ADAS Workshop
Ringberg castle
Oct. 4 - 7, 2009

Atomic structure investigations of heavy atoms and ions

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Collaborations with :

- **THEORY:**

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- **LIF LIFETIME MEASUREMENTS :**

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Also collaboration with :

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Outline

The heavy elements and ions

Introduction

1) The lanthanides
   A. General characteristics
   B. Results
   C. The database DREAM

2) The sixth row of the periodic table
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   B. The database DESIRE

3) The fifth row of the periodic table
   Some chosen results

4) Conclusions

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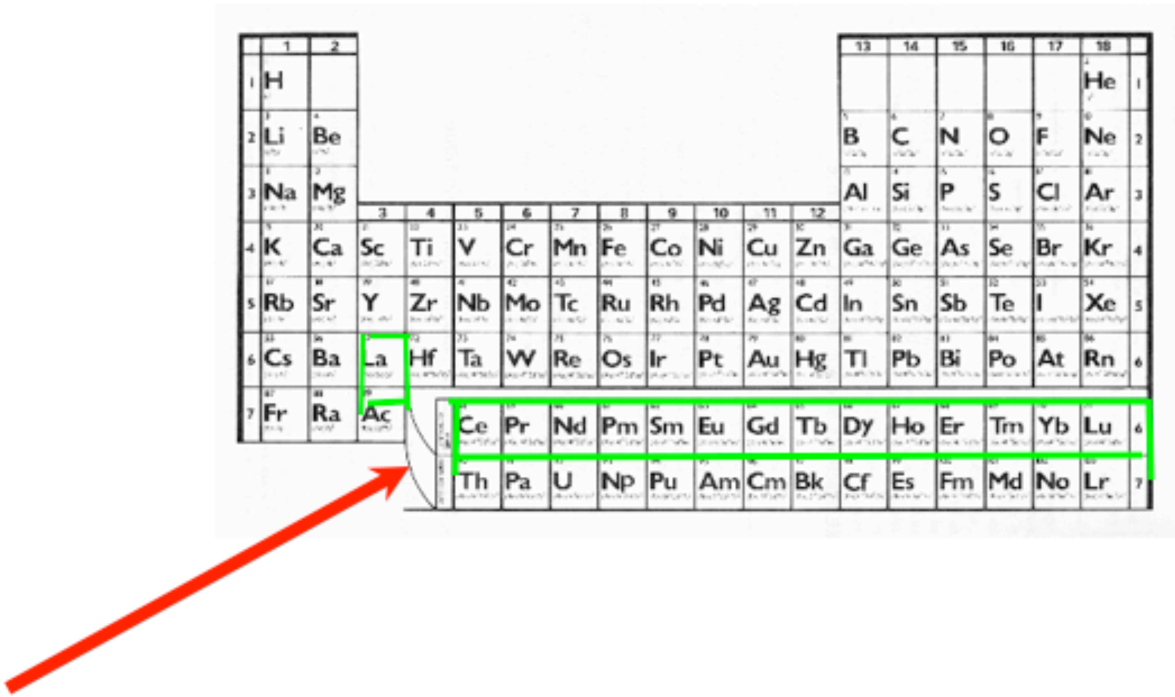
1. THE LANTHANIDES ($57 \leq Z \leq 71$)

[La, Ce, Pr, Nd, (Pm), Sm, Eu, Gd, Tb, Dy, Er, Tm, Yb, Lu]



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THE LANTHANIDES

- **Little investigated up to now**
  - Very complex spectroscopic structures with unfilled 4f shell
  - Laboratory analyses still very fragmentary or missing for many ions
  - Rather low cosmic abundances in astrophysics

- **More interest in recent years**
  - Substantial progress made in computer development
  - High resolution spectra now available in astrophysics requiring new atomic data
  - Progress in lifetime measurements with selective laser excitation

Beside their interest for astrophysics (large overabundances observed in some CP stars), these elements and ions are also interesting for solid state physics, for developing new light sources, …

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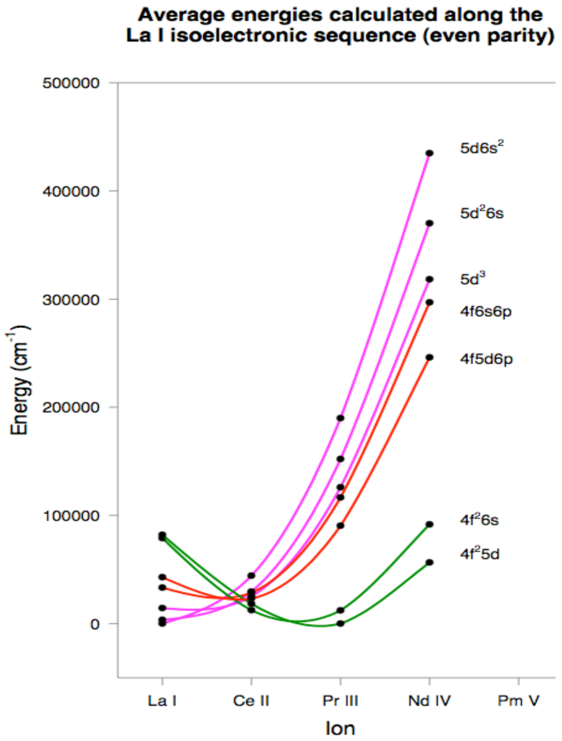
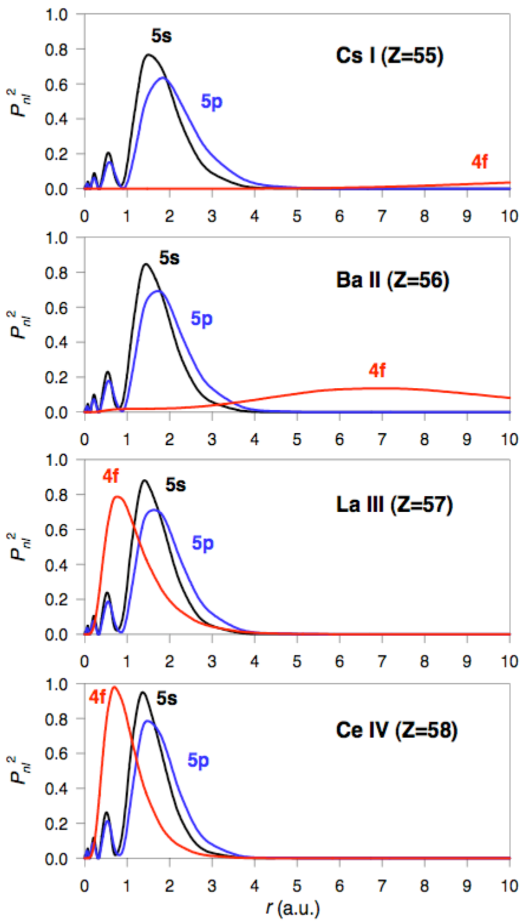
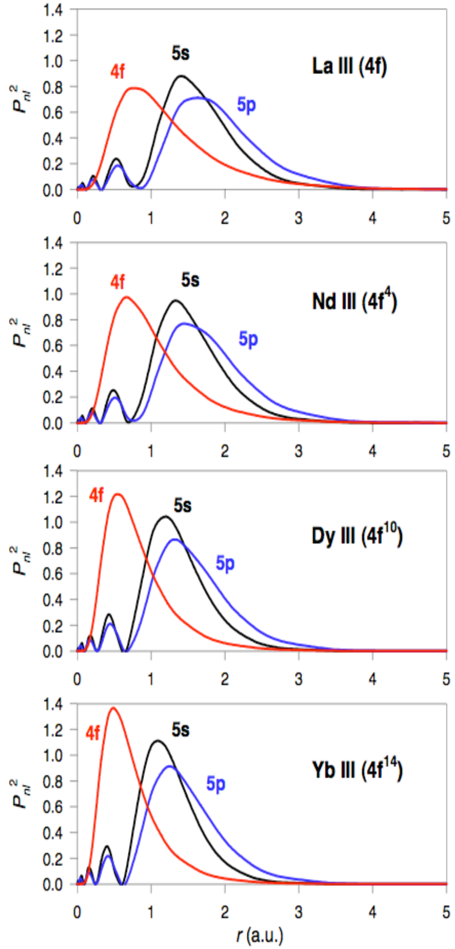
General characteristics of RE (lanthanides)







