Atomic-structure calculations for ultracold gases of lanthanides

Maxence Lepers

In memory of Jean-François Wyart

Conference ASOS, Paris, July 10, 2023

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Ultracold gases (cold atoms)

- Temperatures in **nK-mK range**
- Bose-Einstein condensation
- Precision measurements (atomic clocks)
- Quantum technologies

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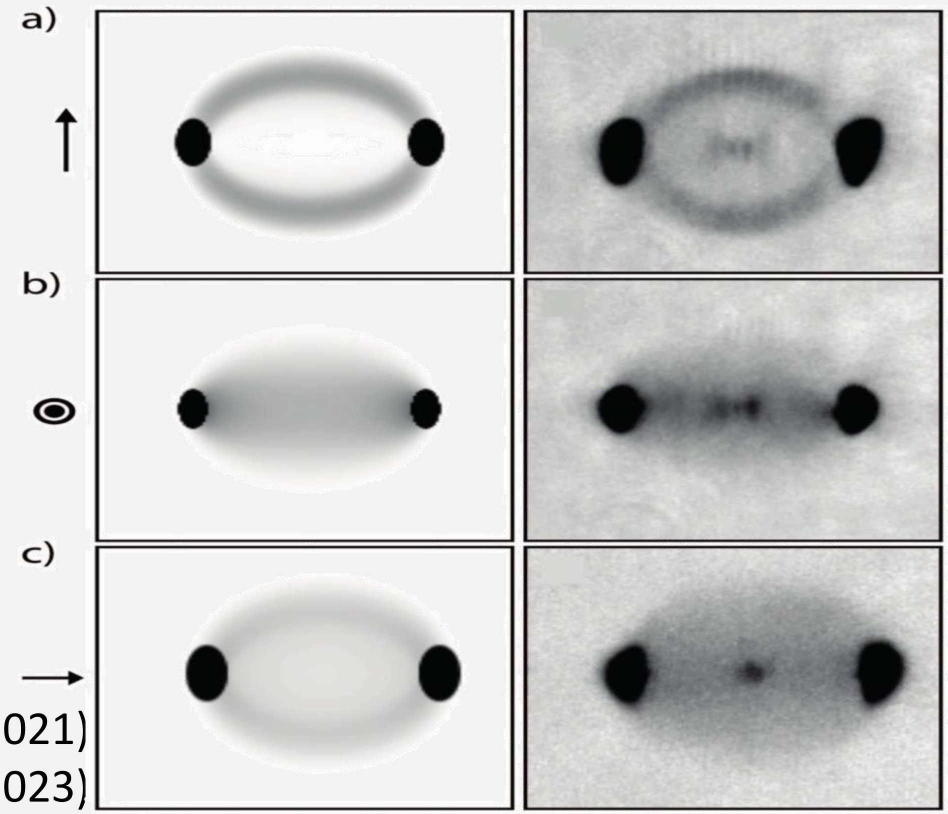
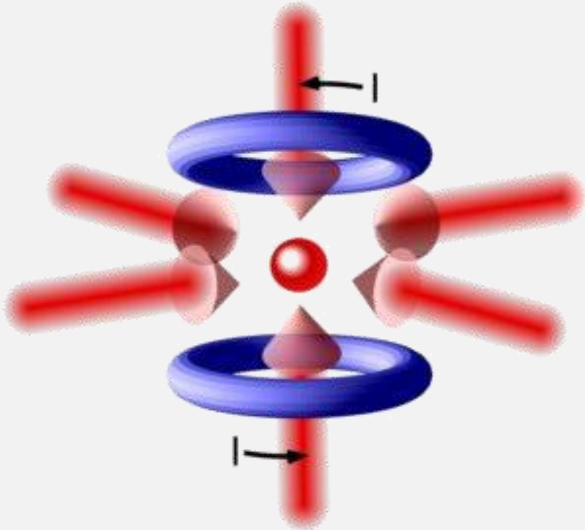
Dipolar gases:

- **Lanthanide magnetic** atoms
- **Anisotropic** and **long-range** dipolar interactions
- Quantum **simulation, many-body** physics, …



M. Norcia & F. Ferlaino, Nat. Phys. **17**, 1349 (2021)
L. Chomaz *et al.*, Rep. Prog. Phys. **86**, 026401 (2023)

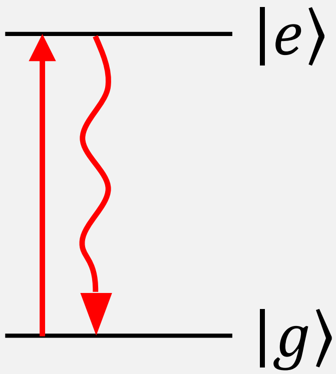
Atomic data for laser-cooling



Vescent website

Many cycles of

absorption – spontaneous emission



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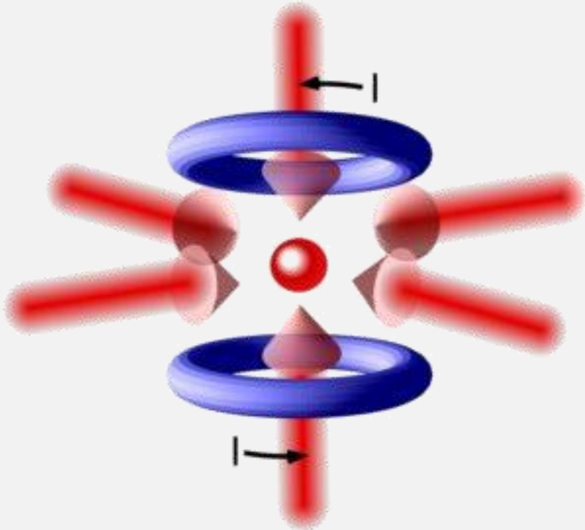
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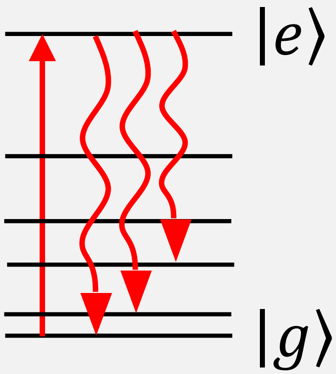
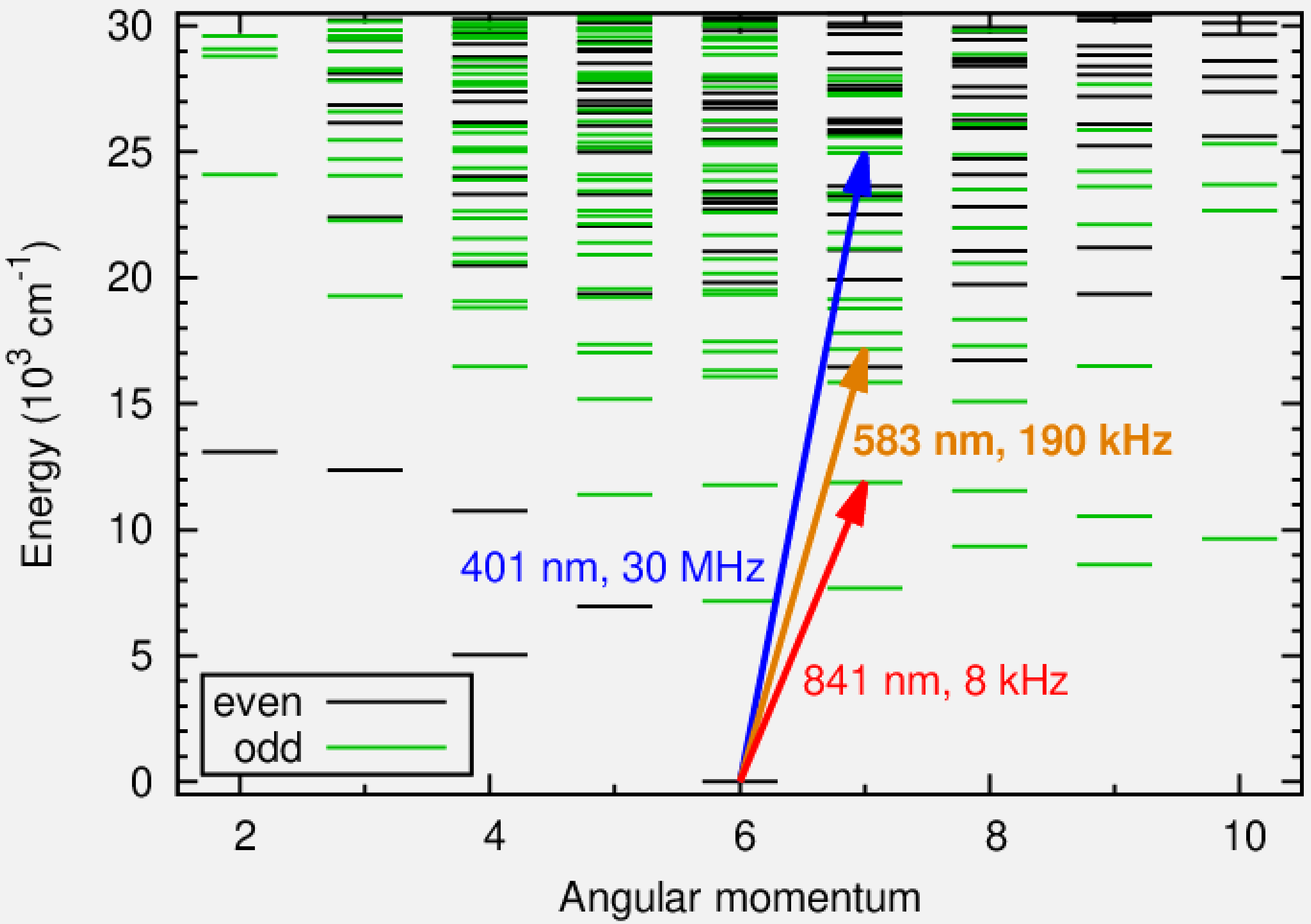


➡ Need A_{ik} Einstein coefficients, branching ratios, line strengths… to characterize **cooling** and **trapping** efficiencies

