Measurement of the Electron Electric Dipole Moment Using YbF Molecules

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The most sensitive measurements of the electron electric dipole moment d_e have previously been made using heavy atoms. Heavy polar molecules offer a greater sensitivity to d_e because the interaction energy to be measured is typically 10^3 times larger than in a heavy atom. We have used YbF to make the first measurement of this kind. Together, the large interaction energy and the strong tensor polarizability of the molecule make our experiment essentially free of the systematic errors that currently limit d_e measurements in atoms. Our first result $d_e = (-0.2 \pm 3.2) \times 10^{-26} e$ cm is less sensitive than the best atom measurement but is limited only by counting statistics and demonstrates the power of the method.

The permanent electric dipole moment (edm) of an elementary particle vanishes unless the discrete symmetries parity (P) and time reversal (T) are both violated [1]. This naturally makes the edm small in fundamental particles of ordinary matter. In the standard model of elementary particle physics, a cancellation of leading order Feynman diagrams further suppresses the expected value of the electron edm d_e to less than $10^{-38} e$ cm [2], but in theories going beyond the standard model, typical predictions are in the range 10^{-26}–$10^{-28} e$ cm [3]. The search for an electron edm is therefore a search for physics beyond the standard model, and particularly it is a search for T violation. T violation is intimately connected to CP violation through the CPT theorem [4]. This is an important and active field at present [5] because the prospects for discovering new physics seem so good. Such research is complementary to experiments on K and B particles where the observed CP violation confirms the standard model Cabibbo-Kobayashi-Maskawa mechanism [6]. The most precise measurement of the electron edm is that of Regan *et al.* [7], who have achieved the result $d_e = (7 \pm 8) \times 10^{-28} e$ cm by searching for a differential Stark shift between the two hyperfine sublevels ($F = 1, m_F = \pm 1$) in the ground state of atomic thallium. Their result imposes stringent constraints on the possible theories of particle physics. Any improvement would make a very important contribution to the further search for new physics beyond the standard model, but their method seems to have reached its limit because of systematic errors associated with stray magnetic fields. Such errors should be greatly suppressed if d_e could be measured using heavy polar molecules instead of atoms [8], but hitherto this has not been technically possible. Here we report on the first experiment to realize this idea, which demonstrates the feasibility of the method and, although not yet as accurate as the thallium experiment, nevertheless obtains the next most significant constraint [9] on d_e. We show that this new molecular method is potentially much more sensitive than the previous atomic experiments.

To leading order in d_e the edm interaction of an electron in an atom or molecule can be described by an effective Hamiltonian $d_e(1 - \gamma_0)\Sigma \cdot \mathbf{E}_{\text{tot}}$, where γ_0 and Σ are standard Dirac matrices and $\mathbf{E}_{\text{tot}}$ is the total electric field at the position of the electron, including the field of the other electrons [10]. The explicit form of this interaction,

$$V = \left\langle \psi_0 \left| \begin{matrix} 0 & 0 \\ 0 & 2d_e\hat{\sigma} \cdot \mathbf{E}_{\text{tot}} \end{matrix} \right| \psi_0 \right\rangle, \qquad (1)$$

