Synthesis of Superheavy Elements at RIKEN

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5. Solution chemistry of Rf and Db
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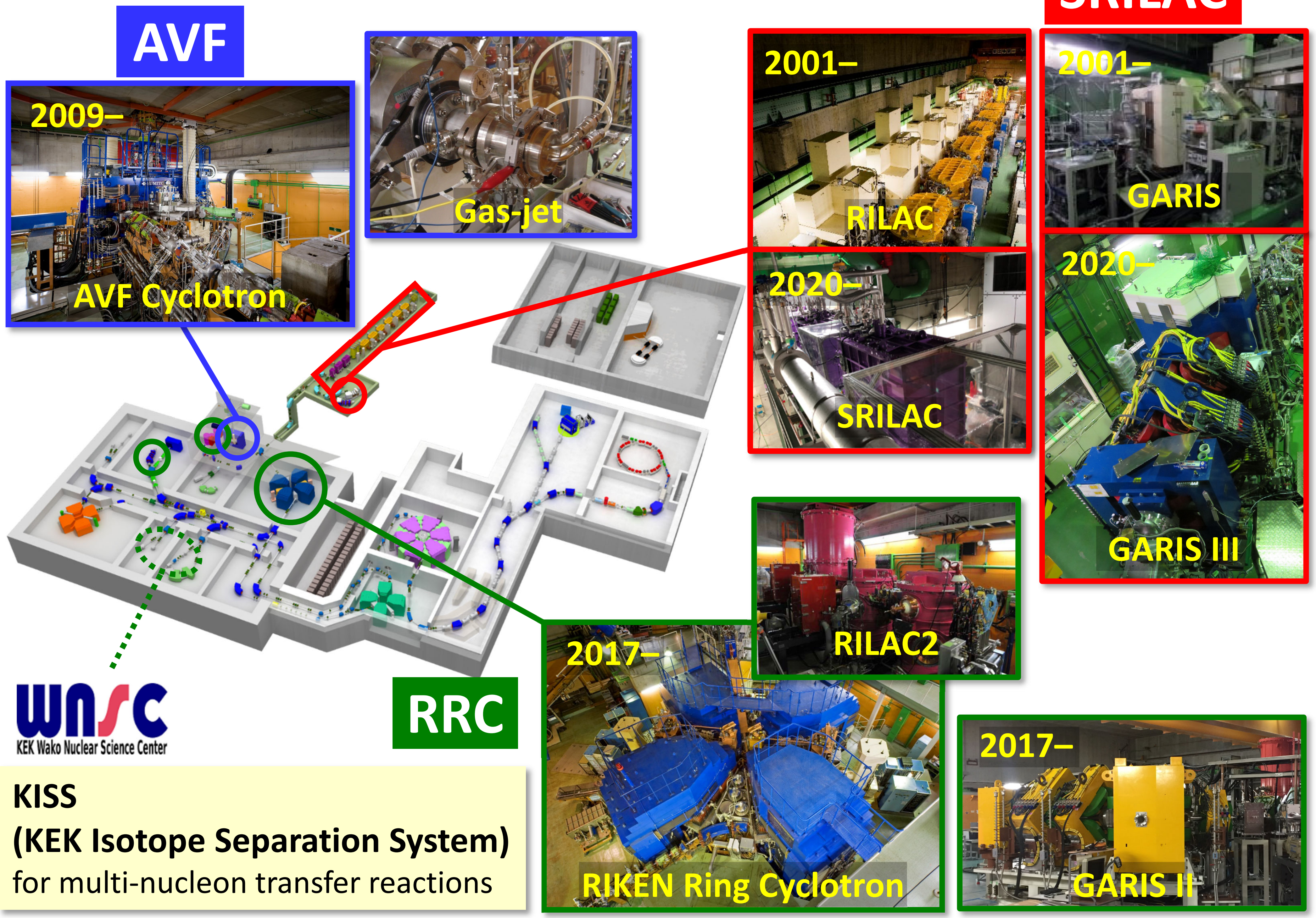
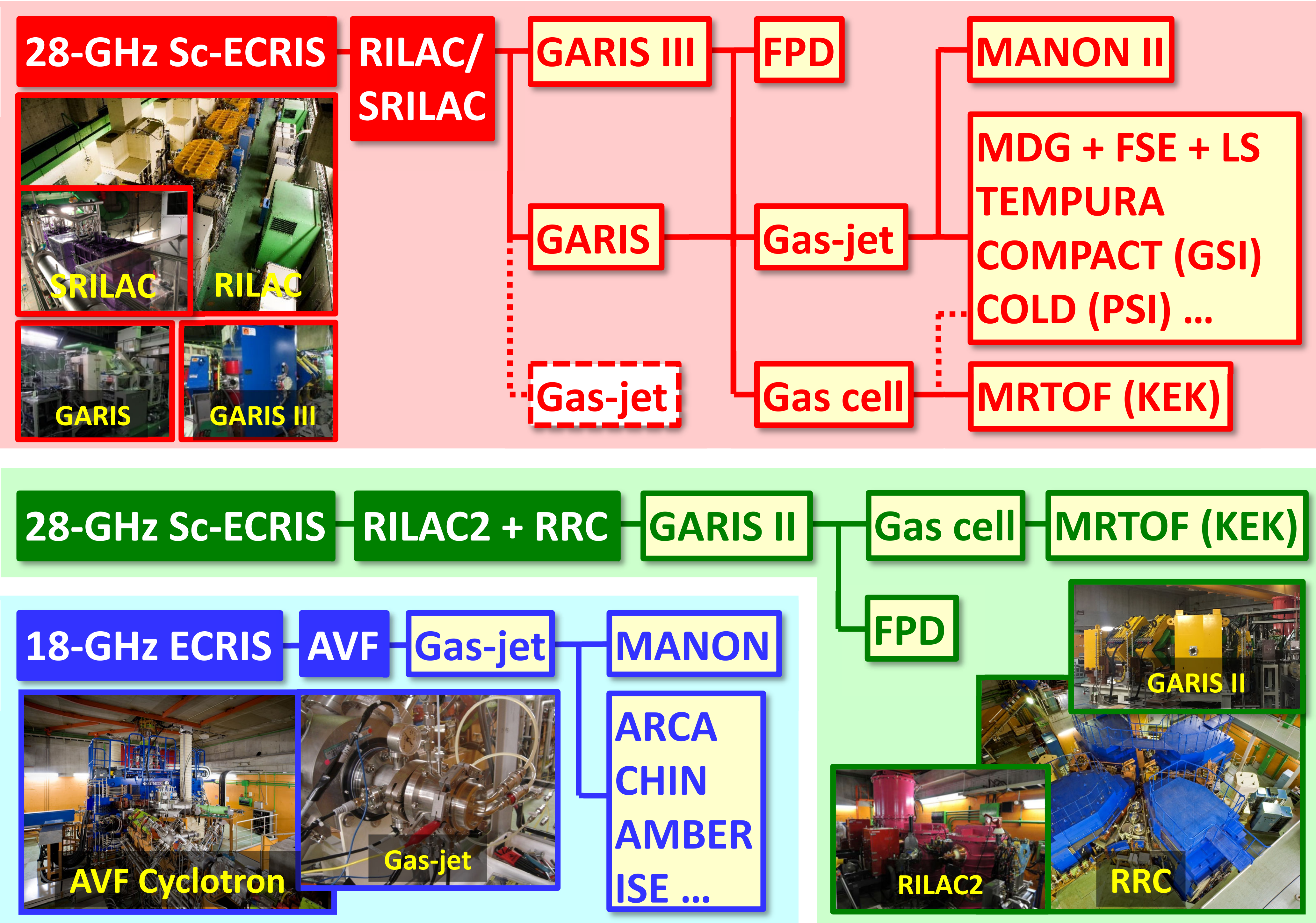
1. Facilities for SHE research at RIKEN RI Beam Factory

