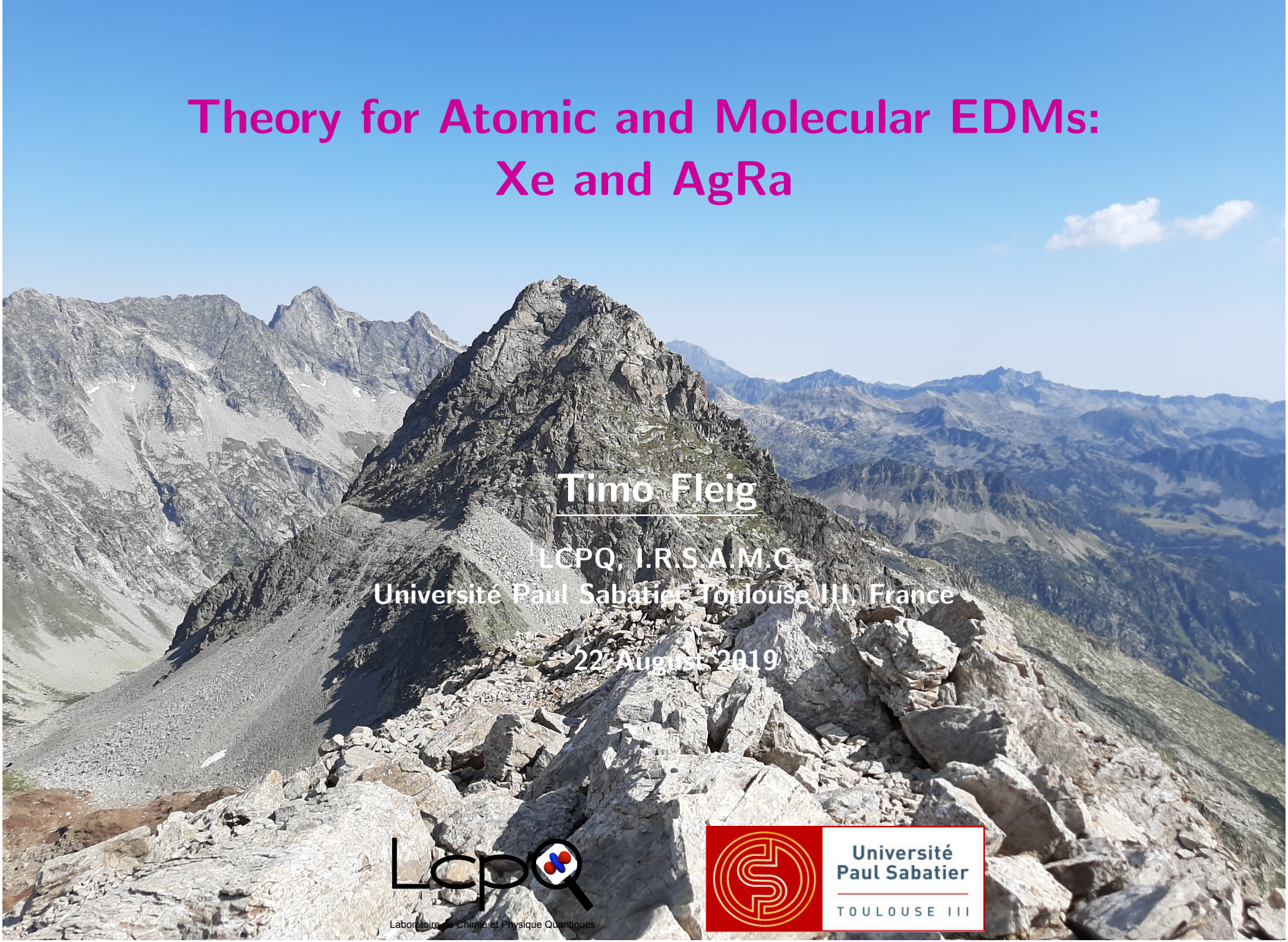
Theory for Atomic and Molecular EDMs: Xe and AgRa



Timo Fleig

LCPQ, I.R.S.A.M.C.

Université Paul Sabatier Toulouse III, France

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Laboratoire de Chimie et Physique Quantiques

Université Paul Sabatier TOULOUSE III

Outline

- Relativistic many-body methods for atomic and molecular structure
- Atomic EDMs : ^{129}Xe
- Molecular EDMs : AgRa

Outline

- **Relativistic many-body methods for atomic and molecular structure**
- Atomic EDMs : ^{129}Xe
- Molecular EDMs : AgRa

Atomic Electric Dipole Moment

- Definition:[1]

$$d_a = -\lim_{E_{\rm ext}\to 0}\left[\frac{\partial(\Delta\varepsilon_{\not P\not T})}{\partial E_{\rm ext}}\right]$$

$\Delta\varepsilon_{\not P\not T}$ is **some** P,T-odd energy shift.

- Example: Lorentz covariant electron EDM interaction

$$\hat{H}_{\rm EDM} = \imath\frac{d_e}{2}\gamma^0\gamma^5\,\frac{\imath}{2}\left(\gamma^\mu\gamma^\nu - \gamma^\nu\gamma^\mu\right)\,F_{\mu\nu}$$

$$\{F_{\mu\nu}\} = \{\partial_\mu A_\nu - \partial_\nu A_\mu\} = \begin{pmatrix} 0 & E_x & E_y & E_z \\ -E_x & 0 & -B_z & B_y \\ -E_y & B_z & 0 & -B_x \\ -E_z & -B_y & B_x & 0 \end{pmatrix}$$

$$\hat{H}_{\rm EDM} = -d_e\gamma^0\left[\boldsymbol{\Sigma}\cdot\mathbf{E} + \imath\boldsymbol{\alpha}\cdot\mathbf{B}\right]$$

- And so

$$d_a = \lim_{E_{\rm ext}\to 0}\frac{\partial}{\partial E_{\rm ext}} d_e\,\left\langle\gamma^0\left[\boldsymbol{\Sigma}\cdot\mathbf{E} + \imath\boldsymbol{\alpha}\cdot\mathbf{B}\right]\right\rangle_{\psi(E_{\rm ext})}$$

[1] E.D. Commins, *Adv. Mol. Opt. Phys.* **40** (1999) *1*

Atomic Electric Dipole Moment

- With the definitions

$$(E+B)_{\text{eff}} = -\left\langle \gamma^0 \left[\boldsymbol{\Sigma} \cdot \mathbf{E} + \imath \boldsymbol{\alpha} \cdot \mathbf{B} \right] \right\rangle_{\psi(E_{\text{ext}})}$$

$$R := \frac{d_a}{d_e} \qquad\qquad R_{\text{lin}} := -\frac{\Delta(E+B)_{\text{eff}}}{\Delta E_{\text{ext}}} = -\frac{(E+B)_{\text{eff}}(2)-(E+B)_{\text{eff}}(1)}{E_{\text{ext}}(2)-E_{\text{ext}}(1)}$$

- the linear-regime enhancement is:

$$R \approx R_{\text{lin}} = -\frac{(E+B)_{\text{eff}}}{E_{\text{ext}}}$$

- $E_{\text{ext}} \in \{10^{-5}, \ldots, 10^{-4}\}$ a.u.

  Corresponding expressions for other P,T-odd effects (Ne-SPS, Ne-TPT, Schiff, etc.)

  Magnetic EDM term will be of importance for constraints[2] on bounds from Xe (near future)

[2] T. Chupp, M. Ramsey-Musolf, *Phys. Rev. C* **91** (2015) *035502*
W. Dekens, J. de Vries, M. Jung, K.K. Vos, *J. High En. Phys.* **1** (2019) *069*

Atomic and Molecular Correlated Wavefunctions

Hamiltonians

- Dirac-Coulomb Hamiltonian + external electric field (atoms)

$$\hat{H}^{\text{Dirac-Coulomb}} + \hat{H}^{\text{Int-Dipole}} = \sum_i^n \left[c\,\boldsymbol{\alpha}_i \cdot \mathbf{p}_i + \beta_i c^2 + \frac{Z}{r_i} \mathbb{1}_4 \right] + \sum_{i,j>j}^n \frac{1}{r_{ij}} \mathbb{1}_4 + \sum_i^n \mathbf{r}_i \cdot \mathbf{E}_{\text{ext}}\, \mathbb{1}_4$$

- Dirac-Coulomb Hamiltonian operator (molecules)

$$\hat{H}^{DC} = \sum_i^n \left[c\,\boldsymbol{\alpha}_i \cdot \mathbf{p}_i + \beta_i c^2 + \sum_A^N \frac{Z}{r_{iA}} \mathbb{1}_4 \right] + \sum_{i,j>i}^n \frac{1}{r_{ij}} \mathbb{1}_4 + \sum_{A,B>A}^N V_{AB}$$

- Dirac-Coulomb-Gaunt Hamiltonian operator (molecules)

$$\hat{H}^{DCG} = \sum_i^n \left[c\,\boldsymbol{\alpha}_i \cdot \mathbf{p}_i + \beta_i c^2 + \sum_A^N \frac{Z}{r_{iA}} \mathbb{1}_4 \right] + \sum_{i,j>i}^n \left(\frac{1}{r_{ij}} \mathbb{1}_4 - \frac{1}{2} \frac{\vec{\alpha}_i \vec{\alpha}_j}{r_{ij}} \right) + \sum_{A,B>A}^N V_{AB}$$

Atomic and Molecular Correlated Wavefunctions

- All-electron Dirac-Coulomb Hartree-Fock (DC(G)HF) calculation
  set of time-reversal paired 4-spinors $\hat{K}\varphi_i = \varphi_{\bar{i}}$ and $\hat{K}\varphi_{\bar{i}} = -\varphi_i$

$$\hat{K}(n) := e^{-\frac{i}{2}\pi\left(\sum_{j=1}^{n} \boldsymbol{\sigma}\otimes\mathbb{1}_2(j)\right)\cdot\vec{e}_y} \prod_{j=1}^{n} \hat{K}_0(j)$$

- Expansion and variation[3] in n-electron sector of Fock space

$$|\psi_k\rangle = \sum_{I=1}^{\dim\mathcal{F}^{\mathrm{t}}(\mathrm{M},\mathrm{n})} c_{kI}\,(\mathcal{S}\overline{\mathcal{T}})_I\,|\ \rangle$$

  unbarred (Kramers up) string $\mathcal{S} = a_i^\dagger a_j^\dagger a_k^\dagger \ldots$
  barred (Kramers down) string $\overline{\mathcal{S}} = a_{\bar{l}}^\dagger a_{\bar{m}}^\dagger a_{\bar{n}}^\dagger \ldots$

