

Mass spectrometry of short-lived nuclides (for origins of heavy elements)

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1. Synthesis of heavy elements in the universe

review of very basics

why we measure atomic masses

very basics but I didn't know in your age

2. Comprehensive mass measurement of short-lived nuclides

mass as finger print

nuclear structure studies

heavy element synthesis

toward islands of stability

Michiharu Wada: Personnel background

- **Tohoku University** 1979~1991

Laser spectroscopy of trapped Sr ions
Alpine expedition to Tibet, China (1986)

- **INS, Univ. Tokyo** 1991~1999

R&D for E-arena, JHP
Innovation of RF-ion guide

- **RIKEN** 1999~2015

Laser spectroscopy of trapped short-lived Be ions
Innovation of RF carpet
R&D of SLOWRI @ RIBF

- **KEK** 2015~Mar2024 retired

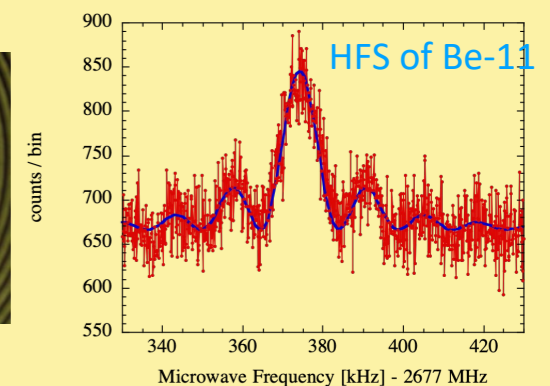
Comprehensive mass measurements with MRTOF
MNT for n-rich nuclides at KISS

★ **Scientific Expedition “for the first time”**

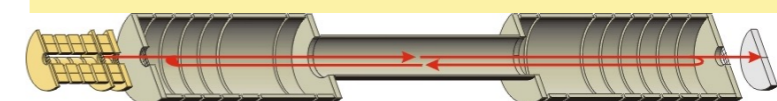
東北大学西藏学術登山隊(1986)



念青唐古拉峰(7162m) First ascent

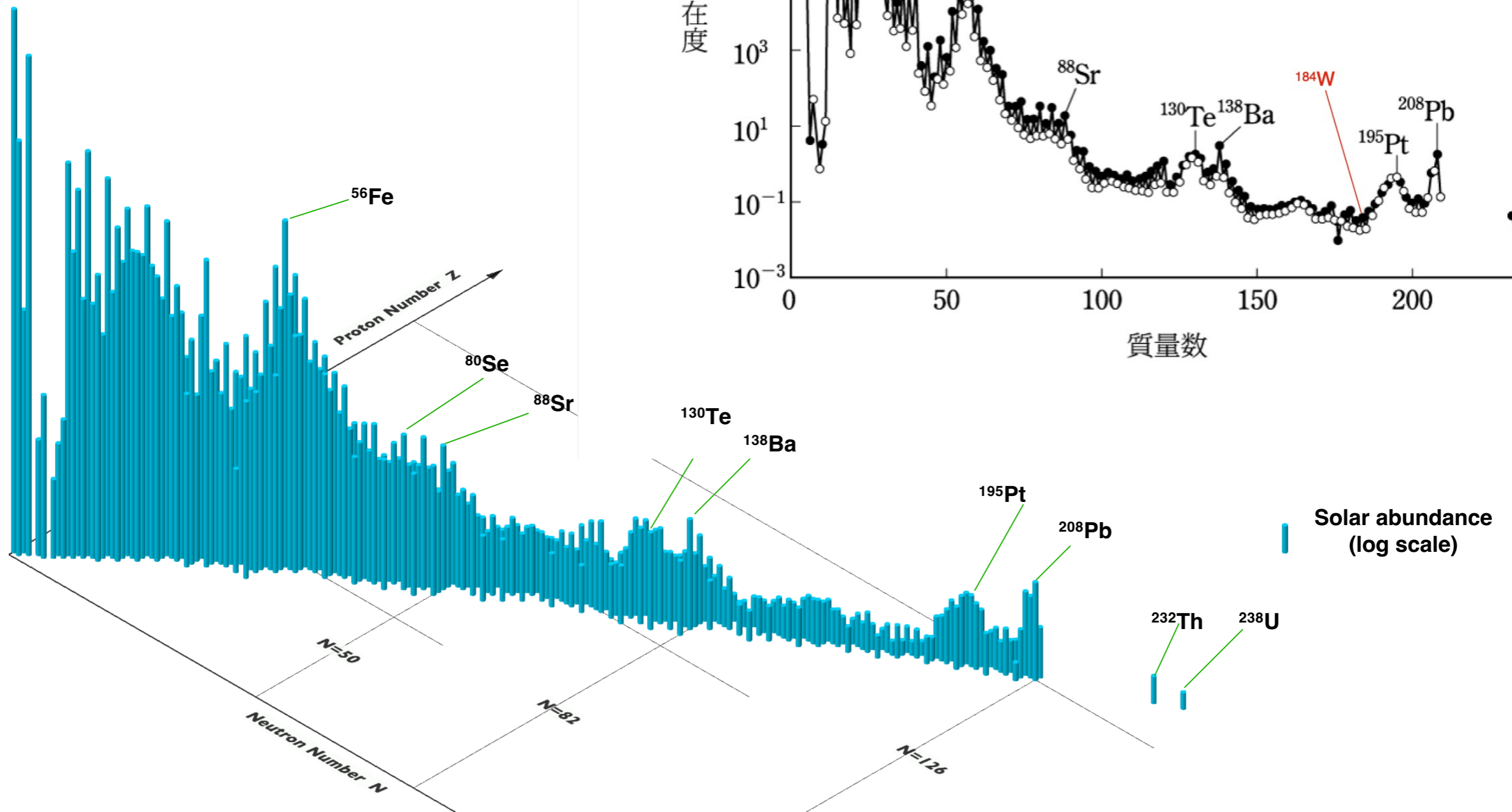


MRTOF mass spectrograph



We have various elements (~83) and their isotopes (~255)

Q1: How do we know?



Optical Spectroscopy

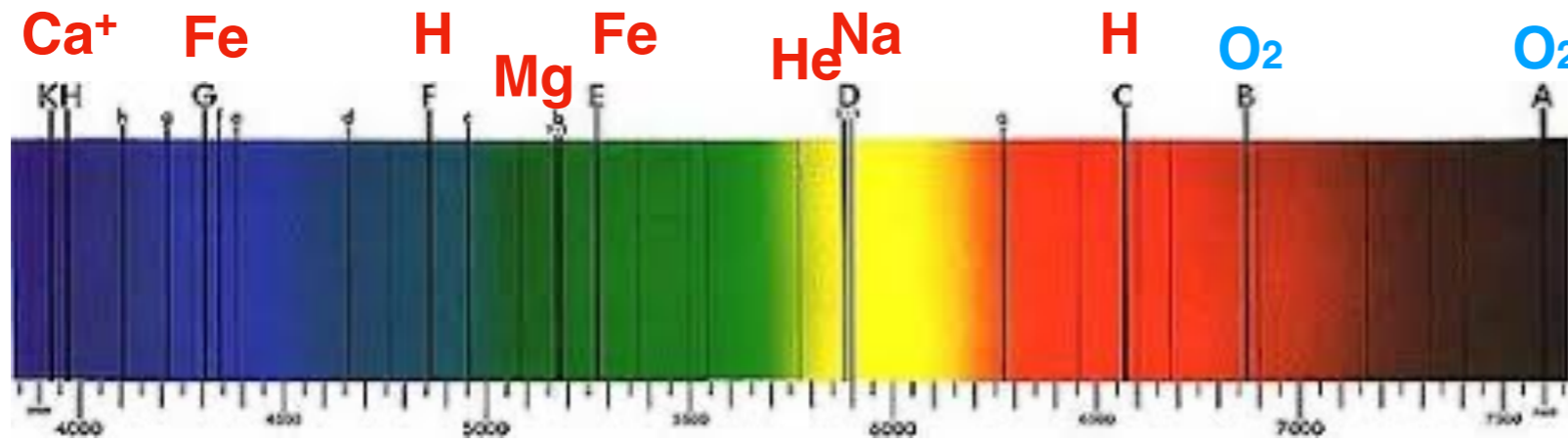
Atmosphere of Earth



Atmosphere of Sun

光球

Sun (Light source)



Absorption lines (or Fraunhofer lines) in optical spectrum

- ★ Volatile elements in “atmosphere” of Sun are observable
- Isotopes identification is difficult
- Contents in atmosphere and core are identical?
 - ▶ Solar quakes can model internal structure of the Sun



1814, J. von Fraunhofer discovered >500 “dark lines” in spectrum. However, the origin of the “lines” were determined by Bunsen & Kirchhoff 40 years later.

Elemental & Isotopic Analyses of Meteorite

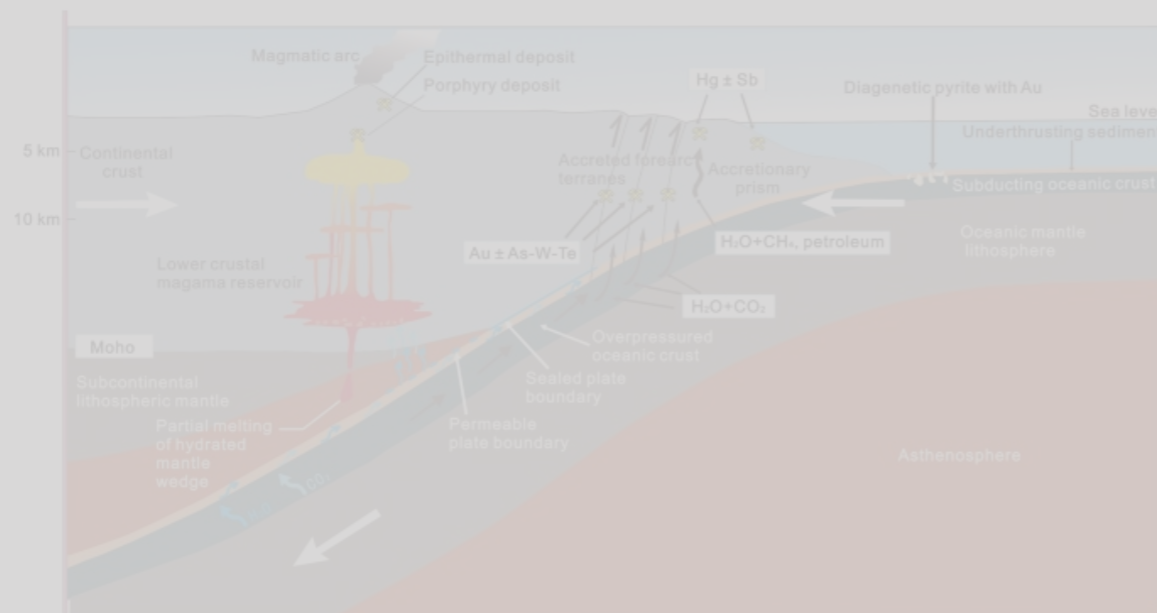
- A specific meteorite contains materials formed 4.5 G year ago.
- Moon stones formed 3.2~4.4 G years ago.
 - ★ non-volatile elements with isotopic ratios
 - How accurately confirm it represent the original abundance?



A flake of Allende meteorite fell in Mexico in 1969.

Q1: Why not stones or soil on the Earth's surface ?

Orogeny (mountain-building processes) has led to the formation of metal deposits, resulting in the uneven distribution of elements.



Moon stone from Apollo 15, 1971

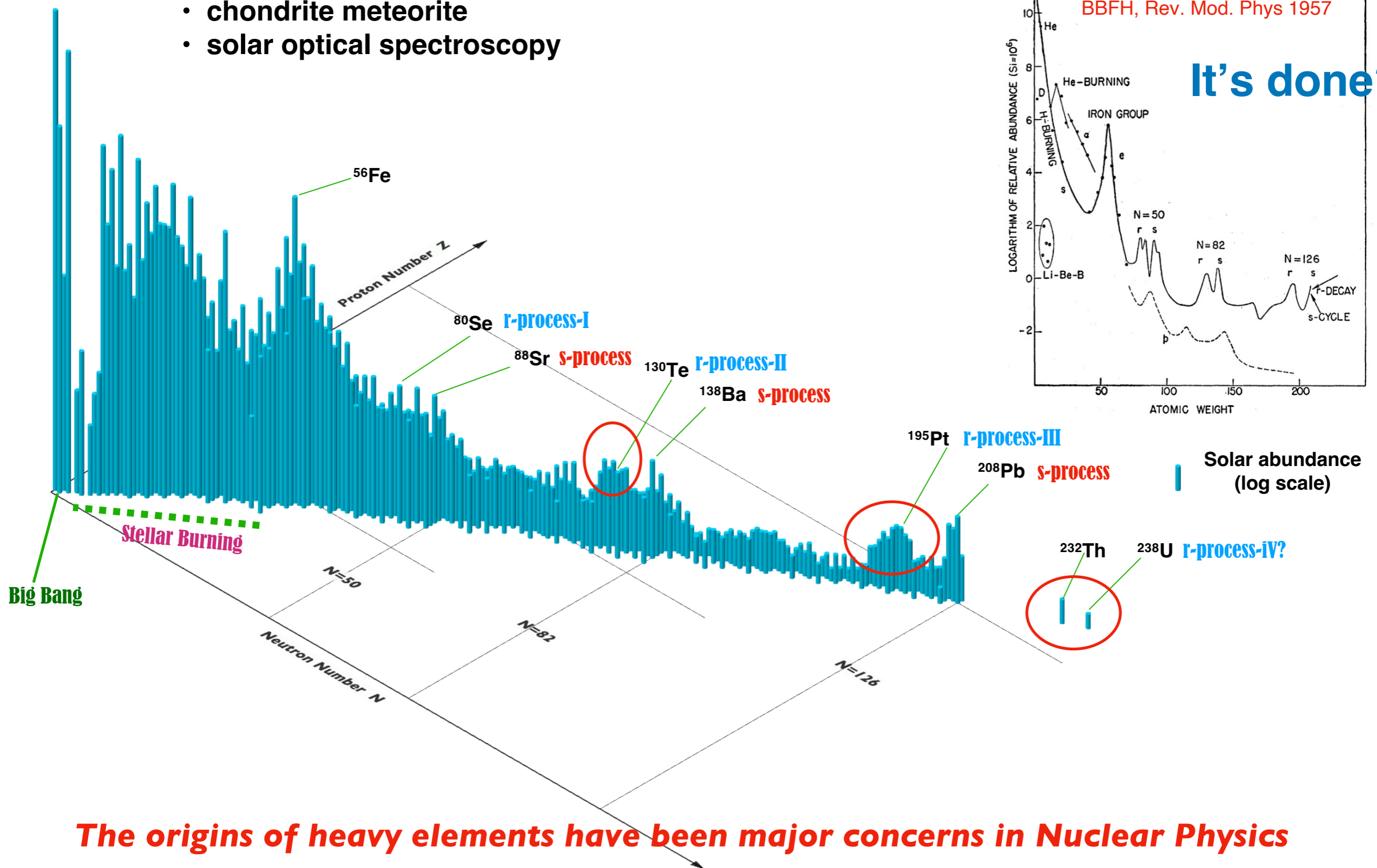
Nuclides Abundance in the Solar System

~83 elements, ~255 nuclides

H: Fe: Pb: Th log-ratio 12 : 7.4 : 2.1 : 0.26

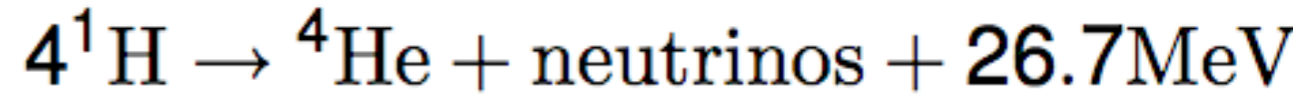
evaluated by

- chondrite meteorite
- solar optical spectroscopy



The origins of heavy elements have been major concerns in Nuclear Physics

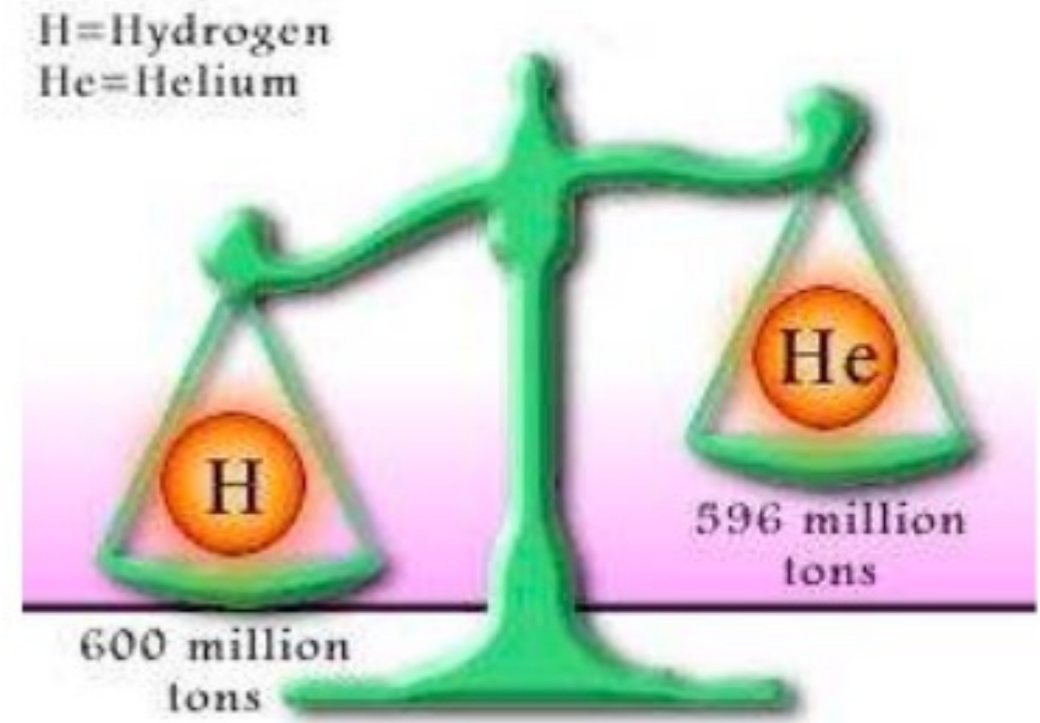
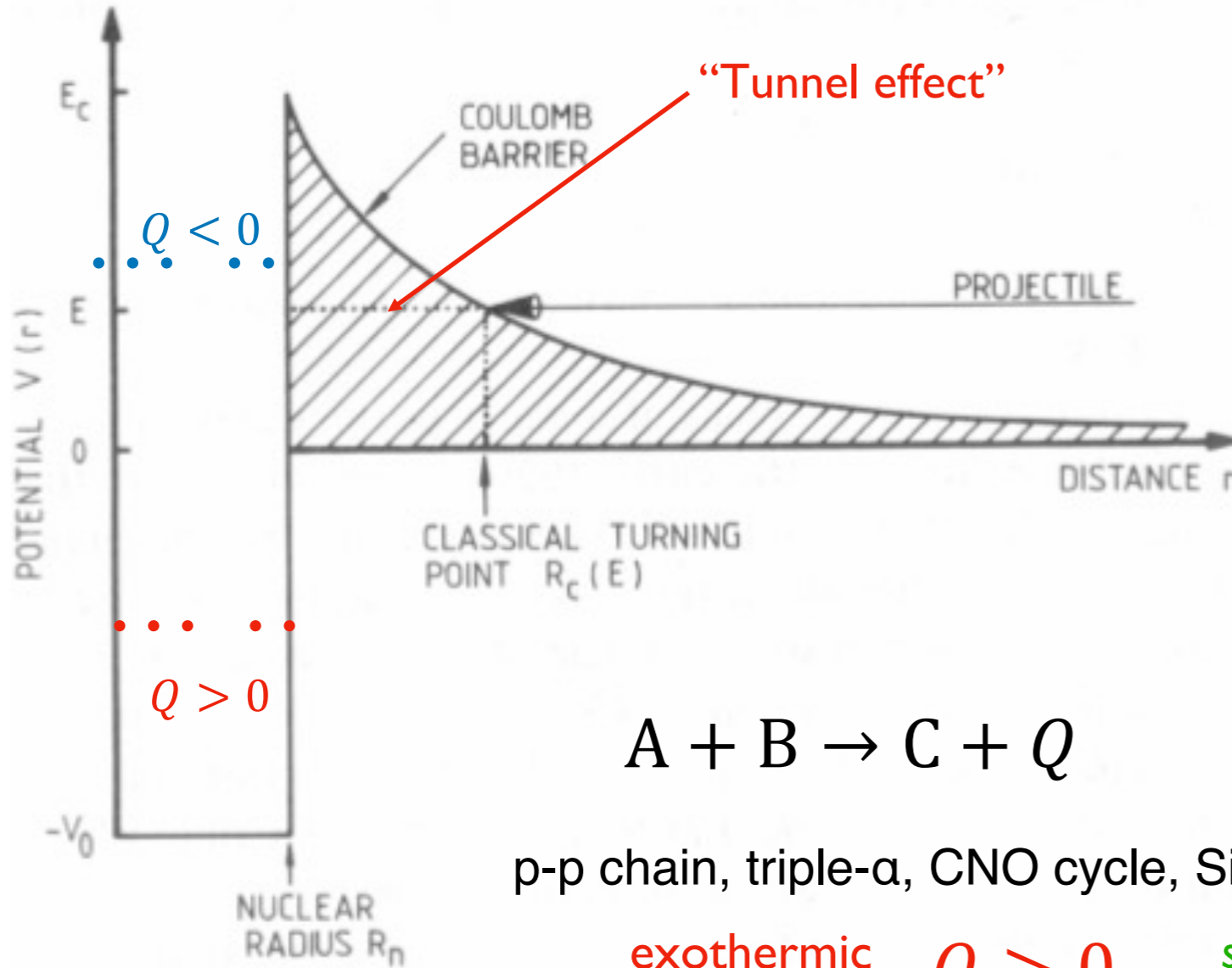
Stellar Burning



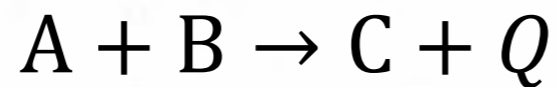
(Thermal) projectile energy is much less than Coulomb barrier

1.35 keV

550keV



600Mt/s of Hydrogen convert to He 596Mt at Sun



p-p chain, triple- α , CNO cycle, Si cycle, etc

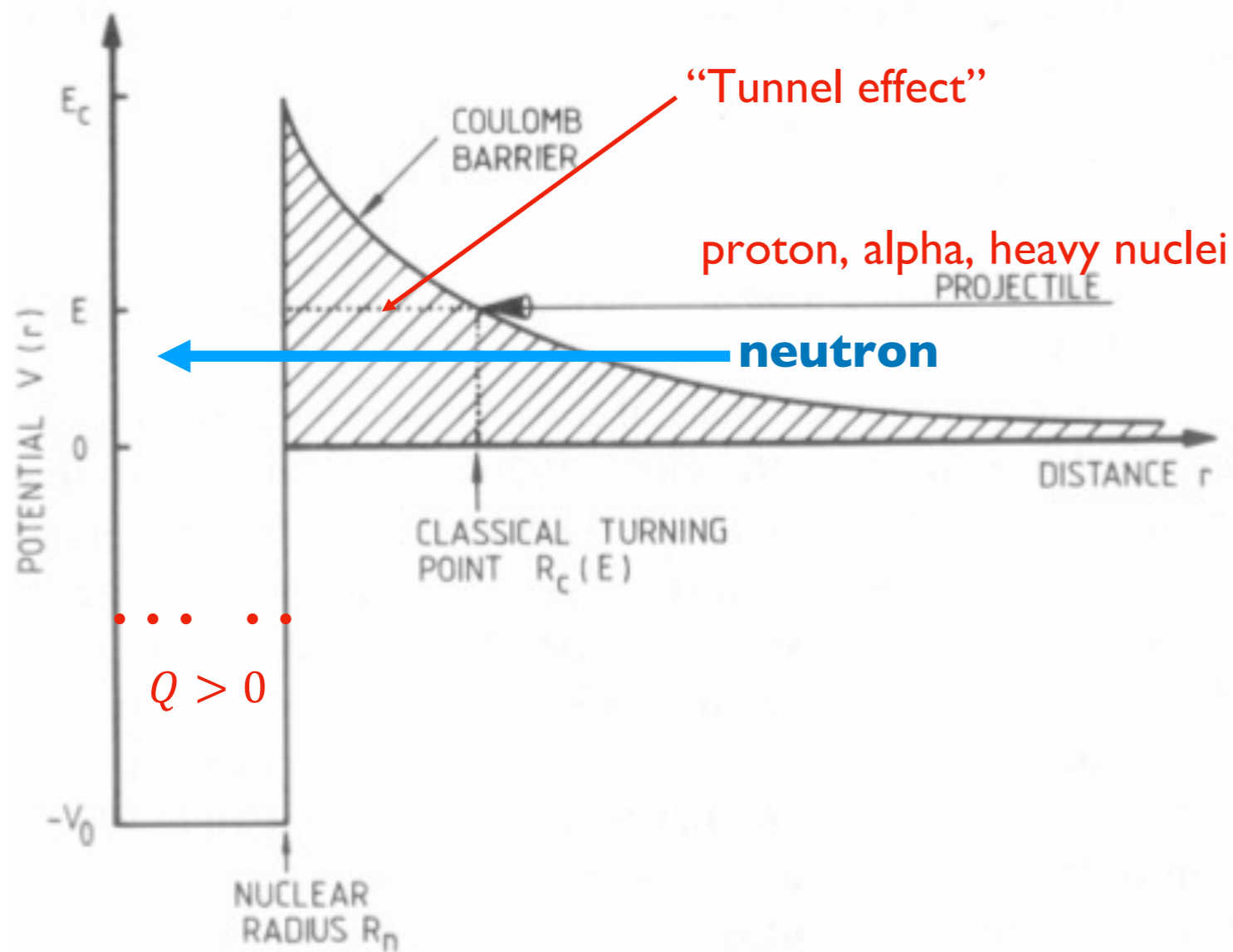
exothermic $Q > 0$ *slowly take place via tunnel effect*

Majority of possible reactions for heavier than Fe, Ni

endothermic $Q < 0$ *difficult to take place with thermal kinetic energies*

What about neutron capture?

No Coulomb barrier



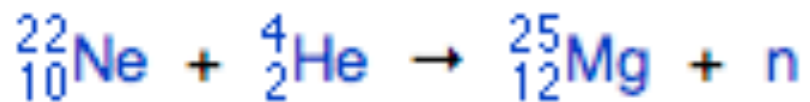
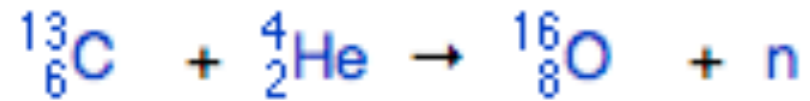
neutron capture and successive β^- decay produces $Z+1$ nuclide

^{57}Cu 196.3 ms	^{58}Cu 3.204 s	^{59}Cu 1.36 m	^{60}Cu 23.7 m	^{61}Cu 3.333 h	^{62}Cu 9.67 m	^{63}Cu 69.15
^{56}Ni 6.075 d	^{57}Ni 35.60 h	^{58}Ni 68.077	^{59}Ni $7.6 \cdot 10^4$ y	^{60}Ni 26.223	^{61}Ni 1.140	^{62}Ni 3.634
^{55}Co 17.53 h	^{56}Co 77.236 d	^{57}Co 271.74 d	^{58}Co 70.86 d ★9.10 h	^{59}Co 100	^{60}Co 5.271 y ★10.467 m	^{61}Co 1.650 h
^{54}Fe 5.845	^{55}Fe 2.744 y	^{56}Fe 91.754	^{57}Fe 2.119	^{58}Fe 0.282	^{59}Fe 44.495 d	^{60}Fe $2.62 \cdot 10^6$ y
^{53}Mn $3.74 \cdot 10^6$ y	^{54}Mn 312.05 d	^{55}Mn 100	^{56}Mn 2.5789 h	^{57}Mn 1.42 m	^{58}Mn ★1.09 m 3.0 s	^{59}Mn 4.59 s

β^- (dashed arrow from ^{59}Fe to ^{59}Co)
 $n\gamma$ (dashed arrow from ^{58}Mn to ^{59}Mn)

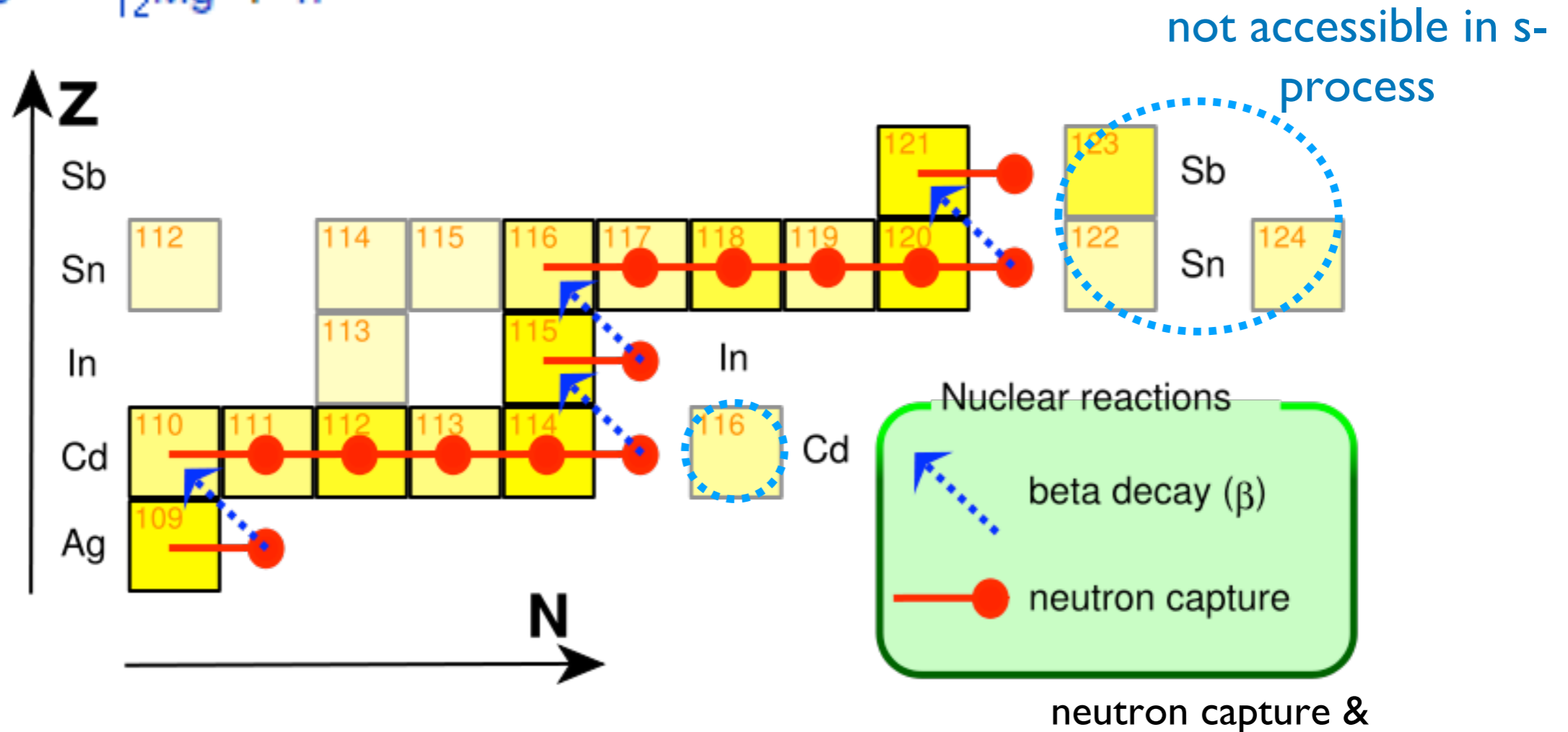
S-process for heavy element synthesis

source of neutrons



Asymptotic Giant Branch of 0.6~10 solar mass stars

at the end of life, 1,000-1,000,000 years of slow process

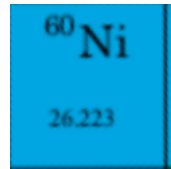


Heavy nuclides can be synthesized up to ${}^{208}\text{Pb}$ and ${}^{209}\text{Bi}$

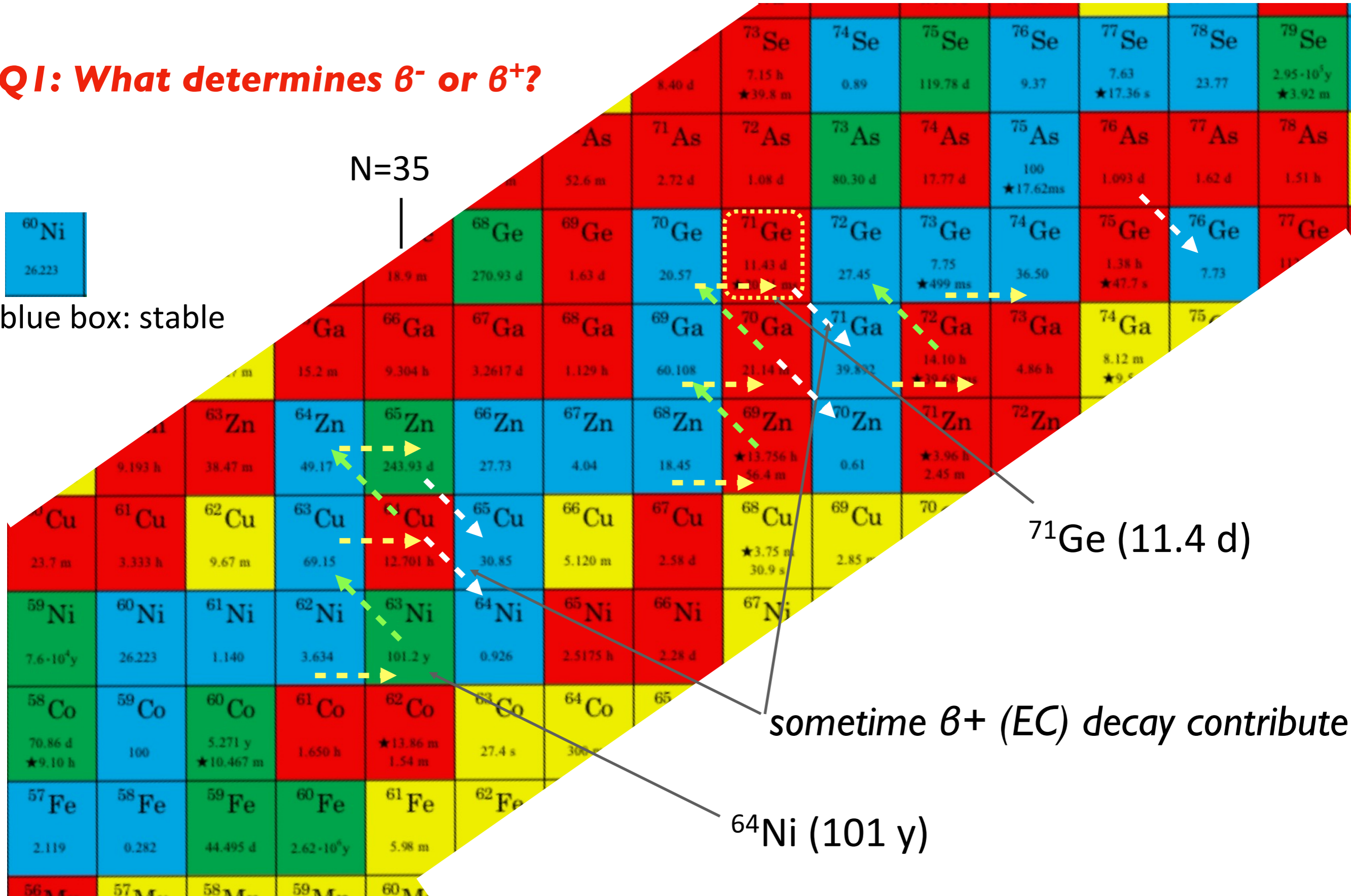
Q1: Have you tried tracing the path?

s-process follows stable nuclides via β^- decay??

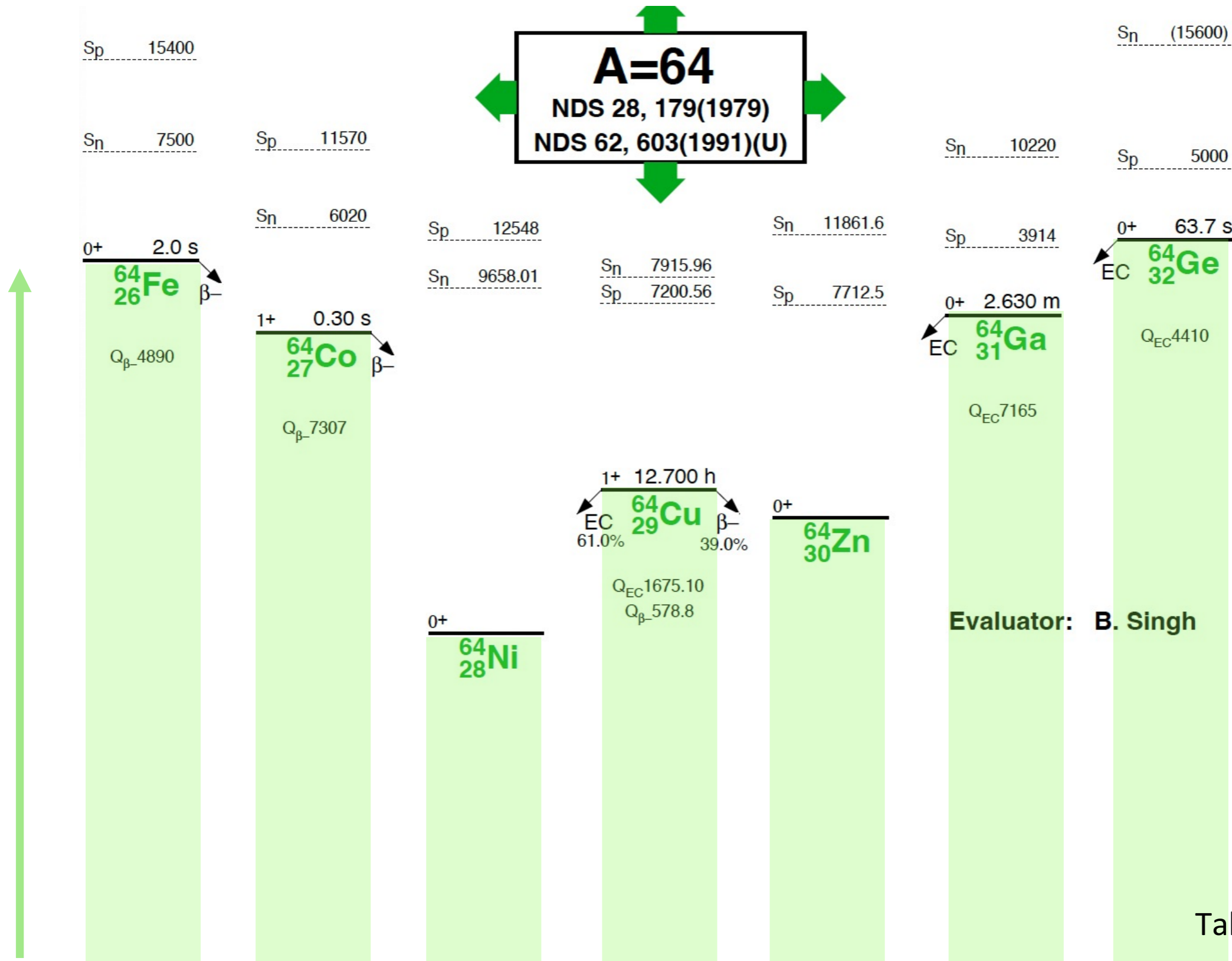
Q1: What determines β^- or β^+ ?



blue box: stable



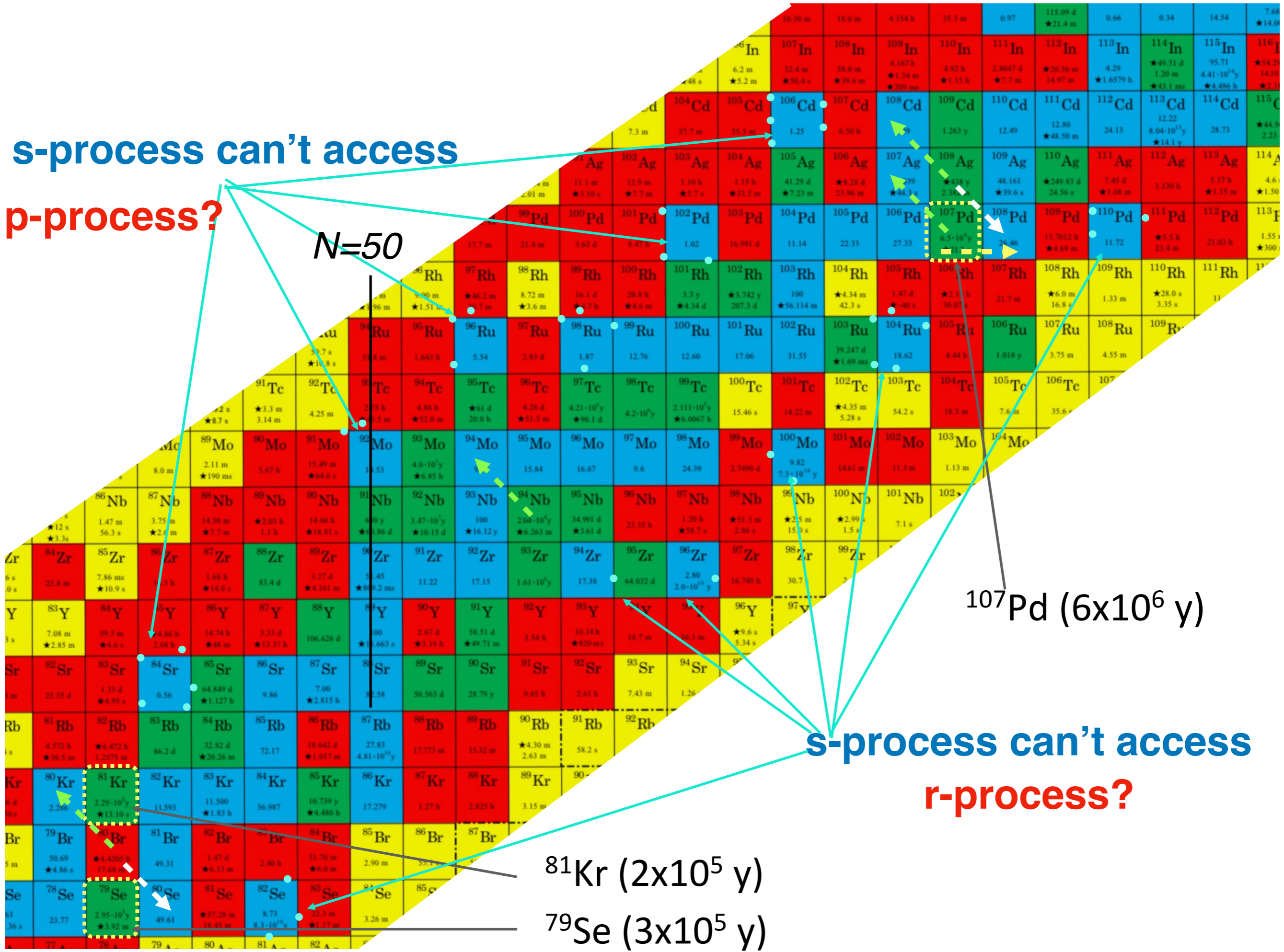
Mass



s-process can't access

p-process?

