Relativistic Coupled-Cluster Calculations for Clock Transition Properties in Al^+ and Pb^{2+}

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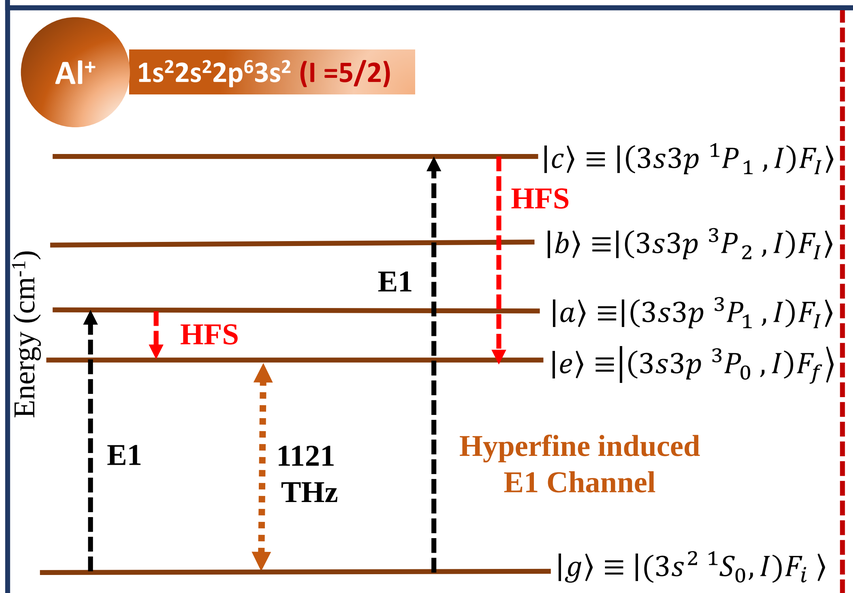
Abstract: We have developed RCC based theories for the properties calculations of closed-shell, one-valence and two-valence atomic systems. These theories are implemented as sophisticated parallel FORTRAN codes [1]. Using our in-house two-valence RCC code, we have probed the clock transition properties in Al^+ [2] and Pb^{2+} [3]. We have computed excitation energies, E1 and M1 transition amplitudes, oscillator strengths, dipole polarizability, and the lifetime of the metastable clock state. For Al^+, our computed lifetime of 20.2 s for $3s3p\ ^3P_0$ clock state is in excellent agreement with the experimental value 20.6 s. For Pb^{2+}, our calculations predict a high lifetime of 9.8×10^6 s for $6s6p\ ^3P_0$ clock state.

Al^+ $1s^22s^22p^63s^2$ (I =5/2)



E1HFS Decay rate:

$$\Gamma_{\text{E1HFS}} = \mu_I^2\,(1+I^{-1})\,\frac{4\,(2\pi)^3}{27\hbar\lambda^3}\left|\frac{\langle {}^1S_0\|D\|{}^3P_1\rangle\langle {}^3P_1\|t^1\|{}^3P_0\rangle}{E_{^3P_0}-E_{^3P_1}}+\frac{\langle {}^1S_0\|D\|{}^1P_1\rangle\langle {}^1P_1\|t^1\|{}^3P_0\rangle}{E_{^3P_0}-E_{^1P_1}}\right|^2$$

E1 & HFS Transition Amplitudes (a.u.)

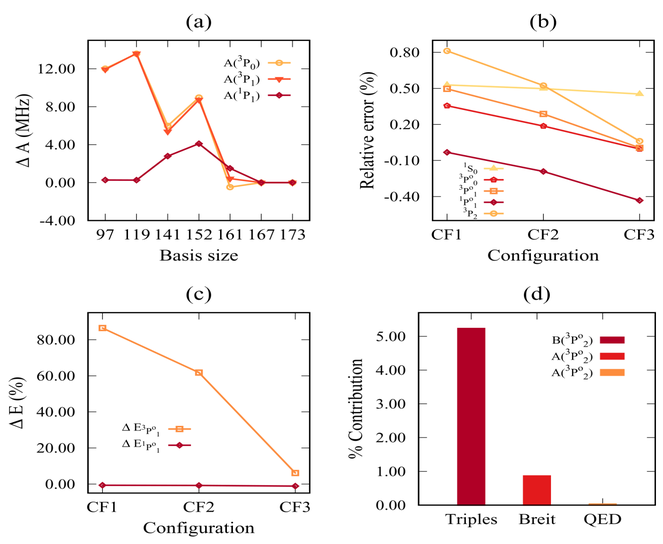
State	RCC	Breit + QED+ P-triples	Total	Others work
$\langle {}^1S_0\|D\|{}^3P_1\rangle$	-1.425 × 10^{-2}	-0.088 × 10^{-2}	-1.513× 10^{-2}	
$\langle {}^1S_0\|D\|{}^1P_1\rangle$	2.841	-0.001	2.840	
$\langle {}^3P_1\|t^1\|{}^3P_0\rangle$	-0.095	0.001	-0.094	-0.120 [4]
$\langle {}^1P_1\|t^1\|{}^3P_0\rangle$	0.079	-0.001	0.078	0.096 [4]

