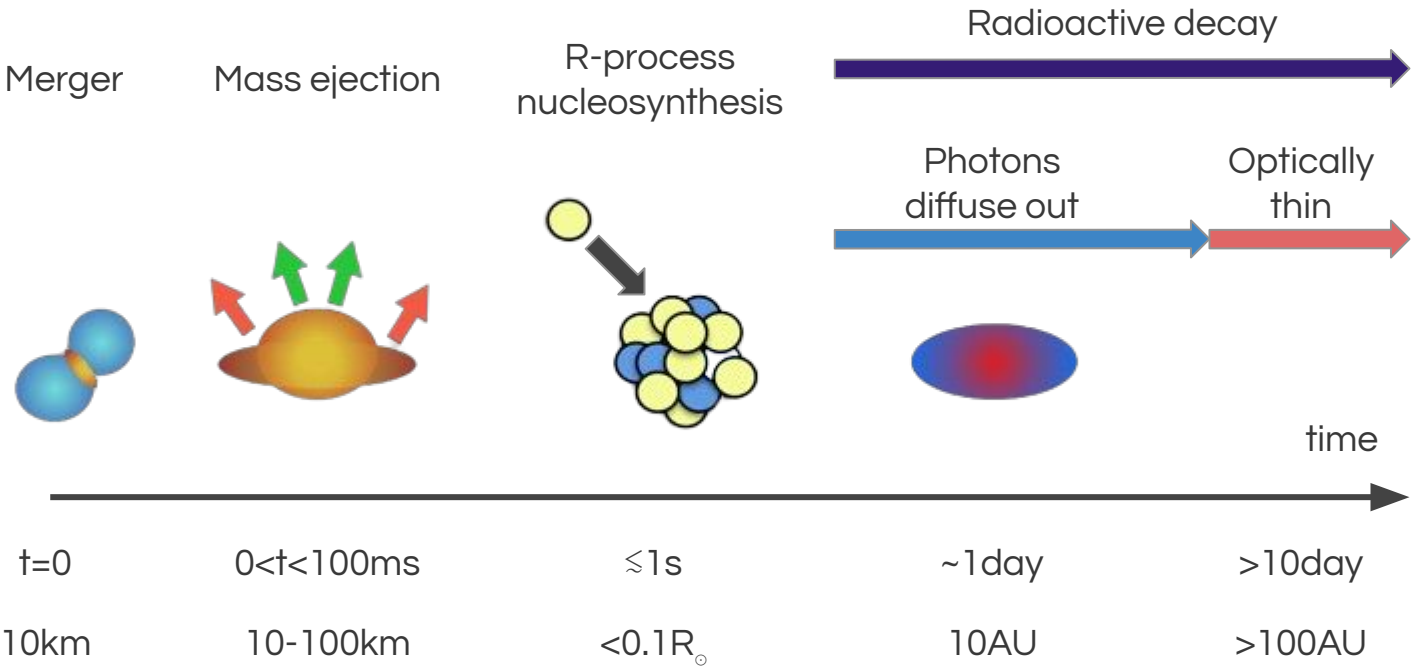
Atomic data requirements for Non-LTE modelling of Kilonovae

Ricardo F. Silva

CompAS 25

Lund, Sweden

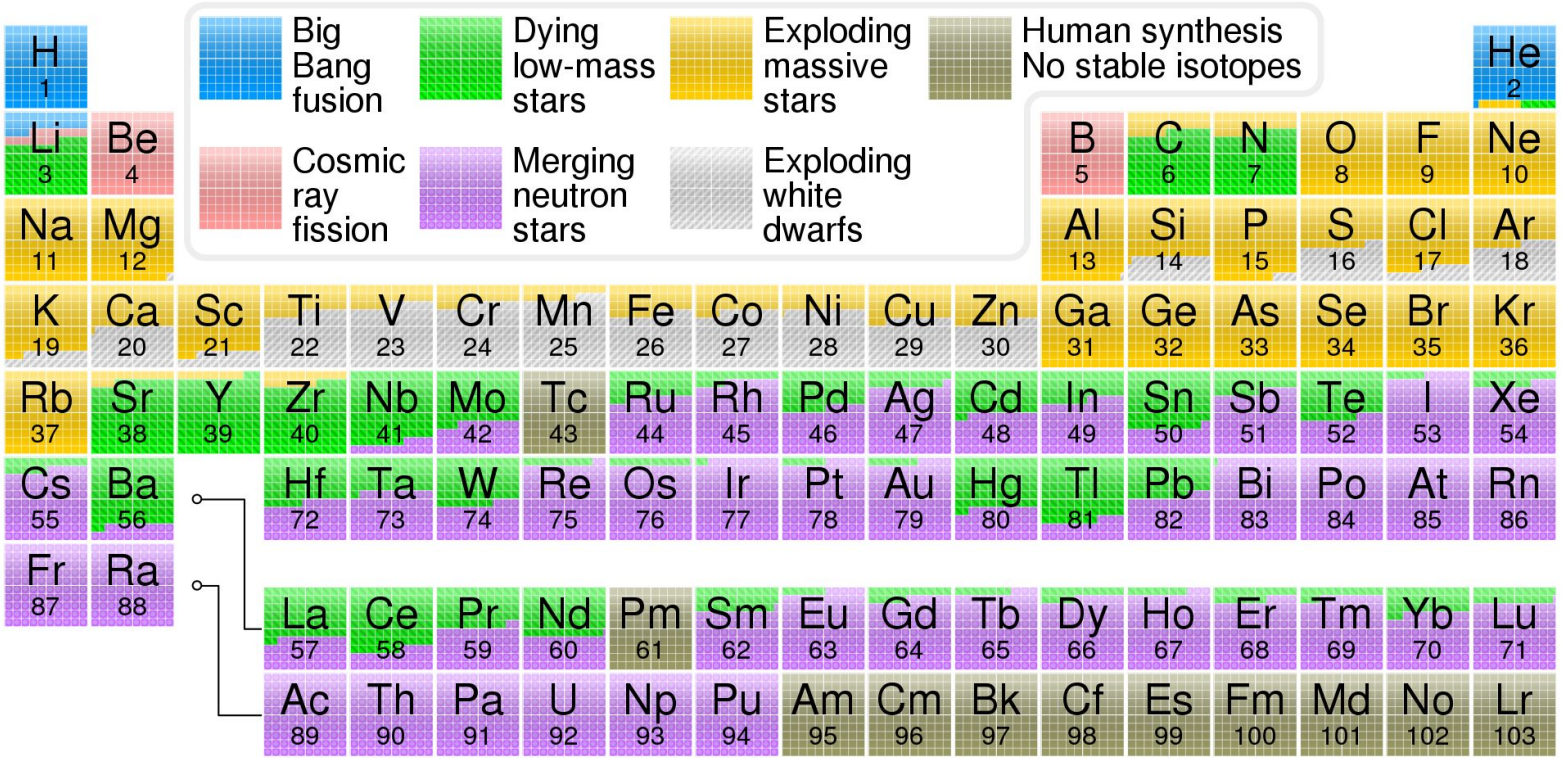
KILONOVA



→ Opacity of the ejecta dominated by photon absorption by **atomic lines**

→ Kilonova light curve and spectra modeling strongly depends on **atomic opacities**

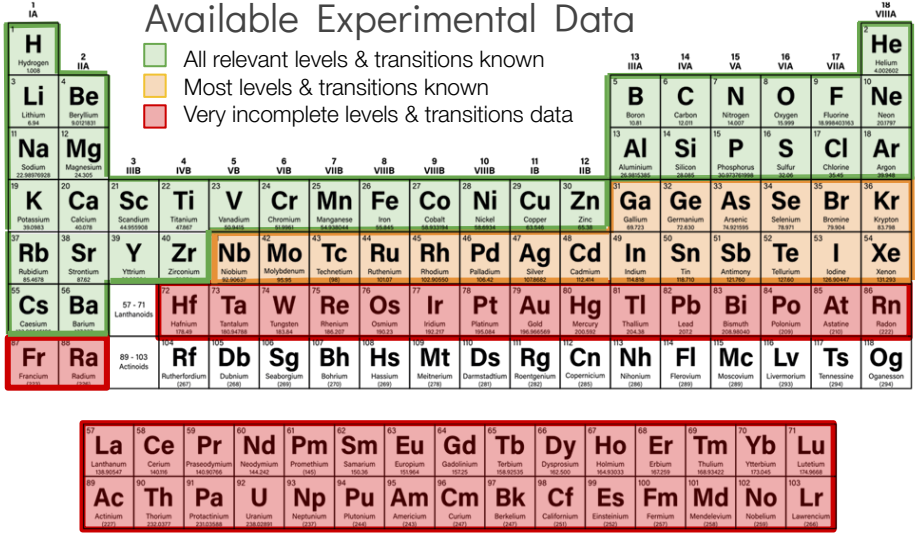
Nucleosynthesis sites



ATOMIC DATA NEEDS

LTE modelling (first few days):
→ **energy levels** and **E1 radiative transitions** required:
Saha & Boltzmann equations
→ bolometric light curves: grey opacities from uncalibrated data good enough
→ spectral models: use of calibrated atomic data essential for line identification and obtaining the relevant spectral features

NLTE modelling (after a few days):
→ requires additional atomic data: **electron-ion impact cross sections, photoionisation & recombination cross sections, forbidden (M1 and E2) transitions**
→ due to lack of atomic data only possible using approximations



Credit: A. Flörs

Atomic data needs

Significant progress on the atomic structure side since 2017!

We now have:

- All relevant energy levels and transitions/opacities for r-process elements
- Data for multiple ionisation stages (neutral to ~4 times ionised)
- Individual important ions investigated in detail
- Calculations from several atomic structure codes
- Most of the data publicly available

Kasen+13, Kasen+17 AUTOSTRUCTURE - Lanthanides
Fontes+ 20, Fontes+ 22 Los Alamos Atomic Physics and Plasma Code - Lanthanides and Actinides (I-IV)
Tanaka +20 , Domoto+22, Banerjee+23, Kato+24 HULLAC - Multiple *r*-process elements (I - IX)
Gaigalas+19, Gaigalas+20, Radžiūtė+20, Radžiūtė+21 GRASP - Multiple lanthanides
Carvajal +22, Deprince+22, Deprince+25 HFR - Multiple Lanthanides and Actinides,
Silva+22, Flörs+23, Silva+25, Flörs+25 GSI + LIP FAC all lanthanides (II, III, IV) calibrated to experimental data
…

Atomic data needs

Energy levels

Recombination rates

Central potential → Wavefunctions

Calibration

Radiative rates (M1, E2 transitions)

Radiative rates (E1 transitions)

Electron-impact excitation rates

Detail in the modelling

Atomic codes

General use codes - multiple atomic processes

- Usually user-input dependent parameters
- Able to calculate a large number of processes
- Limited accuracy
- Fast and efficient
  - 100 000+ levels and transitions in hours/days
- E.g. - **FAC**, Hullac, **Autostructure** , Los Alamos Suite, JAC …

High accuracy structure codes

- Fully *ab-initio* using MC(D)HF or MBPT approaches
- Focused on structure and some radiative properties
- High accuracy
- Computationally demanding
  - Can take months for large scale calculations depending on the ion
- E.g. - GRASP*, ATSP*, MCDFGME*, AMBiT, CI-MBPT…

*Can be (usually) coupled to R-matrix codes for computation of other properties

Method - FAC

- Allows for a complete set of data for plasma modelling with speed and utility in mind
  - ➢ Structure, radiative and collisional processes

- Uses a Dirac-Fock-Slater Hamiltonian with a local central potential, computed for a fictitious mean configuration (FMC) with fractional occupation numbers

  - ➢ Orthogonality is ensured automatically → Speed increase

  - ➢ Potential not optimized for a single configuration → Accuracy issues

  - ➢ Choice of FMC is mostly arbitrary and usually constructed by hand → Major source of uncertainty

FAC (CURRENT IMPLEMENTATION)

Badnel (ASOS 23), Gu 23, private com.

FAC takes a DHS like potential

$$V(r) = \sum_{\beta} \omega_\beta Y^0_{\beta\beta}(r) - a\frac{3}{2}\left[\frac{24}{\pi}\rho(r)\right]^{1/3}$$

$$\rho(r) = \frac{1}{4\pi r^2}\sum_{\alpha} \omega_\alpha \rho_{\alpha\alpha}$$

with $rV(r) \to N$ as $r \to \infty$

$$V(r) = \min\left(V(r), \frac{N-1}{r}\right)^*$$

Latter (1955)

*differences in recent FAC versions comes from different cut-off formalisms introduced

User input:

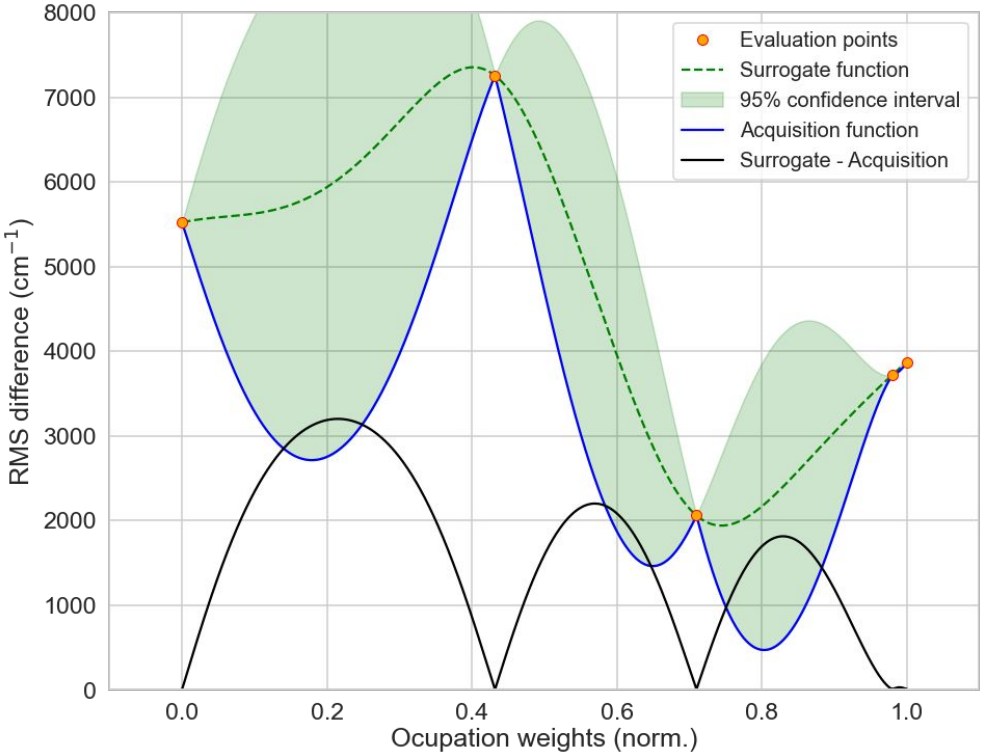
1. User provides a set of configurations (and weights) and the (weighted) mean of the occupation numbers ω_α over all the configurations is taken

