New atomic data calculations in the Yb I isoelectronic sequence (Ta IV - Pt IX) relevant to nuclear fusion diagnostics

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Table of contents

1. Introduction
   - Ionic impurities in the fusion plasma
   - Plasma diagnostics

2. HFR+CPOL preliminary results
   - Pseudo-relativistic Hartree-Fock method with the core polarization corrections (HFR+CPOL)
   - Model + least squares adjustment
   - The configuration interactions and the core-valence correlations
   - Radiative decay rates

3. MCDHF preliminary results
   - Multiconfigurational Dirac-Hartree-Fock (MCDHF) method
   - Model and convergence
   - Radiative decay rates (comparisons)

Introduction

Ionic impurities in the fusion plasma

- Divertor will be made of tungsten: high fluxes of heat and particles (neutrons)

- Transmutation products after few years irradiation time: rhenium, osmium, tantalum, hafnium, iridium and platinium → impurities

- Brittleness of pure tungsten → alloying elements (tantalum, rhenium, titanium or vanadium)

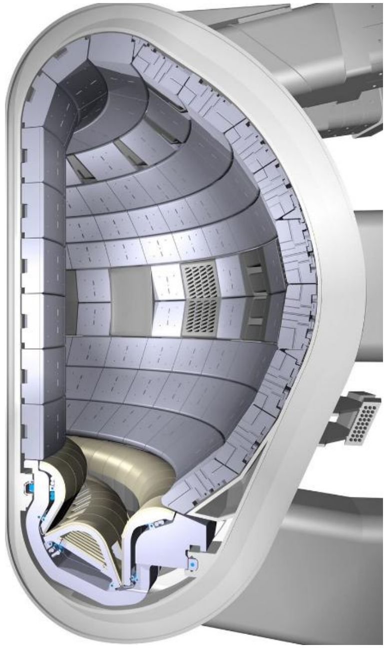
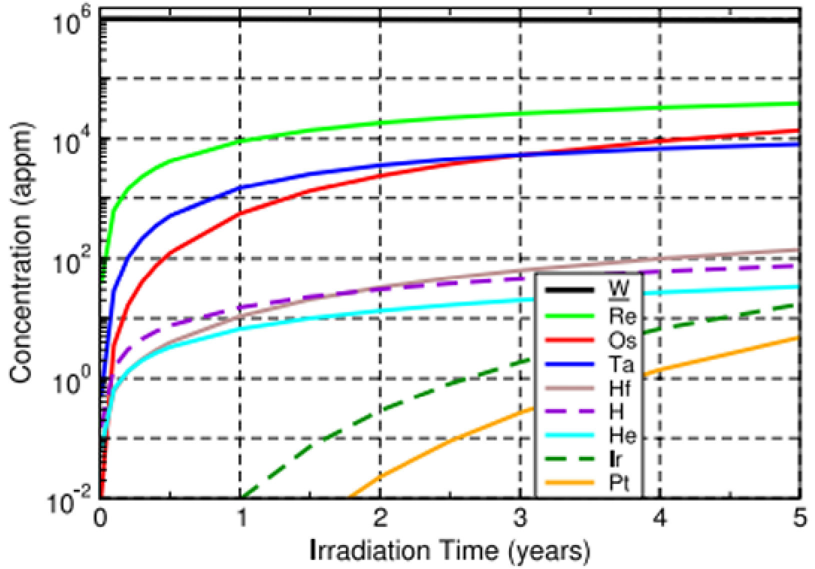


Illustration of the inside of the Tokamak. (https://encyclopedia.pub/entry/37629)



Evolution of the concentration (Atomic Part Per Millions) of all the elements produced by the transmutation of W for a five-years irradiation [Gilbert, M. R and Sublet, J.Ch , 2011]

Plasma diagnostics

- Ionic impurities will contribute to the radiation losses/allow to diagnose the fusion plasma

- From observed intensity ratio and radiative parameters [HJ. Kunze, 2009] :

➢ $$\frac{\varepsilon_Z(p\to q)}{\varepsilon_Z(p'\to q')} = \frac{\lambda_{p'q'}}{\lambda_{pq}} \frac{A_Z(p\to q)}{A_Z(p'\to q')} \frac{A_Z(p'\to)}{A_Z(p\to)} \frac{X_Z(g\to p)(T_e)}{X_Z(g\to p')(T_e)} \Rightarrow T_e$$

➢ $$\frac{\varepsilon_Z(p\to q)}{\varepsilon_Z(p'\to q')} = \frac{\lambda_{p'q'}}{\lambda_{pq}} \frac{A_Z(p\to q)}{A_Z(p'\to q')} \frac{A_Z(p'\to)}{A_Z(p\to)} \frac{X_Z(g\to p)(T_e)}{X_Z(g\to p')(T_e)} \left[1 + n_e \frac{X_Z(p'\to)}{A_Z(p'\to)}\right] \Rightarrow n_e$$

HFR+CPOL preliminary results

HFR method

General procedure [Cowan, R. D. ,1981]

- Solve $H\Psi = E\Psi$ where $H = \sum_{i=1}^{N}\left(-\frac{1}{2}\Delta_i + V(r_i)\right)$ (central field approximation)
- $H_i\varphi_i = E_i\varphi_i \rightarrow \varphi_i(r_i,\theta_i,\phi_i,s_i) = \frac{1}{r_i}P_{n_il_i}(r_i)Y_{l_i}^{m_i}(\theta_i,\phi_i)\sigma_{m_{s_i}}(s_i)$
- $P_{n_il_i}(r_i)$? → solve Hartree-Fock equations (Self-Consistent Field method)
- HF equations obtained by variationnal principle on the average energy of each eletronic configuration
- Atomic State Functions (ASFs) : $\Psi(\alpha, P, J, M_J) = \sum_{r=1}^{n_c} c_r\Phi(\alpha_r, P, L_r, S_r, J, M_J)$

Configuration State Functions (CSFs) are built thanks to Slater determinants

HFR+CPOL method

Core polarisation correction

- Valence electron correlations represented by configuration interactions (CI) and other correlations by core-polarisation potential

- Quinet, P. et al [1999, 2002]: pseudo potentiel have one-body and two-body part :

  - $V_{P1} = -\frac{1}{2}\alpha_D \sum_{i=1}^{N} \frac{r_i^2}{\left(r_i^2 + r_c^2\right)^3}$ and $V_{P2} = -\alpha_D \sum_{i>j} \frac{\vec{r_i} \cdot \vec{r_j}}{\left[\left(r_i^2 + r_c^2\right)\left(r_j^2 + r_c^2\right)\right]^{3/2}}$

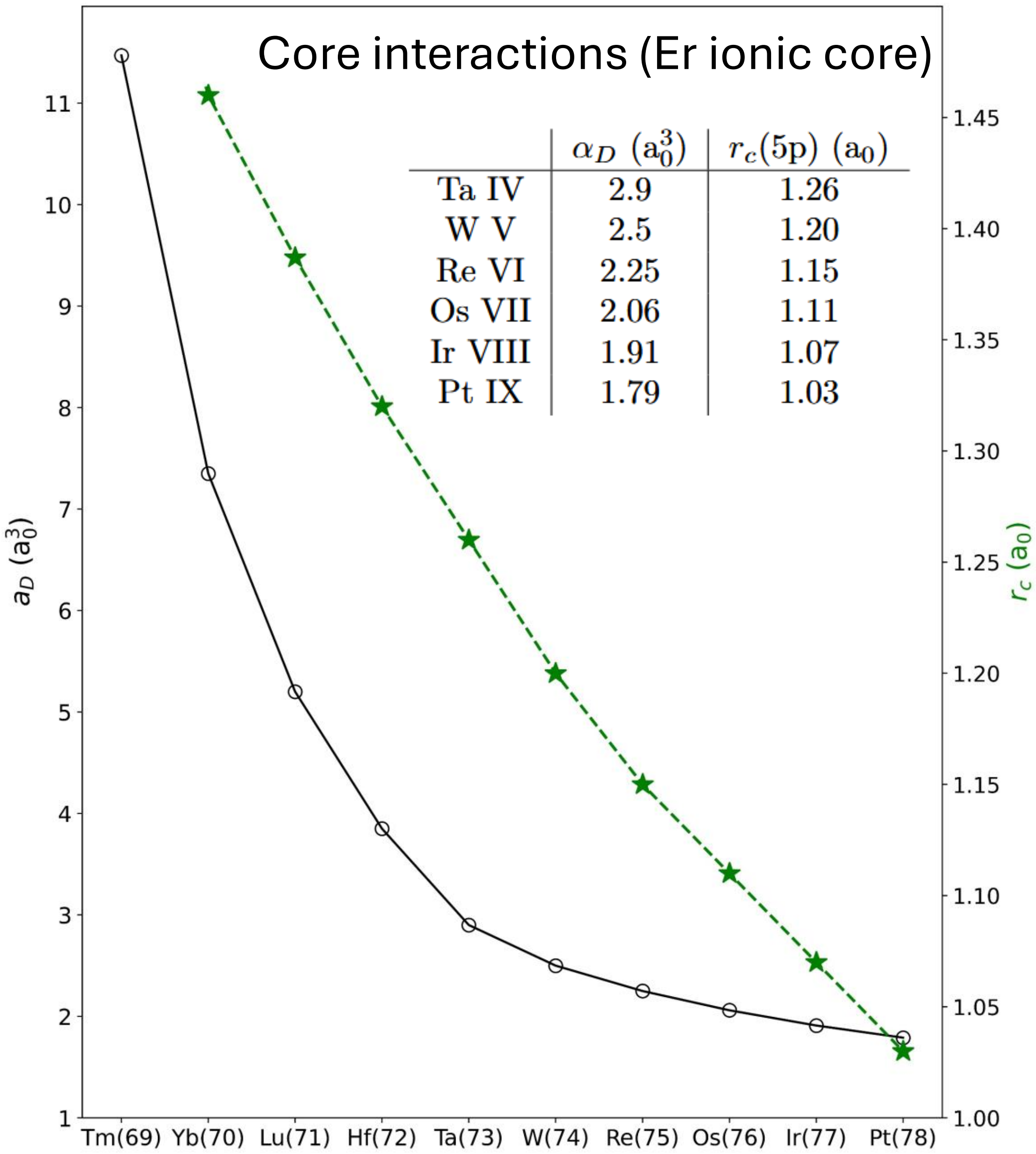
  - α_D: dipole polarisability; r_c: ionic core radius

