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Study of the hyperfine structure of Ba-like elements: an MCDHF approach for modeling the first excited levels

Lorenzo Nezosi[1*], Lucas Maison[1], Patrick Palmeri[1], Per Jönsson[2], Michel Godefroid[3]

1: Physique Atomique et Astrophysique, Université de Mons, Belgium
2: Materials Science and Applied Mathematics, Malmö University, Sweden
3: Spectroscopy, Quantum Chemistry and Atmospheric Remote Sensing, Université Libre de Bruxelles, Belgium

INTRODUCTION & AIM

The study of r-processed elements, such as barium (Ba) and strontium (Sr) is key to understand the formation of heavy elements in the Universe. The hyperfine structure (HFS) of their atomic levels is commonly used by astrophysicists to determine abundances via synthetic spectra.

Identifier of the dominant process: $f_{\mathrm{odd,Ba}} = \frac{N(^{135}\mathrm{Ba}) + N(^{137}\mathrm{Ba})}{\sum N(\mathrm{Ba\ isotopes})}$

Using the Multiconfiguration Dirac-Hartree-Fock (MCDHF) method [1], as implemented in the General Relativistic Atomic Structure Package (GRASP) [2], the magnetic dipole A and electric quadrupole B HFS constants were determined for the ground level and first excited levels of **^{135,137}Ba II** isotopes, as well as for **^{137}Ba I** and **^{87}Sr II** to assess the robustness of the developed model. New code developments such as natural orbitals [3], polarization effects [4] and the use of Configuration State Function Generators (CSFGs) as implemented in GRASPG [5] were tested for these heavy elements. Moreover, this work provides an estimate of the theoretical uncertainty; a feature often lacking in similar studies.

The aim is to develop a general MCDHF model for describing the HFS of elements with little or no available experimental data

METHOD

Based on the Dirac equation, the MCDHF method [1] focuses on multi-electron systems using the spinorbitals

$$\psi(r,\theta,\varphi) = \frac{1}{r}\begin{pmatrix} P_{n\kappa}(r)\chi_{\kappa m}(\theta,\varphi) \\ iQ_{n\kappa}(r)\chi_{-\kappa m}(\theta,\varphi) \end{pmatrix}$$

in the central field approximation. The wave function is approximated by an atomic state function (ASF), $\Psi(\Gamma\pi JM)$, given as a linear combination of configuration state functions (CSFs), $\Phi(\gamma\pi JM)$

$$\Psi(\Gamma\pi JM) \equiv |\Gamma\pi JM\rangle = \sum_i^{N_{CSFs}} c_i^{\Gamma\pi J}\Phi(\gamma_i\pi JM)\,.$$

The MCDHF equations are then obtained for the radial parts of the spin-orbitals, which are solved iteratively using the self-consistent field method.

The hyperfine interaction, arising from the interaction of electronic and nuclear magnetic moments, is described by

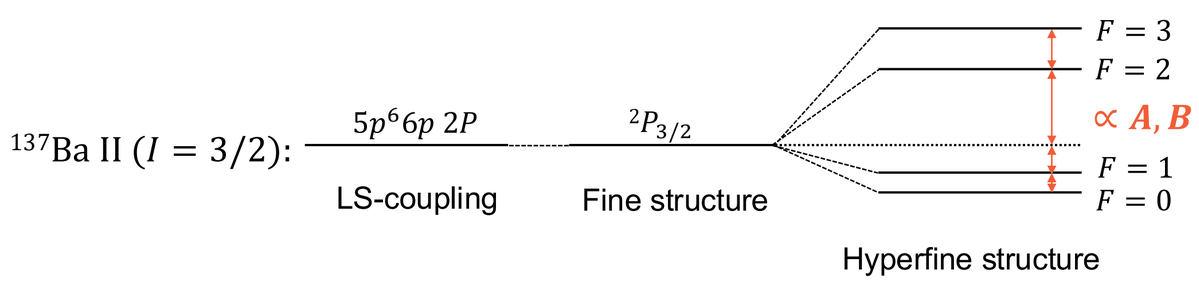
$$H_{\mathrm{HFS}} = \sum_{k\geq 1} \boldsymbol{T}^{(k)} \cdot \boldsymbol{M}^{(k)}\,,$$

with $\boldsymbol{T}^{(k)}$ and $\boldsymbol{M}^{(k)}$ being spherical tensor operators in the electronic and nuclear space, respectively.
When the hyperfine interaction is weak, H_{HFS} can be treated perturbatively, giving the first-order energy contribution

$$E_{\mathrm{HFS}} = \langle \Gamma JIFM_F|H_{\mathrm{HFS}}|\Gamma IJFM_F\rangle = E^{(1)}_{\mathrm{HFS}} + E^{(2)}_{\mathrm{HFS}}$$

$$E^{(1)}_{\mathrm{HFS}} \propto A_{\Gamma J} = \frac{\mu_I}{I}\frac{1}{J(J+1)2(J+1)}\langle \Gamma J||\boldsymbol{T}^{(1)}||\Gamma J\rangle$$

$$E^{(2)}_{\mathrm{HFS}} \propto B_{\Gamma J} = 2Q\sqrt{\frac{J(2J-1)}{(J+1)(2J+1)(2J+3)}}\langle \Gamma J||\boldsymbol{T}^{(2)}||\Gamma J\rangle$$



