^{163}Ho implantation in the HOLMES experiment

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for the HOLMES collaboration

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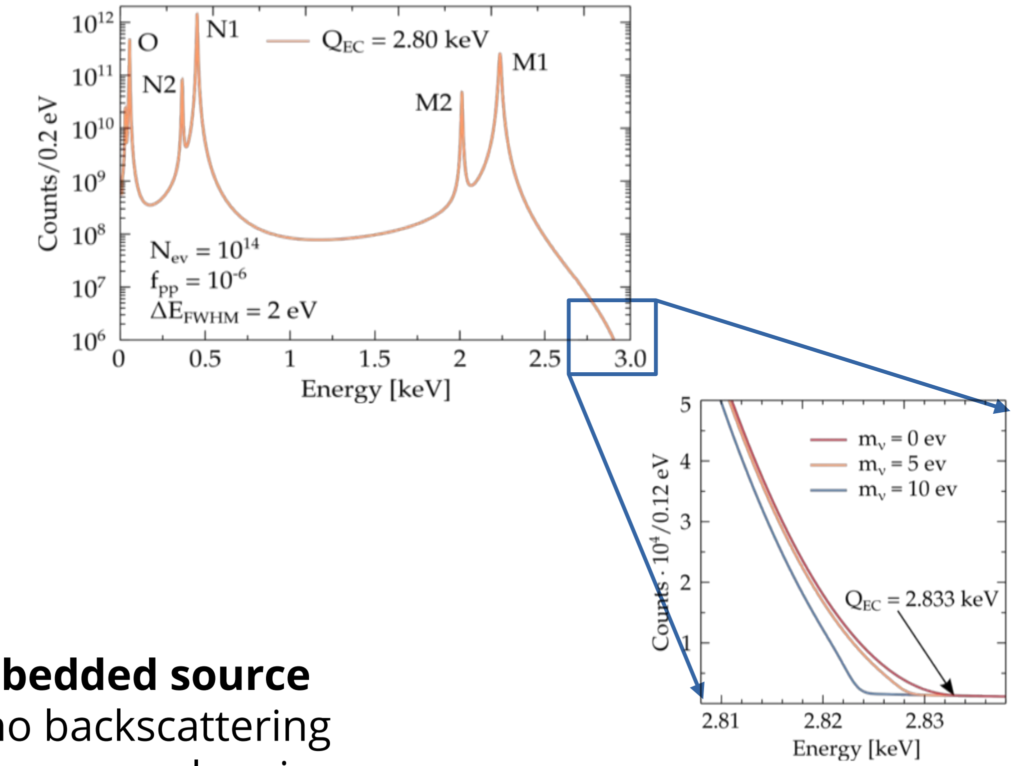
Direct neutrino mass measurement approaches

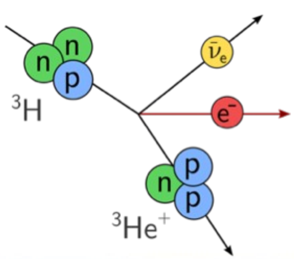
General requirements

- energy resolution order ~eV or below (comparable with m_ν)
- high statistics in the end-point region of beta decay / EC capture
  - ➔ low Q-value (stat ~$1/Q^3$)
  - ➔ high activity/efficiency of the source
- small systematic effects



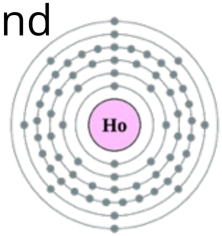
External source

\+ high statistics
\+ high energy resolution (below eV)
− systematics due to the source (energy loss)
− systematics due to decay to excited states
− background



Embedded source

\+ no backscattering
\+ no energy loss in source
\+ no solid state excitation
\+ no atomic/molecular final state effects
\+ good energy resolution (~eV)
− limited statistics
− systematics due to pile-up
− background

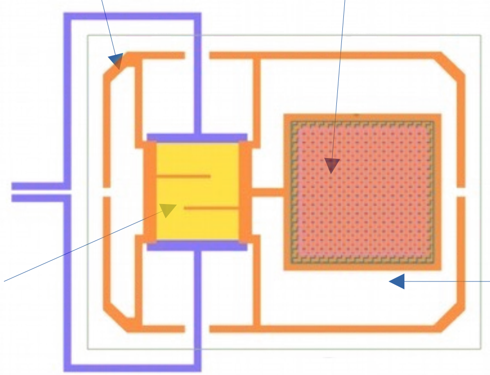


$$^{163}\mathrm{Ho} + e^- \rightarrow {}^{163}\mathrm{Dy}^* + \nu_e \rightarrow {}^{163}\mathrm{Dy} + E_c + \nu_e$$

HOLMES TES detector

Requirements

- energy resolution ΔE~ 1eV, time Δt~ 1μs
- 6.5 x 10^{13} nuclei/det, A(EC) ~ 300 Bq/det (challenging!)
- 1000 channels array: 6.5 x 10^{16} total nuclei (≈18 μg)
- O(10^{13}) events / year, data taking ~ 3 years
- pile up fraction $f_{pp} \approx A \times \Delta t = 3\times10^{-4}$



HOLMES technique

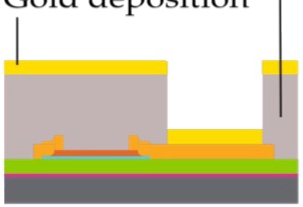
• Au absorber with ^{163}Ho inside coupled to TES Mo/Cu (T_c ~ 100 mK) thermometer

• Ho sandwiched between two 1 μm thick gold layers for a total electron containment

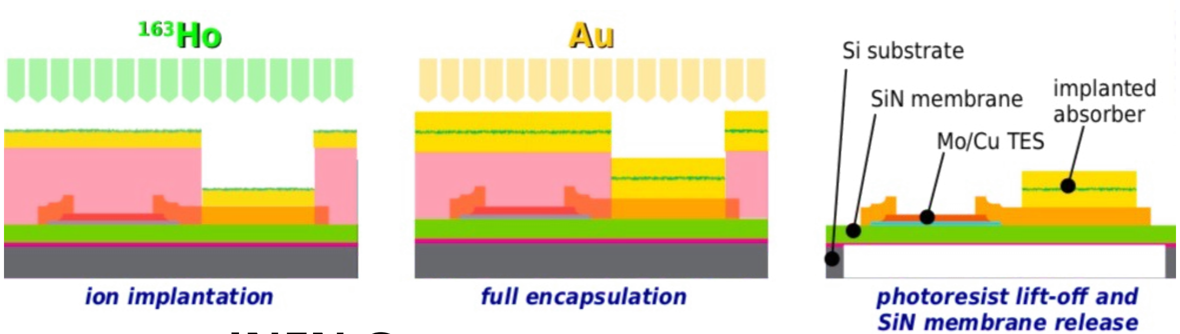
• very steep R vs T dependency in transition region

• fast detectors to reduce pile-up

- adjustable rise time ~ L/R
- decay time dependent on detector characteristics C/G



NIST



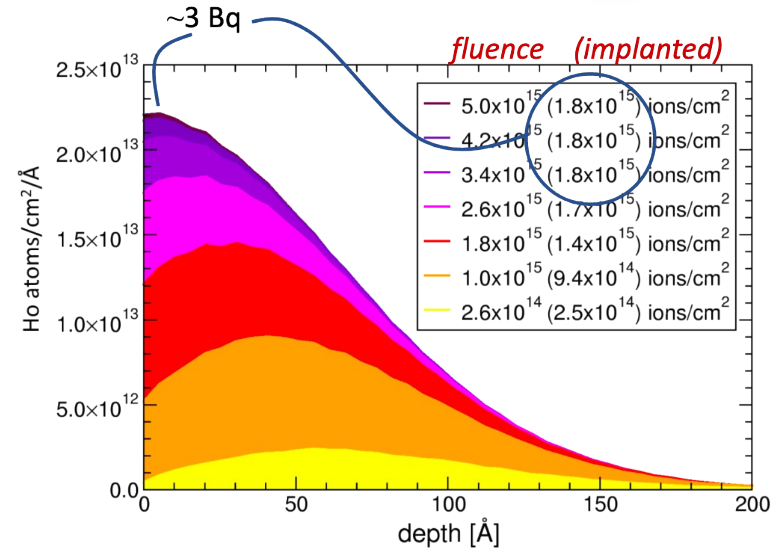
INFN Genoa **Milano-Bicocca U.**

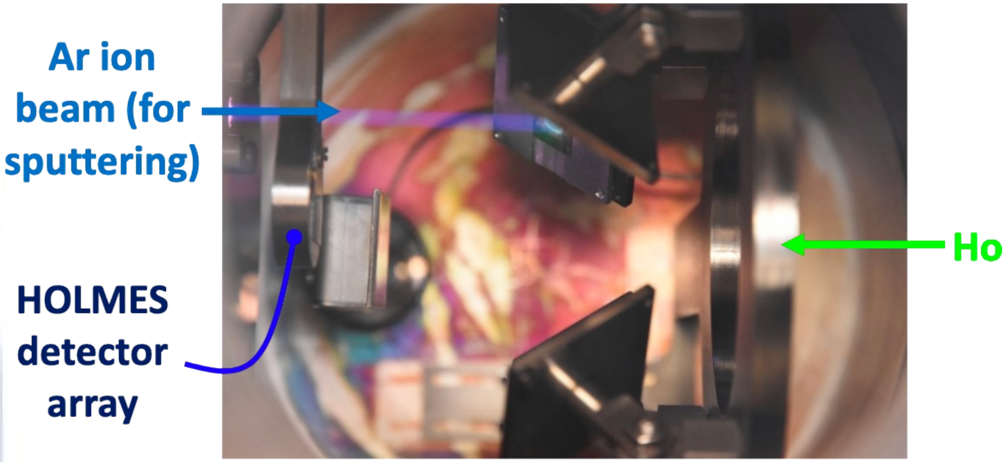
- TESs made by NIST
- Ho implantation in Genoa
- final machining and cryogenic operation in Milan

[A. Giachero et al., 10.1109/TASC.2021.3051104]

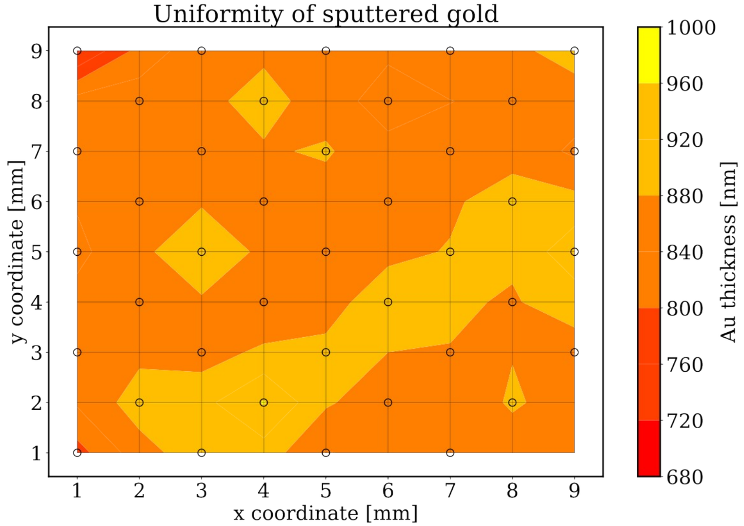
Au co-evaporation

Ho concentration in absorbers saturates due to incoming Ho sputtering deposited Ho.
→ compensation by a Au co-evaporation.
→ full encapsulation by deposition of final 1 μm Au layer to avoid Ho oxidation





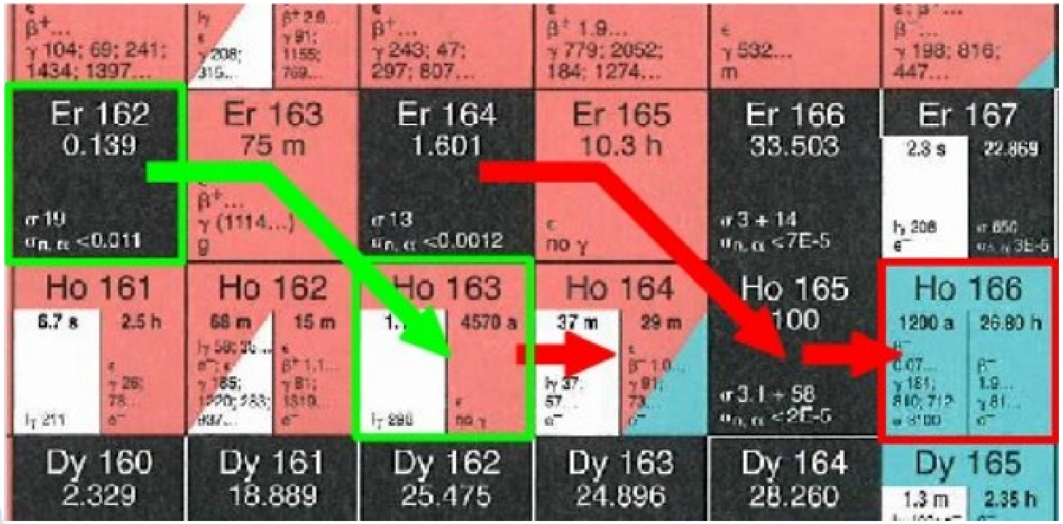
A dedicated target chamber designed and commissioned in Milano Bicocca:



- uniformity check: sputtering Au for ~22 hours on a Si slab 1 × 1 cm^2 with a drilled mask
  Thickness with profilometer:
  d = 865 ± 40 nm
- Deposition rate > 50 nm/hours
  → 1 μm Au can be deposited in around 20 hours.

