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Time reversal symmetry violation in the YbF molecule

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Abstract We present a summary of the techniques used to test time reversal symmetry by measuring the permanent electric dipole moment of the YbF molecule. The results of a recent measurement [Nature **473**, 493 (2011)] are reported. We review some systematic effects which might mimic time reversal violation and describe how they are overcome. We then discuss improvements to the sensitivity of the apparatus, including both short term technical enhancements as well as a longer term goal to use laser cooled YbF in the experiment.

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1 Introduction

More than ten orders of magnitude separate the current experimental limit on the size of the dipole moment of the electron, d_e, from the prediction of the standard model. A non-zero value of d_e would violate time (T) reversal symmetry. Via the CPT theorem, the existence of permanent dipole moments (EDMs) of fundamental particles is intimately related to CP violation. The known CP violation in the standard model generates very small EDMs [1], but many extensions to the standard model [2] include new sources of CP violation which give rise to EDMs near the current experimental limits . As an example, CP violation in supersymmetric models is already strongly constrained by the need to keep the predicted values of EDMs small. Most models of baryogensis in the early universe also require new sources of CP violation in order to give a matter dominated universe [3]. Thus experiments to measure EDMs give insight into CP physics beyond the standard model.

Our experiment [4] uses the YbF molecule to search for T symmetry violation. In its ground state YbF has a single unpaired electron (a $^2\Sigma^+$ ground

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state). We interpret our results in terms of d_e, although as Le Dall and Ritz discuss in this issue [5], the electron EDM can only be distinguished from other T violating effects by comparing different atomic and molecular systems. This paper presents the techniques we used to make this measurement and discusses future improvements which should increase the experimental sensitivity.

2 Technique

The ground electronic, vibrational and rotational state of YbF has hyperfine structure consisting of an $F = 1$ triplet and an $F = 0$ singlet separated by about 170MHz. The energies of these states shift in an electric field [6], but if T is a good symmetry then the $|F, m_F\rangle = |1, +1\rangle$ and $|1, -1\rangle$ remain degenerate. If d_e is not zero, these states have a relative shift $2d_e\eta E_{eff}$, where η gives the polarization factor of the molecule due to the external electric field and E_{eff} parameterizes the enhancement of the dipole moment due to the molecular structure. It has long been recognized [7] that in heavy atoms and in molecules with a heavy nucleus E_{eff} can be much larger than any achievable laboratory field. This enhancement is a relativistic effect [8] and also depends on details of the electronic wavefunction. For YbF these details have been calculated by several independent methods which within 20% agree that $E_{eff} = -25$ GV/cm [9]. The effective field for some more exotic molecules which might be used in future T violation experiments have recently been estimated [10]. In our experiment a laboratory field of 10 kV/cm is applied, which gives the product $\eta E_{eff} = -14.5$ GV/cm.

Figure 1 presents an overview of the apparatus used for the 2011 measurement in YbF. A more detailed description can be found in [11]. YbF molecules were produced in a pulsed ablation source [12]. They were entrained in Ar carrier gas whose supersonic expansion cools the translational and rotational temperature of the YbF to about 3K. This left a significant fraction of the molecules in the ground rotational state which participates in the EDM experiment, at the expense of an increase in forward velocity to around 590 m/s. After a skimmer, the packet of YbF molecules passed through a pump laser beam whose frequency was tuned to excite the $F = 1$ component of the ground state to the $A^2\Pi_{1/2}$ electronic state, from which the population decayed to higher rotational states [13] leaving the YbF population of interest in the $F = 0$ state. Each packet then entered a magnetically shielded region which contained current loops and high voltage electrodes which produced uniform magnetic and electric fields. These electrodes doubled as a radiofrequency transmission line. The radiation was carried to the plates along non-magnetic semi-rigid coaxial cable and capacitively coupled to the corner of each end of the plates. This rather primitive coupling method produced a significant circular component of the rf field near the end of the plates [14].