  Linear expansion: **Configuration Interaction**

  Exponential expansion: Coupled Cluster

[3] S. Knecht, H.J.Aa. Jensen, T.F., *J Chem Phys* **132** (2010) *014108*

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

using correlated wavefunctions

Expectation values over relativistic Configuration Interaction wavefunctions[4]

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

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

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

First-term contribution to expectation value

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

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

Tensor-Pseudotensor $\mathcal{P},\mathcal{T}$-odd Nucleon-Electron Interaction

Lagrangian density[5] for Ne neutral weak current ($\sigma_{\rho\sigma} = \frac{\imath}{2}\left(\gamma^\rho\gamma^\sigma - \gamma^\sigma\gamma^\rho\right)$):

$$\mathcal{L}_{\text{Ne-TPT}} = \frac{1}{2}\frac{G_F}{\sqrt{2}} C_T \sum_N \varepsilon^{\mu\nu\rho\sigma}\, \overline{\psi}_N \Sigma_{N\,\mu\nu} \psi_N \; \overline{\psi}\sigma_{\rho\sigma}\psi$$

Corresponding effective first-quantized Hamiltonian:

$$\hat{H}^{\text{eff}}_{\text{Ne-TPT}} = -\frac{1}{2}\frac{G_F}{\sqrt{2}}\, C_T\, \rho_N(\mathbf{r})\gamma^0 \varepsilon^{\mu\nu\rho\sigma}\Sigma_{N\,\mu\nu}\sigma_{\rho\sigma}$$

Using $\frac{1}{2}\varepsilon^{\mu\nu\kappa\lambda}\sigma_{\kappa\lambda} = -\imath\gamma^5\sigma^{\mu\nu}$ we get:

$$\hat{H}^{\text{eff}}_{\text{Ne-TPT}} = \frac{\imath G_F}{\sqrt{2}}\, C_T\, \rho(\mathbf{r})\gamma^0\Sigma_{N\,\mu\nu}\gamma^5\sigma^{\mu\nu}$$

Since $\Sigma_{N\,\mu\nu}\gamma^5\sigma^{\mu\nu} = 2\gamma^0\boldsymbol{\Sigma}_N\cdot\boldsymbol{\gamma}$ it follows that

$$\hat{H}^{\text{eff}}_{\text{Ne-TPT}} = \imath G_F\sqrt{2}\, C_T\, \boldsymbol{\Sigma}_N\cdot\boldsymbol{\gamma}\rho(\mathbf{r})$$

Nuclear state chosen as $|I, M_I = I\rangle$ gives many-electron Hamiltonian:

$$\hat{H}^{\text{eff}}_{\text{Ne-TPT}} = \imath G_F\sqrt{2}\, C_T\, \langle\Sigma_N\rangle_{\Psi_N} \sum_{j=1}^{n} \left(\gamma_j\right)^3\, \rho_N(\mathbf{r}_j)$$

[5] K. Yanase, N. Yoshinaga, K. Higashiyama, N. Yamanaka *Phys. Rev. D* **99** (2019) *075021*

Scalar-Pseudoscalar $\mathcal{P},\mathcal{T}$-odd Nucleon-Electron Interaction

via magnetic hyperfine interaction

Solve for $K \in$ CI STATES

$$\left[\sum_j^N \left[c\,\boldsymbol{\alpha}_j\cdot\mathbf{p}_j+\beta_j c^2+\frac{Z}{r_j}\mathbb{1}_4\right]+\sum_{j,k>j}^N \frac{1}{r_{jk}}\mathbb{1}_4+\sum_j \mathbf{r}_j\cdot\mathbf{E}_{\text{ext}}\,\mathbb{1}_4\right]\left|\psi_K^{(0)}\right\rangle=\varepsilon_K^{(0)}\left|\psi_K^{(0)}\right\rangle$$

First-order hyperfine-perturbed CI wavefunctions:

$$\left|\psi_J^{(1)}\right\rangle=\left|\psi_J^{(0)}\right\rangle+\sum_{K\neq J}\frac{\left\langle\psi_K^{(0)}\right|-\frac{1}{2c\,m_p}\frac{\mu\boldsymbol{I}}{I}\cdot\sum_{i=1}^n\frac{\boldsymbol{\alpha}_i\times\boldsymbol{r}_i}{r_i^3}\left|\psi_J^{(0)}\right\rangle}{\varepsilon_J^{(0)}-\varepsilon_K^{(0)}}\left|\psi_K^{(0)}\right\rangle$$

To leading order the SPS-ne energy shift is

$$(\Delta\varepsilon)_J=\frac{1}{\langle\psi_J^{(1)}|\psi_J^{(1)}\rangle}\left\langle\hat{H}_{\text{S-PS-ne}}\right\rangle_{\psi_J^{(1)}}$$

Atomic EDM due to Ne-SPS interaction $d_a=\alpha_{C_S}C_S$ and so

$$\alpha_{C_S}(\psi_J)=\frac{-A\frac{G_F}{\sqrt{2}}}{E_{\text{ext}}\left\langle\psi_J^{(1)}\middle|\psi_J^{(1)}\right\rangle}\left[\sum_{K\neq J}\frac{\left\langle\psi_K^{(0)}\middle|\hat{H}_{\text{HF}}\middle|\psi_J^{(0)}\right\rangle\left\langle\psi_J^{(0)}\middle|\imath\sum_e\gamma_e^0\gamma_e^5\rho(\mathbf{r}_e)\middle|\psi_K^{(0)}\right\rangle}{\varepsilon_J^{(0)}-\varepsilon_K^{(0)}}+h.c.\right]$$

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

Interaction constants / enhancement factors for n-electron system

- Electron eEDM interaction constant[6] / enhancement

$$W_d := \frac{2\imath c}{\Omega\, e\hbar} \left\langle \sum_{j=1}^{n} \gamma_j^0 \gamma_j^5\, \vec{p}_j{}^{\,2} \right\rangle_{\psi_k^{(0)}} \qquad E_{\text{eff}} = -\Omega\, W_d \qquad R \approx R_{\text{lin}} = -\frac{E_{\text{eff}}}{E_{\text{ext}}}$$

- S-PS nucleon-electron interaction constant / ratio

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

- T-PT nucleon-electron interaction constant

$$R_T = \sqrt{2} G_F \left\langle \Sigma_N \right\rangle_{\Psi_N} \left\langle \psi_I^{(0)} \left| \imath \sum_{j=1}^{n} (\gamma_j)^3\, \rho(\mathbf{r}_j) \right| \psi_I^{(0)} \right\rangle$$

[6] E. Lindroth, E. Lynn, P.G.H. Sandars, *J. Phys. B: At. Mol. Opt. Phys.* **22** (1989) *559*, stratagem II

Generalized Active Spaces

Parameterization of the correlated wavefunction

		# of Kramers pairs	accumulated # of electrons min.	max.
III	*(100 a.u.)* *Virtual Kramers pairs*	111	32	32
II	*5s5p*	4	29	32
I	*4s4p* *3s3p* *2s2p*	12	22	24
	Frozen core	(11)		

Outline

- Relativistic many-body methods for atomic and molecular structure
- **Atomic EDMs : ^{129}Xe**

  $|d_{\mathsf{Xe}}| < 1.5 \times 10^{-27} e$ cm. Allmendinger *et. al.* (Mainz, Heidelberg), *Phys. Rev. A* **100** (2019) *022505*

  $|d_{\mathsf{Xe}}| < 4.81 \times 10^{-27} e$ cm. Sachdeva *et. al.* (Ann Arbor *et. al.*), arXiv:1902.02864 [physics.atom-ph] (2019)

  Sato *et. al.* (Tokyo *et. al.*), *Hyperfine Interact.* **230** (2015) *147*

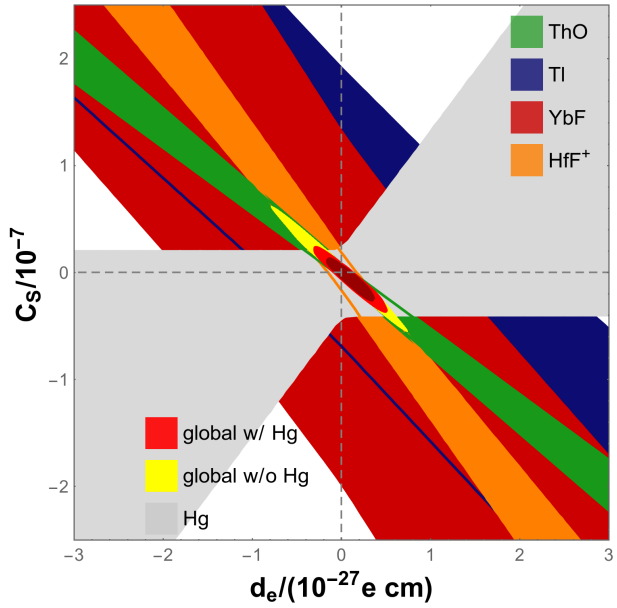
- Molecular EDMs : AgRa

^{129}Xe EDM

Leading[7] (and Subleading) Contributions

$$d_{\mathrm{Xe}} = \rho_Z^N d_N + \alpha_S S - \alpha_{C_T} C_T \; (+\alpha_{C_S} C_S + \alpha_{d_e} d_e)$$

- Atomic coefficients for leading contributions available (α_{C_T}) and in progress (α_S)
- Subleading contributions affect constraints obtained from global fits[8]



Example: Electron EDM and SPS-Ne coupling

- Measurements and calculations on systems with different ratios of atomic/molecular coefficients
- Global fit[9] constrains multiple possible EDM sources

[7] T. Chupp, M. Ramsey-Musolf, *Phys. Rev. C* **91** (2015) *035502*
[8] W. Dekens, J. de Vries, M. Jung, K.K. Vos, *J. High En. Phys.* **1** (2019) *069*
[9] T. F., M. Jung, *J. High En. Phys.* **7** (2018) *012*

A digression on the TI EDM

Leading contributions to **EDM** of a **paramagnetic atom**

$$d_a = R\, d_e + \alpha_{C_S}\, C_S \qquad\qquad \alpha_{C_S} = S\, A\, \frac{G_F}{\sqrt{2}}$$

