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SUPPLEMENTARY INFORMATION

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Improved limit on the electric dipole moment of the electron

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SUPPLEMENTARY METHODS

Description of Mechanisms Causing Systematic Error

We describe in detail the mechanisms behind the systematic shifts described in the main text, starting with shifts which are proportional to magnetic field gradients. We describe the systematic shift due to $\partial\mathcal{B}_z/\partial z$ associated with a translation of the center of mass of the molecules that are successfully prepared by STIRAP, which is the dominant contribution (the systematic effect is entirely analogous for both $\partial\mathcal{B}_z/\partial z$ and $\partial\mathcal{B}_z/\partial y$ gradients). We nominally tune the STIRAP 2-photon detuning, δ, to resonance ($\delta = 0$), where the STIRAP transfer efficiency, η, is maximal ($\eta(\delta = 0) = \eta_0 \approx 75\%$). When $\delta = 0$, by definition the derivative of the transfer efficiency vs. detuning is $\partial\eta/\partial\delta = 0$. However, if slightly off resonance, $\partial\eta/\partial\delta$ will be nonzero: $\partial\eta/\partial\delta \propto \delta$ (for small δ). A small change in the 2-photon detuning, $\mathrm{d}\delta$, can thus lead to a change in the transfer efficiency given by $\mathrm{d}\eta = \partial\eta/\partial\delta \cdot \mathrm{d}\delta$. Spatial dependence of δ on z, $\partial\delta/\partial z$, will then cause a z-dependence of η, described by

$$\eta(z) = \frac{\partial\eta}{\partial\delta}\frac{\partial\delta}{\partial z}z + \eta_0. \tag{1}$$

Such a dependence of δ on z can arise from different sources, including, for example, spatially inhomogeneous $\mathcal{E}$-fields that Stark shift the molecular resonance. In a simple model with a constant molecular density along z, this effect shifts the center of mass of the molecules that are successfully prepared by the STIRAP lasers along z by

$$\mathrm{d}z_{\mathrm{CM}} = \frac{a^2}{3\eta_0}\frac{\partial\eta}{\partial\delta}\frac{\partial\delta}{\partial z}, \tag{2}$$

where a is the half-width of the molecular beam in z. Such translations in molecular beam position, in the presence of a $\mathcal{B}$-field gradient, cause a shift $\mathrm{d}\omega \propto \partial\mathcal{B}_z/\partial z \cdot \mathrm{d}z_{\mathrm{CM}}$ in the precession frequency. If $\partial\delta/\partial z$ has a component correlated with $\tilde{\mathcal{N}}\tilde{\mathcal{E}}$, then $\partial\delta^{\mathcal{NE}}/\partial z$ can lead to a systematic error in the measurement of $\omega^{\mathcal{NE}}$.

We have observed that a nonzero $\partial\delta^{\mathcal{NE}}/\partial z$ can be caused by a gradient in the non-reversing $\mathcal{E}$-field component along z, $\partial\mathcal{E}^{\mathrm{nr}}/\partial z$. $\mathcal{E}^{\mathrm{nr}}$ and its gradients can be generated by patch potentials and technical voltage offsets. These non-reversing $\mathcal{E}$-field components cause $\tilde{\mathcal{N}}\tilde{\mathcal{E}}$-correlated Stark shifts with associated laser detunings $\delta^{\mathcal{NE}} = D\mathcal{E}^{\mathrm{nr}}$, where D is the H state electric dipole moment (Fig. 1a). While we can exaggerate $\mathcal{E}^{\mathrm{nr}}$ by applying an asymmetric voltage on the $\mathcal{E}$-field plates, we have no experimental means to apply $\mathcal{E}^{\mathrm{nr}}$ gradients. However, since this systematic depends on three imperfections ($\partial\mathcal{E}^{\mathrm{nr}}/\partial z$, δ, and $\partial\mathcal{B}_z/\partial z$), amplifying any one of these individually allows us to accurately find settings that null the product of the other two imperfection-related parameters. Then, under ordinary run conditions (when all controllable parameters are nulled), the product of the residual imperfections is (at least) second-order small.

