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Simulation of Pm-Like Bismuth Spectra in an EBIT

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LIP — LABORATÓRIO DE INSTRUMENTAÇÃO E FÍSICA EXPERIMENTAL DE PARTÍCULAS
partículas e tecnologia

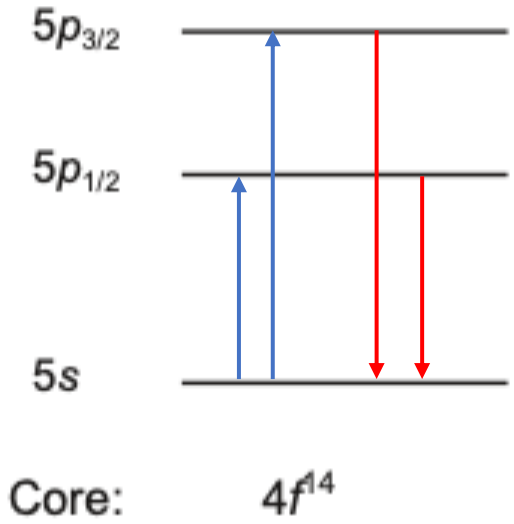
A prediction and a problem

- In 1980 Curtis and Ellis [1] theoretically predicted that Promethium-like ions have an alkali metal-like configuration, [Kr]$4d^{10}$ $4f^{14}$ 5s, for elements with $Z \geq 78$:
  - Thus, their spectra should exhibit strong 5s-5p resonance line emissions.
  - This theoretical prediction was confirmed by several other authors
- Following these predictions an effort was made to identify the prominent 5s-5p resonance
  - However, no evidence of this suggested resonance was experimentally found for W, Au, Pb, and U Pm-like ions

[1] L. J. Curtis and D. G. Ellis, Phys. Rev. Lett. 45, 2099 (1980).
[2] E. Träbert, M.J. Vilkas,and Y. Ishikawa, Journal of Physics: Conference Series 163, 012017(2009).

An experiment

- However, in 2014 Kobayashi et. al [3] showed, using an EBIT with an electron density of 10^{10} cm^{-3}, that in Pm-like Bi the 5s-5p resonance is negligible weak. But…
  - the excitation rate from the ground-state is largest for the 5p levels; this would lead to a prominent resonance, if the ground-state population is dominant.
- So, where is the population?



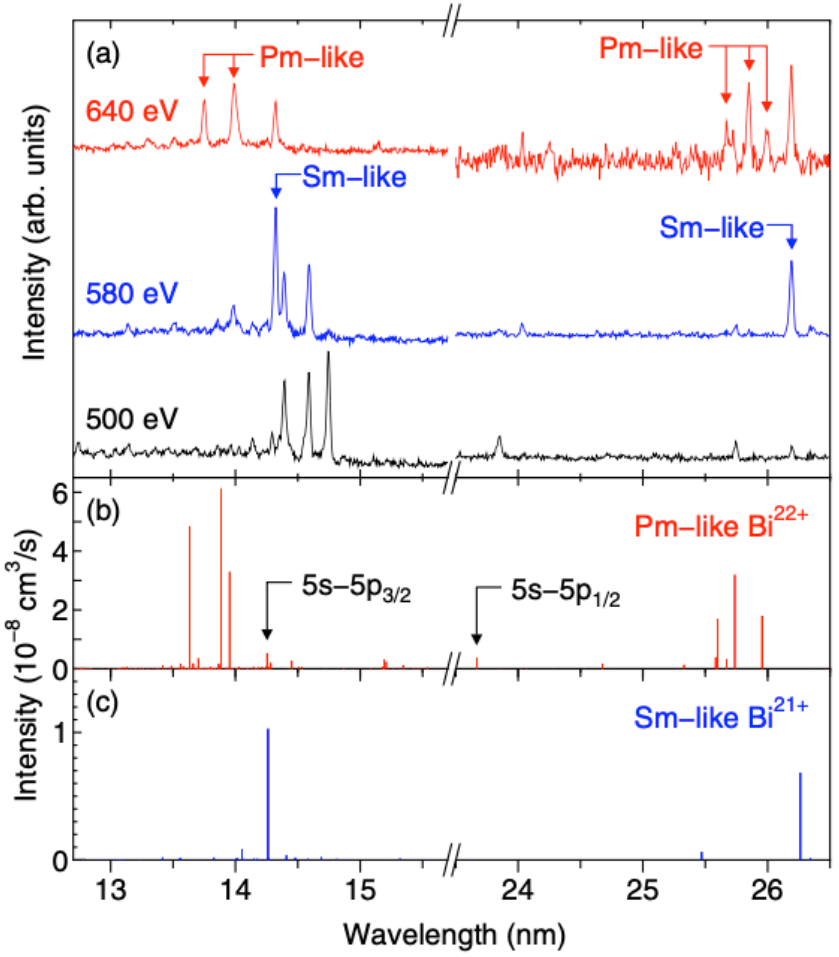
[3] Y. Kobayashi, D. Kato, H. A. Sakaue, I. Murakami, and N. Nakamura, Phys. Rev. A 89, 010501(R) (2014).

