RIKEN GARIS as a promising interface for SHE chemistry

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CONTENTS

Periodic table of the elements (2012)

	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)

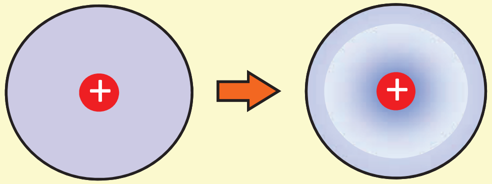
1. Chemistry of new elements

Frontiers in chemistry

- **Chemical properties, periodicity, and electronic structure of new elements? Relativistic effects on chemical reactions?**
- **On-line single-atom chemistry with accelerators**

Direct

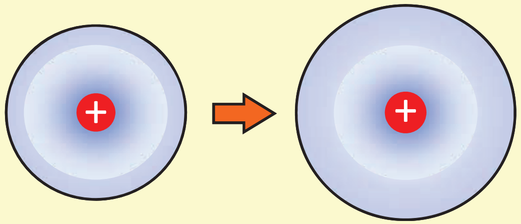
Contraction of inner orbital (*s*, *p*)



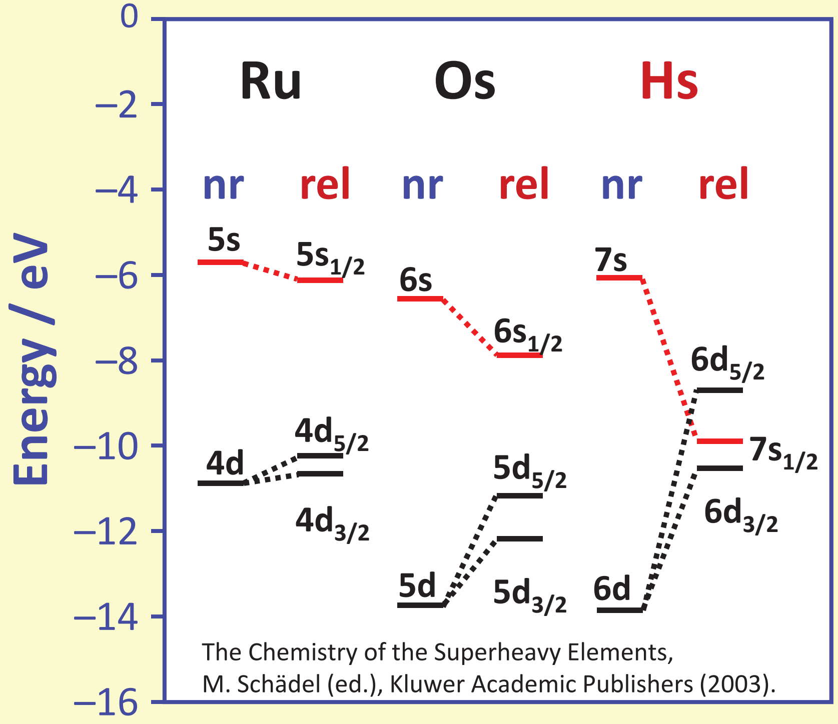
$$m = \frac{m_0}{\sqrt{1-(v/c)^2}} \qquad a_B = \frac{\hbar^2}{me^2} = a_B^0\sqrt{1-(v/c)^2}$$

Indirect

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



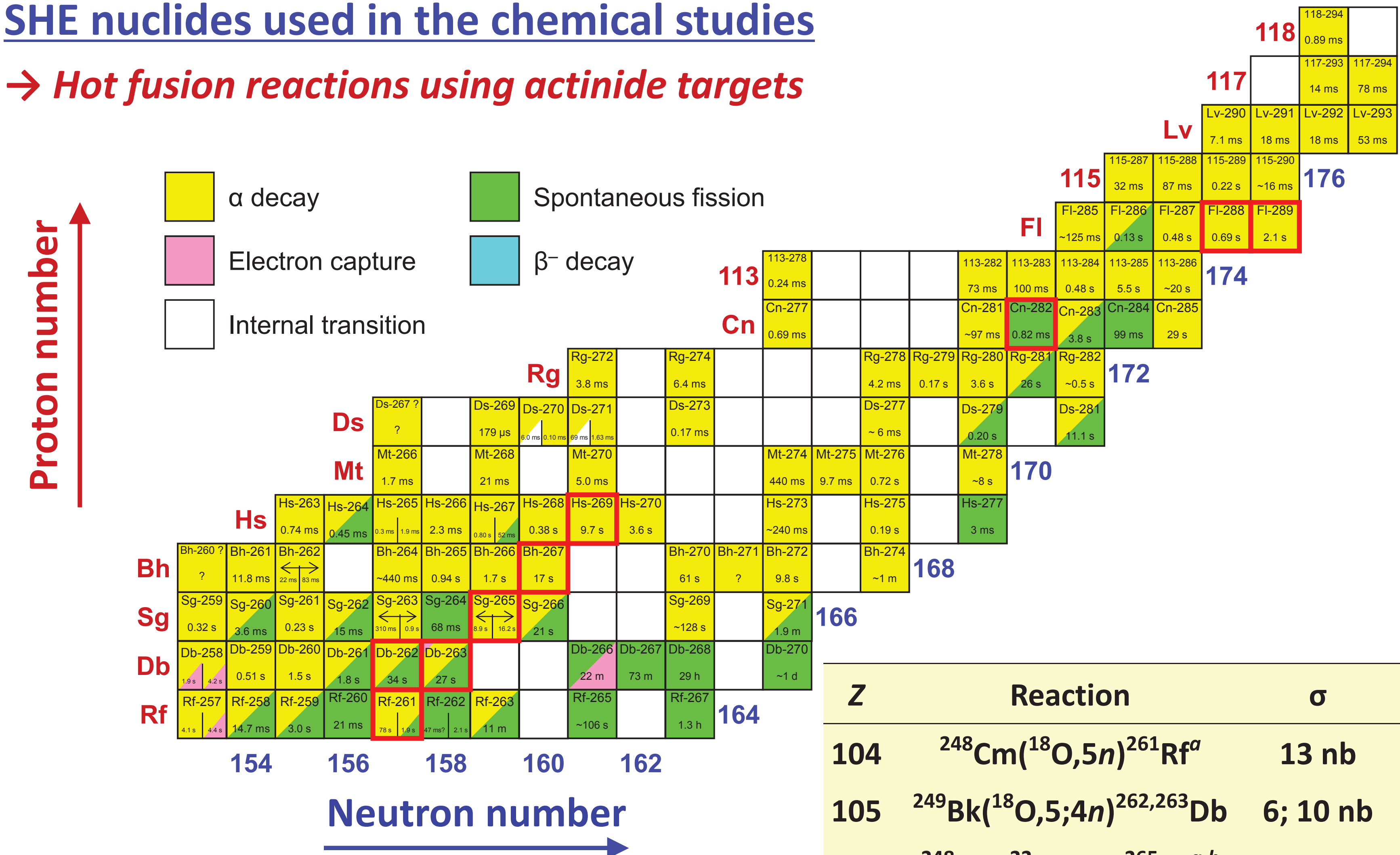
Energy levels of valence electrons



The Chemistry of the Superheavy Elements, M. Schädel (ed.), Kluwer Academic Publishers (2003).

SHE nuclides used in the chemical studies

→ Hot fusion reactions using actinide targets



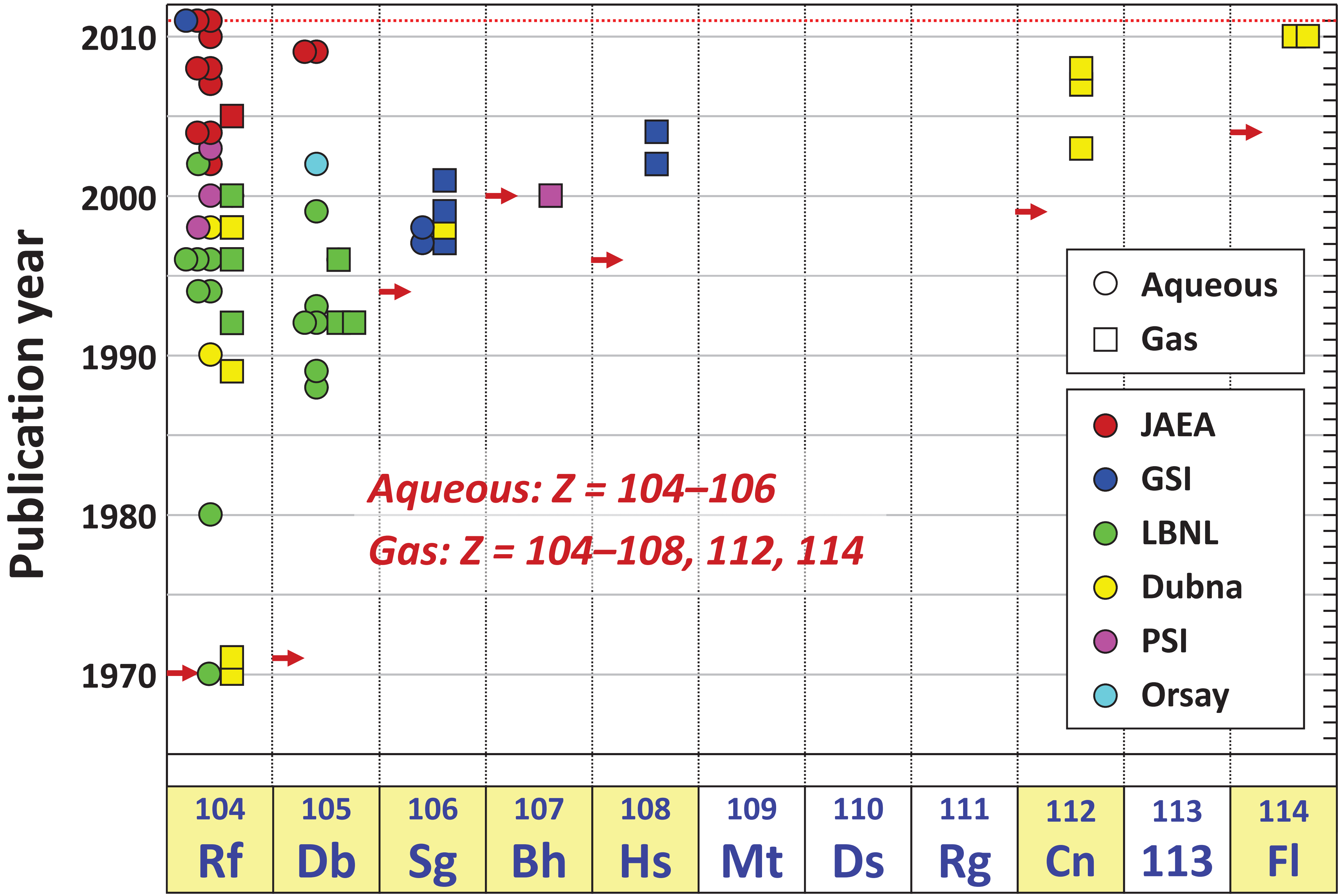
Single-atom chemistry

Small cross sections and short half-lives

→ Rapid and effective chemical experiments
with "single atoms" on-line at accelerators