The relativistic Hartree-Fock (HFR) approximation is well adapted for investigating the atomic structure of RE ions

- Originally proposed by Cowan (1981)

- The most important relativistic effects are included (spin-orbit, Darwin term, mass-velocity contributions,..).

- The computer time needed is rather limited which is interesting for large-scale calculations.

- Convergence problems do occur very rarely. Most of them are encountered in neutrals or for very high Rydberg states.

- This is a multiconfigurational approach : CI cannot be neglected.
  Configuration interaction can be introduced in an extended way in the HFR code.

- The HFR method can be used both in an *ab initio* or in a semi-empirical way (i.e. combined with a LSF of the calculated eigenvalues to the observed energy levels). This approach however can be used only if reliable experimental energy levels are available.

- The HFR results (E_k, λ, gA,…) generally agree generally well with fully relativistic results for highly ionized atoms and with experiment for low ionization stages provided CORE-POLARIZATION EFFECTS ARE INCLUDED IN THE CALCULATIONS.

Importance of CP effects : lifetimes in Lu III (in ns)

Method	6p $^2P°_{1/2}$ 38401 cm^{-1}	6p $^2P°_{3/2}$ 44705 cm^{-1}
HF	1.49	1.00
HFR	1.57	1.06
HFR+POL	2.03	1.38
HFR+POL+PEN	2.23	1.47
Experiment[a]	2.20 ± 0.20	1.55 ± 0.20

[a] TR-LIF spectroscopy [Biémont et al. J.Phys. B32, 3409(1999)]

! Theoretical methods need to be checked by experiment !!

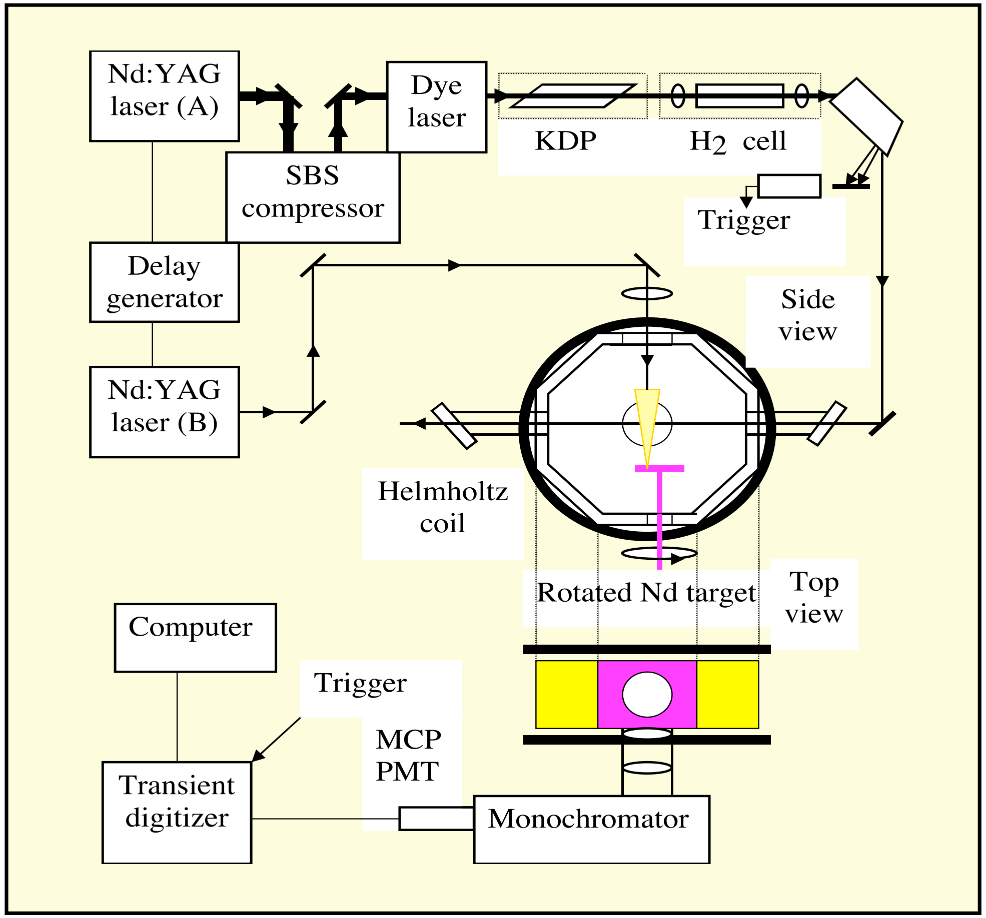
METHOD CHOSEN FOR LARGE SCALE LIFETIME MEASUREMENTS IN LANTHANIDES

(Collaboration Sweden-Belgium-China)

