University of New South Wales
School of Physics

Honours Thesis

Contribution of Core Nucleons to CP-violating Nuclear Magnetic Quadrupole Moment

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Abstract

Investigations into CP violation in the hadron sector may be done by studying CP-violating electromagnetic moments in molecular systems. Recently there have been experimental developments in using paramagnetic molecules to observe CP-violating phenomena, with a recent experiment strengthening the current limit of the electron electric dipole moment (EDM). These paramagnetic molecules allow us to study the lowest CP-violating magnetic moment of the nucleus, the nuclear magnetic quadrupole moment (MQM). The MQM is expected to improve the limits on fundamental CP-violating properties. In this thesis we focus on finding the contribution of core polarisation to the nuclear MQM. We perform calculations of of T,$P-$odd effects in the paramagnetic molecules TaN, ThO, ThF^+,HfF^+,YbF,HgF and BaF induced by MQMs with the core contribution. We compare these results to the valence only contribution and find that the core contribution has a large impact for deformed nuclei. We express the nuclear MQM and molecular frequency shifts in terms of the CP-violating quark EDMs, quark chromo-DMs and the $\tilde{\theta}$ term in QCD.

Contents

Note on Units

In this thesis we use natural units such that $\hbar = c = 1$. We present our results using experimental convention. For example we use units $e \cdot \mathrm{cm}$ for the electric dipole moments and express the energy shift in units of frequency.

Acknowledgments

I would like to thank and express my deep gratitude to Prof. Victor Flambaum and Julian Berengut for introducing me to this interesting problem and for their help and supervision on this thesis.

I would also like to thank Ben Roberts and Yevgeny Stadnik for their helpful discussions and insight, particularly in the context of the axion and its relation to the current work.

Also I would like to thank my peers in honours for their support and friendship, making it a great year all around.

Preface

The purpose of this thesis is to convince the reader of the completeness of the work conducted and the implications of the subsequent results. It is also intended to give an overview on the topic and review the relevant, current literature to create an appropriate setting for this research topic.

The structure of this thesis is to make it as self contained as possible. It can be separated into three parts, the first is a review of the literature and history of the field with all necessary information for the current research (Chapters 1 and 2). The second part is the calculations and major results of the thesis (Chapter 3). The final part is the discussion of results and conclusion (Chapter 4).

As this research topic is quite theoretically involved, to facilitate to reader I have included an appendix to present important yet laborious concepts and calculations.

At the beginning of Chapter 3 I have written a statement of originality to make clear which work is original in this thesis.

Chapter 1

Introduction

In nature there are three fundamental discrete symmetry transformations, parity (P), time reversal (T) and charge conjugation (C). Parity is the inversion of spacial coordinates ($\mathbf{r} \overset{P}{\mapsto} -\mathbf{r}$), time reversal is the transformation which reverses the instantaneous velocity of all particles concerned ($t \overset{T}{\mapsto} -t$) and charge conjugation is the replacement of all particles with their corresponding anti-particles ($m \overset{C}{\mapsto} \bar{m}$, an example of these transformations are shown in Figure 1.1). Up until the 1950s it was unknown whether these symmetries were invariant in nature or if their violation had yet to be observed. It was known there was no violation in the electromagnetic or strong interaction (the discovery of strong CP violation came much later, see Section 2.4) however there was no reason for the weak force to conserve these symmetries. Although many had their suspicions [1, 2] there was no evidence until it was experimentally observed by Wu *et al.* in 1957 [3] that P and C were violated in the Beta decay of ^{60}Co. The prediction of this result a year previous rewarded Lee and Yang with the Nobel prize for their work [4] on parity violating mechanisms in the electroweak theory. Since then, the search for the violation of discrete symmetries in other mechanisms has rapidly grown and now spans many fields including atomic, nuclear and high energy particle physics.

In this chapter we will introduce the concept of CP violation and discuss instances where it has been observed and current attempts to observe this violation in different systems. We will also discuss the inconsistencies with the current Standard Model and how understanding CP violation could lead to theories which amend these inconsistencies.

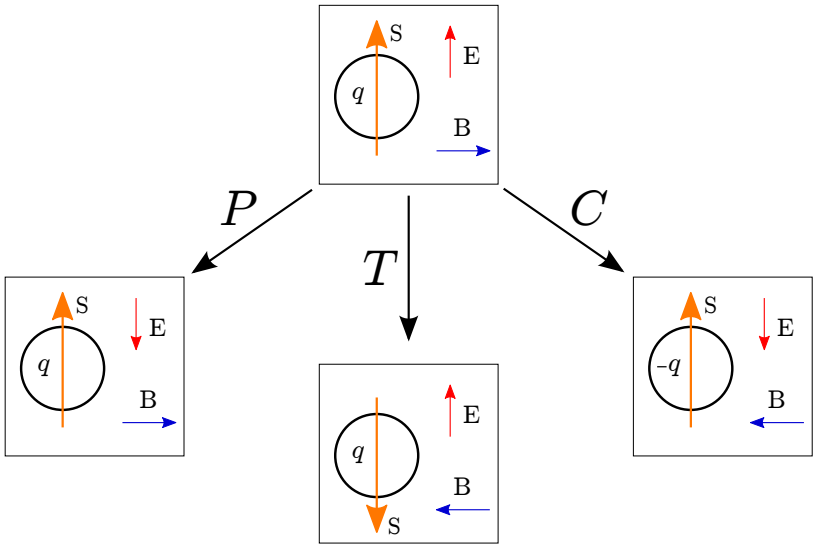

Figure 1.1: This diagram shows the effect of P, T and C transforms on a particle with charge q and spin S. Here E and B are the electric and magnetic fields produced by the charge q.

1.1 CP and T Violation

In 1964 after the discovery of P violation Christenson *et al.* [5] discovered that in the 2π decay of the K_L^0 meson the combined charge parity symmetry (CP) is violated. Before this experiment there where two known decay eigenstates of the neutral kaon K^0 and its anti-particle $\bar{K}^0$. These are CP eigenstates given by [6],

$$
\begin{aligned}
|K_S^0\rangle &= \frac{1}{\sqrt{2}}\left(|K^0\rangle + |\bar{K}^0\rangle\right) \\
|K_L^0\rangle &= \frac{1}{\sqrt{2}}\left(|K^0\rangle - |\bar{K}^0\rangle\right), \\
\text{Where}: \; CP\,|K_S^0\rangle &= +\,|K_S^0\rangle \\
CP\,|K_L^0\rangle &= -\,|K_L^0\rangle\,.
\end{aligned}
$$

Assuming conservation of CP, K_S^0 decays into two pions and K_L^0 decays into three pions (as both pion decays are C-even and pions have negative parity). Also K_S^0 is a short lived eigenstate and K_L^0 a long lived eigenstate[6]. However the experiment by Christenson *et al.* showed that approximately 1 in 500 K_L^0 particles underwent a 2π decay,

$$K_L^0 \to \pi^+ + \pi^-$$

where $\pi^{\pm}$ are charged pions. This decay from the CP-odd K_L^0 state into the CP-even decay state above shows that CP is not conserved (CP-violating) and in 1980 J. Cronin and V. Fitch were awarded the Nobel prize for this discovery. We will not discuss CP-violation in the kaon decay too deeply as this is in the realm of particle physics, we refer you to references [6, 7, 8, 9] for an overview of the kaon decays. Instead for the remainder of the thesis we will focus on the CP-violating electromagnetic moments.

Any Lorentz invariant physical theory that can be represented by a Lagrangian must be CPT invariant (see Section 2.1) and therefore CP-violating theories must also be T-violating. Since the discovery of CP-violating kaon decay, many more theories and experiments have been proposed to observe CP-,T- odd phenomena. Although many of these have been in the particle physics sector another possible avenue which has been considered for the past 50 years is the presence of permanent CP-violating moments in fundamental particles (electrons, quarks) and composite particles (nucleons, nuclei, atoms, molecules).

It was shown quite early that the presence of a finite permanent electric dipole moment (EDM) would constitute a CP-violating phenomenon [1, 10]. This is because the EDM is defined as,

$$\mathbf{d} = e \int \rho\left(\mathbf{r}\right) \mathbf{r} d^3 r \tag{1.1}$$

where ρ is the density of the charge.The EDM must be parallel to the angular momentum as it is the only vector which characterises the direction of the system. Therefore we can represent the dipole moment as [11],

$$\mathbf{d} = d_z \frac{\mathbf{I}}{I} \tag{1.2}$$

Under P and T transformations (1.1) and (1.2) contradict. To see this we have for (1.1)

$$\mathbf{d} \overset{P}{\mapsto} -\mathbf{d}, \qquad \mathbf{d} \overset{T}{\mapsto} \mathbf{d}.$$

Also noting that spin is a pseudo-vector which is T-odd the transform of (1.2) with P and T is

$$\mathbf{d} \overset{P}{\mapsto} \mathbf{d}, \qquad \mathbf{d} \overset{T}{\mapsto} -\mathbf{d}.$$

Therefore (1.1) and (1.2) contradict under P and T transformations and therefore a non-zero EDM is a P-,T-violating phenomenon. This is demonstrated in Figure 1.2.

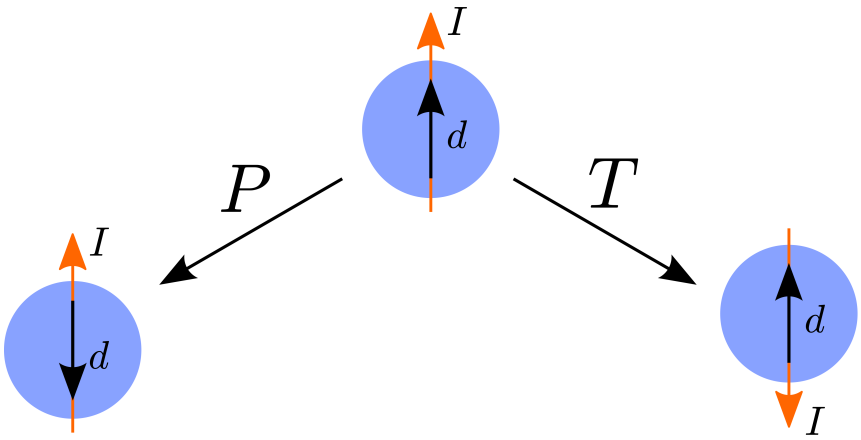

Figure 1.2: This diagram show the P and T transforms of a particle with angular momentum I and a finite EDM $\mathbf{d}$. The spatial EDM defined in (1.1) is P-odd and T-even. However as the EDM must be in the direction of the angular momentum of the particle which is P-even and T-odd there is a contradiction and the EDM violates P and T.

Other than the EDM there are many other P-,T-violating permanent electromagnetic moments, in the context of nuclear moments these are shown in Table 1.1 and derived in Appendix A. Due to the angular momentum requirements higher order moments only exist in composite systems such as nuclei. In this thesis we will focus on the nuclear MQM.

On the path to understanding CP violation electromagnetic moments present a promising approach for many reasons. Experiments focusing on CP-violating moments are low energy in comparison to the alternate route of high energy particle physics. Therefore the study of these moments is beneficial as for a relatively low cost they can be used as a sensitive probe to find and test new physics beyond the standard model.

In the past half century a great amount of work have been done on these moments, for some excellent reviews on the field we direct you to references [12, 13, 14, 15, 16] .

These tests have primarily focused on measuring the electric dipole moments (EDMs)

Nuclear Moment	P-,T-	Tensor Order	Angular Momentum
EDM, Schiff	Violating	1	$I > 0$
Electric Quadrupole (EQM)	Conserved	2	$I > 1/2$
Electric Octupole (EOM)	Violating	3	$I > 1$
Magnetic Dipole (MDM)	Conserved	1	$I > 0$
Magnetic Quadrupole (MQM)	Violating	2	$I > 1/2$
Magnetic Octopole (MOM)	Conserved	3	$I > 1$

Table 1.1: This table shows which nuclear moments violate and conserve P and T symmetries as well as their tensor order and angular momentum restriction, where I is the nuclear spin.

of fundamental particles such as the electron ($\mathbf{d}_e$) and also composite particles such as the neutron ($\mathbf{d}_n$) and proton ($\mathbf{d}_p$). These endeavors have not been futile because though there have only been null measurements for the permanent EDM the upper bounds of various particle EDMs have been strengthened by many orders of magnitude since the first experiments for the electron [17, 18], neutron [19] (these were before CP violation had be discovered in the decay of kaons) and proton [20] EDMs (see Table 1.2 in Section 1.2).

1.2 Physics Beyond the Standard Model

With the recent confirmation of the Higgs boson, the standard model has stood up to every test thus far, however the majority of physicists still believe the standard model to be incomplete as there are many phenomena it can not explain[16, 21, 12, 22]. CP-violation exists within the standard model in the context of the Cabibbo–Kobayashi–Maskawa matrix [23, 24, 14]. Kobayashi and Maskawa showed that for three quark generations the quark mixing matrix has a CP-violating quark phase. For fewer than three quarks generations this phase can be rotated out, however for three or more generations the CP-violating phase is non-vanishing in the standard model [24, 11]. In this phenomenological approach it can be shown that this CP-violating phase will result in a finite EDM for the quarks and leptons however this comes out of a high order correction and therefore the produced EDM is vanishingly small [25]. For example the electron EDM is thought to be of the order $\approx 10^{-38}$ [25, 16, 14] which it is well outside the current measurement limits and any foreseeable experiments in the future.

There is a large inconsistency between the standard model and what has been observed in nature. In the universe there is a large asymmetry between matter and

anti-matter, most of the matter is comprised of baryons (neutrons and protons) and therefore this asymmetry is intimately tied to the baryogenesis of the universe. This asymmetry is typically presented in the ratio of baryons to photons, η. Experimentally it has been shown that in our universe $\eta \approx 10^{-10}$ [26, 27]. In 1967 Sakharov [28] showed that at least three conditions are required to account for this asymmetry, one of them being CP violation. The issue with the standard model prediction is the CP-violating mechanism produces an asymmetry of the order $\eta \approx 10^{-20}$[26, 28]. This suggests that the current Standard Model is incomplete and it must be extended to incorporate a greater degree of CP violation.

There have been many candidate theories for this unknown source of CP violation and among the most popular are the supersymmetric theories (SUSY), multi-Higgs theories, left-right symmetric and superweak theories. The CP-violating mechanisms in these theories all predict EDMs of fundamental particles orders of magnitude greater than the standard model counterpart, these are presented in Table 1.2 [29]. These predictions give us quite significant insight for discovering new physics as we continuously decrease the upper bound of the fundamental EDMs with more accurate experiments we will slowly discount more theories.

Other than the matter anti-matter asymmetry there are other very important unanswered questions in fundamental physics are connected to CP violation and specifically CP violating electromagnetic moments. These are the strong CP problem and dark matter and dark energy. In the strong CP problem there is the puzzling observation that QCD does not appear to violate the CP symmetry[30, 31, 32, 33, 34, 35](see Section 2.4). In 1977 R.D. Peccei and H.R. Quinn proposed that a possible solution to the strong CP problem would be the introduction of a massive bosonic particle known as the axion [30, 31]. It has been noted that the axion may also be a promising cold dark matter candidate. Thus axions, if detected, could resolve both the dark matter and strong CP problems [36, 37, 38].

Currently there have only been null measurements for EDMs of fundamental particles however the current experimental limit of the electron EDM (8.7×10^{-29} $e \cdot \text{cm}$ [39]) already tightly constrains supersymmetric theory predictions as the parameter space is very limited already [16]. Therefore research into improving the limits on CP-violating moments is on the forefront of modern physics. The electron and neutron EDM predictions of different models are presented in Table 1.2 along with the current

experimental limit.

Model	$\lvert d_e \rvert$ $(e \cdot \text{cm})$	$\lvert d_n \rvert$ $(e \cdot \text{cm})$
Standard model	$< 10^{-38}$	$\approx 10^{-32}$ [40]
SUSY	$10^{-26} - 10^{-28}$	$10^{-25} - 10^{-26}$ [41, 42]
Multi-Higgs	$10^{-26} - 10^{-28}$	N/A
Left-right symmetric	$10^{-26} - 10^{-28}$	N/A
Current Experimental Limit	$< 8.7 \times 10^{-29}$ [39]	$< 2.9 \times 10^{-26}$ [43]

Table 1.2: This table shows an order of magnitude prediction of for the electron and neutron EDM for various models which contain a CP violating mechanism. All values come from reference [29] unless cited otherwise.

