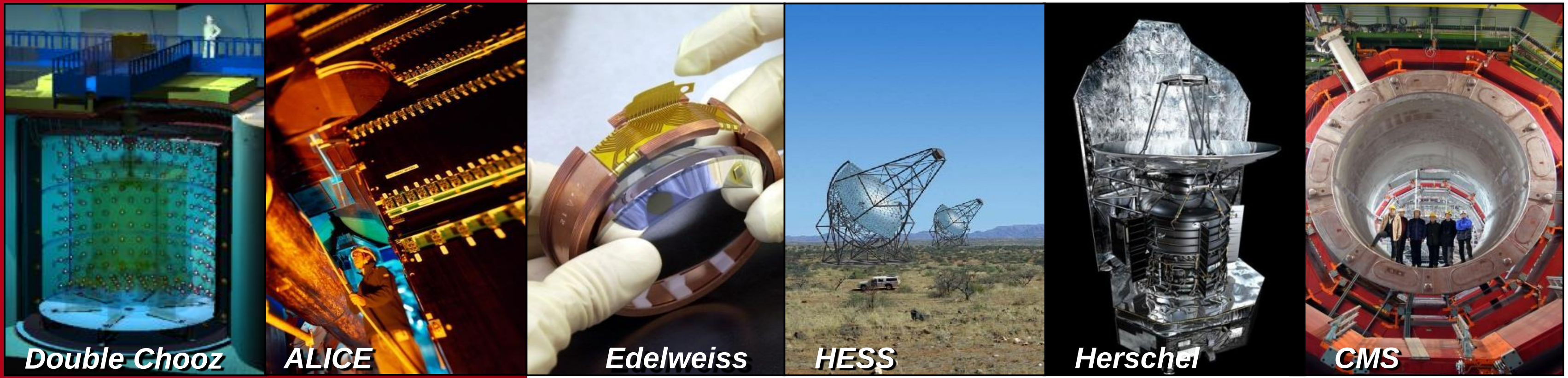
DE LA RECHERCHE À L'INDUSTRIE

# Atomic Layer Deposition:
# An innovative approach for next generation particle accelerators

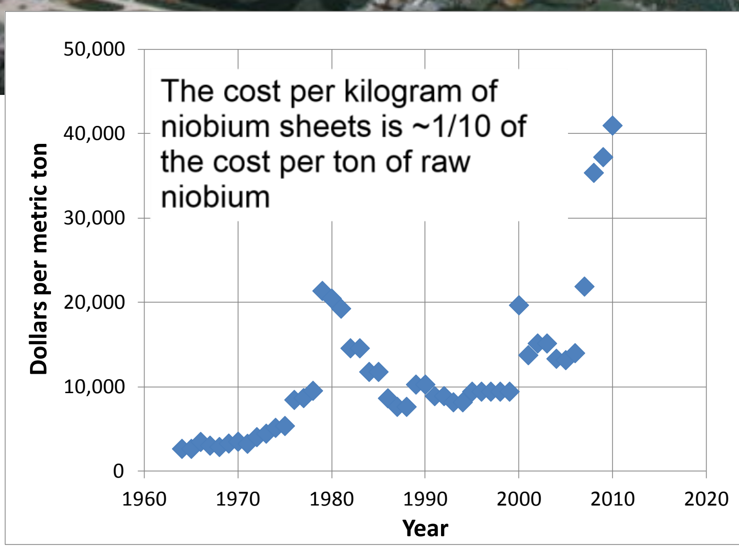



*Déchiffrer les rayons de l'Univers*

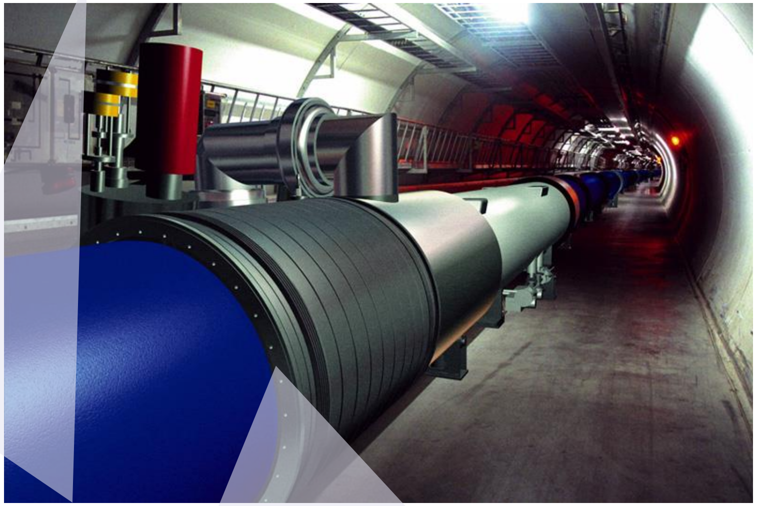
Thomas Proslier
Claire Antoine

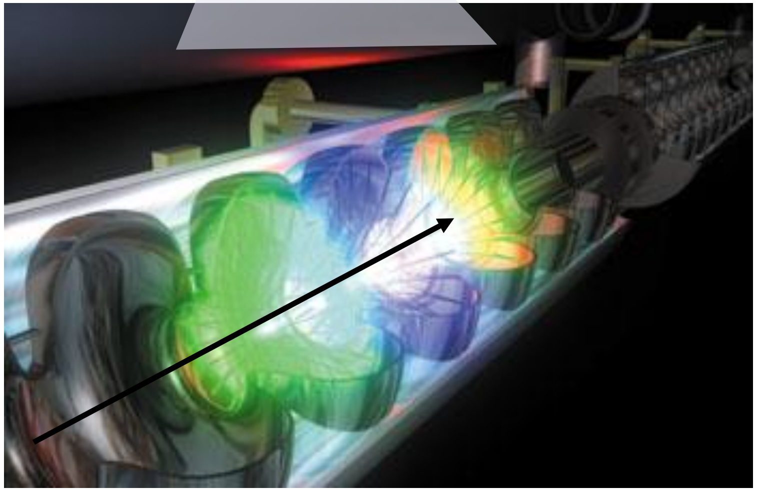
# Particle accelerators

## CERN




## ILC = 11 km





# Atomic Layer Deposition (ALD)

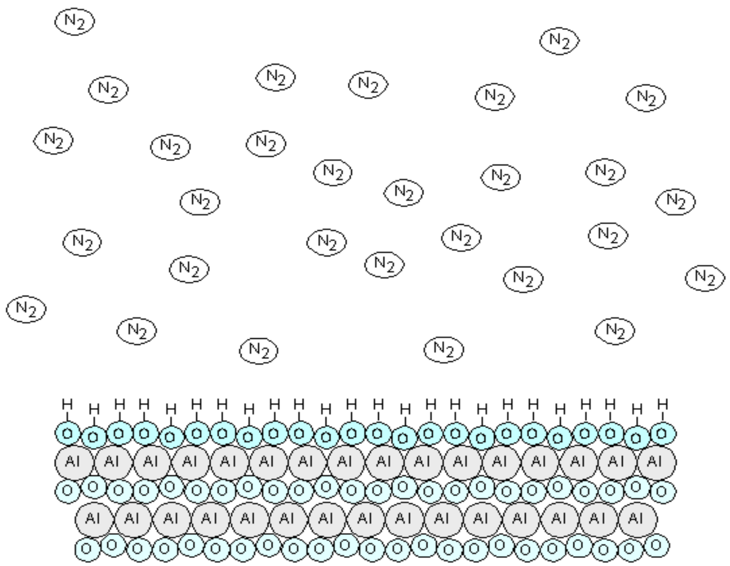
