

PAUL SCHERRER INSTITUT



A. Amato :: Paul Scherrer Institute :: NUM Division

Use of muons to investigate materials:

Not only μ SR...., but also the
Muon-Induced X-ray Emission (MIXE) Technique

- **S. Biswas, L. Gerchow, G. Janka, M. Heiss, C. Chen, A. Yamato, H. Luetkens, Th. Prokscha et al.,** [LMU - PSI](#)

- I. Megatli, K. Peter, L. Raselli, [Museum Augustiniana Raurica, Switzerland](#)

- A. Remhof, R. Asakura, [EMPA](#)

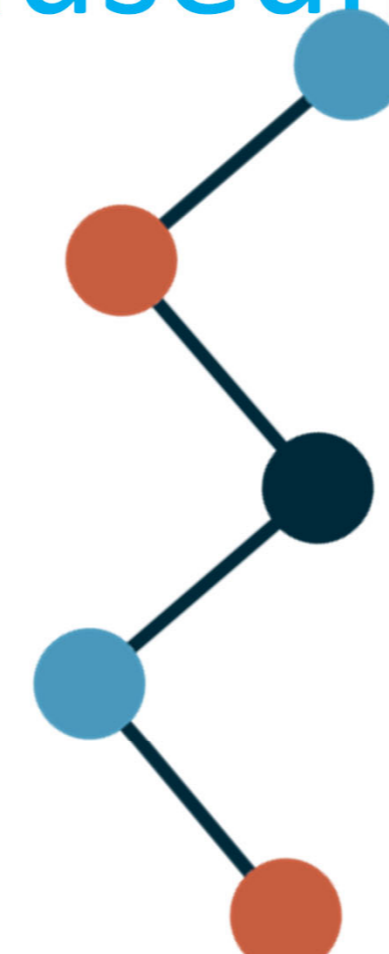
- B. Hoffmann, [Bern](#)

- K. Ninomiya and A. Sato, [Osaka University](#)

- **and (part of the muX Collaboration)**

A. Antognini^{1,2}, N. Berger³, T. Cocolios⁴, R. Dressler¹, P. Indelicato⁵, K. Jungmann⁶, K. Kirch^{1,2}, **A. Knecht**¹, J. Nuber^{1,2}, A. Papa^{1,7}, R. Pohl⁸, M. Pospelov^{9,10}, E. Rapisarda¹, P. Reiter¹¹, N. Ritjoho^{1,2}, S. Roccia¹², N. Severijns⁴, A. Skawran^{1,2}, **S. Vogiatzi**^{1,2}, F. Wauters³, and L. Willmann⁶

¹Lab. for Particle Physics, Paul Scherrer Institut, Villigen, Switzerland; ²ETH Zürich, Switzerland; ³University of Mainz, Germany; ⁴KU Leuven, Belgium; ⁵LKB Paris, France; ⁶University of Groningen, The Netherlands; ⁷University of Pisa and INFN, Pisa, Italy; ⁸Johannes Gutenberg Universität Mainz, Germany; ⁹University of Victoria, Canada; ¹⁰Perimeter Institute, Waterloo, Canada; ¹¹Universität zu Köln, Germany; ¹²Université Grenoble Alpes, France



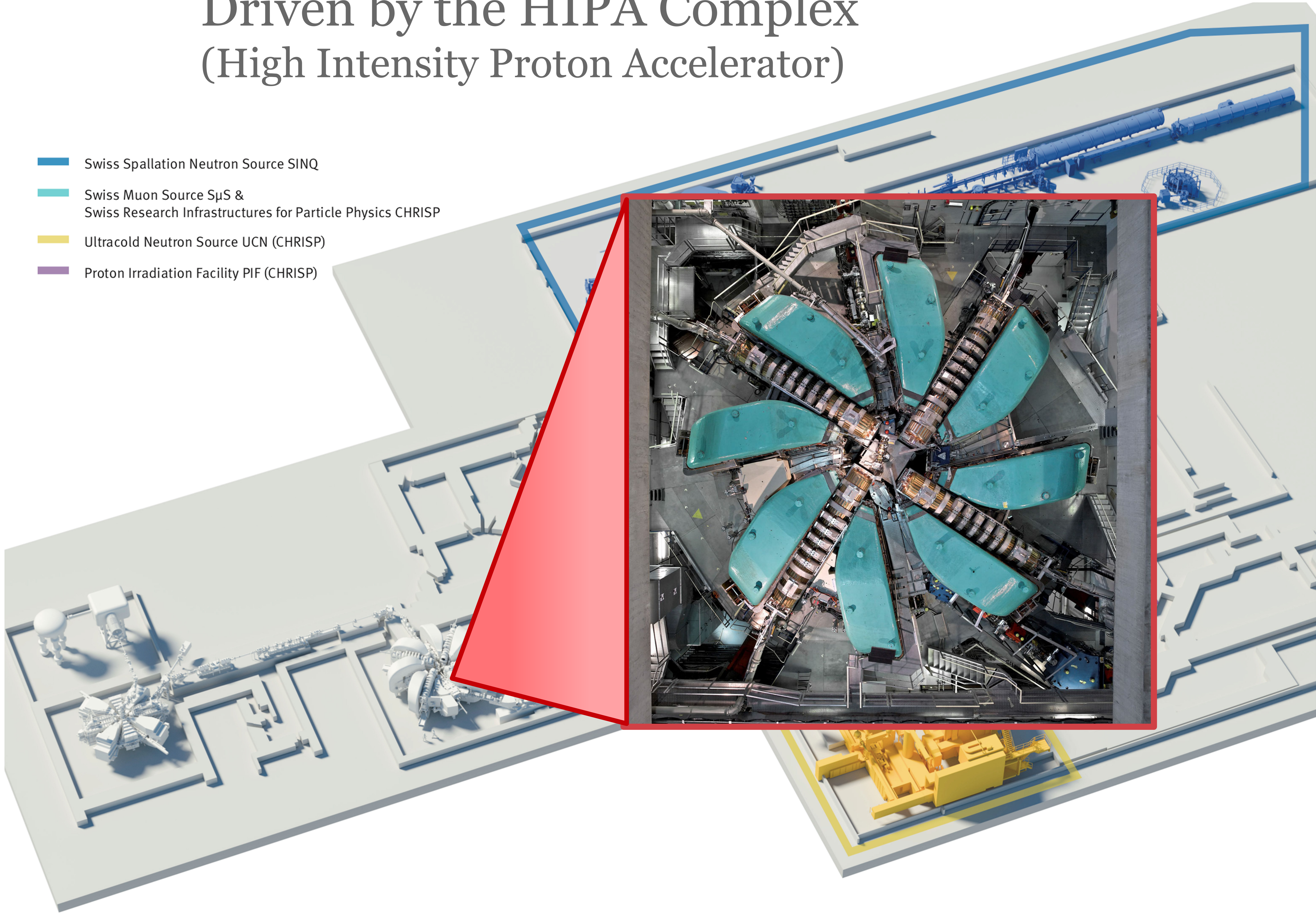
Swiss National
Science Foundation



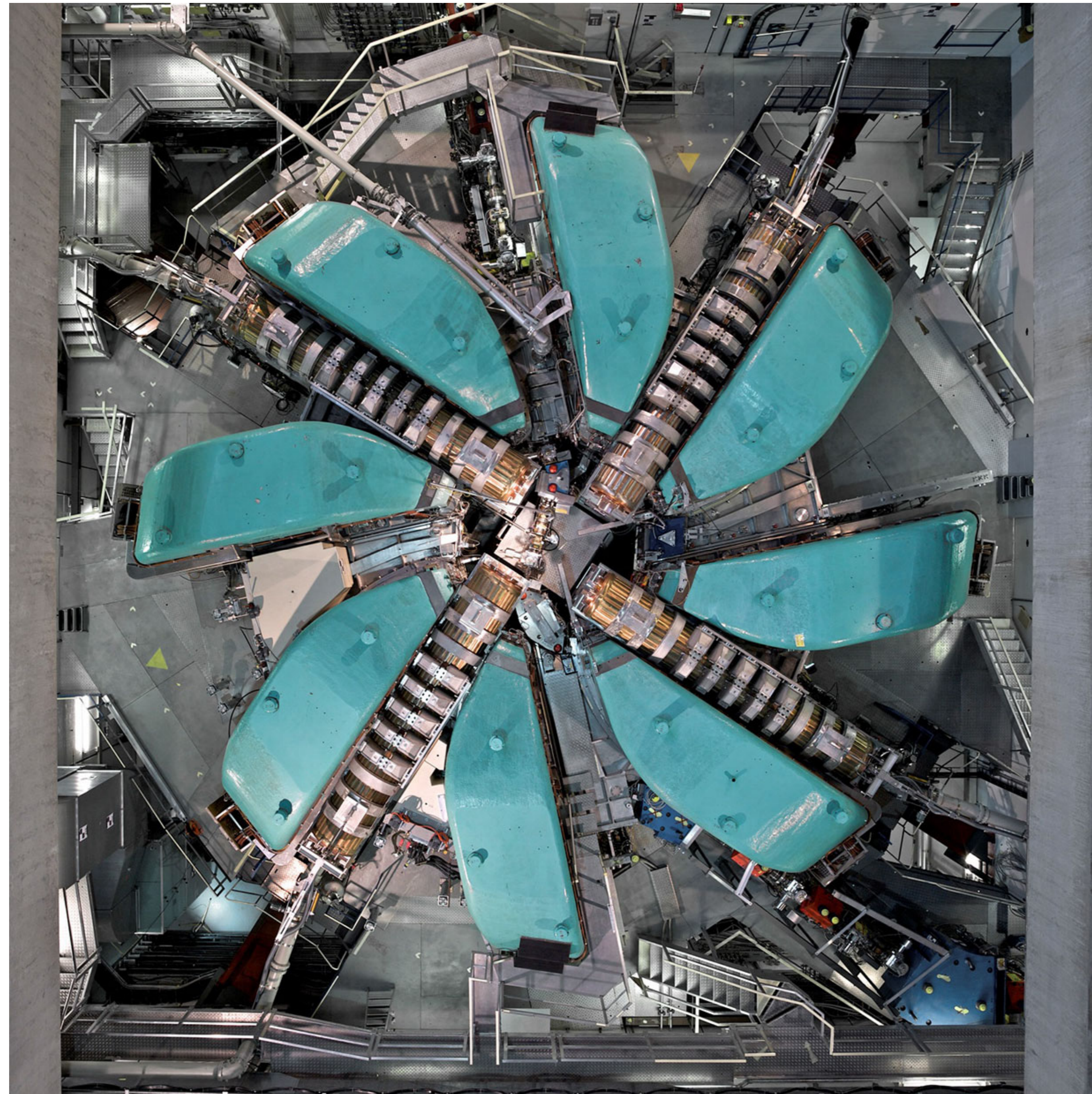


Large Research Facilities at PSI Driven by the HIPA Complex (High Intensity Proton Accelerator)

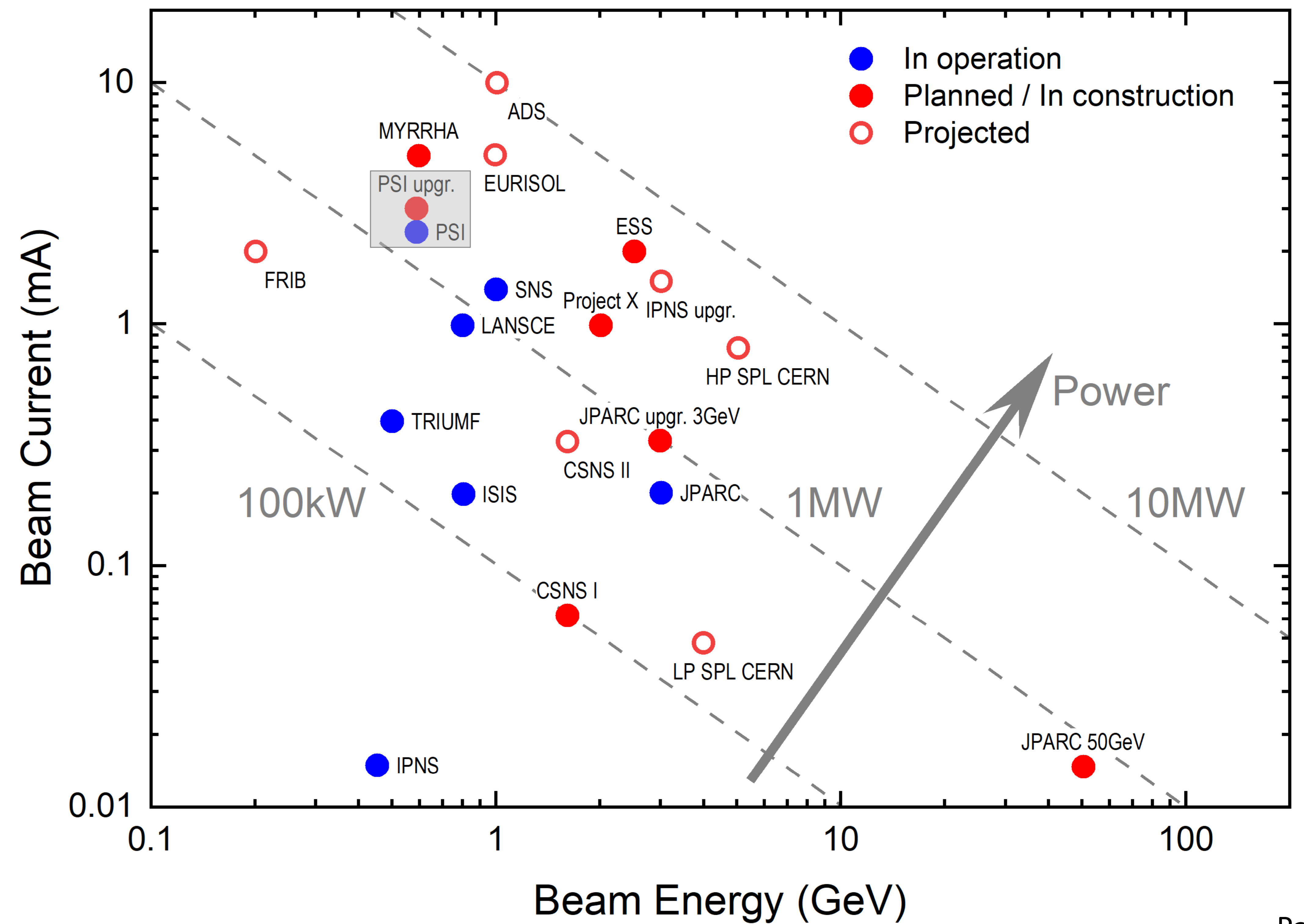
- Swiss Spallation Neutron Source SINQ
- Swiss Muon Source μ S & Swiss Research Infrastructures for Particle Physics CHRISP
- Ultracold Neutron Source UCN (CHRISP)
- Proton Irradiation Facility PIF (CHRISP)



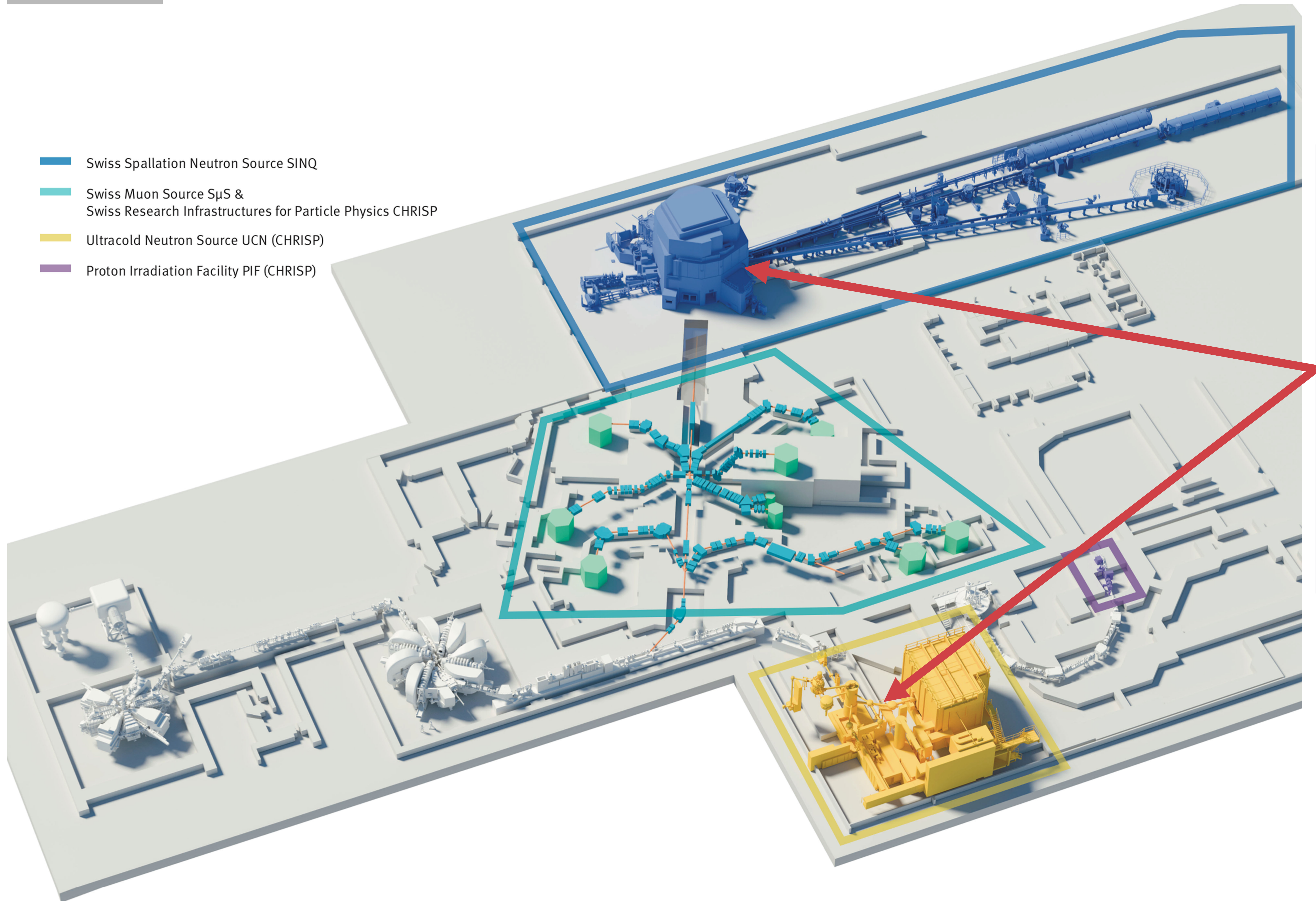
HIPA Proton Accelerator (Overview)



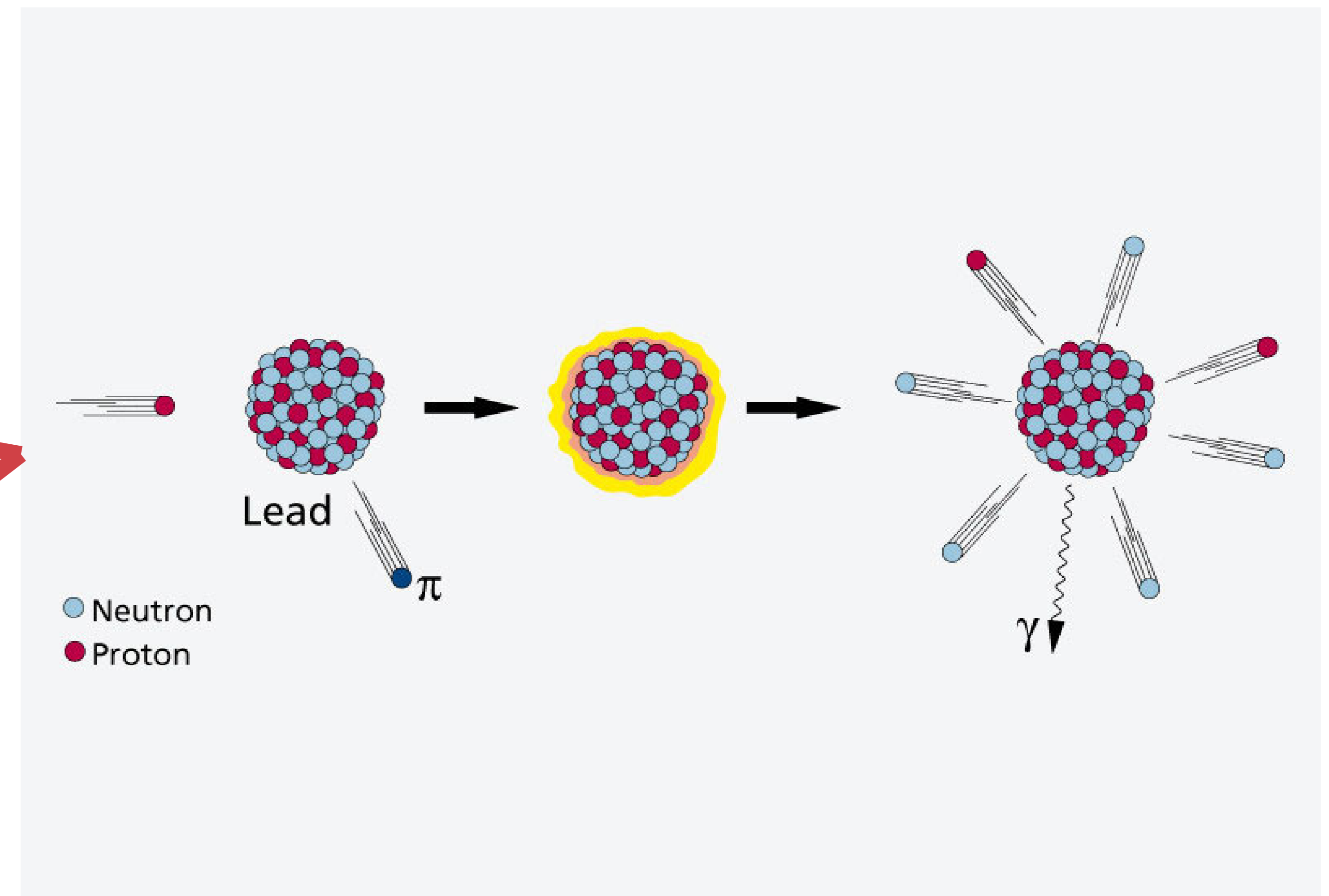
Injection Energy	72 MeV
Extraction Energy	590 MeV
Extraction Momentum	1.2 GeV/c
Energy spread (FWHM)	ca. 0.2 %
Beam Emittance	ca. 2 pi mm x mrad
Beam Current	2.2 mA DC
Accelerator Frequency	50.63 MHz
Time Between Pulses	19.75 ns
Bunch Width	ca. 0.3 ns
Extraction Losses	ca. 0.03 %



