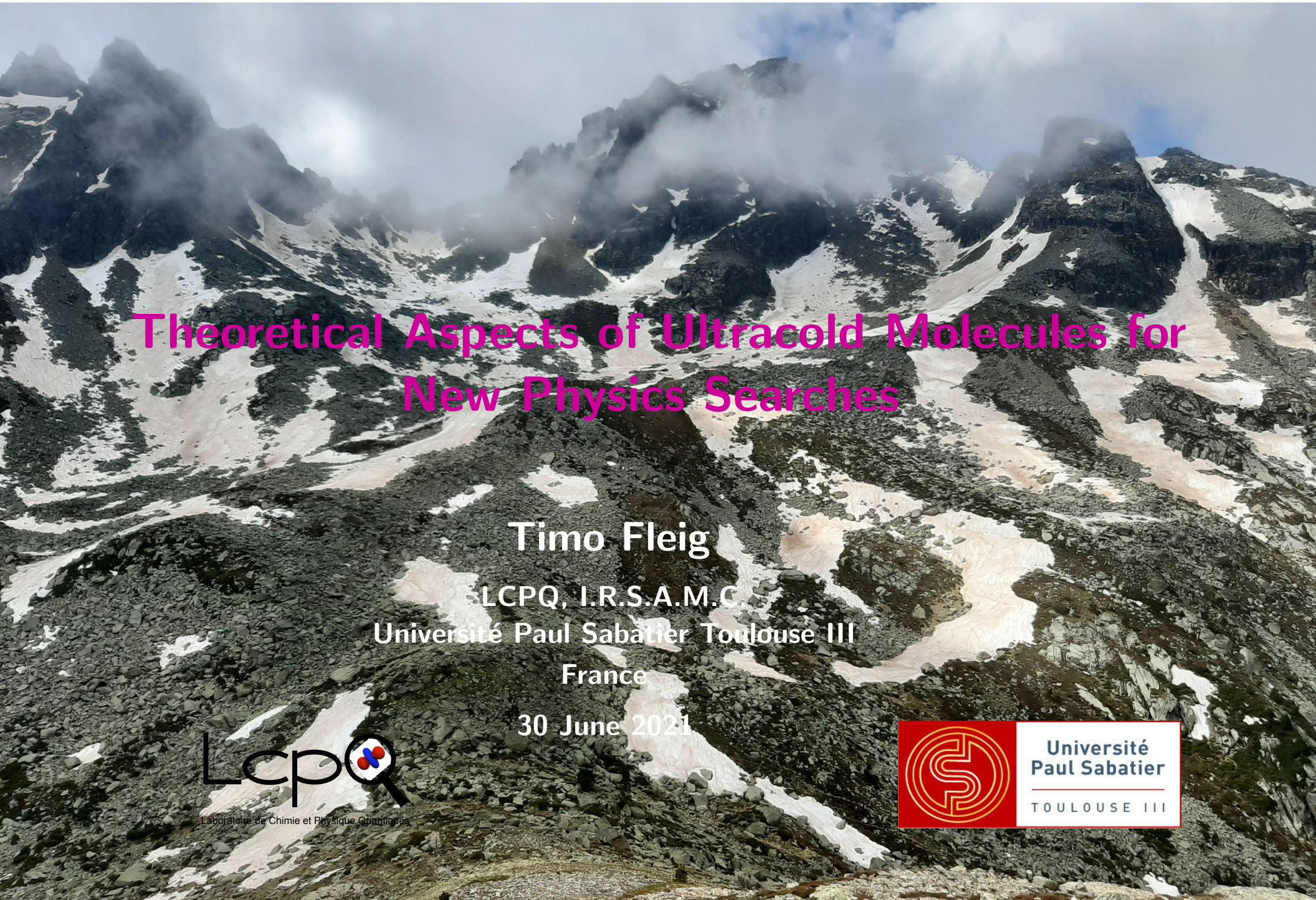


Theoretical Aspects of Ultracold Molecules for New Physics Searches

Timo Fleig

LCPQ, I.R.S.A.M.C.
Université Paul Sabatier Toulouse III
France

30 June 2021

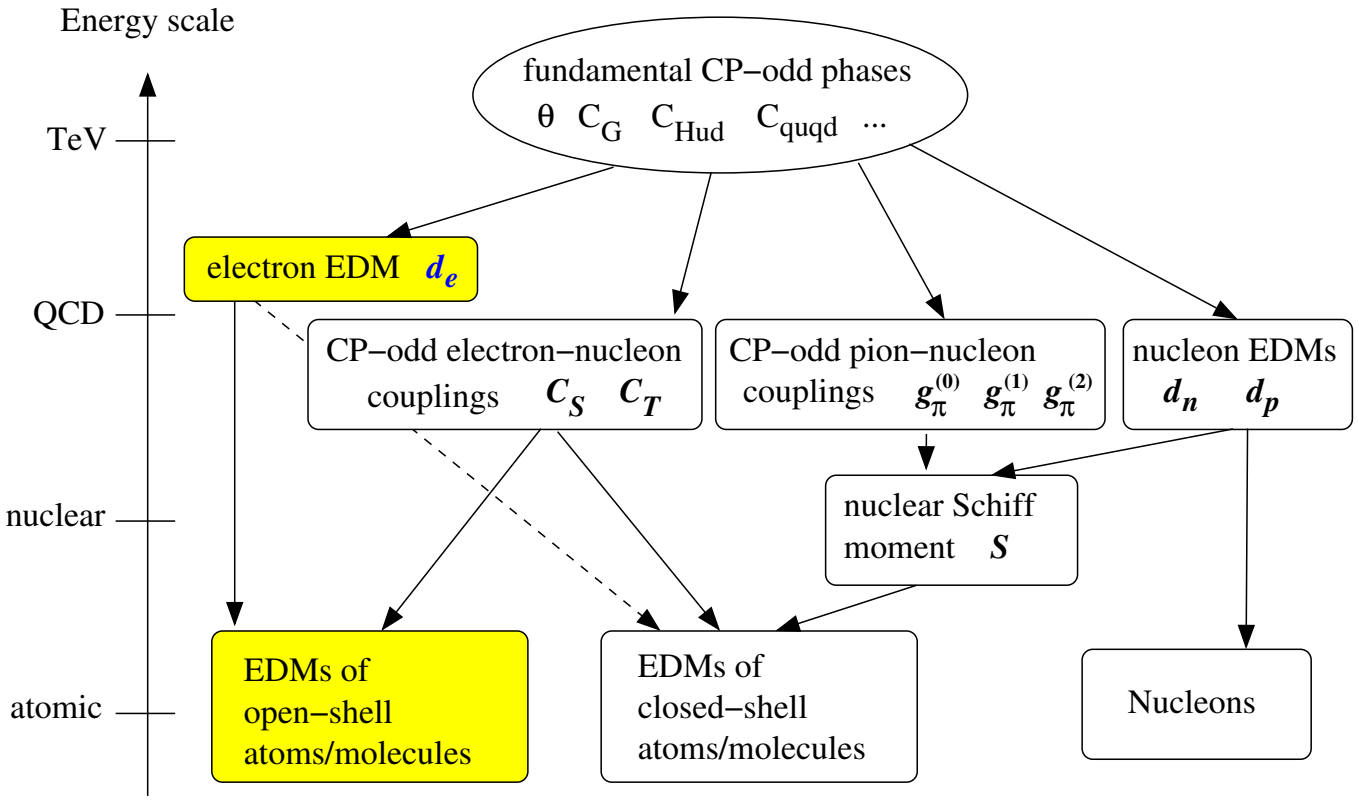
Laboratoire de Chimie et Physique Quantiques

