Radiative data calculations for electric dipole transitions in the Yb I isoelectronic sequence (Ta IV - Pt IX) of interest to nuclear fusion diagnostics

Maxime Brasseur

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UMONS
University of Mons

Physique Atomique et Astrophysique

Faculty of Science

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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)

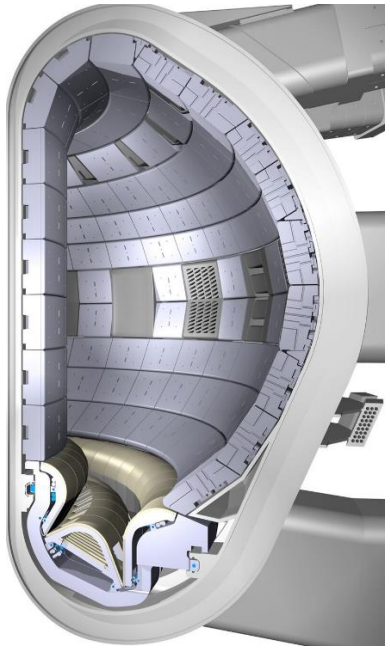
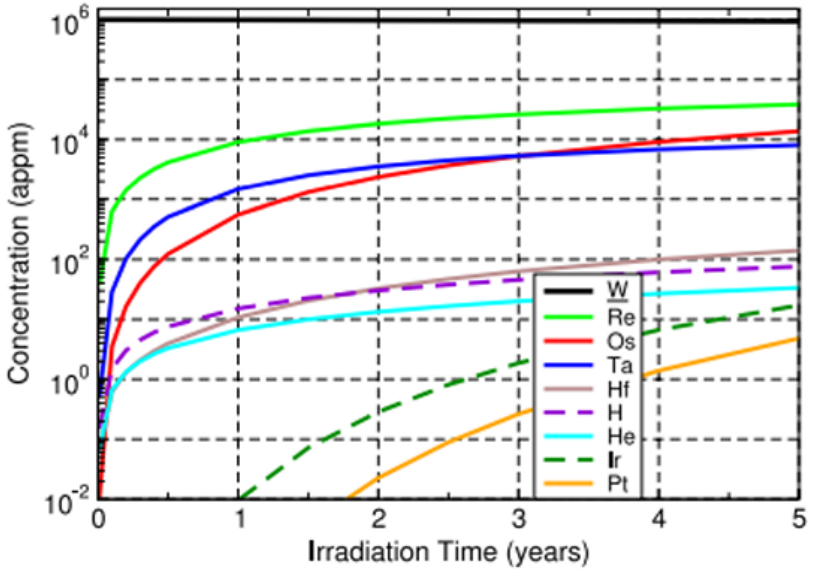


Illustration of the inside of the Tokamak. (https://encyclopedia.pub/entry/37629)



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] :

  - $$\frac{\varepsilon_z(p\rightarrow q)}{\varepsilon_z(p'\rightarrow q')} = \frac{\lambda_{p'q'}}{\lambda_{pq}} \frac{A_z(p\rightarrow q)}{A_z(p'\rightarrow q')} \frac{A_z(p'\rightarrow)}{A_z(p\rightarrow)} \frac{X_z(g\rightarrow p)(T_e)}{X_z(g\rightarrow p')(T_e)} \Rightarrow T_e$$

  - $$\frac{\varepsilon_z(p\rightarrow q)}{\varepsilon_z(p'\rightarrow q')} = \frac{\lambda_{p'q'}}{\lambda_{pq}} \frac{A_z(p\rightarrow q)}{A_z(p'\rightarrow q')} \frac{A_z(p'\rightarrow)}{A_z(p\rightarrow)} \frac{X_z(g\rightarrow p)(T_e)}{X_z(g\rightarrow p')(T_e)} \left[1 + n_e \frac{X_z(p'\rightarrow)}{A_z(p'\rightarrow)}\right] \Rightarrow n_e$$

