Effects of $CP$-violating internucleon interactions in paramagnetic molecules

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We demonstrate that electron electric dipole moment (eEDM) experiments with molecules in paramagnetic state are sensitive to $P,T$-violating nuclear forces and other $CP$-violating parameters in the hadronic sector. These experiments, in particular, measure the coupling constant $C_{SP}$ of the $CP$-odd contact semileptonic interaction. We establish relations between $C_{SP}$ and different $CP$-violating hadronic parameters including strength constants of the $CP$-odd nuclear potentials, $CP$-odd pion-nucleon interactions, quark-chromo EDM and QCD vacuum angle. These relations allow us to find limits on various $CP$-odd hadronic parameters.

I. INTRODUCTION

The existence of non-vanishing electric dipole moments (EDMs) of elementary particles was conjectured nearly seventy years ago [1–4], but they have not been observed so far. Their discovery would be a crucial step in the study of charge and parity ($CP$) violations. On the one hand, the Standard Model of elementary particles predicts non-vanishing values for the EDMs of the electron and nucleons, but these values are so minuscule that they are practically unobservable in current experiments. On the other hand, it is known that the $CP$-violation is needed to explain the apparent matter-antimatter asymmetry in the universe [5]. It is therefore an important challenge for experimental physics to measure the EDMs of elementary particles as well as EDMs of composite objects such as nucleons, nuclei and atoms.

In the last decade or so, tremendous progress has been achieved in the experiments with paramagnetic molecules which measure the electron EDM through specific energy level shifts [6, 7] (see also Refs. [6, 8] for reviews). As was demonstrated in the recent works [9, 10, 11], these experiments are also sensitive to the $CP$-violating interactions in the hadronic sector, which originate, in particular, from the nucleon EDMs. The aim of the current work is to extend the results of the papers [9, 10, 11] and to study the sensitivity of paramagnetic EDM experiments to $P,T$-violating nuclear forces.

It is a well-known fact that the $CP$-violating effects are significantly enhanced in heavy atoms [12]. In this paper, we consider a single-particle nuclear model in which the valence nucleon interacts with a heavy nuclear core through a nuclear potential $U(r)$. The $P,T$-violating nuclear forces are taken into account by the phenomenological interaction Hamiltonian $H_{\text{odd}} = \xi \sigma \cdot \nabla U(r)$, where $\sigma/2$ is the nucleon spin operator and $\xi$ is the coupling constant of dimension of length. This coupling constant will be denoted by $\xi_p$ for proton and $\xi_n$ for neutron. The authors of Ref. [13] demonstrated that the leading contributions to this interaction arise due to $\pi$ meson exchange between the valence nucleon and the nuclear core. In general, however, this interaction may arise due to the $CP$-violating $\pi NN$, $\eta NN$, three-pion and four-nucleon interactions. The last two interactions were considered, in particular, in Ref. [14].

In this paper, we will focus on the contributions to the atomic EDM arising due to the $P,T$-odd nuclear force with the Hamiltonian $H_{\text{odd}}$ regardless of the underlying fundamental interaction. In principle, experimental limits on the phenomenological parameters $\xi_{\text{p,n}}$ may be converted into limits on the parameters of more fundamental hadronic interactions. We stress that the interaction $H_{\text{odd}}$ considered in this work is an independent source of $CP$-violating effects, separate from the contributions due to nucleon EDMs considered in Refs. [10–11].

The most stringent experimental constraint on the electron EDM was obtained by the ACME collaboration [6] which measured specific energy level shifts in the $^{232}\text{ThO}$ molecule. This experiment also placed a limit on the $CP$-odd electron-nucleon interaction coupling constant (90% C.L.),

$$|C_{SP}|_{\text{Th}} < 7.3 \times 10^{-10}.$$  

In a heavy nucleus with $Z$ protons and $N = A - Z$ neutrons this coupling constant is a linear combination of independent electron couplings to protons ($C_{SP}^p$) and neutrons ($C_{SP}^n$), $C_{SP} = C_{SP}^p Z/A + C_{SP}^n N/A$. The latter two coupling constants correspond to the following $CP$-odd semileptonic operators

$$\mathcal{L} = \frac{i G_F}{\sqrt{2}} C_{SP}^p \bar{e} \gamma_5 e \bar{p} p + \frac{i G_F}{\sqrt{2}} C_{SP}^n \bar{e} \gamma_5 e \bar{n} n,$$  

where $G_F$ is the Fermi coupling constant, $e$, $p$ and $n$ are respectively the electron, proton and neutron fields.

Note that the subscript ‘$SP$’ denotes the nucleon-scalar and electron pseudoscalar two-fermion bilinears.

Our goal is to establish the leading-order relation between the coupling constant $C_{SP}$ and the parameters of the $P,T$-odd internucleon interaction $\xi_p$ and $\xi_n$, $C_{SP} = C_{SP}(\xi_p, \xi_n)$. This will allow us to find limits on these couplings originating from the experimental constraint [11]. Then, using known relations between the constants $\xi_p, \xi_n$ and more fundamental CP-violating hadronic parameters, we will establish leading-order relation between $C_{SP}$ and $CP$-odd pion-nucleon couplings $g^{(0,1,2)}_\pi$, quark-chromo EDMs $d_{u,d}$ and QCD vacuum angle $\theta$.

Note that this problem involves the third order perturbation theory in the nuclear part and second order in the
electron-nucleus interaction. To get through these complications we have to make some approximations in the nuclear part of the problem where we perform the calculations analytically. While all these approximations are common and justifiable, we cannot pretend that the accuracy of our results is better than a factor of two. However, this accuracy is comparable to that in other calculations of the hadronic contributions to atomic EDM where the limits on the CP-violating parameters are often presented on the logarithmic scale. For instance, current limit on QCD vacuum angle reads $|\theta| < 10^{-10}$, see, e.g., reviews $[8,9]$ and updated limits presented in Ref. $[15]$.

The rest of the paper is organized as follows. In Sect. III we present an estimate for the atomic EDM arising from the CP-odd nuclear forces. In Sect. III we compare this contribution to the atomic EDM with that of the contact electron-nucleon interaction and the nucleon permanent EDMs and find relations between the constant $C_{SP}$ and CP-violating hadronic parameters. In Sect. III we give a summary of our results and provide some comments on assumptions and precision. Technical details of calculations of electronic and nuclear matrix elements are collected in appendices.

Throughout this paper we use natural units with $c = \hbar = 1$.

II. CONTRIBUTIONS TO THE ATOMIC EDM FROM P,T-ODD NUCLEAR FORCES

In this section, we determine the contributions to the atomic EDM arising due to nuclear $P,T$-odd interactions. In Sect. II A we start with a review of the $P,T$-perturbed nuclear wave functions, which were found in Ref. $[13]$. These wave functions will be used in Sect. II B for the computation of the nuclear matrix elements contributing to the atomic EDM.

A. Nuclear wave functions perturbed by $P,T$-odd nuclear interactions

The nucleons in a nucleus can exhibit different $P,T$-odd interactions originating both from the Standard Model and beyond. Independently of their nature, in the non-relativistic limit, these short-range interactions may be taken into account by the following phenomenological single-particle Hamiltonian $[13]$

$$H_{\text{odd}} = \frac{G_F \cdot \eta \cdot \sigma \cdot \nabla \rho(r)}{\sqrt{2} 2m_p},$$

where $\sigma/2$ is the spin of the valence non-relativistic nucleon, $\rho(r)$ is the density of the nuclear core, $\eta$ is the effective coupling constant and $m_p$ is the proton mass.