Time-resolved laser-induced fluorescence (TR-LIF) technique

- **Selective excitation - no cascading problem**
- **Many levels accessible through the use of different dyes**
- **Different ionization degrees can be considered in laser-produced plasmas (I, II, III, IV)**
- **Accurate lifetime measurements (within a few %)**
- **Large range of lifetime values accessible (1 - 300 ns)**
- **Lifetime measurements must be combined with BFs (theoretical or experimental)**

LLC experimental device



Summary : lifetimes measured at the LLC (Sweden)

ION	τ	CONFIGURATIONS
La I	20	$5d^26p$, 5d6s6p, 4f5d6s
La III	2	6p
Ce II	18	4f5d6p, 4f6s6p, $5d^3$
Ce IV	2	5p65d
Pr I	18	$4f^36s6p$?
Pr II	20	$4f^36p$
Pr III	8	$4f^26p$, $4f5d^2$
Nd I	15	$4f^35d6s^2$, $4f^46s6p$
Nd II	24	$4f^46p$
Nd III	5	$4f^35d$
Sm II	47	$4f^66p$?, $4f^55d6s$?
Sm III	6	$4f^55d$
Tb III	4	$4f^86p$
Dy III	5	$4f^96p$, $4f^95d$
Ho III	9	$4f^{10}6p$, $4f^{10}5d$
Er III	7	$4f^{11}6p$
Tm III	8	$4f^{12}6p$, $4f^{12}5d$
Yb II	10	$4f^{13}5d6s$, $4f^{13}6s6p$, $4f^{14}7s$
Yb III	2	$4f^{13}6p$
Lu I	26*	5d6s6p, $6s^25f$, $6s^27p$, $6s^26d$, $5d^26s$
Lu II	18*	5d6p, 6s6p
Lu III	2*	$4f^{14}6p$

Total : 276 lifetimes

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First specific example : Yb II

$4f^{14}6s\ ^2S_{1/2}$ - $4f^{14}5d\ ^2D°_{3/2,5/2}$ transitions

Electric quadrupole (E2) and magnetic dipole (M1) contributions

Lifetime value :

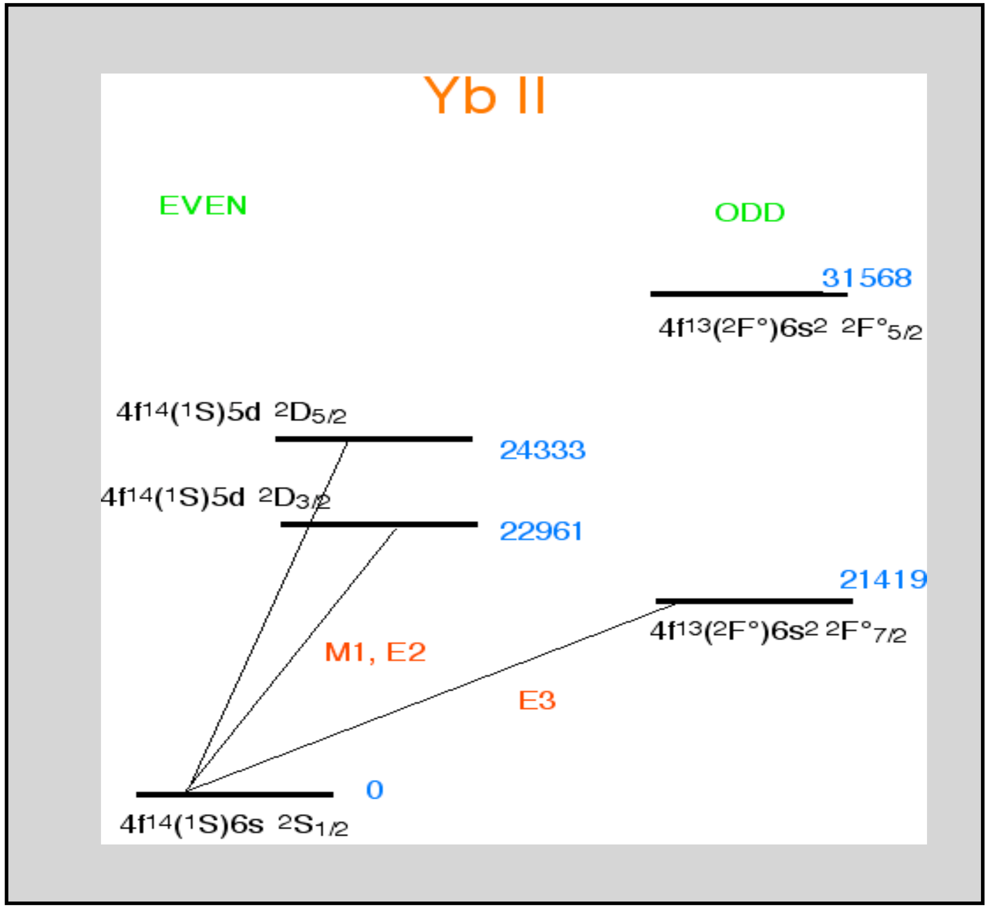
$^2D_{3/2}$

Experiment : 52.7(2.4) ms (a)
52(1) ms (b)
Theory (HFR + CPOL): 52.8 ms

$^2D_{5/2}$

Experiment : 7.0(0.4) ms (a)
7.2(0.3) ms (b)
Theory (HFR + CPOL): 6.9 ms

(a) Yu and Maleki, Phys. Rev. A61, 022507 (2000)
(b) Taylor et al., Phys. Rev. A56, 2699 (1997)