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

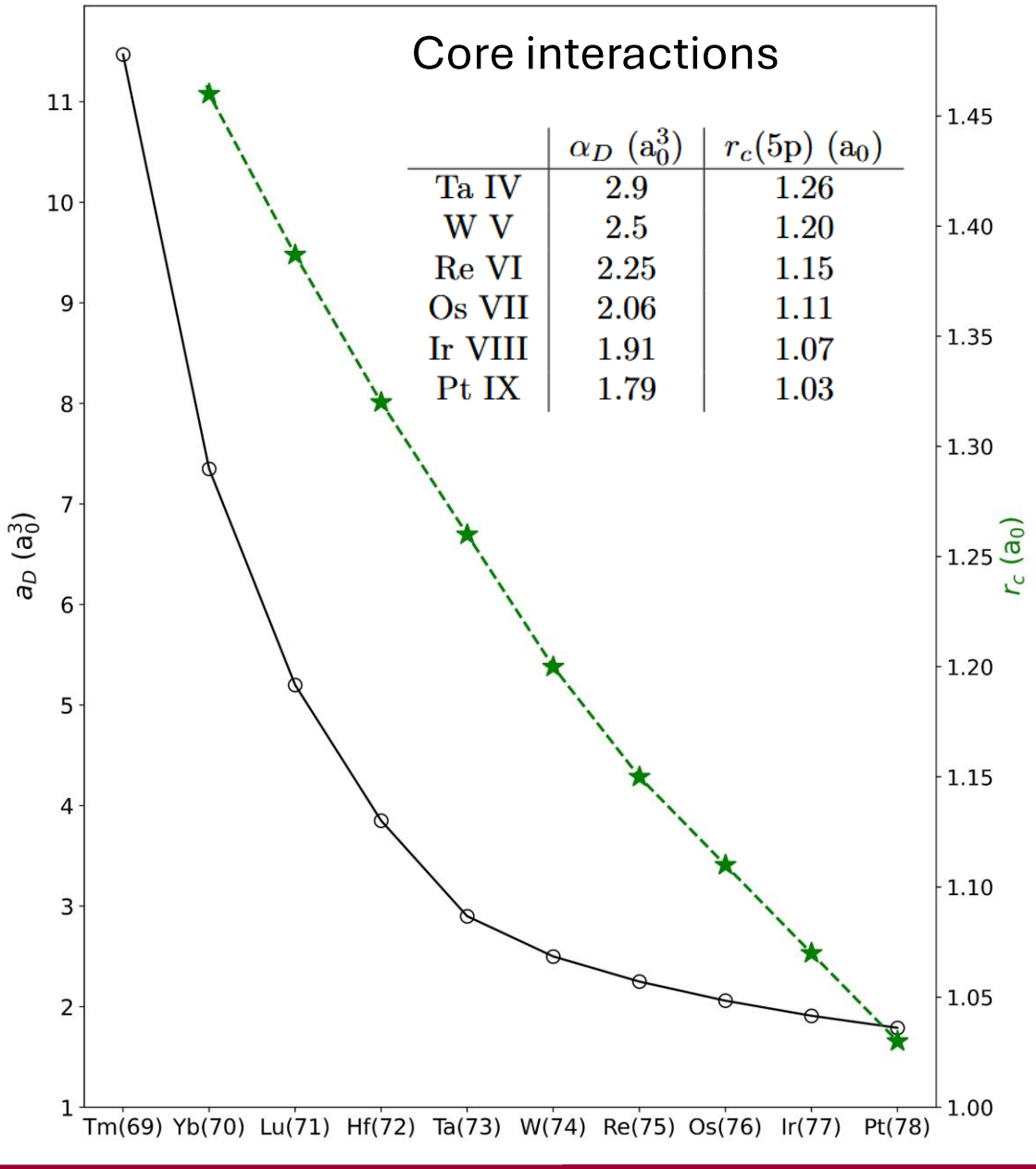
Core polarisation correction

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



Least squares method

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

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

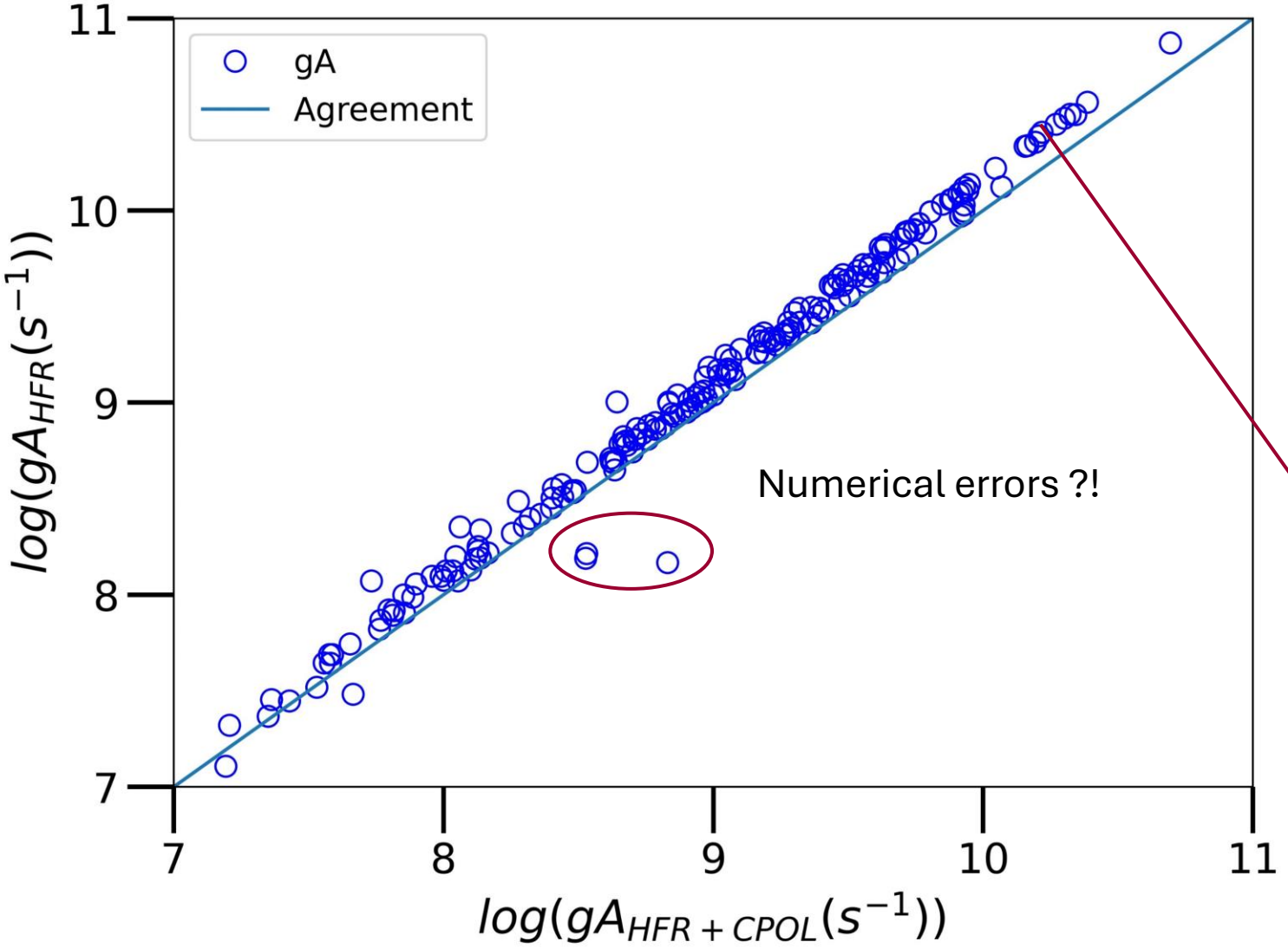
	Even parity		Odd parity	
	nbr of fitted levels	σ (cm^{-1})	nbr of fitted levels	σ (cm^{-1})
Ta IV	14	23	44	145
W V	13	20	44	132
Re VI	13	30	29	347

[Churilov, S.S. et all, 1996 ; Meijer, F.G and Metsch, 1978 ; Kildiyarova, R.R et all, 1996 ; Kramida , A. et all, 2024 ; Sugar, J. et all, 1994 ; Yoca, S.E et all, 2012]

Radiative decay rates

Ta IV

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



213 observed electric dipole lines between 551.422 – 3076.060 Å

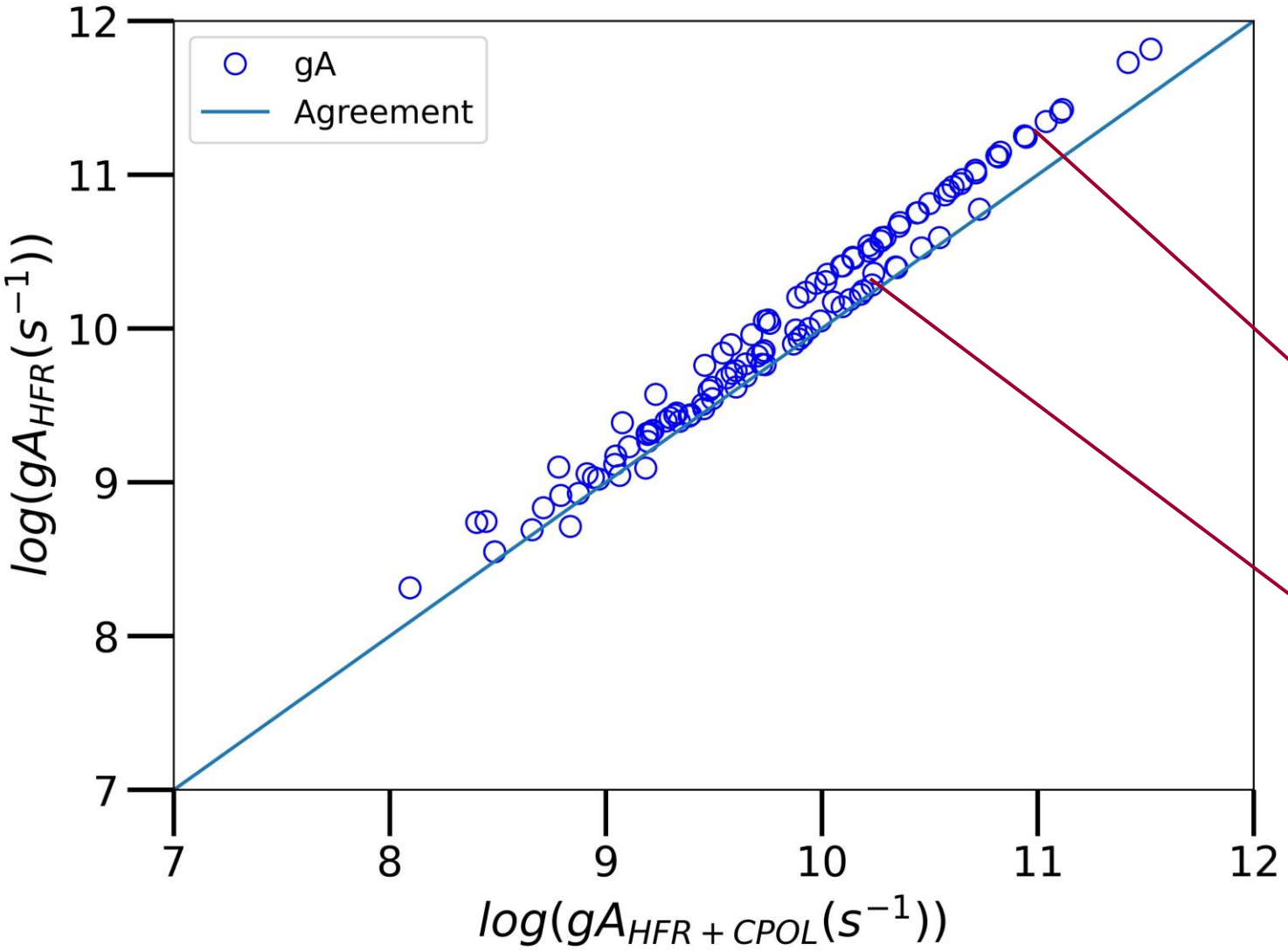
Mean ratio $gA_{HFR+CPOL}/gA_{HFR}$=0.83

