

Order of Magnitude Smaller Limit on the Electric Dipole Moment of the Electron

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The Standard Model (SM) of particle physics is known to be incomplete. Extensions to the SM, such as weak-scale Supersymmetry, posit the existence of new particles and interactions that are asymmetric under time-reversal (T), and nearly always predict a small, yet potentially measurable (10^{-27} - 10^{-30} e cm) electron electric dipole moment (EDM, d_e). The EDM is an asymmetric charge distribution along the spin (\vec{S}) that is also asymmetric under T. Using the polar molecule thorium monoxide (ThO), we measure $d_e = (-2.1 \pm 3.7_{\text{stat}} \pm 2.5_{\text{syst}}) \times 10^{-29}$ e cm. This corresponds to an upper limit of $|d_e| < 8.7 \times 10^{-29}$ e cm with 90 percent confidence, an order of magnitude improvement in sensitivity compared to the previous best limit. Our result constrains T-violating physics at the TeV energy scale.

The exceptionally high internal effective electric field (\mathcal{E}_{eff}) of heavy neutral atoms and molecules can be used to precisely probe for d_e via the energy shift $U = -\vec{d}_e \cdot \vec{\mathcal{E}}_{\text{eff}}$, where $\vec{d}_e = d_e \vec{S}/(\hbar/2)$ and \hbar is the reduced Planck constant. Valence electrons travel relativistically near the heavy nucleus, making \mathcal{E}_{eff} up to a million times larger than any static laboratory field (1–3). The previous best limits on d_e came from experiments with thallium (Tl) atoms (4) ($|d_e| < 1.6 \times 10^{-27}$ e cm), and ytterbium fluoride (YbF) molecules (5, 6) ($|d_e| < 1.06 \times 10^{-27}$ e cm). The latter demonstrated that molecules can be used to suppress the motional electric fields and geometric phases that limited the Tl measurement (5) (this suppression is also present in certain atoms (7)). Insofar as polar molecules can be fully polarized in laboratory-scale electric fields (\mathcal{E}), \mathcal{E}_{eff} can be much greater than in atoms. The $^3\Delta_1$ electronic state in the molecule ThO provides an $\mathcal{E}_{\text{eff}} \approx 84$ GV/cm, larger than those previously used in EDM measurements (8, 9). Its unusually small magnetic moment reduces its sensitivity to spurious magnetic fields (10, 11). Improved systematic error rejection is possible because internal state selection allows the reversal of \mathcal{E}_{eff} with no change in $\vec{\mathcal{E}}$ (12, 13).

To measure d_e we perform a spin precession measurement (10, 14, 15) on pulses of $^{232}\text{Th}^{16}\text{O}$ molecules from a cryogenic buffer gas beam source (16–18). The pulses pass between parallel plates that generate a laboratory electric field $\mathcal{E}_z \hat{z}$ (Fig. 1A). A coherent superposition of two spin states, corresponding to a spin aligned in the xy plane, is prepared

using optical pumping and state preparation lasers. Parallel electric ($\vec{\mathcal{E}}$) and magnetic ($\vec{\mathcal{B}}$) fields exert torques on the electric and magnetic dipole moments, causing the spin vector to precess in the xy plane. The precession angle is measured with a readout laser and fluorescence detection. A change in this angle as $\vec{\mathcal{E}}_{\text{eff}}$ is reversed is proportional to d_e .

In more detail, a 943 nm laser beam optically pumps molecules from the ground electronic state into the lowest rotational level, $J = 1$, of the metastable (lifetime ~ 2 ms) electronic $H^3\Delta_1$ state manifold (Fig. 1B), in an incoherent mixture of the $\tilde{N} = \pm 1$, $M = \pm 1$ states. M is the angular momentum projection along the \hat{z} axis. \tilde{N} refers to the internuclear axis, \hat{n} , aligned (+1) or anti-aligned (−1) with respect to $\vec{\mathcal{E}}$, when $|\vec{\mathcal{E}}| > \sim 1$ V/cm (11). The linearly polarized state-preparation laser's frequency is resonant with the $H \rightarrow C$ transition at 1090 nm (Fig. 1B). Within the short-lived (500 ns) electronic C state there are two opposite parity $\tilde{P} = \pm 1$ states with $J = 1$, $M = 0$. For a given spin precession measurement, the laser frequency determines the \tilde{N}, \tilde{P} states that are addressed. This laser optically pumps the bright superposition of the

two resonant $M = \pm 1$ sublevels out of the H state, leaving behind the orthogonal dark superposition that cannot absorb the laser light; we use this dark state as our initial state (19). If the state preparation laser were polarized along \hat{x} , the prepared state, $|\psi(\tau=0), \tilde{N}\rangle$, has the electron spin aligned along the \hat{y} axis. The spin then precesses in the xy plane by angle ϕ to

$$|\psi(\tau), \tilde{N}\rangle = \frac{\exp(-i\phi)|M=+1, \tilde{N}\rangle - \exp(i\phi)|M=-1, \tilde{N}\rangle}{\sqrt{2}} \quad (1)$$

As $\vec{\mathcal{E}}$ and $\vec{\mathcal{B}}$ are aligned along \hat{z} , the phase ϕ is determined by $|\mathcal{B}_z| = |\vec{\mathcal{B}} \cdot \hat{z}|$, its sign, $\tilde{\mathcal{B}} = \text{sgn}(\vec{\mathcal{B}} \cdot \hat{z})$, and the electron's EDM, d_e :

$$\phi \approx \frac{-(\mu_B g \tilde{\mathcal{B}} |\mathcal{B}_z| + \tilde{N} \tilde{\mathcal{E}} d_e \mathcal{E}_{\text{eff}}) \tau}{\hbar} \quad (2)$$

where $\tilde{\mathcal{E}} \equiv \text{sgn}(\vec{\mathcal{E}} \cdot \hat{z})$, τ is the spin precession time, and $\mu_B g$ is the magnetic moment (15) of the H , $J = 1$ state where $g = 0.044(1)$ and μ_B is the Bohr magneton. The sign of the EDM term, $\tilde{N} \tilde{\mathcal{E}}$, arises from the relative orientation between $\vec{\mathcal{E}}_{\text{eff}}$ and the electron spin, as illustrated in Fig. 1B.

