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Testing Time Reversal Symmetry Using Molecules

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Abstract

Some years ago while on sabbatical leave in Oxford I discussed with George Series the experiments in my group to measure the electric dipole moments of the proton and electron using the magnetic resonance in molecules. He asked about the current interest in this kind of measurement and he wanted to find out why we use molecules rather than something simple like the hydrogen atom. After about an hour, George was satisfied with the answers I gave him, but on several occasions after that, he encouraged me to write up some notes about all this. Well George, here they are.

1. Discrete symmetries and fundamental interactions

There are three interrelated reflection symmetries to consider: space inversion (parity, P), interchange of particles and anti-particles (charge conjugation, C), and time reversal (T). Until the early 1950s, it was axiomatic that all of these were symmetry operations, i.e. that the properties of physical systems were invariant under the action of any of them. This point of view was challenged by Purcell and Ramsey [1] who proposed looking for a permanent electric dipole moment (EDM) of the neutron, and by Lee and Yang (1956) [2] who discussed in some detail the possibility that weak interactions may not have parity symmetry. In 1957 a famous experiment [3] by Wu *et al.* showed that weak interactions do indeed violate P symmetry very strongly (in fact, maximally). The method they used was to polarize ^{60}Co nuclei in a cryostat by means of nuclear demagnetisation and to search for a correlation $\langle \boldsymbol{\sigma} \cdot \boldsymbol{p} \rangle$ between the nuclear spin $\boldsymbol{\sigma}$ and the momentum $\boldsymbol{p}$ of the electron emitted in β-decay. They found the strong correlation shown schematically on the left of Fig. 1, in which the electrons prefer the left-handed final state $\langle \boldsymbol{\sigma} \cdot \boldsymbol{p} \rangle < 0$. When space is inverted by reflection in a fictitious "P mirror" we obtain on the right of Fig. 1 the P-transformed version of the experiment. In this version, the decay is right-handed, preferring to have $\langle \boldsymbol{\sigma} \cdot \boldsymbol{p} \rangle > 0$. Because nature prefers the first over the second, the experiment shows that P-symmetry is violated in nature. The left-handedness of weak interactions is now a very well-established phenomenon, tested by numerous experiments in particle, nuclear, and atomic physics [4].

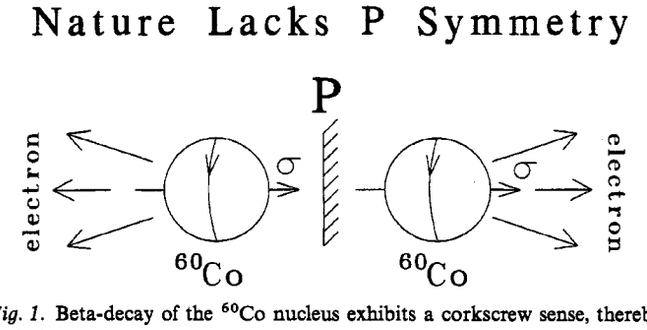

Fig. 1. Beta-decay of the ^{60}Co nucleus exhibits a corkscrew sense, thereby violating P symmetry.

Soon after the discovery of P-violation, Landau (1957) [5] proposed that nature might still be symmetrical but at a deeper level; the combined action CP of charge conjugation and space inversion could be a symmetry operation. His main argument in favour of this idea was the intuitive one that a complete rejection of symmetry "would seem to be extremely strange". By adopting CP one restores the symmetry of *space* because particles which are their own anti-particles have to conserve P. Particles with charge (non-zero quantum numbers) are free to violate P as long as their anti-particles do so in the opposite way, but this is just a property of the charge, not a property of space. In addition, he pointed out, the idea leads naturally to neutrinos and anti-neutrinos that are massless with opposite helicity, and this in turn provides a simple explanation for the known electron energy spectrum in muon decay. The top of Fig. 2 shows the experiment on cobalt, together with a CP-transformed version on the right. CP symmetry would require the rates to be exactly equal and the angular distributions exactly opposite. To demonstrate CP symmetry, we could imagine (somewhat unrealistically) repeating the cobalt experiment with anti-cobalt as in the lower part of Fig. 2. In that case the world under CP inversion is indistinguishable from the real world, as illustrated in Fig. 2, and P-violation conspires with C-violation to produce CP symmetry.

This happy resolution of broken P-symmetry did not last long. In 1964 Christenson *et al.* [6] discovered that the long-lived neutral kaon, K_L^0, can decay occasionally into two pions as well as the more usual three. Since the 2π and 3π final states have opposite CP symmetry, one is forced to conclude that K_L^0 is not an eigenstate of CP and/or the

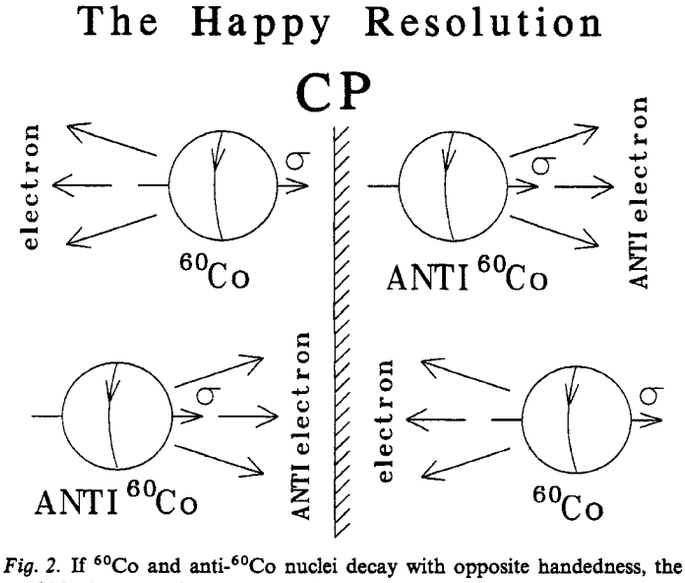

Fig. 2. If ^{60}Co and anti-^{60}Co nuclei decay with opposite handedness, the world is the same after a CP transformation.

decay process itself can change the CP symmetry of the system. In either case CP symmetry is violated. Our current understanding is that K_L^0 is close to being an equal anti-symmetric superposition $1/\sqrt{2}(K^0 - \bar{K}^0)$ of the strong eigenstate K^0 and its charge conjugate $\bar{K}^0$. If the two coefficients were exactly equal to $\pm 1/\sqrt{2}$, this state could not decay into 2π because the amplitudes for $K^0 \to 2\pi$ and $\bar{K}^0 \to 2\pi$ would cancel exactly. The small decay branch into 2π ($\sim 2 \times 10^{-3}$) indicates that this cancellation is not perfect, and it is thought that indeed the K_L^0 spends a little more of its time being a K^0 than it does being a $\bar{K}^0$. This CP non-symmetric state is illustrated in Fig. 3.

