Enhancement of the electric dipole moment of the electron in diatomic molecules YbF and HgF

M. G. Kozlov. Zh. Exp. Teor. Fiz. **89**, 1933 (1985)

M. G. Kozlov, V. F. Ezhov. Phys. Rev. A, **49**, 4502 (1994)

SEMI-EMPIRICAL WAVE FUNCTION

The main assumption of the method is that the WF of the molecule can be written as follows

$$|\Lambda, \Omega\rangle =$$

$$|\lambda = \Lambda, \omega = \Omega\rangle_{unpaired} |\Lambda = 0, \Omega = 0\rangle_{coupled}$$

In the vicinity of the heavy nucleus (Hg or Yb) the WF can be expanded in spherical waves

$$|\lambda, \omega\rangle = \sum_{l,j(=l\pm 1/2)} C_{l,j} |l, j, \omega\rangle$$

Here $|l, j, \omega\rangle$ are normalized four-component spherical waves:

$$|l, j, \omega\rangle = \begin{pmatrix} f_{l,j} Y_{j,\omega}^l \\ ig_{l,j} Y_{j,\omega}^{l'} \end{pmatrix}$$

f and g are radial functions, $Y_{j,\omega}^l$ is the spherical spinor, $l' = 2j - l$.

At the small distances solutions of the Dirac equation are:

$$\begin{pmatrix} f_{l,j} \\ g_{l,j} \end{pmatrix} = \frac{k}{|k|} \frac{a_l}{Z^{1/2}r} \begin{pmatrix} (\gamma + k)J_{2\gamma}(x) - \frac{x}{2}J_{2\gamma-1}(x) \\ \alpha Z J_{2\gamma}(x) \end{pmatrix}$$

where

$$x = \sqrt{8Zr} \quad ,$$

$$\gamma = \sqrt{(j + 1/2)^2 - \alpha^2 Z^2} \quad ,$$

$$k = (l - j)(2j + 1)$$

For each $l \neq 0$ a pair of functions with $j = l - 1/2$ and $j = l + 1/2$

on the large distances have to form nonrelativistic function

$|l, m_l = \lambda, \omega\rangle$. It means, that

$$C_{1,3/2} = -\sqrt{2}C_{1,1/2},$$

$$C_{2,5/2} = -\sqrt{3/2}C_{2,3/2}, \quad etc.$$

For small r molecular WF is defined by the products

$$\sigma_s = C_{0,1/2}a_0, \quad \sigma_p = C_{1,1/2}a_1, \quad etc.,$$

The hyperfine axial tensor $\mathbf{A}$:

$$A = \frac{A_\| + 2A_\perp}{3}$$

$$= \frac{4}{3}C_{-1}^2 h_{-1,-1} + \frac{4}{9}C_1^2 h_{1,1} + \frac{8}{9}C_{-2}^2 h_{-2,-2}$$

$$+ \frac{8}{15}C_2^2 h_{2,2} + \frac{4}{5}C_{-3}^2 h_{-3,-3}$$

$$- \frac{8\sqrt{2}}{9}C_1 C_{-2} h_{1,-2} - \frac{8\sqrt{6}}{15}C_2 C_{-3} h_{2,-3}$$

$$A_d = \frac{A_\| - A_\perp}{3}$$

$$= -\frac{8}{9}C_1^2 h_{1,1} - \frac{8}{45}C_{-2}^2 h_{-2,-2}$$

$$- \frac{8}{15}C_2^2 h_{2,2} - \frac{8}{35}C_{-3}^2 h_{-3,-3}$$

$$- \frac{2\sqrt{2}}{9}C_1 C_{-2} h_{1,-2} - \frac{2\sqrt{6}}{15}C_2 C_{-3} h_{2,-3}$$

Single index k is used instead of l, j;

$$h_{i,j} = -\frac{g_n \alpha}{2m_p} \int_0^\infty (f_i g_j + g_i f_j)dr$$

g_n is G-factor of the nucleus; m_p is the proton mass.