Hyperfine Structure Constant (MHz)

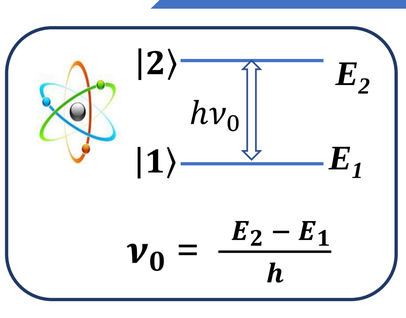
Methods	MHFS (A)			QHFS (B)		
	$^3P_1$	$^3P_2$	$^1P_1$	$^3P_1$	$^3P_2$	$^1P_1$
CCSD	1385.41	1188.02	292.59	-16.17	25.55	27.88
CCSD(T)	1393.73	1184.16	283.93	-16.65	24.27	28.31
CCSD(T) + Breit + QED	1389.81	1174.29	286.08	-16.65	24.27	28.31
Others[5]	1348	1149		-15.62	31.42	

Lifetime (s)

Methods	$E1_{HFS}$	τ
CCSD	5.153×10^{-5}	21.33
CCSD(T)	5.316×10^{-5}	20.04
CCSD(T) + Breit + QED	5.295×10^{-5}	20.20 ± 0.91
Experiments		20.6 ± 1.4[6]



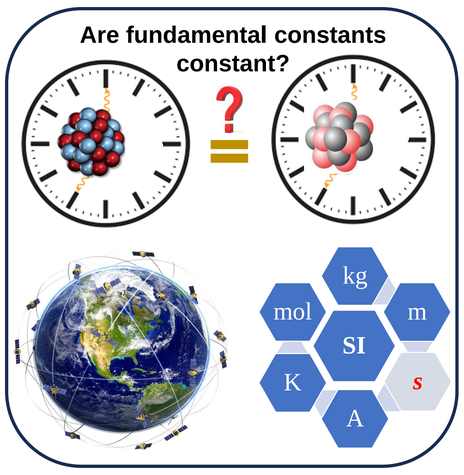
Introduction



1. Fractional frequency uncertainty $= \frac{\Delta f}{f_0}$

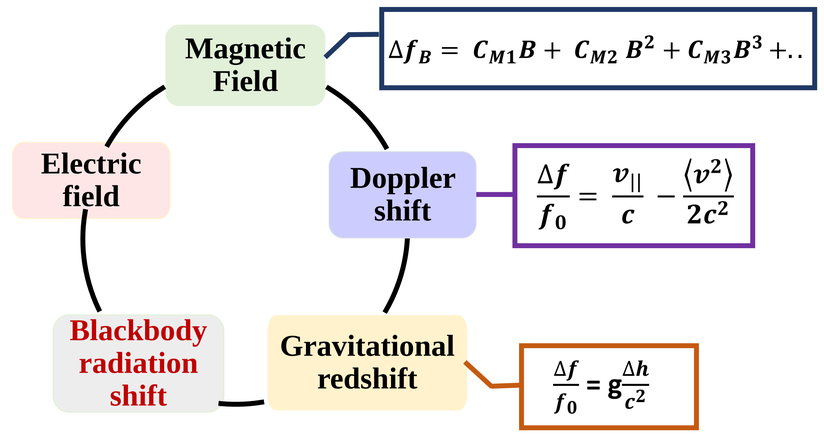
2. Instability $= \sigma(\tau) = \frac{1}{2\pi f_0\sqrt{NT_m\tau}}$