$N=50$



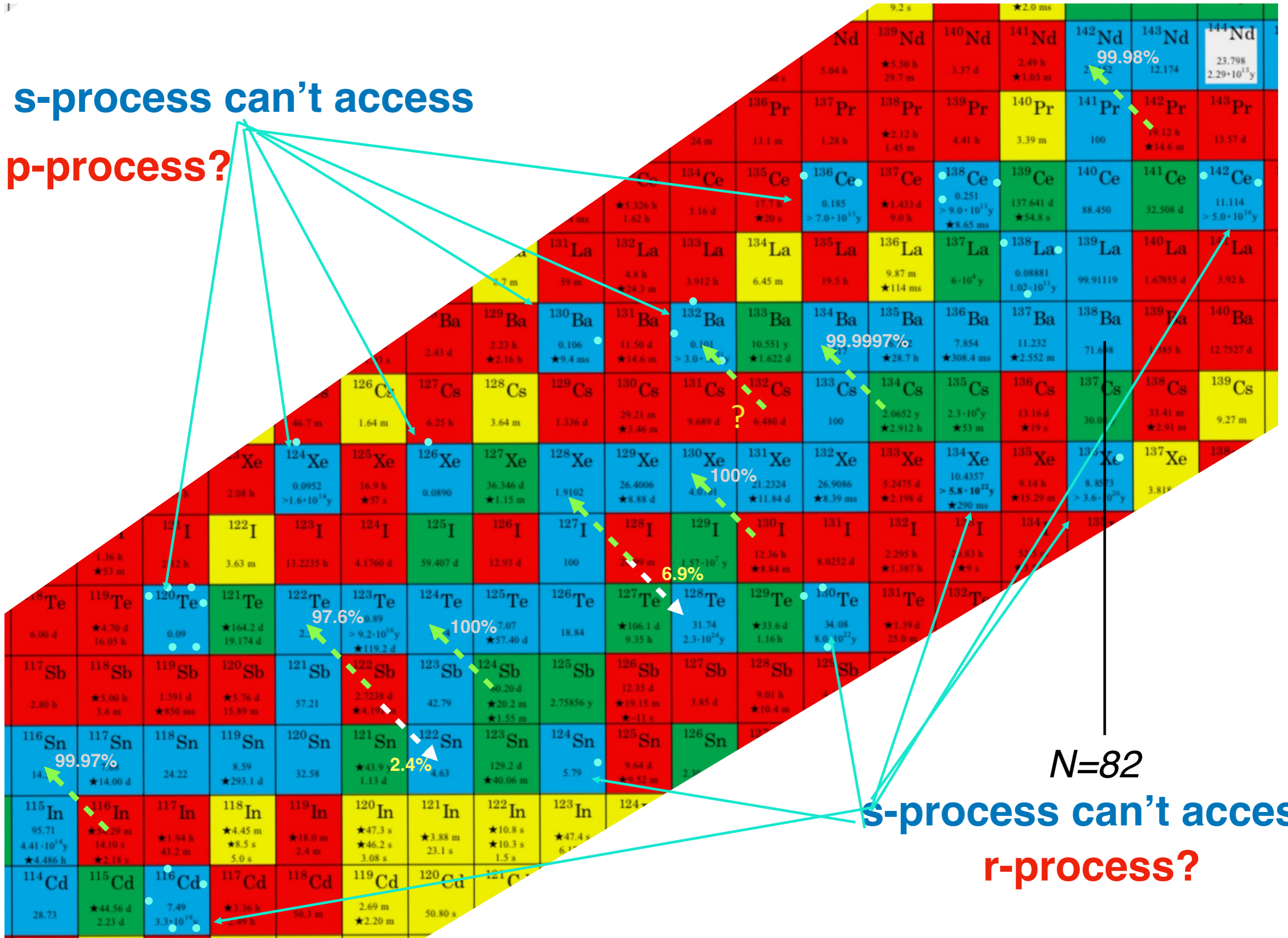
^{107}Pd (6×10^6 y)

s-process can't access
r-process?

^{81}Kr (2×10^5 y)

^{79}Se (3×10^5 y)

s-process can't access
p-process?



$N=82$

s-process can't access
r-process?

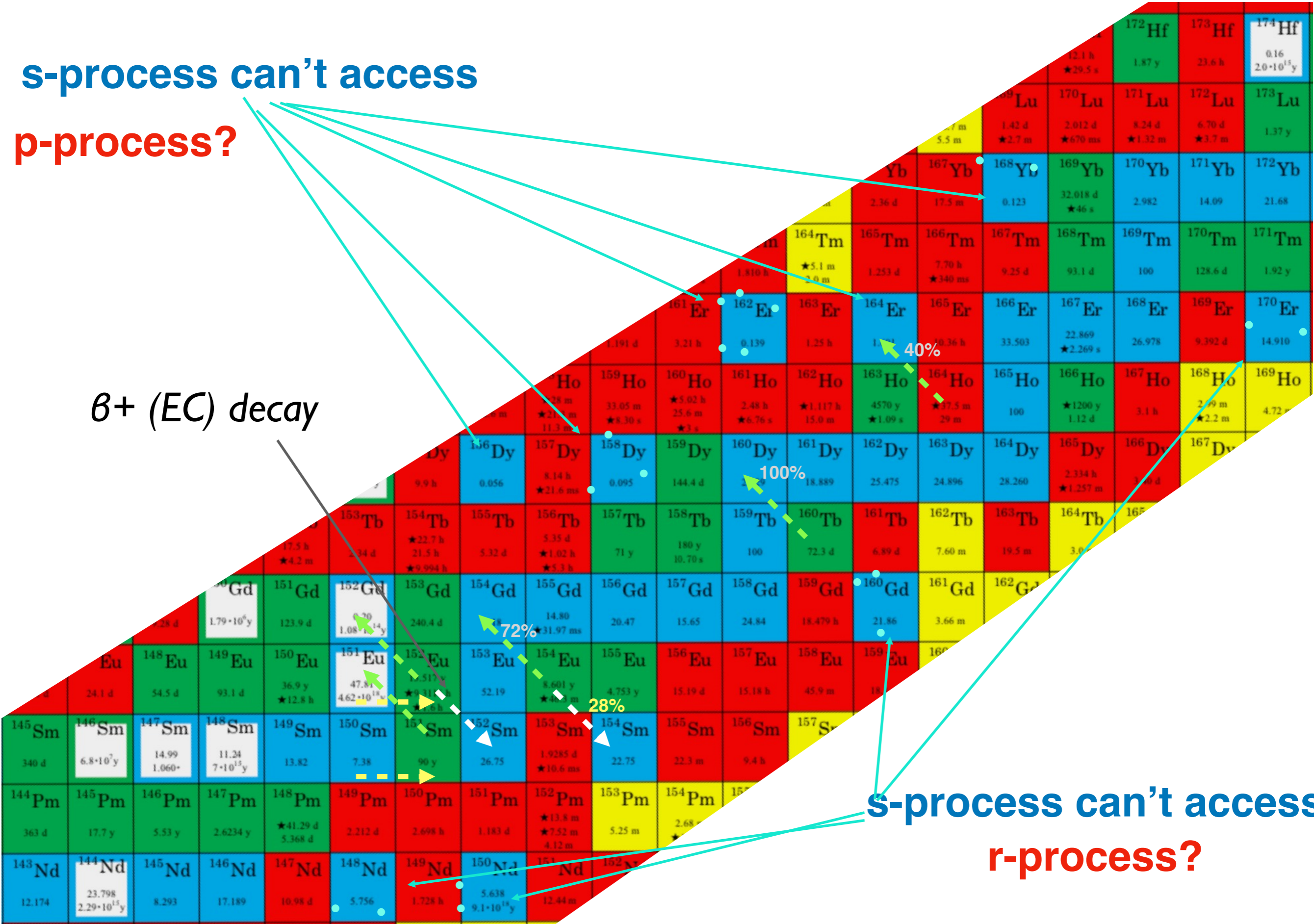
s-process can't access

p-process?

β^+ (EC) decay

s-process can't access

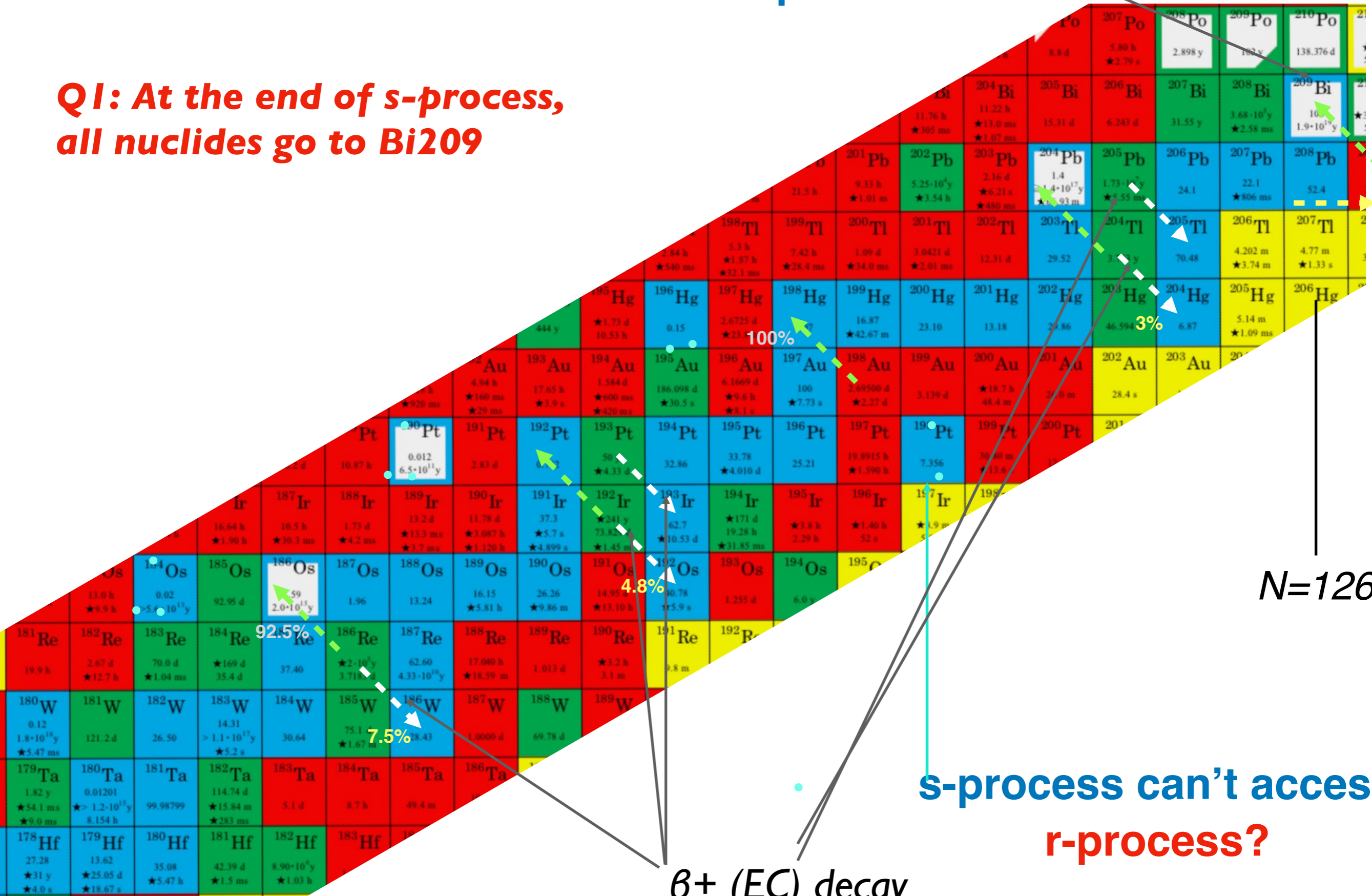
r-process?



End point of s-process

^{209}Bi

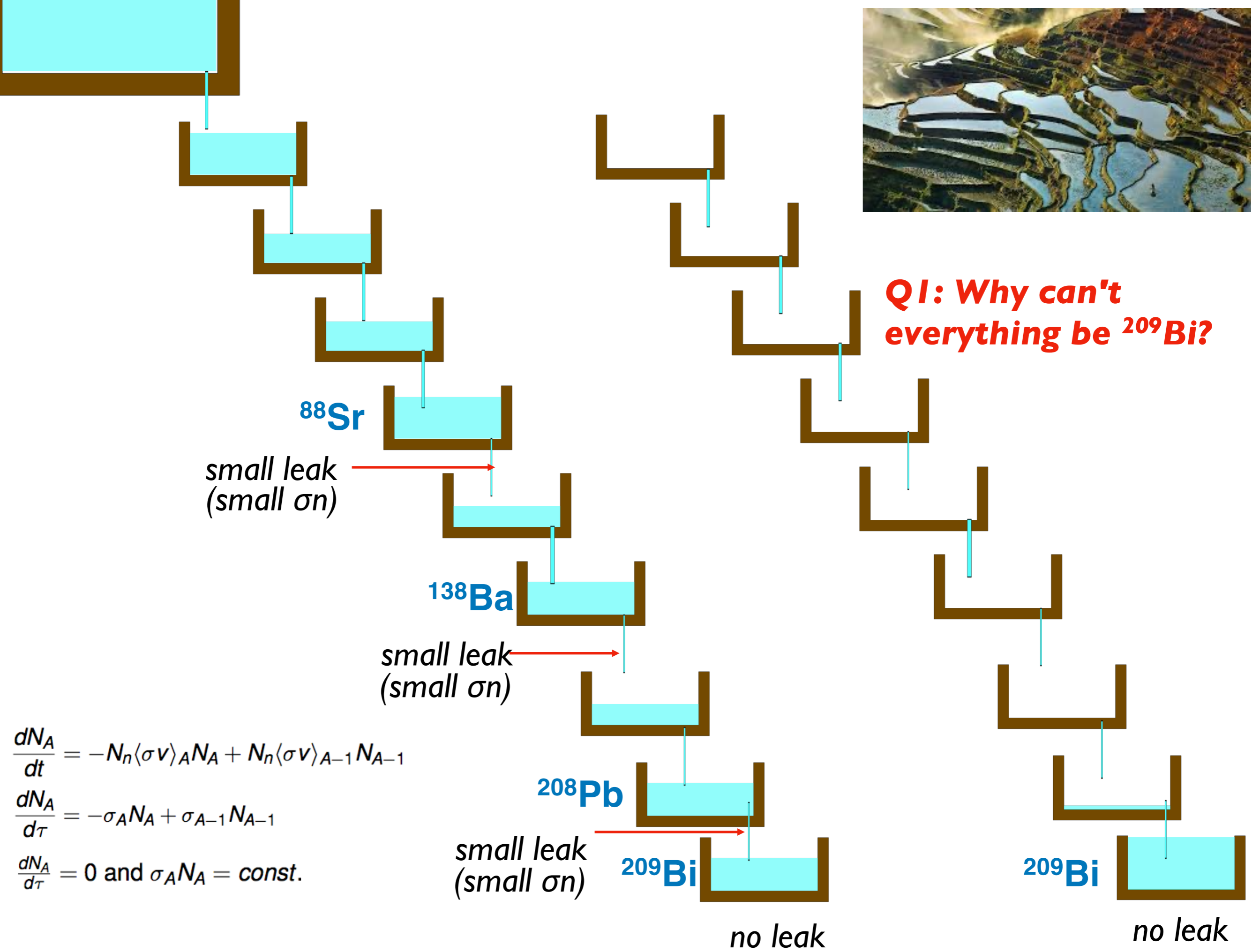
Q1: At the end of s-process, all nuclides go to Bi209



$N=126$

s-process can't access r-process?

β^+ (EC) decay

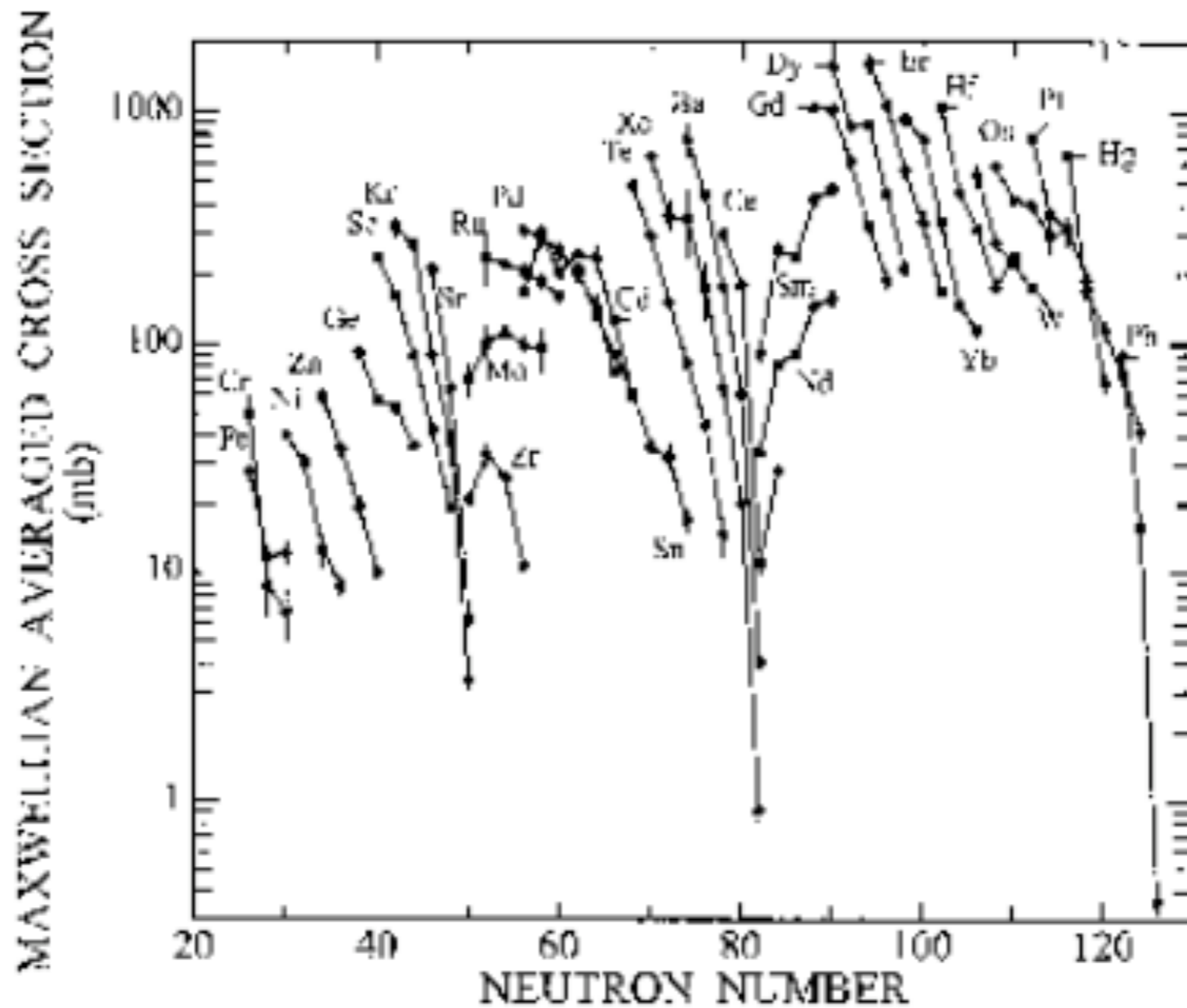


$$\frac{dN_A}{dt} = -N_n \langle \sigma v \rangle_A N_A + N_n \langle \sigma v \rangle_{A-1} N_{A-1}$$

$$\frac{dN_A}{d\tau} = -\sigma_A N_A + \sigma_{A-1} N_{A-1}$$

$$\frac{dN_A}{d\tau} = 0 \text{ and } \sigma_A N_A = \text{const.}$$

n-capture cross section becomes minimum at n-magic number



$$\sigma_A N_A = \text{const.}$$

At minimum σ_n , Abundance max

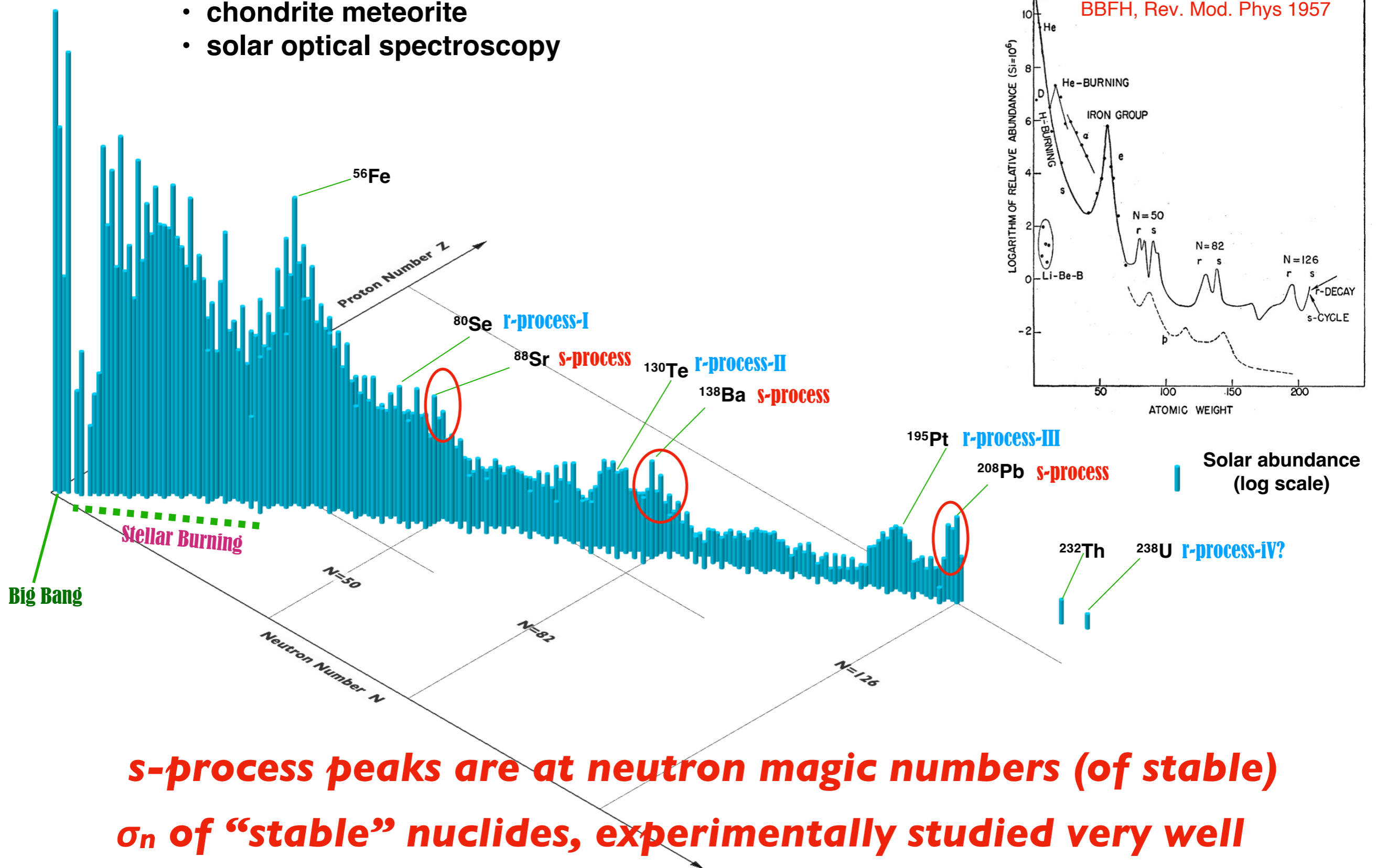
Nuclides Abundance in the Solar System

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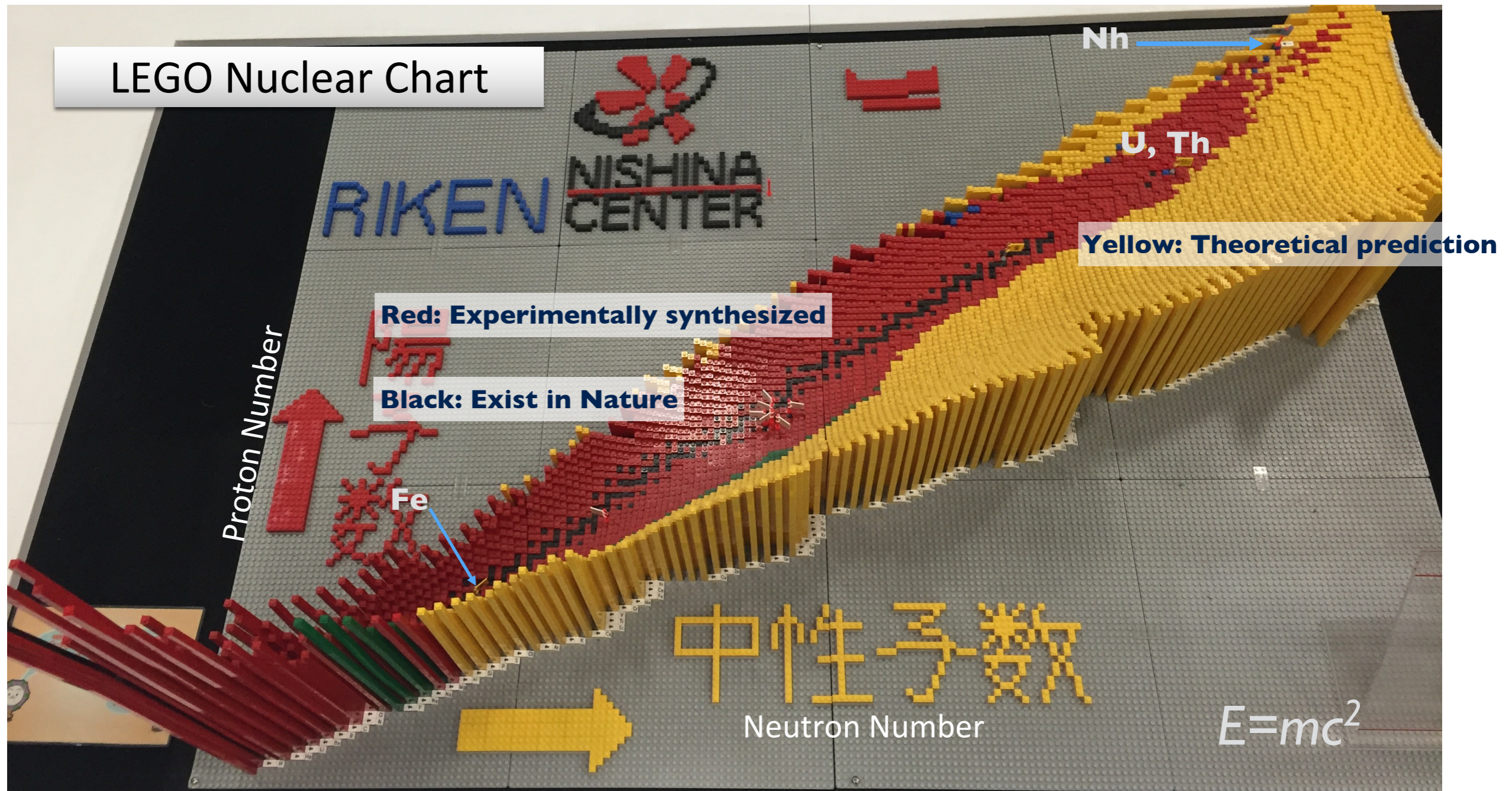
evaluated by

- chondrite meteorite
- solar optical spectroscopy



s-process peaks are at neutron magic numbers (of stable) σ_n of "stable" nuclides, experimentally studied very well

LEGO Nuclear Chart



Q1: What is the height of pillar?

The heights indicates the mass

But divided by A and magnify around mass/A =1

「全原子核の地図」核図表とは

核図表の見かた

核図表

Chart of Nuclides

原子核の大きさ

私たちの体は星くずでできている

皆さんご存知の周期表は、元素を原子番号順に並べた表ですが、性質の似た元素が縦に揃うよう工夫してあるため、凹型という不思議な形状をしています。

一方、核図表は単純に原子番号(つまり陽子⁺の数)を縦軸に、中性子⁰の数を横軸に並べた原子核の表です。原子核は陽子数と中性子数の数によっておよそ1万種類あると言われていて、同じ陽子数(元素)でも様々な中性子数の原子核が存在することがこの表からわかります。同じ元素で、違った中性子数である元素を「同位元素^{*}」と言います。

この表の「高さ」というか「谷の深さ」はそれぞれの原子核の結合エネルギー[†]を示しています(低い方が強い)。結合力の強い安定核(黒)が、あたかも谷筋を流れる川のようなので、これを「ハイゼンベルグ[‡]の谷」と呼びます。

縦軸は陽子数であり、元素の種類でもあります。下から上に向かって陽子数が増えていくため、周期表の順番と一緒です。横軸は中性子数で左から右に向かって中性子の数が増えていきます。つまり横一線では同じ陽子数で、違う中性子数で構成される同位元素になります。

黒い所は安定核^{※6}といい、天然に存在する原子核です。オレンジ色の所はこれまでに発見・合成された原子核です。白い所は理論的に存在するとされる原子核で未発見の原子核です。高さは結合エネルギー[†]を表していて、高いほど原子核が不安定といえます。

原子核の表記方法

陽子数 + 中性子数 元素記号

例えばヘリウム4では

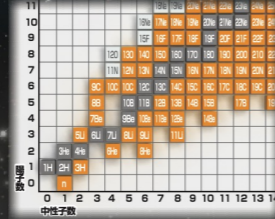


例えば炭素13では

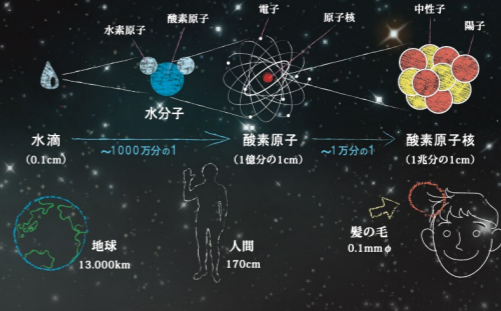


と表す。(※右の核図表では陽子数は省略)

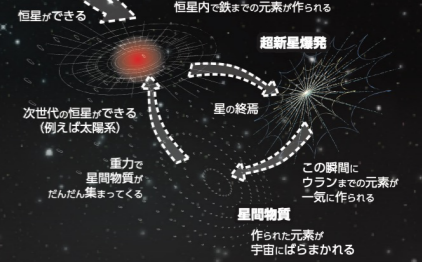
真上からみた立体核図表



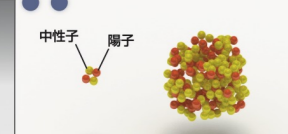
原子の大きさは約1000万分の1mm。最新鋭の顕微鏡でぎりぎり見える大きさです。しかし元素の本体は原子の中心にある原子核です。原子核の大きさは原子のさらに10万分の1ですから、まったく見ることは出来ません。たとえ見えなくても私たちは原子核が陽子(+)の電気をもち、中性子(電気を持たない)という2種類の粒子の塊であることを知っています。



宇宙では常に元素合成を繰り返しています。宇宙誕生時には水素とヘリウムしかありませんでした。それから3億年ほどで星が誕生し、その星の内部や、星の爆発で原子番号が大きい元素は作られています。



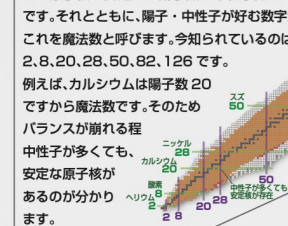
1 2 原子核=陽子+中性子



ヘリウム4
●×2
●×2

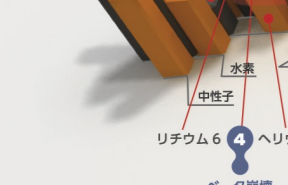
ウラン235
●×92
●×143

3 4 原子核の崩壊—アルファ崩壊、ベータ崩壊

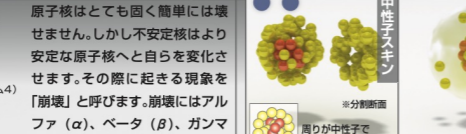


7 魔法数(マジックナンバー)

原子核の安定には陽子数と中性子数のバランスが大切です。それとともに、陽子・中性子が好む数字があり、これを魔法数と呼びます。今知られているのは、2, 8, 20, 28, 50, 82, 126です。例えば、カルシウムは陽子数20ですから魔法数です。そのためバランスが崩れる程、中性子が多くても、安定な原子核があるのが分かります。

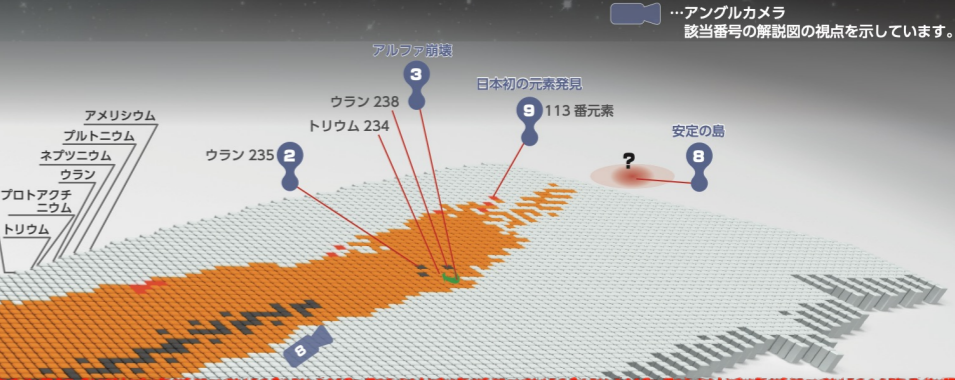


5 6 塊だけじゃない—色々な原子核



原子核はとても固く簡単には壊れません。しかし不安定核はより不安定な原子核へと自らを変化させます。その際に起きる現象を「崩壊」と呼びます。崩壊にはアルファ(α)、ベータ(β)、ガンマ(γ)の3種類あります。α崩壊は左上図のように、原子核からヘリウム原子核(陽子2、中性子2)が飛び出す現象です。当然軽くなり原子番号が2つ減ります。また飛び出したヘリウム原子核をα線と呼びます。β崩壊は左下図のように、原子核内の中性子が陽子に変化する反応です。その時、電子と反ニュートリノを放出します。重さはほぼ変わりませんが、陽子がふえますから、原子番号が1つ増えます。飛び出した電子をβ線と呼びます。なお崩壊した直後の原子核は興奮して熱くなっています。これが冷える時に光を出します。この光をγ線と呼びます。

今から137億年前、ビッグバンによって私たちの宇宙は生まれました。でもその時に存在した元素は水素とヘリウムだけ。それから3億年ほど経て星が生まれると、その中で重い元素が創られ始め、星の終焉に起きる超新星爆発では、より重い元素が一気に創られたと考えられています。私たちの体を含め、宇宙を構成する物質は全てこれらの元素から出来ています。一方、元素の本体は陽子と中性子からなる原子核です。陽子と中性子の微妙なバランスからなる原子核の成り立ちを調べることは、物質の起源を調べることにほかなりません。ここに示す核図表は全ての原子核を示した地図であり、元素合成と宇宙の歴史も刻まれています。原子核は果たしてどのように生まれたのか、またどのようなものなのか、核図表と一緒に見てみましょう。



あるのが分かります。陽子数(元素の種類) 中性子数(横軸は同じ元素)

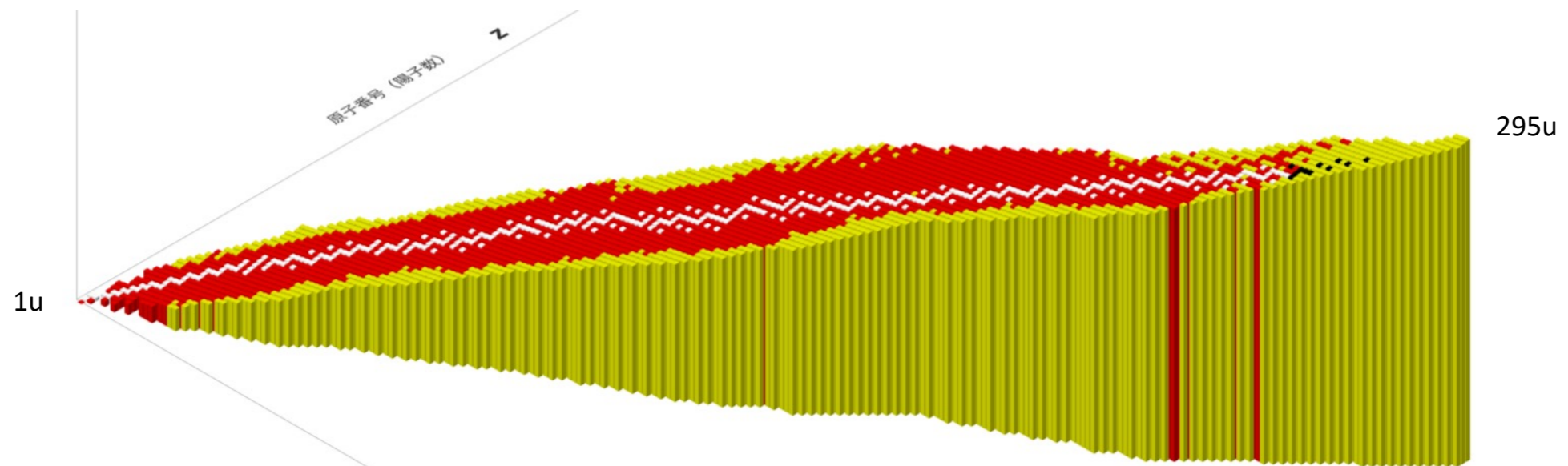
binding energy 結合エネルギー



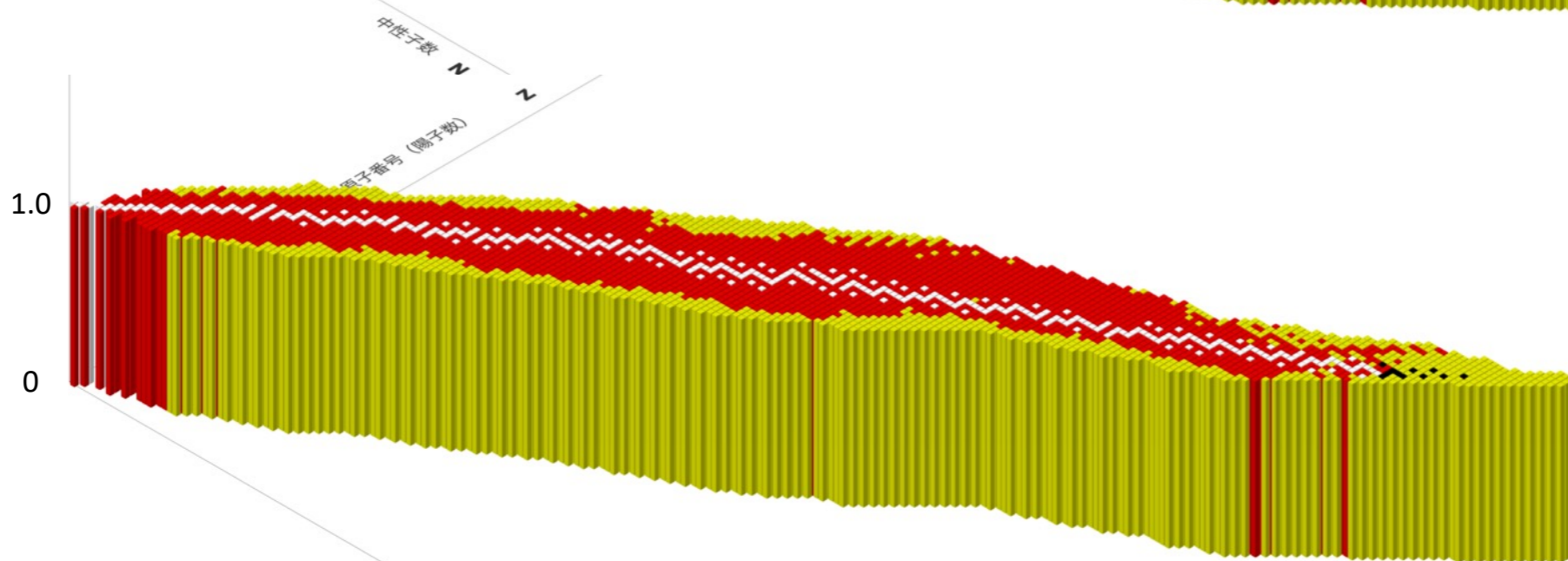
※1 「核図表」: 核図表中央の核図表は都合で高さの違うのが分かりますが、一般に「ハイゼンベルグの谷」は縦軸に上図のように深く凹谷で表現します。なお、深い谷にすると内側が見えなくなってしまう... というのが都合です。
 ※2,3 「陽子」「中性子」: 原子核はほぼ同じ重さの2種類の粒子で出来ています。プラス電荷を持つのが陽子、電荷を持たないのが中性子です。通常は原子核は電子で取りまわされていて、その電子はマイナス電荷を持ち重さは陽子の約2000分の1という軽さです。
 ※4 「同位元素」: 同位元素ともいいます。同位元素には放射性崩壊を起こす「放射性同位元素(RI)」と、崩壊しない「安定同位元素」があります。
 ※5 「ハイゼンベルグ」: ドイツの理論物理学者、ヴェルナー・カール・ハイゼンベルグ。行列力学と不確定性原理によって量子力学を完成させた一人です。
 ※6 「安定核」: 崩壊しない原子核のことを安定核といいますが、ただし、ウランのように半減期が地球の年齢より長くも長くないものも、ここでは安定核としています。
 ※7 「結合エネルギー」: 陽子と中性子がバラバラではなく、結合し原子核になったときに失われるエネルギーを結合エネルギーと呼びます。

Poster of a laboratory

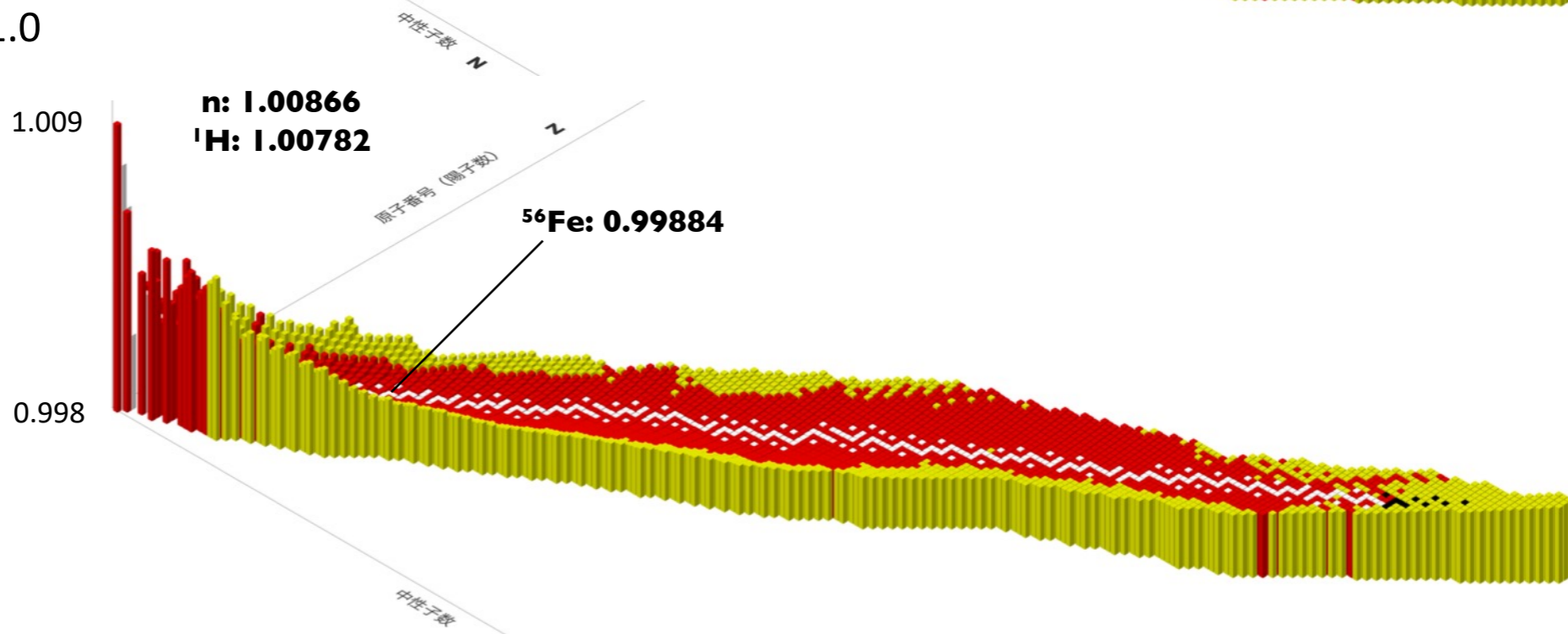
mass



mass/A

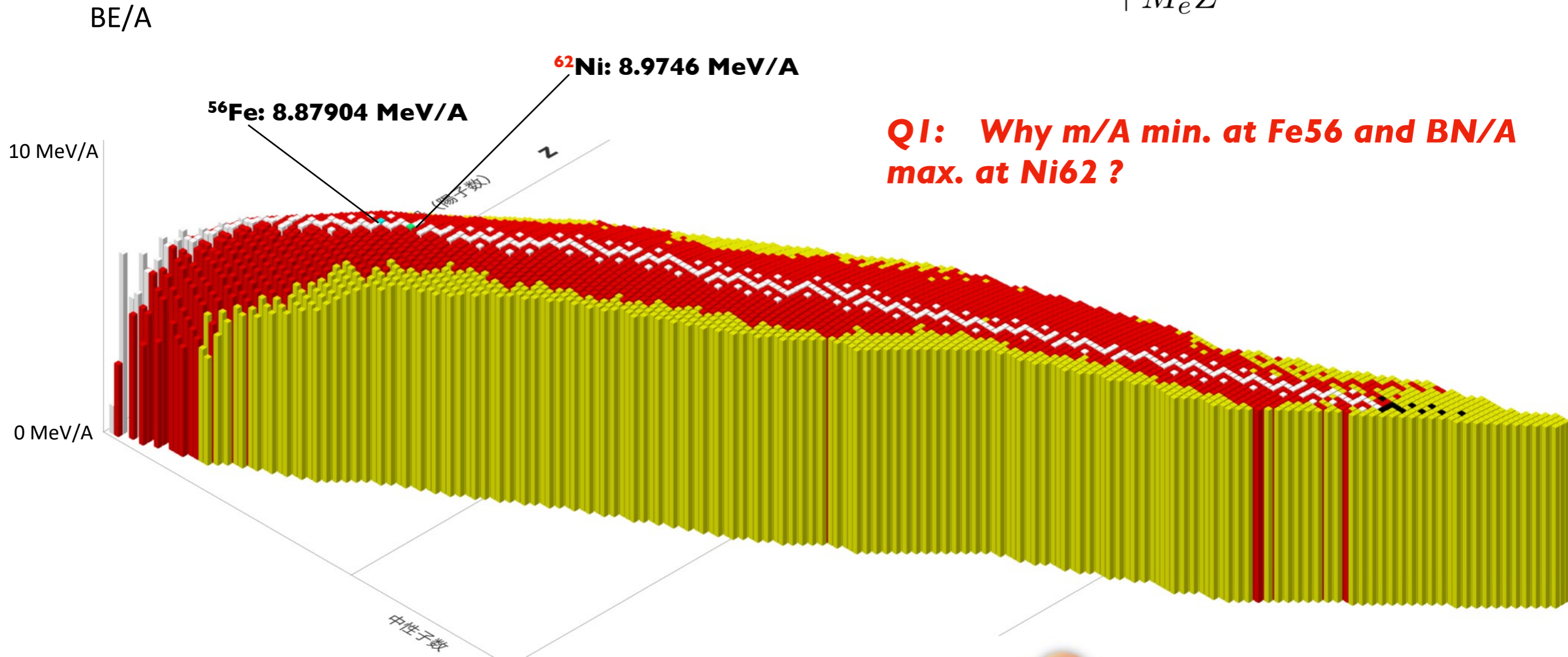


Expand m/A=1.0

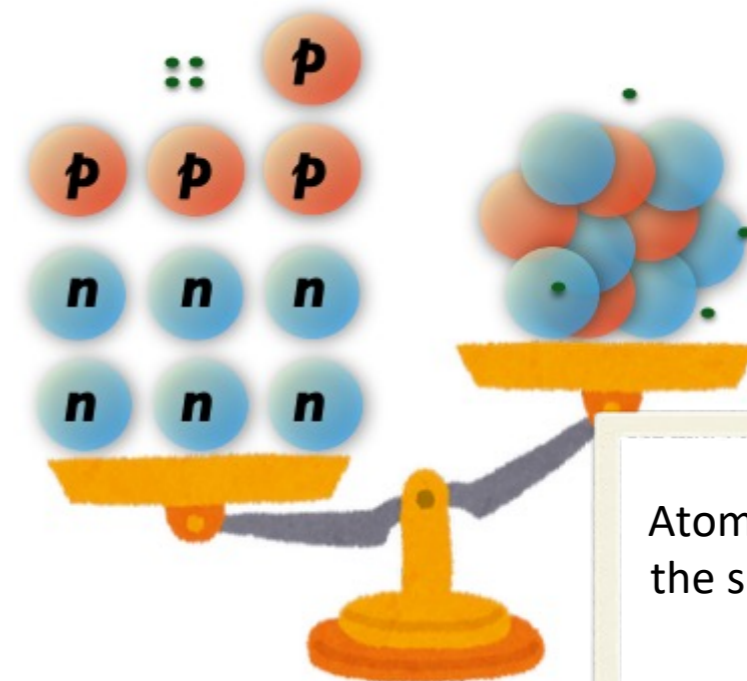
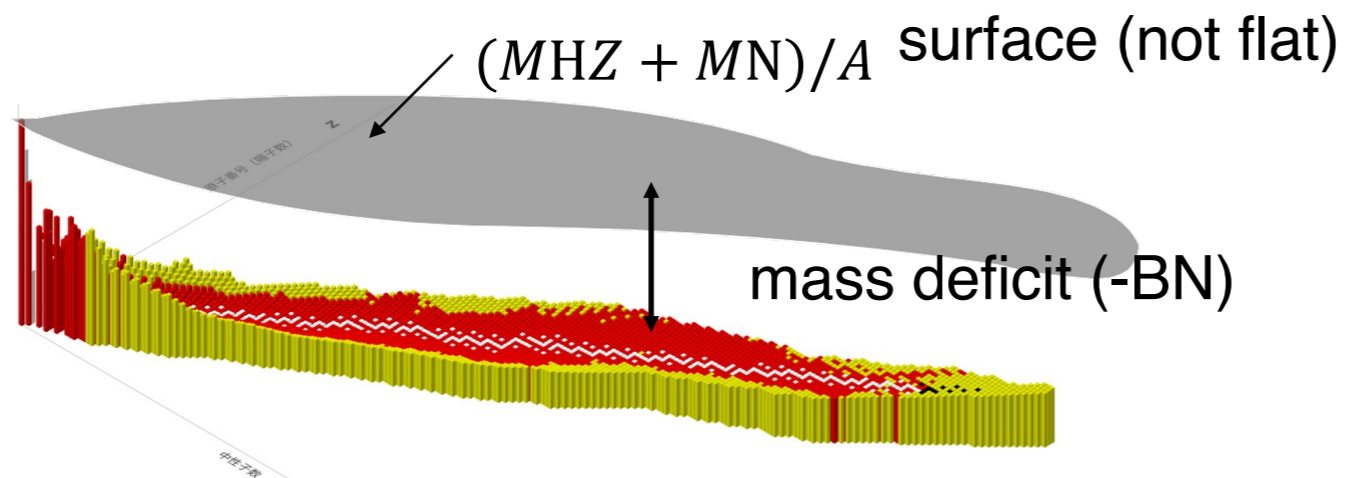


What is the Binding Energy(BE)?

$$B_{Z,N} = M_p Z + M_n N - M_{Z,N} + M_e Z$$



Q1: Why m/A min. at Fe56 and BN/A max. at Ni62 ?