When the YbF packet was about 13cm into the electric field region, an 18μs rf pulse was applied which drove the population from the state $|0, 0\rangle$ to the superposition state $(|1, +1\rangle + |1, -1\rangle)/\sqrt{2}$. This state evolved for a time

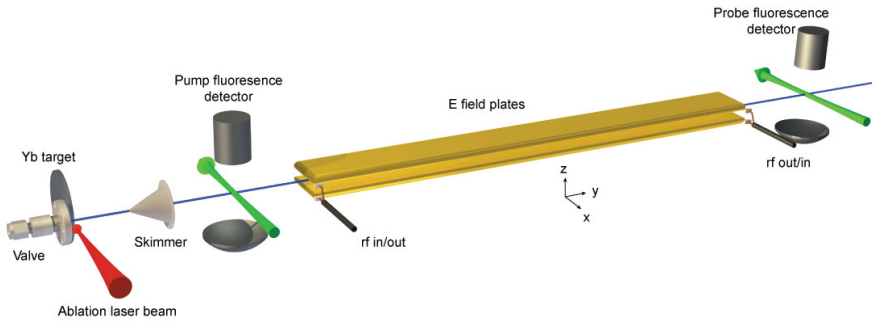

Fig. 1 Schematic diagram of the pulsed beam apparatus.

$\tau = 0.64$ms in the E and B fields, acquiring a phase shift $(e^{-i\varphi}|1,+1\rangle + e^{i\varphi}|1,-1\rangle)/\sqrt{2}$, where $\varphi = \varphi_e + \varphi_b = -d_e\eta E_{eff}\tau/\hbar + g\mu_B B\tau/\hbar$. For $F = 1$ states, g is very nearly equal to 1. A second rf pulse transfered the population back to the $|0,0\rangle$ state with an amplitude which depended on φ. Due to the need to avoid the ends of the field plates, the molecules only traveled 38cm of the 75cm length of the field plates during their free evolution. After the second rf pulse, the molecular packet exited the plates and the magnetically shielded region and passed through a probe laser beam which excited the $|0,0\rangle$ state. The collected fluorescence measured the population of this state, yielding a signal

$$S = S_B + S_0 \cos^2 \varphi, \tag{1}$$

where S_0 is proportional to the number of $|0,0\rangle$ molecules and S_B is the background. The molecular pulse has a velocity width of about 10% of its forward velocity, so the time-of-flight (TOF) signal seen at the probe detector is about 190μs FWHM. Molecules which arrive at different times experienced the rf pulses and free evolution at different spatial positions, as their timing was fixed.Varying the time gate used to analyze the TOF signal probed the effects of these different regions of the beam line [15].

Figure 2 shows the probe collection optics in more detail. A spherical mirror and a pair of spherical lenses were used to collect the fluorescence light and image it onto the face of a photomultiplier tube (PMT). The figure shows a calculation of the focused fluorescence intensity from the molecular beam on the face of the PMT; a field stop was used to mask off-axis light from laser scatter. The calculated efficiency of the collection optics is 5.1%, but small imperfections in alignment could easily reduce this by 20%. The quantum efficiency of the PMT photocathode was 10%; we calculate only 0.4–0.5% of the $|0,0\rangle$ population was actually detected. Our 2011 measurement averaged about 1000 photoelectrons per YbF packet, about half of which contributed to the measurement of the EDM.

The signal S given in eq. 1 is most sensitive to small values of φ_e when $\varphi_b = \pm\pi/4$, i.e. at the steepest slope of the curve shown in fig. 3. To produce this value of φ_b required a magnetic field of $\pm$13.6nT. An additional step

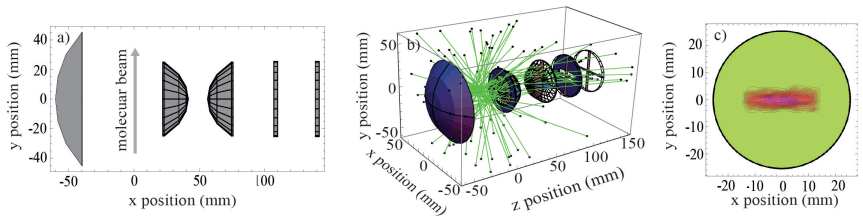

Fig. 2 The probe region collection optics showing a) a cross section of the mirror (left), lens pair (middle), field stop and PMT face (right) , b) a ray tracing analysis of the fluorescence light collection efficiency, and c) the distribution of collected fluorescence at the field stop.

in the magnetic field, $\delta B = \pm 1.7$nT was also applied in order to calibrate the measurement. An additional six parameters of the rf pulses and the laser frequency were modulated. Online analysis of these signals allowed us to apply feedback to keep the transitions on resonance in the face of slow drifts, and to counteract changes in the background magnetic field [16].

