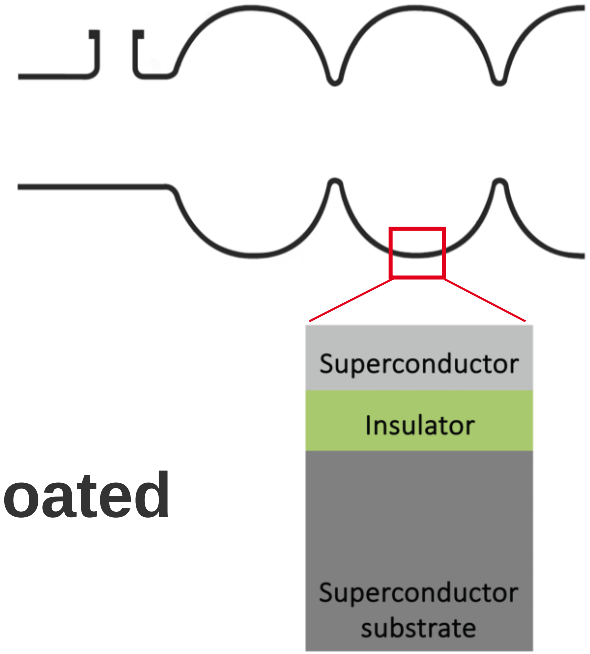
Magnetic Characterization of PEALD Coated Thin Films for SRF Cavity Research



Lea Preece

10.07.2024 Master Colloquium

Motivation

- SRF cavities approach thermodynamic limit of niobium

Goals

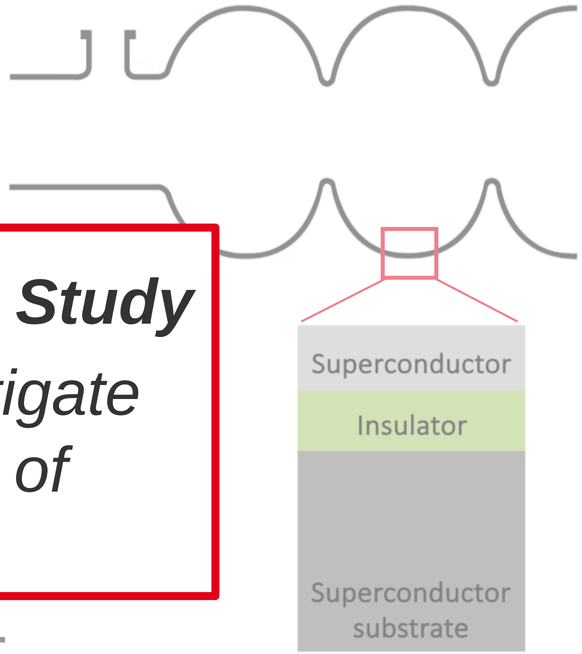
- To achieve lower
  - Coat inside

    since R_{BCS} ∝

- To enhance acce
  - Surpass field limit H_{sh} by coating **thin Superconductor-Insulator-Superconductor (SIS) multilayers**

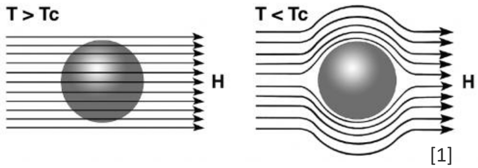
Magnetic Characterization Study

⇛ comprehensively investigate the magnetic properties of SIS layers



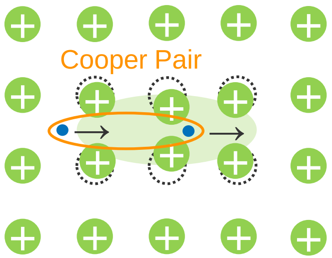
Superconductivity in Particle Accelerators

Meissner effect



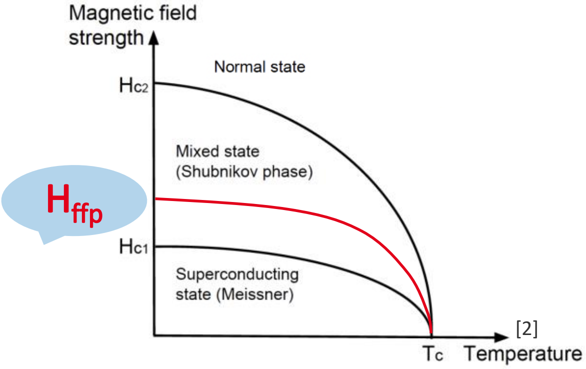
BCS and GL theory

Microscopic electron-lattice-interaction



$$\kappa_{GL} = \frac{\lambda_L}{\xi_{GL}} \begin{cases} < \frac{1}{\sqrt{2}} & \text{Type I SC} \\ > \frac{1}{\sqrt{2}} & \text{Type II SC} \end{cases}$$

Type II SCs



Niobium as material of choice for SRF cavities

Highest known T_C of **9.27 K**

Highest known H_{C1} of **180 mT**

⇛ Upper limit for rf vortex penetration even higher

Superheating field

H_{sh} ~ 200 to 250 mT

Limiting Performance Factors

Surface Resistance

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

R_{BCS} vanishes for $T \rightarrow 0$ K

Everything *except* R_{BCS}

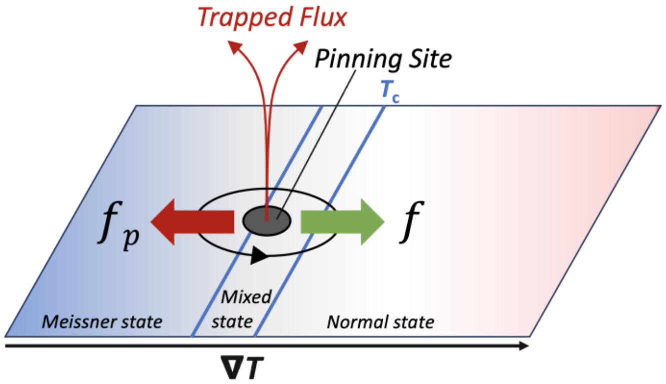
- Weakly coupled grain boundaries, surface oxides etc.
- **Trapped magnetic flux**

Q_0 improves with lower R_S

... but is limited by various factors!

→ Magnetic Quenching when $H_{applied} > H_{C1}$

→ Dissipation becomes unmanagable!

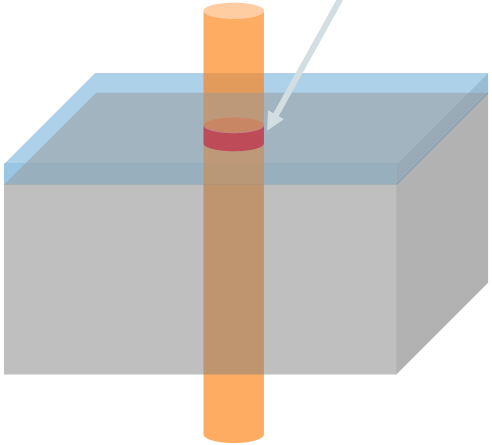


E_{acc} is limited by dissipation because of high surface fields!

Thin Films and Screening Currents

A. Gurevich, APL 88 (2006), T. Kubo, SUST 30 (2017)

- *Gurevich:* Vortices are energetically supressed in SC thin film
  - $d < \lambda_L$
  - **Pancake vortices**
- *SIS* multilayer idea: insulator as vortex barrier
  → global vortex penetration is prevented
- *Kubo's new argument:* Vortex penetration shifts to H_{ffp}>H_{C1} through "**counter current**" at interfaces
  - Layer must be thin enough so that rf field sees counter currents



Vortex dissipation becomes energetically unfavourable

SIS Multilayer Theory

- Top superconductor sees majority of rf field
  → **Surface resistance improves**
- Insulating layer creates more interfaces
  → **More counter currents**
- Layers must be thinner than $\lambda_{L,Top}$
  → **RF field is affected by counter currents**

Theoretical maximum surface field a SIS coated cavity can withstand:

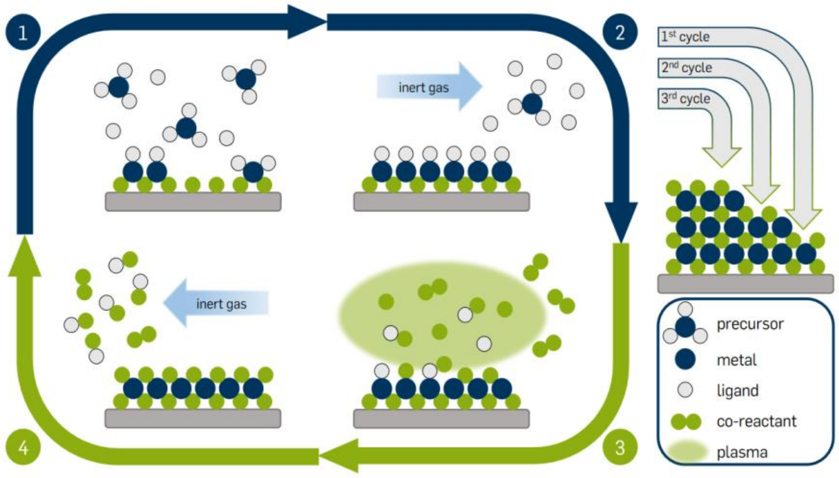
$$H_{max} = \min\{\tilde{\gamma}_1^{-1} H_{sh,S}, \tilde{\gamma}_2^{-1} H_{sh,sub}\}$$

$$\tilde{\gamma}_1 = \frac{\sinh\frac{d_S}{\lambda_S} + \frac{\lambda_{sub}+d_I}{\lambda_S}\cosh\frac{d_S}{\lambda_S}}{\cosh\frac{d_S}{\lambda_S} + \frac{\lambda_{sub}+d_I}{\lambda_S}\sinh\frac{d_S}{\lambda_S}}$$

$$\tilde{\gamma}_2 = \frac{1}{\cosh\frac{d_S}{\lambda_S} + \frac{\lambda_{sub}+d_I}{\lambda_S}\sinh\frac{d_S}{\lambda_S}}$$

