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Electric dipole moments of Hg, Xe, Rn, Ra, Pu, and TlF induced by the nuclear Schiff moment and limits on time-reversal violating interactions

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We have calculated the atomic electric dipole moments (EDMs) induced in ^{199}Hg, ^{129}Xe, ^{223}Rn, ^{225}Ra, and ^{239}Pu by their respective nuclear Schiff moments S. The results are [in units $10^{-17}S(e\,\mathrm{fm}^3)^{-1}e$ cm]: $d(^{199}\mathrm{Hg})=-2.8$, $d(^{129}\mathrm{Xe})=0.38$, $d(^{223}\mathrm{Rn})=3.3$, $d(^{225}\mathrm{Ra})=-8.5$, and $d(^{239}\mathrm{Pu})=-11$. We have also calculated corrections to the parity- and time-invariance-violating (P,T-odd) spin-axis interaction constant in TlF. These results are important for the interpretation of atomic and molecular experiments on EDMs in terms of fundamental P,T-odd parameters.

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I. INTRODUCTION

Recently, a stringent upper limit on the ground-state atomic electric dipole moment (EDM) of ^{199}Hg was obtained [1],

$$d(^{199}\mathrm{Hg})=-(1.06\pm0.49\pm0.40)\times10^{-28}e\ \ \mathrm{cm}. \tag{1}$$

(The respective errors are statistical and systematic.) This is the best experimental upper limit on an atomic EDM to date. Combined with calculations, this limit can be interpreted in terms of limits on fundamental parity- and time-invariance-violating (P,T-odd) parameters. These limits tightly constrain competing theories of CP violation.

Hg has closed electronic subshells, $J=0$. The measurement (1) is therefore sensitive to P,T-violating mechanisms that originate from the nucleus. The P,T-odd nuclear moment that can induce a Hg EDM is the Schiff moment (the nuclear EDM is screened by atomic electrons [2] and the magnetic quadrupole moment does not contribute due to zero electron angular momentum). Note that there are other mechanisms by which the Hg EDM can be induced, such as the P,T-odd electron-nucleon interaction (see, e.g., Ref. [3]) and the electron EDM (it contributes due to the hyperfine interaction) [4,5].

A value for the EDM of Hg induced by the Schiff moment S was estimated in Ref. [6], $d(^{199}\mathrm{Hg})=-4\times10^{-17}S(e\ \ \mathrm{fm}^3)^{-1}e$ cm. This value has been used for the interpretation of the measurements of the Hg EDM in terms of P,T-odd nuclear parameters. However, this value was obtained indirectly from an atomic calculation [7] of the EDM of Hg induced by the P,T-odd electron-nucleon tensor interaction.

In this work we have performed numerical calculations of the EDMs induced by S in Hg and in other diamagnetic atoms of current experimental interest. Our result for Hg,

$$d(^{199}\mathrm{Hg})=-2.8\times10^{-17}\left(\frac{S}{e\ \ \mathrm{fm}^3}\right)e\ \ \mathrm{cm}, \tag{2}$$

differs from the previous estimate (about 30% smaller) and places a more conservative constraint on S, and hence the underlying P,T-odd mechanisms that induce it.

The other atoms we have studied in this work are ^{129}Xe, ^{223}Rn, ^{225}Ra, and ^{239}Pu, in their ground states. There has been a recent measurement of the EDM induced in ^{129}Xe [8],

$$d(^{129}\mathrm{Xe})=(0.7\pm3.3\pm0.1)\times10^{-27}e\ \ \mathrm{cm}. \tag{3}$$

A calculation for the Xe EDM induced by S has previously been performed at the Hartree-Fock level [9], $d(^{129}\mathrm{Xe})=0.27\times10^{-17}S(e\ \ \mathrm{fm}^3)^{-1}e$ cm. In this work we have found that with core polarization included, this value becomes

$$d(^{129}\mathrm{Xe})=0.38\times10^{-17}\left(\frac{S}{e\ \ \mathrm{fm}^3}\right)e\ \ \mathrm{cm}. \tag{4}$$

This value is 40% larger than the previous calculation and hence places a tighter constraint on the P,T-odd parameters extracted from the Xe measurement.