To measure a value for d_e, the relative signs of the applied E and B fields were switched between different beam pulses and the signal correlated with the sign of $\mathbf{E} \cdot \mathbf{B}$ extracted. A "block" of 4096 beam pulses was a measurement of d_e. The uncertainty in the value for d_e, σ_d was calculated from the distribution of the individual block values using a bootstrap analysis [16].

It is instructive to estimate the shot noise limit in our experiment, assuming the lineshape of eq. 1. Expanding around $\varphi_b = \pm\pi/4$ with the assumption that φ_e is small yields a signal S for each pulse linear in d_e:

$$S = S_B + S_0(\frac{1}{2} \mp d_e \eta E_{eff} \tau/\hbar). \tag{2}$$

For the 2011 measurement calibration of the analog PMT amplifier gave $S_0 = 500$ photoelectrons per pulse, with S_B the equivalent of 200 photoelectrons. The shot noise uncertainty for a single molecular pulse is

$$\sigma_d^{(1)} = \frac{\hbar\sqrt{S_B + S_0/2}}{\eta E_{eff} \tau S_0}. \tag{3}$$

Defining the contrast of the interference curve (fig. 3) as

$$\mathcal{C} = \frac{S_{max} - S_{min}}{S_{max} + S_{min}} = \frac{S_0}{S_0 + 2S_B} \tag{4}$$

gives a particularly simple expression for the shot noise sensitivity for $\mathcal{N}$ beam pulses:

$$\sigma_d^{(\mathcal{N})} = \frac{\hbar}{\eta E_{eff} \tau \sqrt{2\mathcal{C}\mathcal{N} S_0}}. \tag{5}$$

For the 2011 measurement $\mathcal{N} = 25$ million and $\mathcal{C} = 0.55$, this gives an expected shot noise sensitivity $\sigma_d^{(\mathcal{N})} = 5 \times 10^{-28}$e.cm. The probe signal was normalized to the pump fluorescence for each shot, this reduced the effect of

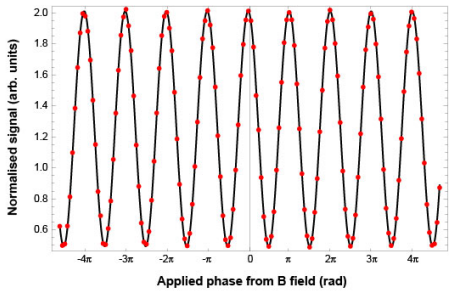

Fig. 3 An example of the probe fluorescence recorded as a function of the applied magnetic field. The signal has been normalized to the pump fluorescence. To avoid the effects of velocity averaging only a very narrow time window of the TOF signal has been selected. The contrast of this interference curve is therefore not representative of the actual EDM data.

source fluctuations but increased the shot noise by a factor of 1.12 due to the counting statistics of the pump detector. In all, the uncertainty in the 2011 measurement, 5.7×10^{-28}e.cm (68% c.l.), was only about 4% above this shot noise estimate.

3 Systematic uncertainties

Although the 2011 measurement was limited by statistical sensitivity, a number of potential systematic effects which could mimic T-violation were investigated. Any non-zero correlation related to the sign of $\mathbf{E} \cdot \mathbf{B}$ will give a false result. A simple example of such an effect would be a magnetic field produced at the molecular beamline by the relays which switch the high voltage for the electric field. We guarded against this particular effect by recording the signal from an array of magnetometers in the lab and by periodically reversing the electric and magnetic connections to the machine manually.

More subtle systematic effects arise because the rf transitions take place in the electric and magnetic fields which are used to measure d_e. The largest example of such an effect in the 2011 measurement was related to a change of the phase in eq. 1 correlated with the rf frequency detuning. By itself, this is not a systematic effect. However when the electric field was reversed its magnitude did not reverse perfectly. This produced an rf detuning which depended on the direction of E via the Stark effect of the hyperfine transition. The combination of these two effects produced a false EDM which could be corrected from the measured size of the individual effects. Details of this and of other, smaller, systematics corrections are given in [16].