By intentionally varying δ and deliberately applying a nonzero $\partial\mathcal{B}_z/\partial z$, we verified our model for how these parameters cause a false shift in $\omega^{\mathcal{NE}}$ (Extended Data Fig. 2c). This model was further confirmed by our observation of an $\tilde{\mathcal{N}}\tilde{\mathcal{E}}$-correlated z-dependent fraction of the fluorescence signal (seen by grouping the PMT signal by spatial location ($+z$ or $-z$)). This independently confirmed the $\tilde{\mathcal{N}}\tilde{\mathcal{E}}$-correlated translation in z, $\mathrm{d}z_{\mathrm{CM}}^{\mathcal{NE}}$, when we amplify the effect by applying deliberate nonzero values of δ.

The second contribution to the $\mathcal{B}$-field gradients class of systematic errors comes from a coupling of the $\mathcal{B}$-field gradients to a translation of the detected molecular population that is related to an $\mathcal{E}^{\mathrm{nr}}$ offset, rather than a gradient in $\mathcal{E}^{\mathrm{nr}}$. We focus our discussion on the effect in the readout beam, which comes from a translation of the molecular beam in z, the propagation direction of the readout laser, therefore coupling to a $\partial\mathcal{B}_z/\partial z$ gradient only (the effect is analogous for $\partial\mathcal{B}_z/\partial y$ in the STIRAP laser beams, which propagate along $\hat{y}$).

After leaving the beam source, molecules travel ballistically through the precession region. Since the molecule beam diverges from the source (effective width ∼6 mm) to the size of the molecular beam in the precession region (where collimators define the beam width to be ∼25 mm) over a distance of 1.1 m, there is a strong correlation (> 0.9 correlation coefficient) between the z position of the molecules and their transverse velocity, $v_z \propto z$. Finite transverse velocity causes a Doppler shift for the readout laser, $\Delta_z^{\mathrm{Dopp}} \propto v_z$, so there is also a correlation between the detuning experienced by a molecule and its position: $\partial\Delta_z^{\mathrm{Dopp}}/\partial z = (\partial\Delta_z^{\mathrm{Dopp}}/\partial v_z)(\partial v_z/\partial z)$. This in turn means that a change in laser detuning, $\mathrm{d}\Delta$, leads to a translation of the detected molecular beam in z, given by $dz_{\mathrm{CM}} = (\partial z/\partial\Delta_z^{\mathrm{Dopp}})\mathrm{d}\Delta$. Such a translation, in concert with a nonzero $\partial\mathcal{B}_z/\partial z$, causes a shift in the precession frequency, $d\omega = -\mu(\partial\mathcal{B}_z/\partial z)(\mathrm{d}z_{\mathrm{CM}})$. Finally, this can lead to a systematic shift in the EDM frequency, $\omega^{\mathcal{NE}}$, if some mechanism leads to an $\mathcal{NE}$-correlated detuning, $\Delta^{\mathcal{NE}}$. This type of correlated detuning can be caused by an overall $\mathcal{E}^{\mathrm{nr}}$ offset, $\Delta^{\mathcal{NE}} = D\mathcal{E}^{\mathrm{nr}}$ (Fig. 1a).

To confirm our model, we intentionally exaggerated $\mathcal{E}^{\mathrm{nr}}$ and verified that it couples to a deliberately applied $\partial\mathcal{B}_z/\partial z$ to cause a shift in the $\omega^{\mathcal{NE}}$ channel, as just described. In addition, we independently verified the coupling of the detected molecule translation, $\mathrm{d}z_{\mathrm{CM}}$, to $\partial\mathcal{B}_z/\partial z$. In particular, we observed a dependence of ω on readout laser detuning, $\mathrm{d}\omega = (\partial\omega/\partial\Delta)\Delta$, when a $\partial\mathcal{B}_z/\partial z$ was applied and the laser detuning Δ was varied. Numerical simulation of the effect agreed with its measured magnitude. Monitoring and suppression methods are described in the main text.

Under typical run parameters, the Rabi frequencies of the Pump and Stokes lasers are kept equal and the STIRAP 2-photon resonance lineshape is reasonably well approximated by a Gaussian[1]. The fits in Extended Data Fig. 2c are for the local slope of the nearly-Gaussian STIRAP 2-photon lineshape, $L(\delta) \equiv \partial\omega^{\mathcal{NE}}/\partial\delta = -c \cdot \delta \cdot \mathrm{Exp}(-\delta^2/(2\sigma_{\mathrm{ST}}^2))$, where the free fit parameters are σ_{ST} and the scaling factor $c \propto a^2/(3\eta_0) \cdot \partial\mathcal{E}^{\mathrm{nr}}/\partial z \cdot \partial\mathcal{B}_z/\partial z$.

