

Methods, Analysis, and the Treatment of Systematic Errors for the Electron Electric Dipole Moment Search in Thorium Monoxide

Response to Referees

Dear Sir/Madam

We would like to thank the referees for the significant time and effort that they put into reviewing our work, and for the many useful comments that helped improve our paper. We also thank the editors for their assistance in this process. We include below our replies to all of the reviewers' comments.

Regards

Adam West

1 Reviewer 1

- *The relativistic factor for the enhancement of the electron electric dipole moment was derived for the first time in the paper Electron electric dipole moment enhancement in heavy atoms. V.V.Flambaum. Yad. Fiz. 24(2), 383, 1976 [Sov. J. Nucl. Phys. 24, 199 (1976)].*

This citation has been added as suggested in two places: above Eq. 6 and again shortly after.

- *The estimates for the molecules and advantages in using omega=1 molecules (like ThO) was for the first time presented for the first time in the paper Effects of parity nonconservation in diatomic molecules. O.P. Sushkov, V.V.Flambaum. JETP 48, 608-611, 1978 (ZhETF 75(10), 1208, 1978).*

This citation has been added as suggested in two places: in the sentence starting ‘In such species with a nucleus of atomic number...’ and in the sentence starting ‘The use of molecules with Ω -doublet structure...’. However, we note that the referee cited the paper’s title incorrectly: it should be ‘Parity breaking effects in diatomic molecules’ not ‘Effects of parity nonconservation in diatomic molecules’.

2 Reviewer 2

- *p.1 l.18 insert [27,42] : “EDMs of the electron [27, 42], neutron [19]”*

We assume that this should actually refer to p.2, and have added citations 27 and 42 as suggested.

- *p.2 l. 27: This should probably be $|d_e|$ not d_e as it is a limit to the magnitude.*

We have replaced d_e in equation 1 with $|d_e|$.

- *p.2 l.55 insert taking $\hbar = c = 1$: an n-loop process [24], taking $\hbar = c = 1$:*

This change made exactly as suggested.

- *p.3 l.6 “...violating phase. [Insert With these units, Eq(1) becomes $(d_e/e) < 5.0 \times 10^{-15} \text{ (GeV)}^{-1}$ and $m_e = 5.1 \times 10^{-4} \text{ GeV}$.] Assuming...”*

This addition was made as suggested. We believe that the value of d_e/e should be (unrounded) $4.7 \times 10^{-15} \text{ (GeV)}^{-1}$: With $\hbar = c = 1$, $1 \text{ eV}^{-1} = 2.0 \times 10^{-5} \text{ cm} \Rightarrow 1 \text{ cm} = 5.1 \times 10^4 \text{ eV}^{-1} = 5.1 \times 10^{13} \text{ GeV}^{-1}$, so $9.3 \times 10^{-29} \text{ cm} = 4.7 \times 10^{-15} \text{ GeV}^{-1}$.

- *p.3 ll.7,8 for one-loop processes of around 10 TeV. Similar analysis shows that our result was sensitive to two-loop effects at around the 1 TeV mass scale. Change 10 TeV to 8 TeV and change 1 TeV to 0.2 TeV.*

The use of equation 2 was intended to give order-of-magnitude estimates. As such we have changed the ‘=’ sign to ‘ \sim ’. We agree with the referee that the values should be changed. We show our working as follows. Note that we also modify Eq. 2 to read $\alpha/2\pi$ rather than $\alpha/4\pi$. For a 1-loop process:

$$\Lambda^2 = e \frac{m_e}{d_e} \frac{\alpha}{2\pi} \sin \phi_{\text{CP}} \quad (1)$$

$$\approx e \frac{m_e}{d_e} \frac{1}{274\pi}. \quad (2)$$

Using $m_e \approx 0.5 \text{ MeV}$, $d_e = 9.3 \times 10^{-29} \text{ e} \cdot \text{cm}$ and the fact that in natural units 1 eV^{-1} is equal to $1.97 \times 10^{-7} \text{ m}$ we then have

$$\Lambda \approx \sqrt{\frac{1 \times 10^{29}}{274\pi}} \text{ eV} \quad (3)$$

$$\approx 11 \text{ TeV}. \quad (4)$$

For the two-loop process one must multiply by an additional factor of $\sqrt{\alpha/2\pi} = 0.034$ to give $\Lambda \approx 0.4 \text{ TeV}$. Since these numbers are intended as order-of-magnitude estimates we then round them to give 10 TeV and 0.1 TeV.

