Production and decay studies of ^{261}Rf, ^{262}Db, and ^{265}Sg at GARIS@RIKEN

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- Future plans

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

Periodic table of the elements (2014)

	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 La	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
Lanthanide				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
Actinide				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
7	87 Fr	88 Ra	89 Ac	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113	114 Fl	115	116 Lv	117	118

Superheavy elements (SHEs)

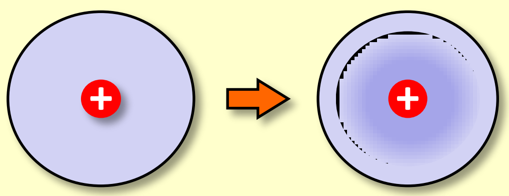
1. Chemistry of Superheavy Elements

Frontiers in chemistry

- **Chemical properties, periodicity, and electronic structure of new elements?**
  Verification of the influence of relativistic effects on chemical reactions
- **Small cross sections (nb or pb) and short half-lives (< 1min)**
  → Rapid and effective chemical experiments with "single atoms" at accelerators

Direct

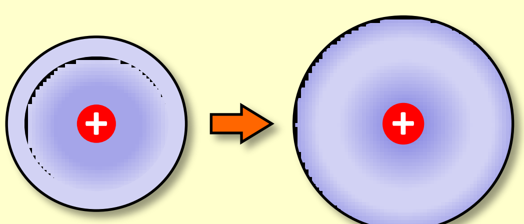
Contraction of inner orbital (*s*, *p*)



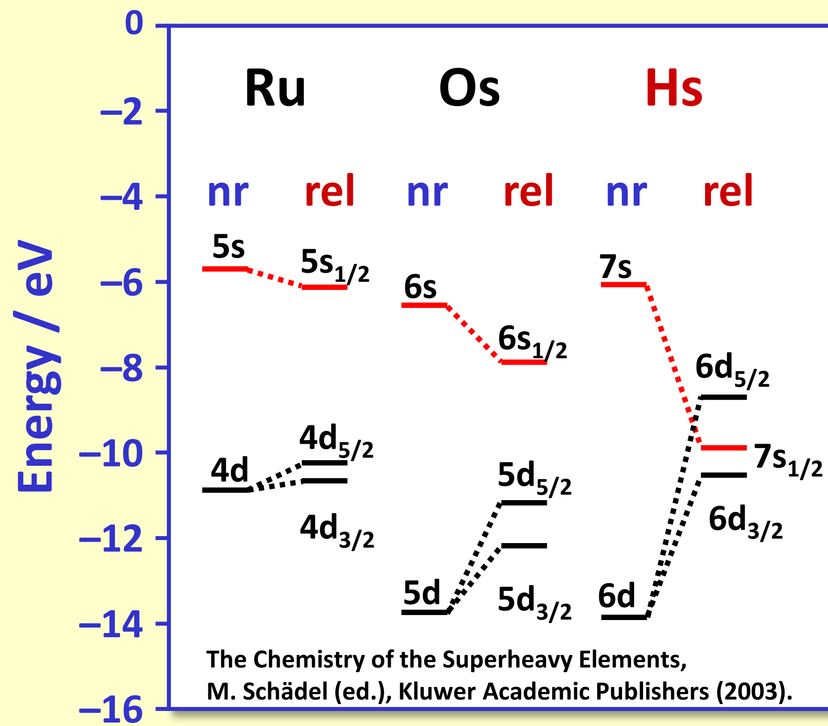
$$m = \frac{m_0}{\sqrt{1-(v/c)^2}} \qquad a_B = \frac{\hbar^2}{me^2} = a_B^0\sqrt{1-(v/c)^2}$$

Indirect

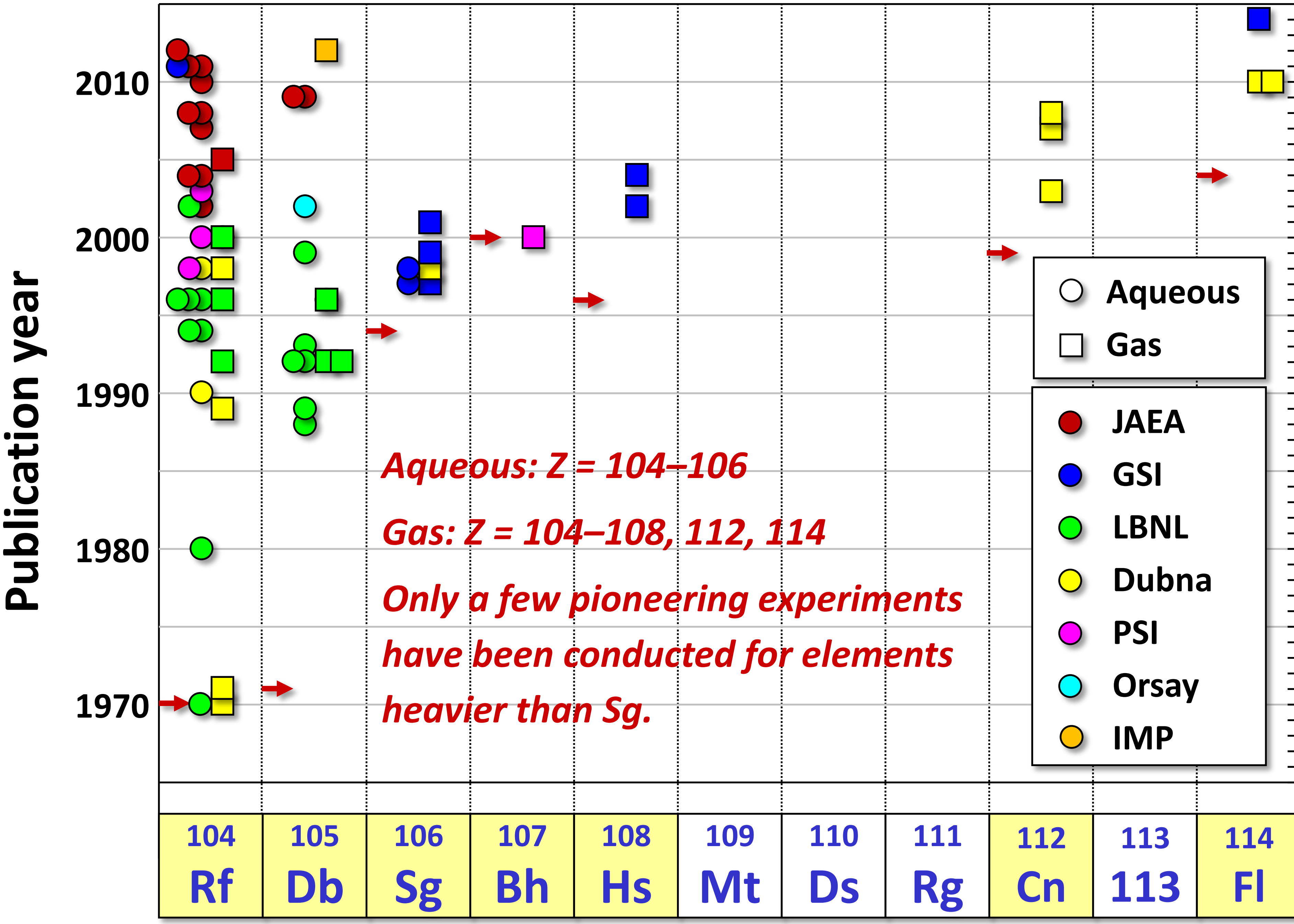
Expansion of outer orbital (*d*, *f*)



Energy levels of valence electrons

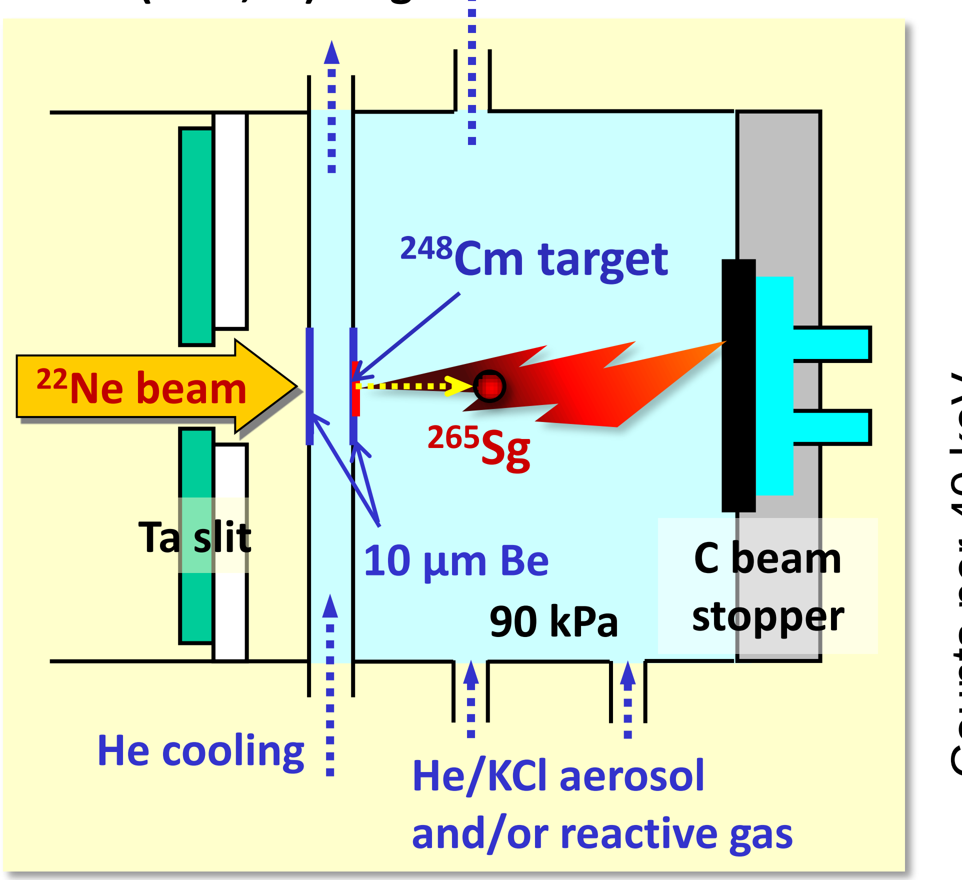


Publications of Experimental Studies on SHE Chemistry



Gas-jet transport technique just behind the target

$^{248}Cm(^{22}Ne,5n)^{265}Sg$

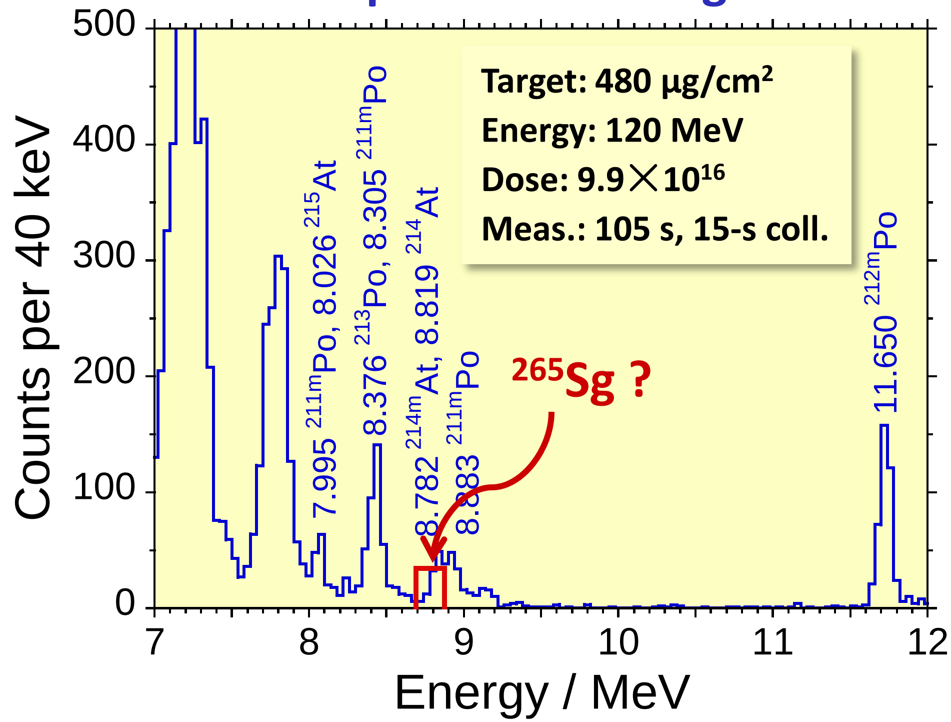


Chemistry apparatuses

Gas or liquid chromatography

→ α/SF spectrometry

α spectrum of ^{265}Sg



Limitations

- Large amount of background radioactivities from unwanted reaction products
- Decrease of gas-jet yields due to plasma condition induced by an intense beam

2. RIKEN GARIS for SHE chemistry

Coupling SHE chemistry to recoil separators

Breakthroughs in SHE chemistry

- **Chemical and physical experiments under low background condition**
- **Stable and high gas-jet transport efficiency**
- **New chemical reactions**