Enhancement factor and S ratio:

$$R := \frac{\left\langle \sum_j \gamma_j^0\, \mathbf{\Sigma}_j \cdot \mathbf{E}_j \right\rangle_\Psi}{E_{\text{ext}}} \qquad\qquad S := -\frac{\left\langle \imath \sum_j \gamma_j^0\, \gamma_j^5\, \rho_N(\mathbf{r}_j) \right\rangle_\Psi}{E_{\text{ext}}}$$

Model for **TI atom**	R	S [a.u.]
vDZ/SD18_CAS_3in3_SDT21/10au	-473	-331
vDZ/SD18_CAS_3in3_SDT21/20au	-479	-335
vDZ/SD8_SDT10_CAS_3in3_SDT21/10au	-471	-331
vDZ/SD18_CAS_3in3_SDTQ21/10au	-469	-329
vTZ/SD18_CAS_3in3_SDT21/20au	-541	-383
vQZ/SD18_CAS_3in3_SDT21/35au[10]	-562	-398
CCSD(T) 53 + core corr. (Skripnikov)[10]	-557	
Literature values		
Porsev *et al.*, *Phys. Rev. Lett.* **108** (2012) *173001*	-573	-411
Nataraj *et al.*, *Phys. Rev. Lett.* **106** (2011) *200403*	-470	
Dzuba *et al.*, *Phys. Rev. A* **80** (2009) *062509*	-582	
Liu *et al.*, *Phys. Rev. A* **45** (1992) *R4210*	-585	

[10] T. F., L.V. Skripnikov, (2019) *in preparation.*

Tensor-Pseudotensor $\mathcal{P},\mathcal{T}$-odd Nucleon-Electron Interaction

Results for ^{129}Xe

Model/virtual cutoff (vDZ,vTZ,vQZ) [*a.u.*]	R_T [10^{-20} $\langle\sigma_N\rangle$ e cm]		
	Basis set		
	vDZ	vTZ	vQZ
RPA/-	0.382	0.473	0.485
SD8/80,100,60	0.360	0.438	0.453
SDT8/80,100,60	0.360	0.435	0.450
SDTQ8/80,12,60	0.357	0.431	
SD16/80,100,60	0.406	0.481	0.496
SD8_SDT16/80,100,60	0.405	0.477	
SD18/80,100,60		0.453	
SD24/80,100,60	0.421	0.497	0.514
SD26/80,100,60		0.493	
S16_SD32/80,100,60		0.507	
SD32/80,100,60	0.431	0.508	0.525
SD36/80,100,60	0.417	0.499	
vQZ/SD32/60 +Δ		**0.536**	
Mårtensson-Pendrill[11] RPA		0.52	
Dzuba *et al.*[12] RPA		0.57	
Singh *et al.*[13] CCSD$_p$T		0.501	

[11] A.M. Mårtensson-Pendrill, *Phys. Rev. Lett.* **54** (1985) *1153*

[12] V.A. Dzuba, V.V. Flambaum, S.G. Porsev, *Phys. Rev. A* **80** (2009) *032120*

[13] Y. Singh, B.K. Sahoo, B.P. Das, *Phys. Rev. A* **89** (2014) *030502(R)*

Scalar-Pseudoscalar $\mathcal{P},\mathcal{T}$-odd Nucleon-Electron Interaction

Results for ^{129}Xe

$$\alpha_{C_S}(\psi_J) = \frac{-A\frac{G_F}{\sqrt{2}}}{E_{\text{ext}}\left\langle\psi_J^{(1)}\middle|\psi_J^{(1)}\right\rangle}\left[\sum_{K\neq J}\frac{\left\langle\psi_K^{(0)}\middle|\hat{H}_{\text{HF}}\middle|\psi_J^{(0)}\right\rangle\left\langle\psi_J^{(0)}\middle|\imath\sum_e\gamma_e^0\gamma_e^5\rho(\mathbf{r}_e)\middle|\psi_K^{(0)}\right\rangle}{\varepsilon_J^{(0)}-\varepsilon_K^{(0)}} + h.c.\right]$$

Basis	# of CI states/X	S [10^{-2} *a.u.*]	α_{C_S} [$10^{-22}\,e$ cm]
cvTZ/40 a.u.	8/S8	0.059	0.063
cvTZ/7 a.u.	8/6s6p	0.055	0.059
cvTZ/14 a.u.	8/6s6p	0.055	0.059
cvTZ/7 a.u.	8/6s6p5d	0.060	0.065
cvTZ/7 a.u.	8/6s6p5d7p	0.067	0.072
cvTZ/7 a.u.	8/6s6p5d7p7s	0.069	0.074
cvTZ/7 a.u.	8/6s6p5d7p7s6d	0.070	0.075
cvTZ/7 a.u.	8/6s6p5d7p7s6d4f	0.070	0.075
cvTZ/7 a.u.	8/6s6p5d7p7s6d4f7d5f8p8s	0.067	0.072
cvTZ/7 a.u.	20/6s6p5d7p7s	0.055	0.059
cvTZ/S10-SDT8-SD18/7au	20/6s6p5d7p	0.05	0.05
cvQZ/100 a.u.	8/S8	0.059	0.064
cvQZ/50 a.u.	1000/S8	0.050	0.053
cvQZ/50 a.u.	8/6s6p	0.048	0.052
cvQZ/50 a.u.	8/6s6p5d7p7s	0.071	0.076
vQZ/1281/S8/100 + ΔS_{corr}		**0.073**	**0.078**

Scalar-Pseudoscalar $\mathcal{P},\mathcal{T}$-odd Nucleon-Electron Interaction

Results for ^{129}Xe

- Two major contributions in the sum over states:

$$C_1 = \frac{\langle 5p{\to}6s\ 0,0|\hat{H}_{\mathsf{HF}}|0,0\rangle\langle 0,0|\imath \sum_e \gamma_e^0\,\gamma_e^5\,\rho(\mathbf{r}_e)|5p{\to}6s\ 0,0\rangle}{\varepsilon^{(0)}_{0,0}-\varepsilon^{(0)}_{5p\to 6s\ 0,0}}$$

$$C_2 = \frac{\langle 5p{\to}6p\ 1,0|\hat{H}_{\mathsf{HF}}|0,0\rangle\langle 0,0|\imath \sum_e \gamma_e^0\,\gamma_e^5\,\rho(\mathbf{r}_e)|5p{\to}6p\ 1,0\rangle}{\varepsilon^{(0)}_{0,0}-\varepsilon^{(0)}_{5p\to 6p\ 1,0}}$$

$|\text{hole spinor} \to \text{particle spinor}\ J, M_J\rangle$

- . . . and a large number of small contributions uncorrected for correlation effects.

Final values for α_{C_T} and α_{C_S} *will lead* to tighter constraints.[14]

[14] T. F., M. Jung, (2019) *in preparation.*

Outline

- Relativistic many-body methods for atomic and molecular structure
- Atomic EDMs : ^{129}Xe
- **Molecular EDMs : AgRa**

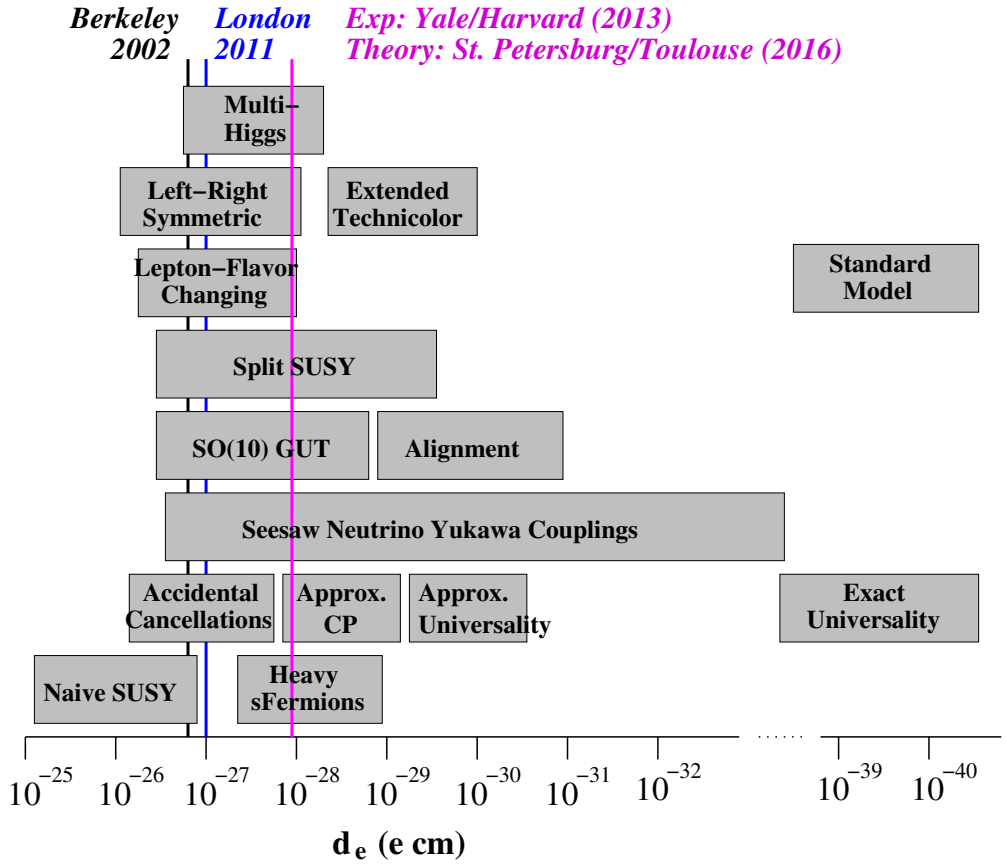
in collaboration with

D. DeMille

Yale University

New Haven, CT 06520, USA

eEDM Constraint on Beyond-Standard-Model Theories[15]
Single-source interpretation



Model	$\lvert d_e \rvert [e \cdot cm]$
Standard model	$< 10^{-38}$
Left-right symmetric	$10^{-28} \ldots 10^{-26}$
Lepton-flavor changing	$10^{-29} \ldots 10^{-26}$
Multi-Higgs	$10^{-28} \ldots 10^{-27}$
Supersymmetric	$\leq 10^{-25}$
Experimental limit (Tl)[16]	$< 1.6 \cdot 10^{-27}$
Experimental limit (YbF)[17]	$< 10.5 \cdot 10^{-28}$
Experimental limit (ThO)[18]	$< 9.6 \cdot 10^{-29}$