Production of neutrons

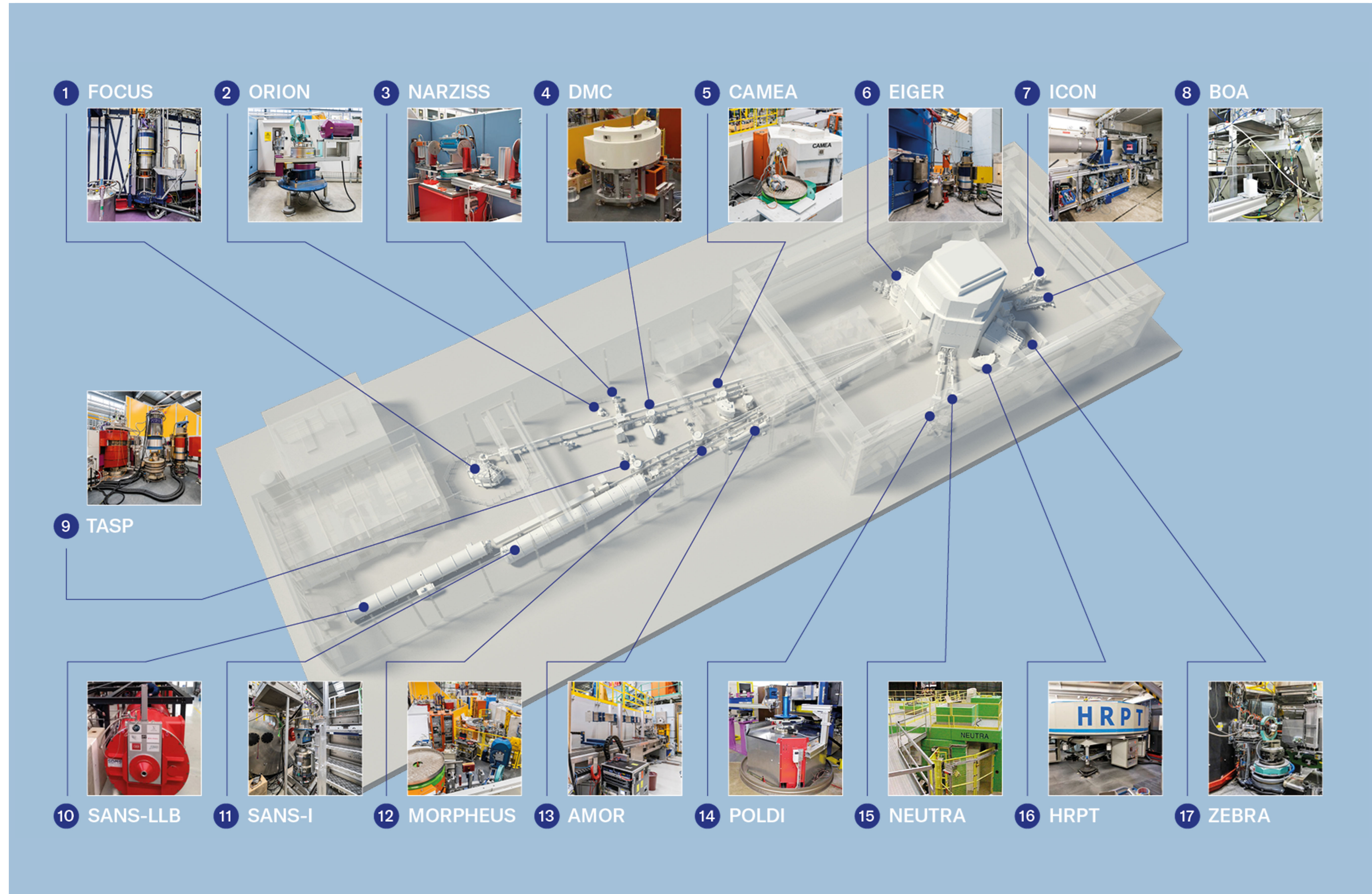


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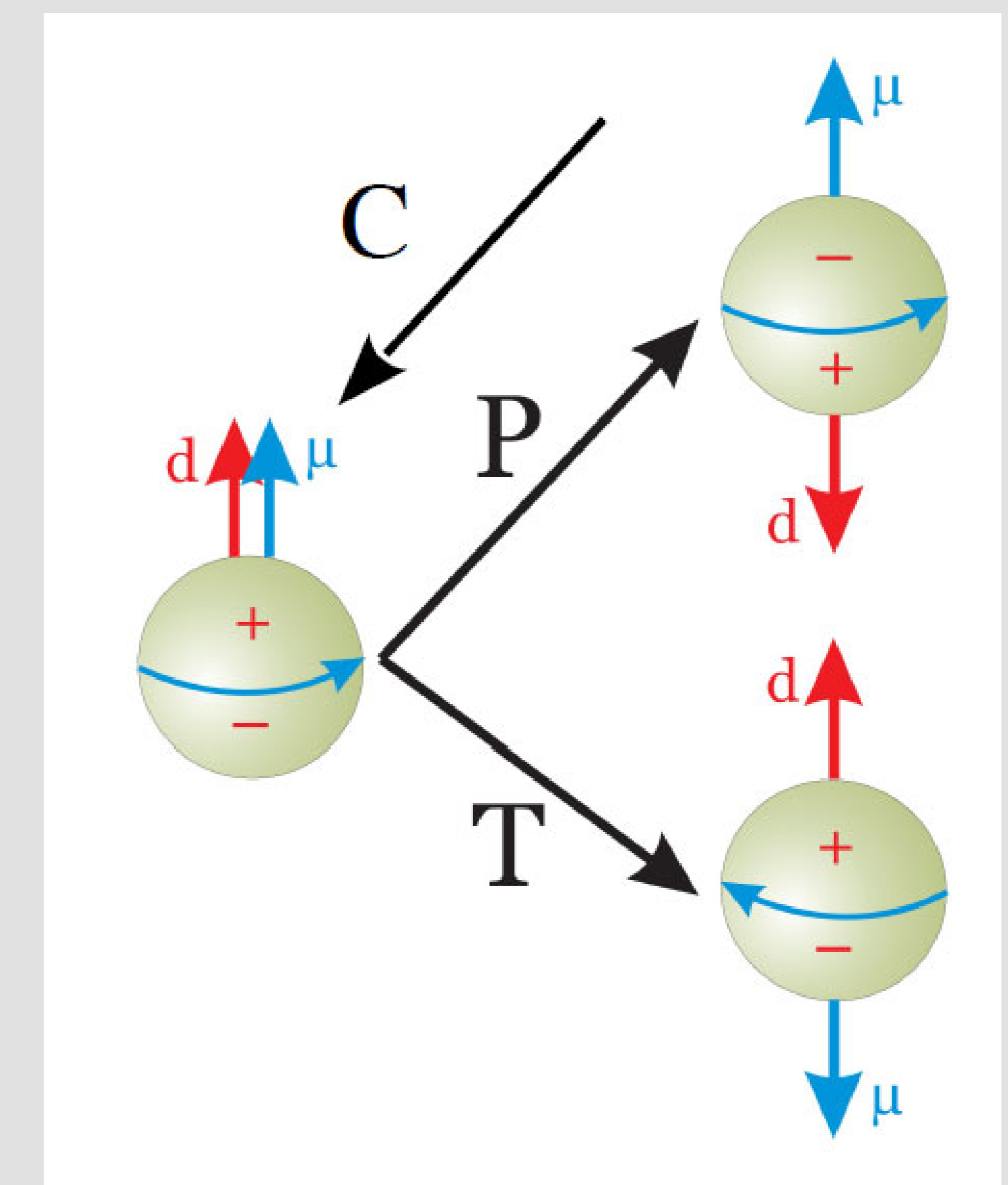
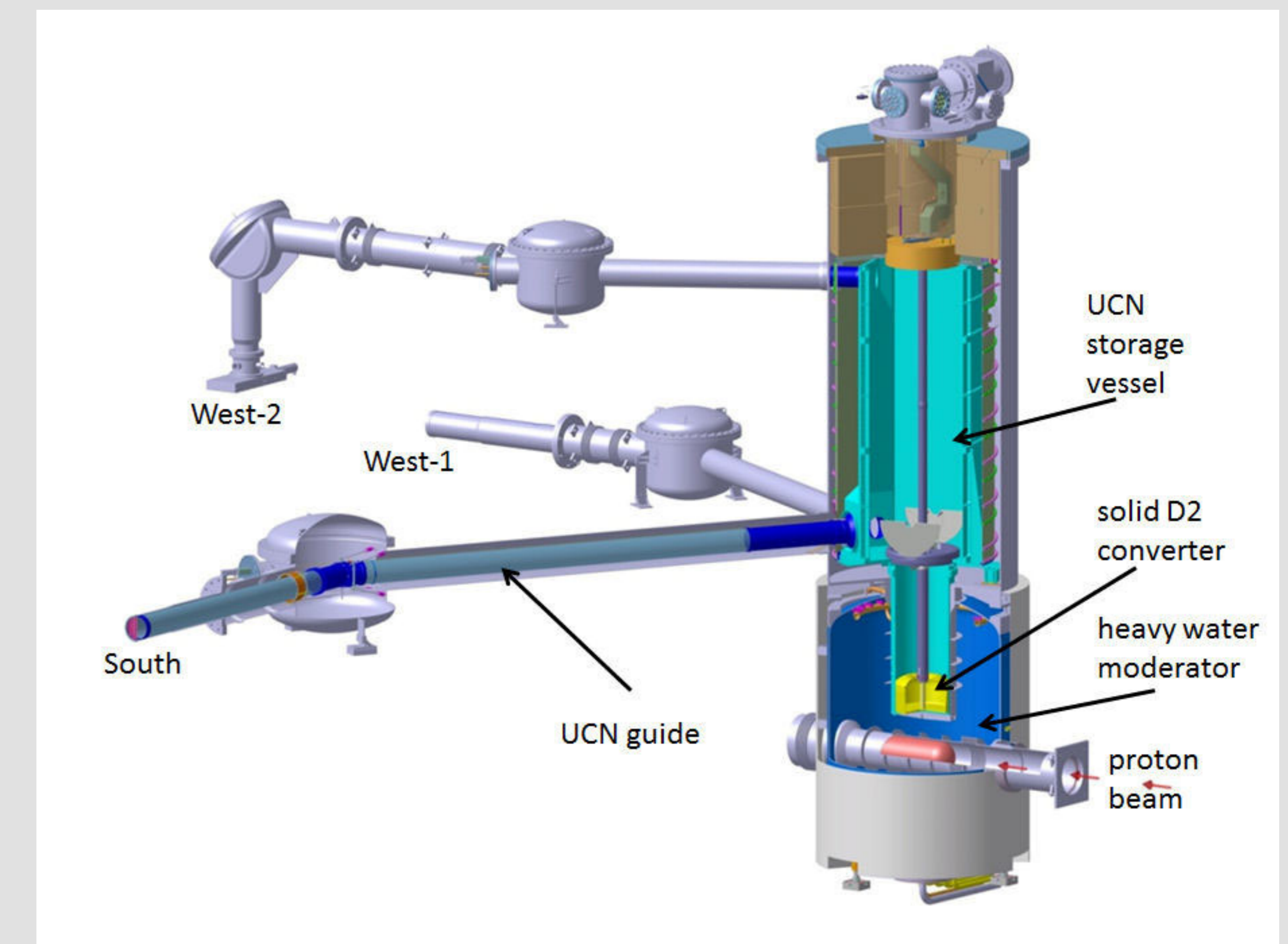
Spallation reactions

Neutrons for Condensed Matter and Particle Physics



SINQ

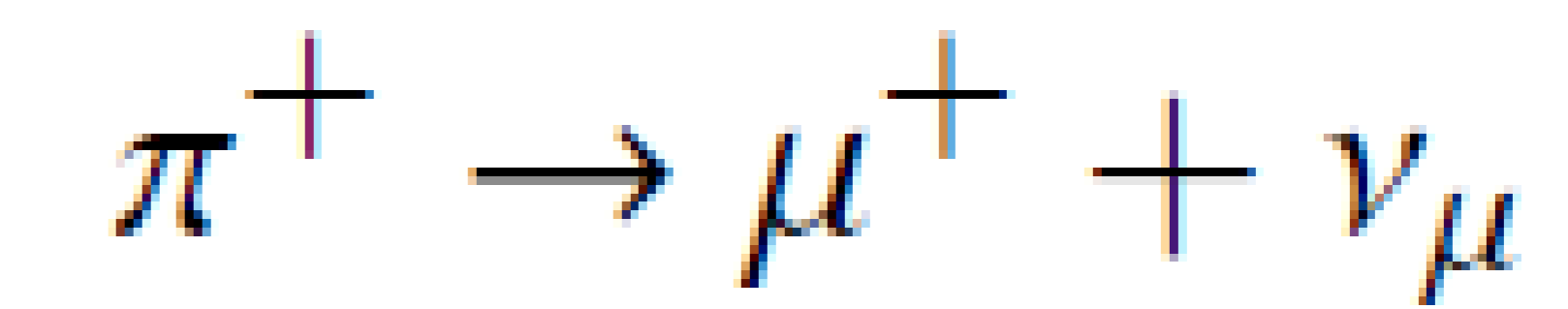
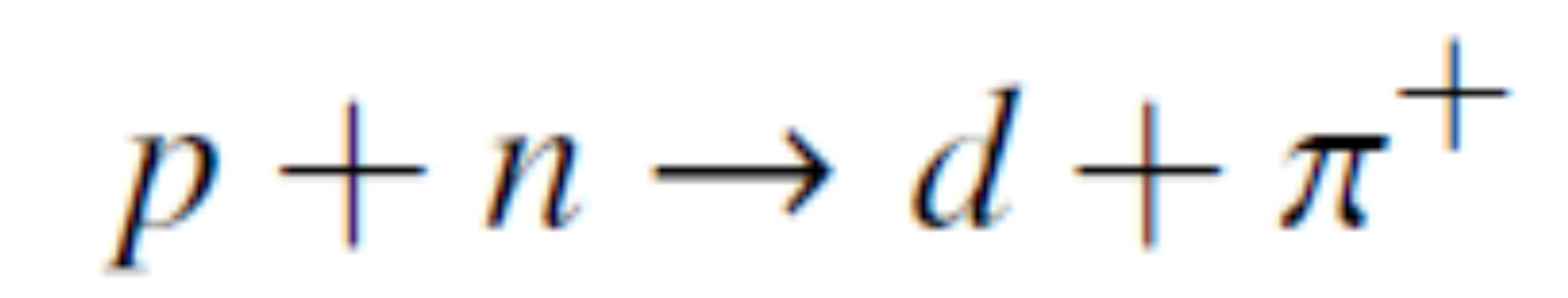
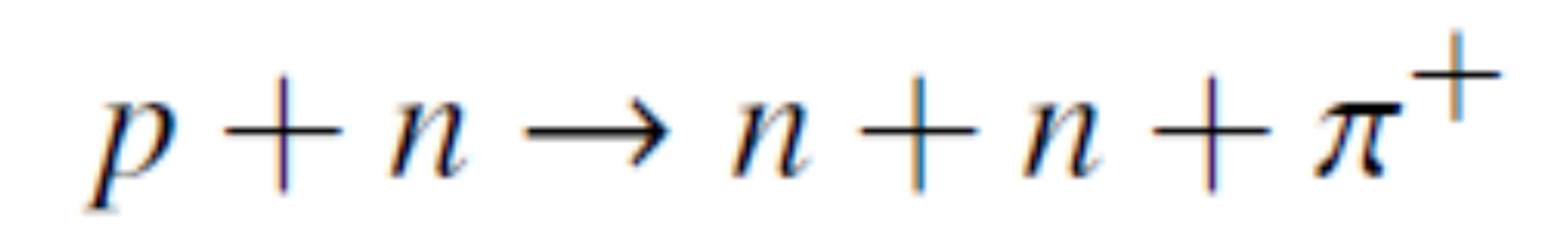
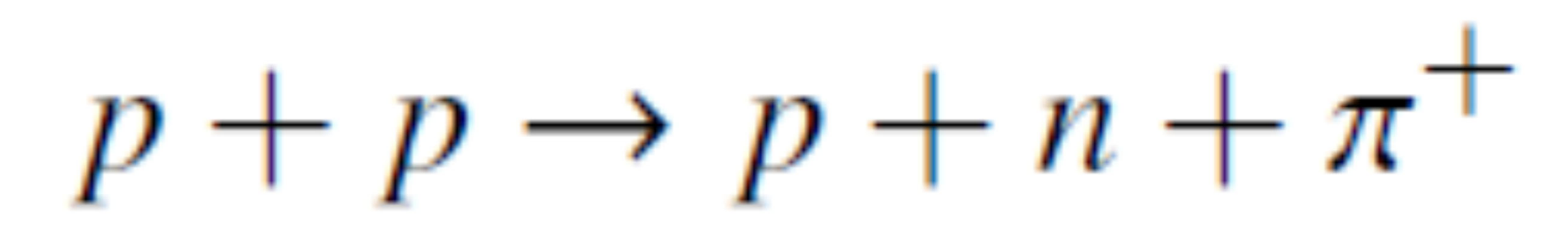
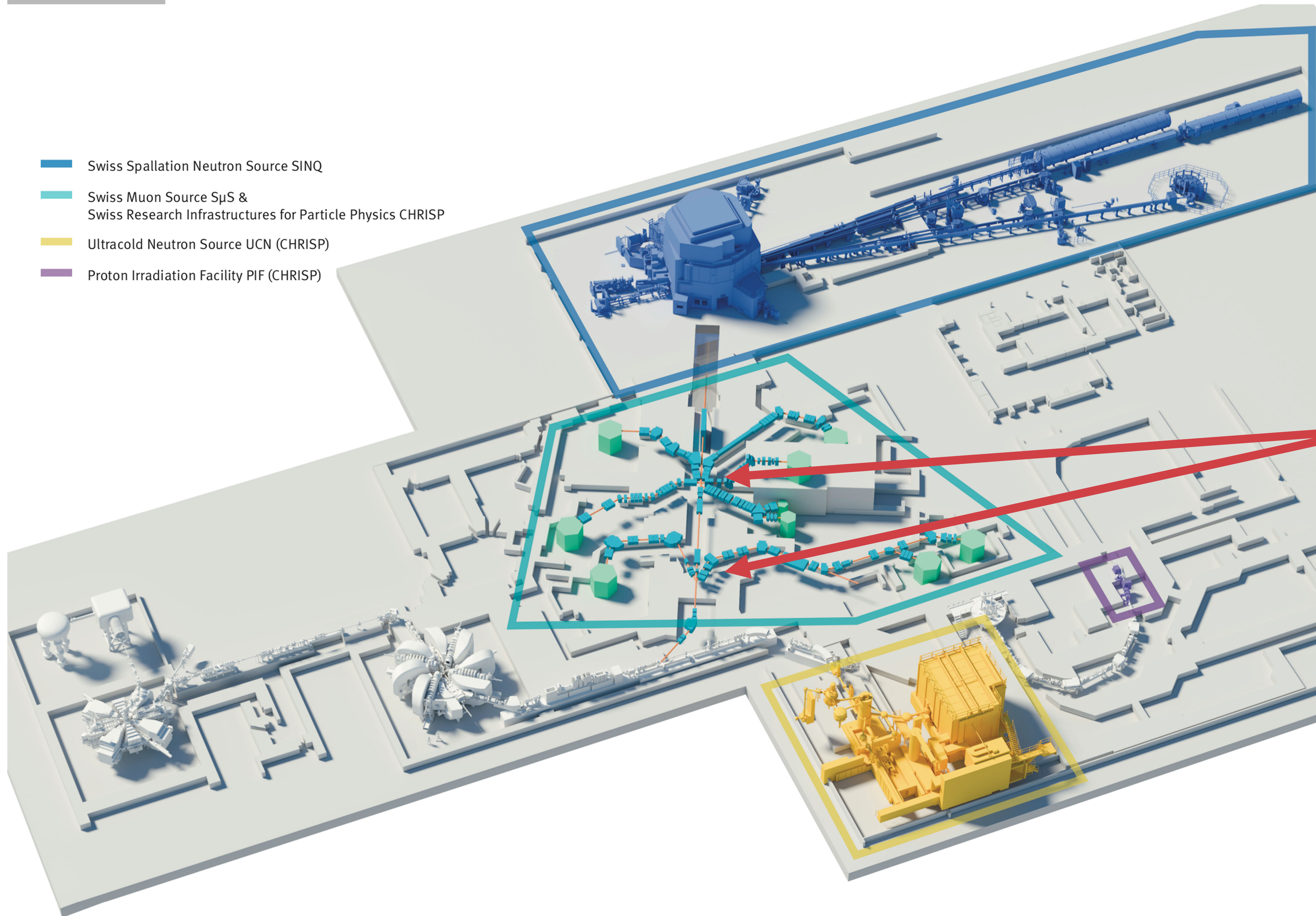
n-EDM (neutron – Electrical Dipole Moment)



Production of muons

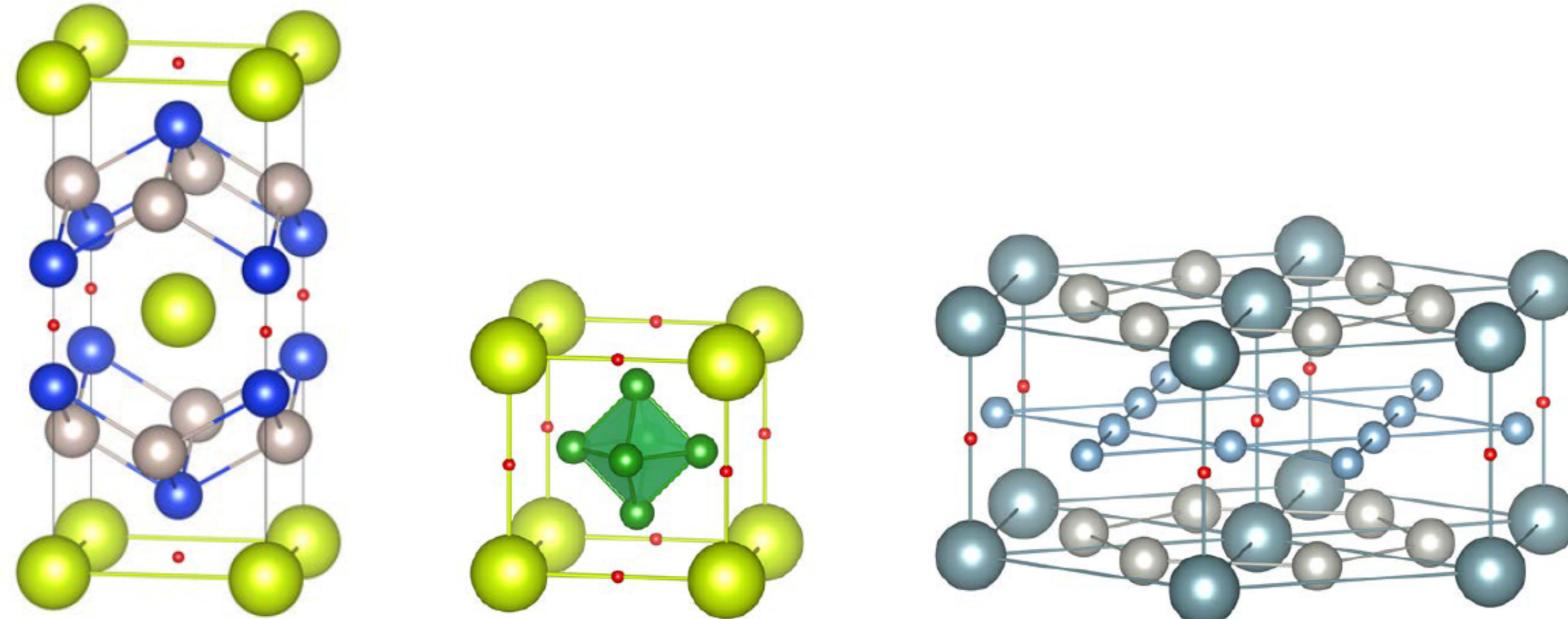


- Swiss Spallation Neutron Source SINQ
- Swiss Muon Source SμS & Swiss Research Infrastructures for Particle Physics CHRISP
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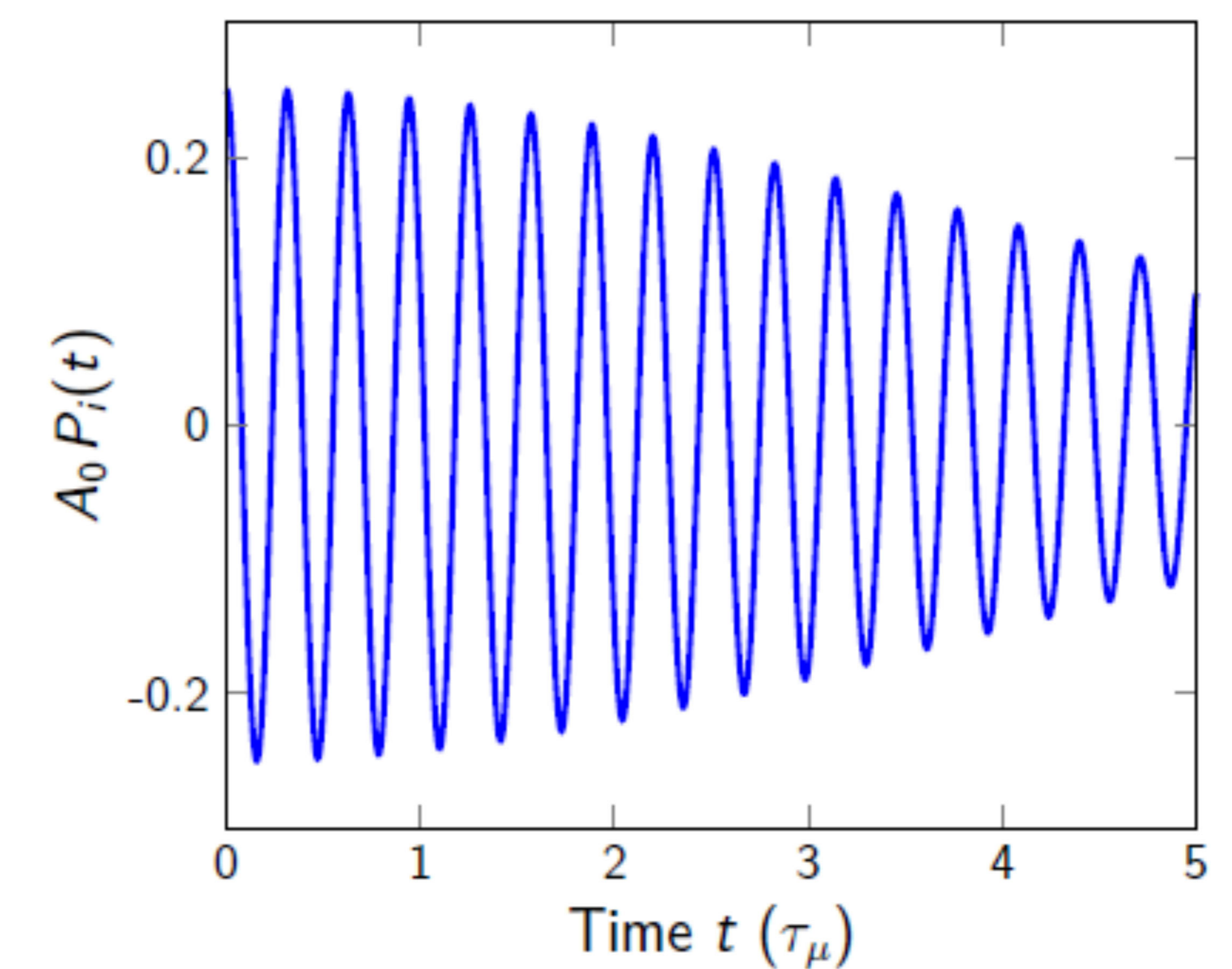
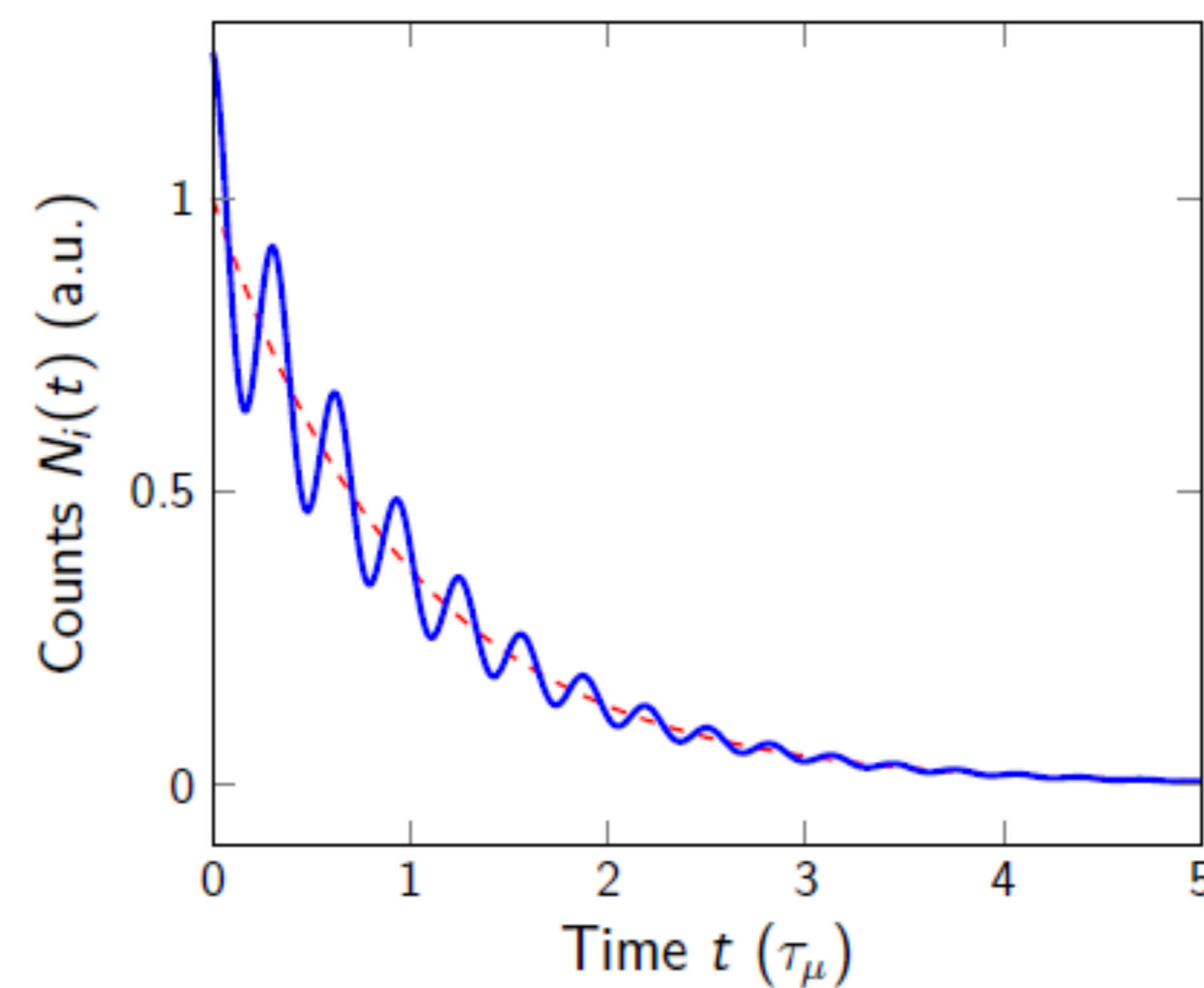
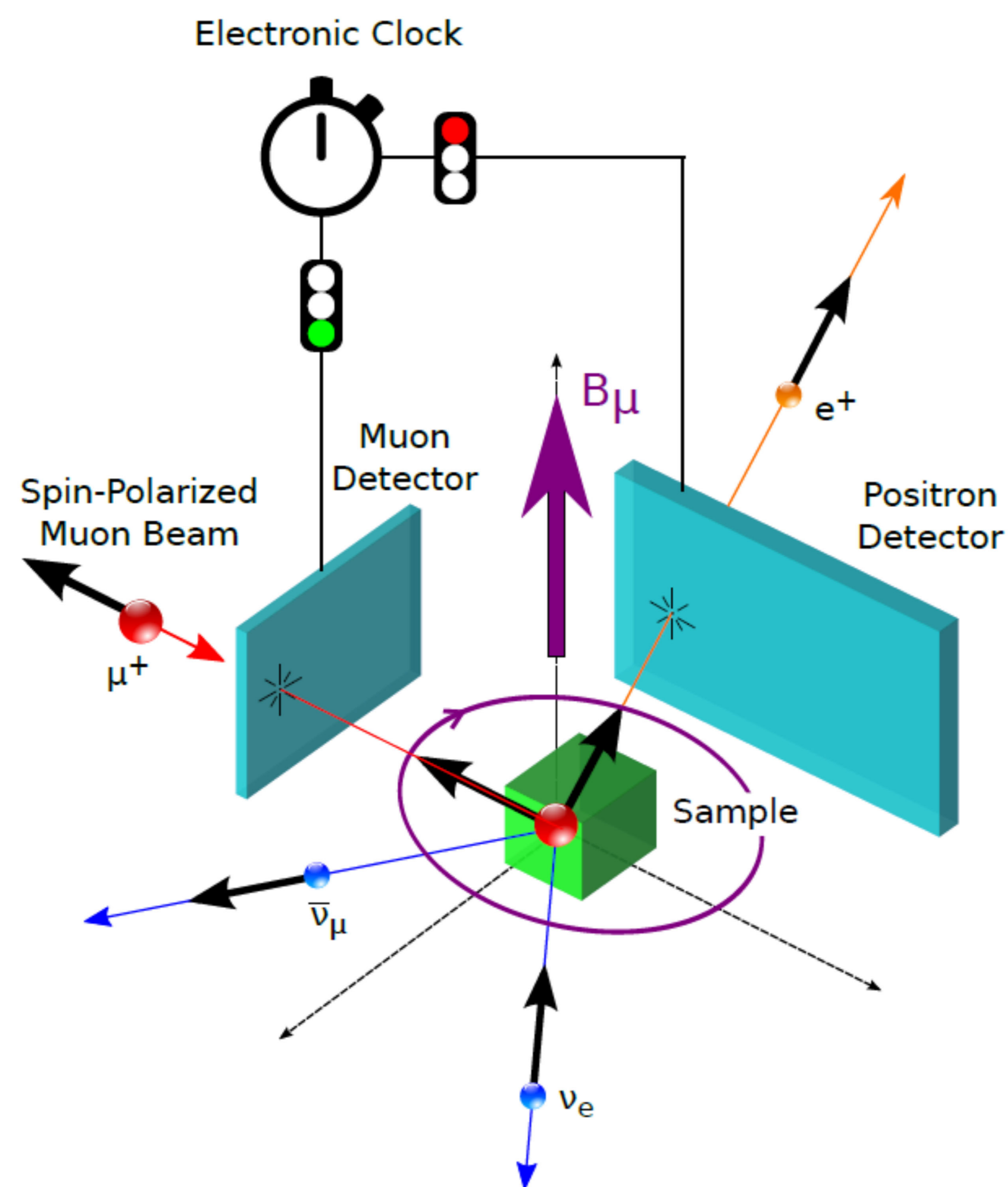