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

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

Re VI

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



Mean ratio $gA_{HFR+CPOL}/gA_{HFR}$=0.69

CPOL corrections reduce more gA-values of about 30%

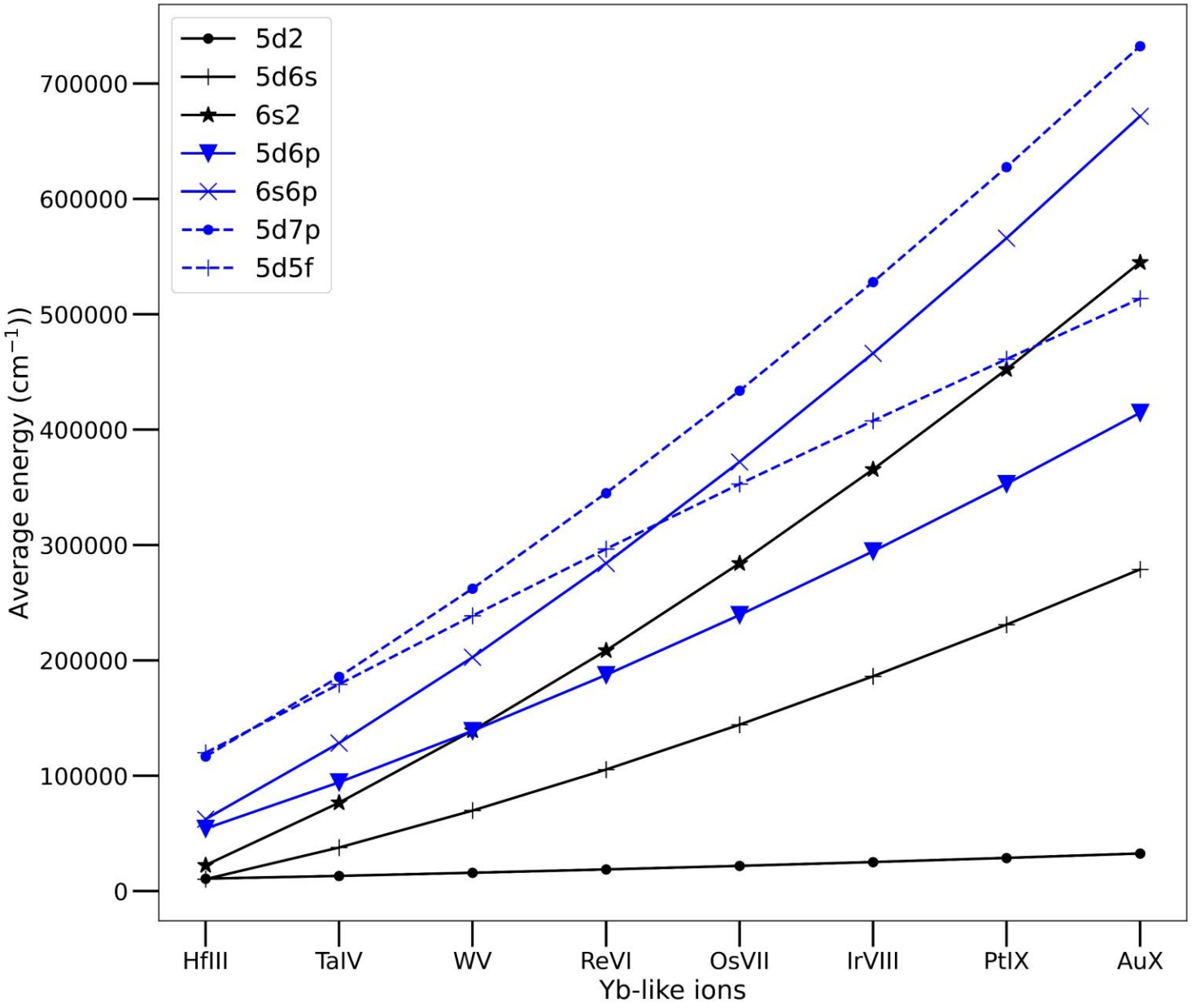
For 5d → 5f ($5d^2$ → 5d5f) : mean ratio $gA_{HFR+CPOL}/gA_{HFR}$ = 0.50

For 5d,6s → 6p ($5d^2$,5d6s → 5d6p) : mean ratio $gA_{HFR+CPOL}/gA_{HFR}$ = 0.85

⇒ E_{av} of configurations with open 4f,5p core orbital closer to valence configurations

