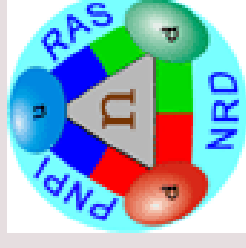


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Enhancement of the electron EDM in atoms, molecules, and crystals

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- Electric dipole moment of the electron (eEDM)
- Atomic enhancement of eEDM
- Additional enhancement of eEDM in diatomic radicals
- Magnetization in electric field
- Reliability of atomic and molecular enhancement factors

Current limit on eEDM

Atomic beam experiment on Tl by Commins' group in Berkeley gave the following upper limit for EDM of Tl atom [Regan et al (2002)]:

$$|d_{\text{Tl}}| < 9 \times 10^{-25} \text{ e cm.}$$

Atomic theory allows to link d_{Tl} with eEDM d_e :

$$d_{\text{Tl}} = -590 \times d_e.$$

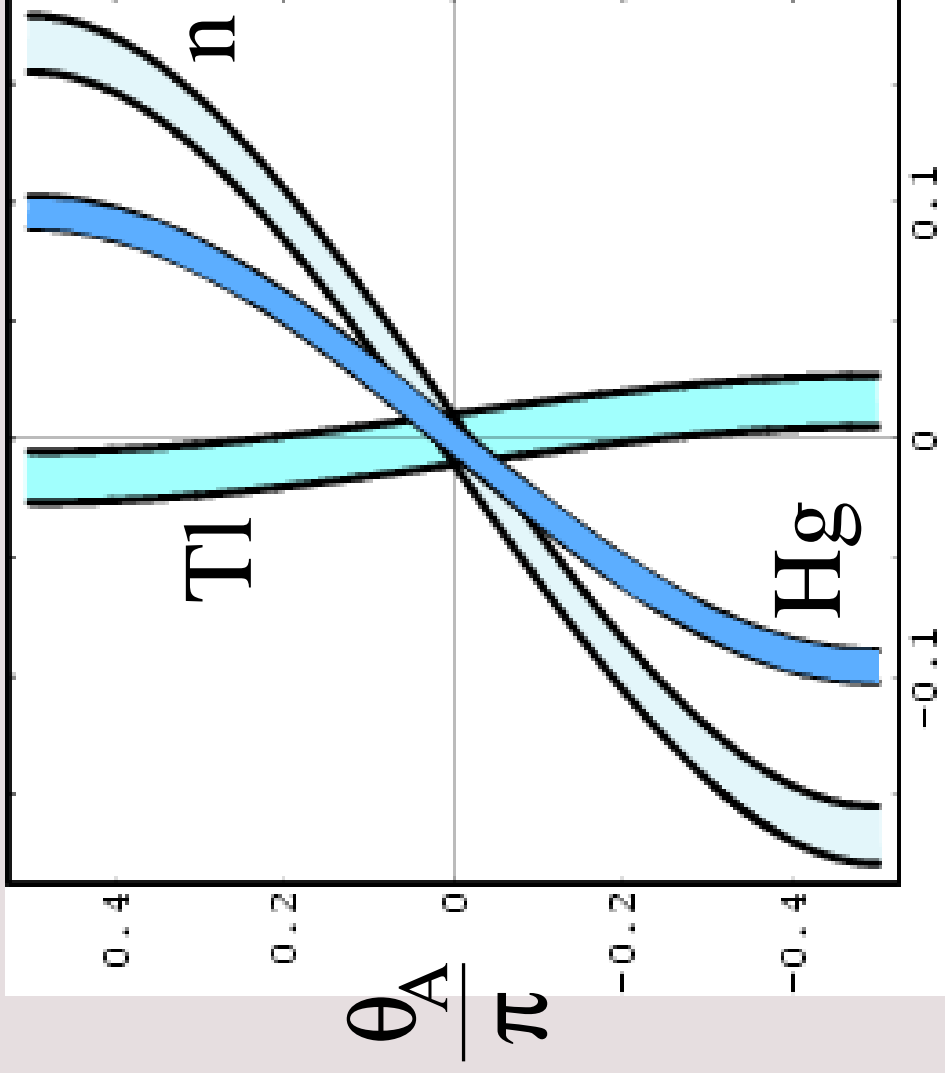
That leads to the upper bound to d_e :

$$|d_e| < 1.6 \times 10^{-27} \text{ e cm.}$$

Experimental limits on the CP-odd phases in the minimal SUSY model from EDM experiments on Tl, Hg, and n.