HFR+CPOL Model

Valence-Valence interactions

Even parity		Odd parity	
$5d^2$	6p6f	5d6p	6p6d
5d6s	6p7f	6s6p	6p7d
5d7s	$6d^2$	5d7p	6d7p
$6s^2$	6d7s	5d5f	6d5f
5d6d	6d7d	5d6f	6d6f
5d7d	$7s^2$	5d7f	6d7f
6s7s	$7p^2$	6s7p	7s5f
6s6d	7s7d	6s5f	7s6f
6s7d	7p5f	6s6f	7s7f
6p7p	7p6f	6s7f	7s7p
$6p^2$	7p7f	6p7s	7s7d
6p5f			

[Yoca et al, 2012]



Least squares method

- Minimise difference between computed energy levels and observed ones with spin-orbit and Slater parameters

- Accuracy of the fit: $\sigma = \left[\frac{\sum_k (E^k - T^k)^2}{N_k - N_p}\right]^{1/2}$, where E^k: energy eigenvalues; T^k: observed energies; N_k: number of fitted levels; N_p: number of fitted parameters

- HFR+CPOL model :

	Even parity		Odd parity	
	nbr of fitted levels	σ (cm^{-1})	nbr of fitted levels	σ (cm^{-1})
Ta IV	14	22	44	142
W V	13	20	44	132
Re VI	13	30	29	347
Os VII	13	31	30	779
Ir VIII	13	35	30	1216
Pt IX	13	38	30	3363

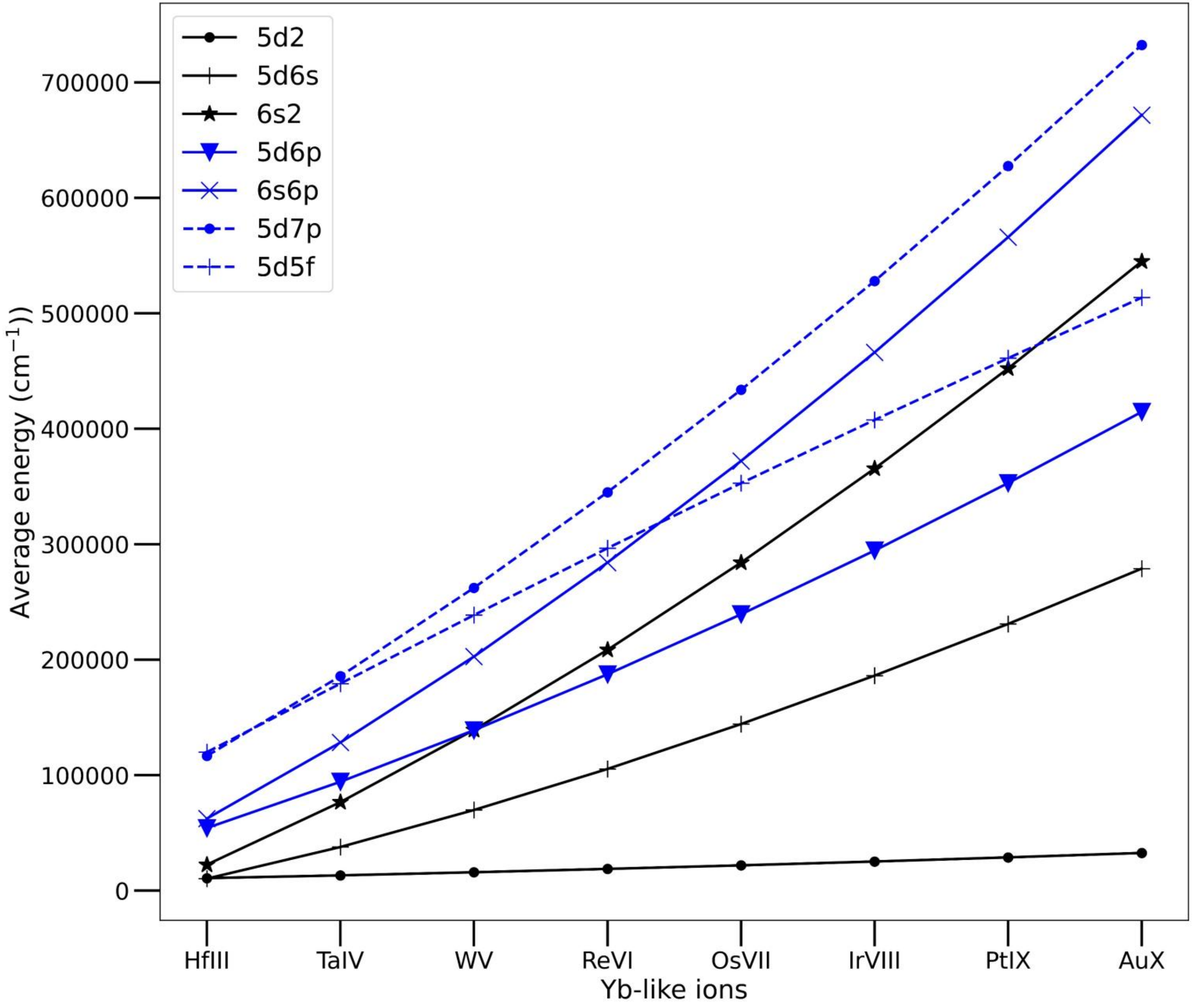
σ decreases due to less mixing

5d7p odd configuration not observed

[Churilov, S.S. et al, 1996 ; Meijer, F.G and Metsch, 1978 ; Kildiyarova, R.R et al, 1996 ; Kramida , A. et al, 2024 ; Sugar, J. et al, 1994 ; Yoca, S.E et al, 2012 ; Van het Hof, G.J. et al, 1995 ; Kildiyarova, R.R et al, 1995 ; Kildiyarova, R.R et al, 1997 ; Azarov V.I. and Churilov S.S , 1999]

Configuration Interactions

The atomic level structure more spread out with increasing ionic charge state → on average less mixing in the level composition

