

MCDF Level Structure and Isotope-Shift Calculations for Medium and Heavy Elements

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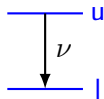
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Outline

1. Isotope-Shift Theory
2. Multi-Configuration Dirac-Fock (MCDF) Method
3. Results
4. Atomic Physics
5. Conclusions

Isotope-Shift Theory

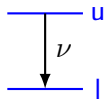
- Observed transition frequency isotope dependent
- Extraction of nuclear parameters
- Transition isotope shift parametrized by



$$\delta\nu^{AA'} = \left(M^{\text{NMS}} + M^{\text{SMS}} \right) \frac{A - A'}{AA'} + F \delta \langle r^2 \rangle^{AA'} \quad A > A'$$

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- Field-shift due to change in charge distribution

$$\delta\nu^{\text{FS}} = F \delta \langle r^2 \rangle^{AA'}$$

Isotope-Shift Theory

Mass Shift

- Transition isotope shift parametrized by

$$\delta\nu^{AA'} = \left(M^{\text{NMS}} + M^{\text{SMS}} \right) \frac{A - A'}{AA'} + F\delta \langle r^2 \rangle^{AA'} \quad A > A'$$

- Mass shift due to nuclear recoil

$$H = \frac{1}{2M} \sum_{ij} p_i p_j$$

- Separation into normal and specific mass shift
- Normal mass shift

$$H = \frac{1}{2M} \sum_i p_i^2 \quad \delta\nu^{\text{NMS}} \approx \nu^\infty \frac{m_e}{M}$$

- Two-electron contribution gives rise to specific mass shift

$$H = \frac{1}{2M} \sum_{i < j} p_i p_j$$

Calculation of Isotope Shifts

- Mass Shift parameters

$$M_{\text{NMS}} \propto \langle \psi | H_{\text{NMS}} | \psi \rangle \qquad M_{\text{SMS}} \propto \langle \psi | H_{\text{NMS}} | \psi \rangle$$

- Estimate of field-shift parameter

$$F \propto \Delta |\psi(0)|^2$$

- Implemented in the GRASP2k package ¹

¹C. Nazé et al., *CPC*, 184pp. 2187–2196

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Alternative approach:

- Calculation of $\delta\nu$ for several isotopes
- Solve system of equations, based on parametrization
- Variation between results interpreted as error estimate

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Multi-Configuration Dirac-Fock Method

General

- Multi-configuration expansion

$$\Psi(PJM) = \sum_{i=1}^N c_i \Phi(\gamma_i PJM) \quad \sum_i |c_i|^2 = 1$$

- Configuration-state functions Φ are superpositions of Slater determinants
- Single-electron orbitals φ_i calculated by minimization of energy functional

$$E = \langle \Psi(PJM) | H_{DC} | \Psi(PJM) \rangle \quad (1)$$

- Variation in c_i is called configuration interaction, leads to eigenvalue problem

$$Hc = Ec$$

- Matrix Elements

$$H_{ij} = \langle \Phi_i | H_{DC} + H' | \Phi_j \rangle$$

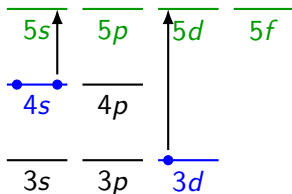
Multi-Configuration Dirac-Fock Method

Generation of basis sets

- Active-space method
- Configuration-state functions obtained by virtual excitations

Example: Mn

Reference configuration:
 $3d^5 4s^2$



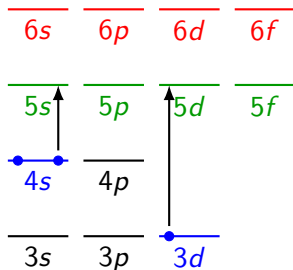
Multi-Configuration Dirac-Fock Method

Generation of basis sets

- Active-space method
- Configuration-state functions obtained by virtual excitations
- Successive enlargement of active space: “correlation layer”
- Layer model allows to monitor convergence

