2020.11.27 Symposium on Nuclear Data 2020 @ RIKEN Wako campus

Isotope production in spallation reaction of ^{93}Nb and ^{93}Zr induced by proton and deuteron

Japan Atomic Energy Agency
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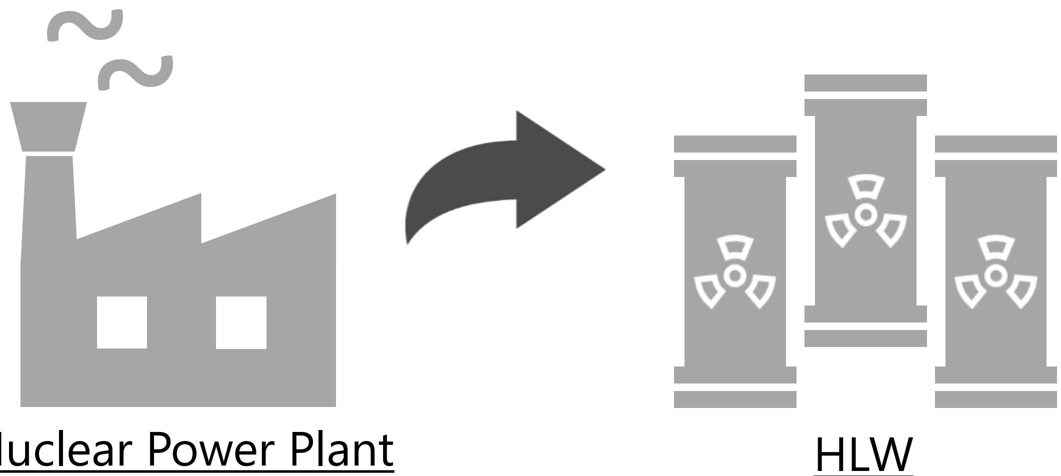
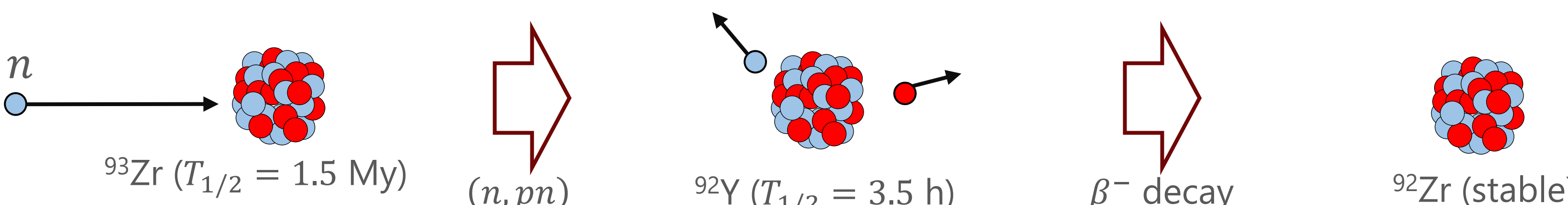
Issue of High-Level Radioactive Waste (HLW)



Minor Actinoids (MAs):
- ^{237}Np ($T_{1/2} = 2.41$ My)
- ^{243}Am ($T_{1/2} = 7.37$ ky)

Long-Lived Fission Products (LLFPs):
- ^{93}Zr ($T_{1/2} = 1.5$ My)
- ^{107}Pd ($T_{1/2} = 6.5$ My)
- ^{129}I ($T_{1/2} = 15.7$ My)

HLW will be disposed of more than 300 meters underground.
However, it is stagnated due to long-term radiotoxicity of long-lived nuclides.
→ Nuclear Transmutation is proposed to be one of the technical options.
But, still no effective method was found for LLFPs.



Typical LLFPs[2]

LLFP	Half-Life	Cumulative Fission Yield from ^{235}U	$\sigma_{n-\mathrm{cap}}$ [b]
^{79}Se	300 ky	0.044%	50.04
^{93}Zr	1500 ky	6.35%	2.24
^{107}Pd	6500 ky	0.146%	9.19
^{126}Sn	230 ky	0.056%	0.09
^{135}Cs	1300 ky	6.52%	8.30
^{99}Tc	210 ky	6.13%	23.68
^{129}I	15700 ky	0.543%	30.33

Among the LLFPs, ^{93}Zr…

- has large fission yield
- has relatively small neutron capture cross section
- can change into minor metals ($^{90,91,92}Zr$, etc…) through transmutation

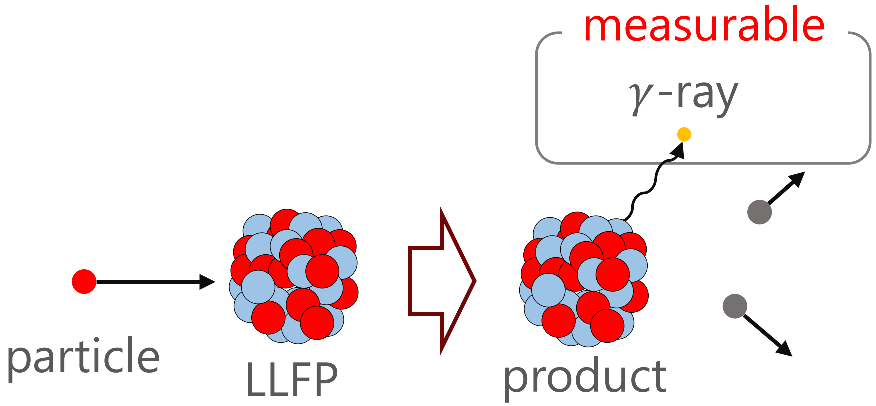
→ We focus on the transmutation of ^{93}Zr using spallation reaction by high-energy charged particles using an accelerator.

[2] K. Shibata et al., J. Nucl. Sci. Technol. 48(1), 1-30 (2011).

Introduction | Inverse Kinematics Method

Activation method was conventionally used
to measure the isotope-production cross sections.
However, activation method is NOT suitable for measurement of LLFP data.

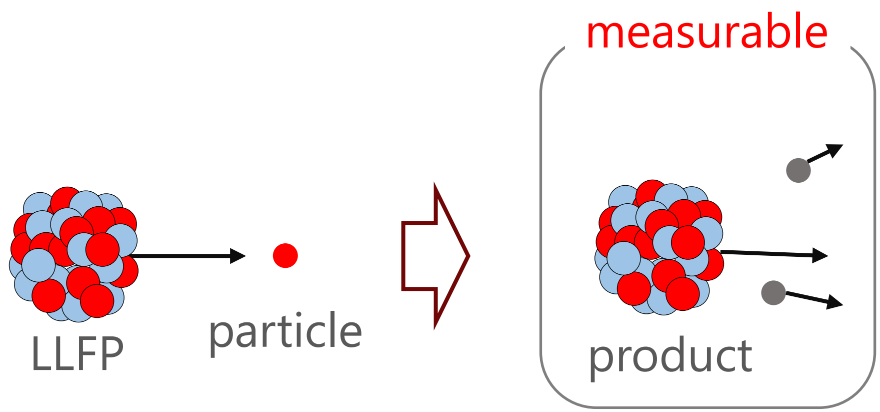
Activation method



In the case of LLFPs,

- ✕ Radioactive target
- ✕ Very difficult to measure yields of stable and short-lived nuclides
- ✓ Simple setup

Inverse kinematics method



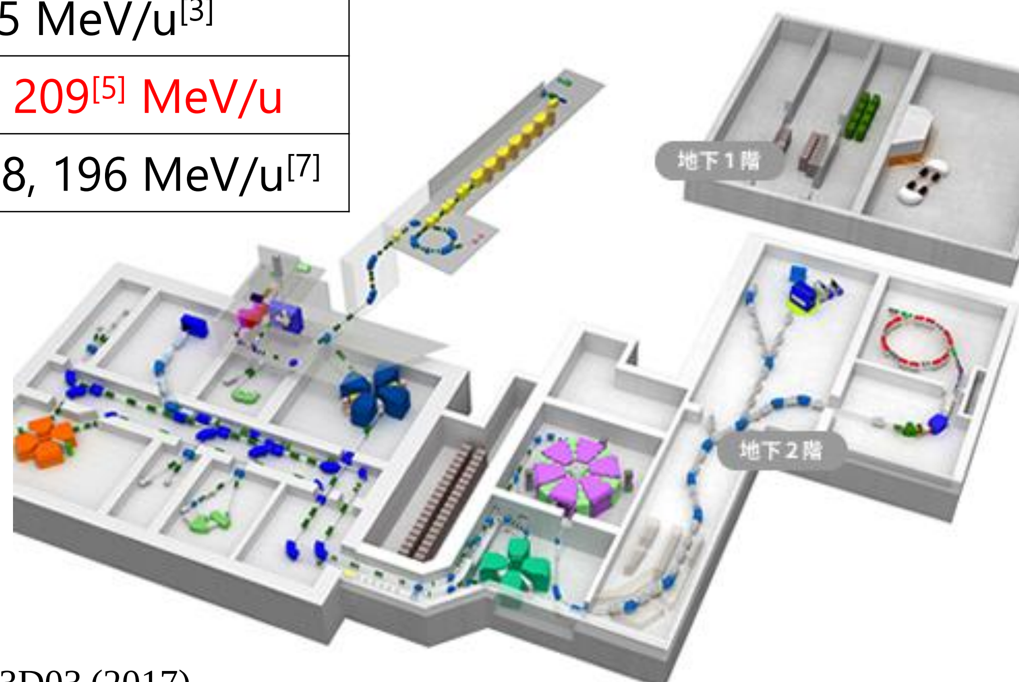
In the case of LLFPs,

- ✓ Stable target
- ✓ Stable and short-lived nuclides are also measurable
- ✕ Complicated setup

Introduction | Previous Works

So far, isotope-production cross sections
for some FPs and LLFPs were measured
using inverse kinematics method at RIKEN RI Beam Factory (RIBF).

Target	Beam (half-life $T_{1/2}$)	Energy
p, d	^{90}Sr (28 y)	185 MeV/u[3]
p, d	^{137}Cs (30 y)	185 MeV/u[3]
p, d	^{93}Zr (1.5 My)	105[4], 209[5] MeV/u
p, d	^{107}Pd (6.5 My)	50[6], 118, 196 MeV/u[7]



Overview of RIBF

[3] H. Wang et al., Phys. Lett. B 754, 104 (2016).
[4] S. Kawase et al., Prog. Theor. Exp. Phys. 2017, 093D03 (2017).
[5] S. Kawase et al., JAEA-Conf2018-001 2018, 111 (2018).
[6] H. Wang et al., Comm. Phys. 2, 2399 (2019).
[7] H. Wang et al., Prog. Theor. Exp. Phys. 2017, 021D01 (2017).