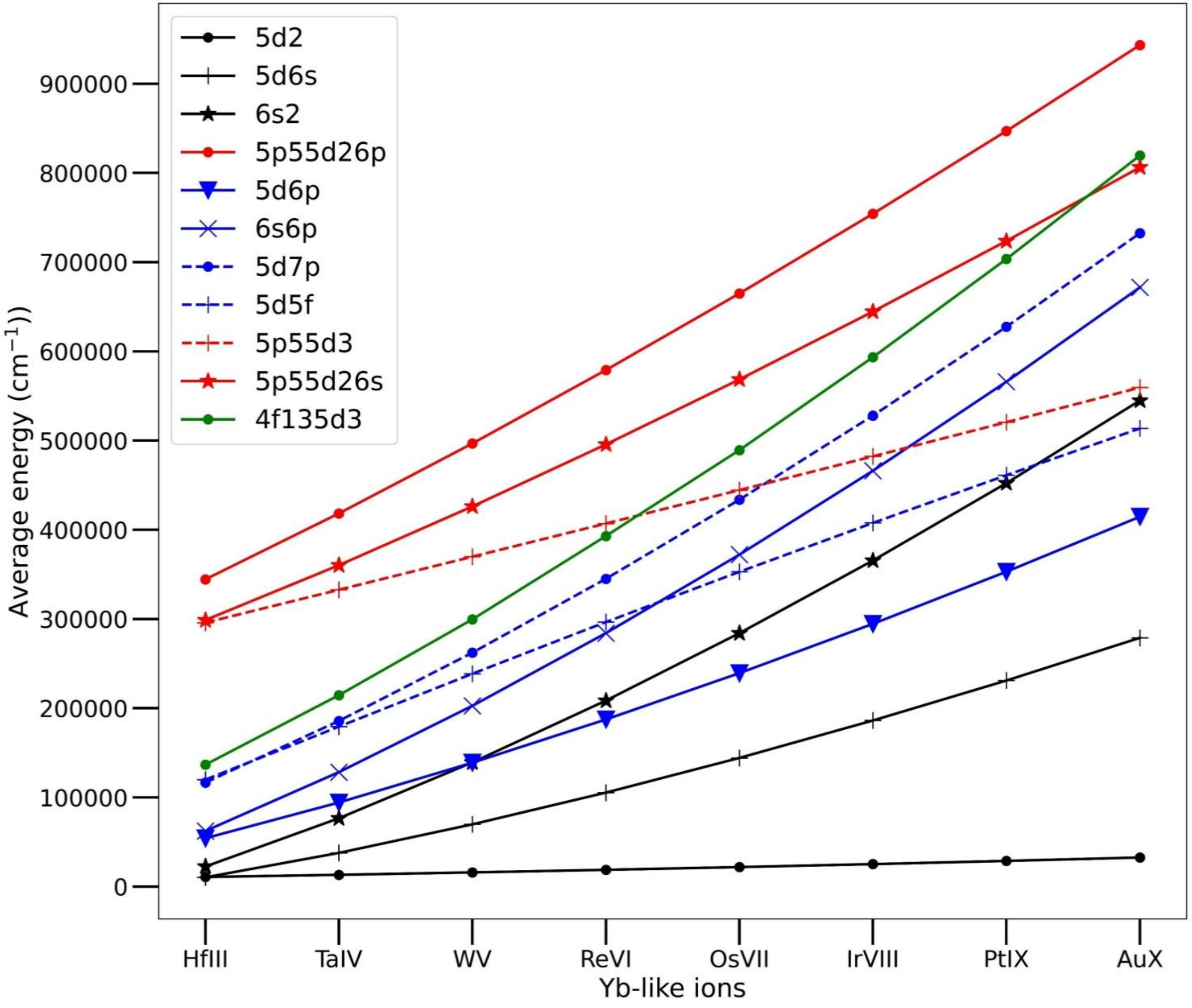


Core-valence correlations

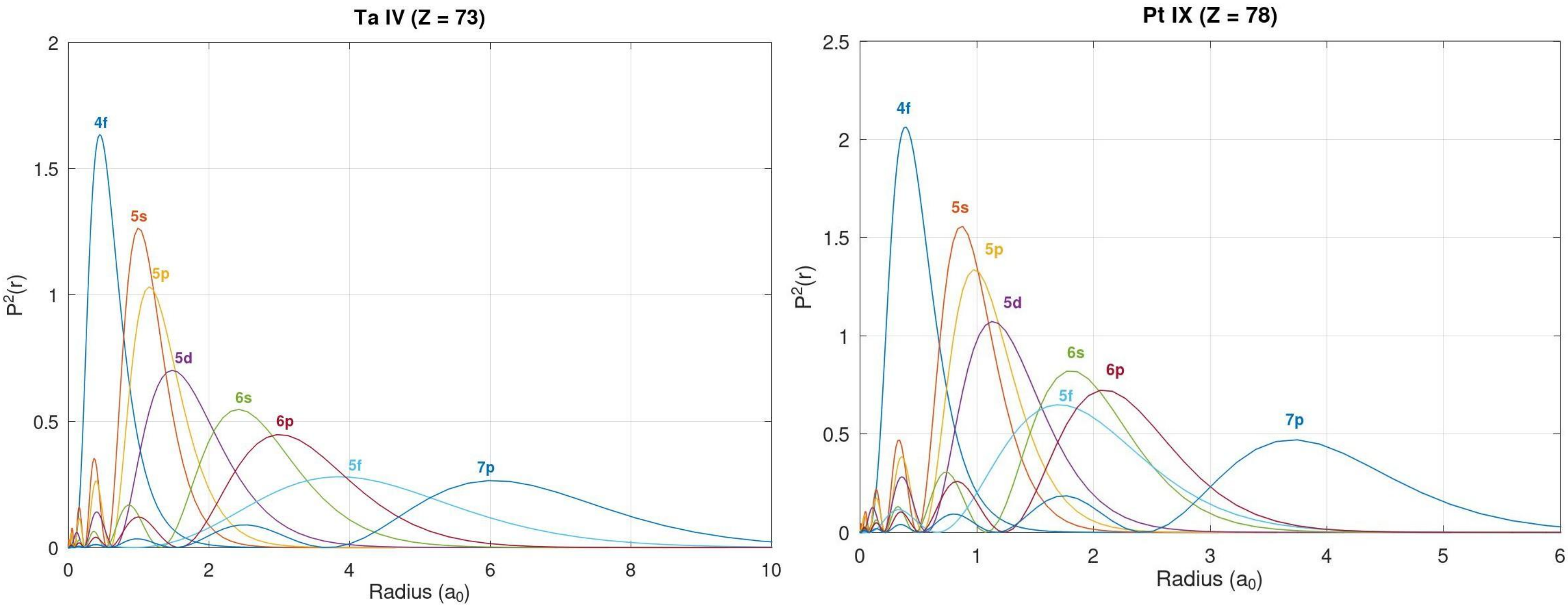
$5p^5 5d^3$ closer to (and overlap) 5d7p, 6s6p and 5d5f

→ core-valence correlations with a pseudo potential no longer valid

→ need to introduce explicitly the configurations with an open 5p,4f core orbitals in our model (through CI)



Large radial part squared computed with the MCDHF method for the 4f, 5s, 5p core orbitals and 5d, 6s, 6p, 5f, 7p valence orbitals after the optimization of the radial part → 5f orbitals moves deeper into the core than the other valence orbitals



HFR(CV) Model

VV + CV interactions

Even parity	
$5d^2$	$5p^5 5d^2 6p$
$5d6s$	$5p^5 6s^2 6p$
$6s^2$	$5p^5 5d6s6p$
$5d7s$	$5p^5 5d^2 5f$
$5d6d$	$5p^5 5d^2 7p$
$5d7d$	$5s5p^6 5d^3$
$6s7s$	$5s5p^6 5d^2 6s$
$6s6d$	$5s5p^6 5d6s^2$
$6s7d$	$4f^{13} 5p^6 5d^2 6p$
$6p^2$	$4f^{13} 5p^6 5d6s6p$
$6p7p$	
$6p5f$	

Odd parity	
$5d6p$	$5p^5 5d^3$
$6s6p$	$5p^5 5d^2 6s$
$5d7p$	$5p^5 5d6s^2$
$5d5f$	$5p^5 5d6p^2$
$5d6f$	$5p^5 6s6p^2$
$5d7f$	$5s5p^6 5d^2 6p$
$6s7p$	$5s5p^6 5d6s6p$
$6s5f$	$4f^{13} 5p^6 5d^3$
$6s6f$	$4f^{13} 5p^6 5d^2 6s$
$6p6d$	
$6p7s$	

	Even parity	
	$\sigma_{HFR+CPOL}$ (cm^{-1})	$\sigma_{HFR(CV)}$ (cm^{-1})
Ta IV	22	24
W V	20	31
Re VI	30	37
Os VII	31	40
Ir VIII	35	47
Pt IX	38	53
	Odd parity	
Ta IV	142	156
W V	132	141
Re VI	347	253
Os VII	779	139
Ir VIII	1216	571
Pt IX	3363	417

HFR(CV) Model

VV + CV interactions

→ σ of the even parity increases slightly due to more level mixing
Ex: the level $5d^2\ {}^3P_0$ observed at 11 166.5 cm^{-1} in Ta IV (Z=73) and 18 617.0 cm^{-1} in Pt IX (Z=78) is composed of the three most dominant spectroscopic terms 93.7% $5d^2\ {}^3P$ + 5.5% $5d^2\ {}^1S$ + 0.1% $6s^2\ {}^1S$ in Ta IV and 87.2% $5d^2\ {}^3P$ + 12.0% $5d^2\ {}^1S$ + 0.2% $5p^5 5d^2 6p\ {}^3P$ in Pt IX

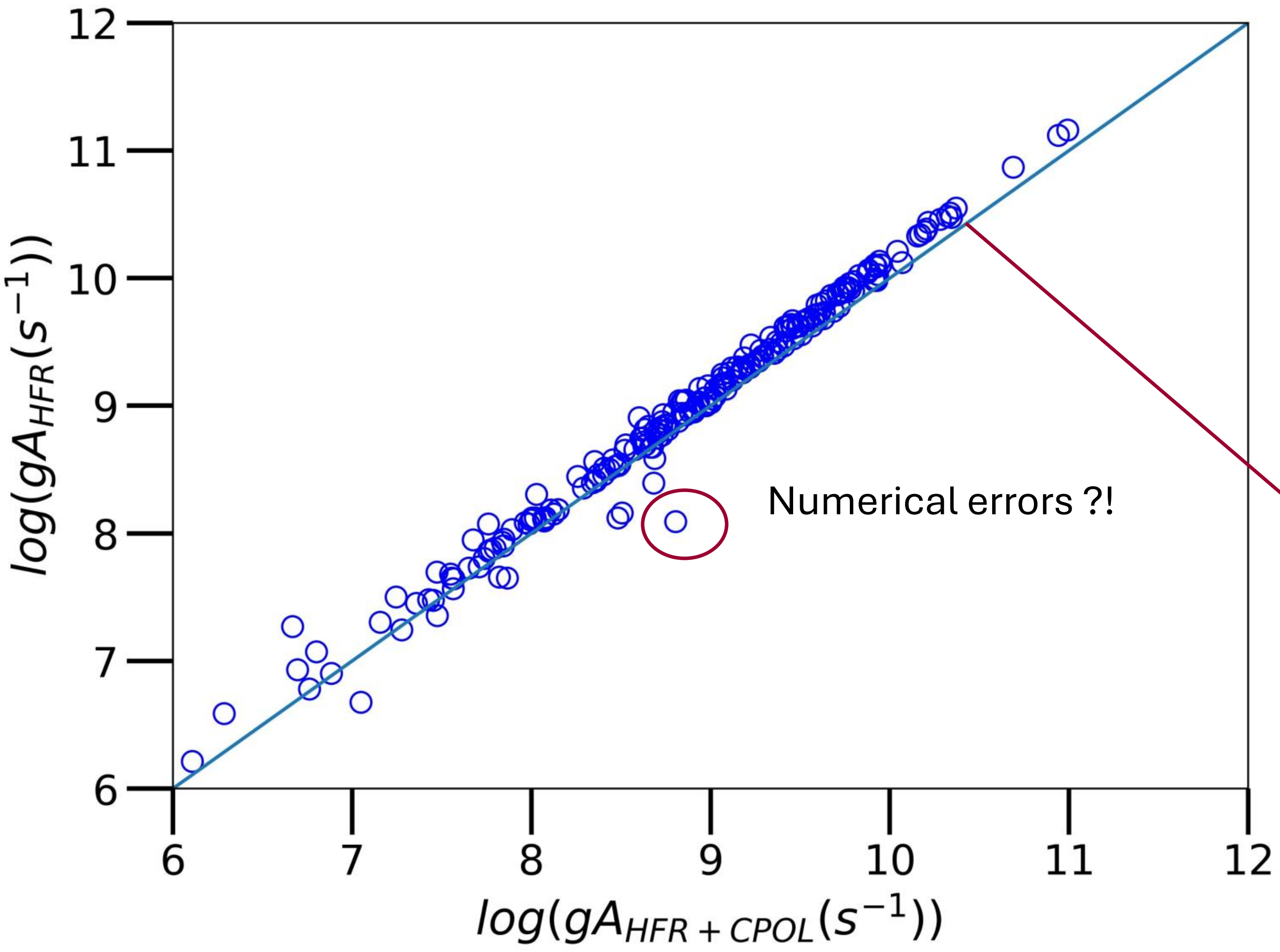
→ σ of the odd parity is significantly improved especially for ions from Re VI to Pt IX that enhance the accuracy of the computed gA's