After the spin precesses over a distance of $L \approx 22$ cm ($\tau \approx 1.1$ ms), we measure ϕ by optically pumping on the same $H \rightarrow C$ transition with the state readout laser. The laser polarization alternates between \hat{X} and

\hat{Y} every 5 s and we record the modulated fluorescence signals S_X and S_Y from the decay of C to the ground state (fig. S1A). This procedure amounts to a projective measurement of the spin onto \hat{X} and \hat{Y} , which are defined such that \hat{X} is at an angle θ with respect to \hat{x} in the xy plane (Fig. 1A). To normalize out molecule number fluctuations, we compute the asymmetry, (10)

$$\mathcal{A} \equiv \frac{S_X - S_Y}{S_X + S_Y} = C \cos[2(\phi - \theta)] \quad (3)$$

where the contrast C is $94 \pm 2\%$ on average. We set $|\mathcal{B}_z|$ and θ such that $\phi - \theta \approx (\pi/4)(2n + 1)$ for integer n , so that the asymmetry is linearly proportional to small changes in ϕ , and maximally sensitive to the EDM. We measure C by dithering θ between two nearby values that differ by 0.1 rad, denoted by $\tilde{\theta} = \pm 1$.

We perform this spin precession measurement repeatedly under varying experimental conditions to (a) distinguish the EDM energy shift from background phases and (b) search for and monitor possible systematic errors. Within a “block” of data (fig. S1C) taken over 40 s, we perform measurements of the phase for each experimental state derived from 4 binary switches, listed from fastest (.5 s) to slowest (20 s): the molecule alignment, $\tilde{\mathcal{N}}$; the \mathcal{E} -field direction, $\tilde{\mathcal{E}}$; the readout laser polarization dither state, $\tilde{\theta}$; and the \mathcal{B} -field direction, $\tilde{\mathcal{B}}$. For each $(\tilde{\mathcal{N}}, \tilde{\mathcal{E}}, \tilde{\theta})$ state of the experiment, we measure \mathcal{A} and C , from which we can extract ϕ . Within each block, we form “switch-parity components” of the phase, ϕ^u , that are combinations of the measured phases that are odd or even under these switch operations (13). We denote the switch-parity of a quantity with a superscript, u , listing the switch labels under which the quantity is odd; it is even under all unlabeled switches. For example, the EDM contributes to a phase component $\phi^{\mathcal{N}\mathcal{E}} = -d_e \mathcal{E}_{\text{eff}} \tau / \hbar$. We extract the mean precession time τ from $\phi^{\mathcal{B}} = -\mu_{\text{BG}} |\mathcal{B}_z| \tau / \hbar$ and compute the frequencies, $\omega^u \equiv \phi^u / \tau$. The EDM value is obtained from $\omega^{\mathcal{N}\mathcal{E}}$ by $d_e = -\hbar \omega^{\mathcal{N}\mathcal{E}} / \mathcal{E}_{\text{eff}}$.

On a slower time scale, we perform additional “superblock” binary switches (fig. S1D) to suppress some known systematic errors and to search for unknown ones. These switches, which occur on the 40-600 s time scales, are: the excited state parity addressed by the state readout lasers, $\tilde{\mathcal{P}}$; a rotation of the readout polarization basis by $\theta \rightarrow \theta + \pi/2$, $\tilde{\mathcal{R}}$; a reversal of the leads that supply the electric fields, $\tilde{\mathcal{L}}$; and a global polarization rotation of both the state preparation and readout laser polarizations, $\tilde{\mathcal{G}}$. The $\tilde{\mathcal{P}}$ and $\tilde{\mathcal{R}}$ switches interchange the role of the \hat{X} and \hat{Y} readout beams and hence reject systematic errors associated with small differences in power, shape, or pointing. The two $\tilde{\mathcal{G}}$ state angles are chosen to suppress systematics that couple to unwanted ellipticity imprinted on the polarizations by birefringence in the electric field plates. The $\tilde{\mathcal{L}}$ switch rejects systematics that couple to an offset voltage in the electric field power supplies. We extract the EDM from $\omega^{\mathcal{N}\mathcal{E}}$ after a complete set of the 2^8 block and superblock states. The $\omega^{\mathcal{N}\mathcal{E}}$ is even under all of the superblock switches.

The total dataset consists of $\sim 10^4$ blocks of data, taken over the course of ~ 2 weeks (fig. S1, E and F). During this dataset, we also varied, from fastest (hours) to slowest (a few days): the \mathcal{B} -field magnitude, $|\mathcal{B}_z| \approx 1, 19, 38$ mG (corresponding to $|\phi| \approx 0, \pi/4, \pi/2$ respectively), the \mathcal{E} -field magnitude $|\mathcal{E}_z| \approx 36, 141$ V/cm, and the pointing direction of the lasers, $\hat{k} \cdot \hat{z} = \pm 1$. Figure 2B shows measured EDM values obtained when the dataset is grouped according to the states of $|\mathcal{B}_z|$, $|\mathcal{E}_z|$, $\hat{k} \cdot \hat{z}$, and

each superblock switch. All of these measurements are consistent within 2σ .

We compute the 1σ standard error in the mean and use standard Gaussian error propagation to obtain the reported statistical uncertainty. The reported upper limit is computed using the Feldman-Cousins prescription (20) applied to a folded normal distribution. To prevent experimental bias, we performed a blind analysis by adding an unknown offset to $\omega^{\mathcal{N}\mathcal{E}}$. The mean, statistical error, systematic shifts, and procedure for calculating the systematic error were determined before unblinding. Figure 2A shows a histogram of EDM measurements. The asymmetry, \mathcal{A} , obeys a ratio distribution, which has large non-Gaussian tails in the limit of low signal to noise (21). We apply a photon count rate threshold cut so that we only include data with a large signal-to-noise, resulting in a statistical distribution that closely approximates a Gaussian. When the EDM measurements are fit to a constant value, the reduced chi-squared is $\chi^2 = 0.996 \pm 0.006$. Based on the total number of detected photoelectrons (~ 1000 per pulse) that contribute to the measurement, the statistical uncertainty is 1.15 times that from shot noise (15).

To search for possible sources of systematic error, we varied over 40 separate parameters (table S1) and observed their effect on $\omega^{\mathcal{N}\mathcal{E}}$ and many other components of the phase correlated with $\tilde{\mathcal{N}}$, $\tilde{\mathcal{E}}$, or $\tilde{\mathcal{B}}$. These parameters are intentionally applied tunable imperfections, such as transverse magnetic fields or laser detunings. These systematic checks were performed concurrently with the 8 block and superblock switches.

We assume that $\omega^{\mathcal{N}\mathcal{E}}$ depends linearly on each parameter P , so that the possible systematic shift and uncertainty of $\omega^{\mathcal{N}\mathcal{E}}$ is evaluated from the measured slope, $S = \partial \omega^{\mathcal{N}\mathcal{E}} / \partial P$, and the parameter value during normal operation (obtained from auxiliary measurements). If S is not monitored throughout the data set, we do not apply a systematic correction but simply include the measured upper limit in our systematic error budget. Data taken with intentionally applied parameter imperfections is used only for determination of systematic shifts and uncertainties. Table 1 lists all contributions to our systematic error.