In a heavy nucleus, the nuclear core creates an effective nuclear potential $U(r)$ in which the valence nucleon moves. In the short-range approximation, this potential is proportional to the density of the nuclear core, $\rho(r) = U(r)\rho(0)/U(0)$. Taking this into account, the Hamiltonian $[13]$ may be represented as

$$H_{\text{odd}} = \xi \sigma \cdot \nabla U(r),$$

where $[13]$

$$\xi = \frac{e G_F \cdot \rho(0)}{2\sqrt{2} 2m_p U(0)} \approx -2 \times 10^{-21} \eta \cdot cm.$$  

The total potential for the valence nucleon is thus given by

$$\tilde{U} = U + H_{\text{odd}} = U + \xi \sigma \cdot \nabla U \approx U(r + \xi \sigma).$$  

Let $|n'\rangle \equiv \psi_{n'}(r)$ be unperturbed wave function of the valence nucleon labeled by some quantum numbers $n'$. This function is supposed to solve for the Schrödinger equation with the potential $U(r)$. Equation (6) suggests that the wave function perturbed by the $P,T$-odd interaction $[13]$ may be represented as

$$\tilde{\psi}_{n'}(r) = \psi_{n'}(r + \xi \sigma) \approx (1 + \xi \sigma \cdot \nabla)\psi_{n'}(r),$$

or, more generally, taking the sum over all nucleons

$$|\tilde{n}'\rangle = \left(1 + \sum_{i=1}^{A} \xi_i \sigma_i \cdot \nabla_i\right)|n'\rangle.$$  

Note that the constants $\xi_i$ are different for proton ($\xi_i = \xi_p$) and neutron ($\xi_i = \xi_n$).

According to Eq. (8), the matrix elements of an operator $O$ may be written up to the first order in coupling constant $\xi$ as

$$\langle \tilde{m}'|O|\tilde{n}'\rangle = \langle m'|O|n'\rangle - \sum_{i=1}^{A} \xi_i \langle m'|[\sigma_i \cdot \nabla_i, O]|n'\rangle.$$  

The $P,T$-perturbed wave functions $[8]$ were found in Ref. $[13]$. In the next subsection we will use these wave functions to compute the nuclear matrix elements contributing to the atomic EDM.

B. Electron-nucleon interaction Hamiltonian

Let us consider a valence electron of charge $-e$ and position vector $R$ interacting with a valence nucleon of charge $q$ which is located at the point $r$. The interaction Hamiltonian considered in this paper is a combination of the electric ($q$) and magnetic ($\mu$) terms,

$$H_{\text{int}} = -H_{q} - H_{\mu},$$

$$H_{q} = \frac{qe}{|R - r|},$$

$$H_{\mu} = \frac{e \mu \cdot [(R - r) \times \alpha]}{|R - r|^{3}},$$

where $\alpha = \sigma \cdot \nabla$ and $\sigma$ is the spin of the valence nucleon. These wave functions will be used in Sect. IV, we present an estimate for the atomic EDM.
where

\[ \mu = \mu_0(g^I + g^s s) \]  

(11)

is the operator of nucleon’s magnetic moment, \( \mu_0 \) is the nuclear magneton, \( g^I \) and \( g^s \) are the orbital and spin factors of the nucleon. Note that \( s = \frac{1}{2} \sigma \) is the nuclear spin operator while \( \alpha = (0 \sigma \sigma) \) are the Dirac matrices acting on electron’s states.

The interaction Hamiltonian \( H_{\text{int}} \) as a function of \( R \) and \( r \) may be expanded into a multipole series. In particular, the leading terms in the expansion of the electric interaction Hamiltonian \( H_{\text{int}} \) are

\[ H_q = qe \left( \frac{1}{R} + \frac{R \cdot r}{R^3} \right) \theta(R - r) \]

\[ + qe \left( \frac{1}{r} + \frac{R \cdot r}{r^3} \right) \theta(r - R) + \ldots, \]  

(12)

where the ellipsis stand for terms with higher multipolarity. In this expansion, the term \( \theta(R - r)/R + \theta(r - R)/r \), after averaging with the nuclear charge density \( \rho(r) \), gives rise to the Coulomb interaction of the electron with the extended nucleus. This interaction is assumed to have already been taken into account by the unperturbed electronic wave functions.

The term \( (R \cdot r)/[\theta(R - r)/R + \theta(r - R)/r] \) in Eq. (12) is the leading dipole one on which we will focus our attention. Evidently, at a distance from the nucleus, this term falls off as \( 1/R^2 \). To find the effective electron-nucleon interaction Hamiltonian inside the nucleus, one needs to average this term over the normalized nuclear density, which, in the leading approximation, may be taken as constant inside the sphere of radius \( R_0 \) and vanishing outside, \( \rho(r) = 3\theta(R_0 - r)/R_0^3 \). As a result, one obtains

\[ f(R) \equiv \int_0^\infty \rho(r) \left[ \frac{1}{R^2} \theta(R - r) + \frac{1}{r^2} \theta(r - R) \right] r^2 dr \]

\[ = \frac{1}{R^2} \theta(R - R_0) + \frac{3R_0 - 2R}{R_0^3} \theta(R_0 - R). \]  

(13)

Using this prescription for the continuation of the \( 1/R^2 \) function to small distances, one may extract the regularized dipole interaction from the operator \( H_{\text{int}} \),

\[ \hat{H}_q \equiv qe(R \cdot r)f(R), \]  

(14)

where \( \hat{R} \equiv R/R \).

The magnetic interaction operator \( \hat{H}_{\mu} \) also behaves as \( 1/R^2 \) at large distances from the nucleus and may also be extended to the short-distance region inside the nucleus according to the prescription \( \hat{H}_\mu \),

\[ \hat{H}_\mu \equiv \epsilon \mu \cdot (\hat{R} \times \alpha)f(R). \]  

(15)

Thus, the dipole part of the interaction \( H_{\text{int}} \) which is regularized at short distances reads

\[ \hat{H}_{\text{int}} = -\hat{H}_q - \hat{H}_\mu, \]  

(16)

with \( \hat{H}_q \) and \( \hat{H}_\mu \) given by Eqs. (14) and (15).

According to Eq. (9), to take into account the \( P,T \)-perturbed nuclear wave functions one has to consider the commutators of the operator \( \sigma \cdot \nabla \) with the interaction Hamiltonians \( H_{\text{int}} \) and \( H_{\mu} \),

\[ [\sigma \cdot \nabla \cdot \hat{H}_q] = qe(\sigma \cdot \hat{R})f(R), \]

(17a)

\[ [\sigma \cdot \nabla \cdot \hat{H}_\mu] = i\epsilon \mu_0(g^s - g^I)\hat{f}(R)(\hat{R} \cdot \alpha) \cdot (\sigma \cdot \nabla \cdot \hat{R}). \]  

(17b)

In deriving Eq. (17b) we have applied the following commutator identities: \( [\sigma_i, \sigma_j] = 2i\varepsilon_{ijk}\sigma_k \) and \( [\nabla_i, \ell_j] = i\varepsilon_{ijk}\nabla_k \).