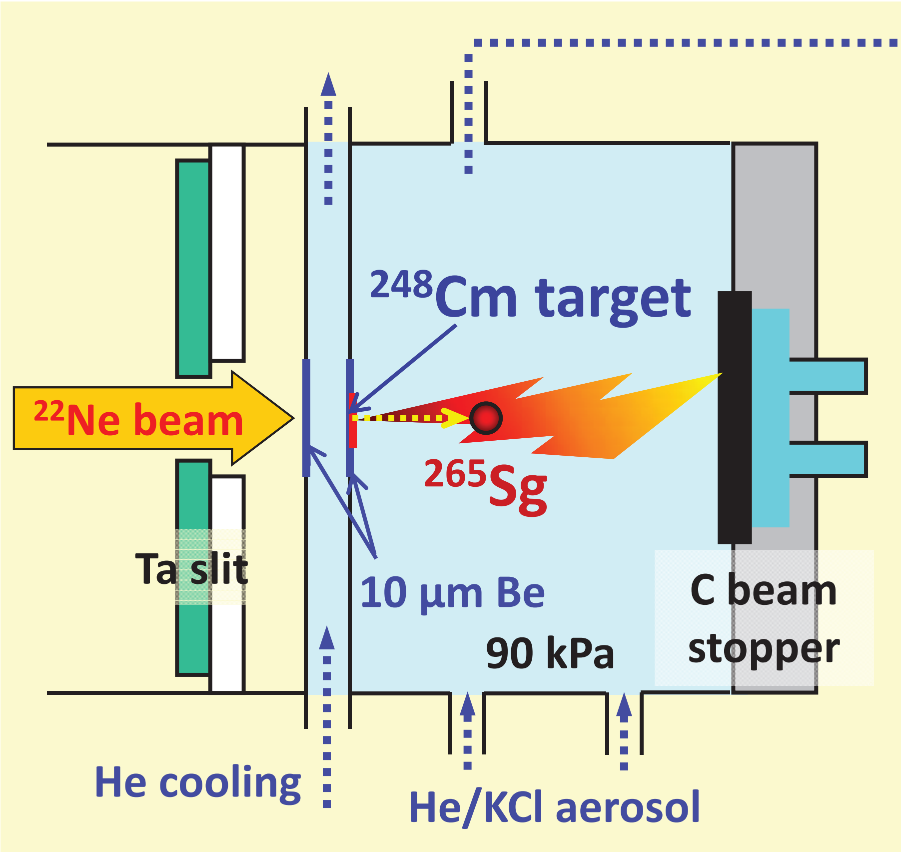
Z	Reaction	σ	$T_{1/2}$ (s)
104	$^{248}Cm(^{18}O,5n)^{261}Rf^{a}$	13 nb	68
105	$^{249}Bk(^{18}O,5;4n)^{262,263}Db$	6; 10 nb	34, 27
106	$^{248}Cm(^{22}Ne,5n)^{265}Sg^{a,b}$	380 pb	8.5, 14.4
107	$^{249}Bk(^{22}Ne,4n)^{267}Bh$	70 pb	17
108	$^{248}Cm(^{26}Mg,5n)^{269}Hs$	7 pb	9.7
112	$^{242}Pu(^{48}Ca,3n)^{287}Fl \rightarrow ^{283}Cn$	3.5 pb	3.8
114	$^{244}Pu(^{48}Ca,3;4n)^{288,289}Fl$	10; 8 pb	0.69, 2.1

Publications of Experimental Studies on SHE Chemistry



Conventional experimental procedure with gas-jet transport technique

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

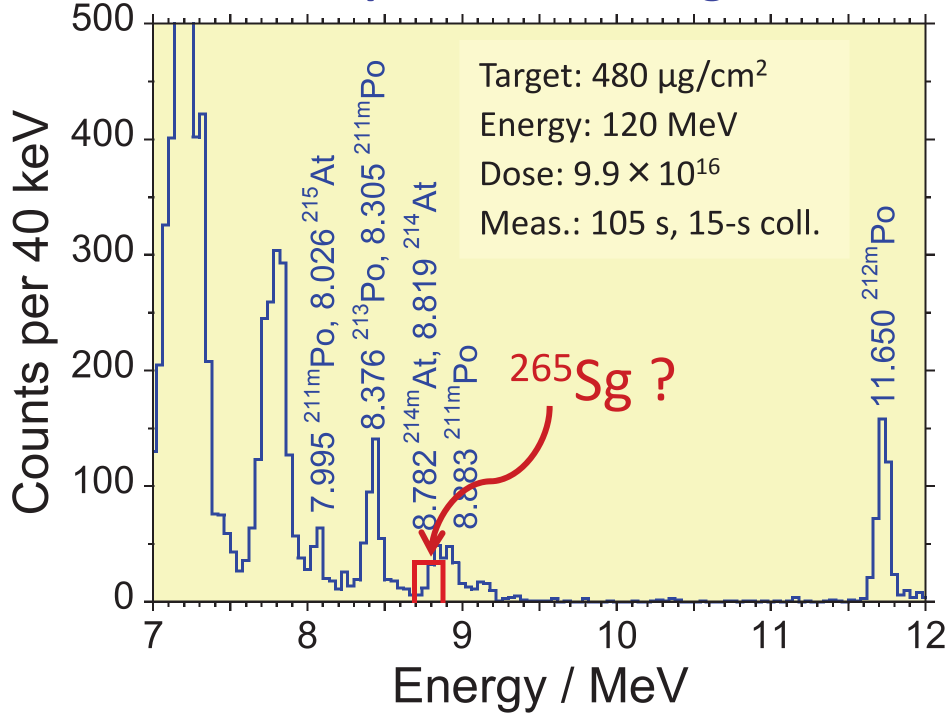


Chemistry apparatuses

Gas or liquid chromatography

→ α/SF spectrometry

α spectrum of ^{265}Sg

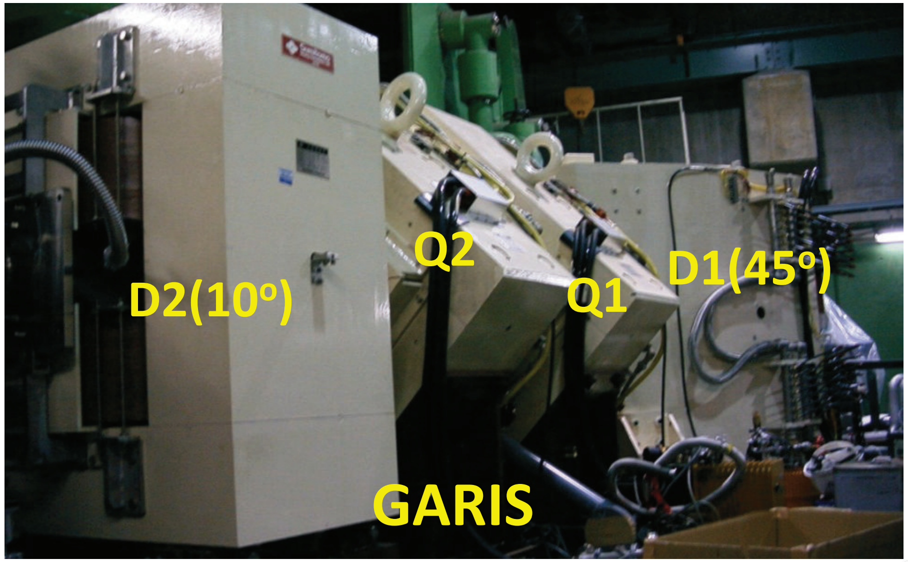


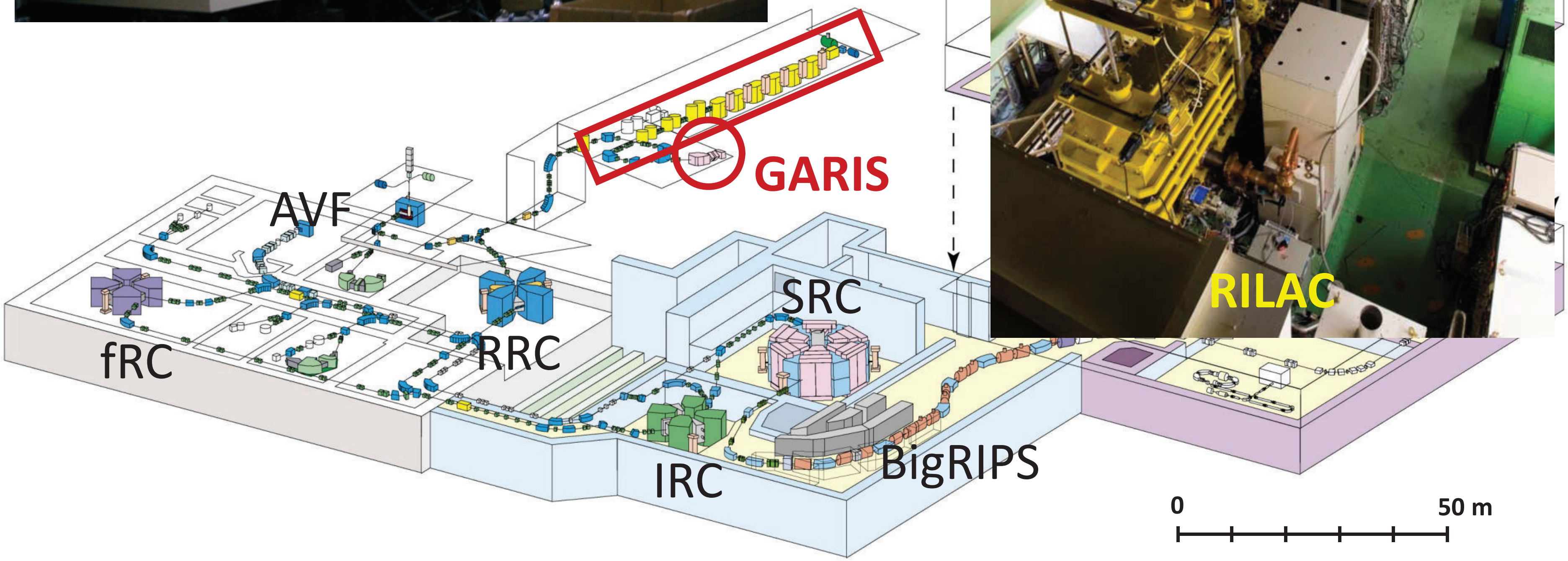
Limitations in the conventional method

- Background radioactivities from a large amount of by-products
- Decrease of gas-jet yields due to plasma condition induced by the beam