ALD is a thin film synthesis technic based on sequential, self limiting surface chemical reactions between precursor in the gas phase. It is a layer by layer deposition method.

**Pros**:

- Control at the atomic level of thickness and chemical composition
- Films are smooth , continuous, and pinhole free on large surfaces
- Excellent conformality complex geometrical structure, aspect ratio up 1:10 000.
- Large Palette of available materials

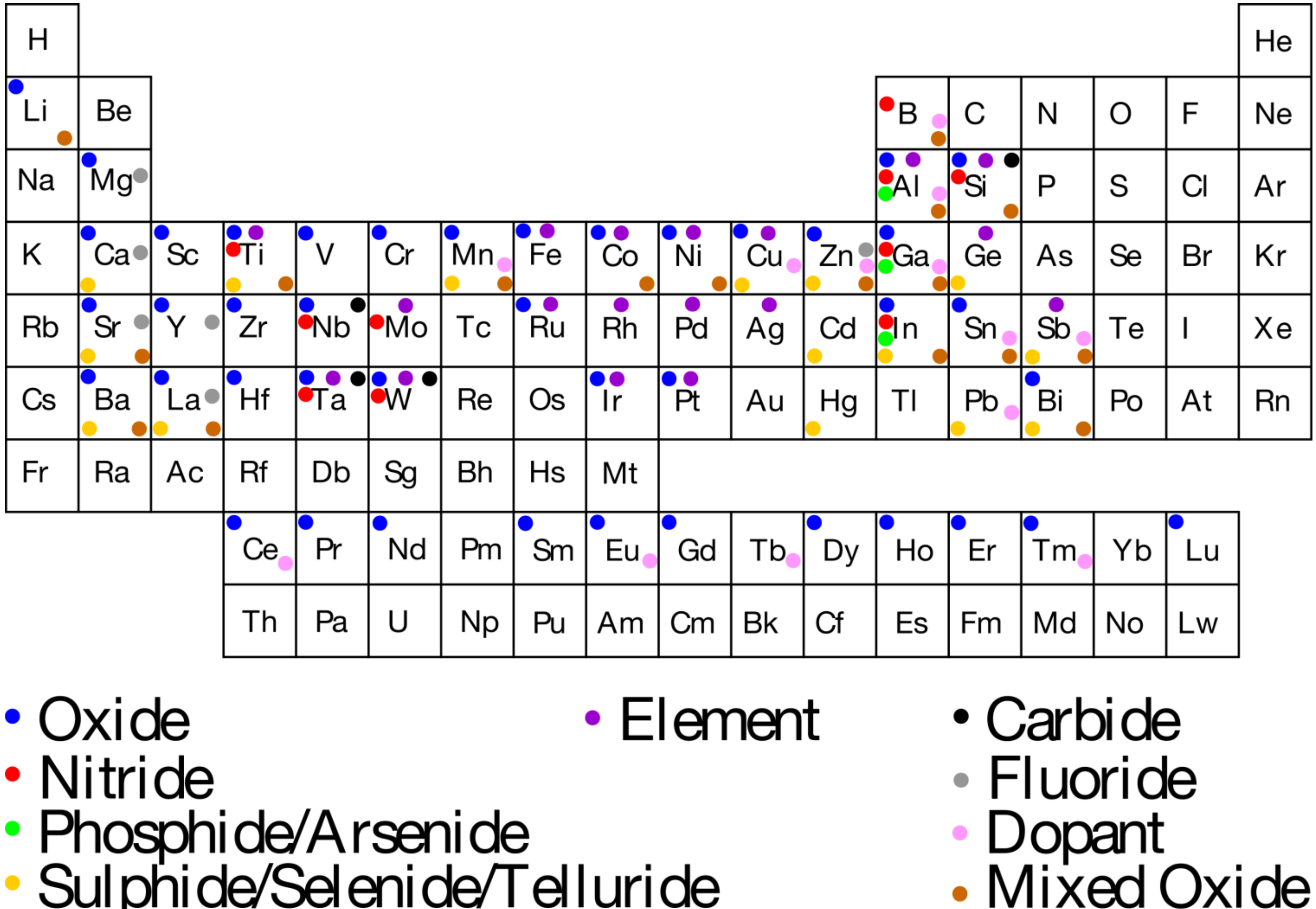
**Limits**:

- Slow growth ~ 1 Å/cy
- New materials require new chemistries.