- *p.3 ll.27 ff. This might seem surprising at first glance, since Schiff’s theorem states that there can be no net electric field acting on a non-relativistic point particle bound in a neutral system [43]. However, in 1958 Salpeter showed that, when relativistic effects are taken into account, a neutral species can experience an energy shift due to an eEDM when an external electric field E_{ext} is applied [44]. It is true that Salpeter first applied the correct relativistic treatment to the electron EDM, but still, the text here does not seem entirely fair to Schiff, who noted in [43] that his theorem would not apply when relativistic effects are considered.*

We have now added a note explaining that Schiff realised the theorem breaks down in the relativistic limit and reworded the following sentence about Salpeter’s treatment.

- *p.3 l. 31 Standards \rightarrow Sandars.*

Changed as described.

- *p. 3 l. 51. electron has significant wavefunction amplitude near a highly-charged nucleus. This is not enough: the electron density has to have a gradient (i.e. a superposition of odd and even parity functions).*

We have modified this explanation in line with the referee’s suggested modification.

- *p. 4 l. 44. Delete spurious it makes no sense in this context. (It means bogus, fake, not genuine, specious, false...).*

‘Spurious’ deleted.

- *p. 4 ll. 50,51. the internuclear axis is nearly aligned or anti-aligned with the applied electric field, [delete: and the alignment orientation is] [insert: as] described by*

Rewording performed as suggested.

- *p. 4. l.51. Define \hat{n} . (The direction of the internuclear axis, I presume). It should be made clear whether that points toward the Th or the O. (reference to appendix A)*

We have defined \hat{n} here and explained our direction convention with reference to the appendix.

- *p.4 l.56 The matrix element of $\vec{D} \cdot \hat{z}$, $|\langle H, J = 1, M = 1, \Omega | D \cdot z | H, J = 1, M = 1, \Omega \rangle|$ is zero. (\hat{z} is an axis in the laboratory). I think here one needs \hat{n} , assuming that is indeed the direction of the internuclear axis as I guessed at line 51.*

This change was made as stated. z is implicitly used (as is a common convention) as a molecule frame coordinate, however since we do not explicitly define it as such, using \hat{n} is much clearer.

- *p.5 l.38 (caption for Fig. 2). red arrows should read red arrow.*

Changed as described.

- *p.5 l.38 (caption for Fig. 2). blue arrows should read blue arrow.*

Changed as described.

- *p.6 starting l 48: It would be clearer to state here that the state preparation is done in the electric field. This only becomes apparent later on in the paper.*

Following sentence added to beginning of that paragraph: Preparation of a spin state occurs in an electric field such that the molecule is polarised and the molecule orientation, specified by \mathcal{N} , can be spectroscopically chosen.

- *p.10 l. 10: apostrophe points the wrong way*

Apostrophe reversed.

- *p.14 l30: Double dash ‘- -’ should be single dash-.*

Double dashes changed to single ‘en’ dash.

- *p.14 l. 41 mixture of tenses in this sentence is confusing.*

This sentence changed to ‘As described in section 3.2.4, we transferred the initial ground state population into $|H, J = 1\rangle$ via optical pumping.’

- *p. 14 section 3.2.3: This is really optical pumping, not rotational cooling. I would argue that the molecular population after pumping is no longer in a thermal distribution and so it doesnt make sense to give it a temperature.*

While the author is of course exactly right that temperature is only well-defined for a thermal distribution, we do not, for that reason, ascribe a temperature to the distribution after the optical pumping processes. In addition, the term ‘cooling’ is used informally in many areas, such as e.g. laser-cooling, where the resulting distribution is again not necessarily thermal, and has been adopted in recent literature on ‘rotational cooling’ of complex molecules and molecular ions in a similar context. We believe that the use of the term ‘cooling’ to describe a phase space compression, such as the compression of rotational population distribution used here, is sufficiently common and sufficiently well understood that it is a reasonable term to use in this instance.

- *p.15 l. 55: Probably the P_i state should have a superscript +, not a subscript.*

Changed as described.

- *p.16 l 55: Should use a different symbol from P for the footnote, as this has just been used for the state parity.*

Per the guide to authors, we are informed that these footnote superscripts will change during the copyediting process. Note to editor: please inform us if this is not the case!

- *p.32 paragraphs beginning on lines 7 and 43: In these paragraphs the authors describe how they subtract the background laser scatter from the molecular fluorescence signal. I feel that the authors should more clearly explain why they have opted to subtract a polarisation independent background $B(t)$ rather than separate backgrounds for the x and y polarised laser beams. Since these backgrounds produce a significant non-reversing phase, they must be different for the two laser beams. Given that no channels of interest changed significantly when the data was analysed with a polarisation dependent background, what is the motivation for using a polarisation independent background?*

We now clarify our choice of background subtraction method with an explanation starting at ‘If polarisation-dependent background subtraction...’.

