Effects of CP -violating internucleon interactions in paramagnetic molecules

V. V. Flambaum^{1,2}, I. B. Samsonov,¹ and H. B. Tran Tan^{1*}

¹*School of Physics, University of New South Wales, Sydney 2052, Australia*

²*Helmholtz Institute Mainz, Johannes Gutenberg University, 55099 Mainz, Germany*

(Received 22 September 2020; accepted 9 December 2020; published 29 December 2020)

We demonstrate that electron electric dipole moment (eEDM) experiments with molecules in a 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.

DOI: [10.1103/PhysRevD.102.115036](https://doi.org/10.1103/PhysRevD.102.115036)

I. INTRODUCTION

The existence of nonvanishing electric dipole moments (EDMs) of elementary particles was conjectured nearly 70 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 nonvanishing 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 past 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. [8,9] for reviews). As was demonstrated in the recent works [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 [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 open-shell nucleon interacts with a heavy nuclear core through a nuclear potential $U(\mathbf{r})$. The P , T -violating nuclear forces are taken into account by the phenomenological interaction Hamiltonian $H_{\text{odd}} = \xi \boldsymbol{\sigma} \cdot \nabla U(\mathbf{r})$, where $\boldsymbol{\sigma}/2$ is the nucleon spin operator and ξ is the coupling constant of the dimension of length. This coupling constant will be denoted by ξ_p for proton and ξ_n for neutron. The authors of Ref. [13] demonstrated that the leading contributions to this interaction arise due to the π meson exchange between the open-shell nucleon and the nuclear core. In general, however, this interaction may arise due to the CP -violating πNN , ηNN , 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_{odd} regardless of the underlying fundamental interaction. In principle, experimental limits on the phenomenological parameters $\xi_{p,n}$ may be converted into limits on the parameters of more fundamental hadronic interactions. We stress that the interaction H_{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}ThO molecule. Based on theoretical calculations [15,16], this experiment also placed a limit on the CP -odd electron-nucleon interaction coupling constant (90% C.L.),

$$|C_{SP}|_{\text{Th}} < 7.3 \times 10^{-10}. \quad (1)$$

*tran.tan@student.unsw.edu.au

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI. Funded by SCOAP³.

In a heavy, spinless 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

$$\mathcal{L} = \frac{iG_F}{\sqrt{2}} C_{SP}^p \bar{e} \gamma_5 e \bar{p} p + \frac{iG_F}{\sqrt{2}} C_{SP}^n \bar{e} \gamma_5 e \bar{n} n, \quad (2)$$

where G_F is the Fermi coupling constant and 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.¹

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 ξ_p and ξ_n , $C_{SP} = C_{SP}(\xi_p, \xi_n)$. This will allow us to find limits on these couplings originating from the experimental constraint (1). Then, using known relations between the constants ξ_p , ξ_n and more fundamental CP -violating hadronic parameters, we will establish a leading-order relation between C_{SP} and CP -odd pion-nucleon couplings $\bar{g}_{\pi NN}^{(0,1,2)}$, quark-chromo EDMs $\bar{d}_{u,d}$, and QCD vacuum angle $\bar{\theta}$.

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 2 [12,13,17]. 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, the current limit on the QCD vacuum angle reads $|\bar{\theta}| < 10^{-10}$; see, e.g., reviews [8,9] and updated limits presented in Ref. [18].

The rest of the paper is organized as follows. In Sec. II, we present an estimate for the atomic EDM arising from the CP -odd nuclear forces. In Sec. 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 Sec. IV we give a summary of our results and provide some comments on assumptions and precision. Technical details of calculations

¹In general, there are also nucleon-pseudoscalar and electron-scalar (PS) as well as tensor-tensor (TT) CP -violating semileptonic interactions. These interactions play a subleading role in atoms with spinless nuclei and, therefore, they will be ignored in this paper.

of electronic and nuclear matrix elements are collected in Appendixes A and B.

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 Sec. 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 Sec. 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 from both the Standard Model and beyond. Independently of their nature, in the nonrelativistic limit, these short-range interactions may be taken into account by the following phenomenological single-particle Hamiltonian [13]:

$$H_{\text{odd}} = \frac{G_F}{\sqrt{2}} \frac{\eta}{2m_p} \boldsymbol{\sigma} \cdot \nabla \rho(\mathbf{r}), \quad (3)$$

where $\boldsymbol{\sigma}/2$ is the spin of the open-shell nonrelativistic nucleon, $\rho(\mathbf{r})$ is the density of the nuclear core, η is the effective coupling constant, and m_p is the proton mass.

In a heavy nucleus, the nuclear core creates an effective nuclear potential $U(\mathbf{r})$ in which the open-shell nucleon moves. In the short-range approximation, this potential is proportional to the density of the nuclear core, $\rho(\mathbf{r}) = U(\mathbf{r})\rho(0)/U(0)$. Taking this into account, the Hamiltonian (3) may be represented as

$$H_{\text{odd}} = \xi \boldsymbol{\sigma} \cdot \nabla U(\mathbf{r}), \quad (4)$$

where [13]

$$\xi = \eta \frac{G_F}{2\sqrt{2}m_p} \frac{\rho(0)}{U(0)} \approx -2 \times 10^{-21} \eta \cdot \text{cm}. \quad (5)$$

The total potential for the open-shell nucleon is thus given by

$$\tilde{U} = U + H_{\text{odd}} = U + \xi \boldsymbol{\sigma} \cdot \nabla U \approx U(\mathbf{r} + \xi \boldsymbol{\sigma}). \quad (6)$$

Now let $\psi_{n'}(\mathbf{r})$ be an unperturbed single-nucleon wave function labeled by some quantum numbers n' . This function is supposed to solve for the Schrödinger equation with the potential $U(\mathbf{r})$. Equation (6) suggests that the wave function perturbed by the P , T -odd interaction (3) may be represented as

$$\tilde{\psi}_{n'}(\mathbf{r}) = \psi_{n'}(\mathbf{r} + \xi\boldsymbol{\sigma}) \approx (1 + \xi\boldsymbol{\sigma} \cdot \nabla)\psi_{n'}(\mathbf{r}), \quad (7)$$

or, more generally, generalizing to all nucleons

$$\tilde{\psi}_{n'}(\mathbf{r}_1, \dots, \mathbf{r}_A) = \left(1 + \sum_{\tau=1}^A \xi_{\tau} \boldsymbol{\sigma}_{\tau} \cdot \nabla_{\tau}\right) \psi_{n'}(\mathbf{r}_1, \dots, \mathbf{r}_A), \quad (8)$$

where $\tilde{\psi}_{n'}(\mathbf{r}_1, \dots, \mathbf{r}_A)$ and $\psi_{n'}(\mathbf{r}_1, \dots, \mathbf{r}_A)$ are multiparticle nuclear wave functions. Note that the constants ξ_{τ} are different for proton ($\xi_{\tau} = \xi_p$) and neutron ($\xi_{\tau} = \xi_n$).

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

$$\langle \tilde{m}' | \mathcal{O} | \tilde{n}' \rangle = \langle m' | \mathcal{O} | n' \rangle - \sum_{\tau=1}^A \xi_{\tau} \langle m' | [\boldsymbol{\sigma}_{\tau} \cdot \nabla_{\tau}, \mathcal{O}] | n' \rangle, \quad (9)$$

where $|n'\rangle$ and $|\tilde{n}'\rangle$ are the kets corresponding to the wave functions $\psi_{n'}(\mathbf{r}_1, \dots, \mathbf{r}_A)$ and $\tilde{\psi}_{n'}(\mathbf{r}_1, \dots, \mathbf{r}_A)$, respectively.

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 $\mathbf{R}$ interacting with nucleons of charge q_{τ} located at the point $\mathbf{r}_{\tau}$. The interaction Hamiltonian considered in this paper is a combination of the electric (q) and magnetic (μ) terms,

$$H_{\text{int}} = - \sum_{\tau=1}^A (H_{\tau}^q + H_{\tau}^{\mu}), \quad (10a)$$

$$H_{\tau}^q = \frac{eq_{\tau}}{|\mathbf{R} - \mathbf{r}_{\tau}|}, \quad (10b)$$

$$H_{\tau}^{\mu} = \frac{e\boldsymbol{\mu}_{\tau} \cdot [(\mathbf{R} - \mathbf{r}_{\tau}) \times \boldsymbol{\alpha}]}{|\mathbf{R} - \mathbf{r}_{\tau}|^3}, \quad (10c)$$

where

$$\boldsymbol{\mu}_{\tau} = \mu_0 (g_{\tau}^l \mathbf{l}_{\tau} + g_{\tau}^s \mathbf{s}_{\tau}) \quad (11)$$

is the nucleon's magnetic moment operator, μ_0 is the nuclear magneton, and g_{τ}^l and g_{τ}^s are the orbital and spin g factors of the nucleon. Note that $\mathbf{s}_{\tau} = \boldsymbol{\sigma}_{\tau}/2$ is the nucleon spin operator while $\boldsymbol{\alpha} = \begin{pmatrix} 0 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & 0 \end{pmatrix}$ are the Dirac matrices acting on the electron's states. We point out that since we consider spinless nuclei in this paper, the interaction (10a) does not arise if the nucleus is treated as a pointlike object.