Positive Muons - μ SR Technique

- Implant positive muons into a target
 - Muon stays polarized
 - interstitial sites (minimum of electrostatic potential)



- Observe the behaviour of the muon spin (through the positron emission)



Frequency is a direct measure of the internal field

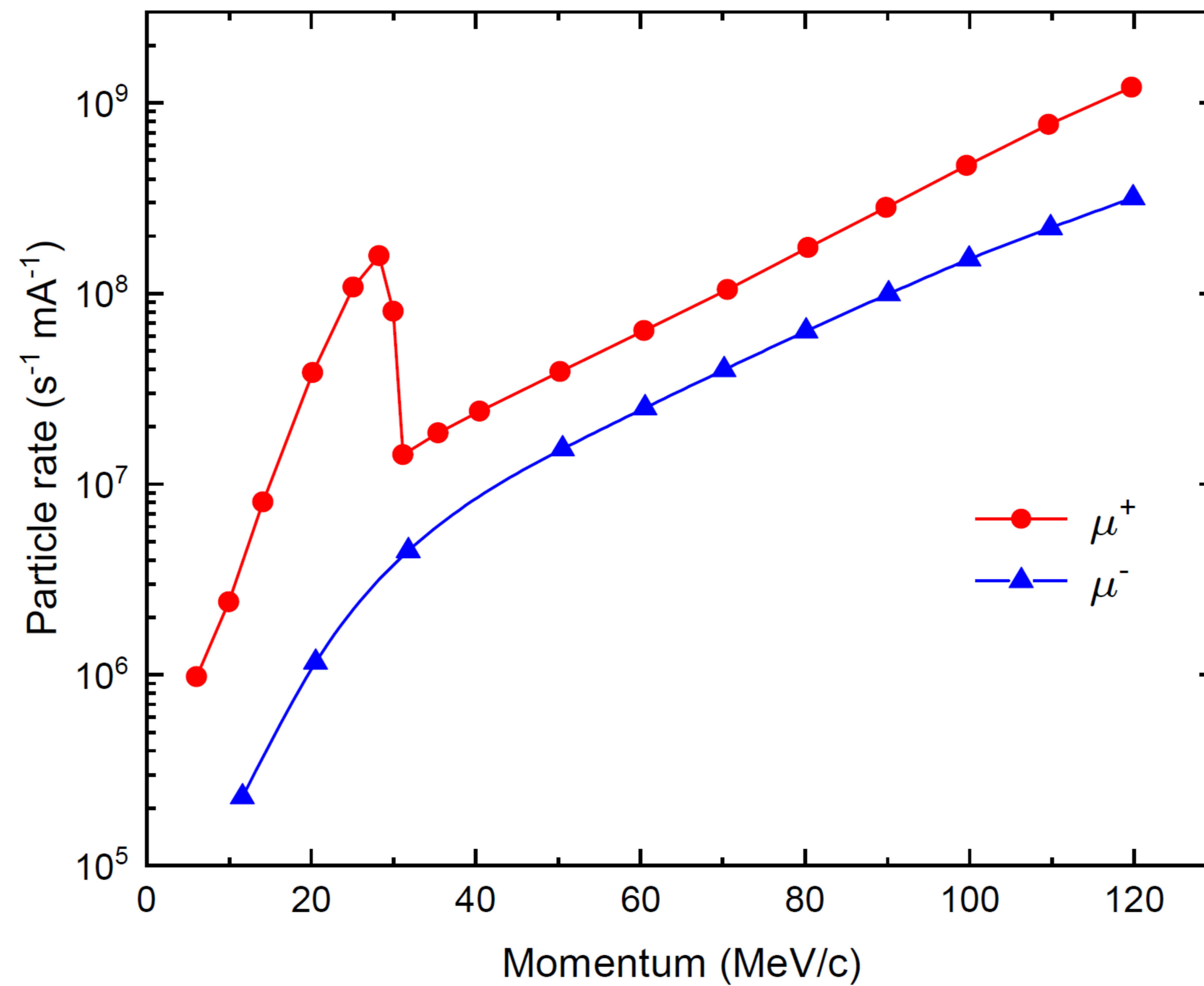
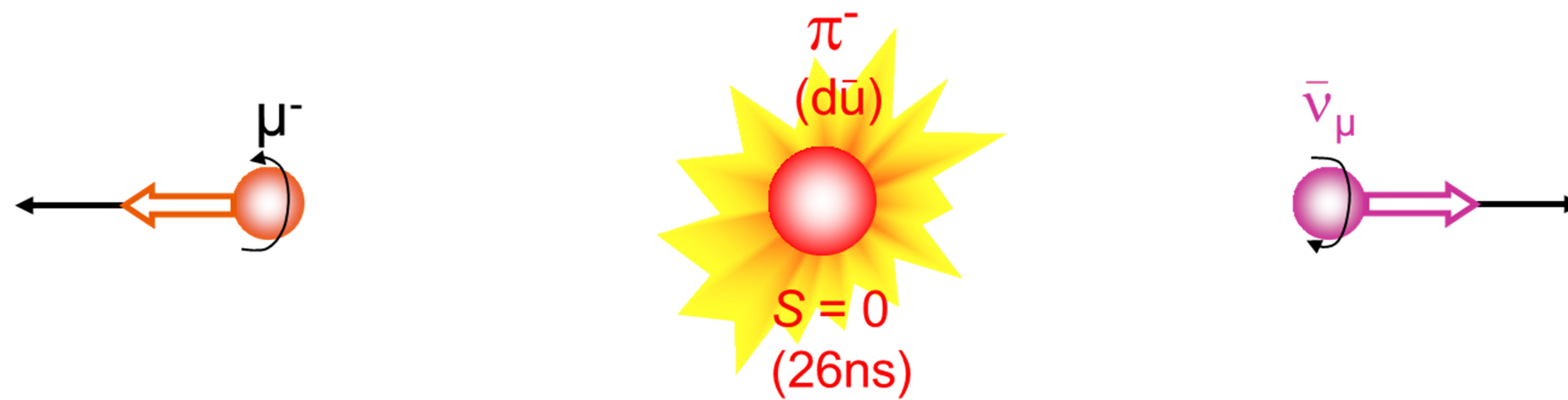
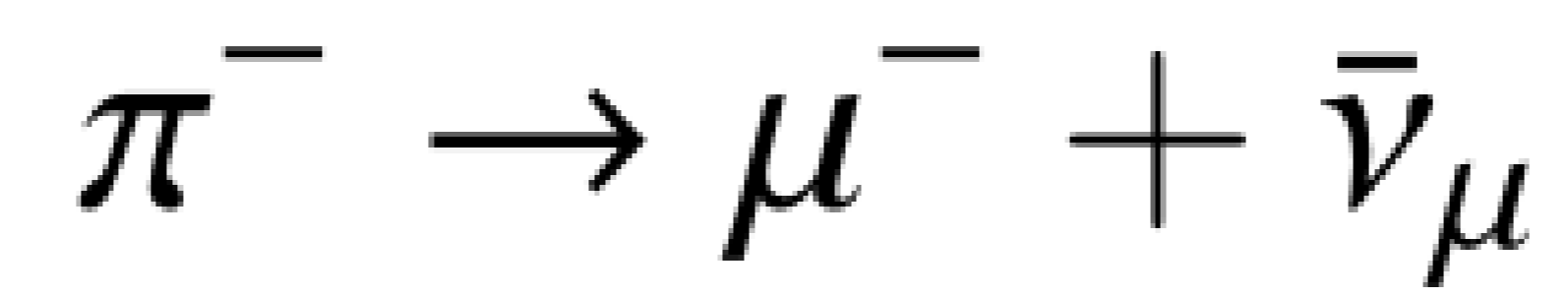
→ magnetism, SC, diffusion, etc

<https://www.psi.ch/de/lmu/lectures>

What about negative muons

Negative pions also produced in the production target, e.g.: $p + n \rightarrow p + p + \pi^-$

The negative pion decay into a negative muon:



What about negative muons

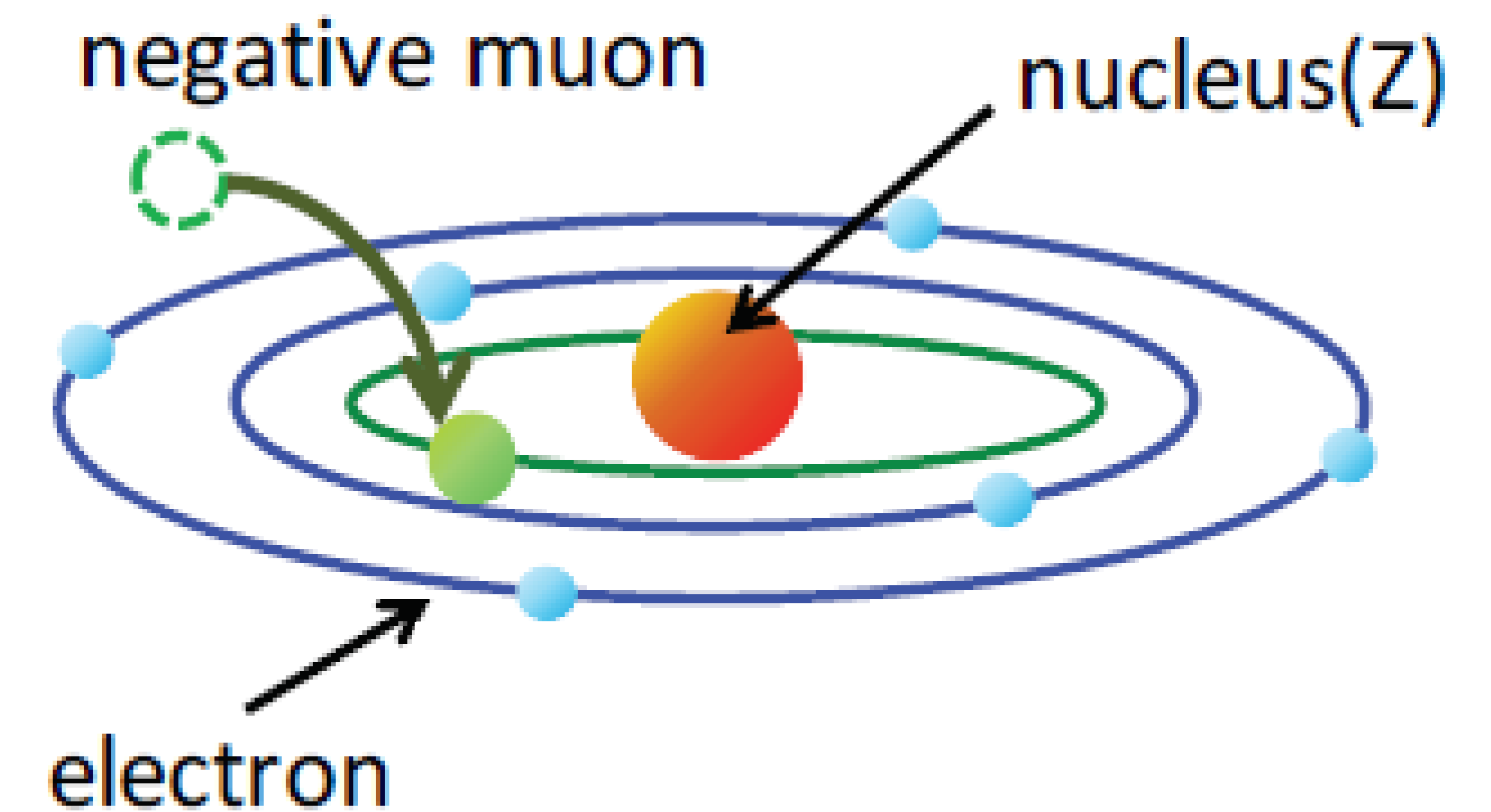
Subsequently, can be sent into a sample (similar to positive muons).

Negative muons are **captured** by an atom of the sample (do not stop at an interstitial site)

→ **Two captures...**

First Capture: the negative muon is captured by the atom

Capture in a «muonic» orbital
(associated with the emission of
Auger electrons)



Very close to nuclei...

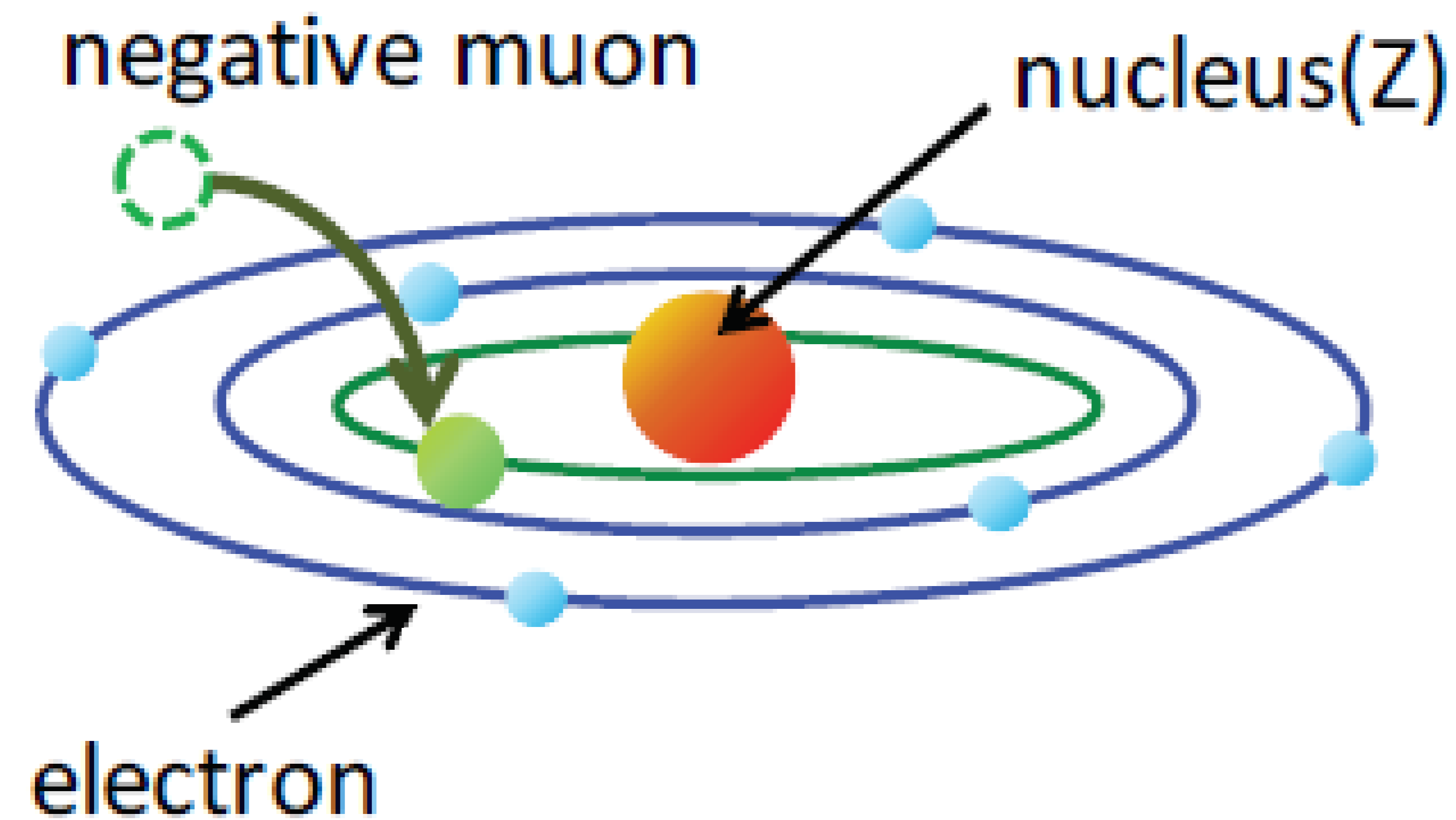
Since the muon has a mass about 207 times higher than the one of the electron, its orbits will be a factor 207 closer to the nucleus than the respective ones of the electron.

$$r_{n,\mu} = \frac{4\pi\epsilon_0\hbar^2 n^2}{\bar{M}_\mu Z e^2} = \frac{\hbar n^2}{\bar{M}_\mu Z c \alpha}$$

$$\alpha = \frac{1}{4\pi\epsilon_0} \frac{e^2}{\hbar c} \quad \text{fine structure constant}$$

What about negative muons – Level energy

The muon cascades down to its 1s ground state



Energy of the levels:

$$E_n = -\frac{\bar{M}_\mu Z^2 e^4}{(4\pi\epsilon_0)^2 2n^2 \hbar^2}$$

A bit more precisely:

$$E_n = -\frac{1}{2} \frac{(Z\alpha)^2 \bar{M}_\mu c^2}{n^2} - \frac{\bar{M}_\mu c^2}{2n^4} \left(\frac{n}{j + \frac{1}{2}} - \frac{3}{4} \right) (Z\alpha)^4 + \dots$$

What about negative muons – X-ray energy

As for the XRF (X-ray fluorescence) technique, X-rays are emitted during the muon cascade

- Remember for the electrons (Moseley law)

$$E_{i \rightarrow f, e} = \frac{\bar{M}_e}{M_e} R_y (Z - S_{scr, e})^2 \left(\frac{1}{n_f^2} - \frac{1}{n_i^2} \right)$$

$$Z_{eff} = (Z - S_{scr, e})$$

$$R_y = \frac{M_e e^4}{8 \epsilon_0^2 h^2}$$

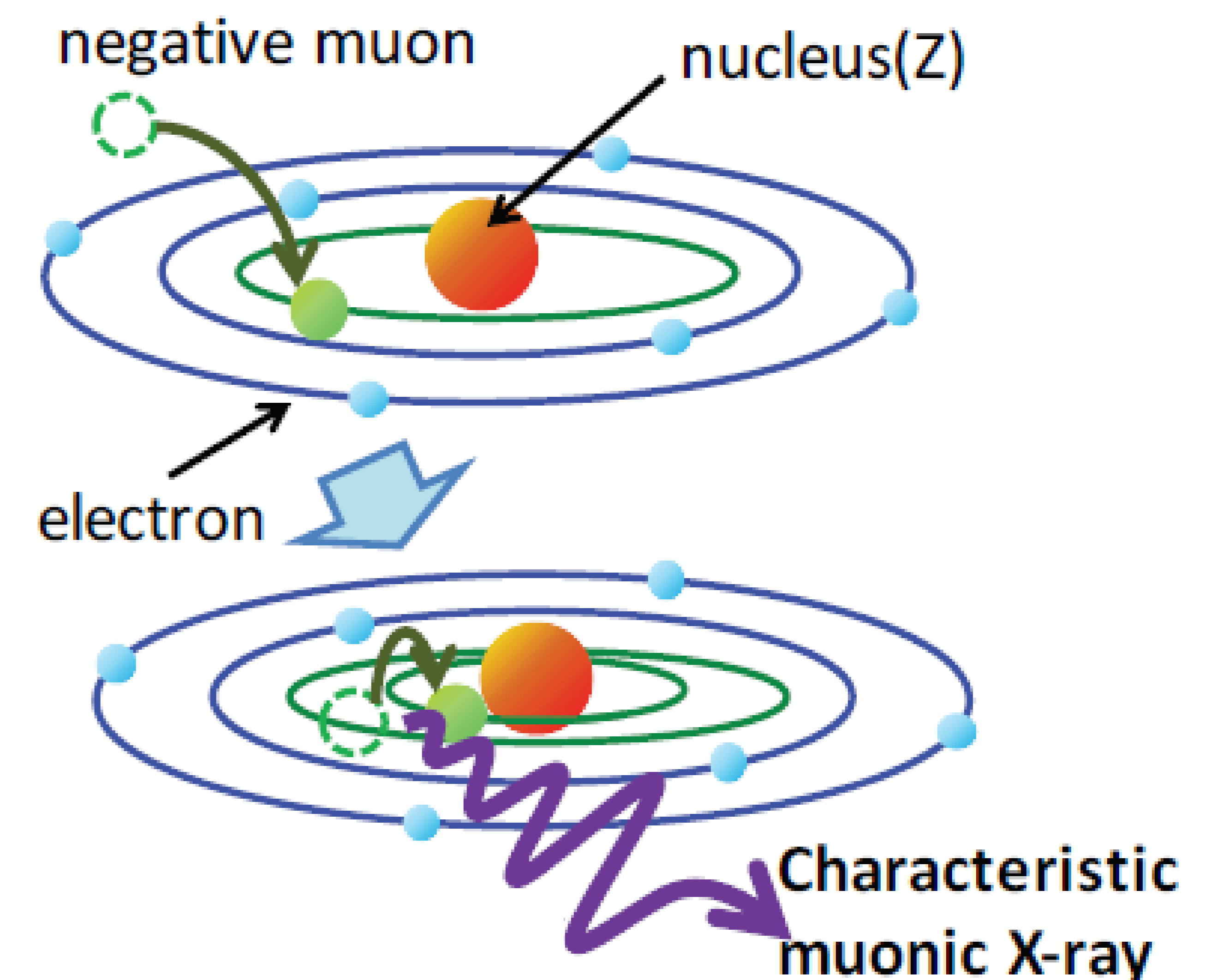
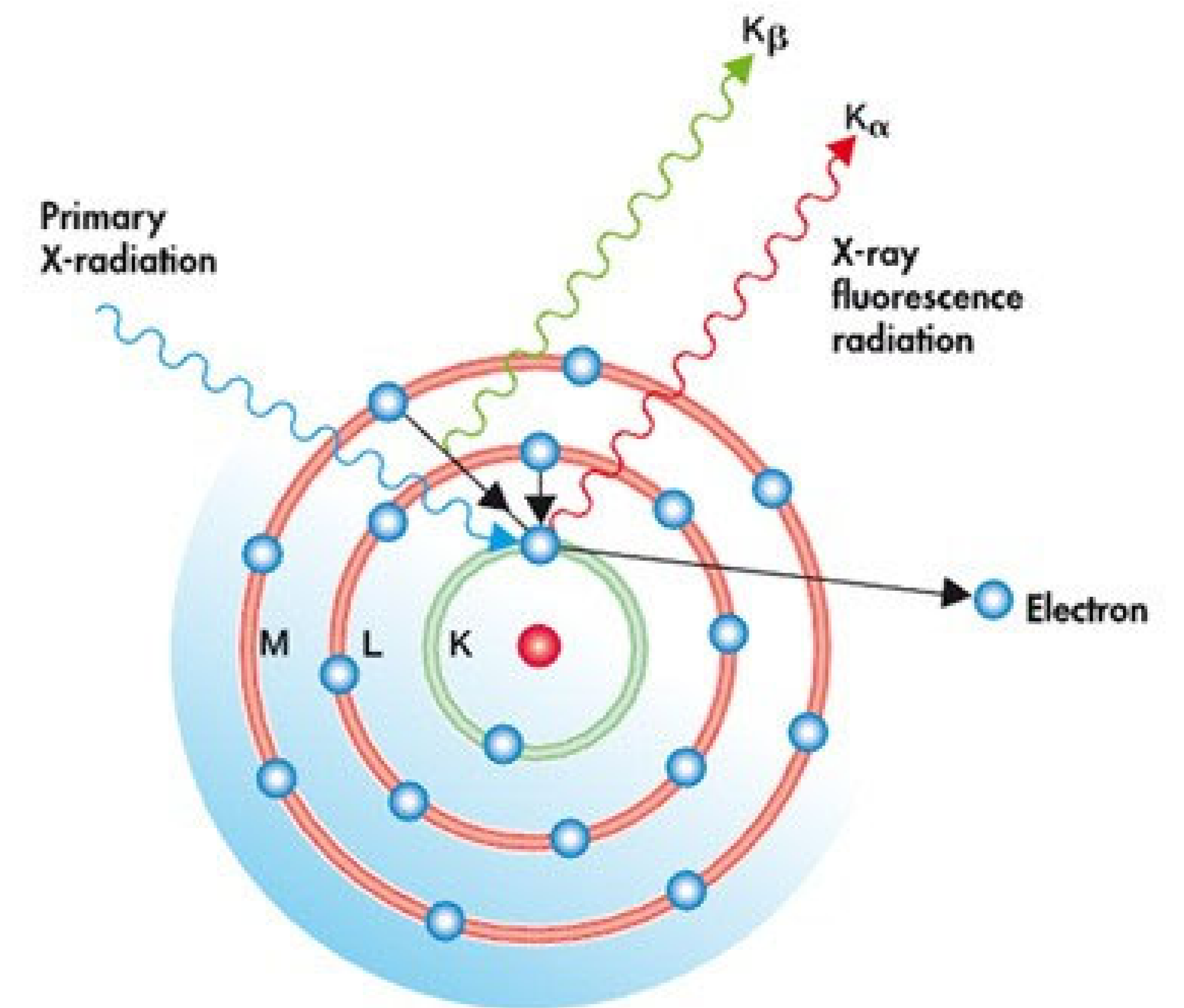
- For the muon:....