C. Atomic EDM due to \( P,T \)-odd nuclear forces

The unperturbed atomic states will be denoted by \( |n \rangle = |n\rangle |n\rangle \), where \( |n\rangle \) and \( |n\rangle \) are electronic and nuclear states, respectively. In what follows, the nuclear quantum numbers will be distinguished from the electronic ones with the apostrophe. As in Sect. III the \( P,T \)-perturbed nuclear wave functions \( \tilde{n} \) are denoted as \( \tilde{n} \).

The atomic EDM arising from the mixed interaction \( H_{\text{int}} \) may be calculated in perturbation theory. The first-order contribution to the atomic EDM vanishes for spinless nuclei which we consider in this paper,

\[ \sum_{n \neq 0} \frac{\langle 0 | -eR | \tilde{n} \rangle \langle \tilde{n} \tilde{n} | \hat{H}_{\text{int}} | \tilde{0} \rangle}{E_0 - E_n} = 0. \]  

(18)

Indeed, it may be shown that \( \langle \tilde{0} \hat{H}_{\text{int}} | \tilde{0} \rangle \propto \langle \tilde{0} | s | 0 \rangle = 0 \), where \( s \) is the nuclear spin operator.

The leading non-vanishing contributions to the atomic EDM thus arise in the second-order perturbation theory,
where the \( \text{sgn}(E_n) \) in the denominators is needed to correctly account for the negative energy electronic states. Indeed, the negative energy electronic states contribute with opposite sign of the nuclear energy because they may be viewed as blocking contributions which prevent the valence electron from directly transitioning into such states which are supposed to be completely occupied in the Dirac sea picture.

The term in the second line in Eq. (19) may be neglected in comparison with the other one because it is suppressed by higher power of nuclear energy in the denominator. Moreover, we assume that the leading contributions to the atomic EDM arise from the matrix elements with \([0] = |s_{1/2}\rangle\) and \(|m\rangle = |p_{1/2}\rangle\) electronic states because these wave functions are significantly enhanced in the vicinity of a heavy nucleus. Taking this into account, Eq. (19) may be cast in the form

\[
d \approx 2 \frac{(s_{1/2})|E_p|p_{1/2}^2|M_{\text{eff}}|s_{1/2}^2)}{E_{p_{1/2}} - E_{s_{1/2}}} , \quad (20)
\]

\[
\langle p_{1/2}|H_{\text{eff}}|s_{1/2}\rangle = - \xi \sum_{nn' \neq 00'} \frac{\langle p_{1/2}|0'\rangle \langle \sigma \cdot \nabla, \tilde{H}_n|n'n\rangle \langle nn'|\tilde{H}_n|0's_{1/2}\rangle}{E_n + \text{sgn}(E_n)\Delta E_{n'}} \quad + (s_{1/2} \leftrightarrow p_{1/2}) . \quad (22)
\]

Here, for brevity, we use the generic symbol \( \xi \) to uniformly denote \( \xi_p \) and \( \xi_n \). The specific proton and neutron contributions will be displayed explicitly in the final section.

Substituting Eq. (16) into Eq. (22), we express this matrix element in terms of the operators (14) and (15).

\[
\langle p_{1/2}|H_{\text{eff}}|s_{1/2}\rangle = \sum_{nn' \neq 00'} \frac{\xi (\mathcal{M}^{1}_{nn'} + \mathcal{M}^{2}_{nn'})}{E_n + \text{sgn}(E_n)\Delta E_{n'}} , \quad (23)
\]

where

\[
\mathcal{M}^{1}_{nn'} = \langle p_{1/2}|0'\rangle \langle \sigma \cdot \nabla, \tilde{H}_n|n'n\rangle \langle nn'|\tilde{H}_n|0's_{1/2}\rangle , \quad (24a)
\]

\[
\mathcal{M}^{2}_{nn'} = \langle p_{1/2}|0'\rangle \langle \sigma \cdot \nabla, \tilde{H}_n|n'n\rangle \langle nn'|\tilde{H}_n|0's_{1/2}\rangle . \quad (24b)
\]

These matrix elements will be calculated in the next subsection.

### D. Calculation of matrix elements

Consider the matrix elements in Eq. (24). Using the identities (15) and (17a), one may separate its electronic and nuclear components as

\[
\mathcal{M}^{1}_{nn'} = g e^2 \langle 0'|\sigma|n'\rangle \langle n'|\mu|0'\rangle \\
\times \langle p_{1/2}|f(R)|n\rangle \langle n|f(R)|\tilde{R} \times \alpha|s_{1/2}\rangle . \quad (25)
\]

Note that in the product of the nuclear matrix elements we may single out the scalar term which gives dominant contribution in spinless nuclei,

\[
\langle 0'|\sigma|n\rangle \langle n'|\mu|0'\rangle = \frac{1}{3} \delta_{ij} \langle 0'|\sigma|n'\rangle \langle n'|\mu|0'\rangle + \ldots , \quad (26)
\]

where the ellipsis stands for the tensor terms which we omit in further calculations. With the use of the definition (11), the expression \(\langle 0'|\sigma|n\rangle \langle n'|\mu|0'\rangle\) reduces to the nuclear spin-flip matrix element

\[
\langle 0'|\sigma|n\rangle \langle n'|\mu|0'\rangle = 2 \mu_0 (g^* - \epsilon g) (0'|s|n')^2 , \quad (27)
\]

where

\[
\epsilon = \left\{ \begin{array}{ll} 1 & \text{for spherical nuclei} \\ 0 & \text{for deformed nuclei} . \end{array} \right. \quad (28)
\]

In Eq. (27), we have taken into account the fact that for spherical nuclei, the states are usually represented in the \( lj \)-basis in which \(\langle 0'|s|n\rangle \langle n'|j|0'\rangle = 0 \) while the states of deformed nuclei are usually represented in the \( ls \)-basis with \(\langle 0'|s|n\rangle \langle n'|l|0'\rangle = 0 \).

With the use of Eqs. (26) and (27), the matrix element (25) may now be cast in the form

\[
\mathcal{M}^{1}_{nn'} = \frac{2}{3} \mu_0 (g^* - \epsilon g) \langle 0'|s|n'\rangle^2 \\
\times \langle p_{1/2}|f(R)|n\rangle \langle n|f(R)|\tilde{R} \times \alpha|s_{1/2}\rangle . \quad (29)
\]

Similarly, one may write the expression (24) for \(\mathcal{M}^{2}_{nn'}\) as

\[
\mathcal{M}^{2}_{nn'} = \frac{i}{3} \epsilon g_0 (g^* - \epsilon g) \langle 0'|\sigma \times \nabla, |n'|\rangle \langle n'|\tau|0'\rangle \\
\times \langle p_{1/2}|f(R)|\tilde{R} \times \alpha|n\rangle \langle n|f(R)|\tilde{R} \times |s_{1/2}\rangle . \quad (30)
\]
Here \(\langle n'|\sigma|0'\rangle\) is the E1 nuclear transition matrix element which may be considered within the giant dipole resonance model. Effectively, this means that the sum over \(n'\) is dominated by the matrix elements \(\langle n'|\sigma|0'\rangle\) which constitute the giant electric dipole resonance with the excitation energy \(\Delta E\). Then having fixed the nuclear energy in the denominator of Eq. (30), one might use the completeness relation for the nuclear states, \(|n'\rangle\langle n'\rangle\), to reduce the nuclear matrix elements in Eq. (30) to the expectation value of the \(1\cdot s\) operator,

\[
\langle 0'|\sigma \times \nabla_r|n'\rangle\langle n'|\sigma|0'\rangle \approx -2i\langle 0'|1\cdot s|0'\rangle \equiv -2i(1\cdot s) . \tag{31}
\]