Université Paul Sabatier
TOULOUSE III

Outline – Part 1

Electron EDM with trapped ultracold molecules

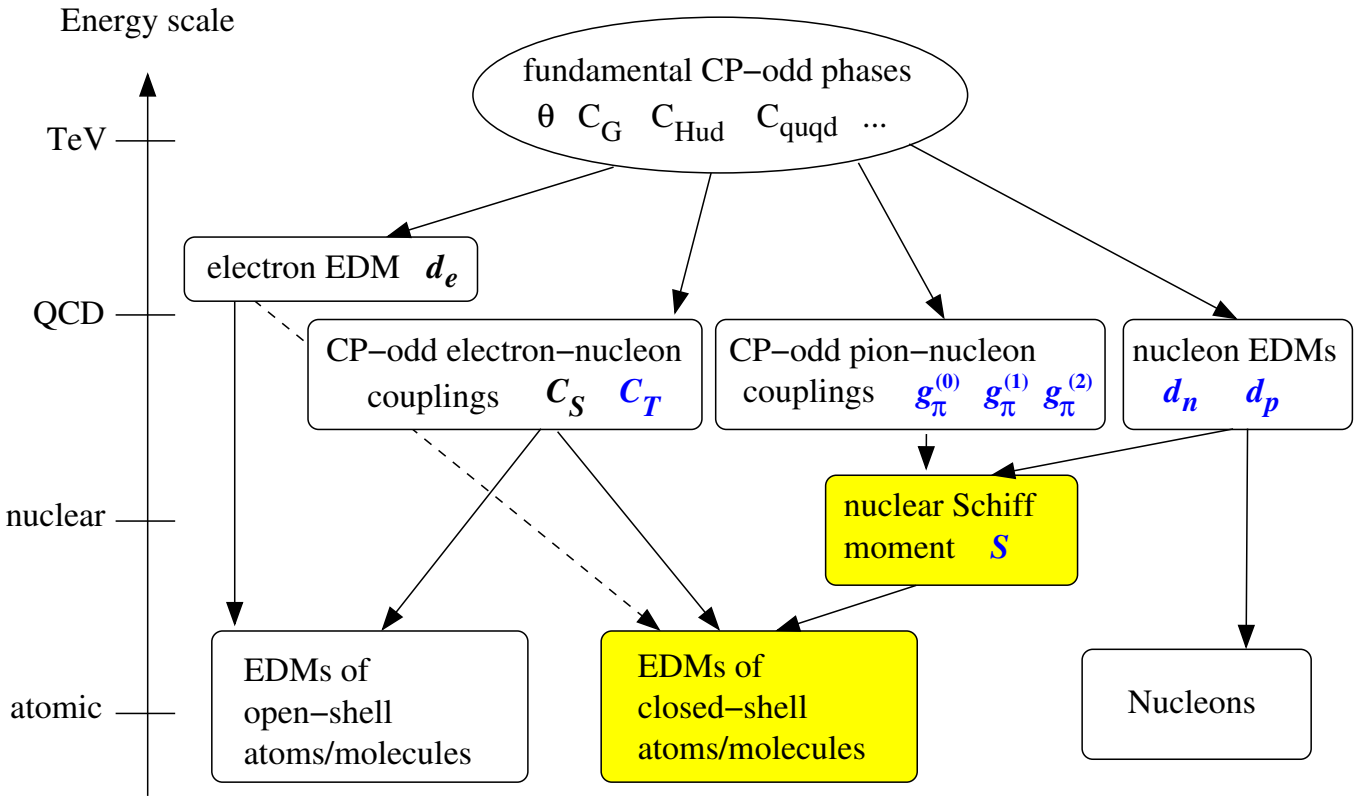
Alkali-radium diatomics and the story of AgRa
Dispersion coefficients with Gaussian basis sets



Outline – Part 2

Toward nuclear Schiff moment with trapped ultracold molecules

Atomic Schiff interactions: Gaussian basis sets . . . again
Next-generation Schiff molecule: AgFr ?



$\mathcal{P}$,$\mathcal{T}$-odd Property Calculations

using correlated wavefunctions

Expectation values over relativistic Configuration Interaction wavefunctions[1]

$$\left\langle \hat{O} \right\rangle_{\psi_k^{(0)}} = \sum_{I,J=1}^{\dim\mathcal{F}^{\mathrm{t}}(\mathrm{M,n})} c_{kI}^* c_{kJ} \left\langle \, | \, (\mathcal{S}\overline{\mathcal{T}})_I^\dagger \left| \hat{O} \right| (\mathcal{S}\overline{\mathcal{T}})_J \, | \, \right\rangle$$

Property operator $\hat{O}$ in basis of Kramers-paired molecular spinors

$$\hat{O} = \sum_{m,n=1}^{P_u} o_{mn}\, a_m^\dagger a_n + \sum_{m=1}^{P_u}\sum_{n=P_u+1}^{P} o_{m\overline{n}}\, a_m^\dagger a_{\overline{n}} + \sum_{m=P_u+1}^{P}\sum_{n=1}^{P_u} o_{\overline{m}n}\, a_{\overline{m}}^\dagger a_n + \sum_{m,n=P_u+1}^{P} o_{\overline{m}\overline{n}}\, a_{\overline{m}}^\dagger a_{\overline{n}}$$

First-term contribution to expectation value

$$W'(\Psi_k)_1 = \sum_{I,J=1}^{\dim\mathcal{F}^{\mathrm{t}}(\mathrm{P,N})} c_{kI}^* c_{kJ} \sum_{m,n=1}^{P_u} o_{mn}^M \left\langle \, | \prod_{p=1}^{N_p\in\mathcal{S}_I} \prod_{\overline{p}=N_p+1}^{N_p\in\mathcal{S}_I+N_{\overline{p}}\in\overline{\mathcal{T}}_I} a_{\overline{p}} a_p \, a_m^\dagger a_n \prod_{q=1}^{N_p\in\mathcal{S}_J} \prod_{\overline{q}=N_p+1}^{N_p\in\mathcal{S}_J+N_{\overline{p}}\in\overline{\mathcal{T}}_J} a_q^\dagger a_{\overline{q}}^\dagger \, | \, \right\rangle$$

[1] S. Knecht, Dissertation, HHU Düsseldorf 2009

$\mathcal{P}$,$\mathcal{T}$-odd Properties as Expectation Values

Interaction constants / enhancement factors for n-electron system

- Electron eEDM interaction constant[2] / enhancement[3]

$$W_d := \frac{1}{\Omega} \left\langle \sum_{j=1}^{n} \gamma_j^0 \, \mathbf{\Sigma}_j \cdot \mathbf{E}_j \right\rangle_{\psi^{(0)}} \approx -\frac{2\imath c}{\Omega \, e\hbar} \left\langle \sum_{j=1}^{n} \gamma_j^0 \gamma_j^5 \, \mathbf{p}_j{}^2 \right\rangle_{\psi^{(0)}}$$

$$E_{\mathsf{eff}} = -\Omega \, W_d \qquad\qquad R \approx R_{\mathsf{lin}} = -\frac{E_{\mathsf{eff}}(E_{\mathsf{ext}})}{E_{\mathsf{ext}}}$$

- S-PS nucleon-electron interaction constant[4] / ratio[5]

$$W_{\mathcal{S}} := \frac{\imath}{\Omega} \frac{G_F}{\sqrt{2}} A(Z) \left\langle \sum_{j=1}^{n} \gamma_j^0 \gamma_j^5 \, \rho_N(\mathbf{r}_j) \right\rangle_{\psi^{(0)}} \qquad S = -\frac{\left\langle \imath \sum_j \gamma_j^0 \, \gamma_j^5 \, \rho_N(\mathbf{r}_j) \right\rangle_{\Psi(E_{\mathsf{ext}})}}{E_{\mathsf{ext}}}$$

---

[2] E. Lindroth, E. Lynn, P.G.H. Sandars, *J. Phys. B: At. Mol. Opt. Phys.* **22** (1989) *559*, stratagem II
TF, M.K. Nayak, *Phys. Rev. A* **88** (2013) *032514*