Chapter 2

Nuclear Moments and CP Violation

In the introduction we mentioned the direct measurement of free particles, however there is a more robust avenue to find permanent CP-violating moments in nature through the study of static nuclear moments in neutral systems.

Measurements of free particles can only get us so far when looking for permanent moments, especially in the context of charged particles like protons and electrons. This is because when we apply an electric field the charged particle will be accelerated out of the experimental apparatus. Therefore we turn to the measurement of neutral systems that consist of these charged particle EDMs to observe CP violation.

We will also see that this allows us to find more fundamental CP violating properties which would have been unobtainable with free particles.

Here I will give a brief overview of the CPT theorem and its consequences, the P-,T-odd electric and magnetic moments which manifest in nuclei, the inter-nucleon P-,T-violating potential of the nucleus, how these moments can be studied with atomic and molecular systems and finally how we can link the CP-violating moments to the strong CP problem in quantum chromodynamics (QCD).

2.1 The CPT Theorem

The CPT theorem is assumed to be fundamental property of nature, it was first suggested by Lüders who provided a proof of the theorem [44] which was later generalised by Pauli [45]and Bell [46]. Essentially the CPT theorem states that under the reflection

of 4-dimensional space all Lorentz invariant systems are unchanged under a combined transformation of C, P and T. This immediately means that all physically realistic systems that can be written as a Lorentz invariant Lagrangian are CPT invariant. The basis of this section is based upon the the work in references [14, 47, 48]. Here will not provide a rigorous proof but an intuitive explanation.

First we will show that the combined CPT transformation results in the reflection of 4-dimensional space-time, as opposed to the naive assumption that just a combined PT transform would be sufficient. To do this we will consider how both true and pseudo-vectors transform under CPT. For the true vector case we will use the example of the electromagnetic 4-vector current $j_\mu = (\rho, \mathbf{j})$, which for a complete reflection we expect $j_\mu \to -j_\mu$. Under the CPT transforms the current is given by

$$\begin{aligned}(\rho, \mathbf{j}) \overset{T}{\mapsto} (\rho, -\mathbf{j}) \overset{P}{\mapsto} (\rho, \mathbf{j}) \overset{C}{\mapsto} (-\rho, -\mathbf{j}) \\ \Rightarrow j_\mu \overset{CPT}{\longmapsto} -j_\mu.\end{aligned}$$

Therefore we see there is only a complete reflection under C, P and T transforms where any subset would be insufficient.

The case for the psuedo-vector is similar however we have to consider an intermediate step due to the Lorentz transform. We will use the spin pseudo-vector s_μ of a particle with mass m and momentum $\mathbf{k}$. In the rest frame of the particle the spin 4-vector is given by $s_\mu = (0, \mathbf{s})$ however for the particle with momentum $\mathbf{k}$ with respect to the frame the Lorentz transform of the spin vector is given by [14],

$$s_\mu = \left(\frac{\mathbf{s} \cdot \mathbf{k}}{m}, \mathbf{s} + \frac{(\mathbf{s} \cdot \mathbf{k})\, \mathbf{k}}{m\,(E + m)} \right)$$

where E is the energy. Therefore we now have a form we can accurately test for reflection. Immediately we now see that under CPT transformation we have a complete

reflection of the pseudo-vector,

$$\left(\frac{\mathbf{s}\cdot\mathbf{k}}{m},\mathbf{s}+\frac{(\mathbf{s}\cdot\mathbf{k})\,\mathbf{k}}{m\,(E+m)}\right)\overset{T}{\mapsto}\left(\frac{\mathbf{s}\cdot\mathbf{k}}{m},-\left(\mathbf{s}+\frac{(\mathbf{s}\cdot\mathbf{k})\,\mathbf{k}}{m\,(E+m)}\right)\right)$$
$$\left(\frac{\mathbf{s}\cdot\mathbf{k}}{m},\mathbf{s}+\frac{(\mathbf{s}\cdot\mathbf{k})\,\mathbf{k}}{m\,(E+m)}\right)\overset{P}{\mapsto}\left(-\frac{\mathbf{s}\cdot\mathbf{k}}{m},\mathbf{s}+\frac{(\mathbf{s}\cdot\mathbf{k})\,\mathbf{k}}{m\,(E+m)}\right)$$
$$\left(\frac{\mathbf{s}\cdot\mathbf{k}}{m},\mathbf{s}+\frac{(\mathbf{s}\cdot\mathbf{k})\,\mathbf{k}}{m\,(E+m)}\right)\overset{C}{\mapsto}\left(\frac{\mathbf{s}\cdot\mathbf{k}}{m},\mathbf{s}+\frac{(\mathbf{s}\cdot\mathbf{k})\,\mathbf{k}}{m\,(E+m)}\right)$$
$$\Rightarrow s_\mu \overset{CPT}{\longmapsto} -s_\mu$$

Therefore we have that only under the operation of combined CPT there is a complete reflection of space-time and therefore both true vectors and pseudo-vectors are CPT-odd.

A principle of Hamiltonians and Lagrangians are that all operators (CPT-odd) are paired and therefore the Hamiltonian and Lagrangian are strictly CPT-even [49]. For example consider the coupling between the vector potential $\mathbf{A}$ and the electromagnetic current 3-vector $\mathbf{j}$, $H_M = \mathbf{A}\cdot\mathbf{j}$. Both of these are true vectors and therefore both are CPT-odd and the Hamiltonian is CPT-even as required.

2.2 Nuclear Moments and Schiff's Theorem

It would seem that to detect the EDM of a system would be analogous to that of the magnetic dipole moment of the system (MDM). The Hamiltonian for the interaction of an electric field ($\mathbf{E}$) and particle EDM ($\mathbf{d}$) is,

$$H_{EDM} = -\mathbf{d}\cdot\mathbf{E}.$$

Therefore measuring the energy shift of the system will mean that the particle has a finite EDM. While this is true when measuring free particles like the electron and nucleon EDMs, in neutral systems such as atoms and molecules an external electric field would result in no atomic EDM due to a theorem by Schiff.

In 1963 a paper by Schiff showed that for a neutral system the permanent EDM would be impossible to observe in the presence of an external electric field to first order for a point particle when only taking in to account electrostatic effects in the neutral system. The Schiff theorem for a neutral atom can be understood in a simple heuristic way.

Consider an neutral atom with a partially filled valence band (paramagnetic) if we were to apply and external electric field the electron would interact with the field and be perturbed and accelerated. However as the atom is neutral it should not accelerate and therefore the perturbing field will be equally matched with an internal restoring field and there will be no net effect. The same argument is valid for any constituent EDM of a neutral system including the EDM of the nucleus.

To see this directly consider the Dirac Hamiltonian of an electron in an external field,

$$H = c\boldsymbol{\alpha}\cdot\mathbf{p} + mc^2\gamma^0 - e\Phi \tag{2.1}$$

$$H_{EDM} = -d_e\gamma^0\boldsymbol{\Sigma}\cdot\mathbf{E} \tag{2.2}$$

$$= -d_e\boldsymbol{\Sigma}\cdot\mathbf{E} - d_e\left(\gamma^0 - 1\right)\boldsymbol{\Sigma}\cdot\mathbf{E} \tag{2.3}$$

where c, $\mathbf{p}$, $\mathbf{E}$ and m are the speed of light, momentum of the electron, electric field at the electron and mass of the electron respectively, Φ is the total electric field acting on the electron (external and internal) and $\boldsymbol{\alpha}$, $\boldsymbol{\Sigma}$ γ^0 and the spin and Dirac matrices given by [47],

$$\boldsymbol{\alpha} = \begin{pmatrix} 0 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & 0 \end{pmatrix},\ \boldsymbol{\Sigma} = \begin{pmatrix} \boldsymbol{\sigma} & 0 \\ 0 & \boldsymbol{\sigma} \end{pmatrix},\ \gamma^0 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

In (2.3) we have separated the Hamiltonian of the EDMs interaction with the external field into non relativistic and relativistic components

$$H_{EDM} = H^{NR}_{EDM} + H^{Rel}_{EDM}$$

$$\text{Where: } H^{NR}_{EDM} = -d_e\boldsymbol{\Sigma}\cdot\mathbf{E} \tag{2.4}$$

$$H^{Rel}_{EDM} = -d_e\left(\gamma^0 - 1\right)\boldsymbol{\Sigma}\cdot\mathbf{E}. \tag{2.5}$$

The $(\gamma^0 - 1)$ matrix only has lower components which vanishes in the non-relativistic case. Therefore for the non-relativistic case the energy shift to first order is given by,

$$\Delta E^{(1)} = \left\langle\psi\right|H^{NR}_{EDM}\left|\psi\right\rangle$$

$$= -d_e\left\langle\psi\right|\boldsymbol{\Sigma}\cdot\mathbf{E}\left|\psi\right\rangle. \tag{2.6}$$

Substituting in the potential, $\mathbf{E} = -\nabla\Phi$, and rearranging we can write this in the form [25],

$$\Delta E^{(1)} = i\frac{d_e}{e}\left\langle \psi \left| \left[\boldsymbol{\Sigma}\cdot\mathbf{p}, e\Phi \right] \right| \psi \right\rangle . \tag{2.7}$$

Therefore rearranging (2.1) for $e\Phi$ and substituting it into (2.7) we are left with,

$$\Delta E^{(1)} = i\frac{d_e}{e}\left\langle \psi \right| \left[\boldsymbol{\Sigma}\cdot\mathbf{p}, H - c\boldsymbol{\alpha}\cdot\mathbf{p} - mc^2\gamma^0 \right] \left| \psi \right\rangle .$$

Using the properties of the Dirac [47] and Pauli matrices[50] it is simple to show that $\boldsymbol{\Sigma}\cdot\mathbf{p}$ commutes with $\boldsymbol{\alpha}\cdot\mathbf{p}$ and $mc^2\gamma^0$. Therefore we are left with,

$$\Delta E^{(1)} = i\frac{d_e}{e}\left\langle \psi \left| \left[\boldsymbol{\Sigma}\cdot\mathbf{p}, H \right] \right| \psi \right\rangle = 0$$

We immediately see that this is zero as $|\psi\rangle$ is an eigenket of H and therefore when the commutator is expanded the terms cancel. Therefore we have shown that Schiff's theorem holds.

For the relativistic case however this energy shift is non zero. Substituting (2.5) into (2.6),

$$\begin{aligned} \Delta E^{(1)} &= \left\langle \Psi \right| H_{EDM}^{Rel} \left| \Psi \right\rangle \\ &\neq 0. \end{aligned}$$

Therefore when considering relativistic effects there is a finite shift in energy due to the electron dipole moment of the electron, violating Schiff's theorem. For the nuclear EDM this correction is insignificant due to the large mass of the nucleus with respect to the electrons.

The other major mechanism which breaks Schiff's theorem for electrostatic moments is that the particle with the EDM has finite size, which is particularly important for the nuclear EDM. As stated above Schiff's theorem assumes a point like particle. Therefore the EDM of the nucleus is non zero although still severely screened by the electrons, in the literature this screened EDM is eponymously named the Schiff moment [11].

2.2.1 Electric Moments in Nuclei

Other than the Schiff moment there are are other CP-violating electric moments of the nucleus. CP-violating moments skip every second generation and therefore the next order moment is the the electric octupole moment (EOM). The Schiff moment and EOM are both derived in Appendix A.2 and both come from the third order expansion of the scalar potential given by,

$$\phi^{(3)}_{octupole}(R) \approx -eq\frac{1}{6}O_{ijk}\partial_i\partial_j\partial_k\frac{1}{R} \tag{2.8}$$

$$\phi^{(3)}_{Schiff}(R) = -S_k\partial_k\partial^2\frac{1}{R} = 4\pi S_k\partial_k\delta(R)$$

$$\text{Where}\quad O_{ijk} = \int\rho_\nu\left(r_ir_jr_k - \frac{1}{5}\left(\delta_{jk}r_i + \delta_{ik}r_j + \delta_{ij}r_k\right)r^2\right)d^3r \tag{2.9}$$

$$S_k = \frac{eq}{10}\left[\int\rho_\nu r_k r^2 d^3r + \frac{5}{3Z}\langle r_k\rangle\int\rho_0 r^2 d^3r\right] \tag{2.10}$$

Here we note that the Schiff Moment retains the vector structure of the EDM. For both the Schiff moment and the EOM an important thing to note it dependence on the charge q. We see that for a valence neutron the valence contribution of the Schiff moment and EOM vanishes and the Schiff moment will come from the core contribution only [51].

It is interesting to note that there is a large difference in magnitude between the higher orders of CP-violating electric moments. Although we have restored an effective nuclear EDM with the Schiff moment we can see it is largely suppressed as $S \propto r_N^2\langle d_N\rangle$ where $\langle d_N\rangle$ is the unshielded nuclear EDM, and $r \approx 1.1$ fm is the nuclear radius. Comparing this to (2.9) for the EOM which scales with r^3 and therefore is greatly suppressed.

2.2.2 Magnetic Moments in Nuclei

Other than the electric nuclear moments it is important to consider the magnetic moments. The lowest order CP-violating magnetic moment is the magnetic quadrupole moment (MQM). The MQM presents a promising way to observe CP-violating effects as it is not screened by the core electrons and therefore is not affected by Schiff's theorem. However with this advantage there are disadvantages associated with the MQM.

The largest restriction of studying the MQM is that we require paramagnetic atoms or

molecules to measure it (see Section 2.5) where the Schiff moment can been observed with both paramagnetic and diamagnetic systems. The second restriction is the nuclear angular momentum restriction for the MQM is, $I > 1/2$ (for the Schiff moment there is a weaker restriction on nuclear spin, $I > 0$). However this problem can be overcome when considering that the MQM displays collective properties in deformed nuclei as the closed shells split into partially filled shells and therefore there are many valence nucleons[52] (see Chapter 3).

2.2.3 Origin of Nuclear Moments

There are many mechanisms which may induce odd electromagnetic moments in the nucleus. Three distinct ones are, [53]

1. Expectation values of nuclear states considering the independent electromagnetic moments of the nucleons.

2. Parity mixing of nuclear states due to the interaction of of the nucleon electric dipole moment with the core of the nucleus.

3. Parity mixing of nuclear states due to the internal T, P-odd internucleon interaction governed by H_{TP} .

We do not consider the second case in this thesis as for the SM and MQM the major contribution comes from the T, P-odd interaction. The first case is important and will be considered in the nuclear MQM which will be presented in section 3.1.

In general the electromagnetic moments of the nucleus are induced through the coupling of a multipole field and the T, P-odd N-N interaction, H_{TP}. We will discuss the form of H_{TP} in greater detail in Section 2.3. For example we will consider the MQM of the nucleus. The total contribution to the MQM by H_{TP} is given by,

$$\begin{aligned} M_{Total} &= \sum_n \frac{\left\langle \psi_0 \left| \hat{M} \right| n \right\rangle \langle n \,|\, H_{TP} \,|\, \psi_0 \rangle}{E_0 - E_n} + \frac{\langle \psi_0 \,|\, H_{TP} \,|\, n \rangle \left\langle n \left| \hat{M} \right| \psi_0 \right\rangle}{E_0 - E_n} \\ &= 2 \sum_n \frac{\left\langle \psi_0 \left| \hat{M} \right| n \right\rangle \langle n \,|\, H_{TP} \,|\, \psi_0 \rangle}{E_0 - E_n} \end{aligned} \tag{2.11}$$

where ψ_0 is the unperturbed wavefunction of the nucleon, $\hat{M}$ is the field operator for the MQM field and $|n\rangle$ is some opposite parity state for the nucleon. We see that (2.11) vanishes for a nonexistent H_{TP} as expected.

Although (2.11) is the complete contribution it can be separated into valence and core contributions. Most current work has only considered the valence contribution which averages over all core nucleons and has the larger contribution of the two. For the effective valence Hamiltonian (see Section 2.3) the MQM contribution can be written as,

$$M^{Val} = 2\sum_n \frac{\left\langle \psi_0 \left| \hat{M} \right| n \right\rangle \left\langle n \left| H_{TP}^{Val} \right| \psi_0 \right\rangle}{E_0 - E_n}$$

which corresponds to the MQM field operating on the valence nucleon only and can be represented by the Feynman diagrams in Figure 2.1.

