

ISOL and laser applications at the SPES facility

Topics:

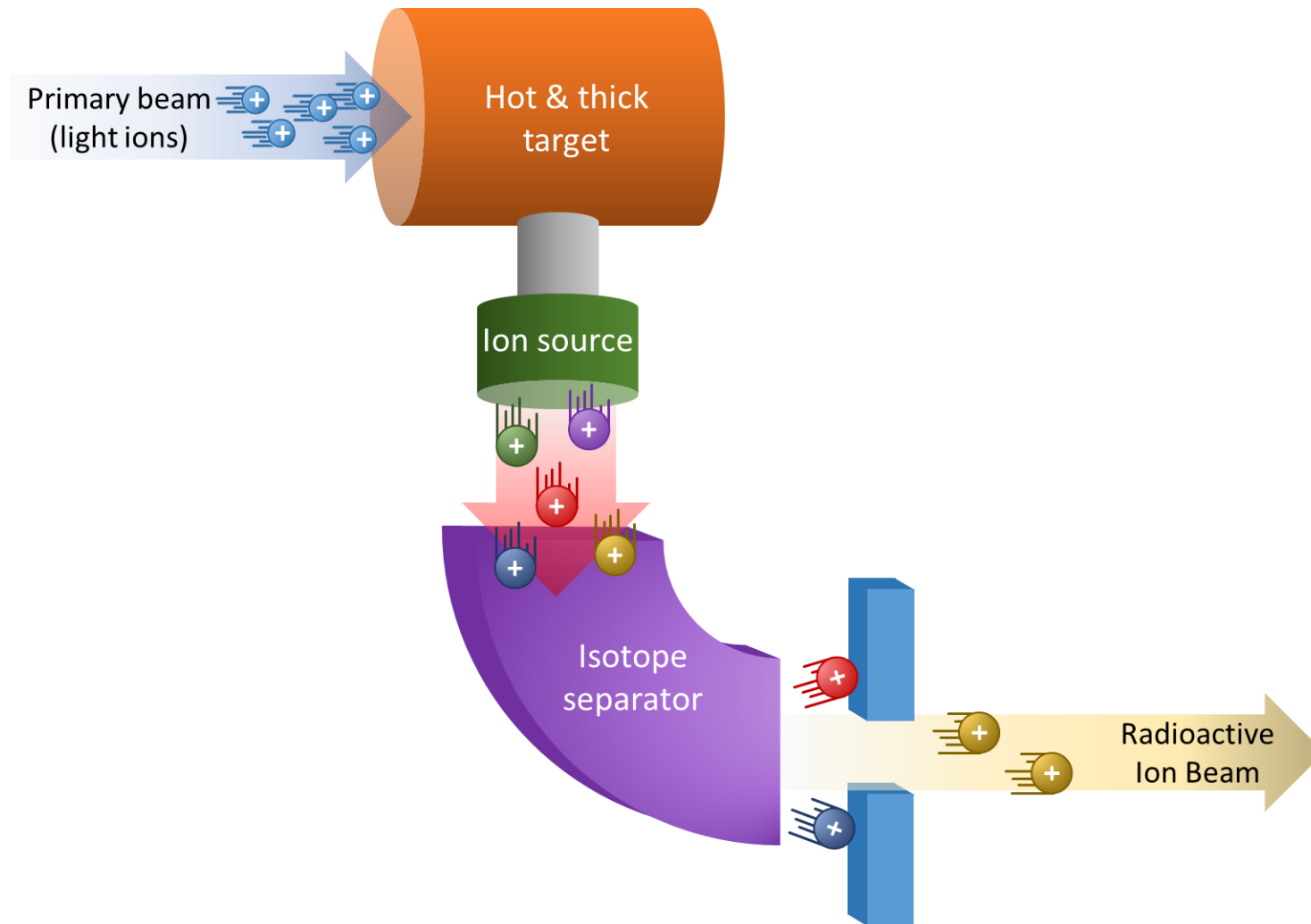
- Laser spectroscopy and applications
- Nuclide production with ISOL for medicine and nuclear physics
- Decay spectroscopy of nuclide of medical interest

Michele Ballan

INFN-LNL, Legnaro, Italy

On behalf of the SPES WP6 & ISOLPHARM
collaboration

The ISOL technique for the production of Radioactive Ion Beams (RIBs)



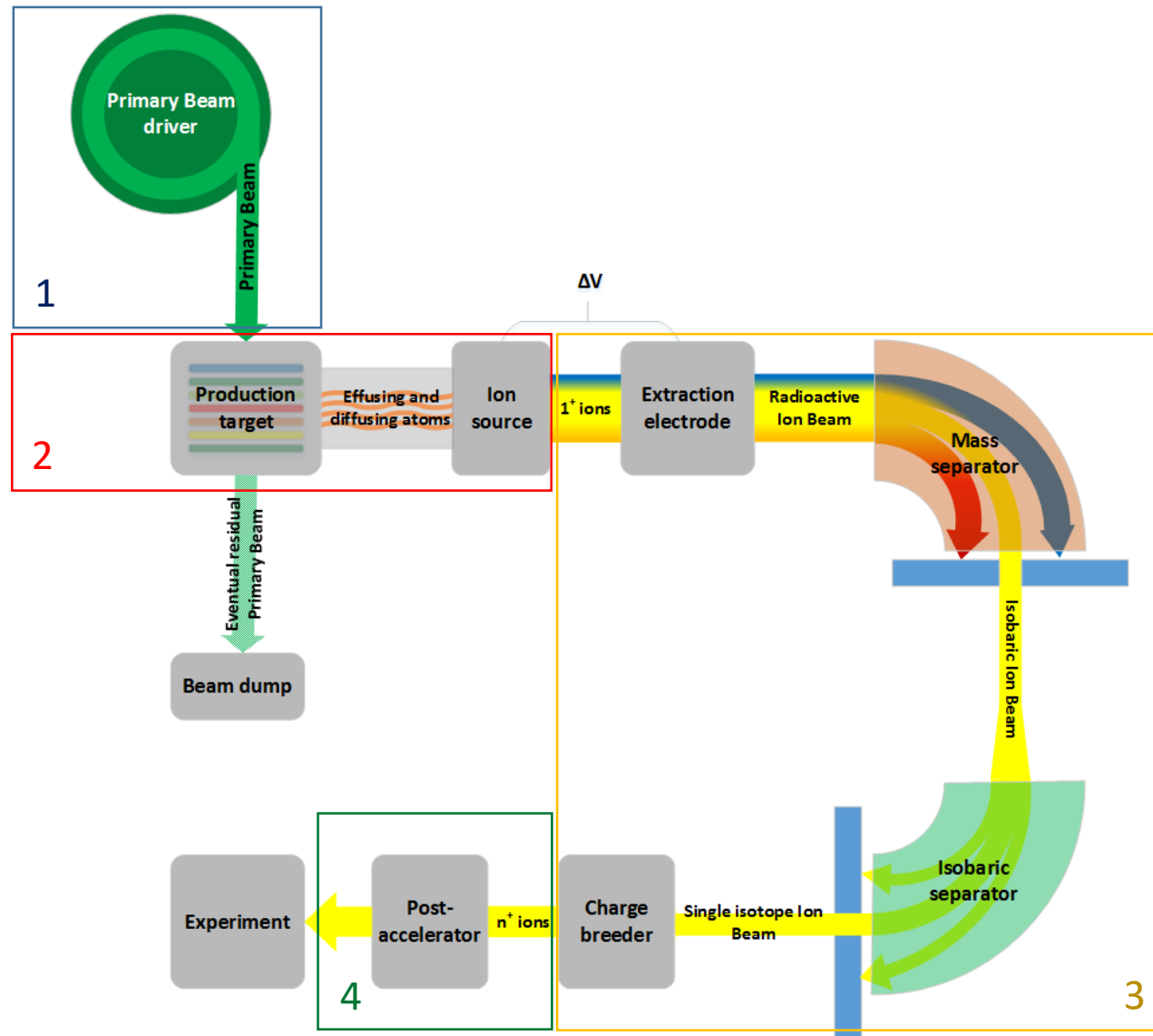
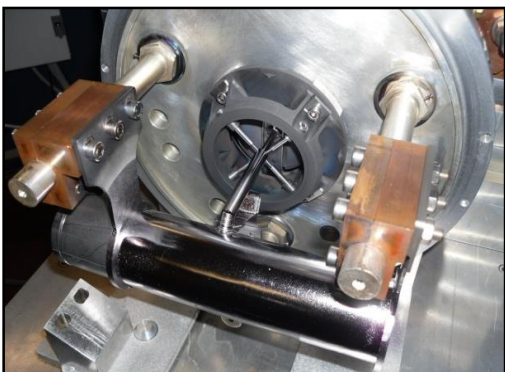
The SPES facility

1 - Driver

70 MeV commercial cyclotron



2 - Target-Ion Source unit



3- RIB manipulation

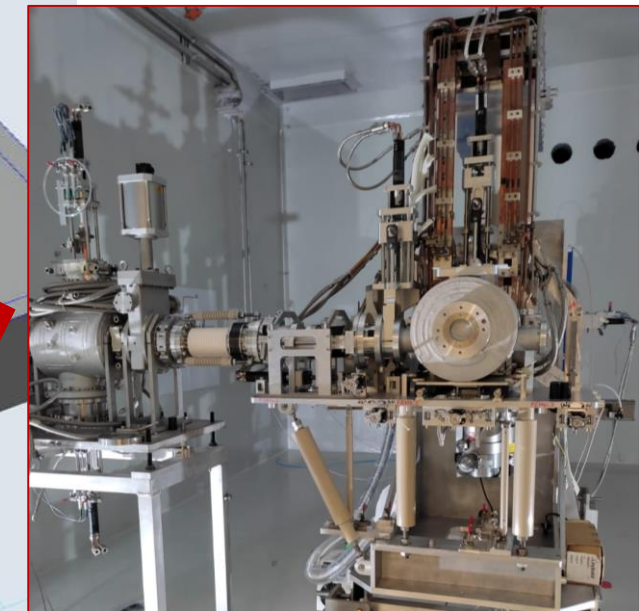
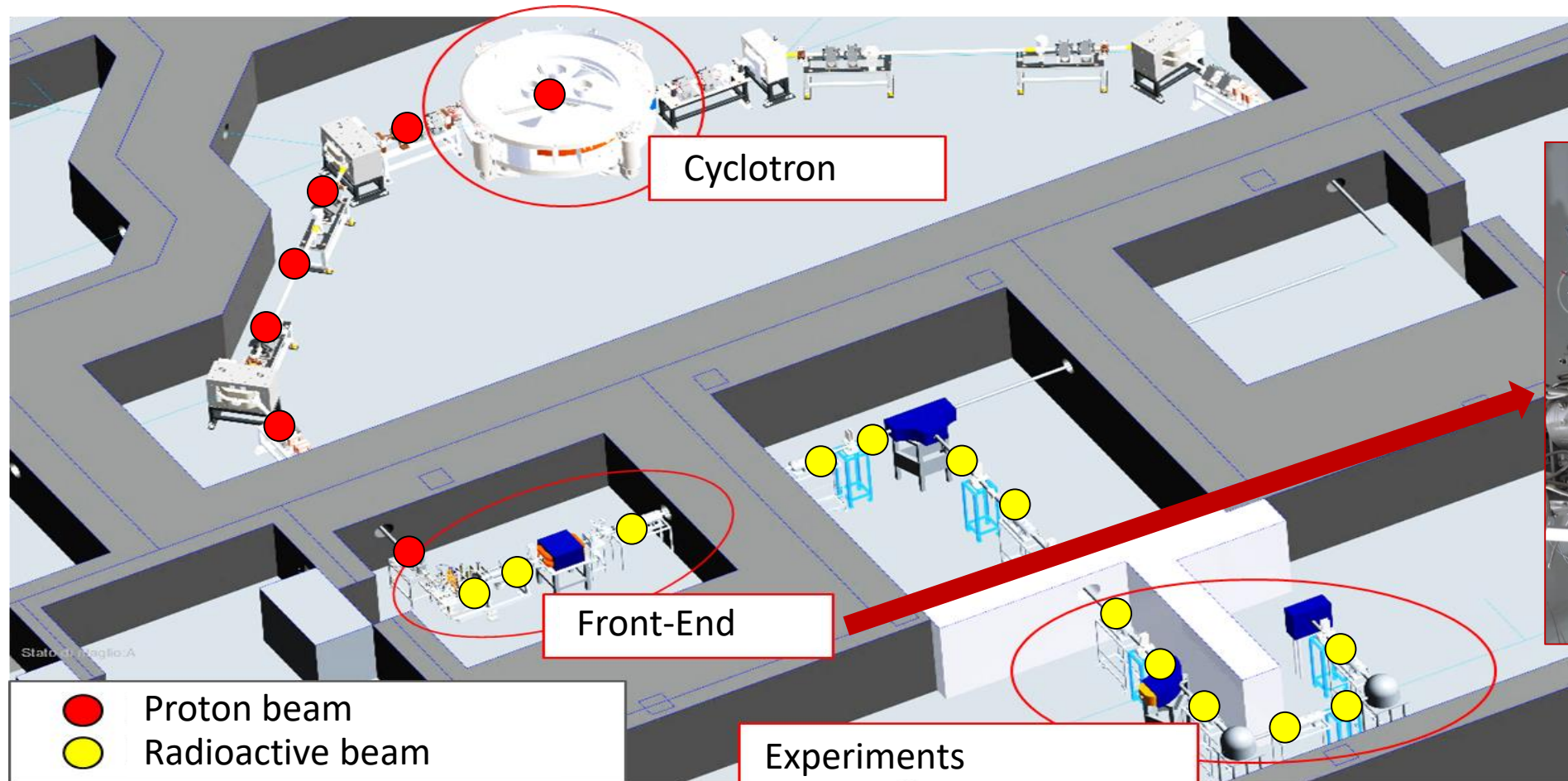
- Mass Separator (WF)
- Beam Cooler
- HRMS
- ECR Charge Breeder
- RFQ