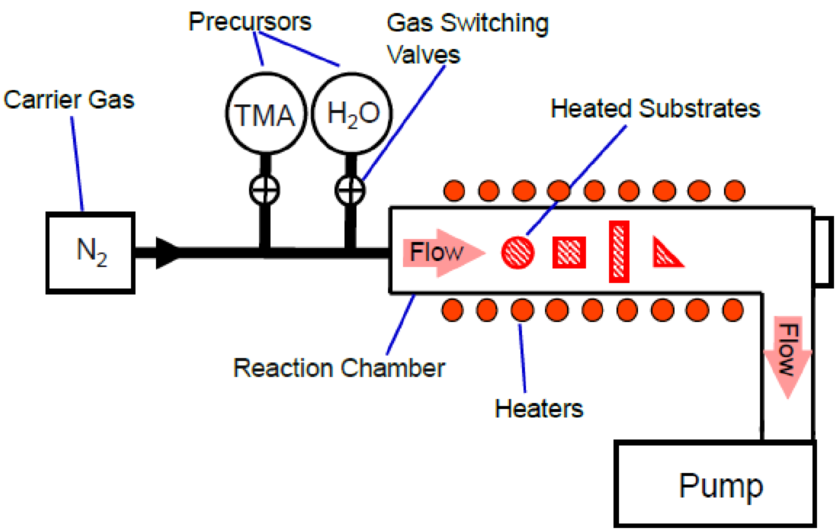

$$Al_2O_3 = TMA(Al(CH3)_3) + H_2O$$

# ALD broad Palette of Materials




See: Miikulainen et al., *J. Appl. Phys.* **113**, 021301 (2013).
Puurunen, *J. Appl. Phys.* **97**, 121301 (2005).

# ALD apparatus: from research to industry



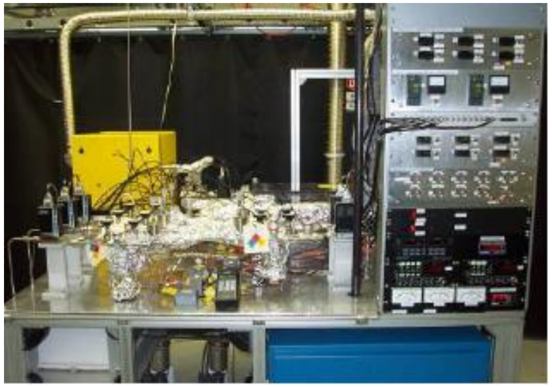

Cluster tool ~1 M€

(Avisa Pantheon)

Small ALD~ 75k€

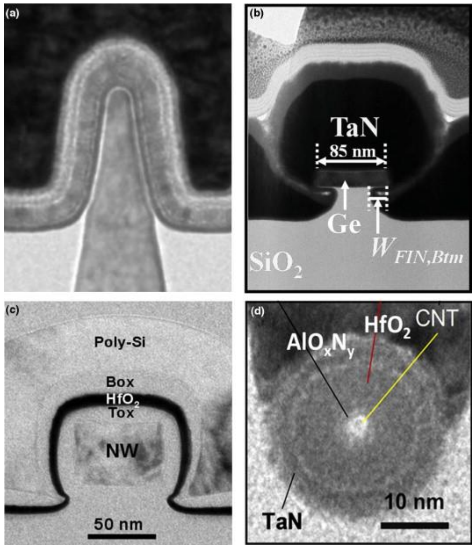
- Temperature ~ TA et 450-500°C
- Pression ~ 1 mbar – laminar flow
- Neutral gas $N_2$ ou Ar
- Précursors: solid, gas or liquid
- Substrats: porous, powders, flats…
- In-Situ Characterisation:
  - Thickness (quartz microbalance)
  - Gas analysis (mass spectrometer)

Home made ~ 100-150 k€

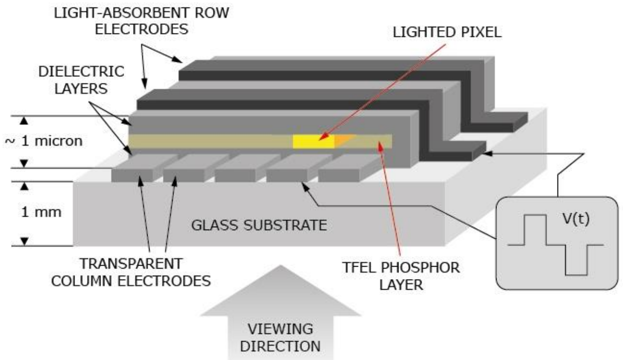


# ALD - Applications

- Microelectronic (high ε thin films): $HfO_2$, $Ta_2O_5$, $La_2O_5$, $Al_2O_3$
- Photovoltaïc (Transparent Conducting Oxide): ZnO:Al, InSnO, PbS
- Biomedical: TiN, ZrN, CrN, AlTiN
- Photonic Crystals: ZnO, ZnS:Mn, TiO2, $Ta_2N_5$
- Batteries: $Al_2O_3$, $LaF_3$, $SnF_2$…
- Electroluminescence: SrS:Cu, ZnS:Mn, ZnS:Tb, SrS:Ce
- Detectors (MCP's, gases…)
- Catalysis: Pt, Ir, Co, $TiO_2$, $V_2O_5$
- Thermoelectric: $Bi_2Se_3$, $Bi_2Te_3$…
- Diffusion barrier/anti-corrosion: $ZrO_2$, TiN…
- Supraconductors: MoN, NbTiN, TiN…
- LED: AlGaN/GaN

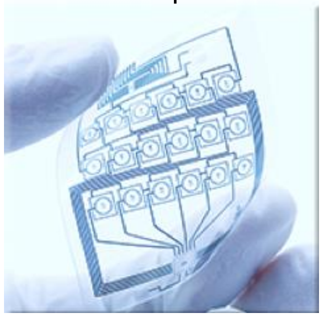



R.W. Johnson et al., *Materials Today* **17**, 236 (2014).

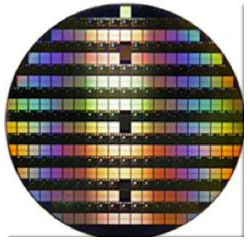



(LUMINEQ, Beneq Oy, Finland)

