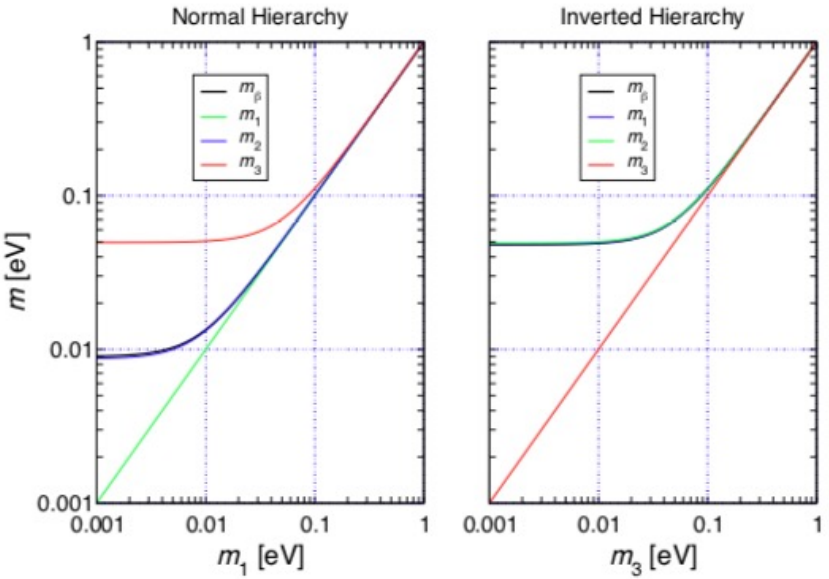
Status of the HOLMES experiment: commissioning of the ion implanter .

Giovanni Gallucci – INFN Genova
On behalf of the HOLMES collaboration

erc European Research Council
INFN Istituto Nazionale di Fisica Nucleare
UNIVERSITA' DEGLI STUDI DI MILANO BICOCCA
UNIVERSITÀ DEGLI STUDI DI GENOVA

The importance of ν-mass

- Understand better the origin of fermion masses
- Extend the Standard Model
- Improve cosmological models (ν-mass affects the large-scale structure and dynamics of the universe)
- Determine neutrino mass hierarchy
- etc…



- From direct measurement :$\lesssim 1.1\ eV$ (KATRIN experiment)
- From Neutrinoless double beta decay: $\lesssim 0.5\ eV$ (only Majorana neutrino)
- From cosmological and astrophysical data : $\lesssim 0.2 - 1.3\ eV$ (model dependent)

Spectrometry vs Calorimetry

General requirements for a ν mass experiment:
- High statistics near the end point
- Low Q-value (stat ~$1/Q^3$)
- High activity/efficiency of the source
- Energy resolution order ~eV or below (comparable with m_ν)
- small systematic effects

Spectroscopy: external source
- high statistics
- high energy resolution (below eV)
- systematics due to the source (energy loss)
- systematics due to decay to excited states
- background

Calorimetry: 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

Electron Capture (EC) in ^{163}Ho

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

$$\frac{d\lambda_{EC}}{dE_c} = \frac{G_\beta^2}{4\pi^2}(Q - E_c)\sqrt{(Q-E_c)^2 - m_\nu^2} \times \sum n_i C_i \beta_i^2 B_i \frac{\Gamma_i}{2\pi} \frac{1}{(E_c - E_i)^2 + \Gamma_i^2/4}$$

Q~2.8keV, capture only from shell ≥ M1
De Rujula & Lusignoli, Phys. Lett. B 118 (1982) 429

same factor as β decay
(total de-excitation energy E_c instead of E_e)

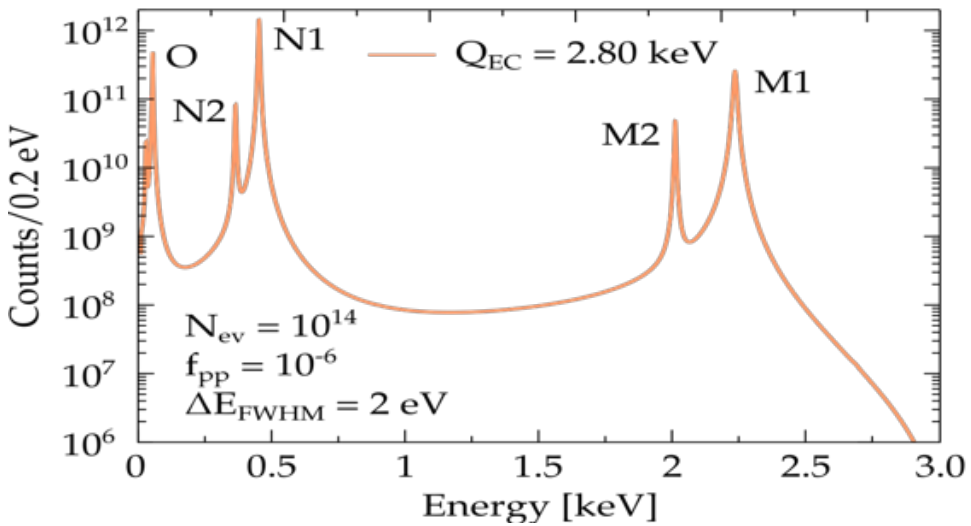
Breit-Wigner shape

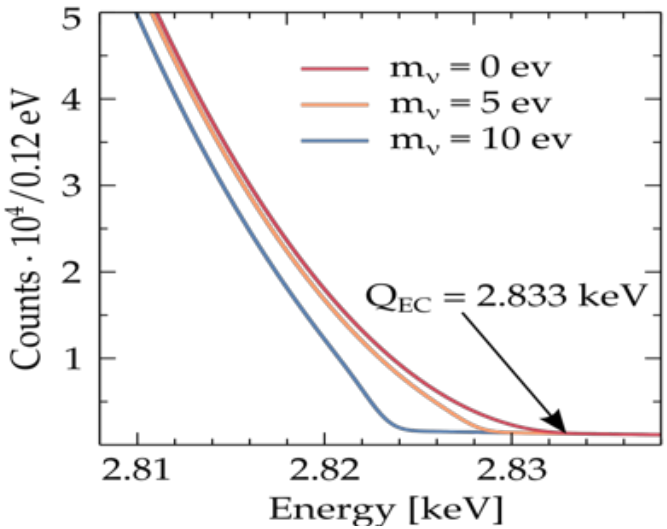
Calorimetric measurement of Dy atomic de-excitations

Q **≈ 2.83 keV** (recently measured with Penning trap)

Rate on the end point depends on (Q-M1): the proximity to M1 resonance peak enhances the statistics at the end point

$\tau \approx$ **4570years** → 2x10^{11} Ho nuclei↔1Bq





HOLMES collaboration

Univ. Milano-Bicocca
INFN Milano-Bicocca
M.Borghesi
M.Faverzani
E.Ferri
A.Giachero
A.Nucciotti
L. Origo
G.Pessina
S.Ragazzi (ERC PI)

INFN Genova
M.Biasotti
G.Gallucci
M.De Gerone
M. Fedkevych
F.Gatti

INFN Roma
M.Lusignoli

INFN LNGS
S.Nisi

NIST
B.Alpert
D.Becker
D.Bennett
J.Fowler
J.Gard
G.Hilton
J.Mates
C.Reintsema
D.Schmidt
D.Swetz
J.Ullom
L.Vale