Configuration Interactions

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

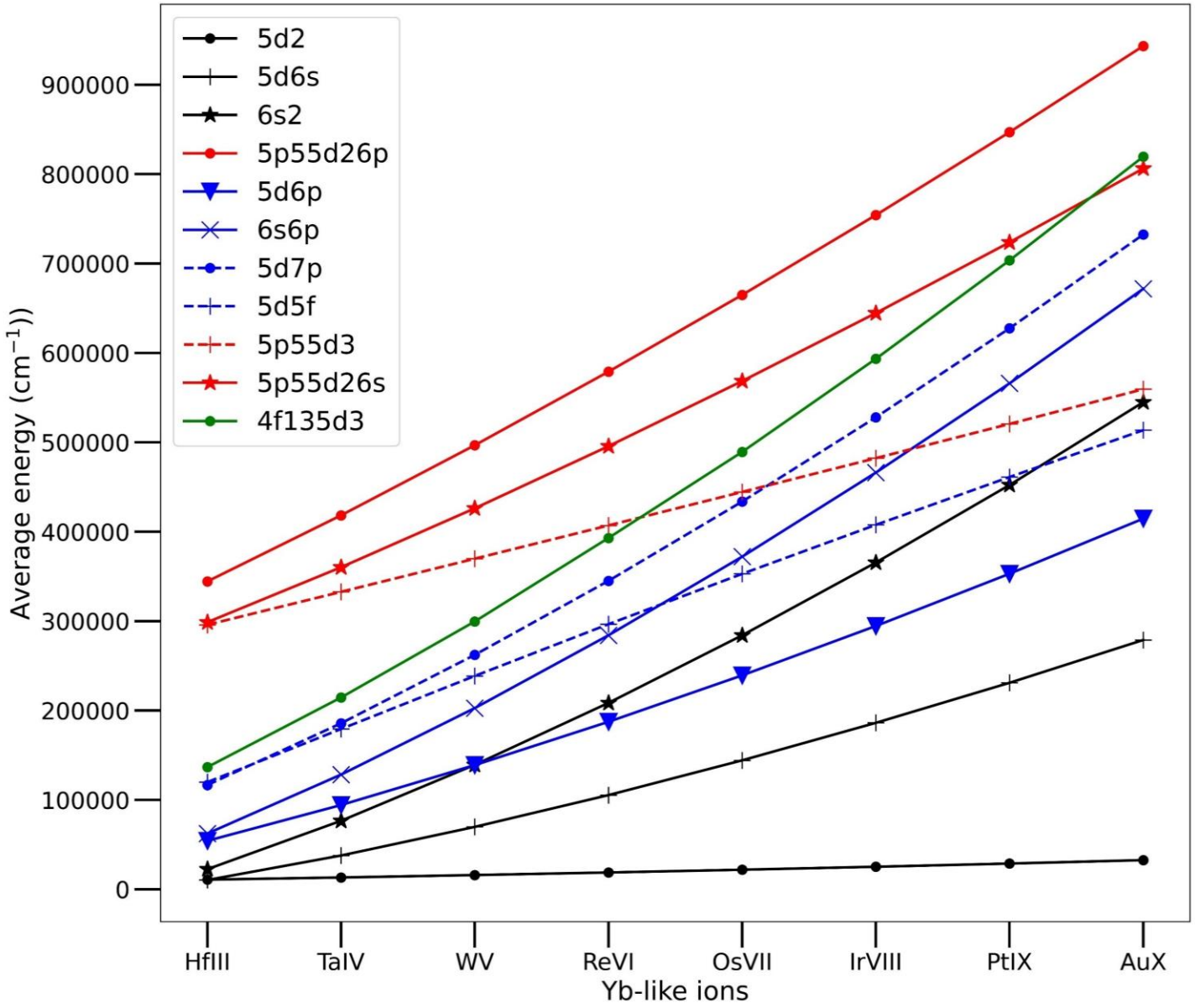


Core-valence correlations

$5p^5 5d^3$ 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)



MCDHF preliminary results

MCDHF method

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

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

- The Atomic State Functions (ASFs) are built with the Active Set approach $(n_{max}l, n'_{max}l', \dots)$:
- 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)	SD (MR{4f(2)}) → (9s,8p,8d,7f)	274 515
CC2 (RCI)	SD (MR{4f,5p}) → (9s,8p,8d,7f)	367 215
CC3 (RCI)	SD (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)	SD (MR{4f(2)}) → (9s,10p,8d,8f)	1 980 405
CC2 (RCI)	SD (MR{4f,5p}) → (9s,10p,8d,8f)	2 574 277
CC3 (RCI)	SD (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
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CC3 (RCI)	SD (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
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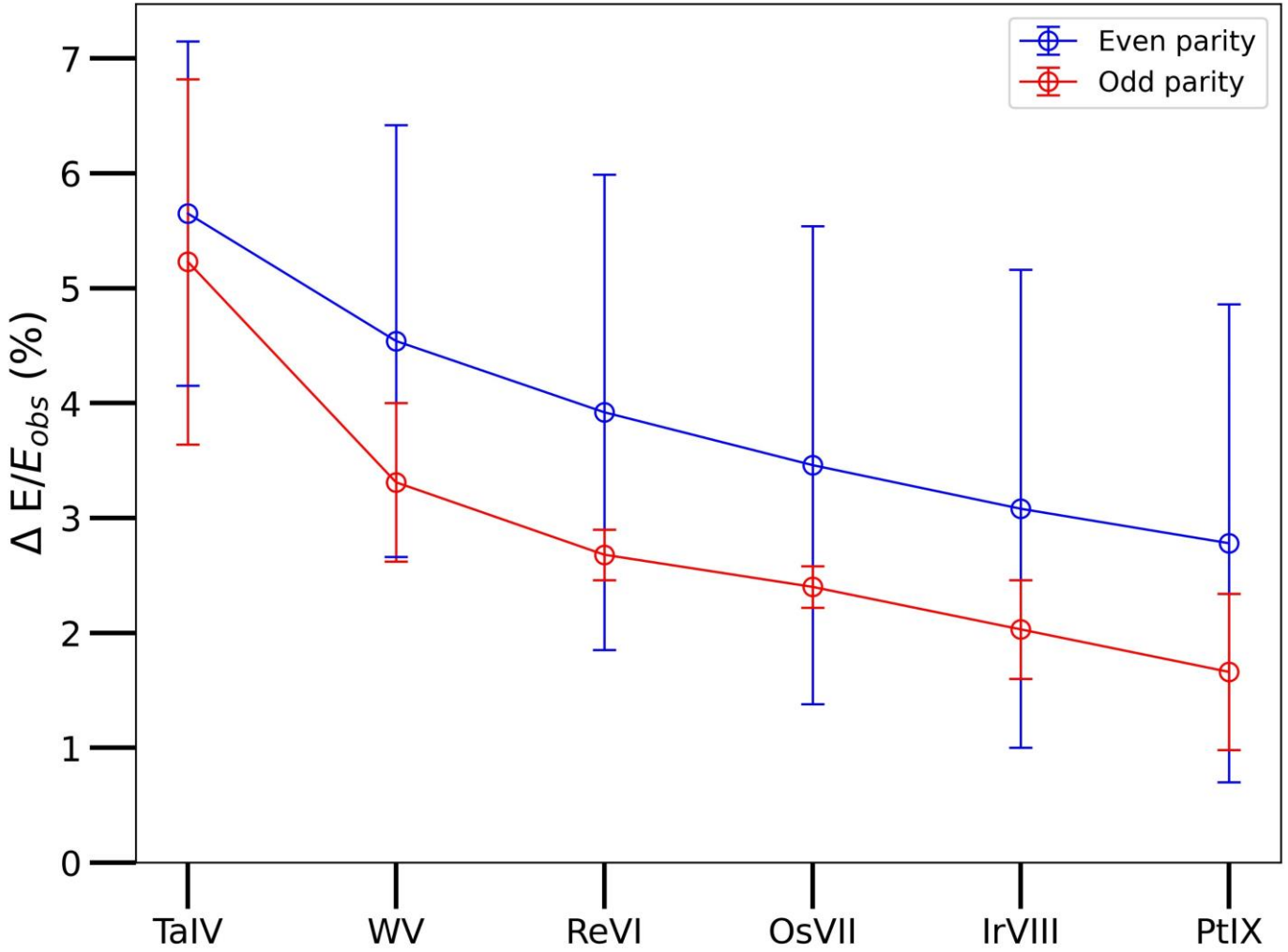
CC3 (RCI)	SD (MR{4f,5p}) + SrD (MR{5s})→ (9s,10p,8d,8f)	2 602 097

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)