Development of a gas-jet transport system coupled to GARIS

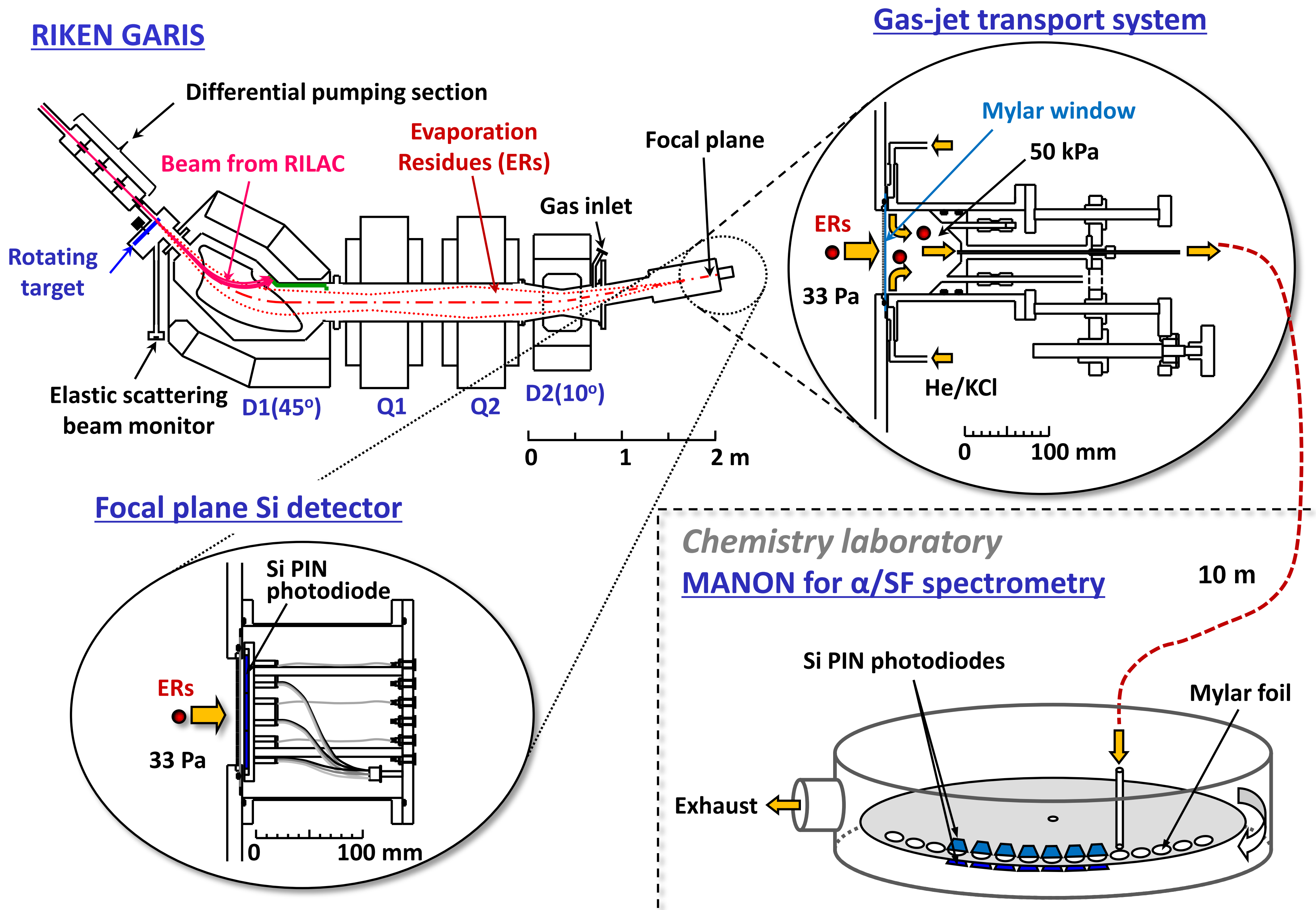
- $\mathbf{^{169}Tm(^{40}Ar,3\mathit{n})^{206}Fr}$; $\mathbf{^{208}Pb(^{40}Ar,3\mathit{n})^{245}Fm}$ [JNRS **8**, 55 (2007); EPJD **45**, 81 (2007)]
- $\mathbf{^{238}U(^{22}Ne,5\mathit{n})^{255}No}$ [JNRS **9**, 27 (2008)]

In this presentation

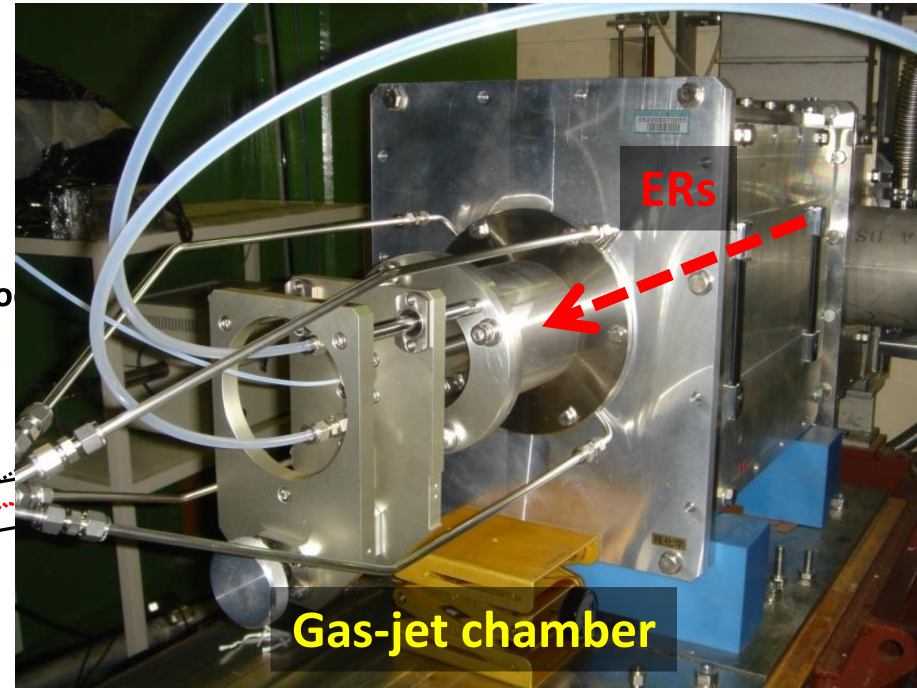
Production and decay studies of $^{261}Rf^{a,b}$, $^{265}Sg^{a,b}$, and ^{262}Db

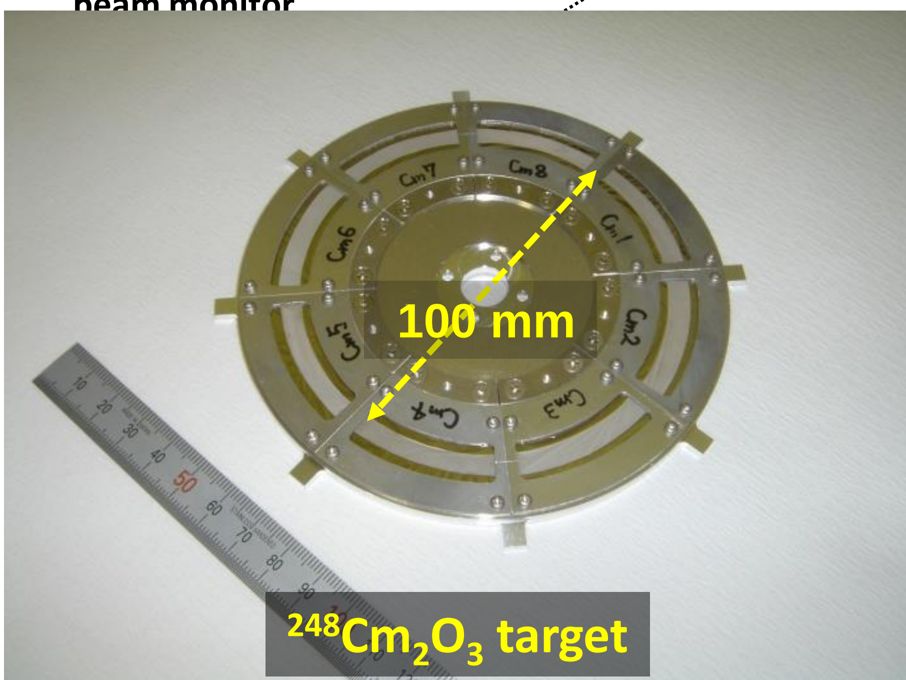
- $\mathbf{^{248}Cm(^{18}O,5\mathit{n})^{261}Rf^{\mathit{a,b}}}$ [Chem. Lett. **38**, 426 (2009); PRC **83**, 034602 (2011); PRC **88**, 024618 (2013)]
- $\mathbf{^{248}Cm(^{22}Ne,5\mathit{n})^{265}Sg^{\mathit{a,b}}}$ [PRC **85**, 024611 (2012)]
- $\mathbf{^{248}Cm(^{19}F,5\mathit{n})^{262}Db}$ [PRC **89**, 024618 (2014)]

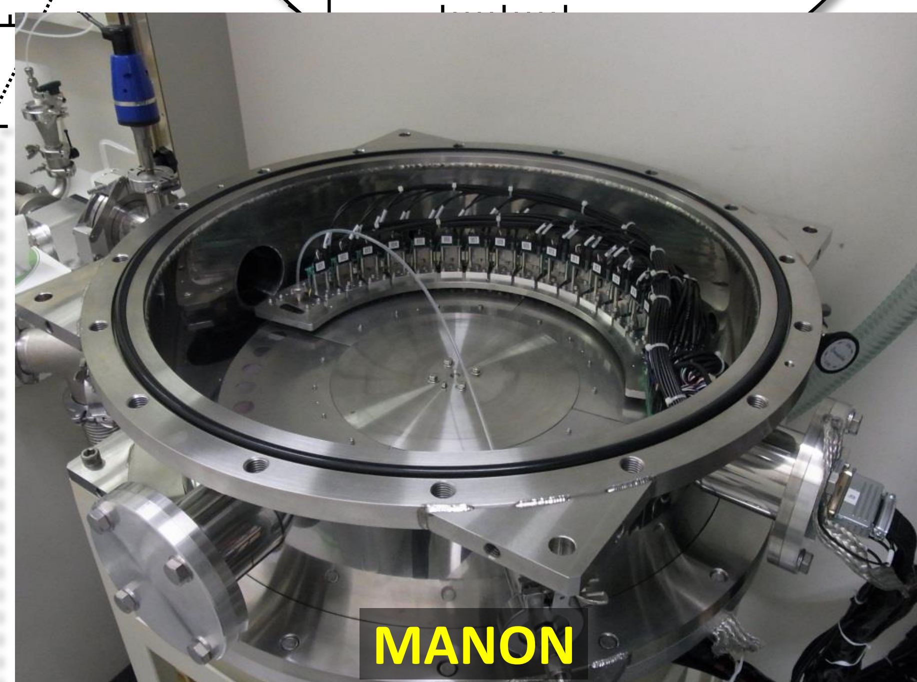
Experimental setup



Experimental setup





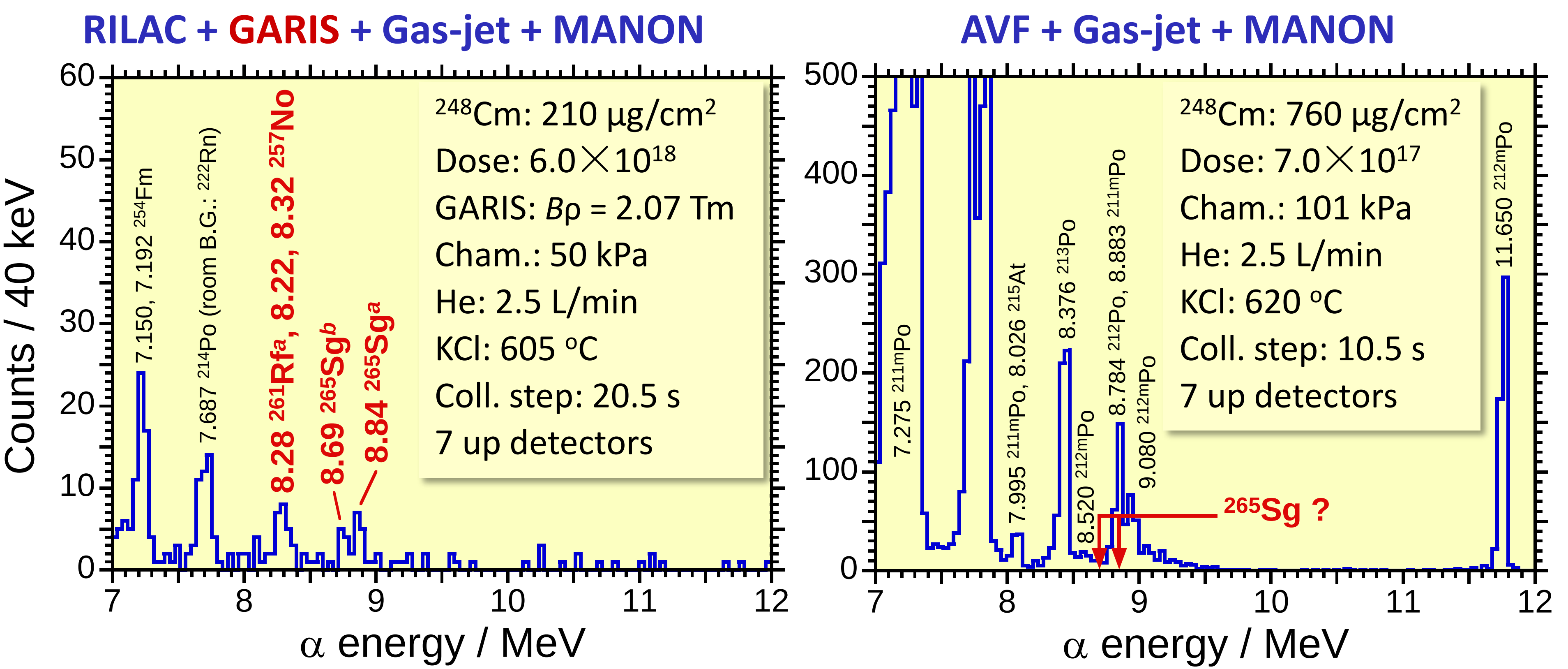


Production of ^{261}Rf, ^{262}Db, and ^{265}Sg using the GARIS gas-jet system

Nuclide	^{261}Rf[a,b] (Z=104)	^{262,263}Db (Z=105)	^{265}Sg[a,b] (Z=106)
Half-life	68, 3 s[1)]	34 s, 27 s[2)]	8.9, 16.2 s[1)]
Reaction	^{248}Cm(^{18}O,5n)	^{248}Cm(^{19}F,5;4n)	^{248}Cm(^{22}Ne,5n)
Cross section (nb)	12[3)], ?	1.5[3)], ?	0.2–0.3[1)] ?
Beam energy (MeV)	95	103, 97.4	118
Beam intensity (pμA)	7	4	3
$^{248}Cm_2O_3$ thickness (μg/cm^2)	280/230	230/290/330	230/280
Magnetic rigidity (Tm)	1.58–2.16	1.73–2.09	1.73–2.16
GARIS He (Pa)	33	32	33
RTC Mylar window (μm)	0.5	0.5	0.7
Honeycomb grid (%)	78/84	84	72/84
Gas-jet He (kPa)	49	47	49
Chamber depth (mm)	20	20	40
He flow rate (L/min)	2.0	2.0	2.0
KCl generator (°C)	620	620	600/605
Step interval of MANON (s)	30.5, 2.0	15.5	20.5/10.5

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).

