is dominated by a few number of defects
- It is very important to measure the penetration field of samples in realistic conditions



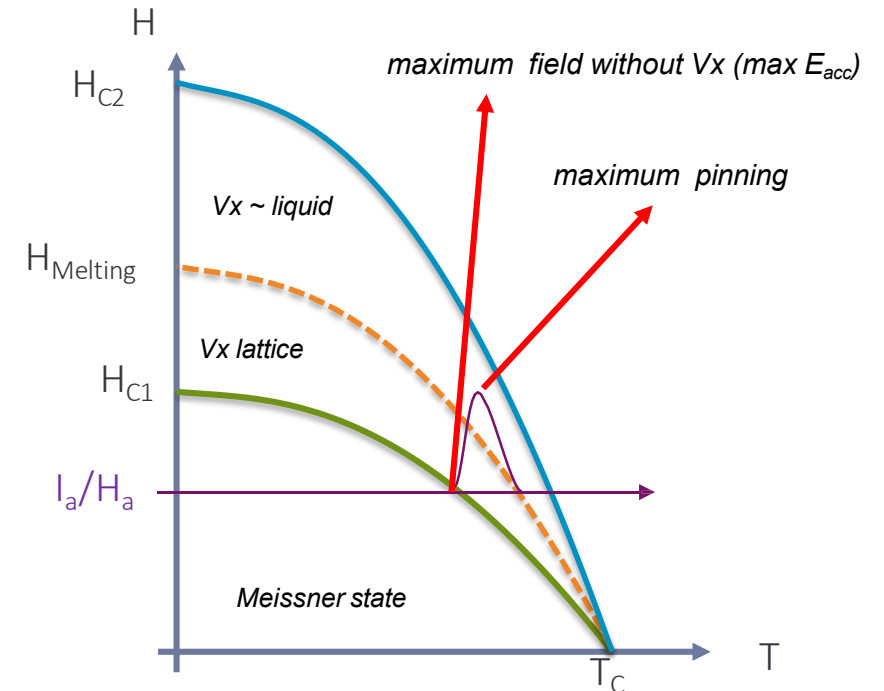
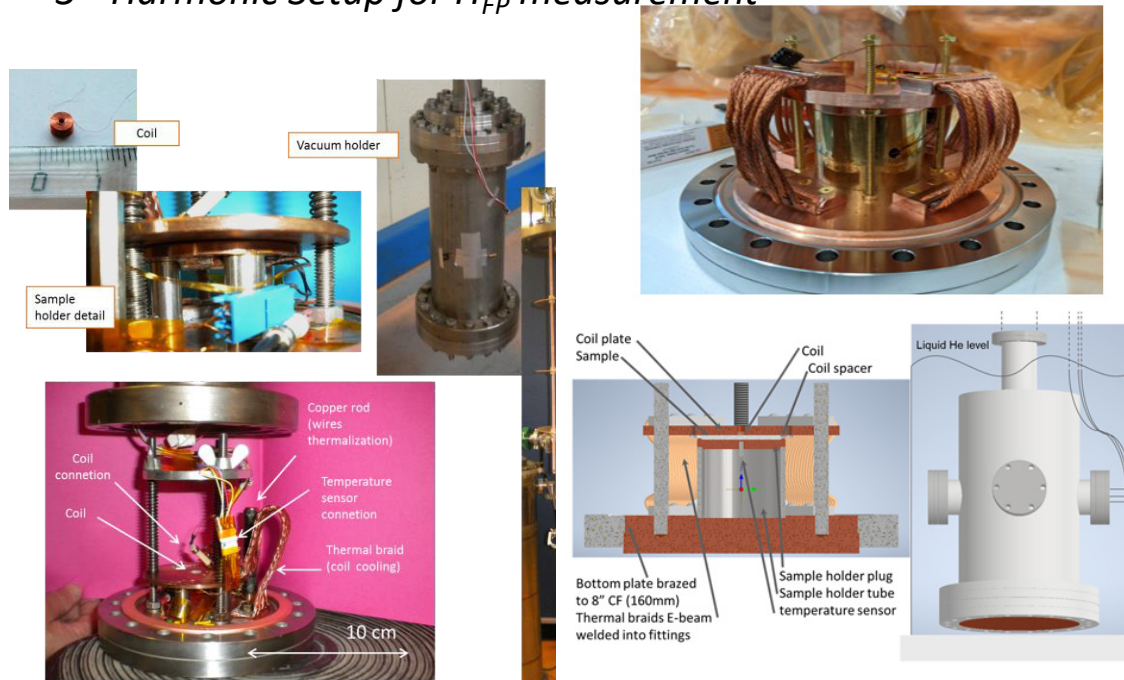
Local magnetometry

- ~ Same geometry as cavities
- No shape/edge effect (vs DC/ Squid magnetometry)
- No demagnetization effect
- Measures actual penetration field wherever it is $H_{fp}/H_{C1}/H_{SH}$



EXPERIMENTAL DETAILS

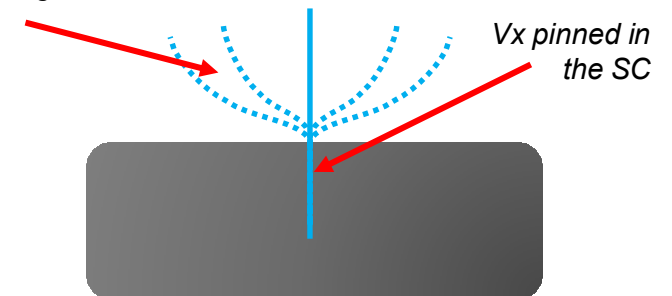
3rd Harmonic Setup for H_{FP} measurement



❑ Low frequency \equiv DC :

- ❑ $0 < H_a < H_{C1} \Rightarrow R=0$, Meissner state state
- ❑ $H_{C1} < H_0 < H_M \Rightarrow V_x$ are trapped, $R=0$, Campbell regime
- ❑ $H_M < H_a < H_{C2} \Rightarrow V_x$ are moving liquid like, $R \neq 0$, Flux flow regime
- ❑ Third harmonic signal arise from flux line tension (affects the e- inside the Cu coil),
- ❑ It does not depend on dissipation inside Nb, BUT depends on #
- ❑ of V_x trapped there (and length).

flux line moving in AC field



Nb – INSULATOR – NbN MODEL

NbN coating by Magnetron Sputtering

- **NbN single layers series**
 - NbN SL / “thick” Nb layer
 - Magnetron sputtered
 - MgO as dielectric layer
 - Far from perfect...



Nb (nm)	MgO (nm) Calc(actual)	NbN (nm) Calc(actual)	T _c (K)
250†	14	0	8.9
250†	14	25	15.5
500	10 (10.3)	50 (65)	15*
500	10 (8.4)	75 (72)	14.1*
500	10 (9.8)	100 (94)	14*
500	10	125	14.3*
500	10 (6.7)	150 (132)	15.9*
500	10 (10.4)	200 (164)	15*

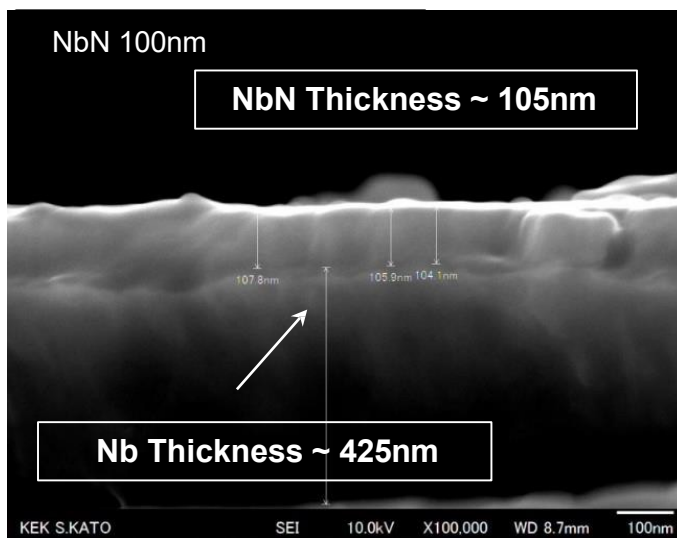
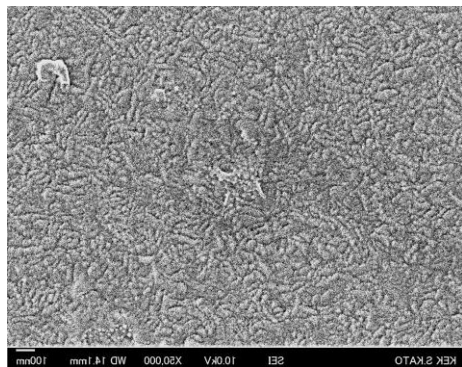
† Not same batch, deposited on the same conditions, but substrate = sapphire

*As determined with magnetometry, see below.