4 - Post Accelerator: ALPI existing complex

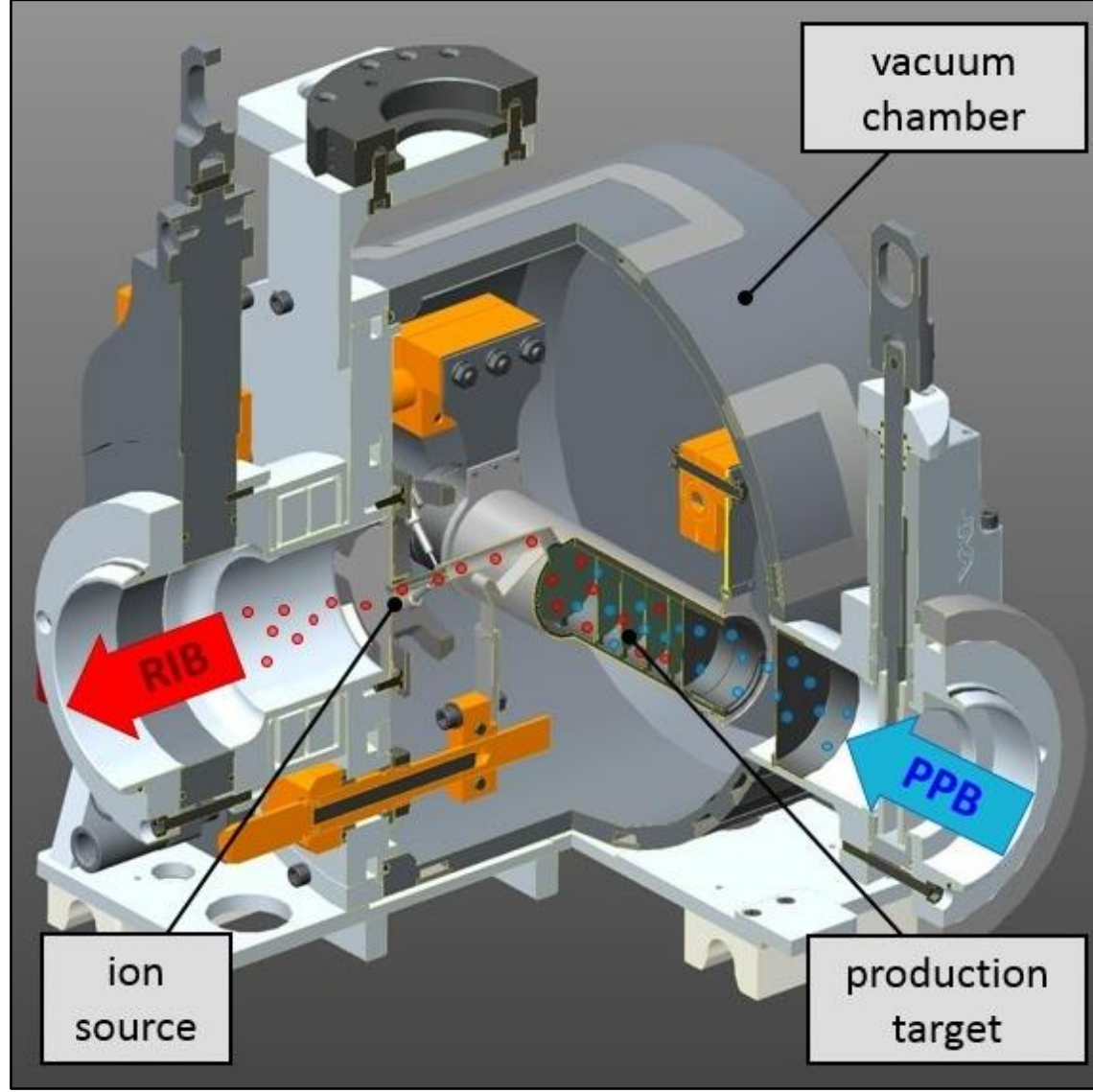


The SPES facility

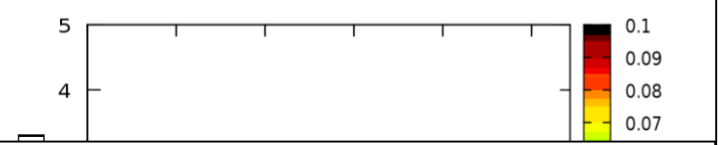
The ISOL machine is currently being installed



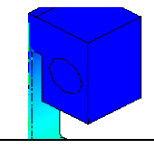
The SPES UC_x target



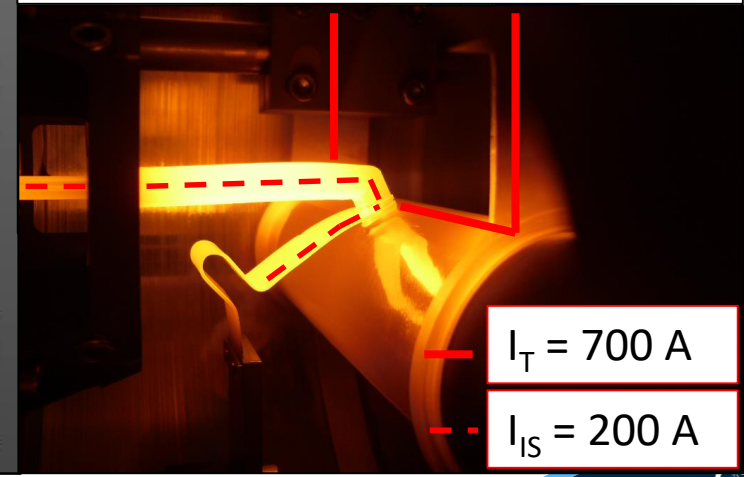
Designed for stopping a 40 MeV 200μA PPB (FLUKA)



Designed for working temperatures above 2000°C (ANSYS®)



Tested and commissioned for high temperature operation



$I_T = 700 \text{ A}$
 $I_{IS} = 200 \text{ A}$



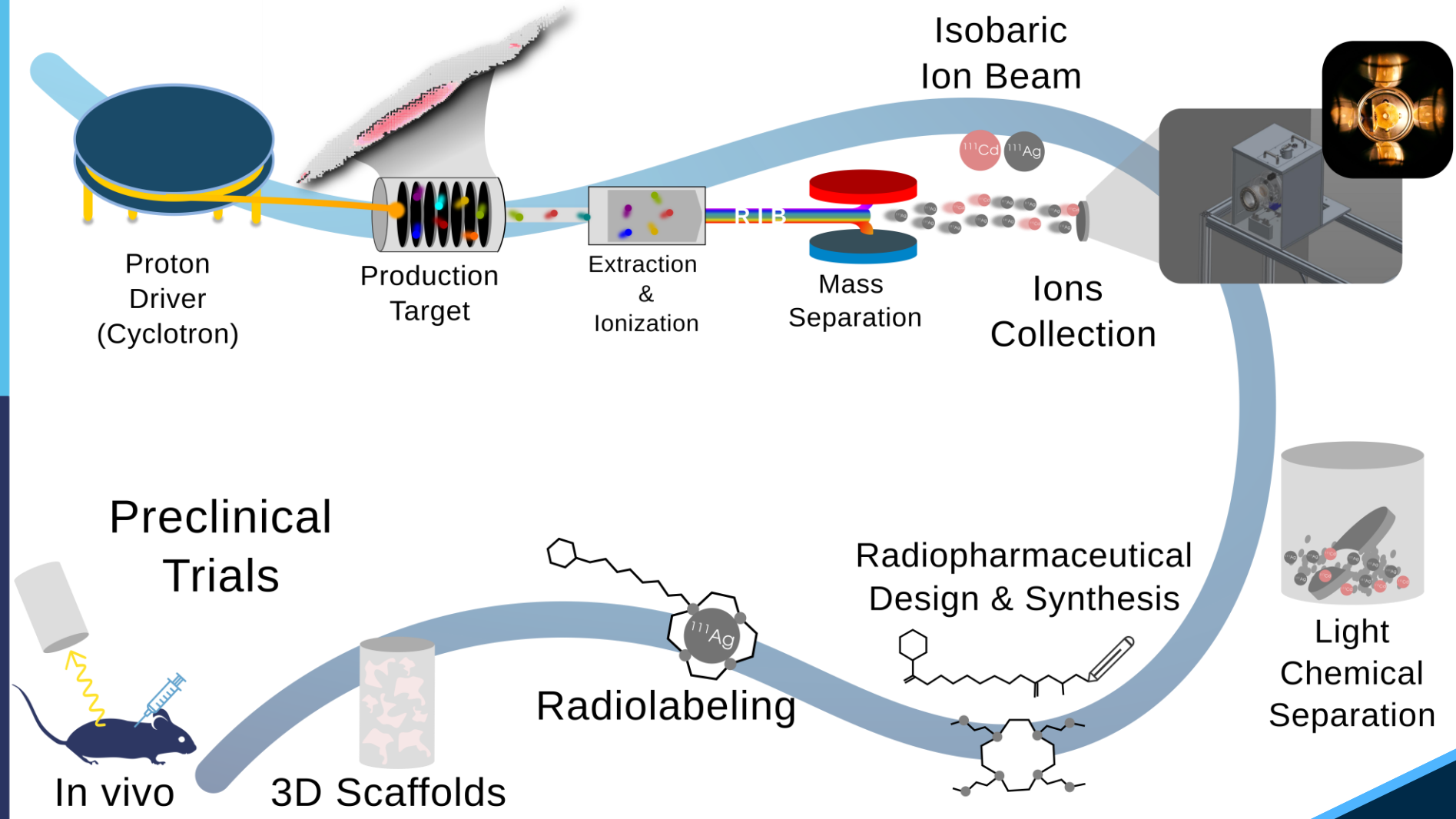
The ISOLPHARM method

INFN-LNL

ISOLPHARM

Collaboration

with universities



The ISOLPHARM method: highlights

1. Method

Innovative production @SPES
of several (medical)
Carrier Free + High Purity
radionuclides
(^{111}Ag , ^{89}Sr , ^{136}Cs ..)



