Interpretation of the parity and time invariance violation in atoms and molecules

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Interpretation of PNC and EDM measurements in atoms and molecules relies on sophisticated calculations of electron structure

- APV

$$E_{PV}(1 \to 2) = \sum_n \left[\frac{\langle 2 | H_{PV} | n \rangle \langle n | D | 1 \rangle}{E_2 - E_n} + \frac{\langle 2 | D | nP \rangle \langle n | H_{PV} | 1 \rangle}{E_1 - E_n} \right] = \xi Q_W$$

- Atomic EDM

$$d_{atom}(1) = 2 \sum_N \frac{\langle 1 | D_z | N \rangle \langle N | H_{PT} | 1 \rangle}{E_1 - E_N} = \xi S$$

ζ and ξ are electron structure factors,
Q_W is weak nuclear charge, S is the Schiff moment.

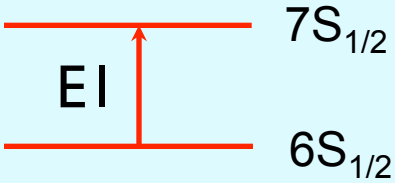
H_{PV} is due to electron-nucleon P-odd interactions and nuclear anapole,
H_{PT} is due to nucleon-nucleon, electron-nucleon PT-odd interactions, electron, proton or neutron EDM.

Atomic wave functions need to be good at *all* distances!

PNC in Cs

- Best measurement for cesium [Boulder 1997]

$$-\mathrm{Im}(E_{PV})/\beta = 1.5935(1 \pm 0.35\%)\mathrm{mV/cm}$$



- Atomic theory required for determination of Q_W

$$E_{PV}(6s \rightarrow 7s) = \sum_n \left[\frac{\langle 7s | H_{PV} | nP \rangle \langle nP | D | 6s \rangle}{E_{7s} - E_{nP}} + \frac{\langle 7s | D | nP \rangle \langle nP | H_{PV} | 6s \rangle}{E_{6s} - E_{nP}} \right] = \zeta Q_W$$

Atomic theory	$\delta E_{PV}/E_{PV}$	Q_W-Q_W^{SM}	Ref.
1% calculations		1.2σ	Dzuba ,Flambaum,Sushkov 1989; Blundell,Johnson,Sapirstein 1990
Reinterpretation 1% to 0.4%		2.5σ	Bennett & Wieman '99
Breit interaction	-0.6%		Derevianko '00
Vacuum polarization	+0.4%		Johnson et al. '01; Milstein & Sushkov '02
Neutron distribution	-0.2%		Derevianko '02
0.5% calculations		2.1σ	Dzuba, Flambaum, Ginges ' 02
Self-energy and vertex radiative corrections	-0.8%		Kuchiev & Flambaum '02; Milstein et al. '02; Sapirstein et al. '03; Shabaev et al. '05; Flambaum & Ginges '05
Final		1.5σ	2013

Ab initio methods of atomic calculations

$$d_{atom}(1) = 2\sum_N \frac{\langle 1 | D_z | N \rangle \langle N | H_{PT} | 1 \rangle}{E_1 - E_N} = \xi S$$

N_{ve}	Method	Accuracy	Examples
0	Rel. Hartree-Fock+RPA	~ 10%	Xe EDM
1	RHF+MBPT All-orders sums	0.1-1%	Cs, Fr, Ba^+, Ra^+ PNC
2-8	RHF+MBPT+CI	1-10%	Yb, Hg, Tl PNC and EDM
2-15	Configuration interaction	10-20%	Dy PNC

N_{ve} *- number of valence electrons*

These methods cover all periodic table of elements

Mono-valence atoms

(Cs, Ba^+, Fr, Ra^+, etc.)

- *Direct summation* approach (Johnson, Derevianko, Safronova)
- *Solving equations* approach (used in our group):

$$E_{PNC} = \left\langle \delta\psi_{6s}^{Br} \,|\, D \,|\, \psi_{7s}^{Br} \right\rangle + \left\langle \psi_{6s}^{Br} \,|\, D \,|\, \delta\psi_{7s}^{Br} \right\rangle + \left\langle \psi_{6s}^{Br} \,|\, \delta V_{DW} \,|\, \psi_{7s}^{Br} \right\rangle$$

$$\left(H_0 - E_v + \Sigma\right)\psi_v^{Br} = 0$$ - Eq. for Brueckner orbitals

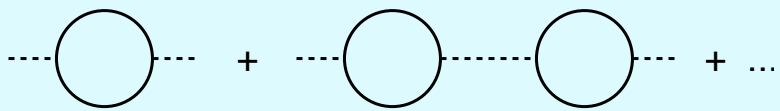
$$\left(H_0 - E_v + \Sigma\right)\delta\psi_v^{Br} = -(F + \delta V_F)\psi_v^{Br}$$ - **RPA** equations