Laser-cooling scheme for neutral Er



$4f^{12}6s^2\ {}^3\mathrm{H}_6$

J. J. McClelland *et al.*, PRL **96**, 143005 (2006)
G. A. Phelps *et al.*, arxiv:2007.10807 (2020)

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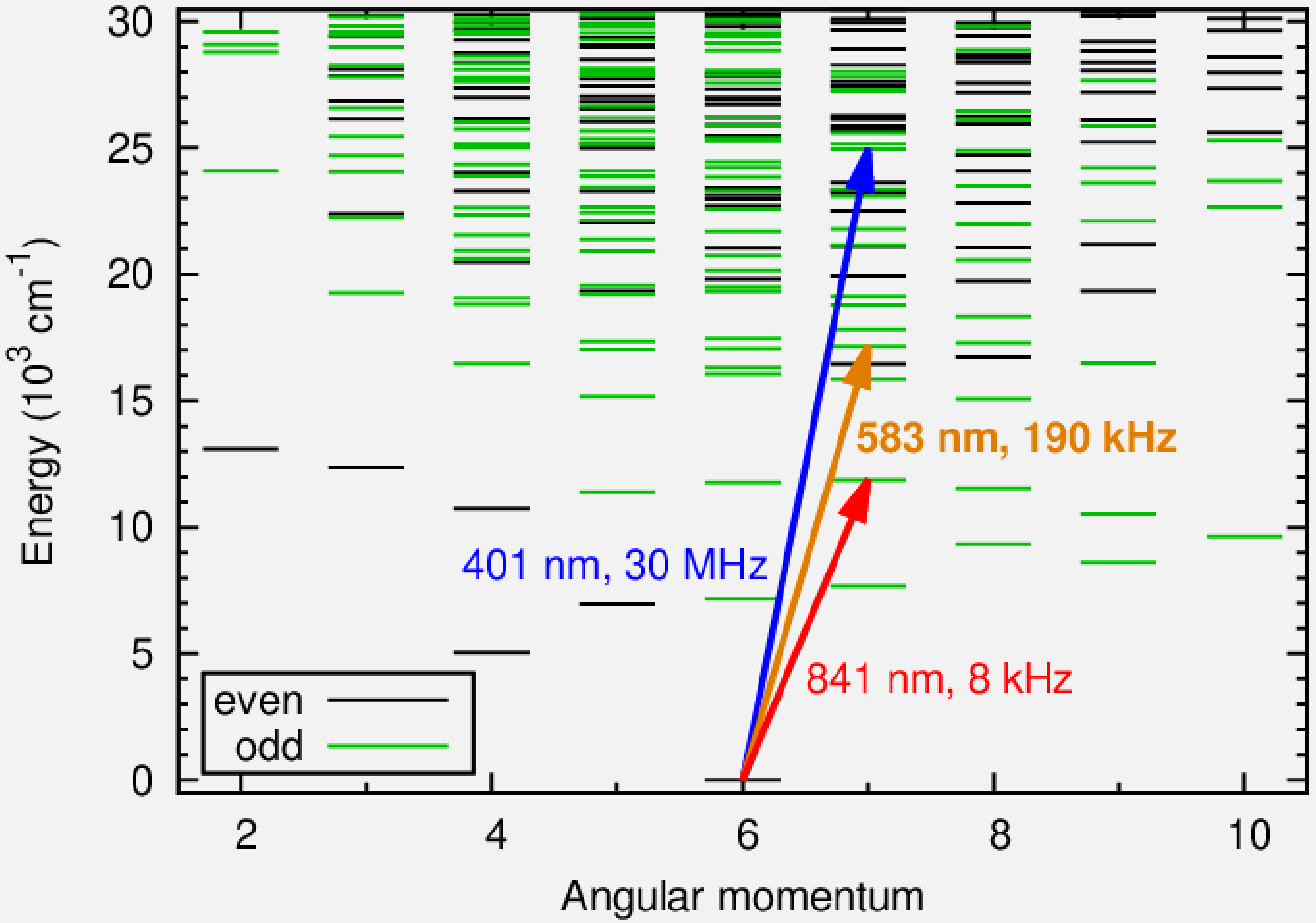

$4f^{12}(^3\mathrm{H}_6)\,6s6p\,(^1\mathrm{P}^o_1)$
$(6,1)^o_7$
$\rightarrow 4f^{12}(^3\mathrm{H}_6)\,5d6s\,(^1\mathrm{D}_2)$

$4f^{12}(^3\mathrm{H}_6)\,6s6p\,(^3\mathrm{P}^o_1)$
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The species under consideration

57 La	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
										Ho_2*	Er^+			
											Er_2			

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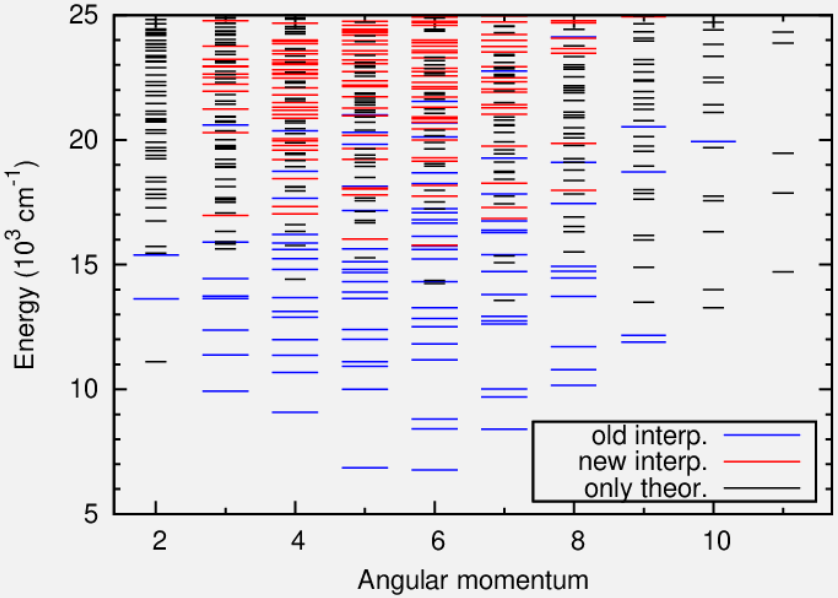
Ho_2* **Er^+**

Er_2



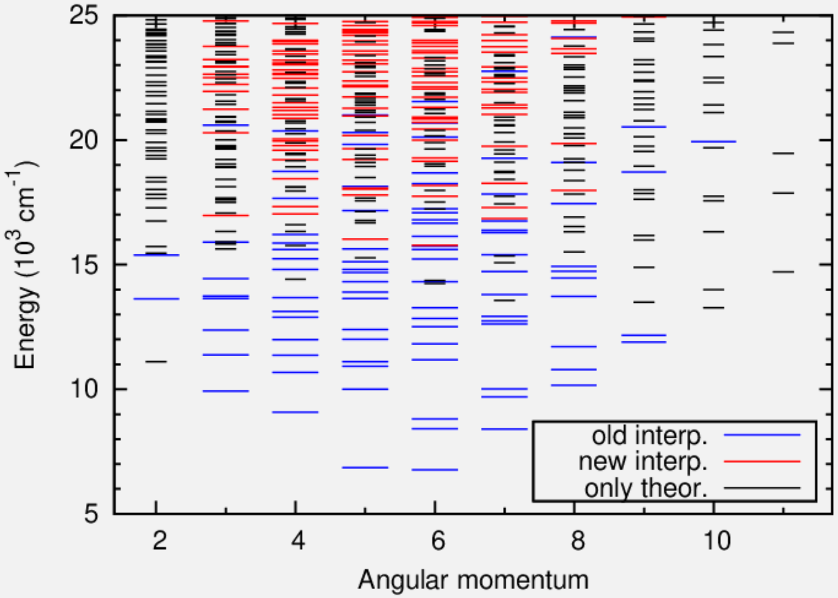
Nd (odd) energy levels:
G. Hovhannesyan & M. Lepers
Phys. Scr. **98** 025407 (2023)

The species under consideration

57	58	59	60	61	62	63	64	65	66	67	68	69	70	71
La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu

Ho_2^* Er^+

Er_2



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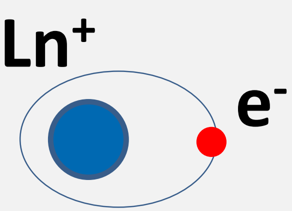


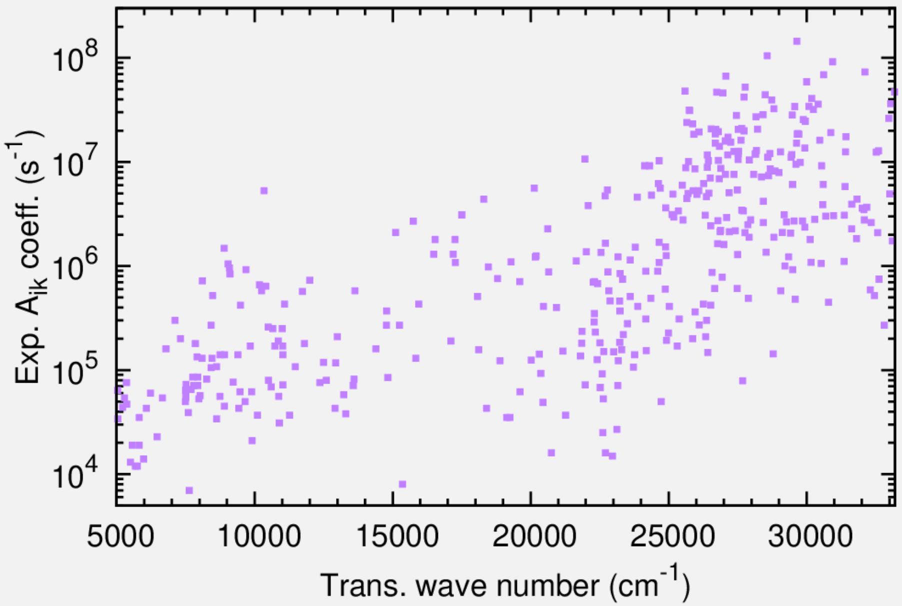
- Ho Rydberg states

J. Hostetter *et al.*, Phys. Rev. A **91**, 012507 (2015)

- **Er Rydberg states**

A. Trautmann *et al.*, Phys. Rev. Research **3**, 033165 (2021)

Einstein A_{ik} coefficients for lanthanides – Er^+



- Many measurements published by the Wisconsin group

Ex: for Er^+, J.E. Lawler *et al.*, ApJSS **78**, 171 (2008)

418 experimental A_{ik} for [289; 1984] nm, levels up to 46750 cm^{-1}

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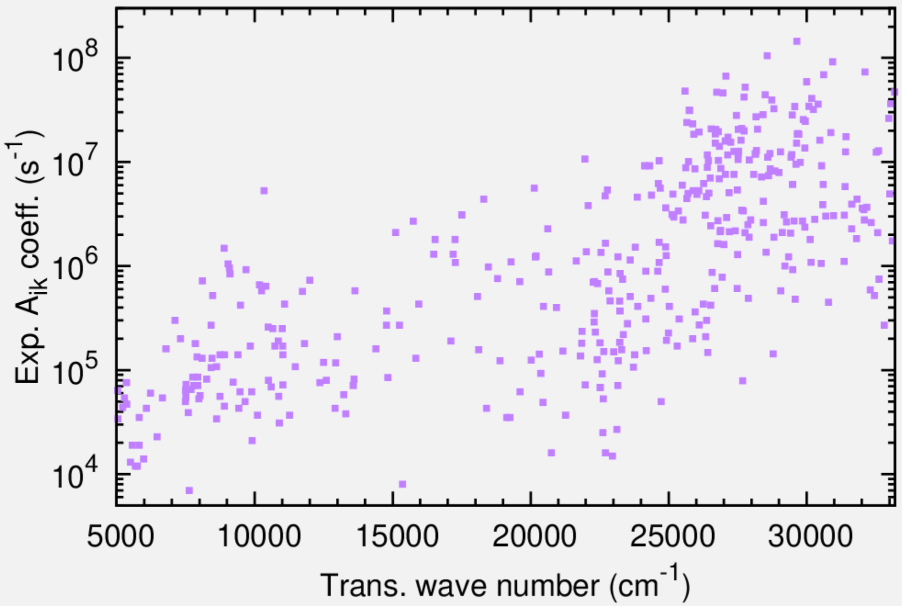

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418 experimental A_{ik} for [289; 1984] nm, levels up to 46750 cm^{-1}

BUT: 21 000+ possible transitions (with E1 approximation)

➡ **Use experimental A_{ik} to adjust a set of appropriate parameters**

➡ **Predict non measured A_{ik} coefficients**

Principle of the calculations

Semi-empirical calculations of energies

*Similar for **energies** and Einstein A_{ik} coefficients*

1. Ab initio calculations (Hartree-Fock + relativistic = **HFR**) for a set of chosen **electronic configurations** => $\{P_{nl}(r)\}$
2. Building the **atomic Hamiltonian** for each **parity and *J***

   H = **radial part $\{P_{nl}(r)\}$** × **angular part (Racah algebra)**

➡ Eigenvalues **(energies)** & eigenvectors (Landé g factors, A_{ik})

3. Least-square fitting of **energies** by adjusting **radial parameters**

Robert D. Cowan's suite of codes
A. Kramida, Atoms **7**, 64 (2019)

