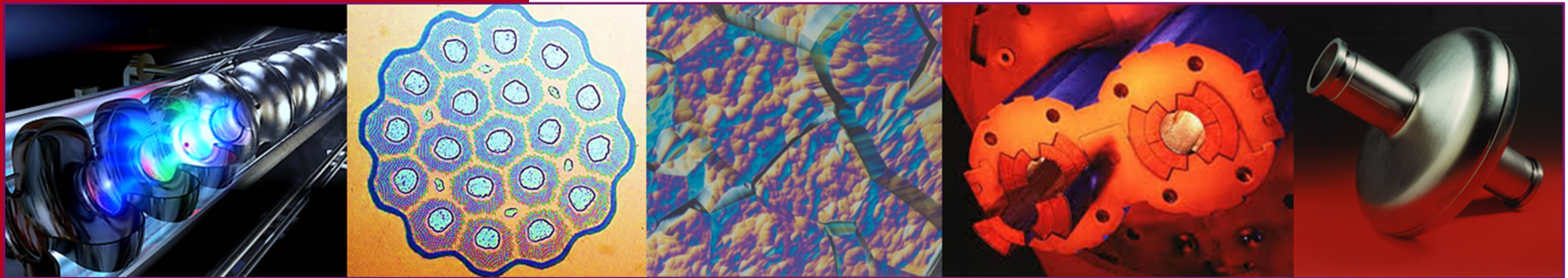


DE LA RECHERCHE À L'INDUSTRIE



MATERIALS FOR SUPERCONDUCTING ACCELERATORS: BEYOND BULK Nb



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SRF 2019 Tutorials

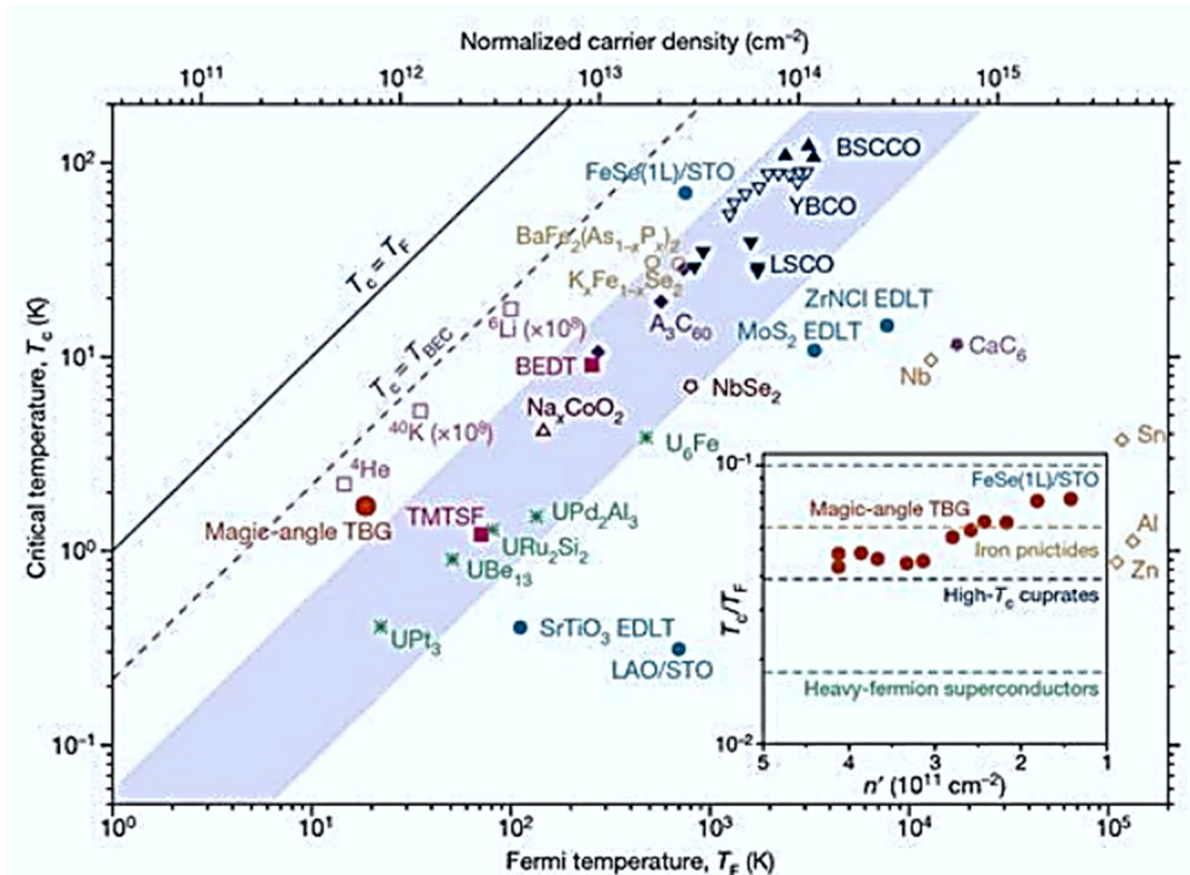
C.Z. Antoine

+ some material gathered by A.M. Valante-Feliciano



CHOICE CRITERIA ?

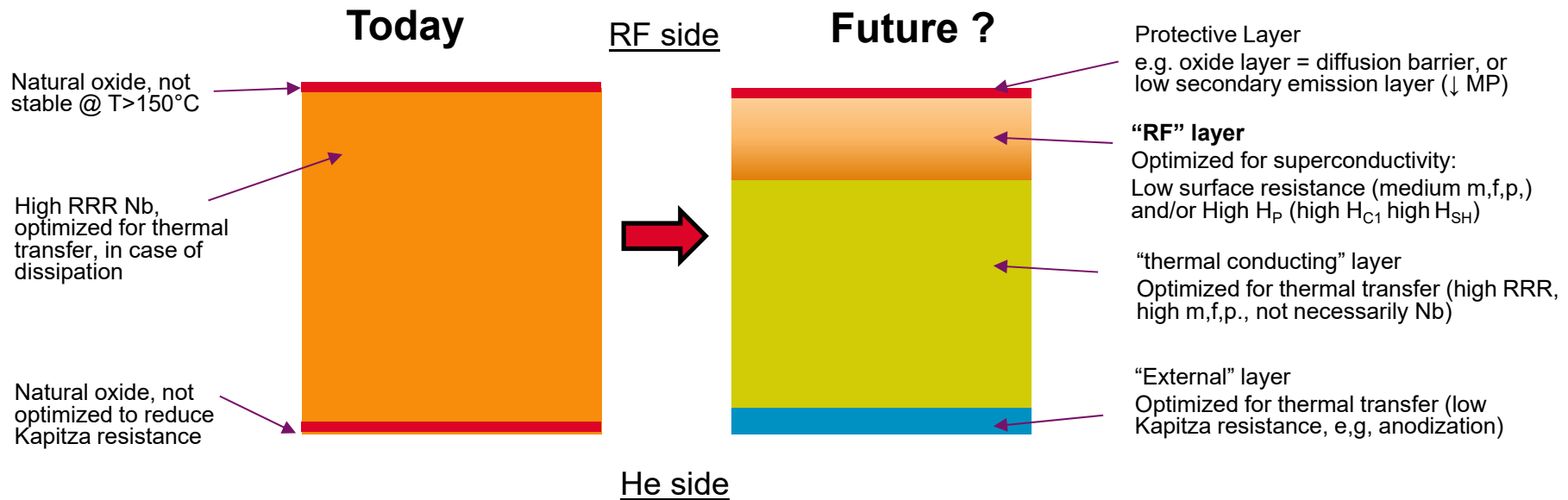
Thousands of SC exist, ~10 are currently used for applications, only bulk Nb works well for SRF !!!



- ✱ Heavy-fermion superconductors
- ▼ Cuprates
- Iron pnictides
- ◇ Conventional superconductors
- BEC in atoms *Bose Einstein Condensation*
- Two-dimensional materials
- Organic superconductors
- ◆ A_3C_{60}
- $NbSe_2$
- △ Na_xCoO_2
- CaC_6
- Magic-angle TBG *Twisted Bilayer Graphene*

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

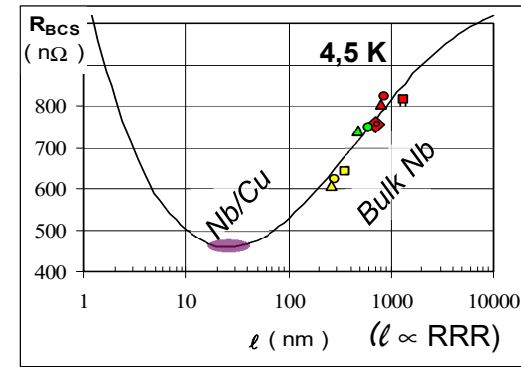
High RRR not required for superconductivity
but for thermal stabilization in case of defects



■ “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



Niobium superconducting radiofrequency cavities

■ Performances

- $E_{acc} \propto H_{RF}$
- $Q_0 (\propto 1/R_S) \propto T_C \Rightarrow \text{Nb}_3\text{Sn, MgB}_2, \text{NbN... (but not YBCO)}$
- Limit = magnetic transition of the SC material @ H_{peak}

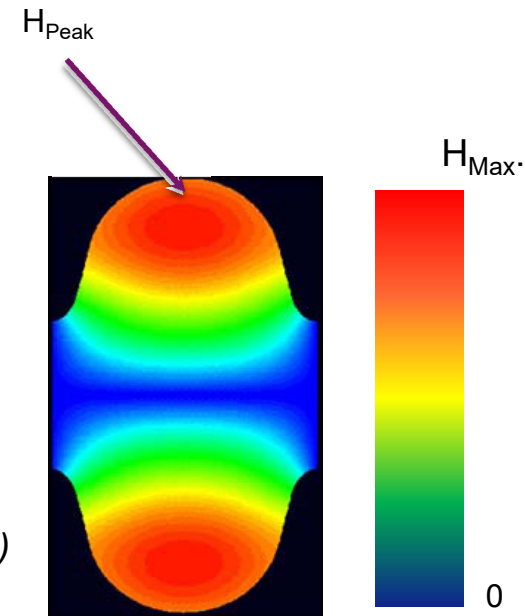
■ Superconductivity only needed inside :

- Thickness $\sim 10 \lambda$, $\sim < 1 \mu\text{m} \Rightarrow$ 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 !)

■ SC phase diagram

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

- Vortices dissipate in RF !

■ SRF $\Rightarrow$ 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:

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

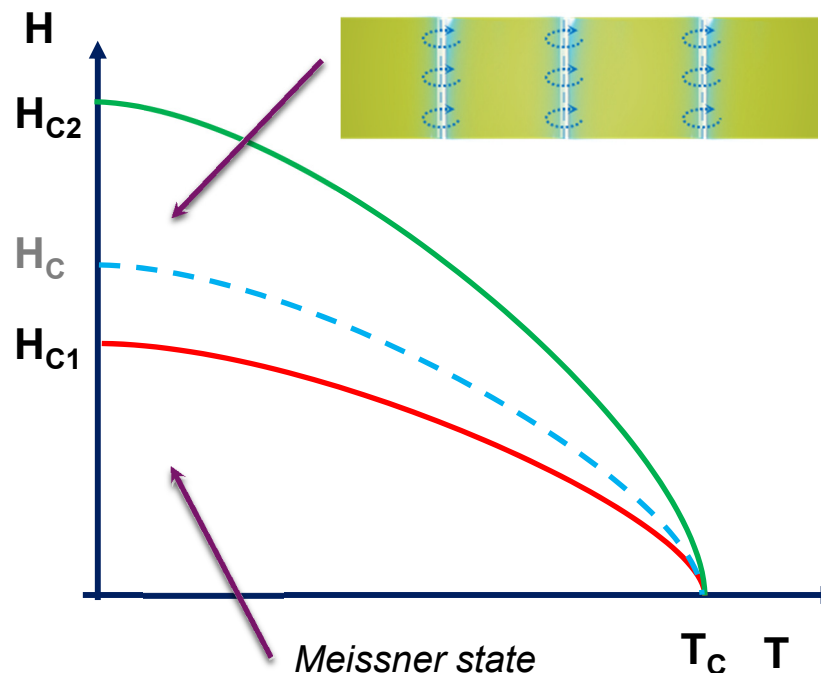
■ High T_c is better

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

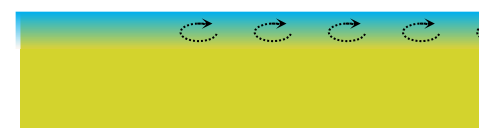
■ Metallic character in NC state is better (ρ_n)

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

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

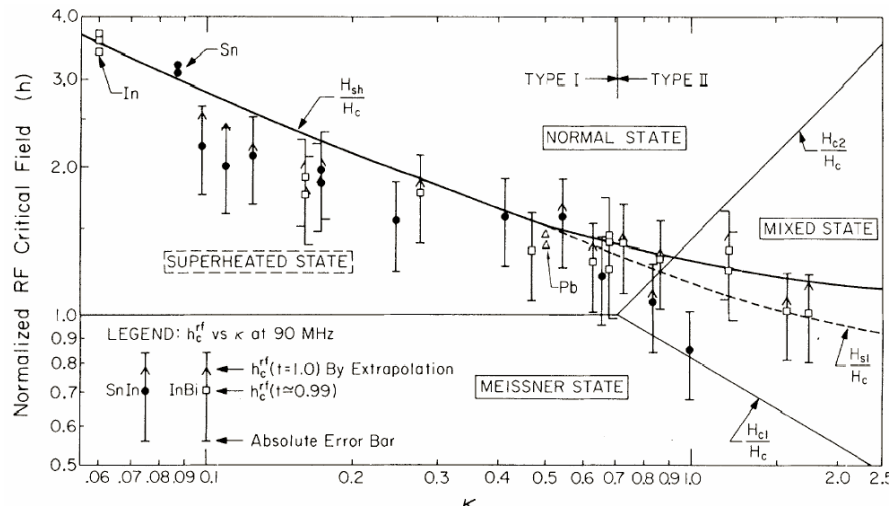


Screening current over λ ,
no magnetic field deeper



Metastable Meissner state above H_{C1}

observed close to T_c in DC/AC



Physical Origin

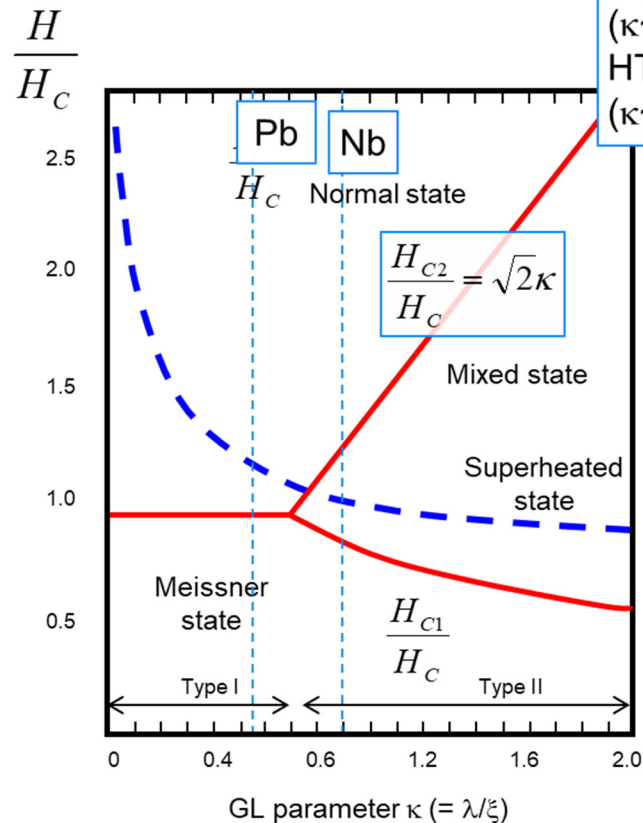
normal zone nucleation $\sim 10^{-6}$ s *

RF $\sim 10^{-9}$ s *

but in fact

1! vortex penetration $\sim 10^{-13}$ s **

SH state favored by H// surface (BL barrier) ***



→
MgB₂, NbTi
($\kappa \sim 40-50$)
HTc
($\kappa \sim 60-2000$)

$H_{SH} \sim 1,2.H_c$ pour $\kappa \sim 1$
 $H_{SH} \sim 0,75.H_c$ pour $\kappa \gg 1$
(Thermodynamics)

* H. Padamsee, J. Knobloch, and T. Hays, "RF superconductivity for accelerators". 1998: J. Wiley & son.

** Gurevich, Brandt, Smethna...

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

SUPERCONDUCTORS FOR SRF ?



Material	T_C (K)	ρ_n ($\mu\Omega\text{cm}$)	$\mu_0 H_{C1}$ (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	9,22	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				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 ?

Surface barrier

(Bean & Livingston, 1964)

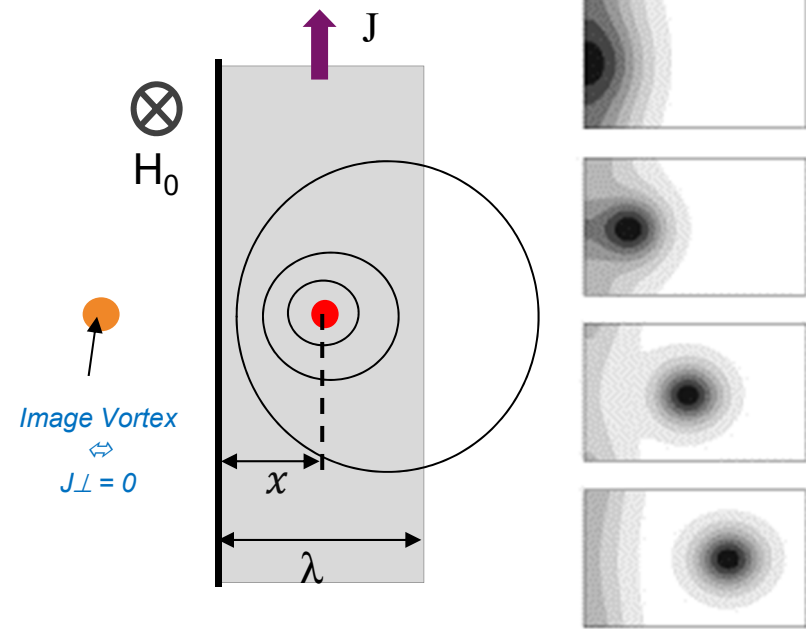
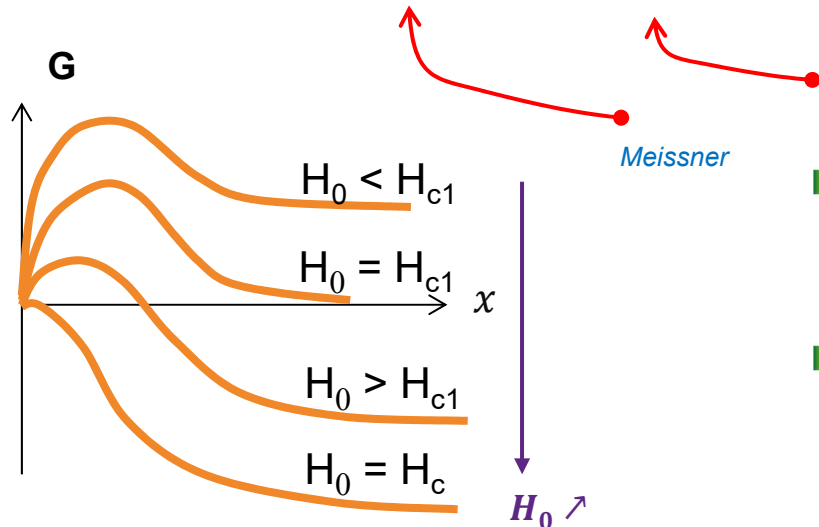
- Boundary condition. ($J_{\perp} = 0$) $\equiv$ “image” vortices

- 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

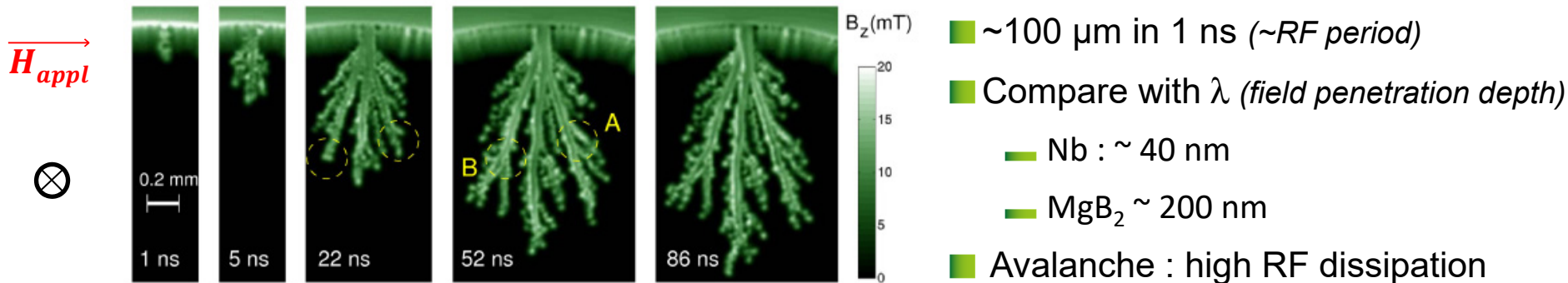
- BUT

- 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

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



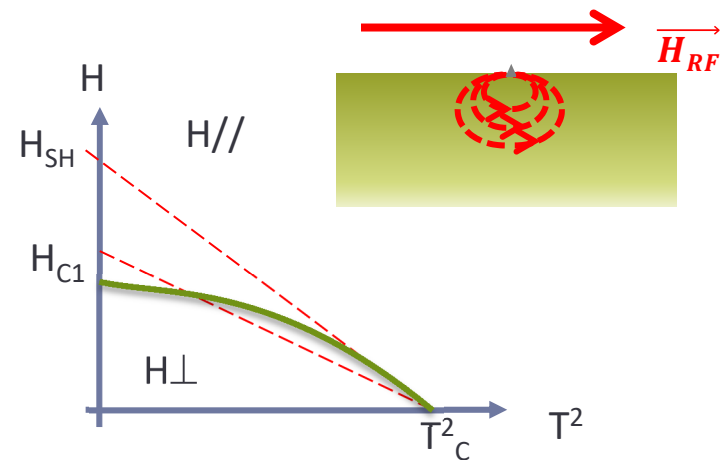
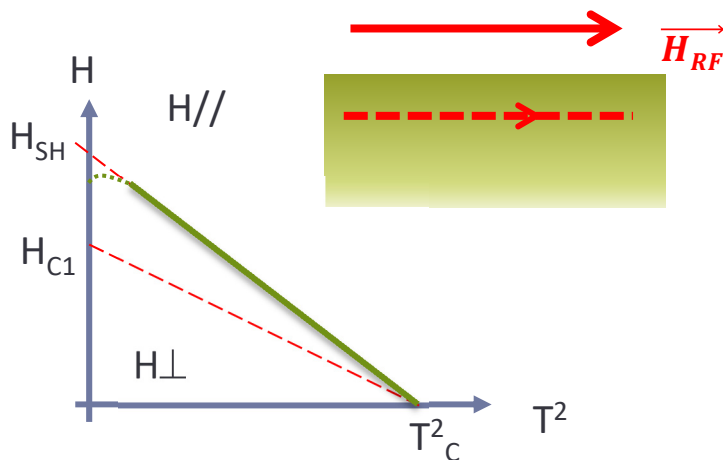
Avalanche penetration/flux jumps



MgB_2 : 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



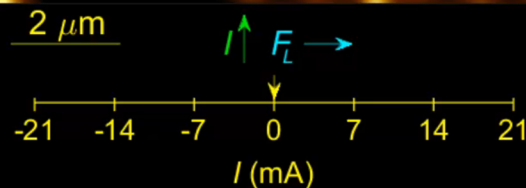
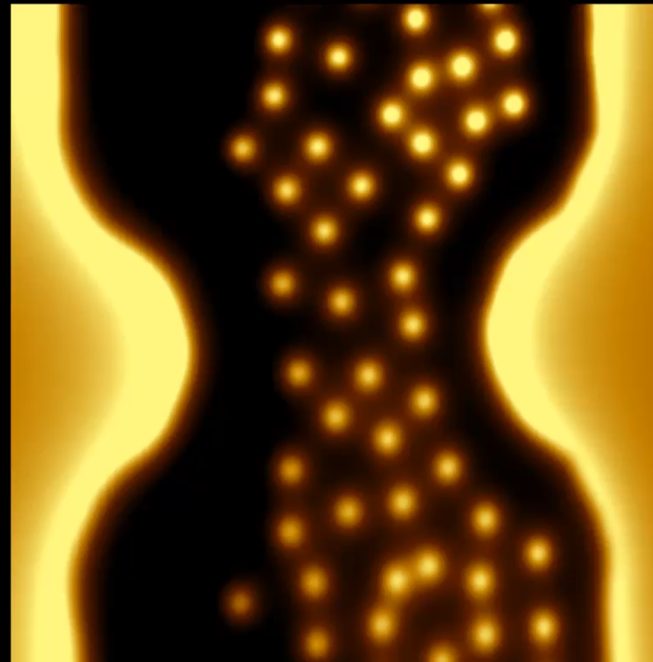
<https://www.eurekalert.org/multimedia/pub/145764.php>

© Dr. Yonathan Anahory

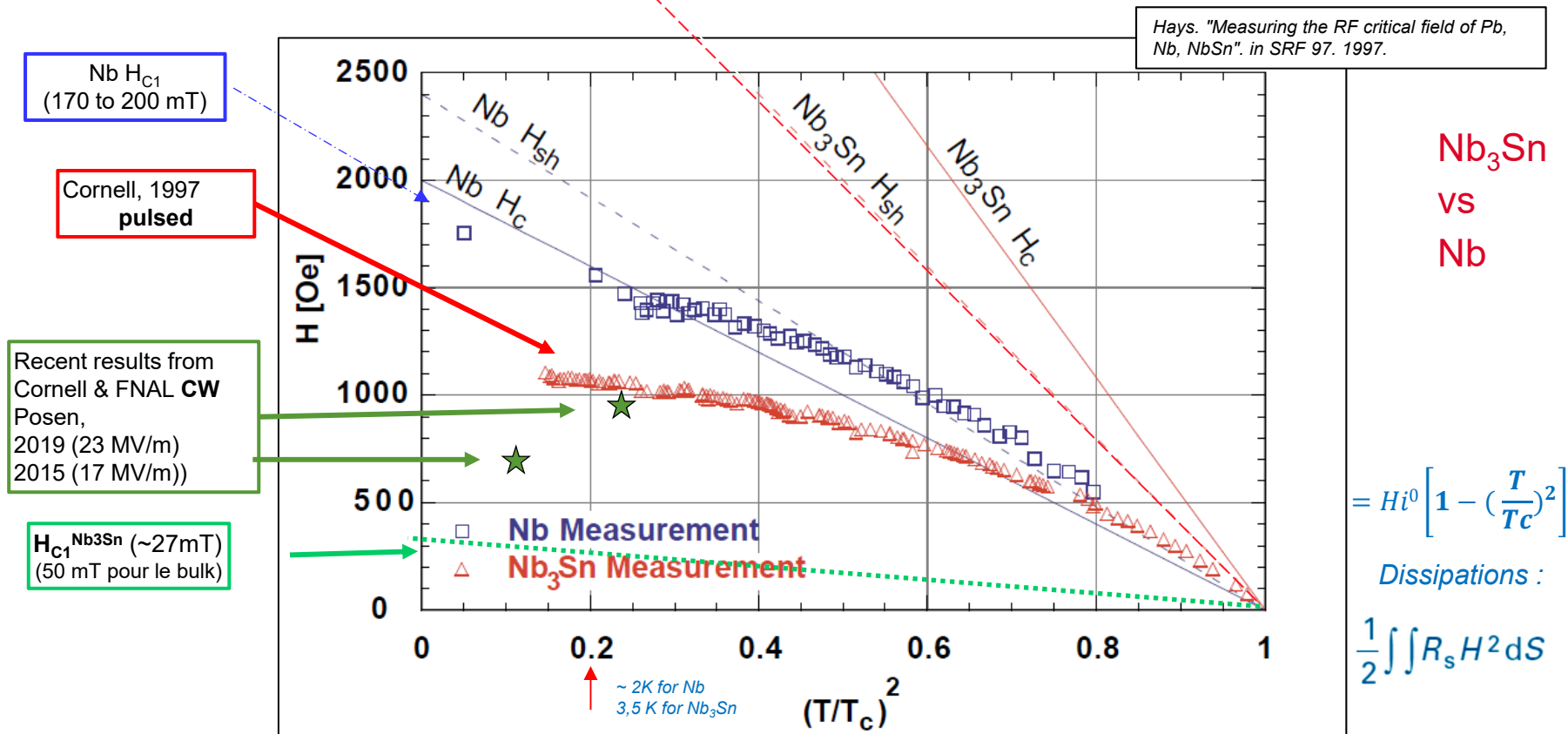
Racah Institute of Physics
The Hebrew University of
Jerusalem

- Lead films
- scanning SQUID-on-tip microscopy technique
- allows magnetic imaging at magnetic sensitivity and high resolution (~ 50 nm)

Vortex dynamics in Pb film at $B_a = 2.7$ mT



At high currents (high drives) vortices move at 20 km/s and appear as smeared line.