Sample Preparation

Plasma-Enhanced Atomic Layer Deposition (PEALD)



[3]

- Uniform layer-by-layer thin film coatings
- Various advantages
  - Low-temperature deposition
  - High quality, high conformality
  - Vast choice of precursors
- Self-regulating process
- Convenient for coating the inside of an SRF cavity

Sample Preparation

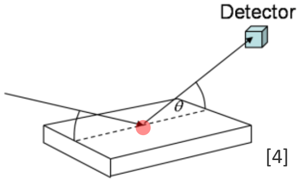
PEALD Process Parameters

Precursor	Target Material
TMA	AlN
TDMAT	TiN
TBTDEN	NbN

- Plasma mix of H_2 and N_2
- Deposition temperature 250 °C

Characterization Measurements

X-Ray Reflectivity (XRR)



[4]

- Determine thin film thickness
- Reflectivity curve shows interference oscillations
  → periodicity related to layer thickness

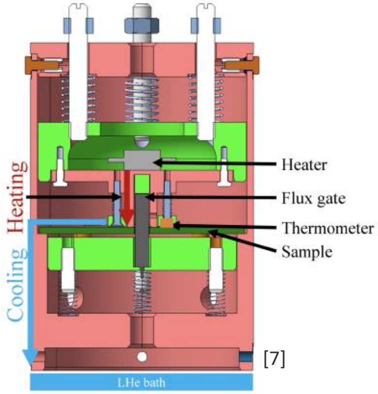
$$d = \frac{\lambda}{2(\Delta\Theta)}$$

Oscillation width

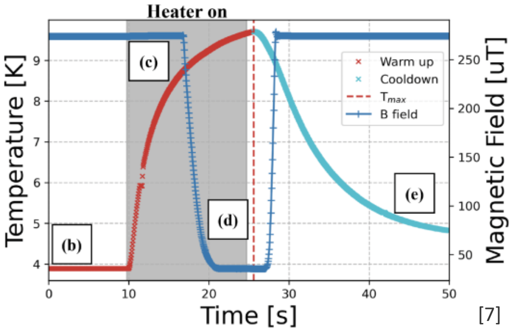
Characterization Measurements

Magnetic Flux Expulsion Lens (MFEL)

- SC/NC sample transition with pulsed heating under controlled cooling conditions



[7]



[7]

PEALD Growth per Cycle for NbN, TiN and NbTiN

1 supercycle NbTiN = 3 x NbN + 1 x TiN

Two-fold results for NbTiN:

1. GPC calculated from measured XRR NbTiN thickness
2. GPC calculated from individual GPCs of NbN and TiN

✓ Results within the expected range

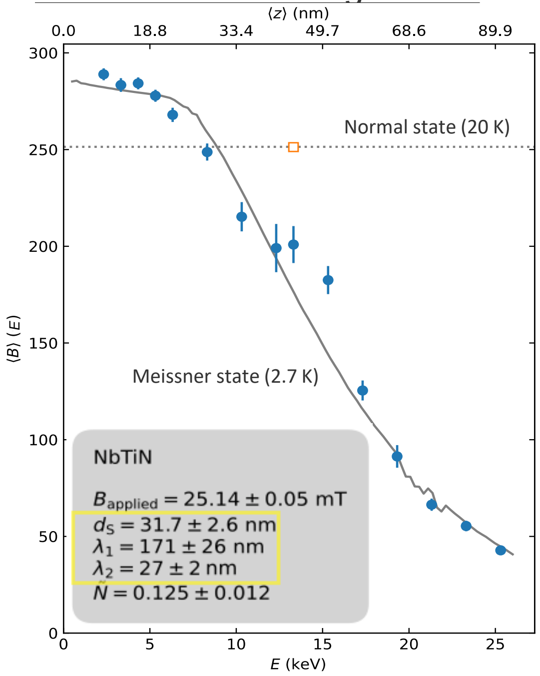
Coating	Cycles/ Supercycles (#)	Estimated thickness (nm)	Measured thickness (nm)	GPC measured (nm/(sup)cyc.)	GPC calculated (nm/supcyc.)
NbN	1000	~ 60	52.23 ± 2.91	0.052 ± 0.003	-
TiN	200	15.95	15.34 ± 0.85	0.077 ± 0.004	-
NbTiN	243	~ 60	56.74 ± 2.91	0.233 ± 0.012	0.233 ± 0.005

XRR results

matching NbTiN GPCs

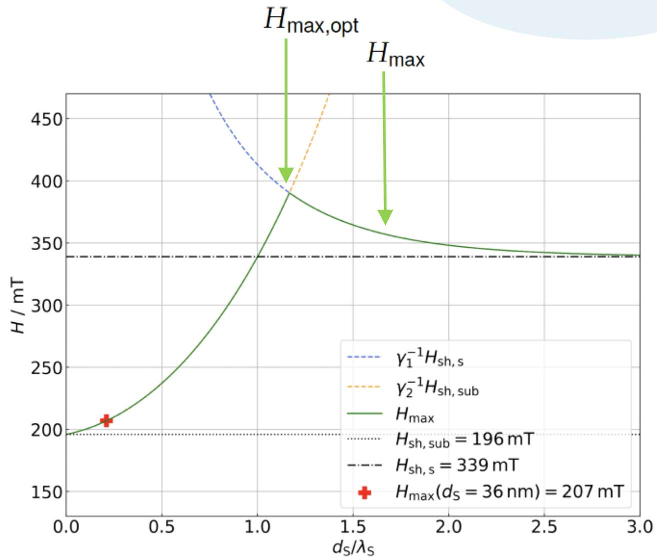
λ_L and the Estimation of H_{ffp}

Meissner Screening Profile



[Credits to Ryan McFadden]

- Nb/NbTiN with d ~ 36 nm
- Kubo's counter current model used for fit
- Expected decrease in B with increasing E and z
- H_{max} **estimated** for Nb/NbTiN SS bi-layer

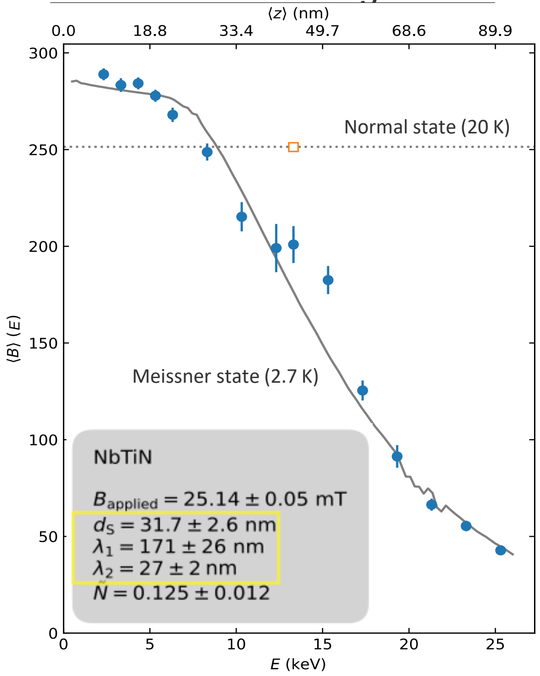


Material	λ (nm)	λ_L (nm)	ξ_0 (nm)	κ (nm)	H_{c1} (mT)	H_c (mT)	H_{sh} (mT)
NbTiN	171	150 [65]	5 [65]	43	24	452	339
Nb	27	28 [56]	35 [65]	0.82	(no calc.)	200 [65]	196

Clean limit of λ_L

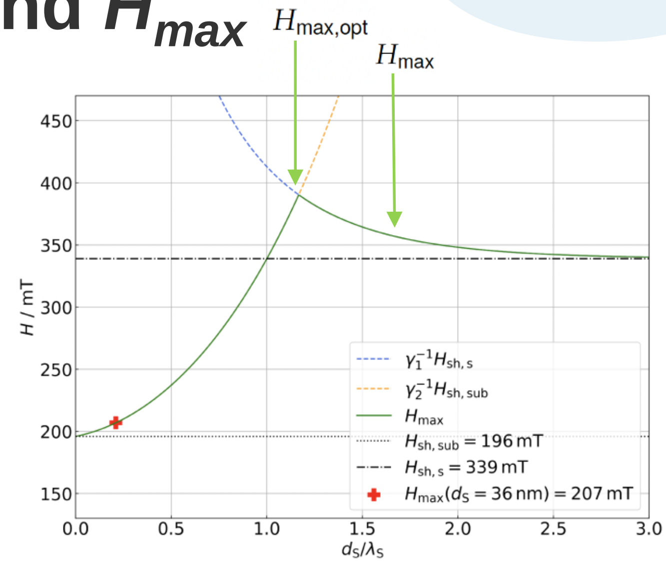
Discussion on Meissner Profile and H_{max}

Meissner Screening Profile

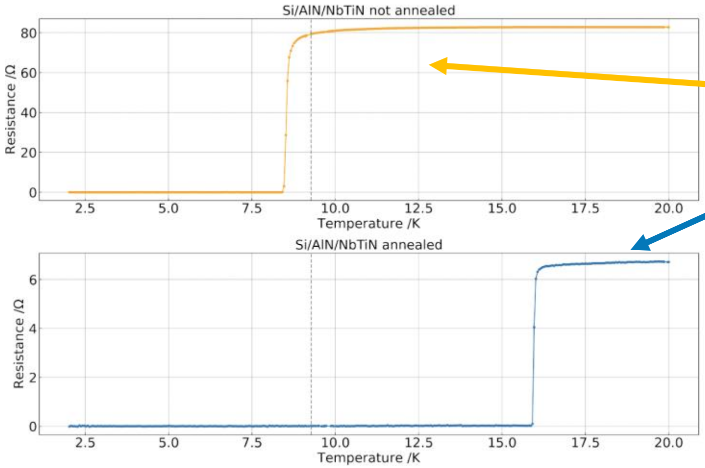


[Credits to Ryan McFadden]