See talk of M. Borghesi

^{163}Ho production and purification

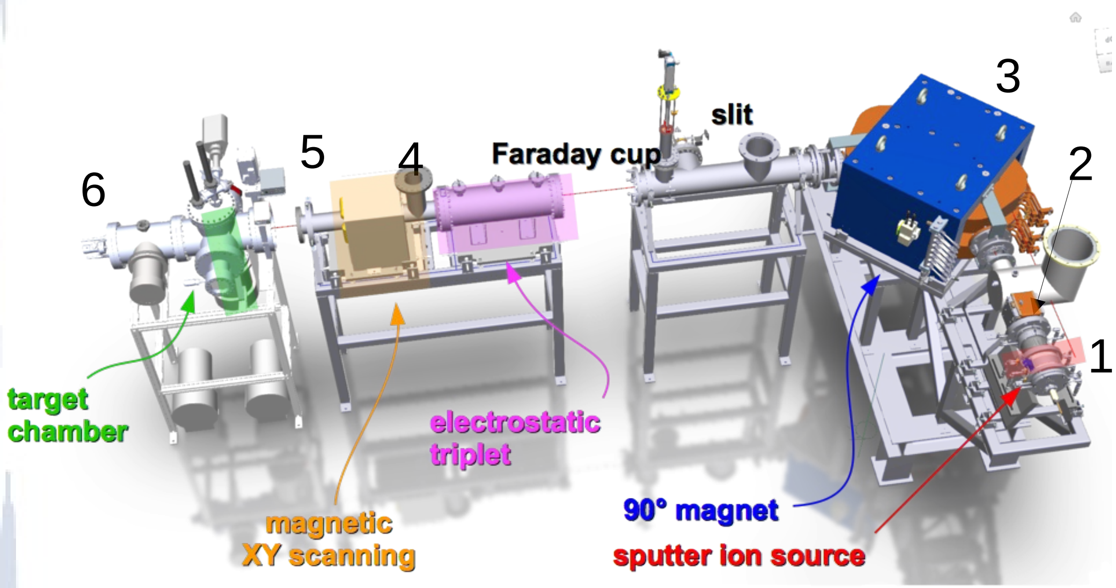


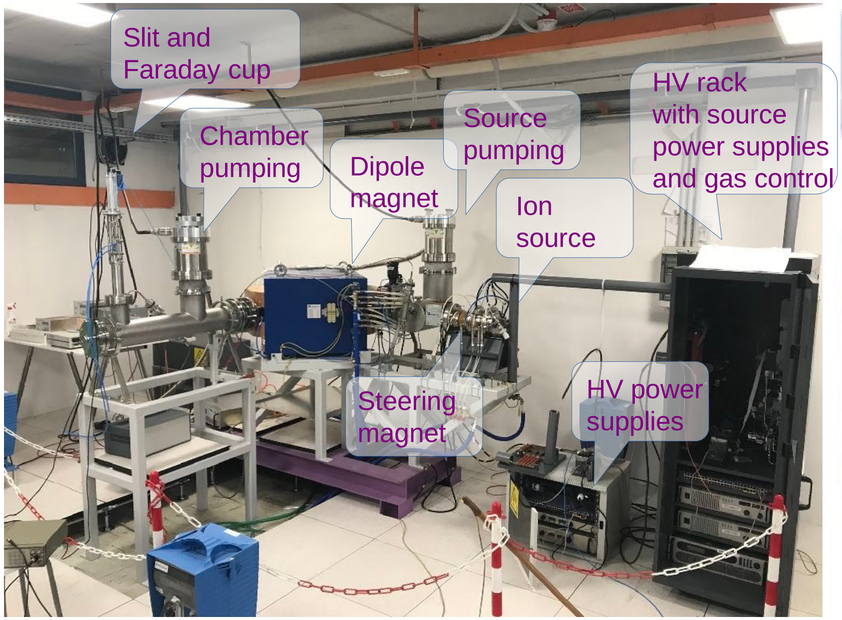
• ^{163}Ho produced by neutron irradiation of Er_2O_3 enriched (30%) in ^{162}Er at the Institut Laue-Langevin (ILL, Grenoble, France). Thermal neutron flux at ILL: 1.3×10^{15} n/cm^2/s.
$^{162}Er(n,\gamma)^{163}Er$ ($\sigma_{thermal}\sim 20$ b),
$^{163}Er + e^- \rightarrow {}^{163}Ho + \nu_e$ ($\tau_{1/2}\sim 75$min).

• **Contaminants**:
1. Other elements (residual Er, rare earth contaminants, decay product, etc…)
   → chemical purification (PSI);
2. Holmium isotopes, in particular
   ^{166m}Ho (β^-, $Q = 1856$ keV, $\tau_{1/2}\sim 1200$ y)
   $A(^{166m}Ho)/A(^{163}Ho) \sim O(1/1000)$
   → isotope separation with ion implanter (INFN Genoa).

• 110 MBq of purified ^{163}Ho available at Genoa (≈250 kBq of ^{166m}Ho) in oxide form (Ho_2O_3) in acid solution (pH<4) to avoid adhesion to the vial wall.
The Er recovered from the purification procedure is available to produce other 80 MBq of ^{163}Ho.

Ion mass separation & implantation



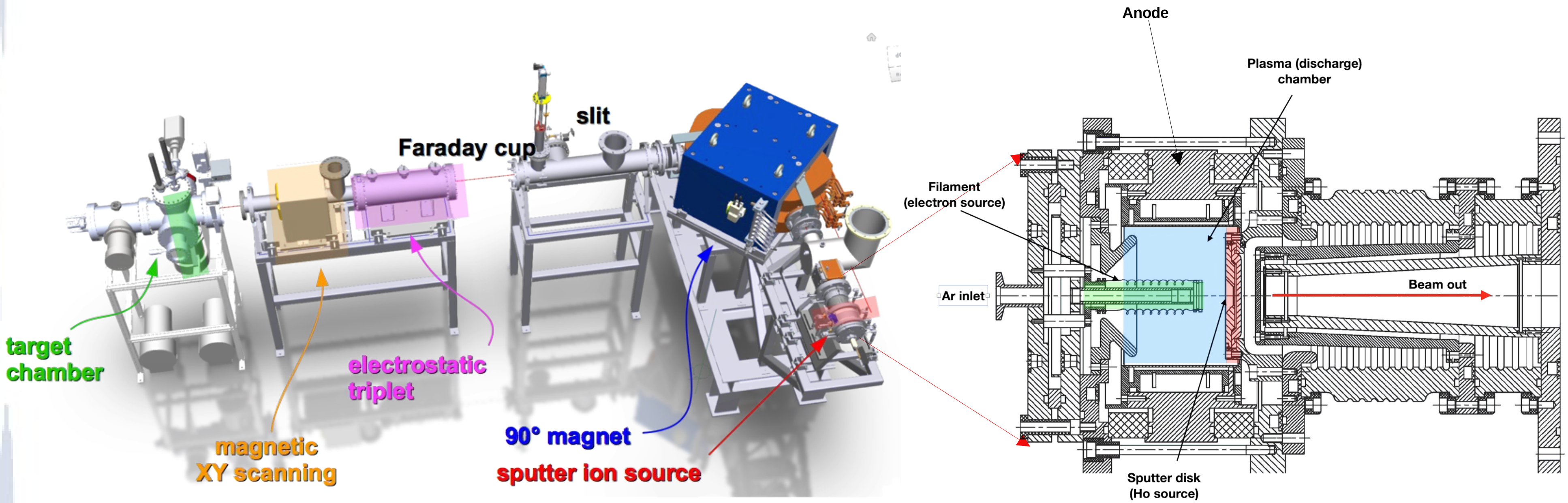


Ion implanter @ Genoa removes contamination of holmium isotopes different from ^{163}Ho & other impurities. Reduced setup has been comissioned.

Components:

1. **an argon sputter ion source** with an acceleration section to reach the beam energy of 50 keV (~50 nm implantation depth)
2. **a steering magnet** right after the ion source to correct the vertical component of the beam direction
3. **a magnetic/electrostatic mass analyzer** with magnetic field until 1.1 Tesla
4. **a focusing electrostatic triplet** (not yet mounted);
5. **a magnetic scanning stage** (not yet mounted);
6. **a target chamber** (not yet mounted).

Ion mass separation & implantation

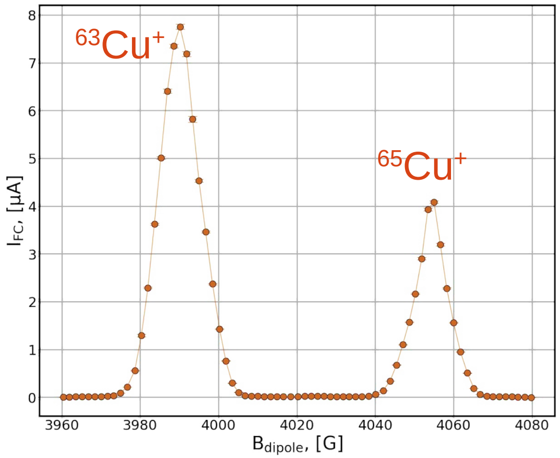


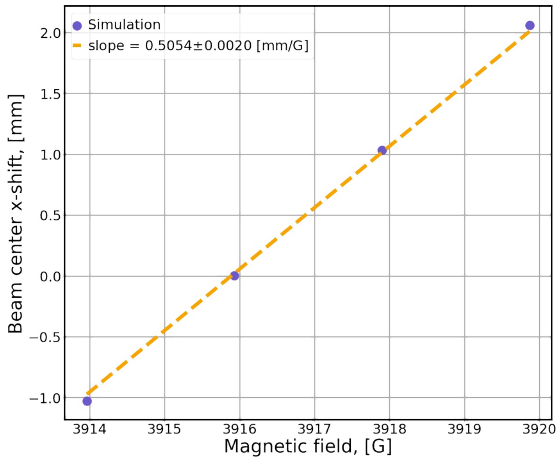
Penning sputter ion source (Danfysik):

- **Ar inlet**: controlled argon flow enters inside the discharge chamber. Argon is ionized and argon plasma burns inside the chamber
- **Filament**: electrons are emitted by thermionic effect. The electrons ionize argon and sputtered materials
- **Sputter disk target** with tunable electrical potential (max 600 V) to attract ions
- **Acceleration section**: ions pass through the hole in the sputter disk and are accelerated in this region (max 50 kV)
- all parts are water cooled but the sputter target reaches temperatures >1000 ^{o}C

Ion peak separation

• use *elegant*[1] simulation package for our beamline
from simulation we found @63 a.m.u., 1G corresponds 0.51 mm
• measured 63 G separation between $^{63}Cu^{+}$ and $^{65}Cu^{+}$ peaks corresponds to ≈ 32.5 mm
evaluated with MC simulation





• From Danfysik expectation, dipole should have a dispersion D(p)=2060mm at image plane where the slit is.

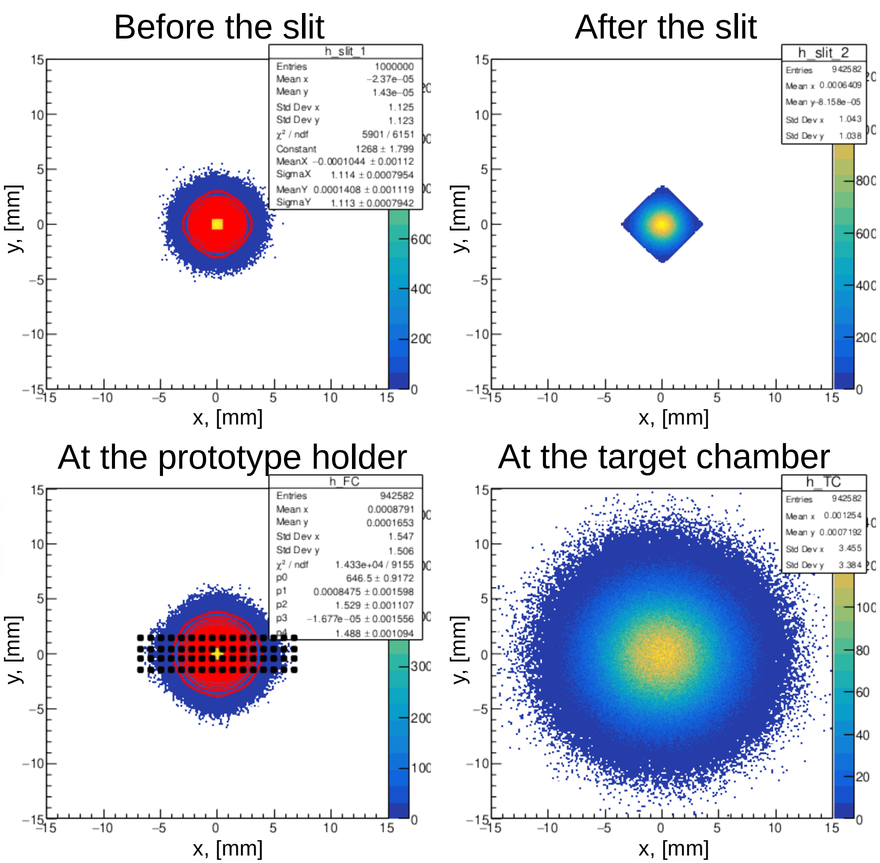
$$\Delta x = D(p)\frac{\Delta p}{p}$$

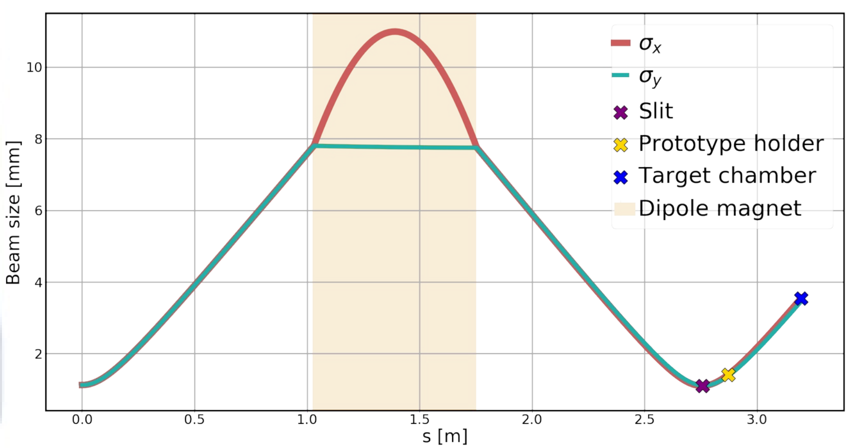
Δp/p @25keV (current nominal acceleration) for 65/63 a.m.u. = 1.575%
2060 mm x 0,01575 ≈ 32.5 mm → it seems Danfysik results are well-reproduced.

• 31 G/a.m.u. @63amu corrisponds to 15 mm/a.m.u. evaluated with simulations → extrapolation to the ROI gives 15.5 mm separation between 163/166 a.m.u. With beam size ~1.5 mm better than 10σ separation

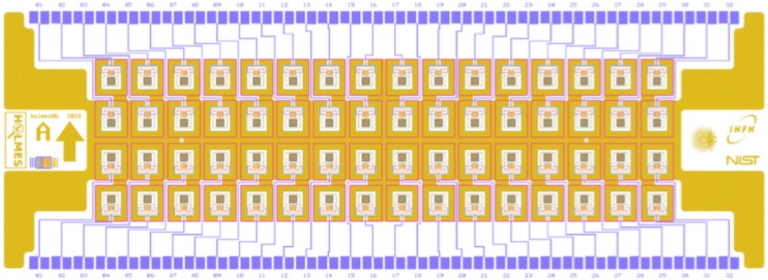
[1] an open-source code from APS for simulating rings, linacs, and beam transport systems, and for tracking particle beams, https://www.aps.anl.gov/Accelerator-Operations-Physics/Software

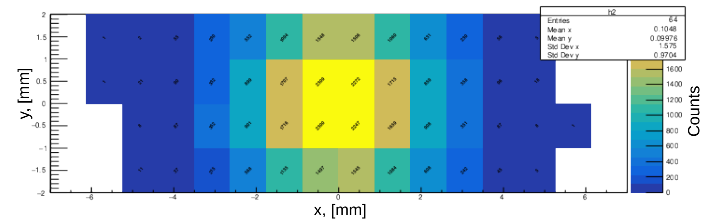
Geometrical efficiency simulations

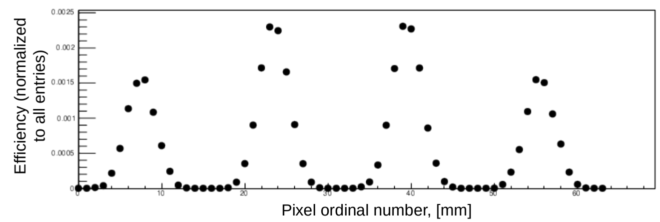


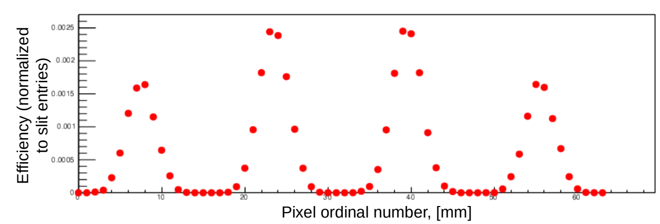


Simulated implantation efficiency
in 4x16 array of 200 μm x 200 μm absorbers on a holder