■ 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 TOWARD OTHER SUPERCONDUCTORS: GENERALITIES

Needed: high T_C , high H_{SH}
(by defect 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

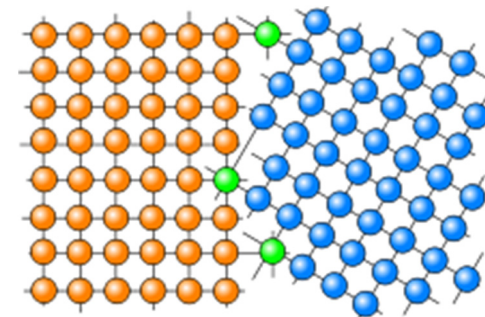
■ Issues with non metallic SC compounds

- 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

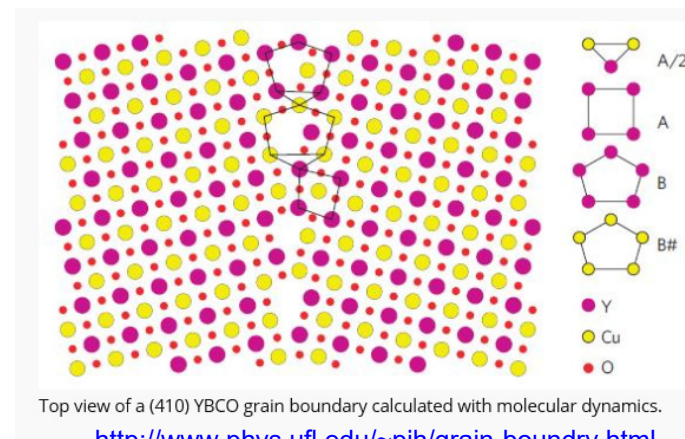
■ Sometimes local disorder =>

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

EX. : Grain boundaries



Some nm $\longleftrightarrow$ Compare with ξ



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



Nb : $\lambda \sim 50$ nm $\Rightarrow$ only a few 100s nm of SC necessary (the remaining thickness= mechanical support) $\Rightarrow$ **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*)

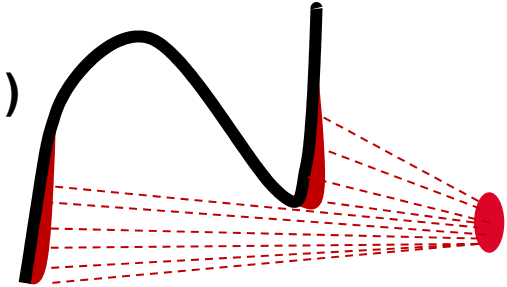
■ Disadvantages

- Fabrication and surface preparation of substrate (*at least*) as difficult as for bulk Nb
- Steep Q_0 reduction often observed by increase of RF field (*sputtered niobium films*)
- 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...*) $\Rightarrow$ **most of the literature recipes are not fitted for SRF application** ☹ ☹ ☹

DEPOSITION TECHNIQUES: 3 MAJORS FAMILIES

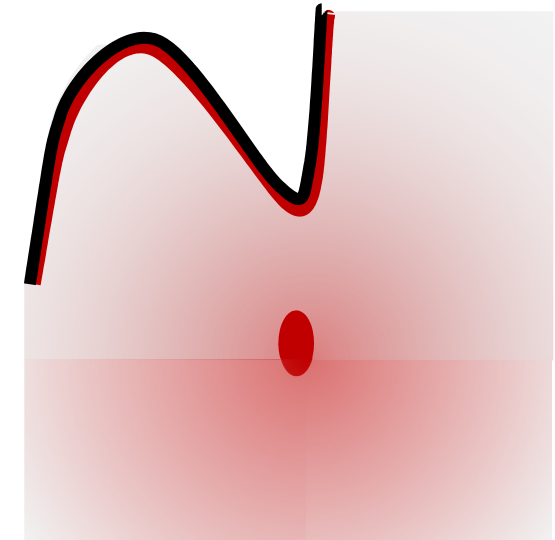
■ Physical deposition techniques (PVD, MS, DS...)

- 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

There are two categories of films

Films: Many techniques, not possible mention all

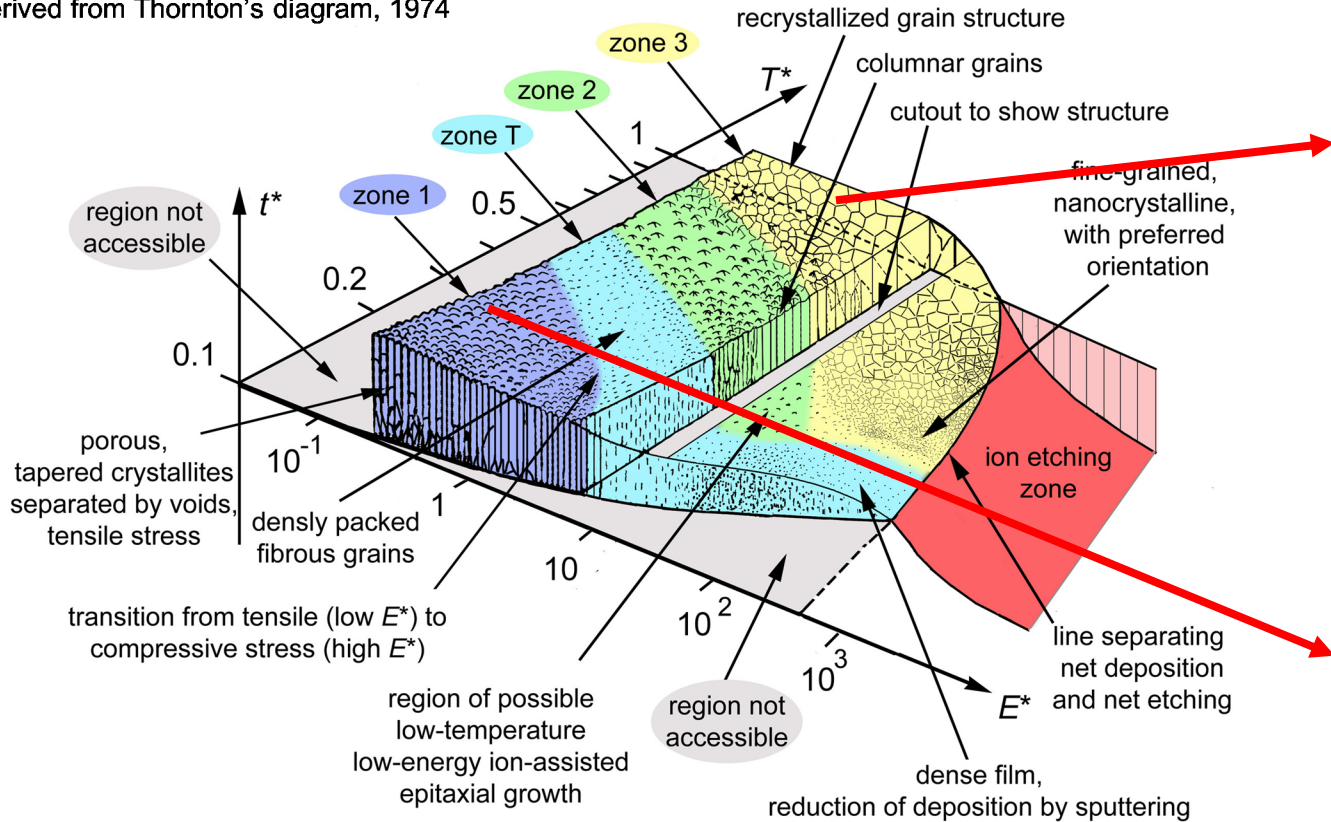
■ Films which are intrinsically films

- Thin, small grains, under stress
- Problems: defects & microstructure, impurities, surface state
- Examples: magnetron sputtered Nb films on oxidized copper

■ The general trend is to move towards films which are bulk-like

- Dense, large grain material
- Examples:
 - high-energy deposition techniques
 - annealed films
 - Nb Cu-clad cavities (hydroformed cavities from bimetallic Nb (2.5 mm), Cu (0.5-1 mm) tube)

Structure zone model (from A. Anders)
derived from Thornton's diagram, 1974



*Energetic deposition
(HPIMS, CED, VAD...)
=> Bulk like films*

*Magnetron sputtering
=> A lot of defects
Cu limits annealing
temperature
recrystallization*

© Andre Anders, 2010

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

11

Unfortunately, more “bulk-like” Nb Films gave disappointing RF results. Not understood yet

Optimizing structure/composition of the films on samples

■ Advantages

- Structure /composition can be optimized with conventional techniques
- Ideal structure and composition can be achieved on model sample (guide for deposition of cavities)
- Cost

■ Disadvantages

- RF performances cannot be directly measured
- **Specific measurement tools need to be developed** (sample cavity, magnetometer...)
- Ultimately a cavity deposition set-up will be needed, but with a known aimed composition & structure

Optimizing deposition inside cavities

■ Advantages

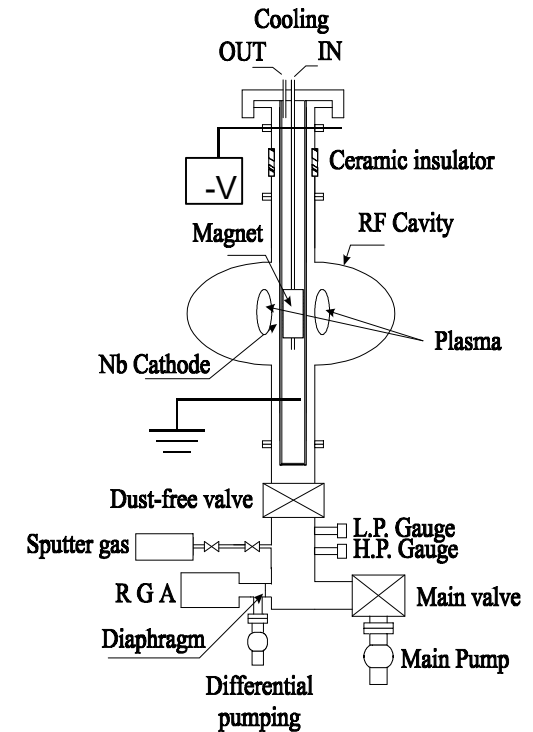
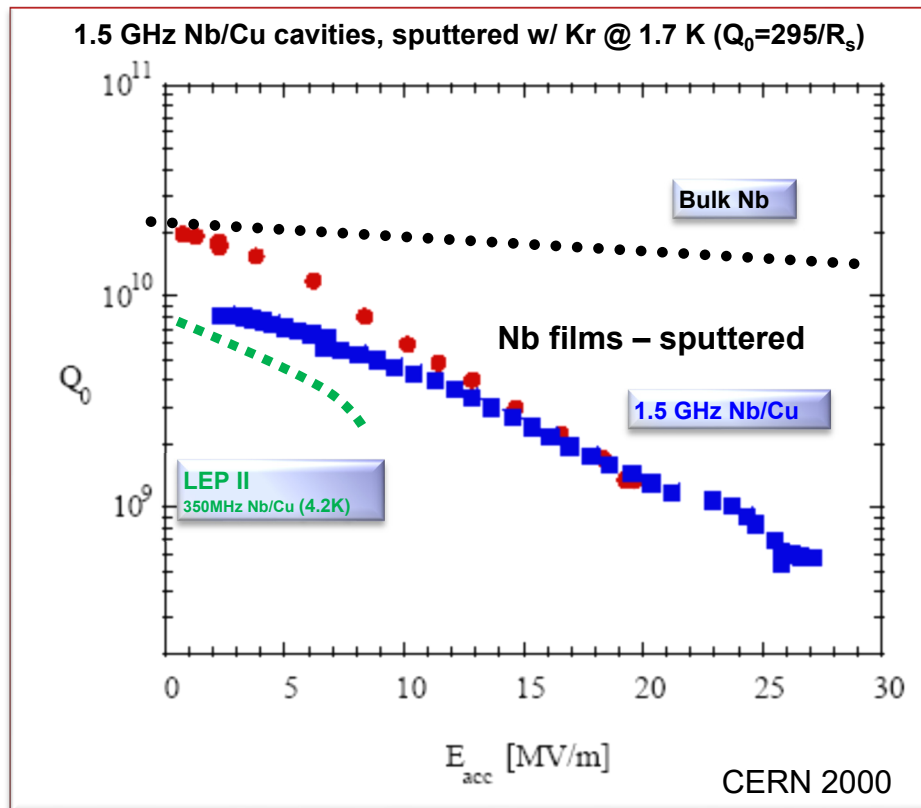
- RF testing easy and gives direct performance
- Work is done only once, direct cavity production

■ Disadvantages

- Very heavy and lengthy, many parameters
- Need to develop a specific cavity deposition set-up
- **Difficult to optimize set-up and films together**
- Optimization of the structure/composition of the film is difficult without structure/ composition info

**Nb/ Cu:
example of the issues
when dealing with thin films**

- The only Nb films deployed in accelerators were made by magnetron or diode sputtering (CERN).
 - Reached relatively low surface fields $\Rightarrow E_{acc} \sim 5 \text{ MV/m}$.
 - Exponential slope in R_s and Q_0

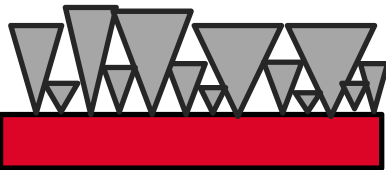


- Possible origin of the slope
 - Depinning of trapped flux
 - Low H_{C1}
 - Early vortex penetration due to roughness

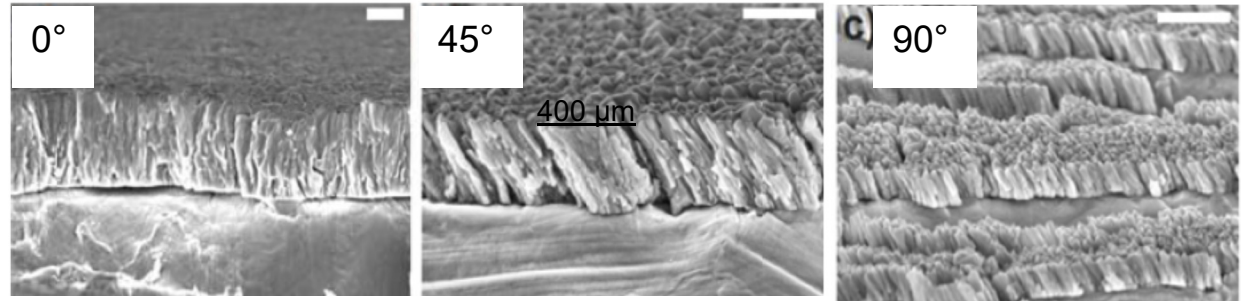
Magn. Sput. Nb

- Line of sight issues
=> porosities

[G. Rozas]

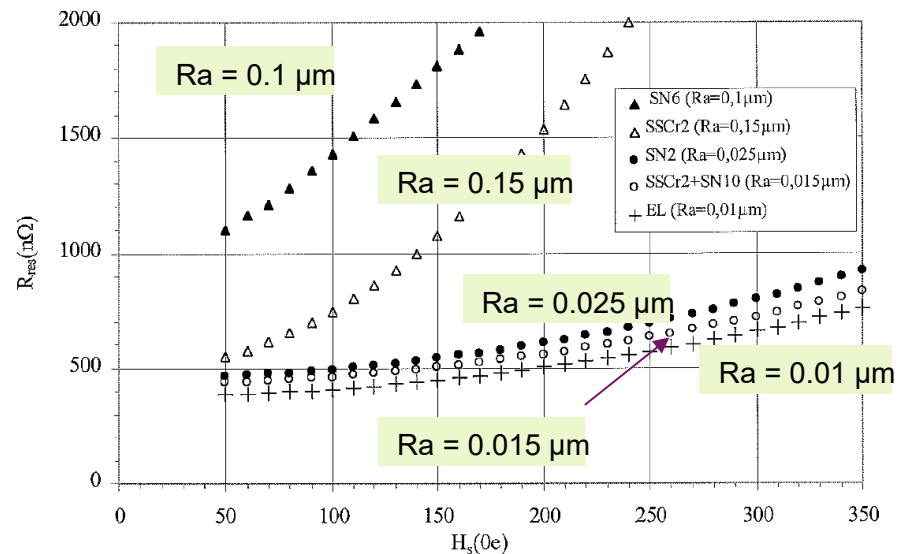


Inverted pyramid crystalline growth



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

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



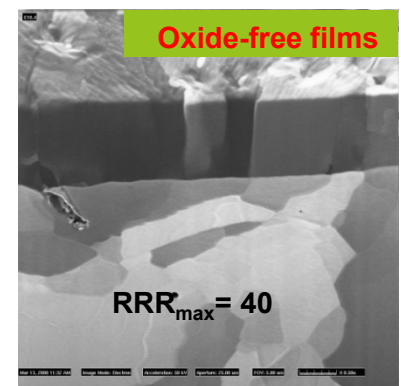
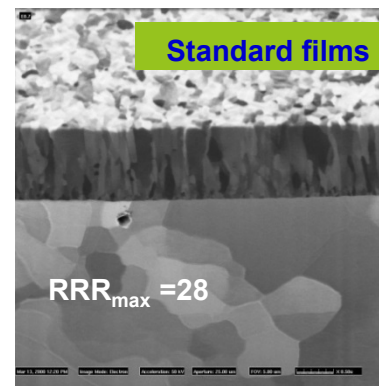
[M. Ribeau, PhD]

- Cu and Nb not miscible (especially in presence of O)
 - Advantage: low interdiffusion
 - Disadvantage: adhesion issues (peeling)
- Best results are not always where expected:
Bulk like films did not perform better ! *(but recent change...?)*



*[data from CERN +
AM Valente-Feliciano]*

	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



Substrate	RRR
Single crystal insulator	
MgO (100)	176
MgO (110)	424
MgO (111)	197
a-Al ₂ O ₃	488
c-Al ₂ O ₃	247

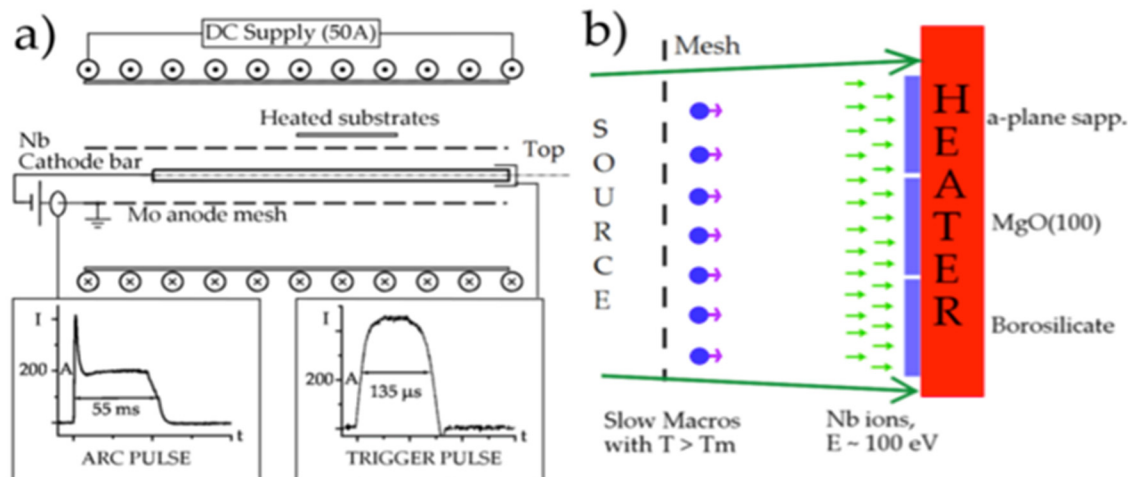
Cu large grains 289

Record 585

- Ions Energy 60-120 eV
- Arc source is scalable for large scale cavity coatings
- UHV and clean walls important

■ Bulk like RRR values

■ Here again, cavities performances disappointing



Cathodic arc plasma.

