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Calculations for heavy atoms with open shells: energy levels and hyperfine structure of Cf, Es, Fm, and Md.

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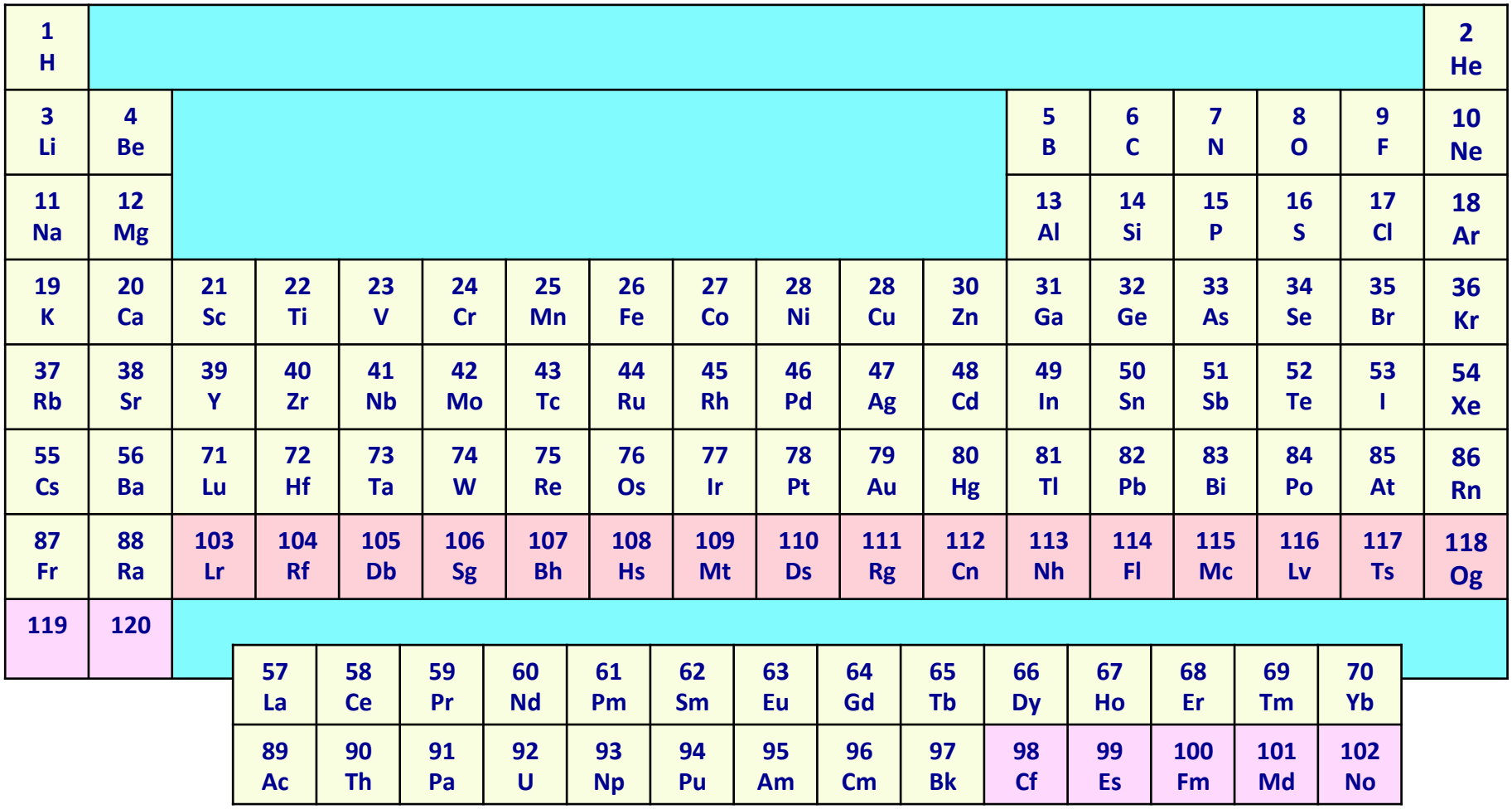


Present work

Atoms with Z>97 have open 5f, 7s, 6d or/and 7p shells.