2. RIKEN GARIS as a pre-separator for SHE chemistry

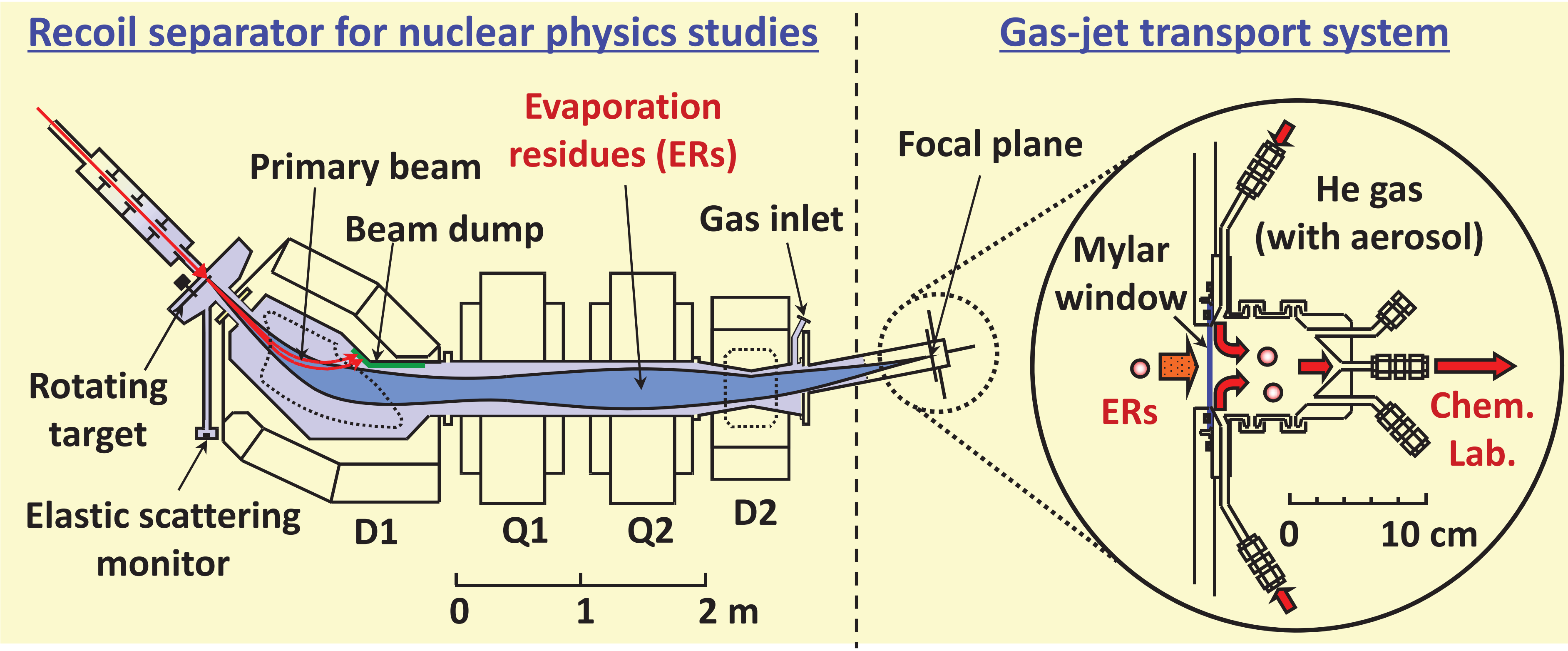
RIKEN RI Beam Factory





Breakthroughs in SHE chemistry

- Chemical and physical experiments under low background condition
- Stable and high gas-jet transport efficiency
- New chemical reaction systems that were not accessible before



- Development of a gas-jet transport system coupled to GARIS

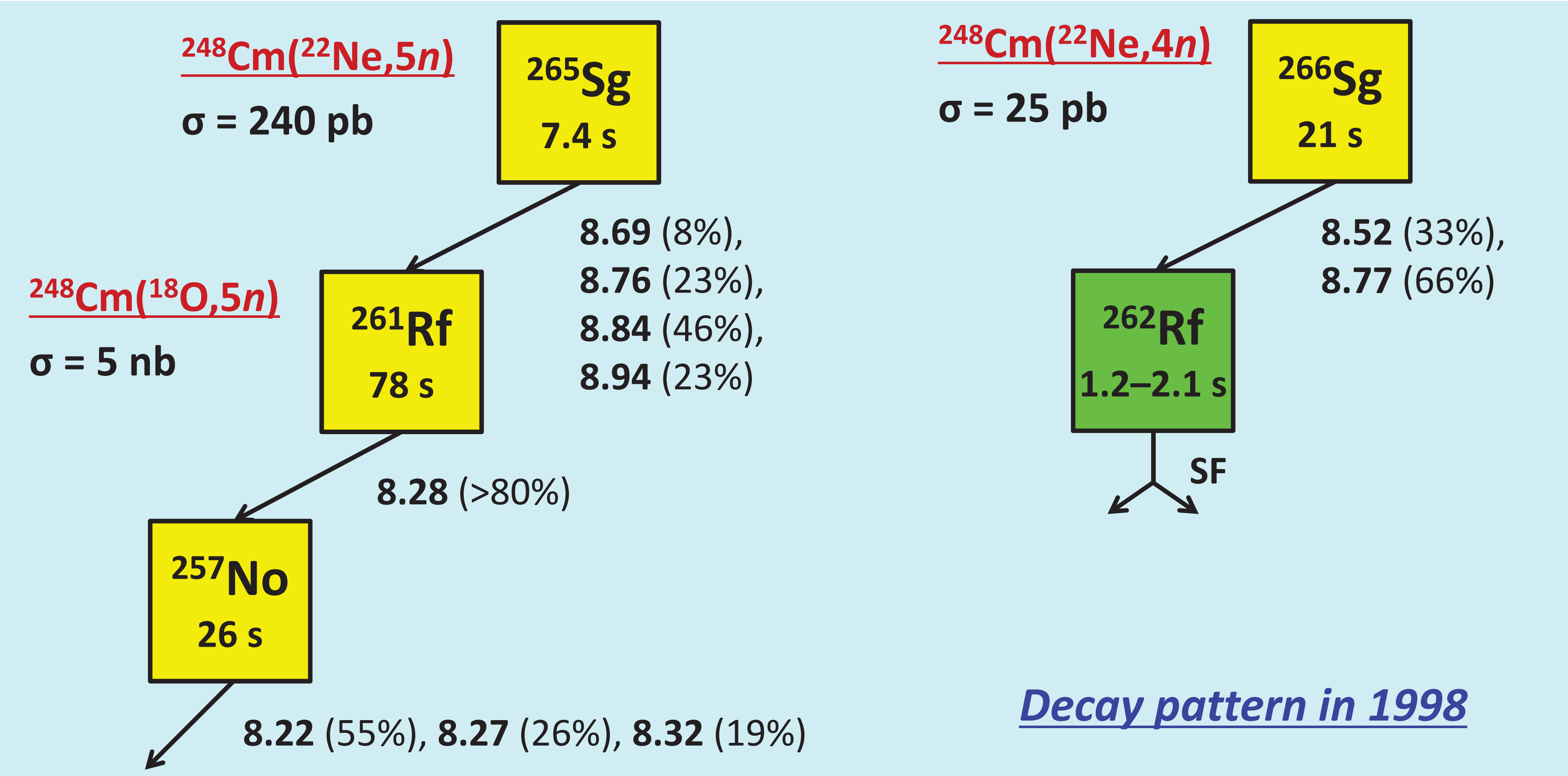
^{169}Tm(^{40}Ar,3n)^{206}Fr; ^{208}Pb(^{40}Ar,3n)^{245}Fm; ^{238}U(^{22}Ne,5n)^{255}No

- Production and decay studies of ^{261}Rf^{a,b} and ^{265}Sg^{a,b} for chemical studies

^{248}Cm(^{18}O,5n)^{261}Rf^{a,b}; ^{248}Cm(^{22}Ne,5n)^{265}Sg^{a,b}

Previous decay studies of ^{261}Rf and ^{265,266}Sg

- Ghiorso *et al.*, PL **32B**, 95 (1970).
  ^{248}Cm(^{18}O,5*n*)^{261}Rf: Gas-jet + Rotating wheel
- Lazarev *et al.*, PRL **73**, 624 (1994).
  ^{248}Cm(^{22}Ne,5;4*n*)$^{265;266}$Sg: DGFRS + FPD (6; 4 events)
- Türler *et al.*, PRC **57**, 1648 (1998).
  ^{248}Cm(^{22}Ne,5;4*n*)$^{265;266}$Sg: Gas-jet + OLGA + ROMA [13(–2.8 as N_R); 3 events]



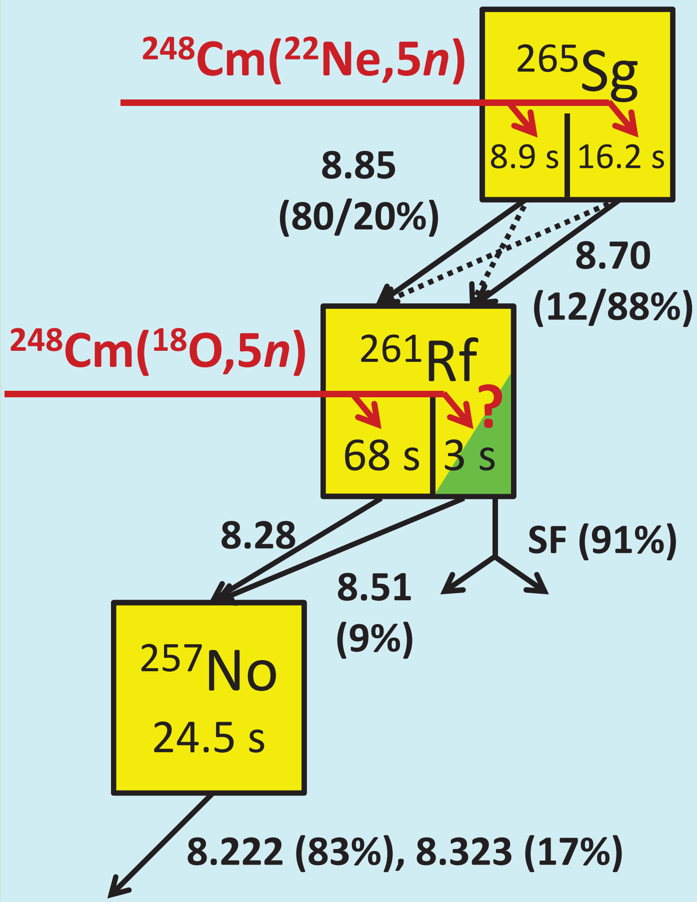
Düllmann and Türler: PRC **77**, 064320 (2008).

- ^{208}Pb(^{70}Zn,*n*)^{277}Cn: SHIP/GARIS + FPD (4 events)
- ^{248}Cm(^{26}Mg,5*n*)^{269}Hs: Gas-jet + COLD/CALLISTO/COMPACT (20 events)
- ^{248}Cm(^{22}Ne,5;4*n*)$^{265;266}$Sg: Reanalysis (36 events)

Refs. ^{248}Cm(^{22}Ne,5*n*)^{265}Sg	Method	E_{beam}	No of. events	σ [pb]
Lazarev *et al.* (1994)[a)]	DGFRS	116	4	80
		121	6	320
Gregorich *et al.* (1996)	MG	121	3	
Türler *et al.* (1998,1999)	OLGA	121/123	19	206
Türler *et al.* (1998)[b)]	PSI Tape	119	1	78
Dressler *et al.* (2000)[b)]	PSI Tape	116	1	73
Hübener *et al.* (2001)[b)]	HITGAS	119	2	92

a) τ(^{265}Sg) was not measured.

