

Prospects for Laser Spectroscopy of Superheavy Elements

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Outline

1. Introduction and Motivation
2. How Sensitive is Optical Spectroscopy?
3. Remarks on **Laser Induced Fluorescence (LIF)** Spectroscopy in a Buffer Gas Trap
4. RAdioactive Decay Detected **Resonance Ionization Spectroscopy (RADRIS)**

Optical Spectroscopy at Americium Fission Isomers ($Z = 96$)

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5. Ion-Guide-Detected Resonance Ionization Spectroscopy (IGRIS)
Investigation of the Atomic Level Scheme of Fermium ($Z = 100$)
 6. RADRIS with the Ion Collection and Atom Re-Evaporation Method
Status of Laser Spectroscopy at Nobelium ($Z = 102$)
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7. Conclusion

Methods

no levels known

1. Introduction and Motivation

Why Atomic Spectroscopy of Heavy Elements?

1 H	Periodic Table																2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
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
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
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	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89 Ac	104 Rf	105 Du	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Uuu	112 Uub		114 Uuq				

Lanthanides

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
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Actinides

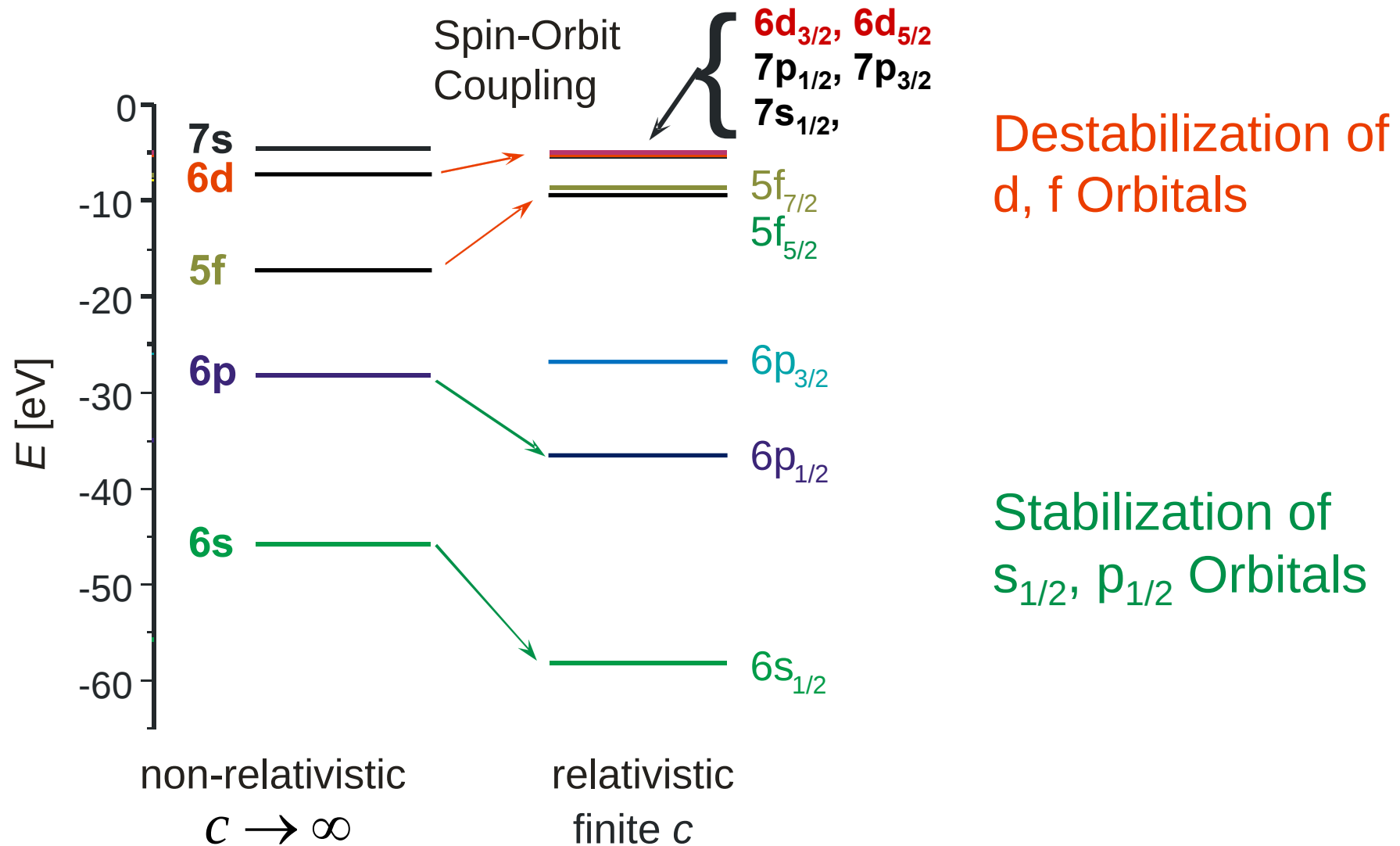
90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr
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6d ²	5f ² 6d	5f ³ 6d	5f ⁴ 6d	5f ⁶	5f ⁷	5f ⁷ 6d	5f ⁹	5f ¹⁰	5f ¹¹	5f ¹²	5f ¹³	5f ¹⁴	5f ¹⁴ 6d
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**no atomic
spectroscopic
data available**

Relativistic Effects for Atomic Level Scheme of Uranium

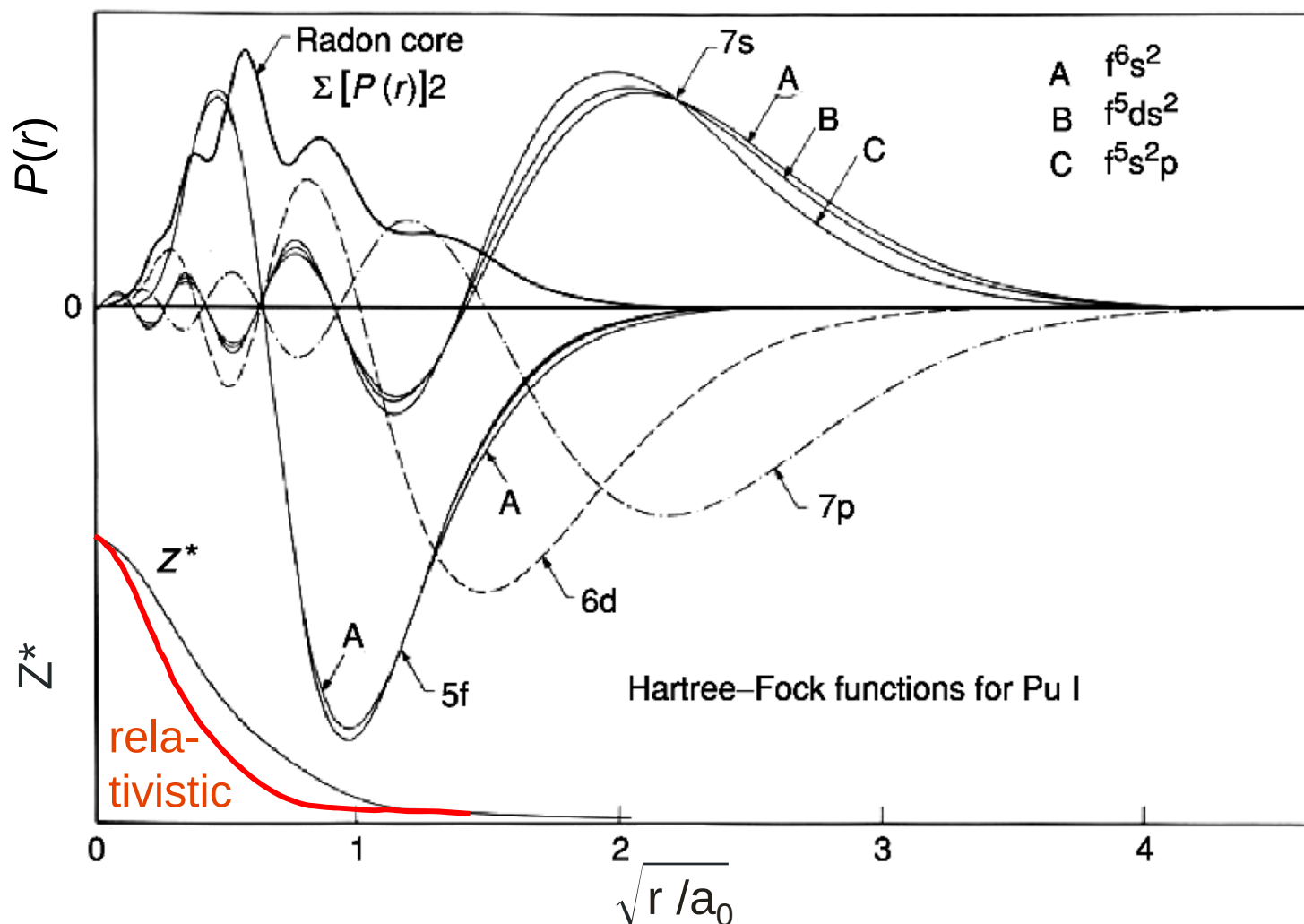


Radial Wave Functions of Relevant Orbits in Pu

Non-relativistic Hartree-Fock calculations

taken from E. F. Worden, Jean Blaise, and Mark Fred and N. Trautmann and J.-F. Wyart

The Chemistry of the Actinide and Transactinide Elements, Third Edition (2008)



Nuclear Ground-State Properties by Atomic Physics Techniques

Hyperfine structure interaction

1. Magnetic dipole hfs

$$A = \frac{\mu_I \langle H_e(0) \rangle}{IJ} \Rightarrow \text{Nuclear magnetic moment } \mu_I$$

2. Electric quadrupole hfs

$$B = eQ_S \langle \varphi_{jj}(0) \rangle \Rightarrow \text{Spectroscopic quadrupole moment } Q_S$$

3. Coupling

$$\vec{J} + \vec{I} = \vec{F} \Rightarrow \text{Nuclear spin } I$$

Isotope shift

$$\delta \langle r^2 \rangle^{A,A'} \Rightarrow \text{change of mean square charge radius}$$

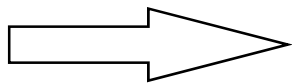
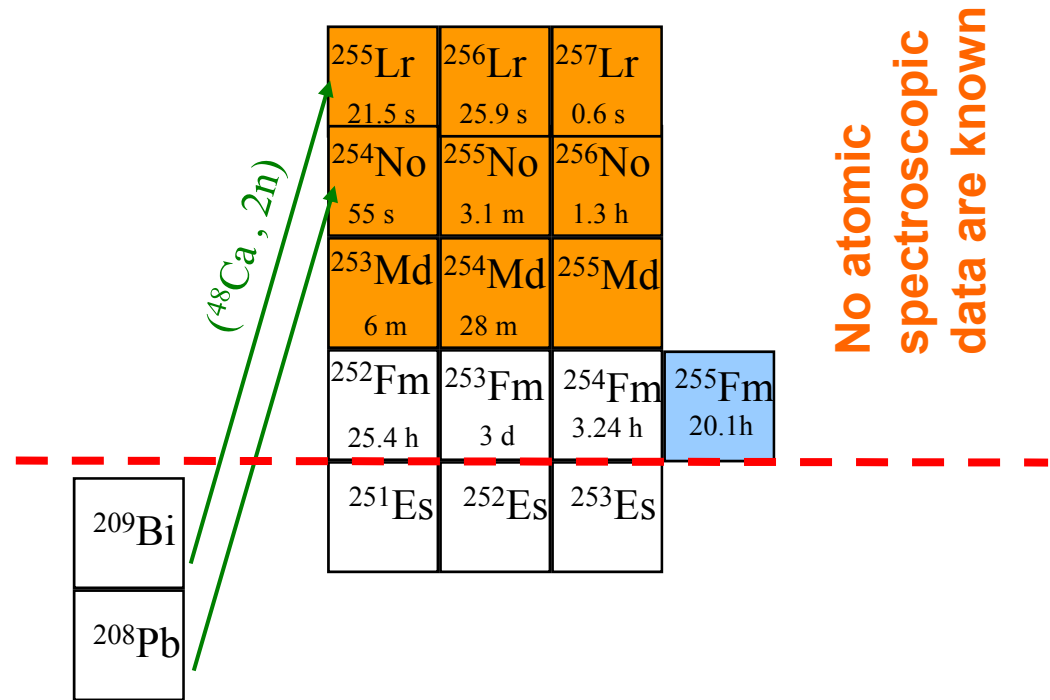
Requirements for Optical Spectroscopy at Trans-Einsteinium Elements

On-line: $Z > 100$ Nuclear reactions
 targets : Pb,U,Pu,Cm,Cf, Es
 beams : p,d, α ,HI (Ca)