[3] TF, L.V. Skripnikov, *Symmetry* **12** (2020) *498*

[4] M. Denis *et al.*, *New J. Phys.* **7** (2015) *043005*

[5] TF, M. Jung, *J. High Energy Phys. (JHEP)* **07** (2018) *012*

$\mathcal{P},\mathcal{T}$-odd Properties as Expectation Values

Interaction constants / enhancement factors for n-electron system

- Quadrupole term of nuclear vector potential in terms of nMQM tensor

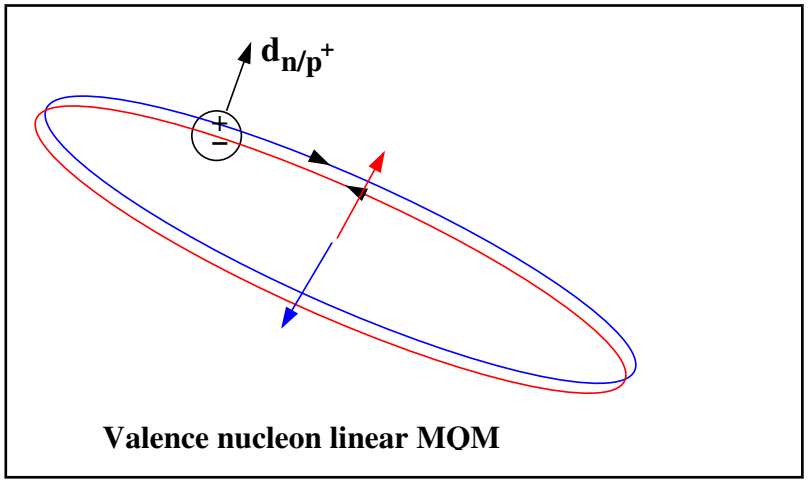
$$\mathbf{A}_Q(\mathbf{r}) = -\sum_{k,n} M_{nk} \frac{1}{2r^5} \sum_{\imath,l} \varepsilon_{\imath l n}\, r_l r_k \mathbf{e}_\imath$$

Nuclear magnetic quadrupole (M)- electronic magnetic field interaction Hamiltonian

$$\hat{H}_{Qe} = -\frac{\boldsymbol{\alpha}\times\mathbf{r}}{2r^5} \cdot (\mathbf{r}\mathbf{M})$$

Nuclear MQM interaction constant (molecules)[6]:

$$W_M = \frac{3}{2\Omega} \left\langle \sum_{j=1}^{n} \left(\frac{\boldsymbol{\alpha}_j \times \mathbf{r}_{jA}}{r_{jA}^5} \right)_z (r_{jA})_z \right\rangle_{\psi_k^{(0)}}$$



[6] TF, M.K. Nayak, *Phys. Rev. A* **93** (2016) *012505*

$\mathcal{P},\mathcal{T}$-Odd and Spectroscopic Constants for X + Ra

	R_e [a.u.]	B_e [cm^{-1}]	D[Debye]	EA(A) [eV]	E_{eff} $\left[\frac{\mathrm{GV}}{\mathrm{cm}}\right]$	W_S [kHz]	W_M $[\frac{10^{33}\mathrm{Hz}}{e\,\mathrm{cm}^2}]$
LiRa(10au)	7.668	0.151	1.36	0.618	**22.2**	**-59.5**	0.652
LiRa(50au)	7.689	0.150	1.34	0.618	**21.7**	**-58.3**	0.641
NaRa	8.703	0.038	0.51	0.548	**12.0**	**-32.2**	0.368
KRa	10.37	0.017	0.39	0.501	**5.44**	**-14.6**	0.167
RbRa	10.75	0.008	0.36	0.486	**5.01**	**-13.6**	0.152
CsRa	11.25	0.006	0.46	0.472	**4.52**	**-12.6**	0.138
FrRa	11.26	0.004	0.24	0.486	**3.44**	**-12.4**	0.137
AgRa	6.241	0.021	4.76	1.304	**63.9**	**-175.1**	1.761

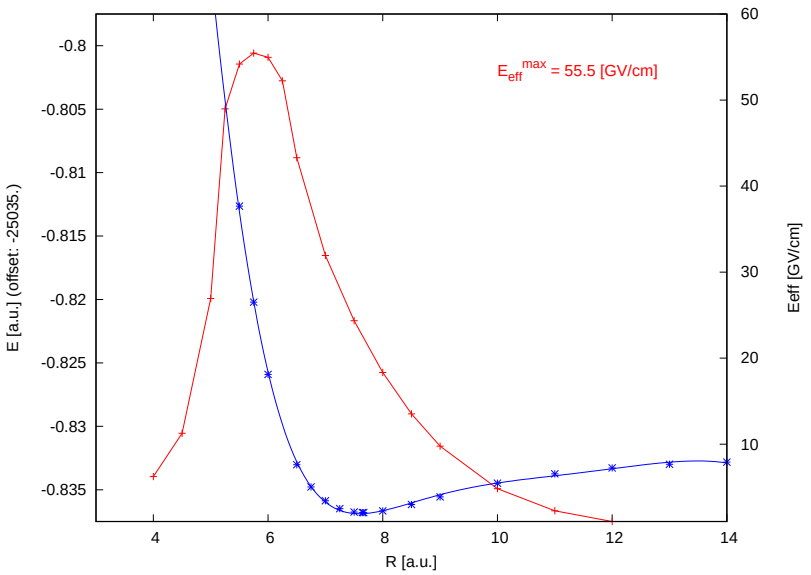
Analytical relationship between E_{eff} and W_S underlying matrix elements[7]

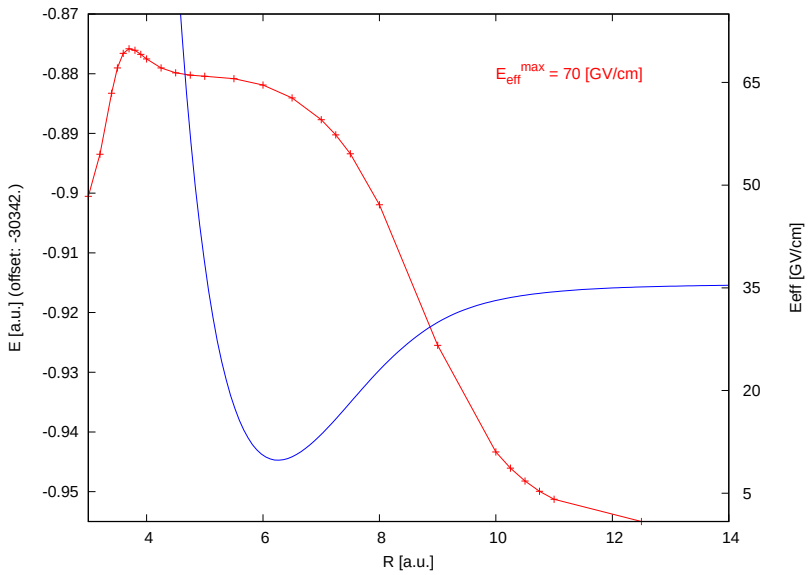
"diamagnetic" systems[8]	$\left[\frac{d_e}{C_S 10^{-18} e\mathrm{cm}}\right]$ Xe	Hg
Ratio $\frac{\langle 0,0\vert 2\imath c\sum_j \gamma_j^0\gamma_j^5\mathbf{p}_j^2\vert p\to s\ 0,0\rangle}{\langle 0,0\vert\frac{AG_F}{\sqrt{2}}\imath\sum_e \gamma_e^0\gamma_e^5\rho(\mathbf{r}_e)\vert p\to s\ 0,0\rangle}$	-158.0	-85.9
Ratio $\frac{\langle 0,0\vert 2\imath c\sum_j \gamma_j^0\gamma_j^5\mathbf{p}_j^2\vert p(s)\to p(s)\ 1,0\rangle}{\langle 0,0\vert\frac{AG_F}{\sqrt{2}}\imath\sum_e \gamma_e^0\gamma_e^5\rho(\mathbf{r}_e)\vert p(s)\to p(s)\ 1,0\rangle}$	-159.2	-85.2