RESULTS & DISCUSSION

^{137}Ba II

$^2S_{1/2}$, $^2D_{3/2}$, $^2D_{5/2}$, $^2P^o_{1/2}$, $^2P^o_{3/2}$

- In-depth study of different classes of correlations showed that s- and p single (S) substitutions were crucial for the HFS of the ground level as deep as the 4s electrons

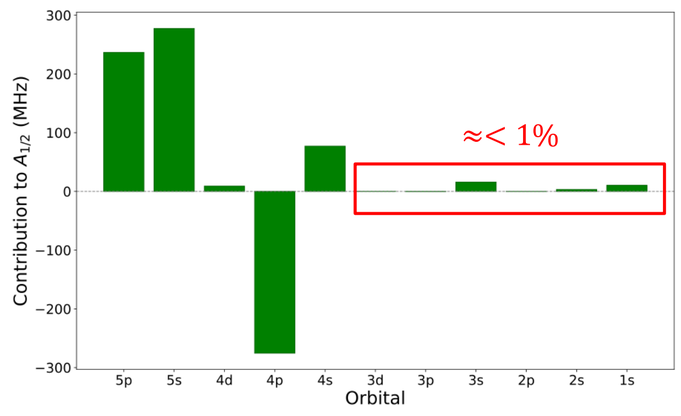


Fig. 1: Contributions of single (S) substitutions relative to the DHF $A(2S_{1/2})$ value in ^{137}Ba II

Chosen model: MCDHF CV SrD $nl \geq 4p$ + RCI CC SD $nl \in \{5p, 5s, 4p, 4s\}$

- Considering independent RCI calculations give a theoretical uncertainty [6], given by $\mu \pm \sigma$, with $\Delta_{\mathrm{expt.}} \leq 10\%$

Level	A (MHz)		B (MHz)	
	This work	Δ Experiment	This work	Δ Experiment
$5p^6 6s\ ^2S_{1/2}$	3901.77 ± 182.01	2.91%	/	/
$5p^6 5d\ ^2D_{3/2}$	178.47 ± 14.62	5.93%	42.56 ± 5.30	4.45%
$5p^6 5d\ ^2D_{5/2}$	0.51 ± 6.12	**104.24%**	56.59 ± 6.99	4.78%
$5p^6 6p\ ^2P^o_{1/2}$	667.18 ± 34.43	10.29%	/	/
$5p^6 6p\ ^2P^o_{3/2}$	118.33 ± 2.93	6.97%	82.43 ± 3.97	10.89%

- Obtained values used to determine $A = 135$ values with appropriate scaling [6]

^{87}Sr II

$^2S_{1/2}$, $^2D_{3/2}$, $^2D_{5/2}$, $^2P^o_{1/2}$, $^2P^o_{3/2}$

- The same model gives similar agreement as in ^{137}Ba II
- $\Delta_{\mathrm{expt.}}[A(^2D_{5/2})] \approx 170\%$ explained by non-relativistic calculations

MCHF: $A \propto a_l, a_{sd}, a_c$

MCDHF: $A \propto \boldsymbol{T}^{(1)}$

^{137}Ba I

$^1D_2$, $^3D_1$, $^3D_2$, $^1P^o_1$, $^3P^o_1$

- Non-negligible configuration mixing ($\geq 5\%$) needs a MR study and improved the problematic values by 30%

CONCLUSIONS

- CV correlations are key to the model; CC correlations decrease the values
- $\Delta_{\mathrm{expt.}} < 10\%$ for all constants except $A(2D_{5/2})$, whose discrepancies are caused by Fermi-contact-like terms
- The uncertainty estimate covers other theoretical studies
- Natural orbitals correct the HFS constants, giving satisfying results with less CSFs
- Polarization orbitals are promising but too costly with available computer resources

FUTURE WORK / REFERENCES

- In-depth study presented in a submitted Atoms paper
- More GRASPG developments on the way [7]
- HFS of Ca and Ra to see if there is a trend related to group 2 elements
- Isotopic shift calculations in the way for a more complete description of the levels

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* : Corresponding author – lorenzo.nezosi@student.umons.ac.be

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