We identified two parameters that systematically shift the value of $\omega^{\mathcal{N}\mathcal{E}}$ within our experimental resolution. Both parameters couple to the AC Stark shift induced by the lasers. The molecules are initially prepared in the dark state with a spin orientation dependent on the laser polarization. If there is a polarization gradient along the molecular beam propagation direction, the molecules acquire a small bright state amplitude. Away from the center of a Gaussian laser profile, the laser can be weak enough that the bright state amplitude is not rapidly pumped away; it acquires a phase relative to the dark state due to their mutual energy splitting, given by the AC Stark shift. An equivalent phase is acquired in the state readout laser. This effect changes the measured phase by $\phi_{\text{AC}}(\Delta, \Omega_r) \approx (\alpha \Delta + \beta \Omega_r)$, where Δ , Ω_r are the detuning from and Rabi frequency of the $H \rightarrow C$ transition, respectively. The constants α , β are measured directly by varying Δ and Ω_r , and depend on the laser’s spatial intensity and polarization profile. These measurements are in good agreement with our analytical and numerical models.

A significant polarization gradient is caused by laser-induced thermal stress birefringence (22) in the electric field plates. The laser beams are elongated perpendicular to the molecular beam axis, which creates an asymmetric thermal gradient and defines the axes for the resulting birefringence gradient. By aligning the laser polarization with the birefringence axes, the polarization gradient can be minimized. We have

verified this both with polarimetry (23) and through the resulting AC Stark shift systematic (Fig. 3A).

Such AC Stark shift effects can cause a systematic shift in the measurement of ω^{ve} in the presence of an $\tilde{\mathcal{N}}\tilde{\mathcal{E}}$ correlated detuning, Δ^{ve} , or Rabi frequency, Ω_r^{ve} . We observe both.

The detuning component Δ^{ve} is caused by a non-reversing \mathcal{E} -field component \mathcal{E}^{nr} , generated by patch potentials and technical voltage offsets, which is small relative to the reversing component, $|\mathcal{E}_z|\tilde{\mathcal{E}}$. The \mathcal{E}^{nr} creates a correlated DC Stark shift with an associated detuning $\Delta^{\text{ve}} = D\mathcal{E}^{\text{nr}}$, where D is the H state electric dipole moment. We measured \mathcal{E}^{nr} via microwave spectroscopy (Fig. 3B), two-photon Raman spectroscopy, and the $\tilde{\mathcal{N}}\tilde{\mathcal{E}}$ -correlated contrast.

The Rabi frequency component Ω_r^{ve} , arises from a dependence of Ω_r on the orientation of the molecular axis, $\hat{n} \approx \tilde{\mathcal{N}}\tilde{\mathcal{E}}\hat{z}$, with respect to laser propagation direction, \hat{k} . This $\hat{k} \cdot \hat{n}$ dependence can be caused by interference between E1 and M1 transition amplitudes on the $H \rightarrow C$ transition. Measurements of a nonzero $\tilde{\mathcal{N}}\tilde{\mathcal{E}}$ -correlated fluorescence signal S^{ve} , and an $\tilde{\mathcal{N}}\tilde{\mathcal{E}}\tilde{\mathcal{B}}$ -correlated phase ϕ^{ve} , both of which changed sign when we reversed \hat{k} , provided evidence for a nonzero Ω_r^{ve} . The ϕ^{ve} channel, along with its linear dependence on an artificial Ω_r^{ve} generated by an $\tilde{\mathcal{N}}\tilde{\mathcal{E}}$ correlated laser intensity, allowed us to measure $\Omega_r^{\text{ve}}/\Omega_r = (-8.0 \pm 0.8) \times 10^{-3}(\hat{k} \cdot \hat{z})$, where Ω_r is the uncorrelated (mean) Rabi frequency (see supplementary materials).

By intentionally exaggerating these parameters we verified that both \mathcal{E}^{nr} and Ω_r^{ve} couple to AC Stark shift effects to produce a false EDM. We tuned the laser polarization for each $\tilde{\mathcal{G}}$ state to minimize the magnitude of the systematic slope $\partial\omega^{\text{ve}}/\partial\mathcal{E}^{\text{nr}}$ (Fig. 3A). The correlations $\partial\omega^{\text{ve}}/\partial\mathcal{E}^{\text{nr}}$ and $\partial\omega^{\text{ve}}/\partial\Omega_r^{\text{ve}}$ were monitored at regular intervals throughout the data set (fig. S1E). The resulting systematic corrections to ω^{ve} were all < 1 mrad/s.

For a subset of our data, the $\tilde{\mathcal{N}}$ -correlated phase ϕ^{v} was nonzero and drifted with time. We identified the cause of this behavior as an $\tilde{\mathcal{N}}$ -correlated laser pointing $\hat{k}^{\text{N}} \cdot \hat{x} \approx 5 \mu\text{rad}$ present in our optical frequency switching setup. We eliminated this effect with improved optical alignment; however, since we were not able to determine the precise mechanism by which \hat{k}^{N} coupled to ϕ^{v} , we chose to include ϕ^{v} variations in our systematic error budget. The slope $\partial\omega^{\text{ve}}/\partial\phi^{\text{v}}$ (consistent with zero) and the mean value of ϕ^{v} established a systematic uncertainty limit of ~ 1 mrad/s on ω^{ve} .

To be cautious, we include in our systematic error budget possible contributions from the following parameters that caused a nonzero EDM shift in experiments similar to ours: stray \mathcal{B} -fields $\mathcal{B}_{x,y,z}^{\text{nr}}$ and \mathcal{B} -field gradients (13); an $\tilde{\mathcal{E}}$ -correlated phase, ϕ^{e} , caused by leakage current, $\vec{v} \times \tilde{\mathcal{E}}$, and geometric phase effects (4); and laser detunings and \mathcal{E} -field ground offsets (5). We obtained direct ω^{ve} systematic limits of $< \sim 1$ mrad/s for each. We simulated the effects that contribute to ϕ^{e} by correlating \mathcal{B}_z with $\tilde{\mathcal{E}}$, which allowed us to place a $\sim 10^{-2}$ mrad/s limit on their combined effect. Because of our slow molecular beam, relatively small applied \mathcal{E} -fields, and small magnetic dipole moment, we do not expect any of these effects to systematically shift ω^{ve} above the 10^{-3} mrad/s level (10, 11).