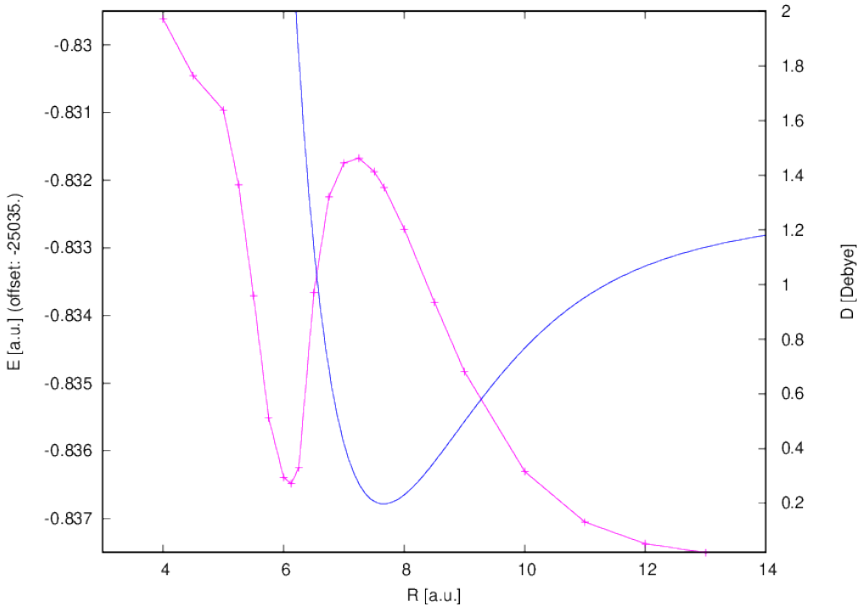
[7] V.A. Dzuba, V.V. Flambaum, C. Harabati, *Phys. Rev. A* **84** (2011) *052108*

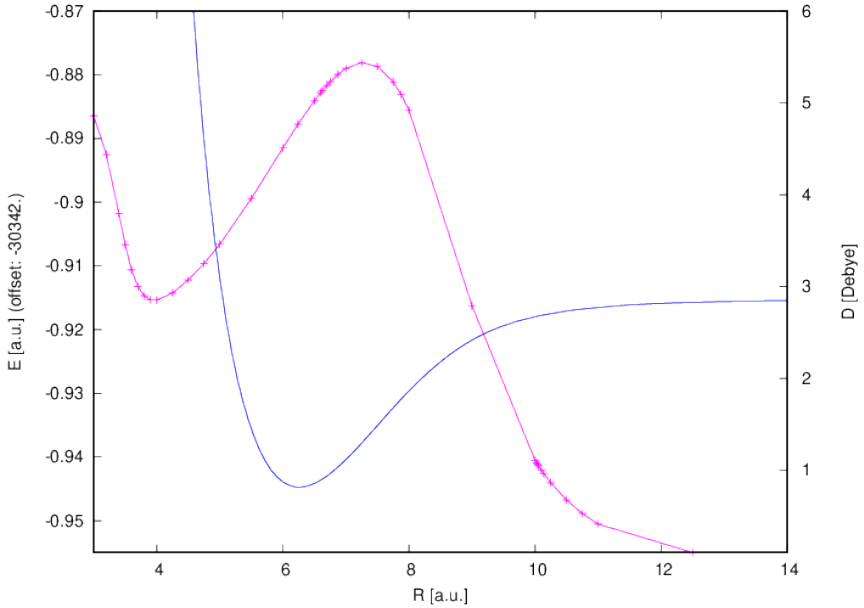
[8] T. F., M. Jung, *Phys. Rev. A* **103** (2021) *012807*

EDM Effective Electric Field : LiRa vs. AgRa









$\mathcal{P},\mathcal{T}$-Odd and Spectroscopic Constants for X + Ra

	R_e [a.u.]	B_e [cm^{-1}]	D[Debye]	EA(A) [eV]	E_{eff} [$\frac{\mathrm{GV}}{\mathrm{cm}}$]	W_S [kHz]	W_M [$\frac{10^{33}\mathrm{Hz}}{e\,\mathrm{cm}^2}$]
LiRa(10au)	7.668	0.151	1.36	0.618	22.2	−59.5	0.652
LiRa(50au)	7.689	0.150	1.34	0.618	21.7	−58.3	0.641
NaRa	8.703	0.038	0.51	0.548	12.0	−32.2	0.368
KRa	10.37	0.017	0.39	0.501	5.44	−14.6	0.167
RbRa	10.75	0.008	0.36	0.486	5.01	−13.6	0.152
CsRa	11.25	0.006	0.46	0.472	4.52	−12.6	0.138
FrRa	11.26	0.004	0.24	0.486	3.44	−12.4	0.137
AgRa	6.241	0.021	4.76	1.304	63.9	−175.1	1.761

Partial charge on partner atom (DCHF)

A	δ_A^- [e] at R_e
Fr	−0.03
Rb	−0.05
Li	−0.08
Ag	−0.24

Rotational constant

$$B_e = \frac{B}{hc} = \frac{\hbar}{4\pi c \mu R_e^2}$$

Electron EDM Using Trapped Ultracold AgRa[9]

State of the Art

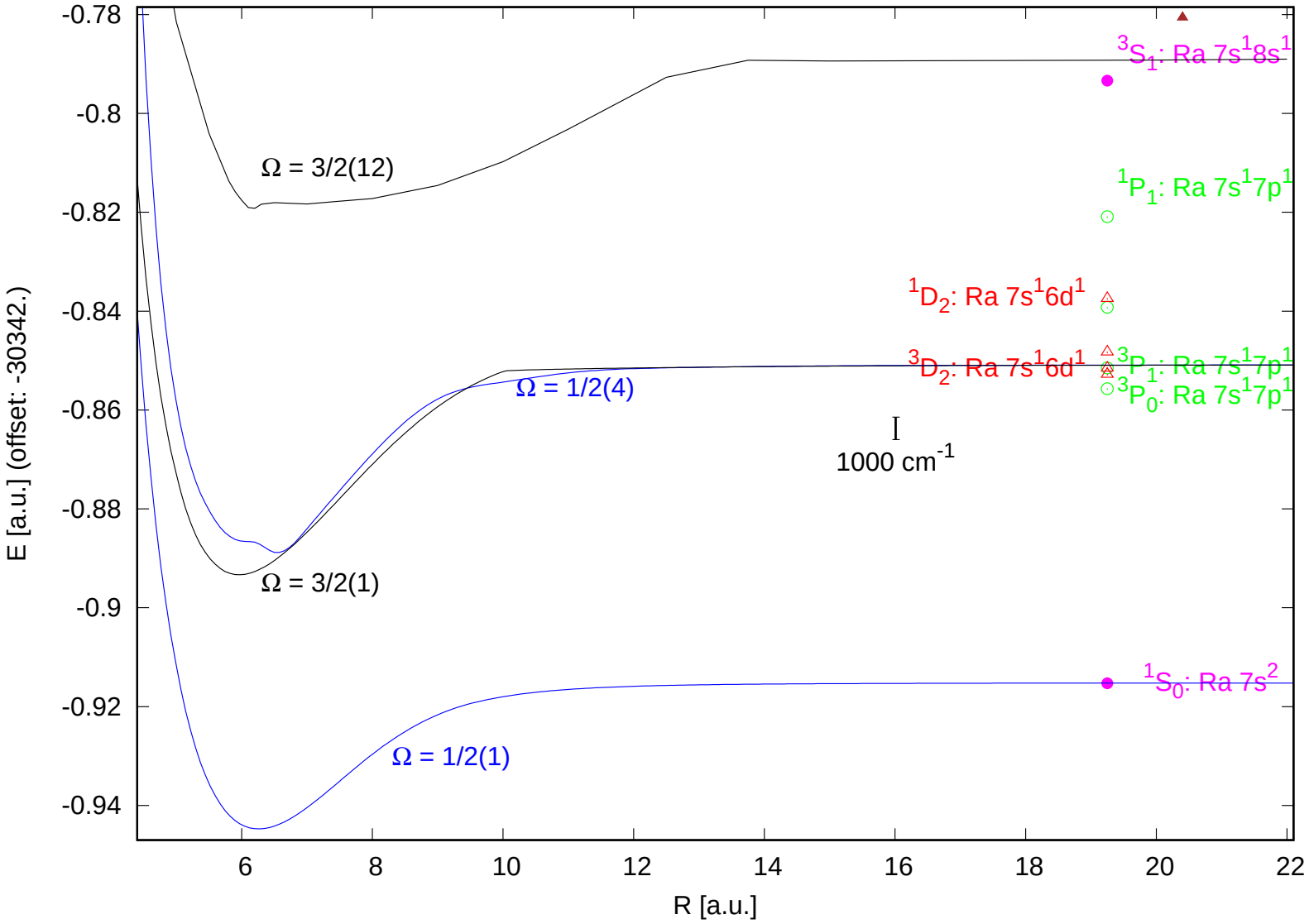
In collaboration with

David DeMille
(Chicago)