*Nb films grown by Jlab and AASC
Almeda Applied Science Corporation.*

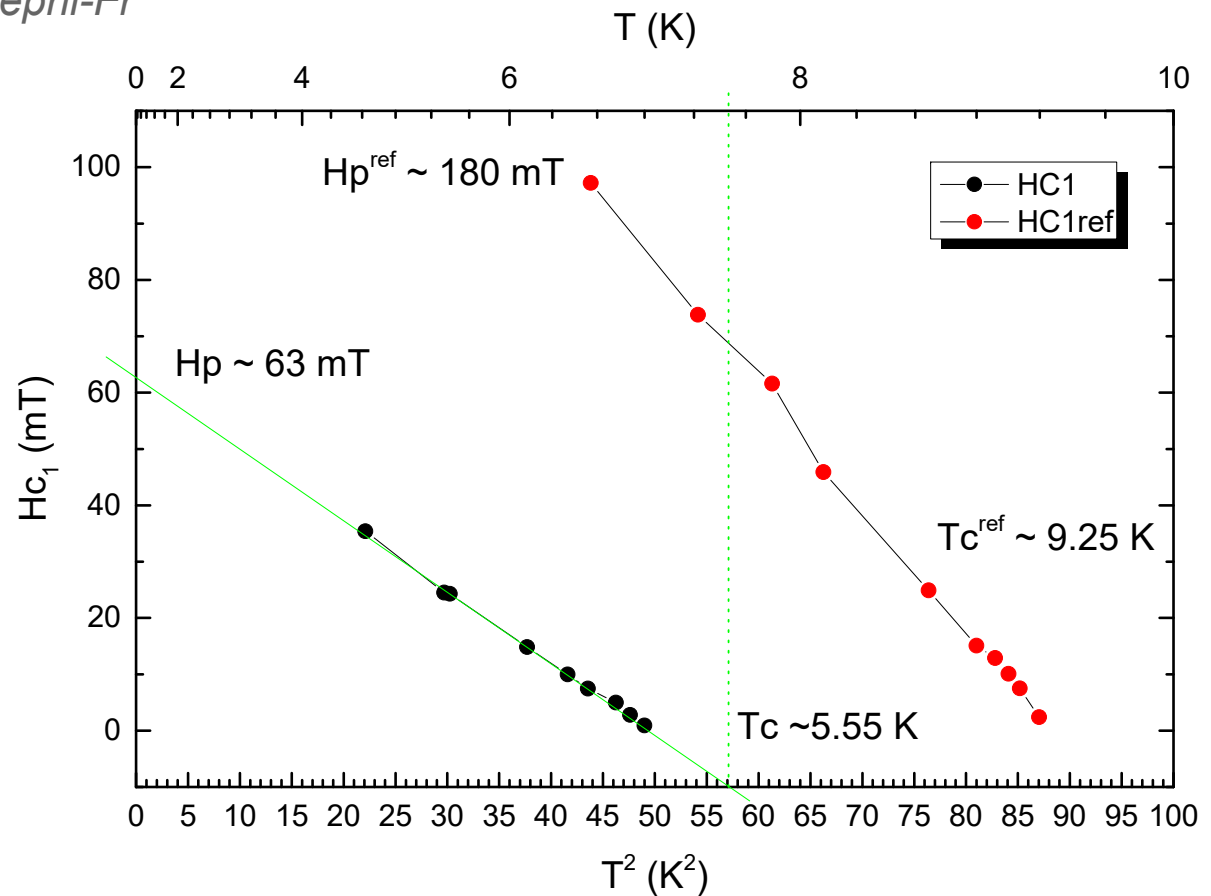
[Ch. Reece; Jlab]



**Coaxial Energetic Deposition
(CED™)**

Thin films

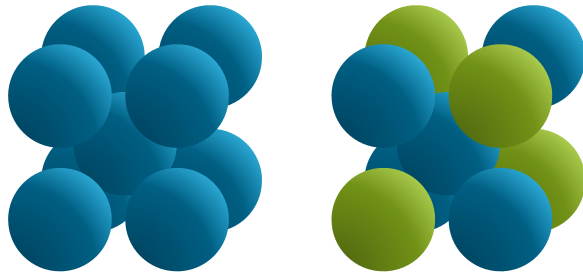
- Thin films : lower H_{C1} , higher H_{C2} / bulk values (because of high λ and low ℓ)
- HPIMS sample from Dephi-Fr



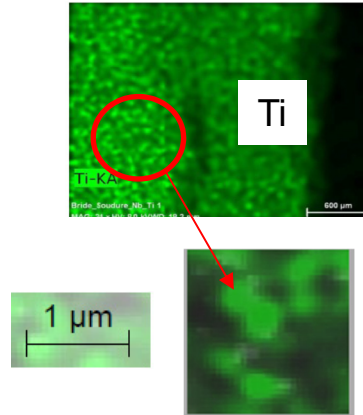
CURRENT SUPERCONDUCTORS:

- **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



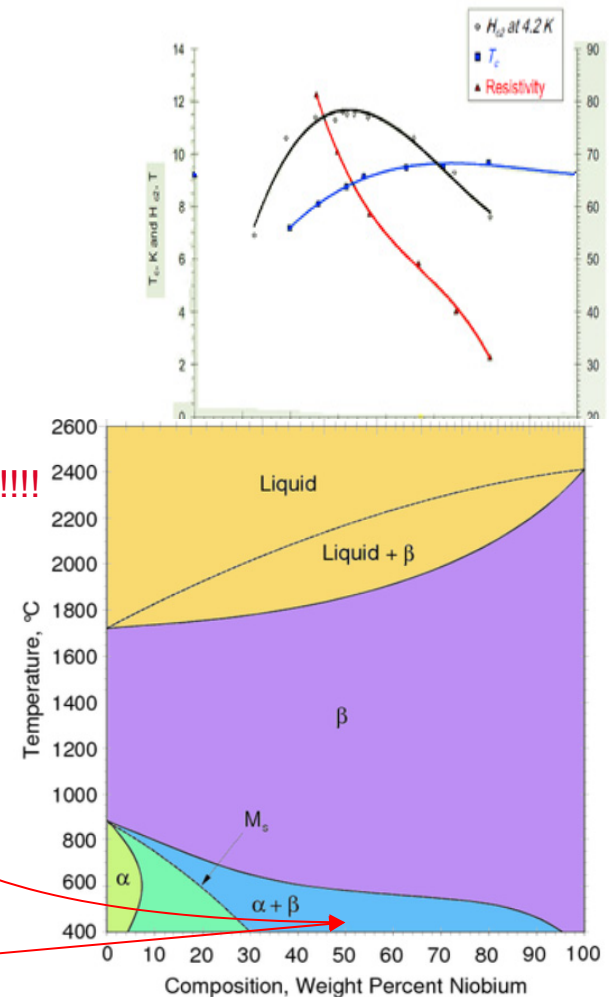
- NbTi widely used in coils
- Available alloys range around 45-55 % Ti
- Ti is not fully miscible inside Nb (Ti precipitates $\exists$ at low T when [Ti] > 5 W%)

=> no RF !!!

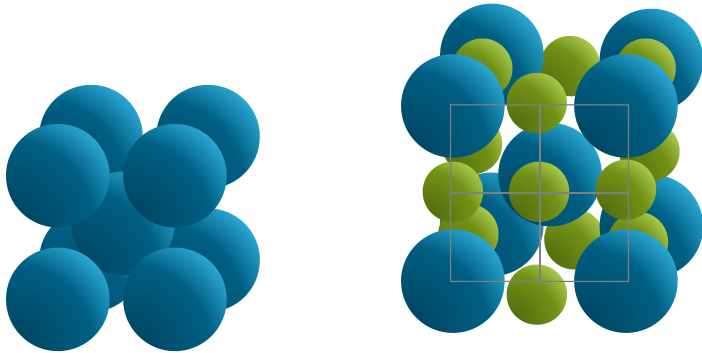
$Ti \varnothing = Nb \varnothing$

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 κ

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



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



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, reactive sputtering...*)
- Good model SC
- Widely used for JJ and SC electronics

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

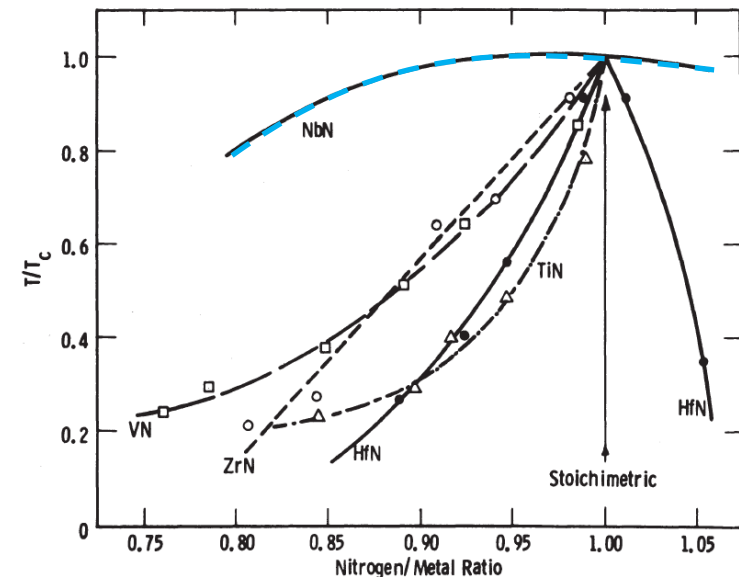


Fig. 20 Critical temperature versus nitrogen-to-metal ratio for various B1-structure nitrides of the transition metals (data assembled by Hulm and Blaugher).

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

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	7			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		

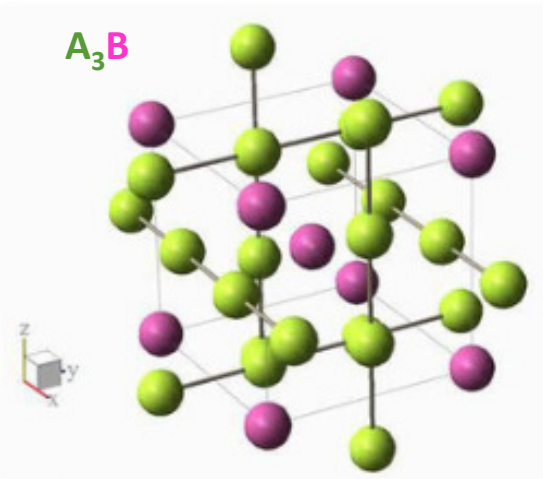
[after Due-Hugues]

■ Extreme brittleness !!!

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

nm $\swarrow$ $\searrow$ μm

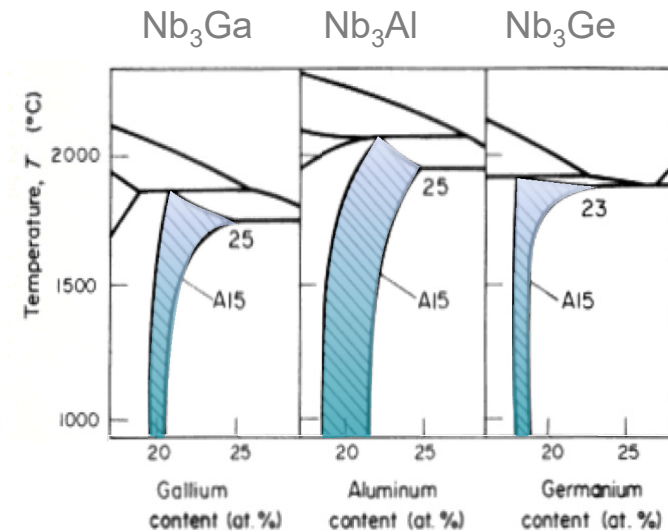
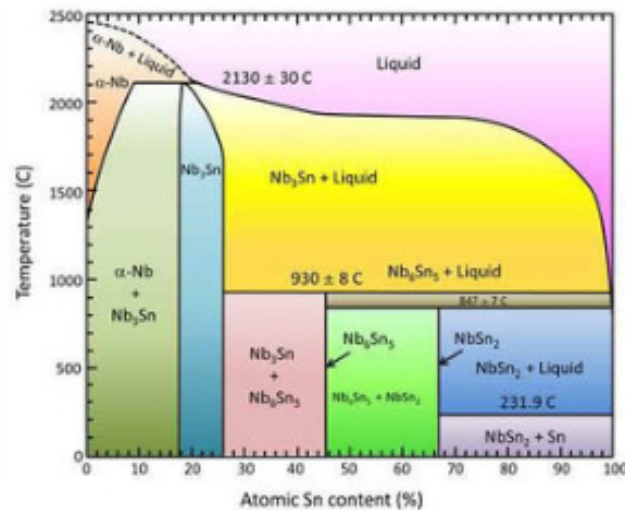
Phases with proper stoichiometry
(A₃B) not stable in normal condition
(RT to Cryogenic temp)



B atoms occupy corners and center of BCC structure

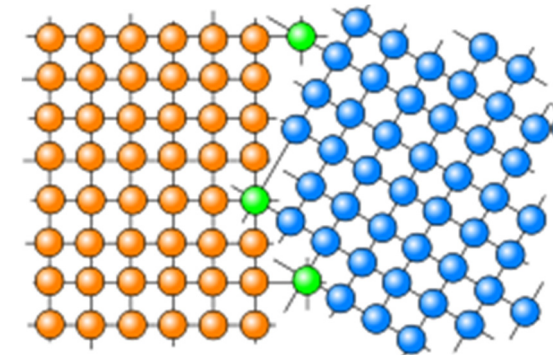
A atoms form orthogonal chains bisecting the faces of the BCC unit cell.

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

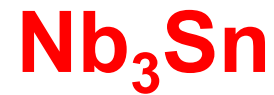


■ Narrow range of concentration for the SC phase:

- 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

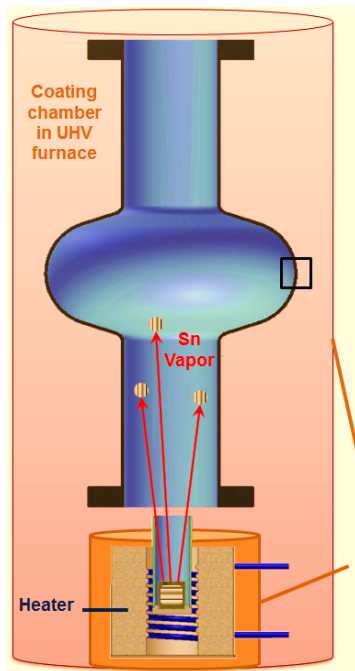
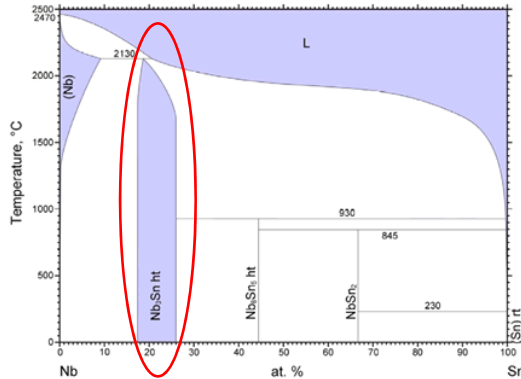


Compare with ξ

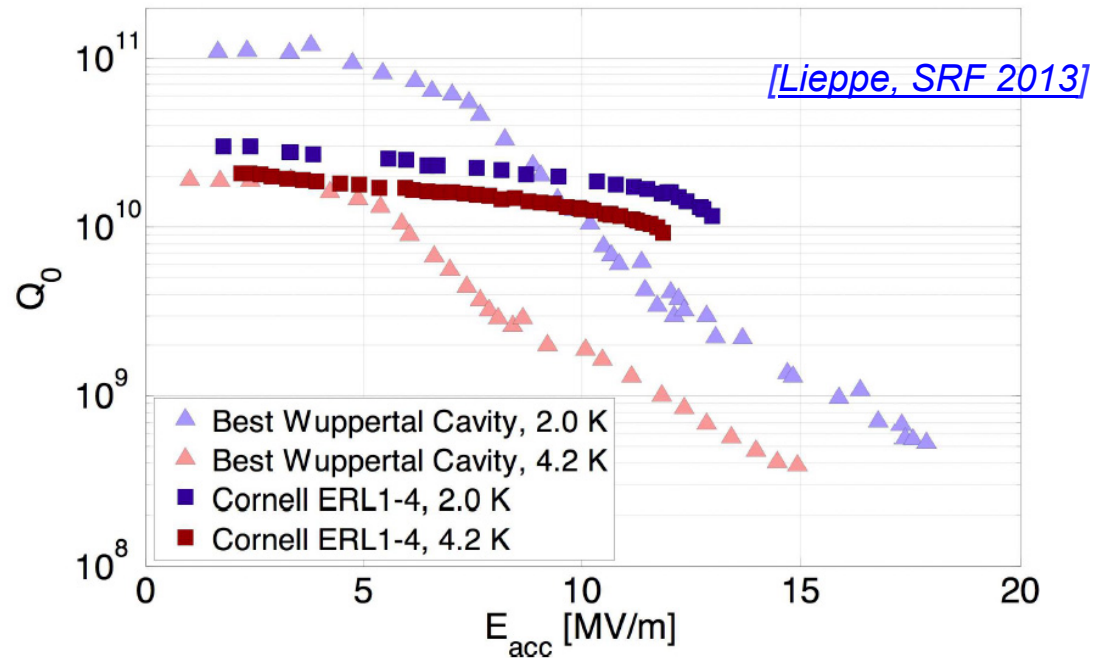


**Special interest for SRF
since the 1980's**

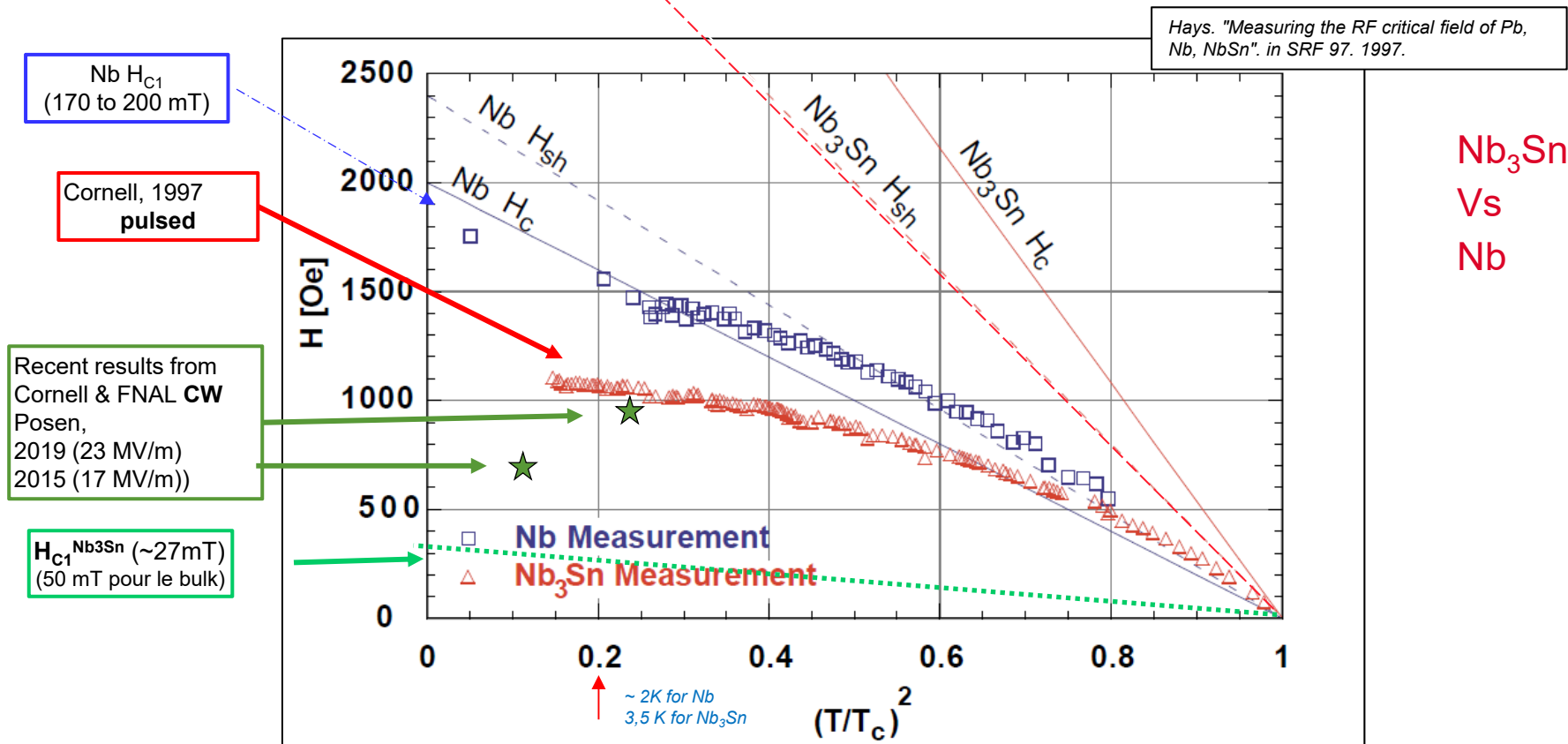
Nb₃Sn ON Nb (thermal way)



Pioneer work: Wuppertal, Cornell



- @ 4,2 K: $Q_0 \times 20$ compare to Nb, @ 2K ~ the same
- Limited in E_{acc} , best results today ~17 MV/m
- Important developments: FNAL, JLAB, CERN, PKU....



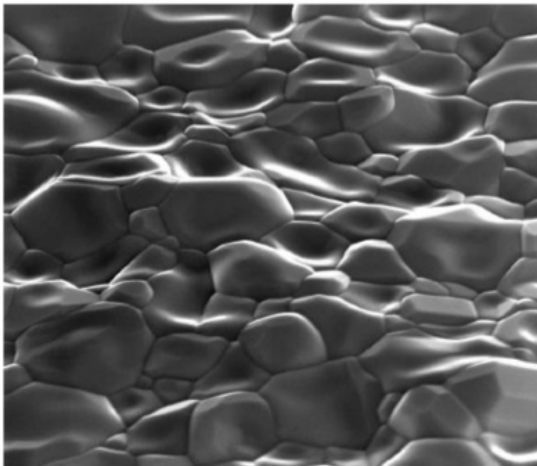
=> We have to reduce defect density

(yes but which ones?)

Candidates of Q-slope and quenching in Nb₃Sn

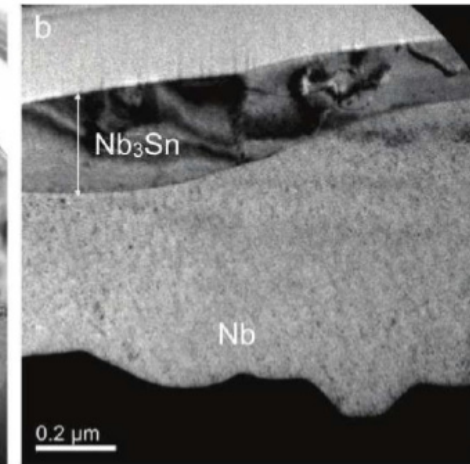
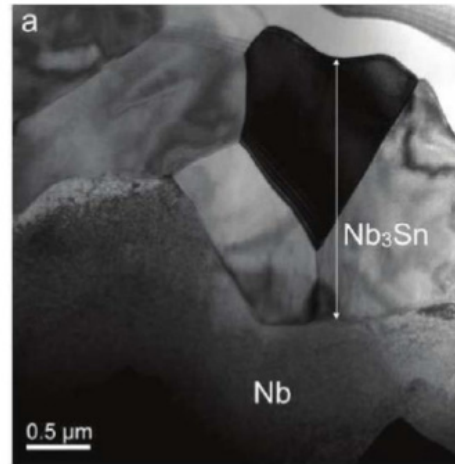
[Jaeyel Lee ttc Milano meeting 2018](#)

1. Surface roughness



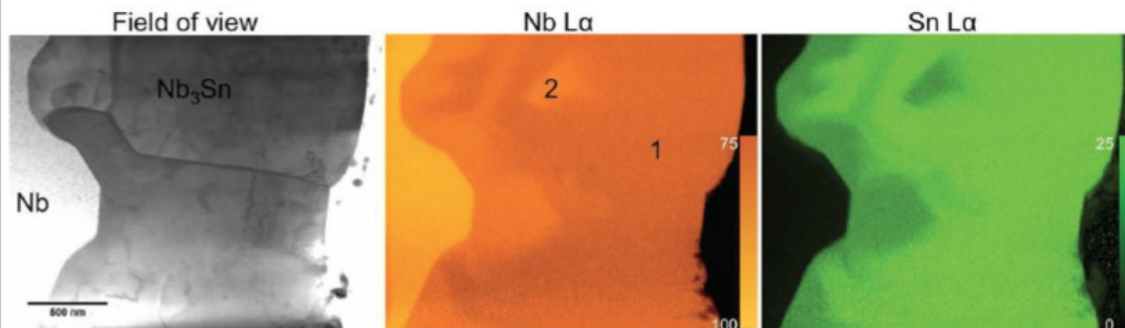
S Posen, PhD thesis, Cornell University (2015)

2. Thin regions



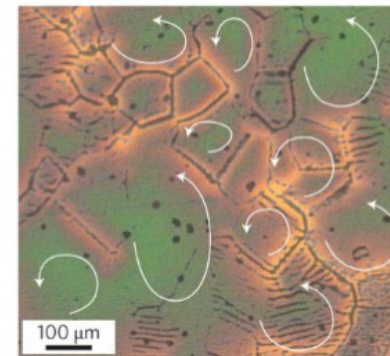
Y Trenikhina et al 2018 Supercond. Sci. Technol. 31 015004

3. Composition variation (Sn-deficient region)



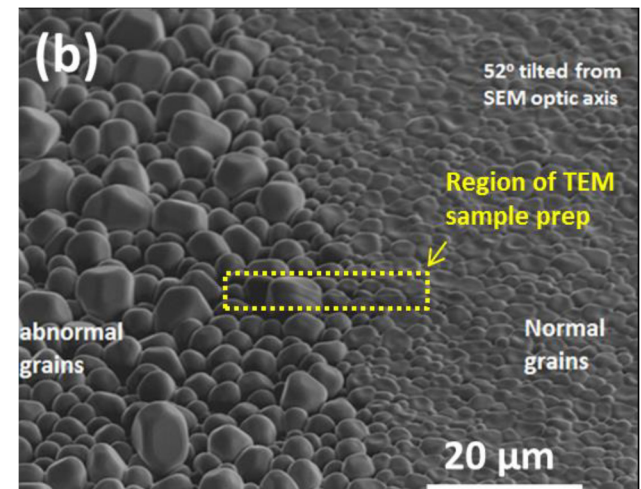
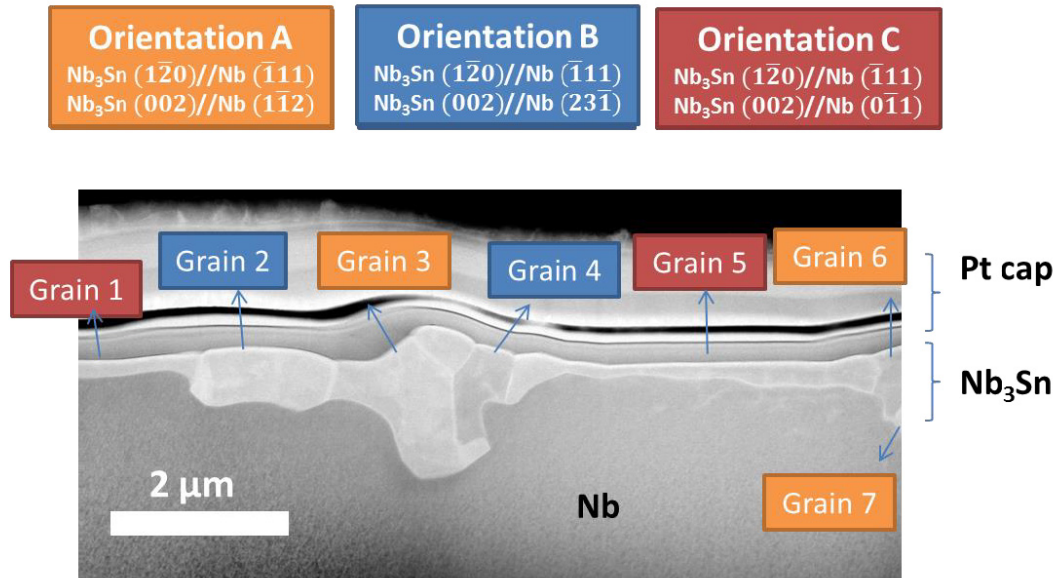
C Becker et al, APL 106, 082602 (2015)