Using experimental values for A and A_d we obtain equations for parameters σ_i. For YbF molecule these equations have the form:

$$\begin{cases} 31170\sigma_s^2 - 1330\sigma_p^2 - 50\sigma_d^2 + \cdots = 7617 \\ 5510\sigma_p^2 + 370\sigma_d^2 + \cdots \quad = 102 \end{cases}$$

The right hand side correspond to the experimental values for A and A_d in MHz measured by Van Zee *et al.*

Coefficients $C_{l,j}$ are normalized to unity. The semi-classical analysis gives $a_l^2 \approx 0.3$. It results in approximate normalization condition

$$\sigma_s^2 + 3\sigma_p^2 + \frac{5}{2}\sigma_d^2 \leq 0.3 \sum_{l,j} C_{l,j}^2 = 0.3$$

The solution which meets this condition correspond to the negligible contribution of the d-wave (less then 5%).

$$\sigma_s^2 = 0.24, \quad \sigma_p^2 = 0.019$$

$$\sigma_s \sigma_p = -0.067$$

SPIN-ORBIT INTERACTION

Mixing of the ground state with the first excited state

$(\lambda = 1, \omega = 1/2)$:

$$\xi = \frac{\langle \lambda = 1, \omega | H_{so} | \lambda = 0, \omega \rangle}{E_0 - E_1}$$

For YbF that is equal to

$$\xi = 0.37 \frac{\langle 6p, \lambda = 1, \omega | H_{so} | 6p, \lambda = 0, \omega \rangle}{18000 \ cm^{-1}} \approx -0.03$$

For HgF molecule this mixing is only slightly larger. It means, that

the accuracy of the equations

$$C_{1,3/2} = -\sqrt{2}C_{1,1/2},$$

$$C_{2,5/2} = -\sqrt{3/2}C_{2,3/2}, \ etc.$$

is about 3%.

P- AND *T*-ODD INTERACTIONS

Interaction of the EDM of the electron d_e with the molecular electric field $(-\nabla\phi)$:

$$H_d = 2d_e \begin{pmatrix} 0 & 0 \\ 0 & \vec{\sigma} \end{pmatrix} (-\nabla\phi)$$

$$\langle \lambda, \omega | H_d | \lambda, \omega \rangle = W_d d_e \omega$$

$$W_d = 8 \sum_i C_{-i} C_i \int_0^\infty g_{-i} g_i \frac{d\phi}{dr} r^2 dr$$

$$= 16\alpha^2 Z^3 \left(\frac{\sigma_s \sigma_p}{\gamma_{1/2}(4\gamma_{1/2}^2 - 1)} - \frac{\sqrt{2}\sigma_p \sigma_d}{\gamma_{3/2}(4\gamma_{3/2}^2 - 1)} \right)$$

For YbF molecule this expression gives

$$W_d = 174\sigma_s\sigma_p - 15\sigma_p\sigma_d + ... = -11.7$$

Electron–nuclear scalar interaction

$$H_S = i\frac{G\alpha}{\sqrt{2}}Zk_S\gamma_0\gamma_5 n(\mathbf{r})$$

G is the Fermi constant; γ_i are the Dirac matrices; $n(\mathbf{r})$ is the nuclear density normalized to unity. Dimensionless factor Zk_S is: $Zk_S = Zk_{S,p} + Nk_{S,n}$.

Interaction of the nuclear magnetic quadrupole moment M with the molecular magnetic field:

$$H_M = -\frac{M}{4I(2I-1)}T_{i,k}\frac{3}{2r^5}\gamma_0\gamma_j r_l(\epsilon_{j,l,i}r_k + \epsilon_{j,l,k}r_i)\ ,$$
$$T_{i,k} = I_i I_k + I_k I_i - \frac{2}{3}I(I+1)\delta_{i,k}$$

P-odd, T-even interaction of the nuclear anapole moment with valence electron:

$$H_A = \frac{G\alpha}{\sqrt{2}}k_A\mathbf{I}\gamma_0\vec{\gamma}n(\mathbf{r})$$

k_A is the anapole moment constant for the ytterbium nucleus (Khriplovich *et al*).