Even so, symmetry is still not entirely lost because according to the CPT theorem [7] the even more complicated transformation CPT is necessarily a symmetry operation. This theorem applies to any field theory which satisfies reasonable constraints to do with causality and relativity, so it is generally assumed to be true. In that case T symmetry must be violated in just the right way to make up for CP-violation. The role of T-violation in the K_L^0 can be seen in the following simple model. K^0 and $\bar{K}^0$ have the same mass, width and internal degrees of freedom (incidentally this is a consequence of CPT invariance), nevertheless the dynamic equilibrium of $K^0 \Leftrightarrow \bar{K}^0$ produces the unbalanced state in Fig. 3. This implies (assuming there is nothing more to the problem) that the rate $\bar{K}^0 \to K^0$ is faster than the reverse process $K^0 \to \bar{K}^0$, in violation of T symmetry. In the rest of this essay, I assume the validity of the CPT theorem and refer to T-violation and CP-violation interchangeably.

Let us come back now to the beginning of this section and the proposal by Purcell and Ramsey that the neutron may have an EDM. The upper diagram in Fig. 4 shows that the existence of a neutron EDM implies a violation of P-symmetry since the EDM must reverse relative to the angular momentum σ under space inversion P. Indeed, the initial interest in the neutron EDM was as a test of parity [1]. However, a similar argument, also illustrated in Fig. 4, shows that the EDM violates T-symmetry as well. This makes the search for an EDM a test of T-symmetry in nature [8], which is more interesting since we already know that P-symmetry is violated by weak interactions. When Smith et al. [9] measured the neutron EDM, they obtained a result consistent with zero and although the precision has

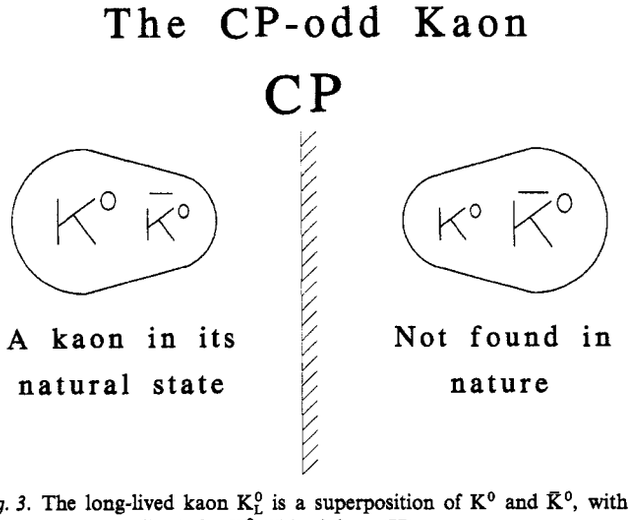

Fig. 3. The long-lived kaon K_L^0 is a superposition of K^0 and $\bar{K}^0$, with a slightly larger amplitude for K^0. This violates CP symmetry.

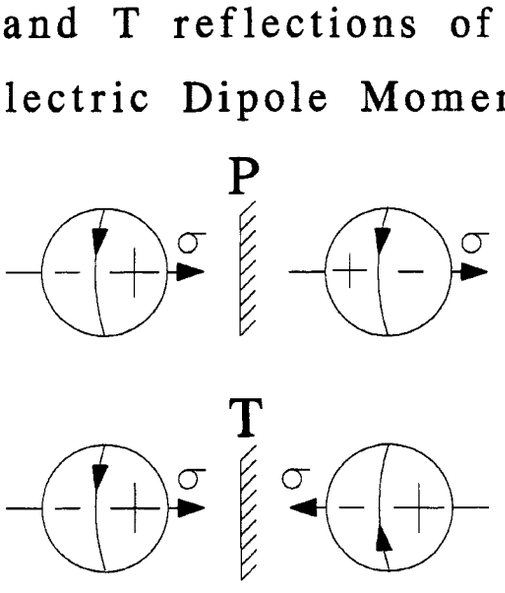

Fig. 4. An elementary system having angular momentum σ and a permanent electric dipole moment. This violates both P and T symmetry.

been improved enormously over the subsequent 40 years, the result is still consistent with zero. Moreover, despite a large number of searches in atomic, nuclear, and particle physics, no instance of T-symmetry violation has been found anywhere apart from the kaon system.

The fact that nature is so selective in violating T-symmetry, imposes a tremendous constraint on the possible theoretical explanations. Out of the dozens of ideas that have been proposed to explain CP-violation since 1964, one has emerged as the most plausible mechanism, and is now part of the "standard model" of elementary particle physics. This is the "KM" mechanism of Kobayashi and Maskawa [10] in which T-violation originates in the complex Yukawa couplings between the three generations of quarks. The decisive virtue of this mechanism is that it can duplicate the CP-violation in K_L^0 without generating any elementary particle EDM large enough to have been detected. There is nothing to say that this KM mechanism is the only one, or even the dominant one, but it is a natural one and it is consistent with experiment. In addition, it would be quite natural within the standard model to have large spontaneous EDMs of the neutron and proton as a result of QCD, the field theory of strong interactions. However, experiment requires that these EDMs be suppressed and this is done by assuming that the "θ parameter" in the theory is less than 10^{-9}. Since θ could reasonably be expected to be roughly unity, the value apparently chosen by nature is astonishingly small. This suggests that θ may be forced to be zero for some reason as yet unclear. There is a variant of QCD in which θ is necessarily zero, but that is not generally accepted as part of the standard model because it requires the existence of a particle called the axion which has so far eluded detection. If the puzzle of the θ parameter could be solved (by the discovery of the axion or by some new theoretical insight into QCD), the standard model would coexist quite comfortably with the current experimental data on T-violation.

The main reason for continuing to make EDM measurements is to test the adequacy of the standard model. The discovery of an EDM of the neutron or proton would indicate either a nonzero value of the θ parameter or some com-

pletely new physics, while an EDM of the electron would be incontrovertible proof of new physics. Although I think that the testing of fundamental laws requires no justification, it is certainly an added incentive to know that most particle theorists believe the standard model to be incomplete. There are many ideas about physics beyond the standard model. For example, the most popular non-standard model at the moment is supersymmetry in which every fermion (e.g. electron, quark, ...) has a bosonic partner (selectron, squark, ...), while the bosons (e.g. photon, gluon, ...) have fermionic partners (photino, gluino, ...). Other ideas include left–right symmetric models and various schemes with more than the minimum number of Higgs particles. As a result of the proliferation of particles, the electric dipole moments of ordinary matter are no longer suppressed as they are in the standard model and most versions of these theories predict relatively huge EDMs for the electron and neutron – EDMs that are comparable with or even much larger than the current experimental limits. Sensitive EDM measurements on low-energy neutrons, atoms and molecules therefore have the potential to provide important new information about the high-energy frontier of fundamental interactions and perhaps to elucidate the present-day uncertainty as to the origin of CP violation in the kaon system.