An experiment

a) Experimental spectra of Bi ions [3]
   Electron energies: 500, 580, 640 eV

b) CR model simulated spectra for 640 eV (HULLAC)

- The strongest lines observed correspond to transitions to $4f^{13}\,5s^2\,J=7/2$ metastable level ($\tau \sim 40$ s)



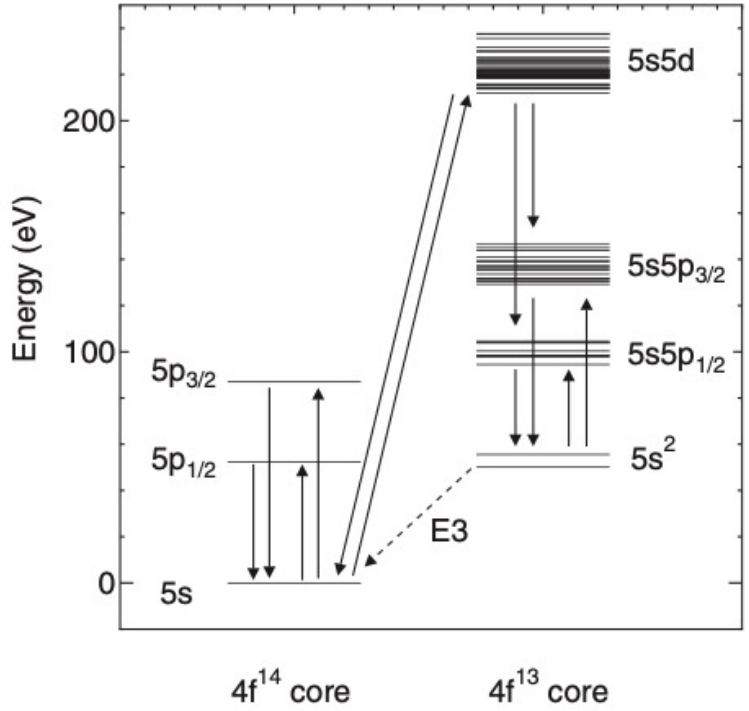
[3] Y. Kobayashi, D. Kato, H. A. Sakaue, I. Murakami, and N. Nakamura, Phys. Rev. A 89, 010501(R) (2014).

… and an explanation

In equilibrium the population can be trapped in the $4f^{13}\ 5s^2\ J=7/2$ metastable level

- leading to the loss of intensity of the 5s-5p resonance.

In 2017, the same group calculated (HULLAC) in more detail the population rates of the excited levels in Pm-like Bi, to explain this population trapping [4].



[3] Y. Kobayashi, D. Kato, H. A. Sakaue, I. Murakami, and N. Nakamura, Phys. Rev. A 89, 010501(R) (2014).

[4] D. Kato, H. A. Sakaue, I. Murakami, and N. Nakamura, Nucl. Instr. Meth. B 408, 16 (2017).

Motivation to re-visit the Pm-like case

- HULLAC calculations use relativistic wavefunctions, collision strengths are in the framework of the DWBA approximation but
  - It uses a central field parametric potential common to many levels and configurations together with 1st order perturbation theory and CI
- We used the state-of-the-art full ab-initio MCDFGME code [5] (Desclaux and Indelicato) to …
  - predict the behaviour of the system in LTE
  - synthesize the radiative spectra for 640 eV incident electron energy
    - for several electron densities in an EBIT.

[5] P. Indelicato and J. P. Desclaux, MCDFGME, a Multi- Configuration Dirac Fock and General Matrix Elements program (release 2022v3), http://kroll.lkb.upmc.fr/mcdf

Calculation ingredients

- Relativity (Dirac-Fock method)
  - Relevant for heavy ions and systems with internal inner holes.
- QED (SE & VP)
  - Precisely known only for H-like ions with point nucleus
- Electronic correlation (Multiconfiguration DF method)
  - Very hard to determine, but not very important for inner shell-transitions in heavy systems with internal holes.

