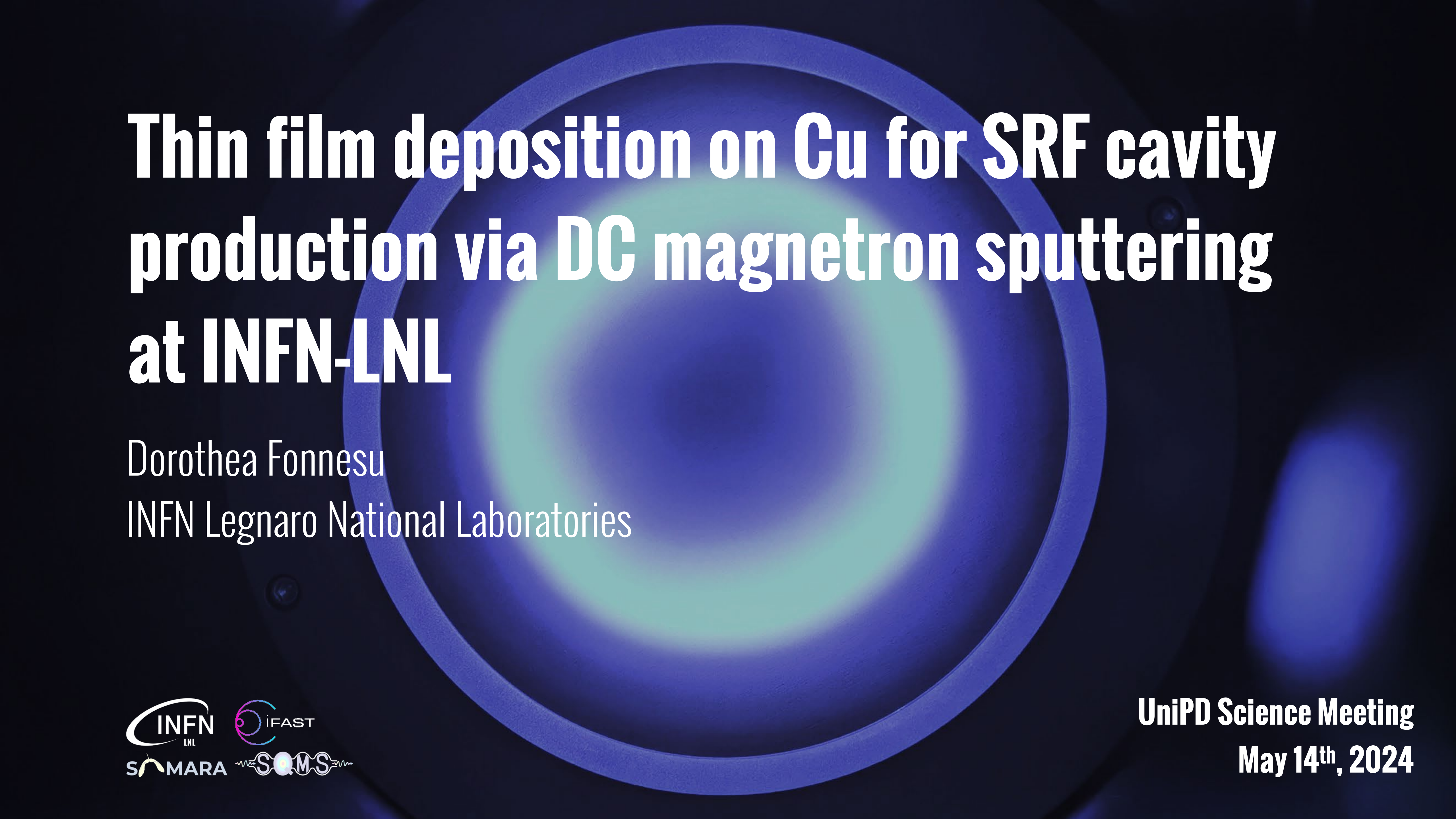


Thin film deposition on Cu for SRF cavity production via DC magnetron sputtering at INFN-LNL

Dorothea Fonnesu
INFN Legnaro National Laboratories

UniPD Science Meeting
May 14th, 2024

Outline

DC Magnetron Sputtering

1

Nb_3Sn for accelerating cavities

2

NbTi for haloscopes

3

Outline

DC Magnetron Sputtering

1

Nb_3Sn for accelerating cavities

2

NbTi for haloscopes

3

dorothea.fonnesu@lnl.infn.it

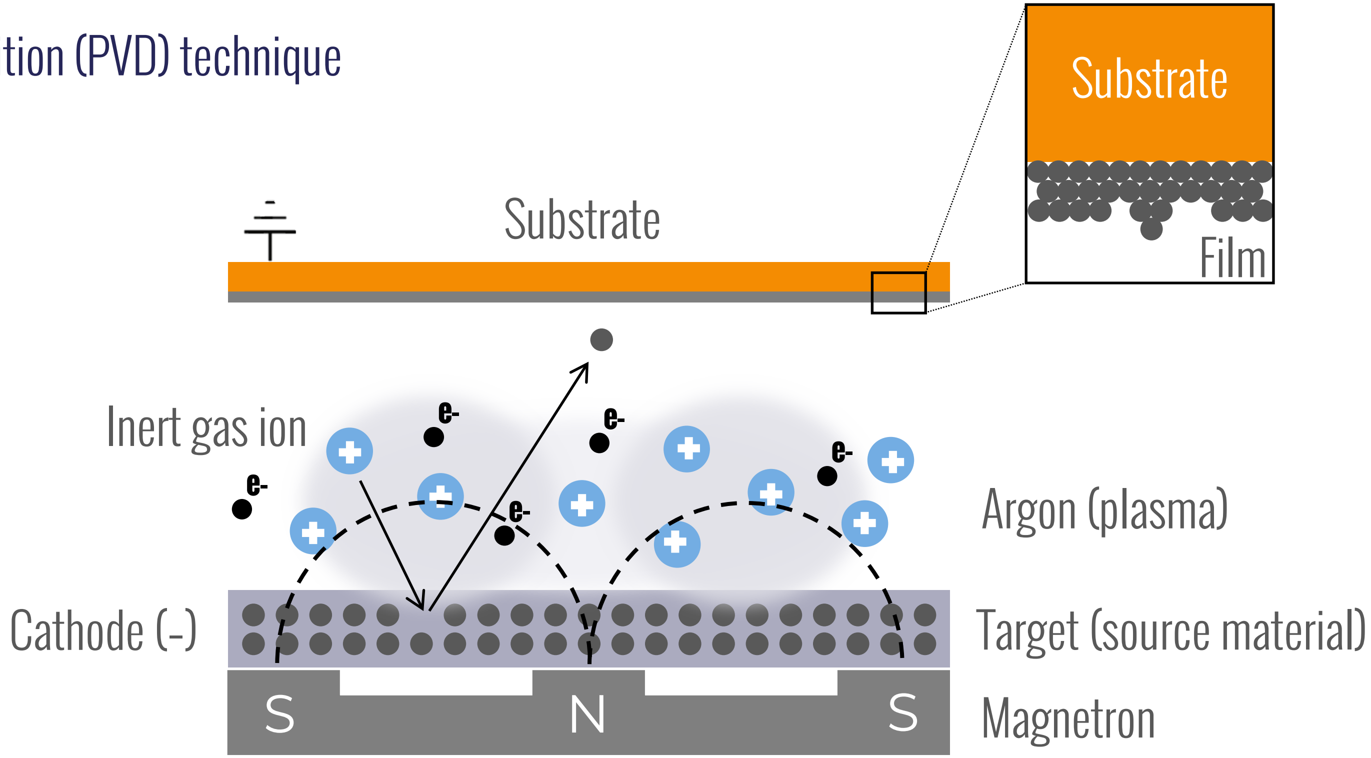
DC Magnetron Sputtering (DCMS)

Physical Vapour Deposition (PVD) technique

Substrate temperature

Gas pressure

Cathode power

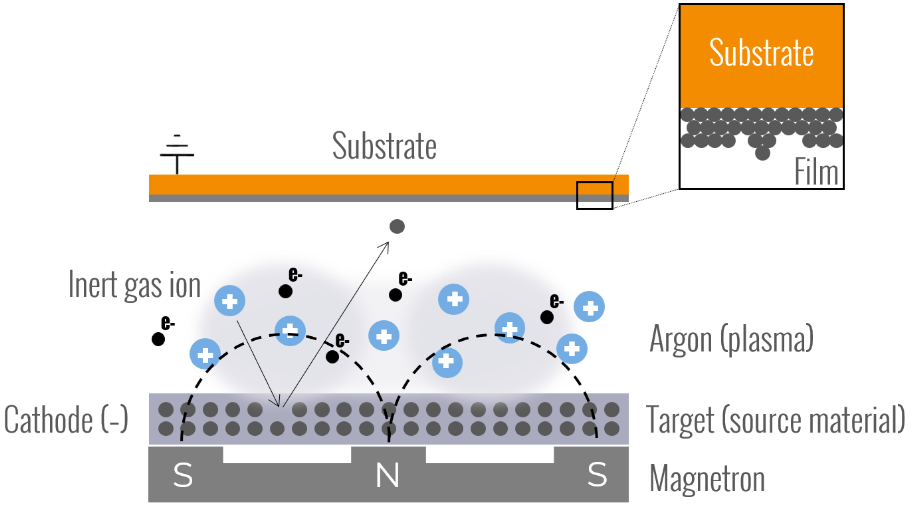


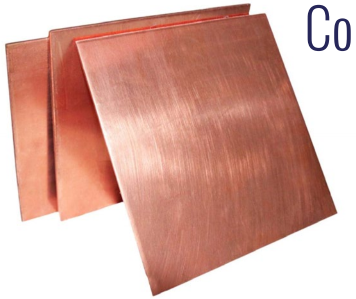
dorothea.fonnesu@lnl.infn.it

DC Magnetron Sputtering

ADVANTAGES

- ➞ Different target materials
- ➞ Different target/magnetron geometries
- ➞ Different substrate materials

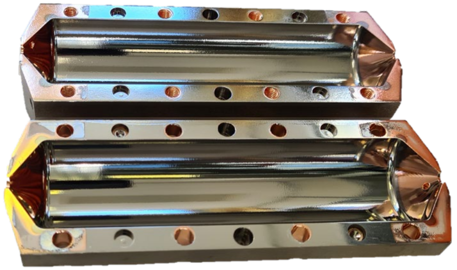



Copper


Sapphire


Niobium

SC thin films for SRF cavities



Accelerating cavities → **Nb_3Sn**

↓

Copper

↑

Haloscopes → **NbTi**

→

Material choice

Advantages

Challenges

Outline

DC Magnetron Sputtering

1

Nb_3Sn for accelerating cavities

2

NbTi for haloscopes

3

dorothea.fonnesu@lnl.infn.it

Nb_3Sn for accelerating cavities: why?

- Nb close to its ideal performance

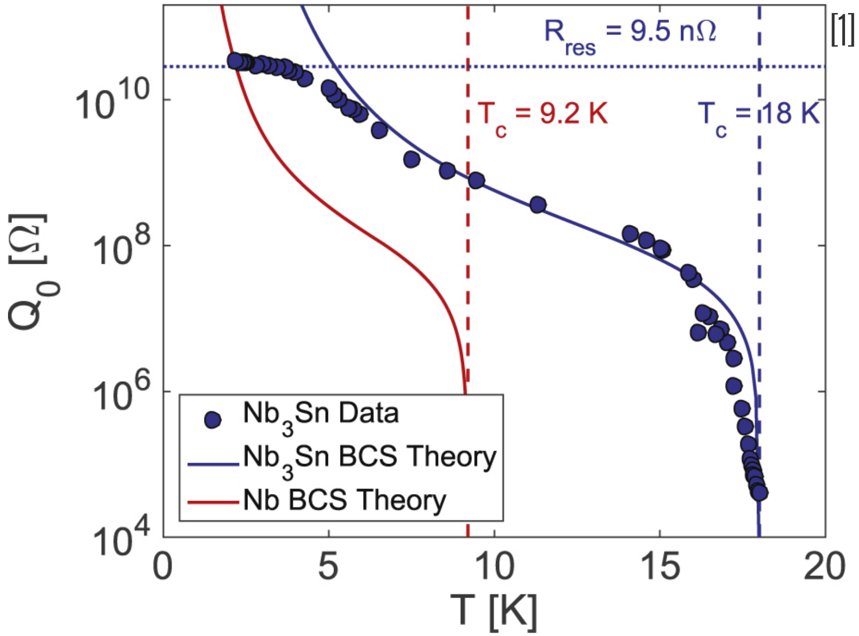
With Nb_3Sn:

- Lower R_{BCS} for a given temperature
- Operation at 4.5 K (instead of 2 K)
- Improve cryo plant efficiency by factor 3-4
- Potential demonstrated by vapour diffusion cavities:

Q_0 ~ 2 x 10^{10} at 20 MV/m (4.4 K, 1.3 GHz) [2]

*predicted

Material	T_c	H_{sh}
Niobium	9.2 K	240 mT
Nb_3Sn	**18.3 K**	**420 mT***



[1]

[1] S Posen and D L Hall 2017 *Supercond. Sci. Technol.* 30 033004
[2] S Posen et al 2021 *Supercond. Sci. Technol.* 34 025007

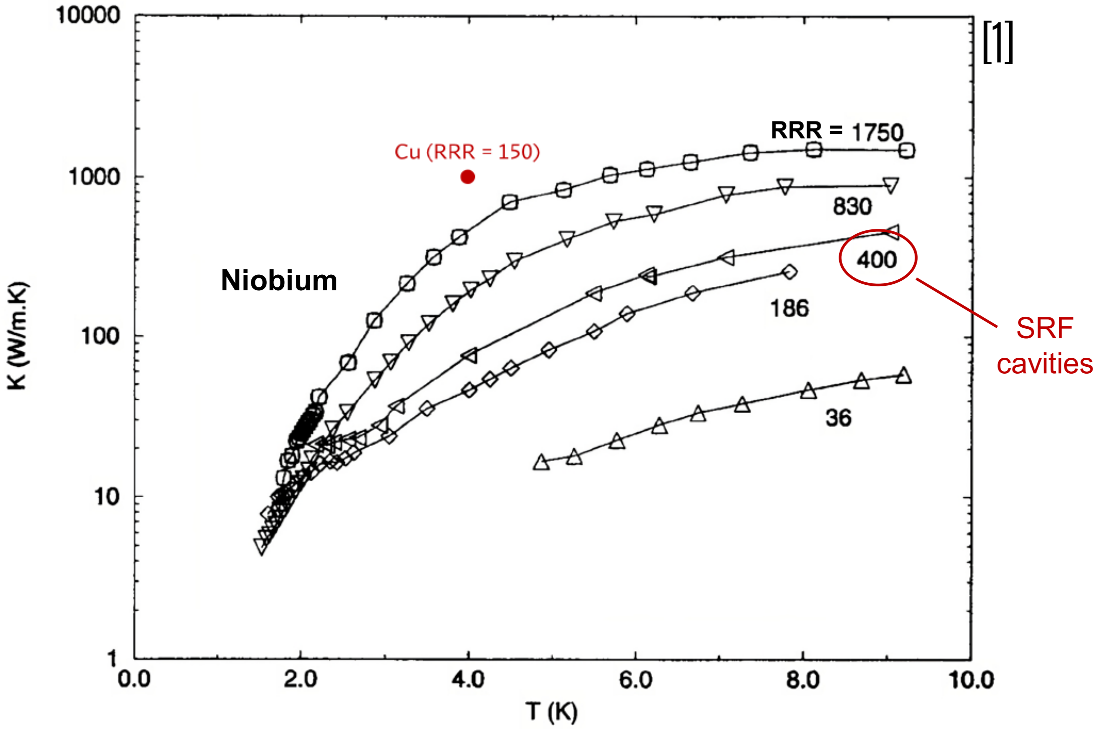
Nb_3Sn on copper: why?