No experiments or direct calculations have been performed for the ground-state EDMs of Rn, Ra, and Pu. In these atoms there is a possibility for an enhanced Schiff moment due to static [10] or even soft [11] octupole deformation.

Measurements of EDMs of Rn isotopes are in preparation [12]. Rn is a heavier analog of Xe. As well as the nuclear enhancement in Rn compared to Xe, there is an electronic enhancement arising from the increase of the electronic density near the nucleus with higher Z. An estimate of the Rn EDM was made in Ref. [10] by extrapolation of the EDM of Xe [9], taking into account the enhancement due to increase in Z.

Also, estimates for Ra [10] and Pu [13] have previously been carried out using the same extrapolation method as for Rn, although here the results were extrapolated from the estimate of the EDM of Hg [6].

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In Sec. IV (Table VII) the values of previous calculations/estimates for the atomic EDMs induced by S in Xe, Rn, Ra, Hg, and Pu and the results of this work are presented.

The experimental study of the P,T-odd effects induced by the nuclear Schiff moments is not restricted to atoms. In the paper [14] the P,T-odd spin-axis interaction in TlF molecule was measured. From this experiment a limit can be placed on the Schiff moment of the ^{205}Tl nucleus. Recently the accuracy of the molecular calculations was significantly increased (see Ref. [15], and references therein) and here we want to use these new molecular calculations to obtain a more reliable limit on $S(^{205}\mathrm{Tl})$. This is dealt with in Sec. III D.

II. METHOD OF CALCULATION

The nuclear Schiff moment S produces a P,T-odd electrostatic potential φ. The interaction Hamiltonian

$$H_W=\sum_i h_W^i=-e\sum_i \varphi(\mathbf{R}_i), \tag{5}$$

mixes states of opposite parity and induces a static atomic EDM. In previous calculations the contact form for the electrostatic potential was used,

$$\varphi(\mathbf{R})=4\pi\mathbf{S}\cdot\boldsymbol{\nabla}\delta(\mathbf{R}). \tag{6}$$

However, for relativistic electrons the matrix element of this potential diverges. Usually this problem is solved by a cutoff of the electron wave functions at the nuclear surface. In the work [13] it was found that there is a more convenient form for φ suitable for relativistic atomic calculations,

$$\varphi(\mathbf{R})=-\frac{3\mathbf{S}\cdot\mathbf{R}}{B}\rho(R), \tag{7}$$

where $B=\int\rho(R)R^4dR$, and $\rho(R)$ is the nuclear density. This expression arises because the Schiff moment produces a constant electric field along the nuclear spin inside the nucleus [13].

The atomic EDM induced in the many-body state N by the P,T-odd interaction H_W can be expressed as

$$d=2\sum_M \frac{\langle N|H_W|M\rangle\langle M|D_z|N\rangle}{E_N-E_M}, \tag{8}$$

where the sum M runs over a complete set of many-body states, E_N and E_M are atomic energies, and D_z is the atomic electric dipole operator.

The atoms we have studied in this work are ^{129}Xe, ^{223}Rn, ^{225}Ra, ^{199}Hg, and ^{239}Pu. Of course, the method of calculation of the electronic states depends on the electronic configuration of the atom. The starting point for the calculations is to self-consistently solve the single-particle relativistic Hartree-Fock (HF) equation,

$$h_0|i\rangle=\epsilon_i|i\rangle, \tag{9}$$

$$h_0=c\boldsymbol{\alpha}\cdot\mathbf{p}+(\beta-1)c^2-Z\alpha/r+V, \tag{10}$$

for each electron $|i\rangle$ contributing to the potential V.

For the noble gases Xe and Rn the calculations are performed in the Hartree-Fock approximation in the V^N potential. This corresponds to solving Eqs. (9) and (10) for the N electrons of the atom, that is, in a self-consistent potential V^N formed from all N electrons.

The atoms Ra and Hg can be treated as closed-shell systems or as atoms with two electrons above closed shells. Correspondingly, we have performed two separate calculations for these atoms: one in the V^N potential; and a more detailed calculation in the V^{N-2} potential [solving Eqs. (9) and (10) for the $N-2$ electrons in the atom] with correlations between the external electrons and between the external electrons and the core included. The interaction of the two external electrons is treated using the configuration interaction (CI) method and the correlations of the external electrons with the core is accounted for by many-body perturbation theory (MBPT). This technique, the CI+MBPT method, was developed in Refs. [16,17].