(a) $^{248}Cm(^{22}Ne,5n)^{265}Sg^{a,b}$



Beam energy (MeV)	$^{248}Cm_2O_3$ target ($\mu g/cm^2$)	Magnetic rigidity (Tm)	Beam dose ($\times 10^{18}$)	Step interval of MANON
117.8	280	1.73	2.07	20.5
117.8	280	1.94	1.91	20.5
117.8	280	1.94	0.431	10.5
117.8	280	2.16	1.57	20.5
117.8	280	2.04	0.639	20.5
117.8	230	2.07	11.2	20.5

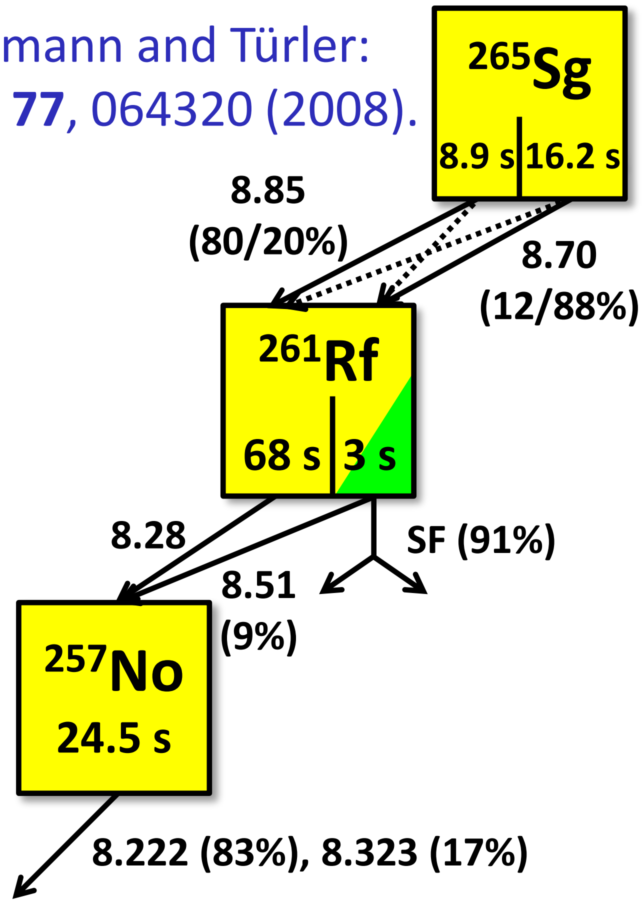
Search for α-α/α-SF correlations

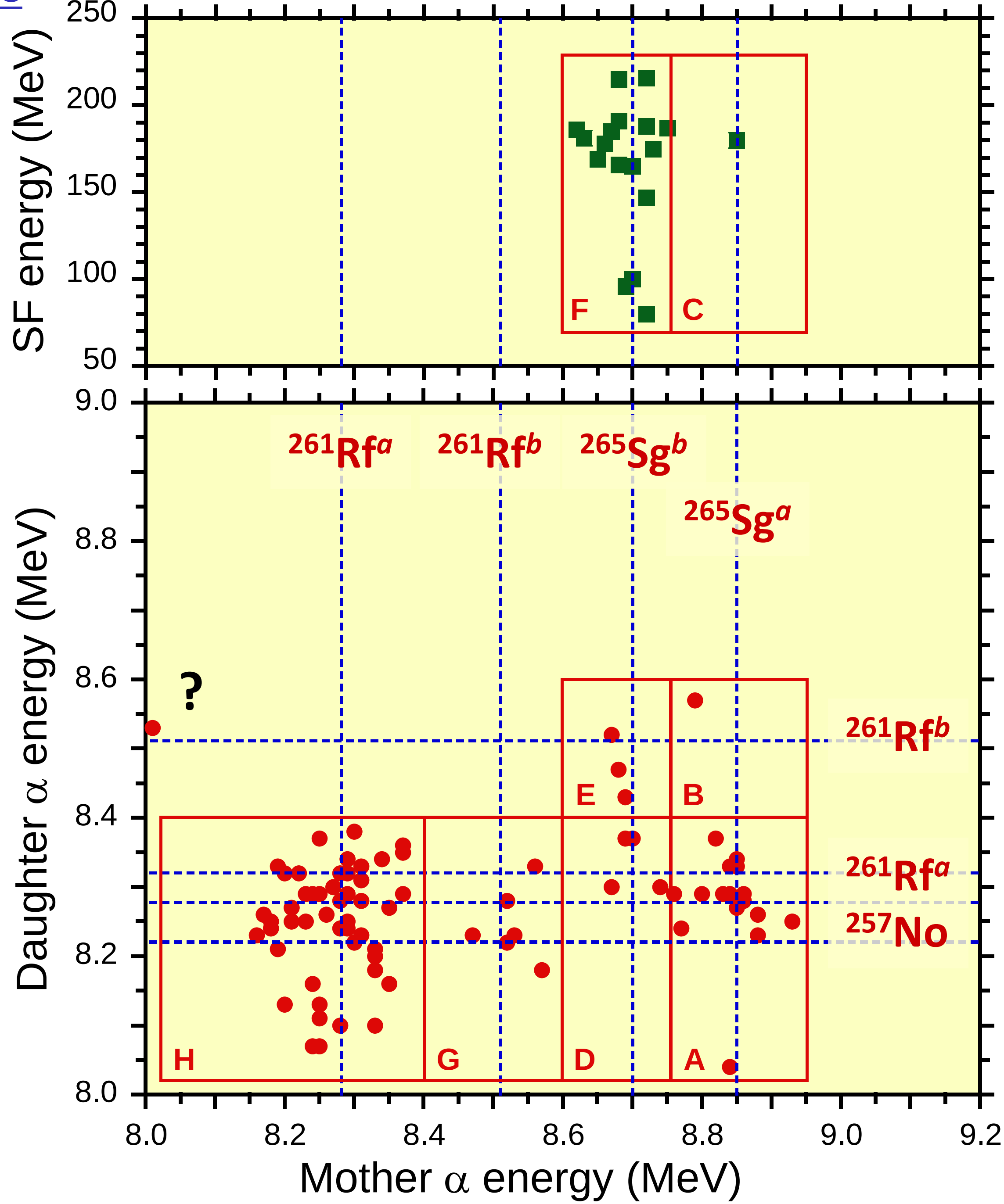
E_α = 8.0–9.0 MeV; $E_{SF} \geq$ 30 MeV

ΔT = 226 s

	Observed	Random
α-α	**97**	< 5.3
α-α-α	**18**	< 0.1
α-SF	**18**	< 2.0

Düllmann and Türler: PRC **77**, 064320 (2008).

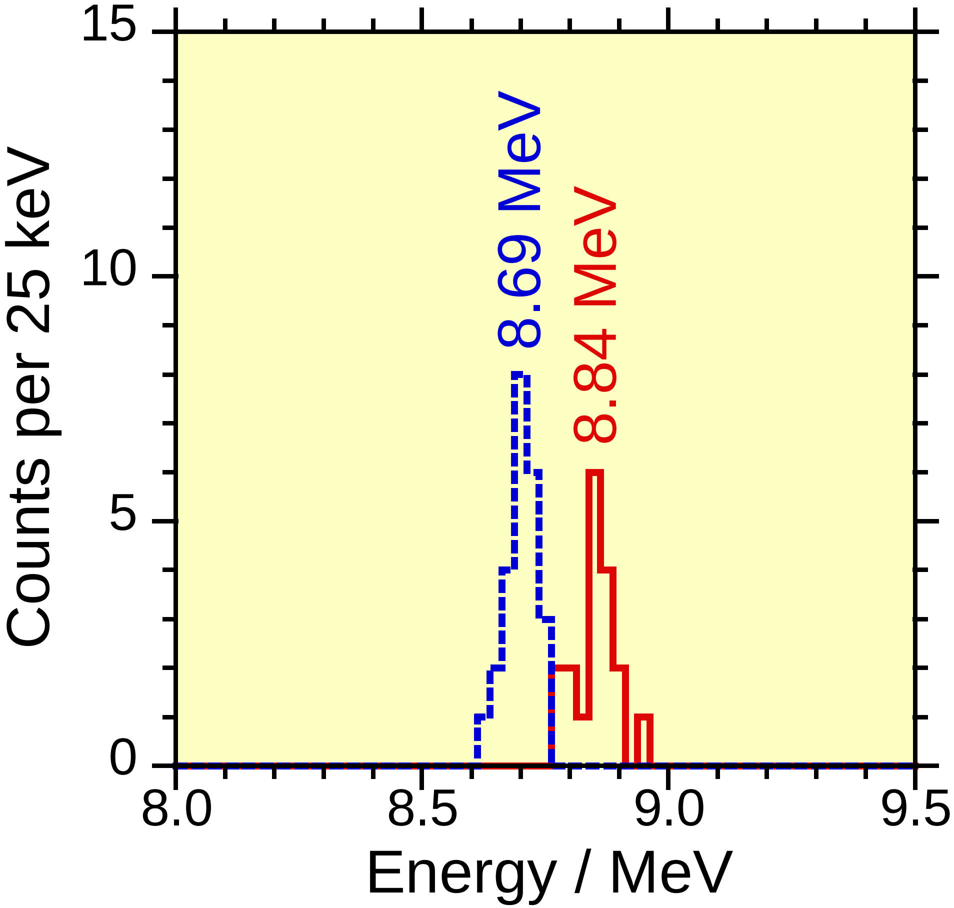




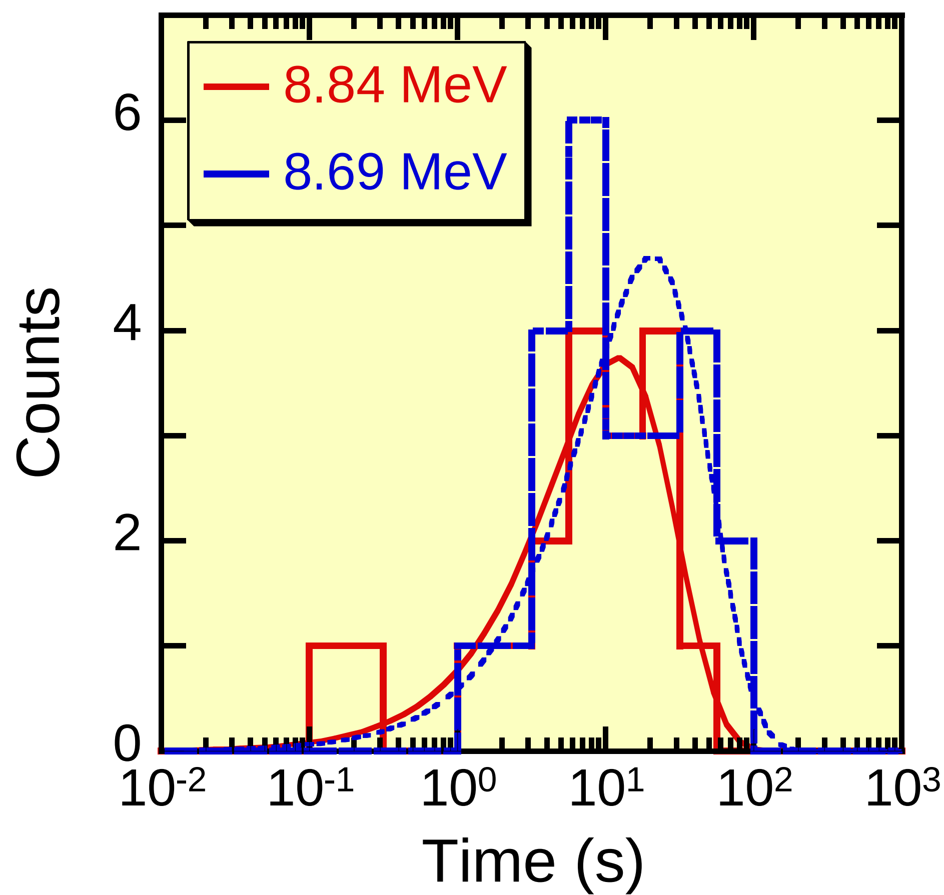
α energy and half-life of $^{265}Sg^{a,b}$

	This work				Düllmann and Türler (2008)		
	n	E_α [MeV]	$T_{1/2}$ [s]	b_{SF} [%]	n	E_α [MeV]	$T_{1/2}$ [s]
$^{265}Sg^a$	**18**	**8.84±0.05**	**$8.5^{+2.6}_{-1.6}$**	**≤ 50**	20	8.85	$8.9^{+2.7}_{-1.3}$
$^{265}Sg^b$	**24**	**8.69±0.05**	**$14.4^{+3.7}_{-2.5}$**	**≤ 51**	24	8.70	$16.2^{+4.7}_{-1.9}$