Atom is ~1% lighter than the sum of constituents:
Mass deficit

What is the difference of mass deficit & mass excess?

Chat GPT 2023

Mass Deficiency and Mass Excess are terms used in the context of nuclear physics.

Mass deficiency refers to the phenomenon where an atomic nucleus is lighter than the sum of the masses of its constituent particles (protons and neutrons). This is because the nucleus possesses binding energy, and energy and mass are mutually convertible. Mass deficiency is explained based on

$E=mc^2$ (Einstein's mass-energy equivalence).

~~Mass excess refers to the situation where **the actual mass of an atomic nucleus is greater than the sum of the masses** of the protons and neutrons that make it up. This excess mass indicates that the nucleus possesses binding energy.~~

Chat GPT 2024/7/30

Mass Deficit

Definition: The mass deficit of a nucleus is the difference between the total mass of its individual protons and neutrons (when they are free and not bound in the nucleus) and the actual mass of the nucleus.

Mass Excess

Definition: The mass excess of a nucleus is the difference between the actual mass of the nucleus and its mass number (A) expressed in atomic mass units (u).

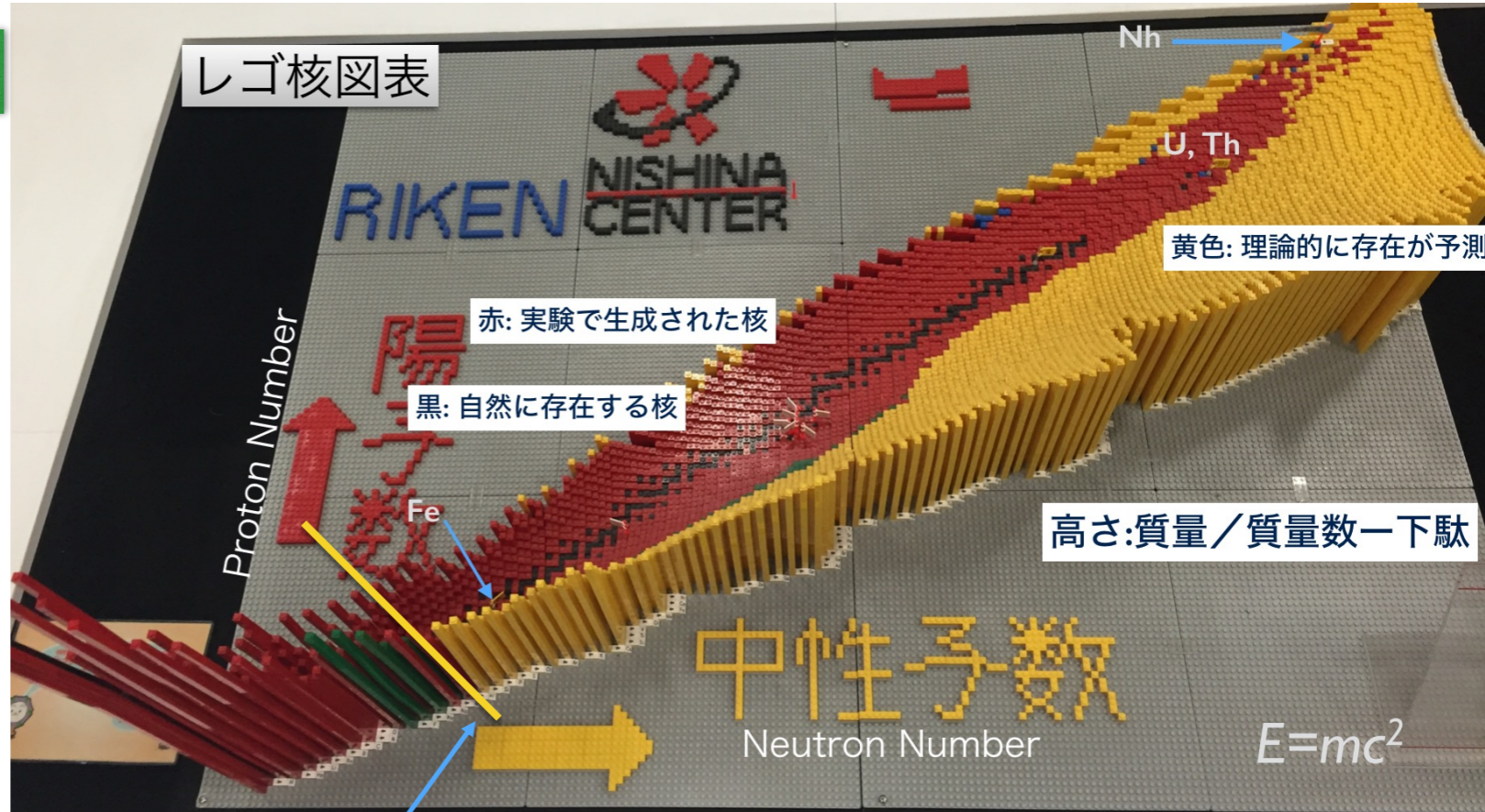
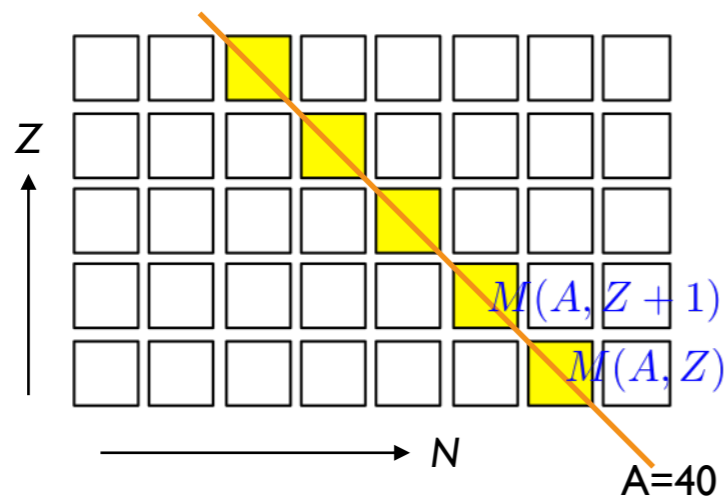
Mass excess is just for convenience.

$$m(^{238}\text{U}) = 238.0507869(16) \text{ u} \quad \text{ME}(^{238}\text{U}) = 0.0507869(16) \text{ u} \quad \text{or} \quad \text{ME}(^{238}\text{U}) = 47307.7(1.5) \text{ keV}$$

Comparison with neighborhood

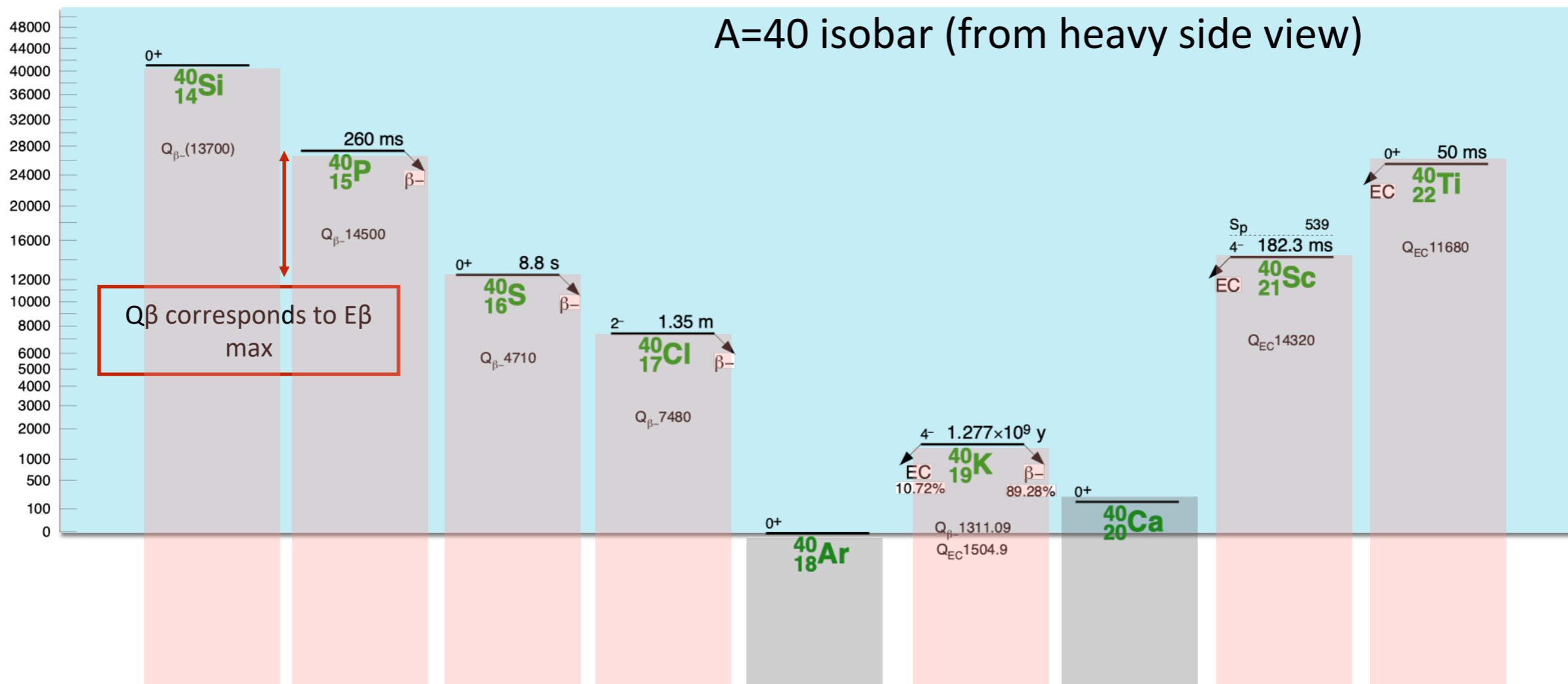
in isobar : beta stability

$$Q_{\beta^-} = M(A, Z) - M(A, Z + 1)$$




Relative mass with 40Ar [keV/c²]

A=40 isobar (from heavy side view)




Comparison with neighborhood 2

separation energies



$$S_p = M(N, Z - 1) + M_p - M(N, Z)$$

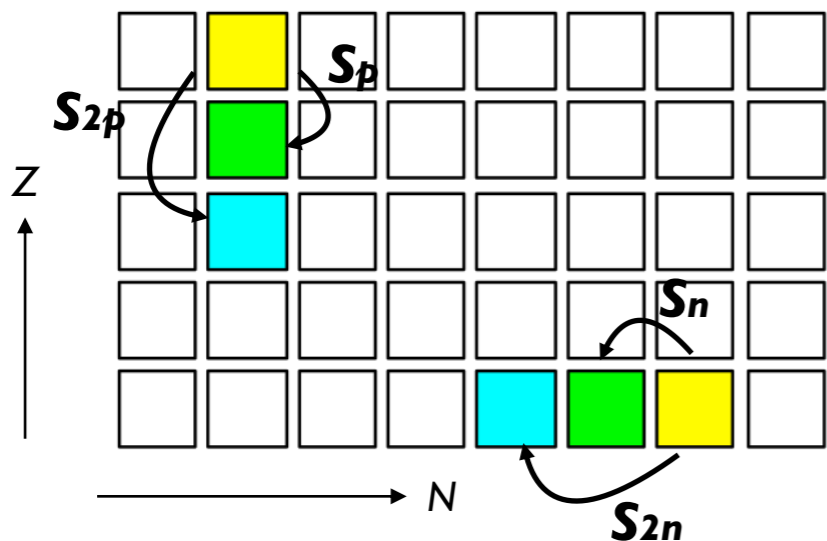
proton separation energy

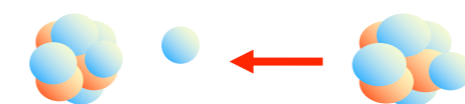


$$S_{2p} = M(N, Z - 2) + 2M_p - M(N, Z)$$

two proton separation energy


Separation energy = 0: Drip line





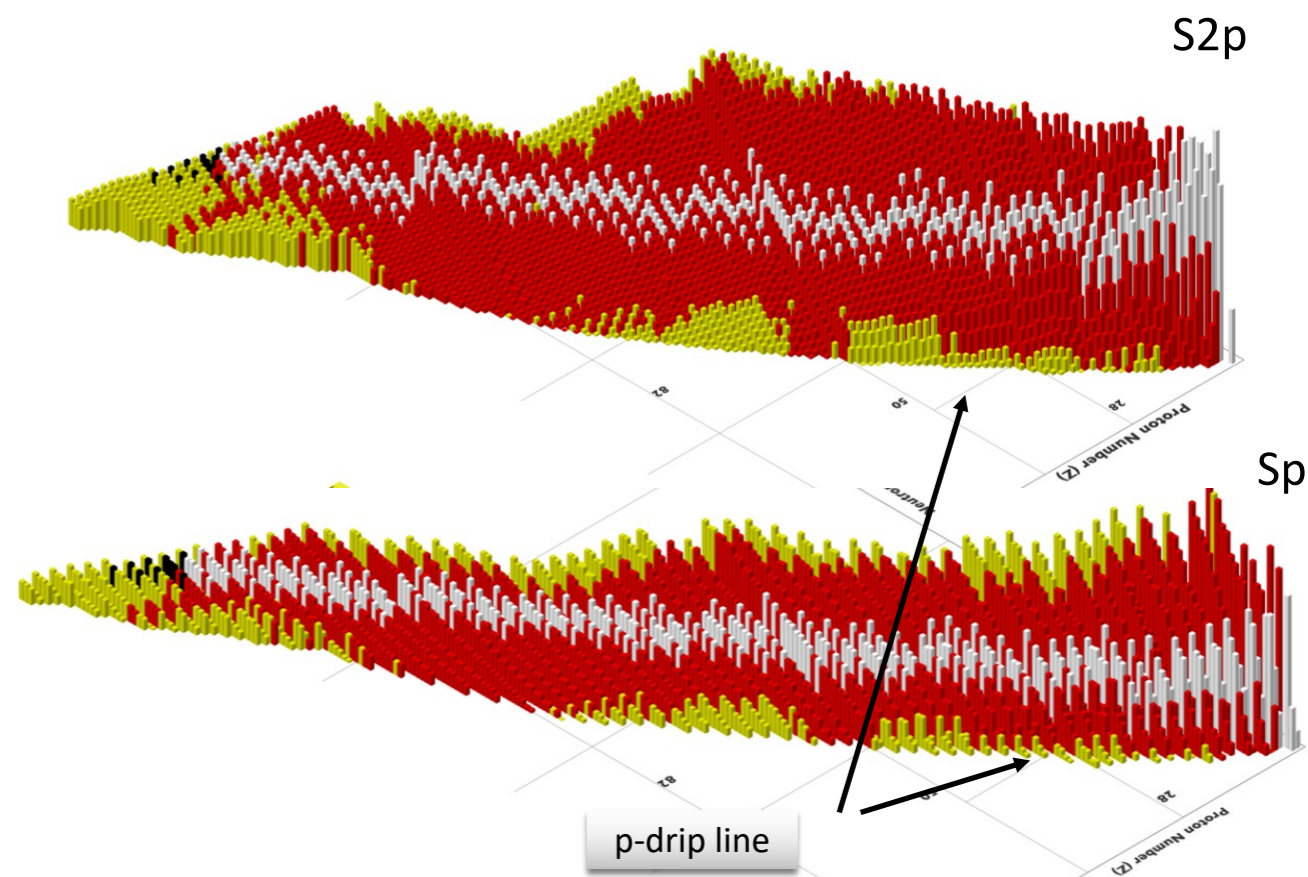
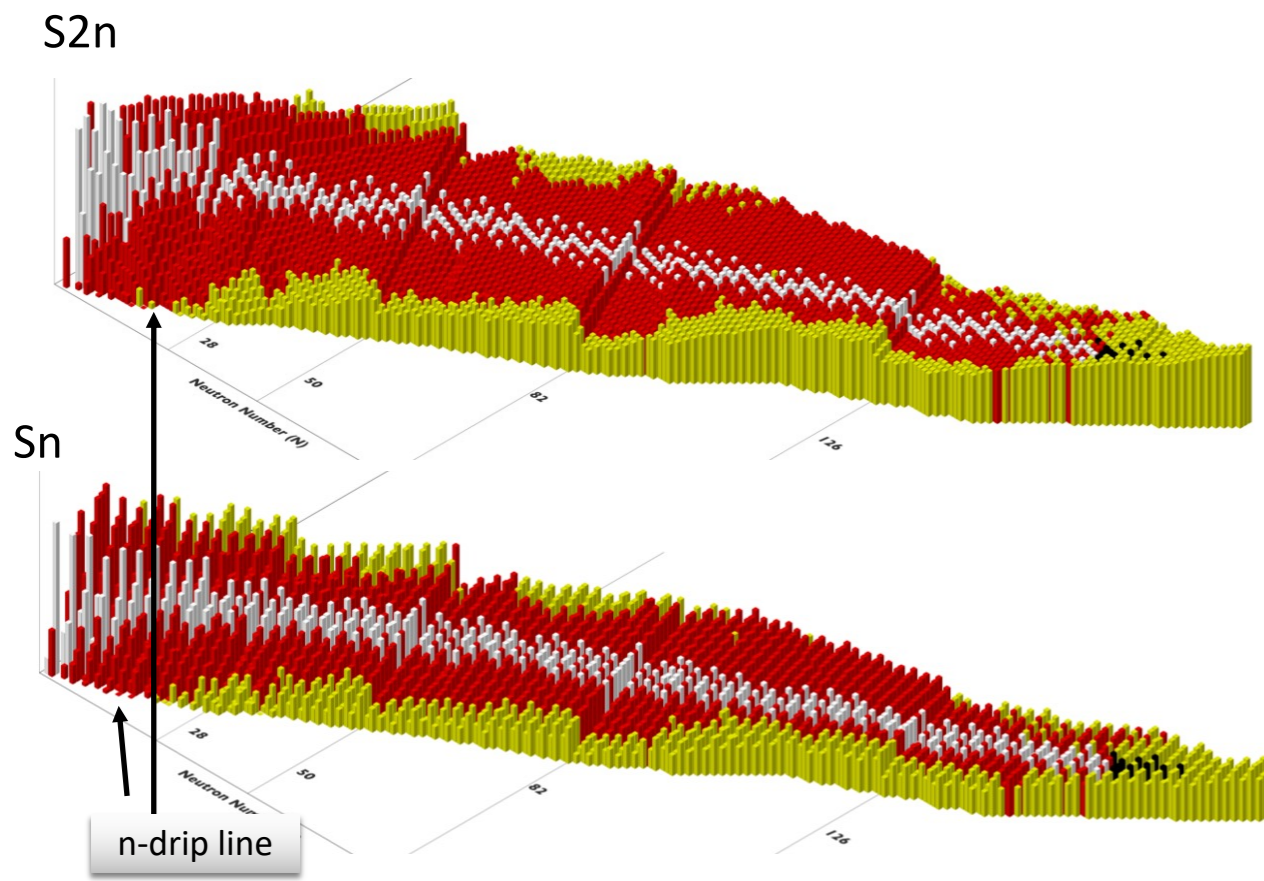
$$S_n = M(N - 1, Z) + M_n - M(N, Z)$$

neutron separation e.



$$S_{2n} = M(N - 2, Z) + 2M_n - M(N, Z)$$

two neutron sep. e.



r-process

Nuclides in Solar System

~83 elements, ~255 nuclides

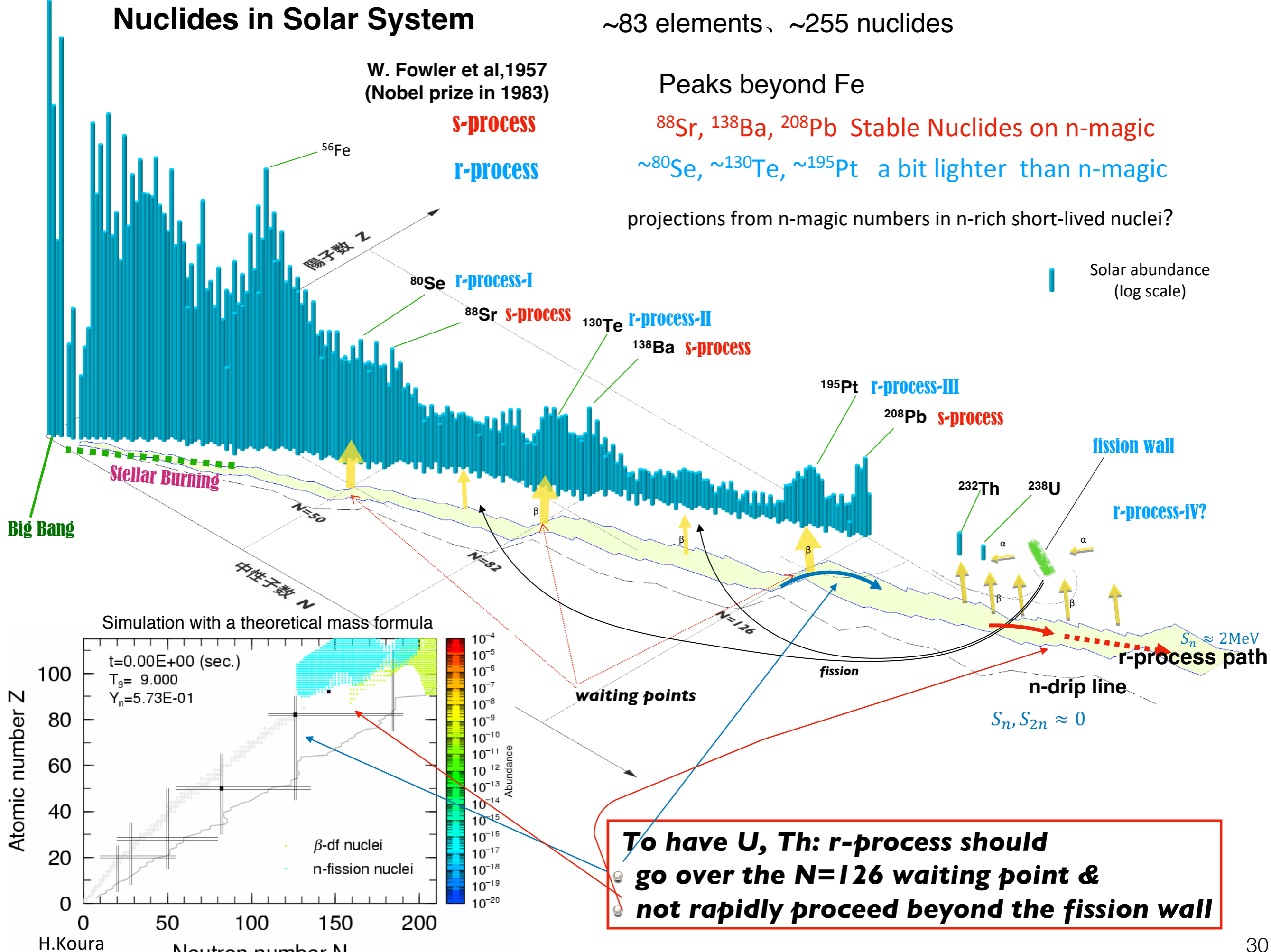
W. Fowler et al, 1957
(Nobel prize in 1983)

Peaks beyond Fe

^{88}Sr , ^{138}Ba , ^{208}Pb Stable Nuclides on n-magic

$\sim^{80}\text{Se}$, $\sim^{130}\text{Te}$, $\sim^{195}\text{Pt}$ a bit lighter than n-magic

projections from n-magic numbers in n-rich short-lived nuclei?



To have U, Th: r-process should go over the $N=126$ waiting point & not rapidly proceed beyond the fission wall

r-process (equilibrium model)

High Temp.

Clearing up

Cool down

- 1) $(n, \gamma) \leftrightarrow (\gamma, n)$ equilibrium
- $$\frac{Y(Z, A+1)}{Y(Z, A)} \propto \rho_n \exp \frac{S_n(Z, A+1)}{kT}$$
- 2) β -decay ($Z=Z+1$)

S_n (Mass) is essential

$T_{1/2}$ is essential

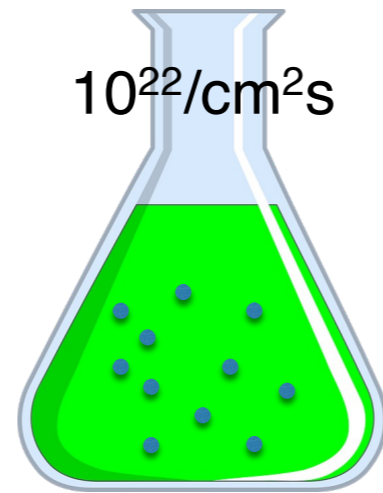
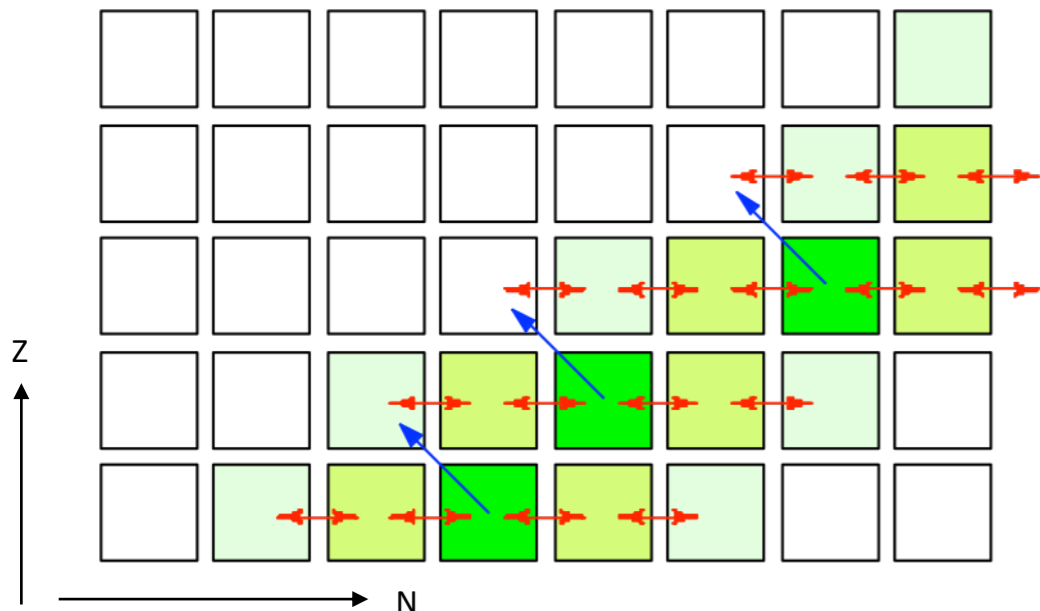
non-equilibrium, intermediate
($A=A+1, A-1$)

S_n, σ_n

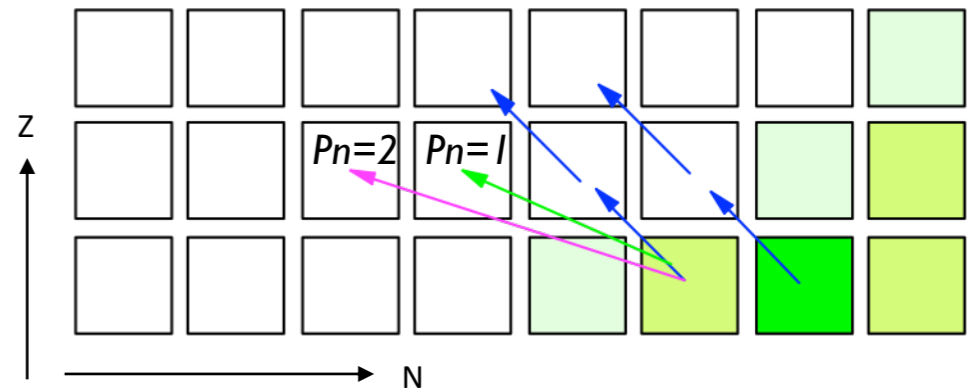
cross sections appear

β -Decay ($Z=Z+1$)
($A=A, A-1, A-2$)
 β -delayed neutron

P_n



In equilibrium, abundance is given by S_n and Temp.
(Chemical process with neutron water)



$(n, \gamma) \leftrightarrow (\gamma, n)$

β -decay ($Z=Z+1$)

$$\exp \frac{\mu}{kT}$$

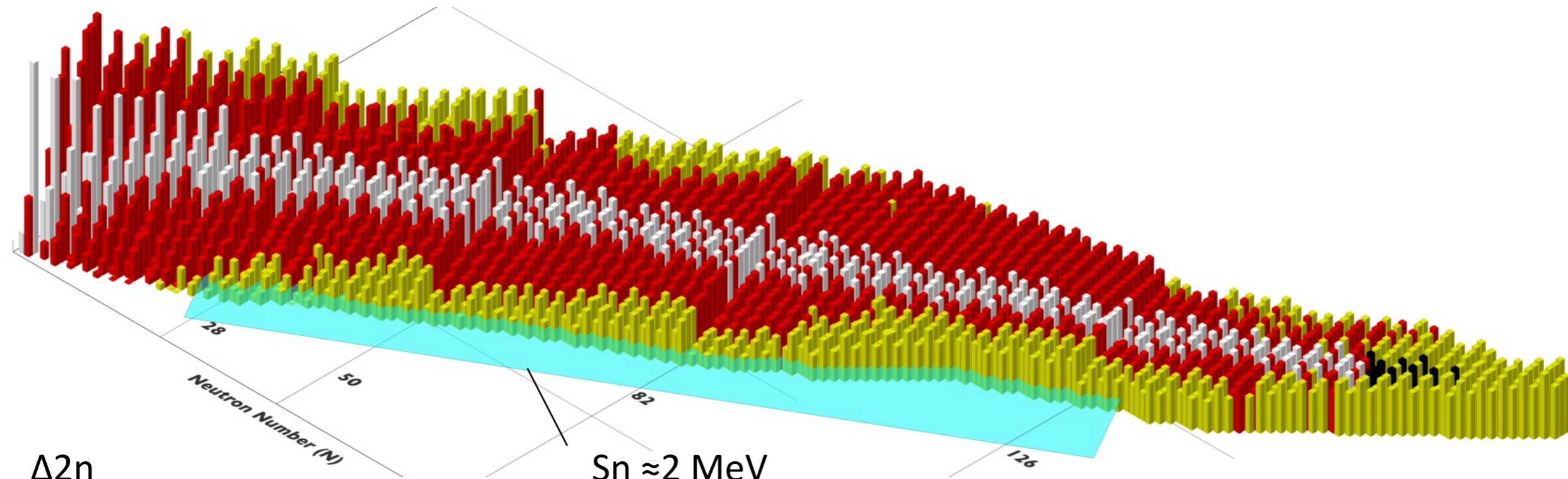
chemical potential μ

neutron separation energy S_n

$S_n \sim 2$ MeV is the r-process path

r-process path and waiting point (neutron-magic)

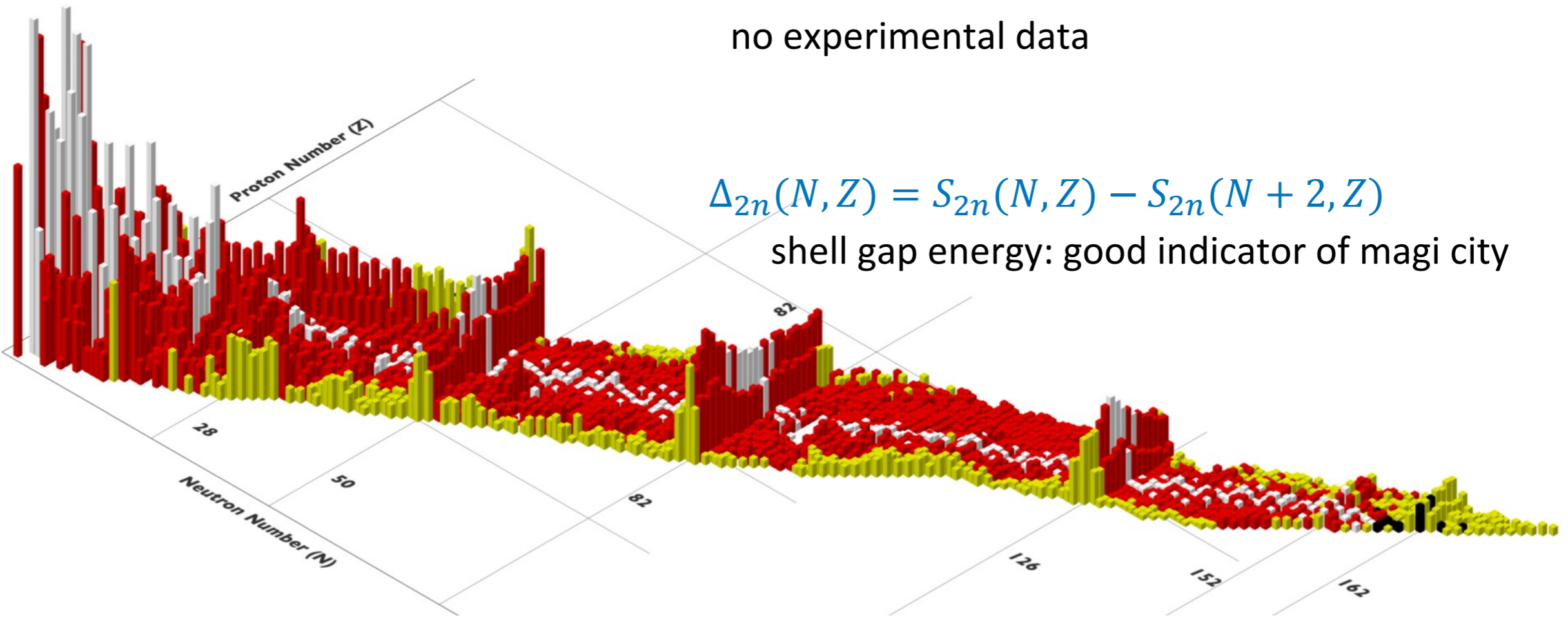
Sn



Sn ≈ 2 MeV

no experimental data

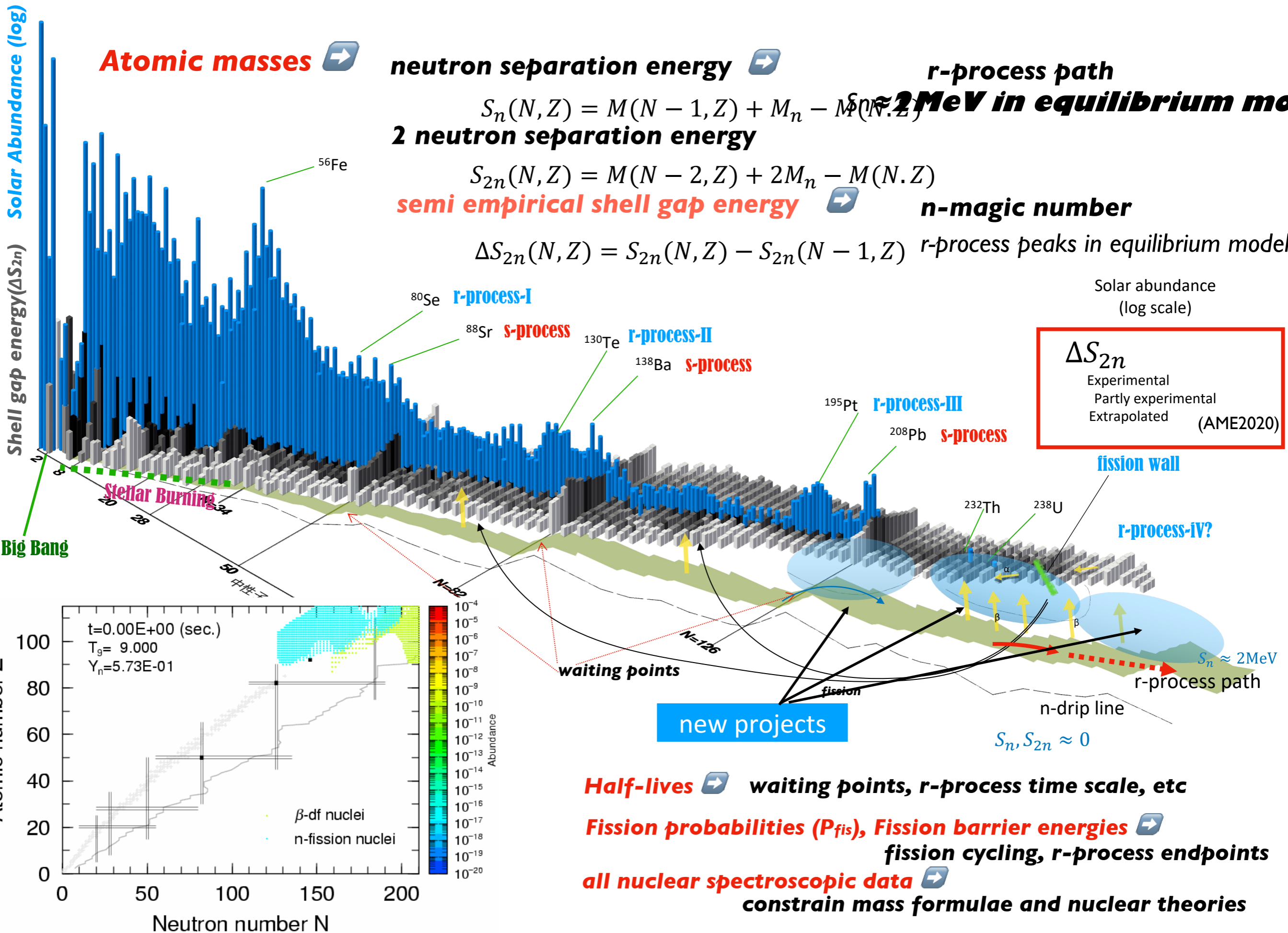
Δ2n



$$\Delta_{2n}(N, Z) = S_{2n}(N, Z) - S_{2n}(N + 2, Z)$$

shell gap energy: good indicator of magicity

Solar abundances and atomic masses (ΔS_{2n})



r-process dynamic calculation

Thomas Rauscher, Essentials for Nucleosynthesis and Theoretical Nuclear Astrophysics, Iop Publishing Ltd, (2020)

$$\begin{aligned}
 \frac{dY_{Z,N}}{dt} = & \rho N_A Y_n \langle \sigma v \rangle_{Z,N-1}^{(n,\gamma)} Y_{Z,N-1} - \rho N_A Y_n \langle \sigma v \rangle_{Z,N}^{(n,\gamma)} Y_{Z,N} + \lambda_{Z,N+1}^{(\gamma,n)} Y_{Z,N+1} - \lambda_{Z,N}^{(\gamma,n)} Y_{Z,N} \\
 & + \sum_{k_n \geq 0} \lambda_{Z-1,N+1+k_n}^{\beta,k_n} Y_{Z-1,N+1+k_n} - \sum_{k_n \geq 0} \lambda_{Z,N}^{\beta,k_n} Y_{Z,N} \\
 & + \rho N_A Y_n \sum_{Z',N'} \sum_{k_n \geq 0} \langle \sigma v \rangle_{Z',N'}^{nf,Z,N,k_n} Y_{Z',N'} - \rho N_A Y_n \sum_{k_n \geq 0} \langle \sigma v \rangle_{Z,N}^{nf,k_n} Y_{Z,N} \\
 & + \sum_{Z',N'} \sum_{k_n \geq 0} \lambda_{Z',N'}^{\beta df,Z,N,k_n} Y_{Z',N'} - \sum_{k_n \geq 0} \lambda_{Z,N}^{\beta df,k_n} Y_{Z,N},
 \end{aligned}$$

n-capture

photo dissociation

β-decay, β-delayed neutron

n-induced fission

β-delayed fission

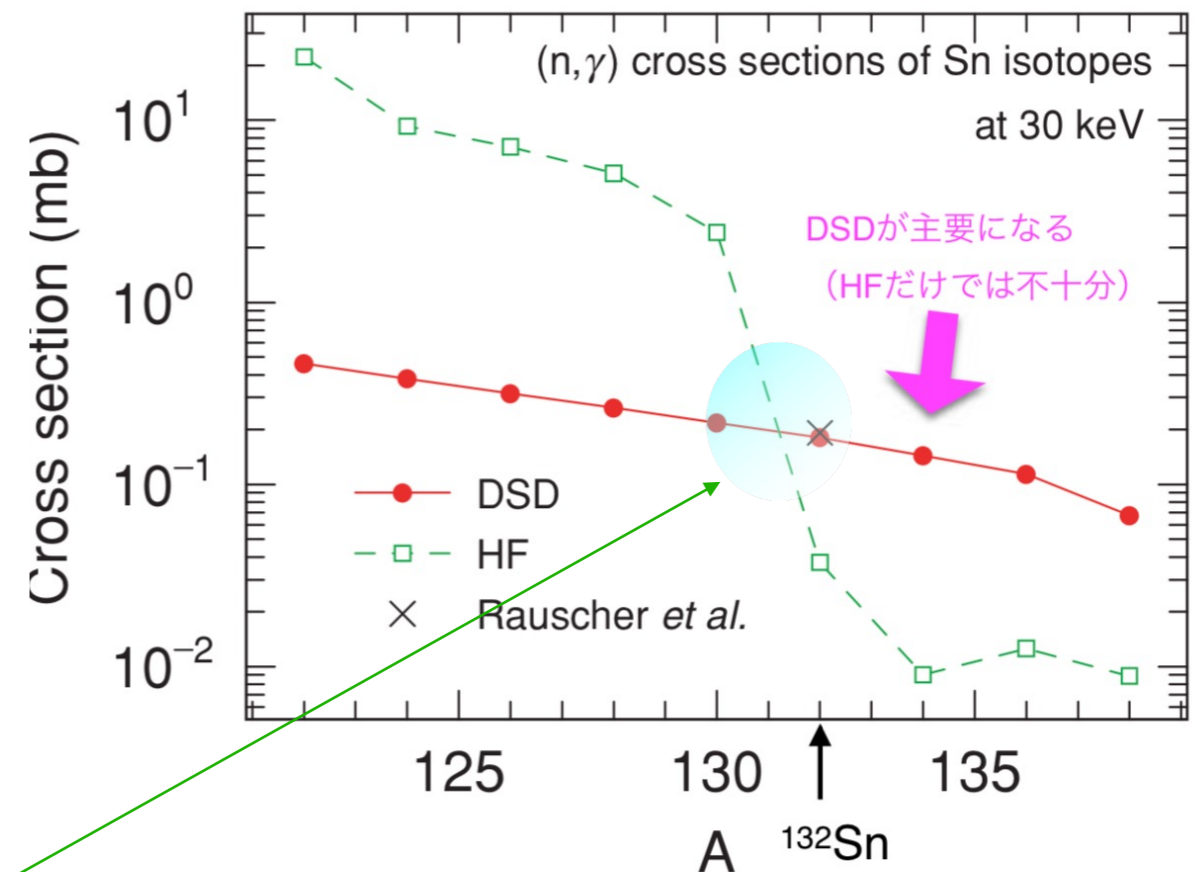
$\langle \sigma v \rangle$ calculation required

σ : Hauser-Feshbach statistical model
Direct-semi-direct reaction (DSD) etc

v : sensitive to resonant state

Mass is essential, but not only mass

Sn: rapid decrease observed



S.Chiba, H. Koura, T. Hayakawa, T. Kawano *et al.*, PRC77 015809 (2008)

●単一粒子準位計算はmodified WS pot (Koura, Yamada, 2000)より

https://www2.yukawa.kyoto-u.ac.jp/~rp2019/slides/190523_09_koura.pdf

A big news in 2017

gravitation wave from n-star merger

金やプラチナ大量生成 中性子星合体の重力波観測で (2017/10/17 11:55)



ツイート mixi チェック シェア B! 使い方は?