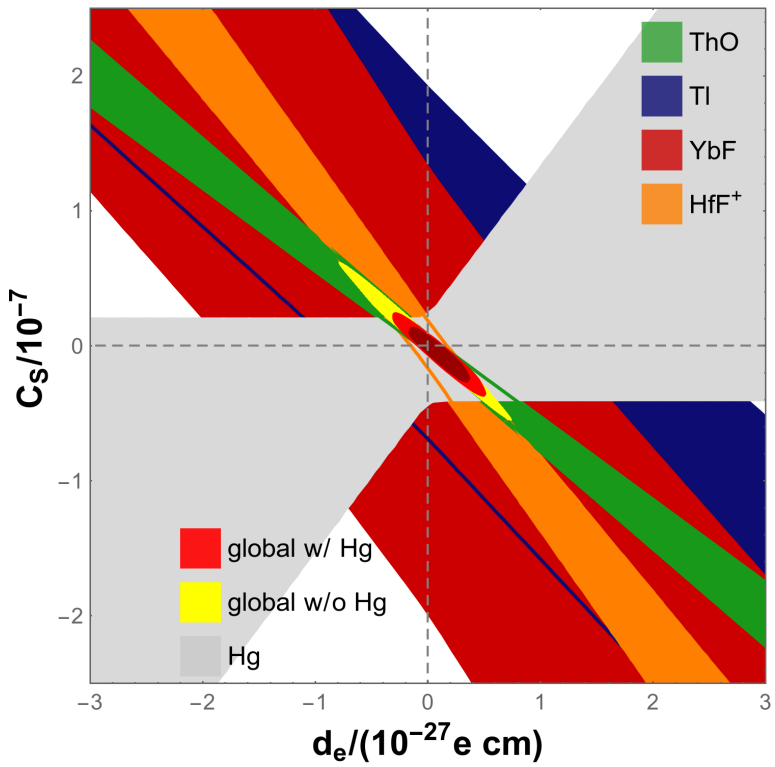
[15] Courtesy: DeMille (2005), Huliyar (2009)

[16] B.C. Regan, E.D. Commins, C.J. Schmidt, D.P. DeMille, *Phys Rev Lett* **88** (2002) *071805/1*

[17] J.J. Hudson, D.M. Kara, I.J. Smallman, B.E. Sauer, M.R. Tarbutt, E.A. Hinds, *Nature* **473** (2011) *493*

[18] D. DeMille, ICAP 2014, Washington D.C., ACME Collaboration, *Science* **6168** (2014) *269*, TF and M. K. Nayak, *J. Mol. Spectrosc.* **300** (2014) *16*, L. V. Skripnikov, A. N. Petrov, A. V. Titov, *J. Chem. Phys.* **139** (2013) *221103*, L. V. Skripnikov, A. V. Titov, *J. Chem. Phys.* **142** (2015) *024301*, M. Denis, TF, *J Chem Phys* **145** (2016) *214307*

New Bounds on (Semi-)Leptonic $\mathcal{CP}$-odd Parameters[19]



Multiple-source picture:

$$\Delta E_{\mathcal{P},\mathcal{T}} = -\langle \mathbf{d}_{\mathrm{sys}} \cdot \mathbf{E}_{\mathrm{ext}} \rangle$$
$$= \left(\alpha_{d_e}\, d_e + \alpha_{C_S}\, C_S\right) \langle \mathbf{n} \cdot \mathbf{z} \rangle \left(E_{\mathrm{ext}}\right)$$

Previous resulting bound:

From HfF^+, ThO, YbF, Tl

$$|d_e|_{2017} < 6.4 \times 10^{-28} e\ \mathrm{cm}$$

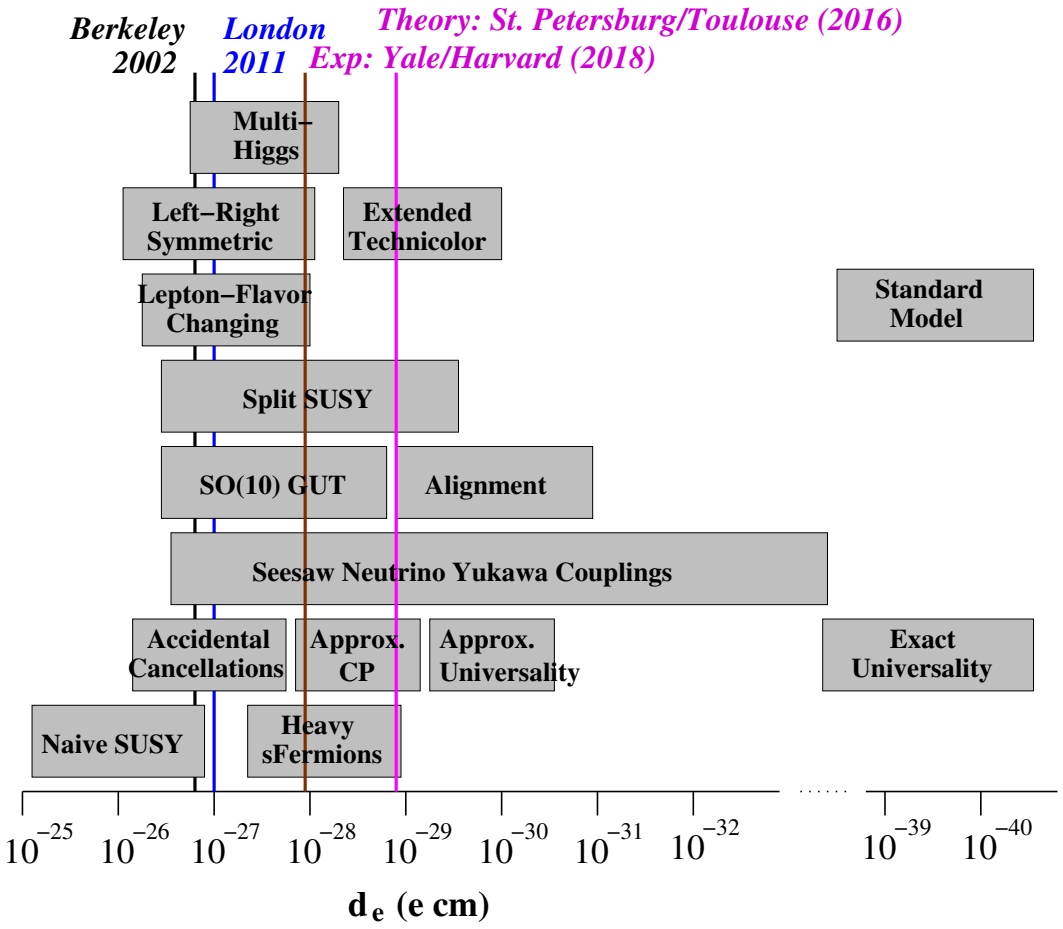
New resulting bounds:

From Hg, HfF^+, ThO, YbF, Tl

$$|d_e|_{2018} < 3.8 \times 10^{-28} e\ \mathrm{cm}$$
$$|C_S|_{2018} < 2.7 \times 10^{-8}$$

[19] TF, M. Jung, *J. High Energy Phys.* **7** (2018) *012*
J. Baron *et al.*, *Science* **343** (2014) *269*
M. Denis, T. F., *J. Chem. Phys.* **145** (2016) *214307*
L. Skripnikov, *J. Chem. Phys.* **145** (2016) *214301*
W.B. Cairncross, D.N. Gresh, M. Grau, K.C. Cossel, T.S. Roussy, Y. Ni, Y. Zhou, J. Ye, E.A. Cornell, *Phys. Rev. Lett.* **119** (2017) *153001*
T. F., *Phys. Rev. A (Rap. Comm.)*, **96** (2017) *040502(R)*
L.V. Skripnikov, *J. Chem. Phys.*, **147** (2017) *021101*

eEDM Constraint on Beyond-Standard-Model Theories
Single-source interpretation (2018)



ACME 2018 result[20] combined with 2016 theory

[20] ACME collaboration, *Nature*, **562** (2018) *355*

Going Ultracold: From beams to traps

PHYSICAL REVIEW A, VOLUME 63, 023405

Loading and compressing Cs atoms in a very far-off-resonant light trap

D. J. Han, Marshall T. DePue, and David S. Weiss
Department of Physics, University of California at Berkeley, Berkeley, California 94720-7300
(Received 25 May 2000; published 12 January 2001)

We describe an experiment in which 3×10^7 Cs atoms are loaded into a 400 μm crossed beam far-off-resonant trap (FORT) that is only 2 μK deep. A high-density sample is prepared in a magneto-optic trap, cooled in a three-dimensional far-off-resonant lattice (FORL), optically pumped into the lowest-energy state, adiabatically released from the FORL, magnetically levitated, and transferred to the final trap with a phase-space density of 10^{-3}. Spontaneous emission in the FORT is negligible, and we have compressed the atoms in the FORT to a spatial density of 2×10^{13} atoms/cm^3. Evaporative cooling under these conditions proceeds rapidly.

- Estimated sensitivity of Cs EDM measurement in DLT[21] is $|d_e| \approx 10^{-29}\, e\text{cm}$

  Cs atom: $\Delta E = R\, E_{\text{ext}}\, d_e$ $\quad E_{\text{int}} \approx 20\ \left[\frac{\text{MV}}{\text{cm}}\right]$

  Ultracold **XY** Molecule: $\Delta E = E_{\text{eff}}\, d_e$ $\quad E_{\text{eff}} \approx 50\ \left[\frac{\text{GV}}{\text{cm}}\right]$

- A factor of $\approx \mathbf{2500}$ gain in sensitivity!

[21] DLT: Dipole light trap; D. Weiss (Penn State), 2014: "Measuring the eEDM using laser-cooled Cs atoms in optical lattices"
S. Chu, J.E. Bjorkholm, A. Ashkin, A. Cable, *Phys. Rev. Lett.* **57** (1986) *314*
C. Chin, V. Leiber, V. Vuletić, A.J. Kerman, S. Chu, *Phys. Rev. A* **63** (2001) *033401*

Towards Ultracold DLT EDM Measurement

Picking the cherry

In the casting:
Alkali(-like) atoms: Li, Na, K, Rb, Cs; Ag, Au
Earth-alkaline atoms: Sr, Ba, Ra; Yb

Jury spreadsheet for X partner of Ra and some contenders:

X	EA(X) [eV]	$E_{\mathrm{eff}_{\max}}\left[\frac{\mathrm{GV}}{\mathrm{cm}}\right]$	$B_v = \left\langle v\left\vert\frac{1}{\mu R^2}\right\vert v\right\rangle\,[\mathrm{cm}^{-1}]$	D [D]	$E_{\mathrm{pol}} = \frac{B_v}{D}\left[\frac{\mathrm{kV}}{\mathrm{cm}}\right]$
Li	0.62	61	−	≈ 1.5	
Na	0.55	58	−	≈ 1	
K	0.50	50	−	≈ 1	
Rb	0.49	48	+	≈ 1	
Cs	0.47	44	+	≈ 1	
Ag	1.30	66	0.021	5.4	0.264
Au	2.31	60	+	≈ 6	
AgBa	1.30	6	+	≈ 3	
RbYb[22]		−0.7	0.001	0.21	5.5
CsYb[22]		0.54	0.007	0.24	3.5

[22] E. R. Meyer, J. L. Bohn, *Phys. Rev. A* **80** (2009) *042508*

Towards Ultracold DLT EDM Measurement

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RbYb		−0.7	0.001	0.21	5.5
CsYb		0.54	0.007	0.24	3.5

Au is forbidden fruit!