2. User provides a set of occupation number ω_α via a fictitious mean configuration (FMC)

Optimization procedure

1. Get a set of initial points
2. **Fit** a surrogate model for a specific loss function
3. Compute **acquisition function** - dynamically chosen between EI, PI and GP-UCB
4. Evaluate new point
5. Repeat 2. - 4. until convergence of loss function evaluation (exploitation) or chosen number of iterations (exploration)
6. **Make recommendation**

Sequential Model-Based Optimization (SMBO)



Example optimization for 1 parameter (4f)
(5d, 6s, 6p) fixed at (0.357,0.0714,0.0714)

Optimization procedure

1. Get a set of initial points
2. **Fit** a surrogate model for a specific loss function
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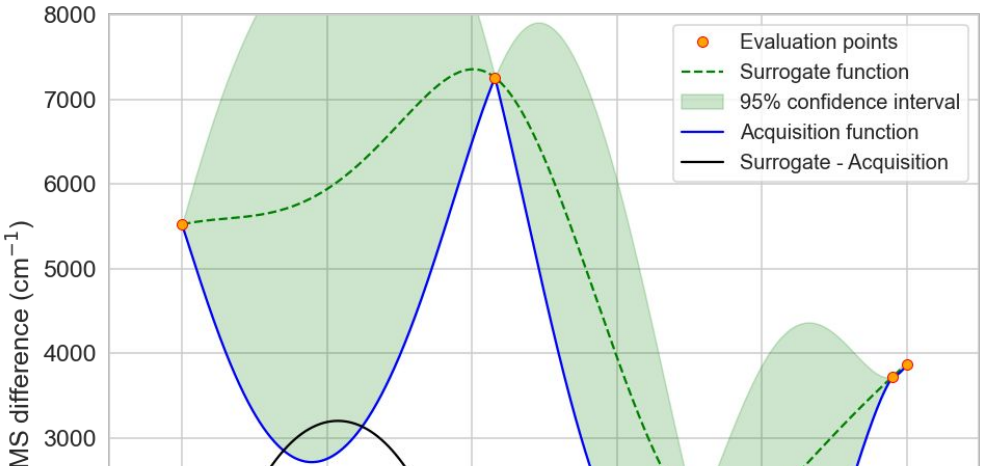

Systematic Bayesian Optimization for Atomic Structure Calculations of Heavy Elements

RFS+25 10.48550/arxiv.2502.13250

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[4] *Institut für Kernphysik (Theoriezentrum), Technische Universität Darmstadt*
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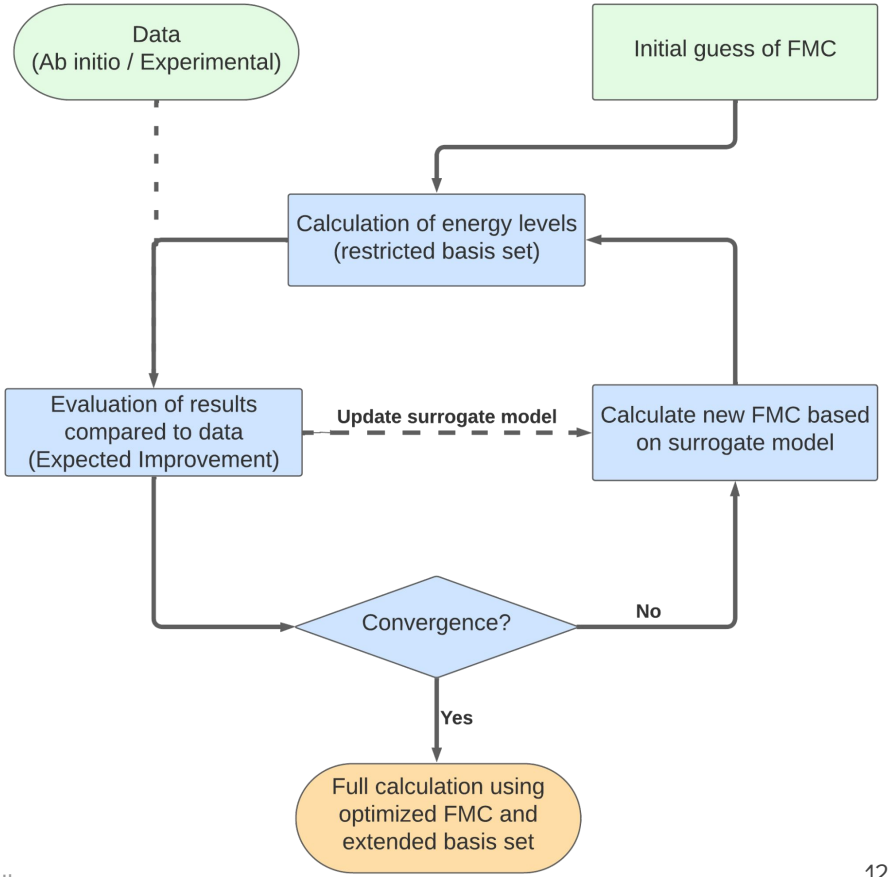
Potential Optimization workflow

- **General** optimization procedure can be applied to multiple codes requiring direct user input for determining local central potential
  - Sequential Model-Based Optimization (SMBO) procedure applied to FAC- RFS+ 25 (10.48550/ARXIV.2502.13250)
  - Similar application had been done already in AUTOSTRUCTURE (M. Mendez PhD Thesis (2021); RFS+ (in prep.)
  - Similar application done for HULLAC (Kato+24)

- **Flexible** loss function - can be adapted to optimized for different needs
  - Energy levels, transition rates, cross sections…

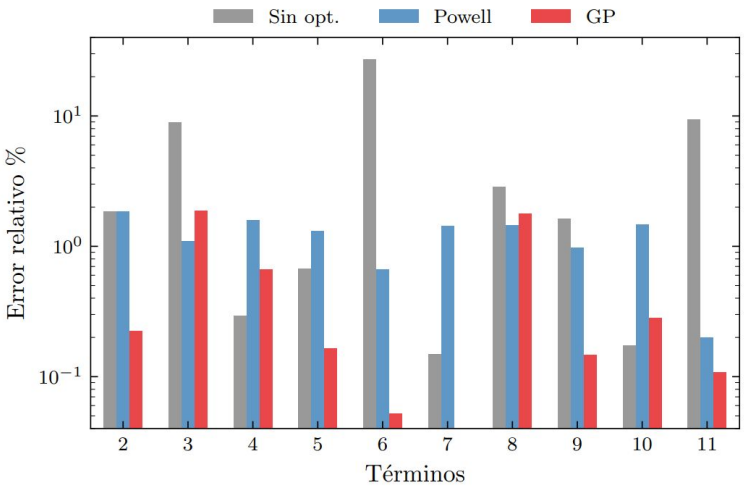


Potential Optimization workflow

M. Mendez PhD Thesis (2021)

i	Término	NIST	Sin opt.	Powell	GP
1	$2s^2\,{}^1S$	0,0000	0,0000	0,0000	0,0000
2	$2s2p\,{}^3P$	0,2003	0,2040	0,1966	0,1998
3	$2s2p\,{}^1P$	0,3879	0,4225	0,3921	0,3952
4	$2s3s\,{}^3S$	0,4746	0,4732	0,4670	0,4714
5	$2s3s\,{}^1S$	0,4983	0,5016	0,4917	0,4991
6	$2p^2\,{}^1D$	0,5184	0,6598	0,5149	0,5181
7	$2s3p\,{}^3P$	0,5368	0,5376	0,5292	0,5368
8	$2p^2\,{}^3P$	0,5440	0,5595	0,5432	0,5537
9	$2s3p\,{}^1P$	0,5485	0,5574	0,5518	0,5493
10	$2s3d\,{}^3D$	0,5655	0,5665	0,5572	0,5639
11	$2s3d\,{}^1D$	0,5871	0,5322	0,5859	0,5877
	Total $2s^2\,{}^1S$	−29,3369	−29,2342	−29,2355	−29,3377

Tabla 4.2: Energía de excitación (en Rydbergs) de los primeros 11 términos espectroscópicos de Be relativos al estado fundamental $2s^2\,{}^1S$.



Atomic data needs

Energy levels

Recombination rates

Central potential → Wavefunctions

Calibration

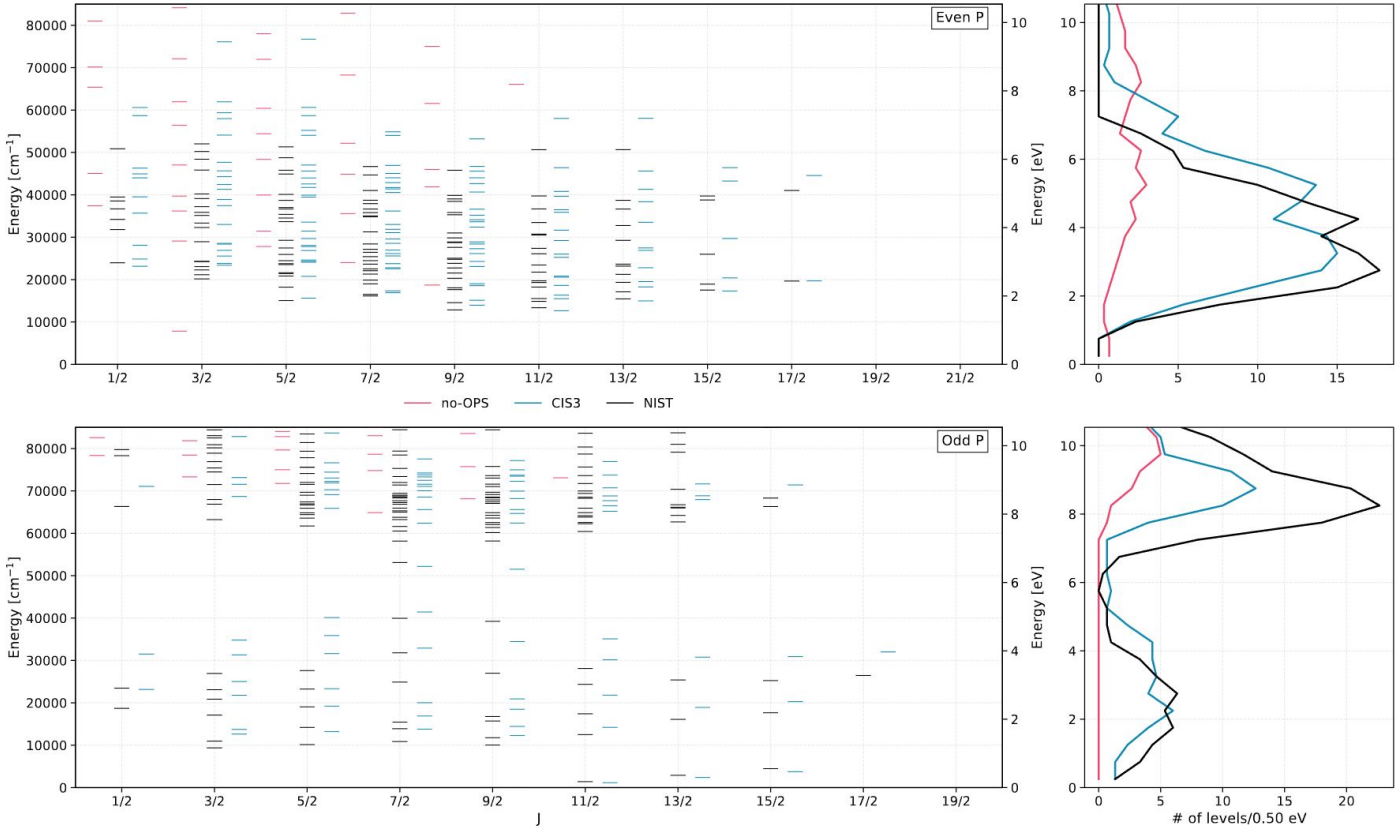
Radiative rates (M1, E2 transitions)

Radiative rates (E1 transitions)

Electron-impact excitation rates

Detail in the modelling

Energy Levels - Lanthanides



- Optimization has the biggest impact in the accuracy of energy levels
- Further calibration (using Term Matching) is then achieved

Calculations for all singly and doubly ionized lanthanides have been achieved

Energy levels of actinides

$$\alpha_{GdII} = 4f^{6.3}5d^{2.08}6s^{0.54}6p^{0.08}$$

$$\alpha^{pred}_{CmII} = 5f^{6.3}6d^{2.08}7s^{0.54}7p^{0.08}$$

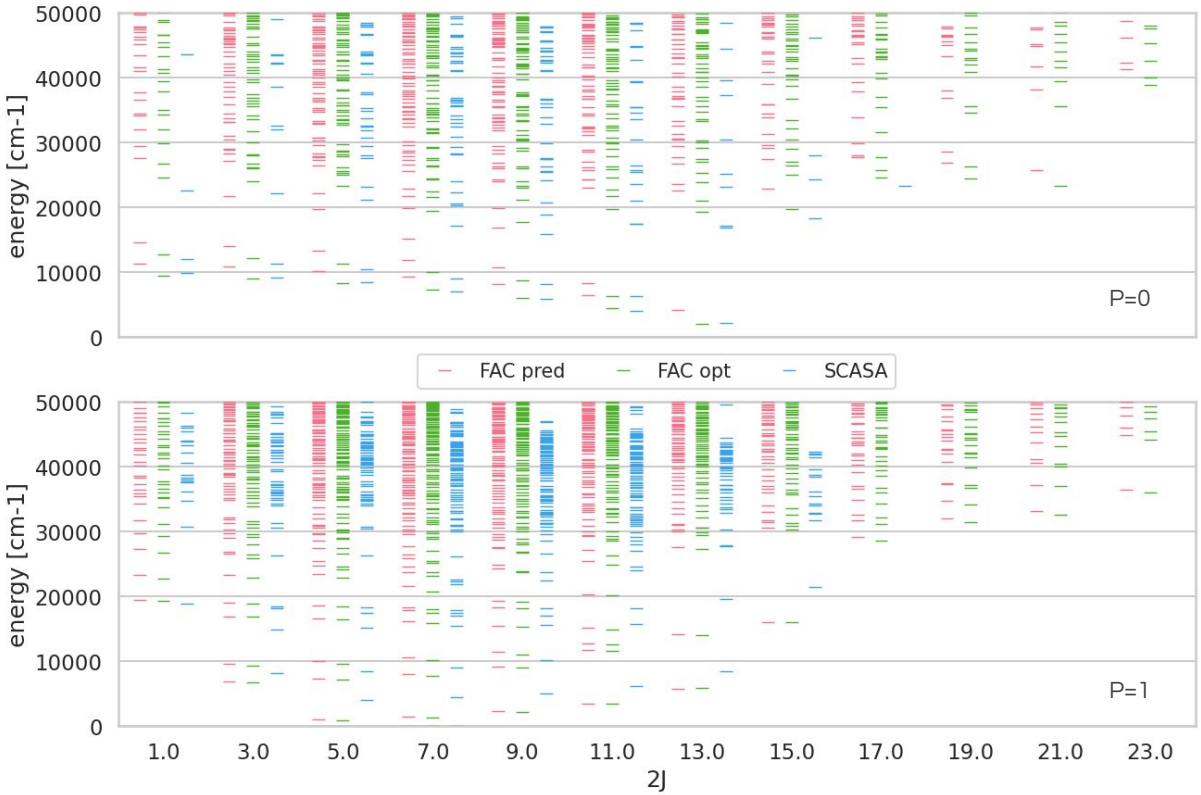
(Δ_{Exp} = 8.4%)

$$\alpha^{opt}_{CmII} = 5f^{6.88}6d^{2.12}7s^{0}7p^{0}$$

(Δ_{Exp} = 7.8%)

- Ability to provide improved data for where no experimental data is available

[R. F. Silva, in prep.]