- Kubo's counter current model suitable to determine λ_L
- H_{max} **estimate** visualizes Kubo's prediction
  - $H_{max} > H_{sh,Nb}$
  - SIS multilayers needed!
- Estimate based on Meissner Fit → improvable!
- Illustrates principle of field enhancement in an SS bi-layer



PPMS T_C Measurements on Si



Sample	Ref. to	Measurement	T_c/K	ΔT_c/K
Si/AlN/NbTiN not annealed	DESY_3_1 DESY_5_1 Tc_5_1	Electr. Transport	8.402 ± 0.271	0.931
Si/AlN/NbTiN annealed	DESY_3_2 DESY_5_2 Tc_5_2	Electr. Transport	15.960 ± 1.952	0.106
Si/AlN/NbTiN annealed	DESY_3_2 DESY_5_2 Tc_5_2	VSM	15.566 ± 0.242	0.666
Si/NbTiN annealed	DESY_2_2 DESY_6_2 Tc_6_2	VSM	14.948 ± 0.409	0.384
Si/AlN/NbN annealed	DESY_4_2	Electr. Transport	2.732 ± 0.070	0.880

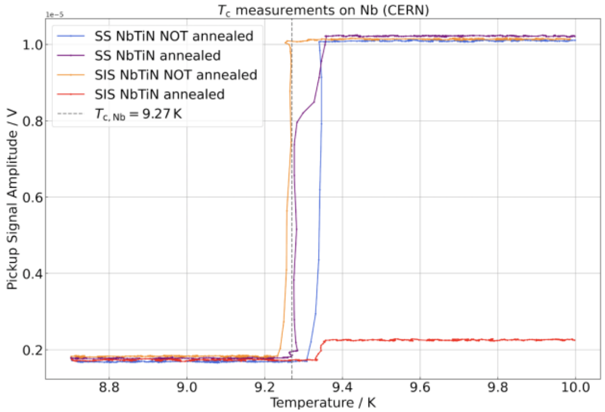
Highest value measured

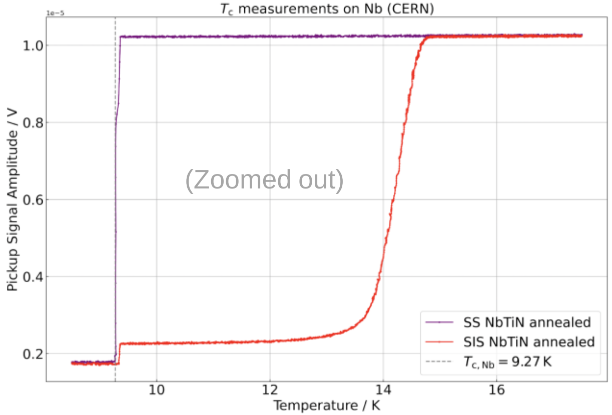
Not the same for Nb substrate!

- T_C can be clearly assigned to the SC thin film
- Highest T_C for **Si/AlN/NbTiN** in both measurement modes
- Measurement of an SIS Si/AlN/NbN sample → very low T_C

Contactless Inductive T_C Measurements on Nb

- As-deposited: T_C not higher than 7 to 8 K
- SS with NbTiN behaves similar as unannealed samples
  - ➢ $T_C \sim T_{C,Nb}$
  - ➢ Two transitions?
- Only **SIS with NbTiN**
  → shows clear stepwise transition
  → reaches high T_c

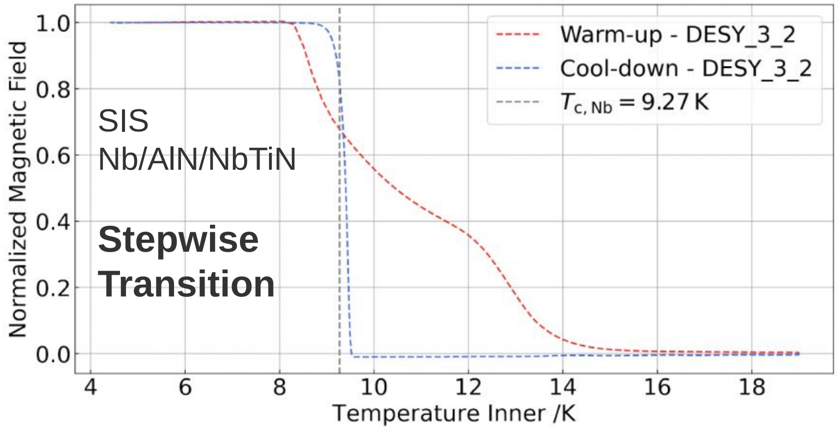




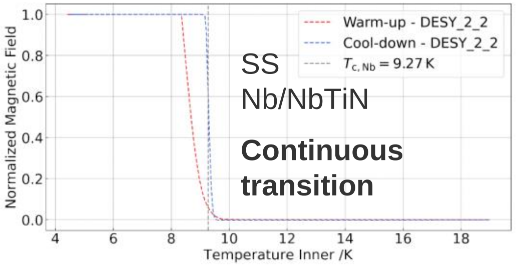
Sample	Structure	$T_{c(1)}/\mathrm{K}$	$\Delta T_{c(1)}/\mathrm{K}$	$T_{c(2)}/\mathrm{K}$	$\Delta T_{c(2)}/\mathrm{K}$
Tc_6_1	Nb/NbTiN not annealed	9.339 ± 6.890	0.032	-	-
Tc_5_1	Nb/AlN/NbTiN not annealed	9.259 ± 6.998	0.023	-	-
Tc_6_2	Nb/NbTiN annealed	9.276 ± 10.663^{8}	0.044	9.340 ± 0.326	0.033
Tc_5_2	Nb/AlN/NbTiN annealed	9.343 ± 1.407	0.0144	14.196 ± 0.014	0.835

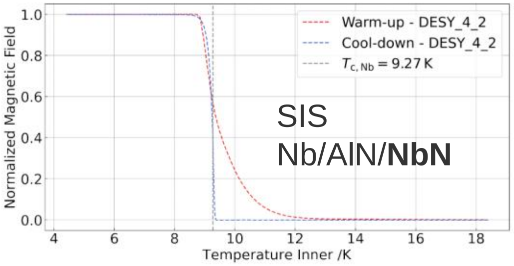
MFEL T_C Measurements

- Annealed Nb SIS and SS samples
- Only **SIS with NbTiN**
  → shows stepwise transition
  → reaches high T_c
- No double transition or significant increase in T_c for
  → SS Nb/NbTiN
  → SIS Nb/AlN/**NbN**
- **All T_c measurements deliver matching results!**



[Credits to Daniel Turner]





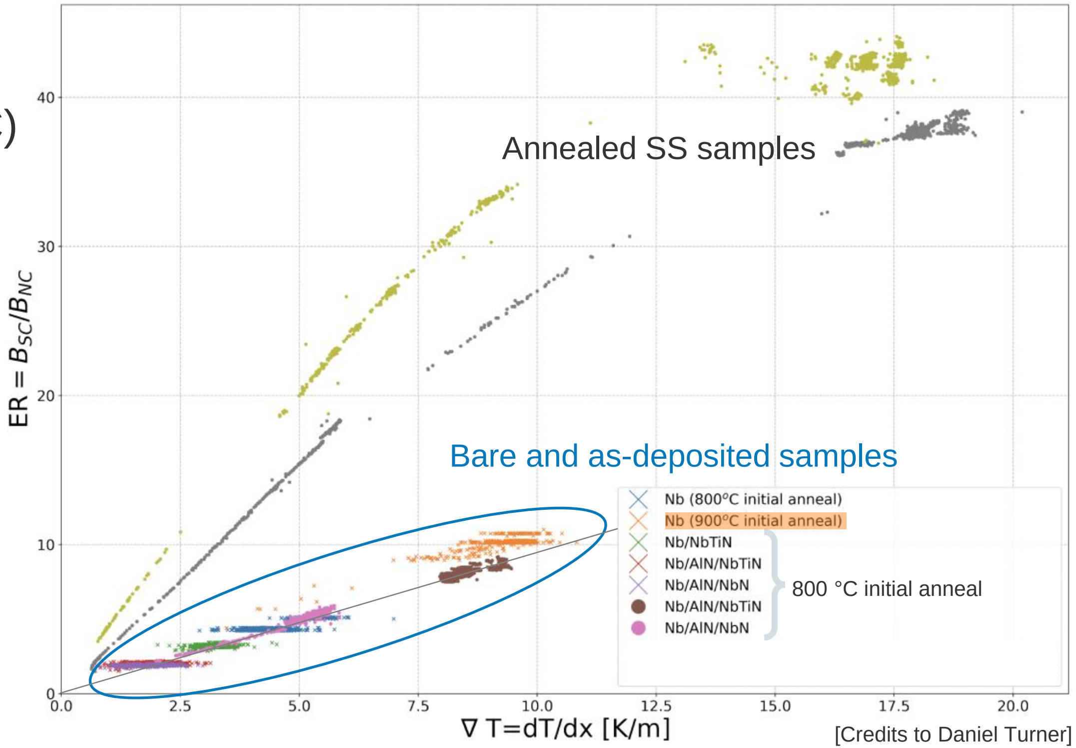
Magnetic Flux Expulsion

<u>As-deposited SIS and SS thin films</u>

- ER(Nb@900 °C) > ER(Nb@800 °C)
- Limited spatial thermal gradient

<u>Annealed SS thin films</u>

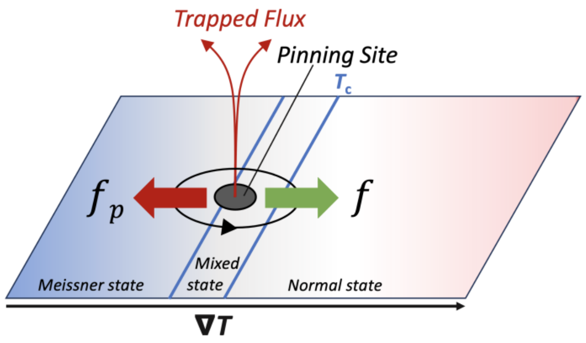
- **Much higher flux expulsion**
- Greater expulsion for 900 °C initial anneal



