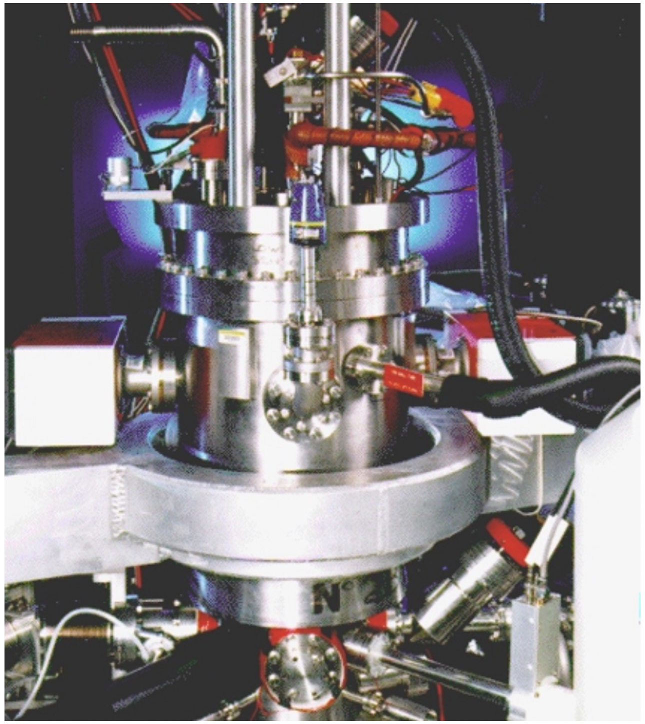
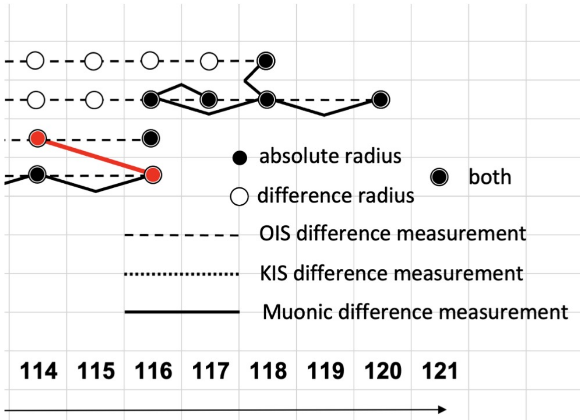
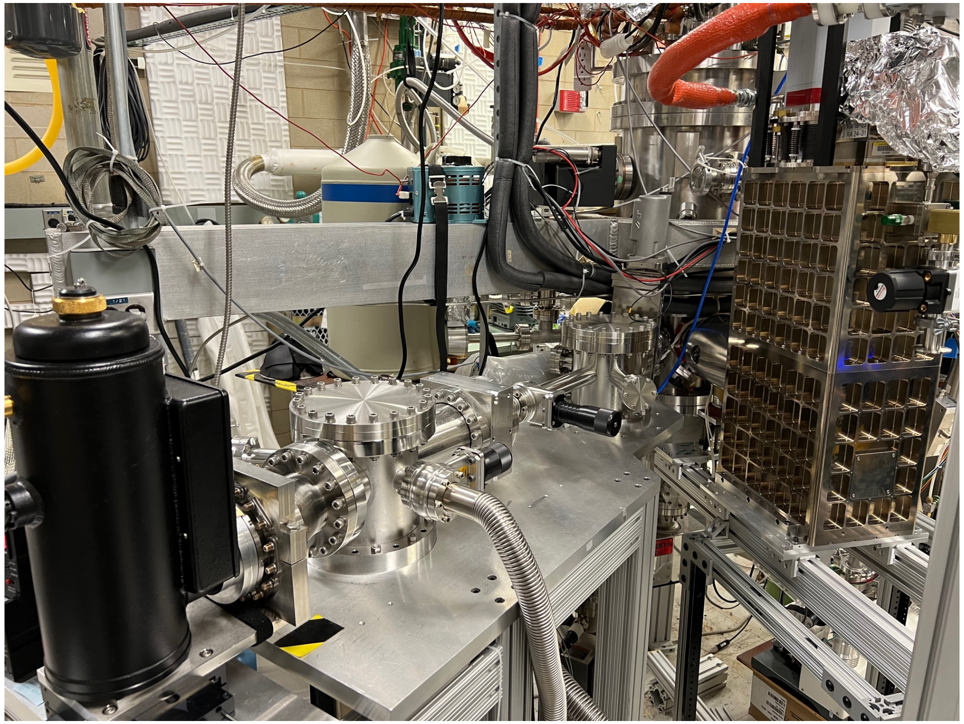
CHARGE RADII OF NUCLEI BY EUV AND X-RAY SPECTROSCOPY OF HIGHLY CHARGED IONS

Endre Takacs

Hunter Staiger, Steven Blundell, Dipti, Gerald Gwinner, Roshani Silwal, Alain Lapierre, John Gillaspy, Galen O'Neil, Joseph Tan, Yuri Ralchenko

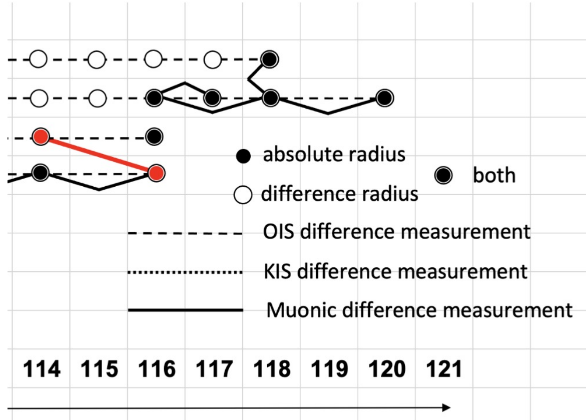




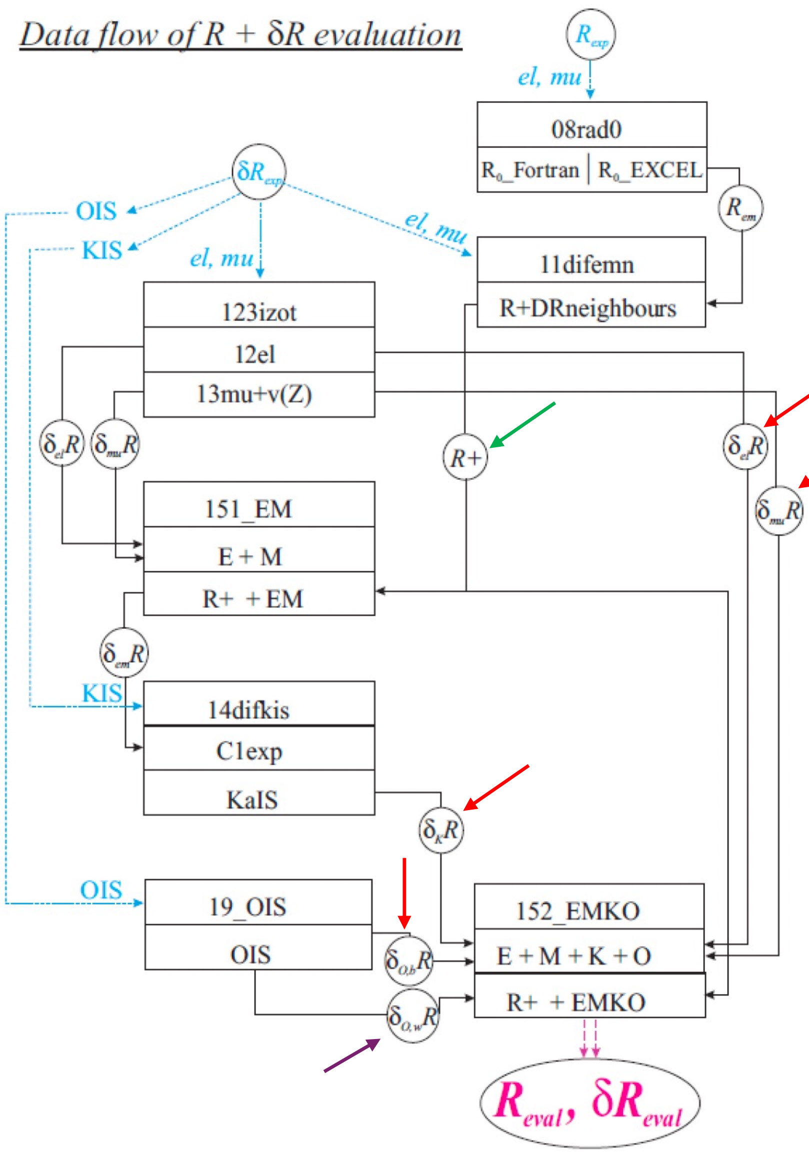


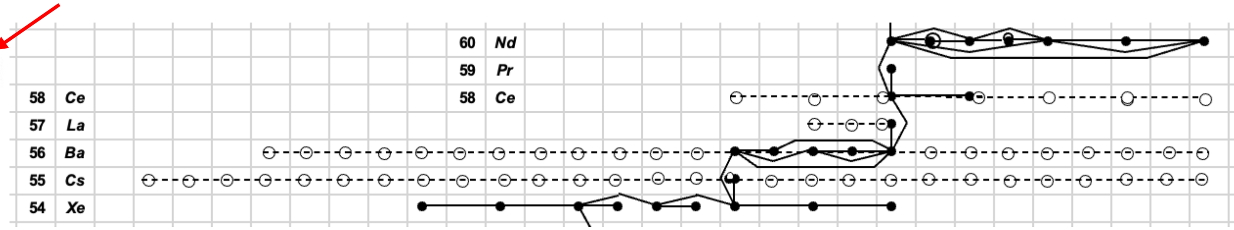
Outline

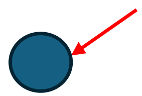
- Note on the Flowchart of the Angeli 2004 Charge Radius Evaluation
- Highly Charged Ions
- Short history of electron beam ion traps
- Spectroscopy of highly charged ions
  - EUV, X-ray
- Nuclear Charge Radius Landscape
- Measurements Using Highly Charged Ions
  - Previous Methods
  - Nuclear Charge Radius Difference – Na-like Ion Spectroscopy
  - Absolute Nuclear Charge Radius – Na-like Ion Spectroscopy
- Cross-element constraints
- Future Directions



Data flow of R + δR evaluation



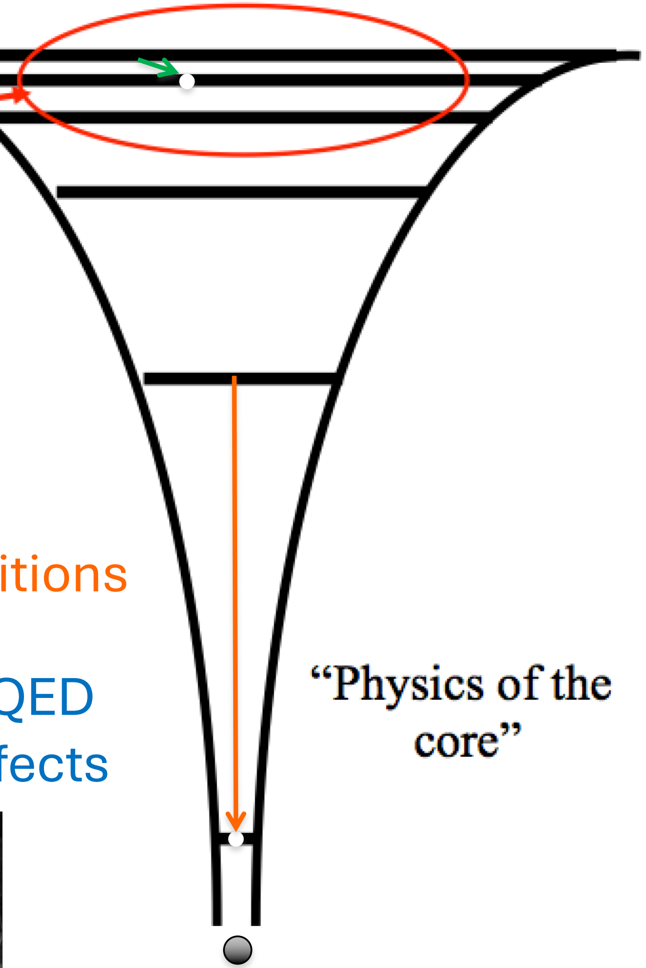




HIGHLY CHARGED IONS

"Everyday atomic physics"

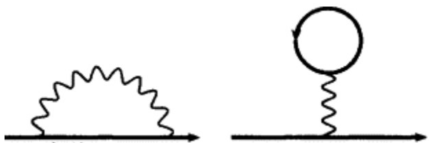
Optical transitions

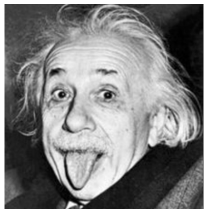


Photon energy ~ Z^2 (E1)

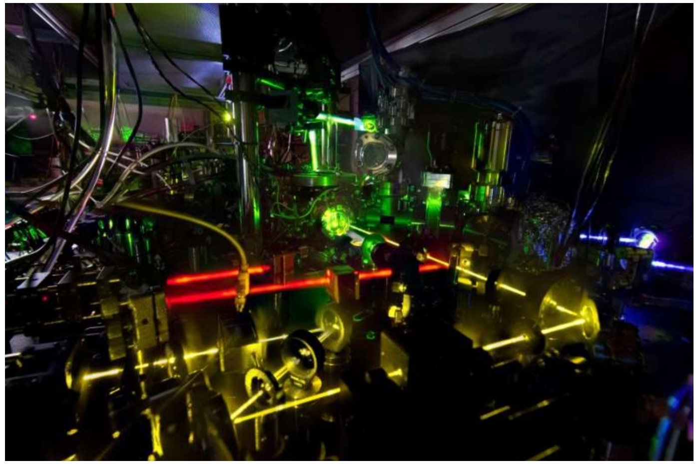
X-ray transitions

Relativistic, QED
& nuclear effects





"Physics of the core"



NIST ytterbium lattice clock

X-ray image of the solar corona

ELECTRON BEAM ION TRAP HISTORY

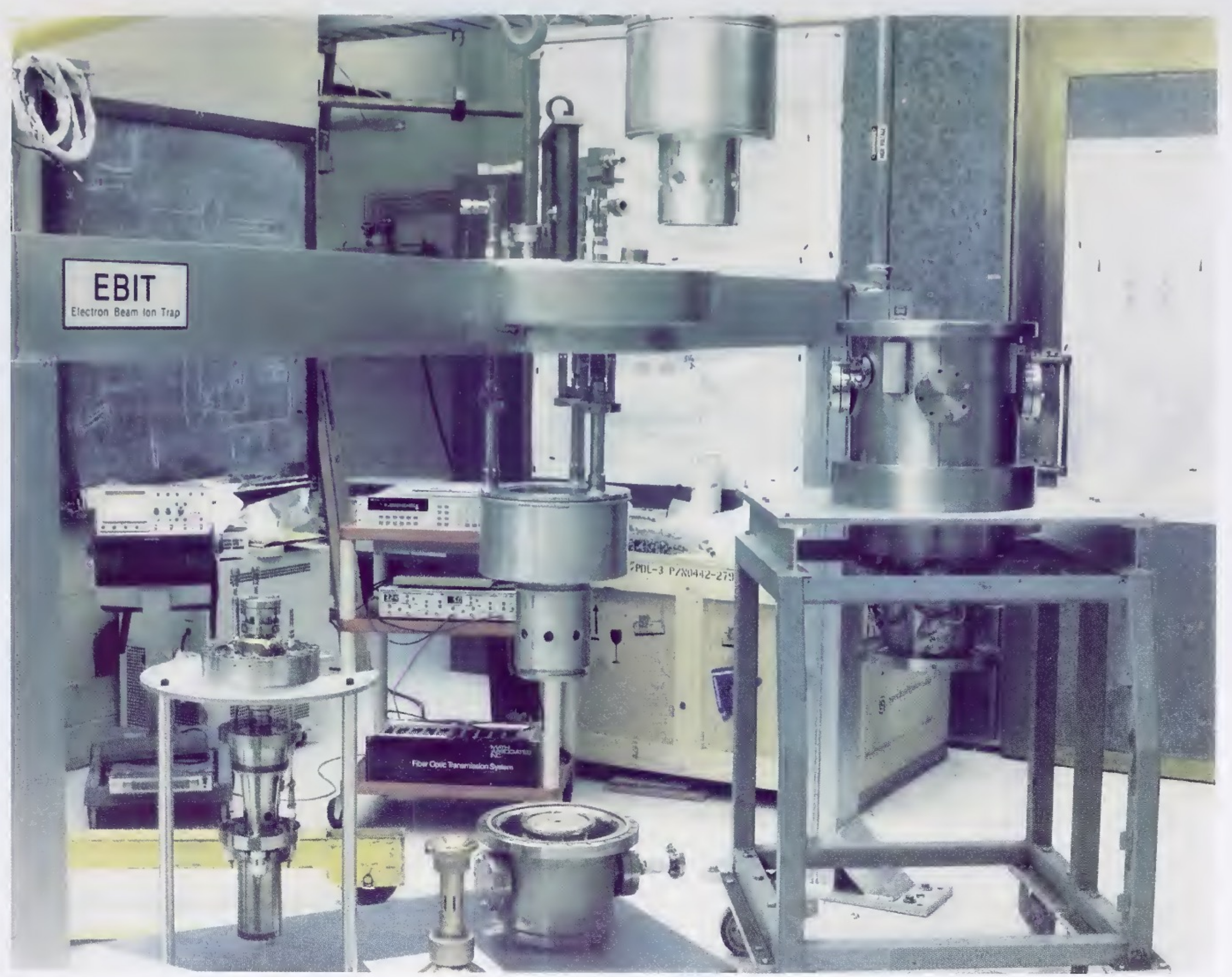
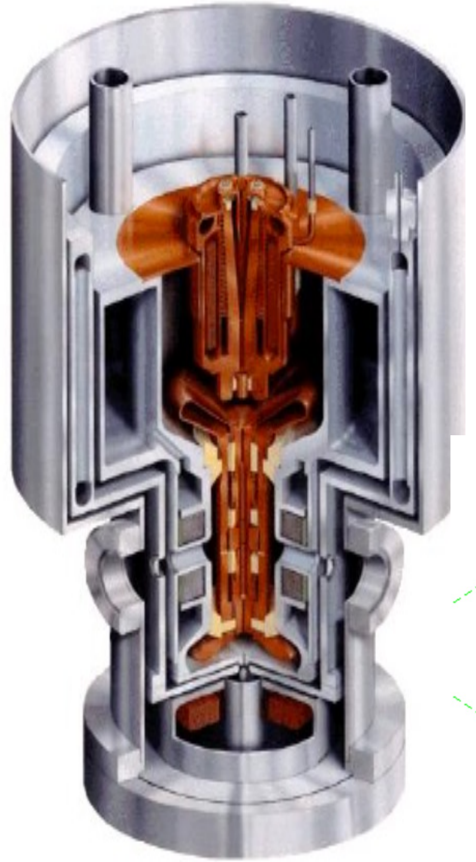
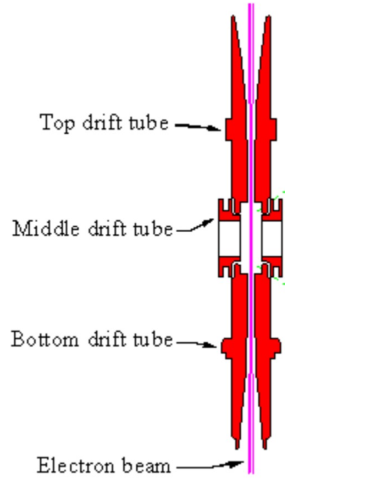
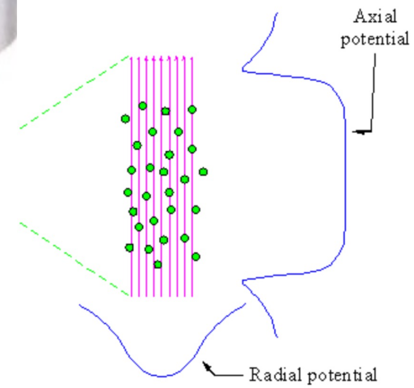