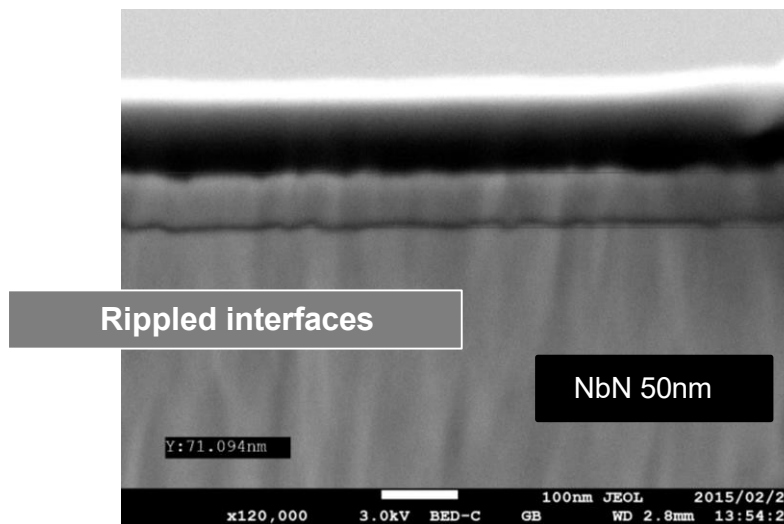
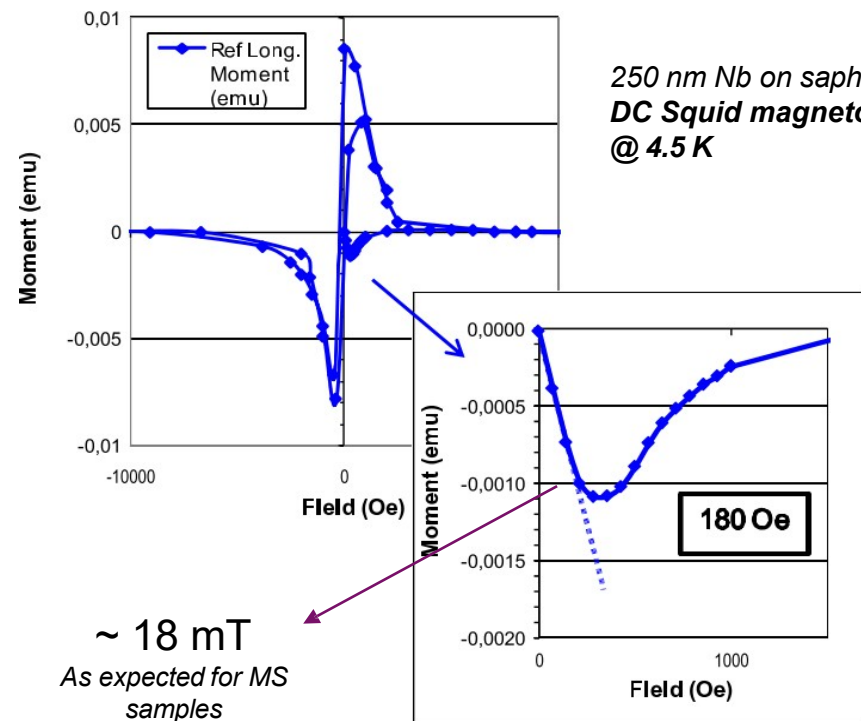
SPUTTERED MATERIALS...

Typical defects...

- Low H_{C1}
- Thickness \neq uniform
- ...

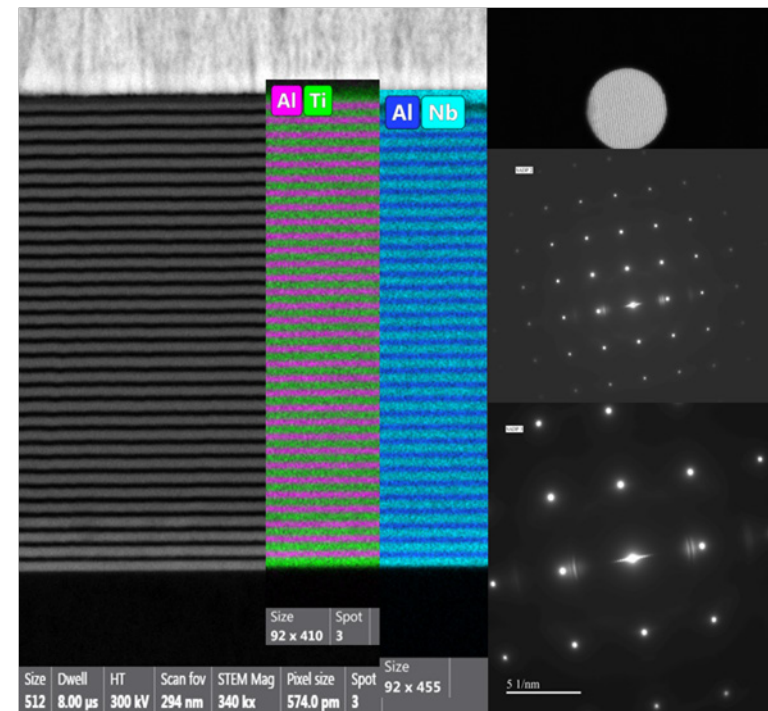


250 nm Nb on sapphire
DC Squid magnetometry
@ 4.5 K



BUT with careful methods:

$32 \times (\text{NbTiN}/\text{AlN})/\text{NbTiN}/\text{MgO}$

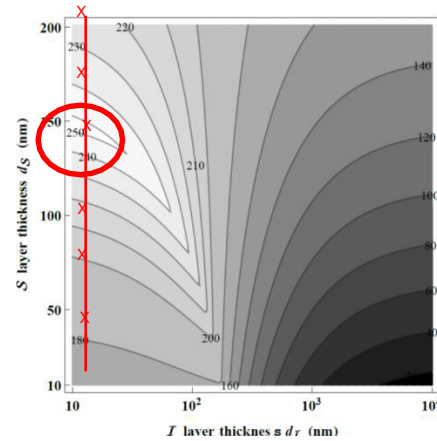


A-M Valente-Feliciano

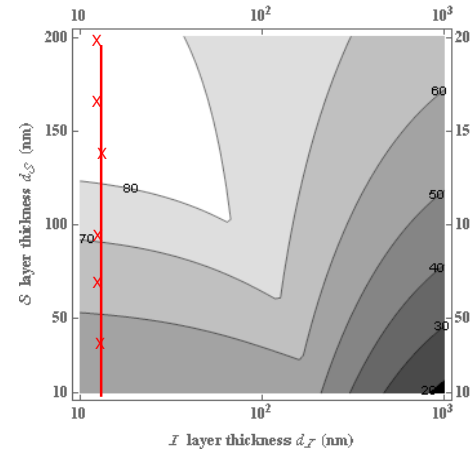
COMPARISON WITH THEORY

Theoretical predictions from T. Kubo (KEK)