The result of this first-generation ThO measurement,

$$d_e = (-2.1 \pm 3.7_{\text{stat}} \pm 2.5_{\text{syst}}) \times 10^{-29} e \text{ cm} \quad (4)$$

comes from $d_e = -\hbar\omega^{\text{ve}}/\mathcal{E}_{\text{eff}}$ using $\mathcal{E}_{\text{eff}} = 84 \text{ GV/cm}$ (8, 9) and $\omega^{\text{ve}} = (2.6 \pm$

$4.8_{\text{stat}} \pm 3.2_{\text{syst}}) \text{ mrad/s}$. This sets a 90 percent confidence limit,

$$|d_e| < 8.7 \times 10^{-29} e \text{ cm} \quad (5)$$

that is 12 times smaller than the previous best limit (4, 5), an improvement made possible by the first use of the ThO molecule and of a cryogenic source of cold molecules for this purpose. If we were to take into account the roughly estimated 15 percent uncertainty on the calculated \mathcal{E}_{eff} (8), and assume that this represents a 1σ Gaussian distribution width, the d_e limit stated above would increase by about 5 percent. Because paramagnetic molecules are sensitive to multiple T-violating effects (24), our measurement should be interpreted as $\hbar\omega^{\text{ve}} = -d_e\mathcal{E}_{\text{eff}} - W_S C_S$, where C_S is a T-violating electron-nucleon coupling, and W_S is a molecule-specific constant (8, 25). For the d_e limit above we assume $C_S = 0$. Assuming instead that $d_e = 0$ yields $C_S = (-1.3 \pm 3.0) \times 10^{-9}$, corresponding to a 90 percent confidence limit $|C_S| < 5.9 \times 10^{-9}$ that is 9 times smaller than the previous limit (26).

A measurably large EDM requires new mechanisms for T violation, which is equivalent to charge conjugation-parity (CP) violation given the CPT invariance theorem (2). Nearly every extension to the SM (27, 28) introduces new CP violating phases ϕ_{CP} . It is difficult to construct mechanisms that systematically suppress ϕ_{CP} , so model builders typically assume $\sin(\phi_{\text{CP}}) \sim 1$ (29). An EDM arising from new particles at energy Λ in an n -loop Feynman diagram will have size

$$\frac{d_e}{e} \sim \kappa \left(\frac{\alpha_{\text{eff}}}{4\pi} \right)^n \left(\frac{m_e c^2}{\Lambda^2} \right) \sin(\phi_{\text{CP}}) (\hbar c)^{-1} \quad (6)$$

where α_{eff} (about $4/137$ for electroweak interactions) encodes the strength with which the electron couples to the new particles, m_e is the electron mass, and $\kappa \sim 0.1$ is a dimensionless prefactor (2, 30, 31). In models where 1- or 2-loop diagrams produce d_e , our result typically sets a bound on CP violation at energy scales $\Lambda \sim 3 \text{ TeV}$ or 1 TeV , respectively (27–29, 31). Hence, within the context of many models, our more precise EDM limit constrains CP violation up to energy scales similar to or higher than those explored directly at the Large Hadron Collider.

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Table 1. Summary of systematic errors. Systematic and statistical errors for $\omega^{N\mathcal{E}}$, in units of mrad/s. All errors are added in quadrature, and are derived from Gaussian 1 σ (68%) confidence intervals. In EDM units, 1.3 mrad/s $\approx 10^{-29}$ e cm.

Parameter	Shift	Uncertainty
\mathcal{E}^{nr} correction	-0.81	0.66
$\Omega_r^{N\mathcal{E}}$ correction	-0.03	1.58
ϕ^e correlated effects	-0.01	0.01
ϕ^N correlation		1.25
Non-reversing \mathcal{B} -field ($\mathcal{B}_z^{\text{nr}}$)		0.86
Transverse \mathcal{B} -fields ($\mathcal{B}_x^{\text{nr}}, \mathcal{B}_y^{\text{nr}}$)		0.85
\mathcal{B} -field gradients		1.24
Prep./read laser detunings		1.31
\tilde{N} correlated detuning		0.90
\mathcal{E} -field ground offset		0.16
Total systematic	-0.85	3.24
Statistical		4.80
Total uncertainty		5.79

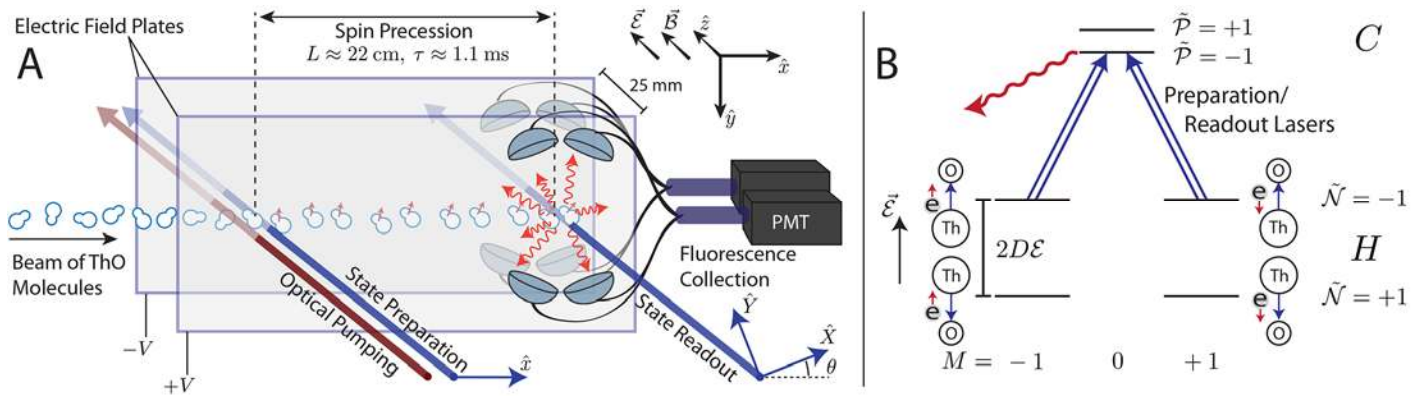


Fig. 1. Schematic of the apparatus and energy level diagram. (A) A collimated pulse of ThO molecules enters a magnetically shielded region (not to scale). An aligned spin state (smallest red arrows), prepared via optical pumping, precesses in parallel electric and magnetic fields. The final spin alignment is read out by a laser with rapidly alternating linear polarizations, \hat{X} , \hat{Y} , with the resulting fluorescence collected and detected with photomultiplier tubes (PMTs). (B) The state-preparation and readout lasers (double lined blue arrows) drive one molecule orientation $\tilde{N} = \pm 1$ (split by $2D\mathcal{E} \sim 100$ MHz, where D is the electric dipole moment of the H state) in the H state to C , with parity $\tilde{P} = \pm 1$ (split by 50 MHz). Population in the C state decays via spontaneous emission, and we detect the resulting fluorescence (red wiggly arrow). H state levels are accompanied by cartoons displaying the orientation of $\vec{\mathcal{E}}_{\text{eff}}$ (blue arrows) and the spin of the electron (red arrows) that dominantly contributes to the d_e shift.