- *p.32 Section 4.2.1: There seemed to be some confusing use of notation in this section in connection with the contrast C , which made it difficult to understand what exactly was being computed in equation 36. In equation 34 the state-averaged contrast is introduced as $C_j(\tilde{N}, \tilde{E}, \tilde{B})$ despite the fact that it is computed from the averages of two groups of asymmetry points, and so to be consistent with the definition of \bar{A} should be labelled $\bar{C}_j(\tilde{N}, \tilde{E}, \tilde{B})$. Then, $\tilde{C}_j(\tilde{N}, \tilde{E}, \tilde{B})$ is introduced as the average (or fit) of $C_j(\tilde{N}, \tilde{E}, \tilde{B})$ over the molecular pulse. If C_j has been averaged over the pulse, why does it still carry the j subscript? Perhaps the authors clarify their use of notation in this section or explain more clearly what is meant.*

There is indeed some confusion here. Firstly we believe the referee means to refer to \bar{A} and \bar{C} here rather than \tilde{A} and \tilde{C} , or perhaps has misread them as such. We have rewritten this section in order to make the way we compute contrast clearer. In particular:

- Section 4.2.1 ‘Accounting for Correlated Contrast’ has been removed and merged with the preceding section 4.2 ‘Computing Contrast and Phase’ and has been reworded throughout.
 - We more clearly distinguish between C_j , the contrast for a particular group j , and the ‘composite’ contrast, \tilde{C}_j , the contrast obtained from multiple groups, either via an average or a fit.
 - We have made our choice of sign convention for contrast more clearly specified within the paper.
 - We have more clearly and explicitly explained how we compute contrast in an experimental-state-dependent way, and how this helps make us resilient against contrast correlations.
 - We now use $|C|$ throughout where possible to avoid the potential ambiguity now explicitly described in section 4.2.
- *p. 36 l. 47: could → could.*

Changed as described.

- *p.37 l 26: Missing parenthesis: two approximately normally distributed random variables ($F_X - F_Y$ and $F_X + F_Y$), should be two approximately normally distributed random variables ($F_X - F_Y$) and ($F_X + F_Y$),*

Indeed — the parentheses were used to separate a grammatical clause, rather than in a mathematical sense. Changed to $(F_X - F_Y)$ and $(F_X + F_Y)$ for clarity.

- *p.44 line 35: As the authors note on p. 47 line 10, $H \rightarrow C$ transition is far from a typical transition with an $M1/E1$ ratio of 0.1. It would motivate the discussion that follows if a sentence could be added at this point to emphasise that in atypical transitions such as the spin-flip forbidden $H \rightarrow C$ or other transitions such as those which are parity forbidden, the corrections to the $E1$ transition moment can be far larger.*

A comment that we may expect higher order corrections to be larger than is typical has been added.

- *p.45 Table 3: -The spin operator in the molecular operator for the $M2$ transition is missing a superscript a .*

Change made as described.

- -In the caption, line 19 the authors should remind the reader that \hat{z} is the direction of the applied static electric field.

Change made as described.

- -The caption should explain what the subscripts i and j refer to in the molecular operators.

We have explained that the subscripts i and j refer to Cartesian components.

- p.45 Equations 48-50: -The matrix elements c_{E1} , c_{M1} and so on are scalars, but the molecular operators appear to be vectors. In order to evaluate expressions $\langle \dots | E1 | \dots \rangle$ should the relevant molecular operators be inserted into the equation at the top of Table 3?

The referee is correct that the relevant molecular operator should be inserted into the matrix elements at the top of Table 3. We have clarified this point in the caption to Table 3: ‘...E2, and M2 operators. To evaluate a transition matrix element one should substitute the operators of interest from the 3rd and 4th columns into the expression at the top of the table.’ To clarify further, substituting the molecular operators into that expression gives scalar quantities which are then multiplied by the polarisation vectors ϵ_{+1}^* and ϵ_{-1}^* to give vector quantities. These are then ‘dotted into’ the light vector \vec{V} , giving a scalar as expected.

- -Do the authors have any calculations or estimates of the values the matrix elements c_{E1} , c_{M1} and c_{E2} that could be included at this point in the text?

Unfortunately we do not have calculations of these matrix elements at the present time, beyond the simple order of magnitude estimates provided in this section which are not believed to be reliable.

- p.45 Equations 53: I think equation 53 should start with a +, or the order of the cross product should be reversed.