Pu corresponds to an open-shell atom, with electron configuration $5f^67s^2$. We have performed a simple calculation for this atom, in the V^N approximation, but with the contribution of the open f shell weighted according to its occupancy. (This simple calculation in the V^N potential, with no correlations accounted for, is justified by comparison of the results of the calculations for Hg and Ra in the potentials V^N and V^{N-2}; see Sec. III B.)

As a test of our wave functions we have performed calculations of the ionization potentials and the scalar polarizabilities α of the ground state for each atom. These can then be compared with the available experimental data. It is easy to calculate the polarizability

$$\alpha=-2\sum_M \frac{|\langle N|D_z|M\rangle|^2}{E_N-E_M} \tag{11}$$

by replacing the operator H_W in Eq. (8) by the dipole operator D_z.

Below we outline the V^N and V^{N-2} calculations.

A. V^N approximation

In the V^N calculation, we can write the atomic EDM induced by the Schiff moment as

$$d=2\sum_n \langle\delta n_W|d_z|n\rangle, \tag{12}$$

where the sum runs over the core states $|n\rangle$, d_z is the single-particle dipole operator, and $|\delta n_W\rangle$ denotes the correction to the state $|n\rangle$ due to the P,T-odd Hamiltonian h_W. The correction $|\delta n_W\rangle$ can be expressed as

$$|\delta n_W\rangle=\sum_\alpha \frac{\langle\alpha|h_W|n\rangle}{\epsilon_n-\epsilon_\alpha}|\alpha\rangle, \tag{13}$$

where $|\alpha\rangle$ corresponds to an excited state. It is found by solving the equation

$$(h_0-\epsilon_n)|\delta n_W\rangle=-h_W|n\rangle. \tag{14}$$

[Of course, calculating the correction to $|n\rangle$ due to the electric dipole ($E1$) field and taking the matrix element of the weak Hamiltonian is equivalent.]

To include polarization of the core due to the fields h_W and d_z, we need only include the polarization due to one field, e.g., by replacing h_W in Eq. (14) by $h_{\tilde{W}}=h_W+\delta V_W$. (The correction δV_W accounts for the change in the self-consistent potential V^N due to the modification of the wave functions. For more on the time-dependent Hartree-Fock (TDHF) method or random-phase approximation (RPA) with exchange, see, e.g., Ref. [18].) This is because all the states $|n\rangle$ belong to the core (that is, they are included in the Hartree-Fock potential), so $\Sigma_n\langle \delta n_{\tilde{W}}|d_z|n\rangle=\Sigma_n\langle \delta n_W|d_z+\delta V_d|n\rangle$.

The scalar polarizabilities are easily calculated by replacing the correction $|\delta n_W\rangle$ due to the P,T-odd field by the correction $|\delta n_d\rangle$ due to the $E1$ field.

B. V^{N-2} approximation

Hg and Ra can be considered as atoms with two valence electrons above closed cores, $[1s\cdots 5d^{10}]$ and $[1s\cdots 5d^{10}6s^2 6p^6]$, respectively. The calculations performed in the V^{N-2} approximation correspond to the CI +MBPT method developed in Refs. [16,17]. An effective Hamiltonian is constructed for the valence electrons, which is formed using MBPT for the interaction of the valence electrons with the core. In this way the correlations between the valence electrons $6s^2$ (for Hg) and $7s^2$ (Ra) are treated using the CI method, while the valence-core correlations are treated using MBPT. (For more on this procedure, please refer to Refs. [16,17].)

Note that calculations in the CI+MBPT method can be performed in a different potential, even in V^N. The final accuracies for V^{N-2} and V^N potentials are comparable; however, calculations in V^{N-2} for Hg and Ra are somewhat simpler — there are no subtraction diagrams, see, e.g., Ref. [16].