PSI
R.Dressler
E.Maugeri
D.Schumann

ILL
U.Koester

CENTRA-IST
M.Ribeiro-Gomes

Holmes experiment in a nutshell

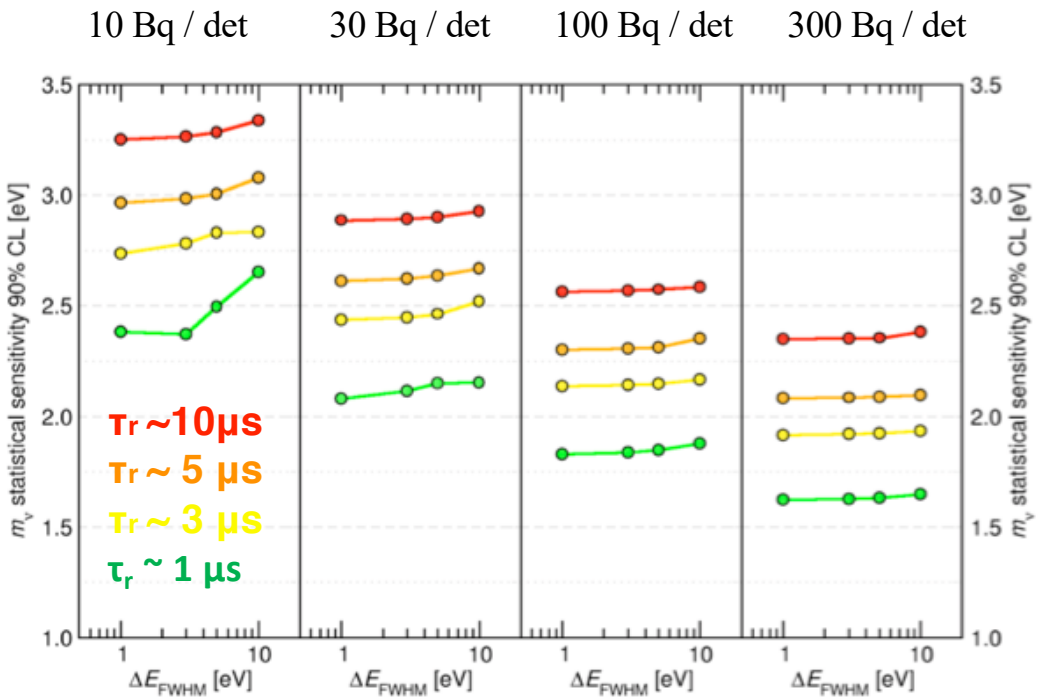
Direct neutrino mass measurement with statistical sensitivity ~ eV.

Usage of Transition edge sensor (TES) based micro-calorimeters with ^{163}Ho implanted and Au absorber:

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

Should prove the technique potential and scalability by:

- assessing EC spectral shape
- assessing systematic error
- sensitivity on m_ν ~ eV



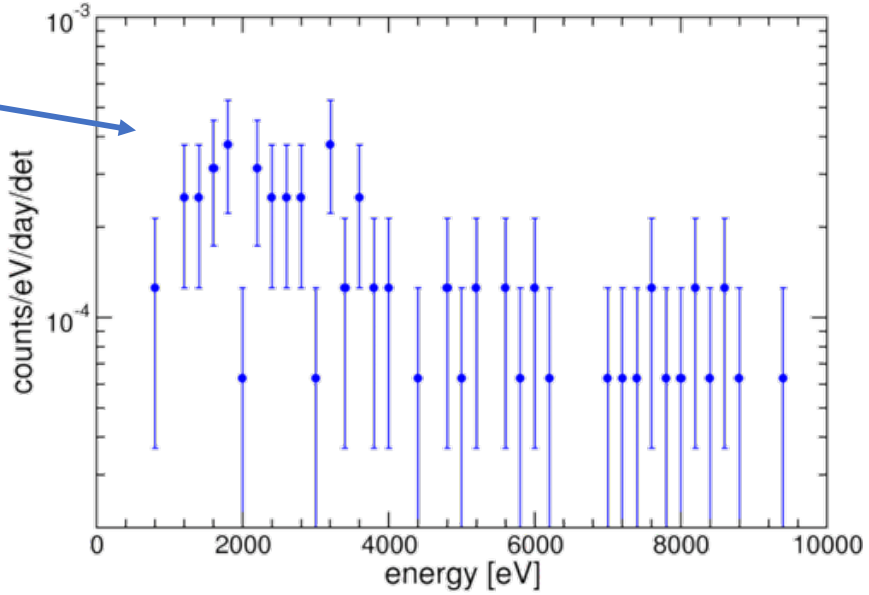
exposure: 1000 det x 3 years

BACKGROUND

HOLMES baseline: ^{163}Ho pile-up rate
$\langle rpp \rangle = A \cdot fpp/2Q$ = 300 Bq x $3 \cdot 10^{-4}/2Q$ =
1.5 counts/eV/day/det

External sources
- Environmental γ radiation
- γ, X and β from close surroundings
- cosmic rays

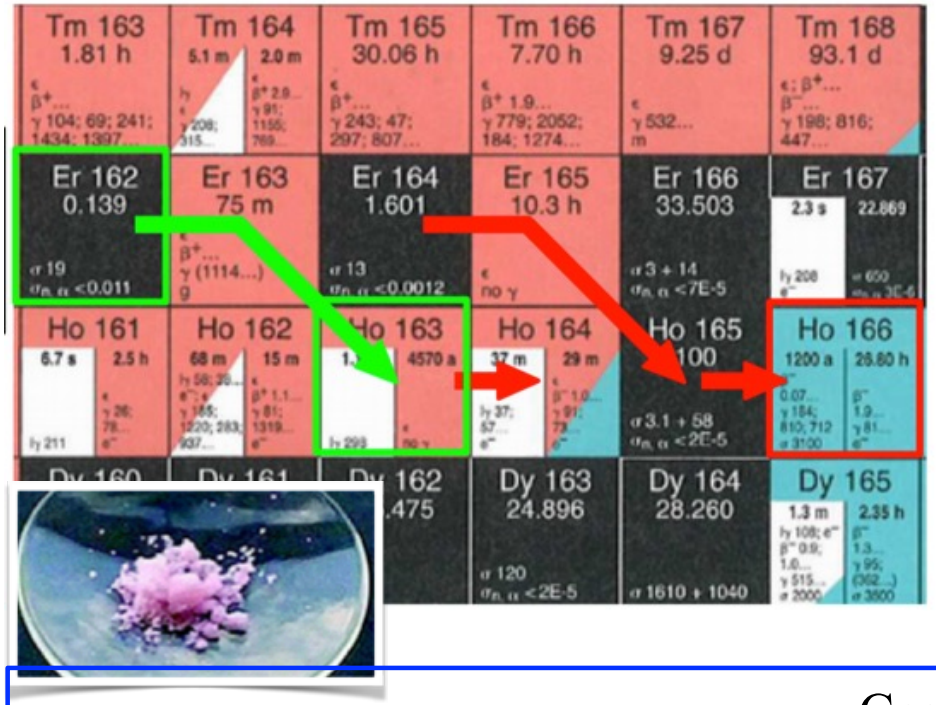
Measured 200x200x2 μm^3 Au absorber (Holmes-like)
→ bkg(1-10 keV) ≈ 5×10^{-3} counts/eV/day/det



Internal source
radionuclides (byproduct of Ho production)
^{166m}Ho, (β^-, Q = 5970 keV, $t_{1/2}$ = 1200 y)
GEANT4 simulation for 200x200x2 mm^3 Au absorber produce
bkg 10^{-11} counts/eV/day/det /^{166}Ho Nucleus

If A(300Bq) and requiring bkg <0.1 counts/eV/day/det
N(163)/N(166) >6000
A(163)/A(166)>1500

163-Holmium 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/cm2/s

$$^{162}\mathrm{Er}\,(n,\gamma)\,^{163}\mathrm{Er} \qquad \sigma_{thermal} \approx 20\mathrm{b}$$

$$^{163}\mathrm{Er} \rightarrow {}^{163}\mathrm{Ho} + \nu_e \qquad \tau_{1/2}^{EC} \approx 75\mathrm{min}$$

high yield (but not all cross sections are well known)

Contaminants:

1. Other elements (residual Er, rare earth contaminants, decay product, etc…)
2. Holmium isotopes, in particular ^{166m}Ho

$A(^{163}\mathrm{Ho})/A(^{166m}\mathrm{Ho}) = 100\text{-}1000$

1) Chemical purification at PSI;
2) Isotopic separation using an implanter.