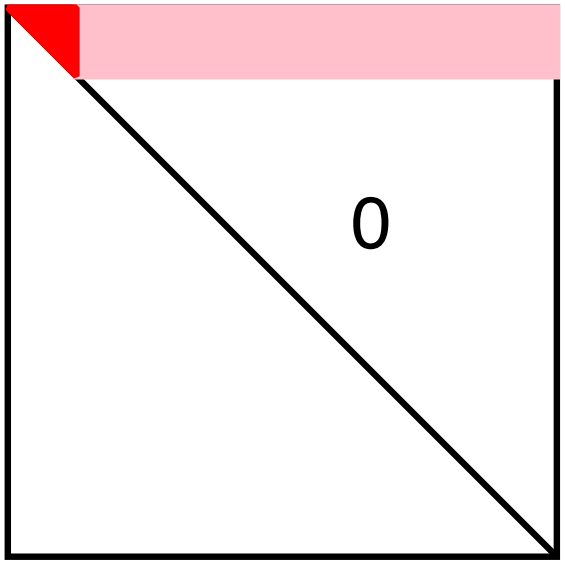
Number of valence electrons from 1 to 16.

Standard CI technique would not work!

The CIPT method

(configuration interaction with perturbation theory)
V. A. Dzuba, J. C. Berengut, C. Harabati, and V. V. Flambaum ,
PRA **95**, 012503 (2017))

The structure of the CI matrix



Use V^{N-1} as good initial approximation

$$\Psi = \sum_i c_i \Phi_i + \sum_m c_m \Phi_m$$

Small correction

Has small number of terms

Neglecting the off-diagonal m.e. corresponds to neglecting the third-order terms

$$\delta E_g = \sum_{i,j} \frac{\langle g|H^{\rm CI}|i\rangle\langle i|H^{\rm CI}|j\rangle\langle j|H^{\rm CI}|g\rangle}{(E_g - E_i)(E_g - E_j)}.$$

Suppressed by large energy denominators

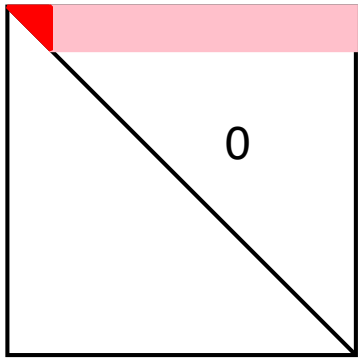
The idea is not new and gaining popularity.

- E. A. Parpia, C. Froese Fischer, I. P. Grant, **GRASP92,** CPC **94**, 249 (1996).

$$\begin{pmatrix} \mathbf{H}_{00} & \mathbf{H}_{01} \\ \mathbf{H}_{10} & \mathrm{diag}(\mathbf{H}_{11}) \end{pmatrix} \qquad (3.15)$$

- E. V. Kahl, J. C. Berengut, **Emu CI**, CPC **238**, 232 (2019).
- M. G. Kozlov , I. I. Tupitsyn, A. I. Bondarev , D. V. Mironova PRA **105**, 052805 (2022).
- Etc...

Current implementation (the CIPT method)



$$(H^{\mathrm{CI}} - EI)X = 0, \qquad (1)$$

$$\langle i|H|j\rangle \rightarrow \langle i|H|j\rangle + \sum_m \frac{\langle i|H|m\rangle\langle m|H|j\rangle}{E - E_m} \qquad (2)$$

Eqs. (1) and (2) give exact solution for the shown CI matrix if energy E in (1) an (2) are the same.

I.e., iterations are needed.
Only first iteration takes time since the numerator can be saved and reused.

Neglecting the off-diagonal m.e. is the only assumption!