**Problem: at $\sigma \sim \mu\text{b}$ small
 production rates $\sim 1/\text{s}$**

**Off-line: $Z \leq 100$ (Fm) breeding
 of material in
 high flux isotope reactors**

**Problem: Experiments at Fm with
 typically 10^{10} atoms only**



Ultra-sensitive Laser-Spectroscopic Methods

2. How Sensitive is Optical Spectroscopy?

How Sensitive is Optical Spectroscopy?

$$\sigma_r(\omega_0) = \frac{\lambda^2}{2\pi} \frac{g_k}{g_i} \approx 10^{-10} \text{ cm}^2$$

⇒ Transition rate per atom

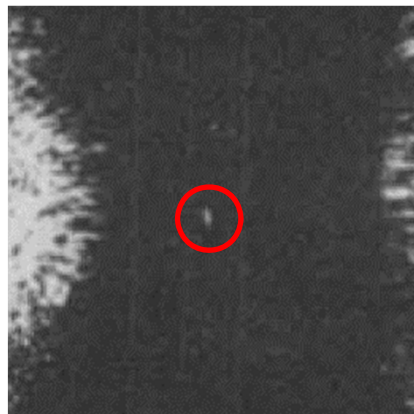
$$P_{ik} = \frac{d\dot{N}}{dA} \cdot \sigma_r(\omega_0) = \frac{1}{\hbar\omega} \frac{dP}{dA} \cdot \sigma_r(\omega_0)$$

Power of tuneable cw dye lasers $P \approx 100 \text{ mW} \dots 1 \text{ W}$

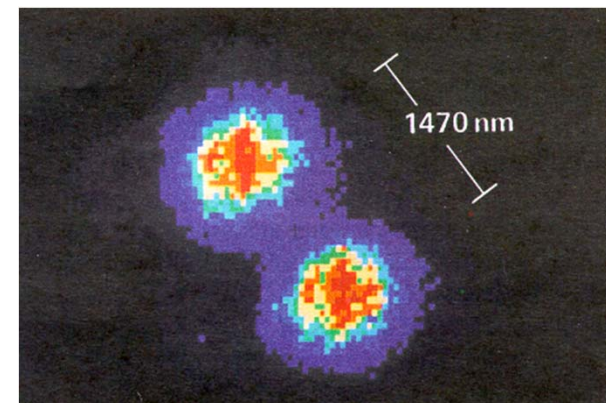
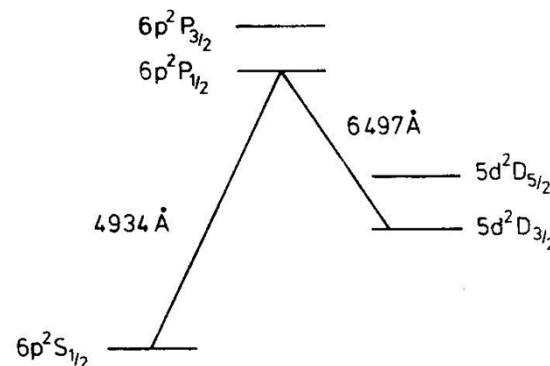
⇒ Flux of photons $\frac{d\dot{N}}{dA} \approx 10^{17} \dots 10^{18} / (\text{s cm}^2) \Rightarrow P_{ik} \approx 10^8 / (\text{s atom})$

“In principle” only 1 atom required

First example: Ba^+ stored in an ion trap

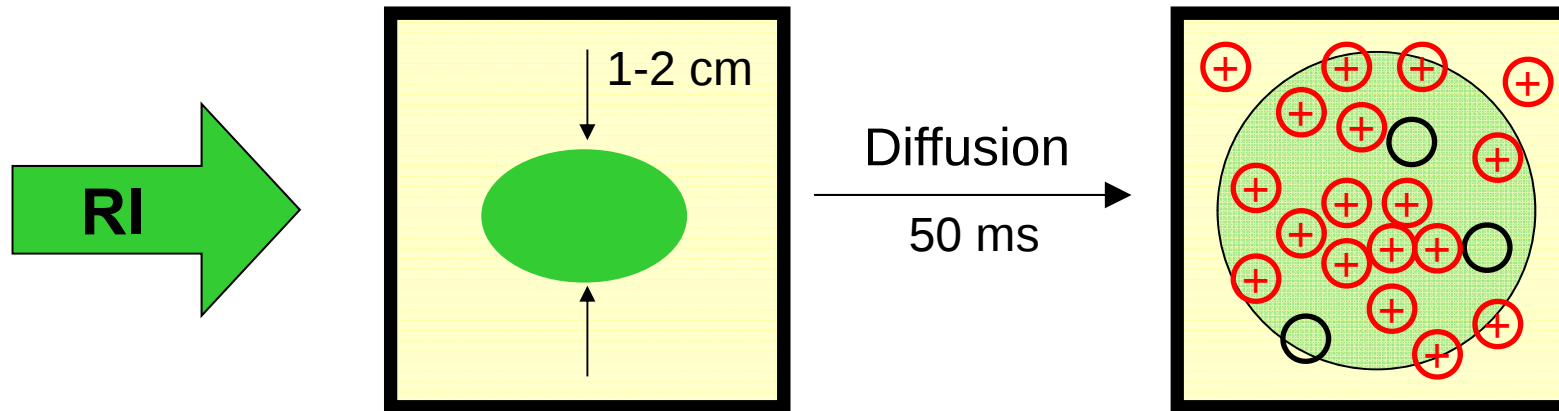


(Toschek, Dehmelt, Neuhauser et al., 1980)



R.G.Voe et al., PRL 76 (1996) 2049

Buffer Gas Trap



**Radioactive Ions
from Separator**
 $T_{1/2} > 1$ ms

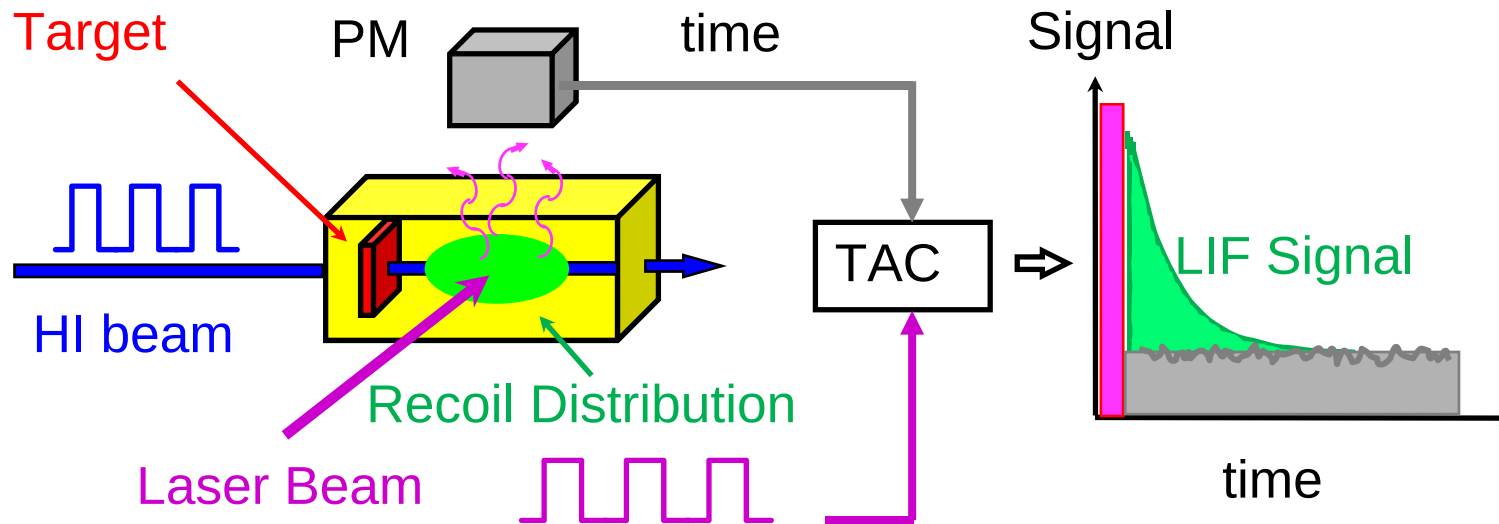
Buffer Gas Cell
He, Ar at ≈ 100 mbar

⊕ 85% charged
○ 15% neutral

- Laser Spectroscopy at Neutral Fraction
- Ion Chemistry and Drift Time Measurements at Charged Fraction
- Cooling and Bunching and Transfer to Traps for Mass Measurements

3. Remarks on Laser Induced Fluorescence (LIF) Spectroscopy in a Buffer Gas Trap

Laser Induced Fluorescence (LIF) Spectroscopy



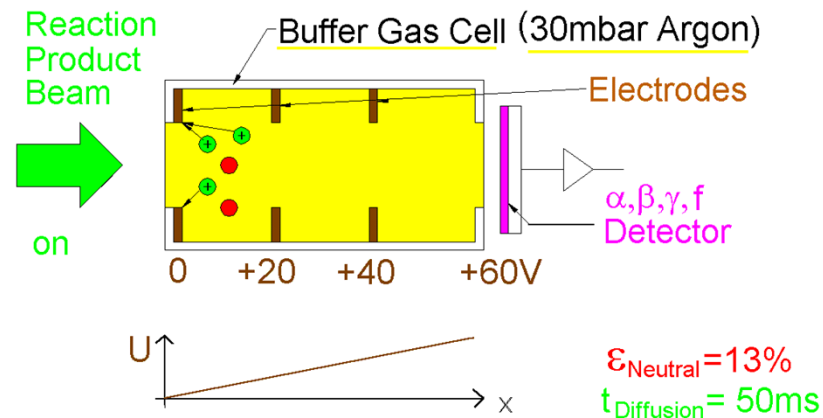
G. D. Sprouse et al.,
Laser Spectroscopy of Light Yb Isotopes On-Line in a Cooled Gas Cell.
Phys. Rev. Lett. **63** (1989) 1463
Sensitivity: Target production rate about $10^3/\text{s}$

4. **R**adioactive **D**ecay Detected **R**esonance Ionisation **S**pectroscopy (**RAD**RI**S**)

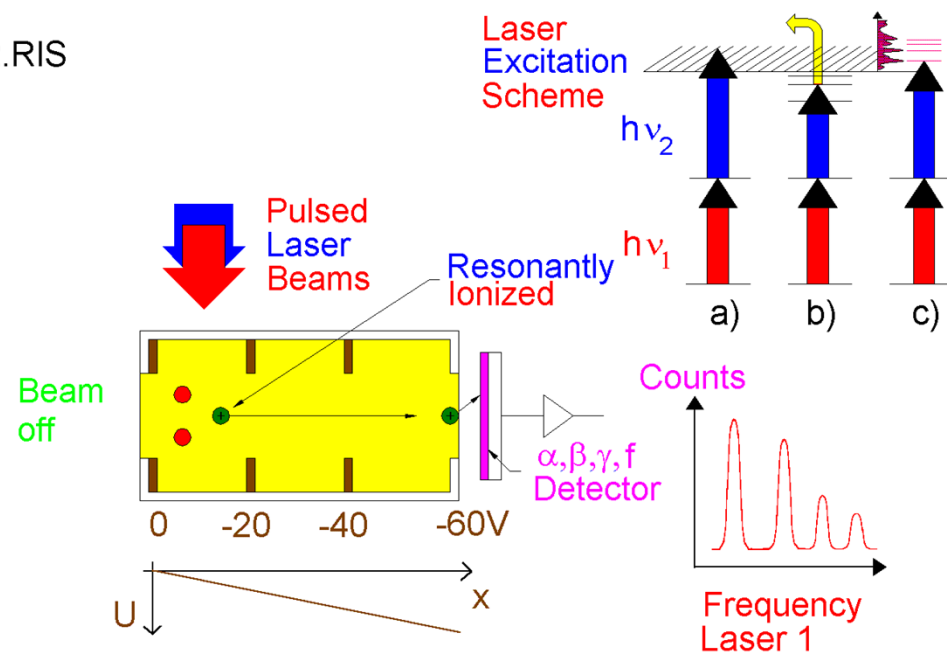
Optical Spectroscopy at Americium
Fission Isomers ($Z = 96$)

RADRadioactive Decay Detected Resonance Ionisation Spectroscopy (RADRIS)