Electronique flexible



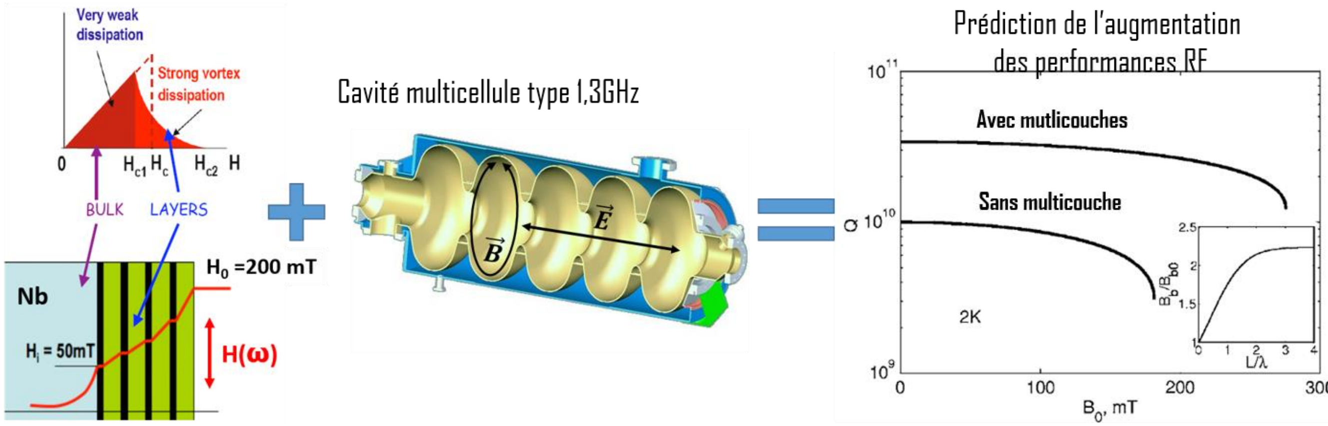
Films diélectrique haut κ



Passivation OLED

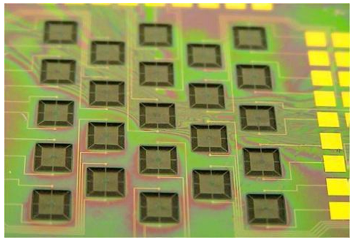
# ALD Applications – Superconductors

- Multilayers: screening magnetic field for SRF cavities – increase current density $J_C$



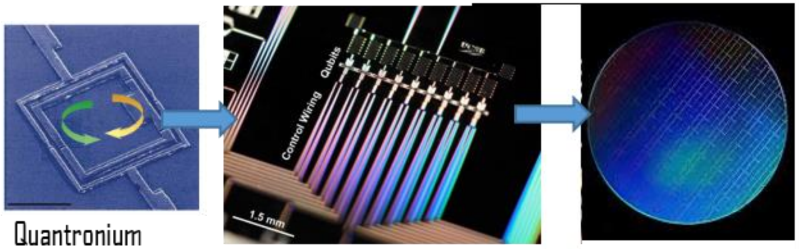

- Bolometers:

  - Thin superconducting films. Control Tc et de ΔTc
  - Cosmological Background Radiation (CMB), dark matter



- Qubits & quantum computing:

  - New superconducting alloys: Nitrides.
  - New applications: quantum phase jump junctions, charge interference junctions, cinetic inductance…




Quantronium

Google device – 9 Qubits

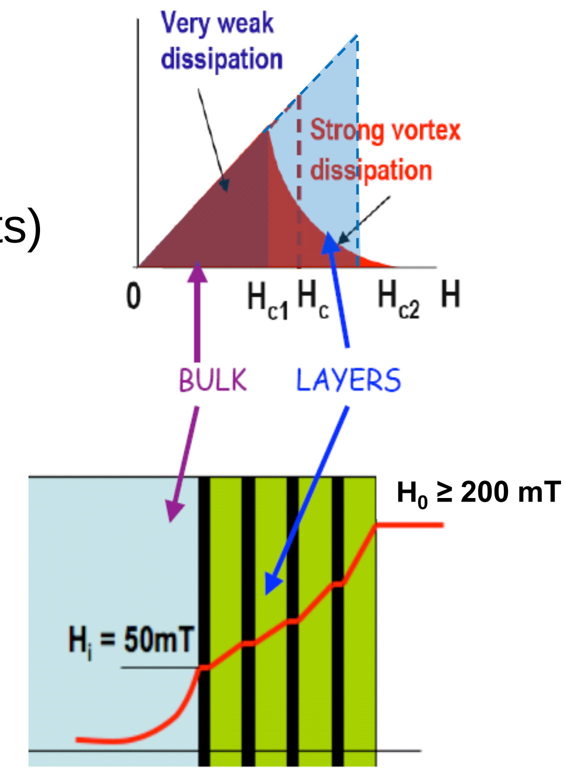
Intel Spin Qubit chip

# Research thrust 1: Multilayers

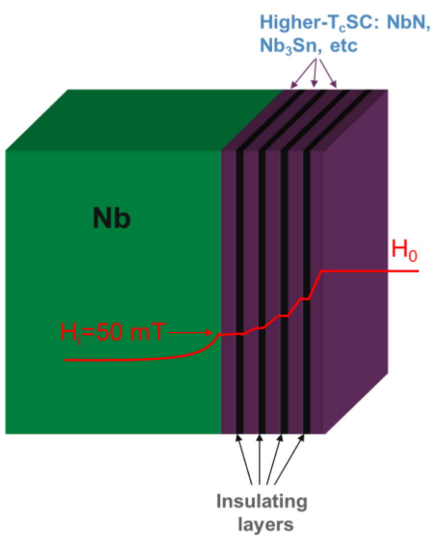
Fields in bulk Nb cavities approaching dc depairing limit for Nb, $H_c(0) \approx 200$ mT

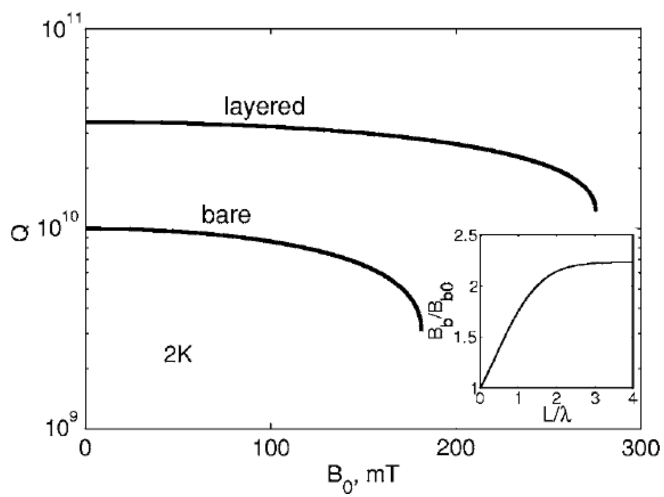
Superconductor-Insulator multilayer Gurevich, *Appl. Phys. Lett.* **88 2006**

- Increase performance
  - Move beyond limits of Nb
- Decrease cost
  - Higher operating temperature (reduce cryogenic costs)
  - Replace bulk Nb with cheaper material (Cu/Al)