Figure 1. The NIST EBIT, just before final assembly of the 6 major subsections (clockwise from the bottom: electron gun, drift tube assembly, collector, liquid helium insert, liquid nitrogen shield, outer vacuum can).

1992 NIST-NRL EBIT
(1st outside LLNL)





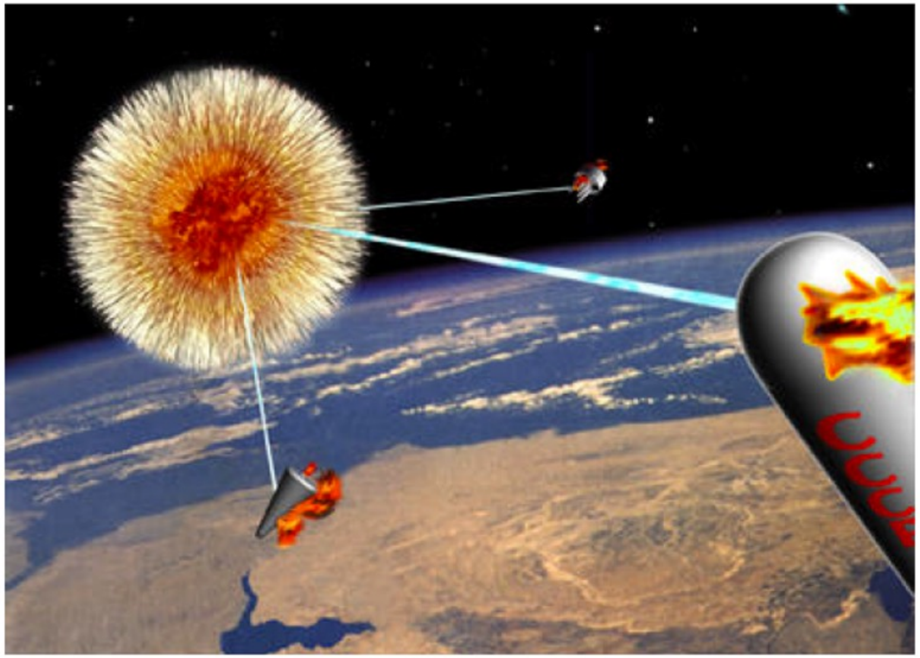


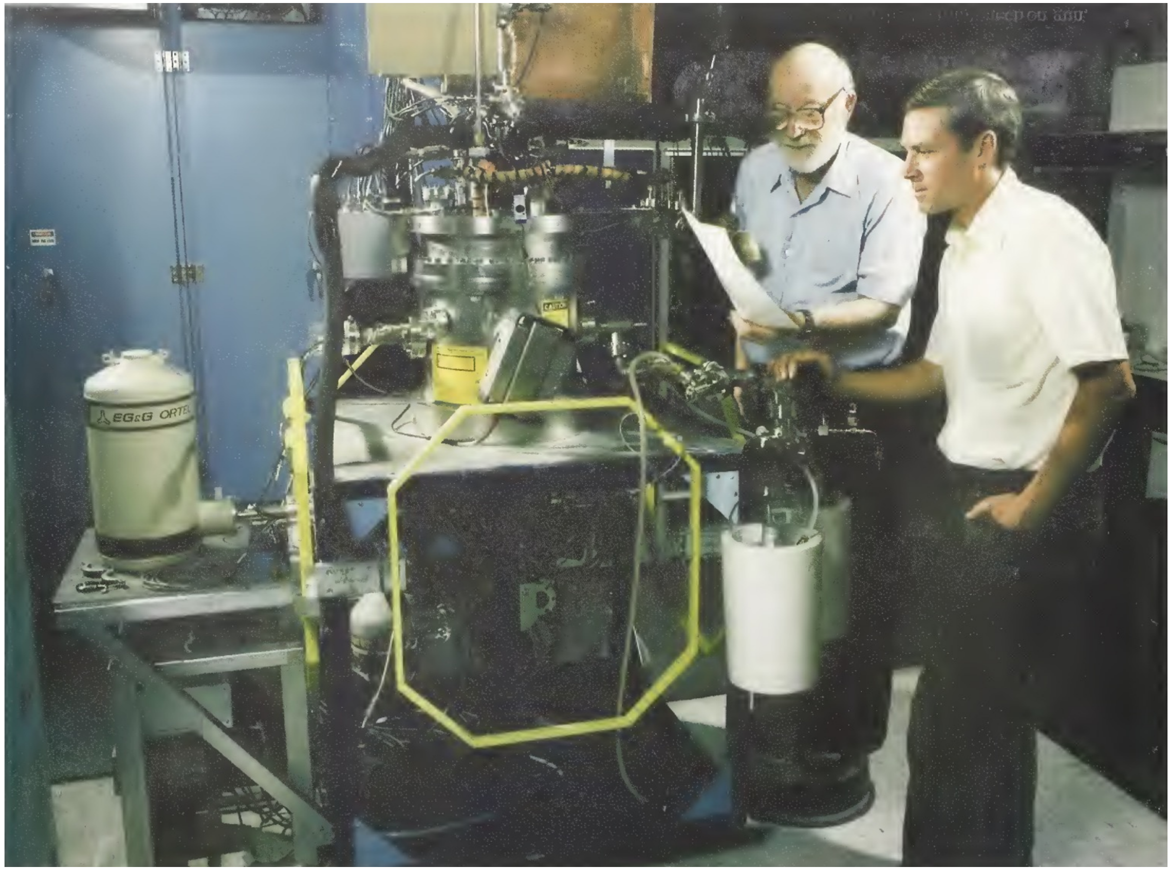
HCI spectroscopy

ELECTRON BEAM ION TRAP HISTORY

Strategic Defense Initiative

Nicknamed as **Star Wars Program**, was first initiated on March 23, 1983 under President Ronald Reagan. The intent of this was to develop a sophisticated anti-ballistic missile system in order to prevent missile attacks from other countries, specifically the Soviet Union.



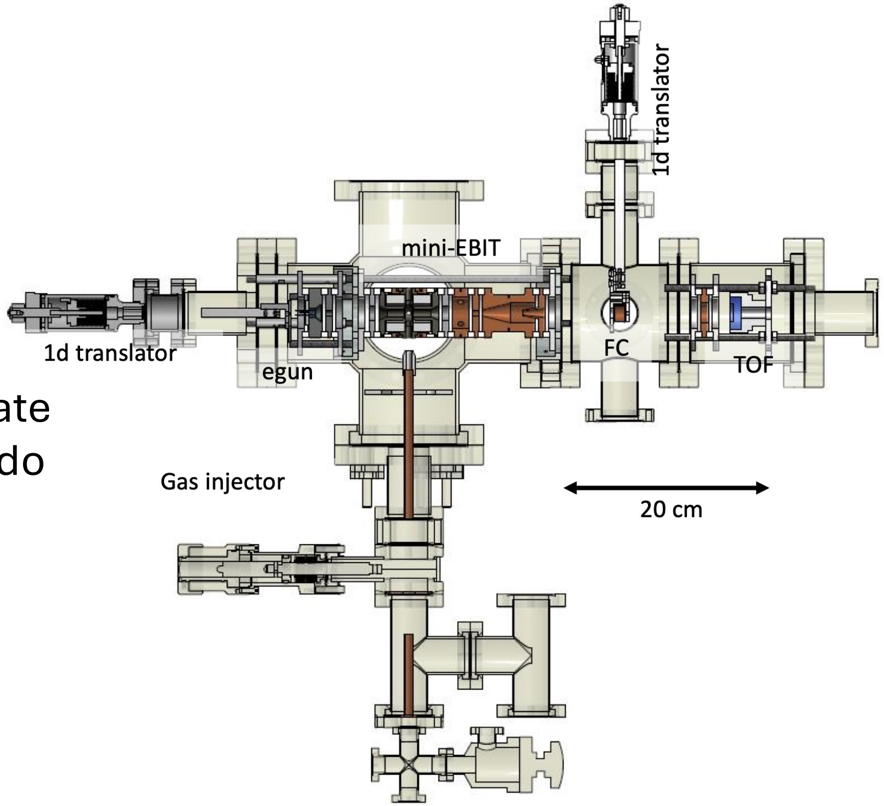


Mort Levine and Ross Marrs 1989 with the first EBIT

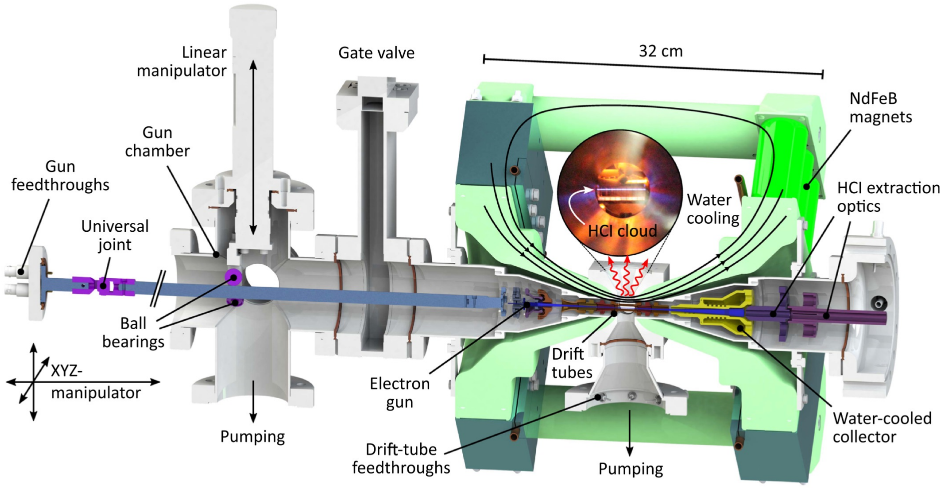
Lawrence Livermore National Laboratory

NEW COMPACT EBIT SOURCES

- NIST
- Heidelberg
- Tokyo
- Shanghai
- Colorado State
- Univ. Colorado
- Clemson

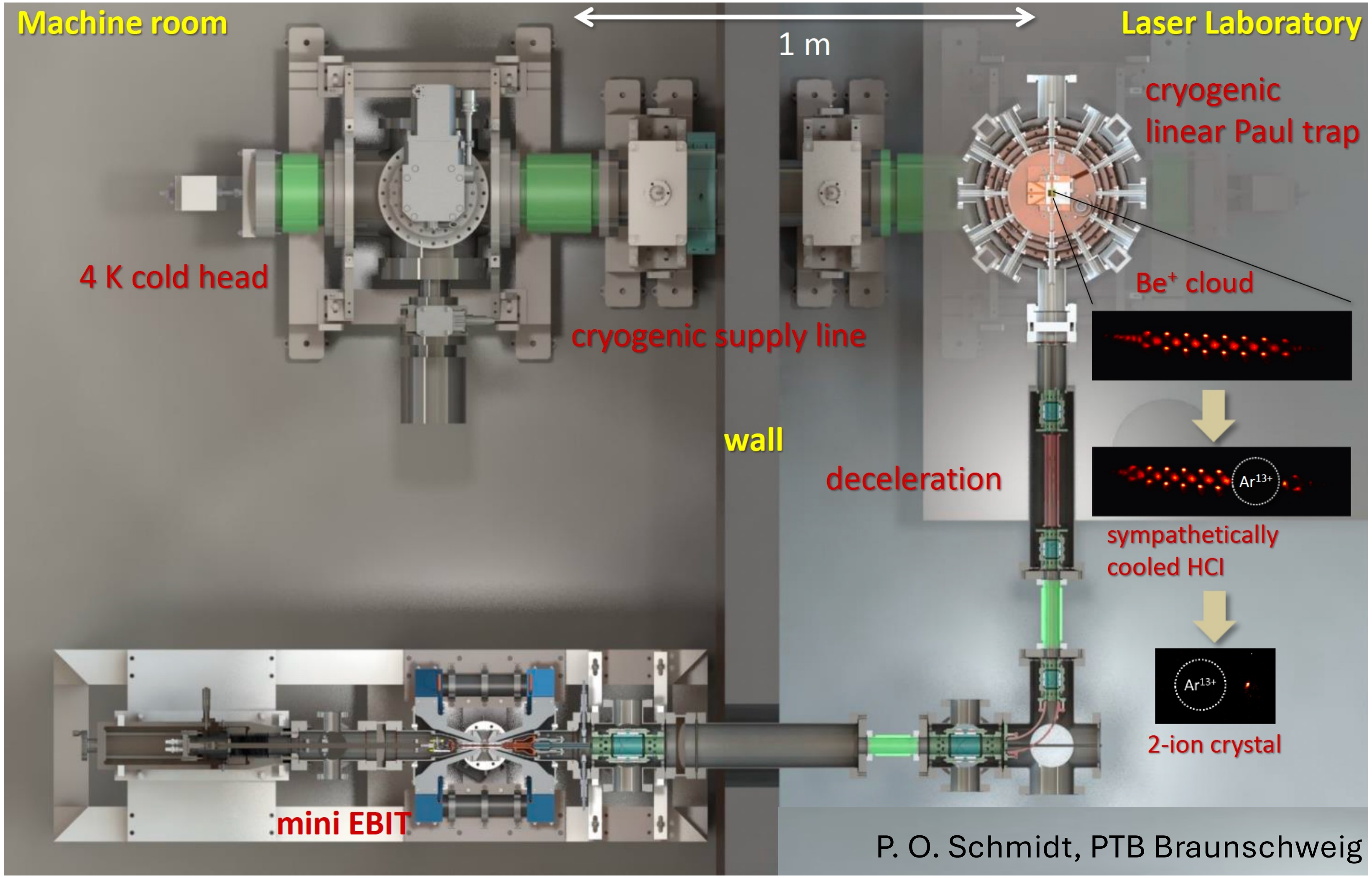


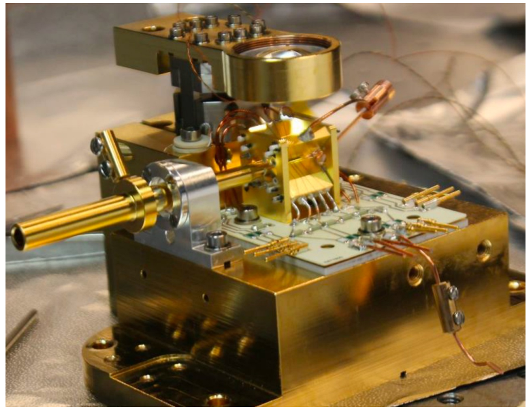
Hoogerheide and Tan, Journal of Physics: Conference Series **583** (2015) 012044

