Spallation reaction study for long-lived fission products in nuclear waste: Cross section measurement for ^{137}Cs and ^{90}Sr on proton and deuteron

He Wang

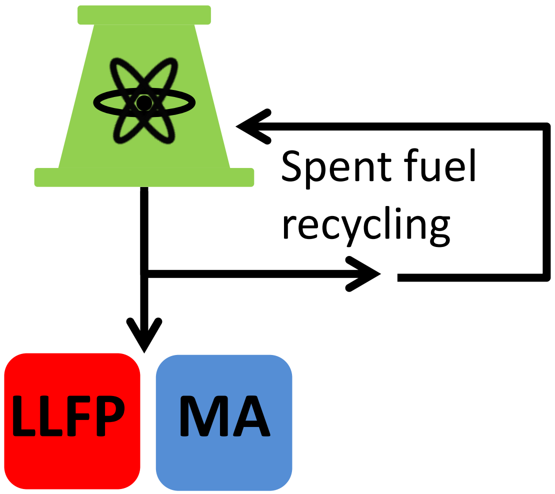
RIKEN Nishina Center

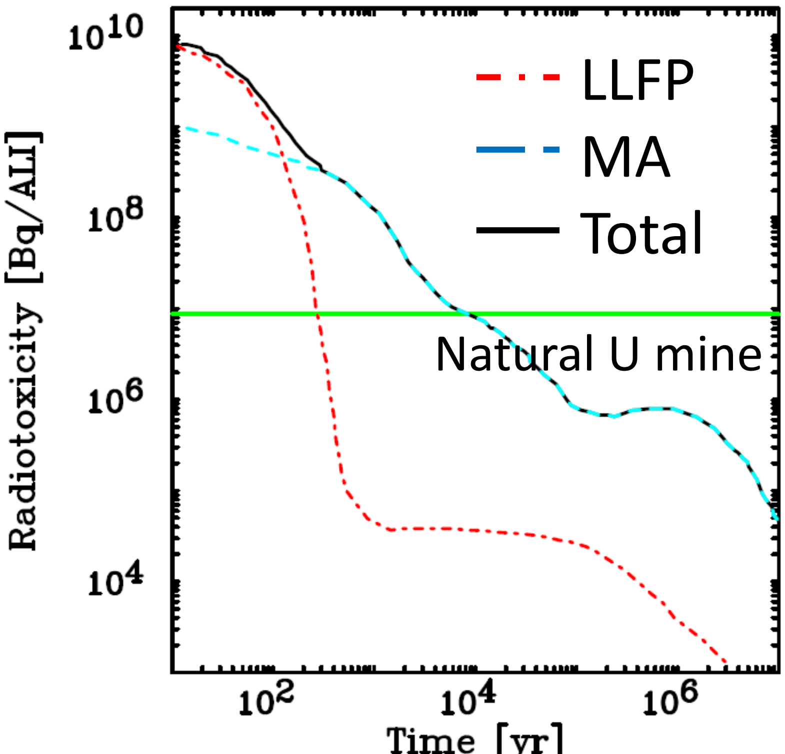
RIBF seminar, RIBF hall, May 10, 2016

Content

- Motivation
- Experiment details
- Results and discussion
- Summary and perspective

Motivation





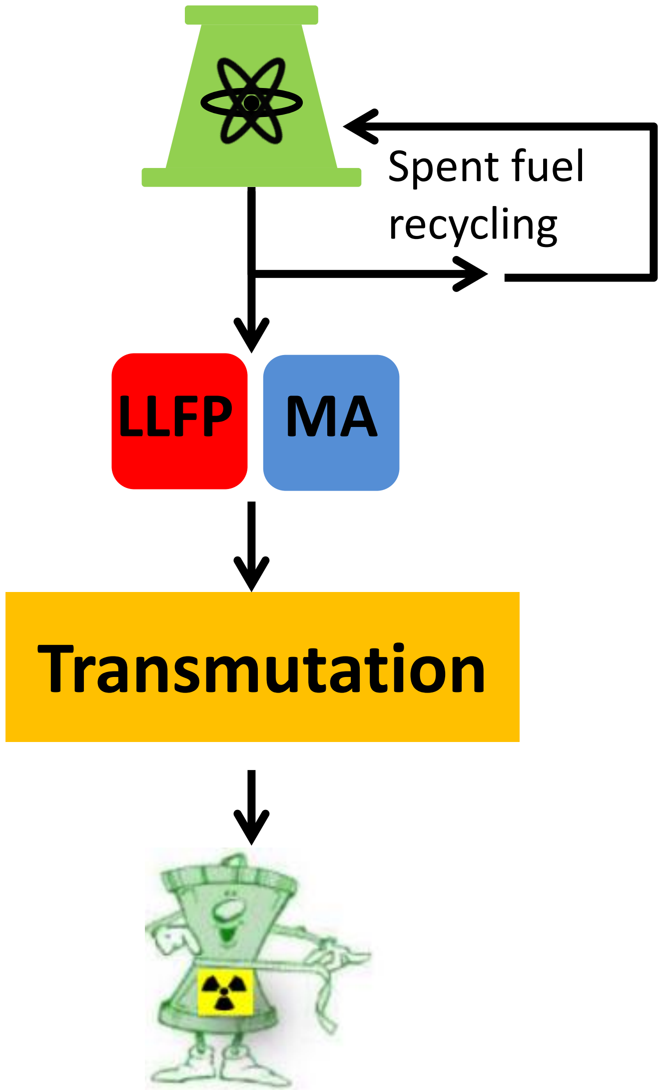
High-level radioactive waste

- Long-lived fission products
  e.g. ^{137}Cs, ^{90}Sr, ^{107}Pd, ^{93}Zr…
- Minor Actinide
  e.g. $^{241,243}Am$, ^{237}Np…

Nuclide	Half-life (year)	Mass (per 1tHM)
Se-79	0.3×10^6	6g
Sr-90	28.8	0.6kg
Zr-93	1.53×10^6	1kg
Tc-99	0.21×10^6	1kg
Pd-107	6.5×10^6	0.3kg
Sn-126	0.23×10^6	30g
I-129	15.7×10^6	0.2kg
Cs-135	2.3×10^6	0.5kg
Cs-137	30.1	1.5kg

Oigawa, CNS summer school, 2016

Partitioning and transmutation (P&T)



Partitioning

-- MA

-- LLFP

  -- Heat generator (Sr, Cs)

  -- Rare metal (Pd)