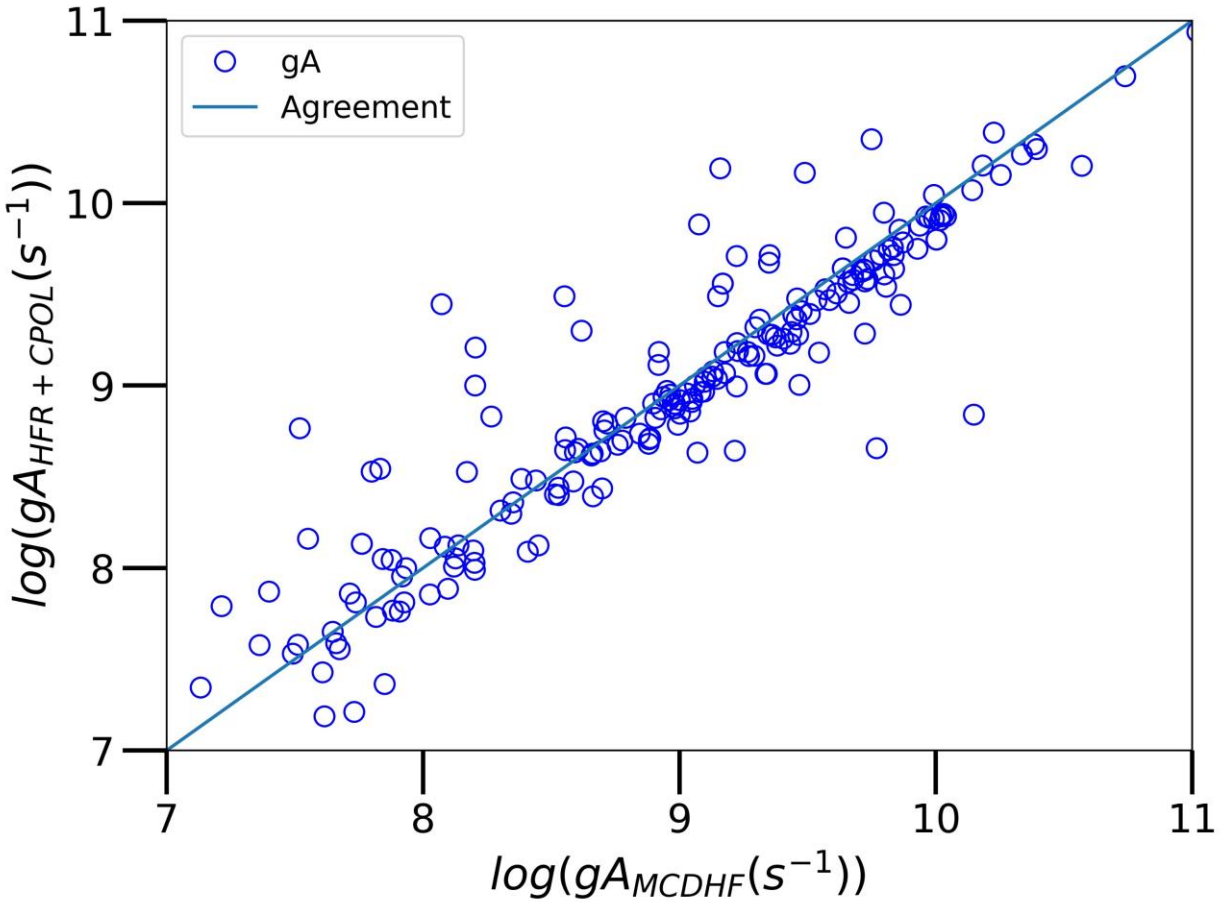
	$\Delta E/E_{obs}$	
	Even parity	Odd parity
TaIV	5.65%	5.23%
WV	4.54%	3.31%
ReVI	3.92%	2.68%
OsVII	3.46%	2.40%
IrVIII	3.08%	2.03%
PtIX	2.78%	1.80%

$5d^2\ {}^3P_0$, $5d^2\ {}^1S_0$: $\Delta E/E_{obs} > 8\%$



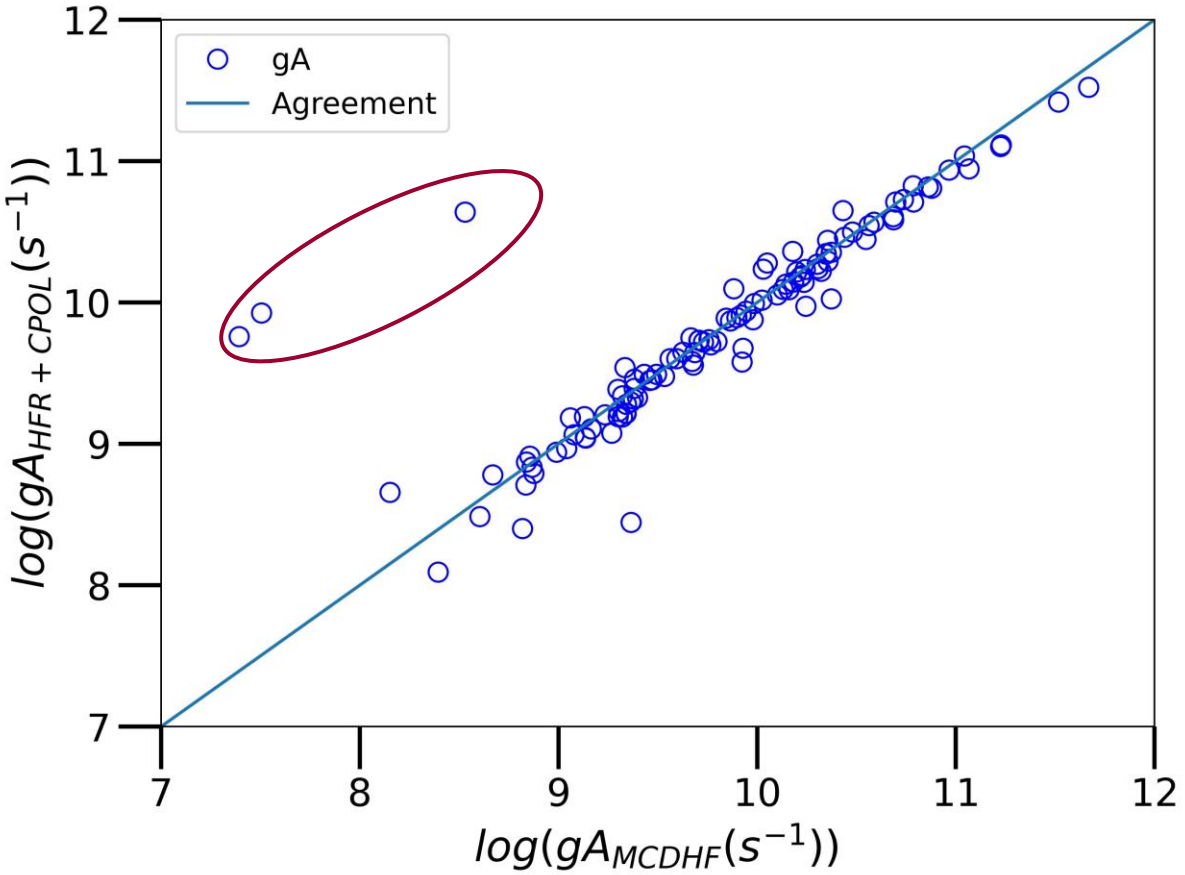
Comparisons between HFR+CPOL and MCDHF gA's

Ta IV



Mean relative difference $\Delta gA/\max(gA_{HFR+CPOL}; gA_{MCDHF})$= 32.3%
Mean ratio $gA_{HFR+CPOL}/gA_{MCDHF}$= 2.16 ± 2.30