The core contribution on the other hand comes from the field operating on the

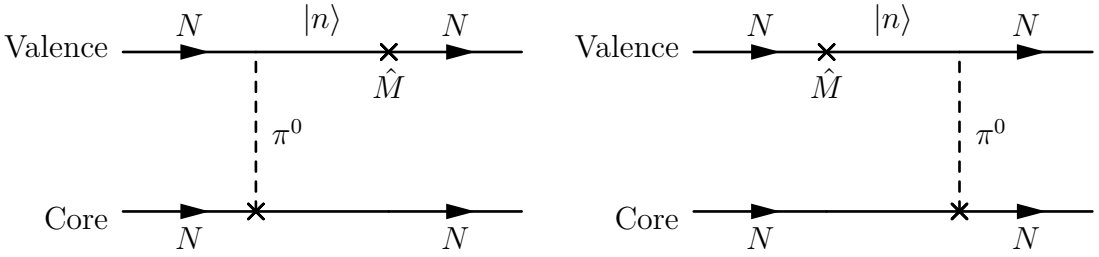

Figure 2.1: The Feynman diagrams showing the magnetic quadrupole moment of the nucleus due to the valence nucleons arising TP-odd pion exchange and the TP-odd MQM field operator. The lower branches are the core nucleons and the upper branches are the valence nucleons where the $\times$ denotes a T-,P-violating vertex. There are two more graphs which have to be summed corresponding to the T-,P-violating vertex of the pion exchange on the valence branch.

core nucleons only. The matrix element representing the core contribution is presented in [54] as,

$$M^{Core} = \sum_{\lambda\lambda'} \langle \nu\lambda \,|\, H_{TP} \,|\, \lambda'\nu \rangle \frac{n_\lambda - n_{\lambda'}}{E_\lambda - E_{\lambda'}} \left\langle \lambda' \left| \hat{M} \right| \lambda \right\rangle$$

where λ are the core nucleon states, ν are the valence nucleon states and n_λ and ϵ_λ are the nuclear occupation numbers and energies respectively. The Feynman diagram of the core interaction is represented in Figure 2.2.

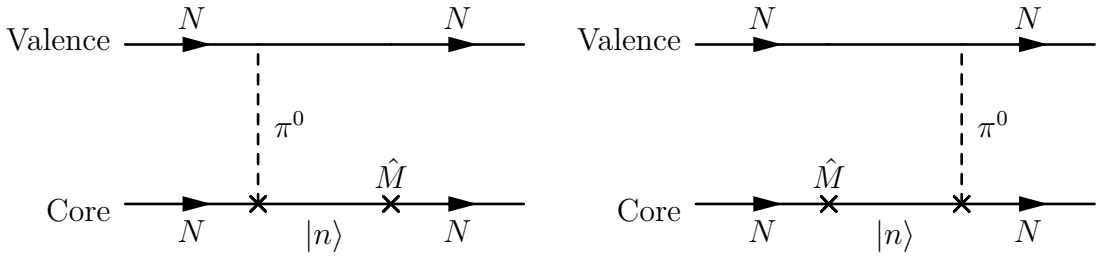

Figure 2.2: The Feynman diagrams showing the magnetic quadrupole moment of the nucleus due to the core nucleons arising TP-odd pion exchange and the TP-odd MQM field operator. The lower branches are the core nucleons and the upper branches are the valence nucleons where the $\times$ denotes a TP-violating vertex. There are two more graphs which have to be summed corresponding to the T-,P-violating vertex of the pion exchange on the valence branch.

2.3 Time and Parity Violating Inter-Nucleon Potential

From the Lagrangian density representation of the T, P-odd nucleon-nucleon interaction in [55, 13] and performing a Fierz transform we can transform it into a coordinate representation of the TP-odd nucleon-nucleon interaction[56, 57, 58, 59]

$$H_{TP}\left(\mathbf{r}_1-\mathbf{r}_2\right)=-\frac{g}{8\pi m_p}\left[\left(\bar{g}_0\boldsymbol{\tau}_1\cdot\boldsymbol{\tau}_2+\bar{g}_2\left(\boldsymbol{\tau}_1\cdot\boldsymbol{\tau}_2-3\tau_1^3\tau_2^3\right)\right)\left(\boldsymbol{\sigma}_1-\boldsymbol{\sigma}_2\right)\right.$$
$$\left.+\bar{g}_1\left(\tau_1^3\boldsymbol{\sigma}_1-\tau_2^3\boldsymbol{\sigma}_2\right)\right]\nabla_1\frac{e^{-m_\pi r_{12}}}{r_{12}} \qquad (2.12)$$

where 1 and 2 correspond to the two nucleons in the pion exchange, $r_{12}=|\mathbf{r}_1-\mathbf{r}_2|$ is the distance between the nucleons and $g\bar{g}_i$ are terms for the ith isopin channel (isoscalar, isovector and isotensor for 0, 1, and 2 respectively) and the bar denotes that the channel is CP-violating.

This Hamiltonian represents the finite range of the pion interaction as seen by the Yukawa term. However we can reduce this into a contact interaction which is of the

form (see Appendix C for details) [56],

$$H_{TP} = \frac{G}{\sqrt{2}}\frac{1}{2m}\left[\left(\eta_{ab}\boldsymbol{\sigma}_1 - \eta_{ba}\boldsymbol{\sigma}_2\right)\nabla\delta\left(\mathbf{r}_a - \mathbf{r}_b\right) + \eta'_{ab}\left[\boldsymbol{\sigma}_a \times \boldsymbol{\sigma}_b\right]\left\{\left(\mathbf{p}_a - \mathbf{p}_b\right), \delta\left(\mathbf{r}_a - \mathbf{r}_b\right)\right\}\right] \tag{2.13}$$

where $\{,\}$ is the anti-commutator. Here η_{ab} and η'_{ab} are the T,P-odd parameters of the direct and exchange channels of the nucleon-nucleon interaction where a and b are protons (p) or neutrons (n). From Appendix C and [57] the T,P-odd parameters for all the channels are given by,

$$\begin{aligned}
\eta_{pp} &= 5\times 10^6 g\left(-\bar{g}_0 + 2\bar{g}_2 - \bar{g}_1\right)\\
\eta_{nn} &= 5\times 10^6 g\left(-\bar{g}_0 + 2\bar{g}_2 + \bar{g}_1\right)\\
\eta_{pn} &= 5\times 10^6 g\left(\bar{g}_0 - 2\bar{g}_2 - \bar{g}_1\right)\\
\eta_{np} &= 5\times 10^6 g\left(\bar{g}_0 - 2\bar{g}_2 + \bar{g}_1\right).
\end{aligned} \tag{2.14}$$

When considering the T,P-odd Hamiltonian (2.13) we typically consider the interaction of the valence nucleon (a) with the core as the major contribution. For this case we can average (2.13) over the core nucleons (noting that $\langle\boldsymbol{\sigma}_{Core}\rangle = 0$) and we can write an effective Hamiltonian for the valence nucleon as,

$$H_{TP}^{Val} = \frac{G}{\sqrt{2}}\frac{1}{2m_p}\eta_a\boldsymbol{\sigma}\nabla\rho\left(r\right), \tag{2.15}$$

$$\text{Where: } \eta_a = \begin{cases} \dfrac{Z}{A}\eta_{pp} + \dfrac{N}{A}\eta_{pn} & \text{for } a = p \\ \dfrac{Z}{A}\eta_{np} + \dfrac{N}{A}\eta_{nn} & \text{for } a = n. \end{cases} \tag{2.16}$$

It is easy to see this effective valence Hamiltonian is P,T-odd as

$$\begin{aligned}
&\boldsymbol{\sigma} \overset{T}{\mapsto} -\boldsymbol{\sigma}, && \boldsymbol{\sigma} \overset{P}{\mapsto} \boldsymbol{\sigma}\\
&\nabla\rho\left(r\right) \overset{T}{\mapsto} \nabla\rho\left(r\right), && \nabla\rho\left(r\right) \overset{P}{\mapsto} -\nabla\rho\left(r\right)
\end{aligned}$$

as the gradient of the density is the radius. Here $\rho\left(r\right)$ is the nuclear density, $\boldsymbol{\sigma}$ is the Pauli spin matrix of the valence nucleon. As the strong interaction is short range we make the approximation that the density profile of the nucleus is the same as the nuclear potential, $U\left(r\right)$ (for the valence case the form is irrelevant however this is not

the case when we consider the core contribution) and therefore we can write [11],

$$\rho(r) = \frac{\rho(0)}{U(0)} U(r).$$

Therefore substituting this into (2.15) we can rewrite the valence nucleon T, P-odd potential as [11],

$$H_{TP}^{Val} = \xi_a \boldsymbol{\sigma} \nabla U(r). \tag{2.17}$$

$$\begin{aligned} \text{Where: } \xi_a &= \frac{G}{\sqrt{2}} \frac{1}{2m_p} \frac{\rho(0)}{U(0)} \eta_a \\ &= -2 \times 10^{-21} \eta_a \quad \text{cm}. \end{aligned} \tag{2.18}$$

We can treat this T-odd Hamiltonian as a perturbation and therefore the perturbed potential of the system can be written as,

$$\begin{aligned} \tilde{U}(r) &= U(r) + H_{TP}, \\ &= U(r) + \xi \boldsymbol{\sigma} \nabla U(r), \\ &\approx U(r + \xi \boldsymbol{\sigma}), \end{aligned}$$

to the first order in a Taylor expansion. Therefore we have that the effective wavefunction of a particle in this potential is given by,

$$\begin{aligned} \tilde{\psi}(r) &= \psi(r + \xi \boldsymbol{\sigma}) \\ &= \psi(r) + \xi \boldsymbol{\sigma} \nabla \psi(r) \end{aligned} \tag{2.19}$$

We will use this effective wavefunction construction for both the MQM and EOM.

2.4 Symmetry violation in QCD

After the discovery of parity violation in β decay it was easily reconciled with the weak interaction. With CP-violation things are not so simple particularly in the hadron sector involving the strong force. In the quantum chromodynamics (QCD) Lagrangian

there exists a term which represents the self interaction of the gluon field,

$$\mathcal{L}_{QCD} \ni \tilde{\theta} G^{\alpha}_{\mu\nu} \tilde{G}^{\alpha}_{\mu\nu} \tag{2.20}$$

where α is the colour index of the gluon field and $\tilde{G}$ is the dual gluon field. In comparison with electrodynamics, (2.20) is similar to the self coupling of the electromagnetic field tensors ($F_{\mu\nu}$ and $\tilde{F}_{\mu\nu}$) in classical electrodynamics [60]. Therefore it serves a purpose to first look at the symmetry violation of the electromagnetic field and then relate it back to the same term in the gluon field for clarity. The CP-violating term in the electromagnetic Lagrangian is given by,

$$\mathcal{L}_{EM} \ni F_{\mu\nu} \tilde{F}_{\mu\nu} = -\mathbf{E} \cdot \mathbf{B} \tag{2.21}$$

Now we see how the symmetries hold up for the electric field $\mathbf{E}$ and magnetic field $\mathbf{B}$ which are produced by some charge q.

$$\begin{aligned}
P &: \mathbf{E} \overset{P}{\mapsto} -\mathbf{E}, & \mathbf{B} &\overset{P}{\mapsto} \mathbf{B}, \\
T &: \mathbf{E} \overset{T}{\mapsto} \mathbf{E}, & \mathbf{B} &\overset{T}{\mapsto} -\mathbf{B}, \\
C &: \mathbf{E} \overset{C}{\mapsto} -\mathbf{E}, & \mathbf{B} &\overset{C}{\mapsto} -\mathbf{B}, \\
\Rightarrow CP &: \mathbf{E} \overset{CP}{\longmapsto} \mathbf{E}, & \mathbf{B} &\overset{CP}{\longmapsto} -\mathbf{B}.
\end{aligned}$$

Therefore from (2.21) we have that,

$$\begin{aligned}
CP &: F_{\mu\nu}\tilde{F}_{\mu\nu} \overset{CP}{\longmapsto} -F_{\mu\nu}\tilde{F}_{\mu\nu}, \\
CPT &: F_{\mu\nu}\tilde{F}_{\mu\nu} \overset{CPT}{\longmapsto} F_{\mu\nu}\tilde{F}_{\mu\nu}.
\end{aligned}$$

The gluon self interaction (2.20) obeys the same relations and therefore is also CP violating. Therefore we see that $\tilde{\theta}$ in (2.20) scales the CP violating term in the Lagrangian. Naively we would expect this factor to be of order unity as as it is dimensionless however it is incredibly small (from EDM experiments, $\tilde{\theta} < 10^{-10}$) which means that CP is not easily violated in QCD. This is known as the strong CP problem and there is currently no known reason for the small value.

Other than the strong CP theta term there are other fundamental CP-,T- violating properties that can be studied using these nuclear systems. The others we will briefly mention in this thesis are the EDMs of the proton, d_p, and neutron, d_n, and

the EDMs of the quarks. There are two dipole moments of these quarks, the first is the standard EDM interaction with the electromagnetic field, that is d_u and d_d for the up and down quarks respectively. The other is the quark dipole moments due to the interaction with the gluon field, these dipole moments are known as chromo-dipole moments (chromo-DMs) and are given by $\tilde{d}_u$ and $\tilde{d}_d$ for the up and down quarks respectively.

As the major contribution of nuclear moments come from the T, P-odd inter-nucleon potential these moments present a great avenue for finding stronger upper-limits on $\tilde{\theta}$ and therefore a greater understanding of CP-violation in QCD. This is because the pion exchange of the nucleon-nucleon interaction has a dependence on $\tilde{\theta}$ is approximately 40 times that of the nucleon EDM dependence [11]. Therefore nuclear moments present a much more fertile approach for QCD CP-violation than single free particle EDM. In this thesis we will present our results in terms of $\tilde{\theta}$ dependence.

2.5 Time Violating Effects in Atoms and Molecules

Now that we have established the presence and origin of CP-,T- violating moments of nuclei we now ask how do we observe these violating properties. Similar to the permanent nuclear moment in atomic and molecular systems a permanent EDM can arise from three different mechanisms [61],

1. The combined EDMs of constituent electrons will result in a net molecular or atomic EDM.

2. The T-,P-odd interaction of the orbital electrons and the nucleons (e-N interaction) will mix opposite electron parity states giving the atom or molecule an EDM.

3. The interaction of the electromagnetic field of the electrons with the T-,P- odd nuclear moments will mix atomic states of opposite parity and result in a molecular and atomic EDM.

For case 1 the total EDM of the system, $\mathbf{D}$, is simply given by,

$$\mathbf{D} = \sum_i \mathbf{d}_i$$

Where $\mathbf{d}_i$ are the individual electron EDMs. However as most of the shells in atomic and molecular systems are closed most of the electron EDMs cancel and therefore this contribution is small.

The EDM of the system due to cases 2 and 3 which are caused by some T-,P-odd operator $\hat{h_{tp}}$ (for example M_{zz} of the MQM of the nucleus) have the same form,

$$\mathbf{D} = 2\sum_n \frac{\left\langle \psi \left| \hat{\mathbf{d}} \right| n \right\rangle \left\langle n \left| \hat{h_{tp}} \right| \psi \right\rangle}{E_0 - E_n}$$

Where $|\psi\rangle$ is the unperturbed electronic wavefunction, $|n\rangle$ are the excited states of opposite parity to $|\psi\rangle$ and $\hat{\mathbf{d}}$ is just a dipole operator along the axis of the system (in molecules this is the molecular axis $\mathbf{n}$, see Appendix D).

In this thesis we do not consider the contribution of the e-N weak interaction and only consider the interaction of the electromagnetic field with the T-,P-odd nuclear moment. We direct you to several thorough reviews on the e-N interaction in [13, 12, 62].

