

Semiempirical calculations of P - and P, T -odd effects in diatomic molecules-radicals

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A semiempirical method of computing the matrix elements of the P - and P, T -odd electron-nucleus interaction operators for diatomic molecules-radicals is proposed. The molecular parameters entering into these matrix elements are extracted from the experimental data on the hyperfine and Zeeman structures of the electronic ground state of the molecules. The accuracy of such a calculation is comparable to the accuracy of the calculations for the heavy atoms. The numerical results given for the molecules BaF, HgH, and HgF confirm the earlier published estimates for the magnitudes of the P - and P, T -odd effects. Thus, the outlook for the use of diatomic molecules-radicals to study P - and P, T -odd interactions and, in particular, to measure the anapole moments of heavy nuclei is made brighter.

1. INTRODUCTION

The effects of the nonconservation of spatial parity P , as well as those of the simultaneous nonconservation of both spatial P and temporal T parities in diatomic molecules have for a long time been the subject of discussion.¹⁻⁶ It was shown in the very first investigations¹⁻³ that, in molecules-radicals possessing a nonzero mean electron angular momentum, the P - and P, T -odd interactions can mix the rotational levels of one electronic term. The rotational-level spacing is four to five orders of magnitude smaller than the characteristic atomic-level spacing, and this leads to the enhancement of the P - and P, T -odd effects.

The interactions in question are described by single-electron operators, and only electrons with uncompensated spins contribute to their matrix elements. This made the P - and P, T -odd effect calculations carried out in Refs. 4-6 for metal-halogen type molecules, which have one unpaired electron, possible. But model unpaired-electron wave functions were used in these calculations, and there is no clear indication of their accuracy.

We propose to use for the determination of the necessary electron wave-function parameters the experimental molecular-hyperfine-structure and electron- g -factor data. This allows us in a number of cases to achieve an accuracy comparable to that achieved in similar calculations for atoms.⁷

For the sake of brevity, we limit ourselves to the consideration of only two types of interaction: the P -odd interaction H_P stemming from the electromagnetic electron-nuclear anapole moment interaction^{5,8} and the P, T -odd electron-nucleon neutral current interaction $H_{P, T}$.^{2,3,6} Another P, T -odd-interaction type that arises as a result of the presence of P, T -odd nuclear forces is considered in Ref. 4.

The operators H_P and $H_{P, T}$ have, in atomic units ($\hbar = e = m = 1$), the form

$$H_P = \frac{G\alpha}{2^h} \kappa_P \mathbf{I} \alpha n(r), \quad (1)$$

$$H_{P, T} = i \frac{G\alpha}{2^h} Z \kappa_{P, T} \gamma_0 \gamma_5 n(r). \quad (2)$$

In these expressions $G = 10^{-5}/m_p^2$ is the Fermi constant; α is the fine-structure constant; α, γ_0 , and γ_5 are the Dirac matrices ($\alpha = \gamma_0 \gamma$); \mathbf{I} is the nuclear spin; $n(r)$ is the nuclear density normalized to unity; Z is the nuclear charge; κ_P and $\kappa_{P, T}$ are dimensionless constants:

$$\kappa_P = \frac{(-1)^{l+h-1} (I+1/2)}{I(I+1)} \kappa_a,$$

(where κ_a is the anapole-moment constant defined in Ref. 8 and l is the orbital angular momentum of the valence nucleon); and $\kappa_{P, T}$ can be expressed in terms of the proton-electron and neutron-electron interaction constants as follows:

$$\kappa_{P, T} = \kappa_{P, T}^p + \frac{A-Z}{Z} \kappa_{P, T}^n.$$

Most often, the ground state of a molecule having an odd number of electrons is either the $^2\Sigma$ or the $^2\Pi_{1/2}$ state. It is precisely these two cases that we consider below.