Holmium chemical purification

Three different batches are produced:
1) 25 mg irradiated for 55 days (2014), A(^{163}Ho) ≈ 5MBq(A(^{166}mHo)≈10kBq)
2) 150 mg irradiated for 50 days (2015), A(^{163}Ho) ≈38MBq(A(^{166}mHo)≈37kBq)
3) 540 mg irradiated 50 days(2017),A(^{163}Ho)≈130MBq(A(^{166}mHo)≈180kBq) (~500 detectors)

The Er/Ho mixture is subjected to a radiochemical separation with ion-exchange resins in hot-cell at PSI.

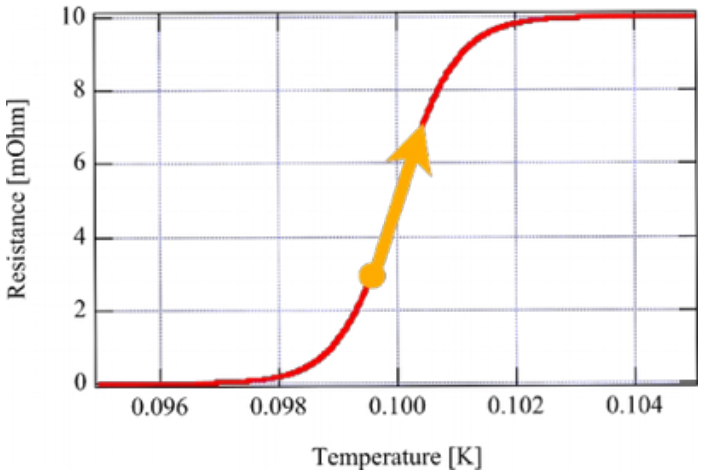
Efficiency > 80%

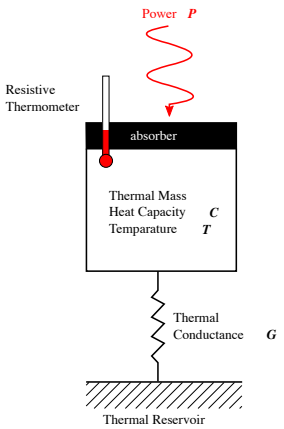
S. Heinitz *et al.*. PLoS One 13 (2018) no.8, e0200910

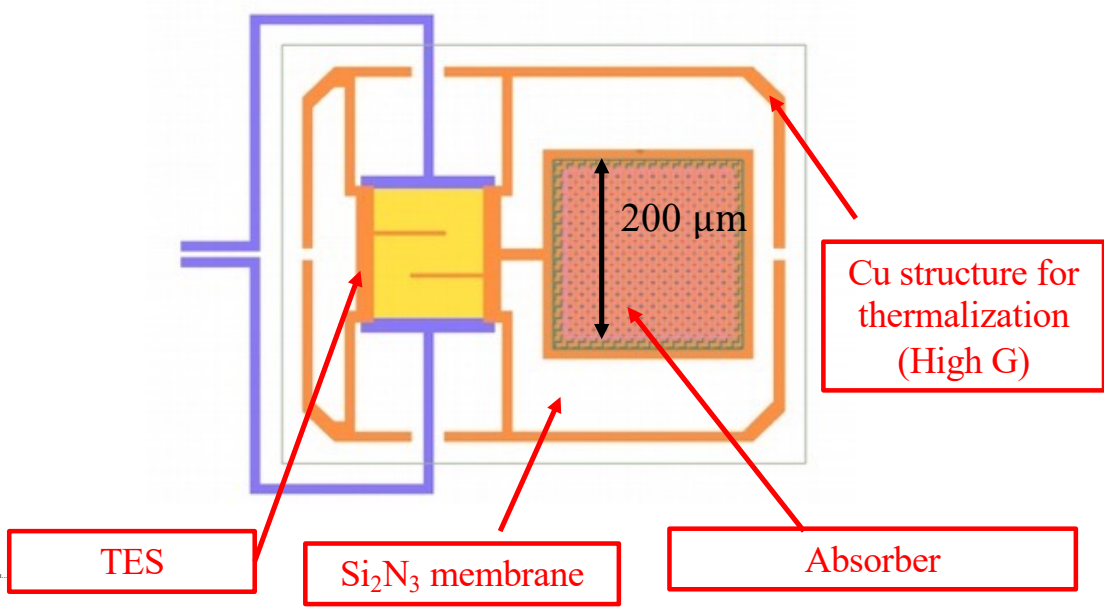
Holmes detector (1)

Transition Edge Sensors Superconductive Detectors (TES)

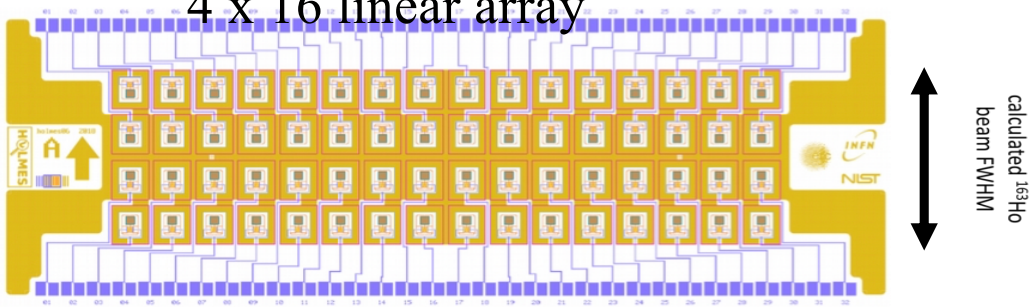
- Molybdenum/copper (T_c ~ 100 mK)
- Very steep R vs T dependency in transition region;
- Gold absorber with ^{163}Ho inside coupled to TES thermometer ;
- Ho sandwiched between two 1 μm thick gold layers for a total electron containment
- Fast detectors to reduce pile-up
  - tunable rise time ~ L/R
  - decay time dependent on detector characteristics C/G





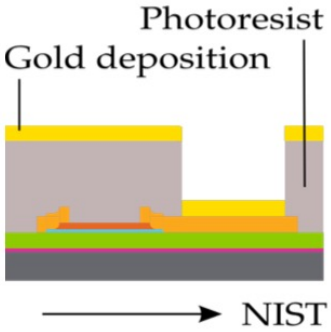


4 x 16 linear array



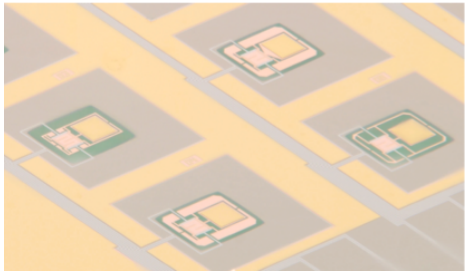
Holmes detector (2)

