ac Stark effect in ThO $H^3\Delta_1$ for the electron electric-dipole-moment search

A. N. Petrov*

National Research Centre "Kurchatov Institute," B.P. Konstantinov Petersburg Nuclear Physics Institute, Gatchina, Leningrad District 188300, Russia

and Division of Quantum Mechanics, St. Petersburg State University, St. Petersburg 198504, Russia

(Received 17 May 2015; published 24 June 2015)

A method and a code for calculations of diatomic molecules in the external variable electromagnetic field have been developed. The code is applied for calculation of systematics in the electron's electric-dipole-moment search experiment on ThO $H^3\Delta_1$ state related to geometric phases, including dependence on the Ω doublet, rotational level, and external static electric field. It is found that systematics decrease cubically with respect to the frequency of the rotating transverse component of the electric field. Calculation confirms that experiment on ThO $H^3\Delta_1$ state is very robust against systematic errors related to geometric phases.

DOI: 10.1103/PhysRevA.91.062509 PACS number(s): 31.30.jp, 32.60.+i, 33.15.Kr, 33.80.−b

I. INTRODUCTION

The experimental measurement of a nonzero electron electric dipole moment (eEDM, d_e) would be a clear signature of physics beyond the standard model [1–4]. The current limit for eEDM, $|d_e| < 9 \times 10^{-29}$ e cm (90% confidence), was set with a buffer-gas-cooled molecular beam [5–7] of thorium monoxide (ThO) molecules in the metastable electronic $H^3\Delta_1$ state. It was shown that due to existence of closely spaced levels of opposite parity of Ω doublet the experiment on ThO is very robust against a number of systematic effects related to magnetic fields [8,9] or geometric phases [10]. However, the upper and lower Ω-doublet states have slightly different properties and systematic effects related to magnetic field imperfections, and geometric phases can still manifest themselves as a false eEDM. The dependence of g factors of the ThO $H^3\Delta_1$ state on Ω doublets and external electric fields was considered in Ref. [9]. The aim of the present work is to consider geometric phase shifts.

II. THEORY

Following the computational scheme of Refs. [9,11], the energies of the rotational levels in the $H^3\Delta_1$ electronic state of the ^{232}Th^{16}O molecule in external *static* electric $\vec{\mathcal{E}} = \mathcal{E}\hat{z}$ and magnetic $\vec{\mathcal{B}} = \mathcal{B}\hat{z}$ fields are obtained by numerical diagonalization of the molecular Hamiltonian ($\hat{\mathbf{H}}_{\rm mol}$) over the basis set of the electronic-rotational wave functions $\Psi_\Omega\theta^J_{M,\Omega}(\alpha,\beta)$. Here Ψ_Ω is the electronic wave function, $\theta^J_{M,\Omega}(\alpha,\beta) = \sqrt{(2J+1)/4\pi}\,D^J_{M,\Omega}(\alpha,\beta,\gamma=0)$ is the rotational wave function, α,β,γ are Euler angles, and M (Ω) is the projection of the molecule angular momentum $\mathbf{J}$ on the laboratory $\hat{z}$ (internuclear $\hat{n}$) axis. Detailed features of the Hamiltonian are described in Ref. [9]. In the paper the $M = \pm 1$ states which represent interest for eEDM search experiment are considered. For electric field $\mathcal{E}$ = 20–200V/cm, used in the experiment, lower rotational levels with $M \neq 0$ can be labeled by $|J,M,\Omega\rangle$ quantum numbers. States $|J,M=1,\Omega=1\rangle$, $|J,M=-1,\Omega=-1\rangle$ correspond to the upper and $|J,M=-1,\Omega=1\rangle$, $|J,M=1,\Omega=-1\rangle$ to the lower Ω-doublet levels. The external magnetic field removes the degeneracy between Ω-doublet components: $\Delta E^u = E(|J,M=1,\Omega=1\rangle) - E(|J,M=-1,\Omega=-1\rangle)$, $\Delta E^l = E(|J,M=1,\Omega=-1\rangle) - E(|J,M=-1,\Omega=1\rangle)$. The relevant energy levels can be seen in Fig. 2 of Ref. [12] or Fig. 3 of Ref. [13]. Provided g factors for upper and lower Ω-doublet levels are close enough that ΔE^u and ΔE^l remain equal unless both parity and time-reversal symmetries are violated. The difference in splitting gives the value for eEDM $d_e = \frac{|\Delta E^l - \Delta E^u|}{4\mathcal{E}_{\rm eff}}$; here $\mathcal{E}_{\rm eff} = 81.5$ GV/cm [12,14] is the effective internal electric field. However, there are systematic effects which can give additional energy shifts $\delta\Delta E^l$ and $\delta\Delta E^u$ for ΔE^l and ΔE^u which manifest as a false eEDM. This leads to a systematic error $\delta d_e(\text{sys}) = \frac{\delta\Delta \mathrm{E^l} - \delta\Delta \mathrm{E^u}}{4\mathcal{E}_{\rm eff}}$. It is also useful to consider systematic effects $\tilde{\delta} d_e(\text{sys}) = \frac{\delta\Delta \mathrm{E^{l(u)}}}{4\mathcal{E}_{\rm eff}}$ related to one of the Ω-doublet components. One of the effects is the interaction with transverse component of the electric field $\vec{\mathcal{E}}(t) = \mathcal{E}_\perp(\hat{x}\cos(\omega_\perp t) + \hat{y}\sin(\omega_\perp t))$, which appears due to spatial inhomogeneties in the applied electric field [13]. Let us consider this effect.