「中性子星」と呼ばれる重力の強い星と星が衝突して合体したことを重力波と光で観測することに成功したとアメリカなどの研究チームが発表しました。世界初の快挙です。

アメリカなどの研究チームは今年8月、地球から1億3000万光年離れた宇宙で、2つの中性子星が衝突して合体した際に生じた重力波を観測しました。この観測を受け、国立天文台など日本の研究チームは衝撃で生じた光を半月にわたって観測しました。この光を分析した結果、中性子星の合体により、金やプラチナなどの重元素が大量に作られたことが分かったということです。

They observe the moment of Au, Pt are born

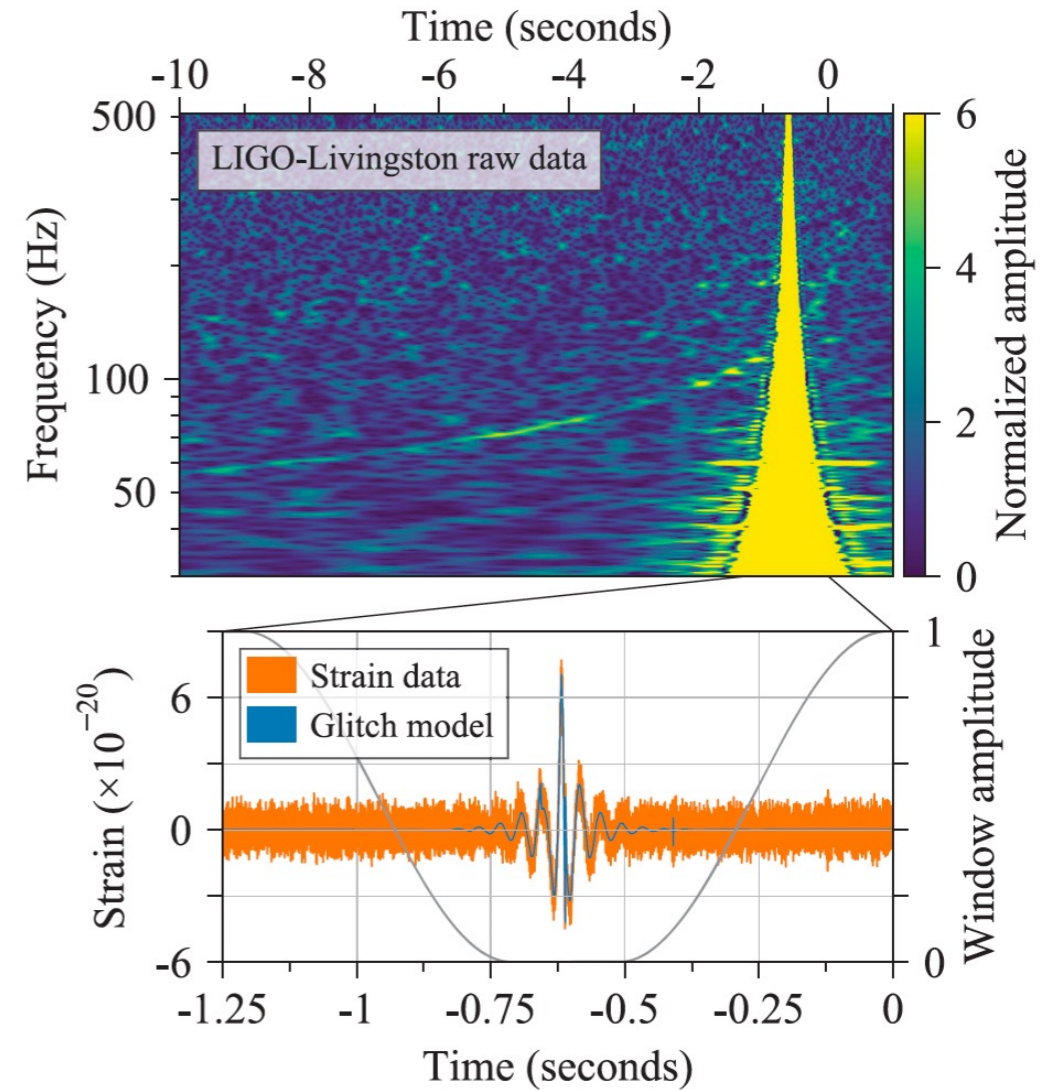
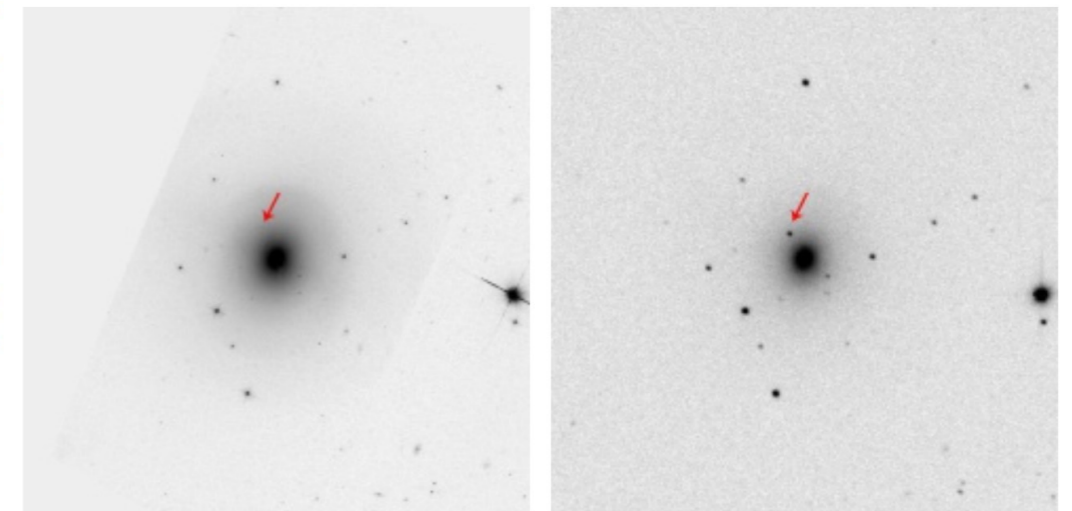


FIG. 2. Mitigation of the glitch in LIGO-Livingston data. Times are shown relative to August 17, 2017 10:41:04 UTC. Top panel

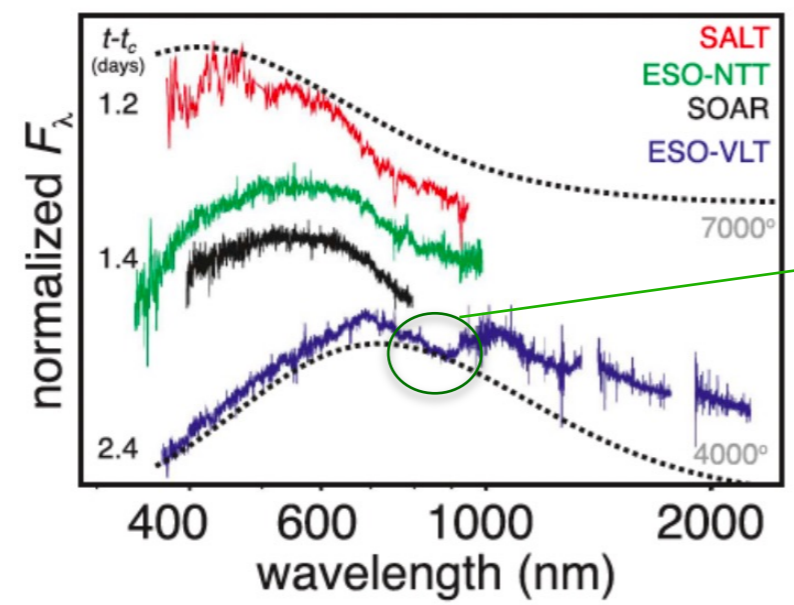
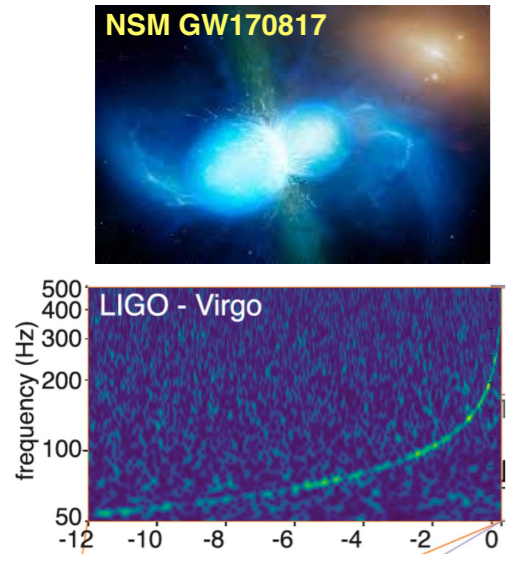


[画像のクリックで拡大表示]

[画像のクリックで拡大表示]

新たな重力波が検出される4カ月前にハッブル宇宙望遠鏡がとらえた楕円形の銀河「NGC4993」(左)。一方、チリのスウォープ望遠鏡の画像(右)には、2017年8月に現れた明るい点が見える。(PHOTOGRAPH BY HUBBLE/STSCI (LEFT) AND

Multi-messenger astronomy after n-star merger



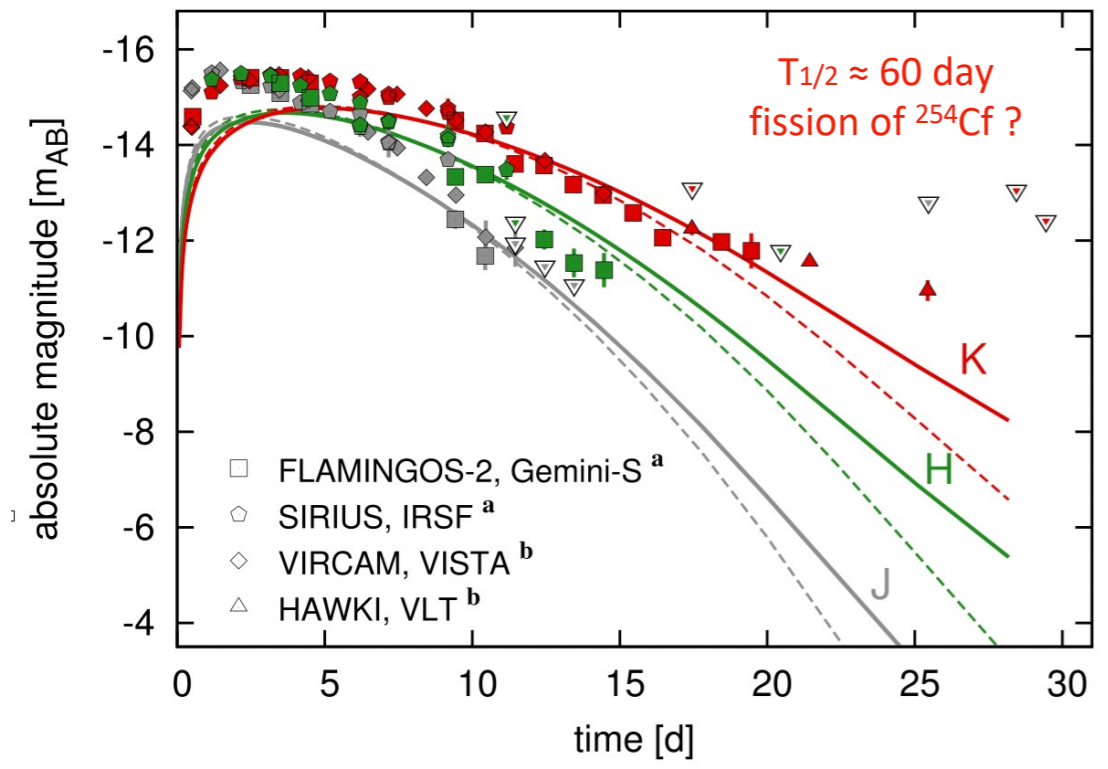
absorption of Sr II ?

Optical spectroscopy of remnant of NSM, kilo nova (r-process nova), wavelength, time profiles agree with predicted r-process !

²⁵³ Fm 3.00 d	²⁵⁴ Fm 3.240 h	²⁵⁵ Fm 20.07 h	²⁵⁶ Fm 2.627 h	²⁵⁷ Fm 100.5 d	²⁵⁸ Fm 370 μs	²⁵⁹ Fm 1.5 s	²⁶⁰ Fm 4 ms
²⁵² Es 471.7 d	²⁵³ Es 20.47 d	²⁵⁴ Es 275.7 d ★1.638 d	²⁵⁵ Es 39.8 d	²⁵⁶ Es ★7.6 h 25.4 m	²⁵⁷ Es 7.7 d	²⁵⁸ Es f28.0 y 40.1 m α 59.8 y	²⁵⁹ Es f13.8 y 6.78 d α 128 y
²⁵¹ Cf 898 y	²⁵² Cf 2.645 y	²⁵³ Cf 17.81 d	²⁵⁴ Cf 60.5 d	²⁵⁵ Cf 1.4 h	²⁵⁶ Cf 12.3 m	²⁵⁷ Cf f278 y 2.39 d α 5480 y	²⁵⁸ Cf f5.76 d α 7800 y
²⁵⁰ Bk 3.212 h	²⁵¹ Bk 55.6 m	²⁵² Bk 1.8 m	²⁵³ Bk 1.60 d α 1120 y	²⁵⁴ Bk 3.70 m	²⁵⁵ Bk 23.4 m	²⁵⁶ Bk 39.7 s	²⁵⁷ Bk 1.63 m
²⁴⁹ Cm 1.069 h	²⁵⁰ Cm 8300 y	²⁵¹ Cm 16.8 m	²⁵² Cm f32.3 y α 1.74·10 ⁶ y	²⁵³ Cm 23.5 m	²⁵⁴ Cm f33.5 y 42.9 m	²⁵⁵ Cm 1.66 m	160

β-delayed fission?

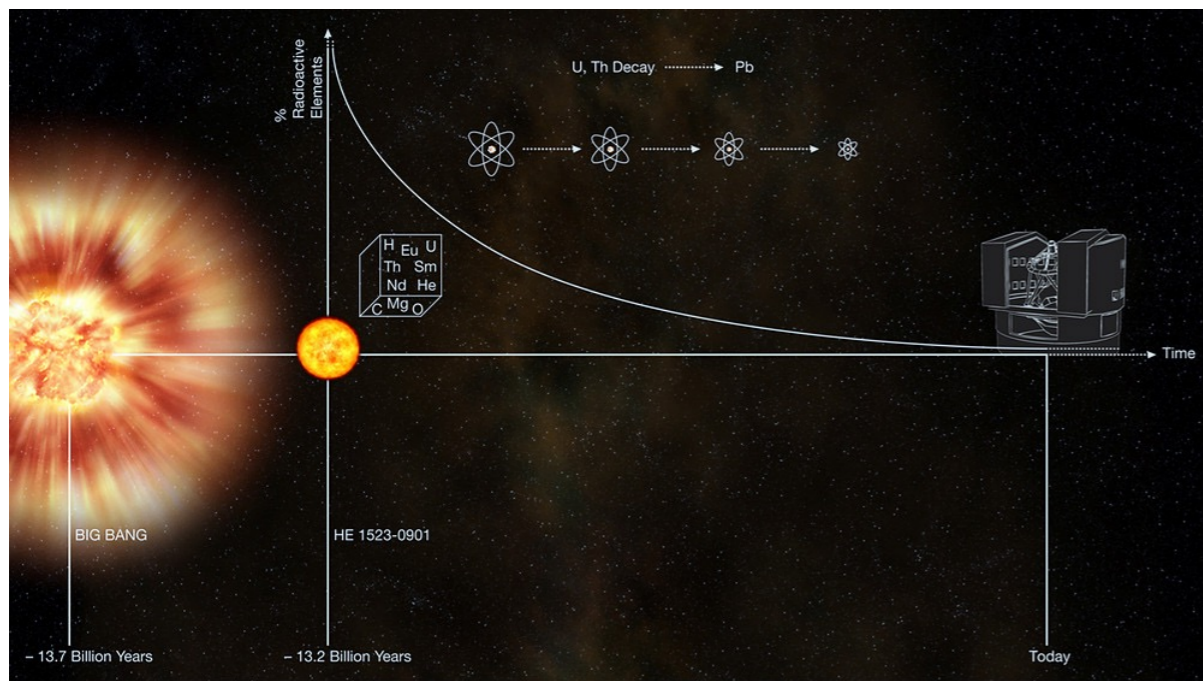
We will provide mass and decay properties of the progenitors of ²⁵⁴Cf



Zhu et al. The Astrophysical Journal Letters, 863:L23 (6pp), 2018 August 20

Wanajo et al. The Astrophysical Journal Letters, 789:L39 (6pp), 2014 July 10 predicted before NSM in 2017

Cosmochronometry



The Cosmic Clock
(Artist's Impression)

ESO Press Photo 23a/07 (10 May 2007)

This image is copyright © ESO. It is released in connection with an ESO press release and may be used by the press on the condition that the source is clearly indicated in the caption.



Using ^{232}Th ($T_{1/2}=14.0$ Gyr) and ^{238}U ($T_{1/2}=4.47$ Gyr)

age of a star can be:

$$t = 46.67\text{Gyr} [\log(\text{Th}/\text{Eu})_{\text{init}} - \log(\text{Th}/\text{Eu}_{\text{obs}})]$$

$$t = 14.83\text{Gyr} [\log(\text{U}/\text{Eu})_{\text{init}} - \log(\text{U}/\text{Eu}_{\text{obs}})]$$

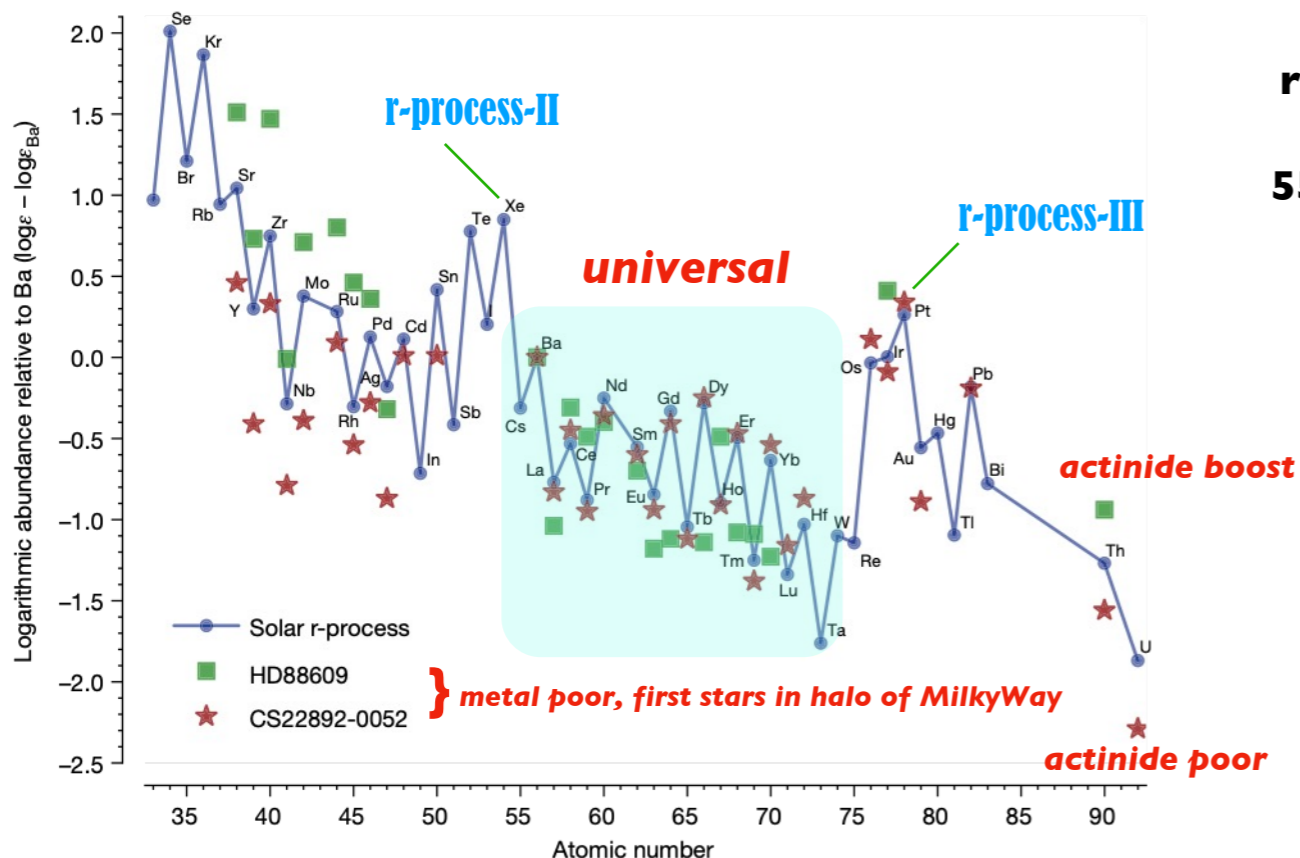
$$t = 21.80\text{Gyr} [\log(\text{U}/\text{Th})_{\text{init}} - \log(\text{U}/\text{Th}_{\text{obs}})]$$

**initial ratios are determined from
r-process model**

If we use bare theoretical mass formulae, the age can be varied
 $t > 14$ Gyr (greater than the age of universe),
 $t < 0$ Gyr (negative age !)

We will provide constraints for more accurate mass formulae

Universality? of r-process



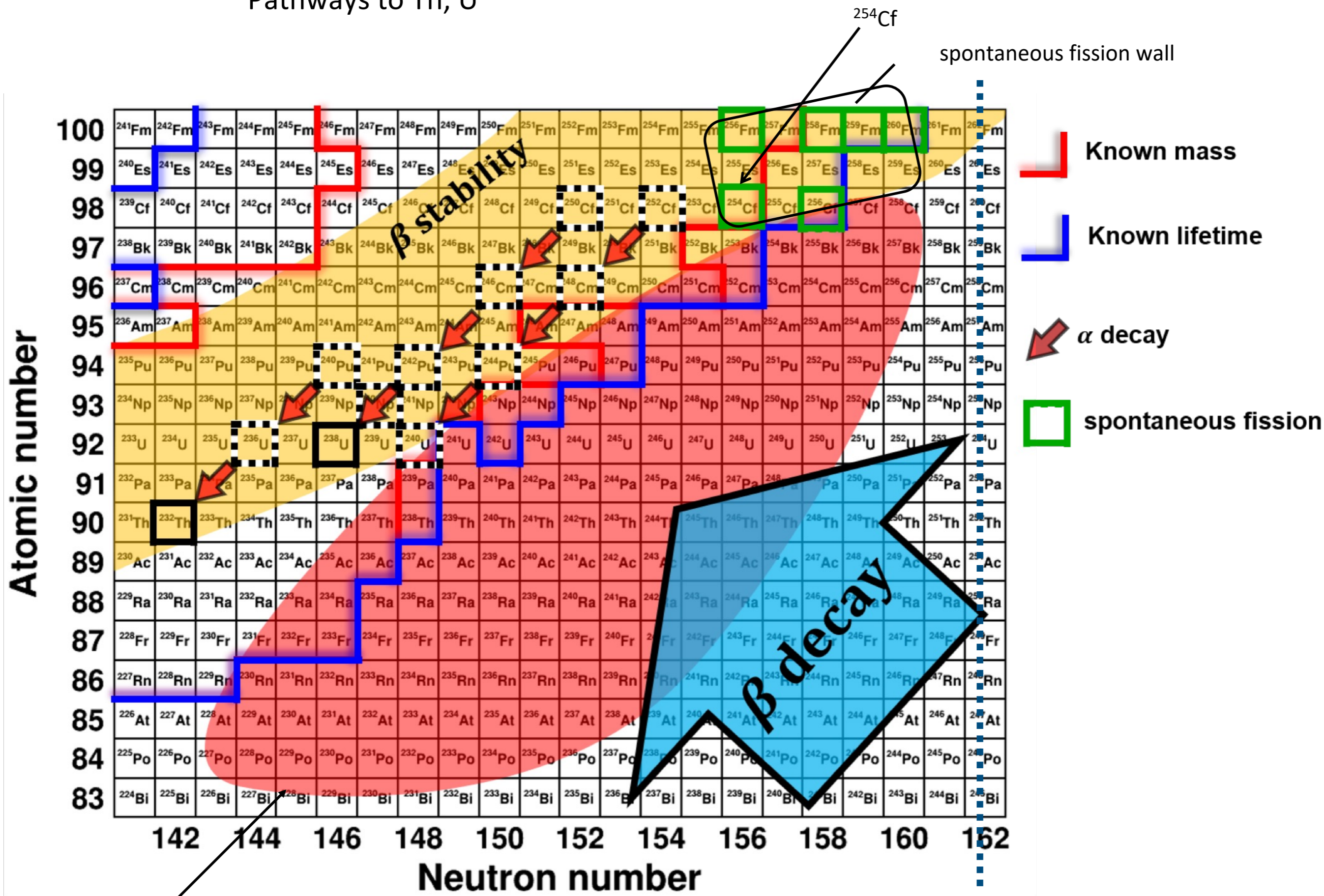
r-process has been considered to be “universal”,

55 < A < 75 is universal, but “actinide boost” and “actinide poor” stars are found in metal poor stars (first stars in halo of our galaxy)

- *not all r-process go over N=126 waiting point?*
- *some r-process rapidly proceed beyond fission wall?*
- *part of 55<A<75 nuclides are considered to be from fission fragments, why they are universal?*

We will provide key experimental data

Pathways to Th, U



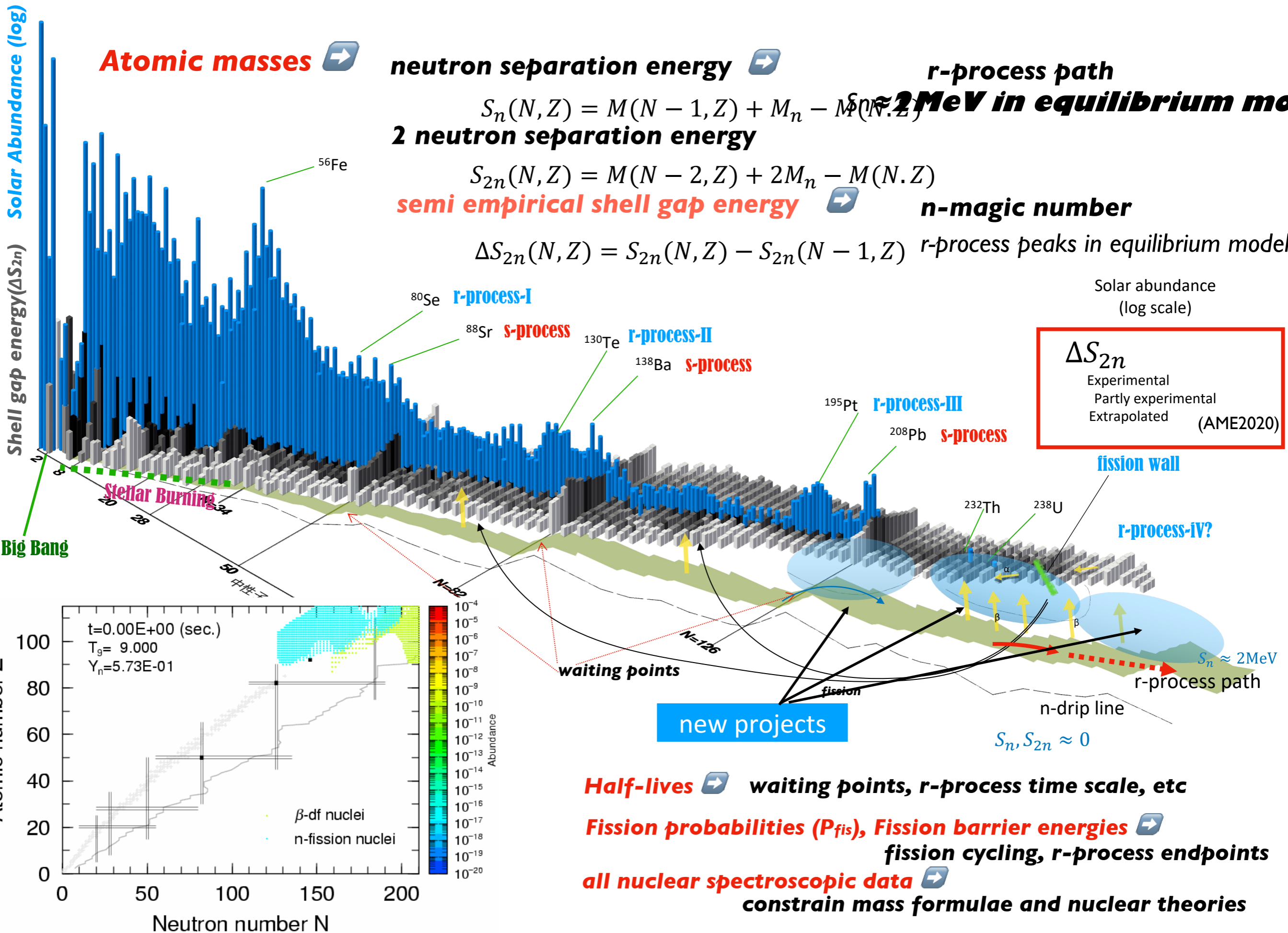
We will access these nuclides

N=162 semi-magic number?

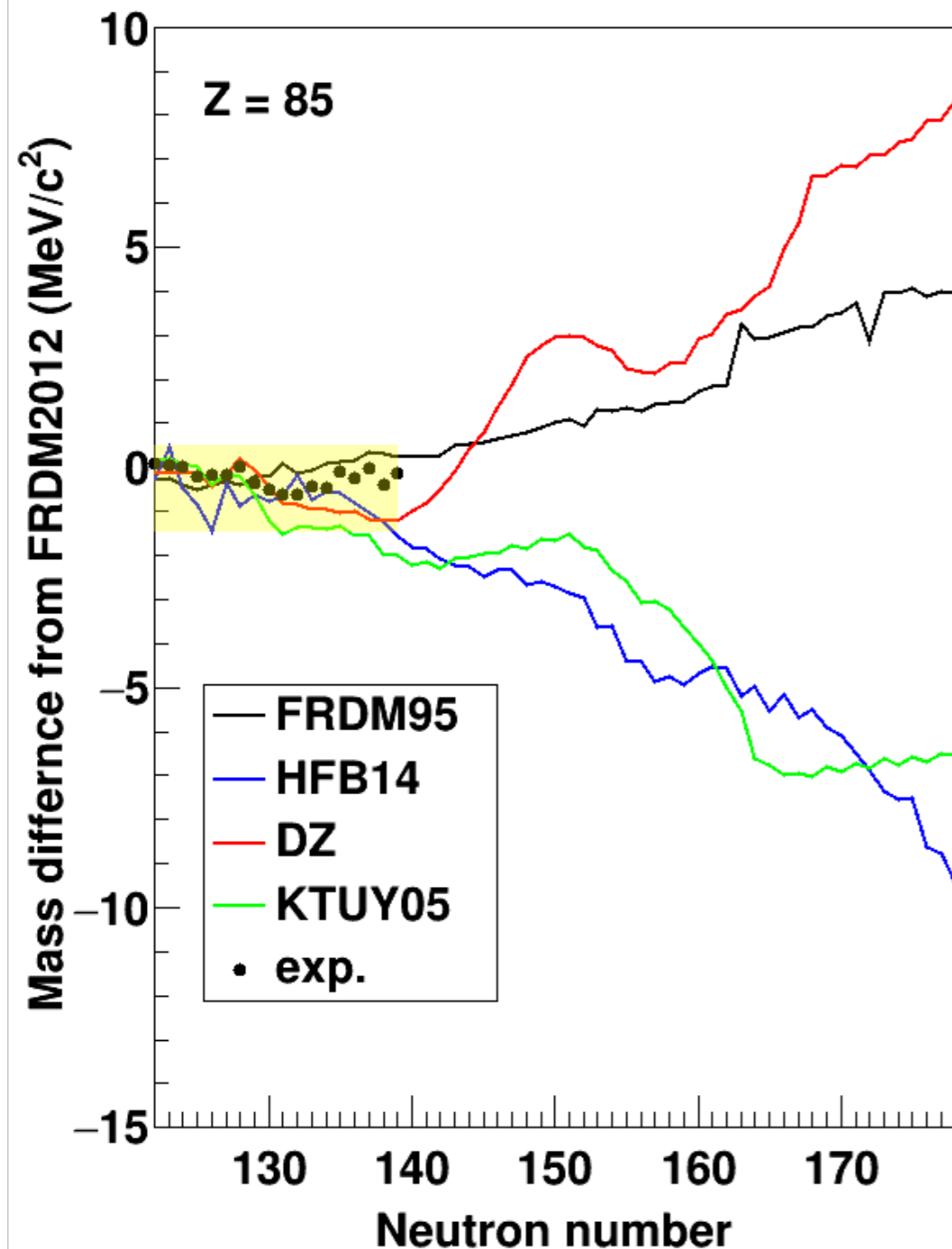
end of part I

part -II

Solar abundances and atomic masses (ΔS_{2n})



Theoretical mass predictions are scattered very much



Experimentally observable data for r-process study

Mass:

- *r*-process path
- waiting points

$T_{1/2}$:

- *r*-process time scale
- waiting points

P_{fission} :

- fission cycling
- *r*-process termination
- final abundance

$P_{\text{delayed-neutron}}$:

- final abundance

Isomeric state:

- *r*-process path
- final abundance

Any nuclear data constrain mass formulae and nuclear theories

Masses to be measured

Experimentally Synthesized: ≈ 3300

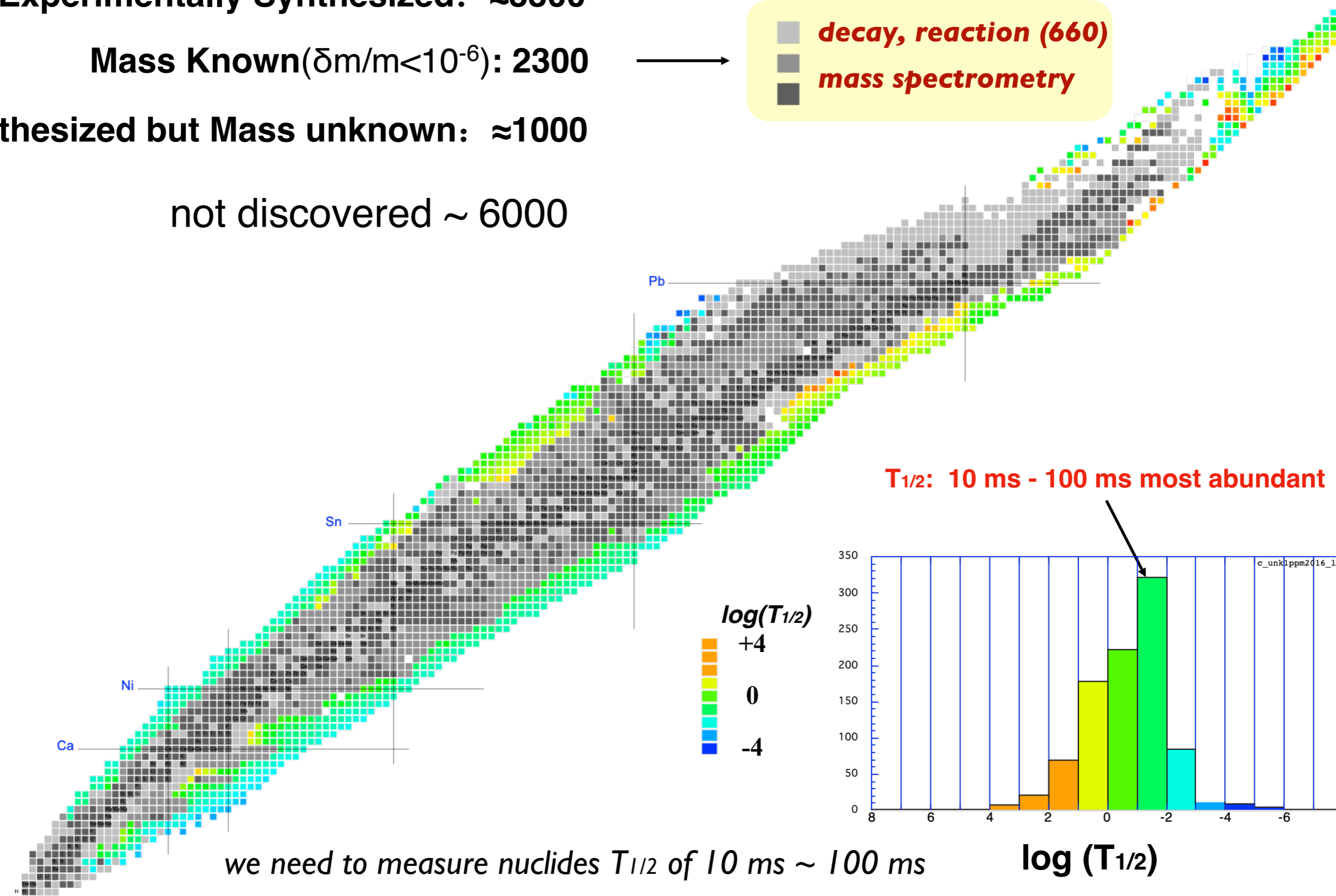
Mass Known ($\delta m/m < 10^{-6}$): 2300

Synthesized but Mass unknown: ≈ 1000

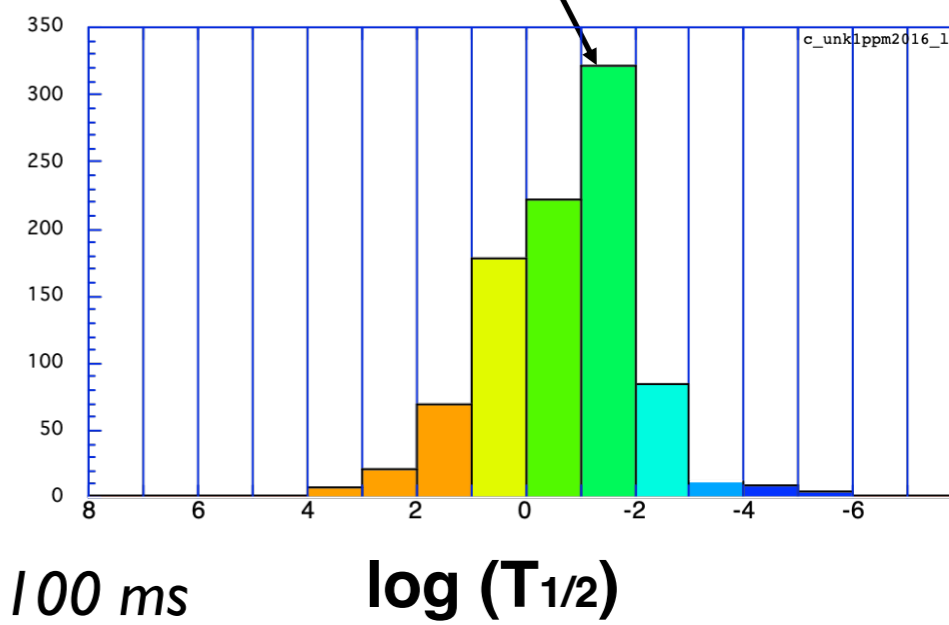
not discovered ~ 6000

note: many known masses were measured indirectly

decay, reaction (660)
 mass spectrometry



$T_{1/2}$: 10 ms - 100 ms most abundant

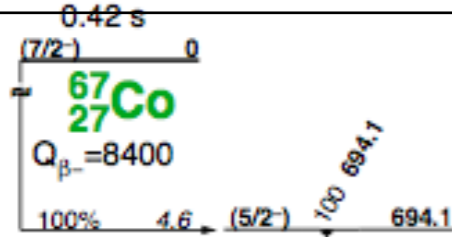


we need to measure nuclides $T_{1/2}$ of 10 ms \sim 100 ms

log (T_{1/2})

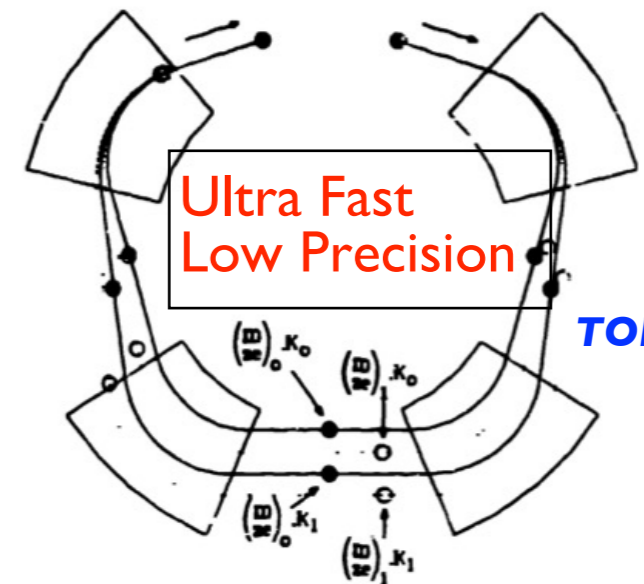
Mass Measurements of Short-lived Nuclei

Q-value (decay or reaction)



indirect : direct

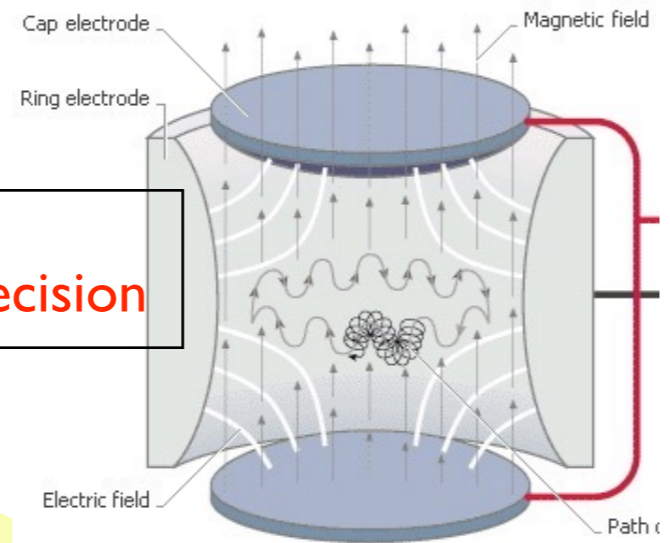
In-flight spectrometer



TOFI, SPEG ..

