TOWARDS A NEW MEASUREMENT OF THE ELECTRON’S ELECTRIC DIPOLE MOMENT

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We present our progress towards a new measurement of the electron electric dipole moment using a beam of YbF molecules. Data are currently being taken with a sensitivity of $10^{-27}$ e.cm/$\sqrt{\text{day}}$.

1. Motivation and historical perspective

The most precise electric dipole moment (edm) measurement\(^1\) on the electron gives $d_e = (6.9 \pm 7.4) \times 10^{-28} \text{ e.cm}$. Although the Standard Model predicts a far smaller value, many modern extensions of particle theory lead quite naturally to a value in the range of $10^{-27}$ e.cm or a little below\(^2\). Our experiment, which aims to be more sensitive than this, is therefore a search for new physics. An edm at this level would also demonstrate a new type of CP violation, beyond the usual CKM mechanism, as is required to understand the matter-antimatter asymmetry of the universe\(^3\).

The interaction between $d_e$ and an applied field $E$ can be expressed by the effective non-relativistic Hamiltonian $-d_e \alpha(E) \hat{\sigma} \cdot E$. For a free electron, $\alpha(E) = 1$ and $\hat{\sigma}$ is a unit vector along the spin. If the electron is part of an atom or molecule, $\hat{\sigma}$ lies along the spin of the system and $\alpha(E)$ is a factor that depends on the structure. Some heavy atoms and molecules have the virtue that $\alpha(E) \gg 1$, and then it is called the enhancement factor\(^4\). This coupling resembles the interaction $-\mu \beta(B) \hat{\sigma} \cdot B$ of the magnetic moment $\mu$ with a magnetic field $B$, where $\beta(B)$ accounts for the atomic or molecular structure. It is instructive to compare these two interactions in the case of a free electron with an edm of, say, $d_e = 5 \times 10^{-28} \text{ e.cm}$, just below the present limit. In a 100 kV/cm field the edm energy is so small that it equals the magnetic energy in a field of only $9 \times 10^{-19} \text{ T}$. Controlling the stray mag-
netic field at that level seems close to impossible, especially when applying the electric field. Heavy atoms such as Cs and Tl alleviate this problem by their large enhancement factors. In particular, $\alpha(E) = -585$ for the thallium atom, which relaxes the necessary field control to the challenging, but achievable $fT$ level. Two magnetic effects are most troublesome. (i) Stray magnetic fields vary both in space and time. (ii) Atoms moving through the large electric field experience a motional magnetic interaction $-\mu \hat{\sigma} \cdot \mathbf{E} \times \mathbf{v}/c^2$. In both cases the unwanted field components are typically many orders of magnitude larger than $fT$ and heroic efforts were needed to reach the current precision.

2. Measuring with molecules

Heavy polar molecules offer substantial relief from these difficulties. First, the enhancement factors are generically much larger because the electron edm interacts with the polarisation of the charge cloud close to the heavy nucleus. In an atom this polarisation follows from the mixing of higher electronic states by the applied electric field. In a polar molecule, these electronic states are already strongly mixed by the chemical bond and it is only rotational states that have to be mixed by the applied field. Since these are typically a thousand times closer in energy, the molecular enhancement factor is larger. For the YbF molecule used in our experiment, the enhancement factor is $\alpha \simeq 10^6$ at our operating field of 13 kV/cm, which relaxes the requirement on field control to the $pT$ level.

There is a second advantage to YbF. Being polar, this molecule has a strong tensor Stark splitting between the Zeeman sublevels, making the applied electric field the natural quantization axis. As a result, the Zeeman shift associated with a perpendicular magnetic field is strongly suppressed, making the molecule insensitive to the motional field. For our typical operating parameters the motion-induced false edm is below $10^{-33}$ e.cm, which is entirely negligible.