2. THE ELECTRONIC MATRIX ELEMENTS OF THE OPERATORS H_P AND $H_{P, T}$

It is well known that the matrix elements of H_P and $H_{P, T}$ must be computed with the Dirac wave functions, since the Schrödinger wave functions approximate the Dirac functions well only at points far from the nucleus. Therefore, the molecular function should be constructed, using the relativistic atomic orbitals (AO). It then turns out that the only nonzero matrix elements are those connecting the $ns_{1/2}$ and $n'p_{1/2}$ AO, for which the semiclassical approximation yields the following expressions⁷:

$$\langle s_h | H_P | p_h \rangle = \frac{iG\alpha^2 Z^2 R}{2^h \pi} \frac{2\gamma_1 + 1}{3} k_s k_i \kappa_P \mathbf{I}, \quad (3)$$

$$\langle s_h | H_{P, T} | p_h \rangle = \frac{G\alpha^2 Z^2 R}{2^h \pi} \gamma_1 k_s k_i \kappa_{P, T}. \quad (4)$$

The matrix element (3) has not been integrated fully over the angular variables: \mathbf{j} is the electron angular-momentum operator.

The coefficients k_i can be expressed in terms of the Z_c of the atomic core (for an atom $Z_c = 1$, while for a positive ion $Z_c = 2$) and the effective principal numbers

$$k_i = Z_c / (v_i)^{1/2}. \quad (5)$$

For the s electron $i = s$ and for the P_j electron $i = 2j$. The relativistic factor R has the form

$$R = [2/\Gamma(2\gamma_1 + 1)]^2 (2Zr_n)^{2\gamma_1 - 2}, \quad (6)$$

where r_n is the nuclear radius and

$$\gamma_1 = [(j + 1/2)^2 - (\alpha Z)^2]^{1/2}. \quad (7)$$

The deviation of R from unity is due to the fact that the relativistic and nonrelativistic functions on the nucleus are different. For $Z = 30$ we have $R \sim 1.5$, while for $Z = 80$ $R \sim 10$.

Let us proceed to consider the molecular matrix elements of H_P and $H_{P,T}$. We shall assume that there is only one heavy atom in the molecule. Then because of the strong dependence of these elements on Z , we can neglect the contribution of the AO of the light atom. That part of the molecular-function expansion which is of interest to us can be written in the form

$$|\omega\rangle = a|s_{1/2}, \omega\rangle - 2\omega b|p_{1/2}, \omega\rangle + c|p_{3/2}, \omega\rangle, \quad (8)$$

where $\omega = \pm 1/2$ is the component of the electron angular momentum \mathbf{j} along the axis of the molecule; $|s_{1/2}, \omega\rangle$ and $|p_{j}, \omega\rangle$ are the relativistic AO of the heavy atom; and a , b , and c are the expansion coefficients.

The function (8) does not possess a definite orbital angular-momentum component Λ along the axis of the molecule. But it may go over at points far from the nucleus into a function having a definite Λ value. In this case $c = 2^{1/2}b$ when $\Lambda = 0$ (i.e., for the ${}^2\Sigma$ state), while $a = 0$ and $c = -2^{-1/2}b$ for $|\Lambda| = 1$ (the ${}^2\Pi_{1/2}$ state). The standard spectroscopic selection rules are determined precisely by this region, and therefore the use of the quantum number Λ in their case is justified. In contrast, the quantum number Λ cannot be used in the case of contact interactions of the H_P and $H_{P,T}$ type.

The molecular matrix elements of H_P and $H_{P,T}$ can be obtained from (3), (4), and (8):

$$\langle \omega' | H_P | \omega \rangle = -i W_{P,T} 2\omega \delta_{\omega', -\omega} \langle \omega' | \mathbf{j} | \omega \rangle, \quad (9)$$

$$W_P = 2(a k_s) (b k_1) \frac{G \alpha^2 Z^2 R}{2^{1/2} \pi} \frac{2\gamma_1 + 1}{3}, \quad (10)$$

$$\langle \omega' | H_{P,T} | \omega \rangle = W_{P,T} \gamma_{\omega', \omega}, \quad (11)$$

$$W_{P,T} = -2(a k_s) (b k_1) \frac{G \alpha^2 Z^2 R}{2^{1/2} \pi} \gamma_1. \quad (12)$$