It might seem strange to search for the dipole moment of molecules when we can observe the dipole properties of polar molecules such as water with a weak electric field. These are not true EDMs (induced not permanent) which do not violate CP. Due to the rotation of the polar molecule, in the absence of an external field there will be no average net dipole. When a weak electric field is applied the resultant energy shift is not a linear stark shift (as with a permanent EDM) but a quadratic stark shift. Therefore the dipole due to the shifted electron density in polar molecules is not a true EDM. A true EDM must have mixed parity states in a weak field approximation [14] (see Figure 2.3). When considering the molecular EDM induced by nuclear moments, the case of the electric an magnetic moments are quite different as the moments interact with the field in different ways. For the electric moments the case is quite simple. The moment will produce some potential, ϕ, of the nucleus which will interact with the electron field simply as,

$$V_{Elec} = -e\phi.$$

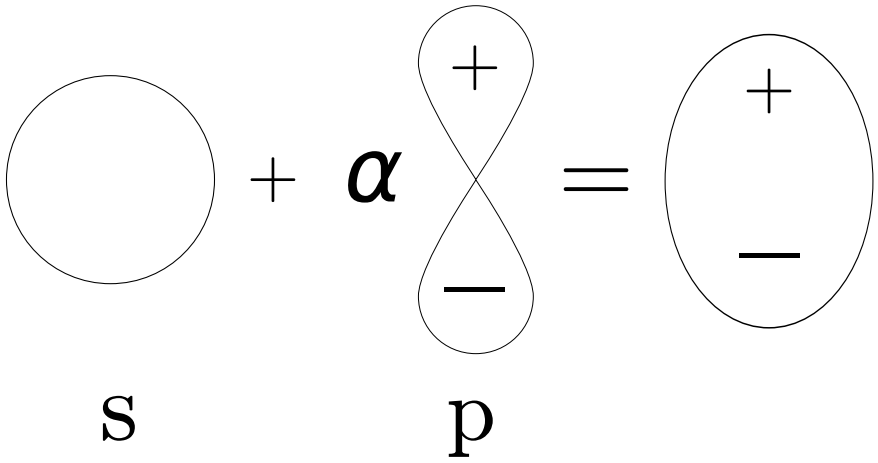

Figure 2.3: This figure depicts the parity mixing of the s-wave state and the opposite parity p-wave state in an atomic system. The sum of these states gives a total spatial charge distribution of the state and therefore a net EDM.

The case for magnetic moments is more restrictive, the energy shift due to the vector potential of the nucleus with the electron field is,

$$V_{Mag} = e\mathbf{A} \cdot \boldsymbol{\alpha}$$

where α is the Dirac vector-matrix. Due to the dependence on $\boldsymbol{\alpha}$ the interaction vanishes for closed shells and therefore to observe the effect of the MQM we can only consider paramagnetic atoms and molecules.

Chapter 3

Magnetic Quadrupole Moment of the Nucleus

Statement of Originality: In this chapter I present the results of my research. As this research builds upon recent work by Flambaum et al. [63] it follows a similar structure and uses many of the results. The research I conducted was the calculation of the core contribution for the MQM and its application to deformed nuclei and the effects on molecules, as well as the contribution of the EOM to molecules. Everything that was not my work has been properly referenced.

The MQM of the nucleus is the first T, P-odd magnetic moment as shown in Appendix A and Table 1.1. Therefore it is an extremely important probe to measure the T-odd and P-odd properties of nuclear, atomic and molecular systems.

3.1 Contributions to the MQM

There are two distinct contributions to the MQM, that of an unpaired nucleon EDM (M^a_{EDM}) and the other is an MQM generated from the internucleon T-P odd potential (as discussed in section 2.3) similar to the Schiff moment. The effect of the internucleon potential can be split into valence and core contributions ($M^a_{Val} + M^a_{Core}$). Therefore we have that,

$$M^a = M^a_{EDM} + M^a_{Val} + M^a_{Core} \tag{3.1}$$

Where a is an unpaired valence nucleon.

The contribution due to the EDM of the nucleon can be understood in a heuristic way, in elementary quantum mechanics we understand that the magnetic dipole moment of a system is due to the orbit of a charged particle. Similarly the MQM is induced in a system by the orbit of an EDM [14]. The MQM of a nucleus due to the EDM shown was by Khriplovich in 1976 [64] to be ,

$$M^a_{EDM} = d_a \frac{(2I-1)\left(\frac{1}{2}-\kappa\right)}{(I+1)}\lambda_p \tag{3.2}$$

$$\text{Where: } \kappa = (-1)^{I+\frac{1}{2}-l}\left(I+\frac{1}{2}\right) \tag{3.3}$$

where d_a is the EDM of the unpaired nucleon, λ_p is the Compton radius of the proton (where we have assumed $m_n \approx m_p$), I is the nuclear spin and l is the orbital angular momentum. As I can be written as $I = l \pm 1/2$, (3.2) is reduced to two cases,

$$M^a_{EDM} = d_a\,(2I-1)\,t_I\lambda_p \tag{3.4}$$

$$t_I = \begin{cases} 1 & I = l+\frac{1}{2} \\ -\dfrac{I}{(I+1)} & I = l-\frac{1}{2} \end{cases} \tag{3.5}$$

The valence contribution to the MQM generated through the inter-nucleon $P-\mathrm{T}-$odd potential (Section 2.3) was first done in [11]. It is found by taking the expectation value of the MQM operator (A.6) over the perturbed wavefunction (2.19). It is given by [11],

$$M^a_{Val} = \xi_a\,(\mu_a - q_a)\,(2I-1)\,t_I\lambda_p \tag{3.6}$$

where ξ^a is defined in (2.18), q is the nucleon charge and, μ_p and μ_n are the proton and neutron magnetic moments respectively.

3.2 Core Contribution

The core contribution due the the MQM was found by Dmitriev in 1996 [65] by using the contact form of the inter-nucleon potential (2.13) and separating out the core contribution from the valence contribution as discussed in section 2.2.2. Doing this

Dmitriev *et al.* found that the core contribution can be written as,

$$M^a_{Core} = M^a_\eta + M^a_{\eta'} \tag{3.7}$$

$$\text{Where : } M^a_\eta = -\frac{G}{\sqrt{2}} e \frac{2I-1}{2I+2} \frac{\left(\mu_p \eta_{pa} \frac{Z}{A} + \mu_n \eta_{na} \frac{N}{A}\right)}{2m^3\omega^2} \left\langle \frac{d^2\rho(r)}{d^2r} - \frac{1}{r}\frac{d\rho(r)}{dr} \right\rangle \tag{3.8}$$

$$M^a_{\eta'} = \frac{G}{\sqrt{2}} e \frac{2I-1}{2I+2} \frac{\left(\mu_p \eta_{pa} \frac{Z}{A} + \mu_n \eta_{na} \frac{N}{A}\right)}{m^3\omega^2} \left(\kappa - \frac{1}{2}\right) \left\langle \frac{1}{r}\frac{d\rho(r)}{dr} \right\rangle \tag{3.9}$$

where κ is defined in (3.3). Now we use the single particle oscillator model for the nuclear potential $U(r)$,

$$U(r) = \begin{cases} \dfrac{m_p\omega^2 r^2}{2} - U_0 & r < R \\ 0 & r > R \end{cases}. \tag{3.10}$$

Ignoring the spin orbit interactions we can assume that the profile of the nuclear density, $\rho(r)$, is the same as that of the central potential[11]. Therefore we have that,

$$\begin{aligned} \rho(r) &= -\frac{\rho(0)}{U(0)} U(r) \\ \Rightarrow \rho(r) &= \begin{cases} b(R^2 - r^2) & r < R \\ 0 & r > R \end{cases} \end{aligned} \tag{3.11}$$

where b is some constant and R is the nuclear radius. Now using the density defined in (3.11) we can find $\rho'(r)$ and $\rho''(r)$,

$$\frac{d\rho}{dr} = \begin{cases} -2br & r < R \\ 0 & r > R \end{cases} \tag{3.12}$$

$$\frac{d^2\rho}{dr^2} = \begin{cases} -2b & r < R \\ 0 & r > R \\ A\delta(r-R) & r = R \end{cases} \tag{3.13}$$

where the discontinuity of the first derivative features as a delta function of the second derivative. Here A normalisation constant of the delta function. This delta function describes the wavefunction at the nuclear surface, however as the wavefunction is small outside the nucleus we can assume it is small at the surface. Therefore for our analytical result we neglect the contribution of this delta term. Therefore using (3.12) and (3.13)

we can write,

$$\frac{d\rho}{dr} = r\frac{d^2\rho}{dr^2}.$$

We immediately see that in this model (3.8) vanishes and the core contribution is completely dependent on the exchange channels of the nucleon-nucleon interaction. Therefore we can rewrite (3.9) as,

$$\begin{aligned} M^a_{Core} = M^a_{\eta'} &= \frac{G}{\sqrt{2}} e \frac{2I-1}{I+1} \frac{\left(\mu_p \eta_{pa} \frac{Z}{A} + \mu_n \eta_{na} \frac{N}{A}\right)}{4m^3\omega^2} \left\langle \frac{2K}{r}\frac{d\rho}{dr} - \frac{1}{r}\frac{d\rho}{dr} \right\rangle \\ &= \frac{G}{\sqrt{2}} e \frac{2I-1}{I+1} \frac{\left(\mu_p \eta_{pa} \frac{Z}{A} + \mu_n \eta_{na} \frac{N}{A}\right)}{4m^3\omega^2} \left\langle \frac{2K}{r}\frac{d\rho}{dr} - \frac{d^2\rho}{dr^2} \right\rangle. \end{aligned} \tag{3.14}$$

Using this same method and model Dmitriev *et al.* [54] found that the valence contribution can be written as,

$$M^a_{Val} = \frac{G}{\sqrt{2}} \frac{e}{4m^3\omega^2} \frac{2I-1}{I+1} \eta_a \left(\mu_a - q_a\right) \left\langle \frac{d^2\rho}{dr^2} - \frac{2K}{r}\frac{d\rho}{dr} \right\rangle. \tag{3.15}$$

Therefore using equations (3.14) and (3.15) the ratio of the core to the valence contribution is,

$$\frac{M^{core}_{\eta'}}{M^a_{\eta}} = -\frac{\left(\mu_p \eta'_{ap} \frac{Z}{A} + \mu_n \eta'_{an} \frac{N}{A}\right)}{\xi_a \left(\mu_a - q_a\right)}. \tag{3.16}$$

Finally rewriting (3.1) using (3.2),(3.6) and(3.16) we have the final contribution of the MQM,

$$\begin{aligned} M &= M^a_{EDM} + M^a_{Val} + M^a_{Core} \\ &= M^a_{EDM} + M^a_{Val}\left(1 + \frac{M^a_{Core}}{M^a_{Val}}\right) \\ &= \left\{ d_a + \xi_a \left(\mu_a - q_a\right) \left[1 - \frac{\left(\mu_p \eta'_{ap} \frac{Z}{A} + \mu_n \eta'_{an} \frac{N}{A}\right)}{\eta_a \left(\mu_a - q_a\right)} \right] \right\} (2I-1)\, t_I \lambda_p \\ &= M^a_0 \,(2I-1)\, t_I \lambda_p, \end{aligned} \tag{3.17}$$

$$\text{Where: } M^a_0 = \left\{ d_a + 2 \times 10^{-21} \left[\eta_a \left(\mu_a - q_a\right) - \left(\mu_p \eta'_{ap} \frac{Z}{A} + \mu_n \eta'_{an} \frac{N}{A} \right) \right] \right\}. \tag{3.18}$$

This is a general form of the contribution of a single unpaired valence nucleon a. We will now examine the difference between the contribution of a proton and neutron.

3.3 Proton and Neutron Contributions

The CP violation parameter η_a from (3.17) can be split up depending on whether the nucleon is a proton or a neutron [66],

$$\eta_a = \begin{cases} \dfrac{Z}{A}\eta_{pp} + \dfrac{N}{A}\eta_{pn} & \text{for } a = p \\ \\ \dfrac{Z}{A}\eta_{np} + \dfrac{N}{A}\eta_{nn} & \text{for } a = n. \end{cases} \tag{3.19}$$

Using the charge ($q_p = 1$ and $q_n = 0$) and magnetic moments ($\mu_p = 2.79$ and $\mu_n = -1.91$) for the proton and neutron we have,

$$\begin{aligned} (\mu_p - q_p)\,\eta_p &= 1.79\left(\frac{Z}{A}\eta_{pp} + \frac{N}{A}\eta_{pn}\right) \\ (\mu_n - q_n)\,\eta_n &= -1.91\left(\frac{Z}{A}\eta_{np} + \frac{N}{A}\eta_{nn}\right). \end{aligned}$$

For large nuclei the ratio of protons and neutrons is approximately $\frac{N}{A} = 0.6$ and $\frac{Z}{A} = 0.4$. Using this approximation we have that,

$$\begin{aligned} (\mu_p - q_p)\,\eta_p &= 0.716\eta_{pp} + 1.074\eta_{pn} \\ (\mu_n - q_n)\,\eta_n &= -1.146\eta_{nn} - 0.764\eta_{np}. \end{aligned}$$

Therefore the MQM contribution for the proton and neutron is given by,

$$M_0^p = \left[d_p - 2\times 10^{-21}\ \mathrm{e}\cdot\mathrm{cm}\left(0.716\eta_{pp} + 1.074\eta_{pn} - 1.116\eta'_{pp} + 1.146\eta'_{pn}\right)\right] \tag{3.20}$$

$$M_0^n = \left[d_n - 2\times 10^{-21}\ \mathrm{e}\cdot\mathrm{cm}\left(-0.764\eta_{np} - 1.146\eta_{nn} - 1.116\eta'_{np} + 1.146\eta'_{nn}\right)\right]\lambda_p \tag{3.21}$$

We can rewrite this in a form with the ratio of the direct (η_{ab}) and exchange (η'_{ab}) parameters,

$$M_0^p = \left[d_p - 2\times 10^{-21}\ \mathrm{e}\cdot\mathrm{cm}\left(0.716\eta_{pp}\left(1 - \frac{1.116}{0.716}\frac{\eta'_{pp}}{\eta_{pp}}\right) + 1.074\eta_{pn}\left(1 + \frac{1.146}{1.074}\frac{\eta'_{pn}}{\eta_{pn}}\right)\right)\right]\lambda_p$$

$$M_0^n = \left[d_n + 2\times 10^{-21}\ \mathrm{e}\cdot\mathrm{cm}\left(1.146\eta_{nn}\left(1 - \frac{\eta'_{nn}}{\eta_{nn}}\right) + 0.764\eta_{np}\left(1 - \frac{1.116}{0.764}\frac{\eta'_{np}}{\eta_{np}}\right)\right)\right]\lambda_p.$$

In Appendix C we showed that the ratio is given by ,

$$\frac{\eta'_{NN}}{\eta_{NN}} = \frac{1}{4.4}. \tag{3.22}$$

It has also been shown that many body corrections reduce the strength of the CP-violating parameters by ≈ 1.5 times[52, 67]. Substituting these and (3.22) into (3.20) and (3.21) gives,

$$M_0^p = \left[d_p - 1.33 \times 10^{-21}\ \mathrm{e}\cdot\mathrm{cm}\,(0.462\eta_{pp} + 1.334\eta_{pn})\right]\lambda_p \tag{3.23}$$

$$M_0^n = \left[d_n + 2 \times 10^{-21}\ \mathrm{e}\cdot\mathrm{cm}\,(0.886\eta_{nn} - 1.018\eta_{np})\right]\lambda_p \tag{3.24}$$

Substituting the dependence of the T, P-odd parameters in terms of the isospin channels using (2.14) into (3.23) and (3.24) we have that,

$$M_0^p \approx \left[\frac{d_p}{1.2 \times 10^{-14}} + g\,(\bar{g}_1 + \bar{g}_2 - 0.5\bar{g}_0)\right]\left(1.2 \times 10^{-14}\ \mathrm{e}\cdot\mathrm{cm}\right)\lambda_p$$

$$M_0^n \approx \left[\frac{d_p}{1.27 \times 10^{-14}} + g\,(\bar{g}_1 - 0.12\bar{g}_2 + 0.07\bar{g}_0)\right]\left(1.27 \times 10^{-14}\ \mathrm{e}\cdot\mathrm{cm}\right)\lambda_p$$

where we have factored out the coefficient of the $\bar{g}_1$ channel. Finally it has been shown that using a Woods-Saxon potential the MQM is increased by ≈ 1.2 times in comparison with analytical calculations. Using this and substituting in for the proton Compton radius $\lambda_p = 2.1 \times 10^{-14}$ cm we find that the MQM of the nucleus induced by a proton and neutron is,

$$M_0^p = \left[\frac{d_p}{1.2 \times 10^{-14}\ e\cdot\mathrm{cm}} + g\,(\bar{g}_1 + \bar{g}_2 - 0.5\bar{g}_0)\right] 3 \times 10^{-28}\ e\cdot\mathrm{cm}^2 \tag{3.25}$$

$$M_0^n = \left[\frac{d_n}{1.27 \times 10^{-14}\ e\cdot\mathrm{cm}} + g\,(\bar{g}_1 - 0.12\bar{g}_2 + 0.07\bar{g}_0)\right] 3 \times 10^{-28}\ e\cdot\mathrm{cm}^2. \tag{3.26}$$

Comparing these with the valence MQM contribution only ($\eta'_{NN} = 0$) from reference [63]

$$M_0^{p/n} = \left[\frac{d_p}{1.4 \times 10^{-14}\ e\cdot\mathrm{cm}} + g\,(\bar{g}_1 + 0.4\bar{g}_2 - 0.2\bar{g}_0)\right] 3 \times 10^{-28}\ e\cdot\mathrm{cm}^2$$

we see that the effect of the core contribution greatly impacted the MQM for the proton and neutron in different ways despite depending only on the exchange channels. For the proton the the $\bar{g}_0$ and $\bar{g}_2$ channels have increased by a factor of 2.5 however

for the neutron the sign of the $\bar{g}_0$ and $\bar{g}_2$ channels has changed as well as reduced the magnitude by a factor of 3. In comparison the isovector channel, $\bar{g}_1$, has largely remained unaffected. As we will see this will have a large affect on the fundamental CP violating quantities depending on these channels.

