Fundamental physics with molecules: From electric dipole moments to dark matter candidates

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TRIUMF, Vancouver, Canada and CENPA, Washington, USA, 4 November 2020

Particle physics, nuclear physics and atomic physics with molecules

"Never measure anything but frequency"
(says Arthur L. Schawlow)

Hänsch, *Rev. Mod. Phys.* **2006**, *78*, 1297

Particle physics, nuclear physics and atomic physics with molecules

"A diatomic molecule is a molecule with one atom too many."
(says Arthur L. Schawlow)

New Scientist, **1981**, *92*, 224-225

Particle physics, nuclear physics and atomic physics with molecules

An atom is an atom too many.
(says the string-theoretician)

Particle physics, nuclear physics and atomic physics with molecules

An atom is a diatomic molecule lacking the essential atom!

Particle physics, nuclear physics and atomic physics with molecules

An atom is at least an atom too few!

Particle physics, nuclear physics and atomic physics with molecules

Let the molecule do the job!

Fundamental symmetries on various levels

Biology

Biochemistry

Chemistry

Molecular physics

Atomic physics

Nuclear physics

Particle physics

- Continuous space-time symmetries (e.g. translation, rotation)
- Discrete symmetries (e.g. **time reversal, space inversion**)
- Permutation symmetry
- Unitary symmetries

Berger in: Relativistic electronic structure theory, Part 2, (Ed: P. Schwerdtfeger), **2004**, 188

Opportunities and Challenges

- Close-lying levels of opposite parity
- Strong enhancement factors
- Large number of levels
- Large number of levels
- Laser cooling
- Theoretical analysis

Effective electroweak Hamiltonian

$$\hat{H}_{\mathrm{ew}} = \hat{H}_{\mathrm{pc}} + \hat{H}_{\mathrm{pv}}$$

$$\hat{H}_{\mathrm{pc}} = \hat{H}_{\mathrm{em}} + \hat{H}_{\mathrm{w,pc}}$$

$$\hat{H}_{\mathrm{pv}} = \hat{H}_{\mathrm{w,pv}}$$

$$\hat{H}_{\mathrm{pc}}|+\rangle = E_{+}|+\rangle$$

$$\mathcal{P}|+\rangle = +|+\rangle$$

$$\hat{H}_{\mathrm{pc}}|-\rangle = E_{-}|-\rangle$$

$$\mathcal{P}|-\rangle = -|-\rangle$$

$$|L\rangle = \left(|+\rangle + |-\rangle\right)/\sqrt{2}$$

$$|L\rangle = \mathcal{P}|R\rangle$$

$$|R\rangle = \left(|+\rangle - |-\rangle\right)/\sqrt{2}$$

$$\begin{pmatrix} E_{+} & V_{\mathrm{pv}} \\ V_{\mathrm{pv}} & E_{-} \end{pmatrix} \begin{pmatrix} C_{+} \\ C_{-} \end{pmatrix} = E_{1,2} \begin{pmatrix} C_{+} \\ C_{-} \end{pmatrix}; \qquad \epsilon \sim \frac{V_{\mathrm{pv}}}{E_{+} - E_{-}}$$

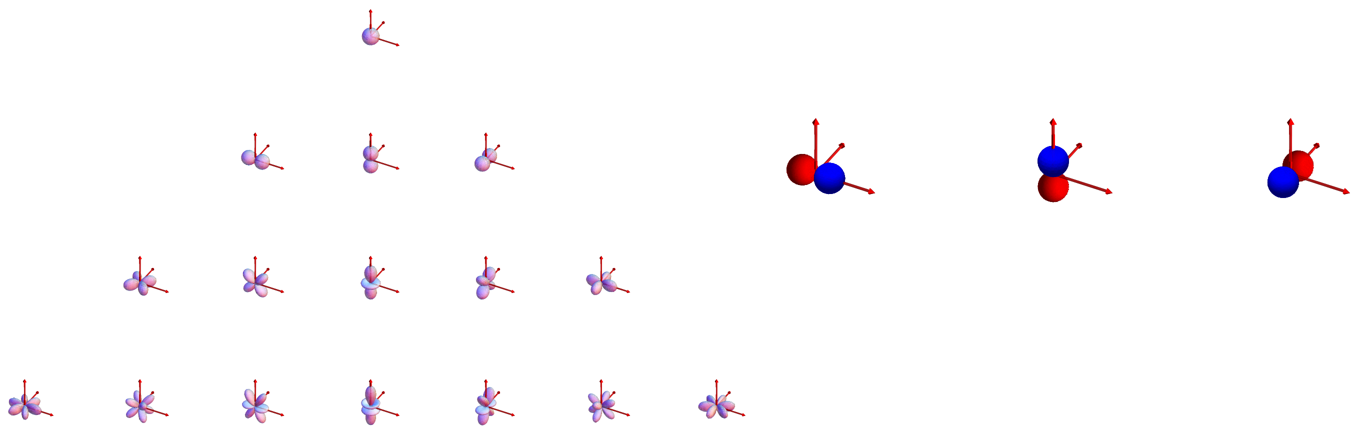
Levels of opposite parity in atoms

$l = 0$ s $|+\rangle$

$l = 1$ p $|-\rangle$

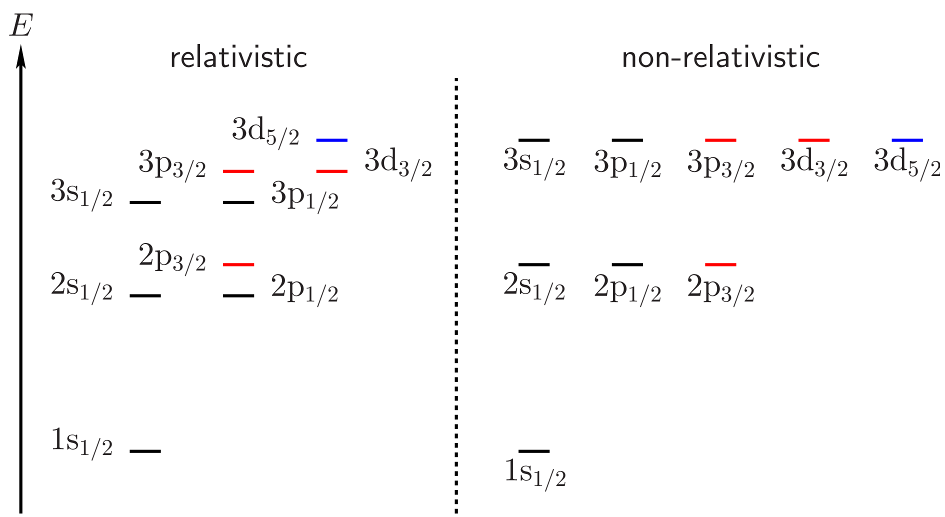
$l = 2$ d $|+\rangle$

$l = 3$ f $|-\rangle$

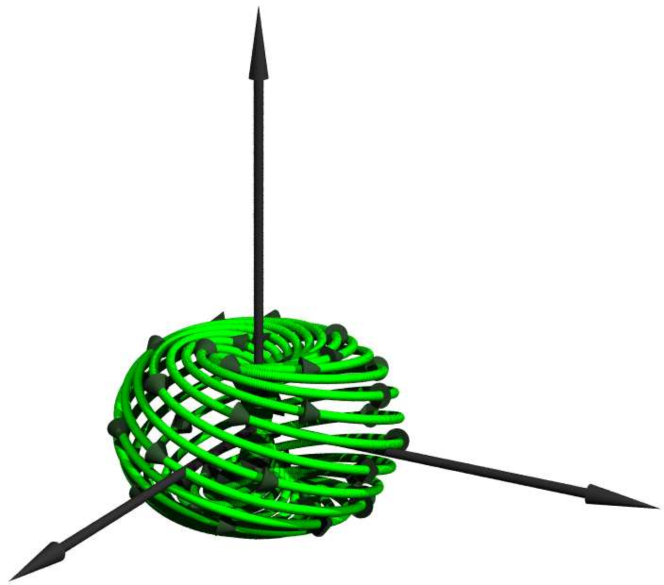


Lamb shift
$[E(2s_{1/2}) - E(2p_{1/2})]/h$:

H	1 GHz
He^{+}	14 GHz
Li^{2+}	63 GHz



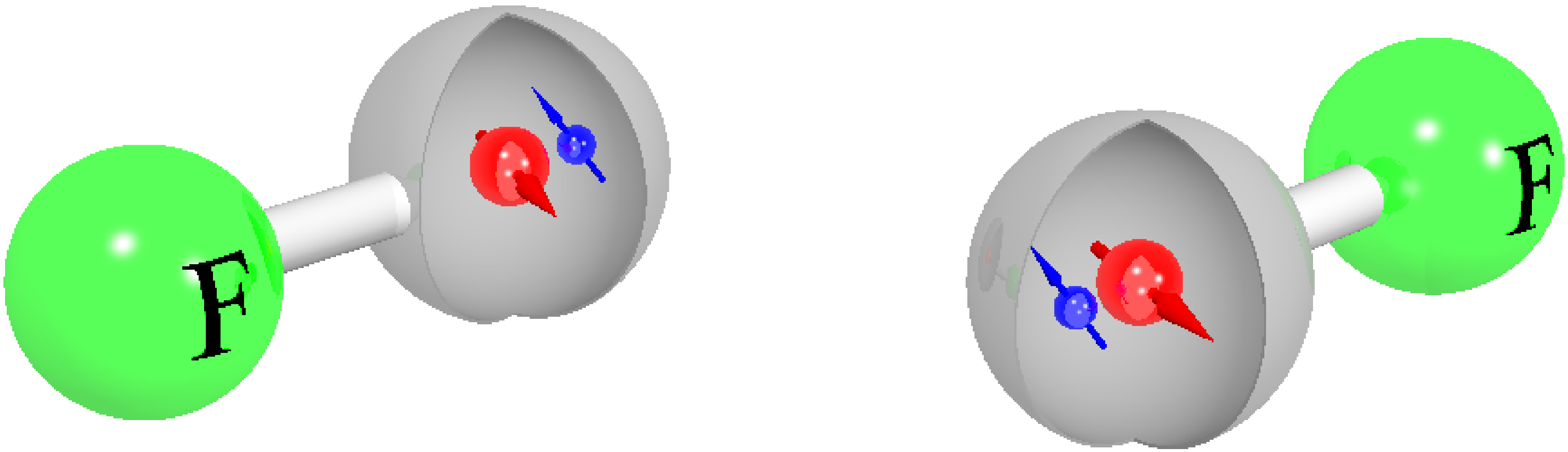
Levels of opposite parity in atoms



Cs $[E(7\mathrm{P}_{1/2}) - E(7\mathrm{S}_{1/2})]/h$: 95 THz

Wood, Bennett, Cho, Masterson, Roberts, Tanner, Wieman, *Science*, **1997**, *275*, 1759

Levels of opposite parity in diatomics

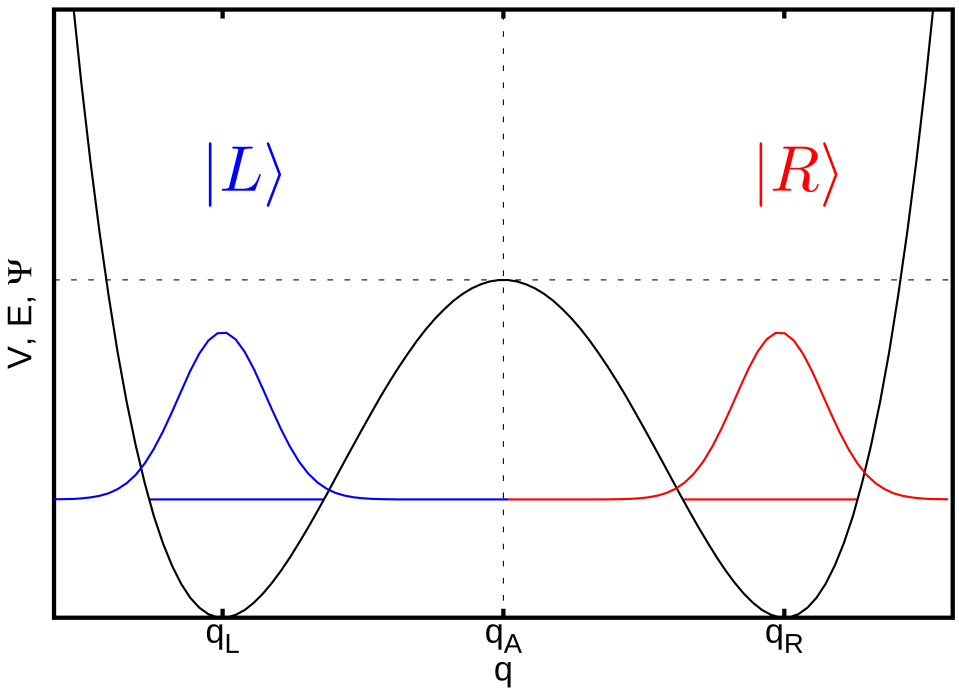


$$|\Omega\rangle = \mathcal{P}| - \Omega\rangle; \quad |\Omega\rangle = \mathcal{T}| - \Omega\rangle$$

Splitting depends on molecule and coupling situation:
On the order of 10 GHz in BaF, 0.3 GHz in HgF

Review: Kozlov, Labzowsky, *J. Phys. B*, **1995**, *28*, 1933; Kozlov, *Sov. Phys. JETP*, **1985**, *89*, 1114

Levels of opposite parity in chiral molecules

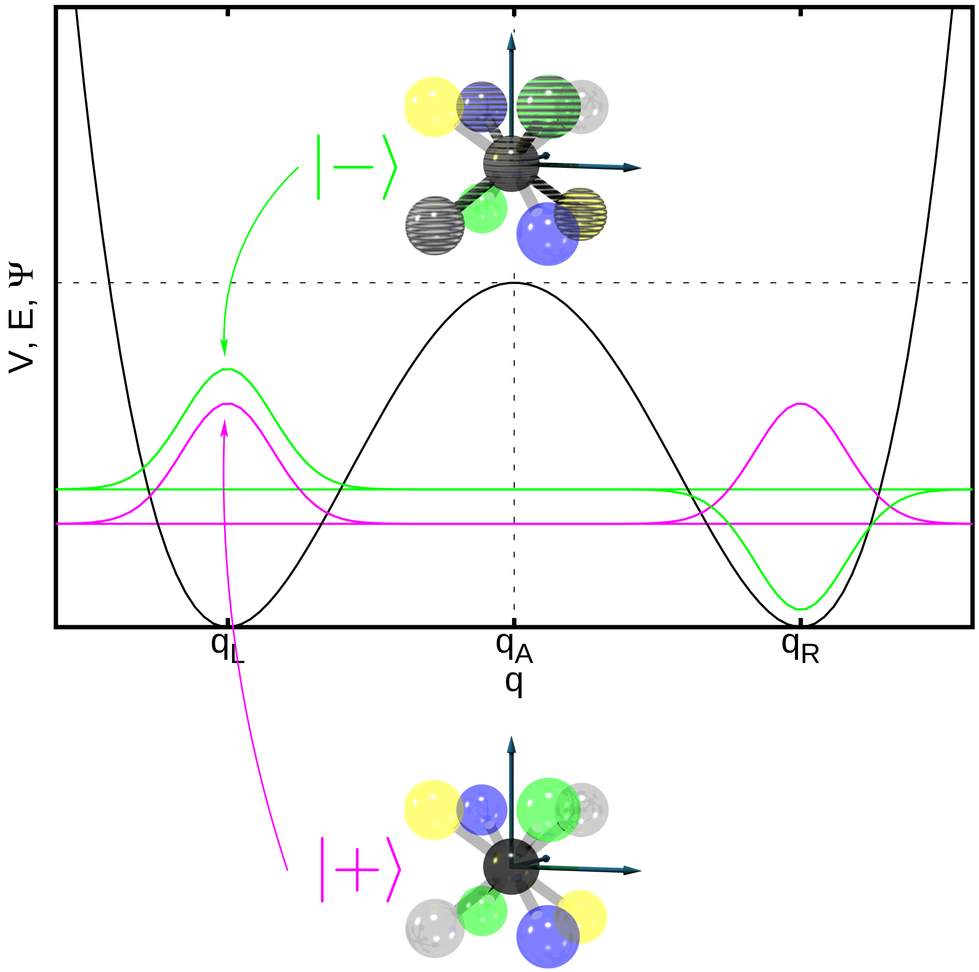


$$|L\rangle = \mathcal{P}|R\rangle$$

$$\begin{aligned}\langle L|\hat{H}_{\mathrm{em}}|L\rangle &= \langle R|\mathcal{P}^{-1}\hat{H}_{\mathrm{em}}\mathcal{P}|R\rangle \\ &= \langle R|\hat{H}_{\mathrm{em}}|R\rangle\end{aligned}$$