SCASA - Selected constants, energy levels and atomic spectra of actinides (Blaise and Wyart 1983)

Atomic data needs

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Radiative rates (E1 transitions)

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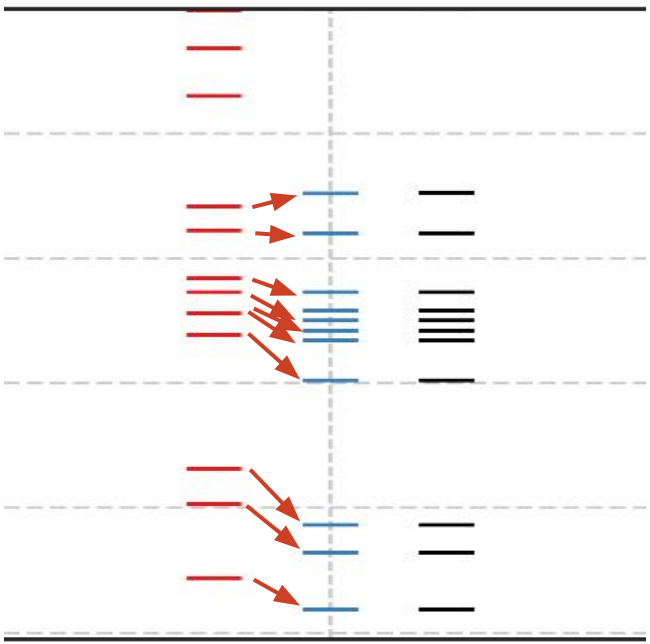
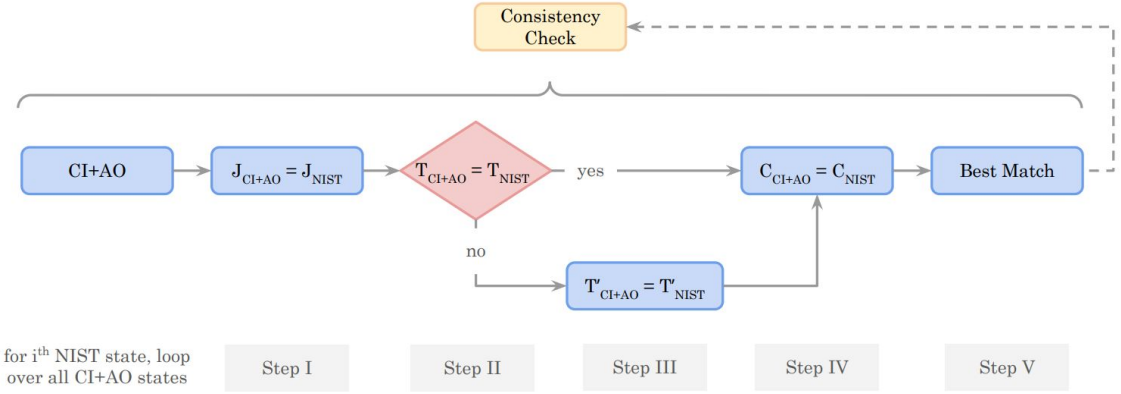
Detail in the modelling

Calibration

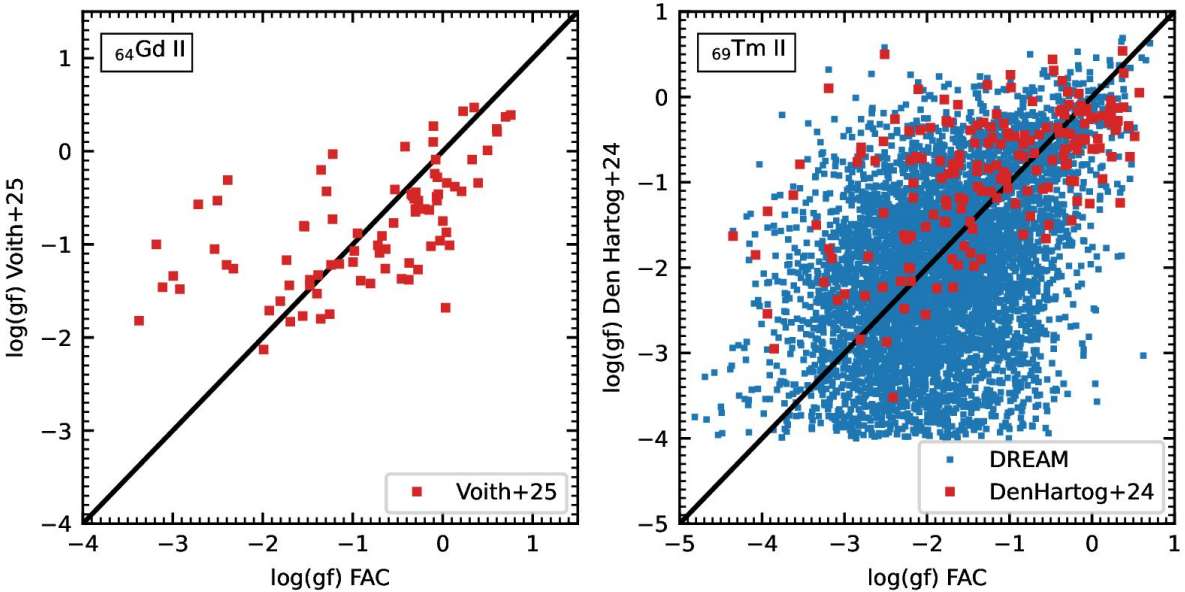


- P, J & spectroscopic term known for FAC & some NIST levels (using the grasp/jj2lsj module by G. Gaigalas)
- Match NIST levels with corresponding FAC levels
- FAC levels without NIST data: use combination of mean correction from P-J group and mean correction from configuration



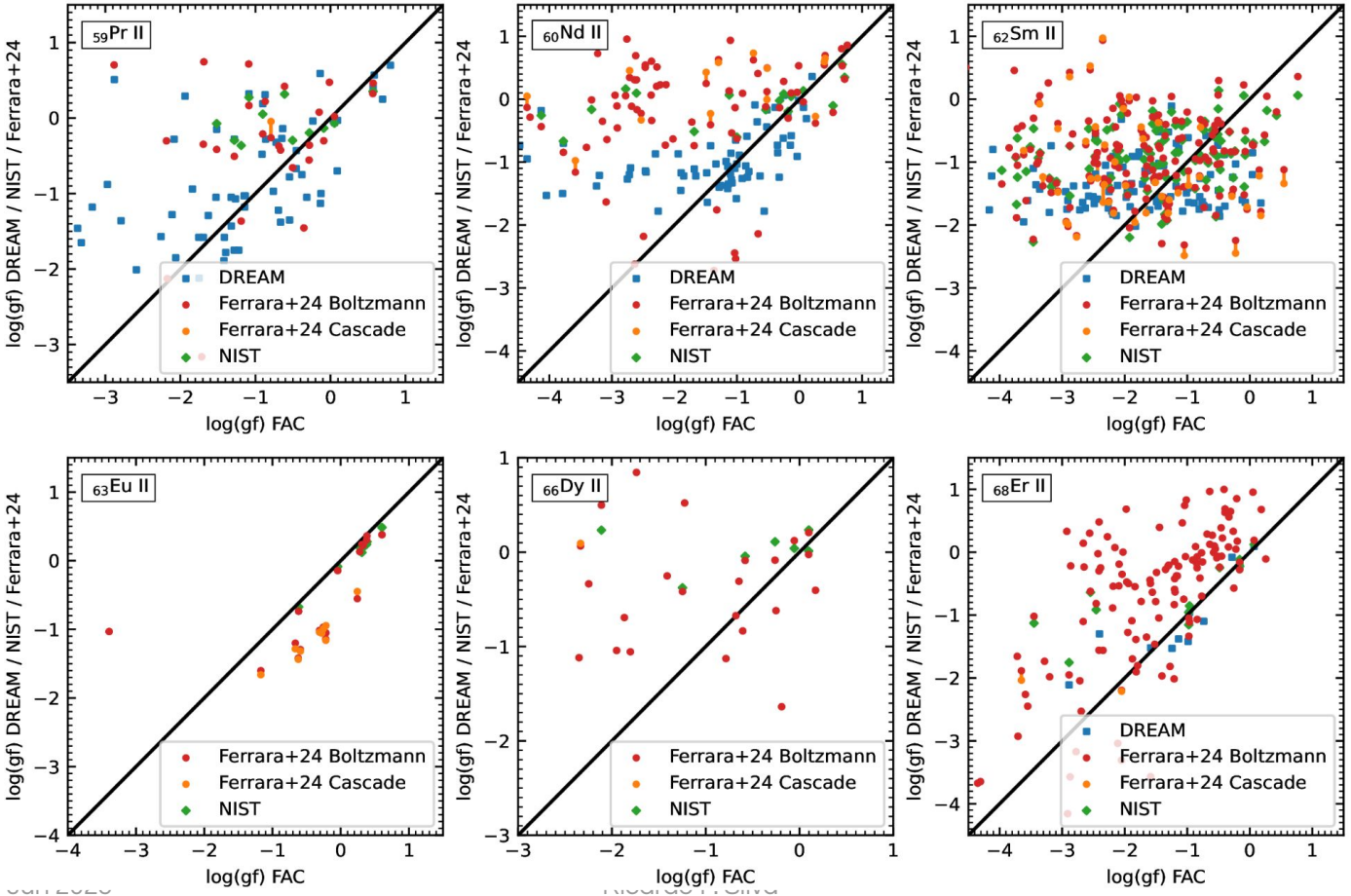
A. Kiruga et al. arXiv:2506.08170

Transition rates

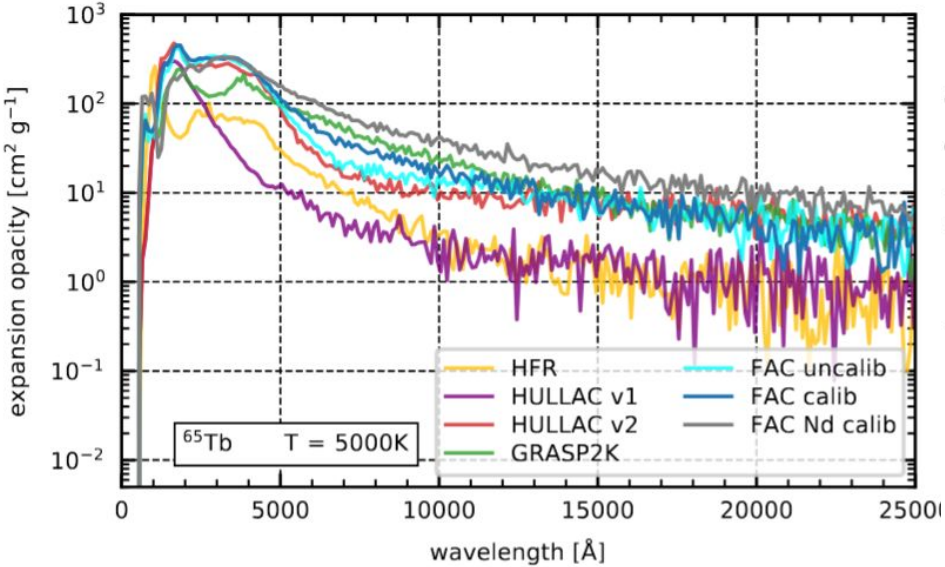


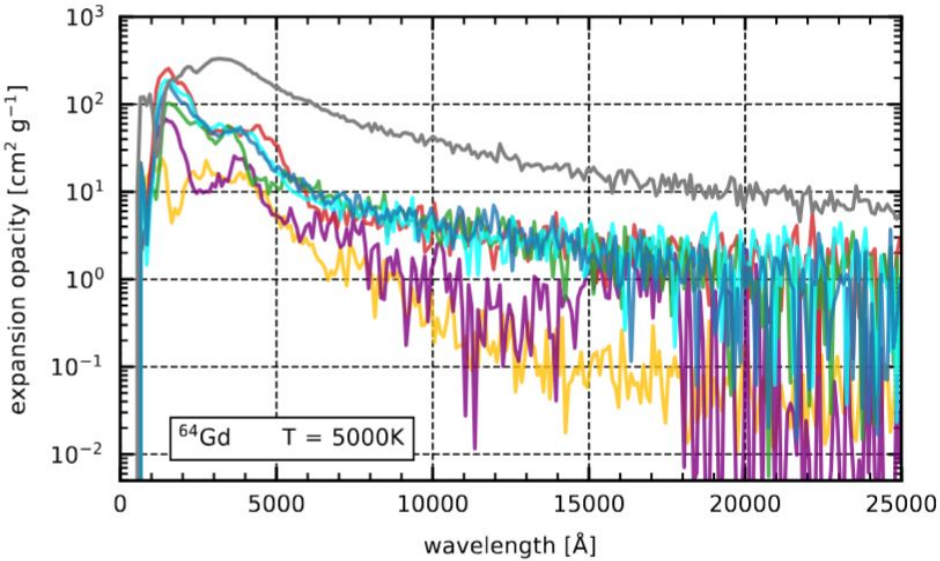
- log(gf) in agreement between calculations, but less so with available experimental data - especially for stronger lines