4 Sensitivity improvements

After the 2011 measurement was completed a number of improvements were made to the apparatus in order to increase its sensitivity. The apparatus had

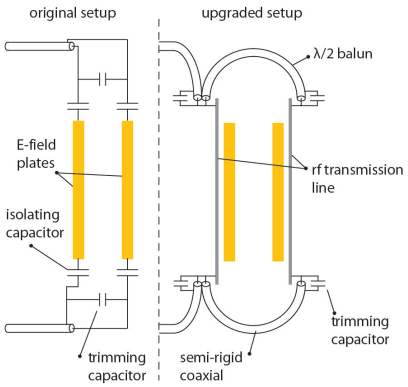

Fig. 4 Coupling of the coaxial radiofrequency feed to the transmission line formed by the high voltage plates. The left side shows the scheme used for the 2011 measurement. The right side shows the improved scheme with auxiliary rf plates and a balanced feed which nearly eliminates the circularly polarized component of the rf radiation.

two layers of magnetic shielding, one outside the vacuum system and one inside which enclosed the electric field plates. This inner shield was a cylinder whose endcaps were 2cm from the ends of the electric field plates. Without increasing the distance between the YbF source and the probe detector, we replaced this shield with a longer version whose endcaps are 12cm from the ends of the field electrodes. This reduced magnetic field gradients over the interaction region and is one change which allowed us to increase the measurement coherence time.

At the same time, the rf coupling to the field plates was changed. Figure 4 illustrates the old and new setups. We installed a pair of auxiliary rf transmission line plates which sit 5mm behind the high voltage electrodes and extend 5cm beyond each end. A balun arrangement which is symmetric about the centerline of the plates couples rf to these auxiliary plates. Compared to the previous arrangement this has two advantages: the circular component of the rf magnetic field is much reduced and a higher voltage can be applied to the electric field plates without excessive leakage. These changes also eliminated the rf frequency induced phase shift, which in turn eliminates the largest systematic correction from the previous measurement.

Reconfiguring the magnetic and rf interaction region allowed us to increase the coherence time τ from 642μs to 980μs, an increase in sensitivity of 1.5. The electric field applied has increased from 10kV/cm to 12.5kV/cm, which increases the polarization factor η by 1.1. For the 2011 measurement the contrast $\mathcal{C}$ was 0.55, with a background mainly due to YbF molecules which experienced imperfect rf transitions due to electric and magnetic gradients along the length of the molecular pulse. By using much higher rf power we are able to shorten the rf transition time, which broadens the rf linewidth and thus reduces the effects of gradients. This reduces the size of some systematics and increases the signal to noise ratio by both allowing more of the TOF curve to

be included and improving the contrast by reducing S_B. We have measured an improvement in sensitivity of 1.25 due to the shorter rf pulses.

Taken together these rather modest improvements in the EDM apparatus have improved its sensitivity by a factor of 2. A third layer of magnetic shielding has been added so that the laboratory magnetic noise floor will continue to be below the statistical sensitivity. We have also improved the reliability of the machine and expect to be able to take about 50% more data than contributed to the 2011 measurement. After making the changes to the apparatus we made extensive checks of possible systematics and can find nothing that is troubling at the level of 6×10^{-29}e.cm. We are therefore confident that we can measure d_e in YbF at the level of 2×10^{-28}e.cm, a factor of three better than our previous result.

The improvements discussed above are not the only way to enhance the experiment. To increase the sensitivity of the YbF measurement even more requires a longer interaction time τ or a more intense source, or both. We have studied the production of YbF in a buffer gas cell filled with gaseous He at 4K [17]. As well as having a low rotational temperature, the YbF extracted from the cell has a velocity three times lower than that produced by our supersonic source and at the same time the beam has up to ten times the flux. Buffer gas sources have been shown to work reliably for other molecular species [18]; one that does so for YbF promises an order of magnitude improvement in the measurement of the electron EDM.

Even further improvement seems possible. Recently it has been shown that atomic laser cooling techniques can be extended to some particular molecules [19]. For these techniques to be practical, it is necessary for the molecule to have a favorable Franck-Condon structure such that only a few vibrational repumping laser frequencies are required. We have recently measured the Franck-Condon factors for YbF [13] and find that laser cooling should be possible. We envision producing YbF in a buffer gas source, using a magnetic guide to separate the YbF beam from the He background, then applying laser cooling and launching the YbF in a molecular fountain to maximize the coherence time of the measurement. We estimate that only 1.5×10^{-8} of the YbF molecules produced in the cell would actually participate in the EDM measurement. However, the YbF density in cell is extremely high, so this will yield a beam flux comparable to our current supersonic source, with a coherence time more than 300 times longer. This effort seems both worthwhile and exciting: at this level of sensitivity YbF would probe new elementary particle physics at energy scales up to 100 TeV and promises to illuminate the CP-violating physics of the very early universe.

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