3. YbF spin interferometer

Our experiment uses a cold, pulsed, supersonic beam of YbF radicals in a magnetically shielded vertical vacuum chamber $\sim 1.5$ m high. The electronic, rotational and vibrational ground state $X^2\Sigma^+_1/2, N = 0, v = 0$ is a hyperfine doublet with states $F = 0, 1$ split by $170 \text{MHz}$. We first

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*a or any system whose tensor Stark splitting greatly exceeds the Zeeman interaction.*
deplete the $F = 1$ state by laser excitation of the $F = 1$ molecules on the $A_{1/2} \leftarrow X$ transition. This laser beam is called the pump. An oscillating field, which we call the first beam splitter, then drives the $F = 0$ molecules into a symmetric coherent superposition of the $F = 1, m_F = \pm 1$ states, as described later in more detail. Next, parallel dc electric and magnetic fields are applied to introduce a phase shift $\Delta \phi = \frac{\hbar}{\pi} \int_0^\tau (dE(t) + \mu \beta(B(t)) dt$ between the two superposed states. Here $E$ and $B$ appear as functions of time because they are the fields in the molecular rest frame. At time $\tau$ the molecules interact with another oscillating field, the recombining beam-splitter, that couples the symmetric part of the $F = 1$ coherence back to the $F = 0$ state. The resulting $F = 0$ state population exhibits the usual $\cos^2 \left( \frac{\Delta \phi}{2} \right)$ fringes of an interferometer. We detect the $F = 1$ population using fluorescence induced by a probe laser on the $A_{1/2} \leftarrow X$ transition. Figure 1 shows the interference fringes observed in this fluorescence when the magnetic field is scanned.

4. The beam splitters

We describe two types of beam splitter, both designed to minimise the distance moved by the molecules whilst they are being split. This is important

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b. The direction of these fields defines the quantisation axis of our basis.

c. Variation of field directions can generate geometric phases, neglected here for simplicity.
4.1. Radio frequency splitter

The radio frequency beam splitter is an rf magnetic field perpendicular to $\mathbf{E}$ along the beam direction. This excites the $F = 1$ coherent superposition with probability

$$ P_{F \rightarrow 1} = \frac{\Omega^2}{\delta^2 + \Omega^2} \sin^2 \left( \frac{1}{2} \sqrt{\Omega^2 + \delta^2} \, t \right), $$

where $\Omega$ is the Rabi frequency and $\delta$ is the detuning of the rf field from resonance. When a pulse of molecules arrives at the centre of the rf loop we subject it to a short rf pulse such that $\Omega t = \pi$, i.e. a $\pi$-pulse, to induce 100% population transfer at resonance. Figure 2 shows the lineshape measured using 50$\mu$s-long rf pulses, together with a fit to eq. 1, showing quite good agreement. However, the peak transition probability is significantly less than unity. This is due to the beam velocity spread, which makes the gas pulse 5 cm long at the first loop and 10 cm at the second. Since the rf field strength varies along the beam line, this spreading produces a distribution of Rabi frequencies. The effect appears more clearly as a damping of the Rabi oscillations, shown in fig. 3(a), in good agreement with the expected damping from the known magnetic field distribution of the rf loop. The
Figure 3. Rabi flopping in the rf beam splitter. Units on axes of all the plots are arbitrary but common. The $\pi$-pulse occurs at a forward rf power to the loop of around 100mW. The dashed lines in (b) show sections taken for (c) and (d).

maximum fraction of the population transferred to $F = 1$ is 66%\textsuperscript{d}.

The arrival of each YbF pulse at our detector is recorded with 1µs resolution. This time-resolved data gives us a spatial resolution of $\sim$ 5 mm at the upper rf loop\textsuperscript{e}. This allows us to resolve the inhomogeneous Rabi frequency, as shown in fig. 3(b). The plot maps rf transition probability

\textsuperscript{d}A longer rf $\pi$-pulse increases this fraction by allowing each molecule to sample a longer distance along the beamline, but this is avoided because of the possible systematic errors due to large movements of the molecules.