α spectrum of $^{265}Sg^{a,b}$



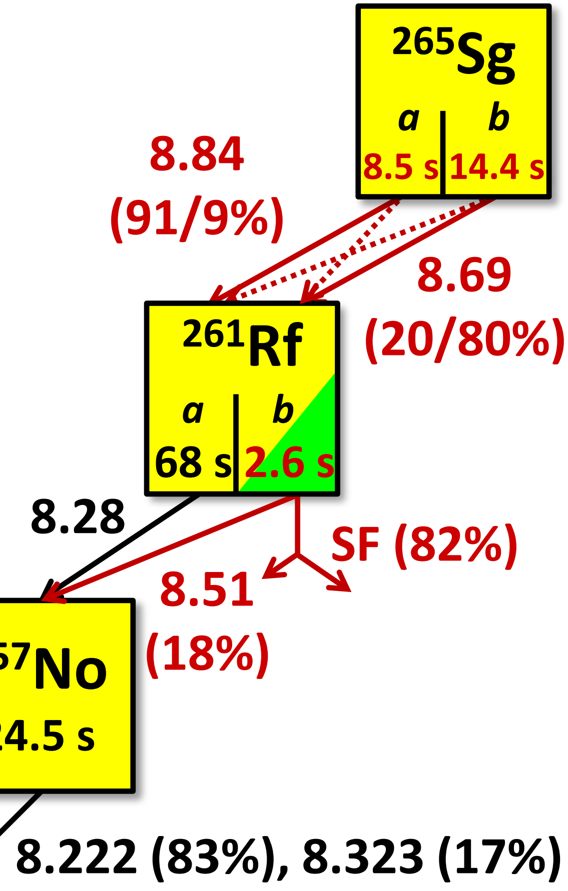
Decay time spectrum of $^{265}Sg^{a,b}$



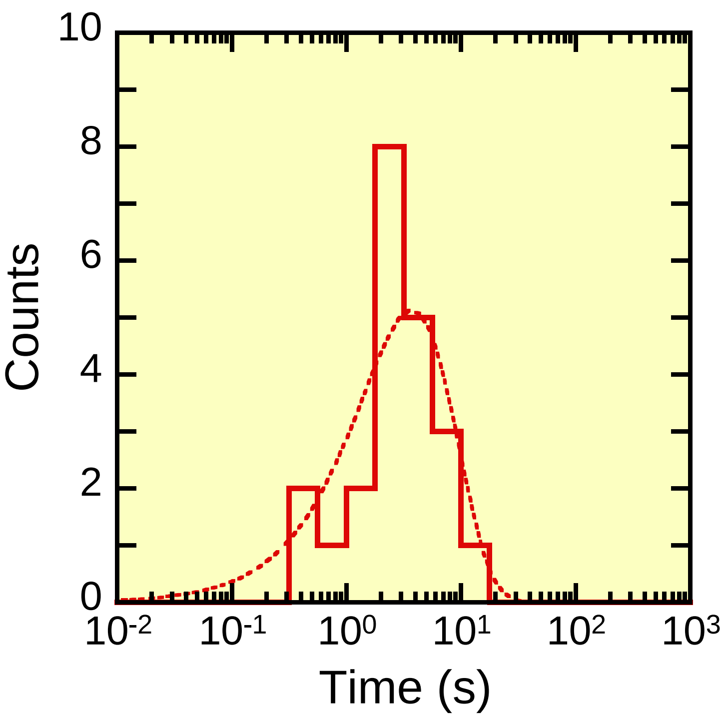
Decay patterns observed in the chain $^{265}Sg^{a,b} \rightarrow {}^{261}Rf^{a,b} \rightarrow {}^{257}No$

^{265}Sg state	→	^{261}Rf state	No. of events (obs.)	No. of events (corr.)	Branching ratio [%] This work	Branching ratio [%] Ref.*
a	→	*a*	**16**	19.9	**91**	80
	→	*b*	**2**	2.0	**9**	20
b	→	*a*	**4**	5.1	**20**	12
	→	*b*	**19**	19.0	**80**	88

*Düllmann and Türler, PRC C **77**, 064320 (2008).



Decay time spectrum of $^{261}Rf^{b}$



α energies and half-lives of $^{261}Rf^{a,b}$ and ^{257}No

	n	E_{α} [MeV]	$T_{1/2}$ [s]	b_{SF} [%]
$^{261}Rf^{a}$	48	8.27±0.06	59±42	
$^{261}Rf^{b}$	**25**	**8.51±0.06**	**$2.6^{+0.7}_{-0.5}$**	**82±9**
^{257}No	54	8.07–8.38	23^{+4}_{-3}	

Cross section

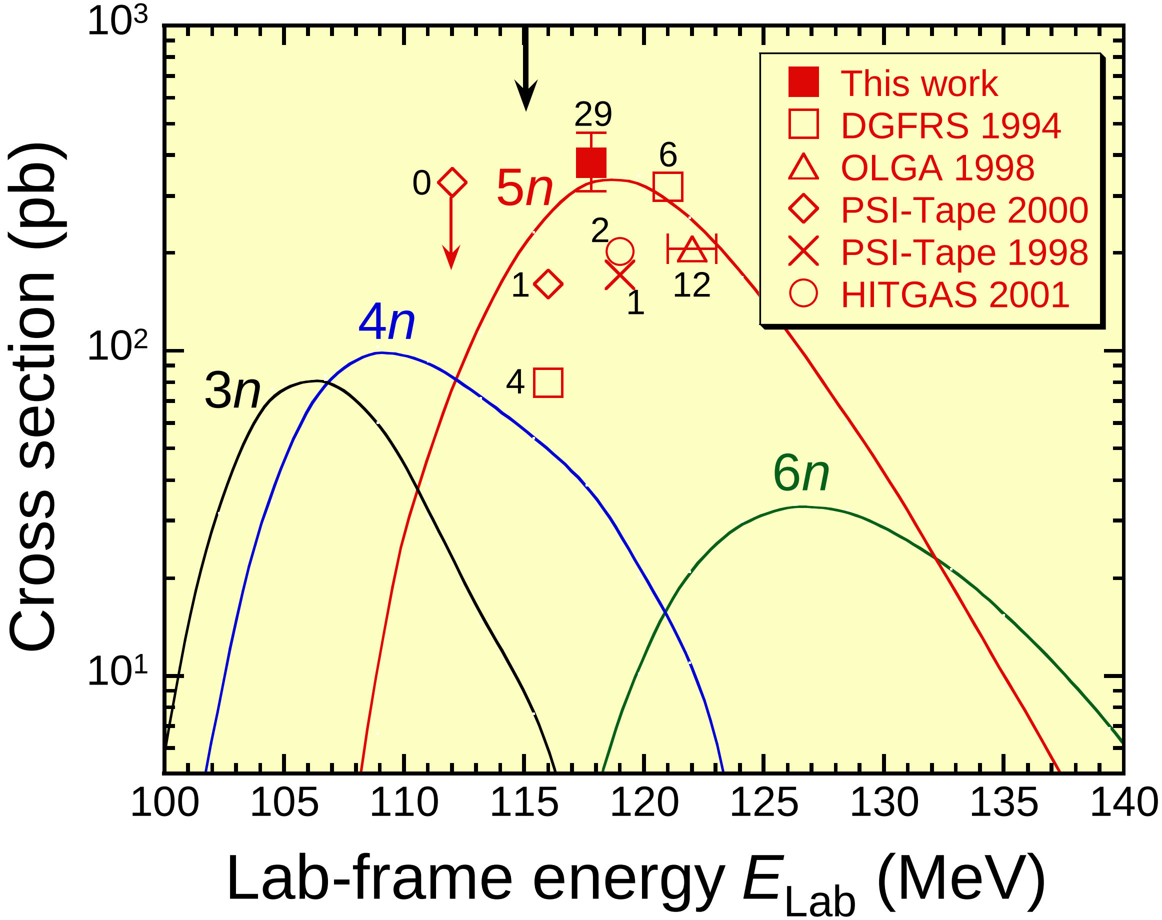
	Cross section at 117.8 MeV [pb]
$^{265}Sg^{a}$	180^{+80}_{-60}
$^{265}Sg^{b}$	200^{+60}_{-50}

$\sigma(^{265}Sg^{a} + {}^{265}Sg^{b}) = 380^{+90}_{-70}$ pb

$\sigma(^{265}Sg^{a})/\sigma(^{265}Sg^{b}) = 1.3 \pm 0.6$

Assumptions: GARIS eff. = 13%; gas-jet eff. = 50%; gas-jet transport time = 3 s

Excitation functions for $^{248}Cm(^{22}Ne,xn)$



HIVAP calculation

Reisdorf and Schädel, ZPA **343**, 47 (1992).

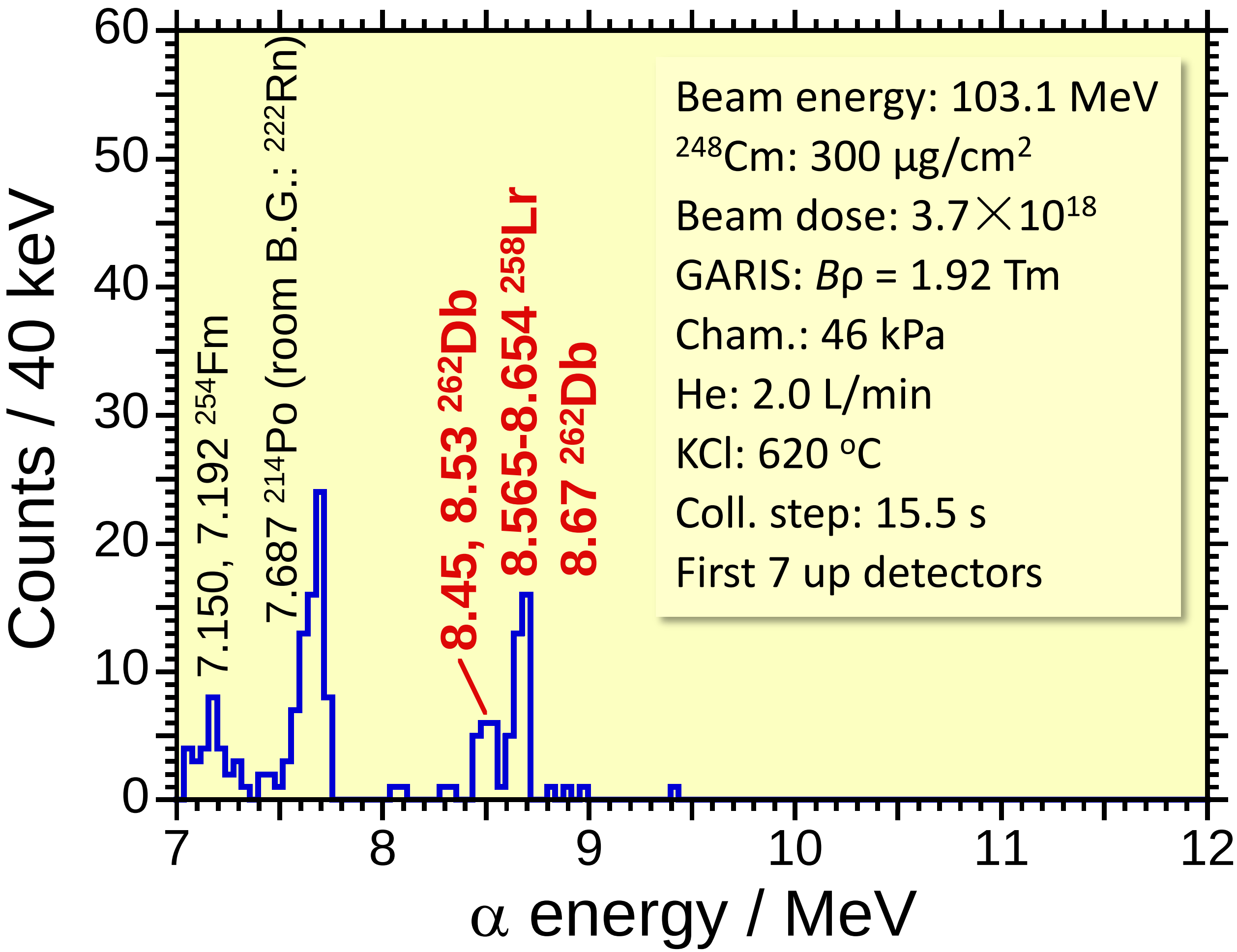
Nishio *et al.*, PRL **93**, 162701 (2004).

Nishio *et al.*, PRC **82**, 024611 (2010).

In the entrance channel, a prolate deformation of the target nucleus was taken into account to calculate the capture cross section.

(b) ^{248}Cm(^{19}F,5*n*)^{262}Db

RILAC + GARIS + Gas-jet + MANON



Beam energy (MeV)	Thickness of ^{248}Cm$_2$O$_3$ (μg/cm^2)	Magnetic rigidity (Tm)	Beam integral (×10^{18})
103.1	230	1.73	0.492
103.1	230	1.80	0.955
103.1	290	1.80	0.930
103.1	230	1.87	1.40
103.1	330	1.92	3.74
103.1	290	1.92	1.23
103.1	230	1.94	1.39
103.1	230	2.02	0.716
103.1	290	2.04	0.885
103.1	230	2.09	0.884
97.4	330	1.92	3.18
97.4	290	1.92	2.43

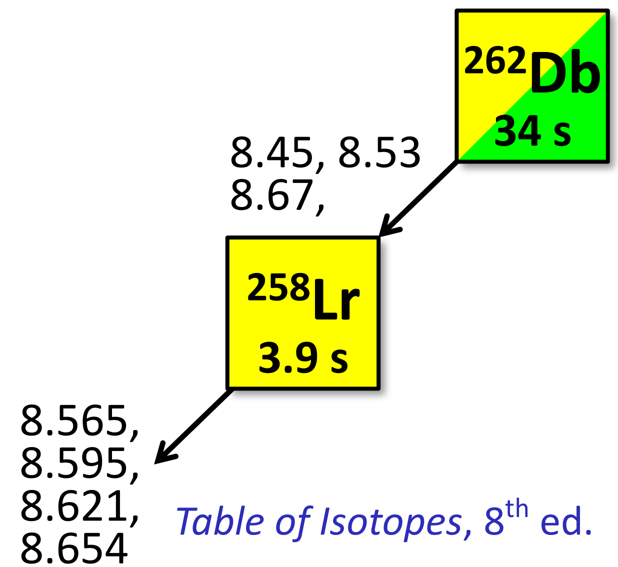


Table of Isotopes, 8th ed.