b) Only sensitive to α-SF chains



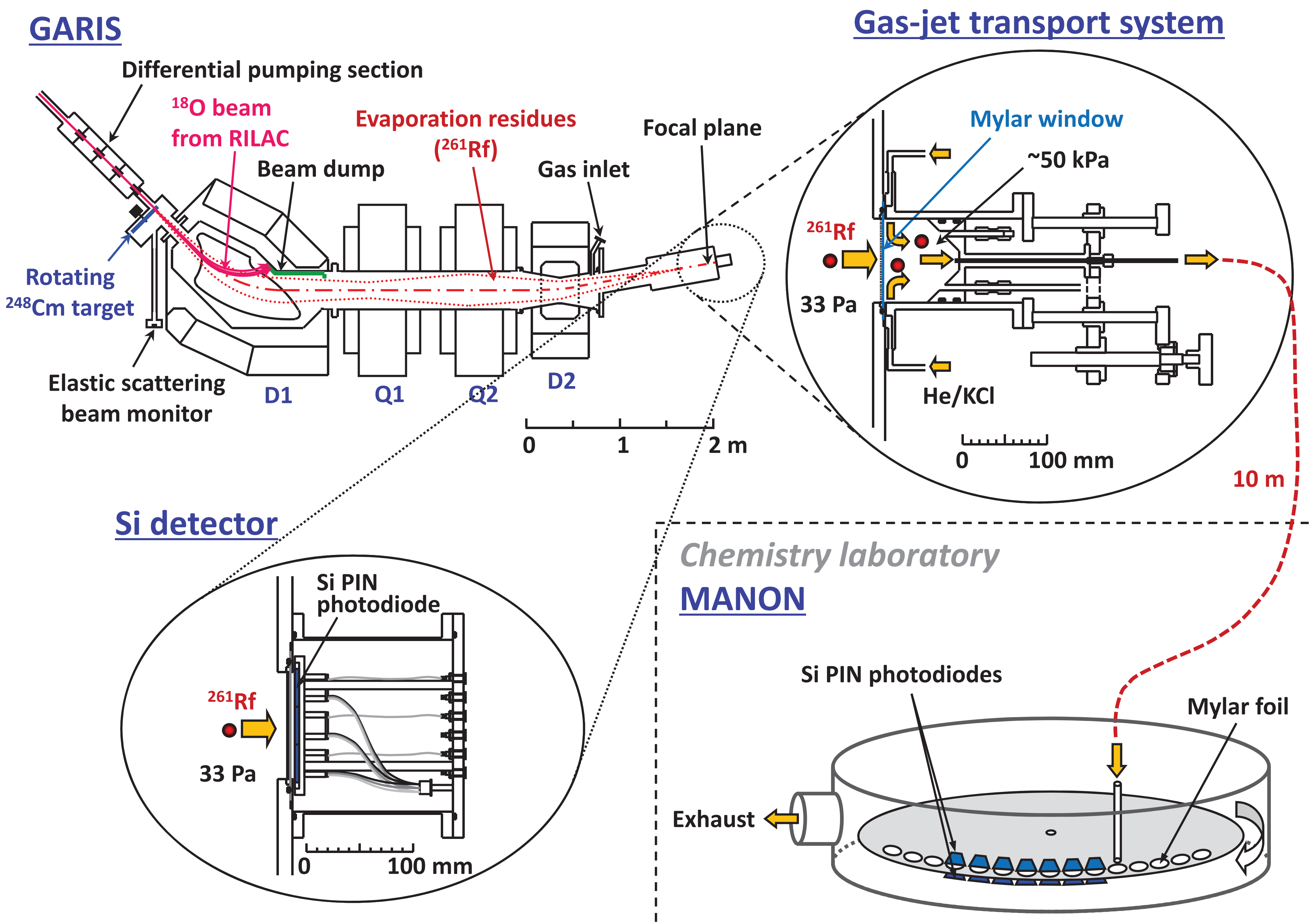
Decay pattern in 2008

For future chemical studies

→ Decay studies on ^{248}Cm(^{18}O,5*n*)^{261}Rf^{a,b}

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

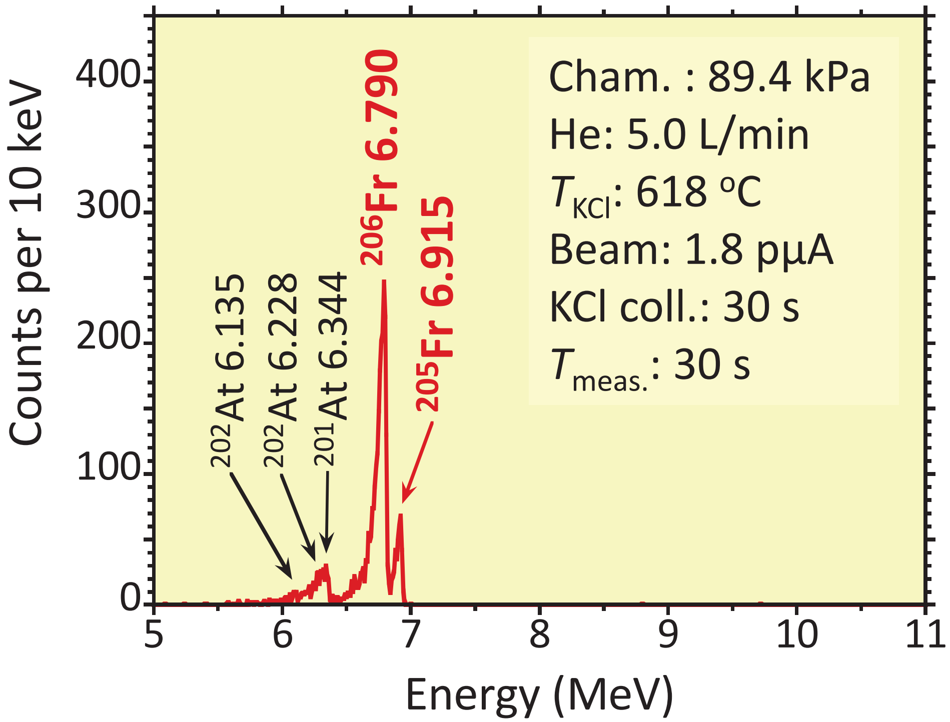
Experimental setup for $^{248}Cm(^{18}O,5n)^{261}Rf$



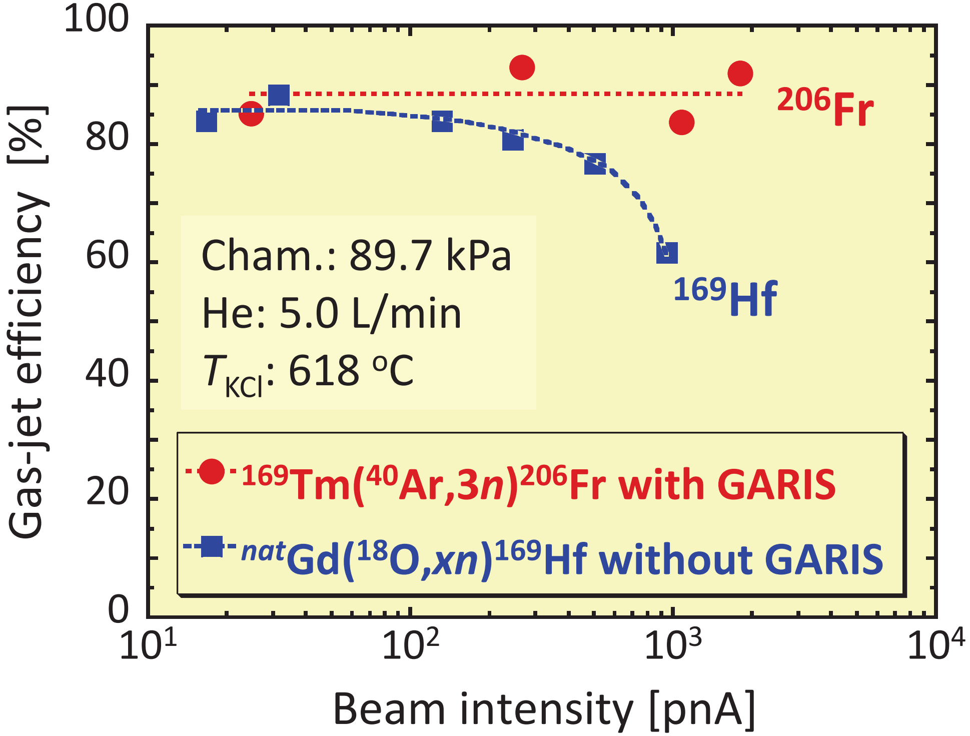
3. Performance of the GARIS/gas-jet system

(a) $^{169}Tm(^{40}Ar,3n)^{206}Fr$

α spectrum of ^{206}Fr



Gas-jet eff. vs. beam intensity

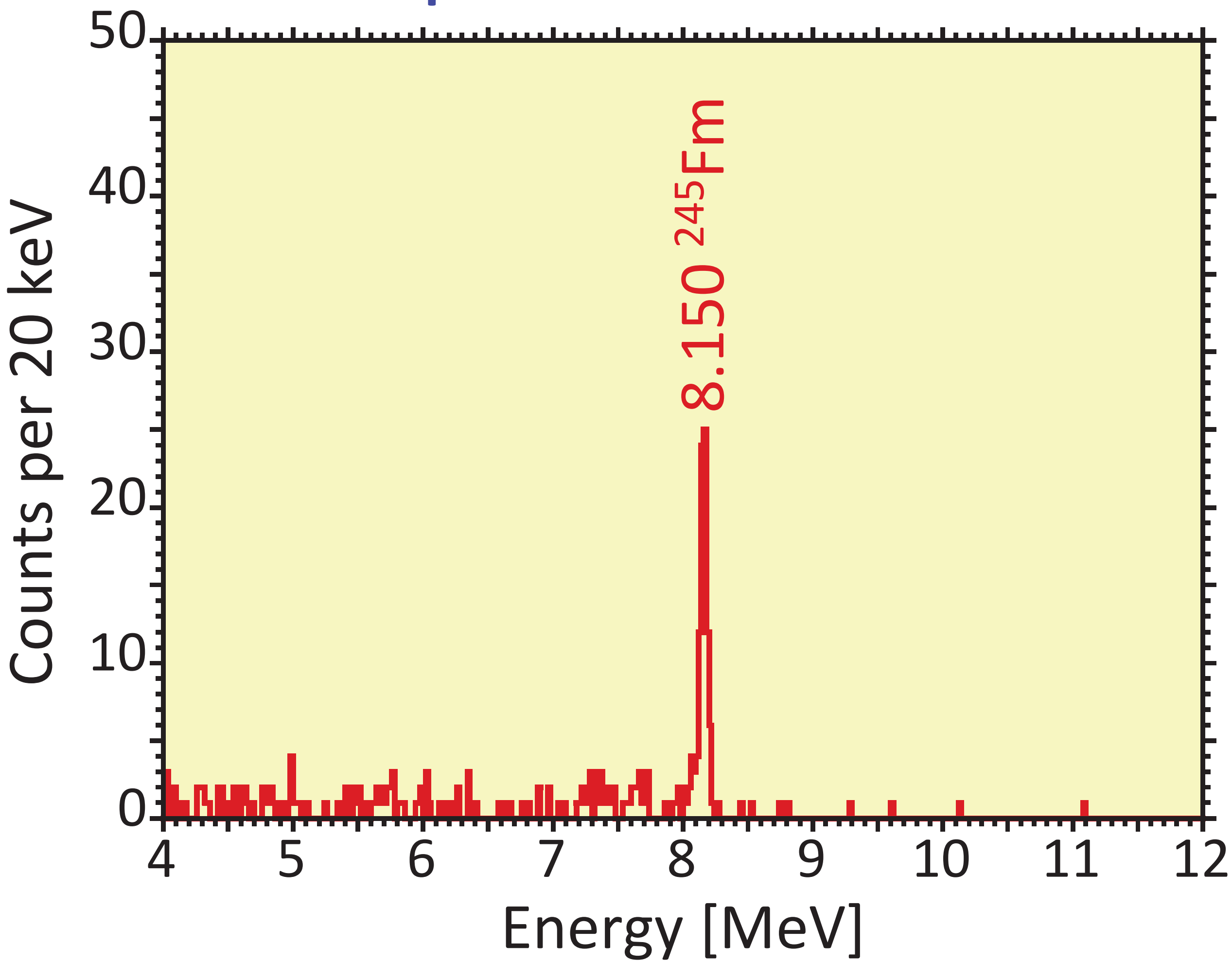


Haba *et al.*, JNRS **8**(2007)55.
Haba *et al.*, EPJD **45**(2007)81.