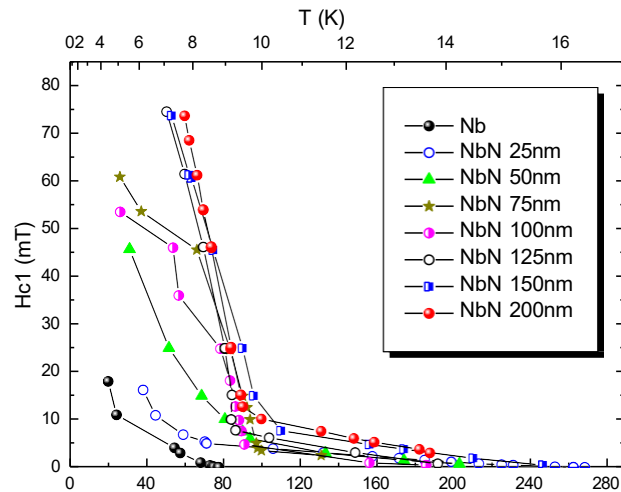
Ideal Nb substrate
with $B_{C1}=170$ mT



Nb with defects*,
with $B_{C1}=50$ mT



* e.g. morphologic
defects that allow earlier
vortex penetration See
SST paper cited earlier

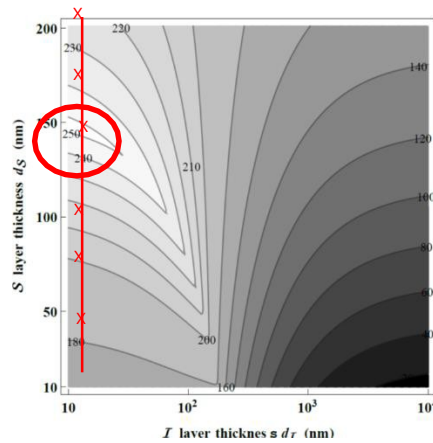


- The enhancement of the field penetration increases with thickness of NbN
- It reaches a saturation at thicknesses > 100 nm

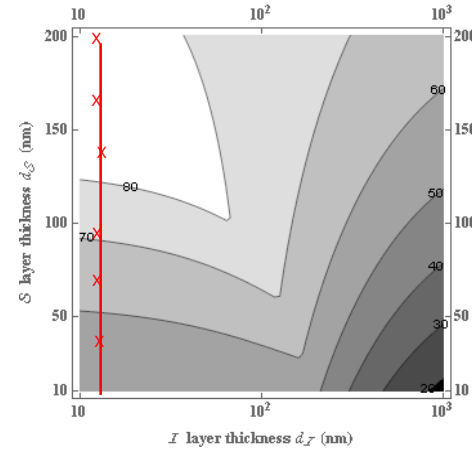
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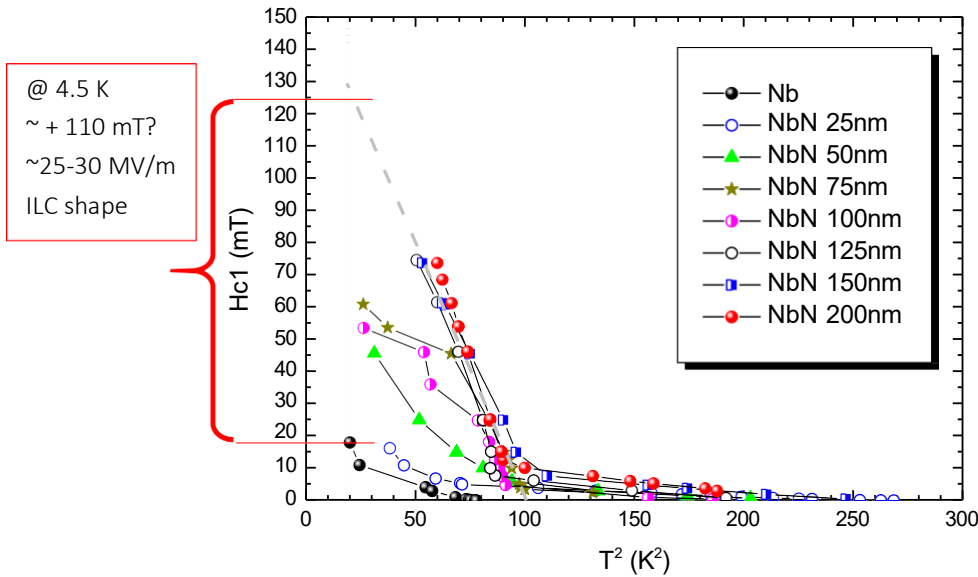
Ideal Nb substrate
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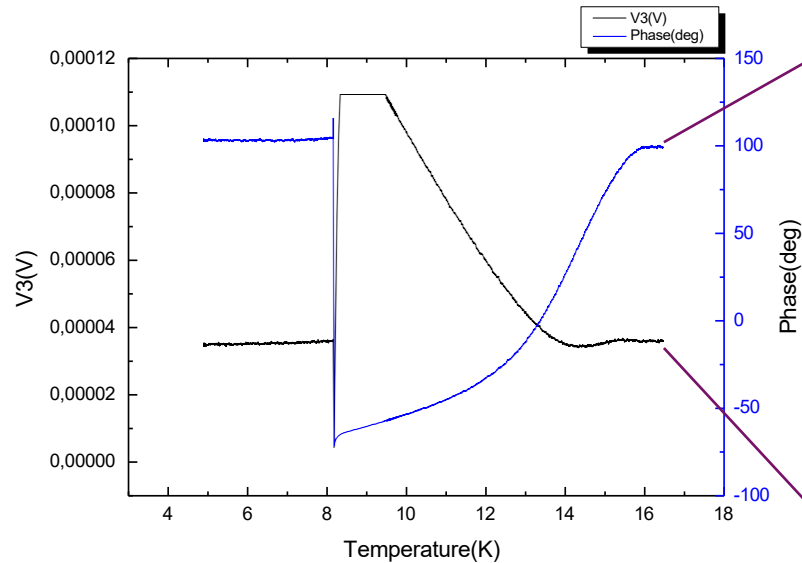
* e.g. morphologic
defects that allow earlier
vortex penetration See
SST paper cited earlier



- The enhancement of the field penetration increases with thickness of NbN
- It reaches a saturation at thicknesses > 100 nm

CLOSEUP OF 3rd HARMONIC SIGNAL

■ For a given H_{appl} , we observe 3 \neq transition temperatures

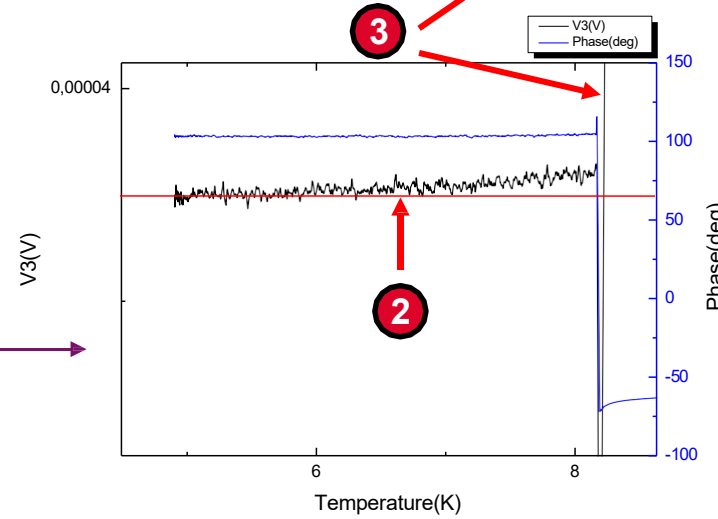
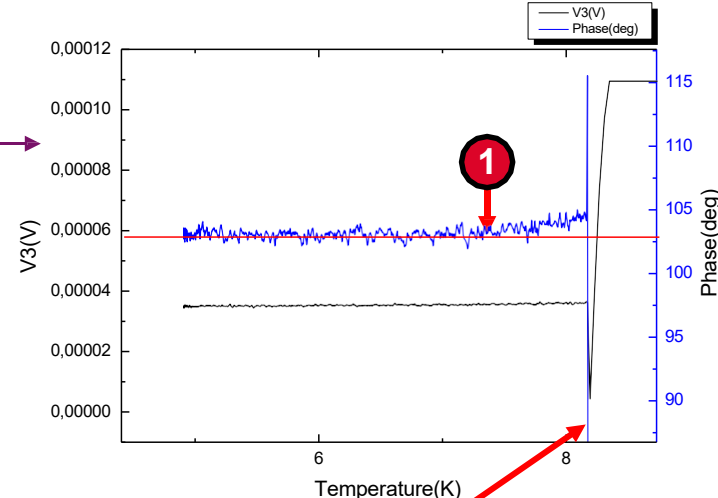