Second specific example : Ce II and Ce III

Results for Ce III

Biémont *et al.*
MNRAS **336** (2002) 1155

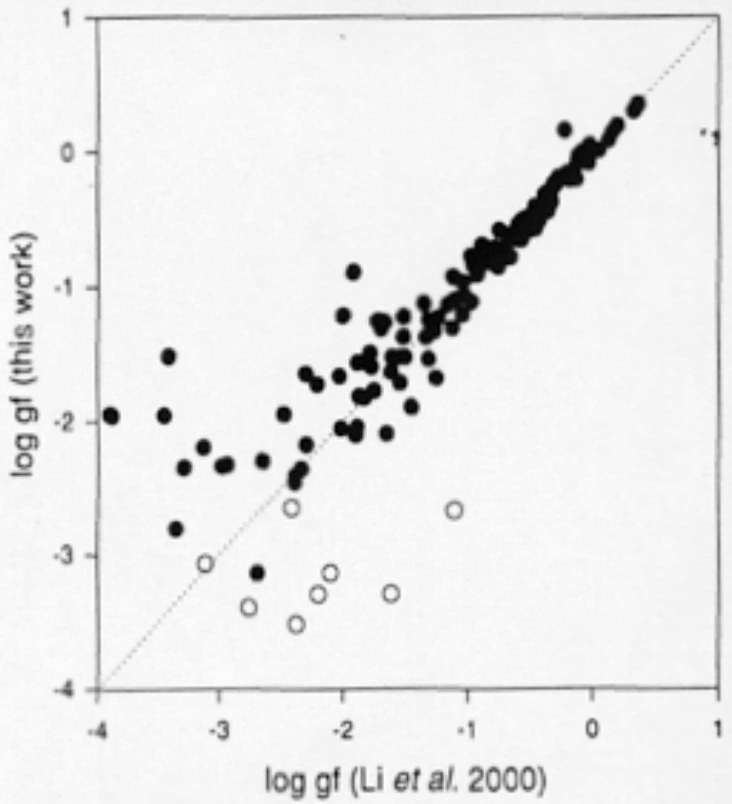


Figure 3. Comparison of the oscillator strengths obtained in this work (HFR+CPOL values) with those derived by Li et al. (2000) from their life-time measurements. The open circles correspond to transitions affected by large cancellation effects.

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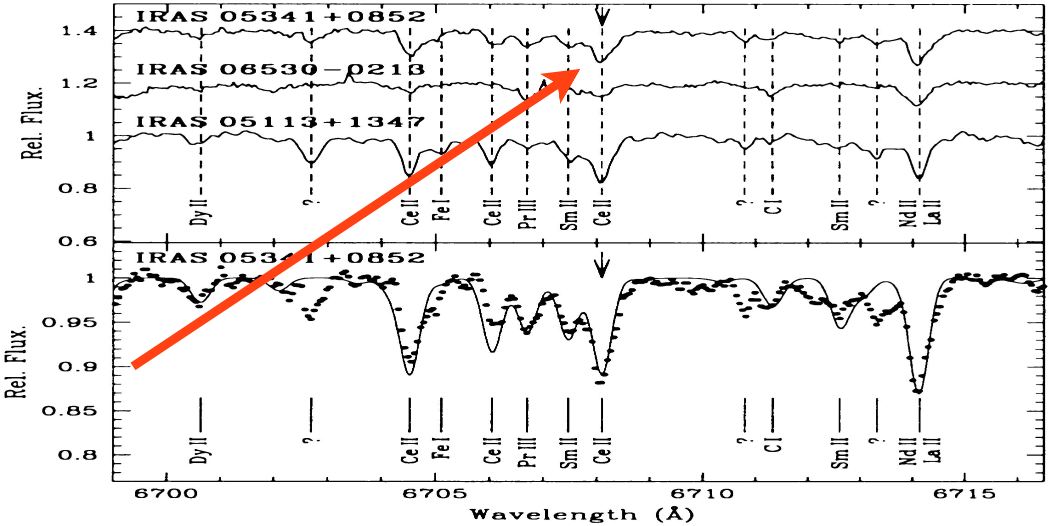
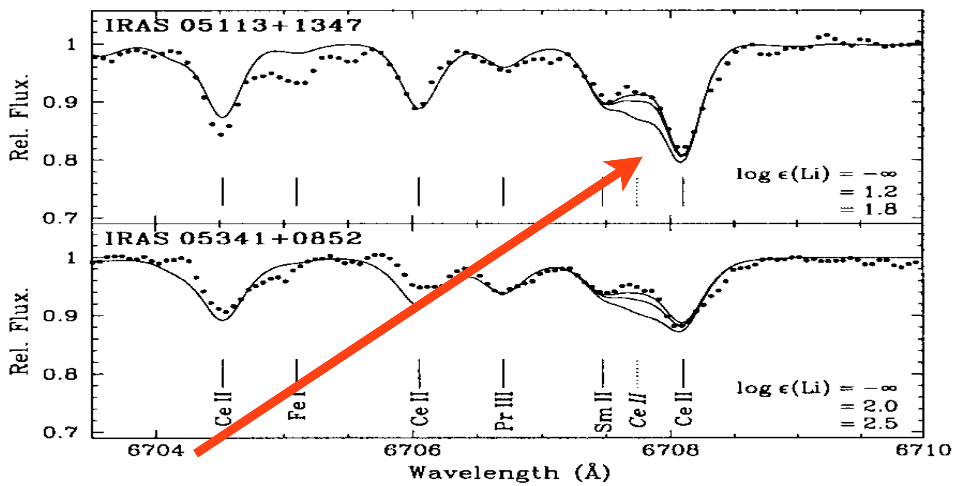
Astrophysical implications for Ce II





Cerium : The Lithium Substitute in post-AGB Stars

See : Reyniers et al. A&A 395, L35 (2002)

Third specific example : Pm II

- Pm (Z=61) has 24 unstable isotopes
- The two longest lived isotopes ^{145}Pm and ^{147}Pm have half lives ot 17.7 and 2.6 yr
- Pm first identified in the lab by Marinsky *et al.* (1947)
- Controversy regarding the possible presence of Pm in Przybylski's star (HD 101065) and in HR 465 (HD 9996)
- No transition probabilities available for Pm II
- Impossible to get experimental information
- Urgent need of theoretical radiative data for solving the astrophysical problem

- Model adopted :
$4f^45d^2 + 4f^45d6s + 4f^46s^2 + 4f^56p + 4f^6 + 4f^46p^2$ (even)
$4f^56s + 4f^55d + 4f^45d6p + 4f^46s6p$ (odd)

- CP effects introduced in the model

- Fitting procedure considered. 169 levels quoted by Martin *et al.* (1978) but configurations known for only 28 of them (23 odd and 5 even). Sd. dev. of the fits : 227 and 51 cm^{-1}.