Hund, *Z. Phys.*, **1927**, *43*, 805

Levels of opposite parity in chiral molecules



$$\hat{H}_{\mathrm{em}}|+\rangle = E_{+}|+\rangle$$

$$\mathcal{P}|+\rangle = +|+\rangle$$

$$\hat{H}_{\mathrm{em}}|-\rangle = E_{-}|-\rangle$$

$$\mathcal{P}|-\rangle = -|-\rangle$$

$$|L\rangle = (|+\rangle + |-\rangle)/\sqrt{2}$$

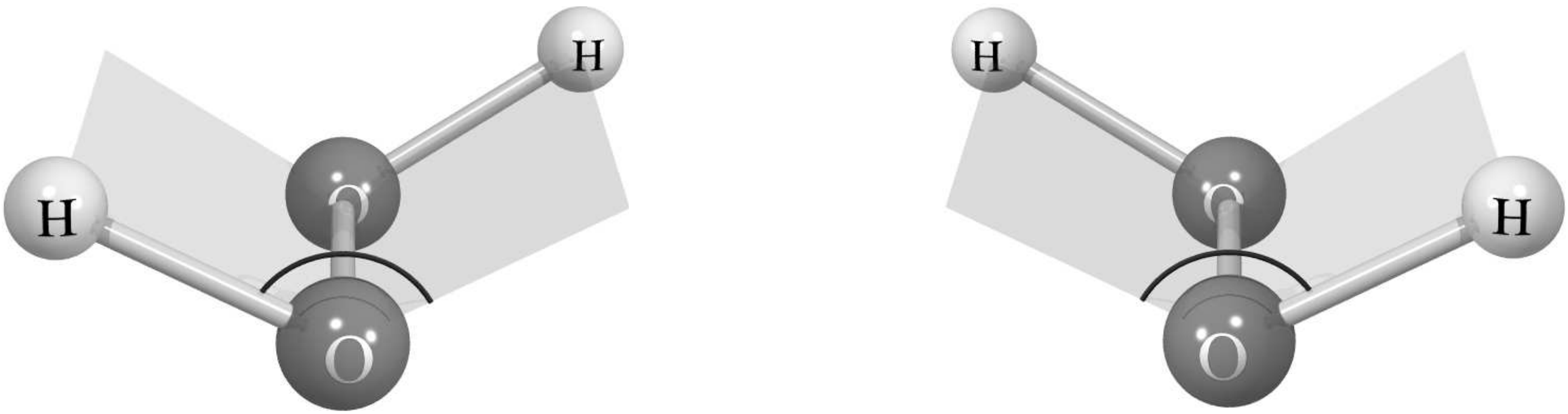
$$|L\rangle = \mathcal{P}|R\rangle$$

$$|R\rangle = (|+\rangle - |-\rangle)/\sqrt{2}$$

Hund, *Z. Phys.*, **1927**, *43*, 805

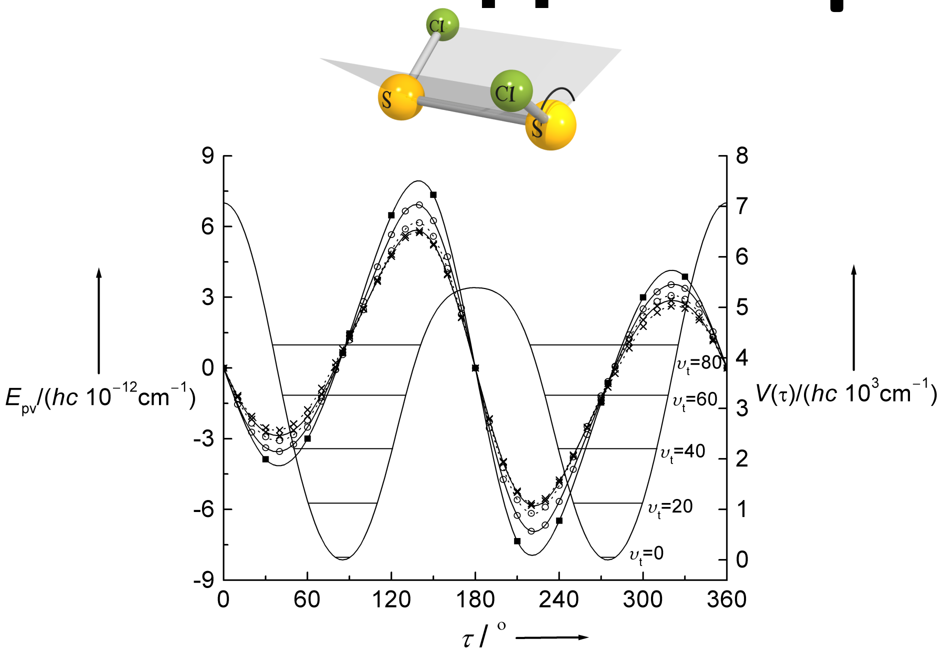
Levels of opposite parity in chiral molecules

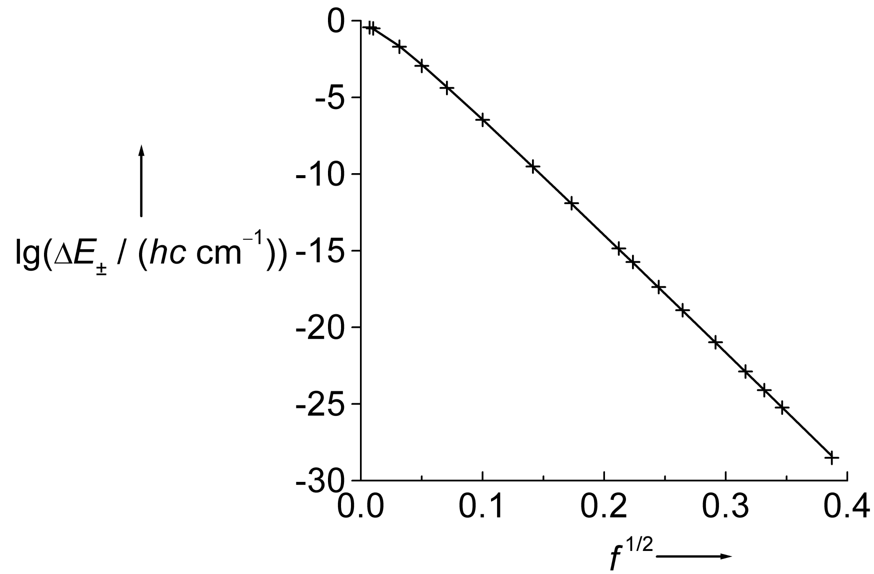
Hydrogen peroxide (H_2O_2)



$|E_+ - E_-|/h$ on the order of 300 GHz

Levels of opposite parity in chiral molecules





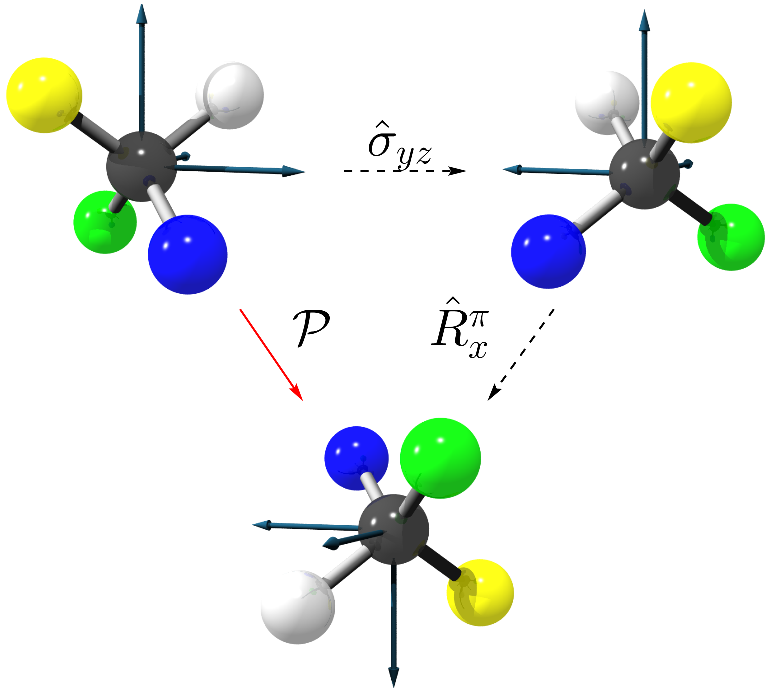
$$\lg\left[\frac{\Delta E_{\pm}}{hc\ \mathrm{cm}^{-1}}\right] = P_1 \lg\sqrt{f} + P_2 - P_3\sqrt{f}$$

$$\Delta E_{\mathrm{pv}}/hc \approx 10^{-12}\ \mathrm{cm}^{-1} \gg 10^{-76}\ \mathrm{cm}^{-1} \approx \Delta E_{\pm}/hc$$

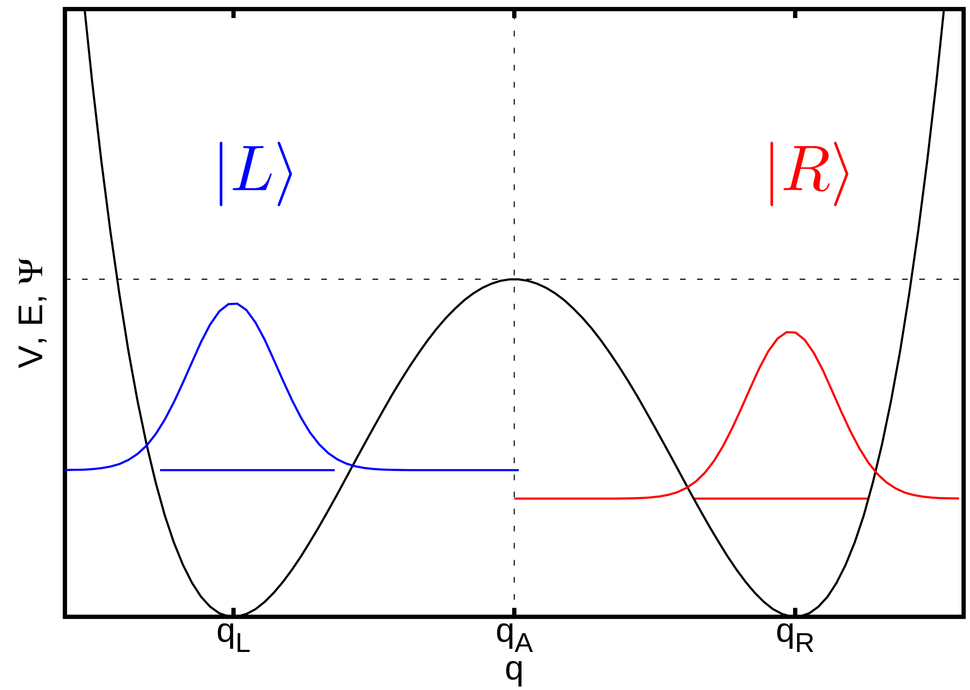
$|E_{+} - E_{-}|/h$ on the order of 10^{-75} GHz or 10^{-42} yHz

Berger, Gottselig, Quack, Willeke, *Angew. Chem. Int. Ed.*, **2001**, *40*, 4195

Molecular parity violation



$$|E_+ - E_-| \ll |E_{\mathrm{pv},L} - E_{\mathrm{pv},R}|$$

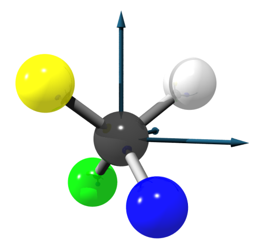


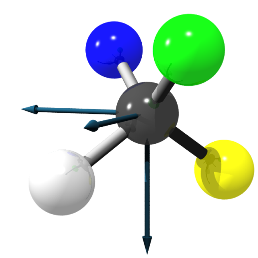
$$\mathcal{P}^{-1}\hat{H}_{\mathrm{pc}}\mathcal{P} = \hat{H}_{\mathrm{pc}}$$

$$\mathcal{P}^{-1}\hat{H}_{\mathrm{pv}}\mathcal{P} = -\hat{H}_{\mathrm{pv}}$$

$$E_{\mathrm{pv},L} = \langle L|\hat{H}_{\mathrm{pv}}|L\rangle$$

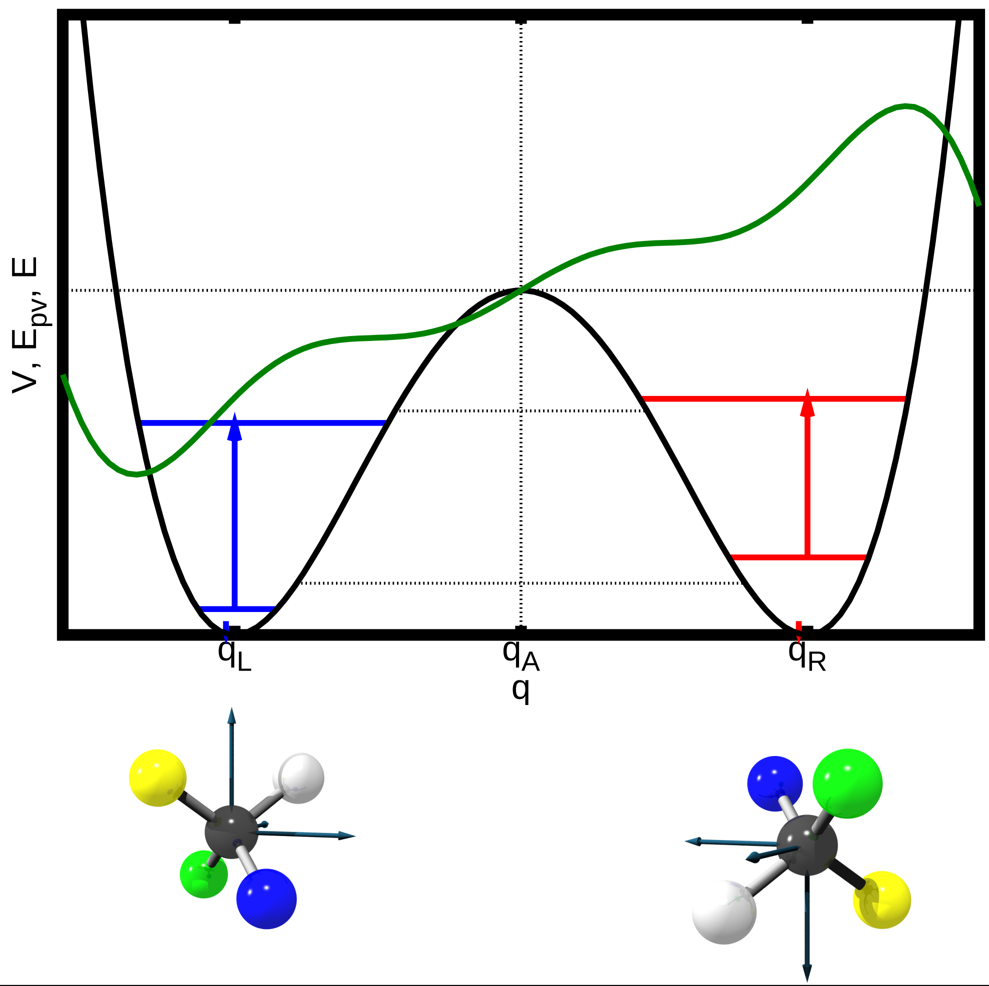
$$= -\langle R|\hat{H}_{\mathrm{pv}}|R\rangle = -E_{\mathrm{pv},R}$$





Reviews: Berger in: Relativistic electronic structure theory, Part 2, (Ed: P. Schwerdtfeger), **2004**, 188; Crassous, Chardonnet, Saue, Schwerdtfeger, *Org. Biomol. Chem.*, **2005**, *3*, 2218; Quack, Stohner, Willeke, *Annu. Rev. Phys. Chem.*, **2008**, *59*, 741

