Sensitivity of the YbOH molecule to $\mathcal{PT}$-odd effects in an external electric field

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An electron electric dipole moment (eEDM) search using laser-coolable triatomics such as YbOH is one of the most sensitive probes for physics beyond the standard model. The eEDM-induced energy shift is proportional to the polarization (P) of the molecule. Similarly to diatomics with an Ω-doubling structure, it was assumed that for triatomics with an l-doubling structure, related to the vibrational angular momentum, P can be easily saturated to almost 100% with a moderate external electric field. We developed a method for the calculation of the properties of triatomic molecules and applied it to a calculation of P of ^{174}YbOH in the first excited $v = 1$ bending mode. Calculations showed that most of the levels reach less than 50% efficiency. We showed that this fact is related to the Hund's case b coupling scheme of YbOH. As the coupling scheme (for $\Omega = 1/2$ molecules) approaches the a (or c) case, polarization increases up to 100%. The results of our calculations should be used for the correct extracting of the eEDM value from YbOH experiments and similar calculations are required for other molecules.

Measuring the electron electric dipole moment (eEDM) is now considered as a most promising test for the existence of physics beyond the standard model [1–4]. The current limit on the electron electric dipole moment (the ACME II experiment), $|d_e| < 1.1 \times 10^{-29}$ e cm (90% confidence), was set by measuring the spin precession using thorium monoxide (ThO) molecules in the metastable electronic $H^3\Delta_1$ state [5]. To successfully perform these kinds of experiments, the possibility of suppressing systematic effects is of high importance. Previously it was shown that due to the existence of Ω-doublet levels the experiments for searching for $\mathcal{PT}$-odd effects on ThO [5–10] or HfF$^+$ [11,12] are very robust against a number of systematics.

In turn, cold polar molecules provide unique opportunities for further progress in the search for the effects of symmetry violations [13]. In such molecules the sensitivity of the experiments can be strongly enhanced due to increased coherence time. Both the possibility of laser cooling and the existence of close levels of opposite parity can be realized with triatomic molecules such as RaOH [14], YbOH [15], etc. In this case the role of Ω doublets used in diatomic molecular experiments is taken over by l doublets of the excited $v = 1$ bending vibrational modes [15,16].

Any eEDM experiment searches for an eEDM-induced Stark shift

$$\delta E = d_e E_{\mathrm{eff}} P, \qquad (1)$$

where d_e is the value of the electron electric dipole moment, E_{eff} is the *effective electric field* acting on the electron in the molecule, which is the subject of molecular calculations

[17–21], and P is the polarization of the molecule by an external electric field. To extract $d_e = \delta E/(E_{\mathrm{eff}} P)$ from the measured shift δE, one needs to know $E_{\mathrm{eff}} P$. Therefore the value of $E_{\mathrm{eff}} P$ directly influences the d_e value extracted from experiments [22,23].

It is well known that for diatomics, due to the existence of the Ω-doublet structure, the P value approaches unity for small laboratory electric fields [6]. The same situation was expected for linear triatomic molecules [15]. In our Letter we show that the l-doubling structure, is, in general, different from Ω doubling, and the P value tends to approach half of the maximum value for molecules with Hund's case b coupling scheme and a value of 100% as the coupling scheme approaches the a (or c) case.

In the present Letter we develop a method for the computation of energy levels and different properties of linear triatomic molecules. We apply it for calculations of the sensitivity of the ^{174}YbOH molecule to eEDM in the ground rotational $N = 1$ level of the first excited $v = 1$ bending mode to eEDM in an external electric field. According to Eq. (1), these calculations should be used to determine the limit on the eEDM.

Together with eEDM one always needs to consider the scalar $\mathcal{PT}$-odd electron-nuclear interaction, since its influence on the spectrum of molecules is identical to eEDM. For brevity, only the influence of eEDM is mentioned in the present Letter.