[Credits to Daniel Turner]

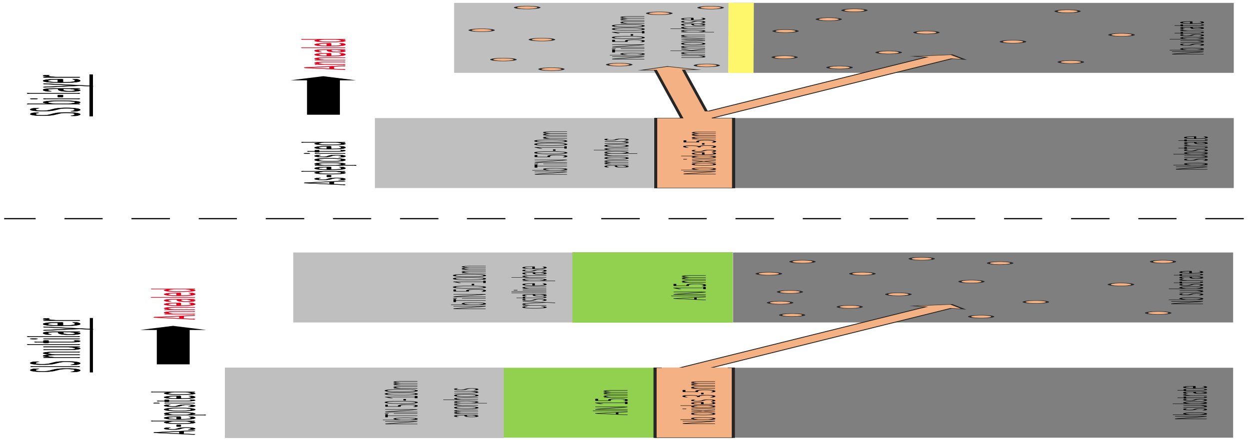
Discussion on T_C and Flux Expulsion

- Better flux expulsion for 900 °C initial anneal
  - Increase in grain size with higher T_{anneal}
  - Reduction in pinning sites
- Post-deposition annealing enhances flux expulsion
  - Even for SS Nb/NbTiN even if no high T_C is achieved
  - Assumption: Pancake effect adding additional force that counteracts pinning force



Discussion on T_C and Flux Expulsion

- High-T annealing required to *activate* the thin film and achieve high T_C
- Insulating layer on Nb required to ensure increase in T_C
  - Barrier layer prevents oxygen from diffusing into NbTiN layer
  - XRD confirms NbTiN δ-phase formation for SS sample

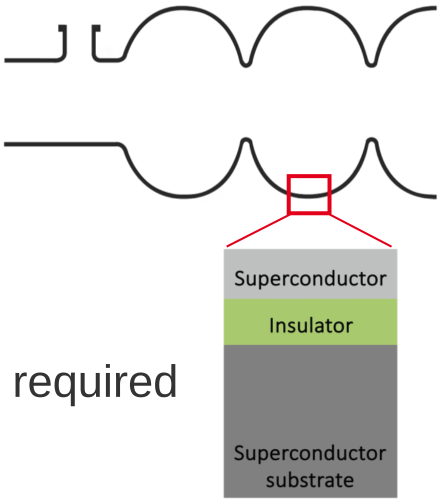


Conclusion

- Unclear if reduction of T_C implies reduction of H_C
  - Experimental determination required
  - Useful comparison with estimation of H_{max}
- Findings on oxygen diffusion allow new considerations on µSR data analysis
  - Intermediate layer?
  - Electron Microscopy probably shows oxygen-enriched phase with varying stoichiometry
- 900 °C annealing of SIS Nb/AlN/**NbN**
  - Does **not** form intended high-T_C δ-phase
  - Ti as stabiliser of the cubic high-T_C δ-phase
  - **NbN excluded from SIS studies**

Summary

- Further improvement of Nb SRF cavity performance neccessary...
- **... possible with PEALD coated SIS multilayer!**
  - Coating thin high-T_C superconducting and insulating layers
  - Pushing the field of first flux penetration H_{ffp}
- More characterization and experimental testing of the SIS theory required



SIS is a promising approach towards new technologies and improved future applications in SRF research!

Thank you! Gracias! Danke!

Isabel, Ricardo,
Rezvan, Jonas, Chris,
Getnet, Marco, Artem,
Chriag, Leon, Michel,
Anton, Thomas, Cem, Marc,
Rakshith, Cornelius, Wolfgang,
Robert, Thorsten, Detlef, Dimitri,
Alexey, Dan, Alick, Ryan, Andreas

References

[1] Irwin Yousept. “Optimal Control of Non-Smooth Hyperbolic Evolution Maxwell Equations in Type-II Superconductivity”. In: SIAM Journal on Control and Optimization 55 (Jan. 2017), pp. 2305–2332. COI: 10.1137/16M1074229

[2] Irwin Yousept. “Optimal Control of Non-Smooth Hyperbolic Evolution Maxwell Equations in Type-II Superconductivity”. In: SIAM Journal on Control and Optimization 55 (Jan. 2017), pp. 2305–2332. COI: 10.1137/16M1074229

[3] Lukas Mai. “Investigation of amino-alkyl coordinated complexes as new precursor class for atomic layer deposition of aluminum, tin and zinc oxide thin films and their application”. doctoralthesis. Ruhr-Universität Bochum, Universitätsbibliothek, 2020. DOI: 10.13154/294-7658

[4] https://de.wikipedia.org/wiki/Röntgenreflektometrie

[5] https://de.wikipedia.org/wiki/Larmorpräzession

[6] https://de.wikipedia.org/wiki/Myon

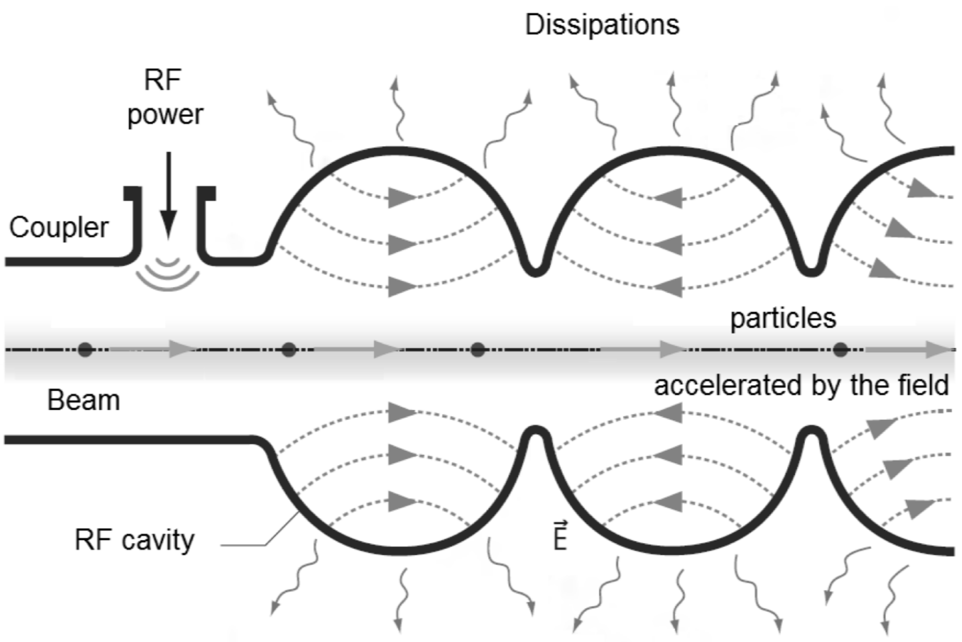
[7] Daniel Turner et al. “Flux Expulsion Lens: Concept and Measurements”. In: JACoW SRF2023 (2023), MOPMB003. DOI: 10.18429/JACoW-SRF2023-MOPMB003

Backup

PEALD System at Universität Hamburg

UHV Furnace at Universität Hamburg

Cavity Resonator



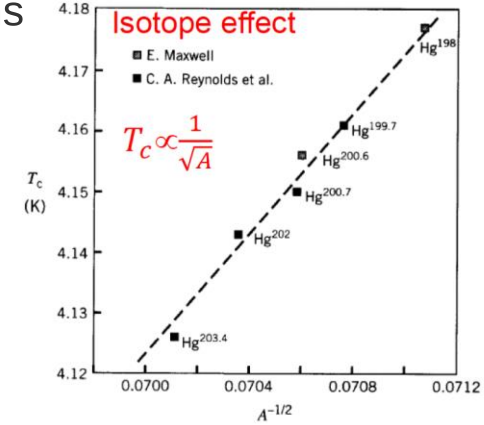
[C. Antoine, EuCARD-BOO-2012-001 (2012)]