TRANSITION RATES



Atomic Data for LTE Modelling





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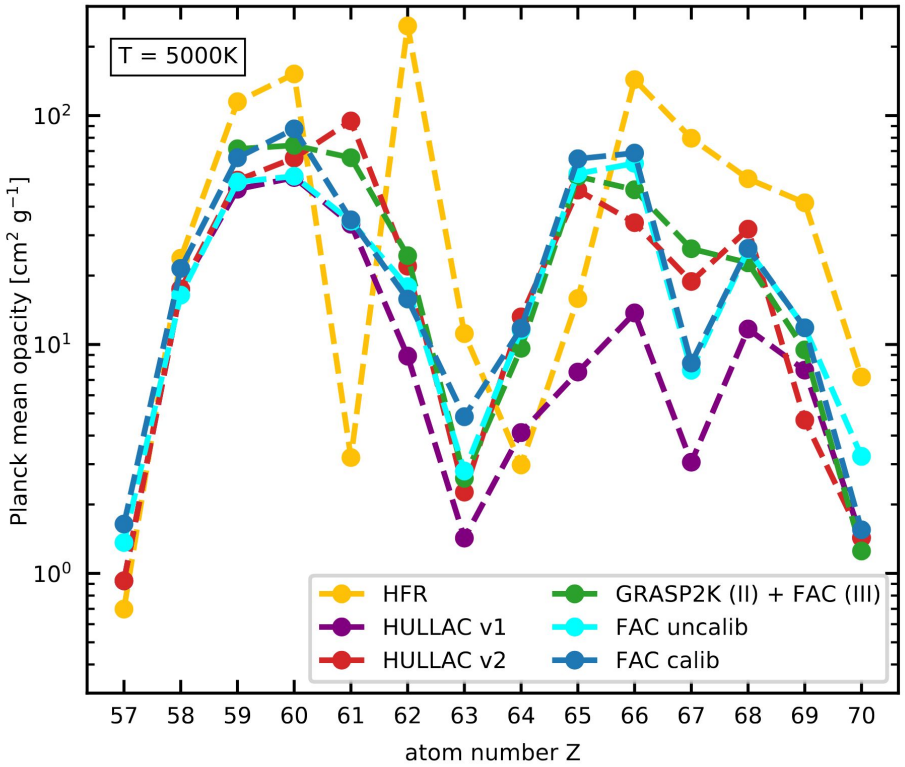

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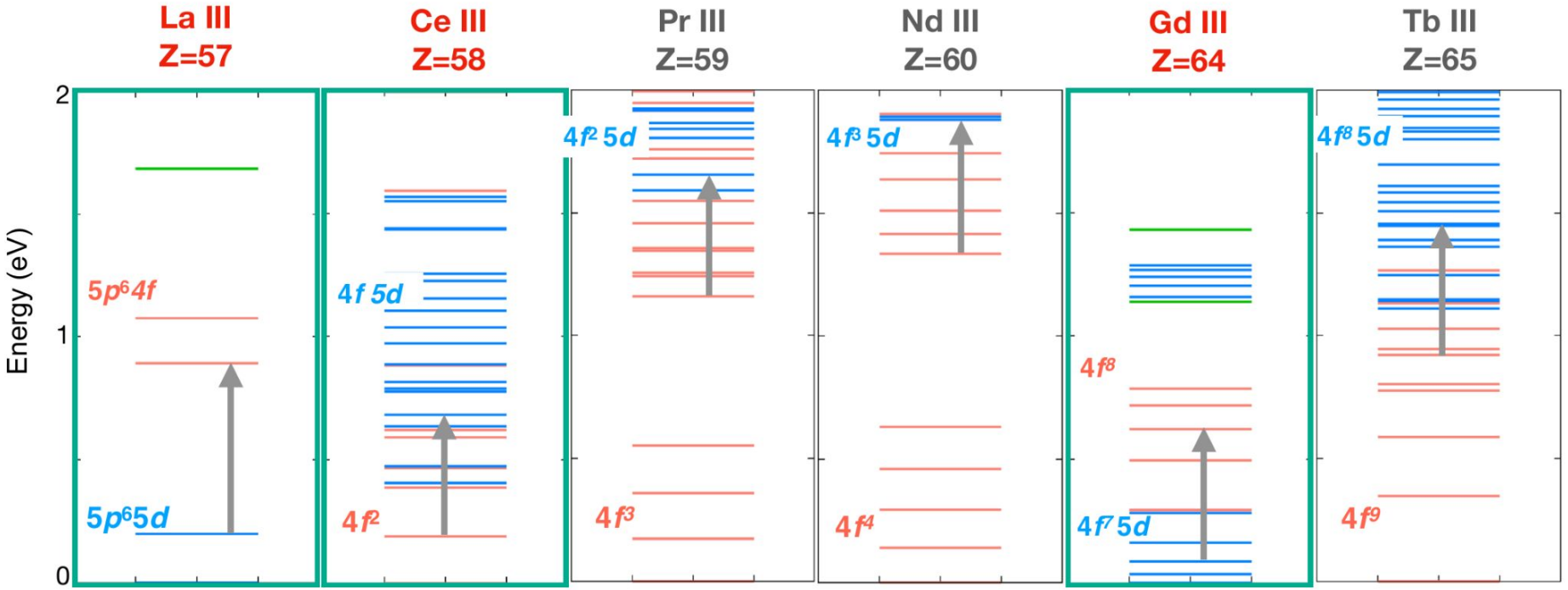
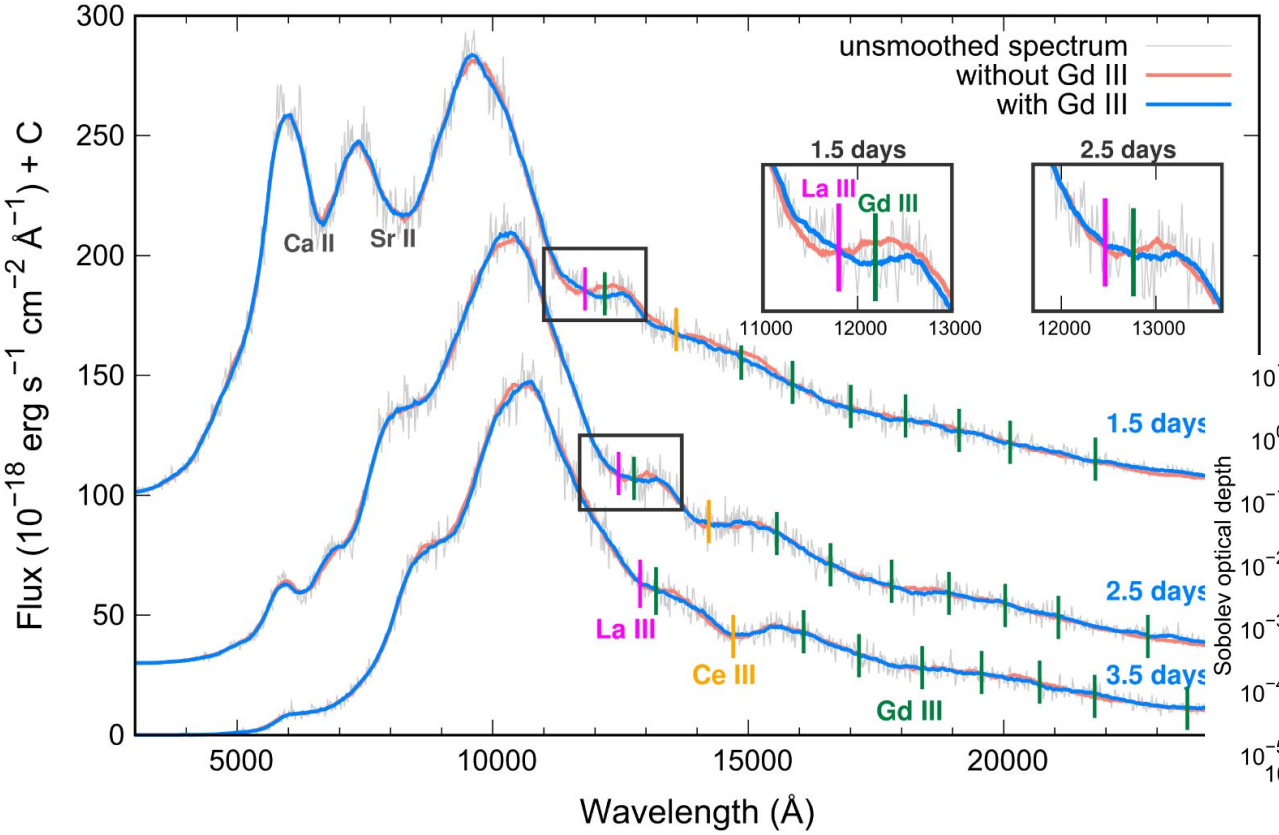


Courtesy of Salma Rahmouni

Atomic Data for LTE Modelling



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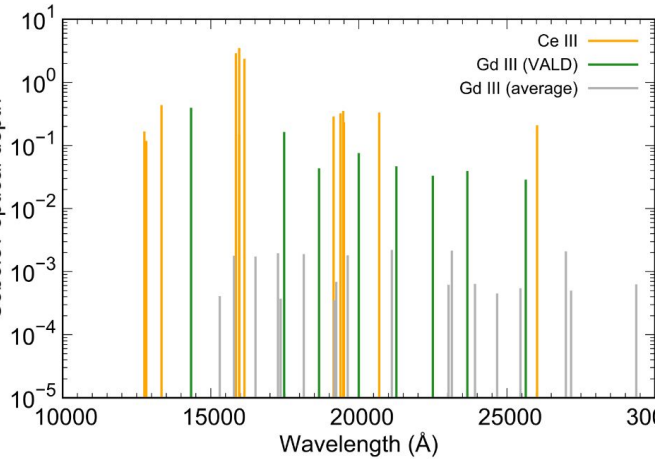


Atomic data needs

Energy levels

Recombination rates

Central potential → Wavefunctions

Calibration

Radiative rates (M1, E2 transitions)

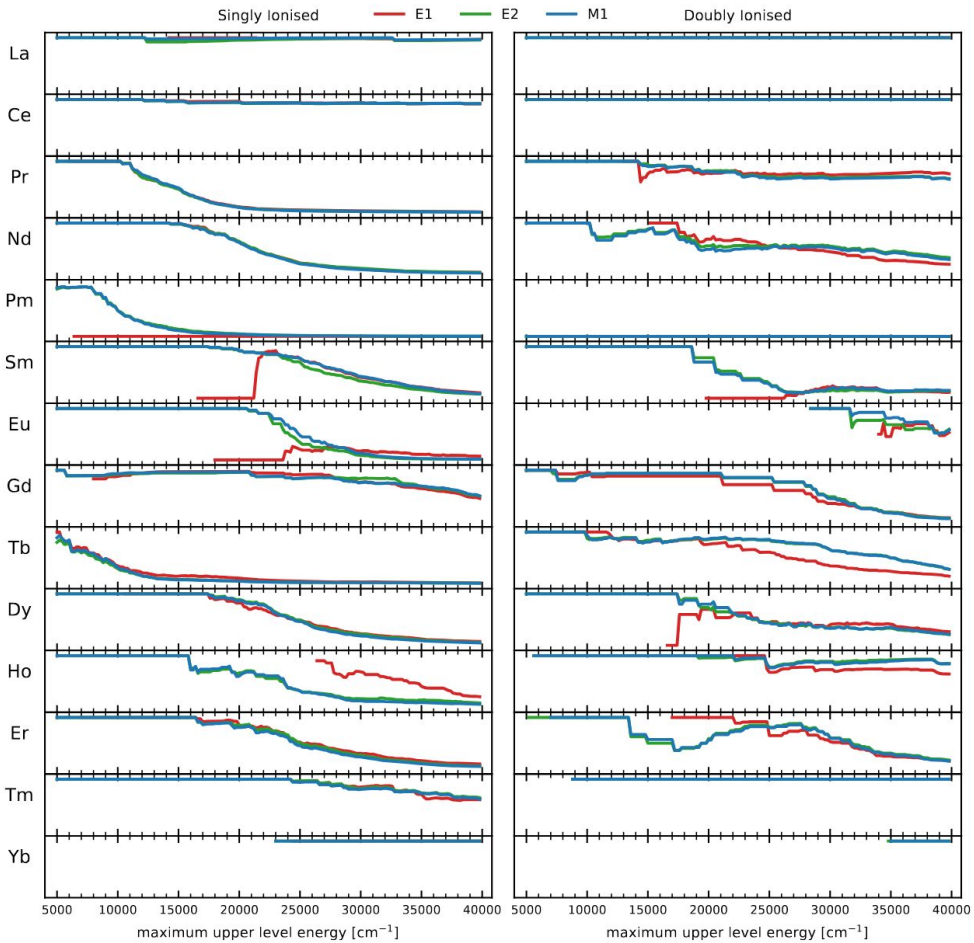
Radiative rates (E1 transitions)

Electron-impact excitation rates

Detail in the modelling

Forbidden Lines

- Doubly ionised lanthanides sufficiently well studied experimentally (exception: Pm III)
- Most permitted & forbidden transitions up to 20 000 - 30 000 cm^{-1} energy calibrated
- Higher number of transitions for singly ionized ions makes the fraction of levels calibrated lower



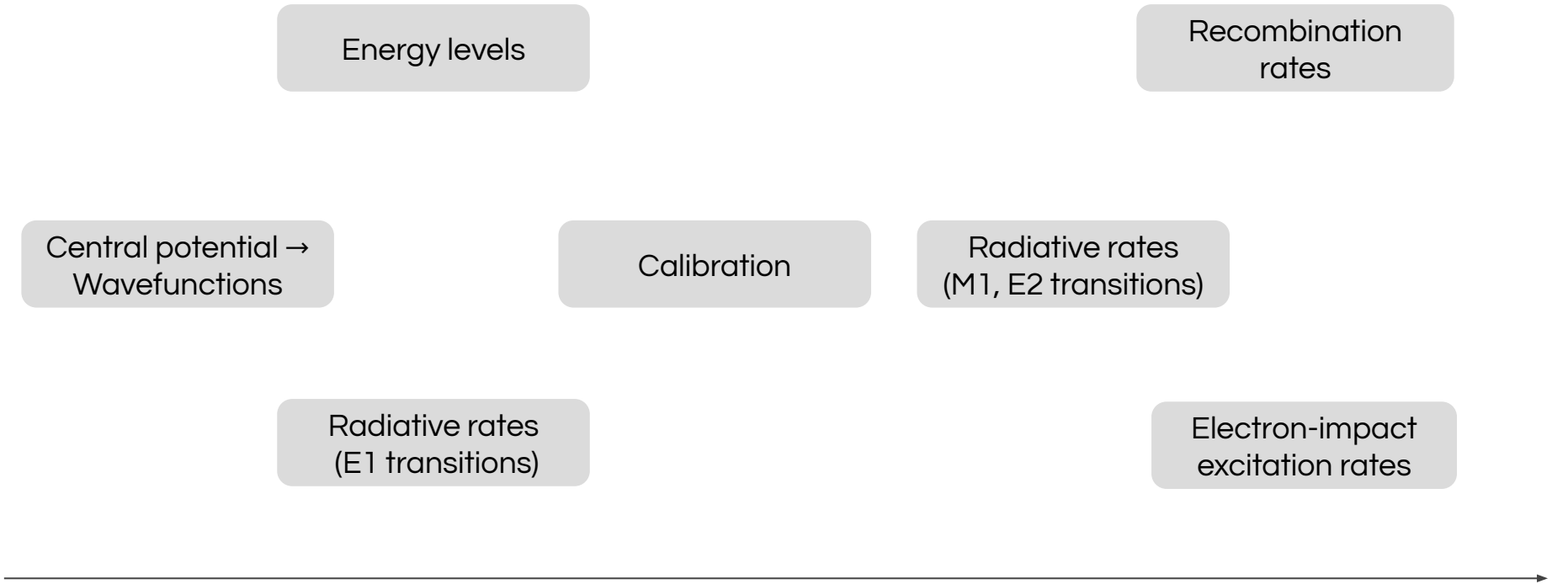
Flörs et al. (in prep.)

