

SRF 2013 Tutorial

CAEN, FRANCE

BEYOND NIOBIUM

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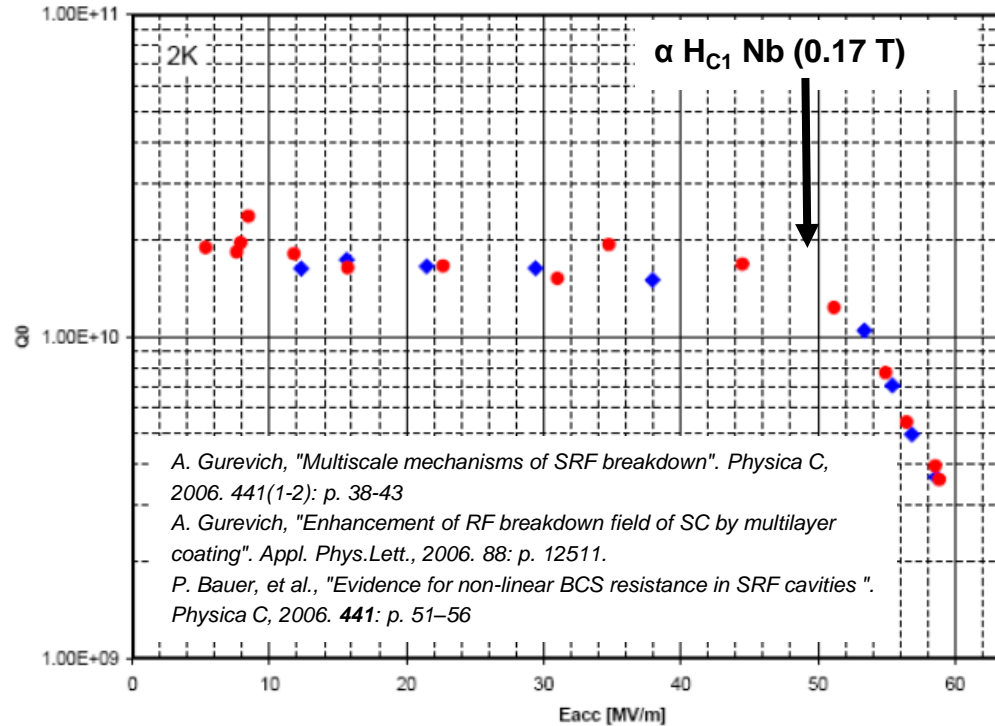
Outline

- Motivation
- Which Superconductors for SRF Cavities?
- Nb compounds: NbN, NbTiN
- A15 Compounds: Nb₃Sn, V₃Si, ...
- MgB₂
- Oxypnictides
- SIS Multilayer Structures
- Concluding Remarks

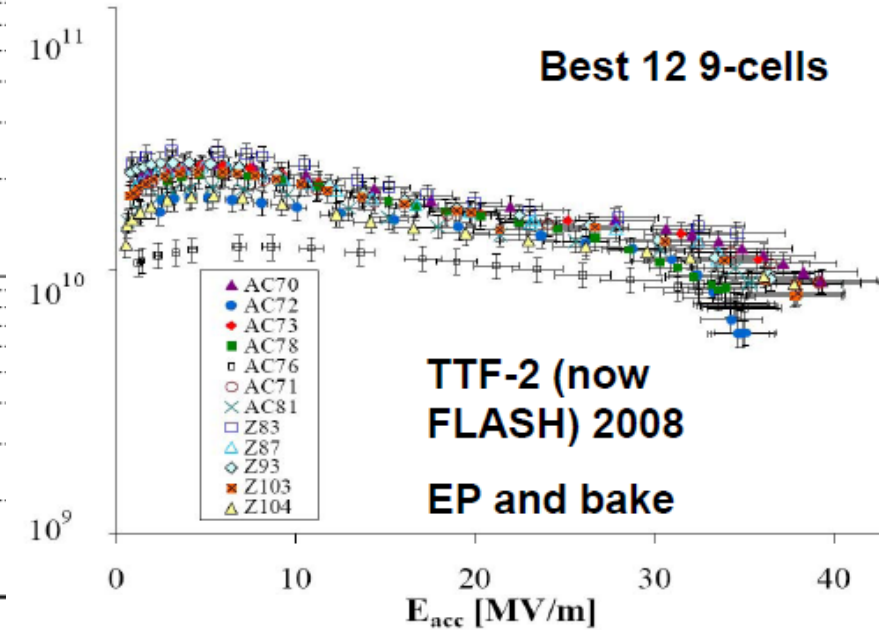
Why looking beyond Nb?

Nb has the highest critical temperature T_c ($\approx 9.25\text{k}$) and the highest lower critical magnetic field H_{c1} ($\approx 180\text{ mT}$) of any elemental superconductor

Cornell 60 mm aperture re-entrant cavity LR1-3 March 14, 2007



Data from the Jlab, KEK, Cornell and other groups



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 $H_{max} = H_C$
 For further improved cavity RF performance, innovation needed

Possibilities to use higher performance superconductors other than Nb?

Looking beyond Nb – Potential Benefits

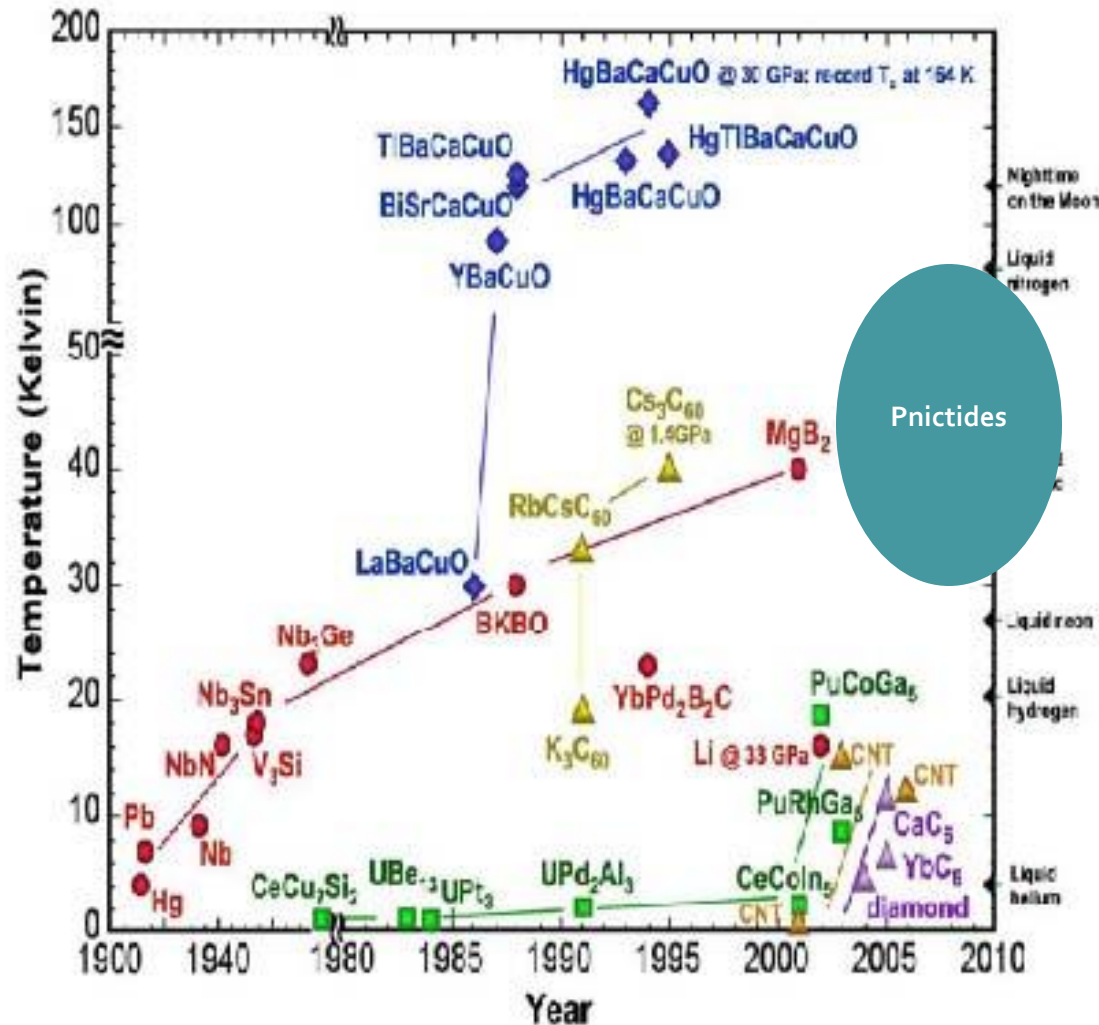
- Reduced material costs
 - Use of inexpensive, highly formable materials with higher thermal conductivity such as Cu or Al
- Simplified engineering, fabrication and assembly
 - Separation of cavity structure from superconducting surface
 - Maximum flexibility & largest variety of options in design of integrated cavity/cryostat structures
- Higher gradients

Increasing gradients reduces capital expense of cryomodules – potentially several \$100M savings & minimize conventional facilities
- Lower RF losses

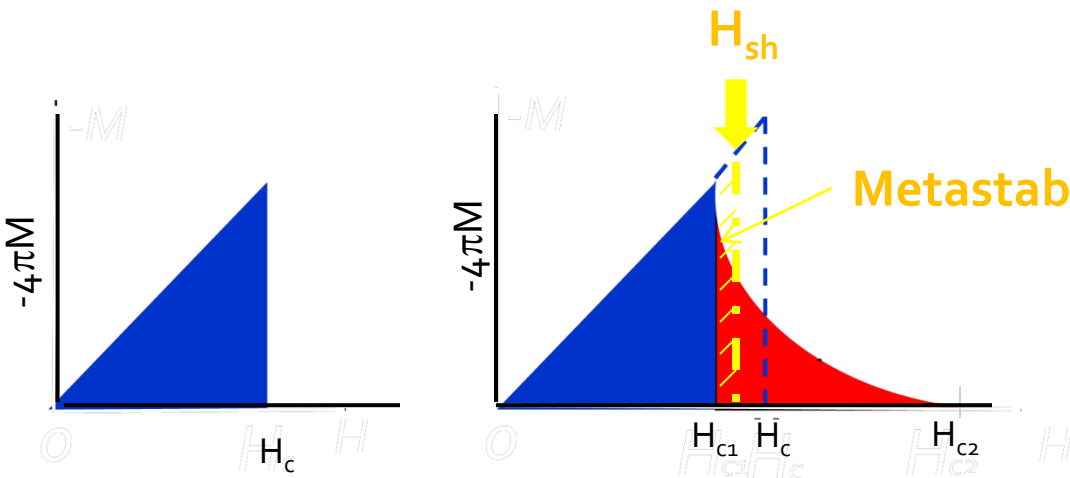
Low loss (high Q) cavities reduce He costs, >\$10M potential capital savings, and several \$M/year in operating costs
- Potentially higher operating temperatures (>4.2K)

Which materials are suitable for SRF cavities?

Highest $T_c = 164\text{K}$ (under GPa)



Critical Field



Boundary between Type I and Type II determined by the Ginzburg-Landau parameter

$$\kappa = \lambda/\xi$$

H_c is the thermodynamic critical field &

$$H_c = H_{c2} / \sqrt{2\kappa}$$

(for type-II superconductors).

Type-I

Type-II

- Meissner state at $0 < H < H_{c1}$
- Mixed vortex state at $H_{c1} < H < H_{c2}$
- Exponentially small R_s at $H < H_{c1}$ ($Q = 10^{10}-10^{11}$)
- Drastic Q drop due to vortex dissipation at $H > H_{c1}$

The superheating field H_{sh} is the field up to which the Meissner state metastably persists above H_{c1}

For type-II superconductors, at $T = 0$ in the clean limit with a Ginzburg-Landau parameter $\kappa = \lambda/\xi \gg 1$, H_{sh} has been calculated to be $\sim 0.75H_c$.

$$H_{RFcrit} \approx H_{sh}$$

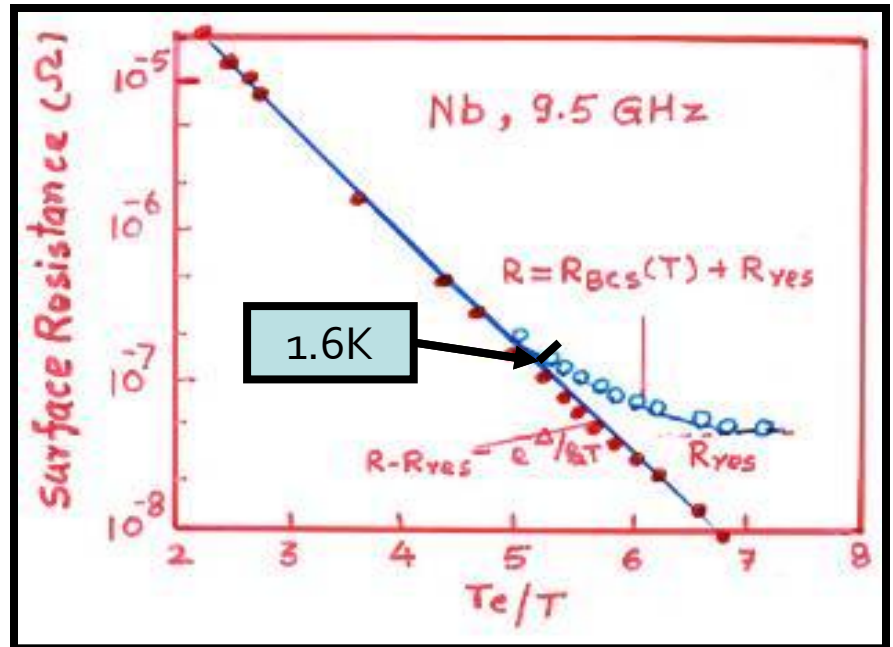
Surface Resistance

The power dissipated per unit area of SC in RF regime $P = R_S \cdot \frac{H^2}{2}$

and

Surface Resistance

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



V. Palmieri, 10th Workshop on RF Superconductivity Proceedings, Tsukuba 2001 (Noguchi)
"New materials for superconducting radiofrequency cavities"

BCS Surface Resistance R_{BCS}

If $T < T_c / 2$, for dirty limit superconductors

$$R_{BCS} \cong \frac{R_n}{\sqrt{2}} \left(\frac{\eta\omega}{\pi\Delta} \right)^{\frac{3}{2}} \frac{\sigma_1}{\sigma_n} = A \sqrt{\rho_n} e^{-\frac{\Delta}{K_B T}} (1 + O(\Delta, \omega, T))$$

A constant weakly dependent on material

ω = RF frequency

ρ_n = Normal State conductivity

Δ = Superconducting gap

T_c = Transition Temperature

dependence on ρ_n and T_c represents an immediate criterion for selecting the most favorable candidates for cavities

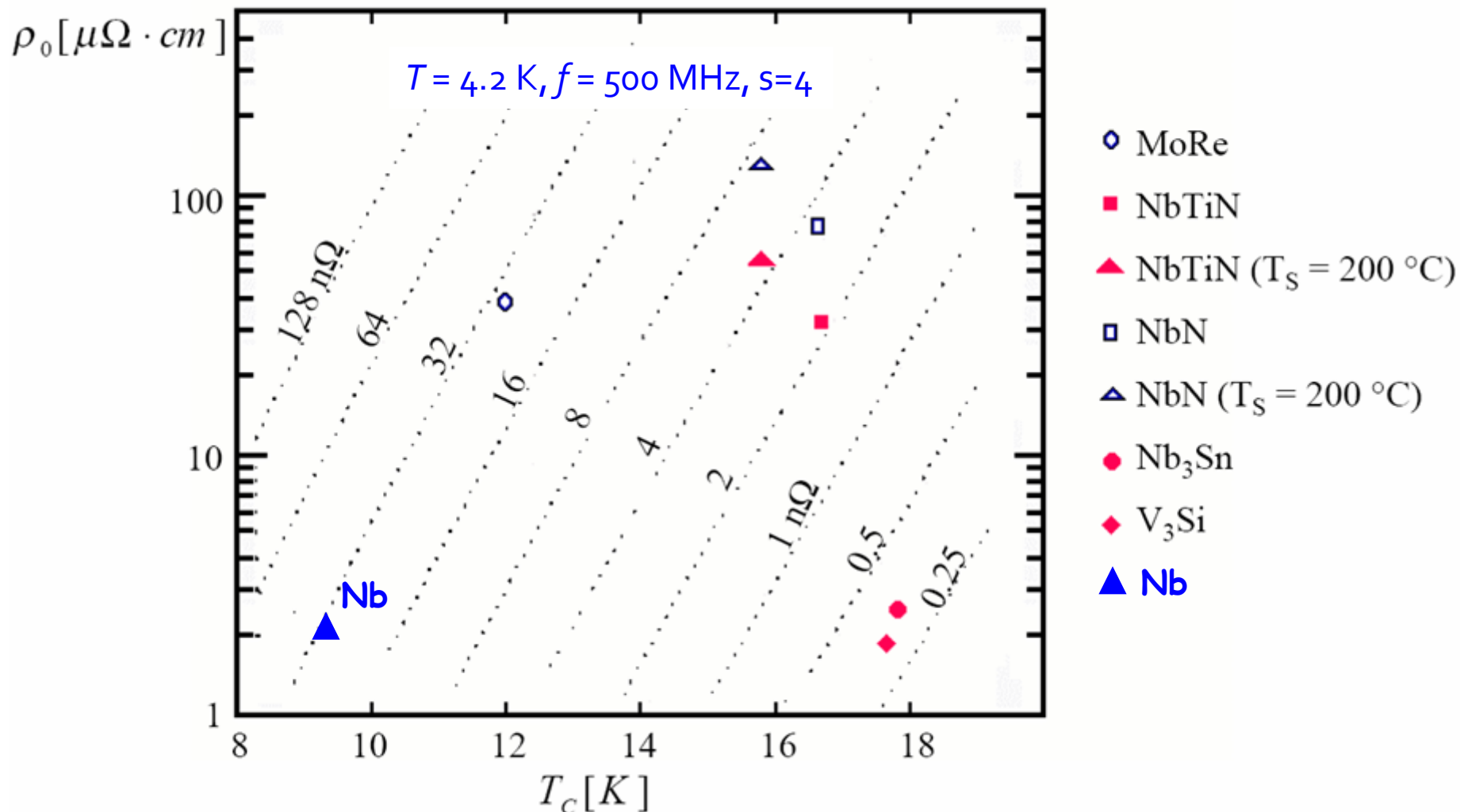
The higher T_c ($= 0.57\Delta$), the smaller the BCS surface resistance

Material with high normal state conductivity and high T_c (high superconducting gap Δ) should be selected

Other materials than niobium

R_{BCS} versus ρ_0, T_c

Vaglio, *Particle Accelerators* 61, 391 (1998)



Residual Resistance R_{res}

Temperature independent

Contributions to residual losses:

Intrinsic:

Inhomogeneity, Metallic Inclusions within I, Grain Boundaries, Oxides

Extrinsic:

Trapped Flux during cooling (can be avoided)

Variety of phenomena involved



Not one formula predicting R_{res}

From literature

Empirically, R_{res} found proportional to at least $\sqrt{\rho_n}$

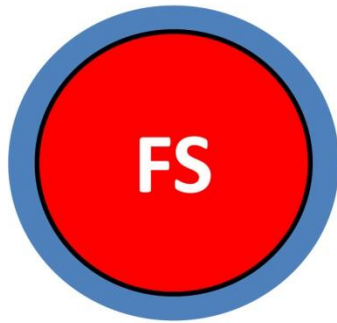
For two materials with the same R_{BCS} and different T_c and r_n ,
the one with the smallest ρ_n should have the smallest R_{res}

Metallic behaviour is favored

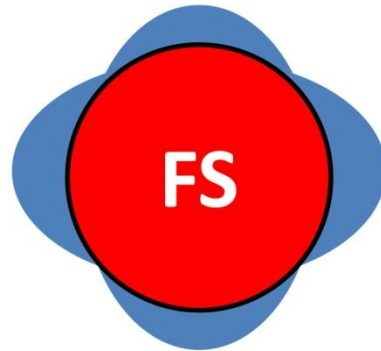
The gap symmetry is important

- The high Q required by SRF cavities results from the exponentially small BCS surface resistance

$$R_s = \frac{A\omega^2}{T} \exp\left(-\frac{\Delta}{T}\right) + R_{res}$$



s-wave: all conventional BCS superconductors, ferropnictides



d-wave: high- T_c cuprates, heavy fermions, borocarbides ferropnictides

- All high- T_c cuprates are d-wave SC with nodes in the gap
- Power-law R_s and gap suppression by nonmagnetic impurities:

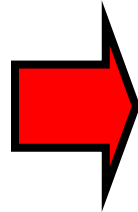
$$R_s \propto T^\alpha, \quad \alpha \cong 2-3$$

d - wave cuprates, or heavy fermions cannot compete with s-wave LTS at low T

Criteria of choice

THERE IS NO IDEAL SUPERCONDUCTOR FOR CAVITY
CHOICE IS BASED ON COMPROMISE

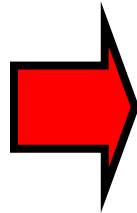
For low RF losses



high T_c

Metallic behavior in the normal state ,
small ρ_n

For high gradients



High H_{sh} , H_c ; small κ

Possible Choices among Superconducting Materials

Material	Critical Temp. T_c [K]	Normal-state resistivity ρ_n ($\mu\Omega\text{cm}$)	Critical Field $H_c(o)$ [T]	Lower Critical field $H_{c1}(o)$ [T]	Upper Critical field $H_{c2}(o)$ [T]	Penetration depth $\lambda(o)$ [nm]	Type
Nb	9.22	2	0.2	0.18	0.28	40	II
Pb	7.2		0.08	N/A	N/A	48	I
NbN	17	70	0.23	0.02	15	200	II, B1 comp.
NbTiN	17.5	35		0.03		151	II, B1 comp.
Nb ₃ Sn	18.3	20	0.54	0.05	30	85	II, A15
V ₃ Si	17				24.5	179	II, A15
Mo ₃ Re	15		0.43	0.03	3.5	140	II, A15
MgB ₂	40		0.43	0.03	3.5	140	II- 2 gaps
YBCO	93		1.4	0.01	100	150	d-wave

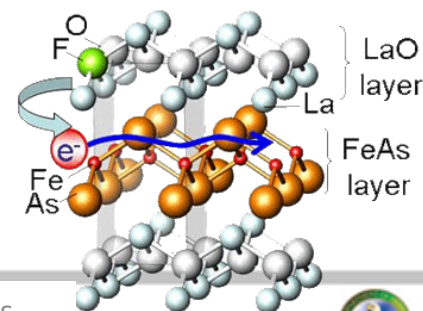
A new family of superconductors: Oxypnictides ReOMPn

–M = Fe, Co, Ni/Pn = As or P/ Re = La, Nd, Sm, Pr

Layered as HTS –superconducting AsFe layers and T_c from <10K to 55K

$\Delta^{\text{oxy}} = 5\text{-}10 \text{ meV} > \Delta^{\text{Nb}_3\text{Sn}} = 3 \text{ meV}$

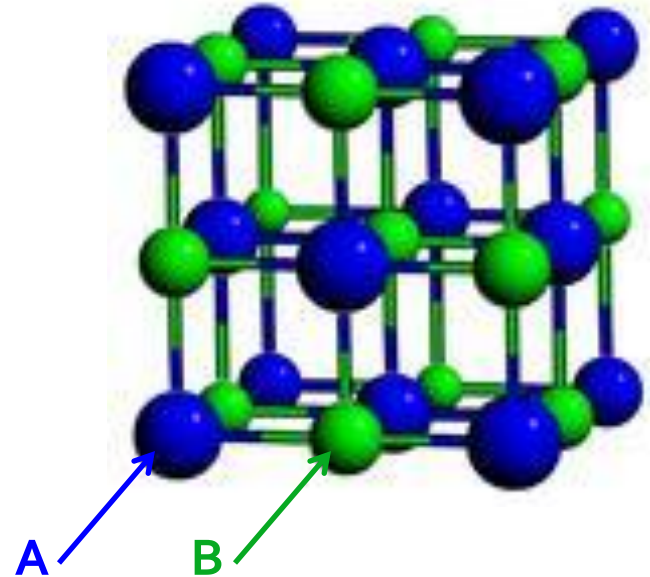
High $\rho_n \sim 1\text{m}\Omega\text{cm} \sim 10\rho_n^{\text{MgB}_2}$, big $\lambda = 180\text{-}250$



Nb Compounds

B₁ compounds – NaCl structure

Metallic atoms A form an fcc lattice
and
non-metallic atoms B occupy
all the octahedral interstices.