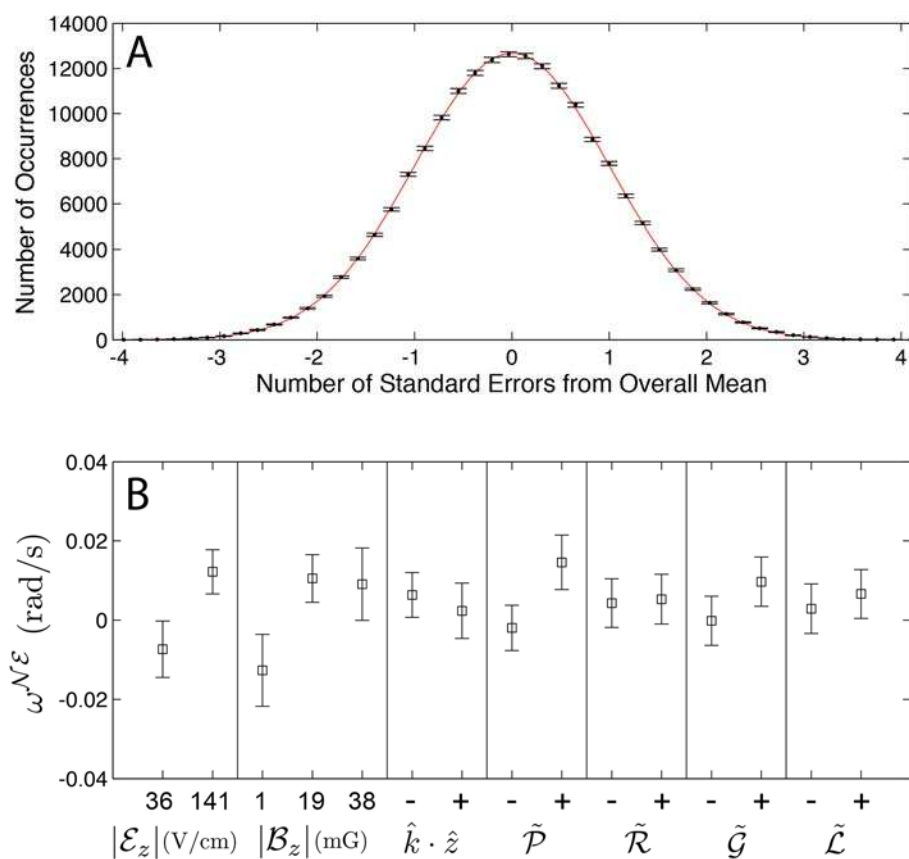


Fig. 2. Statistical spread of $\omega^{\text{N}\varepsilon}$ measurements. (A) Histogram of $\omega^{\text{N}\varepsilon}$ measurements for each time point (within molecule pulse) and for all blocks. Error bars represent expected Poissonian fluctuations in each histogram bin. (B) Measured $\omega^{\text{N}\varepsilon}$ values grouped by the states of $|\mathcal{B}_z|$, $|\mathcal{E}_z|$, $\hat{k} \cdot \hat{z}$, and each superblock switch, before systematic corrections.

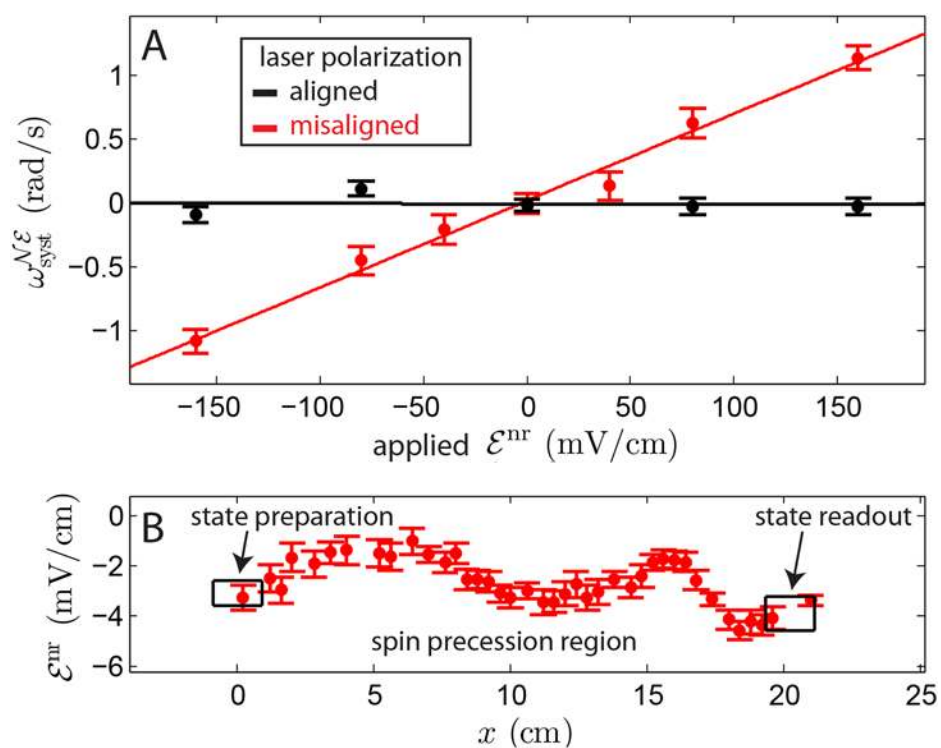


Fig. 3. The ϵ^{nr} systematic. (A) Tuning out laser polarization gradient and $\partial\omega^{\text{nr}}/\partial\epsilon^{\text{nr}}$ (see text for details). The red (black) points were taken with the polarization misaligned (aligned) with the birefringence axes of the electric field plates. (B) Microwave spectroscopic measurement of ϵ^{nr} during normal operation along the molecule beam axis, x .



