

Improvements to Permanent Electron Electric Dipole Moment Measurements using Clock Transitions in Cold Molecules

Z0981459

The Standard Model of particle physics represents decades of advancements in our understanding of the structure of matter and the forces binding it together. Whilst the Standard Model is capable of accurately predicting the results of atomic interactions it nevertheless fails to explain one of the most fundamentally vital questions still taunting modern physicists; the question of the uneven proportion of matter to antimatter that exists in the observable Universe [1, 2].

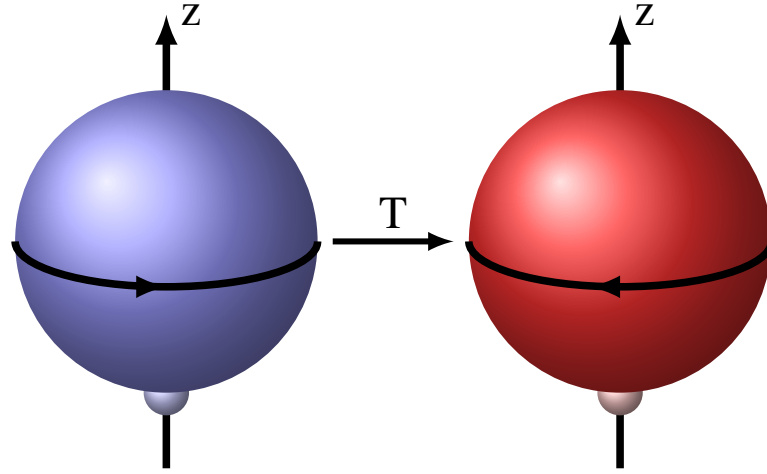


Figure 1: Demonstration of Time invariance violation. Two electrons of opposite spin rotation (indicated by equatorial black arrow) visualised as solid balls of charge. Each electron represents a time reversed version of the other (transformation T). The EDM is represented as a smaller lump of charge attached to the underside of the electron. The arrow aligned along z indicates the direction in which the electron spin points [4].

One promising discovery that poses as a potential answer to this question, is the accurate measurement of the value of the permanent electric dipole moment (EDM). For an electron, a permanent EDM can be visualised as an asymmetric distribution of charge, parallel to the axis of average angular momentum. For a spin-1/2 particle this is along the spin axis \mathbf{S} , as demonstrated in figure 1 [3]. This slight charge displacement leads to the classical result of the formation of a moment, where the EDM (d_e) can be described as $\mathbf{d} = d_e \mathbf{S}$. With corresponding Hamiltonian: $H = \mathbf{d} \cdot \mathbf{E}_{eff}$ [5, 6]. Where \mathbf{E}_{eff} is an effective electric field approximated using molecular theory as $|E_{eff}| \approx 8Z^3\alpha^2 E_{ext}$. Where Z is the nuclear charge, α is the fine structure constant and E_{ext} is an externally applied electric field. For a molecule $|E_{eff}|$ can then be scaled using the strength of the molecule's polarisation [7].

Fundamentally, the search for a permanent EDM is a search for large scale CP violation. CP violation describes a difference in the laws of physics under two transformations: charge conjugation C (the symmetry between particles and antiparticles) and parity inversion P (spatial inversion) [8]. The

alignment of the spin and EDM of an electron results in CP violation, as it is non-invariant under time reversal (T) transformations. This is due to combined CPT symmetry, where violations under T imply a corresponding CP violation [4, 8]. In order to explain the Universal asymmetry between matter and anti-matter, more CP violation than is currently predicted by the Standard Model is required [9]. Therefore suggesting the existence of physics beyond the Standard Model. Theoretical models, such as supersymmetry (SUSY), explain this new physics through introducing additional sources of CP violation. These theories anticipate electron EDM values which are several orders of magnitude larger than the value predicted by the Standard Model and which are within range of modern precision measurement techniques. Validation of such models, following a successful value measurement, would drastically alter the next generation of particle physics theories and would question the viability of the Standard Model [7].

One technique applied in the search for the electron EDM uses clock transitions within heavy polar molecules. These molecules include atomic hyperfine states where the transition energy can be determined to a high degree of accuracy, as states possess small natural linewidths due to spontaneous emission [10, 12]. Experiments measure the energy shift/ population change of quantum states due to the linear Stark interaction of the electron EDM with an electric field. The EDM induces an energy level shift of $\Delta E = -d_e E_{eff}$ which is then detectable in the frequency needed to drive a transition [7, 11]. New methods of producing cold molecules for use in EDM searches has increased the sensitivity and accuracy of these experiments. This is because large ensembles of trapped polar molecules have greater sensitivity to P,T-violations due to their longer lifetimes [13]. Molecules such as Ytterbium Silver ($^{174}\text{Yb}^{107}\text{Ag}$) are used as it possesses magnetically-insensitive hyperfine states or clock states [10]. Crucially, molecules share a lower Zeeman interaction with a motional magnetic field than single atoms, due to their strong tensor polarisability. An ideal condition as this interaction can mimic the effect of the EDM interaction and skew conclusions [7].