Nb Compounds

Only few Nitrides and Carbides of the IV, V and VI group Transition Metals have critical temperatures higher than Niobium.

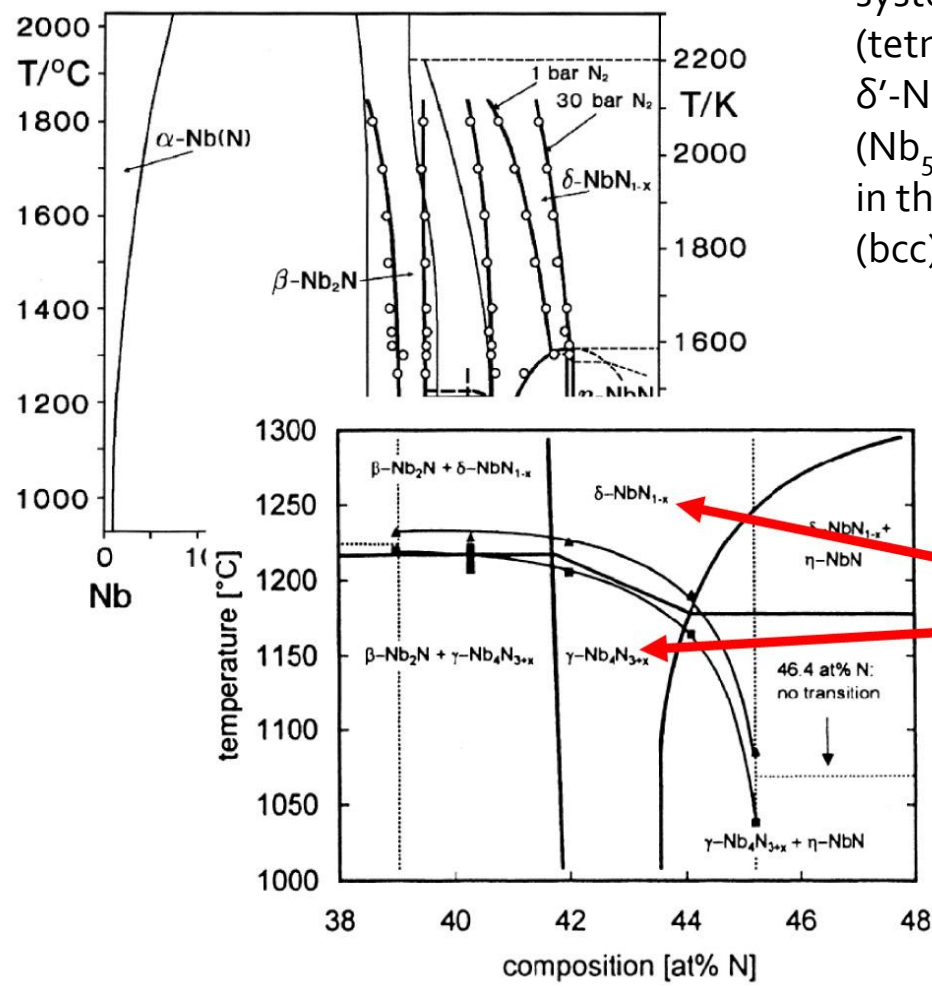
B \ A	Sc	Y	La	Ti	Zr	Hf	V	Nb	Ta	Cr	Mo	W	Re
B					3.4	3.1							
C	<1.38	<1.38		3.42	<0.3	<1.20	0.03 3.2*	12	10.35		14.3	10.0	3.4
N	<1.38	<1.4	1.35	5.49	10.7	8.83	8.5	17.3	6.5	<1.28	5.0	<1.38	
P			<1.68										
Sb		<1.02	<1.02										
O				2.0			<0.3	1.39					
S	<0.33	1.9	0.87		3.3								
Se	<0.33	2.5	1.02										
Te		2.05	1.48										

* $T_c = 3.2$ K was registered in vanadium carbide after implantation of C^+ ions

Superconductivity of Transition Metals, their Alloys and Compounds,
S.V Vonsovsky, Y.A. Izyumov, E.Z. Kurmaev, Springer-Verlag, 1982

NbN phase diagram

Different nitrides are present in the Nb–N system: β -Nb₂N (hexagonal), γ -Nb₄N_{3±x} (tetragonal), δ -NbN_{1-x} (fcc), η -NbN (hexagonal), δ' -NbN (hexagonal) and other N-rich phases (Nb₅N₆, Nb₄N₅). In addition, nitrogen dissolves in the metal forming α -Nb(N) solid solution (bcc).



δ -NbN \rightarrow $T_c \sim 15 - 17.3$ K
 γ -NbN \rightarrow $T_c \sim 12 - 15$ K

RF cavity studies mostly focus on δ phase

Nb Compounds - NbN

The only B₁ simple compound that has widely tested for accelerating cavities

Mainly two different techniques have been investigated for this application:

- Thermal diffusion of N into Nb followed by rapid quench cooling
- Reactive Sputtering on metallic or ceramic substrates to Nb cavities

Thermal Diffusion:

Bulk Nb (RRR 300) annealed @ 1550°C for 2h
+
reacted in N₂ vapor(150mbar) @ 1400°C for 4h
Rs=1.3 10⁻⁶ @ 4.2K and 4 10⁻⁹ @1.8K @7.9GHz

G.Gemme et al., J.Appl.Phys. 77(1), Jan. 1995

Reactive Sputtering:

Sputtering from high purity Nb target in Ar+ N₂ in DC triode magnetron sputtering system
Highest T_c for substrate temp. > 500°C, P_{Ar}=8.10⁻³mbar, P_{N₂}=1.10⁻³mbar

A. Nigro et al., Physica Scripta Vol. 38, 483-485, 1988

Nb Compounds - NbN

Good SC properties, even if deposited at low temperature
Low secondary emission coefficient
Very stable surface properties

The right B1-NbN superconducting phase is the so-called δ -phase

$T_c = 17.2$ K for δ -phase (lattice parameter = 4.388 Å)

T_c very sensitive to Nitrogen stoichiometry

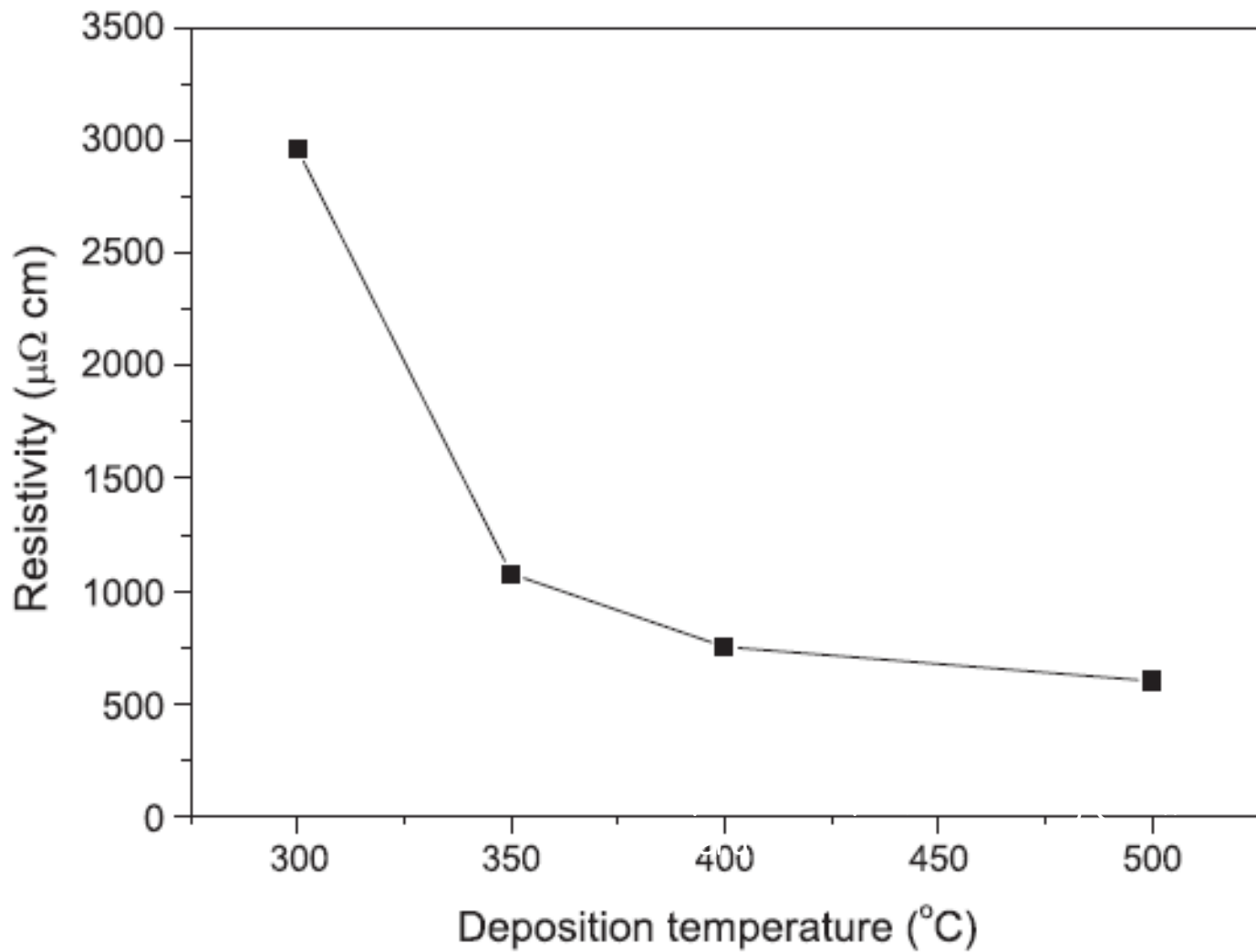
. In sputtered films, the δ -phase can be found mixed to some other low T_c phases

Even if no grain boundaries are present and δ -phase single crystal is considered
the single grain resistivity is not so low.

Anomalously high resistivity of NbN in the normal state, often higher than $100 \mu\Omega\text{cm}$ due to both
metallic and gaseous vacancies randomly distributed in both sublattices

Equiatomic composition is $\text{Nb}_{0.987}\text{N}_{0.987}$ not $\text{Nb}_{1.0}\text{N}_{1.0}$

Common problem for B1 compounds

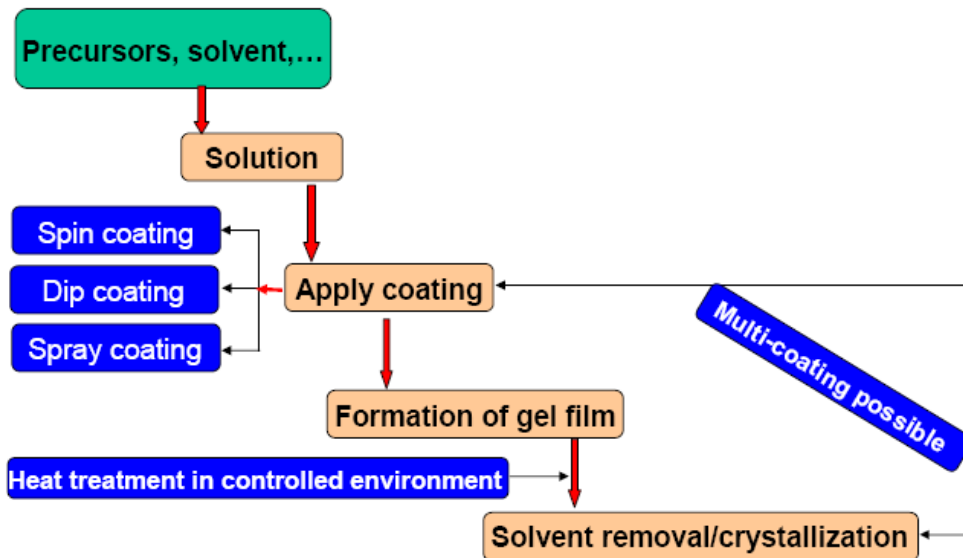


P. Alen, M. Ritala, K. Arstila, J. Keinonen, and M. Leskelae, *Thin Solid Films* 491:235 (2005).

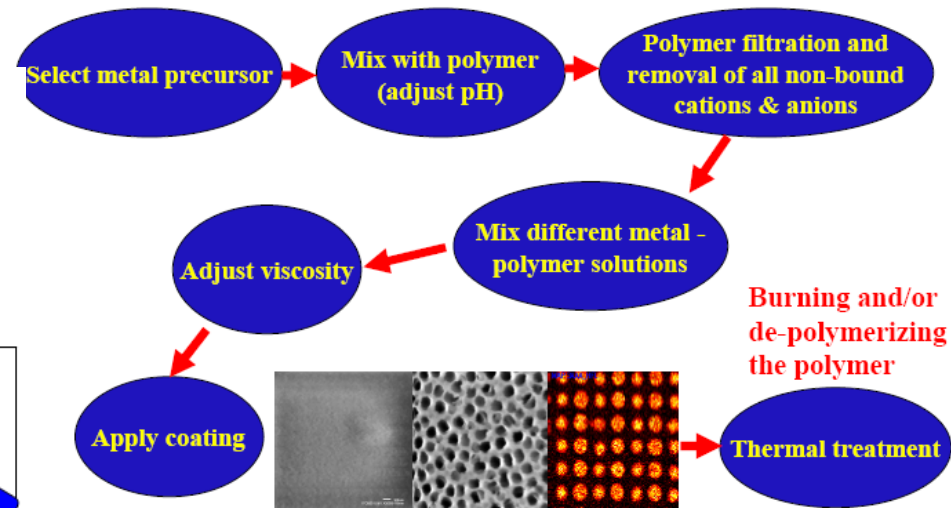
Polymer Assisted Deposition (PAD) at LANL (Q. Jia et al.)

- A chemical solution technique to deposit films of nearly any metal-oxide using aqueous solution by mixing metal precursors with water-soluble polymers
- Polymer plays a critical role in metal-oxide films

Typical chemical solution deposition process flowchart



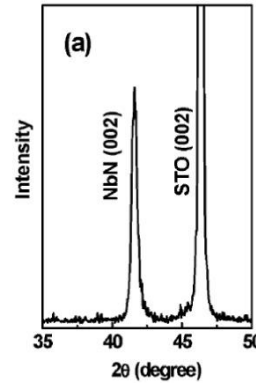
• Process



NbN grown by Polymer Assisted Deposition

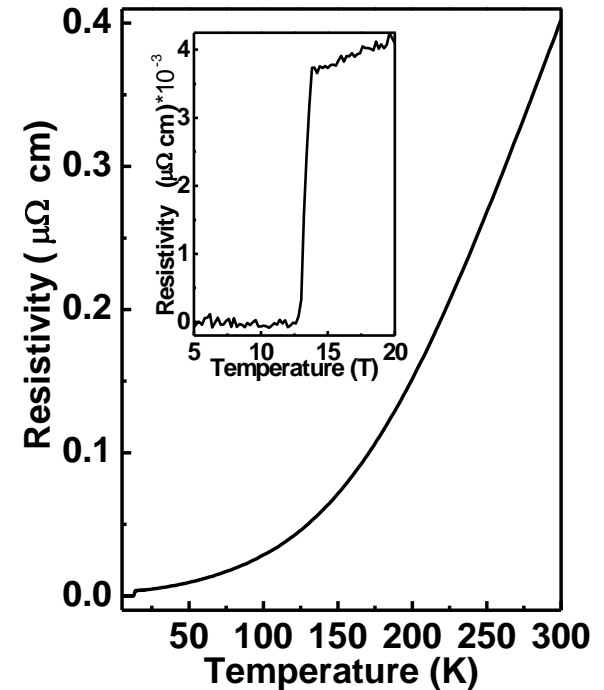
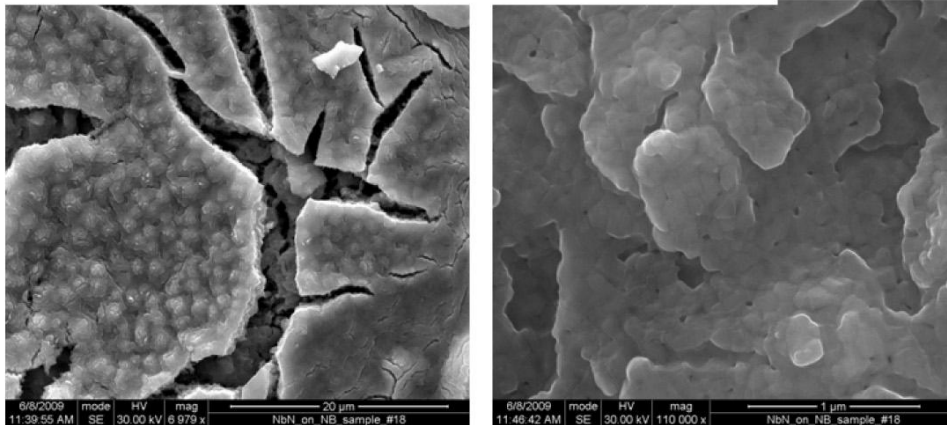
High-quality epitaxial films grown by PAD: an aqueous solution of niobium ion bound to polymer spin coated on substrate, and then annealed at 900 °C for 5 hours in gaseous ammonium.

LANL: First attempts to deposit on polycrystalline niobium – unsuccessful, working with single-crystal Nb now



Unusually high RRR = 98.4
and low $\rho_{20} \approx 0.4 \mu\Omega \text{ cm}$

$T_c \approx 14 \text{ K}$ (γ - NbN)



Nb Compounds - NbTiN

Ternary Nitride $\text{Nb}_{1-x}\text{Ti}_x\text{N}$

Presence of Ti found to reduce significantly the resistivity
And facilitate formation of a pure cubic structure.
The d-phase remains thermodynamically stable even at RT.
 T_c as high as for good quality NbN, for Nb fraction $(1-x) > 0.5$

extreme hardness, excellent adherence on various substrates, very good
corrosion and erosion resistance, high-sublimation temperature, and
relative inertness

**More metallic nature and better surface properties than NbN
should result in better RF performance**

Nb Compounds - NbTiN

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

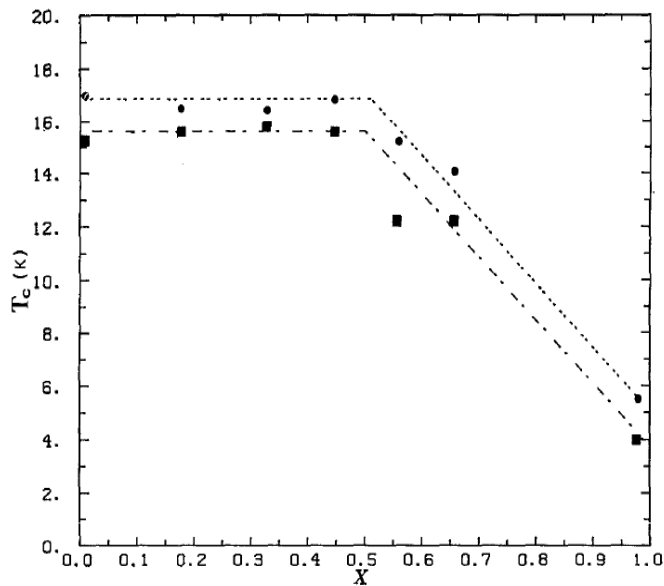


Fig. 1. Superconducting critical temperature T_c as a function of the titanium composition (x) for the (Nb_{1-x}Ti_x)N films deposited at $T_s = 600^\circ\text{C}$ (circles) and at $T_s = 200^\circ\text{C}$ (squares).

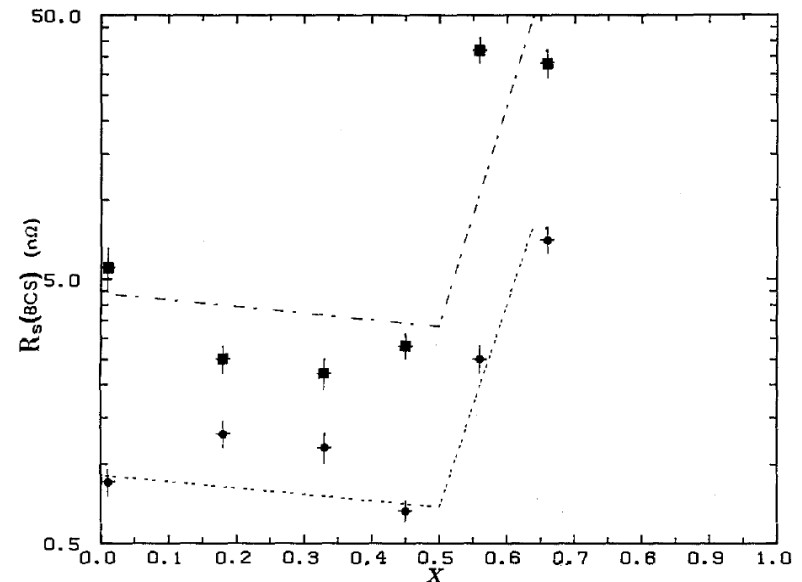


Fig. 3. Calculated BCS surface impedance $R_s(\text{BCS})$ as a function of the titanium composition (x) for the (Nb_{1-x}Ti_x)N films deposited at $T_s = 600^\circ\text{C}$ (circles) and at $T_s = 200^\circ\text{C}$ (squares). The continuous lines correspond to the values of the lines through the data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

Nb Compounds - NbTiN

Reactive Magnetron Sputtering:

CEA Saclay :

NbTiN films deposited on 12 cm copper disks by magnetron sputtering and tested in a cylindrical TE_{011} cavity

reached RF field levels of 35 mT

low residual surface resistance ($< 100 \text{ n}\Omega$ 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.

R_s slope significantly decreased when coating with bias ranging from -50V to -100V

P. Bosland et al.

S. Cantacuzène et al.

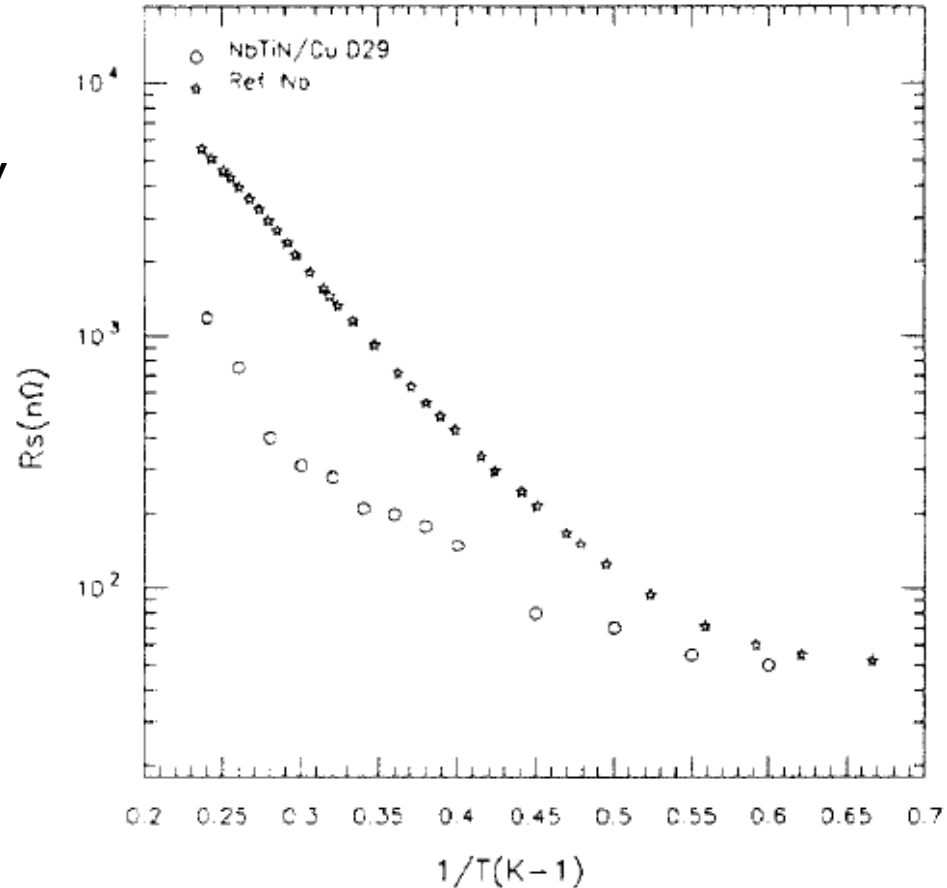


Figure 3 Surface resistance vs temperature for a NbTiN sample, at 4 GHz.