The interaction Hamiltonian (10) as a function of $\mathbf{R}$ and $\mathbf{r}_{\tau}$ may be expanded into a multipole series. In particular,

the leading terms in the expansion of the electric interaction Hamiltonian (10b) are

$$H_{\tau}^q = eq_{\tau} \left(\frac{1}{R} + \frac{\mathbf{R} \cdot \mathbf{r}_{\tau}}{R^3} \right) \theta(R - r_{\tau}) + eq_{\tau} \left(\frac{1}{r} + \frac{\mathbf{R} \cdot \mathbf{r}_{\tau}}{r^3} \right) \theta(r_{\tau} - R) + \dots, \quad (12)$$

where the ellipsis stands for terms with higher multipolarity. In this expansion, the term $\theta(R - r_{\tau})/R + \theta(r_{\tau} - R)/r_{\tau}$, after averaging with the nuclear charge density $\rho(\mathbf{r})$, 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 $(\mathbf{R} \cdot \mathbf{r}_{\tau})[\theta(R - r_{\tau})/R^3 + \theta(r_{\tau} - R)/r_{\tau}^3]$ in Eq. (12) is the leading dipole one on which we will focus our attention.² 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, $\rho(r) = 3\theta(R_0 - r)/R_0^3$. Since $\rho(r)$ is only needed for the calculation of electronic matrix elements, where a dominant contribution comes from outside the nucleus, this approximation does not greatly affect our results. Indeed, a computation which uses a more realistic Woods-Saxon model for the nuclear density only gives a difference of about 3%. The constant density approximation has the advantage of giving analytical form for the regularized potentials. In fact, it was used in the calculation of Lamb shift due to nuclear polarizability [19–22] and hyperfine structure which gave perfect agreement with experiments.

Using this prescription, one may obtain the following continuation of the $1/R^2$ function to a small distance:

$$f(R) \equiv \int_0^{\infty} \rho(r) \left[\frac{1}{R^2} \theta(R - r) + \frac{1}{r^2} \theta(r - R) \right] r^2 dr = \frac{1}{R^2} \theta(R - R_0) + \frac{3R_0 - 2R}{R_0^3} \theta(R_0 - R), \quad (13)$$

with which the regularized dipole interaction operator (12) takes the form

$$\bar{H}_{\tau}^q \equiv eq_{\tau} (\hat{\mathbf{R}} \cdot \mathbf{r}_{\tau}) f(R), \quad (14)$$

where $\hat{\mathbf{R}} \equiv \mathbf{R}/R$.

²This expansion is needed to describe the interaction between the nucleus and the electron. The contribution inside the nucleus is relatively small, the dominant contribution comes from the region outside the nucleus. This justifies the use of the dipole term for the electronic variables.

The magnetic interaction operator (10c) 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 (13),

$$\bar{H}_\tau^\mu \equiv e\boldsymbol{\mu}_\tau \cdot (\hat{\mathbf{R}} \times \boldsymbol{\alpha})f(R). \quad (15)$$

Thus, the dipole part of the interaction (10a) which is regularized at short distances reads

$$\bar{H}_{\text{int}} = - \sum_{\tau=1}^A (\bar{H}_\tau^q + \bar{H}_\tau^\mu), \quad (16)$$

with $\bar{H}_\tau^q$ and $\bar{H}_\tau^\mu$ 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 $\boldsymbol{\sigma}_\tau \cdot \nabla_\tau$ with the interaction Hamiltonians (14) and (15),

$$[\boldsymbol{\sigma}_\tau \cdot \nabla_\tau, \bar{H}_\tau^q] = eq_\tau (\boldsymbol{\sigma}_\tau \cdot \hat{\mathbf{R}})f(R), \quad (17a)$$

$$[\boldsymbol{\sigma}_\tau \cdot \nabla_\tau, \bar{H}_\tau^\mu] = ie\mu_0(g_\tau^s - g_\tau^l)f(R) \times (\hat{\mathbf{R}} \times \boldsymbol{\alpha}) \cdot (\boldsymbol{\sigma}_\tau \times \nabla_\tau). \quad (17b)$$

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

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

The unperturbed atomic states will be denoted by $|nn'\rangle = |n\rangle|n'\rangle$, 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 prime. As in Sec. II, the P , T -perturbed nuclear wave functions (8) are denoted as $|\tilde{n}'\rangle$. Electronic and nuclear ground states are denoted as $|0\rangle$ and $|\tilde{0}'\rangle$ (or $|0'\rangle$ for an unperturbed state), respectively.

The atomic EDM arising from the mixed interaction H_{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,

$$\sum_{n \neq 0} \frac{\langle 0| -e\mathbf{R}|n\rangle \langle n\tilde{0}'|\bar{H}_{\text{int}}|\tilde{0}'0\rangle}{E_0 - E_n} = 0. \quad (18)$$

Indeed, it may be shown that $\langle \tilde{0}'|\bar{H}_{\text{int}}|\tilde{0}'\rangle \propto \langle 0'|\mathbf{s}|0'\rangle = 0$, where $\mathbf{s} \equiv \sum_{\tau=1}^A \mathbf{s}_\tau$ is the nuclear spin operator.

The leading nonvanishing contributions to the atomic EDM thus arise in the second-order perturbation theory,

$$\begin{aligned} \mathbf{d} = & 2 \sum_{m \neq 0, n\tilde{n}' \neq 0\tilde{0}'} \frac{\langle 0| -e\mathbf{R}|m\rangle \langle m\tilde{0}'|\bar{H}_{\text{int}}|\tilde{n}'n\rangle \langle n\tilde{n}'|\bar{H}_{\text{int}}|\tilde{0}'0\rangle}{(E_m - E_0)[E_n - E_0 + \text{sgn}(E_n)(E_{\tilde{n}'} - E_{\tilde{0}'})]} \\ & + \sum_{m \neq 0, n\tilde{n}' \neq 0\tilde{0}'} \frac{\langle 0\tilde{0}'|\bar{H}_{\text{int}}|\tilde{n}'n\rangle \langle n| -e\mathbf{R}|m\rangle \langle m\tilde{n}'|\bar{H}_{\text{int}}|\tilde{0}'0\rangle}{[E_n - E_0 + \text{sgn}(E_n)(E_{\tilde{n}'} - E_{\tilde{0}'})][E_m - E_0 + \text{sgn}(E_m)(E_{\tilde{n}'} - E_{\tilde{0}'})]}, \end{aligned} \quad (19)$$

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 the 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 a 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 $|0\rangle = |s_{1/2}\rangle$ 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

$$\mathbf{d} \approx 2 \frac{\langle s_{1/2}|e\mathbf{R}|p_{1/2}\rangle \langle p_{1/2}|H_{\text{eff}}|s_{1/2}\rangle}{E_{p_{1/2}} - E_{s_{1/2}}}, \quad (20)$$

where we have introduced the effective interaction Hamiltonian

$$H_{\text{eff}} \equiv \sum_{n\tilde{n}' \neq 0\tilde{0}'} \frac{|m\rangle \langle m\tilde{0}'|\bar{H}_{\text{int}}|\tilde{n}'n\rangle \langle n\tilde{n}'|\bar{H}_{\text{int}}|\tilde{0}'0\rangle \langle 0|}{\Delta E_n + \text{sgn}(E_n)\Delta E_{\tilde{n}'}}. \quad (21)$$

Here $\Delta E_n = E_n - E_0$ and $\Delta E_{\tilde{n}'} = E_{\tilde{n}'} - E_{\tilde{0}'}$. Our goal is to calculate the matrix element $\langle p_{1/2}|H_{\text{eff}}|s_{1/2}\rangle$ which is responsible for the leading-order contributions to the atomic EDM.

Using the identity (9), one may single out the part of the matrix element of the effective Hamiltonian (21) which is linear in the P , T -odd nuclear interaction coupling ξ ,

$$\langle p_{1/2}|H_{\text{eff}}|s_{1/2}\rangle = - \sum_{\tau=1}^A \sum_{nn' \neq 00'} \frac{\langle p_{1/2}0'|[\xi_\tau \boldsymbol{\sigma}_\tau \cdot \nabla_\tau, \bar{H}_{\text{int}}]|n'n\rangle \langle nn'|\bar{H}_{\text{int}}|0's_{1/2}\rangle}{\Delta E_n + \text{sgn}(E_n)\Delta E_{n'}} + (s_{1/2} \leftrightarrow p_{1/2}). \quad (22)$$

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

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

where

$$\mathcal{M}_{nn'}^{1\tau} \equiv \langle p_{1/2} | O' [\boldsymbol{\sigma}_{\tau} \cdot \nabla_{\tau}, \bar{H}_{\tau}^q] | n' n \rangle \langle n n' | \bar{H}_{\tau}^{\mu} | 0 s_{1/2} \rangle, \quad (24a)$$

$$\mathcal{M}_{nn'}^{2\tau} \equiv \langle p_{1/2} | O' [\boldsymbol{\sigma}_{\tau} \cdot \nabla_{\tau}, \bar{H}_{\mu}] | n' n \rangle \langle n n' | \bar{H}_{\tau}^q | 0 s_{1/2} \rangle. \quad (24b)$$

These matrix elements will be calculated in the next subsection.

