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Atomic electric dipole moment induced by the nuclear electric dipole moment: The magnetic moment effect

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We have considered a mechanism for inducing a time-reversal violating electric dipole moment (EDM) in atoms through the interaction of a nuclear EDM d_N with the hyperfine interaction, the "magnetic moment effect". We have derived the operator for this interaction and presented analytical formulas for the matrix elements between atomic states. Induced EDMs in the diamagnetic atoms ^{129}Xe, ^{171}Yb, ^{199}Hg, ^{211}Rn, and ^{225}Ra have been calculated numerically. From the experimental limits on the atomic EDMs of ^{129}Xe and ^{199}Hg we have placed the following constraints on the nuclear EDMs, $|d_N(^{129}\mathrm{Xe})| < 1.1 \times 10^{-21}\,|e|$ cm and $|d_N(^{199}\mathrm{Hg})| < 2.8 \times 10^{-24}\,|e|$ cm.

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I. INTRODUCTION

Since the discovery of *CP* violation in the decay of long-lived K^0 mesons, there have been many efforts to observe *CP* or time-reversal (*T*) violation in other systems. The latter is equivalent to *CP* violation assuming the validity of the *CPT* theorem. In particular, the possible existence of a permanent electric dipole moment (EDM) of a particle would imply the violation of both parity and time-reversal invariance (see, e.g., [1]). The EDMs of the particles predicted by the standard model are too small to be detected at the present level of experimental accuracy. However, different extensions of the standard model (such as supersymmetry) predict much larger EDMs of the particles that could, in principle, be found using modern experimental techniques (see, e.g., [2,3]).

Recently, Griffith *et al.* [4] reported a sevenfold improvement on the limit of the atomic EDM of ^{199}Hg, $|d(^{199}\mathrm{Hg})| < 3.1 \times 10^{-29}\,|e|$ cm. This is the most stringent limit on an atomic EDM. In order to interpret the measurement in terms of fundamental *P,T*-violating parameters, atomic calculations are required. In our recent paper [5] (see also [6–8]) we calculated atomic electric dipole moments induced by the nuclear Schiff moment, the *P,T*-odd electron-nucleon interaction, and the electron electric dipole moment and placed limits on the coupling constants of the *P,T*-odd interactions from the new Hg result.

In this work we consider one more *P,T*-odd interaction which gives rise to an atomic EDM in the second order of perturbation theory. This interaction was first discussed by Schiff in Ref. [9]. He showed that if a nucleus has a permanent EDM $\mathbf{d}_N$, an atomic EDM may be induced due to the interaction of $\mathbf{d}_N$ with the magnetic field created by the electrons at the nucleus.

It is worth noting that the existence of a nonzero $\mathbf{d}_N$ alone is insufficient for producing observable EDM effects in neutral atoms due to electronic screening of an applied electric field at the nucleus [9]. This screening is exact for the case of a point-like nucleus experiencing electrostatic forces. One can circumvent this screening by accounting for the finite size of the nucleus or the hyperfine interaction. The former leads to the appearance of the nuclear Schiff moment; the latter "magnetic moment effect" is the subject of this work.

In his work [9] Schiff evaluated the EDMs of H and He induced by the magnetic moment effect. Hinds and Sandars later calculated the effect in TlF [10]. In the current work we present a general analysis of the effect for atoms and perform calculations for diamagnetic atoms of experimental interest.

The volume effect (nuclear Schiff moment) is generally considered to be the dominant mechanism inducing EDMs in heavy diamagnetic atoms with nuclear spin $I = 1/2$. However, it has been discovered recently that the nuclear Schiff moment is very sensitive to many-body corrections (see, e.g., [11] and references therein). These corrections suppress the bare values for the Schiff moments for all considered nuclei. Other than a general suppression, there is yet no agreement between the many-body approaches. The nuclear EDM contribution to the atomic EDM (through the magnetic moment effect) should therefore not be disregarded before the nuclear many-body problem is well understood and specific *CP*-violating models considered.

Further motivation for this work comes from the growing interest in measuring nuclear EDMs in ion storage rings [12–15]. In ions the nuclear EDM is not screened and can be measured directly. The relations obtained in this work between the nuclear and atomic EDMs enable one to place limits on nuclear EDMs from neutral atom measurements. These limits may be considered as an accuracy benchmark for proposed nuclear EDM measurements with ions.

The paper is organized as follows. In Secs. II and III we derive analytical expressions for the *P,T*-odd operator and the matrix elements of this operator between atomic states. In Sec. IV we present the equation for the atomic EDM $\mathbf{d}_{\mathrm{at}}^N$ and discuss different contributions to $\mathbf{d}_{\mathrm{at}}^N$. In Sec. V we obtain simple analytical formulas that can be used for an estimate of the atomic EDM (and this is compared to the atomic EDM induced by the nuclear Schiff moment), we describe our numerical method for EDM calculations of diamagnetic atoms, and we present results for ^{129}Xe, ^{171}Yb, ^{199}Hg, ^{211}Rn, and ^{225}Ra. Section VI contains concluding remarks.