A. Gurevich, APL 88, 012511



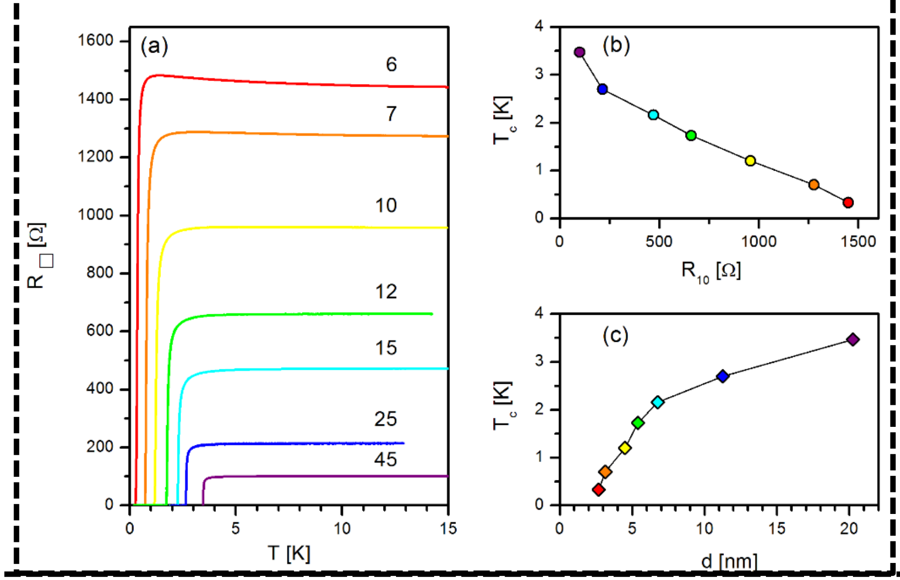

$$B_{c1} = \frac{2\phi_0}{\pi d^2} \ln \frac{d}{\tilde{\xi}}, \quad d < \lambda,$$

- Coat inside Nb SRF cavity with precise, layered structure → ALD

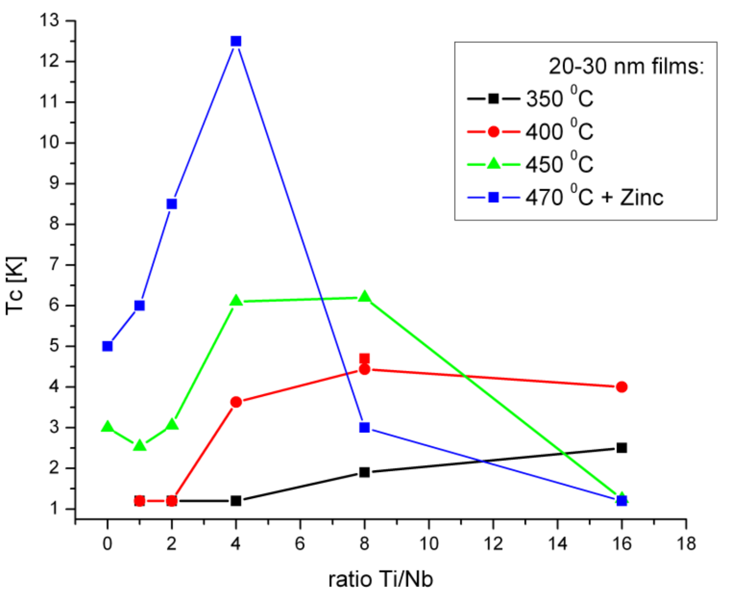
# ALD – Applications – Superconductors

- Niobium Carbide: NbC 1.7 K
- Niobium Carbo-Nitride: $NbC_{1-x}N_x$ 3.8 K
- Niobium Silicide: NbSi 3.1 K
- Titanium Nitride: TiN 3.9 K
- Molybdenum Nitride: MoN 12 K
- Niobium Titanium Nitride: $Nb_{1-x}Ti_xN$ 14 K