With this expression for the nuclear matrix element, Eq. (30) simplifies to

\[
\mathcal{M}_{n'n}^2 = \frac{2}{3}e^2q\mu_0(g^* - g')(0'|1\cdot s|0') \times \langle p_{1/2}|f(R)\hat{R} \times \alpha|n\rangle\langle n|f(R)\hat{R}|s_{1/2}\rangle . \tag{32}
\]

Substituting Eqs. (30) and (32) into Eq. (29), one may represent the matrix element of the effective operator \(\mathcal{H}_{\text{eff}}\) in the compact form

\[
\langle p_{1/2}|\mathcal{H}_{\text{eff}}|s_{1/2}\rangle = 2\xi q\mu_0 \left[ \sum_{n'} (g^* - g')(0'|s|n')^2 + (g^* - g')(\Delta E)|0'|1\cdot s|0'\rangle \right] , \tag{33}
\]

where

\[
M(E) = \frac{e^2}{3} \sum_n \frac{\langle p_{1/2}|f(R)\hat{R}|n\rangle\langle n|f(R)\hat{R} \times \alpha|s_{1/2}\rangle}{\Delta E_n + \text{sgn}(E_n)E} \nonumber \tag{34}
\]

Note that the sum in Eq. (34) contains only single-particle nucleon excitations. It is instructive to separate proton \((p)\) and neutron \((n)\) contributions with nuclear excitation energies denoted by \(\Delta E_p\) and \(\Delta E_n\) as

\[
\langle p_{1/2}|\mathcal{H}_{\text{eff}}|s_{1/2}\rangle = 2\rho_0 \sum_{i=p,n} \xi_i q_i \left[ (g^i_p - g^i_n)M_i + (g^i_n - g^i_p)M(1\cdot s)_i \right] , \tag{35}
\]

where

\[
M_p \equiv \sum_{\Delta E_p} \langle 0'|s|n'\rangle_p^2 M(\Delta E_p) , \tag{36}
\]

\[
M_n \equiv \sum_{\Delta E_n} \langle 0'|s|n'\rangle_n^2 M(\Delta E_n) , \tag{37}
\]

\[
\bar{M} \equiv M(\Delta E) . \tag{38}
\]

We recall that the nucleon g-factors are \(g^p = 1\), \(g^p = 5.586\) for proton and \(g^n = 0\), \(g^n = -3.826\) for neutron. The effective nucleon charge is modified by the recoil effect: \(q = q_p \equiv eN/A\) for proton and \(q = q_n \equiv -eZ/A\) for neutron.

**E. Matrix elements of the effective Hamiltonian for some heavy atoms**

In this section, we present the results of numerical calculation of the matrix element \(\langle 35\rangle\) for different heavy atoms of experimental interest including \(^{138}\text{Ba}\), \(^{206}\text{Pb}\), \(^{208}\text{Pb}\), \(^{172}\text{Yb}\), \(^{174}\text{Yb}\), \(^{176}\text{Yb}\), \(^{178}\text{Hf}\), \(^{180}\text{Hf}\), \(^{226}\text{Ra}\), \(^{232}\text{Th}\). These atoms, as parts of various paramagnetic molecules, have been considered or are proposed for consideration in recent and future eEDM experiments.

The expression \(\langle 34\rangle\) depends on different nuclear matrix elements and corresponding energies of nuclear transitions. In particular, \(|0'|s|n'\rangle_p\) and \(|0'|s|n'\rangle_n\) are matrix elements for nuclear spin-flip proton and neutron transitions with energies \(\Delta E_p\) and \(\Delta E_n\), respectively. These matrix elements and energies may be estimated within the Nilsson nuclear model \(\langle 16\rangle\) which takes into account single-particle excitations only. This model allows one to estimate also the expectation value of the \(1\cdot s\) operator for proton \((1\cdot s)_p\) and neutron \((1\cdot s)_n\) states. The details of calculation of these matrix elements and the corresponding energies are given in Appendix A see Tables IV and V. These tables contain also the energies of giant dipole resonance \(\Delta E\) which enter in the last term in Eq. (35).

The sum over the intermediate electronic states in Eq. (35) is taken into account with the function \(\langle 34\rangle\) which should be evaluated for each nuclear energy. This function involves electronic bound states \(|s_{1/2}\rangle\) and \(|p_{1/2}\rangle\), as well as intermediate excited electronic states \(|n\rangle\). For simplicity, the intermediate electronic states are restricted to the continuum because the states in the discrete spectrum may be shown to give negligible contributions (see, e.g., \(\langle 14\rangle\), \(\langle 15\rangle\)).

Note that the operators in the matrix elements in Eq. (35) are short-range because the function \(\langle 34\rangle\) falls off as \(1/R^2\) outside the nucleus. Therefore, these matrix elements receive their main contributions from the region \(0 < R \ll a_B/Z^{1/3}\), where \(a_B\) is the Bohr radius. In this region, the inter-electron interaction and screening are negligible as compared with the electron-nucleus Coulomb interaction. Therefore, the states \(|s_{1/2}\rangle\) and \(|p_{1/2}\rangle\) may be described by the unscreened Dirac-Coulomb wave functions which are appropriately regu-
larized inside the nucleus; see Appendix B for further details.

The intermediate electronic states \(|n⟩⟩\) in Eq. (34) are given by the Dirac-Coulomb wave functions in the continuous spectrum (see Appendix B.2). Using these wave functions, we calculate numerically the radial integrals in the matrix elements (34) for each particular nuclear energy (see Appendix B.3). The results of these calculations are collected in Table VI.

Using the values of the nuclear matrix elements from Tables IV and V and the values of the electronic matrix elements from Table VI we find the matrix element (35) for various atoms,

\[
⟨p_{1/2}|H_{\text{eff}}|s_{1/2}⟩ = 2c_{s_{1/2}c_{p_{1/2}}/A_B}(\tilde{λ}_pξ_p + \tilde{λ}_nξ_n),
\]

TABLE I: Results of numerical calculations of coefficients \(\tilde{λ}_p\) and \(\tilde{λ}_n\) which specify the leading-order dependence of the matrix element (39) on \(P,T\)-odd coupling constants \(ξ_p\) and \(ξ_n\).

| \(148\)Ba | \(206\)Pb | \(208\)Pb | \(172\)Yb | \(174\)Yb | \(176\)Yb | \(178\)Yb | \(180\)Hf | \(208\)Ra | \(212\)Th |
|---|---|---|---|---|---|---|---|---|---|
| \(λ_p/100\) | 0.65 | 7.0 | 7.1 | 3.6 | 3.6 | 5.4 | 5.4 | 11 | 15 |
| \(λ_n/100\) | 0.68 | 5.8 | 4.8 | 2.4 | 2.9 | 2.3 | 2.6 | 3.2 | 6.7 | 11 |

III. COMPARISON WITH THE CONTACT CP-ODD ELECTRON-NUCLEON INTERACTION

In this section, we will compare the matrix element (39) with that of the contact interaction (2). This will allow us to determine the dependence of the coupling constant \(C_{SP}\) on the \(P,T\)-odd nuclear force coupling constants \(ξ_p\) and \(ξ_n\). Then, employing the experimental constraint (1), we will determine the limits on \(ξ_p\) and \(ξ_n\) originating from the EDM experiments with paramagnetic atoms and molecules.

