Search for the Electron Electric Dipole Moment Using PbO*

Experiment:
D.DeMille, D. Kawall, F. Bay, S. Bickman, Y. Jiang, Y. Gurevich, C. Cheung
Yale University

Theory:
M. Kozlov (PNPI, St. Petersburg), D.D.

- Electron EDM--why? how?
- E-field enhancement in atoms & molecules
- The state of the art
- Why PbO?
- Current status: a proof of principle
- The future (near and far term)

Funding: Research Corporation, Packard Foundation, NSF, NIST, Sloan Foundation, CRDF
How does an electron EDM arise?

\[ \mathcal{L}_d = \frac{d_e}{2} \gamma_5 \sigma_{\mu\nu} \psi F_{\mu\nu} \]

Experimental limit: \(|d_e| < 1.6 \times 10^{-27} \text{ e}\cdot\text{cm}\)

| CP-violation model                  | Predicted |\( |d_e| \) |
|-------------------------------------|-----------|-----------|
| Standard Model                      | \(
\sim 10^{-41} \text{ e}\cdot\text{cm}\) |
| Supersymmetry                       | \(< 10^{-25} \text{ e}\cdot\text{cm}\) |
| Left-right symmetric                | \(10^{-26}-10^{-28} \text{ e}\cdot\text{cm}\) |
| Lepton flavor-changing              | \(10^{-26}-10^{-29} \text{ e}\cdot\text{cm}\) |
| Multi-Higgs                         | \(10^{-27}-10^{-28} \text{ e}\cdot\text{cm}\) |
| Technicolor                         | \(~10^{-29} \text{ e}\cdot\text{cm}\) |

\(d_e\) is a powerful probe for new physics!
General method to detect an EDM:
General method to detect an EDM:

\[ \delta \omega = 2 \mu B \]

Energy level picture:
General method to detect an EDM:

\[ \delta \omega = 2 \mu B + 2dE \]

Energy level picture:

Figure of merit for statistical sensitivity:

\[
\frac{\text{shift}}{\text{resolution}} \propto \frac{E}{(1/T)(S/N)^{-1}} = E \cdot T \cdot \sqrt{N}
\]
General method to detect an EDM:

\[ \delta \omega = 2 \mu B - 2dE \]

Energy level picture:

Figure of merit for statistical sensitivity:

\[
\frac{\text{shift}}{\text{resolution}} \propto \frac{E}{(1/T)(S/N)^{-1}} = E \cdot T \cdot \sqrt{N}
\]
Q. Can an electron in a neutral atom/molecule feel a net E-field?

A. No! (Classically)

Schiff's Theorem: \( \mathbf{E}_{\text{tot}} = \mathbf{E}_{\text{ext}} + \mathbf{E}_{\text{int}} = 0! \)

Simple proof: \( \langle \mathbf{E}_{\text{tot}} \rangle \propto \langle \mathbf{F}_{\text{tot}} \rangle = 0 \)
Evasion of Schiff’s Theorem

d_e “feels” E-field because of magnetic forces:

\[ \langle \mathbf{F}_{\text{tot}} \rangle = \langle \mathbf{F}_{\text{el}} + \mathbf{F}_{\text{mag}} \rangle = 0, \]

so \[ \langle \mathbf{E}_{\text{eff}} \rangle = -\langle \mathbf{F}_{\text{mag}} \rangle / e \]

\[ \mathbf{F}_{\text{mag}} = \nabla (\mu \cdot \mathbf{B}) \sim \mu_B \frac{\partial}{\partial r} \left( \frac{\mathbf{E} \times \mathbf{v}}{c} \right) \]

Near nucleus, v, E, \nabla large + substantial amplitude for valence e⁻:

\[ r \sim a_0/Z; \quad v \sim Z \alpha c; \]
\[ E \sim Ze/r^2; \quad \mu_B \sim \alpha e a_0; \quad \psi(0) \sim Z^{1/2} \]

requires polarization \( \mathcal{P} \) to unbalance vector sum

\[ |\langle \mathbf{E}_{\text{eff}} \rangle| \approx Z^3 \alpha^2 \left( e/a_0^2 \right) \cdot \mathcal{P} \]

\[ \sim \mathcal{P} \cdot 10^{11} \text{ V/cm @} \ Z \sim 80! \]
A new direction in EDM searches: using molecules to search for $d_e$