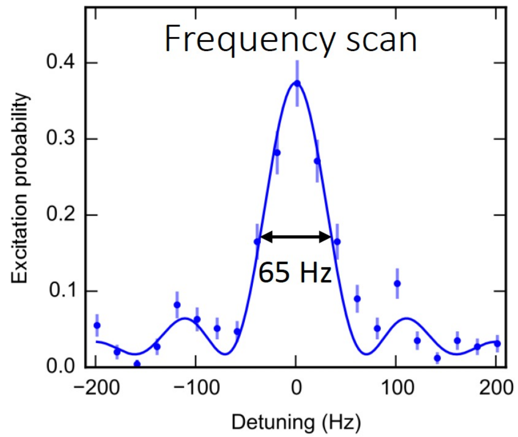


Micke et al., Rev. Sci. Instrum. **89**, 063109 (2018)

OPTICAL CLOCK BASED ON Ar^{13+} HCI (PTB AND MPI)

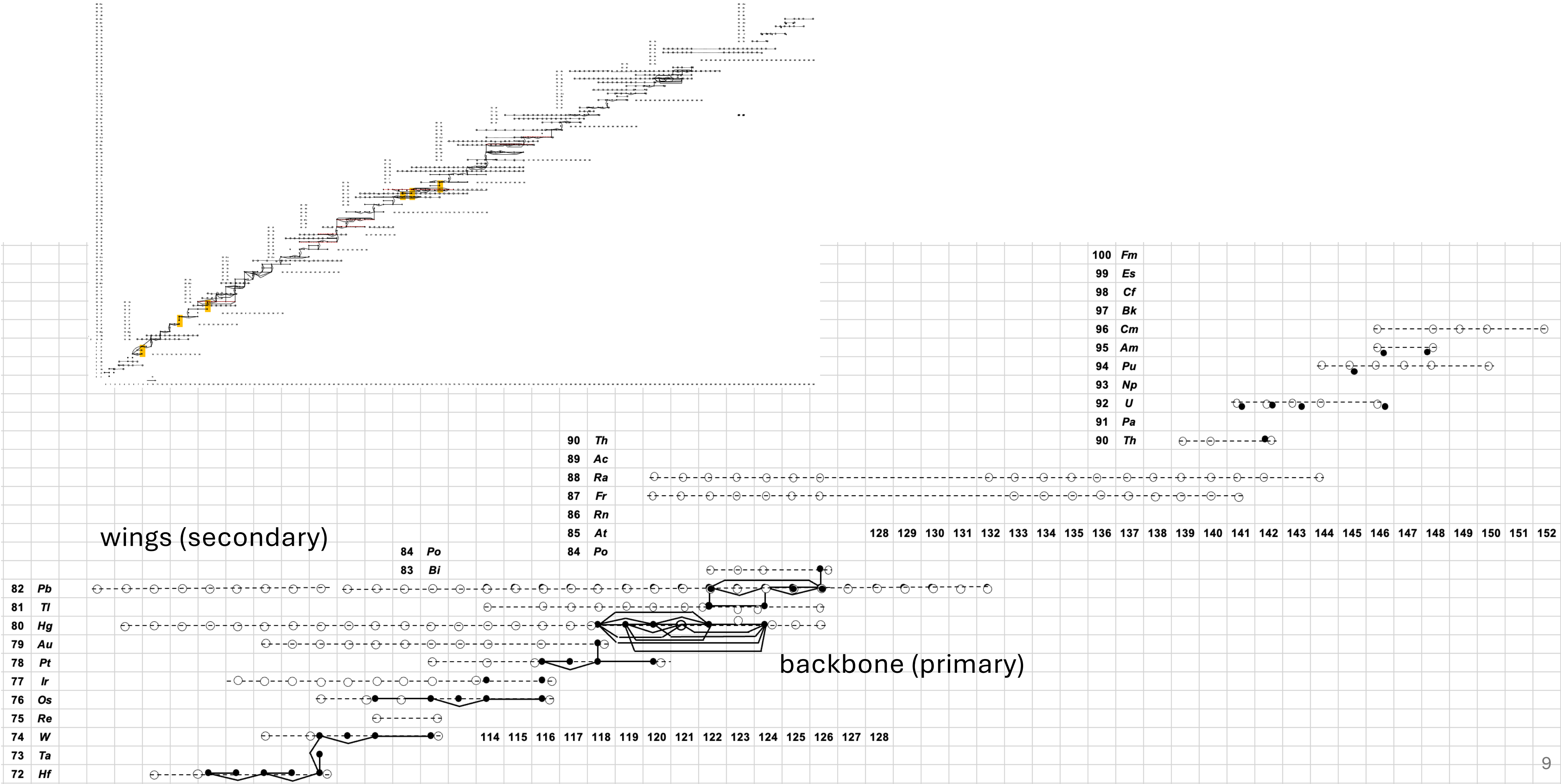






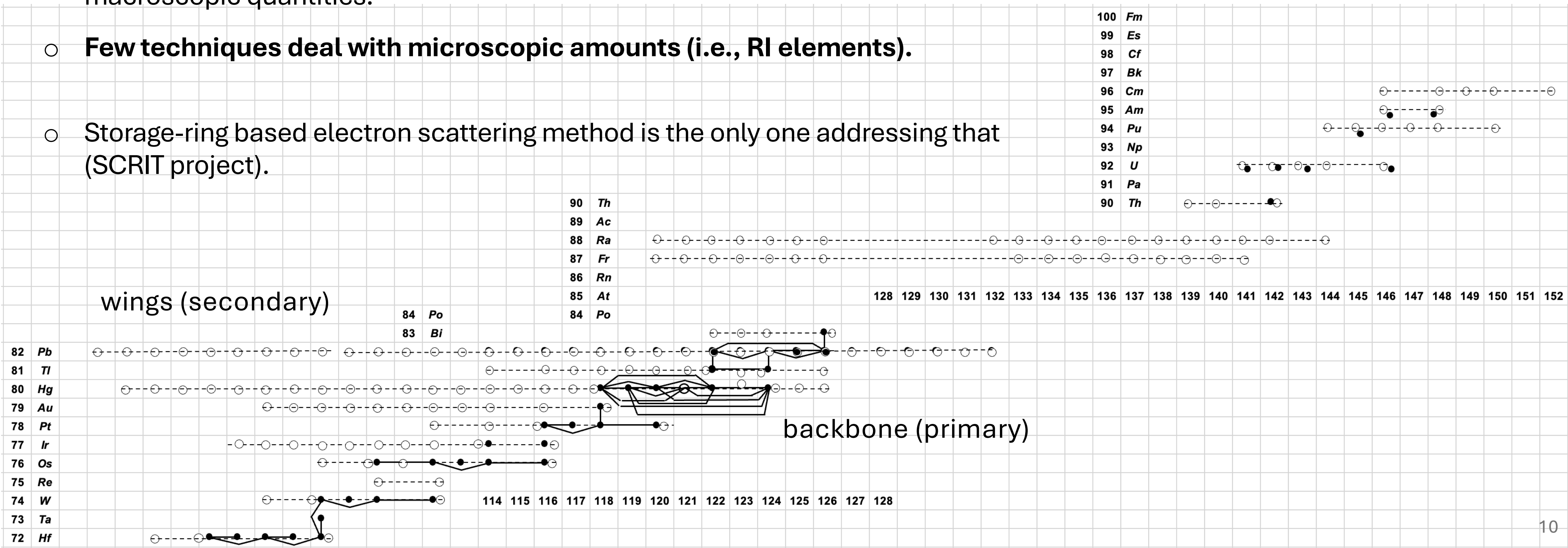
1 part in 10^{16} accuracy

MOTIVATIONS FOR A NEW CHARGE RADIUS MEASUREMENT METHODS

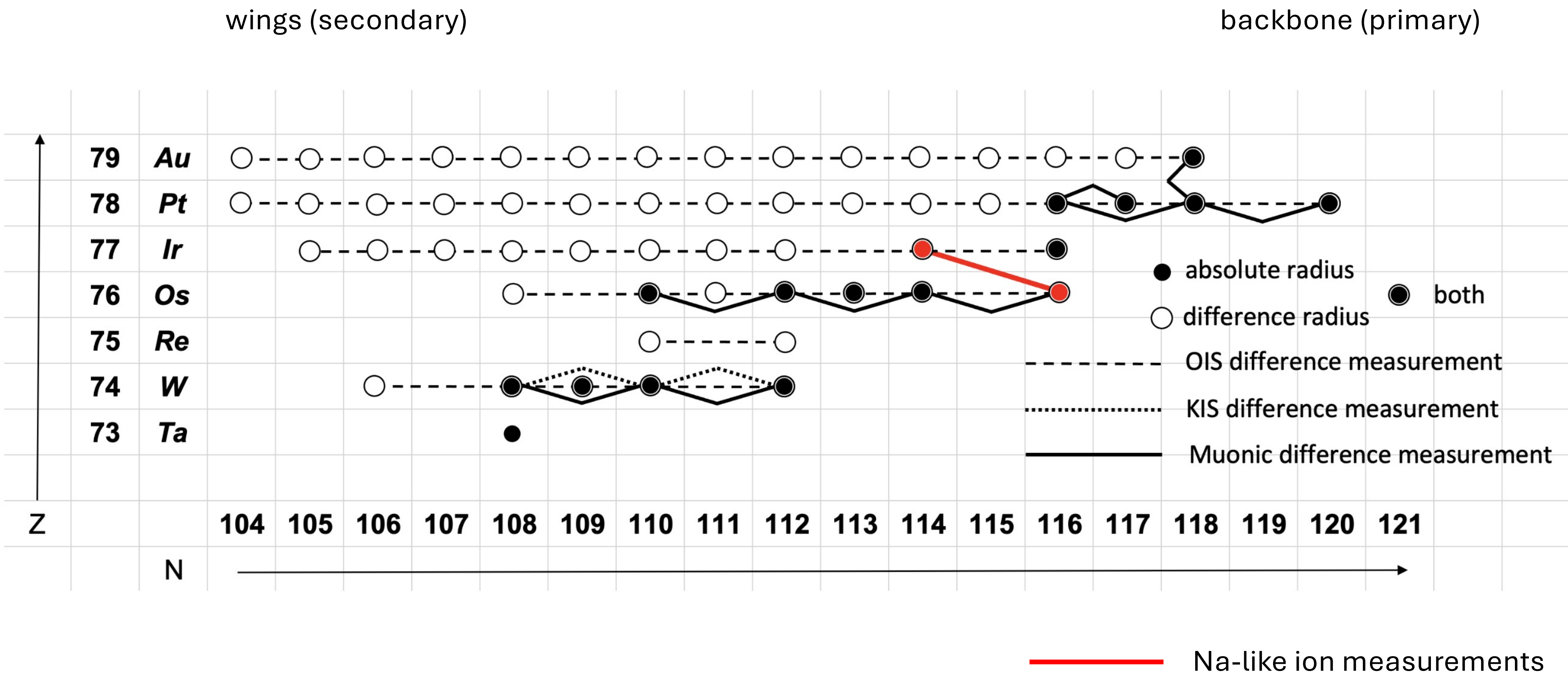


MOTIVATIONS FOR A NEW CHARGE RADIUS MEASUREMENT METHODS

- Except for a few isotopes, no **absolute** charge radius measurements for unstable isotopes exist heavier than Bi.
  - The absolute charge radii of francium, radium, and radon have never been measured.
  - <u>Apparent reason</u>: current techniques (electron scattering / muonic x-ray spectro.) need macroscopic quantities.
  - **Few techniques deal with microscopic amounts (i.e., RI elements).**
  - Storage-ring based electron scattering method is the only one addressing that (SCRIT project).

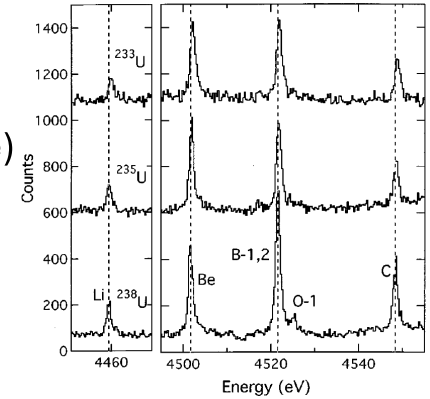


NUCLEAR CHARGE RADIUS DIFFERENCE CONSTRAINTS



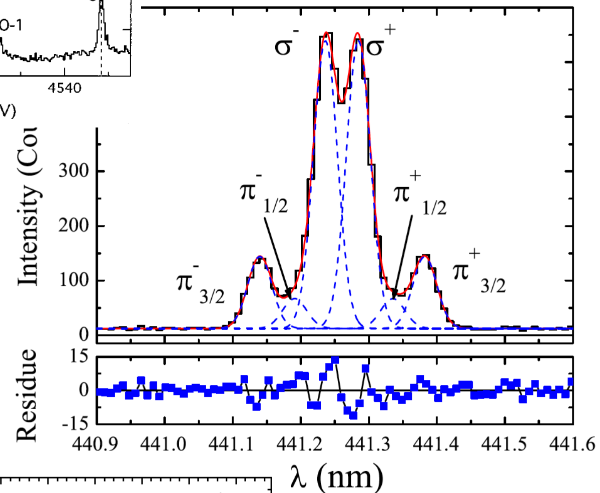
HIGHLY CHARGED IONS AND NUCLEAR CHARGE RADIUS

Note: mass shift scales linearly with Z – field shift scales Z^4



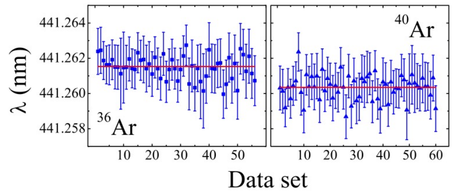
MS/FS ~ 0.03

- Precision X-ray spectroscopy of few electron U ions (Li- though C-like)
  S. R. Elliott *et al*. Phys. Rev. C **57**, 583 (1998)

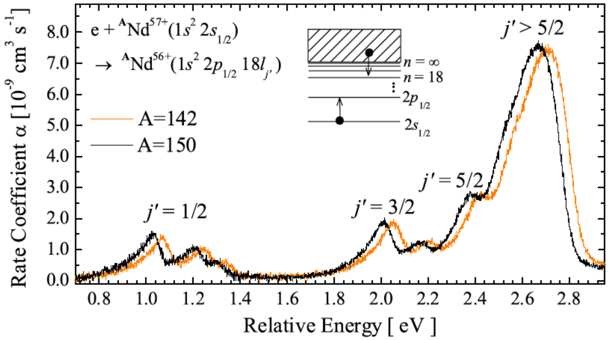


- Magnetic-dipole transitions of Be-like and B-like Ar isotopes in the visible range
  R. S. Orts *et al*., Phys. Rev. Lett. **97**, 103002 (2006)
  Dissertation, Inst. fur Kernphysik Frankfurt (2005)

MS/FS ~ 100



- Dielectronic recombination measurements of quasi few-electron ions (K-like Pb, Li-like Nd)
  R. Schuch et al., Phys. Rev. Lett. **95**, 183003 (2005)
  C. Brandau *et al*., Phys. Rev. Lett. **100**, 073201 (2008)



MS/FS ~ 0.5

MEASUREMENTS NEEDED FOR FUNDAMENTAL SYMMETRY TESTS

- Francium and radium are candidates in searches for physics beyond the Standard Model:
    - Ra-225: Permanent Electric Dipole Moments (EDM)
    - Fr: Atomic Parity Non-Conservation (APNC)
- The absolute charge radii of Fr and Ra were never directly measured.
- The absolute charge radius of Fr in the literature is obtained from extrapolations.
- **Need for absolute charge radius measurements.**

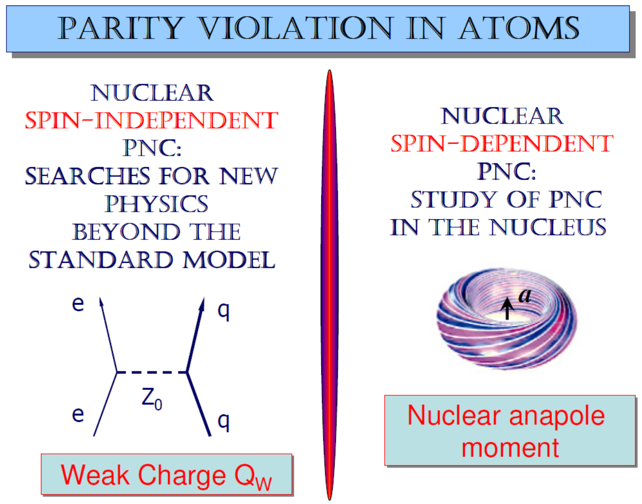
Why is it important ?