- First A_{ki} values for 46 Pm II transitions

- Detailed results in Fivet *et al.* MNRAS, <u>380</u> (2007) 771

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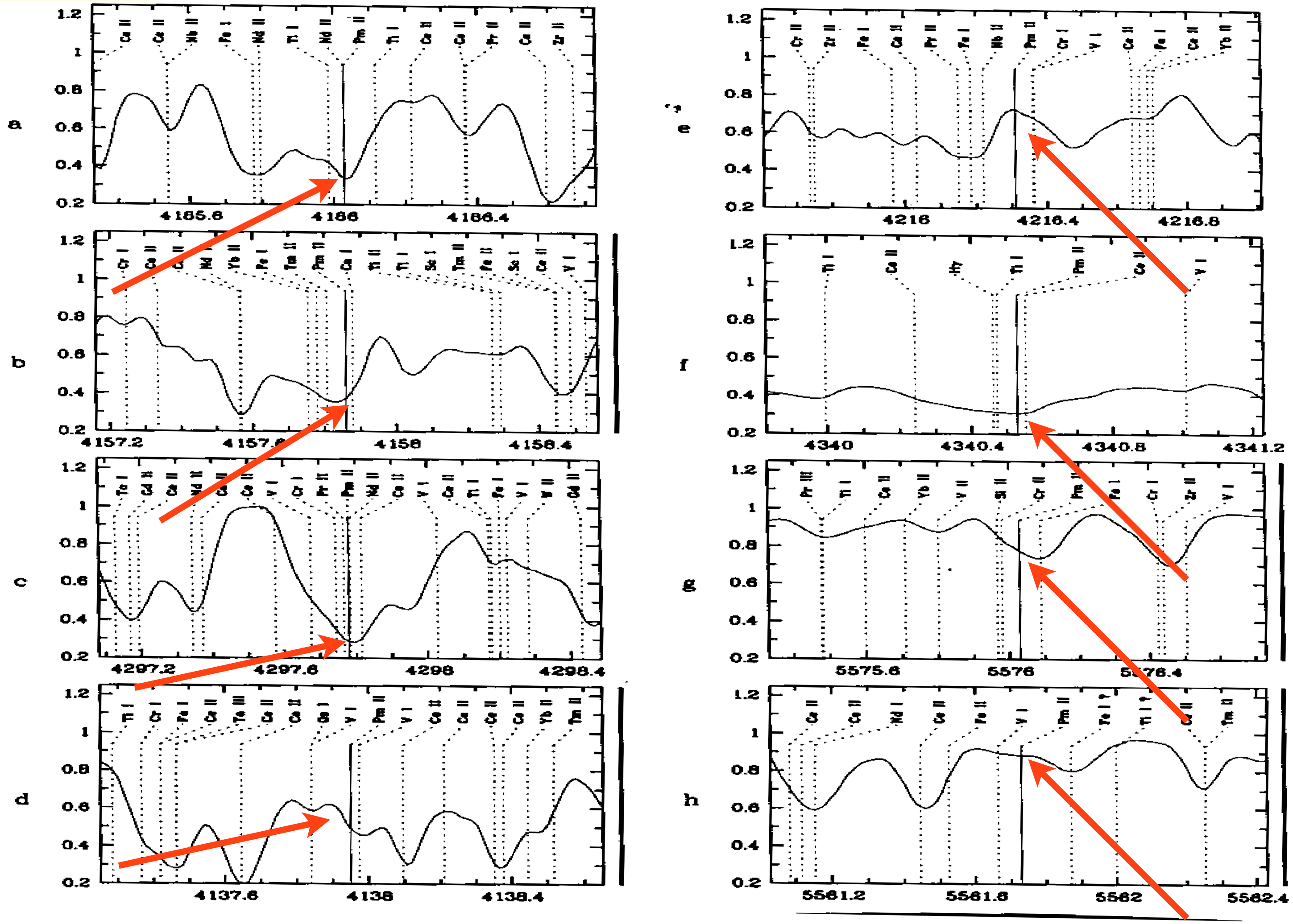
Third specific example : Pm II

Przybylski's star



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DREAM

Database on Rare Earths At Mons University

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Maintained by Pascal Quinet

- **- EASY ACCESS TO THE WEB SITE:**

http://www.umh.ac.be/~astro/dream.shtml

All the relevant references can be found on that site
that is regularly updated

- **- ALSO AVAILABLE THROUGH ANONYMOUS ftp AT THE ADDRESS:**

umhsp02.umh.ac.be/pub/ftp_astro/dream

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DREAM : Sample of results : Yb III

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Radiative transitions in Yb III (see Ref. [18] in the D.R.E.A.M. main page)