Universal Ambiguity from levels

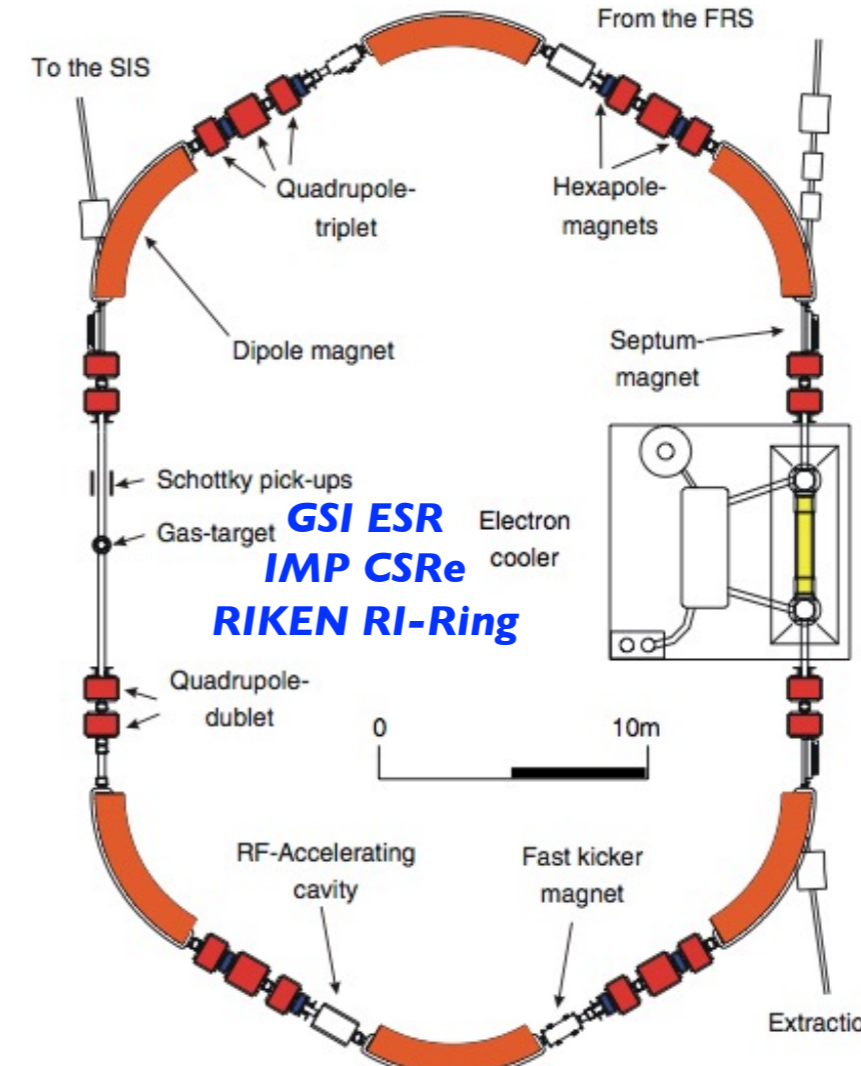
Penning Trap



Slow Ultra precision

ISOLDE, JYFL...

Storage Ring



Electron Cooling

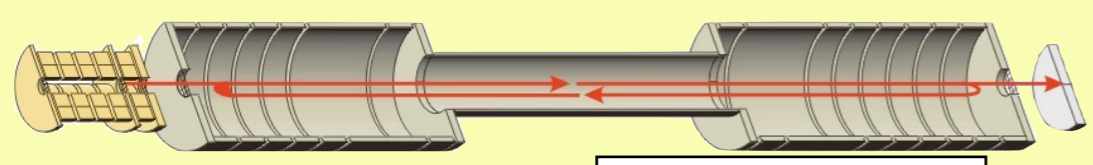
Very Slow High Precision

Isochronous

Fast Low Precision

New method

MRTOF (multi-reflection TOF)

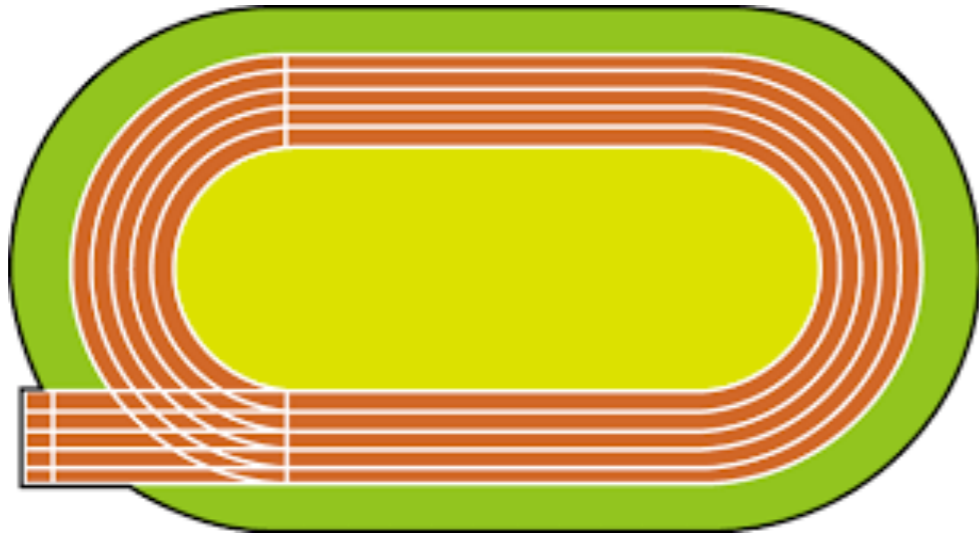


Fast High Precision

RIKEN-KEK, Giessen, ISOLDE ..

Time of Flight Mass Measurement

徒競走 ($\text{Time} \propto \sqrt{\text{Mass}}$)
if kinetic energy is constant



Use of a Track extends the distance



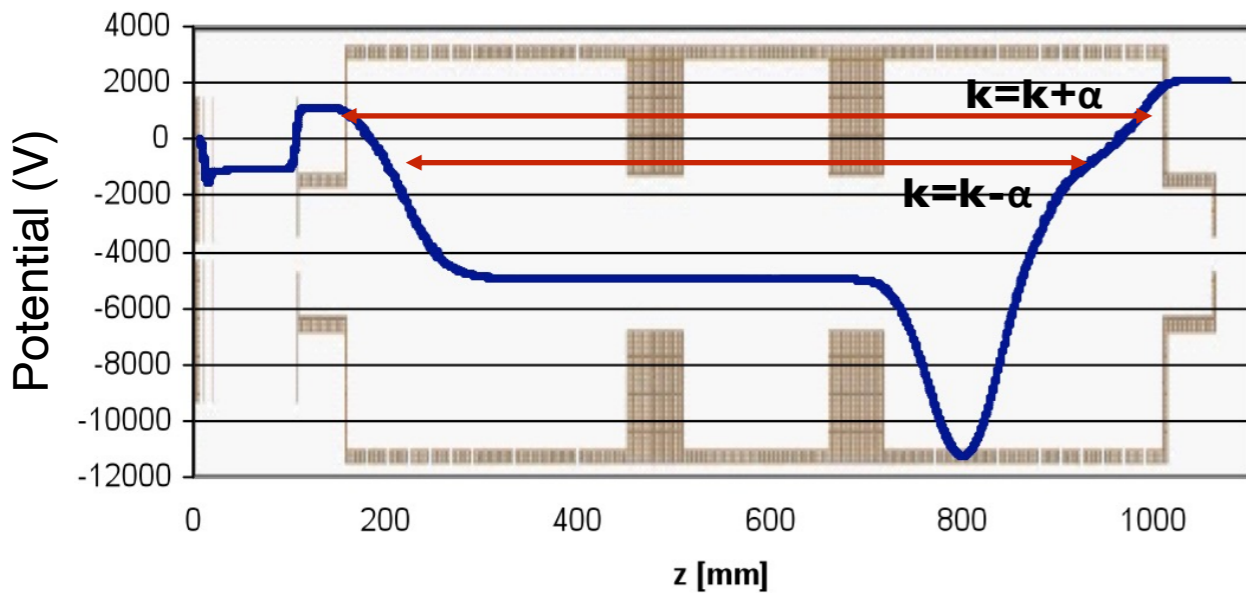
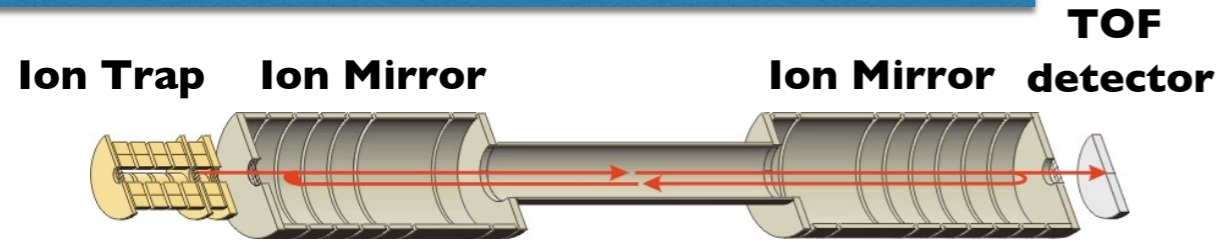
Not much difference in 100m race



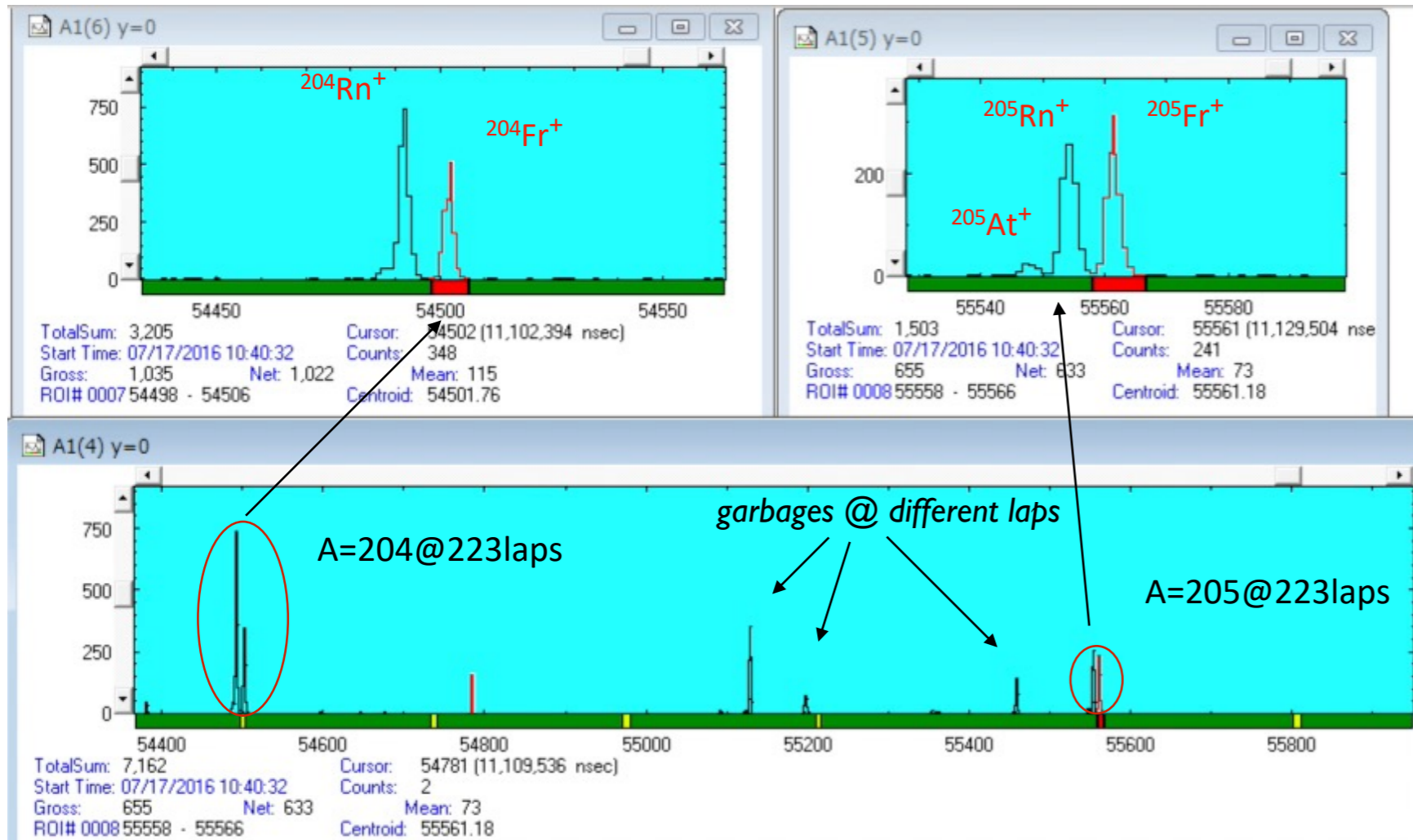
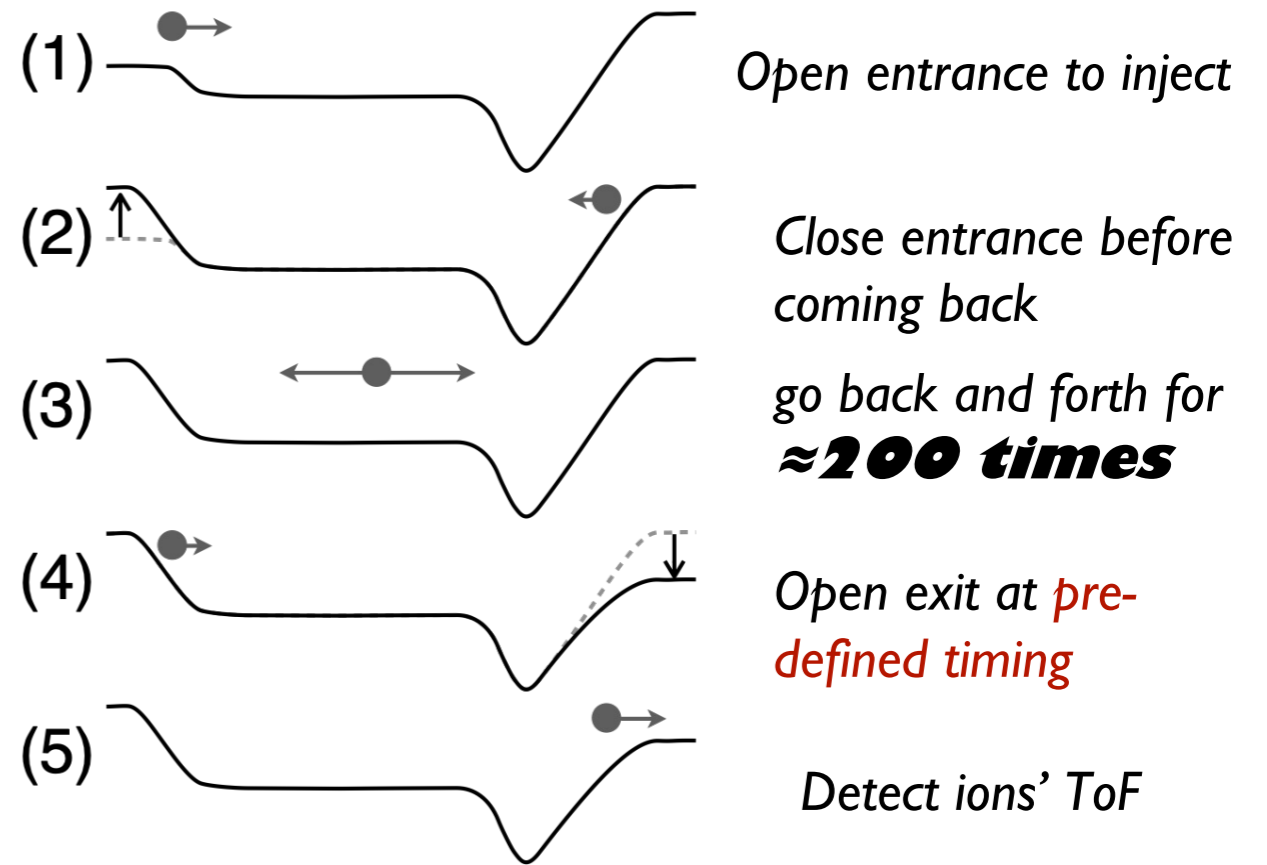
go back and forth in a swimming pool

For atoms (ions), go
back and forth between
a pair of (electric)
mirrors.

MRTOF Mass Spectrograph



(Multi Reflection Time of Flight...)



$$m_x = \left(\frac{ToF_x}{ToF_r} \right)^2 m_r$$

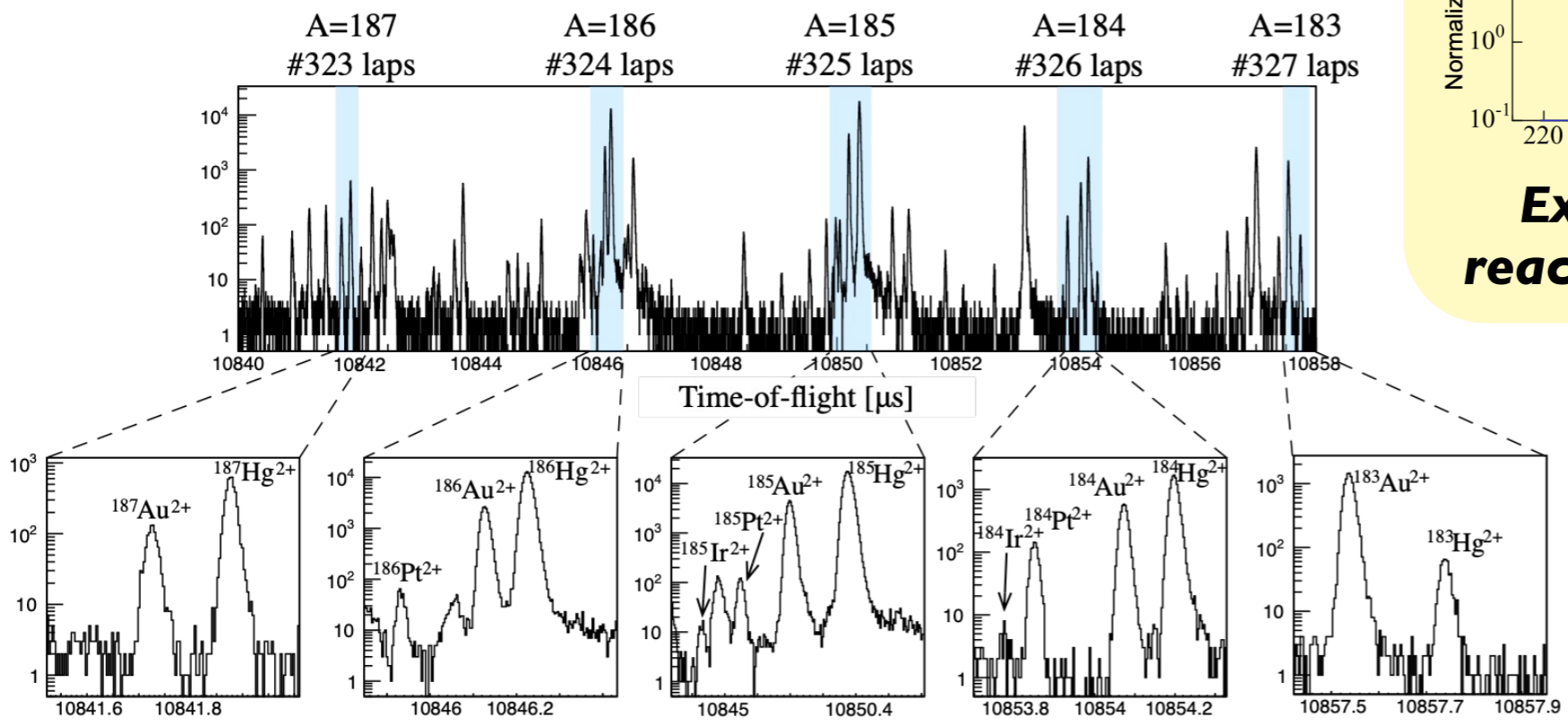
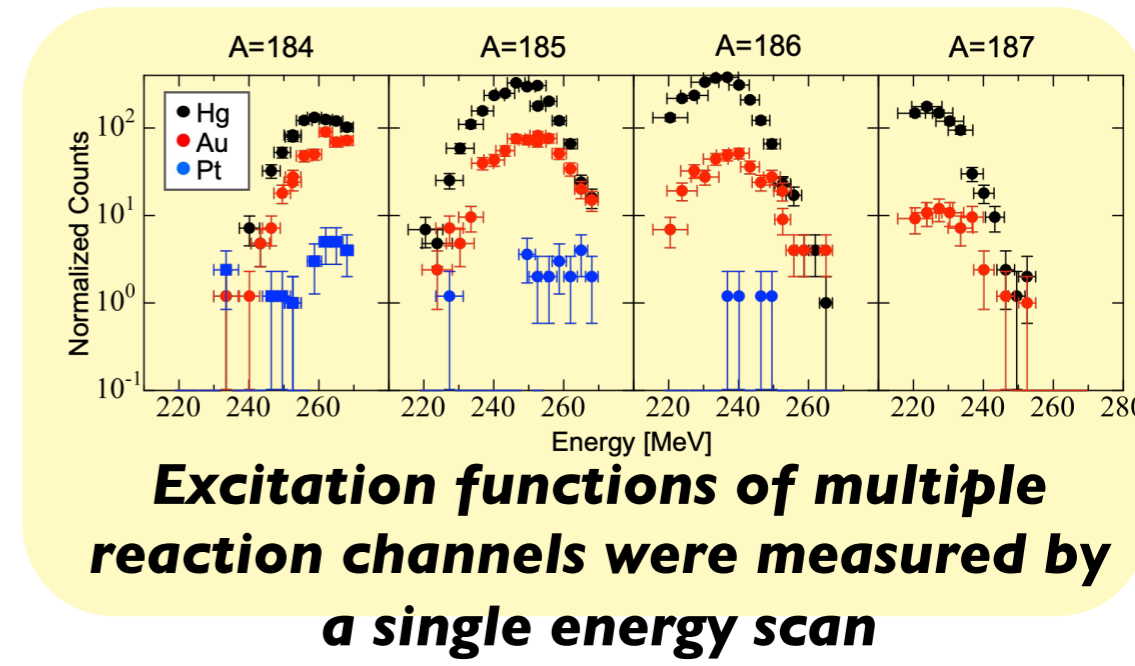
- fast (~10 ms)
- high precision (MRP~1M)
- high accuracy
- high efficacy

● **Example of high efficient measurement**



<i>xn</i>	^{183}Hg	^{184}Hg	^{185}Hg	^{186}Hg	^{187}Hg	^{188}Hg	^{189}Hg	^{190}Hg
<i>pxn</i>	^{182}Au	^{183}Au	^{184}Au	^{185}Au	^{186}Au	^{187}Au	^{188}Au	^{189}Au
<i>αxn</i>	^{181}Pt	^{182}Pt	^{183}Pt	^{184}Pt	^{185}Pt	^{186}Pt	^{187}Pt	^{188}Pt
<i>αpxn</i>	^{180}Ir	^{181}Ir	^{182}Ir	^{183}Ir	^{184}Ir	^{185}Ir	^{186}Ir	^{187}Ir

Products of multiple reaction channels were measured in a single ToF spectrum



Why we use MRTOF?

- High efficacy

Multiple species at once
(no scan, no pre-purification)

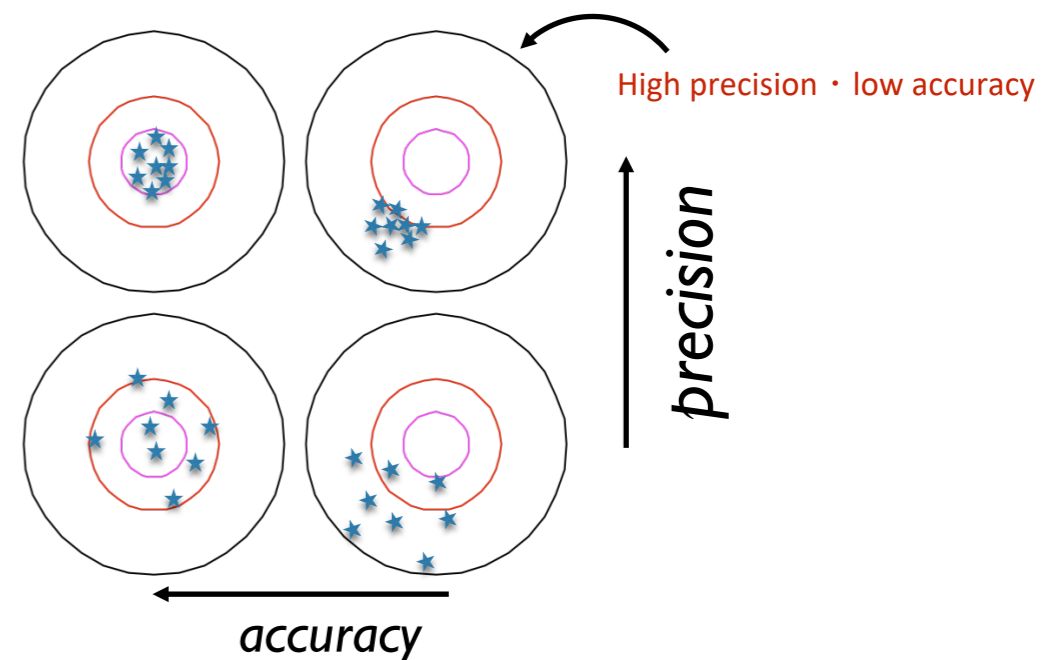
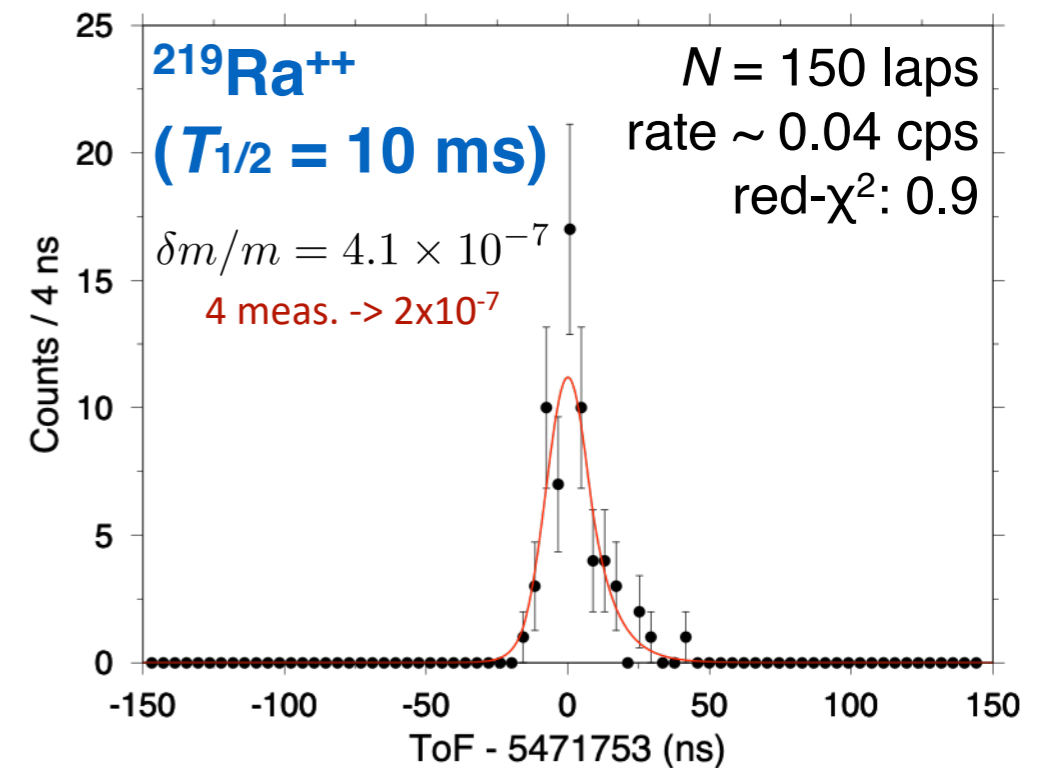
- *Short-lived, Heavy nuclides*

$T_{1/2} = 10$ ms, $A > 200$ no loss in precision

- High precision, High accuracy

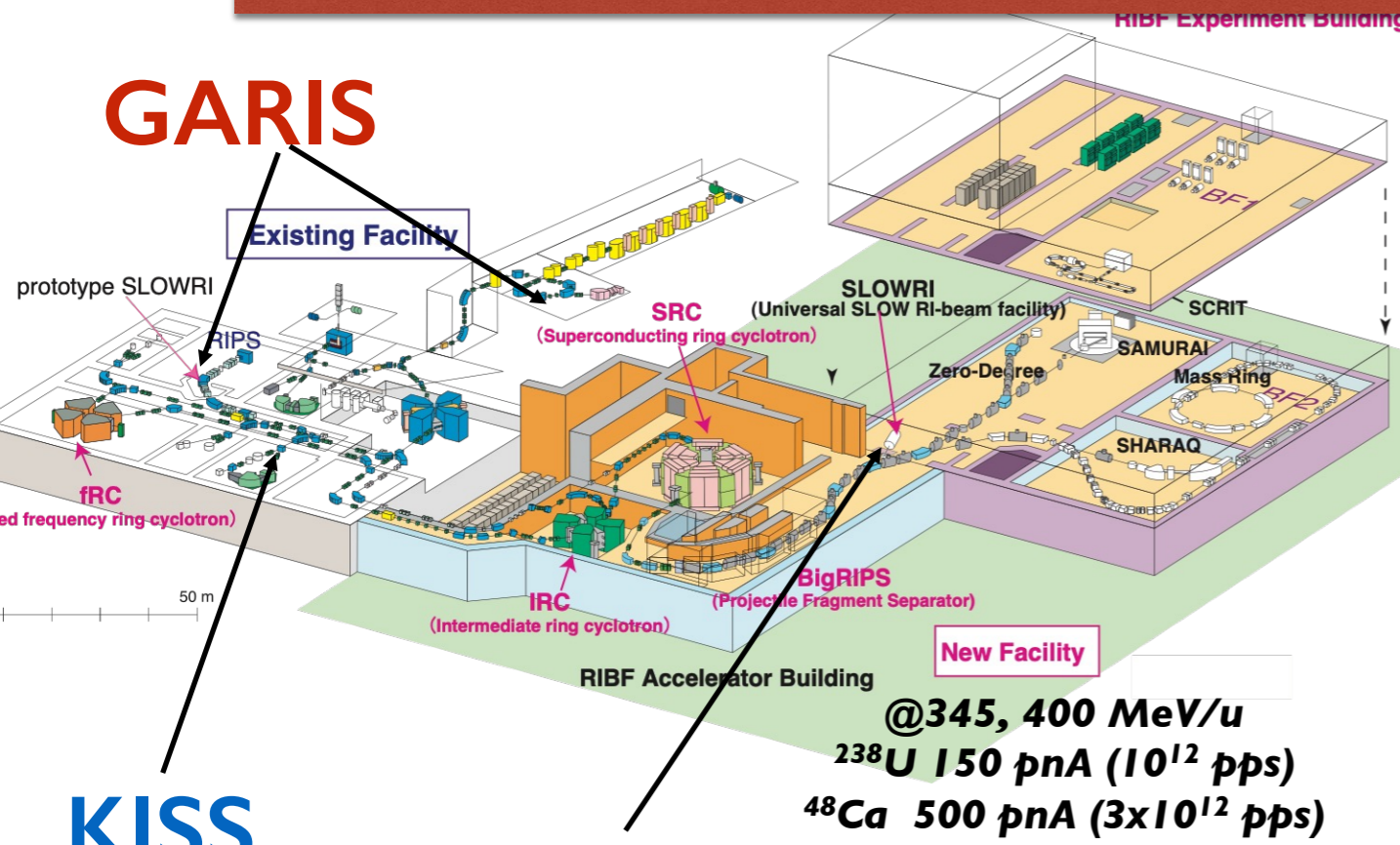
Mass resolving power ($R_m = 1,000,000$)

Excellent referencing methods



Q1: What is the best?

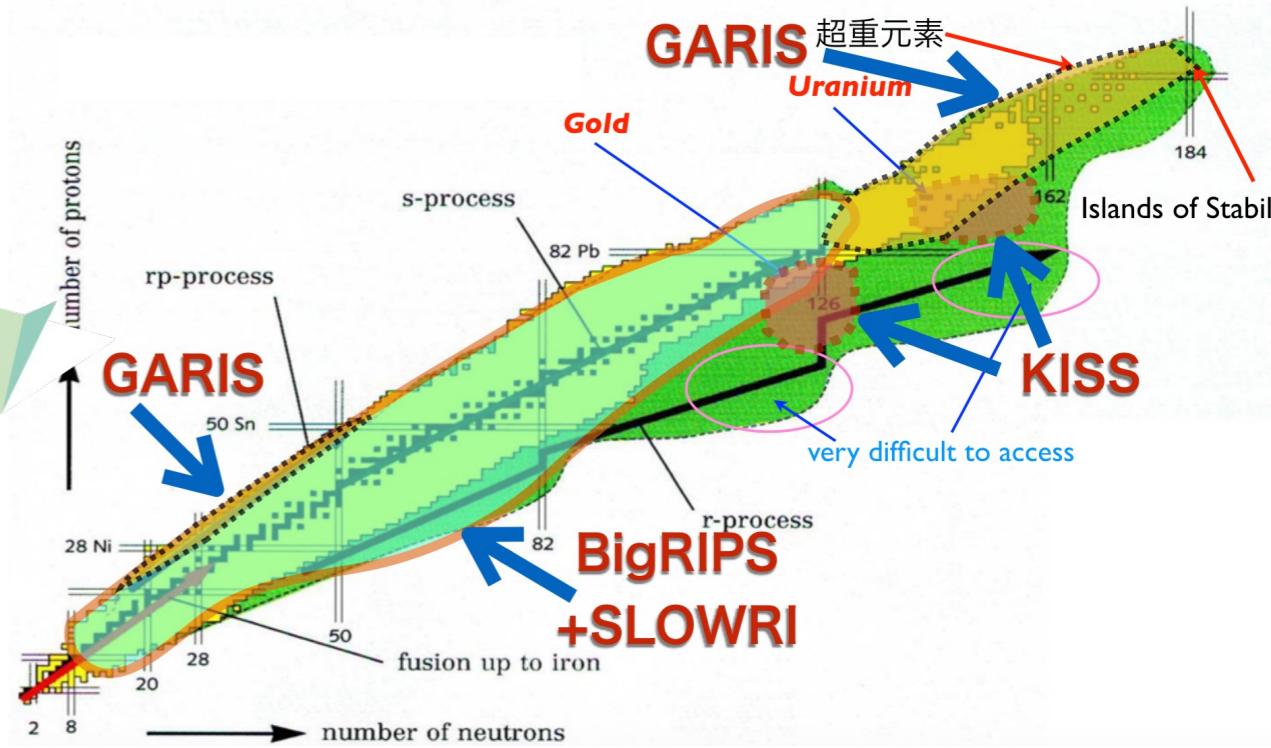
Comprehensive Mass measurements at RIBF with MRTOF



GARIS

KISS

**BigRIPS
+SLOWRI**



GARIS: SHE, fusion products

BigRIPS: fragmentation/fission lower than U

KISS: n-rich nuclides via MNT

RIBF provides world highest number of nuclides
from Hydrogen to Nihonium

slogan:

Measure masses of all available possible nuclides

Other Mass spectrometers at RIKEN RIBF (BigRIPS)

Cover article of RIKEN Accel. Prog. Rep. 56 (2023)

B ρ -ToF at SHARAQ

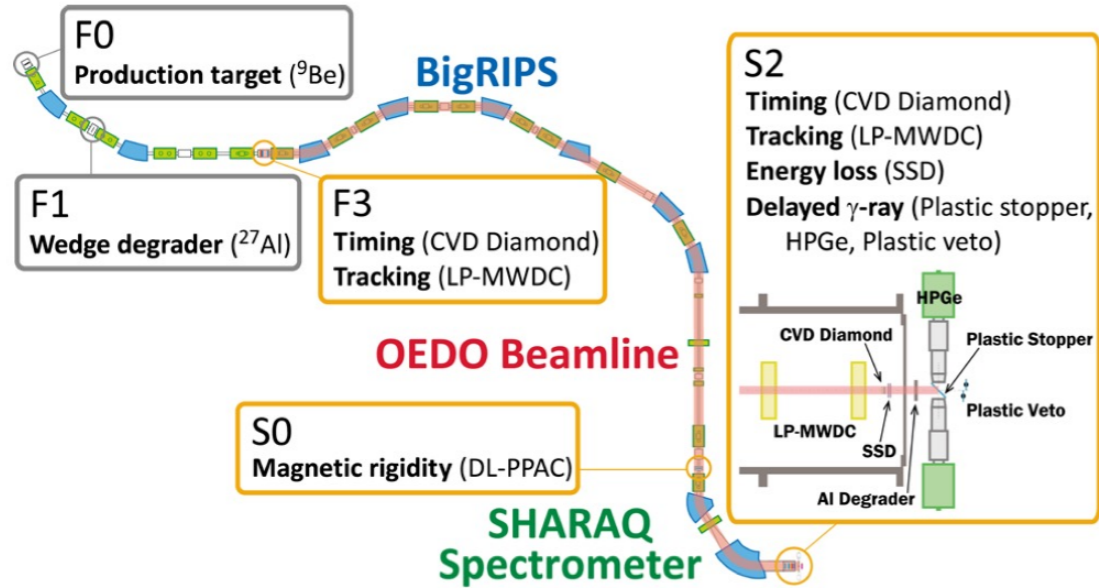


Fig. 2. Experimental detector setup in the OEDO-SHARAQ system for the direct mass measurements.

Isochronous storage ring: R3

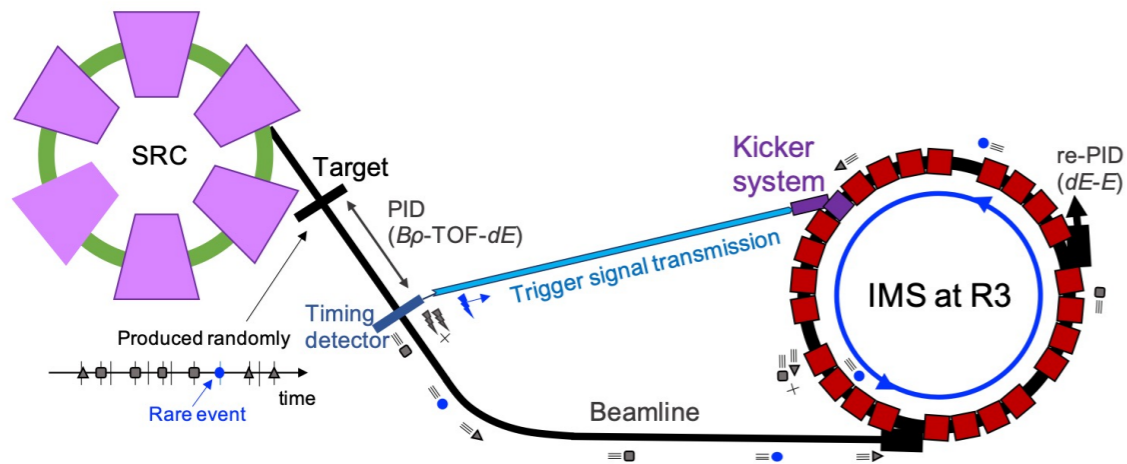


Fig. 3. Conceptual design and method for measuring the mass of short-lived rare nuclei using R3.

