Atomic structure and radiative data calculations for heavy elements of interest in fusion plasma research

Theoretical challenges and recent advances

Pascal Quinet
quinet@umons.ac.be

Introduction – Motivations

Fifth row Sixth row Lanthanides
4f^6 5d^1 4f^6 5d^1
4f^6 5d^1 4f^6 5d^1
4f^6 5d^1 4f^6 5d^1
... ... ...

Very complex spectroscopic structures (unfilled 4d, 5d, 4f shells)
→ little investigated up to now (both theoretically and experimentally)

Interest in many other fields:
- Astrophysics (CP stars, nucleosynthesis, ...)
- Solid state physics (doped crystals, ...)
- New light sources (lasers, lamps, ...)
- Plasma physics (fusion, ITER, ...)

Progress in computers & laser spectroscopy
→ New systematic investigations possible

Determination of atomic data

Theoretically and experimentally...

Atomic structure parameters
- Energy levels, E_k
- Transition energies, A_k
- Wavelengths, λ_k

Radiative parameters
- Transition probabilities, A_{ij}
- Oscillator strengths, f_{ij}
- Radiative lifetimes, τ_k = 1/λ_k A_{ij}

Theoretical approach

Relativistic Hartree-Fock method (HFR)

Based on the Schrödinger equation (atom with N electrons)

\[ H \Psi = E \Psi \]

with

\[ H = \sum_{ij} \left[ \frac{\hbar^2}{2m} \frac{\partial^2}{\partial x_i \partial x_j} + \frac{1}{4\pi \epsilon_0} \sum_{k \neq i} \frac{e^2}{r_{ij}} \right] \]

Central field approximation

\[ \phi_{\text{cusp}}(\sigma) = \frac{1}{2} \phi_{\text{2p}}(\sigma) \prod_{i=1}^{N} (\sigma_i) \prod_{i=1}^{N} (\sigma_i) \]

Slater determinant

\[ \Psi(q_1,q_2,...,q_N) = \frac{1}{\sqrt{N!}} \phi_1(q_1) \phi_2(q_2) ... \phi_N(q_N) \]

Relativistic Hartree-Fock method equations

\[ \left\{ \begin{array}{l}
\frac{\hbar^2}{2m} \frac{\partial^2}{\partial x_i \partial x_j} + \frac{1}{4\pi \epsilon_0} \sum_{k \neq i} \frac{e^2}{r_{ij}} \\cr
E_k = \sum_k \left[ \int \Psi^* \left\{ \frac{\hbar^2}{2m} \frac{\partial^2}{\partial x_i \partial x_j} + \frac{1}{4\pi \epsilon_0} \sum_{k \neq i} \frac{e^2}{r_{ij}} \right\} \Psi \right] \end{array} \right. \]

Resolution of Hartree-Fock equations

Iterative method → self-consistent field method
**Theoretical approach**

Relativistic Hartree-Fock method (HFR)

Multiconfiguration approach

\[
\Psi_f = \sum_n \psi_n
\]

Relativistic effects
- Included perturbationally (spin-orbit, mass-velocity, Darwin term)
- Good agreement with fully relativistic methods

Ab initio or semi-empirical approach
- Experimental energy levels can be used to optimize the radial parameters

Core-polarization effects (HFR + CPOL)
(see e.g. Quinet et al., M.N.R.A.S. 307, 514, 1999; Quinet et al., J. Alloys Compd 344, 255, 2002)

Intravalence correlation considered within a configuration interaction scheme

Core-valence correlation represented by a core-polarization model potential depending on two parameters (dipole polarizability \(\alpha_d\) and cut-off radius \(r_c\))

\[
V_{cc} = -\frac{1}{2} \sum_n \frac{\alpha_d}{(r' + r_c)^{1/2}} P_n(r')
\]

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\]

Correction to the dipole radial integral

\[
\int P_n(r) r P_n(r) \, dr \quad \text{replaced by} \quad \int P_n(r) r \left[1 - \frac{\alpha_d}{(r + r_c)^{1/2}} \right] \, dr
\]