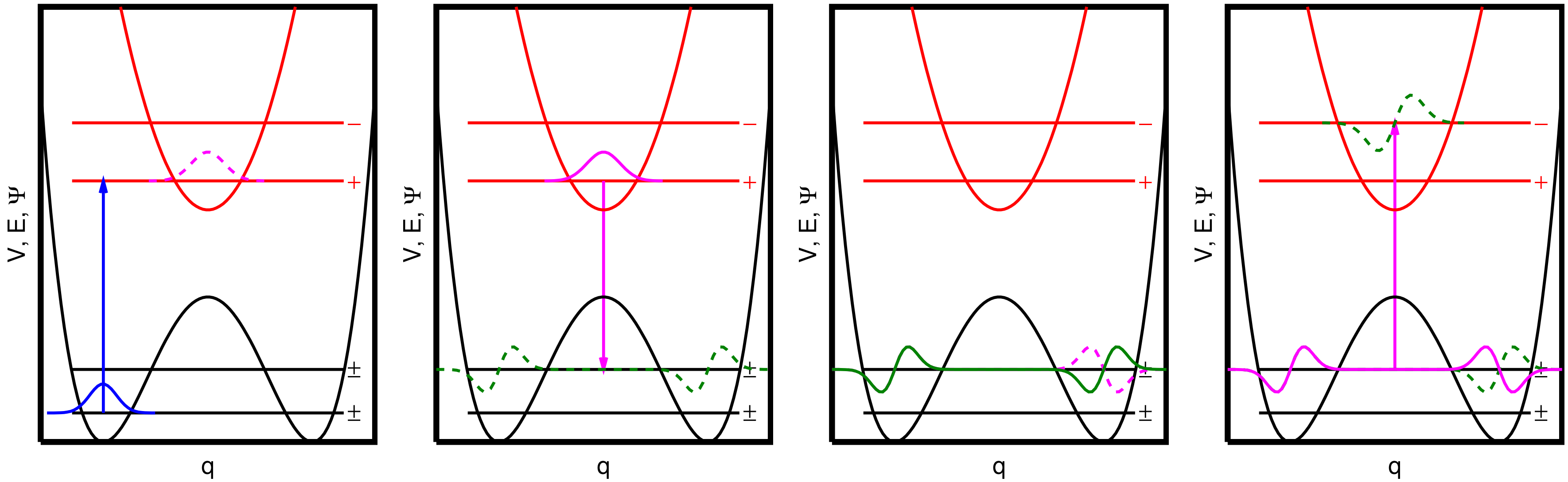
Molecular parity violation



$$\Delta\nu_{\mathrm{pv}} = \nu_{\mathrm{L}} - \nu_{\mathrm{R}}$$

Letokhov, *Phys. Lett. A*, **1975**, *53*, 275; Kompanets, Kukudzhanov, Letokhov, Gervits, *Opt. Commun*, **1976**, *19*, 414; Arimondo, Glorieux, Oka, *Opt. Commun.*, **1977**, *23*, 369; Daussy, Marrel, Amy-Klein, Nguyen, Bordé, Chardonnet, *Phys. Rev. Lett.*, **1999**, *83*, 1554; Berger, Quack, Sieben, Willeke, *Helv. Chim. Acta*, **2003**, *86*, 4048; Berger, Laubender, Quack, Sieben, Stohner, Willeke, *Angew. Chem. Int. Ed.*, **2005**, *44*, 3623;

Molecular parity violation



$$\begin{pmatrix} E_+ & V_{\rm pv} \\ V_{\rm pv} & E_- \end{pmatrix} \begin{pmatrix} C_+ \\ C_- \end{pmatrix} = E_{1,2} \begin{pmatrix} C_+ \\ C_- \end{pmatrix}$$

$$|C_+|^2 = \sin^2 \{\Delta E_{\rm pv} t/\hbar\}$$

Diatomics:

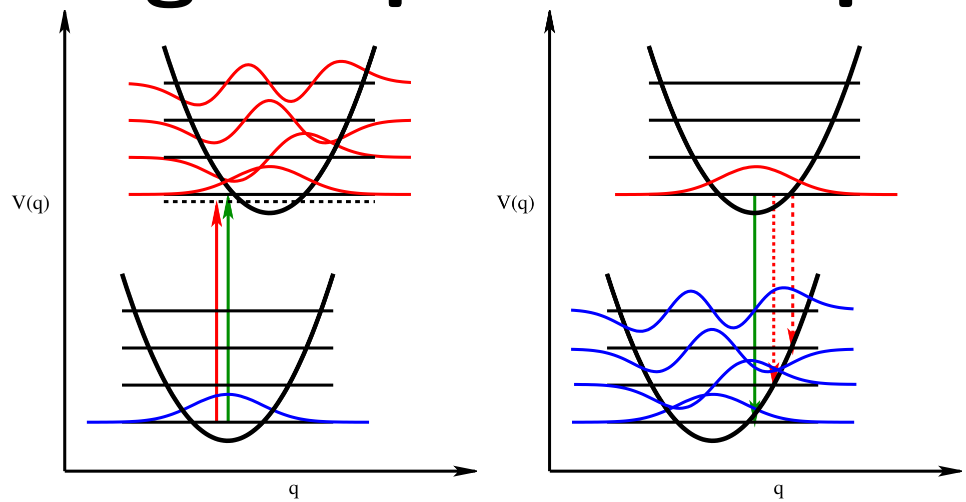
$$\begin{pmatrix} E_+ & dE + {\rm i}W_{\rm pv} \\ dE - {\rm i}W_{\rm pv} & E_- \end{pmatrix} \begin{pmatrix} C_+ \\ C_- \end{pmatrix} = E_{1,2} \begin{pmatrix} C_+ \\ C_- \end{pmatrix}$$

Chiral molecules: Quack, *Chem. Phys. Lett.*, **1986**, *132*, 147; Berger, *Phys. Chem. Chem. Phys.*, **2003**, *4*, 395–399;
Diatomic molecules: DeMille, Cahn, Murphree, Rahmlow, Kozlov, *Phys. Rev. Lett.*, **2008**, *100*, 023003

Opportunities and Challenges

- Close-lying levels of opposite parity
- Strong enhancement factors
- Huge number of levels
- Huge number of levels
- Laser cooling
- Theoretical analysis

Laser cooling for precision spectroscopy



FCF	1 000 cyc.	10 000 cyc.	100 000 cyc.	1 000 000 cyc.
0.9	0	0	0	0
0.99	0.000043	0	0	0
0.999	0.368	0.0000452	0	0
0.9999	0.9048	0.3679	0.0000454	0
0.99999	0.99005	0.90484	0.36788	0.0000453

Classes for cooling of molecules with lasers:

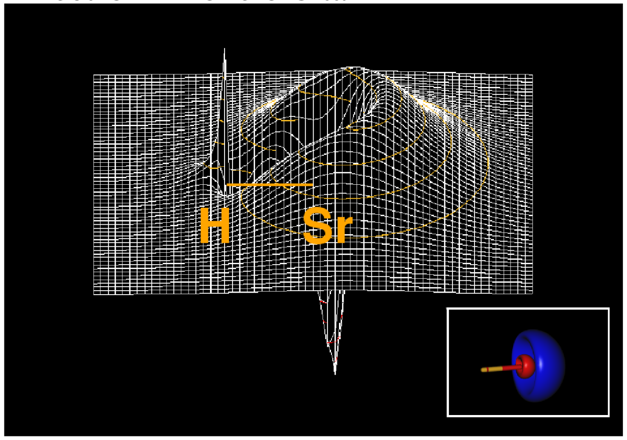
Class 1: Electron in lone orbital; Class 2: Electron in atom-like orbital;

Class 3: Electron in diffuse orbital (heavy element)

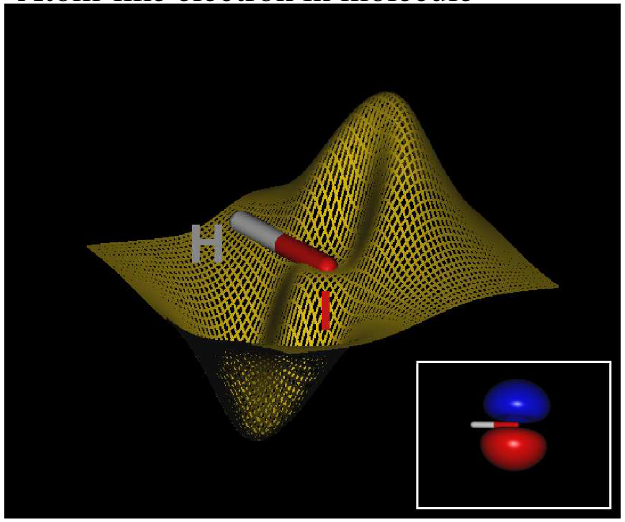
Diatomics: DiRosa, *Eur. Phys. J. D*, **2004**, *31*, 395; Classification: Isaev, Hoekstra, Berger, *Phys. Rev. A*, **2010**, *82*, 052521; Experiment: Shuman, Barry, DeMille, *Nature*, **2010**, *467*, 820; Review: Isaev, Berger, *Chimia*, **2018**, *72*, 375

Classes for cooling with lasers

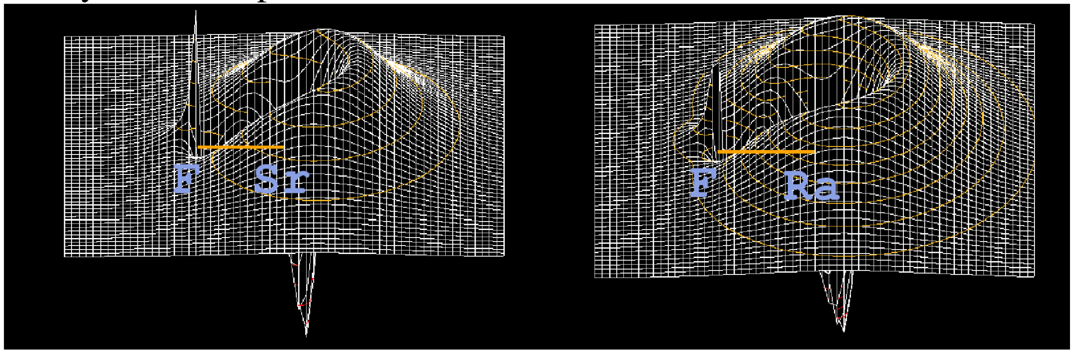
1. "Electron in lone orbital"



2. "Atom-like electron in molecule"

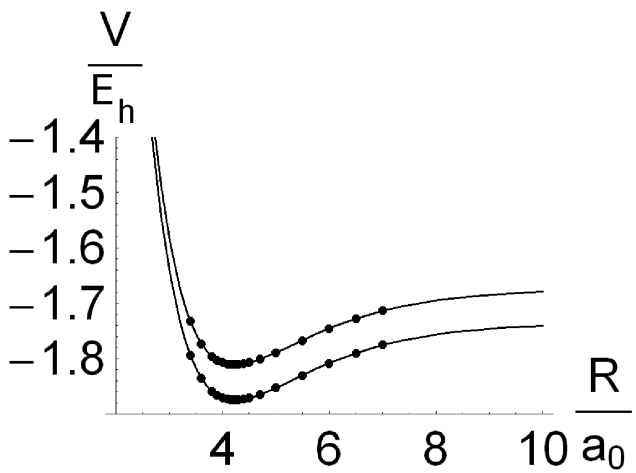


3. Heavy-atom compounds



Shuman, Barry, DeMille, *Nature*, **2010**, *467*, 820; Isaev, Hoekstra, Berger, *Phys. Rev. A*, **2010**, *82*, 052521; Hummon, Yeo, Stuhl, Collopy, Xia, Ye, *Phys. Rev. Lett.*, **2013**, *110*, 143001

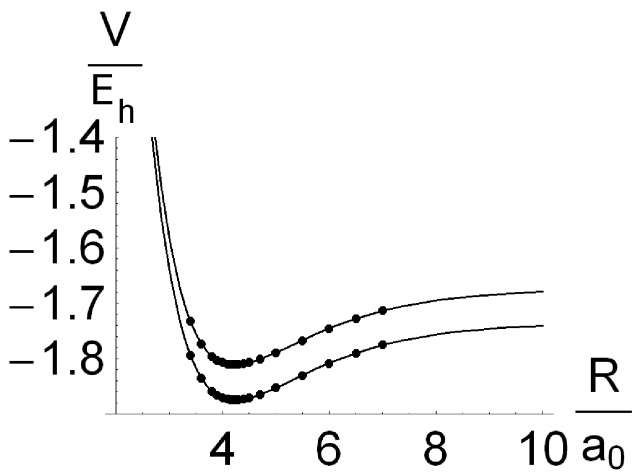
Molecular parameters of RaF



	RaF
	4c FS-CCSD
$r_e(^2\Pi_{1/2})/a_0$	4.24
$r_e(^2\Sigma_{1/2})/a_0$	4.24
$\tilde{\omega}_e(^2\Pi_{1/2})/\text{cm}^{-1}$	428
$\tilde{\omega}_e(^2\Sigma_{1/2})/\text{cm}^{-1}$	432
$\tilde{D}_e(^2\Pi_{1/2})/\text{cm}^{-1}$	3.13 10^4
$\tilde{D}_e(^2\Sigma_{1/2})/\text{cm}^{-1}$	3.21 10^4
$\tilde{T}_e/\text{cm}^{-1}$	1.40 10^4

Isaev, Hoekstra, Berger, *Phys. Rev. A*, **2010**, *82*, 052521; Isaev, Berger, *arXiv:1302.5682*

Molecular parameters of RaF



	RaF
	4c FS-CCSD
$r_{\mathrm{e}}(^2\Pi_{1/2})/a_0$	4.29
$r_{\mathrm{e}}(^2\Sigma_{1/2})/a_0$	4.29
$\tilde{\omega}_{\mathrm{e}}(^2\Pi_{1/2})/\mathrm{cm}^{-1}$	428
$\tilde{\omega}_{\mathrm{e}}(^2\Sigma_{1/2})/\mathrm{cm}^{-1}$	431
$\tilde{D}_{\mathrm{e}}(^2\Pi_{1/2})/\mathrm{cm}^{-1}$	4.24 10^4
$\tilde{D}_{\mathrm{e}}(^2\Sigma_{1/2})/\mathrm{cm}^{-1}$	4.26 10^4
$\tilde{T}_{\mathrm{e}}/\mathrm{cm}^{-1}$	1.33 10^4

Improved estimates: Isaev, Berger, *arXiv:1302.5682*; Isaev, Hoekstra, Willmann, Berger, *arXiv:1310.1511*

Proposed synthesis of RaF

Thermoneutral fluorination:

$$\mathrm{Ra}^+ \xrightarrow{\mathrm{CF}_4} \mathrm{RaF}^+ \xrightarrow{\mathrm{Cs}} \mathrm{RaF}^*(^2\Pi) \xrightarrow{-h\nu} \mathrm{RaF}(^2\Sigma)$$

$$\mathrm{Ra}^+ \xrightarrow{\mathrm{CF}_4} \mathrm{RaF}^+ \xrightarrow{\mathrm{Na}} \mathrm{RaF}(^2\Sigma)$$

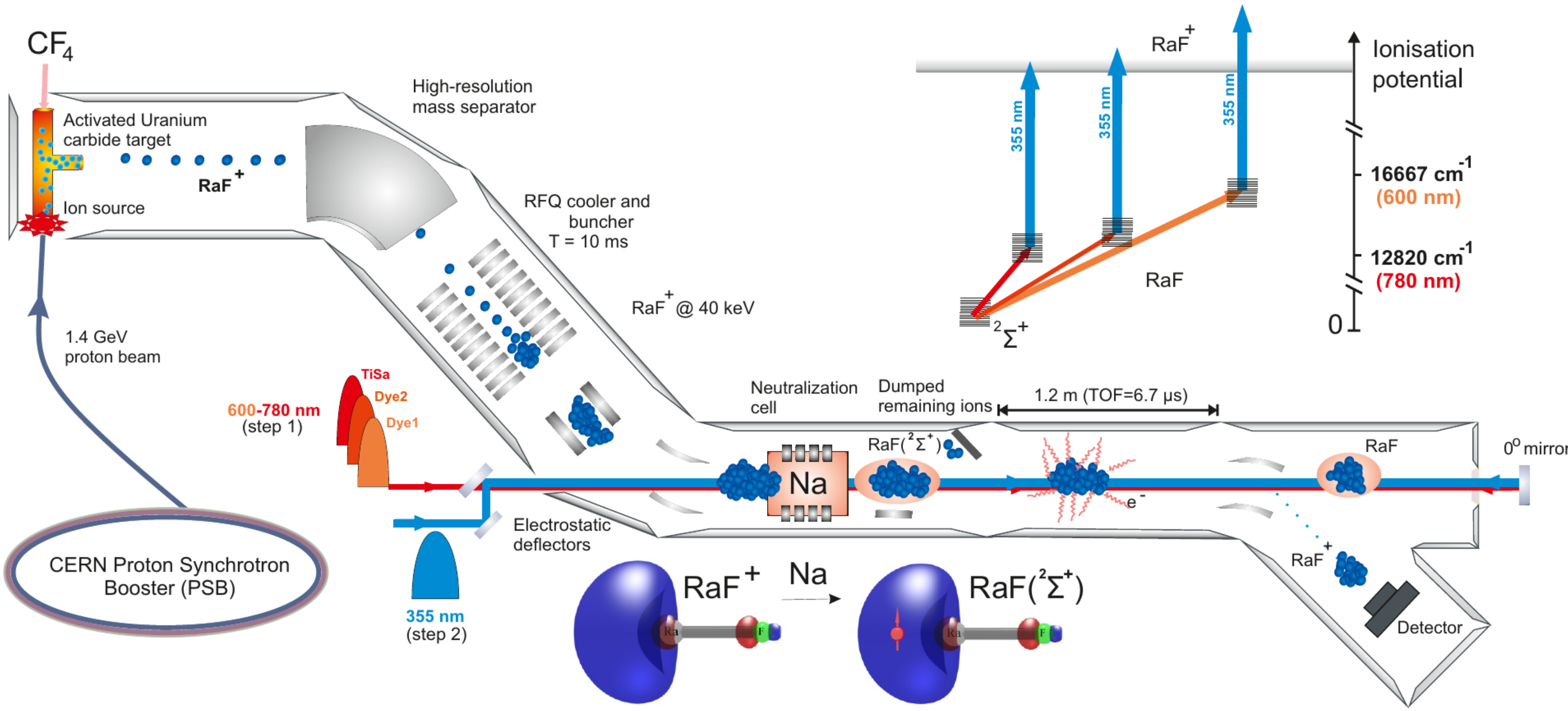
Endothermic fluorination:

$$\mathrm{Ra}^+ \xrightarrow{\mathrm{SiF}_4} \mathrm{RaF}^+ \xrightarrow{\mathrm{Cs}} \mathrm{RaF}^*(^2\Pi) \xrightarrow{-h\nu} \mathrm{RaF}(^2\Sigma)$$

$$\mathrm{Ra}^+ \xrightarrow{\mathrm{SiF}_4} \mathrm{RaF}^+ \xrightarrow{\mathrm{Na}} \mathrm{RaF}(^2\Sigma)$$