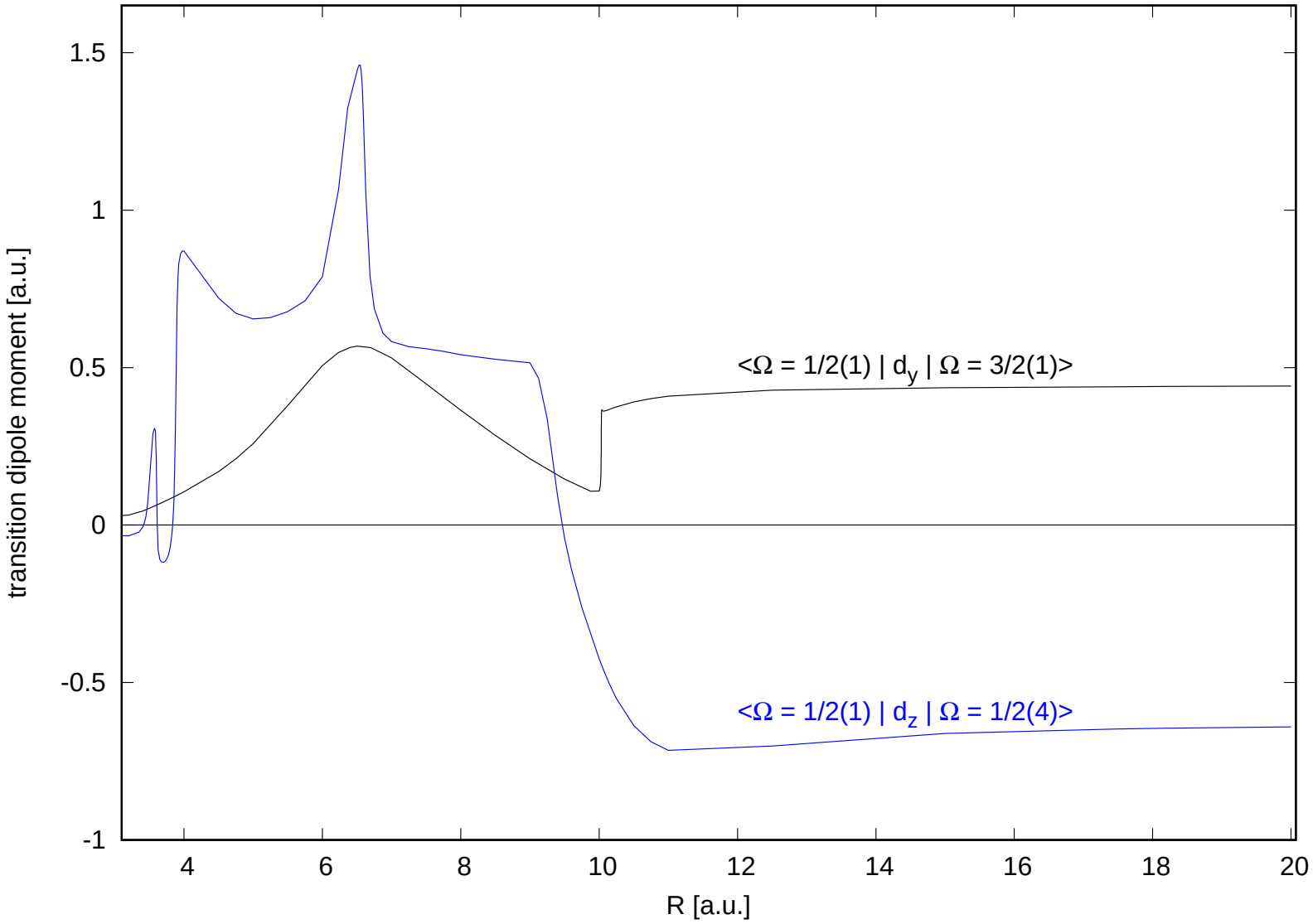
- Important molecular potentials and constants
- Relevant $\mathcal{P},\mathcal{T}$-odd constants at R_e
- Electric transition dipole matrix elements (partially)
- Vibronic transition moments for trapped-bound transitions
- Vibronic transition moments for bound-bound transitions

[9] T. F., O. Grasdijk, D. DeMille, (2021) *in preparation*

PECs for AgRa



E1 Matrix Elements for AgRa



Long-Range Theory – Dispersion Coefficients

- Interaction energy correct to lowest order for heteronuclear neutral atom pair:

  $V(R) \approx -\frac{C_6}{R^6}$

  Hund's case C formulation of leading dispersion coefficient[10]:

  $$C_6(\Omega) = \sum_{j=|J_A-1|}^{J_A+1} \sum_{J=|J_B-1|}^{J_B+1} A_{jJ}(\Omega)\, X_{jJ}$$

  Representation of angular factor:

  $$A_{jJ}(\Omega) = \sum_{\mu m M_J} \left\{ (1+\delta_{\mu 0}) \begin{pmatrix} J_A & 1 & j \\ -M_{J_A} & \mu & m_j \end{pmatrix} \begin{pmatrix} J_B & 1 & J \\ -M_{J_B} & -\mu & M_j \end{pmatrix} \right\}^2$$

  with $\Omega = M_{J_A} + M_{J_B} = m_j + M_J$

  X factor with reduced matrix elements:

  $$X_{jJ} = \sum_{\alpha_l, \alpha_k} \frac{\left|\left\langle \alpha_A\, J_A || \hat{\mathbf{D}} || \alpha_l\, J_l{=}j \right\rangle\right|^2 \left|\left\langle \alpha_B\, J_B || \hat{\mathbf{D}} || \alpha_k\, J_k{=}J \right\rangle\right|^2}{E_l - E_A + E_k - E_B}$$

[10] S.G. Porsev, M.S. Safronova, A. Derevianko, and C.W. Clark, *Phys. Rev. A* **89** (2014) *012711*
S.G. Porsev, M.S. Safronova, A. Derevianko, and C.W. Clark, *Phys. Rev. A* **89** (2014) *022703*

Transition Properties Using Gaussian Basis Sets

- Adapted form of WET using TDM norm:

$$\left\langle \alpha\, J||\hat{\boldsymbol{D}}||\alpha'\, J' \right\rangle = \frac{\left|\left|\left\langle \alpha\, J\, M_J|\hat{\boldsymbol{D}}|\alpha'\, J'\, M'_J \right\rangle\right|\right| \sqrt{2J+1}}{\left\langle J'\, 1\, M'_J\, q|J'\, 1\, J\, M_J \right\rangle}$$

with $q = M_J - M'_J$

Via general definition of CGC

$$\left\langle j_1 j_2 m_{j_1} m_{j_2}|j_1 j_2 j m_j \right\rangle = \delta(m_j, m_{j_1} + m_{j_2}) \sqrt{\frac{(j_1+j_2-j)!(j+j_1-j_2)!(j+j_2-j_1)!(2j+1)}{(j+j_1+j_2+1)!}}$$

$$\times \sum_k \frac{(-1)^k \sqrt{(j_1+m_{j_1})!(j_1-m_{j_1})!(j_2+m_{j_2})!(j_2-m_{j_2})!(j+m_j)!(j-m_j)!}}{k!(j_1+j_2-j-k)!(j_1-m_{j_1}-k)!(j_2+m_{j_2}-k)!(j-j_2+m_{j_1}+k)!(j-j_1-m_{j_2}+k)!}$$

Finally E1 oscillator strengths:

$$f_{IF} = \frac{2}{3} \frac{\varepsilon_F - \varepsilon_I}{2J_I+1} \left| \left\langle \alpha_I\, J_I||\hat{\boldsymbol{D}}||\alpha_F\, J_F \right\rangle \right|^2$$

Transition Properties Using Gaussian Basis Sets

Assessment of Accuracy

Li

	present			experiment (NIST)	literature
Excited state	RME	$\Delta\varepsilon$ [cm^{-1}]	f	$\Delta\varepsilon$ [cm^{-1}]	f
$^2P_{1/2}(2p^1)$	3.3197	14909	0.2495	14903.66	
$^2P_{3/2}(2p^1)$	4.6948	14910	0.4991	14904.00	0.7470 (^2P) [11]
$^2P_{1/2}(3p^1)$	0.1794	30916	0.0015	30925.38	
$^2P_{3/2}(3p^1)$	0.2536	30917	0.0030	30925.38	0.00482 (^2P) [12]
$^2P_{1/2}(4p^1)$	0.3138	39039	0.0058	36469.55	
$^2P_{3/2}(4p^1)$	0.4437	39039	0.0117	36469.55	

