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Atomic layer deposition of superconductors for SRF

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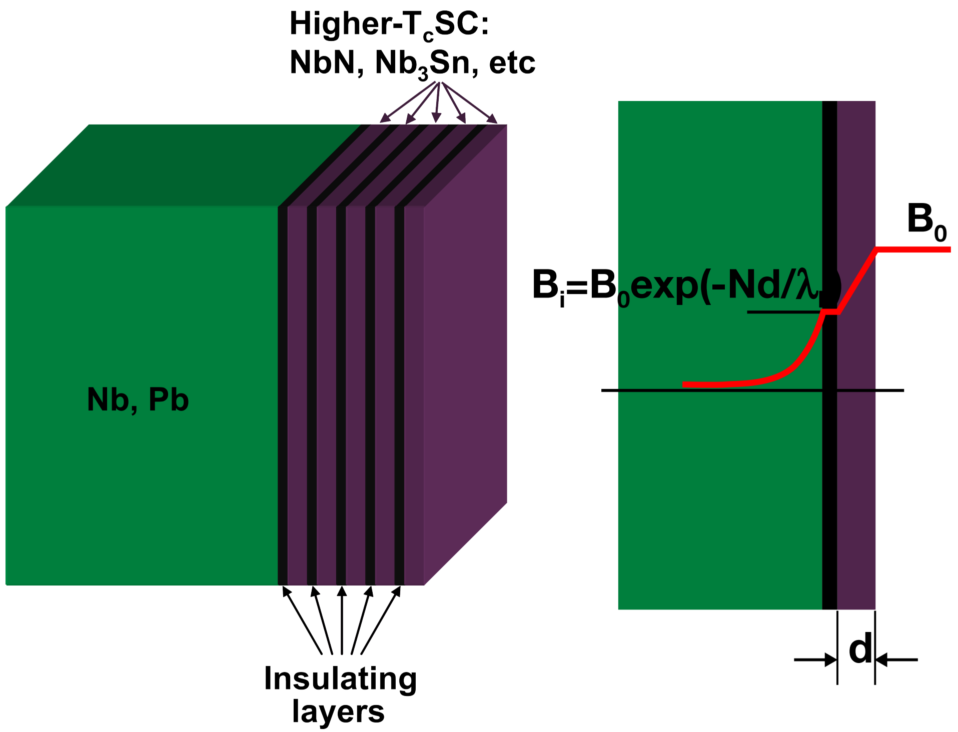
Outline

- Background
- ALD of superconductors: current status
  - Nitride superconductors (MoN, $Nb_{1-x}Ti_xN$)
  - $Nb_{1-x}Ti_xN$ / AlN superconductor-insulator multilayers
- Ongoing and future work
  - New precursors for superconducting selenides and tellurides
  - Plasma-enhanced ALD

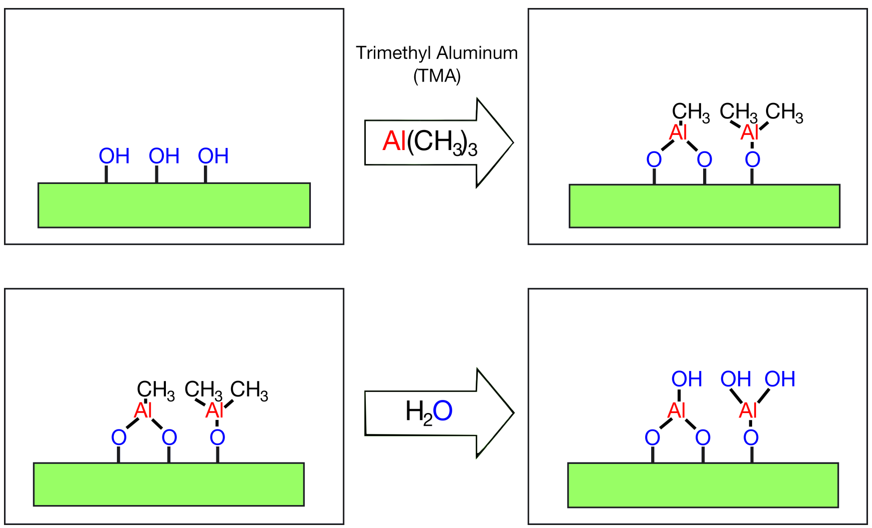
Multilayer thin films for SRF

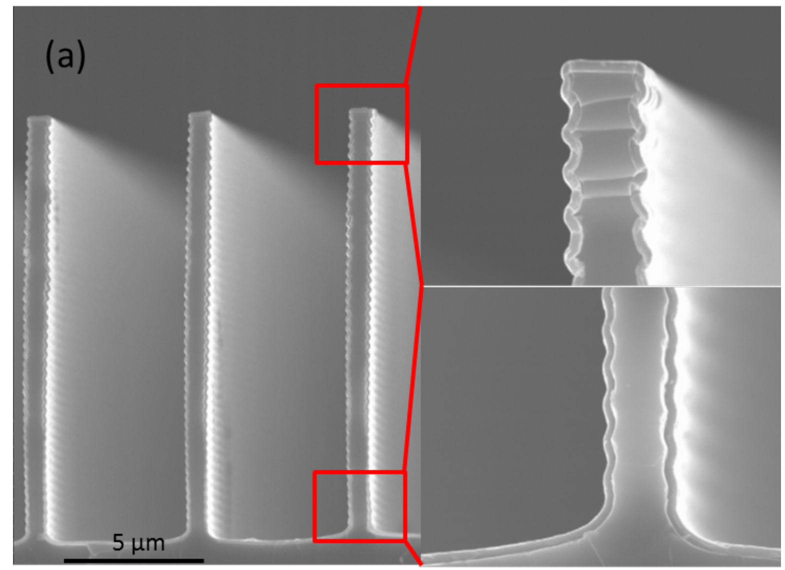
- Superconductor-Insulator multilayer [Gurevich, *Appl. Phys. Lett.* **88**, 012511 (2006)]
  - Use material with larger gap than Nb (lower surface resistance)
  - Exploit thin film B_{c1} enhancement ($d < \lambda_L$) to prevent vortex dissipation
  - Use N higher-T_c SC layers separated by insulating layers to screen field
- Increase performance
  - Move beyond limits of Nb
- Decrease cost
  - Higher operating temperature (reduce cryogen costs)
  - Replace bulk Nb with cheaper material (Cu/Al)
- Coat inside Nb SRF cavity with precise, layered structure