Theory of Superconductivity

$$\Delta(0) = 1.76 k_B \cdot T_c$$

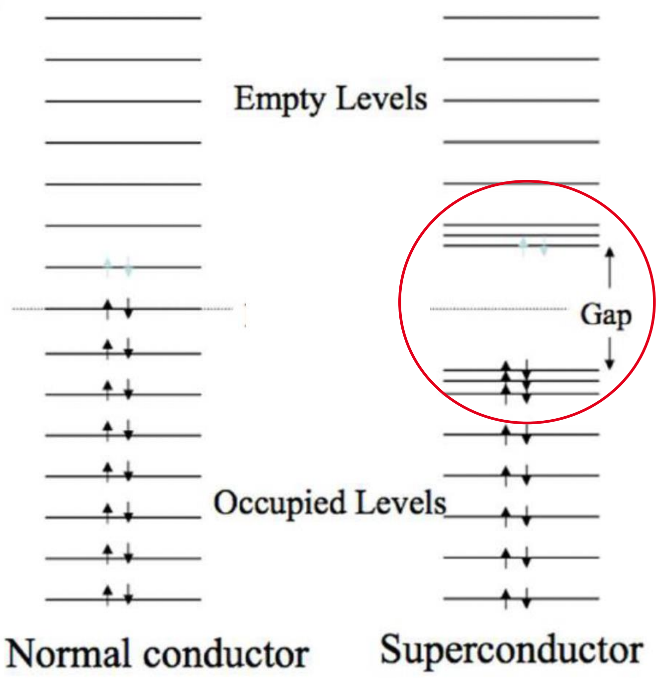
Isotope effect

- First clue that SC is linked to atomic lattice, not just electrons



SC Gap

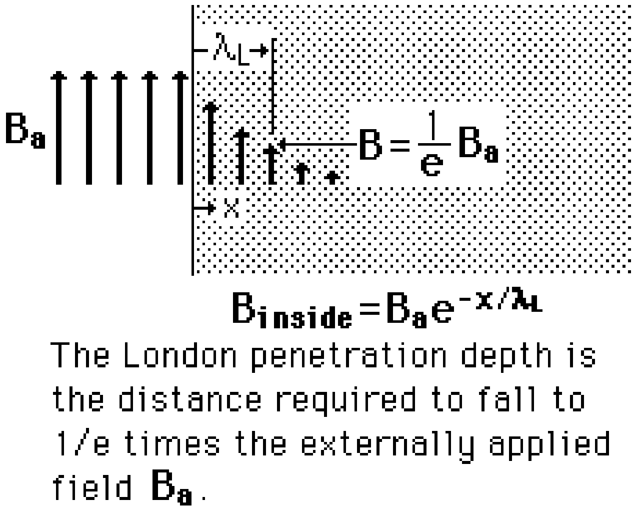
- Sc electrons (bound state) only conduction electrons that interact with lattice at finite phonon frequency
- Energy gap around E_F



Theory of Superconductivity

London Penetration depth

$$\lambda_L(T) = \frac{\lambda_L(T = 0\,\mathrm{K})}{\sqrt{1 - \left(\frac{T}{T_c}\right)^4}}$$



Surface Resistance

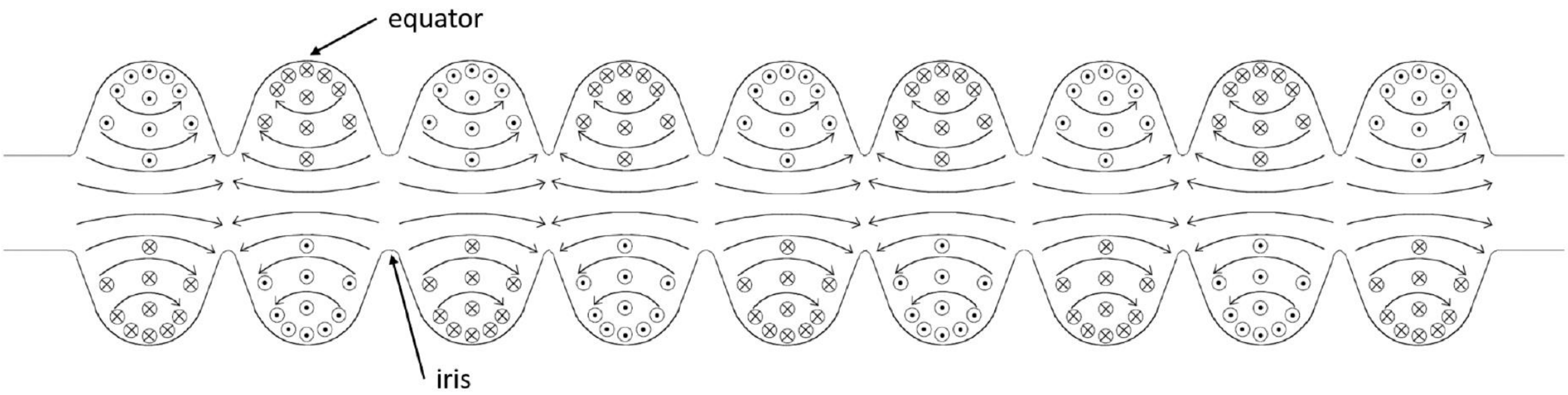
$$R_S = A\frac{\omega^2}{T}\exp\left(-\frac{\Delta}{k_B T}\right) + R_{res}$$

R_{BCS} vanishes for $T \to 0$ K

- R_{BCS} decreases exponentially with respect to the temperature (higher T more electron will participate in the losses)
- Proportional to the square of the frequency (higher frequency means less effective shielding)
- **Non-vanishing AC resistance**
- Cooper pairs have inertia
  - Can not follow the RF field instantly
  - Do not shield RF field perfectly
- 'Normal' electrons are accelerated and dissipate power

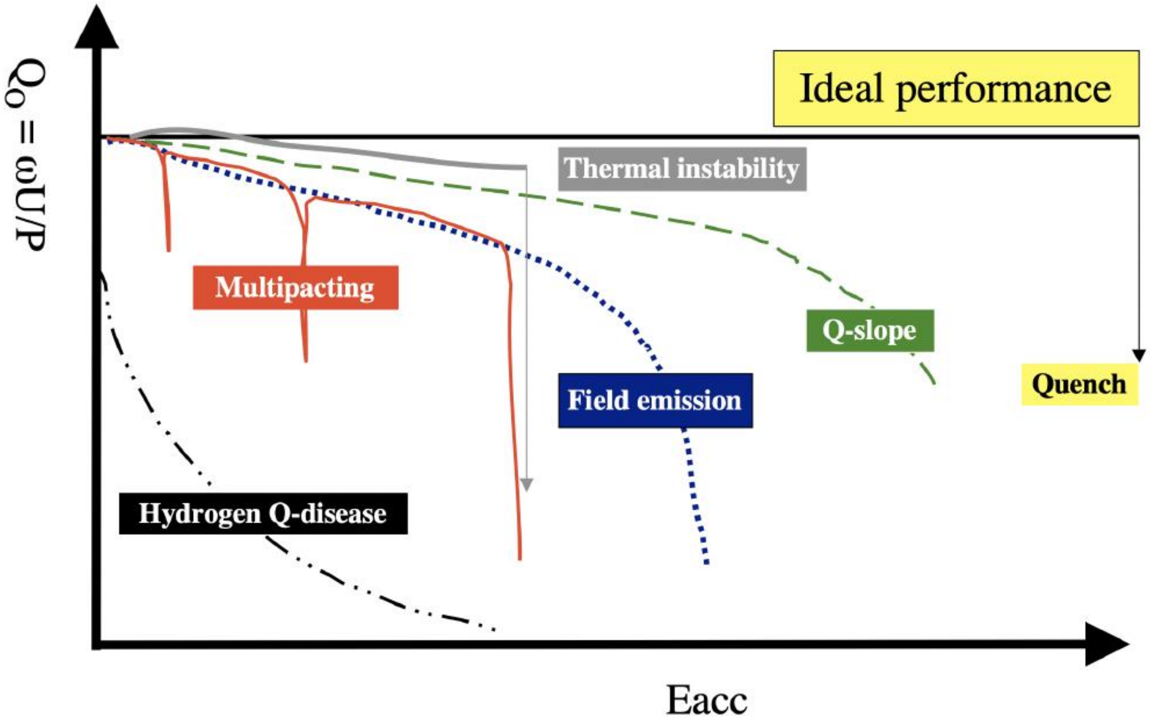
9-cell TESLA-type niobium cavity

$$E_{\text{acc}} = \frac{1}{L}\int_{-L/2}^{L/2} E(z)\cos\left(\frac{\omega z}{v}\right) dz$$



Quality Factor Q_0

$$Q_0 = \frac{\omega_0 U}{P_{\text{diss}}} \equiv \frac{G}{R_S}$$



Thin Films and Pancake Vortices (Gurevich)

- Nucleation of parallel magnetic vortices is energetically suppressed in SC thin films
- Yield of H_{C1}:

$$H_{c1} = \frac{2\phi_0}{\pi d^2} \ln \frac{d}{\tilde{\xi}}$$

- A coating of N superconducting thin layers then screens the interface field H_i on the Nb surface down to

$$H_i = H_0 \exp\left(\frac{-Nd}{\lambda}\right)$$

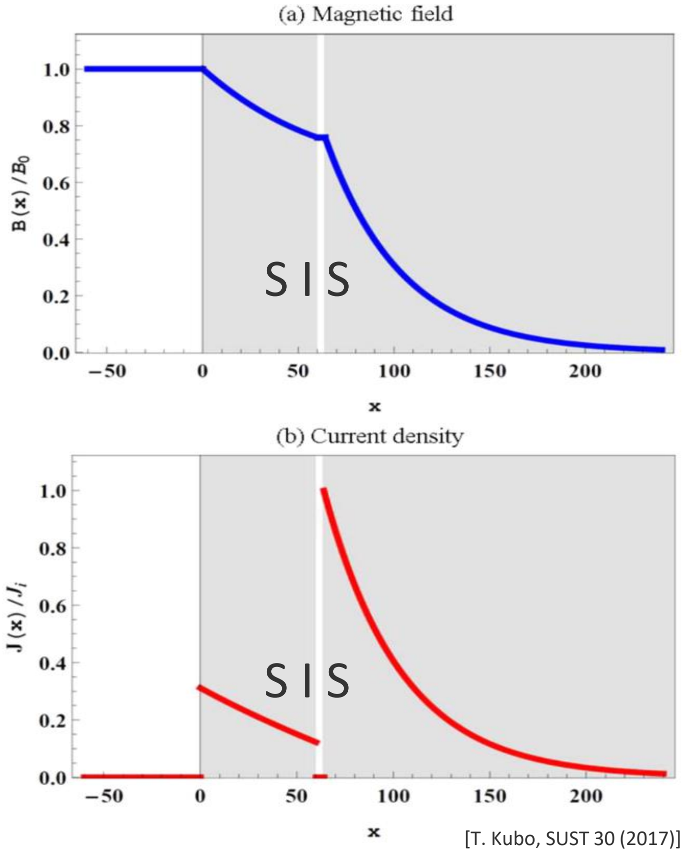
Kubo's Theory

Theoretical maximum surface field a SIS coated cavity can withstand:

$$H_{\rm max} = \min\{\tilde{\gamma}_1^{-1} H_{\rm sh,S}, \tilde{\gamma}_2^{-1} H_{\rm sh,sub}\}$$

$$\tilde{\gamma}_1 = \frac{\sinh\frac{d_S}{\lambda_S} + \frac{\lambda_{\rm sub}+d_I}{\lambda_S}\cosh\frac{d_S}{\lambda_S}}{\cosh\frac{d_S}{\lambda_S} + \frac{\lambda_{\rm sub}+d_I}{\lambda_S}\sinh\frac{d_S}{\lambda_S}}$$

$$\tilde{\gamma}_2 = \frac{1}{\cosh\frac{d_S}{\lambda_S} + \frac{\lambda_{\rm sub}+d_I}{\lambda_S}\sinh\frac{d_S}{\lambda_S}}$$