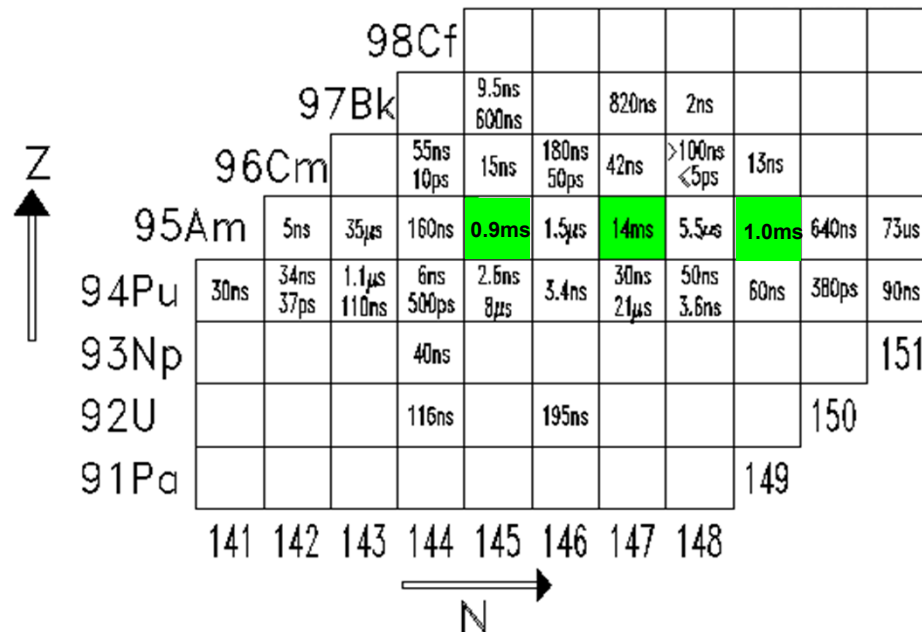
1. Loading



2. RIS

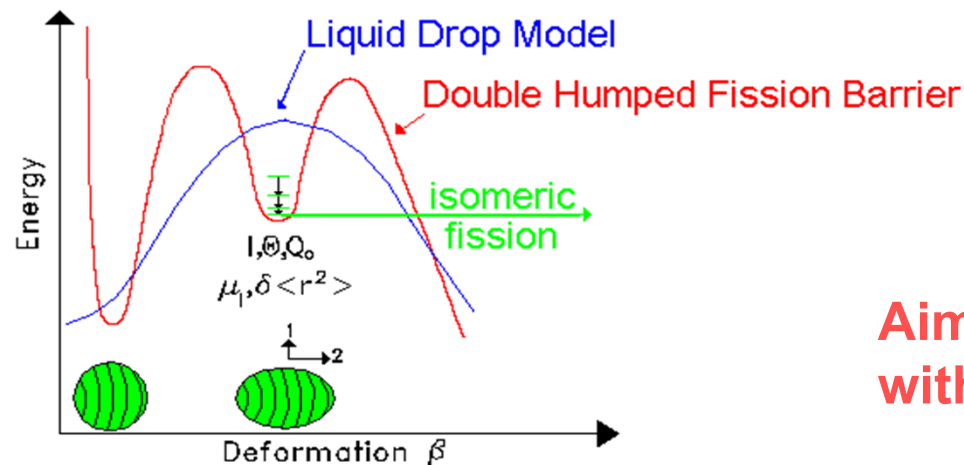


Americium Fission Isomers

 ^{240}fAm ($T_{1/2} = 0.9 \text{ ms}$) ^{242}fAm ($T_{1/2}=14$ ms)
$$^{244}\text{fAm} \text{ (} T_{1/2} = 1 \text{ ms)}$$

Dubna 1962

- conversion electron spectroscopy
- lifetime measurements of excited states (charge plunger method)
- g-factor measurements
- γ -ray spectroscopy
- optical spectroscopy

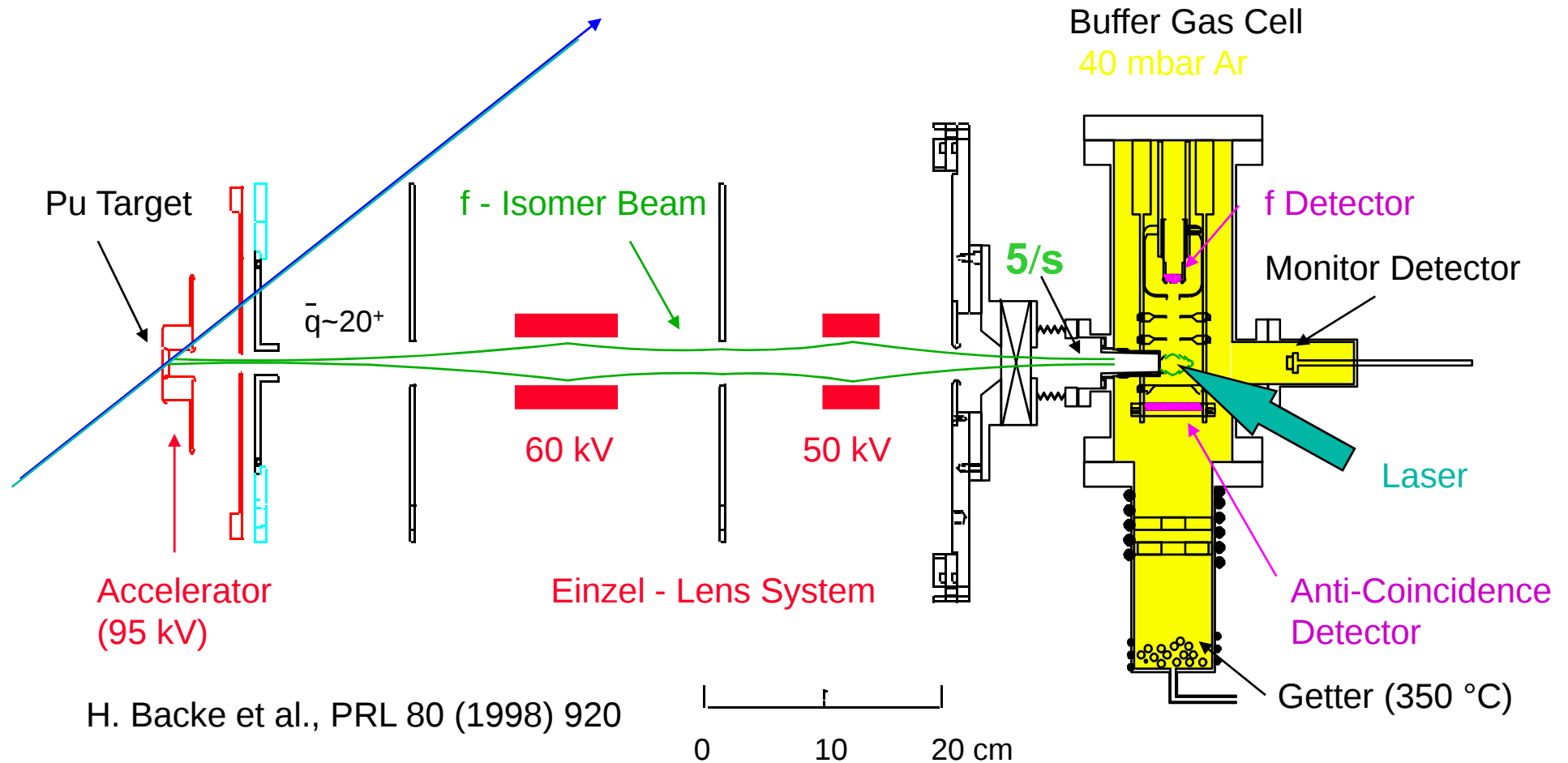


**Aim: $\delta \langle r^2 \rangle$, Q_{20} , I , g
with Optical Spectroscopy**

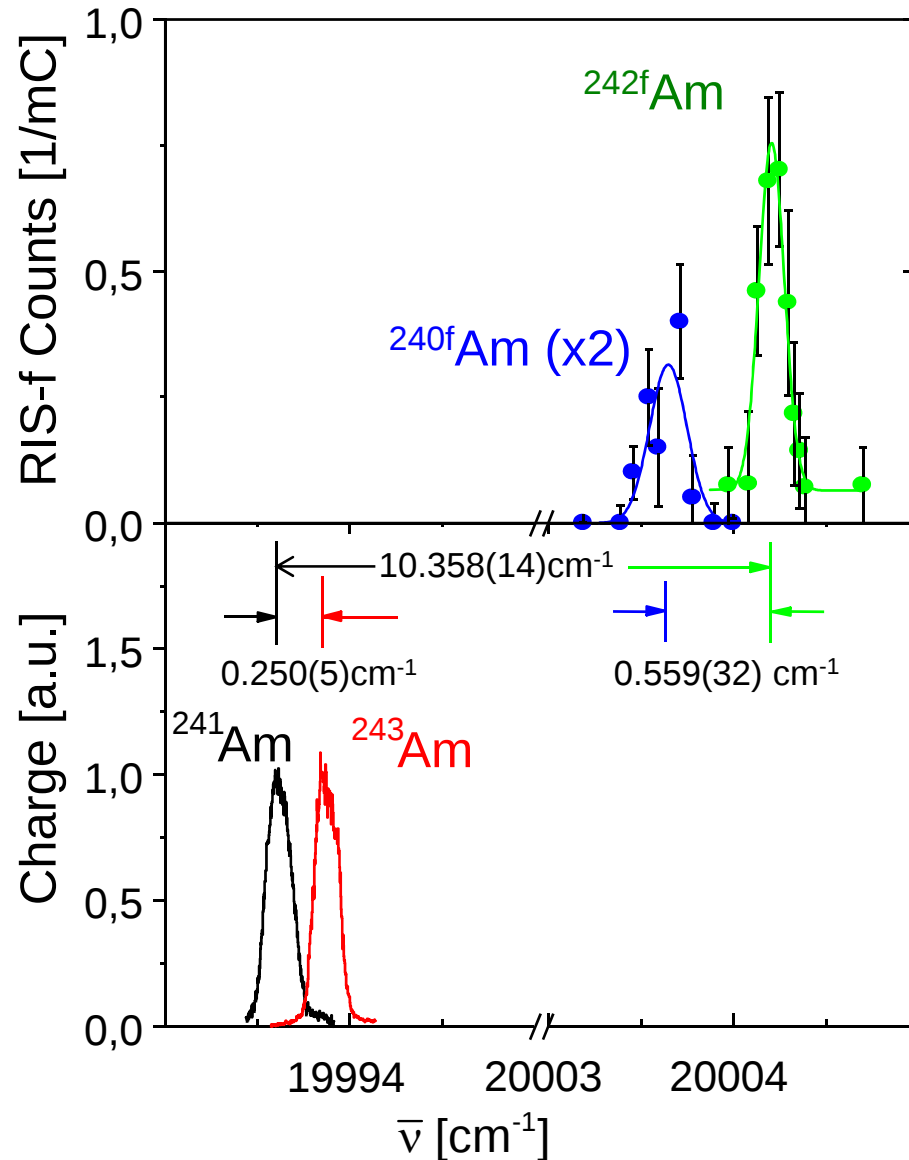
Experimental Setup

^{242}Pu (d,2n) ^{242}fAm $T_{1/2}=14\text{ms}$ d Beam 6ms on / 6ms off 12 MeV, $I=3\mu\text{A}$
 ^{242}Pu (p,3n) ^{240}fAm $T_{1/2}=0.9\text{ms}$ p Beam 2ms on / 2ms off 24 MeV, $I=3\mu\text{A}$
 ^{244}Pu (d,2n) ^{244}fAm $T_{1/2}=1\text{ms}$ d Beam 2ms on / 2ms off 14 MeV, $I=2\mu\text{A}$

$$\sigma = (8 \pm 3) \mu\text{b}$$

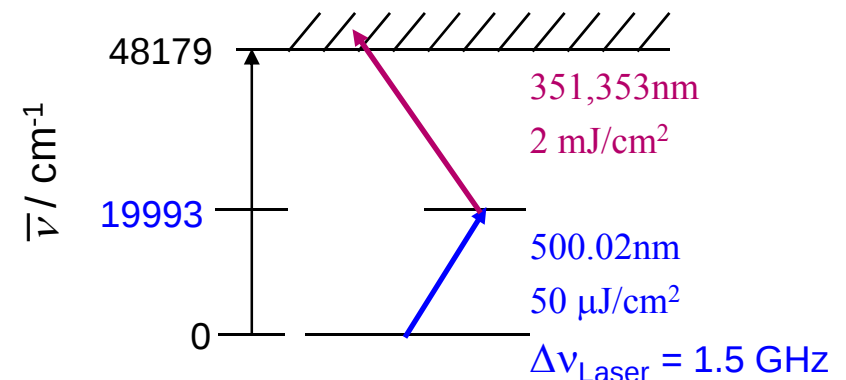


Isotope Shift Measurement in the Second Potential Minimum



$^{242}\text{Pu}(\text{d},2\text{n})^{242\text{f}}\text{Am}$, $\sigma = 8\text{ }\mu\text{b}$, 12 MeV

$^{242}\text{Pu}(\text{p},3\text{n})^{240\text{f}}\text{Am}$, $\sigma = 8\text{ }\mu\text{b}$, 24 MeV