Atomic layer deposition (ALD)

A thin film synthesis process based on sequential, self-limiting surface reactions between vapors of chemical precursors and a solid surface to deposit films in an atomic layer-by-layer manner.

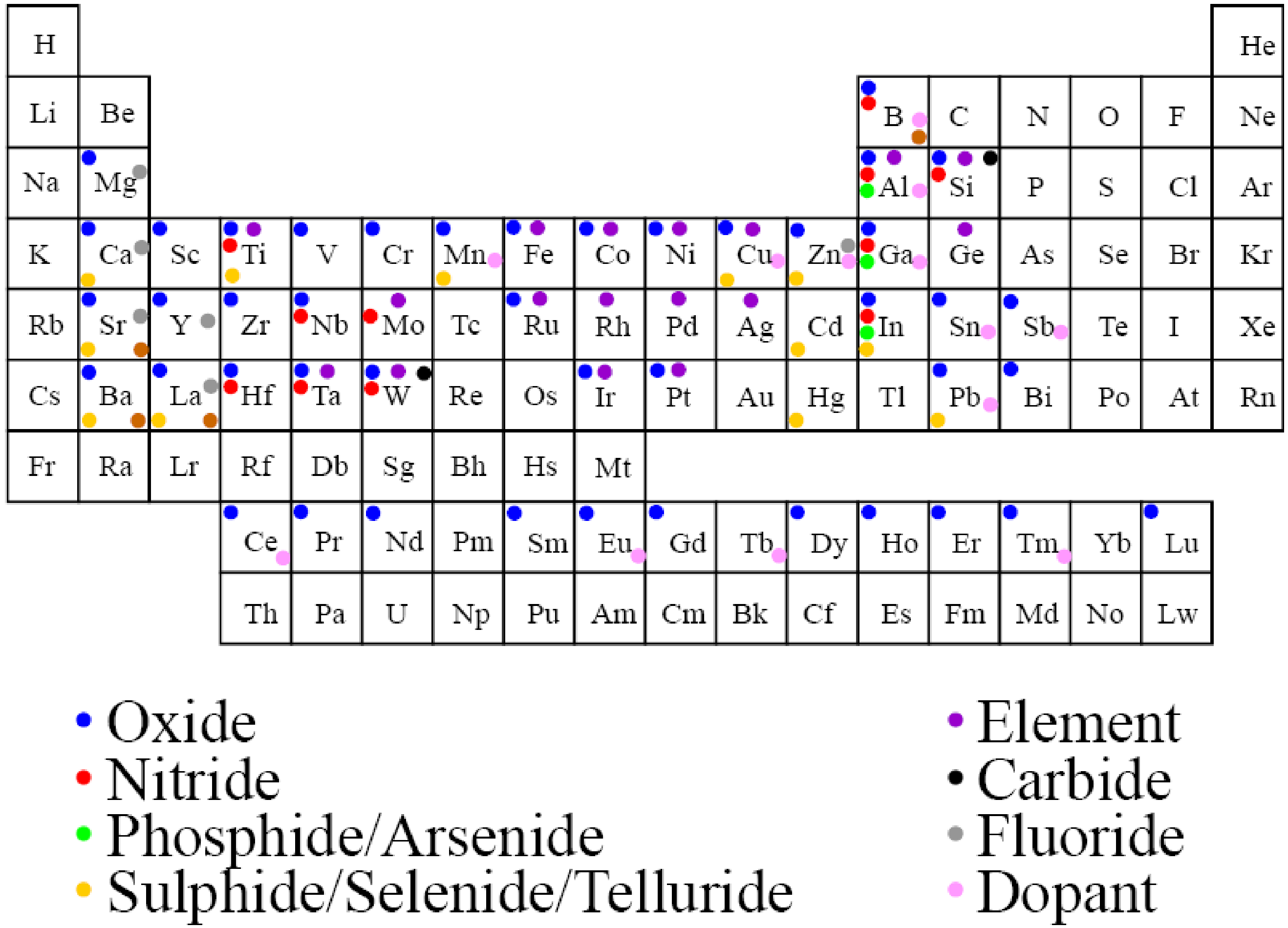




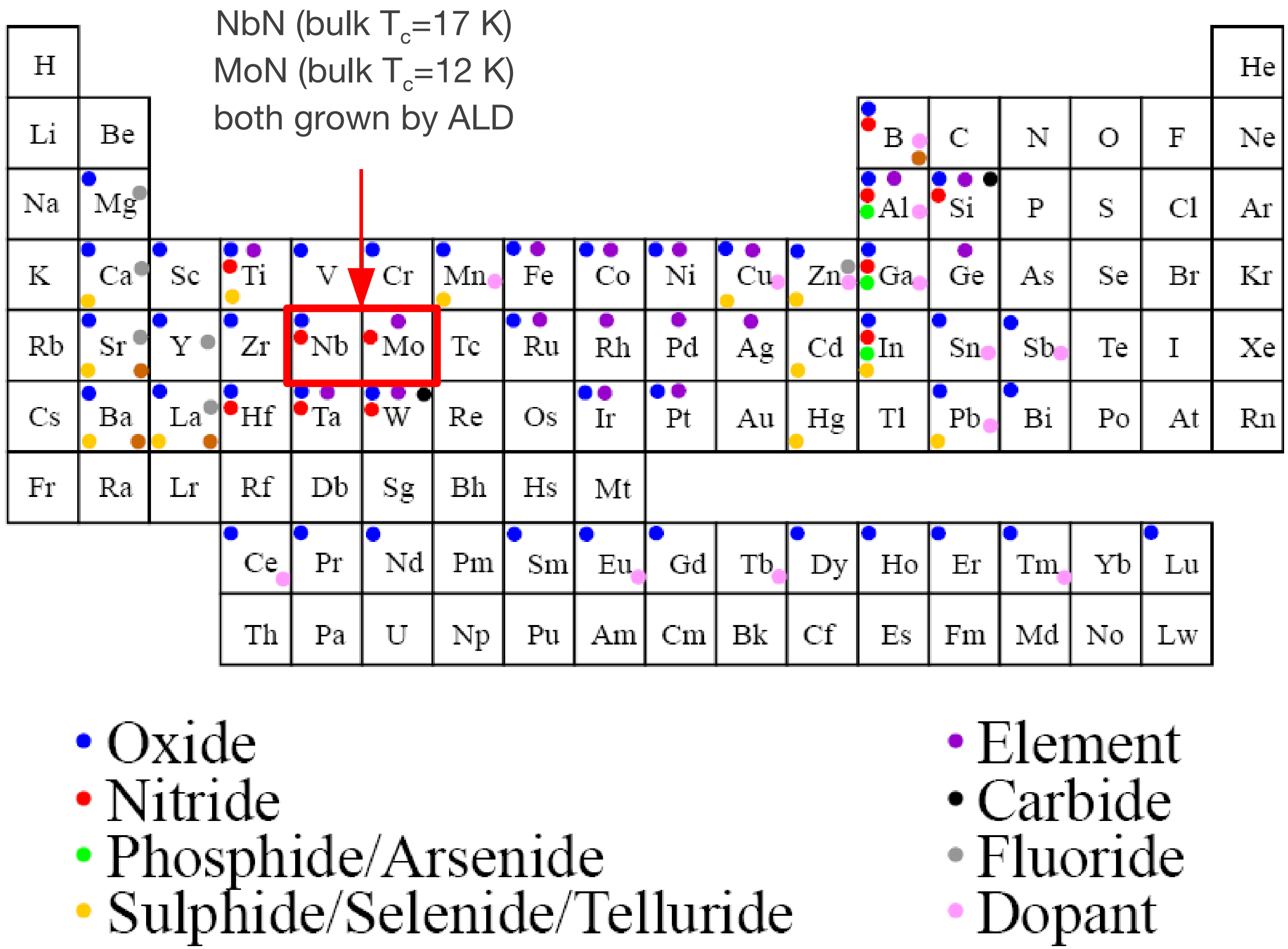
Advantages:

- Atomic-level control of thickness and composition
- Smooth, continuous, pinhole-free coatings on large area substrates
- No line-of-sight limits → excellent conformality over complex shaped surfaces

ALD thin film materials



ALD superconductors?



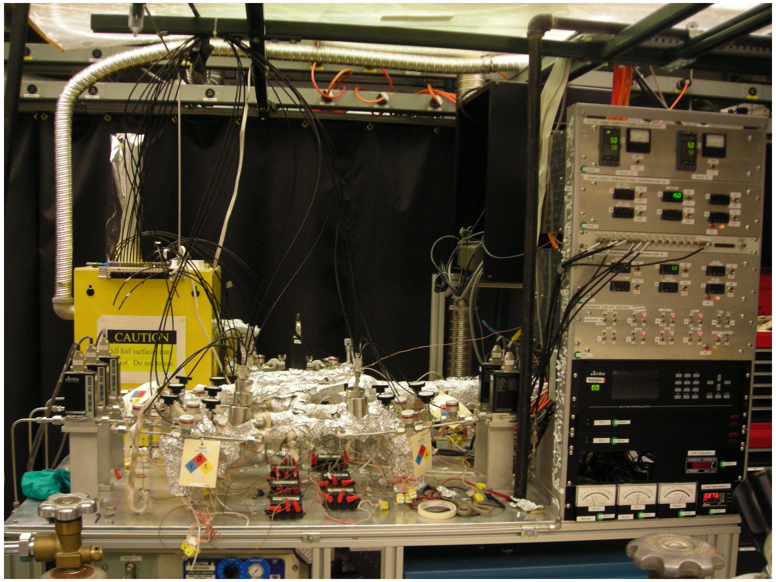
Except in one paper, superconductivity has been ignored...