DCMS Nb/Cu successful technology (LEP-II, LHC, ALPI, ISOLDE, FCC-ee)

Compared to bulk Nb:

- significant **thermal stability**
- much **cheaper** than bulk Nb

  → large scale production
- cryocooler based **conduction cooling**



[1]

[1] DOI: 10.5170/CERN-1996-003.191

Nb_3Sn on copper via DCMS: goals

Long term: scalable process to coat Nb_3Sn/Cu SRF accelerating cavities

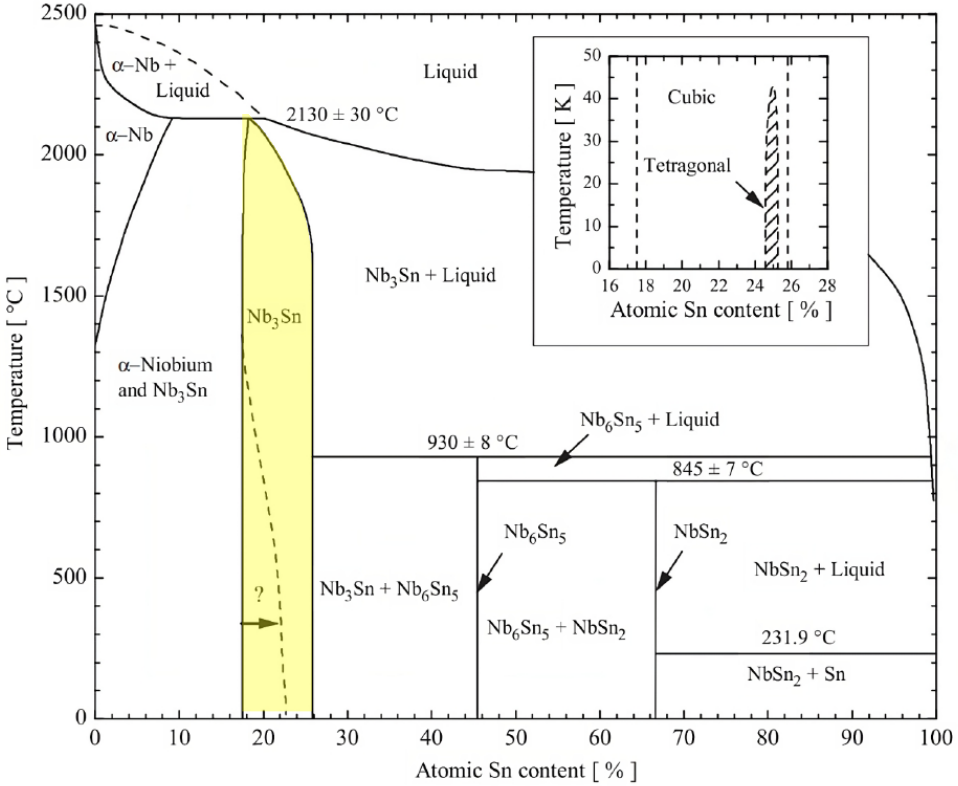
Short term: produce Nb_3Sn films on Cu (small samples) which exhibit

- bulk like T_c
  - correct stoichiometry (Nb-Sn ratio)
  - correct A15 phase
- dense and crack-free morphology

Conditions must be satisfied before further development toward SRF application

Nb_3Sn on copper: challenges

- Complicated phase diagram:
  - ~ 18 - 26 Sn At%
  - > 930 °C to form only A15 Nb-Sn
- Temperature limit = **650 °C** (6 GHz)
  - → weakening point of copper (400 °C)
- Sn and Cu are miscible
  - → intermediate buffer layer needed



[1]

[1] A Godeke 2006 *Supercond. Sci. Technol.* 19 R68

Standard sample production process

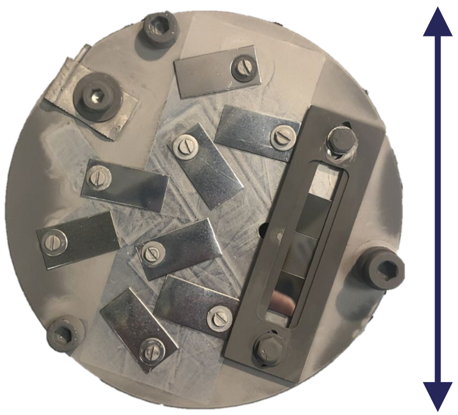
(1) Set parameters:

Cathode power P

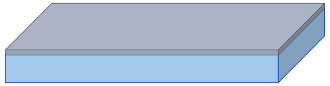
Deposition pressure p_{dep}

Deposition temperature T_{dep}

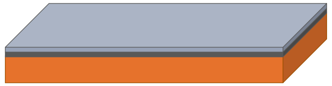
small samples
on holder plate



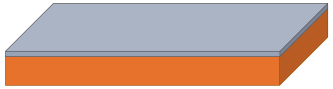
(2) 1 μm Nb_3Sn



Sapphire



Cu + **1 μm** Nb buffer layer (BL)



Cu

+ 24 h annealing

(3) Characterisation:

T_c measurement

SEM + EDS (Sn composition)

XRD

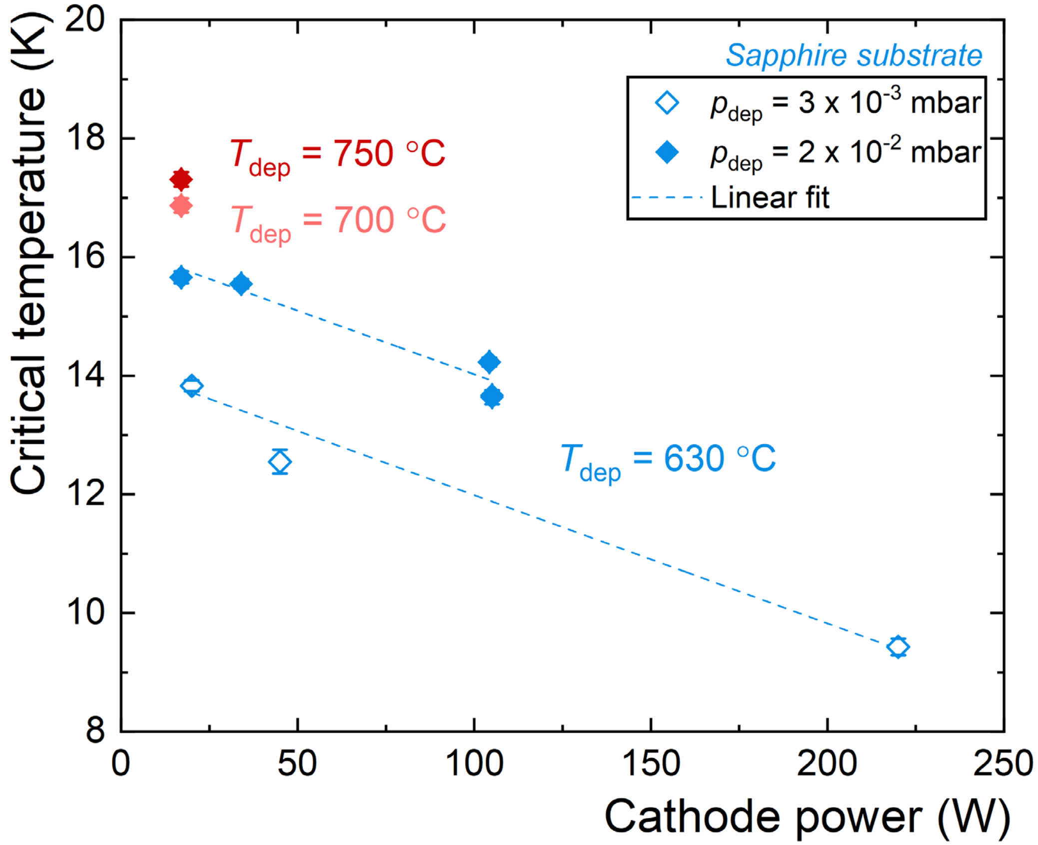
Tune next parameters based on trends (step 1)

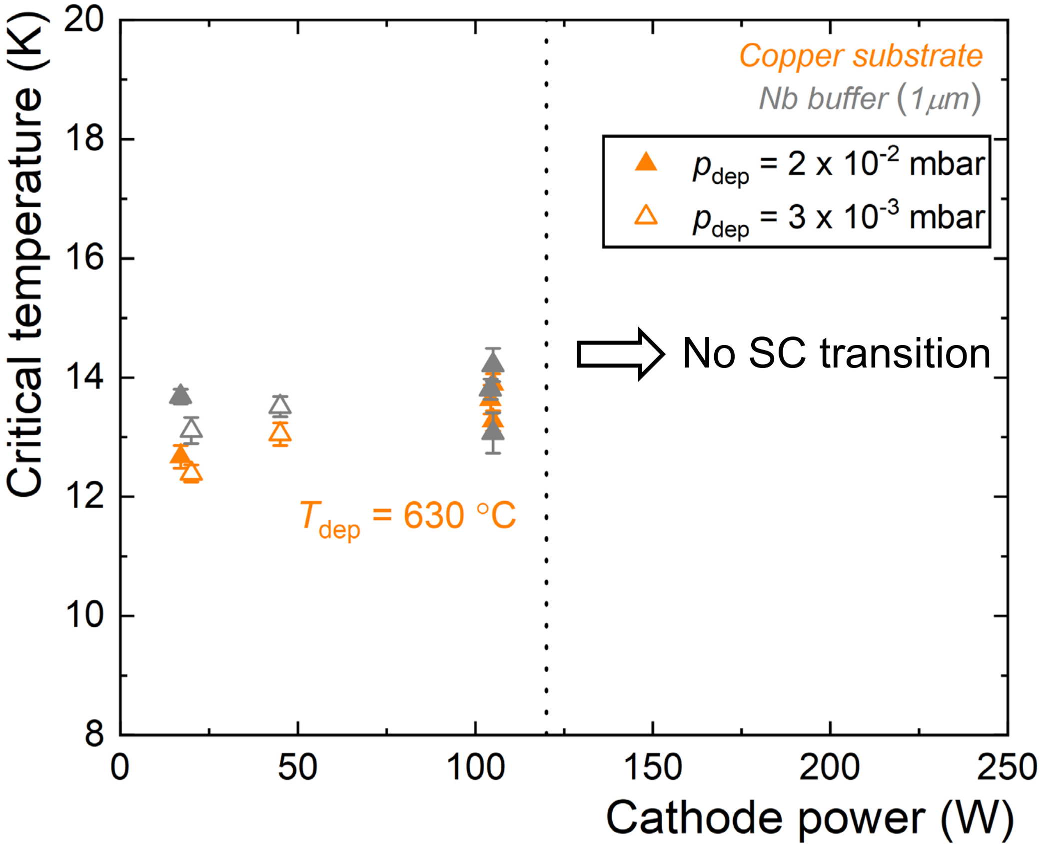
dorothea.fonnesu@lnl.infn.it

Nb_3Sn on copper: first trends

Sapphire as reference

#1 - T_c-oriented study of dependencies from sputtering parameters



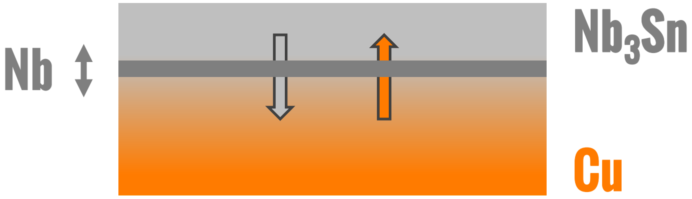


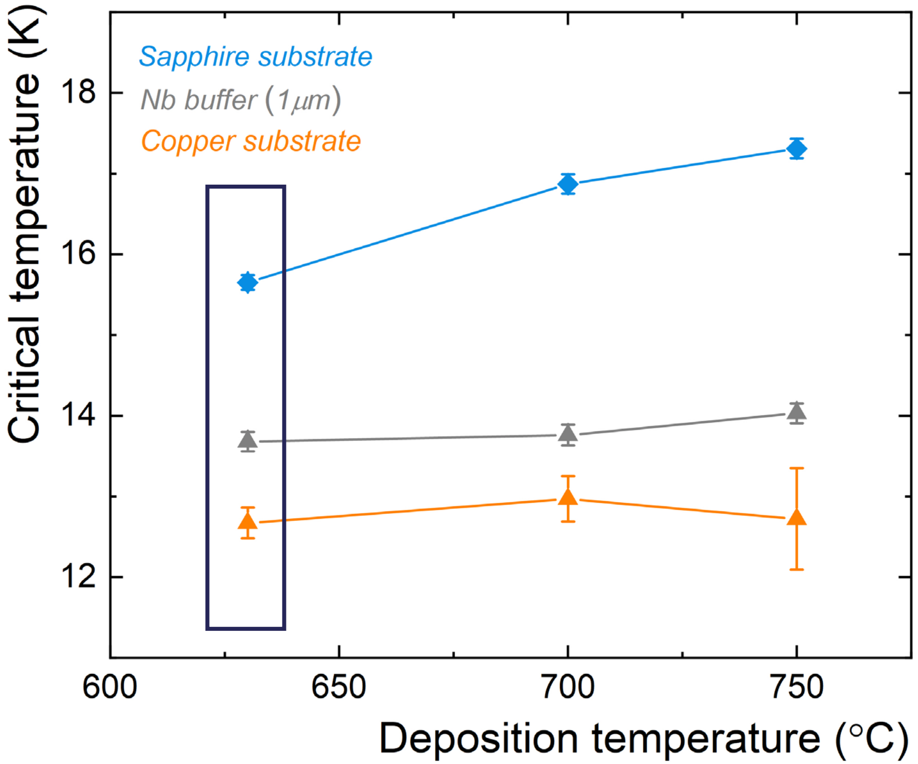
dorothea.fonnesu@lnl.infn.it

Nb_3Sn on copper: where to look

#2 – Fix cathode power and deposition pressure, keep temperature low

- Samples with best T_c on sapphire
- stick to lowest T_{dep} (≤ 630 °C)
- Investigate role of:
    1. annealing time → decrease
    2. buffer layer thickness → increase

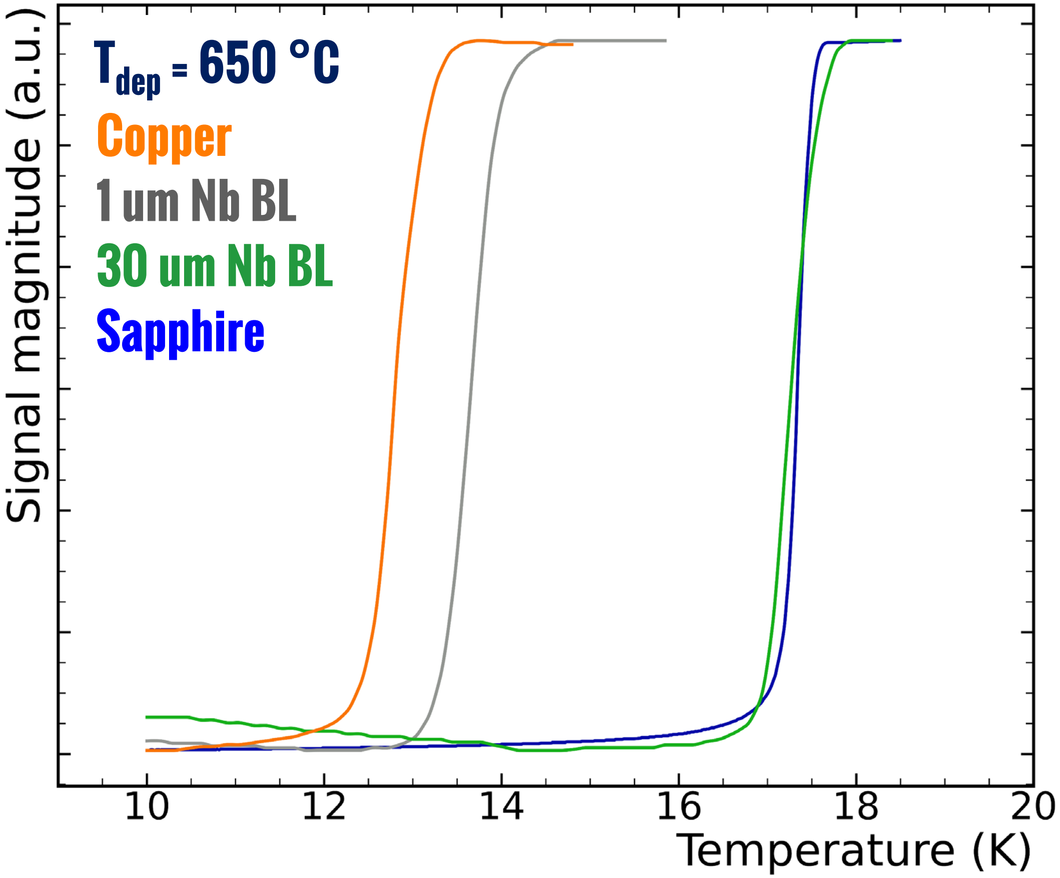




dorothea.fonnesu@lnl.infn.it

Nb_3Sn on copper: SC transition curves

Recipe: 1 um Nb_3Sn T_{dep} = 650 °C $p_{dep} = 2 \times 10^{-2}$ mbar $P_{cathode}$ = 15 W **No annealing**



Buffer layer thickness

dorothea.fonnesu@lnl.infn.it

Nb_3Sn on copper: where we are

- shorter t_{ann} has no effect on Cu and Cu+Nb 1 um BL
- shorter t_{ann} has positive effect on sapphire
- thicker Nb BL (1 μm → 30 - 50 μm) comparable to sapphire

Role of buffer layer?