In this paper I will discuss the possibilities for detecting T-violation by molecular spectroscopy on a diatomic molecule. The basic idea (as with the neutron and with atoms) is to look for an EDM due to some P- and T-violating interactions within the molecule. Whereas the normal molecular dipole moment lies along the internuclear axis and vanishes on average in the laboratory frame of reference (unless an electric field is applied), this T-violating EDM lies along the angular momentum and is nonzero, even in the absence of an electric field. Experiments based on this idea give the best limit on the EDM of the proton and a new experiment being pursued in our laboratory at Sussex promises to push the electron EDM to a very interesting level.

2. Schiff's theorem

Let us suppose that the electron has an intrinsic EDM along its spin axis $\boldsymbol{\sigma}$. One might expect this could be detected by looking at the interaction of an atom with an external field $\boldsymbol{E}_{\rm ext}$, provided the atom has at least one unpaired electron spin. Of course $\boldsymbol{E}_{\rm ext}$ also induces a dipole moment, giving rise to the usual Stark shift, but the two effects can be distinguished simply by reversing the sign of $\boldsymbol{E}_{\rm ext}$. This changes the sign of the T-violating interaction but does not affect the ordinary Stark shift.

Let us approximate an atom as a nonrelativistic collection of point charges (including the nucleus) interacting only through Coulomb forces. To begin, we will take the EDMs of all the charged particles to be zero. In an external electric field, the Hamiltonian can be written as a sum of kinetic and potential terms $\mathscr{H} = T + V$ where V includes the external potential due to $\boldsymbol{E}_{\rm ext}$. Now let particle i at position $\boldsymbol{r}_i$ have a small EDM $\boldsymbol{d}_i$ as well as charge q_i. This EDM interacts with the total electric field $\boldsymbol{E}_i$ at $\boldsymbol{r}_i$ according to

$$\mathscr{H}^1 = -\boldsymbol{d}_i \cdot \boldsymbol{E}_i = \boldsymbol{d}_i \cdot [\nabla_i, V/q_i]. \tag{1}$$

The kinetic term T in our nonrelativistic Hamiltonian $\mathscr{H}^0$ commutes with ∇_i, so we may equally well write

$$\mathscr{H}^1 = \left[\frac{\boldsymbol{d}_i \cdot \nabla_i}{q_i}, \mathscr{H}^0\right]. \tag{2}$$

The first-order contribution of $\mathscr{H}^1$ to the energy of the system is $\langle\psi^0|\mathscr{H}^1|\psi^0\rangle$, where the unperturbed state, $|\psi^0\rangle$, is an eigenfunction of $\mathscr{H}^0$. Because $\mathscr{H}^1$ can be written as a commutator with $\mathscr{H}^0$ [eq. (2)], this diagonal matrix element vanishes. We conclude therefore that in an applied electric field, there is no interaction energy to first order in $\boldsymbol{d}_i$; the atom or molecule has no permanent EDM even if the constituents do!

This result is derived in a famous 1963 paper by Schiff [7] and is often called Schiff's theorem. However, the main point of Schiff's paper was to show how first-order effects *can* occur in spite of the theorem. Schiff proposed two mechanisms. (*i*) The volume effect: the nucleus is not a point, but rather an extended object with distributions of charge and electric dipole moment. (*ii*) The relativistic effect: when the problem is treated relativistically, $\mathscr{H}^1$ can no longer be written as a commutator with $\mathscr{H}^0$. Let us turn first to the case of a nonrelativistic atom in which the nucleus has finite size and EDM, giving rise to a first-order energy shift in an external field through the volume effect.

3. The volume effect

Let the nucleus have charge Q_N and an electric dipole moment $\boldsymbol{D}_N$. The expectation value of $\boldsymbol{D}_N$ must lie along the nuclear angular momentum direction $\hat{\boldsymbol{\sigma}}$, allowing us to define $\langle\boldsymbol{D}_N\rangle = D_N\hat{\boldsymbol{\sigma}}$. Let the charge in an elementary volume d^3x of the nucleus be $Q_N\rho_c(\boldsymbol{x})\,\mathrm{d}^3x$ and let the EDM of the element be $D_N\hat{\boldsymbol{\sigma}}\rho_d(\boldsymbol{x})\,\mathrm{d}^3x$. (We are assuming for the sake of simplicity that the spin of each volume element averages along the axis of total spin.) This defines the normalised distributions ρ_c and ρ_d of charge and EDM.

For a neutral atom or molecule, the net force on the nucleus, $\langle\boldsymbol{F}_N\rangle$ is zero even in the presence of uniform applied electric field $\mathrm{E}_{\rm ext}$, so

$$\langle\boldsymbol{F}_N\rangle = Q_N\int \mathrm{d}^3x\rho_c\langle\psi^0|\boldsymbol{E}(\boldsymbol{x})|\psi^0\rangle = 0. \tag{3}$$

Here $\boldsymbol{E}(\boldsymbol{x})$ is the total electric field acting on the nuclear volume element d^3x due to both $\boldsymbol{E}_{\rm ext}$ and the electrons. The electric dipole interaction energy is

$$\langle\mathscr{H}^1\rangle = -D_N\hat{\boldsymbol{\sigma}}\cdot\int \mathrm{d}^3x\rho_{\rm d}\langle\psi^0|\boldsymbol{E}(\boldsymbol{x})|\psi^0\rangle. \tag{4}$$

Since $\langle\boldsymbol{F}_N\rangle = 0$ we are free to add $\hat{\boldsymbol{\sigma}}\cdot\langle\boldsymbol{F}_N\rangle D_N/Q_N$ to eq. (4), with the result

$$\langle\mathscr{H}^1\rangle = -D_N\hat{\boldsymbol{\sigma}}\cdot\int \mathrm{d}^3x(\rho_d-\rho_c)\langle\psi^0|\boldsymbol{E}(\boldsymbol{x})|\psi^0\rangle. \tag{5}$$

This is an important result to which I will return many times. When $\rho_d = \rho_c$ as in the case of a point nucleus, we recover Schiff's theorem, namely, $\langle\mathscr{H}^1\rangle = 0$. This more microscopic derivation of the theorem shows that the physical cause of the Schiff theorem is the vanishing of $\langle\boldsymbol{F}_N\rangle$, that is, the shielding of the external field by the electrons. In fact every charged particle is shielded from $\boldsymbol{E}_{\rm ext}$ by the polarization of the rest. However, if the distribution of charge and

EDM (ρ_c and ρ_d) are different $\boldsymbol{E}_{\text{ext}}$ is not entirely hidden from the EDM and the residual interaction is given by eq. (5).