Facilities for SHE research in RIKEN RIBF

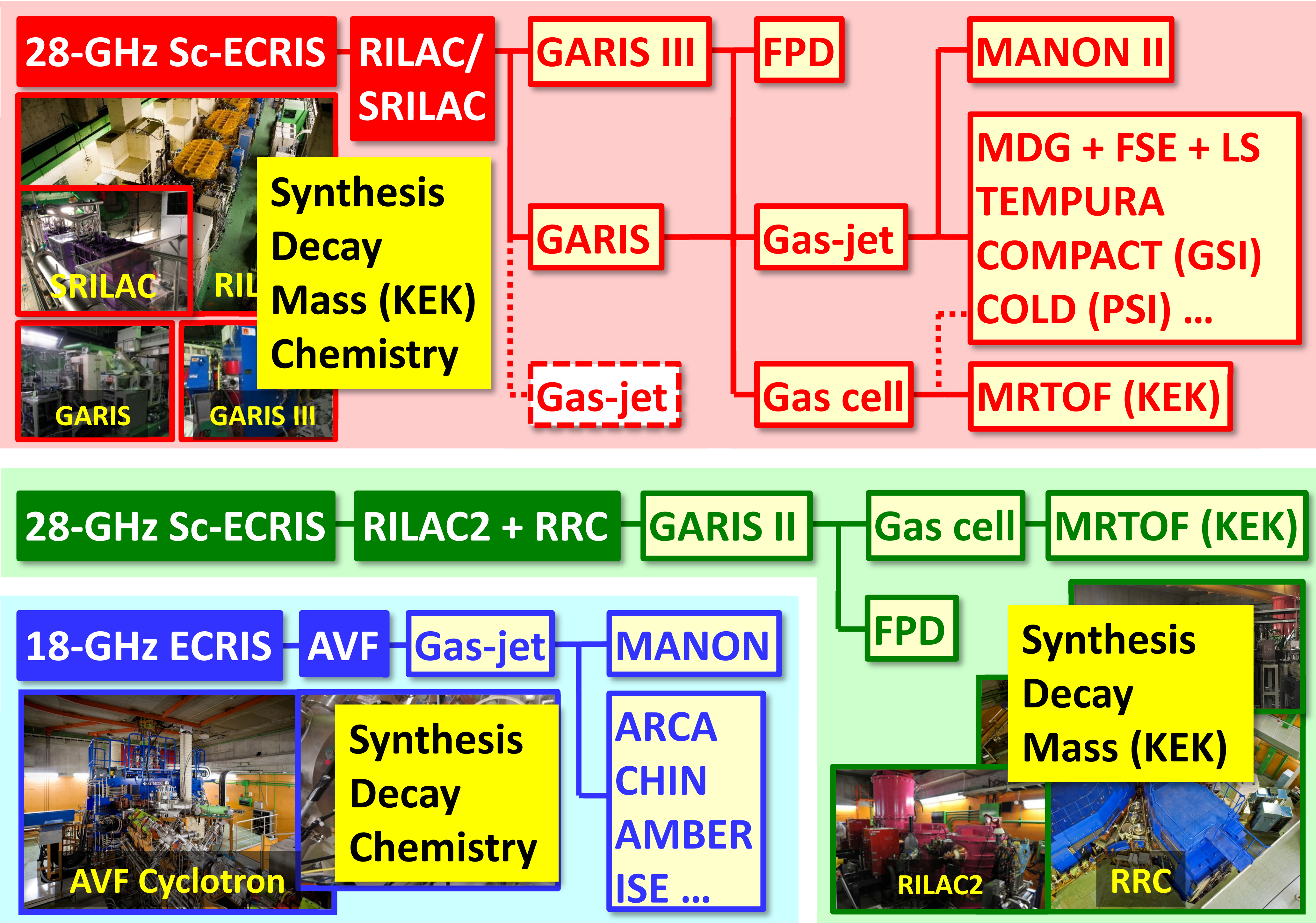


KISS
(KEK Isotope Separation System)
for multi-nucleon transfer reactions

Facilities for SHE research in RIKEN RIBF

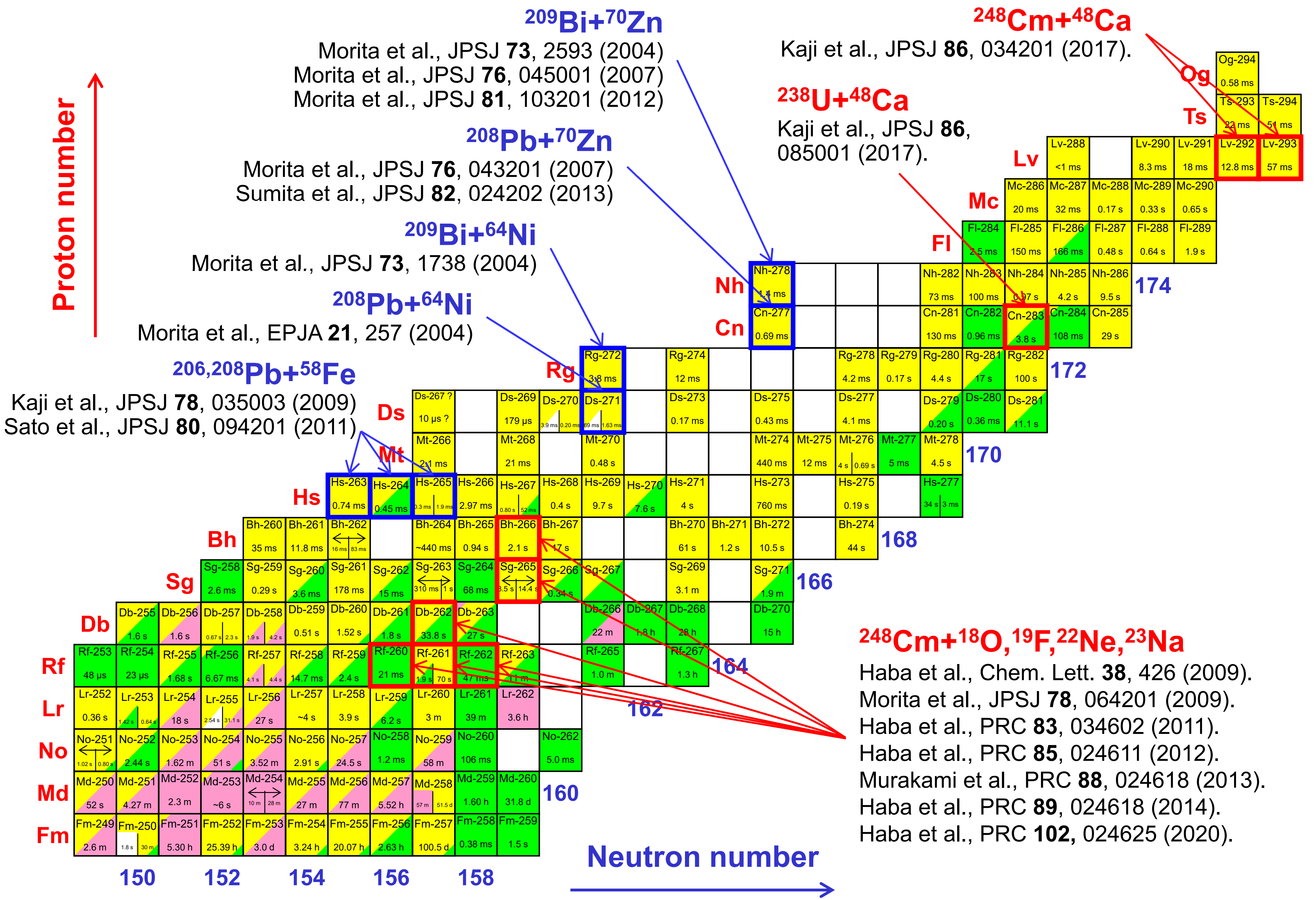


Facilities for SHE research in RIKEN RIBF

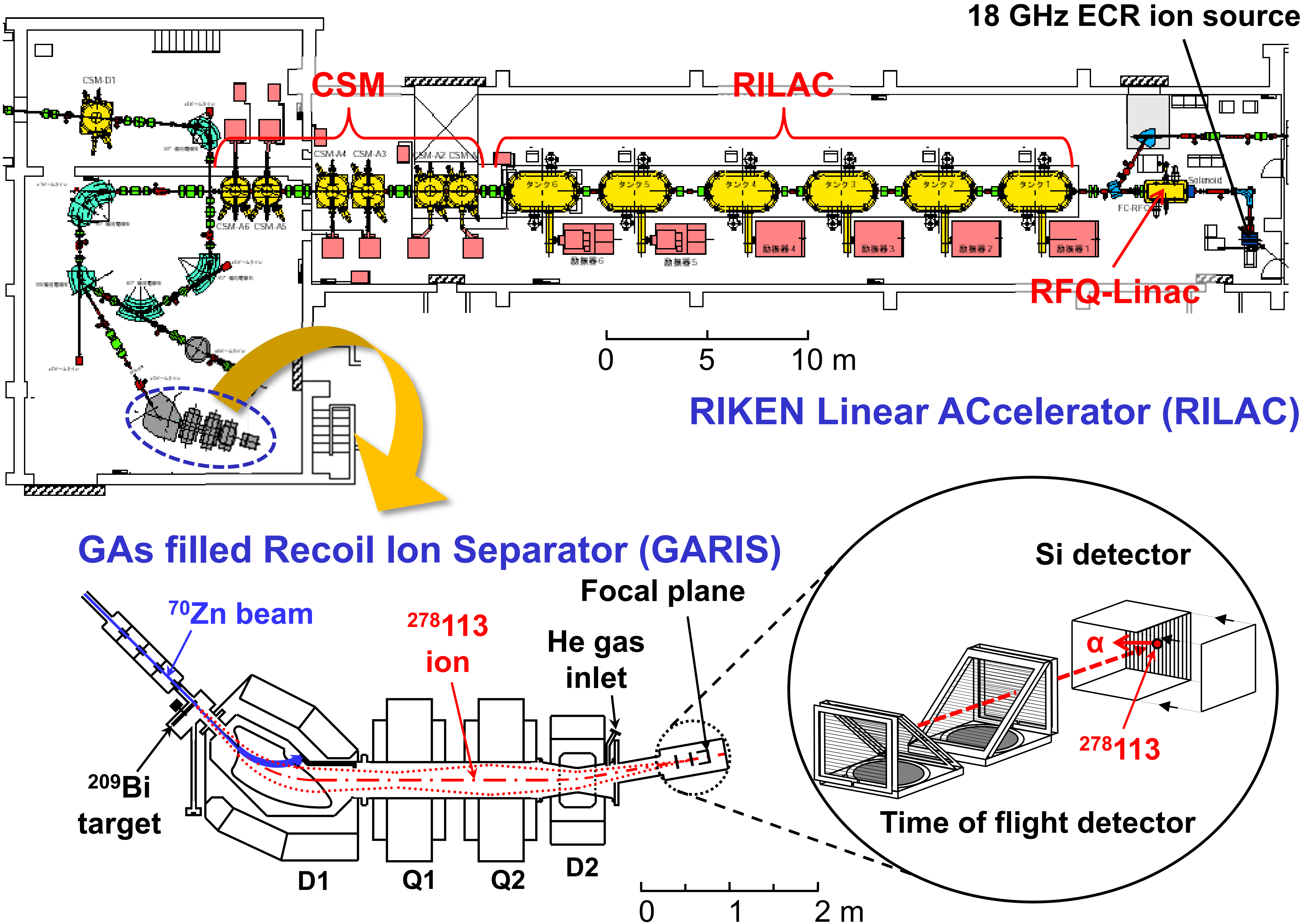


2. Synthesis of element 113
by cold fusion

Synthesis of superheavy elements at RIKEN since 2001

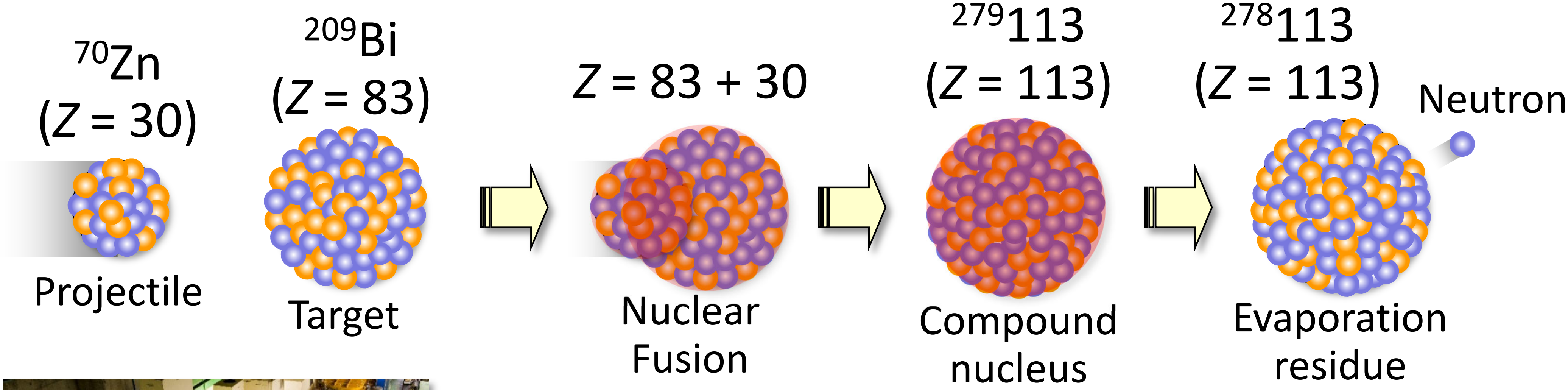


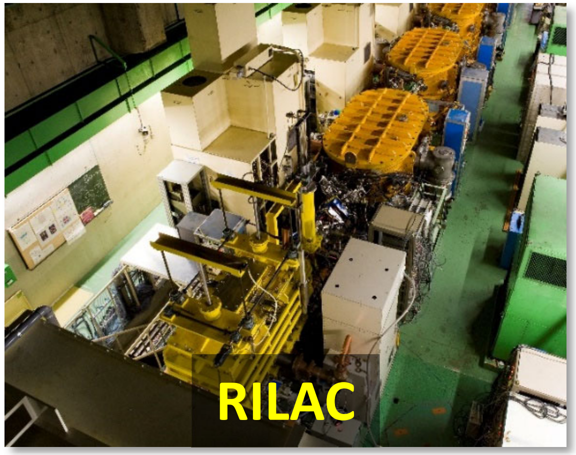
Experimental setup for synthesis of element 113



Cold fusion reaction to produce element 113

^{209}Bi(^{70}Zn,*n*)^{278}113

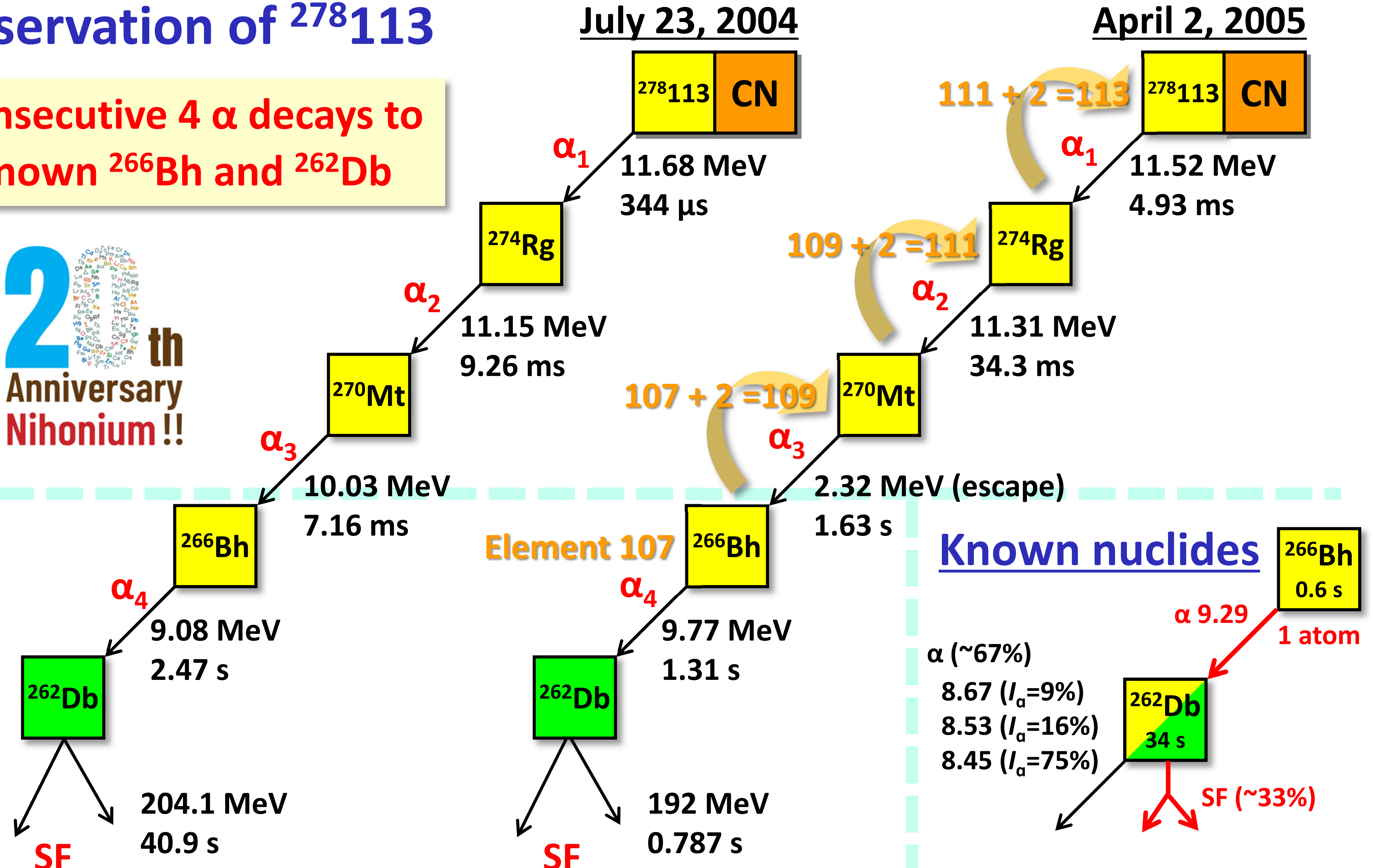




Period	Sept. 5, 2003 – Aug. 18, 2012
Irradiation time	13274 hours (553 days)
Experimenters	43
Beam energy	348 MeV in the middle of the target
Beam intensity	0.47 pμA (2.8 x 10^{12} s^{-1})
Beam integral	1.35 x 10^{20} (15 mg)
Target thickness	0.45 mg cm^{-2} (1.3 x 10^{18} cm^{-2})
GARIS eff.	80%
PSD + SSD eff.	94%

Observation of ^{278}113

Consecutive 4 α decays to known ^{266}Bh and ^{262}Db



Morita et al., JPSJ **73**, 2593 (2004).

Morita et al., JPSJ **76**, 045001 (2007).

Wilk et al., PRL **85**, 2697 (2000).
Table of Isotopes, 8th ed. (1996).

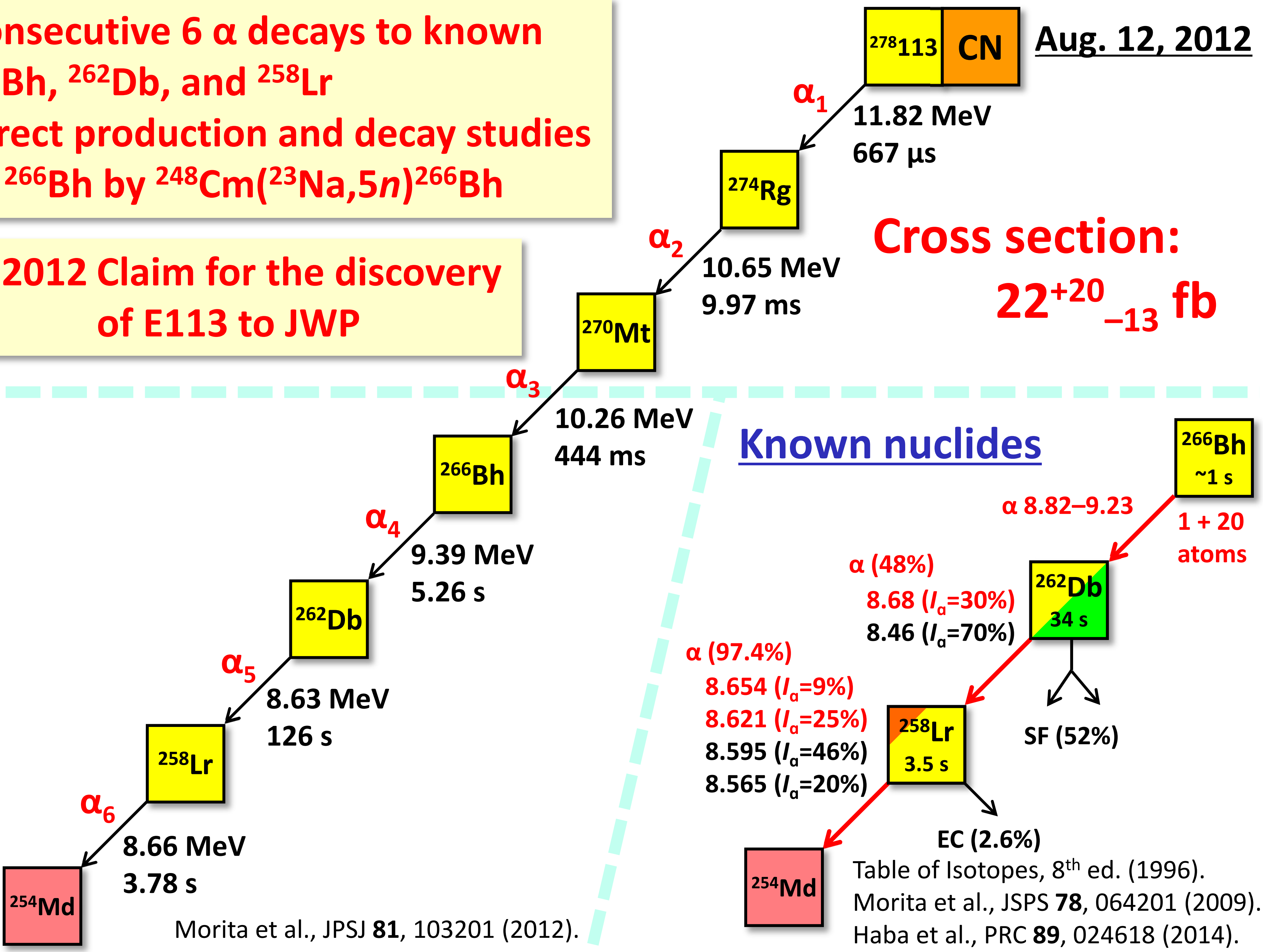
2006 Claim for the discovery of ^{278}113 to JWP of IUPAC and IUPAP
2011 Not approved due to few data on ^{278}113 and ^{266}Bh

Observation of the 3rd event of ^{278}113

- Consecutive 6 α decays to known ^{266}Bh, ^{262}Db, and ^{258}Lr
- Direct production and decay studies of ^{266}Bh by ^{248}Cm(^{23}Na,5n)^{266}Bh

➡ 2012 Claim for the discovery of E113 to JWP

Aug. 12, 2012



Cross section: 22^{+20}_{-13} fb

Morita et al., JPSJ **81**, 103201 (2012).

Known nuclides

Table of Isotopes, 8th ed. (1996).
Morita et al., JSPS **78**, 064201 (2009).
Haba et al., PRC **89**, 024618 (2014).

IUPAC

International Union of Pure and Applied Chemistry

Highlights

- The IUPAC Network
- Periodic Table of the Elements

< ICSU publications on climate change

30 Dec 2015 23:50 Age: 2 days
Category: Press Releases

Discovery and Assignment of Elements with Atomic Numbers 113, 115, 117 and 118

IUPAC announces the verification of the discoveries of four new chemical elements: The 7th period of the periodic table of elements is complete.

The fourth IUPAC/IUPAP Joint Working Party (JWP) on the priority of claims to the discovery of new elements has reviewed the relevant literature for elements 113, 115, 117, and 118 and has determined that the claims for discovery of these elements have been fulfilled, in accordance with the criteria for the discovery of elements of the IUPAP/IUPAC Transfermium Working Group (TWG) 1991 discovery criteria. These elements complete the 7th row of the periodic table of the elements, and the discoverers from Japan, Russia and the USA will now be invited to suggest permanent names and symbols. The new elements and assigned priorities of discovery are as follows:

Element 113 (temporary working name and symbol: ununtrium, Uut)
The RIKEN collaboration team in Japan have fulfilled the criteria for element Z=113 and will be invited to propose a permanent name and symbol.

http://www.iupac.org/

Nihonium – *The first element discovered in Asian countries* –

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For Immediate Release 30 November 2016

IUPAC Announces the Names of the Elements 113, 115, 117, and 118

Elements 113, 115, 117, and 118 are now formally named nihonium (Nh), moscovium (Mc), tennessine (Ts), and oganesson (Og)

Research Triangle Park, NC (USA): On 28 November 2016, the International Union of Pure and Applied Chemistry (IUPAC) approved the names and symbols for four elements: nihonium (Nh), moscovium (Mc), tennessine (Ts), and oganesson (Og), respectively for element 113, 115, 117, and 118.