Sn-Cu interdiffusion

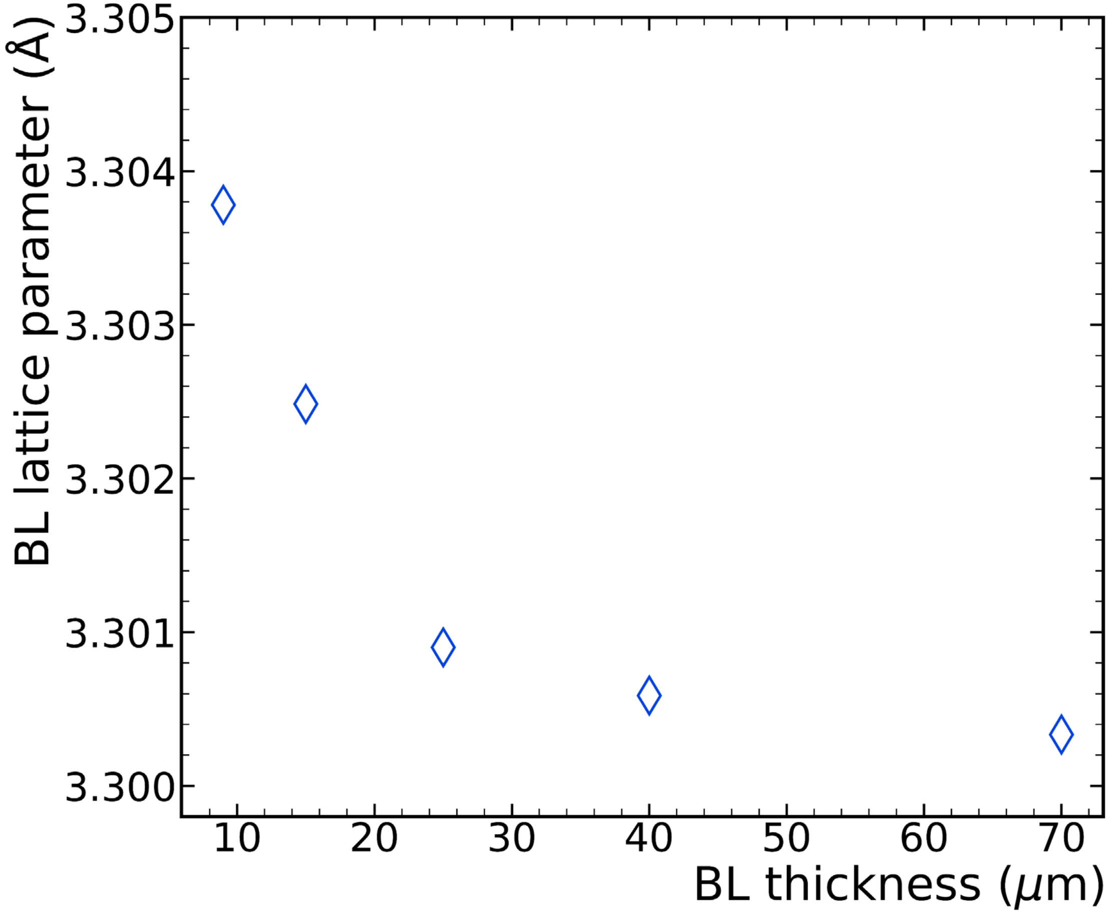
Film accommodation

Temperature limit

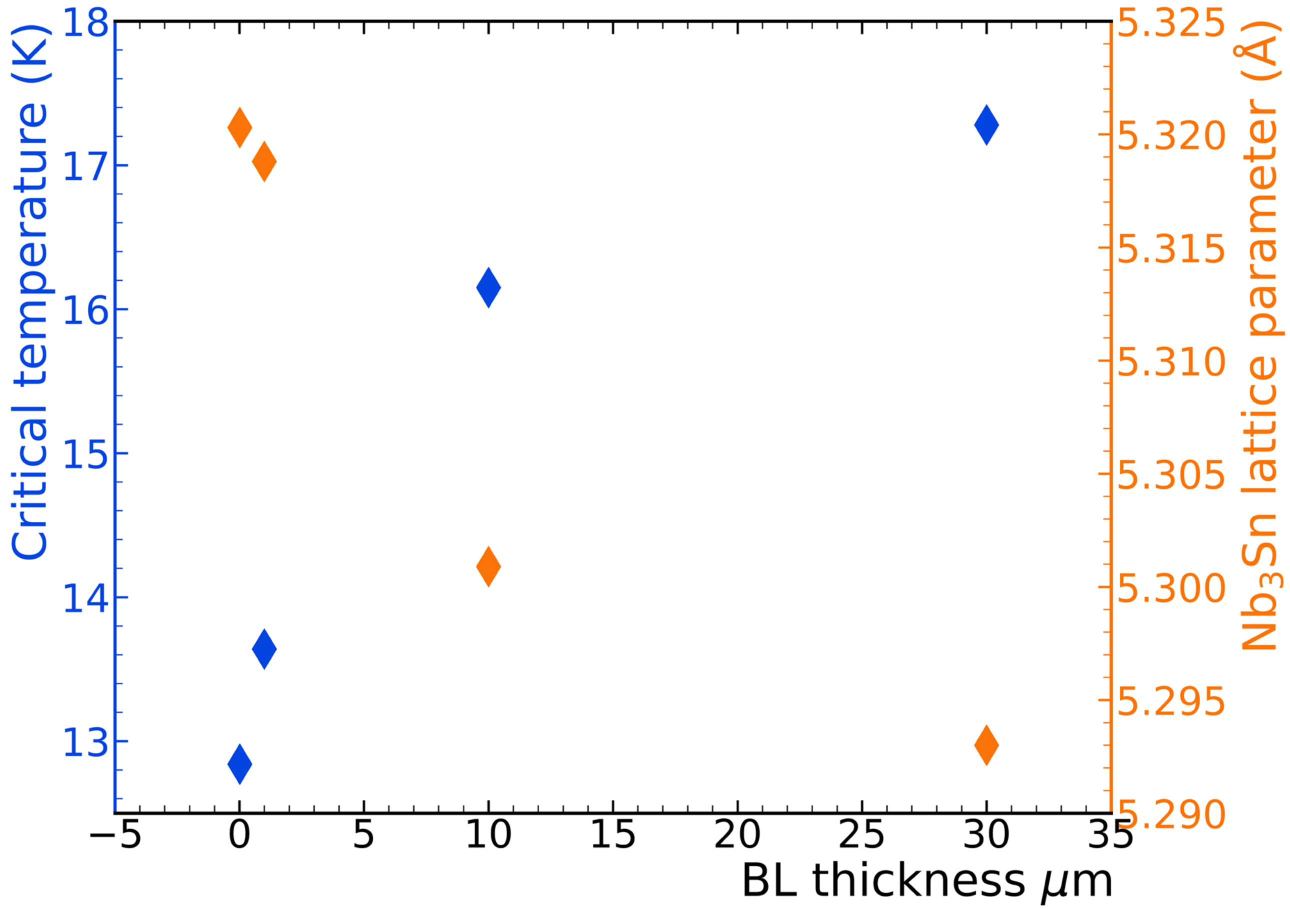
T_c = 17.33 ± 0.25 K on Cu+50 um BL at T_{dep} = 600 °C

Accomodation effects

Nb buffer layer

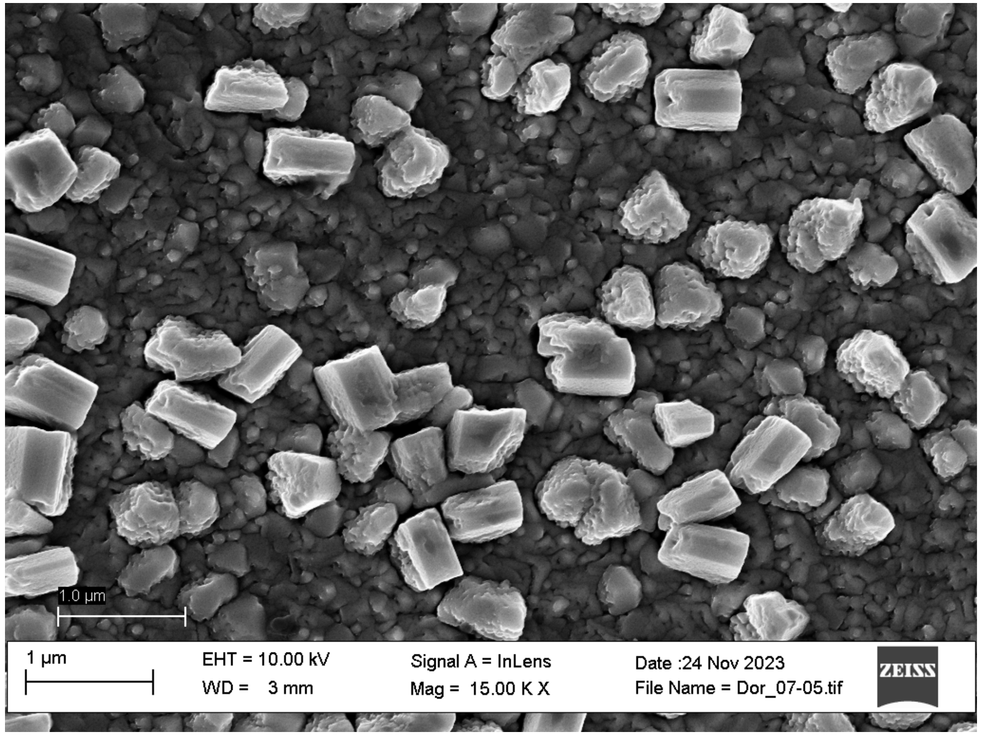


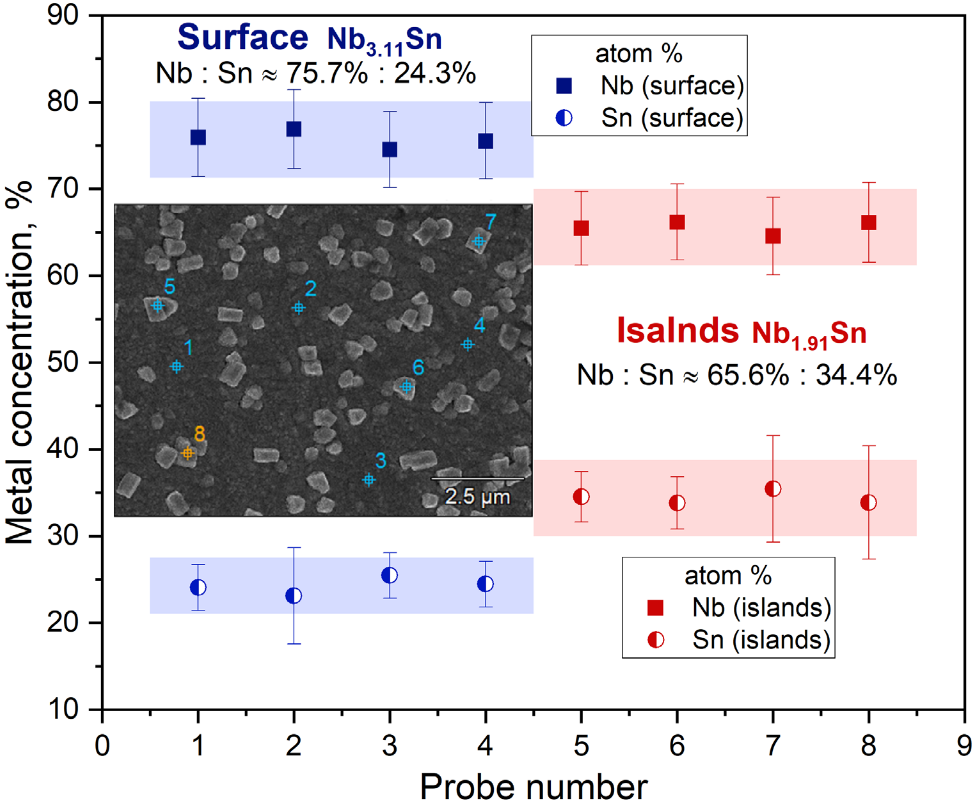
Nb3Sn/Nb/Cu 650 °C



dorothea.fonnesu@lnl.infn.it

Morphology and composition

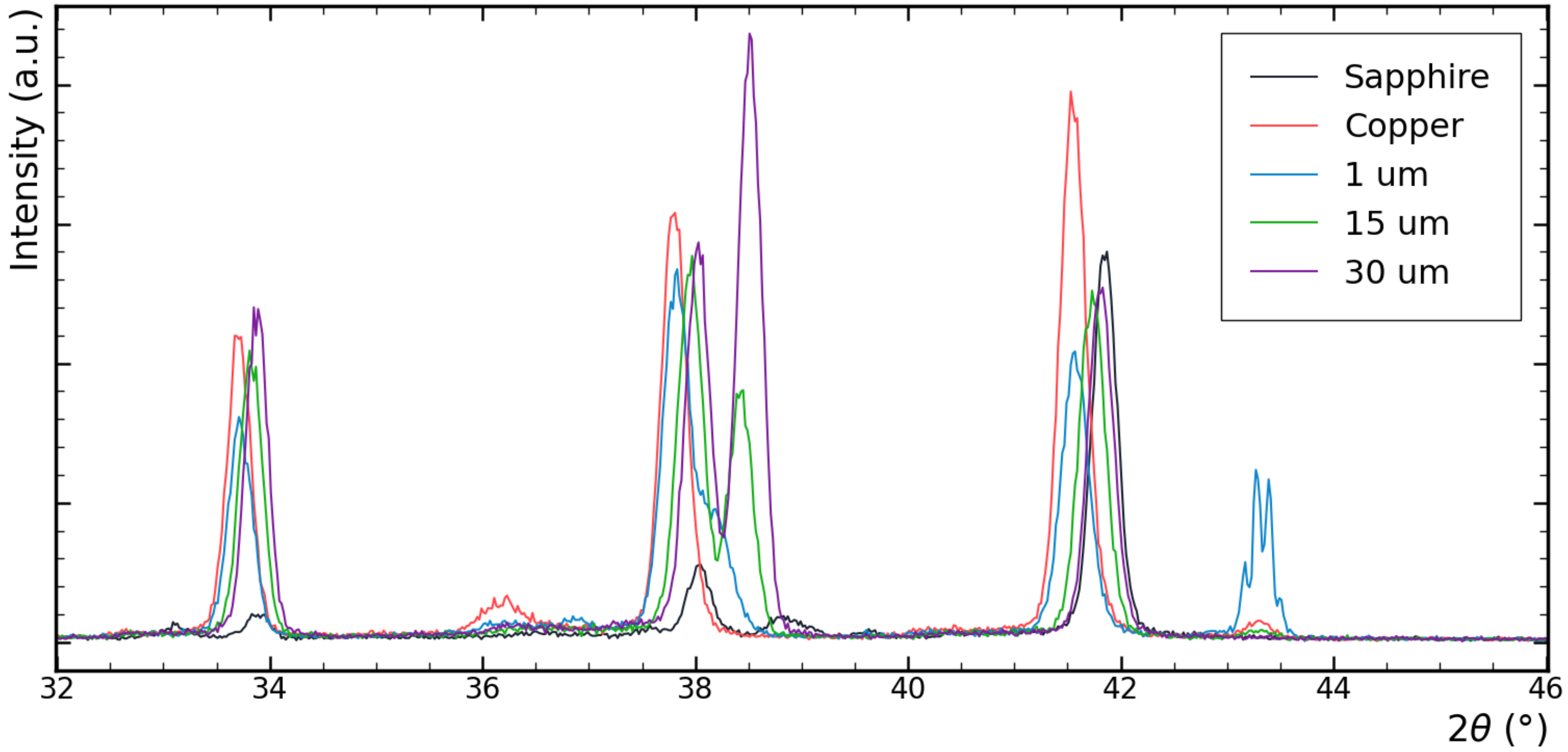




XRD patterns

Nb3Sn peaks								
(002)	**(012)**	(112)	(023)	(123)	(004)	(024)	(124)	(233)
33.870	**38.012**	41.801	63.352	66.041	71.263	81.284	83.736	86.176

Nb Im-3m peaks				Cu peaks			
(011)	(002)	(112)	(022)	**(111)**	(002)	(022)	(113)
38.575	55.696	69.796	82.695	**43.342**	50.479	74.174	89.622