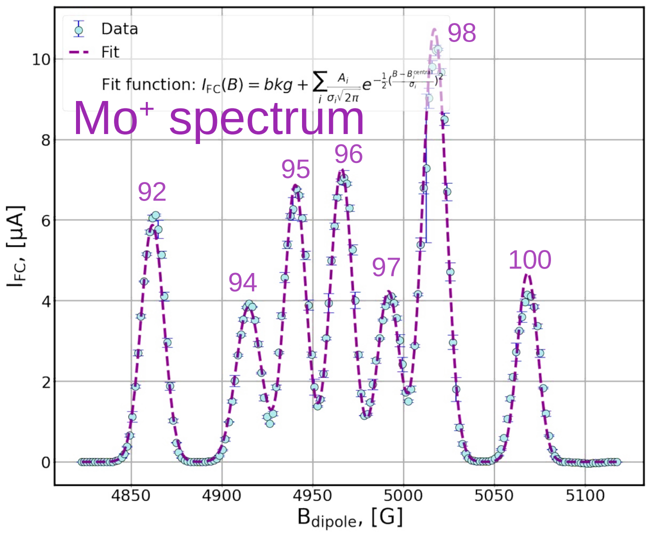


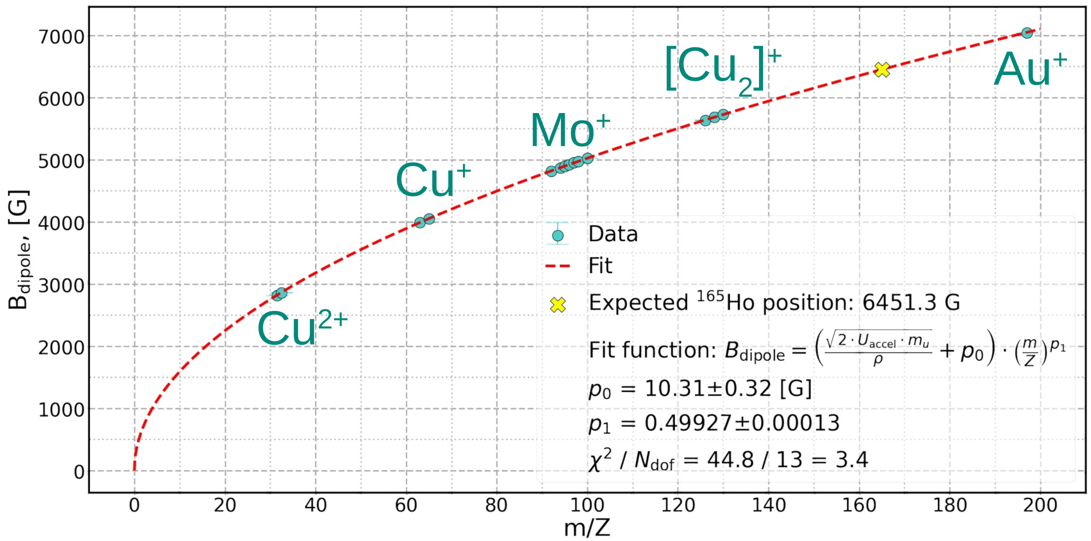
At the target chamber where the final array will be placed the beam will be more de-focused so more uniform implantation

Orient the array in vertical direction during implantation and swipe with beam steering for better uniformity

Dipole magnet calibration

- The dipole magnetic field is independently measured with a gauss probe
- Different materials inside the chamber
  (*e.g.* Cu/Au from sputter target/holder, Mo from the anode *etc.*) allow for a magnetic field vs mass-to-charge ratio calibration

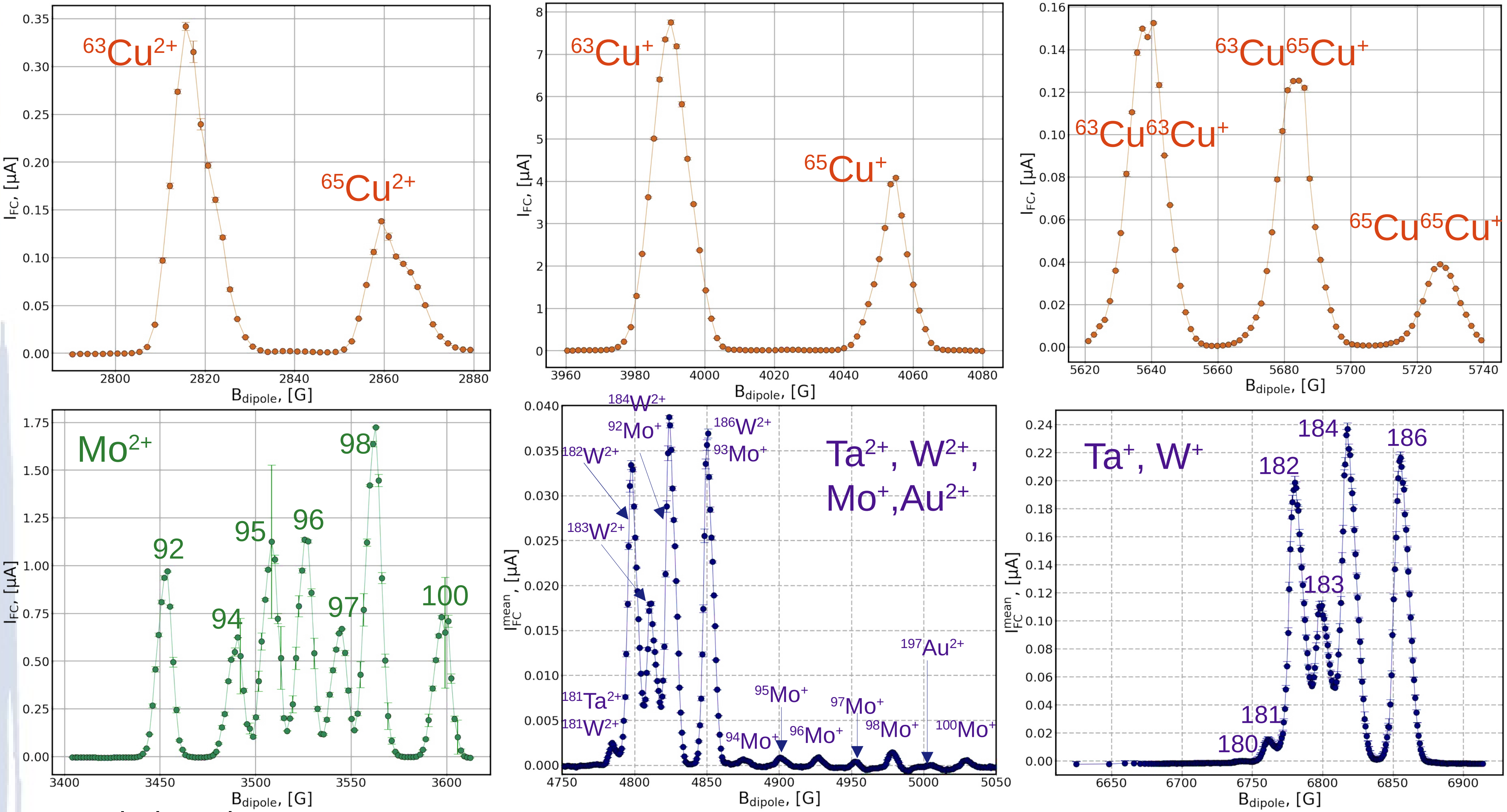




Natural abundances:
^{63}Cu: 69.17% ^{65}Cu:30.83%
^{92}Mo: 14.84% ^{94}Mo: 9.25% ^{95}Mo: 15.92% ^{96}Mo: 16.68% ^{97}Mo: 9.55% ^{98}Mo: 24.13% ^{100}Mo: 9.63%
^{197}Au: 100%

Currents of different ion species

The source produces also multiple-ionized and dimeric ions from the same material, which can also be used for calibration



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^{92}Mo: 14.84% ^{94}Mo: 9.25% ^{95}Mo: 15.92% ^{96}Mo: 16.68% ^{97}Mo: 9.55% ^{98}Mo: 24.13% ^{100}Mo: 9.63%

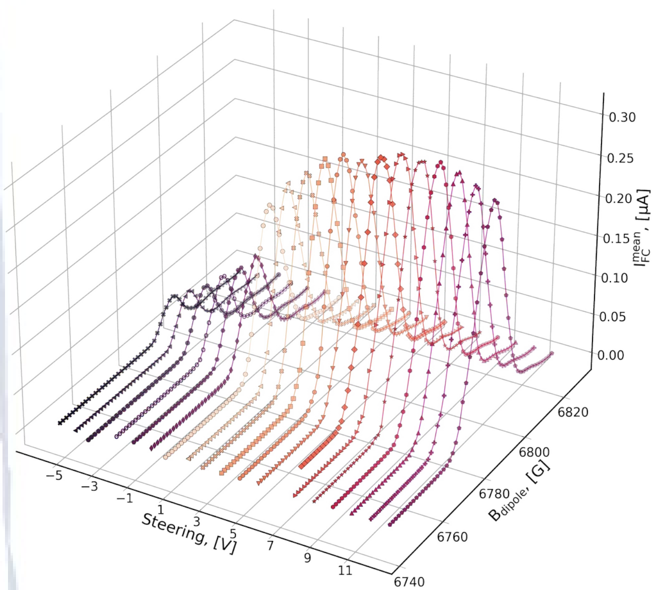
^{180}Ta: 0.012% ^{181}Ta: 99.988%

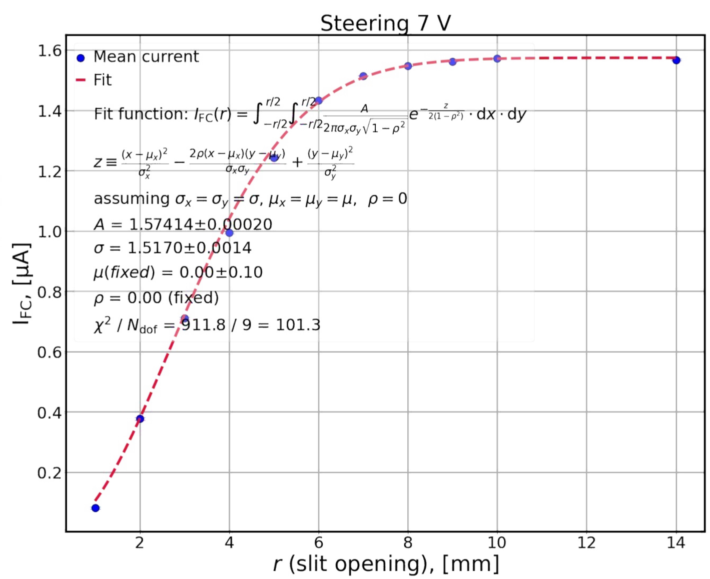
^{180}W: 0.12% ^{182}W: 26.50% ^{183}W: 14.31% ^{184}W: 30.64% ^{186}W: 28.41%

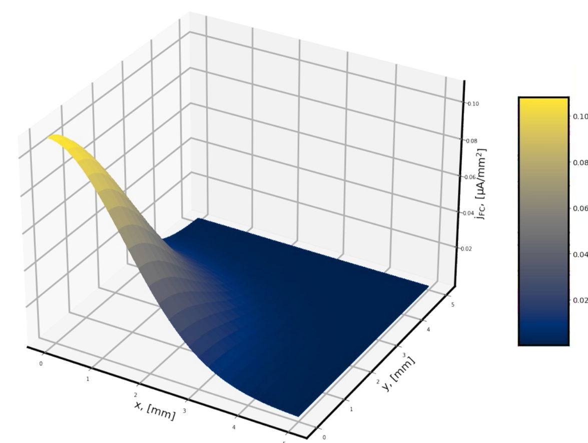
Beamline commissioning

- Steering magnet allows to center the beam, scan the peak with magnet to find maximum. Misalignment depends on ion mass and charge, other studies and simulation are in progress.

- Centering around the peak maximum with dipole and steering magnet, measure current for different slit apertures. Fitting with 2D symmetric Gaussian cumulative function we estimated a beam size σ≈1.5 mm.
  More advanced fit would take into account a finite 2-dimensional slit aperture

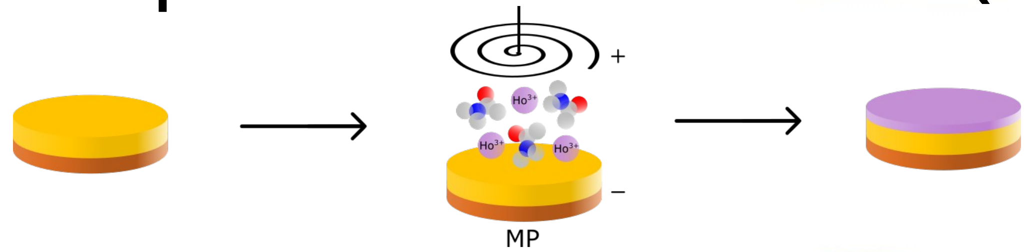




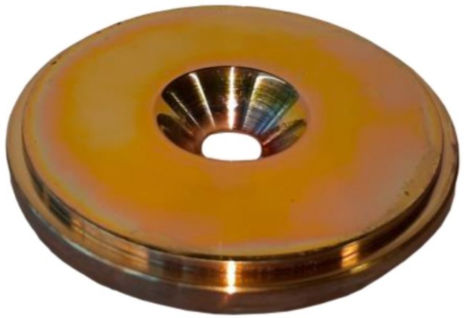


Targets with ^{165}Ho compound on surface (PSI)

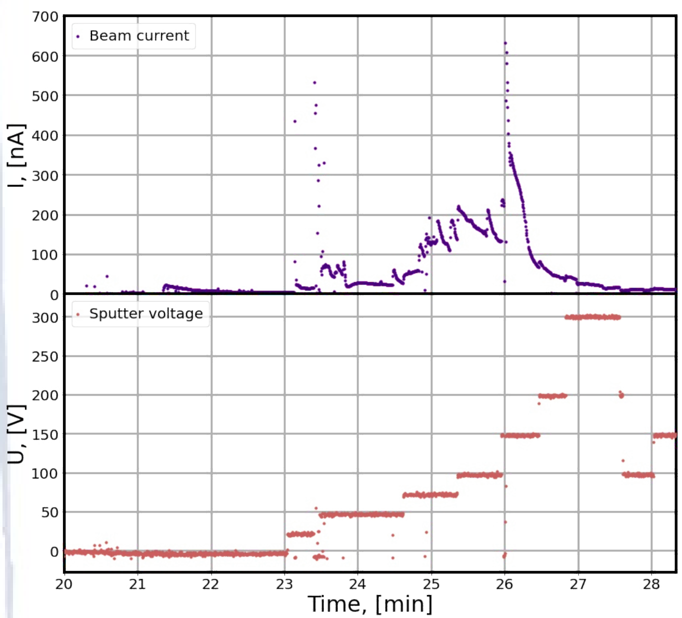
Molecular plating (MP)
$Ho(OH)_3$
PSI



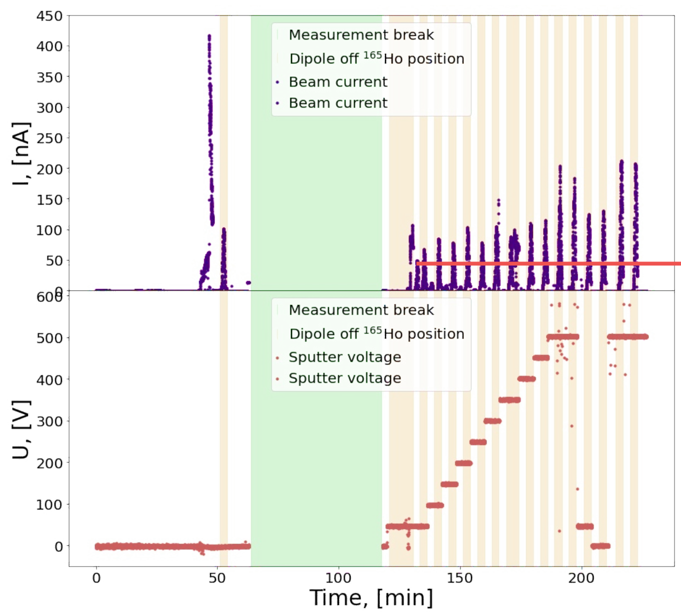
- deposition of Ho complexes in an organic solvent at high voltages with high uniformity and efficiency