Effects on atomic levels

	$Z_{eff}(nl) \leq 20$	$20 \leq Z_{eff}(nl) \leq 60$	$Z_{eff}(nl) \geq 60$
Correlation	**Dominant**	**Important**	Needed
Relativity	Needed	**Important**	**Dominant**
QED	Marginal	Needed	**Important**

$$Z_{eff}(n\ell) = Z - \sigma_{n\ell}$$

MCDHF method

- The total wavefunction is calculated with the help of the variational principle. The total energy of the atomic system is the eigenvalue of the equation

$$H_{DCB}\Psi_{\Pi,J,M}(\cdots,\mathbf{r_i},\cdots) = E_{\Pi,J,M}\Psi_{\Pi,J,M}(\cdots,\mathbf{r_i},\cdots)$$

- Dirac-Coulomb-Breit Hamiltonian

$$H_{DCB} = \sum_{a=1}^{N} h_a^D + \sum_{a=1}^{N-1}\sum_{b=a+1}^{N} V_{ab}^{CB}$$

- One electron Dirac Hamiltonian

$$h_a^D = \boldsymbol{\alpha_a}\cdot\boldsymbol{p_a} + (\beta_a - 1) + V_a$$

Electron-nucleus interaction

Dirac Matrices

MCDHF method

- Electron-electron interaction (Coulomb gauge) $V_{ab}^{CB} = V_{ab}^{C} + V_{ab}^{B} = \frac{1}{r_{ab}}$ ← Coulomb interaction

Breit interaction = Magnetic (Gaunt) + Retardation

$$- \frac{\boldsymbol{\alpha}_a \cdot \boldsymbol{\alpha}_b}{r_{ab}} \cos(\omega_{ab} r_{ab})$$

$$+ (\boldsymbol{\alpha}_a \cdot \boldsymbol{\nabla}_a)(\boldsymbol{\alpha}_b \cdot \boldsymbol{\nabla}_b) \frac{\cos(\omega_{ab} r_{ab}) - 1}{\omega_{ab}^2 r_{ab}}$$

MCDF atomic wavefunctions (ASF) are calculated in the framework of the variational principle and are written as linear combinations of configuration state functions (CSFs) describing fundamental and excited configurations

$$|\Psi_{\Pi,J,M}\rangle = \sum_{\nu} c_\nu \, |\nu \Pi J M\rangle$$

MCDHF method

- CSF's are antisymmetric products of one-electron wave functions expressed as a linear combination of Slater determinants

$$|\nu \Pi J M\rangle = \sum_i d_i \begin{vmatrix} \Phi_1^i(r_1) & \cdots & \Phi_N^i(r_1) \\ \vdots & \ddots & \vdots \\ \Phi_1^i(r_N) & \cdots & \Phi_N^i(r_N) \end{vmatrix}$$

of 4-component Dirac spinors $\boldsymbol{\Phi_{n\kappa\mu}(r) = \frac{1}{r}\begin{bmatrix} P_{n\kappa}(r)\chi_{\kappa\mu}(\theta,\phi) \\ iQ_{n\kappa}(r)\chi_{-\kappa\mu}(\theta,\phi) \end{bmatrix}}$