Nb Compounds - NbTiN

CERN:

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

Best cavity result for thicker film (4.3 μ m) and lower deposition temperature (265 $^{\circ}$ C)

$R_s = 330\text{n}\Omega @ 4.2\text{K}$

Q_0 at zero field is higher than the Q-value of Niobium cavities but E_{acc} limited under 10 MV/m

As for NbN, N stoichiometry critical to obtain the right SC phase

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

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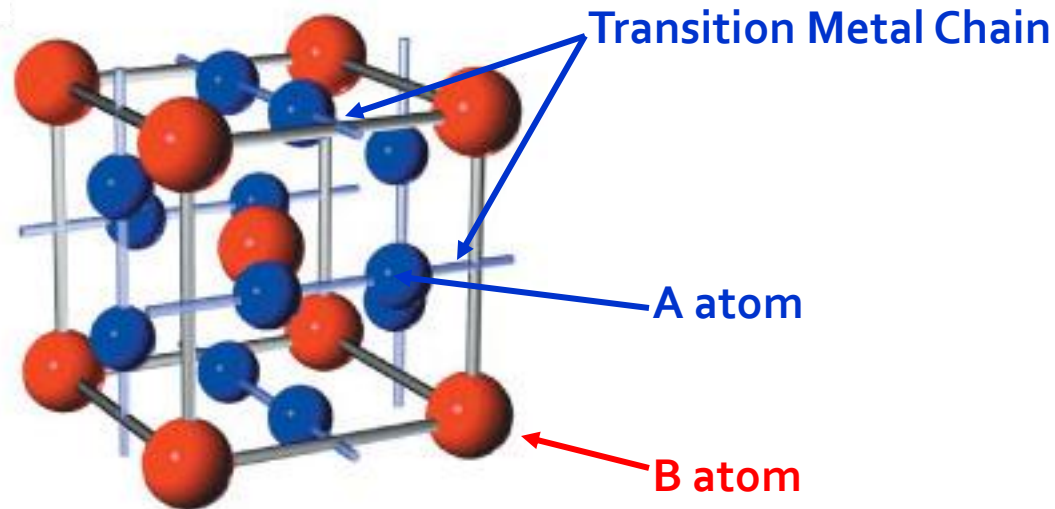
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A₁₅ Compounds - Structure

A atoms = Transition elements of group IV, V or VI

B atoms = Non transition or transition elements



B atoms occupy corners and centre of BCC structure

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

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

A15 Compounds – Potential candidates for RF Cavities



- Among the Nb and V based high T_c (15 – 20 K)
- Nb_3Ga and Nb_3Ge do not exist as stable bulk materials at 3:1 stoichiometry
- Nb_3Al exists only at high temperature causing excessive atomic disorder
- Production of above materials need non equilibrium processes
- V_3Ga , V_3Si & Nb_3Sn are stable bulk material and have high T_c
- Another A-15 compound holding promise is Mo_3Re ($T_c=15\text{K}$)

Sharma, International Workshop on Thin Films and New Ideas for Pushing The Limits Of RF Superconductivity, LNL-INFN, Oct. 2006

A15 Compounds – Preparation Methods I

Extreme brittleness so A-15 bulk structure cannot be formed

The A-15 should be produced as thin layer on the interior of the already formed structure

Such a layer need to be only 1 or 2 microns thick

$$\lambda_L(\text{Nb}_3\text{Sn}) = 65 \text{ nm}$$

Thin film route ideal

A15 Compounds – Preparation Methods II

Co-Sputtering

- Considerable success achieved in synthesizing difficult materials like Nb_3Ge with highest T_c (~23k) or V_3Si
- Typically two constituents are sputtered simultaneously onto a temperature controlled substrate
- Stoichiometry dependent on relative positions of target and substrate (can be manipulated to get perfect stoichiometry)
- Stoichiometry control difficult over large areas like accelerating system and if stoichiometry range for A-15 phase is narrow.

Sputtering

To sputter from a single target of correct stoichiometry (prepared by powder sintering)

Stoichiometry, Substrate Temperature, Deposition Rate, Deposition Thickness
Can be varied independently

A15 Compounds – Preparation Methods III

Chemical Vapor Deposition (CVD)

MOCVD (*Metal Organic Chemical Vapour Deposition*) is a particular case of CVD in which the precursor is a metallorganic compound

Process in which one or more precursors, present in vapor phase, chemically react on an appropriate warm substrate, giving rise to a solid film

Deposition rate and structure of the film depend upon temperature and reagent concentration

⇒ Uniformity of temperature and flow of gaseous over entire cavity surface may be difficult with complex geometry

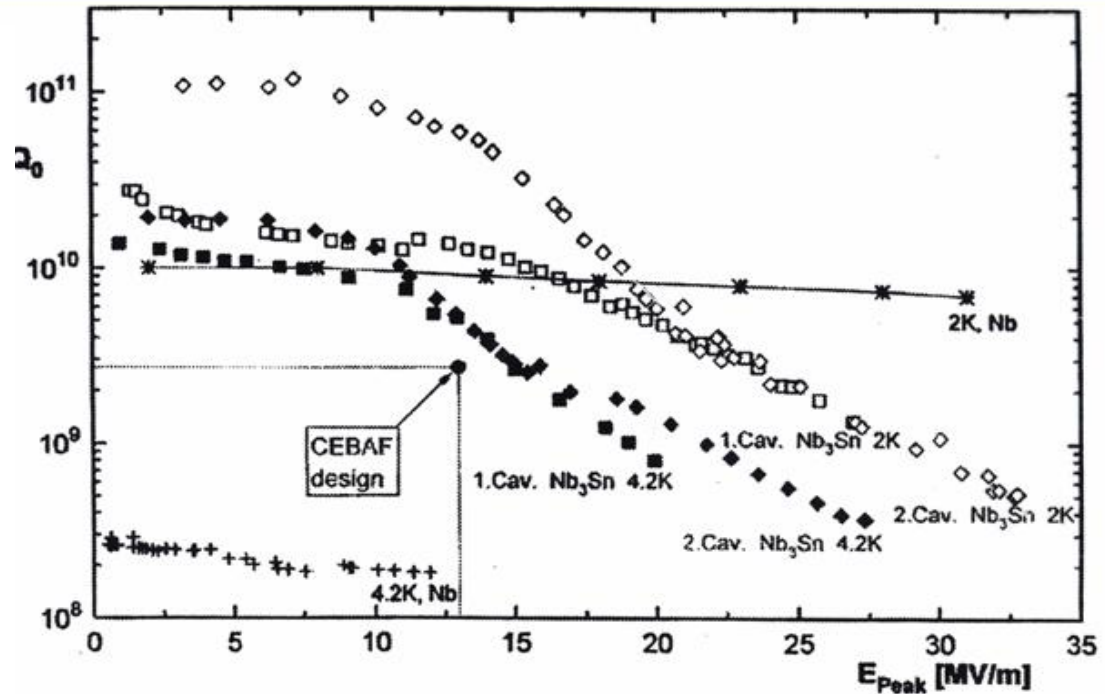
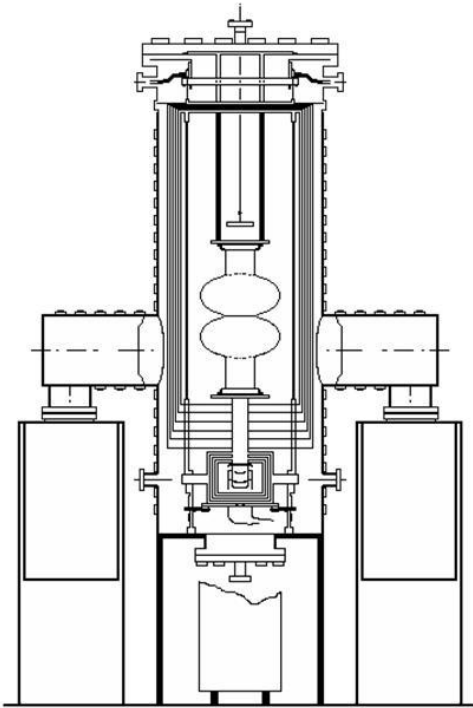
Diffusion Reaction

Technique proved successful for magnet conductor application
Simple equipment compared to sputtering and CVD

A15 Compounds – Nb₃Sn

Wuppertal, end '80s :

Nb₃Sn cavity (1.5 GHz) obtained through Sn vapour phase diffusion @ 1200°C



Q vs. E_{peak} of the 1st two Nb₃Sn-coated 1.5GHz single cell cavities in comparison to pure Nb at 4.2K and 2K from CEBAF

5-cell 1.5GHz cavity also coated: $Q_0 \sim 10^9$, $E_{\text{acc}} = 7 \text{ MV/m}$ with $Q = 8 \cdot 10^8$

G. Müller et al.,

M. Peiniger & H. Piel, *IEEE Trans. On Nucl. Sc.* Vol NS-32, n°5, Oct. 1985

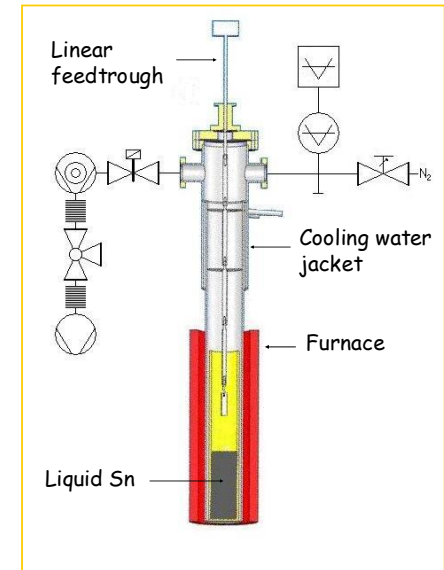
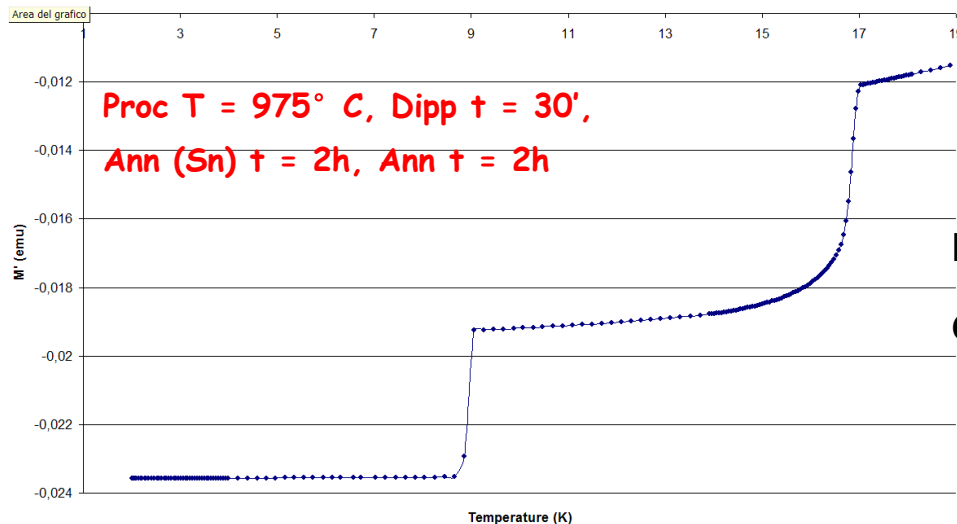
A15 Compounds – Nb₃Sn through liquid diffusion

S. Deambrosis, Sharma, *International Workshop on Thin Films and New Ideas for Pushing The Limits Of RF Superconductivity, LNL-INFN, Oct. 2006*
S. Deambrosis et al., *Physica C 441 (2006) 108-113*

Nb₃Sn coatings on Nb by liquid diffusion method at INFN Legnaro "Hybrid" Process"

- Substrate thermalization (30 min - 1 h)
- Dipping (few min - 2 h)
- Sample annealing with Sn vapor for a few hours
- Sample annealing without Sn vapor for a few hours

Nb₃Sn 42 1: 975°C x 30' + 2h; 975°C x 2h



No Residual Sn traces on the sample surface

Good Nb₃Sn film superconductive properties

T_c = 16.8 K, DT_c = 0.16 K

No Sn rich Phases

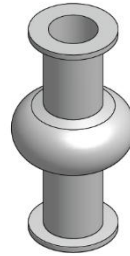
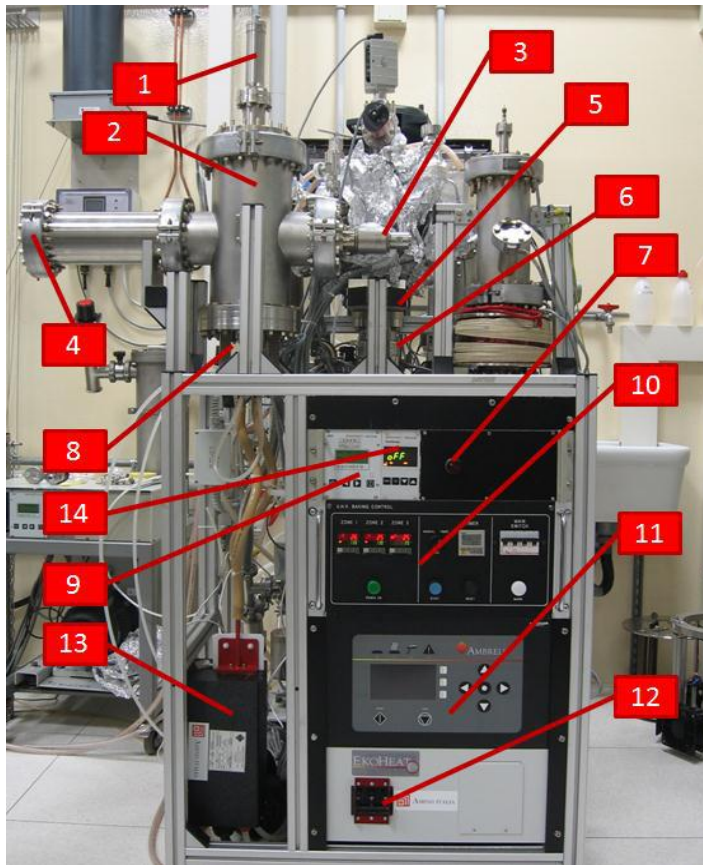
Diffusion temperature to be kept above 930°C to avoid formation of low T_c phases like Nb₆Sn₅ (2.6 K) and NbSn₂ (2.1 K)

Diffusion time optimize to obtain desired Nb₃Sn thickness

Post diffusion heat reaction important to get rid of the outer Sn layer

Post diffusion annealing to have enlarged grains and perfect ordering

A15 Compounds – Nb₃Sn through liquid diffusion



High temperature annealing for thermally diffused Nb₃Sn,
Atroshchenko Konstantin
 Master Thesis, INFN LNL

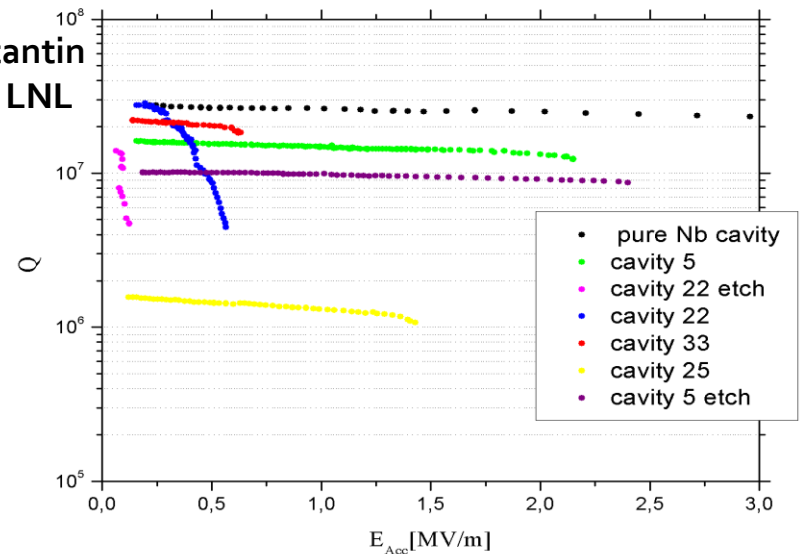
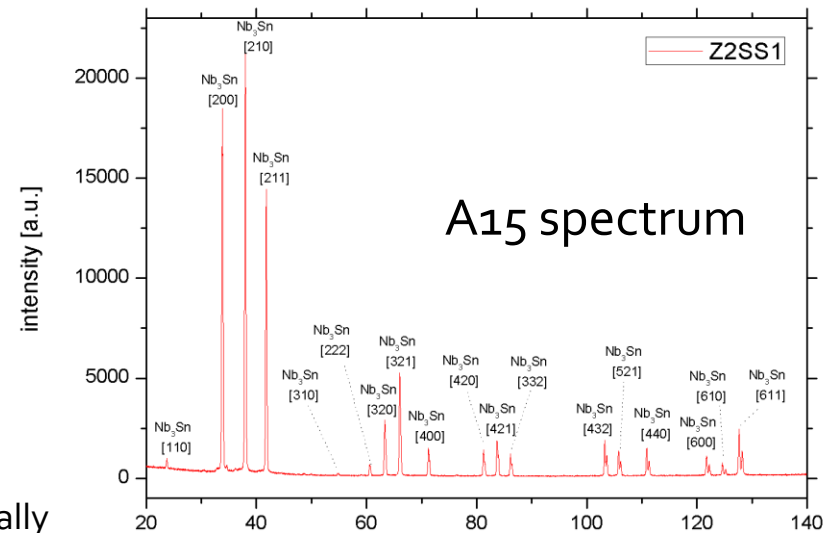
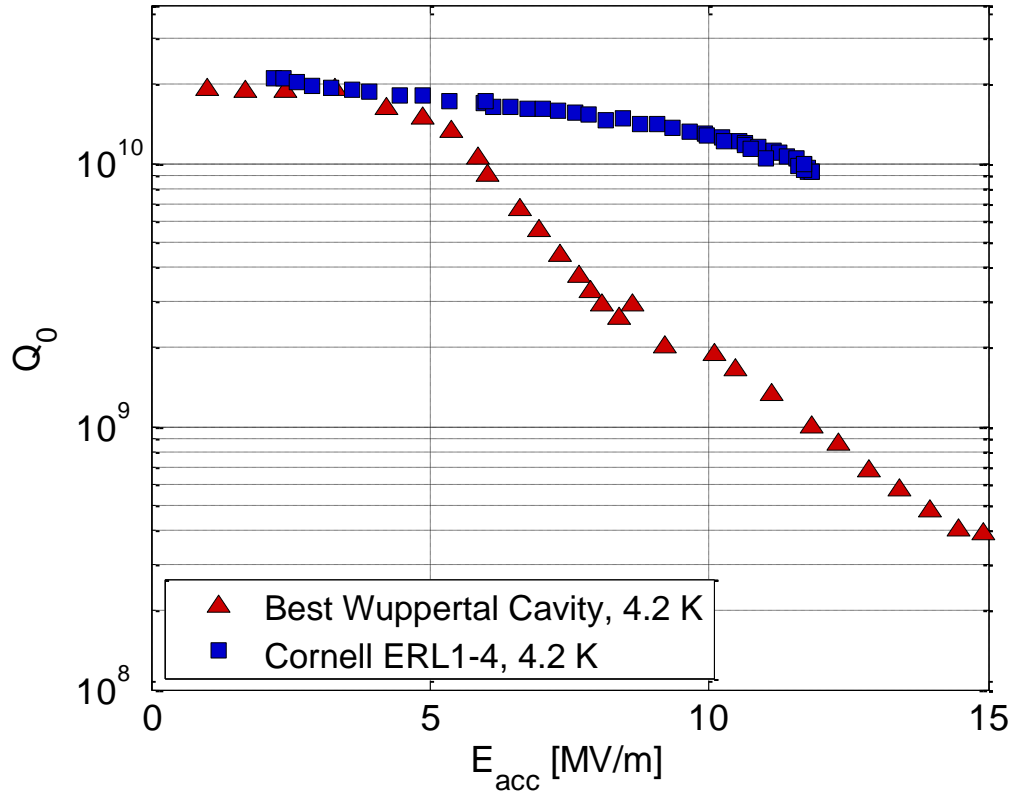


Figure 7.3. Inductive annealing stand
 1-manipulator; 2-vacuum chamber; 3-whole metal valve; 4-window flange; 5-pneumatic gauge; 6-turbomolecular pump; 7-gauge switcher; 8-inlet of work coil; 9-pump controller; 10-baking control unit; 11-induction heater controlling and inverter block; 12-inductor main

Nb₃Sn @ Cornell University – Vapor diffusion

Before
Coating

- New cavity fabricated, coated with Nb₃Sn, and tested at Cornell U. by Sam Posen and Matthias Liepe
- First ever accelerator cavity made with an alternative superconductor to far outperform Nb at useable gradients



After
Coating



Nb₃Sn @ Jefferson Lab– Vapor diffusion

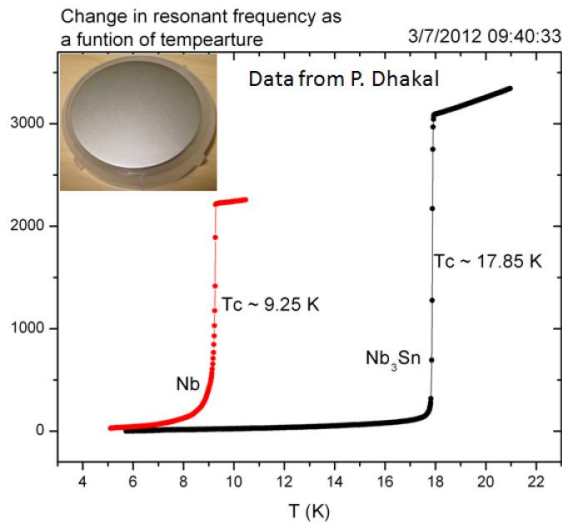
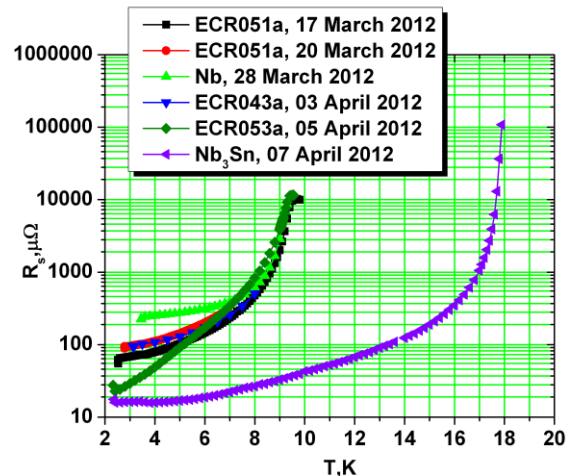
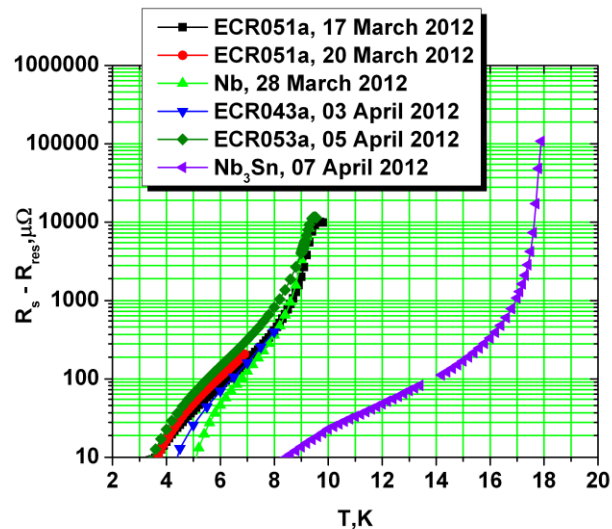


Figure 4: Test results on the coaxial sample. The data shows that the transition temperature of the coaxial sample was 17.95 K. In the top left corner the surface of the best sample is shown.