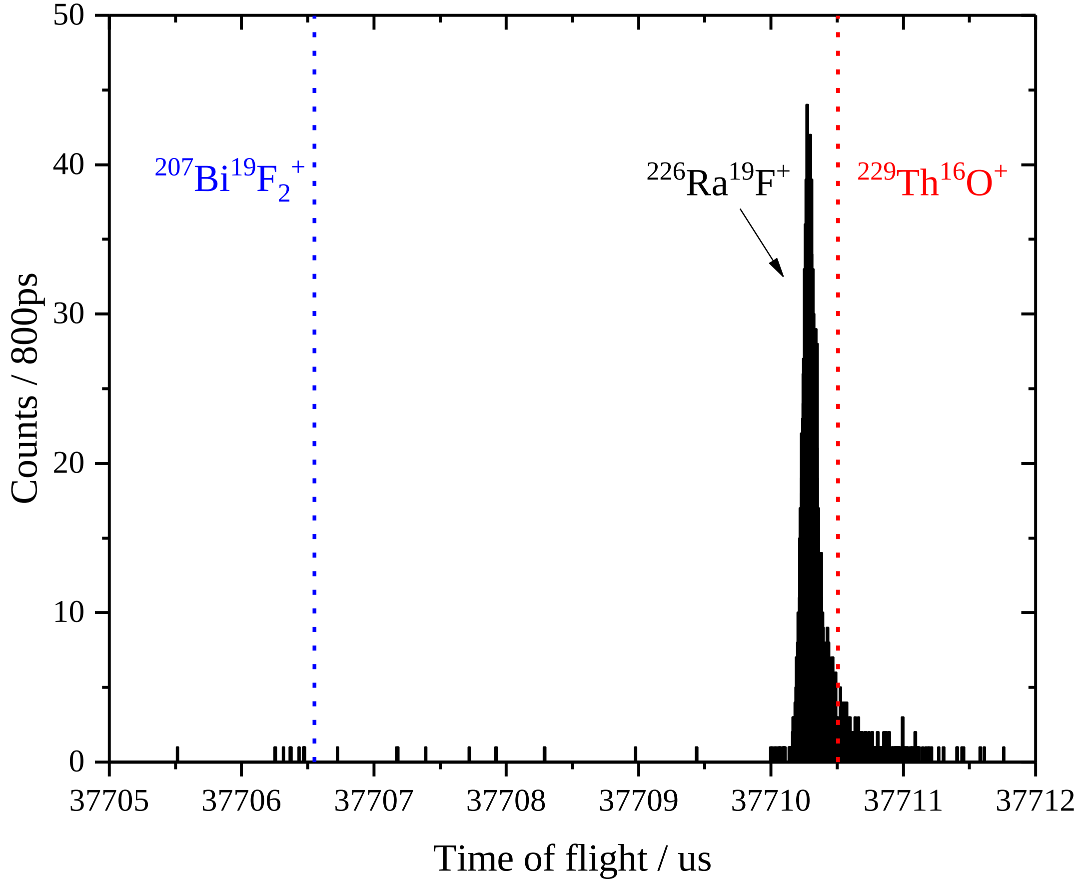
Isaev, Hoekstra, Willmann, Berger, *arXiv:1310.1511*

RaF



Garcia Ruiz, Berger, Billowes, Binnersley, Bissell, Breier, Brinson, Chrysalidis, Cocolios, Cooper, Flanagan, Giesen, de Groote, Franchoo, Gustafsson, Isaev, Koszorús, Neyens, Perrett, Ricketts, Rothe, Schweikhard, Vernon, Wendt, Wienholtz, Wilkins, Yang, *Nature*, **2020**, *581*, 396

RaF



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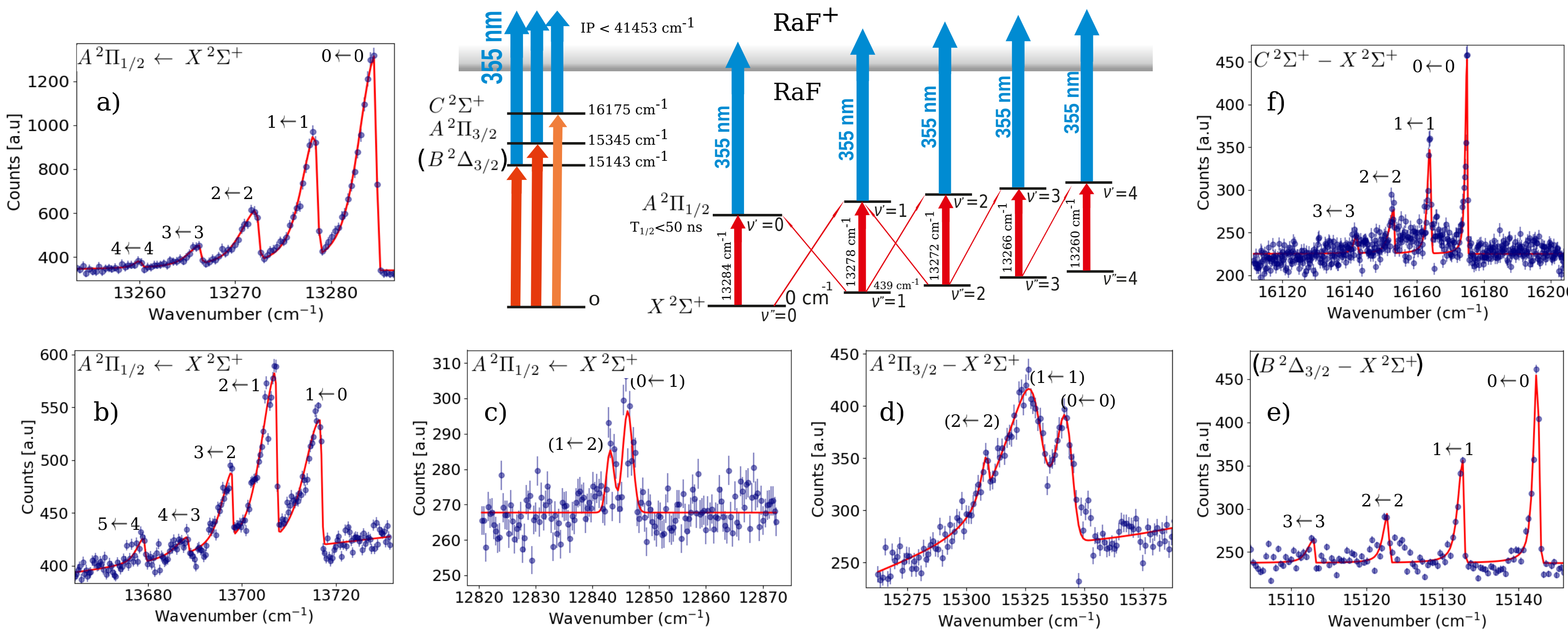
RaF

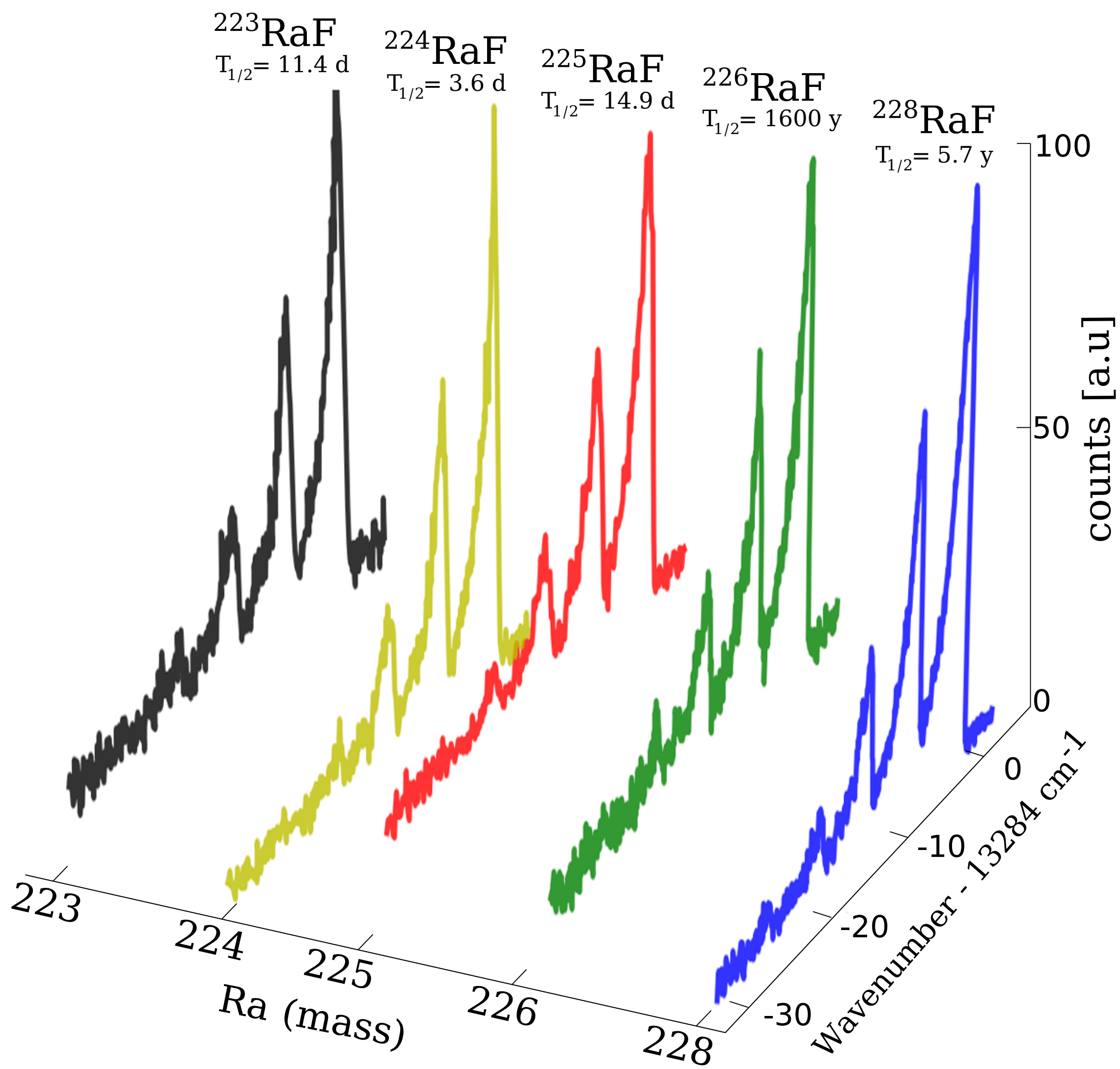


$$\text{A } {}^2\Pi_{1/2} \leftarrow \text{X } {}^2\Sigma^+,\ 0' - 0 \text{ transition}$$

$$\tilde{\nu}_{\text{exp}} = 13284.7(5)\ \text{cm}^{-1}; \tilde{\nu}_{\text{theo}} = 13300(1200)\ \text{cm}^{-1}$$

Garcia Ruiz, Berger, Billowes, Binnersley, Bissell, Breier, Brinson, Chrysalidis, Cocolios, Cooper, Flanagan, Giesen, de Groote, Franchoo, Gustafsson, Isaev, Koszorús, Neyens, Perrett, Ricketts, Rothe, Schweikhard, Vernon, Wendt, Wienholtz, Wilkins, Yang, *Nature*, **2020**, *581*, 396

RaF



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Cooling of polyatomics with lasers?

M=Mg, Ca, Sr, Ba, Ra; MF $\rightarrow$ MX

X = pseudohalogen

NC	CN
PC	CP
NCO	OCN
NCS	SCN
NCSe	SeCN
N_3	

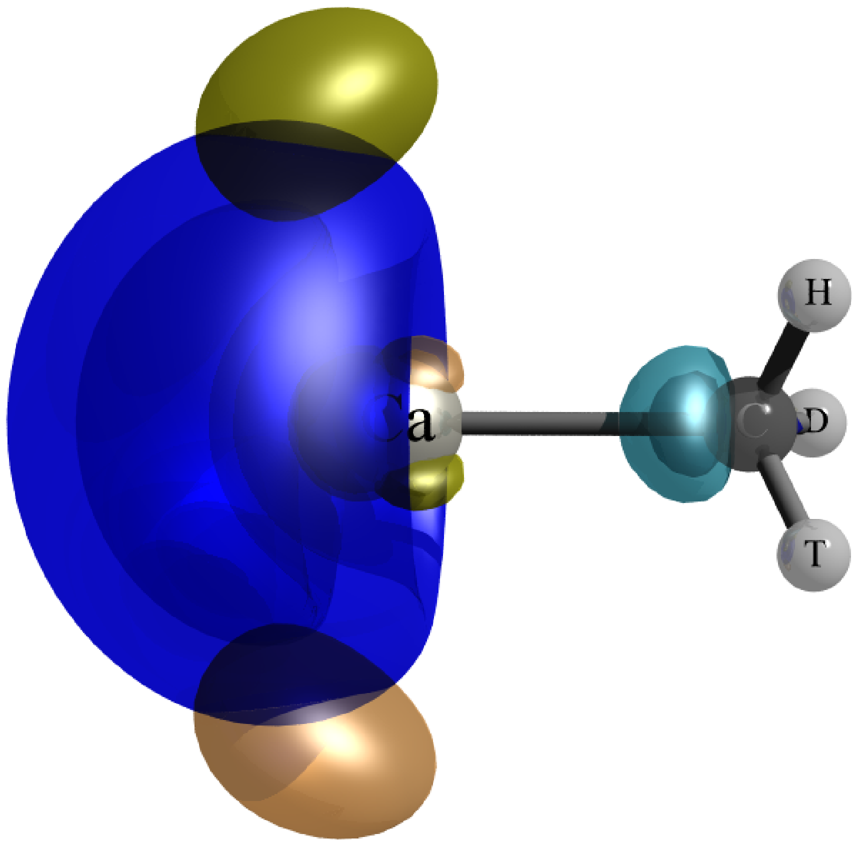
X = residues such as OH, SH, CH_3

Isaev, Berger, *Conference on Cold and Controlled Molecules and Ions*, Monte Verita, Switzerland, **2014**;
Isaev, Berger, *arXiv:1504.08326*; *Phys. Rev. Lett.*, **2016**, *116*, 063006;
Pseudohalogens: Birckenbach, Kellermann, *Ber. Dtsch. Chem. Ges.*, **1925**, *58*, 786

Cooling of polyatomics with lasers

Proposed Candidates:

CaOH, CaNC, $MgCH_3$, $CaCH_3$, CaCHDT . . .



Isaev, Berger, *Conference on Cold and Controlled Molecules and Ions*, Monte Verita, Switzerland, **2014**;
Isaev, Berger, *arXiv:1504.08326*; *Phys. Rev. Lett.*, **2016**, *116*, 063006; Review: Isaev, Berger, *Chimia*, **2018**, *72*, 375
Review on chiral methyl group: Floss, Lee, *Acc. Chem. Res.*, **1993**, *26*, 116.