Forbidden Lines

Flörs et al. (in prep.)

Ion	FAC calib. below E_{ion} # config	# levels	# E1 lines	# lines E1 calib.	# E2 lines	# lines E2 calib.	# M1 lines	# lines M1 calib.
La II	44	472	17 743	1 239	17 277	1 335	16 551	1 286
La III	23	41	219	219	232	232	125	125
Ce II	40	2 829	408 639	14 592	387 677	20 456	499 746	15 873
Ce III	24	295	7 083	3 680	8 140	4 638	5 736	3 182
Pr II	18	3 689	571 453	881	687 044	1 012	708 795	848
Pr III	15	1 105	63 935	9 436	77 435	12 855	54 138	8 389
Nd II	27	9 994	3 336 077	5 434	3 284 715	6 512	4 100 622	5 215
Nd III	21	4 580	667 955	3675	720 857	4 718	848 074	4 228
Pm II	7	4 990	913 018	0	1 355 037	93	1 303 516	66
Pm III	14	5 526	797 829 s	0	1 046 057	0	1 267 671	0
Sm II	6	6 145	1 111 875	1 106	1 776 621	1 038	1 767 334	1 033
Sm III	16	11 177	1 911 972	95	2 189 469	237	2 835 854	206
Eu II	10	6 781	888 646	224	2 281 580	271	2 430 630	198
Eu III	11	8 470	1 308 810	893	2 218 726	934	2 640 346	821
Gd II	8	12 394	3 070 318	2 909	5 926 233	4 333	6 533 689	3 295
Gd III	10	6 298	814 002	85	1 665 995	94	1 844 924	64
Tb II	8	16 092	5 245 495	445	9 641 783	526	10486 628	433
Tb III	9	8 385	1 335 501	928	1 886 633	1071	2 546 384	806
Dy II	9	12 493	3 580 925	4 617	7 062 482	5 456	7 117 517	4 836
Dy III	6	3 910	355 148	1 246	863 920	1 223	747 452	960
Ho II	8	1 517	86 064	101	141 936	107	125 433	104
Ho III	14	4 658	462 551	1 162	601 531	1 617	696 321	1 149
Er II	16	5 072	838 067	983	1 153 835	1 272	1 416 125	1 016
Er III	18	2 837	195 156	216	234 072	238	238 953	163
Tm II	17	1 517	114 318	7 307	172 834	6 911	152 396	7 406
Tm III	11	650	16 361	1 200	24 834	1 792	18 229	1 461
Yb II	34	348	8 849	3 711	11 223	3 963	11 052	4 414
Yb III	14	916	1 585	202	2 457	347	1 987	264
total		143 181	28 129 594	66 586	45 440 635	83 281	50 416 228	67 841

Atomic data needs

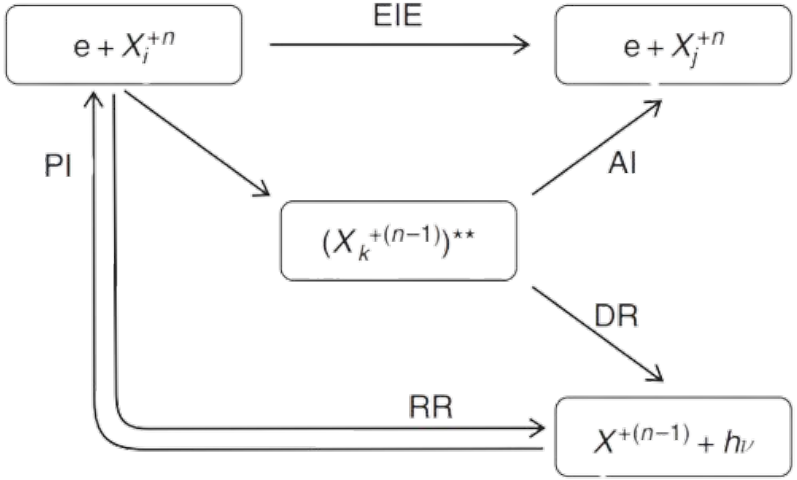


Atomic processes with the continuum

Two main methods:

- Distorted Wave (DW)
  - Neglects interaction between channels (resonances)
  - Fast and efficient
  - FAC, AUTOSTRUCTURE, HULLAC
- R-matrix
  - Resonances are treated consistently
  - Very computationally demanding
  - GRASP^0+DARC

(Pradhan and Nahar, "Atomic Astrophysics and Spectroscopy", Cambridge 2011)



Progress so far…

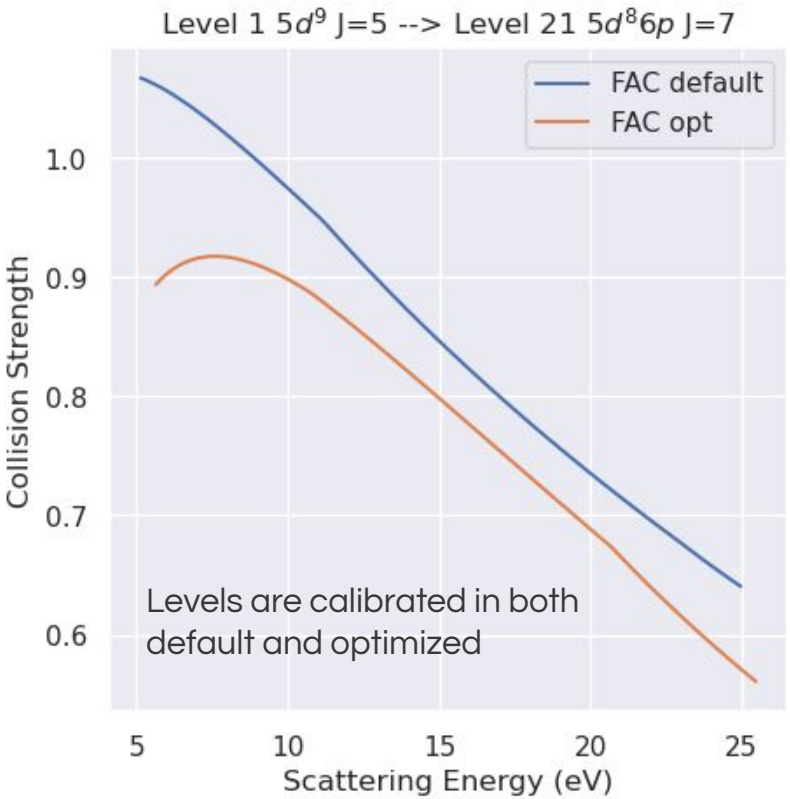
Electron impact excitation:

McCann+21, Bromley+23, Mulholland+24, McCann+25 GRASP0 + DARC - Pt + Au (I- III), Sr II, Y II

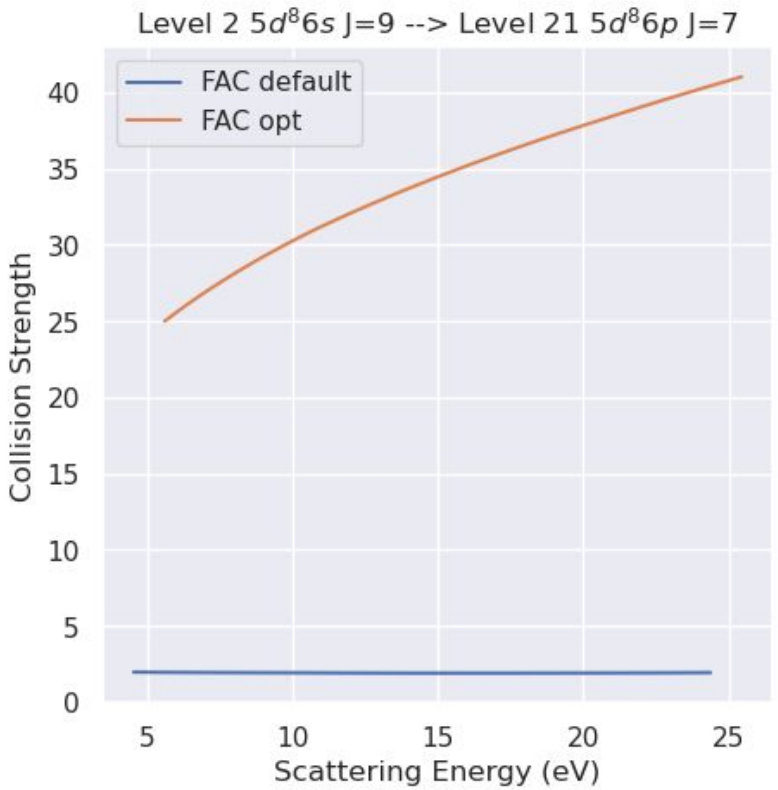
Recombination:

Barnerjee+25 Hullac - Se, Rb, Sr, Y, Zr (I- IV)

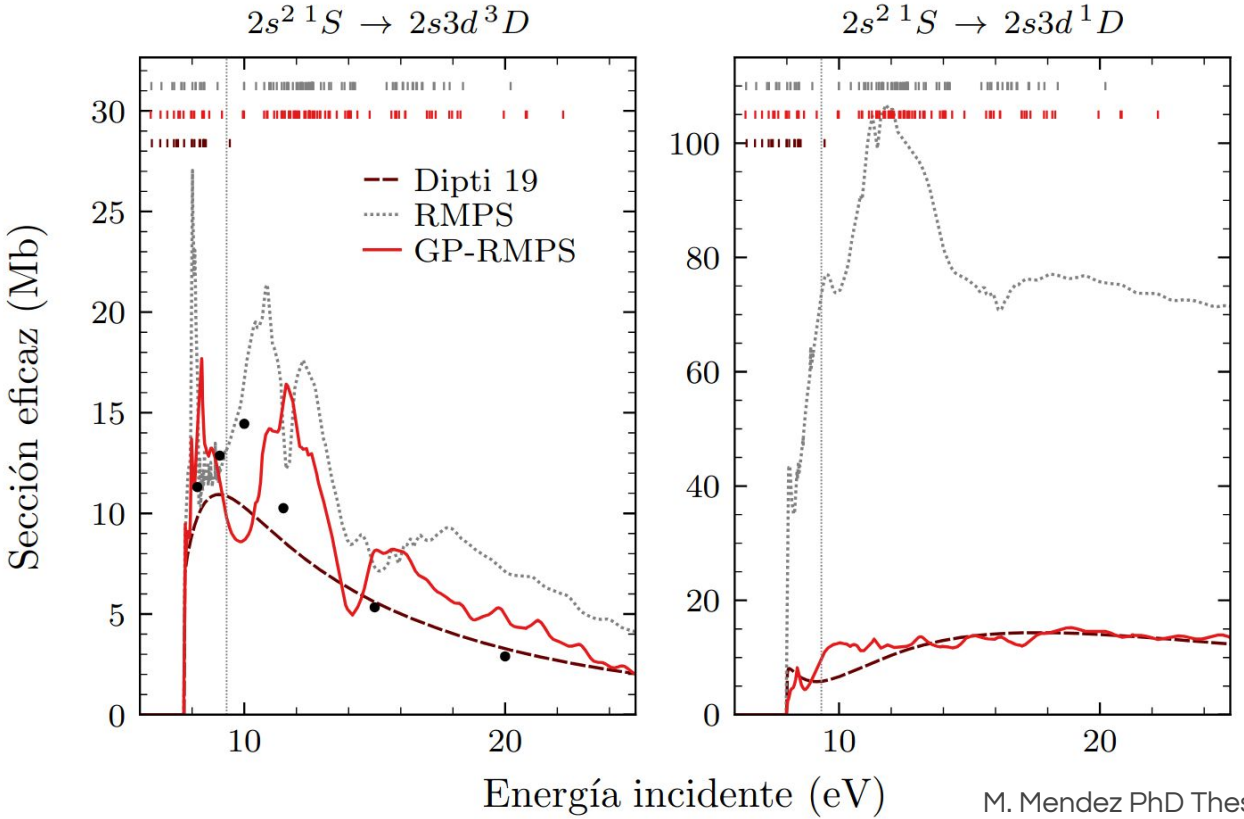
Impact of optimization on EIE



Pt II



Impact of optimization on EIE

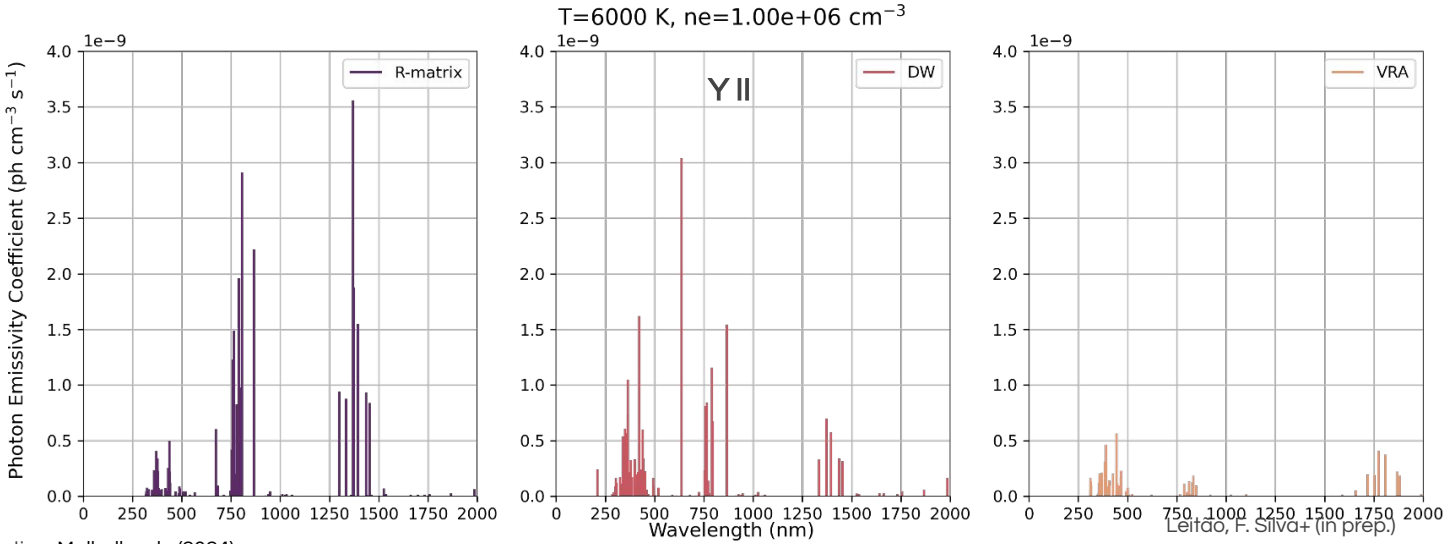


M. Mendez PhD Thesis (2021)

- Similar optimization procedure for improved central potential have shown similar effects on collisions strengths