Data measured so far

Target	Beam (half-life $T_{1/2}$)	Energy
p, d	^{90}Sr (28 y)	185 MeV/u[3]
p, d	^{137}Cs (30 y)	185 MeV/u[3]
p, d	^{93}Zr (1.5 My)	105[4], 209[5] MeV/u
p, d	^{107}Pd (6.5 My)	50[6], 118, 196 MeV/u[7]

Purpose:

- To investigate the energy dependence of isotope-production cross section, and to accumulate fundamental knowledge for nuclear transmutation.
  → ^{93}Zr + p, d @ 50 MeV/u measurement
- To investigate the target dependence of isotope-production cross section.
  → ^{93}Nb + p, d,C @ 113 MeV/u measurement

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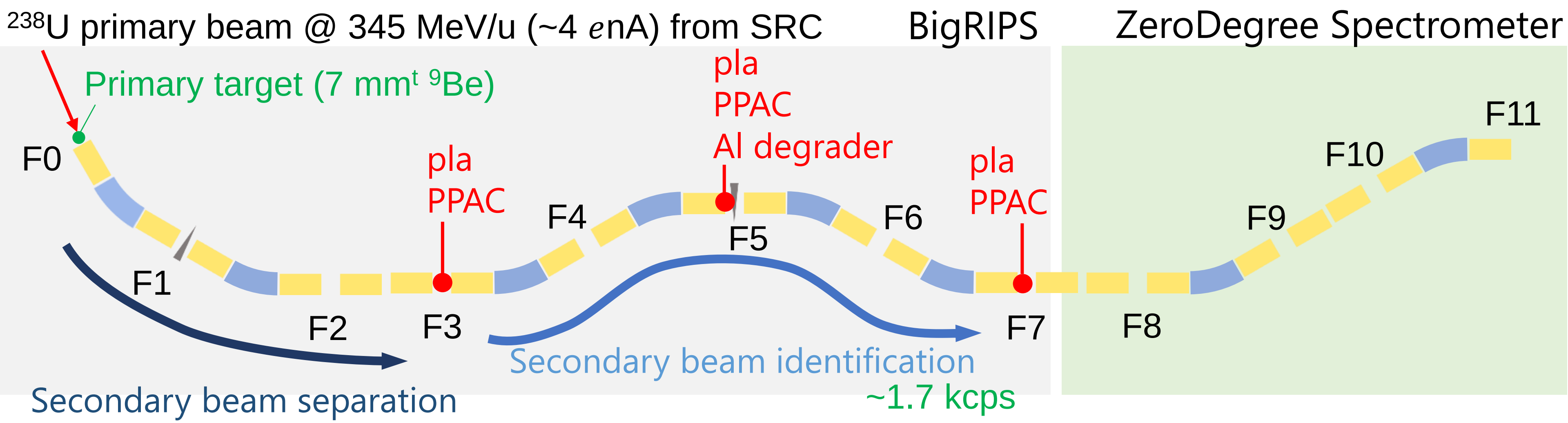
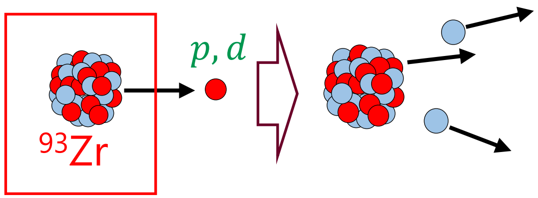
Experiment Ⅰ (^{93}Zr + p, d @ 50 MeV/u)

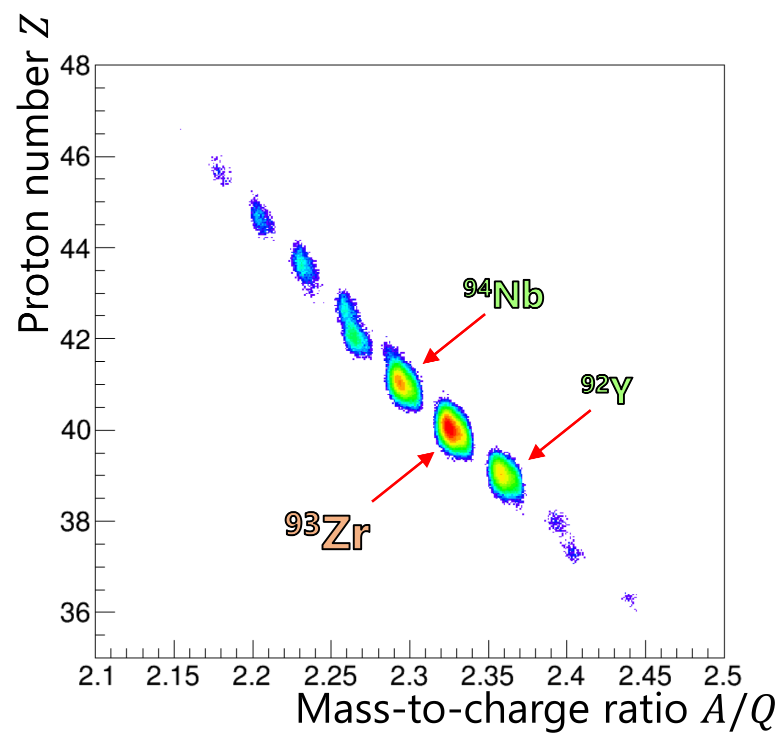


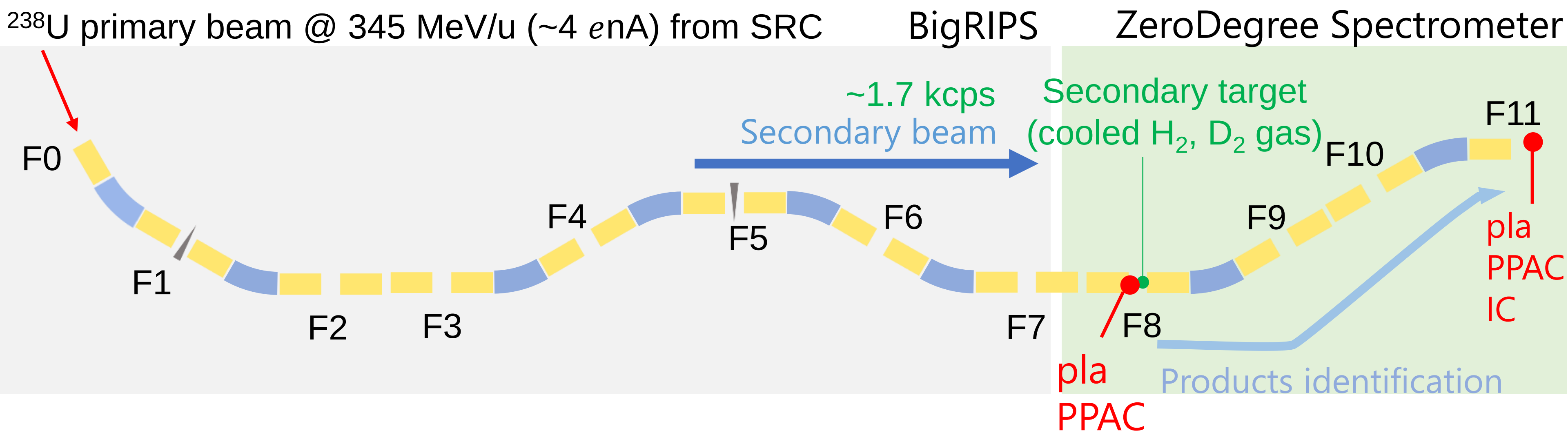
Identification of secondary beam by TOF−$B\rho$ − ΔE method



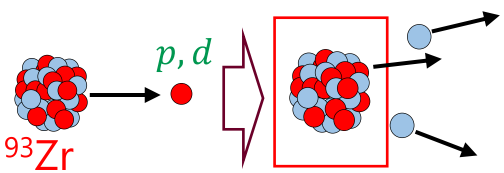
Plastic scintillator (pla) → β, γ
PPAC → $B\rho$ (magnetic rigidity)
Energy loss at Al degrader → ΔE

Proton number: $Z \propto \beta\sqrt{\Delta E}$
Mass-to-charge ratio: $A/Q \propto B\rho/\beta\gamma$



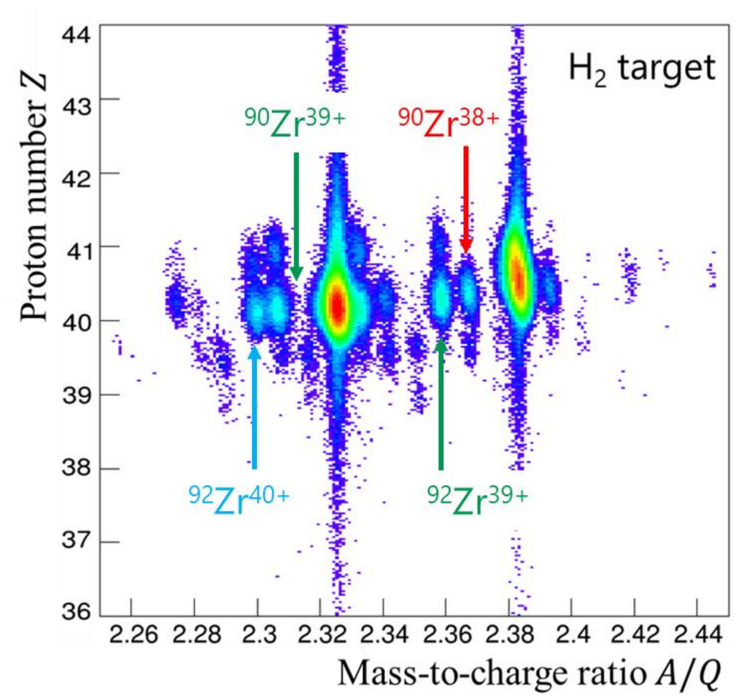


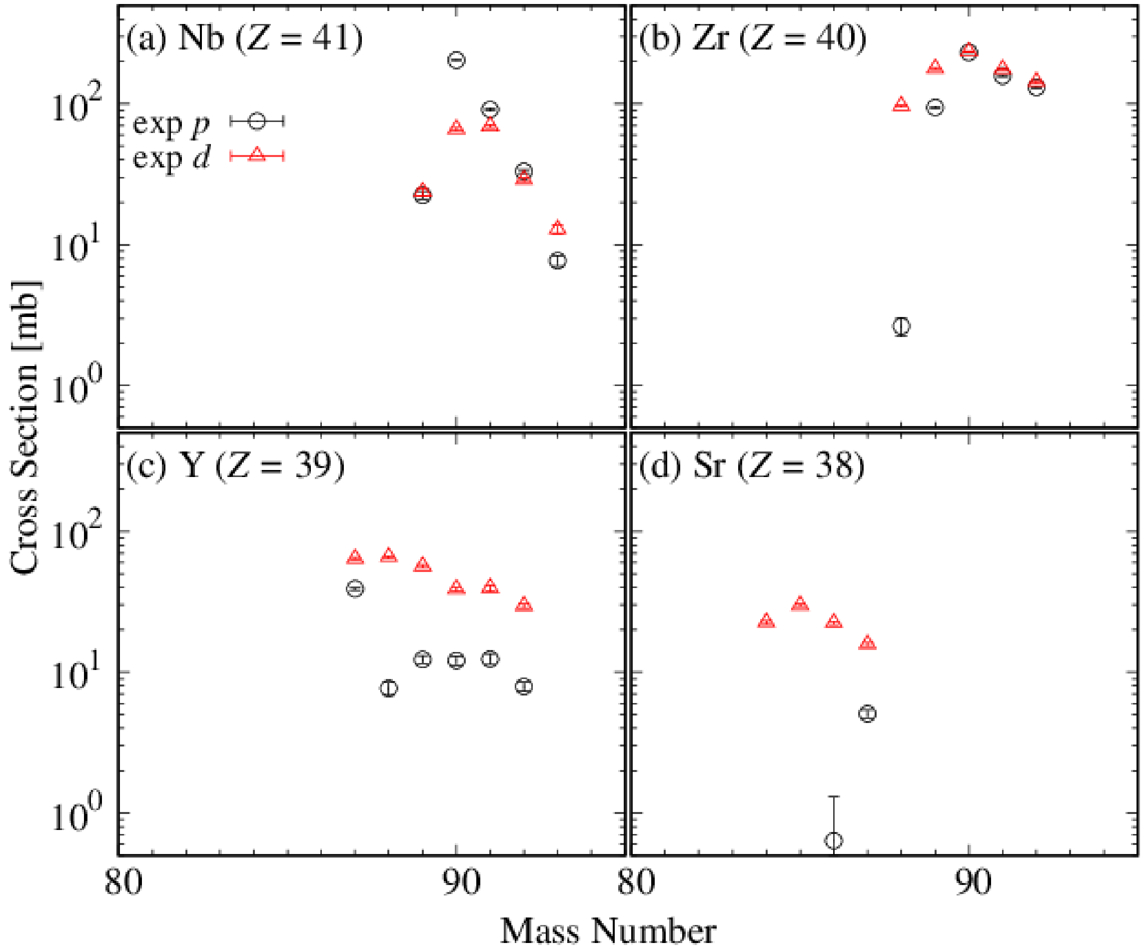
Identification of reaction products by TOF$-B\rho-\Delta E$ method