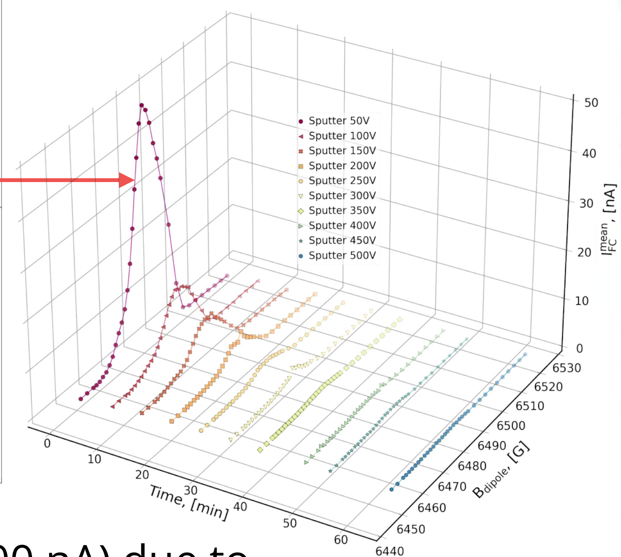
Sample 1:
Au 100 nm sticking layer
on Cu bulk



Sample 2:
Au 100 nm + Ti 18 nm sticking layer
on Cu bulk



^{165}Ho peak



- No $^{165}Ho^+$ current with sputter off
- Switching on the sputter, get $^{165}Ho^+$ current O(100 nA), spike ~700 nA and then vanishing in few minutes

- Initial $^{165}Ho^+$ current spike O(400 nA) due to filament warming, then from ~50 nA vanishing over 30 min

Targets with ^{165}Ho compound on surface (PSI)

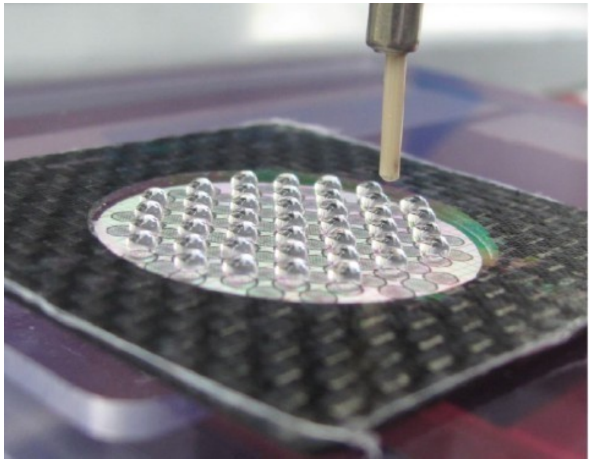
Drop-on-demand inkjet printing (IJP)

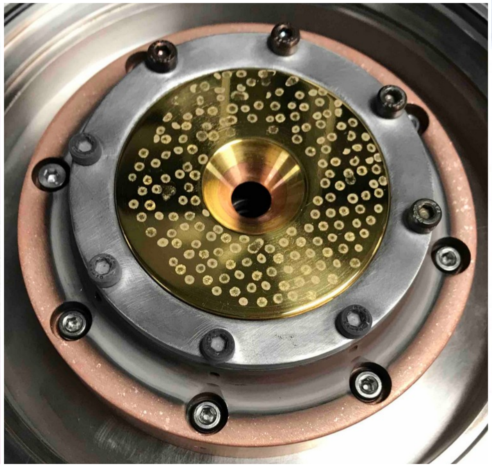
$Ho(CH_3COO)_3$

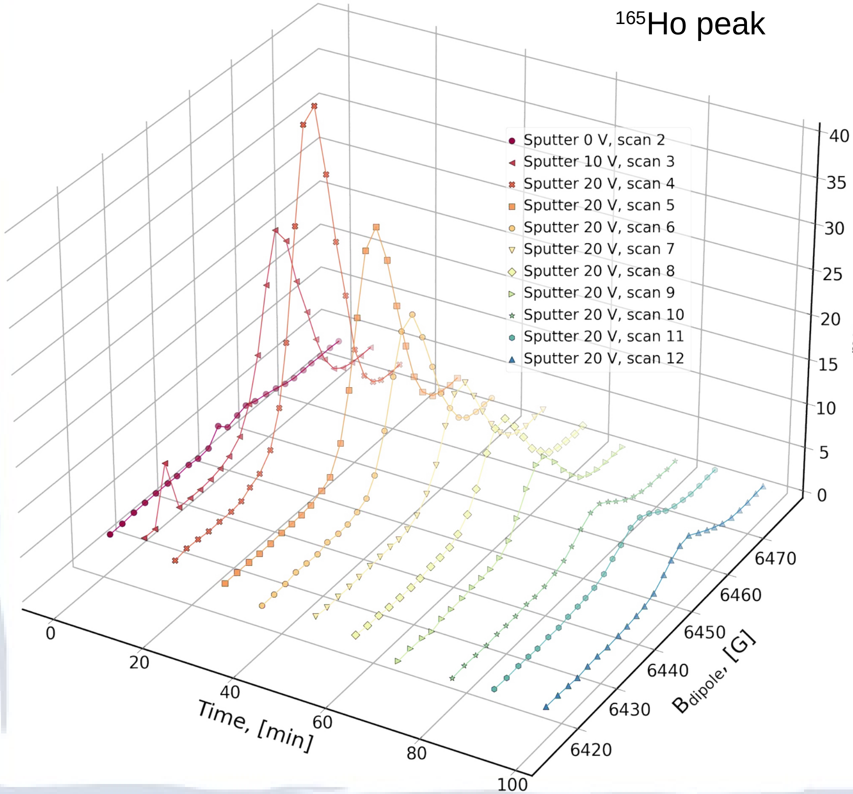
PSI

- put droplets of solution containing compound and let solvent evaporate to deposit the dissolved compound

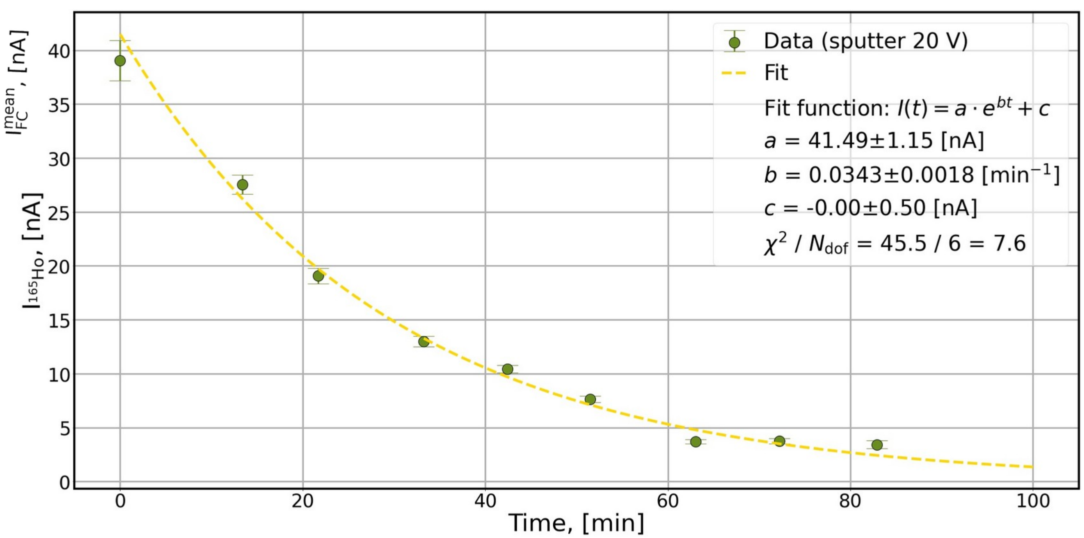
Au 100 nm + 20 nm Ti
sticking layer on Cu bulk







• Initial $^{165}Ho^{+}$ current ~40 nA
Exponential current decay over O(2 h)



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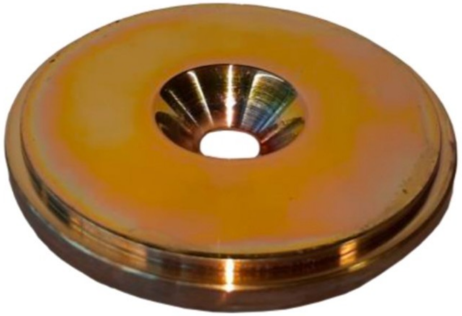
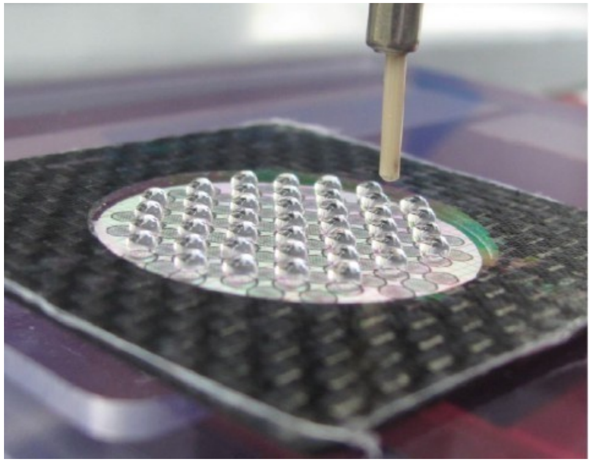
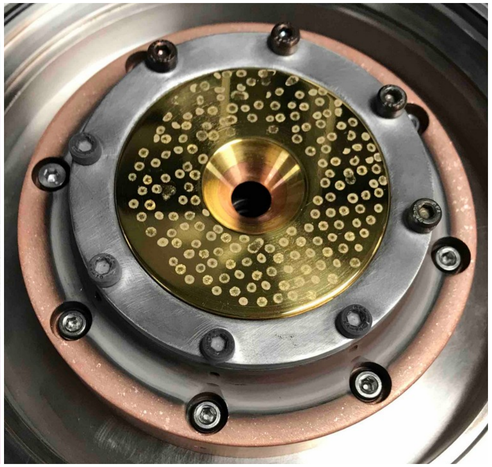
Molecular plating (MP)
$Ho(OH)_3$
PSI



Drop-on-demand inkjet printing (IJP)
$Ho(CH_3COO)_3$
PSI





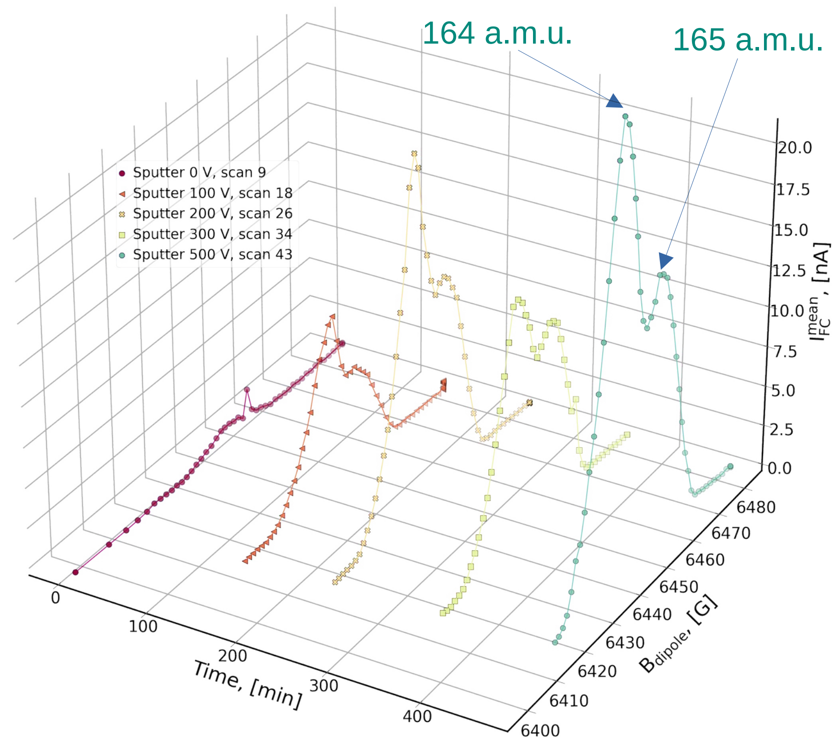
+ Safe fabrication
+ Possibility to use various materials to optimize properties
- Holmium as thin fragile film on surface

Sintered targets (UniGe)

Sinter metallic Ho 5%, Ti 36%, Ni 41%, Sn 18% on Cu holder

- Ho chips mixed with other fine-grained powders (≲40 μm) of Ti, Ni, Sn on a copper support and pressed at 350 bar/cm^2
- baked at 950 ^{o}C and at 10^{-4} mbar (low-oxygen environment) for 2 days

(create intermetallic compounds Ti_2Ni_2Sn/HoNiSn)



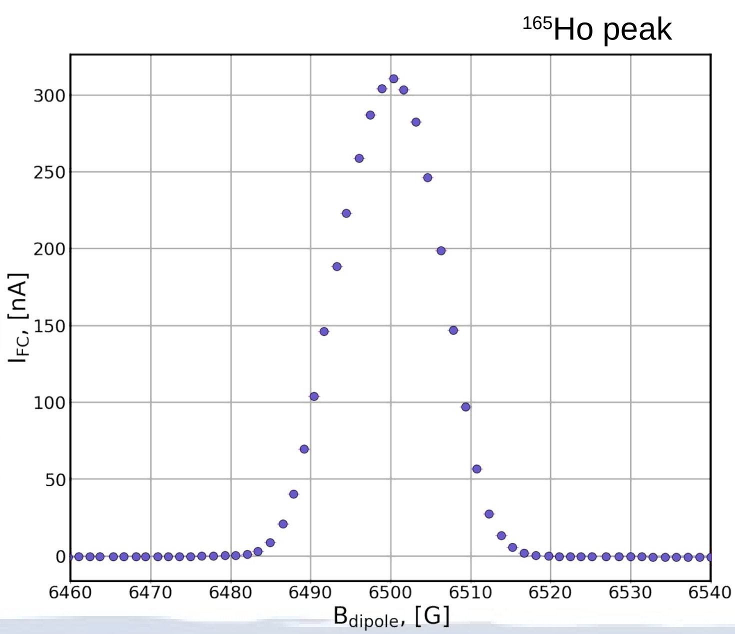
- Parasitic peak at 164 a.m.u. (presumably from $^{48}Ti^{116}Sn^{+}$) along with 165 a.m.u.
- Sustained $^{165}Ho^{+}$ current O(10 nA), no signs of extinguishing but the target melted after 7 h run