For Hg the CI states were formed from the one-electron basis sets $1s$-$12s$, $2p$-$13p$, $3d$-$14d$, and $4f$-$15f$. The core $1s$-$5d$ and the states $6s$, $6p$, $6d$, $5f$ were HF ones, while the rest were virtual orbitals. The virtual orbitals were formed using the recurrent procedure, similar to that used in Ref. [19]. Full CI was made for the two valence electrons. Two sets of virtual orbitals were constructed to test the saturation of the CI space. The results appeared to be very close, so we concluded that saturation was reached.

MBPT calculations generally require more basis functions for high energies and more partial waves, so the number of virtual orbitals was increased to construct the MBPT basis set: it included $1s$-$21s$, $2p$-$22p$, $3d$-$23d$, $4f$-$21f$, $5g$-$18g$, and $6h$-$19h$. For Ra the CI basis set was somewhat larger and included orbitals $1s$-$15s$, $2p$-$16p$, $3d$-$17d$, and $4f$-$17f$. The extended basis set for MBPT was similar to that used for Hg.

III. RESULTS

A. Xenon and radon

The binding energies of the ground state of Xe and Rn performed in the HF approximation in the V^N potential are presented in Table I. It is seen that the Hartree-Fock calculation serves as a good approximation for the noble gases, with the calculated and experimental ionization potentials in disagreement only at the level of 1%.

TABLE I. Ionization potentials of Xe, Rn, Hg, Ra, and Pu. The calculated values are obtained from the Hartree-Fock approximation in the V^N potential. The experimental values are presented in the last column. Units are cm^{-1}.

Atom	HF	Experiment [a]
Xe	96525	97834.4
Rn	84285	86692.5
Hg	71996	84184.1
Ra	36485	42577.35
Pu	41463	48890(200) [b]

[a] From Ref. [20] unless otherwise stated.

[b] Ref. [21].

The ground-state scalar polarizabilities α are listed in Table II. Core polarization increases α only very slightly (about 1%) from the Hartree-Fock value. The value for Xe is in perfect agreement with the experimental value.

Our results for the electric dipole moments induced in Xe and Rn due to the nuclear Schiff moment are presented in Table III. The effect of core polarization increases the value of the EDMs in Xe and Rn by about 30%. Due to the increase in Z, the EDM induced in Rn is ten times larger that that induced in its lighter analog Xe.

A calculation for the Xe EDM has previously been performed at the HF level, with the result $d(^{129}\mathrm{Xe})=0.27\times 10^{-17}S(e\ \ \mathrm{fm}^3)^{-1}e\ \ \mathrm{cm}$ [9]. Our HF result is in agreement with this value.

No direct atomic calculation has previously been performed for the atomic EDM of Rn, or for any of the remaining atoms in this work. The estimate for Rn, based on extrapolation of the HF result from Xe, is $(^{223}\mathrm{Rn})=2.0\times 10^{-17}S(e\ \ \mathrm{fm}^3)^{-1}e\ \ \mathrm{cm}$ [10]. This value is not significantly different from our HF value.

TABLE II. Scalar polarizabilities of Xe, Rn, Hg, Ra, and Pu obtained in HF and TDHF approximations in V^N. Experimental values are presented in the last column (a.u.).

Atom	HF	TDHF	Experiment [a]
Xe	26.87	26.97	27.06
Rn	34.42	35.00	
Hg	40.91	44.92	34
Ra	204.2	297.0	
Pu	147.3	201.3	

[a] Ref. [22].

B. Mercury and radium

The ionization potentials of Hg and Ra performed in the V^N approximation are listed in Table I alongside the results

TABLE III. Electric dipole moments d induced in Xe, Rn, Hg, Ra, and Pu by the nuclear Schiff moment S. We present results obtained using the HF and TDHF approximations in the V^N potential. Units are 10^{-17} $[S/(e\ \text{fm}^3)]e$ cm.

Atom	HF	TDHF
Xe	0.289	0.378
Rn	2.47	3.33
Hg	-1.19	-2.97
Ra	-1.85	-8.23
Pu	-2.66	-10.9

for the noble gases. The deviation from experiment for Hg and Ra is $\sim -14\%$, indicating the importance of correlations.