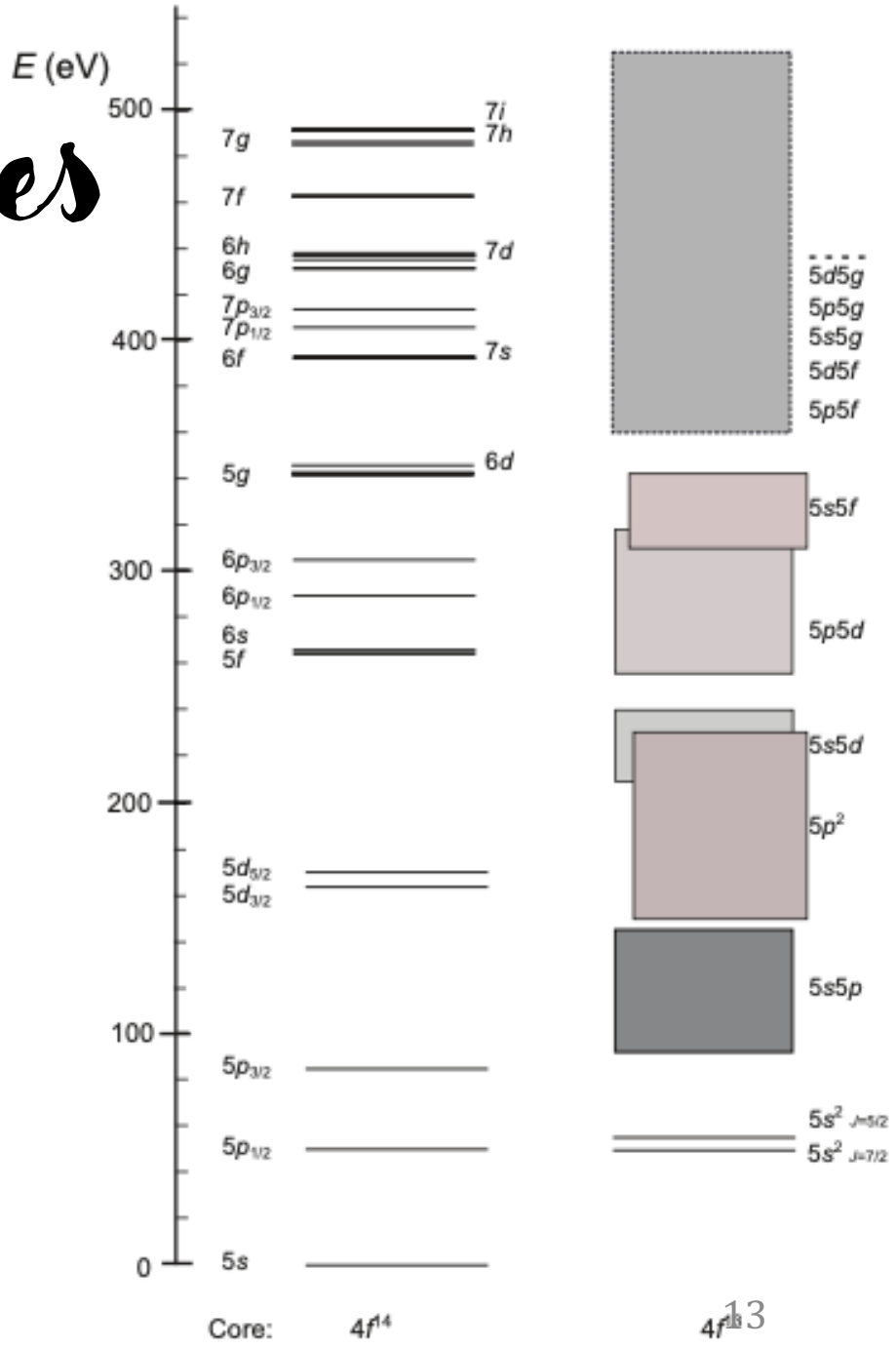
Computational cost grows rapidly with the number of CSF's included.

In summary

- **MCDFGME**
  - Uses DCB Hamiltonian and includes a full SCF treatment of electron-electron interaction (Coulomb + Breit)
  - SE and VP may be included in the SCF process
  - CSF's are calculated with full relaxation
  - Calculates both radiative and radiationless (Auger) transitions rates.
  - Calculates electron excitation cross-sections

Our calculation includes

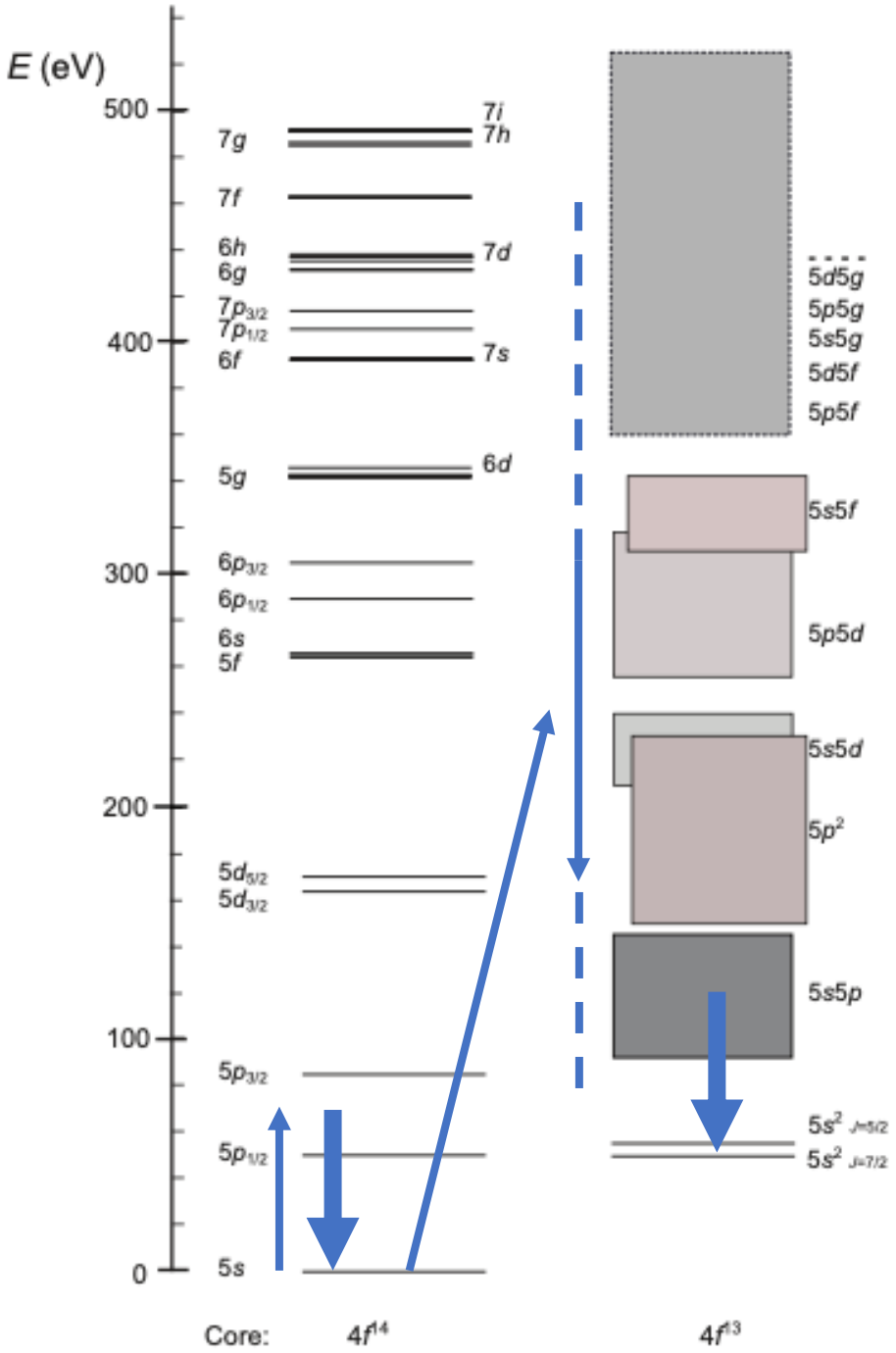
- 557 energy levels belonging to the configurations in the figure
- Rates of all possible transitions between levels
- All possible electron excitation cross-sections calculated from the $4f^{14}$ 5s ground level (using the First-Born approximation)