	Even parity	
	$\sigma_{HFR+CPOL}$ (cm^{-1})	$\sigma_{HFR(CV)}$ (cm^{-1})
Ta IV	22	24
W V	20	31
Re VI	30	37
Os VII	31	40
Ir VIII	35	47
Pt IX	38	53
	Odd parity	
Ta IV	142	156
W V	132	141
Re VI	347	253
Os VII	779	139
Ir VIII	1216	571
Pt IX	3363	417

Radiative decay rates

Ta IV

[Churilov, S.S. et al, 1996 ; Meijer, F.G and Metsch, 1978 ; Kildiyarova, R.R et al, 1996]



229 observed electric dipole lines between 551.422 – 3076.060 Å

Mean ratio $gA_{HFR+CPOL}/gA_{HFR}$=0.82

CPOL corrections reduce gA-values of about 20%, as expected.

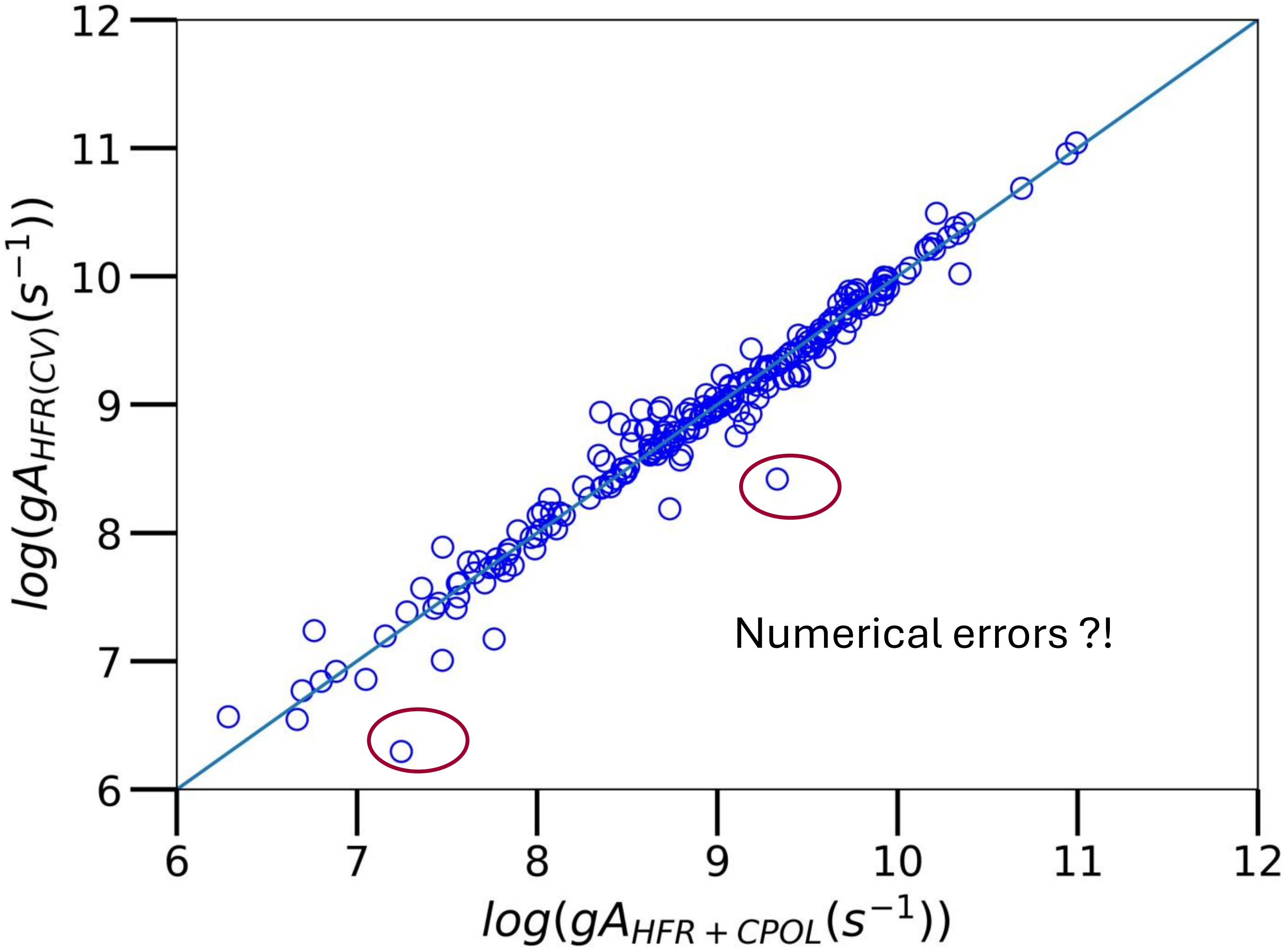
For 5d → 5f,7p ($5d^2$ → 5d5f,5d7p) : reduction is on average 30%

Radiative decay rates

Ta IV

[Churilov, S.S. et al, 1996 ; Meijer, F.G and Metsch, 1978 ; Kildiyarova, R.R et al, 1996]

229 observed electric dipole lines between 551.422 – 3076.060 Å

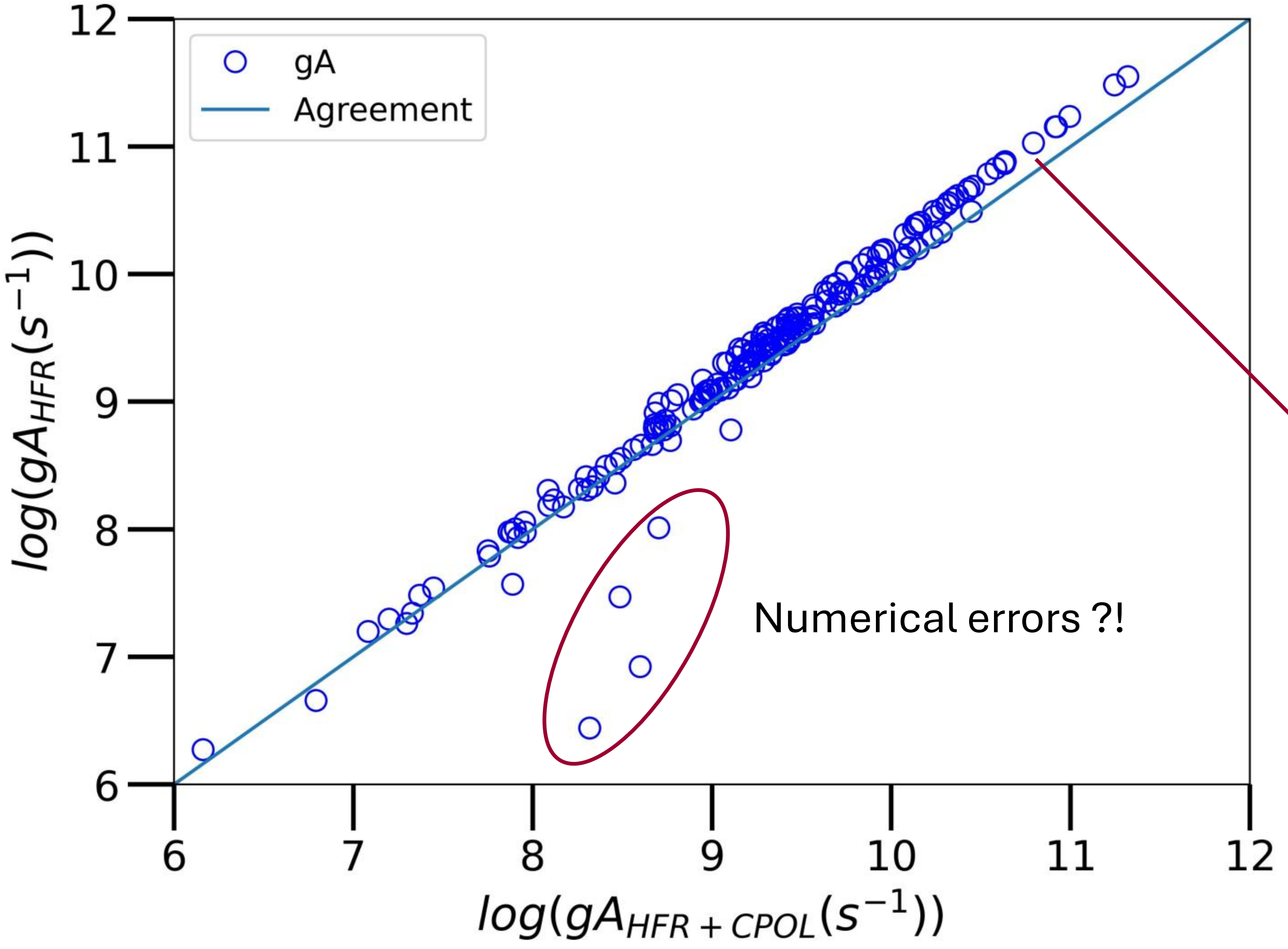


Mean ratio $gA_{HFR(CV)}/gA_{HFR+CPOL}$ = 1.08 ± 0.24

Radiative decay rates

W V

[Nist, Kramida and Shirai, 2009]



193 observed electric dipole lines between 391.566 – 2187.270 Å

Mean ratio $gA_{HFR+CPOL}/gA_{HFR}$=0.79

CPOL corrections reduce gA-values of about 20%, as expected.