Transmutation

High-level radioactive waste

→ stable/short-lived isotopes

To minimize the long-term dose

Nuclear Transmutation

Transmutation in Alchemy

- Philosopher's stone
- Metal into Gold

Natural nuclear transmutation

- α-decay, β-decay

Artificial nuclear transmutation

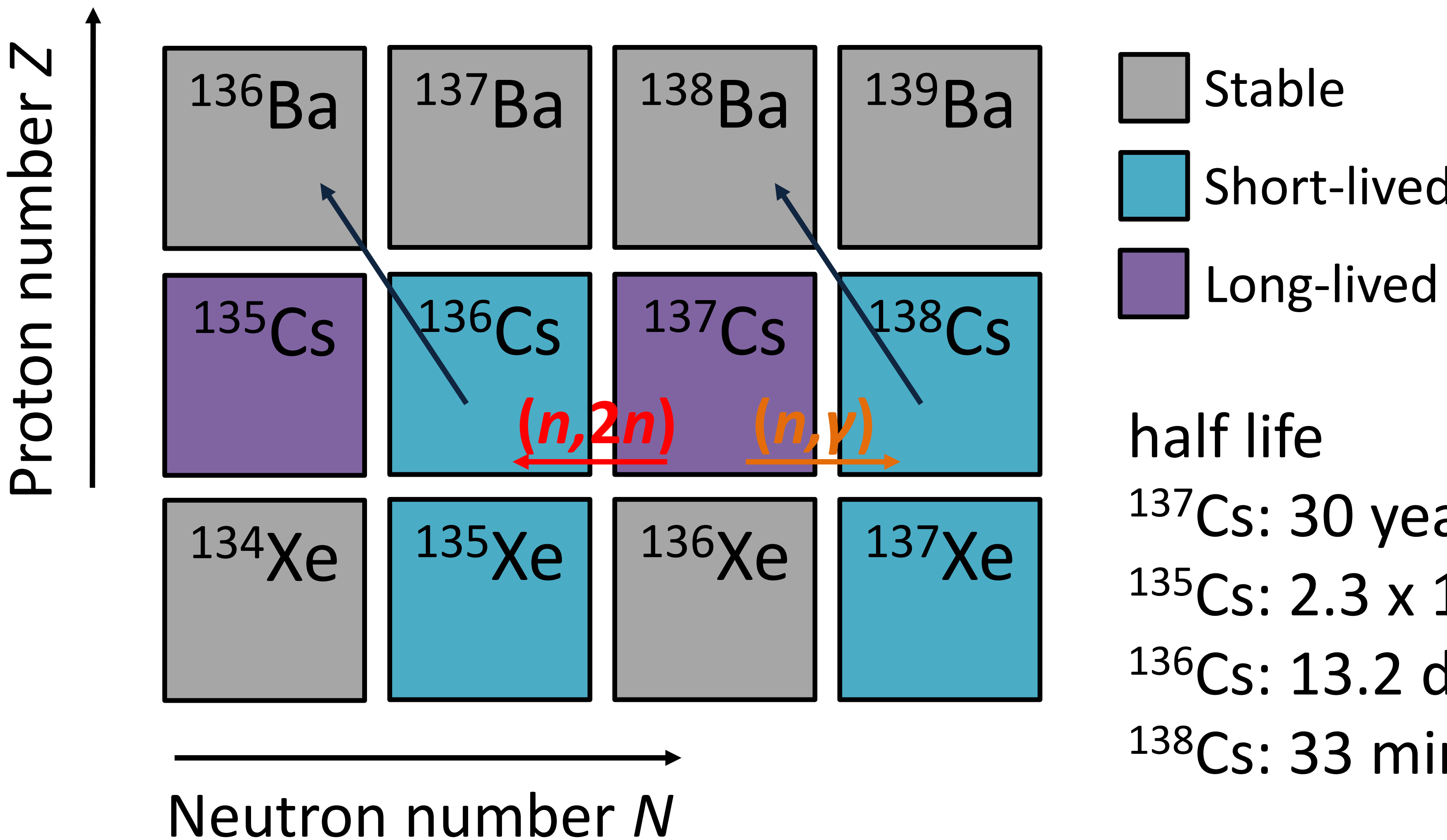
- Reactions in reactor

One chemical element/isotope into another

Possible reaction for the LLFP transmutation

(n,γ): Neutron capture reaction

($n,2n$): Neutron knockout reaction



half life

^{137}Cs: 30 years

^{135}Cs: 2.3 x 10^6 years

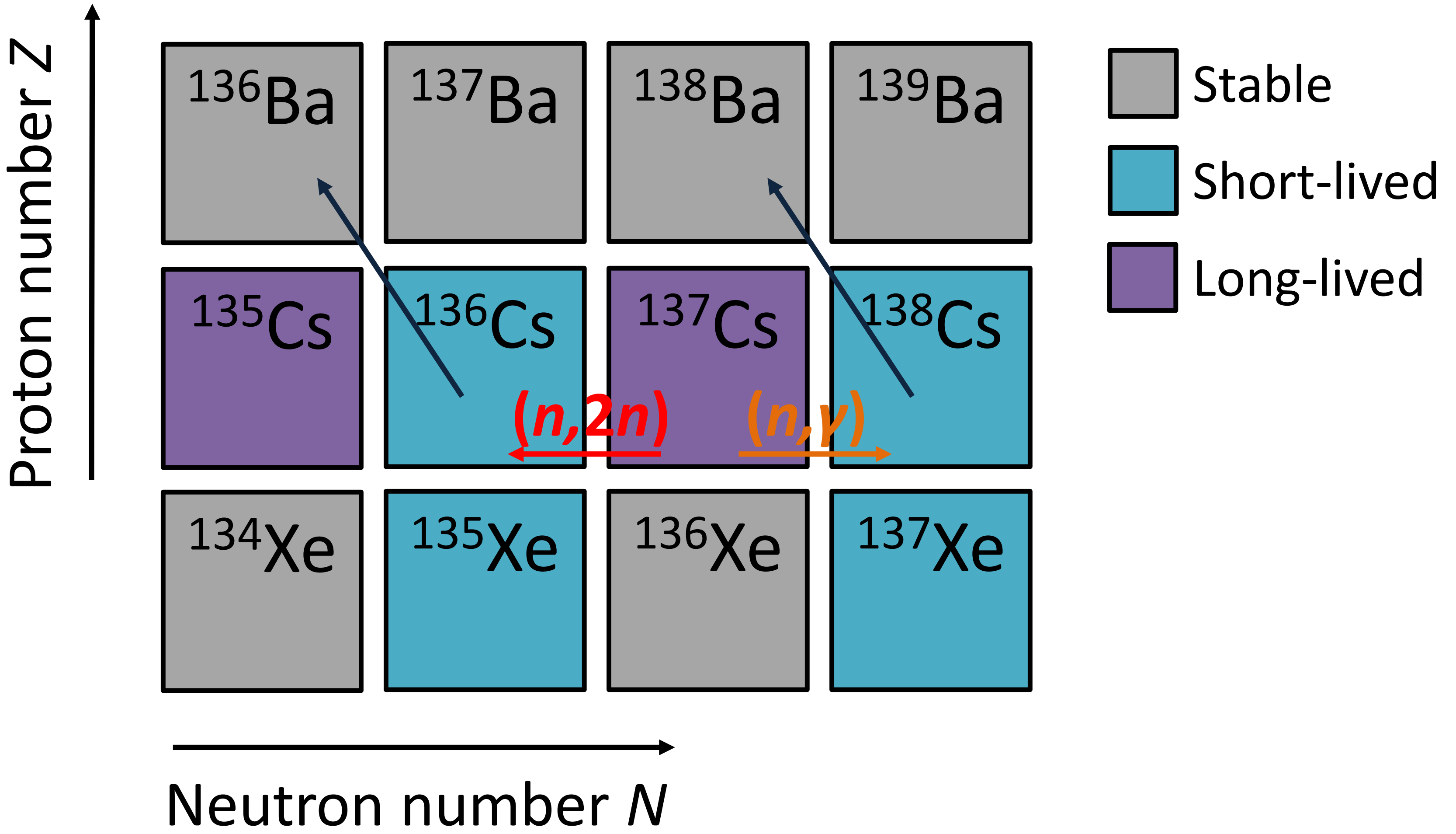
^{136}Cs: 13.2 days

^{138}Cs: 33 minutes

Possible reaction for the LLFP transmutation

(n,γ): limited study

(n,2n): no data

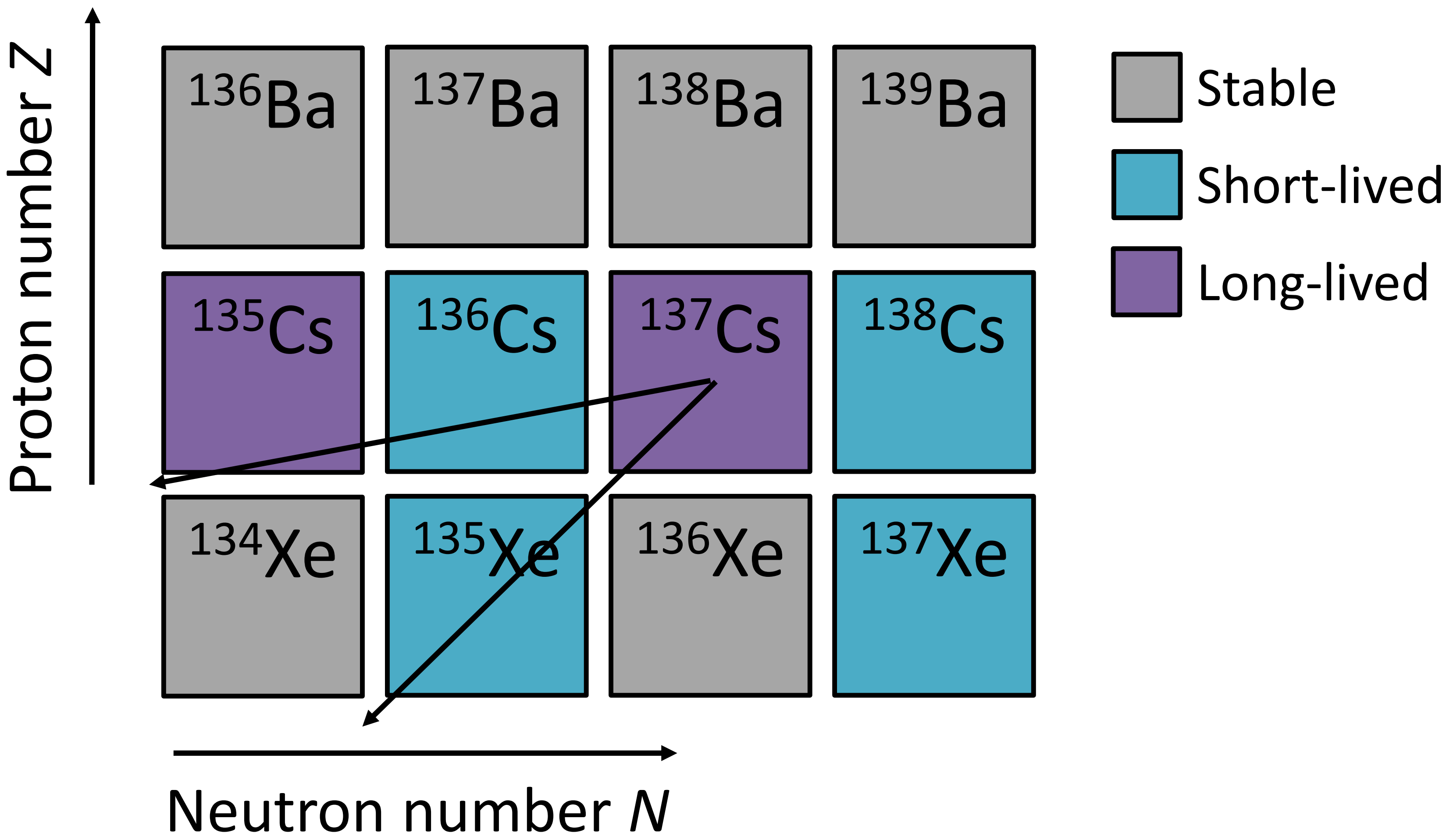