The ground-state polarizabilities for Hg and Ra performed in the V^N approximation are listed in Table II. We see here that core polarization increases α, from the HF value, by about 10% for Hg. The effect of core polarization on Ra is more significant, with an increase of 45%. For both Hg and Ra, essentially the entire HF and TDHF results for α arise from the $6s^2$ and $7s^2$ contributions, respectively. We see that the result for Hg is strongly overestimated (about 30% higher than the experimental value). This indicates that the V^N calculations produce wave functions that are very poor at large distances from the nucleus for systems with two electrons above closed shells.

Our results for the EDMs induced in Hg and Ra by the nuclear Schiff moment, in the V^N approximation, are presented in Table III. The effects of core polarization in Hg and Ra are dramatic, with the EDM increased by factors of 2.5 and 4.5, respectively. The dominating contributions to the EDMs of Hg and Ra come from $6s^2$ and $7s^2$, respectively. These contributions are larger than, and of opposite sign to, the contributions from the core. The instability of the results motivates us to study these atoms more carefully, taking into account valence-valence and valence-core correlations.

The V^{N-2} approximation is more appropriate for calculations of two-electron atoms. The results of the calculations for the binding energies of relevant states of Hg and Ra are presented in Table IV. The calculated value for the removal energy of both s electrons for Hg and Ra deviates from experiment by about -10% at the CI level, and is then improved to $+2\%$ with MBPT included. This is a significant improvement compared to the V^N result. The accuracy of the calculations of the energies of other states is not as good. These states are not directly relevant to our calculations of the EDMs, however, we have presented them in the table as an indication of the accuracy of wave functions produced at various stages of the CI+MBPT method. At the CI stage of the calculations the energies of the *ss* and *sp* states are underestimated by 10–20 %. Note that for *sd* states the energies are overestimated by about the same amount. With MBPT corrections included, the accuracy of the energies improves to 2–9 %.

Results of the calculations of the ground-state scalar polarizabilities α of Hg and Ra in the V^{N-2} approximation are listed in Table V. We see that for Hg at the CI level of calculation α is strongly overestimated (by about 60%). However, both the MBPT and TDHF corrections reduce the value so that the final result is in excellent agreement with experiment (compare 32.99 a.u. with 34 a.u. [22]). We see that for Ra the MBPT and TDHF corrections bring about a cancellation of the CI value similar to what we see for Hg.

The results of the calculations of the EDM of Hg and Ra in the V^{N-2} approximation are presented in Table VI. We see here that for Hg the MBPT and TDHF corrections increase the CI approximation by a factor of 2. The corrections for Ra are huge, the final value being 3.6 times that of the CI value.

TABLE IV. Binding energies of low states of Hg and Ra calculated in the V^{N-2} approximation. The removal energy for both $6s$ electrons for Hg and both $7s$ electrons for Ra is presented in the first row of each respective atom. Energies of excited states are presented relative to the ground state. Units are cm^{-1}.

Atom	State		CI	+MBPT	Experiment [a]
Hg	$6s^2$	$^1S_0$	207659	240912	235464
	$6s6p$	$^3P_0$	29336	40012	37645.080
		$^3P_1$	31009	41753	39412.300
		$^3P_2$	34794	46776	44042.977
		$^1P_1$	46142	55395	54068.781
	$6s7s$	$^3S_1$	51460	65044	62350.456
		$^1S_0$	54187	67090	63928.243
Ra	$7s^2$	$^1S_0$	115318	127248	124419.66
	$7s6d$	$^3D_1$	15910	14012	13715.85
		$^3D_2$	16067	14465	13993.97
		$^3D_3$	16625	15921	14707.35
	$7s7p$	$^3P_0$	10424	14268	13078.44
		$^3P_1$	11289	15159	13999.38
		$^3P_2$	13535	17937	16688.54
		$^1P_1$	18835	21663	20715.71

[a] Ref. [20].

TABLE V. The scalar polarizabilities α for Hg and Ra calculated in the potential V^{N-2}. The valence and core contributions are separated into different rows and their sum is presented. The HF result, with CI included into the valence orbital contribution, is presented in the first column of results. In the next columns the Brueckner and TDHF contributions are added. Units are (a.u.)