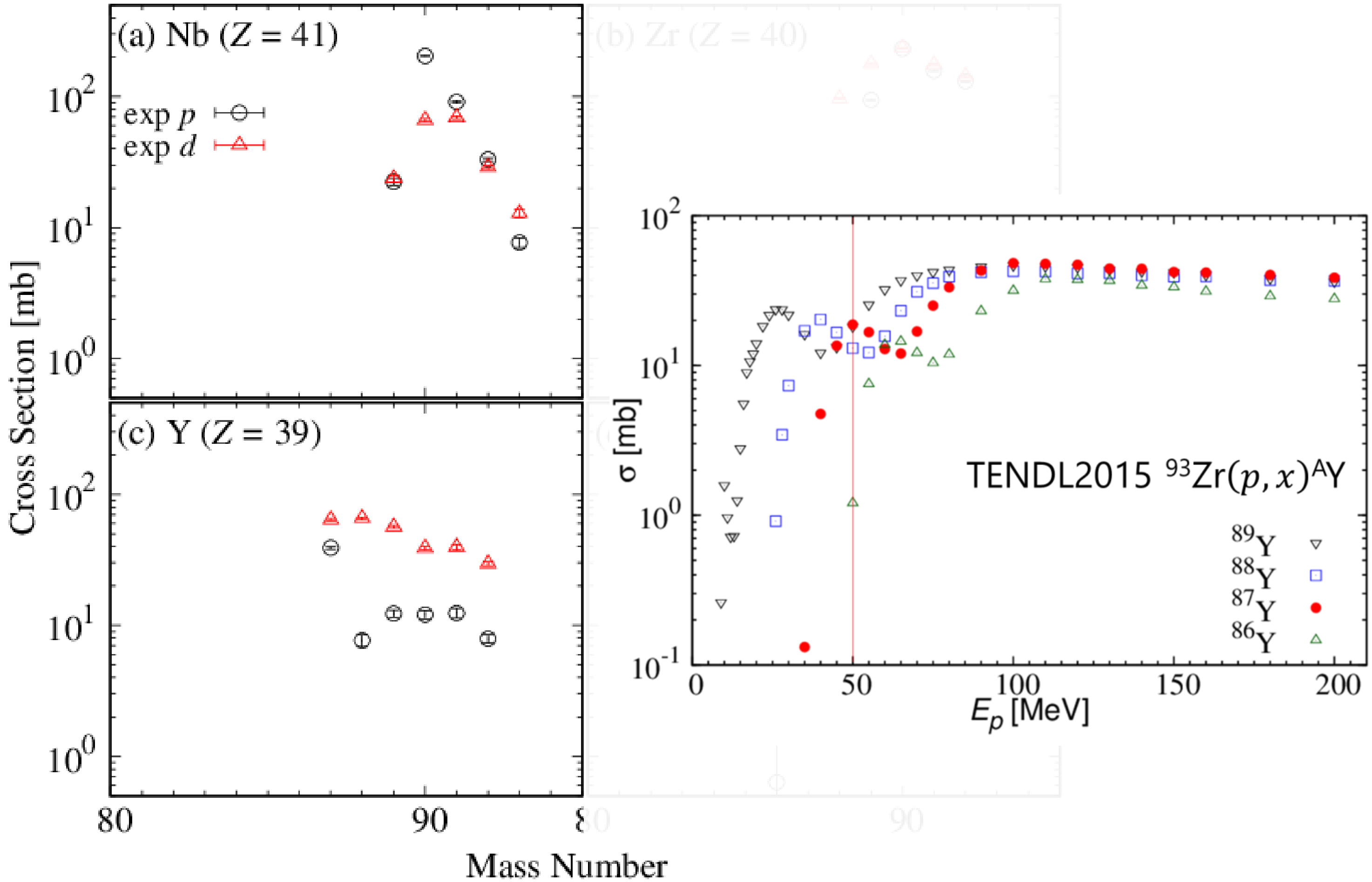
Plastic scintillator (pla) → β, γ
PPAC → $B\rho$ (magnetic rigidity)
Ionization chamber (MUSIC) → ΔE

Proton number: $Z \propto \beta\sqrt{\Delta E}$
Mass-to-charge ratio: $A/Q \propto B\rho/\beta\gamma$



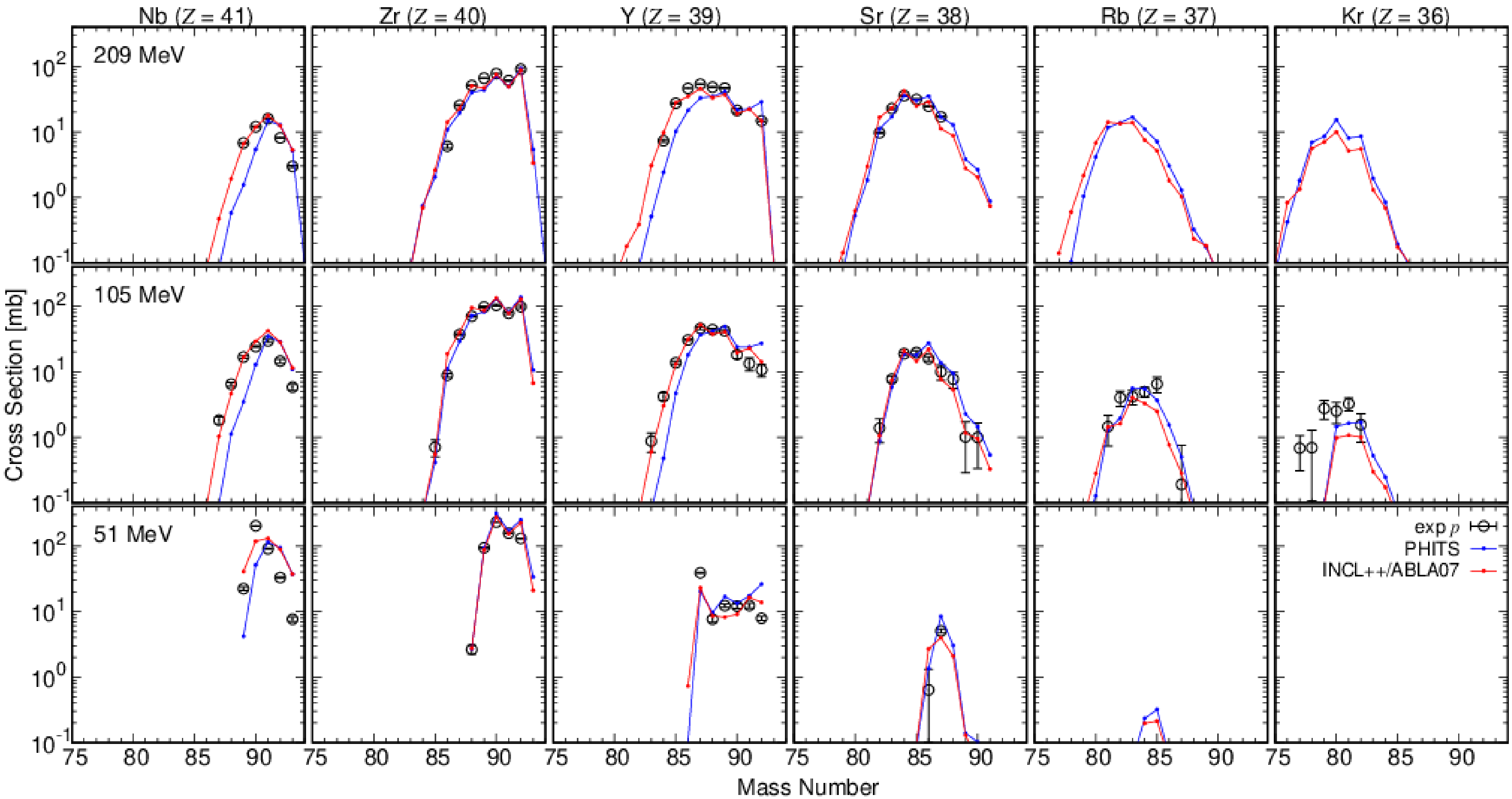


- We obtained σ_p and σ_d for 18 and 20 isotopes, respectively.
  → Advantage of the inverse kinematics method.



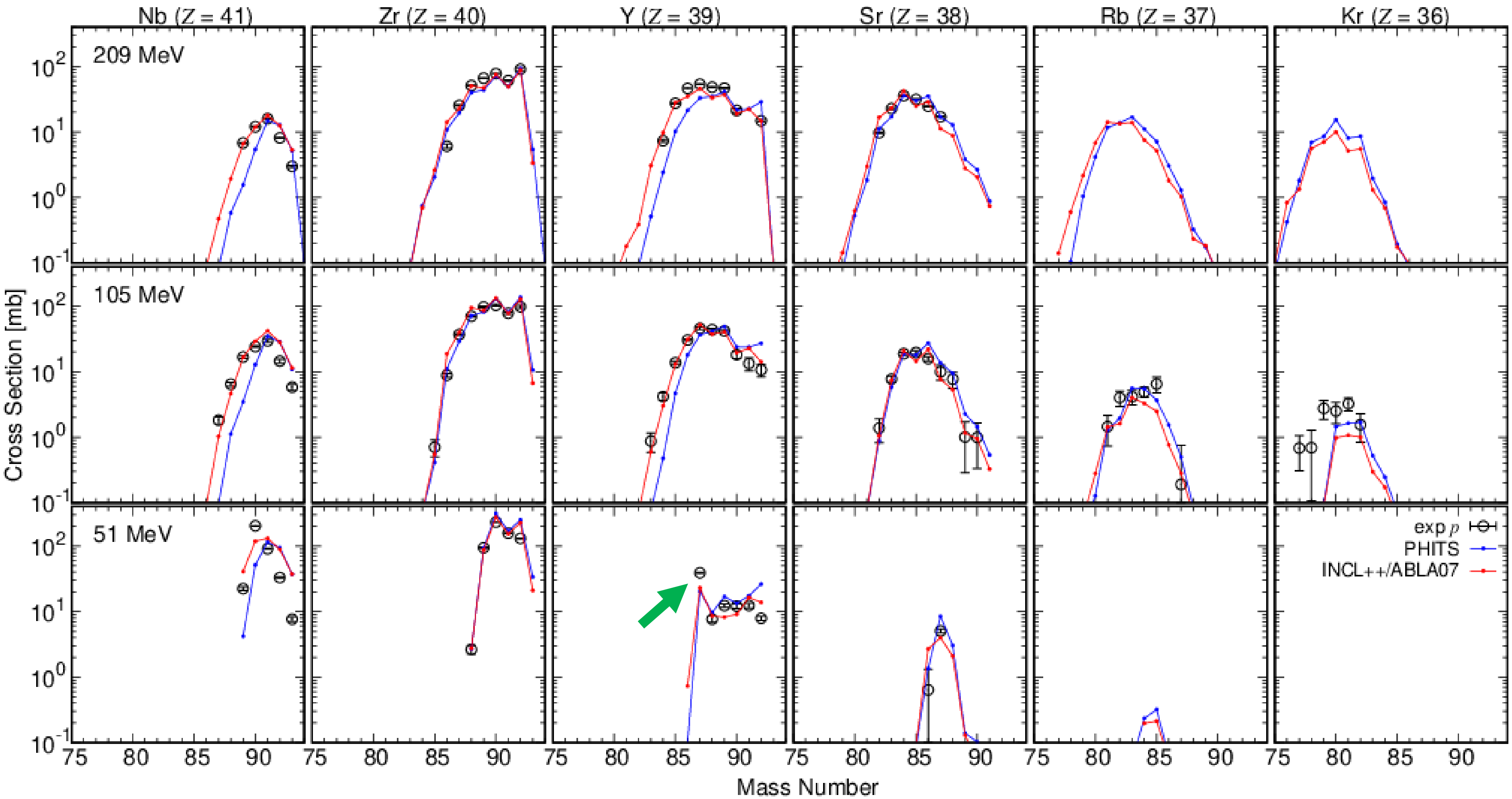
- ^{87}Y has noticeable large production cross section.
  → Incident energy of 50 MeV corresponds to the first peak.