Consider the molecular Hamiltonian for the electronic-nuclear motion, excluding the nuclear hyperfine structure, and with a fixed OH ligand stretch. In the Jacobi coordinates (represented in Fig. 1) in the *laboratory* frame it reads as

$$\hat{\mathbf{H}}_{\mathrm{mol}} = -\frac{\hbar^2}{2\mu}\frac{\partial^2}{\partial R^2} + \frac{\hat{\mathbf{L}}^2}{2\mu R^2} + \frac{\hat{\mathbf{l}}^2}{2\mu_{\mathrm{OH}} r^2} + \hat{\mathbf{H}}_{\mathrm{el}}, \qquad (2)$$

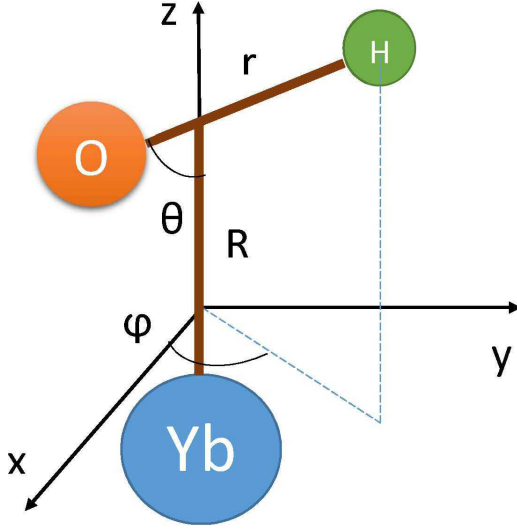———————
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FIG. 1. Orientation of the YbOH relative to the molecular frame. Orientation of the molecular frame relative to the laboratory one is given by Euler's angles (not presented in the figure) α, β, $\gamma = 0$. R, r, θ are Jacobi coordinates.

where μ is the reduced mass of the Yb-OH system, μ_{OH} is the reduced mass of the OH ligand, $\hat{\mathbf{L}}$ is the angular momentum of the rotation of the Yb atom and OH around their center of mass, $\hat{\mathbf{l}}$ is the angular momentum of the rotation of the OH, and $\hat{\mathbf{H}}_{\mathrm{el}}$ is the electronic Hamiltonian. For a number of applications it is convenient to rewrite the Hamiltonian in a molecular reference frame defined by the Euler angles α, β, γ (see, e.g., Fig. 20 in Ref. [24]), with α, β corresponding to the azimuthal and polar angles of the axis (it will be the z axis of the molecular frame) going through Yb and the center of mass of OH, and $\gamma = 0$. In this case the Hamiltonian (2) is rewritten as

$$\hat{\mathbf{H}}_{\mathrm{mol}} = -\frac{\hbar^2}{2\mu}\frac{\partial^2}{\partial R^2} + \frac{(\hat{\mathbf{J}} - \hat{\mathbf{J}}^{e-v})^2}{2\mu R^2} + \frac{\hat{\mathbf{l}}^2}{2\mu_{\mathrm{OH}}r^2} + \hat{\mathbf{H}}_{\mathrm{el}}, \quad (3)$$

where $\hat{\mathbf{J}}$ is the total molecular less nuclear spin angular momentum, $\hat{\mathbf{J}}^{e-v} = \hat{\mathbf{J}}^e + \hat{\mathbf{J}}^v$, $\hat{\mathbf{J}}^e$ is the total electronic momentum, $\hat{\mathbf{J}}^v$ is the vibrational momentum, $\hat{J}_z^v = -i\hbar\frac{\partial}{\partial\varphi}$, φ is the angle between the xz plane of the molecular reference frame and the plane of the molecule (see Fig. 1), $\hat{\mathbf{l}}^2 = \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\sin\theta\frac{\partial}{\partial\theta} + \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\varphi^2}$, and θ is the angle between OH and the z axes. The condition $\theta = 0$ corresponds to the linear configuration where the O atom is between the Yb and H ones.

For the purpose of the present Letter, we also include hyperfine interactions and interactions with an external electric field. Finally, our Hamiltonian reads as

$$\hat{\mathbf{H}} = \hat{\mathbf{H}}_{\mathrm{mol}} + \hat{\mathbf{H}}_{\mathrm{hfs}} + \hat{\mathbf{H}}_{\mathrm{ext}}, \quad (4)$$

where

$$\hat{\mathbf{H}}_{\mathrm{hfs}} = g_{\mathrm{H}}\mathbf{I} \cdot \sum_a \left(\frac{\boldsymbol{\alpha}_a \times \boldsymbol{r}_a}{r_a^3}\right) \quad (5)$$