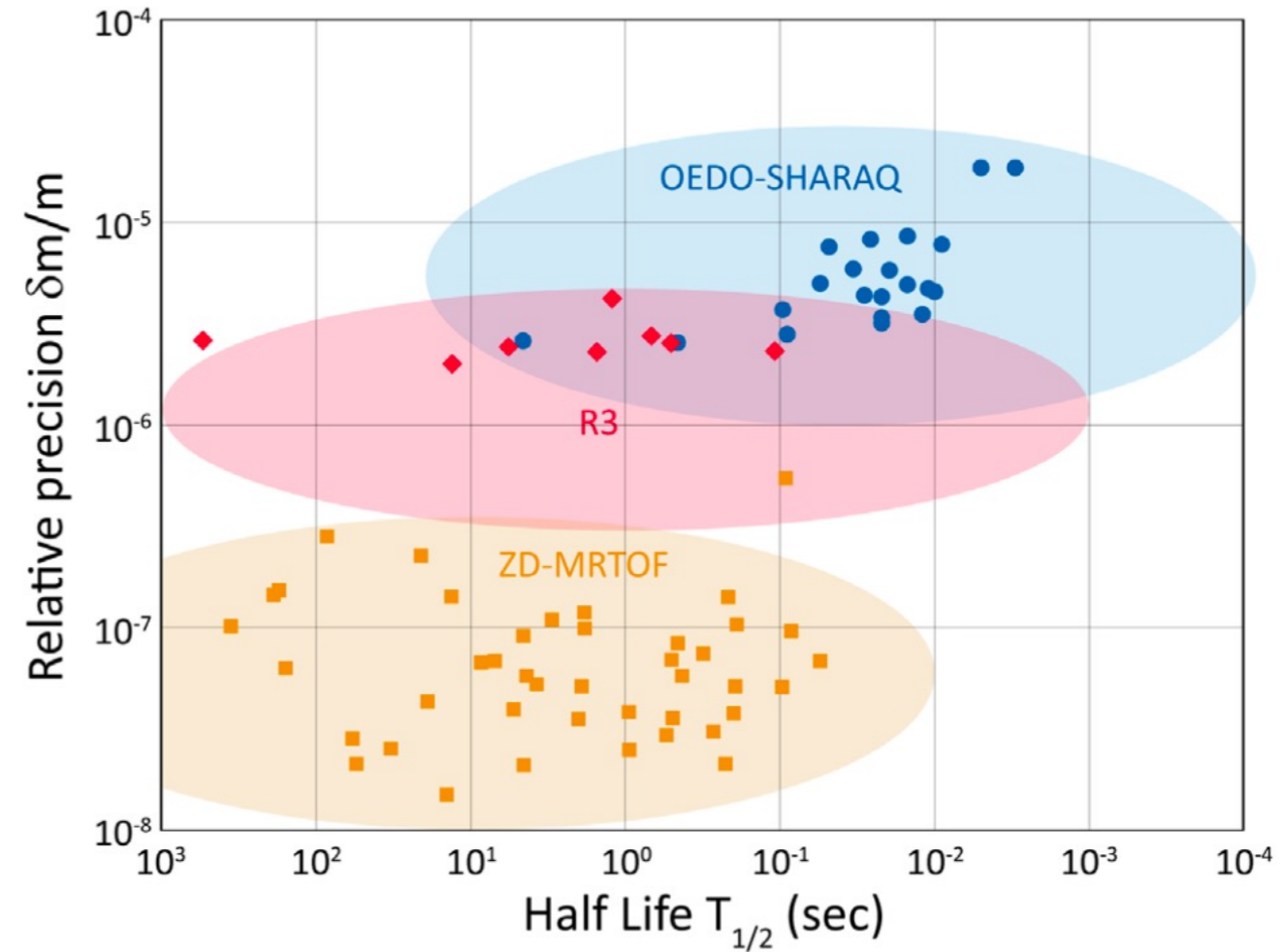
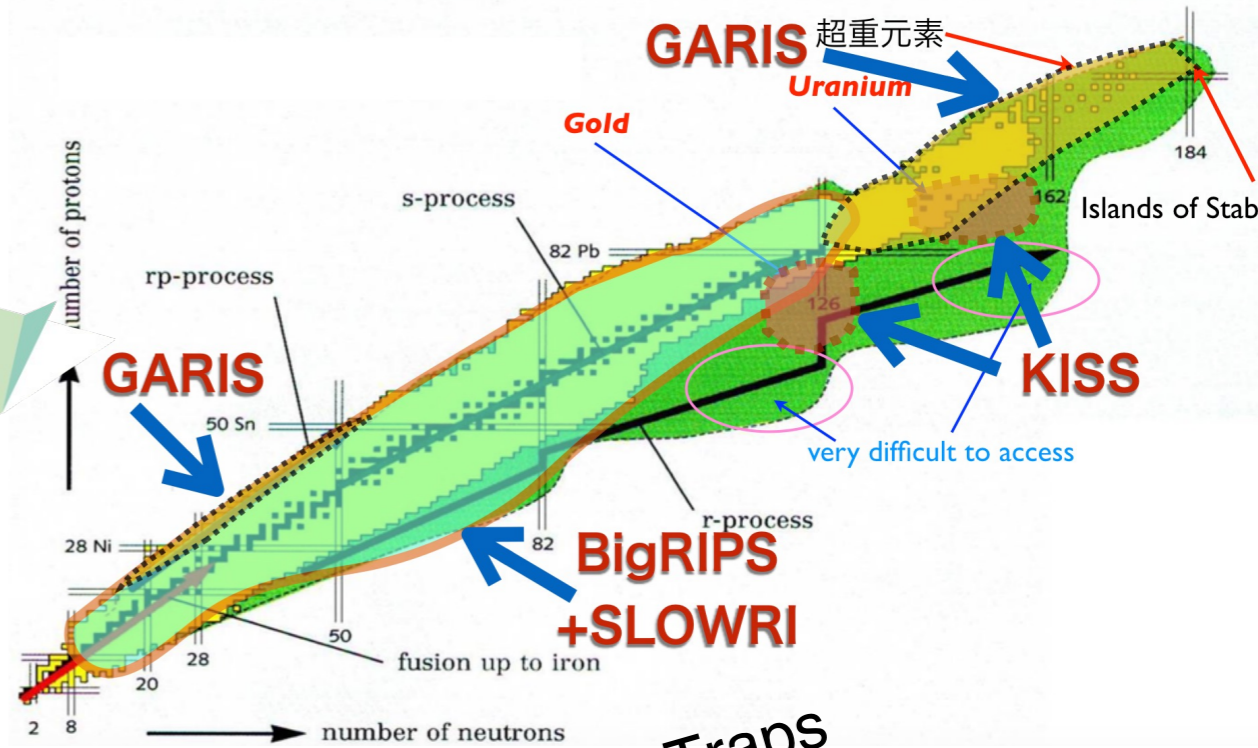
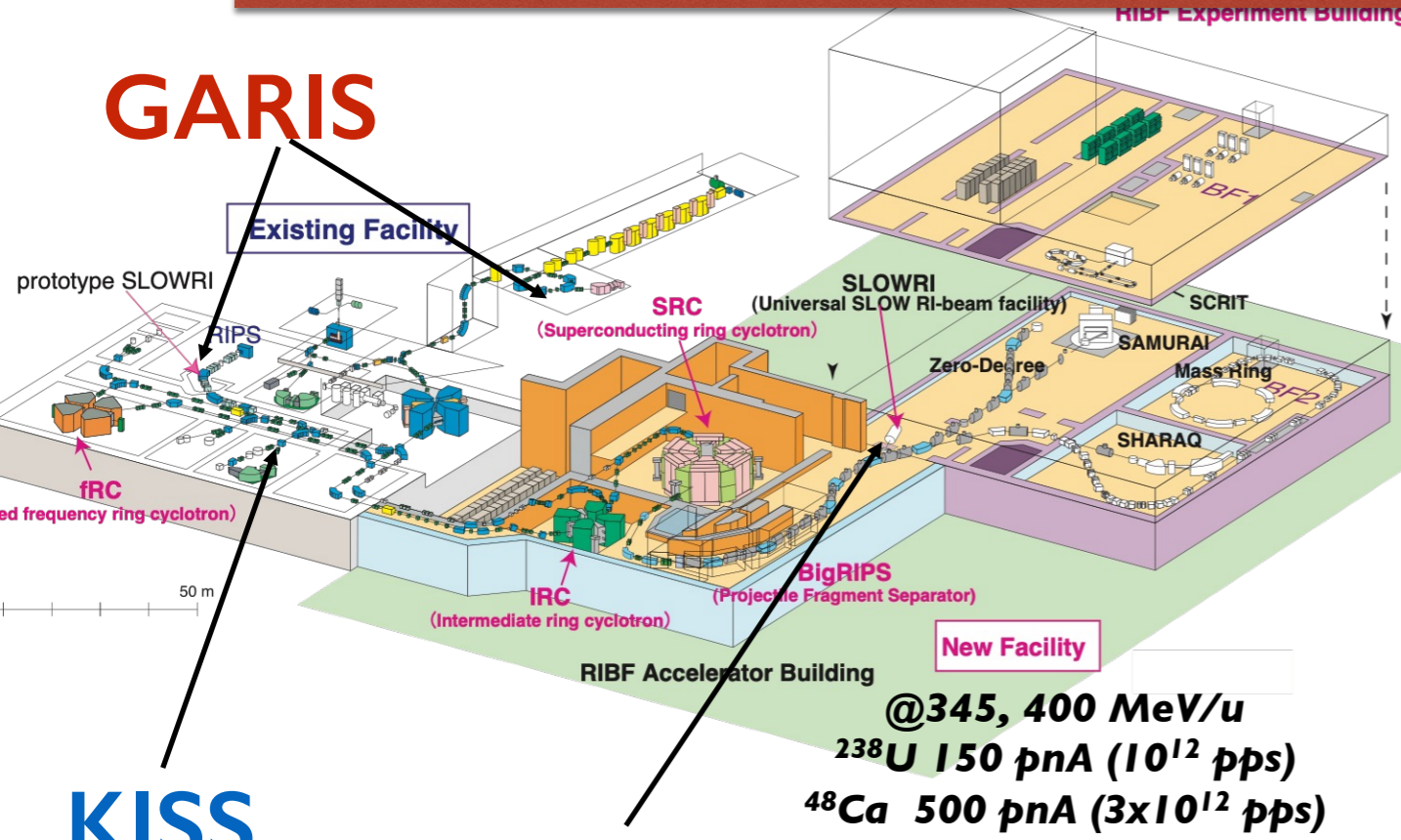


Fig. 1. Mass precision and nuclear half-life regions covered by the three mass measurement methods.

Comprehensive Mass measurements at RIBF with MRTOF

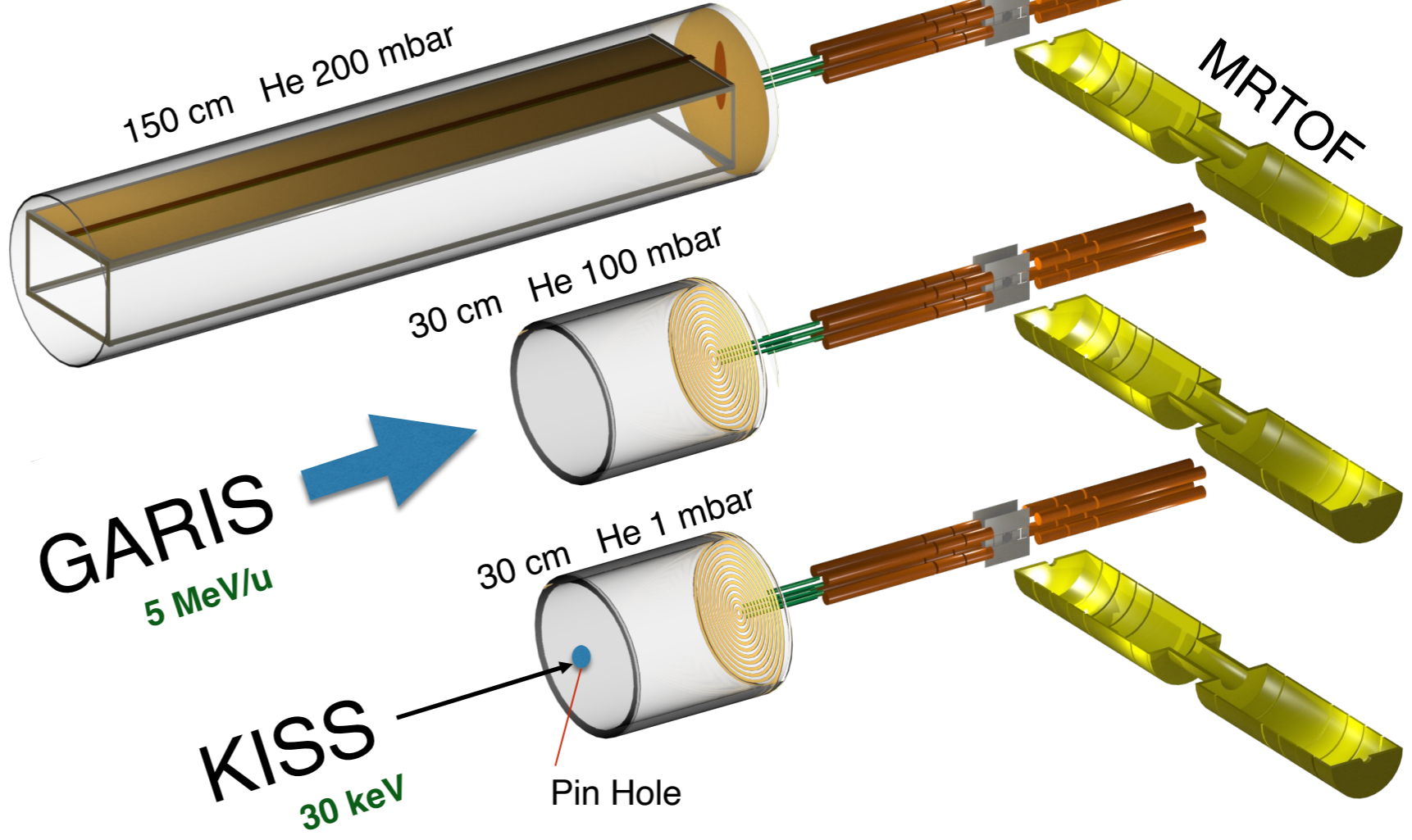


KISS

**BigRIPS
+SLOWRI**

@345, 400 MeV/u
 ^{238}U 150 pA (10^{12} pps)
 ^{48}Ca 500 pA (3×10^{12} pps)

BigRIPS
200 MeV/u



GARIS
5 MeV/u

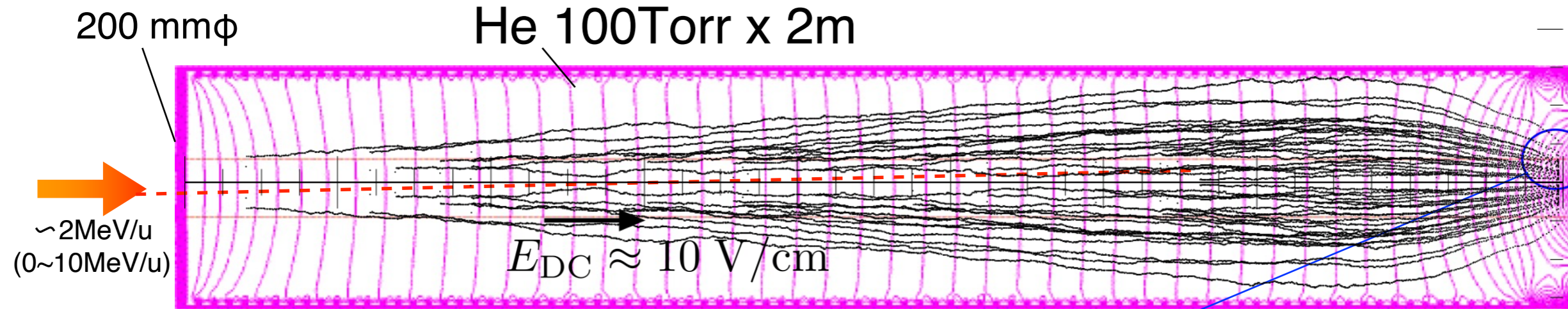
KISS
30 keV

1. Thermalize in He gas
2. Extraction by RF-carpet
3. Trap in Ion-Traps
4. Mass measurements with MRTOF

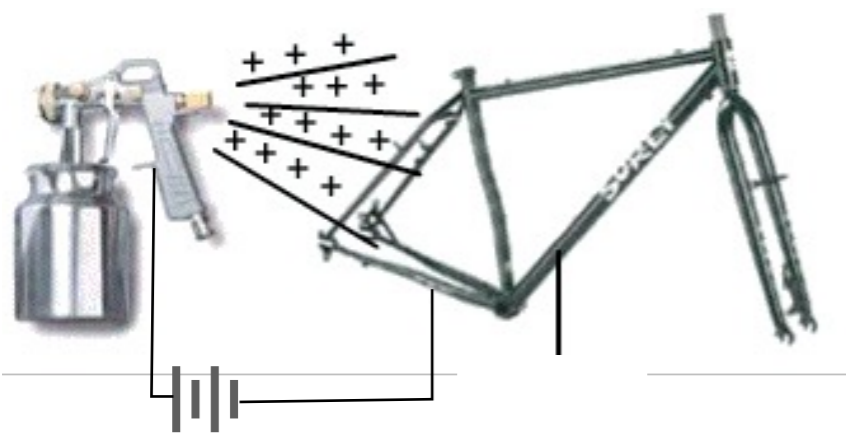
Big Gas Cell -- traditional type

- Energetic ions can be stopped in He gas as ions, extracted by DC+ inhomogeneous RF fields
- fast, efficient, and universal conversions energetic RI to trapped RI ions in ion traps

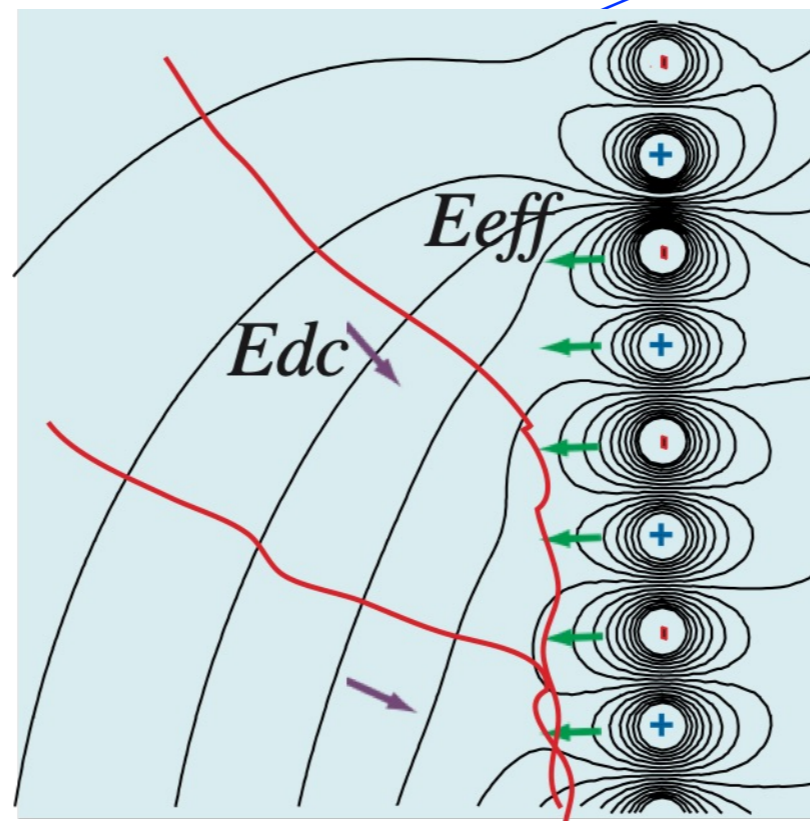
nozzle 0.7mmΦ



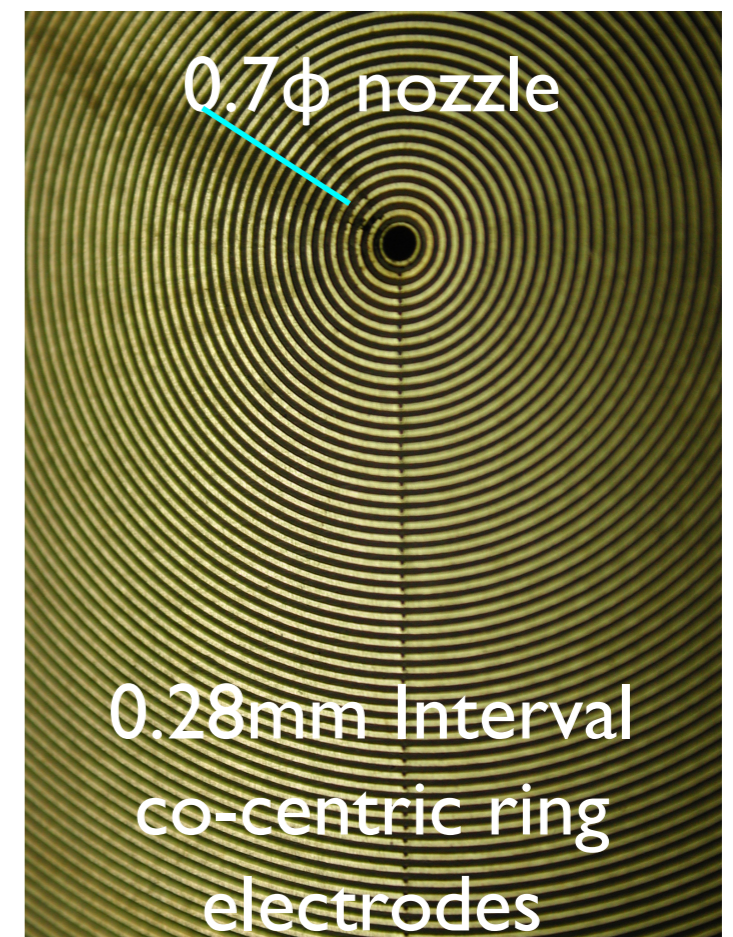
How to paint skeleton?



Electric force lines always terminated at cathode, even if it is a mesh !



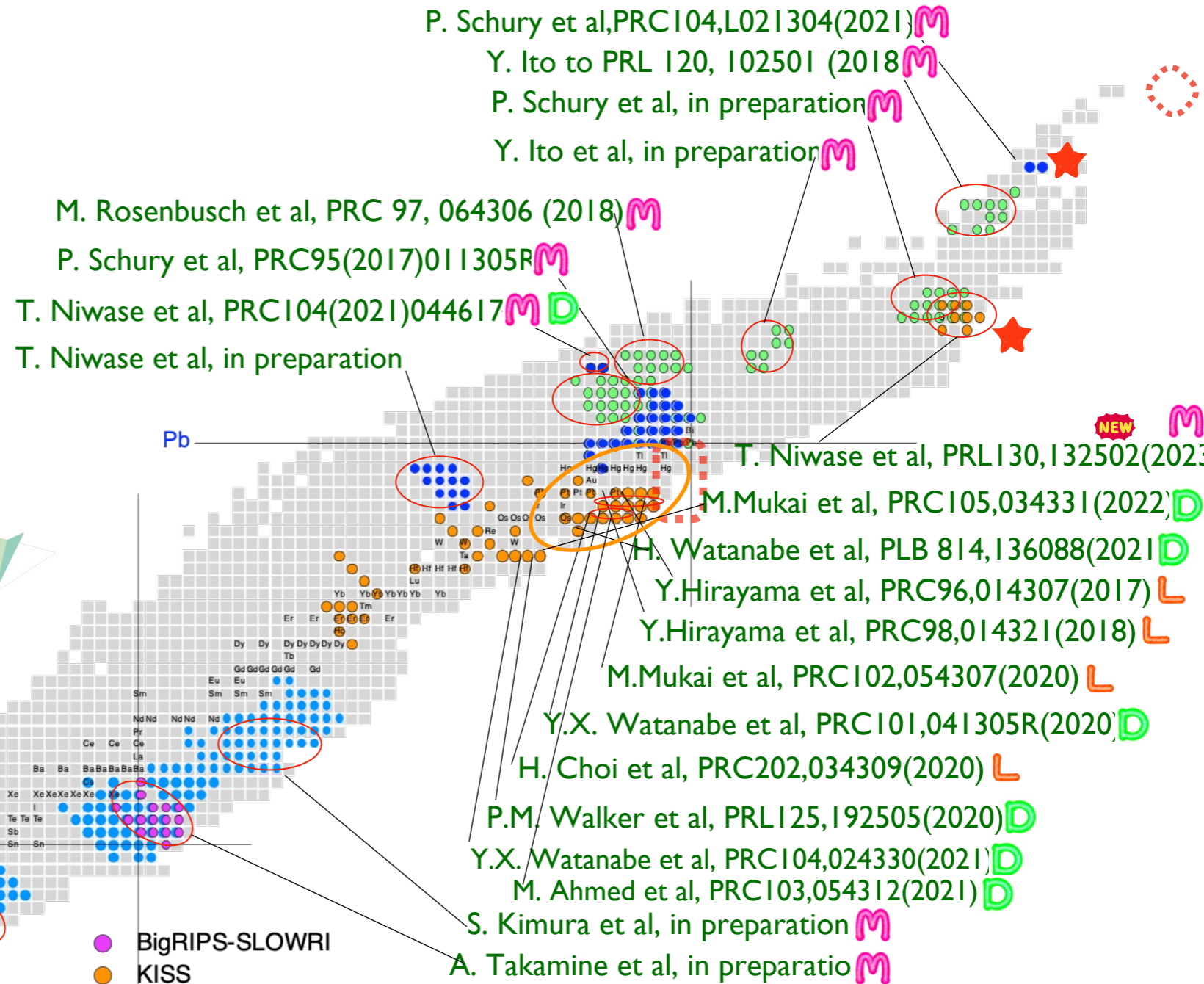
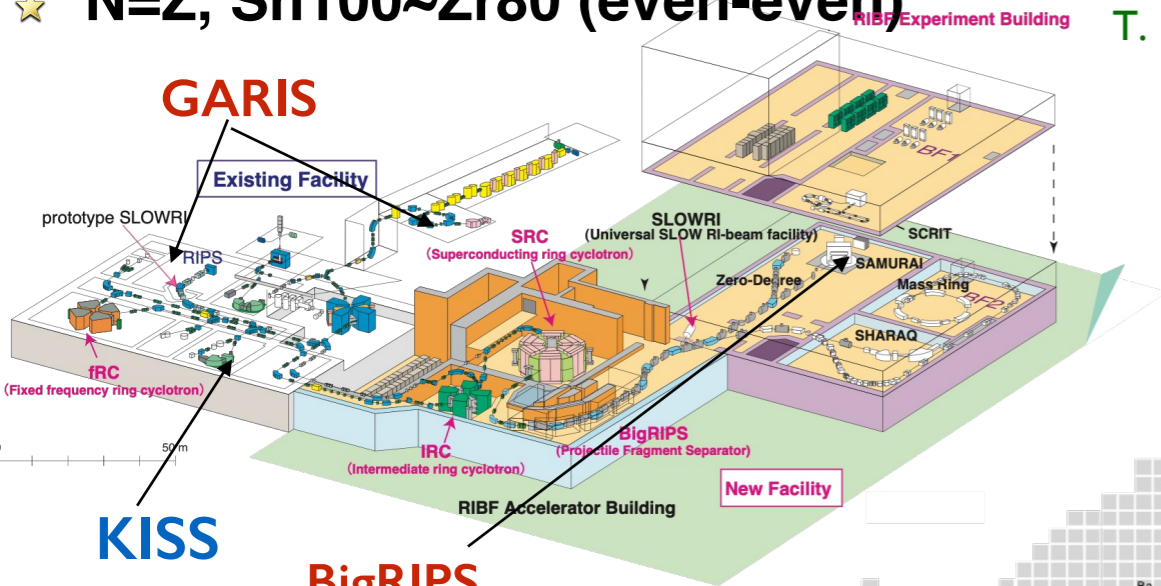
Ion Barrier by RF gradient fields



KEK Wako Nuclear Science Center

~recent highlights~

- ★ Discovery of new n-rich uranium isotope @KISS
- ★ First direct mass measurements of Superheavy element, Db @GARIS
- ★ Disappearance of N=34 magic number for Ti, Sc @BigRIPS-SLOWRI
- ★ N=Z, Sn100~Zr80 (even-even)



Summer 2024, in preparation

NEW

- BigRIPS-SLOWRI
- KISS
- GARIS-II
- AME2020 ($\delta m/m < 1 \text{ ppm}$)

- S. Kimura et al, Nature, in preparation
- S. Kimura et al, PRC submitted M NEW
- D. Hou et al, to PRC108,054312(2023) M NEW
- W. Xian et al, PRC109,035804(2023) M NEW
- M. Rosenbusch et al, in preparation M
- S. Kimura et al, IJMS 430, 134-142 (2018) M
- S. Kimura et al, PRL 130, 012501(2023) M NEW

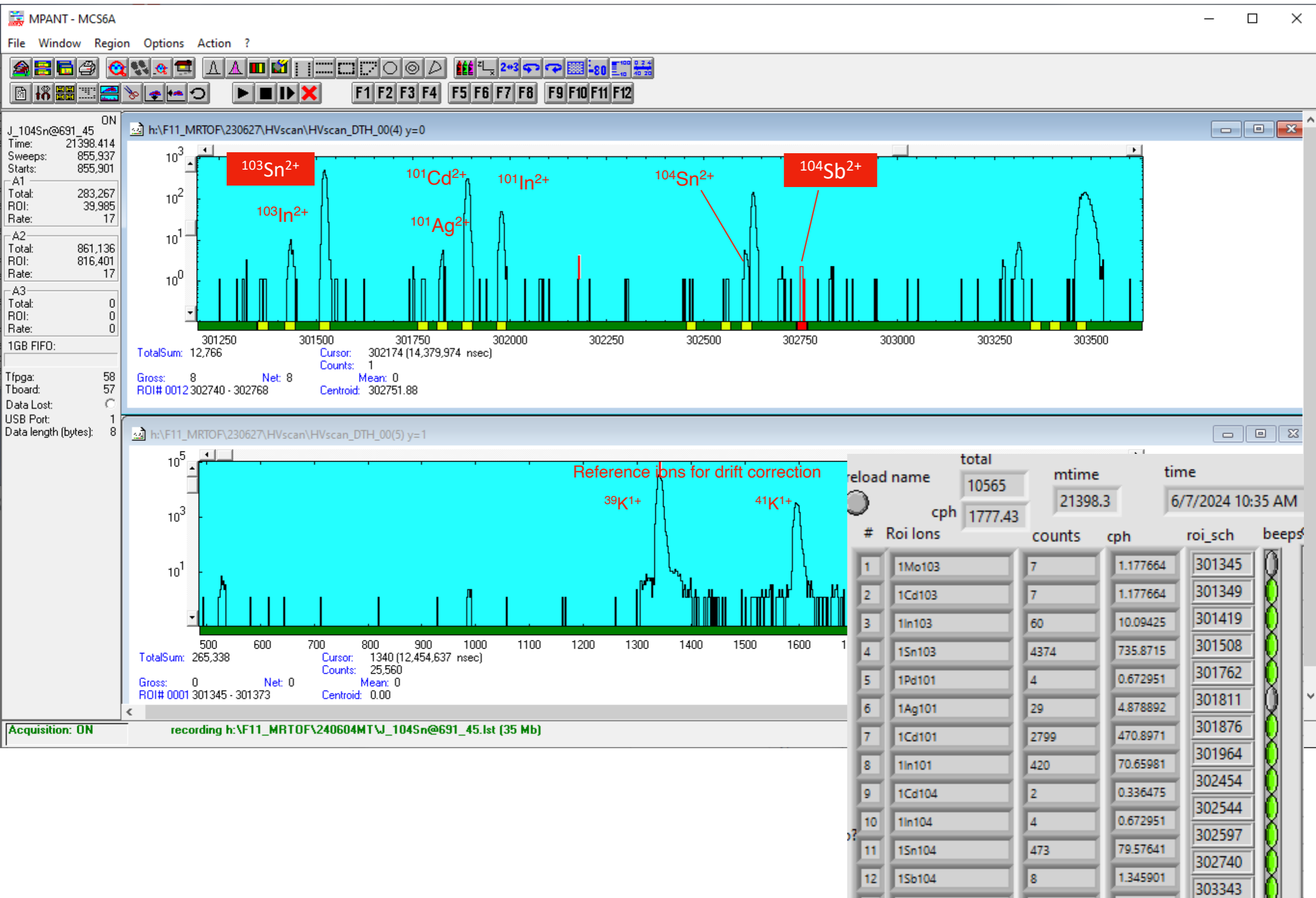
D Decay spectroscopy

L Laser spectroscopy

M Mass spectrometry

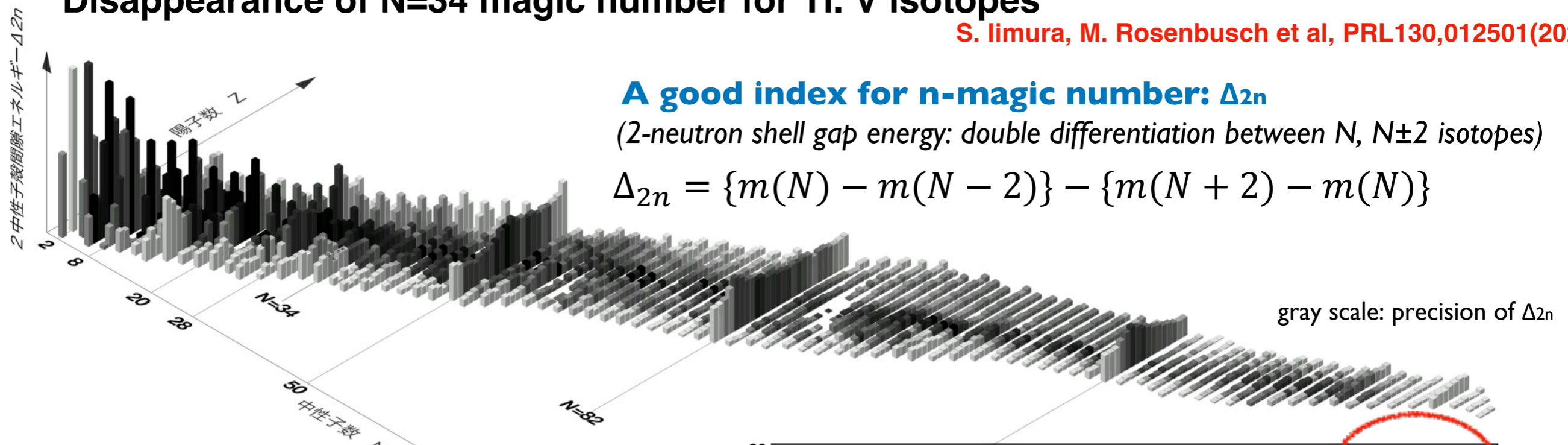
Nuclides studied at WNSC 2017~

RIBF211, MRTOF part (7 June, day 4) Sb104, Sn103,104



Disappearance of N=34 magic number for Ti, V isotopes

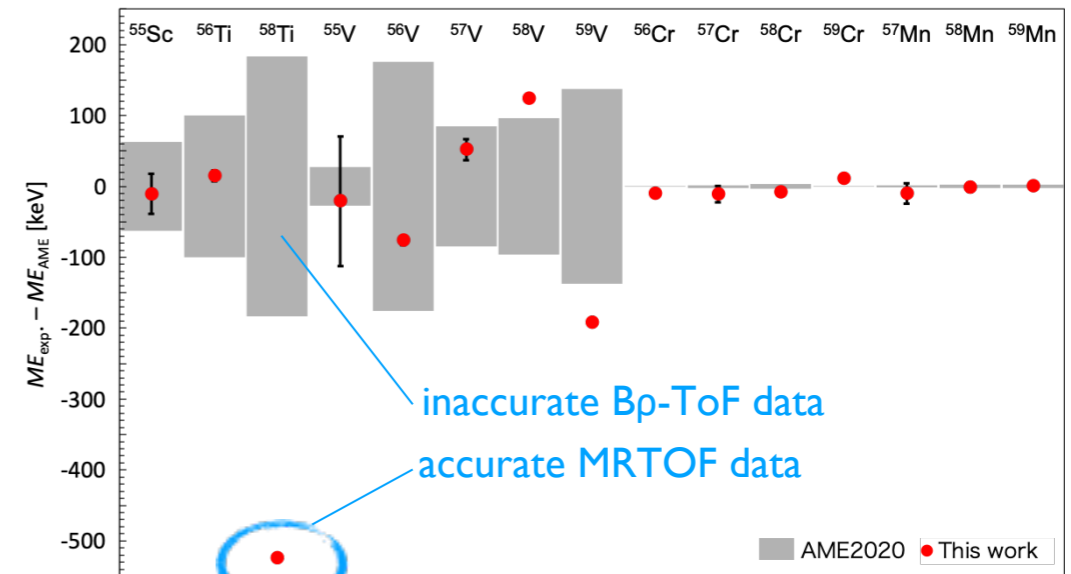
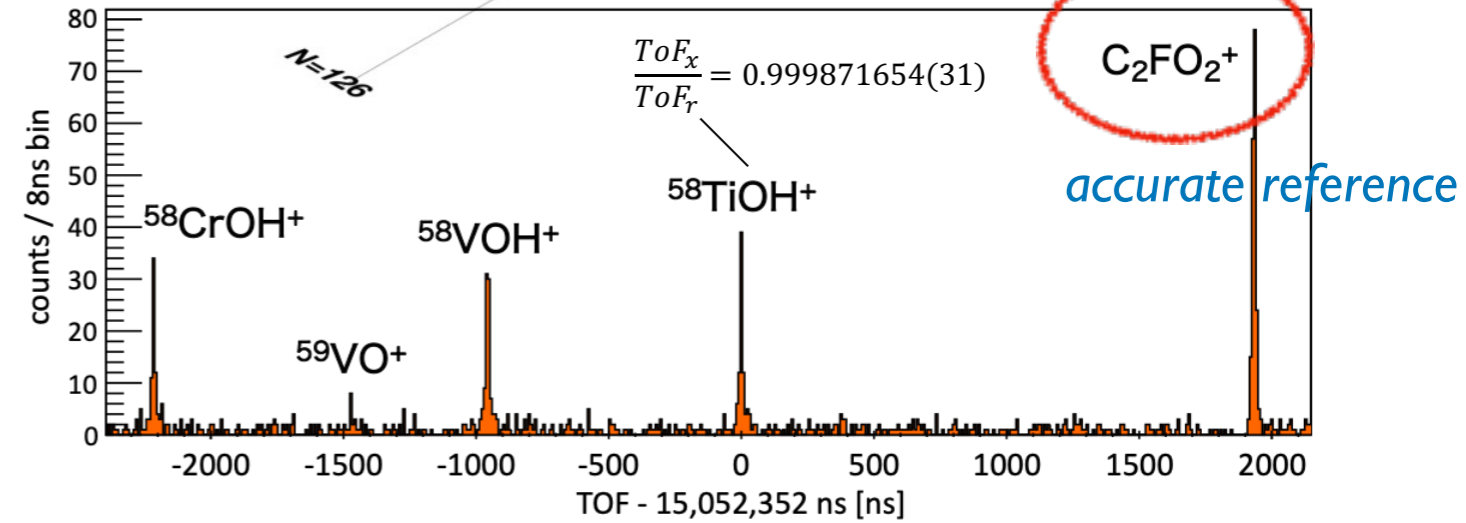
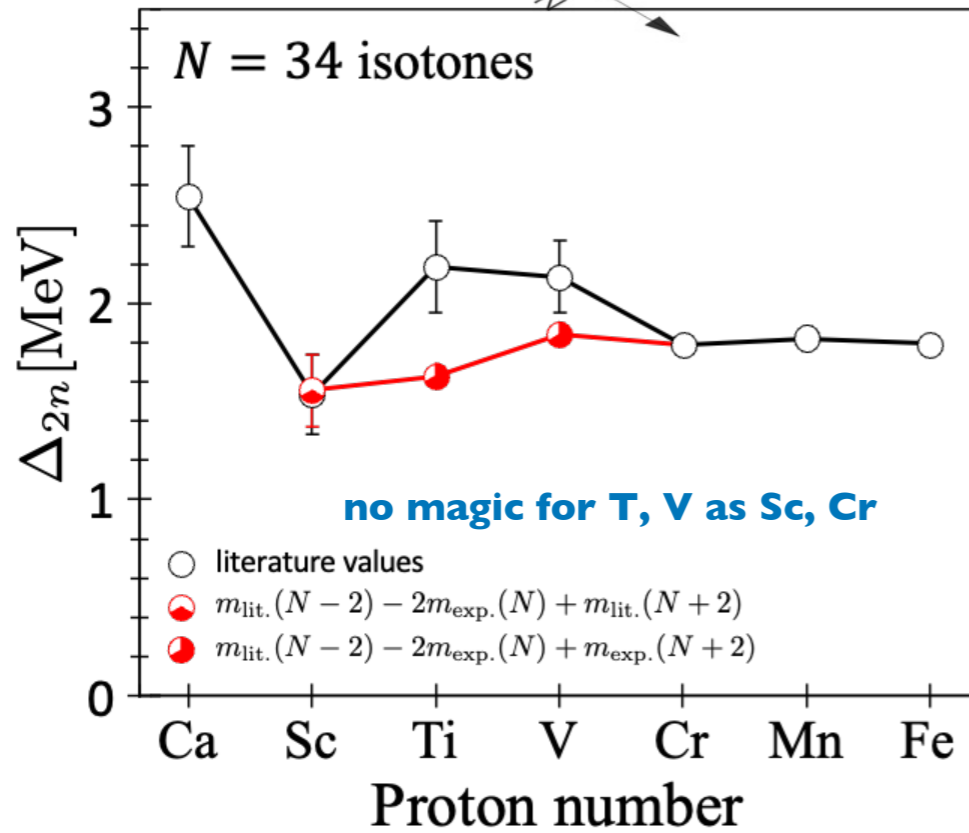
S. Iimura, M. Rosenbusch et al, PRL130,012501(2023)



A good index for n-magic number: Δ_{2n}

(2-neutron shell gap energy: double differentiation between $N, N\pm 2$ isotopes)

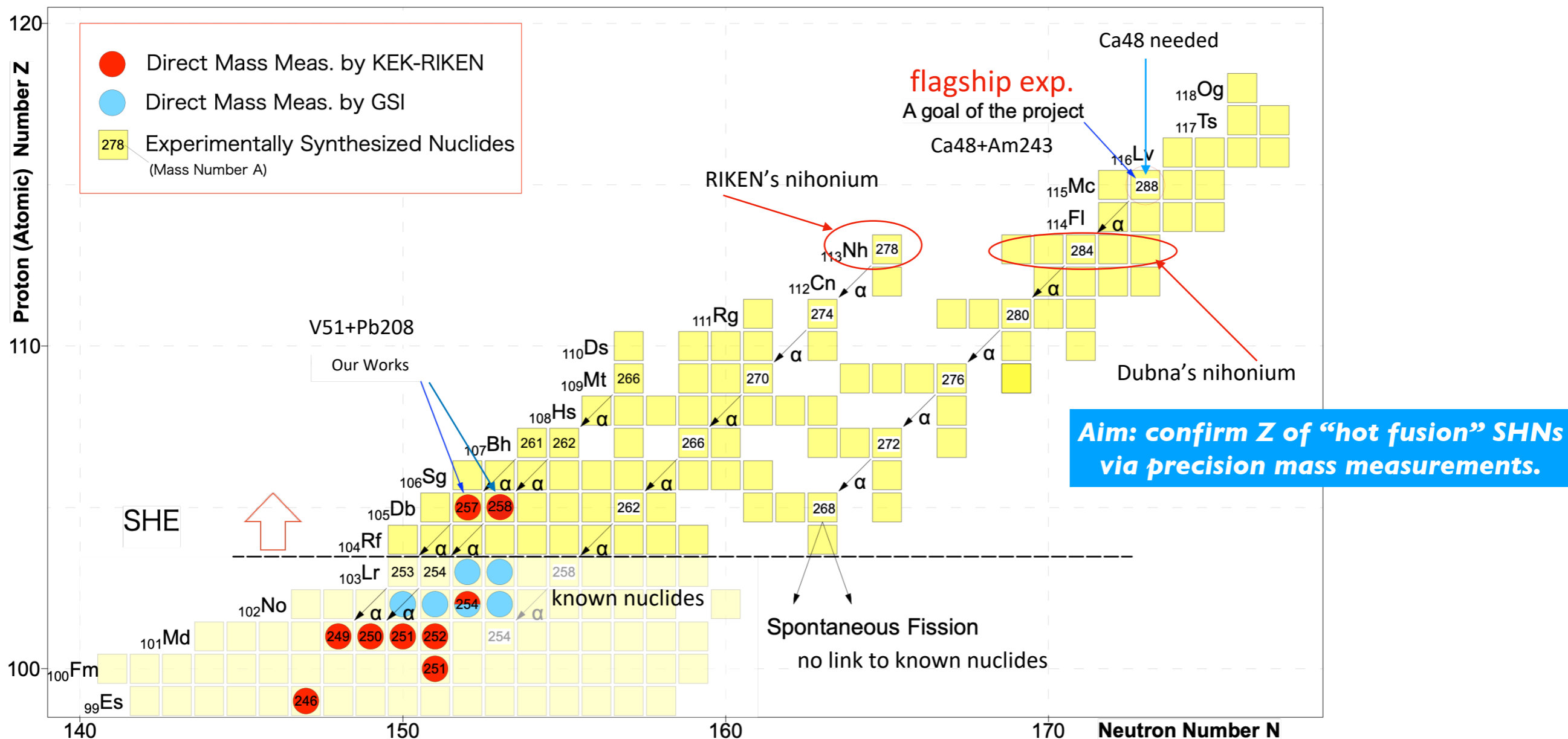
$$\Delta_{2n} = \{m(N) - m(N - 2)\} - \{m(N + 2) - m(N)\}$$



Comparison of this work with AME2020

A previous study showed that a new magic number of N=34, found in Ca isotopes, was present in Ti and V isotopes, even though it was absent in Sc. The high precision and accuracy of the mass measurement in this study proved that there is no magic number nature in Ti and V isotopes.

Mass Measurement of Superheavy elements @ GARIS-II MRTOF



- **Proposed even before Nh granted.**
- **PAC provided "S"-grade with 40 days MT**
- **Setup has been ready for exp., however, difficult to obtain Ca48.**

Dev. of a self-purification system with CANDLE project (ββ)

IUPAC Technical Report

Paul J. Karol^{a,*}, Robert C. Barber, Bradley M. Sherrill, Emanuele Vardaci
and Toshimitsu Yamazaki

Discovery of the elements with atomic numbers $Z = 113, 115$ and 117 (IUPAC Technical Report)

150 — P. J. Karol *et al.*: Discovery of the elements with atomic numbers $Z = 113, 115$ and 117

DE GRUYTER

(in time and position) alpha decays that have a vanishingly small probability to be random coincidences. When corresponding chains are observed in cross reactions of $(X,2n)$, $(X,3n)$ and $(X,4n)$ reactions and/or in the decays of heavier elements made at more than one laboratory, the assignments are made beyond a reasonable doubt.

The new elements identified in the claims considered here have distinct features from their assigned $Z = 114$ and $Z = 116$ neighbors [5]. The nature of the alpha energy spectra observed in the decays of nuclides with atomic numbers 113, 115, and 117 differ from their even- Z neighbors and show a wider energy spread corresponding to decay to excited states. This is further evidence that new atomic number has been produced in these studies and disfavor charged-particle emission in the evaporation process or electron capture in the decay chains. As a result a large group of super heavy nuclides are now on an island without connection to the main peninsula of known nuclei where reliable identification of Z, N becomes more and more difficult. Firmly connecting this island to the nuclear mainland should remain a priority. We encourage development of direct physical methods to determine Z . Particularly promising are the prospects for X-ray measurements and identification as was now attempted [22].

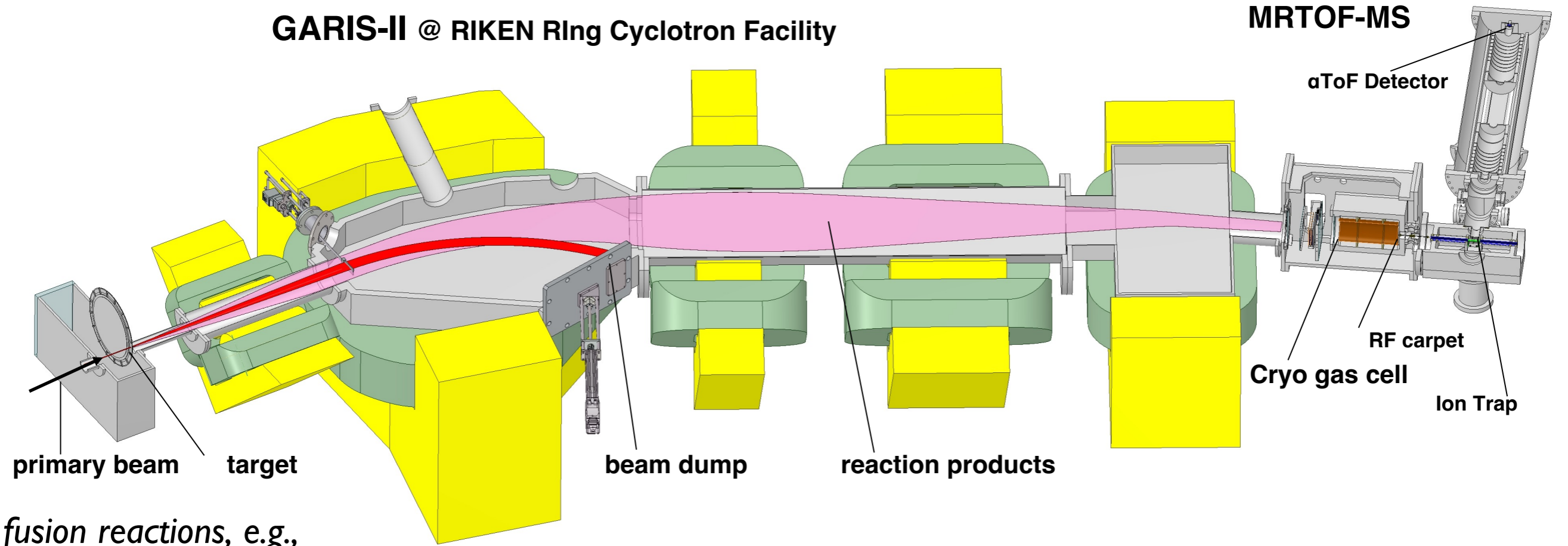
In light of the utility of applying the sum energy check for odd nuclei alpha energies and check of consistency of lifetimes, research groups are encouraged to publish or make readily available the decay data for individual events and not just report averages or mean lifetimes. In addition, research groups are encouraged to make readily available all the raw data (alpha energies, lifetimes, etc.), no matter how well or poorly they fit to a claimed level scheme.