Using YbF molecules as an example, a proposed measurement technique which will use clock state transitions to determine the value of the electron EDM is now discussed. The proposed experiment yields an EDM measurement sensitivity which is far beyond current limits [10]. The increase in precision of this technique can be attributed to advances in the development of atomic clocks and atomic spectroscopy [12]. Molecules of YbF are first assembled from their constituent atoms at ultracold temperatures in an optical trap. Laser cooling then used to populate the Zeeman sub-states of the $N = 0$ level, where N corresponds to the rotational state quantum number. The respective levels for a molecule of YbF are shown in figure 2 [7, 14].

For a molecule in the $X^2\Sigma^+$ electronic ground state of YbF (indicated in figure 2 as $X^2\Sigma^+$) the effective interaction with an external electric \mathbf{E} and magnetic \mathbf{B} field is expressed as the Hamiltonian

$$H_{eff} = -\mu_B(g_s\mathbf{S} + g_I\mathbf{I}) \cdot \mathbf{B} + W_{PT}\mathbf{S} \cdot \hat{n} - D\hat{n} \cdot \mathbf{E}, \quad (1)$$

where g_s and g_I are the electron and nuclear spin g-factors. \mathbf{S} and \mathbf{I} are the valence electron spin and nuclear spin of the molecule respectively. And where: D is the molecular dipole moment, \hat{n} is a unit vector directed parallel to the internuclear molecule axis and μ_B is the Bohr magneton constant. The effect of the EDM is encoded in the term $W_{PT}\mathbf{S} \cdot \hat{n}$, where W_{PT} is analogous to the electron EDM [5].

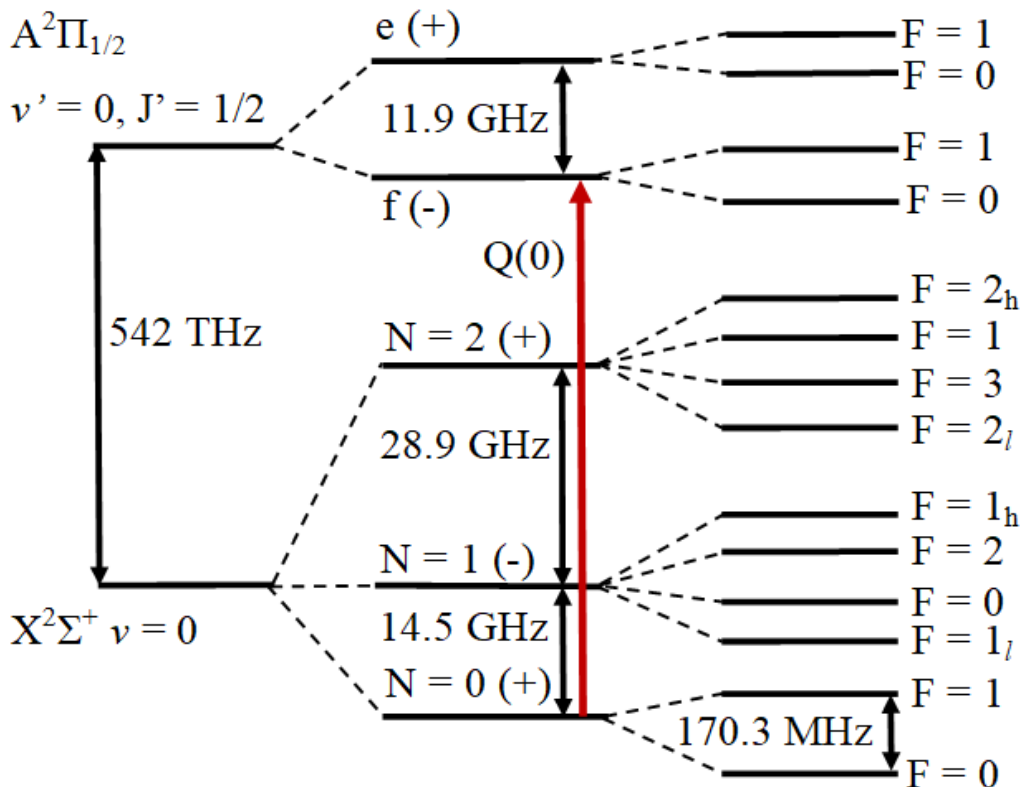


Figure 2: YbF energy level diagram: Indicated are the energies required for a transition between states used in the EDM measurement. Q(0) (red) refers to an optical transition used for population measurements. The relevant hyperfine states ($F=1$ and $F=0$) have been indicated using an associated energy difference. Where N and ν are vibrational and rotational state quantum numbers. J is the total angular momentum. [7, 14].