Atomic matrix element

$$H^{(1)} = \frac{G}{2\sqrt{2}} \gamma_5 \, Q_W \, \rho(r)$$

Weak coupling constant for symmetry tests

Charge density distribution



Atomic matrix element

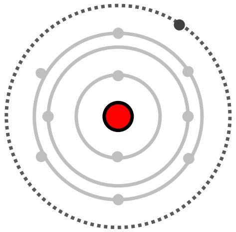
$$H^{(2)} = \frac{G}{\sqrt{2}} \kappa_2 \, \alpha \cdot \mathbf{I} \, \rho(r)$$

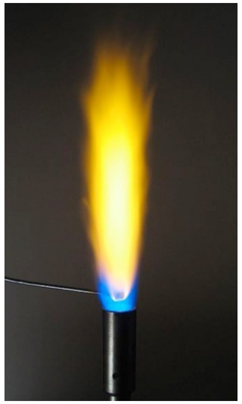
Nuclear spin

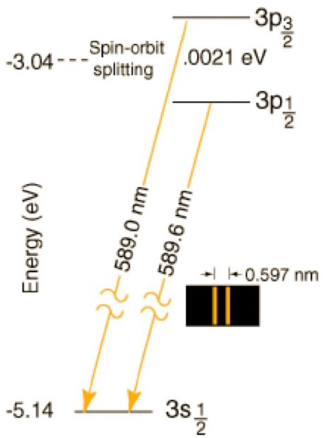
Charge density distribution

M. A. Bouchiat & C. Bouchiat, Rep. Prog. Phys. **60** (1997) 1351

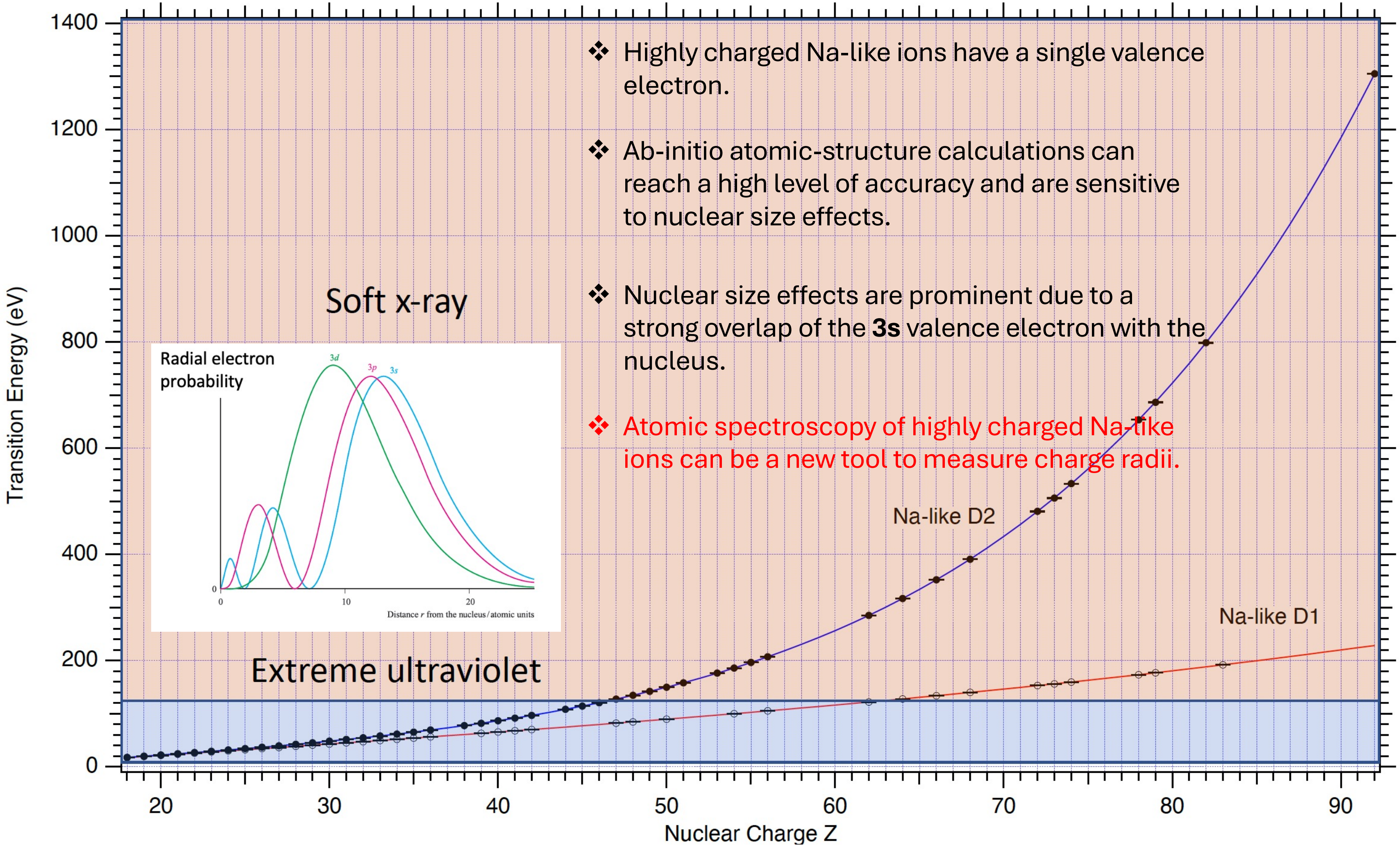
WHY NA-LIKE IONS THEORETICALLY?







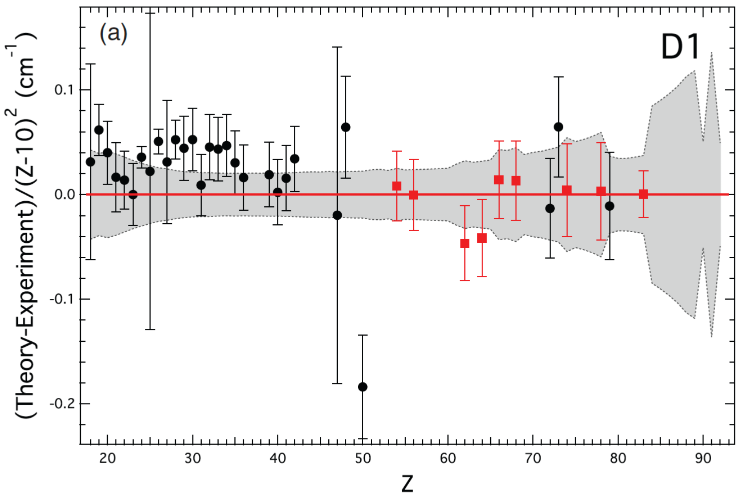
- Highly charged Na-like ions have a single valence electron.
- Ab-initio atomic-structure calculations can reach a high level of accuracy and are sensitive to nuclear size effects.
- Nuclear size effects are prominent due to a strong overlap of the **3s** valence electron with the nucleus.
- Atomic spectroscopy of highly charged Na-like ions can be a new tool to measure charge radii.

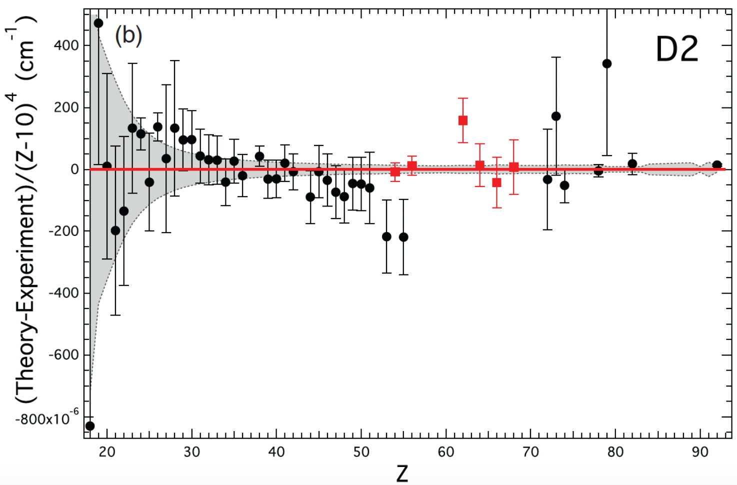


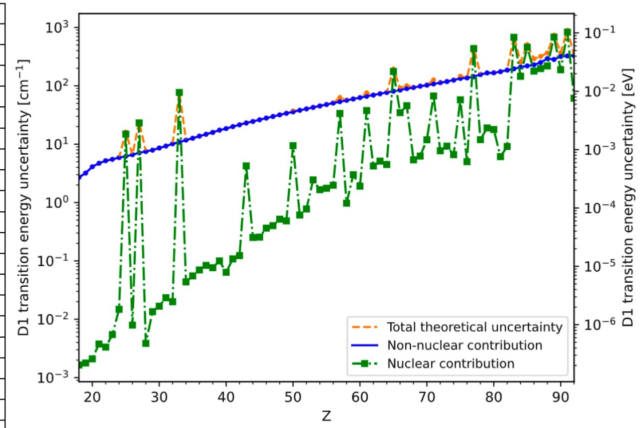
STATE-OF-THE-ART AB INITIO THEORIES ARE AVAILABLE
(RMBPT, MCDHF, S-MATRIX)

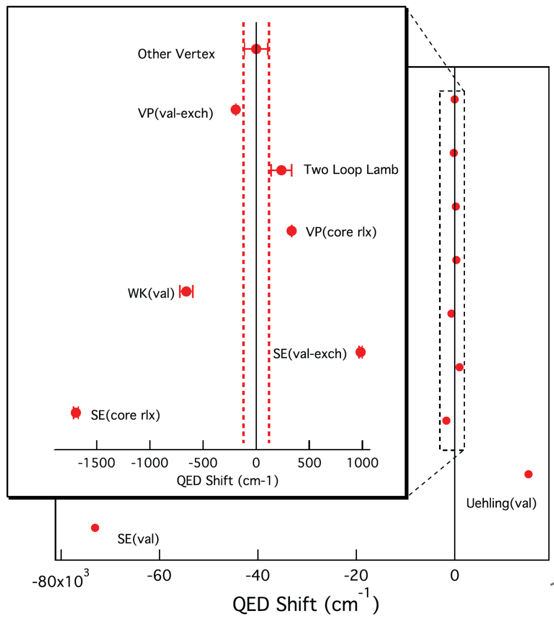
TABLE III. Contributions (cm^{-1}) to the total calculated wave numbers σ and their estimated uncertainties for Bi ($Z = 83$). Values between the dotted lines are from the QED terms.

	$\sigma(D_1)$	Unc.	$\sigma(D_2)$	Unc.
Dirac Hartree Fock	1 559 528	37	6 836 929	41
B(1)	52 830	0	−1481	0
B(rpa)	−1238	0	−299	0
BB(rpa)	−127	0	15	0
Ret(1)	499	0	−8402	0
Ret(rpa)	53	0	−70	0
Other retardation	0	107	0	209
CC(2)	−2616	2	−354	1
BC(2)	−544	1	−265	0
CCC(3)	16	0	−7	0
Nuclear recoil	−68	25	−76	28
SE(val)	−73 091	3	−71 432	3
Uehling (val)	15009	0	17 318	0
WK (val)	−657	62	−781	50
SE (val-exch)	983	14	1029	14
VP (val-exch)	−192	0	−200	0
SE (core rlx)	−1697	23	−814	11
VP (core rlx)	334	0	186	0
Other vertex	0	110	0	73
Two-loop Lamb (val)	238	96	222	90
Total	1 549 261	199	6 771 519	249









Gillaspy et al., Physical Review A **87**, (2013) 062503

WHY NA-LIKE IONS EXPERIMENTALLY?

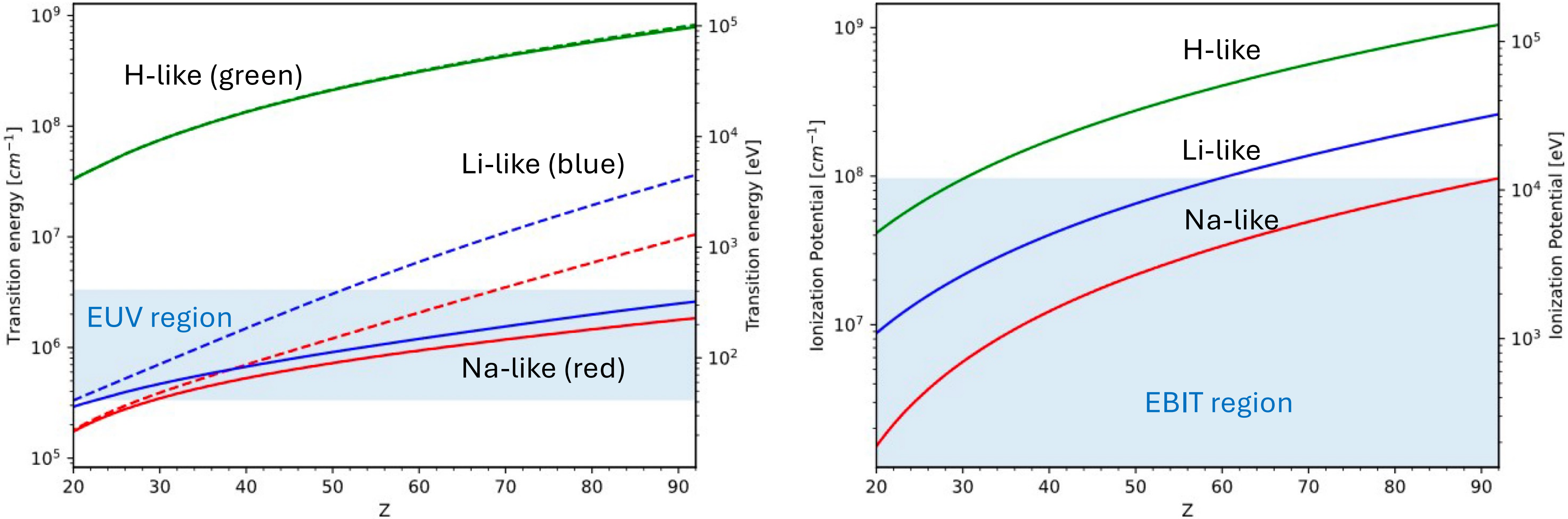
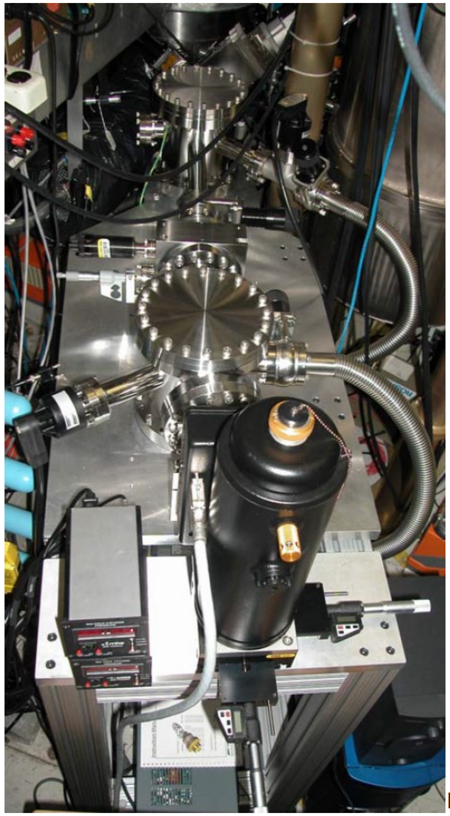


Figure 1: (Left) Plot of transition energies from Z = 20 to Z = 92, with Na-like D1 $3s\ ^2S_{1/2} \rightarrow 3p\ ^2P_{1/2}$ [23] (red solid), Na-like D2 $3s\ ^2S_{1/2} \rightarrow 3p\ ^2P_{3/2}$ [23] (red dashed), Li-like $1s^22s\ ^2S_{1/2} \rightarrow 1s^22p\ ^2P_{1/2}$ [24] (blue solid), Li-like $1s^22s\ ^2S_{1/2} \rightarrow 1s^22p\ ^2P_{3/2}$ [24] (blue dashed), H-like $1s\ ^2S_{1/2} \rightarrow 2p\ ^2P_{1/2}$ [25] (green solid), and H-like $1s\ ^2S_{1/2} \rightarrow 2p\ ^2P_{3/2}$ [25] (green dashed). The shaded blue region indicates EUV range (3 to 30 nm). (Right) Plot of ionization potential necessary to create Na-like (red), Li-like (blue) and H-like (green) ions. The shaded blue region indicates the typical range of optimum electron beam energies used to generate highly charged ions [26]).