4. Grain boundary



A. Gurevich, Nature Materials 10, 255–259 (2011)

<https://export.arxiv.org/ftp/arxiv/papers/1807/1807.03898.pdf>

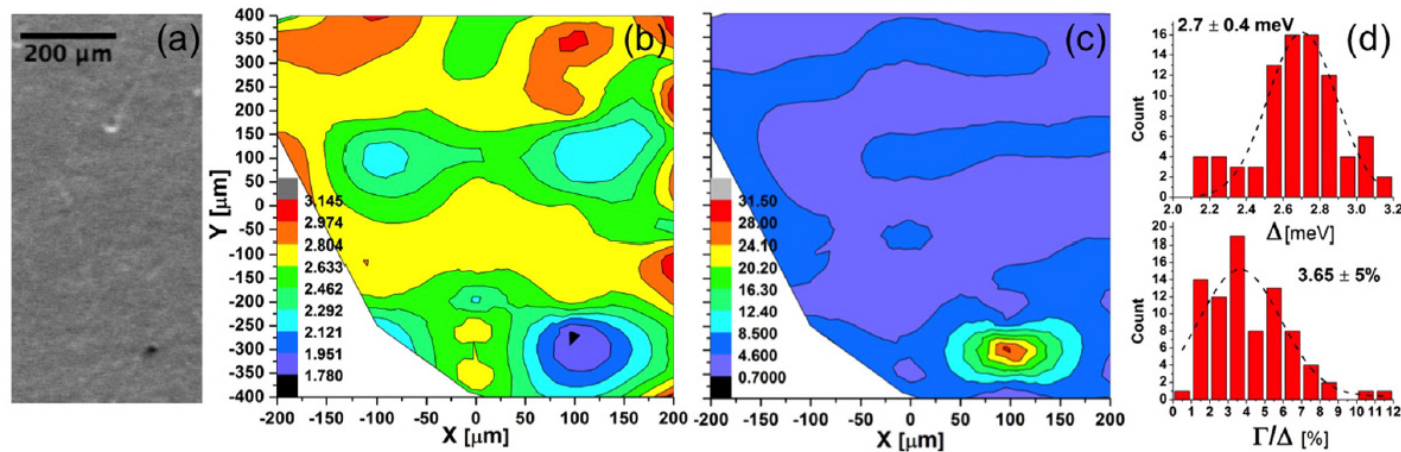


■ Nb₃S film grows in epitaxy

- Regions with poor lattice mismatch grow slower
- Thinner regions tend to be depleted in Sn => lower T_C, early transition
- Can be partially mitigated with interlayer (*e.g. anodization of Nb*)

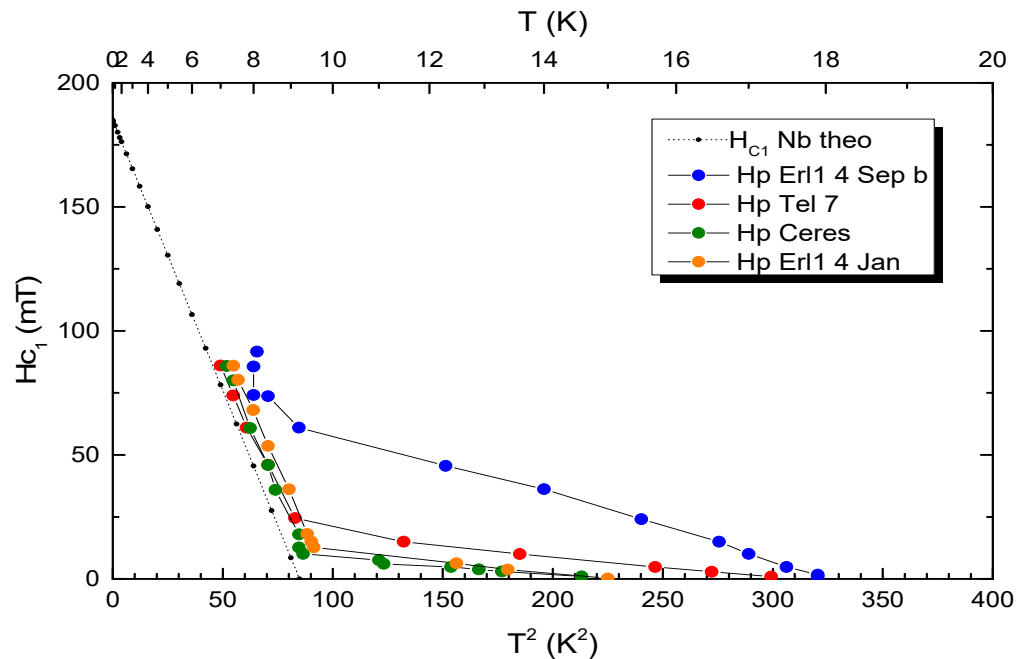
■ Complex materials:

- Real difficult to master
- Very sensitive to defects



Nb₃Sn series

- Along with cavity results and PCT [T. Proslie]
- Magnetometry follows the trends observed on RF tests

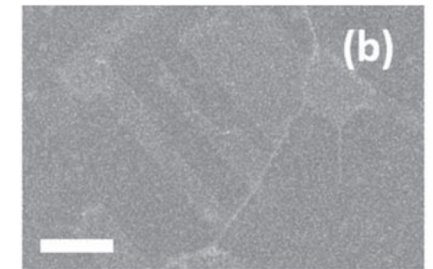
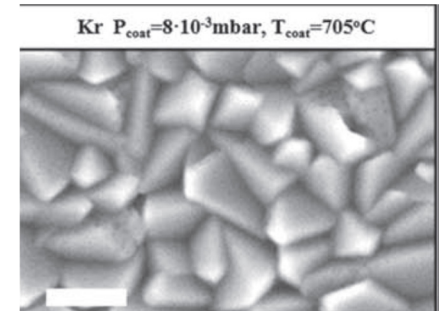


■ Sputtered Nb₃S films on copper

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

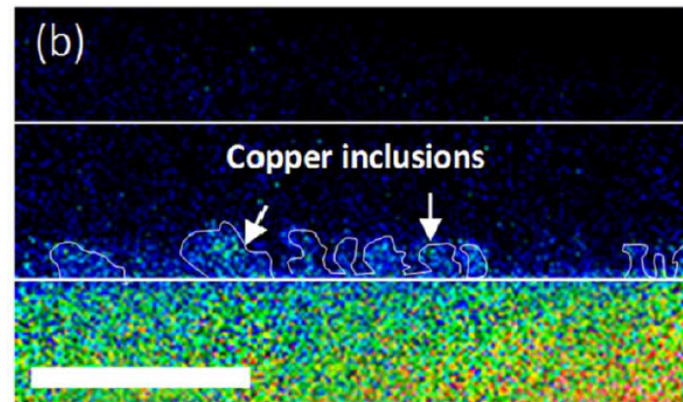
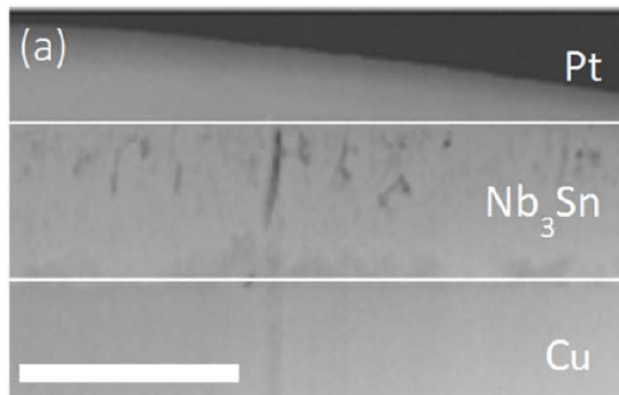
■ Other issues

- 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*



■ Electrochemical deposition + diffusion through copper

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

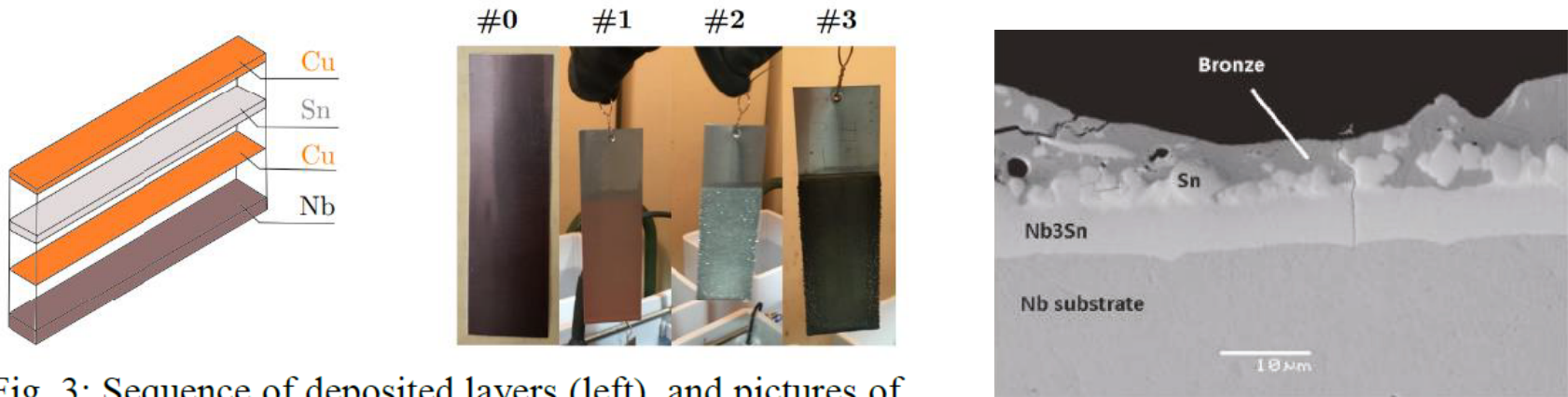


Fig. 3: Sequence of deposited layers (left), and pictures of sample at each deposition step (right).

- Multilayer is heated => solid state diffusion
- Cu lowers the formation T_p° of A15 phase and suppresses the unwanted NbSn_2 and Nb_6Sn_5 phases.

2-D SC (Compounds, anisotropic)

- MgB_2
- Cuprates, Pnictides
- Multilayers

BCS type superconductor

■ $T_C \sim 40$ K, two-gap nature

■ **Advantages:**

- Very high T_C (higher temp operation)
- Semimetal, cheap (fertilizer !)
- ξ, λ of high quality* MgB_2 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 :

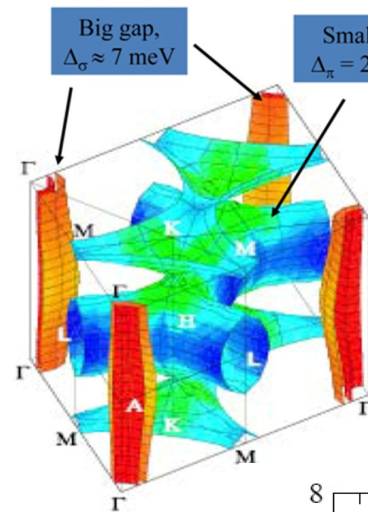
$$\Delta^{\text{Nb}} = 1.5 \text{ meV} < \Delta_{\pi}^{\text{MgB}_2} = 2.3 \text{ meV} < \Delta^{\text{Nb}_3\text{Sn}} = 3.1 \text{ meV}$$

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

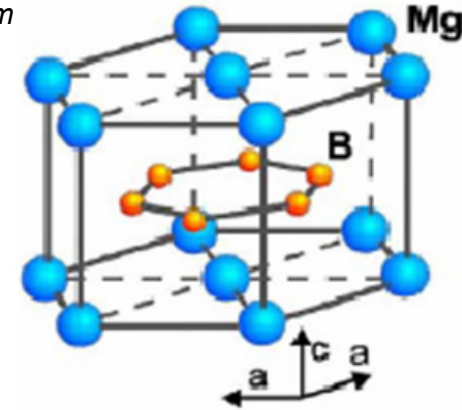
- Sensitive to H_2O (capping necessary ?)
- Thin film routes difficult

* wire quality MgB_2 : $\xi \sim 1\text{-}3$ nm, $\lambda \sim 250$ nm
(by playing on m.f.p.: crystal structure, grain size, impurities...)

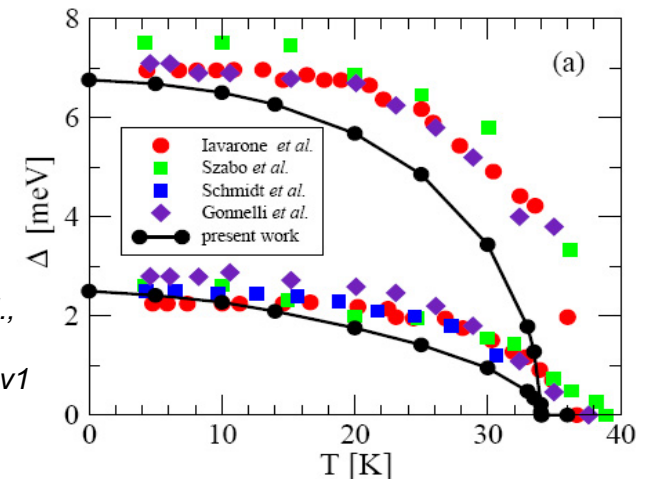
Graphite-type boron layers
separated by hexagonal close-
packed layers of magnesium



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

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 MgB_2 $T_{\text{melt}}/2 = 540^\circ\text{C} \Rightarrow P_{\text{Mg}} \sim 11$ Torr
- Too high for UHV deposition techniques (PLD, MBE...)

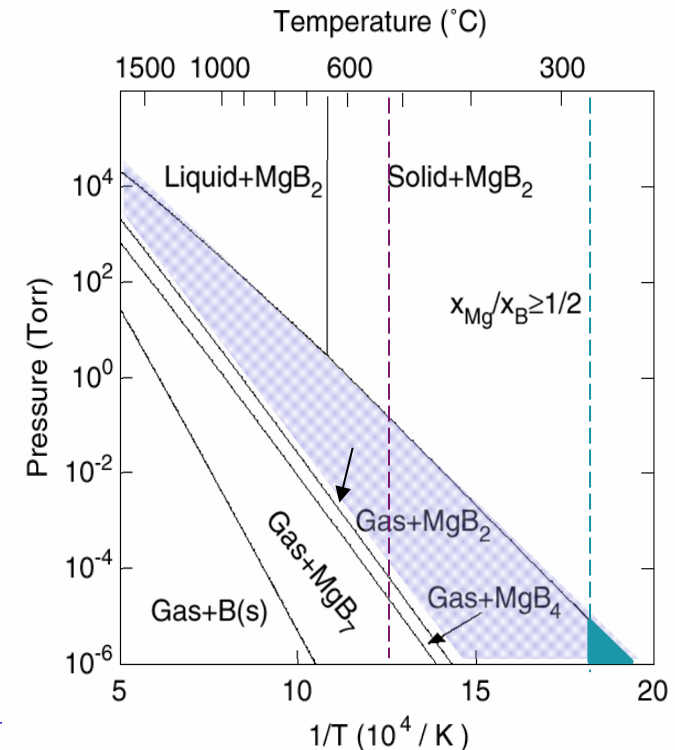
■ **At $P_{\text{Mg}} = 10^{-4}$ - 10^{-6} Torr, and $T_{\text{sub}} \sim 400^\circ\text{C}$**

- Compatible with MBE, and other deposition techniques
- MgB_2 is stable, but no MgB_2 formation:
 - Mg atoms re-evaporate before reacting with B

■ **At $P_{\text{Mg}} = 10^{-4}$ - 10^{-6} Torr, and lower T**

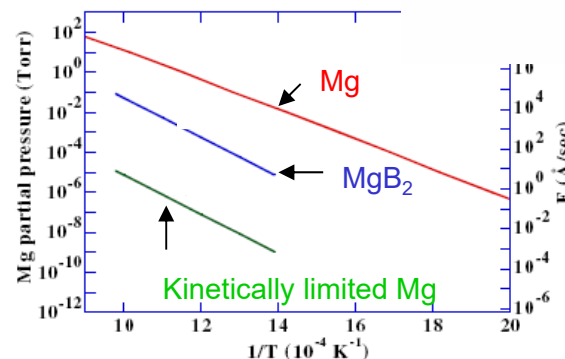
- MgB_2 is stable,
- If $T_{\text{sub}} > 250^\circ\text{C}$, free Mg is lost because the re-evaporation rate is higher than the impinging rate
- If $T_{\text{sub}} < 250^\circ\text{C}$
- **Growth rate is very slow,**
(kinetically limited by available Mg)

evaporation pressure of Mg from MgB_2 < decomposition curve of MgB_2 < Mg vapor pressure



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

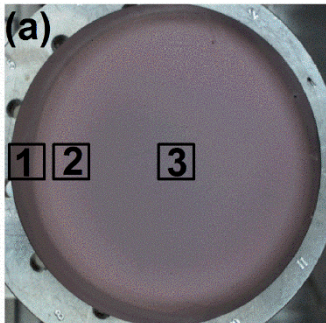
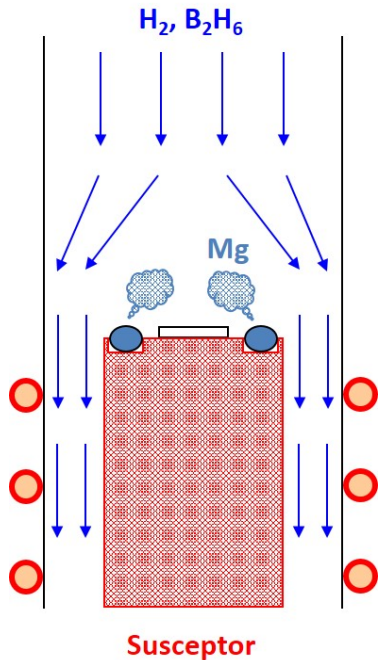
M. Naito and K. Ueda, SUST 17 (2004) R1



MgB₂ – HPCVD ON METAL SUBSTRATES

HYBRID PHYSICAL CHEMICAL VAPOR DEPOSITION

[X. Xi- TempleUniversity]

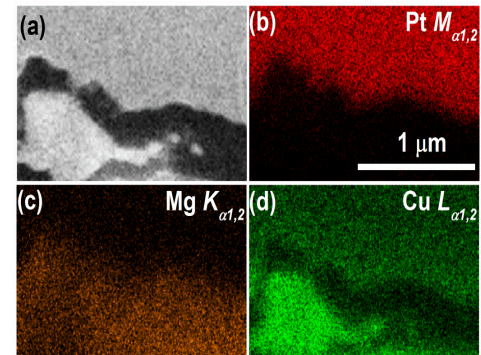
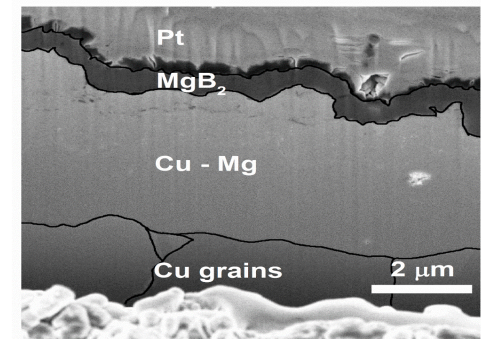
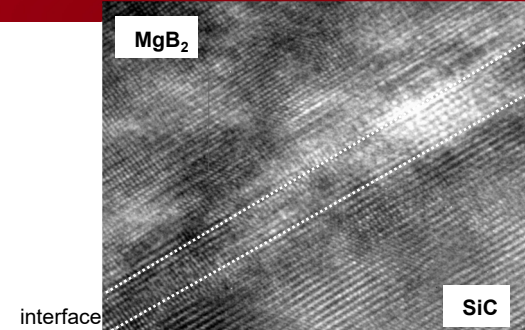


■ Polycrystalline MgB₂ films deposited:

- On stainless steel, Nb, TiN, and other substrates.
- Flat samples and tubes (*conformational*)
- Fitted for SRF apps:
 - $RRR > 80$
 - low resistivity ($< 0.1 \mu\Omega$) and long mean free path
 - high $T_c \sim 42 \text{ K}$ (due to tensile strain),
 - low surface resistance, short penetration depth
 - smooth surface (RMS roughness $< 10 \text{ \AA}$ with N_2 addition)
 - good thermal conductivity (free from dendritic magnetic instability)

■ Keys to high quality MgB₂ thin films:

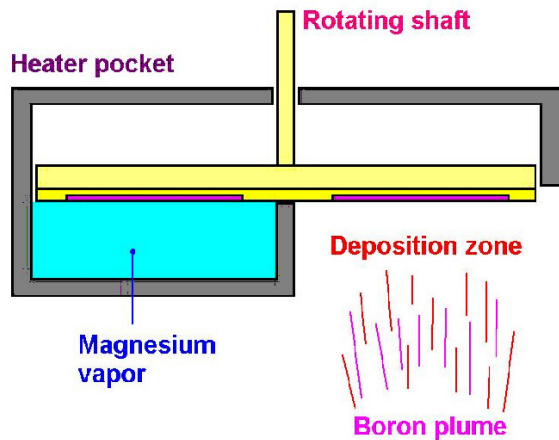
- High Mg pressure for thermodynamic stability of MgB₂
- Oxygen-free or reducing environment
- Clean Mg and B sources
- Prevent formation of spurious phase (e.g. Mg-Cu alloy islands on a Cu substrate)



Reactor/reaction designs require complex calculation in thermodynamics and hydrodynamics

In-situ reactive evaporation @ 550°C

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



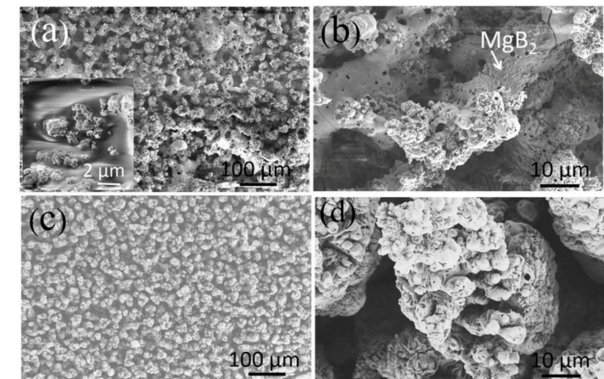
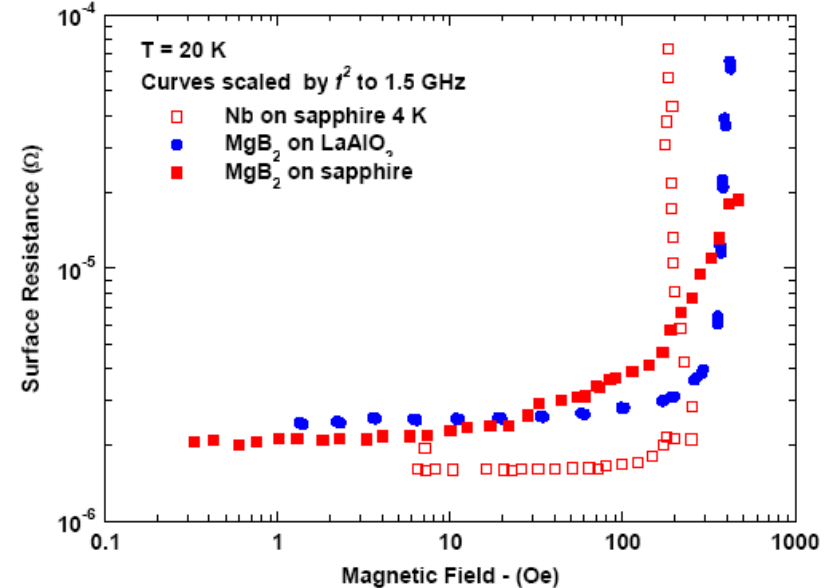
[T. Tajima, LANL]

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

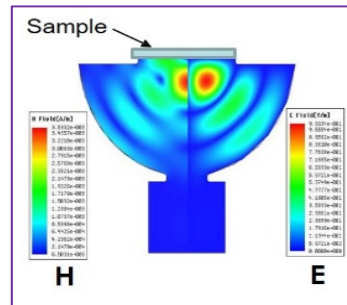
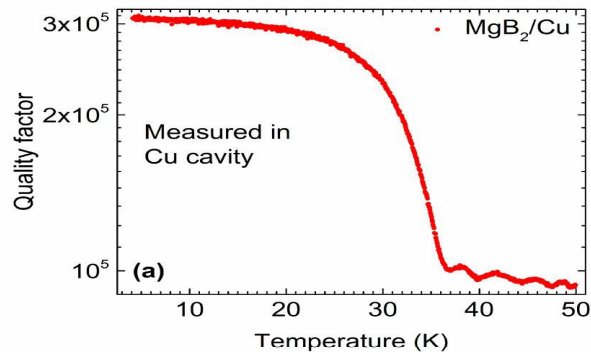
RF measurement @ MIT/Lincoln Lab



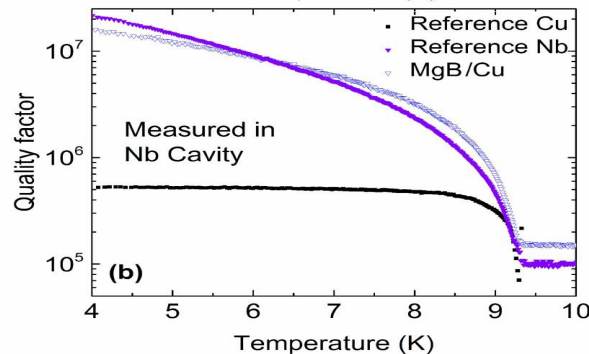
[R. Valizadeh, STFC]

11.4 GHz TE013 cavity @ SLAC

- The MgB_2 coatings were also characterized at 11.4 GHz 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.