is the hyperfine interaction between electrons and the hydrogen nucleus, g_{H} is the g factor of the hydrogen nucleus, $\boldsymbol{\alpha}_a$ are the Dirac matrices for the ath electron, $\boldsymbol{r}_a$ is its radius vector

in the coordinate system centered on the H nucleus, and index a enumerates (as in all equations below) electrons of YbOH. Here,

$$\hat{\mathbf{H}}_{\mathrm{ext}} = -\mathbf{D} \cdot \mathbf{E} \quad (6)$$

describes the interaction of the molecule with an external electric field, and $\mathbf{D}$ is the dipole moment operator.

Wave functions were obtained by numerical diagonalization of the Hamiltonian (4) over the basis set of the electronic-rotational-vibrational wave functions

$$\Psi_{\Omega m\omega}P_{lm}(\theta)\chi(R)\Theta^J_{M_J,\omega}(\alpha, \beta)U^{\mathrm{H}}_{M_I}. \quad (7)$$

Here, $\Theta^J_{M_J,\omega}(\alpha, \beta) = \sqrt{(2J+1)/4\pi}D^J_{M_J,\omega}(\alpha, \beta, \gamma = 0)$ is the rotational wave function, $U^{\mathrm{H}}_{M_I}$ is the hydrogen nuclear spin-wave functions, M_J is the projection of the molecular (electronic-rotational-vibrational) angular momentum $\hat{\mathbf{J}}$ on the laboratory axis, ω is the projection of the same momentum on the z axis of the molecular frame, $M_I = \pm 1/2$ is the projection of the nuclear angular momentum $I = 1/2$ of hydrogen on the laboratory axis, and $P_{lm}(\theta)$ is the associated Legendre polynomial,

$$\Psi_{\Omega m\omega} = e^{i(\omega-1/2\sum_a \sigma_a)\varphi} \sum_i \Psi^e_{\Lambda_i}\Psi^s_{\Sigma_i}, \quad (8)$$

where σ_a are electronic spin variables, $\Psi^e_{\Lambda_i}$ and $\Psi^s_{\Sigma_i}$ are orbital and spin-wave functions, respectively, Λ_i and Σ_i are projections of electronic orbital and spin momenta on the z axis, and i enumerates all possible nonrelativistic wave functions.

Equation (8) is the expansion of the electronic wave function in terms of the nonrelativistic functions of a linear configuration. For a linear configuration only terms with $\Omega = \Lambda_i + \Sigma_i$ contribute. $\Psi_{\Omega m\omega}$ depends on m since ω is chosen so that $\omega = \Omega + m$. $\Psi_{\Omega m\omega}$ is the eigenfunction of the electronic-vibrational momentum:

$$\hat{J}_z^{e-v}\Psi_{\Omega m\omega} = \hbar\omega\Psi_{\Omega m\omega}. \quad (9)$$

To prove Eq. (9), we note that nonrelativistic wave functions depend on φ as $\Psi^e_{\Lambda_i} \sim e^{-i\Lambda_i\varphi}$ and that $1/2\sum_a \sigma_a\Psi^s_{\Sigma_i} = \Sigma_i\Psi^s_{\Sigma_i}$. For the lowest vibrational levels the configuration of the molecule is close to linear, therefore the main contribution in sum (8) is given by the terms with $\Omega = \Lambda_i + \Sigma_i$. Therefore, we also have approximate relations

$$\hat{J}_z^e\Psi_{\Omega m\omega} \approx \hbar\Omega\Psi_{\Omega m\omega}, \quad (10)$$

$$\hat{J}_z^v\Psi_{\Omega m\omega} \approx \hbar m\Psi_{\Omega m\omega}. \quad (11)$$

In this calculation functions with $\omega - m = \Omega = \pm 1/2$, $l = 0$–30 (as in Ref. [21]), $m = 0, \pm 1, \pm 2$, and $J = 1/2, 3/2, 5/2$ were included in the basis set (7). Note that the ground mode $v = 0$ corresponds to $m = 0$, the first excited bending mode $v = 1$ to $m = \pm 1$, the second excited bending mode has states with $m = 0, \pm 2$, etc. Using the basis set (7) allows us to employ (with a little modification) the codes developed for the diatomic calculations [7,9,10,12,25,26], and paves the way for computations of the g factors, g-factor difference, different systematics in an external electromagnetic field, etc. These calculations are important for the optimization of experimental conditions for the eEDM experiment.