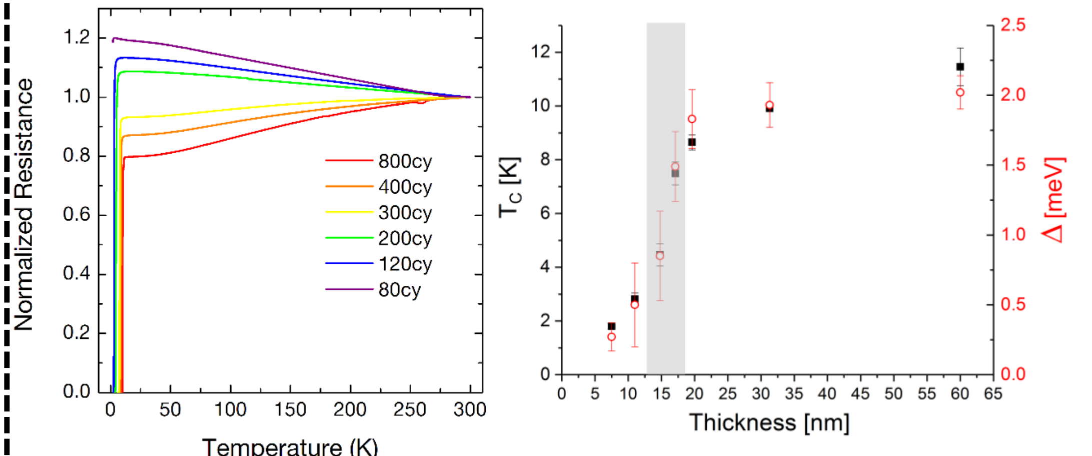
## NbSi – Tc vs thickness



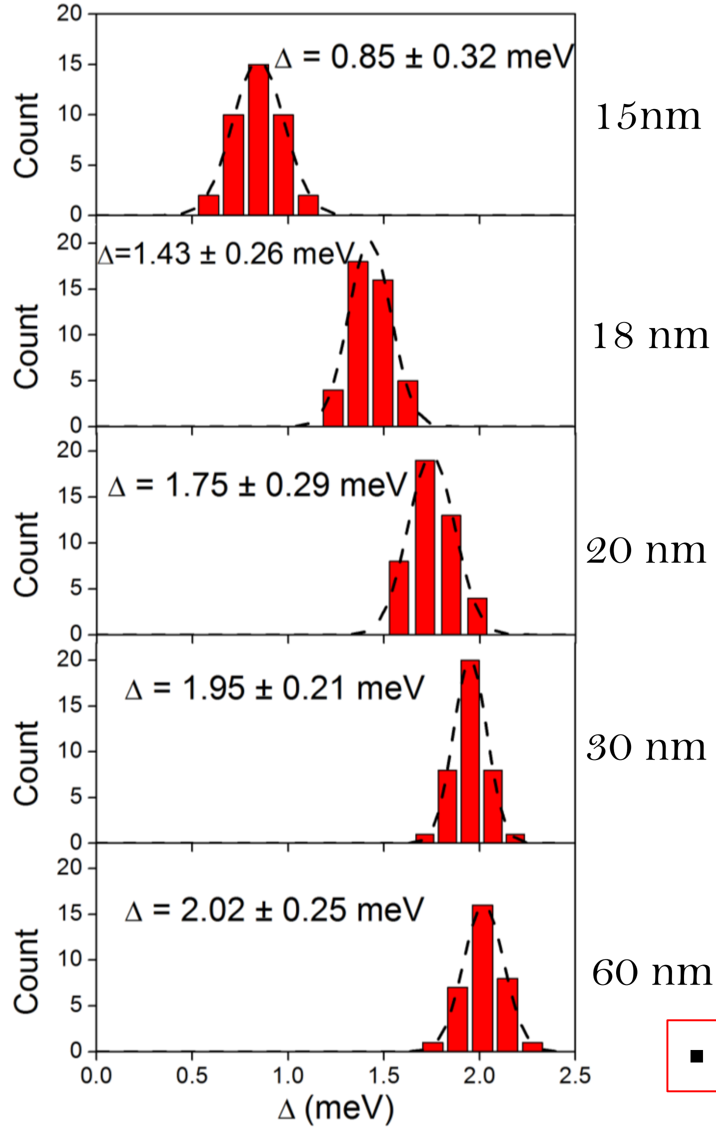

## $NbTi_xN$ – Tc vs Composition

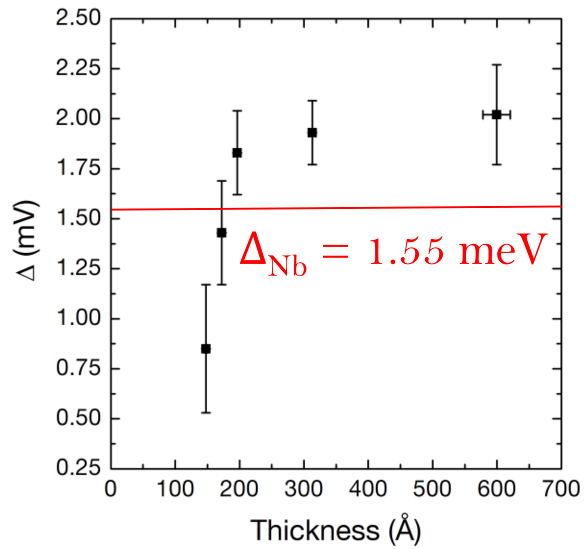


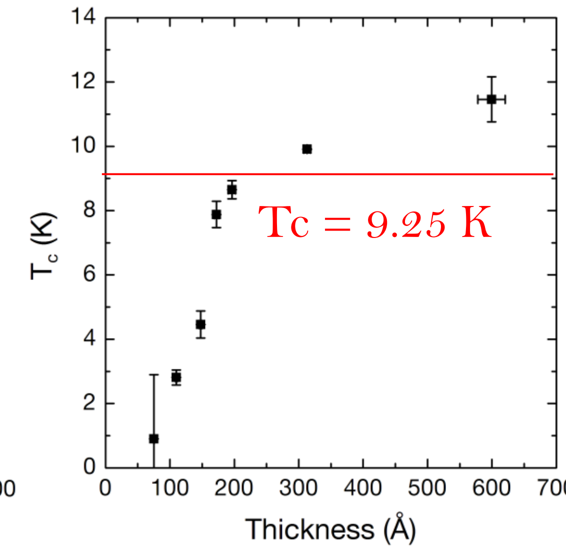

## MoN – Tc vs thickness

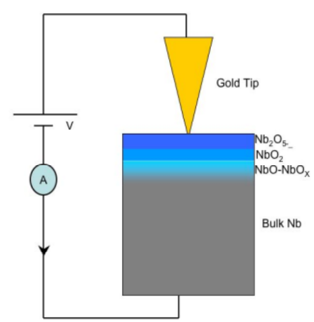


# $MoN_x$ ALD: Superconducting gap vs. thickness

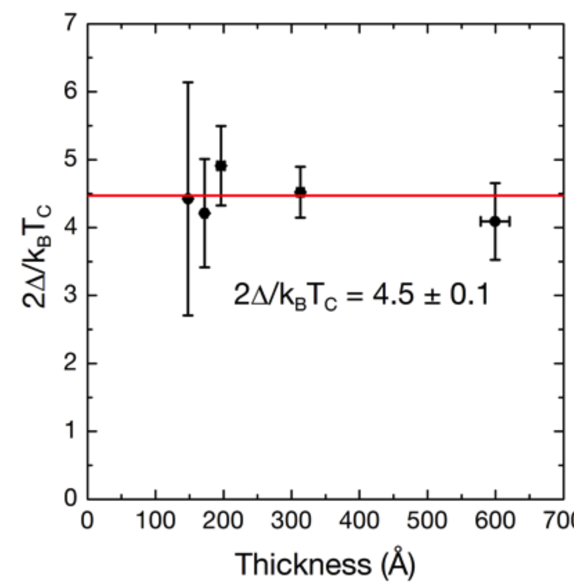


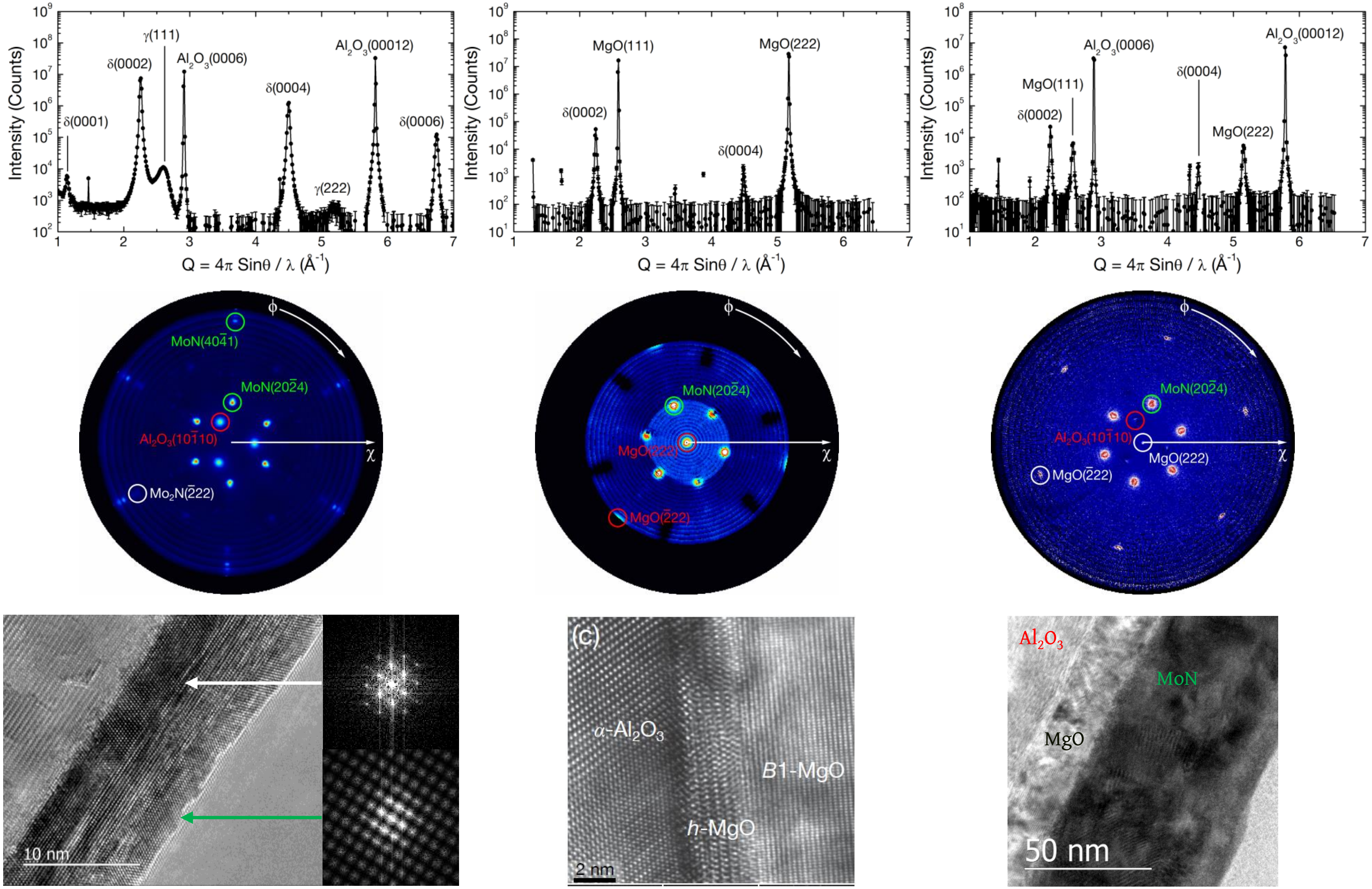




- Bulk for thickness > 50 nm



# High Quality superconductors by ALD? Epitaxy MoN



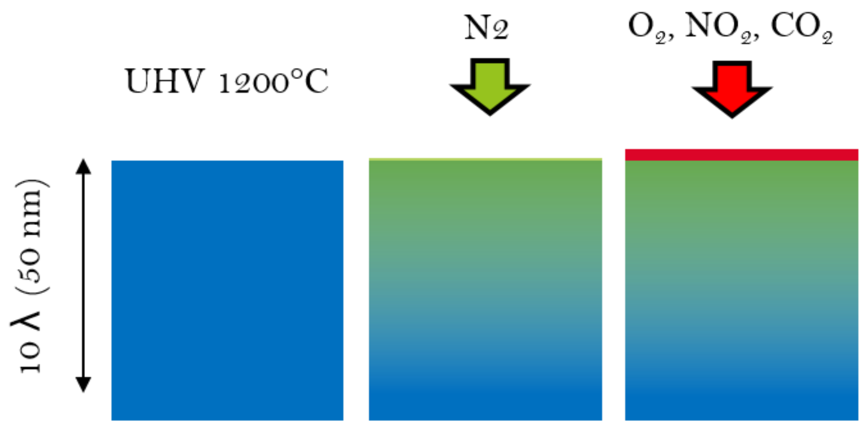

TEM: C. Alvarez (ANL/NU)