F is weak or E1 or both

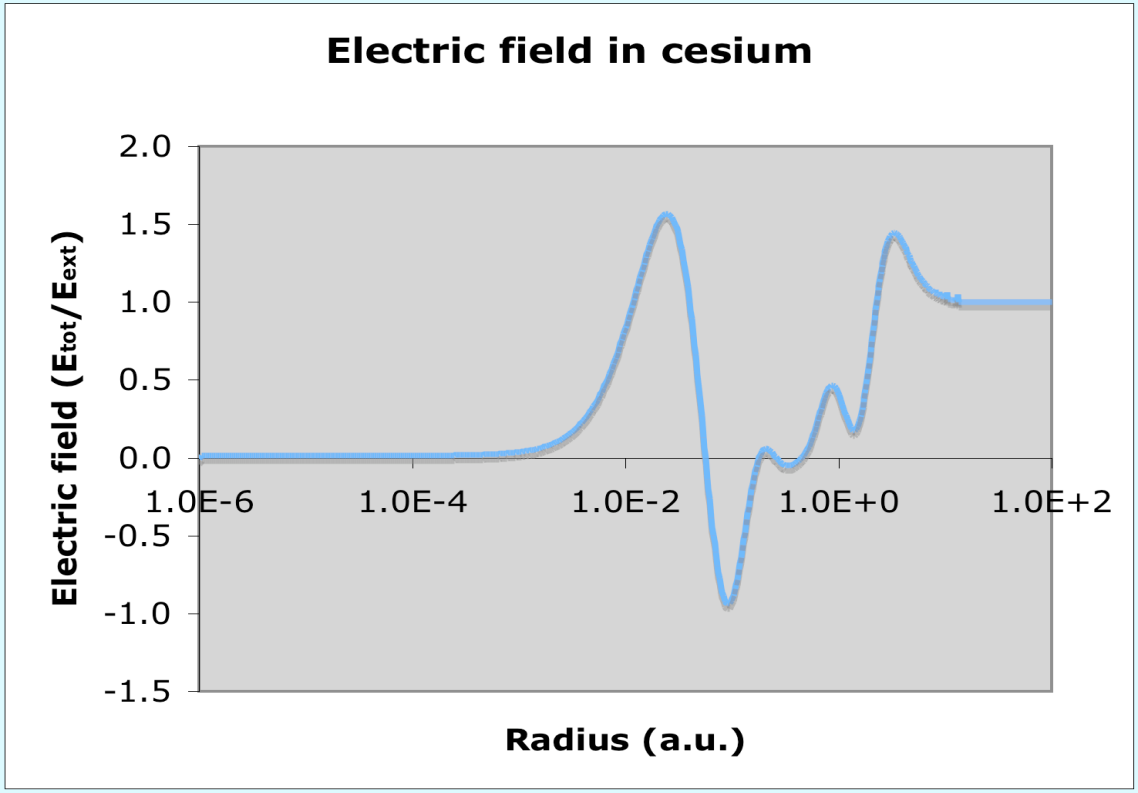
Σ is the correlation potential

Σ includes infinite chains of dominating higher-order diagrams:

- Screening of Coulomb interaction



= QPQ + QPQPQ + …



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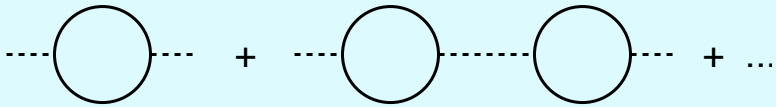
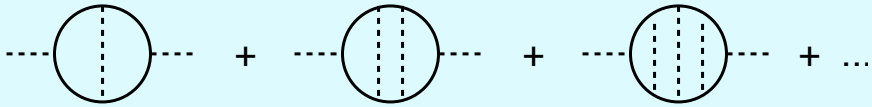
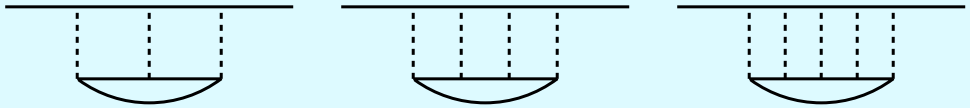


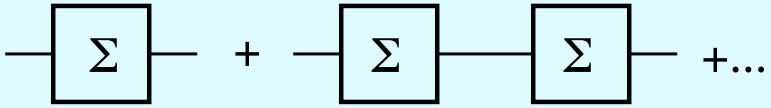
- Hole-particle interaction



- Ladder diagrams



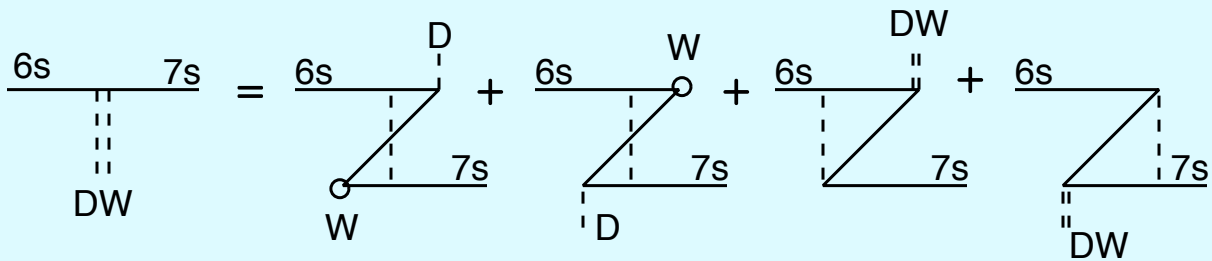
- Iterations of **Σ** are included by solving Eq. for BO



