Towards an improved limit on the electron electric dipole moment.

Russell Stutz  
Laura Sinclair  
Aaron Leanhardt  
Eric Cornell  
JILA (CU Physics and NIST)  $$:$$ NSF, NIST, Keck Found., Marsico.

Local Theory  
John Bohn  
Ed Meyer  
JILA

“Q: Who are your influences?”  
The Commitments (1991)

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A: John Lee Hooker, Aretha Franklin
JJ Cale,
Ramsey, Commins, Wieman
Hinds, Demille

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Petersburg Nuclear Physics Institute
The Lessons of History: eEDM

Limit on eEDM (e-cm)

- Gould, Sandars, Cs beams
- Hunter, Cs vapor cell
- Commins, Ti beam

The Lessons of History: eEDM

The smooth march of progress into the future.... or....
The Lessons of History: eEDM

...or “Impulse Progress”?

Limit on eEDM (e-cm)

10^{-23} 10^{-24} 10^{-25} 10^{-26} 10^{-27} 10^{-28} 10^{-29}

The Lessons of History: eEDM

...or "Impulse Progress"?
Why Use Molecular Ions?

Why use molecules?
  • Large internal electric fields.
  • Molecules with \( \Omega > 1/2 \) have closely spaced levels of opposite parity → fully polarized with \( E \sim 10 \text{ V/cm} \).
  • Can get \( E_{\text{eff}}/E_{\text{lab}} = 10^9 \)

Why use ions?
  • Ions are easy to trap.
  • Potential for long spin coherence times.
Candidate Molecular Ions

HfF\(^{+}\) and ThF\(^{+}\)
- \(^{3}\Delta\) ground states $\rightarrow <$1 V/cm to fully polarize
- strong atomic 6s character $\rightarrow$ large $E_{\text{eff}}$

Meyer and Bohn “jiffycalc” points in blue. PRA 73, 062108 (2006)
Full-on “one-calculation-equals-one-publication”, various authors, in black, arXiv:physics/0506038 and refs. therein
Why Use $^3\Delta_1$ state of molecule?

$\vec{L} \cdot \vec{z} = 2, \quad \vec{s} \cdot \vec{z} = -1$

$g \approx 0 \quad (= 0.03 \mu_B)$

Thallium: $E_{lab} = 10^5 \text{V/cm} \quad E_{eff} = 6 \times 10^7 \text{ V/cm} \quad \mu_{mag} = 1.0 \mu_B$

HfF$^+$ or ThF$^+$: $E_{lab} = 10^1 \quad E_{eff} = 1.5 \times 10^{10} \quad \mu_{mag} = 0.03$

Figure-of-merit: $E_{eff}/(E_{lab} \mu_{mag})$

Our experiment is $>10^7$ to the good. Probably will not even need mu-metal shielding.
Why Use $^3\Delta_1$ state of molecule?

\[ \bar{L} \cdot \bar{z} = 2, \quad \bar{s} \cdot \bar{z} = -1 \]
\[ g \approx 0 \quad (= 0.03 \mu_B) \]

Thallium: \[ E_{\text{lab}} = 10^5 \text{V/cm} \quad E_{\text{eff}} = 6 \times 10^7 \text{ V/cm} \quad \mu_{\text{mag}} = 1.0 \mu_B \]

HfF$^+$ or ThF$^+$: \[ E_{\text{lab}} = 10^1 \quad E_{\text{eff}} = 1.5 \times 10^{10} \quad \mu_{\text{mag}} = 0.03 \]

Figure-of-merit: \[ \frac{E_{\text{eff}}}{(E_{\text{lab}} \mu_{\text{mag}})} \]

Our experiment is $>10^7$ to the good.

But even 10 V/cm is enough to make an ion scoot away?
Use rotating E-field bias!!!!

- E-field defines quantization axis
- Excellent rejection of lab-frame residual B-field.

\[ \omega_{\text{rot}} \]

\[ \omega_{\text{rot}} \text{ is:} \]
BIG enough that radius of “micromotion” circle is small compared to trap size.