$(\mathcal{P},\mathcal{T})$-odd properties of AgRa

- Electron EDM effective electric field[23]

$$E_{\rm eff} = \frac{2\imath c}{e\hbar} \left\langle \sum_{j=1}^{n} \gamma_j^0 \gamma_j^5 \, \vec{p}_j{}^{\,2} \right\rangle_{\psi^{(0)}}$$

- S-PS nucleon-electron interaction constant[24]

$$W_{\mathcal{S}} := \frac{\imath}{\Omega} \frac{G_F}{\sqrt{2}} Z_{\rm heavy} \, \langle \Psi_\Omega | \sum_{j=1}^{n} \gamma_j^0 \gamma_j^5 \, \rho_N(\vec{r}_j) \, | \Psi_\Omega \rangle$$

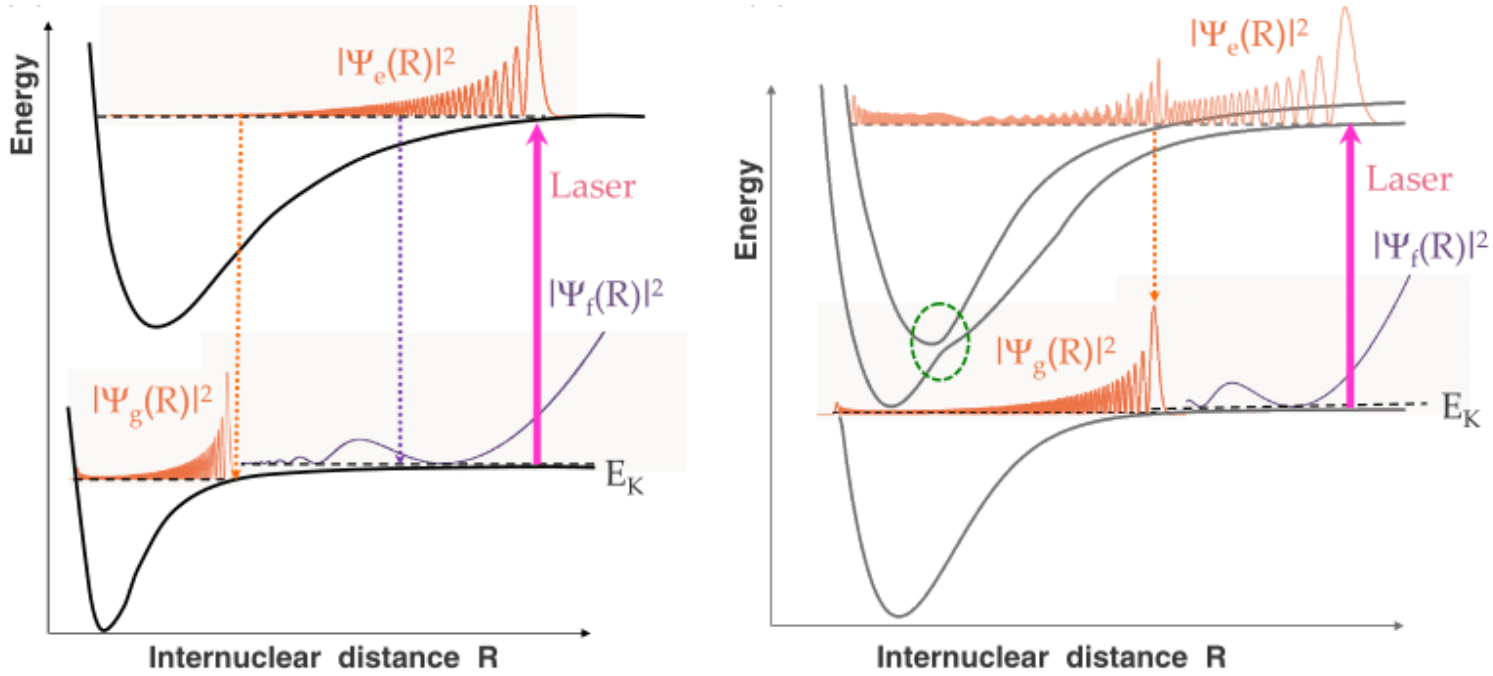
		$^3\Delta_1$		$^2\Sigma_{1/2}$		
	ThO	HfF^+	ThF^+	YbF	AgRa	
$\lvert E_{\rm eff}\rvert$	78	23	35	25	64	$\left[\frac{\rm GV}{\rm cm}\right]$
$\lvert W_S\rvert$	106	20	51	40	175	[kHz]

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

[24] V. G. Gorshkov, L. N. Labzovski, and A. N. Moskalev, *Zh. Eksp. Teor. Fiz.* **76** (1979) *414*
M. Denis *et al.*, *New J. Phys.* **7** (2015) *043005*

Devising a AgRa DLT EDM Experiment

- Photoassociating ultracold atoms into ultracold molecules[25]



- Does its electronic spectrum allow for efficient energy transfer (remove binding energy without heating) ?
- Which states are candidates for photoassociation ?

[25] L. D. Carr, D. DeMille, R. V. Krems, J. Ye, *New J. Phys.* **11** (2009) *055049*

AgRa - Electronic-structure model

		# of Kramers pairs	accumulated # of electrons min.	max.
III	*(4 a.u.)* *Virtual Kramers pairs*	71	21	21
II	*Ag: 5p,6s* *Ra: 7p,8p,6d* *Ag: 5s* *Ra: 7s* σ	18	19	21
I	*Ra: 6s, 6p* *Ag: 4d*	9	17	18
	Frozen core	(57)		

Determinant classes

$I^{18}\ II^{3}\ III^{0}$ (Reference space)

$I^{18}\ II^{2}\ III^{1}$ (Singles)

$I^{18}\ II^{1}\ III^{2}$ (Valence correlating)

$I^{17}\ II^{4}\ III^{0}$ (Singles)

$I^{17}\ II^{3}\ III^{1}$ (Singles)

$I^{17}\ II^{2}\ III^{2}$ (Core-val. correlating)

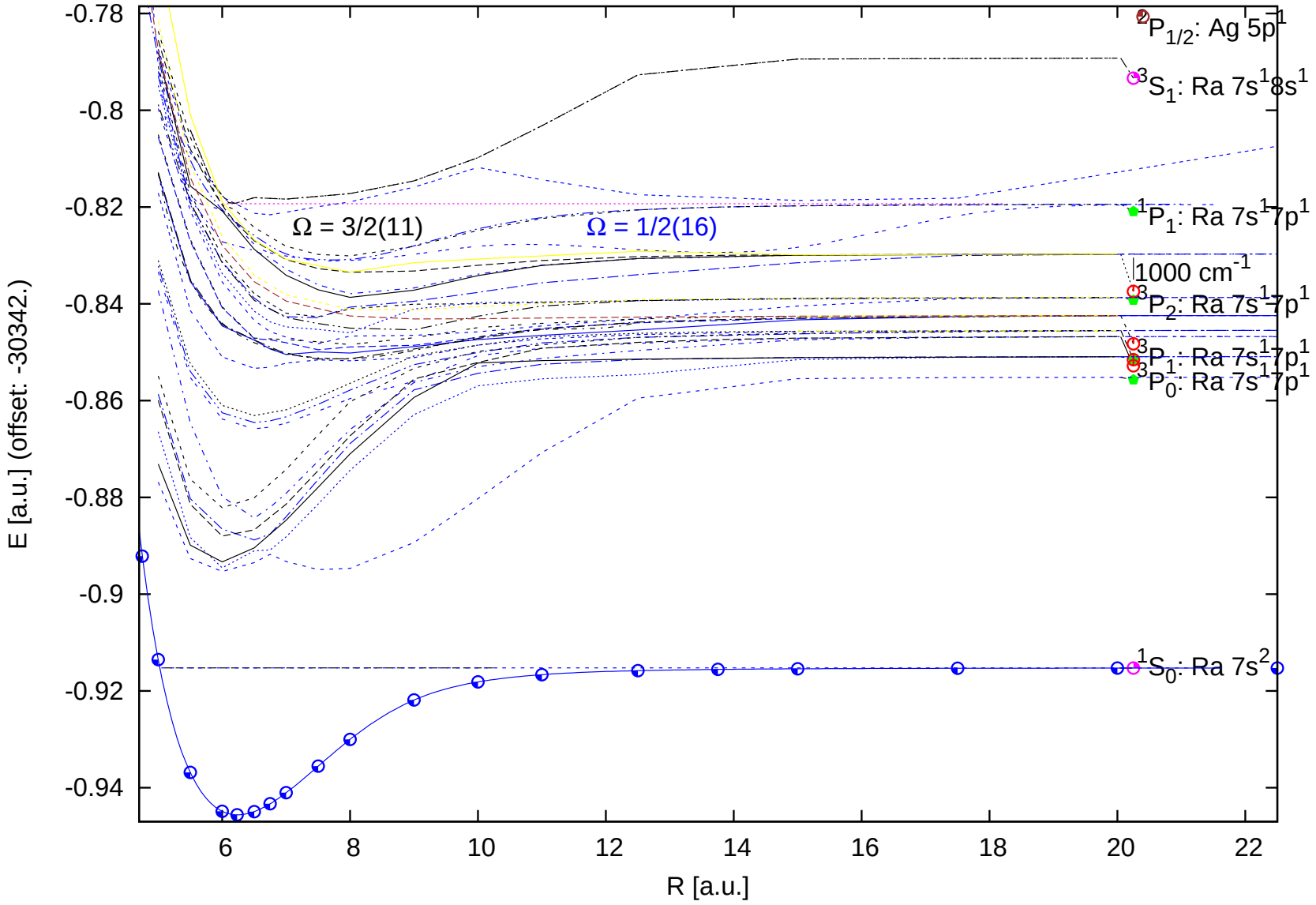
17.6×10^{6} expansion terms ($\Omega = 1/2$)

cvTZ bases[26] **KRCI** program[27]