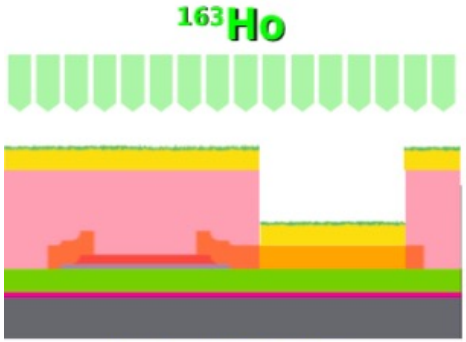
TES design, production and preliminary test is done @NIST



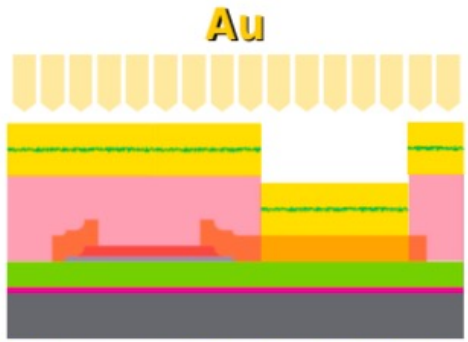
- Holmium implantation and gold co-evaporation (implanter)
- Final 1 μm Au deposition
- Lift-off of photoresist and membrane release

(several procedures tested, work in progress)

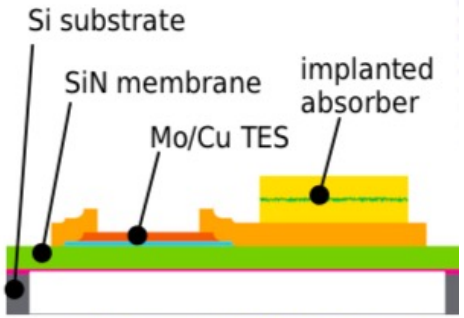




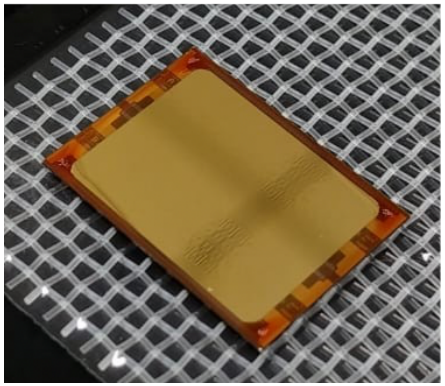
ion implantation



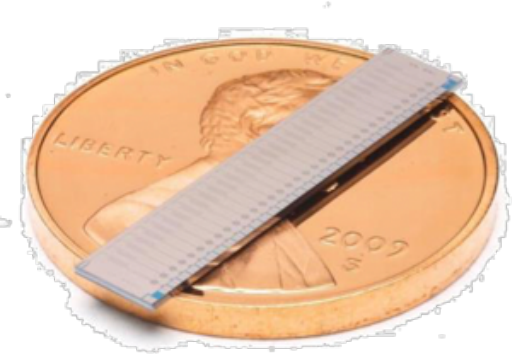
full encapsulation by sputtering



photoresist lift-off and SiN membrane release



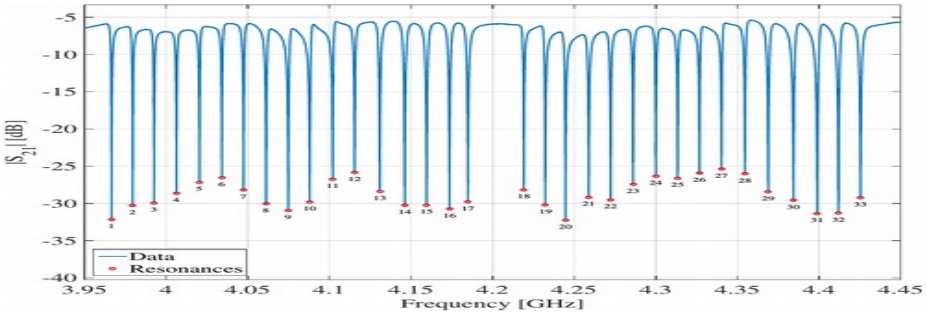
Detector readout and test



The detector array is with a microwave multiplexing system (ROACH2):

- Each TES is coupled to an RF-Squid
- Each RF-Squid is coupled to a common ramp
- Each RF-Squid is coupled to a resonant circuit (frequency multiplexing)
- 33 resonances in 500 MHz, width 2 MHz, separation 14 MHz

A comb of signals probe the resonators at their characteristic resonant frequency

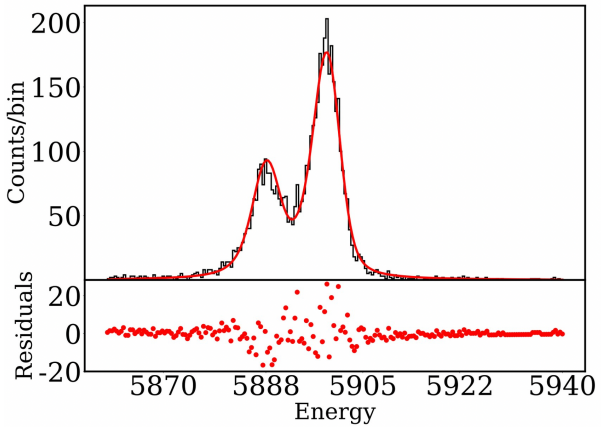


D. T. Becker at al., JINST 14 (2019) 10, P10035

Sources: ^{55}Fe+ fluorescence sources (range 1.6 - 6 keV)

- Energy resolution ~ (4.5 ± 0.3) eV
- Best performing detector: (4.15 ± 0.10) eV @Mn Kα

$\tau_r \sim 1.5\ \mu m$



IEEE Trans.Appl.Supercond. 31 (2021) 5, 2100205

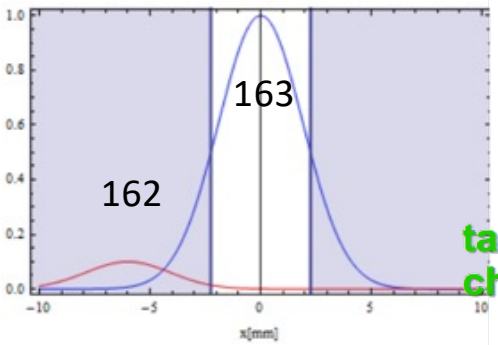
Mass separation and ion implantation

A dedicated ion implanter will be used to remove contamination of holmium isotopes different from ^{163}Ho as well as other impurities.