Results | Comparison with model calculations ($^{93}Zr + p$)



- Shapes and quantities are well reproduced by PHITS and INCL++/ABLA07
  - Peak at ^{87}Y in σ_{p51}
  - Jumps at $N = 50$ originated from neutron magic number

Results | Comparison with model calculations ($^{93}Zr + p$)



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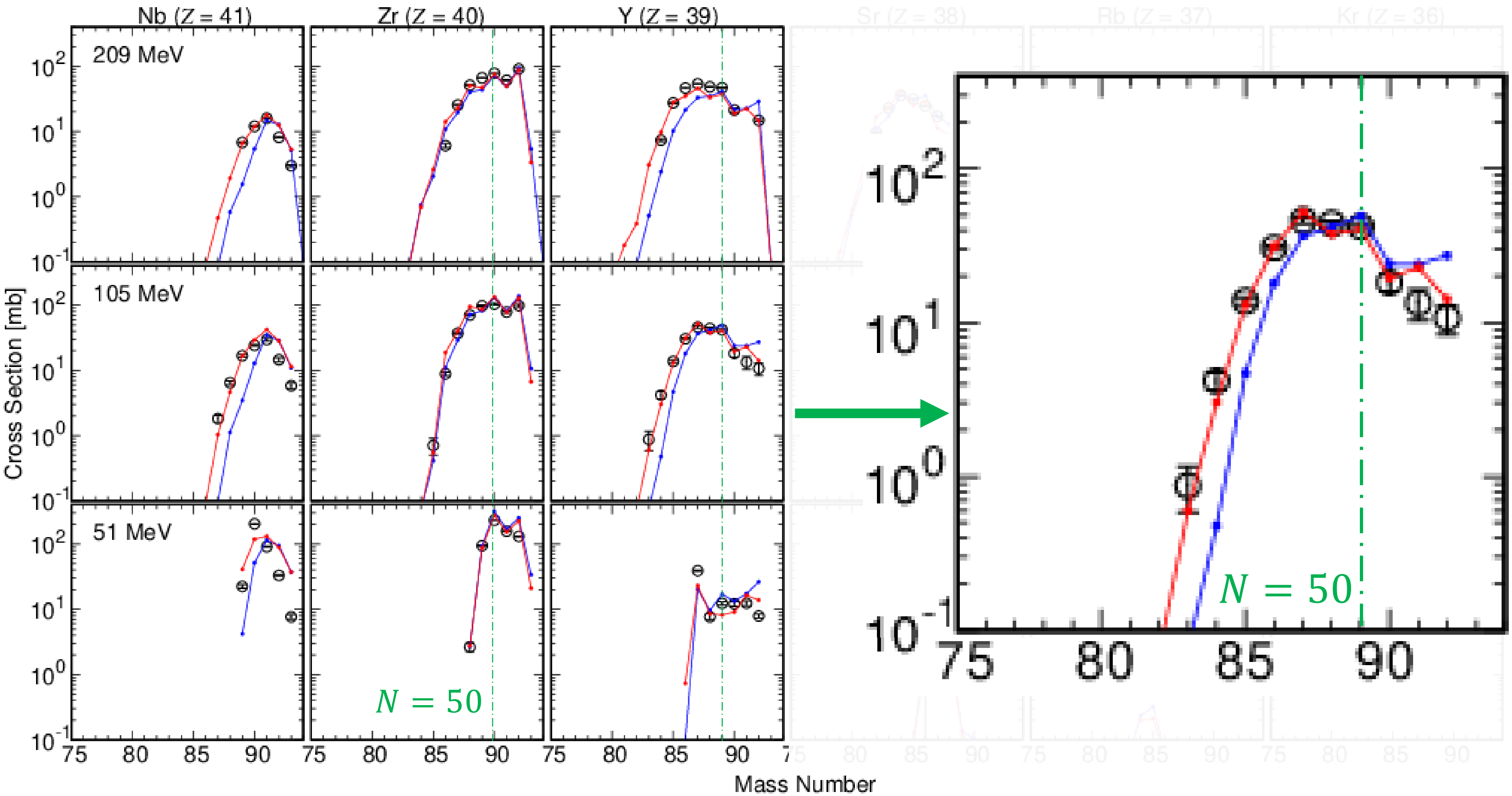

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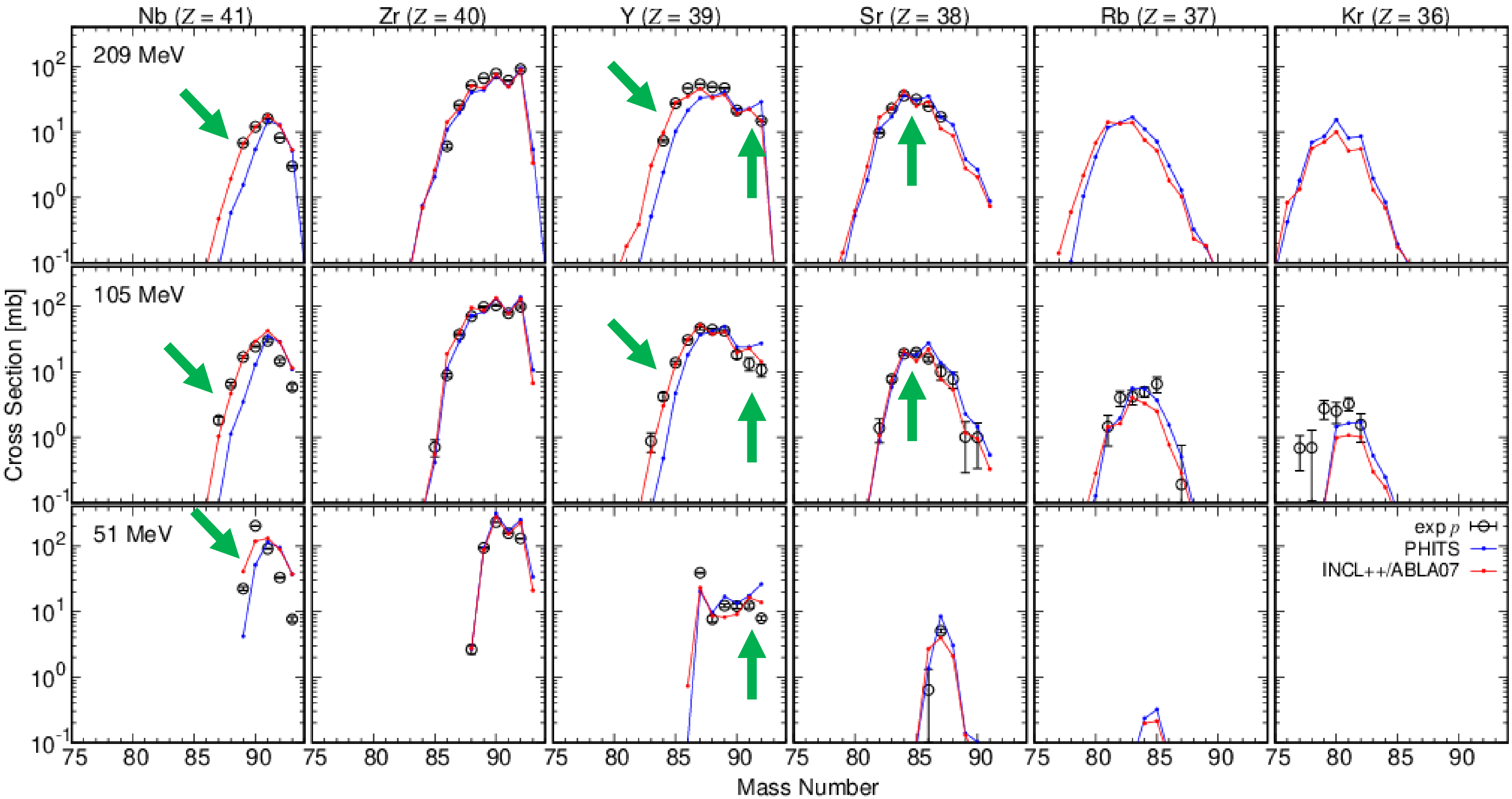
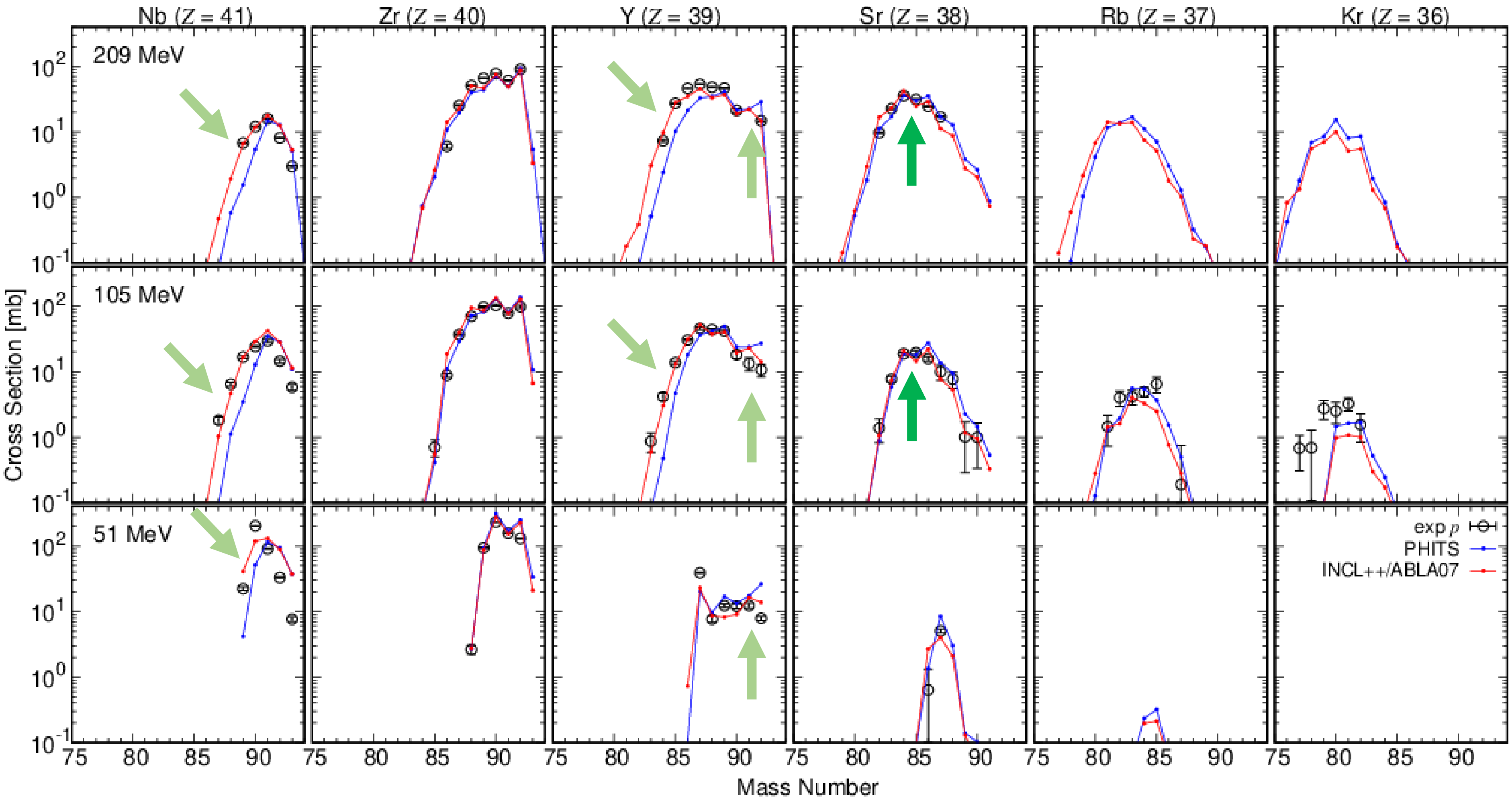