[T. Kubo, SUST 30 (2017)]

Prediction of maximum surface field H_{max}

$$H_{sh} \approx 0.75\, H_c \quad \text{for} \quad \kappa \gg 1$$

$$H_{sh} \approx \frac{0.89}{\sqrt{\kappa_{GL}}} H_c \quad \text{for} \quad \kappa \ll 1$$

$$\kappa = \frac{\lambda}{\xi_{GL}} = \frac{2\sqrt{3}}{\pi} \frac{\lambda^2}{\xi_0 \lambda_L}$$

$$H_c = \frac{\sqrt{2}\kappa H_{c1}}{\ln \kappa}$$

$$H_{c1} = \frac{\Phi_0}{4\pi\lambda^2}(\ln(\kappa) + 0.497)$$

Material	λ (nm)	λ_L (nm)	ξ_0 (nm)	κ (nm)	H_{c1} (mT)	H_c (mT)	H_{sh} (mT)
NbTiN	171	150 [65]	5 [65]	43	24	452	339
Nb	27	28 [56]	35 [65]	0.82	(no calc.)	200 [65]	196

Clean limit of λ_L

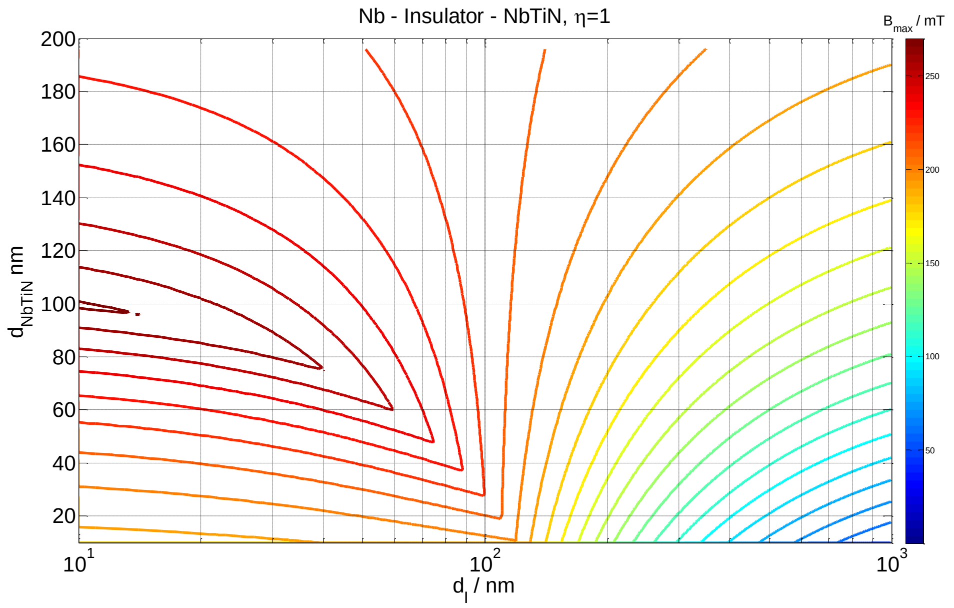
Prediction of maximum surface field H_{max}

- Field enhancement depends on H_{C1}, λ_L of both sc layer and substrate and η

- Assume Nb - Insulator - NbTiN
  - $H_{C1,Nb}$ 180 mT
  - $H_{C1,\, NbTiN}$ 213 mT
  - $\eta = 1$

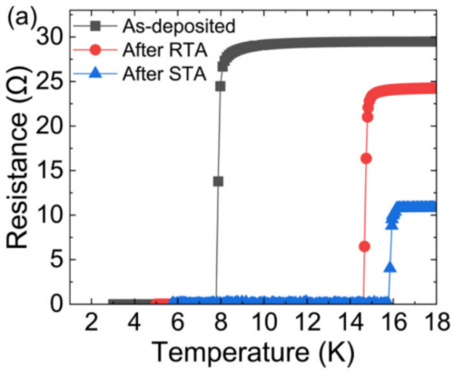
➢ **Isolator 15 nm, NbTiN 100 nm**
  - H_{C1} = 270 mT oder 63 MV/m
  - $\eta = 0.7$

➢ **Isolator 10 nm, NbTiN 60 nm**
  - H_{C1} = 224 mT oder 52 MV/m
  - $\eta = 0.7$



Post-Deposition Annealing

→ **Improvement of T_C and H_{C1}**



$\mu_0 H_{C1}$ = 15 mT
$\mu_0 H_{C1}$ = 81 mT
$\mu_0 H_{C1}$ = 98 mT

$\mu_0 H_{C1,bulk}$ ≈ 33 mT

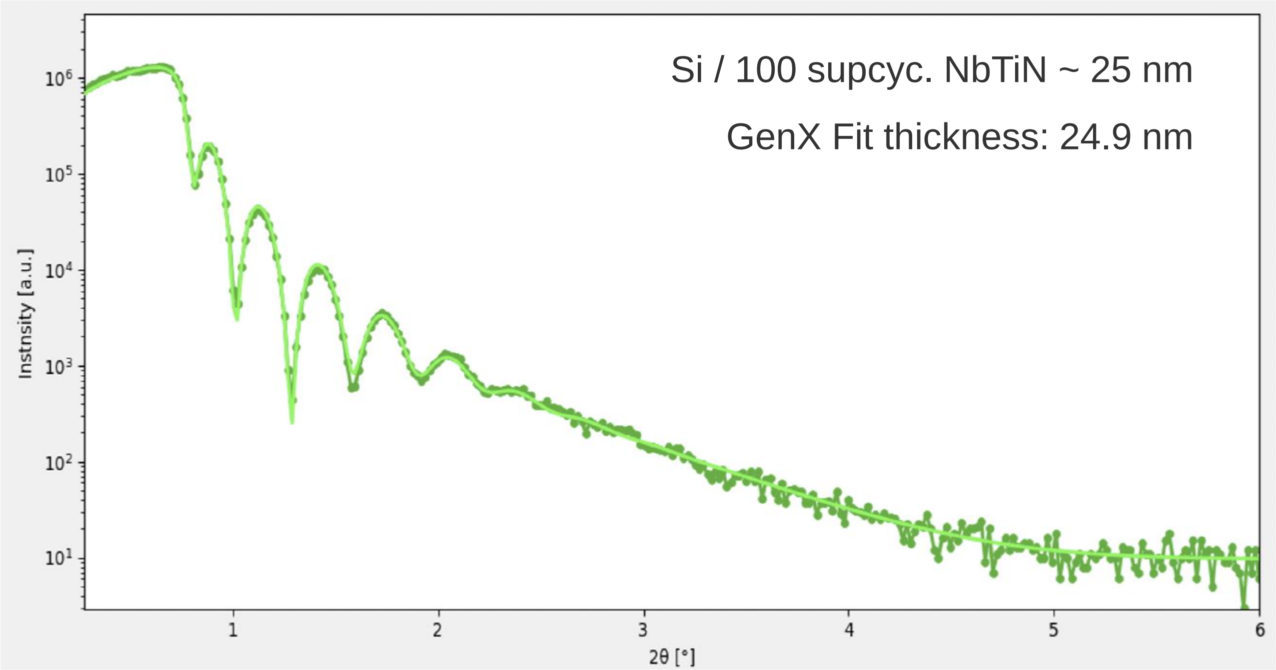
[Gonzalez.I. et al., J. Appl. Phys. 134, 159902 (2023)]
[Gonzalez.I. et al., J. Appl. Phys. 134, 035301 (2023)]

900 °C annealing for 1 hour

→ heating rate 3 °C/min

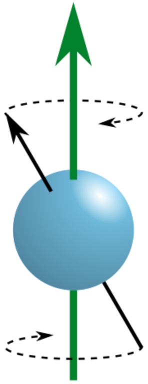
→ controlled cooling rate ~ 1.45 °C/min for 8 hours to 200 °C

XRR

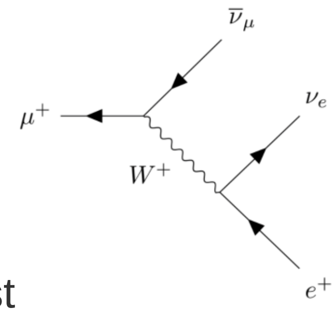

Si / 100 supcyc. NbTiN ~ 25 nm
GenX Fit thickness: 24.9 nm
Instnsity [a.u.]
10^6
10^5
10^4
10^3
10^2
10^1
1
2
3
4
5
6
2θ [°]

How to measure the Meissner Profile with λ_L

- Positively charged, spin-polarized muons are implanted into the material sample to be examined
- passage of a muon through the muon detector determines the time zero of the implantation
- come to rest on interstitial sites or atomic defects
- Interaction of the muon spin with the magnetic moments of the host lattice leads to a characteristic temporal evolution of the spin polarization → different precession frequencies because of field distribution
- The original phase relationship of the individual spins is increasingly lost → rotation signal therefore exhibits a temporal attenuation