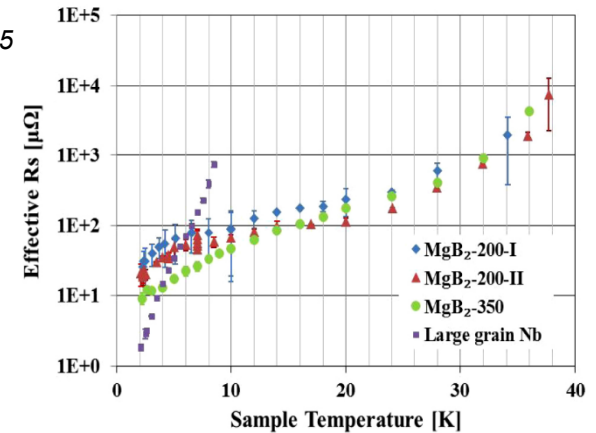


[P. Welander, SLAC]



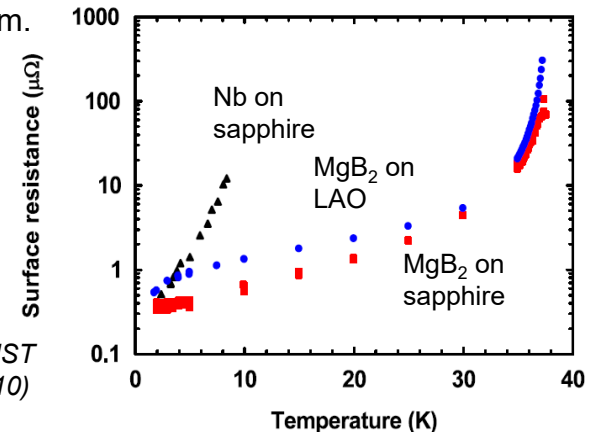
7.5 GHz sapphire-loaded TE011 cavity at JLab

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



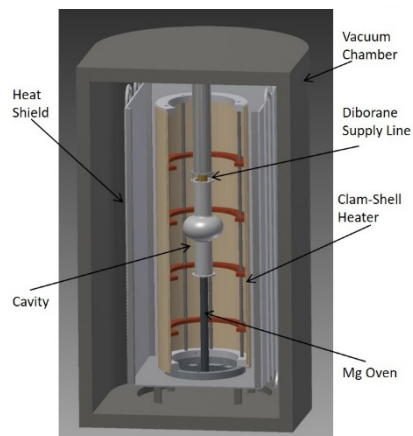
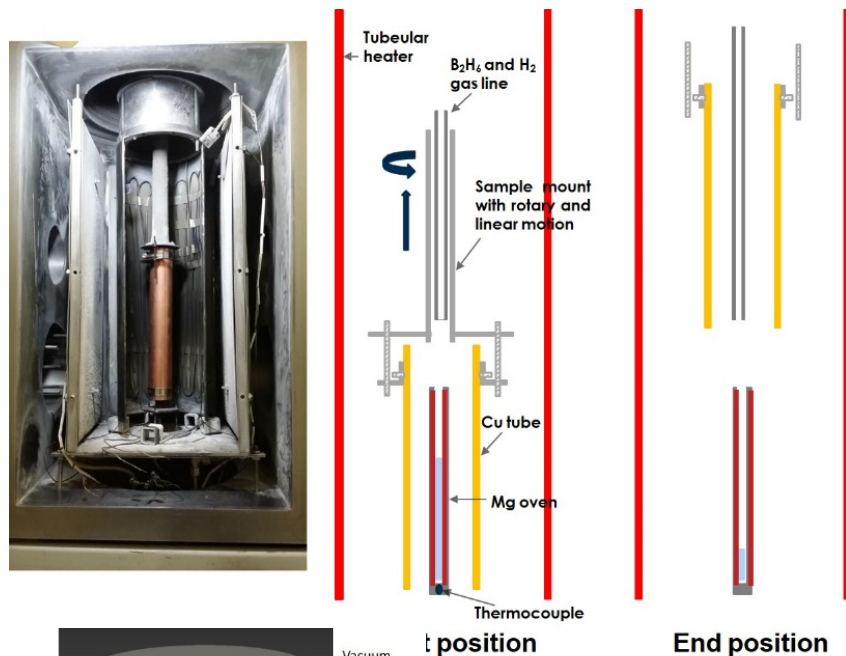
Stripline resonator

- Scaled to 1,5 Ghz
- Lower surface resistance comparable to Nb film.



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

Coating SRF Cavity with a 2-Step Process



[X. Xi, Temple Univ.](#)

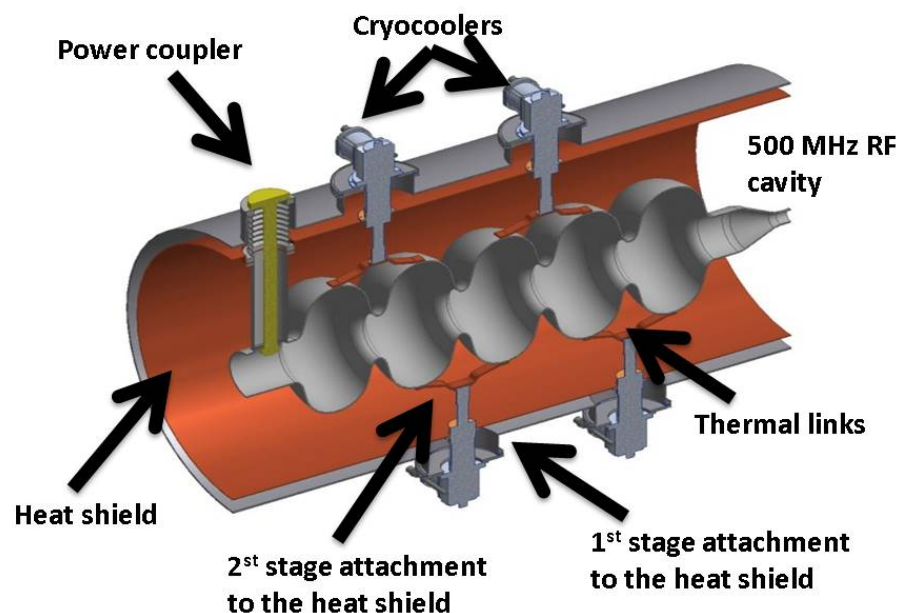
Cryocooler-Cooled MgB₂-Coated Cu Cavities

[A. Nassiri et al., ANL](#)

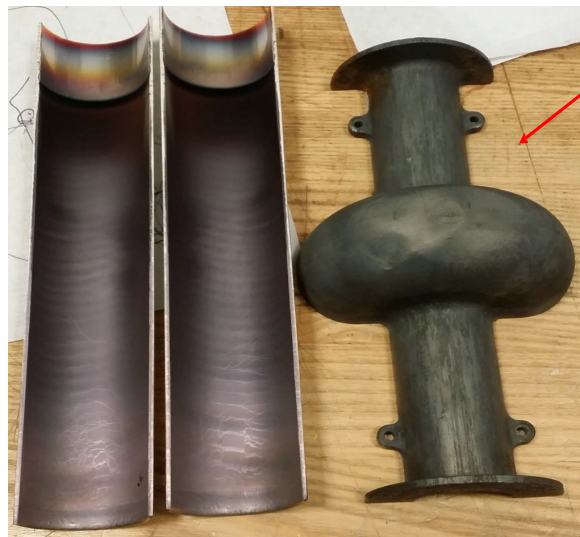
Coating of MgB₂ /Cu makes operation at 8-12 K possible

Temperature range can be achieved with efficient cryocoolers, providing significant benefit with reduced cost.

Goal: 500 MHz MgB₂-coated Cu cavity



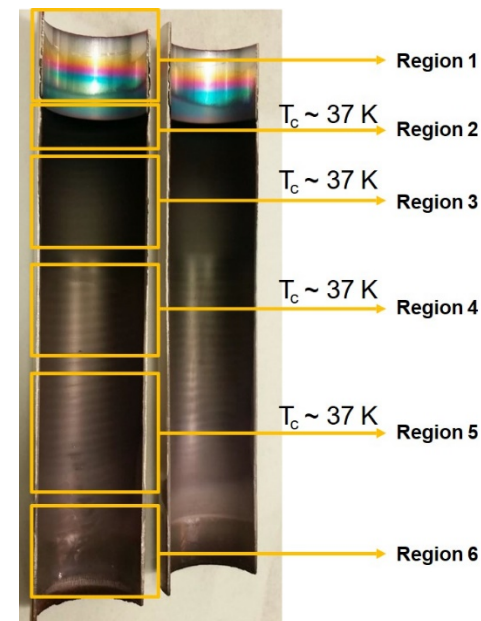
HPCVD MGB₂: CU TUBE COATING-TESTING FOR 3.9 GHZ CAVITY



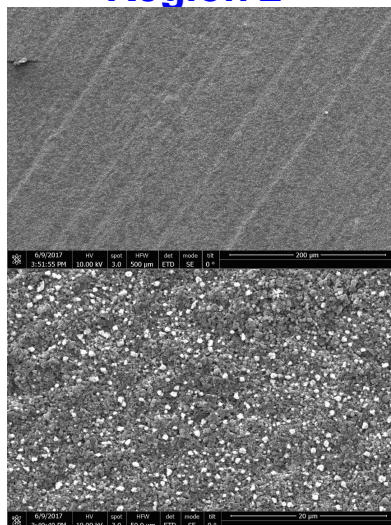
3.9 GHz mock cavity

- B₂H₆ flow rate: 20 sccm
- H₂ flow rate: 100 sccm
- Total pressure: 5 Torr
- Deposition time: 20 - 30 min

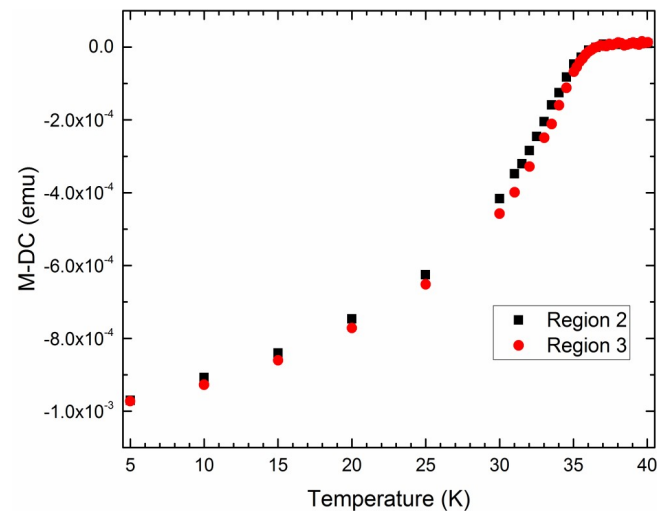
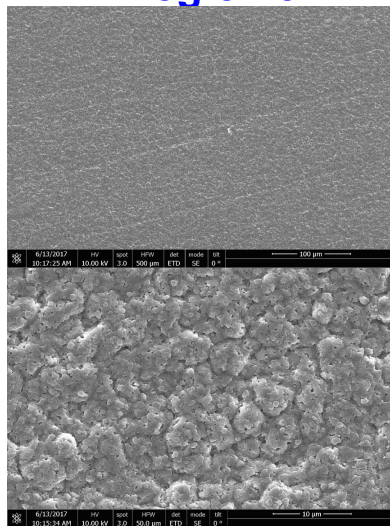
Small piece from each region
are tested for superconducting
and surface properties



Region 2



Region 3



■ MonoXtal : J_c maximum for (a,b) planes and minimum when // c axis

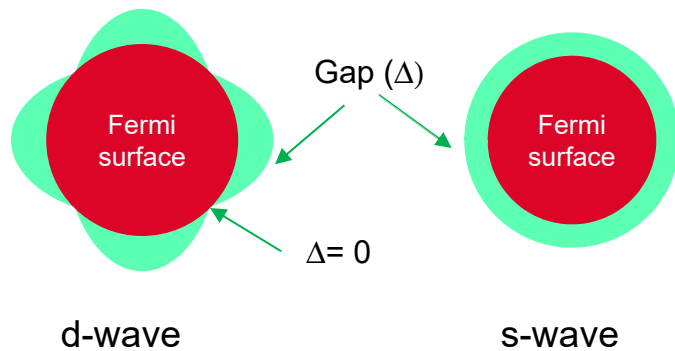
- ξ_c (~0,03 nm) $\ll \xi_a, \xi_b$ (~1-2 nm) $\Rightarrow$ "layered material"

■ Realistic material : polycrystalline, ceramic, fragile...

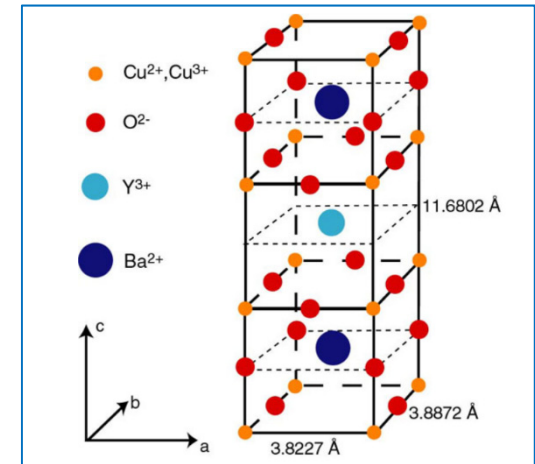
- $\xi_c \ll$ disordered area at G.B $\Rightarrow$ grains are decoupled (weak links)
- $\Rightarrow$ try to introduce preferential orientation (epitaxy): difficult to get on a cavity (but is applied to fabricate tapes for magnets)

■ D-symmetry of the gap

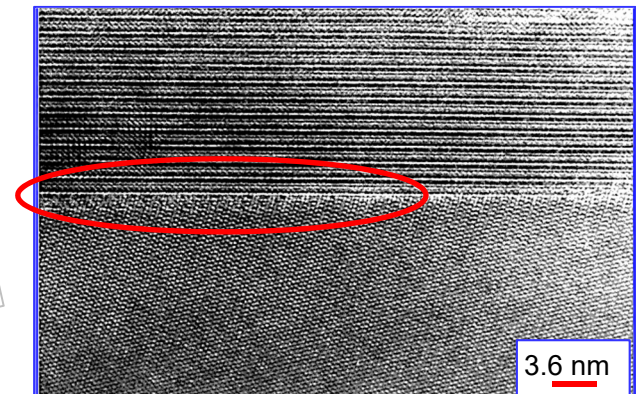
- superconducting gap is also anisotropic
- = zero at four line nodes located at the diagonals of the Brillouin zone
- $\Delta = 0 \Rightarrow$ power law for R_S : $R_S \propto T^2 - T^3$
- For the recall: gaps of conventional SC have s symmetry: isotropic and $R_S \propto e^{-\Delta/T}$ (BCS resistance)



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



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



Twin boundary in YBCO

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

■ Oxypnictide base: ReOMPn

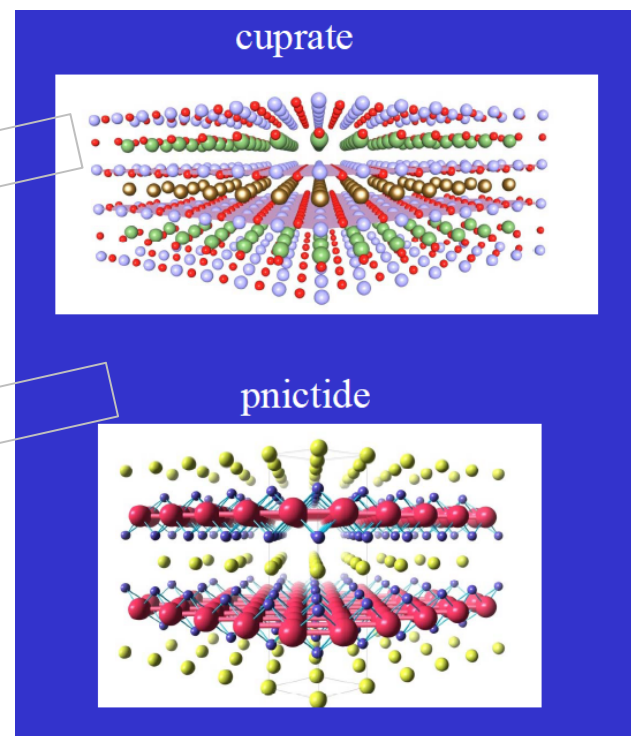
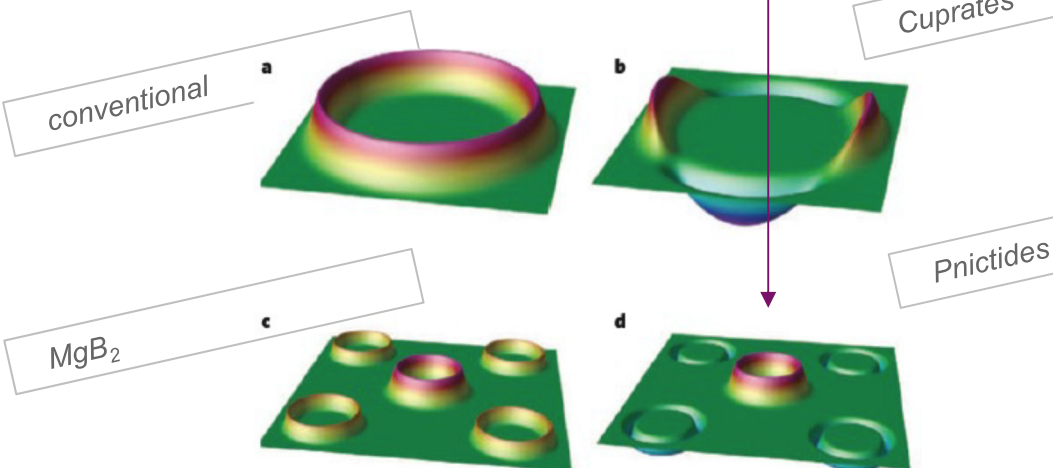
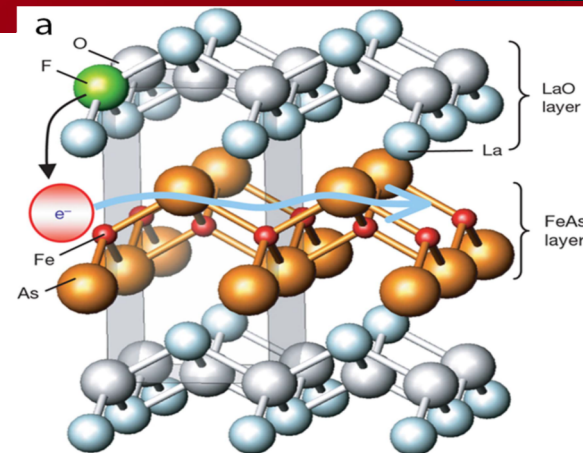
- $M = \text{Fe, Co, Ni}$
- $\text{Pn} = \text{As or P}$
- $\text{Re} = \text{La, Nd, Sm, Pr}$

■ A lot of 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 ?

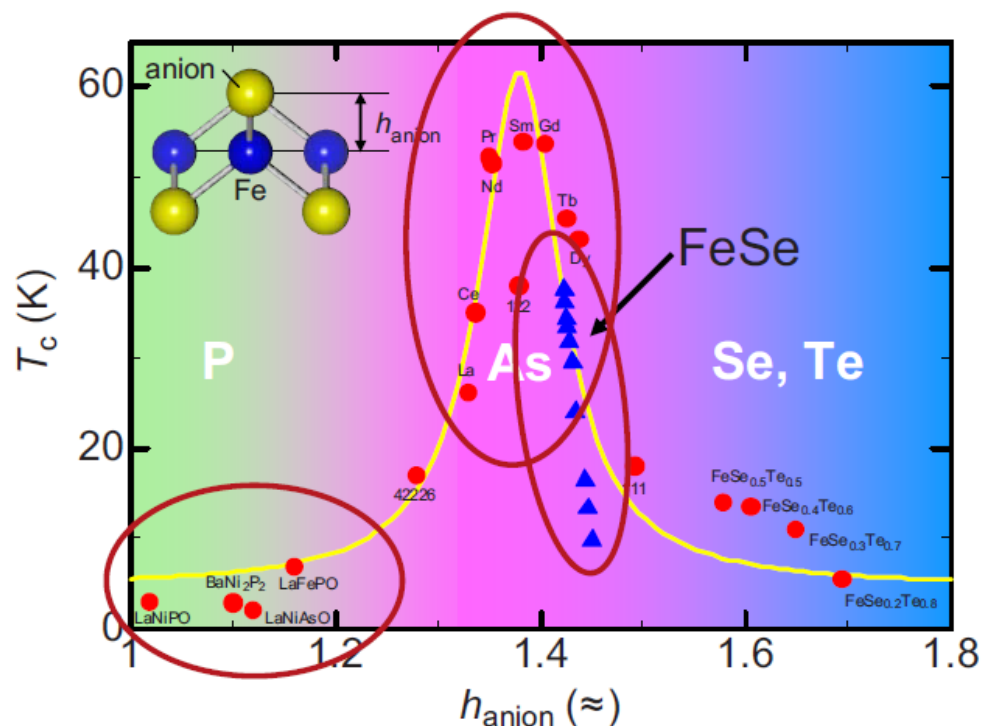
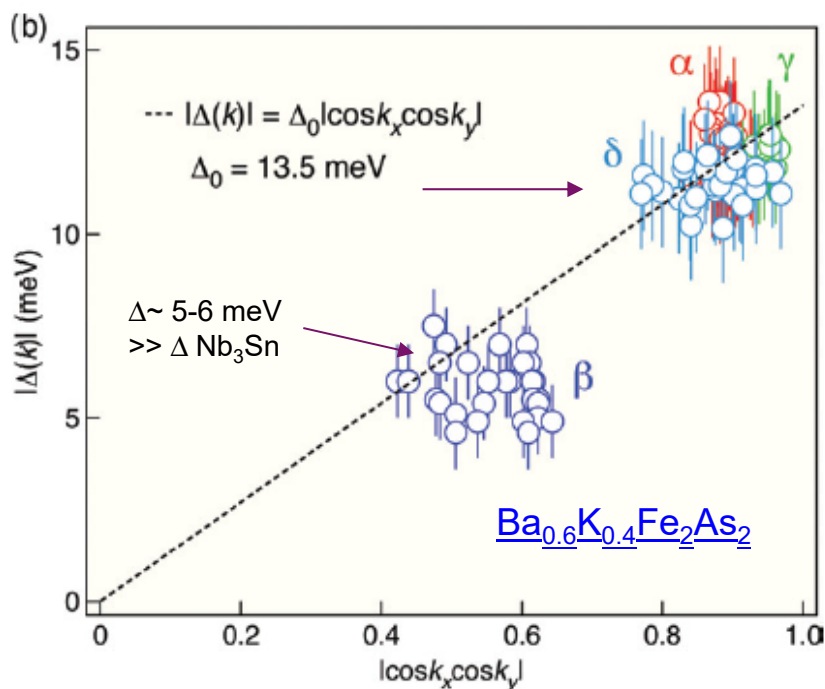


■ A lot of common with YBCO

- 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



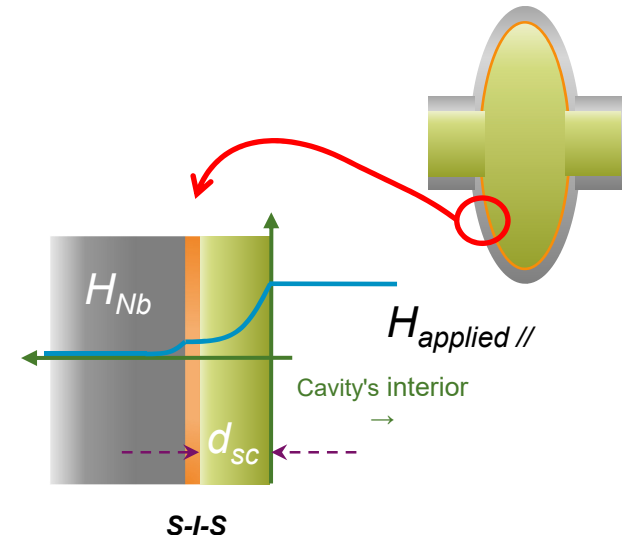
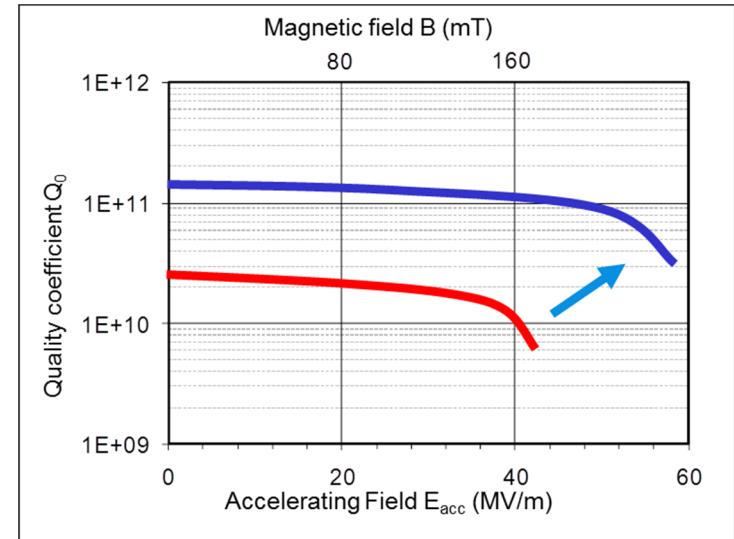
Structures proposed by A. Gurevich in 2006, SRF tailored

■ Dielectric layer

- Small $\perp$ vortex (short $\rightarrow$ low dissipation)
 - Quickly coalesce (w. RF)
 - Blocks avalanche penetration
- $\Rightarrow$ **Multilayer** concept for RF application

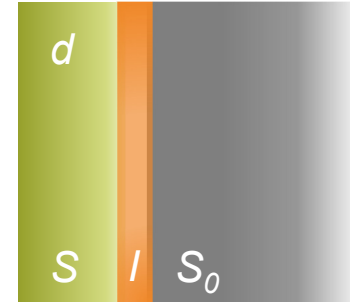
■ Nanometric I/S// layers deposited on Nb

- 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): $R_s^{NbN} \approx \frac{1}{10} R_s^{Nb}$
 $\Rightarrow Q_0^{multi} \gg Q_0^{Nb}$



■ 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



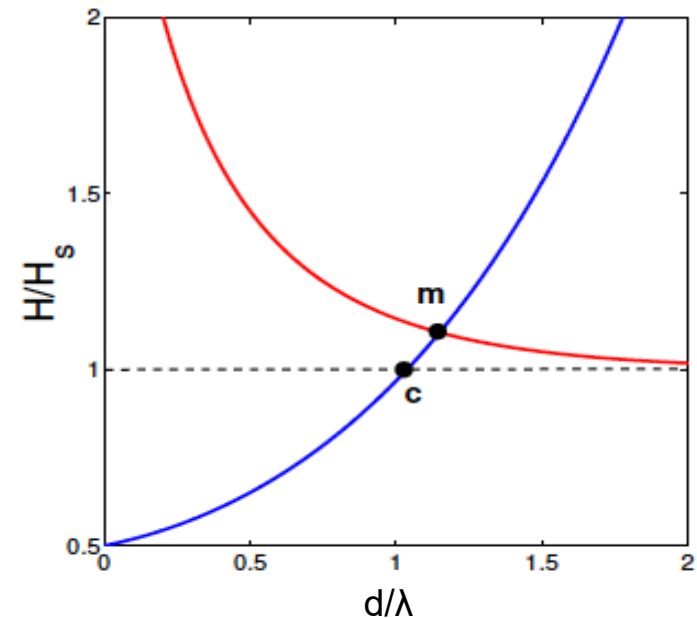
■ ∃ an optimum thickness and a maximum screening field!!!