A second experiment imperfection that causes an EDM shift is birefringence in the $H - C$ STIRAP laser beam.This can occur if this beam's original polarization axis does not lie along the axis of undesired birefringence of optical elements in the path of the laser beam (e.g., vacuum chamber windows or

beam shaping optics) and if this birefringence is not spatially uniform across the area of the laser beam.

This ellipticity gradient causes a shift in the precession frequency, $\omega_{\rm ST}$, due to AC Stark shift effects. This frequency shift, which is linear in $\theta_{\rm ST}^{H-C}$ for small angles, in turn is proportional to the STIRAP 2-photon detuning, δ. If δ has an $\mathcal{NE}$-correlated component, $\delta^{\mathcal{NE}}$, this effect will cause a STIRAP-prepared $\tilde{\mathcal{N}}\tilde{\mathcal{E}}$-correlated spin precession frequency component, $\omega_{\rm ST}^{\mathcal{NE}} = (\partial\omega_{ST}^{\mathcal{NE}}/\partial\theta_{\rm ST}^{H-C})\cdot\theta_{\rm ST}^{H-C}$. (This is similar to one of the dominant systematic effects in ACME I[2,3].) The $\mathcal{NE}$-correlated detuning component, $\delta^{\mathcal{NE}}$ can arise from $\mathcal{E}^{\rm nr}$, which causes $\tilde{\mathcal{N}}\tilde{\mathcal{E}}$-correlated Stark shifts with associated laser detunings $\delta^{\mathcal{NE}} = D\mathcal{E}^{\rm nr}$, where D is the H state electric dipole moment (Fig. 1a).

The refinement laser minimizes the slope $S_{\theta_{\rm ST}^{H-C}} = \partial\omega^{\mathcal{NE}}/\partial\theta_{\rm ST}^{H-C}$ by reprojecting the STIRAP-prepared spin alignment, $\vec{S}_{\rm ST}$, along the polarization axis of the refinement beam, $\vec{\epsilon}_{\rm ref}$. The slope is given by $S_{\theta_{\rm ST}^{H-C}} = (\partial\omega_{\rm ST}^{\mathcal{NE}}/\partial\theta_{\rm ST}^{H-C})/A_{\rm ref}$, where $A_{\rm ref} = 1/(\partial\omega^{\mathcal{NE}}/\partial\omega_{\rm ST}^{\mathcal{NE}})$ is a factor we refer to as the refinement attenuation. Its value depends on the properties of the refinement laser beam (power, spatial profile, and detuning).

A third shift in the EDM is due to the $P^{\mathcal{NE}}$ parameter. We typically attempt to perfectly align $\vec{\epsilon}_{\rm ref}$ with $\vec{S}_{\rm ST}$. However, if the two vectors are misaligned by an angle $\theta_{\rm ST}^{\rm ref}$, the component of $\vec{S}_{\rm ST}$ orthogonal to $\vec{\epsilon}_{\rm ref}$ is reduced only by the factor $A_{\rm ref}$; then the post-refinement spin alignment $\vec{S}$ will deviate from the ideal $\vec{\epsilon}_{\rm ref}$ axis by an angle $\approx \theta_{\rm ST}^{\rm ref}/A_{\rm ref}$. Since $A_{\rm ref}$ is dependent on the power of the refinement laser, $P_{\rm ref}$, a variation of its power, $\mathrm{d}P_{\rm ref}$, causes a shift in the prepared phase: $\mathrm{d}\omega_{\rm ref} = -(\theta_{\rm ST}^{\rm ref}/A_{\rm ref}^2)(\partial A_{\rm ref}/\partial P_{\rm ref})\mathrm{d}P_{\rm ref}/\tau$. Then, if $P_{\rm ref}$ has an $\mathcal{NE}$-correlated component, $P_{\rm ref}^{\mathcal{NE}}$, this mechanism will cause a shift in $\omega^{\mathcal{NE}}$. To confirm this model, we verified that when deliberately applying a $P_{\rm ref}^{\mathcal{NE}}$, in the presence of a large $\theta_{\rm ST}^{\rm ref}$, we observe the expected slope $S_{P_{\rm ref}^{\mathcal{NE}}} = \partial\omega^{\mathcal{NE}}/\partial P_{\rm ref}^{\mathcal{NE}}$.