Hosier et al., Journal of Physics B **57**, (2024) 195001

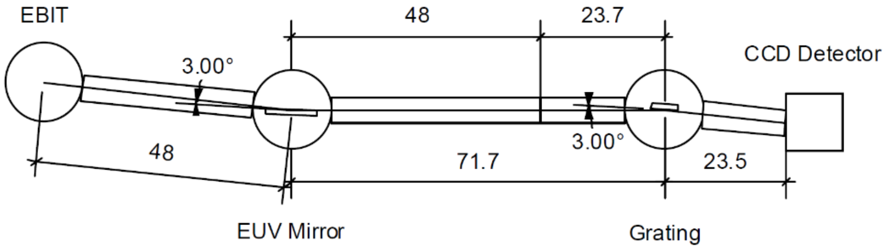
MEASUREMENTS USING HIGHLY CHARGED NA-LIKE IONS

NIST EBIT

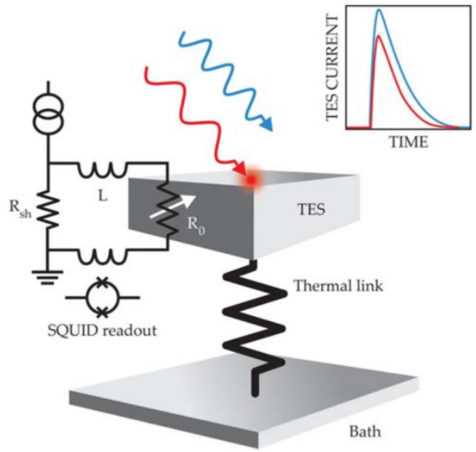


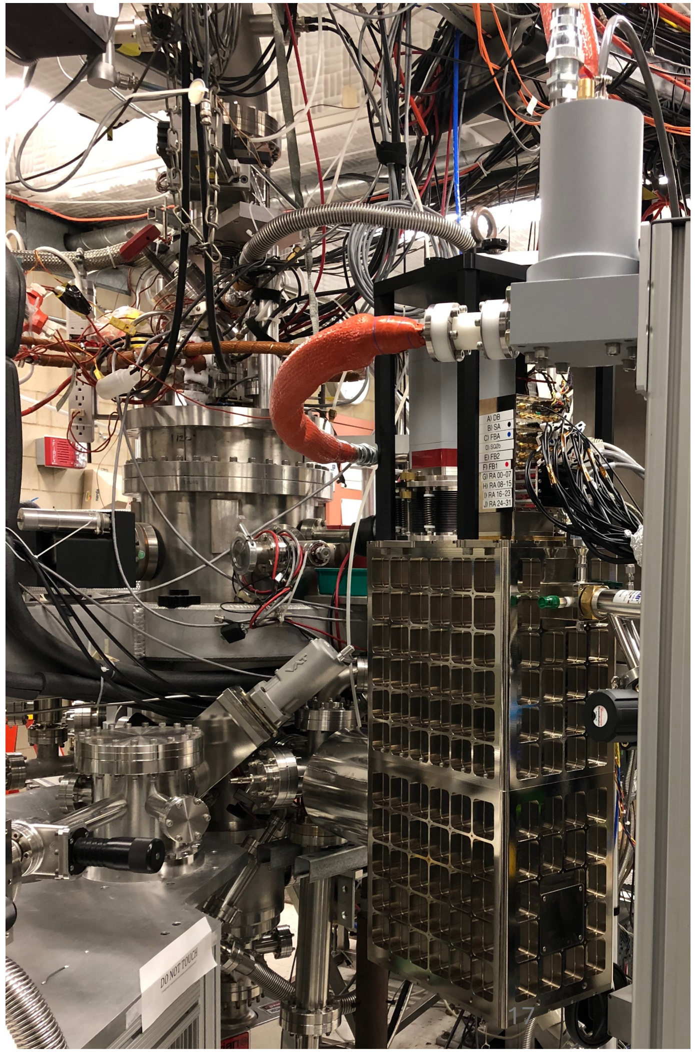
EUV

lit



X-ray





EXPERIMENTAL AND THEORETICAL D LINE SEPARATIONS

Silwal R et al., Phys. Rev. A **98** (2018) 052502; Silwal R et al., Phys. Rev. A **101** (2020) 062512

$\delta E_k^{A,A'}(Exp.) = E_k^{A} - E_k^{A'}$ = Mass shift + Field shift

$$\delta E_k^{A,A'} = \delta E_{k,MS}^{A,A'} + \delta E_{k,FS}^{A,A'}$$

$$= (\mathrm{NMS} + \mathrm{SMS})\frac{(\mathrm{M'} - \mathrm{M})}{\mathrm{MM'}} + \mathrm{F}\lambda^{A,A'}$$

$$\delta E_{FS}^{A,A'} = F_0\delta\langle r^2\rangle^{A,A'} + F_2\delta\langle r^4\rangle^{A,A'} + F_6\delta\langle r^6\rangle^{A,A'} + F_8\delta\langle r^8\rangle^{A,A'} + \ldots$$

$$= \left(F_0 + F_2\frac{\delta\langle r^4\rangle^{A,A'}}{\delta\langle r^2\rangle^{A,A'}} + F_6\frac{\delta\langle r^6\rangle^{A,A'}}{\delta\langle r^2\rangle^{A,A'}} + F_8\frac{\delta\langle r^8\rangle^{A,A'}}{\delta\langle r^2\rangle^{A,A'}} + \ldots\right)\delta\langle r^2\rangle^{A,A'}$$

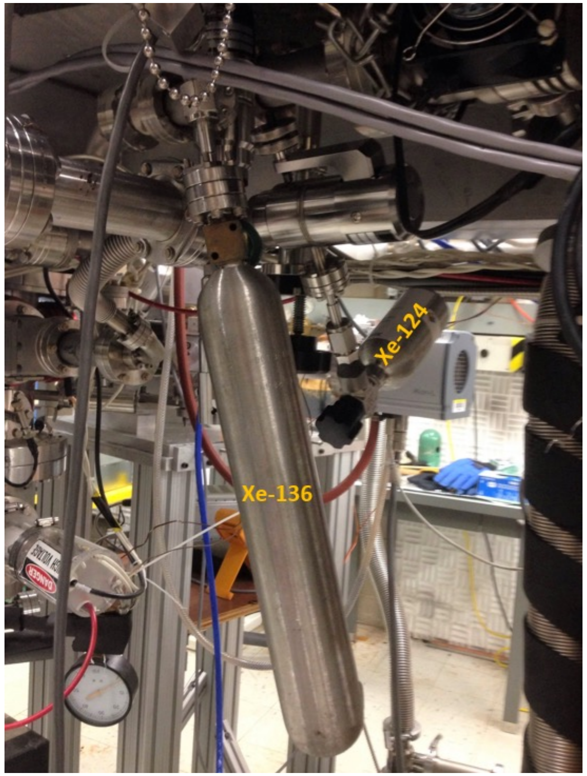
$$= F\delta\langle r^2\rangle^{A,A'}$$

$$\delta\langle r^2\rangle^{A,A'} = \frac{\delta E_k^{A,A'}(Exp.) - \delta E_{k,MS}^{A,A'}}{F}$$

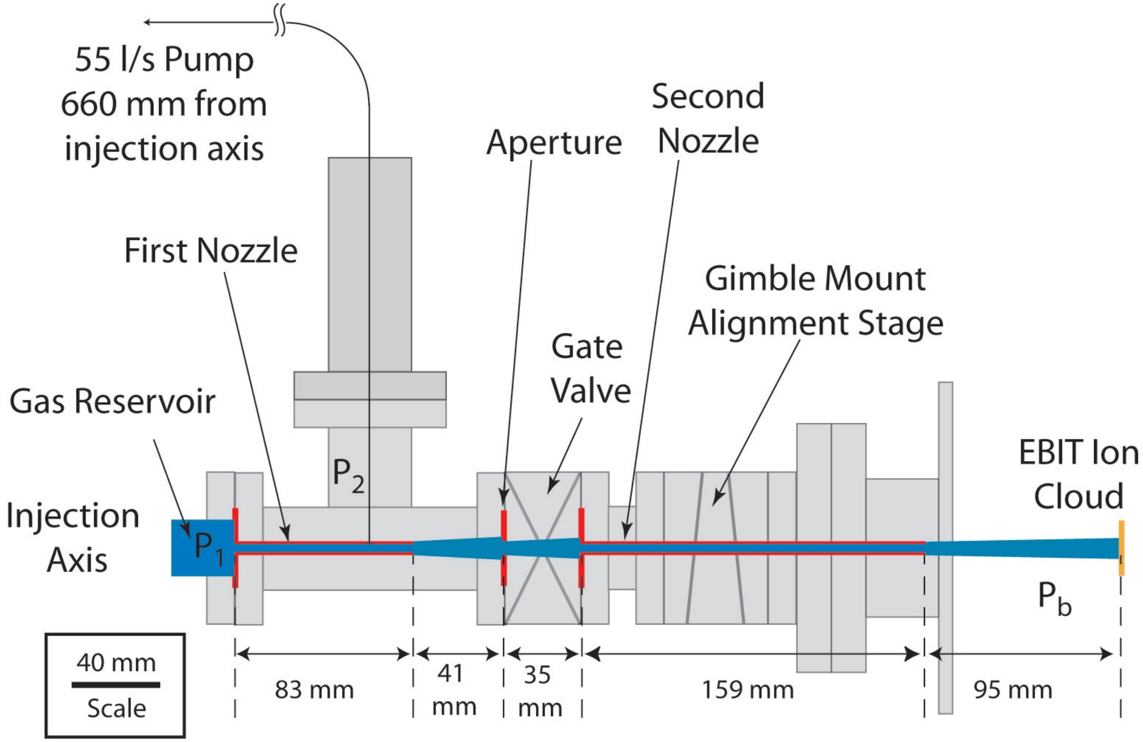
RMBPT or MCDHF calculates these accurately for Na-like ions (with uncertainties)

(Steven Blundell RMBPT, Dipti GRASP2K)

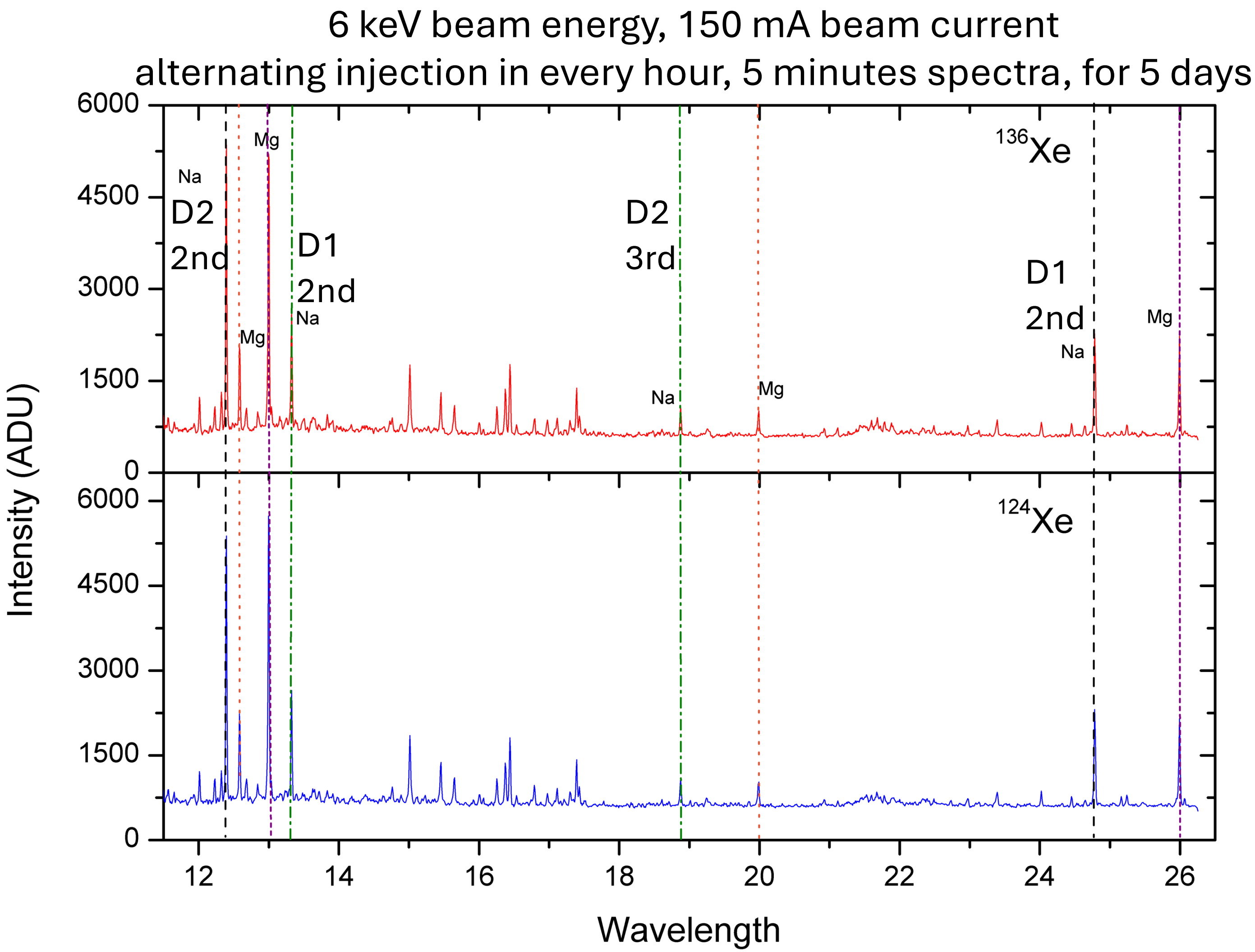
ISOTOPICALLY PURE Xe^{136} AND Xe^{124} NEUTRALS WERE INJECTED USING A BALLISTIC GAS INJECTION SYSTEM



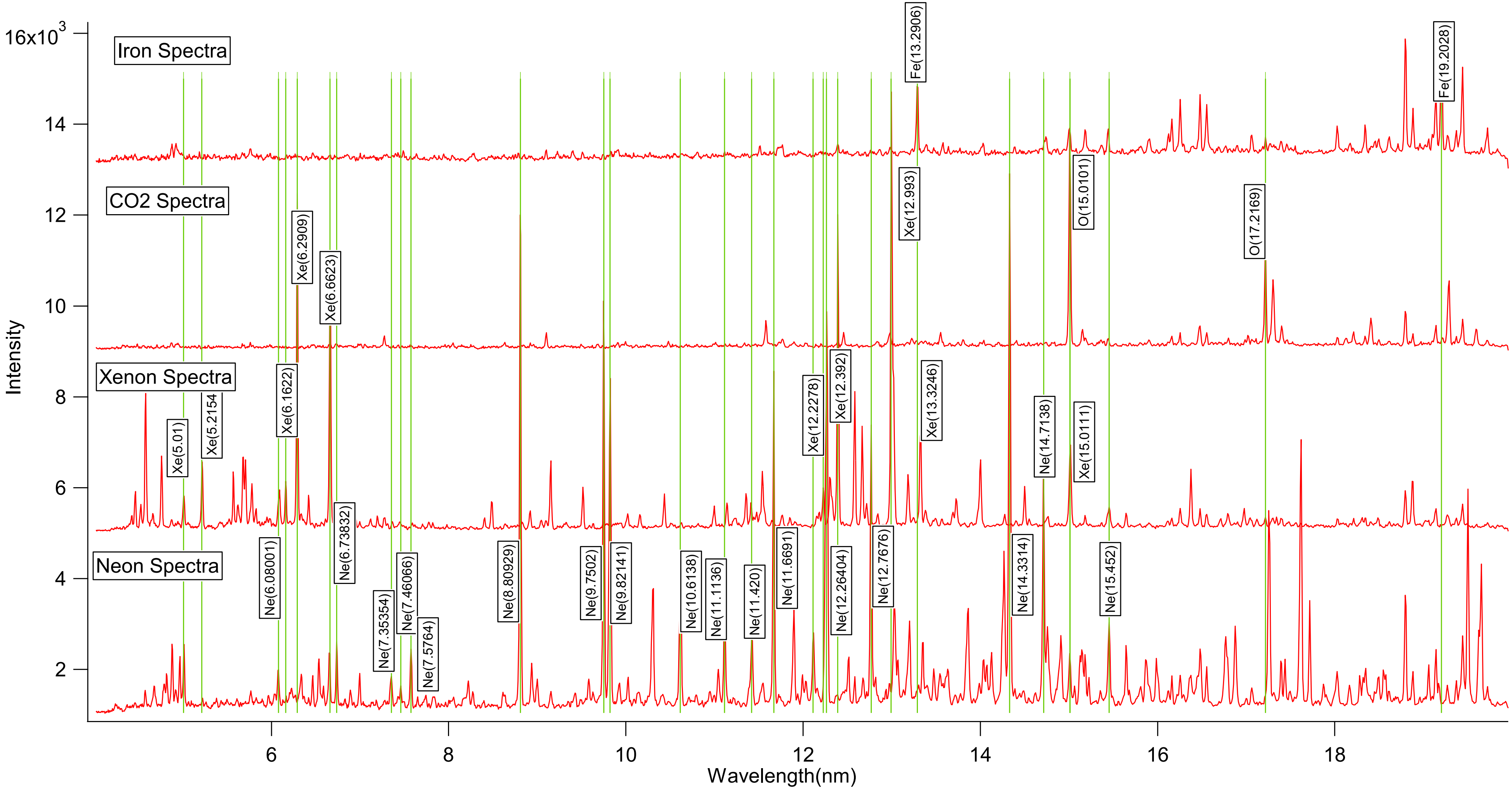
- both have zero magnetic moment
- no hyperfine effect
- discrepancy exists between muonic and optical measurements



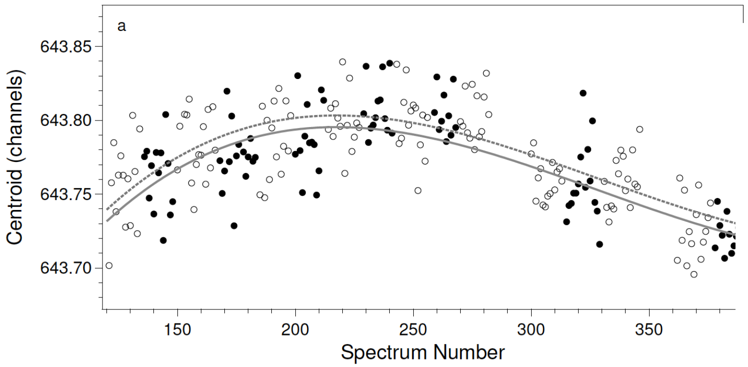
PROOF OF PRINCIPLE: Xe^{136} and Xe^{124} EUV SPECTROSCOPY