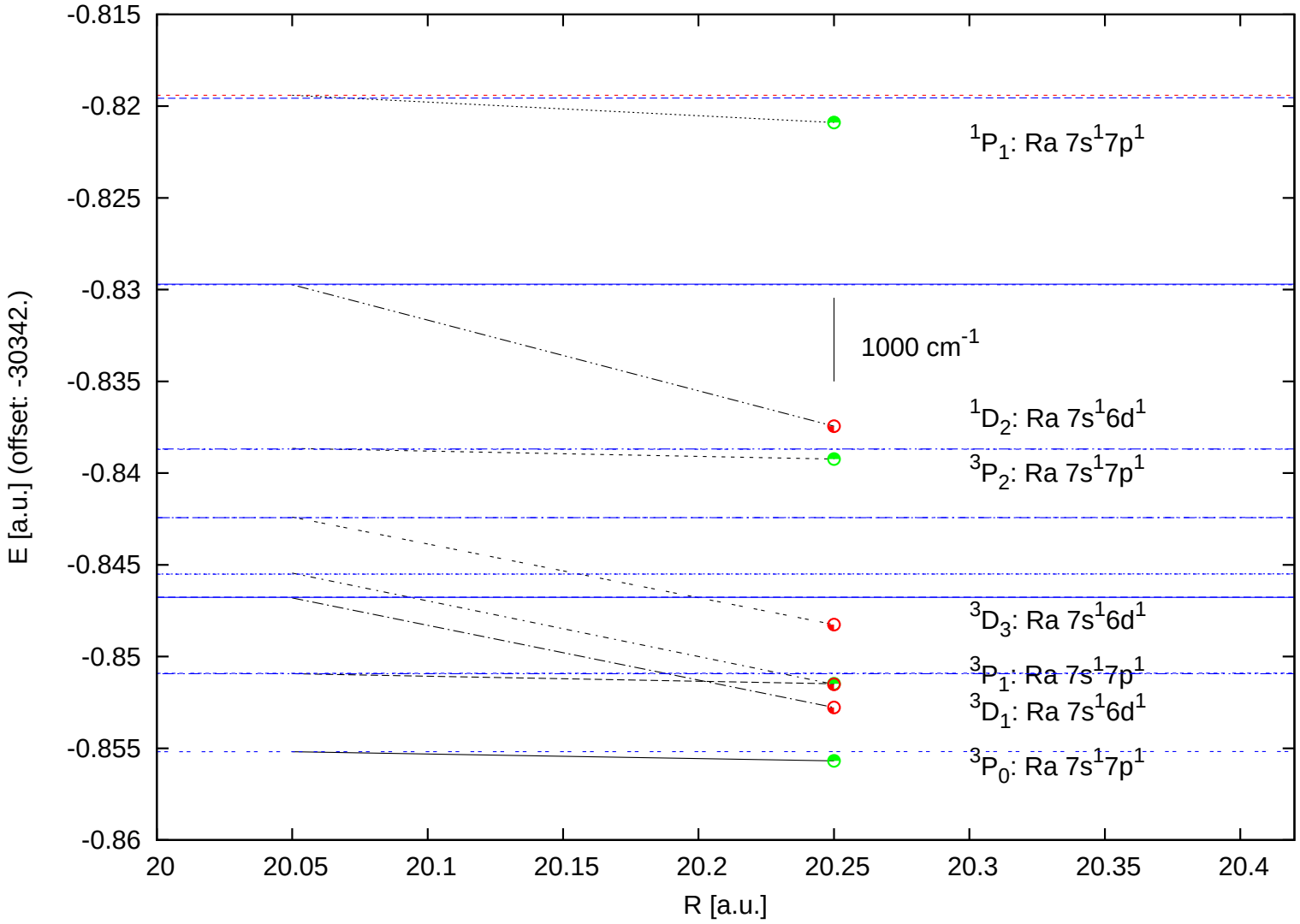
[26] K.G. Dyall, *Theoret. Chim. Acta* **131** (2012) *1217*
K.G. Dyall, A.S.P. Gomes, *Theoret. Chim. Acta* **125** (2010) *97*

[27] S. Knecht, H.J.Aa. Jensen, T.F., *J. Chem. Phys.* **132** (2010) *014108*
DIRAC15 package (locally modified)

AgRa - PECs

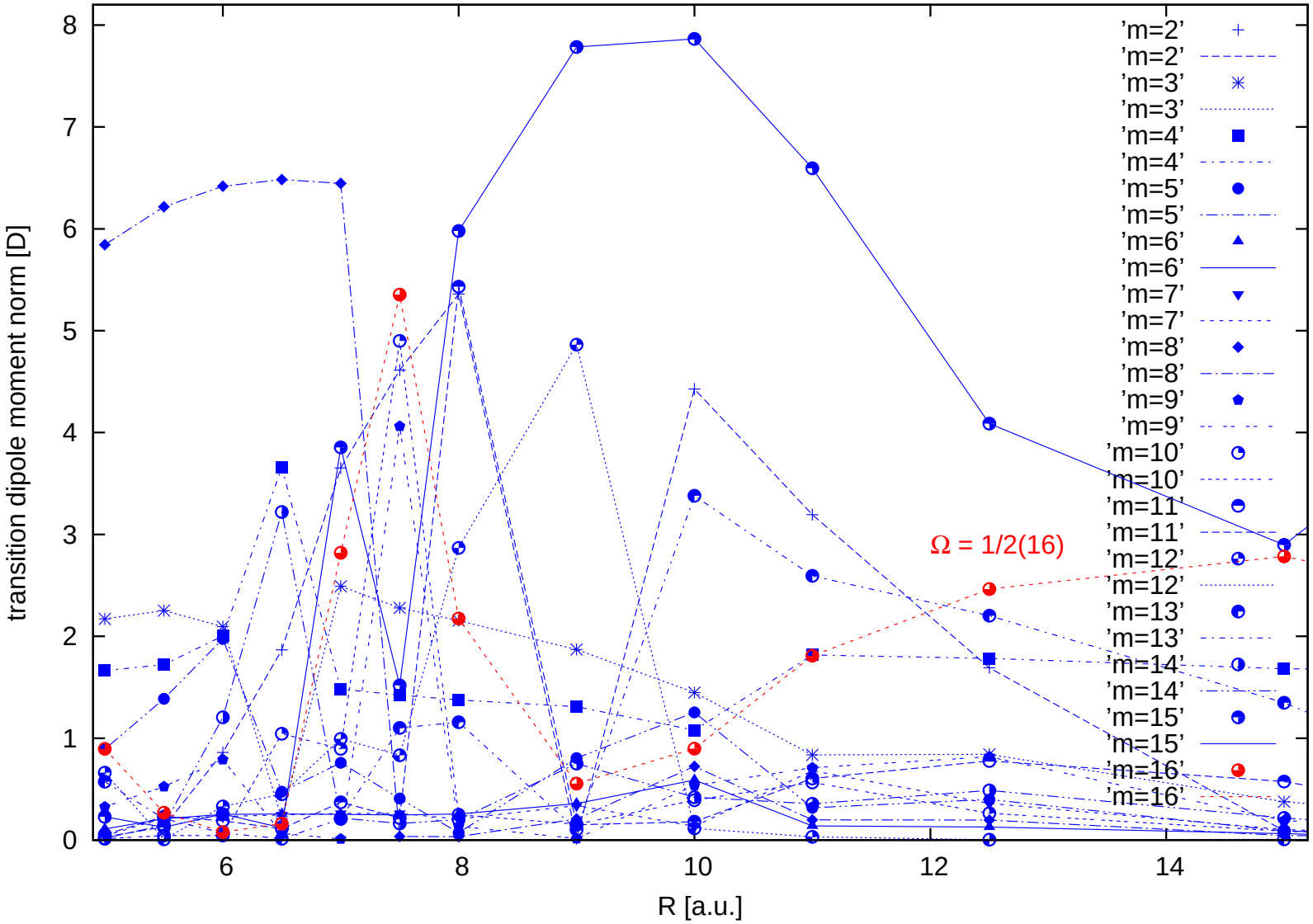

-0.78
-0.8
-0.82
-0.84
-0.86
-0.88
-0.9
-0.92
-0.94
E [a.u.] (offset: -30342.)
6
8
10
12
14
16
18
20
22
R [a.u.]
Ω = 3/2(11)
Ω = 1/2(16)
1000 cm-1
2P1/2: Ag 5p1
3S1: Ra 7s18s1
1P1: Ra 7s17p1
3P2: Ra 7s17p1
3P1: Ra 7s17p1
3P0: Ra 7s17p1
1S0: Ra 7s2

Radium excited states - Comparison with experiment[28]