In PHITS calculation,

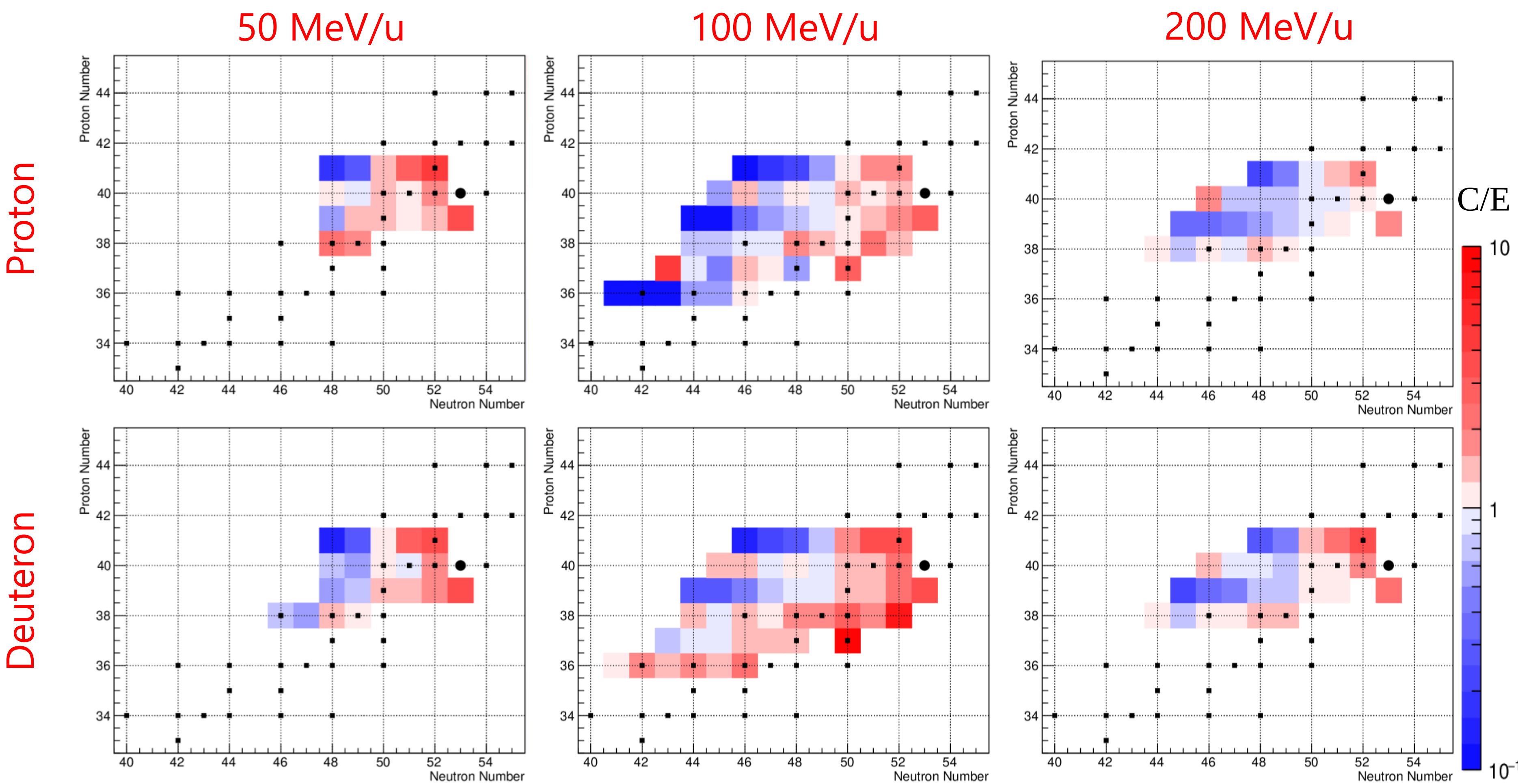
- underestimation in n-deficient region in odd-Z.
- overestimation in isotope near the target nucleus ^{93}Zr.
- exaggerated even-odd staggering.



INCL++/ABLA07 calculation

- well reproduces σ_p in n-deficient region in odd-Z and $(p, 2p)$ reactions
- overestimate even-odd staggering and σ_p in (p, pn) and (p, n) reactions

Results | Comparison with model calculations (^{93}Zr + p, d)

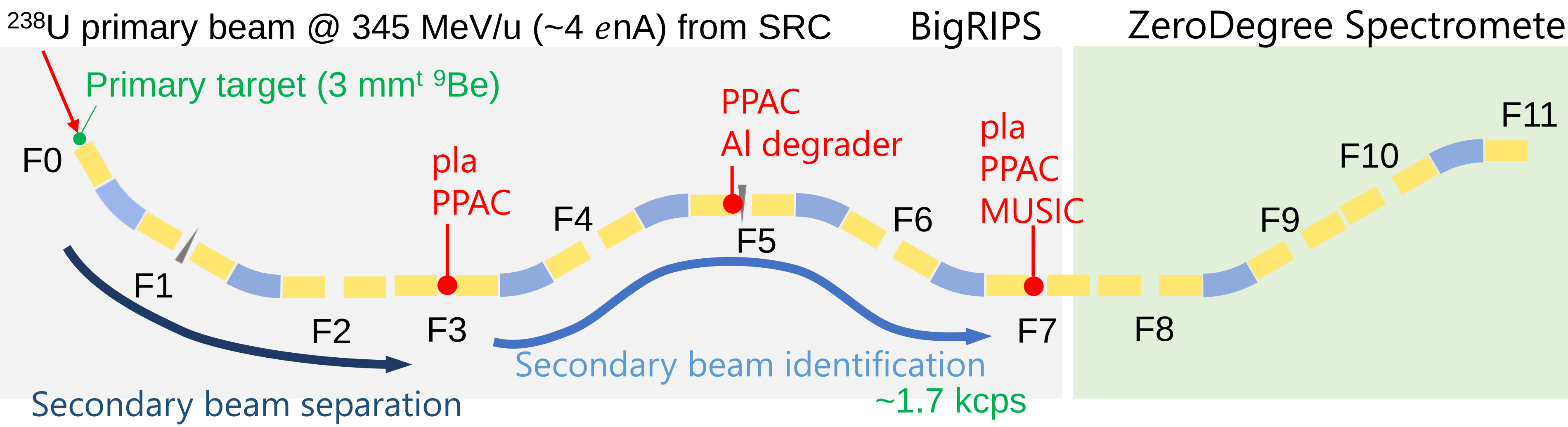


C/E plot of PHITS calculation on nuclear chart

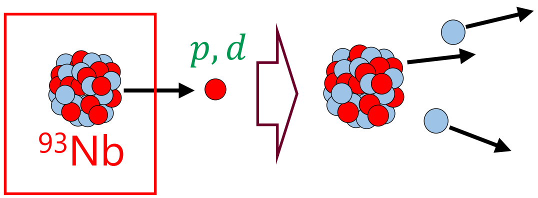
- underestimation in n-deficient region in odd-Z.
- overestimation in isotope near the target nucleus ^{93}Zr.
- exaggerated even-odd staggering.

Contents

Experiment Ⅱ (^{93}Nb + p, d @ 113 MeV/u)

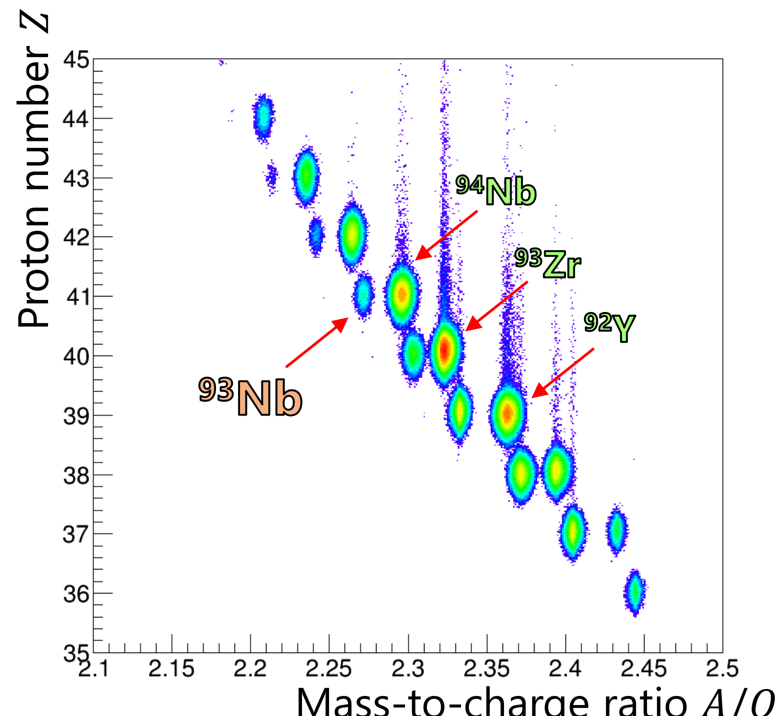


Identification of secondary beam by TOF−$B\rho$ − ΔE method

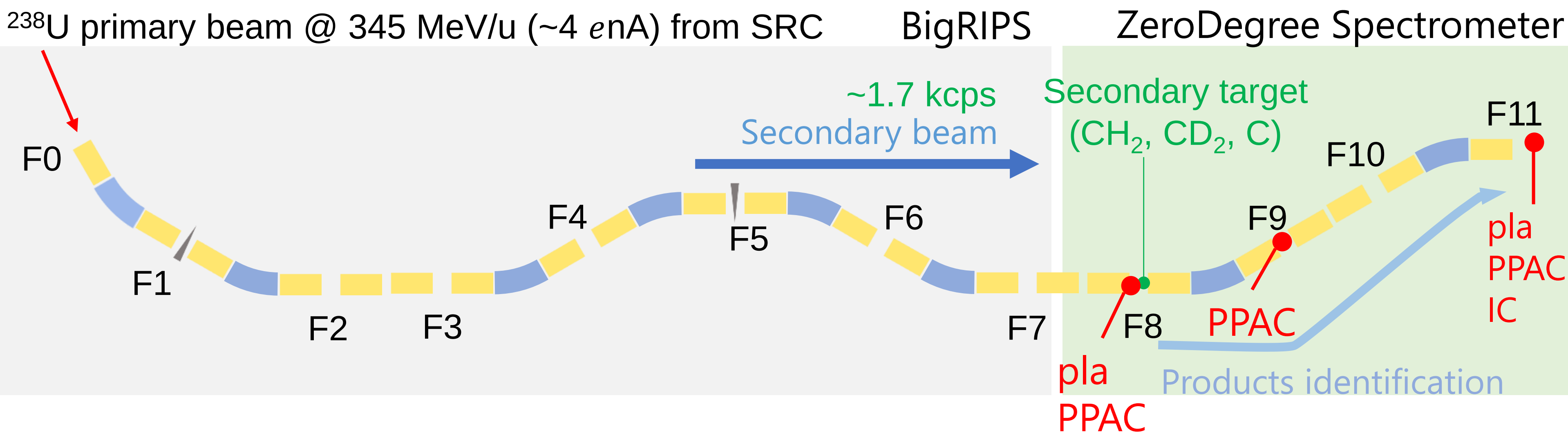


Plastic scintillator (pla) → β, γ
PPAC → $B\rho$ (magnetic rigidity)
Ionization chamber (MUSIC) → ΔE

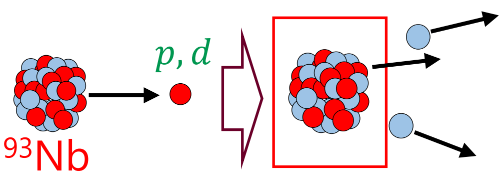
Proton number: $Z \propto \beta\sqrt{\Delta E}$
Mass-to-charge ratio: $A/Q \propto B\rho/\beta\gamma$



Experiment Ⅱ (^{93}Nb + p, d @ 113 MeV/u)