Atom	Contribution	CI	+MBPT	+TDHF
Hg	$6s^2$	46.54	36.06	25.69
	Core	8.05	8.05	7.30
	Sum	54.59	44.11	32.99
Ra	$7s^2$	321.4	260.7	218.2
	Core	13.6	13.6	11.8
	Sum	335.0	274.3	229.9

Even though these corrections are so large, it is certainly interesting that the result for $d(\mathrm{Hg})$ in the V^{N-2} approximation (with correlations) is in agreement ($\approx 10\%$) with the simple calculation in the V^N approximation; compare the results of Hg and Ra in Table VI with those in Table III. This coincidence of the results is an argument in favor of the stability of the final values. Also it indicates that the simple V^N approximation gives accurate results for EDM calculations of closed-shell atoms. From a consideration of the results, we take as our final value for the EDM in Hg induced by S,

$$d(^{199}\mathrm{Hg}) = -2.8\times 10^{-17}\left(\frac{S}{e\ \ \mathrm{fm}^3}\right) e\ \ \mathrm{cm}. \tag{15}$$

We take as our final value for the EDM induced in Ra,

$$d(^{225}\mathrm{Ra}) = -8.5\times 10^{-17}\left(\frac{S}{e\ \ \mathrm{fm}^3}\right) e\ \ \mathrm{cm}. \tag{16}$$

We can compare these new values for Hg and Ra with estimates calculated in Refs. [6] and [10], respectively. The value for Hg, $d(^{199}\mathrm{Hg}) = -4\times 10^{-17} S(e\ \ \mathrm{fm}^3)^{-1} e$ cm, was obtained indirectly from an atomic calculation [7] of the Hg EDM induced by the P,T-odd electron-nucleon tensor interaction. The radium estimate, $d(^{225}\mathrm{Ra}) = -7\times 10^{-17} S(e\ \ \mathrm{fm}^3)^{-1} e$ cm, was found by extrapolation of the Hg EDM estimate, taking account of the enhancement due to higher atomic number Z.

TABLE VI. The EDMs induced in Hg and Ra calculated in the V^{N-2} potential. The valence and core contributions are separated into different rows and their sum is presented. The HF result, with CI included into the valence orbital contribution, is presented in the first column of results. In the next columns the Brueckner and TDHF contributions are added. Units are 10^{-17} $[S/(e\ \ \mathrm{fm}^3)]e$ cm.

Atom	Contribution	CI	+MBPT	+TDHF
Hg	$6s^2$	−1.90	−2.77	−3.04
	Core	0.63	0.63	0.34
	Sum	−1.26	−2.14	−2.70
Ra	$7s^2$	−4.03	−6.08	−10.10
	Core	1.62	1.62	1.40
	Sum	−2.41	−4.46	−8.70

C. Plutonium

We performed a simple calculation for Pu. We basically used the V^N approximation. The unfilled f shell was accounted for by weighting the corresponding angular coefficients according to the occupation number of the shell. In calculations of the atomic EDM, we can expect that Pu behaves in a similar way to Ra and Hg (there is a closed s shell, $7s^2$, and an open f shell). The f shell does not contribute to the EDM at the HF level (the f shell does not penetrate the nucleus due to the centrifugal barrier, and so does not contribute to the Schiff matrix element). However, it can contribute due to core polarization. Because the V^N calculations for Ra and Hg turned out to be sufficient, we expect that the same is true for Pu.

Our result for the ionization potential of Pu (see Table I) deviates by −15% from experiment. The polarizability α is listed in Table II. As we saw for Hg and Ra, essentially the entire value is due to the $7s^2$ contribution. The results for the EDM of Pu are presented in Table III. These values are essentially due to the contribution of the $7s$ electrons, which are of opposite sign to the contributions of the core and f electrons. With core polarization, the contribution of the f electrons amounts only to 10% of the final value. The effect of core polarization increases the HF value four times. This is similar to what we see in Ra.

An estimate for the Pu EDM was performed in Ref. [13] by extrapolation from the estimate of the Hg EDM, $d(^{239}\mathrm{Pu}) = -10\times 10^{-17} S(e\ \ \mathrm{fm}^3)^{-1} e$ cm. Our calculation is in agreement with this estimate.