Phase signal

$T_1 \sim T_2$
 $T_3 \gg T_2$

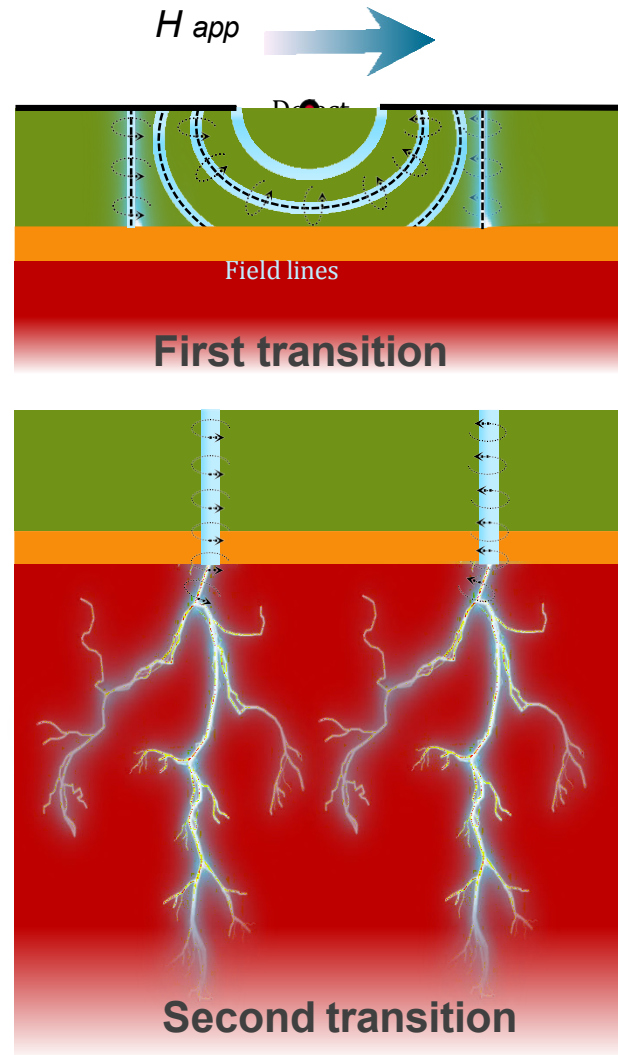
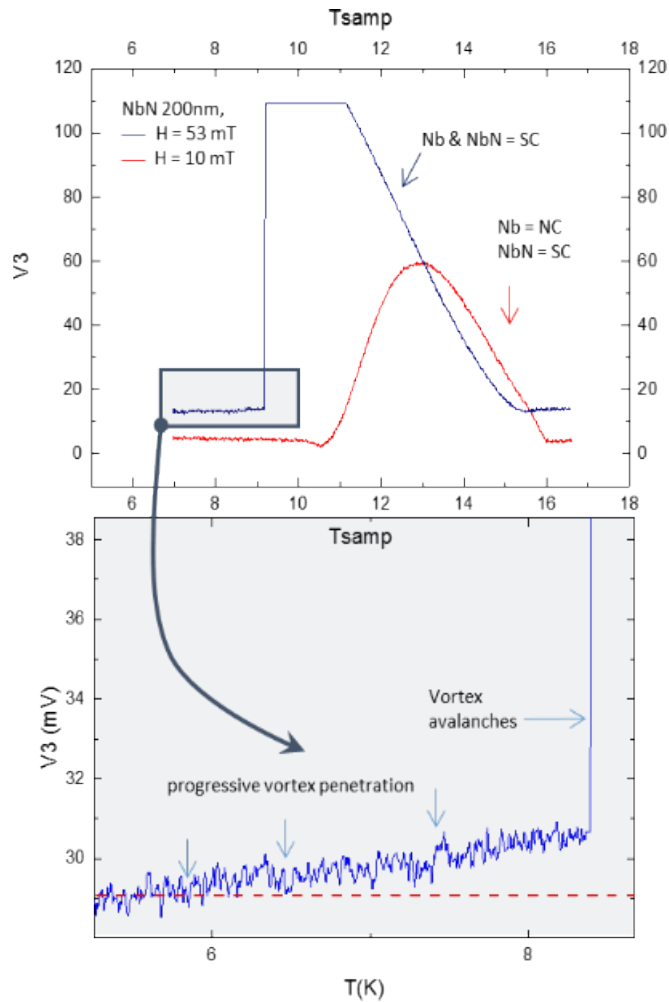
Voltage signal

■ $T_1 \sim T_2$: within noise level
■ $T_3 \gg T_2$: dramatic transition



ROLE OF THE DIELECTRIC LAYER !

Why do we have two transitions ?



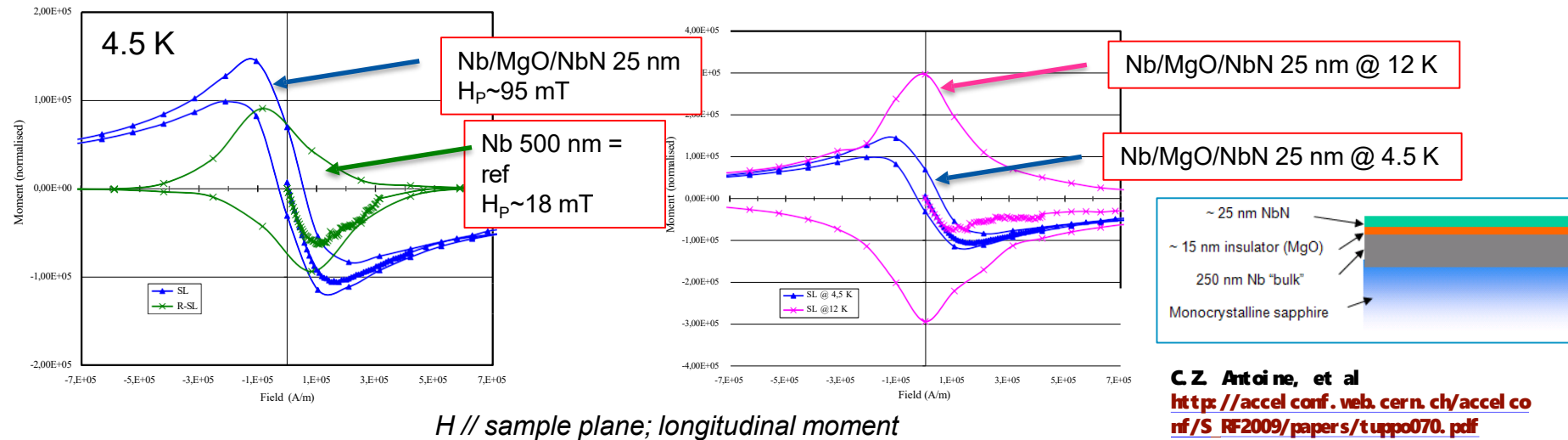
- Thin SC layer NbN
- Insulator MgO
- Thick SC layer Nb

$H \parallel$ surface \Rightarrow surface barrier
 A defect locally weakens the surface barrier
 1st transition, vortex
 blocked by the insulator
 ~ 100 nm \Rightarrow low dissipation.
 2nd transition, propagation of vortex
 avalanches (~ 100 μ m) \Rightarrow high dissipation.