For 5d →7p (5d^2 → 5d7p) : reduction is on average 30%

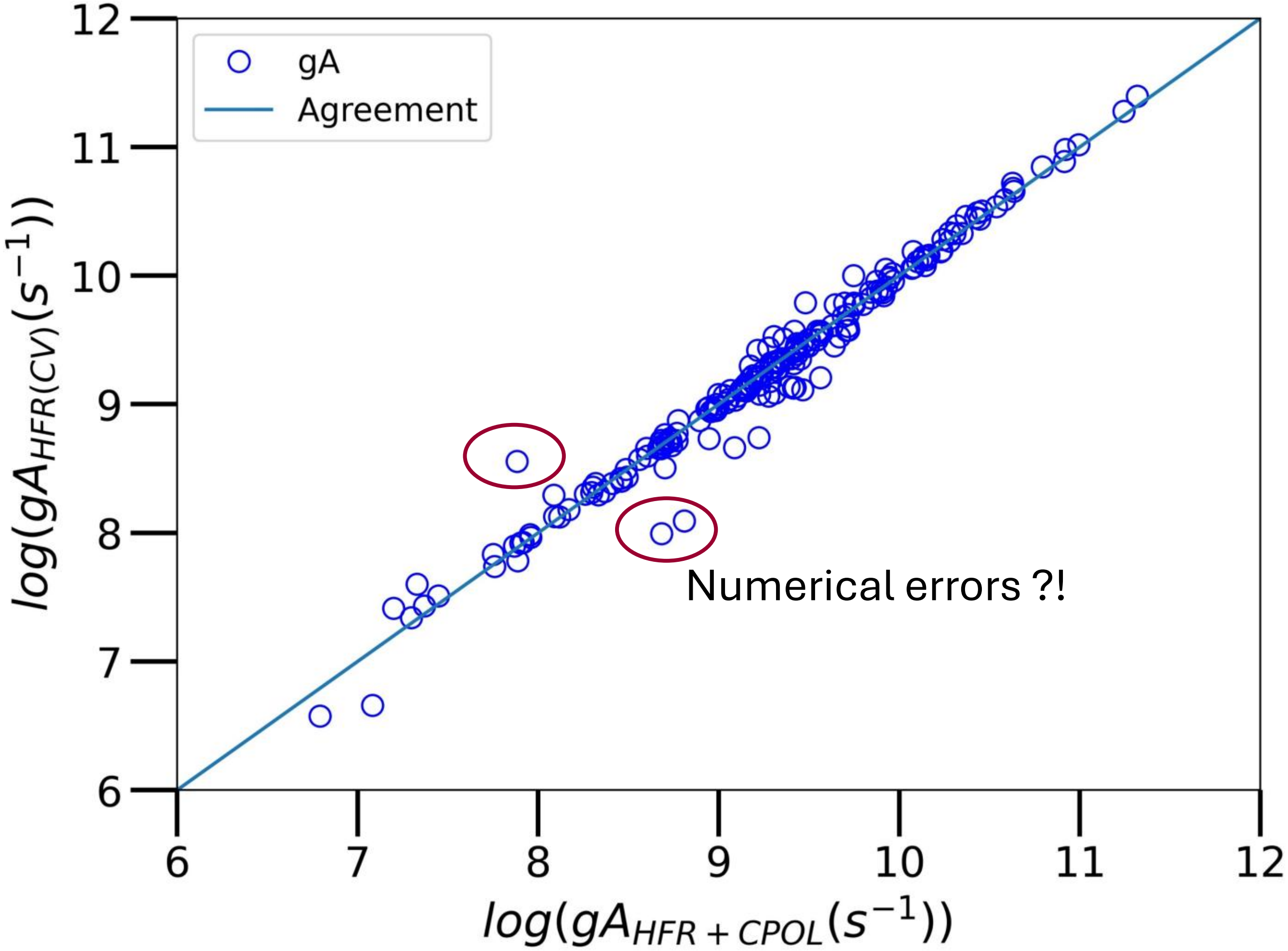
For 5d →5f (5d^2 → 5d5f) : reduction is on average 40%

Radiative decay rates

W V

[Nist, Kramida and Shirai, 2009]

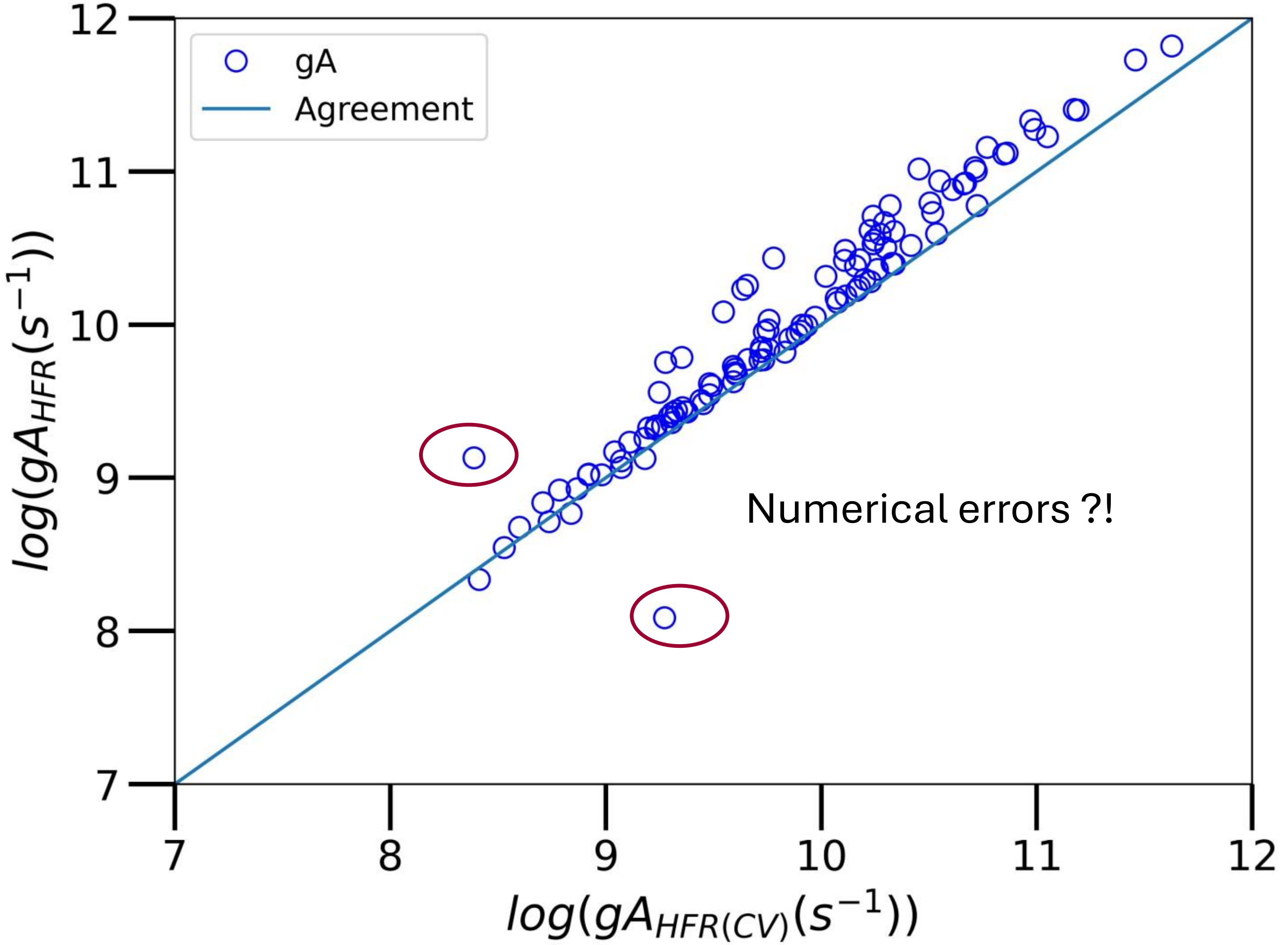
193 observed electric dipole lines between 391.566 – 2187.270 Å



Mean ratio $gA_{HFR(CV)}/gA_{HFR+CPOL}$ = 1.02 ± 0.17

Re VI

112 observed electric dipole lines between 332.044 – 1513.957 Å [Sugar, J. et al, 1994]



Mean ratio $gA_{HFR(CV)}/gA_{HFR}$=0.70 $\pm$ 0.18

CV correlations reduce gA-values of about 30%

Os VII: 149 observed electric dipole lines between 288.274 – 1319.811 Å
Mean ratio $gA_{HFR(CV)}/gA_{HFR}$= 0.74 $\pm$ 0.28 [van het Hof, G.J. et al, 1995 ; Kildiyarova, R.R et al, 1995]

Ir VIII: 129 observed electric dipole lines between 237.098 – 1172.084 Å
Mean ratio $gA_{HFR(CV)}/gA_{HFR}$= 0.65 $\pm$ 0.31 [van het Hof, G.J. et al, 1995 ; Kildiyarova, R.R et al, 1995]

Pt IX: 131 observed electric dipole lines between 216.575 – 1055.171 Å
Mean ratio $gA_{HFR(CV)}/gA_{HFR}$= 0.66 $\pm$ 0.38 [Kildiyarova, R.R et al, 1997 ; Azarov V.I. and Churilov S.S , 1999]

MCDHF preliminary results

MCDHF method

General procedure [Grant, I. P., 2007]

- $H_{DC} = \sum_{i=1}^{N} h_{D_i}$ with $h_{D_i} = c\vec{\alpha}\cdot\vec{p_i} + (\beta - 1)c^2 + V(r_i)$ ($\alpha^j = \gamma^0\gamma^j$ and $\beta = \gamma^0$)

- Each electron: $h_D\varphi = E\varphi \;\rightarrow\; \varphi(r,\theta,\phi) = \frac{1}{r}\begin{pmatrix} P_{n,\kappa}(r)\chi_{\kappa,m}(\theta,\phi) \\ iQ_{n,\kappa}(r)\chi_{\kappa,m}(\theta,\phi)\end{pmatrix}$ where $P_{n,\kappa}(r)$ and $Q_{n,\kappa}(r)$ are large and small radial part, respectively.