Atomic number	Element name	Element symbol
113	nihonium	Nh
115	moscovium	Mc
117	tennessine	Ts
118	oganesson	Og

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	1 H																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	57-71 *	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	89-103 †	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og

*Lanthanoide	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
†Actinoide	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

3. Search for element 119 in the $^{248}Cm(^{51}V,xn)^{299-x}119$ reaction

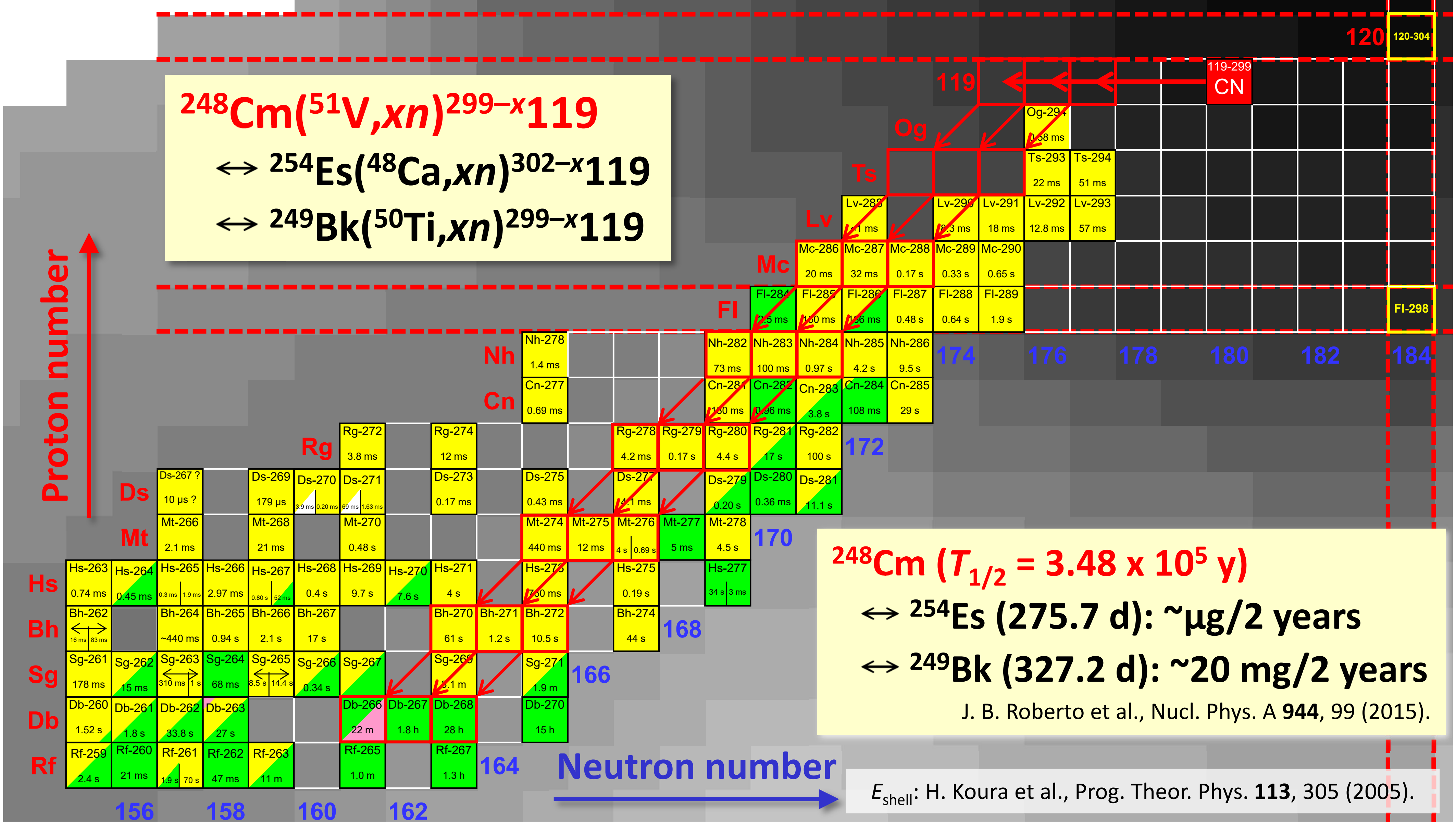
Search for element 119 at RIBF

RIKEN – ORNL – UTK - Kyushu Univ. – Niigata Univ. – Saitama Univ. – Osaka Univ. – Tohoku Univ. – JAEA – Yamagata Univ. – IPHC – IMP – ANU – NCBJ Collaboration

^{248}Cm(^{51}V,*xn*)$^{299-x}$119

↔ ^{254}Es(^{48}Ca,*xn*)$^{302-x}$119

↔ ^{249}Bk(^{50}Ti,*xn*)$^{299-x}$119



^{248}Cm ($T_{1/2}$ = 3.48 x 10^5 y)

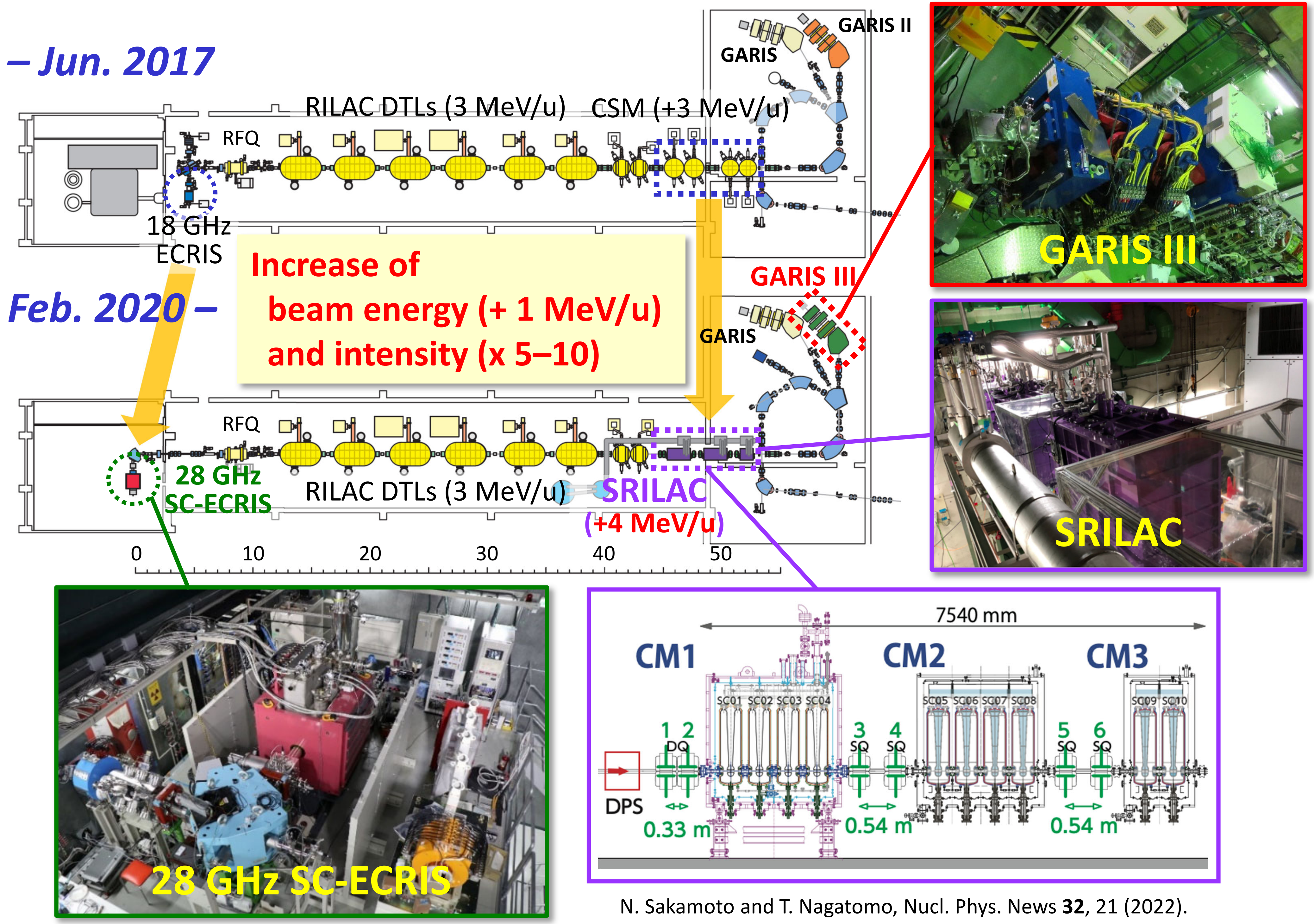
↔ ^{254}Es (275.7 d): ~µg/2 years

↔ ^{249}Bk (327.2 d): ~20 mg/2 years

J. B. Roberto et al., Nucl. Phys. A **944**, 99 (2015).

E_{shell}: H. Koura et al., Prog. Theor. Phys. **113**, 305 (2005).

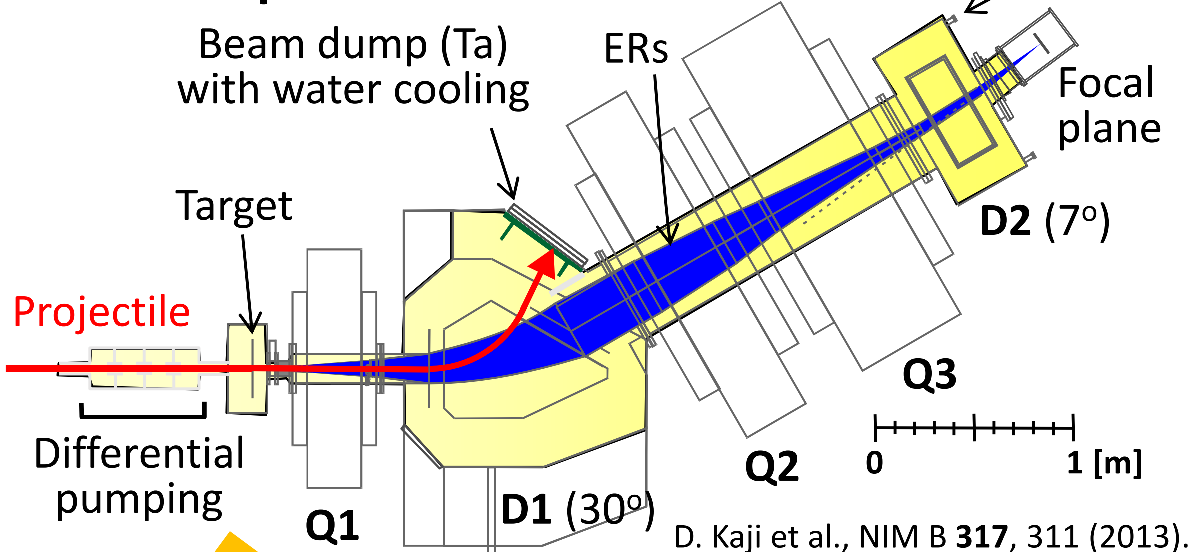
Upgrade of RILAC (June 2017–February 2020)



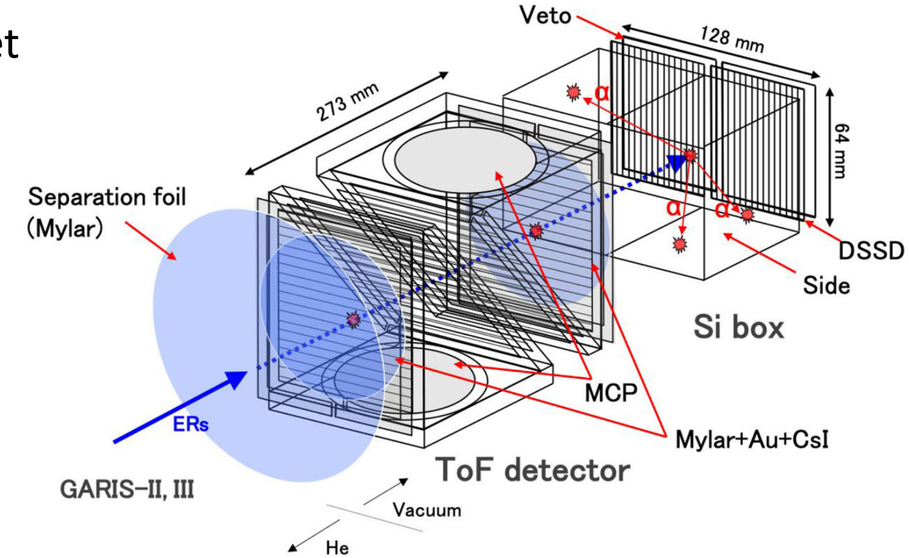
N. Sakamoto and T. Nagatomo, Nucl. Phys. News **32**, 21 (2022).

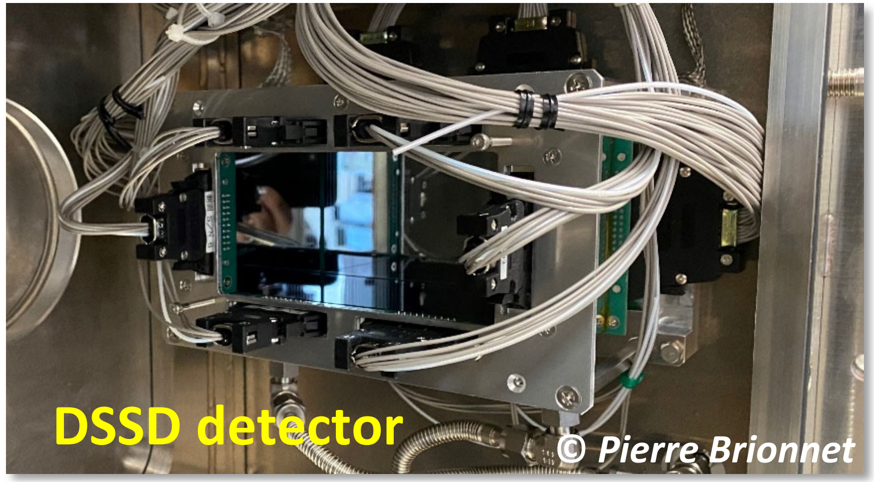
Gas-filled recoil ion separator, GARIS III

Same specifications as GARIS II

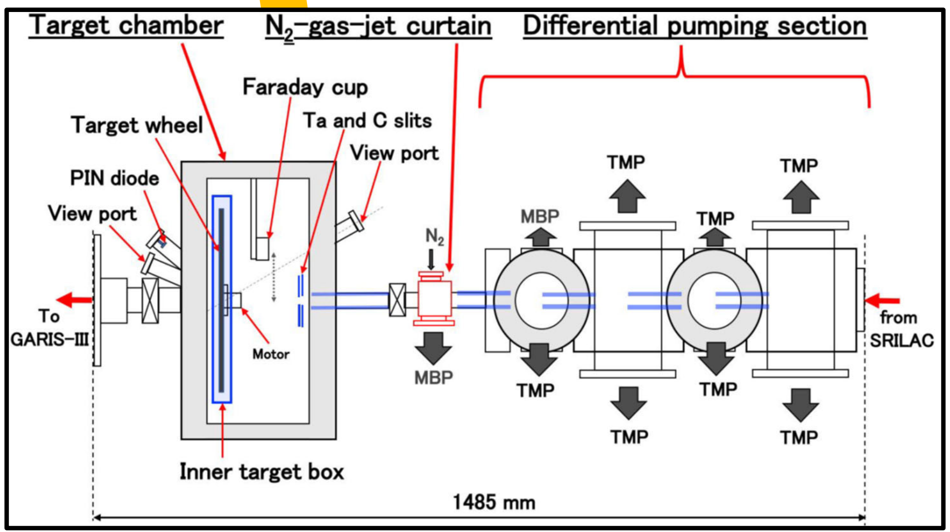


D. Kaji et al., NIM B **317**, 311 (2013).





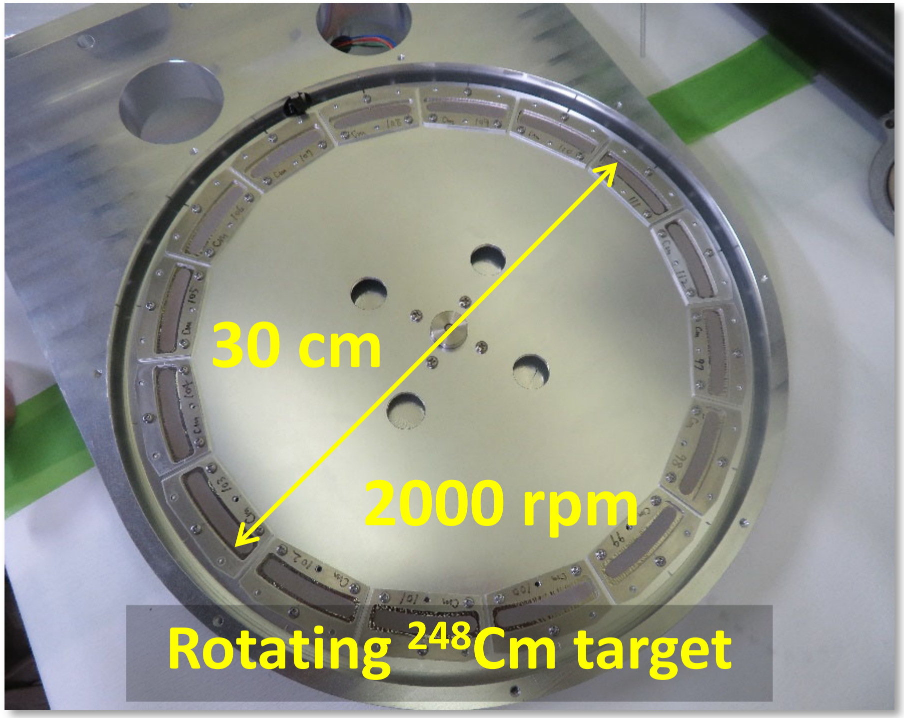
	GARIS	GARIS II&III
Δθ (mrad)	±67	±55
Δϕ (mrad)	±58	±120
ΔΩ (msr)	≈12	≈20