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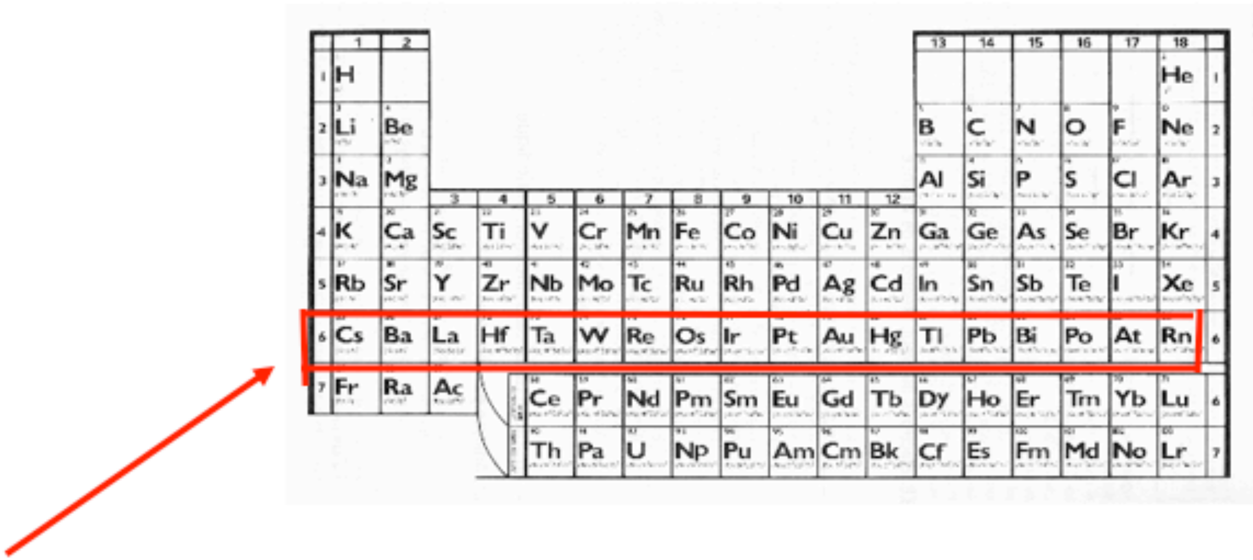
Wavelength	Lower level	Upper level	log gf	gA	CF
2073.950	88498 (e) 4.0	136700 (o) 3.0	-0.61	3.77E+08	-0.674
2078.056	72140 (e) 3.0	120247 (o) 4.0	0.16	2.21E+09	0.897
2082.464	82546 (e) 3.0	130551 (o) 3.0	-0.21	9.46E+08	-0.897
2086.288	34991 (o) 3.0	82907 (e) 2.0	-2.30	7.63E+06	-0.003
2086.534	82546 (e) 3.0	130457 (o) 2.0	0.03	1.64E+09	0.758
2087.375	39721 (o) 1.0	87613 (e) 1.0	-1.73	2.85E+07	0.071
2087.446	34656 (o) 4.0	82546 (e) 3.0	-2.52	4.67E+06	-0.083
2087.985	72487 (e) 4.0	120365 (o) 3.0	0.16	2.18E+09	0.690
2088.224	88977 (e) 2.0	136849 (o) 3.0	0.36	3.49E+09	0.940
2091.230	78183 (e) 2.0	125987 (o) 2.0	0.29	2.96E+09	-0.864
2091.841	78020 (e) 5.0	125810 (o) 4.0	-0.38	6.34E+08	0.892
2092.269	78779 (e) 3.0	126559 (o) 3.0	0.41	3.92E+09	-0.757
2093.135	72487 (e) 4.0	120247 (o) 4.0	-0.10	1.20E+09	-0.982
2094.778	88977 (e) 2.0	136700 (o) 3.0	-0.53	4.47E+08	0.145
2095.310	78020 (e) 5.0	125731 (o) 6.0	0.89	1.18E+10	0.971
2096.791	78779 (e) 3.0	126456 (o) 4.0	0.45	4.30E+09	0.899
2098.249	82907 (e) 2.0	130551 (o) 3.0	0.03	1.63E+09	-0.917
2102.132	34991 (o) 3.0	82546 (e) 3.0	-2.05	1.34E+07	-0.008

Number of transitions Listed in the database DREAM : about 65 000 transitions

2. SIXTH ROW OF THE PERIODIC TABLE

[Cs (Z=55) to Rn (Z=86)]

(Cs, Ba, Hf, Ta, W, Re, Os, Ir, Pt, Au, Hg, Tl, Pb, Bi, Po, At, Rn)



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SIXTH ROW OF THE PERIODIC TABLE

- It is necessary in stellar nucleosynthesis to clarify the relative importance of the *r* and *s* processes for the production of heavy elements in the Galaxy.

- The test of theoretical models requires accurate atomic data for the heavy elements around Z=77 (Ir)

- Tungsten belongs to this group

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Elements Cs to Rn

(Cs, Ba, Hf, Ta, W, Re, Os, Ir, Pt, Au, Hg, Tl, Pb, Bi, Po, At, Rn)

Ba II	Gurell *et al.* Phys. Rev. A75 (2007) 052506
Hf I, II, III	(in preparation)
Ta I	Fivet *et al.*, EPJD 37 (2006) 29
Ta II	Quinet *et al.* A & A (to be submitted)
Ta III	Fivet *et al.* J. Phys. B 41 (2008)015702
W II	Nilsson *et al.* EPJD 49 (2008) 13
W III	Palmeri et al. Phys. Scr. 78 (2008) 015304
Re I	Palmeri *et al.* Phys. Scr. 74 (2006) 297
Re II	Palmeri *et al.*, MNRAS 362 (2005) 1348
Os I, Os II	Quinet *et al.*, A&A 448 (2006) 1207
Ir I, Ir II	Xu *et al.*, JQSRT 104 (2007) 52
Pt II	Quinet *et al.* Phys. Rev. A77 (2007) 022501
Au I, Au II	Fivet *et al.* J. Phys. B39 (2006) 3587
Au II	Biémont *et al.* MNRAS 380 (2007) 1581
Au III	Enzonga Yoca *et al.* Phys. Scr. 78 (2008) 025303
Hg I	Blagoev *et al.* Phys. Rev. A66 (2002) 032509
Tl I	Biémont *et al.* J. Phys. B38 (2005) 3547
Pb I	Li *et al.* Phys. Rev. A57 (1998) 3443; Biémont et al. MNRAS 312 (2000) 116
Pb II	Quinet *et al.* J. Phys. B40 (2007) 1705
Bi II	Palmeri *et al.* Phys. Scr. 63 (2001) 468
Bi III	Quinet *et al.* J. Phys. B40 (2007) 1705

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Some specific examples (4) : W II, W III

9 lifetimes measured in W II and 2 in W III
- The calculated lifetimes agree well with expriment

Table 2. Comparison between measured and calculated lifetimes in W III.

Level	E (cm^{-1})	τ_{exp} (ns)	τ_{calc} (ns)
57231^o_2	57231.04	2.9±0.3[a]	2.61[c], 2.86[d]
60196^o_1	60195.86	1.78[b]	1.52[c], 1.71[d]
61488^o_3	61488.36	2.5±0.3[a]	2.13[c], 2.33[d]
62822^o_2	62821.85	2.10[b]	1.68[c], 1.88[d]
67732^o_5	67731.94	2.42[b]	1.92[c], 2.12[d]

[a] TR–LIF measurements (this work).

[b] TR–LIF measurements by Schultz-Johanning *et al* [20]. The errors are between 5% and 10%.

[c] HFR+CP(A) calculations (this work).