In the case of the Σ states the rotational motion of the molecule mixes the states $|\pm \omega\rangle$ (case b coupling). In this case we should leave the electronic matrix elements unaveraged over the angular variables. Let us, taking account of the fact that for a pure Σ state

$$\langle \omega' | \mathbf{j} | \omega \rangle = \langle \omega' | s | \omega \rangle,$$

rewrite the ω -dependent parts of the expressions (9) and (11) in the form of matrix elements of the spin operators:

$$2\omega \delta_{\omega', -\omega} \langle \omega' | \mathbf{j} | \omega \rangle = i \langle \omega' | \mathbf{n} [s \times \mathbf{I}] | \omega \rangle,$$

$$\omega \delta_{\omega', \omega} = \langle \omega' | s \times \mathbf{n} | \omega \rangle,$$

where \mathbf{n} is the unit vector oriented along the axis of the molecule. Omitting the averaging, we obtain

$$\langle \Sigma | H_P | \Sigma \rangle = W_{P,T} \gamma_{\Sigma} \mathbf{n} [s \times \mathbf{I}], \quad (13)$$

$$\langle \Sigma | H_{P,T} | \Sigma \rangle = W_{P,T} \gamma_{\Sigma} \mathbf{n} s. \quad (14)$$

The expression (13) coincides with the one obtained in Ref. 5.

3. LEVELS WITH OPPOSITE PARITIES

A. Molecules in the ${}^2\Sigma$ -state. Practically all the molecules that have ${}^2\Sigma$ as their ground state pertain to Hund's case- b coupling, i.e., are described by the quantum numbers Λ , S , and N , where N is the rotational angular momentum of the molecule. The parity of the electronic-rotational level (for the Σ^+ terms) is $p = (-1)^N$, and the rotational energy

$$E_r = BN(N+1).$$

Two types of coupling are possible in the case of heavy molecules when we consider the nuclear spin I (Ref. 9):

$$\text{case-}b_J \text{ coupling: } J = N + S, \quad F = J + I$$

$$\text{case-}b_{F_1} \text{ coupling: } F_1 = I + S, \quad F = F_1 + N.$$

The type of coupling is determined by the relation between the isotropic hyperfine structure constant A and the spin-doubling constant γ , which respectively determine the spacing of the levels with different F_1 and J .

The effective spin-rotation Hamiltonian describing the two coupling cases can be written in the form

$$H_{sr} = BN(N+1) + AIS + \gamma NS. \quad (15)$$

It is the eigenvalues of this Hamiltonian that determine the separation of levels with opposite parities.

B. Molecules in the ${}^2\Pi_{1/2}$ state. Heavy molecules in the ${}^2\Pi_{1/2}$ state pertain to a type of coupling that is intermediate between Hund's case a and case c couplings. The rotational Hamiltonian in this case has the form

$$H_r = B(\mathbf{J} - \mathbf{J}^e)^2 = B\mathbf{J}^2 - 2B\mathbf{J}^e \mathbf{J} + B\mathbf{J}^{e2}, \quad (16)$$

where \mathbf{J}^e and \mathbf{J} are the electron and total angular momenta. This Hamiltonian is diagonal in the space of the functions^{2,6}

$$|JM\omega\rangle = 2^{-1/2} (|\omega\rangle |JM\omega\rangle + (-1)^{J+1/2} p |-\omega\rangle |JM-\omega\rangle), \quad (17)$$

where $|J, M, \omega\rangle$ is the rotational wave function. The rotational energy

$$E_{J,p} = BJ(J+1) + (-1)^{J-1/2} p 2B \langle \omega | J_x^e | -\omega \rangle (J+1/2), \quad (18)$$

and the distance between levels with opposite parities¹¹

$$\Delta = E_{J,-} - E_{J,+} = (-1)^{J+1/2} 4B \langle \omega | J_x^e | -\omega \rangle (J+1/2). \quad (19)$$