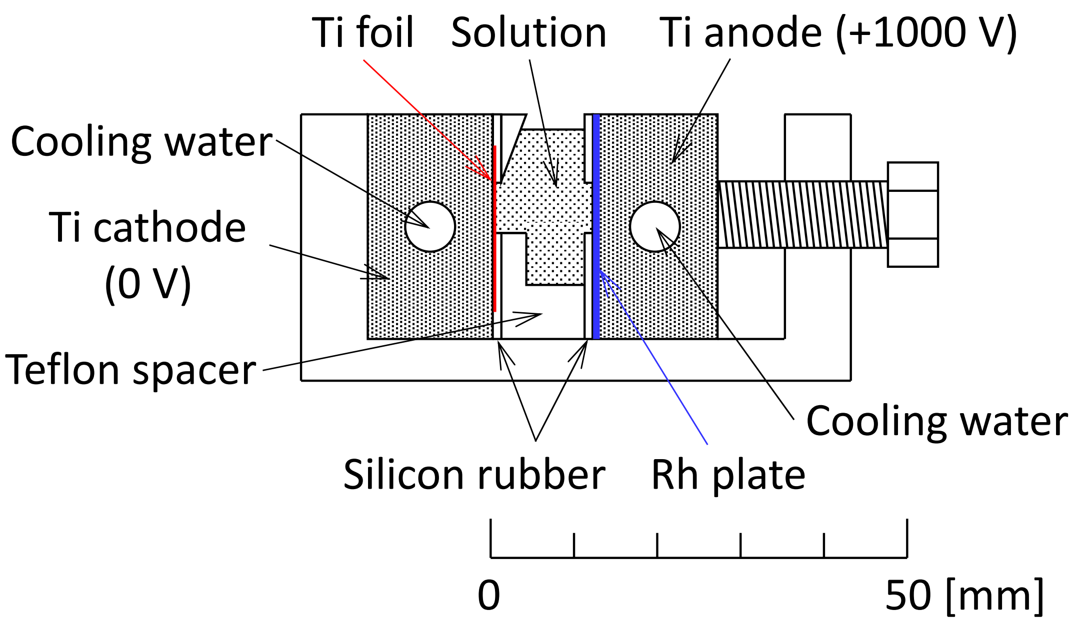


Hamamatsu Photonics K. K.

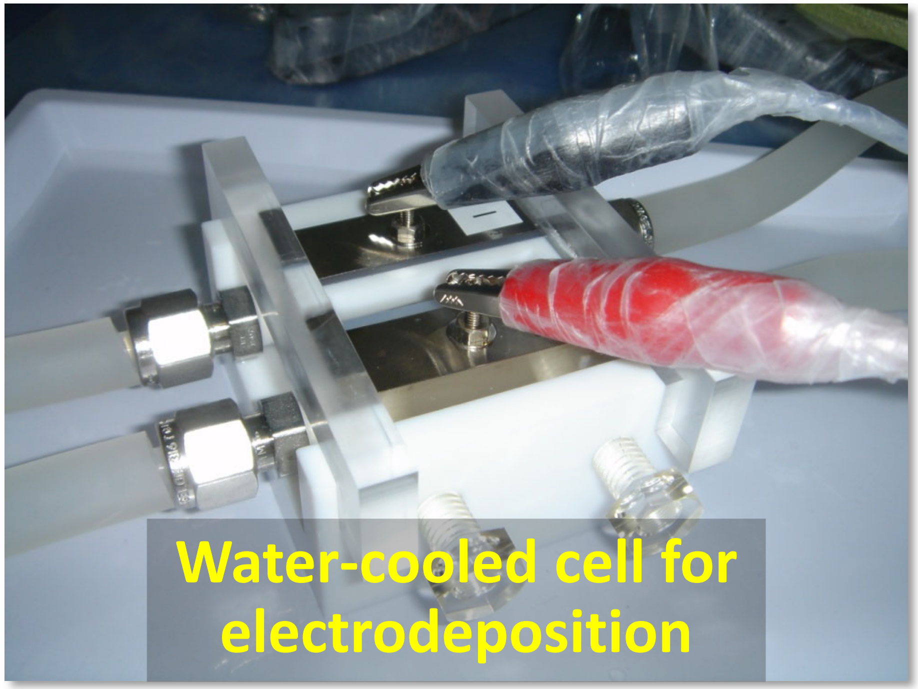
- DSSD: 320 μm, 60 x 123 mm^2, 64 x 64 strips
  pixel size: ~1 x 2 mm^2
- Side detector: 320 μm
  Small: 60 x 60 mm^2; Large: 60 x 123 mm^2
- Veto detector: 650 μm, 60 x 123 mm^2 (4 pads)

^{248}Cm target





Y. Kudou et al., RIKEN Accel. Prog. Rep. **42**, 265 (2009).



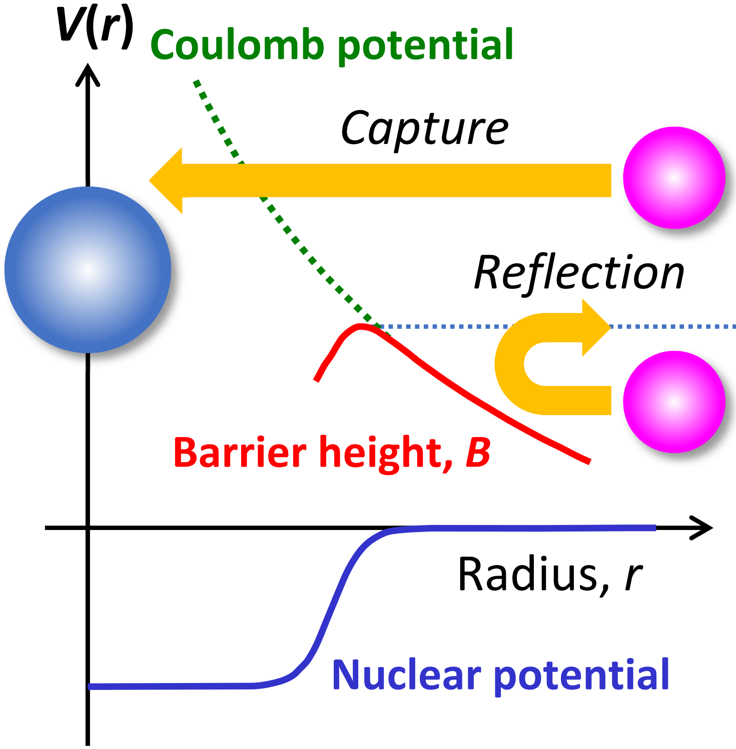
What is the optimal reaction energy ?

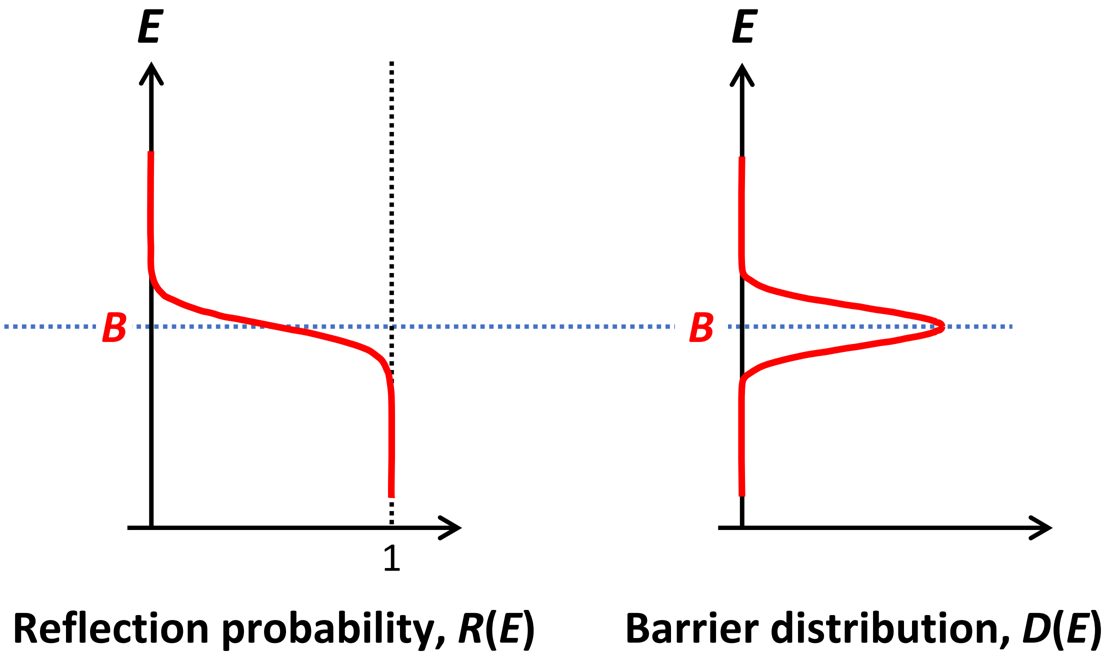
$$\sigma_{ER} = \sigma_{cap} \times P_{CN} \times P_{surv}$$

Capture — *CN formation* — *Survival of ER*

Measurement of excitation function of quasielastic backscattering to Rutherford scattering ($d\sigma_{QE}/d\sigma_{Ruth}$) by detecting non-captured projectiles

→ Optimal reaction energy for capture process

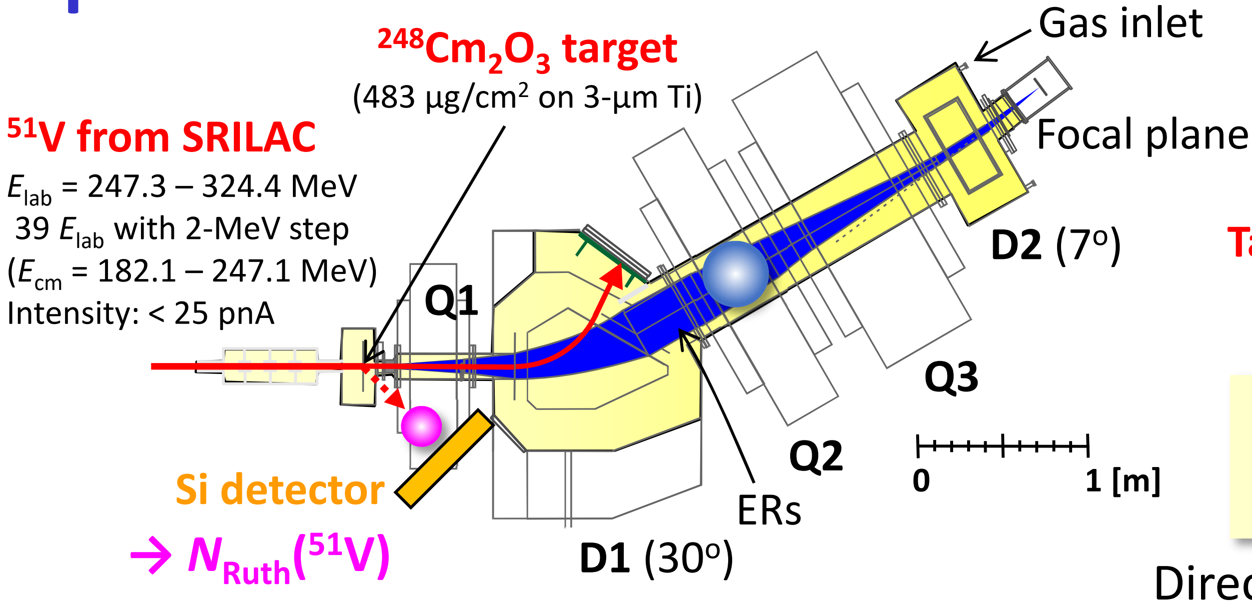


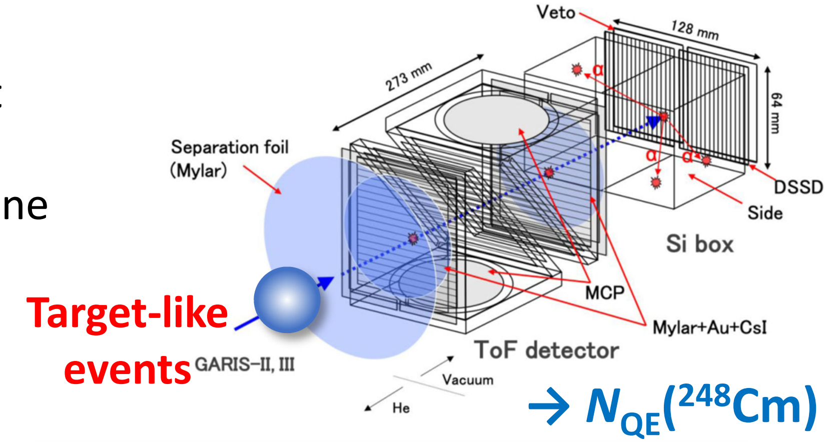


$$R(E) = \frac{d\sigma_{QE}}{d\sigma_{Ruth}}$$

$$D(E) = -\frac{dR(E)}{dE}$$

Experimental method

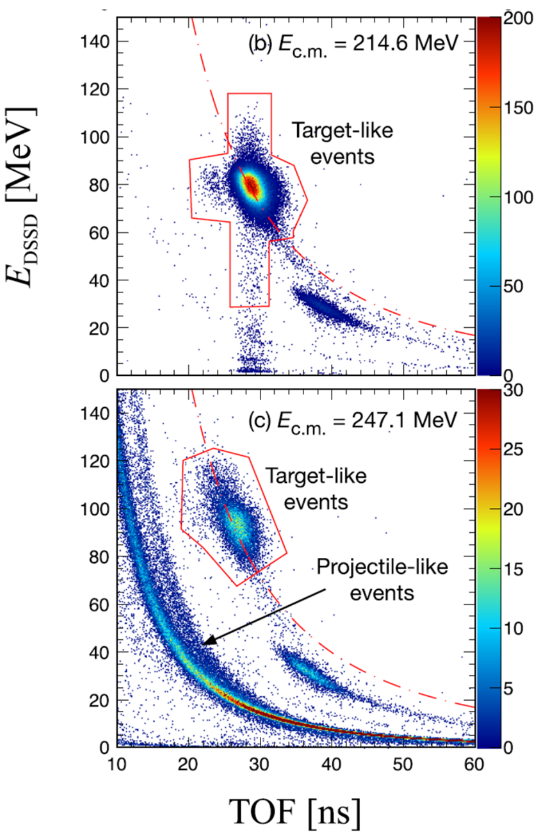


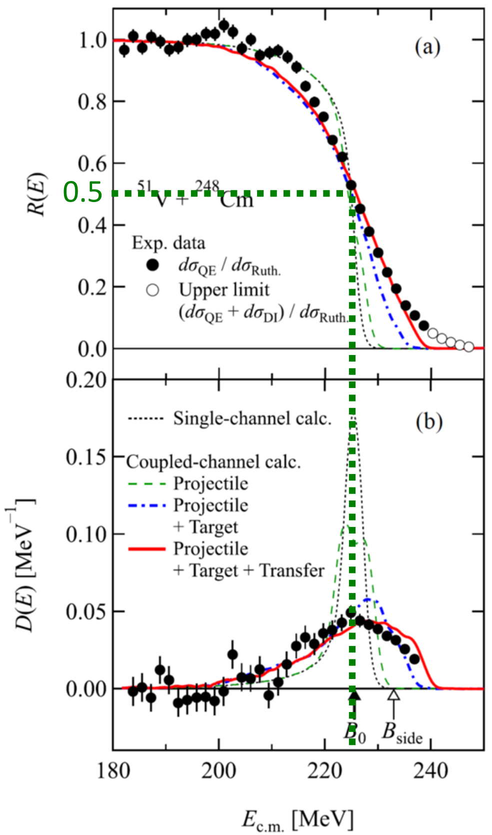


$$R(E) = \frac{d\sigma_{QE}}{d\sigma_{Ruth}} \propto \frac{N_{QE}(^{248}Cm)}{N_{ruth}(^{51}V)}$$

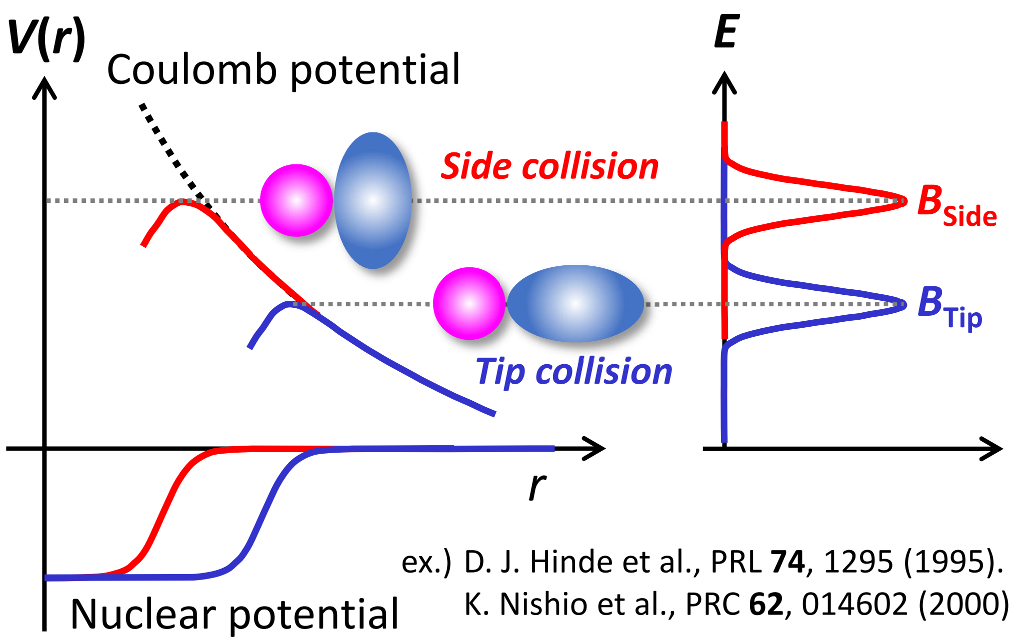
Direct measurement of QE barrier at L ~ 0

T. Tanaka et al., J. Phys. Soc. Jpn. **87**, 014201 (2018).
T. Tanaka et al., Phys. Rev. Lett. **124**, 052502 (2020).
M. Tanaka et al., J. Phys. Soc. Jpn. **91**, 084201 (2022).