The corresponding part of the Hamiltonian is

$$\hat{\mathbf{H}}_{\rm tf} = -\vec{d}\cdot\vec{\mathcal{E}}(t) = -\mathcal{E}_\perp/2(d_+e^{-i\omega_\perp t} + d_-e^{i\omega_\perp t}), \qquad (1)$$

where $d_\pm = d_x \pm i d_y$. It is more convenient to describe the interaction of the molecule with the quantized electromagnetic fields. The corresponding Hamiltonian is

$$\hat{\mathbf{H}}_{\rm int} = \hbar\omega_\perp a^+ a - \sqrt{\frac{2\pi\hbar\omega_\perp}{V}}(d_+a^+ + d_-a), \qquad (2)$$

where a^+ and a are creation and annihilation operators and V is a volume of the system. To work with Hamiltonian (2) one need to add the quantum number $|n\rangle$, where $n = \frac{V\mathcal{E}_\perp^2}{8\hbar\omega_\perp}$ is number of photons. The approach is similar to the formalism outlined in Ref. [10]. For this paper we consider the case $\mathcal{E}_\perp \ll \mathcal{E}$, such that the additional energy

*alexsandernp@gmail.com

 ©2015 American Physical Society

shifts can be calculated in the framework of the second-order perturbation theory:

$$\delta\Delta E^{u} = \mathcal{E}_{\perp}^{2}/4\times\left(\sum_{J',\Omega'}\frac{|\langle J,M=1,\Omega=1|d_{+}|J',M=0,\Omega'\rangle|^{2}}{E(J,M=1,\Omega=1)-E(J',M=0,\Omega')+\hbar\omega_{\perp}}+\sum_{J',\Omega'}\frac{|\langle J,M=1,\Omega=1|d_{-}|J',M=2,\Omega'\rangle|^{2}}{E(J,M=1,\Omega=1)-E(J',M=2,\Omega')-\hbar\omega_{\perp}}\right.$$
$$\left.-\sum_{J',\Omega'}\frac{|\langle J,M=-1,\Omega=-1|d_{+}|J',M=-2,\Omega'\rangle|^{2}}{E(J,M=-1,\Omega=-1)-E(J',M=-2,\Omega')+\hbar\omega_{\perp}}-\sum_{J',\Omega'}\frac{|\langle J,M=-1,\Omega=-1|d_{-}|J',M=0,\Omega'\rangle|^{2}}{E(J,M=-1,\Omega=-1)-E(J',M=0,\Omega')-\hbar\omega_{\perp}}\right), \tag{3}$$

$$\delta\Delta E^{l} = \mathcal{E}_{\perp}^{2}/4\times\left(\sum_{J',\Omega'}\frac{|\langle J,M=1,\Omega=-1|d_{+}|J',M=0,\Omega'\rangle|^{2}}{E(J,M=1,\Omega=-1)-E(J',M=0,\Omega')+\hbar\omega_{\perp}}+\sum_{J',\Omega'}\frac{|\langle J,M=1,\Omega=-1|d_{-}|J',M=2,\Omega'\rangle|^{2}}{E(J,M=1,\Omega=-1)-E(J',M=2,\Omega')-\hbar\omega_{\perp}}\right.$$
$$\left.-\sum_{J',\Omega'}\frac{|\langle J,M=-1,\Omega=1|d_{+}|J',M=-2,\Omega'\rangle|^{2}}{E(J,M=-1,\Omega=1)-E(J',M=-2,\Omega')+\hbar\omega_{\perp}}-\sum_{J',\Omega'}\frac{|\langle J,M=-1,\Omega=1|d_{-}|J',M=0,\Omega'\rangle|^{2}}{E(J,M=-1,\Omega=1)-E(J',M=0,\Omega')-\hbar\omega_{\perp}}\right). \tag{4}$$