where $\hat{\sigma}$ is a unit vector along the electron spin, shows that the effect is relativistic: the operator connects only the small components of the Dirac wave function $|\psi_0\rangle$. The sensitivity to d_e is therefore largest in heavy systems where the electrons are most relativistic. If there is no applied electric field, the small component of $|\psi_0\rangle$ has definite parity and therefore the expectation value of this odd-parity operator vanishes. However, an applied field $E\hat{z}$ polarizes the wave function and yields an energy V of the form $-d_e E_{\text{eff}}\langle\hat{\sigma} \cdot \hat{z}\rangle$. A strong laboratory field applied to an atom induces only a small mixing of atomic orbitals and therefore E_{eff} is essentially proportional to E, with the ratio scaling roughly as $8Z^3\alpha^2$ [11], Z being the nuclear charge and α the fine structure constant. The cesium atom used by Hunter's group to measure d_e [12] is a particularly favorable case with the applied field being effectively amplified by 119 [3]. Thallium has an even larger amplification of -585, a key factor in the more sensitive result of Ref. [7]. Diatomic polar molecules have a great advantage over atoms [8] because the atomic orbitals are naturally strongly polarized along the internuclear axis $\hat{\lambda}$. This gives rise to an internal effective electric field $E_{\text{int}}\hat{\lambda}$ that can be very large when one of the atoms is heavy and has strong s-p hybridization of its orbitals. The molecular rotation averages this strong field to zero unless an external field $E\hat{z}$ is applied. However, only a modest field is needed to polarize $\hat{\lambda}$ along $\hat{z}$ because this involves mixing rotational states, which are much closer together than the electronic states of an atom. The edm interaction energy V once again takes the form $-d_e E_{\text{eff}}\langle\hat{\sigma} \cdot \hat{z}\rangle$, but E_{eff} is now given by $E_{\text{int}}\langle\hat{\lambda} \cdot \hat{z}\rangle$. Figure 1 shows E_{eff} of the
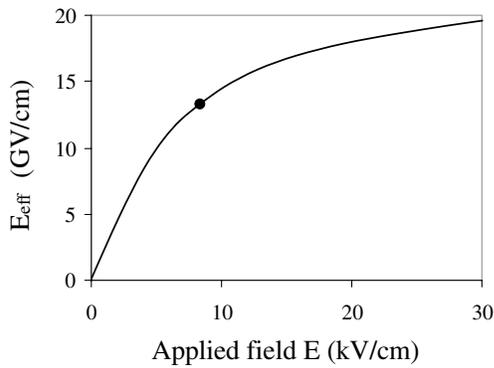
FIG. 1. Effective electric field interacting with the electron edm in YbF versus applied electric field. The dot shows the field at which we operate.

YbF molecule, rising towards its asymptotic value E_{int} of 26 GV/cm [13] as the applied field is increased. In our experiment the applied field is 8.3 kV/cm, for which E_{eff} is 13 GV/cm. Such a large effective field is not particular to YbF but can be found in a variety of other heavy polar diatomic molecules, some of which are listed in Table I. In short, the edm interaction in heavy polar molecules can be thousands of times larger than in heavy atoms.

Our experiment uses ^{174}YbF in the electronic, vibrational, and rotational ground state $X^2\Sigma^+(v = 0, N = 0)$. The electron spin (1/2) and the fluorine nuclear spin (1/2) combine to produce a hyperfine singlet $F = 0$ and triplet $F = 1$, separated by 170 MHz [17] as shown in the inset in Fig. 2. The applied electric field lowers the energy of the $|F, m_F\rangle = |1, 0\rangle$ state relative to $|1, \pm1\rangle$ by an amount Δ ($\Delta/h = 6.7$ MHz for the 8.3 kV/cm used in our experiment) [18]. This strong tensor splitting reflects the nonspherical symmetry of the molecule's internal structure. The two states $|1, +1\rangle$ and $|1, -1\rangle$ remain degenerate as a consequence of time-reversal symmetry. Their degeneracy is lifted by the edm interaction, which causes a splitting of $2d_e E_{eff}$ that we seek to measure in the experiment. A magnetic field small compared with Δ/μ_B causes an additional splitting [19] of $2\mu_B B_z[1 - \frac{1}{2}(\mu_B B_\perp/\Delta)^2]$ plus higher order corrections (here the g factor, both expected

TABLE I. Effective electric fields for some heavy polar molecules.