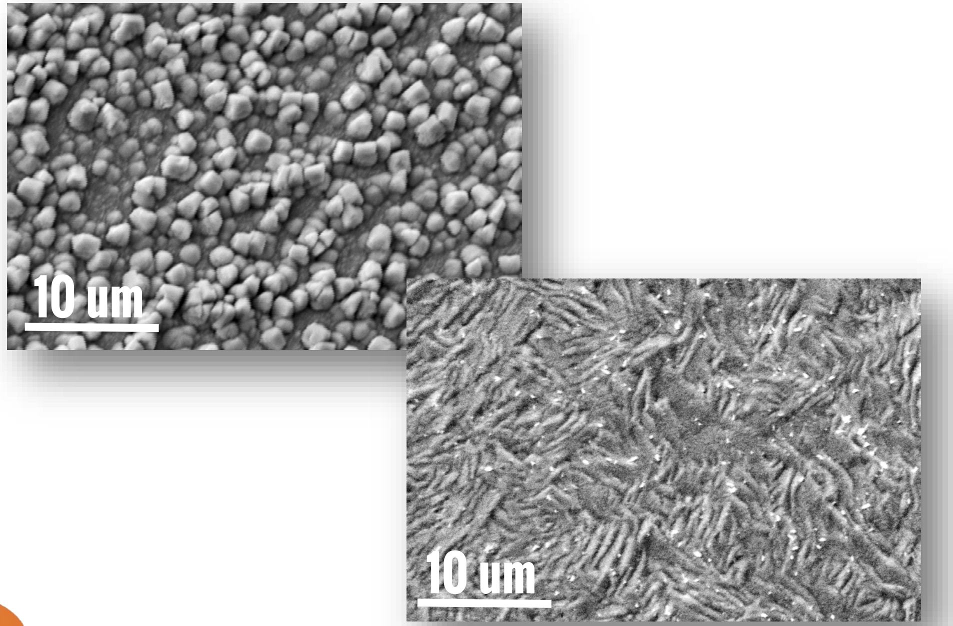
Nb_3Sn on copper: outlook

ONGOING (on our side):

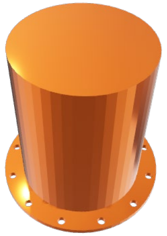
- T_{dep} = 500 °C → find lower limit
- buffer layer thickness dependencies
- EDS composition analysis
- XRD analysis

ONGOING (within I.FAST):

- SQUID magnetometry
- SEM surface and FIB cross-section imaging



Toward QPR sample for RF test



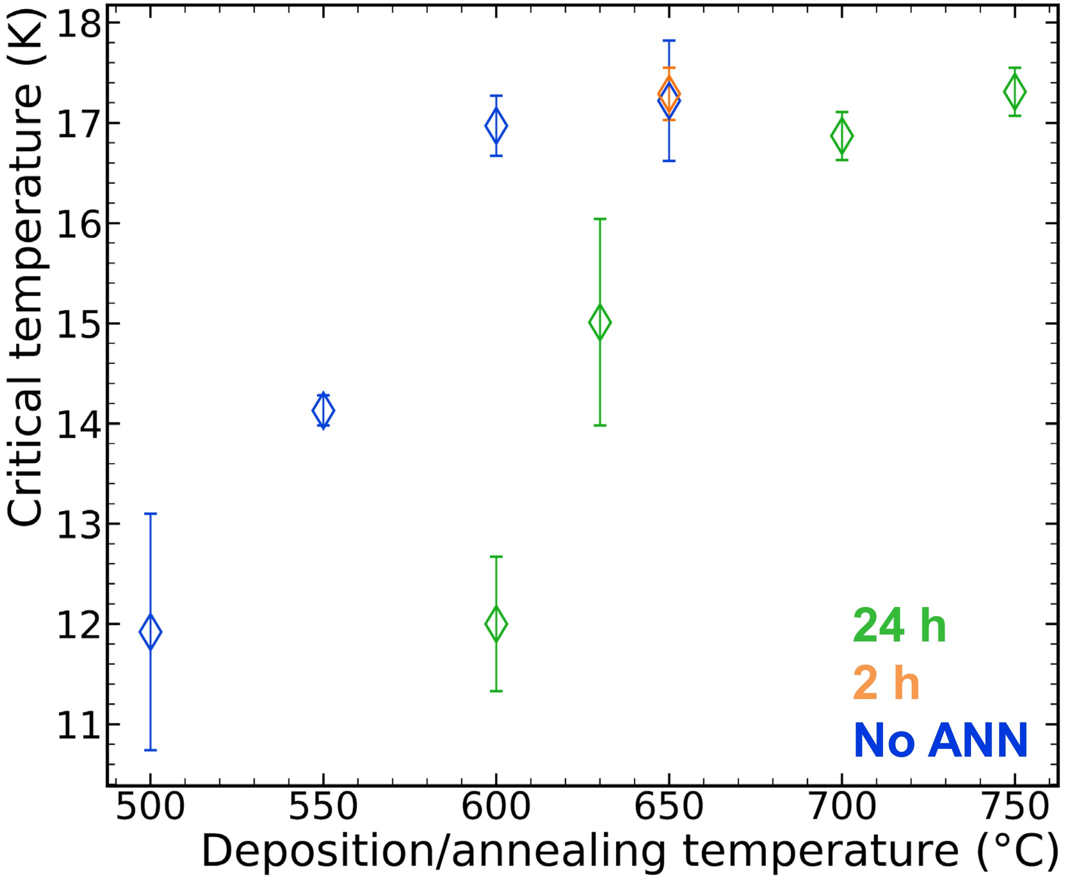
dorothea.fonnesu@lnl.infn.it

Thank you!

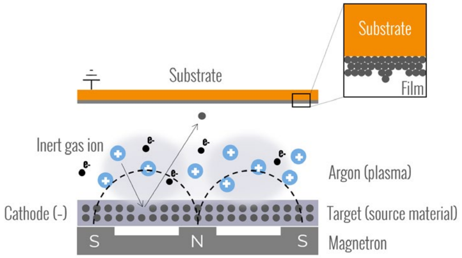

Giovanni Marconato
Davide Ford
Eduard Chyhyrynets
Cristian Pira
Dorothea Fonnesu
Roberta Caforio
Alessandro Salmaso

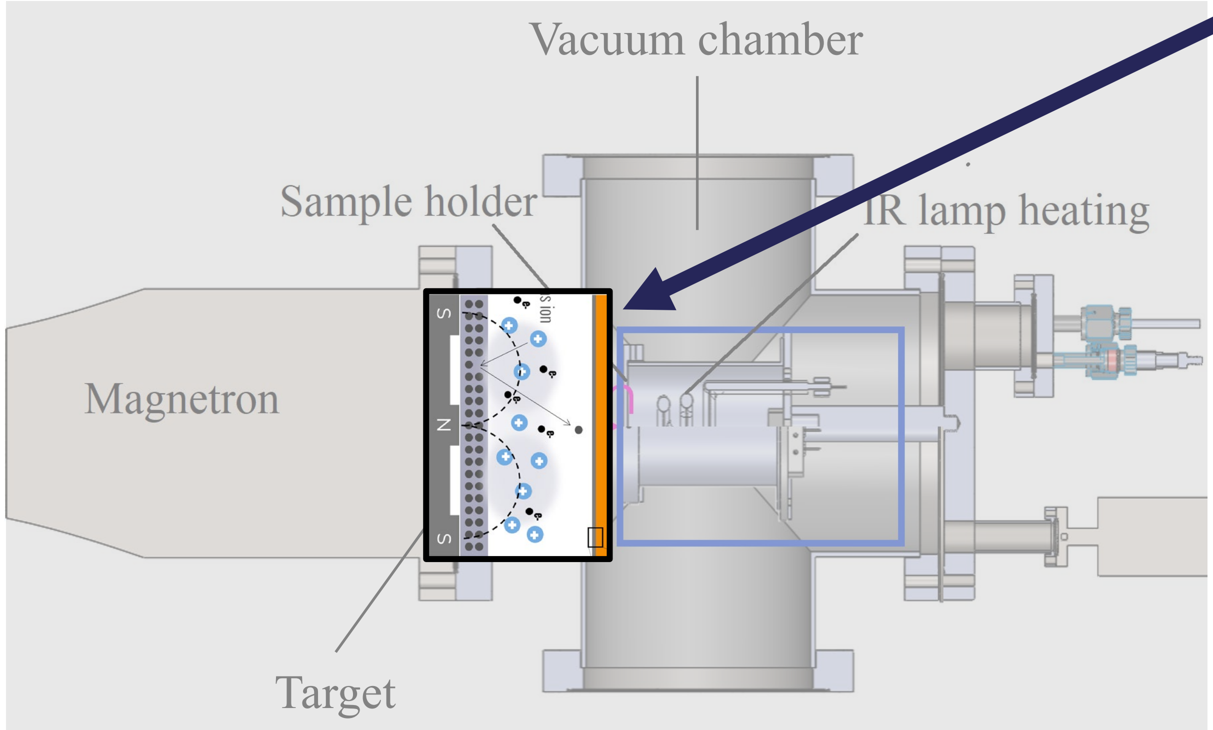
Backup

ANNEALING TIME
Sapphire substrate



Sputtering chamber



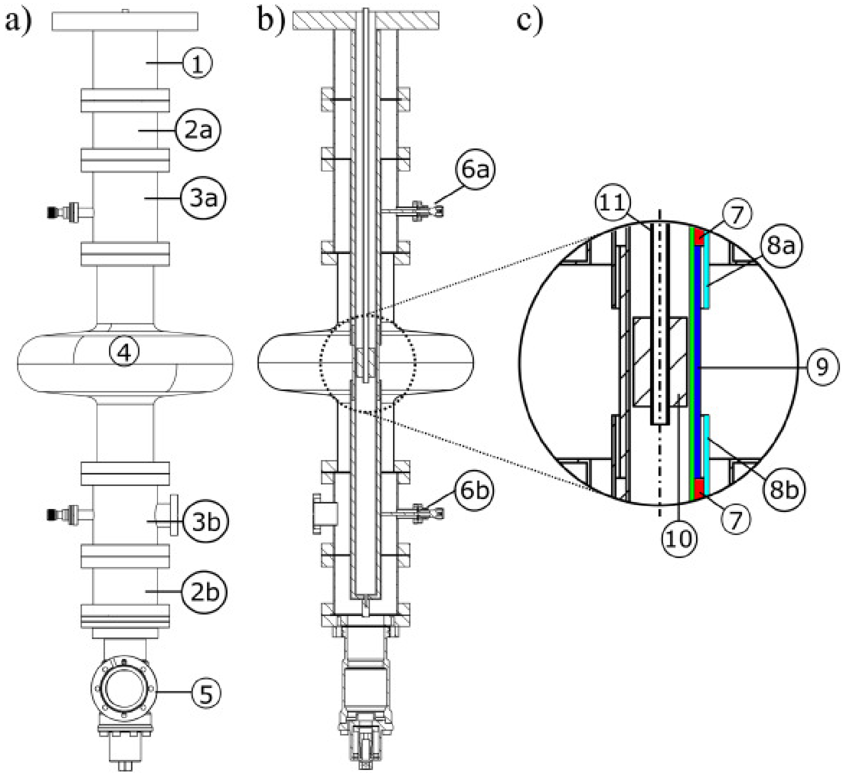


side view

- Planar DCMS on small flat samples
- Single Nb_3Sn stoichiometric target
- Argon atmosphere
- T substrate regulated via IR lamps
- Film thickness 1 μm

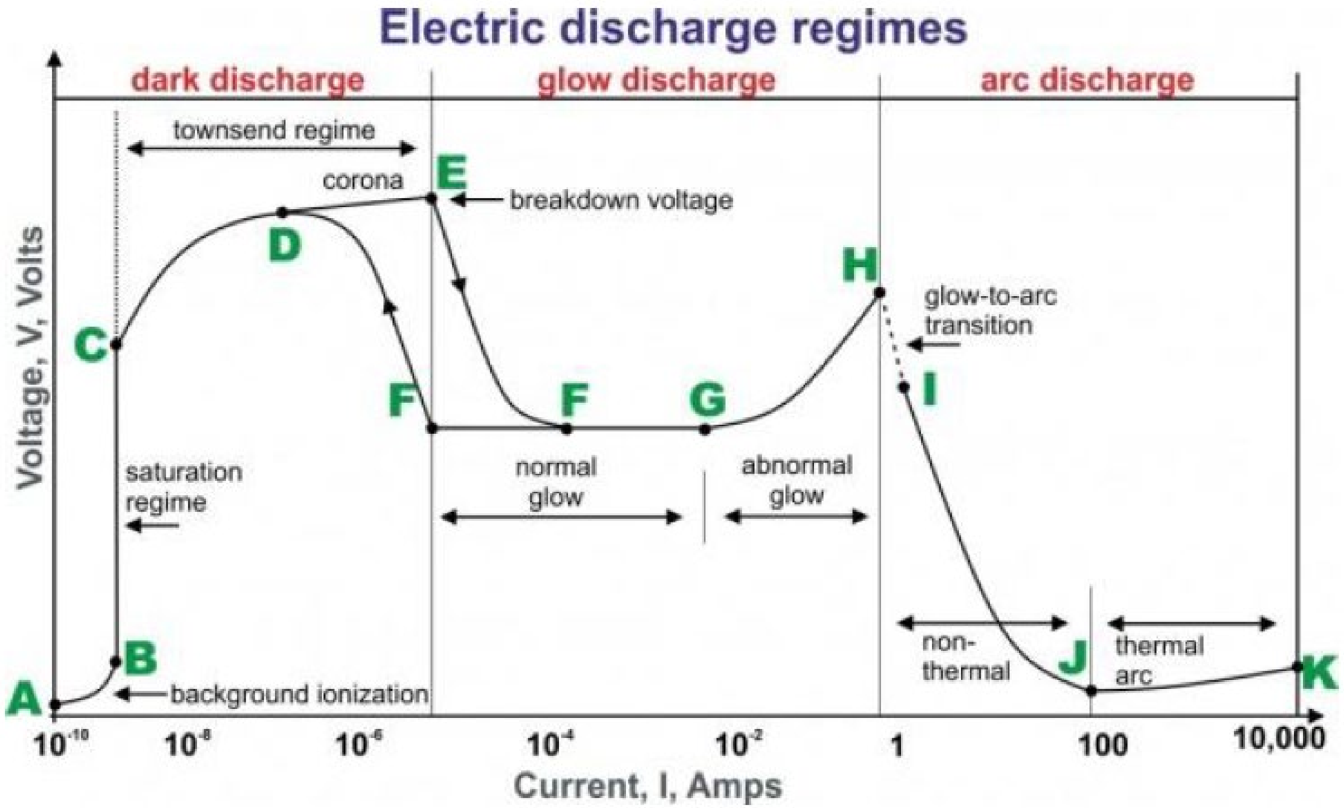
dorothea.fonnesu@lnl.infn.it

DCMS cylindrical configuration



dorothea.fonnesu@lnl.infn.it

Plasma

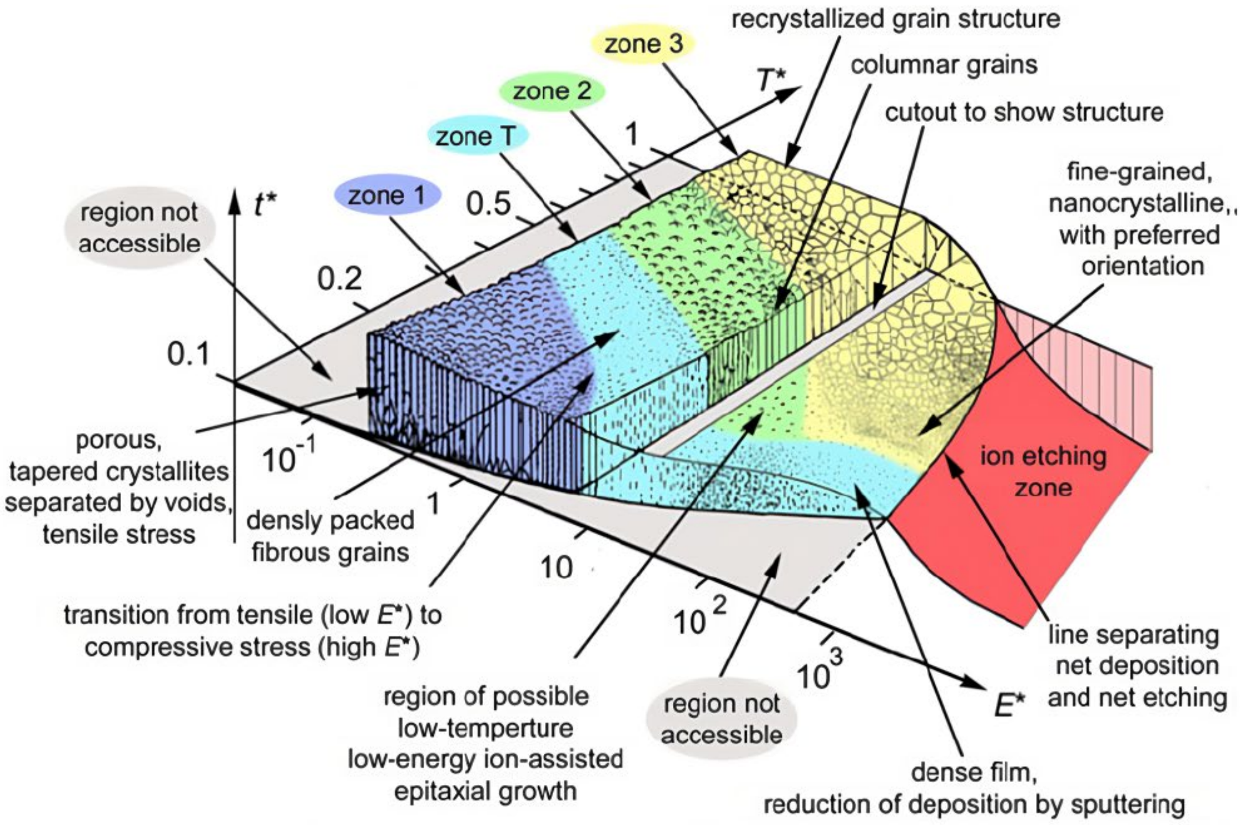

Electric discharge regimes
dark discharge
glow discharge
arc discharge
townsend regime
corona
E
breakdown voltage
D
H
glow-to-arc
transition
C
F
F
G
I
saturation
regime
normal
glow
abnormal
glow
non-
thermal
J
thermal
arc
B
A
background ionization
K
Voltage, V, Volts
10⁻¹⁰
10⁻⁸
10⁻⁶
10⁻⁴
10⁻²
1
100
10,000
Current, I, Amps