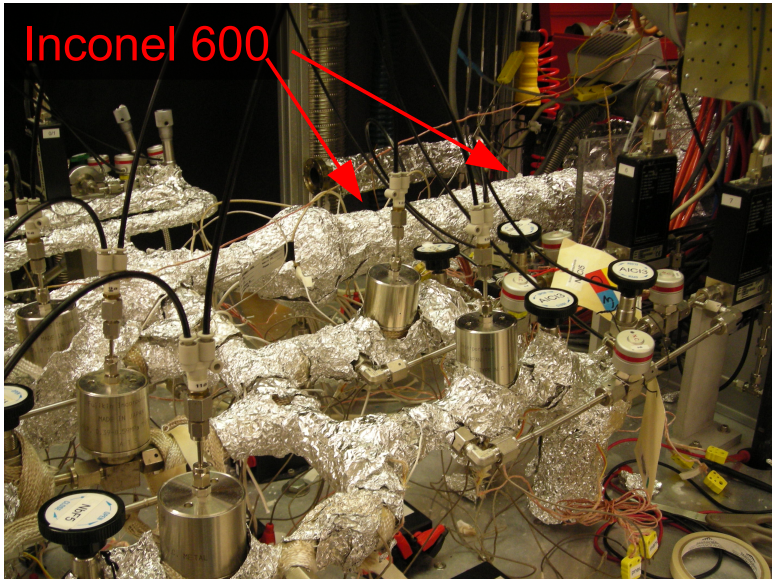
- Reported T_c = 10 K for NbN [Hiltunen, *et al.*, *Thin Solid Films* **166**, 149 (1988)]

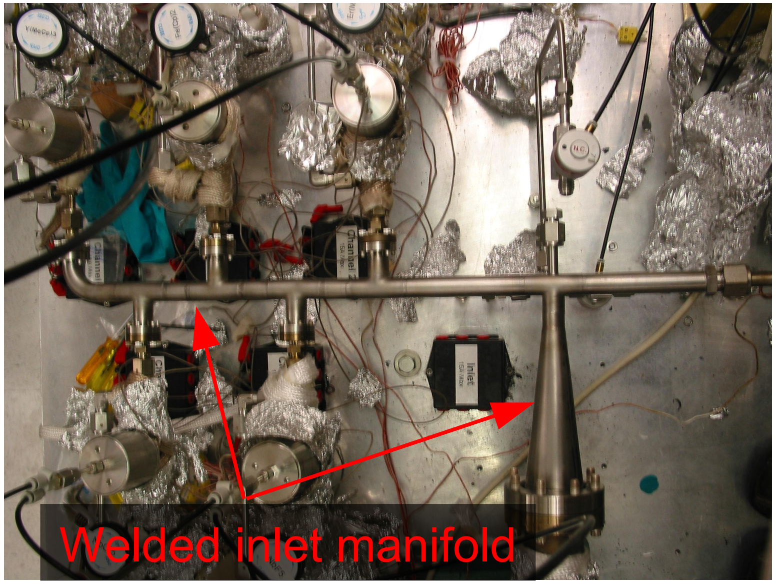
Argonne ALD Reactor 1

Key features:

- Inconel 600 reactor tube (corrosion resistance superior to 316 SS)
  - Halide precursors ($NbCl_5$, $TiCl_4$, etc.)
- All-welded precursor inlet manifold (reduced sites for potential leaks)
  - Oxygen contamination in nitride films







Thin film characterization

ANL resources

- X-ray photoemission spectroscopy (XPS)
- X-ray reflectivity (XRR)
- X-ray diffraction (XRD)
- Synchrotron grazing-incidence x-ray diffraction (GIXRD) — **APS**
- Scanning electron microscopy (SEM)
- Transmission electron microscopy (TEM)

  (SEM and TEM) — **EMC**
- DC electrical transport (down to 1.6 K)
- SQUID magnetometry