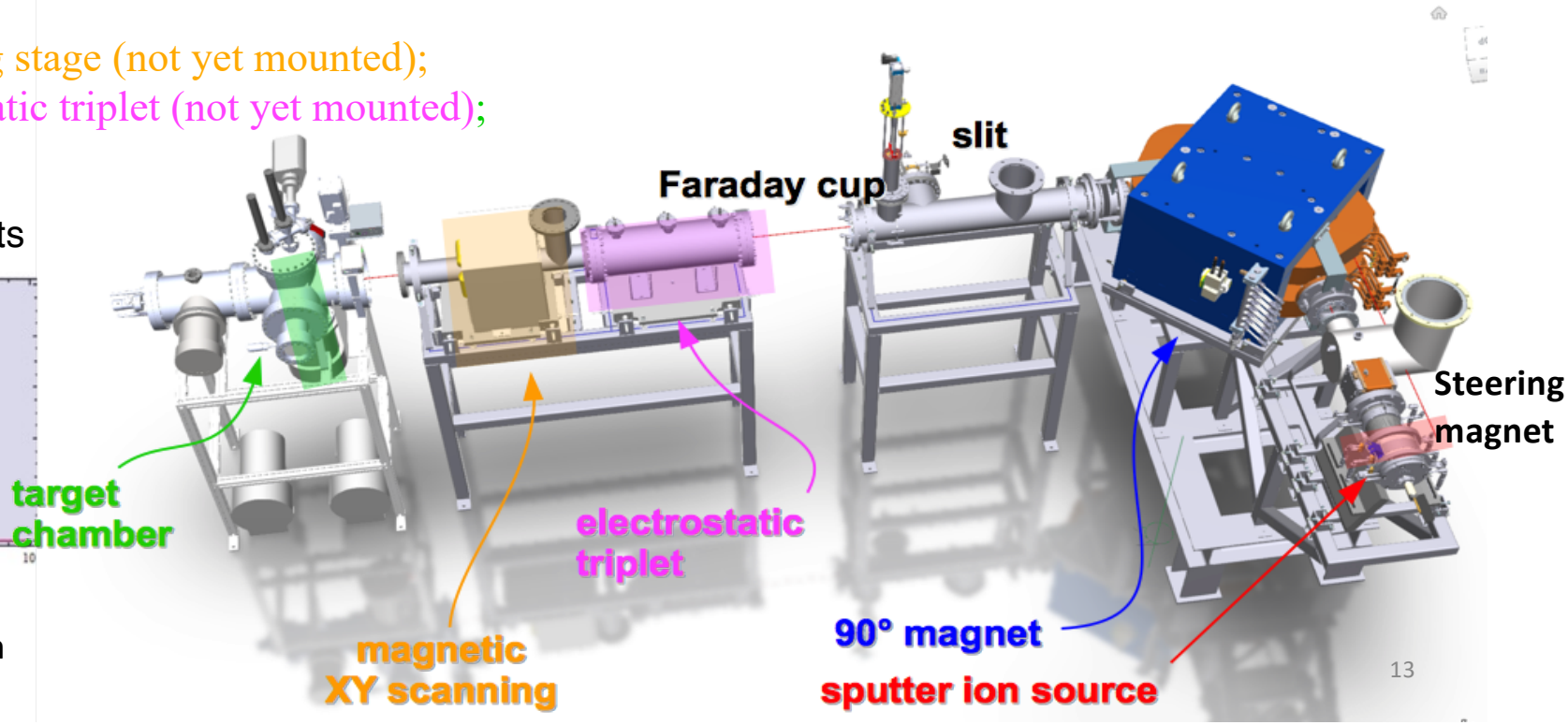
The ion implanter consists of six main components:

1. an argon sputter ion source;
2. an acceleration section to reach the beam energy of 50 keV (~50 nm implantation depth)
3. a magnetic/electrostatic mass analyzer with magnetic field until 1.1 Tesla ^{163}Ho/^{166m}Ho separation better than 10^5
4. a magnetic scanning stage (not yet mounted);
5. a focusing electrostatic triplet (not yet mounted);
6. a target chamber.

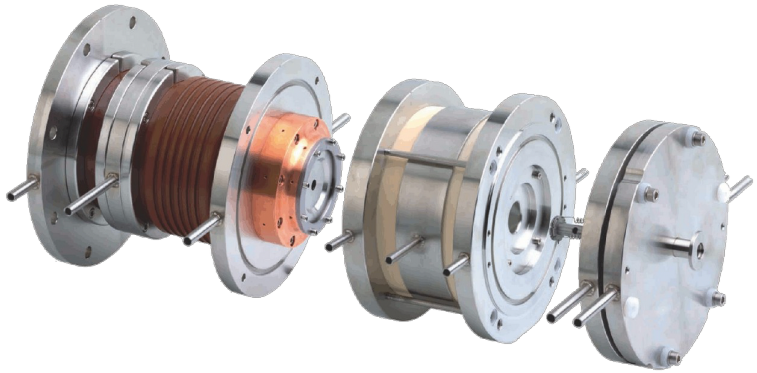
4mm FWHM at the slits

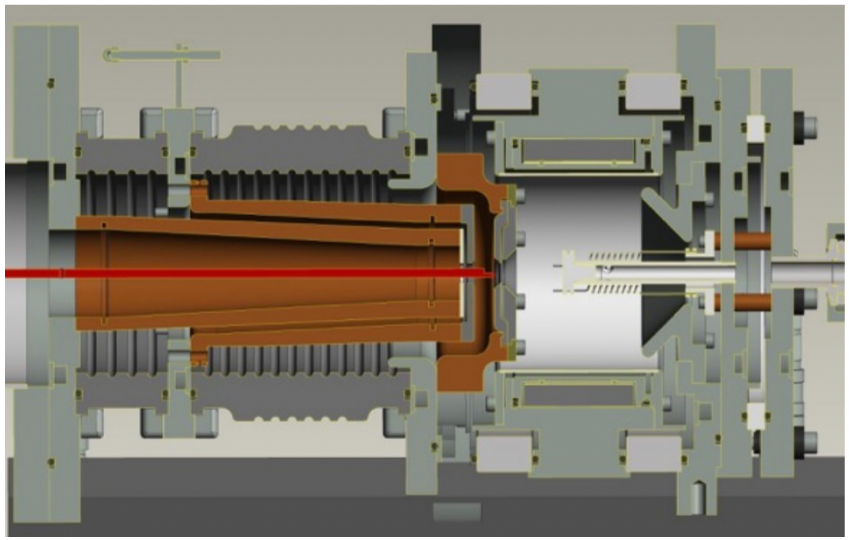


From *elegant* simulation



The ion source





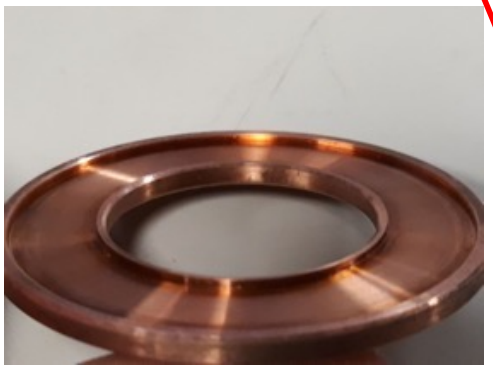
- Sputter gas entrance : controlled argon flow enters inside the sputter chamber. Argon is ionized and argon plasma burns inside the chamber.
- Filament: electrons are emitted by thermionic effect. The electrons ionized argon and sputtered materials
- Sputter target: the sputter target has a tuneable electrical potential (max 600 V) to attract ions.
- Acceleration region: some ions pass through the hole and are accelerated in this region (max 50 kV)
- All parts are water cooled but the sputter target reaches temperatures more than 1000 ^{O}C

The sputter ion source of the implanter needs a metallic cathode.
Two different approaches:
- Sintered target
- Support target with deposited Ho

Sintered target

Sintered targets are developed and produced in collaboration with prof. Manfrinetti (from Chemistry Department of Genoa University)

- Ho metallic powder is produced with a thermoreduction in furnace at T>1600°C
- The Ho powder (5%) is mixed with other materials in fine grained powder (< 40 μm) Ti(36%), Ni(41%), Sn(18%) on a copper support.
- Pressed at 350 bar/cm^2 and heated at 950 °C temperature and at 10^{-4} mbar pressure for 2 days to improve the mechanical properties of the sinter.
- Two different intermetallic compound are created Ti_2Ni_2Sn/HoNiSn