SMALL enough so that \( d_{\text{mol}} E \gg \omega_{\text{rot}} \) and the molecule axis stays aligned with \( E \).

One does Zeeman-level spectroscopy then in the rotating frame.
Experimental Procedure

HfF$^+ \ ^3\Delta_1$ J=1 ground state
• $\Omega$-doublet splitting $\sim$ 1 MHz

Energies not to scale.
Nuclear spin of $\frac{1}{2}$ excluded for clarity.
Experimental Procedure

HfF$^+$ $^3\Delta_1$ J=1 ground state

- Electric field 1 V/cm mixes states of opposite parity.

Energies not to scale.
Experimental Procedure

HfF$^+ \, ^3\Delta_1 \, J=1$ ground state

- Magnetic field lifts degeneracy between $|m|=1$ levels.

Energies not to scale.
Experimental Procedure

HfH$^+ \ 3\Delta_1$ J=1 ground state

- Electron EDM shifts the $|m|=1$ levels in opposite directions in the two $\Omega$-doublet levels.

\[ \mu \parallel B \pm 2d_E \]

\[ \text{Science signal} = 4d_E < 28 \text{mHz}, \quad \text{out of "Berry's offset" of 250 kHz} \]
Experimental Procedure

HfH\(^{+} 3\Delta_1\) J=1 ground state

- Perform electron spin resonance (ESR) frequency measurement via the Ramsey Method.
- Photodissociate one spin state and count HfH\(^{+}\) and Hf\(^{+}\) ions.

\[ 2\mu_m B - 2d_e E_{\text{eff}} \]

\[ 2\mu_m B + 2d_e E_{\text{eff}} \]

\[ m = -1 \quad m = 0 \quad m = +1 \]

Energies not to scale.
Make, cool the molecules,

He or Ne Seed Gas

Pulse valve

Laser Pulse

(Hf or Th) Ablation target

Plus a little SF6
Make, cool the molecules,

He or Ne
Seed Gas

Pulse valve

Laser Pulse

(Hf or Th)
Ablation target

Plus a little SF6

ion/neutral
Experimental Setup

- Laser ablation creates molecular ions.
- Expansion cools ions rotation, vibration, translation (T ~ 2 K).
Experimental Setup

- Linear Paul trap holds ions for measurement.

Trapped $T_{\text{int}} = 2\, \text{K}$, trapped $T_{\text{ext}} = 600\, \text{K}$

Paul trap
Experimental progress: We can, make, stop, trap, and store for many seconds many more HfF\(^+\) (or ThF\(^+\) ions) than we will ever need (or want!) for precision rf spectroscopy of trapped molecular ions.
Experimental Setup

- Linear Paul trap holds ions for measurement.
- Rotating E-field and B-field are applied.
- Rf applied for ESR via Ramsey Method.
- Photodissociation laser pulse to detect spin states.

Trapped $T_{int} = 2K$, trapped $T_{ext} = 600K$
Experimental Setup

• Linear Paul trap holds ions for measurement.
• Rotating E-field and B-field are applied.
• Rf applied for ESR via Ramsey Method.
• Photodissociation laser pulse to detect spin states.
• Channeltron counts atomic or molecular ions.
HfF⁺ electronic states

- $^1\Sigma$ ground state?
- $^3\Delta_1 \sim 800 \text{ cm}^{-1}$ above ground state

S. Petrov, K. Mosyagin, T. Isaev, A. Titov
Experimental Procedure, HfF⁺

\[ \begin{align*}
&1\Pi_1 \\
&3\Pi_1 \\
&3\Delta_1 \\
&1\Sigma \\
\end{align*} \]

\( \tau \sim 1\text{sec} \quad \sim 14000\text{cm}^{-1} \quad \sim 800\text{cm}^{-1} \)

\( m = -1 \quad m = 0 \quad m = +1 \) \quad \text{\( \Omega \)-doublet splitting \( \sim 2 \text{MHz} \)}