Calculation comparison with other authors (calc.) for...

$$4f^{13}5s5p \rightarrow 4f^{13}5s^2$$

$$4f^{14}5p \rightarrow 4f^{14}5s$$



Energies and transition wavelengths

$4f^{13}5s5p \rightarrow 4f^{13}5s^2$

- Wavelength (nm) for the most intense lines compared with Kobayashi et. al values [3]

Transition	MDFGME	HULLAC	Expt.
$5s5p_{1/2}\ J=7/2 \rightarrow 5s^2\ J=7/2$	13.54	13.63	13.77
$5s5p_{1/2}\ J=9/2 \rightarrow 5s^2\ J=7/2$	13.83	13.89	14.0
$5s5p_{1/2}\ J=5/2 \rightarrow 5s^2\ J=7/2$	13.95	13.86	
$5s5p_{3/2}\ J=5/2 \rightarrow 5s^2\ J=7/2$	25.79	25.60	25.7
$5s5p_{3/2}\ J=9/2 \rightarrow 5s^2\ J=7/2$	25.98	25.74	25.86
$5s5p_{3/2}\ J=7/2 \rightarrow 5s^2\ J=7/2$	26.17	25.96	26.01

$4f^{14}5p \rightarrow 4f^{14}5s$

- Wavelength (nm) for the 5p-5s resonance lines compared with Safronova et. al calculations [6]

Initial level	MDFGME	RMBPTB	COWAN
$5p_{1/2}$	24.76	24.686	24.719
$5p_{3/2}$	14.61	14.504	14.424

[6] U. I. Safronova, A. S. Safronova, and P. Beiersdorfer, Phys. Rev. A 88, 032512 (2013)