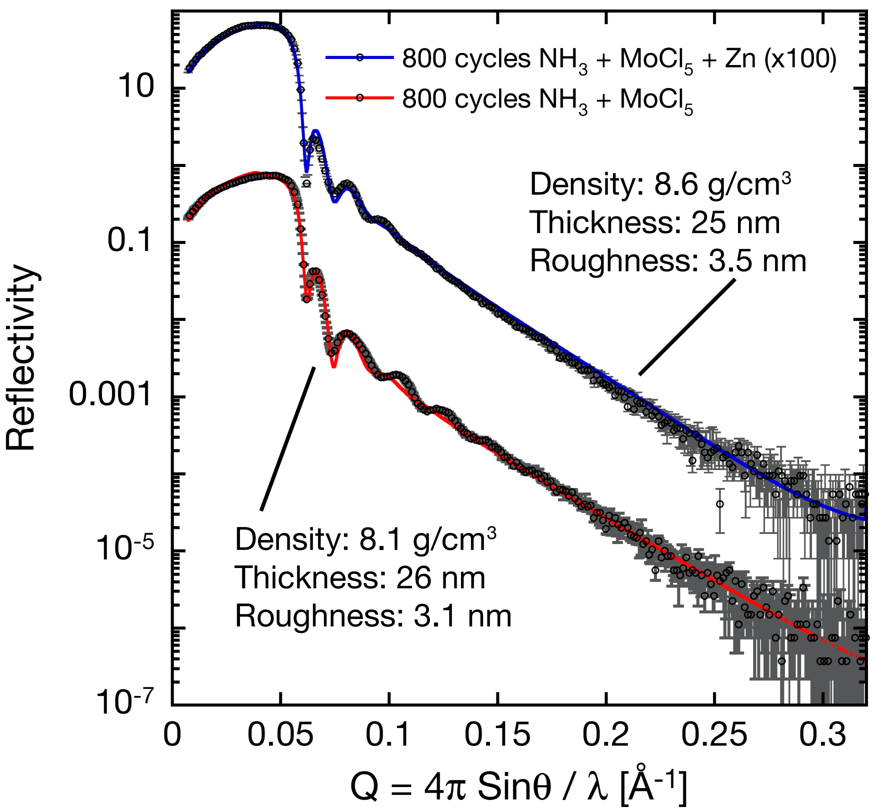
Outside ANL

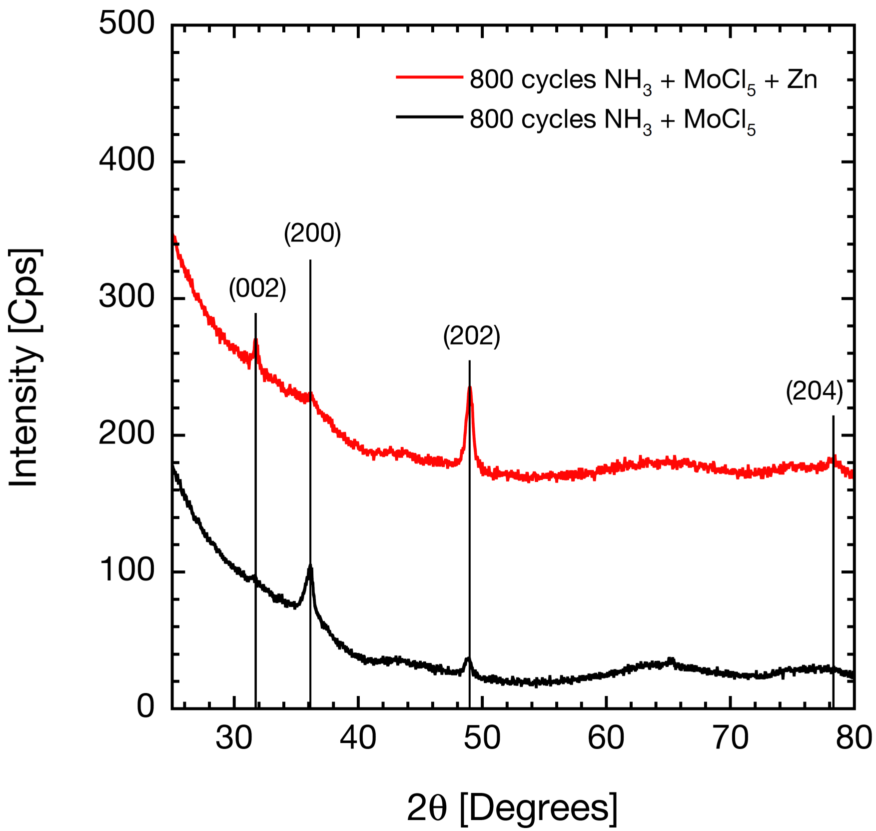
- Atom probe tomography (APT) [Seidman, NU]
- Rutherford backscattering spectroscopy (RBS) [Evans Analytical]

Molybdenum nitride: MoN

Effects of intermittent Zn pulse

- Chemistry: $MoCl_5 + NH_3$ / $MoCl_5 + Zn + NH_3$ at 450°C
- Hexagonal MoN in both cases, higher density & change in texture with Zn

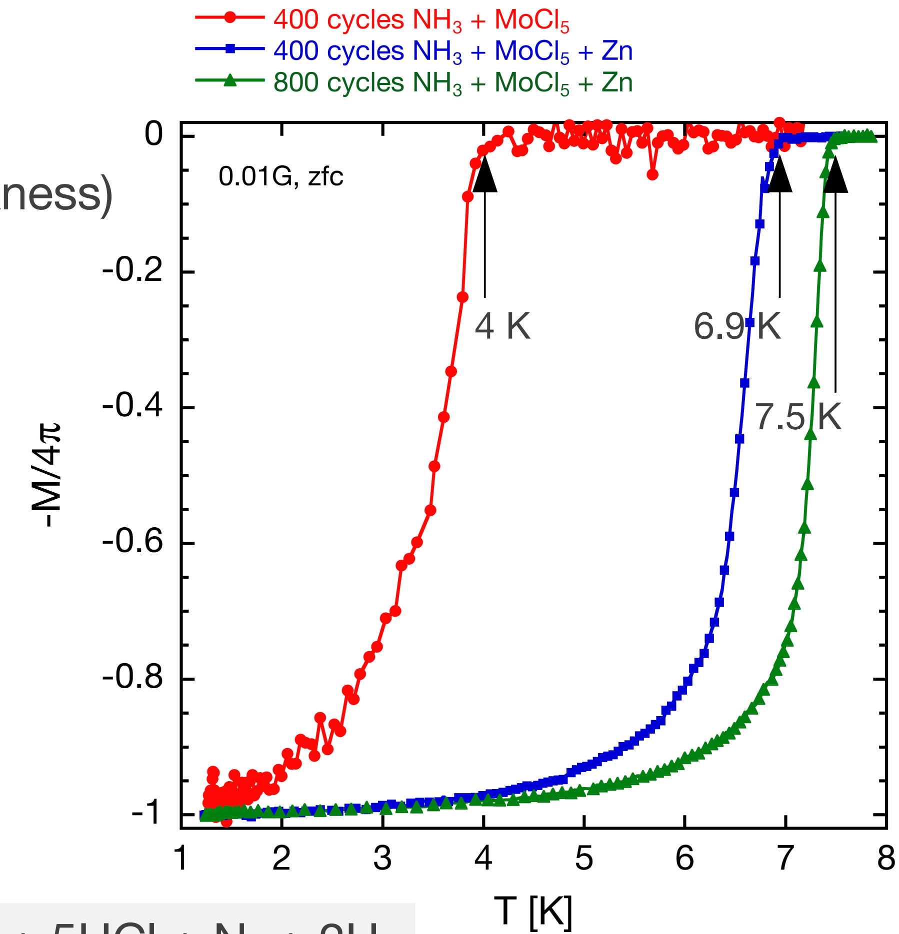




MoN: Superconducting T_c (SQUID)