II. GENERAL FORMALISM

Let us assume that the nucleus has a *P,T*-odd EDM, $\mathbf{d}_N \equiv \langle \mathbf{d}_N \rangle = d_N \mathbf{I}/I$. Here $\langle \mathbf{d}_N \rangle$ denotes the expectation value of the

nuclear electric dipole moment with the exact nuclear ground state wave function [16].

The *P,T*-odd operator corresponding to the magnetic moment effect can be written as [9]

$$U = -i[Q, H_M], \tag{1}$$

where square brackets in Eq. (1) denote a commutator. The operator Q is determined as (if not stated otherwise we use atomic units $\hbar = m = |e| = 1$)

$$Q = \frac{\mathbf{d}_N \cdot \mathbf{p}_N}{Z} = -\frac{1}{Z}\sum_{k=1}^{Z} \mathbf{d}_N \cdot \mathbf{p}_e^{(k)}, \tag{2}$$

where Z is the nuclear charge and $\mathbf{p}_N$ and $\mathbf{p}_e^{(k)}$ are momentum operators for the nucleus and electrons, respectively. H_M is the operator for the hyperfine interaction (HFI) which may be interpreted as the interaction of the nuclear magnetic moment $\boldsymbol{\mu}_N$ with the magnetic field of the electrons. It can be represented by the sum of the single-electron operators

$$H_M = \sum_{i=1}^{Z} \frac{\mathbf{r}_i \times \boldsymbol{\alpha}_i}{r_{i>}^3} \boldsymbol{\mu}_N, \tag{3}$$

where $r_{i>} \equiv \max(r_i, R)$ with R the nuclear radius and $\boldsymbol{\alpha} = \left(\begin{smallmatrix} 0 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & 0 \end{smallmatrix}\right)$ are the Dirac matrices. $\boldsymbol{\mu}_N = \mu_N \mathbf{I}/I$ and $\mu_N = \mu/(2\,m_p\,c)$, where μ is the nuclear magnetic moment, m_p is the nucleon mass, and the speed of light $c = 1/\alpha \approx 137$.

A note on the origin of the P,T-odd operator Eq. (1). Schiff showed (we limit the explanation to the case of an atom with nuclear EDM) that when an atom is acted upon by electrostatic forces only and the nuclear EDM and charge distributions are the same, the full Hamiltonian may be expressed as $H = H_0 + i[Q, H_0]$, where the full Hamiltonian H includes the nuclear EDM and H_0 does not. The eigenvalues of H are therefore the same as those of H_0 to first order (the expectation value of the commutator containing H_0 is zero) and do not contain the nuclear EDM, that is, there is no linear Stark shift and no observable EDM of the atom. When the hyperfine interaction is taken into account, the full Hamiltonian may be written as $H' = H_0' + i[Q, H_0'] - i[Q, H_M]$, where the prime denotes that the magnetic interaction is included. Only the commutator containing H_M may lead to observable EDM effects. We refer the reader to Schiff's landmark work Ref. [9] for details.

In Eq. (2) we assumed that the center of mass is at rest. As a result, the momentum operator of the nucleus $\mathbf{p}_N$ can be replaced by the sum of the electronic momenta $\mathbf{p}_e^{(k)}$ with the opposite sign. In the following we will deal with the electronic momenta only and omit the subscript e, that is, we denote $\mathbf{p} \equiv \mathbf{p}_e$. Then we obtain for the operator U:

$$U = -i\frac{d_N \mu_N}{ZI^2} \sum_{k=1}^{Z} \left[\frac{\mathbf{r}_k \times \boldsymbol{\alpha}_k}{r_{k>}^3} \mathbf{I}, \mathbf{I}\mathbf{p}_k \right] \equiv \sum_{k=1}^{Z} U_k \,. \tag{4}$$

We will consider the single-electron operator U_k (omitting for brevity the index k) since, as seen from Eq. (4), a generalization to the case of a many-electron atom is straightforward. In addition, we restrict ourselves to consideration of the most interesting case of nuclear spin $I = 1/2$. Then for the Cartesian components m and i of the nuclear spin $\mathbf{I}$ we have

$$I_m I_i = \frac{1}{4}\delta_{mi} + \frac{i}{2}\varepsilon_{mil} I_l. \tag{5}$$

Substituting Eq. (5) into Eq. (4) and taking into account that the terms $\sim\delta_{mi}$ will be canceled out, after simple transformations we obtain

$$U = \frac{d_N \mu_N}{Z} \left[\boldsymbol{\alpha}\boldsymbol{\sigma}_N \left\{ \mathbf{p}, \frac{\mathbf{r}}{r_>^3} \right\}_+ - \left\{ \frac{\boldsymbol{\sigma}_N \mathbf{r}}{r_>^3}, \boldsymbol{\alpha}\mathbf{p} \right\}_+ \right]. \tag{6}$$

Here $\boldsymbol{\sigma}_N = 2\mathbf{I}$ and $\{\cdots\}_+$ is an anticommutator. Now we take into account that

$$\mathbf{p} = -i\boldsymbol{\nabla} = -i\mathbf{n}\frac{\partial}{\partial r} - \frac{\mathbf{n} \times \mathbf{L}}{r},$$

with $\mathbf{L} = \mathbf{r} \times \mathbf{p}$ the orbital momentum operator.