A. Limits on \(P,T\)-odd nuclear interaction couplings

In an atom, the contact interaction (2) yields the following interaction Hamiltonian between a valence electron and a nucleus (13):

\[
H_{\text{cont}} = \frac{iG_F}{\sqrt{2}} AC_{SP}r_0^7γ_5ρ(\mathbf{R}),
\]

where \(γ_0\) and \(γ_5\) are the Dirac matrices and \(ρ(\mathbf{R})\) is the normalized nuclear charge density. The matrix element of this operator with the \(s_{1/2}\) and \(p_{1/2}\) states was calculated where \(c_{s_{1/2}}\) and \(c_{p_{1/2}}\) are the normalization coefficients of the wave functions (12) and

\[
\tilde{λ}_p = \frac{A-Z}{A} [(ε_p^s - ε_p^l)M_p + (g_p^s - g_p^l)\vec{M}\cdot \vec{s}_p], \quad (40)
\]

\[
\tilde{λ}_n = \frac{Z}{A} [(g_n^s - g_n^l)M_n + (g_n^s - g_n^l)\vec{M}\cdot \vec{s}_n]. \quad (41)
\]

Numerical values of these coefficients are given in Table I. Equation (39) represents one of the main results on this paper as it specifies the leading-order dependence of the atomic EDM (20) on the \(P,T\)-odd coupling constants \(ξ_p\) and \(ξ_n\).

in Ref. (11),

\[
⟨p_{1/2}|H_{\text{cont}}|s_{1/2}⟩ = -c_{s_{1/2}}c_{p_{1/2}}/A_B \frac{G_F C_{SP}}{10\sqrt{2}\pi} \times \left[ 1 + 4\gamma \frac{AZ\alpha}{\Gamma(2\gamma + 1)^2} \frac{2ZR_0}{a_B} \right]^{2\gamma},
\]

where \(\gamma = \sqrt{1 - Z^2α^2}\) is the relativistic factor.

Let us now compare the matrix elements (39) and (13). Setting \(⟨p_{1/2}|H_{\text{cont}}|s_{1/2}⟩ = ⟨p_{1/2}|H_{\text{eff}}|s_{1/2}⟩\) allows us to find the leading-order dependence of the contact interaction constant \(C_{SP}\) on the \(P,T\)-odd nuclear interaction couplings \(ξ_p\) and \(ξ_n\),

\[
C_{SP} = (λ_pξ_p + λ_nξ_n) \times 10^{13}\text{cm}^{-1},
\]

where the dimensionless coefficients \(λ_{p,n}\) are

\[
λ_{p,n} = -\frac{eμ_0}{a_B} \frac{20\sqrt{2}\pi}{G_F} \frac{1 + 4\gamma}{(2\gamma + 1)^2} \frac{ZR_0}{AZ\alpha} \frac{a_B}{2ZR_0}^{2\gamma} \tilde{λ}_{p,n} \times 10^{-13}\text{cm} .
\]

The numerical values of these coefficients may be found from the corresponding values for \(λ_{p,n}\) listed in Table I. We present them in Table I below.

The relation (44) may be used to derive limits on the couplings \(ξ_{p,n}\) which follow from the experimental constraints (1), yielding

\[
|ξ_p| < 2.2 \times 10^{-23}\text{cm}, \quad |ξ_n| < 3.0 \times 10^{-23}\text{cm} .
\]
Similar limits on $\xi_{p,n}$ obtained from the $^{180}$Hf$^{14+}$ experiment \cite{20} are about an order of magnitude weaker, $|\xi_p| < 2.6 \times 10^{-22}$cm, $|\xi_n| < 4.5 \times 10^{-22}$cm.

To summarize, we have presented a mean to relate the experimentally measured quantity $C_{SP}$ with the phenomenological parameters of the CP-odd nuclear interaction. We now proceed to express this relation in terms of the coupling constants of more fundamental CP-odd nuclear forces.

**B. Relation between $C_{SP}$ and CP-odd pion-nucleon coupling constants**

The tree-level pion exchange in known to give dominant contribution to the CP-odd internucleon interaction \cite{3}. The authors of Refs. \cite{13, 21} established the leading-order dependence of the constants $\xi_p$ and $\xi_n$ on the CP-odd pion-nucleon couplings $\tilde{g}_{\pi NN}^{(0)}$, $\tilde{g}_{\pi NN}^{(1)}$ and $\tilde{g}_{\pi NN}^{(2)}$ as

$$\xi_p = -\xi_n = 10^{-14} g(\tilde{g}_{\pi NN}^{(1)} - 0.2\tilde{g}_{\pi NN}^{(0)}) \text{cm},$$

where $g \approx 13.6$ is the strong interaction constant. Substituting this relation into Eq. (43) allows us to find the leading-order relation between the constant $C_{SP}$ and CP-violating pion-nucleon couplings

$$C_{SP} = \lambda_0 \tilde{g}_{\pi NN}^{(0)} + \lambda_1 \tilde{g}_{\pi NN}^{(1)} + \lambda_2 \tilde{g}_{\pi NN}^{(2)},$$

where the numerical values of the coefficients $\lambda_0 = -0.272(\lambda_p - \lambda_n)$, $\lambda_1 = 1.36(\lambda_p - \lambda_n)$ and $\lambda_2 = 0.544(\lambda_p - \lambda_n)$ are collected in Table II for various paramagnetic atoms.

**C. Relation between $C_{SP}$ and quark chromo-EDM**

In this subsection, we consider the chromo-EDM of up and down quarks denoted by $\tilde{d}_u$ and $\tilde{d}_d$, respectively. Assuming that these quantities are the only sources of CP-violating internucleon forces, the authors of Refs. \cite{21, 24} established the following relations:

$$g\tilde{g}_{\pi NN}^{(0)} = 0.8 \times 10^{15} (\tilde{d}_u + \tilde{d}_d) \text{cm}^{-1}, \quad (49a)$$

$$g\tilde{g}_{\pi NN}^{(1)} = 4.0 \times 10^{15} (\tilde{d}_u - \tilde{d}_d) \text{cm}^{-1}. \quad (49b)$$

We substitute these relations into Eqs. (47) and (48) and ignore the last term $\propto \tilde{g}_{\pi NN}^{(2)}$ because its relation to the quark chromo-EDMs is not known. As a result, we find the leading-order dependence of $C_{SP}$ on $\tilde{d}_u$ and $\tilde{d}_d$,

$$C_{SP} = (\lambda_u \tilde{d}_u + \lambda_d \tilde{d}_d) \times 10^{14} \text{cm}^{-1}. \quad (50)$$

Numerical values of the coefficients $\lambda_u = 3.84(\lambda_p - \lambda_n)$ and $\lambda_d = -4.16(\lambda_p - \lambda_n)$ are given in Table II below.