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

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

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

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



H_m at the optimum thickness **exceeds the bulk superheating fields of both Nb and the layer material** because of counterflow induced by Nb in the S layers with $\lambda > \lambda_0$

For $\lambda \gg \lambda_0$, practically for $\lambda > 160$ nm for a S layer on the Nb cavity with $\lambda_0 = 40$ nm, H_m approaches the limit

$$H_m \rightarrow \sqrt{H_s^2 + H_{s0}^2}$$

- **Dirty Nb layer:** $H_c = 200$ mT, $H_s = 170$ mT, $l = 2$ nm, and $\lambda = \lambda(\xi_0/l)^{1/2} = 180$ nm
 $H_m = 288$ mT, $E_{acc} = 70$ MV/m, $d_m = 0.44\lambda = 79$ nm. +20% compared to $H_{SH}^{clean Nb} = 240$ mT
- **Nb₃Sn:** $H_s = 0.84H_c = 454$ mT and $\lambda = 120$ nm (moderately dirty):
 $H_m = 507$ mT, $E_{acc} = 120$ MV/m, $d_m = 1.1\lambda = 132$ nm x2 $H_{SH}^{clean Nb}$
- **Fe-pnictides on Nb:** $H_s = 0.84H_c = 840$ mT and $\lambda = 200$ nm:
 $H_m = 872$ mT, $E_{acc} = 206$ MV/m, $d_m = 1.78\lambda = 356$ nm x4 $H_{SH}^{clean Nb}$

Kubo et al, APL, 104, 032603 (2014); SUST, 30, 023001 (2017);
 (London calculations)

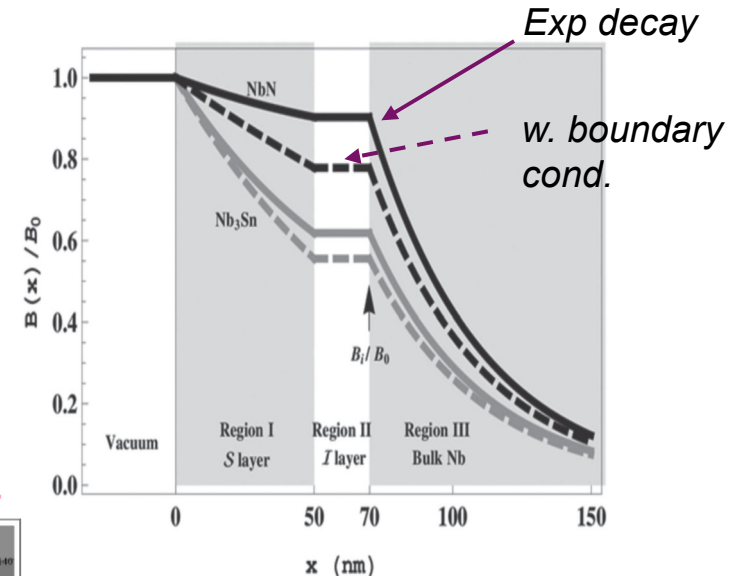
Posen, et al, Phys. Rev. Appl. 4, 044019 (2015)
 (London and GL calculations)

A. Gurevich, SUST 30, 034004 (2017)

First approach: trilayers

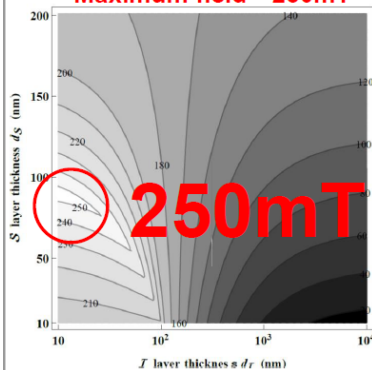
- Boundary conditions implemented (including effect of an insulating layer. finite thickness))
- H_{SH} determined initial y in London approx., further improved w. quasiclassical theory (valid @ $T < T_C$)
- Initially assume “perfect conditions”
- (bulk values, field // to surface)

A. Gurevich, T. Kubo



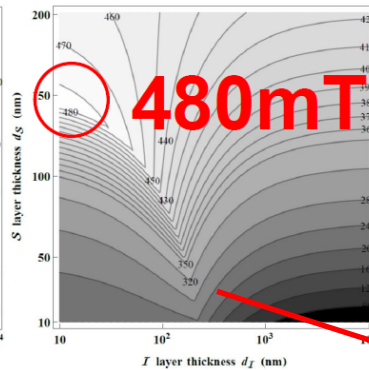
Dirty Nb // Nb

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



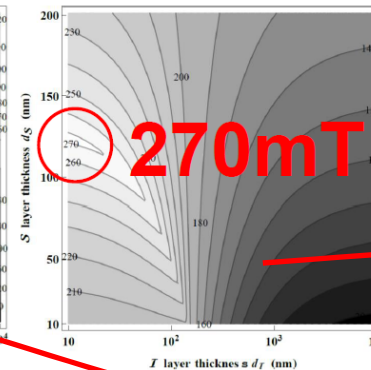
Nb₃Sn // Nb

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



NbN // Nb

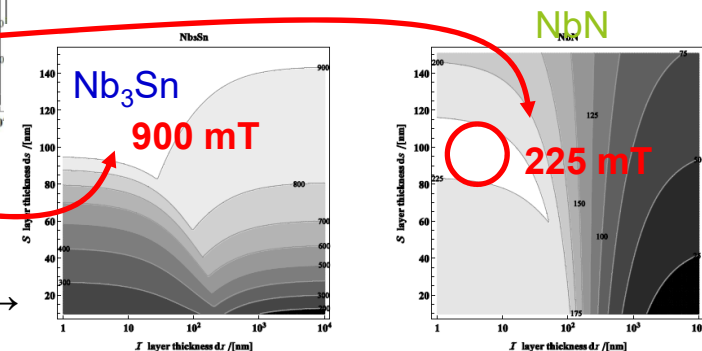
Optimum $d_s \sim 130\text{nm}$
Maximum field $\sim 270\text{mT}$



← Quasiclassical Approach,
Can be further improved...

[Kubo 2013-17]

Previous calculation w. London theory only: not realistic →

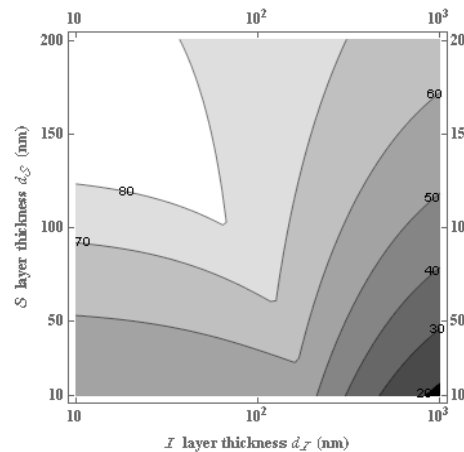
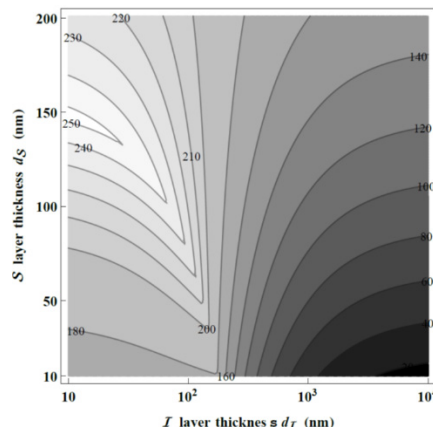


A. Gurevich, T. Kubo

■ Go for realistic condition

- layers present defects, non-negligible surface roughness, non-uniform thickness.
- $\rightarrow H_{SH}^S$ suppressed due to of the 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 dm^S can be determined w. surface topographical data

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



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

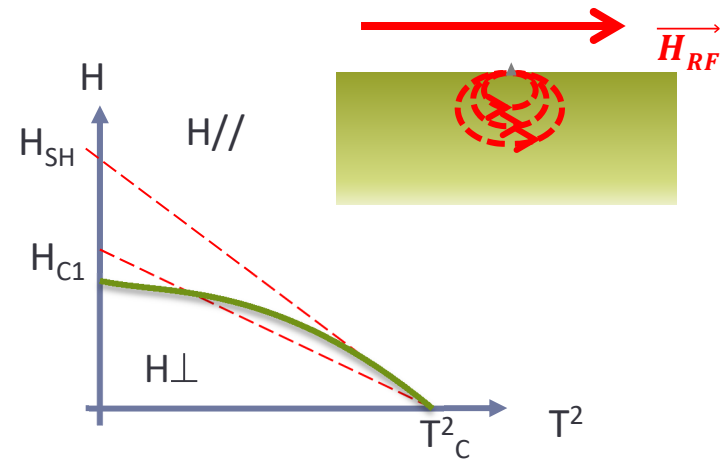
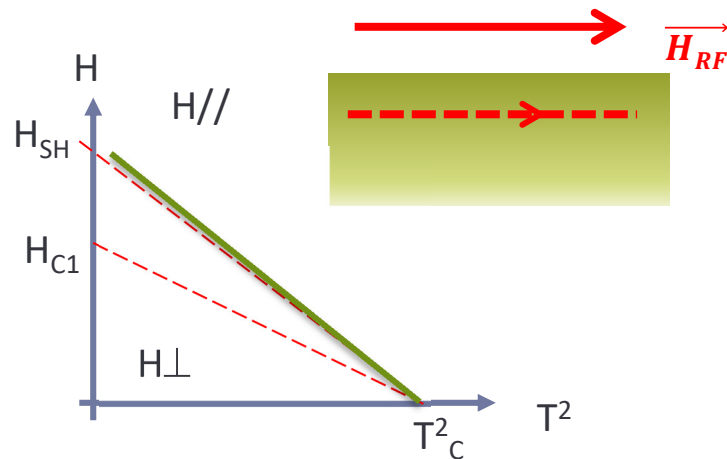
** e.g. morphologic
defects that allow earlier
vortex penetration*

See exp proof later on

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

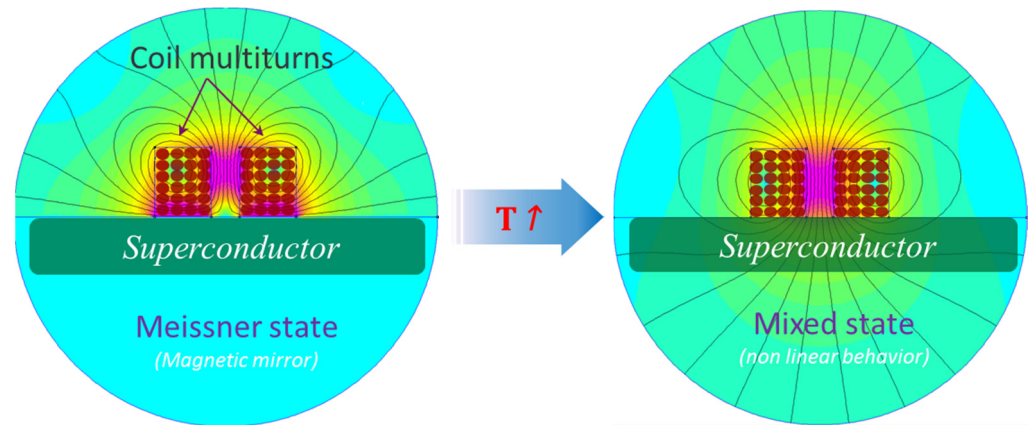


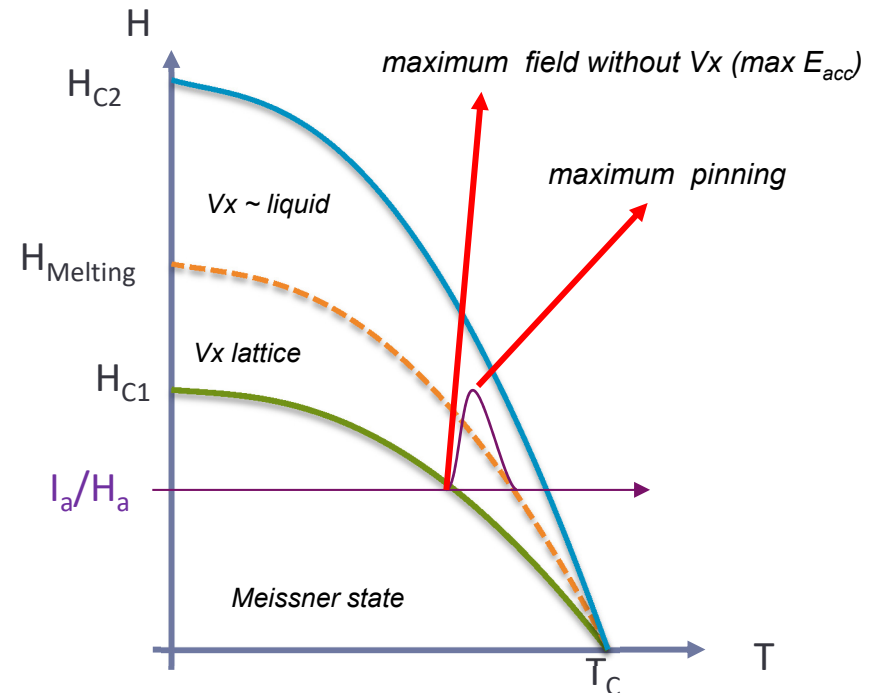
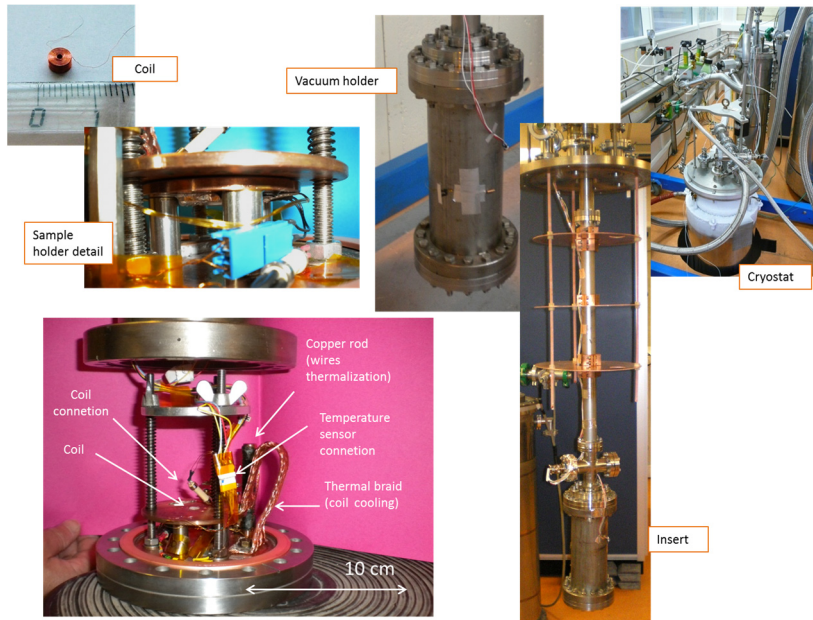
- Real world cavities behavior is dominated by a few number of defects
- It is very important to measure the penetration field of samples in realistic conditions



Local magnetometry

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

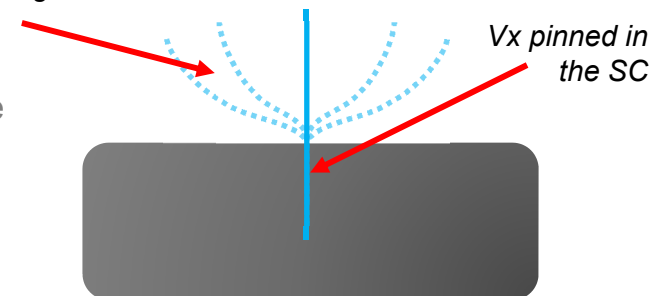




Low frequency $\equiv$ DC :

- $0 < H_a < H_{C1} \Rightarrow R=0$, Meissner state
- $H_{C1} < H_a < H_M \Rightarrow Vx$ are trapped, $R=0$, Campbell regime
- $H_M < H_a < H_{C2} \Rightarrow Vx$ 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 Vx trapped there (and length).

flux line moving in AC field



NbN coating by Magnetron Sputtering

■ NbN single layers series

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



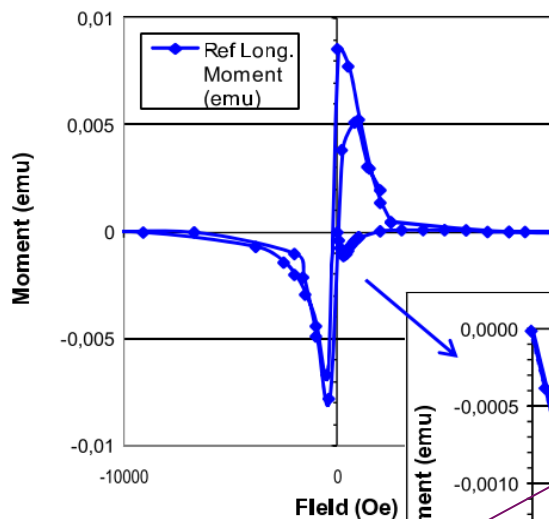
Nb (nm)	MgO (nm) Calc(actual)	NbN (nm) Calc(actual)	T _c (K)
250 [†]	14	0	8.9
250 [†]	14	25	15.5
500	10 (10.3)	50 (65)	15*
500	10 (8.4)	75 (72)	14.1*
500	10 (9.8)	100 (94)	14*
500	10	125	14.3*
500	10 (6.7)	150 (132)	15.9*
500	10 (10.4)	200 (164)	15*

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

*As determined with magnetometry, see below.

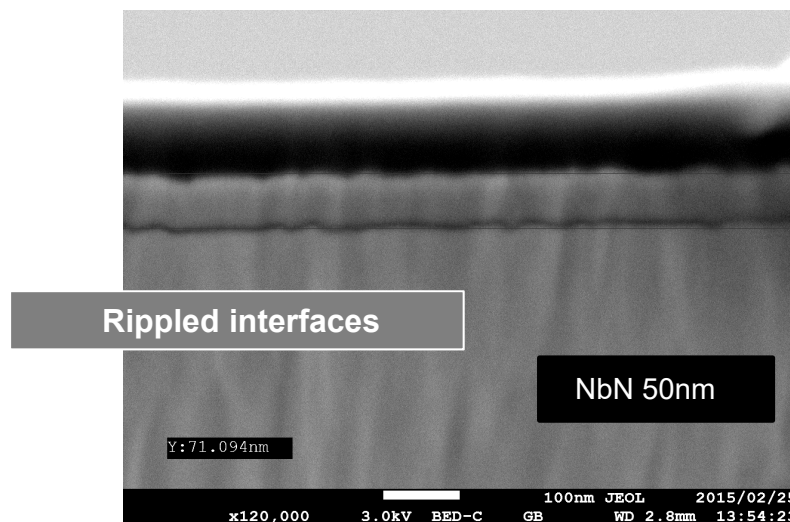
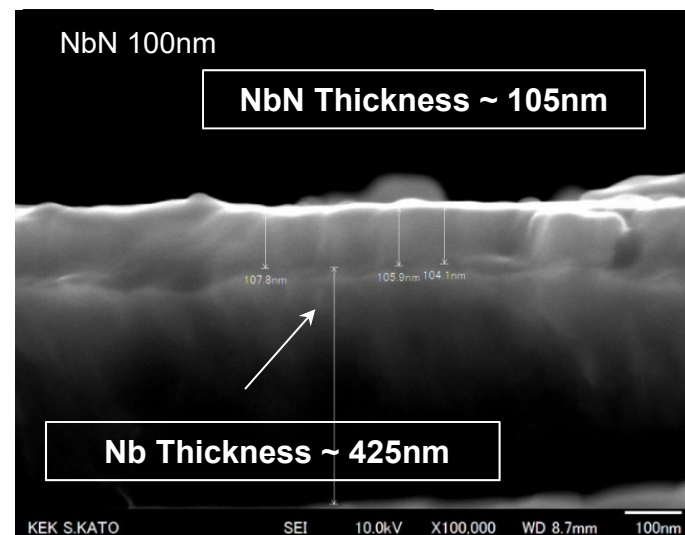
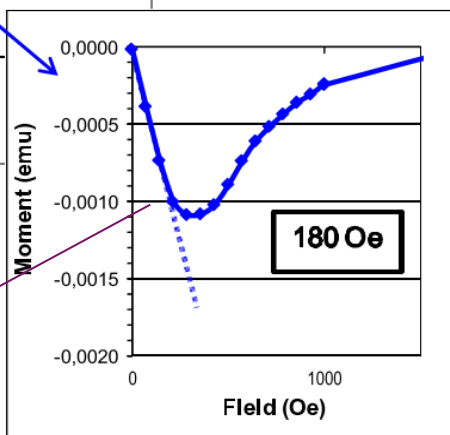
Typical defects...

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



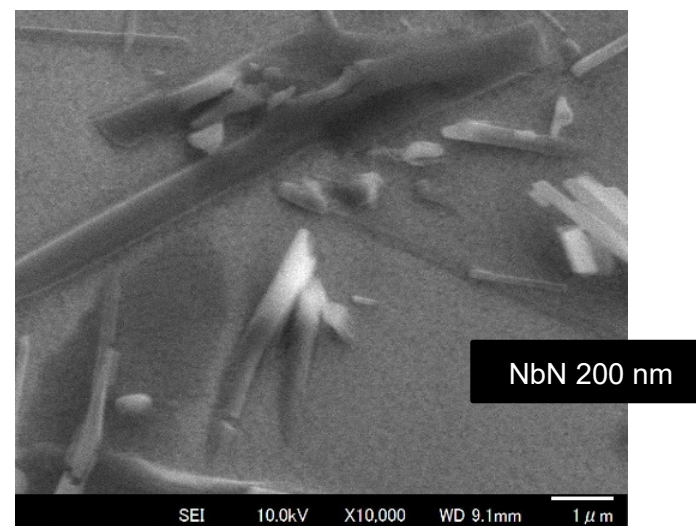
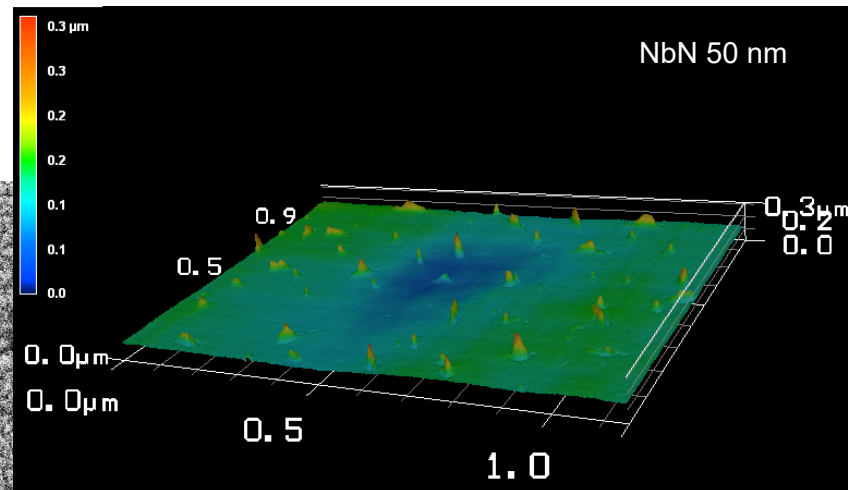
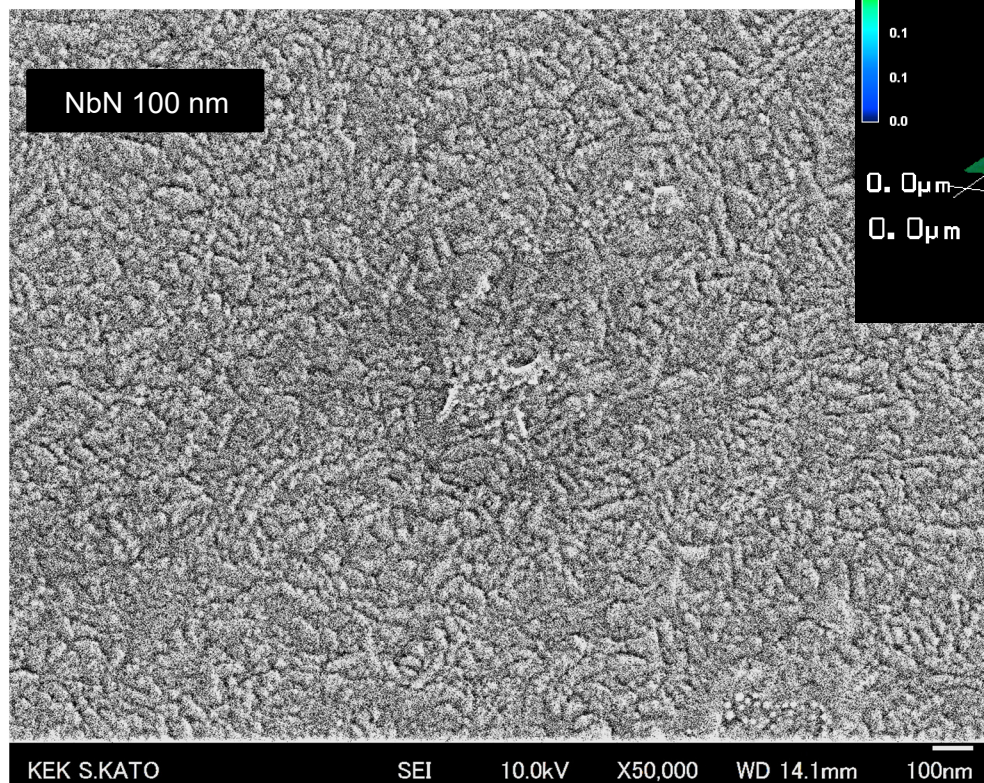
250 nm Nb on sapphire
DC Squid magnetometry @ 4.5 K