Search for correlations

E_α = 8.0–9.0 MeV; E_{SF} ≥ 30 MeV

ΔT ≤ 59.5 s

	Observed	Random
α-α	**75**	< 2.9
α-SF	**2**	< 0.6

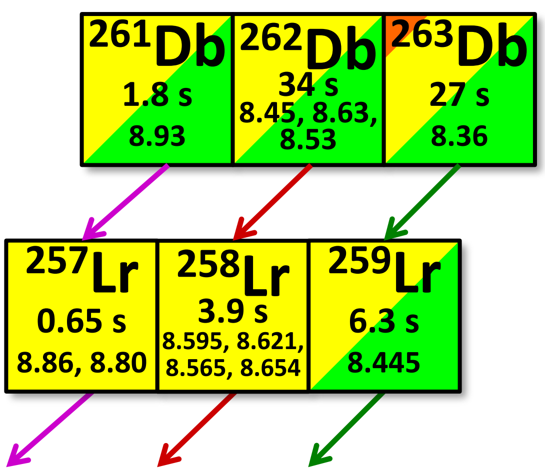
$^{248}Cm(^{19}F,5n)^{262}Db \rightarrow {}^{258}Lr$: 76

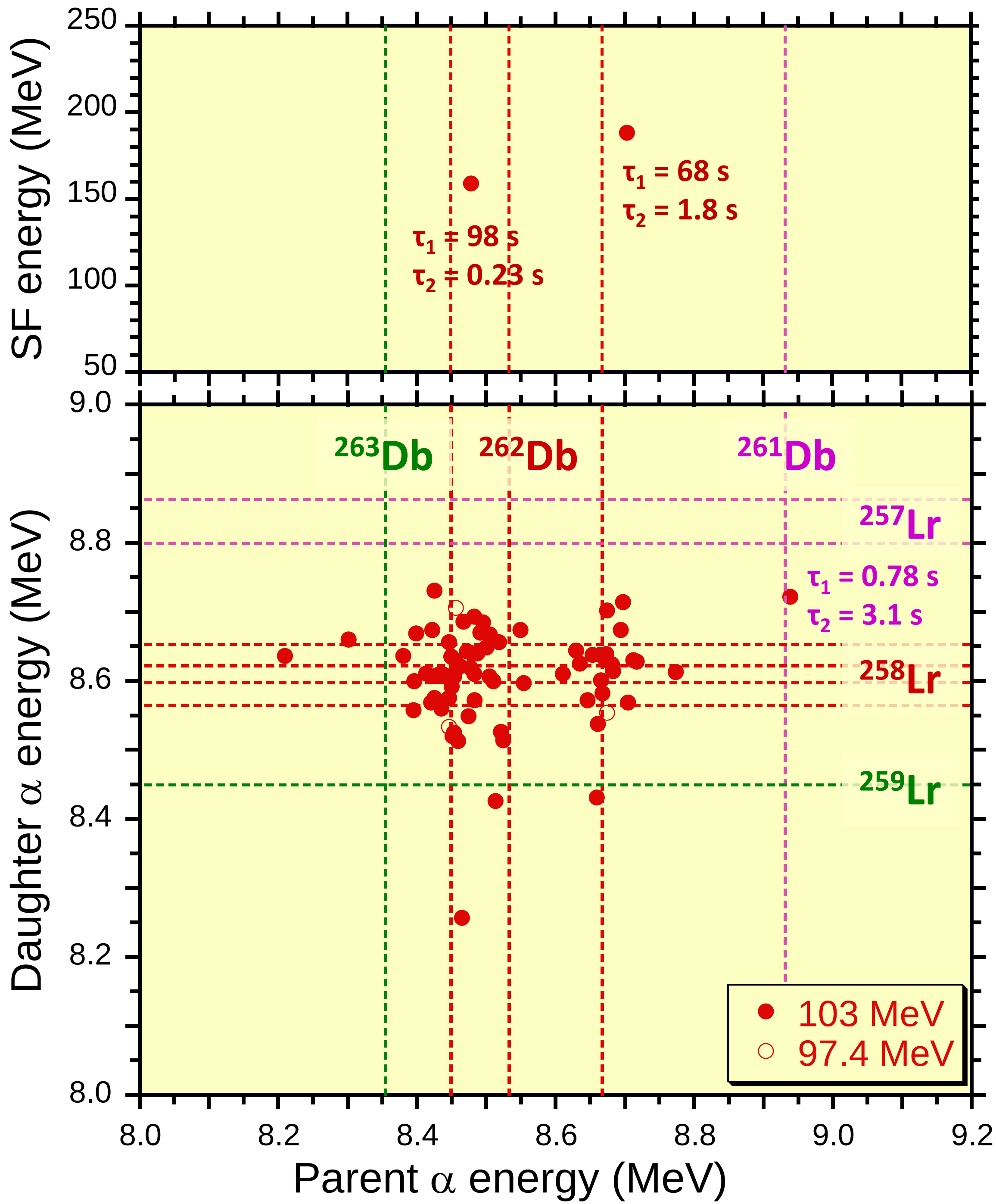
$^{248}Cm(^{19}F,6n)^{261}Db \rightarrow {}^{257}Lr$: 1

$^{248}Cm(^{19}F,4n)^{263}Db \rightarrow {}^{259}Lr$: 0

Single SF events: 123

***Table of Isotopes*, 8th ed.**

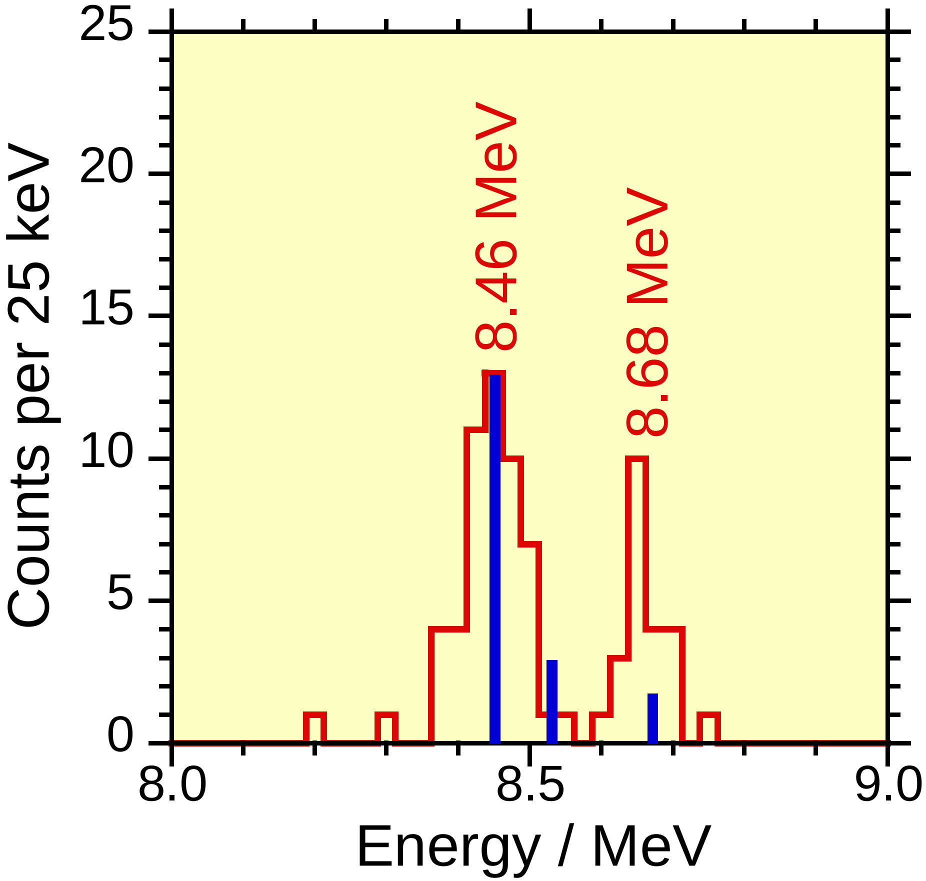




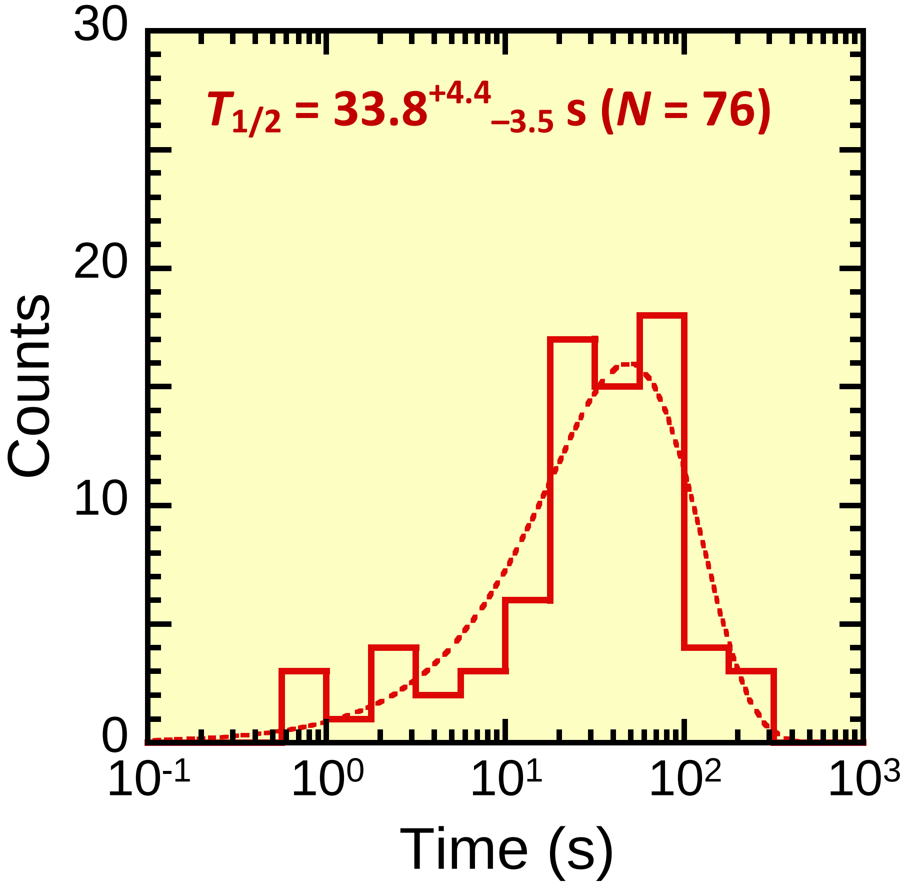
α energy and half-life of ^{262}Db

This work				Table of Isotopes 8th ed.			
$T_{1/2}$ [s]	E_α [MeV]	I_α [%]	b_{SF} [%]	$T_{1/2}$ [s]	E_α [MeV]	I_α [%]	b_{SF} [%]
	8.46±0.04	**70±5**			8.45±0.02	75	
$33.8^{+4.4}_{-3.5}$			**52±4**	34±4	**8.53±0.02**	**16**	**≈33**
	8.68±0.04	**30±5**			8.67±0.02	9	