Same results with the residual resistance subtracted. Note resistance measurement error is estimated to be about 20%.



B. P. Xiao, G. V. Eremeev, M. J. Kelley,
H. L. Phillips, C. E. Reece, LINAC'12



Measurement of coated Nb₃Sn sample as well as some Nb and thin film niobium samples

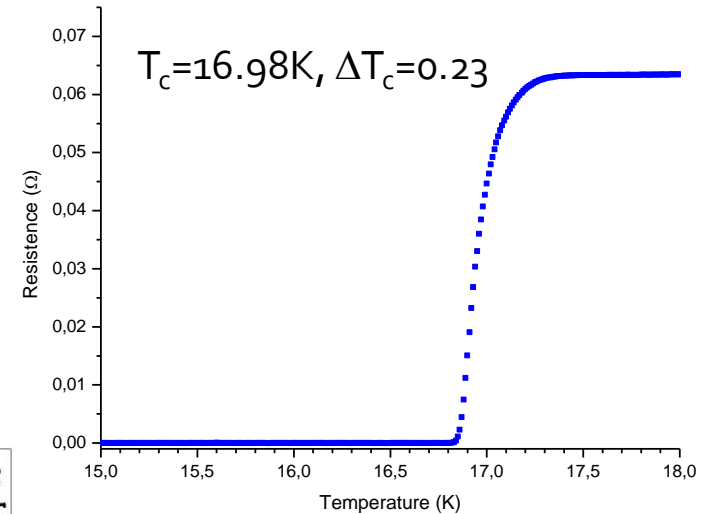
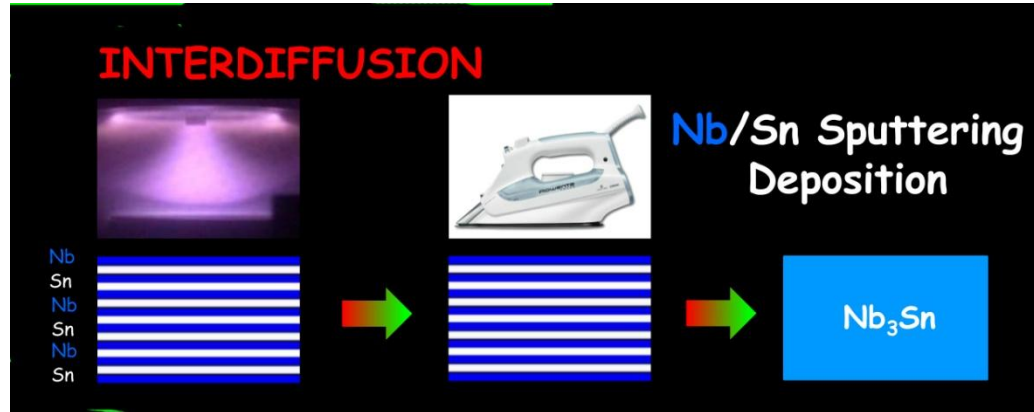


A15 Compounds - Nb₃Sn through Multilayer Coating

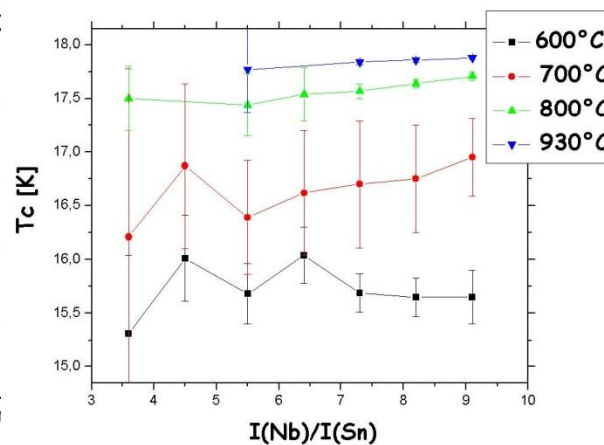
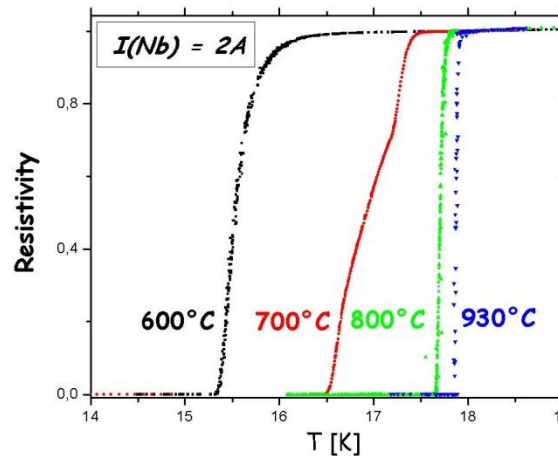
Deambrosis, Keppel, LNL/INFN, International Workshop on Thin Films and New Ideas for Pushing The Limits Of RF Superconductivity, LNL-INFN, Oct. 2006 & SRF 2009

Coat alternate layers of Nb and Sn and subject to diffusion reaction

Preliminary results: a sharp transition and a T_c of 17 K has been obtained



Thickness Nb = 4.5 x Thickness Sn
Annealed after sputtering for 3 hours at 975 °C

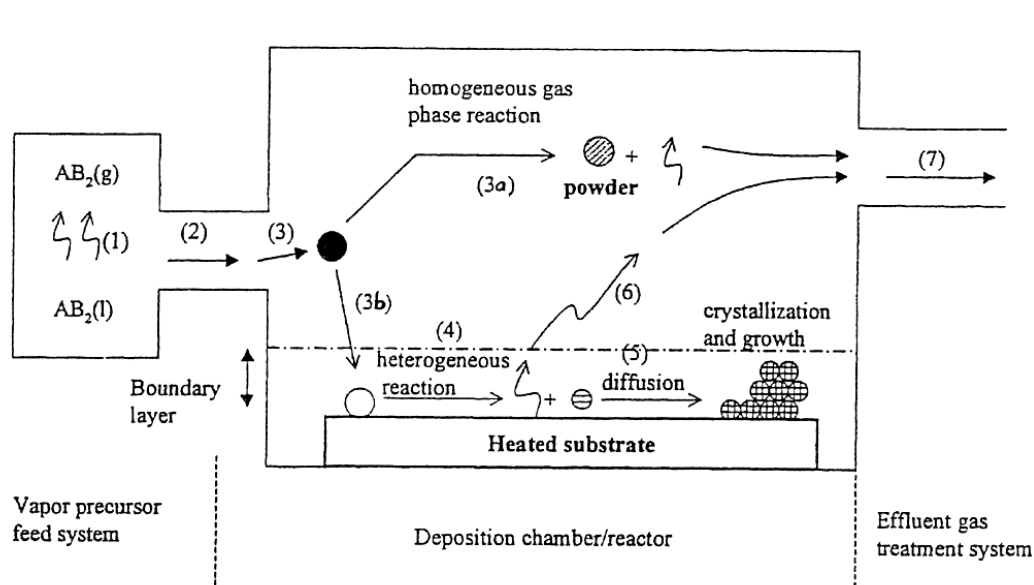


Sputtering Target	Voltage (V)	Current (A)	Power (W)
Sn	613	0.18	108
Nb	407	1.99	800

A15 Compounds - Nb₃Sn through MOCVD process

G. Carta et al. International Workshop on Thin Films and New Ideas for Pushing The Limits Of RF Superconductivity, LNL-INFN , Oct.2006

MOCVD technique using bis(cyclopentadienil)niobium borohydride, (cyclopentadienil)niobium tetramethyl and tributyltin hydride as Nb and Sn precursors respectively.



Tin from Bu ₃ SnH		Niobium from CpNbMe ₄	
Bath temperature	60°C	Bath temperature	80°C
Line temperature	100°C	Line temperature	100°C
Carrier gas flux	N ₂ /H ₂ (25%) 10 scc/min	Carrier gas flux	N ₂ /H ₂ (25%) 100scc/min
Deposition temperature		550°C	
Pressure		3,7 mbar	
Co-reactant gas flux		N ₂ /H ₂ (25%) : 350 scc/min	
Deposition time		45 min	

Sample characterization by XRD, SEM and RBS analyses: presence of niobium (I, II, V) and tin (II) oxides on the surface.

Problems:
great oxophilic character of Nb

A15 Compounds – V₃Si

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

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

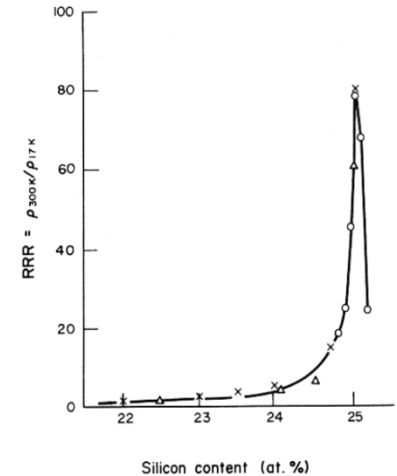
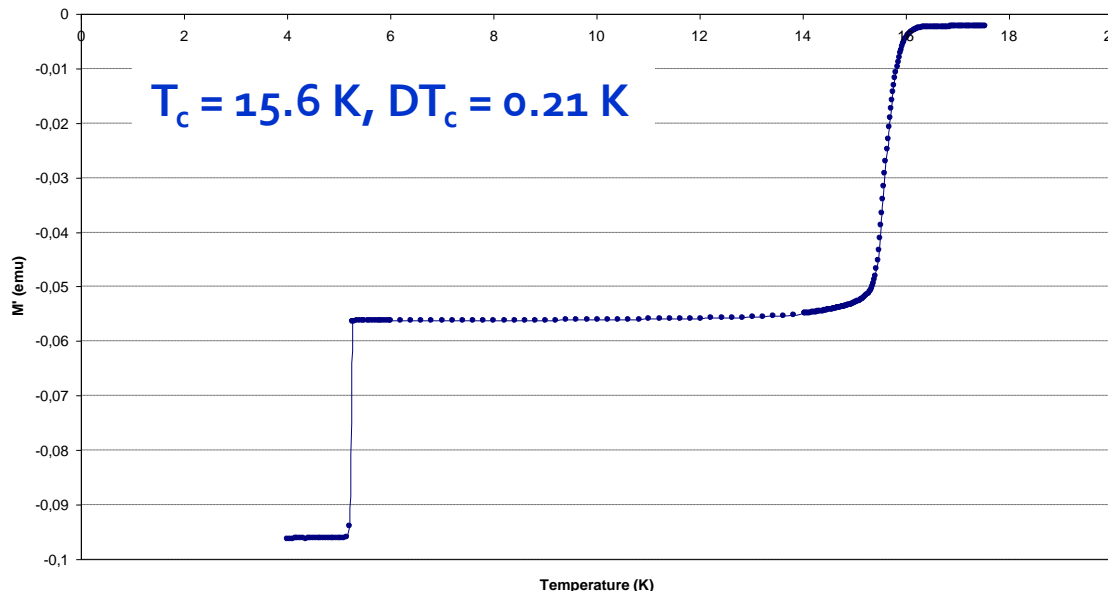
V₃Si layers by silanization of V substrate and Thermal Diffusion

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

825°C, 4h+8h



* 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

A15 Compounds – Mo₃Re

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. Deambrosis 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

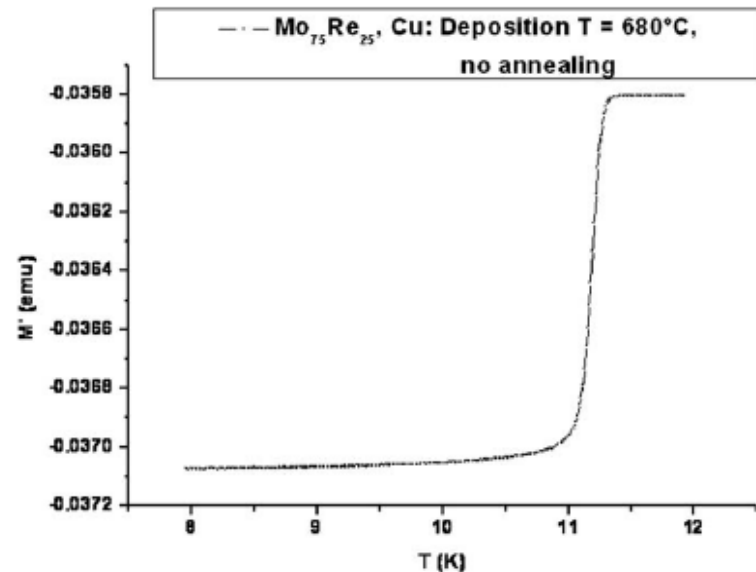


Fig. 4. A Mo₇₅Re₂₅ film deposited on Cu transition curve: deposition $T = 680^\circ\text{C}$, $T_c = 11.18$, $\Delta T_c = 0.08$ K.

Magnesium Diboride (MgB_2)

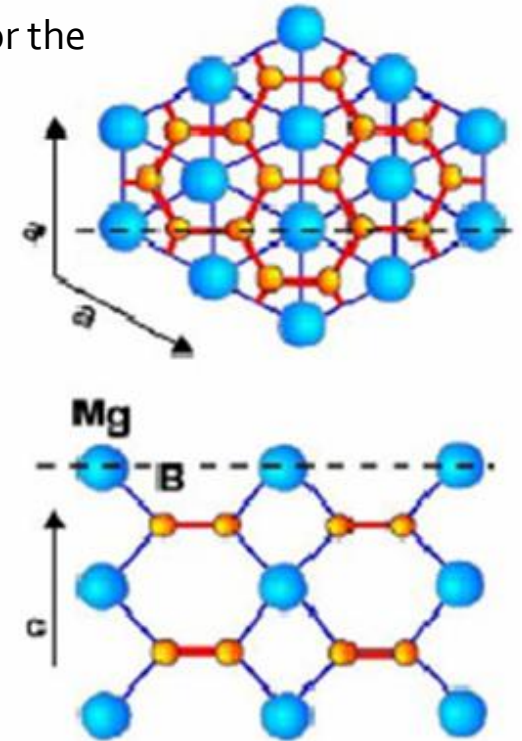
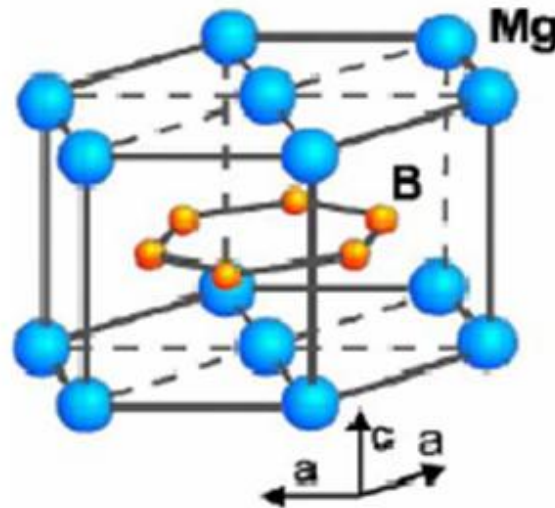
Graphite-type boron layers separated by hexagonal close-packed layers of magnesium

Superconductivity comes from the phonon-mediated Cooper pair production similar to the low-temperature superconductors except for the **two-gap nature**.

$T_c \sim 40 \text{ K}$

Compared to cuprates:

- Cheaper
- Lower anisotropy
- Larger coherence length
- Transparency of grain boundaries to current flows

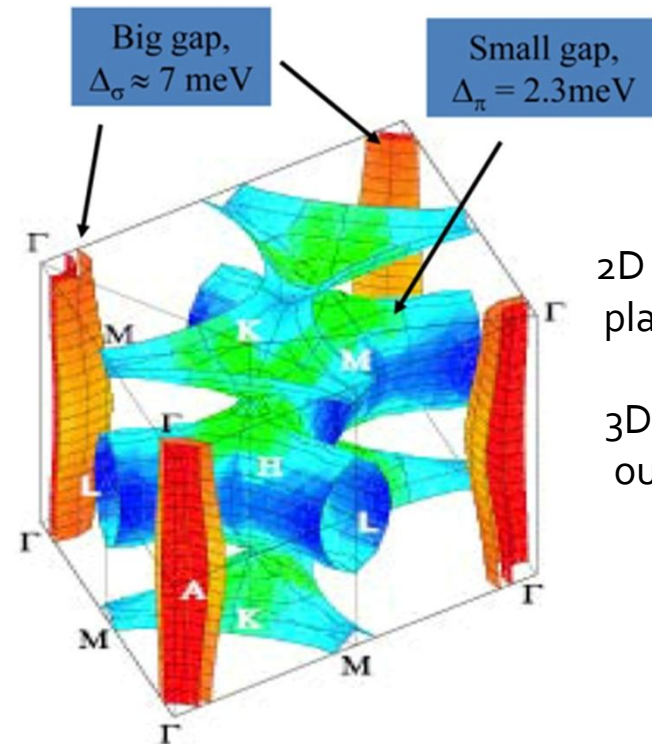
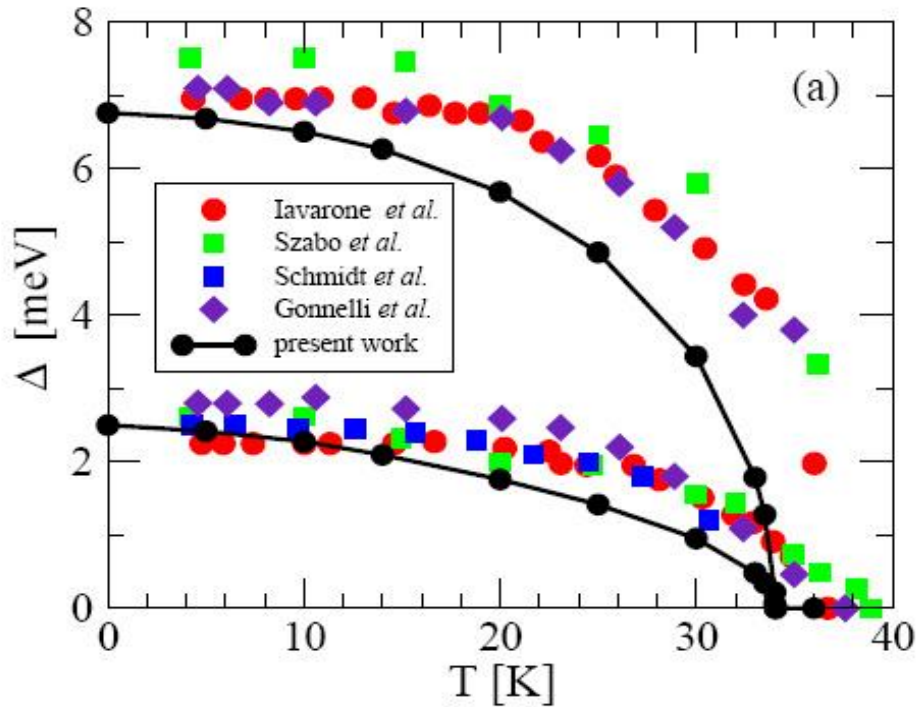


attractive for RF applications.

C. Buzea and T. Yamashita, Superconductor Sci. Technol. 14 (2001) R115.

MgB₂: Two Energy Gaps

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



2D big gap for in-plane σ -orbitals & 3D small gap for out-of-plane π -orbitals

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

R_s is dominated by the smaller gap, so MgB₂ may not be better than Nb₃Sn because $\Delta_{\pi}^{\text{MgB}_2} = 2.3 \text{ meV} < \Delta^{\text{Nb}_3\text{Sn}} = 3.1 \text{ meV}$, but better than Nb ($\Delta^{\text{Nb}} = 1.5 \text{ meV}$)

RF response has shown lower energy gap behavior. This must be compared to $\Delta \approx 1.5 \text{ meV}$ for Nb. There is room for better performance than Nb, since the resistivity can also be made quite low (best values are $\leq 1 \mu\Omega\text{cm}$).

MgB₂ - A comparison with conventional SC for RF applications

X. Xi, International SRF Thin Films Workshop, Padua, Italy, 2006

	MgB ₂	Nb
T _c (K)	39	9.2
ρ ₀ (mΩcm)	0.1-10	0.05
RRR	3-30	300
Δ _{p,s} (meV)	2, 7	1.2
2 Δ _{p,s} /K _B T _c (meV)	1.6, 4	3-9
x _{p,s} (nm)	50, 12	40
l (nm)	85	80
m ₀ H _{c2} (T)	6-50	0.2
R _{BCS} @ 4K, 500MHz (nΩ)	2.5/2.3x10 ⁻⁵	69

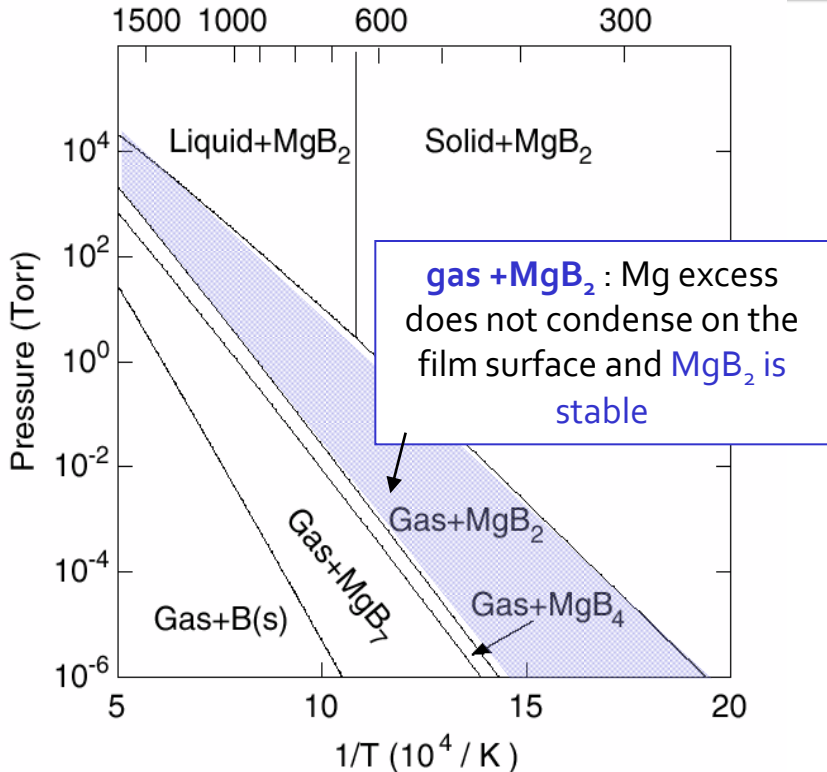


from
$$R_{BCS} (n\Omega) = \left(\frac{1}{T} \right) 10^5 \nu_{GHz}^2 e^{(-\Delta/KT_c)}$$

F. Collings et al. SUST 17 (2004)

MgB₂ -Thin films growth

Temperature (°C)



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

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

optimal T for epitaxial growth ~ T_{melt}/2

For MgB₂, 540° C → it requires P_{Mg} ~11 Torr

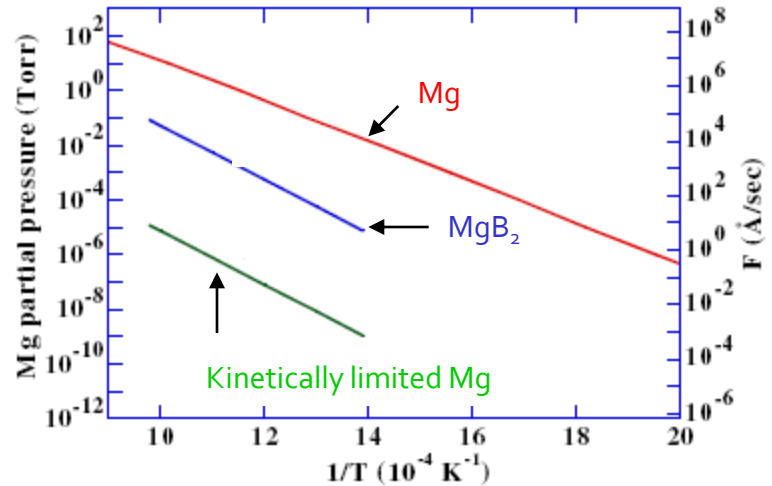
Too high for UHV deposition techniques (PLD, MBE...)