Insufficient reaction study for LLFP

Possible reaction for the LLFP transmutation

Proton, deuteron induced reaction

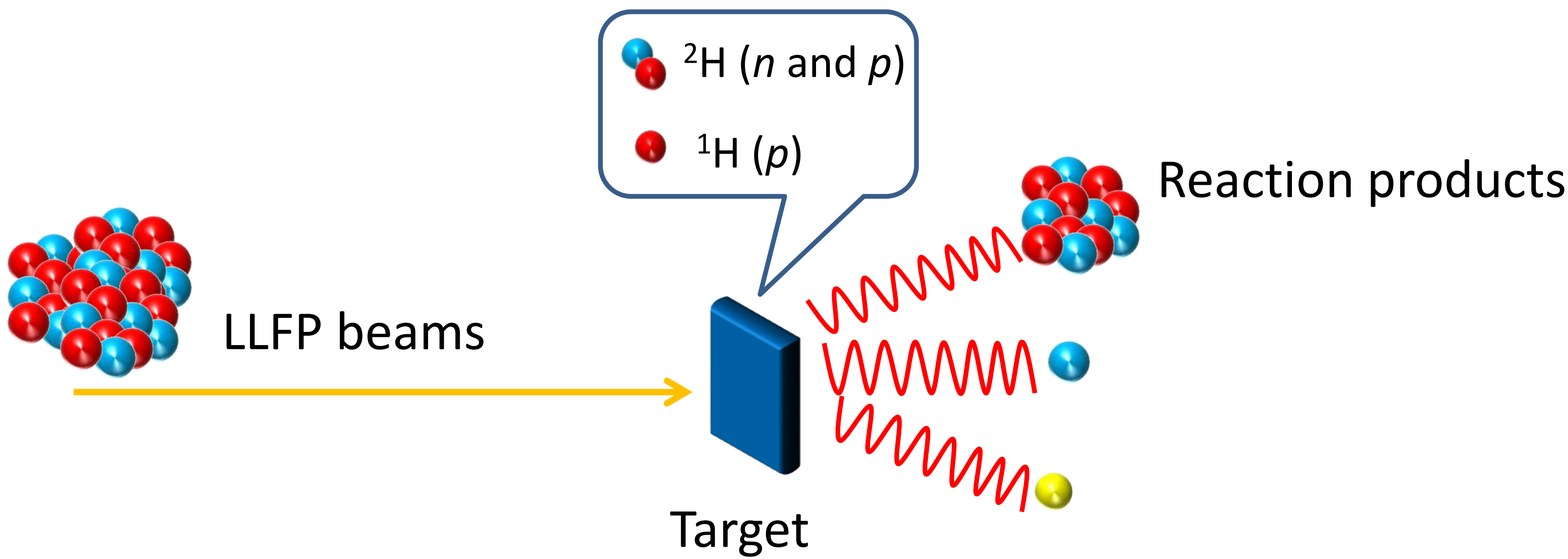
$p/d + {}^{137}Cs \rightarrow$



Reaction study for LLFP at RIBF

Cross section measurements

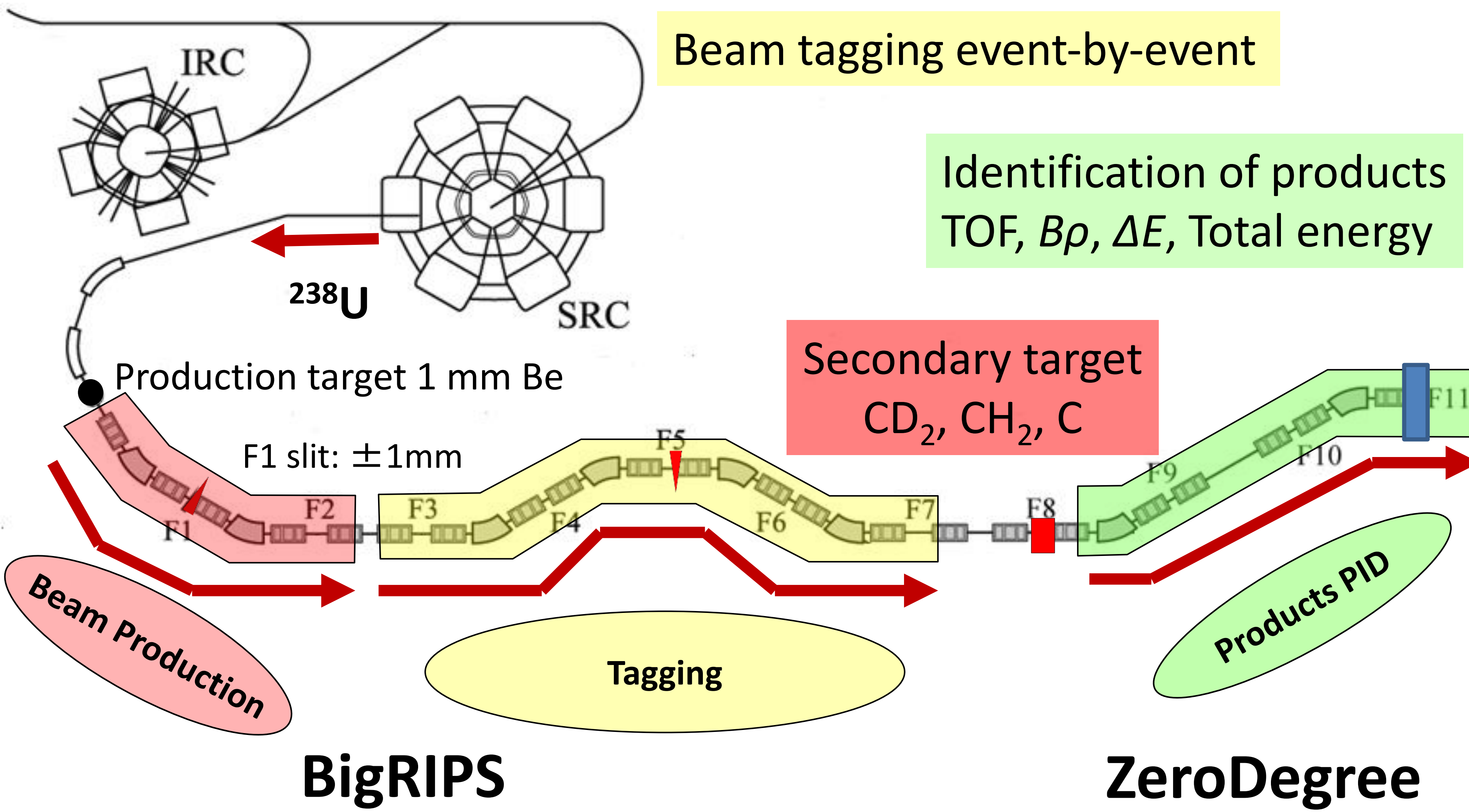
- Inverse kinematics
- Proton and deuteron → information on neutron
  -- Difficulty to make neutron target
- Different reaction energy
  -- 200 MeV/*u* as starting point



Experimental setup

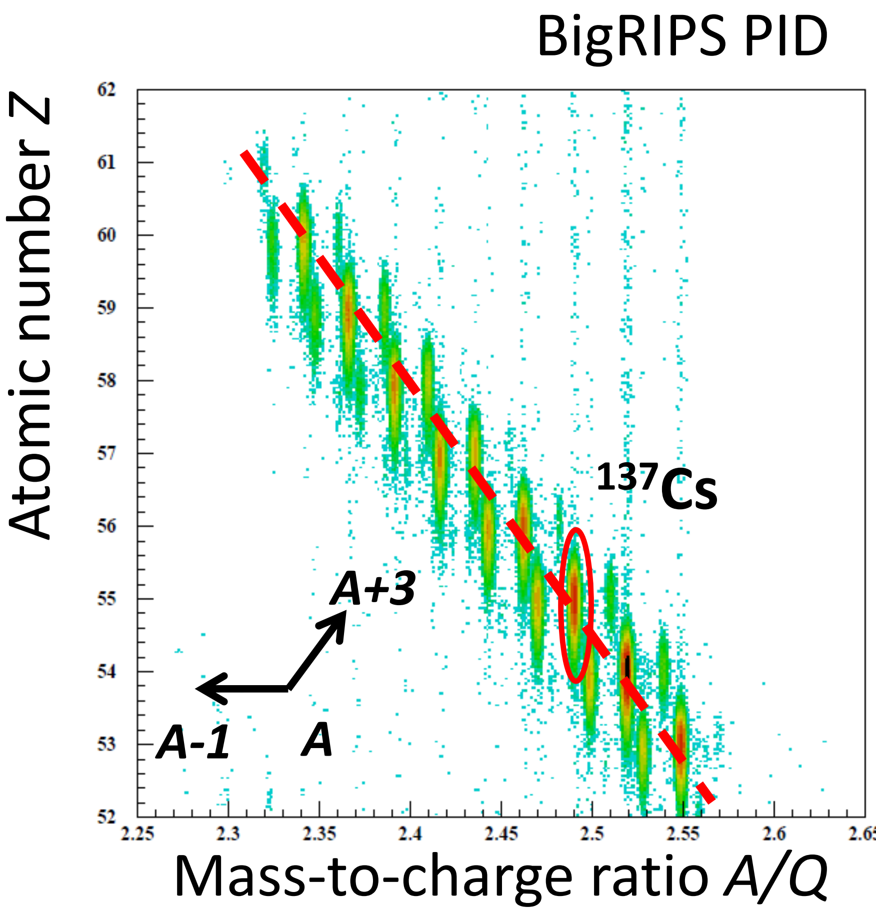
^{137}Cs and ^{90}Sr beams production

Beam energy: 185MeV/*u*



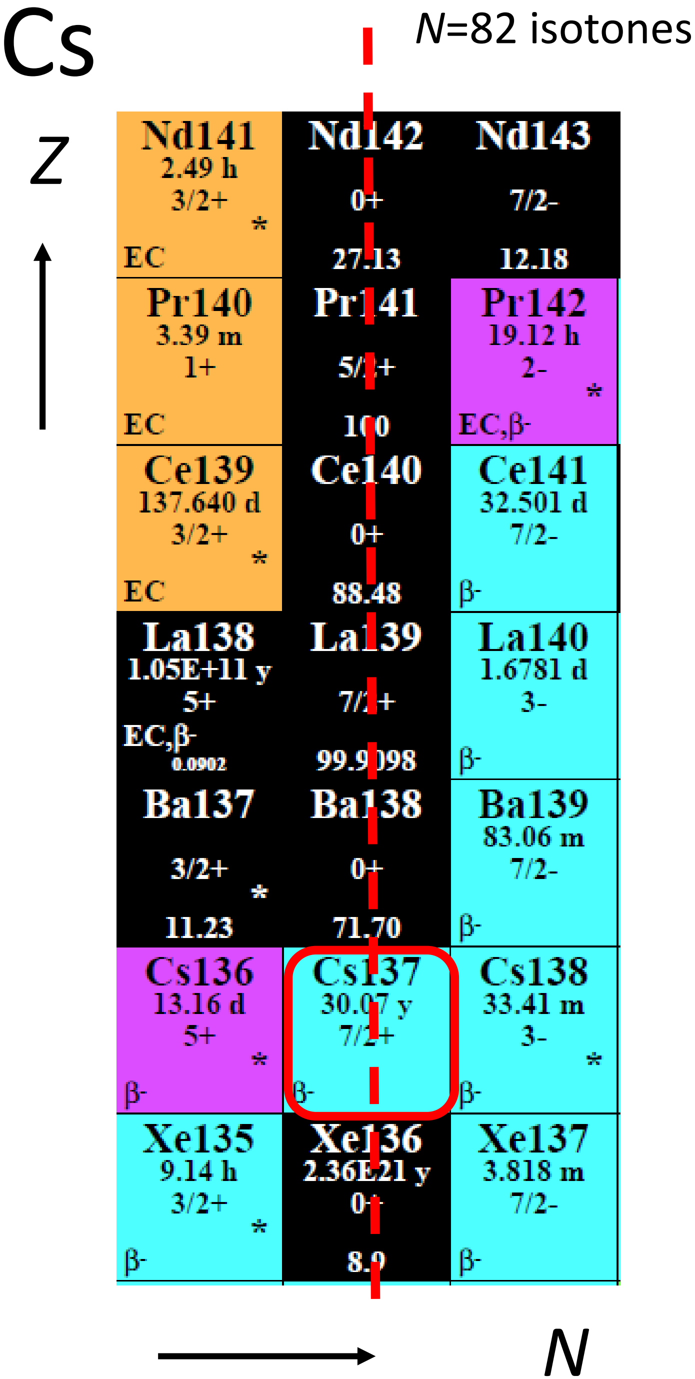
Particle identification for ^{137}Cs

^{137}Cs setting



A/Q resolution=3.43 x 10^{-3} (FWHM)

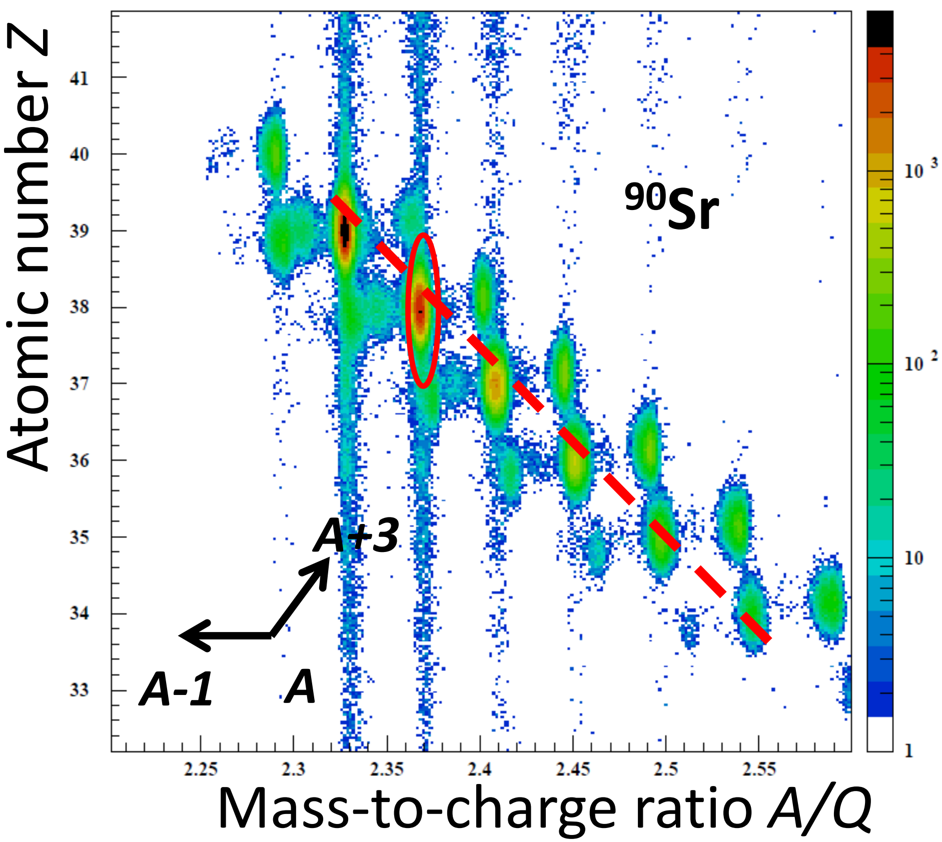
Z resolution=0.55 (FWHM)