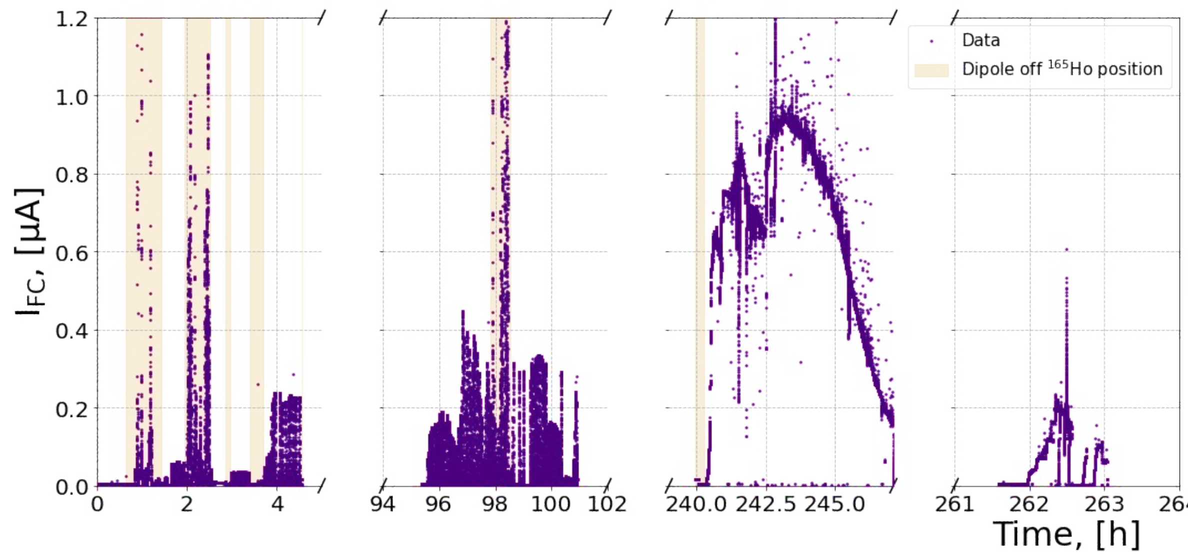
Sintered targets (UniGe)

Sinter
Zr 95%, Y 5%
on Ti holder
with $Ho(NO_3)_3$

- ^{165}Ho in form of $Ho(NO_3)_3$
- Y chips and Zr fine-grained powder (≲100 μm) prepared pressed at 350 bar/cm^2
- baked at 950 ^{o}C and at 10^{-4} mbar (low-oxygen environment) for 3 hours.
- $Ho(NO_3)_3$ deposited on the Zr-Y sinter and dryed on a hot plate in Ar atmosphere to avoid oxidation

- Ti holder for better sinter attachment





- O(200 nA - 1 μA) $^{165}Ho^+$ current sustained over O(15 h)

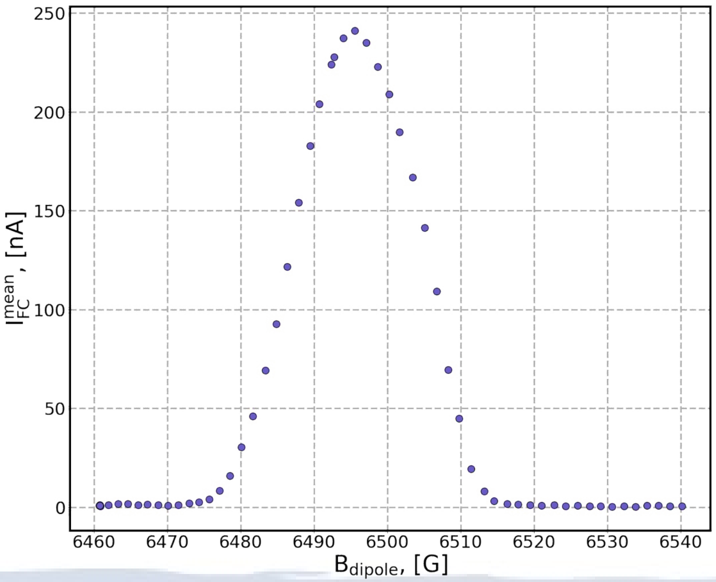
Sintered targets (UniGe)

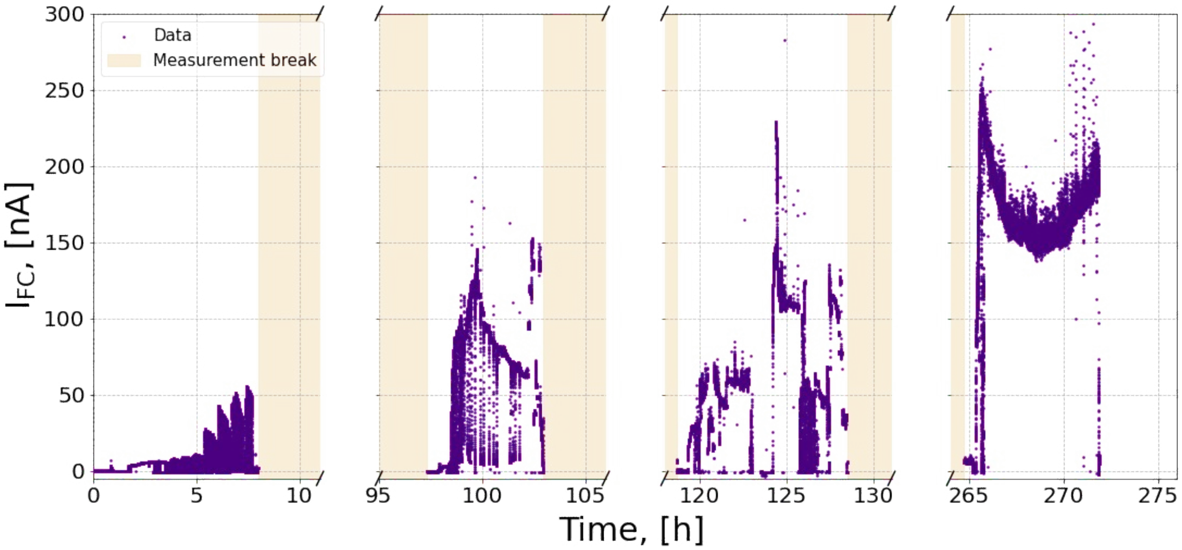
Sinter
Zr 95%, Y 5%
on Mo holder
with $Ho(NO_3)_3$

- ^{165}Ho in form of $Ho(NO_3)_3$
- Y chips and Zr fine-grained powder (≲100 μm) prepared pressed at 350 bar/cm^2
- baked at 950 °C and at 10^{-4} mbar (low-oxygen environment) for 3 hours.
- $Ho(NO_3)_3$ deposited on the Zr-Y sinter and dryed on a hot plate in Ar atmosphere to avoid oxidation

- Mo holder for better thermal properties

^{165}Ho peak





- O(100-200 nA) $^{165}Ho^+$ current sustained over O(15 h)

Sintered targets (UniGe)

Sinter
Zr 95%, Y 5%
on Mo holder
with $Ho(NO_3)_3$

Sinter
Zr 95%, Y 5%
on Ti holder
with $Ho(NO_3)_3$

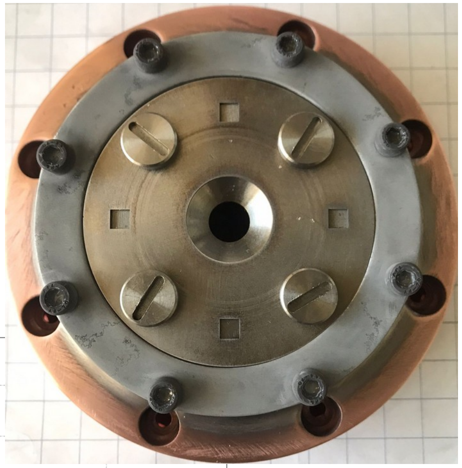
Sinter
metallic Ho 5%,
Ti 36%, Ni 41%,
Sn 18%
on Cu holder

+ Holmium is distributed over the entire thickness of the target
+ Possibility to use various materials to optimize properties
- Careful handling of radioactive powder

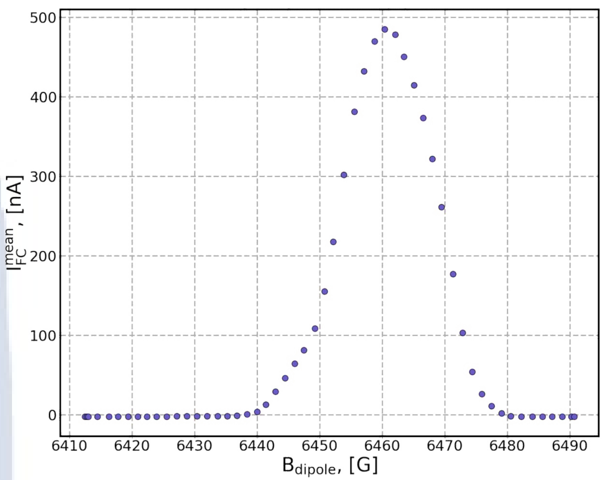
Other target types

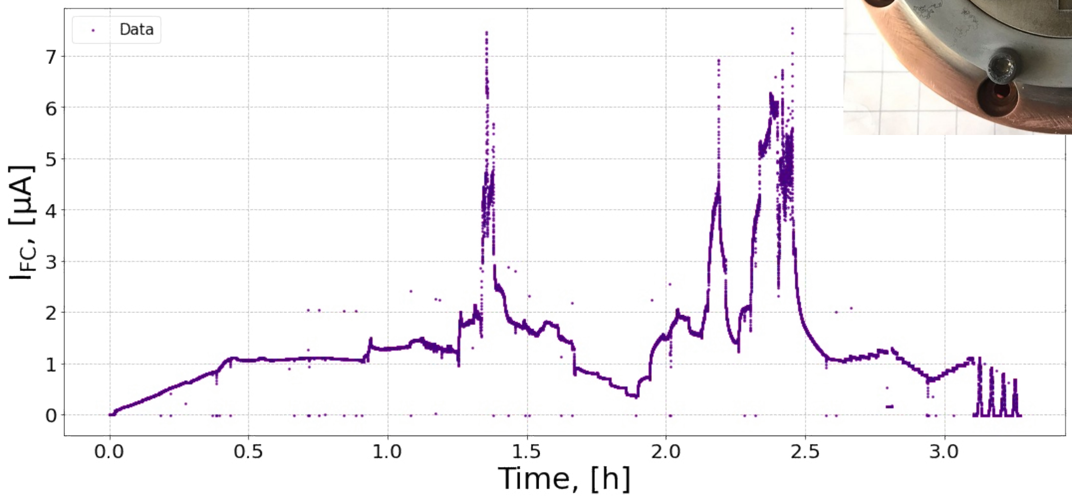
Ho foils
UniGe

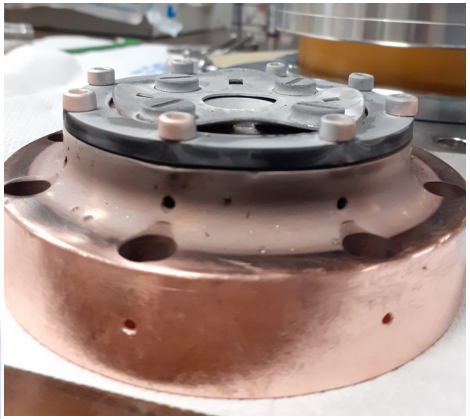
- 4 square pieces of Ho foil (≲0.5 g) pressed to Bulk Ti holder by a Ti mask with 4 square holes correspondingly

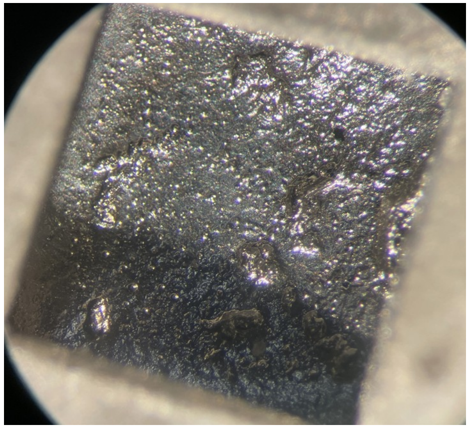


^{165}Ho peak







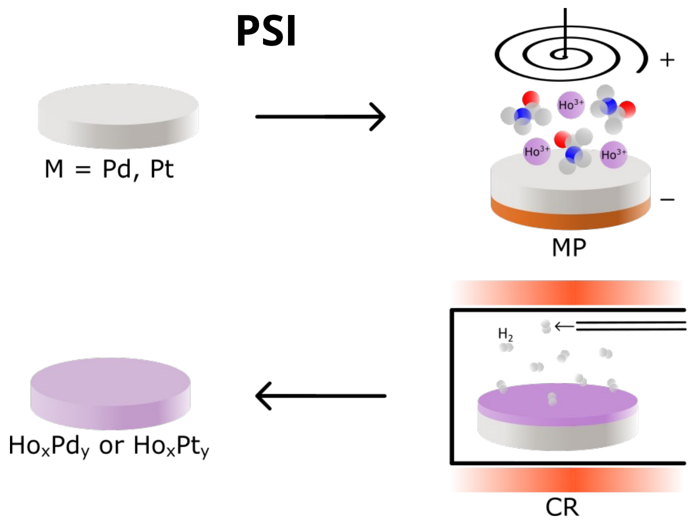


- Current O(μA) current for at least a few hours
- The test was stopped due to a short circuit created inside the source (presumably by a deposit)
- After the test was finished and the source opened, we wound the Ti mask deformed;
- Ho foils were welded on the holder and signs of sputtering are seen under the microscope