In the present Letter we applied the method for the calculation of the sensitivity of ^{174}YbOH to $\mathcal{PT}$-odd effects in the external electric field. As a test, we exactly reproduced with the described method the frequencies of the bending and stretching vibrational modes (319 and 550 cm^{-1}, respectively) and a value of 26 MHz of the l doubling for the ^{174}YbOH molecule obtained with the Hamiltonian (2) in Ref. [21].

For the purposes of the present Letter, we found that ignoring the R dependence leads to an error of about 1% or less for the polarization P of ^{174}YbOH. Therefore, we reduced the Hamiltonian (3) and basis set (7) to

$$\hat{\mathbf{H}}_{\text{mol}} = \frac{(\hat{\mathbf{J}} - \hat{\mathbf{J}}^{e-v})^2}{2\mu R^2} + \frac{\hat{\mathbf{l}}^2}{2\mu_{\text{OH}} r^2} + \hat{\mathbf{H}}_{\text{el}}, \quad (12)$$

and

$$\Psi_{\Omega m\omega} P_{lm}(\theta)\Theta^J_{M_J,\omega}(\alpha, \beta)U^{\text{H}}_{M_I}, \quad (13)$$

with $R = 3.9$ a.u., respectively. In this approximation we neglect the influence of the stretching modes but nevertheless take into account the bending ones with fixed R. Provided that the *electronic-vibrational* matrix elements are known, the matrix elements of $\hat{\mathbf{H}}$ between states in the basis set (13) can be calculated with the help of the angular momentum algebra [24] in the same way as for the diatomic molecules [25]. Note, however, that pure electronic matrix elements, in general, depend on θ, and the selection rules for the Ω quantum number can be violated. In turn, the selection rules for the ω quantum number are rigorous and the same as for the Ω quantum number in the diatomics. For example, the matrix element for the perpendicular hyperfine structure constant,

$$A_\perp = g_{\text{H}}\langle\Psi_{\Omega=1/2m\omega}P_{lm}|\sum_a \left(\frac{\boldsymbol{\alpha}_a \times \boldsymbol{r}_a}{r_a^3}\right)_+$$

$$\times |\Psi_{\Omega=1/2m-1\omega-1}P_{l'm-1}\rangle, \quad (14)$$

is, in general, nonzero. Here,

$$\left(\frac{\boldsymbol{\alpha}_a \times \boldsymbol{r}_a}{r_a^3}\right)_+ = \left(\frac{\boldsymbol{\alpha}_a \times \boldsymbol{r}_a}{r_a^3}\right)_x + i\left(\frac{\boldsymbol{\alpha}_a \times \boldsymbol{r}_a}{r_a^3}\right)_y,$$

and similarly for other vectors. Since the hyperfine interaction on hydrogen is small, in the present Letter we neglected their θ dependence and used the values

$$A_\parallel = \frac{g_{\text{H}}}{\Omega}\langle\Psi_{\Omega m\omega}P_{lm}|\sum_a \left(\frac{\boldsymbol{\alpha}_a \times \boldsymbol{r}_a}{r_a^3}\right)_z |\Psi_{\Omega m\omega}P_{l'm}\rangle$$

$$= 6.436\delta_{ll'} \text{ MHz}, \quad (15)$$

$$A_\perp = g_{\text{H}}\langle\Psi_{\Omega=1/2m\omega}P_{lm}|\sum_i \left(\frac{\boldsymbol{\alpha}_i \times \boldsymbol{r}_i}{r_i^3}\right)_+ |\Psi_{\Omega=-1/2m\omega-1}P_{l'm}\rangle$$

$$= 3.977\delta_{ll'} \text{ MHz} \quad (16)$$

from Ref. [16]. The spin-rotational interaction is modeled by the matrix element [16]

$$p = \frac{1}{\mu R^2}\langle\Psi_{\Omega=1/2m\omega}P_{lm}|J^e_+|\Psi_{\Omega=-1/2m\omega-1}P_{l'm}\rangle$$