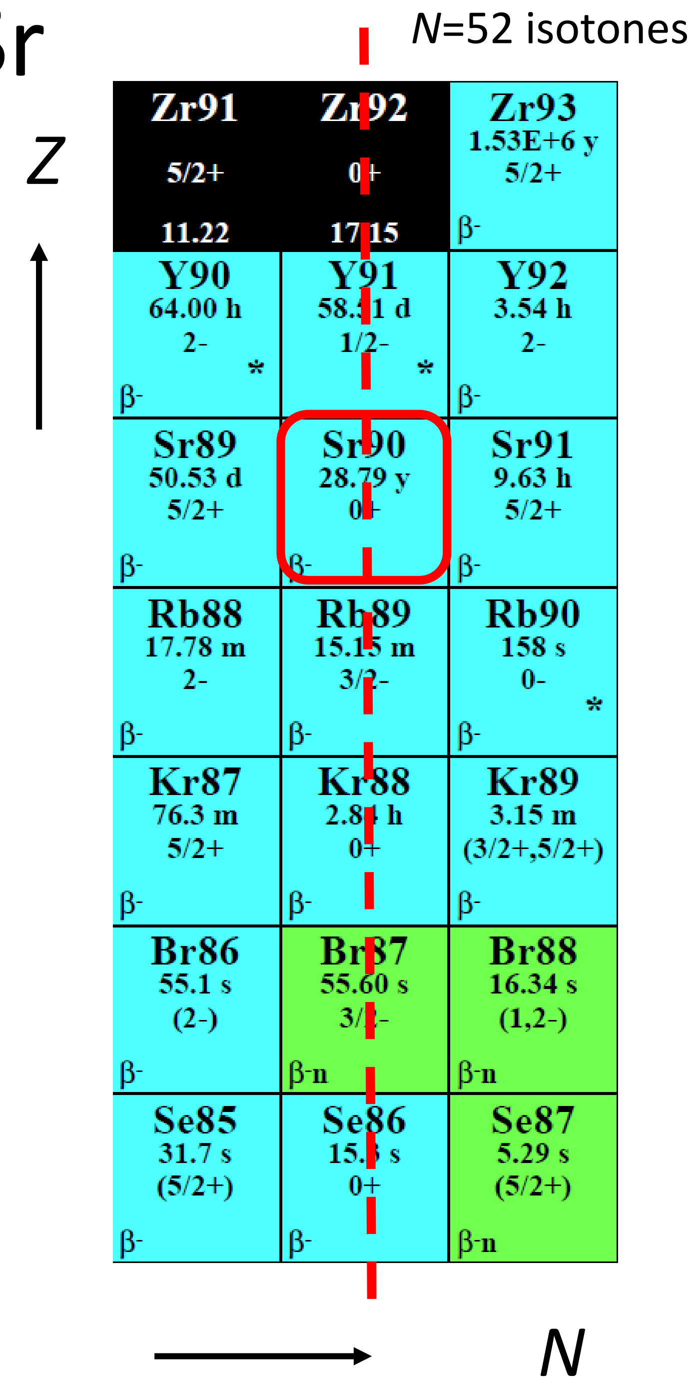


Particle identification for ^{90}Sr

^{90}Sr setting

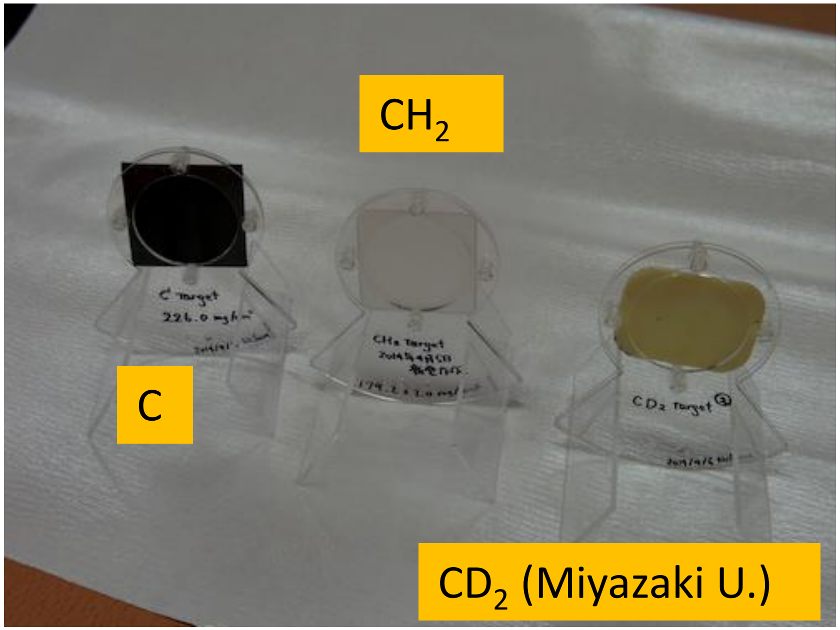
BigRIPS PID

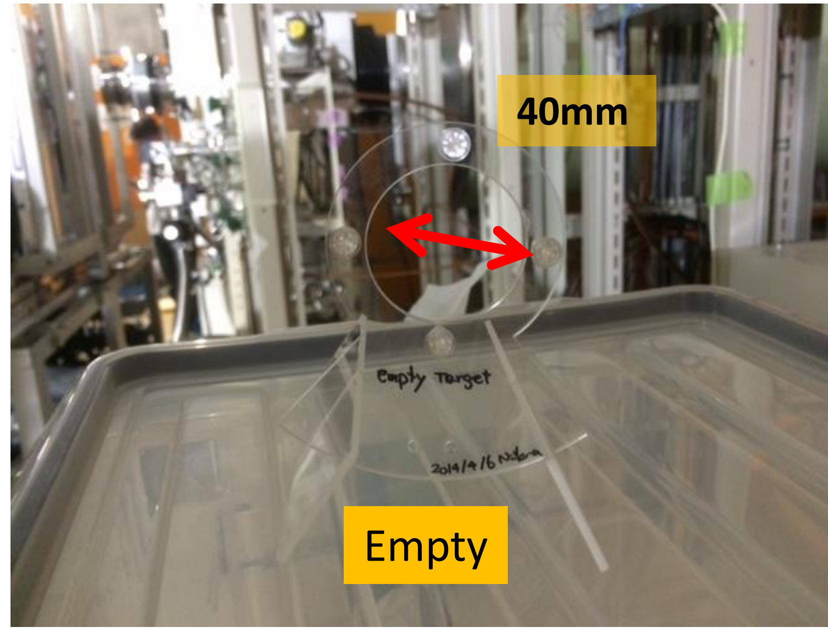




Experimental setup

Secondary targets





3 targets: C, CH_2, CD_2 + empty

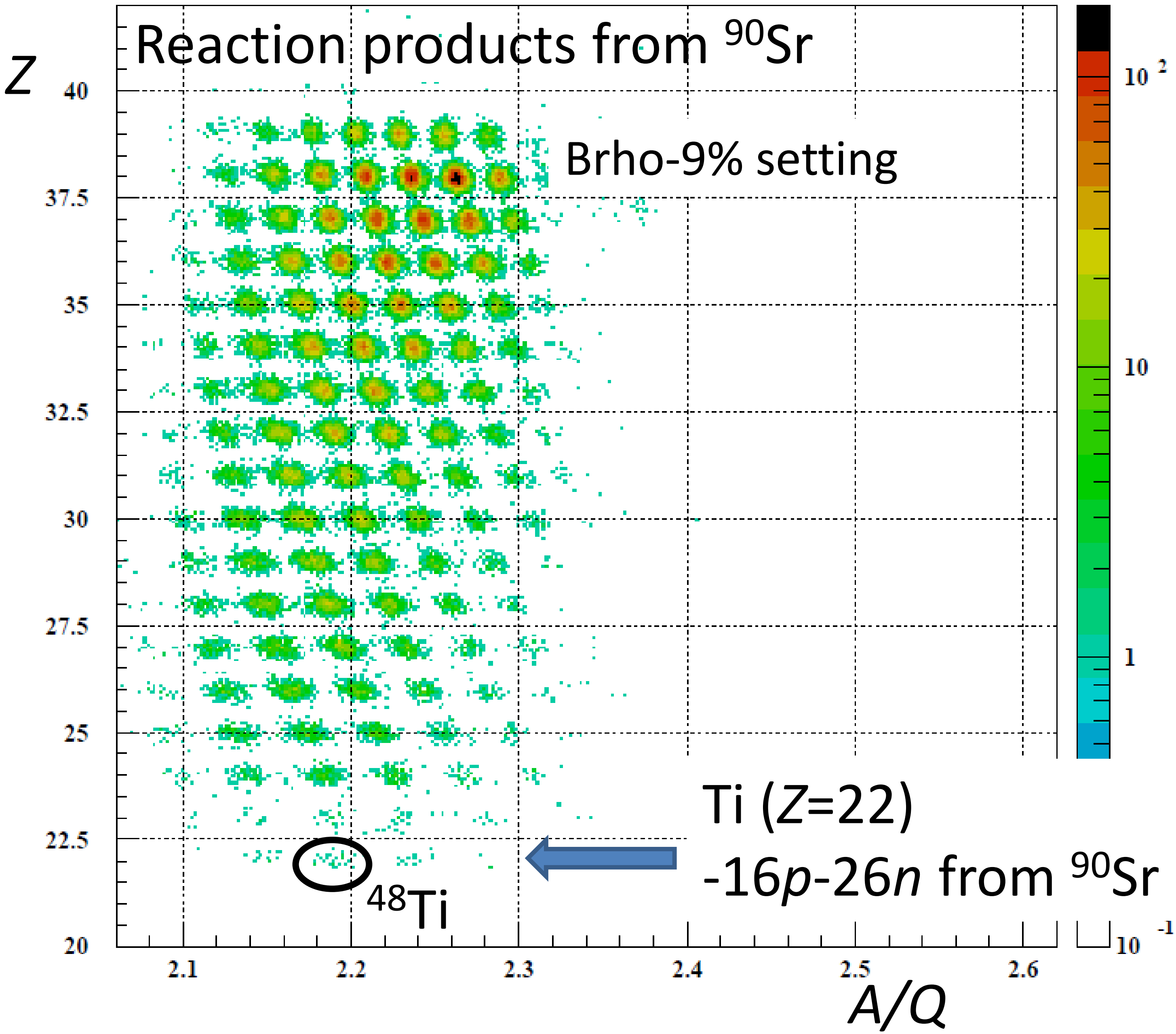
ZeroDegree spectrometer

Mode	*p* res.	*p* acce.	Ang. acce.
Large acceptance	1240	6%	90mrad(H)x60mrad(V)

5 brho settings:

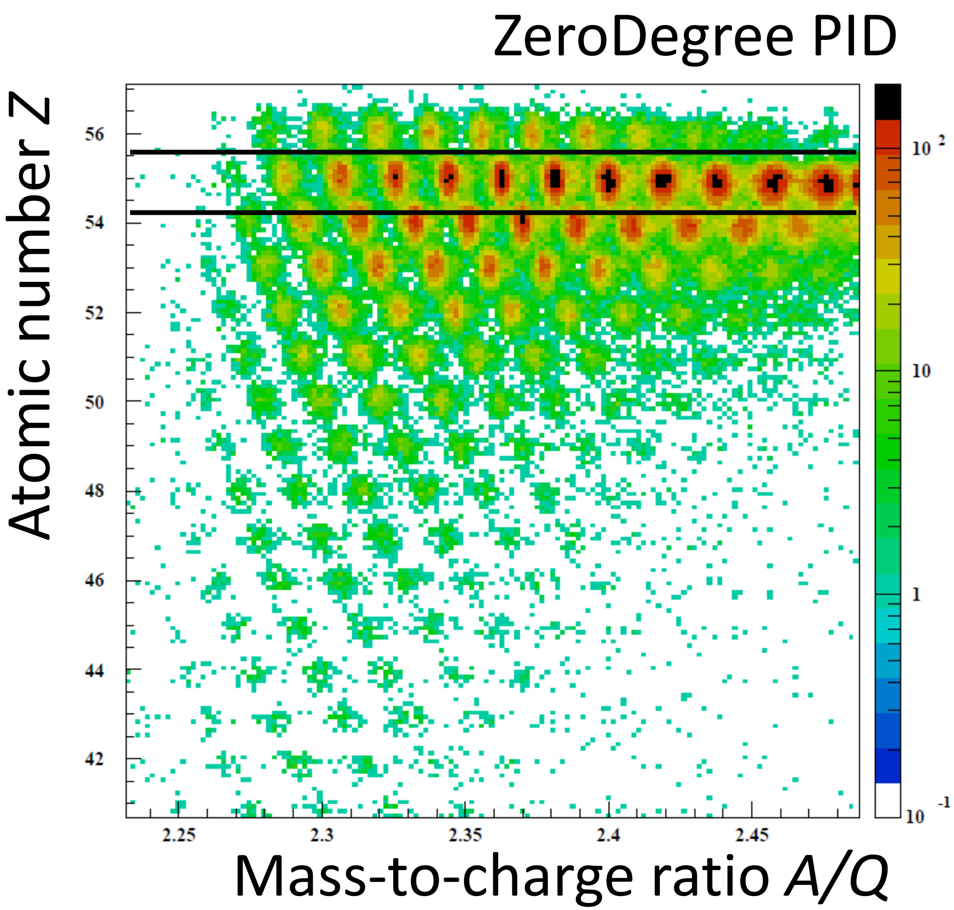
+3%, 0%(≡Brho of secondary beam), -3%, -6%, -9%

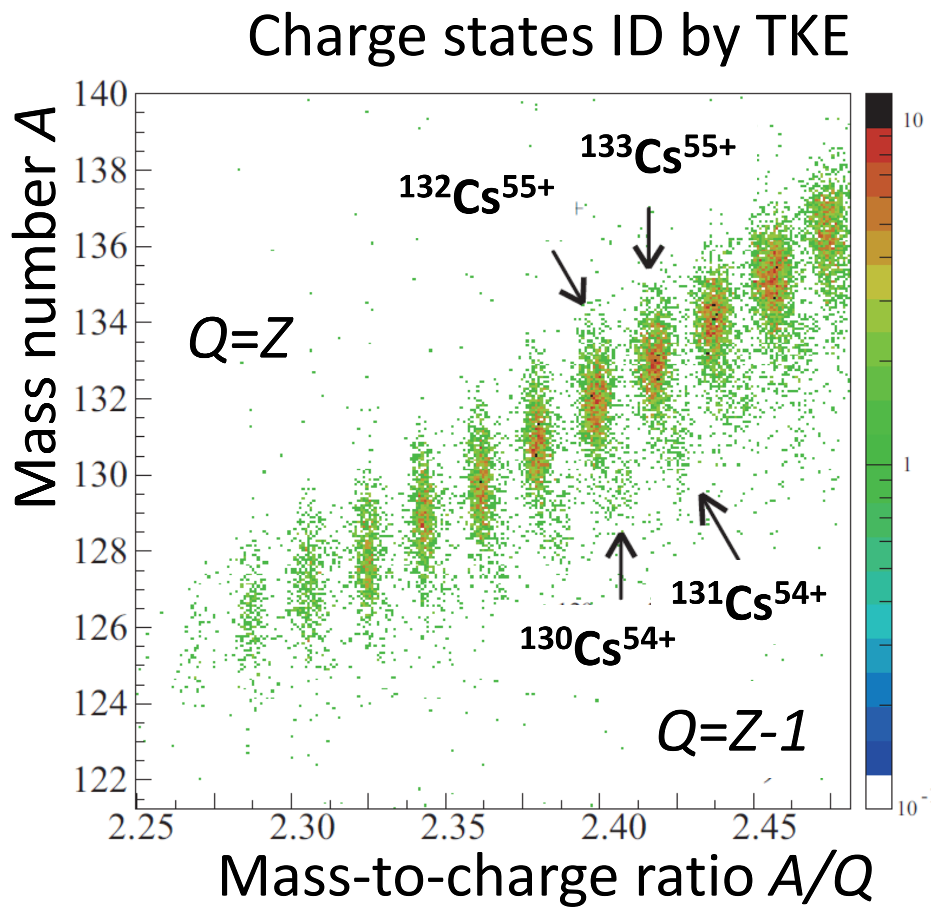
Particle identification



Charge states identification

Reaction products from ^{137}Cs

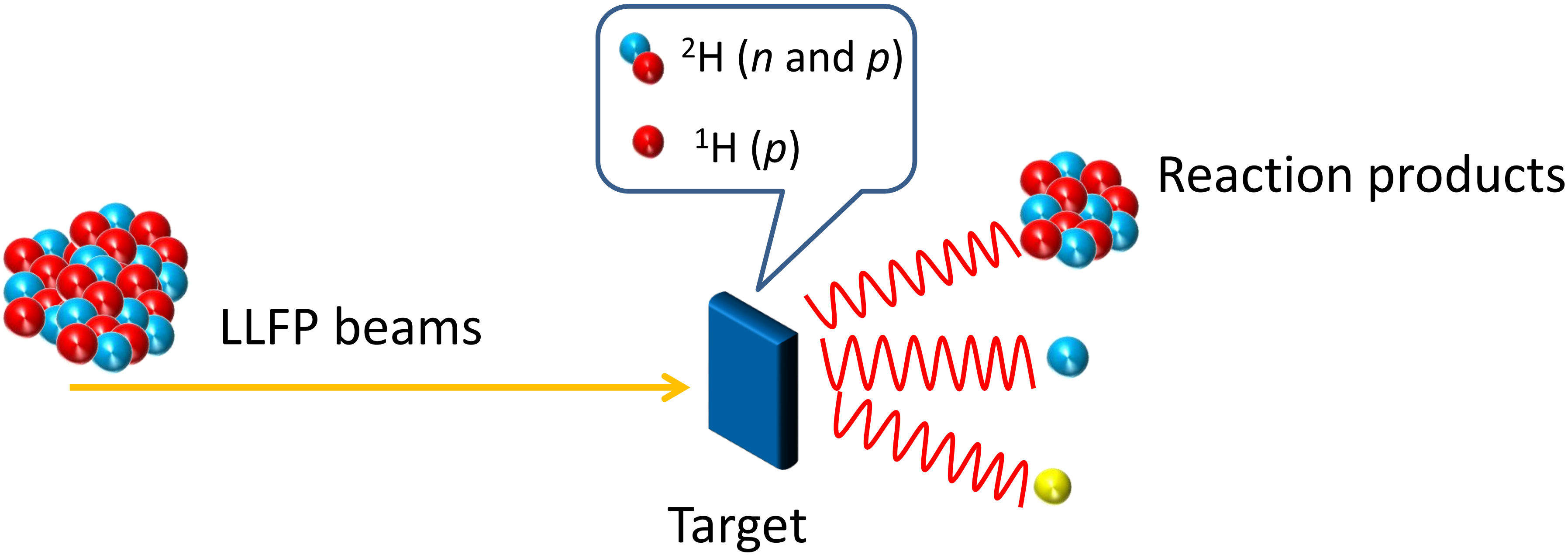




A/Q resolution=4.8 x 10^{-3} (FWHM)

Z resolution=0.47 (FWHM)

Inverse kinematics

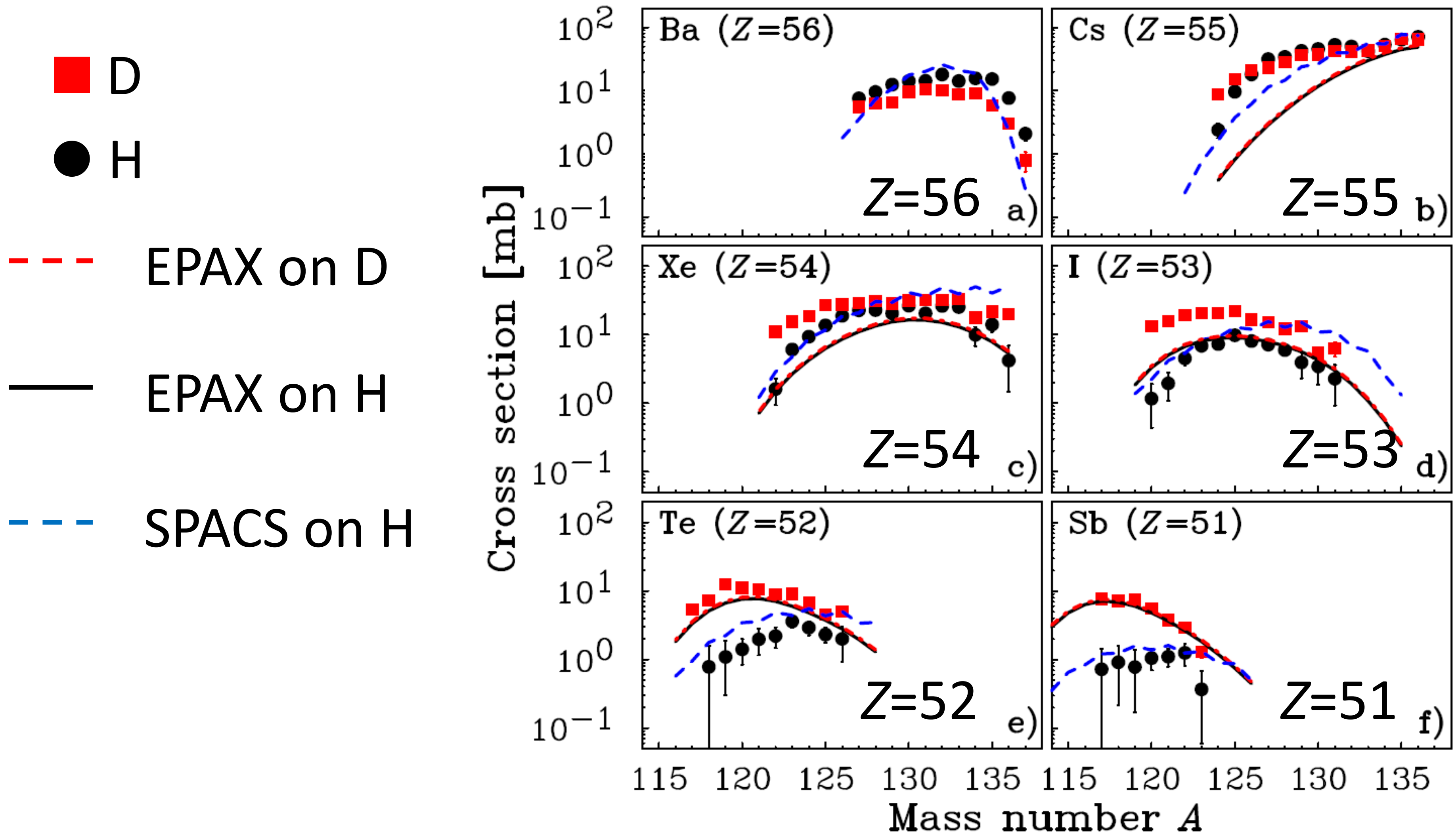


$$\sigma = \frac{Yields}{N_{beam} \times n_{target}}$$

- Cross section on proton: $\sigma_p = (\sigma_{CH2} - \sigma_C)/2$
- Cross section on deuterium: $\sigma_d = (\sigma_{CD2} - \sigma_C)/2$