Electron excitation cross sections

Transition		σ (m^2)	%
$4f^{14}5s \rightarrow$	$4f^{14}5p$	4.60×10^{-22}	10.1
	$4f^{14}5d$	4.76×10^{-23}	1.04
	$4f^{14}5f$	1.16×10^{-23}	0.25
	$4f^{14}5g$	4.55×10^{-24}	0.10
	$4f^{14}6s$	1.21×10^{-23}	0.26
	$4f^{14}6p$	3.68×10^{-24}	0.08
	$4f^{14}6d$	2.19×10^{-24}	0.05
	$4f^{14}6f$	1.40×10^{-24}	0.03
	$4f^{14}6g$	8.70×10^{-25}	0.02
	$4f^{14}6h$	6.67×10^{-25}	0.01
	$4f^{14}7s$	1.49×10^{-24}	0.03
	$4f^{14}7p$	8.51×10^{-25}	0.02
	$4f^{14}7d$	6.00×10^{-25}	0.01
	$4f^{14}7f$	4.51×10^{-25}	0.01
	$4f^{14}7g$	2.86×10^{-25}	0.01
	$4f^{13}5s^2$	1.96×10^{-24}	0.04
	$4f^{13}5s5p$	2.05×10^{-23}	0.45
	$4f^{13}5s5d$	3.98×10^{-23}	0.87
	$4f^{13}5s5f$	3.93×10^{-21}	85.9
	$4f^{13}5s5g$	3.23×10^{-23}	0.71
	$4f^{13}5p5d$	0.00	0.00
	$4f^{13}5p5f$	1.71×10^{-25}	0.00
	$4f^{13}5d5f$	7.61×10^{-26}	0.00
$4f^{13}5s^2\ J = 7/2 \rightarrow$	$4f^{13}5s5p$	1.71×10^{-21}	93.1
	$4f^{13}5s5d$	9.36×10^{-23}	5.1
	$4f^{13}5s5f$	2.32×10^{-23}	1.3
	$4f^{13}5s5g$	8.95×10^{-24}	0.5
	$4f^{13}5s^2$	2.94×10^{-25}	0.02
$4f^{13}5s^2\ J = 5/2 \rightarrow$	$4f^{13}5s5p$	8.54×10^{-22}	87.2
	$4f^{13}5s5d$	9.35×10^{-23}	9.6
	$4f^{13}5s5f$	2.30×10^{-23}	2.3
	$4f^{13}5s5g$	9.01×10^{-24}	0.9

- Excitations to $4f^{13}$ 5s5f are 2-5 orders of magnitude higher than to other configurations
  - This is mainly due to the excitation to $4f^{13}$ 5s5f J=1/2 levels (see table below)
    - In a comparison with other authors there are differences that still need to be investigated.

MCDFGME (this work)		FAC (this work)		HULLAC	
ΔE	σ	ΔE	σ	ΔE	σ
311.86	8.23×10^{-25}	310.52	6.00×10^{-24}	312.2	2.43×10^{-25}
313.11	3.29×10^{-26}	312.67	1.73×10^{-23}	314.3	4.42×10^{-25}
315.46	2.21×10^{-23}	315.27	3.40×10^{-23}	317.2	1.46×10^{-25}
317.29	1.25×10^{-25}	315.52	1.27×10^{-25}	317.7	3.18×10^{-25}
319.70	6.57×10^{-25}	321.21	1.83×10^{-23}	323.7	5.77×10^{-25}
342.28	3.90×10^{-21}	335.68	2.07×10^{-22}	349.2	6.95×10^{-23}