$\uparrow$ \textit{Extremely} large effective E-field with lab-size external field:

$\mathcal{P} \sim 1 \Rightarrow E_{\text{eff}} \sim 10^{11} \text{ V/cm}$

(For atoms, $\mathcal{P} \sim 10^{-3}$ @ $E_{\text{ext}} \sim 100\text{kV/cm}$)

$\downarrow$ Smaller signals due to thermal distribution over rotational levels ($\sim 10^{-4}$)

$\downarrow$ Molecules with unpaired electron spins are thermodynamically disfavored

$\Rightarrow$ high temperature chemistry, even smaller signals
Addressing some problems with molecules: the metastable $a(1)[^3\Sigma^+]$ state of PbO

- PbO is thermodynamically stable (routinely purchased and vaporized)
  $a(1)$ populated via laser excitation (replaces chemistry)

- $a(1)$ has very small $\Omega$-doublet splitting (like any $|\Omega|=1$ state)
  $\Rightarrow$ complete polarization with very small fields ($>15$ V/cm), equivalent to $E \sim >10^7$ V/cm on an atom!

$\Rightarrow$ can work in vapor cell
(Larger density and volume than beam)
**Cell vs. Beam Comparison**

**PbO cell:** Density $n \lesssim 10^{14}/\text{cm}^3$

*Upper limit* on PbO density from PbO*-PbO collisions:
require $T_2 \gtrsim \tau_a \sim 100 \mu\text{s}$, and
assume cross-section $\sigma \lesssim 10^{-14} \text{ cm}^2$

**PbO cell:** Volume $V \sim d^3 \sim 100 \text{ cm}^3$

*Lower limit* on cell dimension from wall collisions:
require $T_2 = d/v \gtrsim \tau_a \sim 100 \mu\text{s}$

$\Rightarrow d \approx 5 \text{ cm}$

**PbO Cell:**
$N = nV \sim 10^{16}$

**Tl Beam:**
$N = nV \sim 10^8$
Searching for Supersymmetry with the electron EDM

Implications of current and ongoing electron EDM searches

AC: Accidental Cancellations
HsF: Heavy sFermions
A-CP: Approx. CP
A-Un: Approx Universality
Align: Alignment
E-Un: Exact Universality
Calculation of the internal E-field of PbO

Basic Method:
[M. G. Kozlov, Sov. Phys. JETP 62, 1114 (1985)]

• MO-LCAO expansion, w/HDF relativistic orbitals as basis
• expansion coefficients from experimental data
  (HFS, fine structure, g-factors)
• $E_{\text{int}}$ mostly from s-p hybridization

Novel aspects of PbO calculation:

• PbO* a(1) state has two valence electrons
• both nominally in $\pi$-type orbitals: no s-wave component
• spin-orbit admixes $\sigma$-type orbitals
• Data indicate large spin-orbit mixing
  $\Rightarrow$ substantial s-p hybridization (much like PbF)

Result:
Lower limit on the observable EDM energy shift $\delta \nu$:

$$\delta \nu = 2 |W_d| d_e \geq 2.4 \times 10^{24} \text{ Hz} \left[ \frac{d_e}{\text{e}\cdot\text{cm}} \right];$$

i.e., $\delta \nu = 24 \text{ mHz} @ d_e = 10^{-27} \text{ e}\cdot\text{cm}$

Slightly larger than for free radicals like YbF, PbF

[M. Kozlov and D. DeMille, Phys. Rev. Lett. 89, 133001 (2002)]
PbO vapor cell: 1st generation