Other parameters of the model are chosen to be:

$$M_{\text{SUSY}} = 500 \text{ GeV},$$
$$\tan \beta = 3.$$



$$\frac{\theta_\mu}{\pi}$$

Atomic enhancement of the eEDM

Non-relativistic consideration of the electron in the stationary state immediately leads to the zero energy shift:

$$\delta\varepsilon = -\vec{d}_e \cdot \langle \vec{E} \rangle; \quad m\vec{a} = -e\vec{E}; \quad \langle \vec{a} \rangle = 0; \quad \Rightarrow \quad \langle \vec{E} \rangle = 0.$$

However, in the electron rest frame there is nonzero magnetic field and the force $\vec{F} \neq -e\vec{E}$. Therefore, generally $\langle \vec{E} \rangle \neq 0$.

Relativistic Hamiltonian for the Dirac electron has the form:

$$H_d = -d_e \begin{pmatrix} \vec{\sigma} & 0 \\ 0 & -\vec{\sigma} \end{pmatrix} \cdot \vec{\nabla} \phi \blacktriangleright \blacktriangleright \blacktriangleright H_d = 2d_e \begin{pmatrix} 0 & 0 \\ 0 & \vec{\sigma} \end{pmatrix} \cdot \vec{\nabla} \phi,$$

where ϕ is atomic potential. In the external electric field \vec{E} the energy correction will be:

$$\delta\varepsilon = 2 \frac{\langle 0 | H_d | n \rangle \langle n | e\vec{r} \cdot \vec{E} | 0 \rangle}{\varepsilon_0 - \varepsilon_n}.$$

Estimate of atomic energy shift due to eEDM (Sandars 1965)

In atomic units ($\varepsilon_0 - \varepsilon_n$) $\sim \langle n|r|0\rangle \sim 1$ and

$$\delta\varepsilon \sim \langle H_d \rangle E = 2d_e E \left\langle \begin{pmatrix} 0 & 0 \\ 0 & \vec{\sigma} \end{pmatrix} \cdot \vec{\nabla} \phi \right\rangle.$$

At large distances $\phi \sim 1$, but for $r \sim \frac{1}{Z}$, $\phi \sim \frac{Z}{r}$ and $\nabla\phi \sim \frac{Z}{r^2} \sim Z^3$. Also, at large distances the small components of the Dirac orbitals $\sim \alpha$, while at short distances they are $\sim \alpha Z \times \psi(0) \sim \alpha Z^3/2$. As a result, the above matrix element is given by the integral over the volume $V \sim Z^{-3}$:

$$\delta\varepsilon \equiv d_{\text{at}} E \sim d_e E (\alpha Z)^2 \psi^2(0) \left(\nabla\phi|_{r \sim 1/Z} V \right) \sim \alpha^2 Z^3 d_e E.$$

We see that $d_{\text{at}} \equiv k_{\text{at}} d_e \sim \alpha^2 Z^3 d_e$ and atomic enhancement factor

$$k_{\text{at}} \sim \alpha^2 Z^3.$$

Calculations of atomic enhancement factors

The estimate $k_{\text{at}} \sim \alpha^2 Z^3$ obtained by Sandars showed that heavy atoms could work as amplifiers of eEDM. That was later confirmed by atomic calculations. For example, for Cs ($Z=55$), $k_{\text{Cs}} \approx 120$ and for Tl ($Z=81$) $k_{\text{Tl}} \approx -590$.

Atomic enhancement factors appear to be very sensitive to electron correlations. In particular, the Dirac-Fock calculation for Tl gives $k_{\text{Tl}} = -1910$.

Core-valence correlations usually increase the answer by 10 – 30%, while valence-valence correlations can work both ways.

Note that k_{at} depends on $|\psi_{\text{val}}(0)|^2$ and, therefore, becomes small when the angular momentum of the valence electron $j > \frac{1}{2}$. For this case correlations can even change the sign and the order of magnitude of the effect.