Hence

$$\left\{ \mathbf{p}, \frac{\mathbf{r}}{r_>^3} \right\}_+ = -i \left\{ \frac{\partial}{\partial r}, \frac{r}{r_>^3} \right\}_+$$

and finally we obtain

$$U = -\frac{d_N \mu_N}{Z} \left[i\,\boldsymbol{\alpha} \cdot \boldsymbol{\sigma}_N \left\{ \frac{\partial}{\partial r}, \frac{r}{r_>^3} \right\}_+ + \left\{ \frac{\boldsymbol{\sigma}_N \cdot \mathbf{r}}{r_>^3}, \boldsymbol{\alpha}\mathbf{p} \right\}_+ \right]. \tag{7}$$

If the typical distances of interest are small one can neglect the eigenvalue and the mass of the electron in comparison with the electrostatic potential. Hinds and Sandars showed in Ref. [10] that in this approximation the operator U can be written in a more simple form:

$$U = \frac{2\,d_N \mu_N}{Z} \boldsymbol{\sigma}_N \frac{\boldsymbol{\alpha} \times \mathbf{L}}{r^3}. \tag{8}$$

In the following sections we will discuss the difference between the two forms of the operator U given by Eqs. (7) and (8).

III. ELECTRONIC MATRIX ELEMENTS

We use the following form for the electronic wave functions:

$$|n\varkappa m\rangle = \begin{pmatrix} f(r)\Omega_{jlm} \\ ig(r)\Omega_{j\tilde{l}m} \end{pmatrix} = \begin{pmatrix} f(r)\Omega_{\varkappa m} \\ ig(r)\Omega_{-\varkappa m} \end{pmatrix},$$

where $\tilde{l} \equiv 2j - l$. Using the expression for the operator U given by Eq. (7) we can derive the electronic matrix elements (MEs). Finally we find (see the Appendix for details of the derivation)

$$\begin{aligned} \langle n'\varkappa' m'|U|n\varkappa m\rangle = -\frac{d_N \mu_N}{Z} \boldsymbol{\sigma}_N \langle \varkappa' m'|\mathbf{n}|\varkappa m\rangle \int_0^\infty \Bigg[\frac{\varkappa + \varkappa'}{r} \\ \times \{f'g(\varkappa - \varkappa' + 1) + fg'(\varkappa' - \varkappa + 1)\} \\ + (\varepsilon' - \varepsilon)(\varkappa - \varkappa')(f'f - g'g) \Bigg] \frac{r^3}{r_>^3} dr. \end{aligned} \tag{9}$$

If we factorize $(\varepsilon' - \varepsilon)$ in the second term of Eq. (9), we can rewrite it as

$$(\varepsilon - \varepsilon')\frac{d_N \mu_N}{Z} \boldsymbol{\sigma}_N \langle n'\varkappa' m'|\, i\,\frac{\mathbf{r} \times \boldsymbol{\sigma}}{r_>^3} |n\varkappa m\rangle. \tag{10}$$

Then Eq. (9) can be represented by

$$\langle n'\varkappa'm'|U|n\varkappa m\rangle = -\frac{d_N\mu_N}{Z}\boldsymbol{\sigma}_N\Bigg[\langle\varkappa'm'|\mathbf{n}|\varkappa m\rangle(\varkappa+\varkappa') \times\int_0^\infty\{f'g(\varkappa-\varkappa'+1) + fg'(\varkappa'-\varkappa+1)\}\frac{r^2dr}{r_>^3} + (\varepsilon'-\varepsilon) \times\langle n'\varkappa'm'|\,i\frac{\mathbf{r}\times\boldsymbol{\sigma}}{r_>^3}|n\varkappa m\rangle\Bigg]. \tag{11}$$

As seen from Eqs. (9) and (11), the first term in these equations disappears if $\varkappa'+\varkappa=0$. It happens for the states with $l'=l\pm1$ and $j'=j$. As a result, the MEs $\langle-\varkappa m'|U|\varkappa m\rangle$ turn out to be proportional to $(\varepsilon'-\varepsilon)$. For heavy atoms these MEs are small. In particular, it means that the MEs $\langle s|U|p_{1/2}\rangle$ contribute less to the atomic EDM than the MEs $\langle s|U|p_{3/2}\rangle$. If we neglect the term $\sim(\varepsilon'-\varepsilon)$ in Eqs. (9) and (11), then Eqs. (7) and (8) lead to the same formula for the MEs.