α spectrum of ^{262}Db

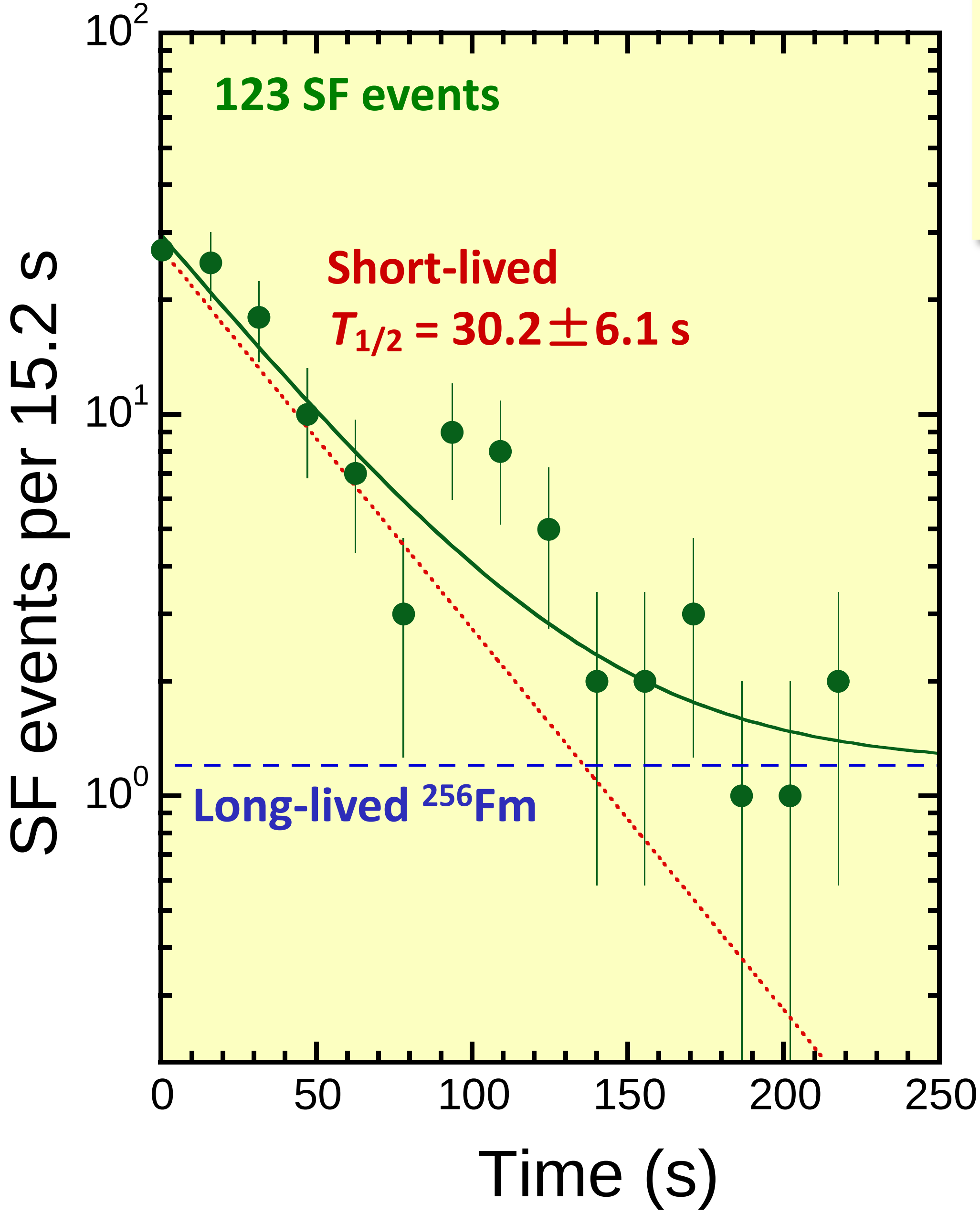


Decay time spectrum of ^{262}Db



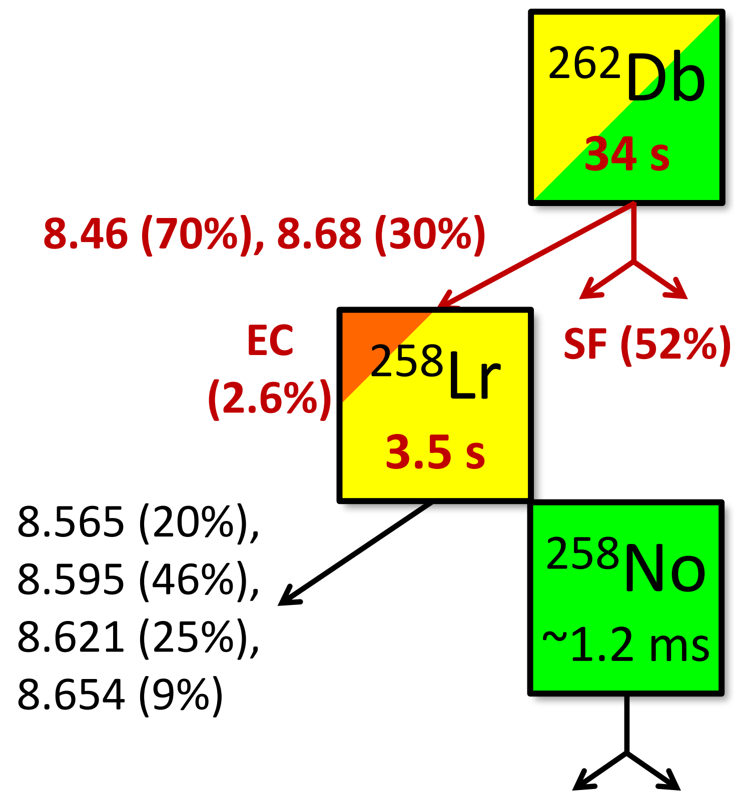
Single SF events

2-components decay curve analysis



- $T_{1/2} = 33.8^{+4.4}_{-3.5}$ s from α decay of ^{262}Db
- No α events of ^{263}Db (b_{α} = 43%)
  → Negligible SF of ^{263}Db (b_{SF} = 57%)

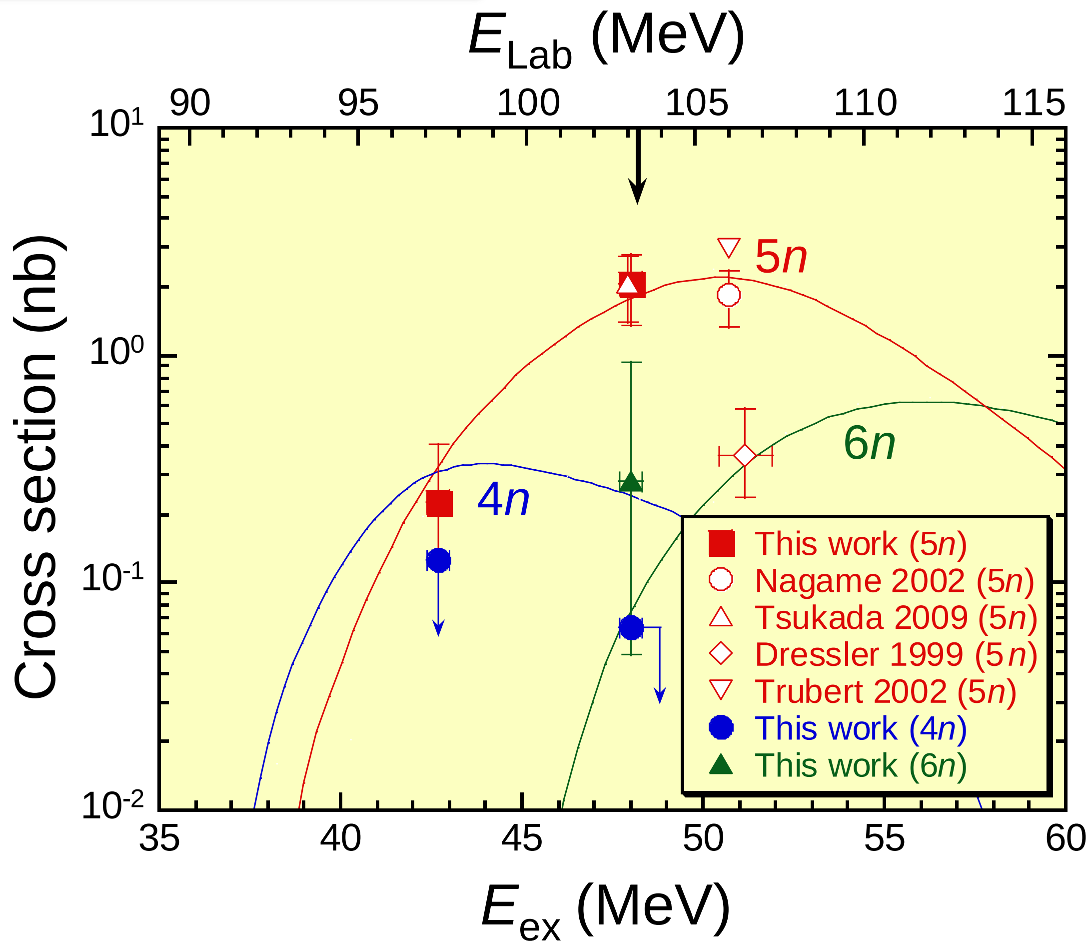
SF branch of ^{262}Db:
b_{SF} = 52±4%



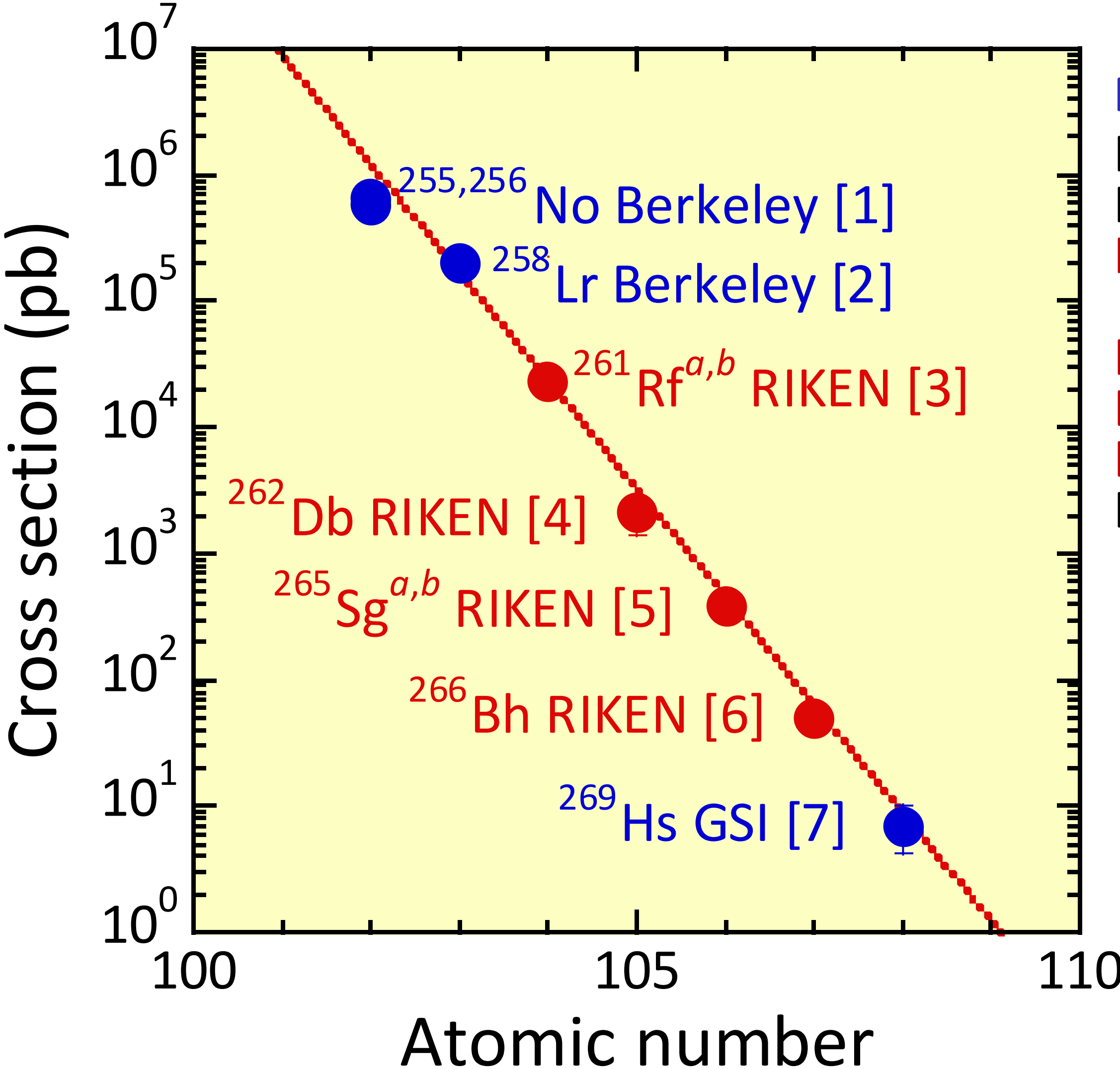
Cross section for $^{248}Cm(^{19}F,xn)^{267-x}Db$

- New decay data of ^{262}Db and ^{258}Lr
  $b_{SF}(^{262}Db)$ = 52%; $b_{EC}(^{258}Lr)$ = 2.6%
- Nagame *et al.*, JNRS **3**, 85 (2002).
  σ = 1.5 ± 0.4 nb at 103 MeV
  for $^{248}Cm(^{19}F,5n)^{262}Db$

Products	Cross sections [nb] 103.1 MeV	Cross sections [nb] 97.4 MeV
^{261}Db (6*n*)	**$0.28^{+0.65}_{-0.23}$**	**< 0.10**
^{262}Db (5*n*)	**2.1 ± 0.7**	**$0.23^{+0.18}_{-0.11}$**
^{263}Db (4*n*)	**< 0.064**	**< 0.13**



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



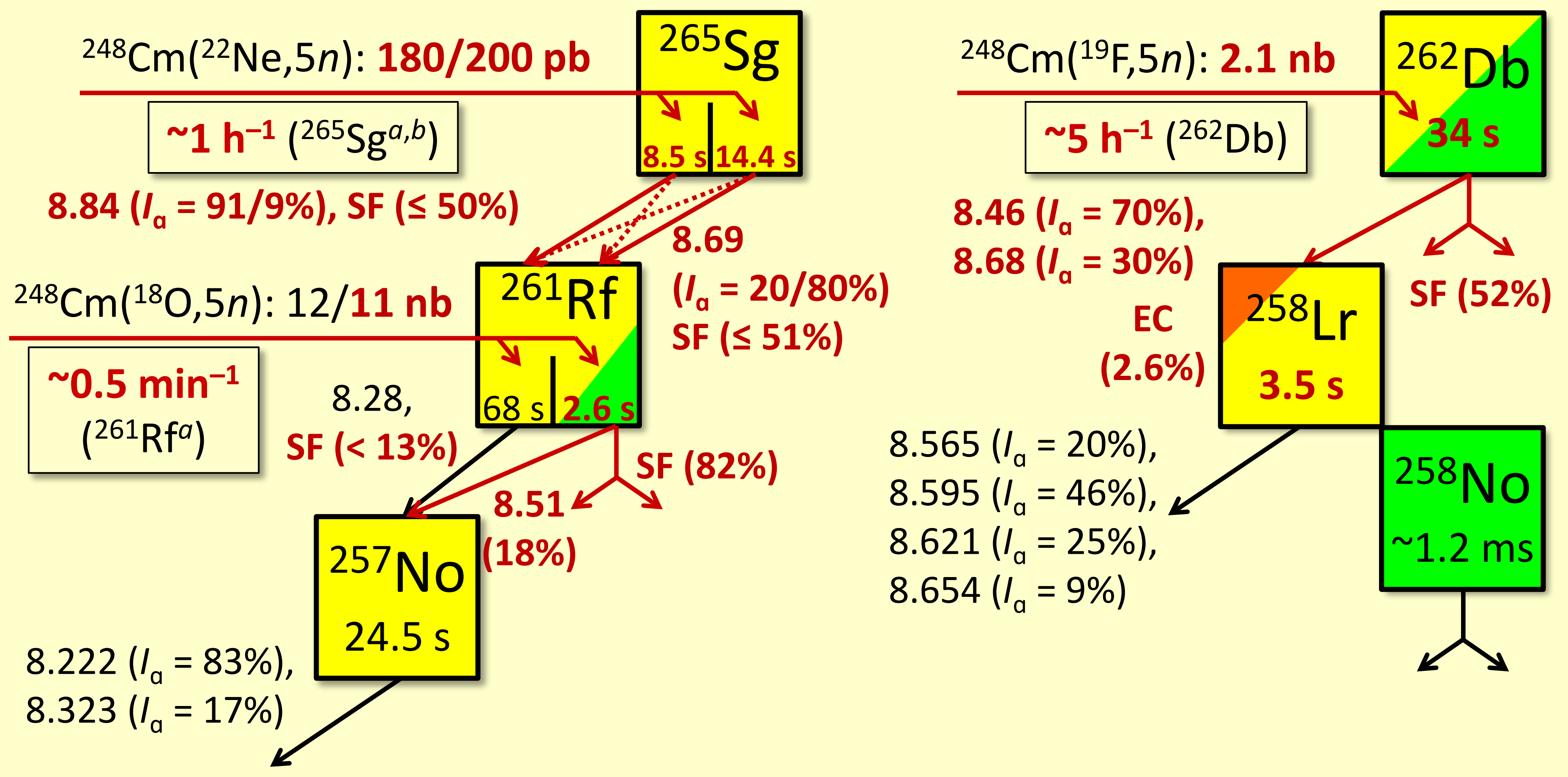
References

[1] Sikkeland *et al.*, PL **172**, 1232 (1968).
[2] Eskola *et al.*, PRC **4**, 632 (1971).
[3] Haba *et al.*, PRC **83**, 034602 (2011); Murakami *et al.*, PRC **88**, 024618 (2013).
[4] Haba *et al.*, PRC **89**, 024618 (2014).
[5] Haba *et al.*, PRC **85**, 024611 (2012).
[6] Morita *et al.*, JPSJ **78**, 064201 (2009).
[7] Dvorak *et al.*, PRL **100**, 132503 (2008).