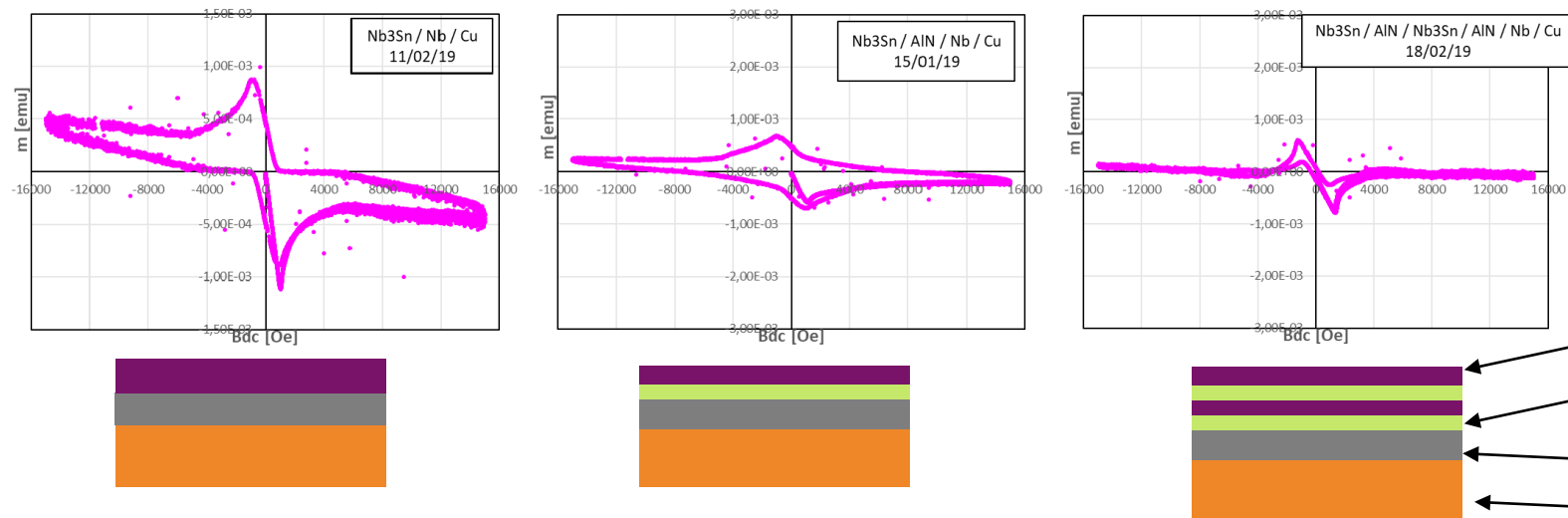
Dielectric layer = efficient protection !!!

†B. Bean and J. D. Livingston, *Phys. Rev. Lett.* 12, 14 (1964).

SIS : IRREVERSIBILITY => NO VORTEX PINNING <= NO VX ENTRY ?



Each individual layer : \exists defects, but combination: seems protected



[Courtesy R. Valizadeh and E. Seiler, 2019]

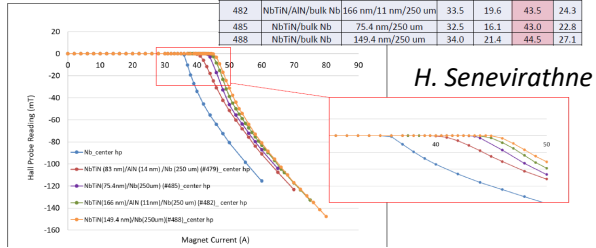
SIS : an intrinsically safe structure ?

SIS Multilayered Structures based on NbTiN

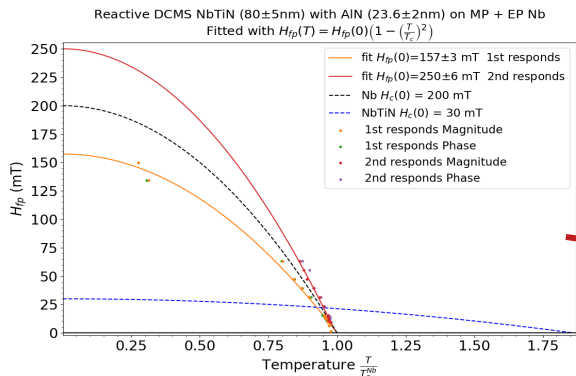
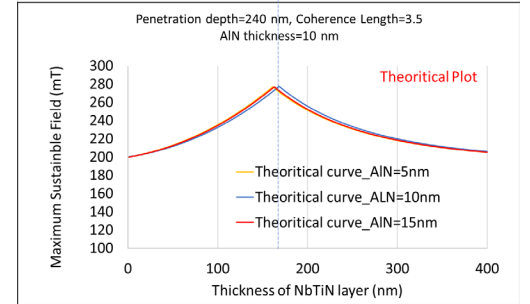
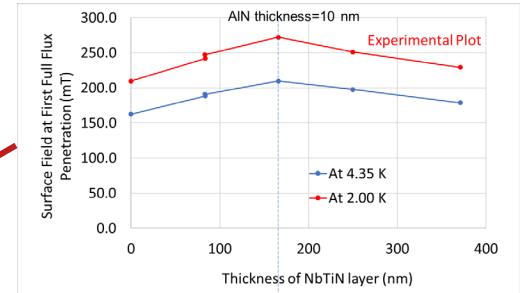
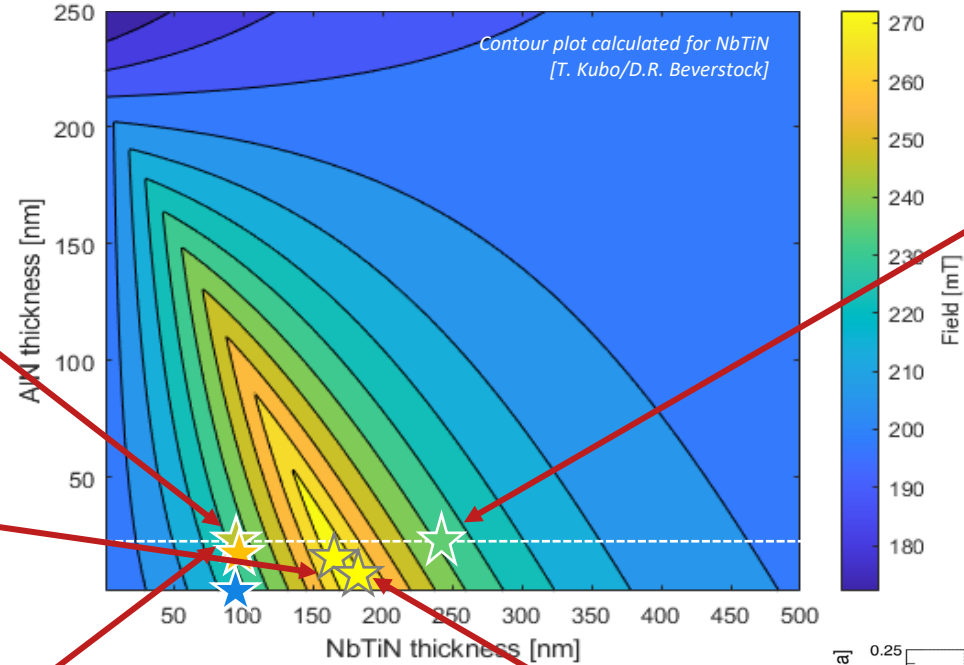
Measurement on NbTiN SIS structures on 1" & 2" Nb substrates