Accuracy for Cs, Ba^+, Fr, Ra^+, etc.:
~ 0.1% for EL, ~ 0.5% for matrix elements

$$E_{PNC} = \left\langle \delta\psi_{6s}^{Br} \,|\, D \,|\, \psi_{7s}^{Br} \right\rangle + \left\langle \psi_{6s}^{Br} \,|\, D \,|\, \delta\psi_{7s}^{Br} \right\rangle + \left\langle \psi_{6s}^{Br} \,|\, \delta V_{DW} \,|\, \psi_{7s}^{Br} \right\rangle$$

Last term (double core polarization)



is often missed in the direct summation approach

Breit interaction

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

includes magnetic interaction and retardation.
Total potential is the sum of Coulomb and Breit parts:

$$V = V_C + V_B$$

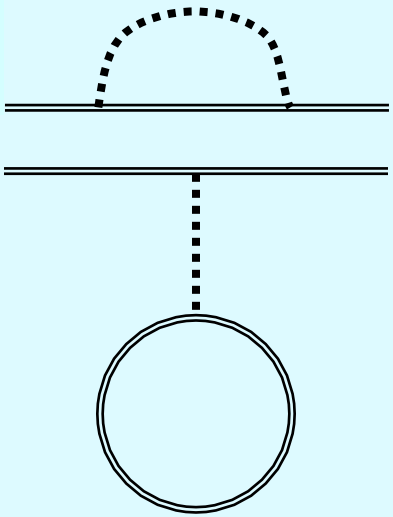
Breit interaction is included in all-orders in Coulomb interaction

Radiative potential for QED

(Flambaum and Ginges , PRA 72, 052115 (2005))

(for energies and E1 amplitudes)

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



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

$\Phi_f(r)$ and $\Phi_f(r)$ have free parameters which are chosen to fit QED corrections to the energies (Mohr, et al) and weak matrix elements (Kuchiev,Flambaum; Milstein,Sushkov,Terekhov; Sapirstein et al)

Direct summation

$$E_{PV}(1 \to 2) = \sum_n \left[\frac{\langle 2 | H_{PV} | n \rangle \langle n | D | 1 \rangle}{E_2 - E_n} + \frac{\langle 2 | D | nP \rangle \langle n | H_{PV} | 1 \rangle}{E_1 - E_n} \right] = \zeta Q_W$$

All matrix elements and energies are calculated independently on each other others using e.g. CC (SD or SDvT) approach.

Advantage: easy to compare with experiment m.e. and energies for dominating terms.

Shortcoming: high accuracy for dominating terms does not guaranty high accuracy for the total amplitude.

Results for cesium

Expt.: $-\mathrm{Im}(E_{PV})/\beta = 1.5935(1 \pm 0.35\%)\mathrm{mV/cm}$ Boulder, 1997

	Epv	Source
	0.908(9)	Dzuba, Flambaum, Sushkov (1989)
	0.909(9)	Blundell, Johnson, Sapirstein (1990)
	0.905(9)	Kozlov, Porsev, Tupitsin (2001)
	0.9078(45)	Dzuba, Flambaum, Ginges (2002)
A	0.8998(24)	Porsev, Derevianko, Beloy (2009)
B	0.9079(40)	Dzuba, Berengut, Flambaum, Roberts (2012)

	A	B	B - A
Core	-0.0020	0.0018	0.0038
Main	0.8823	0.8678	N/A
Tail	0.0195	0.0238	0.0043
	Total correction		0.0081
Finaly, E_{PNC} = 0.8998 + 0.0081 = 0.9079			

Results for cesium

Expt.: $-\mathrm{Im}(E_{PV})/\beta = 1.5935(1 \pm 0.35\%)\mathrm{mV/cm}$ Boulder, 1997

Theory:

Epv	Source
0.908(9)	Dzuba, Flambaum, Sushkov (1989)
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0.9078(45)	Dzuba, Flambaum, Ginges (2002)
0.8906(24)	Porsev, Derevianko, Beloy (2009)
0.9079(40)	Dzuba, Berengut, Flambaum, Roberts (2012) (terms missed in Porsev et al are added)

Q_W-Q_W^{SM}=0.65(43) (1.5σ)

Isospin conserving parameter S=-0.81(54)

For extra Z_x-boson: M_{Zx} > 710 GeV/c2

PNC in analogs of Cs
(Ba^+, Fr, Ra^+, Ac^{2+},Th^{3+})

Advantages:

- PNC effect in Fr, Ra^+, Ac^{2+}, Th^{3+} is 15 times lager than in Cs.
- Correlations are relatively smaller in ions.
- Accuracy for s-d transitions might be better.

$$E_{PV}(6d \rightarrow 7s) = \sum_n \left[\frac{\langle 7s | H_{PV} | np \rangle \langle np | D | 6d \rangle}{E_{7s} - E_{np}} + \frac{\langle 7s | D | np \rangle \langle np | H_{PV} | 6d \rangle}{E_{6d} - E_{np}} \right]$$

this term dominates

Calculations are done in our and other groups.

Experiments:

Seattle (Ba^+), TRIUMF (Fr), Groningen (Ra^+).