→ $^{248}Cm(^{27}Al,5n)^{271}Mt$ (Z = 109): ≈ 1 pb

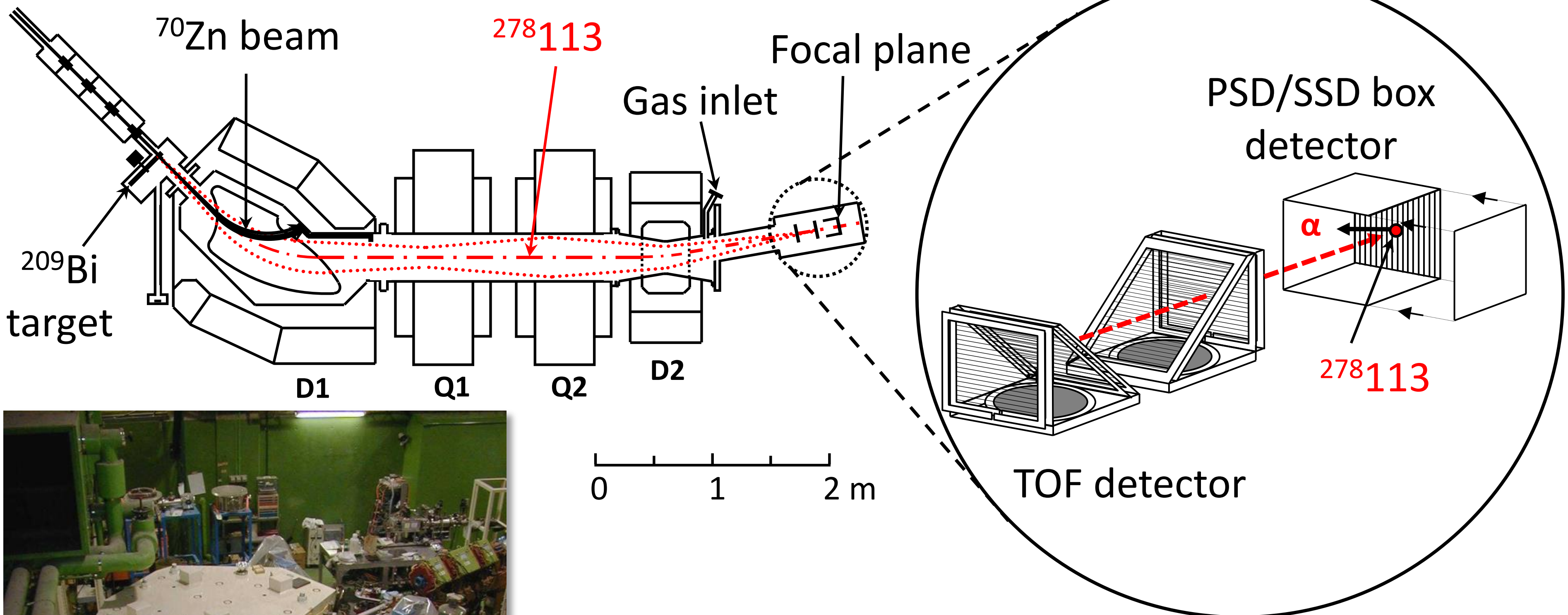
3. Summary

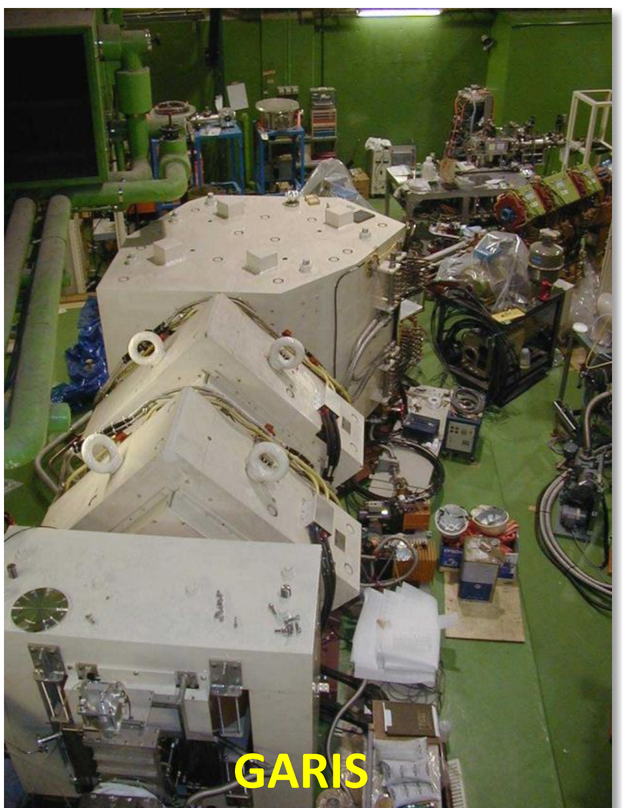
- **The gas-jet transport system was installed in RIKEN GARIS for SHE chemistry.**
- **The production and decay properties of ^{261}Rf, ^{262}Db, and ^{265}Sg for chemical studies were investigated using MANON under low background conditions attained by the GARIS gas-jet system.**

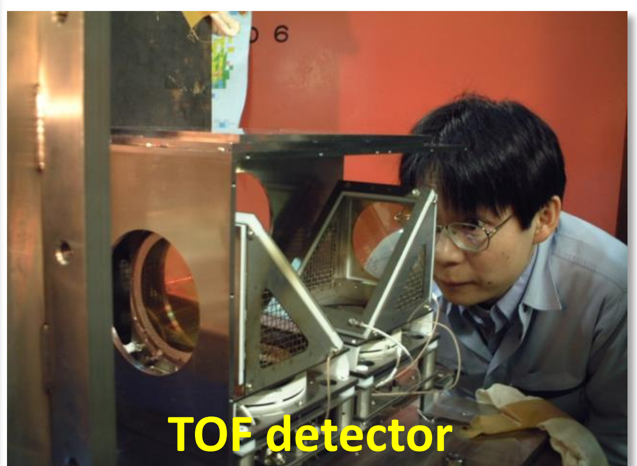


Summary of the $^{209}Bi(^{70}Zn,n)^{278}113$ experiment

Experimental setup







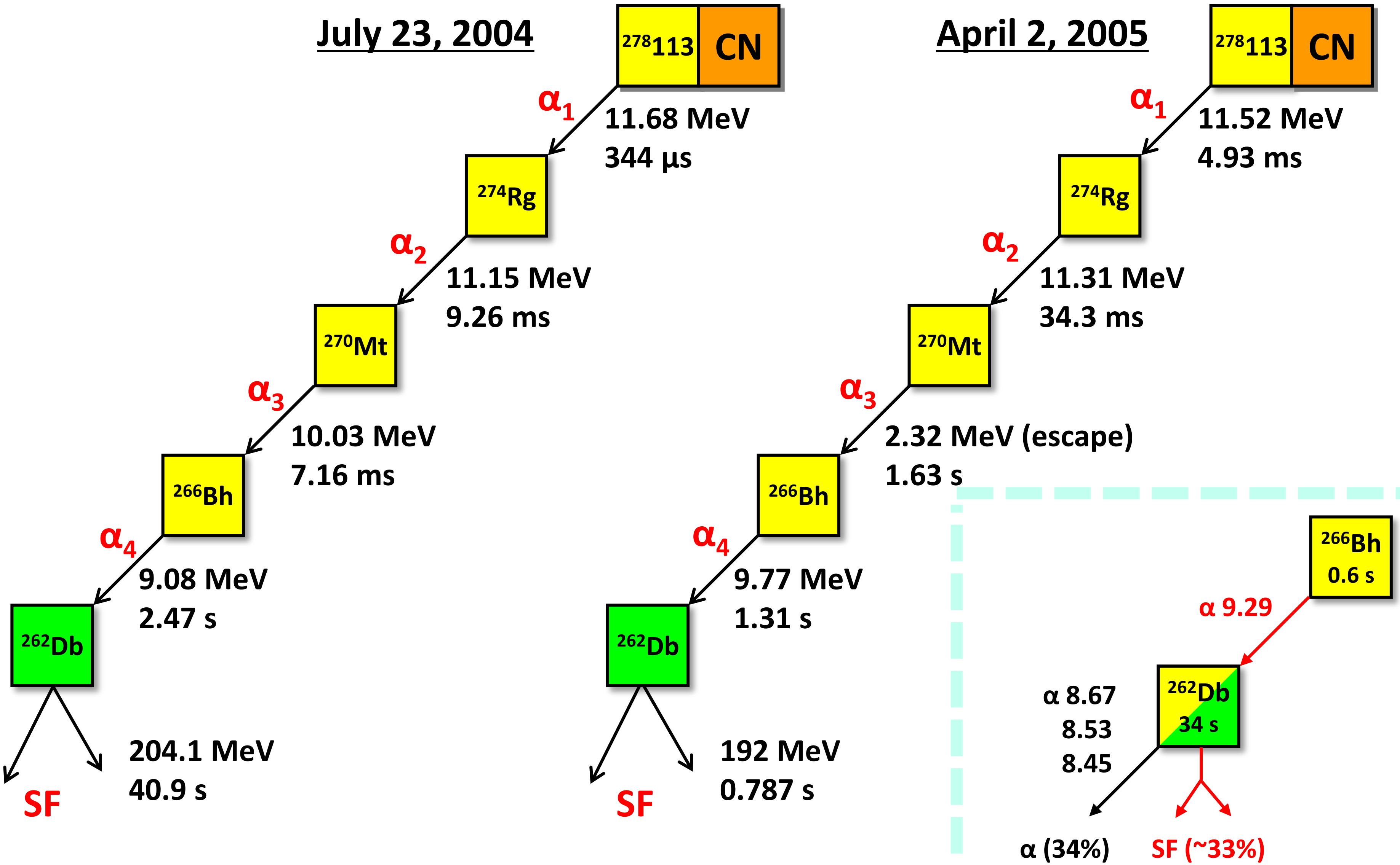
Experimental conditions

Reaction	$^{209}Bi(^{70}Zn,n)^{278}113$
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 (2.8×10^{12} s^{-1})
Beam integral	1.35×10^{20} (15 mg)
Target thickness	0.45 mg cm^{-2} (1.3×10^{18} cm^{-2})
GARIS eff.	80%
PSD + SSD eff.	94%

Summary of the element 113 experiment

Experimental period		Irradiation	Beam integral	No. of events
Year	Date (month/day)	[d]	[$\times 10^{19}$]	
2003	9/5 - 12/29	57.9	1.24/1.24	0
2004	7/8 - 8/2	21.9	0.51/1.75	1
2005	1/20 - 1/23	3.0	0.07/1.82	0
2005	3/20 - 4/22	27.1	0.71/2.53	1
2005	5/19 - 5/21	2.0	0.05/2.58	0
2005	8/7 - 8/25	16.1	0.45/3.03	0
2005	9/7 - 10/20	39.0	1.17/4.20	0
2005	11/25 - 12/15	19.5	0.63/4.83	0
2006	3/14 - 5/15	54.2	1.37/6.20	0
2008	1/9 - 3/31	70.9	2.28/8.48	0
2010	9/7 - 10/18	30.9	0.52/9.00	0
2011	1/22 - 5/22	89.8	2.01/11.01	0
2011	12/2 - 12/19	14.4	0.33/11.34	0
2012	1/15 - 2/9	25.0	0.56/11.90	0
2012	3/13 - 4/17	33.7	0.79/12.69	0
2012	6/12 - 7/2	15.7	0.25/12.94	0
2012	7/14 - 8/18	32.0	0.57/13.51	1
Total		553	13.5	3