- Successful extraction of ^{206}Fr to MANON after the GARIS separation
- Gas-jet efficiency: 90%
- Beam-Independent gas-jet yield

(b) ^{208}Pb(^{40}Ar,3*n*)^{245}Fm

α spectrum of ^{245}Fm

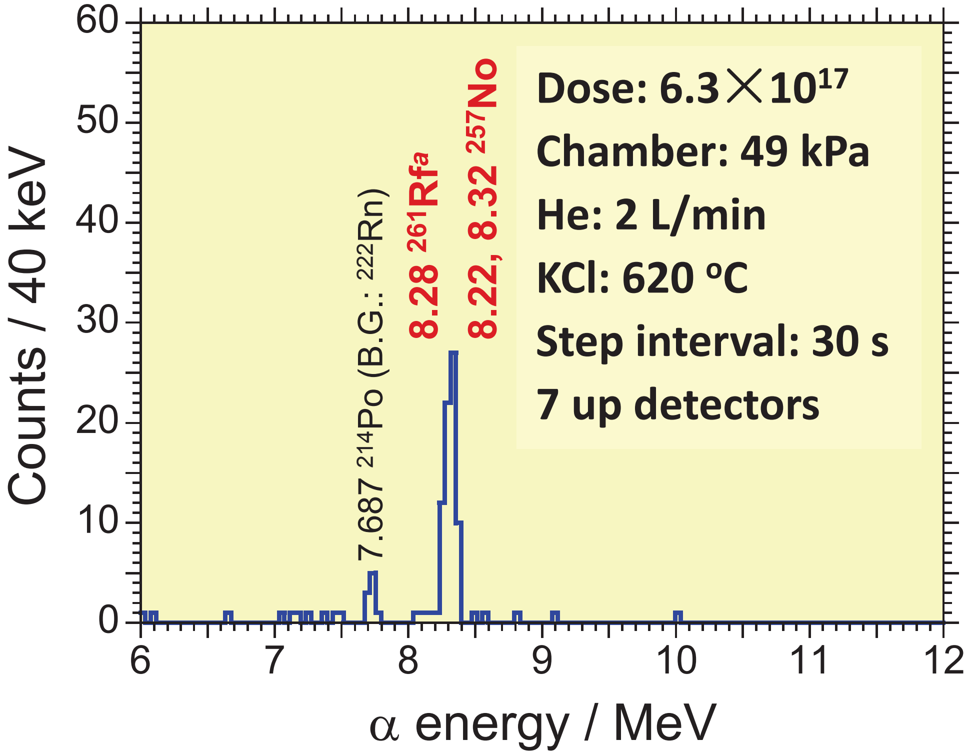


Target: 420 μg/cm^2
Beam dose: 9.5 × 10^{16}
Meas.: 1.9–16.1 s

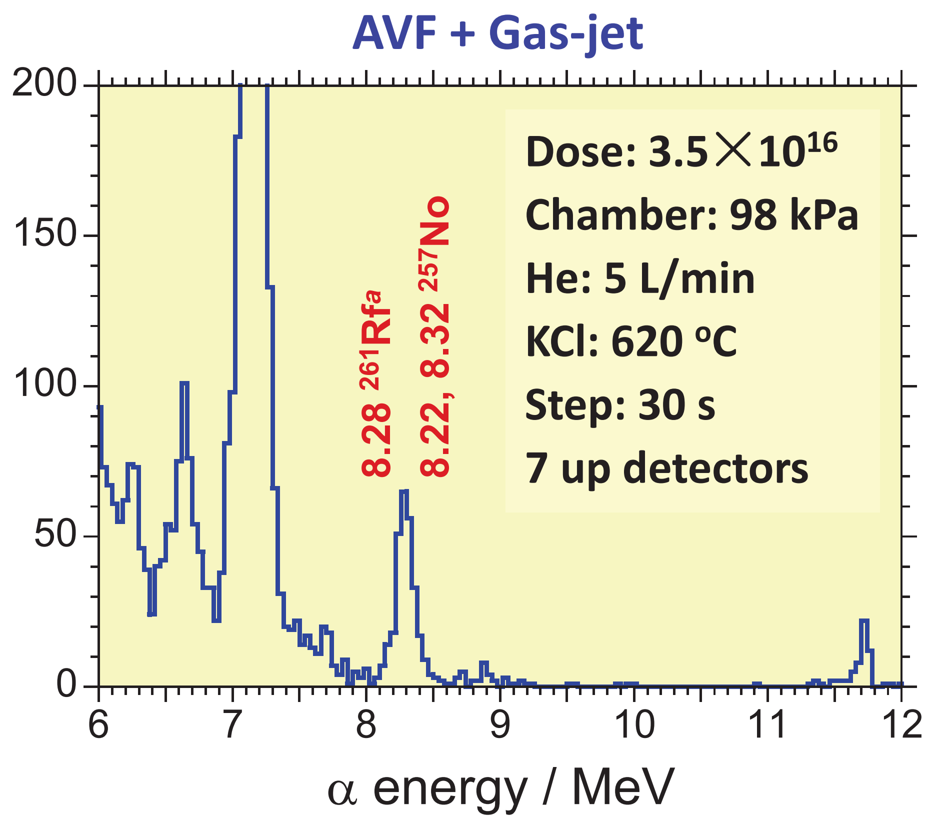
Haba *et al.*, JNRS **8**(2007)55.
Haba *et al.*, EPJD **45**(2007)81.

- **Separation factors for ^{211}Bi and ^{211m,212m}Po: > 10^4**
- **GARIS eff.: 43±4% for Φ60 mm (assumption: σ = 15 nb)**
- **Gas-jet eff.: 83±9%**

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

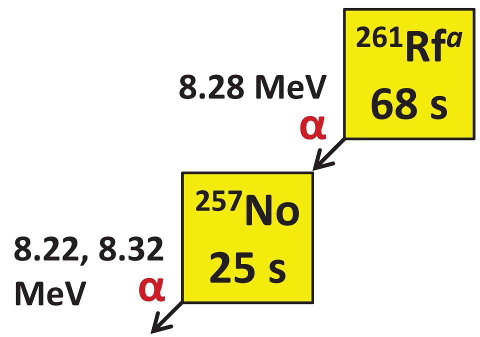


Haba *et al.*, Chem. Lett. **38**(2009)426.



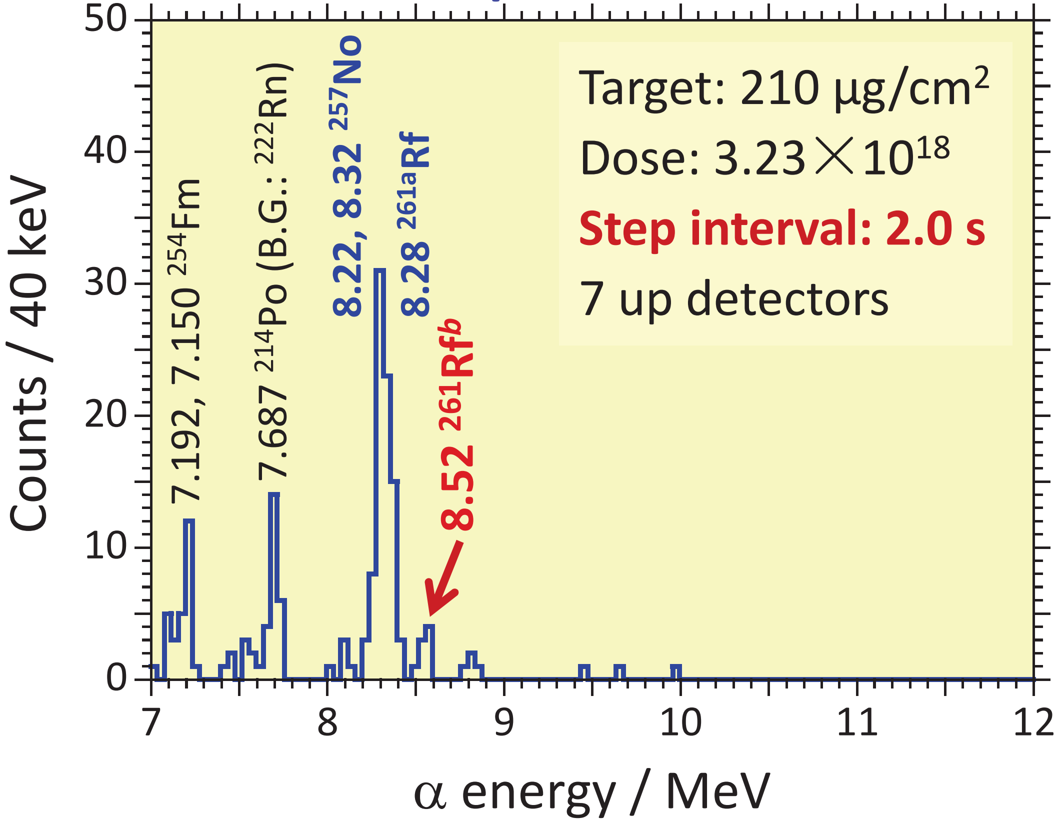
Haba *et al.*, EPJD **45**(2007)81.

- 161 α (58 α-α) / 8.2-h meas.
- GARIS eff.: 7.8±1.7% for Φ100 mm (σ = 13 nb@JAEA)
- Gas-jet eff.: 52±12%
- Yield of $^{261}Rf^{a}$ at the chemistry laboratory: ~0.5 atoms/min



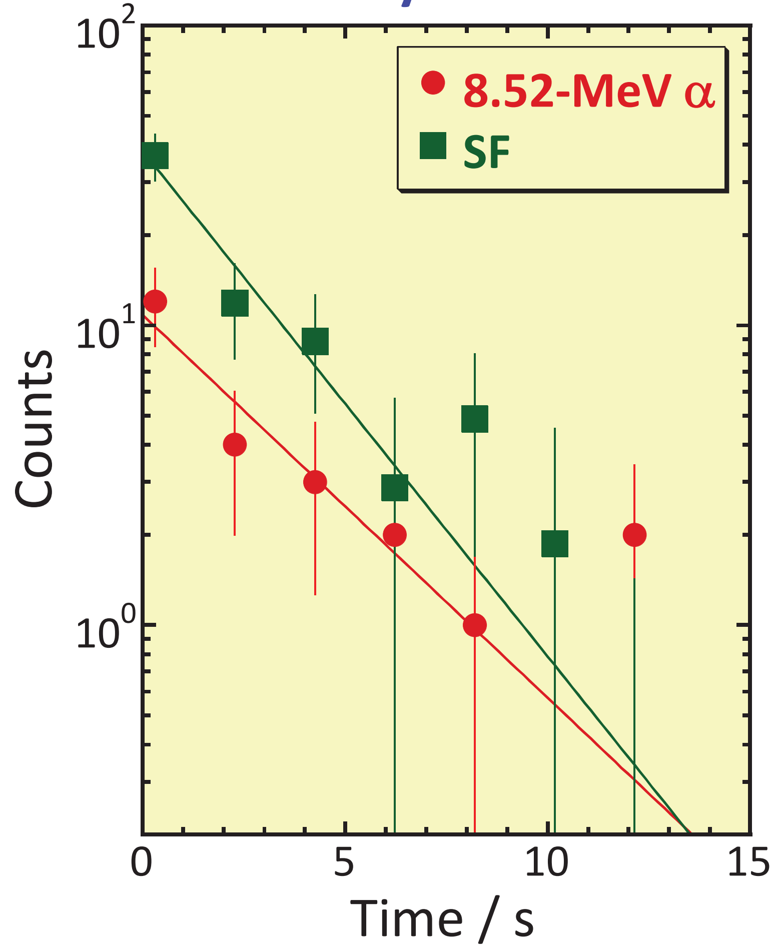
(d) $^{248}Cm(^{18}O,5n)^{261}Rf^{b}$

α spectrum



Haba *et al.*, PRC **83**(2011)034602.