PNC in a chain of isotopes

$$\frac{E_{PNC}^{N_1}}{E_{PNC}^{N_2}} = \frac{K_{PNC} Q_W^{N_1}}{K_{PNC} Q_W^{N_2}} = \frac{Q_W^{N_1}}{Q_W^{N_2}}$$

Dzuba, Flambaum, Khriplovich 1986

Rare-earth atoms:

- close opposite parity levels-enhancement
- Many stable isotopes

Fortson,Pang,Wilets 1990

There is a neutron distribution problem.

Test of Standard model or neutron distribution?

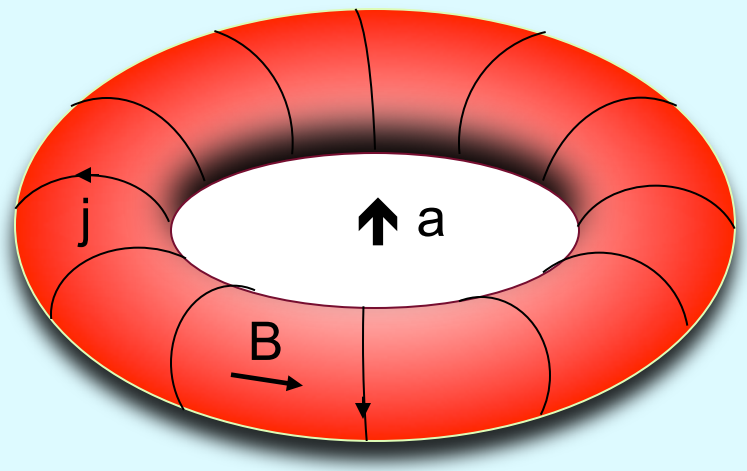
Brown, Derevianko,Flambaum 2009.

Uncertainties in neutron distributions cancel in differences of PV effects in isotopes of the same element.

Measurements of ratios of PNC effects in isotopic chain can compete with other tests of Standard model!

Nuclear anapole moment

- Source of nuclear spin-dependent PNC effects in atoms
- Nuclear magnetic multipole violating parity
- Arises due to parity violation inside the nucleus



- Interacts with atomic electrons via usual magnetic interaction (PNC hyperfine interaction):

$$h_a = e\vec{\alpha}\cdot\vec{A} \propto \kappa_a \vec{\alpha}\cdot\vec{I}\,\rho(r)\ , \quad \kappa_a \propto A^{2/3}$$

[Flambaum,Khriplovich,Sushkov]

$E_{PV} \propto Z^2 A^{2/3}$ measured as difference of PV effects for transitions between hyperfine components

Cs: |6s,F=3> – |7s,F '=4> and |6s,F'=4> – |7s,F=3>

Nuclear anapole moment is produced by PNC nuclear forces. Measurements + calculations give the strength constant g.

Experiment:

- Boulder, Cs: g_p=6(1) G_F
- Seattle, Tl: g_p=-2(3) G_F

Nuclear calculations:

- Flambaum,Hanhart, 1993;
- Dmitriev, Khriplovich,Telitsin, 1994;
- Auerbach, Brown, 1999;
- Haxton,Liu,Ramsey-Musolf 2001-2002;

problem remains.

New measurements are highly disable.

Experiments and proposals

- Dy, Yb, Berkeley.
- Hg, Xe, Berkeley, Crete.
- Yb^+, Los Alamos.
- Ba^+, Seattle.
- Ra^+, Groningen.
- Fr, Rb, TRIUMF

Most of these experiments consider both *isotope chain* and *anapole moment* measurements.

Enhancement of nuclear anapole effects in molecules

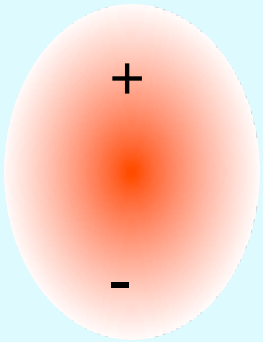
10^5 enhancement of the anapole contribution in diatomic molecules due to mixing of close rotational levels of opposite parity. Theorem: only nuclear-spin-dependent (anapole) contribution to PV is enhanced (Labzovsky;Sushkov,Flambaum 1978). Weak charge can not mix opposite parity rotational levels and Λ−doublet.

Ω=1/2 terms: $\Sigma_{1/2}$, $\Pi_{1/2}$. Heavy molecules, effect $Z^2 A^{2/3} R(Z\alpha)$
YbF,BaF, PbF,LuS,LuO,LaS,LaO,HgF,…Cl,Br,I,…BiO,BiS,…

Cancellation between hyperfine and rotational intervals-enhancement.
Interval between the opposite parity levels may be reduced to zero by magnetic field – further enhancement.

Molecular experiments : Yale, Groningen, NWU.
New calculations for many molecules and molecular ions: Borschevsky,Ilias,Beloy,Dzuba,Flambaum,Schwerdtfeger 2012

Atomic electric dipole moments



- Electric dipole moments violate parity (P) and time-reversal (T)

$$\vec{d} \equiv \vec{r} \propto \vec{J}$$

- T-violation ≡ CP-violation by CPT theorem

CP violation

- Observed in K^0, B^0
- Accommodated in SM as a single phase in the quark-mixing matrix (Kobayashi-Maskawa mechanism)

However, not enough CP-violation in SM to generate enough matter-antimatter asymmetry of Universe!