Semi-empirical optimization

Radial parameters (average energies, electrostatic integrals, spin-orbit parameters) adjusted in order to minimize the discrepancies between computed Hamiltonian eigenvalues and experimental energy levels

- Optimization of the transition energies and wavefunctions
- Optimization of the radiative decay rates

\[
\lambda_i = \frac{64\pi^2 |\alpha_i|}{3h} \left(\frac{\Delta E_i}{2J_i + 1}\right)^{1/2} \left|\langle \alpha_i | V | \beta_i \rangle\right|^{1/2}
\]

Radioactive lifetimes for \(6p^5 \text{P}_{1/2}\) (38401 cm\(^{-1}\)) and \(6p^5 \text{P}_{3/2}\) (44705 cm\(^{-1}\))

\[
\Delta E_i = 2.0621 \times 10^{-11} \text{cm}^{-1} \left|\langle \alpha_i | V | \beta_i \rangle\right|^{1/2}
\]

- A good experimental knowledge of the level structure for the atom (or ion) considered is needed

**Experimental measurements**

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

Accurate measurements of radiative lifetimes (within a few %)
- Lifetime range from 1 ns to 300 ns
- Selective excitation (no cascading problems)
- Many levels accessible (using different laser dyes)
- Different ionization degrees accessible in the laser-produced plasma (neutral, singly-, doubly- and trebly-ionized atoms)
Radiative lifetimes, transition rates (fifth row)

Mo II: lifetimes measured for 16 energy levels belonging to 4d 3p 5/2 D) 5p z (sixth row)

Some recent results: Nb II, Mo II, Rh II

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The specific case of tungsten

Great interest in plasma physics

Fusion reactors

ITER

International Thermonuclear Experimental Reactor
The specific case of tungsten (W I, W II, W III)

Transition rates in W I, W II and W III


W II: 263 experimentally known energy levels within the 5d6s, 5d6s6p, 5d6p and 5d6s6p configurations (Kramida & Shrih, J. Phys. Chem. Ref. Data 35, 423, 2006)

W III: 235 experimentally known energy levels within the 5d6s, 5d6s6p, 5d6p and 5d6s6p configurations (Gleines et al., J. Res. Natl Inst. Stand. Tech. 94, 221, 1989)

Transition probabilities and oscillator strengths calculated for:

- The specific case of tungsten (W IV, W V, W VI)
- Transition rates in W IV, W V and W VI

Critical evaluation of available data

- New decay rates for forbidden lines
- New radiative data (lifetimes, transition probabilities, oscillator strengths) obtained for a large number of lines belonging to heavy neutral and lowly ionized atoms (fifth row, sixth row, lanthanides)

Wavefunction purities (in LS coupling) for W I odd-levels below 45 000 cm⁻¹ (bold lines represent purities smaller than 15%)

Conclusions

- New radiative data (lifetimes, transition probabilities, oscillator strengths) obtained for a large number of lines belonging to heavy neutral and lowly ionized atoms (fifth row, sixth row, lanthanides)
- New data very useful in astrophysics, plasma physics, ...
- Level structure still too poorly known for many ions to perform semi-empirical calculations (new term analyses needed)
Atomic structure calculations
Astrophysics & Spectroscopy, UMONS, Belgium (* also ULg, Liège, Belgium)
(E. Biémont*, V. Fivet, P. Palmeri, P. Quinet*)
Department of Physics, University of Brazzaville, Congo
(S. Enzonga Yoca)
Experimental measurements
Department of Physics, Lund University, Lund, Sweden
Department of Physics, Jilin University, Changchun, China
(Z. Dai, L. Han, Z. Jiang, P. Li, Z. Ma, G. Sun, S. You, J. Xu, W. Zhang, Y. Zhang)
Also collaboration with
Institute of Solid State Physics, Bulgarian Academy of Sciences, Sofia, Bulgaria
(K. Blagoev, G. Malcheva)
Faculty of Physics, Universidad Complutense de Madrid, Spain
(R. Mayo, M. Ortiz)

Thank you for your attention!