Example: Mn

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Results

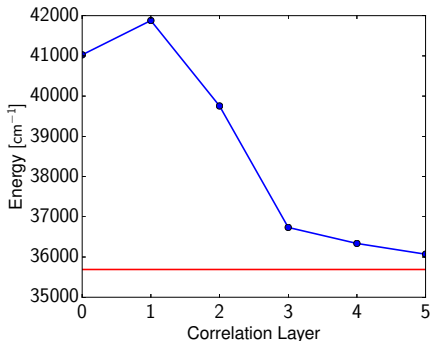
Manganese

- $3d^5 4s^2 ({}^6S_{5/2}) - 3d^5 4s 4p ({}^6P_{3/2}^o)$, $\Delta E = 35\,690 \text{ cm}^{-1}$
- Reference Configurations: $3d^5 (4s^2 + 4p^2)_{5/2}$, $(3d^5 4s 4p + 3d^6 4p)_{3/2}$

Correlation Model

Layer	Exc.	Orbitals	N_{CSF}
0		3d 4s 4p	232
1	SDTQ	3d 4s 4p	8 295
2	SD	5s 5p 5d 5f	94 779
3	SD	6s 6p 6d 6f	243 621
4	SD	7s 7p 7d 7f	454 821
5	SD	8s 8p 8d 8f	728 379

Transition energy

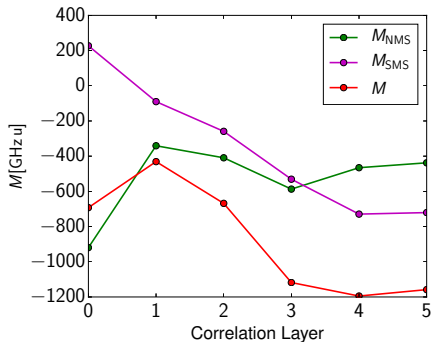


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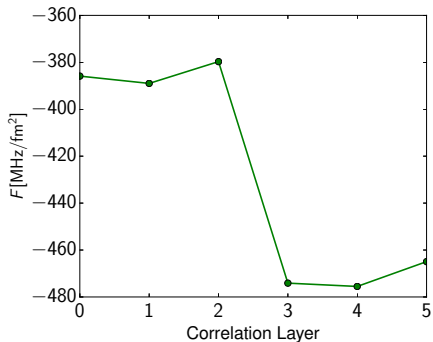
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Mass-shift parameters



Field-shift parameter



Atomic Physics Toolkit: JAC

General

- Package for atomic physics calculations
- Bound states and interaction with continuum states: Photo-ionization, auto-ionization, recombination...
- Many existing Tools in RATIP ²
- Wave function generation using GRASP ³
- Integrate existing software
- Data conversion and management

²S. Fritzsche, *CPC*, 183pp. 1525–1559

³P. Jönsson et al., *CPC*, 184pp. 2197–2203

Atomic Physics Toolkit: JAC

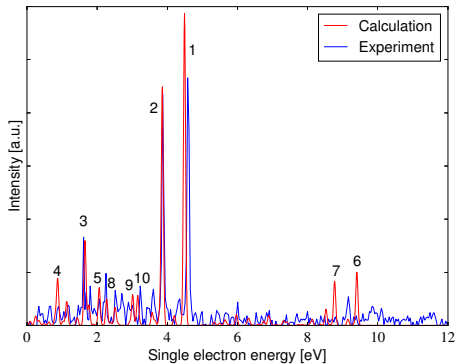
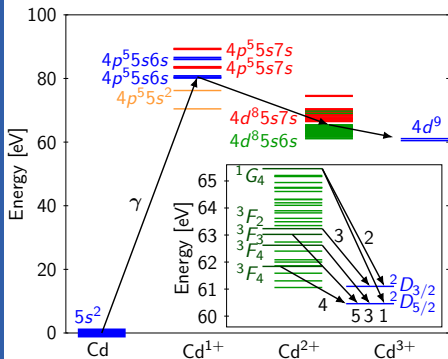
Example

The screenshot shows the JAC software interface with the following configuration:

- Working Directory: /home/randolf/Temp/Uni/Fe23+
- Atomic Properties | Atomic Structure | **Atomic Transitions** | Level Analyzer | About JAC
- Transition Probabilities | Photo Ionization | **Auger Decays**
- Name: demo
- Initial Levels: fe-23+ - Ct: Cl
- Final Levels: F-24+-dummy - Ct: Cl
- Interactions to include in addition to Coulomb: Breit Exchange interaction of continuum orbitals
- Properties to be calculated in addition to Auger rates and lifetimes:
 - Calculate angular distribution parameters
 - Calculate spin polarization parameters
- ▶ Calculate all Transitions
- ▶ Use standard exponential grid
- ▼ Modify default options
 - Apply biorthonormal transformation
 - Expand Wave functions in Slater Determinants
 - Print all selected transitions and energies before their computation
 - Sort all transitions by energy
- Minimal free electron energy:
- ▶ Do not use experimental Energies
- Settings Overwrite existing files Debug mode Use MPI 0
- Completed Auger: demo