➔ Must be some non-SM CP-violation

- Excellent way to search for new sources of CP-violation is by measuring EDMs
  - SM EDMs are hugely suppressed
  - → Theories that go beyond the SM predict EDMs that are many orders of magnitude larger!

e.g. electron EDM

Theory	d_e (*e* cm)
Std. Mdl.	$< 10^{-38}$
SUSY	10^{-28} - 10^{-26}
Multi-Higgs	10^{-28} - 10^{-26}
Left-right	10^{-28} - 10^{-26}

Best limit (90% c.l.): $|d_e| < 1.6 \times 10^{-27}$ *e cm* Berkeley (2002)

- Atomic EDMs $d_{atom} \propto Z^3$ [Sandars]

Sensitive probe of physics beyond the Standard Model!

Atomic EDMs

Best limits

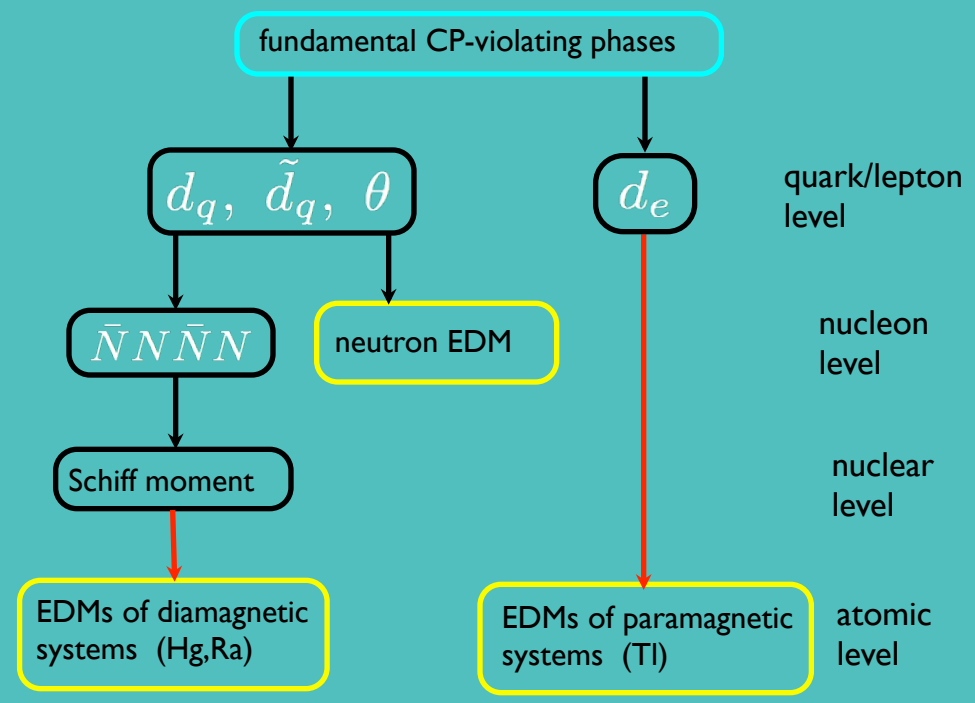
$|d(^{199}\mathrm{Hg})| < 3 \times 10^{-29}\ e$ cm
(95% c.l., Seattle, 2009)

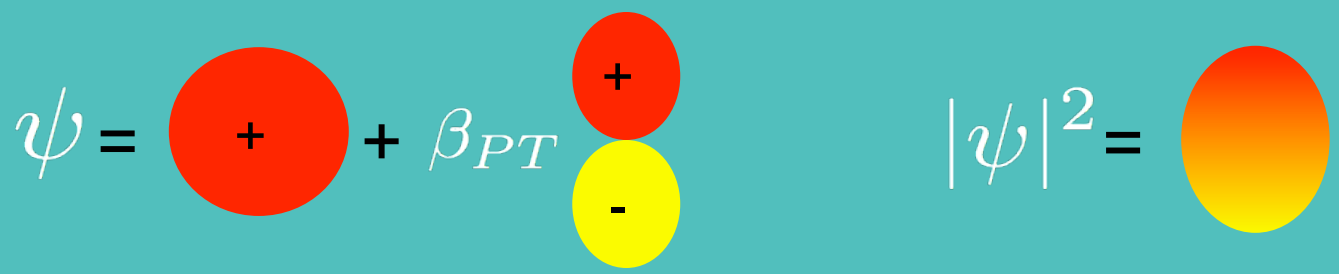
$|d(^{205}\mathrm{Tl})| < 9.6 \times 10^{-25}\ e$ cm
(90% c.l., Berkeley, 2002)

$|d(\mathrm{n})| < 2.9 \times 10^{-26}\ e$ cm
(90% c.l., Grenoble, 2006)

$|d_e| < 10.5 \times 10^{-28}\ e$ cm
(YbF, 90% c.l., London, 2011)

Leading mechanisms for EDM generation





EDM of Tl

d(Tl) = Rd_e < 9.4×10^{-25} e cm (Berkeley, 2002)
Assuming R=-585 (Liu and Kelly, 1992)
lead to d_e < 1.6×10^{-27} e cm

There is strong 6s-6p interaction in Tl.