Should the recommendations of the JWP prove, through future experiments, to be subject to reversal, there should be no issue with authorizing revisions as this has occurred in the past, *viz* with nobelium.

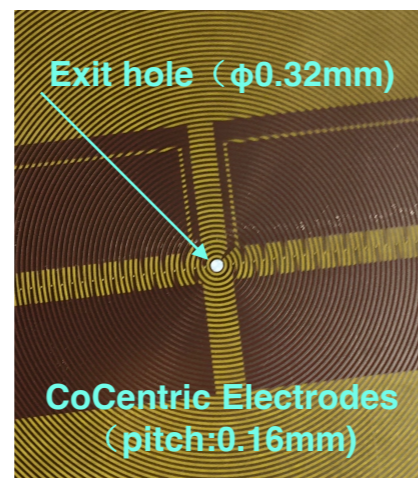
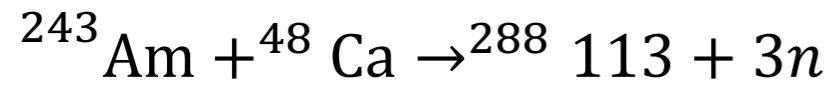
Experimental Setup at GARIS-II *(the structure and the location have been changed for several times)*

GARIS-II @ RIKEN Ring Cyclotron Facility

MRTOF-MS



fusion reactions, e.g.,

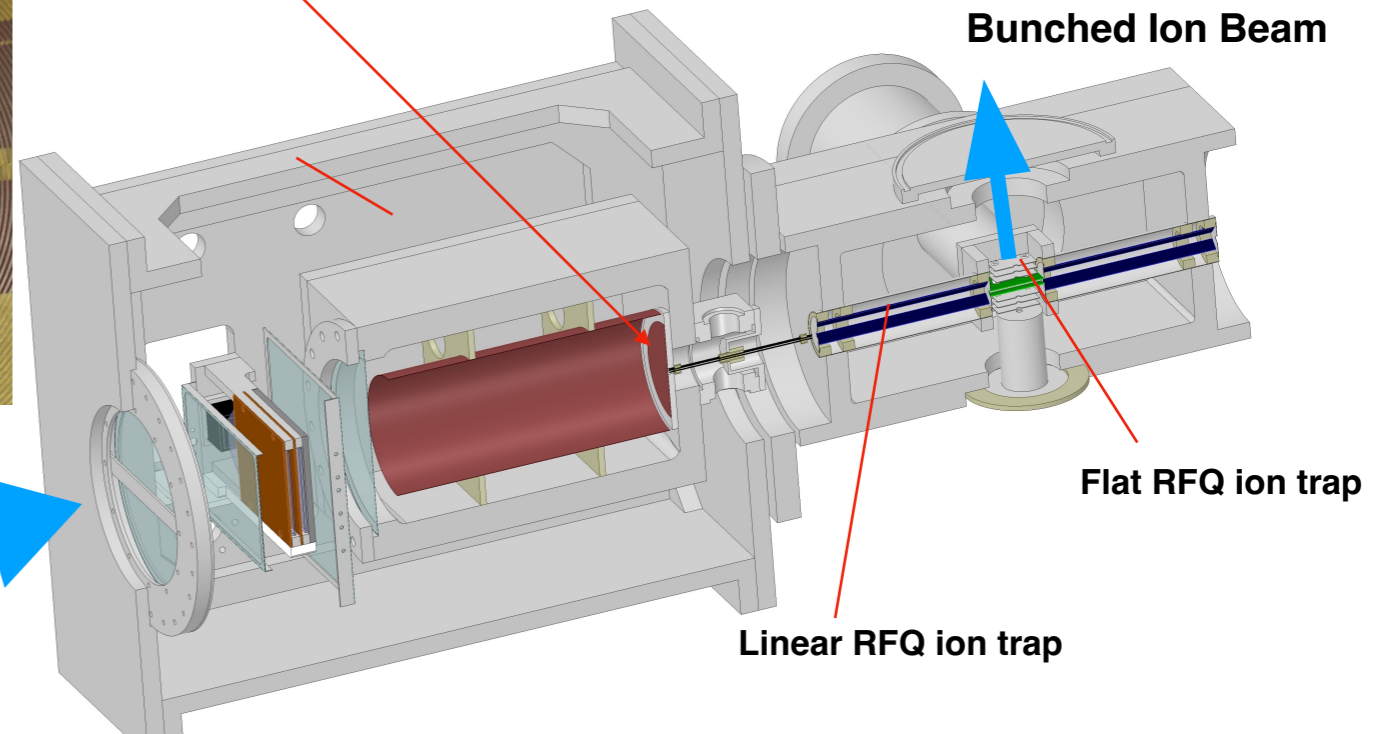


RF-Carpet(traveling wave)

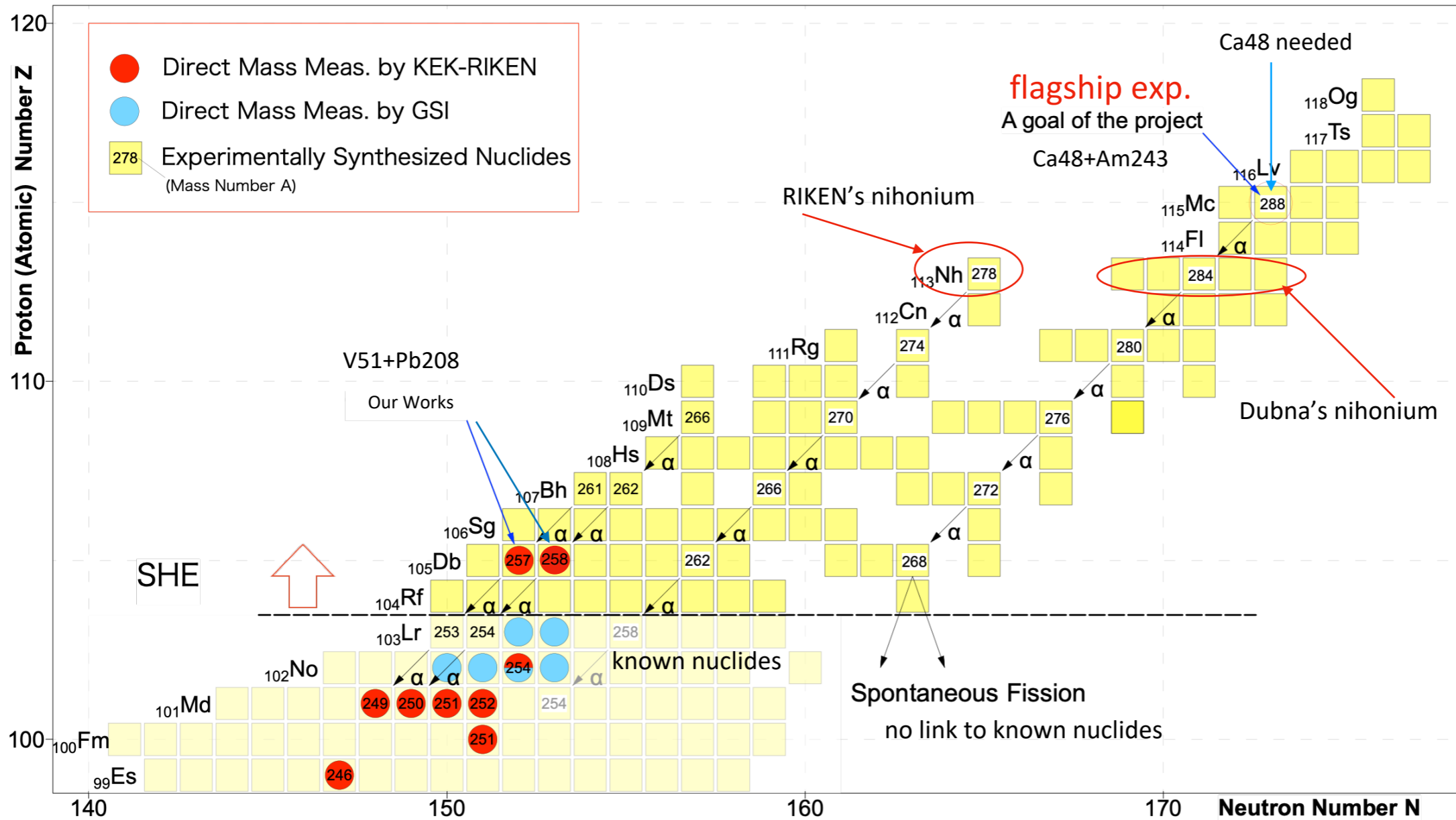
**Db⁺⁺, Db⁺⁺⁺, Md⁺⁺,
Bunched Ion Beam**

Db, Md, Es...

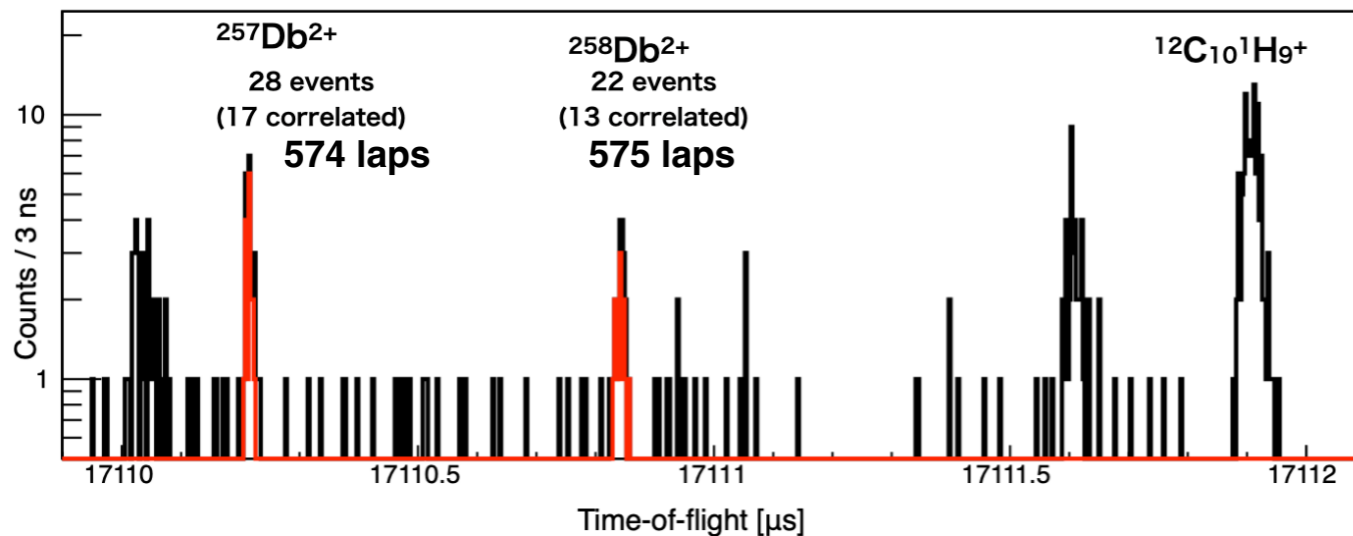
Recoil Ions



Mass Measurement of Superheavy elements @ GARIS-II MRTOF



ToF Singles Spectrum for Db isotopes



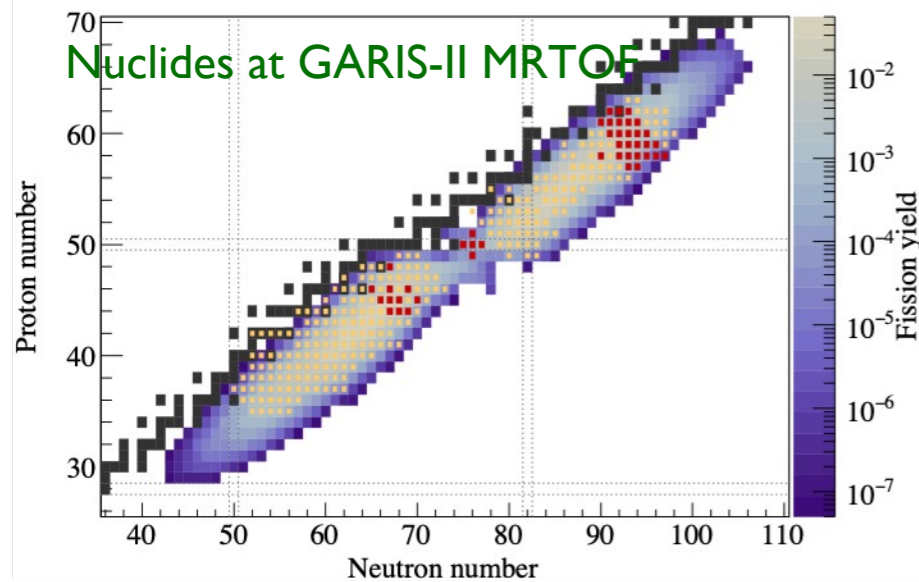
- **Setup has been ready for exp., however, difficult to obtain Ca48.**

Dev. of a self-purification system with CANDLE project

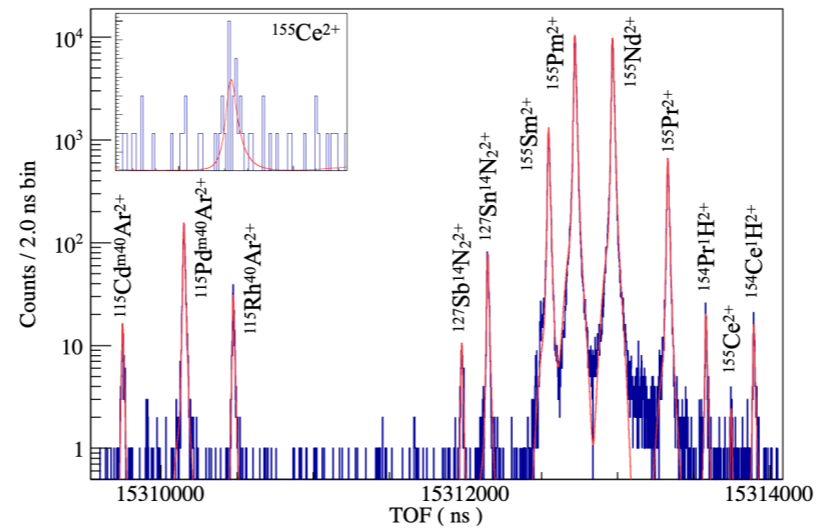
- **First SHE mass Db in NP2020-LINAC07**
 P. Schury et al, PRC104,L02I304(2021) & one in preparation
- **α -ToF detector R&D exp. for Ra isotopes**
 T. Niwase et al, NIMA 953(2020)163198
 T. Niwase et al, PRC104(2021)044617
independent $T_{1/2}$ determination of $^{206,207g,m}\text{Ra}$ and branching ratios

Off-line studies with Fission Source (252Cf, 248Cm)

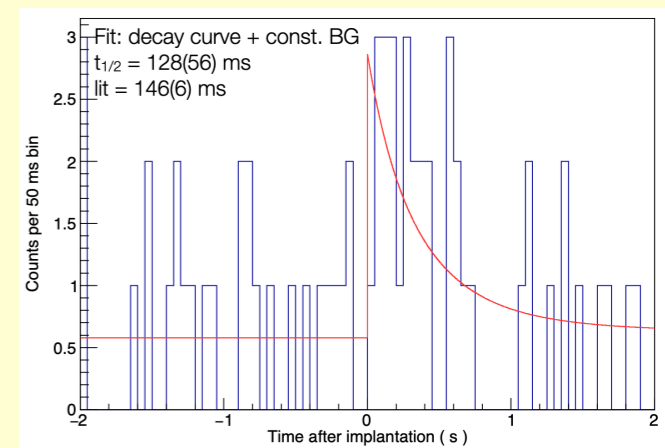
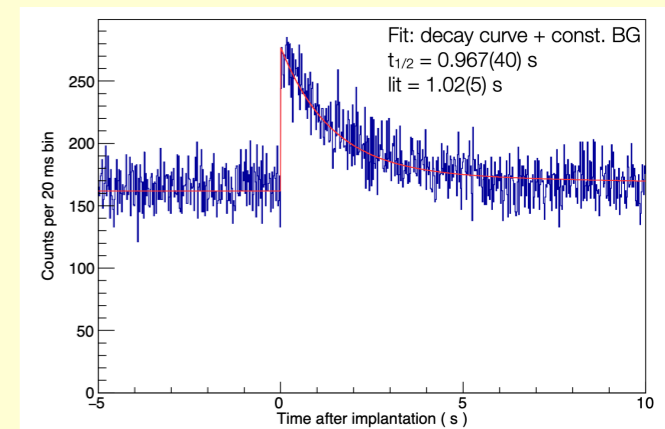
S. Kimura et al, submitting to PRC



ToF of new mass for Ce155



Decay spectra with beta-ToF detector



- >300 Nuclide's Mass were measured (a few *1st masses*, several improved masses).
- *High accuracy* with isobars and molecular ions in the same A/q.
- *beta-ToF* detector allows T1/2 determination and confirms short-lived nuclides (not molecular bg).
- New *atomic physics studies* with variety of elements, nuclear spins at very constant yields.
- *Health check* of the gas cell and total system

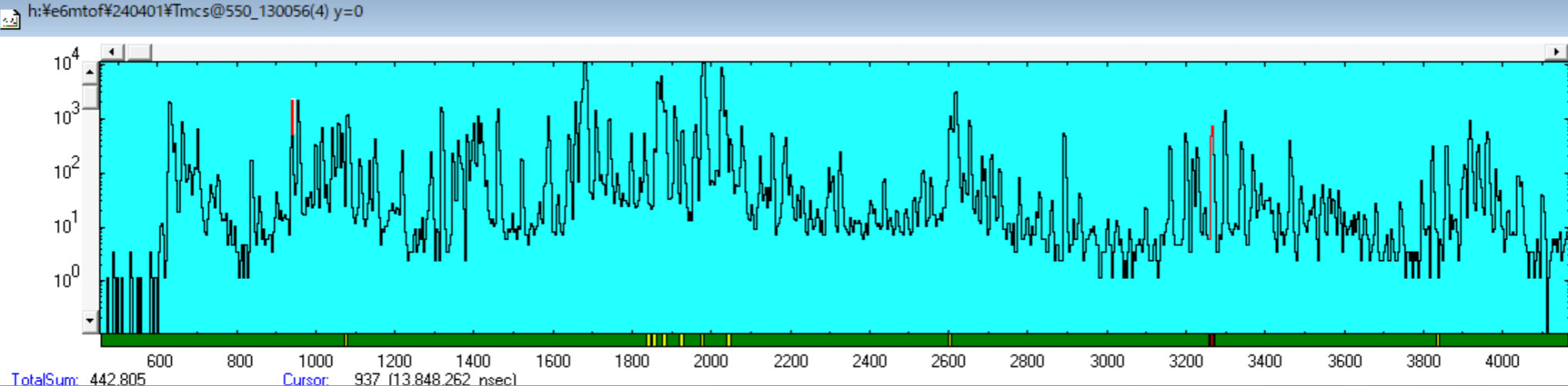
CoFP_153Nd@550_83
 Time: 512.740
 Sweeps: 20,510
 Starts: 20,490

-A1
 Total: 482,953
 ROI: 426,863
 Rate: 945

-A2
 Total: 0
 ROI: 0
 Rate: 0

-A3
 Total: 0
 ROI: 0
 Rate: 0

1GB FIFO:



no MF

MPANT - MCS6A

File Window Region Options Action ?

CoFP_153Nd@550_82
 Time: 500.805
 Sweeps: 20,023
 Starts: 19,986

-A1
 Total: 36,810
 ROI: 24,645
 Rate: 0

-A2
 Total: 0
 ROI: 0
 Rate: 0

-A3
 Total: 0
 ROI: 0
 Rate: 0

1GB FIFO:

Tfpga: 56
 Tboard: 54
 Data Lost: 0



MF for
 151,152,153,154

MPANT - MCS6A

File Window Region Options Action ?

CoFP_153Nd@550_87
 Time: 506.129
 Sweeps: 20,240
 Starts: 20,202

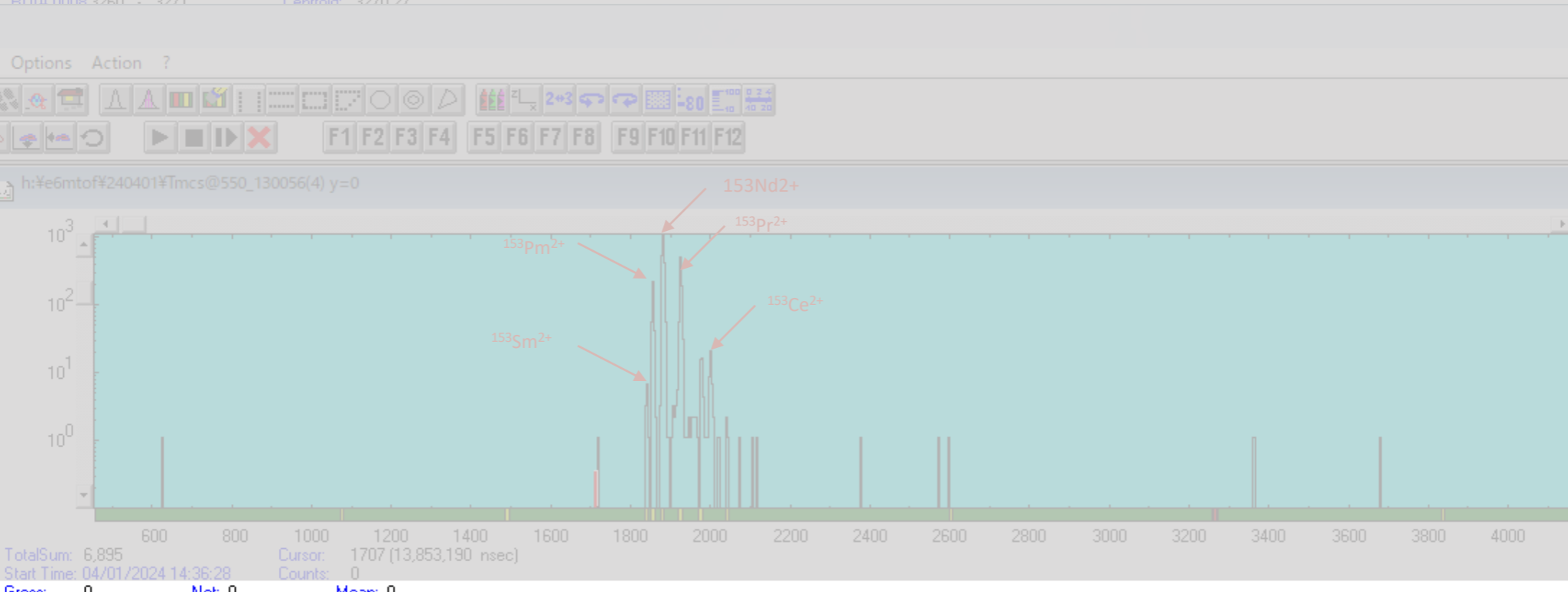
-A1
 Total: 20,295
 ROI: 6,915
 Rate: 15

-A2
 Total: 0
 ROI: 0
 Rate: 0

-A3
 Total: 0
 ROI: 0
 Rate: 0

1GB FIFO:

Tfpga: 56
 Tboard: 54

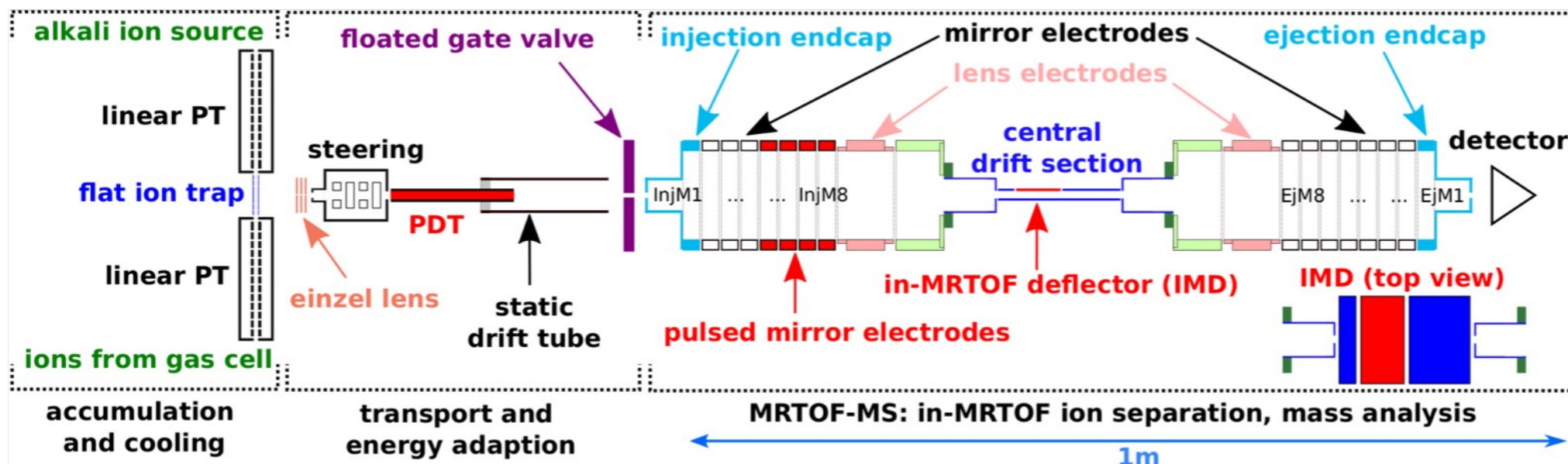


MF for 153

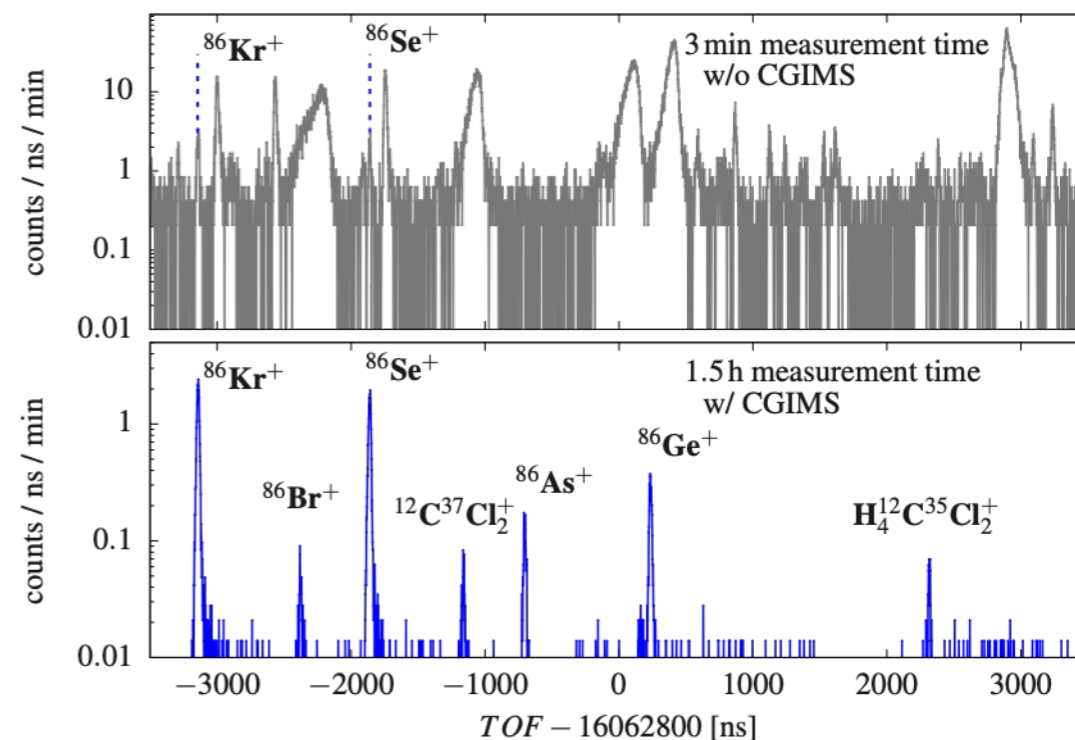
How to eliminate garbage ions?

M. Rosenbusch et al, NIMA1047, 167824 (2023)

In-MRTOF deflector



- A deflector in the flight tube of MRTOF with sophisticate pulse train kicks out unwanted mass/q ions
- Multiple (~5) sets of A/q numbers can be selected

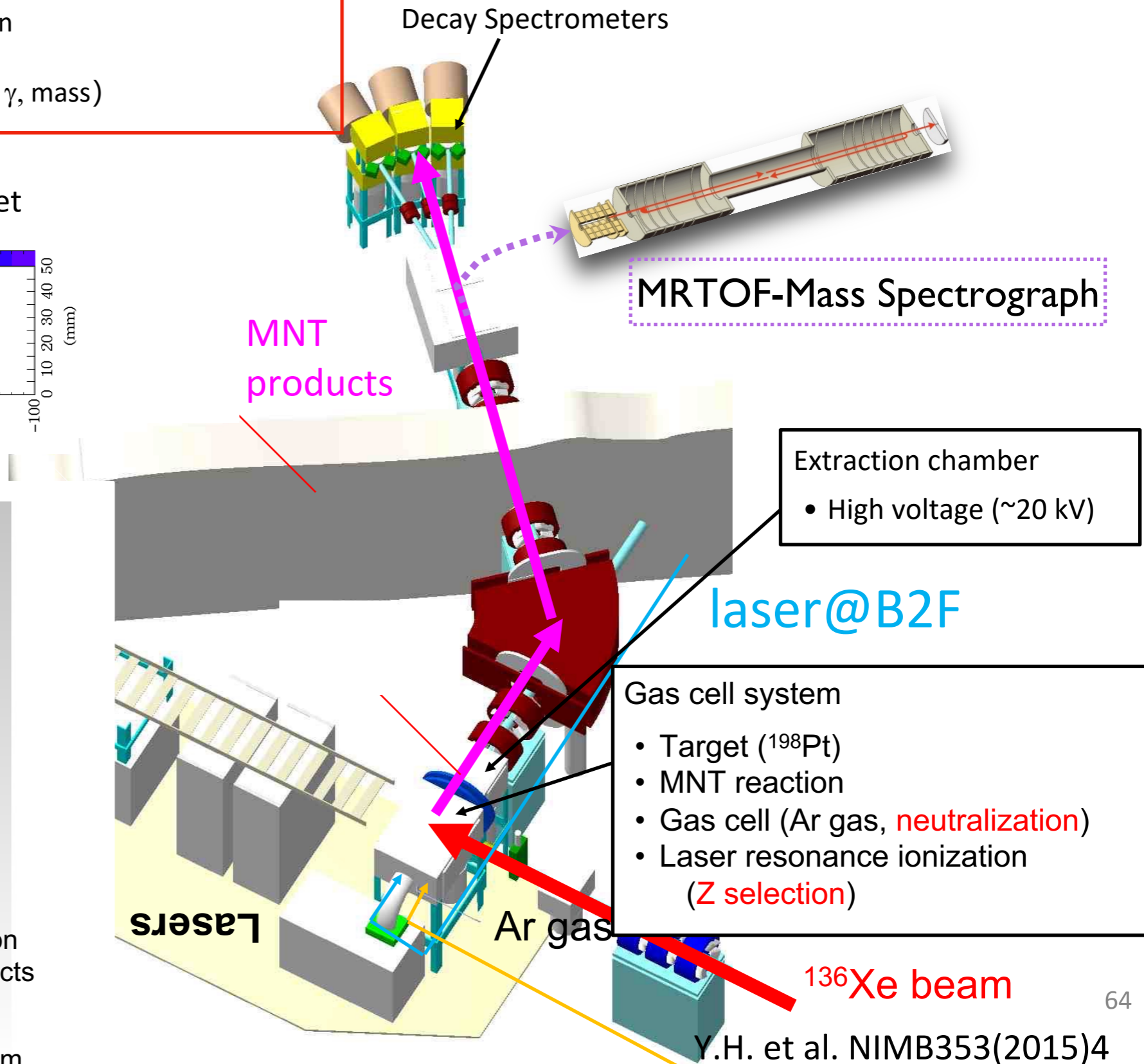
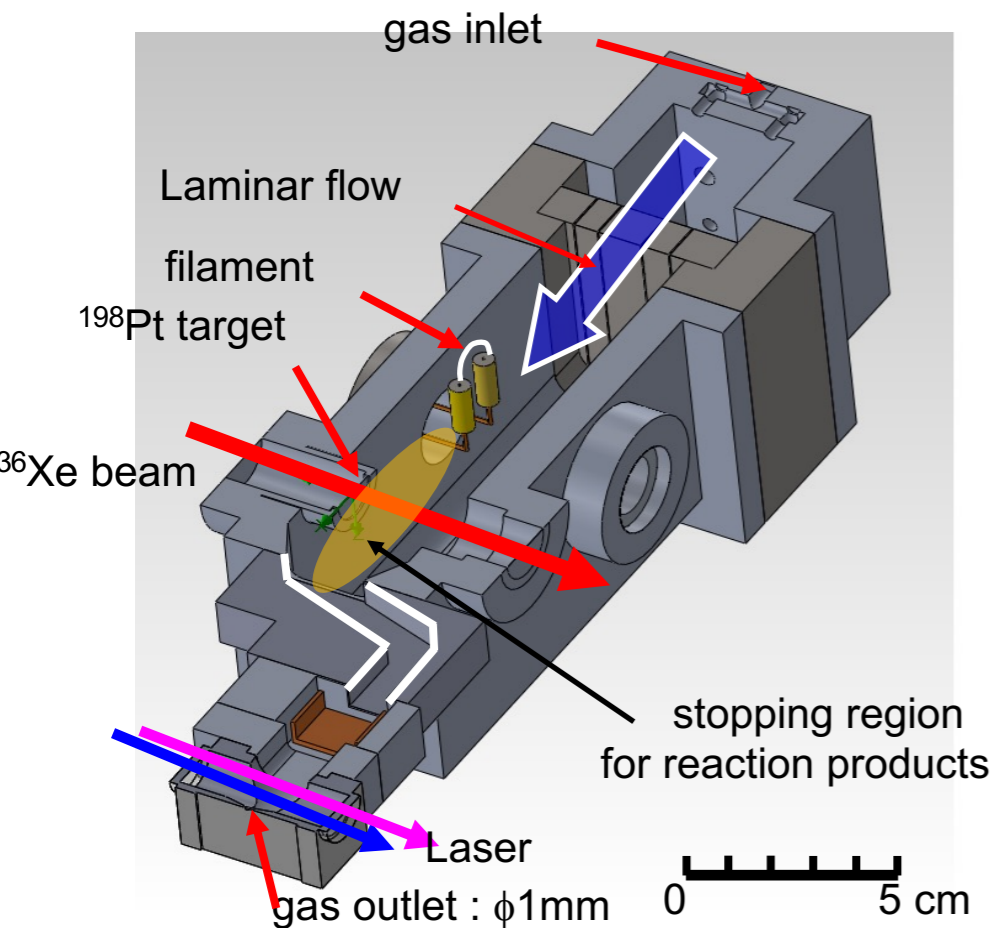
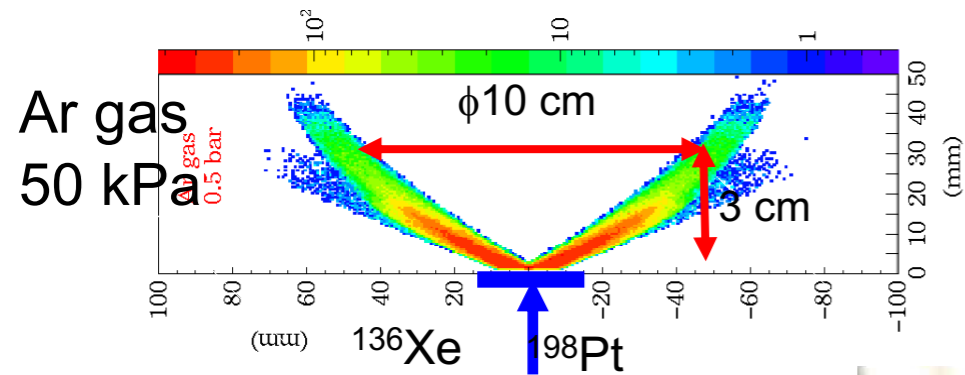


KISS: First and unique ISOL facility for online precision spectroscopy with MNT

Miyatake, Jeong et al

1. MNT fragments thermalize and neutralize in Ar
2. Element selection via resonant laser ionization
3. Mass separation via magnetic separator
4. precision spectroscopy with pure nuclide (β , γ , mass)

MNT reaction: ^{136}Xe beam + ^{198}Pt target



^{136}Xe beam

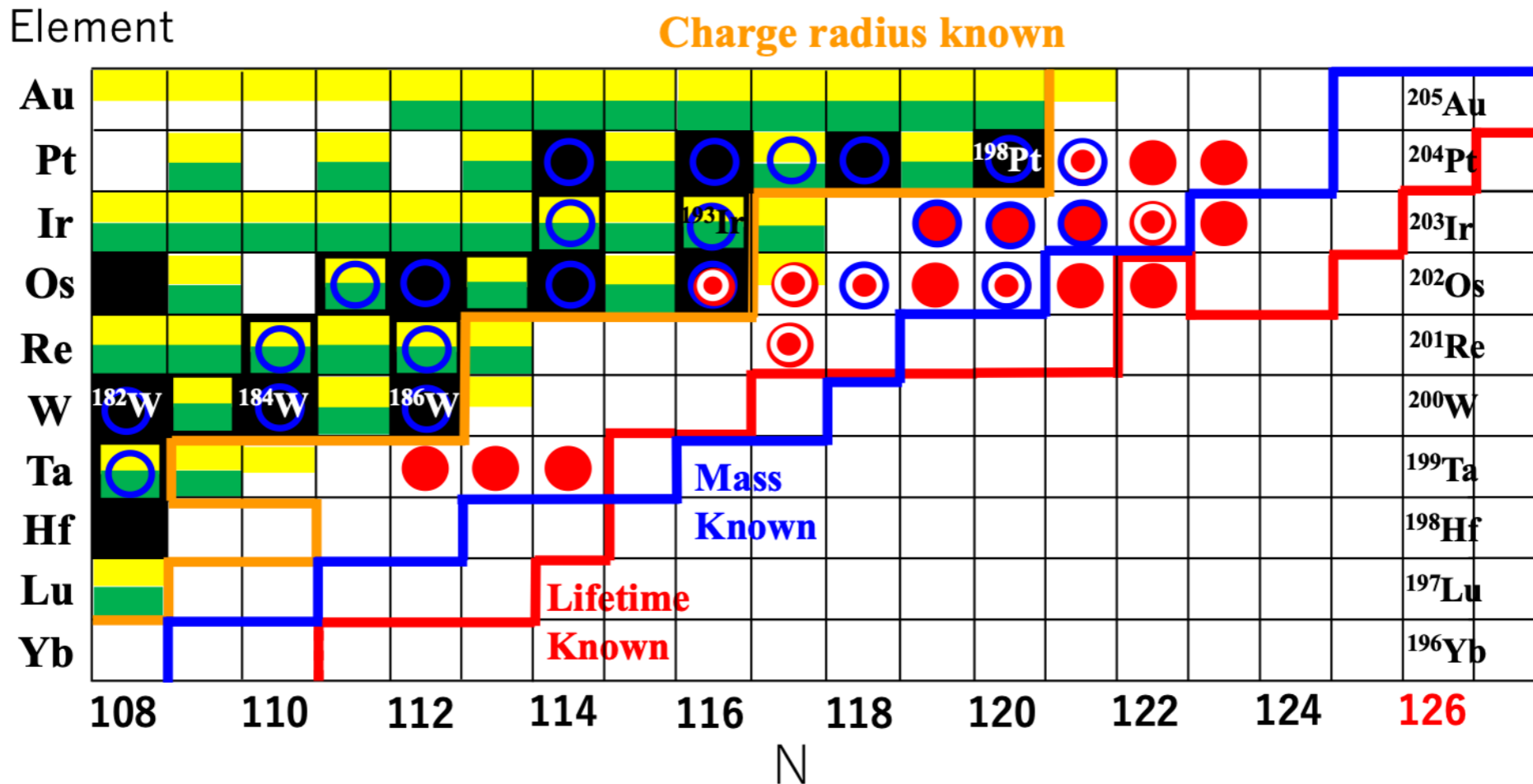
Nuclear spectroscopy at KISS

^{198}Pt , nat.Pt, nat.W targets

■ Stable Known

● β - γ spectroscopy at KISS ○ Mass at KISS

○ Laser spectroscopy at KISS



Not yet reached N=126, though

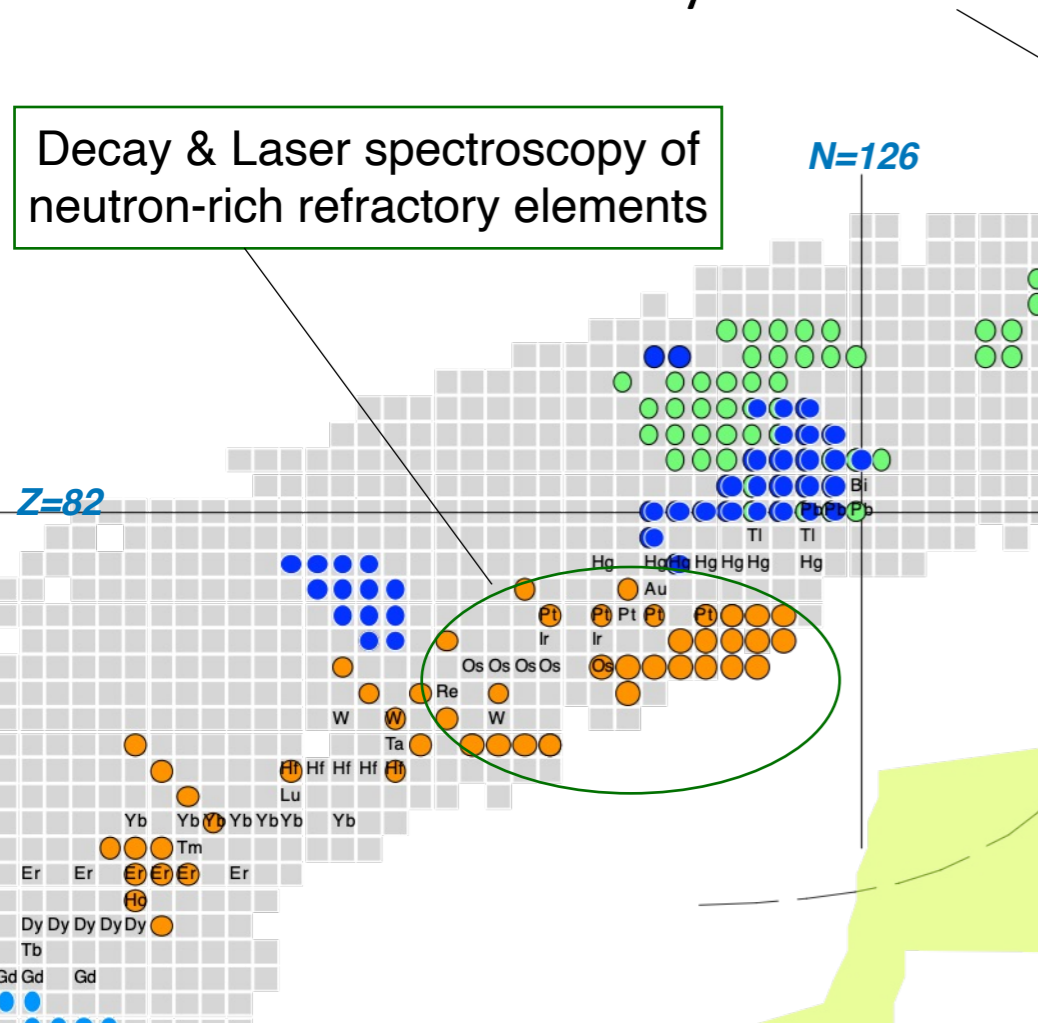
← Low primary beam intensity (10 pA)
Low gas cell efficiency (0.1%)

- Laser Spectroscopy (Charge radii, moments)
- $\beta\gamma$ spectroscopy for “difficult” refractory elements (T1/2, Decay mode)
- Mass Spectroscopy with MRTOF