Energy of the Muonic X-rays:

$$E_{i \rightarrow f, \mu} = \frac{\bar{M}_\mu}{M_e} R_y (Z - S_{scr, \mu})^2 \left(\frac{1}{n_f^2} - \frac{1}{n_i^2} \right)$$

$$\simeq \frac{\bar{M}_\mu}{M_e} \times E_{i \rightarrow f, e}$$

$$\simeq 207 \times E_{i \rightarrow f, e} .$$



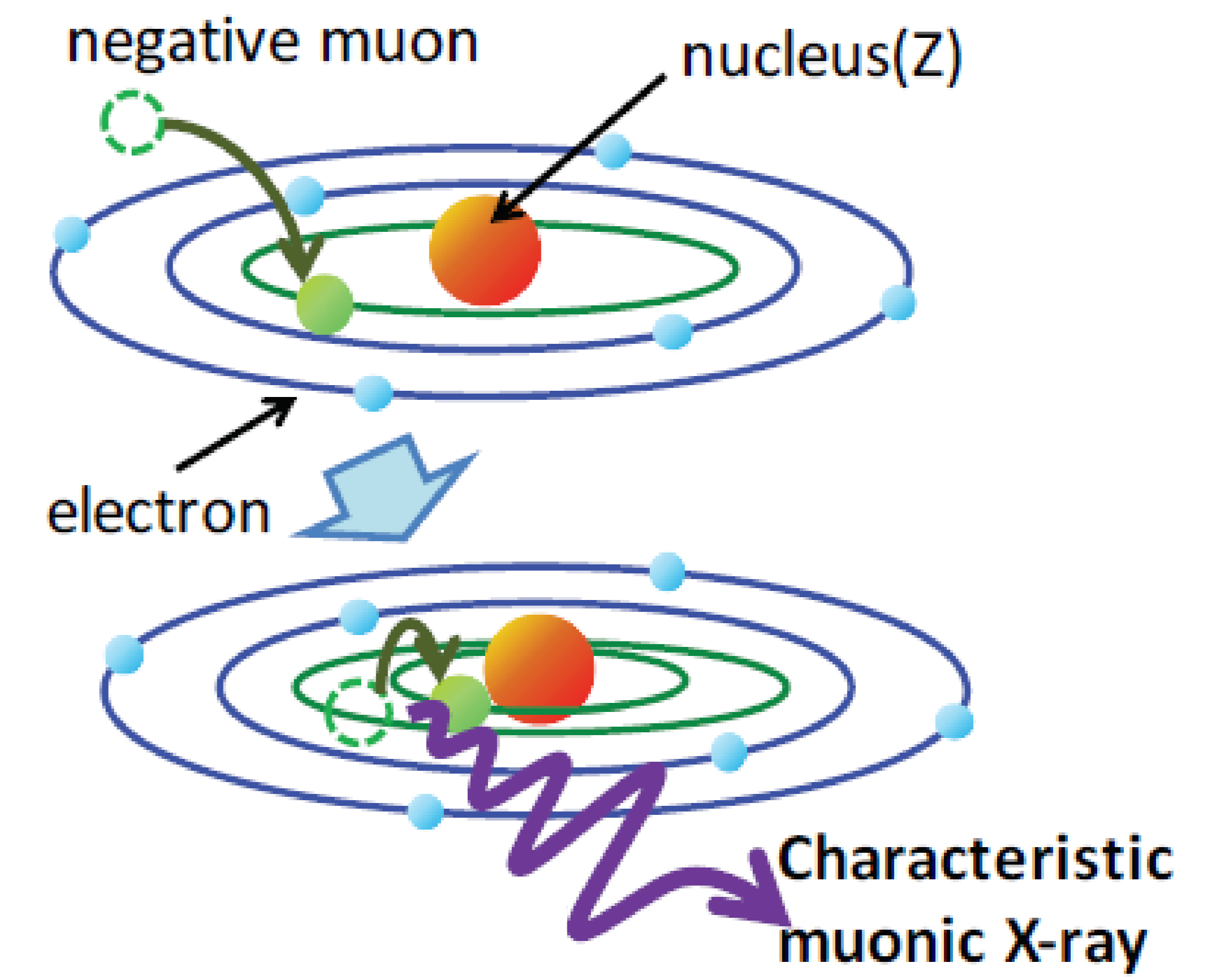
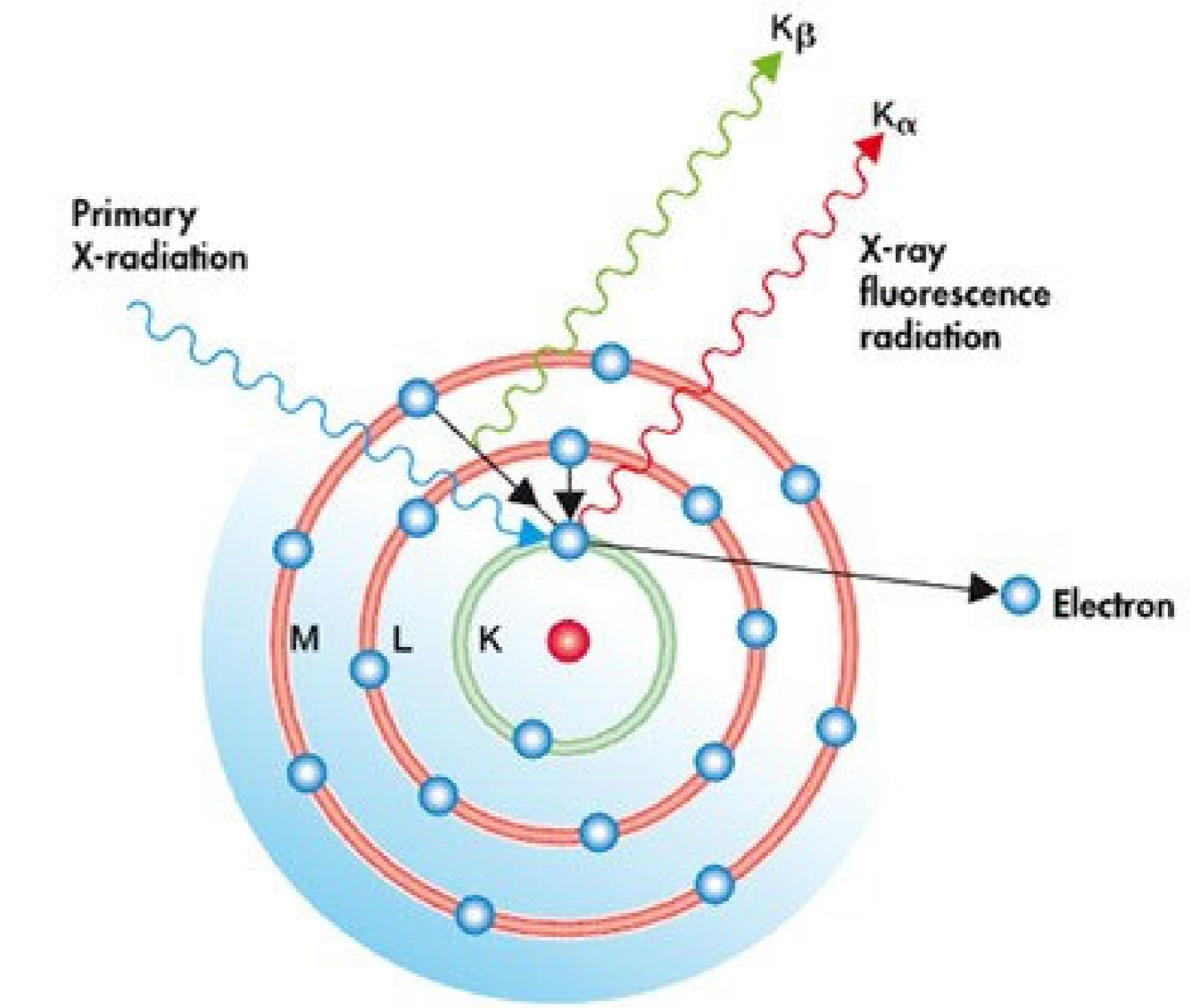
What about negative muons – X-ray energy

Energy of the X-rays:

$$2p_{1/2} \rightarrow 1s_{1/2}$$

$$E_{i \rightarrow f, e} = \frac{\bar{M}_e}{M_e} R_y (Z - S_{scr, e})^2 \left(\frac{1}{n_f^2} - \frac{1}{n_i^2} \right)$$

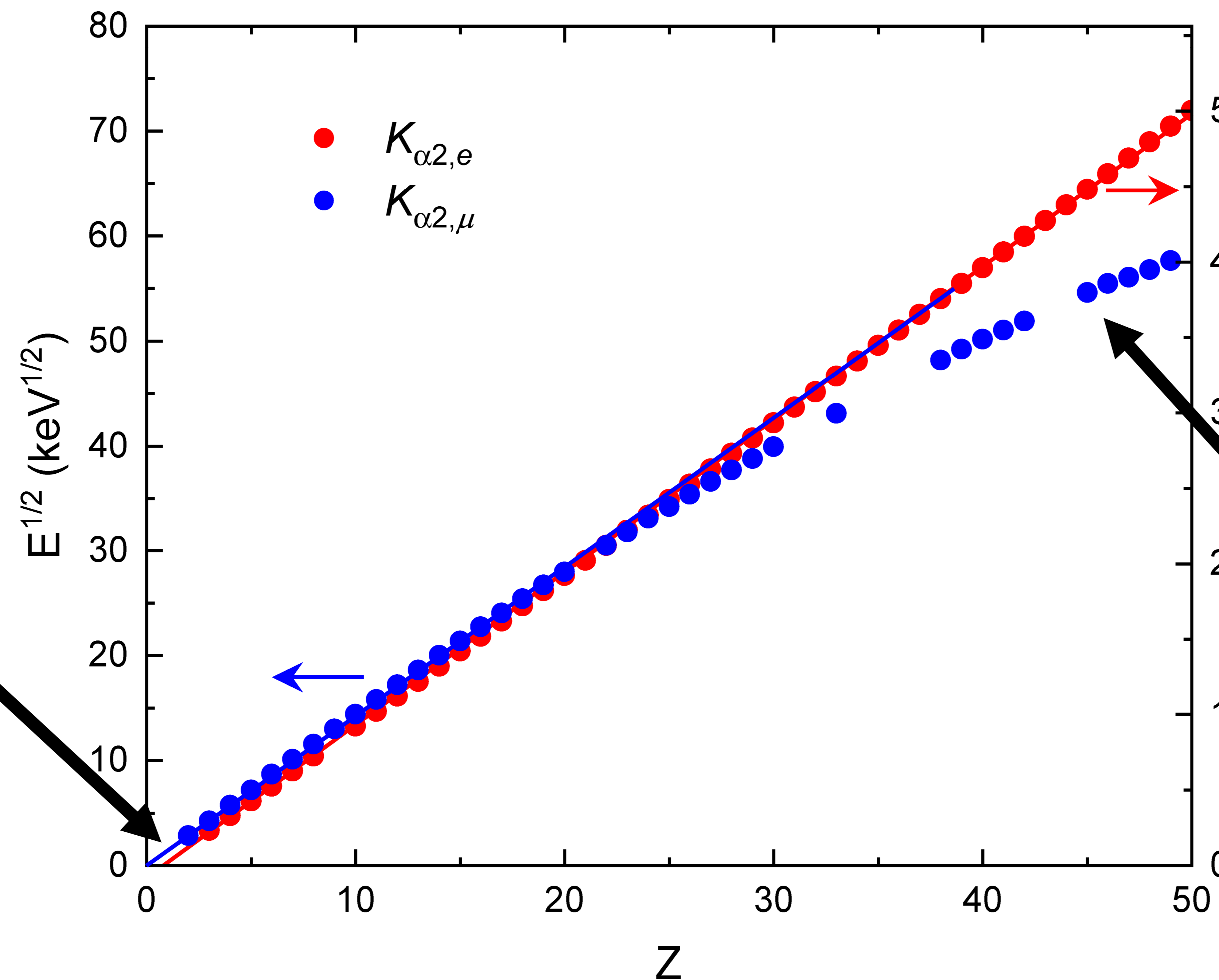
$$E_{i \rightarrow f, \mu} = \frac{\bar{M}_\mu}{M_e} R_y (Z - S_{scr, \mu})^2 \left(\frac{1}{n_f^2} - \frac{1}{n_i^2} \right)$$



Note:

$$S_{scr, \mu} = 0$$

$$S_{scr, e} = 1$$



?

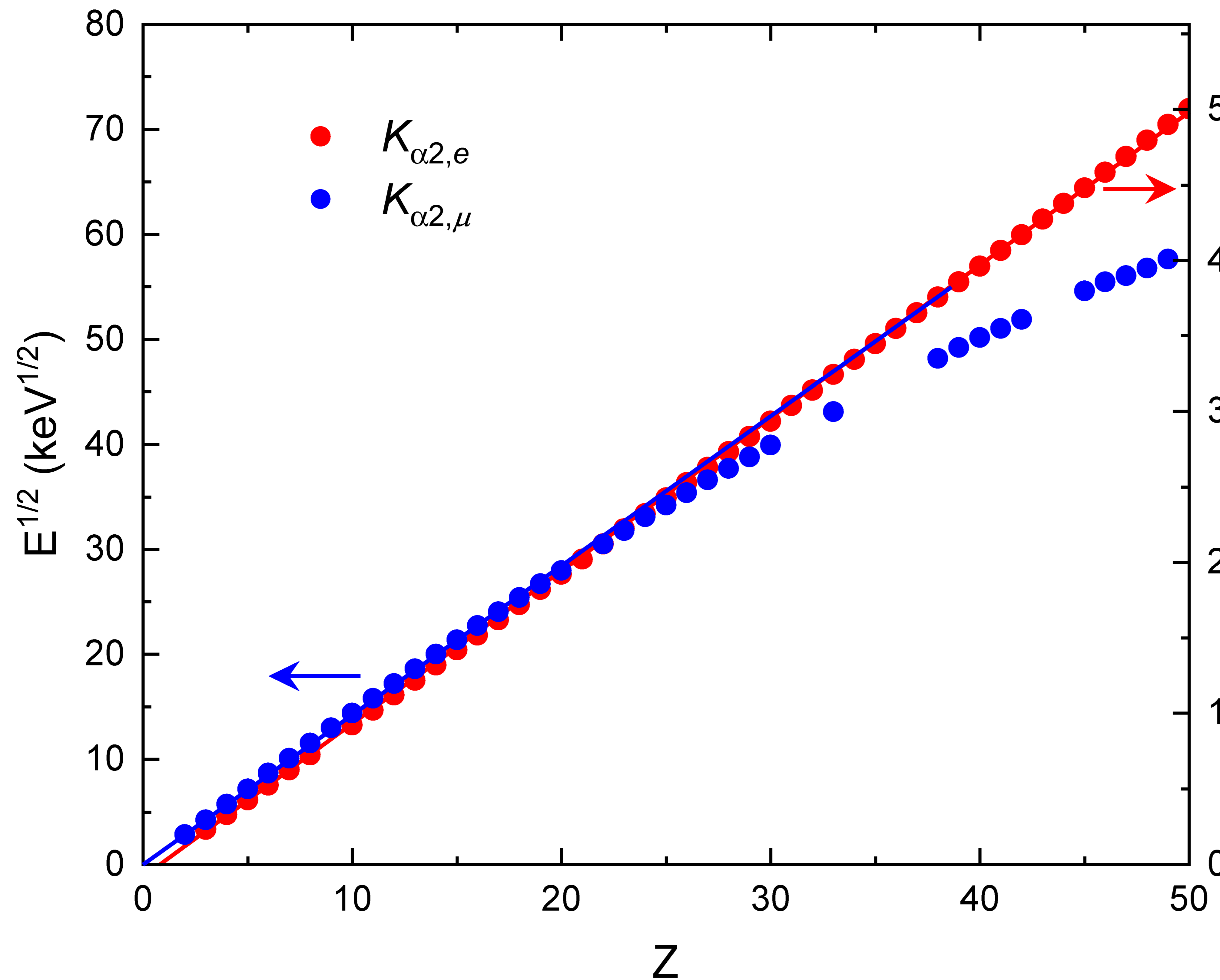
What about negative muons – X-ray energy

Energy of the X-rays: Size effects

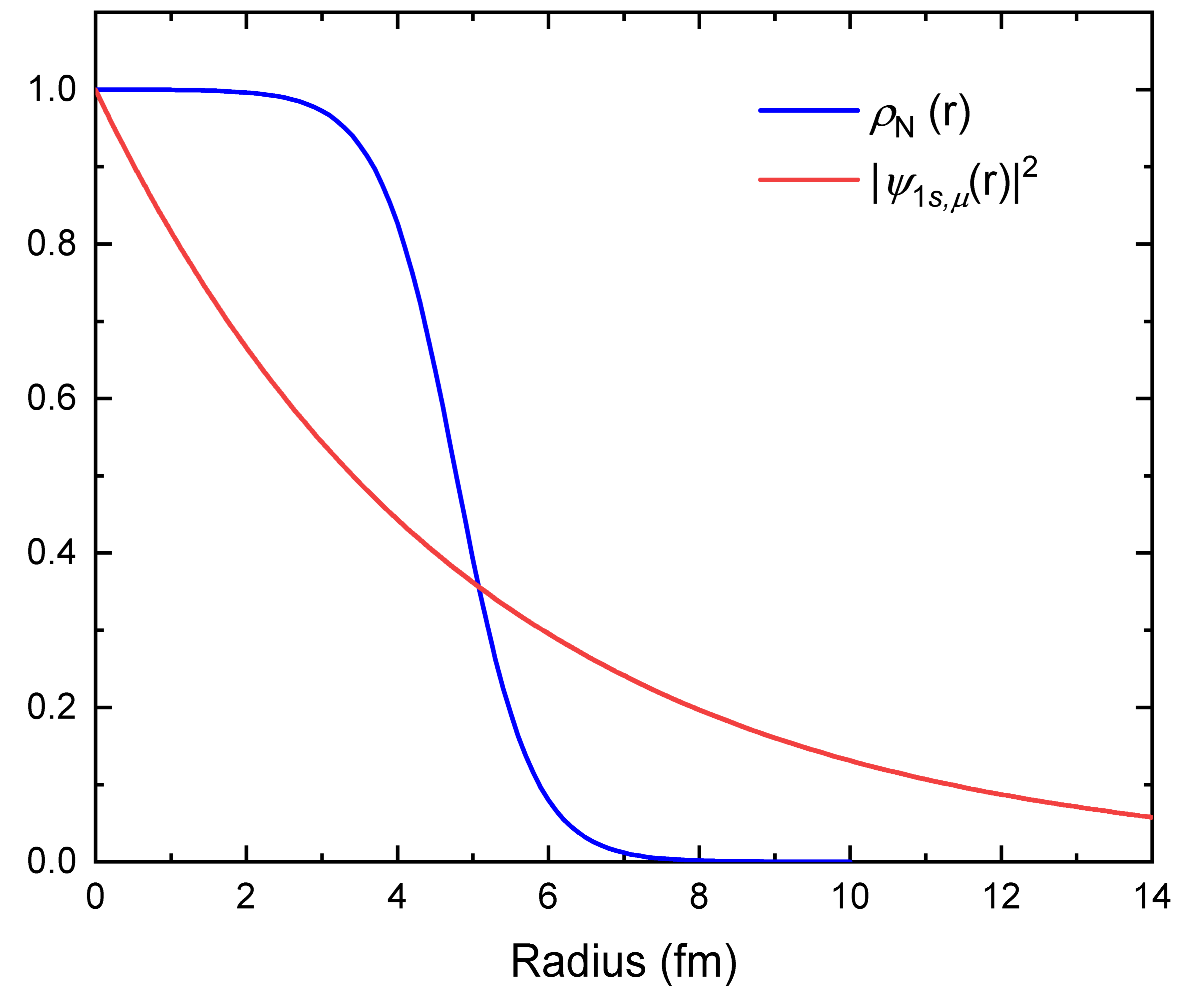
For large Z atoms, the muons is so close to the nucleus (radius proportional to $1/Z$) that the nucleus can no more be considered as point-like \rightarrow **size effects**

Can be used to determined the charge radius of nuclei.

This effect decreases for final levels in the d or f shell...



Example Iron ($Z = 26$)

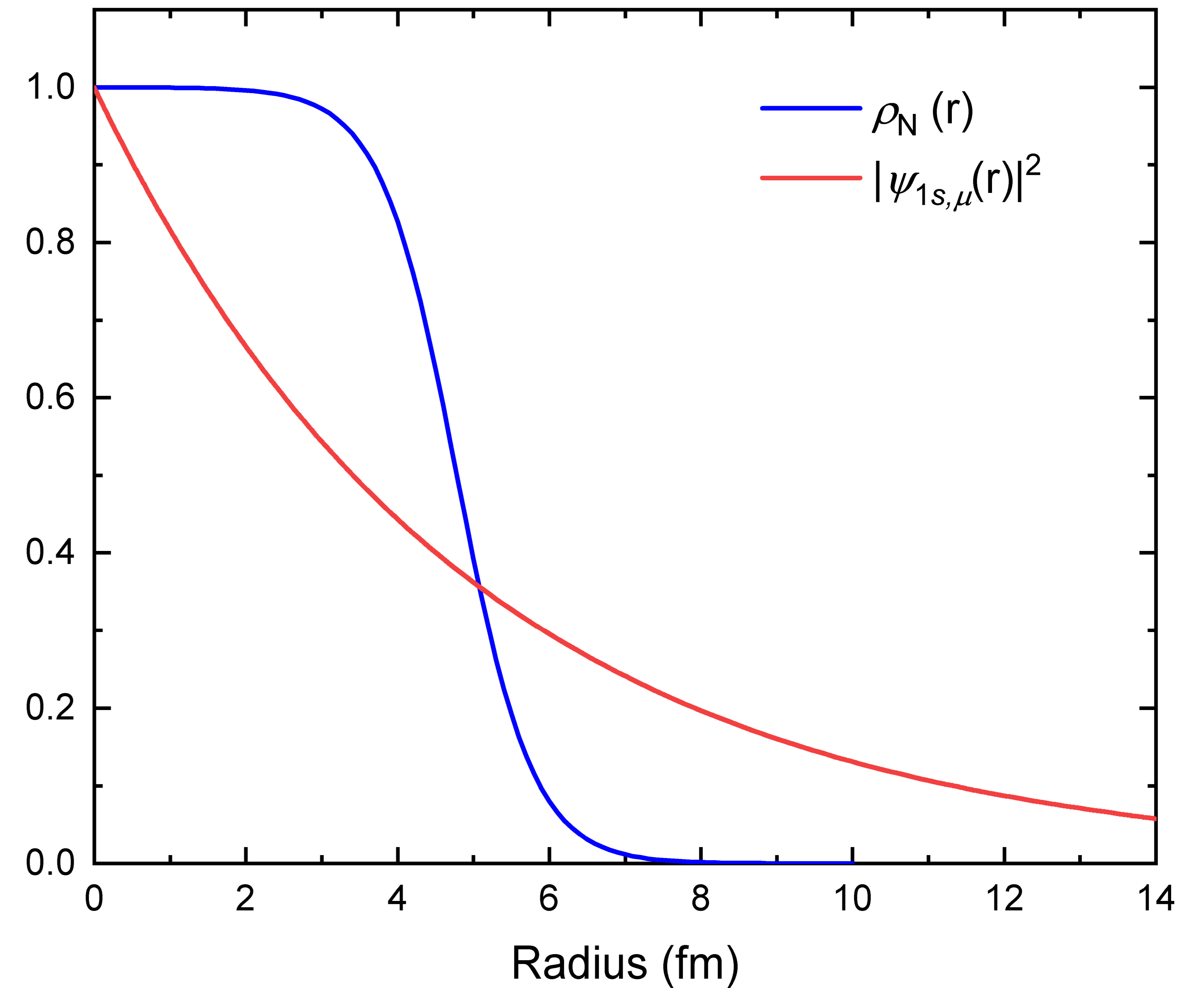
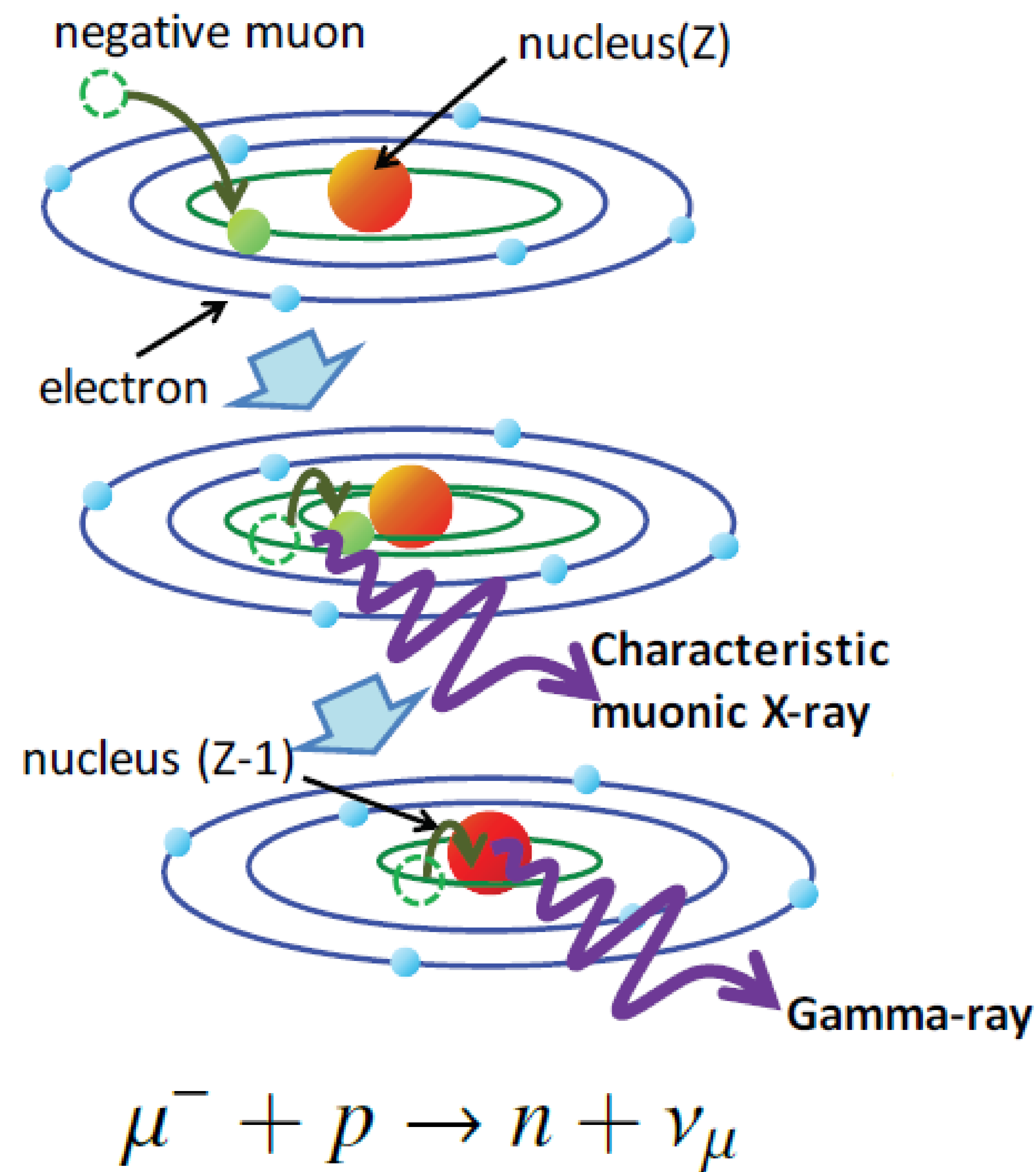


$$\rho_N(r) = \frac{\rho_0}{1 + \exp\left(\frac{r-R}{a}\right)} \quad \begin{array}{l} a \simeq 0.5 \text{ fm} \\ R = r_0 A^{1/3} \end{array}$$

$$|\psi_{1s, \mu}(r)|^2 = \frac{1}{\pi a_{0, \mu}^3} \exp\left(-\frac{2r}{a_{0, \mu}}\right)$$

What about negative muons – Nucleus capture

Second Capture:

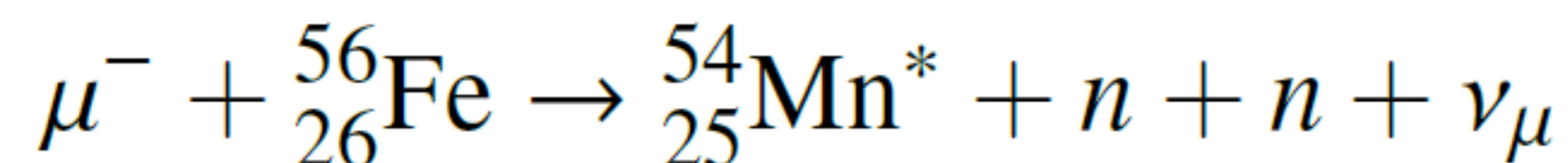
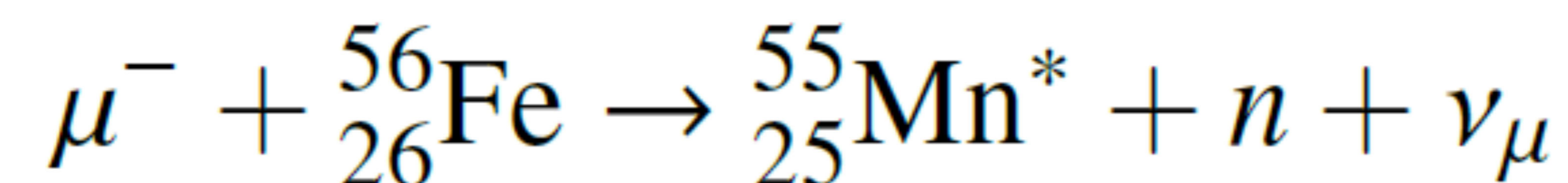