D. Calculation of matrix elements

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

$$\begin{aligned} \mathcal{M}_{nn'}^{1\tau} &= e^2 q_{\tau} \langle 0' | \boldsymbol{\sigma}_{\tau} | n' \rangle \langle n' | \boldsymbol{\mu}_{\tau} | 0' \rangle \\ &\times \langle p_{1/2} | f(R) \hat{\mathbf{R}} | n \rangle \langle n | f(R) \hat{\mathbf{R}} \times \boldsymbol{\alpha} | s_{1/2} \rangle. \end{aligned} \quad (25)$$

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

$$\langle 0' | \sigma_{\tau}^i | n' \rangle \langle n' | \mu_{\tau}^j | 0' \rangle = \frac{1}{3} \delta_{ij} \langle 0' | \sigma_{\tau} | n' \rangle \langle n' | \mu_{\tau} | 0' \rangle + \dots, \quad (26)$$

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

$$\langle 0' | \sigma_{\tau} | n' \rangle \langle n' | \mu_{\tau} | 0' \rangle = 2\mu_0 (g_{\tau}^s - \epsilon g_{\tau}^l) |\langle 0' | s_{\tau} | n' \rangle|^2, \quad (27)$$

where 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

$$\begin{aligned} \mathcal{M}_{nn'}^{1\tau} &= \frac{2}{3} \mu_0 (g_{\tau}^s - \epsilon g_{\tau}^l) e^2 q_{\tau} |\langle 0' | s_{\tau} | n' \rangle|^2 \\ &\times \langle p_{1/2} | f(R) \hat{\mathbf{R}} | n \rangle \langle n | f(R) \hat{\mathbf{R}} \times \boldsymbol{\alpha} | s_{1/2} \rangle. \end{aligned} \quad (28)$$

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

$$\begin{aligned} \mathcal{M}_{nn'}^{2\tau} &= \frac{i}{3} e^2 q_{\tau} \mu_0 (g_{\tau}^s - g_{\tau}^l) \langle 0' | \boldsymbol{\sigma}_{\tau} \times \nabla_{\tau} | n' \rangle \langle n' | \mathbf{r}_{\tau} | 0' \rangle \\ &\times \langle p_{1/2} | f(R) \hat{\mathbf{R}} \times \boldsymbol{\alpha} | n \rangle \langle n | f(R) \hat{\mathbf{R}} | s_{1/2} \rangle. \end{aligned} \quad (29)$$

Here $\langle n' | \mathbf{r}_{\tau} | 0' \rangle$ 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 $\langle n' | \mathbf{r}_{\tau} | 0' \rangle$ which constitute the giant electric dipole resonance with the excitation energy $\Delta \bar{E}$. Then having fixed the nuclear energy in the denominator of Eq. (23), one might use the completeness relation for the nuclear states, $|n'\rangle\langle n'| = 1$, to reduce the nuclear matrix elements in Eq. (29) to the expectation value of the $\mathbf{l}_{\tau} \cdot \mathbf{s}_{\tau}$ operator,

$$\langle 0' | \boldsymbol{\sigma}_{\tau} \times \nabla_{\tau} | n' \rangle \langle n' | \mathbf{r}_{\tau} | 0' \rangle \approx -2i \langle 0' | \mathbf{l}_{\tau} \cdot \mathbf{s}_{\tau} | 0' \rangle \equiv -2i \langle \mathbf{l}_{\tau} \cdot \mathbf{s}_{\tau} \rangle. \quad (30)$$

With this expression for the nuclear matrix element, Eq. (29) simplifies to

$$\begin{aligned} \mathcal{M}_{nn'}^{2\tau} &\approx \frac{2}{3} e^2 q_{\tau} \mu_0 (g_{\tau}^s - g_{\tau}^l) \langle 0' | \mathbf{l}_{\tau} \cdot \mathbf{s}_{\tau} | 0' \rangle \\ &\times \langle p_{1/2} | f(R) \hat{\mathbf{R}} \times \boldsymbol{\alpha} | n \rangle \langle n | f(R) \hat{\mathbf{R}} | s_{1/2} \rangle. \end{aligned} \quad (31)$$

Substituting Eqs. (28) and (31) into Eq. (23), one may represent the matrix element of the effective operator (21) in the compact form

$$\langle p_{1/2} | H_{\text{eff}} | s_{1/2} \rangle = 2 \sum_{\tau=1}^A \xi_{\tau} q_{\tau} \mu_0 \left[\sum_{n'} (g_{\tau}^s - \epsilon g_{\tau}^l) M(E_{n'}) |\langle 0' | s_{\tau} | n' \rangle|^2 + (g_{\tau}^s - g_{\tau}^l) M(\Delta \bar{E}) \langle 0' | \mathbf{l}_{\tau} \cdot \mathbf{s}_{\tau} | 0' \rangle \right], \quad (32)$$

where

$$\begin{aligned} M(E) &\equiv \frac{e^2}{3} \sum_n \frac{\langle p_{1/2} | f(R) \hat{\mathbf{R}} | n \rangle \langle n | f(R) \hat{\mathbf{R}} \times \boldsymbol{\alpha} | s_{1/2} \rangle}{\Delta E_n + \text{sgn}(E_n) E} \\ &+ (s_{1/2} \leftrightarrow p_{1/2}). \end{aligned} \quad (33)$$

Note that the sum in Eq. (32) contains only single-particle nucleon excitations. It is instructive to separate

proton (p) and neutron (n) contributions with nuclear excitation energies denoted by ΔE_p and ΔE_n as

$$\begin{aligned} \langle p_{1/2} | H_{\text{eff}} | s_{1/2} \rangle &= 2\mu_0 \sum_{i=p,n} \xi_i q_i [(g_i^s - \epsilon g_i^l) M_i + (g_i^s - g_i^l) \bar{M} \langle \mathbf{l} \cdot \mathbf{s} \rangle_i], \end{aligned} \quad (34)$$

where

$$M_p \equiv \sum_{\Delta E_p} |\langle 0' | \mathbf{s} | n' \rangle_p|^2 M(\Delta E_p), \quad (35)$$

$$M_n \equiv \sum_{\Delta E_n} |\langle 0' | \mathbf{s} | n' \rangle_n|^2 M(\Delta E_n), \quad (36)$$

$$\bar{M} \equiv M(\Delta \bar{E}). \quad (37)$$

We recall that for a free nucleon, the g factors are $g_p^l = 1$, $g_p^s = 5.586$ for proton, and $g_n^l = 0$, $g_n^s = -3.826$ for neutron. In a nuclear medium, the g factors should be reduced by about a factor of 0.7. 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 (34) for different heavy atoms of experimental interest including ^{138}Ba , ^{206}Pb , ^{208}Pb , ^{172}Yb , ^{174}Yb , ^{176}Yb , ^{178}Hf , ^{180}Hf , ^{226}Ra , ^{232}Th . These atoms, as parts of various paramagnetic molecules, have been considered or are proposed for consideration in recent and future electron electric dipole moment (eEDM) experiments.

The expression (34) depends on different nuclear matrix elements and corresponding energies of nuclear transitions. In particular, $\langle 0' | \mathbf{s} | n' \rangle_p$ and $\langle 0' | \mathbf{s} | n' \rangle_n$ are matrix elements for nuclear spin-flip proton and neutron transitions with energies ΔE_p and ΔE_n , respectively. Since the nuclear operators we use are single-particle operators, these matrix elements and energies may be estimated within the Nilsson nuclear model [23] which takes into account single-particle excitations only. This model allows one to estimate also the expectation value of the $\mathbf{l} \cdot \mathbf{s}$ operator for proton $\langle \mathbf{l} \cdot \mathbf{s} \rangle_p$ and neutron $\langle \mathbf{l} \cdot \mathbf{s} \rangle_n$ states. The details of calculation of these matrix elements and the corresponding energies are given in Appendix A (see Tables V and VI). These tables contain also the energies of giant dipole resonance $\Delta \bar{E}$ which enter in the last term in Eq. (34).

The sum over the intermediate electronic states in Eq. (34) is taken into account with the function (33) 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 $|n\rangle$, which include

those from the discrete spectrum and both negative and positive continua.

Owing to the fact that the function $f(R)$ peaks at the surface of the nucleus where the overlap between different bound states is small, matrix elements that involve intermediate bound states are typically orders of magnitude smaller than those coming from intermediate continuum states. The nuclear energy $\Delta E_{n'}$ (in the MeV range) in the denominator of Eq. (23) provides further suppression of the contribution from discrete states. As a result, in the computation below, the intermediate electronic states are restricted to the continual spectra as in Refs. [19,21]. This is in contrast with the situation in Ref. [24], where contributions of the discrete states are significantly enhanced due to small electronic energy denominators (eV range).

We also point out that one can use Coulomb electron wave functions for the intermediate continuum states since the function $f(R)$ peaks at the surface of the nucleus where the electron screening of the Coulomb nuclear potential is negligible. We tested the accuracy of our numerical calculation of the electronic matrix elements by computing the Lamb shift in heavy atoms due to nuclear polarizability, which have similar matrix elements, and obtained agreement within 5% of known results [19–21].

Note that the operators in the matrix elements in Eq. (34) are short range because the function (13) 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 interelectron 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 regularized inside the nucleus; see Appendix B 1 for further details.

The intermediate electronic states $|n\rangle$ in Eq. (33) 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 (33) for each particular nuclear energy (see Appendix B 3). The results of these calculations are collected in Table I.

We observe that the values of $M_{p,n}$ and $\bar{M}$ generally increase with increasing nuclear charge Z . This is due to the fact that the electronic matrix element (33) receives the most contribution in the region near the nucleus, as can be

TABLE I. Numerical values for the electronic matrix elements M_p , M_n , and $\bar{M}$ for several atoms of interest.