$$= 0.5116\delta_{ll'} \text{ cm}^{-1}. \quad (17)$$
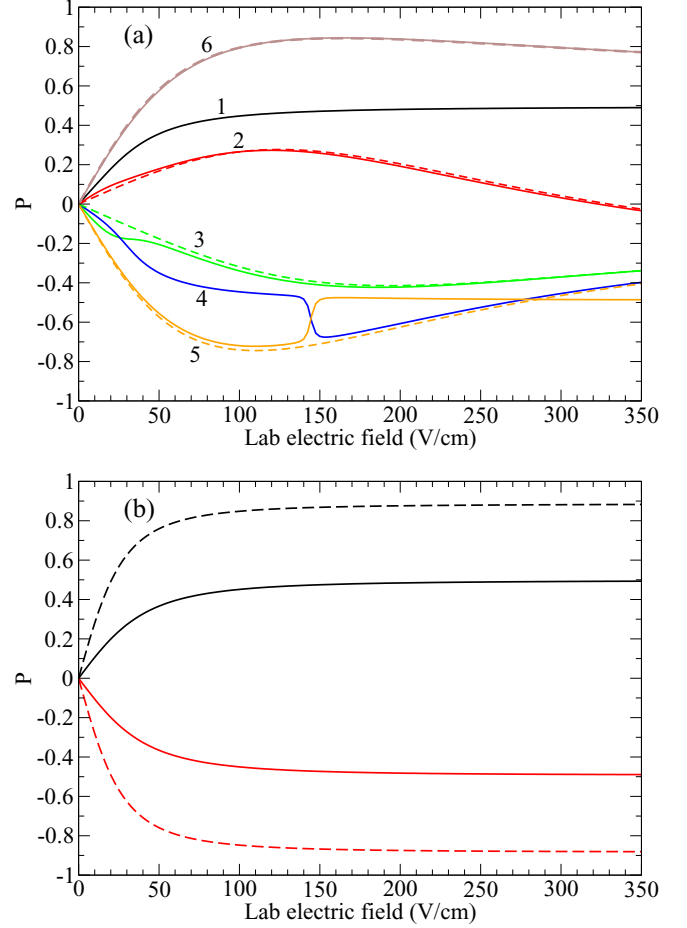

FIG. 2. Polarization P [see Eq. (1)] for the lowest $N = 1$ rotational level of the first excited $v = 1$ bending vibrational mode of ^{174}YbOH as a function of the external electric field. (a) Solid lines correspond to the $M_F = M_J + M_I = 1$ levels. Dashed lines correspond to the $M_J = 1/2$ levels. Colors (numbering) of lines correspond to colors (numbering) of lines in Fig. 3. (b) $M_F = M + M_I = 2$ levels. Solid lines correspond to the real matrix element (17), and dashed lines correspond to the reduced matrix element (19).

The matrix element for the dipole moment operator

$$\langle\Psi_{\Omega m\omega}P_{lm}|\mathbf{D}|\Psi_{\Omega m\omega}P_{l'm}\rangle = 0.433\delta_{ll'} \text{ a.u.} \quad (18)$$

was taken from Ref. [18]. The adiabatic potential was taken from Ref. [21].

In an experiment to search the $\mathcal{PT}$-odd effects, the opposite parity levels of the molecule are mixed in the external electric field to polarize the molecule. For a completely polarized ($P = \pm1$) ^{174}YbOH the $\mathcal{PT}$-odd interaction energy shift approaches the maximum value given by $\delta E^{\text{max}} = E_{\text{eff}}d_e$.

The absolute value of the finite-electric-field shift (1) is smaller than that of δE^{max}. In Fig. 2 the calculated polarization P for the lowest $N = 1$ rotational level of the first excited $v = 1$ bending vibrational mode of the ^{174}YbOH as a function of the external electric field is presented. The function $E_{\text{eff}}(R, \theta)$ from Ref. [21] was used for calculations.

Calculations showed that most of the levels have a polarization equal to or less than 50%. In particular, the stretched $M_F = M_J + M_I = 2$ states smoothly approach a 50%

efficiency value. This is in contradiction with Ref. [15] where about 100% efficiency was found.