2. Collaboration

Broad, focused on **radio-pharmaceutical development**:
synthesis, characterization
and preclinical tests
(*vitro/vivo*)



4. Experiment

Financed by CSN5 (physics
application): ISOLPHARM_Ag +
ISOLPHARM_EIRA.



3. Set-up

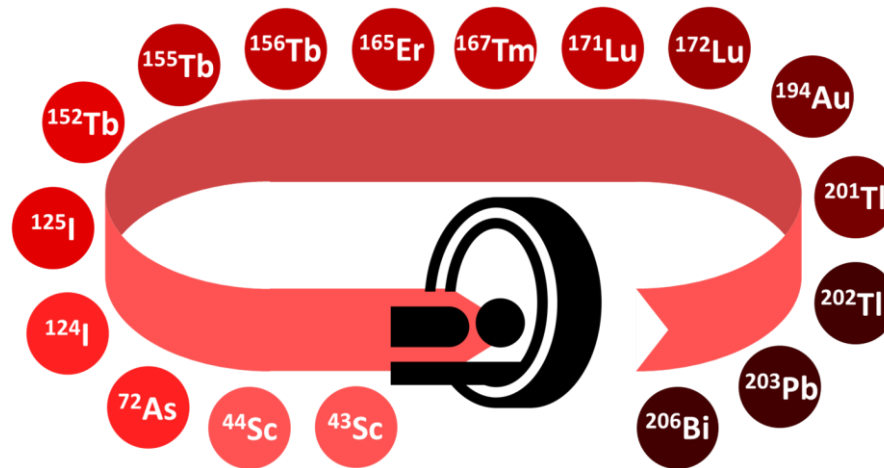
An **irradiation station**,
using the SPES
Low Energy Beams, is
planned in the SPES building



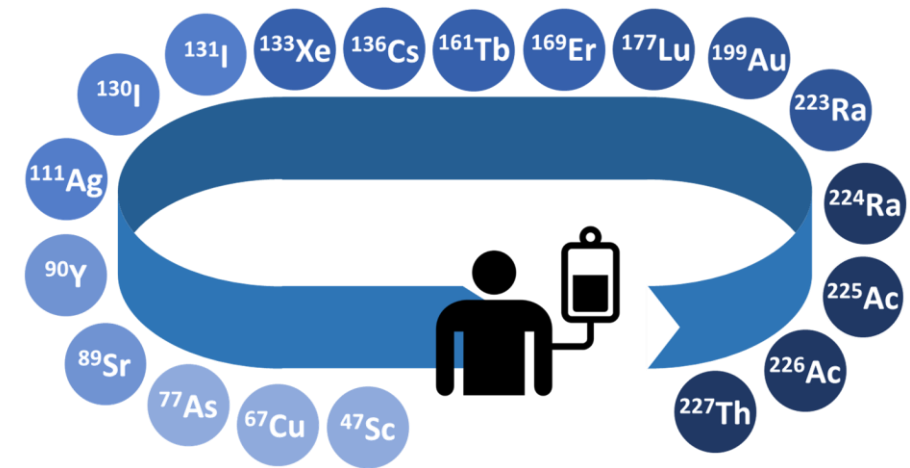
ISOLPHARM
SPES exotic beams for medicine

Possible ISOL nuclides of medical interest

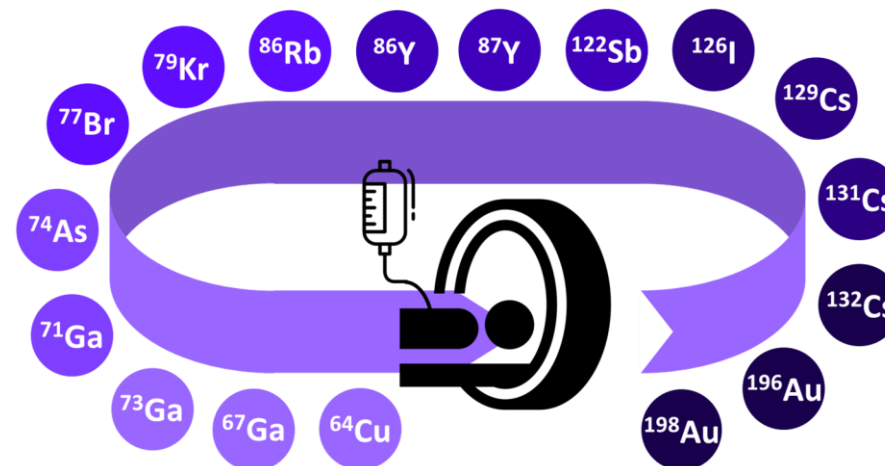
Diagnostic isotopes



Therapeutic isotopes

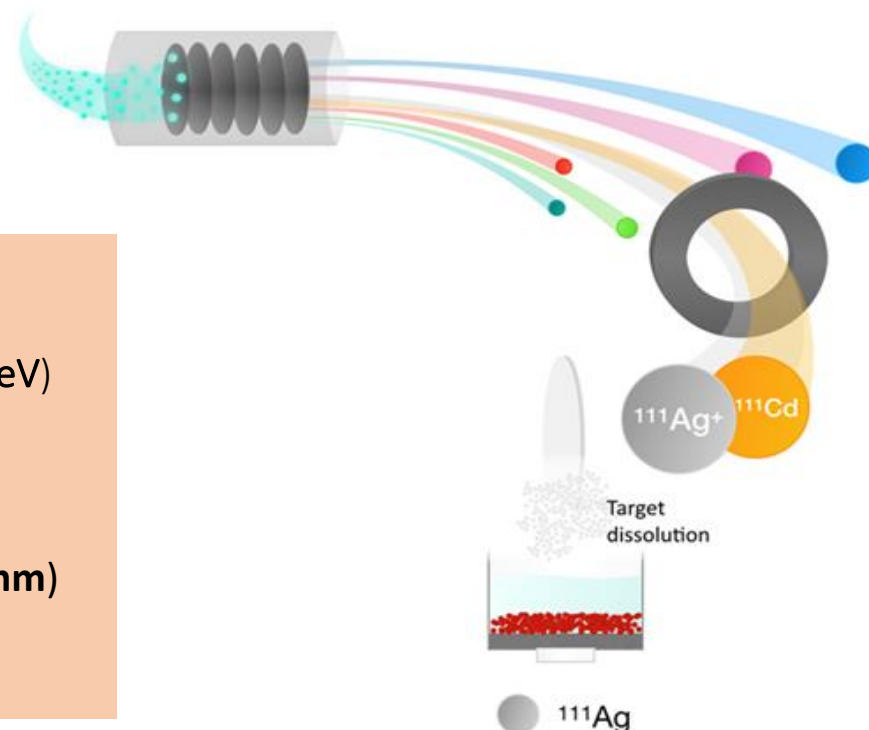


Theragnostic isotopes



Among the wide set of **ISOL producible nuclides**, a large number shows relevant properties for medicine, in terms of **half-life**, **decay radiation** and **chemical behavior**

Possible ISOL nuclides of medical interest: the case of ^{111}Ag



^{111}Ag properties

- β^- emitter (average energy 360 keV)
- Good half-life (7.45 days)
- Average tissue penetration (**1.8 mm**)
- Medium energy γ rays -> SPECT

In the market **No radiopharmaceuticals Silver-based!**

^{111}Ag can be produced @ SPES **with high purity & also with high production rate.**

No Isobaric radioactive contamination in the secondary target (also with LASER off)!



111 Isobaric chain

111 Isobaric chain	$t_{1/2}$	Decay	Target Yield
$^{111}\text{Cadmium}$	Stable		Low yield production
$^{111}\text{Silver}$	7.45 days	β^-	Good yield production
$^{111}\text{Palladium}$	23.4 min	β^-	Bad release, short $T_{1/2}$
$^{111}\text{Rhodium}$	11 sec.	β^-	No release, very short $T_{1/2}$

ISOLPHARM: the collaboration

ISOLPHARM started as a scientific gathering of local competences, now coordinated by INFN-LNL



Trento Institute for Fundamental Physics and Applications



UNIVERSITÀ DI TRENTO

BIOTech BIOtecj
Biomedical Technologies



Centre for Integrative Biology



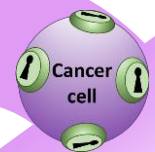
UNIVERSITÀ DI SIENA 1240



UNIVERSITÀ DEGLI STUDI DI BRESCIA



Development of cell-lines, *in-vitro* and *in-vivo* studies, imaging



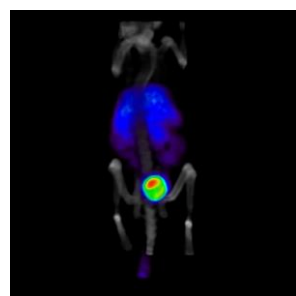
Unconventional radionuclide (e.g. ¹¹¹Ag) production with an innovative method



Development and synthesis of small molecules for cancer treatment



Study and development of tumor targeting agents for specific targets (e.g. CCK2R)



PET imaging of a mouse treated with a ⁶⁴Cu-labelled radiopharmaceutical developed by the ISOLPHARM collaboration

Development of chemical purification procedures and innovative chelators



Laser spectroscopy and applications

Contributors:

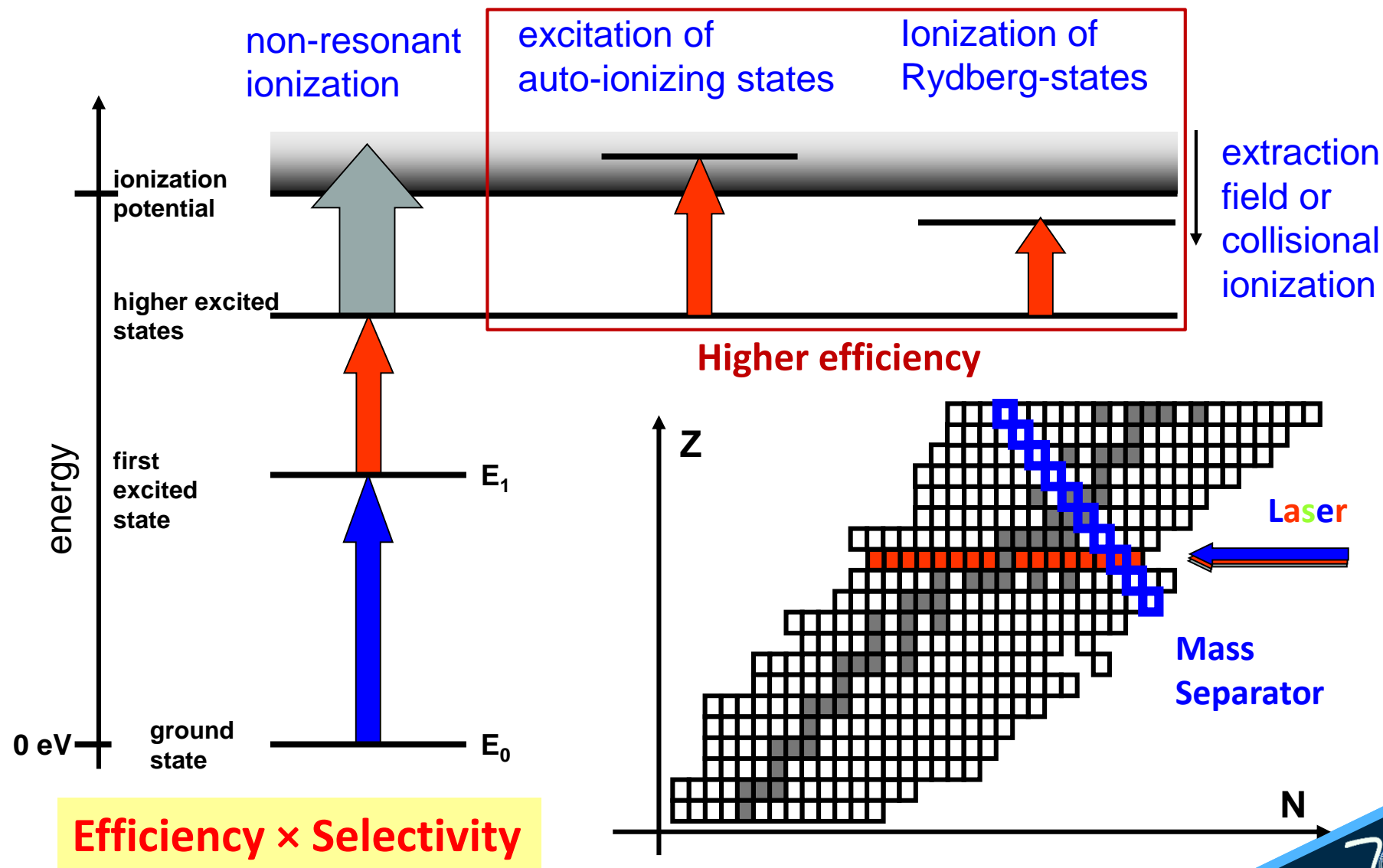
Omorjit Singh Khwairakpam, *UNISI & INFN-LNL*

Daniele Scarpa, *INFN-LNL*

Emilio Mariotti, *UNISI*

Alberto Arzenton, *UNISI & INFN-LNL*

Principles of resonant laser ionization



Challenges to high efficiency laser ionization

- More than 50 % of the elements for which photoionization has been realized **rely on a non-resonant ionization step**.
- For these elements, the ionization step is **not saturated**, and the efficiency is proportional to the **available pump laser power** for this step.
- The efficiency can be significantly **improved if** the final step populates **an Auto-Ionizing State (AISs)**.
- The main limitations in looking for AISs are
 - 1) time-consuming process
 - 2) For many elements, a suitable AIS does not exist.

Such activity benefits of the active collaboration with the community of Laser facilities



The Laser laboratories at SPES

Offline: spectroscopy studies

3 Dye Laser @ 10 Hz rep. rate

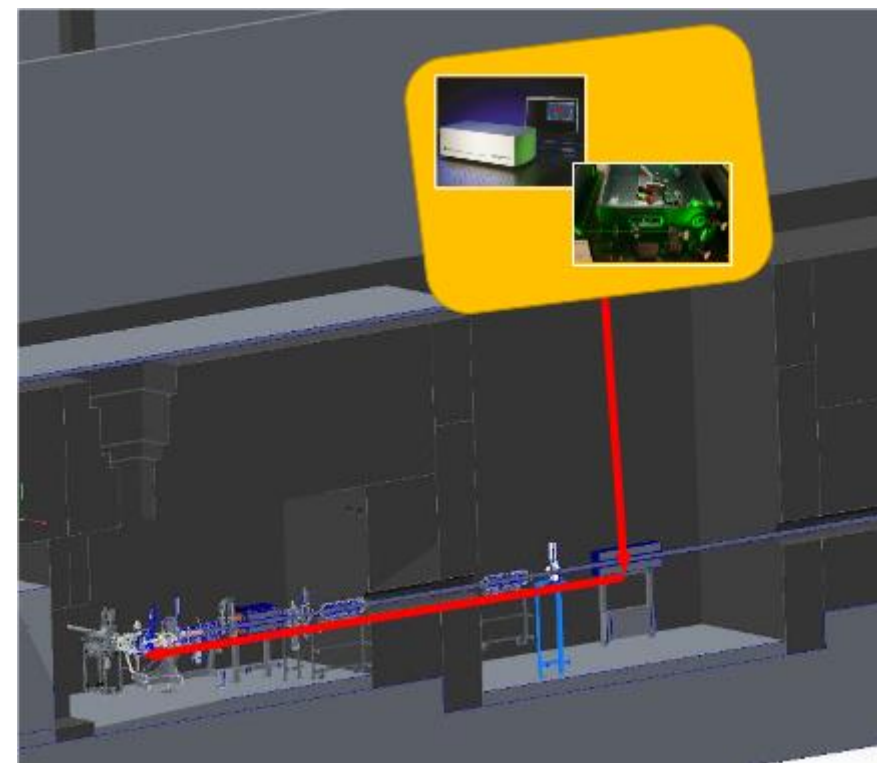


Diagnostic tools:

- Monochromator
- HCL
- ToF Mass Spectrometer

Online (SS laser): RIB production

3 Dye Laser @ 10 Hz rep. rate



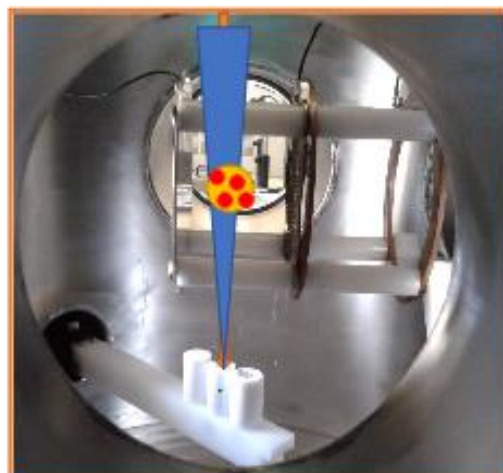
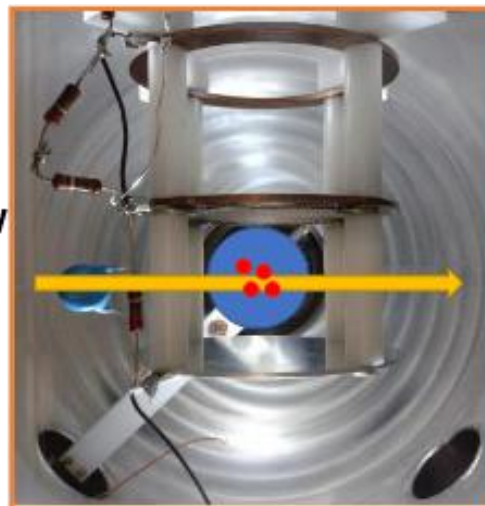
Diagnostic tools:

- Λ -meter
- Alignment system
- Ion-beam

Home-made Time-of-Flight (TOF)

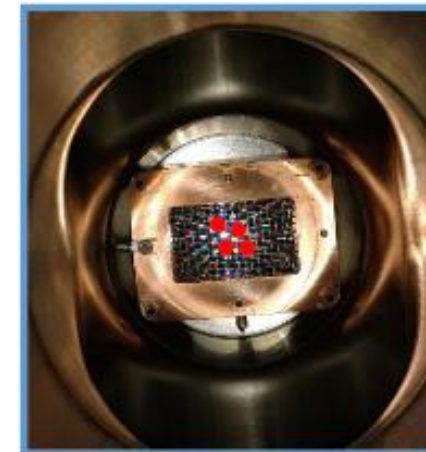
Measure Sequence:

Top view



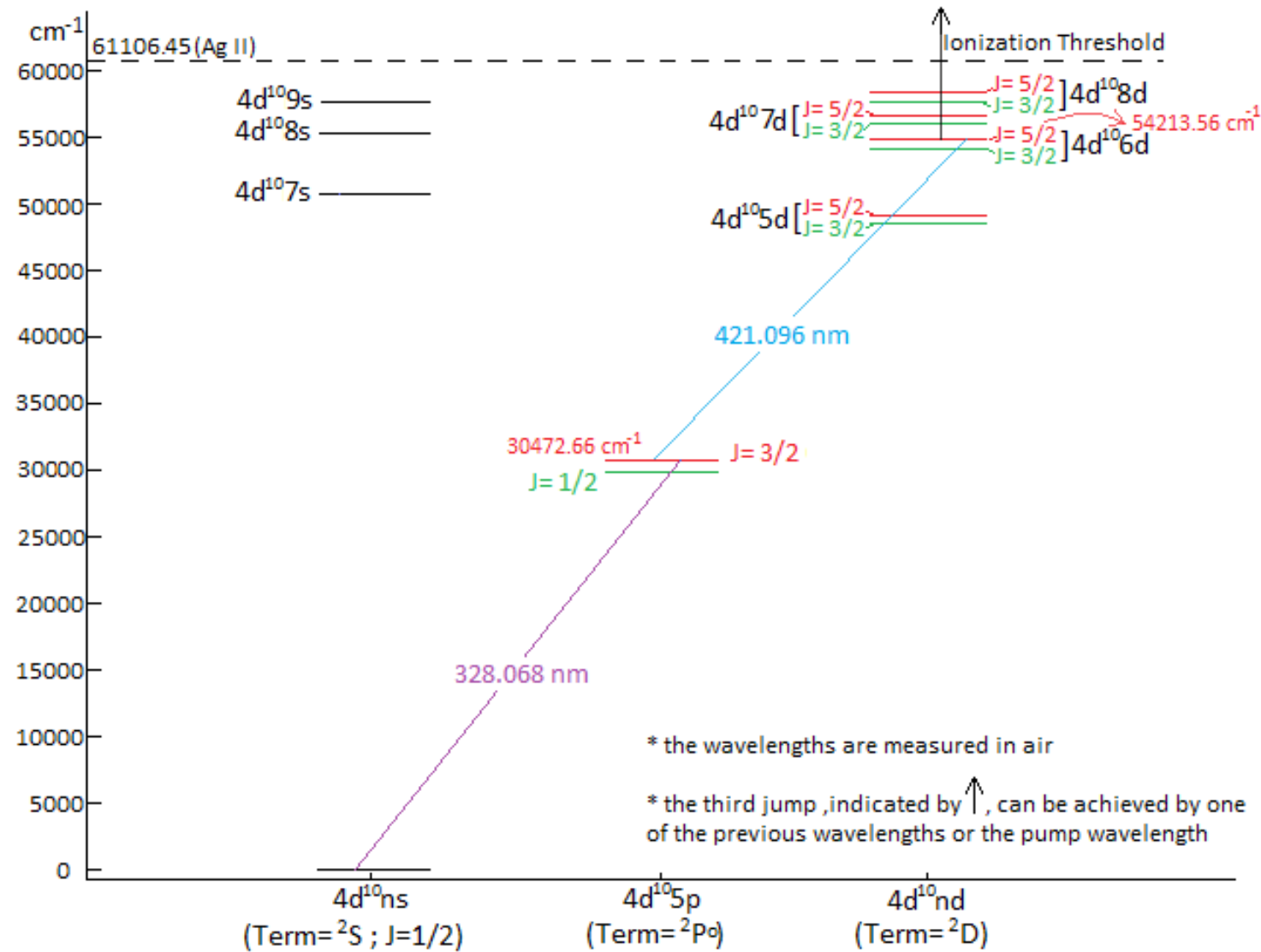
Front view

(as seen by the ionization laser)