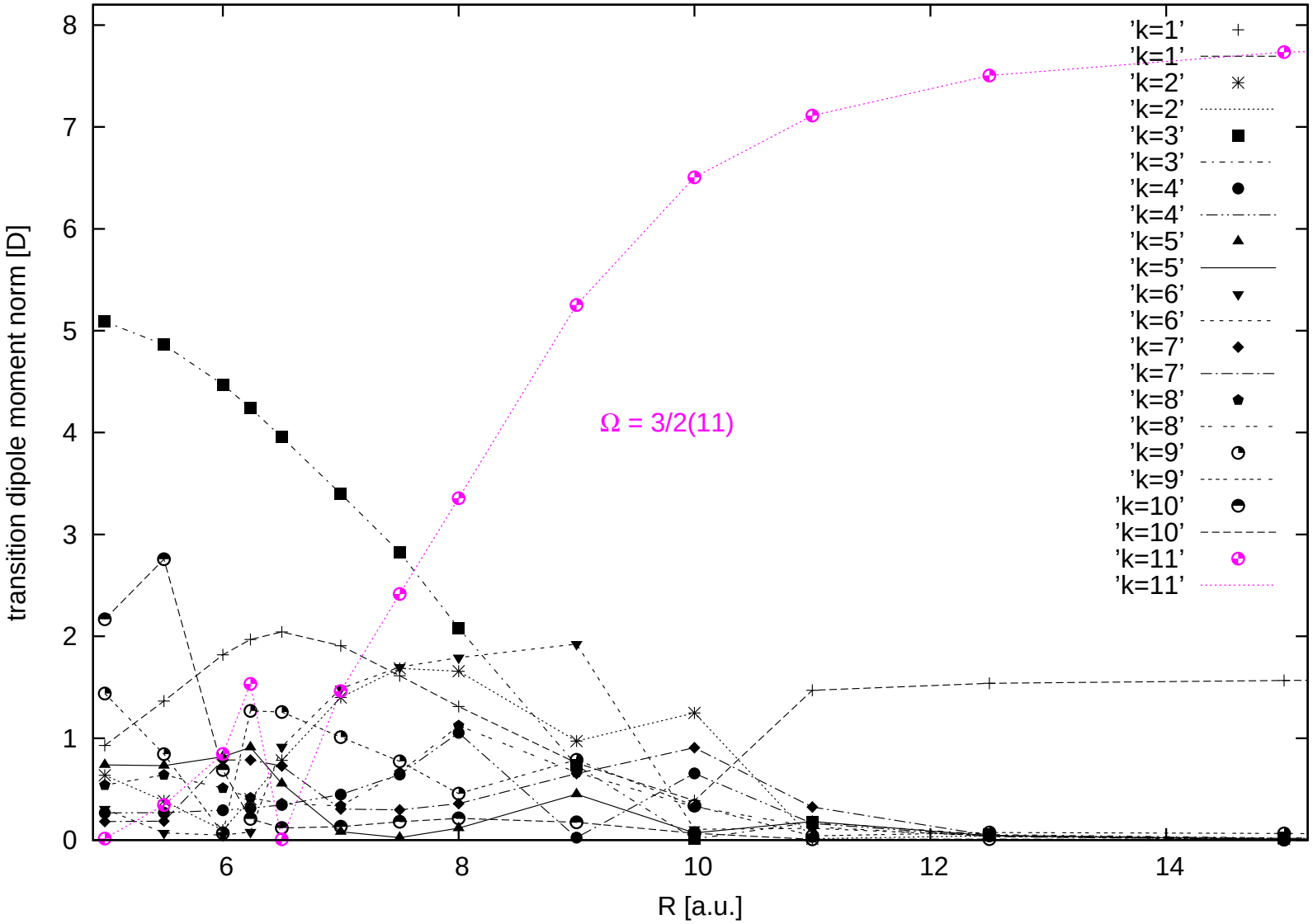


[28] NIST Atomic Spectra Database (ver. 5.5.1)

Electronic Transition Dipole Moments : $\Omega = 1/2$ states



Electronic Transition Dipole Moments : $\Omega = 3/2$ states



Long-range interactions : theory

- Motivation: Find least-bound vibrational level of ground potential
- Problem: PECs at long range not sufficiently accurate, even with very large basis sets !
- Solution: Go "back" to atomic physics

Dispersion interaction for two neutral heteronuclear atoms

Van der Waals interaction potential:

$V(R) = -\frac{C_6}{R^6} - \frac{C_8}{R^8} - \frac{C_{10}}{R^{10}} - \dots$

Dispersion coefficients from oscillator strengths[29]

Ground state:

$$C_6^{\Omega=1/2(1)} = \sum_{n_c(\ell_c=1),n_d(\ell_d=1)} \frac{3}{2} \frac{f_{ac}^{(1)} f_{bd}^{(1)}}{\Delta E_{ca} \Delta E_{db} \left(\Delta E_{ca} + \Delta E_{db}\right)}$$

$a = {}^2S_{1/2}(5s^1)$ for Ag and $b = {}^1S_0(7s^2)$ for Ra

$$f_{KL}^{(1)} = 2 \left(E_L - E_K\right) \left|\left| \langle \psi_K | \sum_{k=1}^{n} \hat{r}(k) \, |\psi_L\rangle \right|\right|^2$$

$|\psi_L\rangle = \left|{}^2P\right\rangle = \left|1, M_L; \frac{1}{2}, \frac{1}{2}\right\rangle$ are expanded as

$$\left|1, 0; \tfrac{1}{2}, \tfrac{1}{2}\right\rangle = \left\langle \tfrac{3}{2}, \tfrac{1}{2} | 1, 0; \tfrac{1}{2}, \tfrac{1}{2}\right\rangle \left|\tfrac{3}{2}, \tfrac{1}{2}\right\rangle + \left\langle \tfrac{1}{2}, \tfrac{1}{2} | 1, 0; \tfrac{1}{2}, \tfrac{1}{2}\right\rangle \left|\tfrac{1}{2}, \tfrac{1}{2}\right\rangle$$

where $\langle J, M_J | L, M_L; S, M_S \rangle$

[29] J.-Y. Zhang, J. Mitroy, *Phys. Rev. A* **76** (2007) *022705*

Dispersion coefficients from oscillator strengths

Atomic Transition Dipole Moments: Ra

Transition	D_{KL}^{0-1} [*a.u.*]	f_{kl}	f_{kl} [30]	$\Delta\epsilon$ [cm^{-1}]
$^1S_0(7s^2) - {}^3P_1(7s7p)$	0.4995	0.0350		15391.
$^1S_0(7s^2) - {}^1P_1(7s7p)$	3.3687	2.1422	1.91	20715.614
$^1S_0(7s^2) - {}^3P_1(7s8p)$	0.3836	0.0402		30000.
$^1S_0(7s^2) - {}^1P_1(7s8p)$	0.8252	0.2039		32857.537

Transition	D_{KL}^{1-J} [*a.u.*]	f_{kl}	$\Delta\epsilon$ [cm^{-1}]
$^1P_1(7s7p) - {}^3S_1(7s8s)$	0.4729	0.0545	26754.02
$^1P_1(7s7p) - {}^1S_0(7s8s)$	3.5065	3.2870	29336.371
$^1P_1(7s7p) - {}^3D_2(7s7d)$	0.2547	0.0189	31993.40
$^1P_1(7s7p) - {}^3D_1(7s7d)$	0.7491	0.1636	32000.78
$^1P_1(7s7p) - {}^1D_2(7s7d)$	1.7717	1.0011	35000.

[30] V.A. Dzuba and V.V. Flambaum, *J. Phys. B* **40** (2007) *227*

Testing the approach: LiBe and RbSr

LiBe ($^2\Sigma_{1/2}$)	lin. CCSD[31]	KRCI(FCI)
C_6[a.u.]	$478(3)$	454

RbSr ($^2\Sigma_{1/2}$)	lin. CCSD[31]	KRCI MR-SD	KRCI MR-SD +TQ
C_6[a.u.]	$3697(10)$	3995	3849

5% residual deviation!

AgRa ($\Omega = 1/2(1)$)	KRCI(MR-SD)
C_6[a.u.]	1163

[31] A. Derevianko, S.G. Porsev, J.F. Babb, *At. Data Nucl. Data Tables* **96** (2010) *323*
M.S. Safronova, W.R. Johnson, A. Derevianko, *Phys. Rev. A* **60** (1999) *4476*

Connecting LR- and SR-Potentials

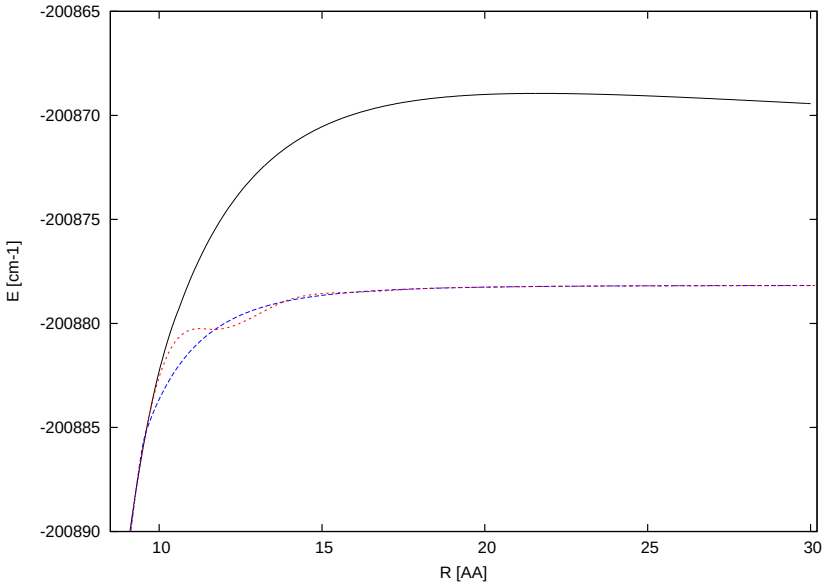
Ground potential $\Omega = 1/2(1)$

LeRoy radius[32]:

$$R_{\rm LR} = 2\left(\langle\hat{r}^2\rangle_A^{1/2} + \langle\hat{r}^2\rangle_B^{1/2}\right)$$

$$\langle\hat{r}^2\rangle_{{\rm Ag}_{5s}} = 13.90 \text{ a.u.} \qquad \langle\hat{r}^2\rangle_{{\rm Ra}_{7s}} = 28.71 \text{ a.u.}$$

$$\Rightarrow R_{\rm LR}^{\Omega=1/2(1)} = 18.2\,\text{a.u.}$$



- Pure short-range potentials produce artefacts! |
- $V(R) \approx -\frac{C_6}{R^6}$ and fit long-range to short-range curves
- Correct physics from careful fitting | |

[32] R.J. LeRoy, *Can. J. Phys.* **52** (1974) *246*
R.J. LeRoy, R. B. Bernstein, *J. Chem. Phys.* **52** (1970) *3869*

Dispersion coefficients from oscillator strengths

Ra(7p) excited states:

$$C_6^{\Omega=1/2(16)} = C_6^{\Omega=3/2(11)} = \sum_{n_c(\ell_c=1),n_d(\ell_d=0)} \frac{3}{4} \frac{f_{ac}^{(1)} f_{bd}^{(1)}}{\Delta E_{ca} \Delta E_{db} \left(\Delta E_{ca} + \Delta E_{db}\right)}$$

$$+ \sum_{n_c(\ell_c=1),n_d(\ell_d=1)} \frac{15}{8} \frac{f_{ac}^{(1)} f_{bd}^{(1)}}{\Delta E_{ca} \Delta E_{db} \left(\Delta E_{ca} + \Delta E_{db}\right)}$$

$$+ \sum_{n_c(\ell_c=1),n_d(\ell_d=2)} \frac{57}{40} \frac{f_{ac}^{(1)} f_{bd}^{(1)}}{\Delta E_{ca} \Delta E_{db} \left(\Delta E_{ca} + \Delta E_{db}\right)}$$