Double-Auger Cascades in Cadmium

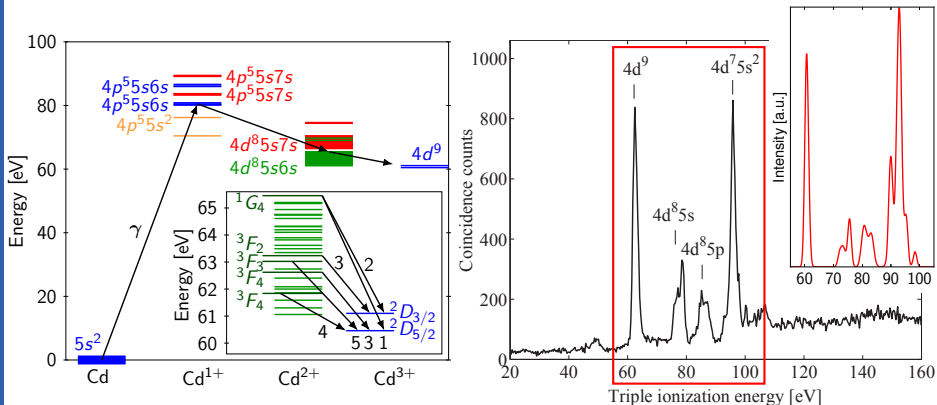
- Inner shell ionization is followed by multiple auger cascades
- Spectrum of emitted electrons calculated
- Process dominated by shake-up transitions $5s \rightarrow 6s$



⁴J. Andersson et al., *Phys. Rev. A*, 92p. 023414

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Shake-up Transitions: Biorthonormal transformation

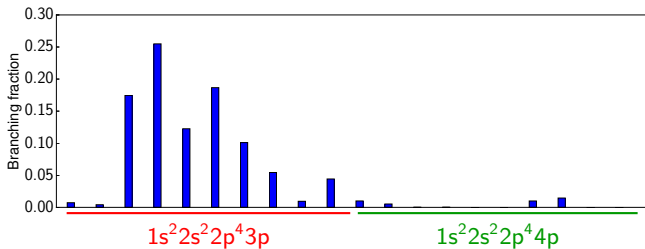
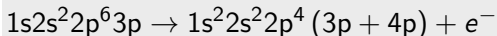
- Evaluation of transition amplitudes

$$A \propto \langle \psi | V | \psi' \rangle$$

$$\langle \varphi_i | \varphi'_j \rangle = \delta_{ij}$$

- Shake-up transitions cannot be calculated in this approximation

Example: Neon 1s Auger decay



⁵J. Olsen et al., *Phys. Rev. E*, 52pp. 4499–4508

Shake-up Transitions: Biorthonormal transformation

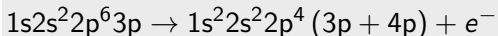
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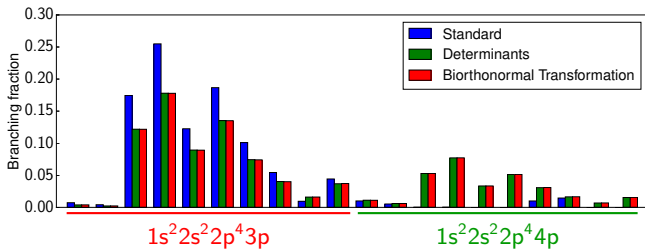
$$\langle \varphi_i | \varphi_j' \rangle = \delta_{ij}$$

- Shake-up transitions cannot be calculated in this approximation

Example: Neon 1s Auger decay



- Biorthonormal transformation ⁵ $\langle \varphi_i' | \varphi_j' \rangle = \delta_{ij}$, $c_i \mapsto c_i'$



⁵J. Olsen et al., *Phys. Rev. E*, 52pp. 4499–4508

Conclusions

- Our strategy on calculation of isotope-shift parameters
- Some results on isotope-shift calculations: Manganese
- Glimpse on our new Atomic Physics Toolkit: JAC
- Example: Biorthonormal transformation applied to calculation of Auger transition rates