- 1) Ablation
- 2) Plume Expansion
- 3) Photoionization
- 4) Flight
- 5) Collection

Example: ionization scheme of Ag



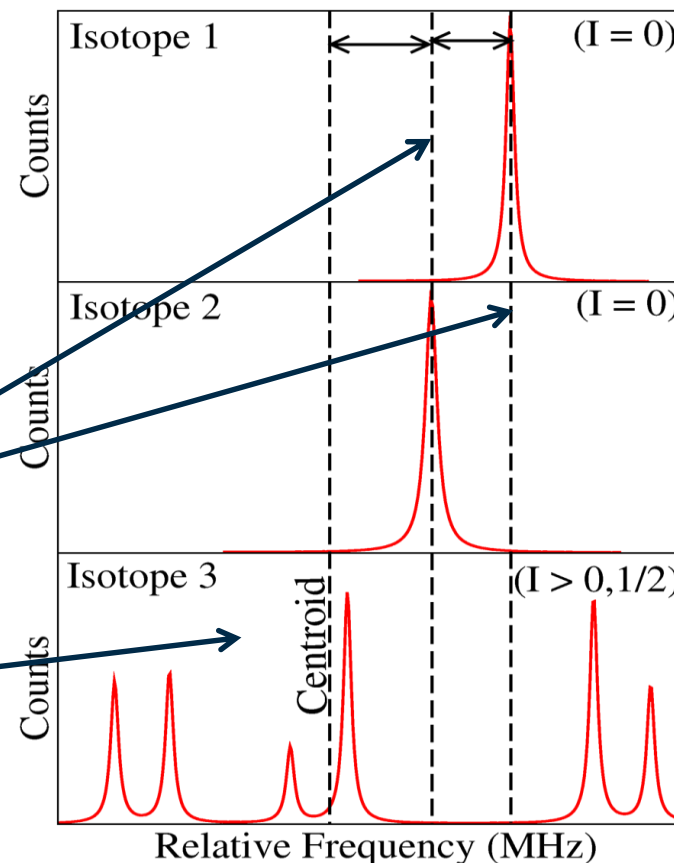
A nuclear fingerprint in the atomic spectrum

The nucleus is not a point charge – the atomic energy levels are perturbed by the electric and magnetic fields at the nucleus (*part per million* effects)

Investigation of

- Isotope shifts
- Isomer shifts
- Hyperfine structure

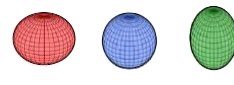
provides *model-independent* data...



$$\delta \langle r^2 \rangle$$

$$\mu$$

$$Q_S$$



$$I$$

Such activity could require an upgrade to the current laser set-ups

Nuclide production with ISOL for medicine and nuclear physics

Contributors:

Alberto Andrichetto, *INFN-LNL*

Mattia Manzolaro, *INFN-LNL*

Stefano Corradetti, *INFN-LNL*

Alberto Monetti, *INFN-LNL*

Giordano Lilli, *INFN-LNL*

Lisa Centofante, *INFN-LNL*

Aldo Zenoni, *UNIBS & INFN-PV*

Antonietta Donzella, *UNIBS & INFN-PV*

João Pedro Ramos, *SCK CEN*

Lucia-Ana Popescu, *SCK CEN*

Feasibility of nuclide production with ISOL for medicine and nuclear physics

Diffusion/Effusion efficiencies (Release efficiency):

Depends on the target (working temperature, porosity/density, chemical affinity with the produced nuclide)

Producible Yield with ISOL:

To be estimated in order to evaluate the feasibility of a specific experiment with ISOL RIBs

Beam transport efficiency:

Depends on the beam optics devices

$$Y = \sigma \Phi N \varepsilon_d \varepsilon_e \varepsilon_i \varepsilon_t$$

Ionization efficiency:

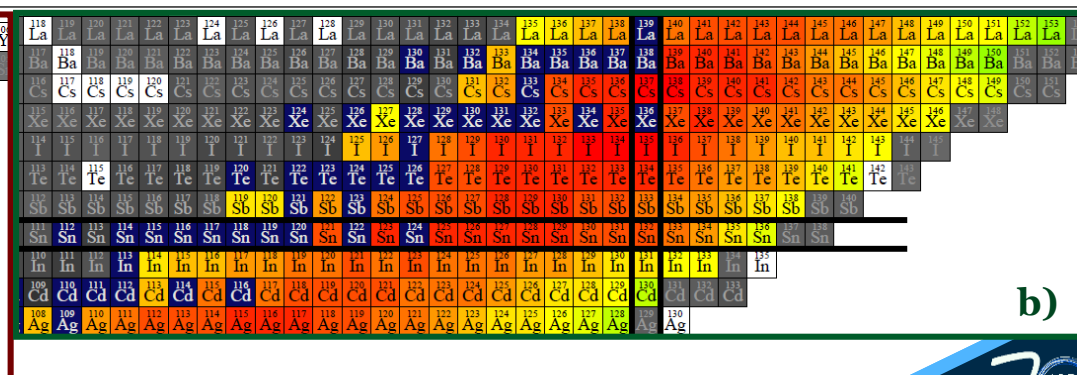
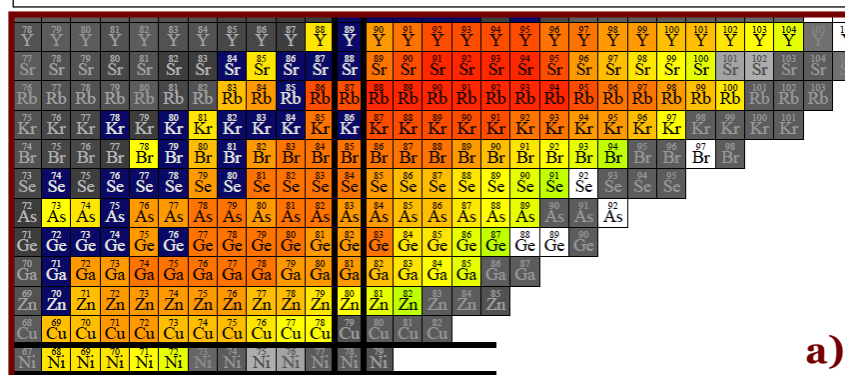
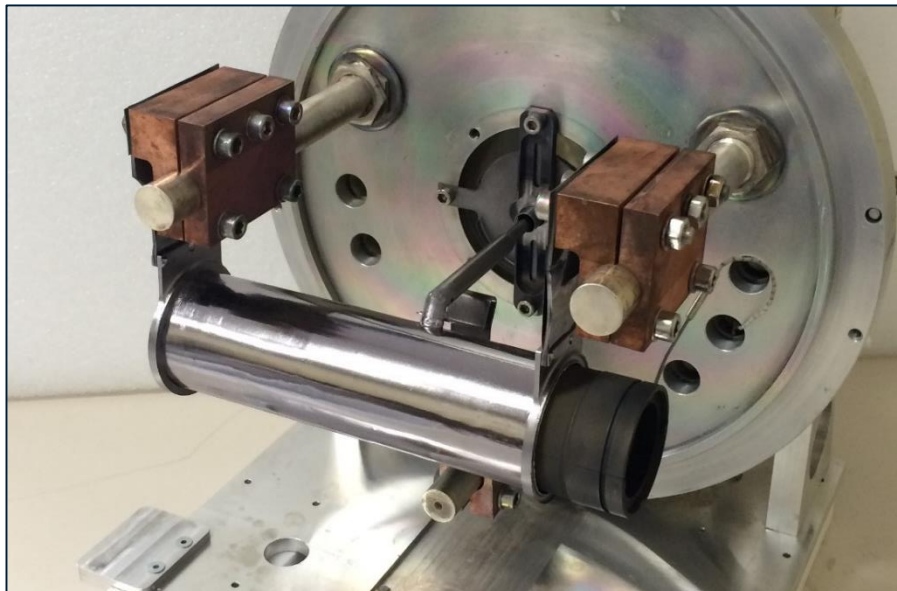
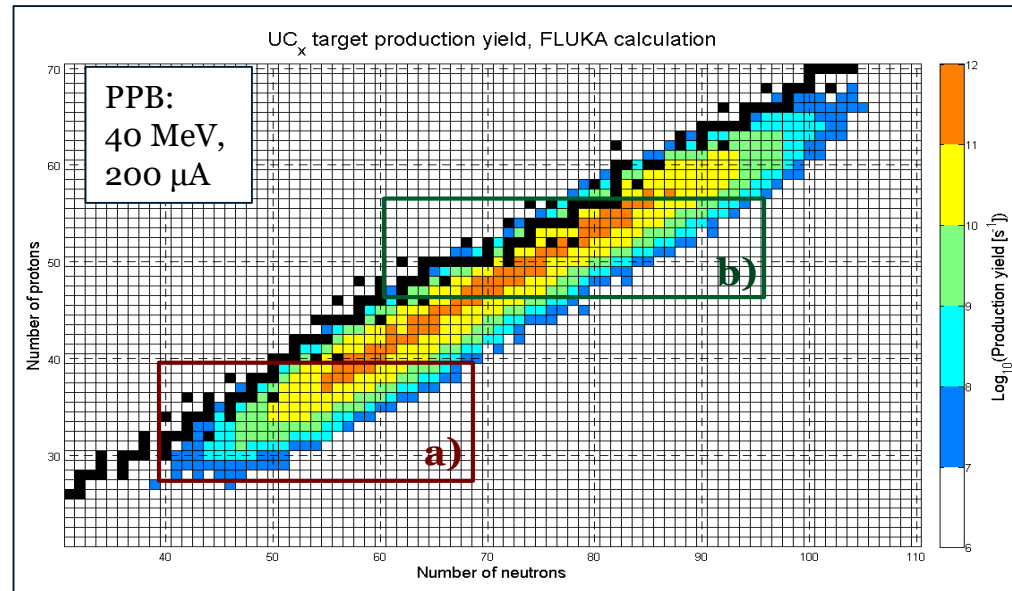
Depends on the ion source technology

In-target yield:

Depends on the beam (particle type, energy) and on the target material

Target-dependent factors

The SPES UC_x target



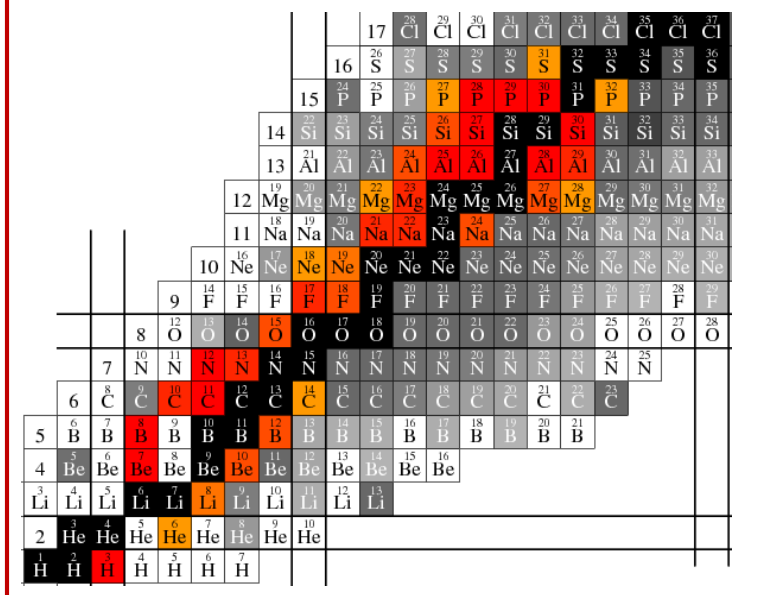
FLUKA calculations experimentally validated at ORNL