Opportunities and Challenges

- Close-lying levels of opposite parity
- Strong enhancement factors
- Huge number of levels
- Huge number of levels
- Laser cooling
- Theoretical analysis

Nuclear physics and BSM physics with molecules

Standard model physics		BSM physics
P-even	P-odd	P,T-odd
Charge radius	Weak nuclear charge	k_s, k_T, k_p
Magnetic dipole moment	Anapole moment	EDMs, Schiff moment
Electric quadrupole moment		Magnetic quadrupole moment
Magnetic octupole moment		Electric octupole moment
		ALPs
		WIMPs
		fuzzy CDM

P,T-odd electric dipole moments

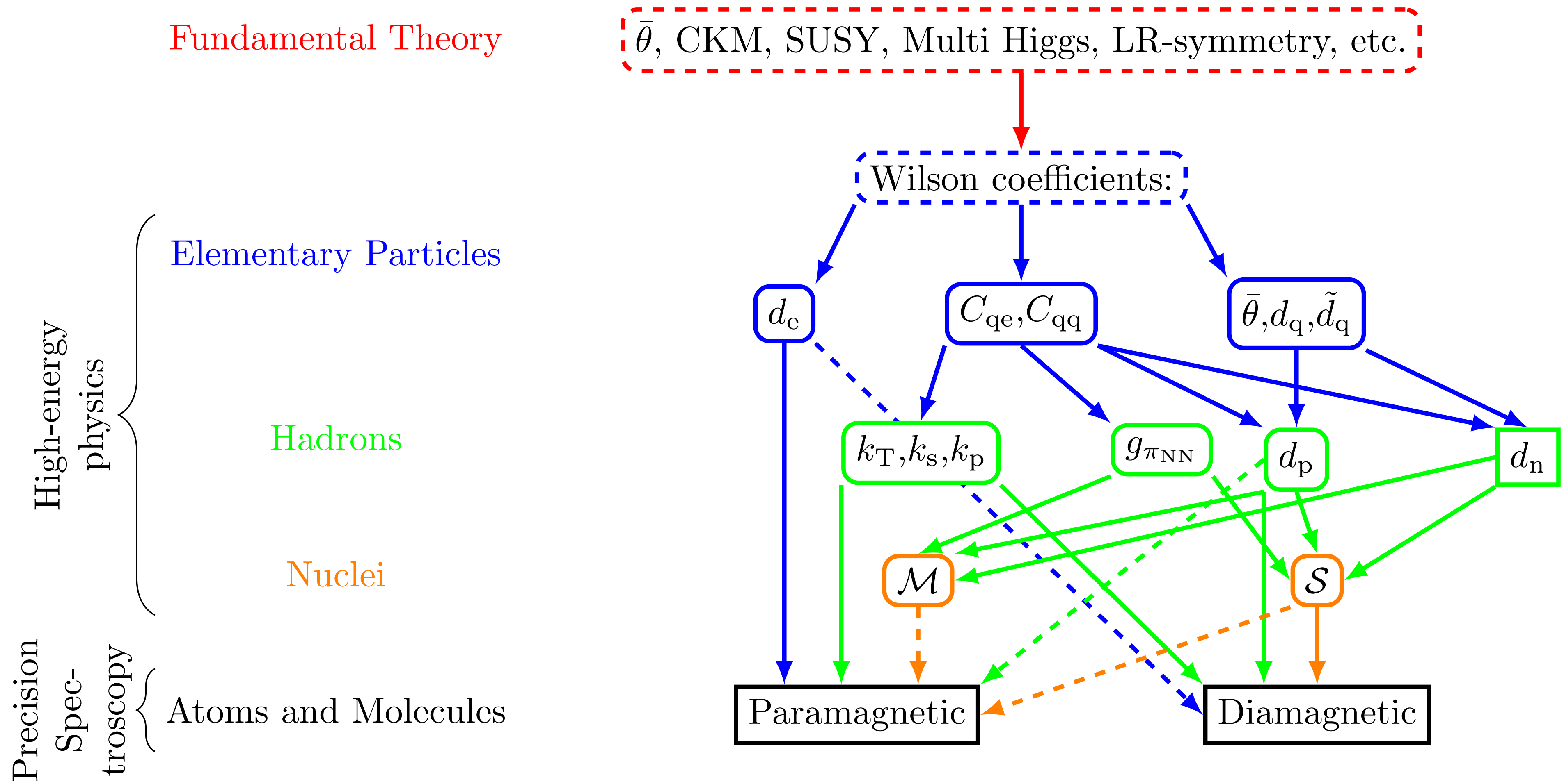
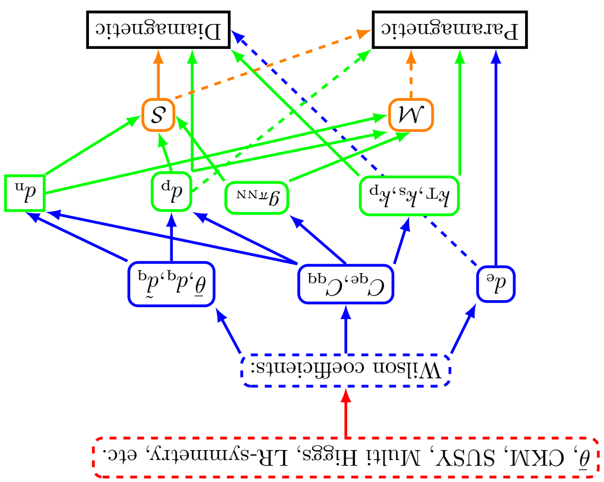


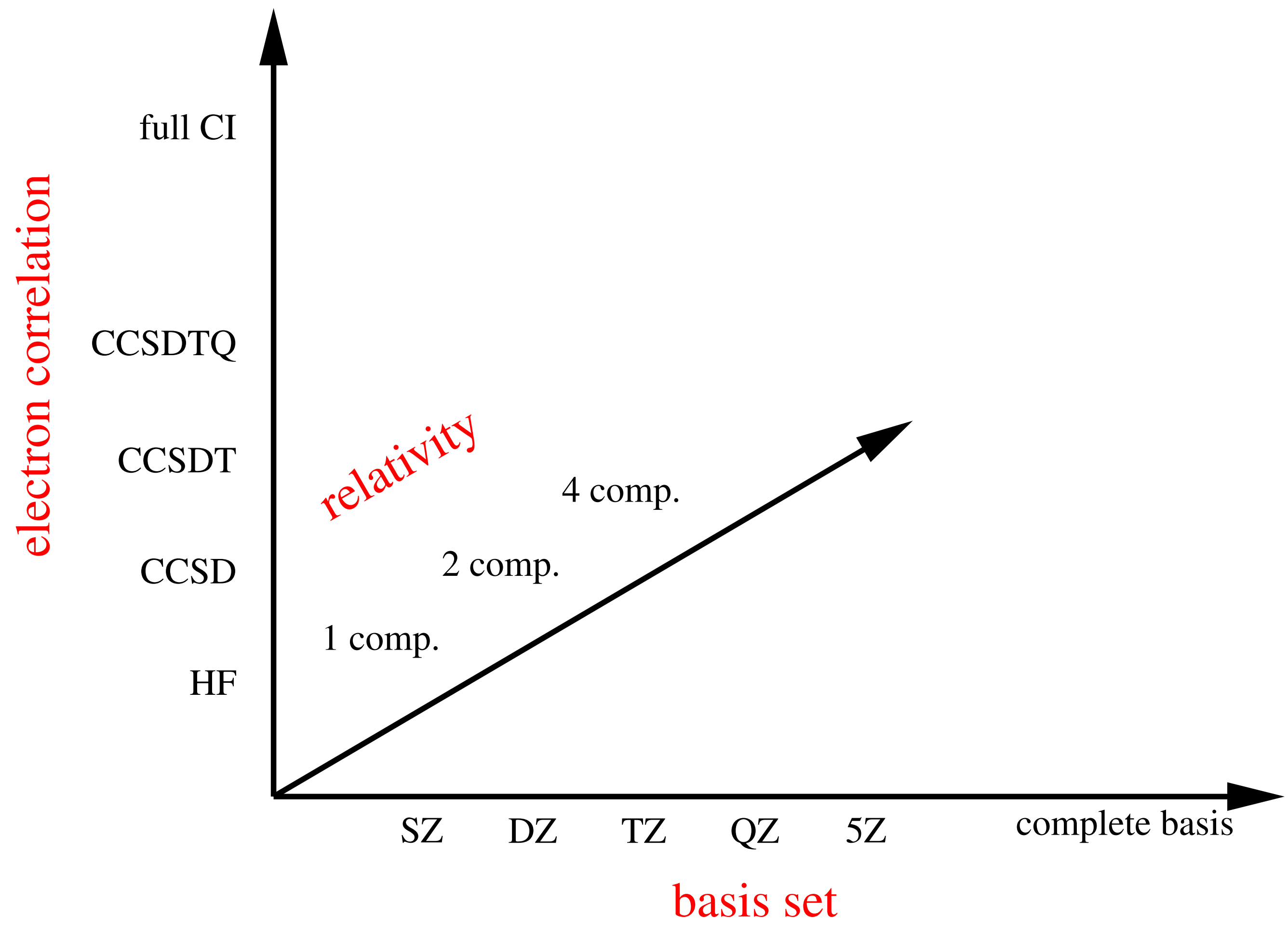
Figure by Konstantin Gaul

P,T-odd electric dipole moments

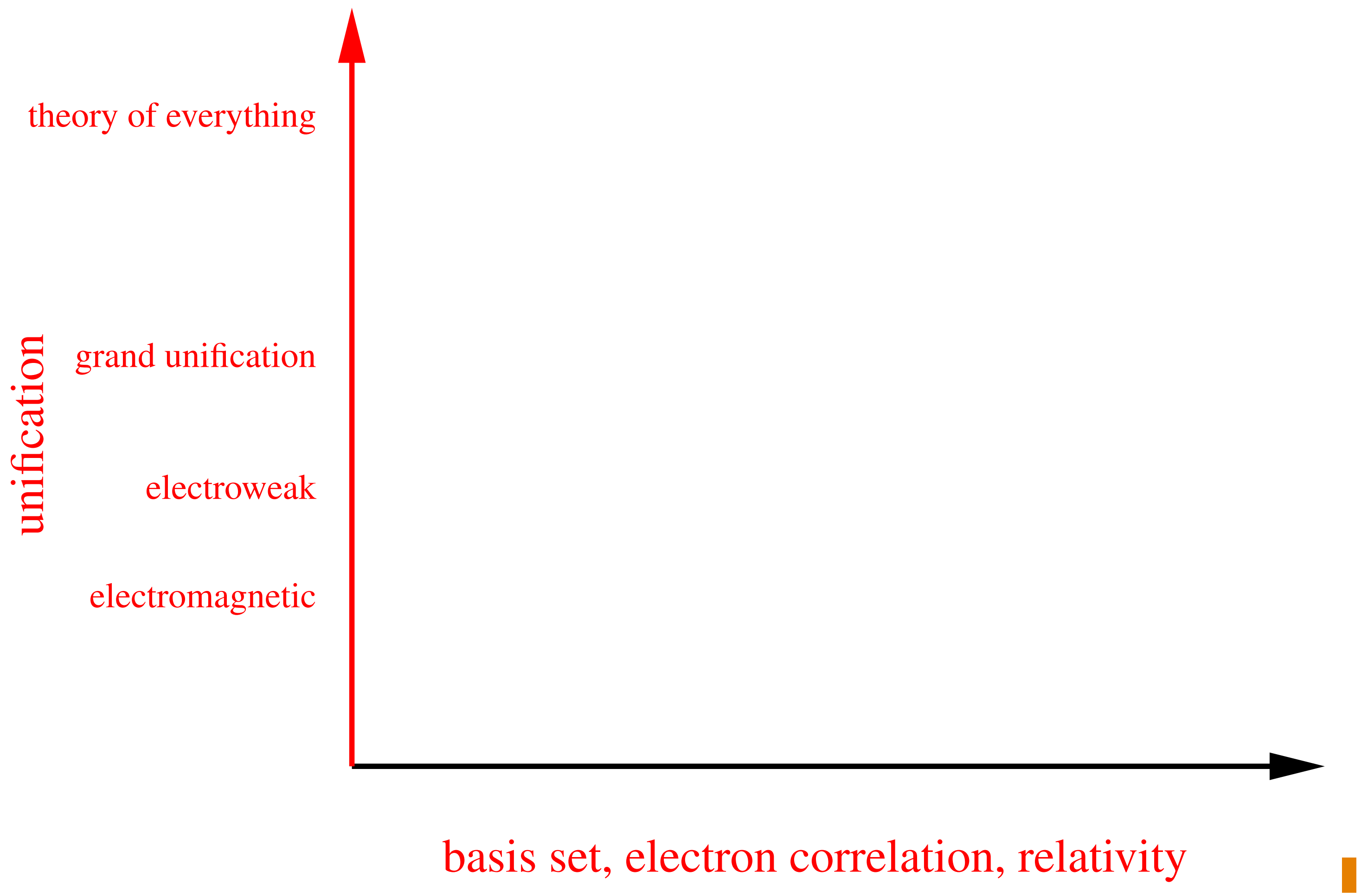


Biology
Biochemistry
Chemistry
Molecular physics
Atomic physics
Nuclear physics
Particle physics

Traditional quantum chemistry



Beyond traditional quantum chemistry



P-odd effects, 1c vs. 4c approaches

$$\hat{H}_{\text{pv}}^{(\text{e}-\text{nucl})} = \hat{H}_{\text{pv}}^{(\text{e}-\text{nucl},1)} + \hat{H}_{\text{pv}}^{(\text{e}-\text{nucl},2)} = \sum_{i=1}^{n} \left[\hat{h}_{\text{pv}}^{(1)}(i) + \hat{h}_{\text{pv}}^{(2)}(i) \right]$$

Schrödinger picture

$$\hat{h}_{\text{pv}}^{(1)} = \frac{G_{\text{F}}}{4\sqrt{2} m_{\text{e}} c} \sum_{A=1}^{N} Q_{\text{w},A} \{ \vec{\sigma} \cdot \vec{p}, \rho_A(\vec{r}) \}_+$$

Dirac picture

$$\hat{h}_{\text{pv}}^{(1)} = \frac{G_{\text{F}}}{2\sqrt{2}} \sum_{A=1}^{N} Q_{\text{w},A} \boldsymbol{\gamma}^5 \rho_A(\vec{r}); \quad \gamma^5 = \begin{bmatrix} \mathbf{0}_{2\times 2} & \mathbf{1}_{2\times 2} \\ \mathbf{1}_{2\times 2} & \mathbf{0}_{2\times 2} \end{bmatrix}$$

Bouchiat, Bouchiat, *J. Phys.*, **1974**, *35*, 899; Hegstrom, Rein, Sandars, *J. Chem. Phys.*, **1980**, *73*, 2329; Wiesenfeld, *Mol. Phys.*, **1988**, *64*, 739; Berger in: Relativistic electronic structure theory, Part 2, (Ed: P. Schwerdtfeger), **2004**

P-odd effects, 1c vs. 4c approaches

$$\frac{1}{2}Q_{\mathrm{w},A}\,\rho_A(\vec{r}) = (Zg_{\mathrm{V}}^{\mathrm{p}} + Ng_{\mathrm{V}}^{\mathrm{n}})\rho_A(\vec{r})$$

$$\approx Zg_{\mathrm{V}}^{\mathrm{p}}\rho_{\mathrm{p},A}(\vec{r}) + Ng_{\mathrm{V}}^{\mathrm{n}}\rho_{\mathrm{n},A}(\vec{r}) = \rho'_{\mathrm{av},A}(\vec{r})$$

P-odd effects, 1c vs. 4c approaches

$$\hat{H}_{\mathrm{pv}}^{(\mathrm{e-nucl})} = \hat{H}_{\mathrm{pv}}^{(\mathrm{e-nucl},1)} + \hat{H}_{\mathrm{pv}}^{(\mathrm{e-nucl},2)} = \sum_{i=1}^{n} \left[\hat{h}_{\mathrm{pv}}^{(1)}(i) + \hat{h}_{\mathrm{pv}}^{(2)}(i) \right]$$

Schrödinger picture

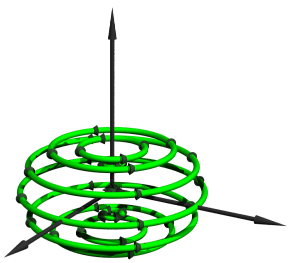
$$\hat{h}_{\mathrm{pv}}^{(2)} = \frac{G_{\mathrm{F}}}{4\sqrt{2} m_{\mathrm{e}} c} \sum_{A=1}^{N} K_{\mathrm{A},A} \left\{ \vec{\sigma} \cdot \vec{p}, \vec{\sigma} \cdot \vec{I} \rho_A(\vec{r}) \right\}_{+}$$

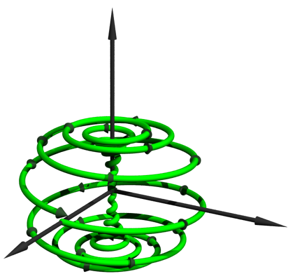
Dirac picture

$$\hat{h}_{\mathrm{pv}}^{(2)} = \frac{G_{\mathrm{F}}}{2\sqrt{2}} \sum_{A=1}^{N} K_{\mathrm{A},A}\, \vec{\alpha} \cdot \vec{I} \rho_A(\vec{r}); \quad \vec{\alpha} = \begin{bmatrix} \mathbf{0}_{2\times 2} & \vec{\sigma} \\ \vec{\sigma} & \mathbf{0}_{2\times 2} \end{bmatrix}$$

Gorshkov, Kozlov, Labzowsky, *Sov. Phys. JETP*, **1982**, *55*, 1042; Barra, Robert, Wiesenfeld, *Phys. Lett. A*, **1986**, *115*, 443; Wiesenfeld, *Mol. Phys.*, **1988**, *64*, 739