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Semi-empirical calculations of A_{ik} coefficients

$$A_{ik} = \frac{\omega_{ik}^3 |\langle i \| \mathbf{d} \| k \rangle|^2}{3\pi\varepsilon_0 \hbar c^3 (2J_i + 1)}$$

$$|i, k\rangle = \sum_b c_b |b\rangle$$

Semi-empirical calculations of A_{ik} coefficients

$$A_{ik} = \frac{\omega_{ik}^3 |\langle i \| \mathbf{d} \| k \rangle|^2}{3\pi\varepsilon_0 \hbar c^3 (2J_i + 1)}$$

$$\equiv A_t = \left(\sum_j a_{tj} \times r_j \right)^2$$

$$|i, k\rangle = \sum_b c_b |b\rangle$$

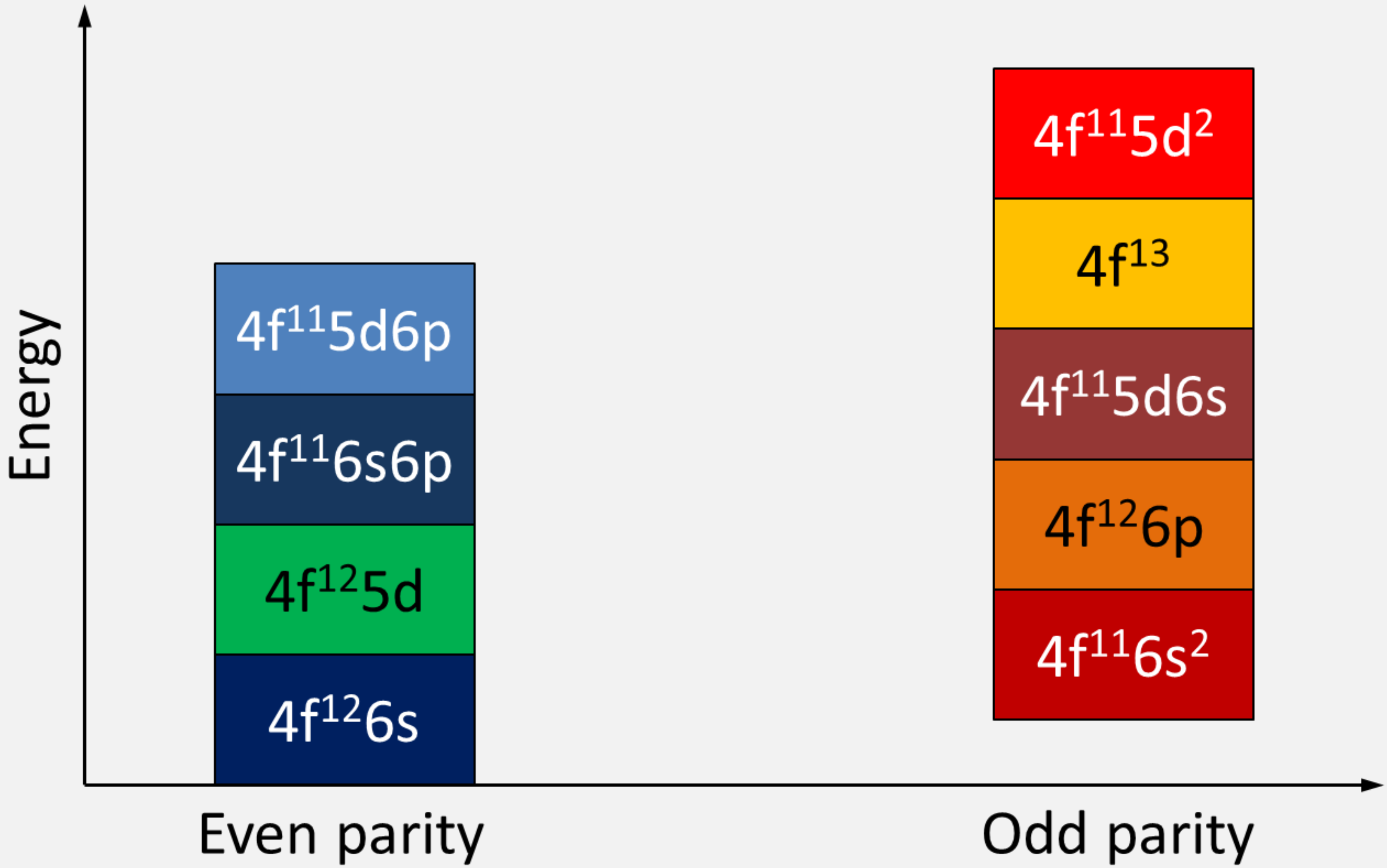
a_{tj} depend on **eigenvalues** and eigenvectors (previous slide)

$r_j = \int_0^{+\infty} dr P_{nl}(r) r P_{n'l'}(r) =$ adjustable parameters

$(nl, n'l')$ safistify the electric-dipole selection rules

$$r_j = f_j \times r_{j,\mathrm{HFR}}$$

Involved configurations in Er^+



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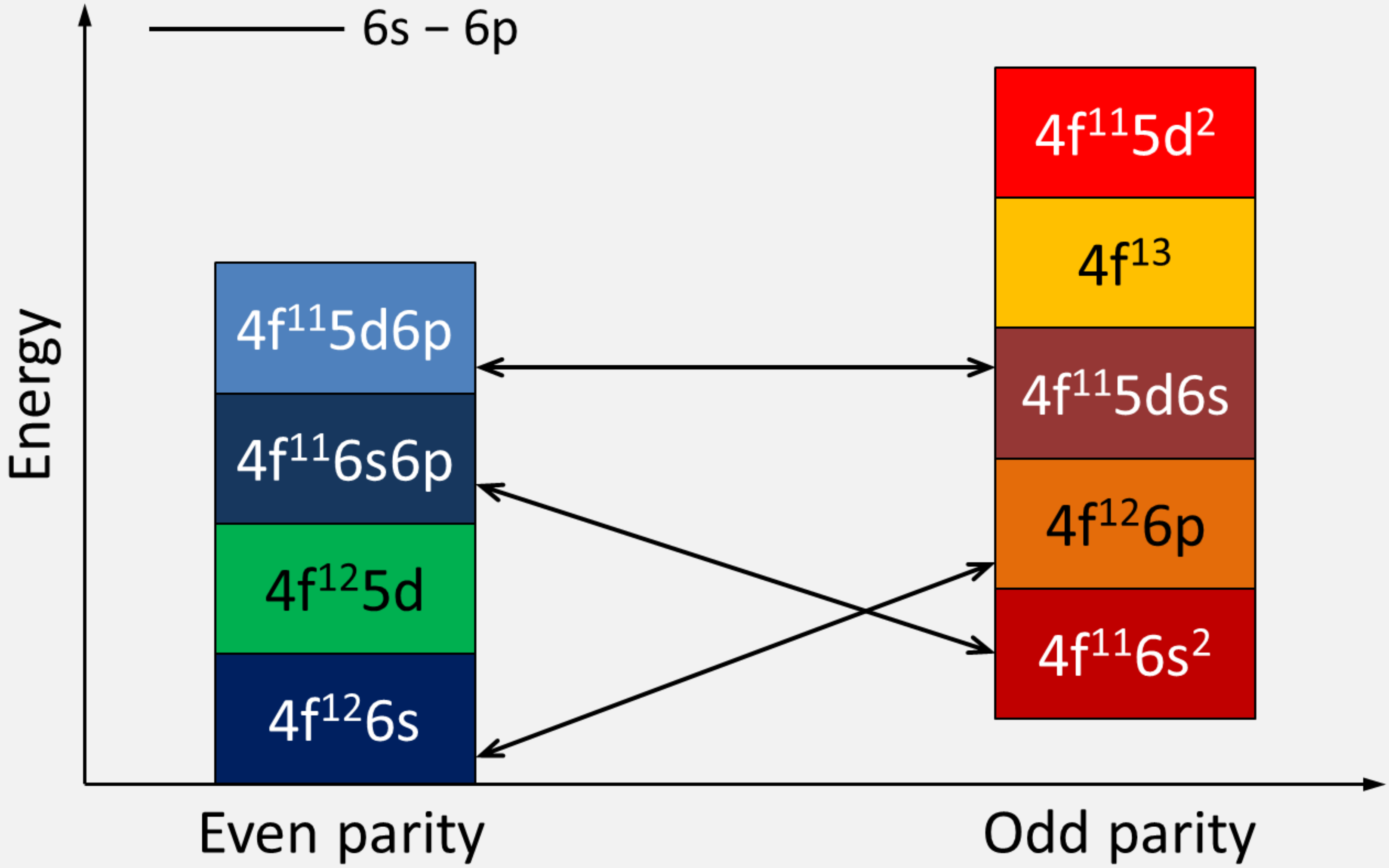

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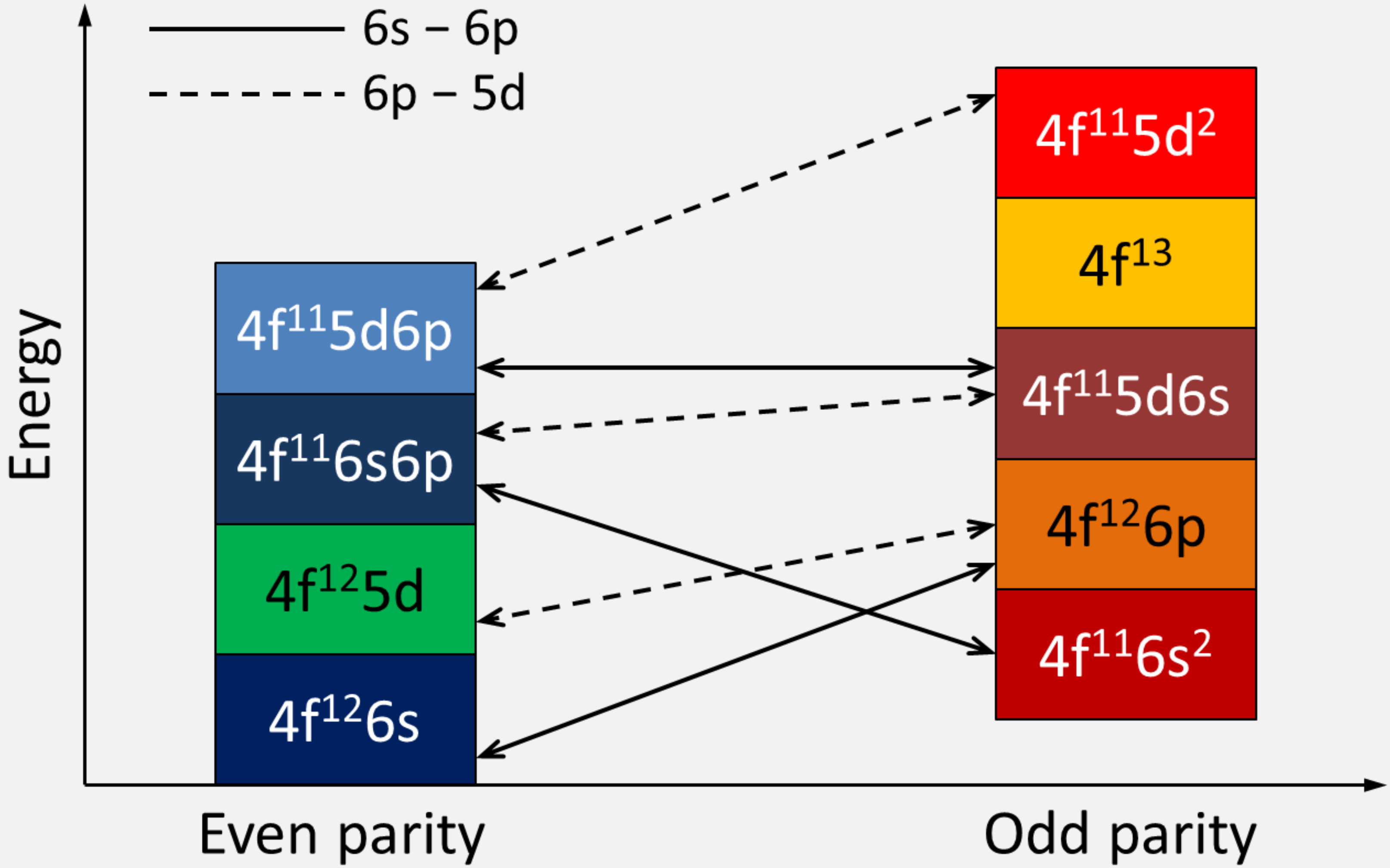