Example of Iron:

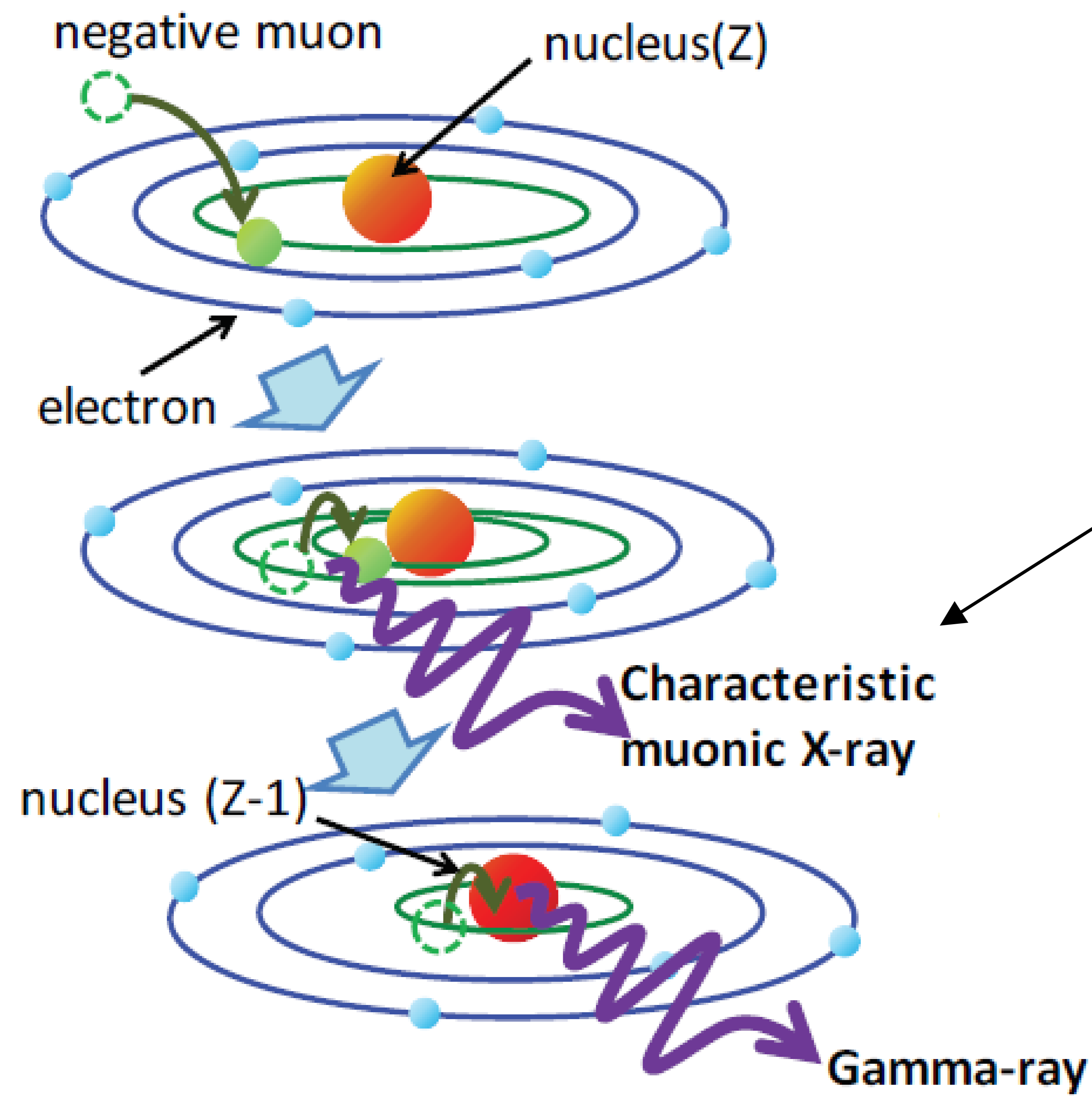


The excited Manganese nucleus will emit Gamma-rays.

Or one or more neutrons (and later Gamma-rays) can be emitted:



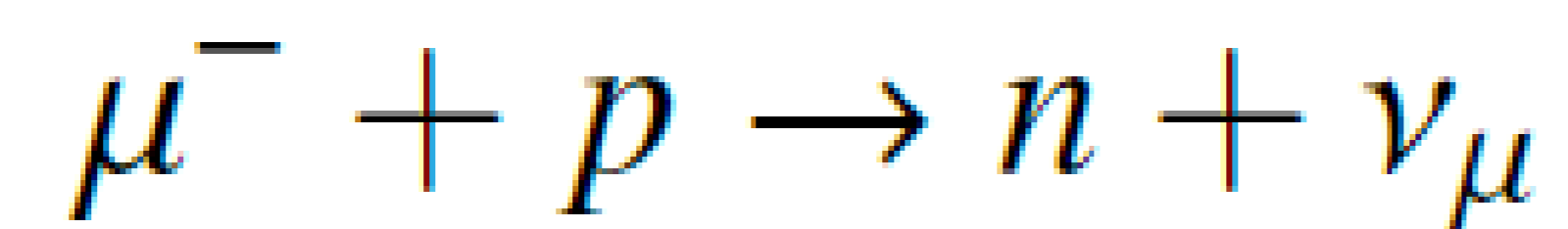
What about negative muons – Nucleus capture



«Prompt» X-rays
(i.e. few picosec. after muon arrival)

«Prompt» and «delayed»
Gamma rays characterized by

$$\frac{1}{\tau_{\mu, \text{Nucl. capt}}(Z)}$$



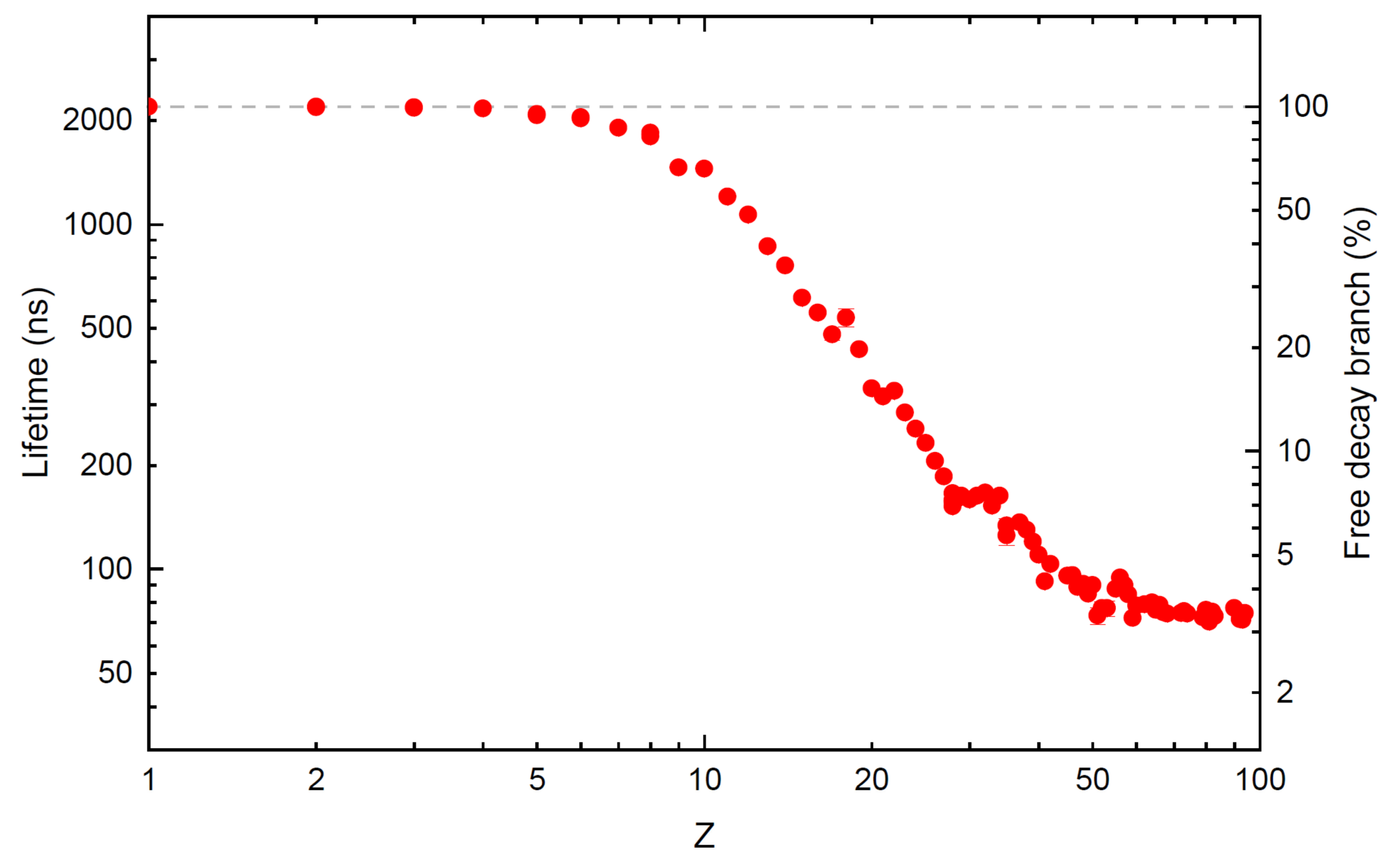
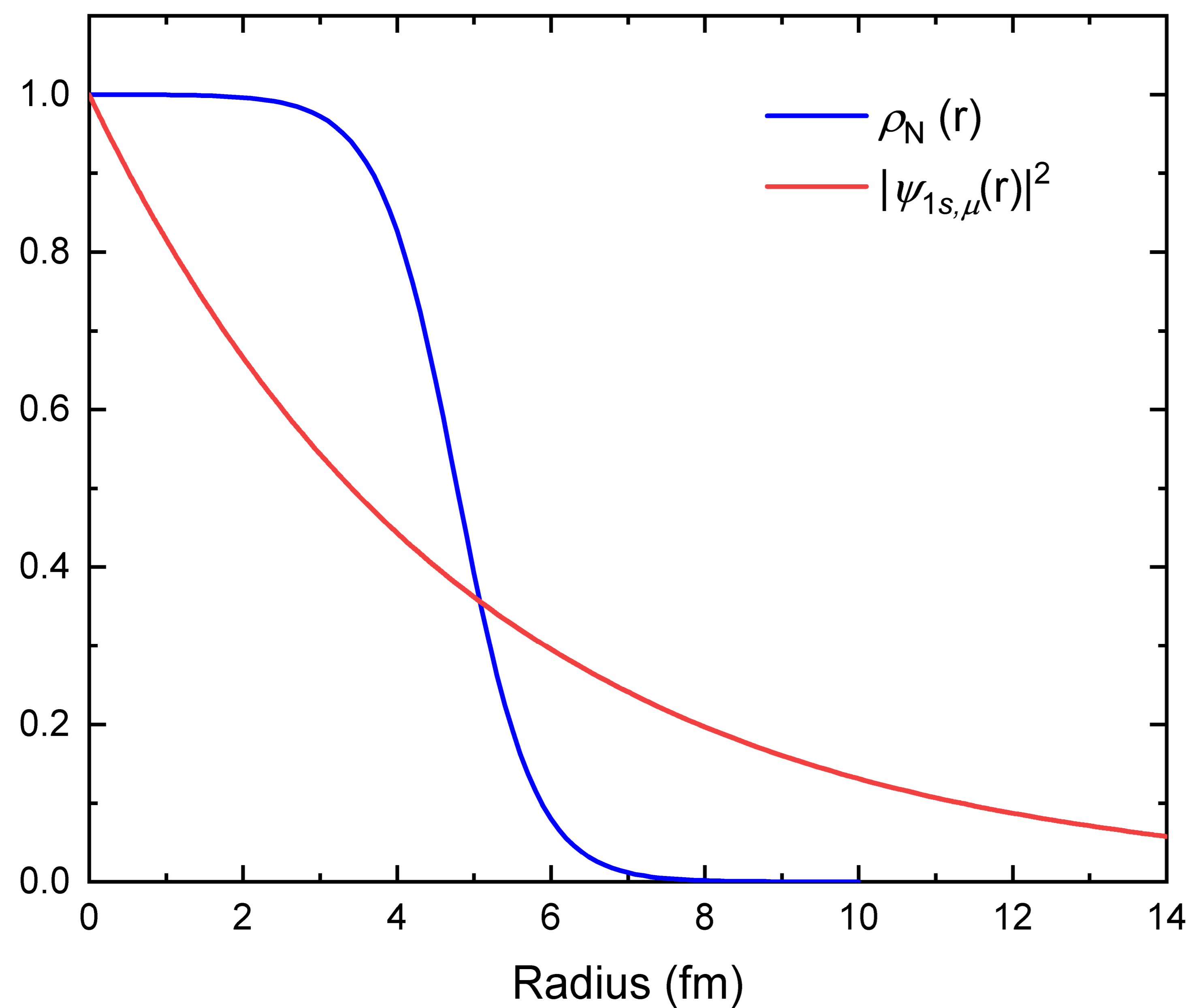
What about negative muons – Lifetime

Free negative muon:

$$\tau_{\mu, \text{free}} = 2.1969811(22) \times 10^{-6} \text{ sec.}$$

For captured muons:

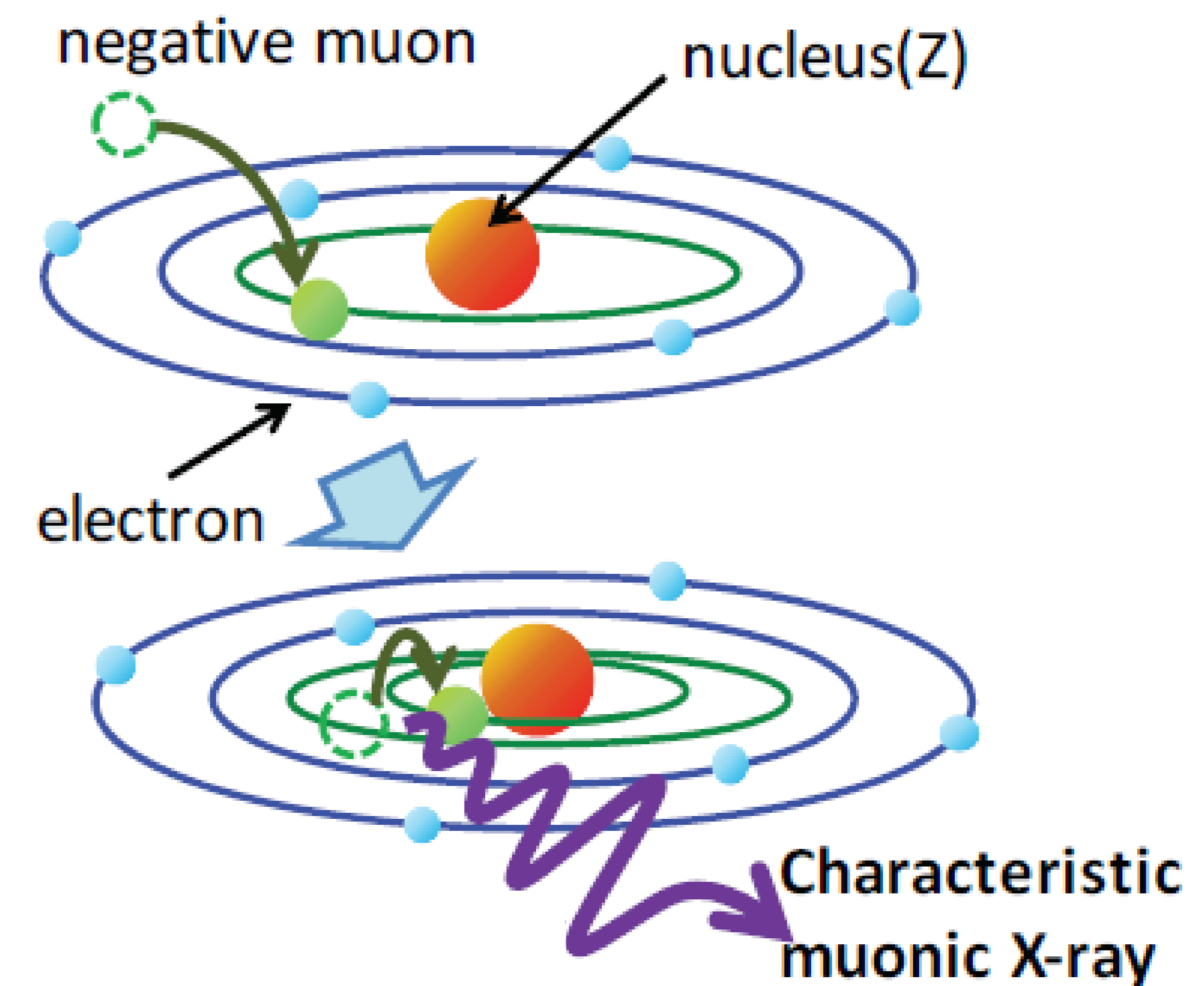
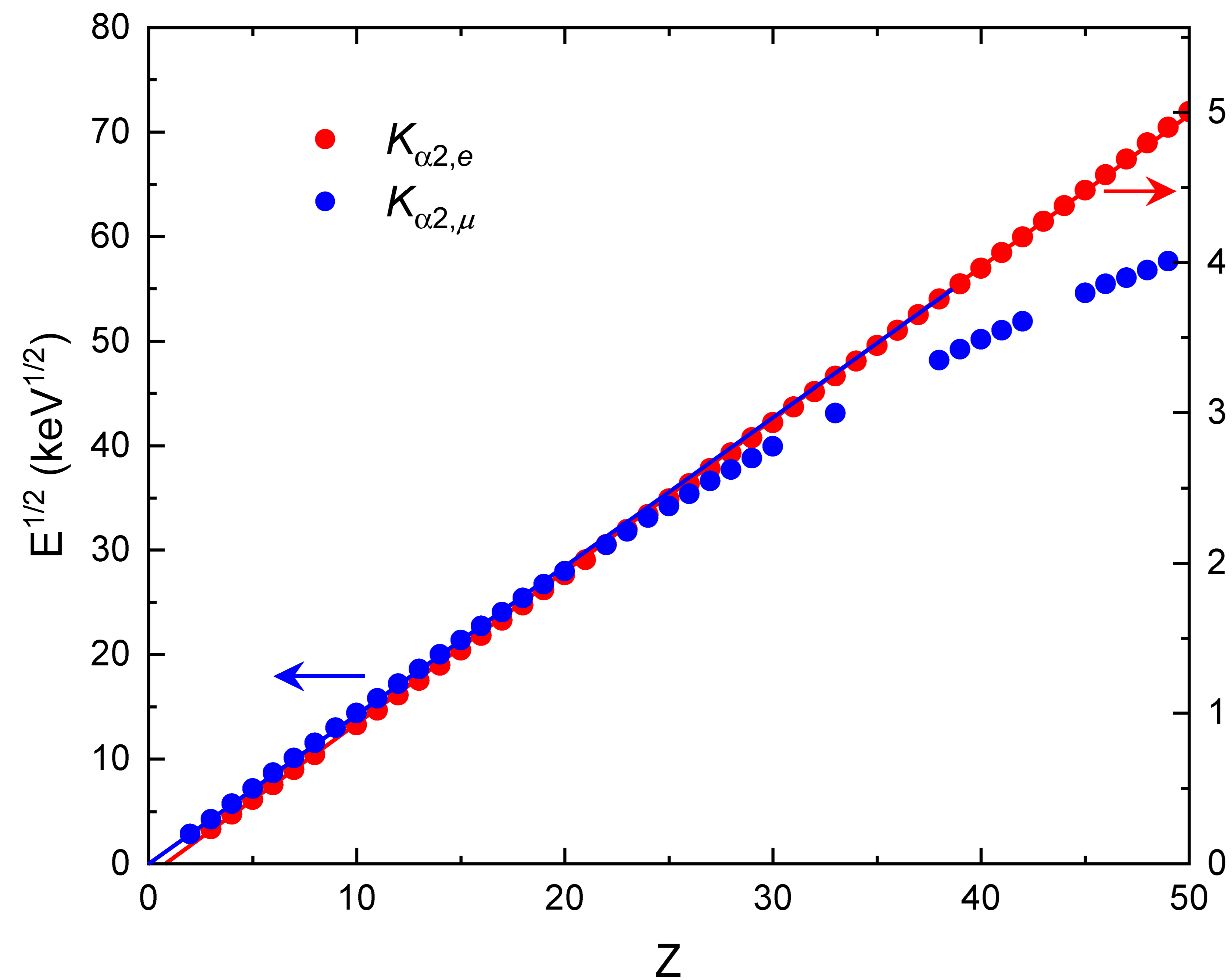
$$\frac{1}{\tau_{\mu}(Z)} = \frac{1}{\tau_{\mu, \text{free}}} + \frac{1}{\tau_{\mu, \text{Nucl. capt}}(Z)}$$



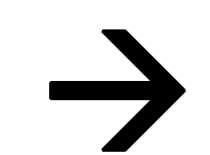
→ Different lifetimes depending on the atom capturing the muon

→ Additional difficulty to perform μ SR with negative muons

What about negative muons – Elemental Analysis



- Possibility to implant the muons at a controlled depth (by changing the muon momentum)
- Characteristic muonic X-rays high energy and can escape from the sample



μ^- : Non-destructive, depth-sensitive probes of elemental composition

APPLICATION OF MUONIC X-RAY TECHNIQUES TO THE ELEMENTAL ANALYSIS OF ARCHEOLOGICAL OBJECTS

E. KÖHLER, R. BERGMANN, H. DANIEL, P. EHRHART and F.J. HARTMANN

Physics Department, Technische Universität München, D 8046 Garching, Germany

Received 27 February 1981

The use of muonic X-rays as a tool for elemental analysis is described. Bulk analyses of modern and archeological fired clay samples are presented. Comparison with chemical and neutron activation analyses supplies standards for future measurements. Scanning techniques are also described.

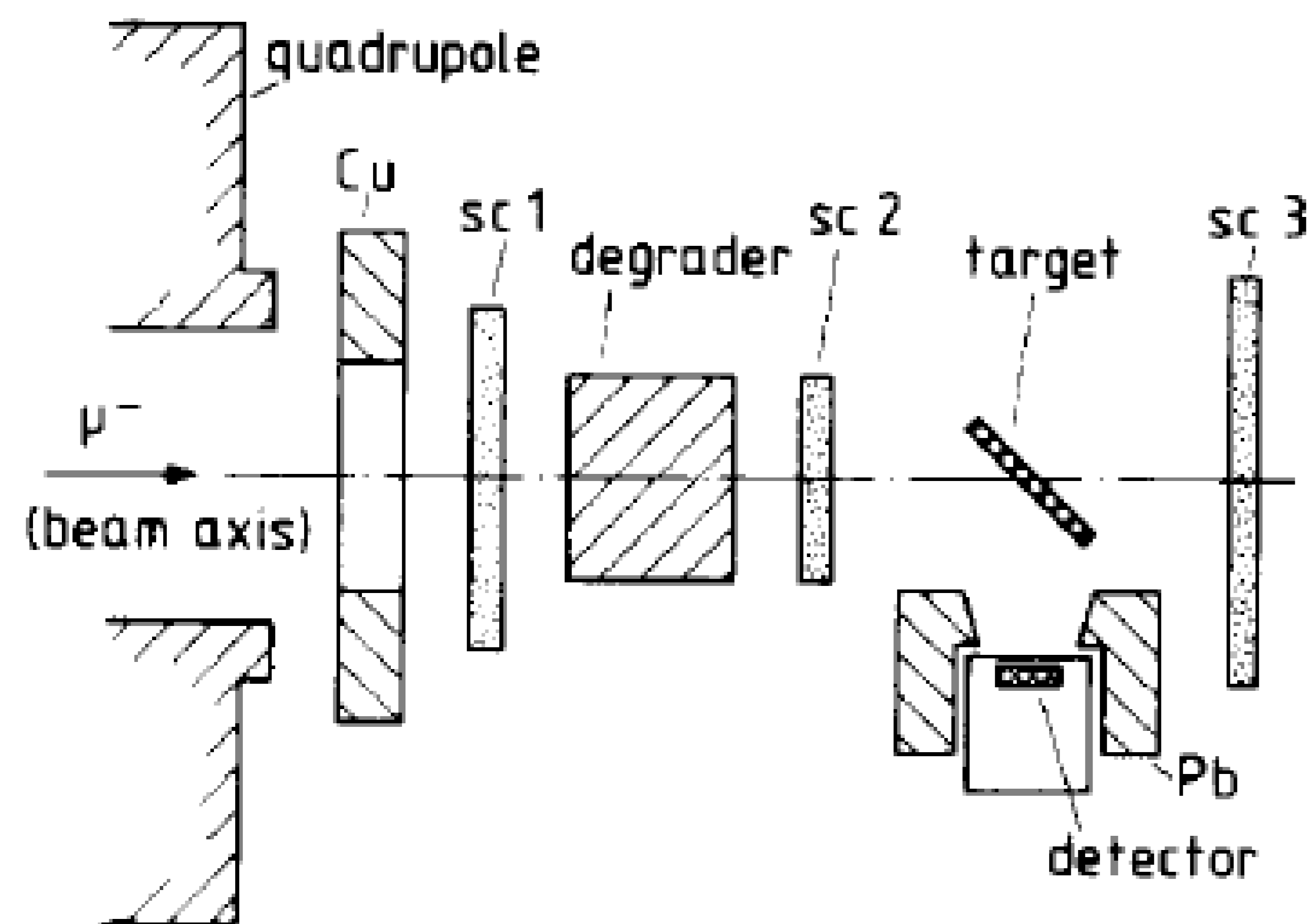


Fig. 1. Experimental setup: Cu—copper shielding, detector—Ge(Li) detector, Pb—lead shielding for detector, sc1, sc2, sc3—scintillation counters forming a telescope.