Improving EIE collision rates

$$\mathrm{PEC}_{j\to i} = \frac{N_j A_{j\to i}}{n_\mathrm{e}}$$



R-matrix calculation: Mulholland+ (2024)

DW seems to provide significant improvements to the current status, based on empirical formulas (VRA):

→ Van Regemorter for allowed, $f_{osc} \geq 10^{-3}$

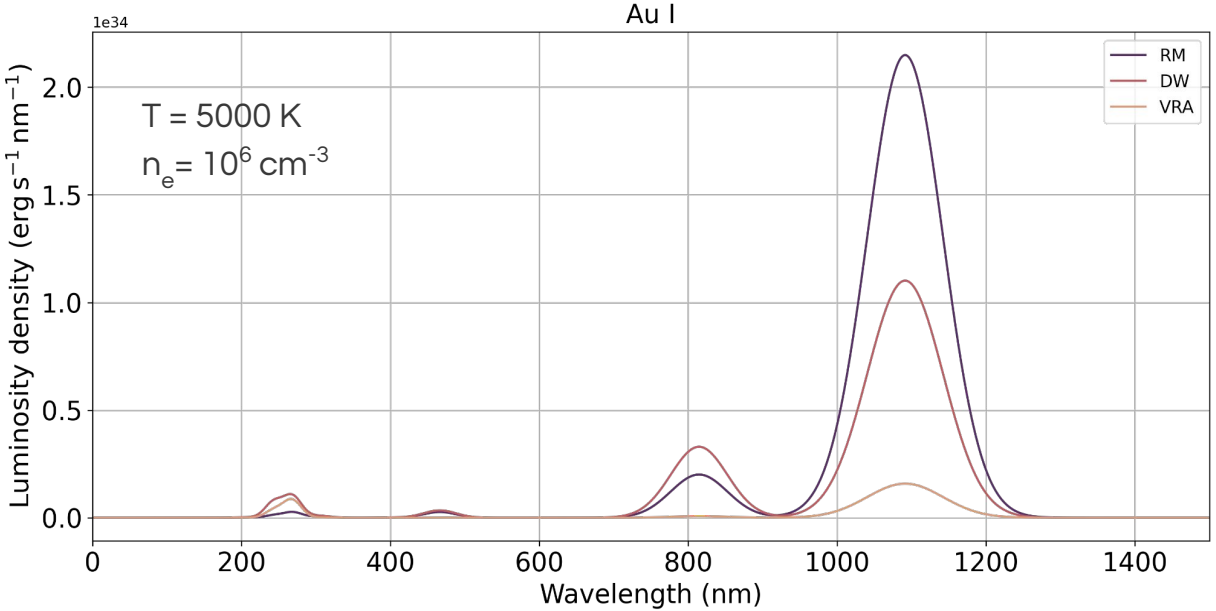
→ Axelrod 1980 for forbidden, $f_{osc} < 10^{-3}$

Empirical approximations produce PECs (“line intensities”) much lower than current calculations

DW provide a much better estimate at a very low computational cost

Improving EIE collision rates

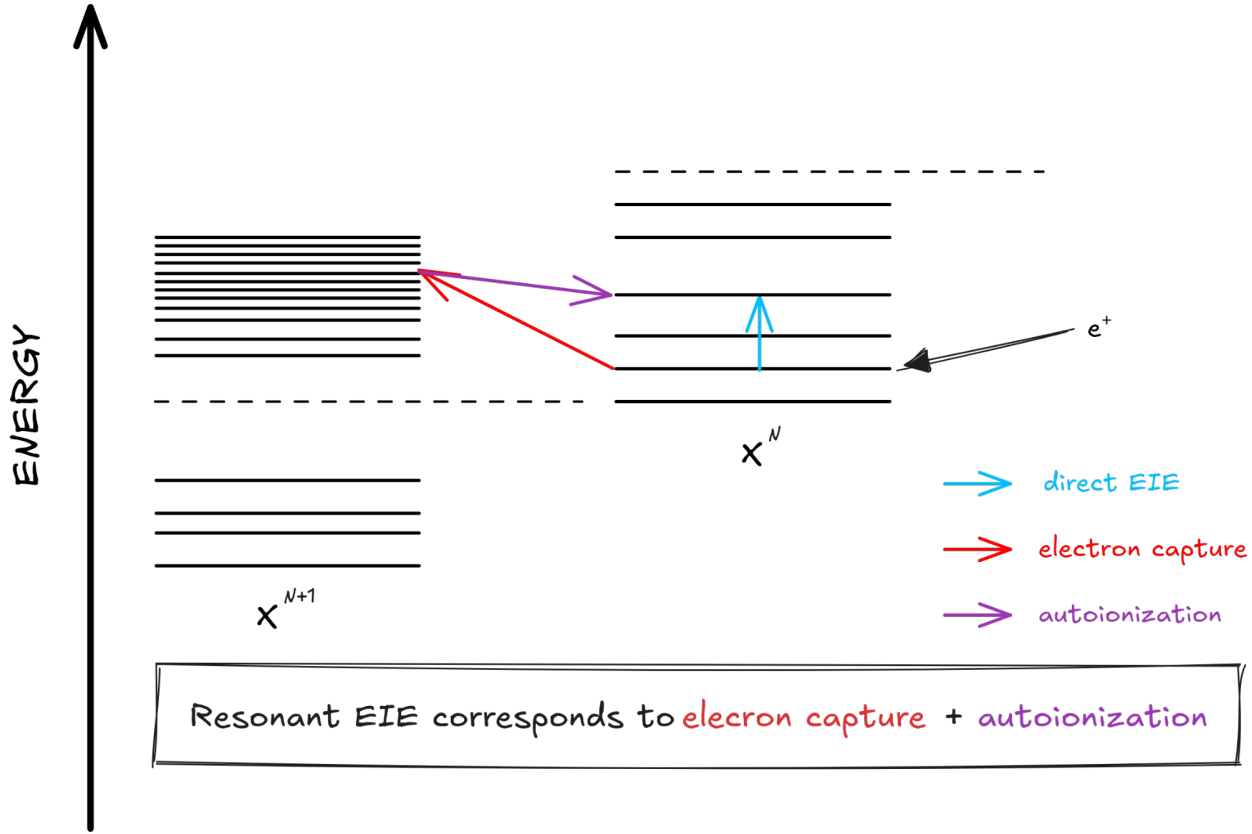
$$L_{j\to i} = \frac{hc}{\lambda_{j\to i}} \frac{n_e \mathrm{PEC}_{j\to i}}{\sum_i N_i} \frac{M_{\mathrm{ion}}}{m_{\mathrm{ion}}}$$



R-matrix calculation: McCann+ (2021)

- DW shows an improvement when compared to just using the VRA approximations -even without the inclusion of resonances.
- "Comparable" to R-Matrix - but at a much lower computational cost
- Calculations for multiple lanthanides have been achived

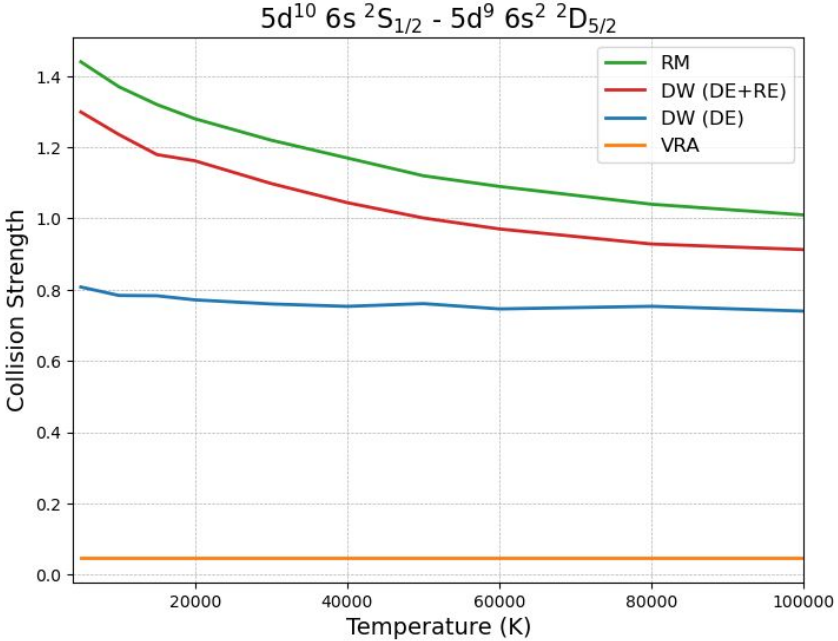
Including Resonances

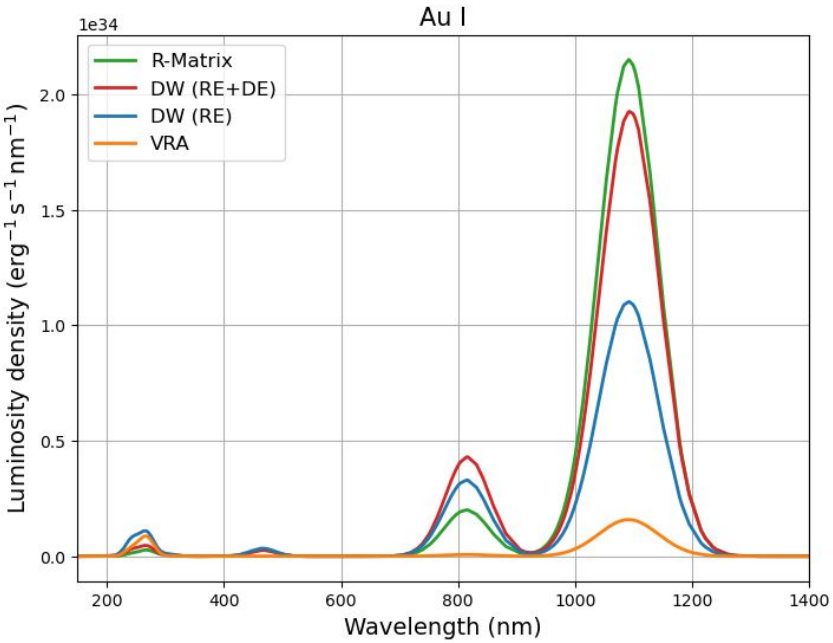


- Resonances can be accounted for in the Independent-Process, Isolated-Resonance Distorted-Wave (IPIRDW) Method (see. e.g. L.Xia et al.. 2017)

- While not as accurate is much more computationally efficient
  - Autoionization and EIE can be computed in parallel

Including resonances (Preliminary)





- Calculation of Resonant EIE for multiple lanthanides ongoing

CONCLUSIONS

- We computed the atomic structure of all singly and doubly ionised lanthanide ions
  → total of 28 ions (27 of them calibrated to experimental data)
  → 120 million (E1 + E2 + M1) transitions, of which 220 000 have calibrated wavelengths

- Inaccurate wavefunctions can have major effects in all atomic data parameters computed
  → Large configurations sets and optimization to available data (when possible) is essential when *ab-initio* calculations are not feasible

- Ongoing calculations of EIE under the DW approximation (DE and RE) for all singly and doubly ionized lanthanides
  → Accuracy of wavefunctions can have a strong impact in collision strengths

COLLABORATION

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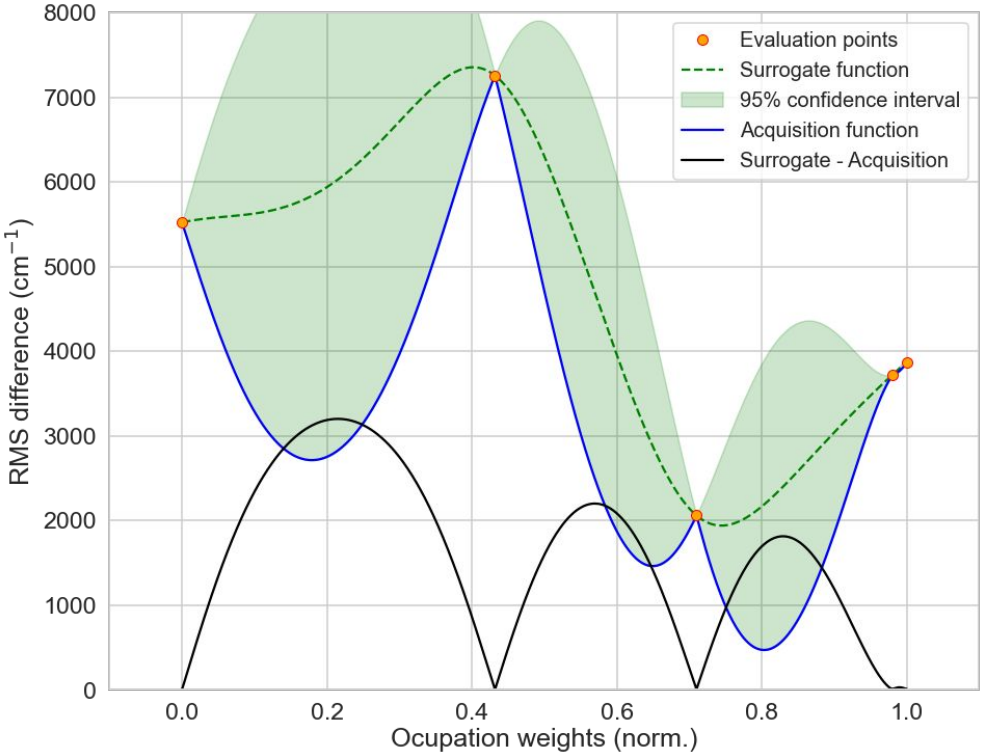
Fundo Europeu de Desenvolvimento Regional

Thank you for your attention!