SPIN-ROTATIONAL HAMILTONIAN

$$H_{sr} = B\mathbf{N}^2 + \gamma \mathbf{S} \cdot \mathbf{N} + \mathbf{S} \cdot \mathbf{A}\mathbf{I} + \mathbf{S} \cdot \mathbf{A}'\mathbf{I}'$$

$$+ \mu_0 \mathbf{S} \cdot \mathbf{G}\mathbf{B} - D\mathbf{n} \cdot \mathbf{E}$$

$$+ W_A k_A \mathbf{n} \times \mathbf{S} \cdot \mathbf{I} + (W_S k_S + W_d d_e)\mathbf{S} \cdot \mathbf{n}$$

$$+ \frac{-1}{4I(2I-1)} T_{i,k} \left(\frac{1}{2}Qq_0 n_i n_k + 2MW_M S_i n_k\right).$$

In this expression $\mathbf{I}$ and $\mathbf{I}'$ are the spins of Yb and F nuclei; $\mathbf{N}$ is the rotational angular momentum; B and γ are the rotational and the spin-doubling constants. Tensors $\mathbf{A}$ and $\mathbf{A}'$ correspond to the hyperfine structures on two nuclei. μ_0 is the Bohr magneton; $\mathbf{n}$ is the molecular axis unit vector directed from Yb (or Hg) to F.

Experimental values for H_{sr} parameters for ^{171}Yb^{19}F molecule:

$$B = 7237 \; MHz;$$

$$A_{\parallel} = 7822 \; MHz, \quad A_{\perp} = 7513 \; MHz \; (I = 1/2);$$

$$A'_{\parallel} = 220 \; MHz, \quad A'_{\perp} = 134 \; MHz \; (I' = 1/2);$$

$$G_{\parallel} = 1.9975, \quad G_{\perp} = 1.9954$$

Constant γ is found from the relation (Knight *et al*)

$$\gamma = -2B(G_{\perp} - G_{\parallel}) \approx 30 \; MHz$$

The rough estimate for D:

$$D = -R_0 + 2C_{0,1/2}C_{1,1/2}\langle 6s|r|6p\rangle$$

$$\approx -5.2 \cdot 10^{-18} \ CGSE,$$

R_0 is the internuclear distance.

Constants q_0 and W_M do not vanish for ^{173}Yb ($I = 5/2$). If $Q = 2.8 \cdot 10^{-24} \ cm^2$, then

$$Qq_0^{val} = 1880 \ MHz.$$

Values of the constants W_i are as follows:

$$W_A = 0.73 \ kHz;$$

$$W_S = -48 \ kHz;$$

$$W_d = -1.5 \cdot 10^{25} \frac{Hz}{e \ cm};$$

$$W_M = 2.1 \cdot 10^{33} \frac{Hz}{e \ cm^2};$$

BEST EXPERIMENTAL LIMITS ON THE CONSTANTS OF P- AND T-ODD INTERACTIONS AND CORRESPONDING FREQUENCY SHIFTS IN MOLECULES

Const.	Group	Year	Object	Upper bound	Frequency shift (Hz)		
					TlF	HgF	YbF
Q_S	Seatle	1993	^{199}Hg	$3 \cdot 10^{-50} e\ cm^3$	$5 \cdot 10^{-5}$	–	–
C_T	”	”	”	$2 \cdot 10^{-8}$	$3 \cdot 10^{-5}$	–	–
C_S	”	”	”	$1 \cdot 10^{-6}$	$1 \cdot 10^{-4}$	0.2	0.05
M	Amherst	1989	^{133}Cs	$1 \cdot 10^{-33} e\ cm^2$	–	1.5	0.7
d_e	Berkeley	1990	Tl	$1 \cdot 10^{-26} e\ cm$	–	0.5	0.15