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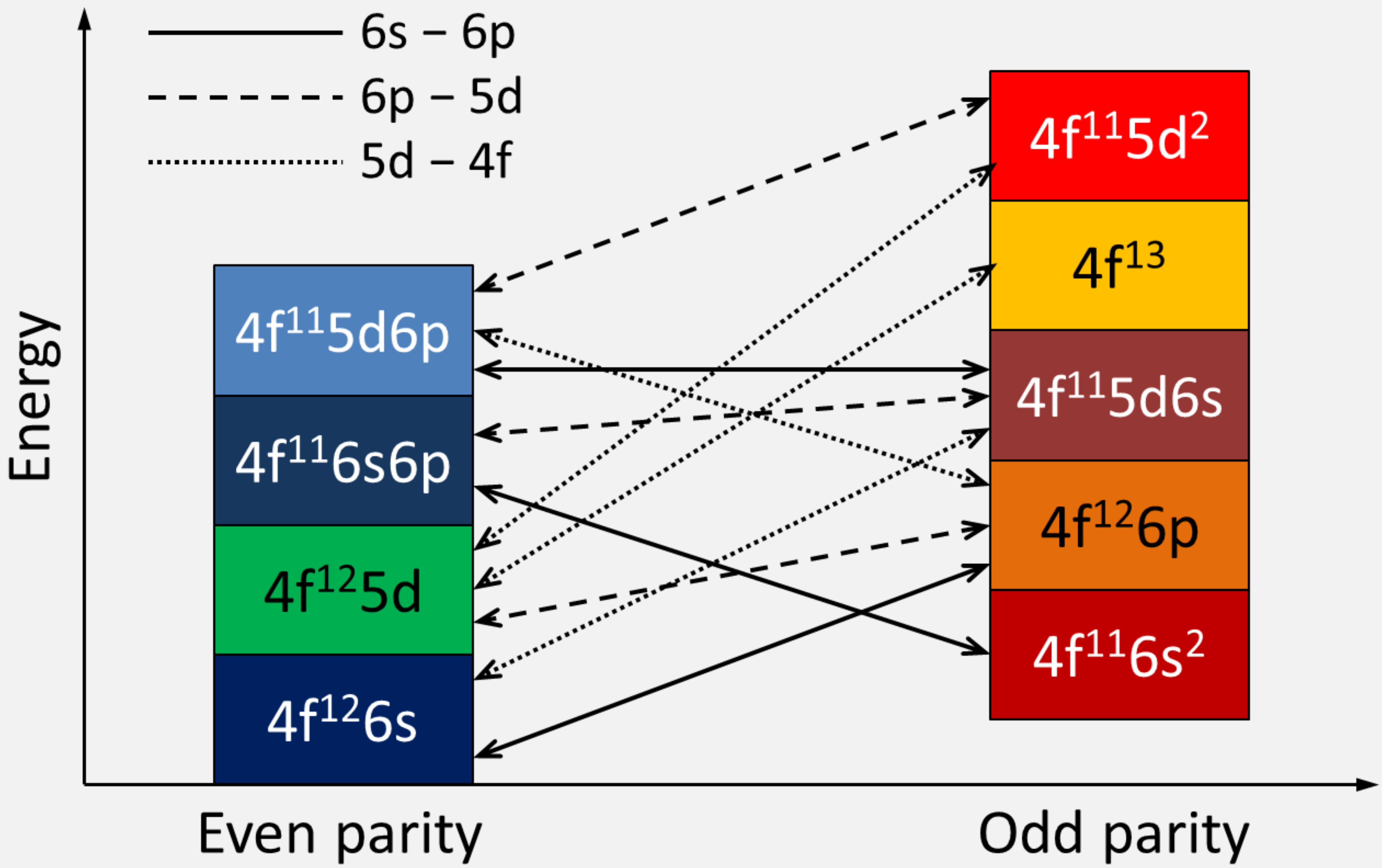
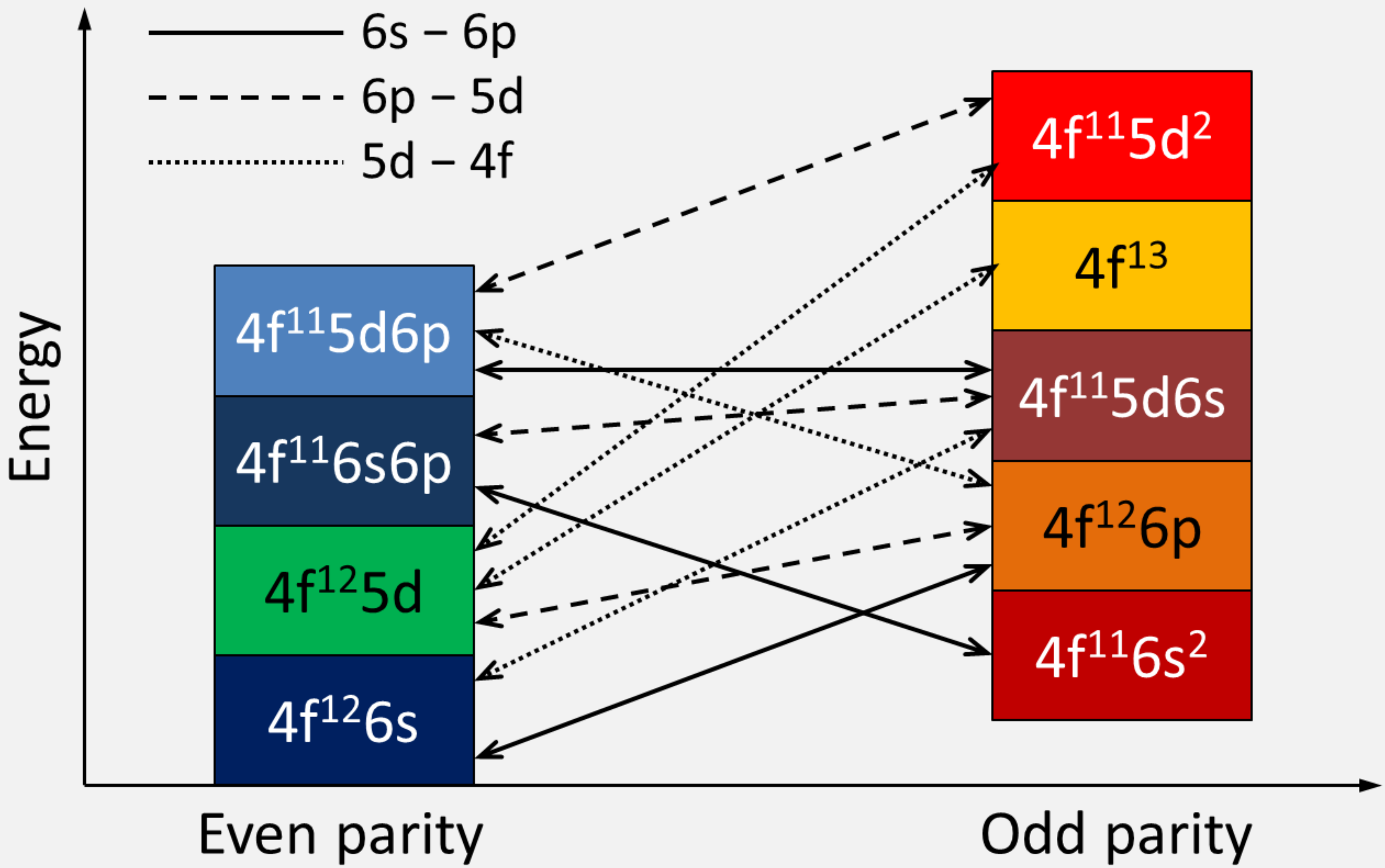
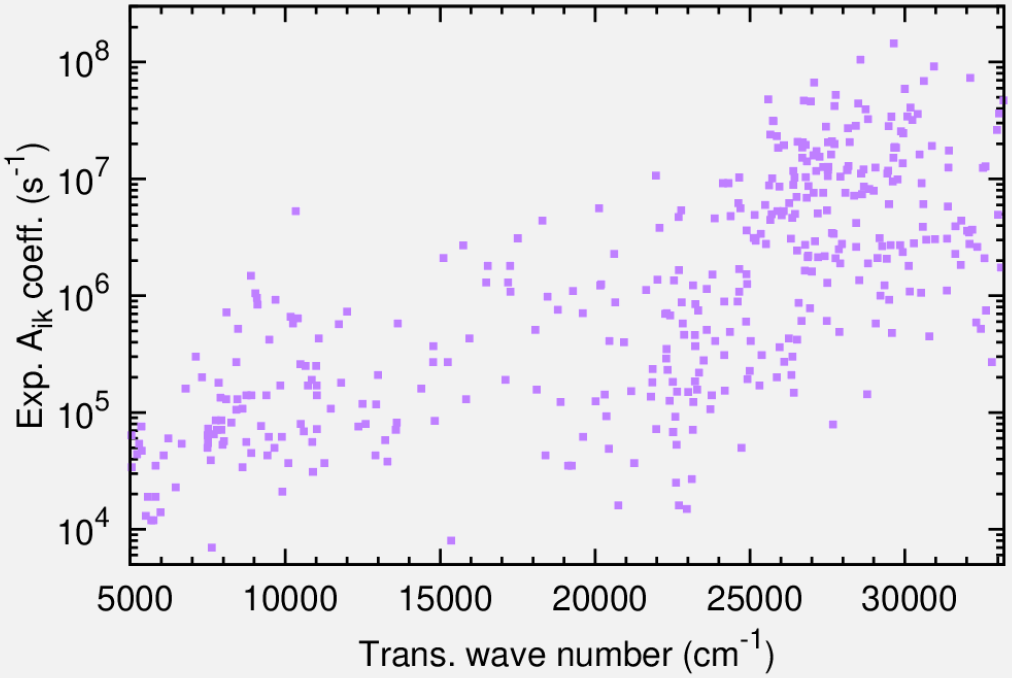


Involved configurations in Er^+



➡ **10 adjustable r_j parameters, 5 groups**

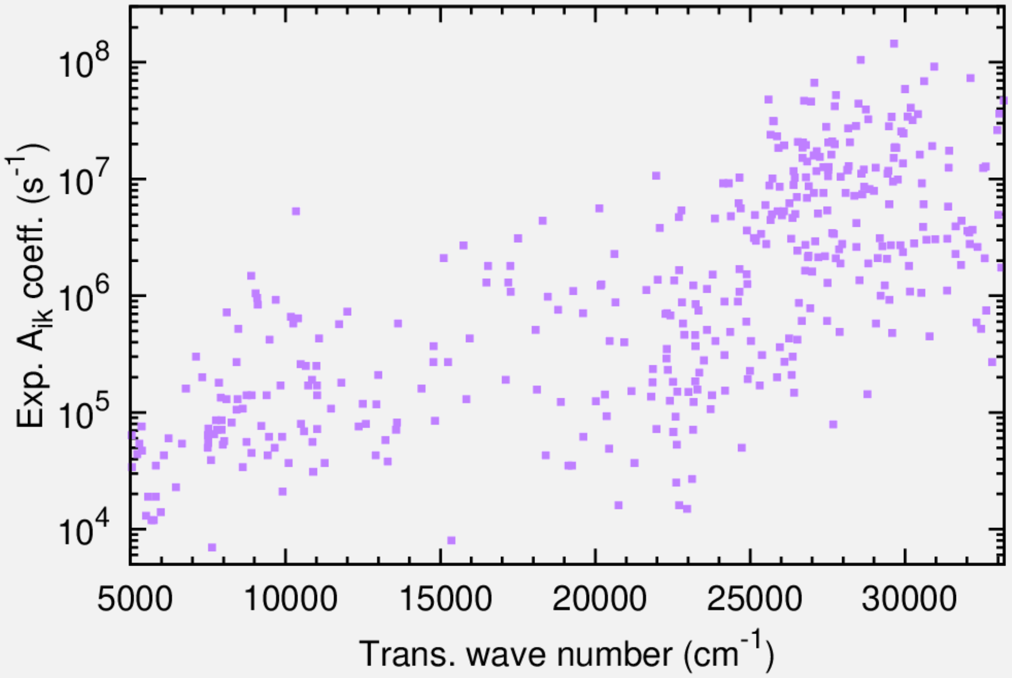
Semi-empirical calculations of A_{ik} coefficients