P-odd effects, 1c vs. 4c approaches

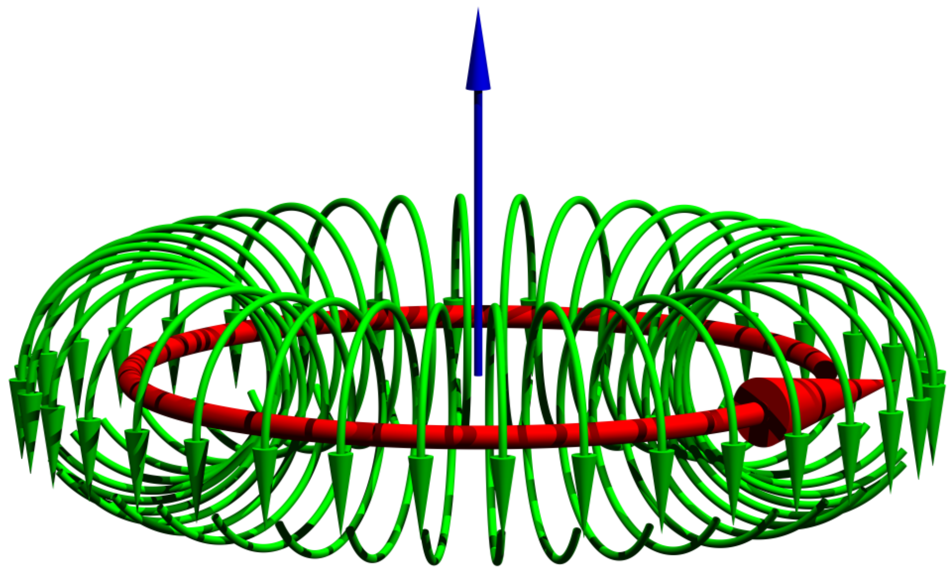




$K^{(2)}_{\mathrm{A},A}\,\vec{I}\rho_A(\vec{r})$

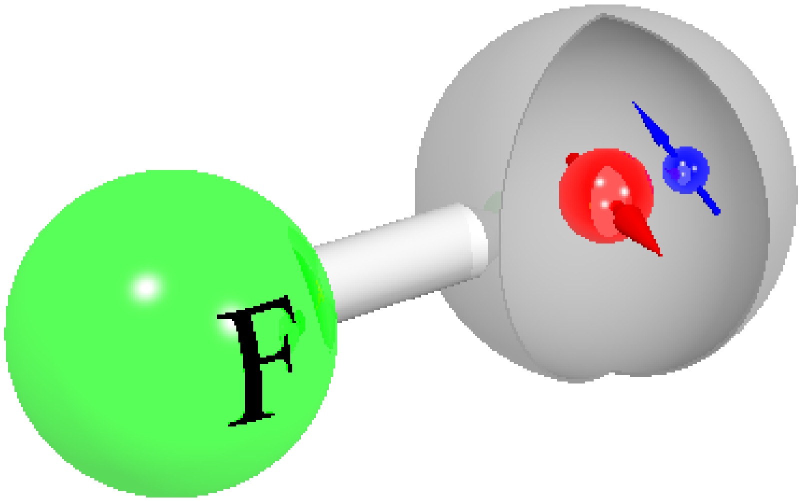
due to

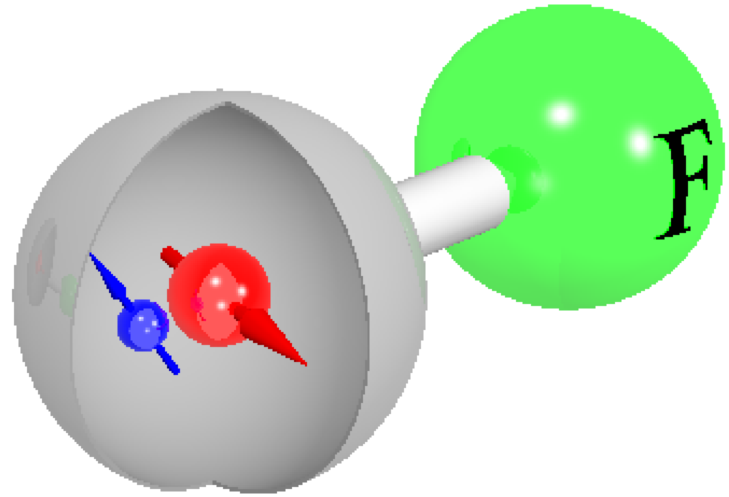
anapole moment



Berger in: Relativistic electronic structure theory, Part 2, (Ed: P. Schwerdtfeger), **2004**; Zel'dovich, *JETP*, **1957**, *33*, 1531; Flambaum, Khriplovich, *Phys. Lett. B*, **1984**, *146*, 367

P-odd and P,T-odd effect in diatomics



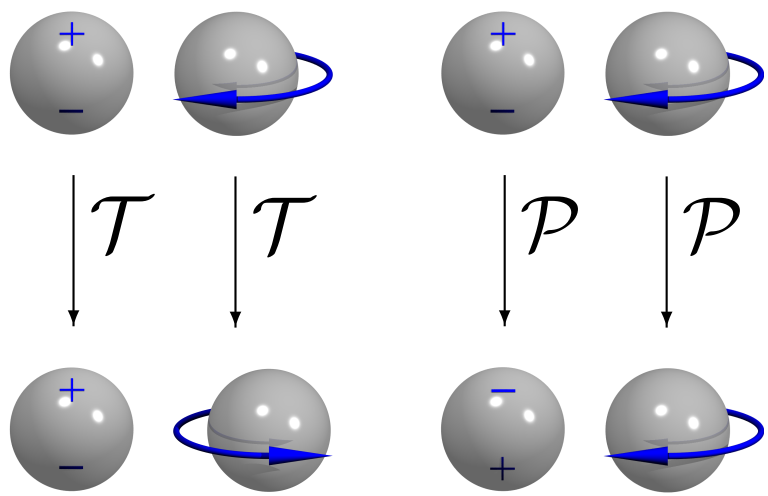


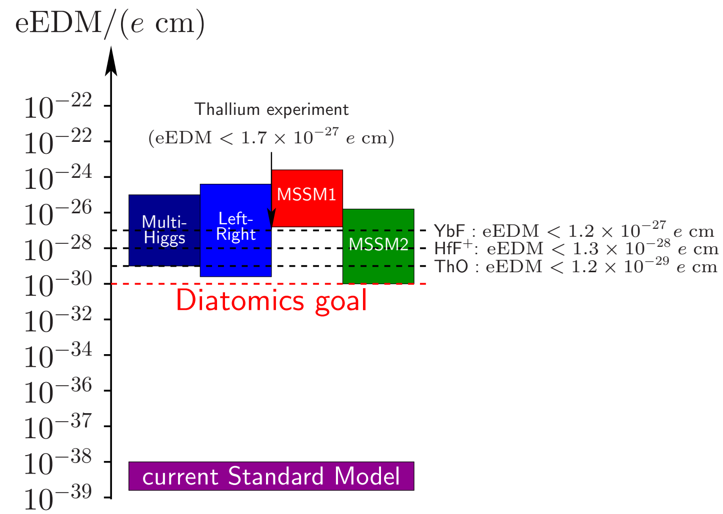
$$\widehat{H}_{\rm sr} = B\vec{\mathcal{N}}^2 + \gamma\,\vec{\mathbf{S}}^{\rm eff}\cdot\vec{\mathcal{N}} + \vec{\mathbf{S}}^{\rm eff}\cdot\widehat{\mathbf{A}}\cdot\vec{\mathbf{I}} + \vec{\mathcal{N}}\cdot\widehat{\mathbf{C}}\cdot\vec{\mathbf{I}}$$

$$+\, W_{\rm a}(K_A/2)[\vec{\lambda}\times\vec{\mathbf{S}}^{\rm eff}]\cdot\vec{\mathbf{I}} + (W_{\rm s}k_{\rm s} + W_{\rm d}d_{\rm e})\vec{\lambda}\cdot\vec{\mathbf{S}}^{\rm eff}$$

Review: Kozlov, Labzowsky, *J. Phys. B*, **1995**, *28*, 1933; Isaev, Hoekstra, Berger, *Phys. Rev. A*, **2010**, *82*, 052521; Isaev, Berger, *Phys. Rev. A.*, **2012**, *86*, 062515; Isaev, Berger, *J. Mol. Spectrosc.*, **2014**, *300*, 26–30; Isaev, Berger, arXiv:1302.5682; Kudashov, Petrov, Skripnikov, Mosyagin, Isaev, Berger, Titov, *Phys. Rev. A*, **2014**, *90*, 052513; Gaul, Berger, *J. Chem. Phys.*, **2017**, *117*, 014109; Gaul, Berger, *J. Chem. Phys.*, **2020**, *152*, 044101

Electron electric dipole moment





$\mathcal{P},\mathcal{T}$-violation: $\quad H_{\rm ptv} + H_{\rm ptc} = (d\vec{E} + m\vec{B}) \cdot \vec{F}/|\vec{F}|$

$\vec{E}$ strongly enhanced in polar molecules (YbF, ThO, RaF)

Regan, Commins, Schmidt, DeMille, *Phys. Rev. Lett.*, **2002**, *88*, 071805; Hudson, Sauer, Tarbutt, Hinds, *Phys. Rev. Lett.*, **2002**, *89*, 023003; Hudson, Kara, Smallman, Sauer, Tarbutt, Hinds, *Nature*, **2011**, *473*, 493; ACME Collaboration, *Science*, **2014**, *343*, 269; Cairncross, Gresh, Grau, Cossel, Roussy, Ni, Zhou, Ye, Cornell, *Phys. Rev. Lett.*, **2017**, *119*, 153001; ACME Collaboration, *Nature*, **2018**, *562*, 355 source (lower figure): Nature

P,T-odd effects, 4c approaches

$$\hat{h}_{\mathrm{s}}^{(1)} = \mathrm{i}\frac{G_{\mathrm{F}}}{\sqrt{2}}\sum_{A=1}^{N} k_{\mathrm{s},A} Z_A \boldsymbol{\gamma}_0 \boldsymbol{\gamma}_5 \rho_A(\vec{r}); \quad \gamma_0 = \begin{bmatrix} \mathbf{1}_{2\times 2} & \mathbf{0}_{2\times 2} \\ \mathbf{0}_{2\times 2} & -\mathbf{1}_{2\times 2} \end{bmatrix}$$

$$W_{\mathrm{s}} = \langle \hat{H}_{\mathrm{s}} \rangle / k_{\mathrm{s}}$$

$$\hat{h}_{\mathrm{e}} = -d_{\mathrm{e}}(\boldsymbol{\gamma}_0 - \mathbf{1}_{4\times 4})\vec{\Sigma} \cdot \vec{E}_{\mathrm{eff}}; \quad \vec{\Sigma} = \begin{bmatrix} \vec{\sigma} & \mathbf{0}_{2\times 2} \\ \mathbf{0}_{2\times 2} & \vec{\sigma} \end{bmatrix}$$

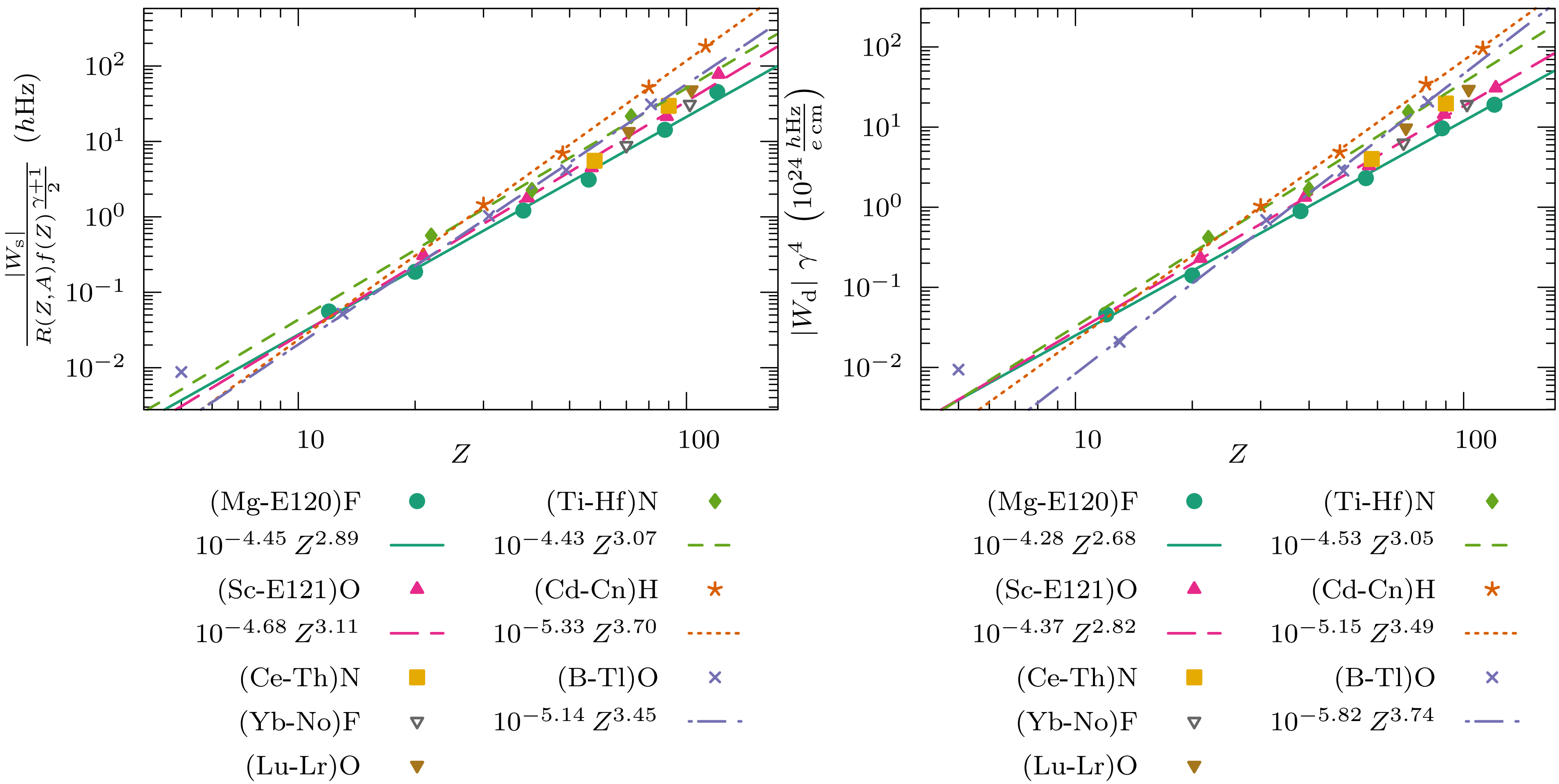
$$W_{\mathrm{d}} = \langle \hat{H}_{\mathrm{e}} \rangle / d_{\mathrm{e}} = -\vec{E}_{\mathrm{eff}} \cdot \vec{d}_{\mathrm{e}} / d_{\mathrm{e}}$$

W_{s} and E_{eff} can be estimated from W_{a}, e.g.:

$$W_{\mathrm{s}}/W_{\mathrm{a}} = Z3\gamma/(2\gamma+1); \gamma = \sqrt{1-(\alpha Z)^2}$$

Kozlov, *Sov. Phys. JETP*, **1985**, *62*, 1114; Dzuba, Flambaum, Harabati, *Phys. Rev. A*, **2011**, *84*, 052108