Supplementary Materials for

Order of Magnitude Smaller Limit on the Electric Dipole Moment of the Electron

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Materials and Methods

Fig. S1

Table S1

References

Supplementary Materials

Apparatus

We create a pulsed molecular beam of ThO using the buffer gas beam technique^{16–18}. Each packet of molecules leaving the source contains $\sim 10^{11}$ ThO molecules in the $J = 1$ rotational level of the ground electronic (X) and vibrational state and are produced at a repetition rate of 50 Hz. The packet is 2–3 ms wide and has a center of mass speed of ~ 200 m/s. The chamber background pressure of $< 10^{-6}$ Torr suggests a ThO-background gas collision probability of $\lesssim 1\%$ during the spin precession measurement which could cause a small decrease in fluorescence signal or contrast.

After leaving the cryogenic beam source chamber, the ground state molecules are in a thermal distribution of rotational states at about 4 K with a rotational constant of about $B_R \approx 10$ GHz. We use a series of lasers and microwaves to enhance the population of the single rotational state, $|X; J = 1\rangle$. The molecules travel through optical pumping lasers resonant with the $|X; J = 2, 3\rangle \rightarrow |C; J = 1, 2\rangle$ transitions, followed by a microwave field resonant with the $|X; J = 0\rangle \leftrightarrow |X; J = 1\rangle$ transition. The optical pumping lasers transfer population from $|X; J = 2, 3\rangle$ into the $|X; J = 0, 1\rangle$ states respectively. The microwaves then mix the populations of $|X; J = 0, M = 0\rangle$ and $|X; J = 1, M = 0\rangle$ resulting in an overall population increase in $|X; J = 1\rangle$ of a factor of ~ 2 .

The molecules then pass through adjustable and fixed collimating apertures before entering the magnetically shielded interaction region, where electric and magnetic fields are applied. The quantization axis is not preserved between the microwave region and the electric field plates so the population in the three M sub-levels of $|X; J = 1\rangle$ are mixed. A retroreflected 943 nm laser optically pumps population from the $|X; J = 1, M = \pm 1\rangle$ states to $|A; J = 0, M = 0\rangle$, which spontaneously decays partially into the $|H; J = 1\rangle$ state in which the EDM measurement is performed.

The spin precession region contains applied electric and magnetic fields, along with lasers to prepare and read our EDM state. The electric field is provided by two plates of 12.7 mm thick glass coated with a layer of indium tin oxide (ITO) on one side, and an anti-reflection coating on the other. The ITO coated sides of the plates face each other with a gap of 25 mm, and a voltage is applied to the ITO to create a uniform electric field.

The spatial profile of the electric field was measured by performing microwave spectroscopy on the ThO molecules. When the molecule pulse is between the state preparation and read-out regions, a 40 μ s burst of microwaves resonant with the DC Stark-shifted $|H; J = 1, M = \pm 1\rangle \rightarrow |H; J = 2, M = 0\rangle$ transitions is introduced by a microwave horn at the end of the apparatus, counterpropagating to the molecular beam. If on resonance, the microwaves drive a transition that spin-polarizes the molecules, similar to the state preparation scheme. We can then detect the spin polarization using the normal readout scheme. The microwave transition width is ~ 5 kHz (dominated by Doppler broadening), so the H -state dipole moment of $D \approx 1$ MHz/(V/cm)¹¹ (for $J = 1$) means that this method is sensitive to mV/cm electric field deviations with spatial resolution of 1 cm, limited by the velocity distribution in the beam. Our measurement indicated that the spatial variation of the electric field plate separation is ~ 20 μ m across the molecule precession region, in very good agreement with an interferometric measurement³². We can also test how well the electric field reverses by mapping the field with equal and opposite voltages on the plates. This measurement indicated that the non-reversing component of the electric field had magnitude $|\mathcal{E}^{\text{nr}}| \approx 1$ –5 mV/cm across the entire molecular precession region, as shown in Figure 3B.

The EDM measurement is performed in a vacuum chamber surrounded by five layers of mu-metal shielding. The applied magnetic field is supplied by a cosine-theta coil, with several shim coils to create a more uniform magnetic field within the precession region, and to allow us to apply transverse magnetic fields and gradients for systematic checks. Changes in the magnetic field are monitored by four 3-axis fluxgate magnetometers inside the magnetic shields, and the magnetic fields were mapped out before and after the experimental dataset was taken by sliding a 3-axis fluxgate down the beamline.

The lasers travel through the electric field plates, so all stages of the spin precession measurement are performed inside the uniform electric field. All laser light in the experiment originates from external cavity diode lasers (ECDL), frequency stabilized via an Invar transfer cavity to a CW Nd:YAG laser locked to a molecular iodine transition³³. All required transition frequencies and state assignments were determined previously^{34–36}. We measured the saturation intensities, radiative lifetimes, electric/magnetic dipole moments, and branching ratios for all required states and transitions.

In order to normalize against drifting molecular beam properties (pulse shape, total molecule number, velocity mean and distribution, etc.), we perform a spin precession measurement every 10 μ s, which is much faster than the molecular beam variations¹⁵, spin precession time, and temporal width of the molecular pulse. The ~ 20 μ s fly-through interaction time with the readout laser allows each molecule to be read-out by both \hat{X} and \hat{Y} polarizations. This is accomplished by sending the detection laser through two different beam paths, combined on the two ports of a polarizing beamsplitter. The two beam paths can be rapidly switched on and off with acousto-optic modulators (AOMs). The maximum rate of the polarization switching is limited by the 500 ns lifetime of the C state (decay rate of $\gamma \approx 2\pi \cdot 0.3$ MHz). A 1.2 μ s delay is inserted between the pulses of \hat{X} and \hat{Y} polarized readout light (Fig. S1A), which minimizes the amount of residual fluorescence overlapping between subsequent polarization states. Since the polarization switching period is longer than the decay time of the C state, we expect $\lesssim 1$ percent of the C state population to spontaneously decay back to the H state while the molecules are in the readout laser beam. Each of these two effects reduces the contrast by about 1 percent. We searched for, but did not observe, changes in ω^{NE} as a function of time within a polarization cycle.

The transparent electric field plates allow us to collect a large fraction of the solid angle of fluorescence from the molecules. Fluorescence travels through the field plates into an eight-lens system (four behind each plate) which focuses the light into an optical fiber bundle. The four bundles on each side are coupled into a fused quartz light pipe, which carries the fluorescence to a PMT (outside the magnetic shields). The net detection efficiency, including collection solid angle and detector quantum efficiency, is about 1%. We typically register around 1000 photon counts per molecule pulse (Fig. S1B). The PMT photocurrents are read as analog signals by a low-noise, high-bandwidth amplifier, and then sent to a 24-bit digitizer operating at 5 megasamples/s. The control and timing for all experimental parameters is managed by a single computer, and the timing jitter is less than one digitizer sampling period.