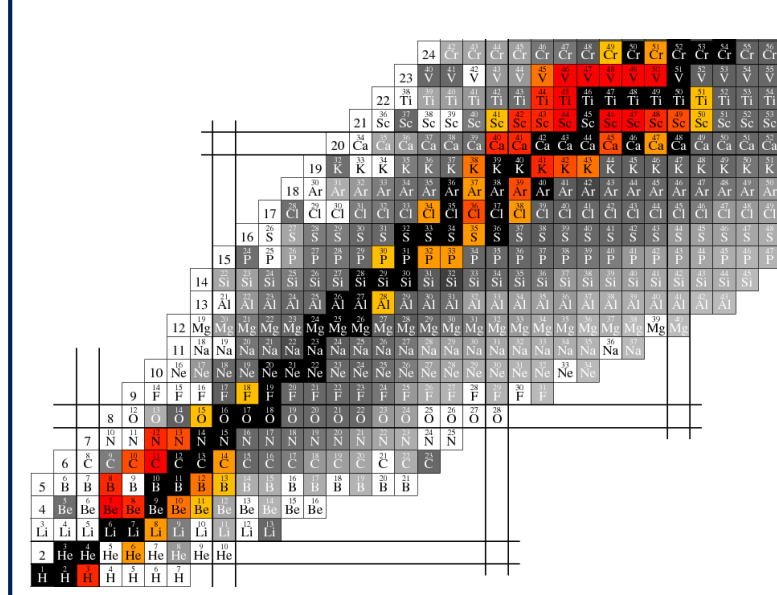
Other possible target materials

(some examples)

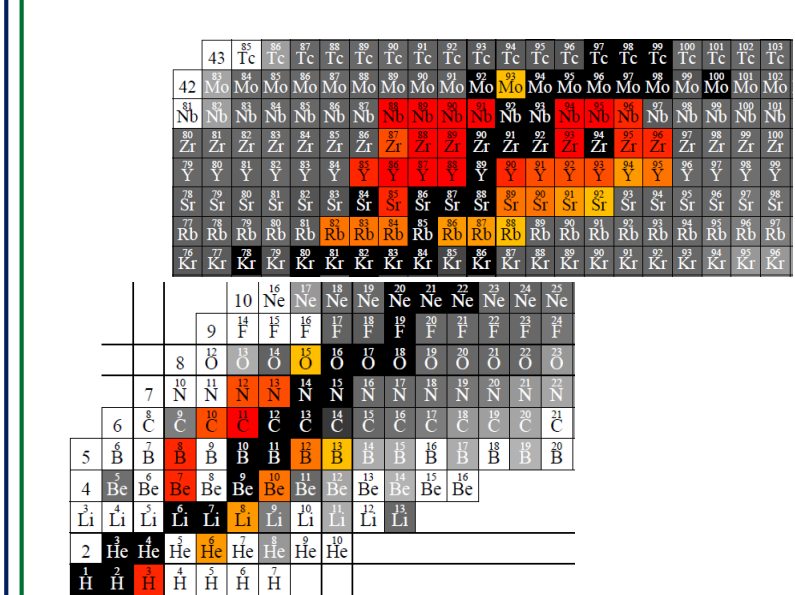
Silicon Carbide (SiC):



Titanium Carbide (TiC):



Zirconium Carbide (ZrC):



In-target FLUKA estimated yields	■	> 10 ¹¹	■	10 ⁹ - 10 ¹⁰	■	10 ⁷ - 10 ⁸	■	10 ⁵ - 10 ⁶	■	10 ³ - 10 ⁴	■	10 - 10 ²	[nuclide/s]
	■	10 ¹⁰ - 10 ¹¹	■	10 ⁸ - 10 ⁹	■	10 ⁶ - 10 ⁷	■	10 ⁴ - 10 ⁵	■	10 ² - 10 ³	■	< 10	

Further examples: B₄C, LaC_x, ZrO₂, HfO₂, CeO₂, ZrGe, CeS

Further information: <https://doi.org/10.1016/j.nimb.2016.01.020>

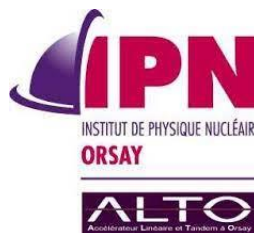
ISOL target material requirements at SPES

In case of SPES, target materials have to meet some specific **mandatory requirements**:

- They have to be **solid** (liquid target are not yet foreseen at SPES)
- They have to be **refractory** (the higher the working temperature the faster the release)
- They have to be **porous** (open porosity enhances the releases of the produced nuclides)
- They have to withstand **extreme conditions**: **high power, thermal stresses, radiation damage**, (direct particle irradiation and very high temperature, in the range of **2000 °C**) with an impact on RIB yields.

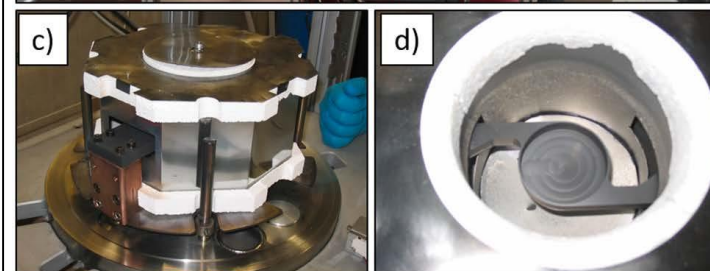
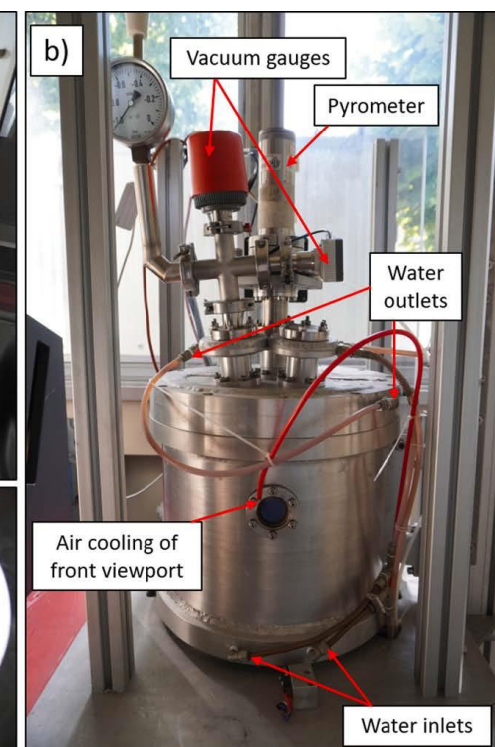
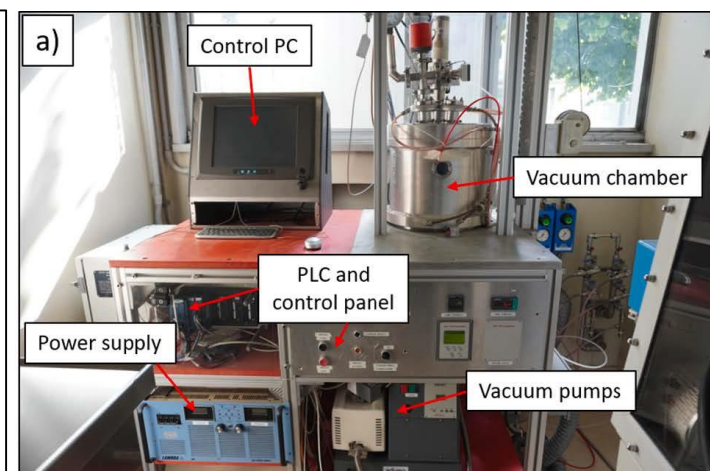
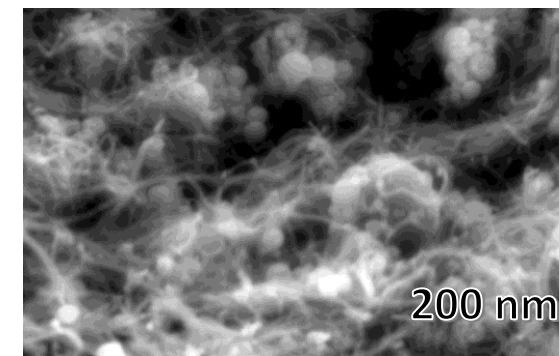
Each new target material requires efforts in its development and characterization before being ready for irradiation

Such activity benefits of the active collaboration with the community of ISOL facilities



Development of target materials

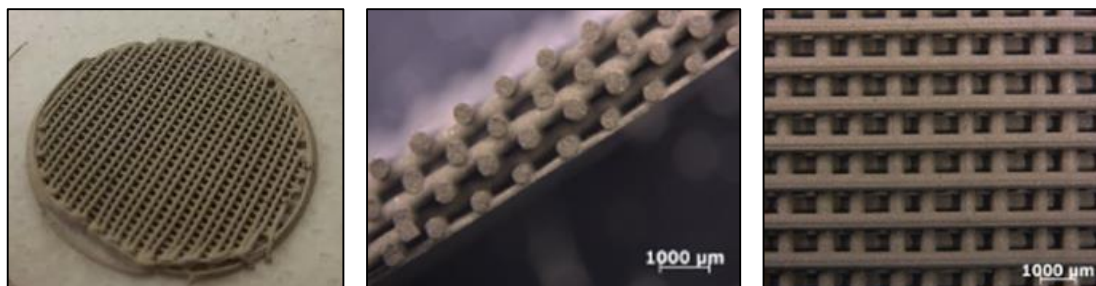
- Typical ceramic targets production methods:
 - Dry method (pressing and sintering of powders)
 - Sol-gel method (powder mixed with a gel solution)
- Micro- or Nano-structured materials with high porosity (usually 30-70%) are the state of the art



Furnaces for target development at INFN-LNL

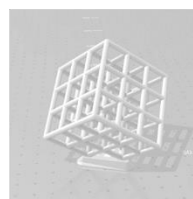
Development of target materials: alternative production techniques

Fabrication of SiC disks by DIW (Direct Ink Writing)

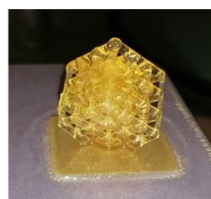


Fabrication of TiC components

PRINTING OF PHOTOCURABLE TITANIUM-SUCROSE-BASED SOL-GEL INK BY DLP



STL file



Printed components



Dried components



Sintered component at 1450°C

→ 3D PRINTED OBJECTS BY A DIGITAL LIGHT PROCESSING (DLP) PRINTER

Additive Manufacturing of ceramic regular structures:

- **Maximization of radiative heat transfer** (increment of the sustainable primary beam intensity)
- **Improved release properties**