IV. ATOMIC EDM

In this section we present our calculations of EDMs induced by the operator U for several diamagnetic atoms of experimental interest having $I=1/2$. The EDM $\mathbf{d}_{\rm at}^N=d_{\rm at}^N\boldsymbol{\sigma}_N$ of an atom in the state $|0\rangle$ in the second order of perturbation theory is given by

$$\mathbf{d}_{\rm at}^N = 2\sum_K\frac{\langle0|\mathbf{D}|K\rangle\langle K|U|0\rangle}{E_0-E_K}, \tag{12}$$

where $\mathbf{D}=-\mathbf{r}$ is the electric dipole operator and the summation goes over all intermediate states $|K\rangle$ allowed by the selection rules.

As follows from the consideration given in the preceding section, it is convenient to present the operator U as a product of two operators, one relating to the electronic part and the other relating to the nuclear part, that is,

$$U=\mathbf{U}_{\rm el}\cdot\boldsymbol{\sigma}_N, \tag{13}$$

where $\mathbf{U}_{\rm el}$ describes the electronic part of the operator U.

Applying the Wigner-Eckart theorem, and summing over magnetic quantum numbers m of the initial and final states in Eq. (12), we obtain for $d_{\rm at}^N$:

$$d_{\rm at}^N = \frac{2}{3}\sum_K\frac{\langle0||r||K\rangle\langle0||U_{\rm el}||K\rangle}{E_K-E_0}. \tag{14}$$

To carry out calculations of atomic EDMs for atoms with closed shells, it is convenient to rewrite Eqs. (12) and (14) in terms of single-electron wave functions. Then the corresponding expressions for $\mathbf{d}_{\rm at}^N$ and $d_{\rm at}^N$ are given by

$$\mathbf{d}_{\rm at}^N = 2\sum_{c,n}\frac{\langle c|\mathbf{r}|n\rangle\langle n|U|c\rangle}{\varepsilon_n-\varepsilon_c} \tag{15}$$

and

$$d_{\rm at}^N = \frac{2}{3}\sum_{c,n}\frac{\langle n||r||c\rangle\langle n||U_{\rm el}||c\rangle}{\varepsilon_n-\varepsilon_c}, \tag{16}$$

where the indices c and n relate to the single-electron core and virtual orbitals and $\varepsilon_{c(n)}$ are the single-electron core (virtual) energies, correspondingly.

Note that if we substitute the second term of Eq. (11) into Eq. (15), then after cancellation of $(\varepsilon_n-\varepsilon_c)$ in the numerator and denominator and applying closure $\sum_n|n\rangle\langle n|=1$, we obtain a term which is proportional to

$$\sum_c\langle c|\,\frac{\mathbf{r}\,[(\mathbf{r}\times\boldsymbol{\sigma})\,\boldsymbol{\sigma}_N]}{r_>^3}\,|c\rangle.$$

It can be readily shown that if we apply again the Wigner-Eckart theorem and sum up over the projections m_c of the total angular momenta j_c, this term goes to zero and therefore does not contribute to $\mathbf{d}_{\rm at}^N$. In particular, it means that the second term in Eq. (11) does not contribute to the atomic EDM for closed-shell atoms.

Keeping in mind that we intend to carry out calculations of EDMs for atoms with closed shells, we can neglect the term $\sim(\varepsilon'-\varepsilon)$ in Eq. (11). Accounting for Eq. (13), we obtain the following expression for the reduced ME of the operator $U_{\rm el}$:

$$\langle n'\varkappa'||U_{\rm el}||n\varkappa\rangle \approx -\frac{d_N\mu_N}{Z}\langle\varkappa'||n||\varkappa\rangle(\varkappa+\varkappa') \times\int_0^\infty\{f'g(\varkappa-\varkappa'+1) + fg'(\varkappa'-\varkappa+1)\}\frac{dr}{r}, \tag{17}$$

where the reduced ME $\langle\varkappa'||n||\varkappa\rangle$ is given by

$$\langle\varkappa'||n||\varkappa\rangle = (-1)^{j'+1/2}\sqrt{(2j'+1)(2j+1)} \times\begin{pmatrix} j' & j & 1\\ -1/2 & 1/2 & 0\end{pmatrix}\xi(l'+l+1) \tag{18}$$

with

$$\xi(x)=\begin{cases}1, & \text{if} \quad x \text{ is even}\\ 0, & \text{if} \quad x \text{ is odd.}\end{cases}$$

In Eq. (17) we also took into account that the integral inside the nucleus is very small and replaced $r_>$ by r. As we mentioned above, the MEs of the operator $U_{\rm el}$ turn to zero if $\varkappa+\varkappa'=0$. It means that MEs between states with the same total angular momentum though different parity like $\langle s||U_{\rm el}||p_{1/2}\rangle$, $\langle p_{3/2}||U_{\rm el}||d_{3/2}\rangle$, etc. are equal to zero.