Decay curves



Numbers of 8.52-MeV α and SF

Step [s]	No. of events 8.52-α	SF	Dose [×10^18]
30	0	13	1.2
2.0	**24**	**86**	2.8

$T_{1/2}$ of 8.52-MeV α and SF

	$T_{1/2}$ [s]
8.52-MeV α	2.4±0.8
SF	1.8±0.4

Correlated events on 8.52-MeV α

$E_{\alpha 2}$ = 8–10 MeV; E_{SF} > 30 MeV, ΔT_2 = 200 s

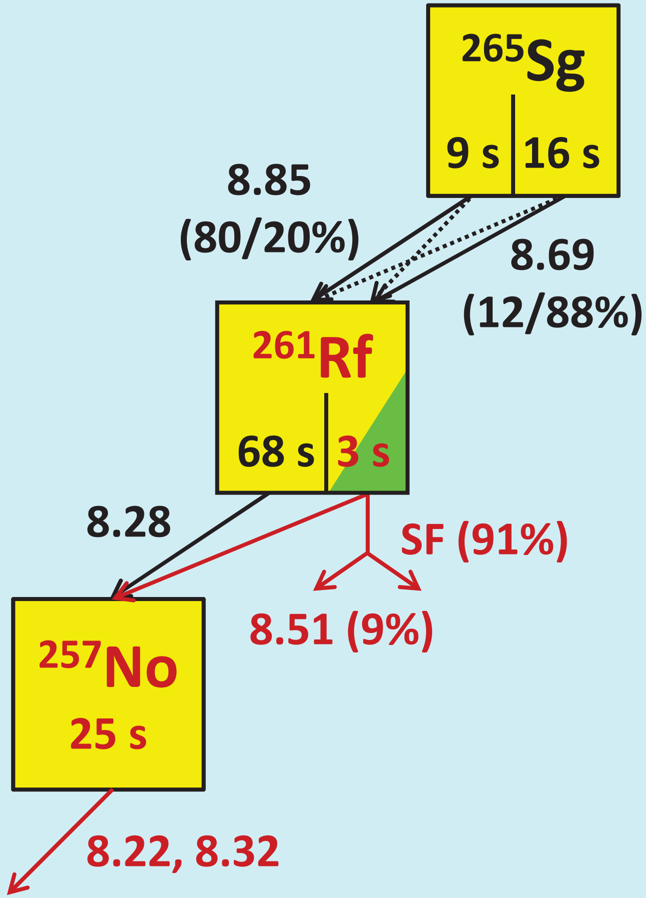
No.	8.52-MeV α		Correlated α	
	E_α (keV)	ΔT (s)	E_α (keV)	ΔT (s)
1	8.47	4.77	8.29	0.59
2	8.54	1.39	8.31	34.60
3	8.45	2.66	8.23	34.01
4	8.54	6.56	8.22	8.31
5	8.44	0.58	8.18	31.96
6	8.51	4.15	8.23	77.31

Random: <0.58

Correlated α

E_α = 8.18–8.31 MeV

$T_{1/2} = 22^{+14}_{-6}$ s



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

Confirmation of $^{261}Rf^b$ by $^{248}Cm(^{18}O,5n)^{261}Rf^b$

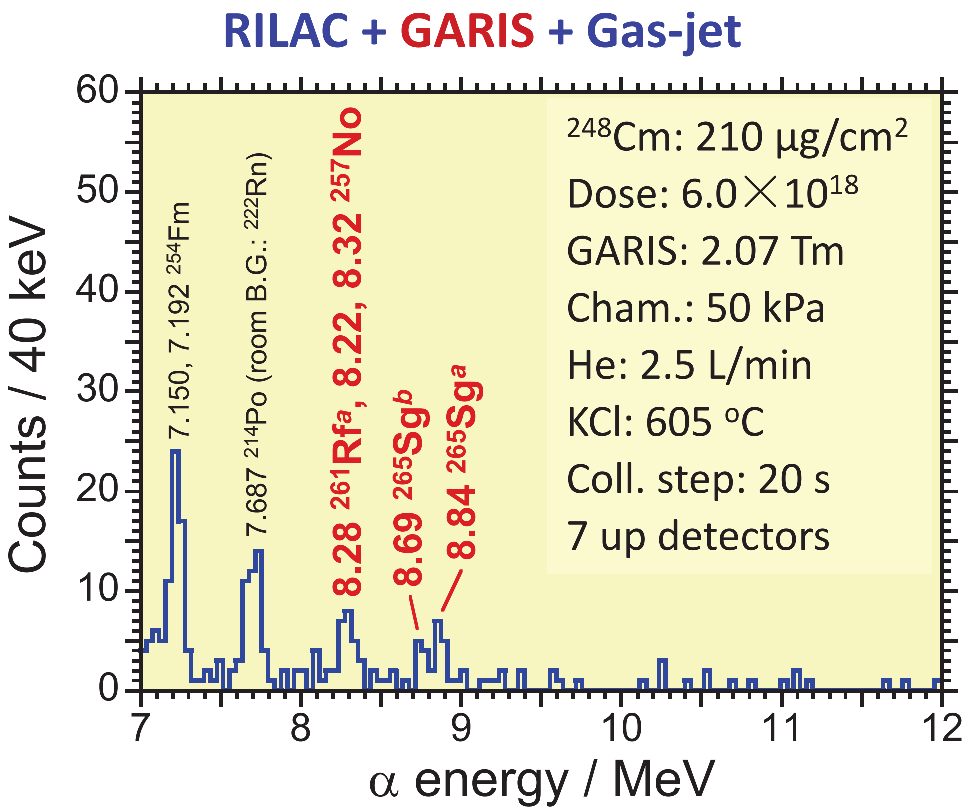
- **E_α = 8.52 ± 0.05 MeV**
- **$T_{1/2}$ = 1.9 ± 0.4 s**
- **Branching ratio (SF/α) = 73 ± 6%/27 ± 6%**
- **σ = 11 ± 2 nb at 95.1 MeV**

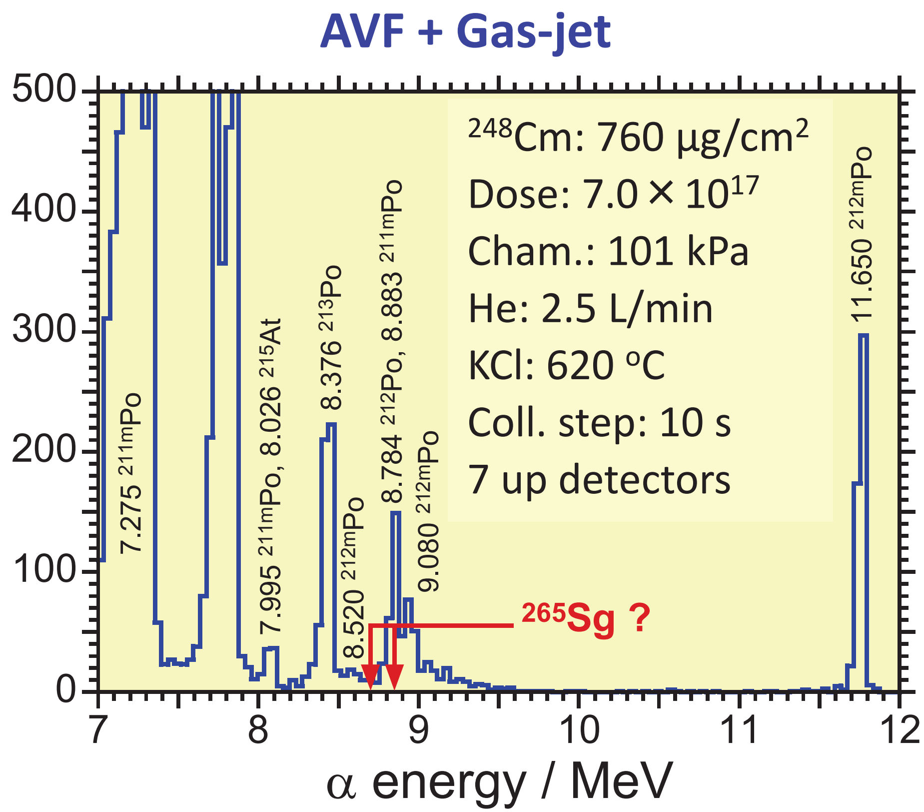
  assumptions: σ($^{261}Rf^a$) = 12 nb@JAEA

  → σ ratio($^{261}Rf^a$/$^{261}Rf^b$) = 1.1 ± 0.2

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

Beam time	Magnetic rigidity (Tm)	Beam dose (× 10^{18})
Sep. 30–Oct. 6, 2008	1.73	2.07
	1.94	1.91
	2.16	1.57
	2.04	0.639
Sep. 19–23, 2009; July 16–20, 2010	2.07	11.2





Haba *et al.*, PRC **85**(2012)024611.