AM targets are expected to ensure higher RIB intensities

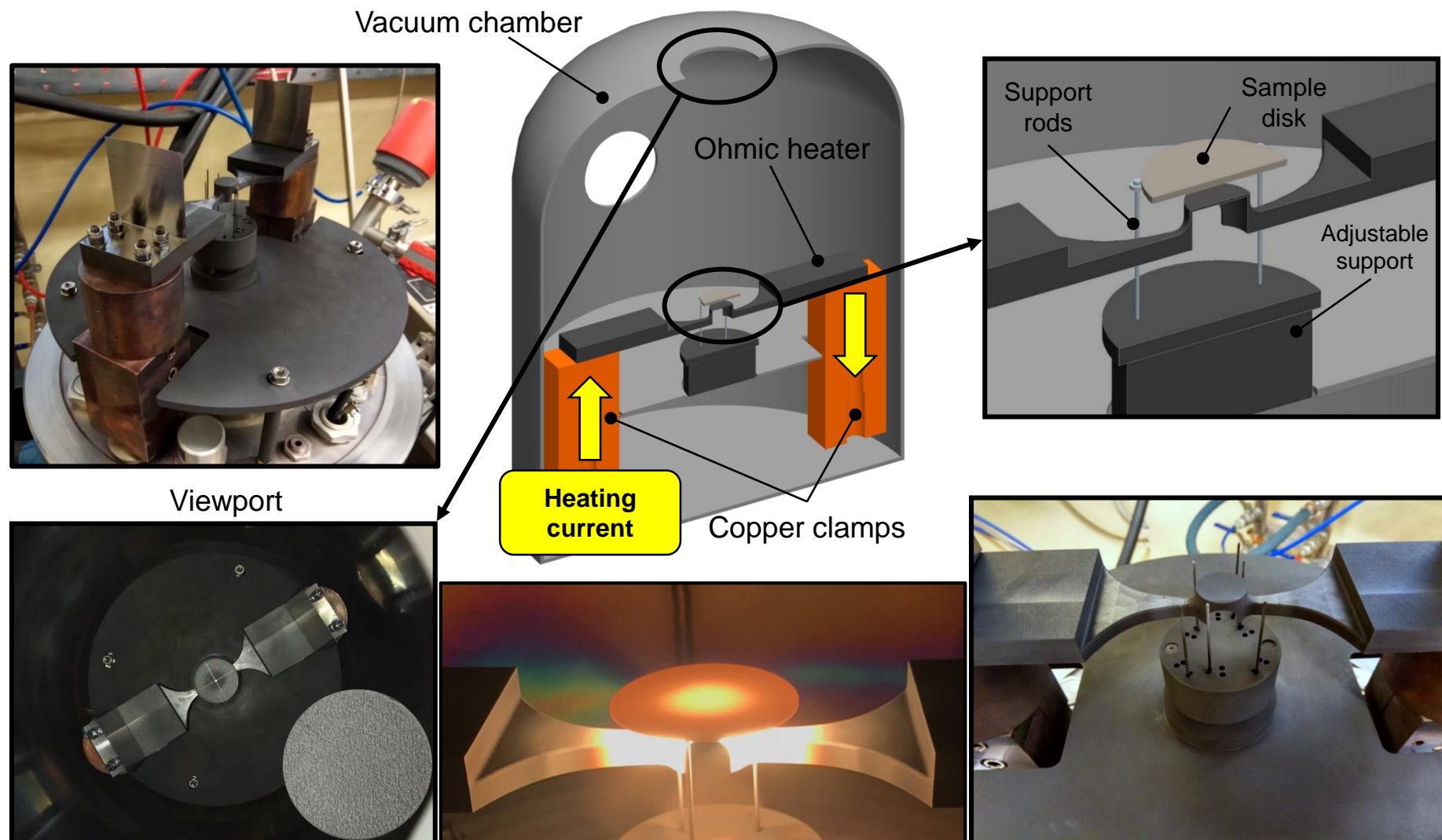
Activity in the framework of the INFN-E Project **AM4INFN**, in collaboration with INFN-PD and UNIPD



UNIVERSITÀ
DEGLI STUDI
DI PADOVA

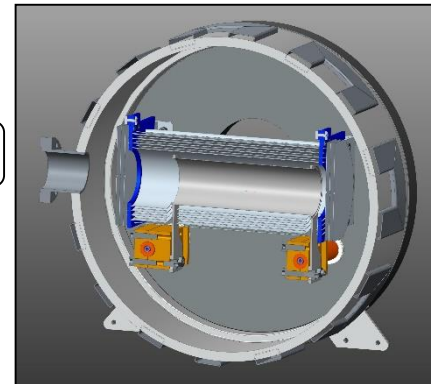
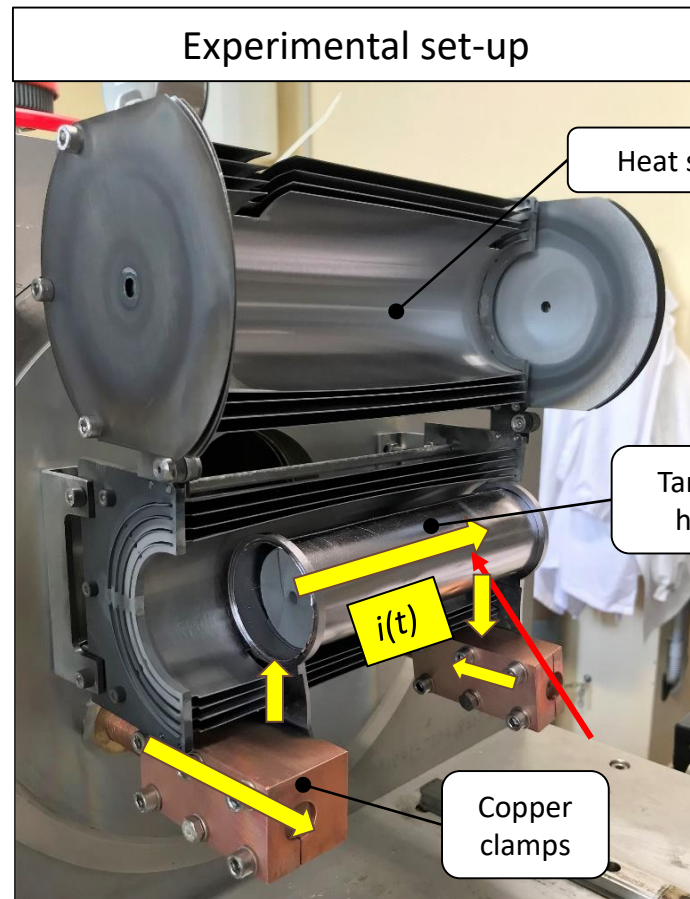


Characterization of target materials: thermal properties (thermal conductivity and emissivity)

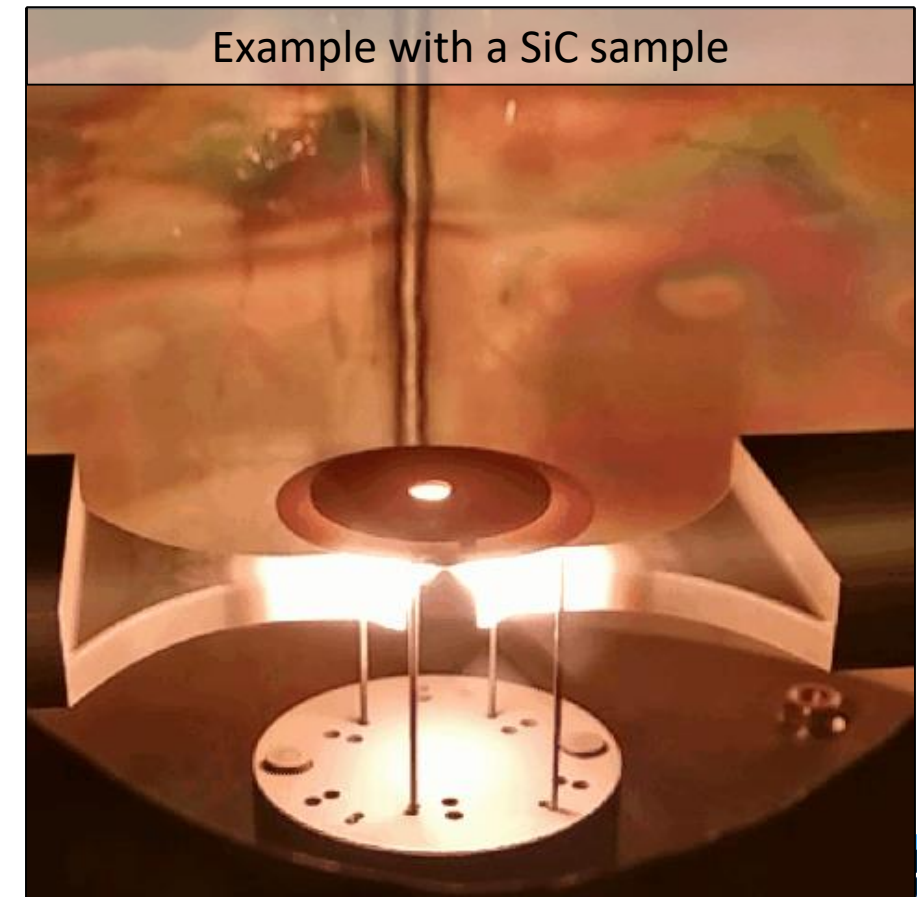


more information at: <https://doi.org/10.3390/ma14102689>

Characterization of target materials: limit temperature and stress resistance



	SiC
T_{lim}	1700 °C



Characterization of target materials: post irradiation analysis

sck cen



Examples of equipment at SCK CEN for nuclear material handling and post irradiation analysis

- Target material research would greatly benefit for a **coordinated approach** and a **facilitated transnational access**.
- **Complimentary** laboratories including characterization techniques are present across all ISOL facilities, e.g.:
 - SCK CEN with very large **service infrastructure** for materials characterization (incl. nuclear materials and post-irradiation examination)
 - INFN-LNL with the expertise of thermal and structural characterization of multi-foil targets
- It also allows the development of **consistent characterization and material production protocols** to allow state of the art operation at all involved facilities.

Decay spectroscopy of nuclides of medical interest

Contributors:

Marcello Lunardon, *UNIPD & INFN-PD*

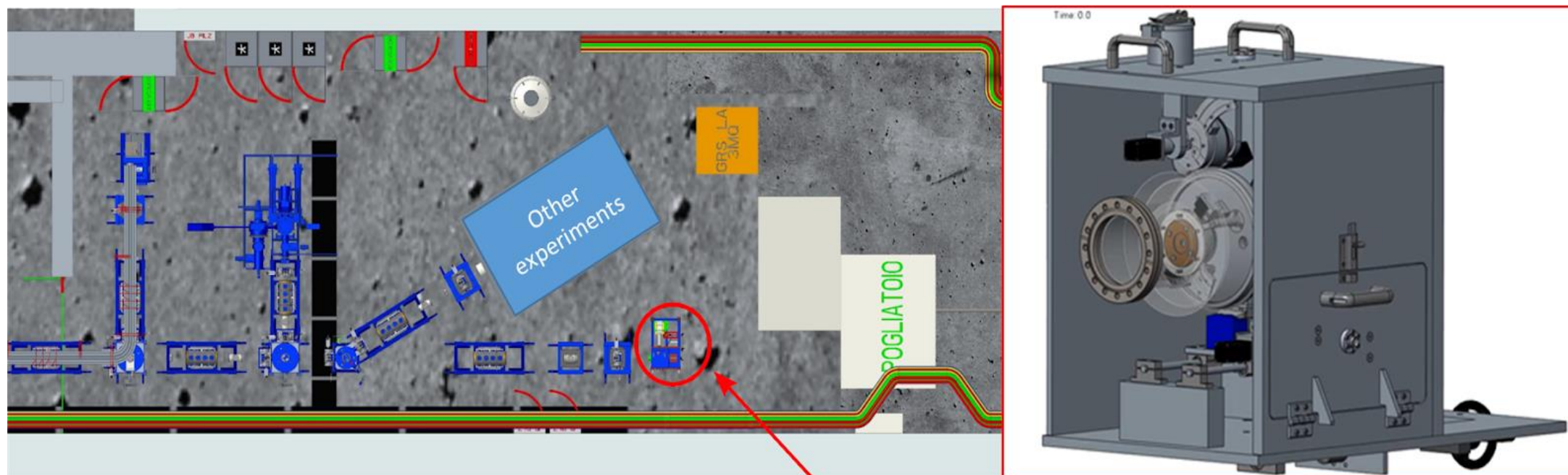
Luca Morselli, *INFN-LNL & UNIFE*

Luca Stevanato, *UNIPD*

IRIS – ISOL(PHARM) Radionuclide Implantation Station

ISOLPHARM required the development of a device able to handle the collection targets and perform spectroscopic analysis for the quality control of the collected radionuclides.