\{ \text{SO mixed} \}

Energies not to scale. Nuclear spin of ½ excluded for clarity.
Experimental Procedure, HfF⁺

\[ \begin{align*}
^1\Pi_1 & \quad \text{m = -1} \quad \text{m = 0} \quad \text{m = +1} \\
^3\Pi_1 & \\
^3\Delta_1 & \quad \text{m = -1} \quad \text{m = 0} \quad \text{m = +1} \\
^1\Sigma & \\
\end{align*} \]

SO mixed

2 photon Raman transition to single Zeeman level

Energies not to scale.
Nuclear spin of \( \frac{1}{2} \) excluded for clarity.
Experimental Procedure, HfF\(^+\)

Apply E and B fields to lift degeneracy of \(|m|=1\) levels. Perform electron spin resonance to measure splitting.

Energies not to scale. Nuclear spin of \(\frac{1}{2}\) excluded for clarity.
Experimental Procedure, HfF$^+$

Photo-dissociate one spin state for ESR read out

$^1\Pi_1$

$^3\Pi_1$

$^3\Delta_1$

$m = -1$  $m = 0$  $m = +1$

Repeat Experiment in other $\Omega$-doublet level

$^1\Sigma$

Energies not to scale.
Nuclear spin of $\frac{1}{2}$ excluded for clarity.
HfF⁺ electronic states

• $^1\Sigma$ ground state?
• $^3\Delta_1 \sim 800 \text{ cm}^{-1}$
  above ground state

S. Petrov, K. Mosyagin, T. Isaev, A. Titov
Uncertainties of 1000 cm\(^{-1}\) !!!
(3 \times 10^{13} \text{ Hz})
HfF⁺ Spectroscopy

• Search for HfF⁺ excited electronic states
• Remove ion lens and perform spectroscopy on ion beam before Paul trap
Experimental “progress”: Laser-induced fluorescence signal from neutral (在玩家) HfF

The R, Q, and P branches of a $v'=0, \Omega'=3/2 \leftarrow v''=0, \Omega''=3/2$ transition in HfF
Current limit, beam of atomic Thallium:


$|d_e| < 1.6 \times 10^{-27} \text{ e}^*\text{cm (90\% c.l.)}$

<table>
<thead>
<tr>
<th></th>
<th>$E_{\text{eff}}$</th>
<th>$\tau$</th>
<th>$\sqrt{N_{\text{eff}}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Commins Tl beam</td>
<td>$6 \times 10^7$ V/cm</td>
<td>2 msec</td>
<td>$10^9$ s$^{-1}$</td>
</tr>
<tr>
<td>Hinds YbF beam</td>
<td>$&gt;$</td>
<td>$&lt;$</td>
<td>$&lt;$</td>
</tr>
<tr>
<td>DeMille PbO vapor cell</td>
<td>$&gt;$</td>
<td>$&lt;$</td>
<td></td>
</tr>
<tr>
<td>Weiss trapped Cs</td>
<td>$&lt;$</td>
<td>$&gt;$</td>
<td>$&lt;$</td>
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<tr>
<td>Heinzen trapped Cs</td>
<td>$&lt;$</td>
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</tr>
<tr>
<td>Gould Cs fountain</td>
<td>$&lt;$</td>
<td>$&gt;$</td>
<td>$&lt;$</td>
</tr>
<tr>
<td>Shafer-Ray PbF beam</td>
<td>$&gt;$</td>
<td>$&lt;$</td>
<td></td>
</tr>
<tr>
<td>Cornell trapped HfF+ or ThF+</td>
<td>$&gt;$</td>
<td>$&gt;$</td>
<td>$&lt;&lt;$</td>
</tr>
</tbody>
</table>

Solid State
Sensitivity Estimate

\[ |d_e| < \frac{h}{2E_{\text{eff}} \tau \sqrt{N}} \]

- \( N = 150 \) ions/shot (\( 10^7 \) ions/day)
- \( E_{\text{eff}} = 1.5 \times 10^{10} \) V/cm
- \( \tau = 1 \) second

proj. sensitivity: \( |d_e| < 5 \times 10^{-29} \) e*cm with 1 day of data
Systematic Error Rejection. Key Chops.