Re VI



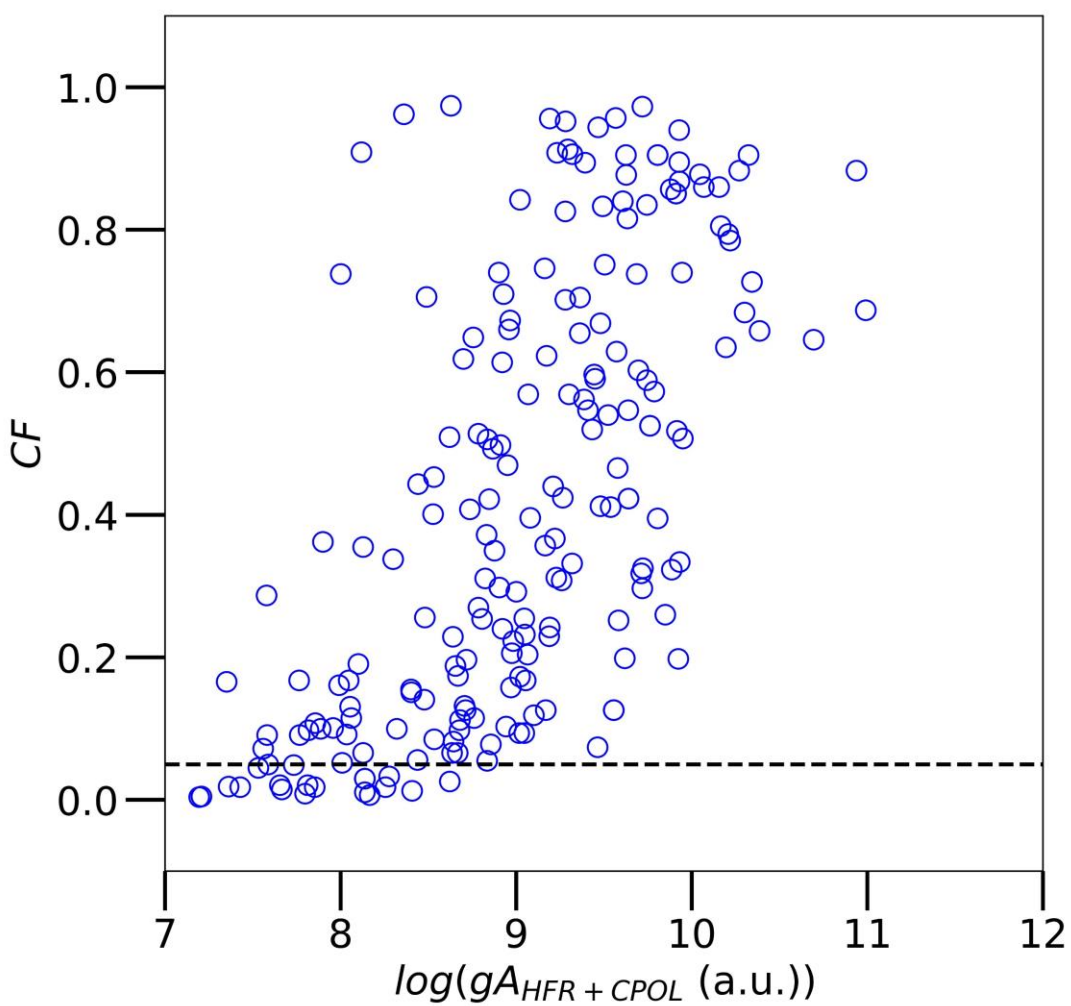
$\Delta gA/\max(gA_{HFR+CPOL}; gA_{MCDHF})$= 18.9%
$gA_{HFR+CPOL}/gA_{MCDHF}$= 6.52 ± 10.81
(three high ratios : 129, 232 and 263)

All gA (and gf) computed in both methods are not reliable

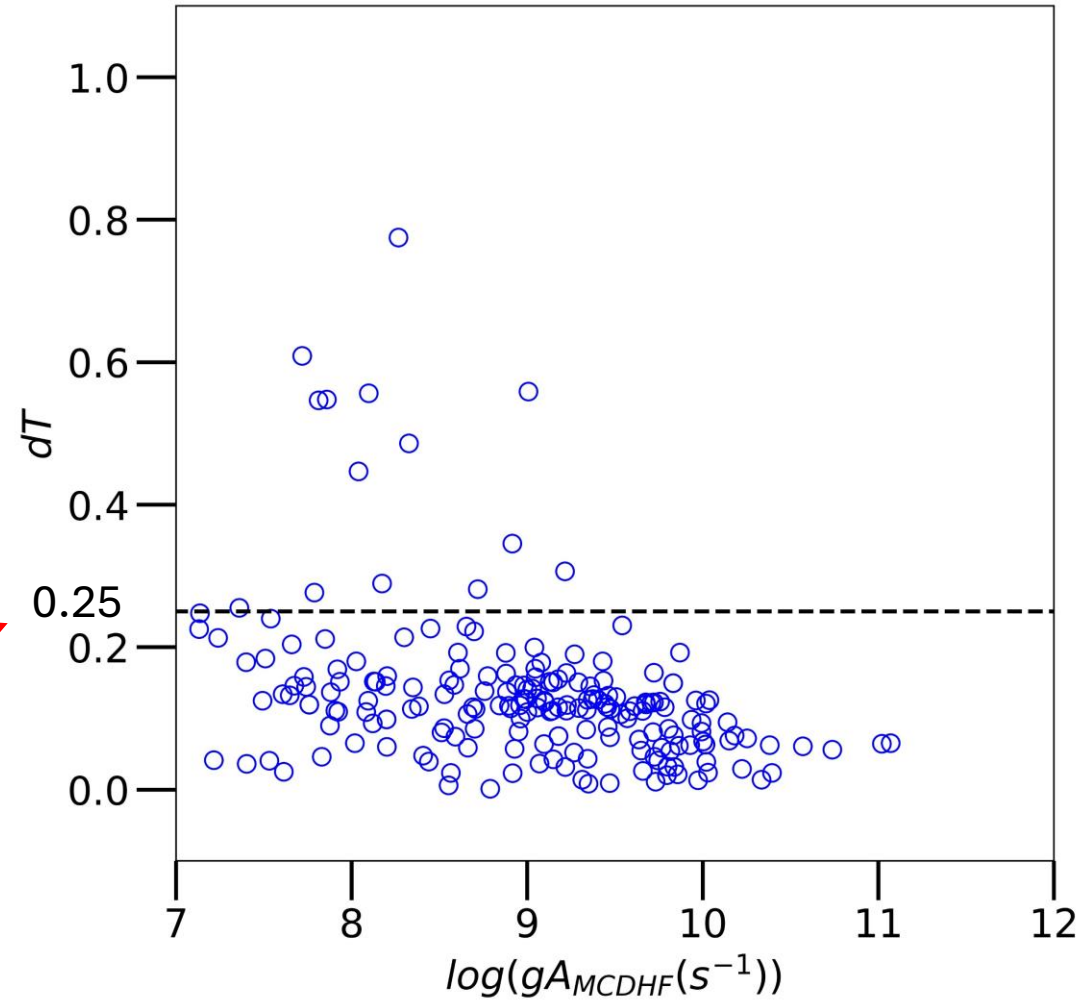
Ta IV

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 : $dT = \frac{|A_B - A_C|}{\max(A_B, A_C)}$



0.05
[Cowan, R. D. ,1981]