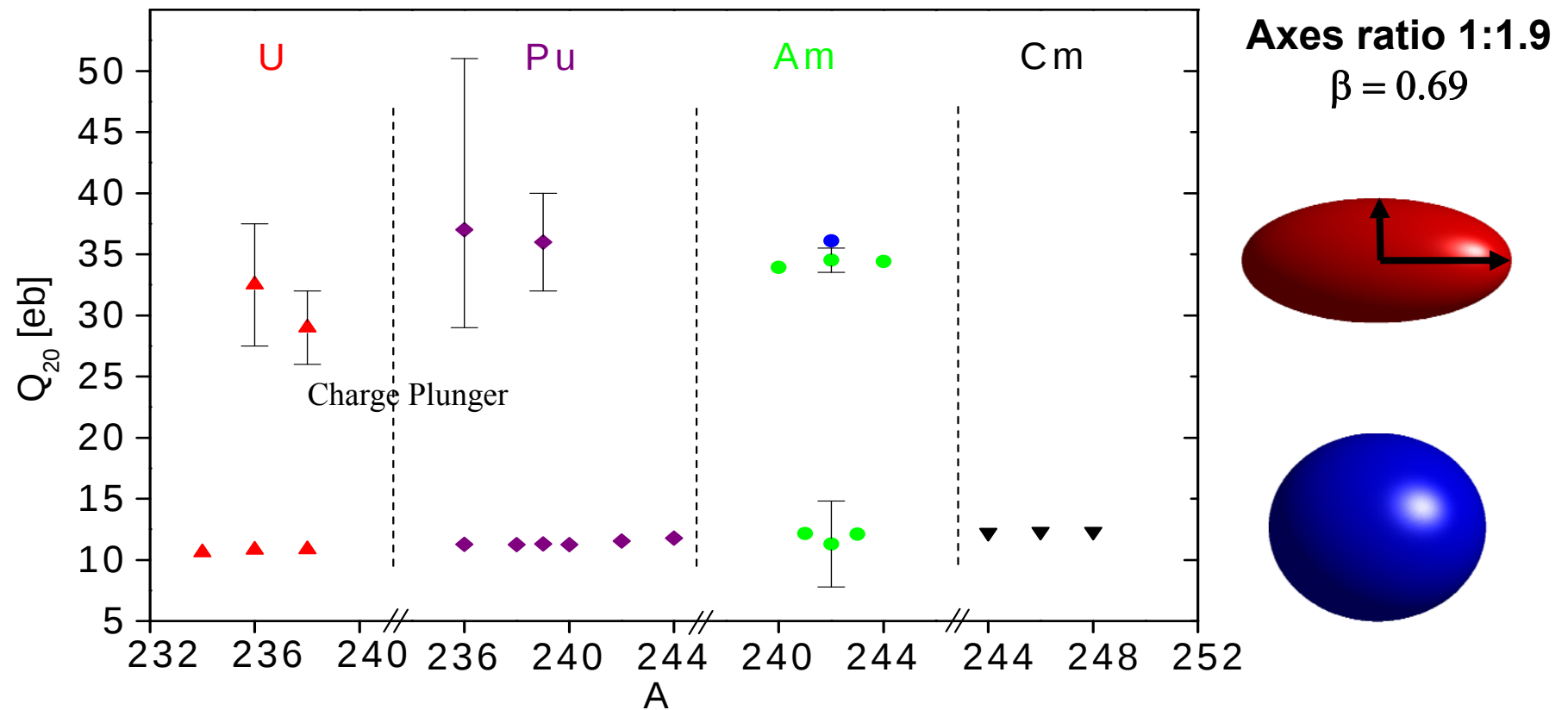
$$X_{\text{exp}}(^{242\text{f}}\text{Am}) = \frac{\text{IS}^{242\text{f},241}}{\text{IS}^{243,241}} = 41.4 (8)$$

$$X_{\text{exp}}(^{240\text{f}}\text{Am}) = \frac{\text{IS}^{240\text{f},241}}{\text{IS}^{243,241}} = 39.2 (8)$$

At variance with

$$X^{240\text{f}} = 26.8 (2.0) \text{ (Bemis et al.)}$$

Quadrupole Moments of Fission Isomers



5. Ion-Guide-Detected Resonance Ionization Spectroscopy (IGRIS)

Investigation
of the Atomic Level Scheme
of Fermium ($Z = 100$)

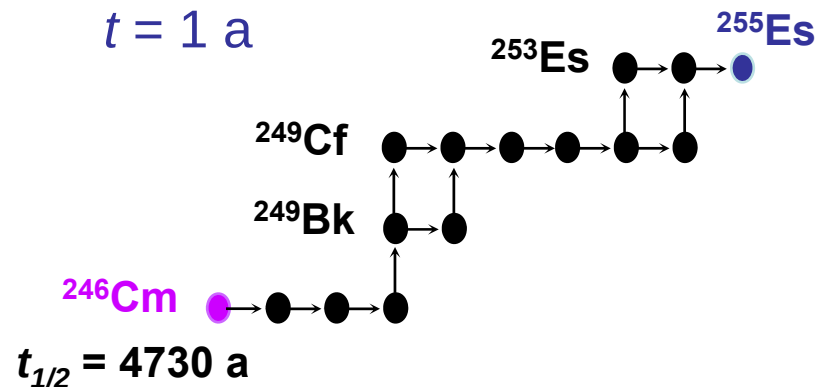
Production of ^{255}Fm

High Flux Isotope Reactor
Oak Ridge National Laboratory

- Breeding of ^{255}Es ($Z = 99$)

$$\Phi_n = 2.6 \cdot 10^{15} / \text{cm}^2 \cdot \text{s}$$

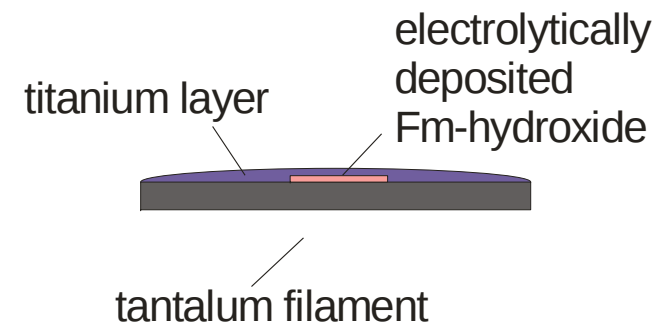
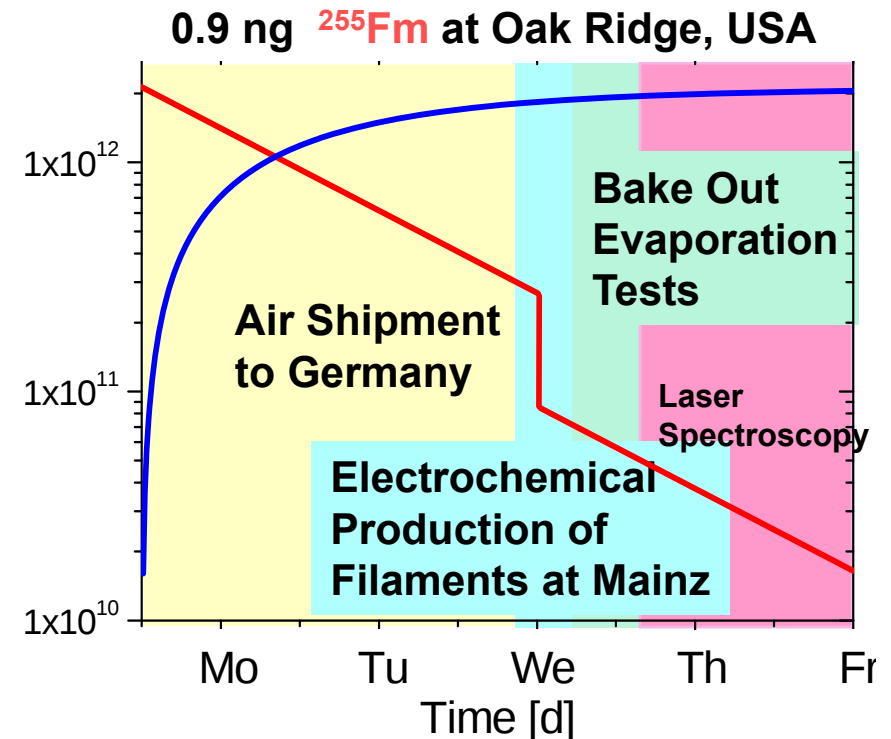
$$t = 1 \text{ a}$$



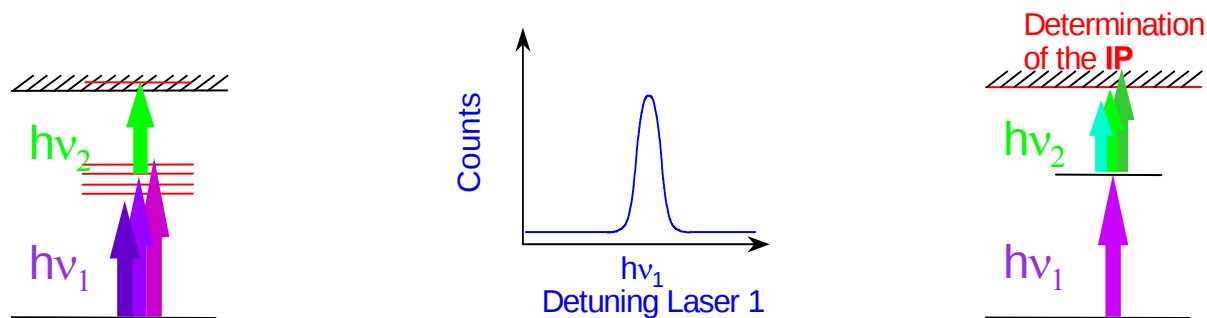
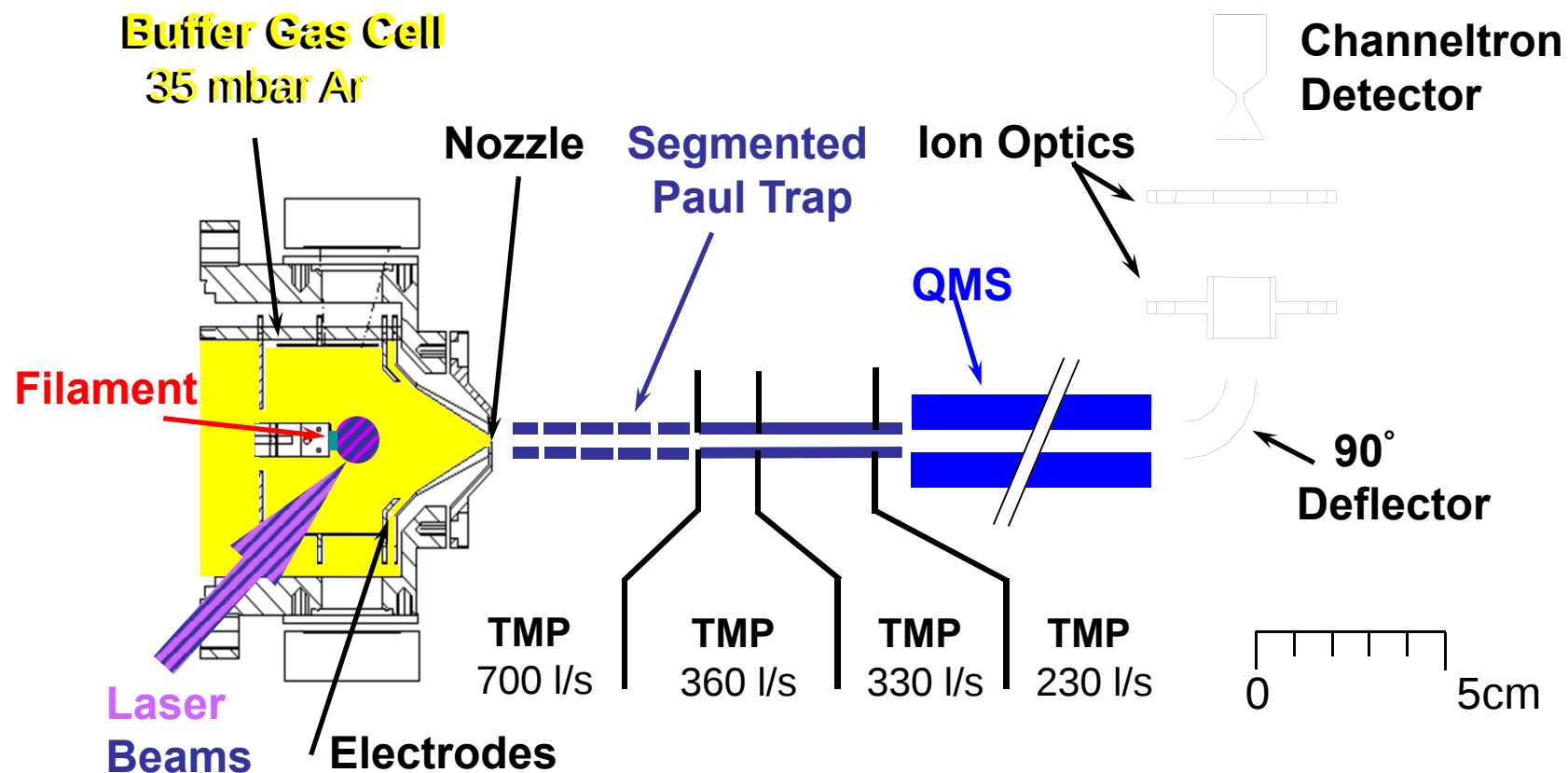
- Chemical Separation of Es,