At P_{Mg} = 10⁻⁴-10⁻⁶ Torr, compatible with MBE, T_{sub} ~ 400° C

MgB₂ is stable, but no MgB₂ formation:

Mg atoms re-evaporate before reacting with B

Kinetic of Mg is also important



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

At P=10⁻⁶ Torr and T> 250°C no accumulation of Mg will take place on the substrate and the growth of the superconducting phase is very slow due to a large kinetic energy barrier.

At low Mg pressure only extremely low deposition temperatures can be used

MgB₂ – HPCVD on metal substrates

X. Xi-TempleUniversity

Hybrid Physical Chemical Vapor Deposition

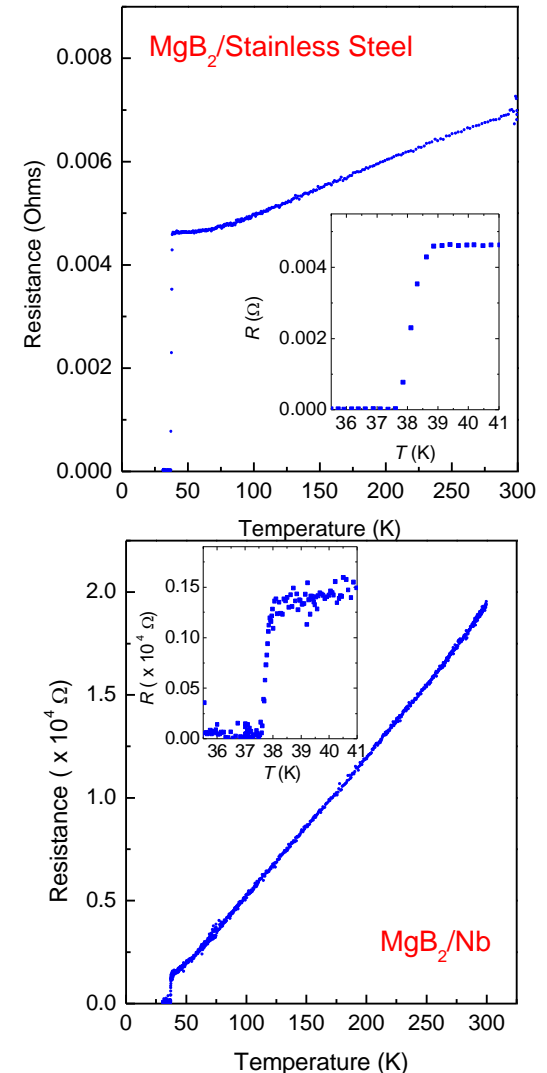
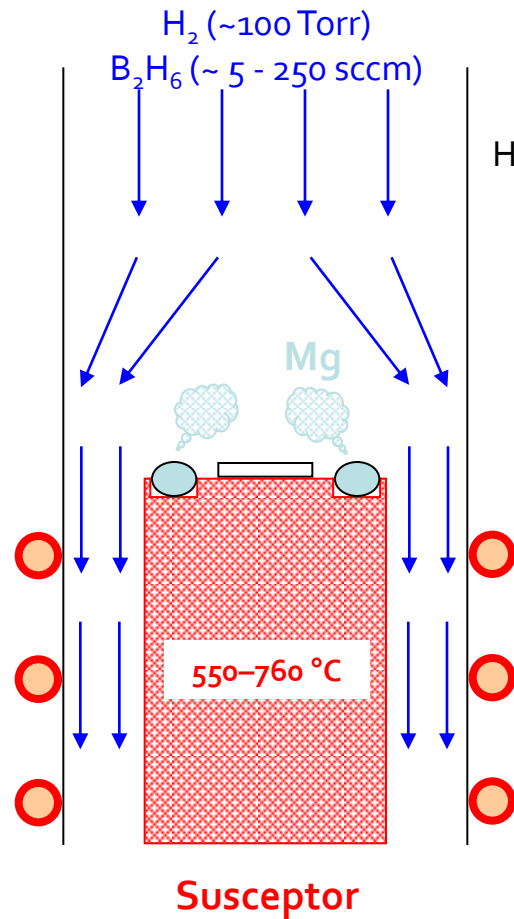
High T_c has been obtained in polycrystalline MgB₂ films on stainless steel, Nb, TiN, and other substrates.

Clean HPCVD MgB₂ thin films with excellent properties:

- RRR > 80
- low resistivity (< 0.1 μΩ) and long mean free path
- high T_c ~ 42 K (due to tensile strain), high J_c (10% depairing current)
- low surface resistance, short penetration depth
- smooth surface (RMS roughness < 10 Å with N₂ addition)
- good thermal conductivity (free from dendritic magnetic instability)

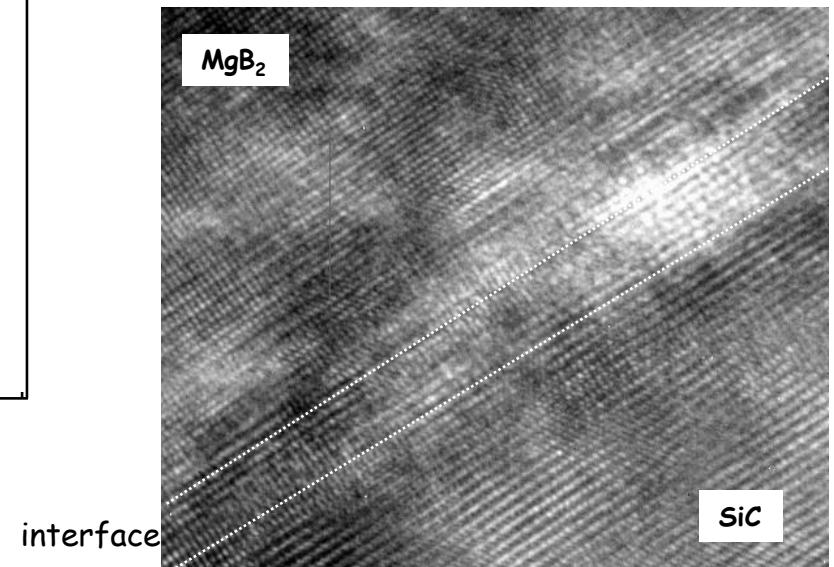
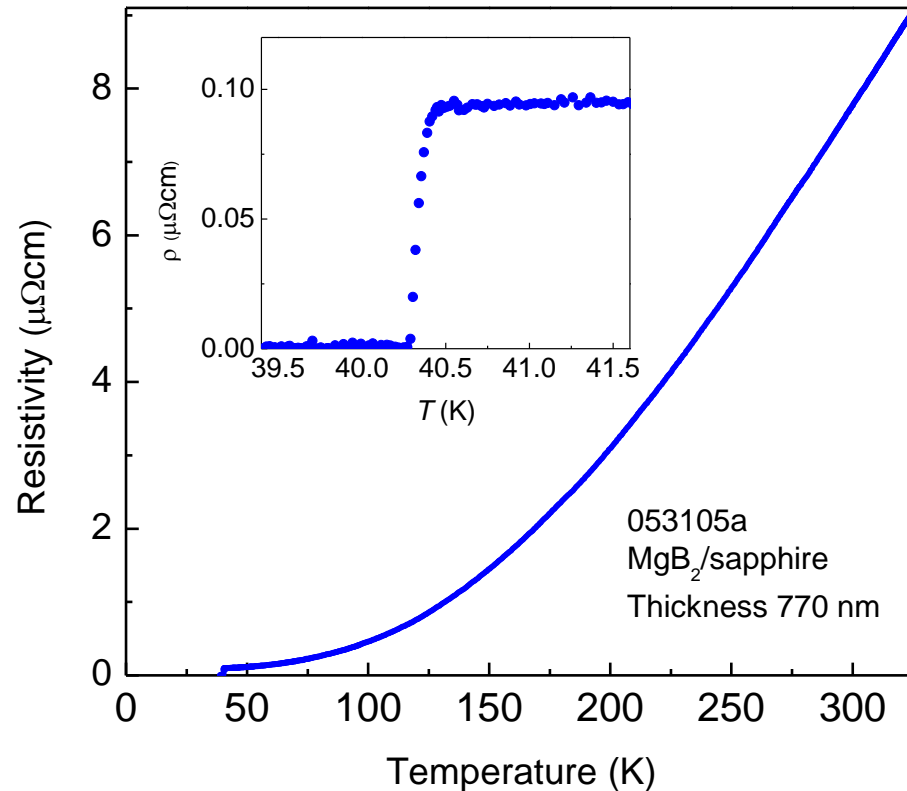
Critical engineering considerations:

- generate high Mg pressure at substrate (cold surface is Mg trap)
- deliver di-borane to the substrate (the first hot surface di-borane sees should be the substrate)



HPCVD at Temple Uni. (X. Xi et al.)

- Example of epitaxial MgB₂ Films by HPCVD: $RRR > 80$

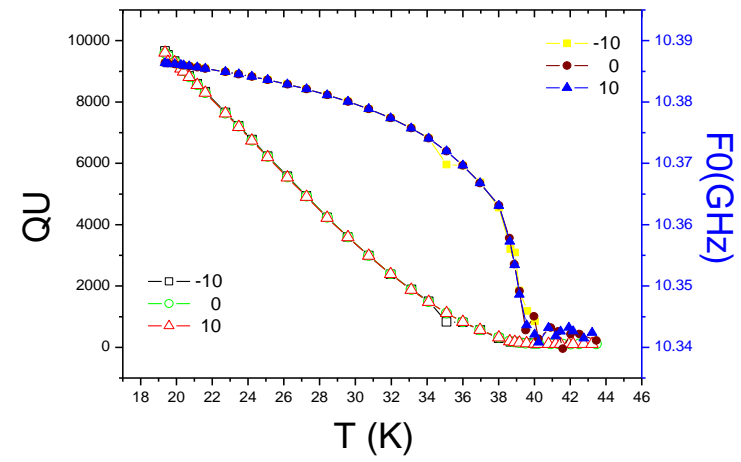
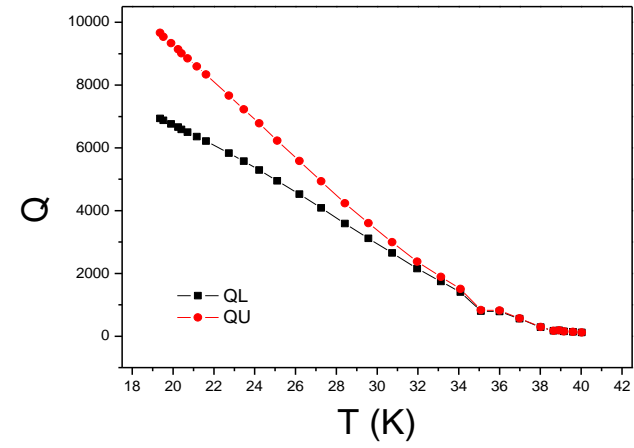
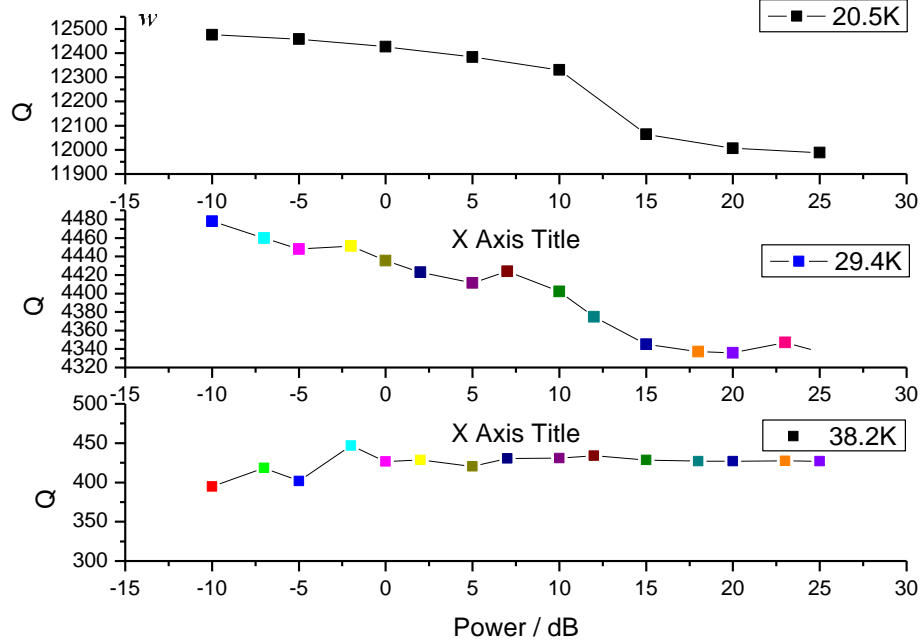
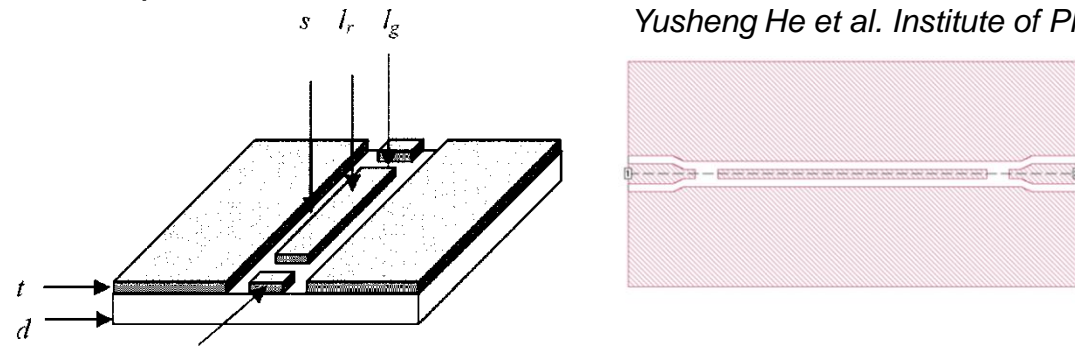


MgB₂ – Microwave Performance on HPCVD Films

X. Xi , private communication

C. Zhuang et al. Peking University

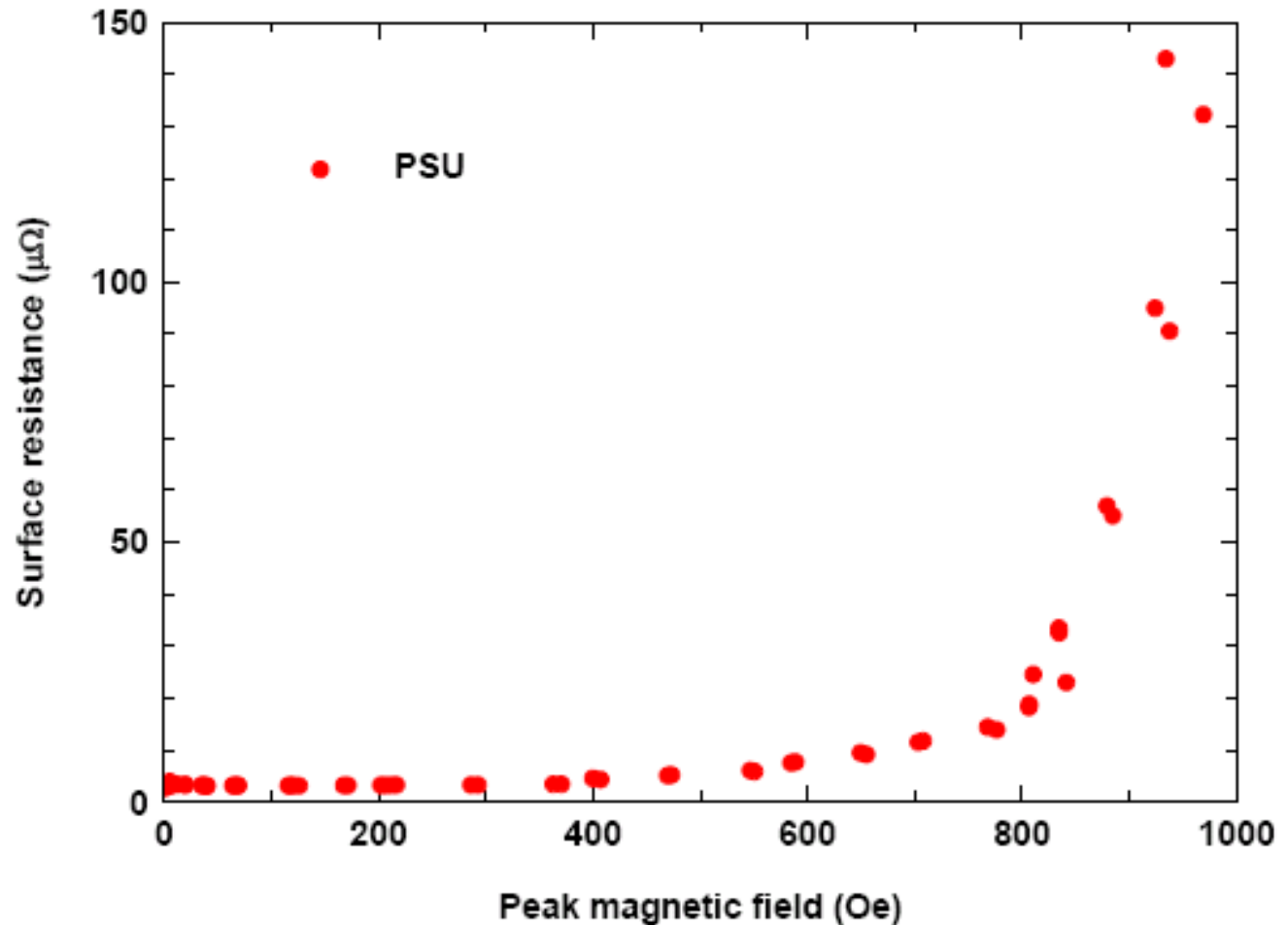
Yusheng He et al. Institute of Physics of Chinese Academy of Sciences



High Q value ~10000 at 20K, sharp transition in Fo-T curve around the critical temperature, no power dependence in QU-T and Fo-T

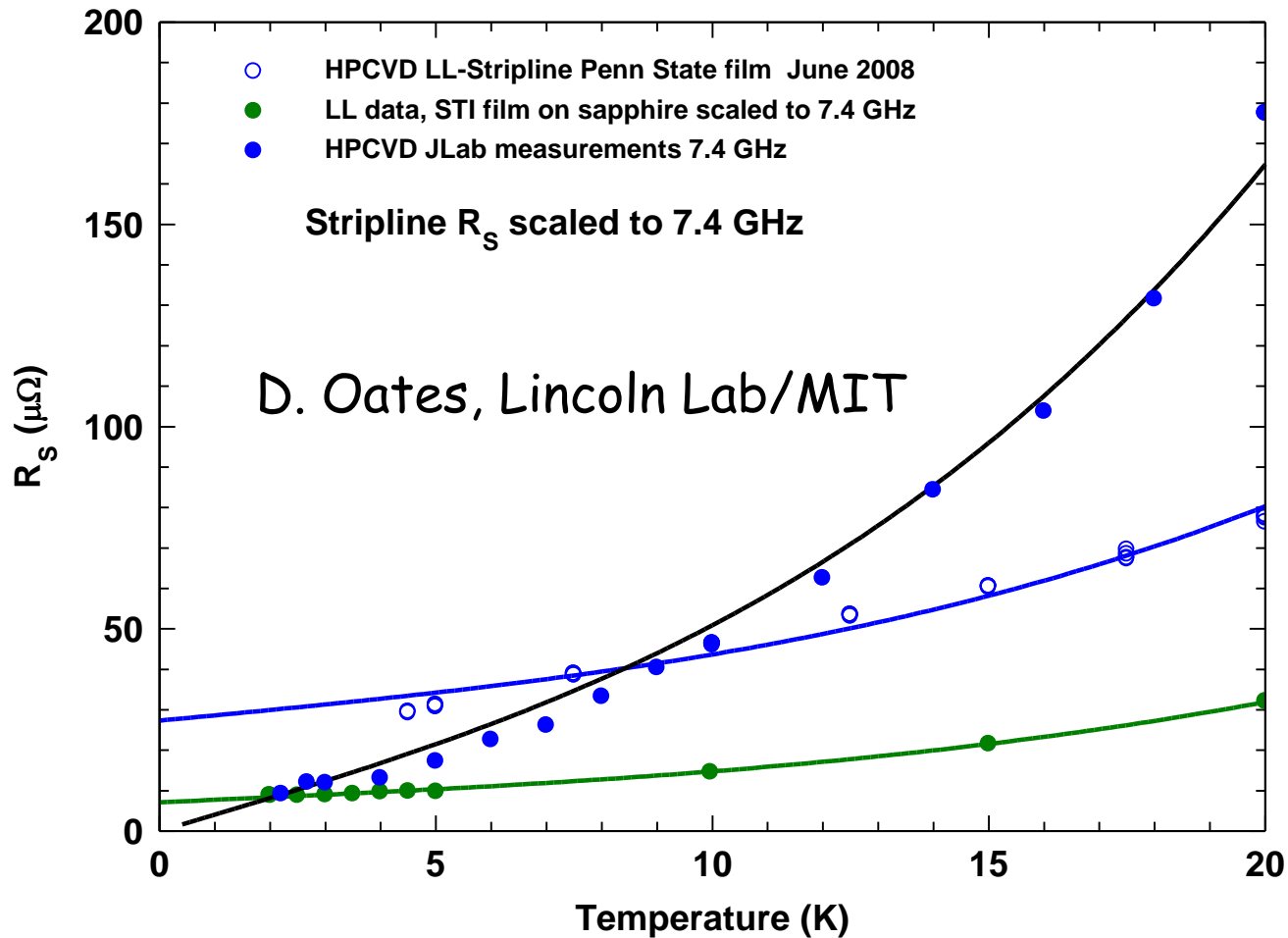
R_s for MgB_2

The achieved field with little RF loss is increasing! (Now ~800 Oe or higher considering the field enhancement effect at the edges) (Xiaoxing Xi at Temple Uni.)



Test at MIT
scaled to 1.5 GHz

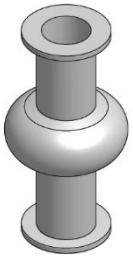
Low-Power Surface Resistance of MgB₂



Fits

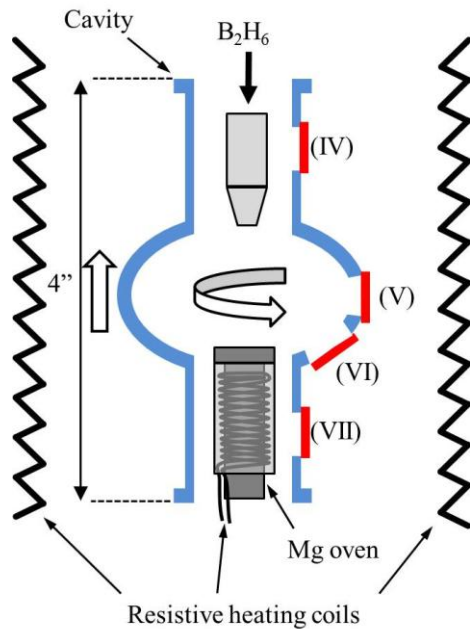
$$R_s(T) = a \left(\frac{1}{\bar{\beta}} + \left(\frac{15}{8} \right) \frac{1}{\bar{\beta}^3} \right) + c, \quad \bar{\beta} = \frac{\Delta_\pi(0)}{k_B T}, \quad \Delta_\pi = 2.3 \text{ meV}$$

MgB₂ @ Temple University – cavity coating

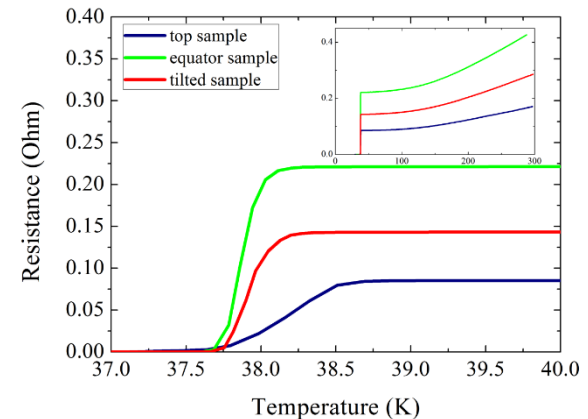
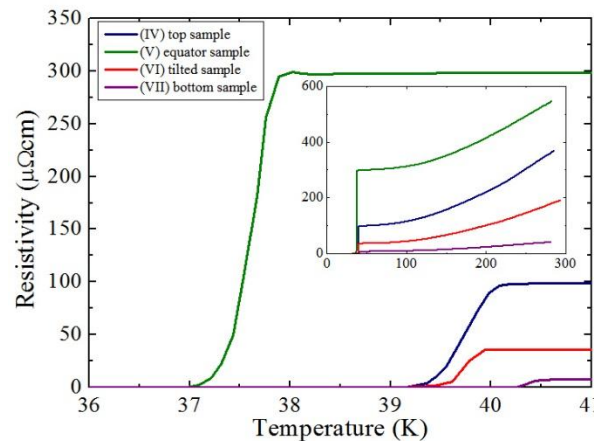


In-situ depositions on dummy cavity

- A stainless steel dummy cavity was fabricated to resemble actual cavity.
- MgB₂ films were deposited, through openings at different locations of the dummy cavity, on c-cut sapphire mounted on the outside.