**D. Relation between $C_{SP}$ and QCD vacuum angle**

The pion-nucleon coupling constants $\tilde{g}_{\pi NN}^{(0)}$ and $\tilde{g}_{\pi NN}^{(1)}$ may be expressed via the QCD vacuum angle $\tilde{\theta}$ as (see, e.g., Refs. \cite{8, 21, 26})

$$\tilde{g}_{\pi NN}^{(0)} = -15.5 \times 10^{-3} \tilde{\theta}, \quad (51a)$$

$$\tilde{g}_{\pi NN}^{(1)} = 3.4 \times 10^{-3} \tilde{\theta}. \quad (51b)$$

Substituting these relations into Eq. (48), we may represent $C_{SP}$ in terms of $\tilde{\theta}$ as

$$C_{SP} = \lambda_0 \tilde{\theta} \times 10^{-2} \tilde{\theta}, \quad (52)$$

where the value of the constant $\lambda_0 = 0.88(\lambda_p - \lambda_n)$ is given in Table II for different atoms. In particular, with the use of the corresponding value for $^{232}$Th, the experimental constraint on $C_{SP}$ \cite{11} implies

$$|\tilde{\theta}| < 9 \times 10^{-8}. \quad (53)$$

This constraint is almost three orders of magnitude weaker than the currently accepted one \cite{27} which is based on the neutron EDM \cite{28} and Hg EDM \cite{29} experiments. However, it is comparable to the constraint on QCD vacuum angle originating from the $^{129}$Xe EDM experiments \cite{30, 51} and from constraints on the nucleon EDM \cite{10, 11} derived from the experiments with paramagnetic molecules. This demonstrates the importance of contributions from the $P, T$-odd nuclear forces to the atomic EDM. For reference, we collect the limits on $\xi_{p,n}$, $\tilde{g}_{\pi NN}$, $\tilde{d}_{u,d}$ and $\tilde{\theta}$ in Table III.

\[ \text{IV. CONCLUSIONS} \]

In this paper, we demonstrated that the eEDM experiments with paramagnetic molecules are also sensitive to the $P, T$-violating nuclear forces, as well as to other sources of CP violation in the hadronic sector. We considered $P, T$-violating internucleon interaction described by the Hamiltonian \cite{11} with coupling constants $\xi_{p,n}$. We established the leading-order relation \cite{11} between these couplings and the constant $C_{SP}$ of the contact semilep-
table II: The results of numerical computations of λ-coefficients in Eqs. (14)-(18) and (52).

| Spherical          | λ₁ | λ₂ | λ₃ | λ₄ | λ₅ | λ₆ |
|--------------------|----|----|----|----|----|----|
| ¹³⁵Ba              | -4.3 | -0.53 | 0.26 | 0.11 | 0.74 | -0.80 |
| ²⁰⁶Pb              | -5.4 | -2.8 | 0.17 | -0.83 | -0.33 | -2.3 |
| ²⁰⁶Pb              | -5.4 | -2.8 | 0.29 | -1.5 | -0.58 | -4.1 |

| Deformed           | dµ | dπ | θ   |
|--------------------|----|----|-----|
| ¹⁷²Yb              | -6.9 | -4.0 | -0.77 |
| ¹⁷²Yb              | -6.9 | -4.0 | -2.9 |
| ¹⁷²Hf              | -6.9 | -4.0 | -2.9 |
| ²³⁰Ra              | -2.9 | -1.8 | 0.29 |
| ²³⁴Th              | -3.3 | -2.4 | 0.25 |

TABLE III: Limits on $\xi_{\mu,n}$, $g_{\pi NN}^{(0,1,2)}$, $\tilde{d}_{u,d}$ and θ obtained from the ThO limit on $|C_{SP}| < 7.3 \times 10^{-10}$.

It is pertinent to make some comments about the accuracy of our results. All our results are based on numerical calculations of the coefficients listed in Table II. These coefficients involve both nuclear and electronic matrix elements as well as nuclear excitation energies. The electronic matrix elements in Eq. (34) contain radial integrals which are calculated numerically with some details given in Appendix A. To estimate the accuracy of our numerical integration methods, we calculated similar radial integrals which are responsible for atomic energy level shifts (contributions to the Lamb shifts) due to nuclear polarizability and compared these shifts with calculated earlier values in Refs. 17, 18. This allows us to conclude that the error in numerical calculation of electronic matrix elements does not exceed 5%. However, the main source of uncertainty is represented by nuclear matrix elements and nuclear excitation energies. In this paper, we use the single-particle nuclear shell model to estimate M₁ spin-flip matrix elements and corresponding energies in heavy nuclei listed in Appendix A. Using these quantities we calculated reduced transition probabilities for M₁ spin-flip transitions for some nuclei (ytterbium and thorium). The results have been compared with analogous quantities calculated using sophisticated many-body methods in Ref. 32. On the basis of this comparison we estimate the error in nuclear transition energies and matrix elements to be under 45-50%.

To conclude, we stress that the EDM-like experiments with molecules in paramagnetic state are sensitive to hadronic $CP$-violating parameters such as $CP$-odd pion-nucleon couplings, quark-chromo EDMs and QCD vacuum angle. We expect that results from the next generation of these experiments will significantly improve the limits on these parameters.

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Appendix A: Nuclear energies and matrix elements

These appendices follow our calculation in Ref. [11] and are presented here to provide the reader with the details facilitating the understanding of the current paper. In the first appendix we estimate the matrix elements and corresponding energies of the nuclear M1 spin-flip single-particle transitions. The details of these computations slightly differ for (nearly) spherical and deformed nuclei. Therefore, we consider these two cases separately.

1. Spherical nuclei

In this section, we focus on the $^{208}$Pb, $^{206}$Pb and $^{138}$Ba nuclei, which are nearly spherical, i.e., they have deformation $\delta < 0.1$. For these nuclei, proton and neutron single-particle states may be labeled as $|n, l, j, m\rangle$, where $n$ is the oscillator quantum number, $l$ and $j$ are the orbital and total momentum numbers, $m$ is magnetic quantum number. In this basis, the nuclear spin operator $s$ provides transitions between fine structure doublets.

In the $^{208}$Pb nucleus, the non-vanishing matrix elements of the spin operator are $\langle 5h_\frac{9}{2}|s|5h_\frac{1}{2}\rangle$ for protons and $\langle 6i_\frac{1}{2}|s|6i_\frac{3}{2}\rangle$ for neutrons. The isotope $^{206}$Pb receives additional contributions from the $\langle 5p_\frac{1}{2}|s|5p_\frac{3}{2}\rangle$ neutron matrix elements. For $^{138}$Ba, non-vanishing proton contributions arise from the matrix elements $\langle 4d_\frac{3}{2}|s|4d_\frac{5}{2}\rangle$ and $\langle 4g_\frac{5}{2}|s|4g_\frac{7}{2}\rangle$ whereas neutron contributions come from $\langle 5h_\frac{9}{2}|s|5h_\frac{1}{2}\rangle$. All these matrix elements may be calculated using the properties of spherical spinors (see, e.g., Ref. [33]). The energies of all these transitions may be estimated with the use of Fig. 5 in Ref. [11]. When the energies are (nearly) degenerate, we give the sum of matrix elements corresponding to the same energy. In the Table [V] below, we collect the values of such matrix elements with the corresponding energies for $^{208}$Pb, $^{206}$Pb and $^{138}$Ba. The values for the nuclear radii $R_0$ and the energy of giant dipole resonance are calculated according to the empirical formulas:

$$R_0 = 1.2A^{1/3} \text{fm},$$

$$\Delta E = 95A^{-1/3}(1 - A^{1/3}) \text{MeV}. \tag{A1}$$

$$M(\Delta E_{n'}) = \frac{\alpha}{c_{s1/2}c_{p1/2}} \sum_n \frac{\langle p_{1/2}|f(R)\tilde{R}_0|n\rangle \langle n|f(R)(\tilde{R} \times \alpha)|s_{1/2}\rangle}{\Delta E_n + \text{sgn}(E_n)\Delta E_{n'}} + (s_{1/2} \leftrightarrow p_{1/2}) \tag{B1}$$

For further computation of the matrix elements in Eq. (B1) the electron wave functions need to be specified.