3.4 QCD CP Violating $\tilde{\theta}$ for MQM

We now find the new dependence of the MQM on the QCD CP violation parameter $\tilde{\theta}$ (Section 2.4). It has been shown that the contribution to $\tilde{\theta}$ of the pion channel is $g\bar{g}_0 = -0.037\tilde{\theta}$ [68] and the proton EDM is $d_p = -1.2 \times 10^{-16}\tilde{\theta}\ e \cdot \text{cm}$ and neutron EDM is $d_n = 1.2 \times 10^{-16}\tilde{\theta}\ e \cdot \text{cm}$.

Therefore for the proton we see from (3.25) that the total contribution of the MQM to $\tilde{\theta}$ is,

$$M_0^p\left(\tilde{\theta}\right) = 5 \times 10^{-29}\tilde{\theta}\ e \cdot \text{cm}^2. \tag{3.27}$$

Comparing this to the valence only contribution of ($M_0^p\left(\tilde{\theta}\right) = 2 \times 10^{-29}\tilde{\theta}\ e \cdot \text{cm}^2$) [63] we see that the inclusion of the core interaction increases the contribution by a factor of 2.5. This is only for one valence proton and therefore this factor will become very important when considering the collective nature of the the MQM in deformed nuclei.

For the neutron we find a dependence of,

$$M_n\left(\tilde{\theta}\right) = -5 \times 10^{-30}\tilde{\theta}\ e \cdot \text{cm}^2. \tag{3.28}$$

Comparing this to the valence on TP odd potential contribution from ($M_0^n\left(\tilde{\theta}\right) = 2 \times 10^{-29}\tilde{\theta}\ e \cdot \text{cm}^2$)[63] we see that the sign has also changed and the contribution is an order of magnitude lower.

These changes could prove quite significant as without the core contribution the proton and neutron contributed the same amount and therefore the MQM was directly proportional to the number of valence nucleons. However with the opposite signs due to the core, the proton and neutron MQMs may result in a canceling out and the total MQM is not necessarily proportional to the number of nucleons but the ratio of valence protons and neutrons.

3.5 Dependence on Nucleon EDM and Quark chromo-DM

Other than the $\tilde{\theta}$ parameter we can also find the MQM dependence on the other fundamental CP-violating parameters discussed in Section 2.4, the nucleon EDM and quark EDMS and chromo-EDMs. As the core contribution only relates to the T, P-odd potential of the nucleus, the MQM generated by the unpaired nucleon has no dependence on the core contribution. Therefore from [63] we see that the dependence of the MQM on the EDM of the nucleon is given by,

$$\begin{aligned} M_0^p\left(d_p\right) &= 2.5 \times 10^{-14} d_p \text{ cm} \\ M_0^n\left(d_n\right) &= 2.5 \times 10^{-14} d_n \text{ cm} \end{aligned}$$

The quark EDMs are slightly less obvious. From [16] the quark EDMs ($d_{u,d}$) and the quark chromo-DMs ($\tilde{d}_{u,d}$) are related the the nucleon EDM and the isospin channels by,

$$\begin{aligned} d_p &= 1.1e\left(\tilde{d}_u + 0.5\tilde{d}_d\right) + 1.4d_u + 0.35d_d, \\ d_n &= 1.1e\left(\tilde{d}_d + 0.5\tilde{d}_u\right) + 1.4d_d + 0.35d_u, \\ g\bar{g}_1 &= 4 \times 10^{15}\left(\tilde{d}_u - \tilde{d}_d\right) \text{ cm}^{-1}, \\ g\bar{g}_0 &= 0.8 \times 10^{15}\left(\tilde{d}_u + \tilde{d}_d\right) \text{ cm}^{-1}. \end{aligned}$$

Therefore substituting these into the proton and neutron MQMs (3.25) and (3.26) we have that the MQM dependence on the chromo-DMs is,

$$\begin{aligned} M_0^n\left(\tilde{d}\right) &= \left(4.0985 \times 10^{15}\tilde{d}_u - 3.85 \times 10^{15}\tilde{d}_d\right) 3 \times 10^{-28}\ e \cdot \text{cm} \\ &\approx 1.2 \times 10^{-12}\left(\tilde{d}_u - \tilde{d}_d\right)\ e \cdot \text{cm}, \\ M_0^p\left(\tilde{d}\right) &= \left(4.5 \times 10^{15}\tilde{d}_u - 4.35 \times 10^{15}\tilde{d}_d\right) 3 \times 10^{-28}\ e \cdot \text{cm} \\ &\approx 1.4 \times 10^{-12}\left(\tilde{d}_u - \tilde{d}_d\right)\ e \cdot \text{cm}. \end{aligned}$$

Therefore comparing this to the valence only contribution in [63], $M_0^n\left(\tilde{d}\right) \approx M_0^n\left(\tilde{d}\right) = 1.2 \times 10^{-12}\left(\tilde{d}_u - \tilde{d}_d\right)\ e \cdot \text{cm}$ we see that the core contribution has little effect on the chromo-DM dependence. This is because with the core contribution the $g\bar{g}_1$ channel remained unchanged (refer to (3.25) and (3.26)) compared to [63]. Therefore as the major contribution to the quark chromo-DM is the $g\bar{g}_1$ channel the inclusion of the

core has little effect.

Therefore considering these results we will now focus on the MQM dependence on $\tilde{\theta}$ term and ignore the quark and nucleon EDM dependence.

3.6 MQM of Spherical and Deformed Nuclei

We now consider the MQM of specific A-odd nuclei. For spherical nuclei the case is simple, there is only one valence nucleon (a) with some spin $I = l \pm 1/2$. Therefore we just substitute this value into (3.17). For example the ^{201}Hg nucleus has a valence neutron with in the state $I = \frac{3}{2}^-$ [69] which implies $l = 1$ (odd parity state) and therefore $t_I = 1$. Substituting this into (3.17) we have that,

$$^{201}\text{Hg} : M = 2M_0^n.$$

We only consider one other spherical nucleus ^{137}Ba, which has an valence neutron in the $I = \frac{3}{2}^+$ state and therefore $l = 2$ which implies $I = l - 1/2$. Using (3.17) the MQM of the ^{137}Ba nucleus can be written as,

$$^{137}\text{Ba} : M = -1.2M_0^n.$$

These are the spherical nuclei cases where we see that there is only a small contribution. This is not the case for deformed nuclei where there is a collective contribution from many valence nucleons.

Approximately 50% of all nuclei are deformed. In a collective model we consider the nucleus as a whole and not just the single valence nucleon as in the spherical case. The motion of nucleons in a nucleus can be decomposed into a collective rotational component and intrinsic component [70]. The Hamiltonian of the system is not separable into these distinct components however it is an acceptable approach. The intrinsic nucleon interaction describes what happens in the nucleus in the rotating frame of the nucleus.

Due to the deformation of the nucleus, in the intrinsic frame the the orbitals with different angular momentum projections split and result in many partially filled shells[71]. Therefore in the intrinsic frame the total MQM will be the sum of all valence nucleons

in the rotational bands,

$$M_{zz}^{Intrinsic} = \sum M_{zz}^{Single}\left(I, I_z, l\right) n\left(I, I_z, l\right), \tag{3.29}$$

where $n\left(I, I_z, l\right)$ is the nucleon occupation number of the rotational band and $M_{zz}^{Single}\left(I, I_z, l\right)$ is the MQM due to a single valence nucleon in the shell with quantum numbers I, I_z and l. Note that orbitals with opposite projections of angular momentum give the same contribution to the MQM whereas for the Schiff moment they cancel.

We need to find the MQM of the state measured from the laboratory, therefore similar to the electric quadrupole moment (EQM) in [71] we can write the transformation as [63],

$$M^{lab} = \frac{I_t\left(2I_t - 1\right)}{\left(I_t + 1\right)\left(2I_t + 3\right)} M_{zz}^{Nucleus} \tag{3.30}$$

where I_t is the total nuclear spin of the nuclei.

For the contribution of the orbital states in deformed nuclei to the proton and neutron MQM we use the results of [63]. These are presented in Table 3.1.

Nucleus	M	Nucleus	M
^{181}Ta	$-14M_0^p - 11M_0^n$	^{229}Th	$0M_0^p - 19M_0^n$
^{173}Yb	$-10M_0^p - 10M_0^n$	^{177}Hf	$-19M_0^p - 14M_0^n$
^{179}Hf	$-13M_0^p - 13M_0^n$	^{137}Ba	$0M_0^p - 1.2M_0^n$
^{201}Hg	$0M_0^p + 2M_0^n$		

Table 3.1: This table presents the dependence of nuclei on the proton and neutron MQMs in (3.25) and (3.26)[63]. The coefficients are found with by using equations (3.30), (3.29) with the nuclear orbitals from [71].

Therefore we can now directly compared the contribution of the core interaction to the valence interaction for each nuclei. The results are shown in Table 3.2.

Here we can clearly see the large impact of the core contribution especially for the nuclei with large dependence due to valence neutrons. Four out of the five candidate collective nuclei showed an increase in magnitude the largest being ^{177}Hf where the core contribution increase the magnitude by a third. However the core contribution

Nucleus	M	Valence Contribution, $M\left(10^{-29}\tilde{\theta}\right)$ [63]	Core Contribution, $M\left(10^{-29}\tilde{\theta}\right)$	Total, $M\left(10^{-29}\tilde{\theta}\right)$
^{181}Ta	$-14M_0^p - 11M_0^n$	-50	-14	-64
^{173}Yb	$-10M_0^p - 10M_0^n$	-40	-5	-45
^{179}Hf	$-13M_0^p - 13M_0^n$	-52	-7	-59
^{201}Hg	$0M_0^p + 2M_0^n$	4	-5	-1
^{229}Th	$0M_0^p - 19M_0^n$	-38	48	9.5
^{177}Hf	$-19M_0^p - 14M_0^n$	-66	-22	-88
^{137}Ba	$0M_0^p - 1.2M_0^n$	2.4	-3	0.6

Table 3.2: This table shows the effective contribution of the valence only and the core only for the considered nuclei, with the total core contribution included.

seems to cast doubt on the ^{229}Th MQM. In all of the deformed nuclei ^{229}Th was the only one where the core contribution did not increase the magnitude, instead the core contribution cancels with the valence contribution to a large degree. This is worrisome as these analytic calculations have a large uncertainty ($\approx 20\%$) and therefore in the case of ^{229}Th we are subtracting a two uncertain numbers which leaves the result dubious at best.

Here we have calculated only the MQM of the nucleus. Experimentally this is not what we observe, instead we observe the nuclear MQM's effect on the atomic or molecular states (see Section 2.5). Therefore we now find the effect of the MQM on difference molecules.

3.7 MQM in Molecules

For the molecular case we wish to find the energy shift of the molecule due to the interaction of the MQM and the electron field. The interaction of the magnetic field with the electron field is,

$$V^{Mag} = e\mathbf{A} \cdot \boldsymbol{\alpha} \tag{3.31}$$

where $\boldsymbol{\alpha}$ is the Dirac matrix for the electron. Therefore for the quadrupole field we can use (A.2) and (A.4) from Appendix A.1 the vector potential for the MQM is,

$$A_i^{MQM} = -\frac{1}{6}\epsilon_{ijk}M_{jk}\partial_j\partial_k\frac{1}{R}$$

Therefore we can set $R = (r_m r_m)^{1/2}$ and expanding out the partial derivatives we can write,

$$\partial_l\partial_k\frac{1}{R} = \frac{3r_i r_k - r^2\delta_{ij}}{r^5}.$$

Therefore as the MQM tensor is symmetric the contraction with the delta and totally antisymmetric tensor vanishes,

$$\begin{aligned}\delta_{ij}\epsilon_{ijk}M_{jk} &= 0\\ \Rightarrow A_i &= -\frac{1}{2}\epsilon_{ijk}M_{jk}r_i r_k.\end{aligned}$$

From (3.31) the interaction of the MQM with the electron field is,

$$V^M = -\frac{1}{2}\epsilon_{ijk}M_{jk}r_i r_k\alpha_i.$$

Using the tensor structure of the MQM (B.1) from Appendix B we can rewrite the interaction as,

$$V^M = -\frac{3}{2}\frac{M}{2I\,(2I-1)}T_{jk}\frac{(\boldsymbol{\alpha}\times\mathbf{r})_j}{r^5}r_k. \tag{3.32}$$

For the molecular component we use the effective T-P odd spin rotational Hamiltonian derived in [62],

$$H_{Mol}^{PT} = W_d d_e \boldsymbol{S'}\cdot\mathbf{n} + W_S\frac{S}{I}\mathbf{I}\cdot\mathbf{n} - \frac{W_M M}{2I\,(2I-1)}\boldsymbol{S'}\hat{\mathbf{T}}\mathbf{n}. \tag{3.33}$$

Here the parameters W_d, W_S and W_M and the electron EDM, nuclear Schiff moment and nuclear MQM molecular parameters respectively, $\mathbf{I}$ is the nuclear spin and $\mathbf{n}$ and $\boldsymbol{S'}$ are the molecular axis and effective electronic spin projection respectively (for more details see Appendix D). Essentially this Hamiltonian represents the total energy shift due the CP-violating electromagnetic moments in molecular systems, the electron EDM dependence (d_e), the SM [11] as derived in Appendix A and the MQM as derived

above. We wish to find the magnitude of the MQM effect in molecules and therefore we will primarily focus on the MQM however it is important that the contribution of the MQM is roughly the same as or larger than the Schiff moment for MQM experiments to be valuable. Here W_M is the average of the MQM interaction with the electron field (3.32) of the molecule over the molecular states [72],

$$W_M = \frac{3}{2\Omega} \left\langle \Psi \left| \sum_i \left(\frac{\boldsymbol{\alpha}_i \times \mathbf{r}_i}{r_i^5} \right)_\zeta r_\zeta \right| \Psi \right\rangle$$

where Ω is the the projection of the total electron angular momentum along the molecular axis ζ (see Appendix D).

The diatomic molecular species we consider consist of those from Table 3.1 and a smaller atom like oxygen (O) or fluorine (F). The benefit of using these diatomics with a large charge asymmetry is that it allows us to consider the molecular state around the large nucleus only as $W_M \propto Z^2 R_M$[11] which means the smaller nucleus gives a negligible contribution, here R_M is the relativistic factor for the MQM atomic matrix elements. Therefore we can assume that the molecular wavefunction is concentrated in the vicinity of the large nucleus with the CP-violating electromagnetic moments. First we consider the molecules with electrons in the state $^2\Sigma_{1/2}$ ^{135,137}BaF, ^{173}YbF and ^{201}HgF.

To calculate the affect the MQM has on a molecule we need to find the MQM molecular parameter W_M for different molecules. Few direct calculations have been done for this parameter, however the electron EDM parameter, W_d, has been calculated for many molecules. Therefore we approach this problem by first finding a relationship between W_M and W_d.