4. SEMIEMPIRICAL WAVE FUNCTIONS OF MOLECULES

For many molecules with a $^2\Sigma$ ground state the electronic wave functions are known from EPR-spectrum measurements.¹⁰⁻¹³ The point is that, for linear molecules, the hyperfine structure constant A and the electronic g factors are axially symmetric second-rank tensors. EPR allows us to determine all the elements of these tensors. We thus obtain six constants:

$$A_{\parallel}^a, A_{\perp}^a, A_{\parallel}^b, A_{\perp}^b, g_{\parallel}, g_{\perp}$$

where a and b number the nuclei. If we know the hyperfine structure constants for the atoms forming the molecule, then we can determine from the hyperfine structure four coefficients of the AO entering into the molecular wave function. Since for the pure Σ state

$$g_{\parallel} = g_{\perp} = g_0 = 2.0023,$$

the g tensor carries information about the admixture of the Π state by the spin-orbit interaction. This admixture leads to the spin doubling, with¹⁰

$$\gamma = 2B(g_0 - g_{\perp}). \quad (20)$$

Let us compute A_{\parallel} and A_{\perp} for the electron wave function (8). The hyperfine interaction operator has the form

$$H_{hf} = \frac{g_n \alpha}{2m_p} \mathbf{I} \frac{[\alpha \otimes \alpha]}{r^3}, \quad (21)$$

where g_n is the nuclear g factor. According to Ref. 14,

$$\langle s | H_{hf} | s \rangle = h_s k_s^2 \mathbf{I}_j, \quad \langle p_j | H_{hf} | p_j \rangle = h_{2j} k_{2j}^2 \mathbf{I}_j, \quad (22)$$

$$\langle p_{\omega}, \omega' | H_{hf} | p_{\omega}, \omega \rangle = (-1)^{j-\omega'+\omega} \begin{pmatrix} 3/2 & 1 & 1/2 \\ -\omega' & q & \omega \end{pmatrix} h_{1,3} k_1 k_3 I_{-q},$$

where

$$h_s = 8/3 \Omega R_{1s}, \quad h_{2j} = [2/3j(j+1)] \Omega R_{2j}, \quad (23)$$

$$h_{1,3} = 3^{-1/2} \Omega R_{1,3}, \quad \Omega = \alpha^2 Z g_n / 2m_p,$$

while the relativistic R factors have the form

$$R_{2j} = \frac{4j(j+1/2)(j+1)}{\gamma_{2j}(4\gamma_{2j}^2-1)}, \quad R_{1,3} = -\frac{4 \sin(\pi(\gamma_3-\gamma_1))}{\pi(\alpha Z)^2}. \quad (24)$$

The matrix elements of the operator H_{hf} , like those of the operators H_P and $H_{P,T}$, are proportional to the products $k_i k_j$; therefore, we can eliminate these factors, going over in the expressions for the molecular functions from the coefficients attached to the AO to the following new parameters:

$$\alpha = ak_s, \quad \beta = bk_1, \quad \gamma = ck_2. \quad (25)$$

In so doing we not only make the computation of the k_i unnecessary, but also take account of a number of corrections to the H_{hf} matrix elements, which also enter into the H_P and $H_{P,T}$ matrix elements.^{15,16}

Writing the H_{hf} -operator matrix in the subspace of the functions $|\pm \omega\rangle$ in the standard form

$$\begin{vmatrix} \frac{1}{2} A_{\parallel} I_z & 2^{-1/2} A_{\perp} I_{-1} \\ -2^{-1/2} A_{\perp} I_1 & -\frac{1}{2} A_{\parallel} I_z \end{vmatrix}, \quad (26)$$

we obtain from (8), (22), and (25) the equations

$$h_s \alpha^2 - 1/3 (h_1 \beta^2 - 5h_3 \gamma^2 + (8/3)^{1/2} h_{1,3} \beta \gamma) = A, \quad (27)$$

$$2h_1 \beta^2 - h_3 \gamma^2 - (8/3)^{1/2} h_{1,3} \beta \gamma = 3A_d, \quad (28)$$

where $A = (A_{\parallel} + 2A_{\perp})/3$ is the isotropic hyperfine structure constant and $A_d = (A_{\parallel} - A_{\perp})/3$ is the dipole constant.