The reason for the fact that 100% efficiency is not reached is the large Coriolis interaction ($\frac{1}{2\mu R^2} J_+^e J_-$) between levels $|\Omega = 1/2, m = 1, M_J = 3/2, M_I = 1/2, J = 3/2\rangle$ and $|\Omega = -1/2, m = 1, M_J = 3/2, M_I = 1/2, J = 3/2\rangle$ (a combination of the above states gives the $N = 1$ state). The interaction is of the same order of magnitude as the separation between close rotational levels. In the Hund's case (b) used in Ref. [15] this fact can be explained by the expression $\langle mNSMM_S|\hat{n}|mNSMM_S\rangle = \text{sgn}(Mm)/N(N+1)$ for projection of the vector $\hat{n}$ on the laboratory axis. Here, $\hat{\mathbf{N}} = \hat{\mathbf{J}} - \hat{\mathbf{S}}$, $\hat{\mathbf{S}}$ is the electronic spin operator, $\hat{n}$ is the unit vector along the molecular axis, and $M, M_S = \pm 1/2$ are the projections of the $\hat{\mathbf{N}}, \hat{\mathbf{S}}$ on the laboratory axis. The factor $1/N(N+1) = 1/2$ for $N = 1$ [$M = \pm 1$ for Hund's case (b)] gives a 50% suppression for the effect. The factor will be different for different rotational levels. For $N = 2$, for example, it is $1/6$ for $M = 1$ and $1/3$ for $M = 2$. The same situation will be for other Hund's case b molecules, e.g., YbOCH$_3$, RaOH, etc. In Ref. [7] it is noted that the $J = 2$ excited rotational state of ThO has the same sensitivity to $\mathcal{PT}$-odd effects as $J = 1$, but is more robust against a number of systematic errors. Using the excited rotational levels of YbOH one should take into account that the sensitivity will be smaller than that for the $N = 1$ level.

The spin-rotation interaction couples $\hat{\mathbf{N}}$ and $\hat{\mathbf{S}}$ to the $\hat{\mathbf{J}}$ angular momentum. The state $|m = 1 N = 1 S J = 3/2 M_J\rangle = 3/4|\Omega = 1/2, m = 1, M_J, J = 3/2\rangle + 1/4|\Omega = -1/2, m = 1, M_J, J = 3/2\rangle$ has $|P| = 3/4 - 1/4 = 1/2$. This is also seen from the consideration above and facts that $|mN = 1SJ = 3/2M_J = 3/2\rangle = |mN = 1SM = 1M_S = 1/2\rangle$ and P is independent of M_J. The state $|m = 1 N = 1 S J = 1/2 M_J\rangle = |\Omega = -1/2, m = 1, M_J, J = 1/2\rangle$ has $|P| = 1$. (Formulas for $m = -1$ can be obtained by substituting $m \to -m, \Omega \to -\Omega$.) The state $J = 1/2$ with a definite quantum number m has a maximum polarization $|P| = 1$. However, the quantum number m implies the mixing of l doublets by some finite electric field. The electric field also mixes the $J = 1/2$ and $J = 3/2$ levels which leads to a decrease of the polarization up to the $|P| = 1/2$ value at the limit. Since at zero electric field $P = 0$, a maximum of the P value as a function of electric field with $1/2 < |P|^{\text{max}} < 1$ is observed. The larger the spin-rotation interaction and the smaller the l doubling, the larger is $|P|^{\text{max}}$.