Optimization procedure

1. Get a set of initial points
2. **Fit** a surrogate model for a specific loss function
3. Compute **acquisition function** - dynamically chosen between EI, PI and GP-UCB
4. Evaluate new point
5. Repeat 2. - 4. until convergence of loss function evaluation (exploitation) or chosen number of iterations (exploration)
6. **Make recommendation**

Sequential Model-Based Optimization (SMBO)



Example optimization for 1 parameter (4f)
(5d, 6s, 6p) fixed at (0.357,0.0714,0.0714)

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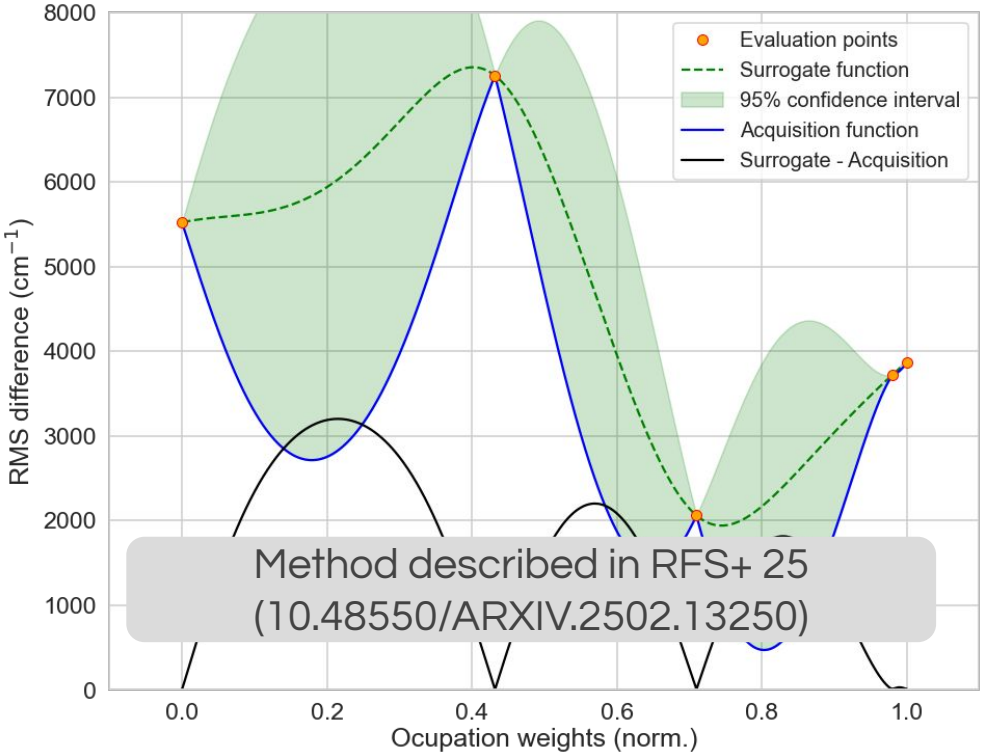
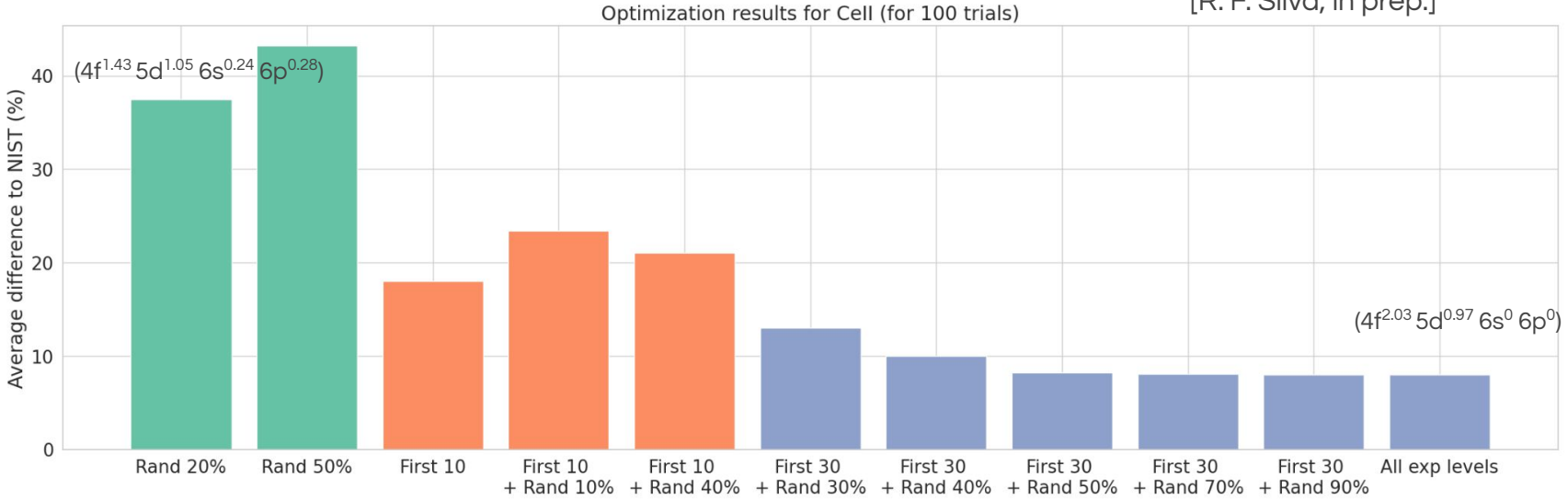


Example optimization for 1 parameter (4f)
(5d, 6s, 6p) fixed at (0.357,0.0714,0.0714)

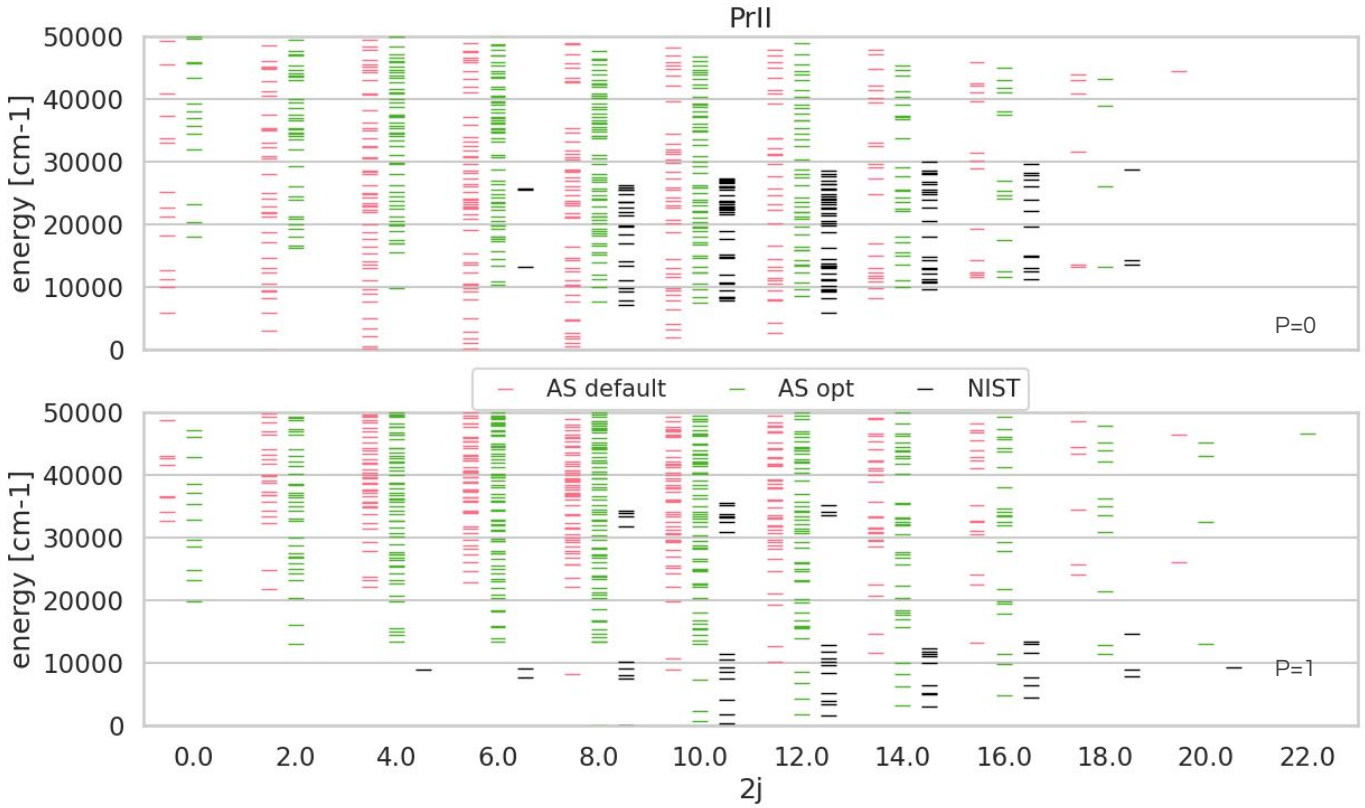
Sensitivity of the Optimization

[R. F. Silva, in prep.]



- Only very small changes to FMC after including more than 50% of the available data for Ce II - maintaining a relative accuracy of ~8%
- Close to ground state levels have the most impact (~10-30 levels)
- Provides confidence on it's predictive value for non-measured levels and robust to low amounts of data

Optimization in AUTOSTRUCTURE

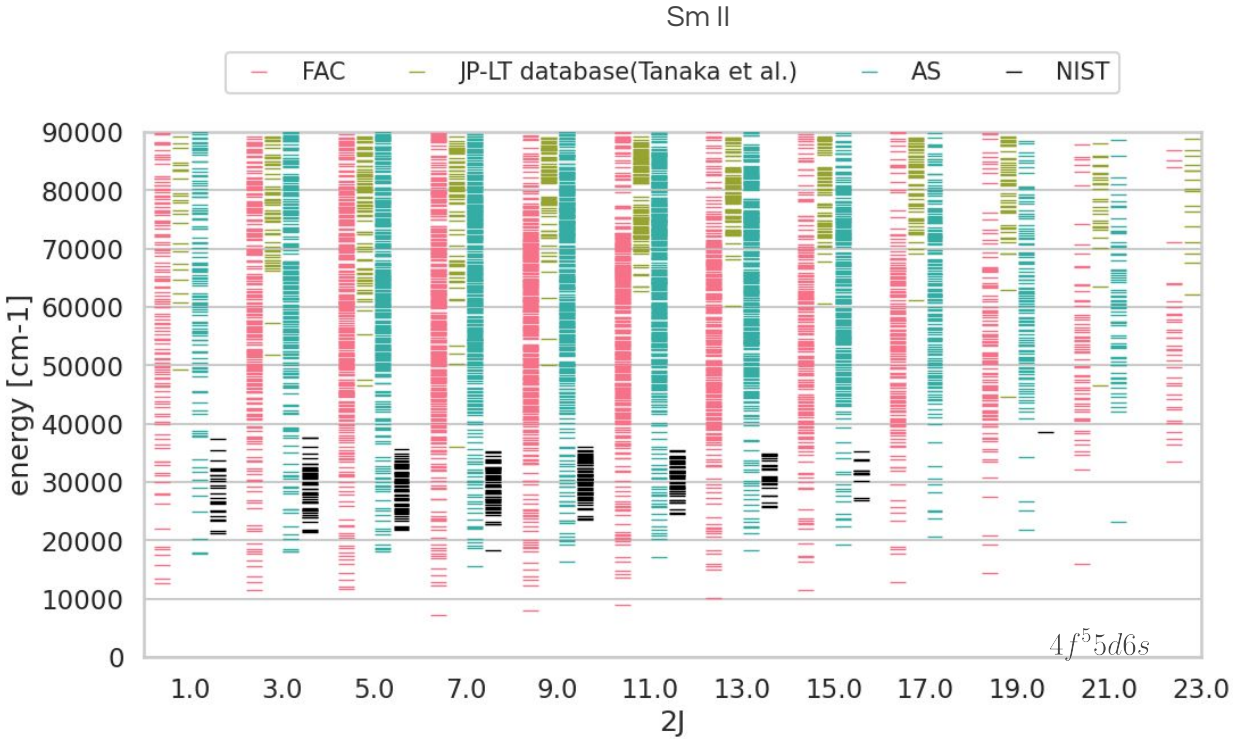


- Faster and better memory handling than FAC
- Not fully relativistic

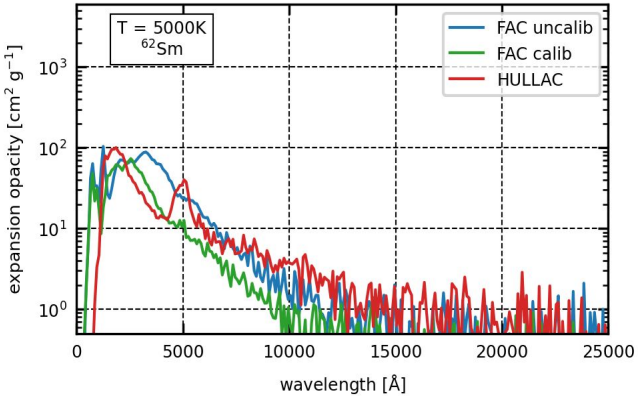
- "AS default" uses one of multiple built in ways to optimized the potential - possible better result if tweaked