Be

	present			experiment (NIST)	literature
excited state	RME	$\Delta\varepsilon$ [cm^{-1}]	f	$\Delta\varepsilon$ [cm^{-1}]	f
$^3P_1(2s^12p^1)$	0.0002	21977	0.0000	21978.93	
$^1P_1(2s^12p^1)$	3.2615	42585	1.3760	42565.35	1.375(^1P) [13]
$^1P_1(2s^13p^1)$	0.2111	60347	0.0082	60187.34	
$^1P_1(2s^14p^1)$	0.1781	70323	0.0068	67034.70	
$^1P_1(2s^15p^1)$	0.2727	87309	0.0197	70120.49	

[11] Z.-C. Yan, M. Tambasco, G.W.F. Drake, *Phys. Rev. A* **57** (1998) *1652*

[12] L. Qu, Z. Wang, B. Li, *Eur. Phys. J. D* **5** (1999) *173*

[13] J. Fleming, M. R. Godefroid, K. L. Bell, A. Hibbert, N. Vaeck, J. Olsen, P. Jönsson, C. Froese Fischer, *J. Phys. B: At. Mol. Opt. Phys.* **29** (1996) *4347*

Dispersion Coefficients Using Gaussian Basis Sets

Assessment of Accuracy

	C_6 [a.u.]	
System/State	present	literature
LiLi $X^1\Sigma_0$	1390	1389 [14]
LiBe $X^2\Sigma_{1/2}$	464	478 [15]

[14] S.G. Porsev, M.S. Safronova, A. Derevianko, and C.W. Clark, *Phys. Rev. A* **89** (2014) *022703*

[15] J. Jiang, Y. Cheng, J. Mitroy, *J. Phys. B: At. Mol. Opt. Phys.* **46** (2013) *125004*

Transition Properties Using Gaussian Basis Sets

Assessment of Accuracy

Na

	present			experiment (NIST)	literature
Excited state	RME	$\Delta\varepsilon$ [cm^{-1}]	f	$\Delta\varepsilon$ [cm^{-1}]	f
$^2P_{1/2}(3p^1)$	3.5865	16799	0.3282	16956.17	0.325 ($^2P_{1/2}$) [16]
$^2P_{3/2}(3p^1)$	5.0724	16817	0.6572	16973.37	0.650 ($^2P_{3/2}$) [16]
$^2P_{1/2}(4p^1)$	0.3101	30001	0.0044	30266.99	
$^2P_{3/2}(4p^1)$	0.4425	30007	0.0089	30272.58	0.016 ± 0.003 (^2P) [17]
$^2P_{1/2}(5p^1)$	0.1422	37122	0.0011	35040.38	
$^2P_{3/2}(5p^1)$	0.2049	37131	0.0024	35042.85	0.0025 ± 0.0005 (^2P) [17]

	C_6 [a.u.]	
System/State	present	literature
NaNa $X^1\Sigma_0$	1642	1564 [16]
NaBe $X^2\Sigma_{1/2}$	514	522 [17]

- $C_6 = 1603$ [a.u.] using exp. data for strongest transition

[16] G. Stephenson, *Proc. Roy. Soc. A* **64** (1951) *458*

[17] P. Erman, J. Brzozowski, and Wm. Hayden Smith *Astrophys. J.* **192** (1974) *59*

Transition Properties Using Gaussian Basis Sets

Assessment of Accuracy

K

	present			experiment (NIST)	literature
Excited state	RME	$\Delta\varepsilon$ [cm^{-1}]	f	$\Delta\varepsilon$ [cm^{-1}]	f
$^2P_{1/2}(4p^1)$	4.2323	12861	0.3499	12985.19	0.330 [18]
$^2P_{1/2}(4p^1)$ (5 a.u.)	4.2238	12857	0.3484		
$^2P_{1/2}(4p^1)$ (SDTQ_SDT 5 a.u.)	4.1328	13195	0.3422		
$^2P_{1/2}(4p^1)$ (+CVC 5 a.u.)	4.2230	12853	0.3481		
$^2P_{1/2}(4p^1)$ (+p 5 a.u.)	4.1919	13240	0.3533		
$^2P_{3/2}(4p^1)$	5.9860	12918	0.7030	13042.90	0.657 [18]
$^2P_{3/2}(4p^1)$ (5 a.u.)	5.9817	12914	0.7018		
$^2P_{3/2}(4p^1)$ (SDTQ_SDT 5 a.u.)	5.8425	13253	0.6871		
$^2P_{3/2}(4p^1)$ (+CVC 5 a.u.)	5.9706	12910	0.6990		
$^2P_{3/2}(4p^1)$ (+p 5 a.u.)	5.9244	13291	0.7085		
$^2P_{1/2}(5p^1)$	0.2797	24412	0.0029	24701.38	
$^2P_{3/2}(5p^1)$	0.4129	24430	0.0063	24720.14	
$^2P_{1/2}(6p^1)$	0.1452	29696	0.0010	28999.27	
$^2P_{3/2}(6p^1)$	0.2197	29715	0.0022	29007.71	

- SDTQ_SDT model halves deviations (residual deviation $\approx 4\%$)

[18] G. Stephenson, *Proc. Roy. Soc. A* **64** (1951) *458*

Transition Properties Using Gaussian Basis Sets

Assessment of Accuracy

Fr

	present			experiment (NIST)	literature
Excited state	RME	$\Delta\varepsilon$ [cm^{-1}]	f	$\Delta\varepsilon$ [cm^{-1}]	f
$^2P_{1/2}(7p^1)$	4.4278	12311	0.3666	12237.41	0.340 [19]
$^2P_{1/2}(7p^1)$ (SDTQ_SDT)	4.3595	12793	0.3693		
$^2P_{1/2}(7p^1)$ (SDTQ_SDT)	"mixed"		0.3532		
$^2P_{1/2}(7p^1)$ (+5d CVC)	4.4584	12381	0.3738		
$^2P_{3/2}(7p^1)$	6.1117	13910	0.7891	13924.00	0.736 [19]
$^2P_{3/2}(7p^1)$ (SDTQ_SDT)	6.0164	14429	0.7932		
$^2P_{3/2}(7p^1)$ (SDTQ_SDT)	"mixed"		0.7655		
$^2P_{3/2}(7p^1)$ (+5d CVC)	6.1548	13949	0.8025		
$^2P_{1/2}(8p^1)$	0.2879	23052	0.0029	23112.96	
$^2P_{3/2}(8p^1)$	0.9112	23613	0.0298	23658.31	
$^2P_{1/2}(9p^1)$	0.1487	29579	0.0010	27118.21	
$^2P_{3/2}(9p^1)$	0.6109	30473	0.0173	27366.20	

- Higher excitation ranks do not lead to improvement for f
  But use of RME and $\Delta\varepsilon$(exp) gives more accurate results
- CVCs lead to slight increase of f

[19] V.A. Dzuba and V.V. Flambaum and O.P. Sushkov, *Phys. Rev. A* **51** (1995) *3454*

Transition Properties for AgRa

Ag

	experiment		present		
Transition	ΔE [cm^{-1}] (NIST)	f_{IF} [20]	ΔE [cm^{-1}]	RME [a.u.]	f_{IF}
$^2S_{1/2}(5s) - {}^2P_{1/2}(5p)$	29552.1	0.232	28401	2.697	0.314
			28904	2.51 (+T)	0.276
$^2S_{1/2}(5s) - {}^2P_{3/2}(5p)$	30472.7	0.476	29240	3.812	0.645
			29734	3.54 (+T)	0.566
$^2S_{1/2}(5s) - {}^2P_{1/2}(6p)$	48297.4	0.005	48883	0.126	0.001
$^2S_{1/2}(5s) - {}^2P_{3/2}(6p)$	48500.8	0.011	49352	0.303	0.007
$^2S_{1/2}(5s) - {}^2P_{1/2}(7p)$	54041.0	0.001			
$^2S_{1/2}(5s) - {}^2P_{3/2}(7p)$	54121.1	0.003			
$^2S_{1/2}(5s) - \infty$	61106.0	0.272			