Systematic Errors

The presence of a nonzero magnetic field component B_z (parallel or antiparallel to the electric field), leads to a nonzero two photon detuning, $\delta = 2\mu_B g \tilde{B} |B_z|$, for the Λ system characterized

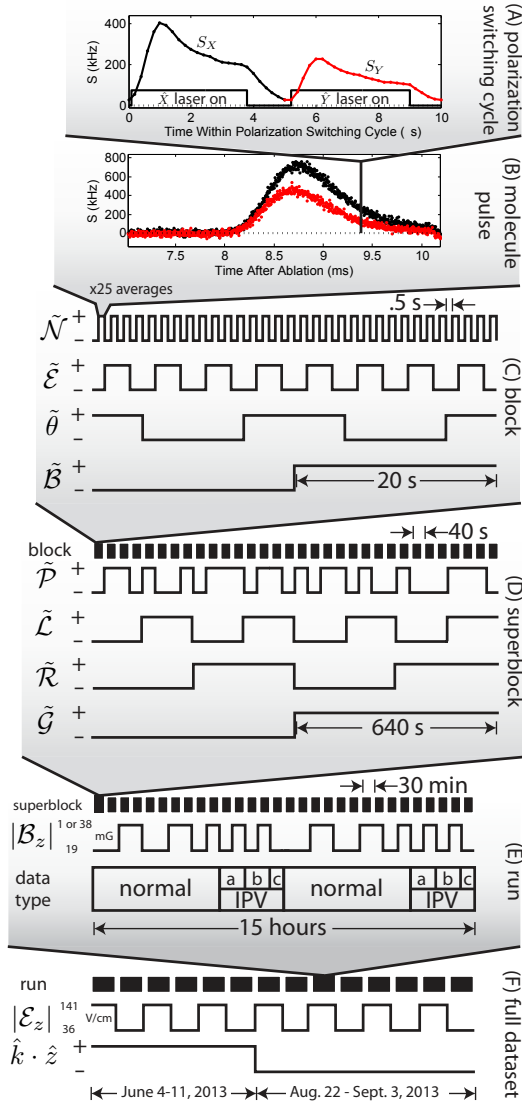


FIG. S1. **Relevant switching timescales** (A) Time within the readout laser polarization switching cycle with a period of 10 μ s. The molecule fluorescence rises rapidly when the laser is switched on, decays with rate $\gamma/2$ to a steady state (due to additional molecules entering the laser beam), and then decays with rate γ after the laser is switched off. Fluorescence is measured in detected photoelectrons/ms (kHz). (B) Time within the ThO molecule pulse after its creation by laser ablation. The fluorescence signals from (A) have been split up according to readout laser polarization to obtain the plotted points labeled by S_x and S_y . The displayed fluorescence traces are averaged over 400 pulses, but 25 pulses are averaged for a given state of the experiment. (C) Switches performed within a block of data. The \tilde{N} switch randomly alternates between a $(-+)$ and $(+-)$ pattern, the \tilde{E} and $\tilde{\theta}$ switches randomly alternate between a $(-++-)$ and $(+--+)$ pattern, and the \tilde{B} switch alternates between a $(-+)$ and $(+-)$ pattern between blocks. (D) Switches performed within a superblock of data. The \tilde{P} state is assigned randomly between blocks, but other superblock patterns are deterministic. (E) Changes in the experimental procedure between superblocks. We alternated between taking data under “normal” conditions and taking data with “intentional parameter variations” (IPV), which were used to monitor systematic effects. The IPVs are a= \mathcal{E}^{nr} , b= Ω_r^{NE} , and c= Δ_{prep} (which was used to monitor the value of \mathcal{E}^{nr} by exaggerating its effect on the $\tilde{N}\tilde{E}$ correlated contrast). (F) An overview of the ~ 2 weeks of data comprising our reported EDM measurement.

Category I Parameters

Magnetic Fields

- Non-Reversing \mathcal{B} -Field: $\mathcal{B}_z^{\text{nr}}$
- Transverse \mathcal{B} -Fields: $\mathcal{B}_x, \mathcal{B}_y$ (even and odd under \tilde{B})
- Magnetic \mathcal{B} -Field Gradients: $\frac{\partial \mathcal{B}_x}{\partial x}, \frac{\partial \mathcal{B}_y}{\partial x}, \frac{\partial \mathcal{B}_y}{\partial y}, \frac{\partial \mathcal{B}_y}{\partial z}, \frac{\partial \mathcal{B}_z}{\partial x}, \frac{\partial \mathcal{B}_z}{\partial z}$ (even and odd under \tilde{B})
- \tilde{E} correlated \mathcal{B} -field: $\mathcal{B}^{\tilde{E}}$ (to simulate $\vec{v} \times \vec{E}$ /geometric phase/leakage current effects)

Electric Fields

- Non-Reversing \mathcal{E} -Field: \mathcal{E}^{nr}
- \mathcal{E} -Field Ground Offset

Laser Detunings

- Detuning of the Prep/Read Lasers: $\Delta_{\text{prep}}, \Delta_{\text{read}}$
- \tilde{P} correlated Detuning: $\Delta^{\tilde{P}}$
- \tilde{N} correlated Detunings: $\Delta^{\tilde{N}}, \Delta\Delta^{\tilde{N}}$

Laser Pointings along \hat{x}

- Change in Pointing of Prep/Read Lasers
- Readout laser \hat{X}/\hat{Y} dependent pointing
- \tilde{N} correlated laser pointing
- \tilde{N} and \hat{X}/\hat{Y} dependent laser pointing

Laser Powers

- Power of Prep/Read Lasers: $P_{\text{prep}}, P_{\text{read}}$
- $\tilde{N}\tilde{E}$ correlated power, $P^{\tilde{N}\tilde{E}}$ (simulating $\Omega_r^{\tilde{N}\tilde{E}}$)
- \tilde{N} correlated power, $P^{\tilde{N}}$
- \hat{X}/\hat{Y} dependent Readout laser power

Laser Polarization

- Preparation Laser Ellipticity

Molecular Beam Clipping

- Molecule Beam Clipping along the \hat{y} and \hat{z} (changes $\langle v_y \rangle, \langle v_z \rangle, \langle y \rangle, \langle z \rangle$ for molecule ensemble)

Category II Parameters

Experiment Timing

- \hat{X}/\hat{Y} Polarization Switching Rate
- Number of Molecule Pulse Averages contributing to an Experiment State