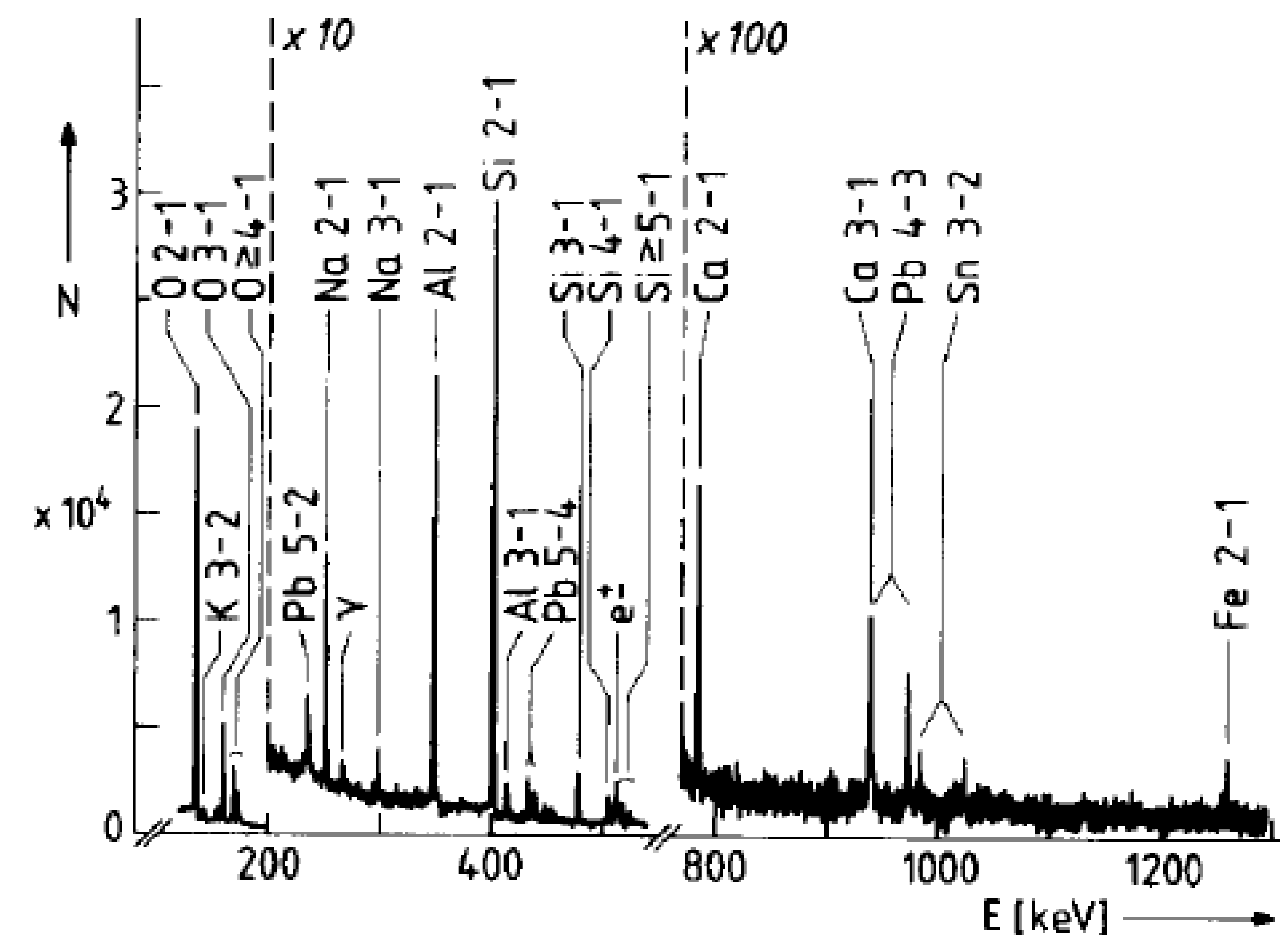
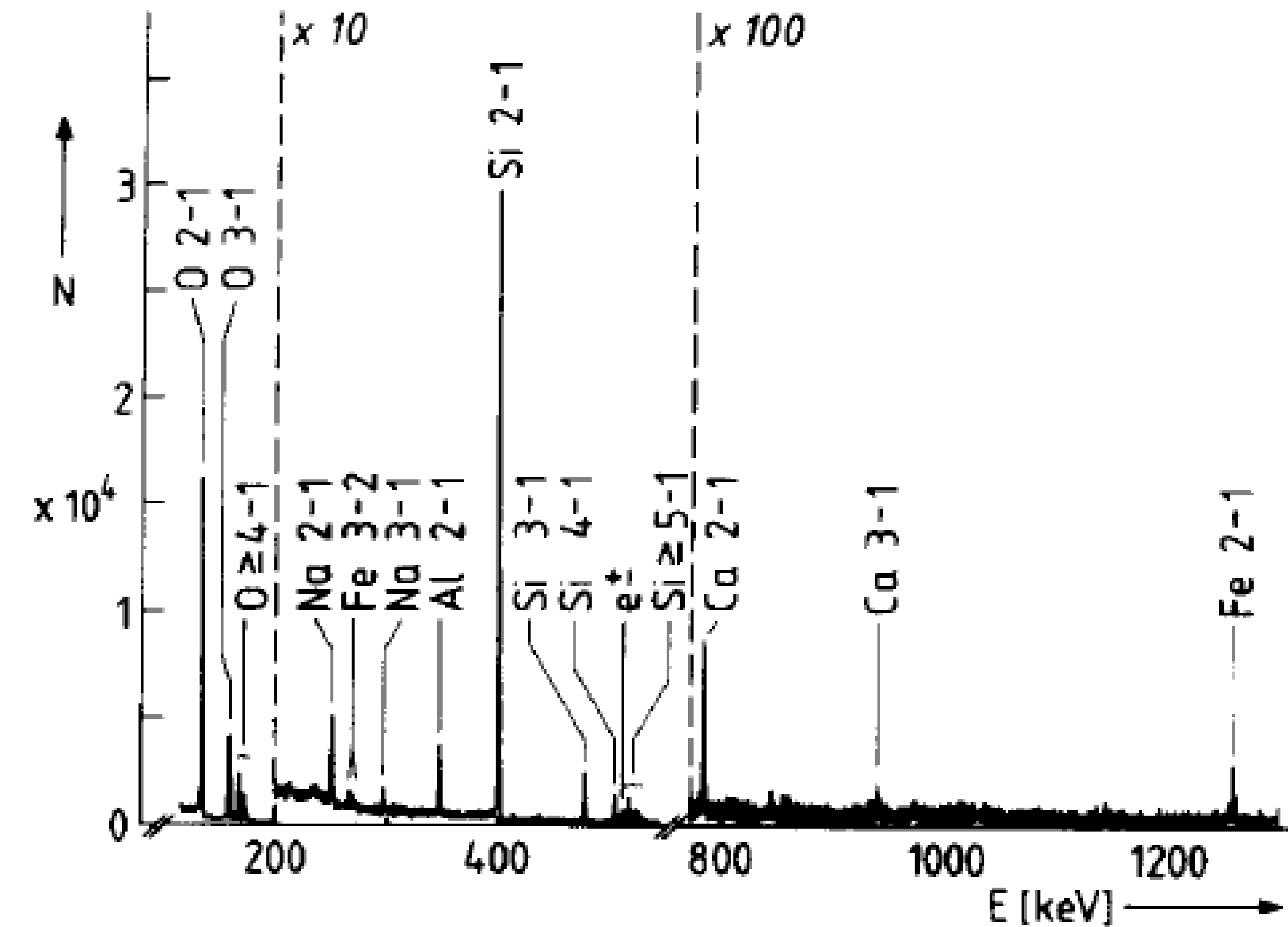
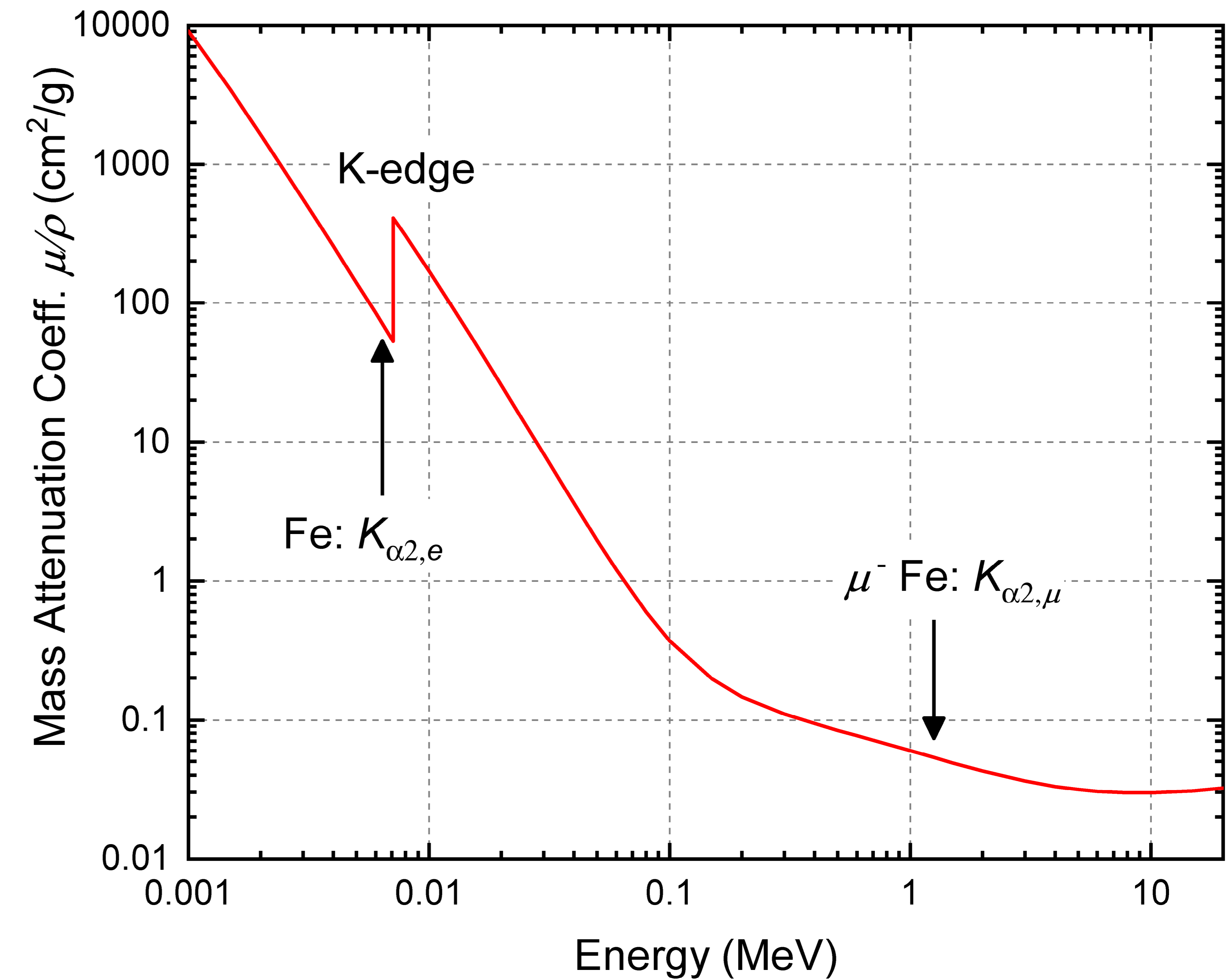
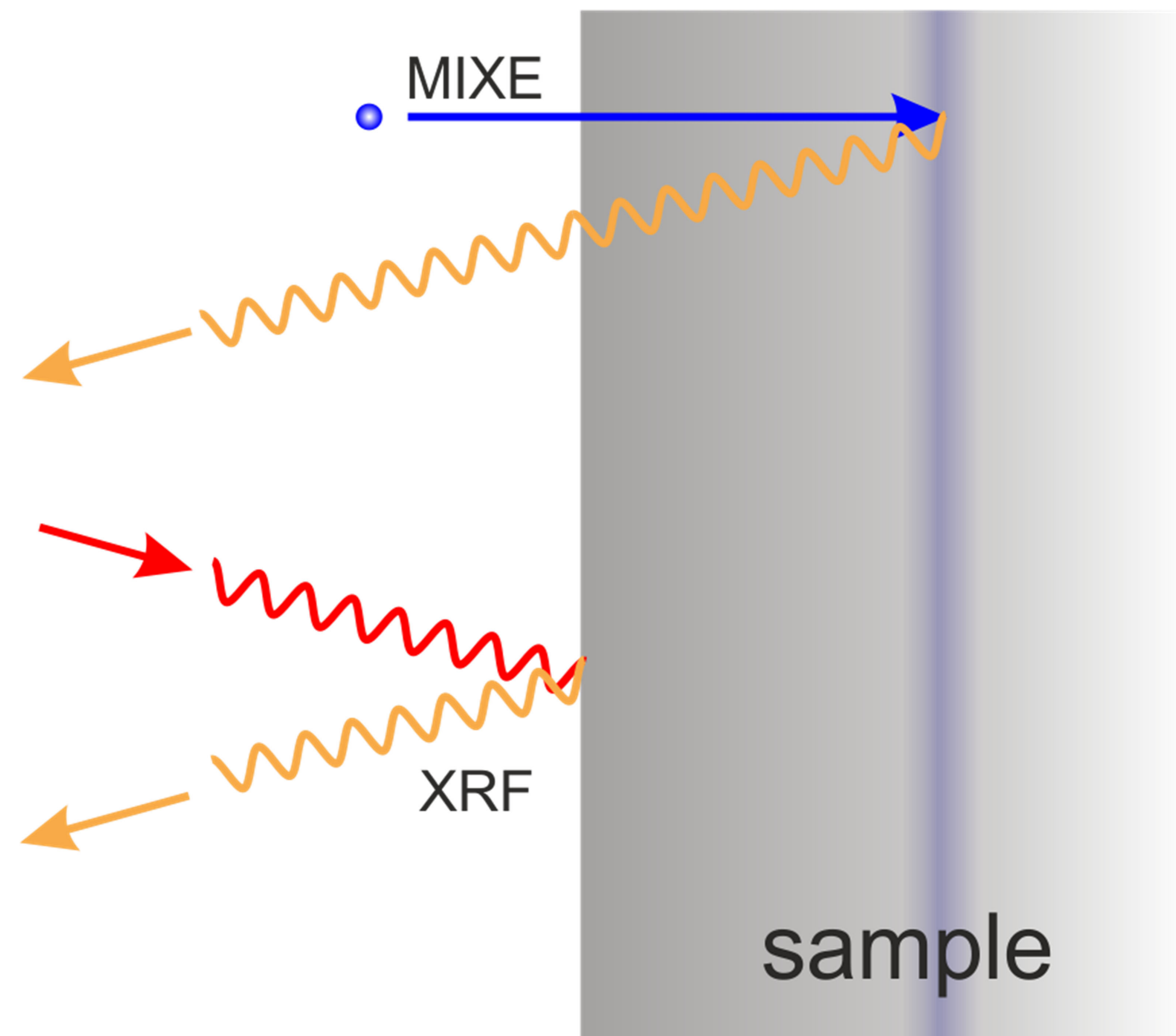


Fig. 2. Spectra of Islamic tile. Above: base material (fired clay); below: glaze.



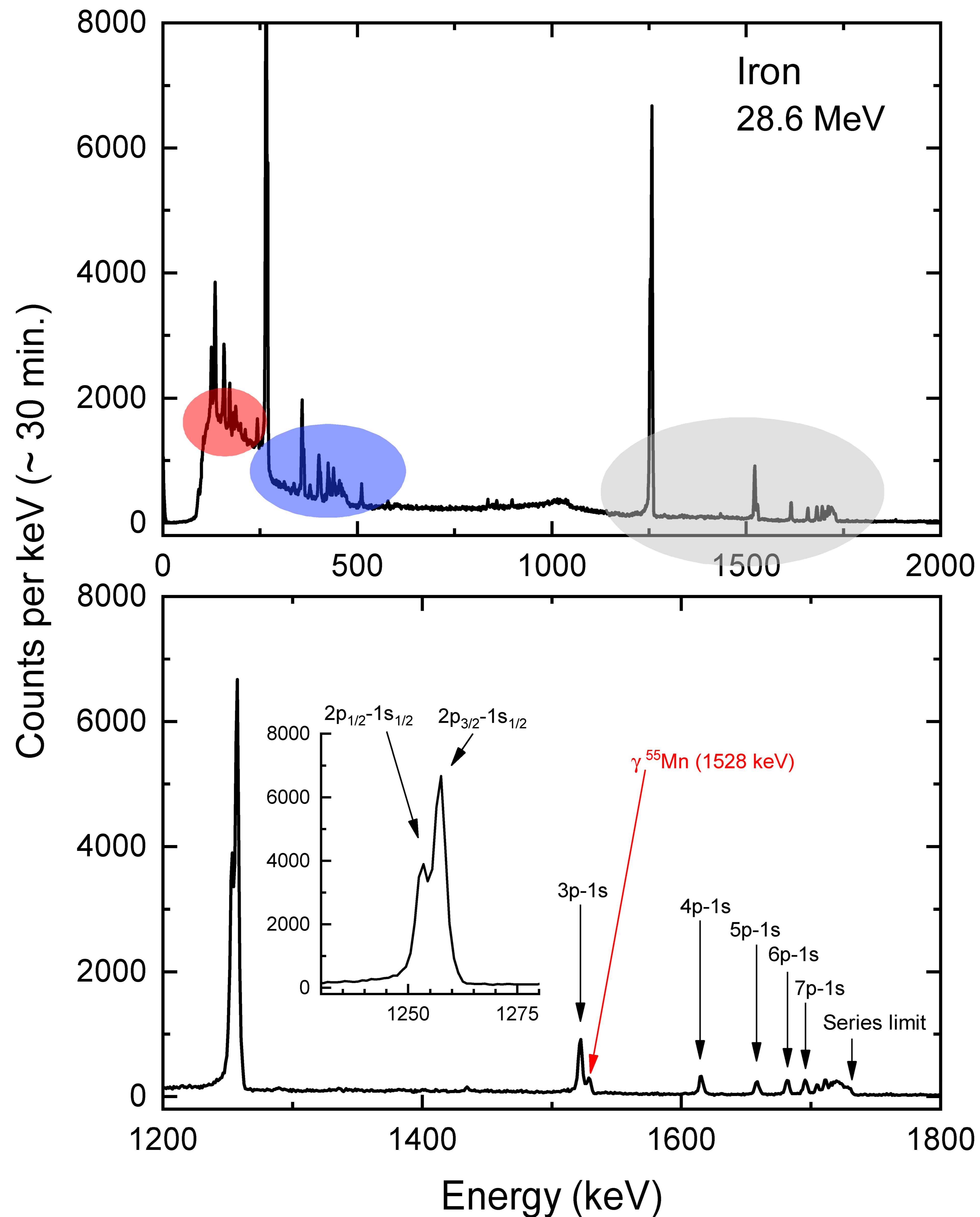
Attenuation of X-rays into a material:

$$I(\ell) = I_0 \exp[-(\mu/\rho)\rho\ell]$$

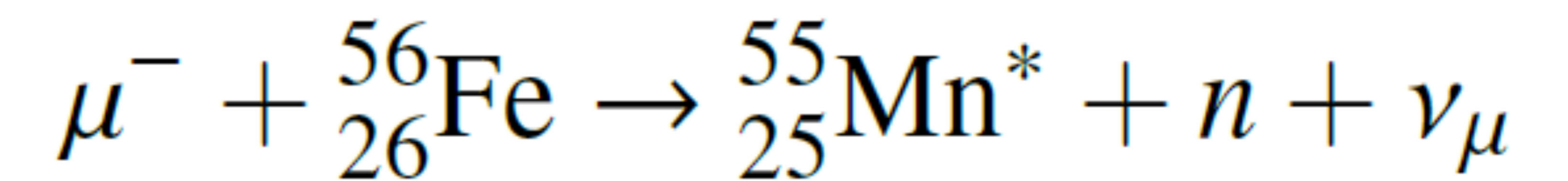
Example of iron

- **Electronic X-ray:** $K_{\alpha 2} = 6.39 \text{ keV}$ → attenuation by a factor e after **25 μm**
- **Muonic X-ray:** $K_{\alpha 2} = 1255 \text{ keV}$ → attenuation by a factor e after **2.5 cm**

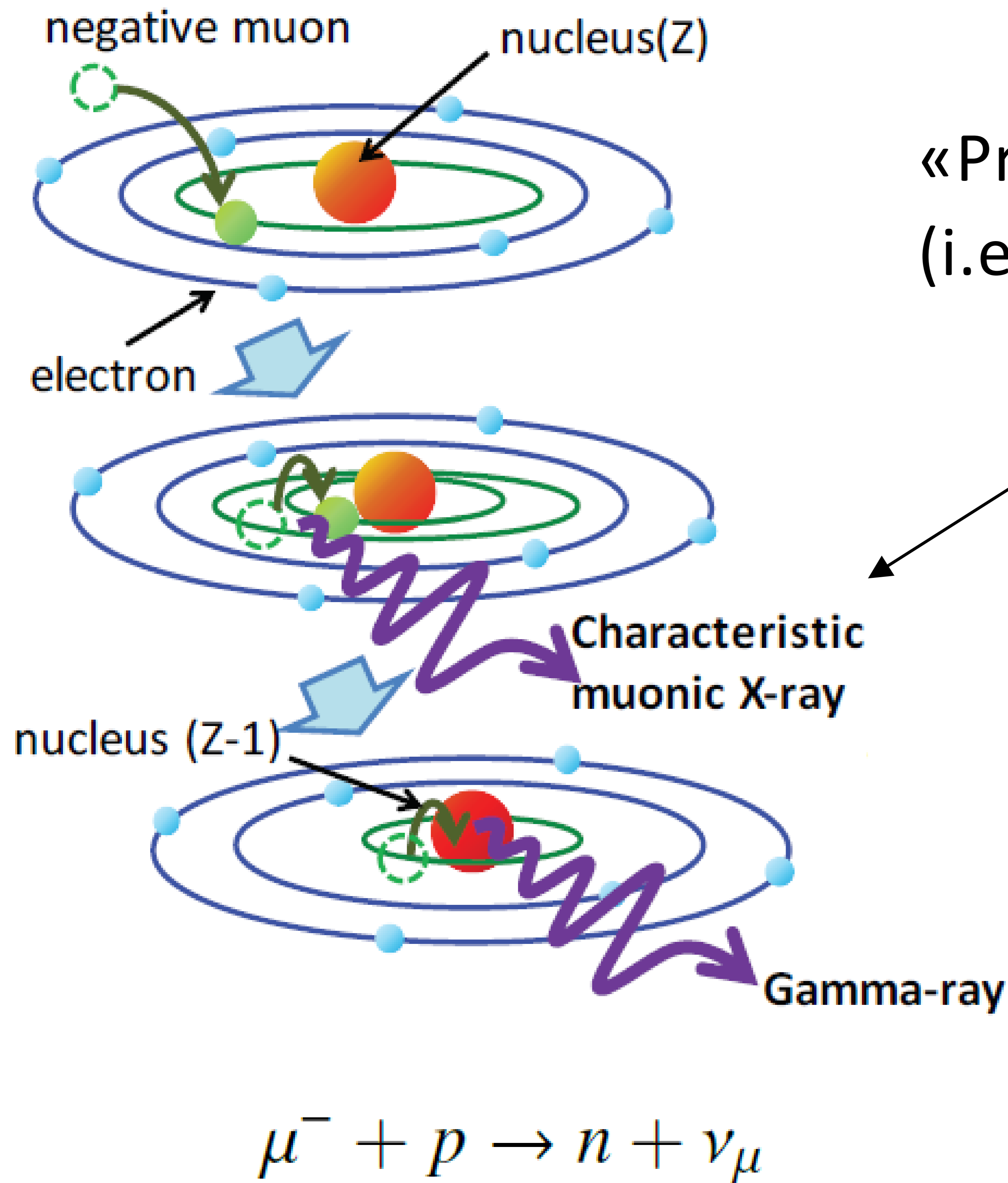
MIXE – Example



- Muon momentum 28.6 MeV/c.
- Penetration about 200 μm .
- Signals from muonic series
 - Lyman ($n_f = 1$, grey region),
 - Balmer ($n_f = 2$, blue region) and
 - Paschen ($n_f = 3$, red region)
- Data were recorded with one HPGe detector for about 30 minutes.
- Note the signal at 1528 keV



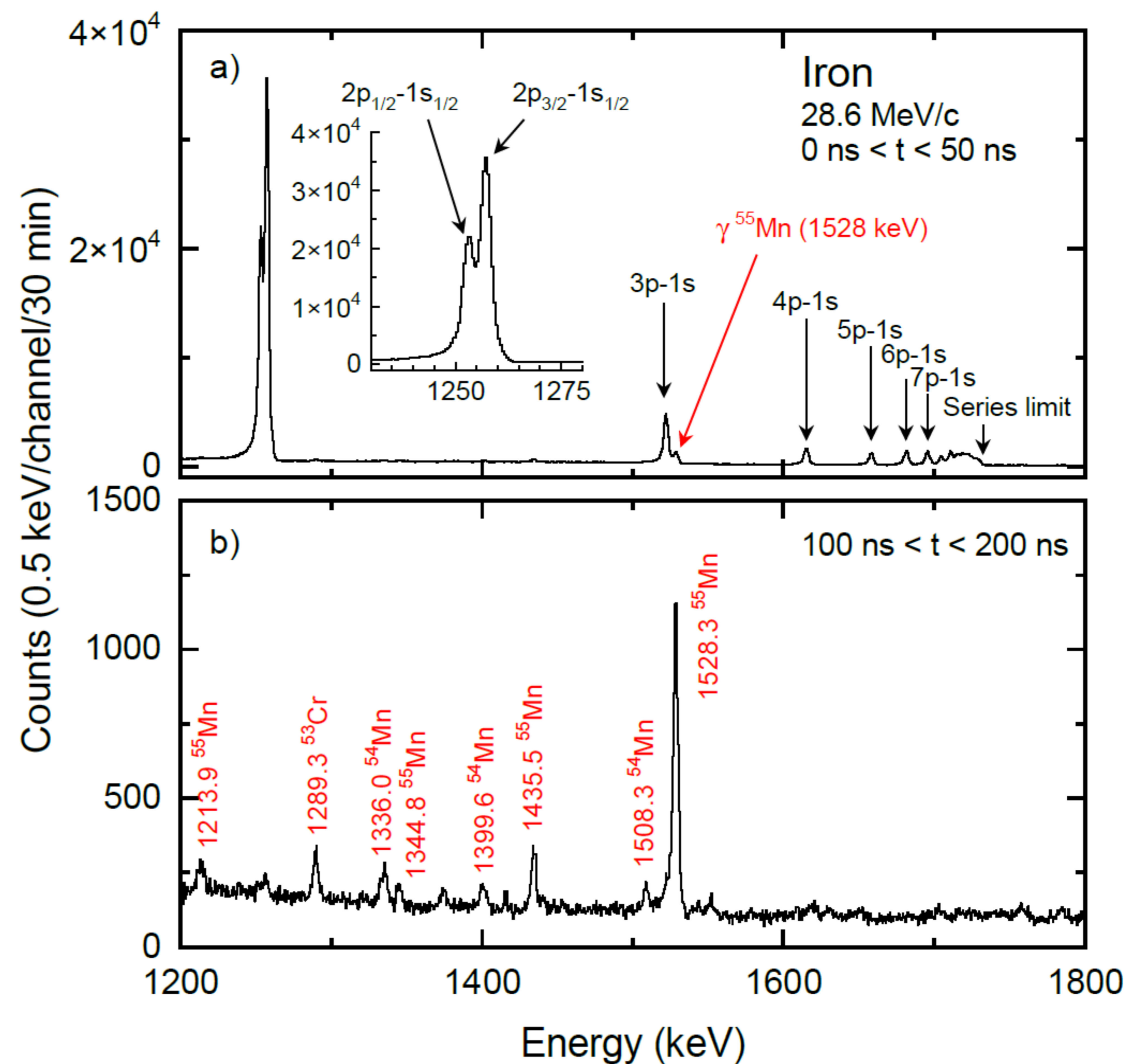
MIXE – Differentiate X-rays from Gamma-rays



«Prompt» X-rays
(i.e. few picosec. after muon arrival)