V. METHOD OF CALCULATION AND RESULTS

A. Analytical estimates

In this section we derive the analytical expression for the matrix element Eq. (17). Outside the nucleus the wave functions $f_{n\varkappa}$ and $g_{n\varkappa}$ can be represented by [17]

$$f_{n\varkappa} = \frac{\varkappa}{|\varkappa|}\frac{1}{\sqrt{Z\nu^3r}}\left[(\gamma+\varkappa)J_{2\gamma}(x)-\frac{x}{2}J_{2\gamma-1}(x)\right],$$
$$g_{n\varkappa} = \frac{\varkappa}{|\varkappa|}\frac{1}{\sqrt{Z\nu^3r}}Z\alpha\,J_{2\gamma}(x), \tag{19}$$

where $x\equiv\sqrt{8Zr}$, $\gamma=\sqrt{\varkappa^2-Z^2\alpha^2}$, and $J_\nu(x)$ are Bessel functions.

Using these expressions for the radial wave functions we obtain for the radial integral

$$\int_0^\infty f_{n'\varkappa'} g_{n\varkappa} \frac{dr}{r} = \frac{Z^2\alpha}{(\nu'\nu)^{3/2}} \lambda R_M. \tag{20}$$

Here

$$\lambda \equiv \frac{\varkappa'\varkappa}{|\varkappa'\varkappa|} 96 \left[\varkappa' - 1 + \frac{1}{4}(\varkappa^2 - \varkappa'^2) \right] A_{j'j}, \tag{21}$$

the relativistic enhancement factor R_M is given by [18]

$$R_M \equiv \frac{1}{A_{j'j}} \frac{\Gamma(\gamma' + \gamma - 2)}{\Gamma(\gamma' - \gamma + 3)\Gamma(\gamma - \gamma' + 3)\Gamma(\gamma' + \gamma + 3)},$$

$\Gamma(\beta)$ are the γ functions and the factor $A_{j'j}$ is determined as follows:

$$A_{j'j} \equiv \frac{(j' + j - 2)!}{(j' - j + 2)!(j - j' + 2)!(j' + j + 3)!}. \tag{22}$$

Let us consider an important particular case of the ME $\langle s||U_{\rm el}||p_{3/2}\rangle$. In this case $\varkappa' = -1$ and $\varkappa = -2$. Then the first term under the integral in Eq. (17) turns to zero while for the second term in the integral we find, using Eqs. (20) and (21),

$$\int_0^\infty f_{np_{3/2}} g_{n's} \frac{dr}{r} = -\frac{1}{2} \frac{Z^2\alpha}{(\nu'_s \nu_{p_{3/2}})^{3/2}} R_M. \tag{23}$$

For the matrix element we therefore have

$$\langle n's||U_{\rm el}||np_{3/2}\rangle \approx d_N \mu_N \frac{2\sqrt{3}\, Z\alpha}{(\nu'_s \nu_{p_{3/2}})^{3/2}} R_M. \tag{24}$$

This analytical expression demonstrates how the atomic EDM depends on Z and nuclear parameters. The enhancement factors R_M for the medium atoms are close to unity. They grow with increasing Z approaching the value 2.2 for Ra. The values of the enhancement factors for the diamagnetic atoms considered in this work are listed in Table I.

A simple analytical estimate gives the following result for the atomic EDM:

$$d_{\rm at}^N \approx \pm 10^{-7} \mu Z R_M d_N, \tag{25}$$

where the upper sign (+) relates to the divalent atoms and the lower sign (−) relates to the noble gases. This formula gives values in reasonable agreement (within a factor of ∼2) with the many-body Dirac-Hartree-Fock (DHF) results presented in the following section.

It is instructive to compare this estimate with a similar one for the atomic EDM induced by the nuclear Schiff moment S,

$$d_{\rm at}^S \approx \mp 10^{-22} Z^2 (R_{1/2} + 2R_{3/2})[S/(|e|{\rm fm}^3)]|e|{\rm cm}, \tag{26}$$

where $R_{1/2}$ and $R_{3/2}$ are relativistic enhancement factors corresponding to $s - p_{1/2}$ and $s - p_{3/2}$ weak matrix elements, respectively (see Ref. [18] for these factors). Again the upper sign is for the divalent atoms and the lower sign is for noble gases. The values obtained from this simple formula also reasonably agree (within a factor of 2) with the many-body DHF results obtained in Refs. [5,7,8].

The atomic EDM induced by the Schiff moment benefits from an extra Z dependence which becomes very important in heavy atoms [17]. In Sec. V C we consider a specific mechanism for inducing the Schiff and nuclear dipole moments and compare the sizes of the induced atomic EDMs.

TABLE I. The nuclear charges Z and the relativistic enhancement factors R_M.

	^{129}Xe	^{171}Yb	^{199}Hg	^{211}Rn	^{225}Ra
Z	54	70	80	86	88
R_M	1.28	1.56	1.83	2.06	2.15

B. Numerical method of calculation and results

Here we describe the simple numerical methods we use for calculations of atomic EDMs for closed-shell atoms. At the first stage we solve DHF equations in the V^N approximation. This means that we include all electrons forming the ground state of the atom in a self-consistency procedure

$$H_0 \psi_c = \varepsilon_c \psi_c. \tag{27}$$

Here H_0 is the relativistic Hartree-Fock Hamiltonian and ψ_c are single-electron wave functions of the core.