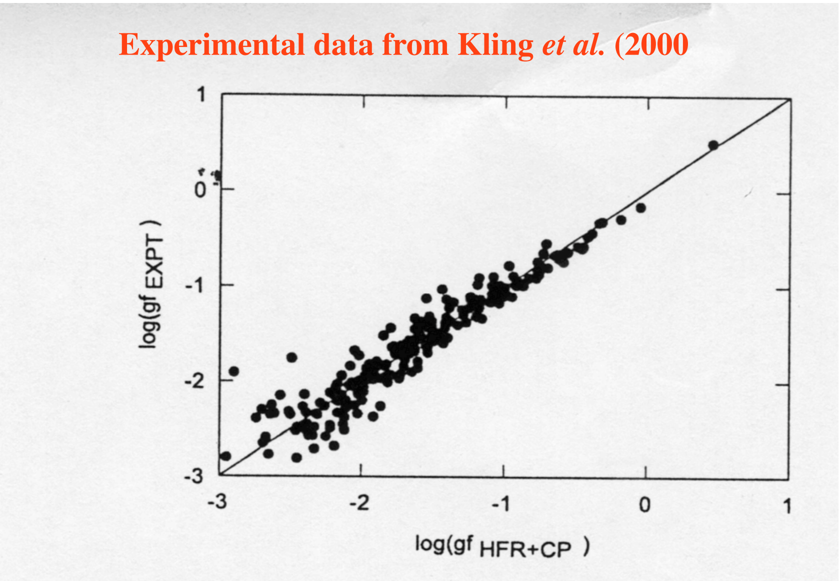
[d] HFR+CP(B) calculations (this work).

From Palmeri et al.
Phys. Scr. 78 (2008) 015304

Two specific examples : W II, W III

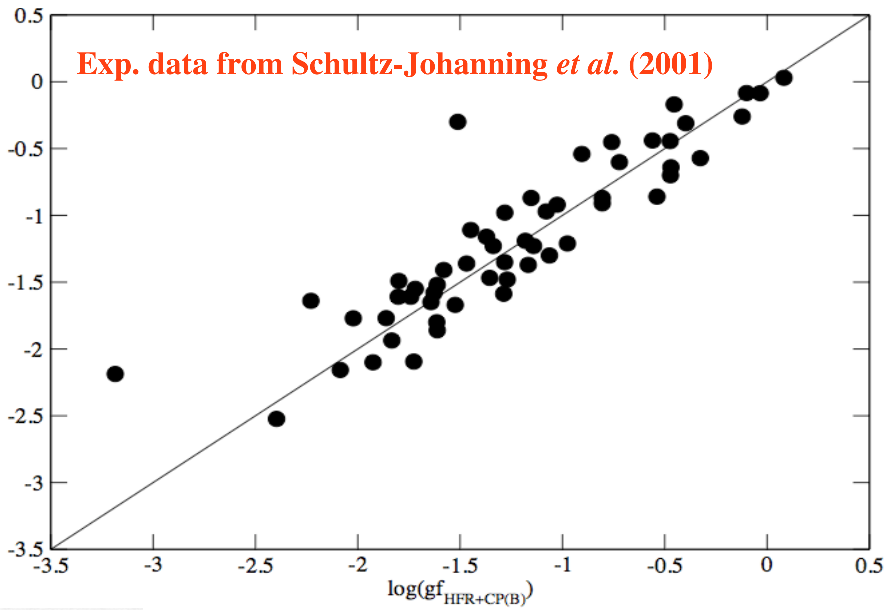
- Transition probabilities obtained for several thousands of transitions.
- They agree well with experimental results (when available)
- The gaps in these two ions are partly filled

W II



Nilsson *et al.* EPJD **49**, 13 (2008)

W III



Palmeri *et al.* Phys. Scr. **78**, 015304 (2008)

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DESIRE

DatabasE on SIxth Row Elements

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Maintained by Pascal Quinet

Address : http://w3.umh.ac.be/~astrødesire.shtml

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Radiative transitions in Os II (see Ref. [5] in the D.E.S.I.R.E. main page)

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Wavelength	Lower level	Upper level	log gf	gA	
2033.959	0 (e) 4.5	49149 (o) 3.5	-1.98	1.67E+07	NORM
2046.295	5592 (e) 1.5	54445 (o) 2.5	-2.74	2.90E+06	NORM
2070.696	3929 (e) 2.5	52206 (o) 3.5	-0.18	1.03E+09	NORM
2075.008	3593 (e) 3.5	51770 (o) 2.5	-3.19	1.00E+06	
2089.573	3929 (e) 2.5	51770 (o) 2.5	-1.63	3.55E+07	
2147.401	7892 (e) 2.5	54445 (o) 2.5	-1.12	1.09E+08	NORM
2150.447	7892 (e) 2.5	54379 (o) 3.5	-0.88	1.90E+08	NORM
2164.838	5592 (e) 1.5	51770 (o) 2.5	-0.54	4.14E+08	
2194.403	3593 (e) 3.5	49149 (o) 3.5	-0.22	8.31E+08	NORM
2210.700	3929 (e) 2.5	49149 (o) 3.5	-2.02	1.31E+07	NORM
2227.980	3929 (e) 2.5	48799 (o) 2.5	-0.49	4.33E+08	NORM
2255.853	0 (e) 4.5	44315 (o) 4.5	0.12	1.73E+09	NORM
2255.896	7892 (e) 2.5	52206 (o) 3.5	-2.60	3.30E+06	NORM
2278.319	7892 (e) 2.5	51770 (o) 2.5	-1.07	1.09E+08	
2282.278	0 (e) 4.5	43802 (o) 3.5	-0.05	1.15E+09	NORM
2313.747	5592 (e) 1.5	48799 (o) 2.5	-0.70	2.49E+08	NORM
2325.663	11460 (e) 3.5	54445 (o) 2.5	-0.36	5.43E+08	NORM
2329.235	11460 (e) 3.5	54379 (o) 3.5	-0.91	1.52E+08	NORM
2336.218	11654 (e) 2.5	54445 (o) 2.5	-1.82	1.86E+07	NORM
2336.805	3593 (e) 3.5	46373 (o) 2.5	-0.20	7.72E+08	NORM

DESIRE : Sample of data for Os II

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Radiative transitions listed in the database DESIRE