Linear standard deviation

$$\sigma_A = \left[\frac{\sum_{i=1}^{N_{\rm tr}} \left(A_{t,\rm cal} - A_{t,\rm exp} \right)^2}{N_{\rm tr} - N_{\rm par}} \right]^{1/2}$$

Semi-empirical calculations of A_{ik} coefficients



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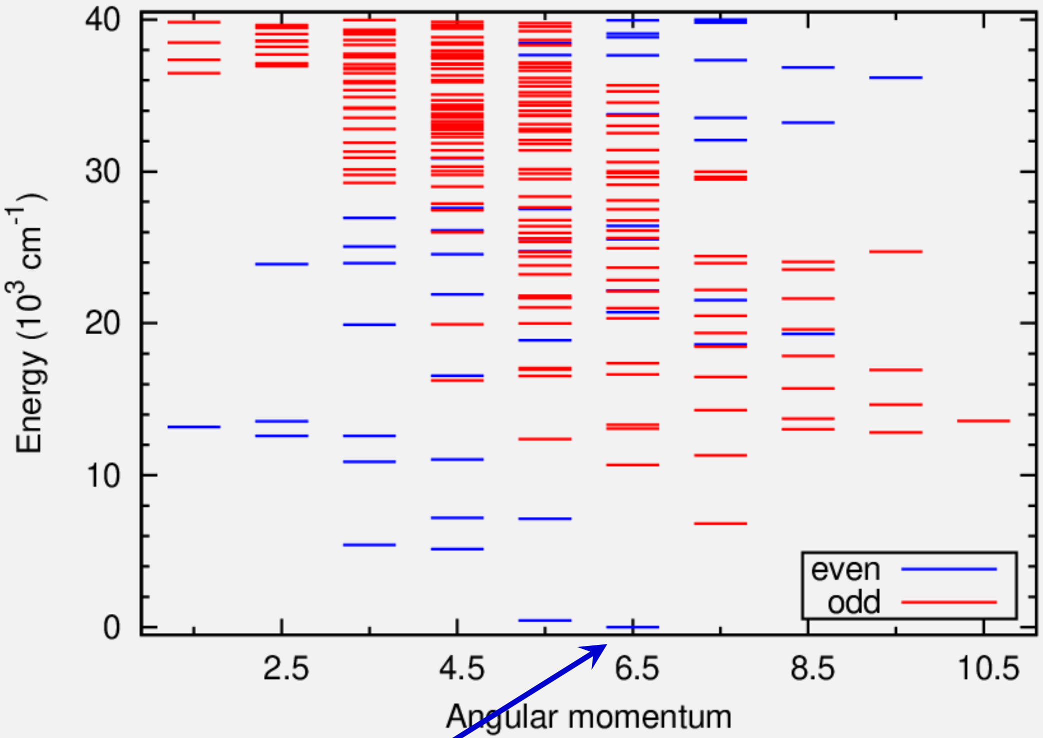
Logarithmic standard eviation

$$\sigma_{{\rm lg}A} = \left[\frac{\sum_{i=1}^{N_{\rm tr}} \log^2 \left(\frac{A_{t,{\rm cal}}}{A_{t,{\rm exp}}} \right)}{N_{\rm tr} - N_{\rm par}} \right]^{1/2} .$$

Results for erbium

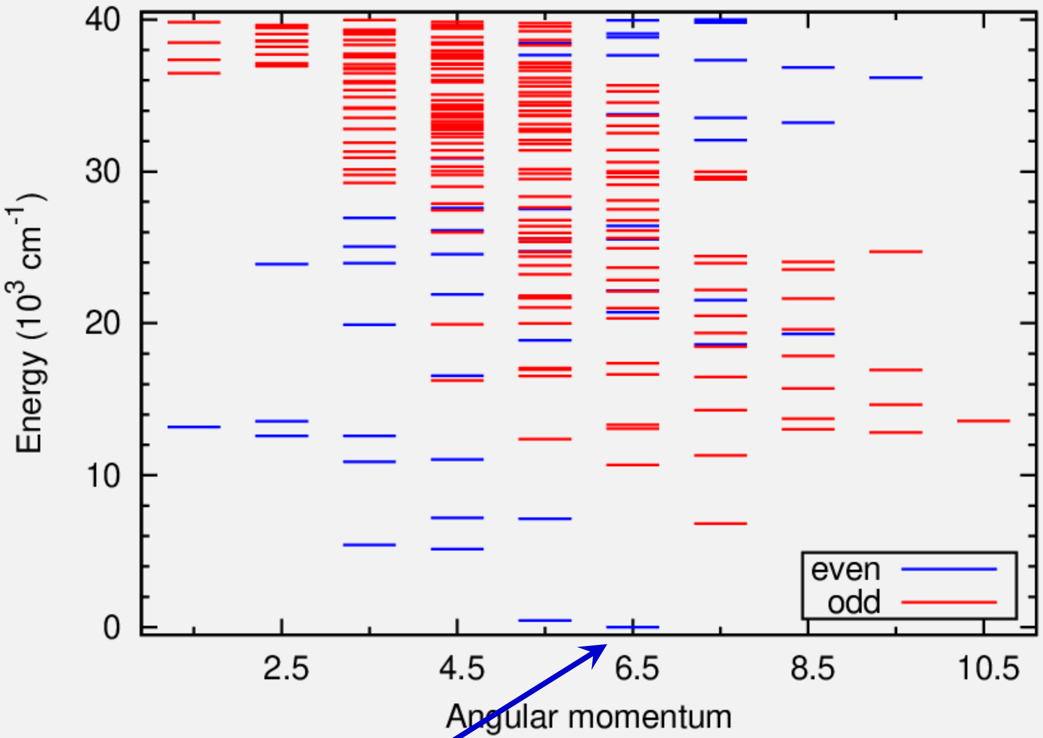
Fitted energy levels



Experimental levels from NIST databasze

Ground level:
[Xe]$4f^{12}(^3H_6)6s\ ^4H_{13/2}$

Fitted energy levels



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J.-F. Wyart and J. E. Lawler, Phys. Scr. **79**, 045301 (2009)

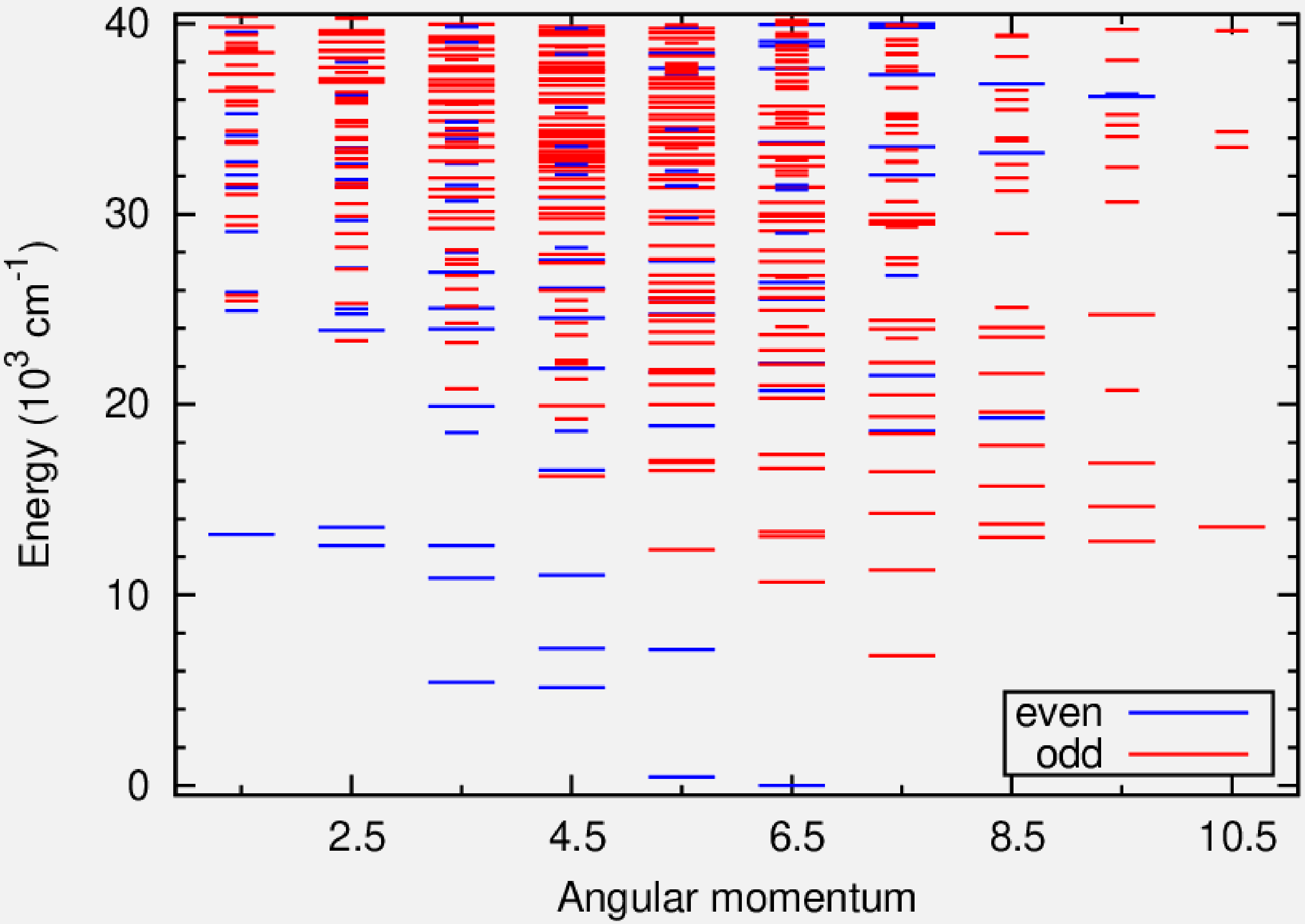
Standard deviation:

$$\boldsymbol{\sigma_E} = \left[\frac{\sum_{i=1}^{N_{\mathrm{lev}}} \left(E_{\mathrm{cal},i} - E_{\mathrm{exp},i}\right)^2}{\boldsymbol{N}_{\mathrm{lev}} - \boldsymbol{N}_{\mathrm{par}}}\right]^{1/2}$$

Parity	even	odd
N_{lev}	130	233
N_{par}	25	21
σ_E (cm^{-1})	55	63

Predicted energy levels

Experimental levels (long dashes) + predicted levels (short dashes)