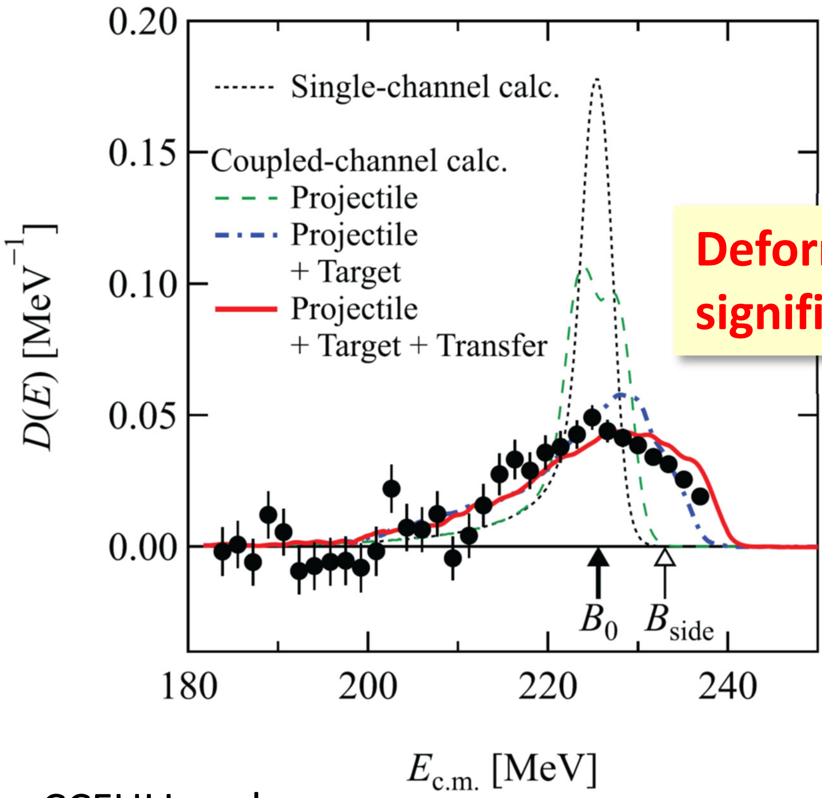




Favorable side collision in hot fusion



ex.) D. J. Hinde et al., PRL **74**, 1295 (1995).
K. Nishio et al., PRC **62**, 014602 (2000).



Deformed ^{248}Cm target significantly affects $D(E)$.

CCFULL code:

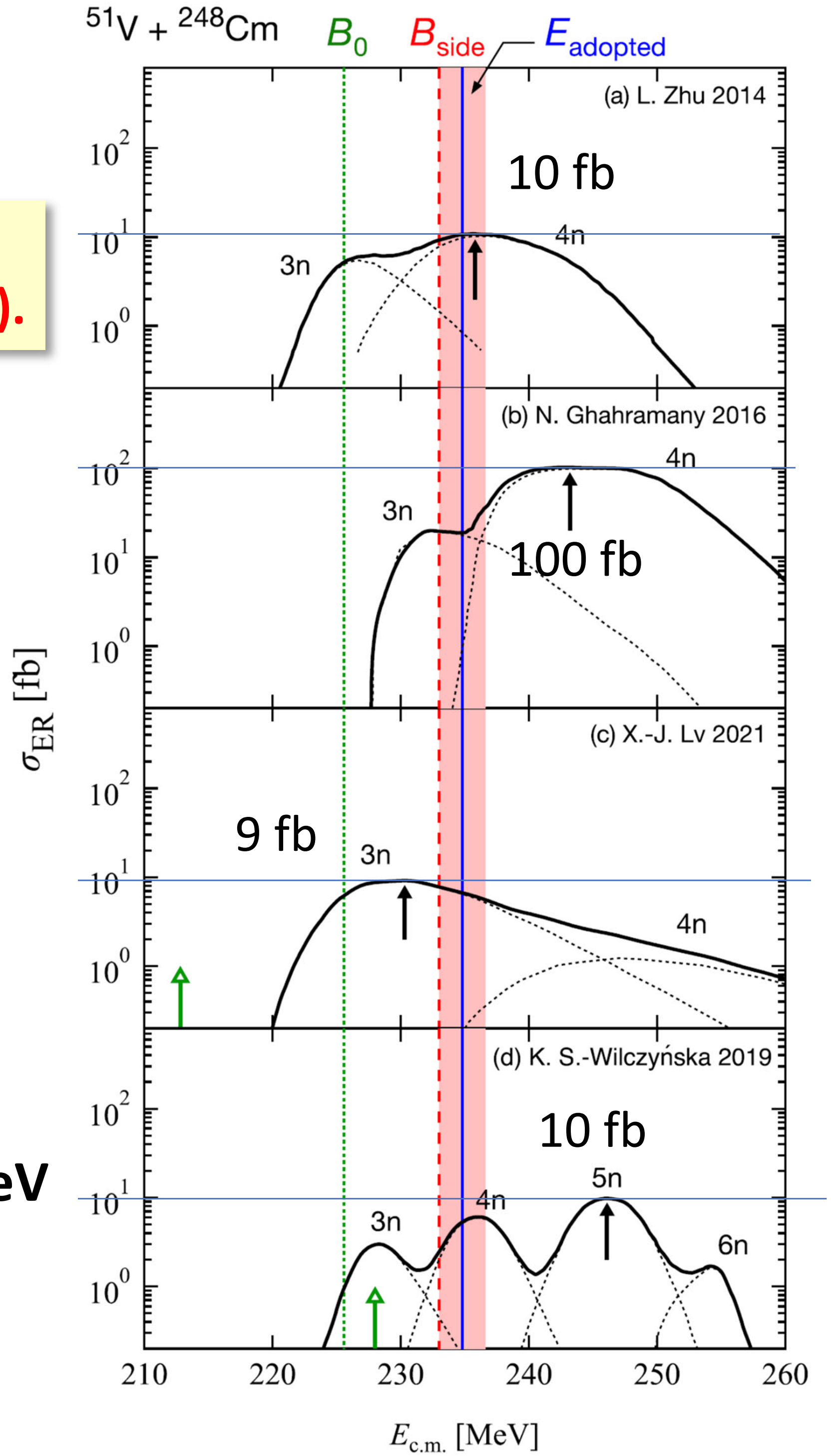
K. Hagino et al., Comput. Phys. Commun. **123**, 143 (1999).

- Average barrier height: B_0 = **225.6(2) MeV**
- Side-collision energy: B_{side} = **233.0(2) MeV** from optical potential $V(r,\vartheta)$ in CC calculation
- Uncertainty between E_{opt} & B_{side}: ΔE_{opt} = **+3.5 MeV** from the ^{48}Ca + ^{248}Cm system (+1.5% for B_{side})

T. Tanaka et al., PRL **124**, 052502 (2020).

→ Adopted energy for the ^{51}V + ^{248}Cm system

$E_{adopted} = B_{side} + 0.5 \times \Delta E_{opt}$ = **234.8 MeV**



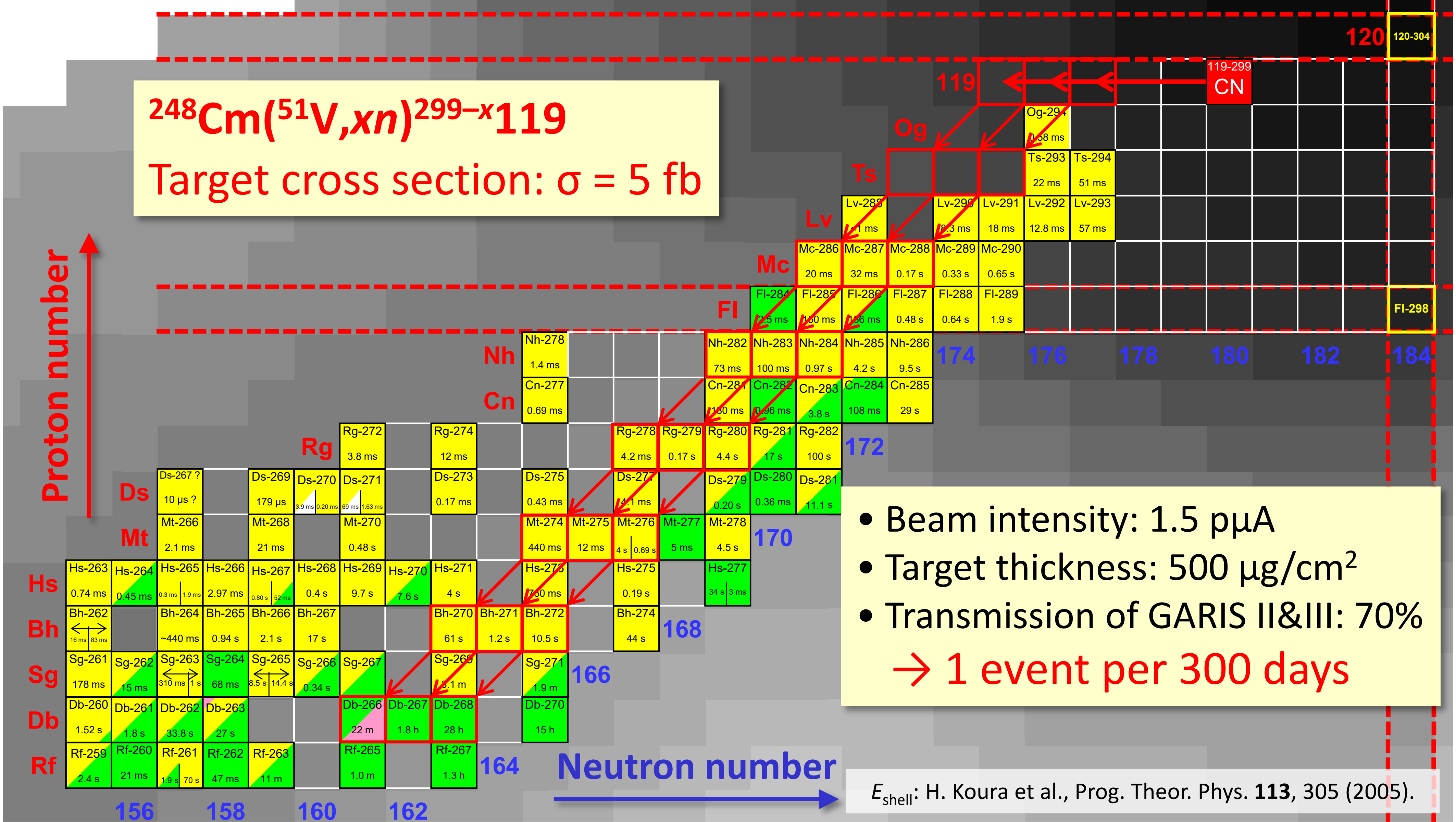
M. Tanaka et al., JPSJ **91**, 084201 (2022).

Search for element 119 at RIBF

RIKEN – ORNL – UTK - Kyushu Univ. – Niigata Univ. – Saitama Univ. – Osaka Univ. – Tohoku Univ. – JAEA – Yamagata Univ. – IPHC – IMP – ANU – NCBJ Collaboration

$^{248}Cm(^{51}V,xn)^{299-x}119$

Target cross section: σ = 5 fb



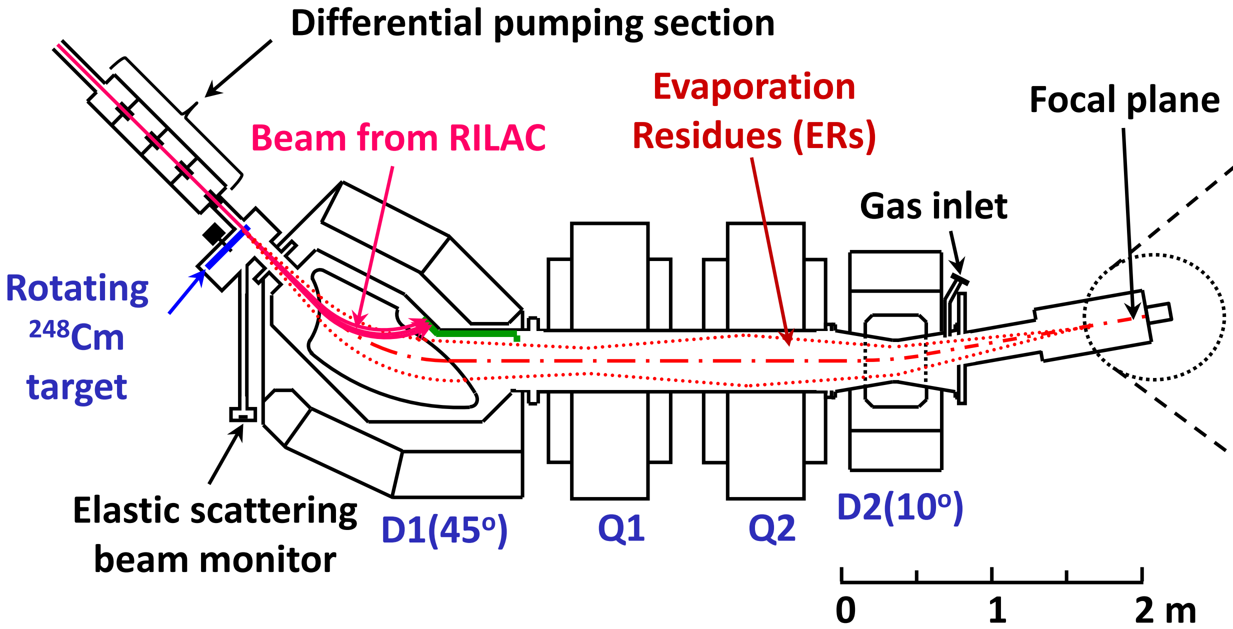
- Beam intensity: 1.5 pμA
- Target thickness: 500 μg/cm²
- Transmission of GARIS II&III: 70%

→ 1 event per 300 days

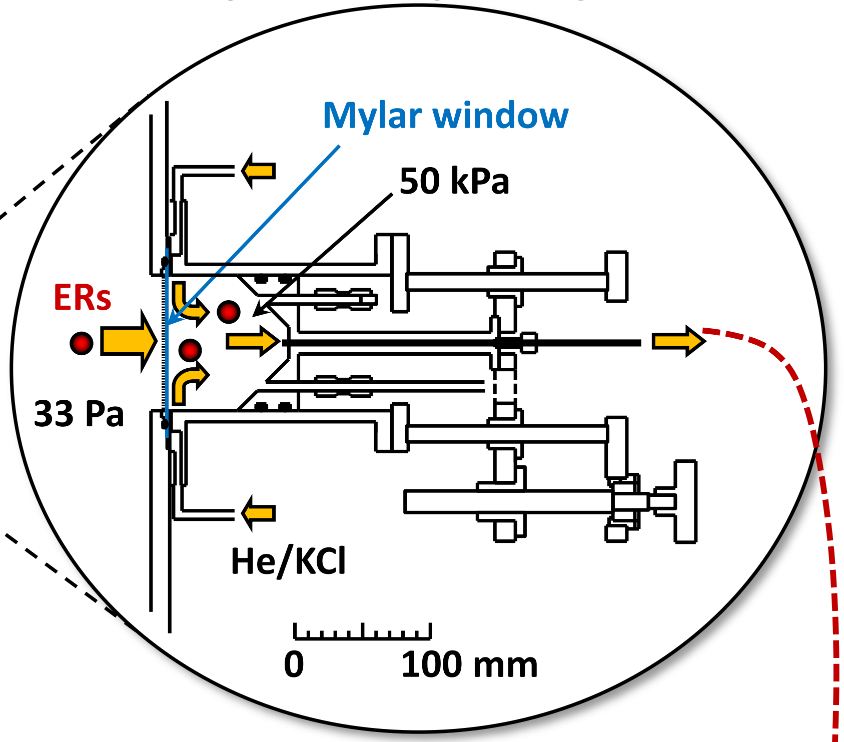
4. Production and decay studies of ^{261}Rf, ^{262}Db, ^{265}Sg, and ^{266}Bh