Another contribution to the systematic error bar comes from correlations between $\omega^{\mathcal{NE}}$ and contrast $\mathcal{C}$. We calculate the EDM frequency by extracting the $\mathcal{NE}$-correlated component from the ratio of asymmetry to contrast, $\omega^{\mathcal{NE}} \approx [\mathcal{A}/(2\mathcal{C})]^{\mathcal{NE}}/\tau$. A first-order expansion of $\omega^{\mathcal{NE}}$ in terms of the individual parity components of all associated quantities results in

$$\omega^{\mathcal{NE}} \approx \frac{(\mathcal{A}\ \mathrm{sgn}(\mathcal{C}))^{\mathcal{NE}}}{2\tau|\mathcal{C}|^{\rm nr}} - \omega^{\mathcal{B}}\frac{|\mathcal{C}|^{\mathcal{NEB}}}{|\mathcal{C}|^{\rm nr}} - \omega^{\rm nr}\frac{|\mathcal{C}|^{\mathcal{NE}}}{|\mathcal{C}|^{\rm nr}} + ..., \quad (3)$$

where we used the fact that $|\mathcal{C}|^{\rm nr} \gg |\mathcal{C}|^u$ for any other parity component u. We looked for possible contributions from any other phase parity component channels, ω^u (not only those shown in Eq. 3), by searching the EDM dataset for nonzero correlations dataset between $\omega^{\mathcal{NE}}$ and $|\mathcal{C}|^u$, quantified by a nonzero value of the slope $S_{|\mathcal{C}|^u} = \partial\omega^{\mathcal{NE}}/\partial|\mathcal{C}|^u$. We have observed such correlations only with two channels: $|\mathcal{C}|^{\mathcal{NE}}$ and $|\mathcal{C}|^{\mathcal{NEB}}$. This is consistent with the fact that only $\omega^{\mathcal{B}}$ and $\omega^{\rm nr}$ have large values; these are due to $\mathcal{B}$-field precession and global offset phases, respectively.

We also describe effects that were not observed to shift $\omega^{\mathcal{NE}}$, but have an effect on the other parity components. The $\tilde{\mathcal{P}}$- and $\tilde{\mathcal{R}}$- odd components of the frequency, $\omega^{u\mathcal{PR}}$, contain understood nonzero offsets that were observed and explained in ACME I[3]. These nonzero components result from differences in the properties (spatial profile, power and pointing) of the $\hat{X}$ and $\hat{Y}$ readout beams and are proportional to correlated detuning components Δ^u. The $\tilde{\mathcal{P}}$ and $\tilde{\mathcal{R}}$ switches, which interchange the roles of the $\hat{X}$ and $\hat{Y}$ beams, prevent offsets in $\omega^{u\mathcal{PR}}$ from contaminating ω^u. When coupled to $\Delta^{\mathcal{NE}}$ (from $\mathcal{E}^{\rm nr}$), the residual differences in the $\hat{X}$ and $\hat{Y}$ readout beams create $\sim 8\sigma$ offsets in the $\omega^{\mathcal{NEPR}}$ channel. We verified numerically that the value of $\omega^{\mathcal{NEPR}}$ is consistent with our model, given the measured size of experimental imperfections that it couples to (pointing between $\hat{X}$ and $\hat{Y}$, $\mathcal{E}^{\rm nr}$). By putting bounds on the possible size of correlations between $\omega^{\mathcal{NEPR}}$ and $\omega^{\mathcal{NEP}}$ (i.e., $\partial\omega^{\mathcal{NEPR}}/\partial\omega^{\mathcal{NEP}}$), and between $\omega^{\mathcal{NEPR}}$ and $\omega^{\mathcal{NER}}$ (i.e. $\partial\omega^{\mathcal{NEPR}}/\partial\omega^{\mathcal{NER}}$), we measured a suppression factor of possible leakage from $\omega^{\mathcal{NEPR}}$ to $\omega^{\mathcal{NE}}$ of > 500. Systematic checks in which we deliberately exaggerate $\omega^{\mathcal{NEPR}}$ by increasing the difference in the parameters between the $\hat{X}$ and $\hat{Y}$ beams (power, pointing asymmetry) give similar or better bounds. The systematic contribution due to $\omega^{\mathcal{NEPR}}$ is therefore limited to < 10 μrad/s. Including this term in the systematic error budget would increase its uncertainty by less than 1%. We did not include it, however, since we did not see direct evidence of these effects shifting $\omega^{\mathcal{NE}}$ near current sensitivity.