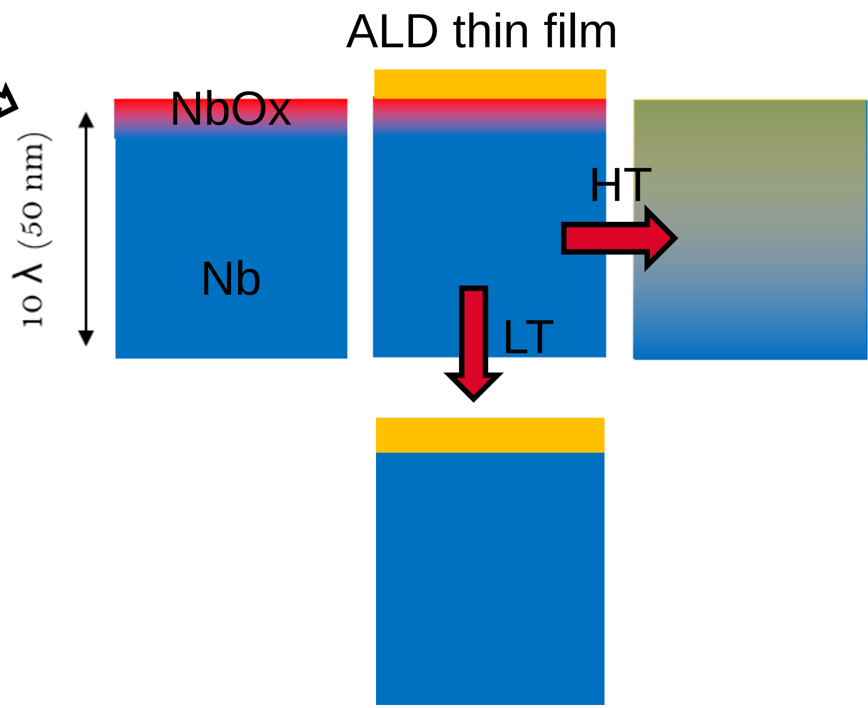
# Research thrust 2: Doping + Protection

- Dissociate influences: doping vs. oxide

- Gases sources:
  - Nitrogen < 300°C et $P_{N2}$ ~ $10^{-2}$ mbar
  - oxide growth: $O_2$, $NO_2$, $CO_2$
    à T ~ 100-150°C et $P_X$ ~ $10^{-2}$ mbar

- Film sources:
  - ALD of: NbN, TiN, MgO, $Al_2O_3$, $Y_2O_3$ …
    thickness ~ 5 nm
  - Low Temp (LT: 400-800°C) annealing:
    Replace NbOx by ALD capping layer with better properties (defects, dielectric, inert)
  - High Temp annealing (HT: 800-1200°C):
    Induce diffusion of metal cation from ALD film into first 50 nm of bulk Nb.