- No extra input needed in "AS opt" - optimization in this work

Assessment of atomic data - Energy Levels

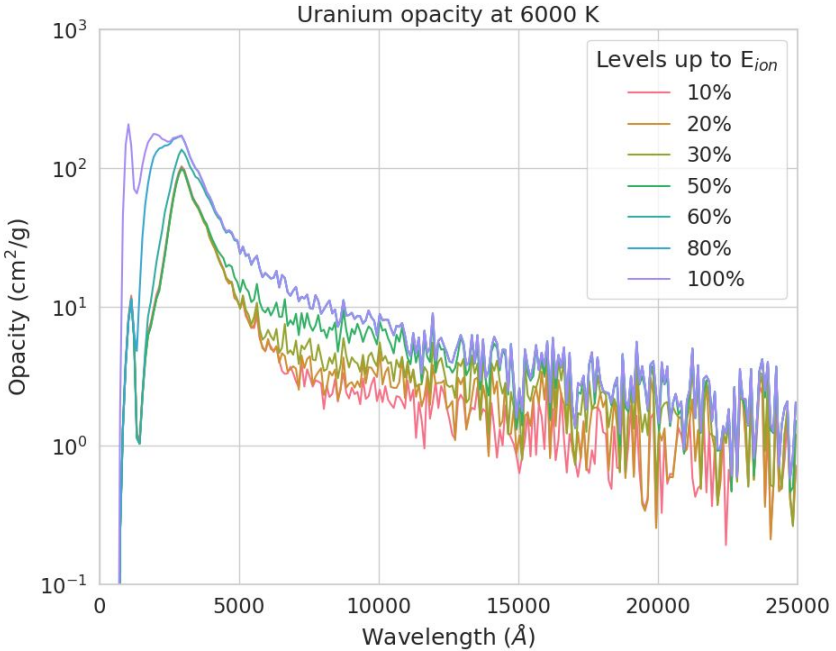


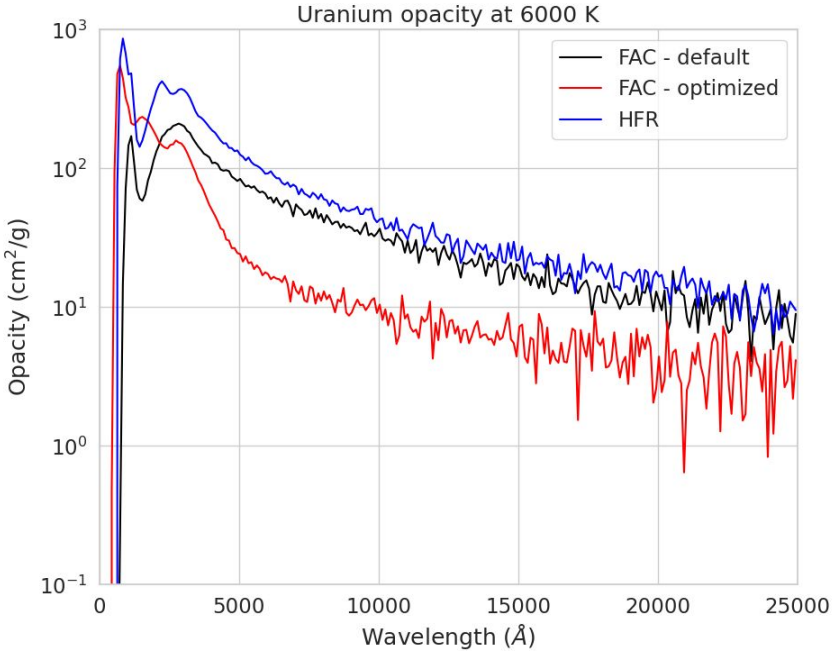
- Systematic discrepancy in $4f^n 5d6s$ configuration
- Consequence of local potential model(?)



- Calibration to experimental data helps but is not sufficient
  → Lack of experimental data
  → Possibly inaccurate wavefunctions

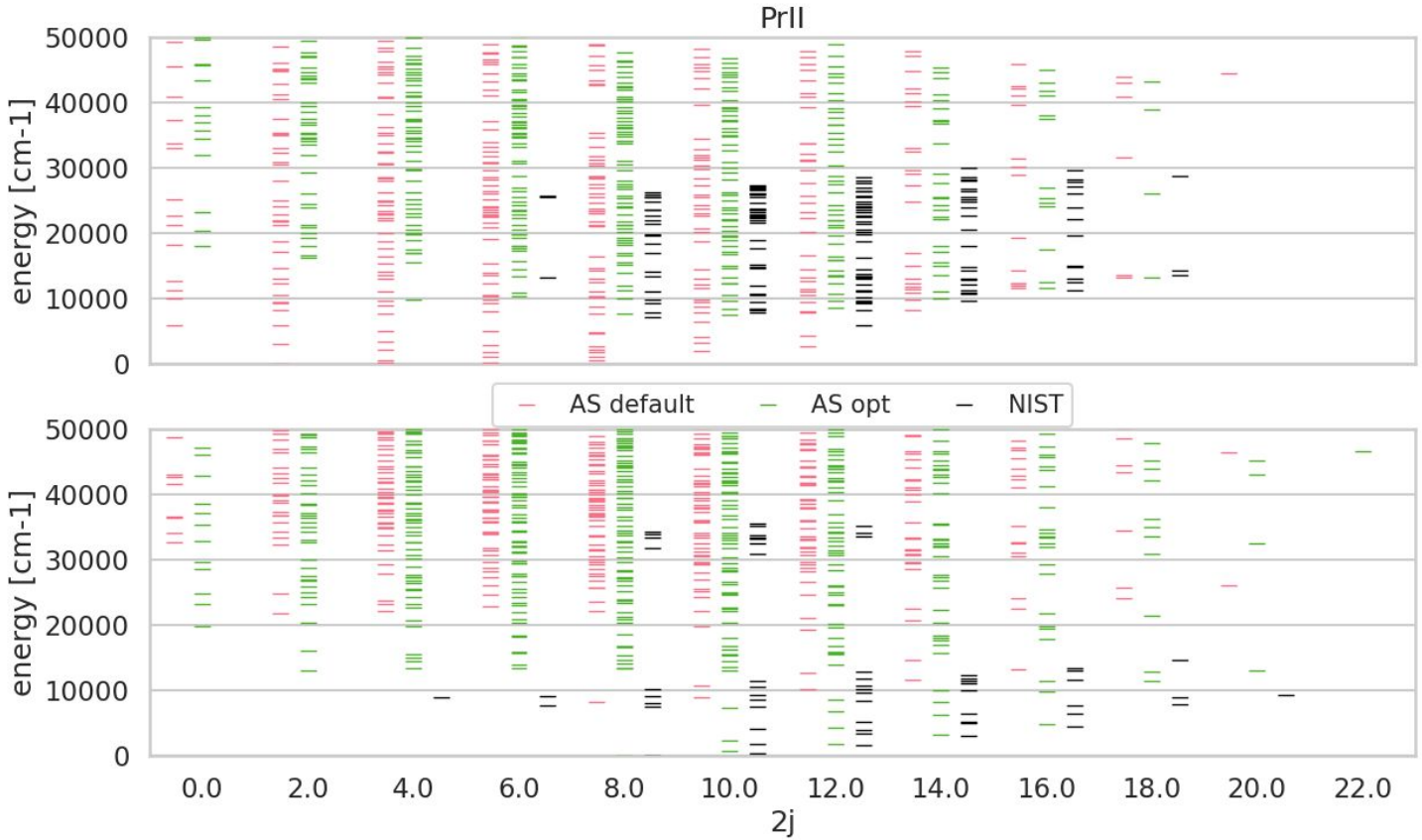
Completeness/Accuracy Duality





- Necessary to ensure convergence
- Differences in atomic data can have significant effect in opacity

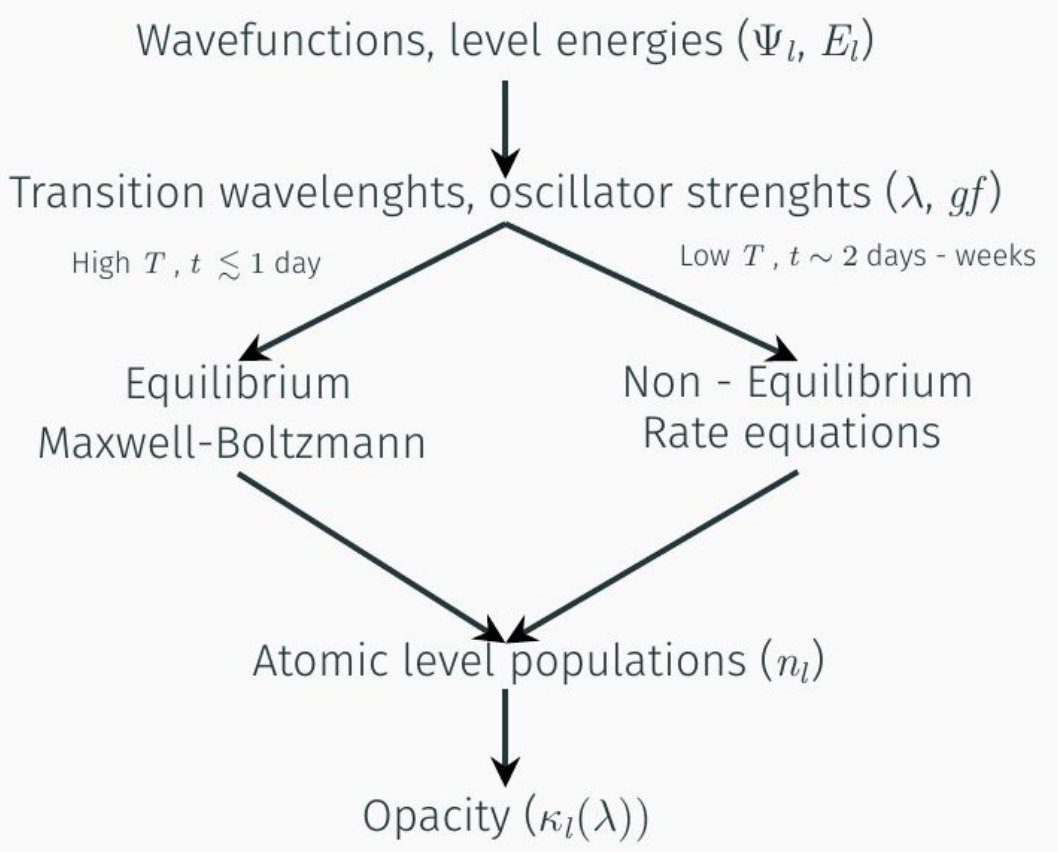
Optimization in AUTOSTRUCTURE



- Faster and better memory handling than FAC - allows for larger computations, essential for CIE, PI and DR (active development by Prof. Nigel Badnel)
- Not fully relativistic

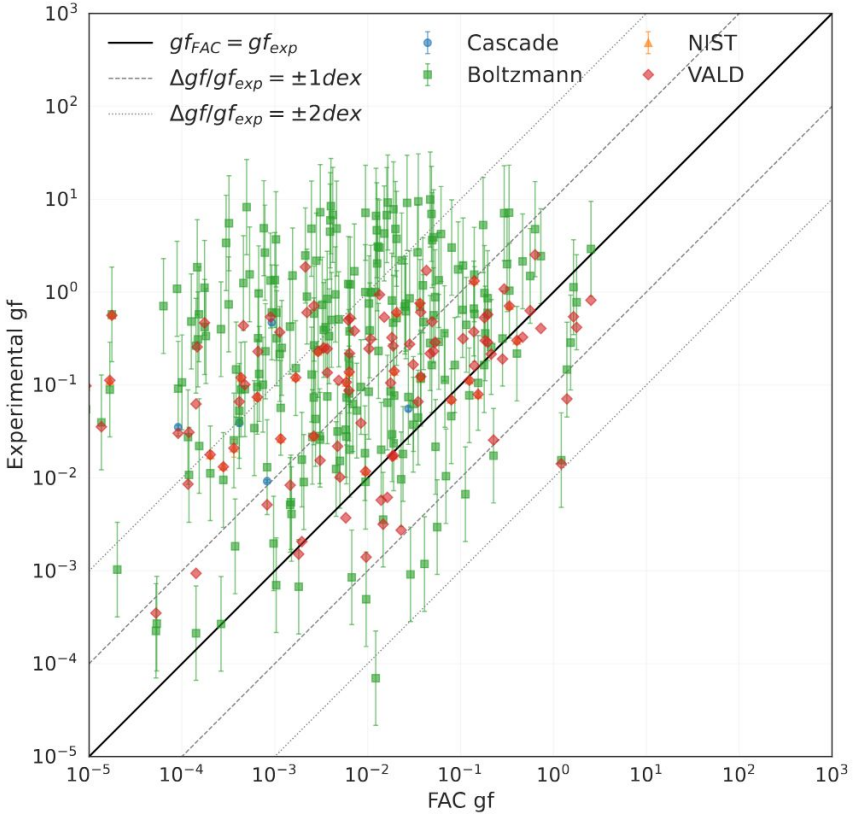
- "AS default" uses one of multiple built in ways to optimized the potential - possible better result if tweaked
- No extra input needed in "AS opt" - optimization in this work

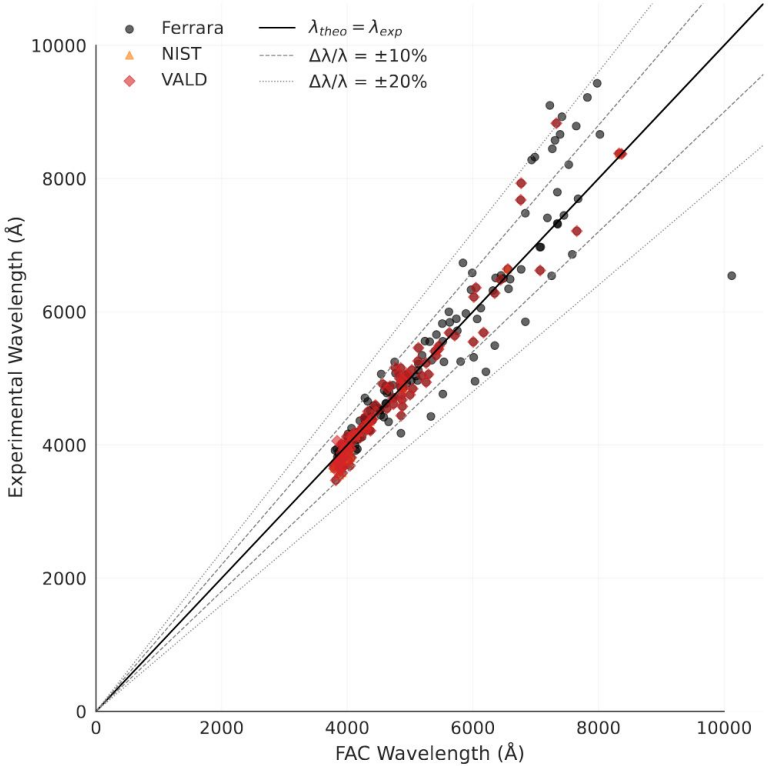
Roadmap to opacity



Allowed Transitions- Lanthanides

RFS+25 10.48550/ARXIV.2502.13250





Assessment of atomic data - Transition rates