In the general case Eqs. (27) and (28) are not sufficient for the determination of the parameters α and β of interest to us. For the pure $^2\Sigma$ state

$$\gamma = 2^{1/2} \beta (k_3/k_1) = 2^{1/2} \beta q, \quad (29)$$

and from (28) we obtain

$$2[h_1 - h_3 q^2 - (2/3^{1/2}) h_{1,3} q] \beta^2 = 3A_d. \quad (30)$$

The admixture of Π states by the spin-orbit interaction destroys the relation (29). If we neglect the contribution to the wave function from the AO of the light atom, then the resulting correction to (30) can be expressed in terms of the quantity $g_{\perp} - g_0$. To do this, let us replace β by $\beta + \epsilon$ in (29). For (8) we obtain in the first-order approximation in

$$g_{\perp} - g_0 = 4\epsilon b,$$

and, consequently,

$$\gamma = 2^{1/2} [q\beta + h_1 k_3 (g_{\perp} - g_0) / 4\beta].$$

Then from Eqs. (27) and (28) we obtain the following expressions:

$$\beta^2 = \frac{3A_d + (h_3 k_3^2 + 3^{-1/2} h_{1,3} k_1 k_3) (g_{\perp} - g_0)}{2(h_1 - h_3 q^2 - 2 \cdot 3^{-1/2} h_{1,3} q)}, \quad (31)$$

$$\alpha^2 = h_s^{-1} [A + 1/3 (h_1 - 10h_3 q^2 + 4 \cdot 3^{-1/2} h_{1,3} q) \beta^2]. \quad (32)$$

For the molecules considered below the spin-orbit correction changes the value of β by only 1-3%. Roughly the same error is made when we replace q by unity in the expression (31) and neglect the second term in (32). The smallness of these corrections allows us to simplify the expressions (31) and (32):

$$\alpha^2 = A/h_s, \quad (33)$$

$$\beta^2 = 3/2 A_d (h_1 - h_3 - 2 \cdot 3^{-1/2} h_{1,3})^{-1}. \quad (34)$$

The formulas (33) and (34) have a significant advantage over (31) and (32), in that they do not contain the coefficients k_i .

The expressions for the H_P and $H_{P,T}$ matrix elements contain the product $\alpha\beta$. The simplified formulas overestimate it slightly, but the error does not exceed 10%.

For the pure Π states, $\alpha = 0$, and, consequently, it is necessary in their case to take account of the mixing with the Σ state. This means that all the three parameters in Eqs. (27) and (28) are independent parameters. We can, in much the same way as we did above for the Σ states, derive a third equation, using the experimental data¹⁷ on the Ω doubling (19) or the g -factor data. But in the general case such an equation will contain two-center (?) integrals. Therefore, the method described here is probably applicable only to molecules with an ionic bond, for which molecules we can neglect the contribution from the AO of the light atom. Such

TABLE I. Molecular parameters of BaF, HgH, and HgF (in MHz).

Molecules	B	ν	A	A_d	g_{\parallel}	g_{\perp}	α	β	$10^4 W_P$	$10^4 W_{P,T}$	Ref.
BaF	6471*	83.3	2420	17.3	2.001	1.995	0,44	0,099	1,80	-0,98	[11]
	6480	81,03	2326	25	-	-	0,43	0,12	2,12	-1,15	[18]
			7000	400			0,34	0,21	18,0	-13,4	
HgH	161600*	64300*			1,976	1,823					[12]
			2610	200			0,38	0,24	19,4	-14,4	
			22127	247			0,70	0,17	25,9	-19,3	
HgF	-	-			1,993	1,961					[13]
			7858	98			0,65	0,17	23,9	-17,8	

*Data taken from Ref. 17.

and is most probably realized for molecules of the metal-halogen type, such as PbF.¹⁷ This molecule is discussed in Refs. 3 and 6.

NUMERICAL RESULTS

In Table I we present the necessary—for the computations—data that we could find in the literature. They all pertain to molecules with a $^2\Sigma$ ground state. With the exception of the data from Ref. 18, which reports the observation of the free-molecule rotational spectrum, all the data were obtained from the EPR spectra of molecules located in an argon matrix. In the table we also present the results of the computations of the parameters α and β and of the quantities W_P and $W_{P,T}$ (see (10) and (12)).

The spin-rotation spectrum of the molecules BaF and HgH can easily be computed with the aid of the Hamiltonian (15). As can be seen from a comparison of γ and A , the first of these molecules pertains to the case b_{F_1} coupling; the second, to the case b_j coupling.