- $P_{n,\kappa}(r)$, $Q_{n,\kappa}(r)$? → solve MCDHF equations (Self-Consistent Field method)

- CI: (ASF) $\Psi(P,J,M) = \sum_{r=1}^{n_c} c_r\,\Phi(\gamma_r,P,J,M)$

MCDHF Models

- MCDHF method implemented in the GRASP18 packages [C. Froese Fischer et al, 2019]
- The Atomic State Functions (ASFs) are built with the Active Set approach ($n_{max}l, n'_{max}l', \ldots$):
- Optimisation of all orbitals (5s,5p,5d,4f) on the 5d^2 $^3F_2$ ground state
- MR : (re)optimize valence orbitals on all levels
- VV1,2,3 : ONLY « new » correlation orbitals are optimized on all levels of the MR
- CV,CC : Relativistic Configuration Interactions (RCI) calculations

Even parity		Number of CSFs
MR ($5d^2$,5d6s,$6s^2$)	(6s,5p,5d,4f)	14
VV1	SD(MR) → (7s,6p,6d,5f)	81
VV2	SD(MR) → (8s,7p,7d,6f)	225
VV3	SD(MR) → (9s,8p,8d,7f)	446
CV1 (RCI)	SrD (MR{4f}) → (9s,8p,8d,7f)	29 249
CV2 (RCI)	SrD (MR{4f,5p}) → (9s,8p,8d,7f)	47 273
CV3 (RCI)	SrD (MR{4f,5p,5s}) → (9s,8p,8d,7f)	53 778
CC1 (RCI)	SrD (MR{4f(2)}) → (9s,8p,8d,7f)	274 515
CC2 (RCI)	SrD (MR{4f,5p}) → (9s,8p,8d,7f)	367 215
CC3 (RCI)	SrD(MR{4f,5p}) + SrD (MR{5s}) → (9s,8p,8d,7f)	373 720
Odd parity		**Number of CSFs**
MR (5d6p,6s6p,5d5f,5d7p)	(6s,7p,5d,5f)	47
VV1	SD(SR) → (7s,8p,6d,6f)	188
VV2	SD(SR) → (8s,9p,7d,7f)	399
VV3	SD(SR) → (9s,10p,8d,8f)	688
CV1 (RCI)	SrD (MR{4f}) → (9s,10p,8d,8f)	115 980
CV2 (RCI)	SrD (MR{4f,5p}) → (9s,10p,8d,8f)	184 664
CV3 (RCI)	SrD (MR{4f,5p,5s}) → (9s,10p,8d,8f)	208 892
CC1 (RCI)	SrD(MR{4f(2)}) → (9s,10p,8d,8f)	1 980 405
CC2 (RCI)	SrD(MR{4f,5p}) → (9s,10p,8d,8f)	2 574 277
CC3 (RCI)	SrD(MR{4f,5p}) + SrD (MR{5s})→ (9s,10p,8d,8f)	2 602 097

Convergence of the models

Ta IV

	Even parity		Odd parity	
	$\Delta E/E_{obs}$	$\Delta E/E_{prev}$	$\Delta E/E_{obs}$	$\Delta E/E_{prev}$
MR	9.99%		1.53%	
VV1	4.12%	5.91%	1.52%	0.35%
VV2	3.75%	0.39%	1.56%	0.05%
VV3	3.69%	0.06%	1.57%	0.01%
CV1	5.25%	1.86%	0.71%	1.44%
CV2	7.71%	3.35%	2.11%	2.08%
CV3	6.40%	1.31%	2.33%	0.21%
CC1	6.59%	1.60%	10.60%	9.48%
CC2	6.90%	0.86%	5.10%	2.83%
CC3	5.65%	0.88%	5.23%	2.74%

303 transitions over 317 have a ratio $gA_{HFR+CPOL}/gA_{MCDHF}$ < 10 with a mean ratio 1.39

299 transitions over 317 have a ratio $gA_{HFR+CPOL}/gA_{MCDHF}$ < 10 with a mean ratio 1.26

Even parity		Number of CSFs
MR ($5d^2$,5d6s,$6s^2$)	(6s,5p,5d,4f)	14
VV1	SD(MR) → (7s,6p,6d,5f)	81
VV2	SD(MR) → (8s,7p,7d,6f)	225
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Odd parity		**Number of CSFs**
MR (5d6p,6s6p,5d5f,5d7p)	(6s,7p,5d,5f)	47
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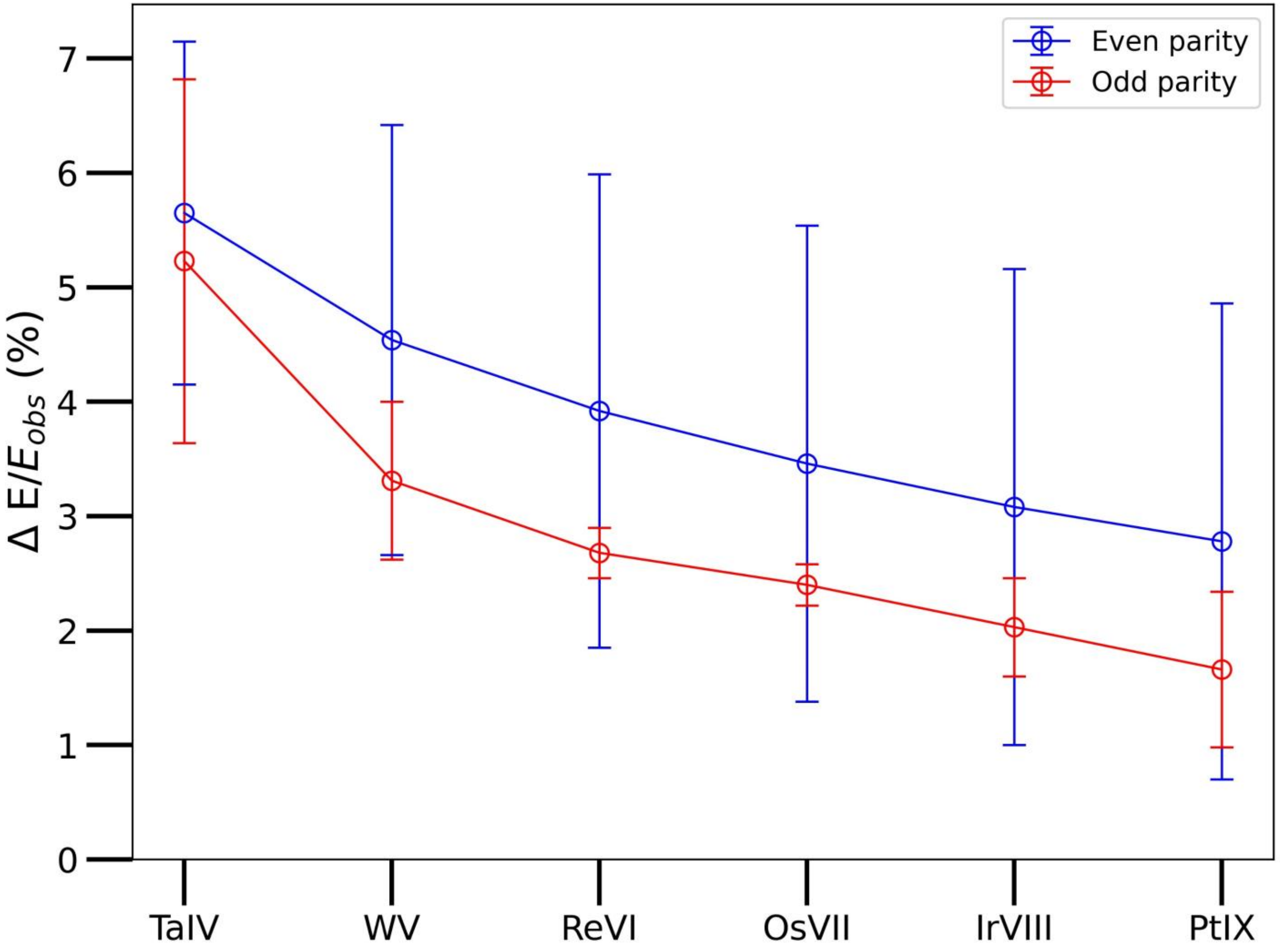
CC2 (RCI)	SrD(MR{4f,5p}) → (9s,10p,8d,8f)	2 574 277
CC3 (RCI)	SrD(MR{4f,5p}) + SrD (MR{5s})→ (9s,10p,8d,8f)	2 602 097