In ACME I, we included in the systematic error budget a contribution from unexplained variations in the $\tilde{\mathcal{N}}$-correlated frequency channel, $\omega^{\mathcal{N}}$[2,3]. Such variations are reduced in magnitude in ACME II. Moreover, they are now understood to be caused by a time-varying relative detuning between the two $\tilde{\mathcal{N}}$ states, $\Delta^{\mathcal{N}}$, coupling to differences in laser beam properties (pointing, power, spatial) of the two probe ($\hat{X}$ and $\hat{Y}$) laser beams[3]. These effects are removed by the $\tilde{\mathcal{P}}$ and $\tilde{\mathcal{R}}$ switches, but will cause noise in the $\omega^{\mathcal{N}}$ channel if the timescale of the $\Delta^{\mathcal{N}}$ variation is faster than that of the $\tilde{\mathcal{P}}$ and $\tilde{\mathcal{R}}$ switches. Such noise is caused primarily by $\sim$1–100 ms excursions in the frequency of the Ti:S readout laser, with magnitude $\Delta^{\mathcal{N}} \sim 400$–$600$ kHz. To ensure this noise does not contribute to $\omega^{\mathcal{NE}}$, we verified that the correlation coefficient between $\omega^{\mathcal{NE}}$ and $\omega^{\mathcal{N}}$ (i.e. $\partial\omega^{\mathcal{NE}}/\partial\omega^{\mathcal{N}}$) for the EDM dataset is consistent with zero.

In ACME I, the leading source of systematic error came from an AC Stark shift effect, associated with an ellipticity gradient across the area of a laser beam, and coupling to the non-reversing component of the electric field ($\mathcal{E}^{\rm nr}$). A thermal stress-induced birefringence gradient, caused by absorption of the laser power by the glass electric field plates and vacuum chamber windows, was responsible for the large ellipticity gradient ($\sim 10\%$/mm circular polarization fraction)[2,3]. ACME II uses redesigned field plates and vacuum windows, with Corning 7980 fused silica instead of Schott Borofloat glass (for an order of magnitude lower thermal expansion and reduced bulk laser absorption). The field plates are coated with a thinner layer of conductive ITO (20 nm compared to 200 nm in ACME I), which reduces thermal absorption. In addition, the refinement and readout excitations are performed on a stronger transition, $H - I$, than that used in

ACME I, $H - C$, which requires significantly less laser power (800 mW compared to 4 W in ACME I). We verified through direct polarimetry that thermal stress-induced birefringence gradients were reduced to below $0.1\%/\mathrm{mm}$[4].

In ACME I, we observed an $\tilde{\mathcal{N}}\tilde{\mathcal{E}}$-correlated Rabi frequency component, $\Omega_{\mathrm{r}}^{\mathcal{NE}}$, which was correlated with the laser propagation direction, $\hat{k} \cdot \hat{z}$[3]. In ACME II, we measured this effect by propagating the laser beams in both $\hat{k} \cdot \hat{z} = \pm 1$ configurations and found no significant $\Omega_{\mathrm{r}}^{\mathcal{NE}}$ component. The difference may be due to the use of a different refinement and probe transition than in ACME I. However, when $\hat{k} \cdot \hat{z} = -1$ (but not $+1$), we found a nonzero value of $\partial\omega^{\mathcal{NE}}/\partial\mathcal{E}^{\mathrm{nr}}$. This slope could be explained[3] by the presence of small ($\sim 1\%/\mathrm{mm}$) ellipticity gradients, caused by mechanical stress birefringence from mounting of the electric field plates and/or vacuum chamber windows, on one side of the apparatus but not the other. Unlike in ACME I, we were not able to minimize this effect by aligning the polarization of the lasers with the birefringence axis of the optics, since the spin alignment is fixed by the STIRAP beams to be along x. While the $\omega^{\mathcal{NE}}$ value was consistent for both $\hat{k} \cdot \hat{z}$ configurations, the larger $\partial\omega^{\mathcal{NE}}/\partial\mathcal{E}^{\mathrm{nr}}$ slope significantly increases (factor of ~ 5) the $\mathcal{E}^{\mathrm{nr}}$ systematic uncertainty for $\hat{k} \cdot \hat{z} = -1$. Hence, in ACME II, we treat $\hat{k} \cdot \hat{z} = -1$ as a parameter variation consistency check and acquire the final EDM dataset only with $\hat{k} \cdot \hat{z} = +1$.

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