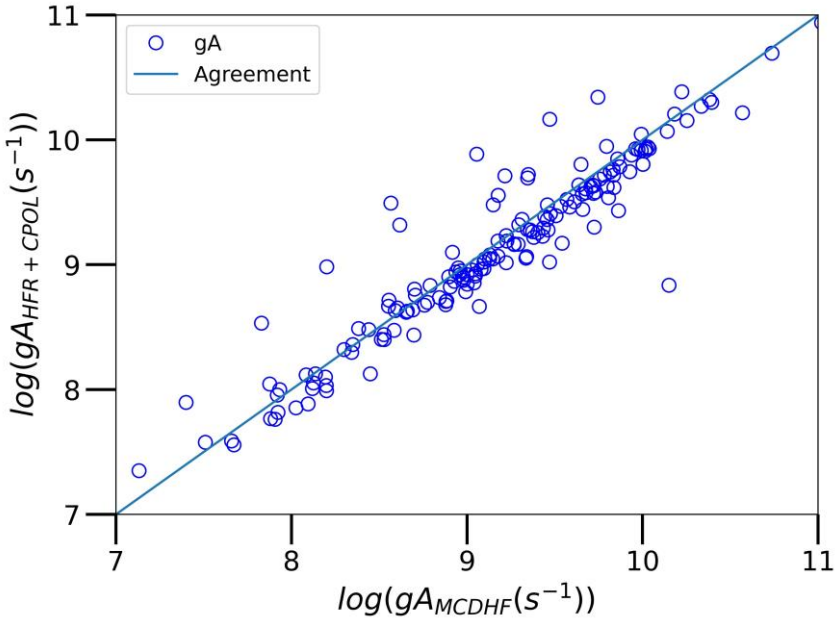
Ta IV

- It remains 170 transitions (among 213) with $CF > 0.05$ and $dT < 0.25$

- The uncertainty on the HFR+CPOL and MCDHF results estimated on average 26%

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

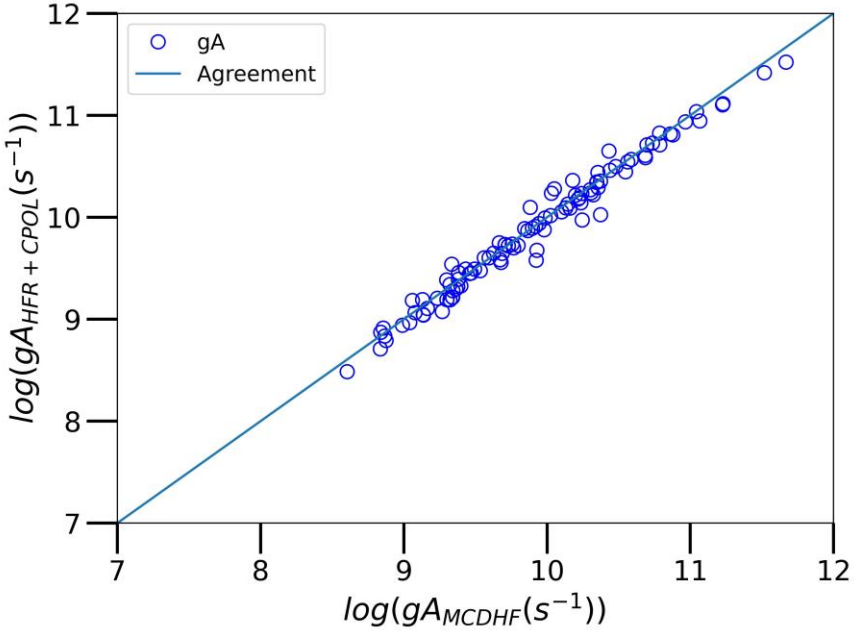
- Mean ratio $gA_{HFR+CPOL}/gA_{MCDHF}$= 1.12 $\pm$ 0.54



Re VI

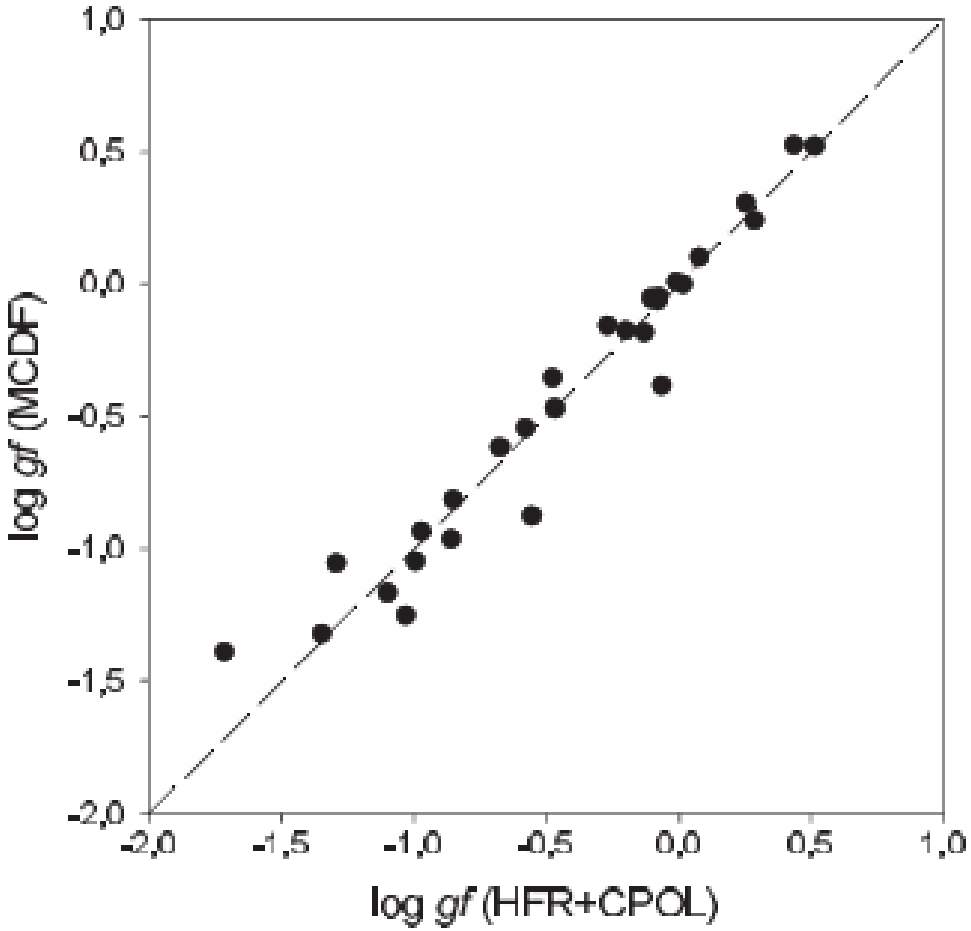
- It remains 101 transitions (among 112) with $CF > 0.05$ and $dT < 0.25$

- The uncertainty estimated on average 15% with a mean ratio 0.96 $\pm$ 0.16



W V

- 193 dipole electric lines between 390 – 2190 Å [Kramida, A. et all, 2024]
- For $5d^2$-5d6p and 5d6s-5d6p transitions :
- $\Delta gf = |gf_{HFR+CPOL} - gf_{MCDHF}|$ = 21% (<30%) [Yoca, S.E et all, 2012]