Applications



1. Global Navigation Satellite Systems (GNSS)
2. Time & Frequency Standards
3. Telecommunications
4. Financial Trading
5. Variation of Fundamental Constant

Systematic Errors in Clocks



BBR Shift

$$\delta\nu_{BBR}^{E1} = \frac{-1}{2}\,(831.9\,V/m)^2\,\Delta\alpha_0^{E1}$$

2nd order Doppler shift

$$\frac{\delta\nu_{Doppler}}{\nu} = \frac{-E_{kinetic}}{mc^2}$$

Coupled-Cluster Theory for Two-valence Atoms

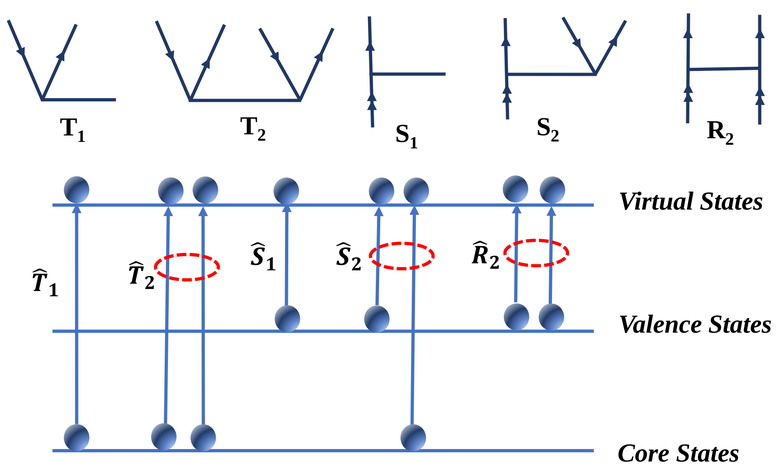
➢ **Dirac-Coulomb Hamiltonian:**

$$\mathrm{H^{DC}} = \sum_{i=1}^{N}[c\boldsymbol{\alpha}_i\,.\,\boldsymbol{p}_i + (\boldsymbol{\beta}_i - \mathbf{1})c^2 - V_N(\boldsymbol{r}_i)] + \sum_{i<j}\frac{1}{r_{ij}}$$

➢ **Many-body Schrodinger equation:** $\mathrm{H^{DC}}|\Psi_i\rangle = E_i\,|\Psi_i\rangle$

➢ **Exact wavefunction in coupled-cluster theory [2, 3]:**

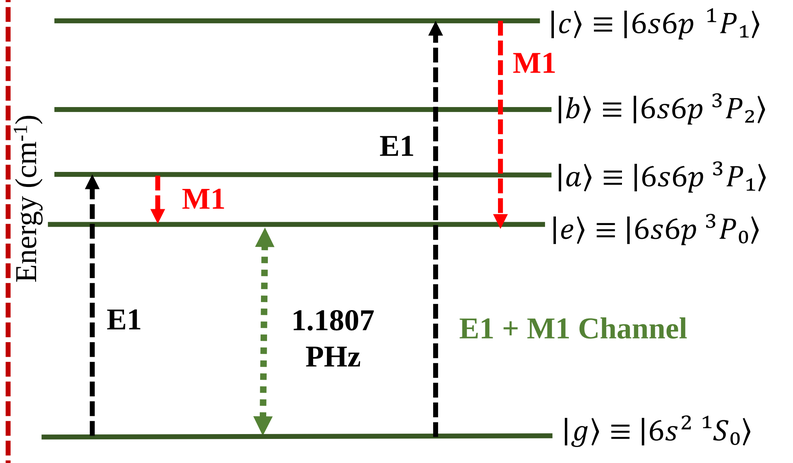
$$|\Psi_i\rangle = e^T[\mathbf{1} + S_1 + S_2 + \tfrac{1}{2}S_1^2 + R_2]|\Phi_i\rangle$$