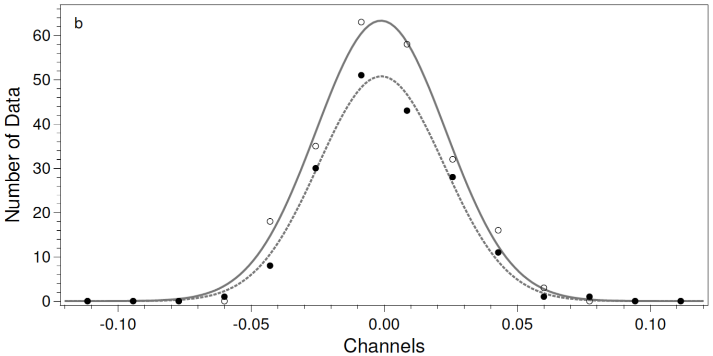


DECADES LONG EXPERIENCE WITH ACCURATE CALIBRATION AT NIST



LINE IDENTIFICATION AND ISOTOPE SHIFT





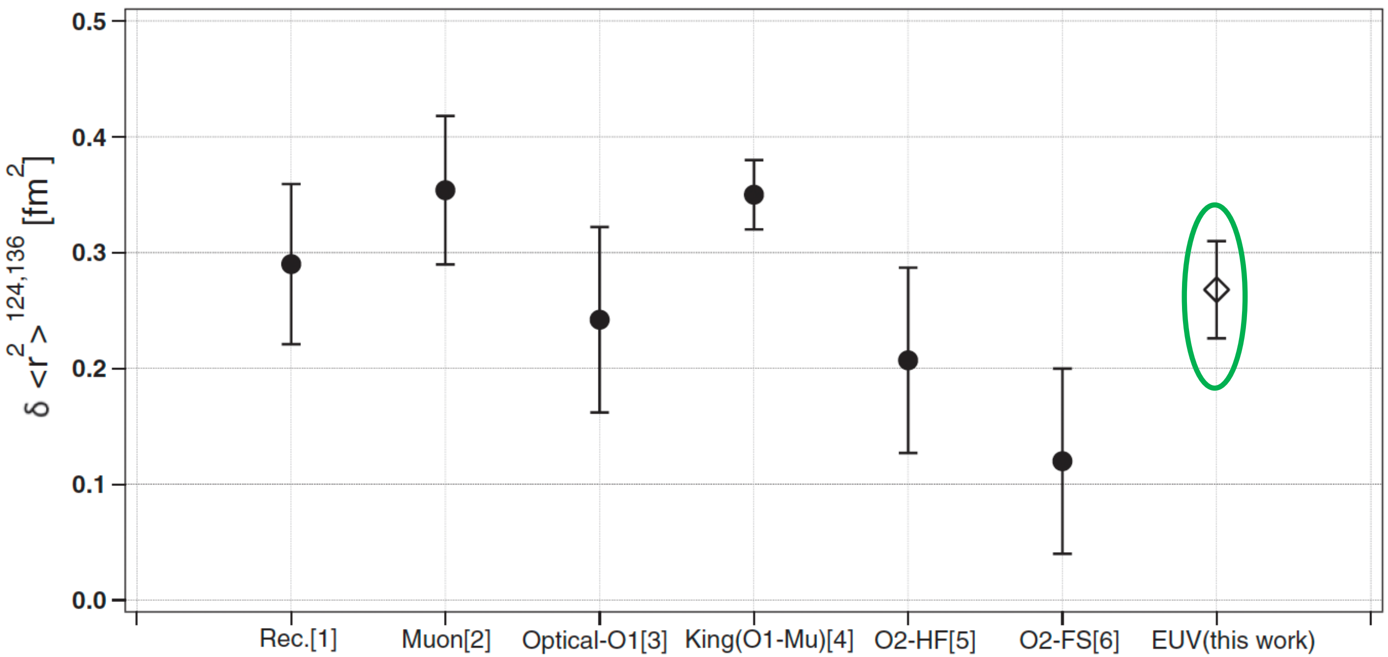
The systematic drift was fitted with an overall function of piece-wise 3rd order polynomials that included a shift between the isotopes (one hundredth of a pixel)

TABLE I. Measured and calculated wavelength values of the isotope shift along with their uncertainties (in units of fm) for the Na-like $D1$ transition $3s\ ^2S_{1/2} - 3p\ ^2P_{1/2}$ for the isotope pair ^{136}Xe–^{124}Xe. The field shift was calculated using the evaluated value of 0.290 fm^2 for $\delta\langle r^2\rangle^{136,124}$ by [20].

	Theory						
	RMBPT		GRASP2K		CIDF [29]	Experiment	
Coefficients	$\delta\lambda$	$\Delta\delta\lambda$	$\delta\lambda$	$\Delta\delta\lambda$	$\delta\lambda$	$\delta\lambda$	$\Delta\delta\lambda$
NMS	−4.8	0.2	−4.8	0.2	−4.8		
SMS	−62.2	3.4	−62.3	3.4	−62.7		
Total MS	−67.0	3.4	−67.1	3.4	−67.5		
FS	143.0	2.8	142.0	2.8	143.0		
Total	76.1	4.4	75.3	4.4	75.8	65.5	20.6 fm

NUCLEAR CHARGE RADIUS DIFFERENCE: $^{136}Xe - ^{124}Xe$

Silwal R et al., Phys. Rev. A **98** (2018) 052502; Silwal R et al., Phys. Rev. A **101** (2020) 062512



Angeli '13

Fricke '95

Borchers '89

Libert '07

Fisher '74

Rec. - Angeli and Marinova

Muon – Muonic atoms

O1 – Optical shift

Exp. Unc.: 0.005 *fm*^{2}

Theor. Unc.: 0.080 *fm*^{2}

King – King plot

O2-HF – OS + Hartree-Fock

O2-FS - OS+ Fermi-Segre

EUV- Present

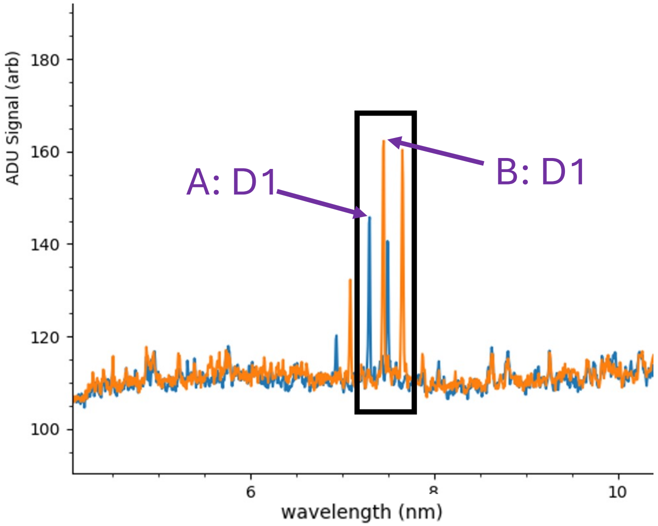
Total Unc.: 0.042 *fm*^{2}

Theor. Unc.: 0.005 *fm*^{2}

$\delta < r^2 >^{136,124} = 0.269(42)$ fm^2

ABSOLUTE NUCLEAR RMS CHARGE RADIUS FROM D LINE SEPARATIONS

Hosier et al., Atoms **11** (2023) 48; Hosier et al., Journal Physics B, **57** (2024) 195001;
Hosier et al., Physical Review Research, (2025) **In print**



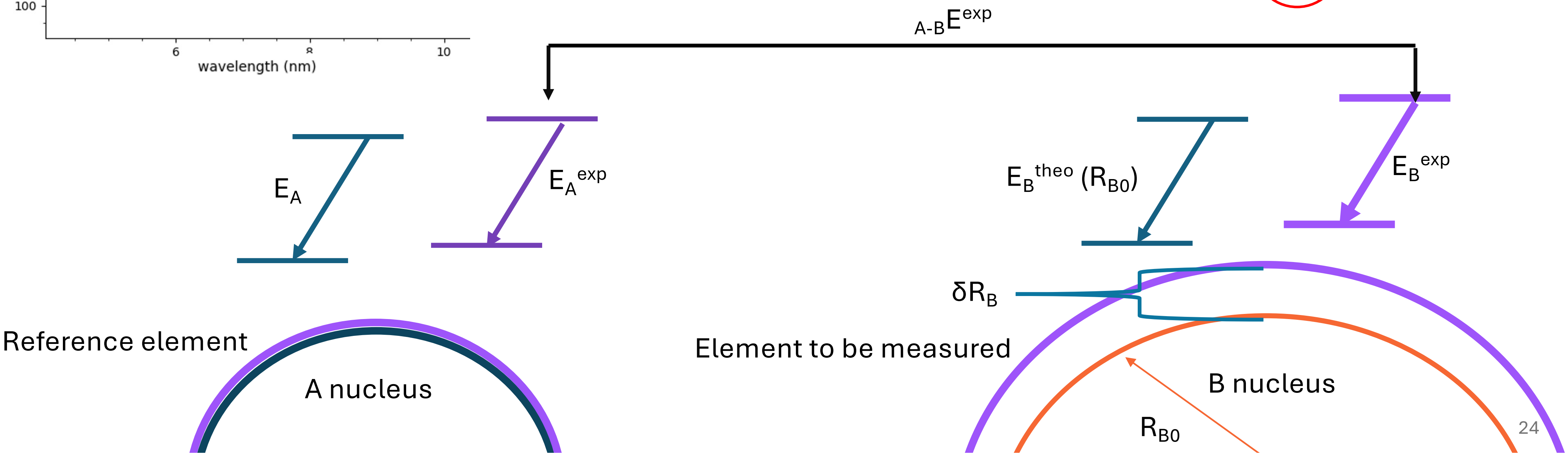
$$\delta E_{AB} \equiv E^{\text{exp}}_{A-B} - E^{\text{th}}_{A-B}$$

theoretical difference has reduced uncertainty (factor of 10!)

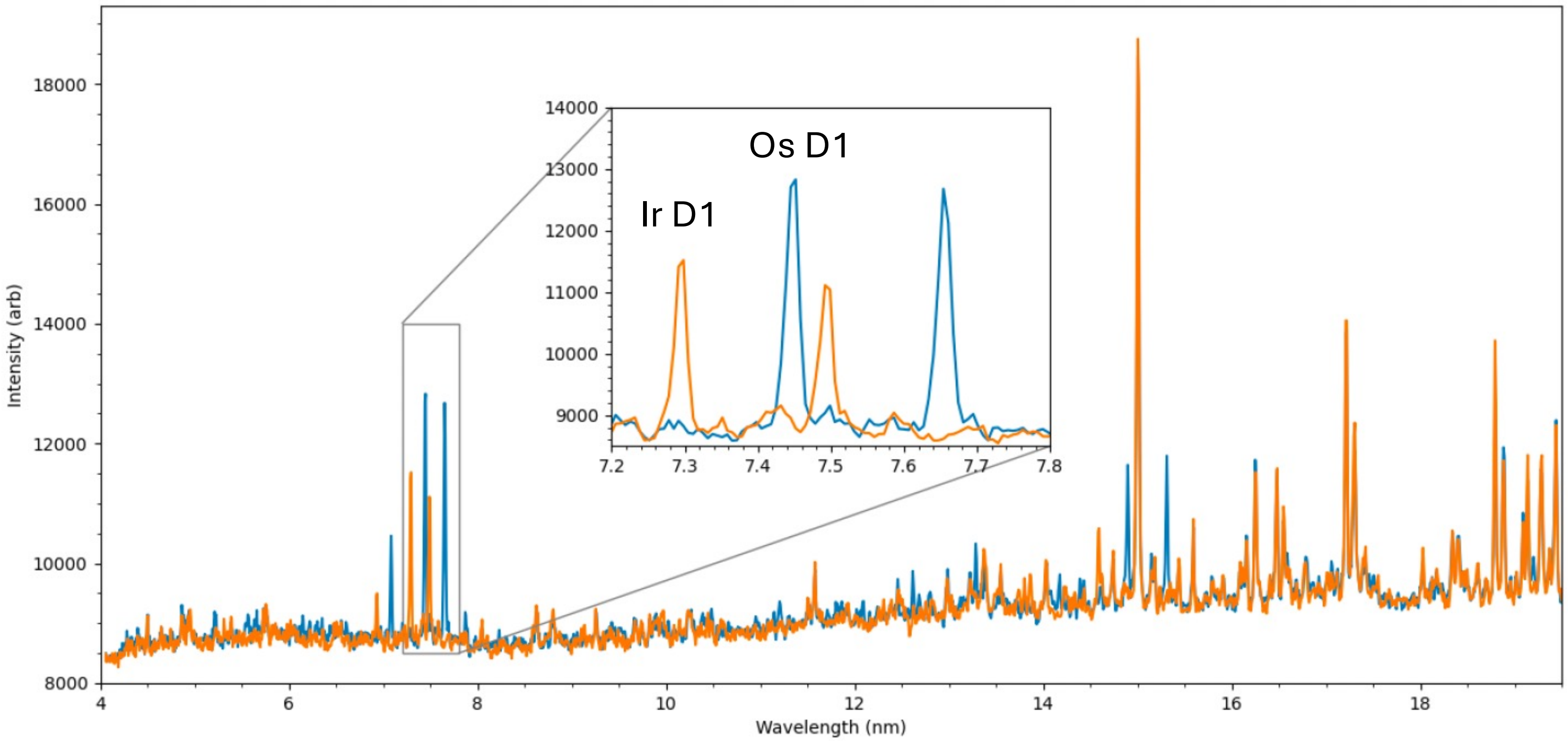
$$\delta E_{AB} = S_B \delta R_B$$

nuclear sensitivity coefficient

$$\delta R_B \equiv R_B - R_{B_0}$$



SPECTRA OF HIGHLY CHARGED Ir AND Os AT 18 keV BEAM ENERGY



EUV spectra of Na-like D1 $3s\ ^2S_{1/2} - 3p\ ^2P_{1/2}$ and Mg-like $3s^2\ ^1S_0 - 3s3p\ ^3P_1$ transitions for both Os and Ir, in orange and blue respectively.

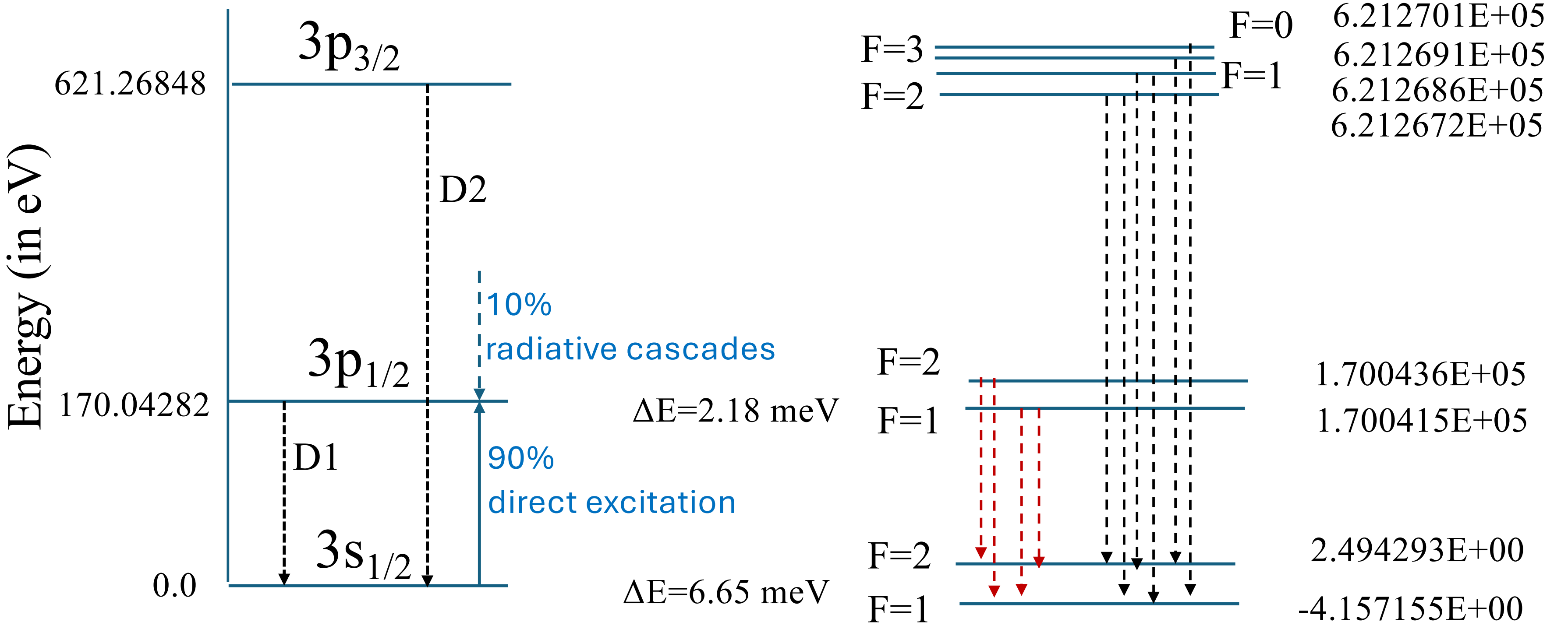
NA-LIKE LEVEL POPULATION MECHANISMS

- ✓ The Ir and Os in the measurements have the natural abundance of their isotopes.
- ✓ Isotope with odd number of nucleons exhibit HF structure.