Comparison with EPAX and SPACS

SPACS: Empirical parameterization for proton/neutron-induced spallation reactions



C. Schmitt, K.-H. Schmidt, and A. Kelic-Heil, Phys. Rev. C **90**, 064605 (2014)

Comparison with PHITS

PHITS: Particle and Heavy Ion Transport code System

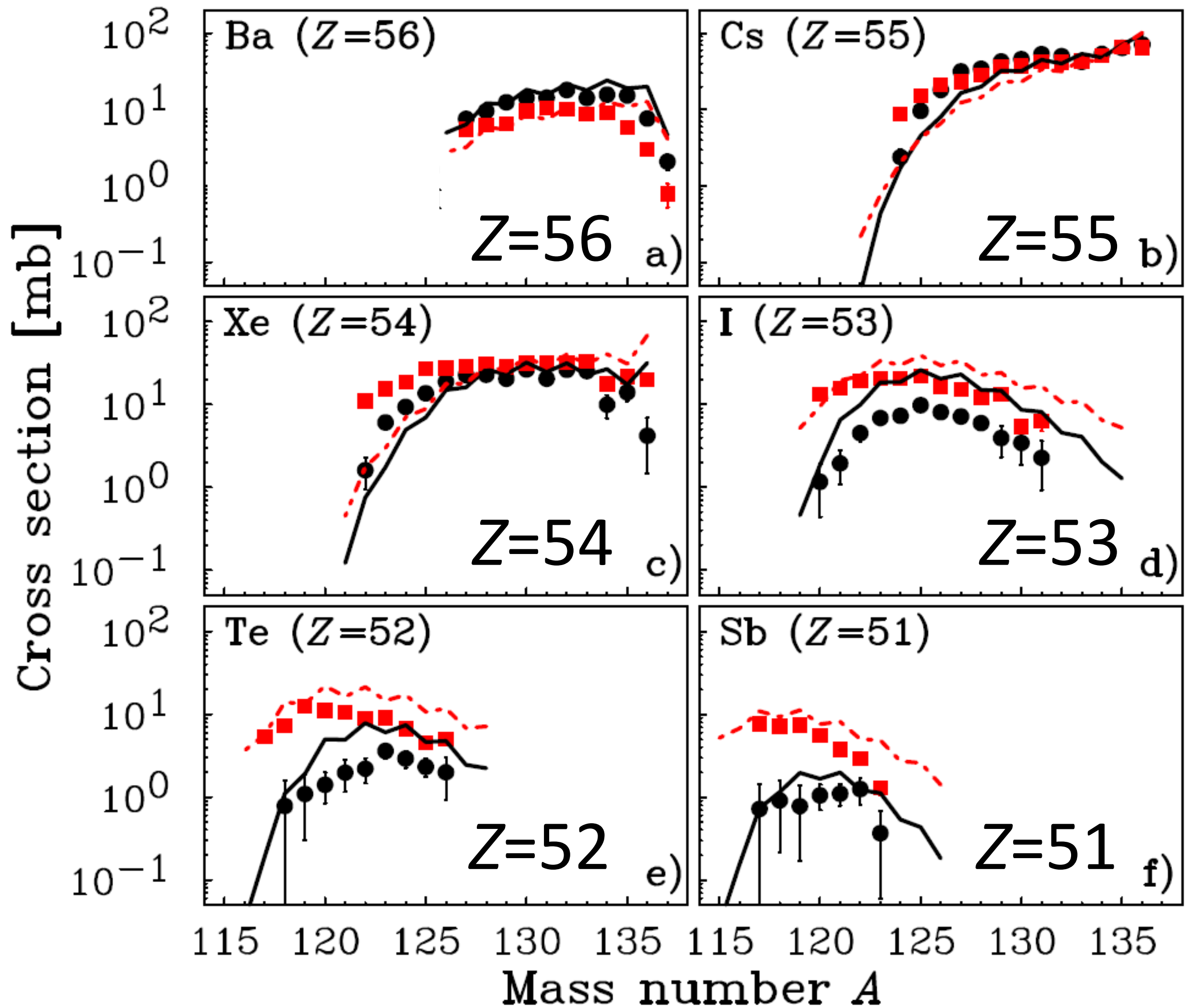
■ D

● H

- - - PHITS on D

—— PHITS on H

Intra-nuclear cascade (INCL)
+
evaporation (GEM)

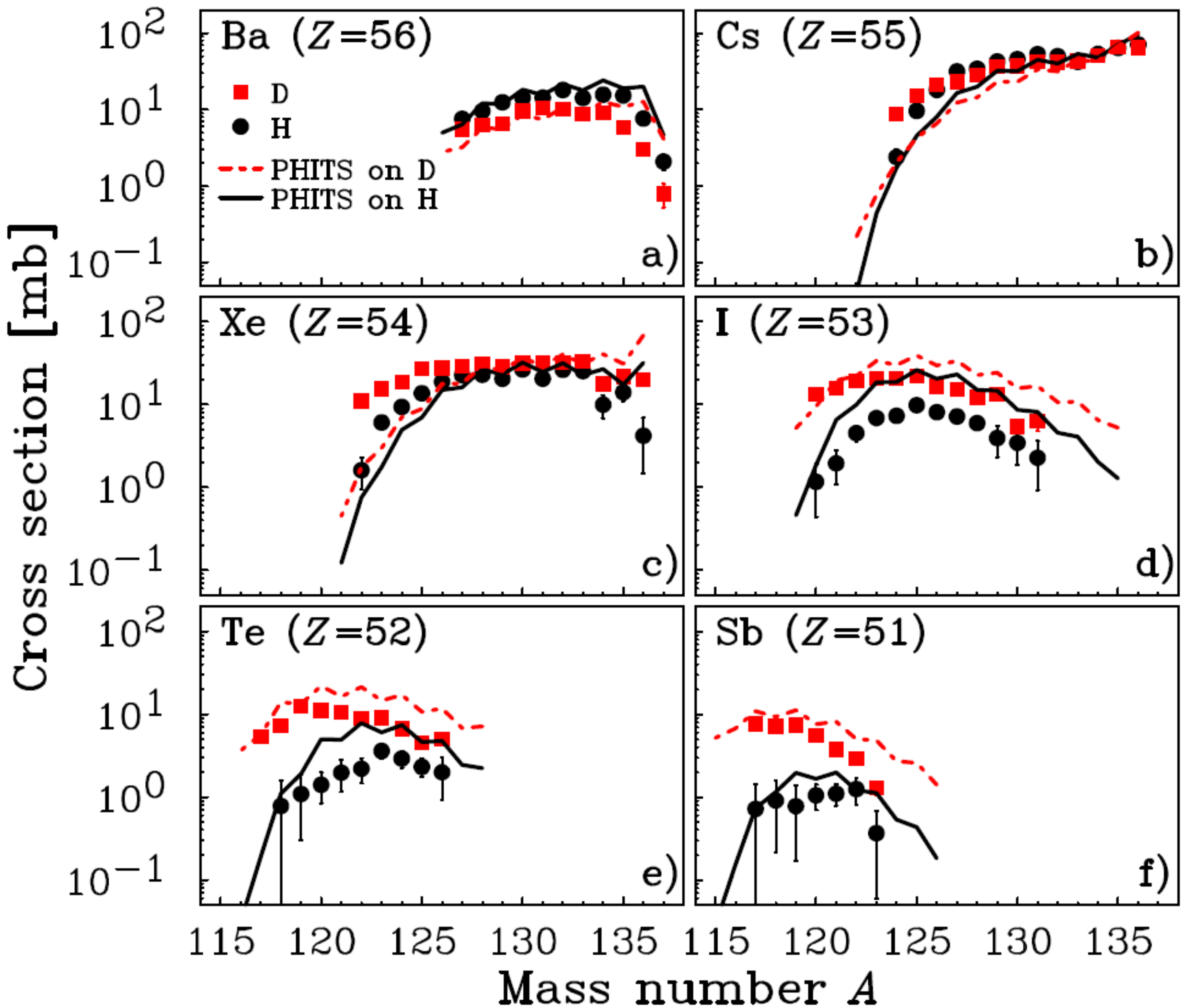


INCL: Boudard et al., Phys. Rev. C 66 (2002) 044615
GEM: Furihata, NIM B 171 (2000) 251

PHITS calculations are provided by Prof. Watanabe's group, Kyushu Univ.