- Moveable plunger for pump-out at temp.
- Flat-on-flat optical polish "seals"
- Solid Fused Silica Light Pipes \( \sim 5\% \times 4\pi \)
- Alumina ceramic cell endcaps & PbO reservoir
- Cell surrounded by vacuum
- Sapphire tube
- Quartz oven body 800 C capability wide optical access w/non-inductive heater for fast switching
Present Experimental Setup

- Pulsed Laser Beam: 5-40 mJ @ 100 Hz
- ∆ν ~ 1 GHz
- ε⊥B
- Larmor Precession ν ~ 100 kHz
- Data Processing
- Solid Quartz Light Pipes
- Vacuum Chamber

Diagram:
- Photo-multiplier tube
- Larmor Precession
- Pulsed Laser Beam 5-40 mJ @ 100 Hz
- ∆ν ~ 1 GHz
- ε⊥B
- Data Processing

Graphs:
- Data processing output
Selective excitation and laser-induced alignment of an $\Omega$-doublet level:

\[ \text{Laser pulse} \quad \lambda \sim 571 \text{ nm} \quad \text{bandwidth} \sim 1 \text{ GHz} \sim \Delta \nu_{\text{Doppler}} \]

\[ \text{X}(0) \left[ ^1\Sigma^+ \right] \quad \rightarrow \quad \text{a}(1) \left[ ^3\Sigma^+ \right] \]

\[ \sim 12 \text{ MHz} \]

\[ \sim 10 \text{ GHz} \]
Vapor cell fluorescence from PbO

\[ X(v'' = 1) \rightarrow a(v' = 5) \text{ excitation (}\lambda = 571 \text{ nm)} \]
\[ a(v' = 5) \rightarrow X(v'' = 0) \text{ detection (}\lambda = 548 \text{ nm)} \]

Integrated over \(\sim 200 \mu s\) after each pulse
Fluorescence vs. time: geometric & collisional effects

(a) Signal reduction from wall quenching
(b) Signal reduction from wall quenching and solid angle effects
(c) Expected Signal (PbO quenching only)
(d) Observed Signal ≈ (b) × (c)
(four parameter fit overlaid)

PbO self-quenching cross-section $\sigma \lesssim 1 \times 10^{-14}$ cm$^2$

Cross section ($10^{-14}$ cm$^2$)

Temperature (Celsius)
Selective excitation and laser-induced alignment of an \( \Omega \)-doublet level:

Zeeman splitting
0-300 kHz typical

Linear polarization \( \varepsilon \perp B \)
\[ |f\rangle = |+1\rangle + |-1\rangle = |\leftrightarrow\rangle \]

Laser pulse
\( \lambda \approx 571 \text{ nm} \)
bandwidth \( \sim 1 \text{ GHz} \) \( \sim \Delta v_{\text{Doppler}} \)

\[ X(0) \ [^1\Sigma^+] \]

\[ a(1) \ [^3\Sigma^+] \]
Zeeman quantum beats in PbO: g-factors

\[ J^P = 1^+ : g_+ = 1.860(8); \quad J^P = 1^- : g_- = 1.857(8) \]

Uncertainties dominated by B-field calibration

Results help constrain calculation of internal E-field

\[ \frac{\Delta g}{g} = \left| \frac{g_+ - g_-}{g} \right| \leq 0.002 \quad (\frac{\Delta g}{g} = 0 \text{ ideal}) \]

Uncertainties dominated by fit systematics

\[ \Rightarrow \Omega \text{-doublet will be near-ideal co-magnetometer} \]
S/N in Zeeman beats

Avg. count rate on $^{208}\text{PbO R0 line} \sim 2 \times 10^7/s$

(consistent with expectations based on current low-efficiency setup)

Short-term frequency fluctuations consistent with expected shot noise: $30 \text{ Hz}/\sqrt{\text{Hz}}$.