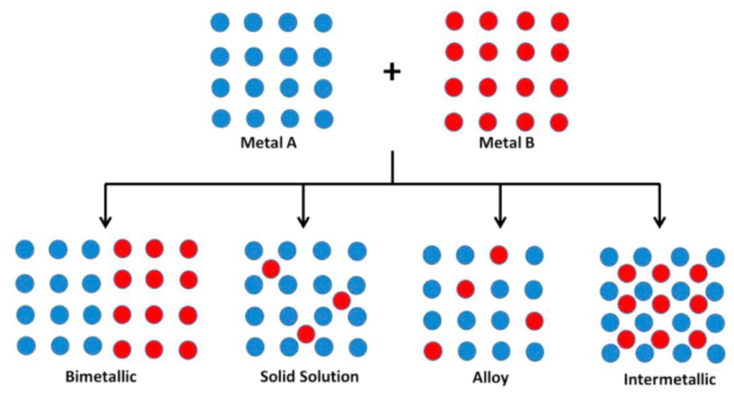
Other target types

Coupled reduction (CR) of $Ho(NO_3)_3$ on Pd

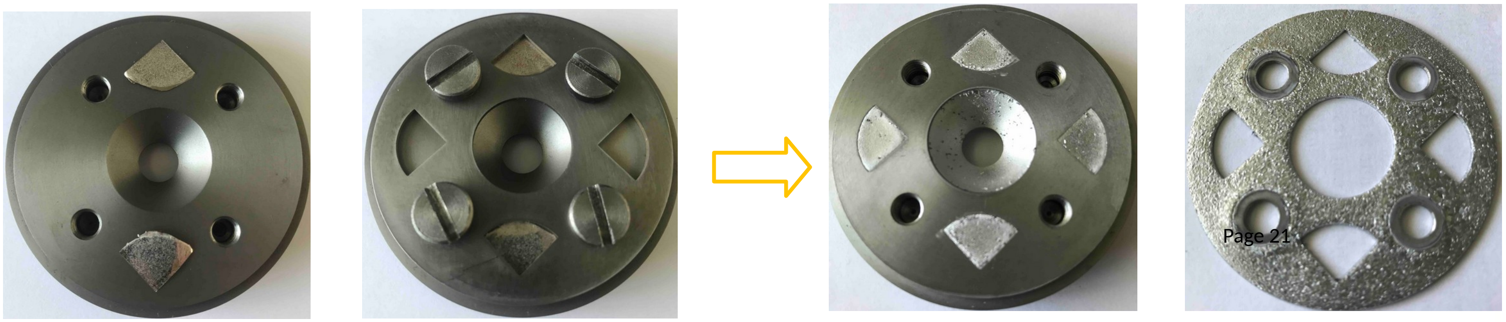
PSI

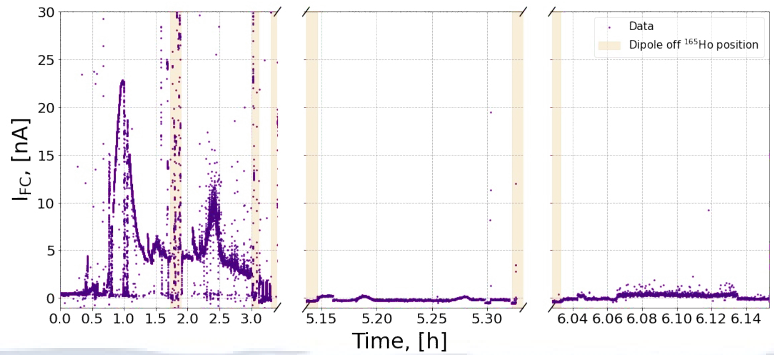


- Ho reduction and diffusion into backing material due to thermodynamically favourable formation of intermetallic compound



• 2 out of 4 wedges of the Pd substrate with CR Ho fixed with Mo mask used in test





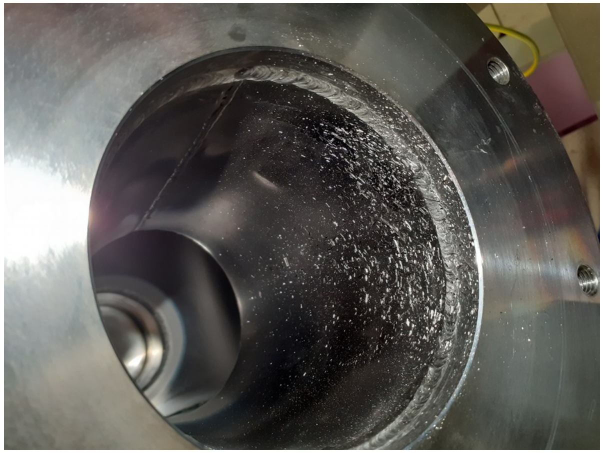
• Initial current ~23 nA vanished over ~5 h
Suspect Ho did not diffuse well in Pd → under investigation

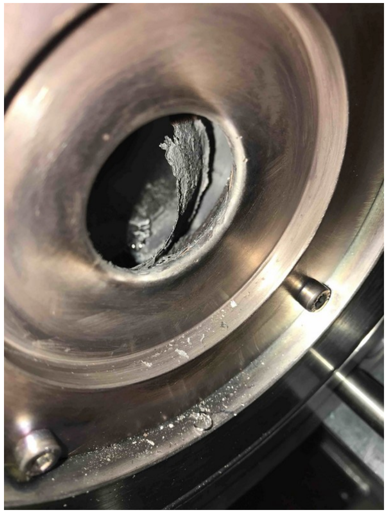
• Try encapsulating one of remaining samples in Pt via discharge welding for the next tests

^{165}Ho extraction efficiency

Target	^{165}Ho atoms in target (estimation)	^{165}Ho atoms extracted (estimation)	Extraction efficiency (lower limit), %
Inkjet printing with acetate	$6 \cdot 10^{18}$	$6.5 \cdot 10^{14}$	0.01
Sinter Zr/Y on Ti holder	$2.3 \cdot 10^{20}$	$1.36 \cdot 10^{17}$	0.06
Sinter Zr/Y on Mo holder	$7 \cdot 10^{19}$	$4.4 \cdot 10^{16}$	0.06
Coupled reduction on Pd	$4.4 \cdot 10^{18}$	$12.7 \cdot 10^{13}$	0.003

Opening the source after tests, we find a lot of target material deposit inside the chamber





Summary

NuMass 2020:

- finished production and purification and stored 110 MBq of ^{163}Ho
- done first test of the implanter setup with dummy copper target

NuMass 2022:

- The ion source with the reduced implantation beamline has been commissioned and showed a good performance – extraction of ion beams of different materials present in the source chamber.
- Calibration procedure is established, we can correct for misalignment and estimate beam size, more detailed investigations and comparison with simulations are ongoing.
- Look for optimal sputter target candidate, ideally chemically pure, having homogeneous distribution of Ho and provide a slow and constant Ho release (and obtained with high yield)
- Tests with different natural ^{165}Ho-containing targets show clear peak to 165 a.m.u.:
  - MP, IJP: current stability not satisfactory, 50 nA at max
  - Sinter and Ho foil: sustain high current over O(15 hours), higher currents achievable

→ targets with Ho distributed over the target volume work significantly better then superficial Ho

Outlook

- Better understanding of implanter setup, source tuning to control better power dissipation on sputter target and decrease its working temperature, and effect of the source parameters on the beam profile

- A more detailed analysis of the data already acquired from various targets is ongoing

- Tests of several new types Ho-containing targets (e.g. CR Ho melted inside Pt drops; combine sinter + IJP + CR) to find the optimal one in terms of Ho extraction efficiency/handling

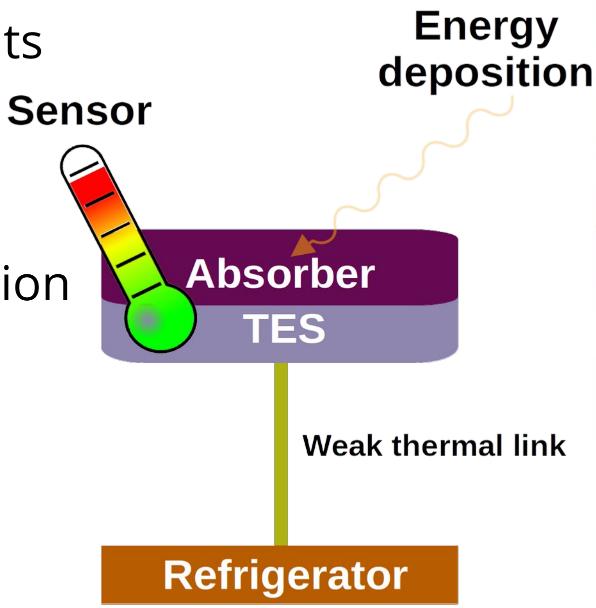
- After the optimal target is chosen, implant a detector array with low dose of ^{163}Ho (~1 Bq/det)

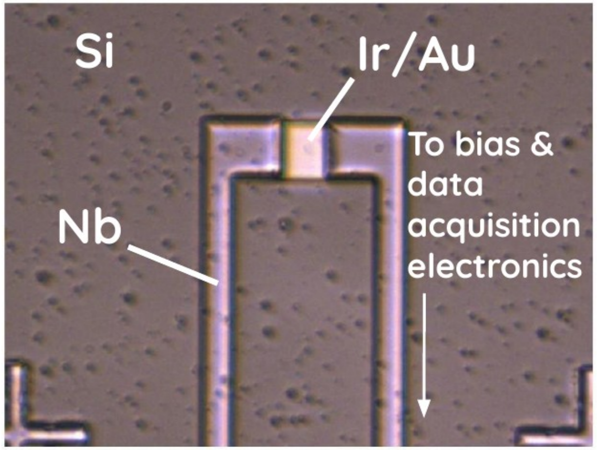
Thanks for your attention!

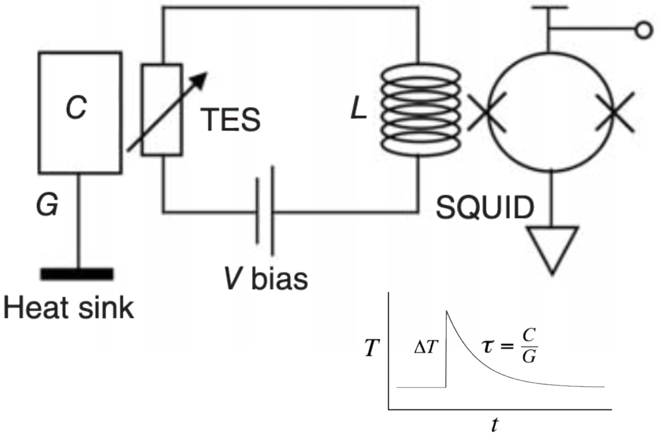
TES detectors

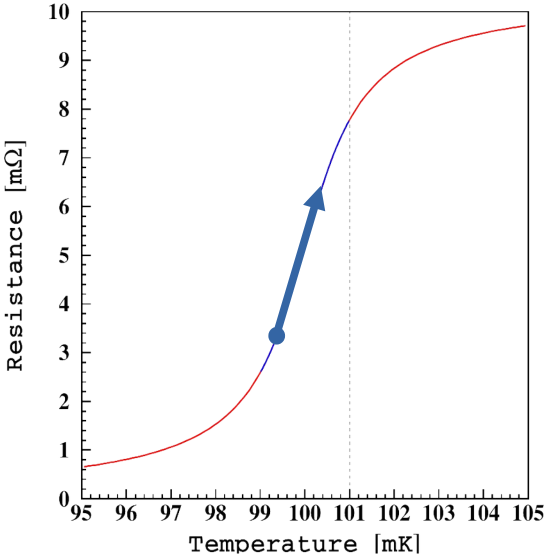
Transition Edge Sensor (TES):

- sensitive film thermometer operated below and close to its transition (critical) temperature T_c (sub-K)
- energy deposited onto TES is transformed to heat which increases TES resistance.
- steep change of resistance with temperature, low operation temperature and narrow bandgap of superconductors => high sensitivity for different applications
- change in resistance causes change in current flowing through TES. This change is read out by a SQUID.

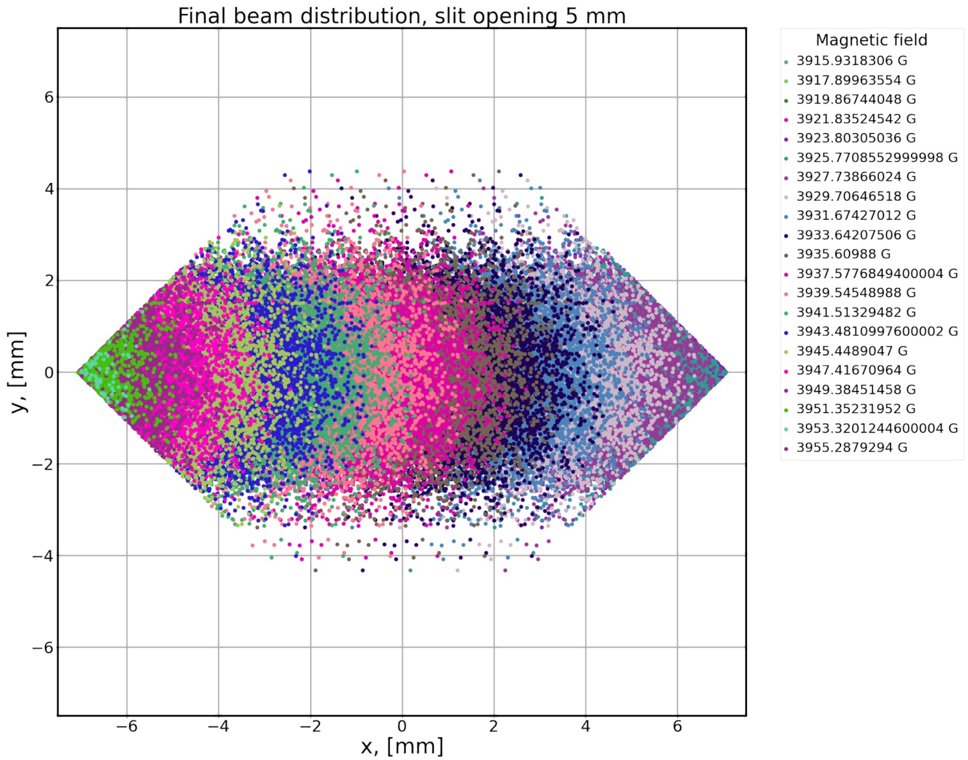


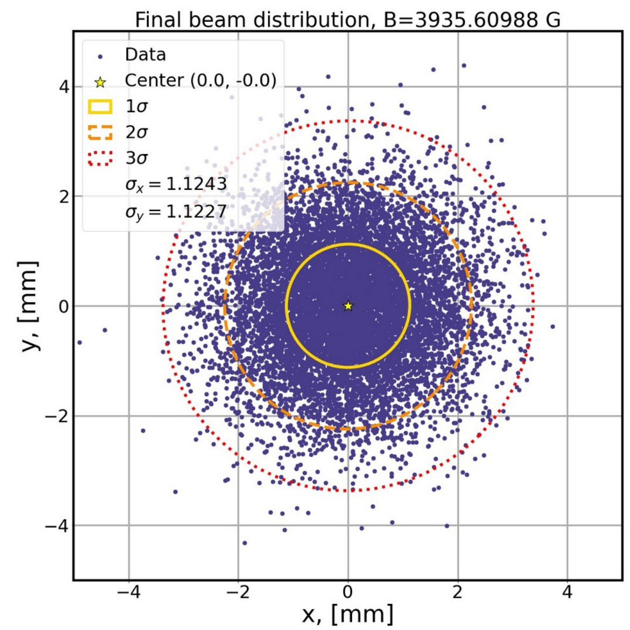


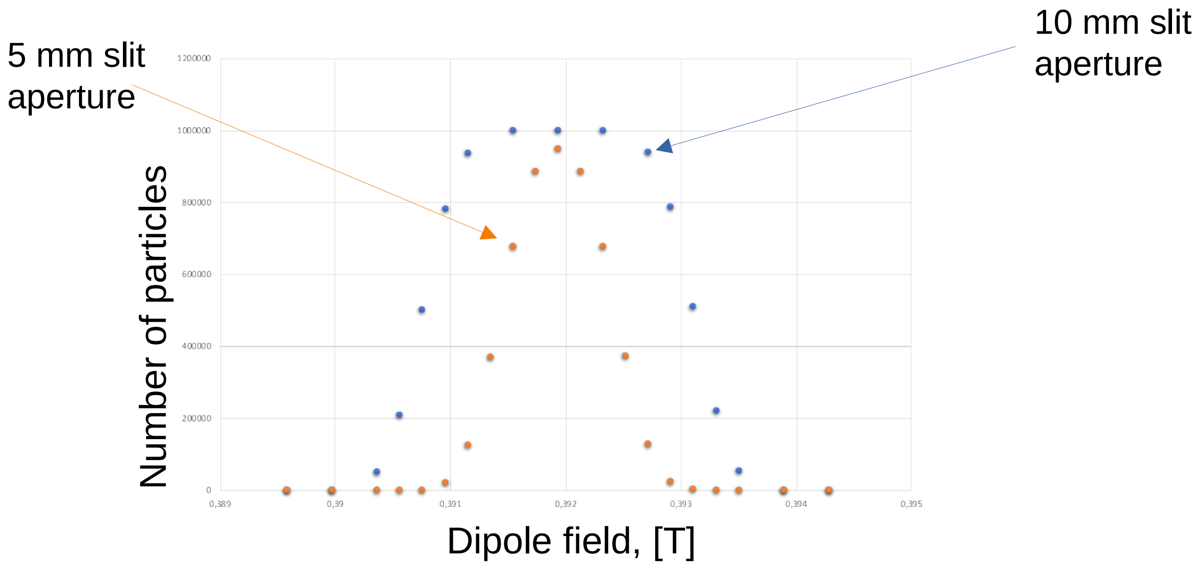




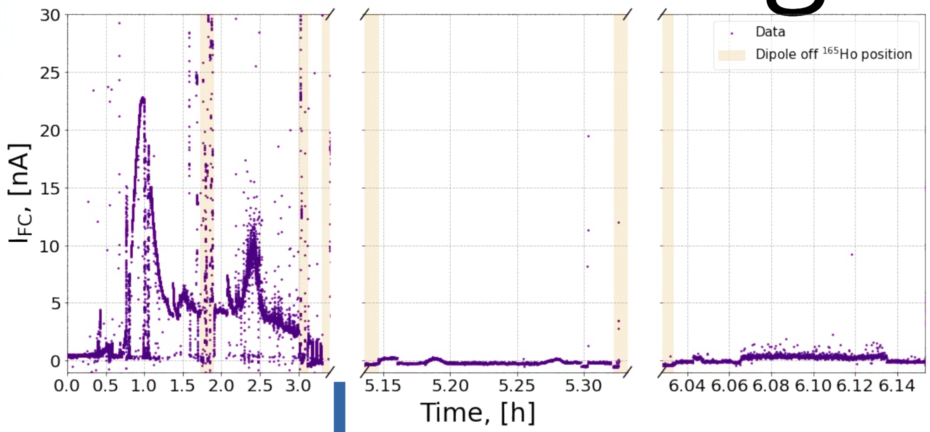
Simulation of ion beam in *elegant* software