Ion	Nb of trans.	Wavelength range (Å)
Ta I	23	2526 - 6506
W II	6265	1434 - 36515
W III	4826	836 - 14940
Re I, II	available soon	
Os I	128	2137 - 8630
Os II	136	1838 - 4501
Ir I	206	2455 - 7184
Ir II	223	1777 - 4391
Au I, II	available soon	
Tl I	available soon	
Bi II	available soon	

Total : 11807 transitions (May 2009)

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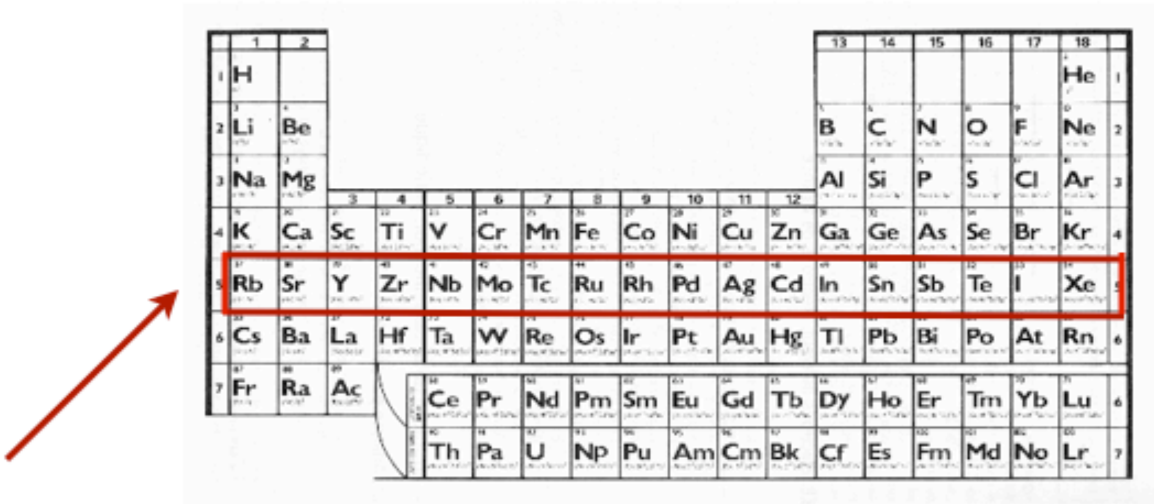
3. FIFTH ROW OF THE PERIODIC TABLE

[Ru (Z=37) to Xe (Z=54)]

(Rb, Sr, Y, Zr, Nb, Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Te, I, Xe)



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Fifth row of the periodic table

Ions considered and results obtained so far : in each case, lifetime measurement and BF calculations and/or measurements, transition probability or oscillator strength determination :

Zr I : **17** lifetimes - Malcheva *et al*. (2009)
Zr II : **16** levels, 242 transitions - Malcheva *et al*. (2006)
Nb II : **17** levels, 109 transitions - Nilsson *et al*. (to be sub.)
Nb III : calculations, 76 transitions - Nilsson *et al*. (to be sub.)
Tc II : calc. for 20 transitions - Palmeri *et al*. (2007)
Ru I : **10** lifetimes, Fivet *et al*. (2009) (MNRAS)
Ru II : **23** levels, Palmeri *et al*. (2009) (MNRAS, submitted)
Sn I : **40** levels Zhang *et al*. PRA (in press) and Xu *et al*. J. Phys. B (in press)

Work in progress : Mo II, Nb II,III, Rh II, Sb I

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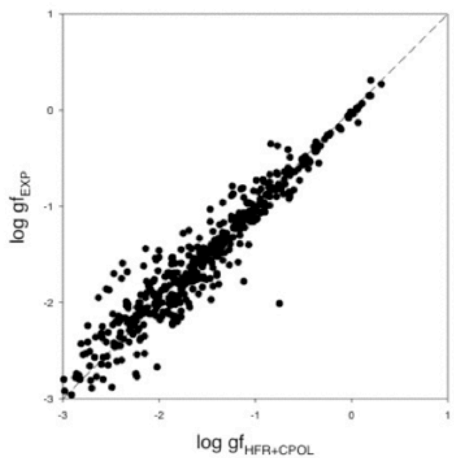
A specific example : Ru I

- In 1984, the solar abundance of ruthenium was determined by Biémont *et al.* from 9 lines of Ru I using a 1D model (Holweger & Müller 1974)
- The result : A(Ru) = 1.84 ± 0.10 (logarithmic scale) was different from the meteoritic result : A(Ru) = 1.76 ± 0.03
- The BFs were derived from the arc measurements of Corliss & Bozman (1962)
- New f values obtained for Ru I transitions in the range 2250-4710 Å. A new 3D model has been proposed by Asplund *et al.* (2009) and Trampedach *et al.* (2009)
- The new abundance value is now A(Ru) = 1.72 ± 0.12, close to the meteoritic result.
- The f values obtained for Ru II show that the lines of this ion are too weak to be observed in the sun.

Ru I [Fivet *et al.* MNRAS (2009) (in press)]



Wickliffe *et al.* 1994

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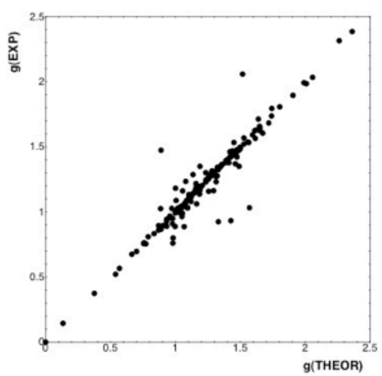

Moore's (1971) compilation

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CONCLUSIONS

- A large number of new experimental lifetimes has been obtained for RE, fifth and sixth-row elements. Theory (HFR + CPOL approach) is able to provide lifetimes for comparison and the agreement Theo.-Exp. is generally very good.

- The new results are useful in many fields of physics including astrophysics (nucleosynthesis, investigation of the chemical composition of stars,..) and also in plasma physics.

- The combination of laser lifetime (TR-LIF) measurements or lifetime calculations with BF measurements or calculations (when experimental data are missing) is useful for providing a large number of new results (A_{ki}, gf, Landé factors,…).

- Further progress is partly hindered by the poor knowledge of the energy levels and spectra.

For further references on the subject, see :

Biémont E. & Quinet P., Phys. Scr. T**105** (2003) 38
Biémont E., Phys. Scr. T**119** (2005) 55