D. TlF molecule

All molecular calculations deal with the following matrix element:

$$X = -\frac{2\pi}{3}\langle \Psi_0 | [\boldsymbol{\nabla}\cdot\mathbf{n}, \delta(\mathbf{R})] | \Psi_0\rangle, \tag{17}$$

TABLE VII. Final results for the atomic EDMs induced by the nuclear Schiff moment. These are compared with the simple calculations/estimates of previous works. Units are 10^{-17} $[S/(e\ \ \mathrm{fm}^3)]e$ cm.

Atom	Other works	This work
Xe	0.27[a]	0.38
Rn	2.0 [b]	3.3
Hg	−4.0 [c]	−2.8
Ra	−7.0 [b]	−8.5
Pu	−10[d]	−11

[a] Ref. [9].
[b] Ref. [10].
[c] Ref. [6].
[d] Ref. [13].

TABLE VIII. Limits on P,T-violating parameters in the hadronic sector extracted from ^{199}Hg [Eq. (25)] compared with the best limits from other experiments. We omit the signs of the central points. Errors are experimental. Some relevant theoretical works are presented in the last column.

P,T-violating term	Value	System	Experiment	Theory
Neutron EDM $d_{\rm n}$	$(17\pm 8\pm 6)\times 10^{-26}e$ cm	^{199}Hg	[1]	[25,3]
	$(1.9\pm 5.4)\times 10^{-26}e$ cm	Neutron	[27]	
	$(2.6\pm 4.0\pm 1.6)\times 10^{-26}e$ cm	Neutron	[28]	
Proton EDM $d_{\rm p}$	$(1.7\pm 0.8\pm 0.6)\times 10^{-24}e$ cm	^{199}Hg	[1]	[25,3,9]
	$(17\pm 28)\times 10^{-24}e$ cm	TlF	[14]	[25,15]
$\eta_{\rm np} i(G/\sqrt{2})\bar{p}p\bar{n}\gamma_5 n$	$\eta_{\rm np}=(2.7\pm 1.3\pm 1.0)\times 10^{-4}$	^{199}Hg	[1]	[6]
$g^0_{\pi NN}$	$(3.0\pm 1.4\pm 1.1)\times 10^{-12}$	^{199}Hg	[1]	[3]
QCD phase $\bar{\theta}$	$(1.1\pm 0.5\pm 0.4)\times 10^{-10}$	^{199}Hg	[1]	[26,3]
	$(1.6\pm 4.5)\times 10^{-10}$	Neutron	[27]	[29]
	$(2.2\pm 3.3\pm 1.3)\times 10^{-10}$	Neutron	[28]	[29]
CEDMs $\tilde{d}$ and	$e(\tilde{d}_{\rm d}-\tilde{d}_{\rm u})=(1.5\pm 0.7\pm 0.6)\times 10^{-26}e$ cm	^{199}Hg	[1]	[30]
EDMs d of quarks	$e(\tilde{d}_{\rm d}+0.5\tilde{d}_{\rm u})+1.3d_{\rm d}-0.3d_{\rm u}$			
	$=(3.5\pm 9.8)\times 10^{-26}e$ cm	Neutron	[27]	[31]
	$=(4.7\pm 7.3\pm 2.9)\times 10^{-26}e$ cm	Neutron	[28]	[31]

where Ψ_0 is the ground-state wave function and $\mathbf{n}$ is the unit vector along the molecular axis. It is clear that this matrix element is related to the contact form of the Schiff moment potential given by Eq. (6):

$$-4\pi\langle\Psi_0|\mathbf{S}\cdot[\boldsymbol{\nabla},\delta(\mathbf{R})]|\Psi_0\rangle=6X\mathbf{S}\cdot\mathbf{n}. \qquad (18)$$

It is not difficult to find the correction coefficient k_1 that accounts for the difference between the contact form of the Schiff moment interaction (6) and the more accurate expression (7).

The latest molecular calculation [15] accounted for the correlations between valence electrons, but neglected core-valence correlations. We saw above that the latter appears to be very important in atomic calculations. The most important core-valence correlations in molecules with one heavy atom are of the same nature as in atoms and can be accounted for by an atomic calculation for the heavy atom [23]. The valence space in the calculation [15] included $5s$, $5p$, $5d$, $6s$, and $6p$ electrons, leaving a rather small core $[1s^2\cdots 4f^{14}]$. For such a compact and rigid core the dominant correlation correction is from the TDHF for the Schiff potential. We account for this correction by the coefficient k_2.