Ground state is $6s^2 6p$
State $6s6p^2$ gives ~ 50% of EDM.

This means that Tl should be treated as a triple-valence system.

CI+MBPT calculations

$$H^{eff}\Psi = E\Psi \qquad \Psi(r_1,\ldots,r_n) = \sum_{i=1}^{n} c_i \Phi(r_1,\ldots,r_n)$$

$$H^{eff} = \sum_{i=1}^{n} h_1(r_i) + \sum_{i<j} h_2(r_i, r_j)$$

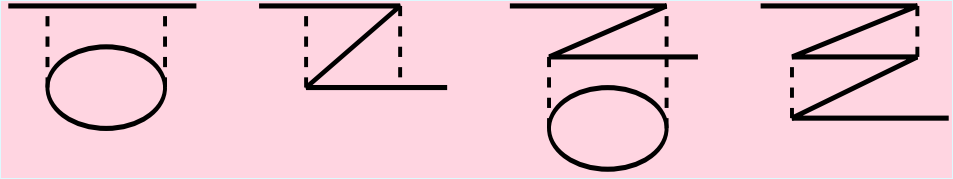
$$h_1(r) = c\alpha p + (\beta - 1)mc^2 - \frac{Ze^2}{r} + V_{core}(r) + \Sigma_1$$

$$h_2(r_1, r_2) = \frac{e^2}{|r_1 - r_2|} + \Sigma_2$$

Σ is the core-valence correlation operator

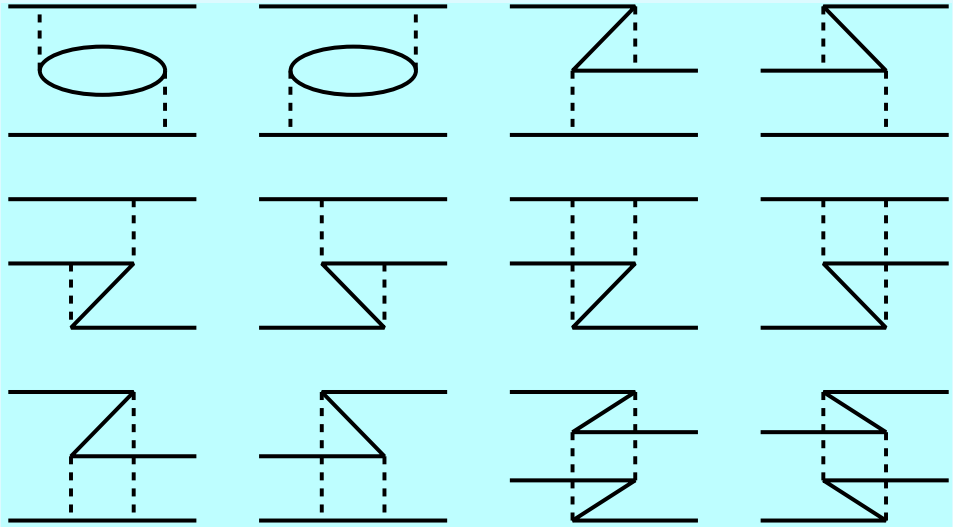
Σ in second-order MBPT:

Σ_1



(Correlation interaction between a valence electron and the core)

Σ_2



(Screening of Coulomb interaction between valence electrons by core electrons)

Results for the EDM enhancement factor R for Tl

R	Authors	Year	Method	Nv
-585	Liu and Kelly	1992	CC	1
-582(20)	Dzuba and Flambaum	2009	CI+MBPT	3
-466(10)	Nataraj et al	2011	CC	1
-573(20)	Porsev, Safronova and Kozlov	2012	CI+MBPT, CI+SD	3

From **Tl** EDM measuraments:

$d_e < 1.6 \times 10^{-27}$ e cm (Berkeley, 2002)

From **YbF** EDM measurements:

$d_e < 1.05 \times 10^{-27}$ e cm (London, 2011)

Schiff moment

Nuclear T,P-odd moments:

- EDM – non-observable due to total screening (Schiff theorem)
- Schiff moment appears when screening of external electric field by atomic electrons is taken into account.

Schiff moment is a source of **EDM**s of *diamagnetic* atoms and molecules

Nuclear electrostatic potential with screening

$$\varphi(\mathbf{R}) = \int \frac{e\rho(\mathbf{r})}{|\mathbf{R}-\mathbf{r}|} d^3r + \frac{1}{Z}(\mathbf{d}\bullet\nabla)\int \frac{\rho(\mathbf{r})}{|\mathbf{R}-\mathbf{r}|} d^3r$$

d is nuclear EDM, the term with **d** is the electron screening term

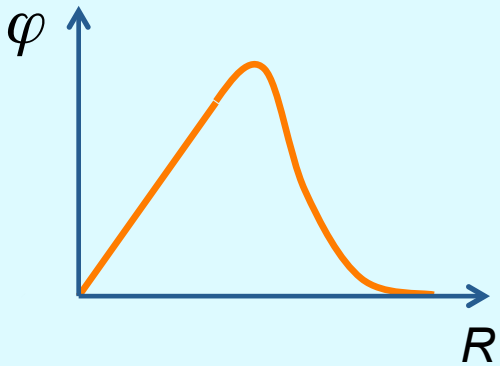
$\varphi(\mathbf{R})$ in multipole expansion is reduced to $\varphi(\mathbf{R}) = 4\pi\mathbf{S}\bullet\nabla\delta(\mathbf{R})$

where $\mathbf{S} = \frac{e}{10}\left[\langle r^2\mathbf{r}\rangle - \frac{5}{3Z}\langle r^2\rangle\langle\mathbf{r}\rangle\right]$ is Schiff moment