3.1. *The field* $\boldsymbol{E}(\boldsymbol{x})$

We turn now to the evaluation of eq. (5), which requires an explicit form for $\boldsymbol{E}(\boldsymbol{x})$:

$$\boldsymbol{E}(\boldsymbol{x}) = \boldsymbol{E}_{\text{ext}} - \frac{e}{4\pi\varepsilon_0} \sum_i \nabla_i \frac{1}{|\boldsymbol{x} - \boldsymbol{r}_i|}. \tag{6}$$

In order to ease the notation, I will set $e/4\pi\varepsilon_0 = 1$ and drop the explicit summation over electrons. Now we expand $|\boldsymbol{x} - \boldsymbol{r}|^{-1}$ as a series of nuclear multipole moments in which only the leading monopole term needs to be written explicitly:

$$\begin{aligned}\boldsymbol{E}(\boldsymbol{x}) &= \boldsymbol{E}_{\text{ext}} + \frac{\hat{r}}{r^2}\,\theta(r - x) + \text{higher multipoles in } \boldsymbol{x} \\ &= \boldsymbol{E}_{\text{ext}} + \frac{\hat{r}}{r^2}\,[1 - \theta(x - r)] + \text{higher multipoles in } \boldsymbol{x},\end{aligned} \tag{7}$$

where θ is the Heaviside step function.

Let us consider the case of a spherical nucleus, which simplifies the mathematics without losing the main point. With spherical ρ_c and ρ_d the higher multipoles of eq. (7) vanish when integrated over nuclear volume d^3x in eq. (5) and they can be omitted. Also the first two terms of eq. (7) contribute nothing because $\boldsymbol{E}_{\text{ext}}$ and $\hat{r}/r^2$ are independent of $\boldsymbol{x}$ and can come outside the integral in eq. (5), leaving

$$\int \mathrm{d}^3x(\rho_d - \rho_c)$$

which vanishes because the distributions are normalized. Thus we can make the very simple effective replacement

$$\boldsymbol{E}(\boldsymbol{x}) \Rightarrow -\frac{\hat{r}}{r^2}\,\theta(x - r), \tag{8}$$

which acts only on electrons *inside* the nucleus, and yields

$$\langle\psi^0 \,|\, \boldsymbol{E}(\boldsymbol{x}) \,|\, \psi^0\rangle \Rightarrow -\int_0^x r^2\,\mathrm{d}r \int_0^{4\pi} \mathrm{d}\Omega\,(\psi^0)^* \frac{\hat{r}}{r^2}\,\psi^0. \tag{9}$$

Since $\hat{r}$ is an odd parity operator, this vanishes when the atom is in free space because ψ^0 is a state of definite parity (neglecting the very small parity impurity due to weak interactions). However, we are interested in the atom subjected to external field $\boldsymbol{E}_{\text{ext}}$ which polarizes the wavefunction. We can expand the polarized ψ^0 in hydrogenic partial waves around the nucleus:

$$\psi^0(r \ll 1) = a_s Y_0^0 + a_p r Y_0^1 + \cdots \tag{10}$$

where the first two terms are the s and p waves and terms of higher angular momentum l involve factors r^l. The spherical harmonic angular factors Y_0^0, Y_0^1 etc. all have $m = 0$ because of cylindrical symmetry around $\boldsymbol{E}_{\text{ext}}$. Since the nuclear radius is $\sim 10^{-4}$ (we are still using atomic units), the contributions of higher partial waves to $\langle\boldsymbol{E}(\boldsymbol{x})\rangle$ are strongly suppressed and we will assume they are negligible in comparison with the s–p contribution. So, finally, the mean field at position $\mathbf{x}$ within the nucleus can be replaced by

$$\langle\psi^0 \,|\, \boldsymbol{E}(\boldsymbol{x}) \,|\, \psi^0\rangle \Rightarrow -x^2\,\frac{a_s a_p}{\sqrt{3}}\,\hat{\lambda}, \tag{11}$$

where $\hat{\lambda}$ is the direction of $\boldsymbol{E}_{\text{ext}}$. Now that we have an effective expression for the field, we can return to the main theme; evaluation of the volume effect interaction given in eq. (5).

3.2. *Effective interaction and Schiff moment*

Inserting eq. (11) into eq. (5) we obtain $\langle\mathscr{H}^1\rangle$ as a product of a nuclear factor and an electronic factor:

$$\langle\mathscr{H}^1\rangle = \left(\tfrac{1}{6}D_N \int \mathrm{d}^3x(\rho_d - \rho_c)z^2\hat{\boldsymbol{\sigma}}\right) \cdot \left(6\,\frac{a_s a_p}{\sqrt{3}}\,\hat{\lambda}\right). \tag{12}$$

The nuclear part is know as the Schiff moment $Q\hat{\boldsymbol{\sigma}}$

$$Q = \tfrac{1}{6}D_N \int \mathrm{d}^3x(\rho_d - \rho_c)x^2 \tag{13}$$

It is interesting to write the electronic part in a different way. Consider the derivative $[\nabla\nabla^2 V]_0$ of the potential V evaluated at the origin. This is related to the electron density through Poisson's equation, yielding

$$\nabla\nabla^2 V = -4\pi\nabla\rho = 4\pi\nabla\,|\,\psi^0\,|^2 \tag{14}$$

(still in cgs atomic units). Using this together with eq. (10) we obtain

$$[\nabla\nabla^2 V]_0 = 6\,\frac{a_s a_p}{\sqrt{3}}\,\hat{\lambda}. \tag{15}$$

Collecting together eqs (12), (13) and (15) we see that

$$\langle\mathscr{H}^1\rangle = Q\hat{\boldsymbol{\sigma}} \cdot [\nabla\nabla^2 V]_0 \tag{16}$$

This interaction is what remains as a result of the finite nuclear size when the electric dipole interaction $-D_N\hat{\boldsymbol{\sigma}} \cdot \boldsymbol{E}_{\text{ext}}$ is shielded by the electrons.