GARIS + gas-jet + MANON

RIKEN GARIS



Gas-jet transport system

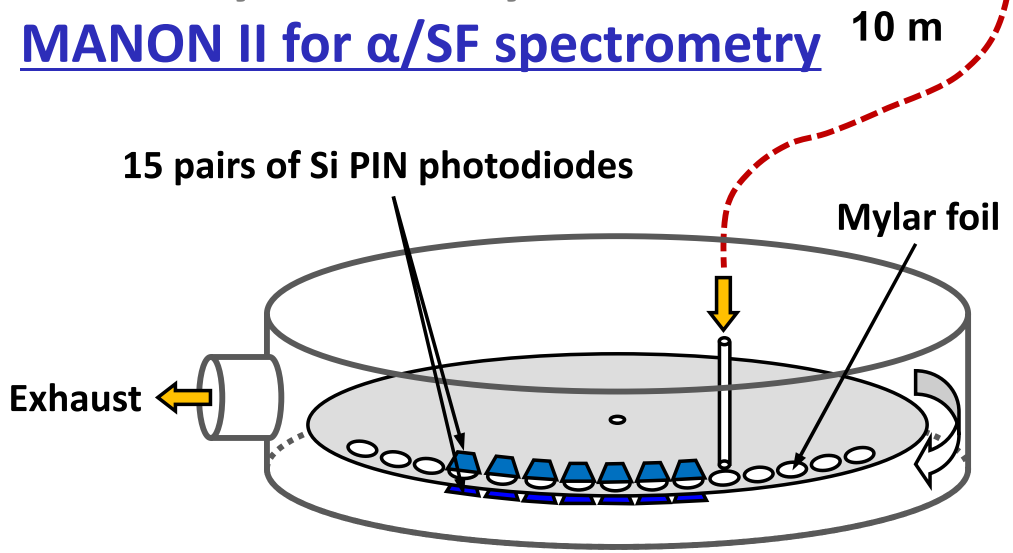


Breakthroughs in SHE chemistry

- **Chemistry experiments under low background radiation**
- **Stable and high gas-jet transport yield**
- **New chemical reactions**

Chemistry laboratory

MANON II for α/SF spectrometry



Production and decay studies of ^{261}Rf, ^{262}Db, ^{265}Sg, and ^{266}Bh

Nuclide	^{261}Rf^{a,b} (Z=104)	^{262,263}Db (Z=105)	^{265}Sg^{a,b} (Z=106)	^{266,267}Bh (Z=107)
Half-life	68, 3 s[1)]	34 s, 27 s[2)]	8.9, 16.2 s[1)]	1.7 s, 17 s[4)]
Reaction	^{248}Cm(^{18}O,5n)	^{248}Cm(^{19}F,5;4n)	^{248}Cm(^{22}Ne,5n)	^{248}Cm(^{23}Na,5;4n)
Cross section (nb)	12[3)], ?	1.5[3)], ?	0.2–0.3[1)] ?	0.05[5)] ?
Beam energy (MeV)	95	103, 97.4	118	135, 131, 126, 121
Beam intensity (pμA)	7	4	3	3
$^{248}Cm_2O_3$ target (μg/cm^2)	280, 230	230, 290, 330	230, 280	290, 260, 270
Magnetic rigidity (Tm)	1.58–2.16	1.73–2.09	1.73–2.16	2.12
GARIS He (Pa)	33	32	33	33
GARIS transmission (%)	7.8±1.7	8.1±2.2	13	15
RTC Mylar window (μm)	0.5	0.5	0.7	0.7
Honeycomb grid (%)	78/84	84	72/84	78
Gas-jet He (kPa)	49	47	49	80
Chamber depth (mm)	20	20	40	20
He flow rate (L/min)	2.0	2.0	2.0	5.0
KCl generator (^{o}C)	620	620	600/605	620
MANON step interval (s)	30.5, 2.0	15.5	20.5, 10.5	5.0, 8.5, 15.0

1) Düllmann and Türler, PRC **77**, 064320 (2008). 2) Firestone and Shirley, *Table of Isotopes*, 8th ed. (Wiley, New York, 1996).
3) Nagame et al., JNRS **3**, 85 (2002). 4) Wilk et al., PRL **85**, 2697 (2000). 5) Morita et al., JSPS **78**, 064201 (2009).

Production and decay studies of ^{266,267}Bh

^{22}Na beam energy (MeV)	Thickness of $^{248}Cm_2O_3$ target (μg/cm^2)	Beam integral (×10^{18})	MANON Step interval (s)
121	257	10.20	8.5
126	256	9.26	8.5
131	290	4.96	5.0
	290	3.99	15.0
	257	8.90	8.5
	257	9.02	8.5
135	256	11.21	8.5

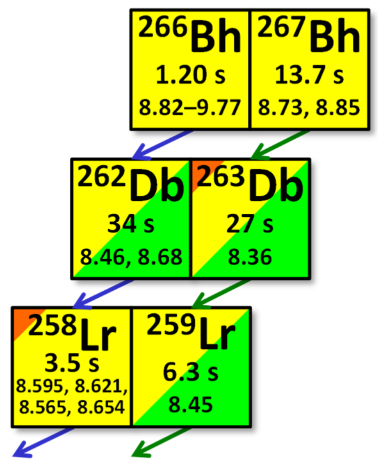
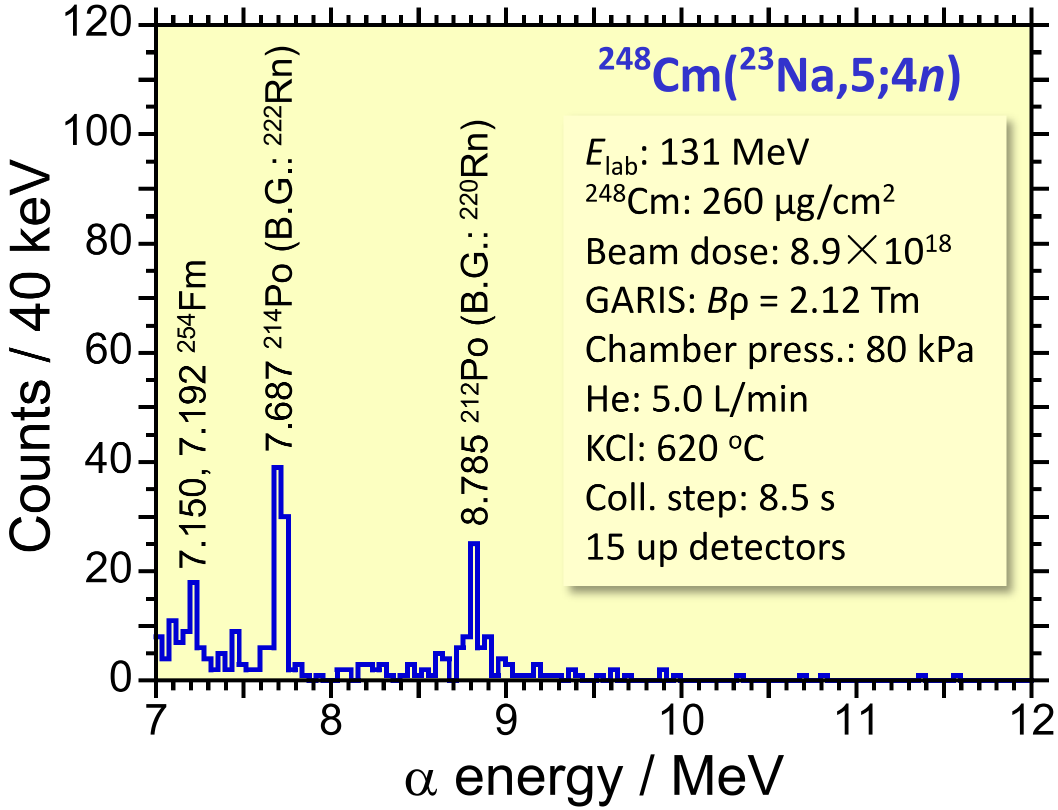


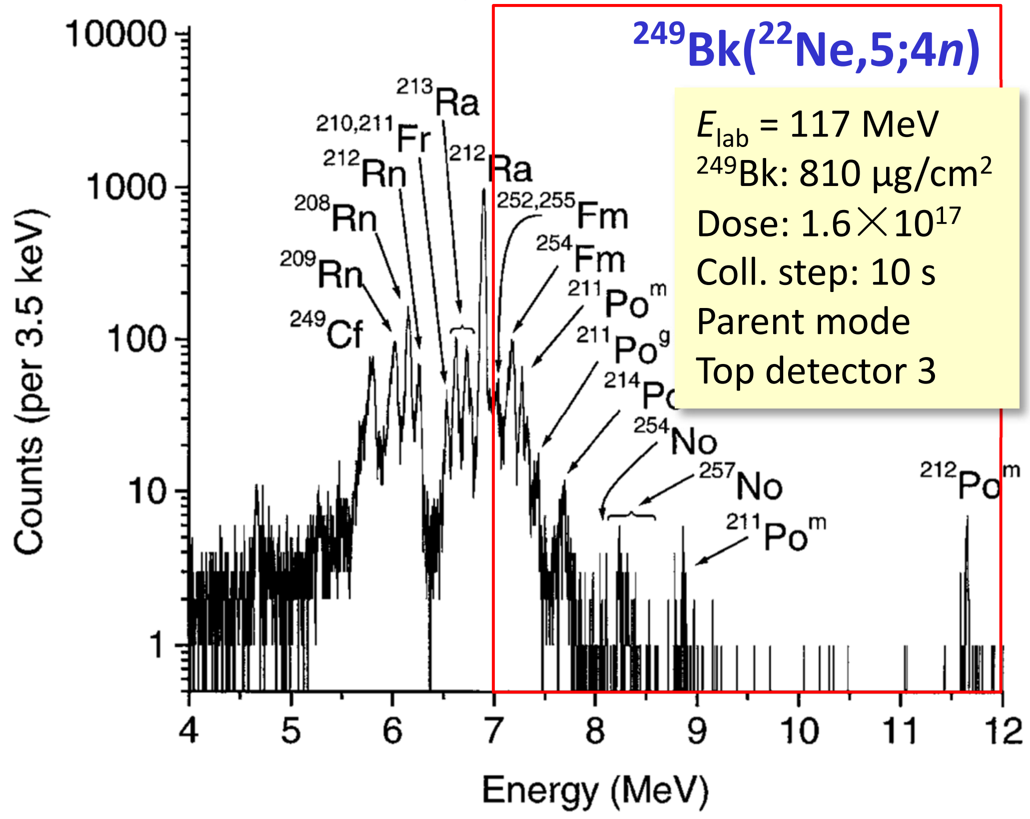
Table of Isotopes, 8th ed. (1996).
Wilk *et al.*, PRL **85**, 2697 (2000).
Eichler *et al.*, Nature **407**, 63 (2000).
Gan *et al.*, EPJA **20**, 385 (2004).
Qin *et al.*, NPR **23**, 400 (2006).
Morita *et al.*, JSPS **78**, 064201 (2009).
Morita *et al.*, JSPS **81**, 103210 (2012).
Haba *et al.*, PRC **89**, 024618 (2014).

GARIS + gas-jet + MANON II



H. Haba et al., Phys. Rev. C **102,** 024625 (2020).

Gas-jet + MG



P. A. Wilk et al., Phys. Rev. Lett. **85**, 2697 (2000).

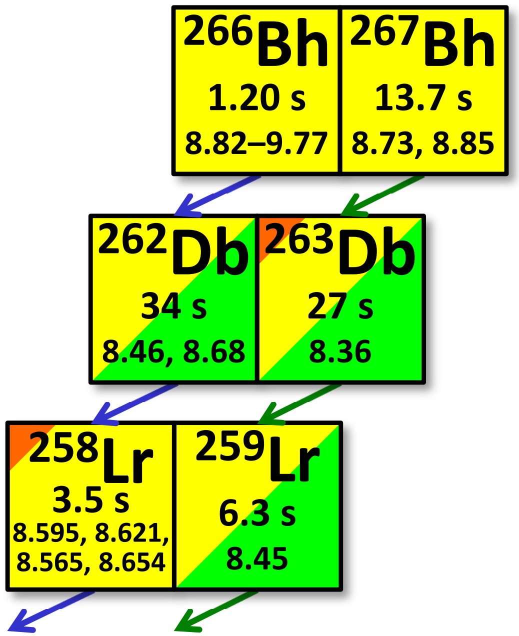
Search for α-α/SF correlations

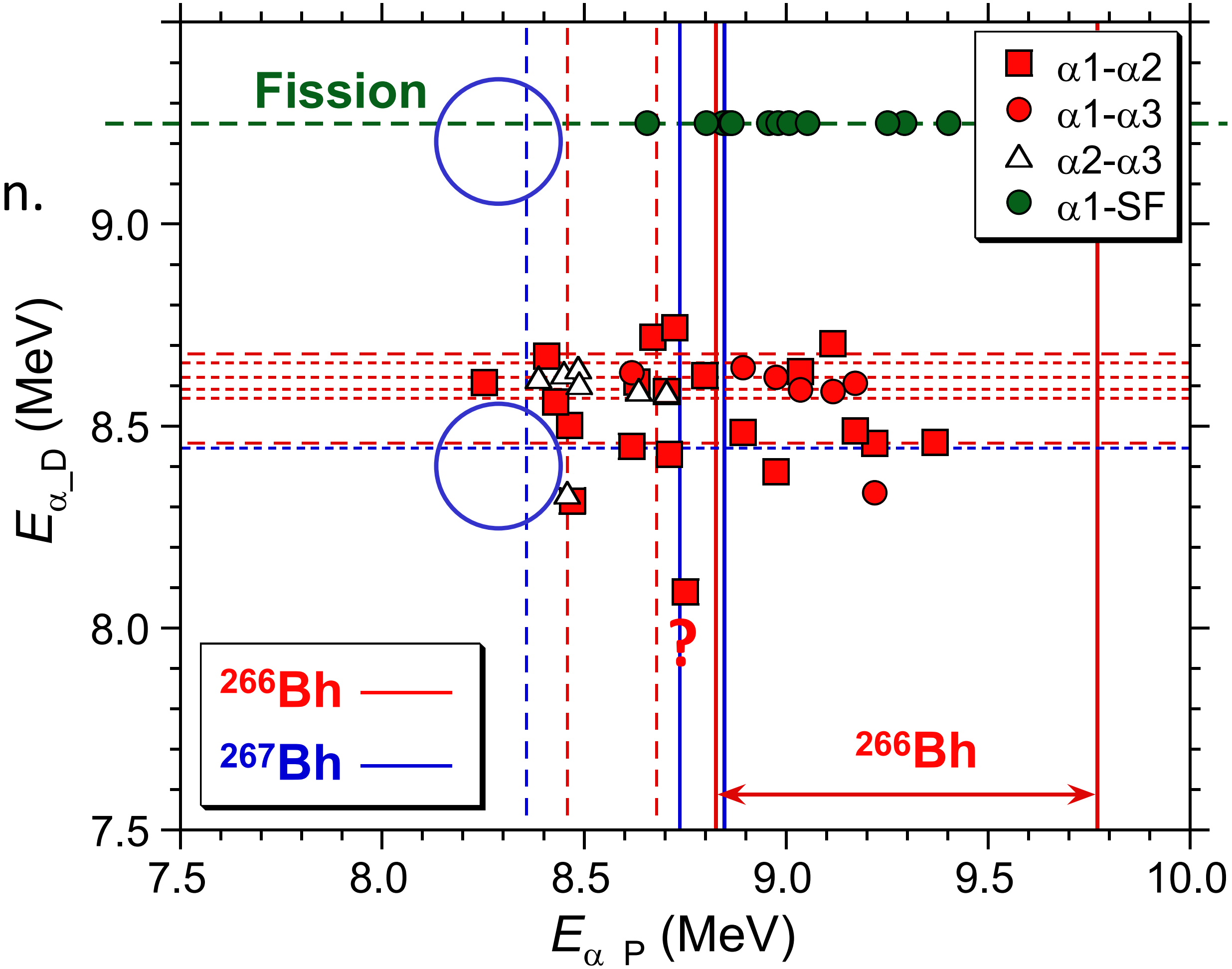
$E_{\alpha 1}$ = 8.00–10.00 MeV

$E_{\alpha 2;\,\alpha 3}$ = 8.00–8.77 MeV

E_{SF} ≥ 20 MeV; Si top & bottom coin.