	Spherical			Deformed						
	^{138}Ba	^{206}Pb	^{208}Pb	^{172}Yb	^{174}Yb	^{176}Yb	^{178}Hf	^{180}Hf	^{226}Ra	^{232}Th
M_p/a_B	11.1	94.1	94.0	69.3	69.3	69.2	121	121	156	244
M_n/a_B	16.5	143	95.7	83.4	106	88.7	96.8	114	196	385
$\tilde{M}/a_B$	1.83	10.7	10.7	4.83	4.83	4.74	5.53	5.54	16.0	18.3

TABLE II. Results of numerical calculations of coefficients $\tilde{\lambda}_p$ and $\tilde{\lambda}_n$ which specify the leading-order dependence of the matrix element (38) on P , T -odd coupling constants ξ_p and ξ_n .

	¹³⁸ Ba	²⁰⁶ Pb	²⁰⁸ Pb	¹⁷² Yb	¹⁷⁴ Yb	¹⁷⁶ Yb	¹⁷⁸ Hf	¹⁸⁰ Hf	²²⁶ Ra	²³² Th
$\tilde{\lambda}_p/100$	0.65	7.0	7.1	3.6	3.6	3.6	5.4	5.4	11	15
$\tilde{\lambda}_n/100$	0.68	5.8	4.8	2.4	2.9	2.3	2.6	3.2	6.7	11

seen from the behavior of the function $f(R)$ in Eq. (13). For large Z , $Z\alpha \sim 1$ and the electronic Coulomb wave function near the nucleus is significantly enhanced due to relativistic effects, thus making $M(E)$ large.

Using the values of the nuclear matrix elements from Tables V and VI, and the values of the electronic matrix elements from Table I, we find the matrix element (34) for various atoms,

$$\langle p_{1/2} | H_{\text{eff}} | s_{1/2} \rangle = 2c_{s_{1/2}} c_{p_{1/2}} \frac{e\mu_0}{a_B} (\tilde{\lambda}_p \xi_p + \tilde{\lambda}_n \xi_n), \quad (38)$$

where $c_{s_{1/2}}$ and $c_{p_{1/2}}$ are the normalization coefficients of the wave functions (B2) and

$$\tilde{\lambda}_p = \frac{A-Z}{A} [(g_p^s - \epsilon g_p^l) M_p + (g_p^s - g_p^l) \tilde{M} \langle \mathbf{l} \cdot \mathbf{s} \rangle_p], \quad (39)$$

$$\tilde{\lambda}_n = -\frac{Z}{A} [(g_n^s - \epsilon g_n^l) M_n + (g_n^s - g_n^l) \tilde{M} \langle \mathbf{l} \cdot \mathbf{s} \rangle_n]. \quad (40)$$

Numerical values of these coefficients are given in Table II. Equation (38) 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 .

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

In this section, we will compare the matrix element (38) 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 [25]:

$$H_{\text{cont}} = \frac{iG_F}{\sqrt{2}} A C_{SP} \gamma_0 \gamma_5 \rho(\mathbf{R}), \quad (41)$$

where γ_0 and γ_5 are the Dirac matrices and $\rho(\mathbf{R})$ is the normalized nuclear charge density. The matrix element of

this operator with the $s_{1/2}$ and $p_{1/2}$ states was calculated in Ref. [11],

$$\begin{aligned} \langle p_{1/2} | H_{\text{cont}} | s_{1/2} \rangle &= -c_{s_{1/2}} c_{p_{1/2}} \frac{G_F C_{SP}}{10\sqrt{2}\pi} \\ &\times \frac{1+4\gamma}{\Gamma(2\gamma+1)^2} \frac{AZ\alpha}{R_0^2} \left(\frac{2ZR_0}{a_B} \right)^{2\gamma}, \end{aligned} \quad (42)$$

where $\gamma = \sqrt{1-Z^2\alpha^2}$ is the relativistic factor.

We stress that at the atomic level the operator (41) describes a general contact CP -odd interaction which may originate from different fundamental CP -violating sources, such as the direct electron-nucleus interaction due to exchange by a heavy intermediate boson, nucleon EDM, QCD vacuum angle, and P , T -odd nuclear forces. Although contributions from different sources to the atomic EDM may partly cancel each other, complete “accidental cancellation” does not seem to be an attractive hypothesis since the scales of such interactions are different in any reasonable model. Therefore, in this paper, we use the traditional approach in EDM studies based on the “single-source assumption” and consider a contribution to the coupling constant C_{SP} of the interaction (41) originating from the CP -violating internucleon interaction with the Hamiltonian (3). In principle, different sources of CP violation may be disentangled by measurements on several systems with significantly different sensitivities to distinct CP -violating parameters (see, e.g., Ref. [24]).

The goal of this section is to determine the leading-order dependence of the contact interaction constant C_{SP} on the P , T -odd nuclear interaction couplings ξ_p and ξ_n . This relation appears upon equating the matrix elements (38) and (42), $\langle p_{1/2} | H_{\text{cont}} | s_{1/2} \rangle = \langle p_{1/2} | H_{\text{eff}} | s_{1/2} \rangle$. As a result, we find

$$C_{SP} = (\lambda_p \xi_p + \lambda_n \xi_n) \times 10^{13} \text{ cm}^{-1}, \quad (43)$$

where the dimensionless coefficients $\lambda_{p,n}$ are

$$\begin{aligned} \lambda_{p,n} &= -\frac{e\mu_0}{a_B} \frac{20\sqrt{2}\pi \Gamma(2\gamma+1)^2}{G_F} \frac{1}{1+4\gamma} \\ &\times \frac{R_0^2}{AZ\alpha} \left(\frac{a_B}{2ZR_0} \right)^{2\gamma} \tilde{\lambda}_{p,n} \times 10^{-13} \text{ cm}. \end{aligned} \quad (44)$$

The numerical values of these coefficients may be found from the corresponding values for $\tilde{\lambda}_{p,n}$ listed in Table II. We present them in Table III below.

TABLE III. The results of numerical computations of λ coefficients in Eqs. (43), (47), (49), and (51).

		λ_p	λ_n	λ_0	λ_1	λ_2	λ_u	λ_d	λ_θ
Spherical	$^{138}_{56}\text{Ba}$	-4.1	-4.3	-0.053	0.26	0.11	0.74	-0.80	0.17
	$^{206}_{82}\text{Pb}$	-3.4	-2.8	0.17	-0.83	-0.33	-2.3	2.5	-0.54
	$^{208}_{82}\text{Pb}$	-3.4	-2.3	0.29	-1.5	-0.58	-4.1	4.5	-0.95
Deformed	$^{172}_{70}\text{Yb}$	-5.6	-3.7	0.50	-2.5	-1.0	-7.1	7.7	-1.6
	$^{174}_{70}\text{Yb}$	-5.6	-4.5	0.31	-1.6	-0.62	-4.4	4.8	-1.0
	$^{176}_{70}\text{Yb}$	-5.5	-3.5	0.56	-2.8	-1.1	-7.9	8.6	-1.8
	$^{178}_{72}\text{Hf}$	-3.9	-2.2	0.47	-2.4	-0.94	-6.7	7.2	-1.5
	$^{180}_{72}\text{Hf}$	-6.9	-4.0	0.77	-3.8	-1.5	-11	-12	-2.5
	$^{226}_{88}\text{Ra}$	-2.9	-1.8	0.29	-1.4	0.58	-4.1	4.4	-0.94
	$^{232}_{90}\text{Th}$	-3.3	-2.4	0.25	-1.2	-0.50	-3.5	3.8	-0.81

The relation (43) may be used to derive limits on the couplings $\xi_{p,n}$ which follow from the experimental constraints (1), yielding

$$|\xi_p| < 2.2 \times 10^{-23} \text{ cm}, \quad |\xi_n| < 3.0 \times 10^{-23} \text{ cm}. \quad (45)$$

Similar limits on $\xi_{p,n}$ obtained from the $^{180}\text{Hf}F^+$ experiment [26] are about an order of magnitude weaker, $|\xi_p| < 2.6 \times 10^{-22} \text{ cm}$, $|\xi_n| < 4.5 \times 10^{-22} \text{ cm}$.

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 is known to give a dominant contribution to the CP -odd internucleon interaction (3). The authors of Refs. [13,27] established the leading-order dependence of the constants ξ_p and ξ_n on the CP -odd pion-nucleon couplings $\bar{g}_{\pi NN}^{(0)}$, $\bar{g}_{\pi NN}^{(1)}$, and $\bar{g}_{\pi NN}^{(2)}$ as

$$\xi_p = -\xi_n = 10^{-14} g (\bar{g}_{\pi NN}^{(1)} + 0.4 \bar{g}_{\pi NN}^{(2)} - 0.2 \bar{g}_{\pi NN}^{(0)}) \text{ cm}, \quad (46)$$

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

$$C_{SP} = \lambda_0 \bar{g}_{\pi NN}^{(0)} + \lambda_1 \bar{g}_{\pi NN}^{(1)} + \lambda_2 \bar{g}_{\pi NN}^{(2)}, \quad (47)$$

where the numerical values of the coefficients $\lambda_0 = -0.272(\lambda_p - \lambda_n)$, $\lambda_1 = 1.36(\lambda_p - \lambda_n)$, and $\lambda_2 = 0.544 \times (\lambda_p - \lambda_n)$ are collected in Table III 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. [27–30] established the following relations:

$$g\bar{g}_{\pi NN}^{(0)} = 0.8 \times 10^{15} (\tilde{d}_u + \tilde{d}_d) \text{ cm}^{-1}, \quad (48a)$$

$$g\bar{g}_{\pi NN}^{(1)} = 4.0 \times 10^{15} (\tilde{d}_u - \tilde{d}_d) \text{ cm}^{-1}. \quad (48b)$$

We substitute these relations into Eqs. (46) and (47) and ignore the last term $\propto \bar{g}_{\pi NN}^{(2)}$ 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 $\tilde{d}_p$,

$$C_{SP} = (\lambda_u \tilde{d}_u + \lambda_d \tilde{d}_d) \times 10^{14} \text{ cm}^{-1}. \quad (49)$$

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 III below.