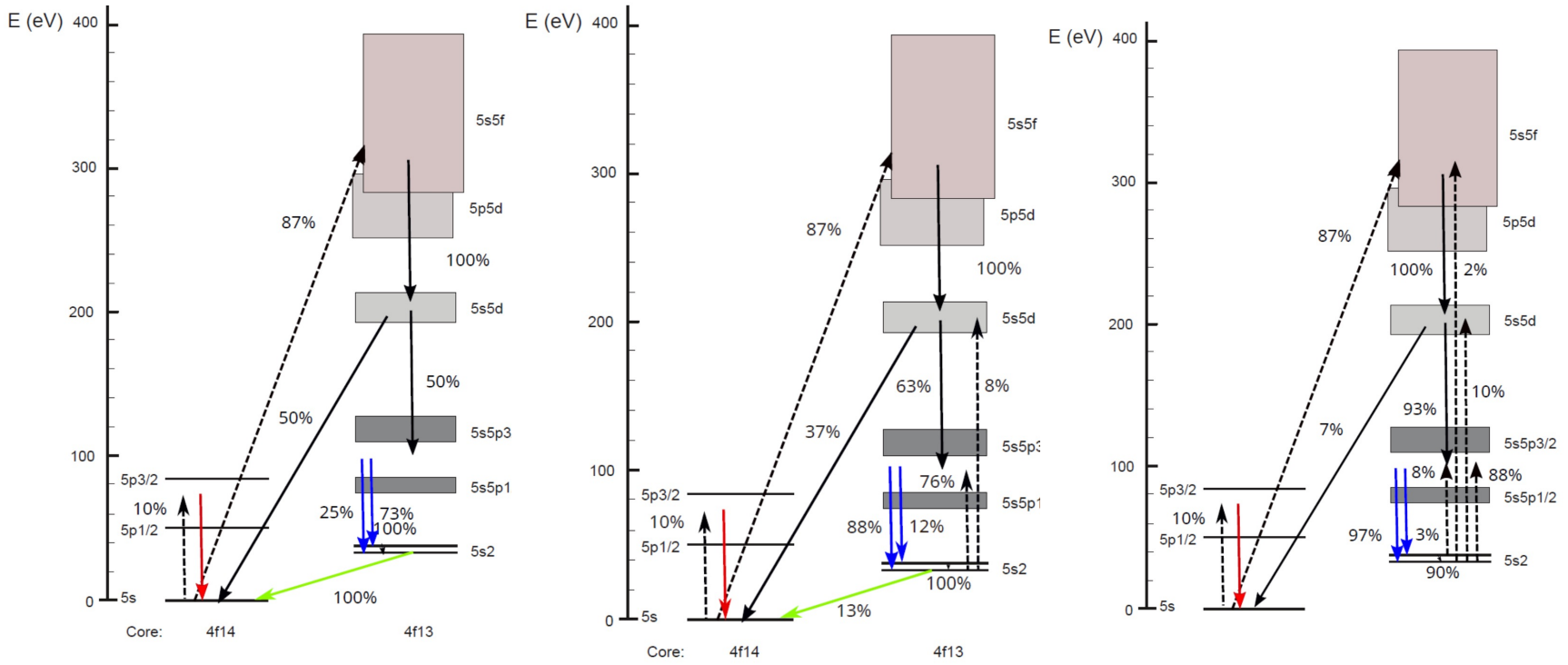
Equilibrium equations

- To obtain the populations we solved a system of 115 equilibrium balance equations, for all levels of the $4f^{14}5s$, $4f^{13}$ $5s5d$, $4f^{13}5s5p$ and $4f^{13}5s^2$ configurations.
- For example, the equation for each of the 24 levels α of the $4f^{13}$ $5s5p$ configuration is

$$
\begin{aligned}
0 \;=\; & n_e v \sigma^{A\to C\alpha} N_A + n_e v \sum_{\alpha'=1}^{24} \sigma^{C\alpha'\to C\alpha} \\
& + n_e v \sigma^{D_{J=5/2}\to C\alpha} N_{D_{J=5/2}} + n_e v \sigma^{D_{J=7/2}\to C\alpha} N_{D_{J=7/2}} \\
& + \sum_{\alpha''=1}^{39} R^{B\alpha''\to C\alpha} N_{B\alpha''} \\
& - \left(R^{C\alpha} + n_e v \sum_{\alpha'=1}^{24} \sigma^{C\alpha\to C\alpha'} \right) N_{C\alpha}. \qquad (2)
\end{aligned}
$$

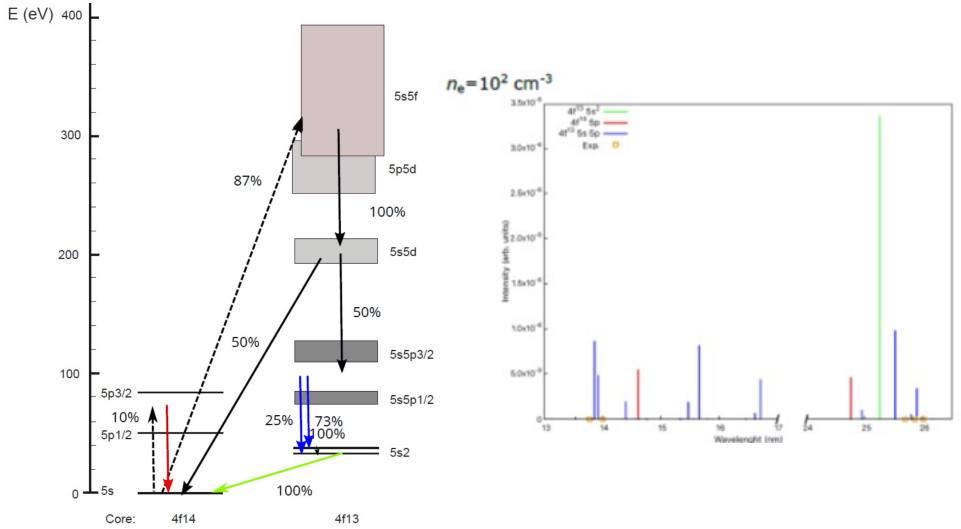
n_e v - electron density and incident electrons velocity
σ, R – electr. exc. cross-sect. and rad. transition prob.
N – level population
i, j – level indexes
A,B,C,D – for $4f^{14}5s$, $4f^{13}$ $5s5d$, $4f^{13}5s5p$ and $4f^{13}5s^2$ confs.