To take into account polarization of the atomic core by external fields (the electric dipole field or the P,T-odd field), we solve the random phase approximation (RPA) equations

$$(H_0 - \varepsilon_c)\delta\psi_c = -(F + \delta V^N)\psi_c, \tag{28}$$

where F is the operator of the external field and δV^N is the correction to the self-consistent potential due to the effect of the external field. The RPA equations (28) are solved self-consistently for all states in the core.

In implementing the DHF and RPA procedures for calculations of the atomic EDMs, we have used two equivalent approaches. The first involves the construction of virtual orbitals and summation over states. The second involves the direct solution of the perturbed orbital $\delta\psi_c$ on the grid.

In the former method the virtual orbitals are constructed by multiplication of the previous orbital of the same partial wave to a smooth function of r with subsequent orthogonalization of this orbital to the rest of the orbitals. This method was described in detail in Refs. [19] and [20].

In the latter method, implemented in Ref. [7] for calculation of the Schiff moments, instead of direct summation over virtual states in Eq. (15), we evaluate $\mathbf{d}_{\rm at}^N = 2\sum_c \langle c| - \mathbf{r}|\delta c^U\rangle$, where $|\delta c^U\rangle = \sum_n \frac{\langle n|U|c\rangle}{\epsilon_c - \epsilon_n}|n\rangle$ is a solution of Eq. (28) for the P,T-odd field. In taking into account core polarization by the fields, the correction goes to one field and not the other to avoid double counting. See Ref. [7] for details.

The results of numerical calculations carried out using the DHF and RPA methods for the considered diamagnetic atoms are presented in Table II. It is seen that inclusion of the RPA

TABLE II. The values of $d_{\rm at}^N$ in units ($10^{-6}\, d_N$) obtained in the DHF and RPA approximations.

	^{129}Xe$_{54}$	^{171}Yb$_{70}$	^{199}Hg$_{80}$	^{211}Rn$_{86}$	^{225}Ra$_{88}$
DHF	4.4	2.6	3.2	−9.1	−9.5
RPA	5.8	11	11	−13	−33

corrections increases the size of the atomic EDM. For the noble gases (Xe and Rn) the RPA corrections contribute at the level of 30% –40%, while for atomic Hg, Yb, and Ra, which have two s electrons above closed shells, the RPA corrections are much larger. In fact, they increase the EDMs of these atoms several times compared to the DHF values. The reason for this increase is that the two s electrons are loosely bound and can be easily excited. As a result, account of higher orders of perturbation theory (like the RPA corrections) leads to a significant change in the "bare" results obtained in the DHF approximation. This result is similar to that obtained for other contributions to the P,T-odd atomic EDM discussed in Refs. [5,7,8] for the same atoms.

We note that in our previous works [5,7] a check on the RPA results for Yb, Hg, and Ra was carried out by performing more sophisticated calculations in the V^{N-2} approximation. In this approach, correlations between the two valence electrons and the core were taken into account using many-body perturbation theory (MBPT) while correlations between the valence electrons were accounted for using the configuration interaction (CI) method (see Refs. [21,22] regarding the CI + MBPT method). It was found in [5,7] that the two approaches (V^N and V^{N-2}) yield results for the EDMs that differ by less than 20% for the various P,T-odd mechanisms. The good agreement of the results obtained using two very different approaches is a strong argument in favor of the stability of the RPA results. Because the operator considered in this work is of similar form to the other P,T-odd operators, we expect that our RPA results for the atomic EDMs are accurate to about 20%.

Using the experimental limits on the P,T-odd atomic electric dipole moments of ^{129}Xe [23]

$$d(^{129}\mathrm{Xe}) = (0.7 \pm 3.3_{\mathrm{stat}} \pm 0.1_{\mathrm{syst}}) \times 10^{-27}\, e\,\mathrm{cm} \longrightarrow |d(^{129}\mathrm{Xe})| < 6.6 \times 10^{-27}\, |e|\,\mathrm{cm} \quad (29)$$

and ^{199}Hg [4]

$$d(^{199}\mathrm{Hg}) = (0.49 \pm 1.29_{\mathrm{stat}} \pm 0.76_{\mathrm{syst}}) \times 10^{-29}\, e\,\mathrm{cm} \longrightarrow |d(^{199}\mathrm{Hg})| < 3.1 \times 10^{-29}\, |e|\,\mathrm{cm} \quad (30)$$

we are able to place constraints on the nuclear EDMs $d_N(^{129}\mathrm{Xe})$ and $d_N(^{199}\mathrm{Hg})$. Using Eqs. (29) and (30), and the results presented in Table II we obtain

$$|d_N(^{129}\mathrm{Xe})| < 1.1 \times 10^{-21}\, |e|\,\mathrm{cm}, \qquad |d_N(^{199}\mathrm{Hg})| < 2.8 \times 10^{-24}\, |e|\,\mathrm{cm}. \quad (31)$$

We have not included the atomic theory error in these limits.