In the vicinity of the large nucleus we can expand the molecular state $|\Omega\rangle$ in terms of the atomic orbitals of the nucleus. Neglecting the spin-orbit interaction and comparing the coupling to the orbital angular momentum axis we can write the molecular state as partial waves up to $l = 1$ as [62],

$$\begin{aligned} |\omega\rangle &= a\,|s\rangle\,|\omega\rangle + b\,|p_0\rangle\,|\omega\rangle + c\,|d_0\rangle\,|\omega\rangle + \dots \\ &= a\,|s_{1/2},\omega\rangle + b\left(-\frac{2\omega}{\sqrt{3}}\,|p_{1/2},\omega\rangle + \sqrt{\frac{2}{3}}\,|p_{3/2},\omega\rangle\right) + \dots \end{aligned} \tag{3.34}$$

where we have used Clebsch-Gordon coefficients to expand the partial waves, and a and b are normalisation constants. Here $\omega = \pm\frac{1}{2}$ is the spin projection of an electron in the molecular state where $\Omega = \sum_i \omega_i$. Now consider the interaction of the electron EDM, $\mathbf{d}$, and MQM, $\hat{\mathbf{M}}$, which mix electron states k_1 and k_2 through the interactions V^d and V^M respectively (here k_1 and k_2 have opposite parity). By the triangle rule for angular momentum and the tensor structure of the operators we have the requirement for the matrix elements,

$$\left\langle k_2 \left| V^d \right| k_1 \right\rangle \neq 0 \Rightarrow k_1 + k_2 \geq 1 \geq |k_1 - k_2| \tag{3.35}$$

$$\left\langle k_2 \left| V^M \right| k_1 \right\rangle \neq 0 \Rightarrow k_1 + k_2 \geq 2 \geq |k_1 - k_2| \tag{3.36}$$

Therefore considering (3.34) the MQM will only mix the $|s_{1/2}, \omega\rangle$ and $|p_{3/2}, \omega\rangle$ (higher order states are also mixed, such as $p_{3/2} \to d_{3/2}$, however the matrix elements are smaller) electron states, and the largest matrix element for the electron EDM will mix the $|s_{1/2}, \omega\rangle$ and $|p_{1/2}, \omega\rangle$ states. Therefore using (3.34) and taking the ratio of the MQM interaction and electron EDM interaction matrix elements we see the normalisation coefficients cancel and we can write,

$$\begin{aligned} \frac{W_M}{W_d} &= \frac{\left\langle \omega \left| V^M \right| \omega \right\rangle}{\left\langle \omega \left| V^d \right| \omega \right\rangle} \\ &= -\frac{\sqrt{2}}{2\omega} \frac{\left\langle s_{1/2}, \omega \left| V^M \right| p_{1/2}, \omega \right\rangle}{\left\langle s_{1/2}, \omega \left| V^d \right| p_{1/2}, \omega \right\rangle} \end{aligned} \tag{3.37}$$

where W^M and W^d are the molecular parameters defined in (3.33).

Now as mentioned above this holds for the non-relativistic electron states far away from the nucleus. However in (3.37) we have represented the ratio of molecular parameters in terms of atomic matrix elements. Using the relativistic form of these atomic matrix elements from [11, 13, 14] we can rewrite this as [63],

$$W_M = \frac{9R_M(Z)}{20r_0\alpha Z R_d(Z)} W_d$$

where R_M, R_d are the relativistic factors for the MQM and electron EDM respectively and r_0 is the Bohr radius.

For the MQM we only consider two molecular states, $^2\Sigma_{1/2}$ and $^3\Delta_1$. For the $^2\Sigma_{1/2}$

states, W_d and W_M have been numerically calculated by Kozlov *et al.* in [73, 62] for the molecules ^{135,137}BaF, ^{173}YbF and ^{201}HgF. These are presented in Table 3.3.

For the $^3\Delta_1$ terms we use recent calculations of W_M in [63]. These are presented in Table 3.3.

Molecule	I_t	Molecular State	$\lvert W_M \rvert$	$\lvert W_M MS \rvert$ (μHz)	
				Valence Only [63]	Core Included
			$\frac{10^{33}\text{Hz}}{e\cdot\text{cm}^2}$	$10^{10}\tilde{\theta}$	$10^{10}\tilde{\theta}$
^{135,137}BaF	$\frac{3}{2}$	$^2\Sigma_{1/2}$	0.83 [62]	1	0.25
^{173}YbF	$\frac{5}{2}$	$^2\Sigma_{1/2}$	2.1 [73]	42	47
^{201}HgF	$\frac{3}{2}$	$^2\Sigma_{1/2}$	4.8 [62]	10	2.5
^{177}HfF$^+$	$\frac{7}{2}$	$^3\Delta_1$	0.5	33	44
^{179}HfF$^+$	$\frac{9}{2}$	$^3\Delta_1$	0.5	26	30
^{181}TaN	$\frac{7}{2}$	$^3\Delta_1$	≈ 1	50	64
^{229}ThO	$\frac{5}{2}$	$^3\Delta_1$	1.9 [72]	72	18
^{229}ThF$^+$	$\frac{5}{2}$	$^3\Delta_1$	1.7	65	16

Table 3.3: This table compares the molecular frequency shift due to the nuclear MQM for the dependence on the strong CP $\tilde{\theta}$ parameter for the valence only contribution and the core contribution

From Table 3.3 we see that the contribution from the the core nucleons has a large effect on the frequency shift on most molecules. There is no overall increase or decrease and the change is case specific. The molecules which are affected the most in the inclusion of the core contribution are the molecules whose major nuclei have a disproportionate dependence on valence neutrons than protons such as thorium (Th) and mercury (Hg). This is due the order of magnitude reduction for the MQM's dependence for the neutron. However due to the increase of the protons dependence, molecules whose major nuclei has a large contribution from valence protons will be increased due to the core contribution, specifically the molecules of hafnium (Hf) and tantalum (Ta).

In summary the inclusion of the core contribution significantly alters the frequency shift due to the nuclear MQM of molecules. Nuclei with a large dependence on valence neutrons are reduced and those with a large dependence on protons are increased. This

casts doubt on the hope of using ^{229}ThO as a candidate for a smaller upper bound on the $\tilde{\theta}$ parameter as stated in recent work such as Ref[63] and Ref[72].

3.8 Molecular contribution of EOM

Other than the Schiff moment and MQM the next highest order CP-violating moment is the EOM. In this section I will present the EOM term of the spin rotational Hamiltonian and show that the EOM contribution to the molecular calculations is negligible in comparison to the MQM.

3.8.1 EOM Contribution to the Spin Rotational Hamiltonian

The interaction of the EOM potential (2.8) is simply,

$$V^O = e\phi^{(3)}_{octupole}$$
$$\text{Where: } \phi^3_{octupole}(R) \approx -eq\frac{1}{6}O_{ijk}\partial_i\partial_j\partial_k\frac{1}{R}$$

expanding out the partial derivatives in the potential gives us,

$$\phi^{(3)}_{octupole}(R) = -eq\frac{1}{6}O_{ijk}\frac{3\left(r_i\delta_{jk}+r_j\delta_{ik}+r_k\delta_{ij}\right)r^2-15r_ir_jr_k}{r^7}$$

As the octupole operator is traceless we immediately have that,

$$\phi^{(3)}_{octupole}(R) = eq\frac{15}{6}O_{ijk}\frac{r_ir_jr_k}{r^7}$$

Now substituting in the tensor form of the octupole moment((B.2) from Appendix B) we have the interaction of the EOM potential with the electron is given by,

$$V^O = e^2q\frac{15}{6}\frac{5O_{zzz}}{6I\left(I-1\right)\left(2I-1\right)}X_{ijk}\frac{r_ir_jr_k}{r^7}.$$

Now to find the EOM contribution to spin rotational Hamiltonian we have to average over the molecular state $|\Psi\rangle$. The interaction is independent on spin and therefore we do not need to introduce an effective spin term like the MQM. We can write the

interaction term as,

$$\begin{aligned} H_{EOM} &= \left\langle \Psi \left| V^O \right| \Psi \right\rangle \\ &= \left\langle \Psi \left| e^2 q \frac{15}{6} \frac{5O_{zzz}}{6I\left(I-1\right)\left(2I-1\right)} X_{ijk} \frac{r_i r_j r_k}{r^7} \right| \Psi \right\rangle \end{aligned}$$

For **n** being the unit vector along the molecular axis we write the Hamiltonian as,

$$H_{EOM} = \frac{15}{6} \frac{W_O O_{zzz}}{I\left(I-1\right)\left(2I-1\right)} X_{ijk} n_i n_j n_k$$

Where W_O is found from molecular calculations. In comparison to the MQM spin rotational Hamiltonian we see that the EOM does not depend on the effective spin, S', of the molecular state.

3.8.2 EOM Molecular Contribution

In this section we show that the EOM gives an insignificant contribution to the molecular calculation. We will give an order of magnitude estimate of the ratio of the EOM to MQM contribution. Similar to Section 3.7 we assume that in the diatomic molecule there is a large nuclei with a nuclear EOM and MQM and a small nucleus which we can neglect. Therefore we expand the molecular state $|\Omega'\rangle$ in terms of the atomic orbitals in the vicinity of the large nucleus. However for this case we assume that total projection of angular momentum of the molecular state is $|\Omega'| = \frac{3}{2}$ and therefore the expansion is given by,

$$\begin{aligned} |\omega'\rangle &= a\,|s\rangle\,|\omega'\rangle + b\,|p_0\rangle\,|\omega'\rangle + c\,|d_0\rangle\,|\omega'\rangle + \ldots \\ &= a\,|s_{3/2},\omega'\rangle + b\left(\frac{2\omega}{3}\sqrt{\frac{3}{5}}\,|p_{3/2},\omega'\rangle + \sqrt{\frac{2}{5}}\,|p_{5/2},\omega'\rangle\right) \\ &+ c\left(\sqrt{\frac{1}{5}}\,|d_{3/2},\omega'\rangle - 2\omega'\sqrt{\frac{2}{35}}\,|d_{5/2},\omega'\rangle + \sqrt{\frac{2}{7}}\,|d_{7/2},\omega'\rangle\right) + \ldots \end{aligned} \tag{3.38}$$

after expanding using Clebsch-Gordon coefficients. Here $\omega' = \pm\frac{3}{2}$ and $\Omega' = \sum_i \omega'_i$. Now as the EOM is a third rank tensor its interaction with the electrons must satisfy the

conditions,

$$\left\langle k_2 \left| V^O \right| k_1 \right\rangle \neq 0 \Rightarrow k_1 + k_2 \geq 3 \geq |k_1 - k_2|$$

where k_1 and k_2 are states of opposite parity. Therefore comparing this to (3.36) we see that both the EOM and MQM have non-zero matrix elements when mixing $|p_{3/2}, \omega'\rangle$ and $|d_{5/2}, \omega'\rangle$. Using (3.38) we write the ratio of the two matrix elements as,

$$\left(\frac{W_O}{W_M}\right)_{p_{3/2}\to d_{5/2}} = \frac{\left\langle p_{3/2}, \omega' \left| V^O \right| d_{5/2}, \omega' \right\rangle}{\left\langle p_{3/2}, \omega' \left| V^M \right| d_{5/2}, \omega' \right\rangle}.$$

Now consider the neutral atom of ^{209}Bi which has a single particle EOM [74]. This atom has a valence electron in the $|6p_{3/2}, 3/2\rangle$ state with the rest of the shells filled. Therefore we wish to find the matrix element of this mixing with the $|6d_{5/2}, 3/2\rangle$ for both the MQM and EOM interaction. These two matrix elements are given in [74] as (suppressing the principal quantum numbers),

$$\left\langle p_{3/2}, 3/2 \left| V^O \right| d_{5/2}, 3/2 \right\rangle \approx 1.9 \times 10^{-13} \eta_a \text{ cm}$$
$$\left\langle p_{3/2}, 3/2 \left| V^M \right| d_{5/2}, 3/2 \right\rangle \approx 1.3 \times 10^{-11} \eta_a \text{ cm}$$

where η_a is the CP-violating parameter from the valence only contribution of the P,T-odd Hamiltonian in Section 2.3. The ratio of these two matrix elements is,

$$\left(\frac{W_O}{W_M}\right)_{p_{3/2}\to d_{5/2}} \approx 0.015$$

This represents an upper-bound on the contribution of the EOM as there are atomic matrix elements for the MQM interaction much larger than $\left\langle p_{3/2}, 3/2 \left| V^M \right| d_{5/2}, 3/2 \right\rangle$ (such as $\left\langle s_{3/2}, 3/2 \left| V^M \right| p_{3/2}, 3/2 \right\rangle$) due to the much larger wavefunction of the $|s_{3/2}\rangle$ state than the $|d_{5/2}\rangle$ near the nucleus) and the ratio is smaller. Therefore we finally have that,

$$\frac{W_O}{W_M} < 0.015.$$

Therefore although there is a contribution from the nuclear EOM in paramagnetic molecules it is very insignificant compared to the MQM and Schiff moment contributions.

Chapter 4

Discussion and Conclusion

Although the study of CP-violating electromagnetic moments is over half a century old, it is still a very rich field on the forefront of physics. The continuous strengthening of limits on CP-violating phenomena promises to unveil new physics beyond the Standard Model, especially since current limits are already constraining possible SUSY theories. Therefore both theoretical and experimental research into possible mechanisms which strengthen these limits is crucial.

Experiments focusing on the MQM effects could be a large step towards measuring these CP-violating moments. Currently one of the best limits on CP-violating phenomena is the Schiff moment of the ^{199}Hg atom ($S\left(^{199}\text{Hg}\right) < 1.2 \times 10^{-12}\ e \cdot \text{fm}^3$ [75]) which in turn put limits on the nucleon EDMs, $d_n = 6.3 \times 10^{-26}\ e \cdot \text{cm}$ and $d_p = 8.6 \times 10^{-25}\ e \cdot \text{cm}$ [76]. These limits are roughly the same order of magnitude as the strongest current limit from direct measurement on the neutron[43] (see Table 1.2) and currently the best limit on the proton EDM. The Effect of the MQM on the molecular EDM is expected to be much larger than the Schiff moment as there is no electron shielding, close molecular levels of opposite parity and the collective effect of deformed nuclei. Therefore experiments into these molecules could place limits on the MQM orders of magnitude greater than the Schiff moment and therefore strengthen the limits on fundamental EDMs. Currently there are ideas for experiments to be performed on paramagnetic molecules focusing on molecular beams [77, 78, 79] and molecular ions traps [80]. These experiments present an excellent opportunity to test the effect of the MQM on molecular systems and obtain stronger upper bounds on fundamental CP-violating constants.

Currently there is interest in studying the ThO molecules due to the high MQM depen-

dent energy shift shown in reference [63]. However the inclusion of the core contribution casts doubt on this possibility as the core contribution cancels the valence contribution resulting in a small energy shift (Table 3.3) compared to other molecules. However this result contains a large degree of uncertainty as the valence and core contribution calculations have a large uncertainty. Therefore further research should be performed to see the degree of this cancellation. Another molecule with great experimental interest is ^{181}TaN, from Table 3.3 we see that including the core contribution has actually enhanced the frequency shift of this molecule by ≈ 1.3. Therefore this molecule is an excellent candidate to measure the effects of the MQM.

There is another topical area of physics where study into the MQM will have a large impact, particularly the strengthening of limits on the CP-violating QCD $\tilde{\theta}$ term. The strong CP problem revolves around this inexplicably small $\tilde{\theta}$, it has been speculated [30] that this CP-violating phenomena is a spontaneously broken symmetry and therefore a Goldstone boson known as the axion could be introduced to describe this dynamic field. Currently the studies on the axion are very topical as it is expected to be a candidate for cold dark matter, which is currently one of the largest cosmological mysteries. It is also suspected that the axion would create oscillating nuclear moments when interacting with different systems [81, 82, 83, 84, 85]. In particular it has been suggested that axions interacting for a recent review on axions and the connection to nuclear moments, particularly the nuclear MQM, and dark matter we refer to the reader reference [86].

To conclude, we have considered the nuclear MQM contribution in nuclei and molecules, primarily focusing on the core contribution. We found that the MQM has a large dependence on the core contribution using a harmonic oscillator model for the potential and density profile. The core contribution produced a large effect on deformed nuclei as it shares the collective properties of the valence contribution. Due to this large dependence it is important to consider the core contribution when calculating the effect the MQM has on the frequency shift in molecular experiments. We also showed that though the effect of a nuclear EOM is present in these paramagnetic molecules, its contribution compared to the nuclear MQM is negligible. As more experiments are performed on the target species we expect an increase in the order of magnitude on the limits of fundamental EDMs and $\tilde{\theta}$. This could have have far reaching implications in multiple fields of physics ranging from particle physics to cosmology.

Appendix A

Multipole Expansions

As the core of this thesis is study of nuclear moments to study CP-violation in atomic and molecular systems we present here the multipole expansion of both the scalar electric potential, $\phi(r)$ and magnetic vector potential,$\mathbf{A}(r)$, at a distance $\mathbf{R}$. For both cases we will use the Taylor expansion,

$$\frac{1}{|\mathbf{R}-\mathbf{r}|} = \frac{1}{R} - r_m \partial_m \frac{1}{R} + \frac{1}{2} r_m r_n \partial_m \partial_n \frac{1}{R} - \frac{1}{6} r_m r_n r_l \partial_m \partial_n \partial_l \frac{1}{R}. \tag{A.1}$$

The derivations of this section are primarily based off references [11, 74] with intermediate steps included.