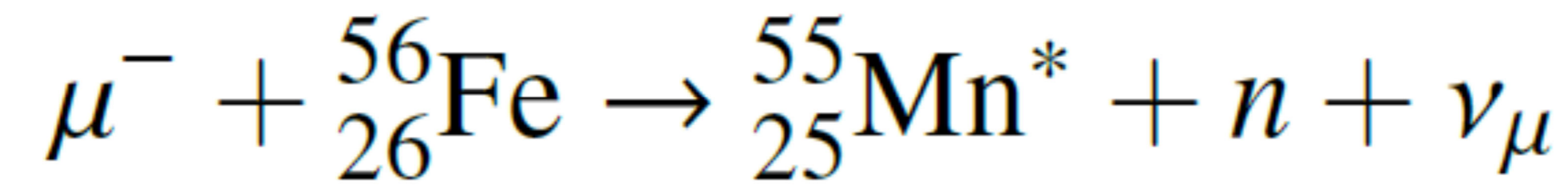
«Prompt» and «delayed»
Gamma rays characterized by

$$\frac{1}{\tau_\mu, \text{Nucl. capt}(Z)}$$

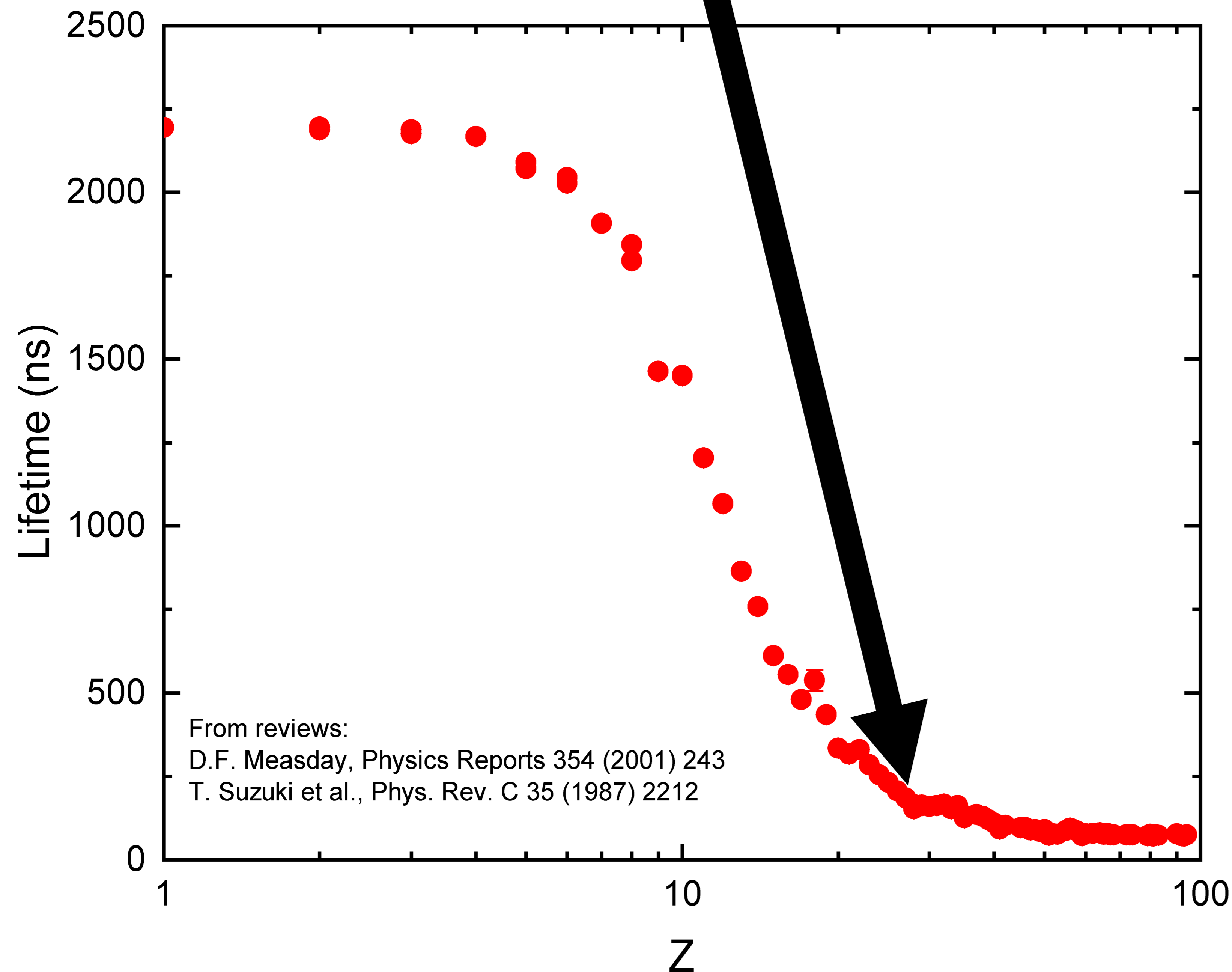
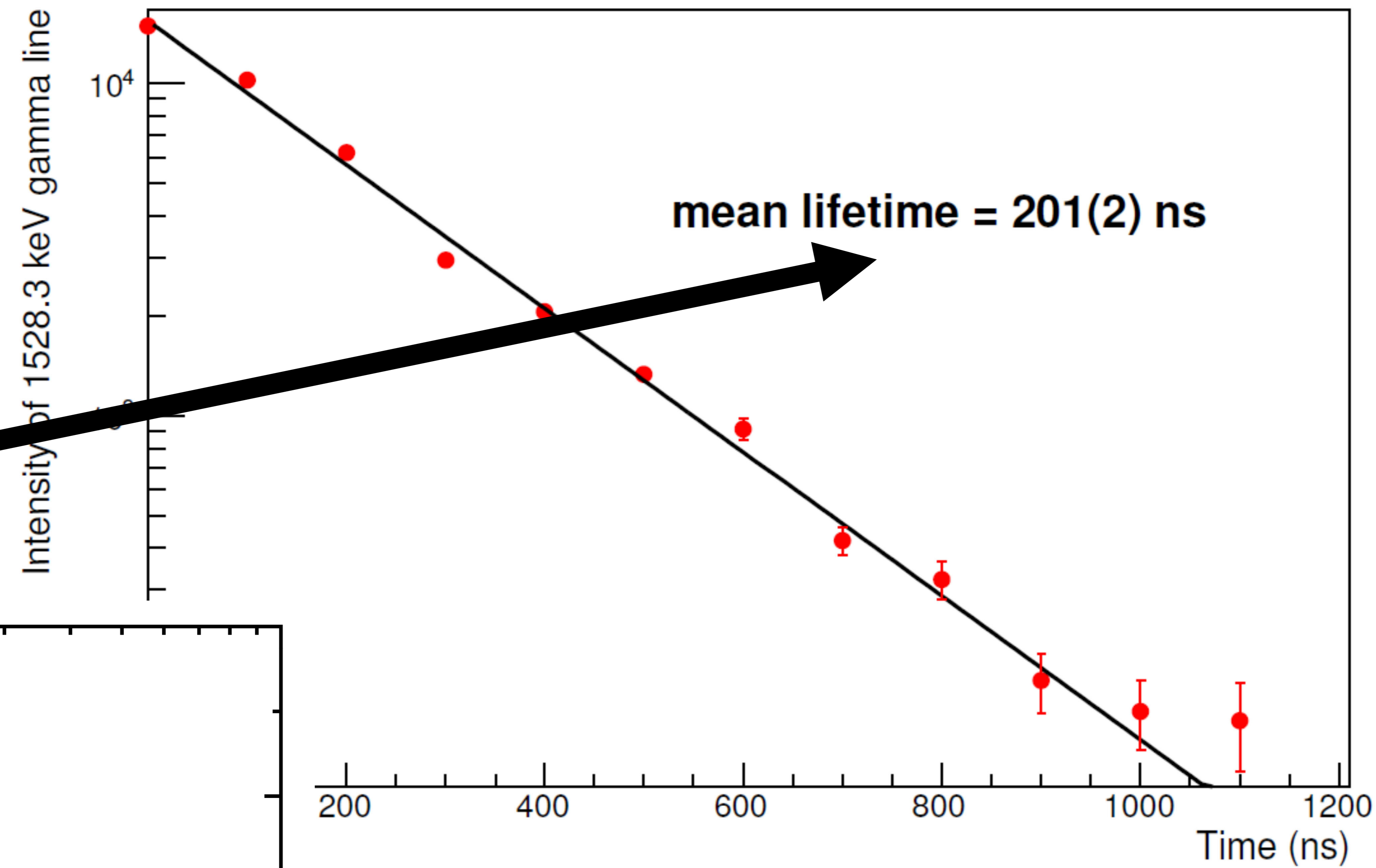


MIXE – Gamma rays

Example:



Emission of gamma-rays → one gamma-ray with energy 1528.3 keV



→ For «heavy» elements, by looking at the gamma-rays, we can also identify the primary atom, which has captured the muon

MIXE -- setup

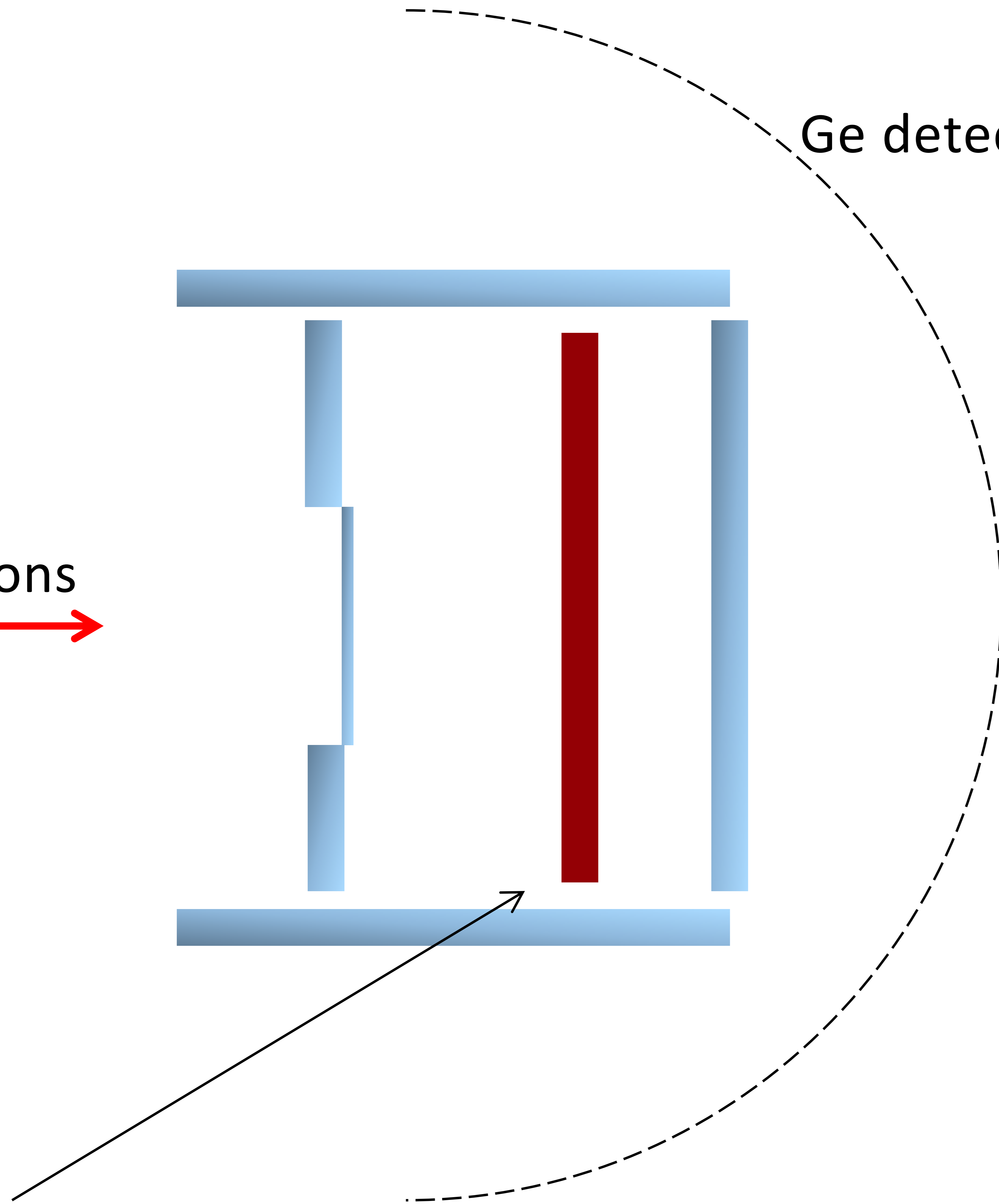


Negative muons

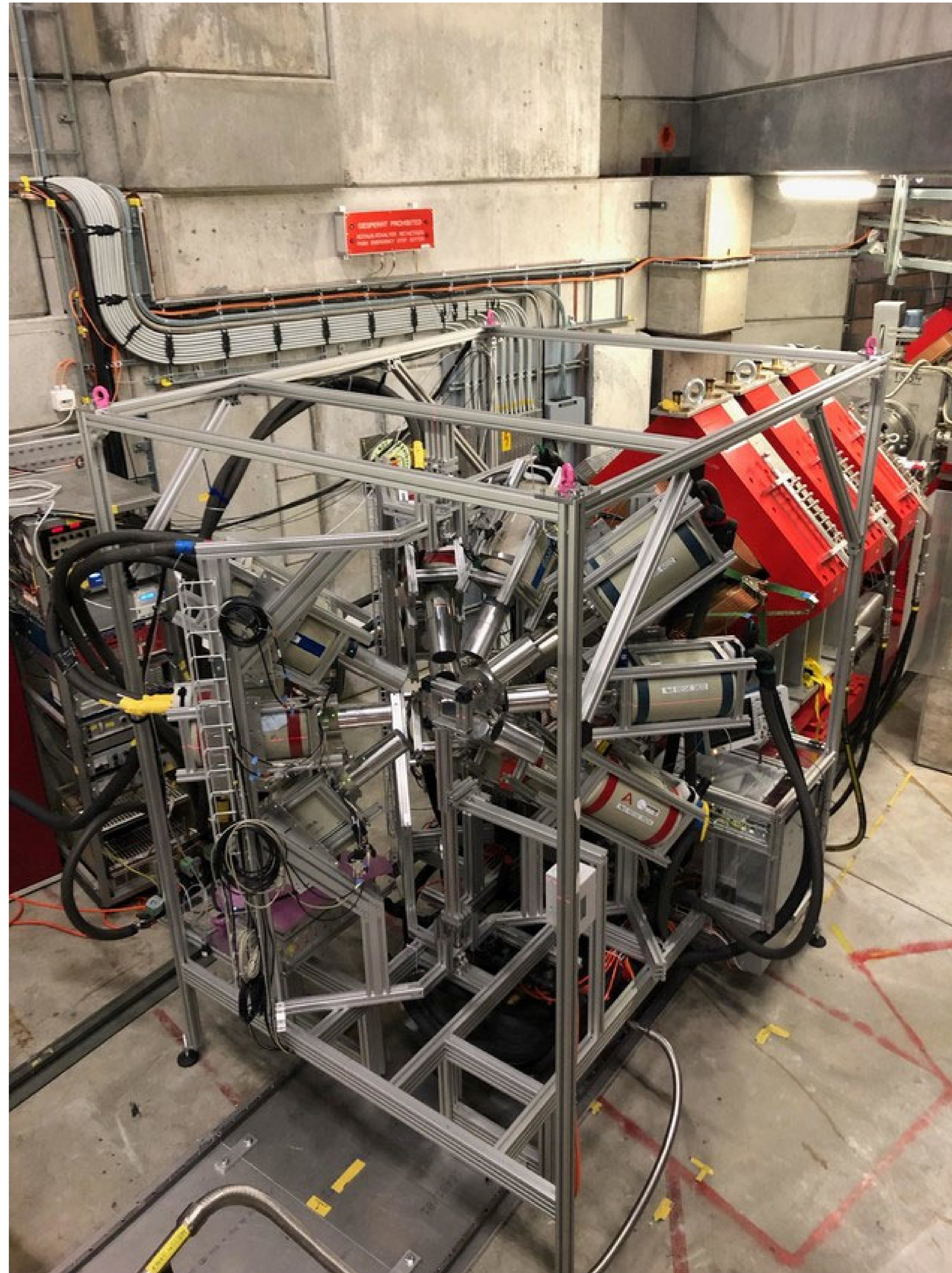


Ge detectors

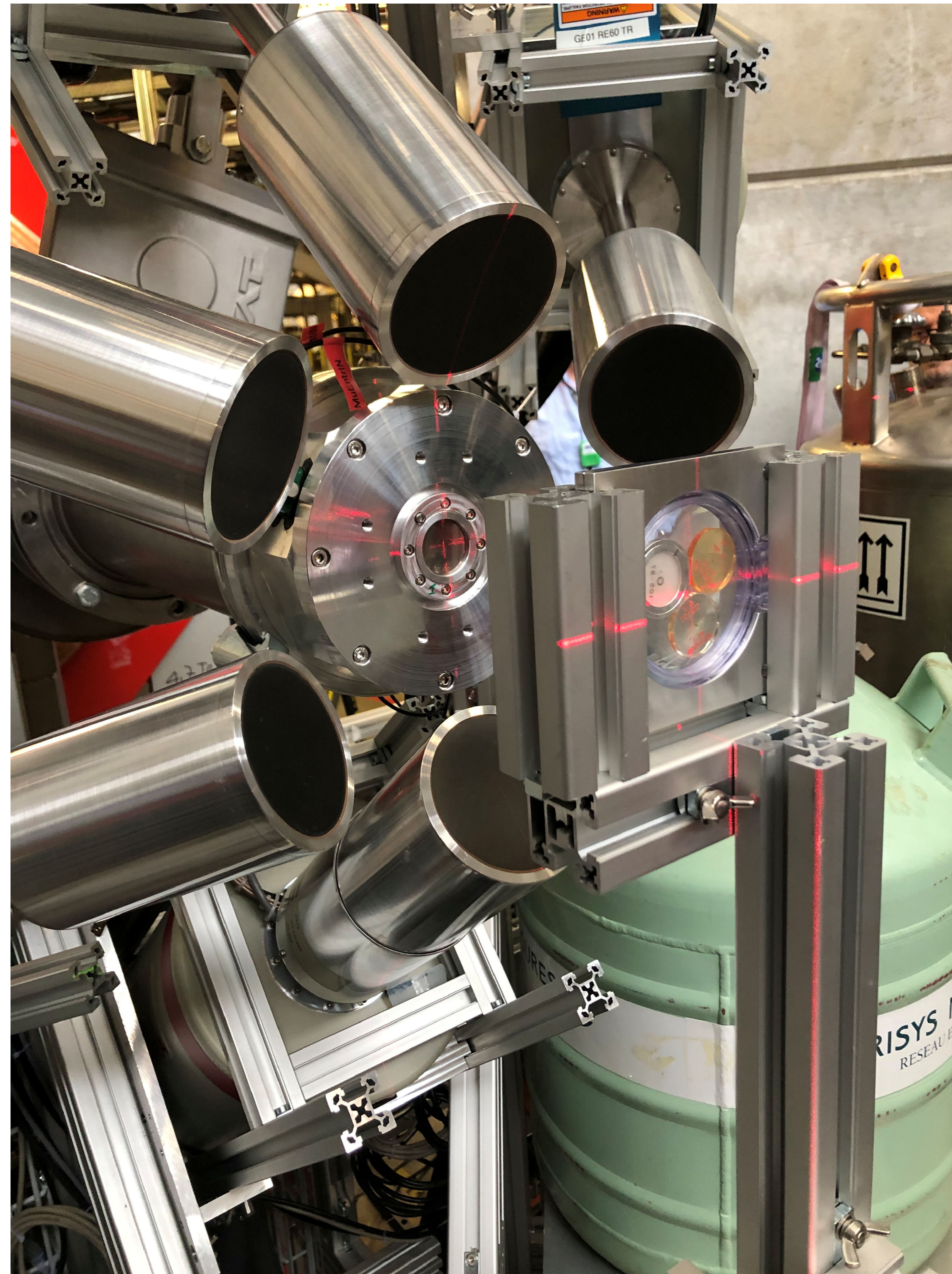
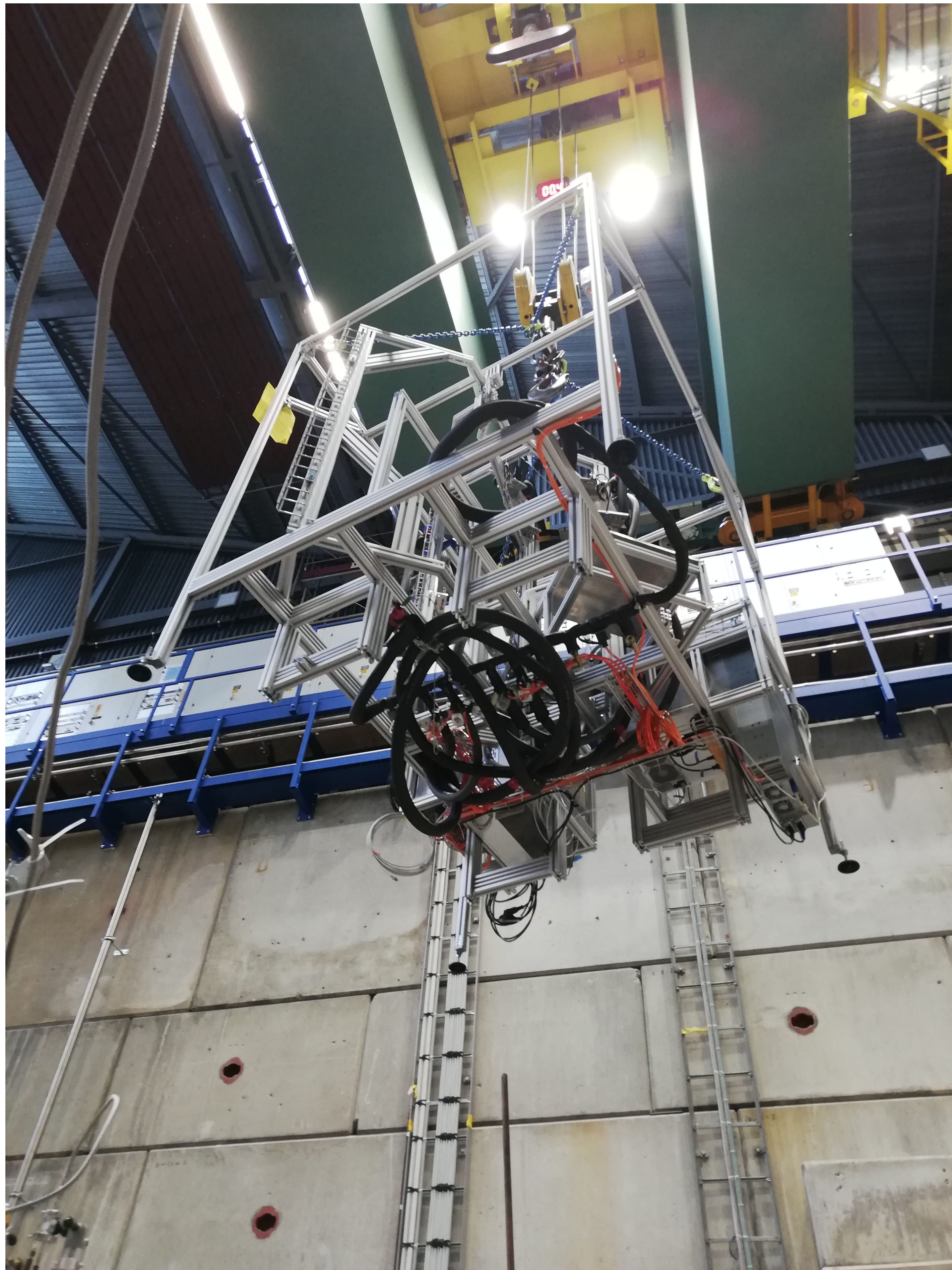
sample

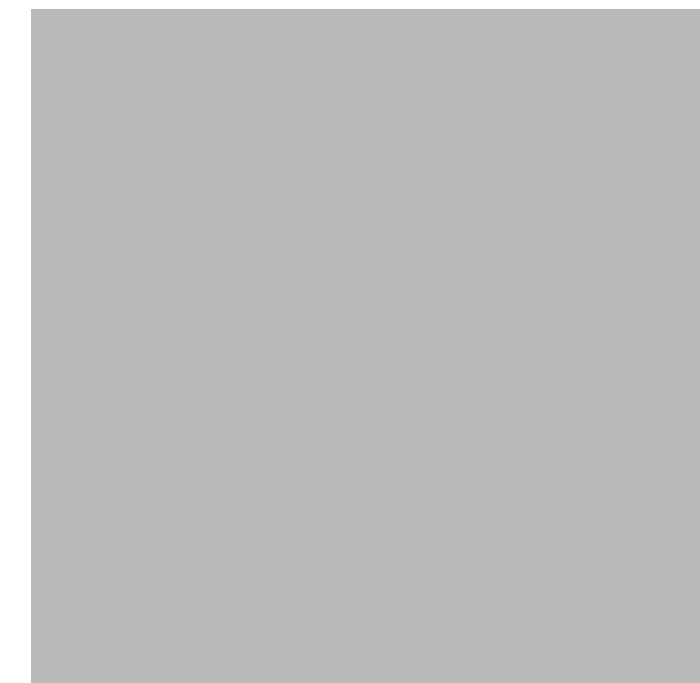


MIXE – Campaign 2021 (piE1)

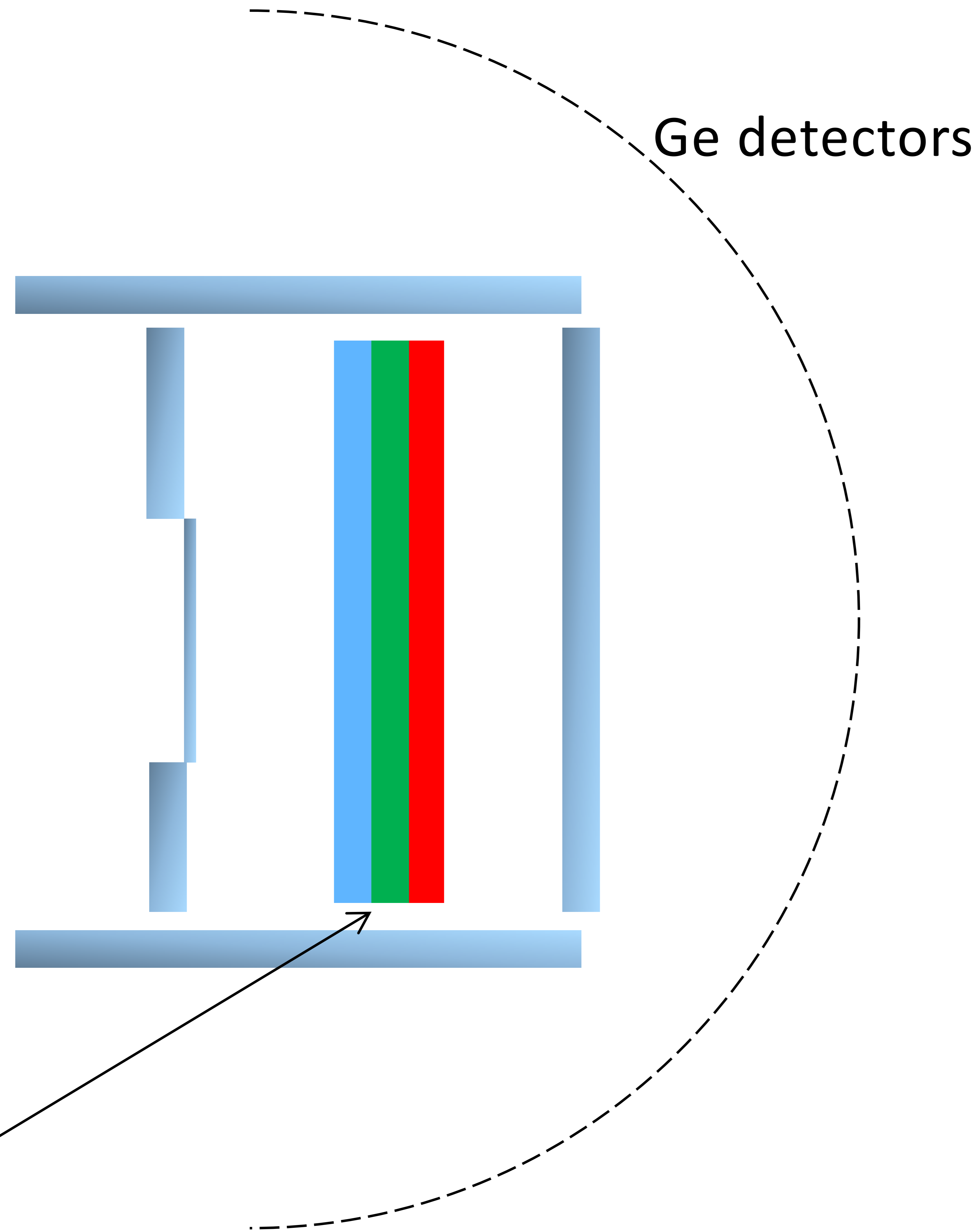


MIXE – Campaign 2021 (piE1)