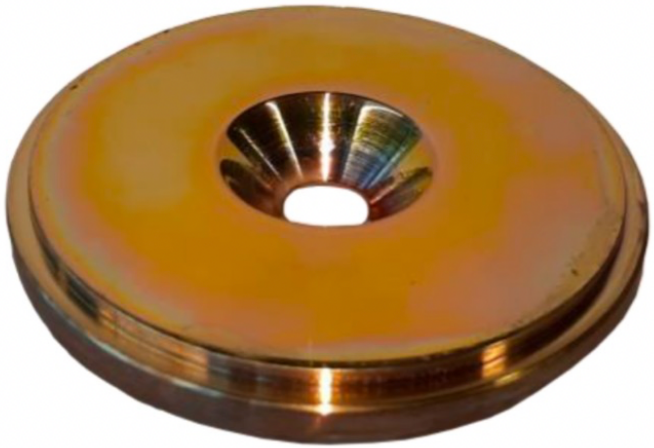


- Holmium is diffused in the entire thickness of the target
- It is possible change mixing materials to obtain different properties
- Careful handling of radioactive powder

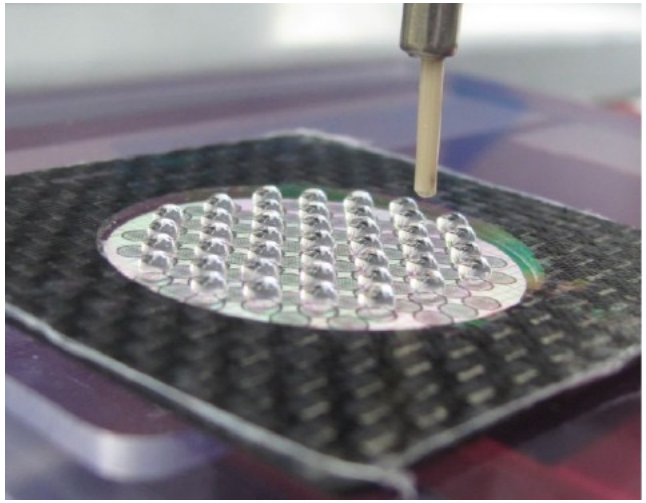
Support target with deposited Ho

Holmium is deposited with different techniques on a Cu target covered by a Ti (20 nm) and Au (100 nm) films. In collaboration with Paul Scherrer Institute (PSI) different targets with different holmium compounds are produced ($Ho(OH)_3$ $Ho(NO3)_3$, $HoCl_3$, HoF_3, Ho_2S_3).

Paper will be published soon



molecular plating



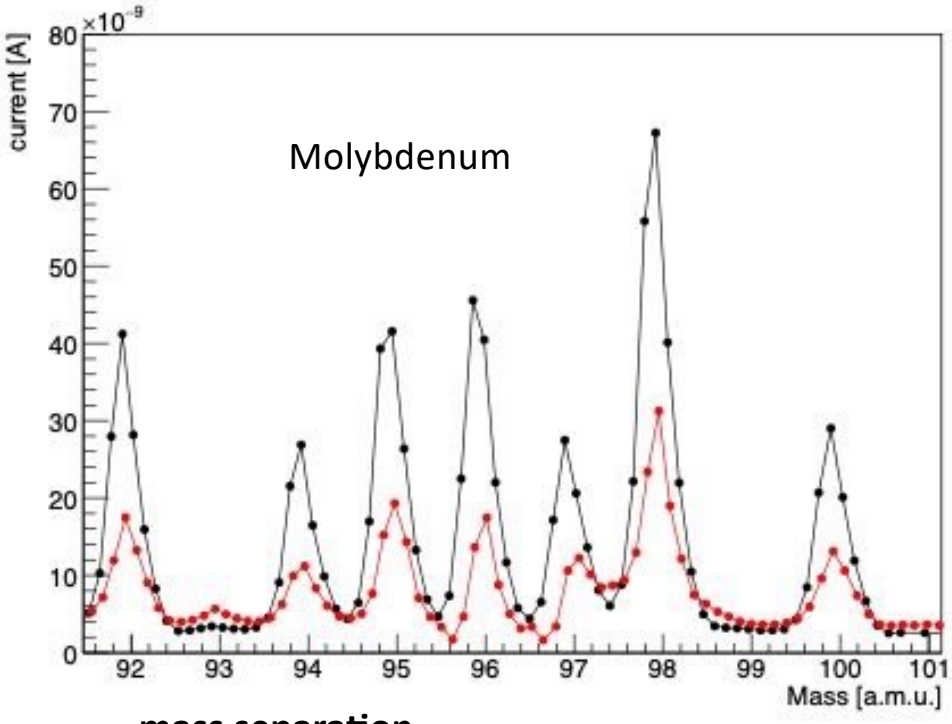
drop-on-demand inkject printing

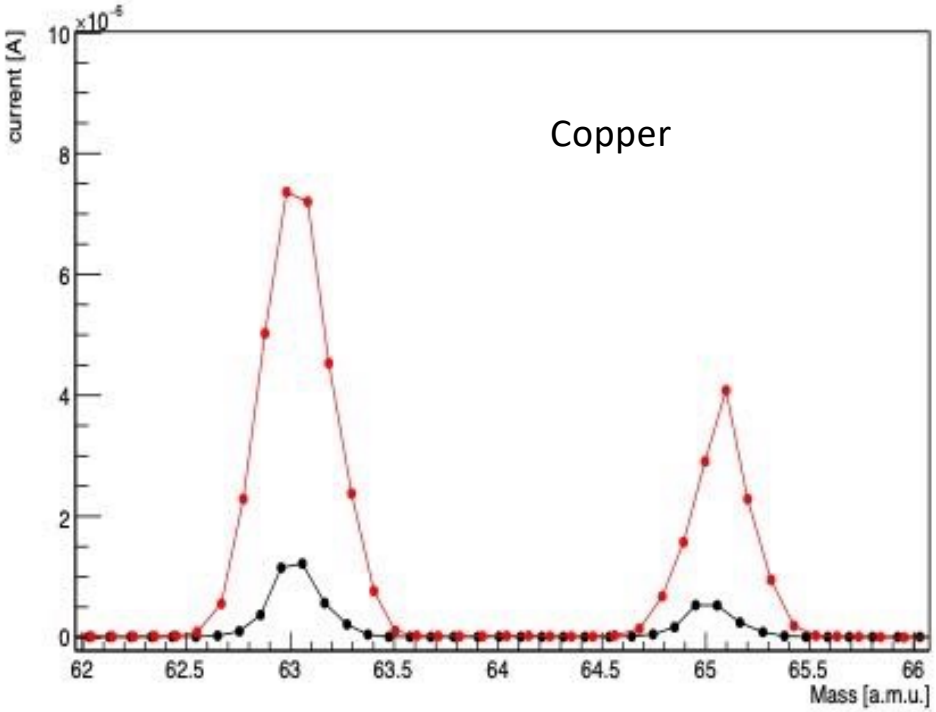
- Safe production
- Different Ho compounds

- Holmium in thin film on surface.
- Possible chemical decomposition of Ho compounds

First tests without holmium

We tested dummies target without holmium (25 keV).
Some outcoming material is not related to the target: molybdenum and iron from the chamber hot parts, tantalum or tungsten from the filament etc.