$\Delta T \leq 340$ s [= 10 $T_{1/2}(^{262}\mathrm{Db})$]





Energy (MeV)	α-α-α		α-α		α-SF		α-α-SF	
	Obs.	RDM	Obs.	RDM	Obs.	RDM	Obs.	RDM
121	**0**	<0.00	**0**	<0.15	**0**	<0.02	**0**	<0.00
126	**0**	<0.00	**1**	<0.16	**3**	<0.03	**0**	<0.00
131	**5**	<0.00	**21**	<1.09	**10**	<0.13	**0**	<0.00
135	**2**	<0.00	**9**	<0.15	**0**	<0.02	**0**	<0.00
Total	**7**	<0.00	**31**	<1.55	**13**	<0.19	**0**	<0.00

Decay properties of ^{266}Bh

- E_α of ^{266}Bh: E_α = 8.62–9.40 MeV.
  ↔ E_α = 8.82–9.77 MeV in Refs.

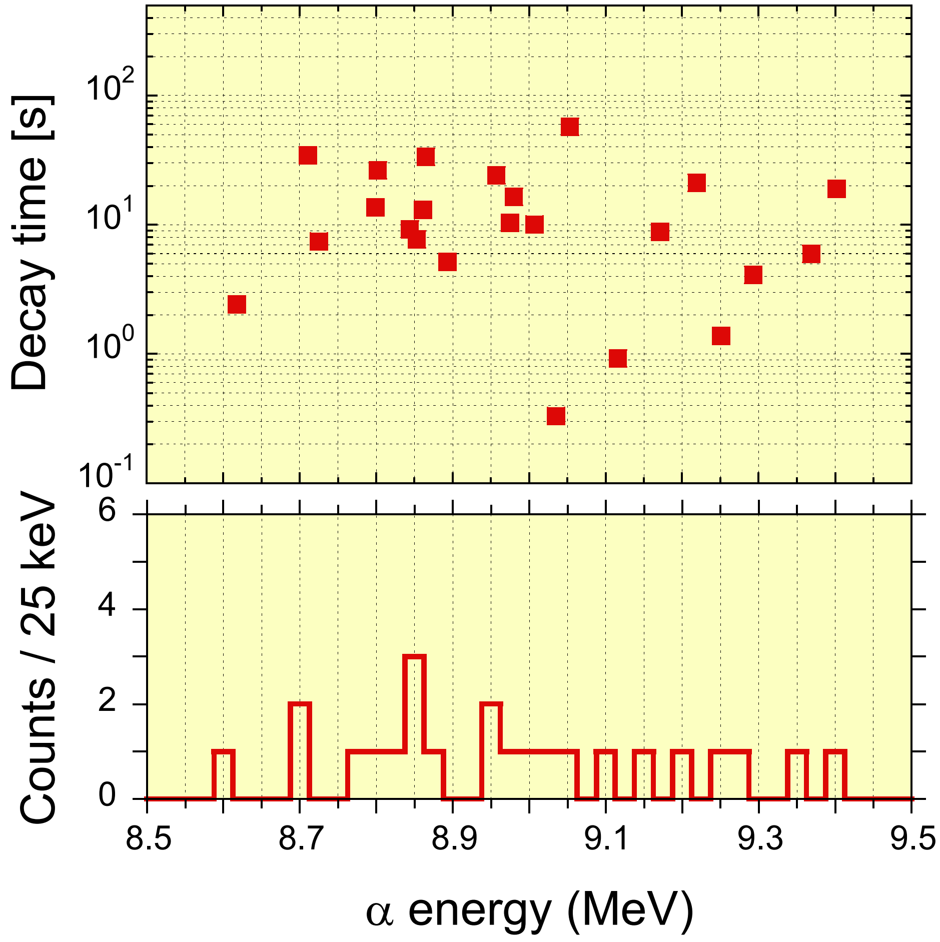
- $T_{1/2}$ = 10.0 s in this work is longer than those of ^{266}Bh in Refs.

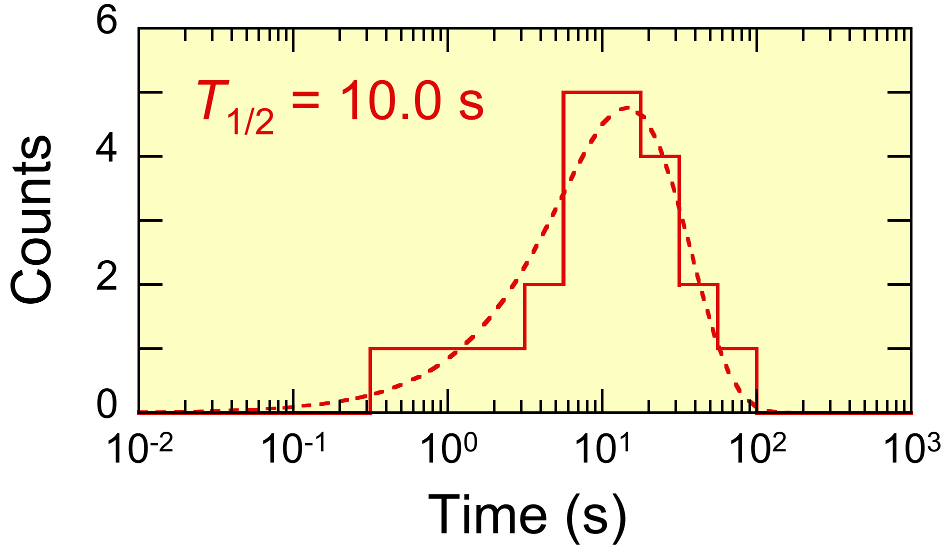
Nuclide	This work		Refs. [1–4]	
	N	$T_{1/2}$ [s]	N	$T_{1/2}$ [s]
^{266}Bh	**23**	$\mathbf{10.0^{+2.6}_{-1.7}}$	**8**	$\mathbf{1.20^{+0.66}_{-0.31}}$
^{267}Bh	**0**	**–**	**11**	$\mathbf{13.7^{+5.9}_{-3.2}}$

[1] ^{249}Bk(^{22}Ne,5;4n)^{266,267}Bh (N = 1, 5): Wilk *et al.*, PRL **85**, 2697 (2000).
[2] ^{249}Bk(^{22}Ne,4n)^{267}Bh (N = 6): Eichler *et al.*, Nature **407**, 63 (2000).
[3] ^{243}Am(^{26}Mg,3n)^{266}Bh (N = 4): Qin *et al.*, Nucl. Phys. Rev. **23**, 400 (2006).
[4] ^{209}Bi(^{70}Zn,n)^{278}113 → ^{266}Bh (N = 3): Morita *et al.*, JPSJ **81**, 103201 (2012).

- Existence of an isomeric state in ^{266}Bh? Miss assignment of ^{266}Bh to ^{267}Bh in the previous experiments?

- The long half-life of ^{266}Bh is good for Bh chemistry.

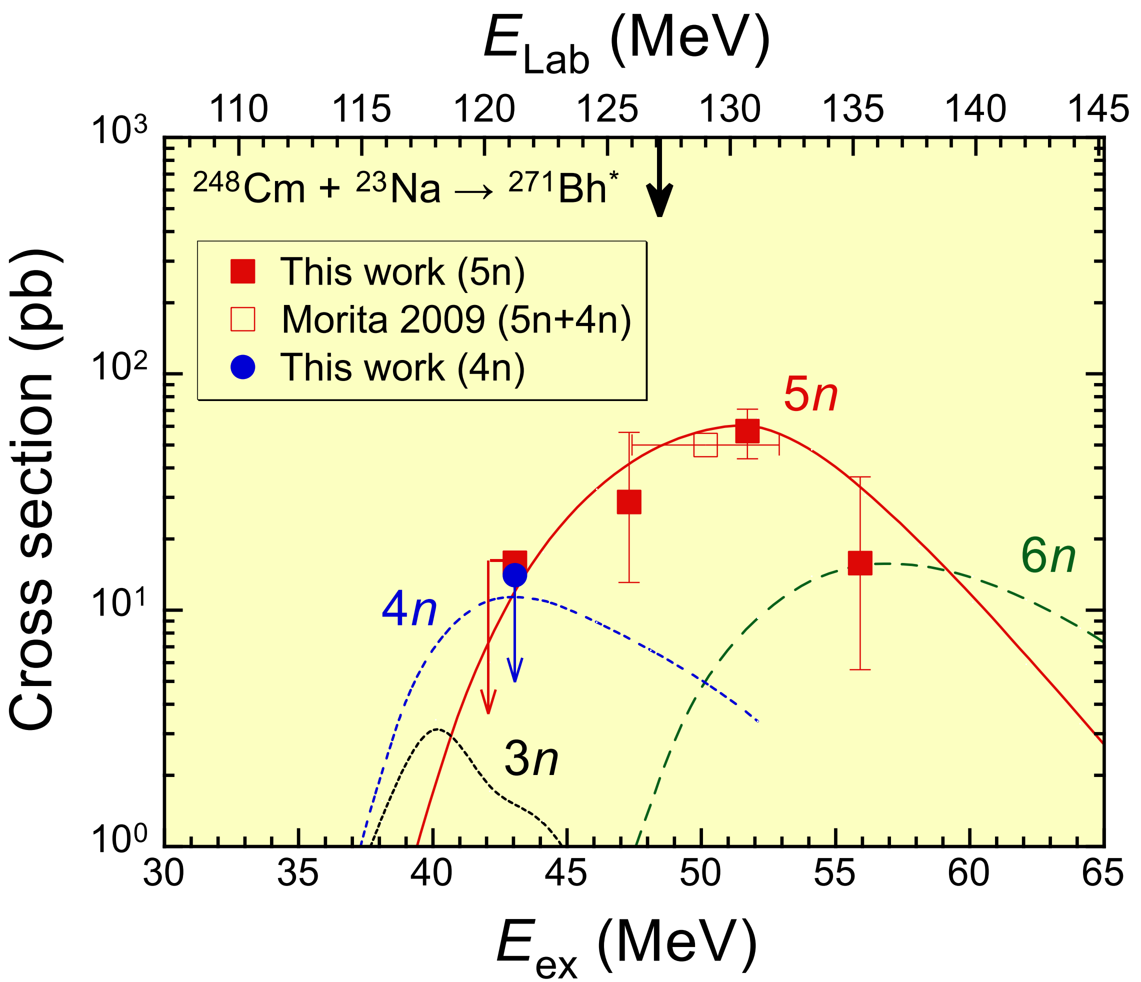




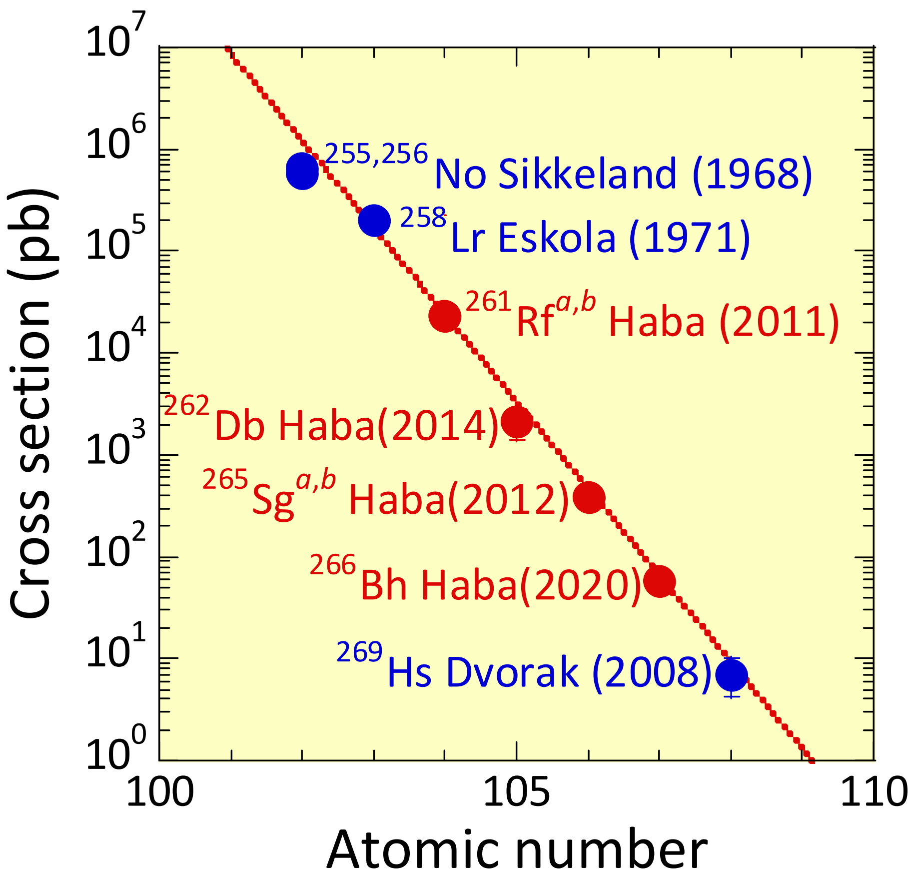
Cross section of $^{248}Cm(^{23}Na,5n)^{266}Bh$

Reaction	Cross section at 131 MeV	Reaction*	Cross sections* at 117/123 MeV
$^{248}Cm(^{23}Na,5n)^{266}Bh$	**57 ± 14 pb**	$^{249}Bk(^{22}Ne,5n)^{266}Bh$	-/25–250 pb
		$^{249}Bk(^{22}Ne,4n)^{267}Bh$	$58^{+33}_{-15}/96^{+55}_{-25}$ pb

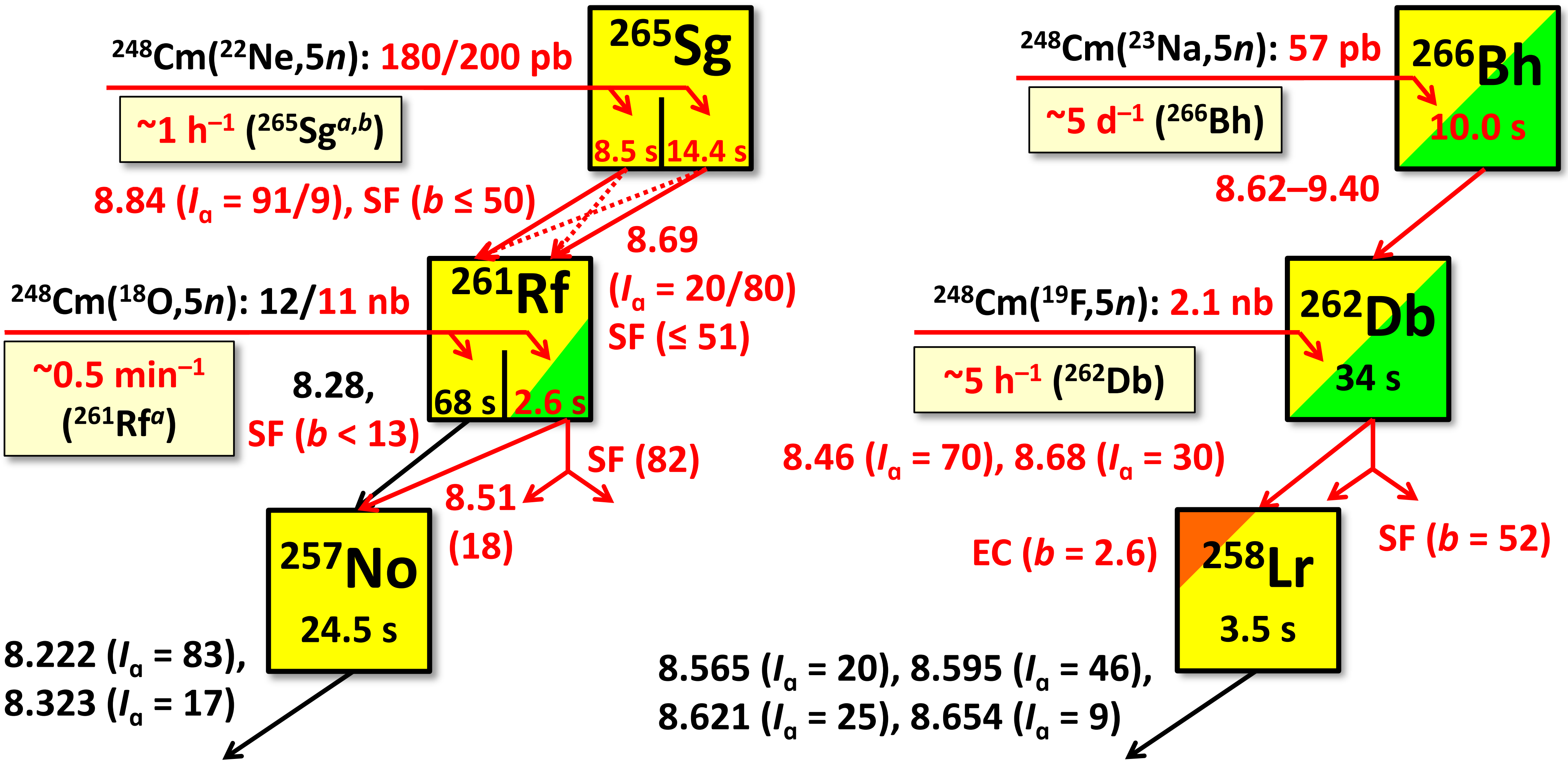
*Wilk *et al.*, PRL **85**, 2697 (2000).