Steady Scientific Results have been obtained

Discovery of a new U isotope at KISS MRTOF via mass spectrometry

Decay & Laser spectroscopy of neutron-rich refractory elements



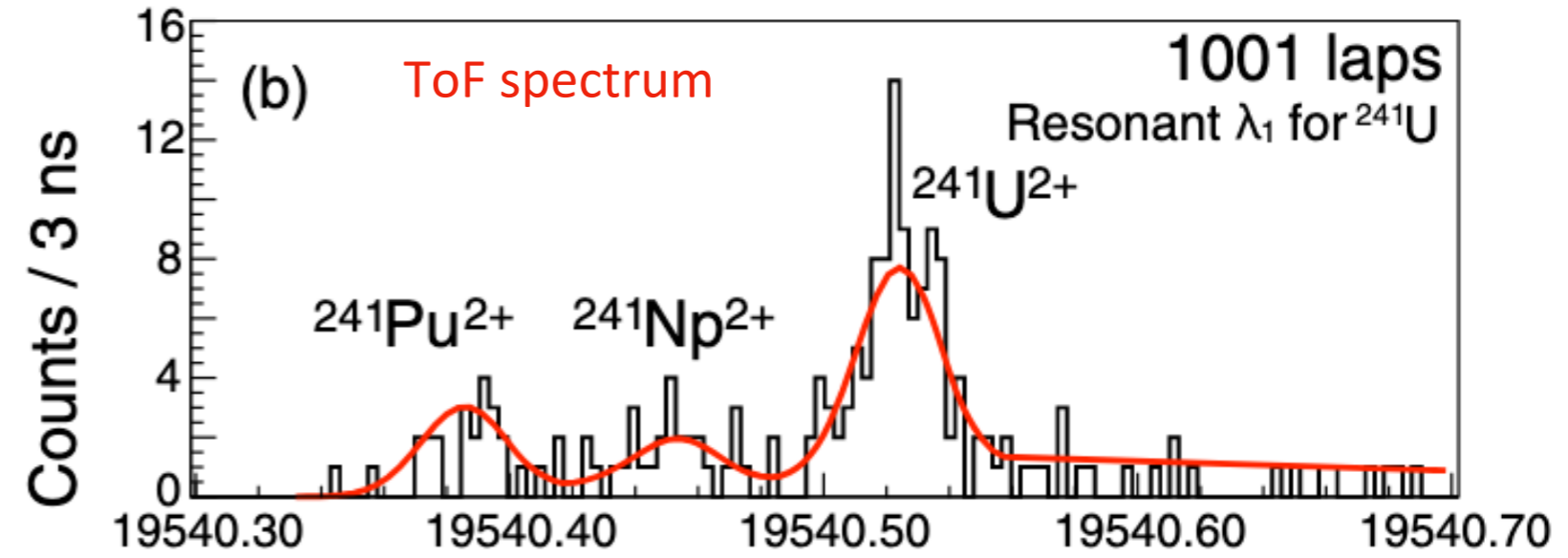
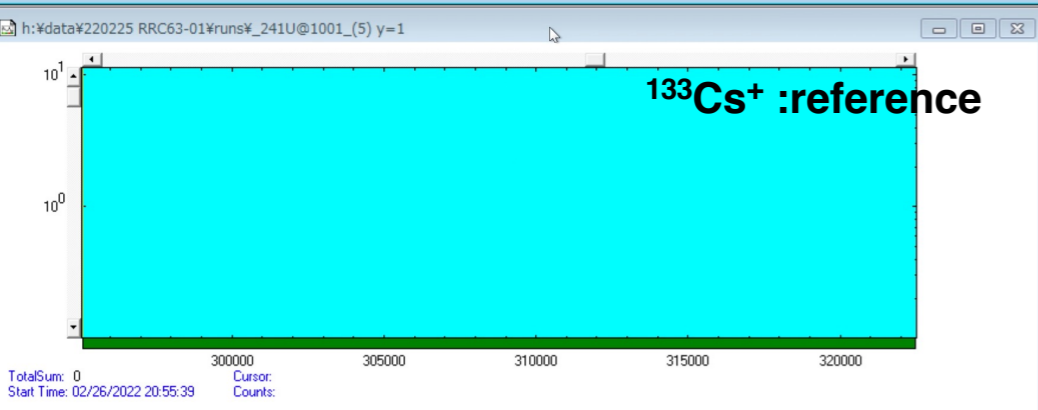
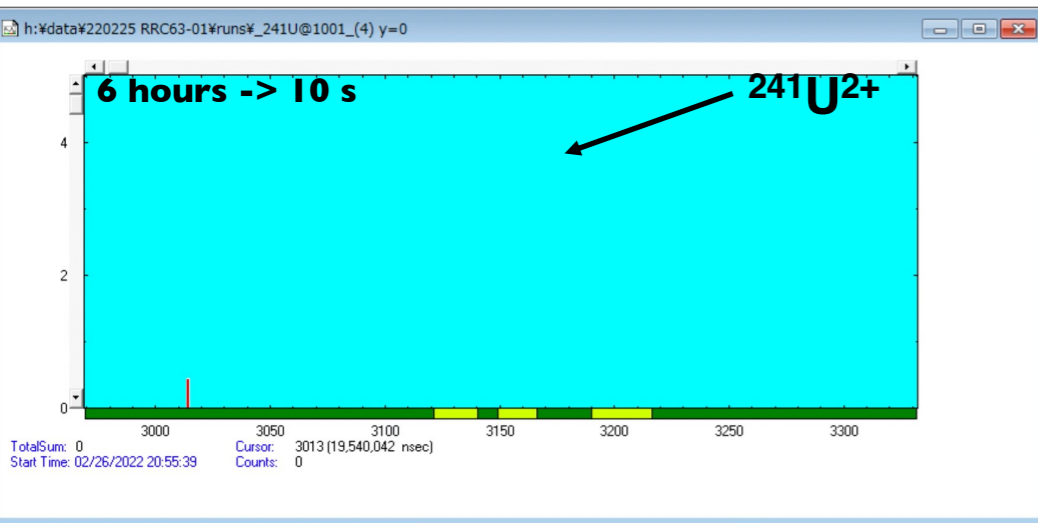
expected KISS-II frontier

mass frontier

²³⁸ Pu 87.7 y	²³⁹ Pu 2.4110·10 ⁴ y	²⁴⁰ Pu 6561 y	²⁴¹ Pu 14.290 y	²⁴² Pu 3.735·10 ⁵ y	²⁴³ Pu 4.956 h	²⁴⁴ Pu 5.11·10 ⁷ y	²⁴⁵ Pu 10.5 h	²⁴⁶ Pu 10.84 d	²⁴⁷ Pu 2.27 d	²⁴⁸ Pu 48.3 m	²⁴⁹ Pu 1.58 m	²⁵⁰ Pu 1.80 m
²³⁷ Np 2.144·10 ⁶ y	²³⁸ Np 2.117 d	²³⁹ Np 2.356 d	²⁴⁰ Np 1.032 h ★7.22 m	²⁴¹ Np 13.9 m	²⁴² Np ★5.5 m 2.2 m	²⁴³ Np 1.85 m	²⁴⁴ Np 2.29 m	²⁴⁵ Np 2.29 m	²⁴⁶ Np 14.5 s	²⁴⁷ Np 27.7 s	²⁴⁸ Np 6.11 s	²⁴⁹ Np 7.24 s
²³⁶ U 2.342·10 ⁷ y	²³⁷ U 6.752 d	²³⁸ U 99.2742 4.468·10 ⁹ y	²³⁹ U 23.45 m	²⁴⁰ U 14.1 h	²⁴¹ U 45.6 m	²⁴² U 16.8 m	²⁴³ U 3.04 m	²⁴⁴ U 2.67 m	²⁴⁵ U 29.2 s	²⁴⁶ U 22.6 s	²⁴⁷ U 7.54 s	156
²³⁵ Pa 24.44 m	²³⁶ Pa 9.1 m	²³⁷ Pa 8.7 m	²³⁸ Pa 2.27 m	²³⁹ Pa 1.8 h	²⁴⁰ Pa 20.1 s	²⁴¹ Pa 32.3 s	²⁴² Pa 6.77 s	²⁴³ Pa 7.18 s	²⁴⁴ Pa 2.59 s	²⁴⁵ Pa 2.77 s	155	
²³⁴ Th 24.10 d	²³⁵ Th 7.2 m	²³⁶ Th 37.3 m	²³⁷ Th 4.8 m	²³⁸ Th 9.4 m	²³⁹ Th 35.4 s	²⁴⁰ Th 33.0 s	²⁴¹ Th 7.08 s	²⁴² Th 5.76 s	153	154		

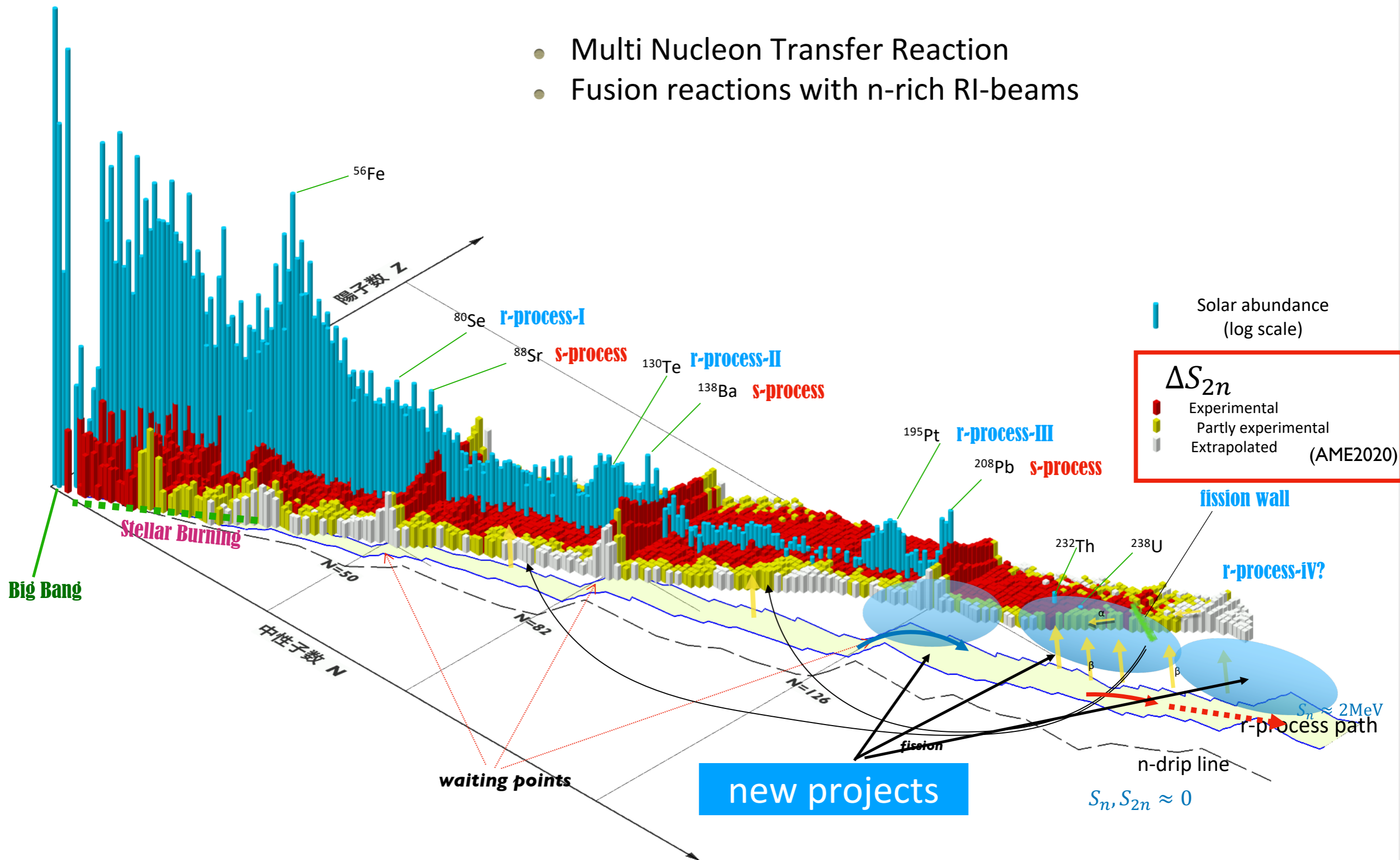
new nuclide discovered by mass measurement

T. Niwase, Y. Watanabe et al, PRL130,132502(2023)



How to access N=126, origin of Th U, end of r-process?

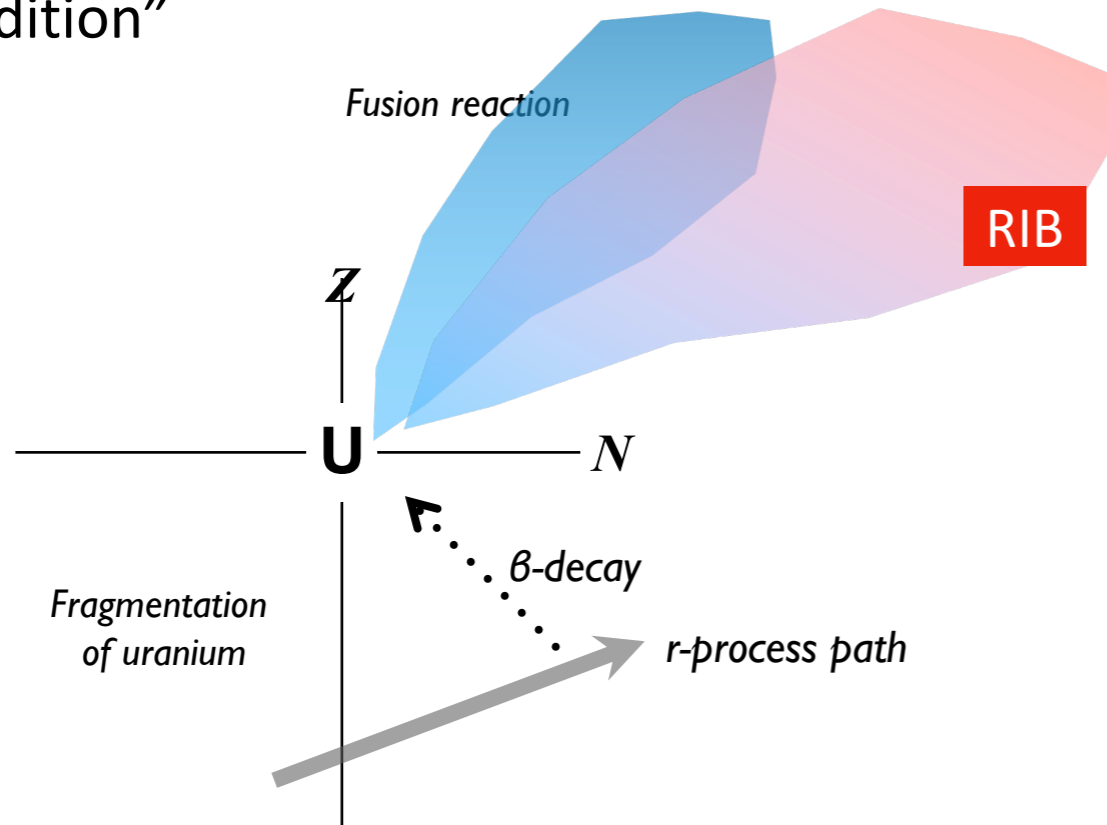
- Multi Nucleon Transfer Reaction
- Fusion reactions with n-rich RI-beams



How to access progenitors of uranium and end of r-process ?

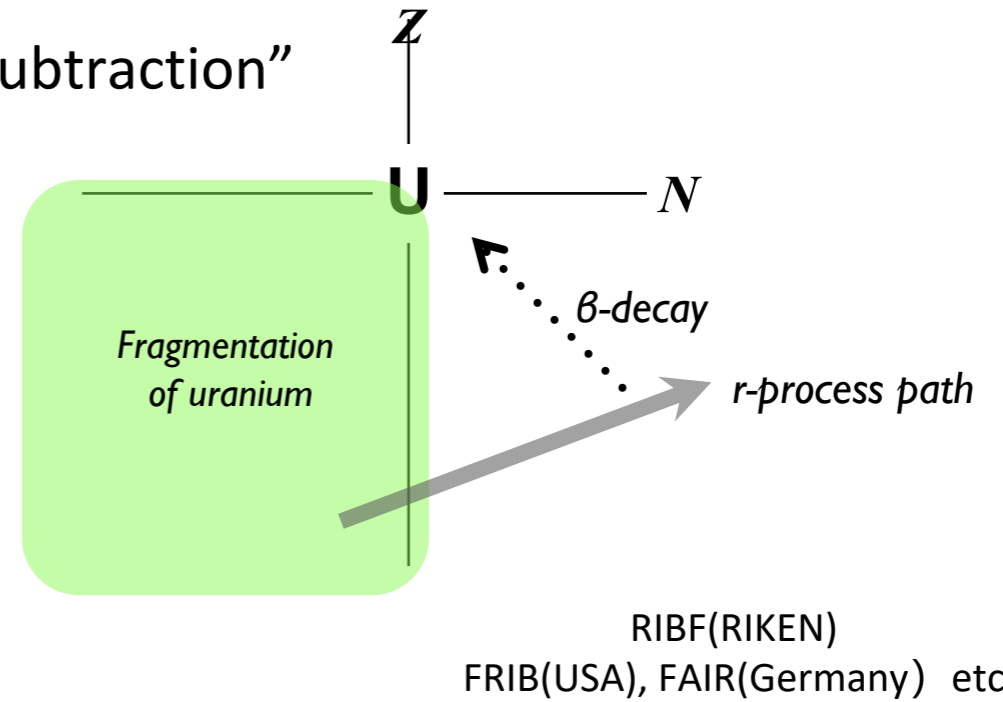
Fusion (~5 MeV/u)

“Addition”



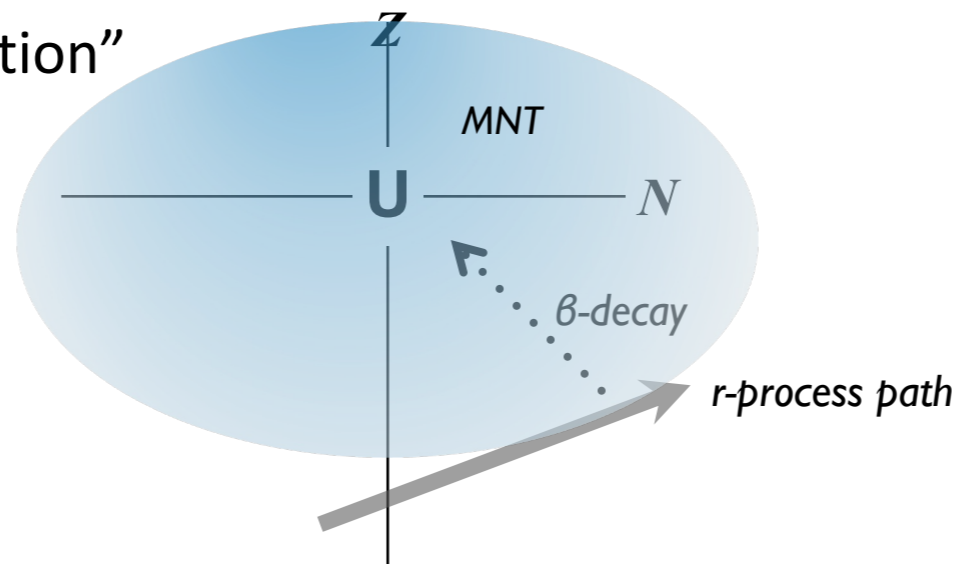
Fragmentation/Spallation (~100 MeV/u)

“Subtraction”



Multi-Nucleon Transfer (5~10 MeV/u)

“Addition” + “Subtraction”



“Add” neutrons
“Subtract” protons
south-east of uranium

KISS, KISS-II
NEXT, N=126 factory,
STAR, HIAF-MNT?

KISS-II “MNT reaction products, measure multiple species at once”

- Primary beam separator: High intensity beam
- RF ion guide He gas cell: Efficient collection of all elements
- MRTOF: Multiple species, Tag for spectroscopy

	primary beam	total efficiency	#nuclides / unit time	total gain
KISS	10 pA	0.1%	1	1
KISS-2	1000 pA	>1%	>10	>10000
	primary separator	RF gas catcher	MRTOF	

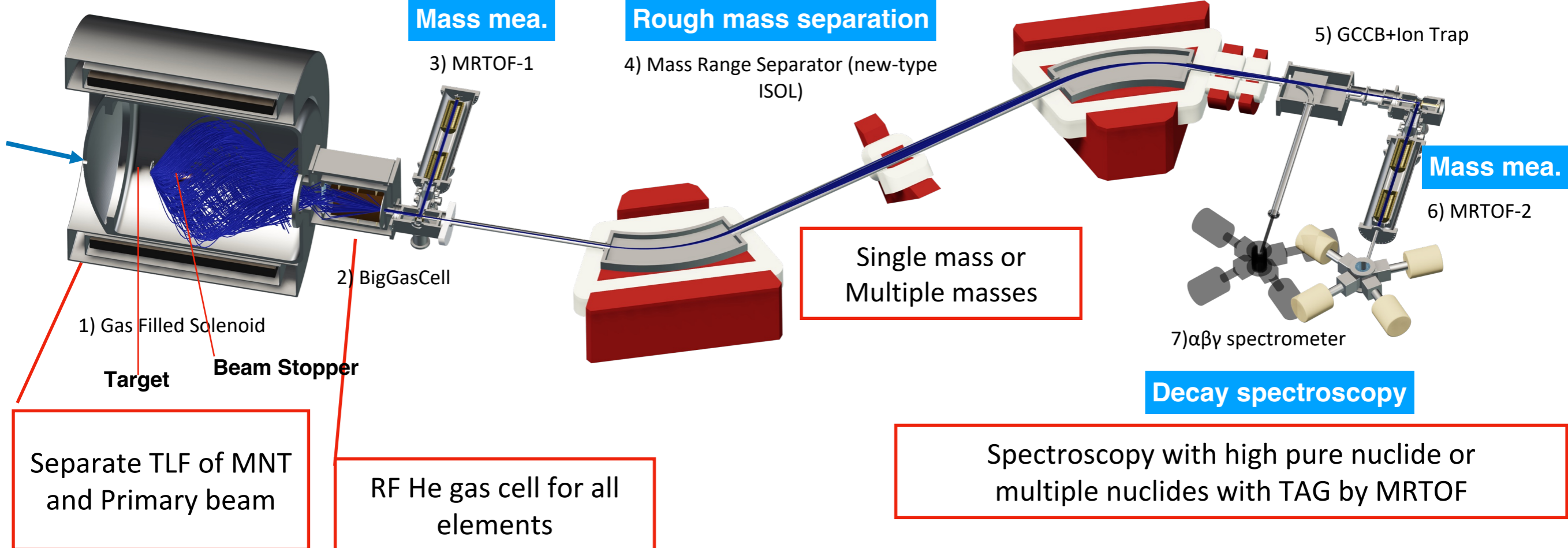
Primary beam separation

Mass mea.

Rough mass separation

Mass mea.

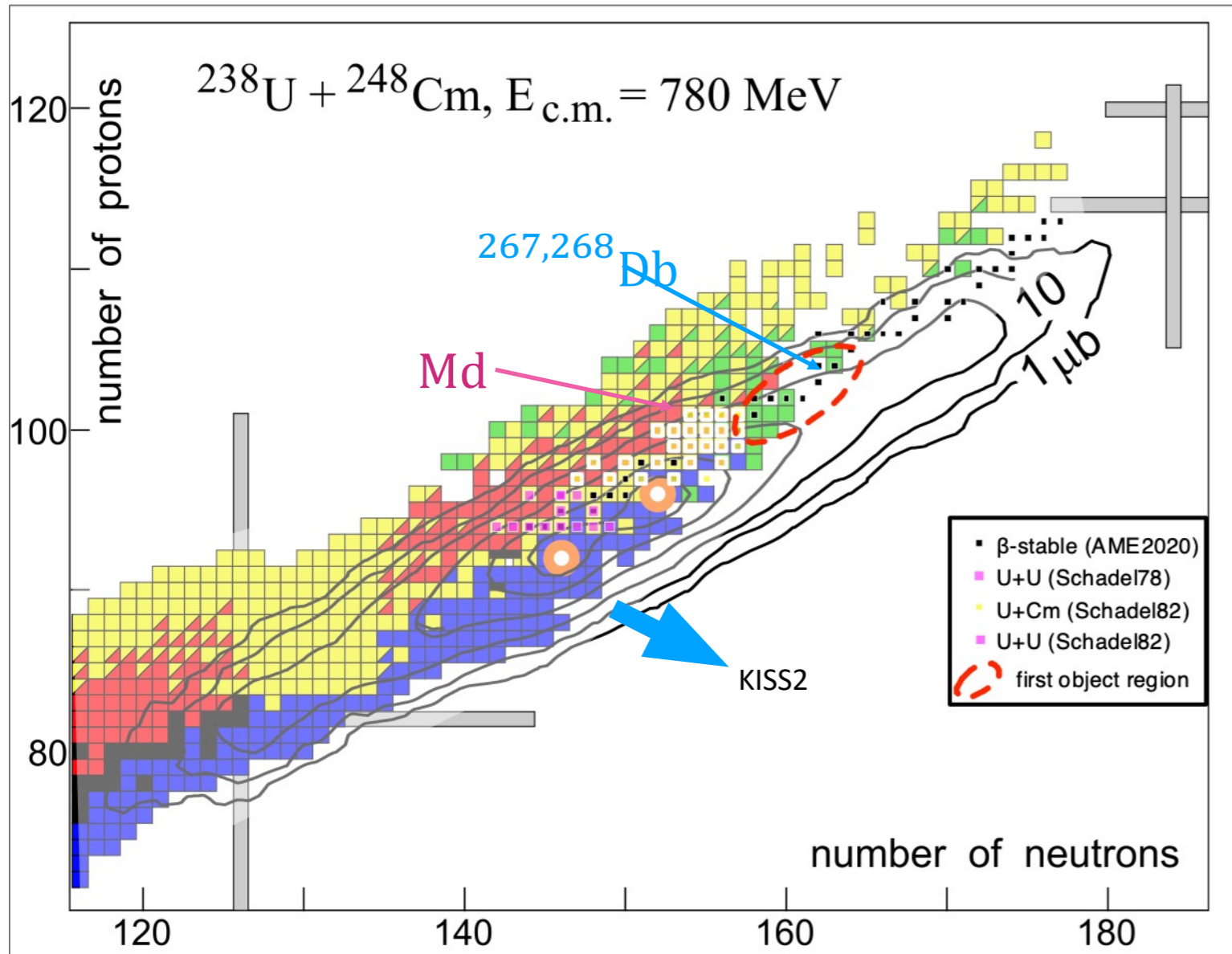
Decay spectroscopy



total budget: \$15M in 5 (setup) + 5 (exp.) years , failed

KISS-1.5: \$5M in 5 years, starting 2024 by Watanabe JSPS

★ Multi-Nucleon Transfer reaction towards SHN

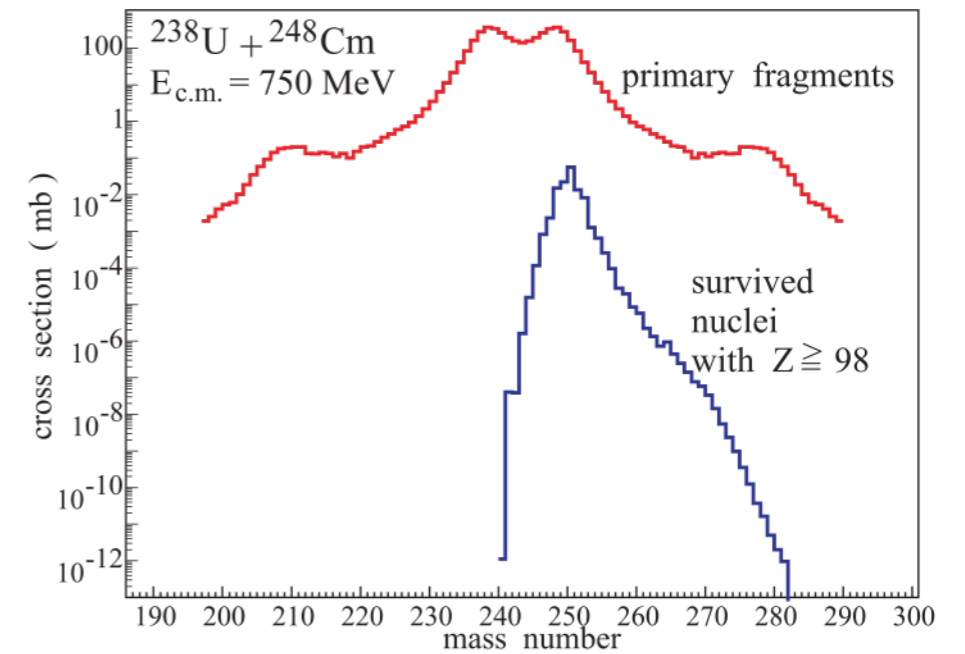


W. Greiner and V. Zagrevaev, Nucl. Phys. A 834, 323c (2010) , retouch by M.W.

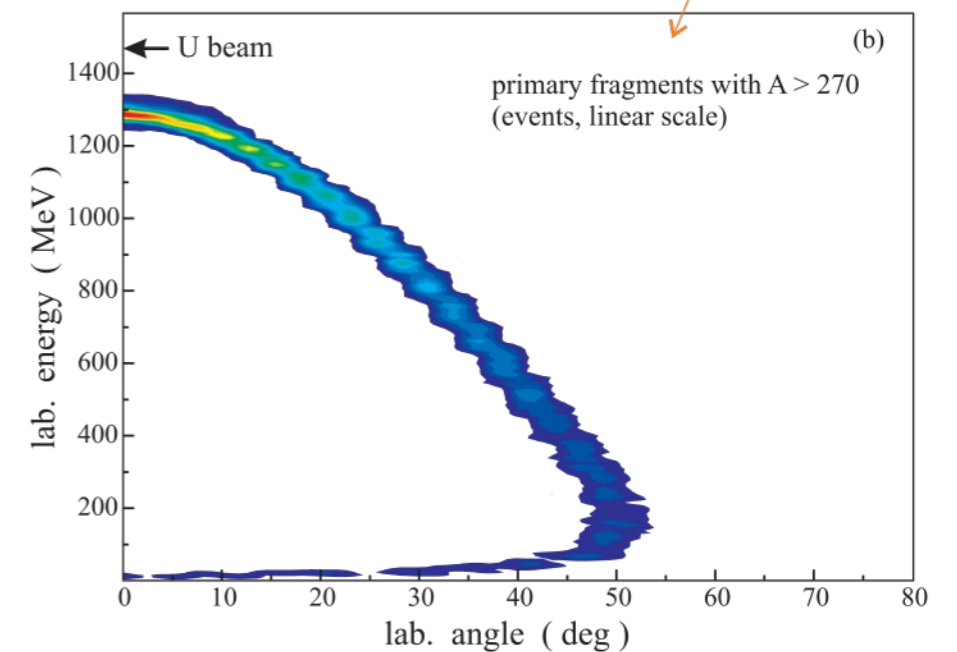
Radiochemical or Successive α -decay

are not applicable to β -decaying, long-lived nuclides

V.I. Zagrevaev and W. Greiner, Phys. Rev. C83, 044618 (2011).

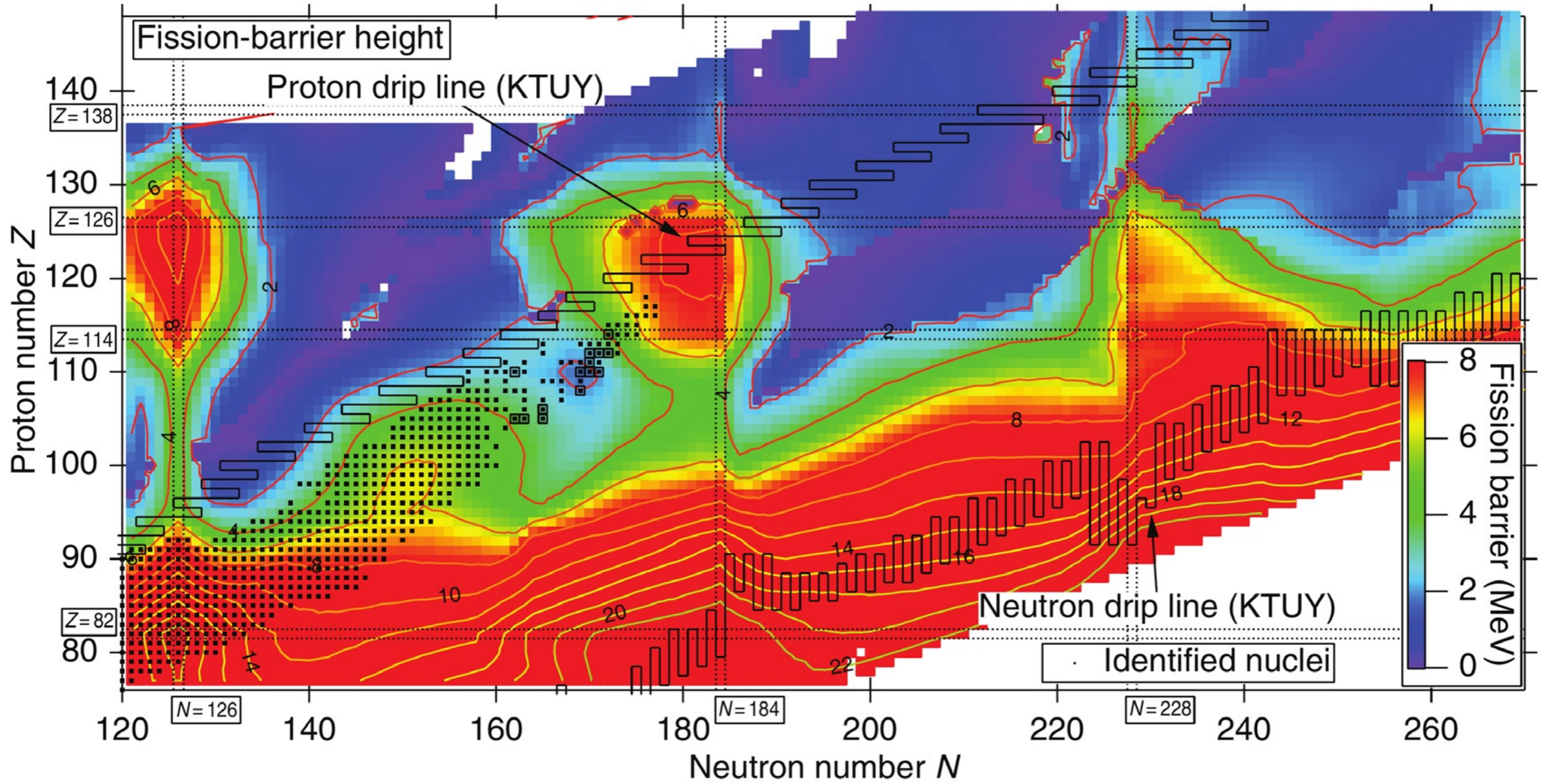


Survival probabilities are orders of mag. low

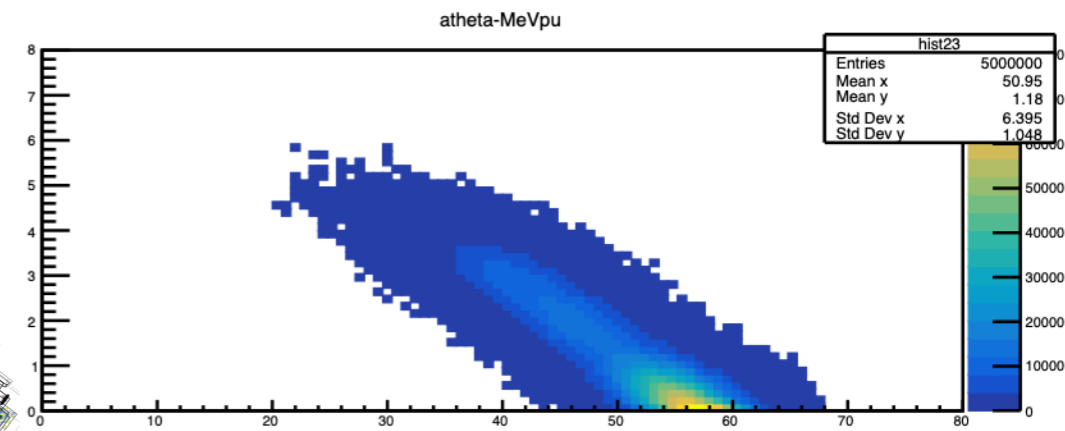
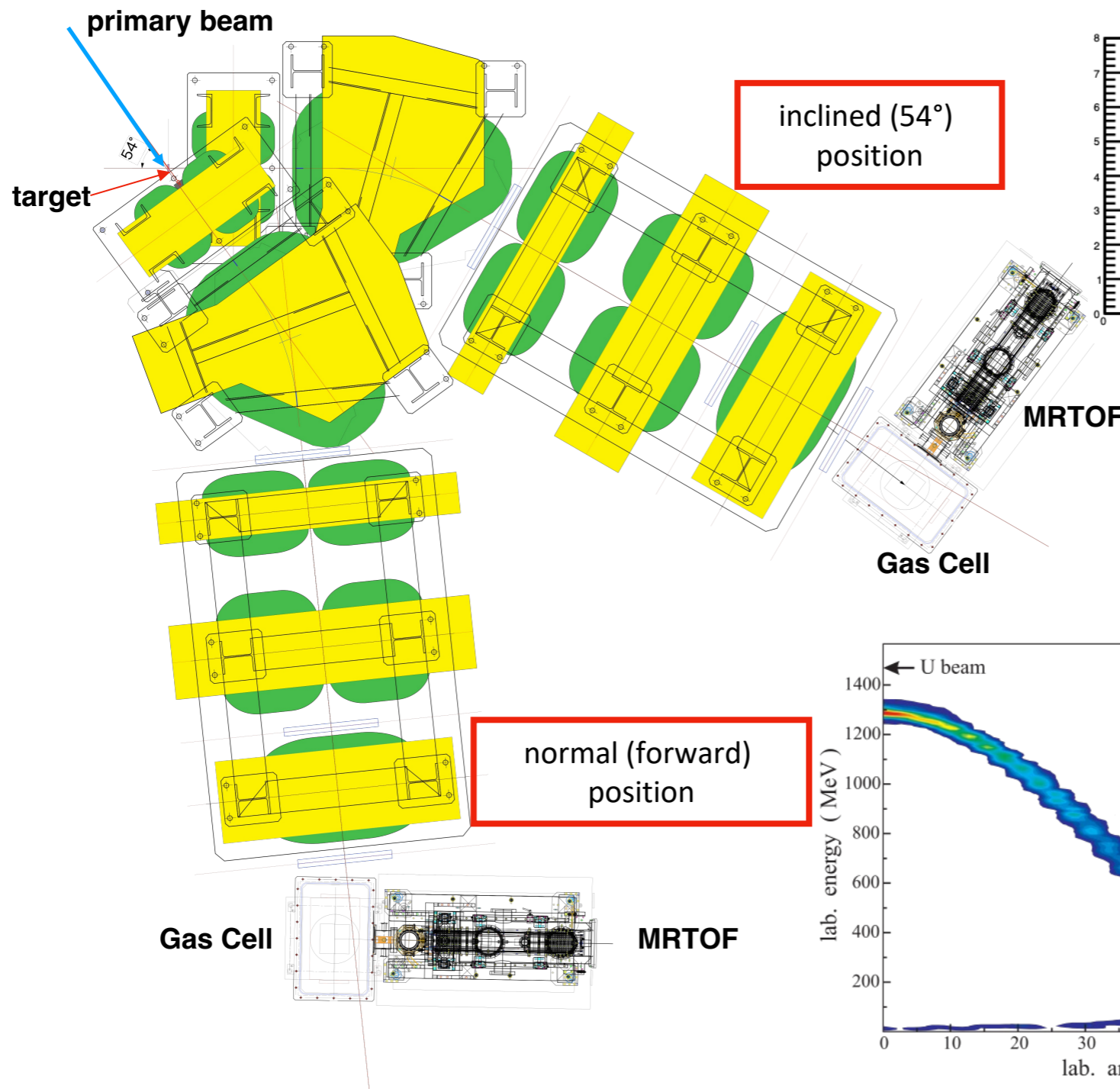


Heavy fragments go forward

Fission barrier increases approaching to IoS



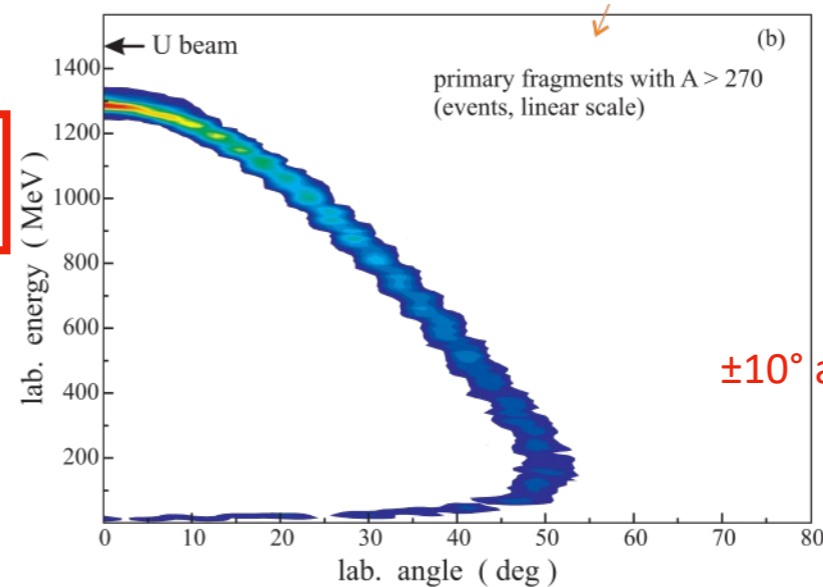
Two Configurations of BigGARIS with MRTOF Mass Spectrograph



n-rich direction (E, SE)
“Large angle horn”

Part of the “Horn”, $\epsilon_{\text{collection}} \sim 5\%$ feasible

KISS 1.7 ?



SHE direction (NE)
“Forward”

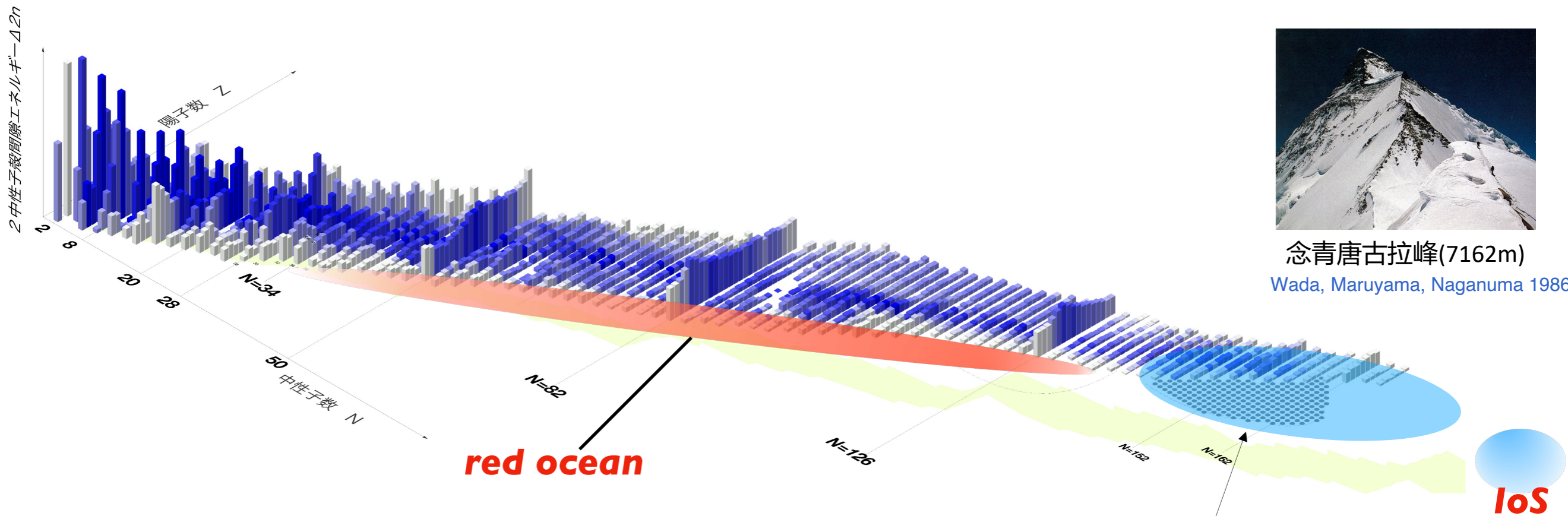
$\pm 10^\circ$ accepts, $\epsilon_{\text{collection}} \sim 50\%$ feasible

V.I. Zagrebaev, W. Greiner, PRC83, 044618(2011)

Cm target 1 mg/cm^2 , U beam 3 puA , $\epsilon_{\text{collection}} \times \epsilon_{\text{measure}} \sim 6\%$:

10 pb ($267, 268 \text{ Db}$) produces 1 event/2 days

“Boldly go where no man has gone before” © Star Trek



念青唐古拉峰(7162m)
Wada, Maruyama, Naganuma 1986



珠穆朗玛峰(8192m)

[Tenzing Norgay](#) and [Edmund Hillary](#), 1953

n-rich actinide is “blue ocean,”
>200 nuclides are waiting to discover