➢ **Working equation for two-valence systems:**

$$\left\langle\Phi_{vw}^{pq}\middle|\bar{H}_N + \{\overline{\bar{H}_N S_1}\} + \{\overline{\bar{H}_N S_2}\} + \frac{1}{2}\{\overline{\bar{H}_N S_1^2}\} + \{\overline{\bar{H}_N R_2}\}\middle|\Phi_{vw}\right\rangle = E_{vw}^{attach}\left\langle\Phi_{vw}^{pq}\middle|S_1 + S_2 + S_1^2 + R_2\middle|\Phi_{vw}\right\rangle$$

Pb^{2+} $1s^22s^22p^63s^23p^63d^{10}4s^24p^64d^{10}4f^{14}5s^25p^65d^{10}6s^2$ (I=0)



E1M1 Decay rate:

$$\Gamma_{\text{E1M1}} = \frac{8}{27\pi}\,\alpha^6\int_0^\infty d\omega_1\,\omega_1^3\int_0^\infty d\omega_2\,\omega_2^3\left|\sum_{n_+}\frac{\langle f\|\mathbf{M1}\|n_+\rangle\langle n_+\|\mathbf{D}\|i\rangle}{E_i+\omega_1-E_{n_+}}+\sum_{n_-}\frac{\langle f\|\mathbf{D}\|n_-\rangle\langle n_-\|\mathbf{M1}\|i\rangle}{E_i+\omega_2-E_{n_-}}\right|^2$$

$$\delta\,(E_f+\omega_1+\omega_2-E_i)$$

E1 & M1 Transition Amplitudes (a.u.)

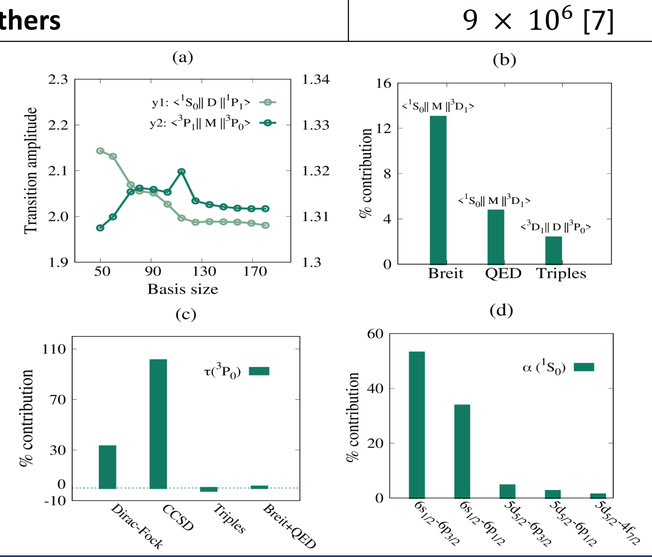
State	RCC	Breit + QED+ P-triples	Total	Others work
$\langle {}^1S_0\|D\|{}^3P_1\rangle$	0.5319	-0.0031	0.5288	0.706 [7], 0.644 [8]
$\langle {}^1S_0\|D\|{}^1P_1\rangle$	1.9854	-0.0324	1.9530	2.350 [7], 2.384 [8]
$\langle {}^3S_1\|D\|{}^3P_0\rangle$	0.5362	-0.0003	0.5359	0.963 [8]
$\langle {}^3D_1\|D\|{}^3P_0\rangle$	-1.4796	-0.0184	-1.4980	-1.516 [8]
$\langle {}^3P_1\|M\|{}^3P_0\rangle$	-1.3117	0.0001	-1.3116	-0.674 [7]
$\langle {}^1P_1\|M\|{}^3P_0\rangle$	0.4972	0.0003	0.4975	0.205 [7]
$\langle {}^1S_0\|M\|{}^3S_1\rangle$	0.0044	-0.0003	0.0041	
$\langle {}^1S_0\|M\|{}^3D_1\rangle$	-0.0143	-0.0026	-0.0169	

Ground State Polarizability (a.u.)

State	Present work	
	Method	α
$6s^2\ ^1S_0$	DF	16.246
	PRCC(T)	14.166
	PRCC(T) + Breit +QED	14.064
	Recommended	14.02 ± 0.21
	Other results	13.3 [8], 13.6(Expt) [9]

Lifetime (s)

Configurations/Methods	τ ($\times 10^6$)
$6s^2$ + 6s6p	9.595
6s7s	0.029
6s6d	0.248
Total CCSD	9.872
CCSD(T) + Breit + QED	9.761
Others	9×10^6 [7]



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Acknowledgments

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