It is important to note that we considered the projection of the vector $\hat{n}$ on the laboratory axis since in Hund's case b the quantization axis for the electron spin is a laboratory axis. For Hund's case a (when both orbital angular and spin angular momenta have definite projections on the molecular axis) $^2\Pi_{1/2}$ states the matrix element of the Coriolis interaction between $|\Omega = 1/2, m = 1, M_J = 3/2, M_I = 1/2, J = 3/2\rangle$ and $|\Omega = -1/2, m = 1, M_J = 3/2, M_I = 1/2, J = 3/2\rangle$ is zero and a $P = 1$ value is reached. For Hund's case c when the $^2\Pi_{1/2}$ state has an admixture of other $\Omega = 1/2$ states (mostly $^2\Sigma$), the Coriolis interaction is not zero again, but less than one given by Eq. (17). In Fig. 2(b), the calculated polarization for the matrix element
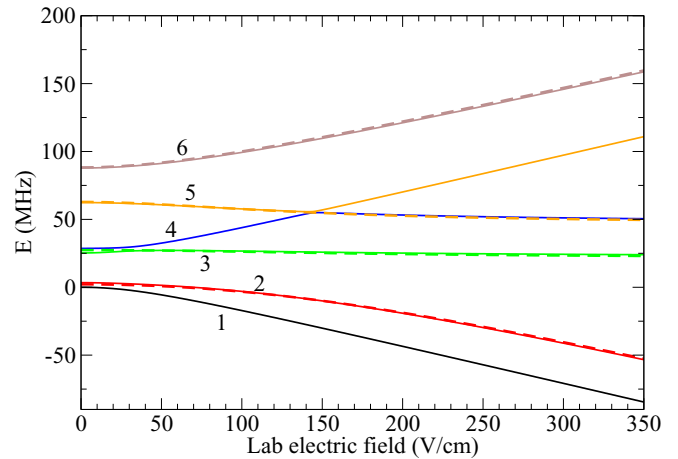
$$p = 0.15 \, \delta_{ll'} \, \text{cm}^{-1}, \tag{19}$$

FIG. 3. Energies of the lowest $N = 1$ rotational level of the first excited $v = 1$ bending vibrational mode of ^{174}YbOH as functions of the external electric field. Solid lines correspond to the $M_F = M_J + M_I = 1$ levels. Dashed lines correspond to the $M_J = 1/2$ levels. Colors (numbering) of lines correspond to colors (numbering) of lines in Fig. 2(a).

corresponding to the structure of the $X^2\Pi_{1/2}$ state of PbF [27] and can be expected on this level for the PbOH molecule, is given. As is expected, for a smaller value of p, a larger polarization P is reached.

For diatomic molecules, for a given value of the external electric field, a smaller value of p implies a larger P value as well. We stress, however, that for the triatomics, we discuss here the saturated value of P which is independent of p in diatomics.

Exact values which take into account the mixing of the levels by the hyperfine interaction should be obtained in molecular calculations as described in our method. Solid lines on Fig. 2(a) show that four of the six levels $M_F = 1$ similarly to $M_F = 2$ have a polarization of less than 50%. Two of them ($m = \pm 1, J = 1/2$ levels) reach values of 84% and -72% for electric fields of 109 and 166 V/cm, respectively, and their absolute values decrease for higher values of the field. Dashed lines on Fig. 2(a) correspond to $M_J = 1/2$ states assuming zero nuclear spin (and therefore the hyperfine interaction is neglected). One sees that a hyperfine interaction only slightly changes the P value.

For dipole moment (18) increased (decreased) by 10%, the extreme values for polarization remain the same but the corresponding values for the electric field shifted to 100 and 150 V/cm (122 and 184 V/cm). The energy levels of the six levels $M_F = 1$ and four levels $M_J = 1/2$ (assuming zero nuclear spin) are shown in Fig. 3. At zero electric field the first four $M_F = 1$ levels are $J = 3/2$ ones and the next two are $J = 1/2$ levels. At an electric field strength of about 150 V/cm the avoided crossing between the fourth and fifth levels $M_F = 1$ is observed. Due to the large change in the corresponding wave functions, a sharp change in P of these levels is observed.

Finally, we considered the polarization of the ^{174}YbOH molecule in the first excited bending mode by the external electric field. We found that values for polarization are smaller than 100% for realistic electric field strengths and decrease

as the rotational quantum number increases. The limit for polarization is $P = Mm/N(N+1)$ (e.g., $P = 1/2$ for $N = 1$, $P = 1/6, 1/3$ for $N = 2$, etc.). The spin-rotation interaction increases the P value, forming a maximum at some electric field. The situation will be the same for other Hund's case b molecules, e.g., $YbOCH_3$, RaOH, etc. A hyperfine interaction can potentially increase the P value up to 100%. For ^{174}YbOH, $P = 0.84$ is reached for an electric field strength of 109 V/cm.

The results of our Letter should be used for the correct extracting of the eEDM value from YbOH experiments and an optimal choice of the quantum state and electric field strength. Our results confirm the fundamental importance of polyatomic molecules as candidates for eEDM measurements.

[1] T. Fukuyama, Int. J. Mod. Phys. A **27**, 1230015 (2012).