~ 18 mT
As expected for MS samples



Morphology

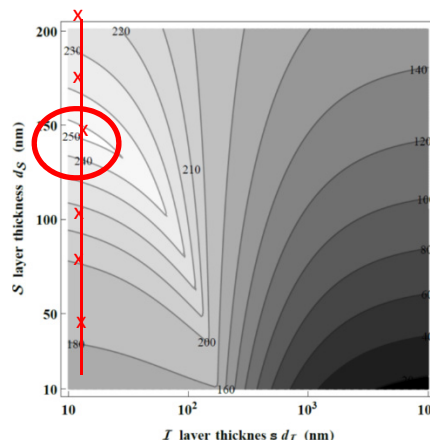
■ Rough surface



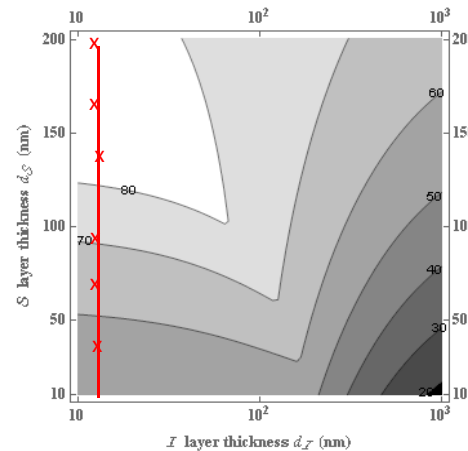
MgO needle ?
(capping material)

Theoretical predictions from T. Kubo (KEK)

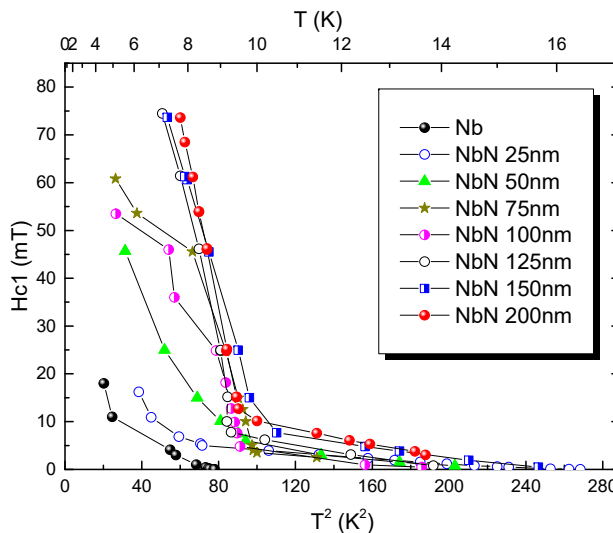
Ideal Nb substrate
with $B_{C1}=170 \text{ mT}$



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



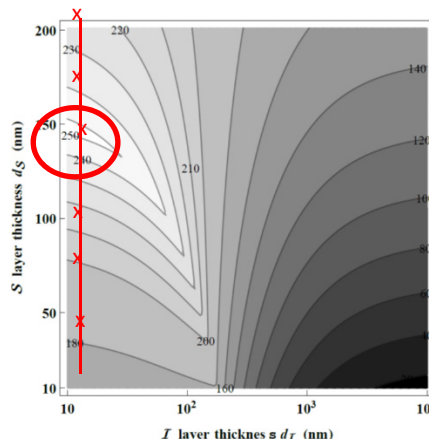
* e.g. morphologic
defects that allow earlier
vortex penetration See
SST paper cited earlier



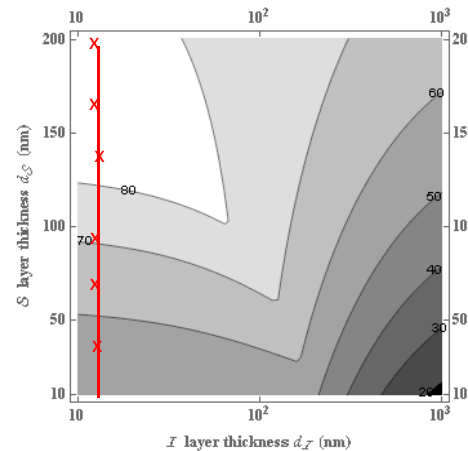
- The enhancement of the field penetration increases with thickness of NbN
- It reaches a saturation at thicknesses > 100 nm

Theoretical predictions from T. Kubo (KEK)

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

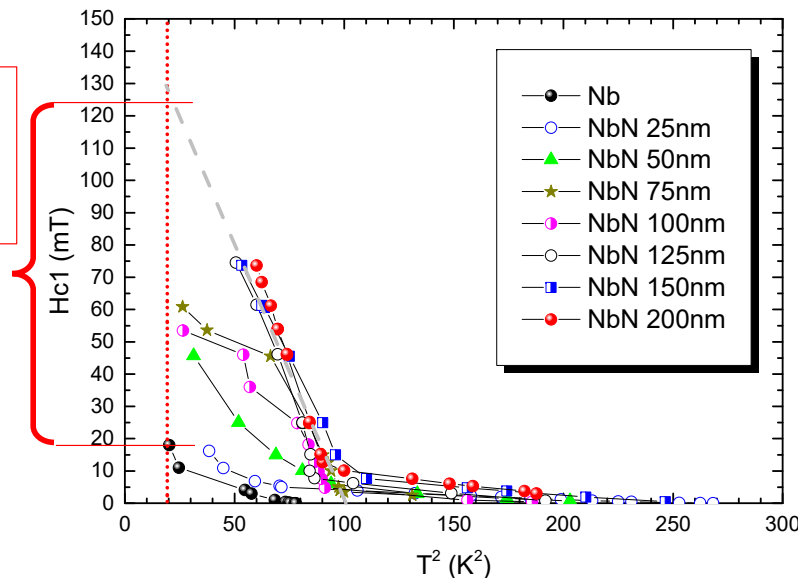


Nb with defects*,
with $B_{C1}=50$ mT



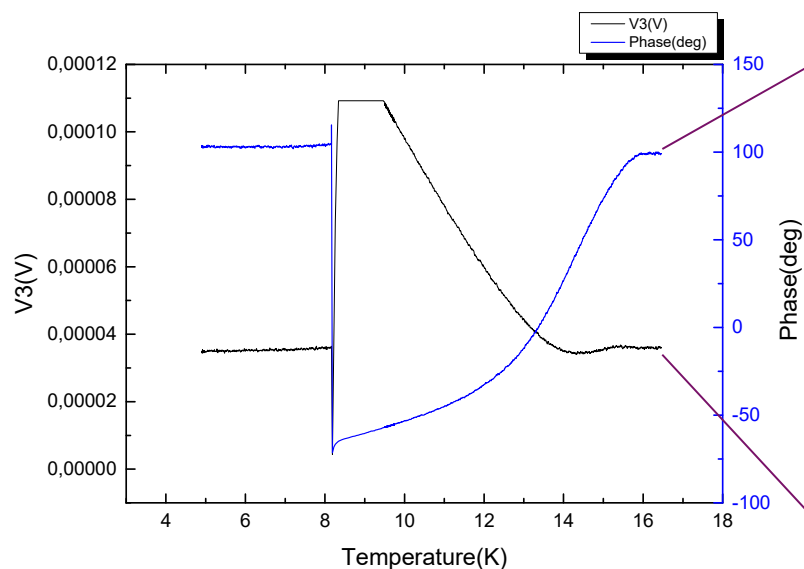
* e.g. morphologic
defects that allow earlier
vortex penetration See
SST paper cited earlier

@ 4.5 K
~ + 110 mT?
~25-30 MV/m
ILC shape

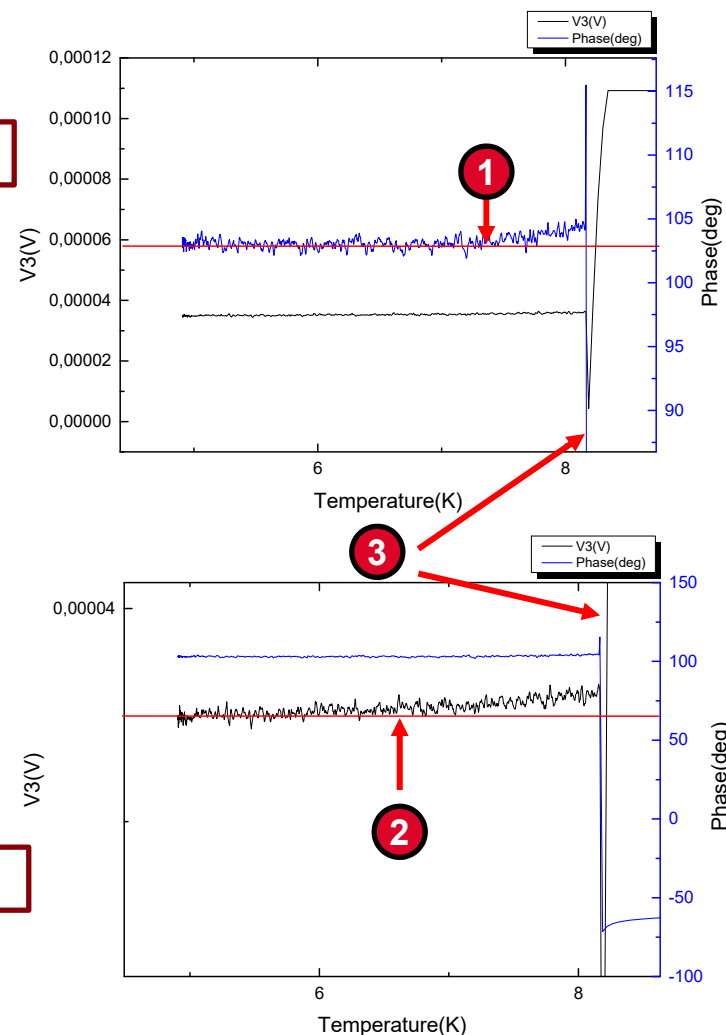


- The enhancement of the field penetration increases with thickness of NbN
- It reaches a saturation at thicknesses > 100 nm

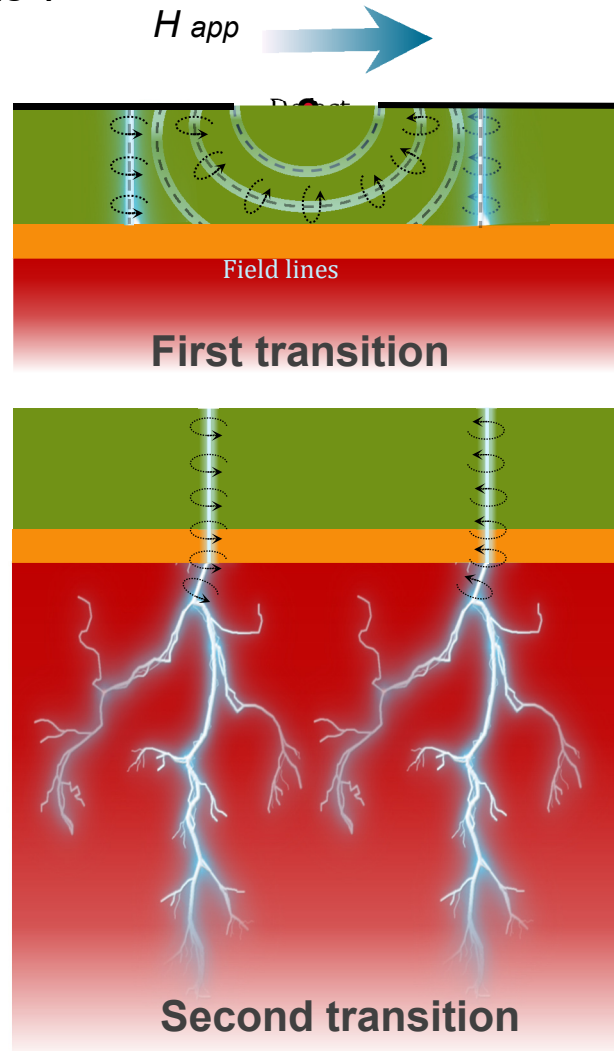
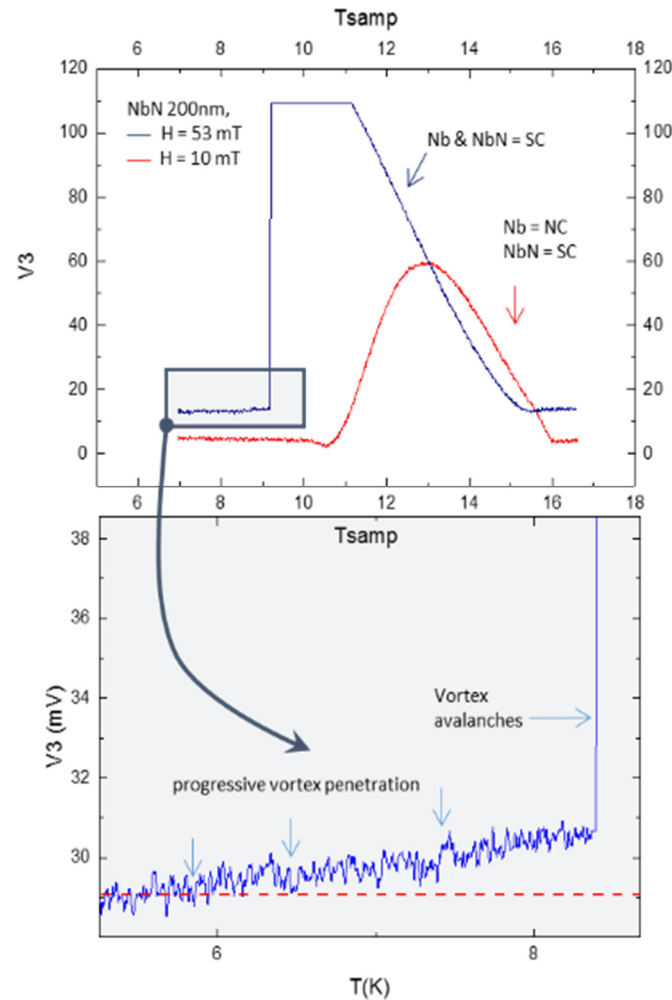
- For a given H_{appl} , we observe 3 $\neq$ transition temperatures



- $T \textcircled{1} \sim T \textcircled{2}$: within noise level
- $T \textcircled{3} \gg T \textcircled{2}$: dramatic transition



Why do we have two transitions ?



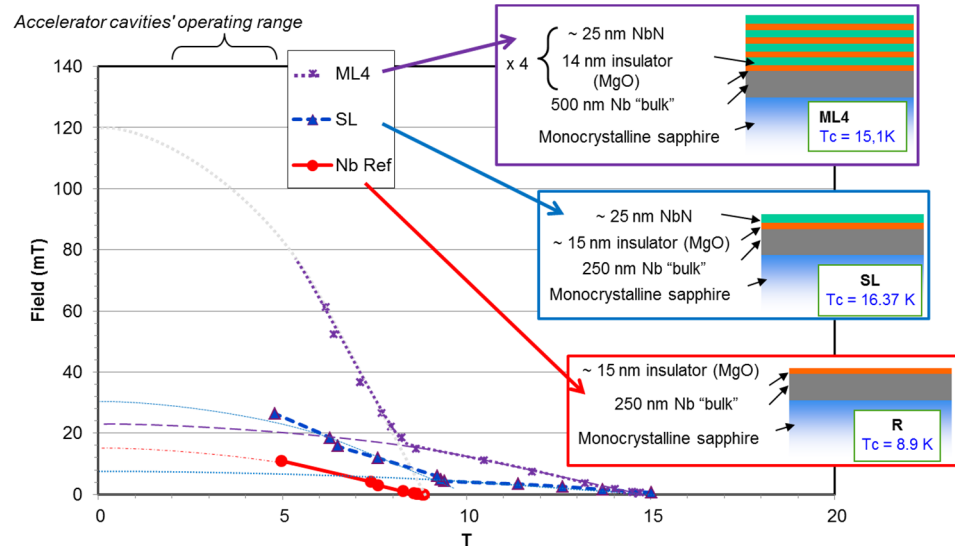
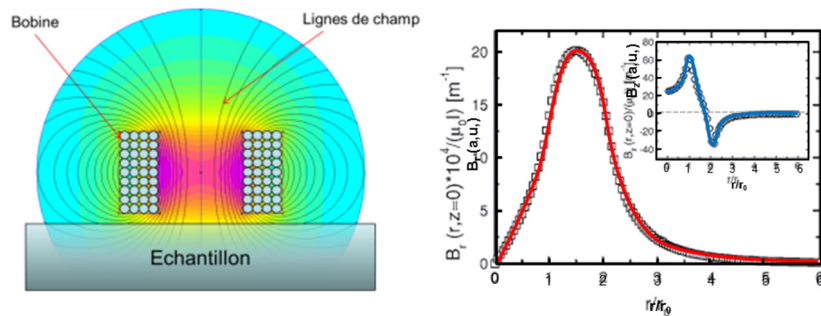
- Thin SC layer NbN
- Insulator MgO
- Thick SC layer Nb

- $H \parallel surface \Rightarrow$ 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 !!!**

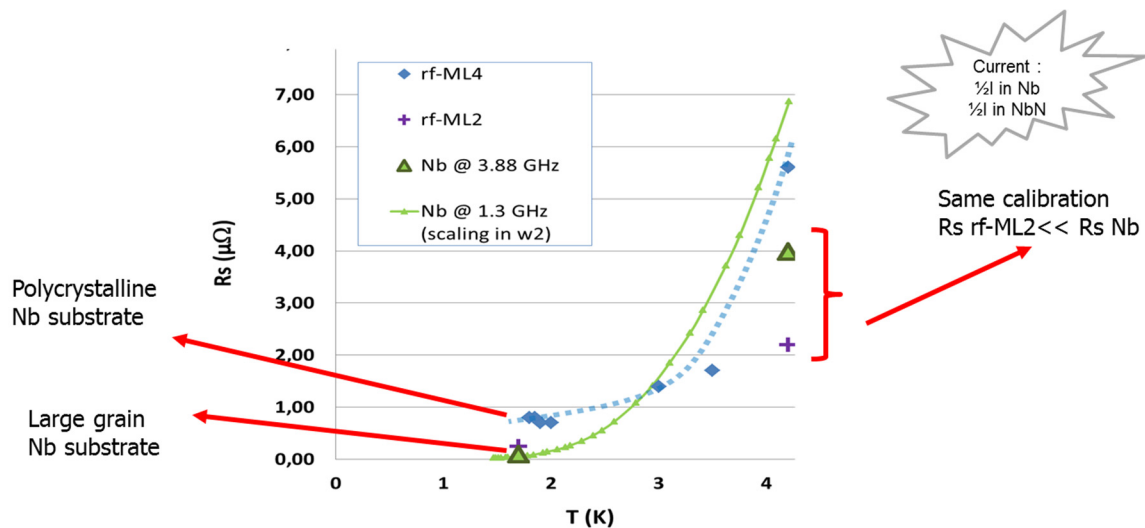
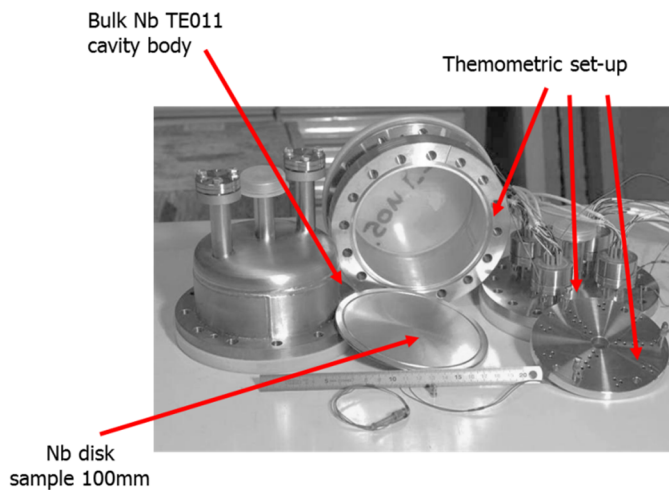
†B. Bean and J. D. Livingston, *Phys. Rev. Lett.* **12**, 14 (1964).

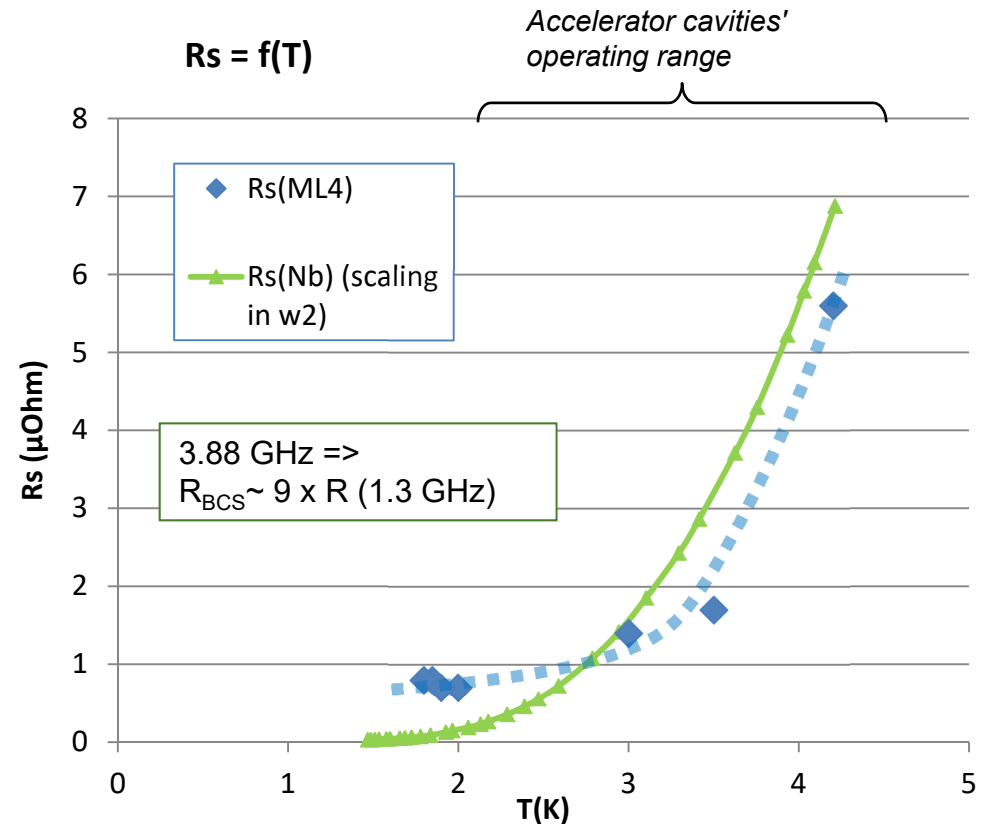
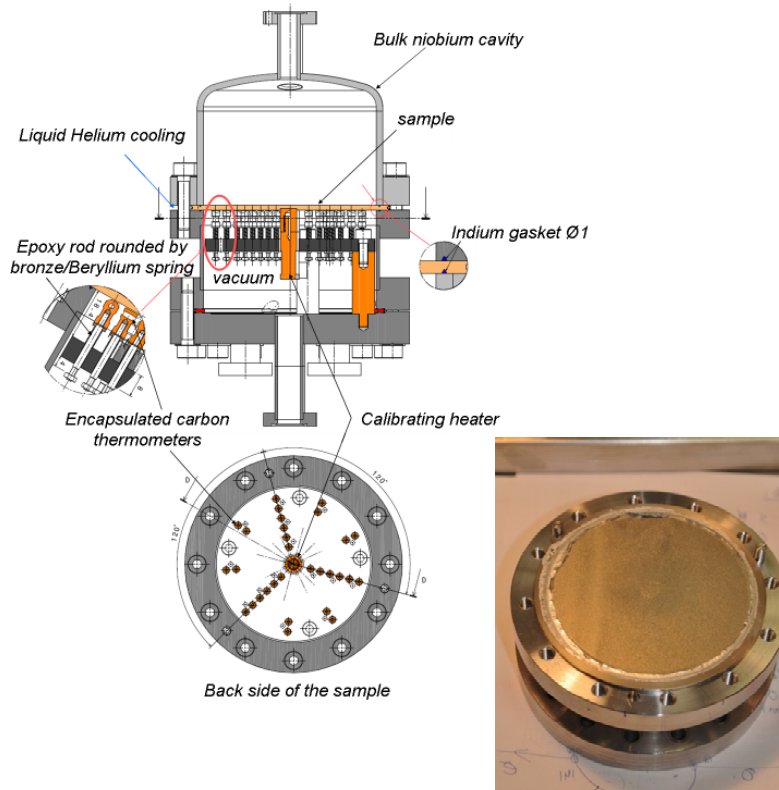
H_{c1} (E_{ACC}^{MAX}) AND R_s (Q_0^{MAX}) ESTIMATION

Local magnetometry:

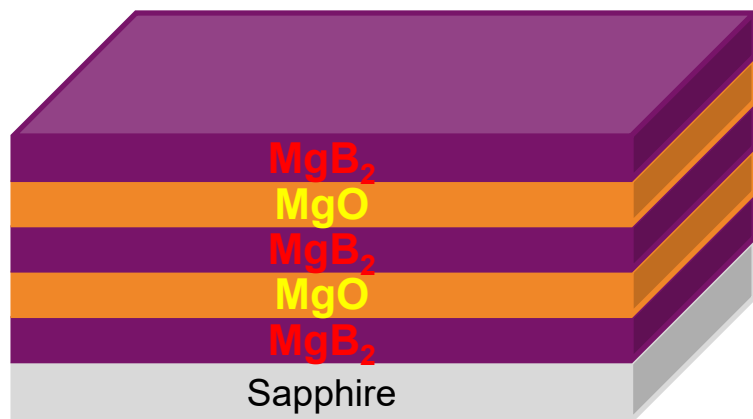


RF test (collaboration IPNO)





- 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.