Limitations of the CIPT method

- Only low-lying states can be calculated.
- Calculations are sensitive to the initial approximation. This may lead to different accuracy for different states.
- Core-valence correlations are not included.
- Half-filled f-shell is hard to treat.

$$\Psi = \sum_i c_i \Phi_i + \sum_m c_m \Phi_m$$

Small correction

Has small number of terms

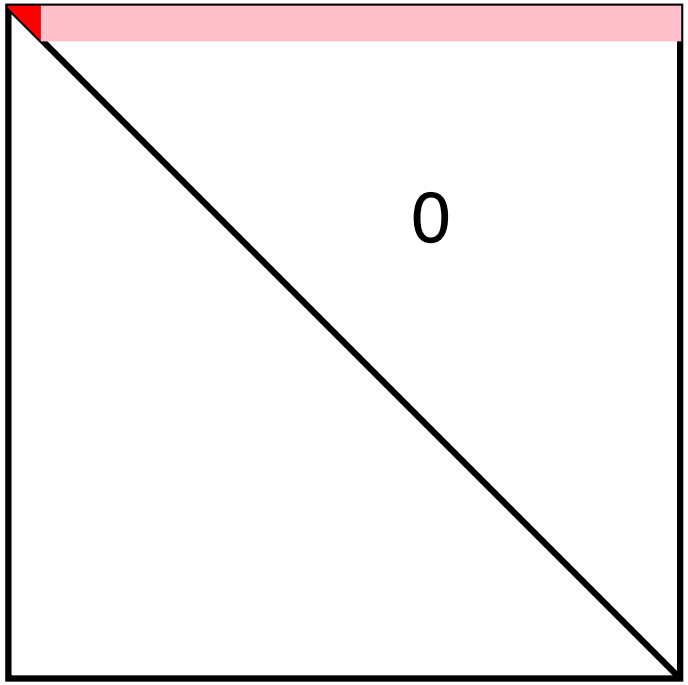
First two limitations can be eased by increasing the size of the effective CI matrix.

In contrast, core-valence correlations require special consideration.

Special use of the CIPT approach: increasing efficiency of the CI+MBPT method.

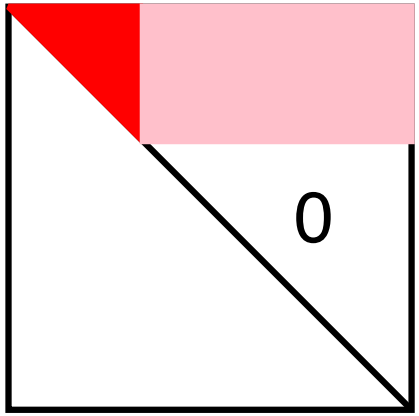
CI matrix

~10 electrons, V^{N-1}



N_{eff} ~ 1 – 100; N_{total} ~ 10^7 - 10^8

~5 electrons, V^{N-M}



N_{eff} ~ 10^3 - 10^4; N_{total} ~ 10^6

- The use of V^{N-M} allows to include core-valence correlations.
- The use of **CIPT** allows to go to larger number of electrons.

Calculation of matrix elements

RPA equations: $(\hat{H}^{\rm RHF} - \epsilon_c)\delta\psi_c = -\left(\hat{f} + \delta V^f_{\rm core}\right)\psi_c,$

Transition amplitudes: $A_{ab} = \langle a|\hat{d} + \delta V^{N-1}|b\rangle,$

Hyperfine structure $\delta\epsilon_v = \left\langle v\middle|\hat{f} + \delta V^f_{\rm core}\middle|v\right\rangle.$

Magnetic dipole hfs: $A_a = \dfrac{g_I \delta\epsilon_a^{(A)}}{\sqrt{J_a(J_a+1)(2J_a+1)}},$

Electric quadrupole hfs: $B_a = -2Q\delta\epsilon_a^{(B)}\sqrt{\dfrac{J_a(2J_a-1)}{(2J_a+3)(2J_a+1)(J_a+1)}}.$