⇒ 10 days to match Berkeley (w/ 2nd detector)

Straightforward (but numerous) improvements should increase count rate ($\sim 10^4$), beat contrast ($\sim 2.5$), and background ($\sim 20$)

⇒ x300 Gain in sensitivity

⇒ $\sim 3$ days to $\delta d_e \sim 10^{-29} \text{ e} \cdot \text{cm}!!$
Early results with E-field in cell

- No discharges up to $E = 200 \text{ V/cm}$
- Leakage current $\sim 1 \mu\text{A}$ @ $E = 50 \text{ V/cm}$; probably good enough, but tests indicate that $0.1 \text{ nA}$ is possible
- Beat contrast vs. E-field features (below) under investigation, but qualitatively understood
Summary: a proof of principle

- PbO vapor cell technology in place
- Collisional cross-sections as expected
  \[ \Rightarrow \text{anticipated density OK} \]
- Signal sizes large, consistent with expectation; straightforward improvements will reach target
- Shot-noise limited frequency measurement using quantum beats in fluorescence
- \( g \)-factors of \( \Omega \)-doublet states match precisely
  \[ \Rightarrow \text{co-magnetometer will be very effective} \]
- E-fields of required size applied in cell; no apparent problems
2nd generation cell development

Surface-oxide bonding with gold foil interlayers allows complex geometries, electrical feedthroughs

2nd-generation prototype now in use

EDM cell design

- Re-entrant electrodes so PbO sees only metal
- Guard ring for homogeneity
- Larger volume
- Flat windows
- Compatible with RF needs
The PbO EDM group
Spin alignment & molecular polarization with applied E-field

\[ m = 0 \]

\[ n \]

\[ m = -1 \]

\[ S \]

\[ m = +1 \]

\[ S \]

\[ B_{rf} \perp z \]

\[ \varepsilon \parallel z \]

\[ X, J=0^+ \]
Spin alignment & molecular polarization with applied E-field

$J=1^-$
$m = -1$

$J=1^+$
$m = +1$

$E$
$n$ $n$

$B_{rf} \perp z$

$\varepsilon \parallel z$

$X, J=0^+$
EDM measurement in PbO*: New mechanisms for suppressing systematics!

Ω-doublet levels = comagnetometer: Most systematics cancel in comparison

Tensor Stark splitting eliminates $\mathbf{v} \times \mathbf{E}$ effects
EDM measurement in PbO*: New mechanisms for suppressing systematics!

- doublet levels = comagnetometer: Most systematics cancel in comparison

Tensor Stark splitting eliminates $v \times E$ effects
EDM measurement in PbO*: New mechanisms for suppressing systematics!

$\Omega$-doublet levels = comagnetometer: Most systematics cancel in comparison

Tensor Stark splitting eliminates $\mathbf{v} \times \mathbf{E}$ effects
EDM measurement in PbO*: 
New mechanisms for suppressing systematics!

Ω-doublet levels = comagnetometer: 
Most systematics cancel in comparison

Tensor Stark splitting eliminates \( \mathbf{v} \times \mathbf{E} \) effects
Conclusions and current status of searches for the electron EDM:

• New limit $|d_e| < 1.6 \times 10^{-27}$ e·cm from Berkeley

• New experiment based on PbO* is underway
  – Proof of principle complete--everything OK
  – 1st-generation statistics and systematics at $\sim 10^{-29}$ e·cm looks feasible in $\sim 3$ years
  – More support from molecular theory highly desirable
  – Ultimate configuration could reach $\lesssim 10^{-31}$ e·cm level (but additional spectroscopic data needed)

• Other approaches also under consideration
  – Solid-state (S. Lamoreaux, nucl-ex/0109014; L. Hunter)
    • Good S/N, but systematics unexplored
  – Laser-cooled & trapped Cs atoms (Penn State, Stanford,..)
    • Long coherence times, small enhancement
    • Systematics due to e.g. trapping fields may be a problem
  – YbF molecular beam (Sussex/Imperial College)
    • Recent work gave limit 30× less sensitive than Berkeley
    • Improved beam source (with cooling) for better statistics?
  – ????????
    • Many groups considering various approaches

• Complementary improvements w/neutrons, diamagnetic atoms developing
  (Los Alamos, Seattle, Princeton, …)