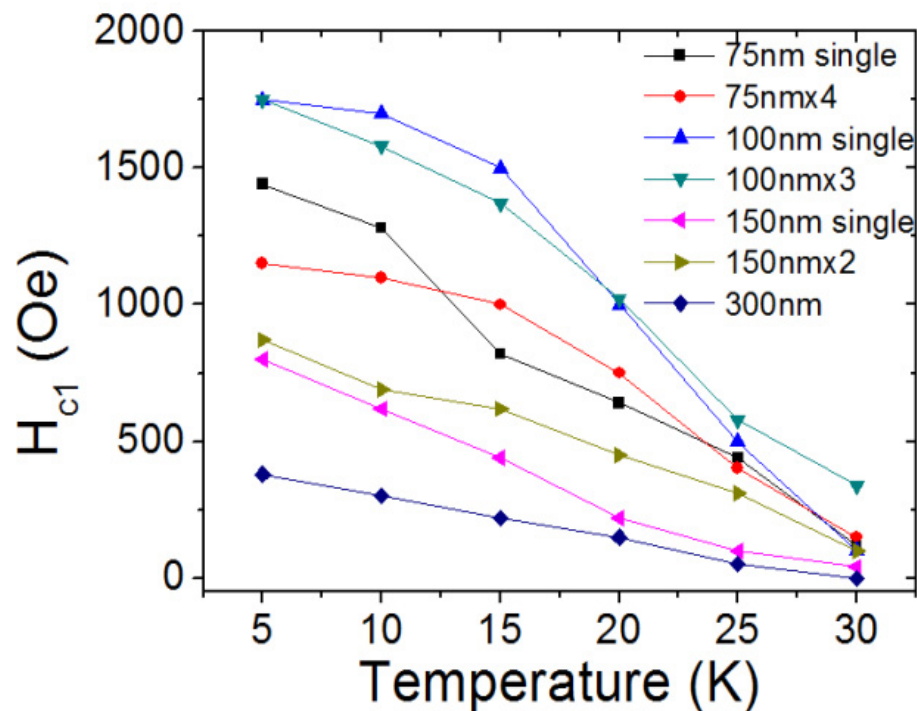


Alternating MgB₂-insulator structures have been fabricated on sapphire substrate:

- 40 nm MgO as insulating layer, sputtered.
- MgB₂ deposited by HPCVD *ex situ*. 150, 100, and 75 nm in thickness.

T_c near 40 K for 100 and 150 nm films. Lower for 75 nm film.

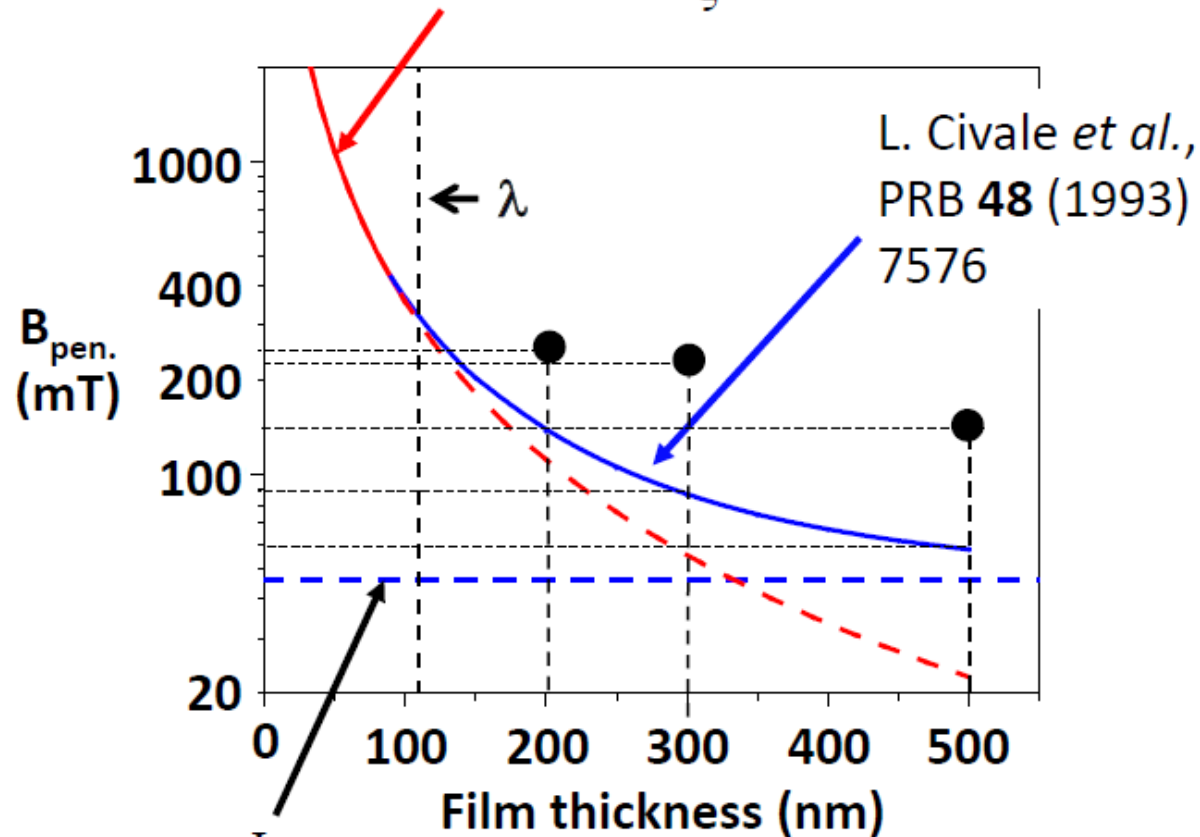
Multilayer films with thin MgB₂ layers show higher H_{c1} than the 300 nm film even though the total thickness are the same.



[Teng, Xi – Temple University]

B_{pen} data are higher than expected B_{c1} ($\lambda=110$ nm, $\xi=6$ nm)

$$H_{c1}(d \ll \lambda) \approx \frac{2\Phi_0}{\pi d^2} \ln \frac{d}{\xi} \quad \text{Gurevich, APL } \mathbf{88} \text{ (2006) 012511}$$

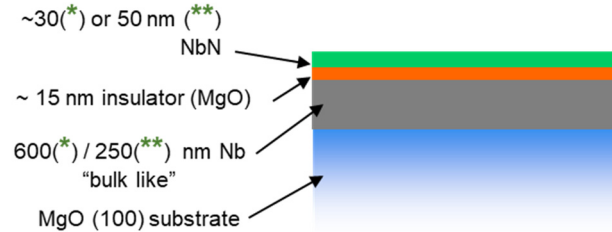


$$H_{c1}(d \gg \lambda) = \frac{\Phi_0}{4\pi\lambda^2} \ln \kappa \sim 46 \text{ mT}$$

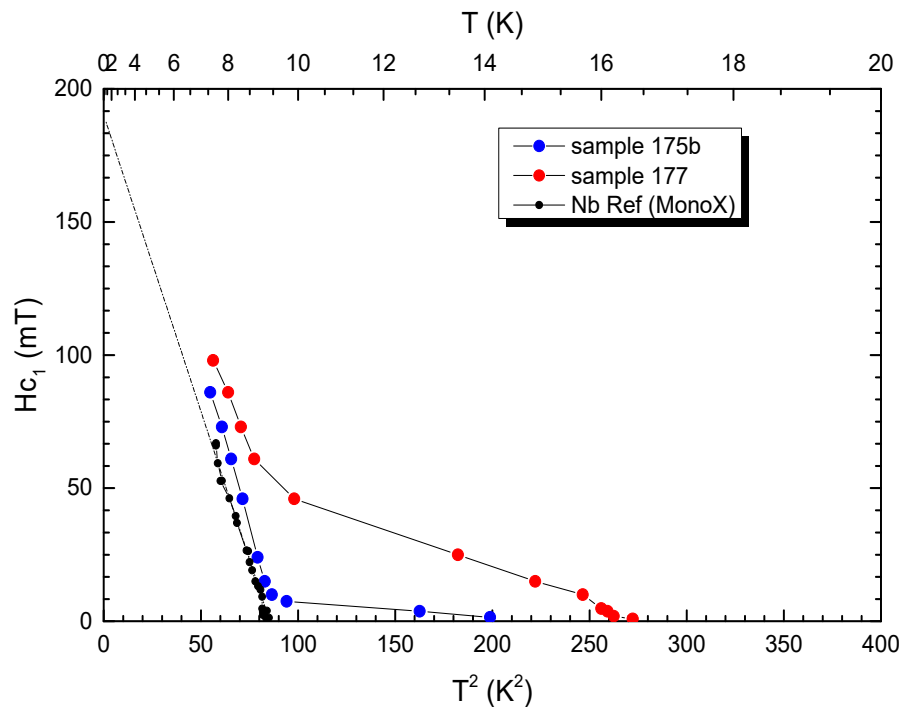
SRF2011, Chicago, IL, USA, 25-29 July 2011

Nb/MgO/NbN Samples

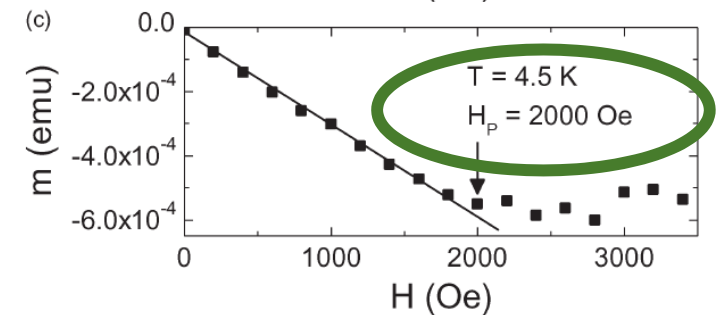
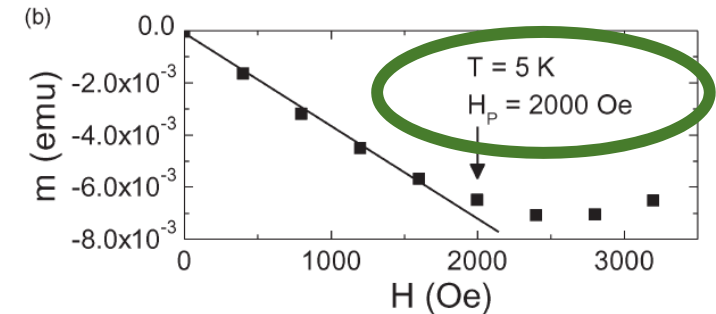
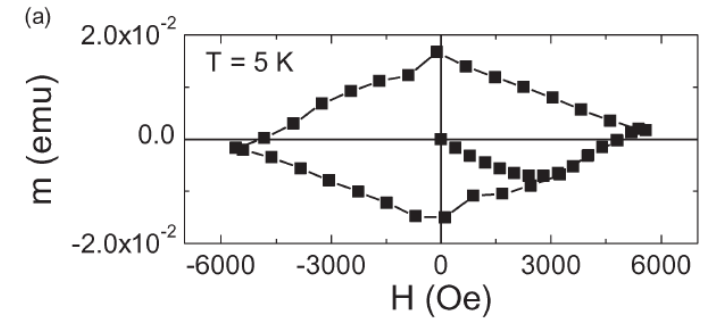
[Lukaszew, 2012]



Nb bulk /AlN/NbTiN Samples [AM Valente-Felicianno, TBP]



**



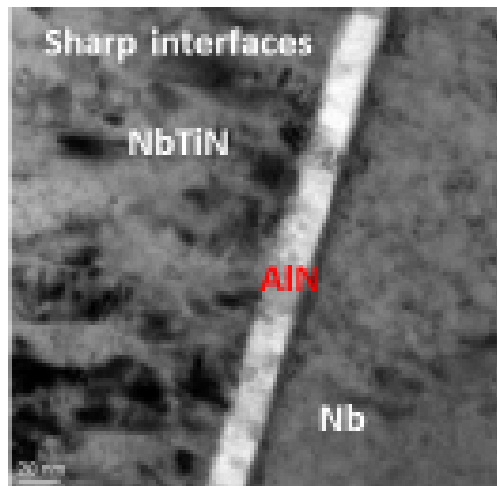
Compare with what is expected for bulk Nb : ~1300 Oe @ 4.5 K !

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.

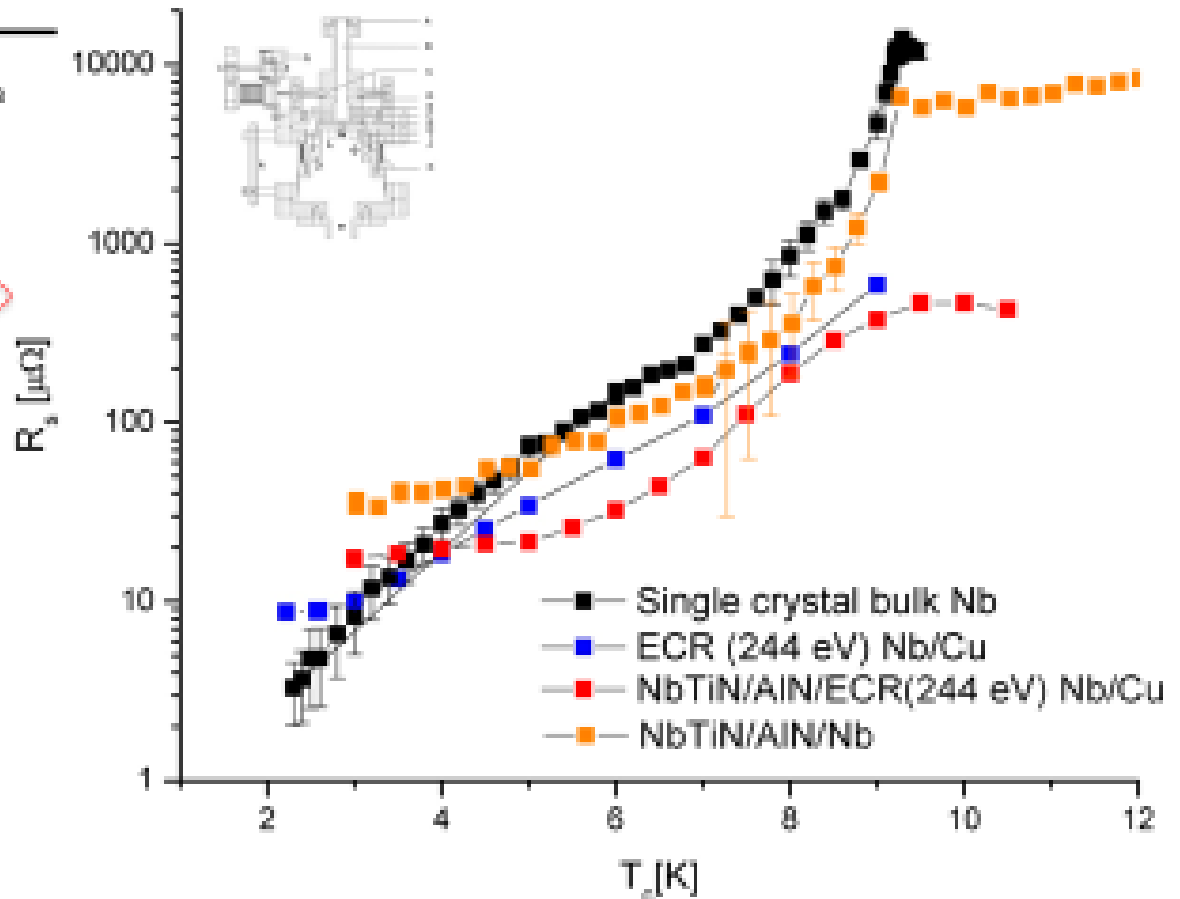
[A.M.Valente-Feliciano]

	AlN	NbTiN
N ₂ /Ar	0.33	0.23
Total pressure [Torr]	2×10^{-2}	2×10^{-2}
Sputtering Power [W]	100	300
Deposition rate [nm/min]	~ 2.5	~ 18
Thickness [nm]	20	150
T _c [K]	N/A	16.9



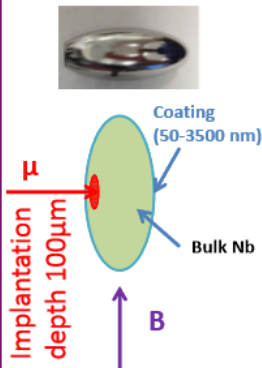
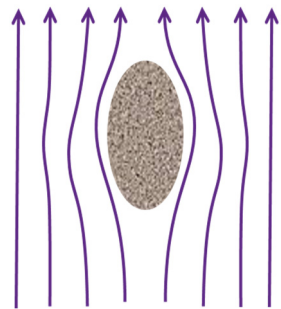
TEM cross-section (FIB cut) of NbTiN/AlN/Nb/Cu structure

RF Measurement in 7.5 GHz sapphire-loaded TE₀₁₁ cavity



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

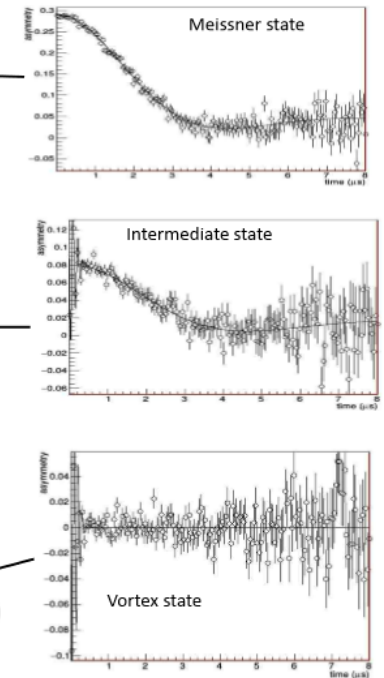
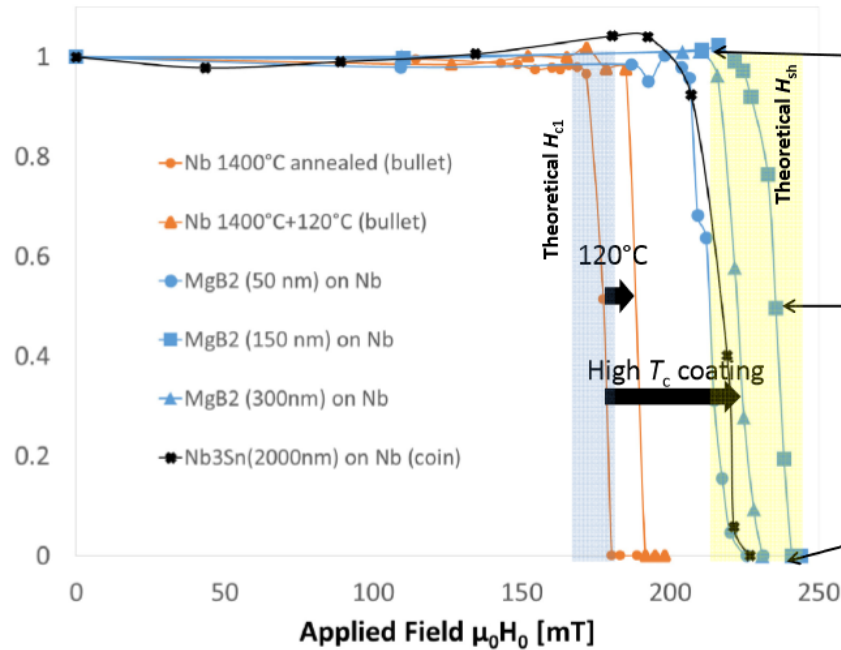
μ -SR



[Junginger, SRF 2017]

Field of first flux entry measurements

Volume fraction in the Meissner state at 0K

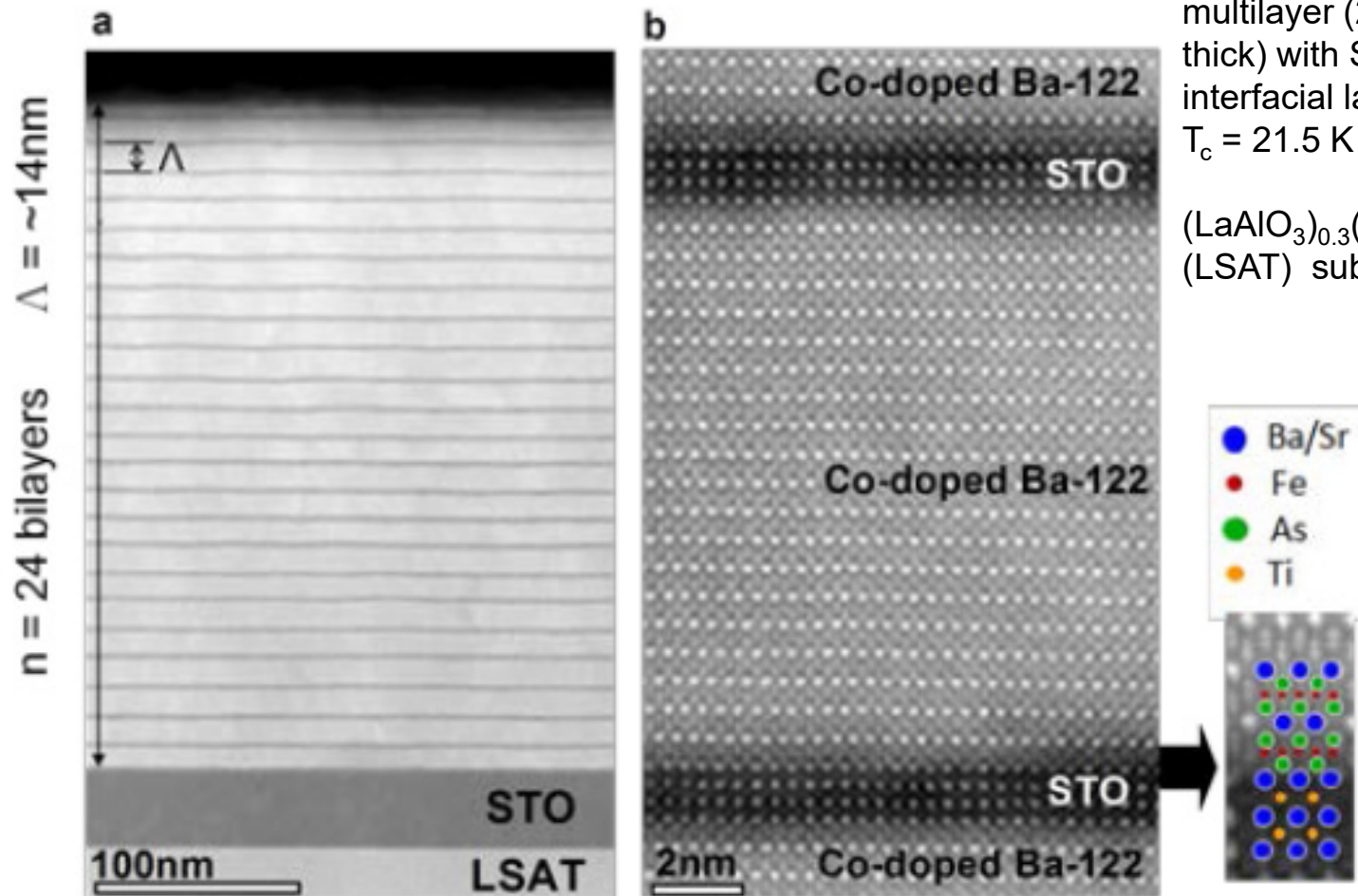


<https://arxiv.org/abs/1705.06383>

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]

S. Lee et al, Nature Materials, 12, 392 (2013)



PLD grown Co-doped BaF_2As_2 multilayer (24 layers, 13 nm thick) with SrTiO_3 (1.2 nm) interfacial layers
 $T_c = 21.5\text{ K}$

$(\text{LaAlO}_3)_{0.3}(\text{Sr}_2\text{TaAlO}_6)_{0.7}$ (LSAT) substrate:

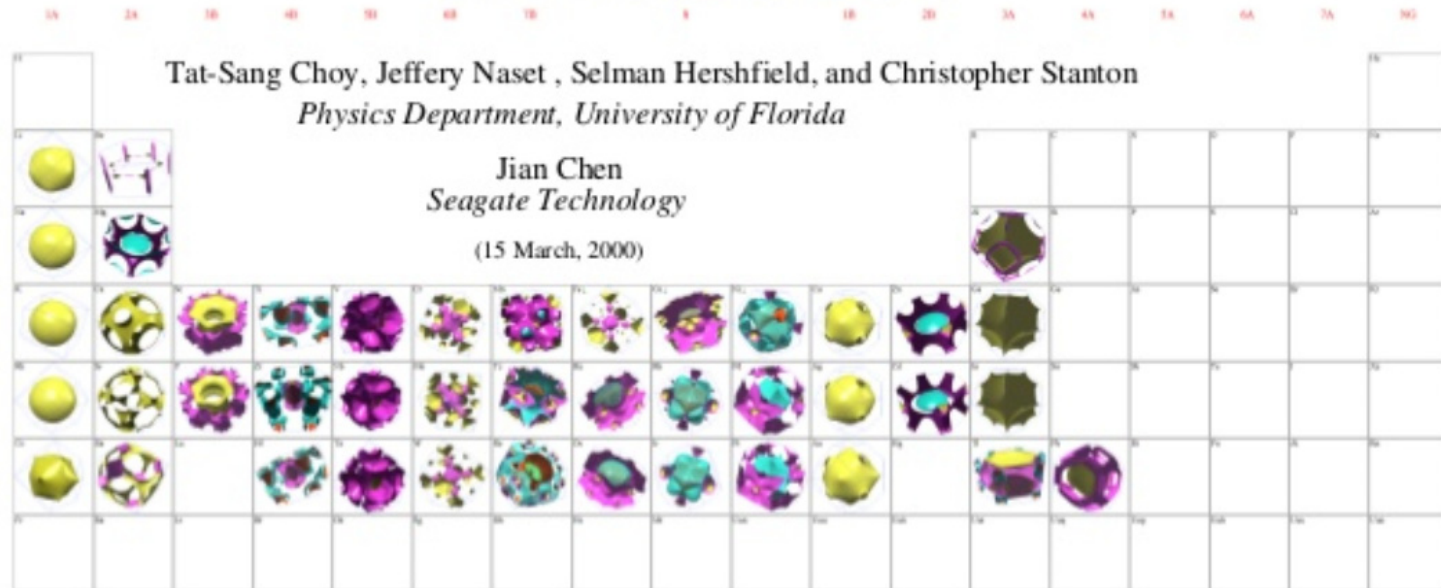
Group of C-B Eom, U Wisconsin

First high quality epitaxial films have been grown by C.-B. Eom (UW)

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

Periodic Table of the Fermi Surfaces of Elemental Solids

<http://www.phys.ufl.edu/fermisurface>



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Ferromagnets:



Alternate Structures:



Source of tight binding parameters (except for fcc Co ferromagnet): D.A. Papaconstantopoulos, *Handbook of the band structure of elemental solids*, Plenum 1986.

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