Os-184	0.02(1) %
Os-186	1.59(3) %
Os-187	1.96(2) %
Os-188	13.24(8) %
Os-189	16.15(5) %
Os-190	26.26(2) %
Os-192	40.78(19) %

Ir-191	37.3(2) %
Ir-193	62.7(2) %

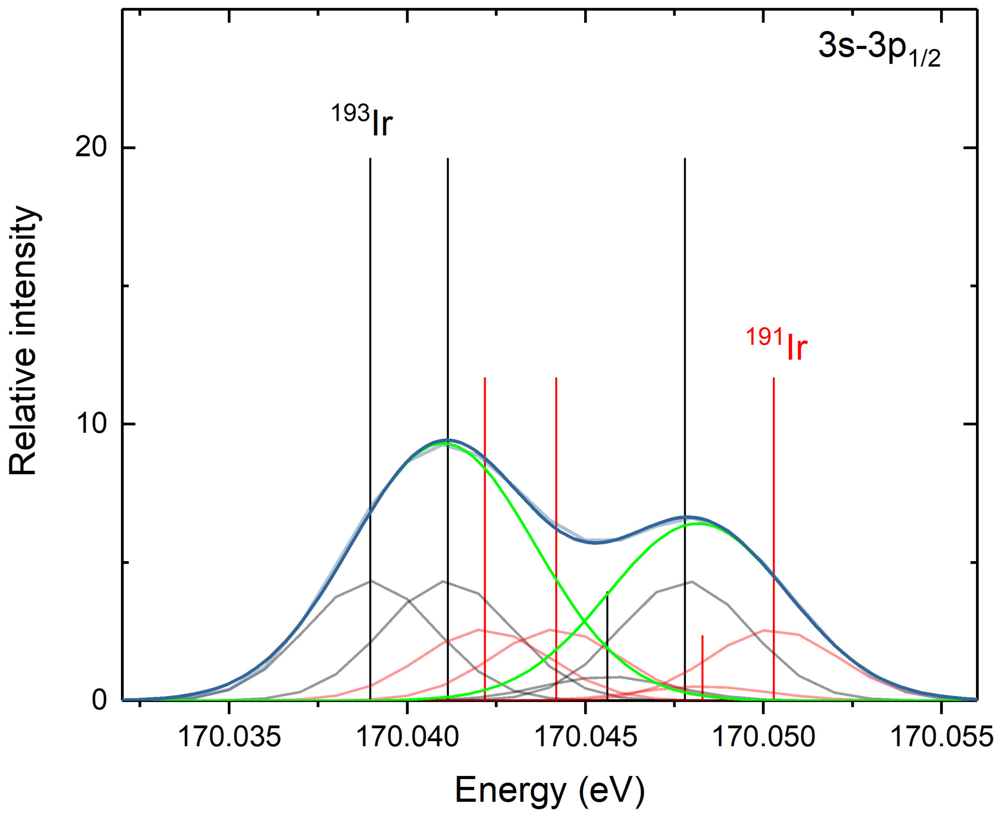
$$\Delta E = \frac{A}{2}K + \frac{B}{4}\frac{1.5K(K+1) - 2I(I+1)J(J+1)}{I(2I-1)J(2J-1)}$$



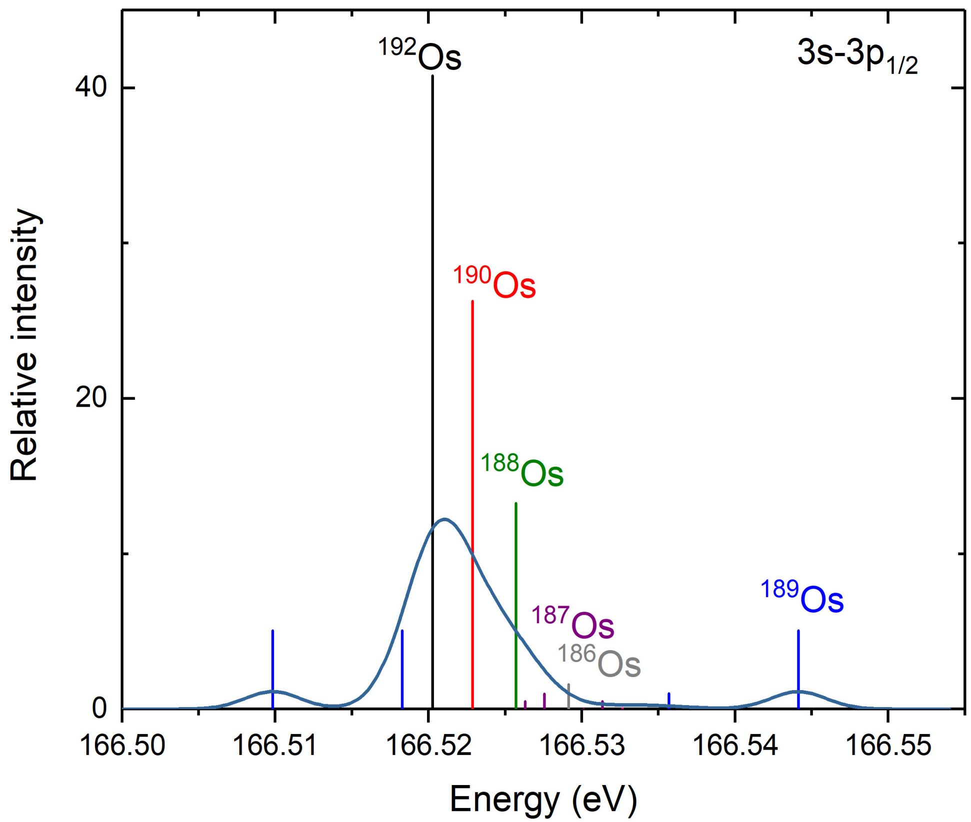
Fine-structure splitting

Hyperfine-structure splitting

HYPERFINE STRUCTURE OF THE Na-like Ir AND Os D1 TRANSITIONS



HF splitting ~ 9 meV



HF splitting ~ 35 meV

Instrument resolution ~ 440 meV

RESULTS: NUCLEAR CHARGE RADIUS OF ^{191}Ir

Hosier et al., Physical Review Research, (2025) **In print**

Units: eV						
	Os D1 [eV]		**Ir D1 [eV]**		**Ir–Os D1 [eV]**	
R(rms):	5.4064	fm	5.4000	fm	(interpolation)	
DF	167.470	(0)	171.025	(0)	3.5550	(0)
B(1)	4.746	(0)	4.977	(0)	0.2310	(0)
B(RPA)	-0.118	(0)	-0.122	(0)	-0.0045	(0)
BB(RPA)	-0.011	(0)	-0.012	(0)	-0.0007	(0)
Ret(1)	0.016	(0)	0.020	(0)	0.0045	(0)
Ret(RPA)	0.004	(0)	0.004	(0)	0.0003	(0)
Ret(other)	0.000	(11)	0.0000	(11)	0.0000	(3)
CC(2)	-0.300	(0)	-0.303	(0)	-0.0032	(0)
BC(2)	-0.053	(0)	-0.055	(0)	-0.0019	(0)
BB(2)	0.011	(0)	0.011	(0)	-0.0001	(0)
GGG(3)	0.005	(0)	0.005	(0)	0.0001	(0)
Nuc. Rec.	-0.007	(2)	-0.007	(2)	0.0000	(0)
RMBPT(tot)	171.762	(11)	175.543	(11)	3.7805	(3)
SE(val)	-6.387	(0)	-6.725	(0)	-0.3375	(0)
Uehl(val)	1.162	(0)	1.245	(0)	0.0825	(0)
WK(val)	-0.045	(4)	-0.049	(4)	-0.0041	(4)
SE(val-x)	0.090	(1)	0.094	(1)	0.0041	(1)
VP(val-x)	-0.016	(0)	-0.017	(0)	-0.0010	(0)
SE(core)	-0.153	(2)	-0.161	(2)	-0.0072	(1)
VP(core)	0.027	(0)	0.029	(0)	0.0017	(0)
Other(vert)	0.000	(10)	0.000	(11)	0.0000	(5)
2-loop	0.019	(7)	0.020	(7)	0.0013	(5)
QED(tot)	-5.304	(13)	-5.564	(14)	-0.2602	(8)
TOTAL	**166.458**	**(17)**	**169.979**	**(18)**	**3.5204**	**(8)**
		(38)		(40)		(18)
		(25)		(27)		(12)
TOTAL [no BB(2)]	***166.447***	***(17)***	***169.968***	***(18)***	***3.5205***	***(8)***

$$\delta R_{\rm Ir} = \frac{1}{S_{\rm Ir}}[\delta E^{\rm exp}_{\rm Ir\text{-}Os} - (E^{\rm th}_{\rm Ir}(R_{\rm Ir}) - E^{\rm th}_{\rm Os}(R_{\rm Os}))]$$

5.4307(77) fm

Uncertainty

key theoretical

experimental

$$\Delta(\delta R_{Ir}) = \Big[\frac{(S_{Os}\Delta R_{Os})^2}{S_{Ir}^2} + \frac{(\Delta[E_{Os}(R_{Os,0}) - E_{Ir}(R_{Ir,0})])^2}{S_{Ir}^2} + \frac{(\Delta E^M_{Os-Ir})^2}{S_{Ir}^2} + (\delta R_{Ir})^2\left(\frac{\Delta S_{Ir}}{S_{Ir}}\right)^2\Big]^{1/2}$$

Nuclear model dependence

$$\rho(r,\theta,\phi) = \frac{\rho_0}{1 + exp[(r - c_{\rm def})/a]} \qquad c_{\rm def}(\theta,\phi) = c[1 + \beta_2 Y_{20}(\theta,\phi)]$$

$$\delta E(R,\beta_2,t) = \delta R\frac{\partial E}{\partial R} + \delta\beta_2\frac{\partial E}{\partial\beta_2} + \delta t\frac{\partial E}{\partial t}$$
$$\equiv S\delta R + S_{\beta_2}\delta\beta_2 + S_t\delta t$$

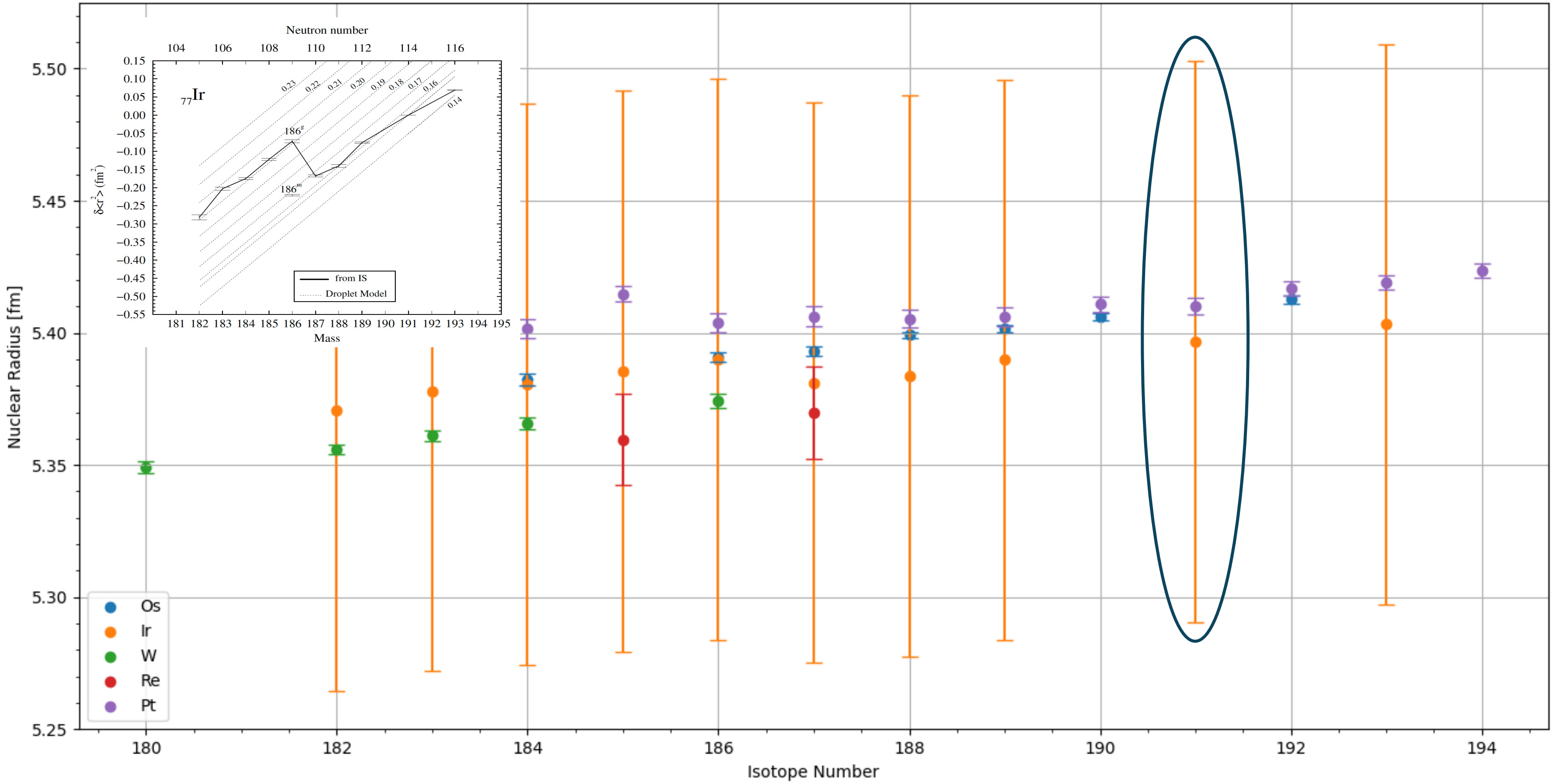
	units	Na-like D_1
S	eV fm^{-1}	-0.4557
S_{β_2}	eV	0.0032
S_t	eV fm^{-1}	0.0025
S_2	eV fm^{-2}	-0.026
$\Delta\beta_2$		0.17
Δt	fm	0.1
$\delta R_{\rm Ir}$	fm	0.04
$\Delta S/S$		0.5%

Included in ΔS

$$(\Delta S/S)\delta R_{\rm Ir} \approx 0.0016{\rm fm}$$

Nuclear charge radius measurement of Ir using Os as a reference element

Hosier et al., Physical Review Research, (2025) **In print**



Angeli and K. P. Marinova, At. Data Nucl. Data Tables **99**, 69 (2013)