One can recognize the Schiff interaction as the first non-zero term in a Taylor expansion. Loosely speaking we expand $\langle\psi^0 \,|\, \boldsymbol{E}(\boldsymbol{x}) \,|\, \psi^0\rangle$ in eq. (5) around the centre of charge as

$$\langle\boldsymbol{E}(\boldsymbol{x})\rangle \sim E(0) + xE' + x^2E'' + \cdots \tag{17}$$

in which $E(0) = 0$ because the net force on the nucleus must be zero. The first-derivative term xE' contributes nothing to the integral in eq. (5) over the spherical nucleus because it is an odd function of x. Hence the leading effect is due to x^2E'' which yields in eq. (5)

$$\langle\mathscr{H}^1\rangle = -D_N\hat{\boldsymbol{\sigma}} \cdot \langle(\rho_d - \rho_c)x^2E''\rangle \tag{18}$$

Since $E'' \sim -V'''$, this has all the essential features of the more rigorous eq. (16).

In deriving the Schiff moment Q [eq. (13)], we assumed a spherical distribution $\rho_d\hat{\boldsymbol{\sigma}}$ of electric dipole moment due, for example, to an intrinsic EDM of the proton or neutron, but the Schiff moment is actually more general. Even when the nucleons have no EDM, it is still possible for the nucleus to acquire an electric dipole distortion if the forces between the nucleons violate P and T symmetry. If the dipolar part of the charge distribution is $\delta(\boldsymbol{x})$, we can define the distributed

EDM as $\boldsymbol{d} = x\delta(\boldsymbol{x})$, and the total EDM as

$$D_N\hat{\boldsymbol{\sigma}} = \int \mathrm{d}^3x\boldsymbol{d}.$$

It can be shown (from the V''' term in a Taylor expansion of $V(\boldsymbol{x})$ [11]) that once again there is a volume effect of the form given in eq. (16), with the Schiff moment being given now by

$$Q = \frac{1}{6}\int \mathrm{d}^3x(\tfrac{3}{5}\boldsymbol{d} - D_N\rho_c\hat{\boldsymbol{\sigma}})x^2 \tag{19}$$

At this point, it is probably useful to summarize the main points about the volume effect. It leads to an effective interaction, given by eq. (16), which is the product of two parts. The first is the Schiff moment $Q\hat{\boldsymbol{\sigma}}$. Q is of order $D_N r_N^2$, where r_N is the nuclear radius and D_N is the nuclear EDM, and it depends also on the details of the distributions of charge and electric dipole moment [see eqs (13) and (19)]. A nonzero value for Q indicates an electric dipole distortion of the nucleus or its constituents due P- and T-violating interactions. The second part is the electronic factor $[\nabla\nabla^2 V]_0$, which is proportional to the gradient of electron density at the nucleus. This can be related to the $l = 0$ and $l = 1$ parts of the electronic wavefunction by eq. (15). In systems where neither the s nor the p part of the wavefunction is large, the Schiff interaction is suppressed since higher partial waves have a factor $(r_N/a_0)^l$ at the origin.

3.3. *Strength of the volume effect in atoms and molecules*

It remains for us to determine the sizes of a_s and a_p defined by eq. (10). For an electron nominally in an s state, the s–p admixture induced by the applied electric field is given to first order in $\boldsymbol{E}_{\mathrm{ext}}$ by $|s\rangle' = |s\rangle + \varepsilon|np\rangle$, where

$$\varepsilon = E_{\mathrm{ext}}\sum_n \frac{\langle s|ez|np\rangle}{W_{np} - W_s}. \tag{20}$$

For a rough numerical estimate, we take the sum to be about one atomic unit, and hence $\varepsilon\hat{\lambda} \sim \boldsymbol{E}_{\mathrm{ext}}$ in atomic units (1 au of electric field is $5 \times 10^9\,\mathrm{V\,cm^{-1}}$). Close to the origin, $|s\rangle \sim Z^{1/2}Y_0^0$ while $|np\rangle \sim Z^{3/2}rY_0^1$ (more exact expressions for the s and p wavefunctions at small radius can be found in [12]), so $a_s a_p\hat{\lambda} \sim Z^2\boldsymbol{E}_{\mathrm{ext}}$. Equation (15) then gives $[\nabla\nabla^2 V]_0 \sim 4Z^2\boldsymbol{E}_{\mathrm{ext}}$. With $Q \sim D_N r_N^2$ it follows from eq. (16) that the effective interaction is

$$\langle\mathscr{H}^1\rangle \sim D_N\hat{\boldsymbol{\sigma}}\cdot 4Z^2(r_N/a_0)^2\boldsymbol{E}_{\mathrm{ext}}, \tag{21}$$

where we have written the Bohr radius explicitly in order to return to normal units. Since $r_N/a_0 \sim 10^{-4}$, we find that in a heavy atom with $Z = 80$, the Schiff shielding effectively suppresses the applied electric field by a factor of order 3×10^{-4}. Heavy atoms are better than light ones because of the factor Z^2, which is due to the enhanced electron density near a highly charged nucleus. A more careful calculation shows that there is an additional relativistic enhancement factor K_r, which can be as large as 10 for heavy atoms [12], but that is beyond the scope of this essay.

A heavy atom becomes much more sensitive to the volume effect when it is on one end of a diatomic polar molecule because the electronic cloud is then strongly polarized along the internuclear axis $\hat{\lambda}$. If there is no external electric field, the angular distribution of this axis averages $[\nabla\nabla^2 V]_0$ to zero, but it is easy to apply enough field to polarize $\hat{\lambda}$ completely in a heavy molecule. Taking $\varepsilon \sim 0.1$ as a typical value, eq. (15) gives $[\nabla\nabla^2 V]_0 \sim 0.4Z^2\hat{\lambda}$ (au), and this is virtually independent of the applied field once $\hat{\lambda}$ is largely polarized along $\boldsymbol{E}_{\mathrm{ext}}$. The same quantity in an atom is $\sim 4Z^2\boldsymbol{E}_{\mathrm{ext}}$ which means that the volume effect is larger by a factor $\sim 1/(10E_{\mathrm{ext}})$ (a.u.) when the atom is part of a polar molecule:

$$\frac{\langle\mathscr{H}^1\rangle_{\mathrm{molecule}}}{\langle\mathscr{H}^1\rangle_{\mathrm{atom}}} \sim \frac{5 \times 10^8}{E_{\mathrm{ext}}} \tag{22}$$

where E_{ext} is the field in $\mathrm{V\,cm^{-1}}$ applied to the atom. For a typical field of $5\,\mathrm{kV\,cm^{-1}}$ this is a factor of 10^5, quite an improvement in sensitivity! Indeed, this factor can be large enough in some cases to overcome the shielding so that the effective electric field $\langle\mathscr{H}^1\rangle_{\mathrm{molecule}}/D_N$ is comparable with the applied field E_{ext} or even larger.