\textsuperscript{e}Note, however, that the molecules move approximately 3 cm during the 50µs rf pulse.
(lighter shade meaning higher probability) versus rf field strength and arrival time at the detector. Contours of constant $\Omega t$ are clearly seen. At the centre of the loop, corresponding to the dashed line (c) in fig. 3(b), we can drive more than two complete Rabi oscillations, as plotted in fig. 3(c). Figure 3(d) shows the much slower Rabi flopping of late-arriving molecules (section (d) of fig. 3(b)) that experienced a weaker rf field. The data follow the expected sinusoidal Rabi-flopping behaviour (lines), demonstrating that the damping in fig. 3(a) is indeed due to rf field inhomogeneity.

We are currently working to shorten the rf pulses further so that the molecules move even less distance during the splitting. Preliminary results with new, higher power, rf amplifiers suggest that we can work with pulses less than 10 $\mu$s long, corresponding to a beam movement below 6 mm.

4.2. Raman splitter

The Raman beam splitter uses two co-propagating 552 nm laser beams with a frequency difference of $\sim 170$ MHz to drive the hyperfine transition. In this case the spatial localisation of the light ensures that the transition occurs in well-defined static fields. An adequate hyperfine transition rate is achieved by tuning the light near the $A_{1/2} \leftarrow X$ transition. At the same time we avoid spontaneous emission from the $A_{1/2}$ state as that tends to leave the molecule in a vibrationally or rotationally excited state. We achieve the required detuning by Stark shifting the $A_{1/2} \leftarrow X$ transition. This allows us to use just one dye laser to generate the Raman beams as well as the pump and probe light - a valuable simplification. We apply an electric field of 2.7 kV/cm to detune the transition by $\Delta = 250$ MHz, which is $\sim 10$ natural linewidths. The excitation probability is then well approximated by eq. 1 with the substitution $\Omega = \Omega_1 \Omega_2 / \Delta$, where $\Omega_{1,2}$ are the Rabi frequencies for the two laser fields. Now, $\delta$ (eq. 1) is the detuning from two-photon resonance and $t$ is the time of flight through the cw Raman splitter. Once again, the velocity spread of the beam makes it impossible to give all the molecules a $\pi$-pulse. However, the spread is only some 40 m/s FWHM on a mean velocity of 580 m/s, allowing a transition probability of 99.7% to be achieved.

Figure 4(a) shows the Raman transition lineshape. The width of this line is dominated by Doppler broadening from the transverse momentum spread of the laser beams, an effect that is less well controlled than we would like in our edm measurement. In fig. 4(b) we show the Rabi flopping of the transition, with the intensity held fixed in one beam (the redder one)
and varied in the other. Limited laser power restricts us to less than a full cycle.

![Graph](image)

**Figure 4.** Raman beam splitter. (a) Transition lineshape. (b) Rabi flopping on the Raman beam splitter transition.

### 5. Results and outlook

We are now making an electron edm measurement. Our current data set, collected over the last two months is displayed in chronological order in fig. 5. There are 3088 measurements each of which took typically 2 minutes. The small variations in average sensitivity are due primarily to changes in the intensity and stability of the molecular beam and, to a lesser degree, of the laser. In the last two weeks there is a period of sharply increased uncertainty as we start to make checks for systematic errors.

![Graph](image)

**Figure 5.** The current data set. Each of the 3088 points is an independent electron edm measurement over approximately two minutes.

Although we cannot present a value for the edm at this conference, we can make some comments on our data set. Since our last edm
measurement\textsuperscript{10}, gains from a cold, pulsed source\textsuperscript{11} and other technical improvements have increased the experimental sensitivity by more than a factor of thirty so that the statistical sensitivity is approximately $10^{-27}\text{e.cm}/\sqrt{\text{day}}$. All the data were taken using Raman beam splitters. We are preparing to take new data with the pulsed rf beam splitters once the current systematic checks are complete. As a further control against any false edm we plan to repeat the experiment using CaF molecules. These provide a good null test as CaF is similar to YbF structurally and magnetically, but has $\sim 40$ times less sensitivity to the electron edm according to the expected $Z^3$ scaling\textsuperscript{7}. We have already performed a trial run and the results look encouraging for making a measurement in the $10^{-28}\text{e.cm}$ range in the near future. Beyond that, we plan to guide and decelerate the beam\textsuperscript{14}, to obtain an anticipated further factor of 100 in sensitivity.

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