Fitting of A_{ik} coefficients

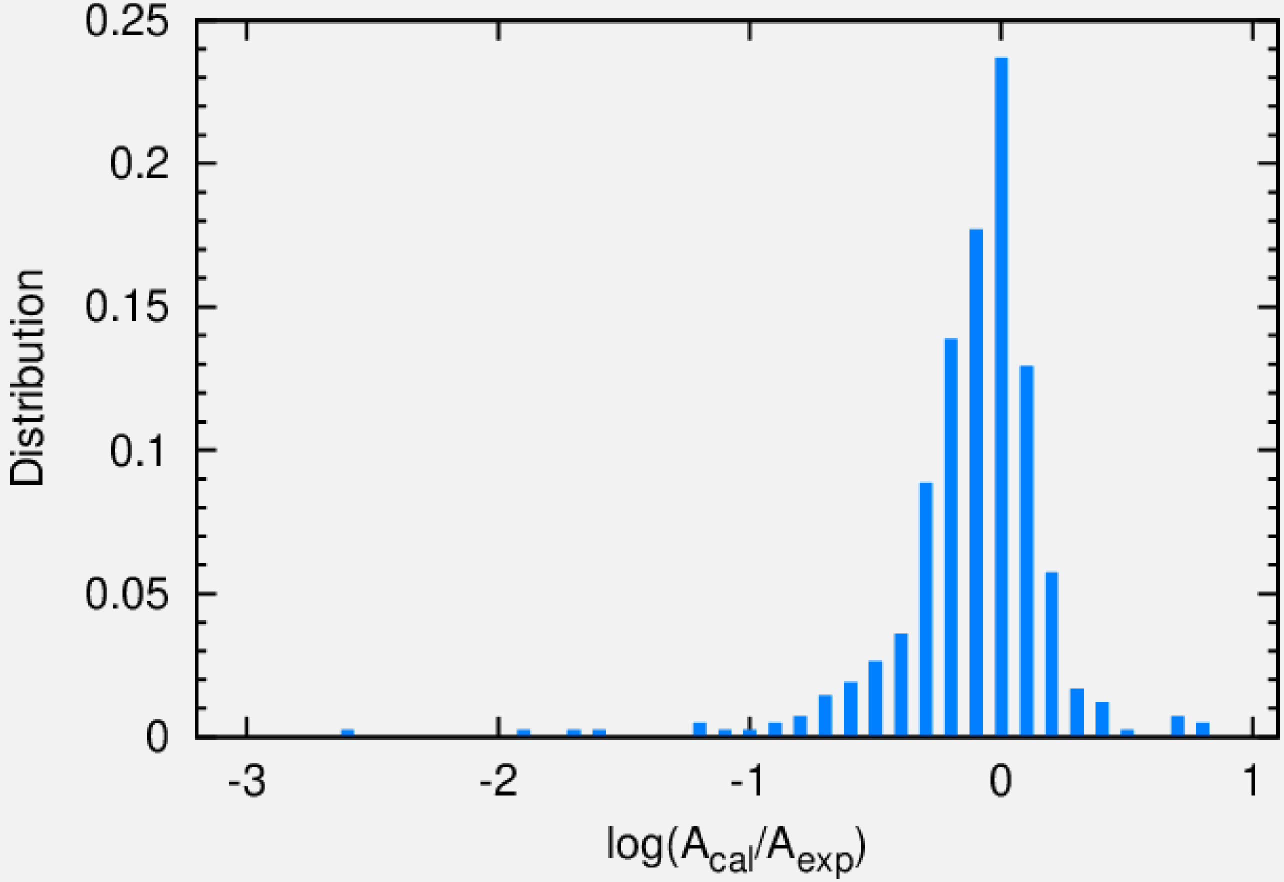
Group Number	Subshell pair	Scaling factor
1	$\langle 4f^{12}\mathbf{6s}\vert r\vert 4f^{12}\mathbf{6p}\rangle$	0.886
2	$\langle 4f^{11}\mathbf{6s^2}\vert r\vert 4f^{12}6s\mathbf{6p}\rangle$	0.876
3	$\langle 4f^{11}5d\mathbf{6s}\vert r\vert 4f^{12}5d\mathbf{6p}\rangle$	0.797
4	All $\langle\mathbf{5d}\vert r\vert\mathbf{6p}\rangle$	0.808
5	All $\langle\mathbf{4f}\vert r\vert\mathbf{5d}\rangle$	0.817
σ_A (s^{-1})		4.66 x 10^6
σ_{lgA}		0.217