AgRa $(\Omega = 1/2(16), \Omega = 3/2(11))$	KRCI(MR-SD)
C_6[a.u.]	773

Connecting LR- and SR-Potentials

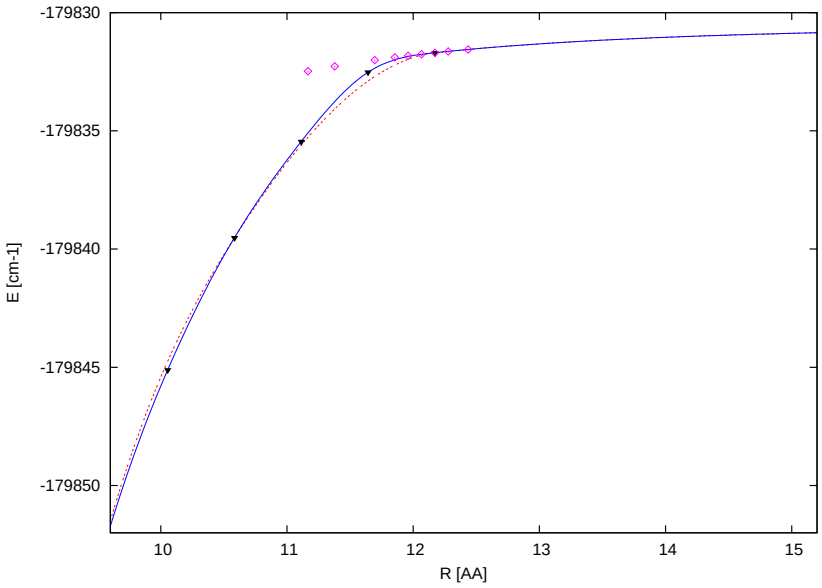
Excited potentials $\Omega = 1/2(16), \Omega = 3/2(11)$

LeRoy radius:

$$R_{\mathrm{LR}} = 2\left(\langle \hat{r}^2 \rangle_A^{1/2} + \langle \hat{r}^2 \rangle_B^{1/2}\right)$$

$$\langle \hat{r}^2 \rangle_{\mathrm{Ag}_{5s}} = 13.90 \text{ a.u.} \qquad \langle \hat{r}^2 \rangle_{\mathrm{Ra}^+_{7p_{3/2}}} = 60.63 \text{ a.u.}$$

$$\Rightarrow R_{\mathrm{LR}}^{\Omega=1/2(1)} = 23.0\,\text{a.u.}$$



- $V(R) \approx -\frac{C_6}{R^6}$ and fit long-range to short-range curves

AgRa FCF for Proposed PA

Largest FCF of excited state with $\Omega = 1/2(1), v = 128$, electronic TDM with excited state, and dispersion coefficient of excited state

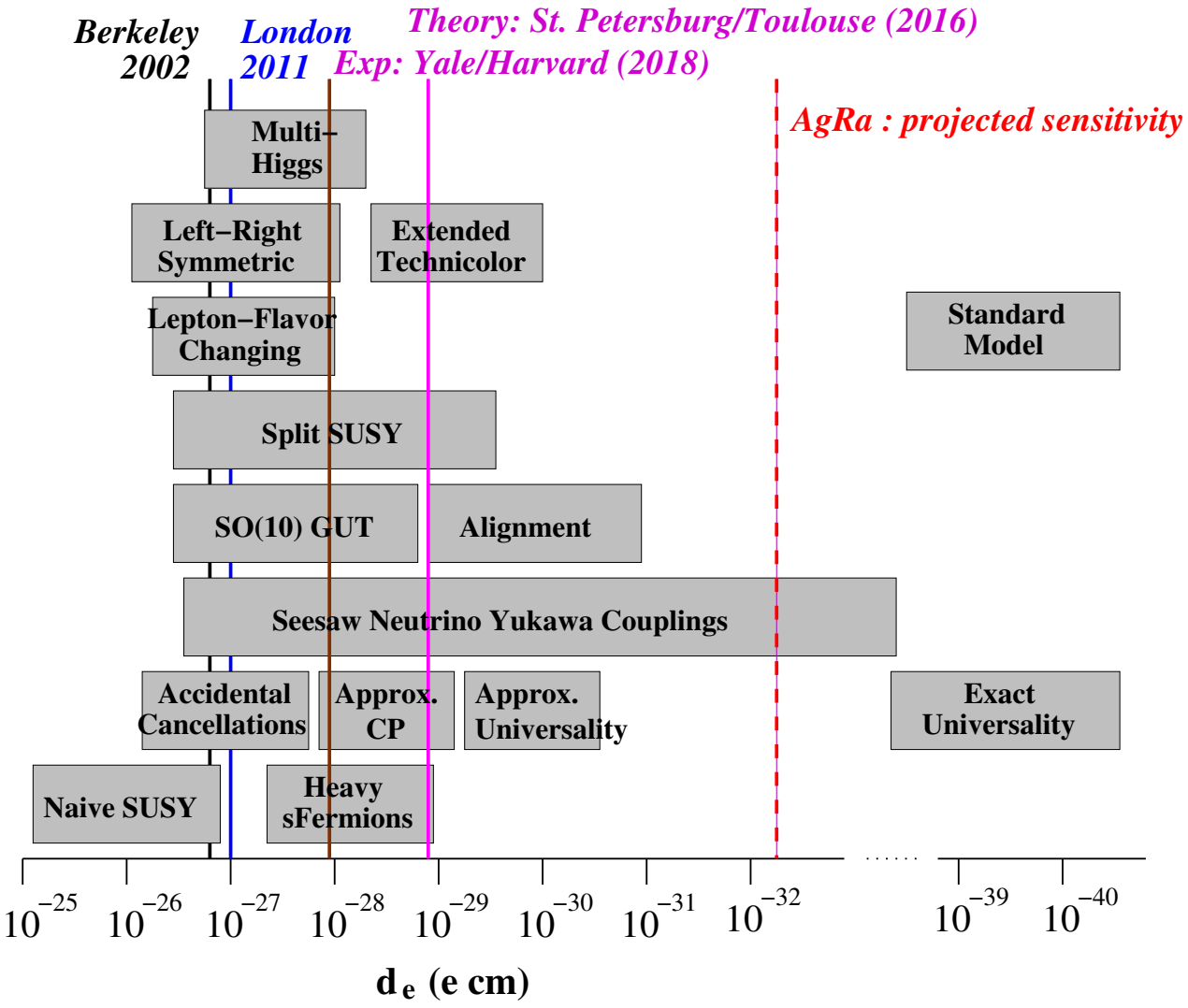
	$\Omega = 1/2(15), v = 191$	$\Omega = 1/2(16), v = 102$	$\Omega = 3/2(11), v = 98$
max. FCF	2.63×10^{-1}	1.26×10^{-2}	1.66×10^{-2}
TDM [D]	4.09	2.46	7.51
C_6 [a.u.]	2336	773	773

Conclusion.[33] Where do we go from here ?

- AgRa $\mathcal{P},\mathcal{T}$-odd properties are competitive
- Electronic spectrum is complex, but PA seems viable
- FCFs for $\Omega = 1/2(1) - \Omega = 1/2(16)$ and $\Omega = 1/2(1) - \Omega = 3/2(11)$ look promising
- One remaining question: Free/trap-bound — bound transitions ?
- AgRa seems prime candidate among ultracold "contenders"

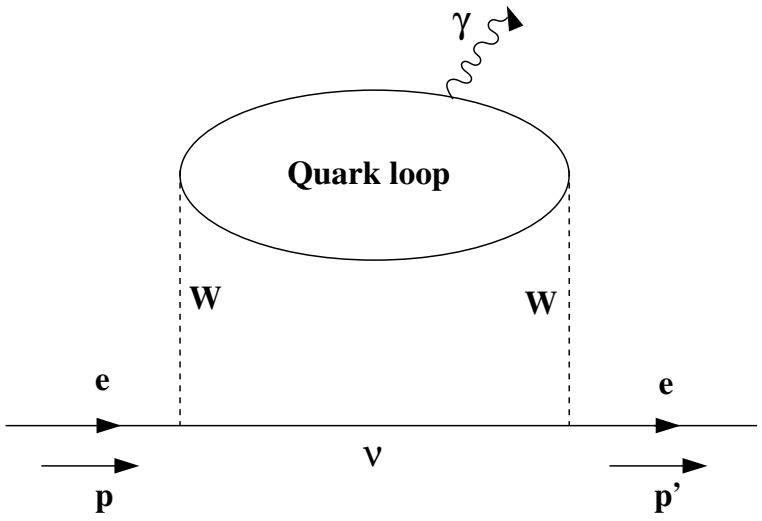
[33] TF, D. DeMille, manuscript in preparation

eEDM Constraint on Beyond-Standard-Model Theories
Single-source interpretation (20??)



The induced fermion EDM

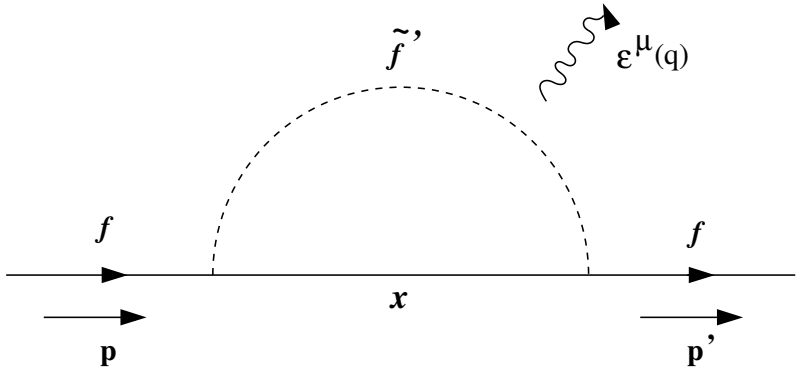
Standard Model Picture[34]



- Three-loop $\mathcal{CP}$-odd contributions zero in the absence of gluonic corrections[35]

  $d_e^{SM} \leq 10^{-38}\, e$ cm

BSM Picture



χ: chargino, neutralino

$\tilde{f}'_j$: supersymmetry (s)-fermion

$\epsilon^\mu(q)$: photon

- MSSM ("naïve SUSY") prediction[36]:
  $d_e \leq 10^{-27}\, e$ cm

[34] E.D. Commins, *Adv At Mol Opt Phys* **40** (1998) *1*

[35] M. Pospelov, I.B. Khriplovich, *Sov J Nuc Phys* **53** (1991) *638*

[36] J. Ellis, J.S. Lee, A. Pilaftsis, *J High Energy Phys* **10** (2008) *049*