The large hyperfine structure of the BaF molecules determines the minimum separation of levels with opposite parity by an amount almost twice the decrease due to the rotational splitting. In this case the closest levels turn out to be separated by the interval

$$\Delta E = E(N=1, F_1=1, F=^3/2) - E(N=0, F_1=2, F=^3/2) = 8090 \text{ MHz.} \quad (35)$$

For these levels the magnitude of the P -odd admixture is equal to

$$\delta = \langle H_P \rangle / \Delta E = 2.4 \cdot 10^{-6} i \kappa_P \langle \mathbf{n} [s, \mathbf{I}] \rangle. \quad (36)$$

For the HgH molecule the minimum level separation

$$E(N=1, J=^1/2, F) - E(N=0, J=^1/2, F) = 2.6 \cdot 10^5 \text{ MHz.} \quad (37)$$

$$\delta = 7.2 \cdot 10^{-9} i \kappa_P \langle [n, s] \mathbf{I} \rangle. \quad (38)$$

Unfortunately, the rotational constant for the HgF molecule is not known, and this makes it impossible to obtain the magnitude δ of the admixture. Nevertheless, it is clear that the quantity obtained here is of the order of, or smaller than, the corresponding quantity for BaF. Thus, if we assume that

$B = 8000 \text{ MHz}$, then for ^{201}HgF

$$E(N=1, F_1=1, F=^3/2) - E(N=0, F_1=2, F=^3/2) = 300 \text{ MHz} \quad (39)$$

and then

$$\delta = 8 \cdot 10^{-6} i \kappa_P \langle \mathbf{n} [s \times \mathbf{I}] \rangle. \quad (40)$$

To compute the P -odd effects in the spin-rotation transitions,^{19,20} we must know besides δ the electric and magnetic transition amplitudes. These amplitudes can be expressed in terms of the electric dipole moment d of the nonrotating molecule and the g tensor.

The magnitude of the P, T -odd correction to the energy of a molecule located in an external electric field E is, according to (14), given by an expression of the form

$$\delta E = W_{P,T} \langle \mathbf{n} \rangle s, \quad (41)$$

where the averaging of the vector \mathbf{n} over the spin-rotation state should be carried out with allowance for the hyperfine structure. It is clear that $\langle \mathbf{n} \rangle$ is a function of dE , and that it is oriented along the field E . Thus, in both cases we must know the quantity d .

CONCLUSION

The similarity between the matrix elements of the P - and P, T -odd interaction operators and those of the hyperfine interaction operator enables us to compute the P - and P, T -odd effects in diatomic molecules-radicals. The experimental data required for this purpose can probably be obtained directly in the course of an experiment on the observation of these effects.

It seems to us that molecules having a $^2\Sigma$ ground state are more suitable for the study of the parity nonconservation effects. The calculations are simpler in their case than in the case of the $^2\Pi$ states, since the spin-orbit mixing plays a significantly greater role in the latter case. Furthermore, the $^2\Pi$ states are, apparently, also less suitable for experiments on the observation of the P -odd effects in spin-rotation transitions. The point is that in their case, because the orbital and spin angular momenta are antiparallel, the g factors are small (in the nonrelativistic limit they are equal to zero). Therefore, the magnetic transitions between the spin-rotation levels are strongly inhibited, and their detection may be difficult.

The comparable magnitudes of the constants A and B in the spin-rotation Hamiltonian for the molecules BaF and HgF make the separation of levels with opposite parities still smaller than the purely rotational separation. This leads to further enhancement of the parity nonconservation effects. An analogous situation obtains for the other similar molecules.⁵

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¹⁾ Let us note that in the formulas (16)–(19) J^e is the angular momentum in the center-of-mass system: $J^e \equiv J_{c.m.}^e = \mathbf{j} + [\mathbf{R}_n - \mathbf{R}_{c.m.}, \mathbf{P}]$, where \mathbf{j} is the angular momentum for the motion relative to the nucleus and \mathbf{p} is the electron momentum. This circumstance can be important for molecules with two heavy nuclei, when $|\mathbf{R}_n - \mathbf{R}_{c.m.}| \sim 1$.

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