[20] J. Carlsson, P. Jønsson, L. Sturesson, *Z. Phys. D* **16** (1990) *87*
G. J. Bengtsson and J. Larsson and S. Svanberg, *Phys. Rev. A* **42** (1990) *5457*
G. J. Bengtsson and P. Jønsson and J. Larsson and S. Svanberg, *Z. Phys. D* **22** (1991) *437*
S. Civiš and I. Matulková and J. Cihelka and P. Kubelik and K. Kawaguchi and V. E. Chernov, *Phys. Rev. A* **82** (2010) *022502*

Transition Properties for AgRa

Ra

	literature[21]			present		
Transition	ΔE [cm^{-1}] (NIST)	RME [a.u.]	f_{IF}	ΔE [cm^{-1}]	RME [a.u.]	f_{IF}
$^1S_0(7s^2) - {}^3P_1(7s7p)$	13999.4	1.218	0.063	13713	0.865	0.031
$^1S_0(7s^2) - {}^1P_1(7s7p)$	20715.6	5.504	1.906	22922	5.835	2.370
$^1S_0(7s^2) - J' = 1$				33511	1.429	0.208
$^1S_0(7s^2) - J' = 1$				46545	0.664	0.062
$^3P_1(7s7p) - {}^3D_1(7s6d)$	−283.6	2.574	−0.002	215	2.571	0.001
$^3P_1(7s7p) - {}^3D_2(7s6d)$	−5.5	4.382	−0.000	510	4.490	0.010
$^3P_1(7s7p) - {}^1D_2(7s6d)$	3081.9	0.344	0.000	5317	0.013	0.000
$^3P_1(7s7p) - {}^3S_1(7s8s)$	12754.7	3.890	0.195	13109	3.792	0.191
$^3P_1(7s7p) - {}^3F_2(6d^2)$	15518[b]	0.542	0.005			
$^3P_1(7s7p) - {}^1D_2(6d^2)$	16571[b]	1.274	0.027			
$^3P_1(7s7p) - (J = 1)$				30420	1.300	0.052
$^3P_1(7s7p) - (J = 1)$				32474	3.113	0.319
$^3P_1(7s7p) - (J = 2)$				32615	4.882	0.787
$^3P_1(7s7p) - (J = 2)$				46215	2.425	0.275
$^3P_1(7s7p) - \infty$				28573.4	3.393	0.333

[21] V. A. Dzuba, V. V. Flambaum, *J. Phys. B: At. Mol. Opt. Phys.* **40** (2007) *227*

Dispersion Coefficients for AgRa[22]

Diss. channel	Ag($^2S_{1/2}$) - Ra($^1S_0$)	Ag($^2S_{1/2}$) - Ra($^3P_1$)			Ag($^2S_{1/2}$) - Ra($^3S_1$)
Molecular state	$\Omega = 1/2(1)$	$\Omega = 1/2(3)$	$\Omega = 1/2(4)$	$\Omega = 3/2(1)$	$\Omega = 3/2(12)$
C_6 [a.u.]	853	733	953	958	4253

[22] T. F., O. Grasdijk, D. DeMille, (2021) *in preparation*

Toward Nuclear Schiff moment with trapped ultracold molecules

In collaboration with

Mickaël Hubert
(EPITA, Toulouse)

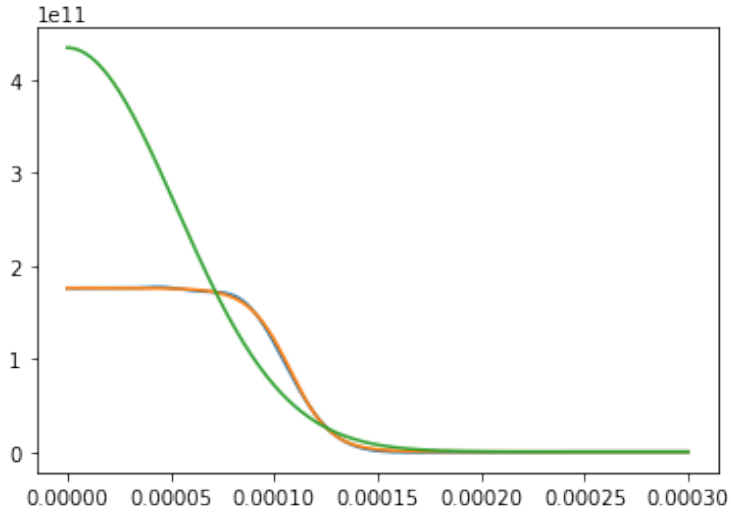
- Two models for nuclear density[23]:

  $\rho_G(r) = Z\left(\frac{\zeta}{\pi}\right)^{\frac{3}{2}} e^{-\zeta r^2}$ (Gaussian)

  $\rho_F(r) = \frac{a}{1+e^{\frac{r-C}{t}}}$ (Fermi)

- Fermi: Expand into Gaussian basis set

  $\rho_F(r) = \sum_{i=1}^{n} \alpha_i e^{-\zeta_i r^2}$ optimize $\{\alpha_i, \zeta_i\}$



[23] M. Hubert and T. F., (2021) *unpublished*

A Glance at Atoms: Xe Schiff Interaction

- Energy shift due to atomic Schiff moment interaction[24]:

$$\Delta\varepsilon_{\mathsf{SM}} = -S_z \frac{3}{B} \left\langle \hat{z}\, \rho(\mathbf{r}) \right\rangle_{\psi(E_{\mathsf{ext}})} \qquad \text{with} \qquad B = \int_0^\infty \rho(\mathbf{r}) r^4 dr$$

- Definition of the atomic interaction constant

$$\alpha_{\mathsf{SM}} := \frac{\Delta\varepsilon_{\mathsf{SM}}}{S_z\, E_{\mathsf{ext}}} = \frac{-\frac{3}{B} \left\langle \hat{z}\, \rho(\mathbf{r}) \right\rangle_{\psi(E_{\mathsf{ext}})}}{E_{\mathsf{ext}}}$$

	$\alpha_{\mathsf{SM}} \left[10^{-17} \frac{e\mathsf{cm}}{e\mathsf{fm}^3}\right]$		
Model	Gauss	Fermi	$\varepsilon_{\mathsf{DCHF}}$
Dyall-cvDZ	-1.220	-1.005	-7446.876435682244
Dyall-cvTZ	-0.379	-0.391	-7446.895053544852
Dyall-cvQZ	0.318	0.234	-7446.895410571442
Dyall-cvQZ-79s67p	0.375	0.293	-7446.895402055156
Dyall-cvQZ-79s67p/SD8-5au	0.354	0.275	
Dzuba *et al.*[25] (RPA, 2002)	0.38		
Ramachandran *et al.*[26] (CPHF, 2014)	0.374		
Sakurai *et al.*[27] (RNCCSD, 2019)	0.32		

[24] V.V. Flambaum, V.A. Dzuba, H.B. Tran Tan, *Phys. Rev. A* **101** (2020) *042501*

[25] V.A. Dzuba, V.V. Flambaum, J.S.M. Ginges, and M.G. Kozlov, *Phys. Rev. A* **66** (2002) *012111*

[26] S. M. Ramachandran and K. V. P. Latha, *Phys. Rev. A* **90** (2014) *042503*

[27] A. Sakurai, B.K. Sahoo, K. Asahi, B.P. Das, *Phys. Rev. A* **100** (2019) *020502*

Schiff Interaction in Molecules

TlF

Model / $R = 3.94$ a.u.	$W_{\mathrm{SM}} = \frac{\Delta\varepsilon_{\mathrm{SM}}}{S_z}$(Tl) [a.u.]
cvQZ/DCHF	42877
cvQZ/SD8_4au	38280
cvQZ/SD18_4au	38648
cvQZ/SD10_SDT18_4au	37341
Flambaum *et al.*[28] (CC, 2002)	40539
Abe *et al.*[29] (CC, 2020)	*41136*