The final expression, which includes the two corrections discussed above, has the form,

$$\langle\Psi_0|H_W|\Psi_0\rangle=6\,k_1\,k_2\,X\mathbf{S}\cdot\mathbf{n}. \qquad (19)$$

The dominant contribution to the molecular matrix element (19) comes from atomic matrix elements for Tl of the form

$$\langle ns|H_W|mp_z\rangle=\frac{1}{3}\langle ns_{1/2}|H_W|mp_{1/2}\rangle+\frac{2}{3}\langle ns_{1/2}|H_W|mp_{3/2}\rangle. \qquad (20)$$

This allows us to approximate the correction coefficients: $k_i\approx\frac{1}{3}k_i(s_{1/2},p_{1/2})+\frac{2}{3}k_i(s_{1/2},p_{3/2})$. Within this method we calculated the coefficients k_i for the Tl atom and found

$$k_1=0.89;\quad k_2=1.10. \qquad (21)$$

We see that both corrections are relatively small and, therefore, our simple model calculation is justified. Note that the core here is much smaller than in our previous atomic calculations. Because of that the TDHF correction here is only 10%. Other core-valence correlations tend to decrease more rapidly with the energy of the core orbitals, and therefore are less important here. Using the result $X=7635$ a.u. from Ref. [15] and Eqs. (19) and (21) we get as our final answer

$$\langle\Psi_0|H_W|\Psi_0\rangle=4.47\times 10^4(\mathbf{S}\cdot\mathbf{n})\ \text{a.u.} \qquad (22)$$

IV. CONCLUSION

The most accurate measurement of the P,T-odd spin-axis interaction in the TlF molecule [14] combined with Eq. (22) gives the limit

$$S(^{205}\text{Tl})=(69\pm 111)\times 10^{-12}e\ \text{fm}^3. \qquad (23)$$

In Table VII we present our final results for the atomic EDMs induced by the Schiff moments S alongside the values of previous calculations/estimates.

The limit on the Schiff moment of ^{129}Xe [Eq. (3), Table VII] is

$$S(^{129}\text{Xe})=(184\pm 868\pm 26)\times 10^{-12}e\ \text{fm}^3. \qquad (24)$$

Our result for Hg puts new constraints on the fundamental P,T-violating parameters. Comparing Eqs. (1) and (15) we obtain for the limit on the nuclear Schiff moment

$$S(^{199}\text{Hg})=(3.8\pm 1.8\pm 1.4)\times 10^{-12}e\ \text{fm}^3. \qquad (25)$$

Comparison of the calculated and measured values of the ionization potentials and polarizabilities as well as a comparison of the calculations in two completely different approximations (in V^N and V^{N-2}) indicate that the error of the atomic calculations probably does not exceed 20%.

In general, the Schiff moment can be induced from a number of P,T-violating mechanisms: due to a permanent EDM of an unpaired nucleon or due to the P,T-violating nucleon-nucleon interaction. Mercury has an unpaired neutron, and so in the shell model its Schiff moment can arise due to the EDM of the neutron. The contribution to the Schiff moment of Hg from a proton EDM can be estimated by comparing the experimental value of the magnetic moment of Hg with that estimated by the nuclear shell model (this allows us to estimate the contribution of proton configurations with unpaired spin); see Ref. [9]. The dominant mechanism for the production of a Schiff moment is the P,T-odd nucleon-nucleon interaction η [24]. Due to this mechanism, the P,T-odd field of the unpaired neutron excites core protons that contribute to the Schiff moment [6]. The magnitude of this interaction is characterized by the dimensionless constant η_{np}. In Table VIII we present the limits on these parameters extracted from Hg. These are compared in the table with the best limits from other experiments. Also presented are limits on P,T-violating parameters at the more fundamental level, the P,T-odd pion-nucleon coupling constant $\overline{g}^0_{\pi NN}$, the P,T-odd QCD phase $\overline{\theta}$, and the chromoelectric dipole moments (CEDMs) and EDMs of quarks. We see here that the limits extracted from the Hg measurement are stronger than those extracted from direct neutron EDM experiments.

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