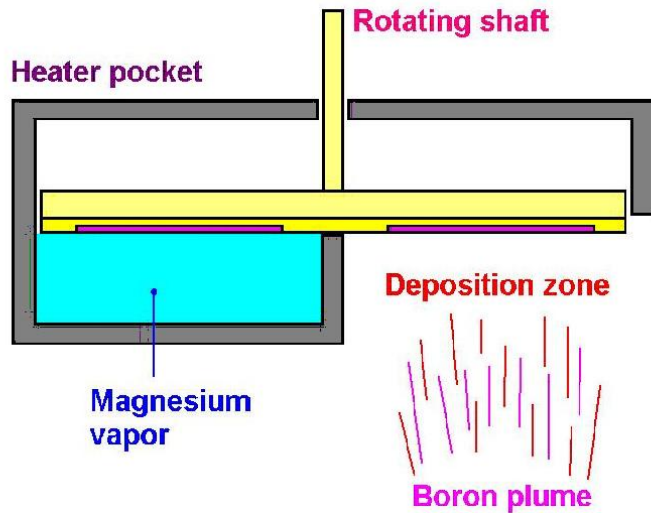
- The films exhibit residual resistivity between 7.4 and 267 $\mu\Omega\text{cm}$ and RRR between 2 and 6.
- $T_c(0)$ ranges between 36.9 K and 40.2 K.



Different sequences of deposition steps on dummy cavity

MgB₂ - Reactive Evaporation

Superconducting Technologies Inc.



T. Tajima, LANL

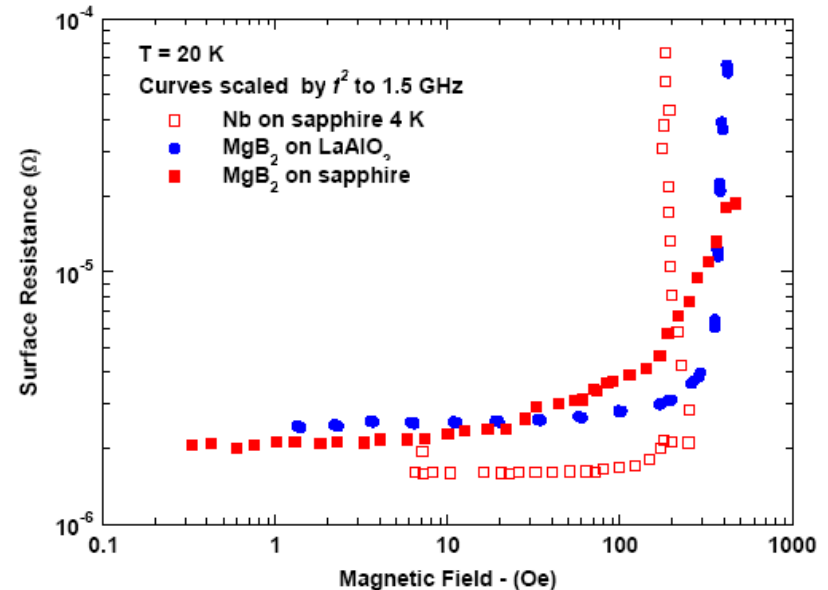
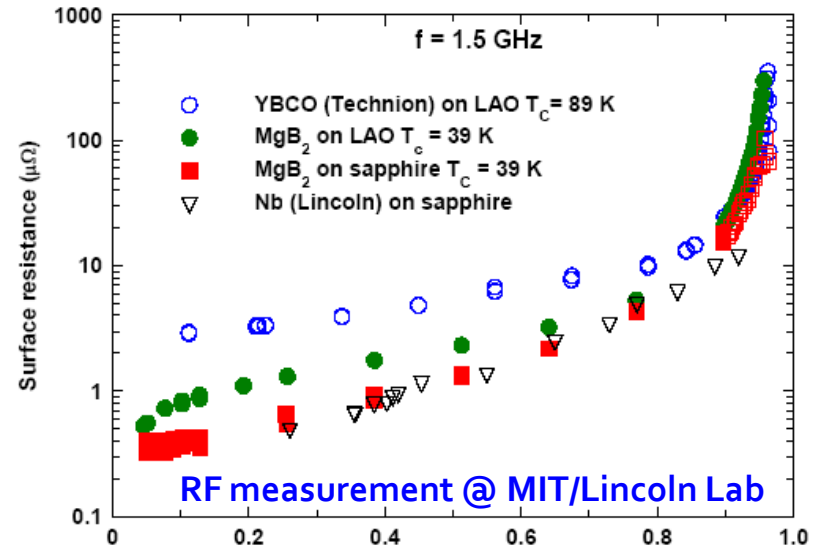
In-situ reactive evaporation @ 550°C

Compared to Nb:
 Higher T_c
 low resistivity
 larger gap
 higher critical field

B.H. Moeckly et al., *IEEE Trans. Appl. Supercond.* 15 (2005) 3308.
 T. Tajima et al, *Proc. PAC05*.

B.H. Moeckly, *ONR Superconducting Electronics Program Review*
 Red Bank, NJ, February 8, 2005

Oates, Agassi, and Moeckly, *ASC 2006 Proceeding*

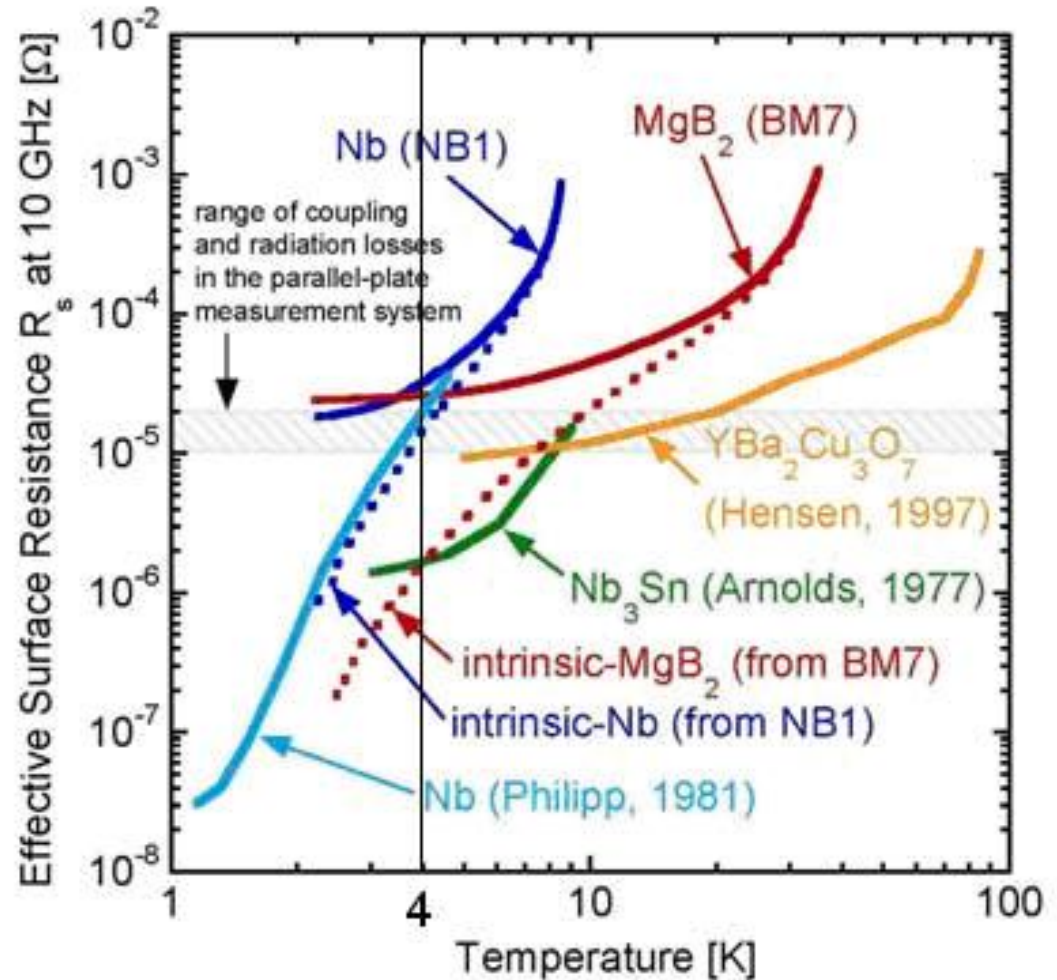


RF Surface Resistance (R_s) at 10 GHz: LANL measurements compared to the data from other references, and prediction

- Dotted line is the predicted BCS resistance by subtracting the residual resistance (temperature independent)
- R_s lower than Nb at 4K
- Still residual resistance dominates at low temperatures

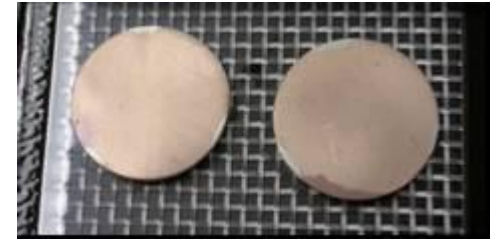
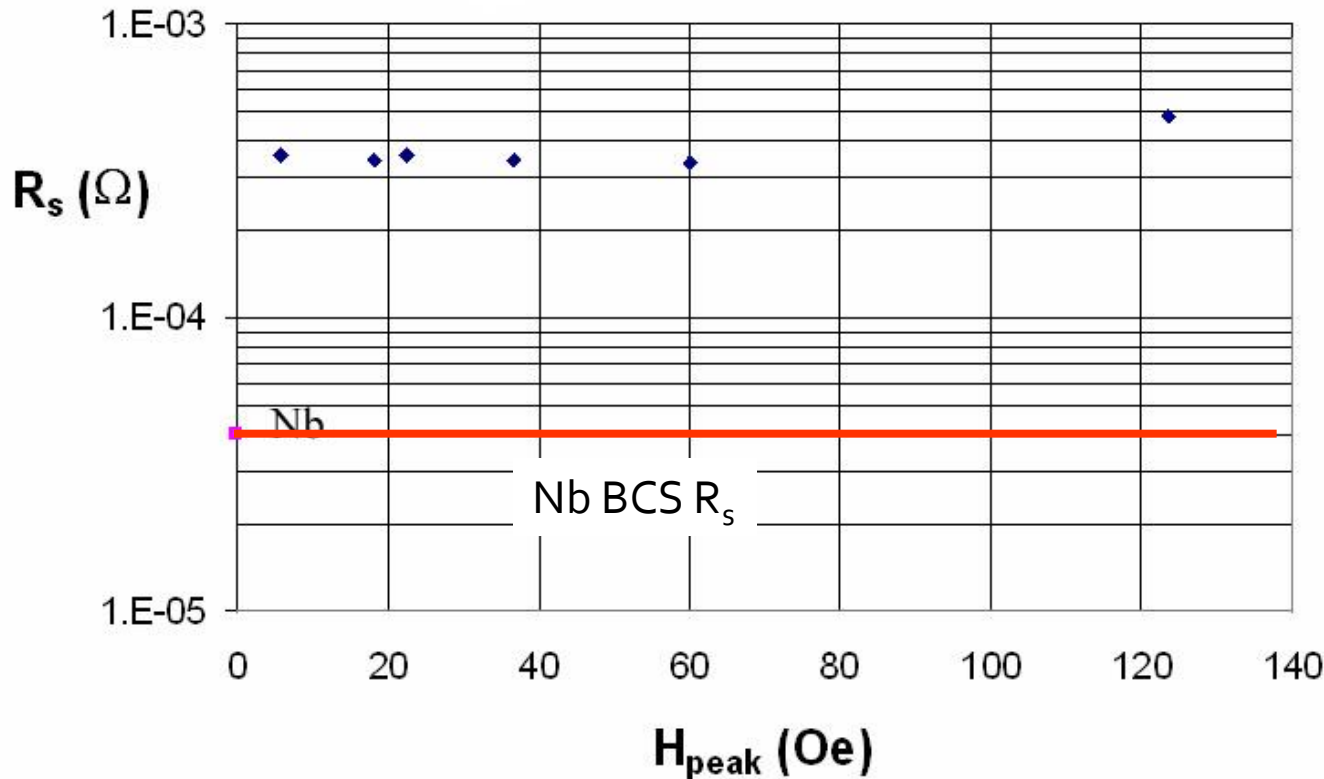
Generally, $R_s \propto f^2$ at $T < T_c/2$

- A.T. Findikoglu et al., NSF/DOE Workshop on RF Superconductivity, Bethesda, MD, Aug. 29, 2003.
- B.H. Moeckly et al., IEEE Trans. Appl. Supercond. 15 (2005) 3308.



R_s for MgB_2

R_s Power Dependence Test of the sample coated on a rough Nb disk at STI
with reactive evaporation method in 2004
Normalized to 10 GHz



First attempt to coat on a Nb substrate (1.5 cm disk). R_s was higher than Nb due to the rough ($R_a \sim 400\text{nm}$) substrate.

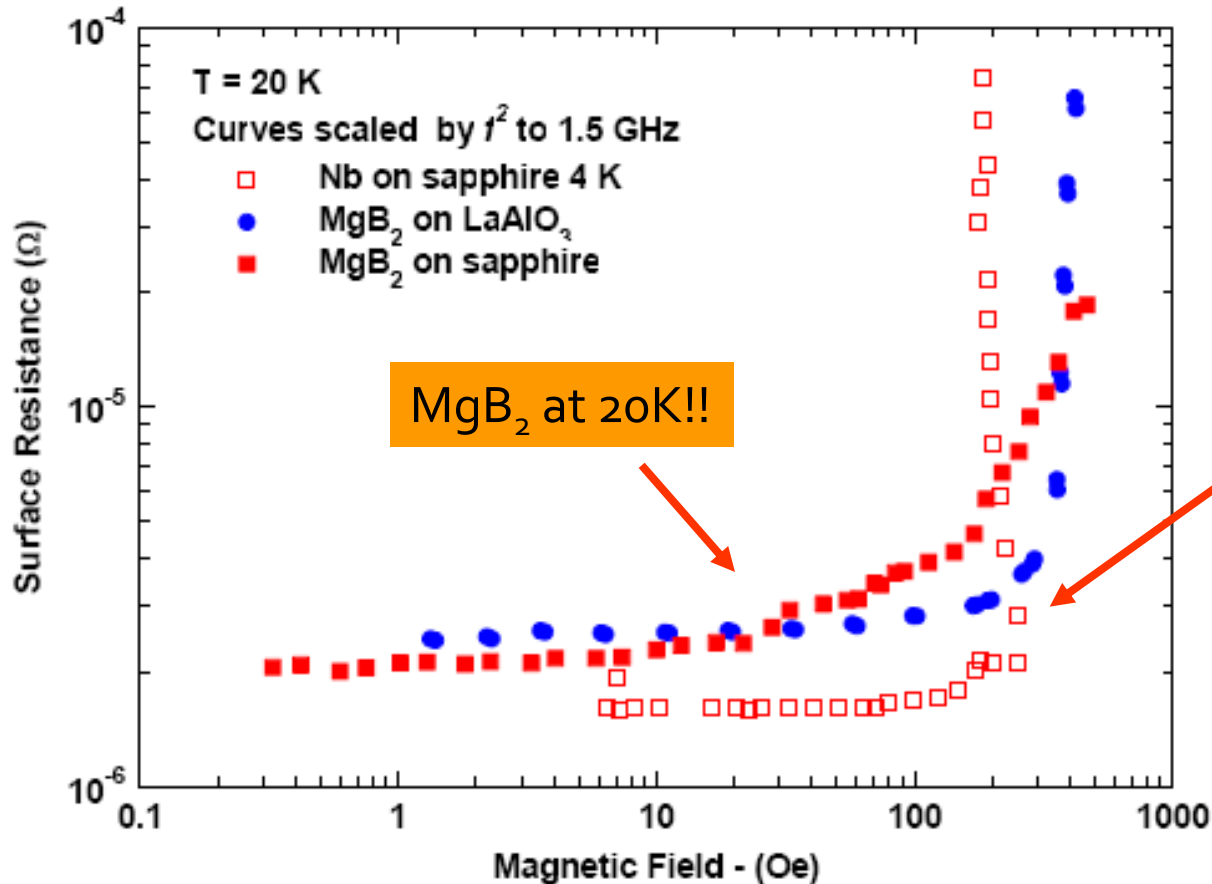
Test at Cornell with TE_{011} Nb cavity at 4.2 K.

T. Tajima et al., Proc. PAC2005, p. 4215

R_s Power Dependence Test showed little increase up to ~ 120 Oe!

R_s for MgB_2

Results at MIT in 2006 showed R_s comparable to Nb even at 20 K! and the field at which the R_s start to increase rapidly was higher than Nb!!



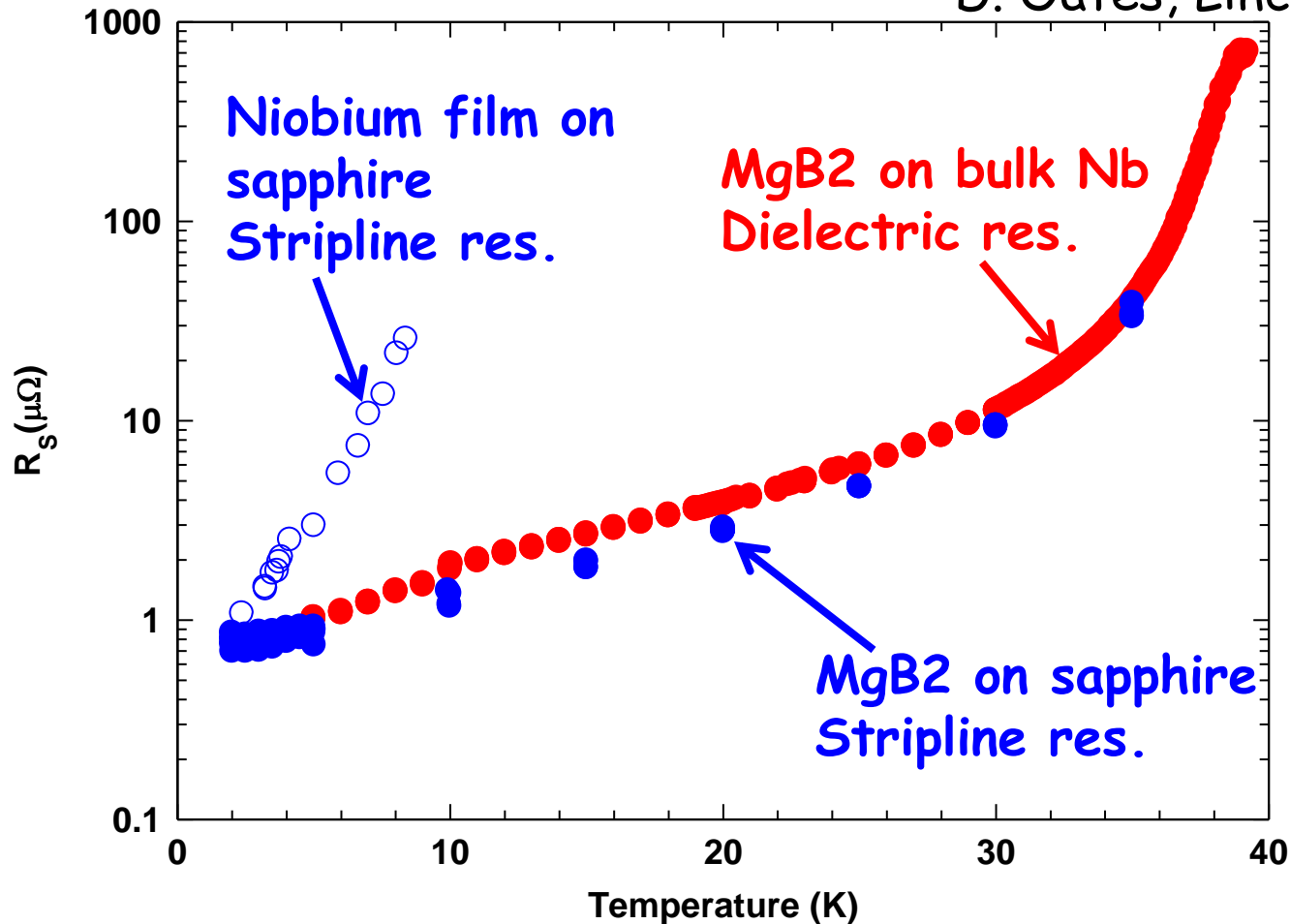
Coated at STI with reactive evaporation
Thickness: 500 nm

Nb at 4.2 K, Enhanced field at the edges may have caused this take off at low gradient.

Oates et al., IEEE Trans. Appl. Supercond. 17 (2007) 2871.

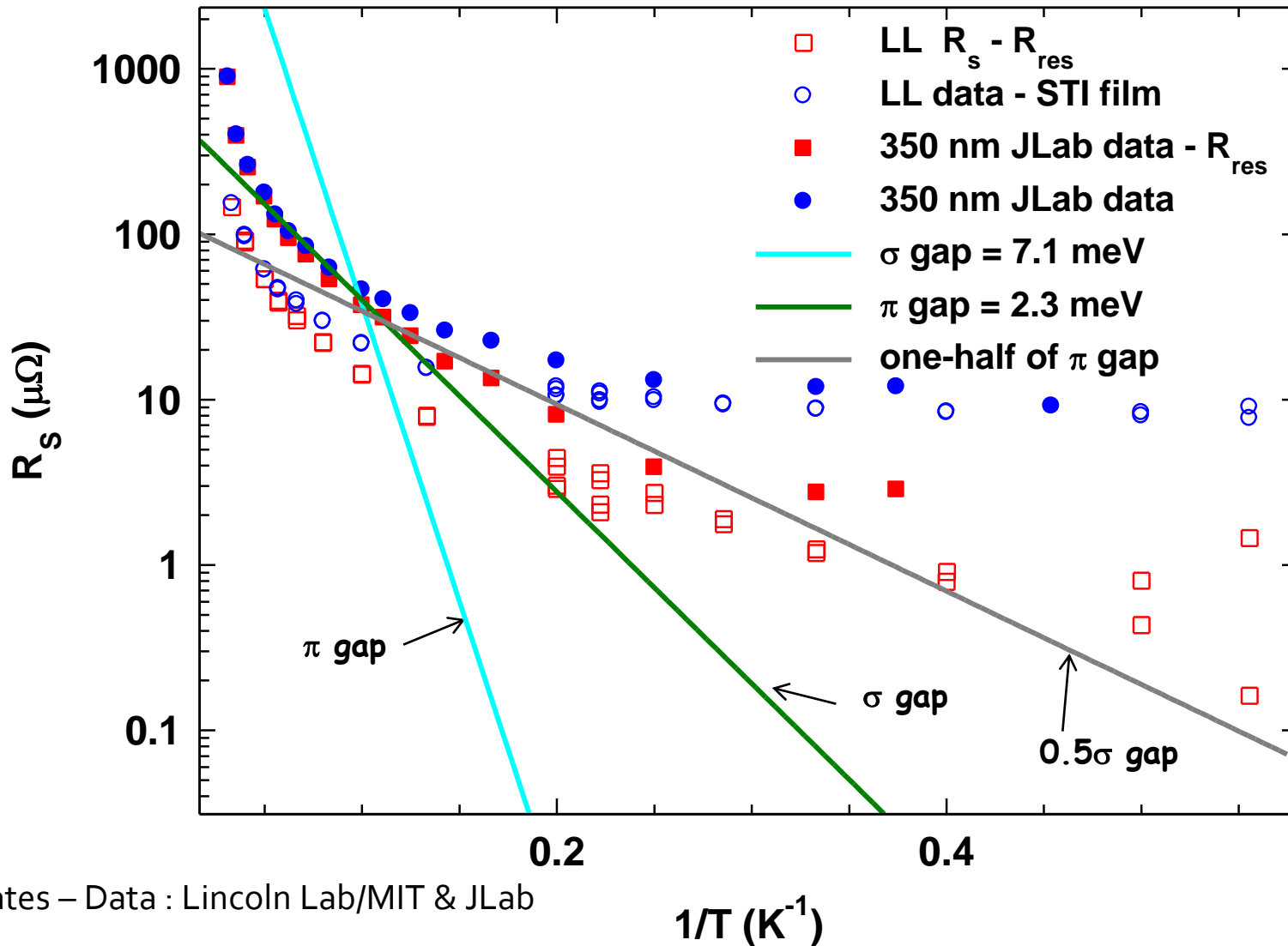
Low-Power $R_S(T)$: MgB_2 and Nb

D. Oates, Lincoln Lab/MIT



R_S extrapolated to 2.2 GHz by f^2 for the dielectric-resonator data

R_s vs $1/T$ Semi-log Plot



D. Oates – Data : Lincoln Lab/MIT & JLab

MgB₂ – Challenges

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

Challenges

Film properties degrade with exposure to moisture: resistance goes up, T_c goes down

Clean cavity surface leads to degradation in water and moisture
... need of a cap layer?