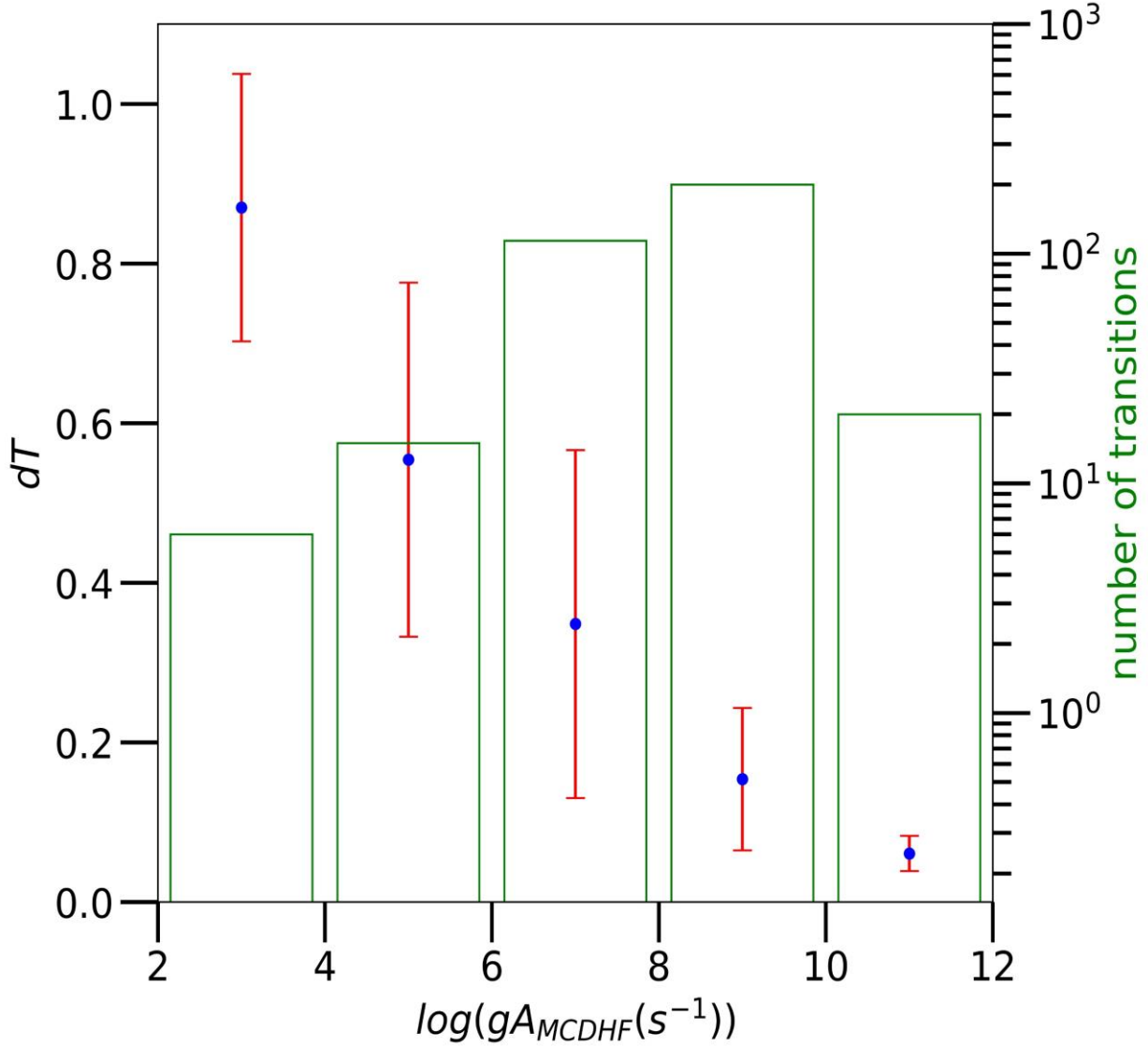


Ta IV

- All 355 transitions computed in the MCDHF method

⇒ 251 transitions have $dT < 0.25$

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



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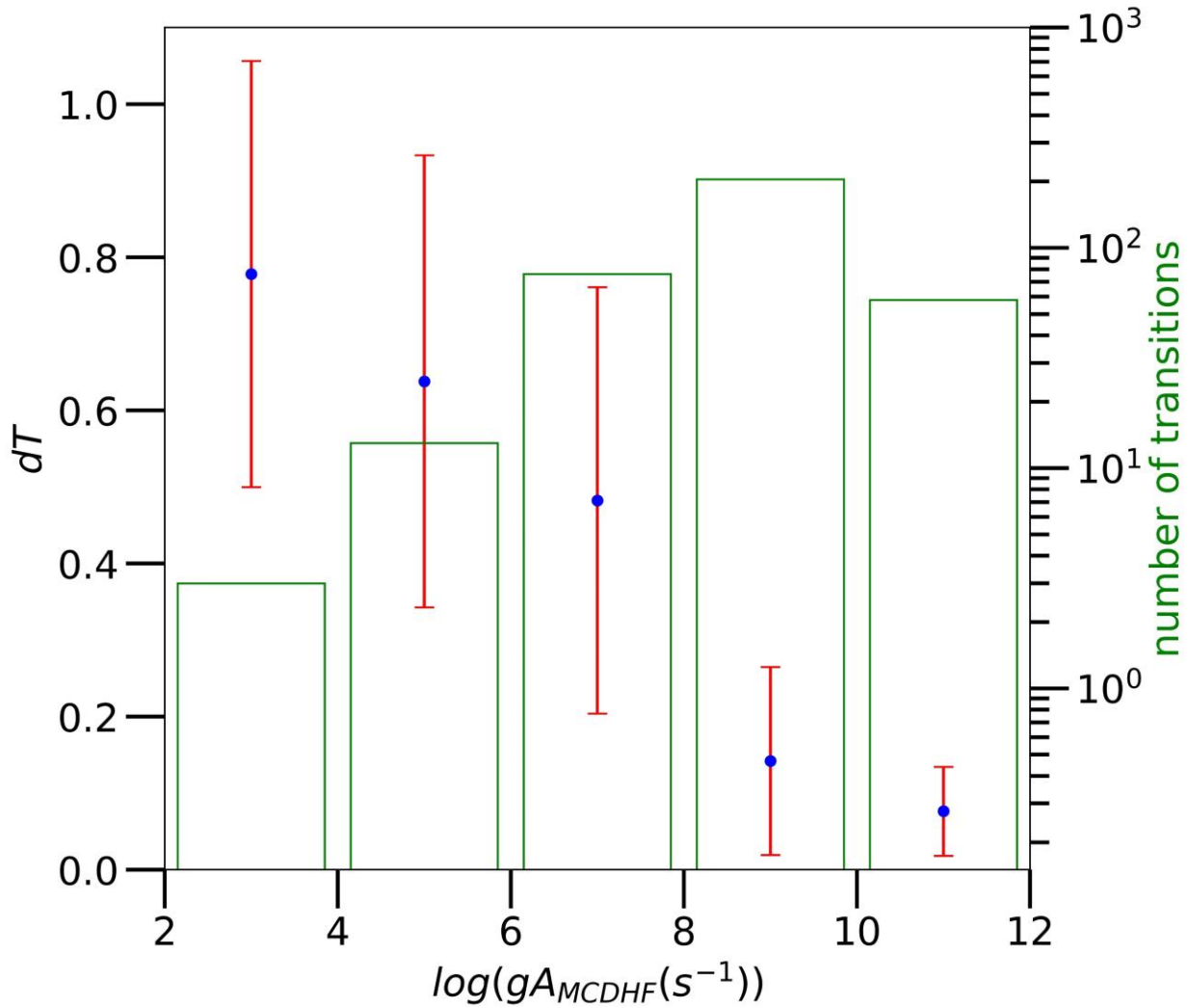

Conclusion

- 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 orbitals begin to overlap the valence configurations → Introduce explicitly these configurations ?!
- Good agreement between HFR+CPOL and MCDHF method and comparisons (170 transitions for TaIV and 101 for ReVI) allow to estimate uncertainty (TaIV:26%; ReVI:15%)
  - ➢ set of new 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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