The hyperfine clock states of YbF can be labelled as $|g\rangle \equiv |F = 0, m_F = 0\rangle$ and $|e\rangle \equiv |F = 1, m_F = 0\rangle$. Where F indicates the total angular momentum and an associated quantum number m_F . These states are separated by an energy of ω_0 . In figure 2, ω_0 is shown as equal to 170.3 MHz. A time (t) dependent polarizing electric field $E_z = E_0 \cos(\omega_E t + \beta)$ and magnetic field $B_z = B_0 \cos(\omega_B t)$ is applied to the molecules. Where ω_B and ω_E are the frequencies at which the magnetic and electric fields are driven respectively. An adjustable phase difference between the fields is expressed as β [5, 10].

The magnetic field stimulates a clock transition between the two states, whilst the electric field causes the molecular orientation to oscillate with frequency ω_E and amplitude ζ_0 . Due to the PT violating term W_{PT} of the Hamiltonian and as a result of the presence of the electron EDM, the oscillating molecular orientation behaves like an effective magnetic field. This induces an extra contribution to the transition amplitude between $|g\rangle$ and $|e\rangle$. This transition amplitude either increases the probability for a transition to occur or decreases it depending on the phase β of \mathbf{E} with respect to \mathbf{B} [10].

Driving the transition of molecules for a time τ , using the oscillating magnetic and electric fields, results in molecules initially prepared in $|g\rangle$ being transferred to $|e\rangle$. Using the Hamiltonian described in equation 1 the final transition amplitude is expressed as $\Omega_{BT}\tau \pm \Omega_{PT}\tau$ (sign determined by β). With the Rabi frequencies due to \mathbf{B} and \mathbf{E} expressed as $\Omega_B = -1/2(g_s - g_I)\mu_B B_0$ and $\Omega_{PT} = 1/2W_{PT}\zeta_0$ respectively, with all terms are previously defined [10].

It is assumed the electric and magnetic fields are driven at the same frequency, $\omega_E = \omega_B = \omega$. The detuning of the imposed fields is calculated as $\Delta = \omega - \omega_0$. Supplying the molecule with fields on resonance ($\Delta = 0$) results in an excited state population:

$$\rho_e = \sin^2 \left[\frac{(\Omega_B + \Omega_{PT} \cos \beta) \tau}{2} \right], \quad (2)$$

with all terms as previously defined. During an EDM measurement \mathbf{B} and τ are locked so that $\Omega_B \tau$ is equal to $\frac{\pi}{2}$. Ensuring that the impact of the EDM on the population of molecules in $|e\rangle$ can be identified through varying the phase between the two fields [10]. The number of molecules present in a quantum state is measured using laser-induced fluorescence. The Q(0) transition indicated in figure 2 is the excitation used for this process. With the resulting photon numbers measured using photomultiplier tubes (PMT) [14, 15].

Following a final measurement of W_{PT} , the result of this measurement technique corresponds to a predicted electron EDM sensitivity of

$$\delta d_e = 10^{-31} \text{ ecm} \left(\frac{10^4}{N} \right)^{1/2} \left(\frac{10 \text{ s}}{\tau} \right) \left(\frac{10 \text{ d}}{T} \right)^{1/2} \left(\frac{17.5 \text{ GV/cm}}{E_{eff}} \right) \left(\frac{1}{\zeta_0} \right). \quad (3)$$

Where N is the number of molecules being examined. The time T is the total time the molecules spend within the apparatus. E_{eff} is the effective electric field and is therefore calculated for the specific molecule being investigated [10].

The described method suffers from a statistical uncertainty σ which is common in most shot-noise limited measurements of d_e . Such uncertainty is related to experimental parameters via

$$\sigma_d = \frac{\hbar}{2E_{eff}\tau\sqrt{N}}, \quad (4)$$

where N is the number of particles detected and all other terms are as previously defined [14]. Extensions to the atomic transitions method have been suggested to reduce the error. For instance, the use of laser cooling of molecular beams has been proposed in experiments using YbF and BaF molecules in fountain arrangements to increase the evolution time τ [17, 18].