Summary for atoms

- Atomic EDM scales as $10\alpha^2 Z^3$ when valence electron has $j = \frac{1}{2}$ and is much smaller otherwise.
- The sign of d_{at} depends on the valence configuration.
- Atomic enhancement factor k_{at} is very sensitive to electron correlations, in particular for the case of $j > \frac{1}{2}$.
- Modern atomic theory allows reliable calculations of k_{at} for most heavy atoms of interest.

eEDM in diatomic molecules

- Internal electric field in the polar molecule $E_{\text{mol}} \sim \frac{e}{R_0^2} \sim 10^9 \text{ V/cm}$, which is 4 – 5 orders of magnitude larger than typical laboratory field in EDM experiment. This field is directed along the molecular axis and is averaged by rotation of the molecule.
- In the external electric field E_{lab} molecule axis is polarized in the direction of the field. For $\Sigma_{1/2}$ -molecule in the rotational level J the complete polarization requires the field $D_{\text{mol}}E_{\text{lab}} \approx 2BJ$. For molecules with Ω -doubling a much smaller field may be sufficient.
- Typical molecular enhancement factor $k_{\text{mol}} \sim k_{\text{at}} \times \frac{E_{\text{mol}}}{E_{\text{lab}}} \sim 10^4 k_{\text{at}}$.

Molecules with large enhancement

- The molecule must have non-zero electronic angular momentum \Rightarrow unpaired electron(s). Thus, it can be either a molecule in excited state [PbO in $a(1)$ state], or a radical [PbF, YbF, etc.].
- Unpaired electron(s) must be in orbitals with $\omega = \frac{1}{2}$. [YbF and PbF have $\sigma_{1/2}$ and $\pi_{1/2}$ electron; PbO has configuration $\pi_{3/2}\pi^*_{-1/2}$.]
- The molecule must have heavy atom and non-zero dipole moment. Unpaired electron must be centered on the heavy atom.
- \Rightarrow HgF, PbF, and YbF are likely to have large enhancement in the ground state. Metastable $a(1)$ state of PbO is also likely to have large enhancement. On the contrary, CsO and IO are not good candidates: for CsO the unpaired electron is on oxygen and IO has ground state $\Pi_{3/2}$.

Molecular calculations

Diatomics in the ground state $\Sigma_{1/2}$ can be calculated with semiempirical and *ab initio* methods. Results of different calculations typically agree within 10 – 20%. [BaF, YbF, HgF]

Semiempirical calculations for $\Pi_{1/2}$ are less reliable, but *ab initio* calculations should be of a similar quality as for $\Sigma_{1/2}$ molecules. [PbF]

State $a(1)$ of PbO is tough for both semiempirical and *ab initio* methods. Results differ by a factor of 2. There is strong configuration mixing.

Molecular ions HBr⁺ and HI⁺ were calculated with atomic perturbation theory and with molecular codes. Last molecular calculation is closer to atomic estimate, but still 2 times smaller. Enhancement is likely to be 2 orders of magnitude smaller than for HgF.

Summary for diatomic molecules

Theory

Ω	Polarization field	E_{eff} (au)	Reliability of calculations	Examples
$\frac{1}{2}$	few kV/cm	$\alpha^2 Z^3$	high	HgF, YbF, PbF
1	few V/cm	$\alpha^2 Z^3$	low	PbO*
$\frac{3}{2}$	\ll V/cm	$0.1\alpha^2 Z^3$	low	HBr ⁺ , HI ⁺

Experiment

At present experiments are going on YbF in London and on PbO in Yale. Experiment on PbF is planned in Oklahoma. Experiments with trapped molecular ions are planned in Boulder.

Solid state

In 1968 Shapiro suggested to look for magnetization of a substance with unpaired electrons in the external electric field. Enhancement factor here is roughly k_{at}/ϵ .

Magnetic flux linked to eEDM is proportional to the spin density and the surface area. Thus, it is advantageous to do experiment with macroscopic solid sample. Such experiment was done in 1978 by Vasil'ev & Kolicheva.

In 2001 Lamoreaux showed that recent advances in magnetometry and new materials, such as gadolinium garnets, allow to raise current sensitivity to eEDM by several orders of magnitude.

Similar experiment can be done with diatomic radicals in a noble gas matrix. In this case we can have 5 orders of magnitude gain in the enhancement factor, while the spin density will be only 2 – 3 orders of magnitude smaller, \Rightarrow the net gain 10^2 .