Addition of Zn leads to:

- ~2x increase in T_c (equivalent thickness)
  - Peak T_c = 7.5 K for 25 nm film
- Decrease in RT resistivity
  - 200 μΩ-cm without Zn
  - 120 μΩ-cm with Zn



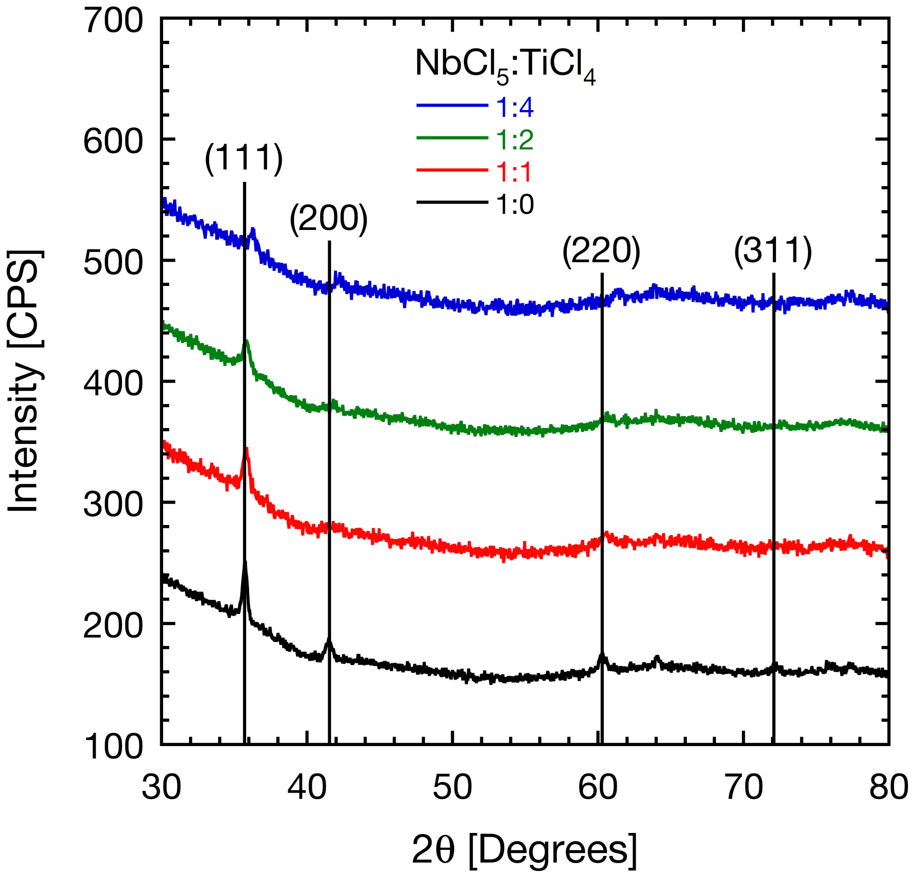
- No chlorine, zinc observed by XPS
- Could be related to film density
  - 88-93% of bulk (9.2 g/cm^3)
- Could be due to hydrogen:

Without Zn: $MoCl_5 + 3NH_3 \rightarrow MoN + 5HCl + N_2 + 2H_2$

With Zn: $MoCl_5 + Zn + NH_3 \rightarrow MoN + ZnCl_2 + 3HCl$

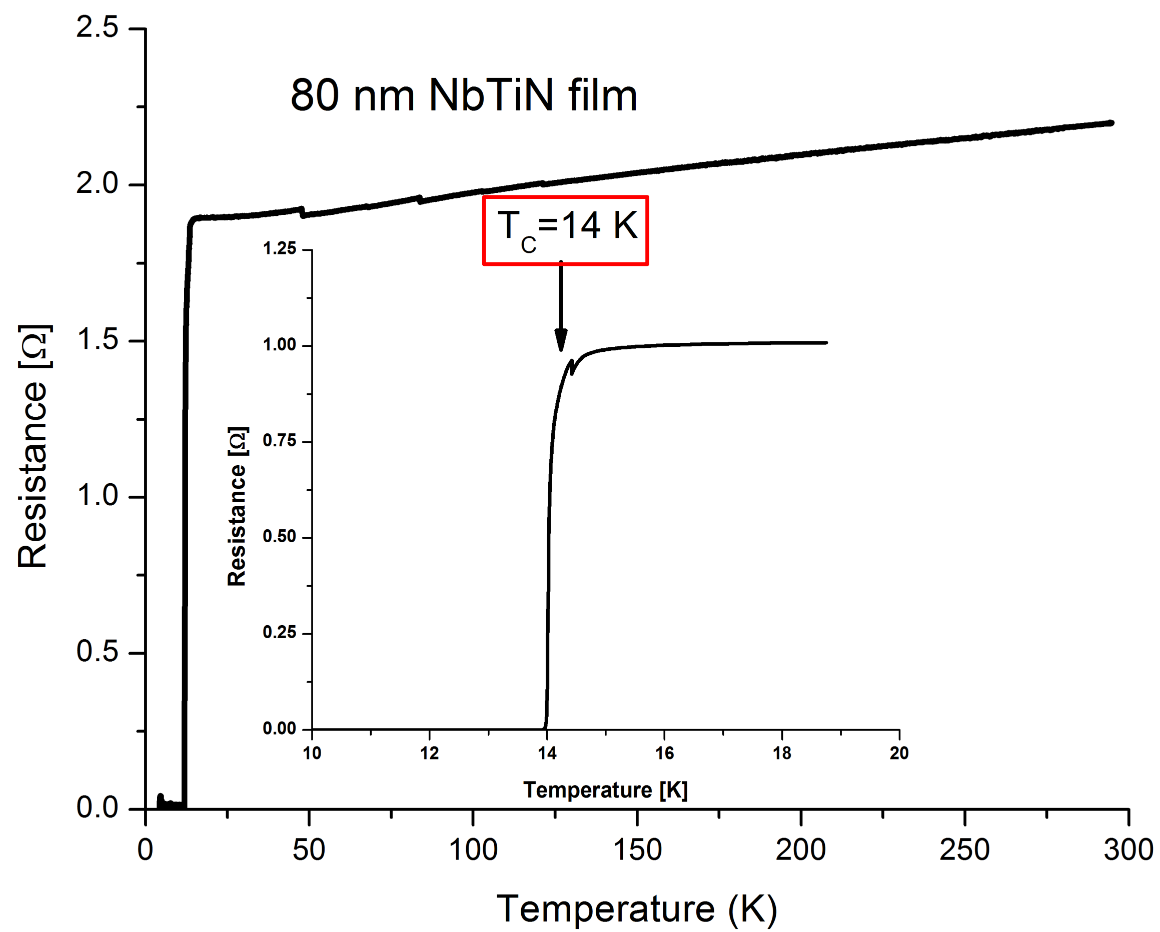
Niobium titanium nitride: $Nb_{1-x}Ti_xN$

- Chemistry: $(NbCl_5:TiCl_4) + Zn + NH_3$ at 450°C, 500°C
- Can vary Ti content with $NbCl_5:TiCl_4$ ratio (1:2 ~ 20% TiN)
- Cubic δ phase, peaks shift to higher angle with increasing TiN
- Density decreases with increasing TiN
  - 7.2 g/cm³ (1:0)
  - 5.7 g/cm³ (1:4)
- RT resistivity decreases with increasing TiN
  - 380 μΩ-cm (1:0)
  - 130 μΩ-cm (1:4)
- Chlorine content: 0.05 atom %
- Are they good superconductors?



Optimized growth of $Nb_{1-x}Ti_xN$

- Achieved superconducting T_c=14 K, ***40% higher than any other ALD film***
- Nearly 5 K higher than Nb



$Nb_{1-x}Ti_xN$-based S-I structures

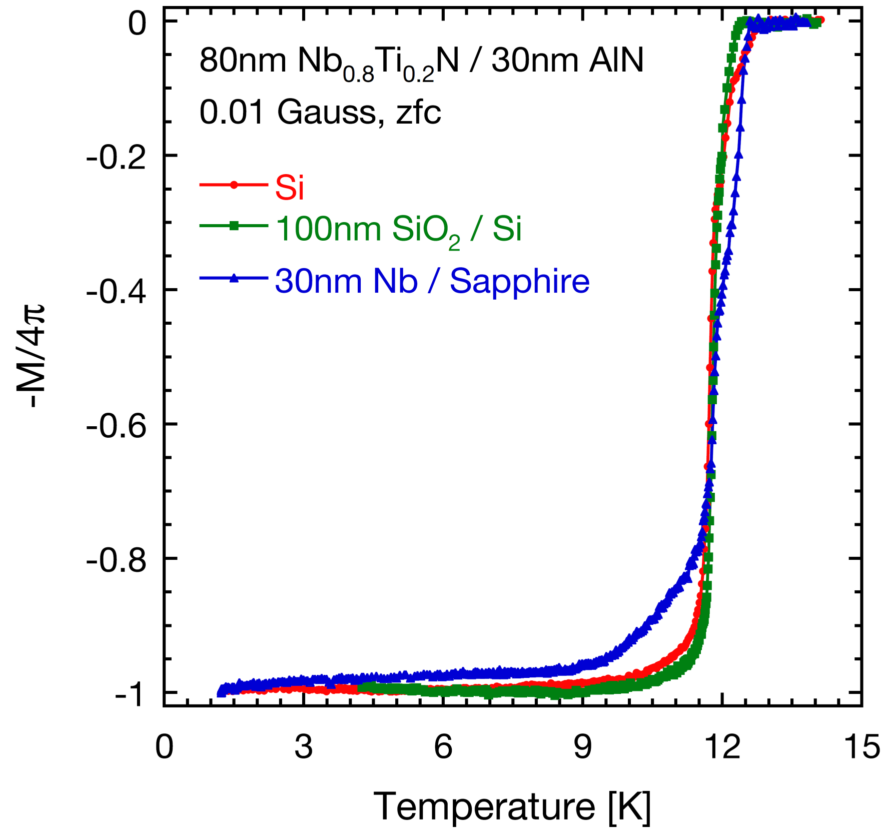
Aluminum nitride: AlN

- Oxygen-free insulator, stable interface with Nb(Ti)N
- Similar structure to Nb(Ti)N
  - 0.27% mismatch between in-plane spacing of (0001)-oriented AlN and (111)-oriented NbN
- Can be grown with $AlCl_3$ and NH_3 at same temperature as Nb(Ti)N
  - No thermal cycling between deposition steps
  - ALD previously demostrated [K.-E. Elers, et al. *J. de Phys. IV* **5** (1995)]