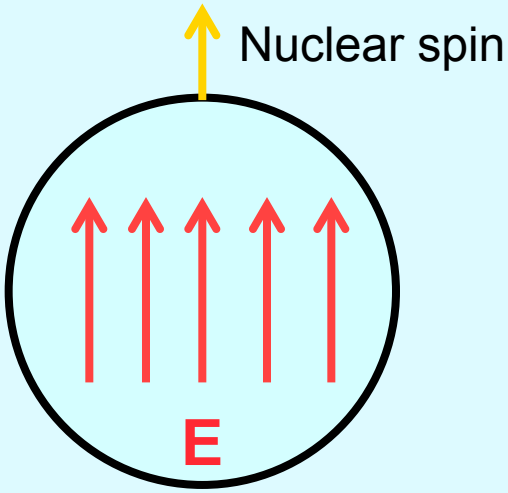
This expression is not suitable for relativistic calculations.

Flambaum,Ginges:
$L=S(1 - c\, Z^2\, \alpha^2)$

$$\phi(\mathbf{R}) = -\frac{3L \bullet \mathbf{R}}{B}\rho(R)$$ where $$B = \int \rho(R) R^4 dR$$



Electric field induced by T,P-odd nuclear forces which influence proton charge density



This potential has no singularities and may be used in relativistic calculations.
SM electric field polarizes atom and produces EDM.

Calculations of nuclear SM: Sushkov,Flambaum,Khriplovich ;Brown et al,Flambaum et al Dmitriev et al,Auerbach et al,Engel et al, Liu et al,Sen'kov et al, Ban et al.

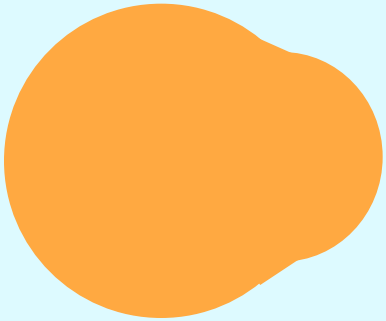
Atomic EDM: Sushkov,Flambaum,Khriplovich; Dzuba,Flambaum,Ginges,Kozlov.

Best limits from **Hg EDM** measurement in Seattle –
Crucial test of modern theories of CP violation (supersymmetry, etc.)

Nuclear enhancement

Auerbach, Flambaum, Spevak 1996

The strongest enhancement is due to octupole deformation (Rn,Ra,Fr,…)

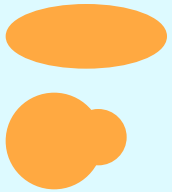


Intrinsic Schiff moment:

$$S_{\text{intr}} \approx eZR_N^3 \frac{9\beta_2\beta_3}{20\pi\sqrt{35}}$$

$\beta_2 \approx 0.2$ - quadrupole deformation

$\beta_3 \approx 0.1$ - octupole deformation



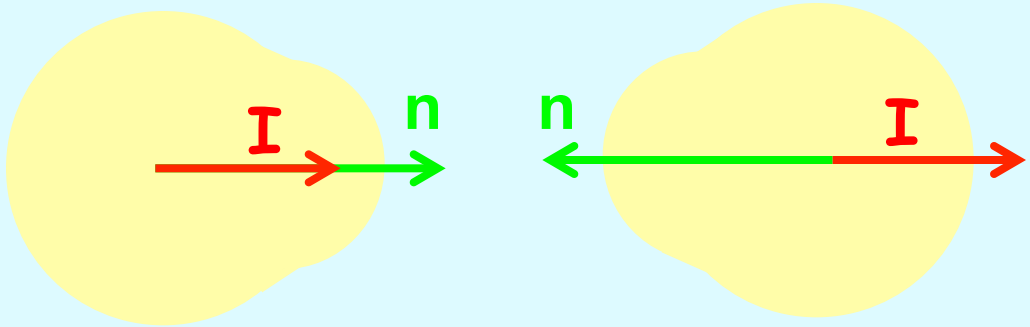
No T,P-odd forces are needed for the Schiff moment and EDM in intrinsic reference frame

However, in laboratory frame S=d=0 due to rotation

In the absence of T,P-odd forces: doublet (+) and (-)

$$\Psi = \frac{1}{\sqrt{2}}\left(\left|IMK\right\rangle + \left|IM-K\right\rangle\right)$$

and $\langle \mathbf{n} \rangle = 0$



$K = (\mathbf{I} \cdot \mathbf{n})$

T,P-odd mixing (β) with opposite parity state (-) of doublet:

$$\Psi = \frac{1}{\sqrt{2}}\left[(1+\beta)\left|IMK\right\rangle + (1-\beta)\left|IM-K\right\rangle\right]$$

and $\langle \mathbf{n} \rangle \propto \beta \mathbf{I}$

EDM and Schiff moment

$$\langle d \rangle, \langle \mathbf{S} \rangle \propto \langle \mathbf{n} \rangle \propto \beta \mathbf{I}$$