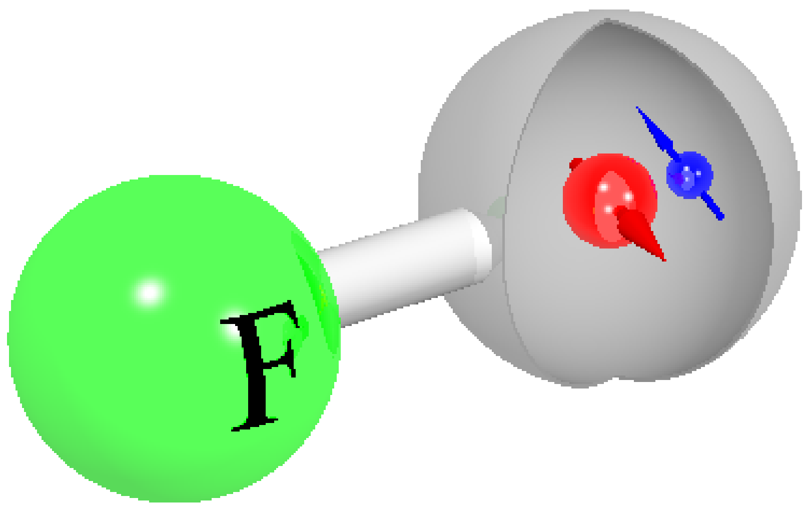
P,T-odd effect in diatomics

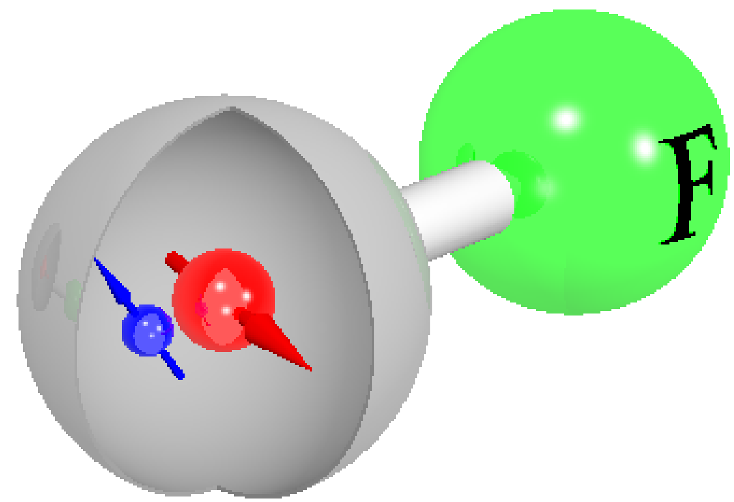


cGHF results

Gaul, Marquardt, Isaev, Berger, *Phys. Rev. A*, **2019**, *99*, 032509

P-odd and P,T-odd effect in diatomics



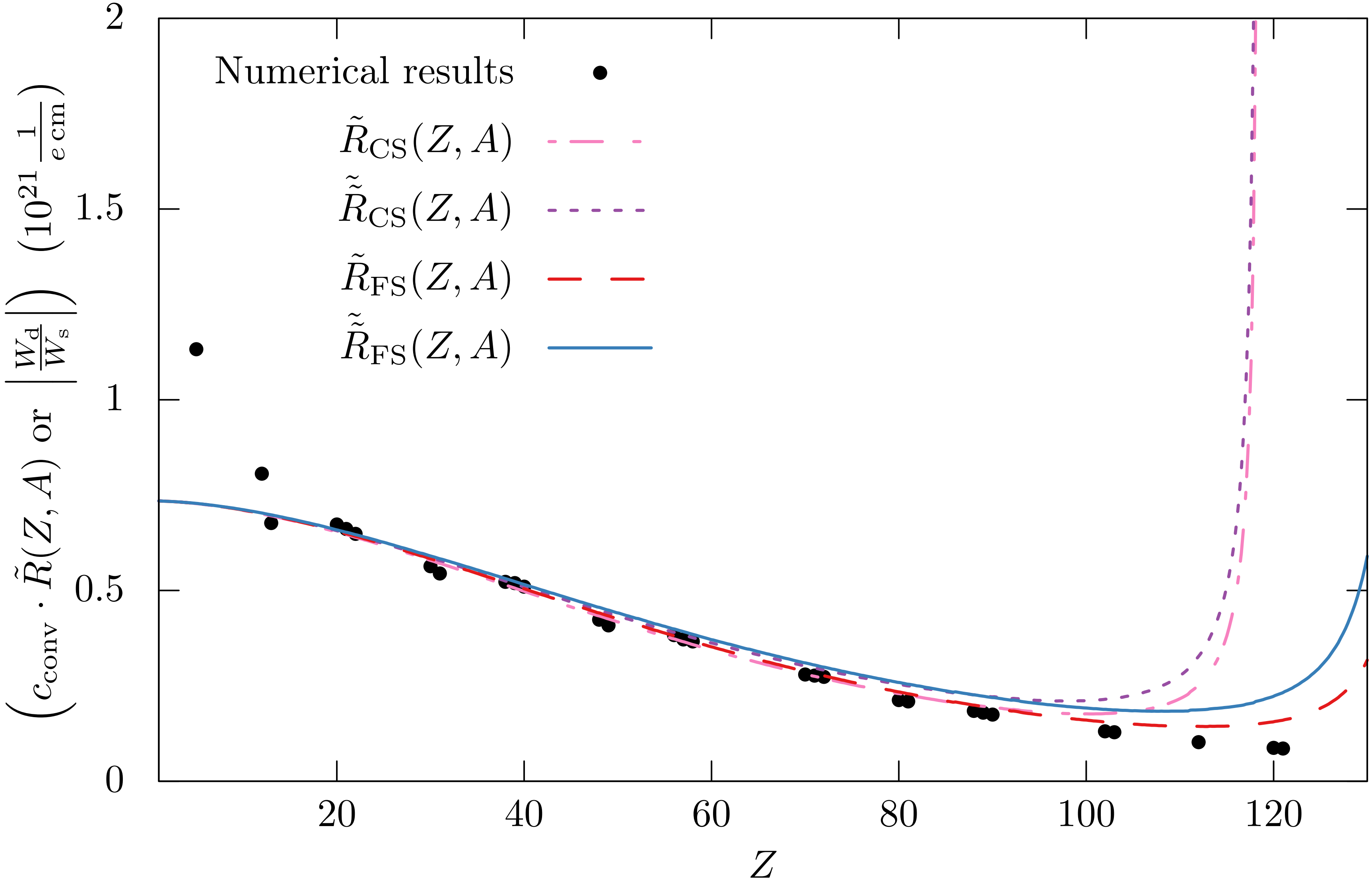


$$\widehat{H}_{\mathrm{sr}} = B\vec{\mathcal{N}}^2 + \gamma\,\vec{\mathbf{S}}^{\mathrm{eff}}\cdot\vec{\mathcal{N}} + \vec{\mathbf{S}}^{\mathrm{eff}}\cdot\widehat{\mathbf{A}}\cdot\vec{\mathbf{I}} + \vec{\mathcal{N}}\cdot\widehat{\mathbf{C}}\cdot\vec{\mathbf{I}}$$

$$+ W_{\mathrm{a}}(K_A/2)[\vec{\lambda}\times\vec{\mathbf{S}}^{\mathrm{eff}}]\cdot\vec{\mathbf{I}} + (W_{\mathrm{s}}k_{\mathrm{s}} + W_{\mathrm{d}}d_{\mathrm{e}})\vec{\lambda}\cdot\vec{\mathbf{S}^{\mathrm{eff}}}$$

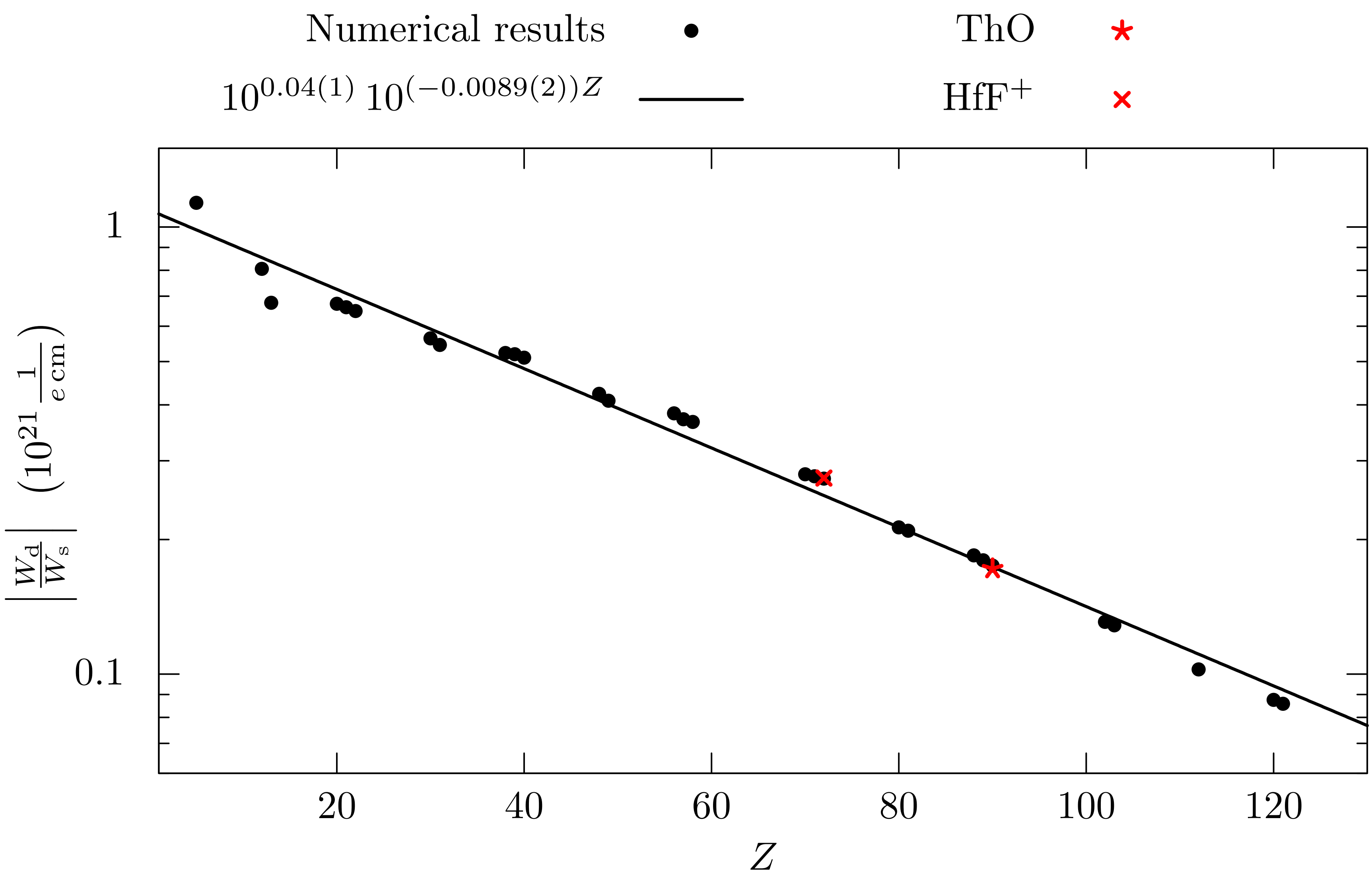
Review: Kozlov, Labzowsky, *J. Phys. B*, **1995**, *28*, 1933; Isaev, Hoekstra, Berger, *Phys. Rev. A*, **2010**, *82*, 052521; Isaev, Berger, *Phys. Rev. A.*, **2012**, *86*, 062515; Isaev, Berger, *J. Mol. Spectrosc.*, **2014**, *300*, 26–30; Isaev, Berger, arXiv:1302.5682; Kudashov, Petrov, Skripnikov, Mosyagin, Isaev, Berger, Titov, *Phys. Rev. A*, **2014**, *90*, 052513; Gaul, Berger, *J. Chem. Phys.*, **2017**, *117*, 014109; Gaul, Berger, *J. Chem. Phys.*, **2020**, *152*, 044101

P,T-odd effect in diatomics



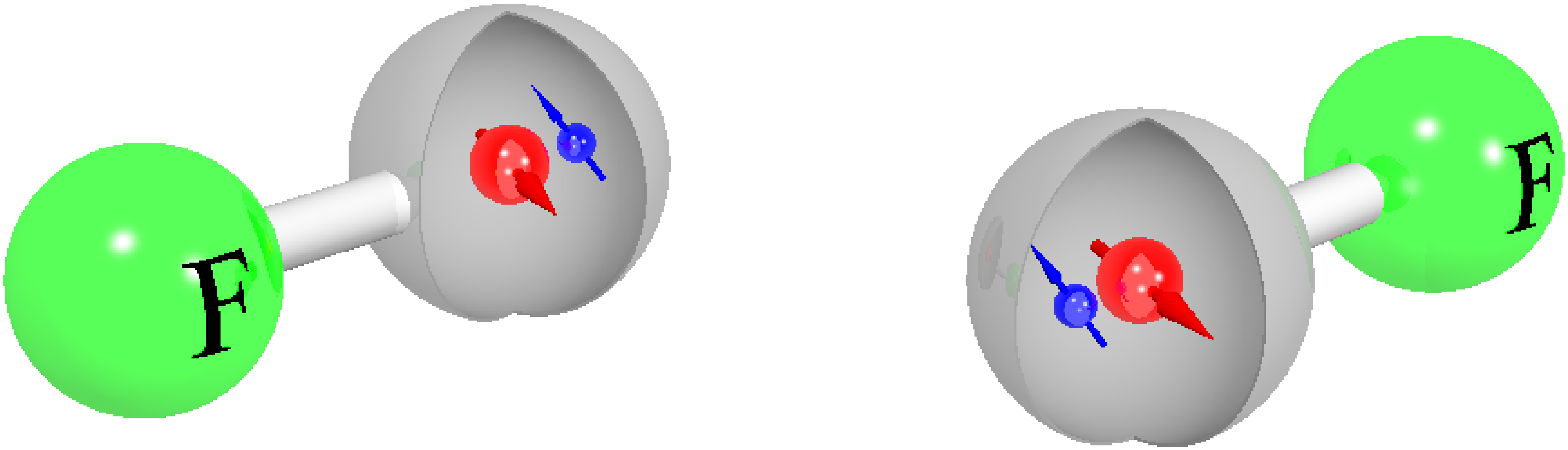
Gaul, Marquardt, Isaev, Berger, *Phys. Rev. A*, **2019**, *99*, 032509

P,T-odd effect in diatomics



Gaul, Marquardt, Isaev, Berger, *Phys. Rev. A*, **2019**, *99*, 032509; ThO, HfF^{+}: Fleig, *Phys. Rev. A*, **2017**, *96*, 040502

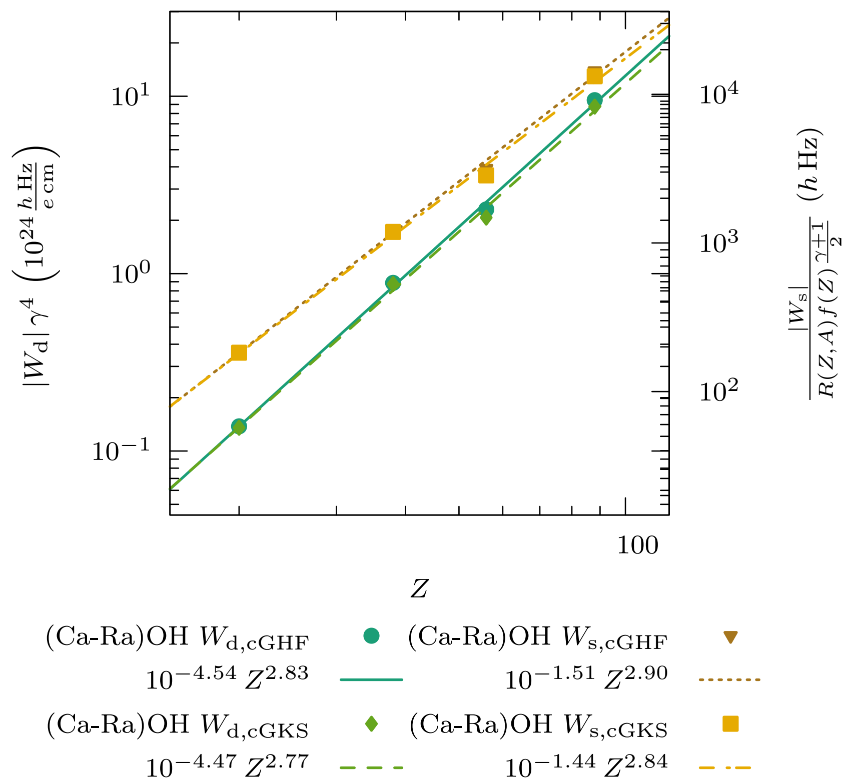
P-odd and P,T-odd effect in diatomics

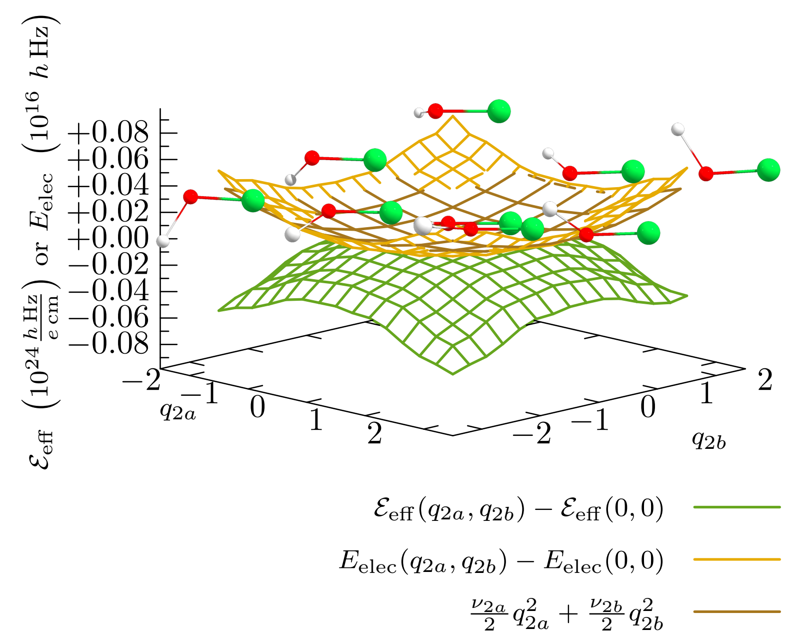


$$\widehat{H}_{\mathrm{sr}} = B\vec{\mathcal{N}}^2 + \gamma\,\vec{\mathbf{S}}^{\mathrm{eff}}\cdot\vec{\mathcal{N}} + \vec{\mathbf{S}}^{\mathrm{eff}}\cdot\widehat{\mathbf{A}}\cdot\vec{\mathbf{I}} + \vec{\mathcal{N}}\cdot\widehat{\mathbf{C}}\cdot\vec{\mathbf{I}}$$
$$+ W_{\mathrm{a}}(K_A/2)[\vec{\lambda}\times\vec{\mathbf{S}}^{\mathrm{eff}}]\cdot\vec{\mathbf{I}} + W_{\mathrm{d}}(k_{\mathrm{s}}W_{\mathrm{s}}/W_{\mathrm{d}} + d_{\mathrm{e}})\vec{\lambda}\cdot\vec{\mathbf{S}}^{\mathrm{eff}}$$