Species: state	E_{eff} (GV/cm)
BaF: $X^2\Sigma^+$	7.4[a]
YbF: $X^2\Sigma^+$	26[b]
HgF: $X^2\Sigma^+$	99[c]
PbF: $X^2\Sigma^+$	−29[c]
PbO: a(1) $^3\Sigma^+$	6[d]

[a]Reference [14].
[b]Reference [13].
[c]Reference [15].
[d]Reference [16].

and measured, is 1). This formula shows that the field parallel to **E** induces a Zeeman splitting $2\mu_B B_z$ between the $m_F = \pm1$ sublevels, whereas the splitting due to the perpendicular field $B_\perp$ is suppressed relative to $\mu_B B_\perp$ by a factor $\mu_B^2 B_z B_\perp/\Delta^2$, which is 3×10^{-10} in our experiment ($B_z \approx 10$ nT, $B_\perp \approx 6$ nT). We separate the splitting due to the edm interaction from that of the magnetic interaction by reversing the directions of the applied electric and magnetic fields, **E** and **B**. The edm part of interest has the symmetry of **E** · **B**, as one might expect for a P-odd, T-odd effect. The suppression of the splitting induced by $B_\perp$ is a critical aspect of the experiment because the motion of the molecules through the electric field generates a 6 nT contribution to $B_\perp$, $B_y^{mot} = Ev/c^2$, which reverses with **E** and therefore has the potential to masquerade as an edm [20]. If $B_\perp$ is entirely motional it does not generate a false edm because the splitting depends on $B_\perp^2$, remaining unchanged when $B_\perp$ reverses. However, if there is also a small y component B_y of the applied magnetic field, the magnitude of $B_\perp$ will change when either **E** or **B** is reversed, leading to an apparent edm given by $\mu_B^3 B_z B_y^{mot} B_y/\Delta^2 E_{eff}$. In our experiment B_y is less than 1 nT and therefore this false d_e is less than $10^{-33}e$ cm. The advantage of a strong tensor polarizability for edm measurements was first demonstrated by Player and Sandars using the $^3P_2$ metastable state of Xe [21]. These two features of heavy polar molecules—large E_{eff} and strong tensor polarizability—give them such excellent suppression of all the known systematic errors that a major improvement in d_e now seems accessible.

Our YbF molecular beam, illustrated in Fig. 3, effuses out of a molybdenum oven containing a mixture of Yb metal and powdered AlF_3 (mass ratio 4:1) heated to ~1500 K. The molecules are detected by dye-laser-induced fluorescence 1 m away from the source, using collection optics with 22% efficiency and a photomultiplier (PMT) of 10% quantum efficiency. The detection laser is tuned to the $F = 0$ component in the $Q(0)$ line of the $A^2\Pi_{1/2}$-$X^2\Sigma^+$ electronic transition (Fig. 2) at
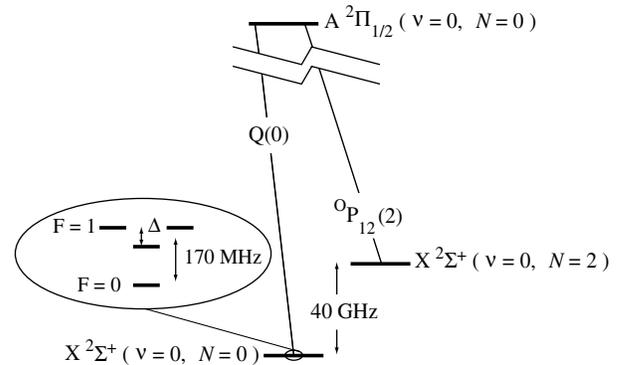