Breit and QED

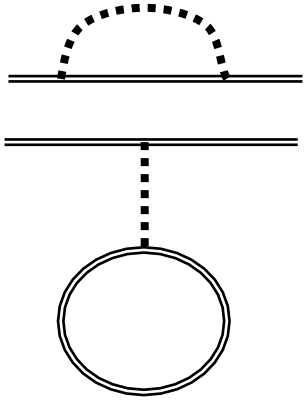
Breit interaction (ω=0):
(magnetic interaction and retardation)

$$H_B = -\frac{\alpha_1\alpha_2 + (\alpha_1 n)(\alpha_2 n)}{2r}$$

Breit potential is formed: $e^2/r \to V^{\mathrm{C}}, \quad H^{\mathrm{B}} \to V^{\mathrm{B}}$

QED. Radiative potential method (Flambaum and Ginges, 2005)

$$\Phi_{\mathrm{rad}}(r) = \Phi_U(r) + \Phi_g(r) + \Phi_f(r) + \Phi_l(r) + \frac{2}{3}\Phi_{WC}^{simple}(r)$$



$\Phi_g(r)$ – magnetic formfactor
$\Phi_f(r)$ – electric formfactor
$\Phi_l(r)$ – low energy electric formfactor
$\Phi_U(r)$ – Uehling potential
$\Phi_{WC}(r)$ – Wichmann-Kroll potential

Both potentials are included into HF: $V^{\mathrm{HF}} \to V^{\mathrm{HF}} + V^{\mathrm{B}} + \Phi_{\mathrm{rad}}$

Relaxation effect in E119

(for Breit and QED)