<table>
<thead>
<tr>
<th>Chop:</th>
<th>B</th>
<th>E</th>
<th>E/E_{eff}</th>
<th>ν</th>
<th>Other</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tl beam</td>
<td>Y</td>
<td>Y</td>
<td>N</td>
<td>Y</td>
<td></td>
</tr>
<tr>
<td>YbF beam</td>
<td>Y</td>
<td>Y</td>
<td>N</td>
<td>N*</td>
<td></td>
</tr>
<tr>
<td>PbO vapor cell</td>
<td>Y</td>
<td>Y</td>
<td>Y</td>
<td>N*</td>
<td></td>
</tr>
<tr>
<td>trapped Cs</td>
<td>Y</td>
<td>Y</td>
<td>N</td>
<td>Trap</td>
<td></td>
</tr>
<tr>
<td>Cs fountain</td>
<td>Y</td>
<td>Y</td>
<td>N</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>PbF beam</td>
<td>Y</td>
<td>Y</td>
<td>N</td>
<td>N*</td>
<td></td>
</tr>
<tr>
<td>Trapped MF+</td>
<td>Y</td>
<td>N</td>
<td>Y</td>
<td>Rotation sense</td>
<td></td>
</tr>
</tbody>
</table>
We've got the chops, and:

Key fact: $\nu_{\text{science}}$ is independent of magnitude of $E$, $B$, and $\omega_{\text{rot}}$. Also should be independent of strength of ion trap confinement, $T$, and $n_{\text{ion}}$. 

<table>
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<tr>
<th>Trapped MF+</th>
<th>Y</th>
<th>N</th>
<th>Y</th>
<th>Y*</th>
<th>Rotation sense</th>
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Rotating electric quantization axis

Stark shift and Berry’s phase:

We are working in a TRAP. Electric field is perforce spatially inhomogenous, \textit{and} it rotates!
Consequences for decoherence? systematics?
Rotating electric quantization axis

**Stark shift and Berry’s phase:**

\[
U(M, E, \Omega, \omega_r) = |d_{mol}E|M\Omega + MA\omega_r + \frac{\alpha MA\omega_r^2}{|d_{mol}E|\Omega} + \frac{\beta MA\omega_r^3}{(|d_{mol}E|\Omega)^2} + \ldots
\]
Rotating electric quantization axis

\[ \Omega = 1 \]

Stark shift and Berry’s phase:

\[
U(M, E, \Omega, \omega_r) = \left| d_{mol} E \right| M\Omega + M A \omega_r + \frac{\alpha MA \omega_r^2}{\left| d_{mol} E \right| \Omega} + \frac{\beta MA \omega_r^3}{\left( \left| d_{mol} E \right| \Omega \right)^2} + \ldots
\]

(A, solid angle = 2\pi)

\[
U = \left( \left| d_{mol} E \right| M\Omega \right) \left[ 1 + A \varepsilon + \alpha A \varepsilon^2 + \beta A \varepsilon^3 \right]
\]

\[
\varepsilon \equiv \frac{\omega_{rot}}{\left| d_{mol} E \right| \Omega}
\]
Stark shift and Berry’s phase:

\[ U(M, E, \Omega, \omega_r) = |d_{mol}E| M\Omega + MA\omega_r + \frac{\alpha MA\omega_r^2}{|d_{mol}E| \Omega} + \frac{\beta MA\omega_r^3}{(|d_{mol}E| \Omega)^2} + \ldots \]