Nuclear charge radius measurement of Ir using Os as a reference element

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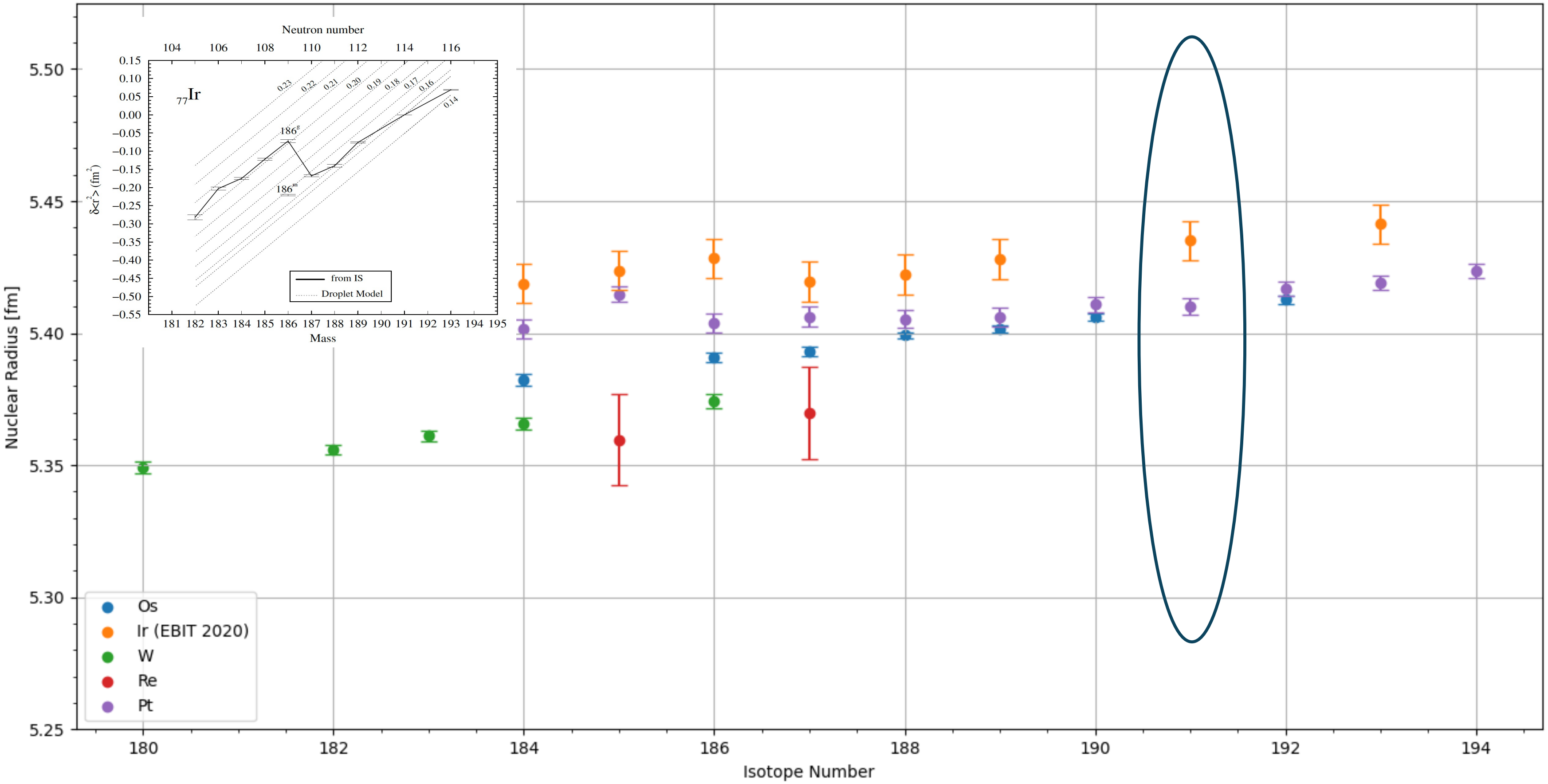
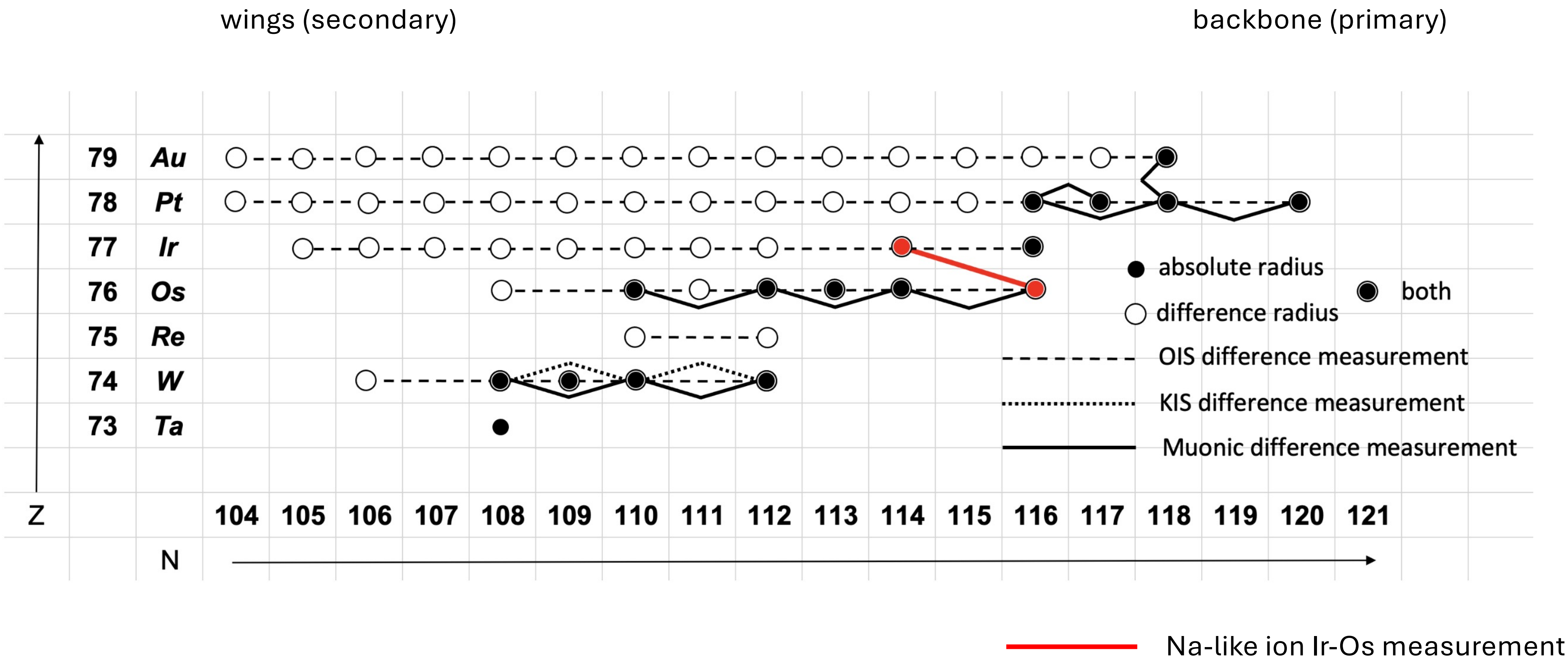
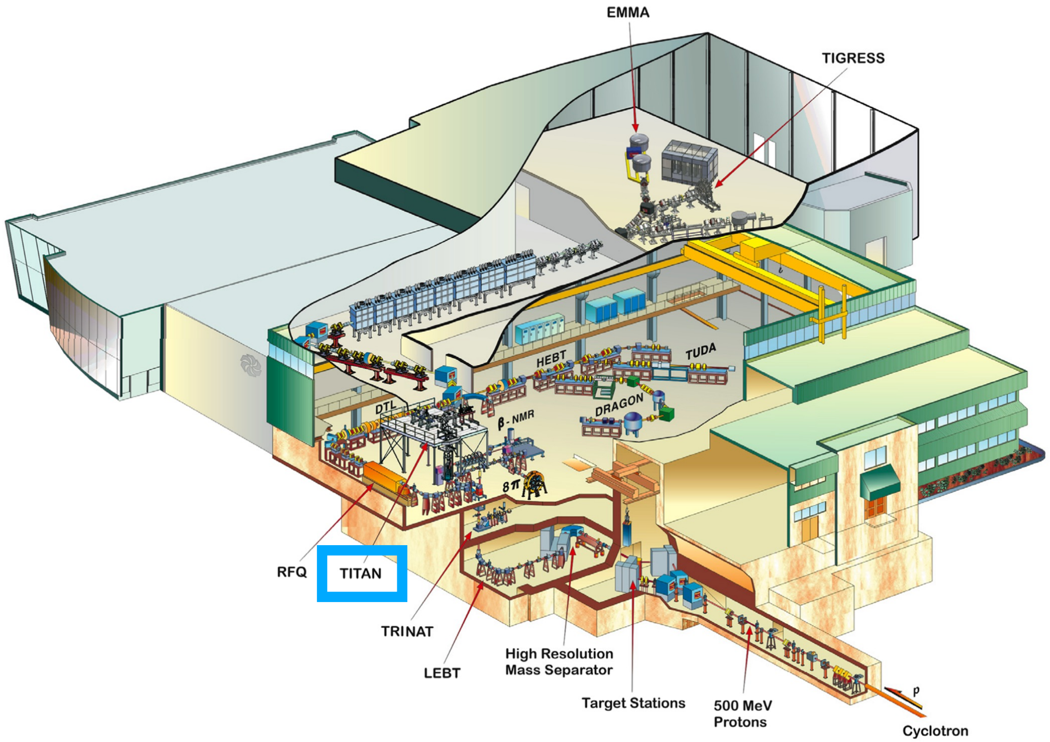


Our measurements combined with optical isotope shift data

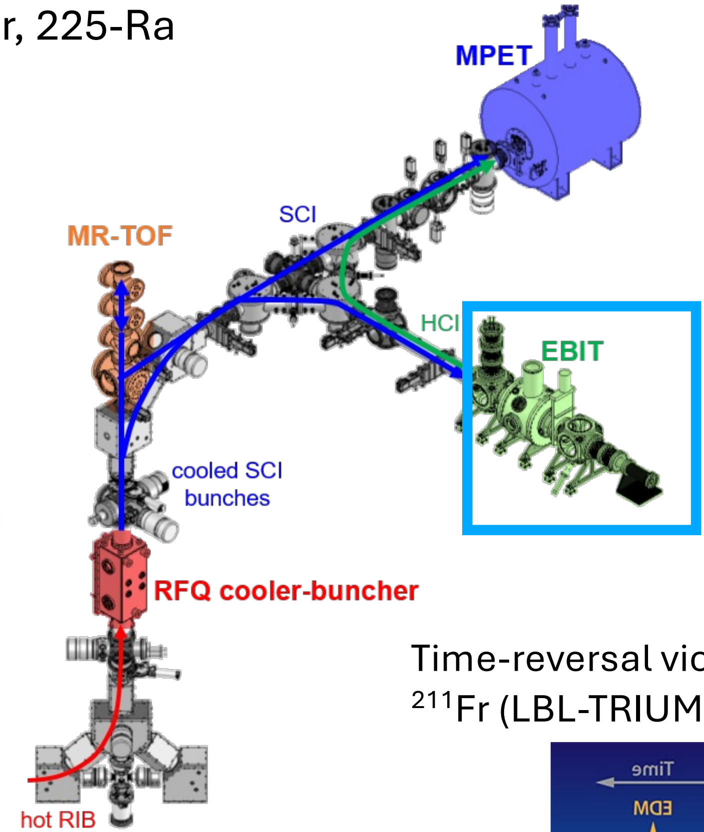
NUCLEAR CHARGE RADIUS CROSS-ELEMENT CONSTRAINT



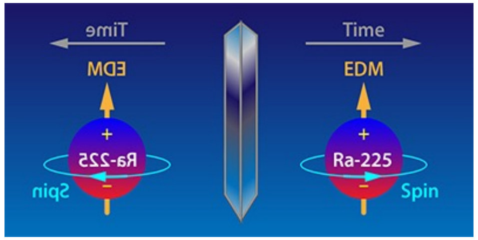
Next step: TRIUMF's Ion Trap for Atomic and Nuclear science (TITAN)



Fr, 225-Ra

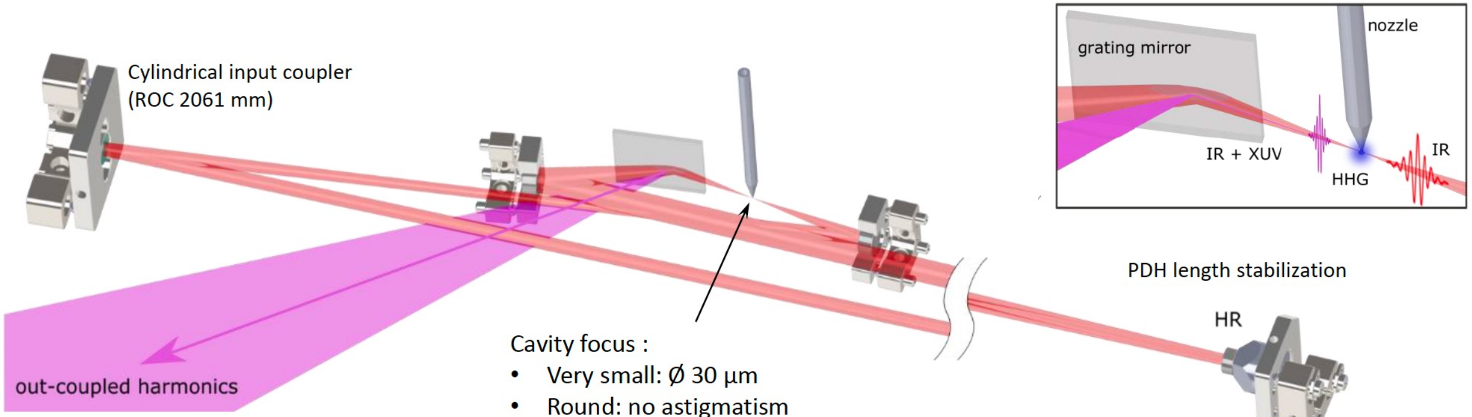


Time-reversal violation measurements:
^{211}Fr (LBL-TRIUMF), ^{225}Ra (Argonne NL)

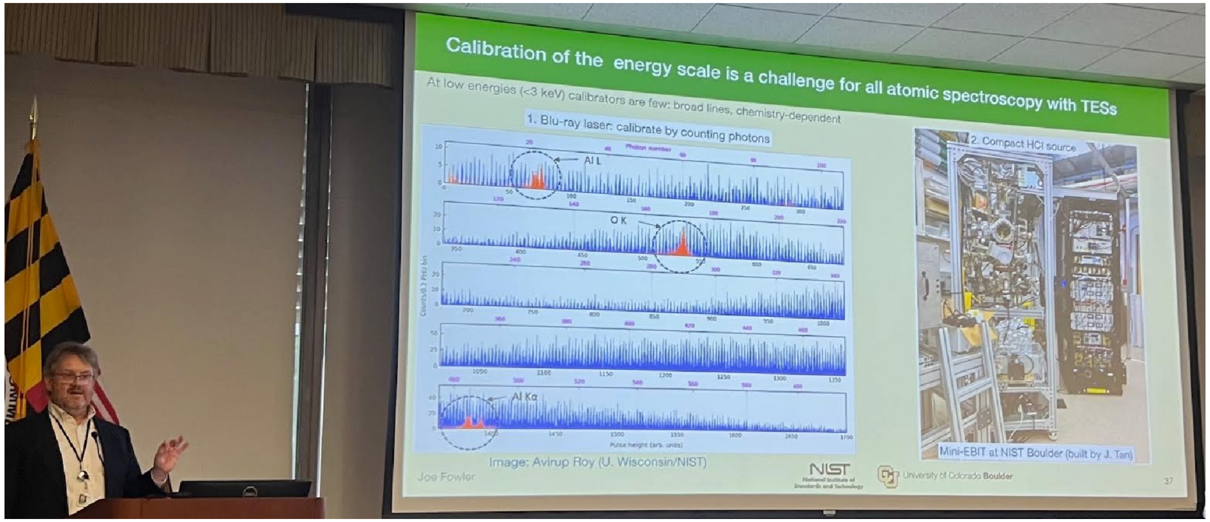


Peter Mueller/Argonne National Laboratory

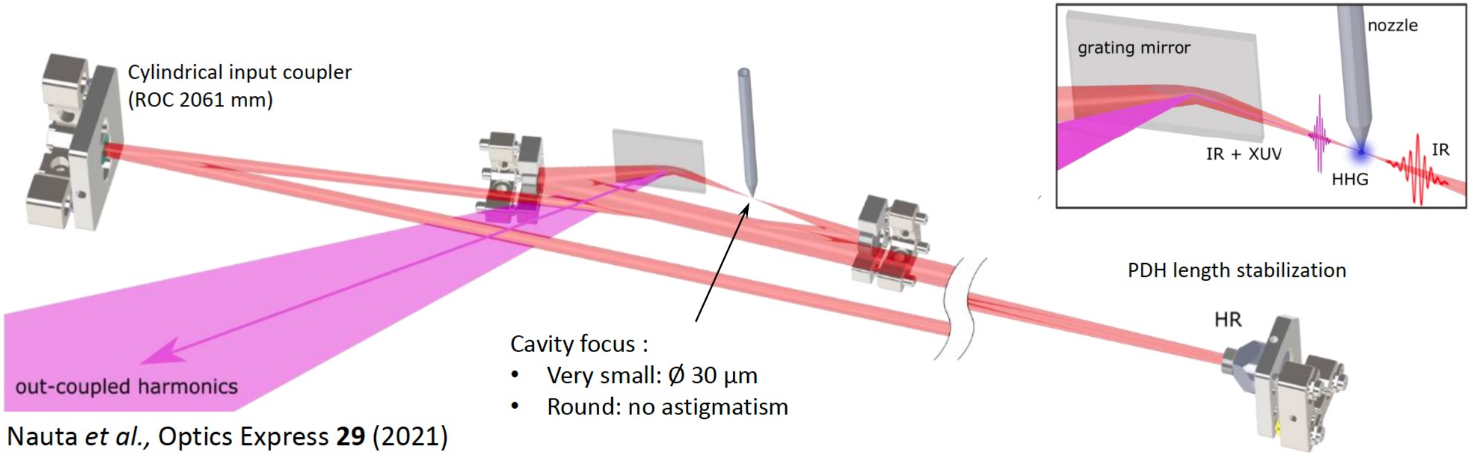
Future Plans: Improving Precision with XUV Frequency Combs, microcaloriemeter



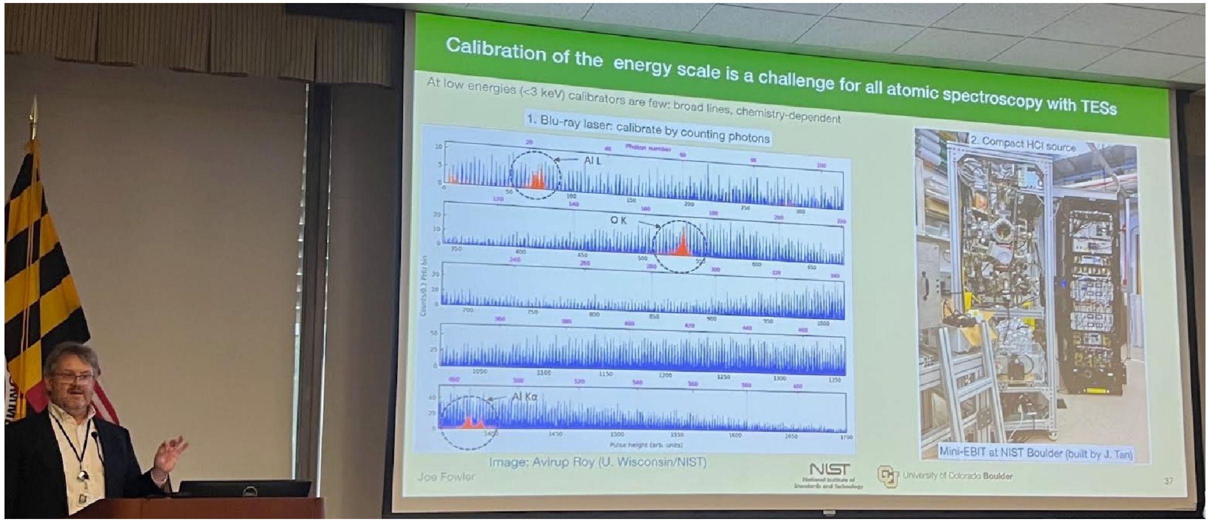
J. Nauta *et al.,* Optics Express **29** (2021)



Future Plans: Improving Precision with XUV Frequency Combs, microcaloriemeter



J. Nauta *et al.*, Optics Express **29** (2021)



Future is bright! Thank you!