- Waiting for $^{255}\text{Es} \xrightarrow[t_{1/2} = 39.8 \text{ d}]{\beta^-} ^{255}\text{Fm}$ decay

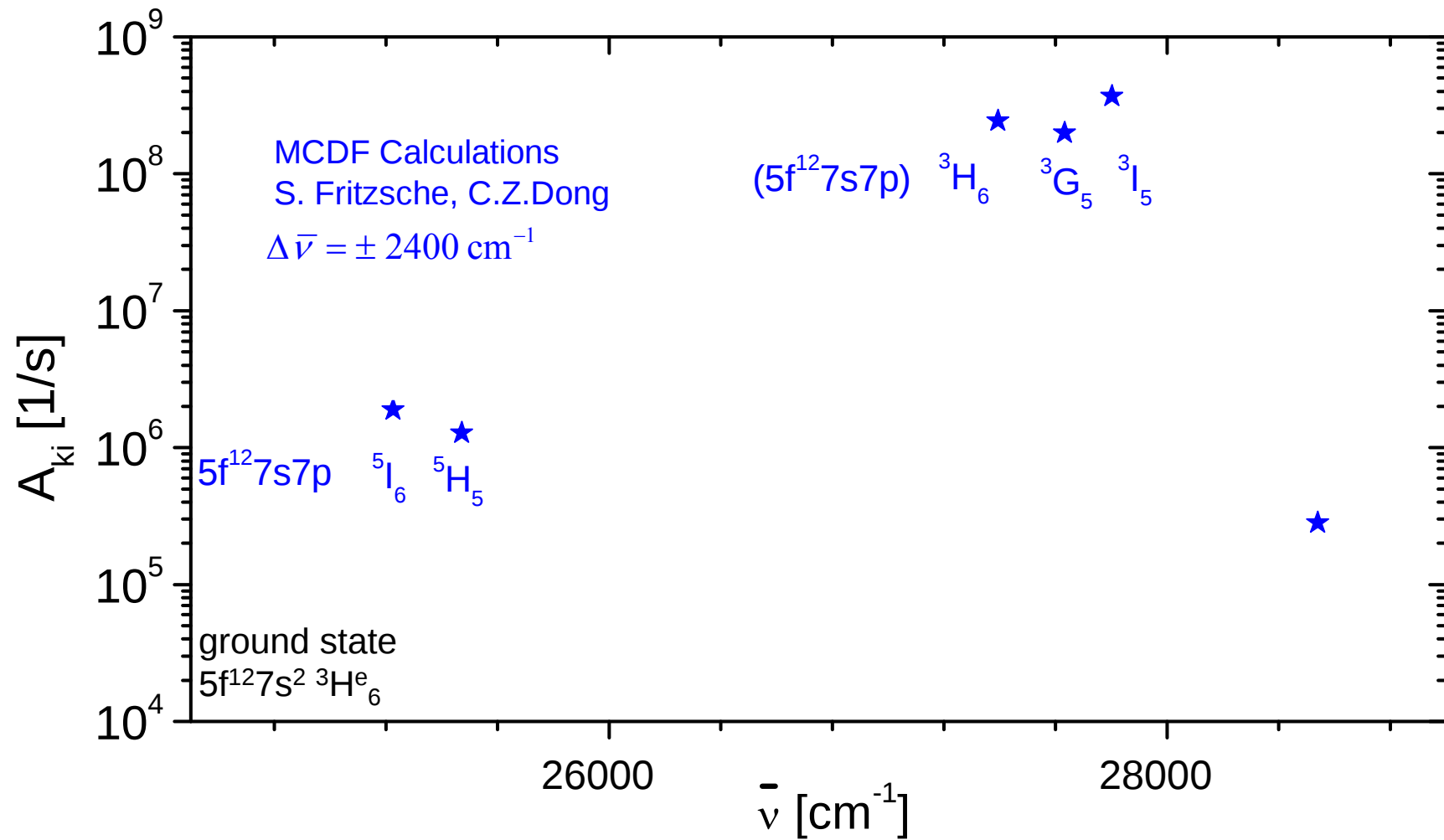
- Chemical Extraction of ^{255}Fm ($t_{1/2} = 20.1 \text{ h}$)



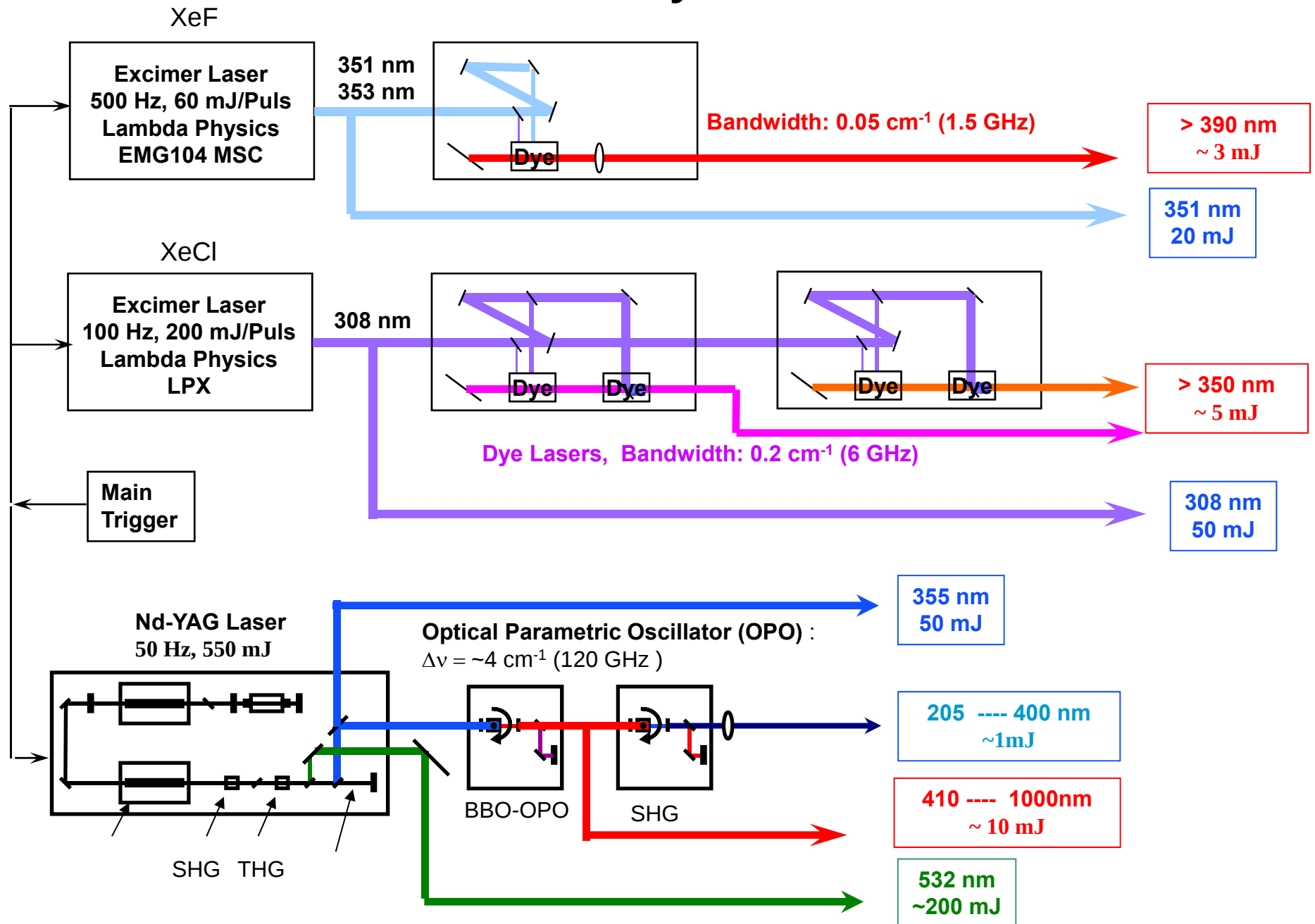
Ion-Guide Detected Resonance Ionization Spectroscopy with Mass Selective Ion Detection



Theoretical Level Predictions for Fermium

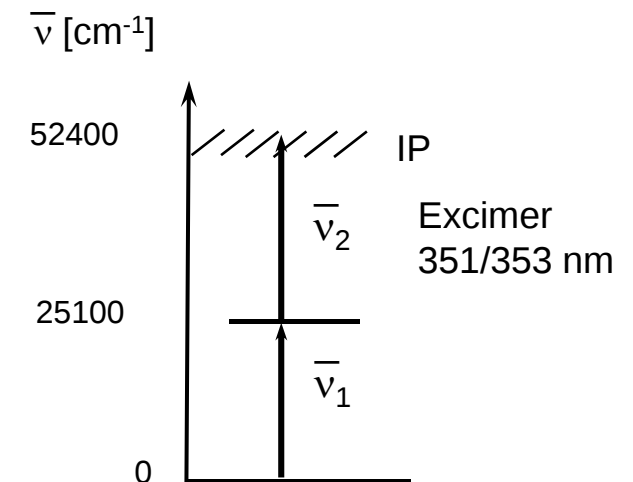
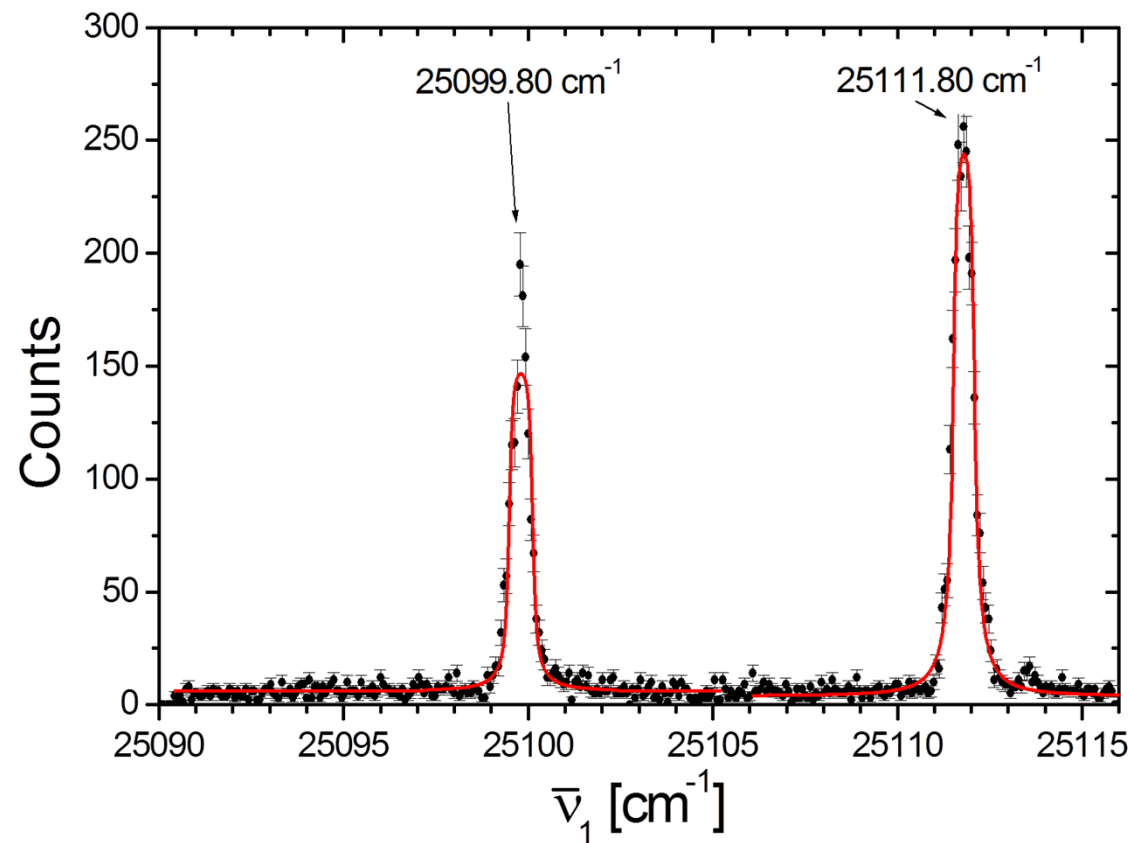