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

The pion-nucleon coupling constants $\bar{g}_{\pi NN}^{(0)}$ and $\bar{g}_{\pi NN}^{(1)}$ may be expressed via the QCD vacuum angle $\bar{\theta}$ as (see, e.g., Refs. [8,30–32])

$$\bar{g}_{\pi NN}^{(0)} = -15.5 \times 10^{-3} \bar{\theta}, \quad (50a)$$

$$\bar{g}_{\pi NN}^{(1)} = 3.4 \times 10^{-3} \bar{\theta}. \quad (50b)$$

Substituting these relations into Eq. (47), we may represent C_{SP} in terms of $\bar{\theta}$ as

$$C_{SP} = \lambda_\theta \times 10^{-2} \bar{\theta}, \quad (51)$$

where the value of the constant $\lambda_\theta = 0.88(\lambda_p - \lambda_n)$ is given in Table III for different atoms. In particular, with the use of the corresponding value for ^{232}Th , the experimental constraint on C_{SP} (1) implies

TABLE IV. Limits on $\xi_{p,n}$, $\bar{g}_{\pi NN}^{(0,1,2)}$, $\tilde{d}_{u,d}$, and $\bar{\theta}$ obtained from the ThO limit on $|C_{SP}| < 7.3 \times 10^{-10}$.

$\frac{ \xi_p }{10^{-23} \text{ cm}}$	$\frac{ \xi_n }{10^{-23} \text{ cm}}$	$\frac{\bar{g}_{\pi NN}^{(0)}}{10^{-9}}$	$\frac{\bar{g}_{\pi NN}^{(1)}}{10^{-9}}$	$\frac{\bar{g}_{\pi NN}^{(2)}}{10^{-9}}$	$\frac{\tilde{d}_u}{10^{-24} \text{ cm}}$	$\frac{\tilde{d}_d}{10^{-24} \text{ cm}}$	$\frac{\bar{\theta}}{10^{-8}}$
2.2	3.0	2.9	0.6	1.5	2.1	1.9	9

$$|\bar{\theta}| < 9 \times 10^{-8}. \quad (52)$$

This constraint is almost 3 orders of magnitude weaker than the currently accepted one [33] which is based on the neutron EDM [34] and Hg EDM [35] experiments. However, it is comparable to the constraint on the QCD vacuum angle originating from the ^{129}Xe EDM experiments [36,37] and from constraints on the nucleon EDM [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 $\xi_{p,n}$, $\bar{g}_{\pi NN}^{(0,1,2)}$, $\tilde{d}_{u,d}$, and $\bar{\theta}$ in Table IV.

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 (4) with coupling constants $\xi_{p,n}$. We established the leading-order relation (43) between these couplings and the constant C_{SP} of the contact semileptonic interaction (2). This relation contains atom-specific coefficients $\lambda_{p,n}$ calculated numerically and presented in Table III. We used this relation to place independent limits (45) on the coupling constants $\xi_{p,n}$ originating from the experimental constraint (1).

The P , T -odd nuclear interaction is described by the phenomenological Hamiltonian which may originate from different fundamental interactions. In particular, it is known [13] that this interaction may appear due to the π meson exchange between nucleons. In this case, the parameters $\xi_{p,n}$ may be expressed via the CP -odd pion-nucleon couplings as in Eq. (46). Then, using the known relations between the pion couplings and the quark-chromo EDMs (48) as well as QCD $\bar{\theta}$ angle (50) we relate the parameters $\xi_{p,n}$ to $\tilde{d}_{u,d}$ and to $\bar{\theta}$ as in Eqs. (49) and (51), respectively. This allows us to place limits on $\tilde{d}_{u,d}$ and $\bar{\theta}$ originating from the experimental constraint (1).

We stress that the limits on $\bar{g}_{\pi NN}^{(0,1,2)}$, $\tilde{d}_{u,d}$, and $\bar{\theta}$ obtained in this paper are independent from similar constraints established in our recent paper [11] because the latter were found by taking into account nucleon EDM while in this paper we consider the P , T -odd nuclear force as the origin for these relations. It is useful to compare these two results. For example, the limit on the CP -odd pion-nucleon coupling

constant $\bar{g}_{\pi NN}^{(2)}$ is obtained in the present paper only, the limits on u -quark chromo-EDM $\tilde{d}_u$ are approximately the same in both papers while the constraint on $\bar{\theta}$ in Eq. (52) is 6 times weaker than the analogous one found in Ref. [11], but it is comparable to the constraint on the QCD vacuum angle originating from the ^{129}Xe EDM experiment [36,37]. Of course, all these constraints on $\bar{\theta}$ are not competitive with currently accepted strongest limit $|\bar{\theta}| < 10^{-10}$ [33], obtained on the neutron EDM [34] and Hg EDM [35] experiments (see also discussion in Ref. [18]).

It is pertinent to make some comments about the accuracy of our results. All our results are based on numerical calculations of the coefficients (39) 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. (33) contain radial integrals which are calculated numerically with some details given in Appendix B 3. 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. [19,21]. This allows us to conclude that the error in the 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. [38]. 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 the 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.

ACKNOWLEDGMENTS

This work was supported by the Australian Research Council Grants No. DP190100974 and No. DP200100150 and the Gutenberg Fellowship.

APPENDIX A: NUCLEAR ENERGIES AND MATRIX ELEMENTS

These appendixes 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

TABLE V. Nuclear radii R_0 , deformation parameters δ , nucleon spin-orbit expectation values $\langle \mathbf{l} \cdot \mathbf{s} \rangle_{p,n}$, E1 giant resonance energies $\Delta \bar{E}$ and reduced matrix elements $|\langle n' || s || 0' \rangle_{p,n}|^2$ and energies $\Delta E_{n'}$ of M1 spin-flip transitions in some spherical nuclei of interest. The pair $[m', m]$ represents the magnetic quantum numbers of the initial and final states of the transitions. For each pair $[m', m]$, there is another pair $[-m', -m]$ with the same transition energy.