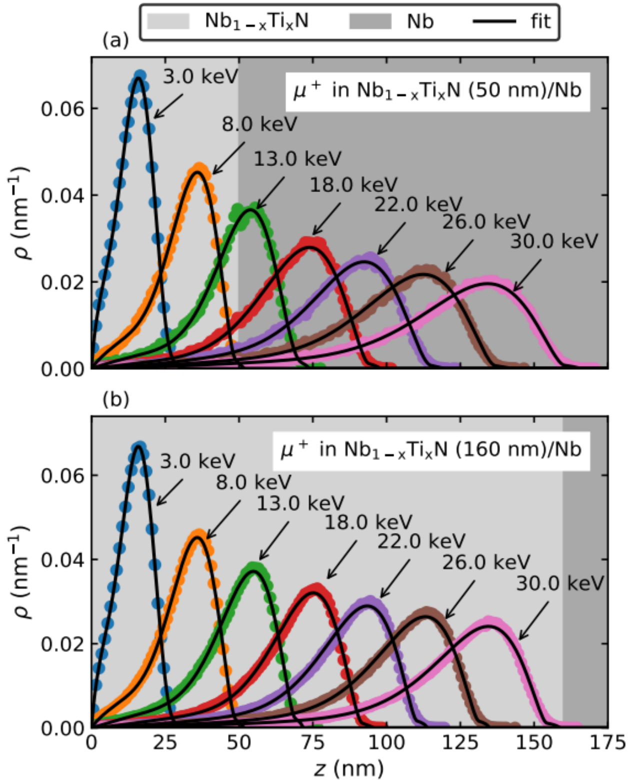


[5]



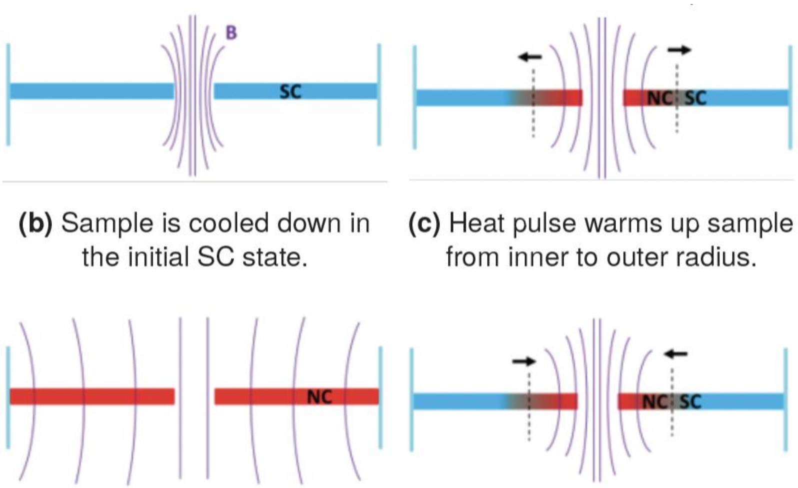
[6]

Stopping Profiles



Asaduzzaman, Md et al. [arXiv:2304.09360v1] (2023)

Flux Expulsion Measurement

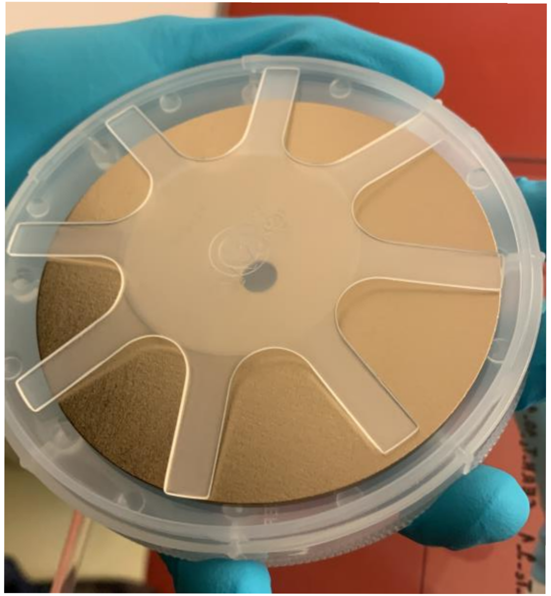


(b) Sample is cooled down in the initial SC state.

(c) Heat pulse warms up sample from inner to outer radius.

(d) Sample is warmed-up and fully normal conducting.

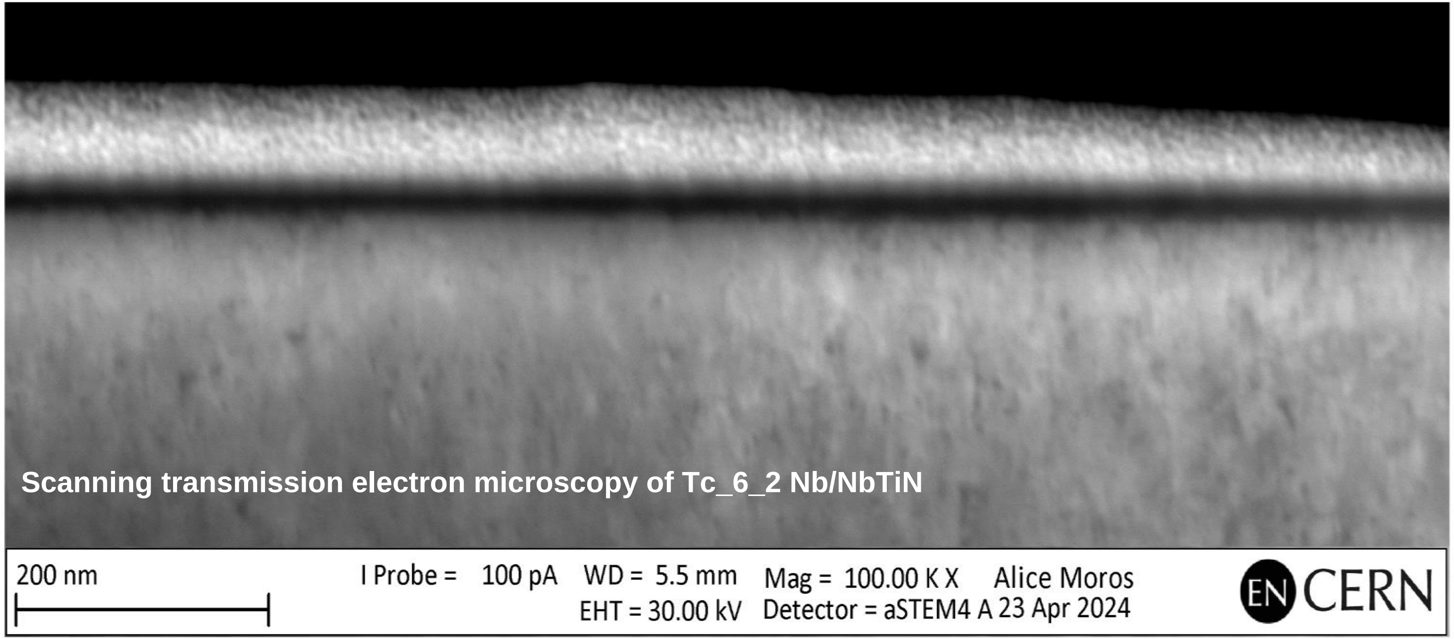
(e) Heat pulse stops and sample cools down again.



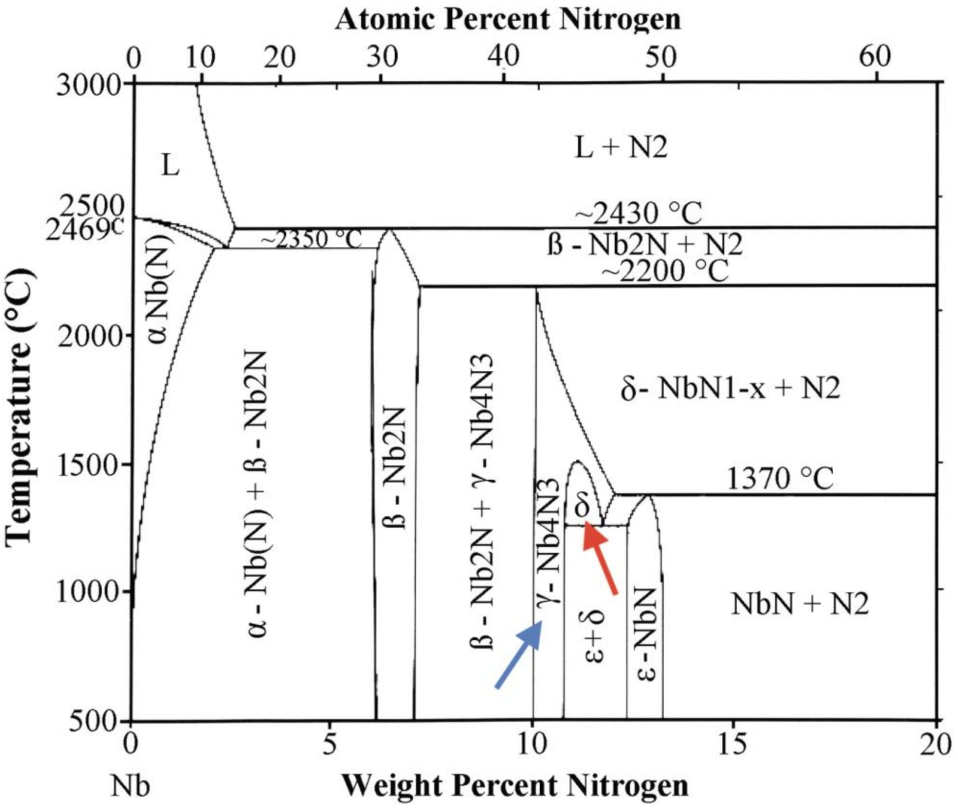
Flux Expulsion Measurement

Sample	Initial annealing T (°C)	Coated structure	I ($\sim 15\,\mathrm{nm}$)	S ($\sim 60\,\mathrm{nm}$)
DESY_0	800	-	-	-
DESY_1	900	-	-	-
DESY_2_1 DESY_2_2	800	SS	-	NbTiN
DESY_3_1 DESY_3_2	800	SIS	AlN	NbTiN
DESY_4_1 DESY_4_2	800	SIS	AlN	NbN
DESY_5_1 DESY_5_2	900	SIS	AlN	NbTiN
DESY_6_1 DESY_6_2	900	SS	-	NbTiN