Identification of reaction products by TOF−$B\rho$ − ΔE method



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PPAC → $B\rho$ (magnetic rigidity)
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Proton number: $Z \propto \beta\sqrt{\Delta E}$
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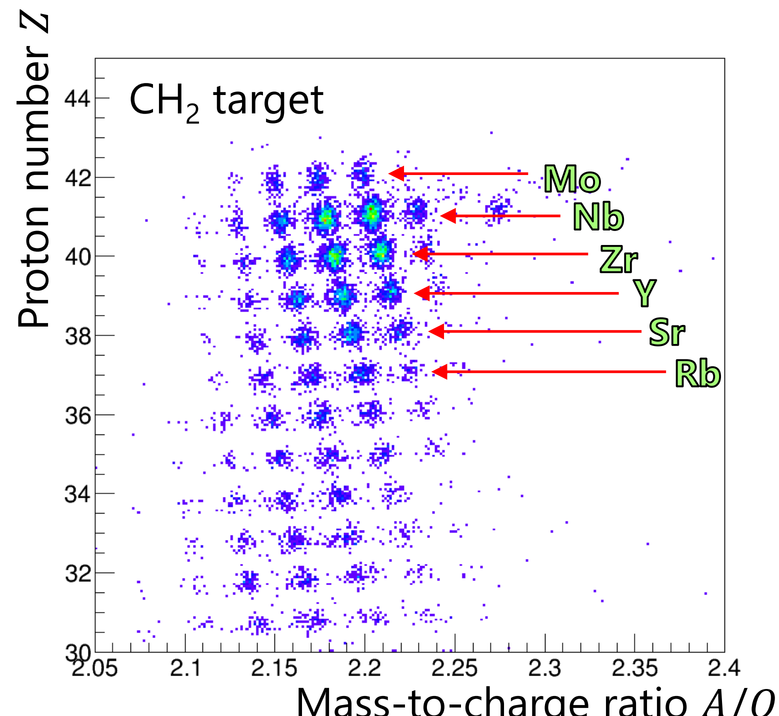
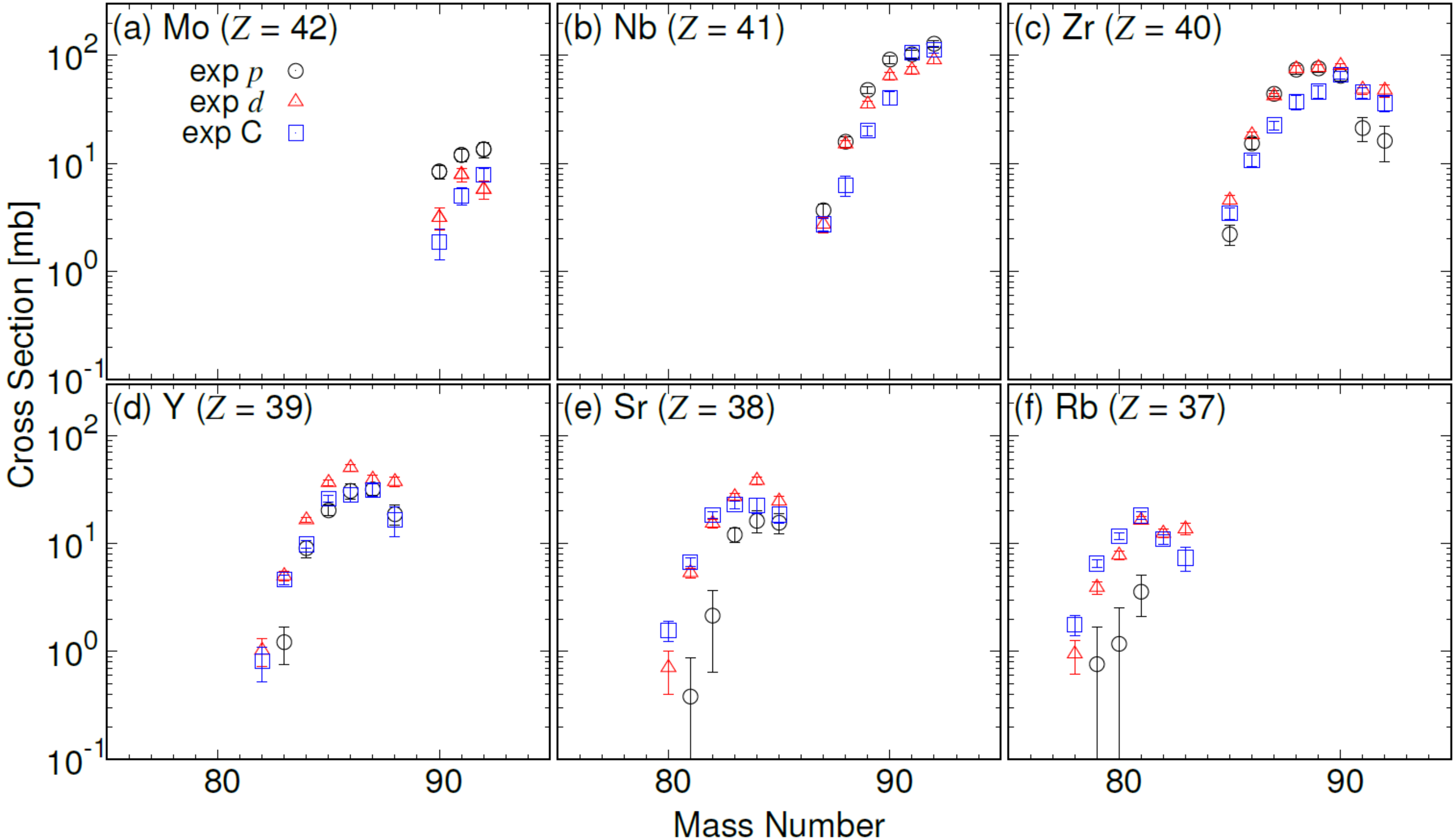
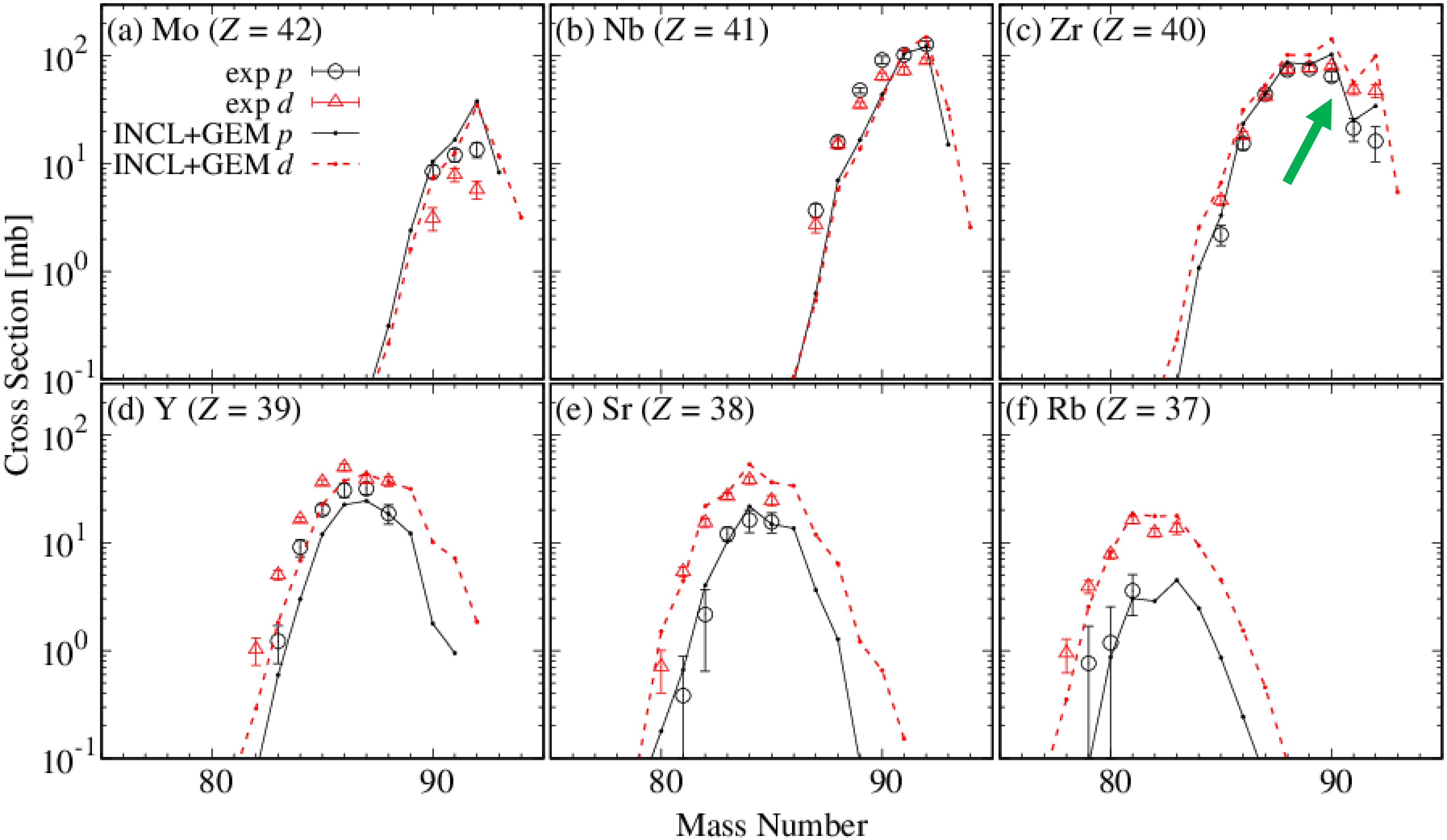
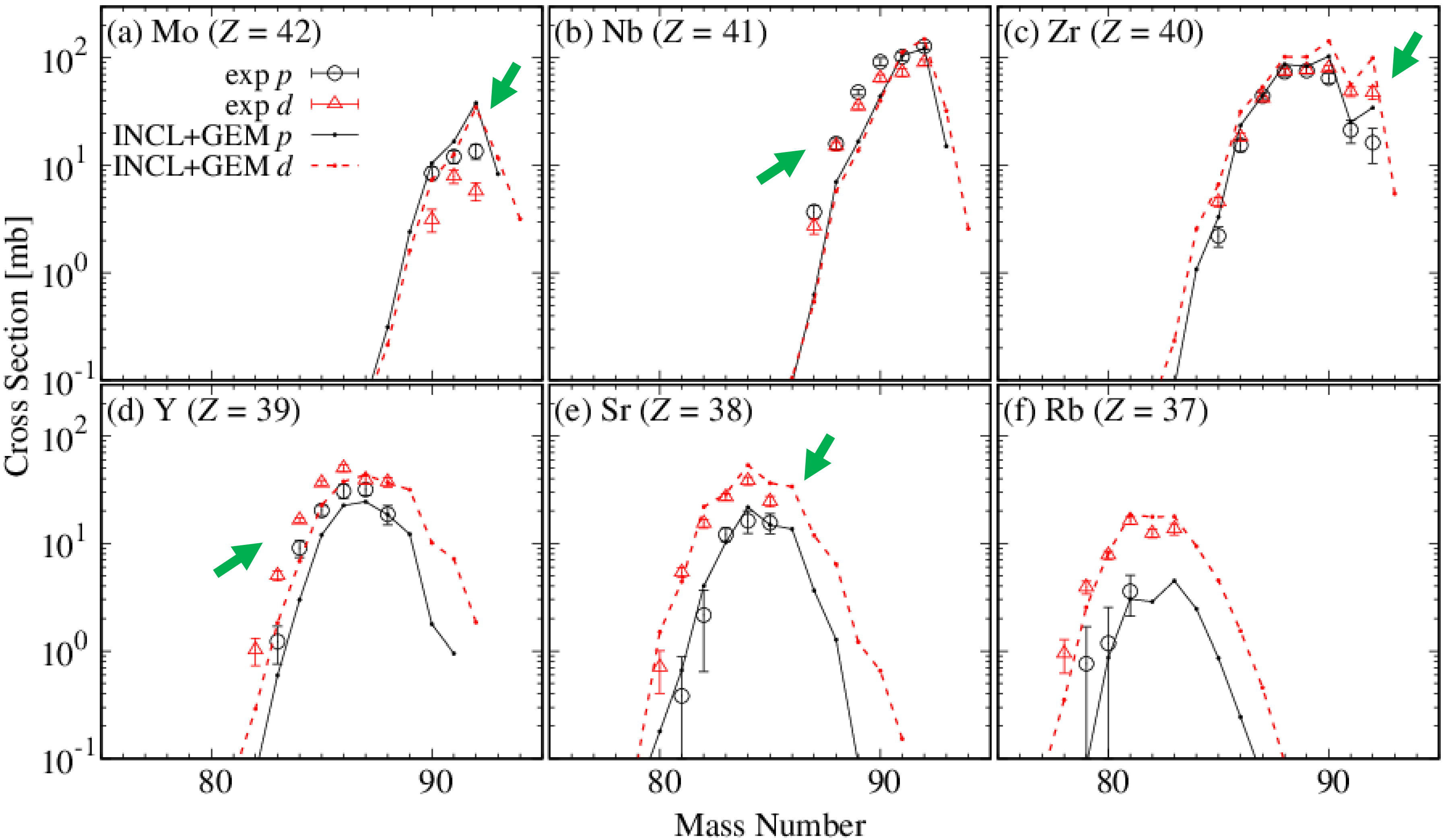