	Proton transitions			Neutron transitions			$\langle \mathbf{l} \cdot \mathbf{s} \rangle_p$	$\langle \mathbf{l} \cdot \mathbf{s} \rangle_n$	R_0 [fm]	$\Delta \bar{E}$ [MeV]	δ
	$[m', m]$	$\Delta E_{n'}$ [MeV]	$ \langle n' s 0' \rangle_p ^2$	$[m', m]$	$\Delta E_{n'}$ [MeV]	$ \langle n' s 0' \rangle_n ^2$					
^{138}Ba	$ 4g_{\frac{9}{2}} m'\rangle \rightarrow 4g_{\frac{7}{2}} m\rangle$		160/9	$ 5h_{\frac{11}{2}} m'\rangle \rightarrow 5h_{\frac{9}{2}} m\rangle$		336/13	15	21	7.11	13.3	0.05
	$[\frac{9}{2}, \frac{7}{2}], [\frac{7}{2}, \frac{5}{2}]$	4.4		$[\frac{5}{2}, \frac{3}{2}], [\frac{7}{2}, \frac{5}{2}]$	5.3						
	$[\frac{5}{2}, \frac{3}{2}]$	4.8		$[\frac{9}{2}, \frac{7}{2}], [\frac{11}{2}, \frac{9}{2}]$							
	$[\frac{7}{2}, \frac{5}{2}], [\frac{3}{2}, \frac{1}{2}]$	5.2		$[\frac{3}{2}, \frac{1}{2}]$	5.6						
	$[\frac{5}{2}, \frac{3}{2}]$	5.5		$[\frac{5}{2}, \frac{3}{2}]$	5.7						
	$ 4d_{\frac{5}{2}} m'\rangle \rightarrow 4d_{\frac{3}{2}} m\rangle$		48/5	$[\frac{3}{2}, \frac{1}{2}], [\frac{7}{2}, \frac{5}{2}]$	5.9						
	$[\frac{1}{2}, \frac{1}{2}], [\frac{1}{2}, \frac{-1}{2}]$	2.7		$[\frac{9}{2}, \frac{7}{2}]$							
	$[\frac{1}{2}, \frac{3}{2}]$	4.0		$[\frac{1}{2}, \frac{1}{2}], [\frac{1}{2}, \frac{-1}{2}]$	6.2						
				$[\frac{3}{2}, \frac{1}{2}]$							
				$[\frac{1}{2}, \frac{3}{2}]$	6.3						
				$[\frac{3}{2}, \frac{1}{2}], [\frac{7}{2}, \frac{5}{2}]$	6.4						
^{206}Pb	$ 5h_{\frac{11}{2}} m'\rangle \rightarrow 5h_{\frac{9}{2}} m\rangle$		240/11	$ 6i_{\frac{13}{2}} m'\rangle \rightarrow 6i_{\frac{11}{2}} m\rangle$		336/13	15	22	7.09	13.3	0.03
	$[\frac{11}{2}, \frac{9}{2}]$	4.5		$[\frac{5}{2}, \frac{3}{2}]$	6.1						
	$[\frac{9}{2}, \frac{7}{2}], [\frac{7}{2}, \frac{5}{2}]$	4.6		$[\frac{9}{2}, \frac{7}{2}], [\frac{7}{2}, \frac{5}{2}]$	6.2						
	$[\frac{5}{2}, \frac{3}{2}], [\frac{3}{2}, \frac{1}{2}]$	4.7		$[\frac{3}{2}, \frac{1}{2}], [\frac{1}{2}, \frac{1}{2}]$							
	$[\frac{1}{2}, \frac{-1}{2}], [\frac{1}{2}, \frac{1}{2}]$	4.8		$[\frac{1}{2}, \frac{-1}{2}], [\frac{3}{2}, \frac{3}{2}]$							
	$[\frac{3}{2}, \frac{3}{2}], [\frac{5}{2}, \frac{5}{2}]$			$[\frac{13}{2}, \frac{11}{2}], [\frac{11}{2}, \frac{9}{2}]$	6.3						
	$[\frac{7}{2}, \frac{7}{2}], [\frac{9}{2}, \frac{9}{2}]$			$[\frac{1}{2}, \frac{3}{2}]$							
	$[\frac{1}{2}, \frac{3}{2}], [\frac{3}{2}, \frac{5}{2}]$	5.0		$[\frac{5}{2}, \frac{3}{2}], [\frac{7}{2}, \frac{7}{2}]$	6.4						
	$[\frac{5}{2}, \frac{7}{2}], [\frac{7}{2}, \frac{9}{2}]$	5.1		$[\frac{3}{2}, \frac{3}{2}], [\frac{5}{2}, \frac{7}{2}]$	6.5						
				$[\frac{9}{2}, \frac{9}{2}], [\frac{11}{2}, \frac{11}{2}]$							
				$[\frac{7}{2}, \frac{9}{2}]$	6.7						
				$[\frac{9}{2}, \frac{11}{2}]$	6.9						
				$ 5p_{\frac{3}{2}} m'\rangle \rightarrow 5p_{\frac{1}{2}} m\rangle$	16/3						
				$[\frac{3}{2}, \frac{-1}{2}]$	1.4						
				$[\frac{1}{2}, \frac{1}{2}], [\frac{1}{2}, \frac{-1}{2}]$	2.0						
^{208}Pb	$ 5h_{\frac{11}{2}} m'\rangle \rightarrow 5h_{\frac{9}{2}} m\rangle$		240/11	$ 6i_{\frac{13}{2}} m'\rangle \rightarrow 6i_{\frac{11}{2}} m\rangle$		336/13	15	21	7.11	13.3	0.05
	$[\frac{11}{2}, \frac{9}{2}]$	4.5		$[\frac{5}{2}, \frac{3}{2}]$	6.1						
	$[\frac{9}{2}, \frac{7}{2}], [\frac{7}{2}, \frac{5}{2}]$	4.6		$[\frac{9}{2}, \frac{7}{2}], [\frac{7}{2}, \frac{5}{2}]$	6.2						
	$[\frac{5}{2}, \frac{3}{2}], [\frac{3}{2}, \frac{1}{2}]$	4.7		$[\frac{3}{2}, \frac{1}{2}], [\frac{1}{2}, \frac{1}{2}]$							
	$[\frac{1}{2}, \frac{-1}{2}], [\frac{1}{2}, \frac{1}{2}]$	4.8		$[\frac{1}{2}, \frac{-1}{2}], [\frac{3}{2}, \frac{3}{2}]$							
	$[\frac{3}{2}, \frac{3}{2}], [\frac{5}{2}, \frac{5}{2}]$			$[\frac{13}{2}, \frac{11}{2}], [\frac{11}{2}, \frac{9}{2}]$	6.3						
	$[\frac{7}{2}, \frac{7}{2}], [\frac{9}{2}, \frac{9}{2}]$			$[\frac{1}{2}, \frac{3}{2}]$							
	$[\frac{1}{2}, \frac{3}{2}], [\frac{3}{2}, \frac{5}{2}]$	5.0		$[\frac{5}{2}, \frac{3}{2}], [\frac{7}{2}, \frac{7}{2}]$	6.4						
	$[\frac{5}{2}, \frac{7}{2}], [\frac{7}{2}, \frac{9}{2}]$	5.1		$[\frac{3}{2}, \frac{3}{2}], [\frac{5}{2}, \frac{7}{2}]$	6.5						
				$[\frac{9}{2}, \frac{9}{2}], [\frac{11}{2}, \frac{11}{2}]$							
				$[\frac{7}{2}, \frac{9}{2}]$	6.7						
				$[\frac{9}{2}, \frac{11}{2}]$	6.9						

n is the oscillator quantum number, l and j are the orbital and total momentum numbers, and m is the magnetic quantum number. In this basis, the nuclear spin operator $\mathbf{s} = \sum_{\tau=1}^A \mathbf{s}_{\tau}$ provides transitions between fine structure doublets.

In the ^{208}Pb nucleus, the nonvanishing matrix elements of the spin operator are $\langle 5h \frac{9}{2} m | \mathbf{s} | 5h \frac{11}{2} m' \rangle$ for protons and

$\langle 6i \frac{11}{2} m | \mathbf{s} | 6i \frac{13}{2} m' \rangle$ for neutrons (henceforth, the ground state will be put on the right side of the operator $\mathbf{s}$). The isotope ^{206}Pb receives additional contributions from the $\langle 5p \frac{1}{2} m | \mathbf{s} | 5p \frac{3}{2} m' \rangle$ neutron matrix elements. For ^{138}Ba , nonvanishing proton contributions arise from the matrix elements $\langle 4d \frac{3}{2} m | \mathbf{s} | 4d \frac{5}{2} m' \rangle$ and $\langle 4g \frac{9}{2} m | \mathbf{s} | 4g \frac{7}{2} m' \rangle$, whereas

TABLE VI. Nuclear radii R_0 , deformation parameters δ , nucleon spin-orbit expectation values $\langle \mathbf{l} \cdot \mathbf{s} \rangle_{p,n}$, E1 giant resonance energies $\Delta \bar{E}$ and matrix elements $|\langle n' | \mathbf{s} | 0' \rangle_{p,n}|^2$ and energies $\Delta E_{n'}$ of M1 spin-flip transitions in some deformed nuclei of interest.

	Proton transitions		Neutron transitions		$\langle \mathbf{l} \cdot \mathbf{s} \rangle_p$	$\langle \mathbf{l} \cdot \mathbf{s} \rangle_n$	R_0 [fm]	$\Delta \bar{E}$ [MeV]	δ
	Transition	$\Delta E_{n'}$ [MeV]	Transition	$\Delta E_{n'}$ [MeV]					
^{172}Yb	$ 523 \frac{7}{2}\rangle \rightarrow \frac{5}{2}\rangle$	4.5	$ 651 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	3.9	10	15	6.67	14.0	0.31
	$ 532 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	4.0	$ 642 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	4.5					
	$ 541 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	4.5	$ 633 \frac{7}{2}\rangle \rightarrow \frac{5}{2}\rangle$	5.0					
	$ 404 \frac{9}{2}\rangle \rightarrow \frac{7}{2}\rangle$	4.1	$ 505 \frac{11}{2}\rangle \rightarrow \frac{9}{2}\rangle$	5.1					
^{174}Yb	$ 523 \frac{7}{2}\rangle \rightarrow \frac{5}{2}\rangle$	4.5	$ 651 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	3.9	10	17	6.70	14.0	0.31
	$ 532 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	4.0	$ 642 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	4.5					
	$ 541 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	4.5	$ 633 \frac{7}{2}\rangle \rightarrow \frac{5}{2}\rangle$	5.0					
	$ 404 \frac{9}{2}\rangle \rightarrow \frac{7}{2}\rangle$	4.1	$ 505 \frac{11}{2}\rangle \rightarrow \frac{9}{2}\rangle$	5.1					
^{176}Yb	$ 523 \frac{7}{2}\rangle \rightarrow \frac{5}{2}\rangle$	4.5	$ 651 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	4.2	10	13	6.72	13.9	0.29
	$ 532 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	4.1	$ 642 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	4.5					
	$ 541 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	4.5	$ 633 \frac{7}{2}\rangle \rightarrow \frac{5}{2}\rangle$	5.0					
	$ 404 \frac{9}{2}\rangle \rightarrow \frac{7}{2}\rangle$	4.0	$ 505 \frac{11}{2}\rangle \rightarrow \frac{9}{2}\rangle$	5.2					
^{178}Hf	$ 523 \frac{7}{2}\rangle \rightarrow \frac{5}{2}\rangle$	4.4	$ 505 \frac{11}{2}\rangle \rightarrow \frac{9}{2}\rangle$	4.2	9	13	6.75	13.8	0.26
	$ 532 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	4.1	$ 512 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	2.4					
	$ 541 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	4.1	$ 633 \frac{7}{2}\rangle \rightarrow \frac{5}{2}\rangle$	5.0					
	$ 402 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	1.9	$ 642 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	4.6					
^{180}Hf	$ 411 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	1.4	$ 631 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	7.8					
	$ 523 \frac{7}{2}\rangle \rightarrow \frac{5}{2}\rangle$	4.4	$ 505 \frac{11}{2}\rangle \rightarrow \frac{9}{2}\rangle$	4.2	9	17	6.80	13.8	0.25
	$ 532 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	4.1	$ 512 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	2.4					
	$ 541 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	4.1	$ 624 \frac{9}{2}\rangle \rightarrow \frac{7}{2}\rangle$	5.3					
^{226}Ra	$ 402 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	1.9	$ 633 \frac{7}{2}\rangle \rightarrow \frac{5}{2}\rangle$	5.0					
	$ 411 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	1.4	$ 642 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	4.6					
			$ 631 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	7.8					
	$ 523 \frac{7}{2}\rangle \rightarrow \frac{5}{2}\rangle$	4.3	$ 624 \frac{9}{2}\rangle \rightarrow \frac{7}{2}\rangle$	5.0	12	16	7.31	13.0	0.20
^{232}Th	$ 514 \frac{9}{2}\rangle \rightarrow \frac{7}{2}\rangle$	4.4	$ 615 \frac{11}{2}\rangle \rightarrow \frac{9}{2}\rangle$	5.0					
	$ 505 \frac{11}{2}\rangle \rightarrow \frac{9}{2}\rangle$	4.4	$ 606 \frac{13}{2}\rangle \rightarrow \frac{11}{2}\rangle$	5.6					
			$ 761 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	4.3					
	$ 651 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	4.5	$ 752 \frac{5}{2}\rangle \rightarrow \frac{3}{2}\rangle$	4.1	13	19	7.37	12.9	0.25
	$ 505 \frac{11}{2}\rangle \rightarrow \frac{9}{2}\rangle$	4.2	$ 761 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	4.0					
	$ 514 \frac{9}{2}\rangle \rightarrow \frac{7}{2}\rangle$	4.0	$ 631 \frac{3}{2}\rangle \rightarrow \frac{1}{2}\rangle$	1.0					
	$ 523 \frac{7}{2}\rangle \rightarrow \frac{5}{2}\rangle$	3.7	$ 624 \frac{9}{2}\rangle \rightarrow \frac{7}{2}\rangle$	5.0					
			$ 615 \frac{11}{2}\rangle \rightarrow \frac{9}{2}\rangle$	4.8					
			$ 606 \frac{13}{2}\rangle \rightarrow \frac{11}{2}\rangle$	5.4					