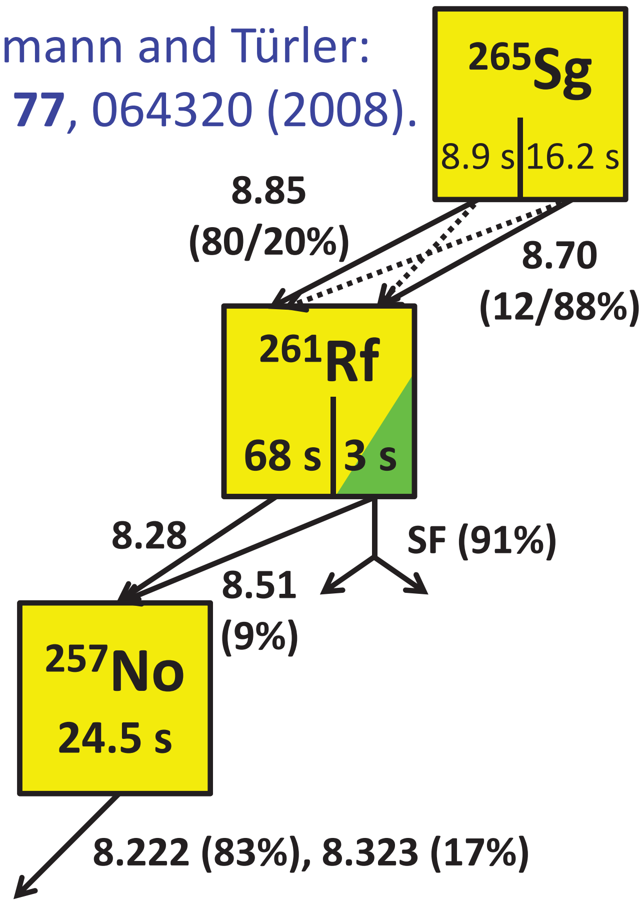
Search for α-α/α-SF correlations

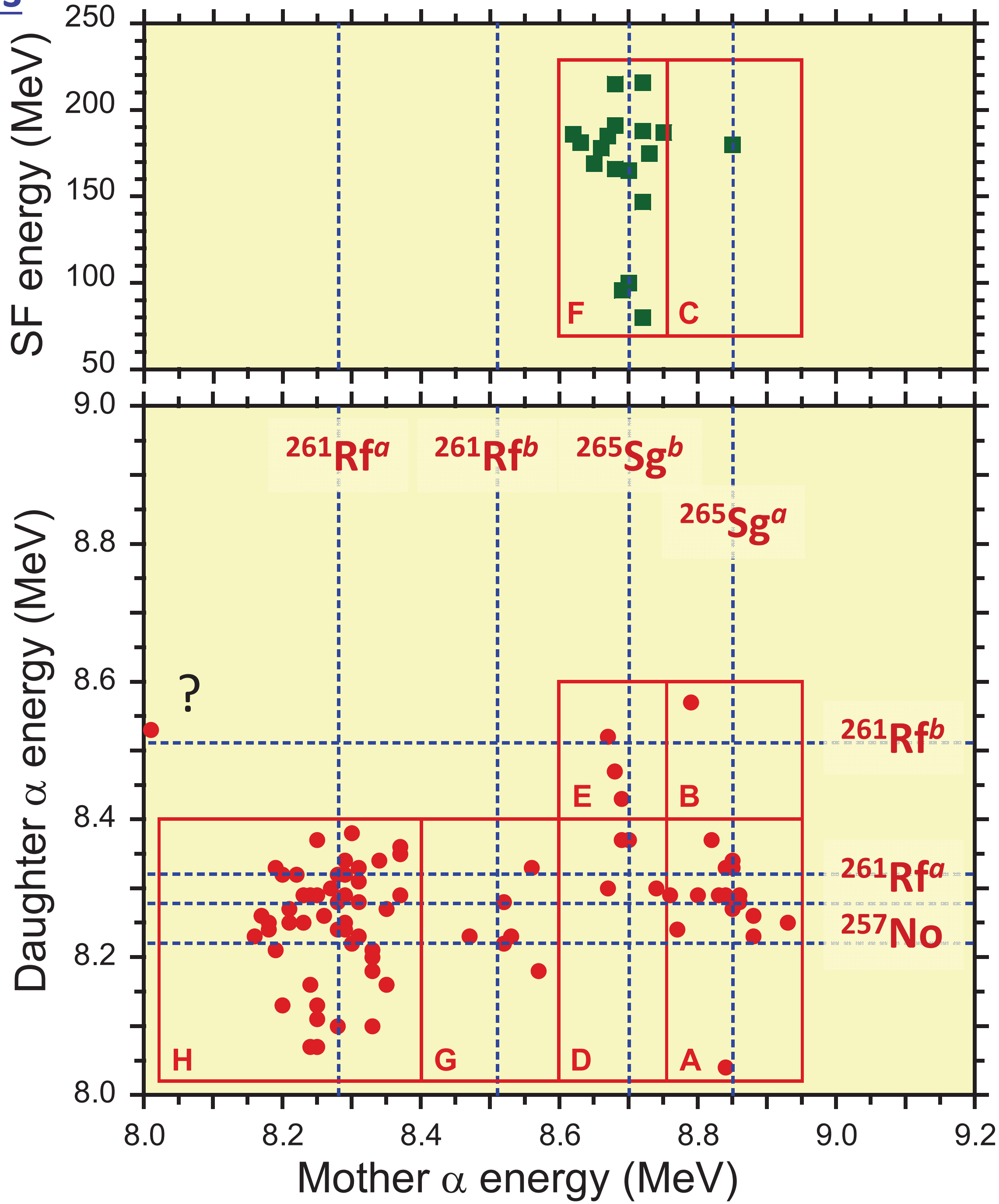
E_{α} = 8.0–9.0 MeV; $E_{SF} \geq 30$ MeV

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).

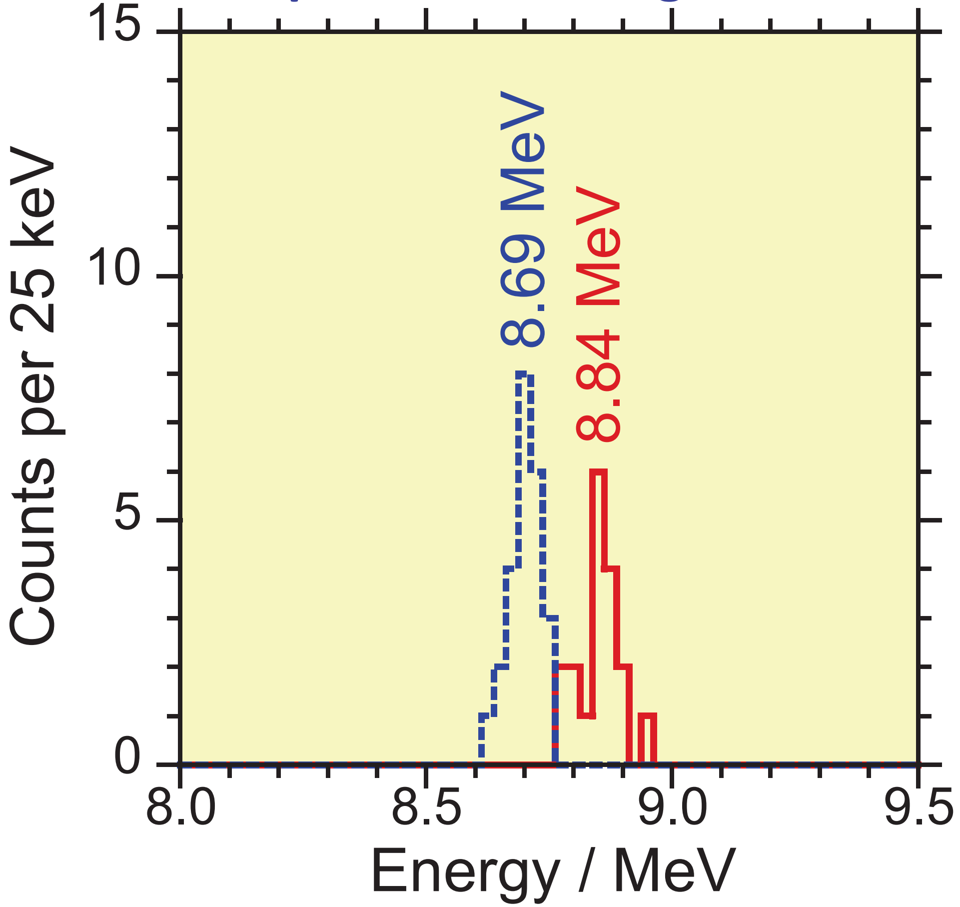




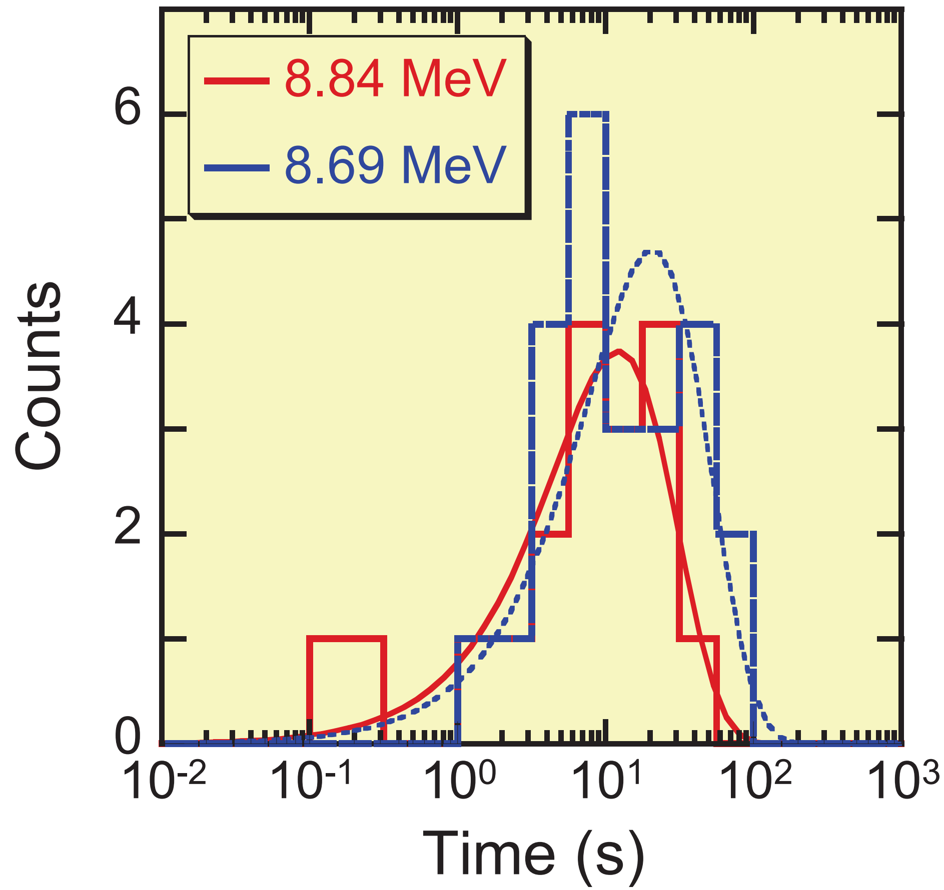
α energy and half-life of $^{265}Sg^{a,b}$

	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 $^{265}Sg^{a,b}$

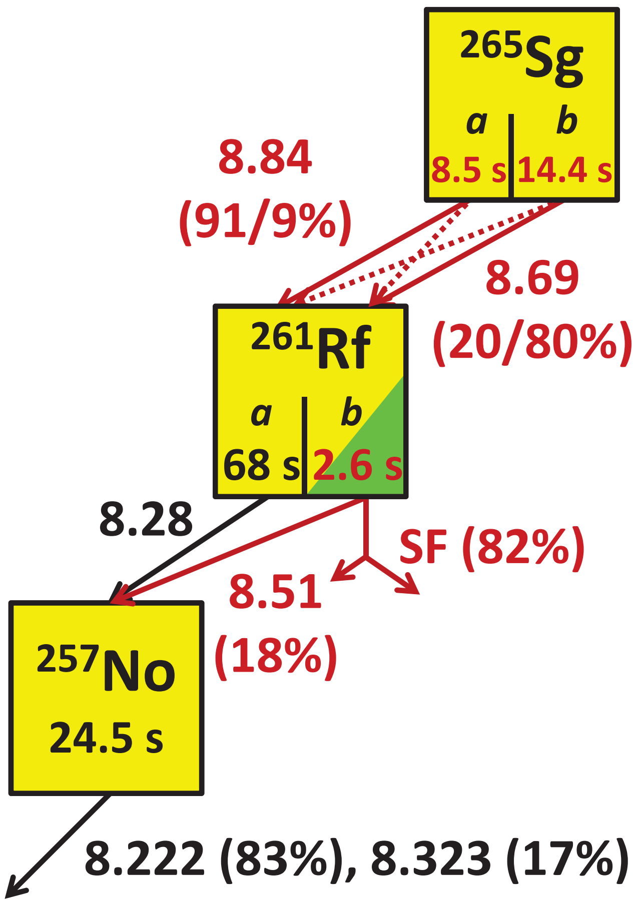


Decay time spectrum of $^{265}Sg^{a,b}$



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 [%]
a	→	*a*	**16**	19.9	**91**
	→	*b*	**2**	2.0	**9**
b	→	*a*	**4**	5.1	**20**
	→	*b*	**19**	19.0	**80**