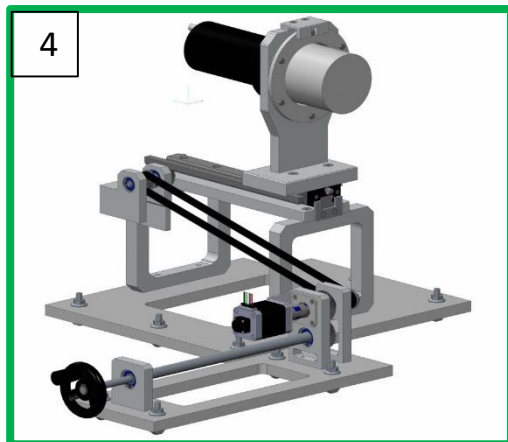
Such device could also be used to produce extremely pure radioactive sources (monoisotopic if laser ionization is used) extremely interesting for nuclear physics applications



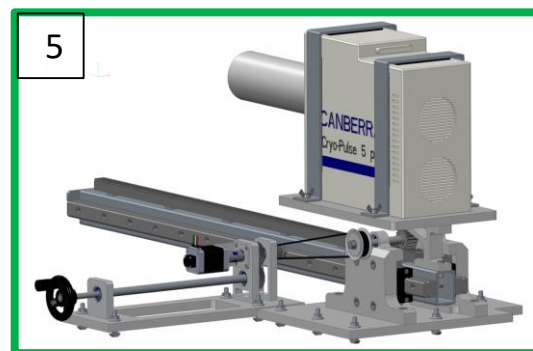
ISOL(PHARM) Radionuclide Implantation Station (IRIS) and its collocation in the SPES low energy experimental hall.

IRIS (ISOLPHARM Radionuclide Implantation Station)

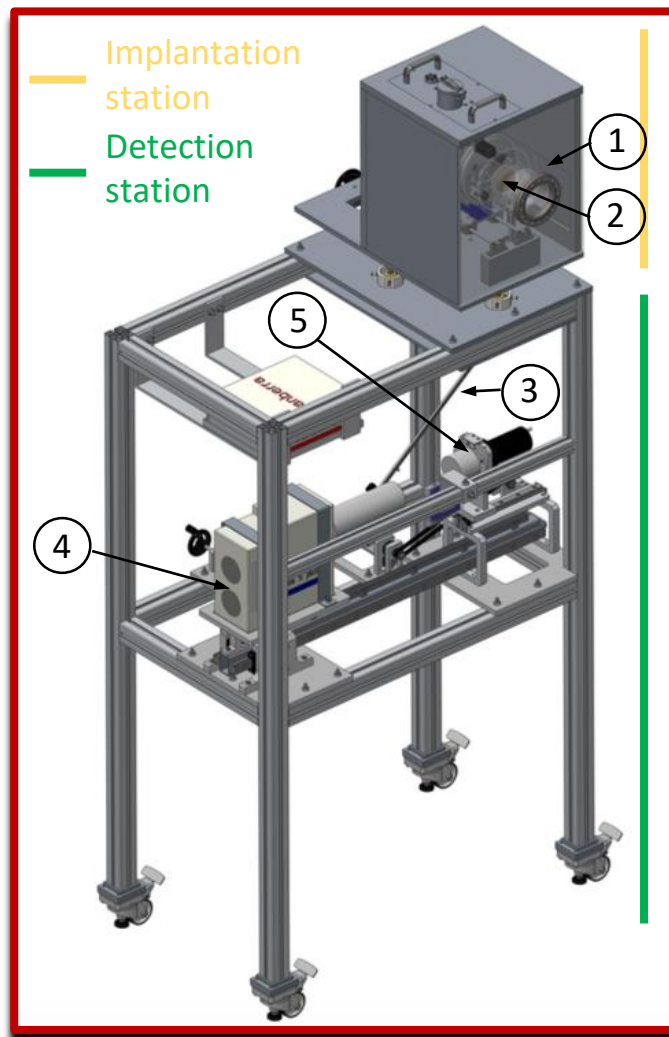
LaBr₃ Detector



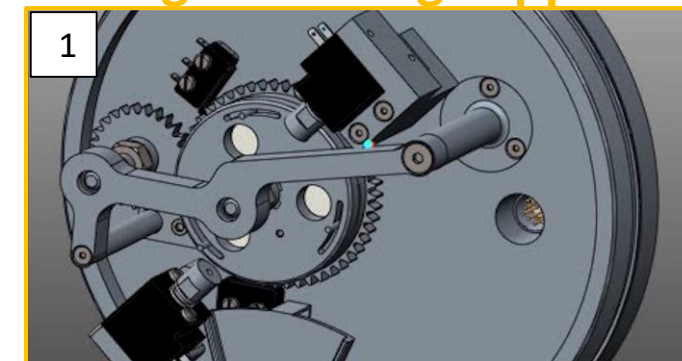
HPGe detector



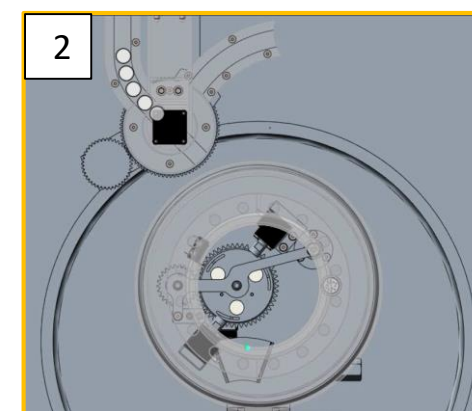
IRIS



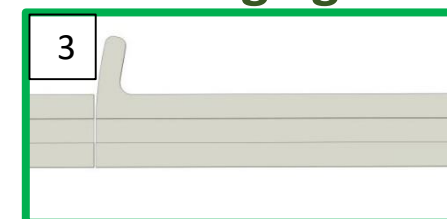
Target rotating support



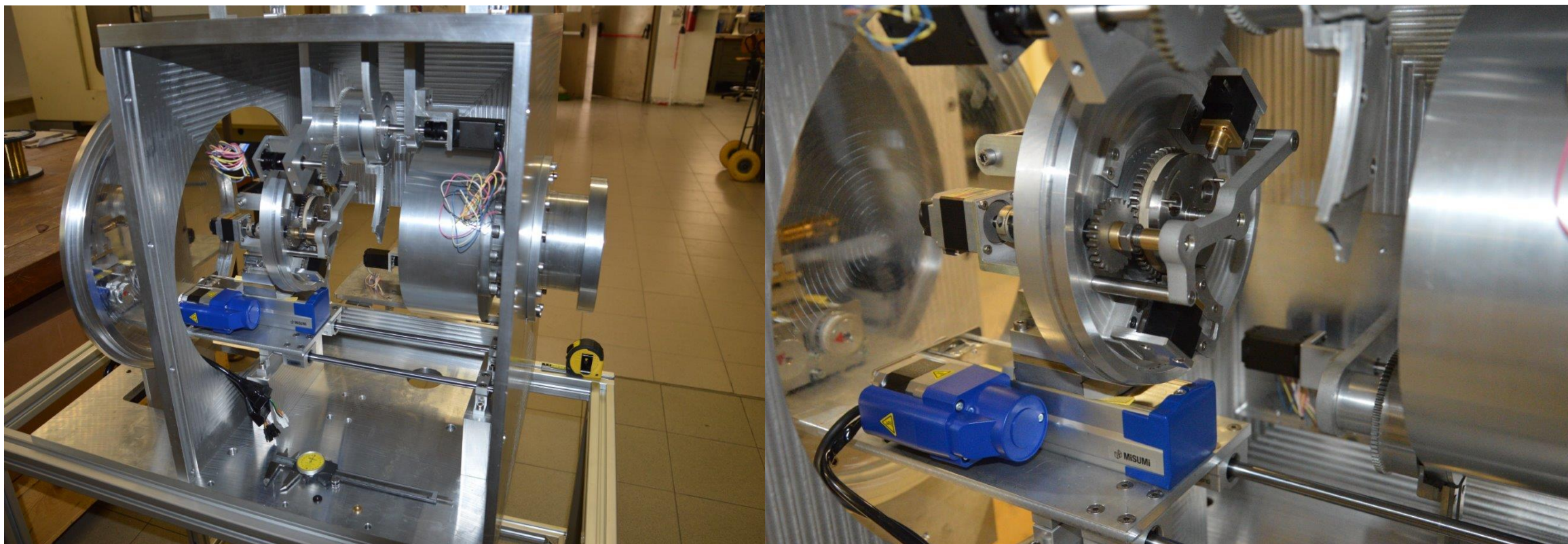
Target refill system



Discharge guide



IRIS deposition station



Deposition station of the ISOLPHARM Radionuclide Implantation Station (IRIS), realized in collaboration with INFN-BO.

IRIS possible spectroscopy system

The system had to satisfy the **following constraints**:

- Allowing for the characterization of elements and isotopes contained inside irradiated sample.
- Short Measuring Session (10-20 minutes)
- Allowing for the counting of pure β -emitters.

The whole system could also be used for decay spectroscopy exploiting β - γ coincidence.

Possible detectors:

3"x3" LaBr₃ Crystal



- Fair energy resolution < 30 keV at 1.3 MeV.
- Fast anode signal ~ 100 ns.
- Fast acquisition rate up to 500 kcps.
- Significant intrinsic background to be considered (~ 1 kHz)

HPGe



- Very good energy resolution: 2 keV at 1.3 MeV.
- Slow preamplified signal (~ 100 μ s).
- Typical acquisition rate < 10 kcps.

Conclusions

Conclusions

- **Laser spectroscopy and applications**
 - Photoionization can ensure high quality RIBs
 - At INFN-LNL the ionization schemes of several elements can be studied with the offline test set-ups
 - With eventual set-up upgrades, photoionization could be selective towards isomers
- **Nuclide production with ISOL for medicine and nuclear physics**
 - Different solid target materials could be used at SPES ensuring the availability of a wide set of RIBs for both nuclear physics studies and nuclear medicine applications
 - The development of an ISOL target is not trivial and requires several steps, that can be performed with the available competences at LNL
 - ISOL target development could benefit of innovative technologies and collaborations with other institutions
- **Decay spectroscopy of nuclide of medical interest**
 - For the collection and quality control of ISOL produced radionuclides, a specific device is being developed and tested
 - Such device could be also used to produce isotopically pure radioactive sources

Thank you for your kind attention