FIG. 2. Important optical transitions $Q(0)$ and $^OP_{12}(2)$ in ^{174}YbF at 553 nm. They are 40 GHz apart. Inset: ground state hyperfine levels $F = 0$, $F = 1$, 170 MHz apart. In static electric field, the $m_F = 0$ sublevel of $F = 1$ is lower than the $m_F = \pm1$ sublevels by an amount Δ.
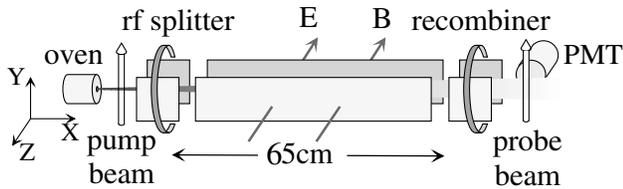
FIG. 3.   Schematic of the YbF molecular beam apparatus.
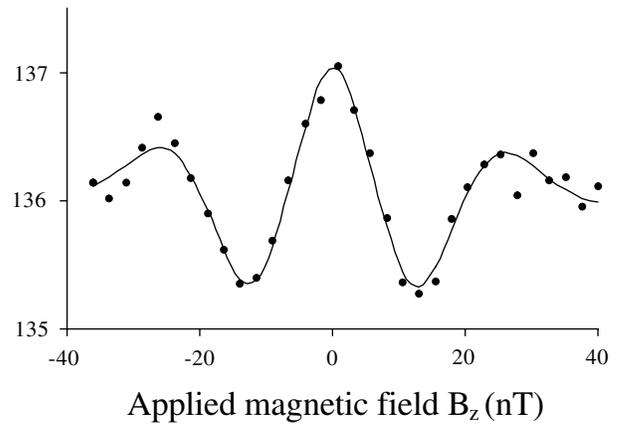

Applied magnetic field B_z (nT)

FIG. 4.   Interference fringe in the $F = 0$ state population versus magnetic field. Each point represents 12 s of integration time. The curve is a velocity-averaged calculation whose only free parameters are the normalization, a field offset, and a sloping background.

553 nm. This induces a count rate of 2.5 kHz due to the ^{174}YbF ground state $F = 0$ molecules and 20 kHz from nearby transitions involving higher rotational states and other isotopes of Yb [18]. There are also backgrounds of ~60 kHz from oven light and ~30 kHz from scattered laser light. The pump beam of Fig. 3 consists of two dye laser beams. One is a 170 MHz red sideband of the probe laser. This removes molecules from all the sublevels of the $F = 1$ ground state by exciting them to $A\,^2\Pi_{1/2}$, which has negligible hyperfine structure [22]. A second dye laser 40 GHz to the red, tuned to the $^{O}P_{12}(2)$ transition (Fig. 2), excites molecules out of the highest hyperfine level of the $N = 2$ rotational manifold. Together these two beams pump molecules into the $F = 0$ ground state, increasing the probe signal to 8 kHz, while emptying out $F = 1$. After pumping, the beam enters an electric field of 3.3 kV/cm, where a 170 MHz rf magnetic field along the x direction excites the molecules to a coherent superposition $\frac{1}{\sqrt{2}}|1, +1\rangle + \frac{1}{\sqrt{2}}|1, -1\rangle$ of the $|F, m_F\rangle$ states. In essence this step is the beam splitter of a molecular interferometer. Next the molecules enter the center of the interferometer, where they evolve for a time T (~1 ms) in combined electric and magnetic fields $(\pm E, \pm B)\hat{z}$, and the two parts of the wave function develop a relative phase shift of $2\varphi = 2(\pm d_e E_{\text{eff}} \mp \mu_B B)T/\hbar$. To measure this phase, a second rf loop drives a transition back to the $F = 0$ state. This is the recombiner in Fig. 3. The state amplitude $\frac{1}{\sqrt{2}}(e^{i\varphi} + e^{-i\varphi})$ gives a final $F = 0$ population proportional to $\cos^2\varphi$. Our detector, measuring fluorescence from molecules excited on the $Q(0)$, $F = 0$ transition, probes this population. Figure 4 shows the central interferometer fringe, measured by scanning the applied magnetic field. To obtain this fringe pattern it is necessary to suppress stray magnetic fields, which we do by means of two 1-mm-thick layers of high-permeability (Ad-MU 80) magnetic shielding. We note that coherences at the 170-MHz rf frequency play no role in this experiment; indeed the two rf loops are driven by two different oscillators to avoid any such Ramsey interferences.