- 80 MHz \hspace{1cm} 250 kHz \hspace{1cm} 800 Hz \hspace{1cm} 2.5 Hz
- 80 MHz \hspace{1cm} 250 kHz \hspace{1cm} 800 Hz \hspace{1cm} 2.5 Hz
The decohering effects of ion-ion collisions:

\[ E_{\text{bias}} + E_{\text{ion-ion}} \]

Ion picks up a little random Berry’s phase with each near miss.

\[ \tau_{\text{cohere}} \propto n_{\text{ion}}^{-1} \]

Sensitivity to EDM fairly flat with \( N_{\text{ion}} \), but \( N_{\text{usable}} / N_{\text{ion}} \) is critical. (And rather uncertain).
Random issues:

Inhomogeneity in $E_{rot}$ can lead to line-broadening but first to problems with ponderomotive potential (an anti-Paul trap.)

Momentum kick associated with optical pumping to one Omega level or the other can be troublesome if one doesn’t insert a suitable dwell time.
Systematics bottom line:
We haven’t thought of a killer systematic at the 10\(^{-28}\) level yet. We will have a number of powerful techniques for smoking out unforeseen ones.

In the end, we’ve got to try it.
Decoherence Effects

- Axial electric field tips molecular axis out of radial plane.
- Rotating spin picks up spatially varying Berry’s phase.
- Net effect cancels out in the absence of ion-ion collisions.
- Collisions lead to phase diffusion and decoherence.

\[ \sqrt{\langle \Delta \varphi^2 \rangle} = \pi \sqrt{N_{\text{coll}}} \sqrt{\frac{k_B T}{U_{\text{rot}}}} \approx \pi \times 0.1 \]
Experimental Progress

- Laser ablation of Hf or Th targets in the expansion gives HfF+, ThF+. We can “catch”, trap, $10^5$/shot, hold time $>> 1$s.
- Comoving beam temperature measured (in Hf and Hf+) $< 2$K.
- Photodissociation of CH+ to C+ and H.

\[
\text{CH}^+ \rightarrow \text{C}^+ + \text{H}
\]
Initial spectroscopy

- Must scan over 1000’s of cm$^{-1}$’s, doppler width of $\sim$50 MHz
  - Course scans with pulsed dye laser
  - Fine scans with cw Ti:Saph laser
- Optimized detection on Hf neutral lines
  - S/N $\sim$10$^4$:1 w/ Ti:Saph laser
  - Observed cooling of Hf fine structure from Helium collisions
Experimental Progress

- Comoving beam temperature measured (in Hf and Hf+) <2 K.
- Photodissociation of CH+ to C+ and H.

T = 2K, Hf laser fluorescence, crossed-beam doppler width.

T~1K Hf+ beam (measured with translatable ion detector)
Sensitivity Estimate

\[ |d_e| < \frac{h}{2E_{\text{eff}} \tau \sqrt{N}} \]

- \( N = 150 \) ions/shot \((10^7 \) ions/day\)
- \( E_{\text{eff}} = 3 \times 10^{10} \) V/cm
- \( \tau = 1 \) second

Inverts EDM signal \( \rightarrow \) 
- Flip B-Field bias gradient.
- Change \( \Omega \)-doublet levels.
- Change sign, mag., freq of \( E_{\text{rot}} \).
- Multiple internal mol. states

Constant EDM signal \( \rightarrow \) 
- Multiple internal mol. states

proj. sensitivity: \( |d_e| < 6 \times 10^{-29} \) e\(^*\)cm with 1 day of data

\[ |d_e| < 1.6 \times 10^{-27} \) e\(^*\)cm \]
E.D. Commins TI Exp. Limit \([\text{PRL 88, 071805 (2002)}]\)
Common traits of (AMO) eEDM searches

- Magnetic and dipole moment of electron must be aligned
  - no other direction to point
- Perform electron spin resonance in the presence of E and B fields

Look for $\omega_1 \neq \omega_2$, $\hbar(\omega_1-\omega_2) = 4d_e \cdot E$

Figure of Merit for experiment

$$E_{\text{eff}} \cdot \tau \cdot \sqrt{N}$$