# ALD: Other applications for Particle accelerators

- 3D printing + ALD: beamlines optics (Fresnell, kinoform Lenses and capillaries): W ALD + PMMA
- Charging of ceramics: mitigation by TiN thin film (2-5 nm) on ceramic windows (coupler)
- Detectors (MCP's): controlling resistivity by doping W:Al2O3 + high SEE layer MgO
- Diffusion barriers or adhesive layers: $Nb_3Sn$/Cu or Nb/Cu > 400°C – TiN/AlN on Cu.
- Low secondary electron emission film ~ nm thick of carbide, nitrides…amorphous Carbon?

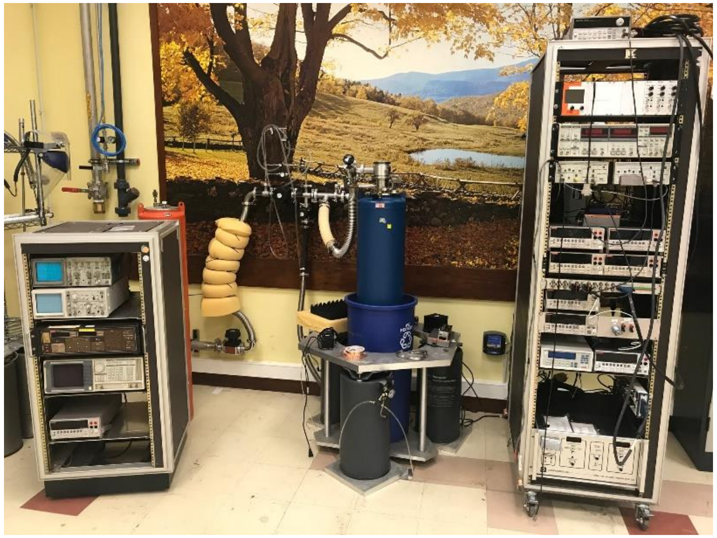
# Status report - ALD

- *<u>Commissioning of the ALD laboratory progressing</u>*:
  - Laboratory set up (gases supply, sample oven, computers..) √
  - Students hired (B. Delatte + R. Dubroeucq)√
  - Funding secured for Oven for ALD on cavities 135 k€ + Faraday cage for spectroscopy 75k€ √
  - Designed ALD chamber to be delivered next week (9 weeks delay - Neyco)√
  - Designed and ordered (Vacom) adaptative parts for ALD on cavities √
  - Oven set up built for clean room gaz thermal treatments. √
  - Preparation of 3 x 1,3 GHz (EPV) cavities baseline for ALD depositions √
  - Future: Receive and install Oven for ALD deposition on cavities and large objects
    Receive and install Faraday Cage.

**ALD - deposition**

**Thermal treatment**

**PCT - characterization**



# Aries POC Budget /total budget

| Items | Description | Cost (k€) |
|---|---|---|
| 1 | Niobium cavity 1.3 GHz Tesla Design | 10 |
| 2 | TOF-SIMS: Depth profile O + metal cations – 12 samples | 6 |
| 3 | ALD System oil pump (Neyco) | 6 |
| 4 | ALD System RGA | 15 |
| 5 | Adaptation flanges cavity – ALD system | 3 |
| 6 | ALD gas purifiers | 4 |
| 7 | Precursors | 5 |
| 8 | Shipping samples/cavity CERN – CEA | 1 |
| Total | | 50 |

Total budget ~ 285 k€

- ~20% internal, 80 % external including Aries and 2 other sources
- 3 years, 14 proposals to secure the budgetin
- Repted PhD student grants asked without sucess

# Timetable-Deliverable

WP1:

- D1 – ALD system commissioning

WP2:

- D2 – Deposition High Quality superconducting film on Nb samples
- D3 – Optimization of screening efficiency on Nb samples
- D4 – Deposition of optimized mutlilayers (MoN and NbTiN) on two Nb cavities

WP3:

- D5 – Deposition of MgO, $Y_2O_3$ and $Al_2O_3$ films on Nb samples
- D6 – Optimization of dopant concentration profile for MgO, $Y_2O_3$, $Al_2O_3$ films
- D7 – Deposition of selected film and post annealing treatment on Nb cavity

| WP\T | T1 | T2 | T3 | T4 | T5 | T6 |
|---|---|---|---|---|---|---|
| WP1-D1 | | | | | | |
| WP2-D2 | | | | | | |
| WP2-D3 | | | | | | |
| WP2-D4 | | | | | | |
| WP3-D5 | | | | | | |
| WP3- D6 | | | | | | |
| WP3-D7 | | | | | | |

# Partnership

Industry - ALD:

- CEA and **AirLiquide** Non-Disclosure Agreement (NDA) on new precursor synthesis and 3D printing of Cu, Ti, Nb, Al pure metals.
- AirLiquide agreed to provide ~ 200 gr of new precursors of Nb, Mo and Mg.
  -> first prove that ALD is working prior to more investment.
- Talk scheduled in September at AirLiquide
  -> prepare for next round of AirLiquide internal funding decision
- CEA-Tech: Start up Incubator. + Paris-Saclay: IncubAlliance.

Collaborations:

**CERN**

- Transport, magnetometry,Tunneling spectroscopy for Nb/Cu or $Nb_3Sn$/Nb and $Nb_3Sn$/Cu
- Cavity tests …

**DESY**:

- Tunneling spectroscopy and X-Ray diffraction, SEM…

**IPNO-LAL**

# THANKS !

Commissariat à l'énergie atomique et aux énergies alternatives
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Etablissement public à caractère industriel et commercial | R.C.S Paris B 775 685 019

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