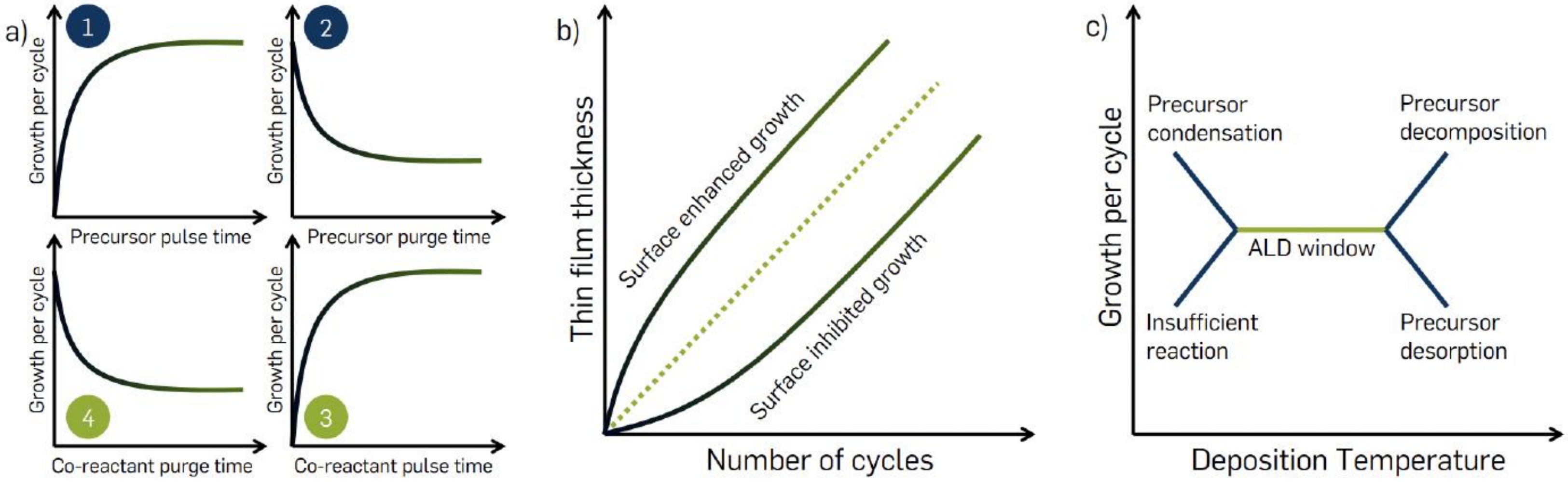
STEM Measurements at CERN



NbN Phase Diagram



PEALD



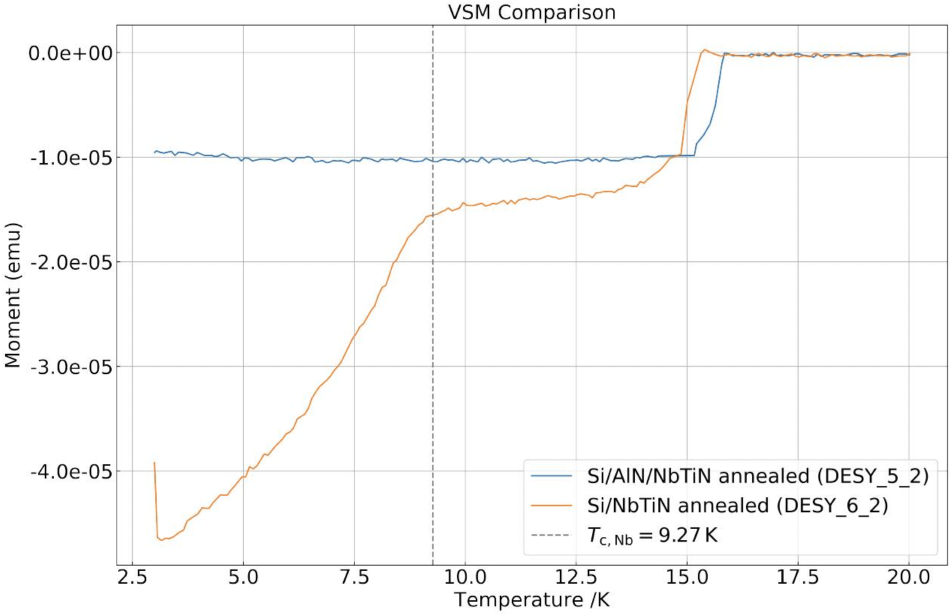
a)
1
Growth per cycle
Precursor pulse time
2
Growth per cycle
Precursor purge time
4
Growth per cycle
Co-reactant purge time
3
Growth per cycle
Co-reactant pulse time
b)
Thin film thickness
Surface enhanced growth
Surface inhibited growth
Number of cycles
c)
Growth per cycle
Precursor condensation
Precursor decomposition
ALD window
Insufficient reaction
Precursor desorption
Deposition Temperature

Deposition specifications

Precursor	TMA trimethyl-aluminum	TDMAT tetrakis (dimethylamino) titanium(IV)	TBTDEN (t-butylimido) tris(diethylamino) niobium(V)
Target Material	AlN	TiN	NbN
Precursor Pulse Duration (s)	0.021	0.5	0.5
Precursor Temperature (°C)	room temperature	70	90
1st Purge Duration (s)	30	40	40
Plasma Exposure Duration (s)	10	60	90
Plasma Flow of H_2, N_2 (SCCM)	30, 10	45, 20	45, 12
2nd Purge Duration (s)	100	120	120

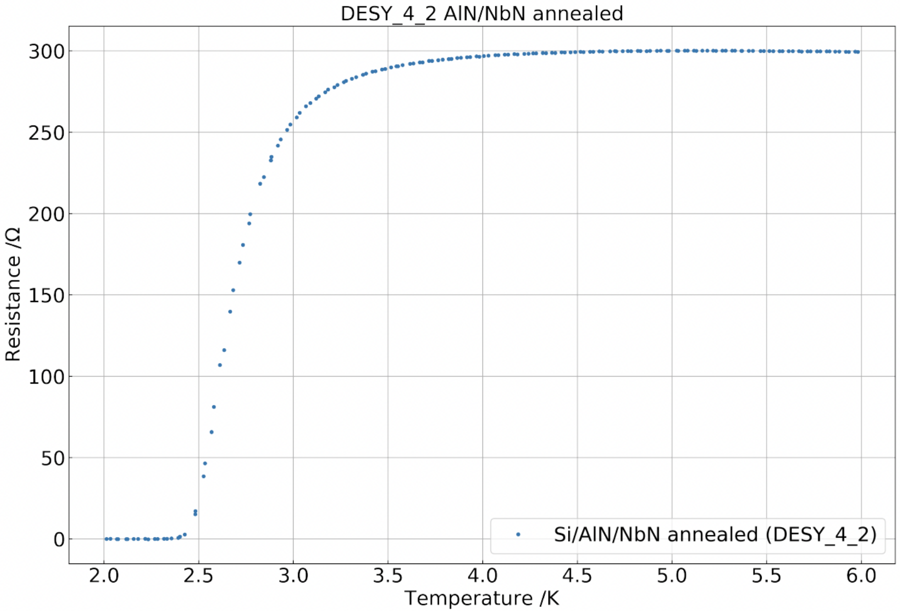
VSM T_c Measurements Results

- text



Electrical-transport T_c Measurement Result for NbN

- 900 °C annealing of SIS Nb/AlN/**NbN**
  - intended high-T_C δ-phase is **not** formed
  - Ti as stabiliser of the cubic high-T_C δ-phase
  - **NbN excluded from SIS studies**

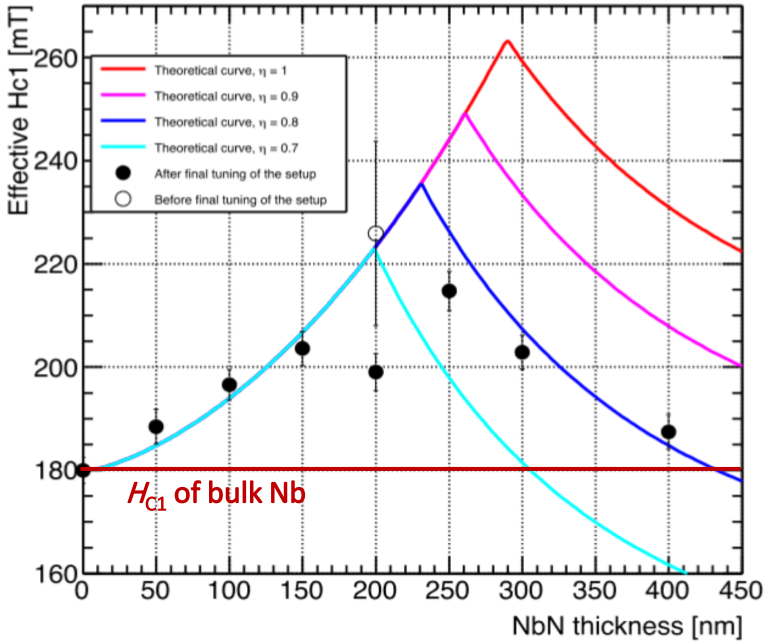


H_{C1} measurement at KEK

- **Determination of $H_{C1}(d)$ for SIS mandatory**
- **Third-harmonic voltage measurement with local magnetometry at KEK in Japan**
  - SIS coated Nb samples with varying sc layer thicknesses

Compare results with theory!

Measured H_{C1} of Nb/SiO_2/NbN samples vs. NbN thickness:



Ito, H. et al. [arXiv: 1907.03410] (2019)