neutron contributions come from $\langle 5h_{\frac{9}{2}}m|s|5h_{\frac{11}{2}}m' \rangle$. All these matrix elements may be calculated using the properties of spherical spinors (see, e.g., Ref. [39]). The energies of all these transitions may be estimated with the use of Fig. 5 in Ref. [23].

In Table V below, we collect the values of reduced matrix elements squared $|\langle n'|s||0' \rangle|^2$ and the transition energies for proton and neutron transitions in ^{208}Pb , ^{206}Pb , and ^{138}Ba nuclei. The matrix element with the initial magnetic quantum number m' summed over $|\langle n'|s||0' \rangle|^2 \equiv \sum_{m'} |\langle n, l, j, m|s|n, l, j', m' \rangle|^2$ may be calculated with the help of the Wigner-Eckart theorem. The transition energy depends on the final quantum number m . When the energies are (nearly) degenerate, we give the sum over m of the matrix elements corresponding to the same energy.

The values for the nuclear radii R_0 and the energy of the giant dipole resonance are calculated according to the empirical formulas:

$$R_0 = 1.2A^{1/3} \text{ fm}, \quad (\text{A1})$$

$$\Delta \bar{E} = 95A^{-1/3}(1 - A^{-1/3}) \text{ MeV}. \quad (\text{A2})$$

For reference, the values of the deformation parameter δ are also presented [40,41].

2. Deformed nuclei

For deformed heavy nuclei with $\delta > 0.1$, it is convenient to use the Nilsson basis [23,42], 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, and Λ and Ω are the projections of angular and total momenta on the deformation axis. Note that $\Omega = \Lambda + \Sigma$ where Σ is the projection of the nucleon's spin on the deformation axis. The dependence of the energy levels on the deformation parameter δ in this model may be inferred from Fig. 5 in Ref. [23]. 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 Σ corresponds to $\pm\Lambda$.

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

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 ($\mathbf{e}_+$, $\mathbf{e}_-$, $\mathbf{e}_0$). The components of the vectors in this basis will be labeled by the $(+, -, 0)$ subscripts. Because of the spherical symmetry, Eq. (33) may be rewritten in terms of the “0” component of the operators $\hat{\mathbf{R}}$ and $\hat{\mathbf{R}} \times \boldsymbol{\alpha}$

$$M(\Delta E_{n'}) = \frac{\alpha}{c_{s_{1/2}} c_{p_{1/2}}} \sum_n \frac{\langle p_{1/2} | f(R) \hat{\mathbf{R}}_0 | n \rangle \langle n | f(R) (\hat{\mathbf{R}} \times \boldsymbol{\alpha})_0 | s_{1/2} \rangle}{\Delta E_n + \text{sgn}(E_n) \Delta E_{n'}} + (s_{1/2} \leftrightarrow p_{1/2}). \quad (\text{B1})$$

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

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 $\Omega_{\mu}^{\kappa}(\hat{\mathbf{R}})$ where μ is the magnetic quantum number and $\kappa = (l - j)(2j + 1)$ as

$$|s_{1/2}\rangle = c_{s_{1/2}} \begin{pmatrix} f_{s_{1/2}}(R) \Omega_{\mu}^{-1}(\hat{\mathbf{R}}) \\ ig_{s_{1/2}}(R) \Omega_{\mu}^1(\hat{\mathbf{R}}) \end{pmatrix}, \quad (\text{B2a})$$

$$|p_{1/2}\rangle = c_{p_{1/2}} \begin{pmatrix} f_{p_{1/2}}(R) \Omega_{\mu}^1(\hat{\mathbf{R}}) \\ ig_{p_{1/2}}(R) \Omega_{\mu}^{-1}(\hat{\mathbf{R}}) \end{pmatrix}, \quad (\text{B2b})$$

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

$$f_{s_{1/2}}(R) = \frac{(-1 + \gamma) J_{2\gamma}(x) - \frac{x}{2} J_{2\gamma-1}(x)}{R}, \quad (\text{B3a})$$

$$f_{p_{1/2}}(R) = \frac{(1 + \gamma) J_{2\gamma}(x) - \frac{x}{2} J_{2\gamma-1}(x)}{R}, \quad (\text{B3b})$$

$$g_{s_{1/2}}(R) = g_{p_{1/2}}(R) = \frac{Z\alpha J_{2\gamma}(x)}{R}, \quad (\text{B3c})$$

where $x \equiv \sqrt{8ZR/a_B}$.

Note that the wave functions (B3) are the zero-energy solutions of the Dirac-Coulomb equations for a pointlike nucleus. For an extended nucleus, the corresponding solution is complicated. At the current level of accuracy, it suffices to use Eqs. (B3) as an approximation to the wave functions. For the region inside the nucleus, $0 \leq R \leq R_0$, the radial wave functions $f_{s,p_{1/2}}$ and $g_{s,p_{1/2}}$ may be continued as follows:

$$f_{s_{1/2}}(R) = \frac{(-1 + \gamma)J_{2\gamma}(x_0) - \frac{x_0}{2}J_{2\gamma-1}(x_0)}{R_0}, \quad (\text{B4a})$$

$$f_{p_{1/2}}(R) = \frac{R[(1 + \gamma)J_{2\gamma}(x_0) - \frac{x_0}{2}J_{2\gamma-1}(x_0)]}{R_0^2}, \quad (\text{B4b})$$

$$g_{s_{1/2}}(R) = \frac{R}{R_0^2}Z\alpha J_{2\gamma}(x_0), \quad (\text{B4c})$$

$$g_{p_{1/2}}(R) = \frac{1}{R_0}Z\alpha J_{2\gamma}(x_0), \quad (\text{B4d})$$

where $x_0 \equiv \sqrt{8ZR_0/a_B}$.

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

2. Excited electronic states of the continuous spectrum

The excited electronic states $|n\rangle$ in the continuous spectrum may be labeled by the quantum number $\kappa = (l - j)(2j + 1)$ and the energy E , $|n\rangle \equiv |E\kappa\rangle$. In spherical coordinates, these functions read (see, e.g., Refs. [43,44]) as

$$|n\rangle \equiv |E\kappa\rangle = \begin{pmatrix} f_\kappa^E(R)\Omega_\mu^\kappa(\hat{\mathbf{R}}) \\ ig_\kappa^E(R)\Omega_\mu^{-\kappa}(\hat{\mathbf{R}}) \end{pmatrix}, \quad (\text{B5})$$

with

$$f_\kappa^E(R) = \frac{(2pR)^\gamma e^{\pi y/2} |\Gamma(\gamma + iy)| \sqrt{|E + m_e|}}{R\sqrt{\pi p}\Gamma(2\gamma + 1)} \times \text{Re}[e^{-ipR + i\eta} {}_1F_1(\gamma + 1 + iy, 2\gamma + 1, 2ipR)], \quad (\text{B6a})$$

$$g_\kappa^E(R) = -\text{sgn}(E) \frac{(2pR)^\gamma e^{\pi y/2} |\Gamma(\gamma + iy)| \sqrt{|E - m_e|}}{r\sqrt{\pi p}\Gamma(2\gamma + 1)} \times \text{Im}[e^{-ipR + i\eta} {}_1F_1(\gamma + 1 + iy, 2\gamma + 1, 2ipR)]. \quad (\text{B6b})$$

Here $p = \sqrt{E^2 - m_e^2}$ is the electron's momentum, $y = Z\alpha E/p$, $e^{i\eta} = \sqrt{-\frac{\kappa - iym_e/E}{\gamma + iy}}$, and ${}_1F_1(a, b, z)$ is the confluent hypergeometric function of the first kind. Note that the wave functions (B5) are normalized as $\langle E'\kappa | E\kappa \rangle = \delta(E' - E)$.