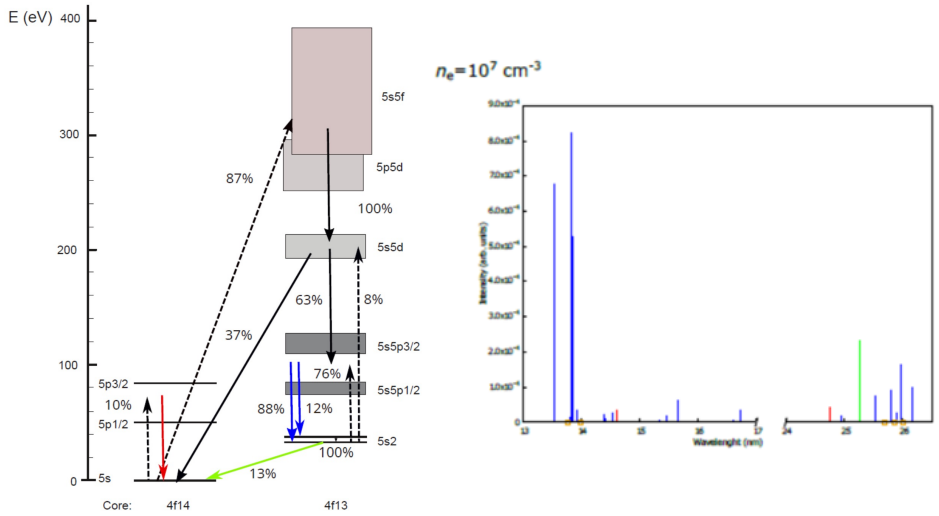
Equilibrium populations (n_e)

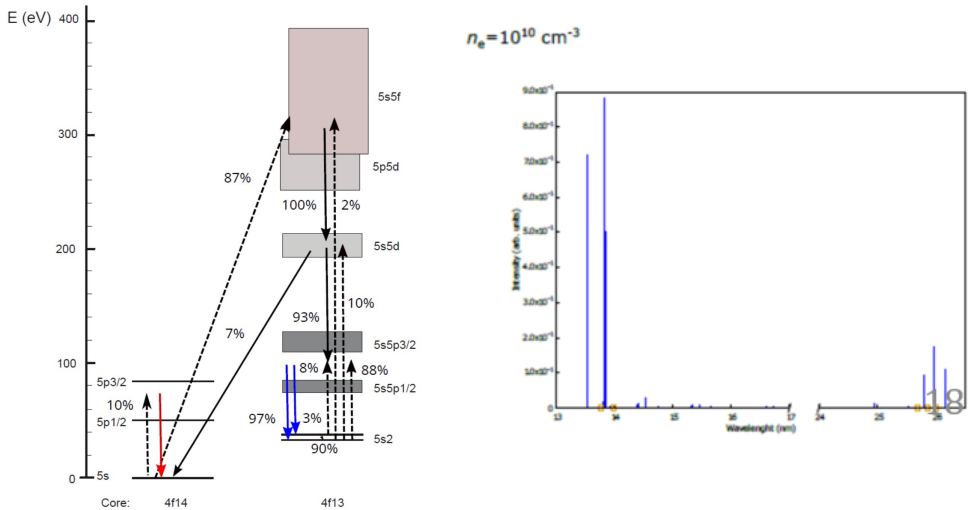


10^2 cm^{-3} 10^7 cm^{-3} 10^{10} cm^{-3}

- The numbers next to each arrow give the relative weight (in %) of the paths from each level.
- Ions in $4f^{13}5s5f$ will cascade down until they reach $4f^{13}5s^2$ or the ground level.
- 5s5d decay to ground decreases as n_e increases







- Main processes in the system for three values of n_e and corresponding synthesized spectra .
- The line colours in the spectra match the corresponding transition arrows in the leftmost diagrams.
- $4f^{13}5s^2$ J=7/2 is metastable (τ=45 s)
  - Population is trapped here
  - leading to the loss of intensity of the 5s-5p resonance.

Conclusions

- The synthesized spectra obtained from this calculation agree with the experimental data of Kobayashi et al. [3] for $n_e=10^{10}$ cm^{-3} but not for $n_e < 10^7$ cm^{-3}
- We predict that
  - **87%** of the ions will be excited to $4f^{13}$ 5s5f
  - only **10%** to $4f^{14}$ 5p
  - 1.5% will be excited to $4f^{14}$ *nl*
  - 1.5% to other levels
  - Excitation rate from ground level to $4f^{13}$ 5s*n*l will be 599.1 s^{-1}
    - This is much higher than the value calculated by Kato et. al (54.2 s^{-1}). This is due to the anomalous excitation cross section to $4f^{13}$ 5s5f J=1 levels.
- Excitation rate to $4f^{14}$ 5p (69 s^{-1}) is closer to the value of Kato (86.2 s^{-1}) ($n_e=10^{10}$ cm^{-3})

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