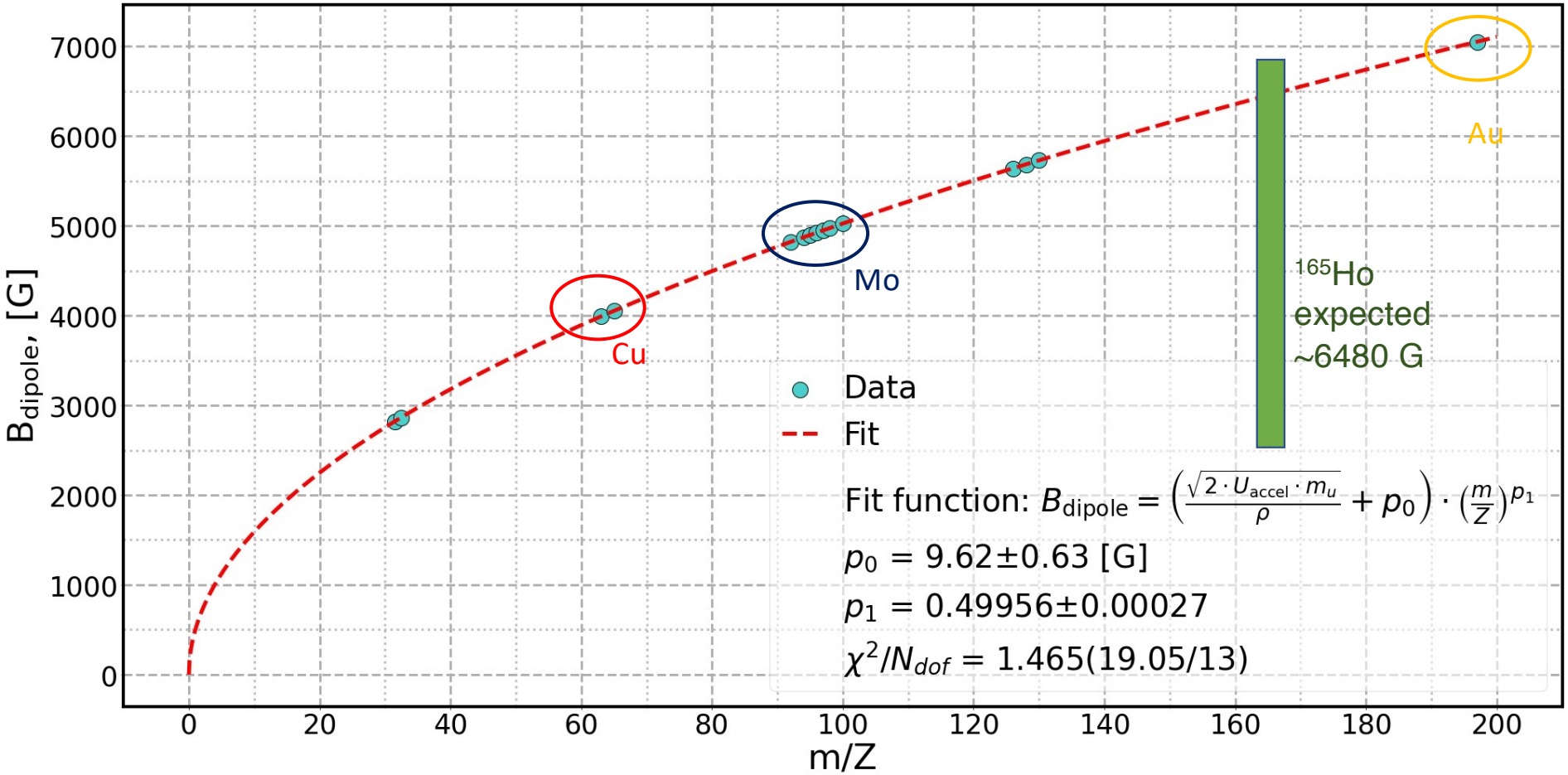
mass separation

63amu - 65amu, ΔB = 62G, Δx = 15mm/amu

Expected holmium isotopes separation: 163amu - 166amu, Δx = 18.8mm

Dipole calibration

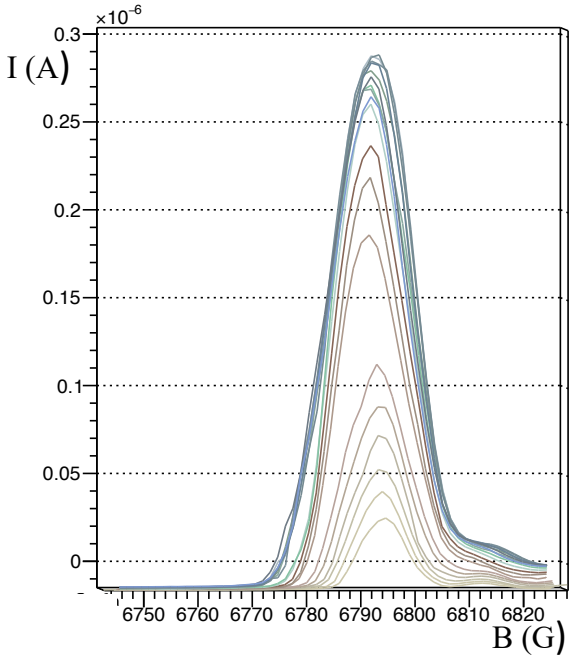
The dipole magnet field is independently measured with a gaussian probe.
The different materials inside the chamber allows a calibration of field vs m/Z curve