The interferometer is most sensitive to small changes in phase when $\varphi = \pm\pi/4$, i.e., when the magnetic field $\pm B_0$ is set near the points of steepest slope at ± 6 nT on either side of the central fringe. We record the fluorescence in the detector under the eight conditions $\pm E$, $\pm(B_0 \pm \delta B)$, where δB is a carefully calibrated change of 1.6 nT in the magnitude of the applied field. Over a period of

82 sec these eight measurements are repeated 128 times, ordered in such a way as to minimize drifts [23]. The duty factor is 60%. This constitutes a "block" of data. In order to maintain operation near the most sensitive points (the residual field drifts by a few nT over a period of hours), we compare the PMT count rates for $+B_0$ and $-B_0$ (averaged over the other reversals) at the end of each block. Any difference indicates a nonzero residual field, which we null by adding a small proportional bias.

The dye in both lasers has a short lifetime (~20 h) as does the molecular beam source (~10 h). Frequent maintenance and the small signal and large background in our detector make the experiment difficult to operate at present. Nevertheless, we have succeeded in taking 24 h of data over a period of four months, a total of 1758 data blocks containing 1.1×10^{10} counts. In our analysis of the data we extract the change in PMT count rate correlated with every combination of E, B, and δB reversals within every block. Two of these combinations give the phase shift due to the edm interaction and the phase shift due to δB. Their ratio, $d_e E_{\text{eff}}/\mu_B \delta B$, yields the edm result $d_e = (-0.2 \pm 3.2) \times 10^{-26} e$ cm, where the uncertainty (1σ) is due entirely to random pulse counting statistics.

All other combinations of the switched parameters are consistent with zero, suggesting that systematic errors are well under control. As a further check, we took the data in four sets with the connections to the electric plates and/or magnetic field wires reversed, and this too revealed no systematic errors. We monitored the leakage current of the plates, which was always below 10 nA on both plates during data taking. At worst, this current (implausibly looping around the edge of each plate) would cause a false edm of $4 \times 10^{-28} e$ cm. This is small compared with the result of Ref. [7] and is insignificant in the present experiment. With a view to future improvements in sensitivity, we checked the effect of a change in field magnitude

when the electric field was reversed. This changed the resonance strength and therefore the slope of the fringe. When combined with an asymmetry of the magnetic field it led to a false edm, but one that we were fully able to understand and correct on the basis of the separate E-reversal and B-reversal signals. We also ran the experiment with $B_\perp$ intentionally amplified by a factor of 50 and saw that this did not induce any false edm at the level of $5 \times 10^{-26} e$ cm. All these effects were negligible in the present experiment and seem to be well under control for a future measurement below $1 \times 10^{-27} e$ cm. Some details of the systematic checks are available in Ref. [19].

This experiment has demonstrated that a molecular measurement of d_e is less sensitive to stray magnetic fields than any previous experiment by several orders of magnitude. This is due to the high value of $E_{\rm eff}$. The cylindrical symmetry of the electronic state also makes the molecule immune to the motional magnetic field effect. The present result could improve considerably with higher beam intensity. To this end, we have developed a new pulsed source in which YbF molecules are produced by laser ablation and cooled below 20 K by supersonic expansion in a jet of carrier gas. This produces 100 times more population in the $N = 0$ rotational state. On the basis of its current performance, we expect that incorporating the supersonic source into our experiment will reduce the statistical uncertainty in d_e to below $1 \times 10^{-27} e$ cm in 24 h. A competing experiment is in preparation at Yale University by the group of DeMille using a cell of PbO vapor excited to the paramagnetic metastable $a(1)$ state [16]. Trapped cold atoms offer a possible alternative to using polar molecules for measuring d_e [24]. They can have transverse spin relaxation times of seconds instead of milliseconds and the atoms move slowly making the $\mathbf{E} \times \mathbf{v}$ effect much less than in an atomic beam. Even so, the sensitivity to stray magnetic fields and to the trapping fields is a very severe problem for paramagnetic atoms and the experiment would have to use two trapped species, a heavy one for measuring d_e and one for monitoring the magnetic field. Trapped cold molecules offer the best prospect of all, but at present the required techniques are only in a very early stage of development [25].

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