[2] M. Pospelov and A. Ritz, Phys. Rev. D **89**, 056006 (2014).

[3] Y. Yamaguchi and N. Yamanaka, Phys. Rev. Lett. **125**, 241802 (2020).

[4] Y. Yamaguchi and N. Yamanaka, Phys. Rev. D **103**, 013001 (2021).

[5] V. Andreev, D. Ang, D. DeMille, J. Doyle, G. Gabrielse, J. Haefner, N. Hutzler, Z. Lasner, C. Meisenhelder, B. O'Leary *et al.*, Nature (London) **562**, 355 (2018).

[6] D. DeMille, F. Bay, S. Bickman, D. Kawall, L. Hunter, D. Krause, Jr., S. Maxwell, and K. Ulmer, in *Art and Symmetry in Experimental Physics*, edited by D. Budker, P. H. Bucksbaum, and S. J. Freedman, AIP Conf. Proc. Vol. 596 (AIP, Melville, NY, 2001), p. 72.

[7] A. N. Petrov, L. V. Skripnikov, A. V. Titov, N. R. Hutzler, P. W. Hess, B. R. O'Leary, B. Spaun, D. DeMille, G. Gabrielse, and J. M. Doyle, Phys. Rev. A **89**, 062505 (2014).

[8] A. C. Vutha, W. C. Campbell, Y. V. Gurevich, N. R. Hutzler, M. Parsons, D. Patterson, E. Petrik, B. Spaun, J. M. Doyle, G. Gabrielse *et al.*, J. Phys. B: At., Mol. Opt. Phys. **43**, 074007 (2010).

[9] A. N. Petrov, Phys. Rev. A **91**, 062509 (2015).

[10] A. N. Petrov, Phys. Rev. A **95**, 062501 (2017).

[11] W. B. Cairncross, D. N. Gresh, M. Grau, K. C. Cossel, T. S. Roussy, Y. Ni, Y. Zhou, J. Ye, and E. A. Cornell, Phys. Rev. Lett. **119**, 153001 (2017).

[12] A. N. Petrov, Phys. Rev. A **97**, 052504 (2018).

[13] T. A. Isaev and R. Berger, Phys. Rev. Lett. **116**, 063006 (2016).

[14] T. A. Isaev, A. V. Zaitsevskii, and E. Eliav, J. Phys. B: At., Mol. Opt. Phys. **50**, 225101 (2017).

[15] I. Kozyrev and N. R. Hutzler, Phys. Rev. Lett. **119**, 133002 (2017).

[16] N. H. Pilgram, A. Jadbabaie, Y. Zeng, N. R. Hutzler, and T. C. Steimle, J. Chem. Phys. **154**, 244309 (2021).

[17] M. Denis, P. A. B. Haase, R. G. E. Timmermans, E. Eliav, N. R. Hutzler, and A. Borschevsky, Phys. Rev. A **99**, 042512 (2019).

[18] V. S. Prasannaa, N. Shitara, A. Sakurai, M. Abe, and B. P. Das, Phys. Rev. A **99**, 062502 (2019).

[19] K. Gaul and R. Berger, Phys. Rev. A **101**, 012508 (2020).

[20] A. Zakharova and A. Petrov, Phys. Rev. A **103**, 032819 (2021).

[21] A. Zakharova, I. Kurchavov, and A. Petrov, J. Chem. Phys. **155**, 164301 (2021).

[22] H. S. Nataraj, B. K. Sahoo, B. P. Das, and D. Mukherjee, Phys. Rev. Lett. **106**, 200403 (2011).

[23] S. G. Porsev, M. S. Safronova, and M. G. Kozlov, Phys. Rev. Lett. **108**, 173001 (2012).

[24] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics*, 3rd ed. (Pergamon, Oxford, UK, 1977).

[25] A. N. Petrov, Phys. Rev. A **83**, 024502 (2011).

[26] A. N. Petrov, L. V. Skripnikov, A. V. Titov, and V. V. Flambaum, Phys. Rev. A **98**, 042502 (2018).

[27] A. N. Petrov, L. V. Skripnikov, A. V. Titov, and R. J. Mawhorter, Phys. Rev. A **88**, 010501(R) (2013).