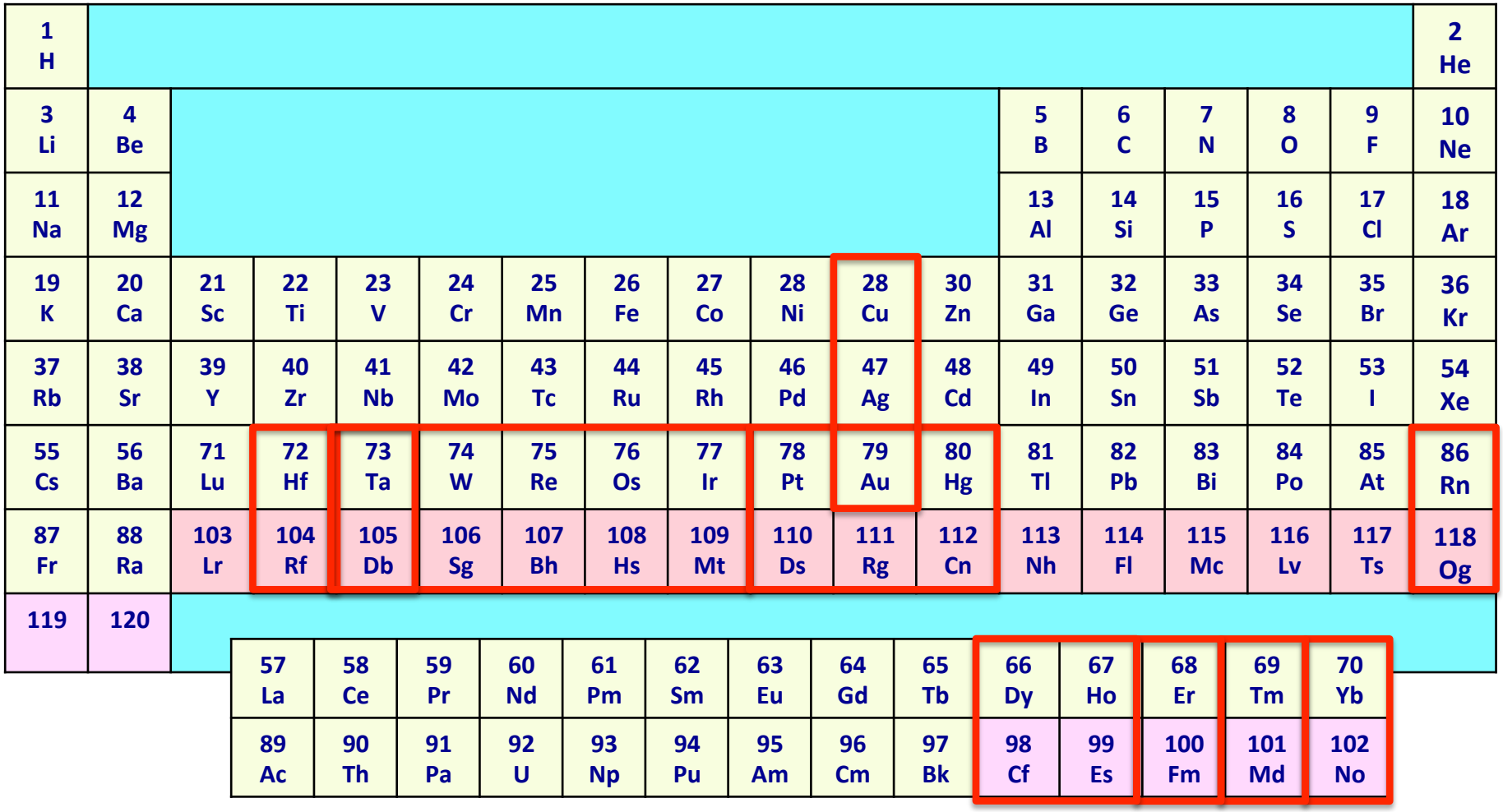
Breit

State	Energy cm^{-1}	$\langle\psi\|H^B\|\psi\rangle$ (no relaxation) cm^{-1}	%	ΔE_B (with relaxation) cm^{-1}	%
8s 1/2	-39697	217	-0.5%	38	-0.1%
8p 1/2	-24482	126	-0.5%	67	-0.3%
8p 3/2	-18626	45	-0.2%	8	-0.04%
7d 3/2	-17926	65	-0.4%	-30	+0.2%
7d 5/2	-17422	46	-0.3%	-28	+0.2%

QED

State	Energy cm^{-1}	$\langle\psi\|\Phi_{rad}\|\psi\rangle$ (no relaxation) cm^{-1}	%	ΔE_{rad} (with relaxation) cm^{-1}	%
8s 1/2	-39697	108	-0.3%	78	-0.2%
8p 1/2	-24482	19	-0.08%	7	-0.03%
8p 3/2	-18626	11	-0.06%	4	-0.02%
7d 3/2	-17926	2	-0.01%	-21	+0.1%
7d 5/2	-17422	3	-0.02%	-16	+0.1%