Sputtering

$$E_{\mathrm{th}} = E_{\mathrm{b}} \begin{cases} \frac{1+5.7(M_{\mathrm{i}}/M_{\mathrm{a}})}{\Lambda} & \text{for } M_{\mathrm{i}} \leq M_{\mathrm{a}} \\ \frac{6.7}{\Lambda} & \text{for } M_{\mathrm{i}} > M_{\mathrm{a}} \end{cases} \qquad \Lambda = \frac{4M_{\mathrm{i}}M_{\mathrm{a}}}{(M_{\mathrm{i}}+M_{\mathrm{a}}^2)}$$

$$Y(E)\mathrm{d}E \propto \frac{1-\sqrt{(E_{\mathrm{b}}+E)/\Lambda E_0}}{E^2(1+E_{\mathrm{b}}/E)^3}\mathrm{d}E$$

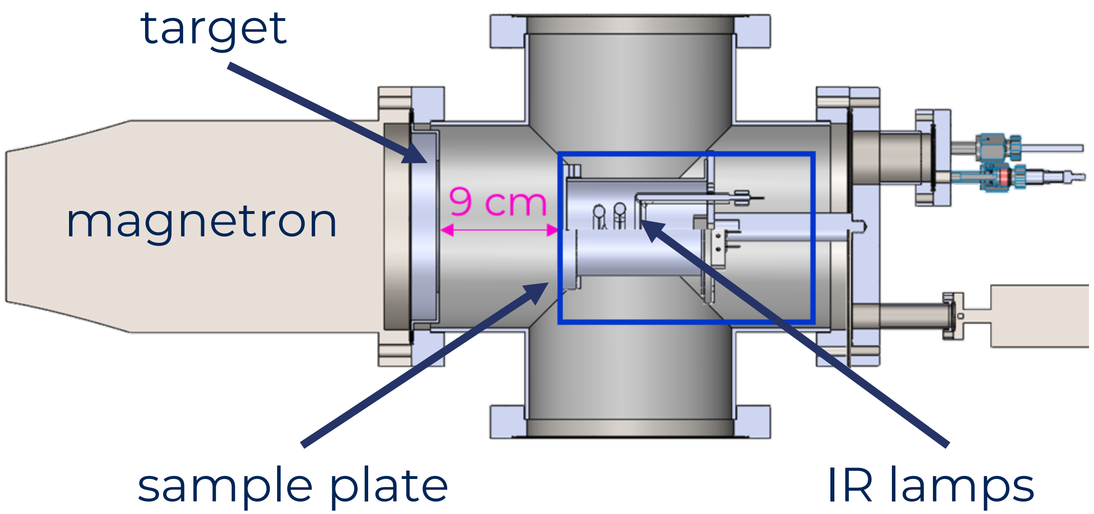
Structure zone diagram

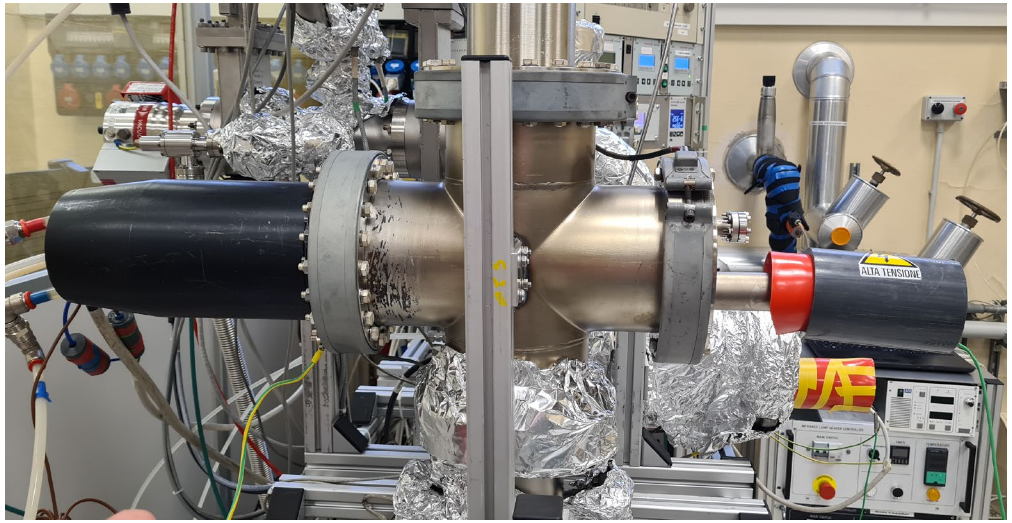


dorothea.fonnesu@lnl.infn.it

Nb_3Sn Experimental Set-up

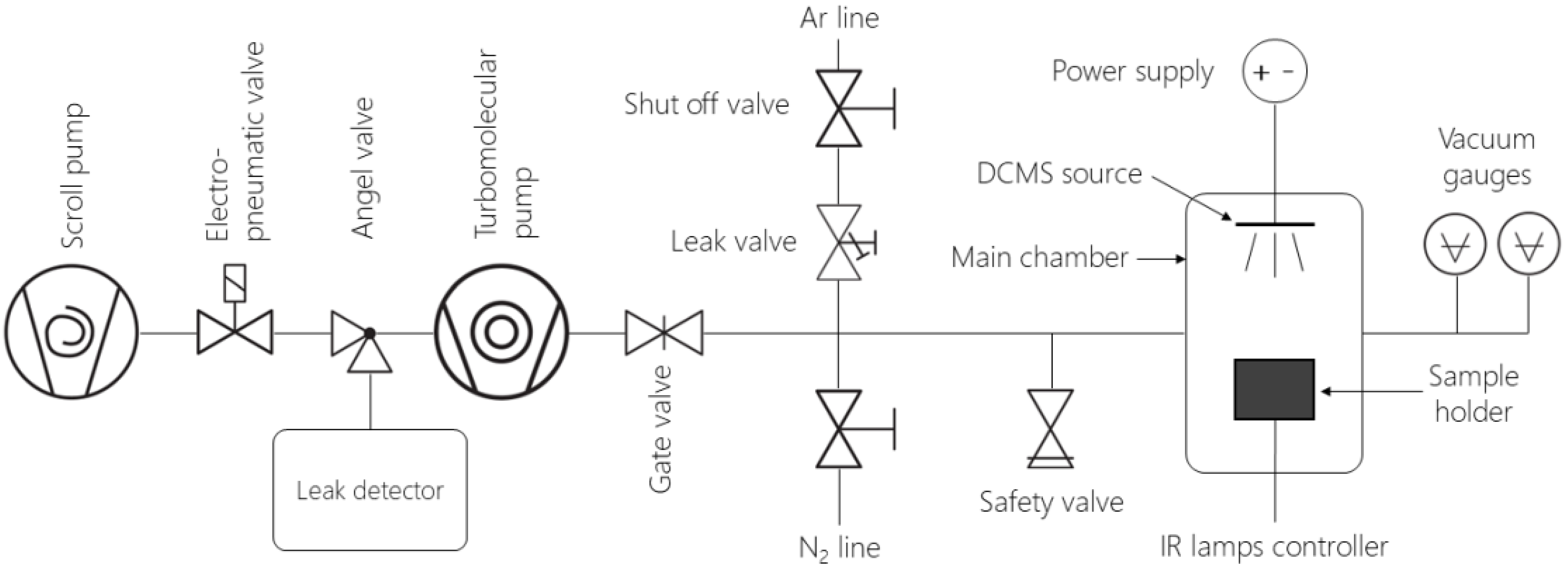
- Commercial Nb_3Sn stoichiometric planar target (4'' diameter)
- System base pressure: 5 x 10^{-9} mbar
- Heated sample holder: up to 950 °C via IR lamps





dorothea.fonnesu@lnl.infn.it

Nb_3Sn Experimental Set-up (PID)



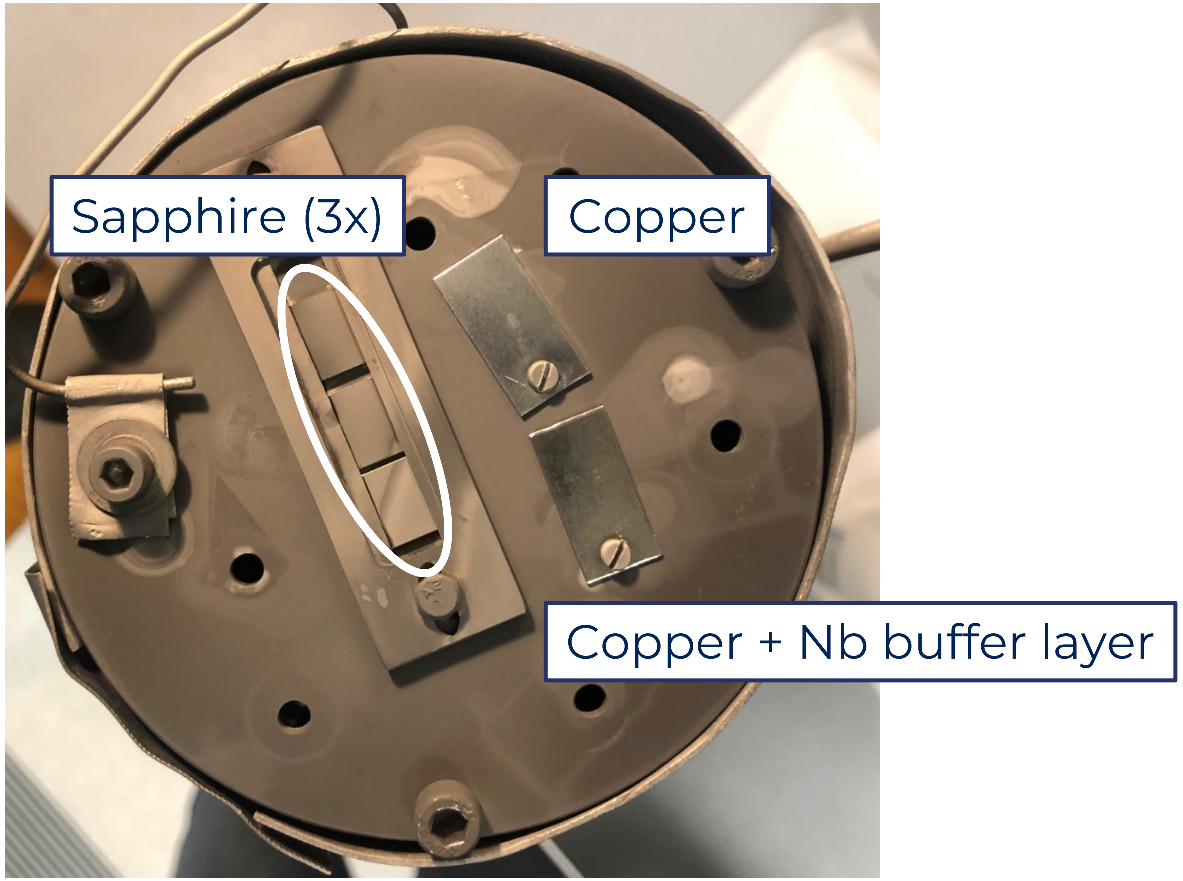
dorothea.fonnesu@lnl.infn.it

Experimental Set-up

Standardized procedure (1)

Fixed number and type of substrates

- 5 substrates per run:
  - 3x sapphire
  - 1x copper
  - 1x copper + Nb buffer layer