A.1 Magnetic Multipoles

First we consider the magnetic multipoles of a system we look at the vector potential for the system,

$$A_i(\mathbf{R}) = \int \frac{j_i(\mathbf{r})\, d^3 r}{|\mathbf{R}-\mathbf{r}|}.$$

Where $j_i(\mathbf{r})$ is the electromagnetic current of the system. Considering the expansion (A.1) we see that the first term, which is the scalar component of the expansion, will correspond to the monopole of the system which is forbidden by Maxwell's laws and therefore we discard this term. Therefore we have up to the third order that the multipole expansion of the vector potential is given by,

$$\mathbf{A}_i(R) = -\int j_i r_m d^3 r \partial_m \frac{1}{R} + \frac{1}{2}\int j_i r_m r_n d^3 r \partial_m \partial_n \frac{1}{R} - \frac{1}{6}\int j_i r_m r_n r_l d^3 r \partial_m \partial_n \partial_l \frac{1}{R}$$

The first term corresponds to the magnetic dipole moment (MDM) and the third term corresponds to the magnetic octopole moment (MOM). As these terms are T, P-conserving we ignore them and only keep the second order term

$$\mathbf{A}_i^{(2)}(R) = \frac{1}{2}\int j_i r_m r_n d^3 r \partial_m \partial_n \frac{1}{R} \tag{A.2}$$

which corresponds to the magnetic quadrupole moment (MQM) of the potential. Using the current conservation law $\partial_n j_n = 0$ we can expand the partial derivative in the integral and rewrite in the form,

$$\begin{aligned} 0 &= \int \partial_p j_p r_m r_n r_k d^3 r \\ &= \int \left[\left(\partial_p j_p\right) r_m r_n r_k + j_p \left(\partial_p r_m\right) r_n r_k + j_p r_m \left(\partial_p r_n\right) r_k + j_p r_m r_n \left(\partial_p r_k\right) \right] d^3 r. \end{aligned}$$

The first term is obviously zero and using $\partial_i r_j = \delta_{ij}$ we can rewrite the integral as,

$$\begin{aligned} 0 &= \int \left[j_m r_n r_k + j_n r_m r_k + j_k r_m r_n \right] d^3 r \\ \Rightarrow \frac{1}{2}\left\langle j_i r_m r_n \right\rangle &\rightarrow \frac{1}{3}\left\langle \left(j_i r_m - j_m r_i\right) r_n \right\rangle \end{aligned} \tag{A.3}$$

Dropping the average notation we have the form of a third order tensor which is anti-symmetric in i and m. Therefore we can write it as,

$$\frac{1}{3}\left(j_i r_m - j_m r_i\right) r_n = \frac{1}{3} t_{imn}.$$

Therefore we can reduce it to a second order tensor by multiplying with the totally antisymmetric tensor,

$$\frac{1}{3} t_{imn} \epsilon_{ima} = \frac{1}{3} T_{na}.$$

Now we can decompose all second order tensors into the sum of a symmetric and anti-symmetric tensor,

$$T_{na} = A_{na} - M_{na}$$

The negative is convention. Multiplying each side again by the totally anti-symmetric tensor gives,

$$\begin{aligned} t_{imn}\epsilon_{ima}\epsilon_{kja} &= \epsilon_{kja}\left(A_{na} - M_{na}\right) \\ t_{imn}\left(\delta_{ik}\delta_{mj} - \delta_{ij}\delta_{mk}\right) &= \epsilon_{kja}\left(A_{na} - M_{na}\right) \\ t_{kjn} - t_{jkn} &= 2t_{kjn} \\ \Rightarrow t_{kjn} &= \frac{1}{2}\left(A_{na} - M_{na}\right) \end{aligned}$$

after contraction of indices and rearranging. We can do a similar decomposition in the anti-symmetric second rank tensor into a vector with the totally antisymmetric tensor we finally write that,

$$\frac{1}{3}t_{kjn} = \frac{1}{6}\epsilon_{kja}\left(\epsilon_{nal}a_l - M_{na}\right) \tag{A.4}$$

where M_{na} and a_l are the MQM and anapole moment (AM) respectively. Now to find the form of the M_{na} we multiply both sides by ϵ_{kjp} which gives,

$$\begin{aligned} \frac{\epsilon_{kjp}}{3}t_{kjn} &= 2\delta_{pa}\left(\frac{1}{4\pi}\epsilon_{nal}a_l - \frac{1}{6}M_{na}\right) \\ \Rightarrow M_{pn} &= -\epsilon_{imp}\left\langle\left(j_i r_m - j_m r_i\right) r_n\right\rangle + \frac{3}{2\pi}\epsilon_{pnl}a_l \end{aligned}$$

Therefore as M_{pn} is a symmetric tensor we can simplify the above as,

$$\begin{aligned} M_{pn} + M_{np} = 2M_{pn} &= -\epsilon_{imp}\left\langle\left(j_i r_m - j_m r_i\right) r_n\right\rangle - \epsilon_{imn}\left\langle\left(j_i r_m - j_m r_i\right) r_p\right\rangle \\ &= -2\epsilon_{imp}\left\langle j_i r_m r_n\right\rangle - \epsilon_{imn}2\left\langle j_i r_m r_p\right\rangle \\ \Rightarrow M_{pn} &= -\int\left(\epsilon_{imp}r_n + \epsilon_{imn}r_p\right) j_i r_m d^3 r. \end{aligned} \tag{A.5}$$

The AM is P-odd and T-even and therefore we will not consider it further as it is unrelated to CP violation.

Now the for the electromagnetic current we use the Pauli current,

$$\mathbf{j} = \frac{ie}{2m}q\left(\psi\nabla\psi^* - \psi^*\nabla\psi\right) + \frac{e\mu}{2m}\left[\nabla\times\psi^*\boldsymbol{\sigma}\psi\right]$$

and rewriting in tensor form is,

$$j_p = \frac{ie}{2m} q \left(\psi \partial_p \psi^* - \psi^* \partial_p \psi\right) + \frac{e\mu}{2m} \left[\epsilon_{pij} \partial_i \psi^* \sigma_j \psi\right]$$

where ϵ is the totally antisymmetric. The first term and second term are the orbital and rotational current respectively. To simplify the calculation we will consider each separately. Substituting the orbital current in gives,

$$\begin{aligned} M_{kn}^{Orbital} &= -\frac{ie}{2m} q \int \left(r_k \epsilon_{npq} + r_n \epsilon_{kpq}\right) r_q \left(\psi \partial_p \psi^* - \psi^* \partial_p \psi\right) d^3 r \\ &= -\frac{ie}{2m} q \int \left(r_k \epsilon_{npq} r_q \psi \partial_p \psi^* + r_n \epsilon_{kpq} r_q \psi \partial_p \psi^* - r_k \epsilon_{npq} r_q \psi^* \partial_p \psi + r_n \epsilon_{kpq} r_q \psi^* \partial_p \psi\right) d^3 r \\ &= -\frac{ie}{2m} q \int \left(r_k \psi \left(\mathbf{r} \times \nabla\right)_n \psi^* + r_n \psi \left(\mathbf{r} \times \nabla\right)_k \psi^* - r_k \psi^* \left(\mathbf{r} \times \nabla\right)_n \psi + r_n \psi^* \left(\mathbf{r} \times \nabla\right)_k \psi\right) d^3 r \end{aligned}$$

We know that the orbital angular momentum is given by, $\mathbf{l} = \mathbf{r} \times \mathbf{p}$ and we can rewrite the derivative in terms of the momentum $\nabla = i\mathbf{p}$. Therefore we have that $\mathbf{r} \times \nabla = i\mathbf{l}$. Substituting this in reduces the above to,

$$M_{kn}^{Orbital} = \frac{e}{m} \left\langle r_k l_n + r_n l_k \right\rangle .$$

The spin contribution is similar, substituting in gives,

$$M_{kn}^{Spin} = -\frac{e\mu}{2m} \int \left(r_k \epsilon_{npq} + r_n \epsilon_{kpq}\right) r_q \epsilon_{pij} \partial_i \psi^* \sigma_j \psi d^3 r$$

Now integrating by parts allows us to rewrite this as,

$$M_{kn}^{Spin} = \frac{e\mu}{2m} \int \left(\partial_i r_k r_q \epsilon_{npq} + \partial_i r_n r_q \epsilon_{kpq}\right) \epsilon_{pij} \psi^* \sigma_j \psi d^3 r$$

Therefore using $\delta_i r_j r_k = \delta_{qi} r_k + \delta_{ik} r_q$ and the totally antisymmetric tensor property $\epsilon_{ijk}\epsilon_{imn} = \delta_{jm}\delta_{kn} - \delta_{jn}\delta_{km}$ and contracting indices we have that,

$$\begin{aligned} M_{kn}^{Spin} &= \frac{e\mu}{2m} \int \left(3 r_k \psi^* \sigma_n \psi + 3 r_n \psi^* \sigma_k \psi - 2\delta_{kn} \psi^* \mathbf{r} \cdot \boldsymbol{\sigma} \psi\right) d^3 r \\ &= \frac{3e\mu}{2m} \left\langle r_k \sigma_n + r_n \sigma_k - \frac{2}{3} \delta_{kn} \mathbf{r} \cdot \boldsymbol{\sigma} \right\rangle \end{aligned}$$

Therefore combining the orbital and spin components we can rite the MQM tensor as,

$$M_{kn} = \frac{e}{2m} \left\langle 3\mu \left(r_k \sigma_n + r_n \sigma_k - \frac{2}{3} \delta_{kn} \mathbf{r} \cdot \boldsymbol{\sigma} \right) + 2q \left(r_k l_n + r_n l_k \right) \right\rangle . \tag{A.6}$$

where the first term is the spin dependent term and the second is the orbital term.

A.2 Electric Multipoles

The electric multipoles are found by expanding the electric potential. However this case is more complicated due to Schiff's theorem. To account for Schiffs theorem we write the scalar electric potential as[11, 74],

$$\phi(\mathbf{R}) = e\int \frac{\delta\rho(\mathbf{r})}{|\mathbf{R}-\mathbf{r}|}d^3r + \frac{1}{Z}(\mathbf{d}\cdot\nabla)\int \frac{\rho_0(\mathbf{r})}{|\mathbf{R}-\mathbf{r}|}d^3r. \tag{A.7}$$

Where $\rho_0(\mathbf{r})$ is the nuclear charge density, $\delta\rho(\mathbf{r})$ is the correction to the potential due to the TP odd inter-nucleon interaction and $\mathbf{d}$ is the EDM of the nucleus. We will see that this ensures a vanishing EDM in accordance to Schiff's theorem.

We now have to account for the recoil of the nucleus due the to orbit of the valence nucleon. the correction to the charge density is given by,

$$\delta\rho(\mathbf{r}) = q\delta\rho_\nu(\mathbf{r}) + \frac{1}{A}\nabla\rho_0(r)\langle\mathbf{r}\rangle$$
$$\text{For: } \langle\mathbf{r}\rangle = \int \delta\rho_\nu(\mathbf{r})\,\mathbf{r}d^3r$$

Now for $\mathbf{r} = q\rho_\nu + \frac{1}{A}\nabla\rho_0\langle\mathbf{r}\rangle$ we have that,

$$\phi(R) = eq\left(\int \frac{\rho_\nu}{|\mathbf{R}-\mathbf{r}|}d^3r + \frac{1}{Z}(\mathbf{d}\cdot\nabla)\int \frac{\rho_0}{|\mathbf{R}-\mathbf{r}|}d^3r\right)$$

Therefore using the expansion (A.1) we have that,

$$\phi(R) = eq\left[\int \rho_\nu\frac{1}{R}d^3r - \int \rho_\nu r_i d^3r\partial_i\frac{1}{R} + \frac{1}{2}\int \rho_\nu r_i r_j d^3r\partial_i\partial_j\frac{1}{R} - \frac{1}{6}\int \rho_\nu r_i r_j r_k d^3r\partial_i\partial_j\partial_k\frac{1}{R} + \right.$$
$$\left. + \frac{1}{Z}\langle r_m\rangle\partial_m\int \rho_0 d^3r\frac{1}{R} - \frac{1}{Z}\langle r_m\rangle\partial_m\int \rho_0 r_i d^3r\partial_i\frac{1}{R} + \frac{1}{2Z}\langle r_m\rangle\partial_m\int \rho_0 r_i r_j d^3r\partial_i\partial_j\frac{1}{R}\right].$$

The dipole terms cancel as $\int \rho_\nu r_i d^3r = \langle r_i\rangle$, $\int \rho_0 d^3r = Z$ and therefore we are left with,

$$\phi(R) = eq\left[\int \rho_\nu\frac{1}{R}d^3r + \frac{1}{2}\int \rho_\nu r_i r_j d^3r\partial_i\partial_j\frac{1}{R} - \frac{1}{Z}\langle r_m\rangle\partial_m\int \rho_0 r_i d^3r\partial_i\frac{1}{R} + \right.$$
$$\left. - \frac{1}{6}\int \rho_\nu r_i r_j r_k d^3r\partial_i\partial_j\partial_k\frac{1}{R}\frac{1}{2Z}\langle r_m\rangle\partial_m\int \rho_0 r_i r_j d^3r\partial_i\partial_j\frac{1}{R}\right]$$

The first three terms are P,T-even multipoles and are of no interest to us. Therefore as expected we are left with only third order P-,T-odd terms,

$$\phi^{(3)}(R) = eq\left[-\frac{1}{6}\int \rho_\nu r_i r_j r_k d^3r + \frac{1}{2Z}\langle r_k\rangle \int \rho_0 r_i r_j d^3r\right]\partial_i\partial_j\partial_k\frac{1}{R} \tag{A.8}$$

Making the transformation,

$$r_i r_j r_k = \left[r_i r_j r_k - \frac{1}{5}\left(\delta_{jk}r_i + \delta_{ik}r_j + \delta_{ij}r_k\right)r^2\right] + \frac{1}{5}\left(\delta_{jk}r_i + \delta_{ik}r_j + \delta_{ij}r_k\right)r^2$$
$$r_i r_j = \left[r_i r_j - \frac{1}{3}\delta_{ij}r^2\right] + \frac{1}{3}\delta_{ij}r^2$$

we see that the terms in the square brackets correspond to the traceless symmetric components of the tensors and the remainder is the trace. The traceless symmetric component corresponds to the octupole and quadrupole moments and the trace is the Schiff moment. Therefore substituting this into (A.8) and contracting the deltas in the traces we have that [74]

$$\phi^{(3)}_{octupole}(R) = -eq\frac{1}{6}O_{ijk}\partial_i\partial_j\partial_k\frac{1}{R} + \frac{eq}{2Z}Q_{ij}\langle r_k\rangle\partial_i\partial_j\partial_k\frac{1}{R} \tag{A.9}$$
$$\phi^{(3)}_{Schiff}(R) = -S_k\delta_k\delta^2\frac{1}{R} = 4\pi S_k\delta_k\delta(R)$$
$$\text{Where}\quad O_{ijk} = \int \rho_\nu\left(r_i r_j r_k - \frac{1}{5}\left(\delta_{jk}r_i + \delta_{ik}r_j + \delta_{ij}r_k\right)r^2\right)d^3r \tag{A.10}$$
$$Q_{ij} = \int \rho_0\left(r_i r_j - \frac{1}{3}\delta_{ij}r^2\right)d^3r \tag{A.11}$$
$$S_k = \frac{eq}{10}\left[\int \rho_\nu r_k r^2 d^3r + \frac{5}{3Z}\langle r_k\rangle\int \rho_0 r^2 d^3r\right] \tag{A.12}$$

Here O_{ijk} represents the EOM moment operator and Q_{ij} is the P-,T-even EQM moment. The EQM deformation will cause a screening effect of the EOM. However as only unpaired protons will contribute to this screening and it is proportional to $1/Z$ it is heavily suppressed especially given that we need large nuclei to find the EOM [74]. Therefore we can ignore this term and the octupole potential is given by,

$$\phi^{(3)}_{octupole}(R) = -eq\frac{1}{6}O_{ijk}\partial_i\partial_j\partial_k\frac{1}{R}$$

Although not largely considered in this thesis we have presented the form of the Schiff moment above. It should be noted that this is a rank 1 tensor and therefore it can be seen to represent an effective EDM of the nucleus despite the original one canceling

as shown above. This is primarily due to our artificial choice of static potential (A.7) such that the EDM canceled. However as Schiff's theorem is violated for a finite sized nucleus the EDM-like Schiff moment was recovered in the third order expansion.