Center Probe at 2.00 K

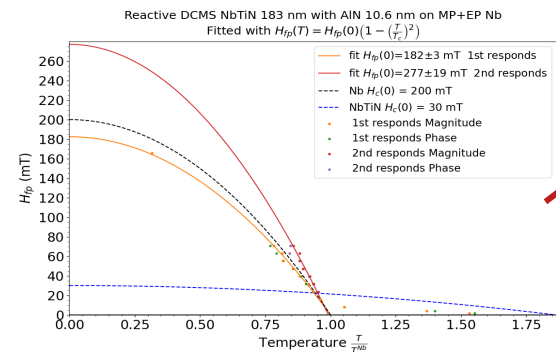
Sample #	Sample	Thickness	Current at first full penetration (A)			
			At the center		At the edge	
			4.35 K	%	2.00 K	%
3	Nb	250 um	28.0	-	35.0	-
479	NbTiN/AlN/bulk Nb	83nm/14 nm/250 um	30.0	7.1	39.5	12.8
482	NbTiN/AlN/bulk Nb	166 nm/11 nm/250 um	33.5	19.6	43.5	24.3
485	NbTiN/bulk Nb	75.4 nm/250 um	32.5	16.1	43.0	22.8
488	NbTiN/bulk Nb	149.4 nm/250 um	34.0	21.4	44.5	27.1



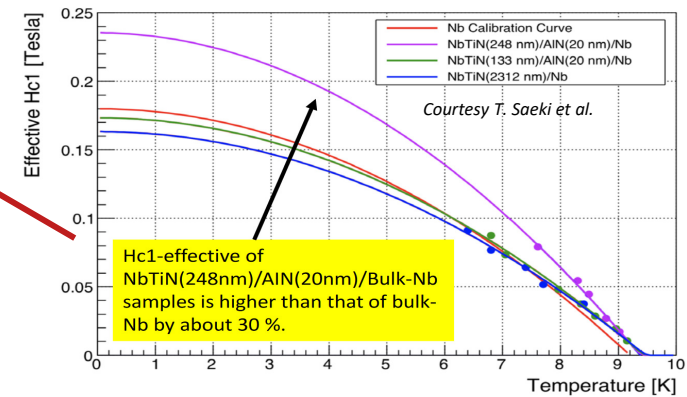
Penetration depth = 240 nm, Coherence length=3.5 nm



D.R. BEVERSTOCK

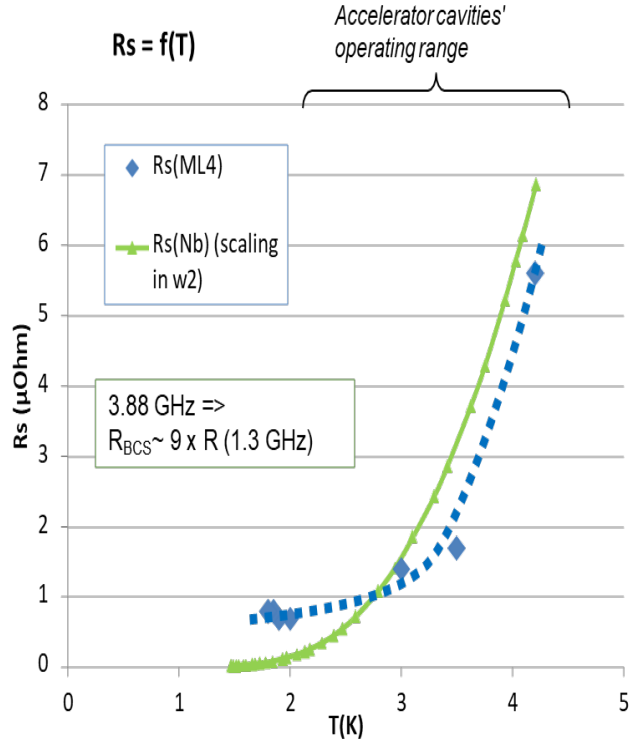
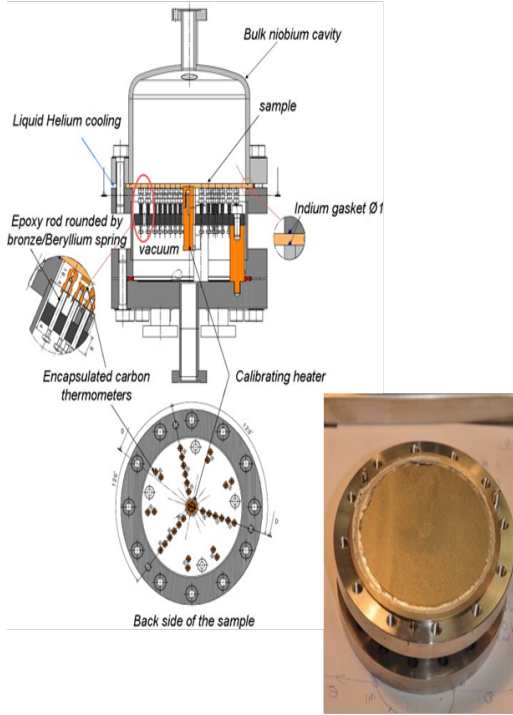


- ❑ Deposited SIS structures fit the theoretical model
- ❑ 3rd harmonic measurements show field enhancement up to 20-60% compared to base bulk Nb.
- ❑ Effect most sensitive to coherence length ξ



1st RF TESTS

1st TEST RF @ 3.88 GHz (4x 25nm NbN LAYERS)

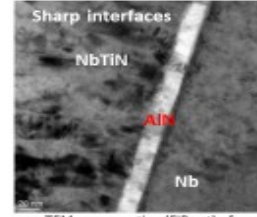


- Comparison is done with a high performance 1.3 GHz Nb cavity (scaling in ω^2)
- Indium gasket presents some defects measured with thermometric map => extra RF losses
- Residual resistance comes from NbN + bulk Nb substrate + indium gasket. Further investigations needed.

NbTiN/AlN/Nb structures - RF characterization

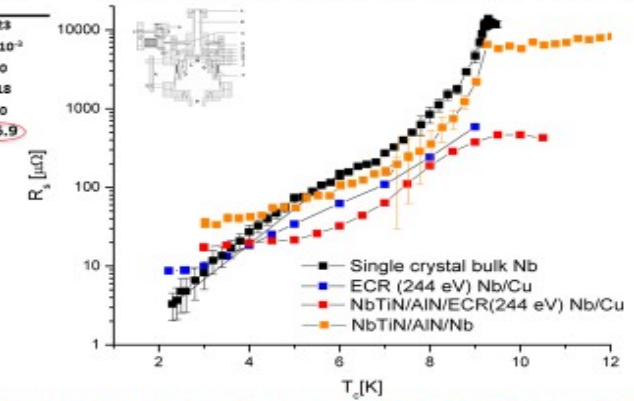
SIS structures coated on ECR Nb/Cu film and bulk Nb: 24h-bake, coating and annealing for 4 h at 450°C.

	AlN	NbTiN
N ₂ /Ar	0.33	0.23
Total pressure [Torr]	2x10 ⁻³	2x10 ⁻²
Sputtering Power [W]	100	300
Deposition rate [nm/min]	~ 2.5	~ 18
Thickness [nm]	20	150
T _c [K]	N/A	16.9



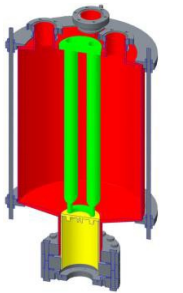
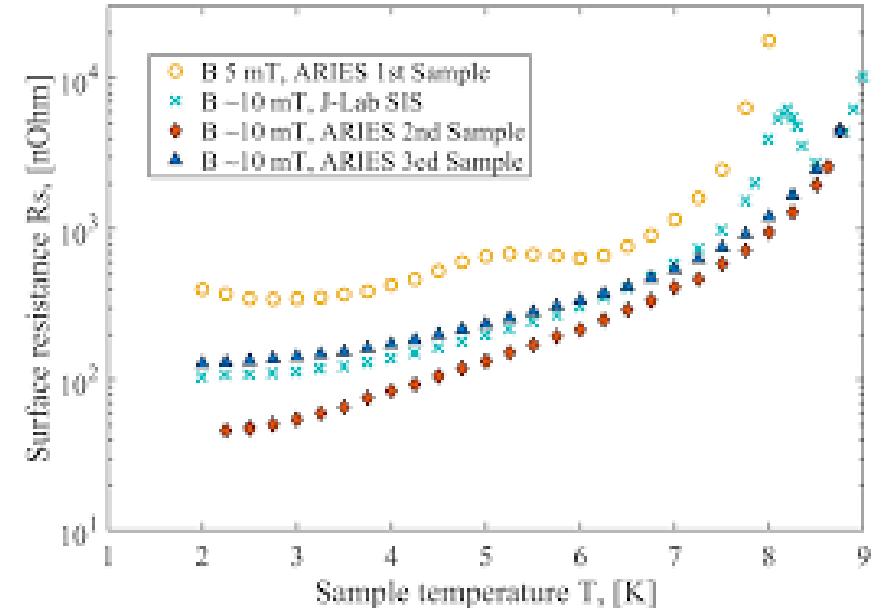
Lower BCS resistance beyond 4 K for SIS coated surfaces compared to standalone ECR film & bulk SC Nb.