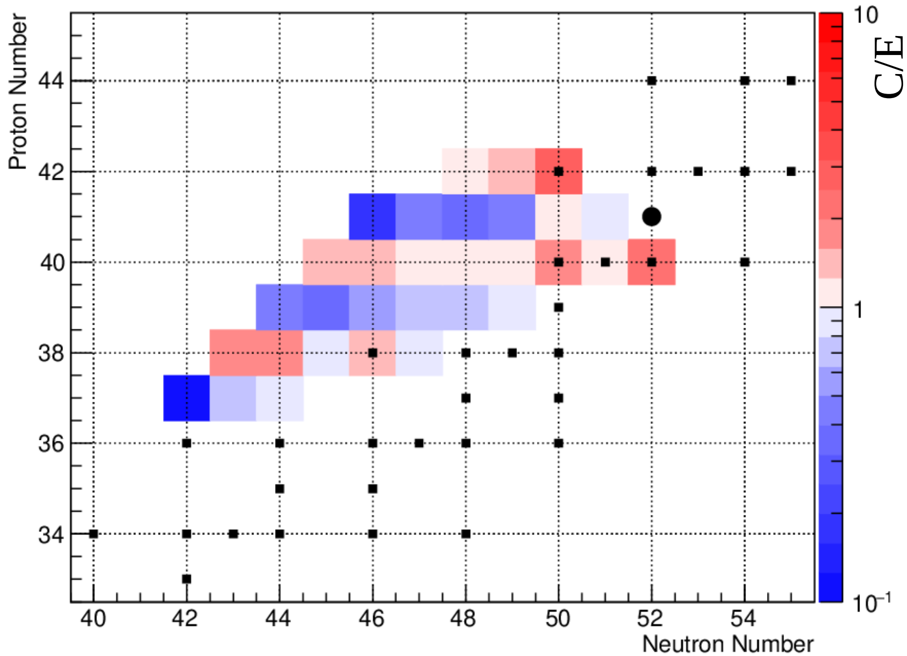
- We obtained $\sigma_p, \sigma_d,$ and σ_C for 33, 36, and 36 isotopes, respectively.
  → Advantage of the inverse kinematics method.



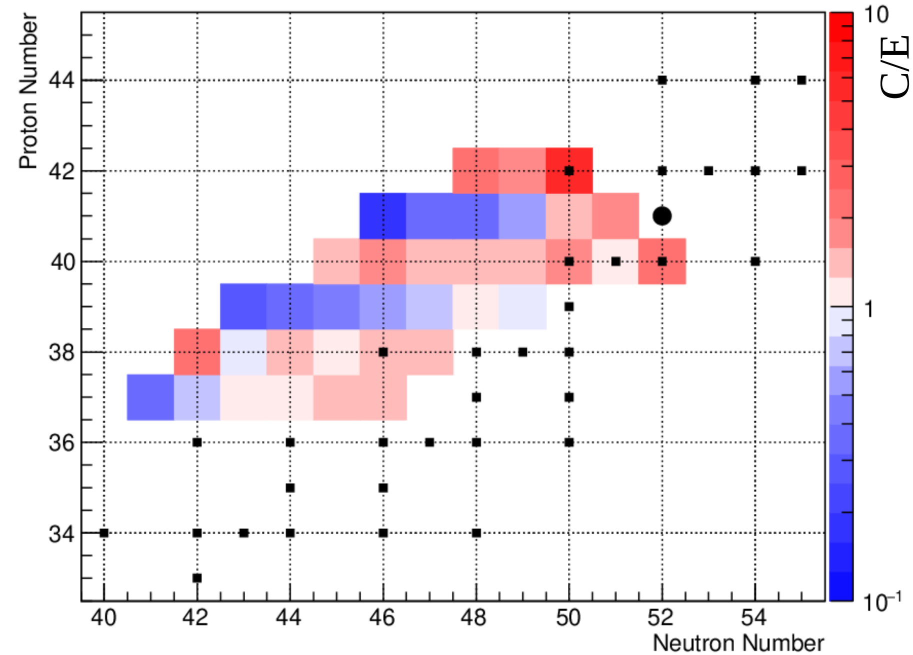
- PHITS calculations show generally good agreement.
- Jump at $N = 50$ is reproduced reasonably well.



- Overestimation in isotopes near the target nucleus
- Exaggerated even-odd staggering
- Underestimation in neutron-deficient region in odd-Z isotopes



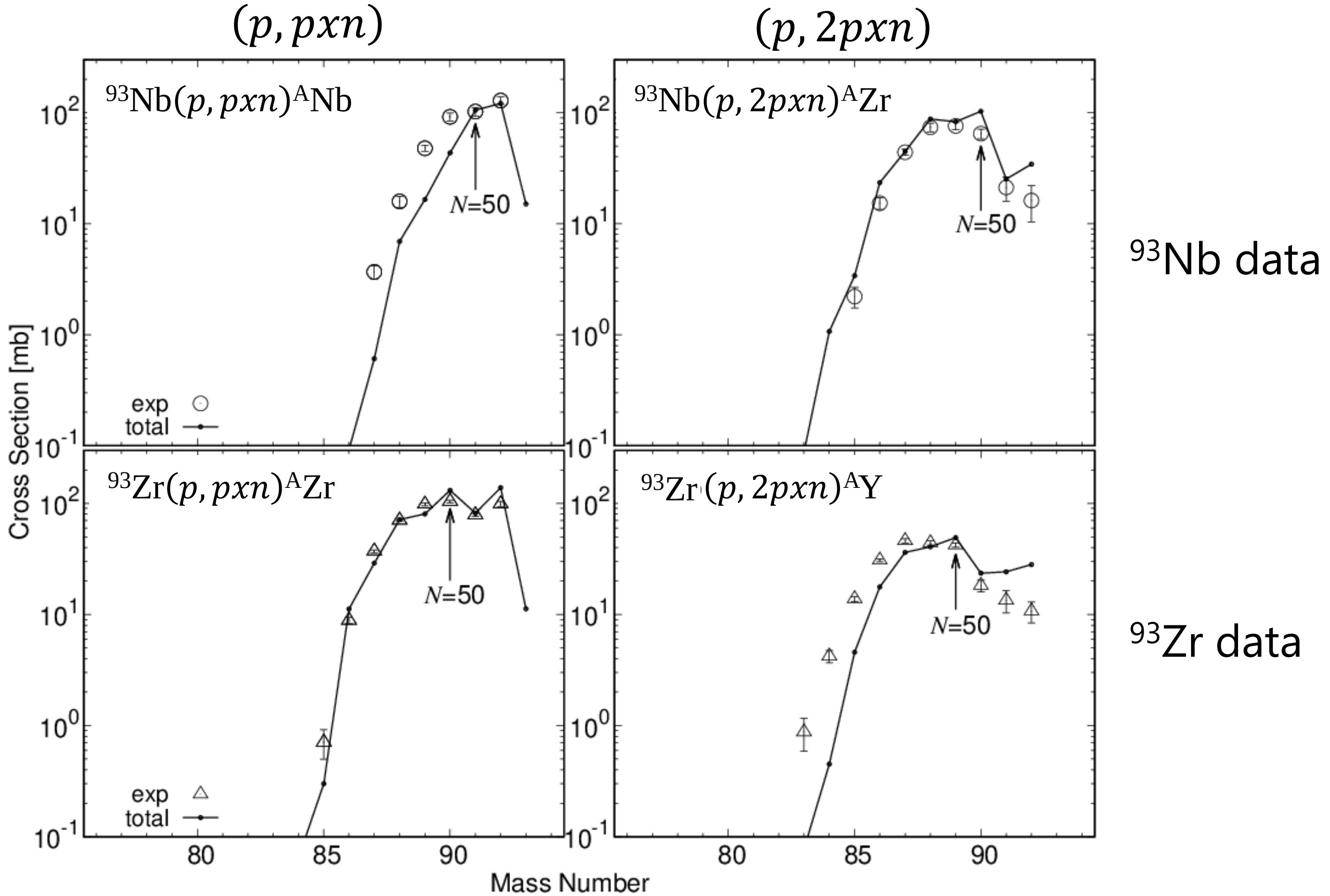
^{93}Nb + p @ 113 MeV



^{93}Nb + d @ 113 MeV/u

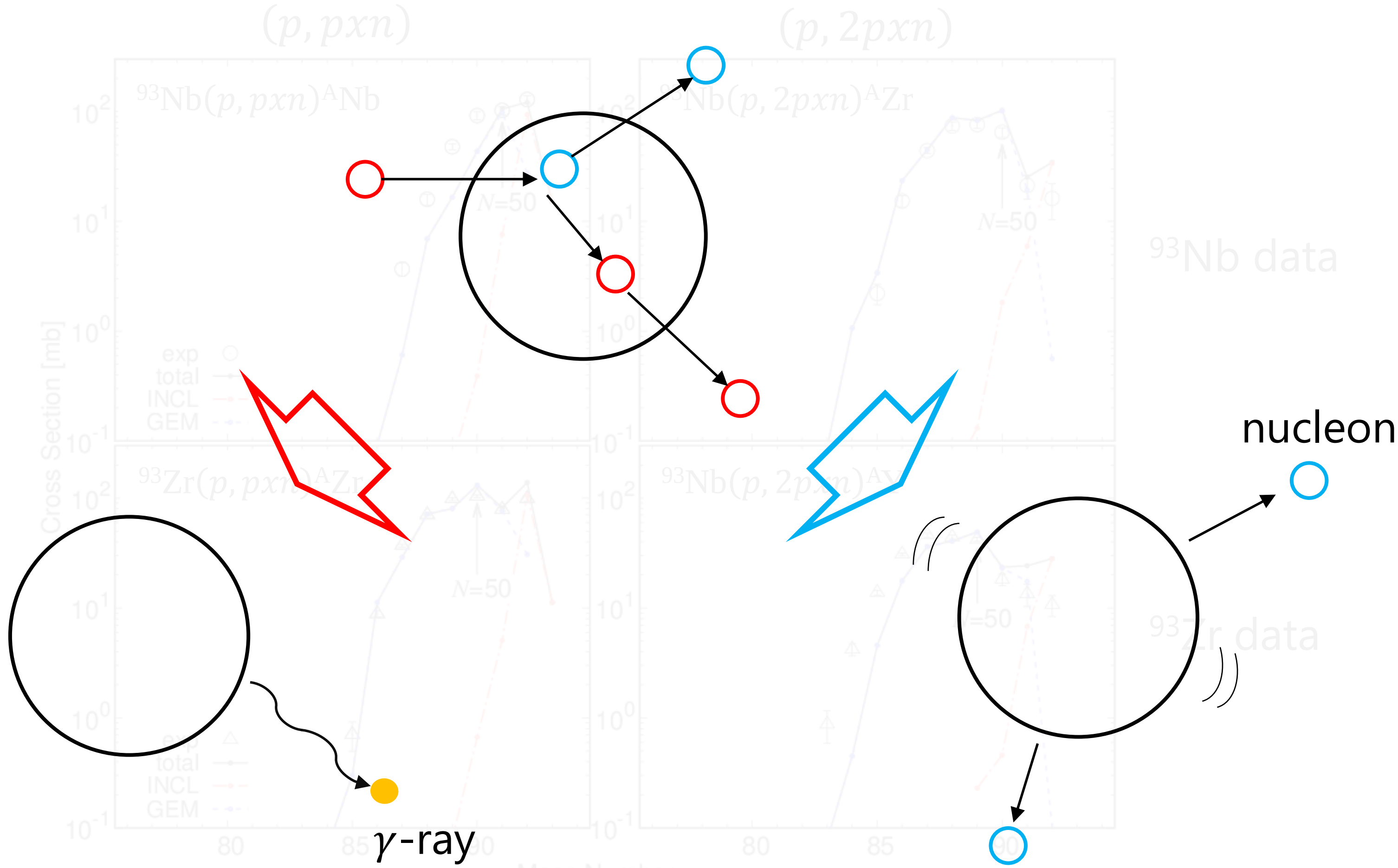
- Exaggerated even-odd staggering both along Z and N are clearly seen in C/E plot in chart of the nuclides