The use of the CIPT method

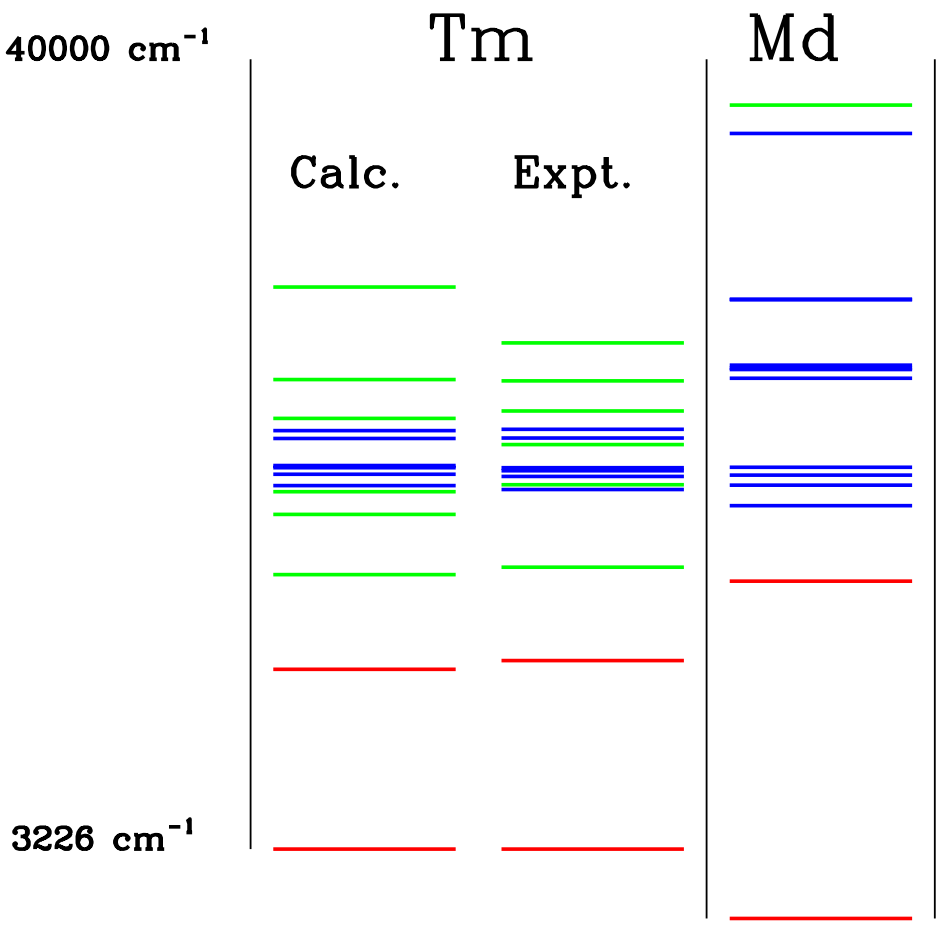


Present work

More than 10 publications

Energy levels of Tm and Md

(thulium and mendelevium)



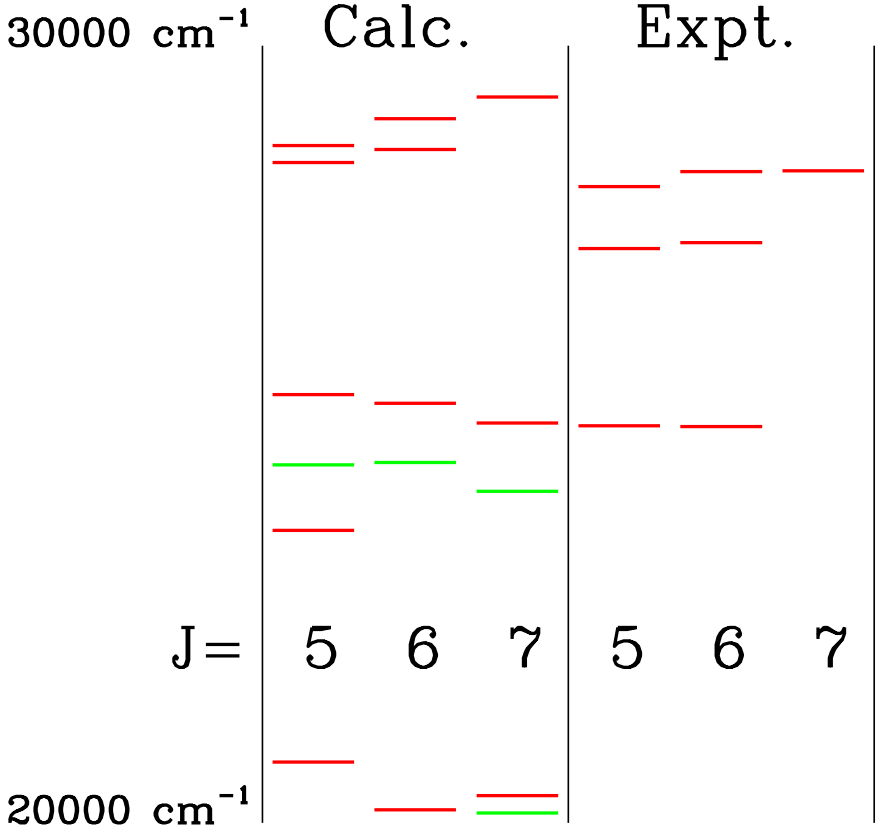
--- $4f^{13}\ 6s^2$ or $5f^{13}\ 7s^2$

--- $4f^{13}\ 6s6p$ or $5f^{13}\ 7s7p$

--- $4f^{12}\ 5d6s^2$ or $5f^{12}\ 6d7s^2$

Energy levels of Fm

(odd states connected to the G.S. by E1).