2.2 % of the best largest A_{exp}

0.61 < most ratios < 1.65

A_{ik} coefficients

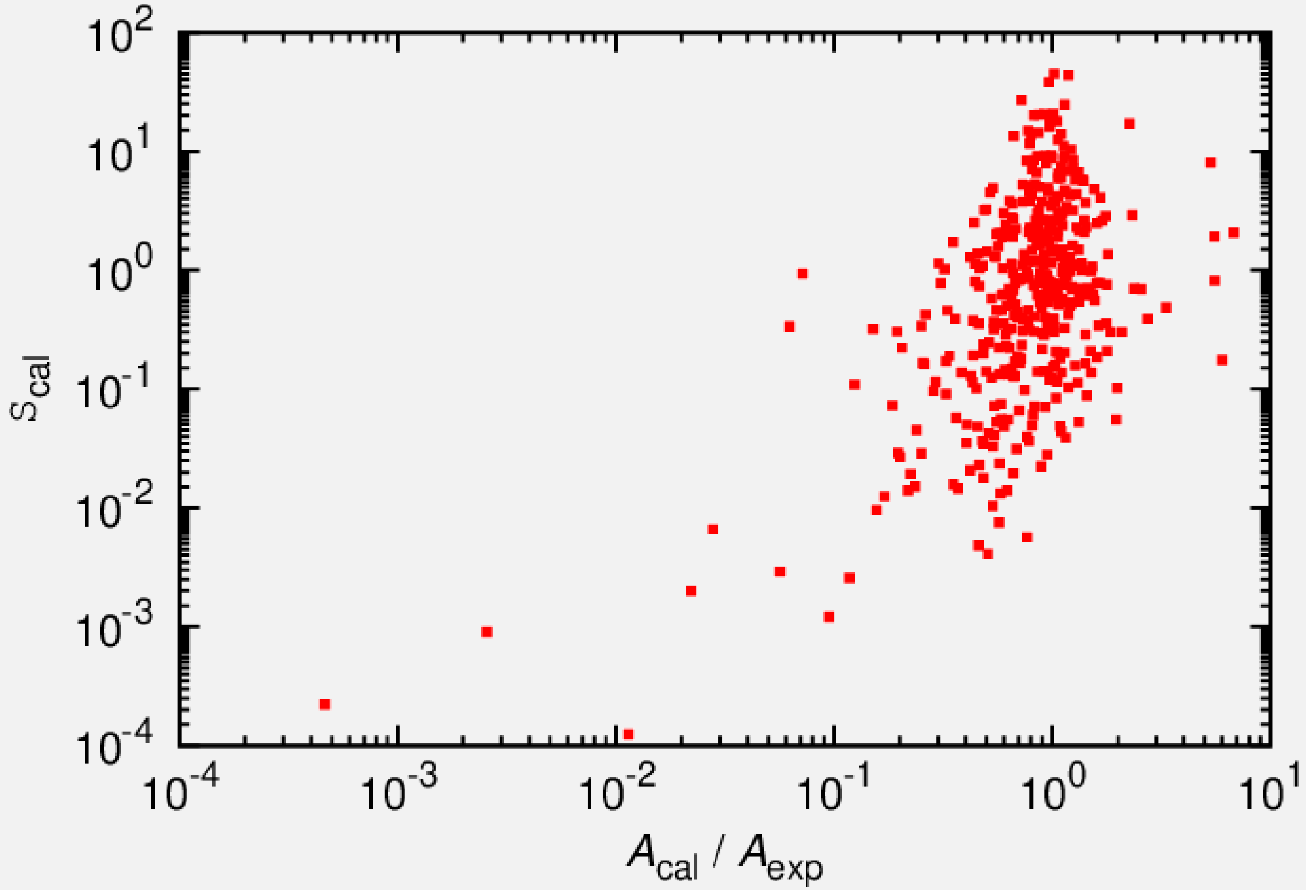
Including all experimental transitions



22 transitions with ratio < 0.2 or > 5

A_{ik} coefficients

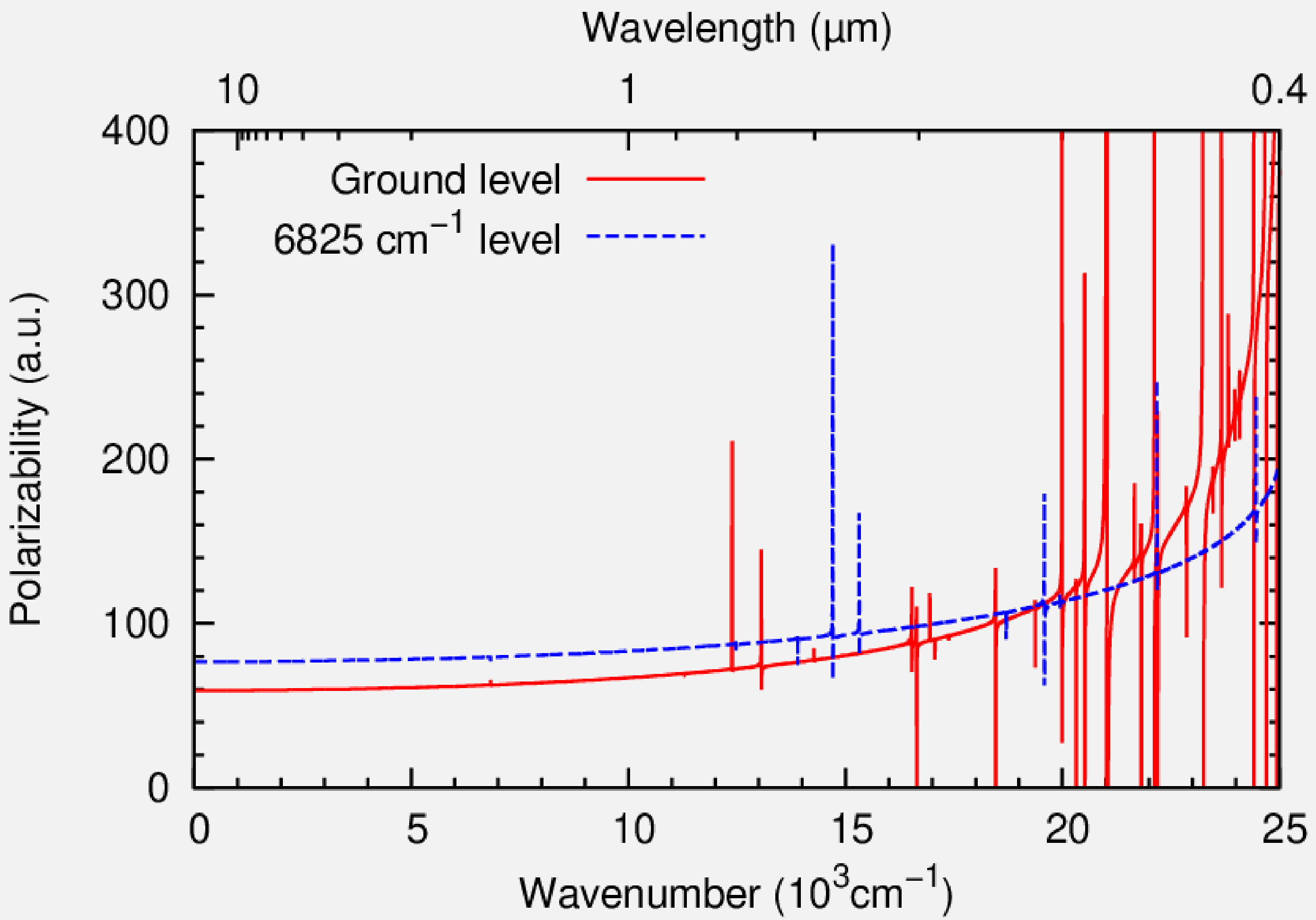
Including all experimental transitions



A. Kramida, FST **63**, 313 (2013)

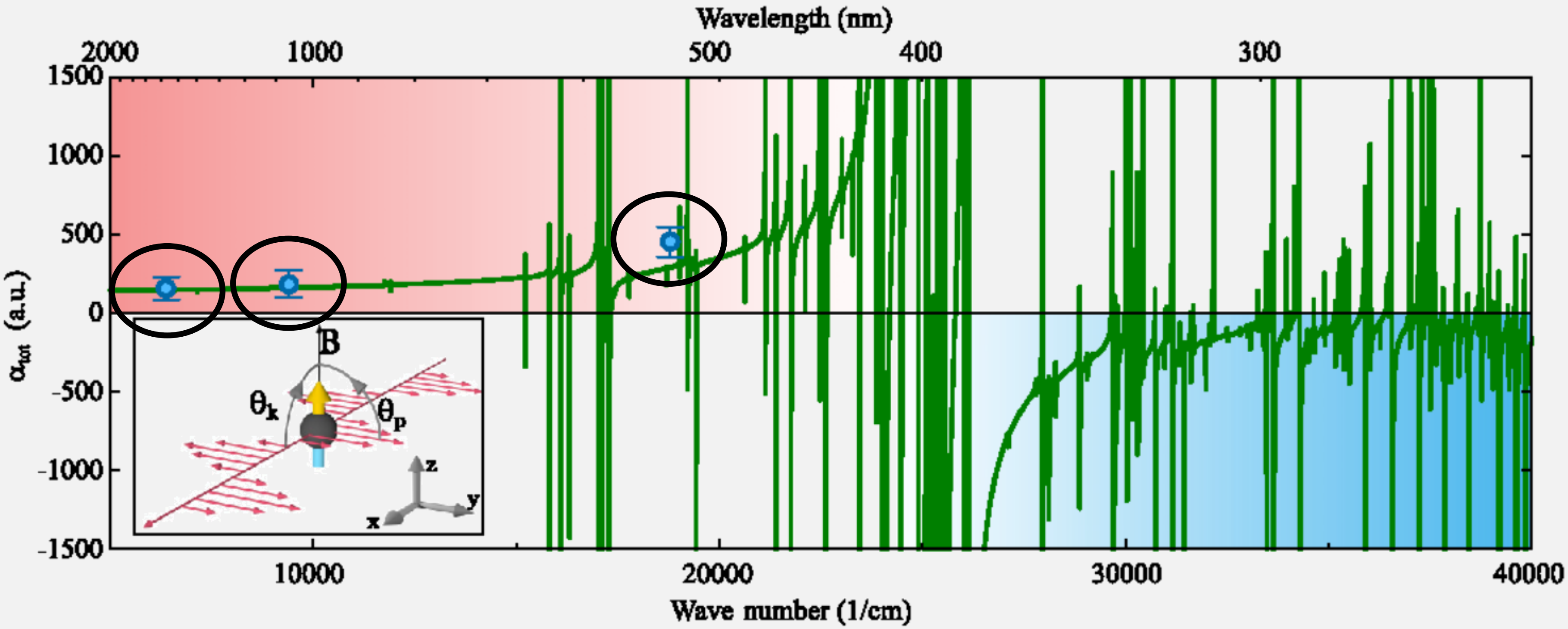
Dynamic dipole polarizability (Er^+)

$$\alpha_{\mathrm{scal}}(\omega) = \frac{2}{3\hbar(2J+1)} \sum_{\beta' J'} \frac{\omega_{\beta' J', \beta J}}{\omega_{\beta' J', \beta J}^2 - \omega^2} |\langle \beta' J' \| \mathbf{d} \| \beta J \rangle|^2$$



Dynamic dipole polarizability (neutral Er)

Comparison with ultracold experiment (Innsbruck)



J. H. Becher *et al.*, Phys. Rev. A **97**, 012509 (2018)

Conclusion & propospects

- Calculation of **A_{ik} coefficients** for **complex cold atoms**
- Using **least-square fitting** to experimental values
- Package *FitAik* working in **interaction with Cowan**

  https://gitlab.com/labicb/fitaik/
- Publication in **CaDDiAcS database**

  https://vamdc.icb.cnrs.fr/caddiacs/
- Results for **Er^+**, but also **Er, Dy, Ho, Cr**, **Nd**
- Future calculations: **Tm, Tm^+, …**

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Thank you for your attention !

Angular part of the Hamiltonian

Angular basis sets (in LS coupling)

$$|b\rangle = |n_1 l_1^{w_1}\, \alpha_1 L_1 S_1, n_2 l_2^{w_2}\, \alpha_2 L_2 S_2, LSJM\rangle$$

- Orbital angular momentum: $\vec{L} = \vec{L}_1 + \vec{L}_2$
- Spin angular momentum: $\vec{S} = \vec{S}_1 + \vec{S}_2$
- Total angular momentum: $\vec{J} = \vec{L} + \vec{S}$

Ex: Er^+ ground level: [Xe]$4f^{12}(^3H)\ 6s(^2S)\ {}^4H_{13/2}$

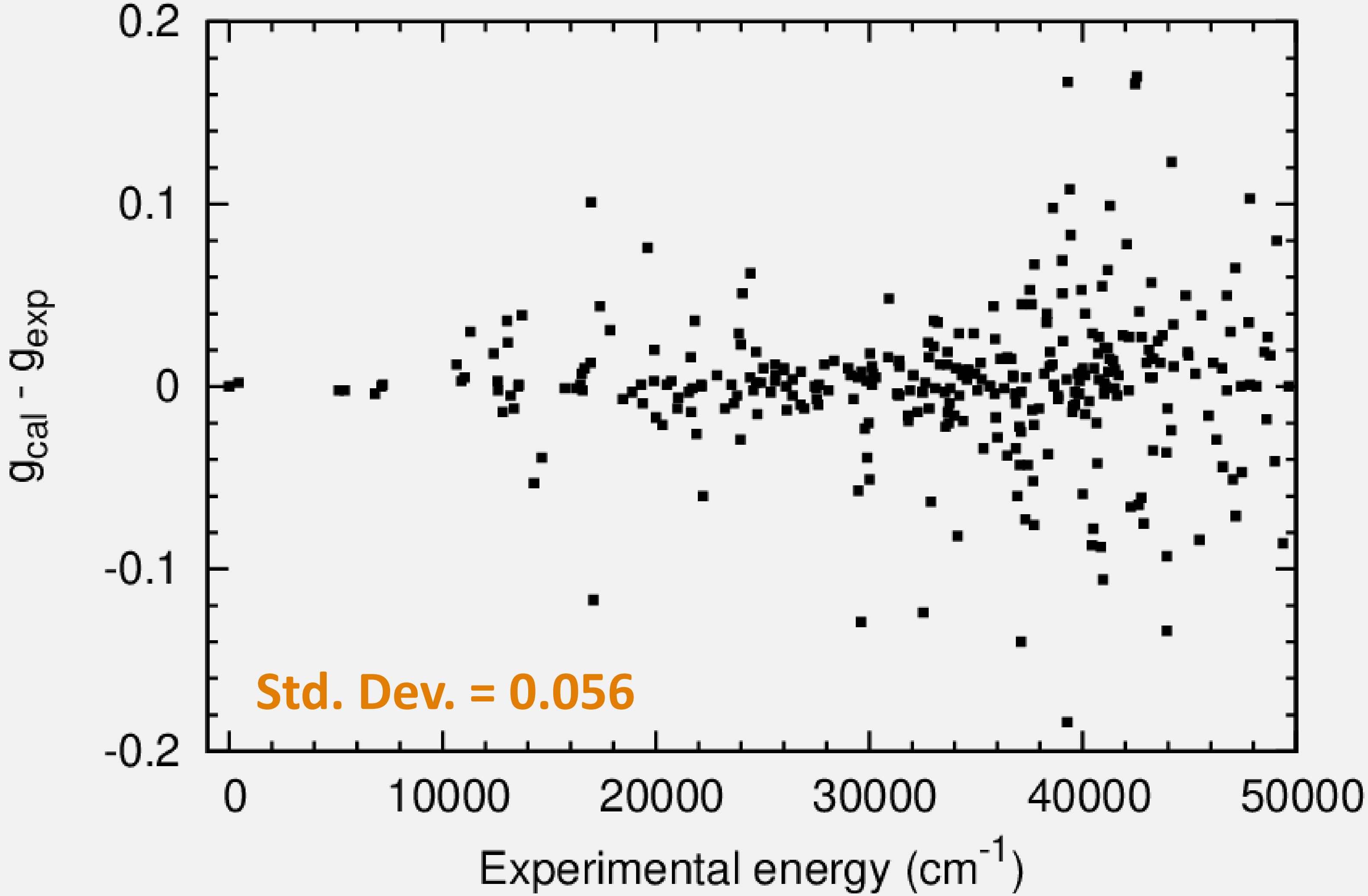
$L_1 = 5$
$S_1 = 1$

$L_2 = 0$
$S_2 = 1/2$

$L = 5$
$S = 3/2$
$J = 13/2$

Landé-g factors

Magnetic moment: $\vec{\mu} = -\mu_B \times g \times \vec{J}$



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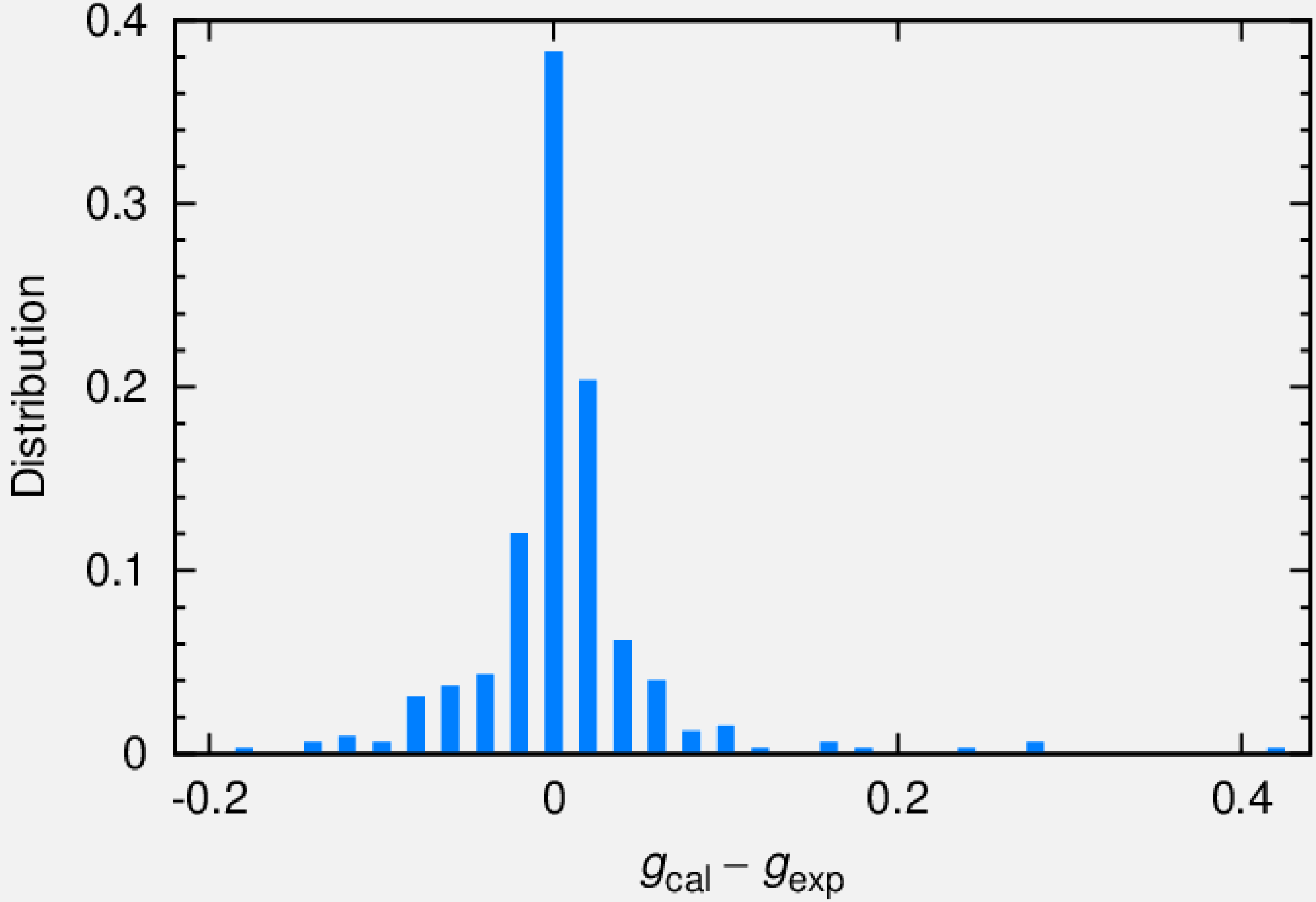