2. Deformed nuclei

For deformed heavy nuclei with $\delta > 0.1$, it is convenient to use the Nilsson basis [16, 34], wherein proton and neutron single-particle states are labeled with $|n_3, n_\perp, \Lambda, \Omega\rangle$, where $n_3$ and $n_\perp$ are the oscillator quantum numbers, $\Lambda$ and $\Omega$ are the projections of angular and total momenta on the deformation axis. Note that $\Omega = \Lambda + \Sigma$ where $\Sigma$ is the projection of the nucleon’s spin on the deformation axis. The dependence of the energy levels on the deformation parameter $\delta$ in this model may be inferred from Fig. 5 in Ref. [11]. From such dependence, one may estimate the energies of the spin-flip transitions. Note that in the basis $|n_3, n_\perp, \Lambda, \Omega\rangle$, each M1 spin-flip matrix element is $\langle m'|s_+|0'\rangle = 1$, and the corresponding energy level is doubly degenerate since each quantum number $\Sigma$ corresponds to $\pm \Lambda$.

The single-nucleon spin-flip transition energies $\Delta E_{n'}$, the energies of giant dipole resonance $\Delta E$, the deformation parameters $\delta$ and the nuclear radii $R_0$ for several nuclei of interest are presented in Table [V].

Appendix B: Evaluation of electronic matrix elements

In this appendix, we provide the details for the numerical calculation of the electronic matrix element (33). For convenience, we use the spherical basis $|e_+e_-e_0\rangle$. The components of the vectors in this basis will be labeled by the $(+, -, 0)$ subscripts. Due to spherical symmetry, Eq. (34) may be rewritten in terms of the ‘0’-component of the operators $\tilde{R}$ and $\tilde{R} \times \alpha$.

1. The $s_{1/2}$ and $p_{1/2}$ wave functions

The valence electron $s_{1/2}$ and $p_{1/2}$ wave functions may be expressed in terms of the spherical spinors $\Omega^s_\mu(\tilde{R})$
where $\mu$ is the magnetic quantum number and $\kappa = (l - j)(2j + 1)$ as

$$|s_{1/2}⟩ = c_{s_{1/2}} \left( f_{s_{1/2}}(R)\Omega^{-1}_\mu(R) \right), \quad |p_{1/2}⟩ = c_{p_{1/2}} \left( f_{p_{1/2}}(R)\Omega^1_\mu(R) \right),$$

where the radial wave functions $f_{s_{1/2}}$ and $g_{s_{1/2}}$ are well approximated in the region $R_0 < R < a_B Z^{1/3}$ by the Bessel functions of the first kind $J_\nu(x)$ (see, e.g., [12]).

Note that the wave functions [13] are the zero-energy solutions of the Dirac-Coulomb equations for a point-like nucleus. For an extended nucleus, the corresponding solution is complicated. At the current level of accuracy, it suffices to use Eqs. [13] as an approximation to the wave functions. For the region inside the nucleus, $0 \leq R \leq R_0$, the radial wave functions $f_{s_{1/2}}$ and $g_{s_{1/2}}$ may be continued as follows

$$f_{s_{1/2}}(R) = \frac{(1 + \gamma)J_{2\gamma}(x_0) - \frac{\pi}{2}J_{2\gamma-1}(x_0)}{R_0}, \quad g_{s_{1/2}}(R) = \frac{R_0 Z_\alpha J_{2\gamma}(x_0)}{x_0},$$

where $x_0 = \sqrt{8ZR/a_B}$.

Note that these functions are the approximate solutions (containing only leading terms at small distance) of the Dirac equation inside the nucleus with constant density.

2. Excited electronic states of the continuous spectrum

The excited electronic states $|n⟩$ in the continuous spectrum may be labeled by the quantum number $\kappa = (l - j)(2j + 1)$ and the energy $E$, $|n⟩ \equiv |En⟩$. In spherical coordinates, these functions read (see, e.g., Refs. [17, 18, 19]):

$$|n⟩ \equiv |E\kappa⟩ = \left( \begin{array}{c} f^E_\kappa(R)p_{1/2}^E(R) \\ ig^E_\kappa(R)\Omega^{-1}_\mu(R) \end{array} \right),$$

with

$$f^E_\kappa(R) = \frac{(2pR)^\gamma e^{\pi y/2} [\Gamma(\gamma + iy)] \sqrt{|E + m_e|}}{R\gamma^{\pi y/2}(2\gamma + 1)} \times \text{Re}[e^{-ipR+i\eta}F_j(\gamma + 1 + iy, 2\gamma + 1, 2ipR)],$$

$$g^E_\kappa(R) = -\text{sgn}(E)(2pR)^\gamma e^{\pi y/2} [\Gamma(\gamma + iy)] \sqrt{|E - m_e|} \times \text{Im}[e^{-ipR+i\eta}F_j(\gamma + 1 + iy, 2\gamma + 1, 2ipR)].$$

Here $p = \sqrt{E^2 - m_e^2}$ is the electron’s momentum, $y = Z\alpha E/p$, $e^{\eta} = \sqrt{-\kappa - i\eta/m_e}$ and $F_j(a, b, z)$ is the confluent hypergeometric function of the first kind. Note that the wave functions [13] are normalized as $|E\kappa|E\kappa⟩ = \delta(E - E\kappa)$.

The functions [16] solve for the Dirac equation with a point-like nucleus. Therefore, we will only use them for outside of the nucleus, $R > R_0$. For the inside of the nucleus, $0 \leq R \leq R_0$, we will consider the following continuation of these functions

$$f^E_\kappa(R) = b_1 R^l, \quad g^E_\kappa(R) = b_2 R^l,$$

where $l = |\kappa + 1/2| - 1/2$ is the orbital angular momentum corresponding to $\kappa$, $l = |-\kappa + 1/2| - 1/2$ is the orbital angular momentum corresponding to $-\kappa$. The values of the coefficients $b_1$ and $b_2$ are determined by matching Eqs. [15] and [17] on the boundary of the nucleus. The wave functions [17] are, to the leading order, solutions to the Dirac equation inside a nucleus of a constant density.

We stress that the extension of the electronic wave functions to the inside region of the nucleus [17] is an approximation which is acceptable at our level of accuracy. We checked the validity of this approximation by computing the Lamb shift in heavy atoms due to nuclear polarizability. Within this approximation, we have 95% agreement with the exact results presented in Refs. [17, 18, 19].