RF Measurement in 7.5 GHz sapphire-loaded TE₀₁₁ cavity



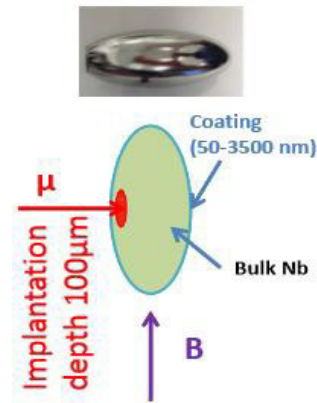
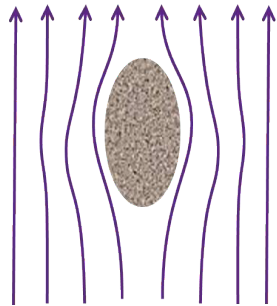
A. M. Valente-Falliciano - SRF Materials other than bulk Niobium

Jefferson Lab



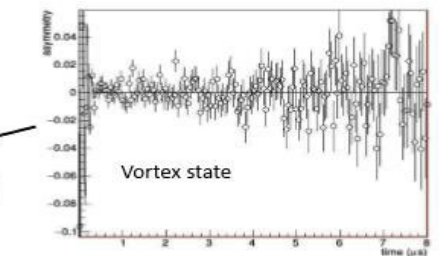
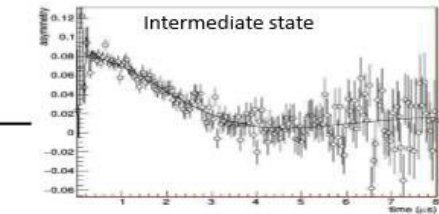
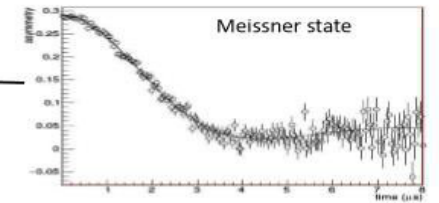
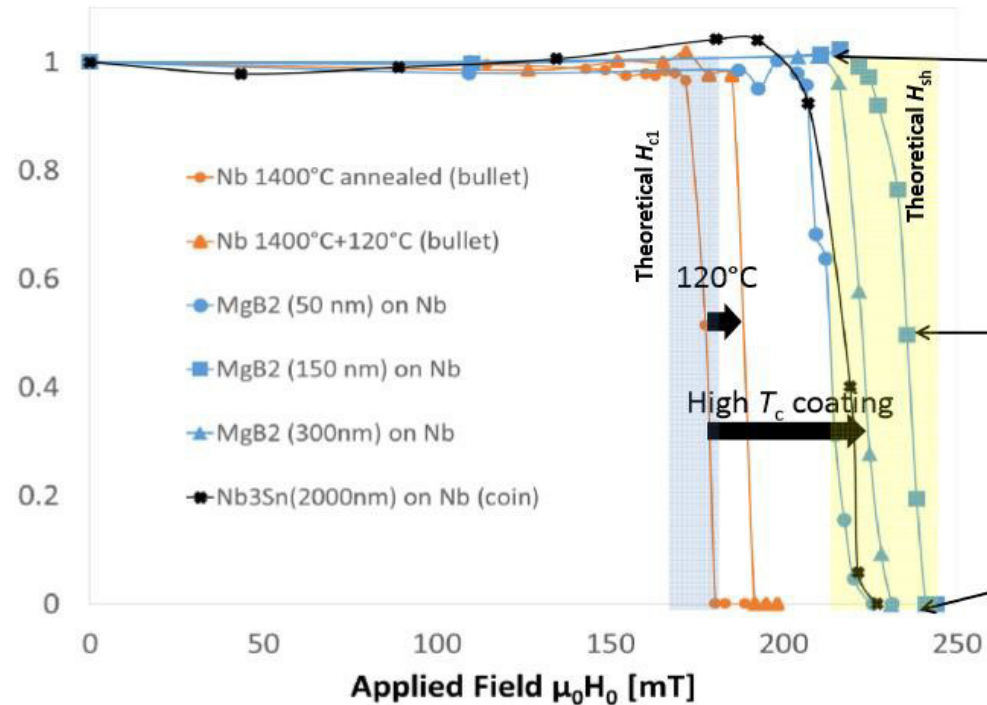
ML WITHOUT DIELECTRIC INTERLAYER

μ -SR



Field of first flux entry measurements

Volume fraction in the Meissner state at 0K



[Junginger, SRF 2017]

<https://arxiv.org/abs/1705.06383>

The SS boundary provides an additional barrier to prevent penetration of vortices. It would not be as robust as the I layer of the SIS structure, but it also contributes to pushing up the onset of vortex penetration.

[Kubo, SST]

Wrapping Up...

WHAT IS A GOOD THIN FILM SUPERCONDUCTOR ?

❑ A compromise between:

- ❖ High superconducting/RF performance (High T_c , High superheating field)
- ❖ Easy fabrication process,
 - ≡ high reproducibility at “industrial scale”
 - Easy process to go from 1-cell to multi-cells or complex shapes
 - Easy process to adapt to various frequencies
- ❖ Tunability
 - Beware of brittle materials !
- ❖ Low sensitivity to trapped flux upon cooling down
 - Few crystalline defects or a structure not too sensitive to them (e.g. SIS)

❑ Thin films on copper = route to help cost savings

- ❖ Cheaper manufacturing
- ❖ Higher operation temperature
- ❖ Higher gradients : lower capital costs