Comparison with PHITS

Overestimation on the magnitude of odd-even staggering



Cross sections for ^{90}Sr on H and D

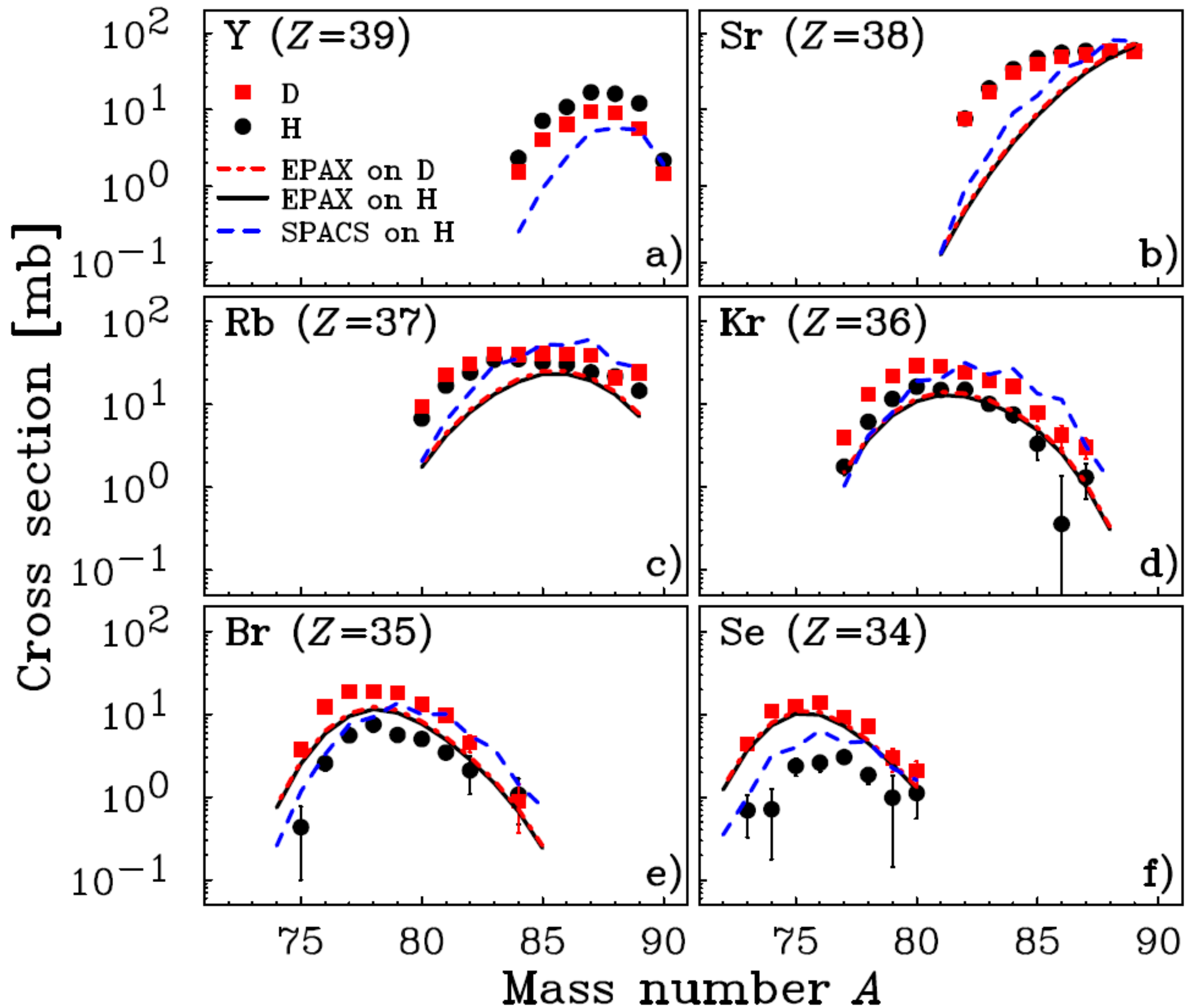
■ D

● H

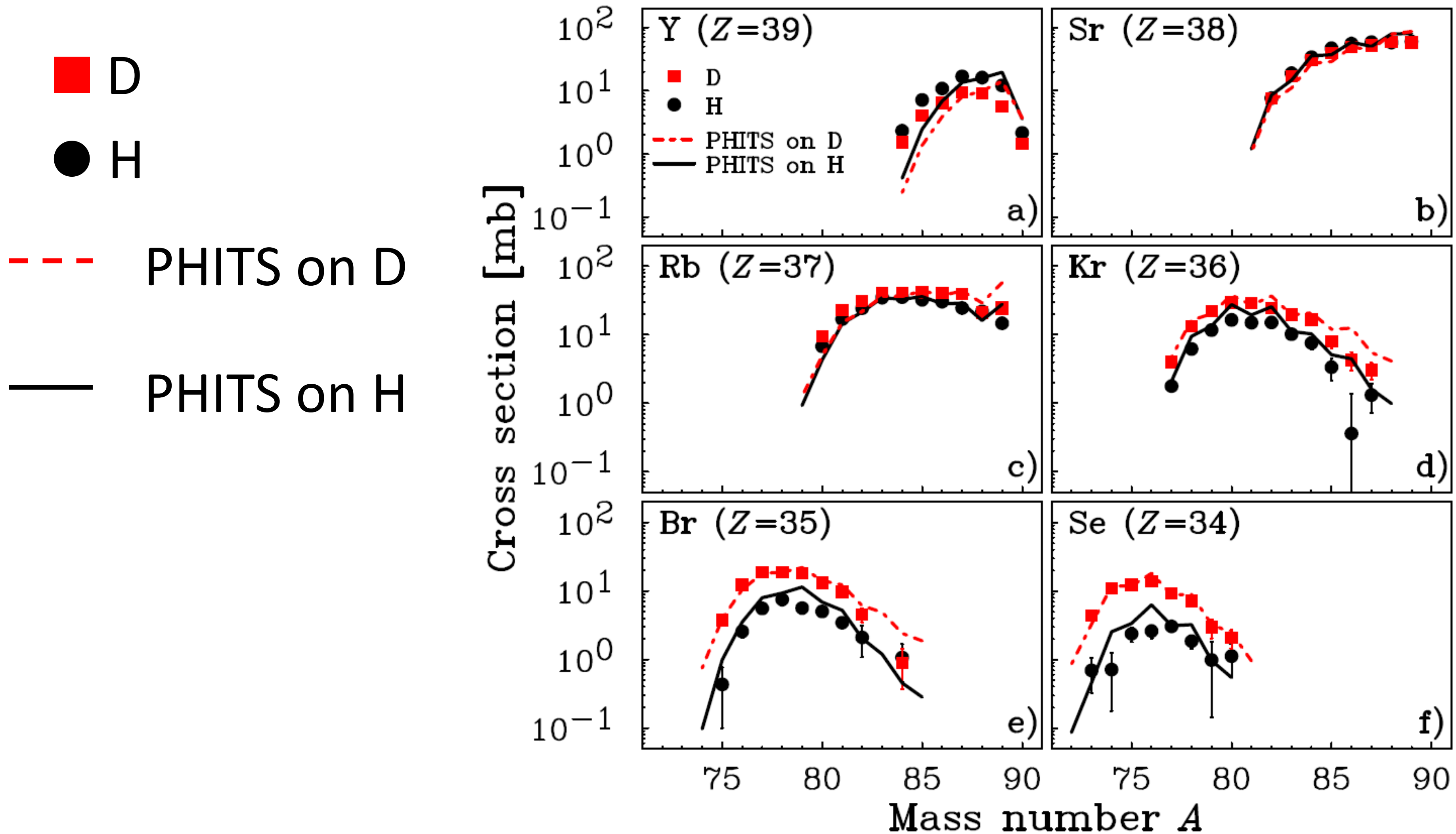
- - - EPAX on D

—— EPAX on H

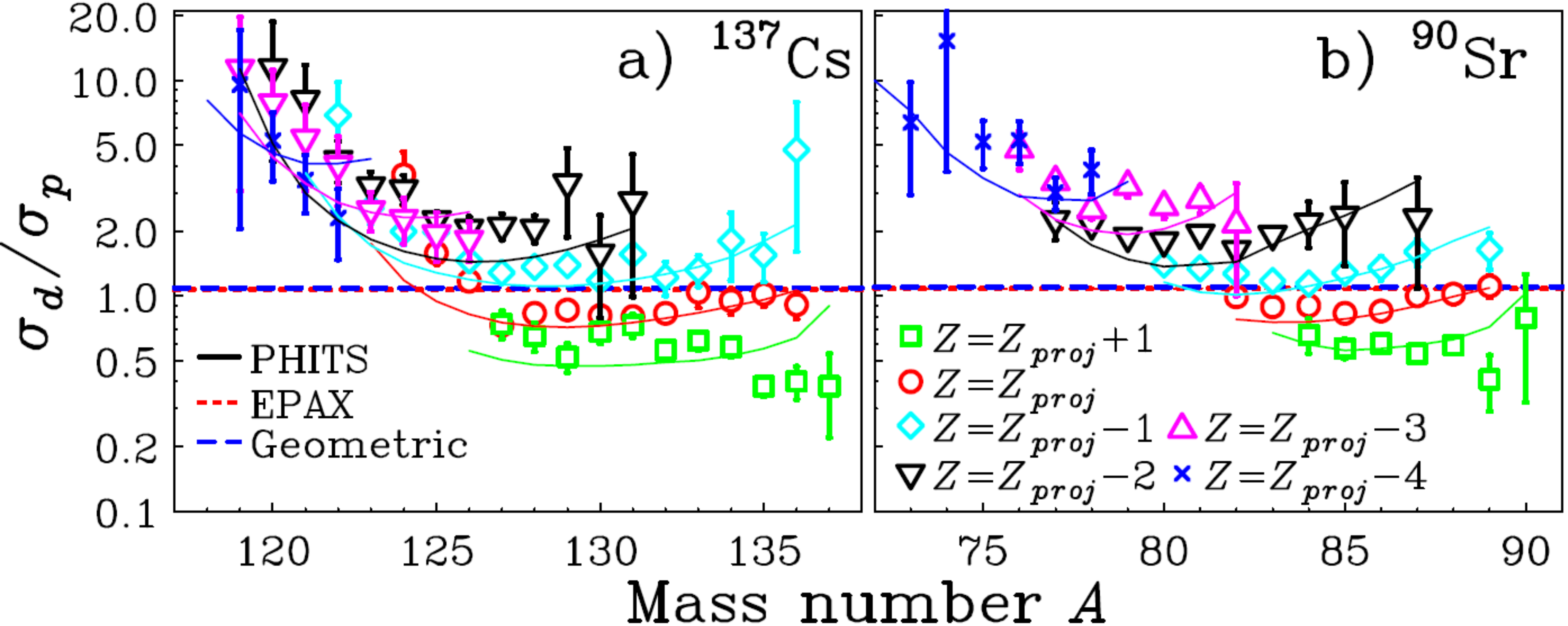
- - - SPACS on H



Cross sections for ^{90}Sr on H and D



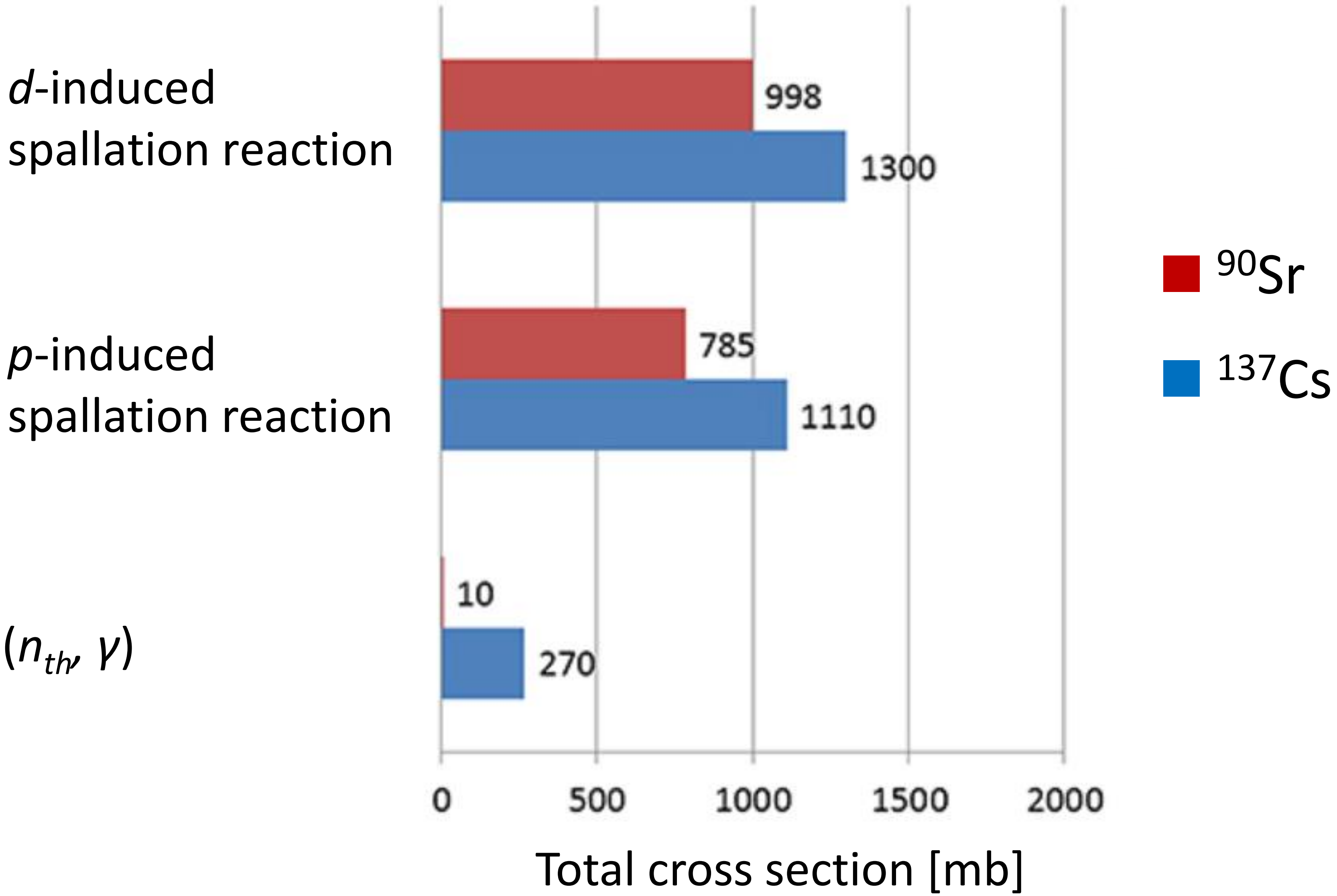
Difference between σ_d and σ_p



- σ_d/σ_p increases towards neutron-deficient side
- ΔZ increases, σ_d/σ_p increases