Negative muons



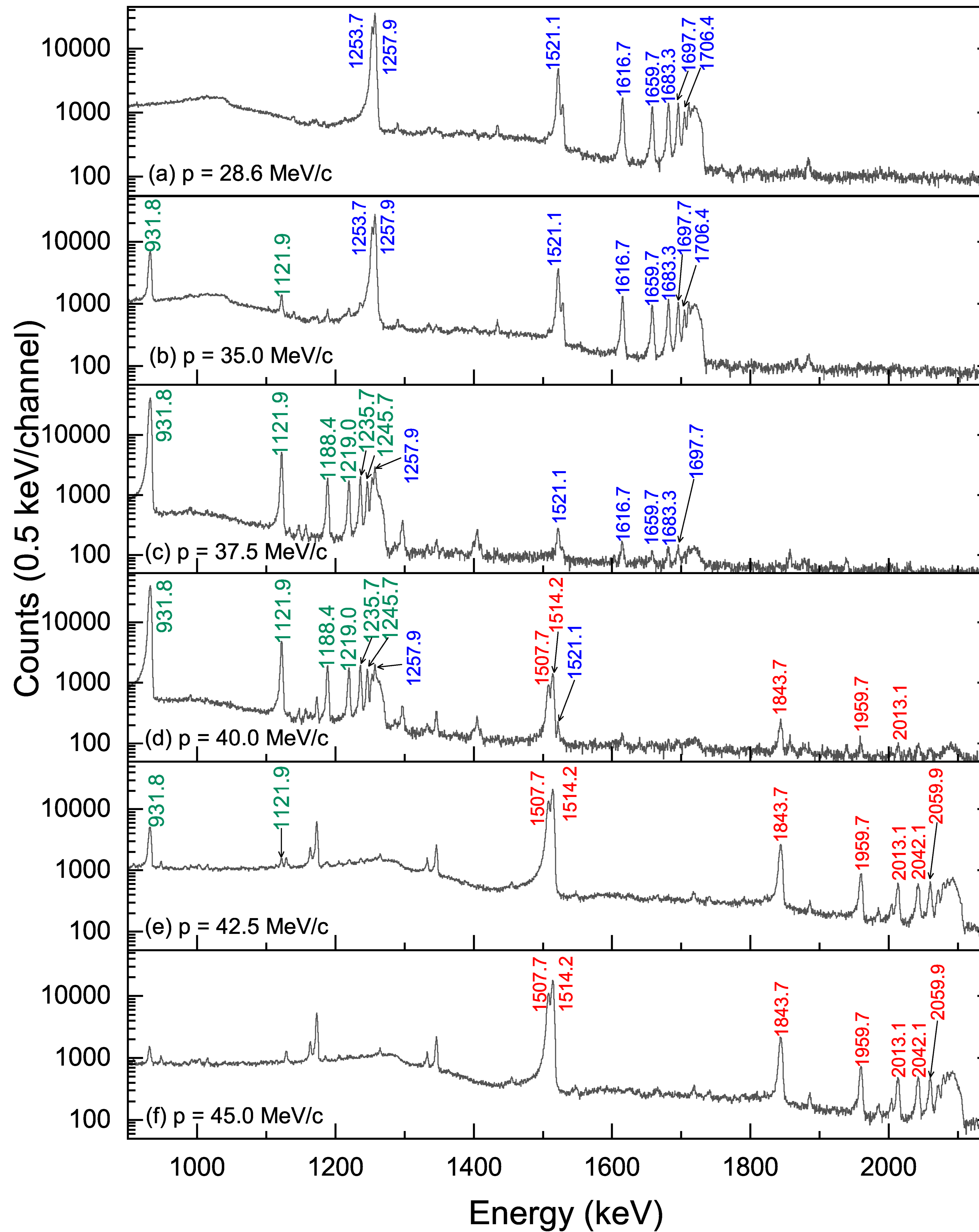
«Sandwich» sample

0.5 mm Fe

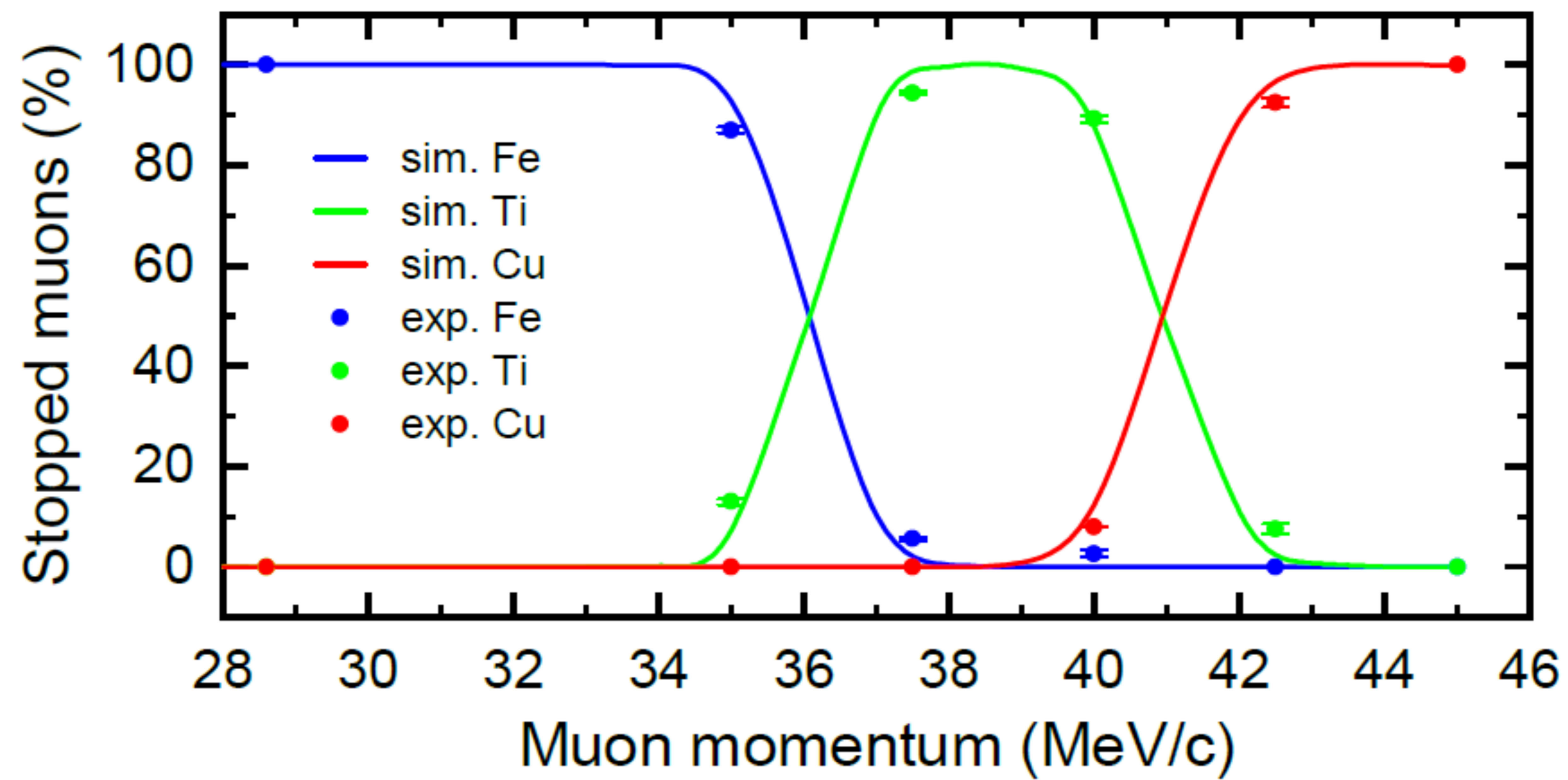
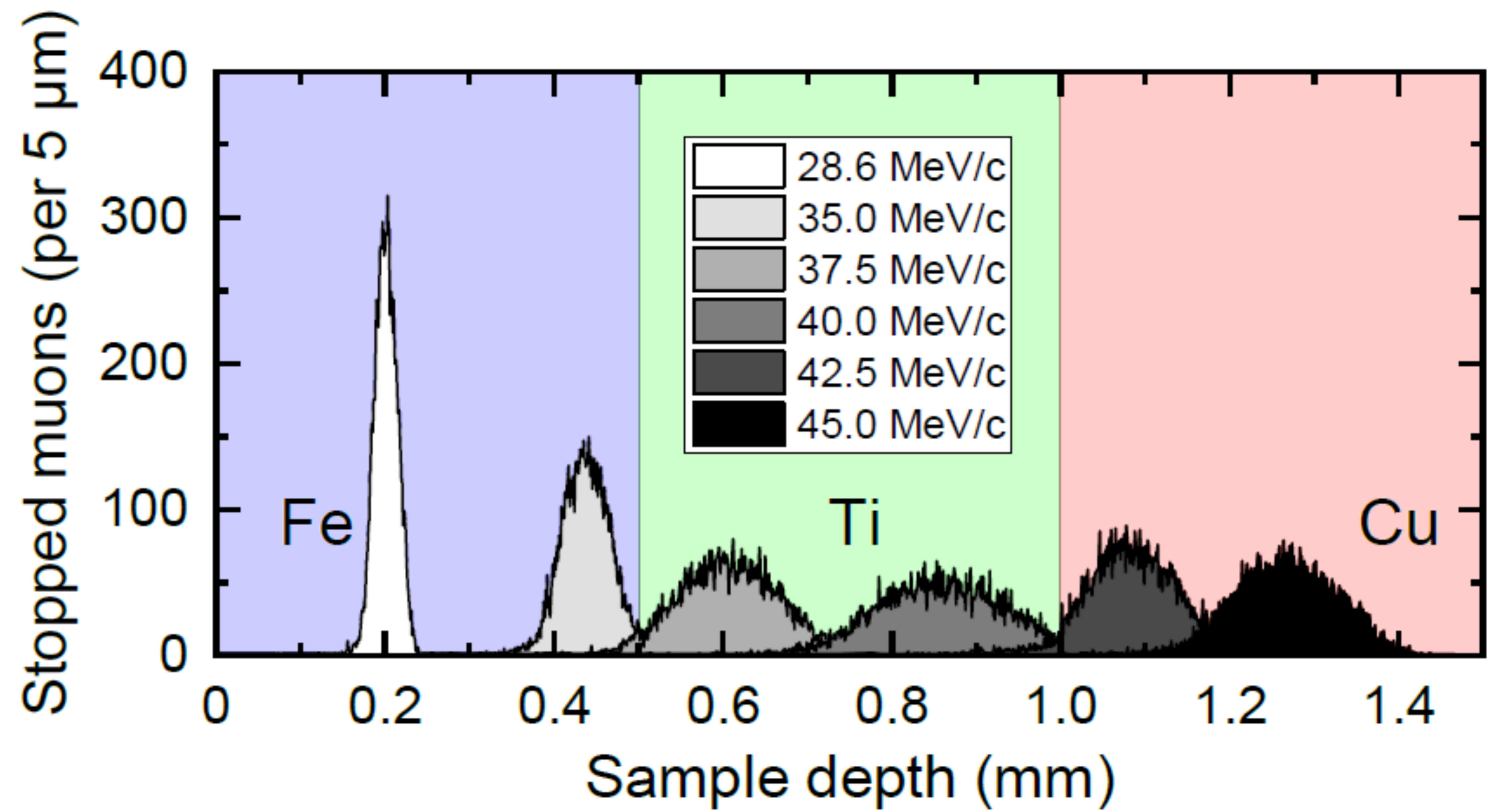
0.5 mm Ti

0.5 mm Cu

MIXE -- test



MIXE -- test



This is an easy test as a muon will only see one type of atom.

What about an alloy?

MIXE in alloys

Binary alloy $Z_k Z_{k'}$

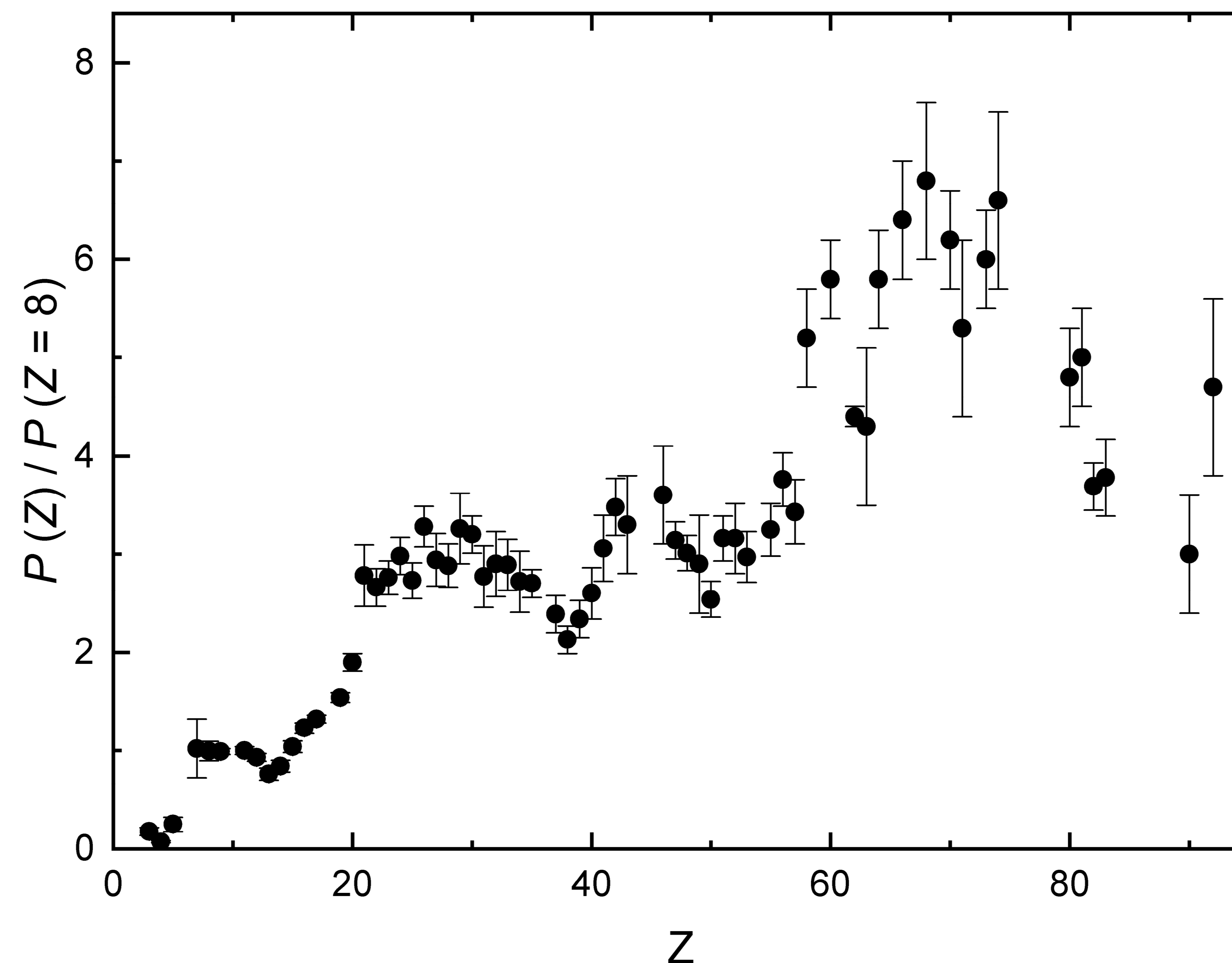
$$R(Z, Z', k, k') = A(Z, Z') \frac{k}{k'}$$

MIXE intensity ratio (including corrections)

Capture ratio between the 2 elements

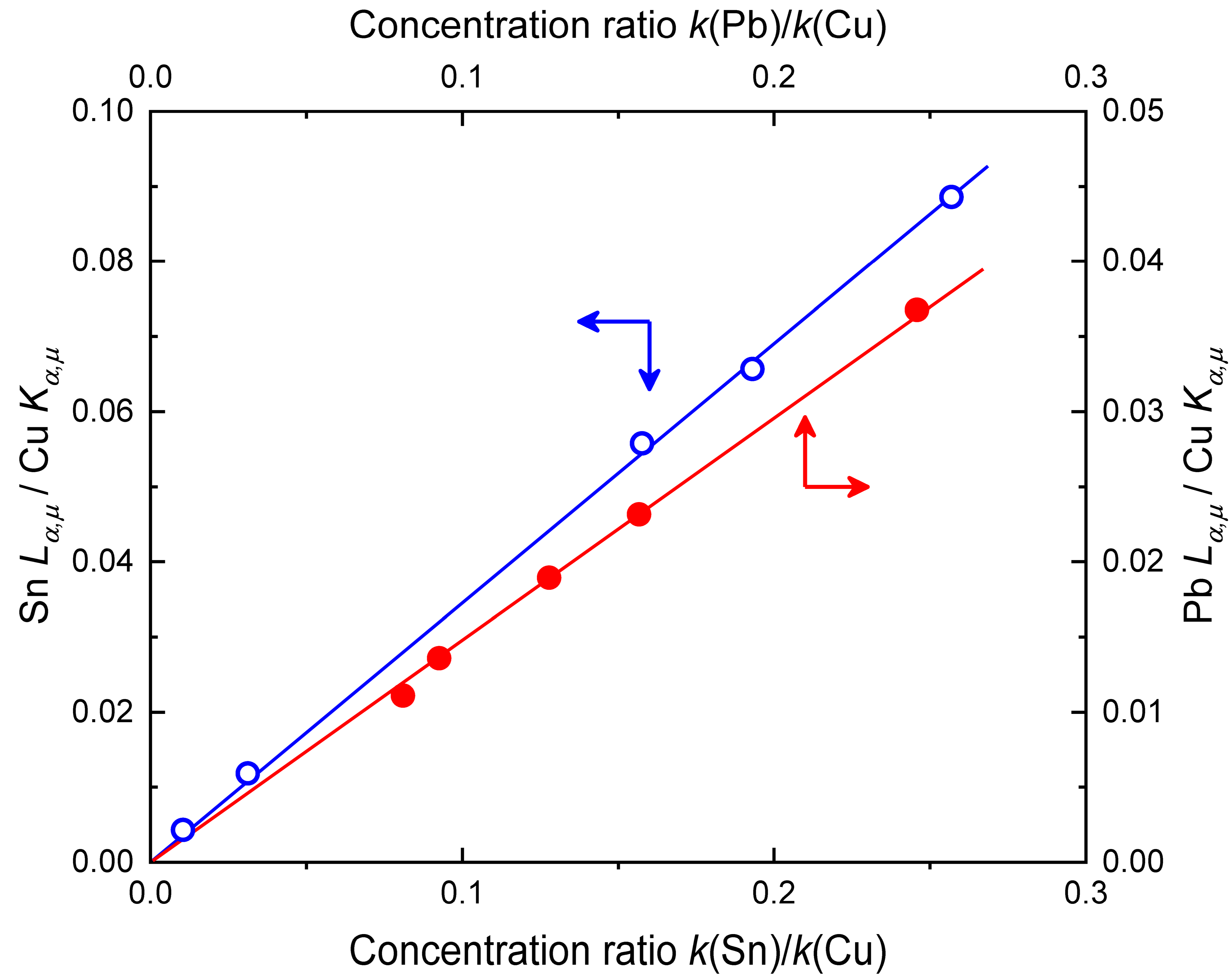
$$A(Z, Z') = \frac{P(Z)}{P(Z')}$$

capture probability: depends on the quantum numbers n and l of the electron involved in the Auger emission during the capture and the atomic number Z



MIXE in alloys

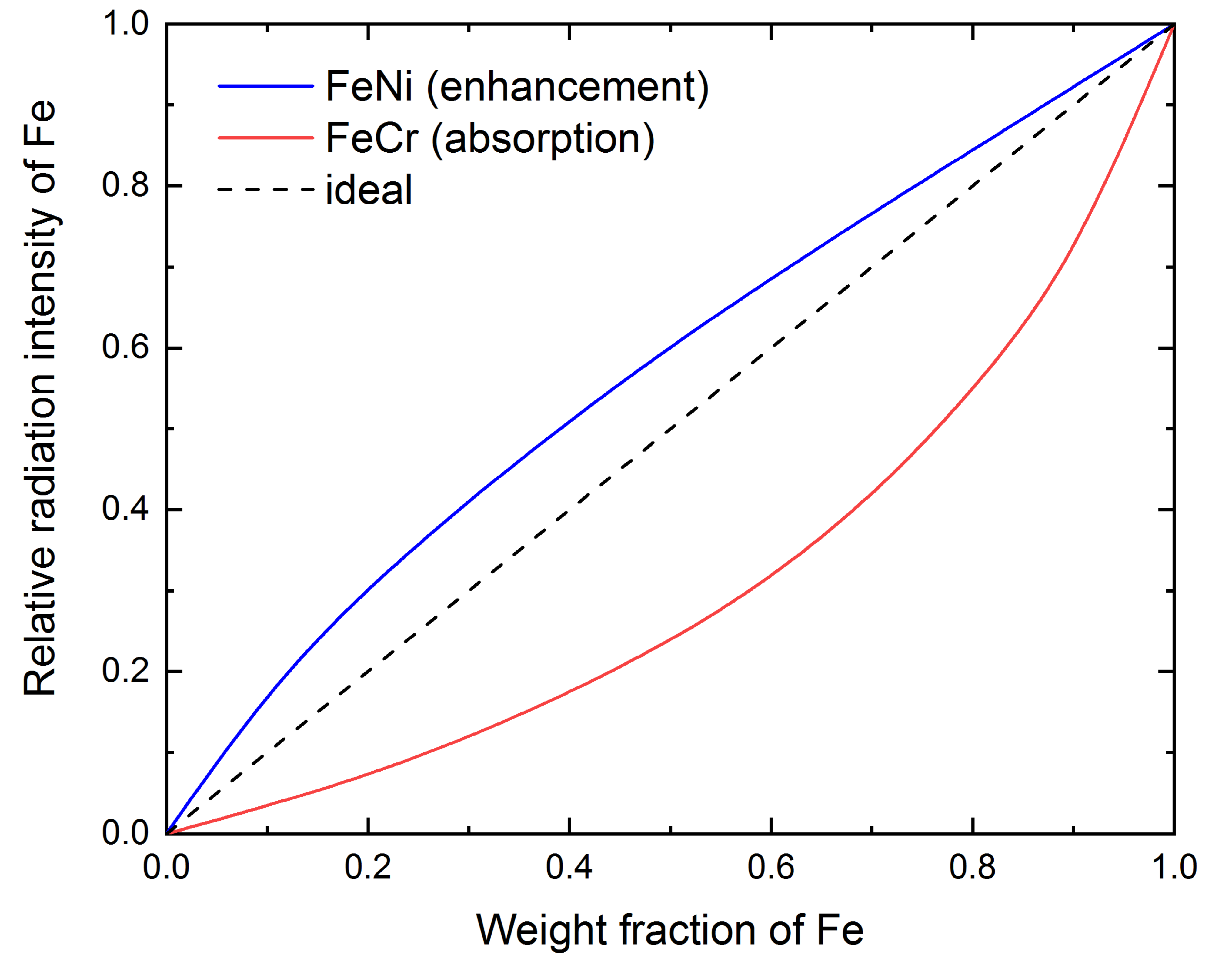
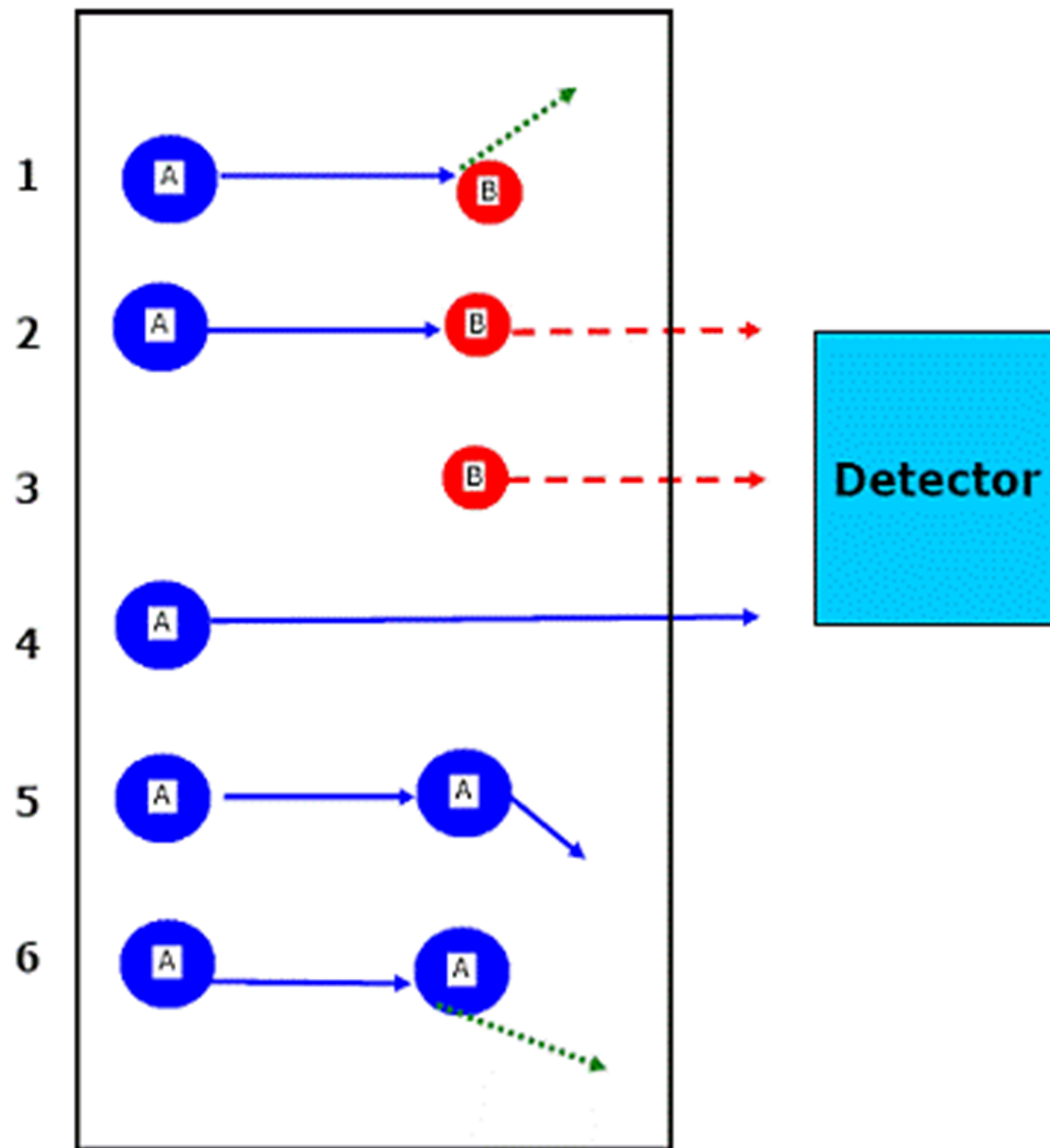
Also for ternary alloys: example $\text{Cu}_{k(\text{Cu})}\text{Sn}_{k(\text{Sn})}\text{Pb}_{k(\text{Pb})}$



$$R(Z, Z', k, k') = A(Z, Z') \frac{k}{k'}$$

Matrix effects in X-ray Fluorescence

- Absorption
- Enhancement



F.G. Banica, 09-03-20

“Quantification in X-Ray Fluorescence Spectrometry”

Rafał Sitko and Beata Zawisza

<https://doi.org/10.5772/29367>

Main characteristics

	<u>Muon-Induced X-ray Emission</u>
Abbreviation	MIXE
Primary Excitation	Negative Muon
Detected Secondary	X-ray
Elemental Range	3-92
Lateral Resolution	1 mm
Detected Depth	Up to several cm
Detection Limit	0.1% at least
Depth Profile	Yes
Destructive	No

Ideal for:

- Large objects
- Valuable objects

Examples:

- Rare objects (archeological artefacts, meteorites, return samples,...)
- Operando devices (batteries)
- ...

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Also: Absence of enhancement and absorption !

Thank you very much!