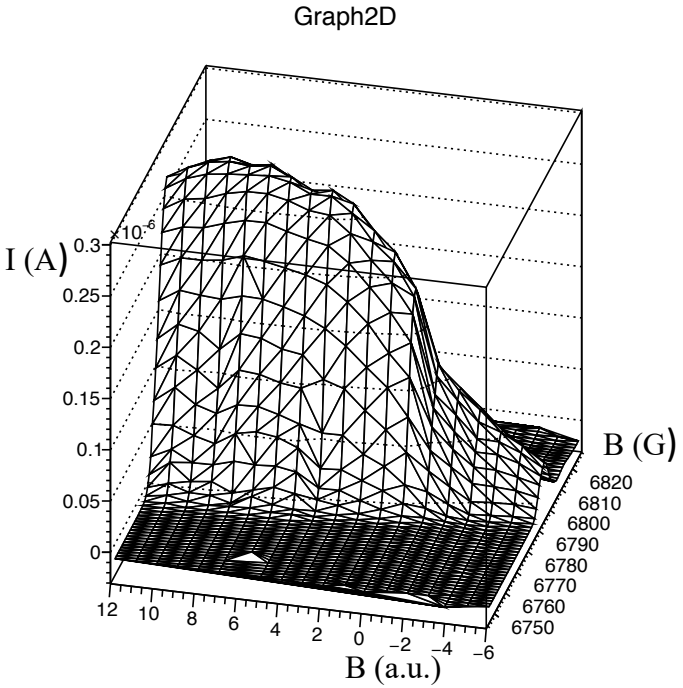


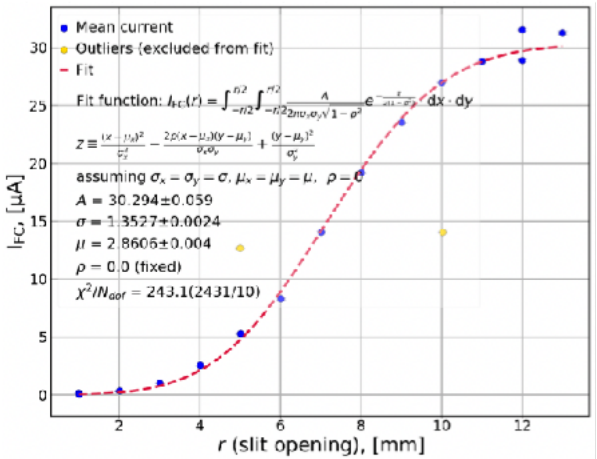
Beam centring and spot

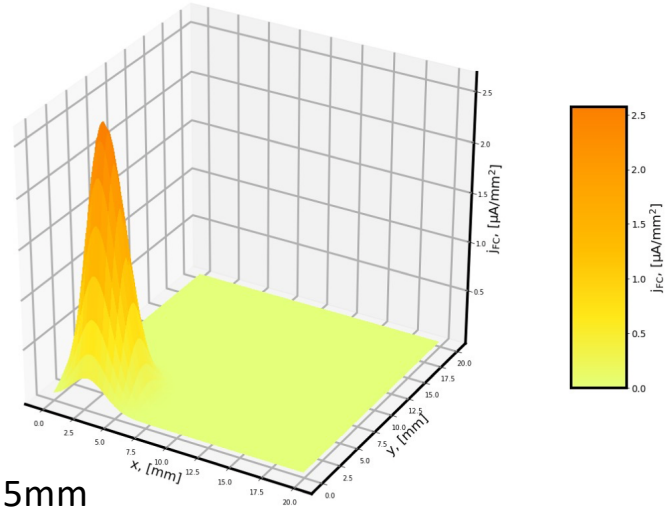
In order to characterize the ion beam exit from source:

- Several measurements are done changing the field of the steerer magnet (vertical position)
- Several measurements are done changing slit opening (from 3 to 15) mm





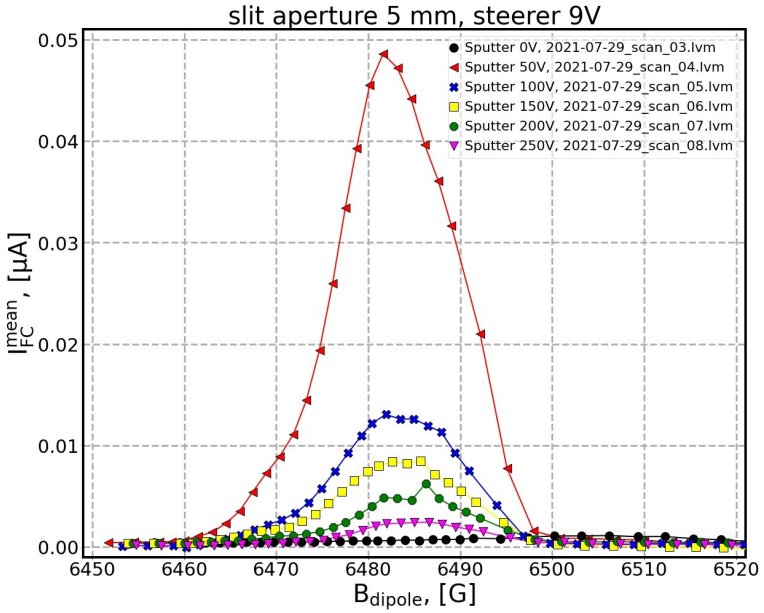




We estimated a vertical misalignment of about 2 mm and a beam profile $\sigma_x \approx \sigma_y \approx 1.5$mm

Misalignment depends on ion mass, other studies and simulation are in progress.

Target with $Ho(OH)_3$: preliminary studies



Tests with other materials and further studies ongoing

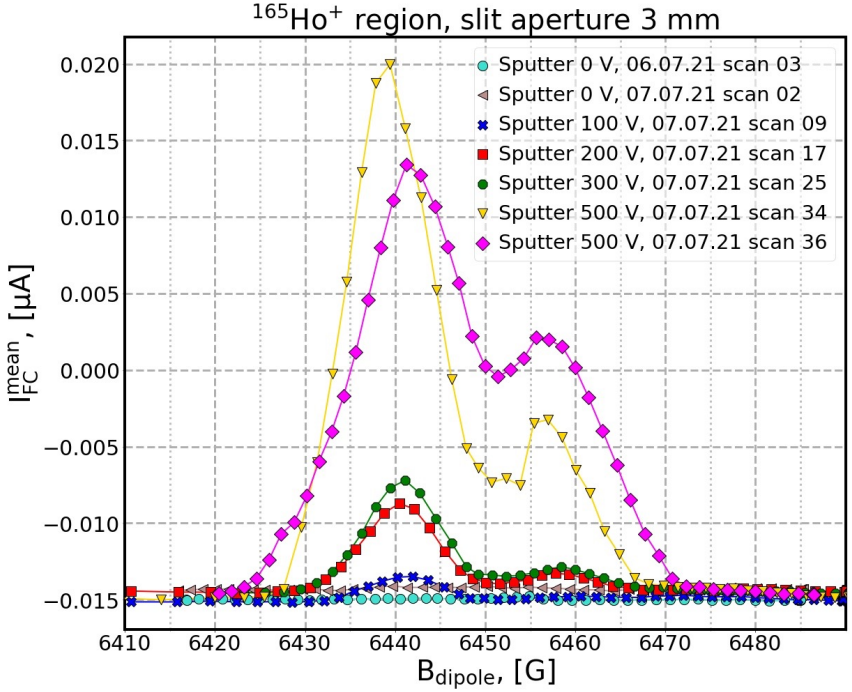
First tests performed with sputter target made with bulk Cu disk covered with 18 nm Ti + 100 nm Au substrate layer with a thin layer of $^{165}Ho(OH)_3$ deposited via molecular plating.

We observed a clear peak at 165 amu but with a low current O(50nA) .
We performed several measurement (~6 minutes each) and observed a decrease of current in about 30 minutes.

Preliminary interpretations:

1. High temperature of sputter target caused chemical dissociation of compound.
2. Sputter splits the $^{165}Ho(OH)_3$ more likely in different parts (HoO, Ho(OH),etc.) than Ho.
3. The presence of Ho compound in a thin layer on target surface could cause a fast sputter speed with respect of extraction. The most part of Ho could remain on the chamber walls and not enter the extraction section.

Sintered target: preliminary studies



First tests performed with a sintered target of Ho Ti, Ni, Sn on a copper support tested.
Analysis is ongoing

We observed a clear peak at 165 amu.
The current was low O(40nA) but more stable during the different measurements but we melted the target after a ~2 hours.
We observed an unexpected peak at 164 a.m.u.

Preliminary interpretations:

1. The chemical and mechanical properties of sinter could have decreased the heat flux from target to cooled support.
2. The high temperature could be dissociate the intermetallic compounds in the target and create some compound at 164.

Studies on other materials to reach high temperature and a deep analysis to understand 164 peak are ongoing.

Summary

- The ion source has been commissioned and showed a good performance. A more detailed analysis of the data already acquired from various targets is ongoing and the calibration procedure is being established. Effect due to misalignment and beam spot are enough understood. New studies are ongoing.
- Results of the first tests with different Ho targets show clear peak to 165 amu (natural holmium) but the current value and stability are not yet satisfactory.

NEXT STEPS

- Better understanding of implanter setup and source tuning. This could help to reduce power dissipation on sputter target and decrease its working temperature (reduce or eliminate possible chemical decomposition)
- Search for the optimal way to prepare and operate Ho targets with different compounds (molecular plating) or different materials (sintered target).
- We are planning to implant a detector array with low dose of ^{163}Ho (~1 Bq/det) in the next few months.