Decay properties 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

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

	Cross section at 118 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±0.5**

4. Perspectives of SHE nuclear chemistry at RIKEN

– SHE studies opened by beam separation and low background condition –

Approved experiments using the GARIS-gas-jet system

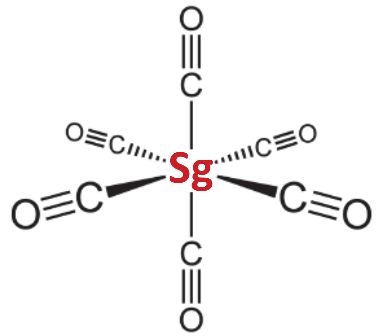
- **Production of ^{261}Rf for chemical studies using the gas-jet transport system coupled to GARIS (H. Haba, RIKEN)**
- **Searching for a new Bk isotope of ^{234}Bk by ^{40}Ar-induced fusion reaction of ^{197}Au (D. Kaji, RIKEN)**
- **α-fine structure spectroscopy of ^{257}Rf and ^{255g,m}Lr (M. Asai, JAEA)**
- **Production of element 105 ^{262}Db for chemical studies using the gas-jet transport system coupled to GARIS (H. Haba, RIKEN)**
- **Gas phase chemistry of trans-actinide elements (H. Kudo, Niigata Univ.)**
- **Electrochemistry of the heaviest actinides (A. Toyoshima, JAEA)**
- **Gas-phase chemistry of $Sg(CO)_6$ –Establishing novel metal-organic chemistry for superheavy elements (Ch. E. Düllmann, Mainz Univ./GSI)**

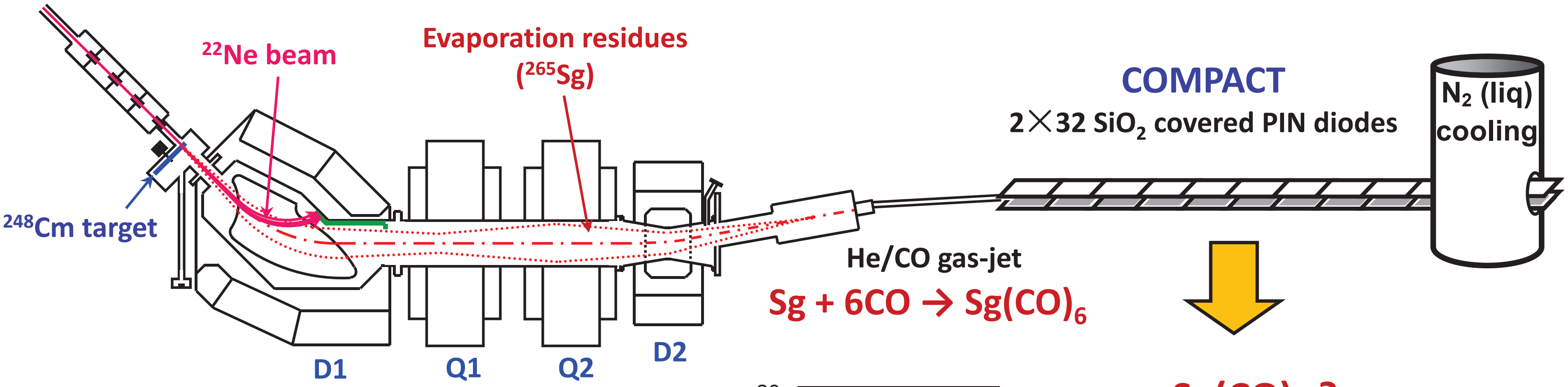
Gas-phase chemistry of $Sg(CO)_6$

–Establishing novel metal-organic chemistry for superheavy elements

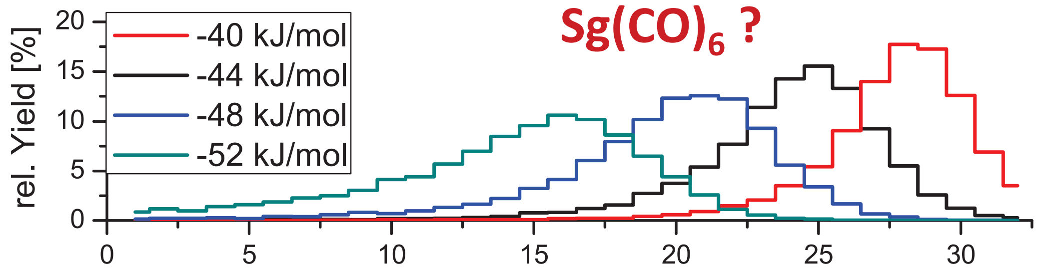
proposed by Ch. E. Düllmann (Mainz Univ./GSI)

- Chemical synthesis and observation of $Sg(CO)_6$
  First metal-organic compound of SHEs
- Measurement of $-\Delta H_{ads}$ for $Sg(CO)_6$ and comparison with theory
  First gas-phase study of a SHE compound in a reduced oxidation state (Sg^0) with π acceptor ligand



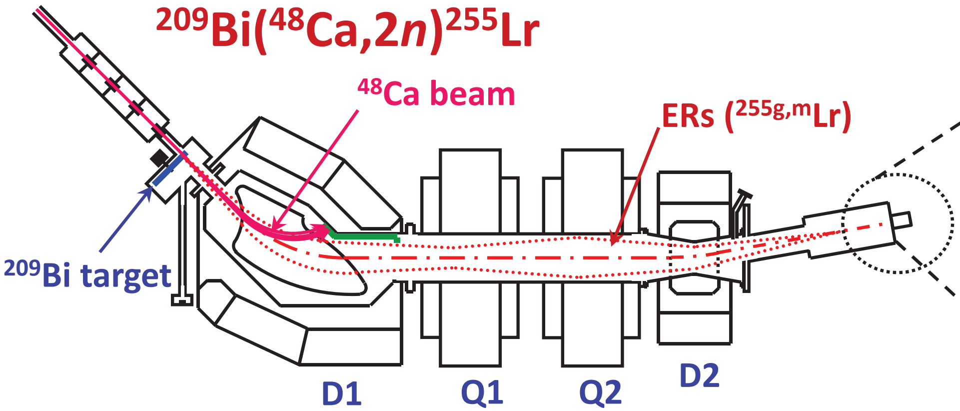


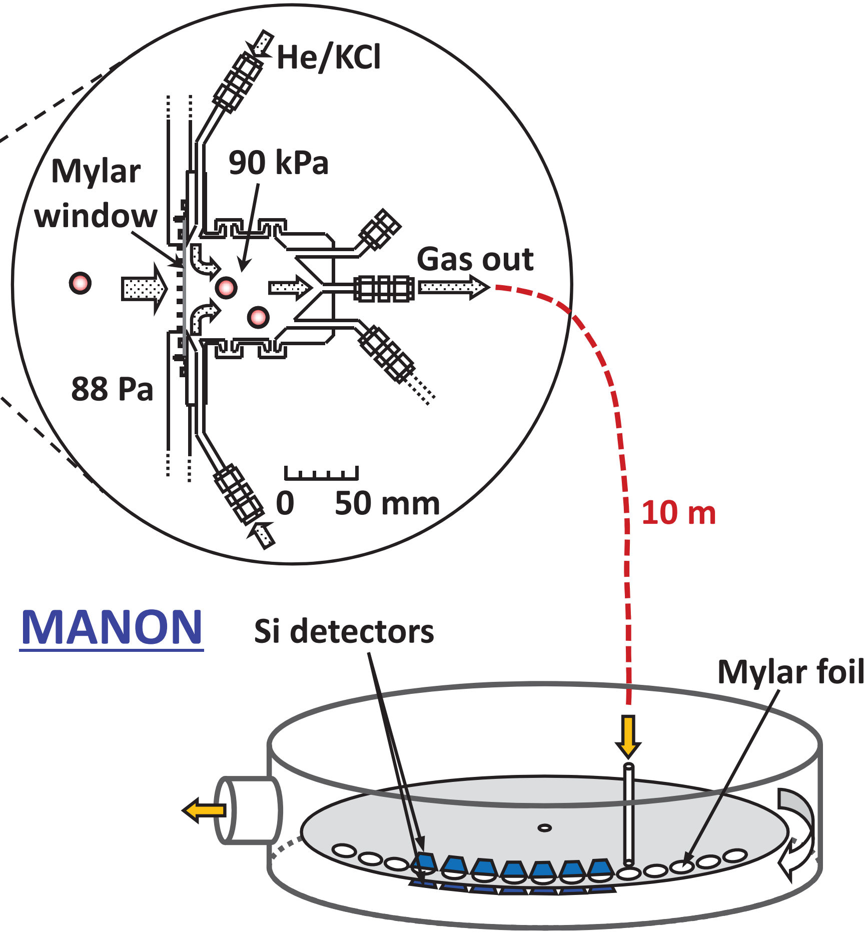
$^{248}Cm(^{22}Ne,5n)^{265}Sg^{a,b}$

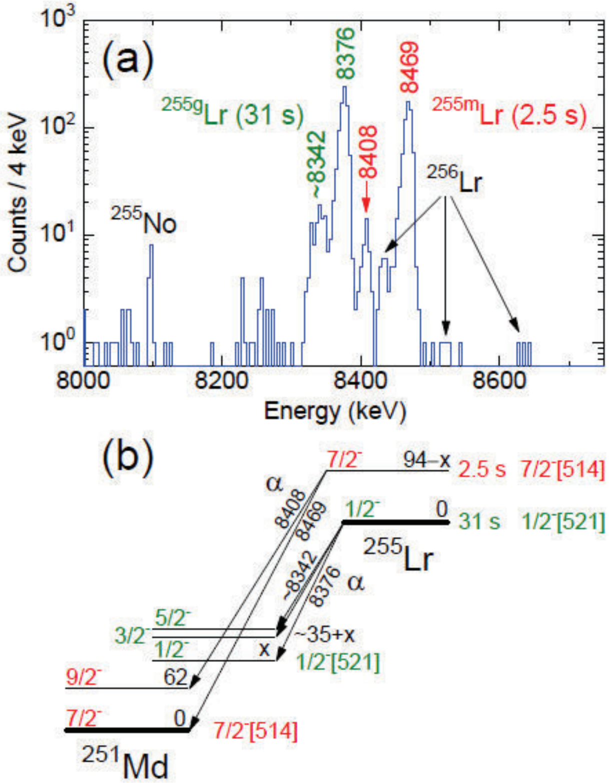


α-fine structure spectroscopy of ^{257}Rf and ^{255g,m}Lr

proposed by M. Asai (JAEA)







Asai *et al.*, RIKEN APR **44**(2011)22.

~12 keV FWHM

- GARIS + Gas-jet system + MANON
  → **High-resolution α fine-structure spectroscopy**
- Rotational band energies and α intensities
  → **Single-particle configuration**

5. Summary

- The gas-jet transport system has been coupled to the RIKEN gas-filled recoil ion separator, GARIS, at RILAC.

- The commissioning of the system was successful.
  ^{169}Tm(^{40}Ar,3n)^{206}Fr; ^{208}Pb(^{40}Ar,3n)^{245}Fm; ^{238}U(^{22}Ne,5n)^{255}No
  → Low background condition
  → High gas-jet transport efficiency irrespectively of beam intensity

- Productions and decay properties of ^{261}Rf^{a,b} and ^{265}Sg^{a,b} were investigated in detail.
  ^{248}Cm(^{18}O,5n)^{261}Rf^{a,b}; ^{248}Cm(^{22}Ne,5n)^{265}Sg^{a,b}

- SHE studies opened by the GARIS gas-jet system were introduced.

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