Analysis

- Signal size cuts, Asymmetry magnitude cuts, Contrast cuts
- Difference between two PMT detectors (checking spatial fluorescence region dependence)
- Variation with time within molecule pulse (serves to check v_x dependence)
- Variation with time within polarization switching cycle
- Variation with time throughout the full dataset (autocorrelation)
- Search for correlations with all ϕ, C , and S switch-parity components
- Search for correlations with auxiliary measurements of \mathcal{B} -fields, laser powers, and vacuum pressure
- 3 individuals performed independent analyses of the data

TABLE S1. **Parameters varied in the search for systematic errors.** Category I: Parameters that were varied far from their values under normal conditions of the experiment. For each of these parameters direct measurements or limits were placed on possible systematic errors that could couple linearly to each by the method described in the main text. Category II: Parameters for which all values are considered consistent with normal conditions of the experiment. Although direct limits on systematic errors cannot be derived, these served as checks for the presence of unanticipated systematic errors.

by $|H, \tilde{N}, M = +1\rangle \leftrightarrow |C, \tilde{P}, M = 0\rangle \leftrightarrow |H, \tilde{N}, M = -1\rangle$. Such a Λ system has a dark eigenstate (a state that has zero excited state $|C, \tilde{P}, M = 0\rangle$ amplitude) in the limit of $\delta = 0$, but for $\delta \neq 0$, all eigenstates have nonzero excited state amplitude. In the limit of

small $\delta/\Omega_r \ll 1$ (in our case, $\delta/\Omega_r \sim 10^{-3}$), the introduction of the magnetic field mixes the bright and dark states with amplitudes proportional to δ/Ω_r . The bright state amplitude acquires an AC Stark shift and results in a change in the measured phase that is correlated with the magnetic field direction,

$$\phi_{AC}^B(\Delta, \Omega_r) = \alpha^B \Delta^2 + \beta^B \Omega_r, \quad (S1)$$

where α^B and β^B are proportional to $|\mathcal{B}_z|$ and depend on the spatial profile of the laser. This model was verified and these coefficients were measured directly from ϕ^B by varying Δ and Ω_r with AOMs.

The coupling of the $\Delta^{\mathcal{N}\mathcal{E}}$ and $\Omega_r^{\mathcal{N}\mathcal{E}}$ to this $\tilde{\mathcal{B}}$ -odd AC Stark shift-induced phase leads to contributions to $\phi^{\mathcal{N}\mathcal{E}B}$:

$$\phi^{\mathcal{N}\mathcal{E}B} = 2\alpha^B \Delta \Delta^{\mathcal{N}\mathcal{E}} + \beta^B \Omega_r^{\mathcal{N}\mathcal{E}}. \quad (S2)$$

This phase is dominated by the $\beta^B \Omega_r^{\mathcal{N}\mathcal{E}}$ term since we operate the experiment on resonance, $\Delta \approx 0$; this was verified by observing that $\phi^{\mathcal{N}\mathcal{E}B}$ reversed sign with $\hat{k} \cdot \hat{z}$ (since $\Omega_r^{\mathcal{N}\mathcal{E}} \propto \hat{k} \cdot \hat{z}$). We used this effect to our advantage to measure the value of $\Omega_r^{\mathcal{N}\mathcal{E}} = \phi^{\mathcal{N}\mathcal{E}B}/\beta^B$ in our system. We measured $\phi^{\mathcal{N}\mathcal{E}B}$ from our EDM dataset, and we measured $\beta^B = \partial\phi^{\mathcal{N}\mathcal{E}B}/\partial\Omega_r^{\mathcal{N}\mathcal{E}}$ by intentionally correlating the laser power of the state preparation and read-out lasers with $\tilde{\mathcal{N}}\tilde{\mathcal{E}}$ using AOMs.

The \mathcal{E}^{nr} and $\Omega_r^{\mathcal{N}\mathcal{E}}$ systematics, which result from AC Stark shift induced phases, were sensitive to the spatial intensity profile and polarization gradients in the prep and readout lasers. A sharper edge to the laser intensity profile reduces the size of the region where the AC stark shift phase accumulates, therefore reducing the systematic slopes proportional to α and β . The dependence on the spatial intensity profile was confirmed by clipping our Gaussian laser beam profile with a razor edge. This data agreed with numerical simulations of the Schrödinger equation under varying spatial intensity profiles. To vary the polarization gradients, an optical chopping wheel was added on the state preparation laser beam, reducing the time averaged energy deposited in the field

plates and hence also the thermally induced birefringence. As expected, the slope of the \mathcal{E}^{nr} systematic was also reduced by half.

The two $\tilde{\mathcal{N}}$ states in which we perform our EDM measurement have magnetic moments differing by about 0.1 percent¹². This difference is proportional to $|\mathcal{E}_z|$ and is the main contribution to $\phi^{\mathcal{N}B}$. Therefore, any effect coupling to the magnetic moment to systematically shift $\phi^{\mathcal{E}}$ will also produce a 1000-times smaller shift in $\phi^{\mathcal{N}\mathcal{E}}$. We verified this by intentionally correlating a 1.4 mG component of \mathcal{B}_z with $\tilde{\mathcal{E}}$, resulting in a large offset of $\phi^{\mathcal{E}}$ and a 1000-times smaller offset of $\phi^{\mathcal{N}\mathcal{E}}$, as expected. Although $\phi^{\mathcal{E}}$ shifts caused by leakage current, $\vec{v} \times \tilde{\mathcal{E}}$, and geometric phase effects were observed in past experiments⁴, we expect to be immune to such effects at our current level of sensitivity¹⁰. Indeed, the measured $\phi^{\mathcal{E}}$ was consistent with zero for our reported data set. The mean and uncertainty of $\phi^{\mathcal{E}}$, divided by the measured suppression factor, is included in our $\phi^{\mathcal{N}\mathcal{E}}$ systematic error budget (see Table 1).

Data was stored and analyzed as a function of time after ablation and time within a polarization switch state. Due to the 10 percent longitudinal velocity dispersion of our molecule beam, the arrival time at our detectors is correlated with different longitudinal velocity classes, and therefore different precession times τ . We did not see any variation in the measured phases $\phi^{\mathcal{E}}$ or $\phi^{\mathcal{N}\mathcal{E}}$ as a function of time after ablation.

Outlook

It is possible to further reduce this experiment's statistical and systematic uncertainty. In a separate apparatus we have demonstrated that electrostatic molecule focusing and EDM state preparation via Stimulated Raman Adiabatic Passage can combine to increase our fluorescence signal by a factor of ~ 100 . Also, a thermochemical beam source may increase our molecule flux by a factor of ~ 10 . The combined signal increase may reduce our statistical uncertainty by a factor of $\gtrsim 10$. The dominant AC Stark shift systematic errors can be further suppressed by implementing electric field plates with improved thermal and optical properties.

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