Test Atom/molecule	Year	Research Group	d_e limit (e cm)
TI	2002	Berkley (US)	1.6×10^{-27}
YbF	2011	Imperial (UK)	1.1×10^{-27}
ThO	2014	ACME I (US)	8.7×10^{-29}
HfF ⁺	2017	JILA (US)	1.3×10^{-28}
ThO	2018	ACME II (US)	1.1×10^{-29}

Table 1: Improvements to the experimental limit of the EDM value [3, 13, 19, 20, 21]

The last decade has seen the value of d_e tentatively decrease towards the theoretically predicted range of $d_e \approx 10^{-27}$ ecm to $d_e \approx 10^{-30}$ ecm [7, 9]. The improvements have almost entirely come from experiments utilising cold molecule techniques, as can be seen in table 1 [22]. The current upper experimental limit of $|d_e| < 1.1 \times 10^{-29}$ ecm was achieved by the ACME group of Yale University [3]. Several key research groups plan to use molecules of YbF for their future measurements and are seeking

to remove smaller and smaller areas of uncertainty (such as excluding background magnetic fields to the sub n-T scale) [14, 15, 16].

The EDM is very sensitive to the mass of weakly interacting particles and can be used to detect their existence. The magnitude of the EDM scales as the inverse of a heavy particle's mass. Hence improving the accuracy of EDM measurements through better spectroscopy measurements provides a much greater mass reach than the corresponding accuracy improvement in any collider. Therefore raising the question of whether expensive experiments, such as the LHC at CERN, are still the most viable techniques for exploring physics beyond the Standard Model [4, 6].

References

- [1] C. Orzel, *New Physics Using Precision Spectroscopy*, Adv. in Atomic, Molecular and Optical Phys. (2018) 67:337
- [2] T. E. Chupp, P. Fierlinger, M. J. Ramsey-Musolf & J. T. Singh, *Electric dipole moments of atoms, molecules, nuclei and particles*, Rev. Mod. Phys. (2019) **91** 015001
- [3] V. Andreev, et al. (ACME Collaboration), *Improved limit on the electric dipole moment of the electron*, Nature (2018) **562** pgs 355-360
- [4] N. Fortson, et al. *The Search for a Permanent Electric Dipole Moment*, Adv. Atomic, Molecular and Optical Phys. (2010) 59:129
- [5] T. Chupp, *Permanent Electric Dipole Moments of Atoms and Molecules*, Adv. Atomic, Molecular and Optical Phys. (2010) 59:129
- [6] Y. Nakai & M. Reece, *The implications of a Precise Electron Measurement*, Phys. Today (2018), DOI:10.1063/PT.6.3.20181114a
- [7] C. J. Ho, et al., *New Techniques for a measurement of the electron's electric dipole moment*, New J. Phys. (2020) **22** 053031
- [8] P. Hamilton, *Preliminary results in the search for the electron electric dipole moment in PbO*, Yale E-Thesis (2010) :
- [9] A. Sakharov, ZhETF Pis'ma (1967) **5** 32
- [10] M. Verma, A. M. Jayich & A. C. Vutha, *Electron electric dipole moment searches using clock transitions in ultracold molecules*, Phys. Rev. Lett. (2020) **125** 153201
- [11] M. S. Soronova, et al., *Search for New Physics with Atoms and Molecules*, Rev Mod Phys. (2018) 90:025008
- [12] C. Orzel, *Searching for New Physics Through AMO Precision Measurements*, Union College Dept. Phys. & Astro. (2012)
- [13] W. B. Cairncross, et al., *A precesion measurement of the electron's electric dipole moment using trapped molecular ions*, Phys. Rev. Lett. (2017) **119** 153001
- [14] N. J. Fitch, J. Lim, B. E. Sauer, E. A. Hinds & M. R. Tarbutt, *Methods for measuring the electron's electric dipole moment using ultracold YbF molecules*, Quantum Sci. Technol. (2021) **6** 014006
- [15] B. E. Sauer, et al., *Prospects for the measurement of the electric electric dipole moment using YbF*, Phys. Procedia (2011) **17** pgs 175 - 180
- [16] H. Loh, et al., *Precision Spectroscopy of Polarized Molecules in an Ion Trap*, Science (2013) **342** pgs 1220 - 1222
- [17] M. R. Tarbutt, B. E. Sauer, J. J. Hudson & E. A. Hinds, *Design for a fountain of YbF molecules to measure the electron's electric dipole moment*, New J. Phys. (2013) **15** 053034

- [18] P. Aggarwal, et al., *Measuring the electric dipole moment of the electron in BaF*, The European Phys. J. D (2018) **72** 197
- [19] B. C. Regan, E. D. Commins, C. J. Schmidt & D. DeMille, *New Limit on the Electron Electric Dipole Moment*, Phys. Rev. Lett. (2002) **88** 071805
- [20] J. J. Hudson, D. M. Kara, I. J. Smallman, et al., *Improved measurement of the shape of the electron*, Nature (2011) **473** 493
- [21] The ACME Collaboration, J. Baron, et al., *Order of Magnitude Smaller Limit on the Electric Dipole Moment of the Electron*, Science (2014) **343** 269
- [22] M. A. Trigatzis, *An ultracold molecular beam for measuring the electric dipole moment of the electron*, Impl Coll. Dept. Phys. E-Thesis (2020)