Laser Systems

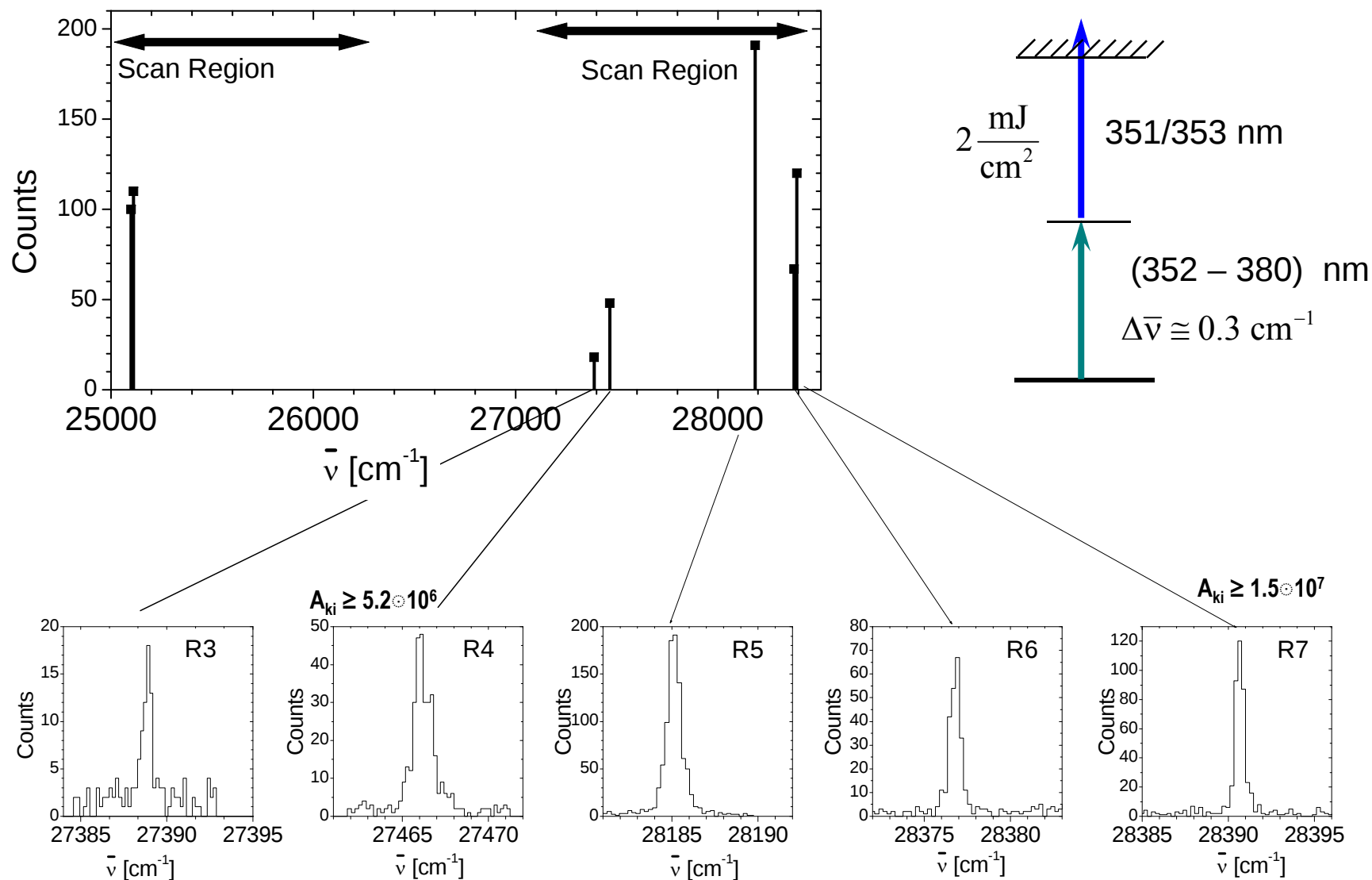


The First Optical Transition ever Observed for Fermium ($Z = 100$)

M. Sewtz et al., Phys. Rev. Letters **90** (2003) 163002



Summary of Level Search



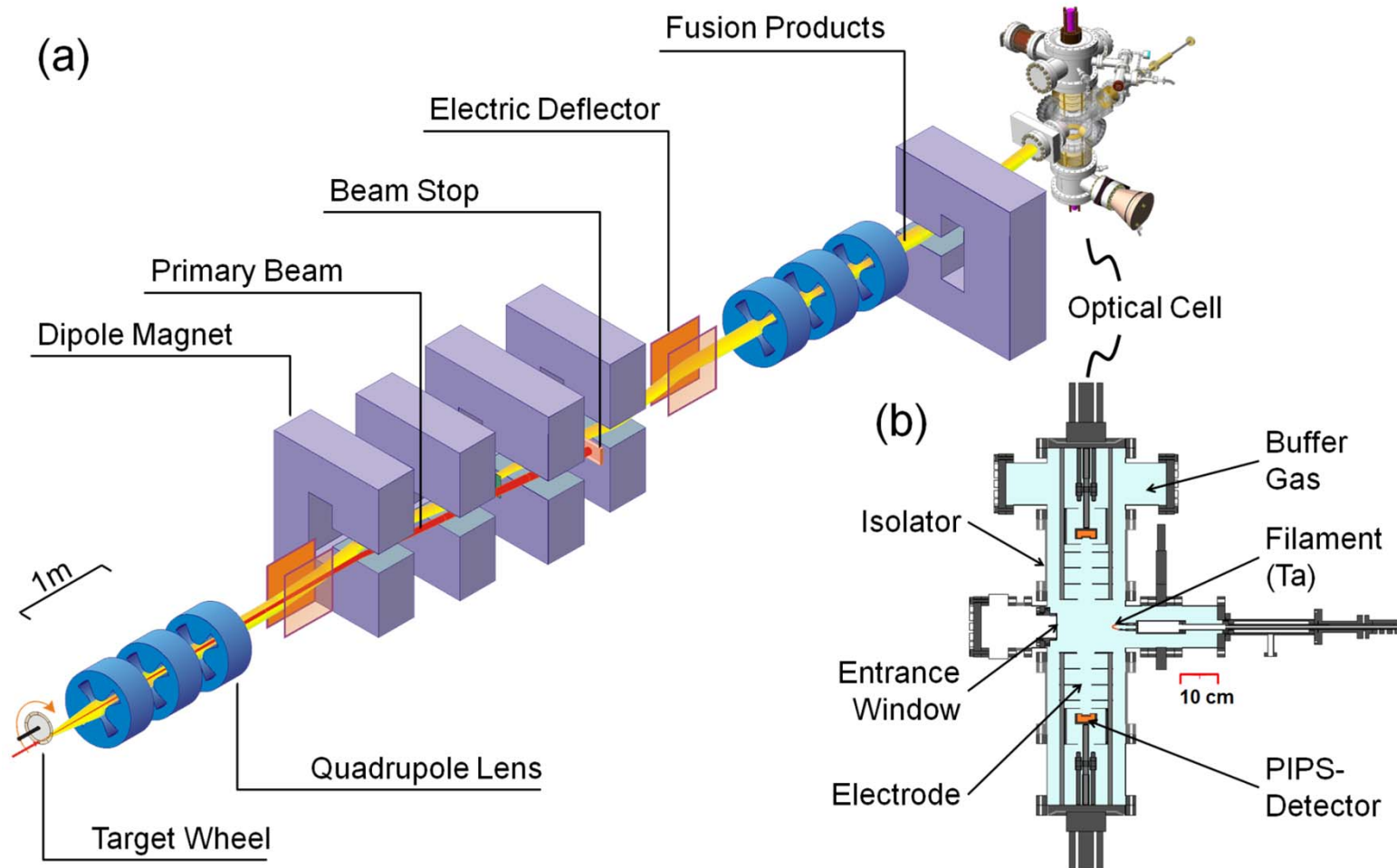
6. RADRIS with the Ion Collection and Atom Re- Evaporation Method

Status of Laser Spectroscopy
at Nobelium ($Z = 102$)

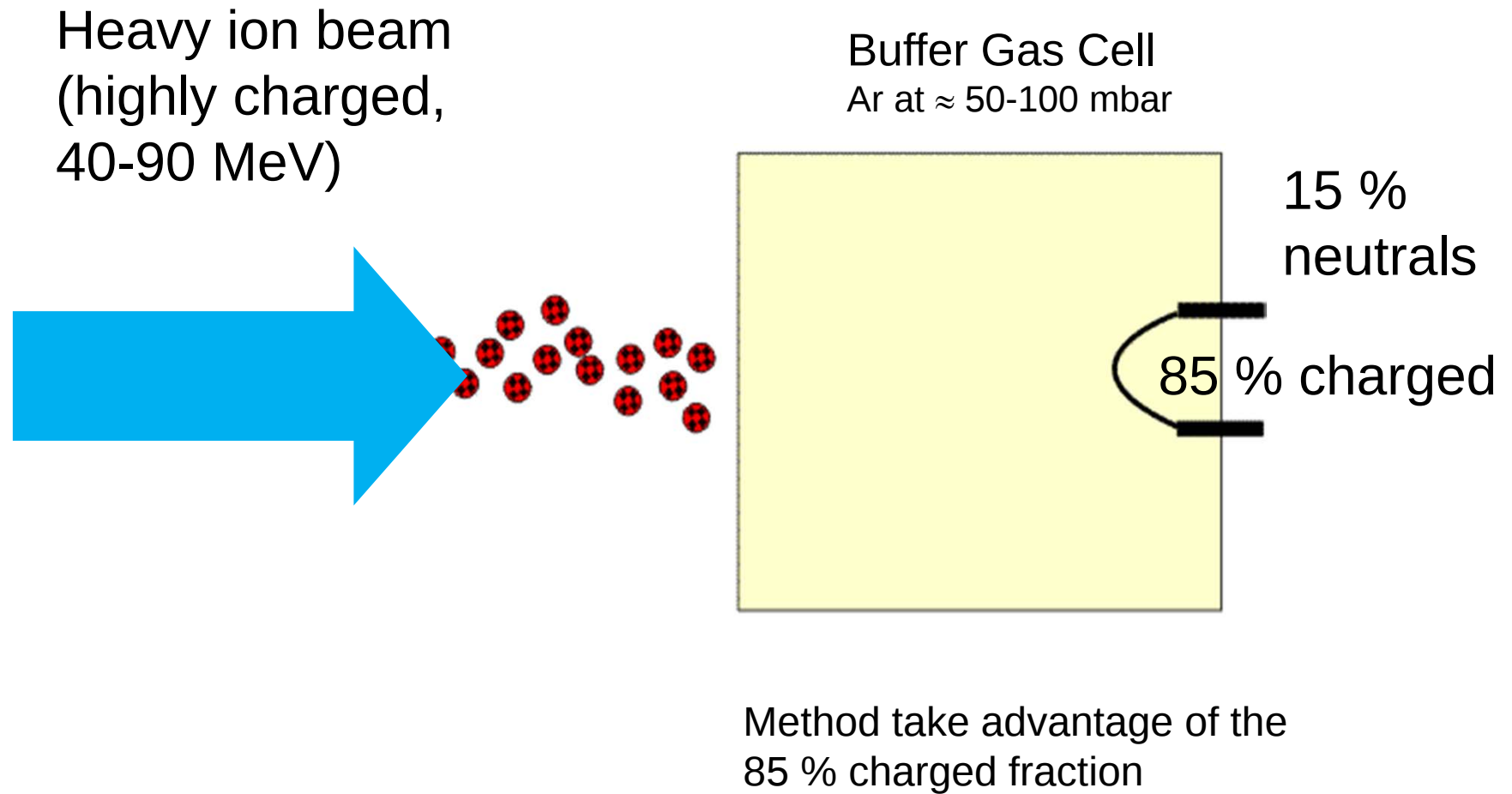
Experimental Setup

^{208}Pb ($^{48}\text{Ca}, 2n$) ^{254}No ($t_{1/2}=55\text{ s}$, α decay)

$\sigma \sim 3.4\text{ }\mu\text{b}$: 5 Ions/s



The Ion Collection and Atom Re-Evaporation Method (ICARE)