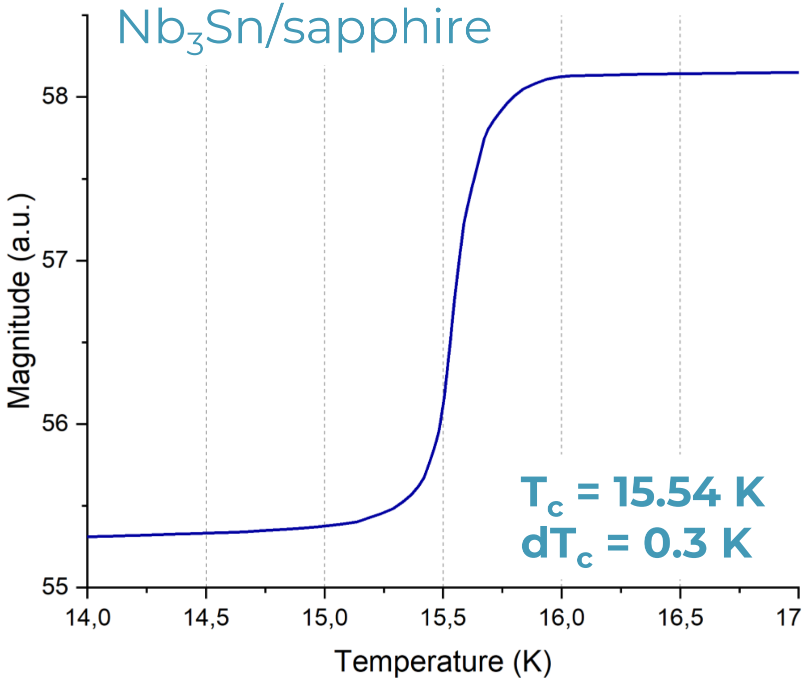


dorothea.fonnesu@lnl.infn.it

Sample characterization

Standardized procedure (2): fixed measurement routine and T_c extraction method

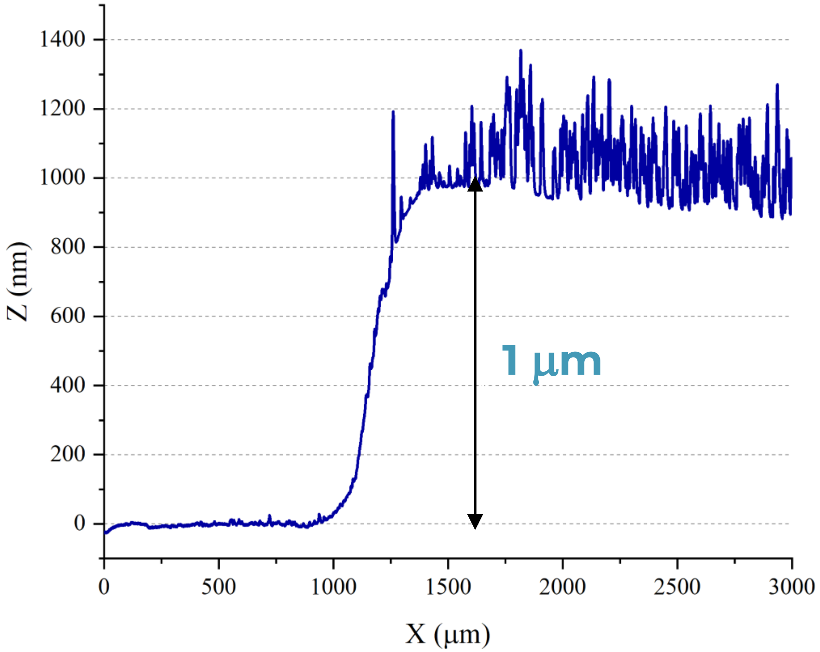
T_c by induction coil



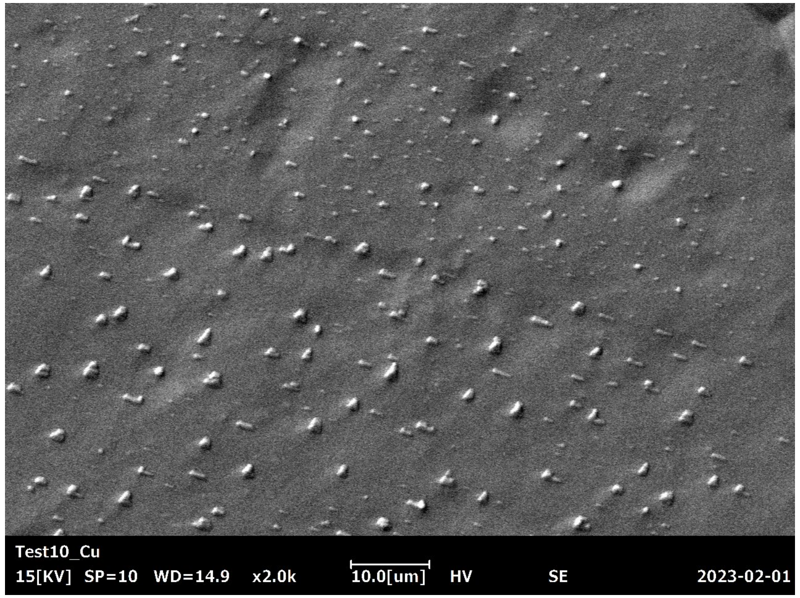
T_c = 15.54 K

dT_c = 0.3 K

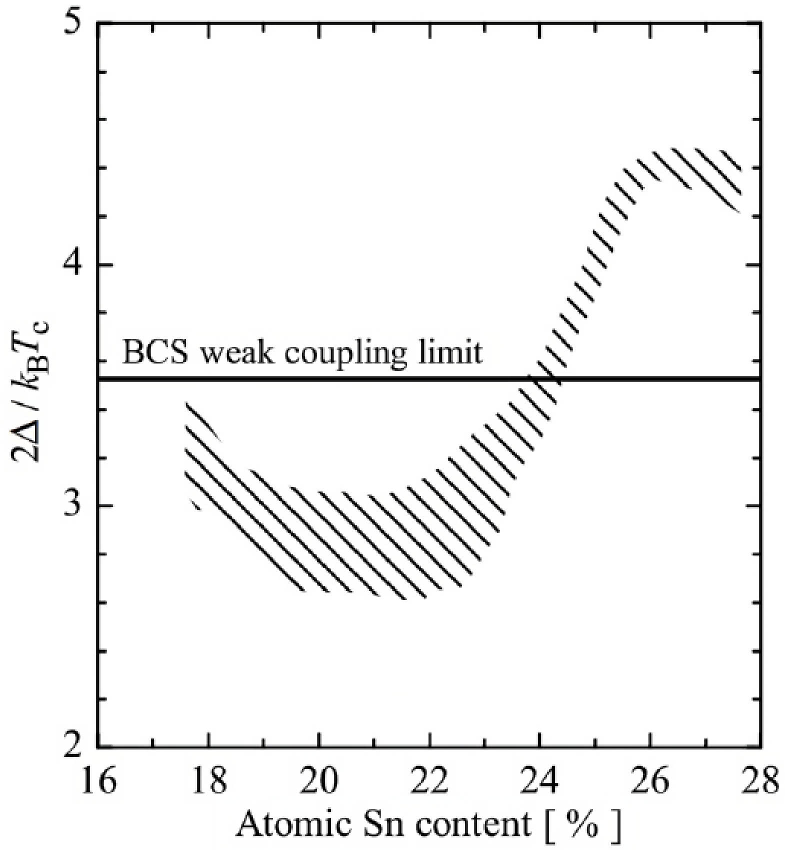
Thickness by profilometer and SEM cross-section

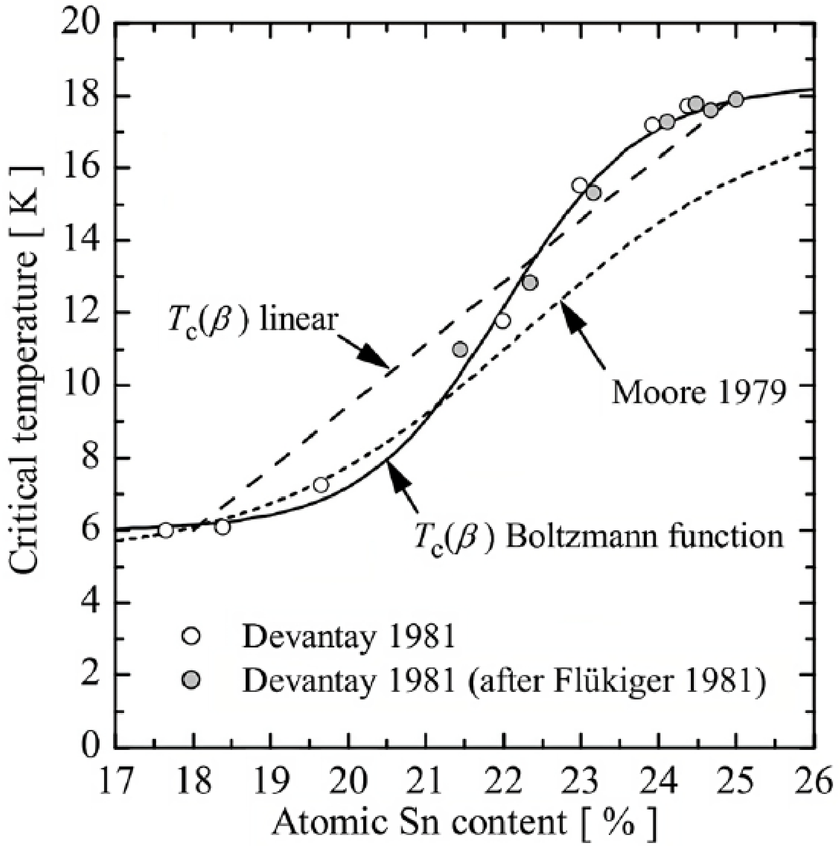


Morphology and composition by SEM + EDS



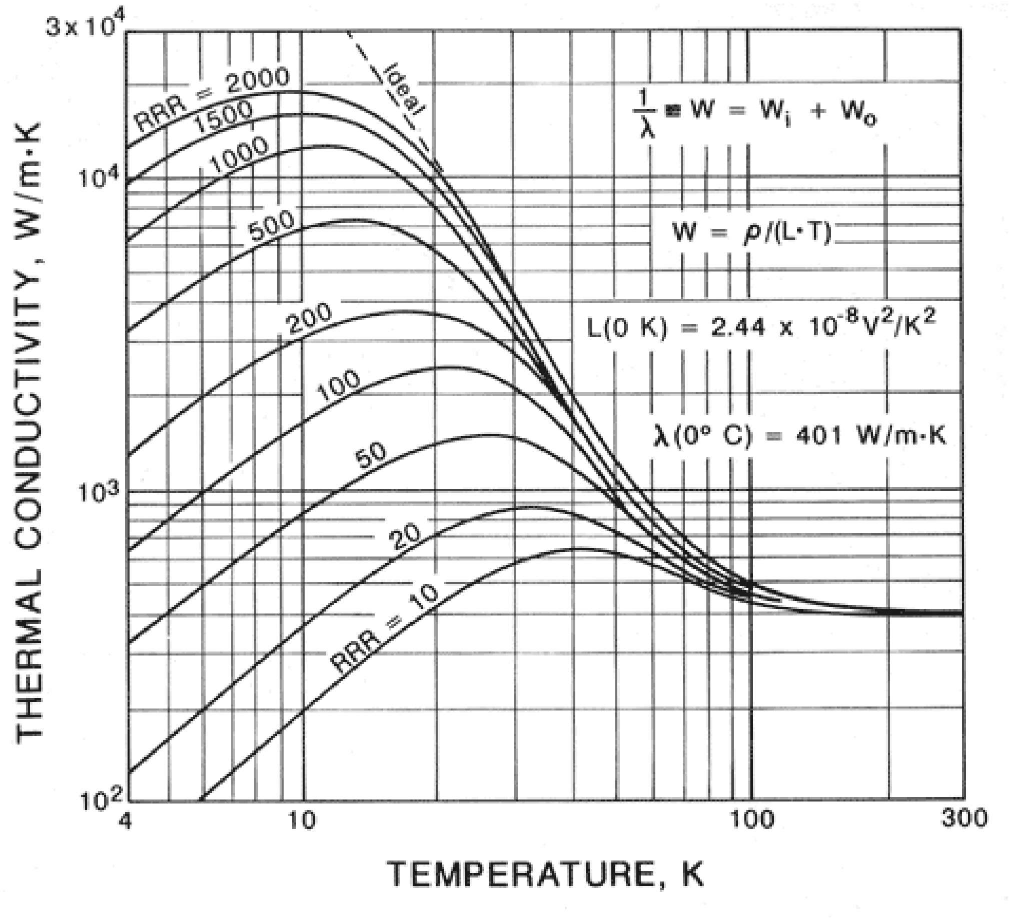
Nb_3Sn coupling





dorothea.fonnesu@lnl.infn.it

Copper



3 x 10^4
10^4
10^3
10^2
THERMAL CONDUCTIVITY, W/m·K
4
10
100
300
TEMPERATURE, K
RRR = 2000
1500
1000
500
200
100
50
20
RRR = 10
Ideal
1/λ = W = W_i + W_o
W = ρ/(L·T)
L(0 K) = 2.44 x 10^-8 V^2/K^2
λ(0° C) = 401 W/m·K

Copper vs. Nb costs

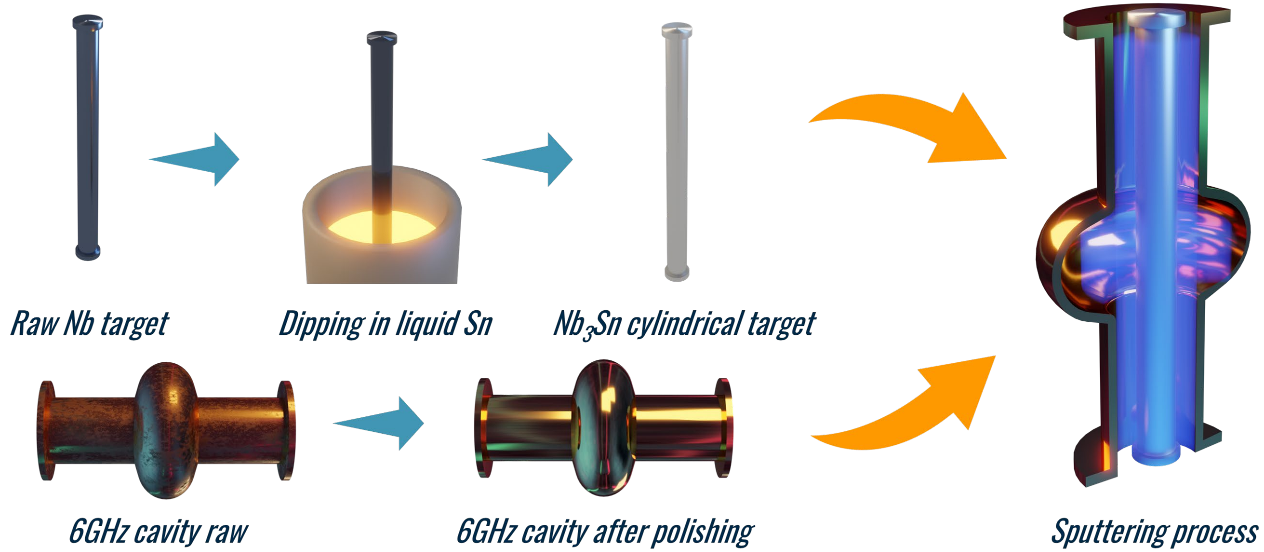
Type I Nb ~ 420 $/Kg (year 2014)

Cu ~ 6 $/kg (year 2014)

~ factor 70 difference

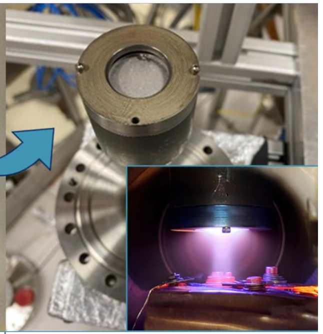
dorothea.fonnesu@lnl.infn.it

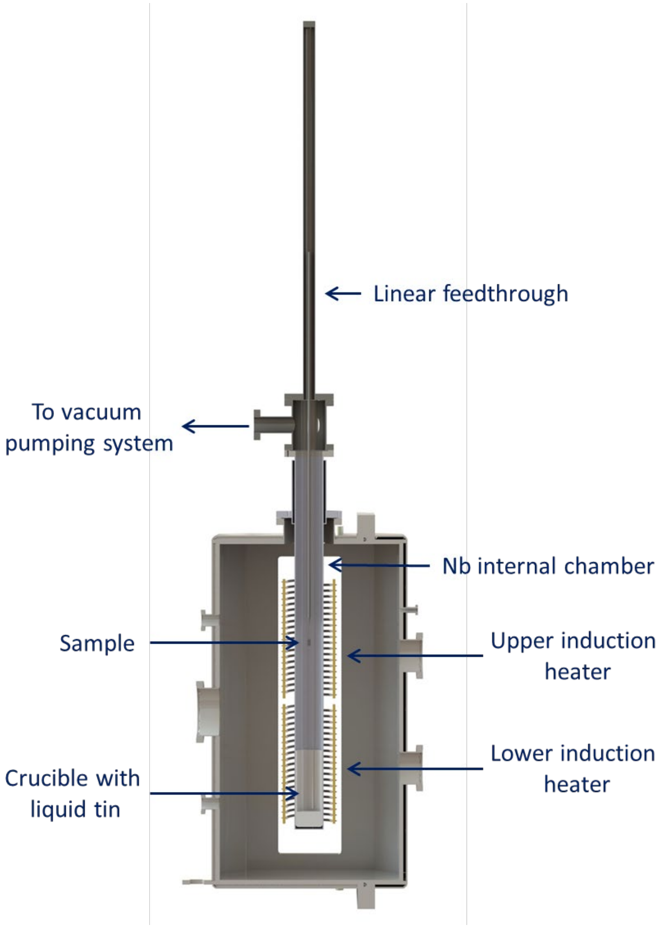
Dipping (Nb_3Sn target production via LTD)



