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Supplementary Materials for

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

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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 $B_{\rm R} \approx 10$ 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 sublevels 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 µs 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 µm across the molecule precession region, in very good agreement with an interferometric measurement[32]. 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 $|\mathcal{E}^{\rm nr}| \approx$ 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 transition[33]. 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 µs, which is much faster than the molecular beam variations[15], spin precession time, and temporal width of the molecular pulse. The ~ 20 µs 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 µs 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 $\omega^{\mathcal{N}\mathcal{E}}$ 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 $\mathcal{B}_z$ (parallel or antiparallel to the electric field), leads to a nonzero two photon detuning, $\delta = 2\mu_B g\tilde{\mathcal{B}}\,|\mathcal{B}_z|$, for the Λ system characterized