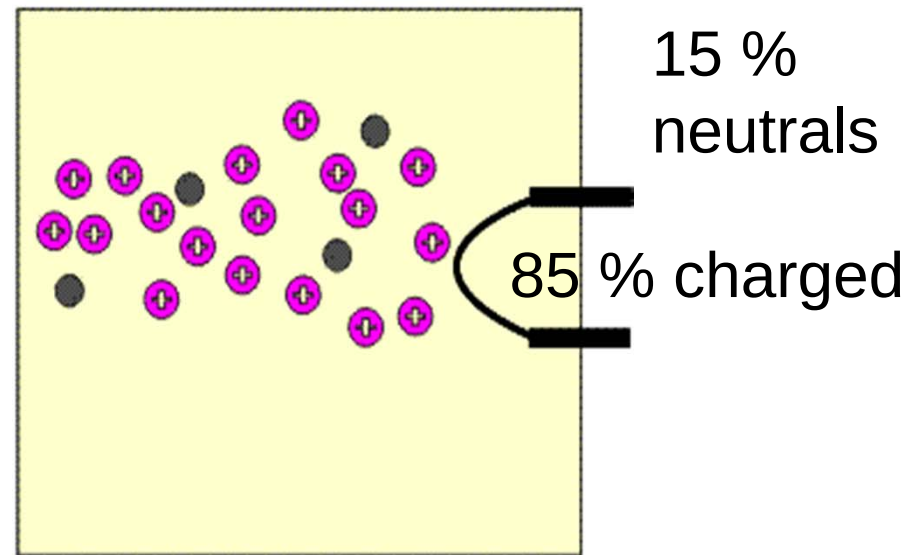


The Ion Collection and Atom Re-Evaporation Method (ICARE)

Heavy ion beam
(highly charged,
40-90 MeV)



Buffer Gas Cell
Ar at $\approx 50\text{-}100$ mbar



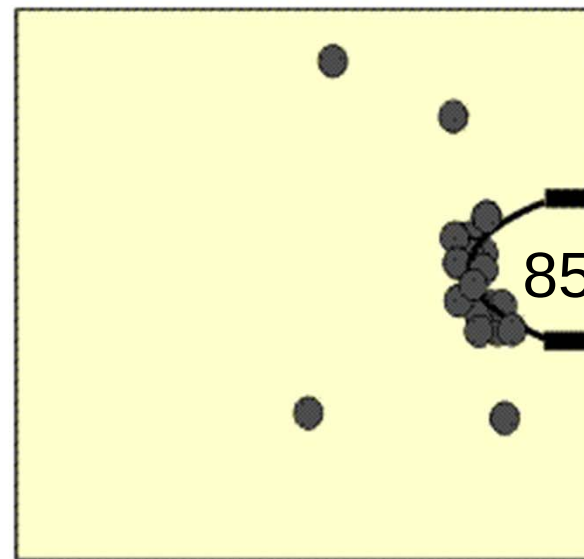
Method take advantage of the
85 % charged fraction

The Ion Collection and Atom Re-Evaporation Method (ICARE)

Heavy ion beam
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Buffer Gas Cell
Ar at $\approx 50\text{-}100$ mbar

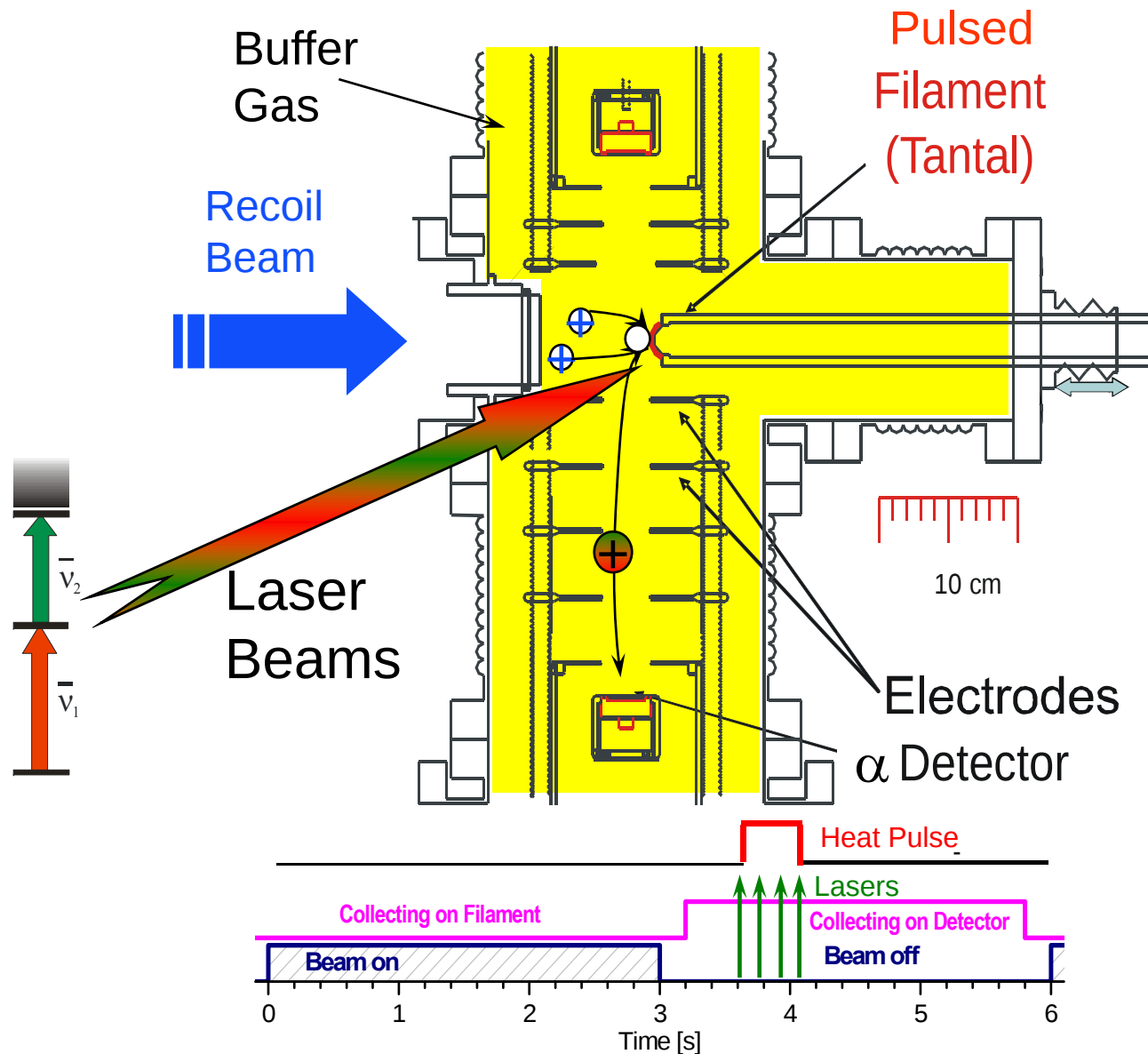


15 %
neutrals

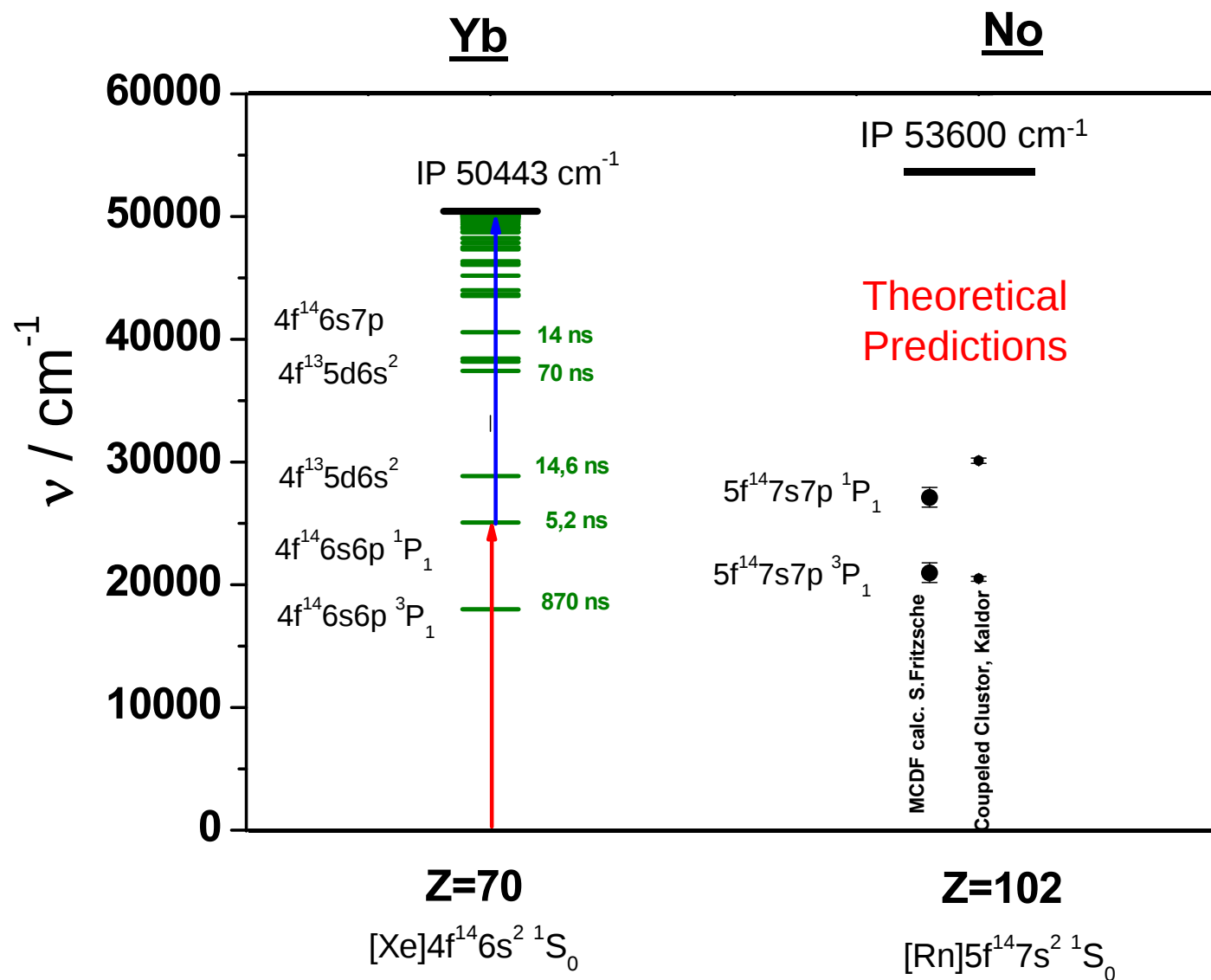
85 % charged

Method take advantage of the
85 % charged fraction

Radioactive Decay Detected Resonance Ionization with the ICARE Method

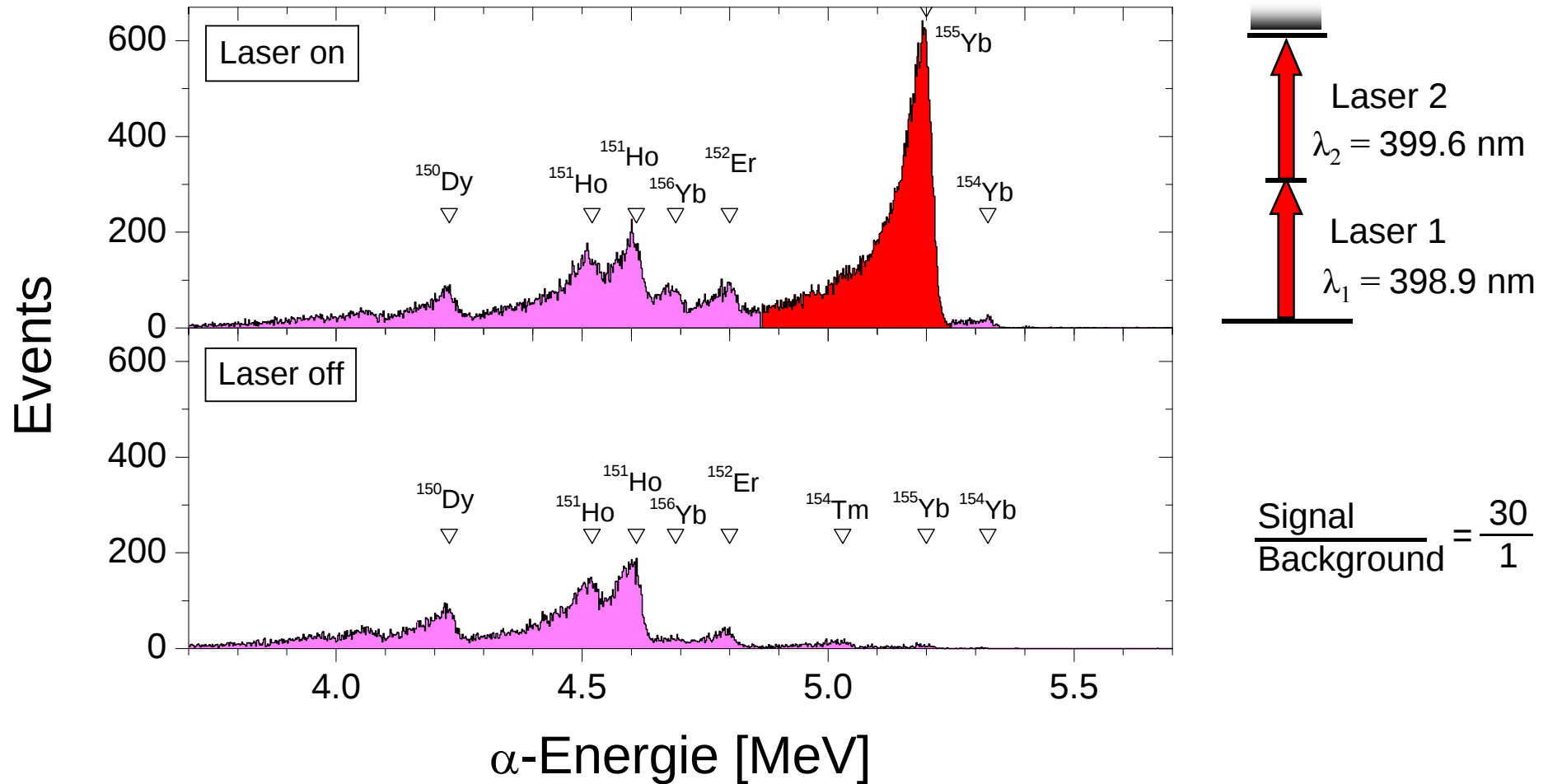


Nobelium as Chemical Homolog of Ytterbium



Test of the Method at ^{155}Yb ($Z=70$)