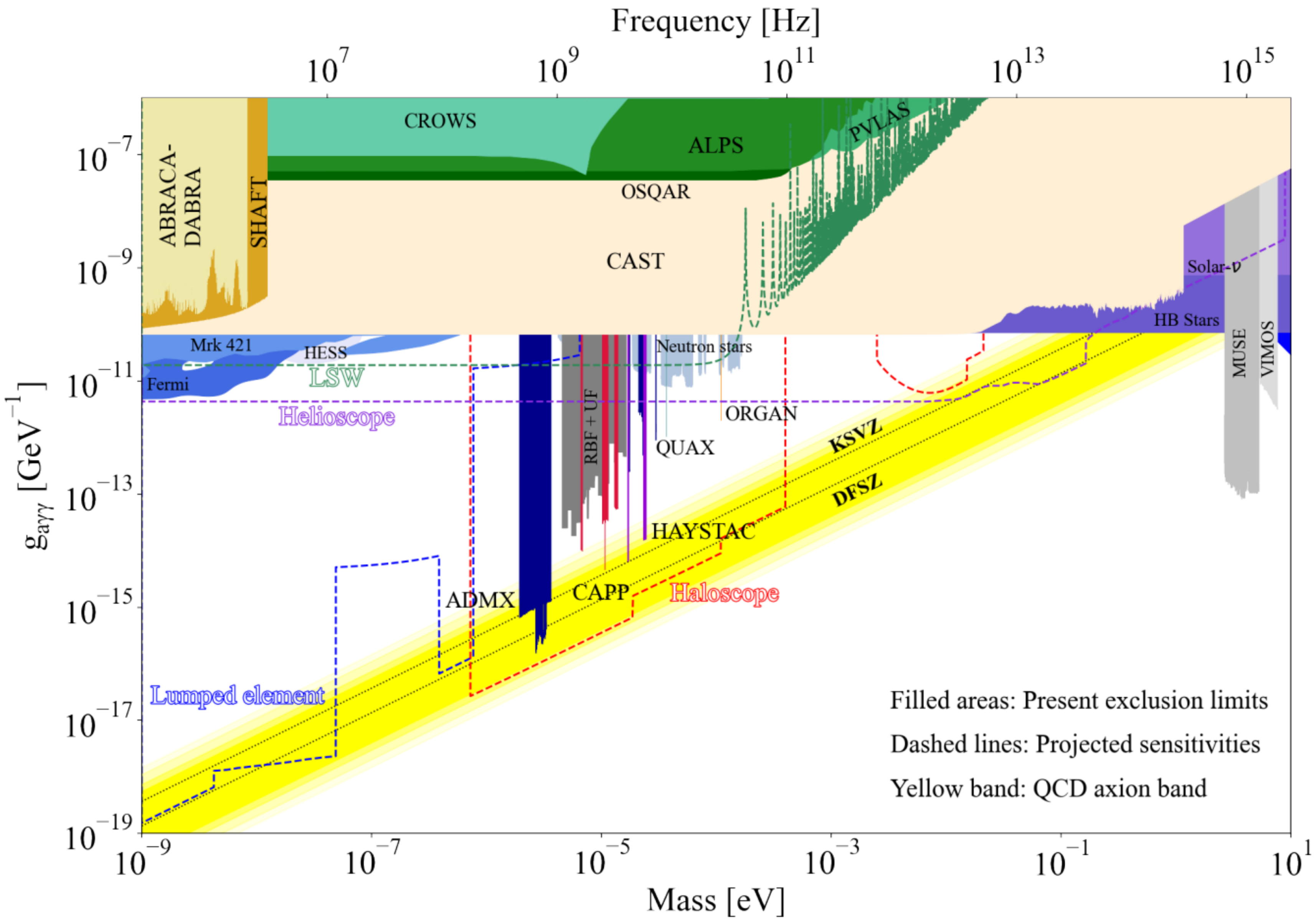

1) Anodized Nb samples


2) Nb_3Sn coated samples


3) 1’’ magnetron sputtering



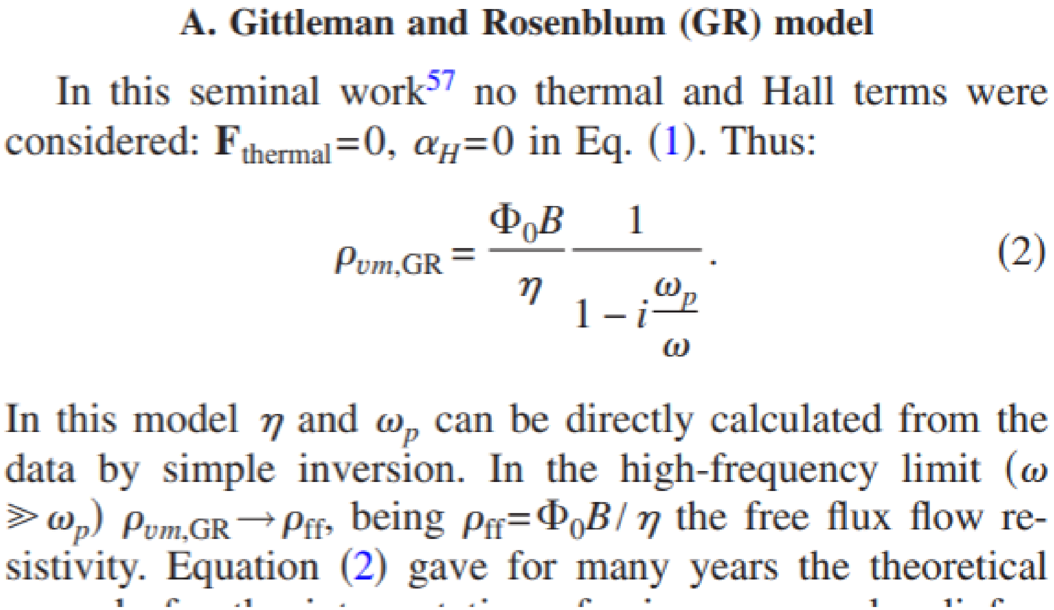
dorothea.fonnesu@lnl.infn.it


Frequency [Hz]
10^7
10^9
10^{11}
10^{13}
10^{15}
10^{-7}
10^{-9}
10^{-11}
10^{-13}
10^{-15}
10^{-17}
10^{-19}
$g_{a\gamma\gamma}$ [GeV^{-1}]
ABRACA-DABRA
SHAFT
CROWS
ALPS
OSQAR
PVLAS
CAST
Solar-ν
HB Stars
MUSE
VIMOS
Mrk 421
HESS
Fermi
LSW
Helioscope
Neutron stars
RBF + UF
ORGAN
QUAX
KSVZ
DFSZ
HAYSTAC
ADMX
CAPP
Haloscope
Lumped element
Filled areas: Present exclusion limits
Dashed lines: Projected sensitivities
Yellow band: QCD axion band
10^{-9}
10^{-7}
10^{-5}
10^{-3}
10^{-1}
10^{1}
Mass [eV]

Fluxons

$$\eta = \frac{\phi_o B_{c2}}{\rho_n}$$

$$k = \frac{2\pi J_c \phi_0}{d}$$



A. Gittleman and Rosenblum (GR) model

In this seminal work[57] no thermal and Hall terms were considered: $\mathbf{F}_{\text{thermal}}=0$, $\alpha_H=0$ in Eq. (1). Thus:

$$\rho_{vm,\mathrm{GR}} = \frac{\Phi_0 B}{\eta}\frac{1}{1-i\frac{\omega_p}{\omega}}. \qquad (2)$$

In this model η and ω_p can be directly calculated from the data by simple inversion. In the high-frequency limit ($\omega \gg \omega_p$) $\rho_{vm,\mathrm{GR}} \to \rho_{\mathrm{ff}}$, being $\rho_{\mathrm{ff}}=\Phi_0 B/\eta$ the free flux flow resistivity. Equation (2) gave for many years the theoretical

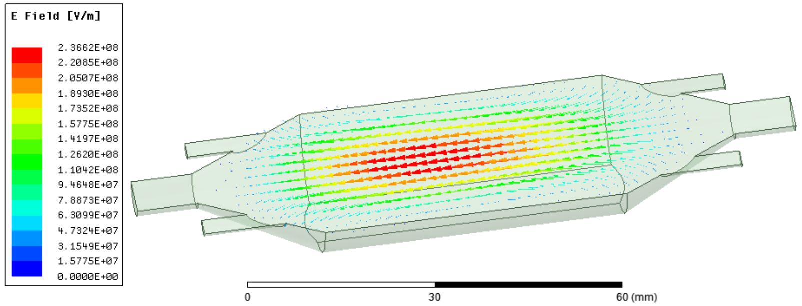
First SC haloscope

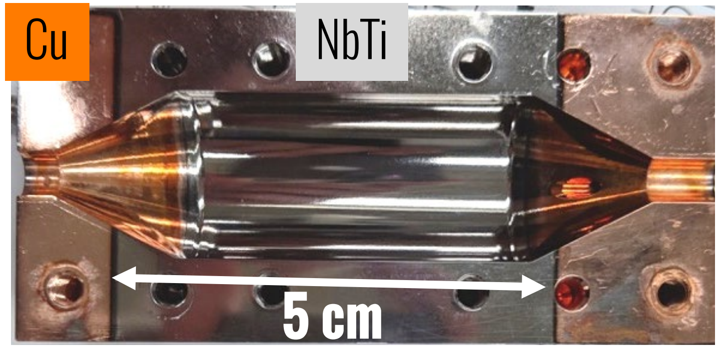
→ Hybrid Geometry

Cavity designed at LNF

Cavity coated at LNL with 4 um NbTi layer

Cu endcaps to reduce vortex motion dissipation





(half cell)

Freq. (TM010)	9.1 GHz
G_{cyl}	6270 W
G_{cones}	482 W
Rs^{Cu}	4.9 mW
Q_0^{Max}	**1.3×10^6**

$$\frac{1}{Q_0} = \frac{R_s^{\mathrm{cyl}}}{G_{\mathrm{cyl}}} + \frac{R_s^{\mathrm{cones}}}{G_{\mathrm{cones}}}$$

Deposition via DCMS:

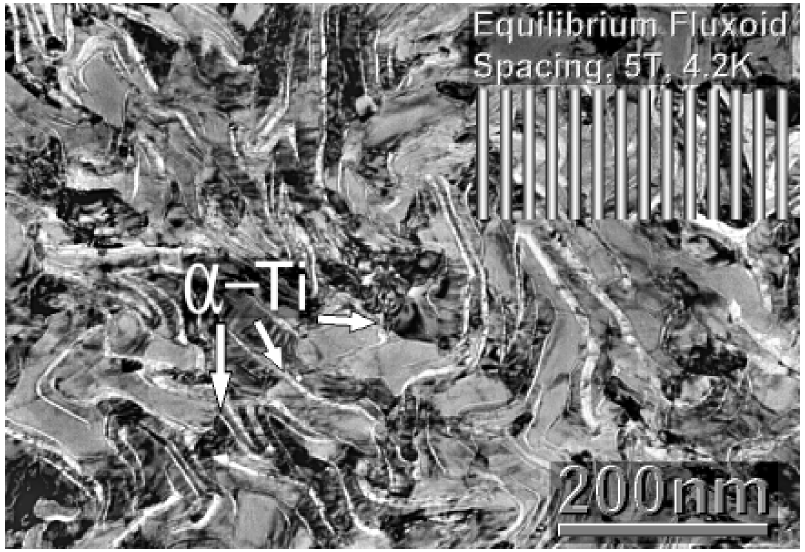
- allows easy deposition (NbTi ≠ Nb_3Sn)
- allows easy exploration of Ti concentration
- allows application of mask for copper cones

[1] D. Alesini, Phys. Rev. D 99, 101101(R) (2019)

dorothea.fonnesu@lnl.infn.it

NbTi pinning

α-Ti precipitates act as pinning centers in NbTi alloys

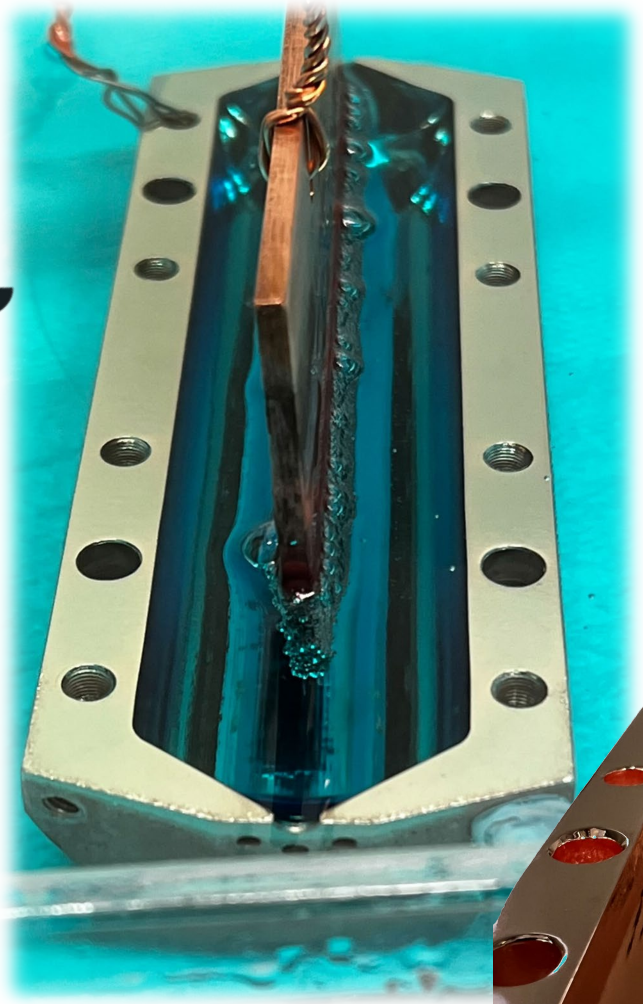


D. C. Larbalestier and P. J. Lee," *Proceedings Particle Accelerator Conference*, Dallas, TX, USA, 1995

- **Pinning Force has a maximum with Ti content** (we expect similar pinning for $Nb_{0,31}Ti_{0,69}$ and $Nb_{0,38}Ti_{0,62}$)
- α-Ti precipitates density and dimension depends on thermal treatments
- Data only for wires → **no data for thin films**

Surface Preparation

1. Ultrasonic degreasing in Rodatel-30 soap
2. Ultrasonic in deionized water
3. **Electropolishing**
   in H_3PO_4: Butanol at 3:2 volume ratio
4. Chemical polishing in SUBU-5 solution
5. Surface passivation with sulfamic acid
6. Ultrasonic, ethanol rinsing and drying
7. 100 bar High Pressure Water Rinsing