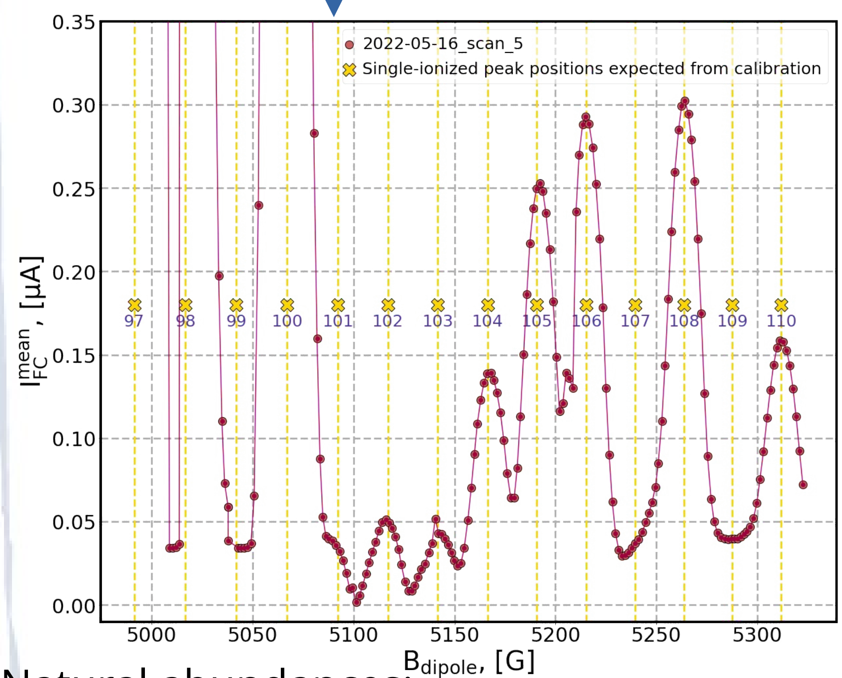






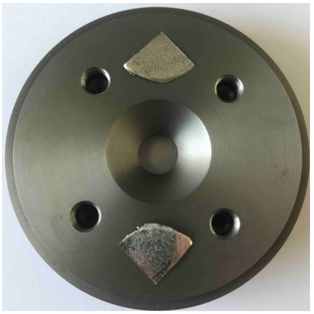
Different target types with ^{165}Ho





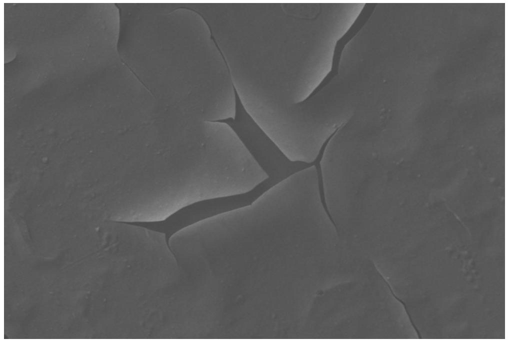
Coupled reduction of $Ho(NO_3)_3$ on Pd

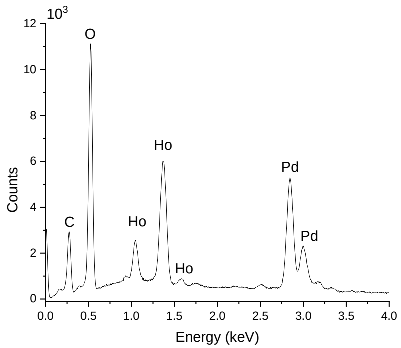
PSI



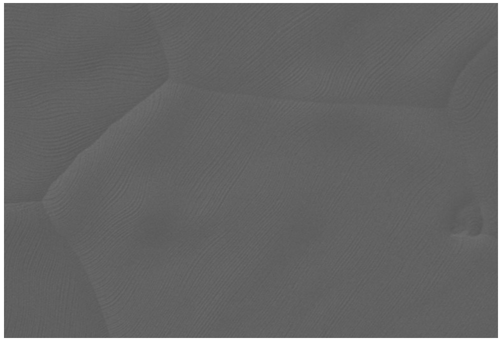
Characterization of test sample (not target)

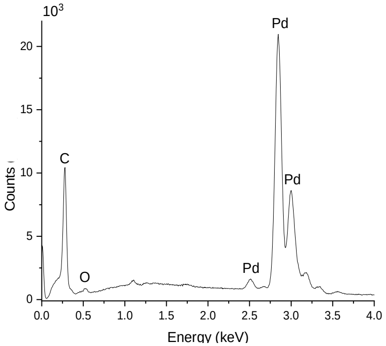
Before CR (after MP)





After CR





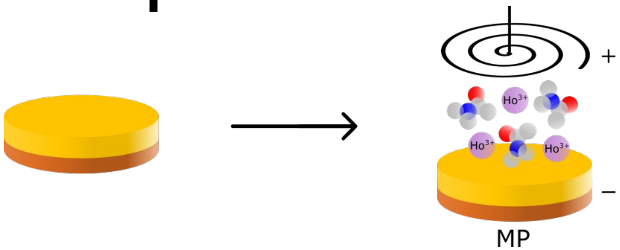
Natural abundances:

^{92}Mo: 14.84% ^{94}Mo: 9.25% ^{95}Mo: 15.92% ^{96}Mo: 16.68% ^{97}Mo: 9.55% ^{98}Mo: 24.13% ^{100}Mo: 9.63%

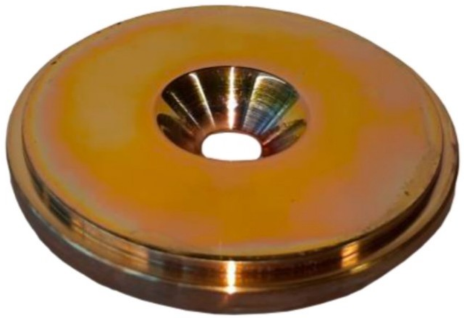
^{102}Pd: 1.02% ^{104}Pd: 11.14% ^{105}Pd: 22.33% ^{106}Pd: 27.33% ^{108}Pd: 26.46% ^{110}Pd: 11.72%

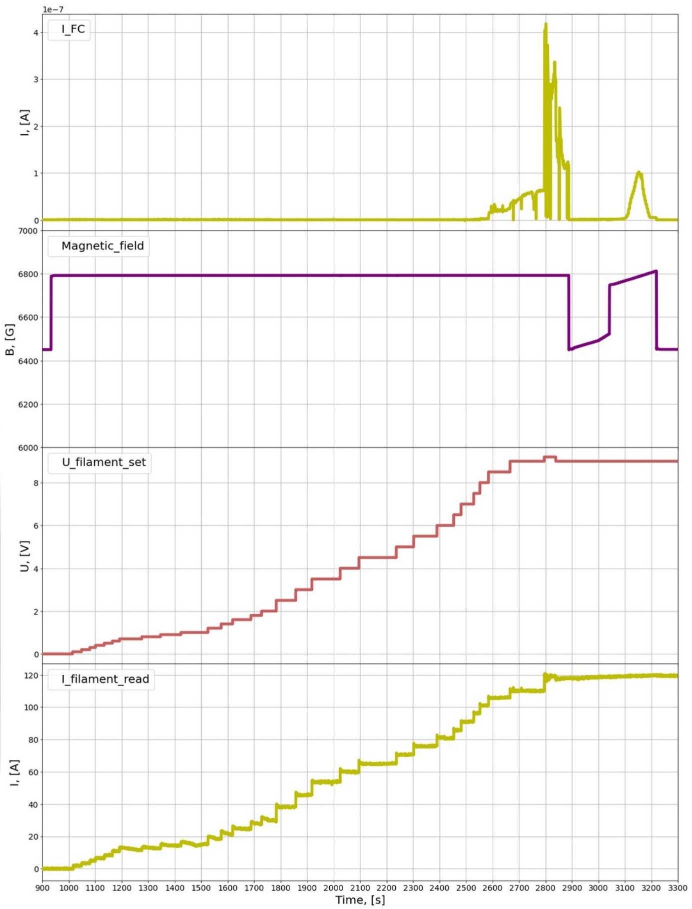
Targets with ^{165}Ho compound on surface (PSI)

Molecular plating (MP)
$Ho(OH)_3$
PSI

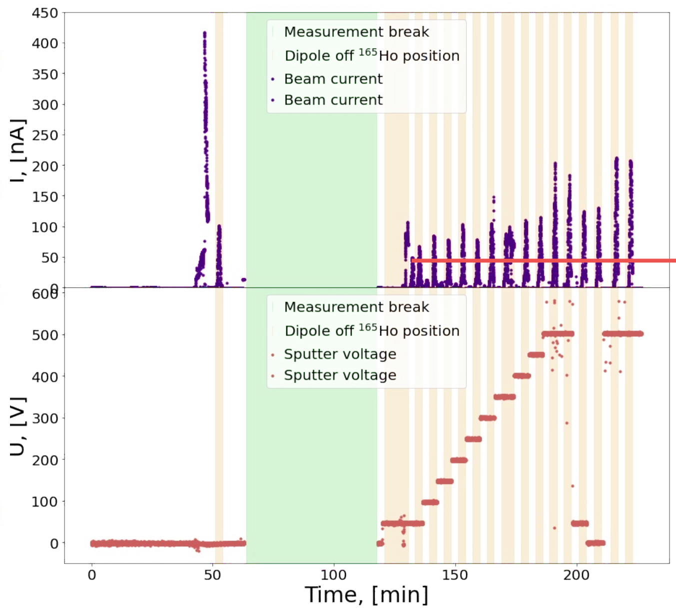


- deposition of Ho complexes in an organic solvent at high voltages with high uniformity and efficiency

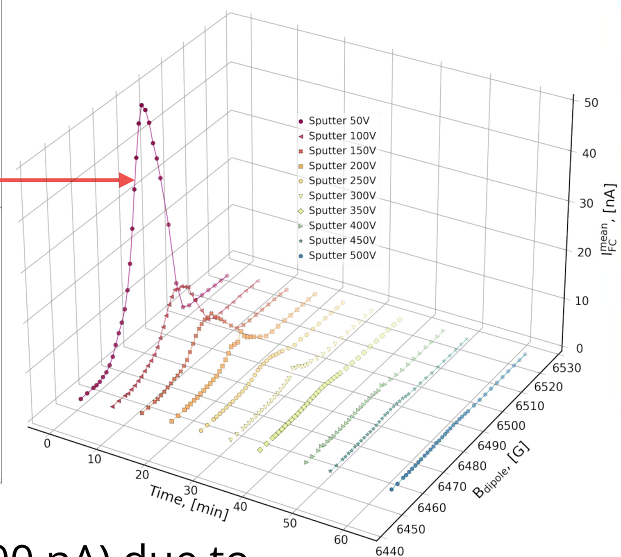




Sample 2:
Au 100 nm + Ti 18 nm sticking layer on Cu bulk

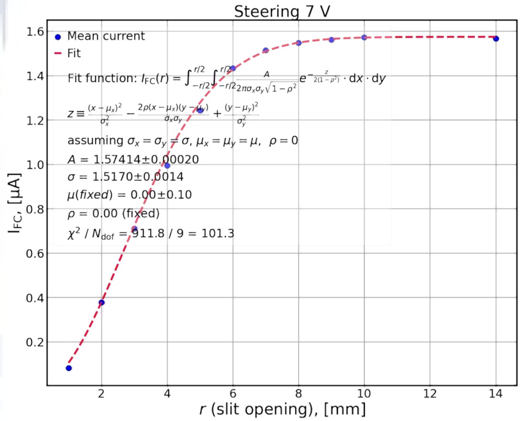


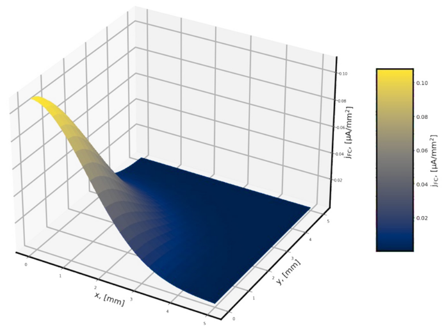
^{165}Ho peak



- Initial $^{165}Ho^{+}$ current spike O(400 nA) due to filament warming, then from ~50 nA vanishing over 30 min