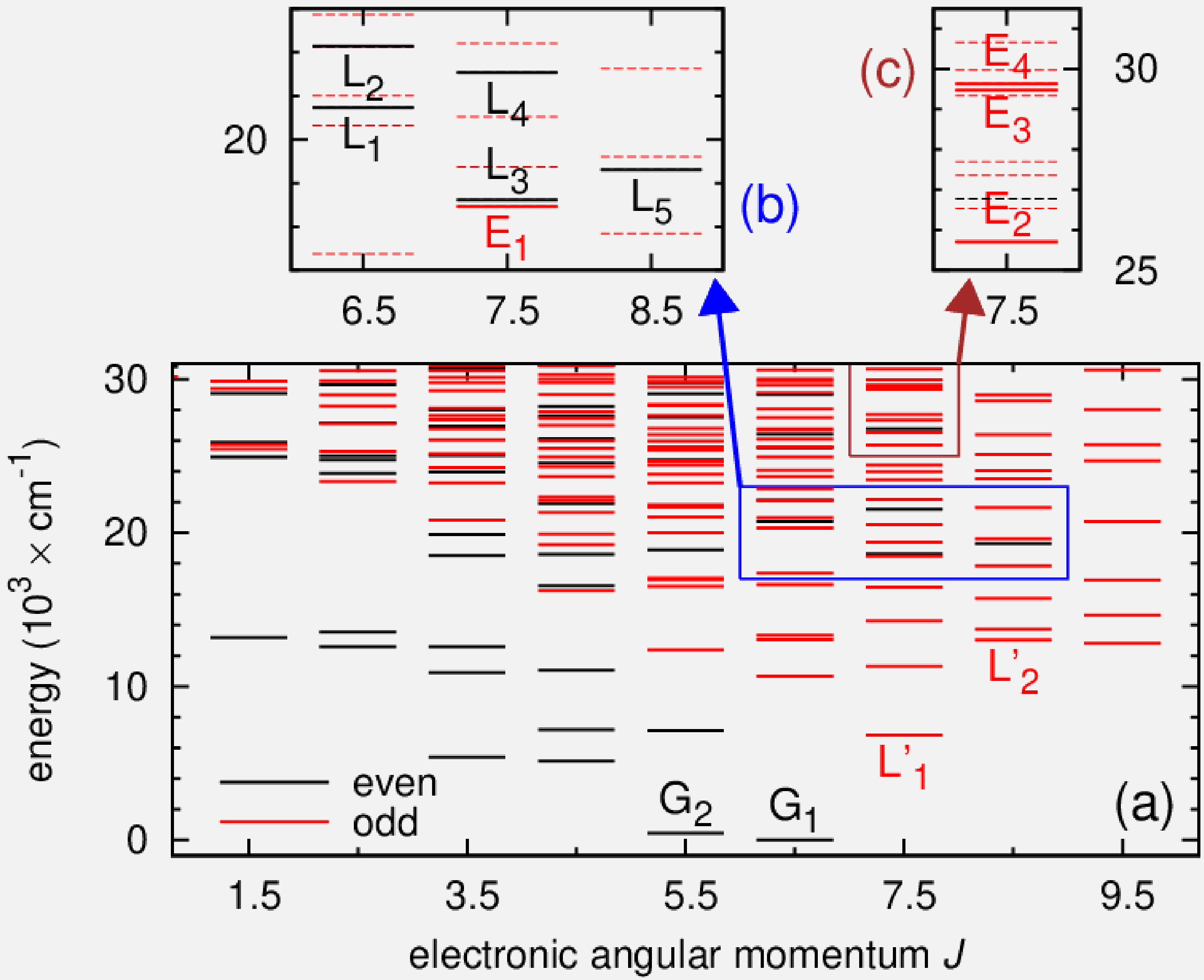
Scaling factors and standard deviations

SF	(1)	(2)	(3)
f_1	0.884 ± 0.056	0.886 ± 0.046	0.987 ± 0.081
f_2	0.877 ± 0.055	0.876 ± 0.044	0.892 ± 0.607
f_3	0.797 ± 0.088	0.797 ± 0.071	0.870 ± 0.187
f_4	0.799 ± 0.493	0.808 ± 0.394	0.857 ± 0.099
f_5	0.822 ± 0.701	0.817 ± 0.569	0.859 ± 0.179
σ_A	5.488×10^6	4.565×10^6	5.891×10^6
$\sigma_{\lg A}$	0.524	0.217	0.199

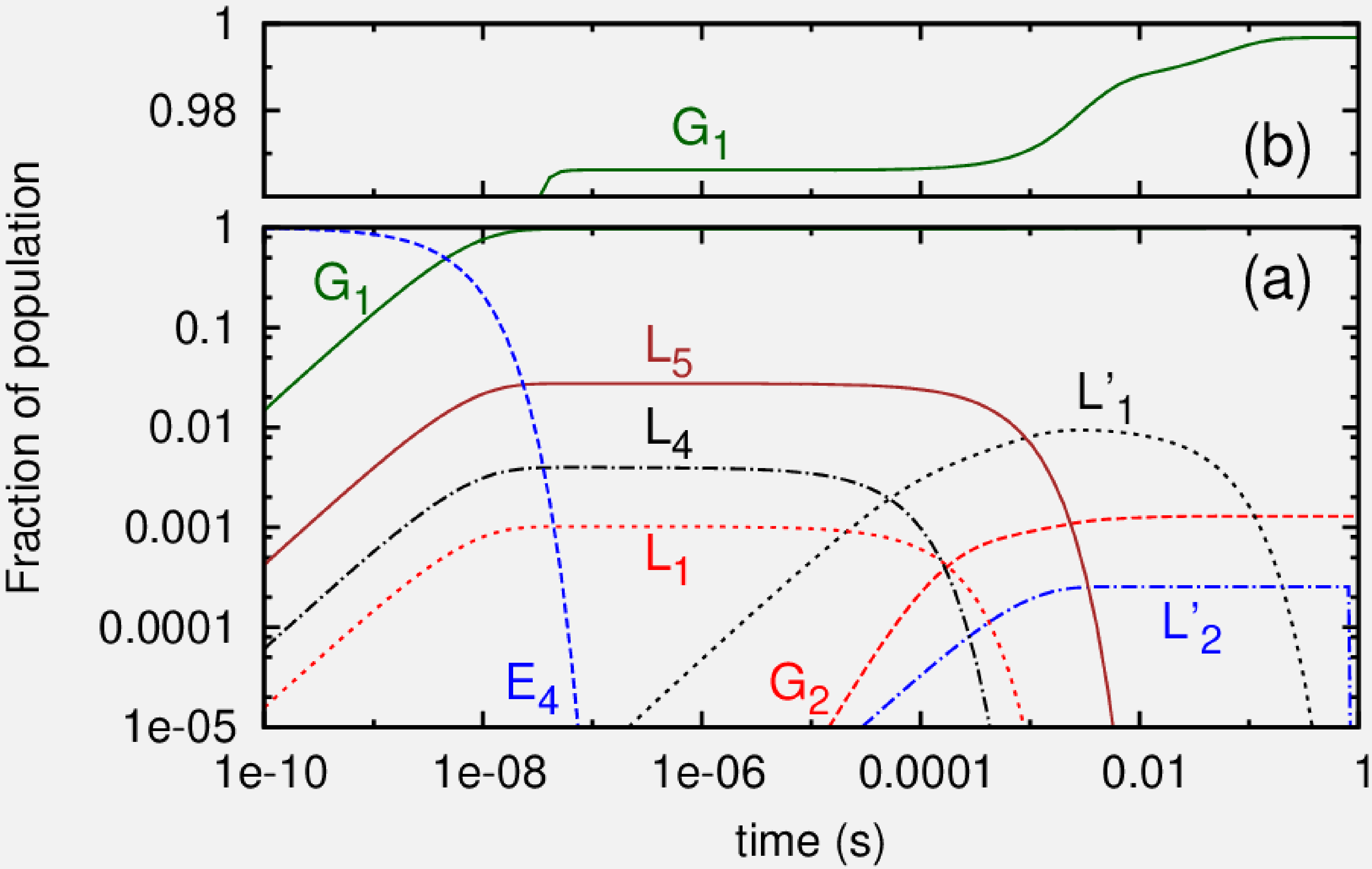
CaDDiAcS database

Wavelength (nm)	Einstein A coeff. (s^{-1})	Lower Energy (cm^{-1})	Lower Angular Momentum	Lower Leading Term	Lower Leading Percentage	Upper Energy (cm^{-1})	Upper Angular Momentum	Upper Leading Term	Upper Leading Percentage
200.7	4.286e+5	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*49849.0*	11/2	$4f^{11}(^2K^o_{13/2})5d_{5/2}$ $(13/2,5/2)^o_6$ $6s_{1/2}$ $(6,1/2)^o$	19.7
200.9	7.828e+5	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*49785.6*	11/2	$4f^{11}(^4F^o_{7/2})5d^2(^3P_2)$ $(7/2,2)^o$	22.4
201.5	2.349e+5	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*49641.7*	13/2	$4f^{11}(^4G^o_{7/2})5d_{5/2}$ $(7/2,5/2)^o_6$ $6s_{1/2}$ $(6,1/2)^o$	24.3
202.1	2.429e+6	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*49482.7*	11/2	$4f^{11}(^4G^o_{7/2})5d6s(^3D_3)$ $(7/2,3)^o$	21.1
202.5	1.152e+5	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*49406.6*	13/2	$4f^{11}(^4F^o_{9/2})5d^2(^1D_2)$ $(9/2,2)^o$	17.8
202.6	3.878e+5	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*49371.4*	11/2	$4f^{11}(^4G^o_{7/2})5d6s(^3D_3)$ $(7/2,3)^o$	15.9
202.6	2.477e+5	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*49361.0*	15/2	$4f^{11}(^4F^o_{9/2})5d^2(^1G_4)$ $(9/2,4)^o$	28.0
203.4	8.767e+5	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*49188.9*	11/2	$4f^{11}(^2K^o_{13/2})5d6s(^3D_3)$ $(13/2,3)^o$	21.4
204.0	4.585e+3	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*49035.1*	11/2	$4f^{11}(^4I^o_{9/2})5d^2(^1D_2)$ $(9/2,2)^o$	16.8
204.5	2.014e+5	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*48919.9*	11/2	$4f^{11}(^4F^o_{5/2})5d^2(^3F)$ $^3[11/2]^o$	11.1
204.6	4.240e+5	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*48889.9*	13/2	$4f^{11}(^4I^o_{9/2})5d^2(^1G_4)$ $(9/2,4)^o$	31.9
205.6	4.104e+5	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*48645.6*	11/2	$4f^{11}(^2K^o_{13/2})5d_{3/2}$ $(13/2,3/2)^o_5$ $6s_{1/2}$ $(5,1/2)^o$	19.3
206.077	4.745e+3	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	48525.656	13/2	$4f^{11}(^4I^o_{9/2})5d^2(^1G_4)$ $(9/2,4)^o$	18.0
207.2	2.968e+5	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*48274.1*	15/2	$4f^{11}(^2K^o_{13/2})5d6s(^3D)$ $^3[15/2]^o$	61.7
207.8	2.886e+4	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*48135.9*	15/2	$4f^{11}(^4F^o_{7/2})5d^2(^3F_4)$ $(7/2,4)^o$	37.2
207.8	7.864e+4	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*48132.3*	11/2	$4f^{11}(^4F^o_{5/2})5d^2(^3F_3)$ $(5/2,3)^o$	12.2
208.2	1.274e+5	0.000	13/2	$4f^{12}(^3H_6)6s_{1/2}$ (6,1/2)	98.1	*48052.5*	11/2	$4f^{11}(^4F^o_{7/2})5d^2(^3F)$ $^3[9/2]^o$	14.4
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

Laser-cooling of Er^+?



Laser-cooling of Er^+?



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