Even parity (CC3) : SD (MR {4f,5p}) + SrD (MR {5s}) → (9s,8p,8d,7f)

Odd parity (CC3) : SD (MR {4f,5p}) + SrD (MR {5s}) → (9s,10p,8d,8f)

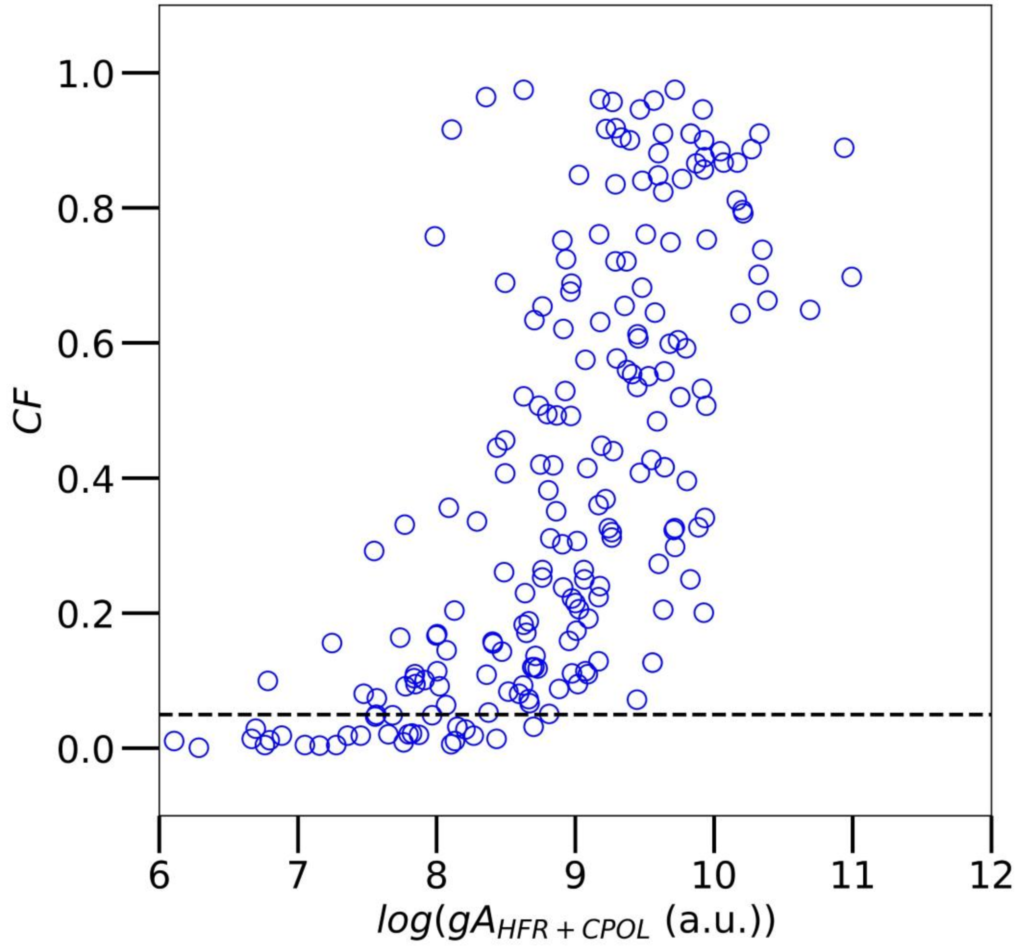
	$\Delta E/E_{obs}$	
	Even parity	Odd parity
TaIV	5.65%	5.23%
WV	4.54%	3.31%
ReVI	3.92%	2.68%
OsVII	3.46%	2.40%
IrVIII	3.08%	2.03%
PtIX	2.78%	1.80%



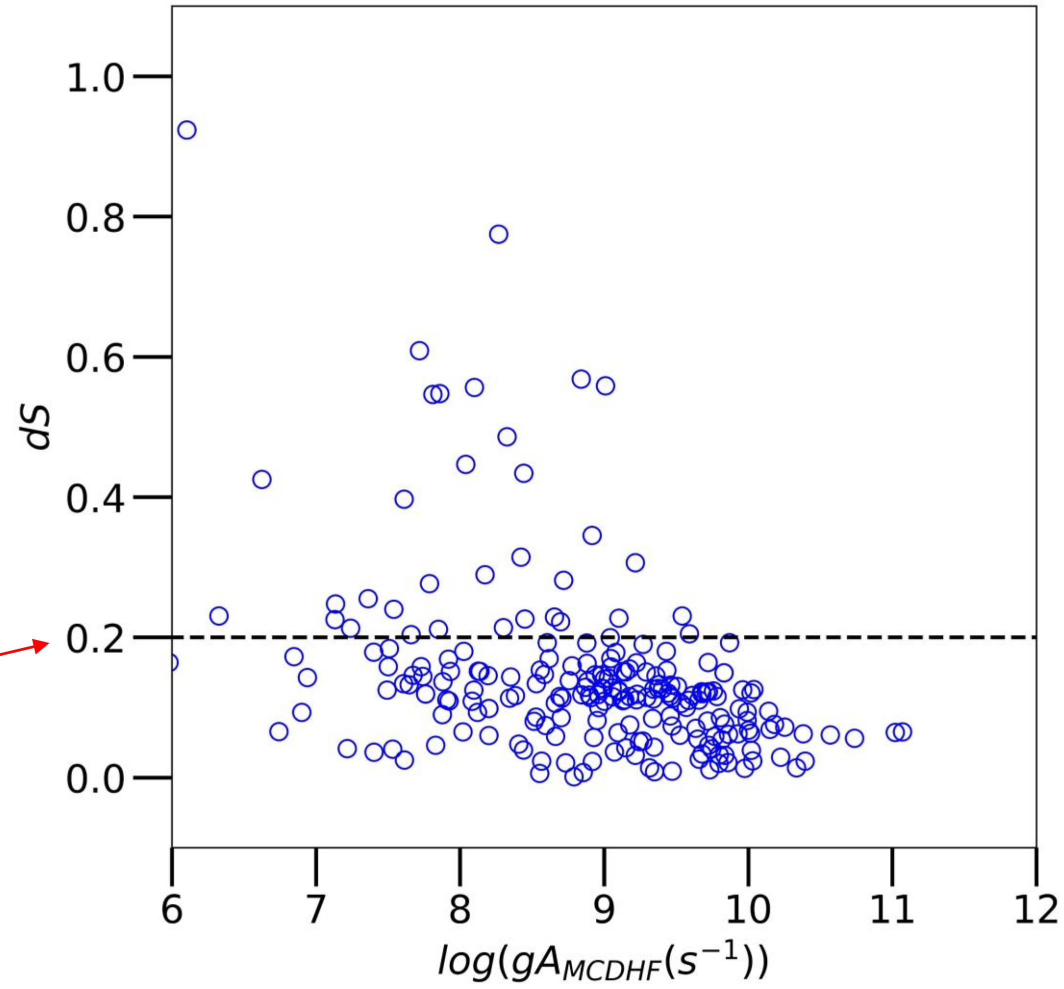
Comparisons between HFR+CPOL and MCDHF gA's Ta IV

Cancellation Factor $CF_{ij} = \left(\frac{|\sum_b \sum_c y_j^b y_i^c \langle \psi_c | \mathbf{P}^{(1)} | \psi_b \rangle |}{\sum_b \sum_c |y_j^b y_i^c \langle \psi_c | \mathbf{P}^{(1)} | \psi_b \rangle |} \right)^2$
[Cowan, R. D. ,1981]

Uncertainty of MCDHF transition rates : $dS = \frac{|S_B - S_C|}{\max(S_B, S_C)}$



0.05
[Cowan, R. D. ,1981]



Comparisons between HFR+CPOL and MCDHF gA's

Ta IV

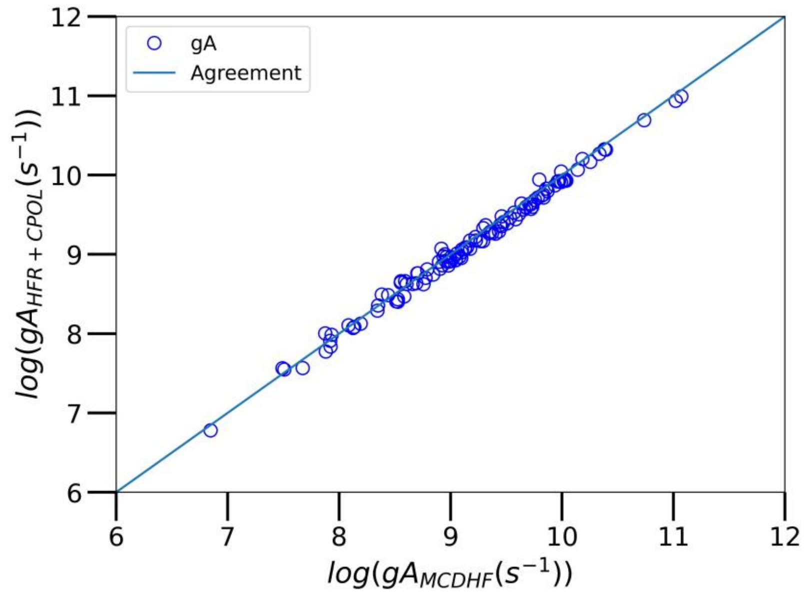
- It remains 170 transitions (among 229) with $CF > 0.05$ and dS < 0.2
- 117 transitions have an uncertainty lower than 30%
- The uncertainty on the HFR+CPOL and MCDHF results estimated at 15% on average