Safety ... procedures for use of diborane

A new superconducting family

Oxyprictide base

ReOMP_n

- M = Fe, Co, Ni
- Pn = As or P
- Re = La, Nd, Sm, Pr

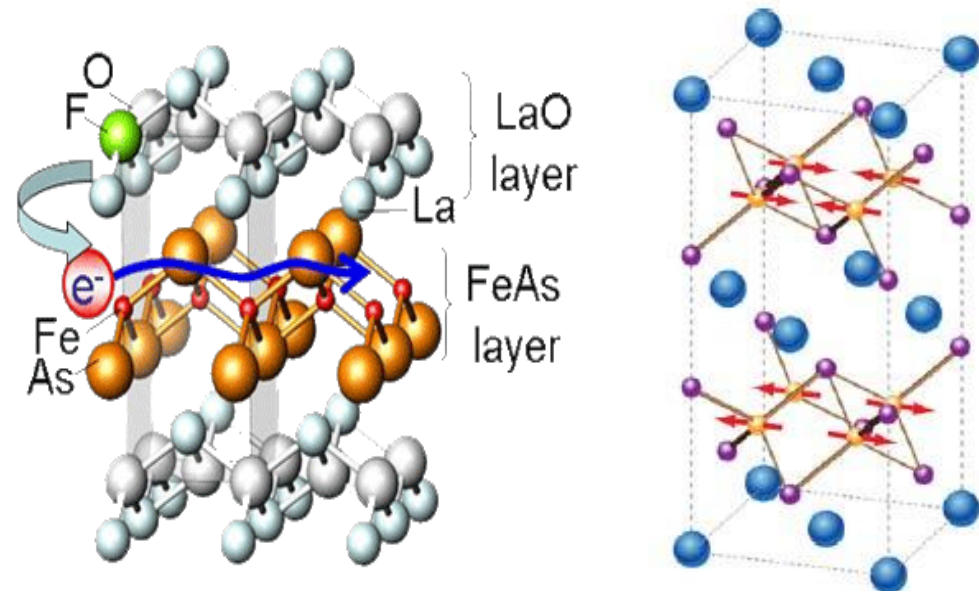
Layered as HTS –superconducting
AsFe layers and T_c from <10K to 55K

Superconductivity occurs on the FeAs layer
with magnetic pair-breaking Fe²⁺ions

Another families: Ba_{1-x}K_xFe₂As₂

Up to a few thousand compounds

Materials usually brittle



Iron-Based Layered Superconductor La[O_{1-x}F_x]FeAs (x = 0.05–0.12)
with T_c = 26 K

Yoichi Kamihara,*† Takumi Watanabe,‡ Masahiro Hirano,^{1,§} and Hideo Hosono^{1,§}

ERATO-SORST, JST, Frontier Research Center, Tokyo Institute of Technology, Mail Box S2-13, Materials and Structures Laboratory, Tokyo Institute of Technology, Mail Box R3-1, and Frontier Research Center, Tokyo Institute of Technology, Mail Box S2-13, 4259 Nagatsuta, Midori-ku, Yokohama 226-8503, Japan

Received January 9, 2008; E-mail: hcsono@mssl.titech.ac.jp

Tests in magnetic fields up to 45 T suggest the H_{c2} of
LaFeAsO_{0.89}F_{0.11} may be ~64 T.
A different La-based material (La_{0.8}K_{0.2}FeAsO_{0.8}F_{0.2})
tested at 6 K predicts H_{c2} ~122 T.

Can pnictides be useful for TFSRF?

For s-wave members of the pnictide family, one can expect a much lower R_s at 2K because

$$\Delta_{\text{oxy}} = 5\text{-}10 \text{ meV} > \Delta_{\text{Nb}_3\text{Sn}} = 3 \text{ meV}$$

Normal skin effect ($l \ll \lambda$): multiple impurity scattering in the λ -belt:

$$R_s \sim (\mu_0^2 \omega^2 \lambda^3 \sigma_n \Delta / T) \exp(-\Delta / T)$$

Anomalous skin effect ($l \gg \lambda$) in the clean limit:

$$\text{Effective } \sigma_{\text{eff}} \sim e^2 n \lambda / p_F$$

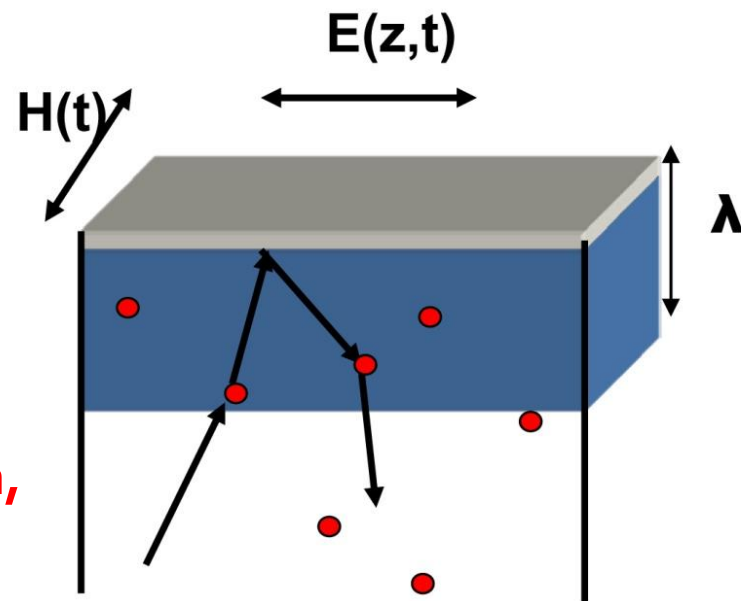
High $\rho_n \sim 1 \text{ m}\Omega\text{cm} \sim 10 \rho_n \text{ MgB}_2$, **big $\lambda = 180\text{-}250 \text{ nm}$,**

low $H_{c1} \sim 10 \text{ mT}$

R_s can be much lower than R_s of Nb_3Sn , but TF multilayer coating is necessary

High quality epitaxial films have been grown –

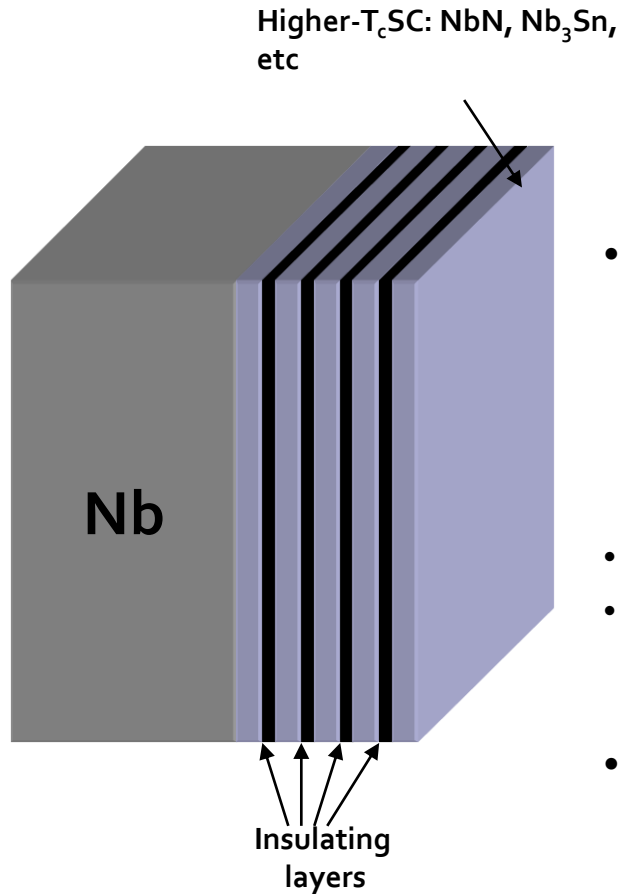
MBE - C.-B. Eom (UW)



SIS Multilayers

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

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 $d < \lambda$

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

- Strong increase of H_{c1} in films allows using RF fields $> H_c$ of Nb, but lower than those at which flux penetration in grain boundaries may become a problem

540 mT for Nb₃Sn or 1T for pnictides

-high H_{c1} => no transition, no vortex in the layer

-applied field is damped by each layer

- insulating layer prevents Josephson coupling between layers
- applied field, i.e. accelerating field can be increased without vortex nucleation
- Strong reduction of BCS resistance because of using SC layers with higher Δ (Nb₃Sn, NbN, etc)

=> low R_{BCS} at low field => higher Q_0

=> Opportunity to move operation from 2K to 4.2K

SIS Multilayers – The Benefits

- Multilayer S-I-S-I-S coating could make it possible to take advantage of superconductors with much higher H_c , than those for Nb without the penalty of lower H_{c1}
- Strong increase of H_{c1} in films allows using rf fields $> H_c$ of Nb, but lower than those at which flux penetration in grain boundaries may become a problem
- Strong reduction of BCS resistance because of using SC layers with higher Δ (Nb_3Sn , NbN , ...)

The significant performance gain may justify the extra cost.

... but ...

Technical challenges, influence of composition on H_{c1} and H_c , influence of the morphology and composition at grain boundaries,

...

NbN/MgO Multilayer @ CEA Saclay/IPN-Orsay/CNRS Grenoble

Local magnetometry

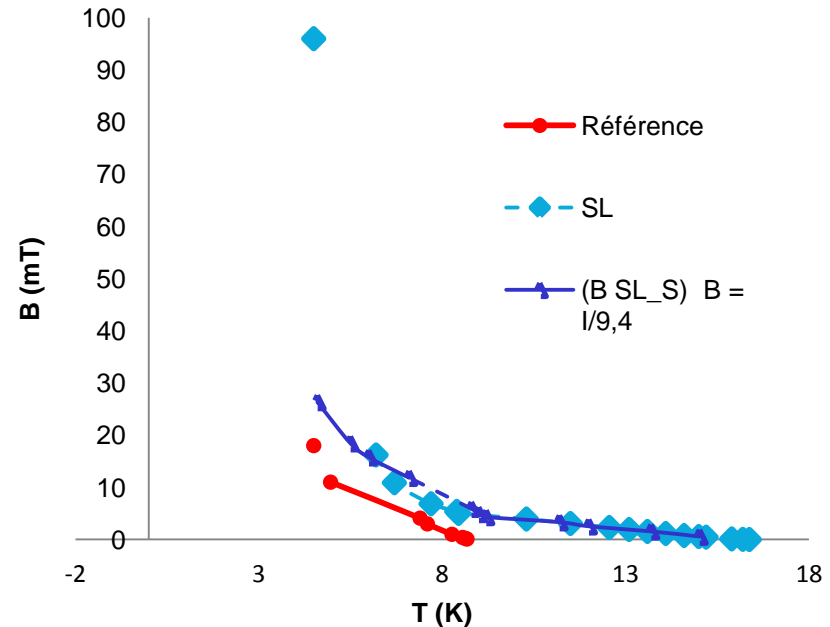
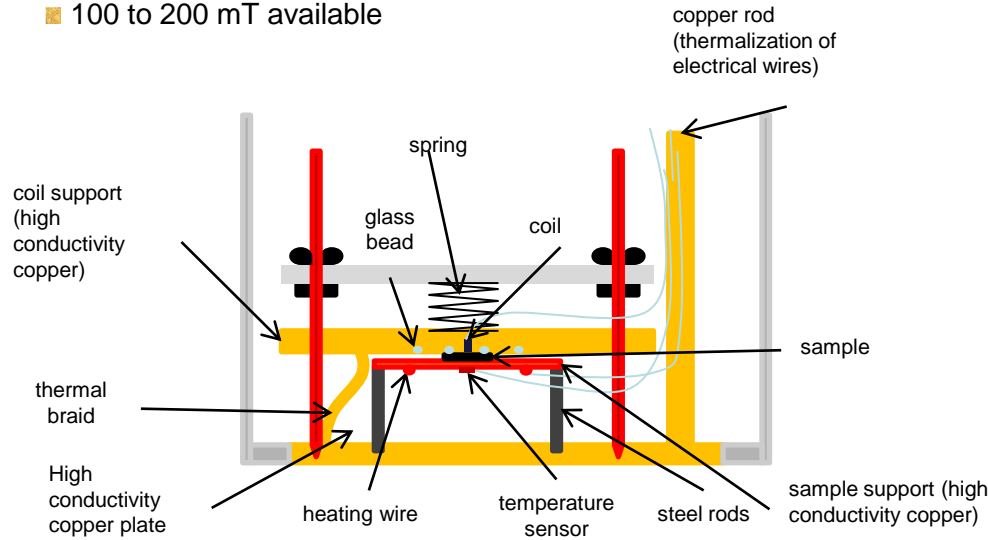
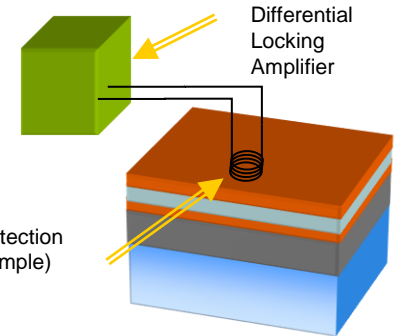
- 3rd harmonic measurement, coll. INFM Napoli

M. Aurino, et al., Journal of Applied Physics, 2005. 98: p. 123901.

Perpendicular field : field distribution can be determined analytically.

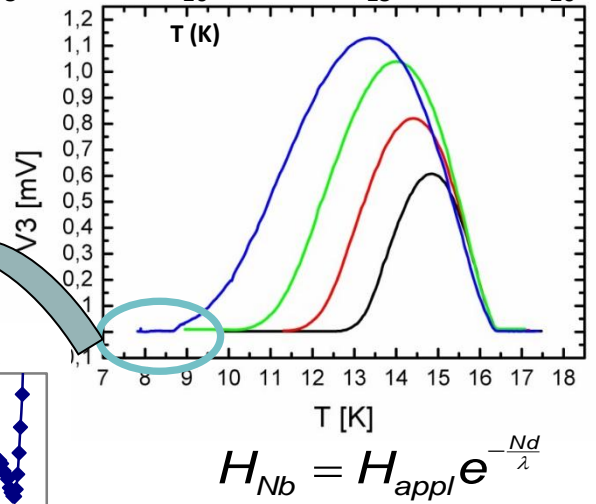
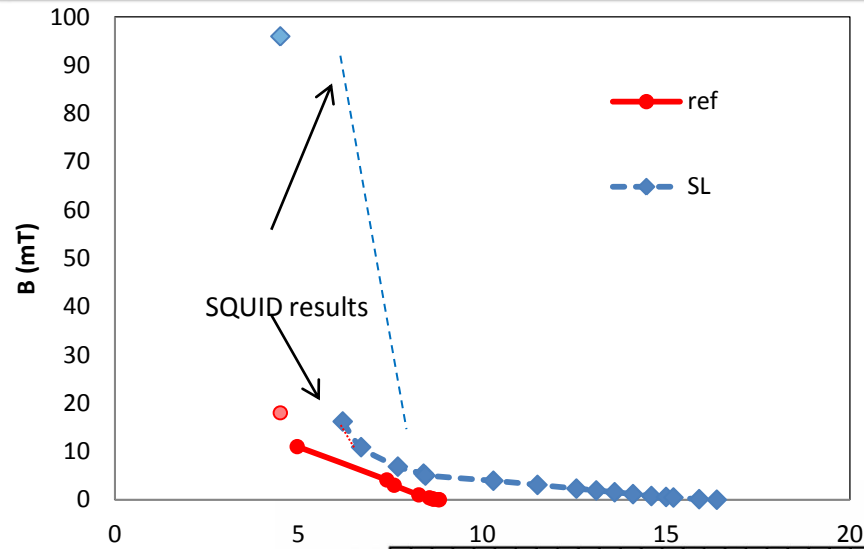
- If $r_{\text{sample}} > 4 r_{\text{coil}}$: Sample \equiv infinite plate approximation
- Applied field : perpendicular, induction (B) // surface (below B_{C1})

- thermal regulation
1.6 K $< T_p <$ 40K, automated
- 100 to 200 mT available



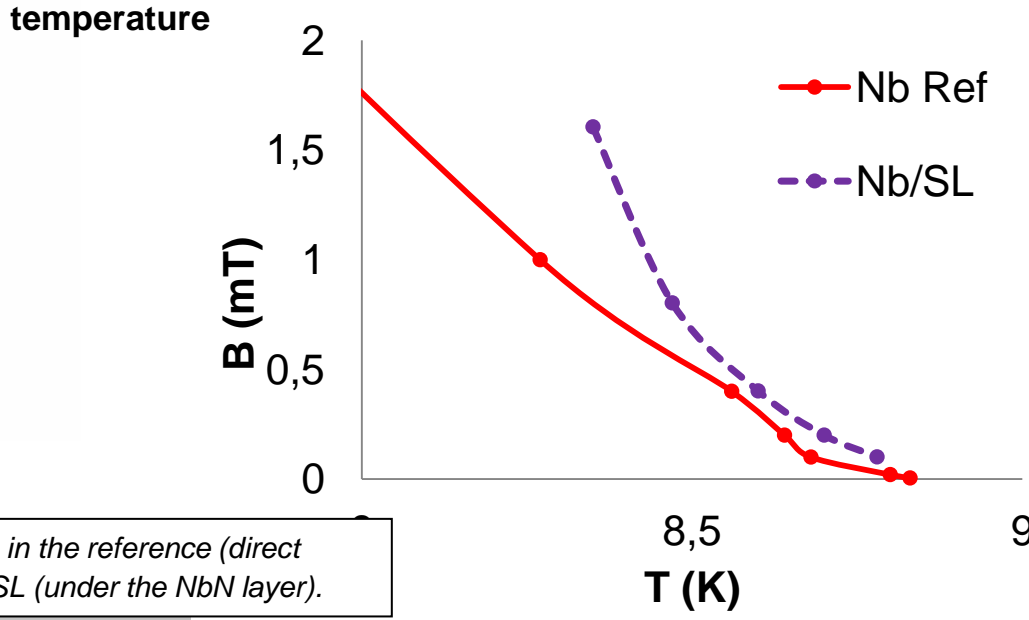
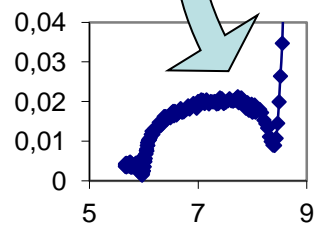
NbN/MgO Multilayer @ CEA Saclay/IPN-Orsay/CNRS Grenoble

- **SL sample** : 250 nm Nb + 14 nm MgO + 25 nm NbN
- $T_p < 8.90\text{K}$, i.e. when Nb substrate is SC, $\Rightarrow B_{C1}^{SL} \gg B_{C1}^{Nb}$
- Sample SL : small Nb signal @ $\sim T_c^{Nb}$: Nb is sensed through the NbN layer !
- Since the Nb layer feels a field attenuated by the NbN layer, the apparent transition field is higher.
- This curve provides a direct measurement of the attenuation of the field due to the NbN layer
- Need to extend measure @ higher field and lower temperature



$$H_{Nb} = H_{appl} e^{-\frac{Nd}{\lambda}}$$

B_{C1} curves for Niobium in the reference (direct measurement) and in SL (under the NbN layer).



NbN, NbTiN based ML -JLAB/W&M Collaboration

SIS Multilayer Structures:

- ✓ Nucleation studies of NbTiN, NbN on dielectrics like MgO, Al₂O₃ and reciprocally.
- ✓ Creation and characterization of a set of NbTiN, NbN.../insulator/Nb samples by UHV multi-target energetic ion deposition with well-controlled, incremented thickness. The variation of rf field properties with temperature as a function of thickness of the superconducting overlayer will provide a direct test of the Gurevich delayed flux entry model.