  -- Possible reason: $\sigma_{pn}>\sigma_{pp}$ @200MeV/u

Potential for LLFP transmutation



http://www.riken.jp/pr/press/2016/20160219_1/

Reduction of radiotoxicity

$^{137}Cs \rightarrow ^{135}Cs$

	Cross section	Halflife [year]
^{137}Cs	1 barn	30
^{135}Cs	64 mb	2.3×10^6

$^{90}Sr \rightarrow ^{79}Se$

	Cross section	Halflife [year]
^{90}Sr	0.9 barn	29
^{79}Se	1 mb	6.5×10^4

Summary

Reaction study on ^{90}Sr and ^{137}Cs

- Inverse kinematics using RIBF facilities
- Cross sections on carbon, *p*, *d*
- Comparison with calculations

Potential for the transmutation on ^{90}Sr and ^{137}Cs

- Large total cross section
- Reduction in the radiotoxicity
- Importance of *d*-induced reaction
- Collaboration with nuclear engineering

Perspective

- Systematic study on other LLFP nuclides

- Energy dependence of cross section
  Starting point (~200 MeV/*u*)
  Low reaction energy

- Energy and angular distribution of neutrons
  Reuse of neutrons

Physics Letters B 754 (2016) 104–108

Contents lists available at ScienceDirect

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Spallation reaction study for fission products in nuclear waste: Cross section measurements for ^{137}Cs and ^{90}Sr on proton and deuteron

H. Wang[a,*], H. Otsu[a], H. Sakurai[a], D.S. Ahn[a], M. Aikawa[b], P. Doornenbal[a], N. Fukuda[a], T. Isobe[a], S. Kawakami[c], S. Koyama[d], T. Kubo[a], S. Kubono[a], G. Lorusso[a], Y. Maeda[c], A. Makinaga[e], S. Momiyama[d], K. Nakano[f], M. Niikura[d], Y. Shiga[g,a], P.-A. Söderström[a], H. Suzuki[a], H. Takeda[a], S. Takeuchi[a], R. Taniuchi[d,a], Ya. Watanabe[a], Yu. Watanabe[f], H. Yamasaki[d], K. Yoshida[a]

[a] RIKEN Nishina Center, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan
[b] Faculty of Science, Hokkaido University, Sapporo 060-0810, Japan
[c] Department of Applied Physics, University of Miyazaki, Miyazaki 889-2192, Japan
[d] Department of Physics, University of Tokyo, 7-3-1 Hongo, Bunkyo, Tokyo 113-0033, Japan
[e] Graduate School of Medicine, Hokkaido University, North-14, West-5, Kita-ku, Sapporo 060-8648, Japan
[f] Department of Advanced Energy Engineering Science, Kyushu University, Kasuga, Fukuoka 816-8580, Japan
[g] Department of Physics, Rikkyo University, 3-34-1 Nishi-Ikebukuro, Toshima, Tokyo 171-8501, Japan

Collaborations

RIKEN Nishina Center

HW, H. Otsu, H. Sakurai, D. S. Ahn, P. Doornenbal, N. Fukuda, N. Inabe, T. Isobe, T. Kubo, S. Kubono, G. Lorusso, Y. Shiga, H. Suzuki, P.-A. Söderström, H. Takeda, S. Takeuchi, Y. Watanabe, K. Yoshida

Hokkaido University

M. Aikawa, A. Makinaga

Japan Atomic Energy Agency

T. Fukahori

Kyushu University

Y. Watanabe

Miyazaki University

S. Kawakami, Y. Maeda

University of Tokyo

T. Ando, S. Koyama, T. Miyazaki, S. Momiyama, M. Niikura, R. Taniuchi, H. Yamazaki

The ImPACT program

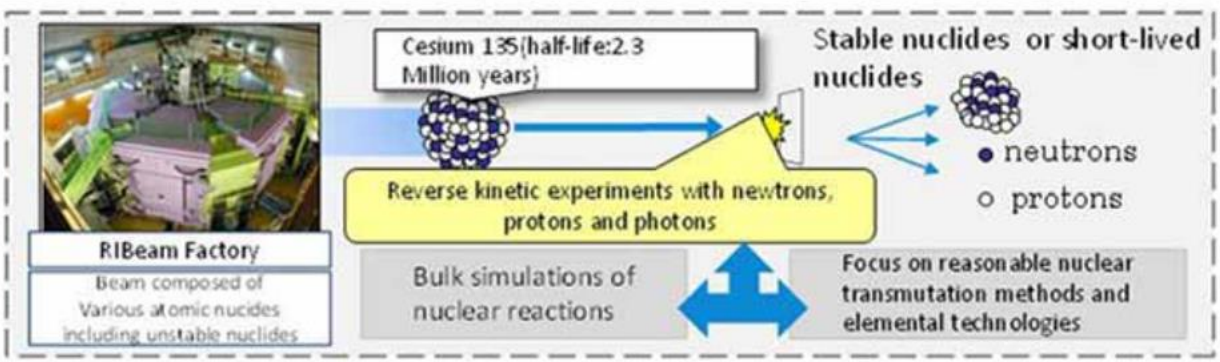
Impulsing Paradigm Change through Disruptive Technologies Program

Reduction and Resource Recycling of High-level Radioactive Wastes through Nuclear Transmutation

Disruptive Innovation

Keys to breakthrough

- To be the first in the world to obtain nuclear reaction data for long-lived fission products, and to confirm the world's first nuclear reaction path for conversion to short lived nuclides or stable nuclides.



http://www.jst.go.jp/impact/en/program/08.html

PL: Mizoguchi (Toshiba)
Project 1: Development of separation and recovery technologies

PL: Shimoura (CNS)/Sakurai(RIKEN)
Project 2: Obtained nuclear reaction data & new nuclear reaction control method

PM:Fujita

PL: Niita (RIST)
Project 3: Reaction theory modeling and simulation

PL: Sakurai (RIKEN)
Project 4: Evaluation of nuclear transmutation system and development of elemental technologies

PL: Tsujimoto (JAEA)
Project 5: Process concept for design

ImPACT in 2015 spring (BigRIPS+ZeroDegree)

Joint programs of ImPACT Project 2

- Fragmentation reaction measurement (Watanabe ; Kyushu)
- Coulomb breakup/excitation (Nakamura ; TITECH)
- Neutron knockout measurement (Otsu ; RIKEN)

Using secondary beams from In-flight fission of 345 MeV/u U beam

- Secondary beams :
  - 100 MeV/u :

    ^{107}Pd

    ^{93}Zr + ^{90}Sr

    ^{135}Cs

  - 200 MeV/u :

    $^{107,108}Pd$

    $^{93,94}Zr$

ImPACT in 2015 autumn (BigRIPS+SAMURAI)

Joint programs of ImPACT Project 2

- Fragmentation reaction measurement (Watanabe ; Kyushu)
- Coulomb breakup/excitation (Nakamura ; TITECH)
- Neutron knockout measurement (Otsu ; RIKEN)

Using secondary beams from In-flight fission of 345 MeV/u U beam

- Secondary beams :
  - 100 MeV/u :
    ^{93}Zr
    ^{79}Se
  - 200 MeV/u :
    $^{93,94}Zr$
    $^{79,80}Se$

Summary

- Reaction study on LLFP

  Pre-ImPACT

  -- ^{137}Cs, ^{90}Sr

  ImPACT2015@ZeroDegree

  --^{107}Pd, ^{93}Zr, ^{135}Cs

  ImPACT2015@SAMURAI

  --$^{93,94}Zr$, $^{79,80}Se$

- Plan for the ImPACT campaign in 2016

  Lower energy with the ZeroDegree spectrometer

Contributor list for ImPACT 2015 spring

RIKEN Nishina Center

H. Otsu, HW, H. Sakurai, S. Chen, N. Chiga, P. Doornenbal, T. Ichihara, T. Isobe, S. Kubono, T. Matsuzaki, Y. Shiga (+Rikkyo Univ.), P.-A. Söderström, S. Takeuchi, Y. Watanabe,

K. Yoshida, N. Fukuda, H. Suzuki, H. Takeda, Y. Shimizu, D. S. Ahn, H. Sato, M. Uesaka, T. Kubo

Kyushu University

Y. Watanabe, S. Kawase (+CNS), K. Nakano, S. Araki, T. Kin

Tokyo Institute of Technology

Y. Togano, M. Shikata, S. Takeuchi (+RNC), T. Nakamura, Y. Kondo, J. Tsubota, A. Saito, Y. Ozaki

Miyazaki University

Y. Maeda, S. Kawakami, T. Yamamoto

University of Tokyo

S. Momiyama , M. Niikura, R. Taniuchi, S. Koyama, T. Ando, S. Nagamine, T. Saito, K. Wimmer

CNS, Univ. of Tokyo

S. Michimasa, M. Matsushita, S. Shimoura

Hokkaido University

A. Makinaga, M. Aikawa

Contributor list for ImPACT 2015 autumn

RIKEN Nishina Center
- Hideaki Otsu
- HW
- Nobuyuki Chiga
- Yasushi Watanabe
- Tadaaki Isobe
- Paer Anders Söderström
- Takashi Ichihara
- Hiroyoshi Sakurai

RNC BigRIPS Team
- Naoki Fukuda
- Koichi Yoshida
- Hiroshi Suzuki
- Hiroyuki Takeda
- Yohei Shimizu
- Deuk Soon Ahn
- Toshiyuki Sumikama
- Hiromi Sato
- Naohito Inabe
- Toshiyuki Kubo

Kyushu U.
- Shoichiro Kawase
- Keita Nakano
- Junki Suwa
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- Yukinobu Watanabe

Tokyo Tech
- Satoshi Takeuchi
- Yasuhiro Togano
- Yosuke Kondo
- Mizuki Shikata
- Junichi Tsubota
- Atsumi Saito
- Tomoyuki Ozaki
- Takahito Tomai
- Akihiro Hirayama
- Takashi Nakamura

Tohoku U.
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Kyoto U.
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U. Tokyo
- Shunpei Koyama
- Takeshi Saito
- Shunsuke Nagamine

CNS, U. Tokyo
- Shin'ichiro Michimasa
- Masashi Matsushita

Rikkyo U.
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Osaka U.
- Masaomi Tanaka
- Yutaro Tanaka
- Hang Du

Niigata U.
- Maya Takechi
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Tokyo U. of Science
- Daiki Nishimura
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Miyazaki U.
- Yukie Maeda

Kochi U. of Tech
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Hokkaido U.
- Masayuki Aikawa

TU Darmstadt, Germany
- Hans Toshihide Törnqvist
- Leyla Atar
- Julian Kahlbow
- Tom Aumann

KVI, Nederlands
- Ali Najafi
- Christiaan Douma

IRB, Croatia
- Igor Gašparić

ELTE, Hungary
- Ákos Horváth

Thank you