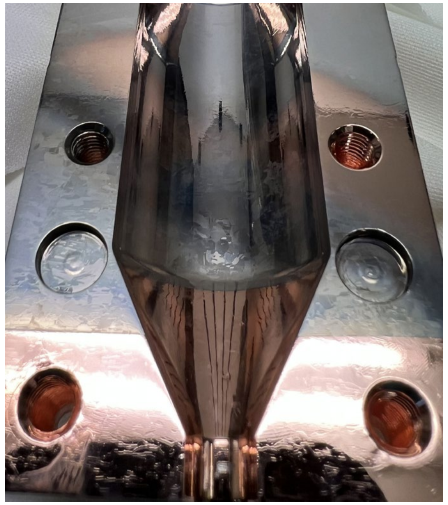


Surface preparation is a key process for SRF cavities

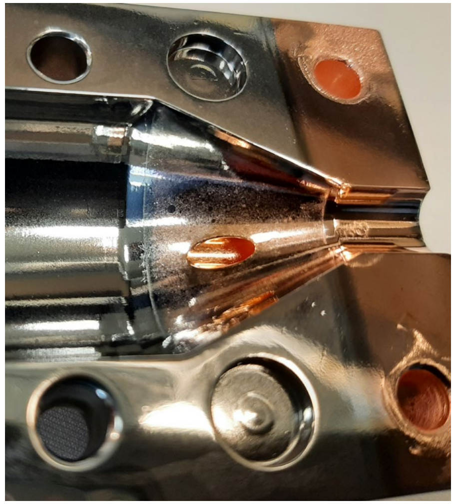
Coating mimate substrate surface

Ra < 100 nm

Defects on the cavity surface

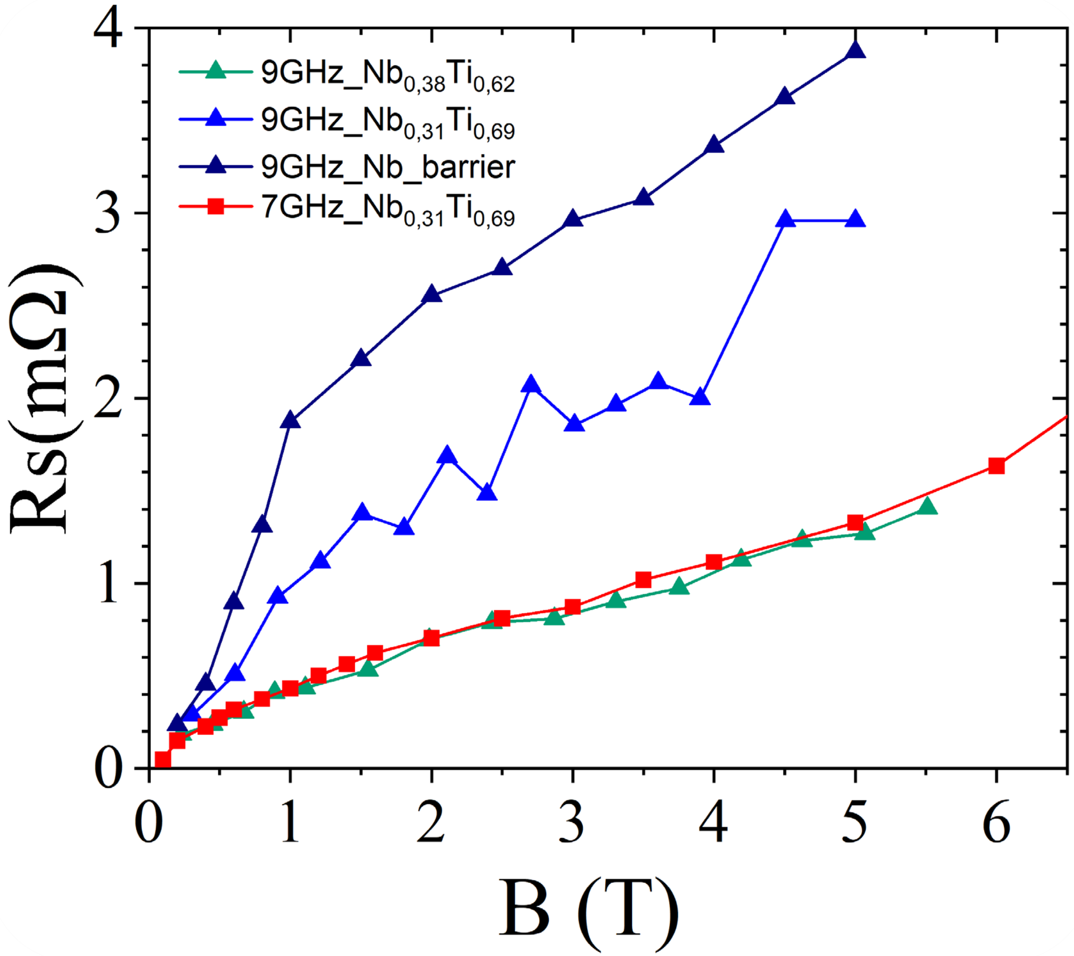


Large grain boundaries



Pitting + NbTi coating on Cu cones

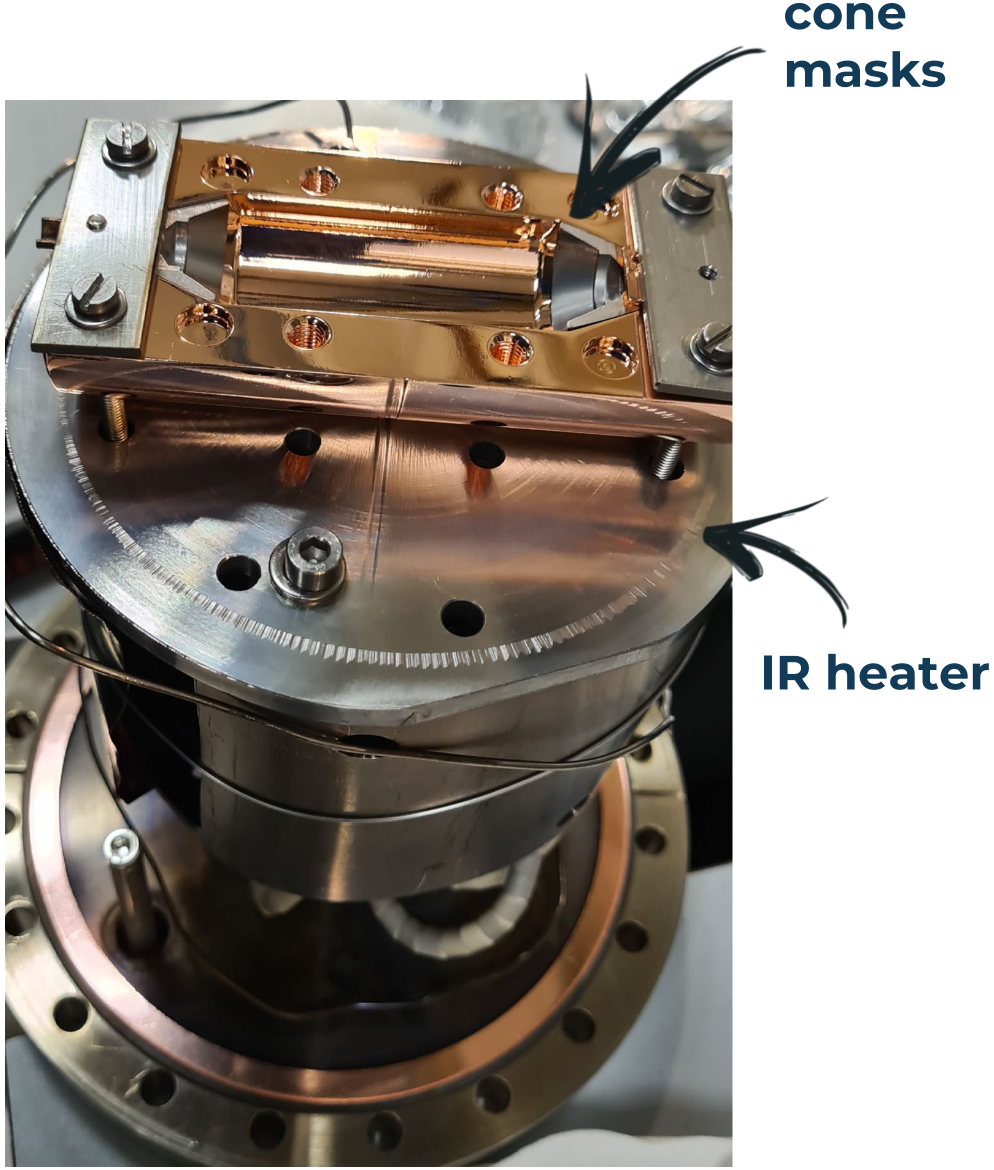
All measurements at 4 K



dorothea.fonnesu@lnl.infn.it

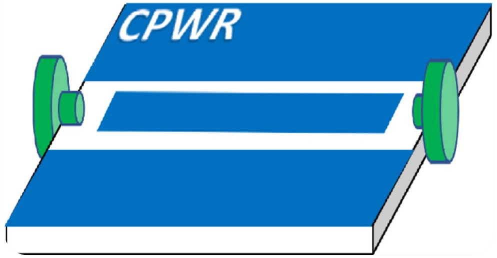
NbTi Coating Set-up

- DC Magnetron Sputtering
- Single NbTi target
- Ar pressure $6 \cdot 10^{-3}$ mbar
- T substrate 500 °C
- Film thickness 2,5 – 3.5 μm
- Base pressure: $< 9 \cdot 10^{-9}$ mbar @roomT

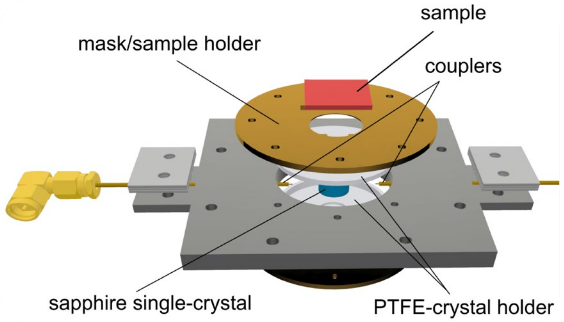


NbTi characterisation

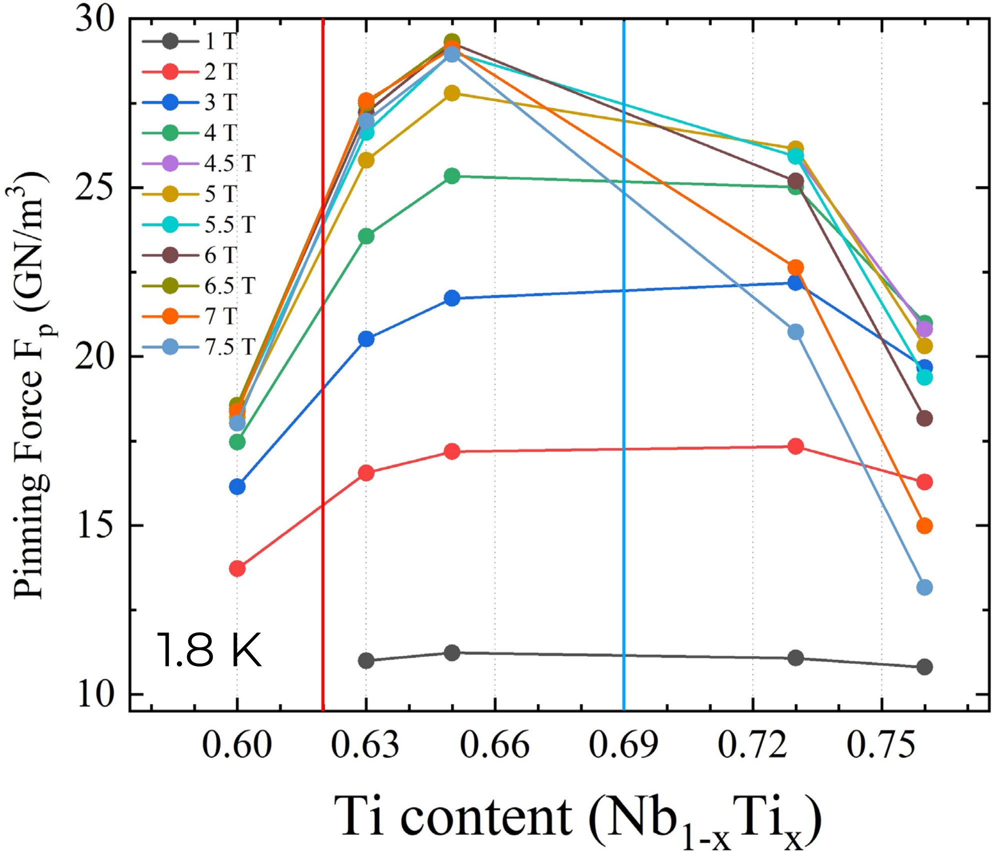
INFN Torino

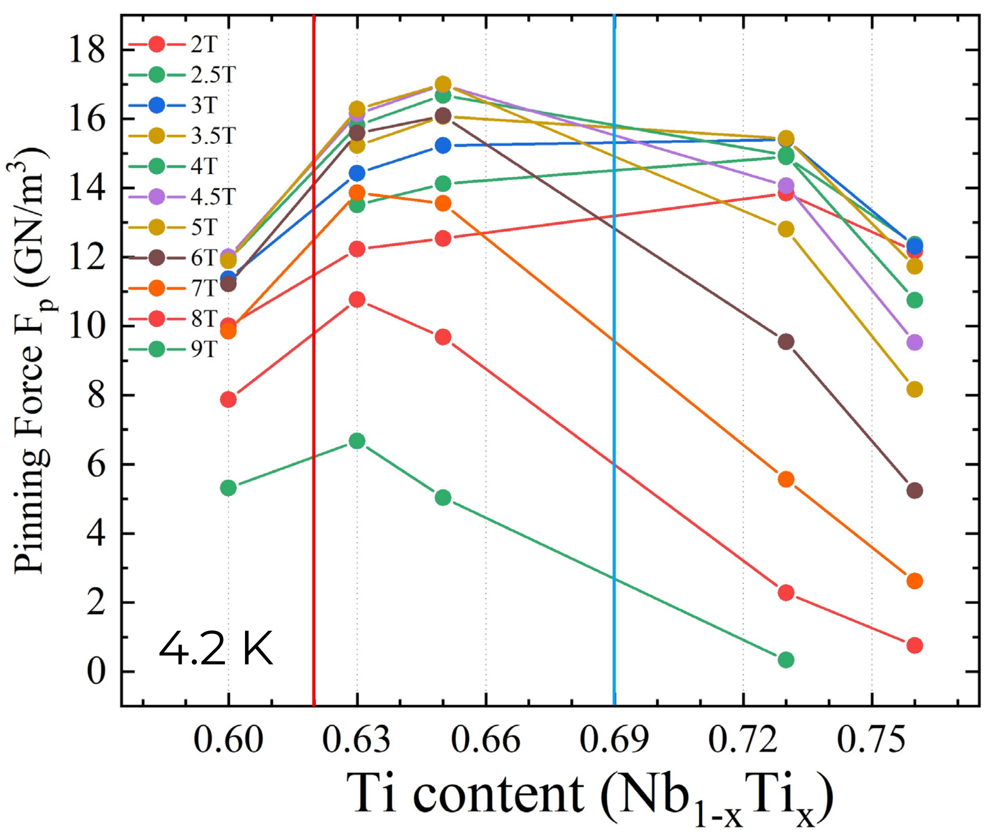


Roma 3 university



NbTi pinning force



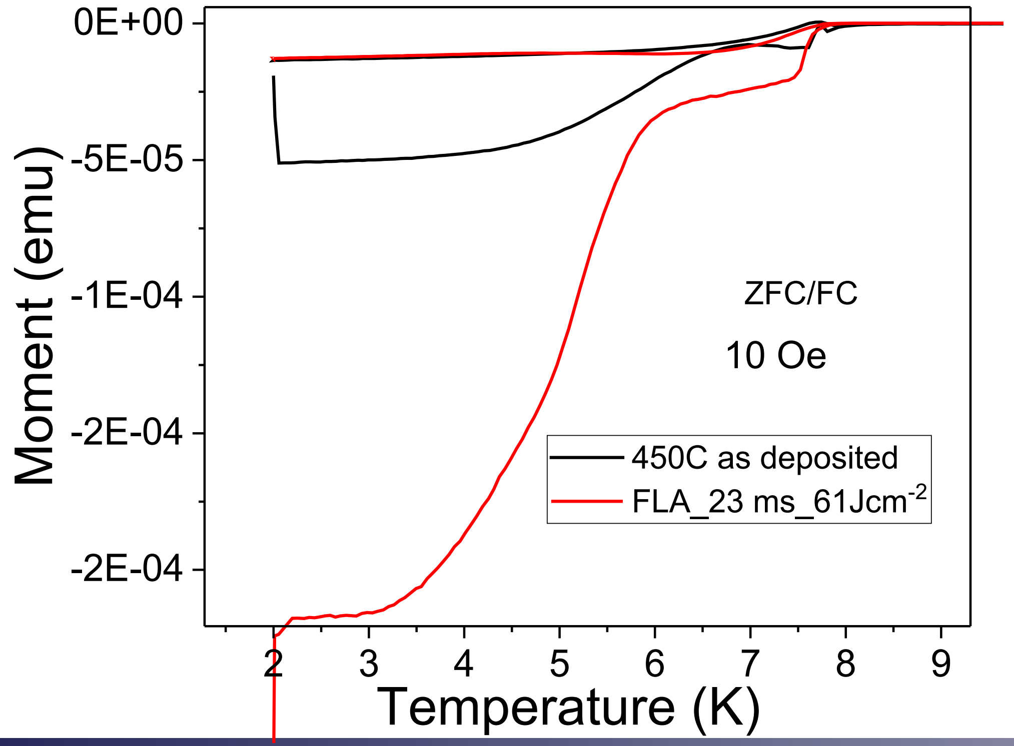


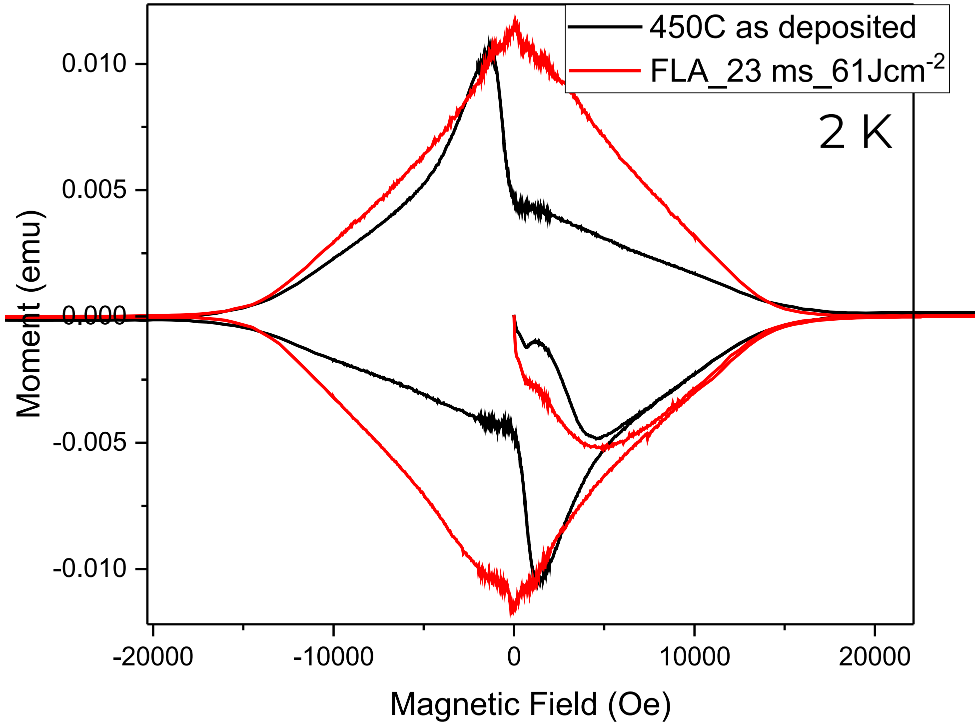
Flash annealing tests (HZDR)

Original idea: can a “bad sample” improve with FLA? (seems not…)

- 1 µm Nb_3Sn on sapphire, Cu, Cu+ 1 µm Nb buff
- coating parameters: T_{coat} = 450 C, p_{coat} = 2 x 10^{-2} mbar, P = 20 W
- two sets:
  - 1 set annealed, t_{ann} = 24 h (shown here)
  - 1 set not annealed

NEXT:
test effect on “good sample”





Technical issues

Two headaches:

1. Leaks
    - runs to be repeated several times due to CF copper gaskets not standing the temperature gradient (both time and temperature-wise)
    - OFHC copper gaskets by Pfeiffer are granted for -196 – 200 °C T range
    - other companies offer up to 400 °C
    - OFE copper might be an option
2. IR lamps
    - not designed to stand high temp for long times, fail each 3 - 4 runs

What's your experience?

Any advice?