We believe that indeed the referee is correct. Our working is as follows: To obtain Eq. 53 we must evaluate the left-hand side of Eq. 52. First we take the definition of $\hat{\epsilon}_{\text{eff}}$:

$$\vec{\epsilon}_{\text{eff}} = \hat{\epsilon} - a_{M1} i \hat{n} \times (\hat{k} \times \hat{\epsilon}) + a_{E2} (\tilde{\mathcal{P}}) i (\hat{k}(\hat{\epsilon} \cdot \hat{n}) + \hat{\epsilon}(\hat{k} \cdot \hat{n})) + \dots \quad (5)$$

and note that $\hat{n} = \tilde{\mathcal{N}} \tilde{\mathcal{E}} \hat{z}$ to give

$$\vec{\epsilon}_{\text{eff}} = \hat{\epsilon} - a_{M1} i \tilde{\mathcal{N}} \tilde{\mathcal{E}} \hat{z} \times (\hat{k} \times \hat{\epsilon}) + a_{E2} (\tilde{\mathcal{P}}) i (\hat{k}(\hat{\epsilon} \cdot \tilde{\mathcal{N}} \tilde{\mathcal{E}} \hat{z}) + \hat{\epsilon}(\hat{k} \cdot \tilde{\mathcal{N}} \tilde{\mathcal{E}} \hat{z})) + \dots \quad (6)$$

We see that the first term, $\hat{\epsilon}$ does not reverse with any experimental switch, so $\hat{\epsilon}_{\text{eff}}^{\text{nr}} = \hat{\epsilon}$. The other two terms switch with $\tilde{\mathcal{N}}$ and $\tilde{\mathcal{E}}$ so we have

$$d\vec{\epsilon}_{\text{eff}}^{\mathcal{N}\mathcal{E}} \approx -a_{M1} i \hat{z} \times (\hat{k} \times \hat{\epsilon}) + a_{E2} (\tilde{\mathcal{P}}) i (\hat{k}(\hat{\epsilon} \cdot \hat{z}) + \hat{\epsilon}(\hat{k} \cdot \hat{z})). \quad (7)$$

Let’s just consider the a_{M1} term. As mentioned, we must use the left-hand side of Eq. 52. The denominator evaluates to 1, so we can ignore that. We are then left with

$$\hat{z} \cdot (\hat{\epsilon}_{\text{eff}}^{\text{nr}} \times d\vec{\epsilon}_{\text{eff}}^{\mathcal{N}\mathcal{E}}) \approx -a_{M1} i \hat{z} \cdot (\hat{\epsilon} \times (\hat{z} \times (\hat{k} \times \hat{\epsilon}))) \quad (8)$$

$$\equiv -a_{M1} i \hat{z} \cdot (\hat{\epsilon} \times (\hat{z} \times \vec{u})) \quad (9)$$

where \vec{u} is used as a placeholder. We can now rewrite the vector triple product to give

$$\hat{z} \cdot (\hat{\epsilon}_{\text{eff}}^{\text{nr}} \times d\vec{\epsilon}_{\text{eff}}^{\mathcal{N}\mathcal{E}}) \approx -a_{M1} i \hat{z} \cdot (\hat{z}(\hat{\epsilon} \cdot \vec{u}) - \vec{u}(\hat{\epsilon} \cdot \hat{z})) \quad (10)$$

$$\approx -a_{M1} i \hat{z} \cdot (\hat{z}(\hat{\epsilon} \cdot (\hat{k} \times \hat{\epsilon})) - (\hat{k} \times \hat{\epsilon})(\hat{\epsilon} \cdot \hat{z})) \quad (11)$$

$$\approx -a_{M1} i \hat{z} \cdot (-(\hat{k} \times \hat{\epsilon})(\hat{\epsilon} \cdot \hat{z})) \quad (12)$$

$$\approx i a_{M1} (\hat{\epsilon} \cdot \hat{z})(\hat{k} \times \hat{\epsilon}) \cdot \hat{z}. \quad (13)$$

Comparing with the result given in Eq. 53 we see that indeed there is a sign error.

- p.57 Equations 57: s is missing a subscript i .

Changed S to S_i on page 47, equation 57.

- p.58 line 40: microwave should be radio-frequency.

Changed as described.

- p.62 Table 4: It seems odd that the Stark interference effect described in section 5.2.2 isn't referred to in the systematic table. I understand the reasoning given in the top paragraph on p. 47, but I think it would be appropriate to mention the maximum estimated size of the effect in the table (or in the caption to the table).

We have repeated our estimate of the maximum size of the Stark interference systematic in this table caption.

- p.63 l 45: insert μ : the location parameter is the unknown \rightarrow the location parameter μ is the unknown.

Changed as described.

- p.67 Appendix 1: This is very clear, but for completeness should the Zeeman hamiltonian also be defined here?

The Zeeman Hamiltonian was in fact already defined, immediately prior to Section A.1.

- p.70 There is a logical problem below Eq(A.14). In order to fix it, line 12, immediately below Eq(A.14), should read where $A = Z + N$, we can write then comes Eq(A.15). Below that comes CS represents a weighted average etc.

These changes were implemented exactly as suggested.

- p. 70 l.21. Y_s has not been defined anywhere that I can find. I suppose it is the projection of \hat{n} onto \hat{z} but that needs to be made clear.

A definition of Y_S was added between Eq. A.15 and Eq. A.16.

- p.70 Eq(A.17) is missing a factor of C_s .

Factor of C_S added to Eq. A.17.