Fit of the slit aperture scan data





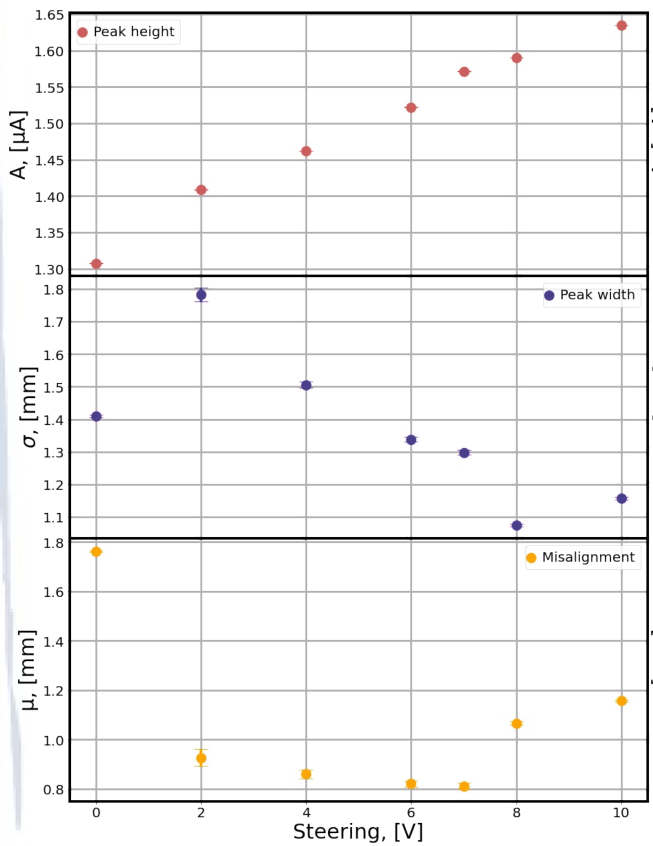
7V: fixed μ=0
→ σ

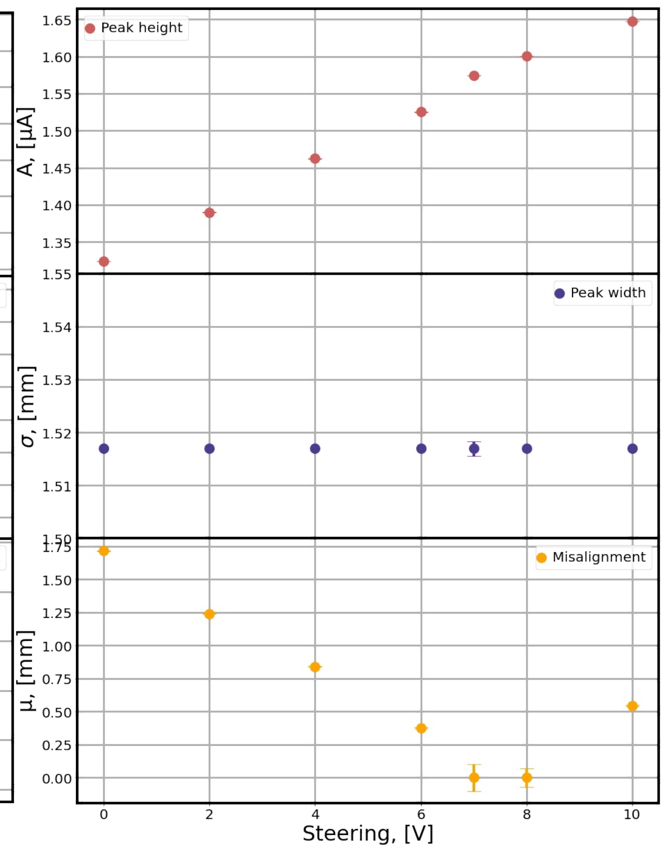
other settings:
σ fixed from 7V,
A and μ free

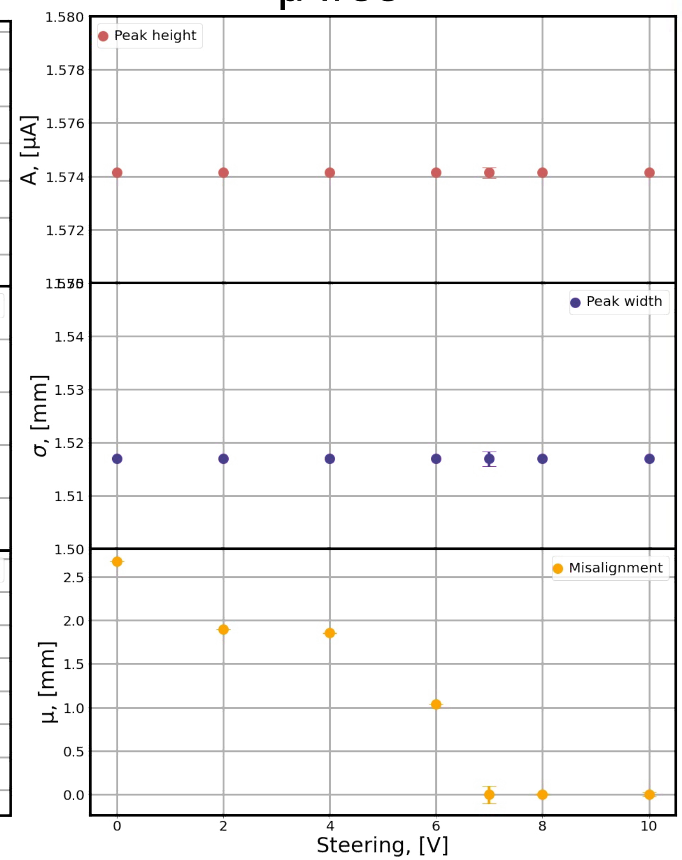
7V: fixed μ=0
→ A, σ

other settings:
σ, A fixed from 7V,
μ free

A, σ, μ free







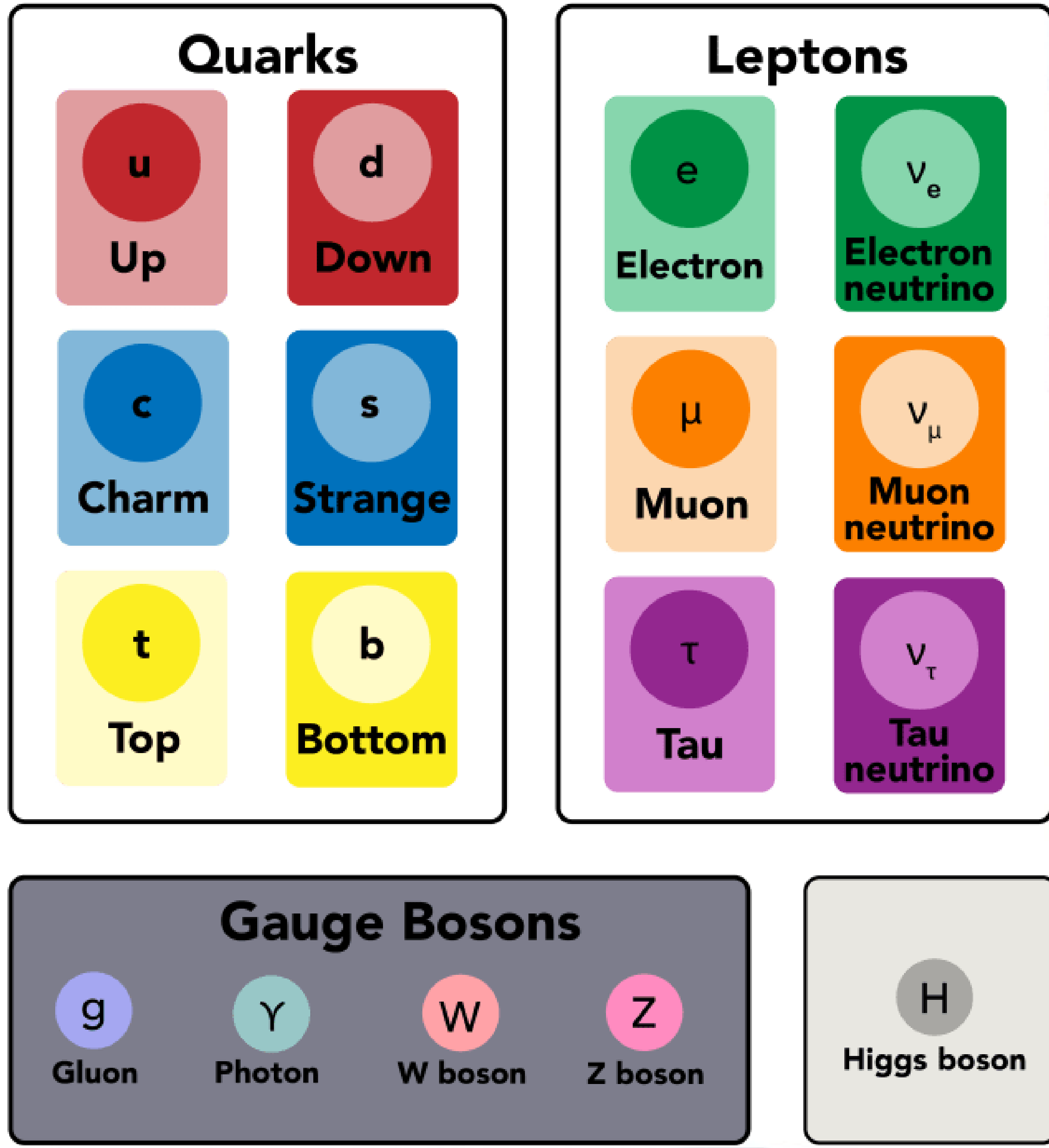
Neutrinos in the Standard Model

Neutrinos in Standard Model:

- Massless
- Left-handed
- Only weak interactions (no electromagnetic, strong)
- 3 flavour states: ν_e, ν_μ, ν_τ

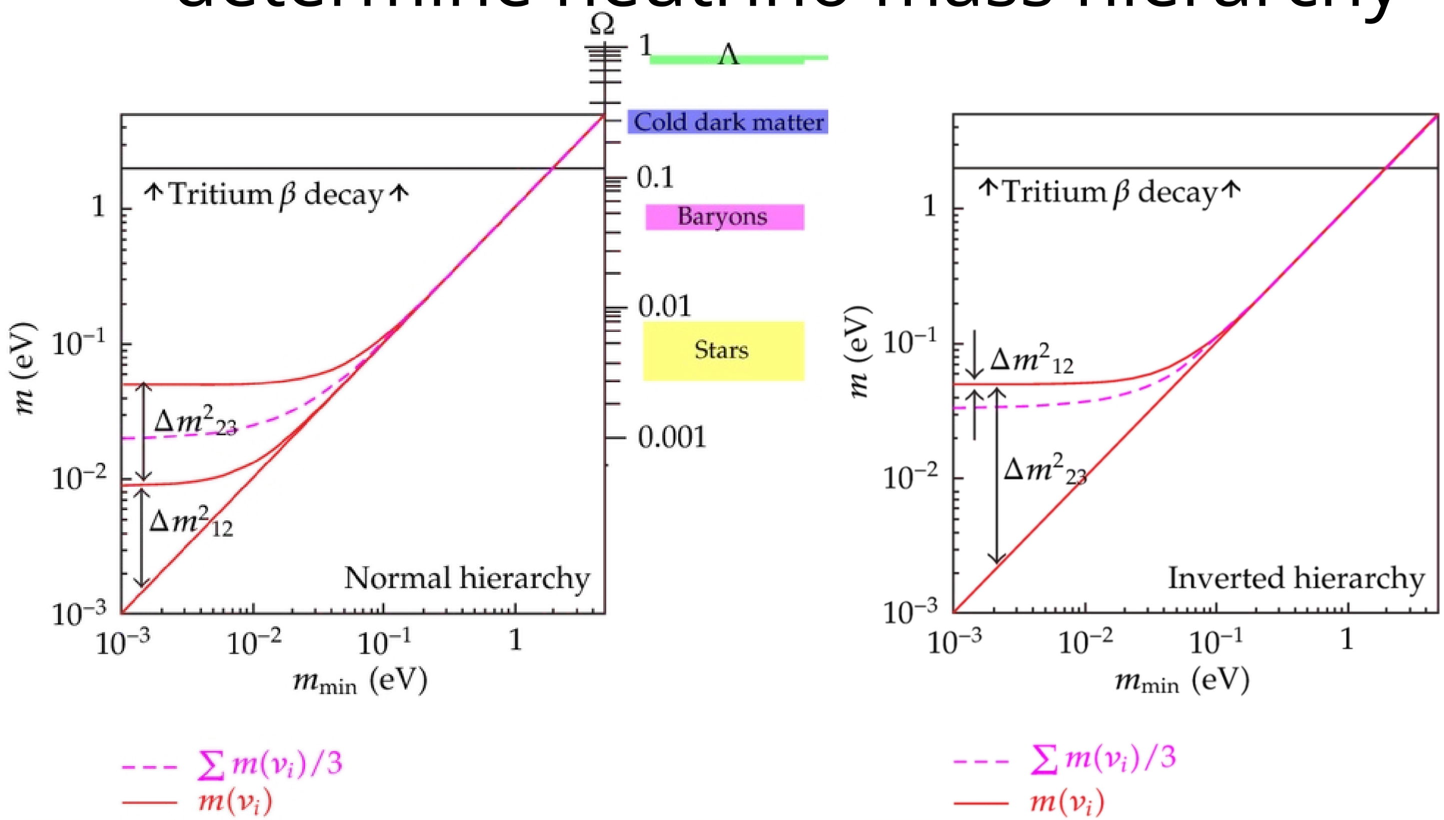
Beyond the Standard Model:

- Neutrino oscillation experiments: neutrinos have mass (no knowledge on the absolute scale)

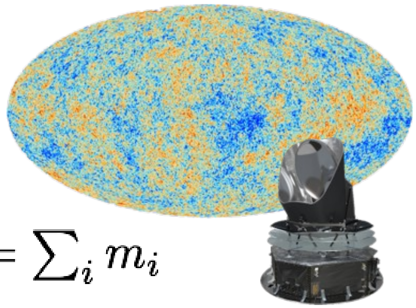


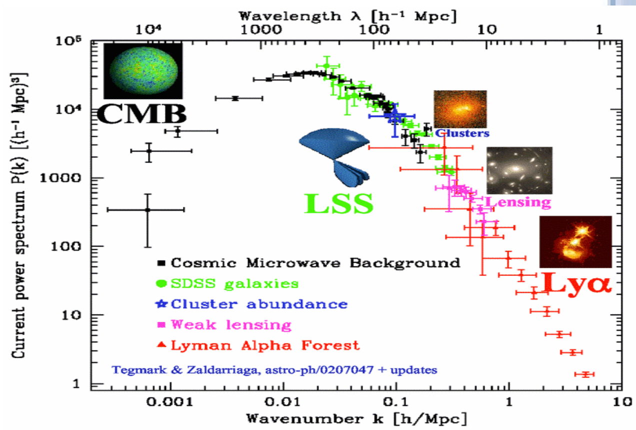
Why measuring neutrino mass?

- origin of fermion masses
- Standard Model extension
- important parameter in cosmology
- determine neutrino mass hierarchy



3 ways to probe neutrino mass





Cosmology

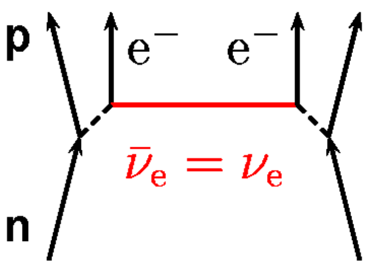
- Very sensitive, but model-dependent
- Present upper limit: 0.12 eV – 0.26 eV

$$M_\nu = \sum_i m_i$$

0νββ decay

- Sensitive to Majorana neutrinos, model dependent
- Present upper limit 0.12-0.4 eV

$$m_{\beta\beta}^2 = \left|\sum_i U_{ei}^2\, m_i\right|^2$$



Direct measurement

- Use $E^2 = p^2c^2 + m^2c^4 \rightarrow m^2(\nu)$
- Time-of-flight measurements from supernova, model-dependen…
- SN1987a (Large Magellan Cloud) : m(νe) < 5.8 eV
- Kinematics of weak decays/ β-decays - model independent!
- β-decay searchs for m(νe) :

$$m_\beta^2 = \sum_i |U_{ei}|^2\, m_i^2$$

  - tritium (KATRIN)
  - [163]Ho electron capture (ECHo, HOLMES, and NuMECS)
- Present upper limit: 1.1 eV (KATRIN)