$^{107}\text{Ag}(^{52}\text{Cr}, p3n)^{155}\text{Yb}$ ($t_{1/2}=1.75$ s, α - decay);
 $\sigma = 15$ mb, $\rho d = 0.455$ mg/cm², $4 \cdot 10^4$ Ions/s

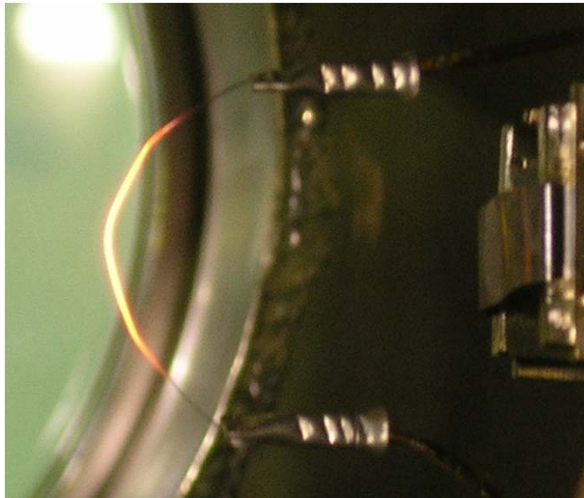


Efficiency

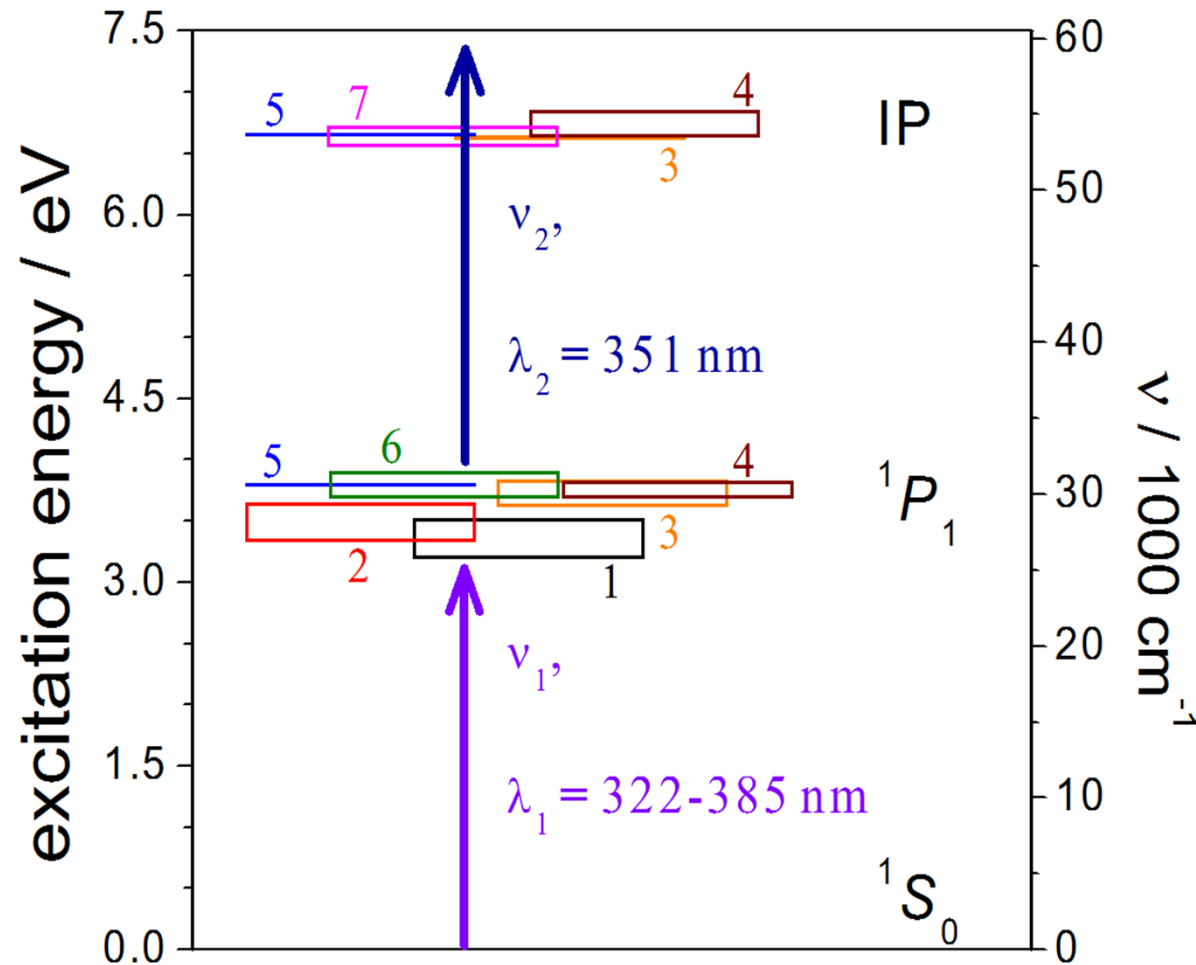
$$\frac{N_{\text{Yb}}^{\text{RIS}}}{N_{\text{Yb}}^{\text{Target}}} = 0.8 \cdot 10^{-2}$$

$$= \epsilon_{\text{SHIP}} \cdot \epsilon_{\text{Cell}} \cdot \epsilon_{\text{Ion}} \cdot \epsilon_{\text{Collect}} \cdot \epsilon_{\text{Decay}} \cdot \underbrace{\epsilon_{\text{Evap}} \cdot \epsilon_{\text{RIS}}}_{0.16} \cdot \epsilon_{\text{Transp.}} \cdot \epsilon_{\alpha\text{Det}}$$

$\downarrow \quad \downarrow \quad \downarrow \quad \downarrow \quad \downarrow \quad \downarrow \quad \downarrow \quad \downarrow$
 $0.5 \cdot 0.7 \cdot 0.85 \cdot 0.8 \cdot 0.59 \cdot 0.16 \cdot 1 \cdot 0.36$



Predictions for the 1P_1 – level



[1,2] (MCDF) S. Fritzsche, Eur. Phys. J. D 33, 15 (2005)

[3] (IHFSCC) A. Borschevsky et al., Phys. Rev. A 75, 042514 (2007)

[4] (RCC) V.A. Dzuba et al., Phys. Rev. A 90 (2014) 012504

[5] (MCDF) Y. Liu, R. Hutton, Y. Zou, Phys. Rev. A 76, 062503 (2007)

[6] (MCDF) P. Indelicato et al., Eur. Phys. J. D45, 155 (2007)

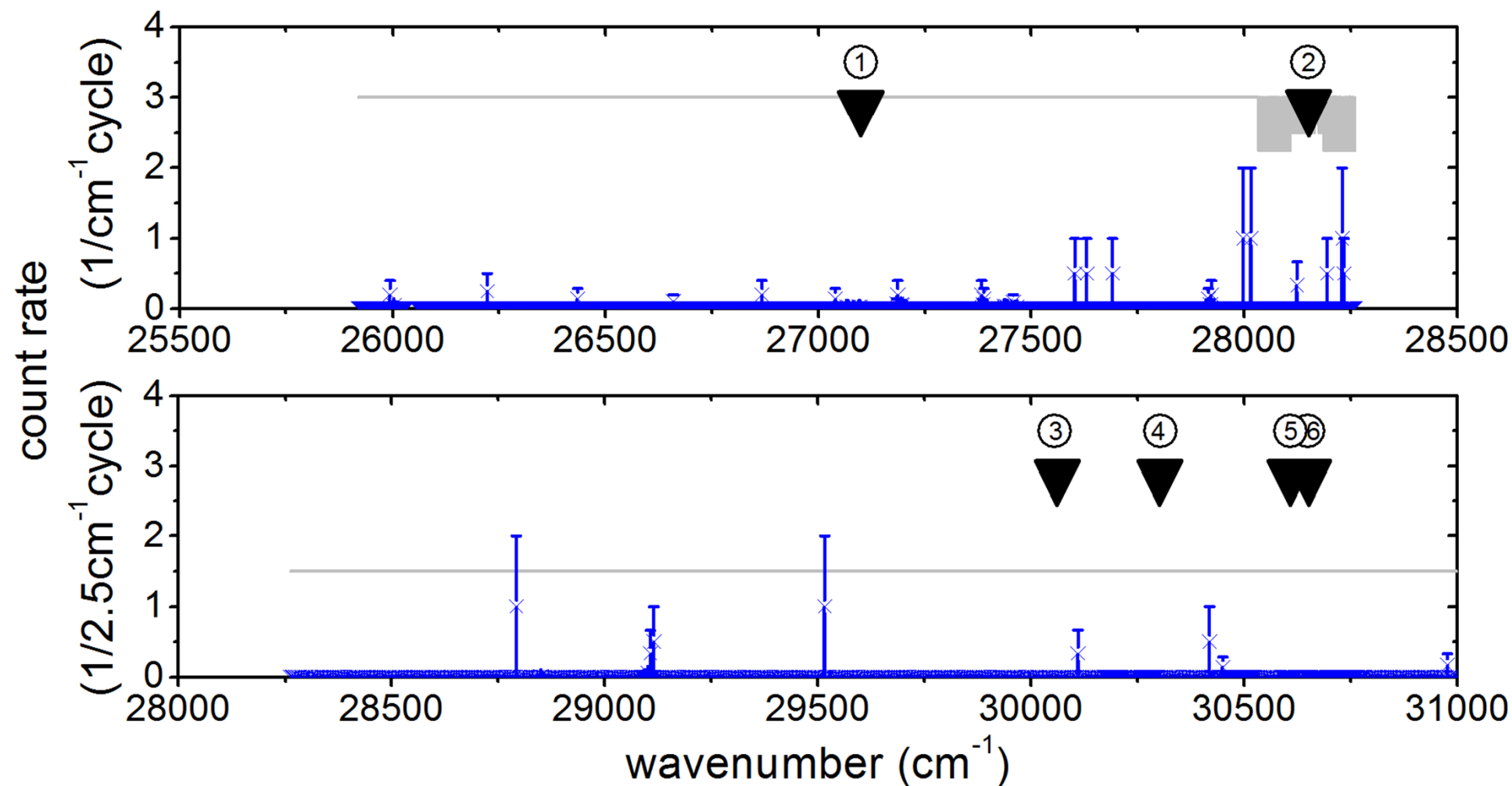
[7] (Extrapolation) J. Sugar, J. Chem. Phys. 60 (1974) 4103

Multi-Configuration Dirac Fock (MCDF)

Intermediate Hamiltonian Fock-space coupled-cluster (IHFSCC)

Relativistic coupled cluster (RCC)

Search for an Atomic Level



7. Conclusions

- Resonance Ionization Spectroscopy (RIS) in gas cells is a powerful laser spectroscopic method at samples in the order of only 10^{10} atoms or production rates as low as 1/s.

• $^{240-244}\text{fAm}$	0.9–14 ms	on-line	$10^6 - 10^7$	RADRIS
• ^{255}Fm	20.1 h	off-line	few 10^{10}	IGRIS

Prospects

- ^{254}No 55 s on-line $10^6 - 10^7$ RADRIS
- Method has been tested with an on-line laser spectroscopy experiment at the chemical homologue ^{155}Yb . An overall efficiency of $\sim 1\%$ has been measured.
- The experiment at ^{254}No at GSI for an atomic level search is well prepared but is difficult and **requires a lot of beam time.**
- Activities are also underway at KU Leuven (Belgium) within the Heavy Element Laser Ionization Project (HELIOS) to be performed at the SPIRAL-2 facility at GANIL (France).

no atomic levels known

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