Cross sections for $^{248}Cm(X,5n)$



Production and decay studies of ^{261}Rf, ^{262}Db, ^{265}Sg, and ^{266}Bh

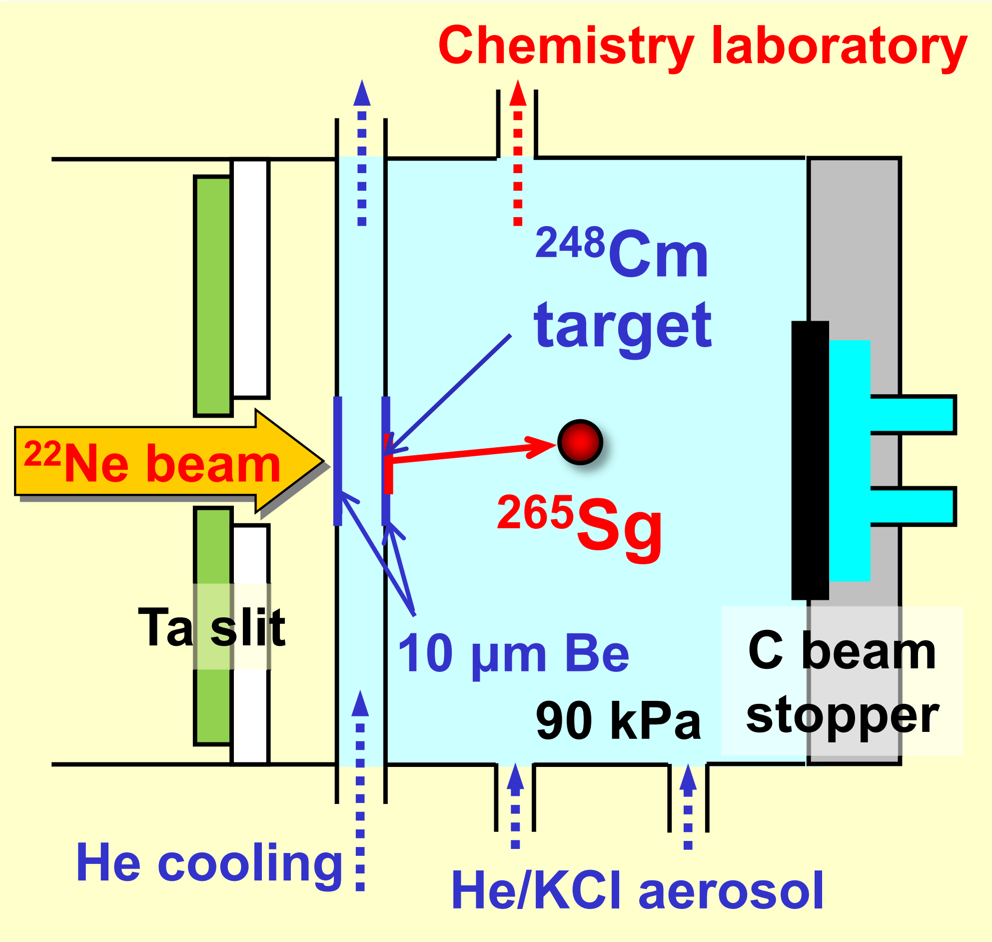


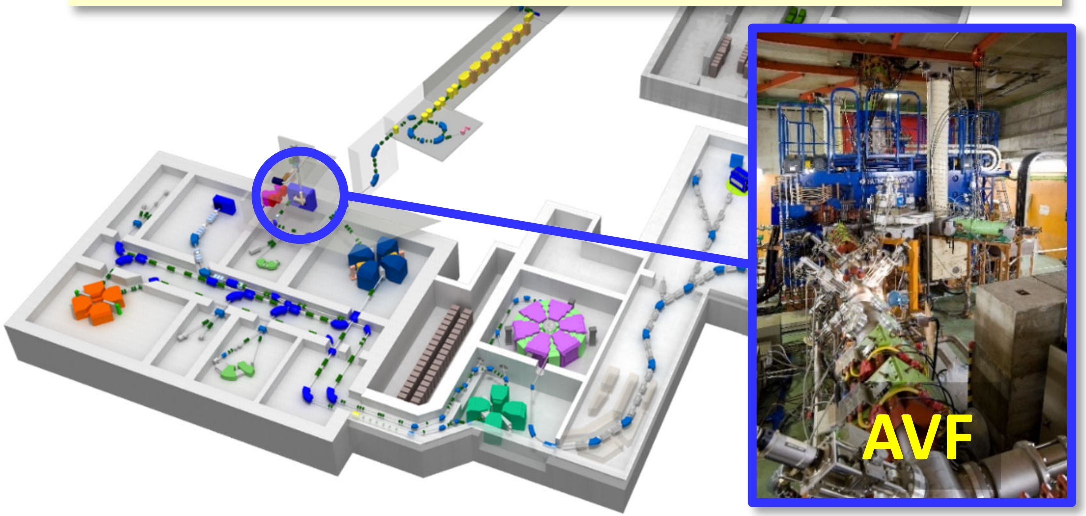
H. Haba et al., Chem. Lett. **38**, 426 (2009).
H. Haba et al., Phys. Rev. C **83**, 034602 (2011).
H. Haba et al., Phys. Rev. C **85**, 024611 (2012).
M. Murakami et al., Phys. Rev. C **88**, 024618 (2013).
H. Haba et al., Phys. Rev. C **89**, 024618 (2014).
H. Haba, EPJ Web Conf. **131**, 07006 (2016).
H. Haba et al., Phys. Rev. C **102,** 024625 (2020).

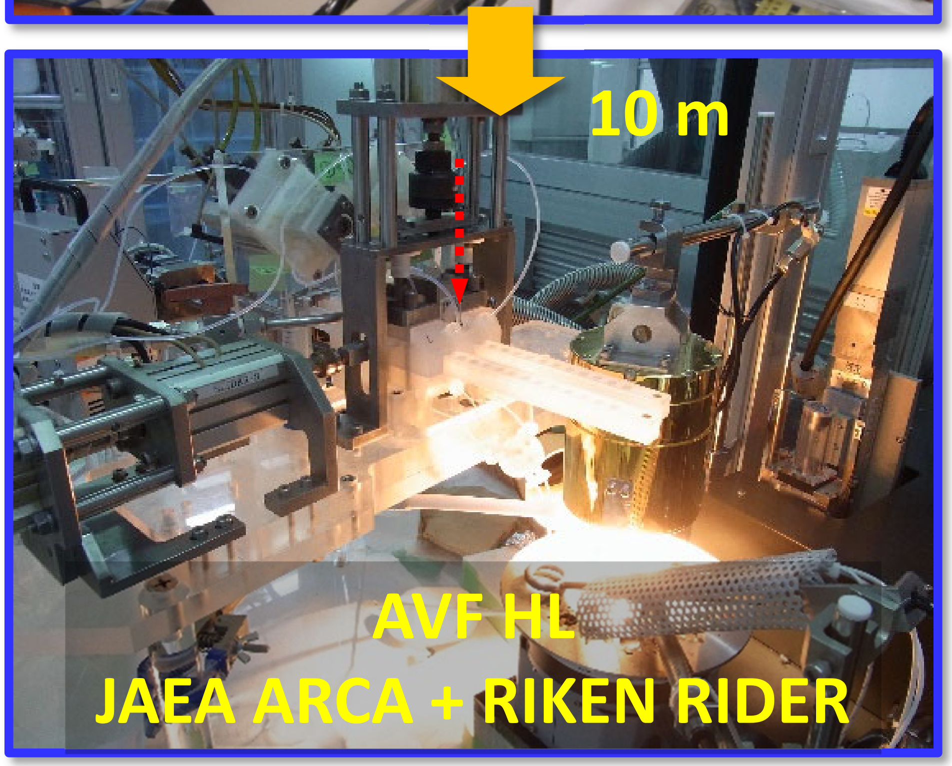
Pre-separated SHE RIs are ready for chemistry experiments.

5. Solution chemistry of Rf and Db

SHE production system at RIKEN AVF cyclotron





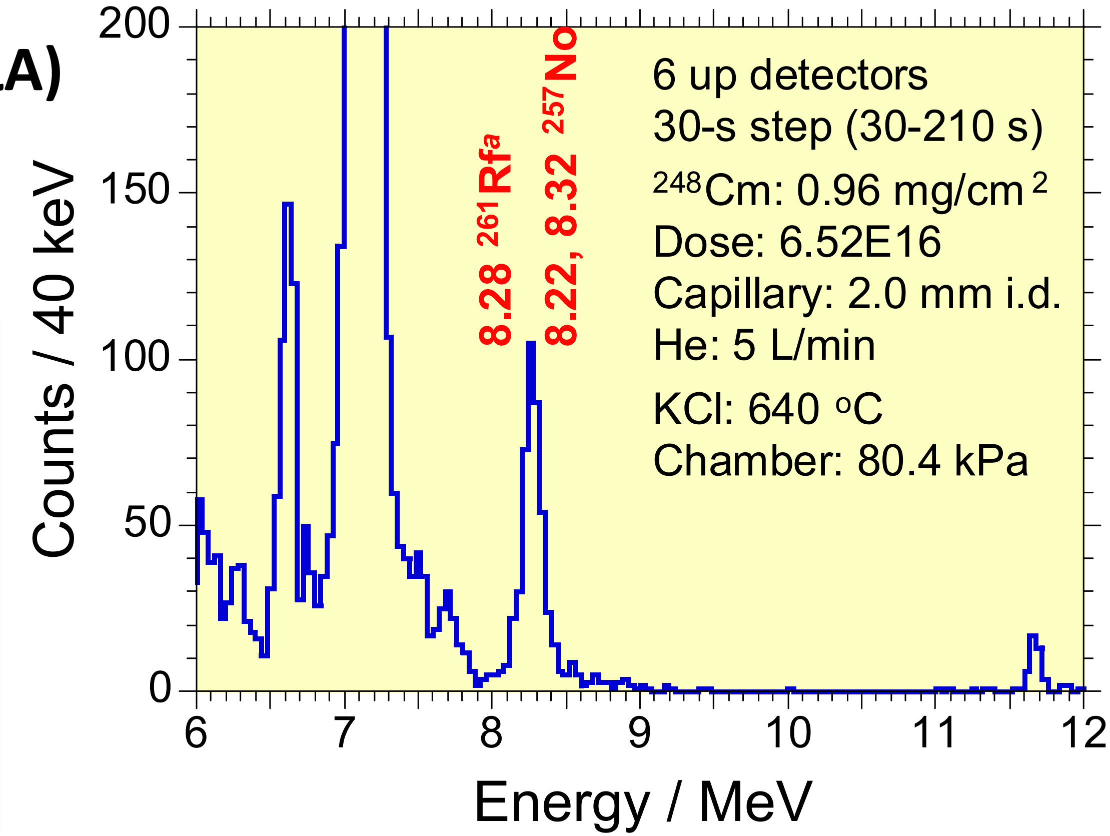


Conventional target and gas-jet system for production of SHEs

- **Heavy-ion beams from AVF:**
  ^{12}C, ^{18}O, ^{19}F, ^{22}Ne, and ^{26}Mg (~1 pμA)
- **High production yields without a recoil separator**

Z	Reaction	Half-life
102	^{248}Cm(^{12}C,5n)^{255}No	3.52 min
104	^{248}Cm(^{18}O,5n)^{261}Rf^{a}	68 s
105	^{248}Cm(^{19}F,5n)^{262}Db	34 s
106	^{248}Cm(^{22}Ne,5n)^{265}Sg^{b}	14.4 s
108	^{248}Cm(^{26}Mg,5n)^{269}Hs	9.7 s

^{248}Cm(^{18}O,5n)^{261}Rf^{a}



Exp.	Beam [MeV]	Beam [pμA]	Irrad. [h]	No. of α events	Gas-jet eff.** [%]	σ*** [nb]	Yield at AVF HL [atoms/min]
^{255}No	79.1	0.944	1.09	2689	46	900	**200**
^{261}Rf^{a}	96.4	0.870	3.34	865*	63	17	**5**

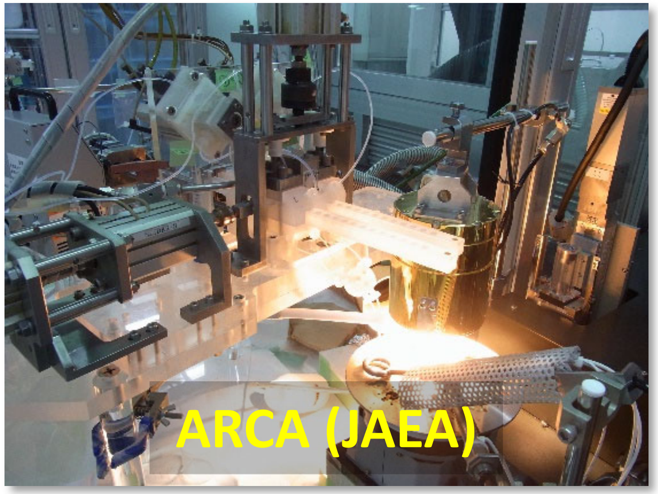
* Including α particles of ^{257}No.

** Estimated from the gas-jet efficiencies of ^{169}Hf produced in ^{nat}Gd(^{18}O,xn)^{169}Hf.

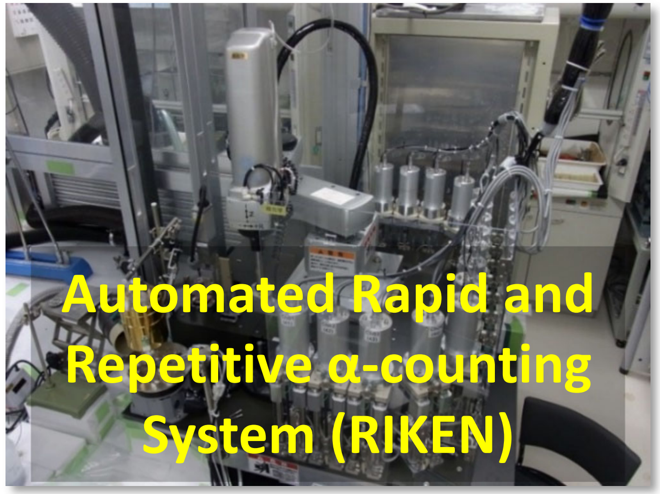
*** The target thicknesses were assumed to be 325 and 569 μg/cm^2 for ^{255}No and ^{261}Rf^{a}, respectively.

Solution chemistry of Rf and Db at AVF

ARCA (JAEA)
  Liq. chromatography
CHIN (Osaka Univ.)
  Co-precipitation
AMBER (Osaka Univ.)
ISE (Osaka Univ.)
  Liq.-liq. flow extraction



+



- **Reversed-phase TTA extraction of Rf fluoride with ARCA** (Kanazawa Univ.)
  A. Yokoyama et al., Radiochim. Acta **107**, 27 (2019). → **Rf < Zr ≈ Hf**
- **Extraction of Rf chloride with Aliquat 336 resin** (Osaka Univ.)
  T. Yokokita et al., Dalton Trans. **45**, 18827 (2016). → **Rf > Zr > Hf**
- **Hydroxide co-precipitation of Rf with $Sm(OH)_3$** (Osaka Univ.)
  Y. Kasamatsu et al., Nat. Chem. **13**, 226 (2021). → **Rf > Zr ≈ Hf**
- **Anion-exchange of Rf sulfate with AMBER** (RIKEN)
  A. Yokokita et al., to be submitted. → **Rf (≈ Th) < Zr ≈ Hf**
- **Reversed-phase TBP extraction of Db fluoride** (Niigata Univ.)
  M. Murakami et al., to be submitted. → **Db ≈ Nb, Db ≠ Ta**

	Group 4
5	$_{40}Zr$
6	$_{72}Hf$
7	$_{104}Rf$

6. Summary

- Present status of RIKEN RIBF facilities for SHE research was introduced.
- Element 113 was synthesized in the cold fusion of ^{209}Bi + ^{70}Zn.
- A synthesis experiment of element 119 in the $^{248}Cm(^{51}V,xn)^{299-x}119$ reaction is ongoing using GARIS III at the upgraded SRILAC facility.
- Production and decay properties of ^{261}Rf, ^{262}Db, ^{265}Sg, and ^{266}Bh were investigated using the GARIS gas-jet system coupled to the rotating wheel apparatus for α and SF spectrometry.
- Formations of chloride, fluoride, hydroxide, and sulphate complexes of Rf and fluoride complexes of Db were investigated using the conventional gas-jet transport system at AVF.