C. Contributions to the nuclear EDM

For spherical nuclei with spin determined by a single unpaired nucleon, there are several main terms that contribute to the nuclear EDM. One of them is characterized by the P,T-odd nucleon-nucleon interaction while the other ones are the contributions from the EDMs of the neutron (d_n) and the proton (d_p). Thus, we can write d_N as [18]

$$d_N = d_N^\eta + t_I d_n + a_p d_p, \quad (32)$$

where we denote by d_N^η the contribution from the P,T-odd nucleon-nucleon interaction; $t_I = 1$ for $I = l_I + 1/2$ and $t_I = -I/(I+1)$ for $I = l_I - 1/2$ (with l_I being the orbital momentum of the unpaired nucleon) and the coefficient a_p is numerically close to 0.1.

In Refs. [18,24] it was shown that the nuclear EDM induced by the P,T-odd nucleon-nucleon interaction can exceed the nucleon EDM by more than two orders of magnitude. If we neglect the terms proportional to d_n and d_p in Eq. (32) and express d_N^η through the T-odd nucleon-nucleon coupling constant η, we obtain [24]

$$d_N \approx 4 \times 10^{-13} \left(q - \frac{Z}{A}\right) t_I \eta\,, \quad (33)$$

where $q = 0$ and 1 for an outer neutron and proton, respectively. Taking into account that for all atoms considered $q = 0$, we obtain for d_N (in units of $|e|$ cm)

$$d_N \approx -2 \times 10^{-21} \frac{Z}{A}\, t_I \eta\, |e|\,\mathrm{cm}. \quad (34)$$

Interactions of the outer neutron with both protons and neutrons of the core contribute to the nuclear EDM; the coupling constant $\eta = \frac{Z}{A}\eta_{\mathrm{np}} + \frac{N}{A}\eta_{\mathrm{nn}}$. Using Eq. (34) and the results for d_{at}^N given in Table II we can express the values of the atomic EDMs through η. These results for ^{129}Xe, ^{171}Yb, ^{199}Hg, and ^{211}Rn are presented in Table III.

To get an idea of the relative size of the induced atomic EDMs compared to those induced by the Schiff moment, we consider the Schiff moment produced by the nucleon-nucleon interaction. For all considered atoms, the nuclei have an outer neutron. This means that there is no direct contribution to the Schiff moment. The protons need to be excited to distort the charge density and create a P,T-odd charge distribution. A simple numerical calculation in the Woods-Saxon potential with spin-orbit interaction included gives for ^{199}Hg $S(^{199}\mathrm{Hg}) = -1.4 \times 10^{-8} \eta_{\mathrm{np}} |e|\mathrm{fm}^3$ [24]. Using this result, the calculation for the atomic EDM induced by the Schiff moment $d_{\mathrm{at}}^S = -2.8 \times 10^{-17} [S/(|e|\mathrm{fm}^3)] |e|\mathrm{cm}$ [5,7], and the result for the atomic EDM induced by d_N through η presented in Table III, we obtain

$$\left|\frac{d_{\mathrm{at}}^N(^{199}\mathrm{Hg})}{d_{\mathrm{at}}^S(^{199}\mathrm{Hg})}\right| \approx 0.01 \frac{(0.4\eta_{\mathrm{np}} + 0.6\eta_{\mathrm{nn}})}{\eta_{\mathrm{np}}}. \quad (35)$$

(The ratio is larger for lighter atoms.) While this indicates that the contribution to the atomic EDM from the nuclear EDM is significantly smaller than that from the nuclear Schiff moment, we remind the reader that we have used a very

TABLE III. I^p is the spin and parity of the nuclear ground state and μ is the magnetic moment expressed in nuclear magnetons [29]. The values of d_{at}^N in units ($10^{-27}\, \eta\, |e|$ cm) are obtained in the RPA approximation.

	I^p	μ	t_I	d_{at}^N
^{129}Xe	$1/2^+$	−0.7780	1	−5.1
^{171}Yb	$1/2^-$	0.4919	−1/3	3.1
^{199}Hg	$1/2^-$	0.5059	−1/3	3.1
^{211}Rn	$1/2^-$	0.60	−1/3	−3.7
^{225}Ra	$1/2^+$	−0.734	1	

simple model for the nucleus. Indeed, it is only recently that many-body calculations have been performed for the nuclear Schiff moment and these have demonstrated that many-body corrections are large and lead to a suppression of the bare results for all nuclei considered (see, e.g., the most recent calculation [11] and references therein). The results of the different many-body approaches, however, are not in agreement. For example, in their RPA approach, Dmitriev and Sen'kov [25] find a suppression of two orders of magnitude in the isoscalar channel of the *P,T*-odd pion-nucleon-nucleon interaction and a suppression of one order of magnitude in the isotensor channel for the case of ^{199}Hg. In the fully self-consistent approach of Ban *et al.* [11] applied to ^{199}Hg, one order of magnitude suppression is seen in the isoscalar and isotensor channels, while instabilities are seen in the isovector channel, with results even varying in sign. Therefore, until the many-body problem is well understood, and specific *CP*-violation models considered, the contribution to the atomic EDM from the nuclear dipole moment should not be dismissed.