Study of growth modes of Superconductor/ Insulator & Insulator/superconductor

Substrates :

Single crystal Nb
Poly crystalline Nb
thick Nb/Cu films

Alternative Materials:

NbTiN, NbN
Nb₃Sn, V₃Si, Mo₃Re

Insulator:

Al₂O₃, MgO, AlN
according to
lattice mismatch
between
I and S materials

Multilayer: S/I/Nb and S/I/S/I/.../S/I/Nb

NbN/MgO Multilayer @ College William & Mary

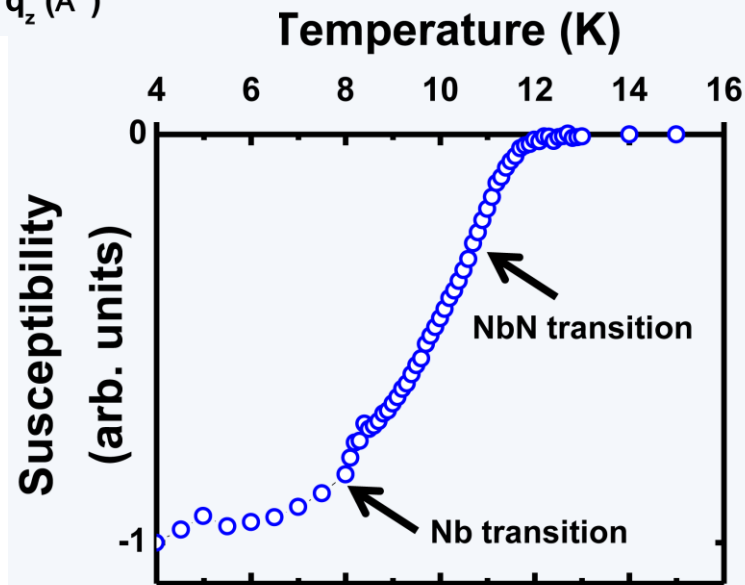
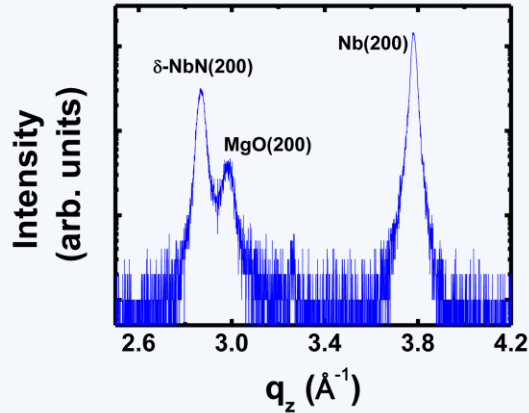
Multilayer Film

50 nm NbN

15 nm MgO

250 nm Nb

MgO (100)



- Further optimization
 - Surface modification (ion gun)
 - Alternative insulators (AlN)

NbTiN/AlN Multilayer @ Jefferson Lab (DC-MS, HiPIMS)

NbTiN deposited with various thicknesses by reactive DC sputtering at 600°C on MgO substrates



NbTiN, NbN, Mo₃Re, V₃Si coatings with Reactive Sputtering and High Power Pulse Magnetron Sputtering & MgO coating with RF sputtering

NbTiN/MgO (100)

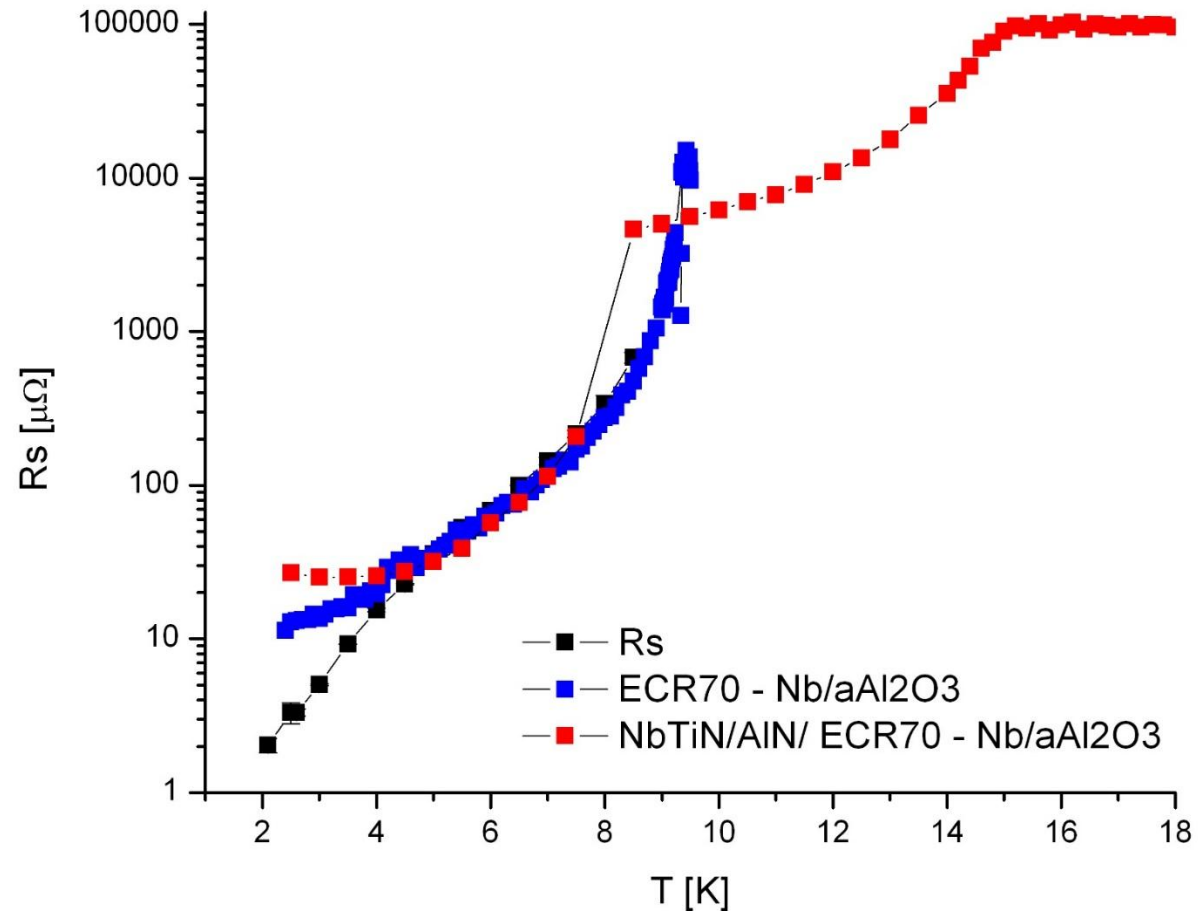
Thickness [nm]	T _c [K]	ΔT _c [K]	a _o [Å]
10	12.11	0.91	4.3487
35	16	0.39	
50	15.97	0.16	4.3644
100	16.57	0.21	4.3657
1625	16.95	0.06	4.3618

NbTiN layers have been coated at various temperatures on MgO (100) and AlN ceramic substrates.

The NbTiN films exhibit the cubic d-phase and T_c above 16K for thicknesses larger than 30-50 nm and coating temperatures of 450°C or higher.

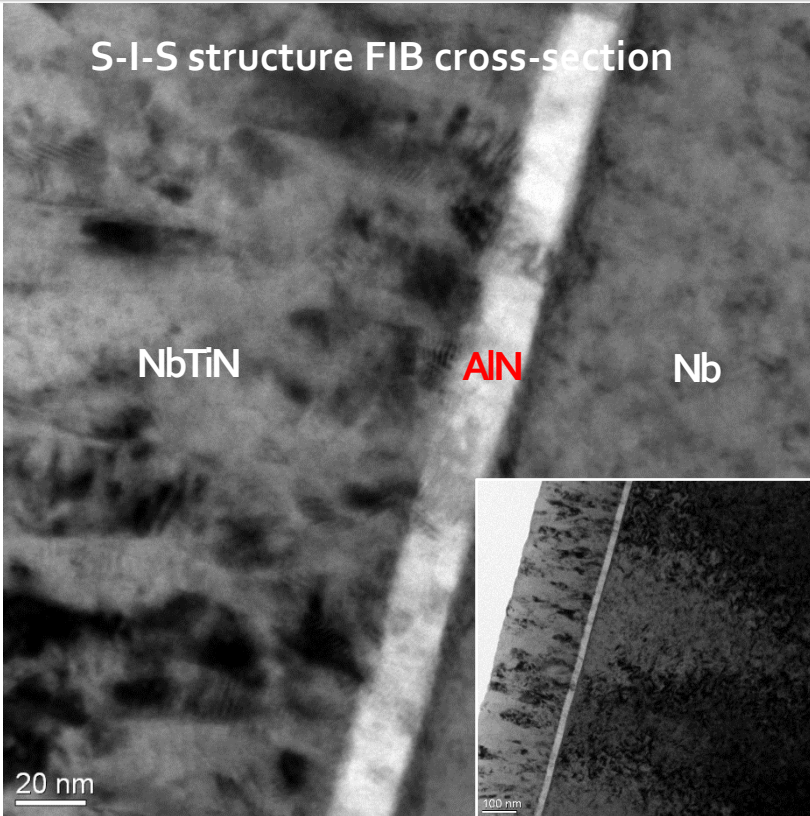
NbTiN/AlN Multilayer @ Jefferson Lab (DC-MS, HiPIMS)

The SIS structure coated on the ECR Nb/(11-20)Al₂O₃ film exhibits a suppressed T_c for the Nb film compared to the measurement prior to the SIS coating. This is most likely due to the Nb oxide reduction and oxygen diffusion during the bake at 600 °C. The NbTiN has a T_c of about 15K.



NbTiN/AlN Multilayer @ Jefferson Lab (DC-MS, HiPIMS)

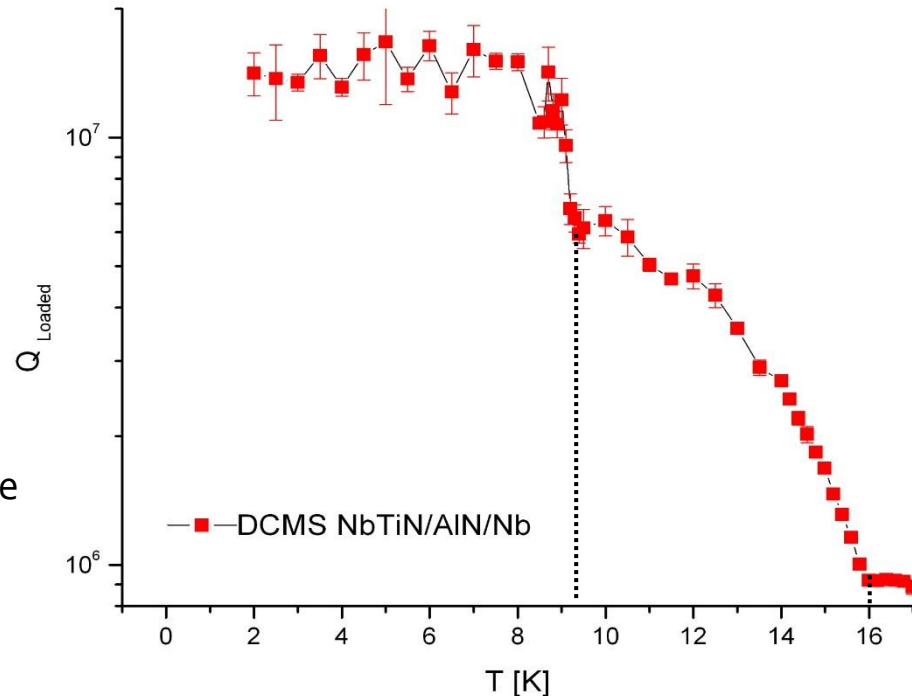
S-I-S structure FIB cross-section



NbTiN/AlN/Nb SIS structure coated at 450°C in-situ after a 24h-bake at 600°C. Annealing at 450°C for 4 hours.

XRD measurements (θ - 2θ) reveal lattice parameters of respectively 3.301Å, 4.041Å and 4.330 Å for the Nb substrate, AlN and NbTiN layers.

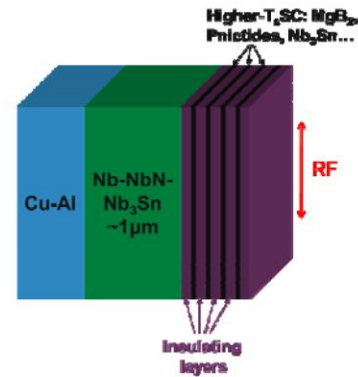
The SIS structure coated on the Nb exhibits a T_c for the NbTiN of 16K . RF measurements are in going.



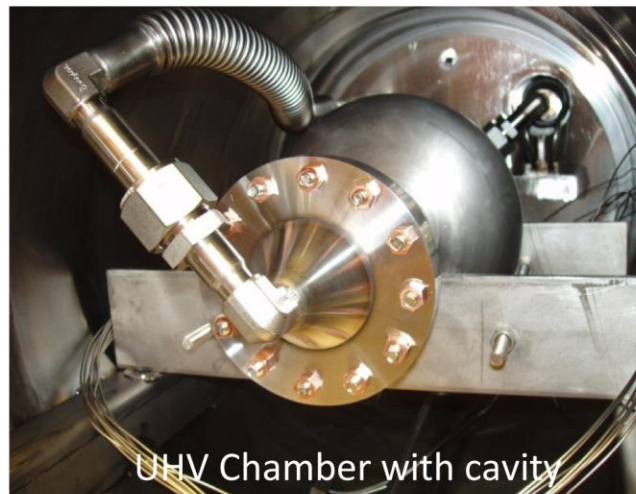
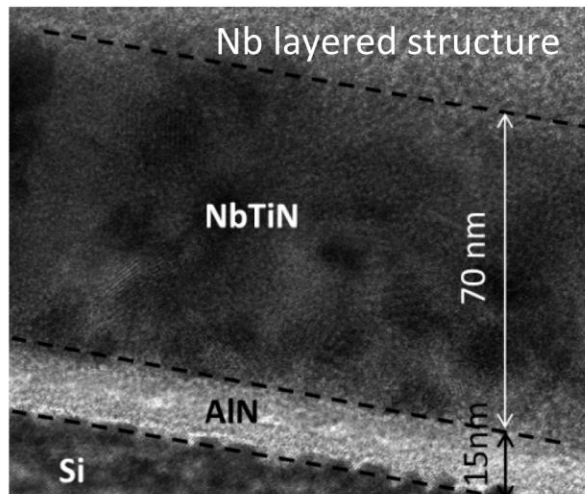
NbTiN/AlN Multilayer @ ANL (Atomic Layer Deposition -ALD)

- Able to synthesize better superconductors than Niobium:
 - NbTiN with new ALD chemistry is very pure (<0.05%)
 - NbTiN $T_c = 14\text{K}$ for 60 nm thick film (bulk=16K)

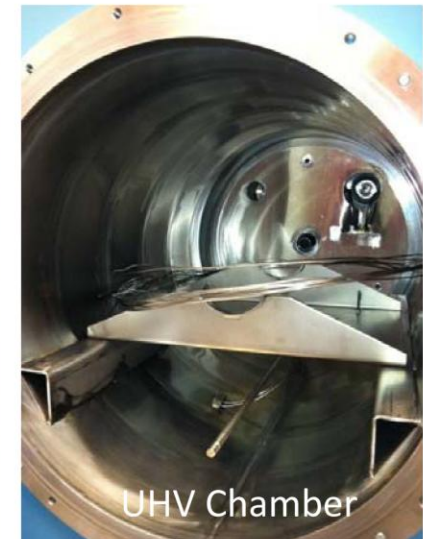
- Multilayer structure:
Aluminum Nitride (AlN) + NbTiN
works perfectly:
(15 nm AlN/ 70 nm NbTiN) x n



ALD UHV Oven



UHV Chamber with cavity



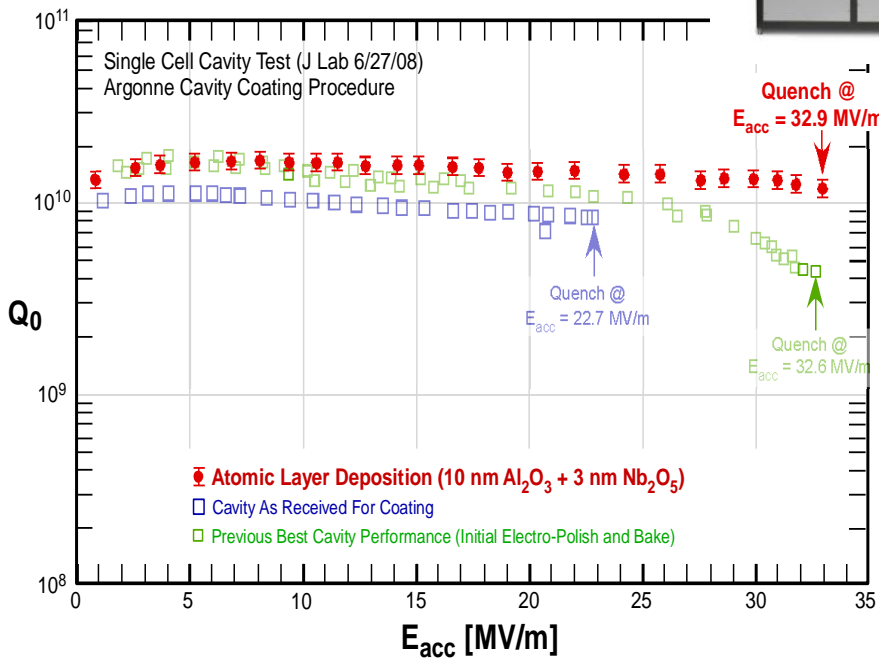
UHV Chamber

NbTiN/AlN Multilayer @ ANL (ALD)

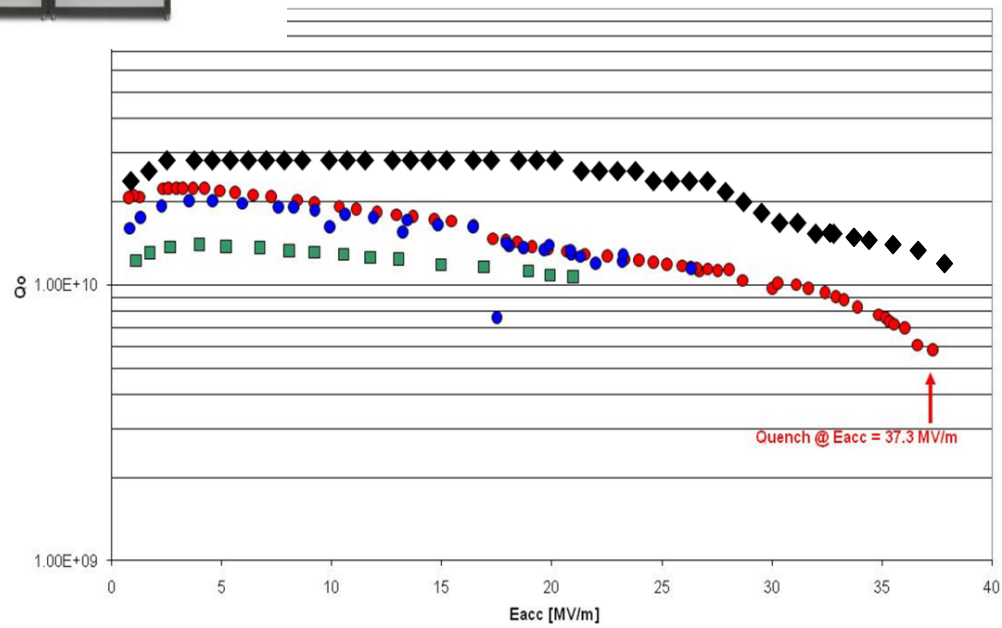


TD_Large Grain_Baked
T = 2K

● Baseline ■ ALD2Test1a ● ALD2Test1



ALD dielectric layer compatible with high E or B

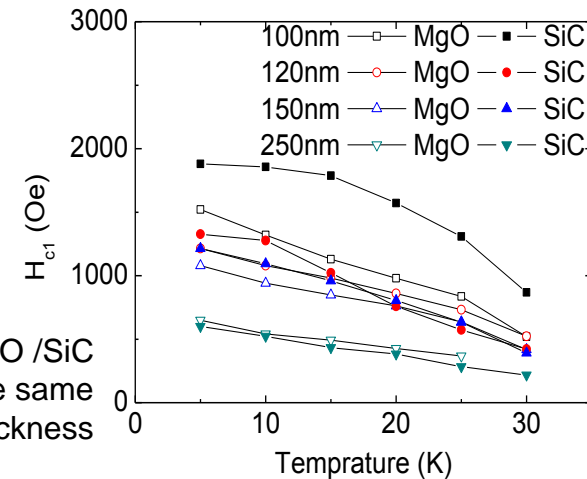


ALD + annealing improves the Q & Q-slope

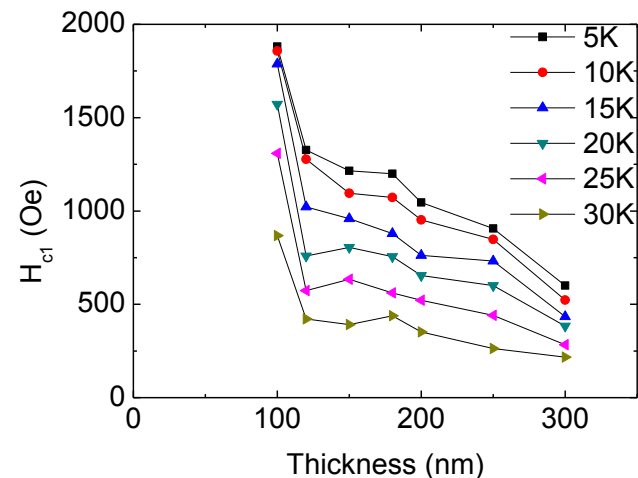
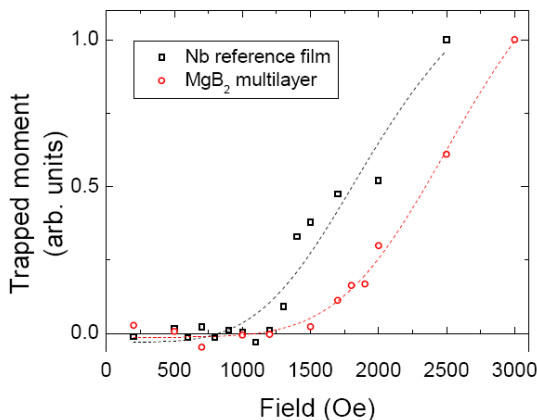
MgB₂ based Multilayer @ Temple University

Enhancement of H_{c1} in thin MgB₂ films

- 2 group of samples were studied:
 - Single crystalline thin films on SiC(0001) substrate
 - Polycrystalline thin films on amorphous MgO layer on SiC(0001)
- $H_{c1}(5K)$ is enhanced when the film thickness decreases, for films on both single crystal SiC substrate and polycrystalline MgO films.
- $H_{c1}(5K) \sim 2000$ Oe in 100nm-thick MgB₂ film.



- Results show that MgB₂ thin films with $d > \lambda$ also have enhanced H_{c1} , which makes it possible to use single layer coating to increase H_{c1} of cavities.
- Films grown on MgO buffer layers with thicknesses of 100-250 nm have the same enhanced H_{c1} as thin films grown on bare SiC substrates.



films on bare SiC substrates

Samples: Temple University/ H_{c1} Measurements: William&Mary

Oxypnictides based Multilayer structures@ ODU

A project on the development of
SIS structures based on
oxypnictides
is starting at
Old Dominion University
(A. Gurevich)
in collaboration with University
Wisconsin-Madison

oxypnictides

Material	T_c (K)
$\text{LaO}_{0.89}\text{F}_{0.11}\text{FeAs}$	26 ^[9]
$\text{LaO}_{0.9}\text{F}_{0.2}\text{FeAs}$	28.5 ^[10]
$\text{CeFeAsO}_{0.84}\text{F}_{0.16}$	41 ^[9]
$\text{SmFeAsO}_{0.9}\text{F}_{0.1}$	43 ^[9]
$\text{La}_{0.5}\text{Y}_{0.5}\text{FeAsO}_{0.6}$	43.1 ^[11]
$\text{NdFeAsO}_{0.89}\text{F}_{0.11}$	52 ^[9]
$\text{PrFeAsO}_{0.89}\text{F}_{0.11}$	52 ^[12]
$\text{GdFeAsO}_{0.85}$	53.5 ^[13]
$\text{SmFeAsO}_{\sim 0.85}$	55 ^[14]

F. Hunte, J. Jaroszynski, A. Gurevich, D. C. Larbalestier, R. Jin, A. S. Sefat, M. A. McGuire, B. C. Sales, D. K. Christen & D. Mandrus (2008). "Two-band superconductivity in $\text{LaFeAsO}_{0.89}\text{F}_{0.11}$ at very high magnetic fields". *Nature* **453** (7197): 903-905.

CONCLUDING REMARKS

- ✓ Over the years, some attempts have been made to study alternative materials to Nb for applications to SRF cavities.
- ✓ Most of the sample/cavities using alternative materials have been produced by reactive magnetron sputtering or thermal diffusion.
- ✓ Use of Energetic Condensation Techniques like Vacuum Arc Deposition, ECR, or ALD? (production of very dense films with nm-scale roughness...Some trials with Vacuum Arc @ INFN-Rome, non conclusive)
- ✓ The multilayer approach opens the door to further potential improvement for SRF cavities, taking benefits from the advantages of higher T_c superconductors without the penalty of lower field onset for vortex penetration and increase the accelerating gradient and Q.
- ✓ Strong reduction of the BCS resistance for superconducting layers with higher Δ (Nb₃Sn, MgB₂, BKBO, NbN ... s-wave, fully gapped superconductors)
- ✓ New non-magnetic members of the oxypnictide family with $15\text{K} < T_c < 55\text{K}$ or BaO_{0.6}Ko_{0.4}BiO₃ with $T_c \approx 30\text{K}$: a possibility to greatly increase Q if TF coating can be developed.

CONCLUDING REMARKS cont.

- ✓ Possibility to move from 2K to 4.2K would mean huge cost saving on refrigeration in LINACS
- ✓ Higher $-T_c$ s-wave materials are often multi-gap superconductors, which do not always have better SRF performance despite higher H_c and T_c
- ✓ First experimental evidence of field enhancement with NbN/MgO/.../Nb samples with SQUID measurement with $H \parallel$ sample plane. Field penetration delayed for NbN layered samples compared to Nb sample. Limitation of method: edge effect due to sample size comparable to measurement setup size (C. Antoine et al, CEA Saclay)
- ✓ Many long-standing problems of condensed matter physics and non-equilibrium superconductivity will have to be addressed to understand nonlinear surface resistance under strong rf fields
- ✓ Multi-parameter materials optimization is required to reveal the full SRF performance potential.
- ✓ The effort for new materials research for SRF cavities is gaining amplitude. But there is still a lot of work ahead!