Appendix B

Multipole Tensor Forms

In the previous appendix we found the form of the magnetic and electric multipoles through the expansion of potentials. However this gives us the electromagnetic for of the operators. There is another form of the multipoles we require, the tensor forms. For a nucleus the only quantity that can characterise a direction is the nuclear spin I, therefore we can write the multipole tensors using only angular momentum, for example the EDM is a vector and therefore it must have the form $\mathbf{d} = d_z I$. We will do this for both the MQM and EOM.

As the MQM is a second rank tensor it must be characterised by two quantum numbers.Also we require the MQM to be a traceless and symmetric we can write in the general form,

$$M_{ij} = A\left[I_i I_j + I_j I_i - \frac{2}{3} I\left(I+1\right)\delta_{ij}\right]$$

where A is some constant and I is the projection of the angular momentum in the z-direction ($I = I_z$). To see that this satisfies the conditions is trivial as $M_{ij} = M_{ji}$ and $\delta_{ij} M_{ij} = 0$.

Now we wish to have the MQM in terms of the maximal projection on the z-axis

and therefore we set $i = j = z$ and solve for A. This gives

$$\begin{aligned} A &= \frac{3}{2}\frac{M_{zz}}{I\left(2I-1\right)} \\ \Rightarrow M_{ij} &= \frac{3}{2}\frac{M_{zz}}{I\left(2I-1\right)}T_{i,k} \\ \text{Where: } T_{i,k} &= \left[I_iI_j + I_jI_i - \frac{2}{3}I\left(I+1\right)\delta_{ij}\right] \end{aligned} \tag{B.1}$$

This is the general tensor form of the MQM, here we can explicitly see the angular momentum restrictions of the MQM as the system is undefined for $I = 0, 1/2$ and therefore we must have $I > 1/2$. We have derived two forms of the MQM, (A.6) and (B.1).

Similarly for the EOM we have a third rank tensor and therefore it must be defined by three angular momentum vectors. It must also be traceless and symmetric, we can write the EOM in the general third rank tensor form,

$$O_{ijk} = A\left[I_iI_jI_k + I_jI_iI_k + I_kI_jI_i + I_iI_kI_j + I_jI_kI_i + I_kI_iI_j - \frac{6I\left(I+1\right)-2}{5}\left(I_i\delta_{jk} + I_j\delta_{ik} + I_k\delta_{ij}\right)\right]$$

Therefore substituting in $i = j = k = z$ the maximal projection of the octupole tensor on the z-axis, O_{zzz} is given by,

$$\begin{aligned} A &= \frac{5O_{zzz}}{6I\left(I-1\right)\left(2I-1\right)} \\ \Rightarrow O_{ijk} &= \frac{5O_{zzz}}{6I\left(I-1\right)\left(2I-1\right)}X_{ijk} \\ \text{Where: } X_{ijk} &= [I_iI_jI_k + I_jI_iI_k + I_kI_jI_i + I_iI_kI_j + I_jI_kI_i + I_kI_iI_j \\ &\quad - \frac{6I\left(I+1\right)-2}{5}\left(I_i\delta_{jk} + I_j\delta_{ik} + I_k\delta_{ij}\right)\Bigg] \end{aligned} \tag{B.2}$$

Therefore we have two forms for the octupole tensor, (A.10) and (B.2).

Appendix C

Isospin Channels

In this appendix we will demonstrate how to reduce the T, P-odd finite range nucleon-nucleon interaction into a contact form. Initially we have the CP violating Hamiltonian of the form [56, 57, 58, 59],

$$H_{TP} = -\frac{g}{8\pi m_p}\left[\left(g_0\boldsymbol{\tau}_1\cdot\boldsymbol{\tau}_2 + g_2\left(\boldsymbol{\tau}_1\cdot\boldsymbol{\tau}_2 - 3\tau_1^z\tau_2^z\right)\right)\left(\boldsymbol{\sigma}_1-\boldsymbol{\sigma}_2\right) + g_1\left(\tau_1^z\boldsymbol{\sigma}_1 - \tau_2^z\boldsymbol{\sigma}_2\right)\right]\nabla_1\frac{e^{-m_\pi r_{12}}}{r_{12}} \tag{C.1}$$

where 1 and 2 are the first and second nucleon, $\mathbf{r}$ is the position , $\boldsymbol{\sigma}$ is the spin matrix and $\boldsymbol{\tau}$ is the isospin matrix. This Hamiltonian is cumbersome and therefore to make it easier we will find a representation in the contact limit of the interaction. In the contact limit we assume the intermediate pion has a large mass , $m_\pi \to \infty$, and in this limit the exponential in (C.1) will reduce to a delta function such that,

$$\frac{e^{-m_\pi r_{12}}}{r_{12}} = A\delta\left(\mathbf{r}_1 - \mathbf{r}_2\right)$$

where A is a normalisation constant for the transformation. Therefore integrating over all space we have that,

$$\begin{aligned} A &= \int_{-\infty}^{\infty}\frac{e^{-m_\pi r}}{r}d^3r \\ &= 4\pi\int_0^\infty e^{-m_\pi r}rdr \end{aligned}$$

Therefore integrating by parts we have that $A = \frac{4\pi}{m_\pi^2}$ and therefore in the contact limit we can rewrite (C.1) as,

$$H_{TP} = \frac{G}{2\sqrt{2}m_p}\left(\frac{-g\sqrt{2}}{Gm_{\pi^2}}\right)\left[\left(g_0\boldsymbol{\tau}_1\cdot\boldsymbol{\tau}_2 + g_2\left(\boldsymbol{\tau}_1\cdot\boldsymbol{\tau}_2 - 3\tau_1^z\tau_2^z\right)\right)\left(\boldsymbol{\sigma}_1-\boldsymbol{\sigma}_2\right)\right.$$
$$\left. + g_1\left(\tau_1^z\boldsymbol{\sigma}_1 - \tau_2^z\boldsymbol{\sigma}_2\right)\right]\nabla_1\delta\left(\mathbf{r}_1-\mathbf{r}_2\right) \tag{C.2}$$

where we have introduced the Fermi weak constant, G. Now we wish to remove the isotopic dependence of the Hamiltonian and write it in an effective form,

$$H_{TP} = \frac{G}{\sqrt{2}}\frac{1}{2m}\left[\left(\eta_{ab}\boldsymbol{\sigma}_1 - \eta_{ba}\boldsymbol{\sigma}_2\right)\nabla\delta\left(\mathbf{r}_a-\mathbf{r}_b\right) + \eta'_{ab}\left[\boldsymbol{\sigma}_a\times\boldsymbol{\sigma}_b\right]\left\{\left(\mathbf{p}_a-\mathbf{p}_b\right),\delta\left(\mathbf{r}_a-\mathbf{r}_b\right)\right\}\right] \tag{C.3}$$

where a and b are either protons (p) or neutrons (n) and η_{ab} are the TP odd parameter for the direct channels. To get η_{ab} in terms of the isoscalar ($g\bar{g}_0$), isovector ($g\bar{g}_1$) and isotensor ($g\bar{g}_2$) components we find the expectation value of (C.2) for all the possible channels. The other parameter η'_{ab} corresponds to the exchange channels. This factor is small and we will only consider it at the end of this appendix.

Here we use the isospin convention where the τ matrices are given by

$$\tau^z = \begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix},\ \tau^x = \begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix},\ \tau^y = \begin{pmatrix}0 & -i\\ i & 0\end{pmatrix}$$
$$\tau^+ = \tau^x + i\tau^y = \begin{pmatrix}0 & 2\\ 0 & 0\end{pmatrix}$$
$$\tau^- = \tau^x - i\tau^y = \begin{pmatrix}0 & 0\\ 2 & 0\end{pmatrix}$$

Therefore we can write,

$$\begin{aligned}\boldsymbol{\tau}_1\cdot\boldsymbol{\tau}_2 &= \tau_1^z\tau_2^z + \tau_1^x\tau_2^x + \tau_1^y\tau_2^y\\ &= \tau_1^z\tau_2^z + \frac{1}{2}\left(\tau_1^+\tau_2^- + \tau_1^-\tau_2^+\right)\end{aligned}$$

Now these act of the proton and neutron states as,

$$|p\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \; |n\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

Therefore we have that the isospin matrix identities are,

$$\begin{aligned} \tau^z |p\rangle &= |p\rangle, & \tau^z |n\rangle &= -|n\rangle \\ \tau^+ |p\rangle &= 0, & \tau^+ |n\rangle &= 2|p\rangle \\ \tau^- |p\rangle &= 2|n\rangle, & \tau^- |n\rangle &= 0. \end{aligned}$$

With these we can find the effect form for all the possible channels. We will present the calculation for the proton-proton channel only, the calculation for the other channels are very similar.

Proton Proton Channel (pp)

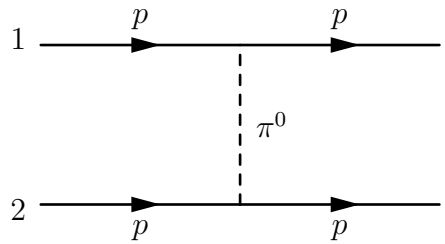

Figure C.1: The proton proton isospin scattering channel. 1 and 2 branch numbers represent to first and second particle in (C.2).

We take the expectation value of C.2 with respect the the state $|1,2\rangle$. Therefore for the direct proton-proton channel (Figure C.1) we have that,

$$\begin{aligned} \langle p,p\,|\,H_{TP}\,|\,p,p\rangle = \frac{G}{2\sqrt{2}m_p}\left(\frac{-g\sqrt{2}}{Gm_{\pi^2}}\right)\langle p,p\,|\,(g_0\boldsymbol{\tau}_1\cdot\boldsymbol{\tau}_2 + g_2\,(\boldsymbol{\tau}_1\cdot\boldsymbol{\tau}_2 - 3\tau_1^z\tau_2^z))\,(\boldsymbol{\sigma}_1-\boldsymbol{\sigma}_2)\,|\,p,p\rangle \\ + \langle p,p|g_1\,(\tau_1^z\boldsymbol{\sigma}_1 - \tau_2^z\boldsymbol{\sigma}_2)\,|p,p\rangle\,\nabla_1\delta\,(\mathbf{r}_1-\mathbf{r}_2) \end{aligned}$$

Therefore using the above relations of the isospin operators we have that,

$$\begin{aligned} \langle p,p\,|\,\tau_1^z\tau_2^z\,|\,p,p\rangle &= 1 \\ \left\langle p,p\,\middle|\,\tau_1^{\pm}\tau_2^{\mp}\,\middle|\,p,p\right\rangle &= 0 \\ \langle p,p\,|\,\tau_1^z\,|\,p,p\rangle = \langle p,p\,|\,\tau_2^z\,|\,p,p\rangle &= 1 \end{aligned}$$

and therefore we can write the above as,

$$\langle p,p\,|\,H_{TP}\left(\mathbf{r}_1-\mathbf{r}_2\right)|\,p,p\rangle=\frac{G}{2\sqrt{2}m_p}\left(\frac{-g\sqrt{2}}{Gm_{\pi^2}}\right)\left(\bar{g}_0-2\bar{g}_2+\bar{g}_1\right)\left(\boldsymbol{\sigma}_1-\boldsymbol{\sigma_2}\right)\nabla_1\delta\left(\mathbf{r}_1-\mathbf{r}_2\right).$$

Therefore comparing this to the contact limit for the pp channel

$$H_{TP}=\frac{G}{\sqrt{2}}\frac{1}{2m}\eta_{pp}\left(\boldsymbol{\sigma}_1-\boldsymbol{\sigma}_2\right)\nabla\delta\left(\mathbf{r}_1-\mathbf{r}_2\right)$$

we immediately see that,

$$\eta_{pp}=\left(\frac{g\sqrt{2}}{Gm_\pi^2}\right)\left(-\bar{g}_0+2\bar{g}_2-\bar{g}_1\right). \tag{C.4}$$

Using the mass of the neutral pion and the value of the Fermi weak constant we can write the factor in front of the TP odd parameter as,

$$\frac{\sqrt{2}}{Gm_\pi^2}=6.7\times10^6$$
$$\Rightarrow\eta_{pp}=6.7\times10^6\left(-\bar{g}_0+2\bar{g}_2-\bar{g}_1\right).$$

However this factor comes from the reduction of the finite pion exchange to a purely contact interaction, we account for this with a finite range correction. Comparing analytical results we derived to a numerical calculation with a finite range interaction we find that the finite range correction is a factor of 0.7 [56]. Therefore performing a similar calculation for the remaining channels we find the TP-odd constants are given by,

$$\begin{aligned}\eta_{pp}&=5\times10^6\left(-\bar{g}_0+2\bar{g}_2-\bar{g}_1\right)\\ \eta_{nn}&=5\times10^6\left(-\bar{g}_0+2\bar{g}_2+\bar{g}_1\right)\\ \eta_{pn}&=5\times10^6\left(g_0-2g_2-g_1\right)\\ \eta_{np}&=5\times10^6\left(g_0-2g_2+g_1\right).\end{aligned}$$

For all the above calculations we have ignored the exchange channels which are represented by η'_{ab} in the contact interaction. Although we could have calculated these in a similar fashion to the direct channels above it is unnecessary. Instead we just find the ratio between the direct and exchange contributions. Compared to the direct interactions the exchange interactions are small, in [11] the correction to the exchange channel

due to the contact limit is $\eta'_{ab} = 0.16\eta_0$ compared to the direct channels $\eta_{ab} = 0.7\eta_0$, where η_0 is the corrected interaction T-P-odd parameter. Therefore we can write the ration of the two channels as,

$$\begin{aligned} \frac{\eta'_{ab}}{\eta_{ab}} &= \frac{0.16}{0.7}\frac{\eta_0}{\eta_0} \\ &= \frac{1}{4.4} \end{aligned} \tag{C.5}$$

Appendix D

Molecular Quantum Numbers

As the focus of this thesis is the observation of T and P violating effects in diatomic molecules. We will present here a short overview of the quantum numbers of molecules. The conventions here are based off of those in [50] and our approach follows [62].

Similar to atomic states we define a molecular state by distinct quantum numbers. For diatomic molecules we say that the molecular axis is represented by the vector $\mathbf{n}$. Therefore we represent the orbital states of the molecule with orbital momentum $\mathbf{L}$, spin $\mathbf{S}$ and total momentum $\mathbf{j}_e$ by their projection on the molecular axis. [62]

$$\begin{aligned} \Lambda &= \langle \mathbf{L} \cdot \mathbf{n} \rangle , \\ \Sigma &= \langle \mathbf{S} \cdot \mathbf{n} \rangle , \\ \Omega &= \langle \mathbf{j}_e \cdot \mathbf{n} \rangle . \end{aligned} \tag{D.1}$$

Here $\langle\rangle$ denotes the average over the electronic states. For well defined quantum numbers Λ and Σ we have that

$$\Omega = \Lambda + \Sigma$$

which is simply that the total angular momentum projection of the molecule is the sum of the spin and orbital projection of the atomic orbitals. Now the classification of orbital angular momentum of molecular states is similar to standard atomic states. Where we represent the atomic states for $l = 0, 1, 2, 3, ...$ by $l = s, p, d, f$ respectively we represent the molecular orbital momentum by the capital greek counterparts D.1.

For molecular states we also have the multiplicity of spin due to degeneracy [50]. If the

Orbital Angular Momentum	0	1	2	3
Atomic Representation, l	s	p	d	f
Molecular Representation, Λ	Σ	Π	Δ	Φ

Table D.1: Comparison on the angular momentum notation between atomic and molecular states.

spin S is non-zero the multiplicity of the state is $2S+1$ as in atoms. Using these two properties we have a simple way of writing out a molecular state. We write the orbital angular momentum of the state with the spin as a subscript and the multiplicity as a superscript, that is, ${}^{2S+1}\Lambda_{\Omega}$. For example we write the molecular state with $\Lambda = 2$, $\Omega = 1$ and $S = 1$ as, ${}^{3}\Delta_{1}$.

It is important to note that all the molecular quantum numbers are T-,P-odd. This can easily be seen as in (D.1) all the quantum numbers depend on a T-odd pseudo-vector (angular momentum) and a T-even true vector (molecular axis $\mathbf{n}$).

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