3. Results of calculation of electronic matrix element

Substituting the wave functions [12] and [15] into Eq. [11] and performing the integration over angular variables, we obtain

$$M(\Delta E) = \frac{2\alpha}{9} \int_{-\infty}^{\infty} T(E) dE,$$

where

$$T(E) = \frac{R^2_0(E)R^2_0(E) - R^{-2}_0(E)R^{-2}_0(E)}{-S^{-1}_p(E)S^{-1}_p(E) + S^{2}_s(E)S^{2}_s(E)}.$$
and the radial integrals $R^\kappa_{s,p}(E)$ and $S^\kappa_{s,p}(E)$ are defined by

\begin{align}
R^\kappa_s(E) & \equiv \int_0^\infty (f_{s_1/2} f_{s}^E + g_{s_1/2} g_{s}^E) f(R) R^2 dR, \\
R^\kappa_p(E) & \equiv \int_0^\infty (f_{p_1/2} g_{s}^E + g_{p_1/2} f_{s}^E) f(R) R^2 dR, \\
S^\kappa_s(E) & \equiv \int_0^\infty (f_{s_1/2} g_{s}^E + g_{s_1/2} f_{s}^E) f(R) R^2 dR, \\
S^\kappa_p(E) & \equiv \int_0^\infty (f_{p_1/2} f_{s}^E + g_{p_1/2} g_{s}^E) f(R) R^2 dR.
\end{align}

Here the radial function $f(R)$ is given by Eq. (13). Note that Eq. (B9) involves only the terms with $\kappa = \pm 1, \pm 2$ which are allowed by the selection rules for transitions from $s_{1/2}$ and $p_{1/2}$ bound electron states.

With the radial wave functions (B3), (B4), (B6) and (B7), the radial integrals (B10) may be computed numerically for any specific electron energy $E$ and nuclear energy $\Delta E_n'$ or $\Delta \bar{E}$. For all values of $\Delta E_n'$ and $\Delta \bar{E}$ presented in Appendix A numerical analysis showed that for $|E| > 500 m_e$, $T(E)/(E_{s_{1/2}} - E \pm \Delta E_n')$ is effectively zero, so the energy integrals in Eqs. (B5) may be cut off at $|E| \approx 500 m_e$. We also point out that the dominant contributions to the energy integrals (B5) come from the region where $E \sim 50m_e$, which is larger than the values of $\Delta E_n'$ or $\Delta \bar{E}$ considered in Appendix A. As a result, $M(\Delta E_n')$ is a slowly varying function of energy.

The energy integrals in Eqs. (B5) are computed numerically, giving $M(\Delta E_n')$ for all values of $\Delta E_n'$ and $\Delta \bar{E}$ presented in Appendix A. The resulting numerical values of the electronic factors $M_n'$, $M_n$ and $M(\Delta \bar{E})$ are presented in Table VI below.

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| Proton transitions | Neutron transitions | \( \Delta E_{n'} \) (MeV) | \( \langle l \cdot s \rangle_p \) | \( \langle l \cdot s \rangle_n \) | \( R_0 \) (fm) | \( \Delta \bar{E} \) (MeV) | \( \delta \) |
|-------------------|-------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| \( |(n'|s|0')_p|^2 \) | \( |(n'|s|0')_n|^2 \) | \( |(n'|s|0')_p|^2 \) | \( |(n'|s|0')_n|^2 \) | \( |(n'|s|0')_p|^2 \) | \( |(n'|s|0')_n|^2 \) | \( |(n'|s|0')_p|^2 \) | \( |(n'|s|0')_n|^2 \) |
| \( 18/25 \) | 2.7 | 170/121 | 5.3 | 7 | 15 | 6.20 | 14.8 | 0.09 |
| \( 2/25 \) | 4.1 | 200/121 | 5.4 | | | | | |
| \( 28/81 \) | 4.3 | 30/121 | 5.5 | | | | | |
| \( 56/81 \) | 4.4 | 136/121 | 5.9 | | | | | |
| \( 16/81 \) | 4.5 | 56/121 | 6.0 | | | | | |
| \( 8/9 \) | 4.6 | 60/121 | 6.2 | | | | | |
| \( 8/81 \) | 5.2 | 8/121 | 6.5 | | | | | |

| Proton transitions | Neutron transitions | \( \Delta E_{n'} \) (MeV) | \( \langle l \cdot s \rangle_p \) | \( \langle l \cdot s \rangle_n \) | \( R_0 \) (fm) | \( \Delta \bar{E} \) (MeV) | \( \delta \) |
|-------------------|-------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| \( |(n'|s|0')_p|^2 \) | \( |(n'|s|0')_n|^2 \) | \( |(n'|s|0')_p|^2 \) | \( |(n'|s|0')_n|^2 \) | \( |(n'|s|0')_p|^2 \) | \( |(n'|s|0')_n|^2 \) | \( |(n'|s|0')_p|^2 \) | \( |(n'|s|0')_n|^2 \) |
| \( 10/11 \) | 4.5 | 72/169 | 6.1 | 15 | 22 | 7.09 | 13.3 | 0.03 |
| \( 162/121 \) | 4.6 | 462/169 | 6.2 | | | | | |
| \( 98/121 \) | 4.7 | 318/169 | 6.3 | | | | | |
| \( 250/121 \) | 4.8 | 132/169 | 6.4 | | | | | |
| \( 32/121 \) | 5.0 | 100/169 | 6.5 | | | | | |
| \( 8/121 \) | 5.1 | 6/169 | 6.7 | 2/169 | 6.9 | | | |
| | | 2/3 | 1.4 | 10/9 | 2.0 | | | |

TABLE IV: Nuclear radii \( R_0 \), deformation parameters \( \delta \), nucleon spin-orbit expectation values \( \langle l \cdot s \rangle_{p,n} \), E1 giant resonance energies \( \Delta \bar{E} \) and matrix elements \( |(n'|s|0')_{p,n}|^2 \) and energies \( \Delta E_{n'} \) of M1 spin-flip transitions in some spherical nuclei of interest.
| Proton transitions | Neutron transitions | (1·s)_p | (1·s)_n | R₀ (fm) | ∆E (MeV) | δ |
|-------------------|-------------------|---------|---------|---------|----------|---|
| ⁷²⁷Yb | | | | | | |
| ⁷³¹Yb | | | | | | |
| ⁷³³Yb | | | | | | |
| ⁷⁷⁸Hf | | | | | | |
| ⁷⁹⁰Hf | | | | | | |
| ⁷⁷⁹Ra | | | | | | |
| ²²⁶Ra | | | | | | |
| ²³²Th | | | | | | |

**Table V:** Nuclear radii R₀, deformation parameters δ, nucleon spin-orbit expectation values ⟨1·s⟩_p, n, E1 giant resonance energies ∆E and matrix elements |⟨n'|s|0⟩⟩_p, n|^2 and energies ∆E_{n',p} of M1 spin-flip transitions in some deformed nuclei of interest.

| Spherical | Deformed |
|-----------|----------|
| ¹³⁸Ba | ²⁰⁶Pb | ²⁰⁸Pb | ⁷²⁷Yb | ⁷²⁷⁷Yb | ¹⁷⁸⁷Yb | ¹⁷⁸⁹Hf | ¹⁷⁸⁹Hf | ²²⁶Ra | ²³²Th |
| M_p/a_M | M_n/a_M | M/M | M/a_M |
| 11.1 | 94.1 | 94.0 | 69.3 | 69.3 | 69.2 | 121 | 121 | 156 | 244 |
| 16.5 | 143 | 95.7 | 83.4 | 106 | 88.7 | 96.8 | 114 | 196 | 385 |
| 1.83 | 10.7 | 10.7 | 4.83 | 4.83 | 4.74 | 5.53 | 5.54 | 16.0 | 18.3 |

**Table VI:** Numerical values for the electronic matrix elements M_p, M_n and M for several atoms of interest.