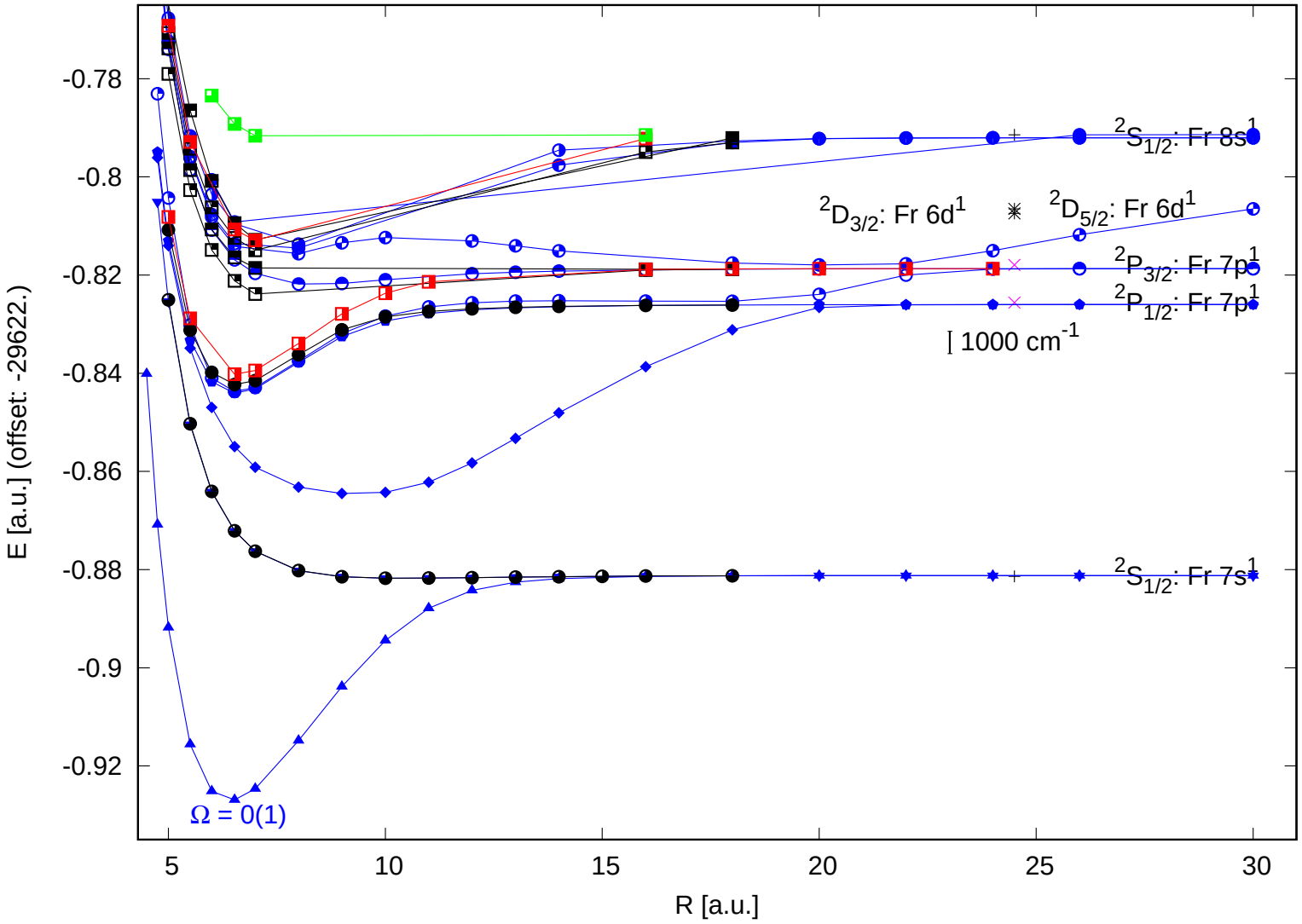
AgFr

Model (at $R = 6.525$ a.u.)	$W_{\mathrm{SM}} = \frac{\Delta\varepsilon_{\mathrm{SM}}}{S_z}$(Fr) [a.u.]
cvTZ/DCHF	26692
cvTZ/S18_CAS2in22_SD20_4au	26075
cvQZ/DCHF	27915
cvQZ/S18_CAS2in22_SD20_4au	27328
cvTZ-cvQZ/DCHF	27922
cvTZ-cvQZ/S18_CAS2in22_SD20_4au	27272

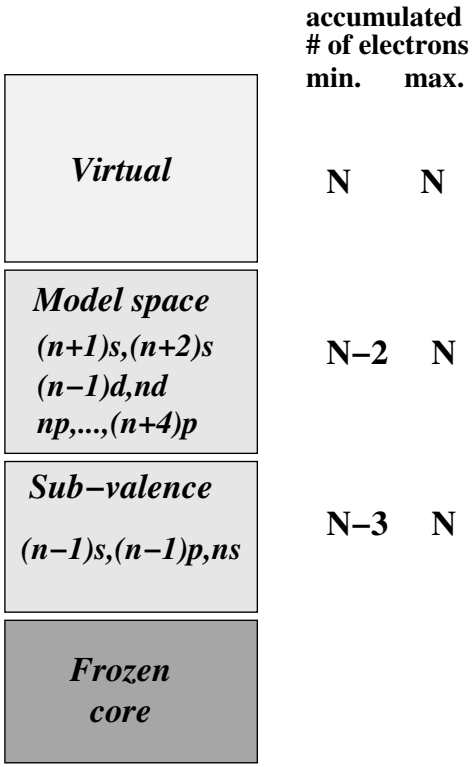
[28] V.V. Flambaum, V.A. Dzuba, H.B. Tran Tan, *Phys. Rev. A* **101** (2020) *042501*
A.N. Petrov, N.S. Mosyagin, T.A. Isaev, A.V. Titov, V.F. Ezhov, E. Eliav, and U. Kaldor, *Phys. Rev. Lett.* **88** (2002) *073001*

[29] M. Abe, T. Tsutsui, J. Ekman, M. Hada, B.P. Das, *Mol. Phys.* **118** (2020) *e1767814*

Schiff Interaction in Molecules: AgFr PEC



Appendix : Technical Definition of Atomic Wavefunctions



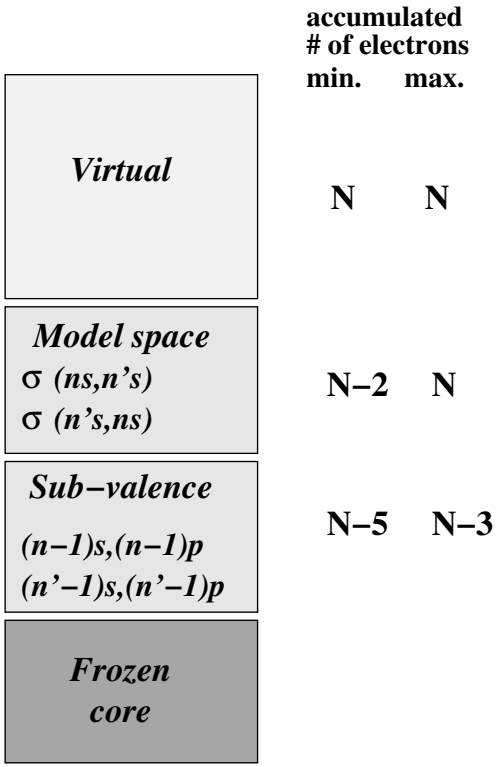
Li $(n = 2)$: $N = 3$, no model space, no frozen core (FCI), Sub-valence is $1s, 2s$ only;

Na $(n = 3)$: $N = 11$, no frozen core; Single excitations out of the $1s$ shell added to expansion for enhanced convergence;

K-Fr $(n = 4 \ldots 7)$: $N = 9$, $np, \ldots, (n+4)p$ for p functions in model space;

The cutoff for the virtual space is set to 100 [a.u.] for all atoms except for Fr where is it 5 [a.u.]

Appendix : Technical Definition of Molecular Wavefunctions



Up to two holes are allowed in the sub-valence spinors which accounts for correlation effects among the sub-valence electrons and with the valence electrons. The model space is restricted to the valence spinors where all occupations are allowed. The cutoff for the virtual space is set to 10 [a.u.]