Simple estimate (Auerbach, Flambaum, Spevak):

$$S_{lab} \propto \frac{\langle + | H_{TP} | - \rangle}{E_+ - E_-} S_{body}$$

Two factors of enhancement:

1. Large collective moment in the body frame
2. Small energy interval (E_+-E_-), 0.05 instead of 8 MeV

$$S \approx 0.05 e \beta_2 \beta_3^2 Z A^{2/3} \eta r_0^3 \frac{\text{eV}}{E_+ - E_-} \approx 700 \times 10^{-8} \eta e \text{fm}^3 \approx 500 S(\text{Hg})$$

^{225}Ra,^{223}Rn, Fr,… -100-1000 times enhancemnt

Engel, Friar, Hayes (2000); Flambaum, Zelevinsky (2003):
Static octupole deformation is not essential, nuclei with soft octupole vibrations also have the enhancement.

Nature 2013 Experiment : Octupole deformation in ^{224}Ra,^{220}Rn,

EDMs of atoms of experimental interest

Z	Atom	[S/(e fm^3)]e cm	[10^{-25} η] e cm	Expt.
2	^{3}He	0.00008	0.0005	
54	^{129}Xe	0.38	0.7	Seattle, Ann Arbor, Princeton
70	^{171}Yb	-1.9	3	Bangalore,Kyoto
80	^{199}Hg	-2.8	4	Seattle
86	^{223}Rn	3.3	3300	TRIUMF
88	^{225}Ra	-8.2	2500	Argonne,KVI
88	^{223}Ra	-8.2	3400	

Standard Model $\eta = 0.3\ 10^{-8}$ $d_n = 5 \times 10^{-24}$ *e* cm η, $d(^{199}\text{Hg})/\ d_n = 10^{-1}$

Extra enhancement in excited states: Ra

$$d_{atom}(1) = 2\sum_N \frac{\langle 1|D_z|N\rangle\langle N|H_{PT}|1\rangle}{E_1 - E_N}$$

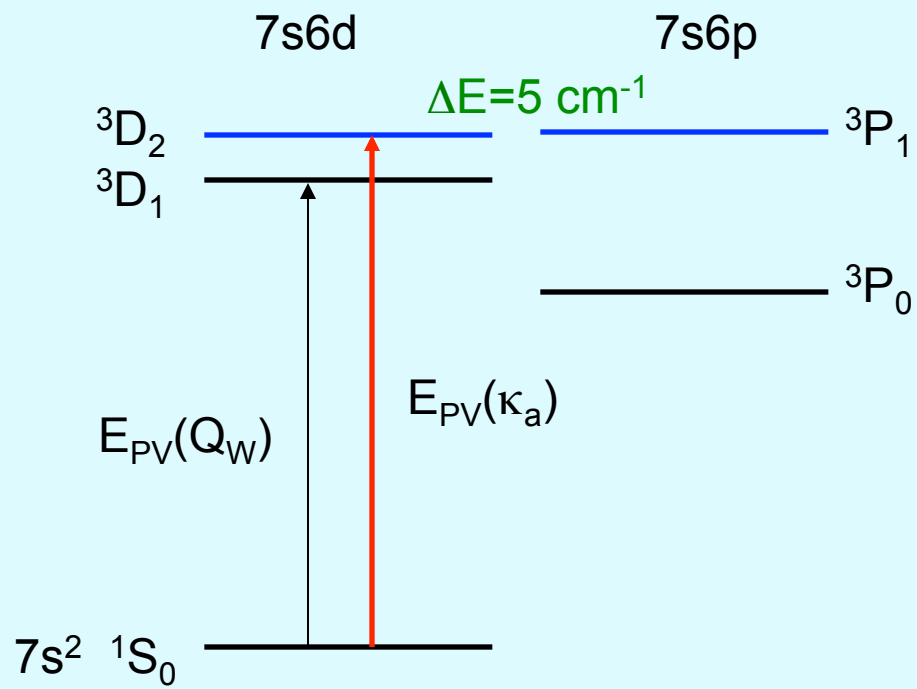
- Extra enhancement for EDM and APV in metastable states due to presence of close opposite parity levels

  [Flambaum; Dzuba,Flambaum,Ginges]

$d(^3D_2) \sim 10^5 \times d(Hg)$

$E_{PV}(^1S_0\text{-}^3D_{1,2}) \sim 100 \times E_{PV}(Cs)$

Comparison of even Ra isotopes



Good to study anapole moment:

- Strongly enhanced ($E_{PV} \sim 10^3\ E_{PV}$ (Cs))
- Q_W does not contribute ($\Delta J = 1$)
- PV in optical or microwave transition

Summary

- Strong constrains on new physics are found from Cs PNC, Tl EDM, Hg EDM, YbF EDM measurements.
- Interpretation of the measurements are based on sophisticated atomic calculations.
- Promising future directions:

  - ✓ Q_W measurements for Fr and Fr-like ions.
  - ✓ *Isotope chain* and *anapole moment* measurements for atoms and ions.
  - ✓ *Anapole moment* measurements for molecules.
  - ✓ **EDM** measurements for atoms and molecules.