COST SAVINGS beyond bulk Nb : Construction & Operations

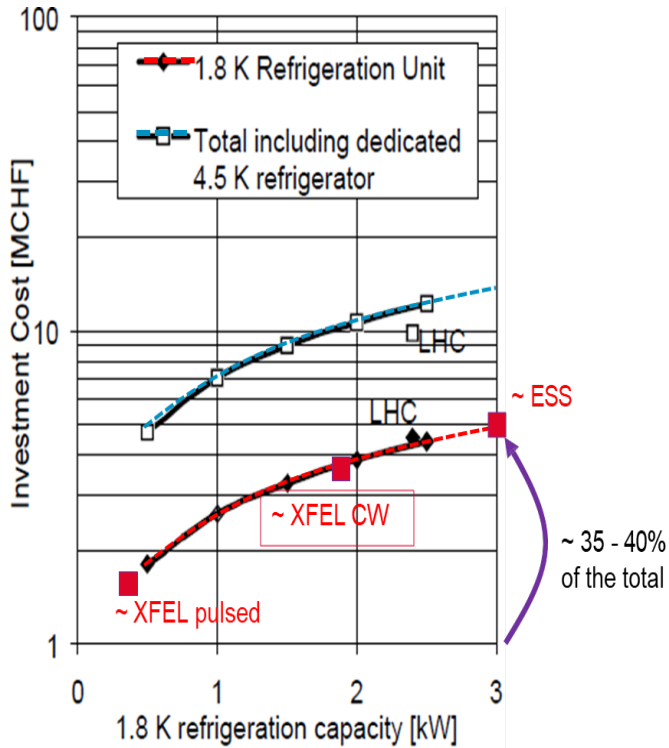


Figure 9. Investment cost of 1.8 K refrigeration

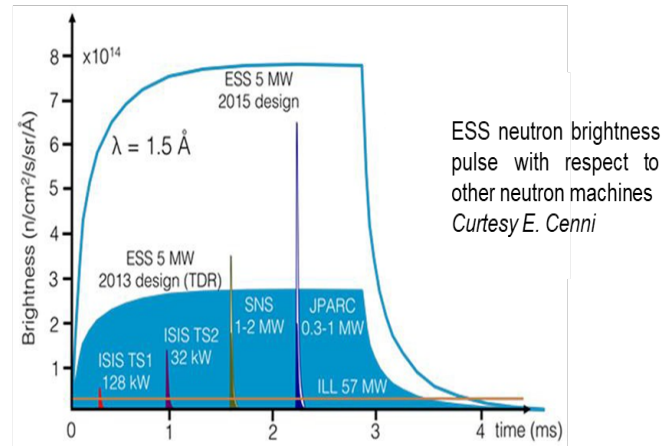
Courtesy P. Lebrun

Courtesy C. Antoine, IPAC 2023

- **LHC project report 317** (S. Claudet, Ph. Gayet, Ph. Lebrun, L. Taviani and U. Wagner, "Economics of large helium cryogenic systems: experience from recent projects at CERN")

- **4.5 K instead of 2 K: investment decreased by ~35-40 % !!!**

- e.g. ESS cryogenic total cost ~40 M€
- ~ 15 M€ savings ?



- **Carnot efficiency η_c** (thermodynamics)
- **Refrigerator efficiency η_{Th}** (real life compared to physics)

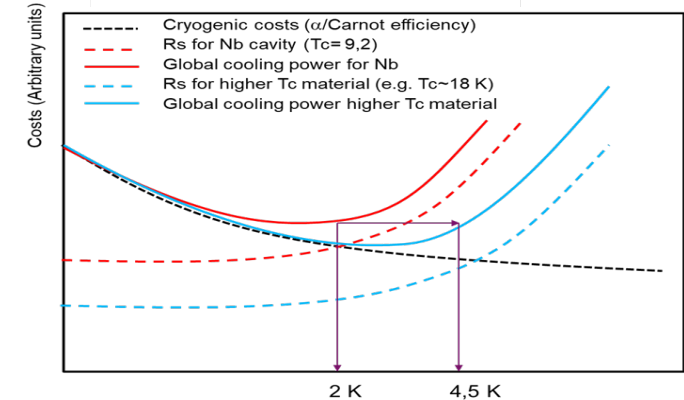
$$\eta_c = \frac{T_c}{T_h - T_c} \approx \begin{cases} 1/70 & \text{for } T_h = 300 \text{ K}, T_c = 4.2 \text{ K} \\ 1/150 & \text{for } T_h = 300 \text{ K}, T_c = 2 \text{ K} \end{cases}$$

$$\eta_{th} = \begin{cases} 25 - 30 \% & \text{at } T = 4.2 \text{ K} \\ 15 - 20 \% & \text{at } T = 2 \text{ K} \end{cases}$$

- To remove 1W @ 80K: ~20W @ 300K is needed
- To remove 1W @ 4.2K: ~250W @ 300K is needed
- To remove 1W @ 2K: ~750W @ 300K is needed

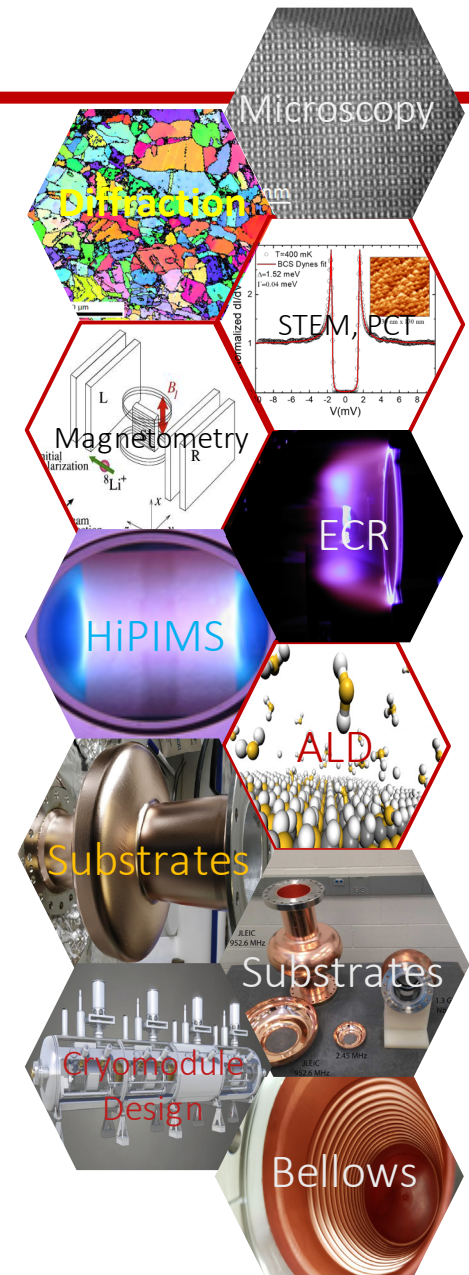
- **RF surface resistance**

$$R_s = R_0 + \frac{A\omega^2}{T} e^{-BT_c/T}$$



- **Higher Tc materials:**
 - Same cooling power @ 4.5 K instead of 2K
 - Or: lower cooling power at 2 K
- **4.5 K instead of 2 K: plug power divided /3 !!!**
 - Less risks of He pollution
 - Easier maintenance...

Path Forward



- **Theoretical and material studies** to gain in-depth understanding of the fundamental limitations of thin film superconductors under radio-frequency fields
- **Advanced coating technology** for Nb/Cu and alternative materials, Nb₃Sn, V₃Si, NbTiN ...
 - Energetic condensation (electron cyclotron resonance (ECR), HiPIMS, kick positive pulse...)
 - Atomic Layer Deposition (ALD)
 - Hybrid deposition techniques
- **Cavity deposition techniques for development of superconductor-insulator-superconductor (SIS) nanometric layers** to further enhance the performance of bulk Nb and Nb/Cu
- **Improved cavity fabrication & preparation techniques**
 - electroforming, spinning, hydroforming, electro-hydro forming, 3D additive manufacturing
 - environmentally friendly electropolishing, diamond cutting, nano-polishing, plasma etching ...)
- **Cryomodule design** optimization
- Improvement of **accelerator ancillaries** with advanced deposition techniques
 - HiPIMS Cu coated bellows, power couplers...

TAKE HOME MESSAGE

- ❑ Superconducting cavities are dominated by their surface quality (Niobium AND other SC !)
- ❑ Niobium is close to its ultimate limits, but can be surface engineered (doping)
- ❑ H_{SH} difficult to reach in real “accelerator cavities” (low T, large scale cavity fabrication, surface defects,...)
- ❑ Renewed activity on bulk-like Nb films (cost issues) and high H_{SH} SC e.g. Nb_3Sn or $NbTiN$ (higher performances)
- ❑ SIS structures seem to be a promising way to go toward realistic complex materials (+ Nb cavity upgrade)
- ❑ Look for higher Q_0 , not only E_{acc} !
- ❑ **A TECHNOLOGICAL REVOLUTION FOR SRF CAVITIES IS WITHIN REACH!**

- **Aknowledgement**

Inspiration and material from earlier lectures from:

Prof. V. Palmieri, INFN, Dr. C. Antoine, CEA Saclay; Prof. A. Gurevich, ODU; ...

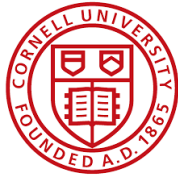
- **Next TFSRF Workshop (11th)**

September 2024

In PARIS Area, France



Acknowledgement



PAUL SCHERRER INSTITUT



Institut national de physique nucléaire et de physique des particules



SRf Tutorials 2023 - Beyond Bulk Nb