Observation of $^{278}113$



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

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

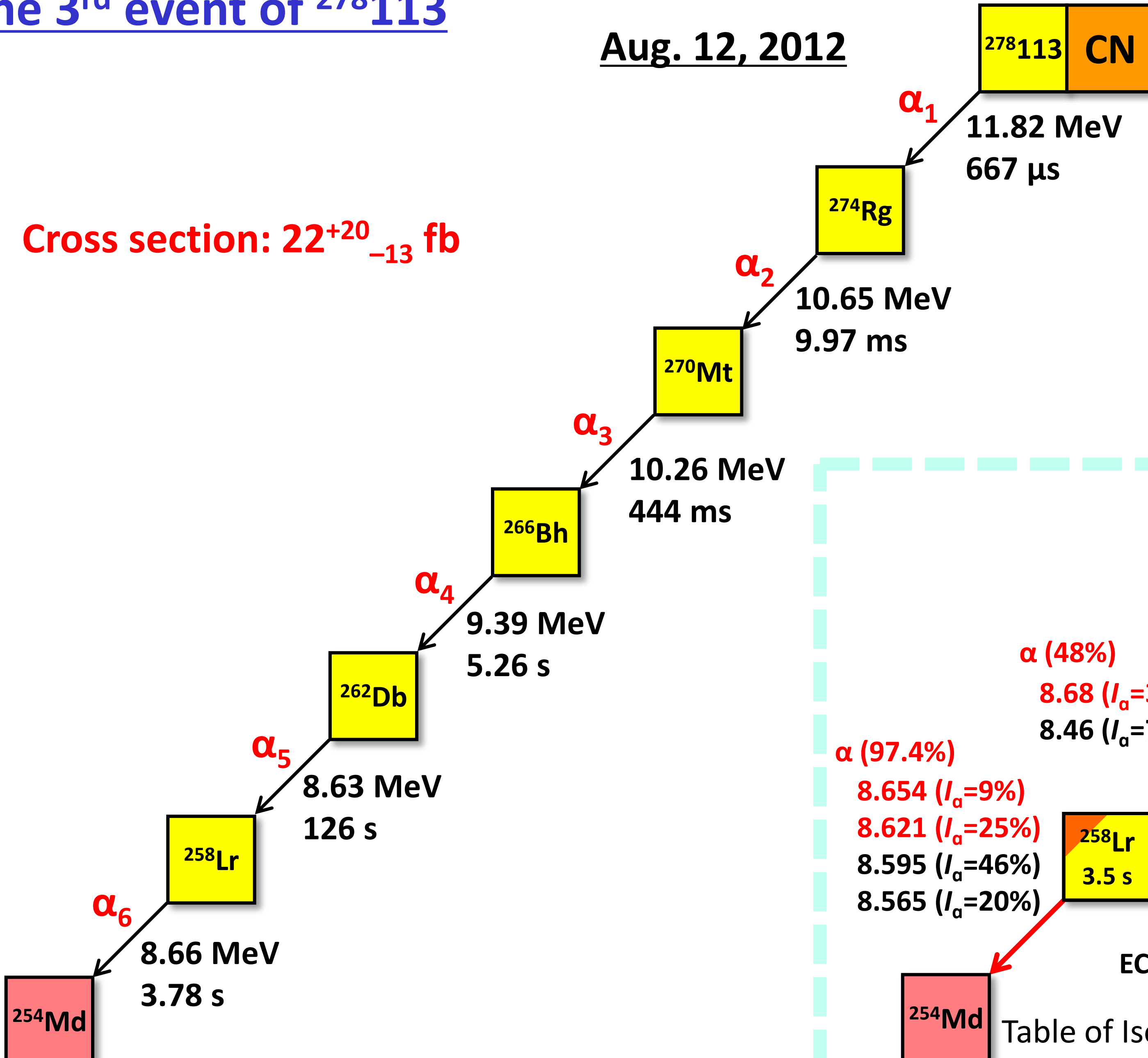
Wilk et al., PRL **85**, 2697 (2000).

Table of Isotopes, 8th ed. (1996).

The 3rd event of $^{278}113$

Aug. 12, 2012

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



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

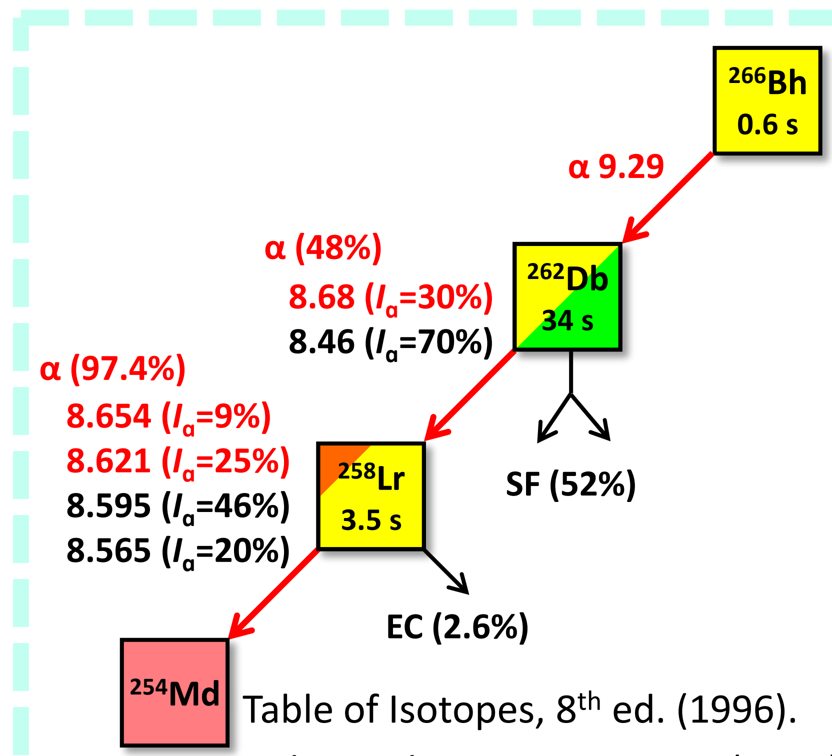
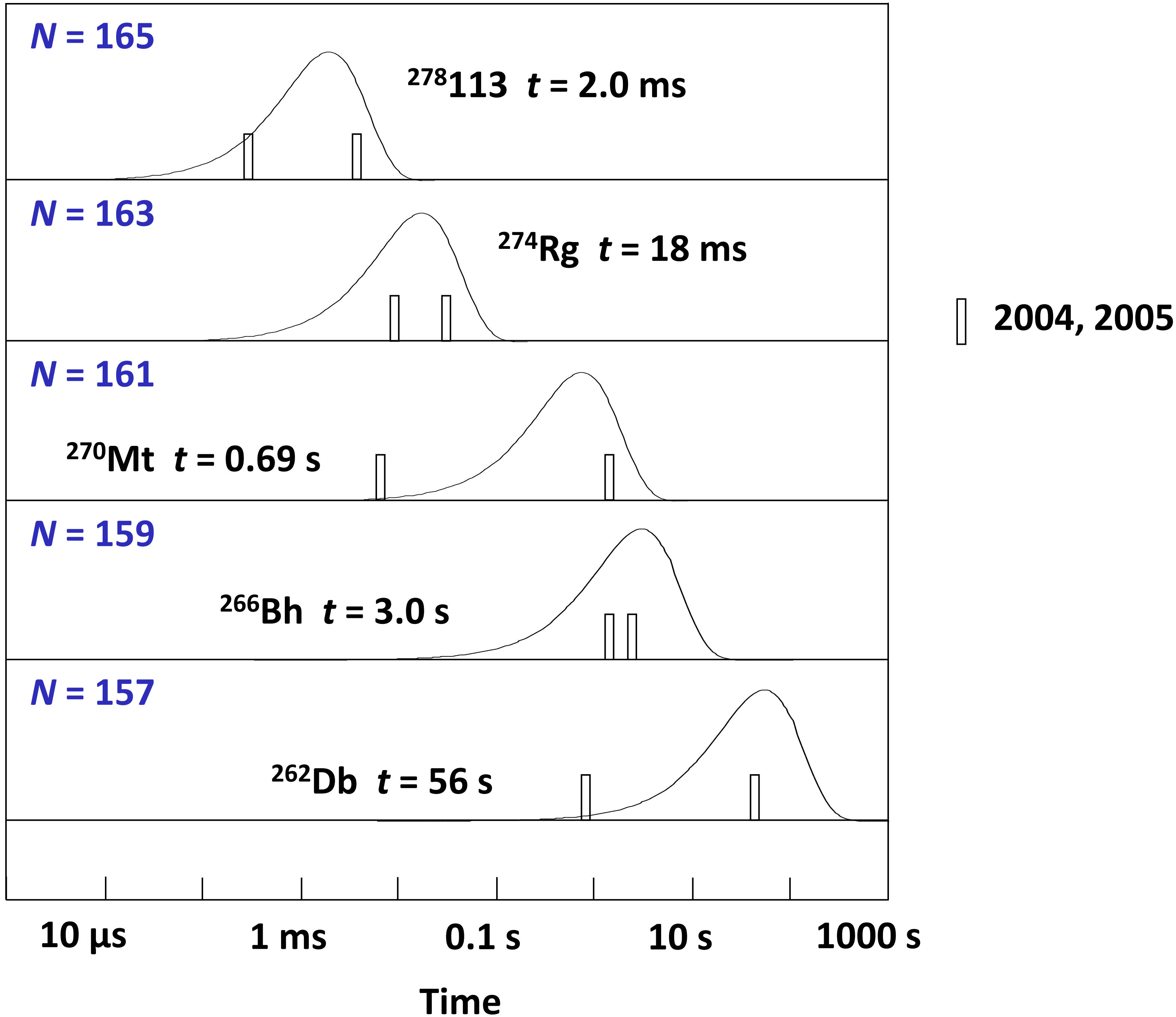
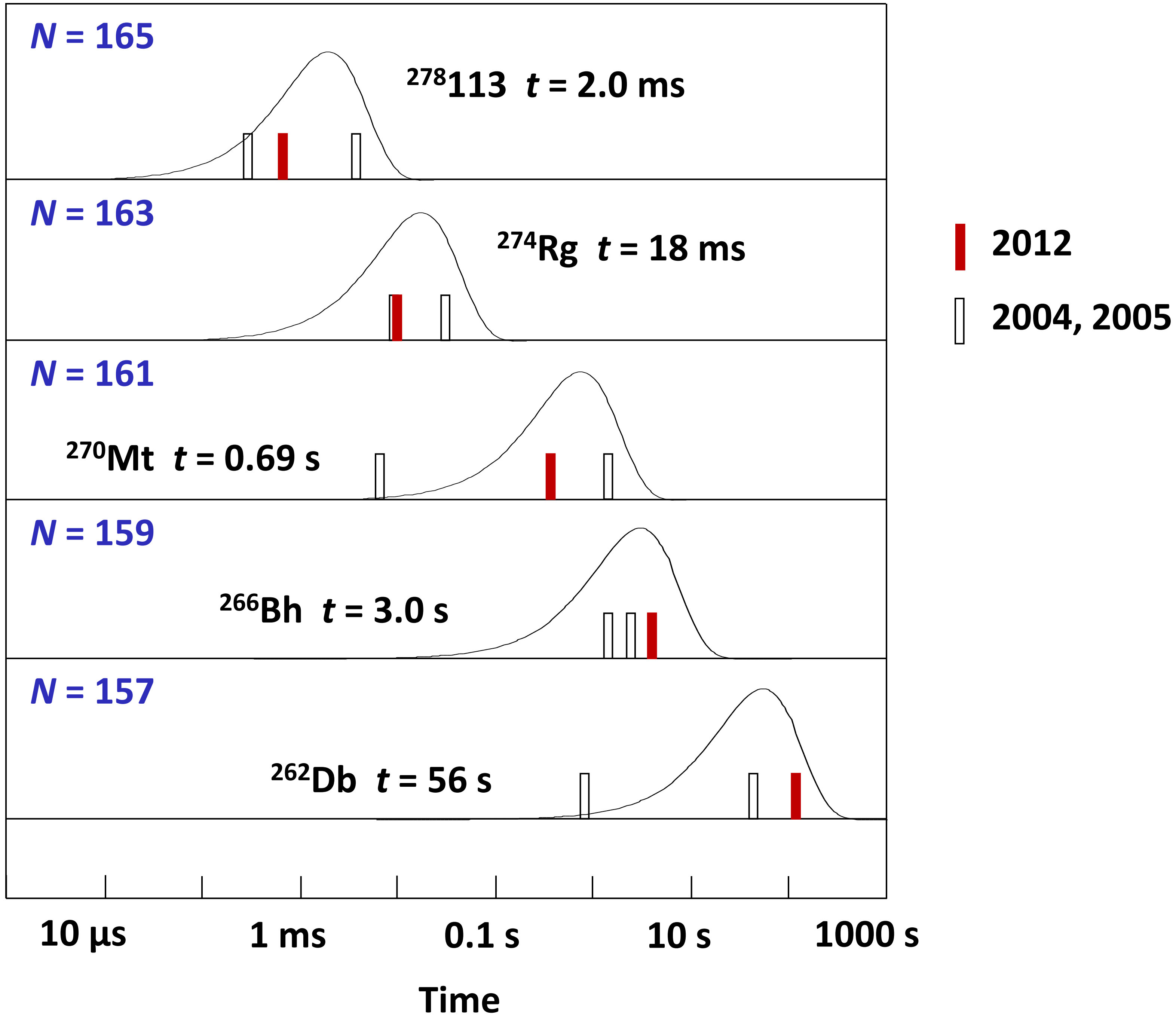


Table of Isotopes, 8th ed. (1996).

Haba et al., PRC **89**, 024618 (2014).



N = 165
278113 t = 2.0 ms
N = 163
274Rg t = 18 ms
N = 161
270Mt t = 0.69 s
N = 159
266Bh t = 3.0 s
N = 157
262Db t = 56 s
2004, 2005
10 μs
1 ms
0.1 s
10 s
1000 s
Time



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2012
2004, 2005
10 µs
1 ms
0.1 s
10 s
1000 s
Time

Future plans

Chemistry using preseparated $^{261}Rf^{a}$, ^{262}Db, and $^{265}Sg^{a,b}$

- Aqueous chemistry by solvent extraction with LS
- Gas chemistry by direct complexation without aerosols
- $^{248}Cm(^{23}Na,4n)^{267}Bh$ (scheduled in 2014)

Syntheses of the heaviest SHEs

- $^{248}Cm(^{48}Ca,xn)^{296-x}Lv$ (in progress)
- $^{248}Cm(^{50}Ti,xn)^{298-x}118$ (scheduled in 2014)
- $^{248}Cm(^{51}V,xn)^{299-x}119$
- $^{248}Cm(^{54}Cr,xn)^{302-x}120$
- Commissioning of GARIS II (in progress)

High precision mass measurement of SHE nuclei ($\delta m/m$ ~ 0.5 ppm)

- GARIS II + RF-Carpet + MRTOF Spectrograph (scheduled in 2014)

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Thank you for your kind attention.