- NbN/AlN multilayers grown previously by sputtering
  - Enhanced J_c at high fields [J.M. Murduck, et al. *Appl. Phys. Lett.* **62** (1988)]
  - Model system for vortex matter in HTS [E.S. Sadki, et al. *Phys. Rev. Lett.* **85** (2000)]

$Nb_{1-x}Ti_xN$ / AlN heterostructures

- 40 nm $Nb_{0.8}Ti_{0.2}N$ / 15 nm AlN (single bilayer and double stack)
- 80 nm $Nb_{0.8}Ti_{0.2}N$ / 30 nm AlN (single bilayer and double stack)
  - Quartz, Si(001), 100 nm SiO_2/Si(001), 30 nm Nb/Sapphire, and cavity-grade Nb



Optimized $Nb_{1-x}Ti_xN$/AlN ALD growth process (T_c = 14 K) is now ready for coating Nb SRF cavities

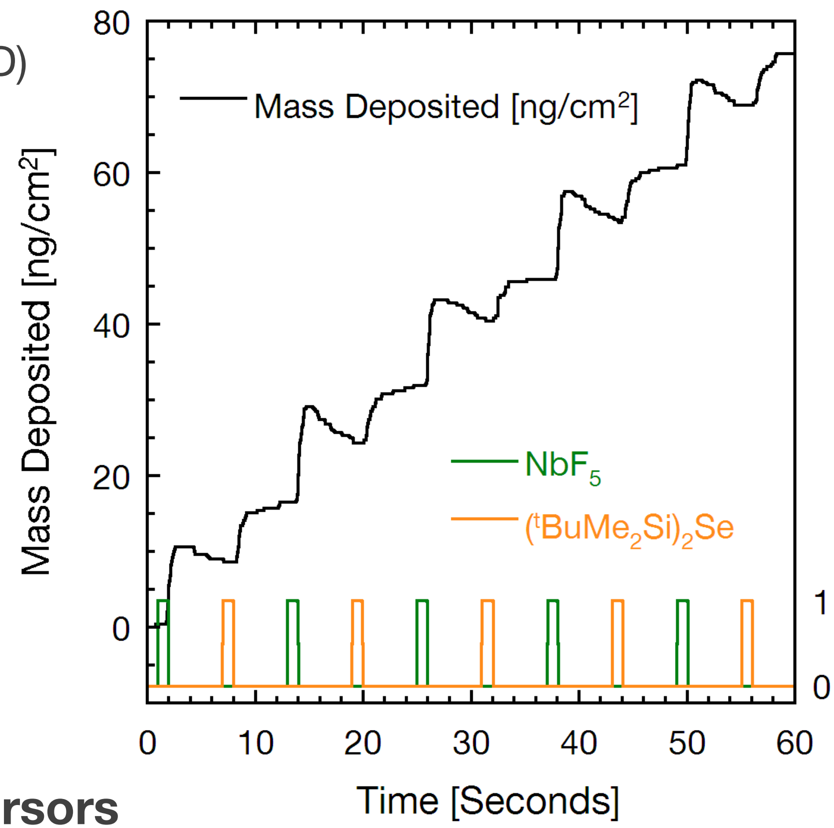
- Will enable testing the effects of S-I multilayer on cavity performance

Alkylsilyl precursors for Se, Te compounds

$$(R_3Si)_2Te(g) + MCl_2(g) \rightarrow MTe(s) + 2R_3SiCl(g) \; ^*$$

New custom precursors (J. Schlueter, MSD)
- $(Et_3Si)_2Te$ / $(Et_3Si)_2Se$
- $(^tBuMe_2Si)_2Te$ / $(^tBuMe_2Si)_2Se$

Promising for ALD growth of new Fe-based superconductors ($FeSe_{1-x}Te_x$)
- T_c reported up to 37 K
- Remain superconducting in high magnetic fields (>45 T)



Initial studies of ANL synthesized precursors
- Growth of $NbSe_x$ with $NbF_5/(^tBuMe_2Si)_2Se$

Plasma-enhanced ALD (PEALD)

- New PEALD system (Cambridge NanoTech Fiji200)
  - Commissioned and in use
- Plasma-enhanced:
  - Increase reactivity with atomic radicals
- Advantages of PEALD:
  - Higher purity films
  - More dense (different phases, better crystallinity)
  - Easier to grow pure metals

- Use new commercial Plasma-enhanced ALD (PEALD) system to develop processes for growth of superconducting Nb metal and other compounds which cannot be grown by traditional (thermal) ALD, e.g. MgB_2 (T_c = 40 K)

Summary

- Growth of single-phase hexagonal-MoN at 450°C
- Demonstrated ~2x increase in T_c in MoN with intermittent Zn dose ($MoCl_5$ + Zn + NH_3)
- Optimized growth of $Nb_{1-x}Ti_xN$ to achieve superconducting T_c = 14 K, 40% higher than any other ALD film and ~5 K higher than Nb
- Demonstrated successful ALD growth of $Nb_{1-x}Ti_xN$/AlN S-I multilayers on flat substrates (Si, SiO_2, Sapphire, Nb)
- New precursors for Fe-based superconductors ($FeSe_{1-x}Te_x$)
- Plasma-enhanced ALD system now online and in use