4. The magnetic effect

So far we have considered the finite nuclear size as a mechanism for avoiding Schiff's theorem. This is the main effect to be considered if the T-violation occurs in the nucleus, but if it is the electron that has an EDM the volume effect does not help because electrons are point particles. In that case we must rely on relativity, the other effect pointed out by Schiff, to overcome the electrostatic shielding.

Since the effect of interest now is relativistic, we take as our zeroth order Hamiltonian the Dirac equation

$$\mathscr{H}^0 = \sum_{\mathrm{electrons}} \beta mc^2 + \boldsymbol{\alpha}\cdot c\boldsymbol{p} + V, \tag{23}$$

where V includes the uniform applied field $\boldsymbol{E}_{\mathrm{ext}}$ as well as the atomic Coulomb interaction. Now we would like to let the electron have an EDM d_e and to treat the electric dipole interaction as a perturbation, but what is the correct relativistic form of the interaction? According to the Dirac equation, the magnetic dipole interaction for an electron has the Hamiltonian density $i\mu_B(\frac{1}{2}\bar{\psi}\gamma_\mu\gamma_\nu\psi F_{\mu\nu})$ [13]. Since an electric dipole has the opposite P and T properties we multiply this by $i\gamma_5$ (following Salpeter [14]) to obtain the electric dipole interaction

$$\mathscr{H}_{E1} = -d_e(\tfrac{1}{2}\bar{\psi}\gamma_5\gamma_\mu\gamma_\nu\psi F_{\mu\nu}) \tag{24}$$

where d_e is the supposed EDM of the electron. Converting from covariant notation to Dirac notation, the interaction becomes

$$\mathscr{H}_{E1} = -d_e\beta(\boldsymbol{\Sigma}\cdot\boldsymbol{E} + ic\boldsymbol{\alpha}\cdot\boldsymbol{B}). \tag{25}$$

Here β, $\boldsymbol{\Sigma}$ and $\boldsymbol{\alpha}$ are the standard Dirac operators. To be concrete, let us choose the representation

$$\beta = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix},\quad \boldsymbol{\Sigma} = \begin{bmatrix} \boldsymbol{\sigma} & 0 \\ 0 & \boldsymbol{\sigma} \end{bmatrix},\quad \boldsymbol{\alpha} = \begin{bmatrix} 0 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & 0 \end{bmatrix}, \tag{26}$$

$\boldsymbol{\sigma}$ being the vector whose Cartesian components are the three Pauli matrices. The familiar classical expression $-\boldsymbol{d}\cdot(\boldsymbol{E} + \boldsymbol{v}\times\boldsymbol{B})$ is the non-relativistic limit of this interaction. In atoms and molecules with an unpaired electron, the electric term in eq. (25) turns out to be much larger then the magnetic one, so we take as our perturbation

$$\mathscr{H}^1 = -d_e\beta\boldsymbol{\Sigma}\cdot\boldsymbol{E}. \tag{27}$$

This is very similar to the operator $-d_e\boldsymbol{\Sigma}\cdot\boldsymbol{E}$, which one might have guessed, but has the additional factor β. It will turn out that this makes all the difference. In a heavy atom we should really sum over electrons, but to keep the notation simple I will just ignore that. The EDM interaction energy of the atom is therefore

$$\langle\mathscr{H}^1\rangle = \langle\psi^0|-d_e\beta\boldsymbol{\Sigma}\cdot\boldsymbol{E}|\psi^0\rangle. \tag{28}$$

Following exactly the path used to derive the Schiff theorem in Section 2 let us note that the total field $\boldsymbol{E}$ on the electron can be written as $-[\nabla, V/q]$ where V is the potential energy in eq. (23). From this it follows that

$$\boldsymbol{\Sigma}\cdot\boldsymbol{E} = -[\boldsymbol{\Sigma}\cdot\nabla, \mathscr{H}^0/q], \tag{29}$$

where $\mathscr{H}^0$ is the relativistic Hamiltonian of eq. (23). But this commutator vanishes, which means that the nonrelativistic interaction $\langle\psi^0|d_e\boldsymbol{\Sigma}\cdot\boldsymbol{E}|\psi^0\rangle = 0$, (i.e. without the β) vanishes in accordance with Schiff's theorem. Finally, since this matrix element is zero, we can add it to the right-hand side of eq. (28) to obtain

$$\langle\mathscr{H}^1\rangle = \langle\psi^0|(1-\beta)d_e\boldsymbol{\Sigma}\cdot\boldsymbol{E}|\psi^0\rangle, \tag{30}$$

which can be written explicitly as

$$\langle\mathscr{H}^1\rangle = \left\langle\psi^0\left|\begin{matrix}0 & 0\\ 0 & 2d_e\boldsymbol{\sigma}\cdot\boldsymbol{E}\end{matrix}\right|\psi^0\right\rangle. \tag{31}$$

This form of the interaction energy obviously satisfies Schiff's theorem since it vanishes in the nonrelativistic limit.

The integral in eq. (31) involves only the small components of the electron wavefunction so it is most sensitive to regions where the speed of the electron approaches the speed of light. This only happens very close to the nucleus where the total field $\boldsymbol{E}$ on the electron can be reasonably approximated by the Coulomb field $Z\hat{r}/r^2$. Writing the two small components of $|\psi^0\rangle$ as $|g^0\rangle$ we obtain the approximation

$$\langle\mathscr{H}^1\rangle = \left\langle g^0\left|2d_e\boldsymbol{\sigma}\cdot\frac{Z\hat{r}}{r^2}\right|g^0\right\rangle. \tag{32}$$

This vanishes unless g^0 is a state of mixed parity (because $\hat{r}$ is an odd-parity operator), which just means that the electric dipole interaction requires an applied field. As in Section 3.3, let us take the electron to be nominally in an s state with a small admixture of p state due to the external field:

$$|g^0\rangle = |g^0_{s_{1/2}}\rangle + \varepsilon|g^0_{p_{1/2}}\rangle + \cdots \tag{33}$$

Close to the nucleus where $r \leqslant 1/Z$, which is the region of interest for us, $|g^0_{p_{1/2}}\rangle$ is roughly independent of the radius, while $|g^0_{s_{1/2}}\rangle \propto r$. We ignore the $p_{3/2}$ part of g^0 because it varies as r^2 and is very small. In this case, the radial integral is a little more complicated than the one involved in eq. (9) so I simply quote the result [15] that

$$\left\langle g^0_{s_{1/2}}\left|2d_e\boldsymbol{\sigma}\cdot\frac{Z\hat{r}}{r^2}\right|g^0_{p_{1/2}}\right\rangle \sim 4Z(Z\alpha)^2 d_e. \tag{34}$$