The functions (B6) solve for the Dirac equation with a pointlike 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:

$$f_\kappa^E(R) = b_1 R^l, \quad g_\kappa^E(R) = b_2 R^{\tilde{l}}, \quad (\text{B7})$$

where $l = |\kappa + 1/2| - 1/2$ is the orbital angular momentum corresponding to κ and $\tilde{l} = |-\kappa + 1/2| - 1/2$ is the orbital angular momentum corresponding to $-\kappa$. The values of the coefficients b_1 and b_2 are determined by matching Eqs. (B6) and (B7) on the boundary of the nucleus. The wave functions (B7) 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 (B7) 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 agreement within 5% of the known results presented in Refs. [19–21].

3. Results of calculation of electronic matrix element

Substituting the wave functions (B2) and (B5) into Eq. (B1) and performing the integration over angular variables, we obtain

$$M(\Delta E_{n'}) = -\frac{2\alpha}{9} \int_{m_e}^{\infty} \frac{T(E)dE}{E - E_{s_{1/2}} + \Delta E_{n'}} - \frac{2\alpha}{9} \int_{-\infty}^{-m_e} \frac{T(E)dE}{E - E_{s_{1/2}} - \Delta E_{n'}}, \quad (\text{B8})$$

where

$$T(E) = R_s^1(E)R_p^1(E) - R_s^{-2}(E)R_p^{-2}(E) - S_s^{-1}(E)S_p^{-1}(E) + S_s^2(E)S_p^2(E), \quad (\text{B9})$$

and the radial integrals $R_{s,p}^\kappa(E)$ and $S_{s,p}^\kappa(E)$ are defined by

$$R_s^\kappa(E) \equiv \int_0^\infty (f_{s_{1/2}} f_\kappa^E + g_{s_{1/2}} g_\kappa^E) f(R) R^2 dR, \quad (\text{B10a})$$

$$R_p^\kappa(E) \equiv \int_0^\infty (f_{p_{1/2}} g_\kappa^E + g_{p_{1/2}} f_\kappa^E) f(R) R^2 dR, \quad (\text{B10b})$$

$$S_s^\kappa(E) \equiv \int_0^\infty (f_{s_{1/2}} g_\kappa^E + g_{s_{1/2}} f_\kappa^E) f(R) R^2 dR, \quad (\text{B10c})$$

$$S_p^\kappa(E) \equiv \int_0^\infty (f_{p_{1/2}} f_\kappa^E + g_{p_{1/2}} g_\kappa^E) f(R) R^2 dR. \quad (\text{B10d})$$

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 $|E| > 500m_e$, $T(E)/(E_{s_{1/2}} - E \pm \Delta E_{n'})$ is effectively zero, so the energy integrals in Eqs. (B8) may be cut off at $|E| \approx 500m_e$. We also point out that the dominant contributions to the energy integrals (B8) 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. (B8) 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_p , M_n , and $M(\Delta \bar{E})$ are presented in Table I.

-
- [1] E. M. Purcell and N. F. Ramsey, *Phys. Rev.* **78**, 807 (1950).
 - [2] T.-D. Lee and C. N. Yang, Technical Report, Brookhaven National Lab., Upton, NY, 1957.
 - [3] L. Landau, *Sov. Phys. JETP* **5**, 336 (1957).
 - [4] L. Landau, *Nucl. Phys.* **3**, 127 (1957).
 - [5] A. D. Sakharov, *Sov. Phys. Usp.* **34**, 392 (1991).
 - [6] V. Andreev, D. G. Ang, D. DeMille, J. M. Doyle, G. Gabrielse, J. Haefner, N. R. Hutzler, Z. Lasner, C. Meisenhelder, B. R. OLeary *et al.* (ACME Collaboration), *Nature (London)* **562**, 355 (2018).
 - [7] H. Loh, K. C. Cossel, M. C. Grau, K.-K. Ni, E. R. Meyer, J. L. Bohn, J. Ye, and E. A. Cornell, *Science* **342**, 1220 (2013).
 - [8] N. Yamanaka, B. Sahoo, N. Yoshinaga, T. Sato, K. Asahi, and B. Das, *Eur. Phys. J. A* **53**, 54 (2017).
 - [9] T. E. Chupp, P. Fierlinger, M. J. Ramsey-Musolf, and J. T. Singh, *Rev. Mod. Phys.* **91**, 015001 (2019).
 - [10] V. V. Flambaum, M. Pospelov, A. Ritz, and Y. V. Stadnik, *Phys. Rev. D* **102**, 035001 (2020).
 - [11] V. V. Flambaum, I. B. Samsonov, and H. B. Tran Tan, *J. High Energy Phys.* **10** (2020) 077.
 - [12] I. B. Khriplovich, *Parity Nonconservation in Atomic Phenomena* (Gordon and Breach Science Publishers, New York, 1991).
 - [13] O. P. Sushkov, V. V. Flambaum, and I. B. Khriplovich, *Zh. Eksp. Teor. Fiz.* **87**, 1521 (1984).
 - [14] J. Bsaisou, U.-G. Meiner, A. Nogga, and A. Wirzba, *Ann. Phys. (Amsterdam)* **359**, 317 (2015).
 - [15] M. Denis and T. Fleig, *J. Chem. Phys.* **145**, 214307 (2016).
 - [16] L. V. Skripnikov, *J. Chem. Phys.* **145**, 214301 (2016).
 - [17] V. V. Flambaum, I. B. Khriplovich, and O. P. Sushkov, *Nucl. Phys. A* **449**, 750 (1986).
 - [18] V. V. Flambaum and V. A. Dzuba, *Phys. Rev. A* **101**, 042504 (2020).
 - [19] G. Plunien, B. Müller, W. Greiner, and G. Soff, *Phys. Rev. A* **43**, 5853 (1991).
 - [20] K. Pachucki, D. Leibfried, and T. W. Hänsch, *Phys. Rev. A* **48**, R1 (1993).
 - [21] G. Plunien and G. Soff, *Phys. Rev. A* **51**, 1119 (1995).
 - [22] A. V. Nefiodov, L. N. Labzowsky, G. Plunien, and G. Soff, *Phys. Lett. A* **222**, 227 (1996).
 - [23] A. Bohr and B. R. Mottelson, *Nuclear Structure*, Vol. 2 (World Scientific, Singapore, 1998).
 - [24] T. Fleig and M. Jung, *J. High Energy Phys.* **07** (2018) 012.
 - [25] V. V. Flambaum and I. B. Khriplovich, *Zh. Eksp. Teor. Fiz.* **89**, 1505 (1985).
 - [26] W. B. Cairncross, D. N. Gresh, M. Grau, K. C. Cossel, T. S. Roussy, Y. Ni, Y. Zhou, J. Ye, and E. A. Cornell, *Phys. Rev. Lett.* **119**, 153001 (2017).
 - [27] V. V. Flambaum, D. DeMille, and M. G. Kozlov, *Phys. Rev. Lett.* **113**, 103003 (2014).
 - [28] M. Pospelov and A. Ritz, *Phys. Rev. Lett.* **83**, 2526 (1999).
 - [29] M. Pospelov and A. Ritz, *Phys. Rev. D* **63**, 073015 (2001).
 - [30] M. Pospelov and A. Ritz, *Ann. Phys. (Amsterdam)* **318**, 119 (2005), Special Issue.
 - [31] J. de Vries, E. Mereghetti, and A. Walker-Loud, *Phys. Rev. C* **92**, 045201 (2015).
 - [32] J. Bsaisou, C. Hanhart, S. Liebig, U.-G. Meissner, A. Nogga, and A. Wirzba, *Eur. Phys. J. A* **49**, 31 (2013).
 - [33] M. Tanabashi *et al.* (Particle Data Group), *Phys. Rev. D* **98**, 030001 (2018).
 - [34] C. Abel, S. Afach, N. J. Ayres, C. A. Baker, G. Ban, G. Bison, K. Bodek, V. Bondar, M. Burghoff, E. Chancel *et al.*, *Phys. Rev. Lett.* **124**, 081803 (2020).
 - [35] B. Graner, Y. Chen, E. G. Lindahl, and B. R. Heckel, *Phys. Rev. Lett.* **116**, 161601 (2016).
 - [36] N. Sachdeva, I. Fan, E. Babcock, M. Burghoff, T. E. Chupp, S. Degenkolb, P. Fierlinger, S. Haude, E. Kraegelloh, W. Kilian *et al.*, *Phys. Rev. Lett.* **123**, 143003 (2019).
 - [37] F. Allmendinger, I. Engin, W. Heil, S. Karpuk, H.-J. Krause, B. Niederländer, A. Offenhäusser, M. Repetto, U. Schmidt, and S. Zimmer, *Phys. Rev. A* **100**, 022505 (2019).
 - [38] P. Sarriuren, E. Moya de Guerra, and R. Nojarov, *Phys. Rev. C* **54**, 690 (1996).
 - [39] R. Szymtkowski, *J. Math. Chem.* **42**, 397 (2007).
 - [40] S. Raman, C. Nestor, and P. Tikkanen, *At. Data Nucl. Data Tables* **78**, 1 (2001).
 - [41] N. Stone, *At. Data Nucl. Data Tables* **90**, 75 (2005).
 - [42] S. G. Nilsson, *Dan. Mat. Fys. Medd.* **29**, 1 (1955).
 - [43] V. B. Berestetskii, E. M. Lifshitz, and L. P. Pitaevskii, *Quantum Electrodynamics*, Vol. 4 (Butterworth-Heinemann, London, 1982).
 - [44] W. Greiner, *Relativistic Quantum Mechanics: Wave Equations*, Vol. 2 (Springer, New York, 2000).