It is worth noting that there are nuclei with octupole deformation (such as, e.g., ^{223}Rn and ^{223,225}Ra). For these nuclei the nuclear EDM cannot be approximated by the simple formula Eq. (34). As shown in [26,27], the *P,T*-odd nuclear forces lead to an enhanced collective dipole moment that can significantly exceed single-particle moments. The enhancement of the nuclear EDM is also possible in nuclei with quadrupole deformations due to mixing of close opposite-parity levels [18,28].

VI. CONCLUSION

We have derived an expression for the *P,T*-odd operator produced by the interaction of a *P,T*-odd electric dipole moment of the nucleus with the operator of the hyperfine interaction. We have presented simple analytical formulas that can be used for an estimate of the EDM for different atoms. Using numerical methods we have found the contributions to atomic EDMs caused by this *P,T*-odd interaction for a number of diamagnetic atoms. Using the experimental limits on the atomic electric dipole moments of ^{129}Xe and ^{199}Hg, we constrain the EDMs of the nuclei of ^{129}Xe and ^{199}Hg to be $|d_N(^{129}\mathrm{Xe})| < 1.1 \times 10^{-21}\,|e|$ cm and $|d_N(^{199}\mathrm{Hg})| < 2.8 \times 10^{-24}\,|e|$ cm, correspondingly. These limits give an accuracy benchmark for proposed measurements of nuclear EDMs in ion storage rings.

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APPENDIX

For the matrix elements of the two operators entering Eq. (7) (see the main text) we obtain after certain transformations

$$\langle n'\varkappa'm'|i\boldsymbol{\alpha}\cdot\boldsymbol{\sigma}_N\left\{\frac{\partial}{\partial r},\frac{r}{r_>^3}\right\}_+|n\varkappa m\rangle$$

$$=\boldsymbol{\sigma}_N\langle\varkappa'm'|\mathbf{n}|\varkappa m\rangle\int_0^\infty\left[(\varkappa'-\varkappa-1)\left(g\frac{df'}{dr}-f'\frac{dg}{dr}\right)\right.$$
$$\left.+(\varkappa-\varkappa'-1)\left(g'\frac{df}{dr}-f\frac{dg'}{dr}\right)\right]\frac{r^3}{r_>^3}dr \qquad \text{(A1)}$$

and

$$\langle n'\varkappa'm'|\left\{\frac{\boldsymbol{\sigma}_N\cdot\mathbf{r}}{r_>^3},\boldsymbol{\alpha}\mathbf{p}\right\}_+|n\varkappa m\rangle$$
$$=\boldsymbol{\sigma}_N\langle\varkappa'm'|\mathbf{n}|\varkappa m\rangle\int_0^\infty\left[g'\frac{df}{dr}-f'\frac{dg}{dr}+g\frac{df'}{dr}\right.$$
$$\left.-f\frac{dg'}{dr}+\frac{\varkappa'+\varkappa}{r}(f'g+fg')\right]\frac{r^3}{r_>^3}dr. \qquad \text{(A2)}$$

Adding (A1) and (A2) we find for the ME of the operator U:

$$\langle n'\varkappa'm'|U|n\varkappa m\rangle=-\frac{d_N\mu_N}{Z}\boldsymbol{\sigma}_N\langle\varkappa'm'|\mathbf{n}|\varkappa m\rangle\int_0^\infty\left[(\varkappa'-\varkappa)\right.$$
$$\times\left(g\frac{df'}{dr}-f'\frac{dg}{dr}-g'\frac{df}{dr}+f\frac{dg'}{dr}\right)$$
$$\left.+\frac{\varkappa'+\varkappa}{r}(f'g+fg')\right]\frac{r^3}{r_>^3}dr. \qquad \text{(A3)}$$

Using the Dirac equations for the radial wave functions

$$\begin{aligned}\frac{df'}{dr}&=-\frac{1+\varkappa'}{r}f'+\left(\varepsilon'+m+\frac{Z\alpha}{r}\right)g',\\ \frac{dg}{dr}&=-\frac{1-\varkappa}{r}g-\left(\varepsilon-m+\frac{Z\alpha}{r}\right)f,\\ \frac{df}{dr}&=-\frac{1+\varkappa}{r}f+\left(\varepsilon+m+\frac{Z\alpha}{r}\right)g,\\ \frac{dg'}{dr}&=-\frac{1-\varkappa'}{r}g'-\left(\varepsilon'-m+\frac{Z\alpha}{r}\right)f',\end{aligned} \qquad \text{(A4)}$$

we can write

$$g\frac{df'}{dr}-f'\frac{dg}{dr}-g'\frac{df}{dr}+f\frac{dg'}{dr}$$
$$=\frac{\varkappa'+\varkappa}{r}(fg'-f'g)-(\varepsilon'-\varepsilon)(f'f-g'g). \qquad \text{(A5)}$$

Now substituting (A5) to (A3) we finally arrive at Eq. (9) given in the main text.

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