The factor of $(Z\alpha)^2$ is due to the size of the small components of the wavefunction over the region near the nucleus, while Z is due to the Coulomb field. Once again we take $\varepsilon\hat{\boldsymbol{\lambda}} \sim \boldsymbol{E}_{\text{ext}}$, then eqs (32)–(34) and give

$$\langle\mathscr{H}^1\rangle \sim 8Z(Z\alpha)^2 d_e E_{\text{ext}}\hat{\boldsymbol{\sigma}}\cdot\hat{\boldsymbol{\lambda}}, \tag{35}$$

where $\hat{\boldsymbol{\sigma}}$ is now a unit vector along the electron spin direction and $\hat{\boldsymbol{\lambda}}$ lies along the applied electric field. For a heavy atom, say $Z = 70$, eq. (35) gives

$$\langle\mathscr{H}^1\rangle_{\text{atom}} \sim 100 d_e\hat{\boldsymbol{\sigma}}\cdot\boldsymbol{E}_{\text{ext}}, \tag{36}$$

which means that in spite of Schiff's theorem, the EDM interaction in a heavy atom is 100 times larger than that of the free electron! This marvellous enhancement factor was first noticed in 1965 by Sandars [16].

The situation in heavy polar molecules is even move favourable since we have typically $\varepsilon \sim 0.1$. In that case

$$\langle\mathscr{H}^1\rangle_{\text{molecule}} \sim 10 d_e\hat{\boldsymbol{\sigma}}\cdot\hat{\boldsymbol{\lambda}}, \tag{37}$$

which is larger by a factor $\sim 5\times 10^8/E_{\text{ext}}$ than the atomic interaction. This enhancement of a molecule relative to an atom is exactly the factor we encountered in Section 3.3 for the volume effect; it is simply a result of the large mixing of s and p orbitals in a polar molecule. Our conclusion is that if the electron has an EDM, the effect in a heavy polar molecule should be similar to that of the free electron in a field of 10 au $= 5\times 10^{10}\,\text{V cm}^{-1}$. This is a huge effective electric field.

5. The search for a nuclear EDM

The most sensitive searches for a nuclear EDM employ three different systems: the neutron [17], the Hg atom [18], and the TlF molecule [19]. Any EDM of the bare neutron should have a normal electric dipole interaction with the applied electric field because there are no electrons to shield it. The Hg atom on the other hand relies on the volume effect, for which the effective interaction given in eq. (16) is proportional to the Schiff moment $Q\hat{\boldsymbol{\sigma}}$ of the Hg nucleus. Consequently the effective electric field at the nucleus ($\langle\mathscr{H}^1/D_N\rangle$) is about four orders of magnitude smaller than the applied field, as I show in Section 3.3. However, the Hg experiment makes up for this disadvantage by having a narrower nuclear resonance line which can detect much smaller electric dipole interactions $\mathscr{H}^1$. At present the these two experiments have similar sensitivities to fundamental T-violating forces and they are the most sensitive of their kind.

The experiment on TlF also relies on the volume effect. It is a search for the Schiff moment of the Tl nucleus. But because TlF is a polar molecule, there is a strong s–p mixing which largely makes up for the shielding and results in an effective electric field of $\sim 10\,\text{kV cm}^{-1}$ – roughly the same as the applied field. In this respect then, the molecule and the neutron are similar. Unfortunately the width of the nuclear resonance line in TlF is approximately 100 Hz, as compared with 7 mHz for the neutron. Other factors enter into the final sensitivity of the experiment allowing TlF to catch up most but not all of the deficit.

If the TlF molecules could somehow be cooled to a few K there could be an enormous improvement in the sensitivity of the experiment for two reasons. First, the velocity of the molecules would be lower, allowing them to spend longer in the interaction region and resulting in a narrower resonance line. Second, the population would be larger in the rotational state of interest because of a more favourable Boltzman factor. The most obvious idea is to try laser cooling, which has been so successful in recent years with atoms. This is based on the scattering of many laser photons through

spontaneous emission. Unfortunately, no one knows how to extend the idea to molecules. The difficulty is that after a few excitations the molecule is optically pumped out of the rotation/vibration state of interest and is no longer near resonance with the light. The standard cooling and trapping technique for molecules is matrix isolation [20] in which the molecules are held inside frozen inert gas, but this is not ideal for EDM experiments because of the perturbations due to the inert gas matrix. An exciting recent development in this direction is the demonstration by Arndt *et al.* [21] that individual atoms trapped in bubbles within solid helium can have long spin relaxation times. This kind of technique could well lead to a new generation of ultra sensitive EDM experiments.

The TlF experiment [19] involved a spin-polarised beam of TlF molecules, passing through an electric field $\boldsymbol{E}_{\text{ext}}$. Nuclear magnetic resonance (NMR) was performed on the Tl nucleus and we looked for a linear Stark effect by searching for a shift of the NMR frequency when $\boldsymbol{E}_{\text{ext}}$ was reversed. The interaction of the Tl nuclear spin $\frac{1}{2}\hbar\hat{\boldsymbol{\sigma}}$ with the rest of the molecule can be described by the effective Hamiltonian

$$\mathscr{H} = -\mu_{\text{Tl}}\hat{\boldsymbol{\sigma}} \cdot \boldsymbol{B}_0 - Dh\hat{\boldsymbol{\sigma}} \cdot \hat{\boldsymbol{\lambda}}. \tag{38}$$

The first term is the usual (T-conserving) hyperfine interaction of the nuclear magnetic dipole moment $\mu_{\text{Tl}}\hat{\boldsymbol{\sigma}}$ with the internal magnetic field $\boldsymbol{B}_0$ of the molecule. The second term describes the P&T-violating electric dipole interaction that we are interested in. Here $\hat{\boldsymbol{\lambda}}$ is a unit vector pointing from the Tl nucleus to the F nucleus, D is a measure of T-violation in TlF and h is Planck's constant. In free space, such an interaction would tip the internuclear axis, giving it a small projection along $\hat{\boldsymbol{\sigma}}$, hence producing a small permanent EDM. We prefer however to detect this interaction by applying the strong field $\boldsymbol{E}_{\text{ext}}$, which substantially polarizes $\hat{\boldsymbol{\lambda}}$, giving an energy of the form $\hat{\boldsymbol{\sigma}} \cdot \boldsymbol{E}_{\text{ext}}$ as given in eq. (21). This appears as a shift of the NMR frequency when $\boldsymbol{E}_{\text{ext}}$ is reversed. Details of the experimental method can be found in Ref. [19].