Results | Comparison with ^{93}Zr data (p-induced reactions)



- Jumps are seen at $N = 50$ except for ^{93}Nb$(p, pxn)^{A}$Nb reactions.
- PHITS calculations show generally good agreement with experimental data
  → discuss the reason why the jump disappears on the basis of calculation

Results | Comparison with ^{93}Zr data (p-induced reactions)

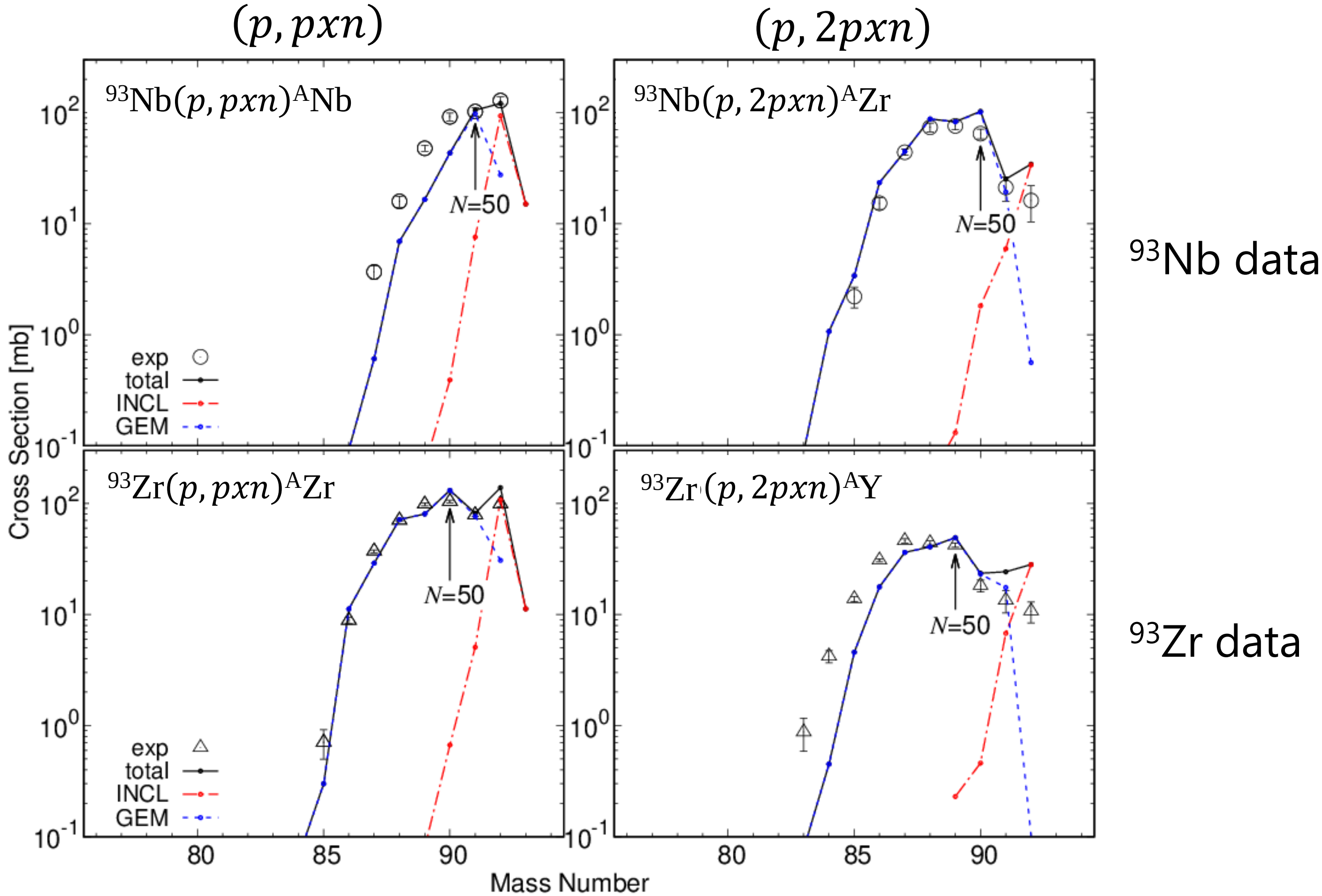


Pre-fragment = reaction product

Pre-fragment ≠ reaction product

- Cross sections by PHITS are decomposed into two components:
  INCL: direct production yield via INC process
  GEM: production by particle evaporation from highly excited pre-fragments

Results | Comparison with ^{93}Zr data (p-induced reactions)



- Cross sections by PHITS are decomposed into two components:
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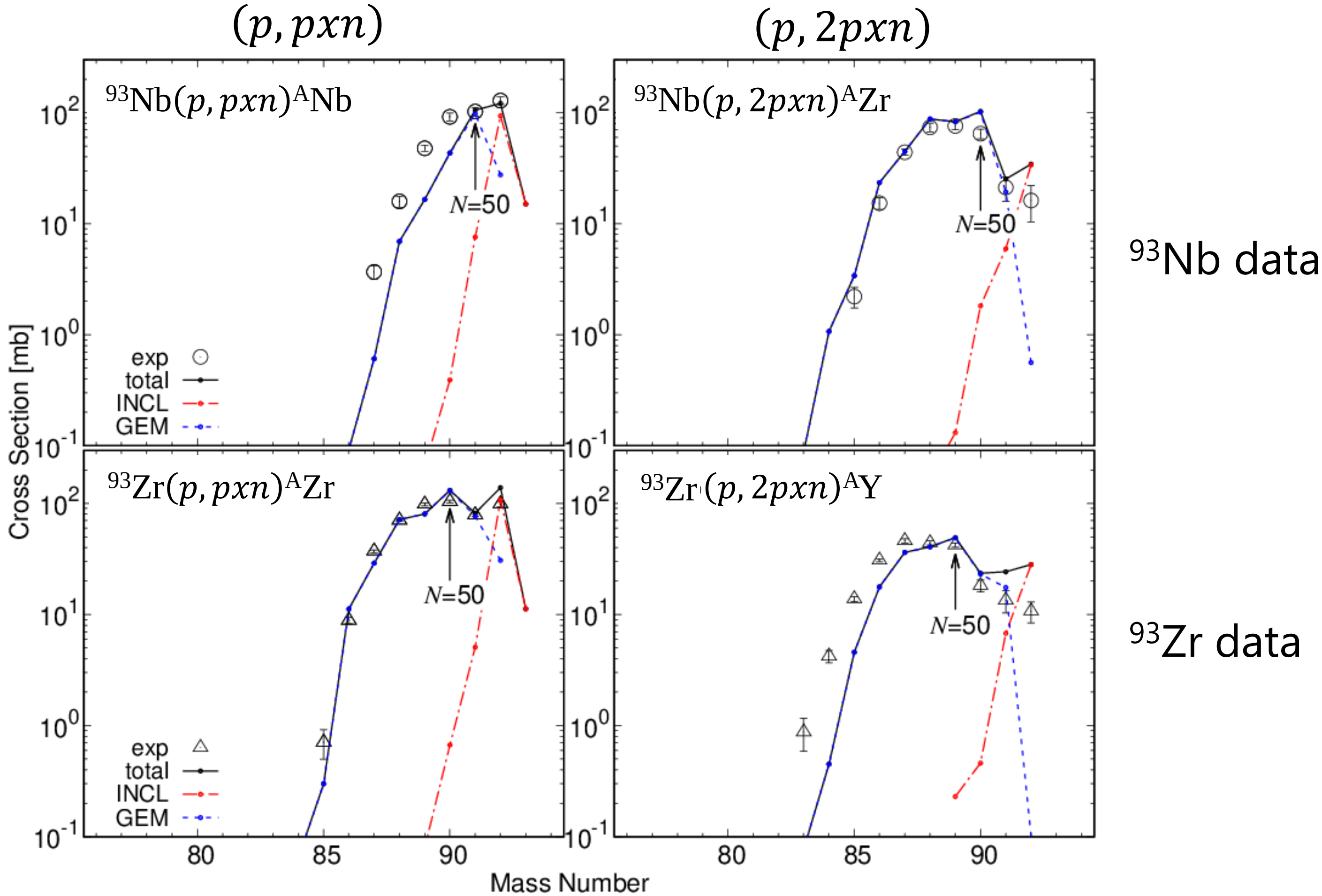
Results | Comparison with ^{93}Zr data (p-induced reactions)



- INCL: maximum values at $A = 92$
- GEM: maximum values at $N = 50$ and jumps appears in all the panels

  → ^{93}Nb$(p, pxn)^{A}$Nb: jump by GEM is smeared out by INCL

  others: INCL DO NOT disturb the jumps seen in GEM components

Contents

Summary

- Isotope-production cross sections for proton- and deuteron-induced reactions on ^{93}Zr and ^{93}Nb were obtained at RIKEN RIBF using inverse kinematics method.

- The calculations by PHITS reproduce the measured data generally well.
- But, further improvement of theoretical models is needed:
  - underestimation in n-deficient region in odd-Z
  - overestimation in isotope near the target nucleus
  - Exaggerated even-odd staggering

- The magic number is reflected in the isotope-production cross sections.
- The appearance of the jump at $N = 50$ depends on the relative fractions of the INC and evaporation components.

Collaborators (^{93}Zr + p,d @ 50 MeV/u exp.)

Kyushu University	S. Kawase, K. Nakano, Yu. Watanabe, J. Suwa
RIKEN Nishina Center	H. Wang, H. Otsu, N. Chiga, T. Sumikama, H. Sakurai, D.S. Ahn, H. Baba, S.D. Chen, M.L. Cortes, P. Doornenbal, N. Fukuda, T. Isobe, S. Kubono, I. Murray, H. Sato, Y. Shimizu, P.-A. Söderström, X.H. Sun, D. Suzuki, H. Suzuki, H. Takeda,K. Yoshida
Tokyo Tech.	S. Takeuchi, A. Hirayama, Y. Kondo, T. Nakamura, T. Ozaki, A. Saito, Y. Togano, T. Tomai, H. Yamada, M. Yasuda
University of Miyazaki	S. Kawakami, Y. Maeda, Y. Soudo
University of Tokyo	S. Koyama, M. Niikura, T. Saito
CNS, University of Tokyo	S. Masuoka, S. Michimasa, R. Nakajima, S. Shimoura
Niigata University	K. Chikaato, R. Hosoda, M. Takechi

Collaborators (^{93}Nb + p,d @ 113 MeV/u exp.)

Kyushu University	S. Kawase, K. Nakano, Yu. Watanabe, S. Araki, T. Kin
RIKEN Nishina Center	H. Wang, H. Otsu, H. Sakurai, D.S. Ahn, S. Chen, N. Chiga, P. Doornenbal, N. Fukuda, T. Isobe, T. Kubo, S. Kubono, M. Kurokawa, T. Matsuzaki, Y. Shimizu, T. Sumikama, P.-A. Söderström, H. Suzuki, H. Takeda, Ya. Watanabe, K. Yoshida
Tokyo Tech.	S. Takeuchi, Y. Togano, T. Nakamura, Y. Kondo, T. Ozaki, A. Saito, M. Shikata, J. Tsubota
University of Miyazaki	Y. Maeda, S. Kawakami, T. Yamamoto
University of Tokyo	S. Koyama, S. Momiyama, S. Nagamine, M. Niikura, T. Saito, K. Wimmer
CNS, University of Tokyo	M. Matsushita, S. Michimasa, S. Shimoura
Hokkaido University	M. Aikawa, A. Makinaga
Rikkyo University	Y. Shiga