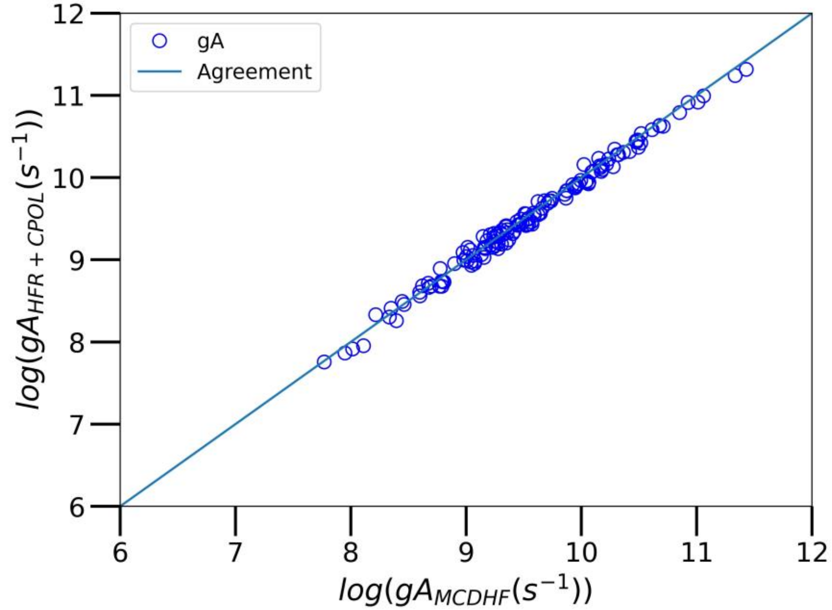
$$\frac{\Delta gA}{\max} \text{ where } \Delta gA = |gA_{HFR+CPOL} - gA_{MCDHF}|$$
and max=max($gA_{HFR+CPOL}; gA_{MCDHF}$)

- Mean ratio $gA_{HFR+CPOL}/gA_{MCDHF}$= 0.91 $\pm$ 0.12

W V

- It remains 158 transitions (among 193) with $CF > 0.05$ and dS < 0.2
- 140 transitions have an uncertainty lower than 30%
- The uncertainty estimated at 13% on average with a mean ratio 0.94 $\pm$ 0.12

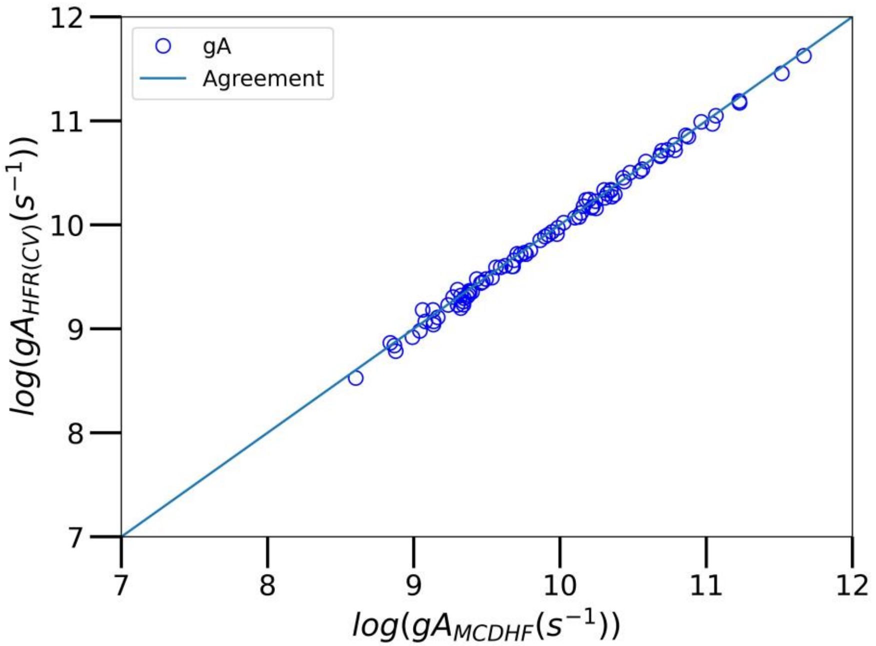




Comparisons between HFR(CV) and MCDHF gA's

Re VI

- It remains 94 transitions (among 112) with $CF > 0.05$ and $dS < 0.2$
- 87 transitions have an uncertainty lower than 30%
- The uncertainty estimated at 9% on average with a mean ratio 0.95 ± 0.07



Os VII

- The uncertainty of 84 transitions (among 149) estimated at 9% on average with a mean ratio 0.96 ± 0.08

Ir VIII

- The uncertainty of 73 transitions (among 129) estimated at 8% on average with a mean ratio 0.98 ± 0.08

Pt IX

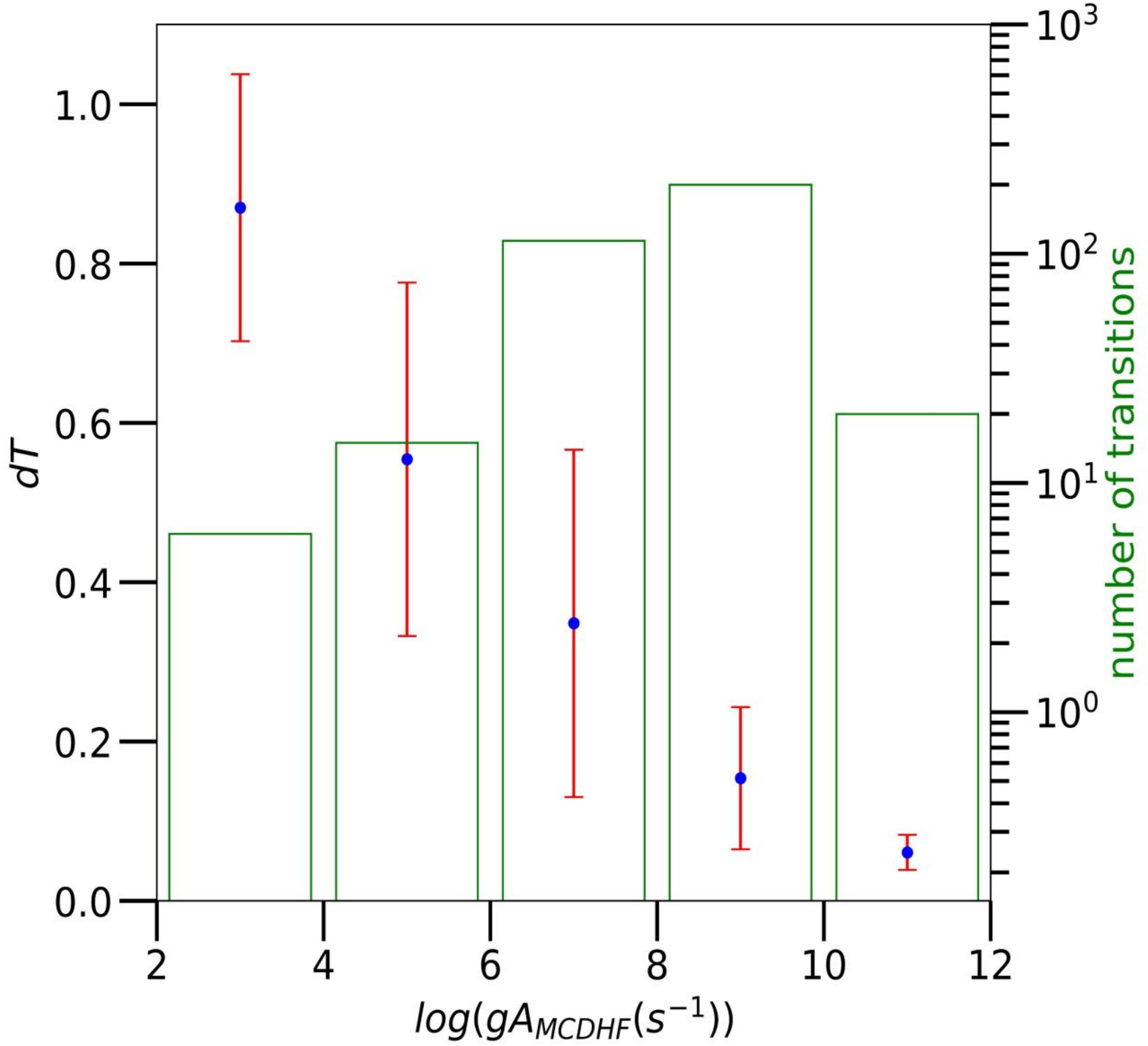
- The uncertainty of 61 transitions (among 131) estimated at 6% on average with a mean ratio 0.99 ± 0.07

Ta IV

- All 355 transitions computed in the MCDHF method

⇒ 251 transitions have $dT < 0.25$

⇒ The more intense the transitions, the lower the dT value



Conclusion

- Origin of Tantalum up to Platinium in the fusion plasma (tungsten transmutation)
- Compute radiative parameters with HFR+CPOL method + least squares adjustement
- Increasing the ionic charge, configurations with an open 4f,5p core orbitals begin to overlap the valence configurations of interest → Introduce explicitly these configurations for Re VI → Pt IX
- Good agreement between HFR+CPOL and MCDHF method and comparisons allow to estimate uncertainty of a selected set of new transition probabilities (TaIV:15%; WV:13%; ReVI:9%; OsVII:9%; IrVIII:8%; PtIX:6%)
  - ➢ set of new accurate atomic data for TaIV → PtIX for plasma diagnostics
- Same procedure for higher ionic charge state of tungsten transmutation products (Ta, Re, Os, Ir, Pt)
- However, few atomic data published

THANK YOU !

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