Gaul, Marquardt, Isaev, Berger, *Phys. Rev. A*, **2019**, *99*, 032509

$\color{blue}{\text{P,T-odd}}$ effect in laser-coolable triatomics

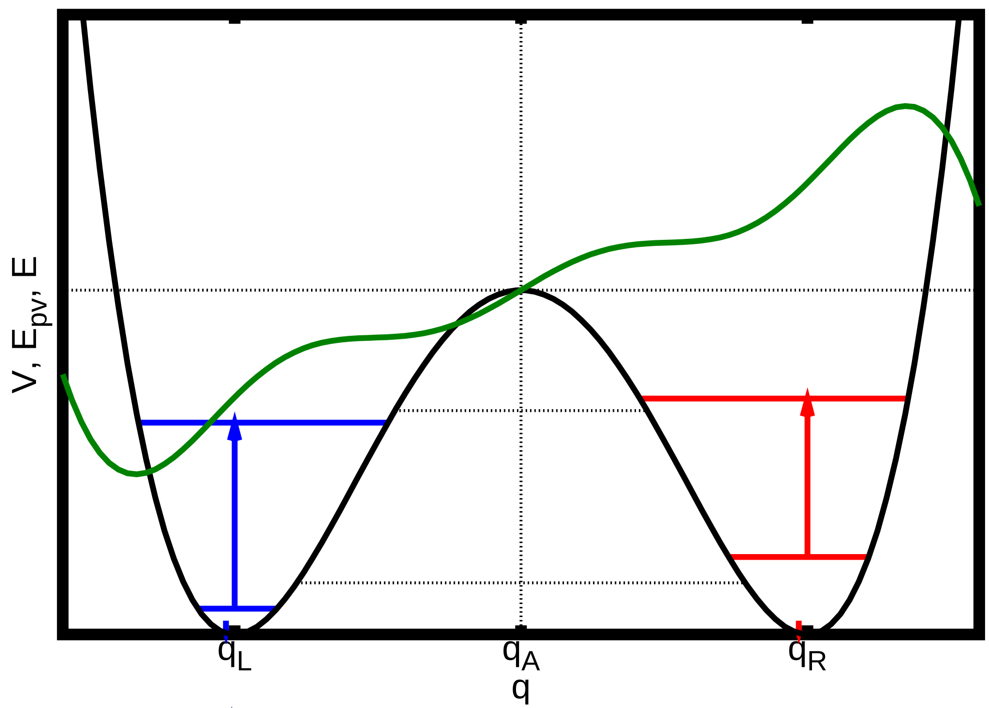




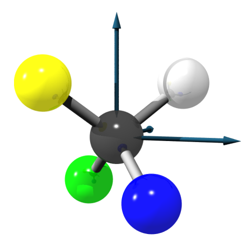
Very similar behaviour for MF and MOH

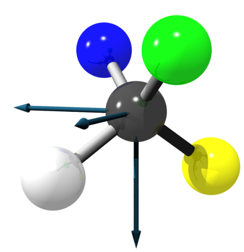
Gaul, Berger, *arXiv:1811.05749*; Gaul, Berger, *Phys. Rev. A*, **2020**, *101*, 012508

Detection of molecular parity violation



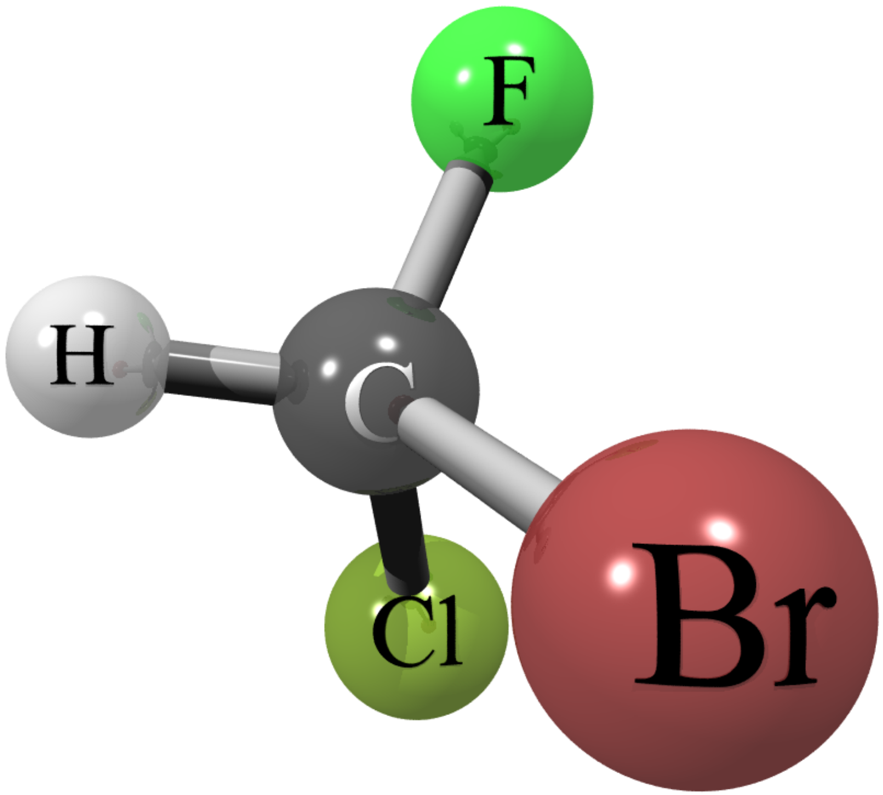
$$\Delta\nu_{pv} = \nu_L - \nu_R$$





Letokhov, *Phys. Lett. A*, **1975**, *53*, 275; Kompanets, Kukudzhanov, Letokhov, Gervits, *Opt. Commun*, **1976**, *19*, 414; Arimondo, Glorieux, Oka, *Opt. Commun.*, **1977**, *23*, 369; Berger, Quack, Sieben, Willeke, *Helv. Chim. Acta*, **2003**, *86*, 4048; Berger, Laubender, Quack, Sieben, Stohner, Willeke, *Angew. Chem. Int. Ed.*, **2005**, *44*, 3623;

Experimental vibrational frequency shifts

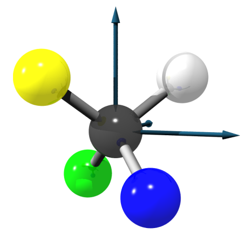


Experimental upper bound for C–F stretch fundamental:

$$\Delta\nu_{\mathrm{pv}}/\nu < 10^{-13}$$

Daussy, Marrel, Amy-Klein, Nguyen, Bordé, Chardonnet, *Phys. Rev. Lett.*, **1999**, *83*, 1554; Ziskind, Daussy, Marrel, Chardonnet, *Eur. Phys. J. D*, **2002**, *20*, 219

Theoretical vibrational frequency shifts



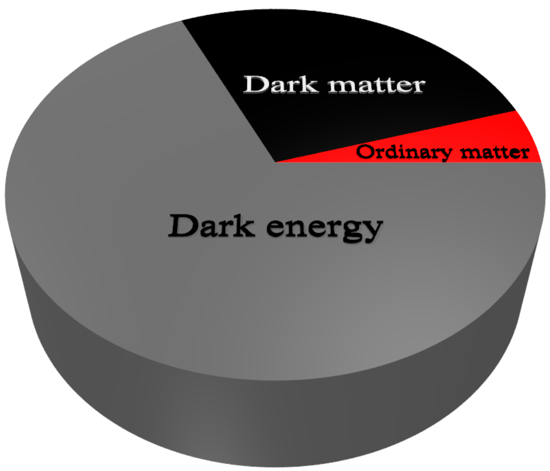
	$E_{pv}/(hc\ 10^{-12}\,\mathrm{cm}^{-1})$			$\Delta\nu_{pv}/(\nu\,10^{-13})$
Molecule	ZORA (2c)	DHFC(4c)[a]	BP(1c)[b]	ZORA(2c)
(S)-CHBrClF	-1.5	-1.2	-1.47	$+0.0007$
(S)-CHBrClI	-24.7			
(S)-CHBrFI	-38.5	-37.1		$+0.0178$
(S)-CHClFI	-13.7	-13.0		$+0.0094$
(S)-CBrClFI	$+5.1$	$+4.4$		
(S)-CHAtFI	$+2314$			-1.072

a) Schwerdtfeger, Laerdahl, Chardonnet, *Phys. Rev. A*, **2002**, *65*, 042508

b) with atomic scaling factors, unscaled value: -0.8; Berger, *J. Chem. Phys.* **2008**, *129*, 154105

Berger, Stuber, *Mol. Phys.*, **2007**, *105*, 41

Dark matter search with chiral molecules



- P-odd interaction with pseudo-scalar cosmic field

$$\hat{H}_{\mathrm{a,PS1}} = \eta\hbar\omega_{\mathrm{a}}\sin(\omega_{\mathrm{a}}t)\gamma^5$$

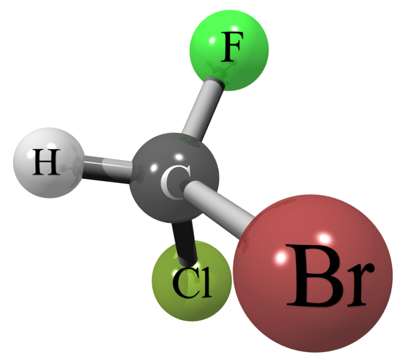
- P-odd interaction with pseudo-vector cosmic field

$$\hat{H}_{\mathrm{pv,cosmic}} = b_0^{\mathrm{e}}(t)\gamma^5$$

$\langle\gamma^5\rangle$ depends on electron helicity in chiral molecules

Gaul, Isaev, Kozlov, Berger, *Phys. Rev. Lett.*, **2020**, *125*, 123004; *Phys. Rev. A*, **2020**, *102*, 032816;
Atoms: Roberts, Stadnik, Dzuba, Flambaum, Leefer, Budker, *Phys. Rev. D*, **2014**, *90*, 096005

Dark matter search with chiral molecules



Experimental upper bound for C–F stretch fundamental:

$$|\Delta\nu| = 9.4 \pm 5.1 \pm 12.7\ \text{Hz};\ \Delta\nu_{\text{pv}}/\nu < 10^{-13}$$

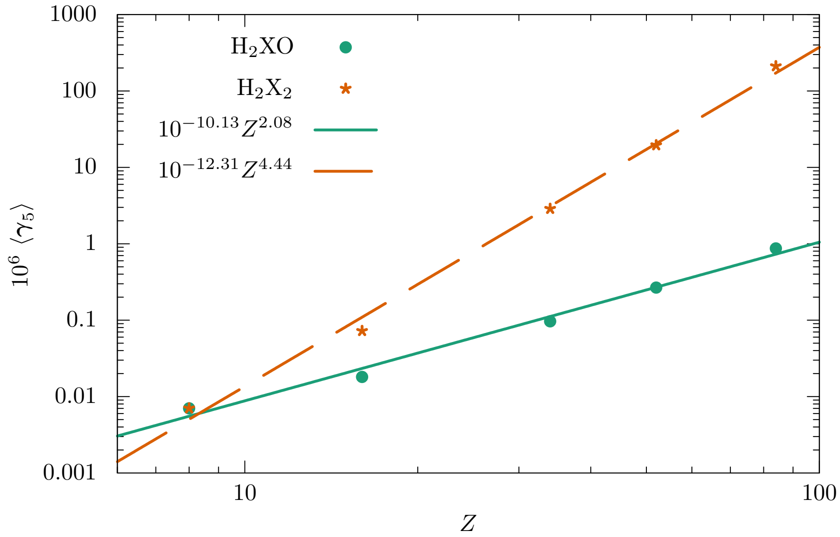
Theoretical analysis:

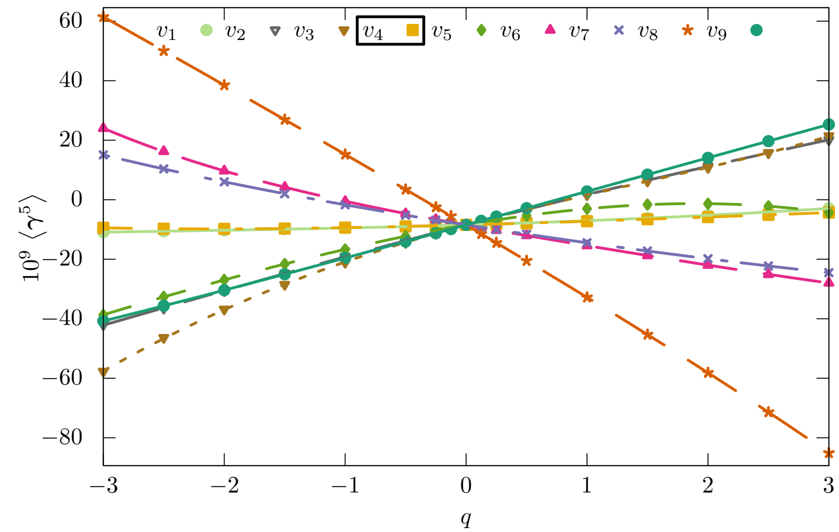
$$\Delta_{(R,S)}\langle\gamma^5\rangle \approx 7.4 \times 10^{-10}$$

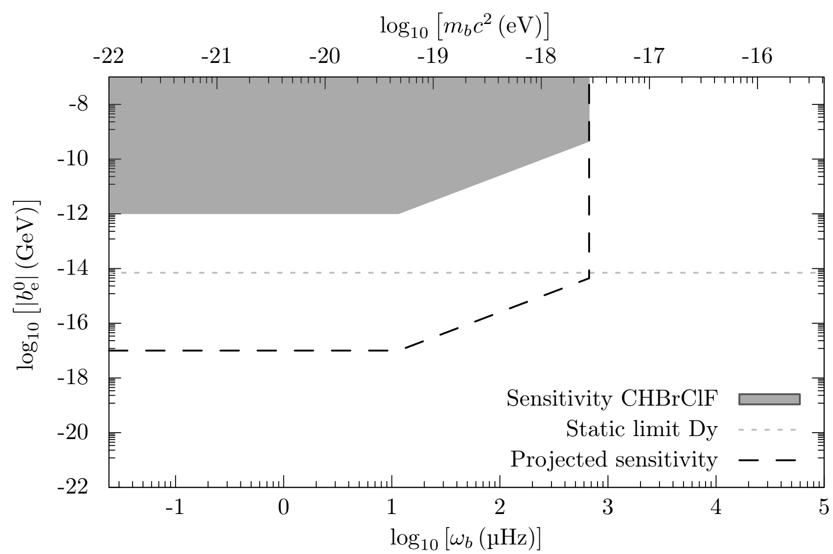
$$|b_0^{\text{e}}| \lesssim \left|\frac{12.7\ \text{Hz}}{\mathcal{O}(10^{-10})}h\right| \sim \mathcal{O}(10^{-12}\ \text{GeV})$$

Gaul, Isaev, Kozlov, Berger, *Phys. Rev. Lett.*, **2020**, *125*, 123004; *Phys. Rev. A*, **2020**, *102*, 032816;
Atoms: Roberts, Stadnik, Dzuba, Flambaum, Leefer, Budker, *Phys. Rev. D*, **2014**, *90*, 096005

Dark matter search with chiral molecules







Gaul, Isaev, Kozlov, Berger, *Phys. Rev. Lett.*, **2020**, *125*, 123004; *Phys. Rev. A*, **2020**, *102*, 032816;
Atoms: Roberts, Stadnik, Dzuba, Flambaum, Leefer, Budker, *Phys. Rev. D*, **2014**, *90*, 096005

Conclusions

- Test of BSM physics with (polyatomic) molecules
- Precision spectroscopy on heavy, radioactive molecules offers ample of opportunities
- Both simple models and accurate predictions play important role for preparation of experiments

Conclusions (short version)

Let the molecule do the job!

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