The result of the experiment was

$$D = -0.13 \pm 0.22\,\text{mHz} \tag{39}$$

for the T-violating coupling constant in eq. (38). In order to extract a Schiff moment of the Tl nucleus from this result, it is necessary to calculate the electronic integral $\langle \nabla V^2 V \rangle$ [see eq. (16)]. This calculation has been done by Coveney and Sandars [22] and when combined with our measurement of D gives

$$Q(^{205}\text{Tl}) = (2.3 \pm 3.9) \times 10^{-10}\,\text{e.fm}^3. \tag{40}$$

The experiment on ^{199}Hg gives

$$|Q(^{199}\text{Hg})| < 0.22 \times 10^{-10}\,\text{e.fm}^3, \tag{41}$$

while the most recent neutron EDM measurement yields

$$d_n = (-3 \pm 5) \times 10^{-13}\,\text{e.fm}. \tag{42}$$

Since the Hg nuclear radius squared is $\sim 50\,\text{fm}^2$, these last two results are of comparable precision. These are small limits indeed: for example, the dipole distortion of the neutron is less than 10^{-12} of its diameter.

When the experimental limits given above are used to constrain possible T-violating elementary particle interactions, the exact sensitivity of each experiment to a given hypothetical effect depends upon the details of the effect. However, the general trend is that Hg and the neutron are similar, while the TlF constraint is typically a factor of ten weaker. The level of sensitivity is significant for particle physics. For example, in all three experiments the EDM observed is smaller than one should expect if supersymmetry is broken near the electroweak energy scale. The nature and extent of T-violation is sufficiently uncertain at present that any one of these experiments could be on the verge of detecting a non-zero nuclear EDM.

6. The search for the electron EDM

The two most sensitive measurements of the electron EDM use the paramagnetic atoms Tl [23] and Cs [24]. The result from Cs is

$$d_e = (-1.5 \pm 5.5 \pm 1.5) \times 10^{-26}\,\text{e.cm} \tag{43}$$

and from Tl

$$d_e = (-1.8 \pm 1.2 \pm 1.0) \times 10^{-27}\,\text{e.cm}. \tag{44}$$

It is interesting to compare these results with the predictions of the various theories of elementary particle physics [25, 26] as listed in Table I. Electron EDM measurements are clearly starting to place significant constraints on physics beyond the standard model and real possibilities exist for the discovery of a nonzero effect. This is true in the case of the nuclear EDM as well, but the electron EDM is much more directly connected to underlying fundamental interactions and the predictions of theory are therefore more certain.

These experiments rely on the magnetic effect outlined in Section 4 to overcome the Schiff screening of the applied field and indeed to enhance the interaction in comparison with the bare electron. For Tl the enhancement factor is -585 ± 30 [27], which is to say the interaction between the Tl atom and the applied field is $+d_e\hat{\boldsymbol{\sigma}} \cdot 585\boldsymbol{E}_{\text{ext}}$. (The sign of the enhancement factor is an interesting story but beyond the scope of this paper.) In the experiment a $100\,\text{kV cm}^{-1}$ field was applied to a Tl atomic beam, resulting in an effective field on the electron spin of $\sim 60\,\text{MV cm}^{-1}$. The Cs experiment on the other hand was done in a cell of vapour which could not support a field above $4\,\text{kV cm}^{-1}$, giving an effective field on the Cs electron spin of $0.5\,\text{MV cm}^{-1}$, a hundred times smaller than in the Tl experiment. This difference is the primary reason that the Tl experiment gives the more precise measurement of the electron EDM.

In a polar molecule it should be possible to achieve an effective electric field of $\sim 50\,\text{GV cm}^{-1}$, a thousand times bigger than the Tl field, as shown in Section 4. Moreover, the width of the electron spin resonance in a molecular

Table I. *Theoretical predictions for the electron EDM in various models of elementary particle physics*

Type of model	Magnitude of electron EDM (e.cm)
Standard model	$<10^{-38}$
Supersymmetry	$<10^{-27}$
Left-right symmetric models	10^{-26}–10^{-28}
Lepton flavour-changing models	10^{-26}–10^{-29}
Higgs models	10^{-27}–10^{-28}

beam should be similar to the width of the Tl atomic beam resonance. Thus it seems possible to achieve a large improvement in the determination of the electron EDM by working with a beam of suitable molecules. Candidates should be diatomic for simplicity, polar for large s–p mixing, heavy for large $Z^3\alpha^2$ and paramagnetic because the interaction is proportional to the electon spin. The most promising molecules for an experiment seem to be YbF and BaF with effective electric fields on the electron spin of 30, and 9 GV cm^{-1} respectively [28]. Until recently, rather little was known about YbF but now our group has managed to produce a beam of these molecular radicals and to measure all the structural information needed to conduct an EDM experiment [29, 30]. The electronic, vibrational and rotational ground state $X^2\Sigma$ ($v = 0$, $N = 0$) of ^{174}YbF has only two angular momenta, the electron spin (1/2) and the fluorine nuclear spin (1/2). These form a singlet F = 0 and a triplet F = 1 which are separated by 170.254 MHz. We have conducted a brief pilot EDM experiment on this $F = 0 \Rightarrow 1$ transition with a rather short 1 cm interaction region. This gives an uncertainty in the electron EDM of order 1×10^{-25} e.cm in a day. We are now increasing the length of the interaction region to obtain a hundred times narrower resonance and the prospects seem good for a new value of the electron EDM in the near future.

7. Conclusion

In this short article I have covered the main points about testing time reversal symmetry using molecules. First, I have discussed how the search for permanent electric dipole moments allows us to probe fundamental interactions beyond the electroweak scale, thereby learning about physics beyond the standard model. These electric dipole moments are particularly sensitive to new physics because there is no "background" from interactions within the standard model. Next, I have described the volume and magnetic effects of Schiff which allow the nuclear and electronic dipoles to be detected in atoms and molecules despite the electrostatic shielding of the applied field. We saw that polar molecules are generically more sensitive than atoms to both these effects because of the strong s-p mixing in the electronic wavefunction. Finally I have outlined the current status of both nuclear and electronic EDM tests, paying particular attention to the role that molecules are playing. On the nuclear front, the long nuclear coherence times available with stored neutrons or Hg atoms in a cell cannot be achieved with molecular beams and this has prevented them from taking the lead. Electron spin resonances on the other hand do not have such long coherence times and the molecular beam technique with YbF seems likely to produce a significant improvement in the electron EDM. A further large improvement is conceivable if techniques can be developed to trap molecules in a cold environment such as solid helium.

I close with a quotation from Steven Weinberg [31]. "*. . . it may be that the next exciting thing to come along will be the discovery of a neutron or atomic or electron electric dipole moment. These electric dipole moments . . . seem to me to offer one of the most exciting possibilities for progress in particle physics.*"

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