$\Delta E/E$ ~ 2-4%

$\Delta E/E$ ~ 1-1.5%

--- $5f^{12}\ 7s7p$

--- $5f^{11}\ 6d7s^2$

GS: $5f^{12}\ 7s^2$ J=6

Expt.: Sewtz et al, PRL **90**, 163002 (2003)

Backe et al, Hyperfine Interactions **162**, 3(2005)

Ionization potentials

(cm^{-1})

Atom	Calculations	NIST	Difference
$_{68}$**Er**	49216	49262	0.1%
$_{69}$**Tm**	50332	49880	0.9%
$_{98}$**Cf**	50821	50663	0.3%
$_{99}$**Es**	51763	51358	0.8%
$_{100}$**Fm**	52902	52600(1050)	~1%
$_{102}$**Md**	53800	53100(600)	~1%

Coclusions:

- Accuracy is good for ground states
- Should be true for HFS as well
- HFS of ground states is sufficient to find A and B.

HFS of the ground state of Dy, Ho, Er, Es. Comparison with experiment (MHz).

	A_{theor}	B_{theor}	A_{expt}	B_{expt}
^{161}Dy	-113	1127	-116.231	1091.577
^{165}Ho	787	-1943	800.583	-1668.089
^{167}Er	-117	-5034	-120.487	-4552.984
^{253}Es	798	-5481	817.153	-4316.254

Expt.: Childs, PRA 28, 3402 (1983).

$\Delta A/A < 3\%$

$\Delta B/B \sim 3\text{-}30\%$

HFS of the ground state of Cf, Es, Fm, and Md (MHz).

Atom	Conf.	Term	A	B
Cf	$5f^{10}\ 7s^2$	$^5I_8$	608 (μ/I)	477 Q
Es	$5f^{11}\ 7s^2$	$^5I^o_{15/2}$	681 (μ/I)	-818 Q
Fm	$5f^{12}\ 7s^2$	$^3H_6$	655 (μ/I)	-1750 Q
Md	$5f^{13}\ 7s^2$	$^2F^o_{7/2}$	826 (μ/I)	-1808 Q

Similar accuracy is expected.

More on Fm

	CIPT Energy	CIPT g-factor	Experimental energy [5, 6]	A/g_I MHz	B/Q MHz	
		Ground state, $J = 6$				
S	0	1.1619		655	-1750	← HFS was measured before
		Odd states with $J = 5$				
P	20782	1.1505		-1668	-1422	
P	23761	1.1834		2693	-446	
D	24605	1.1918		1927	-497	
P	25512	1.1212	25111.8(0.2)	253	-1787	← HFS was measured before
P	28497	1.1776	27389(1.5)	1449	-1164	
P	28715	1.2343	28185(1.5)	3086	135	
D	30669	1.1147		553	592	
		Odd states with $J = 6$				
P	18964	1.2565		2580	-1458	
D	19595	1.2846		730	-621	
P	20167	1.0880		-752	-1211	
D	24638	1.1643		651	-956	
P	25397	1.1854	25099.8(0.2)	1850	-1956	← HFS was measured before
P	28663	1.2501	27466(1.5)	3063	406	
P	29060	1.1721	28377(1.5)	481	-1885	
		Odd states with $J = 7$				
D	20128	1.2341		821	-1039	
P	20352	1.1953		2823	-1732	
D	24267	1.1526		623	-956	
P	25143	1.2344		2394	-1775	
P	29337	1.1455	28391(1.5)	-359	-1080	
D	33110	1.1481		439	250	
D	33900	1.0641		547	225	

$S = 5f^{12}\ 7s^2$

$P = 5f^{12}\ 7s\ 7p$

$D = 5f^{11}\ 6d\ 7s^2$

Expt.: Sewtz et al, PRL **90**, 163002 (2003)
Backe et al, HI **162**, 3(2005)

Conclusion

The CIPT method is a valuable tool in studying open-shell atoms and helping fundamental research in many ways.