The major contribution to $\delta\Delta E^{u(l)}$ comes from coupling of states with the same J. The most simple is the picture for $J=1$ state. $|J=1,M=1,\Omega,n\rangle$ interact with $|J=1,M=0,\Omega,n-1\rangle$ and $|J=1,M=-1,\Omega,n\rangle$ with $|J,M=0,\Omega,n+1\rangle$. Note that $\hat{\mathbf{H}}_{\rm mol}$ can only couple the states with the same n, whereas $\hat{\mathbf{H}}_{\rm int}$ couples the states with $\Delta n=\pm1$. Energies of states $|J,M=0,\Omega,n+1\rangle$ and $|J,M=0,\Omega,n-1\rangle$ are different by $2\hbar\omega_{\perp}$, and states $|J=1,M=1,\Omega,n\rangle$ and $|J=1,M=-1,-\Omega,n\rangle$ by $2\mu\mathcal{B}$. This leads to different energy denominators in Eqs. (3) and (4) and results in different energy shift for $|J=1,M=1,\Omega,n\rangle$ and $|J=1,M=-1,-\Omega,n\rangle$. However, for $J=1$ level, it was shown in Ref. [13] that (considering the interaction with $|J=1,M=0,\Omega\rangle$ states only) provided tensor Stark ($\Delta E_{ST}=E(J,M=\pm1,\Omega)-E(J,M=0,\Omega)$) and Zeeman splitting are the same for upper and lower component of the Ω doublet the ac Stark shifts $\delta\Delta E^{u}$ and $\delta\Delta E^{l}$ will also be equal. This allows one to reject systematic errors due to geometric phases by performing measurements in both Ω-doublet states. However, the tensor Stark splittings do not coincide exactly. Also including interaction with other states lifts the degeneracy. It is particularly important to include the interaction with the neighbor rotational levels. The latter interaction increases value for δd_e(sys) on several orders of magnitude, whereas $\tilde{\delta}d_e$(sys) is almost unaffected by this interaction. See also influence of perturbation by $J=2$ level on $J=1$ g factors in Refs. [9,11,15,16].

III. RESULTS AND DISCUSSION

Tables I and II list the calculated $\tilde{\delta}d_e$(sys) and δd_e(sys) as a functions of $\omega_{\perp}$ and $\mathcal{E}$ for $J=1$ and $J=2$, correspondingly. Though for smaller $\mathcal{E}$ the $\mathcal{E}_{\perp}$ value will be smaller as well, for the calculation I take the same $\mathcal{E}_{\perp}=10\,$mV/cm given in Ref. [13] for all $\mathcal{E}$. Using the fact that $\tilde{\delta}d_e$(sys) and δd_e(sys) are quadratic functions of $\mathcal{E}_{\perp}$, the results can be easily recalculated for any $\mathcal{E}_{\perp}$. For static magnetic field the value $\mathcal{B}=40\,$mG used in the experiment [5] is used. One can see that δd_e(sys) is two orders of magnitude larger for $J=2$ than for $J=1$ though much smaller than the current limit on $d_{\rm e}$.

Calculation for $\omega_{\perp}/2\pi$ less than 250 kHz is not performed due to the limited computational accuracy. For smaller $\omega_{\perp}$ one can expect further decreasing of $\tilde{\delta}d_e$(sys) and δd_e(sys). Each term in Eqs. (3) and (4) has the form $\frac{b_{u(l)}^2}{a_{u(l)}+\hbar\omega_{\perp}}-\frac{b_{u(l)}^2}{a_{u(l)}-\hbar\omega_{\perp}}$. By retaining terms up to the third order in $\omega_{\perp}$ we have

$$\delta\Delta E^{u(l)}\approx-2\frac{B_{u(l)}^2}{A_{u(l)}}\left(\frac{\hbar\omega_{\perp}}{A_{u(l)}}+\frac{\hbar^3\omega_{\perp}^3}{A_{u(l)}^3}\right). \tag{5}$$

Formulae for $B_{u(l)}$ and $A_{u(l)}$ for $J=1$ are given below. Equation (5) explains the fact that $\delta\Delta E^{u(l)}$ decreases linearly with small $\omega_{\perp}$ listed in Tables I and II. Similar to $\delta\Delta E^{u(l)}$ the major contribution to difference $\delta\Delta E^{l}-\delta\Delta E^{u}$ comes from coupling of states with the same J [terms with $J'=J$ in Eqs. (3) and (4)]. However, an important role is played by the perturbation by the closest rotational levels which makes matrix elements and denominators for upper and lower components of the Ω doublet slightly different. Let us consider this effect for the simplest case, the $J=1$ level. Without perturbation by the $J=2$ level the parameters

$$A=\Delta E_{ST}=-\mathcal{E}\langle J=1,M=1,\Omega|d_z|J=1,M=1,\Omega\rangle$$
$$=-\mathcal{E}dM\Omega/J(J+1) \tag{6}$$

and

$$B=-\mathcal{E}_{\perp}/2\langle J=1,M=1,\Omega|d_{+}|J=1,M'=0,\Omega\rangle$$
$$=-\frac{\mathcal{E}_{\perp}d\Omega}{2}\frac{\sqrt{(J-M+1)(J+M)}}{J(J+1)} \tag{7}$$

up to the sign are the same for upper and lower Ω-doublet levels. ΔE_{ST} is positive for upper and negative for lower levels. Note that dipole moment $d<0$. Equations (5) and (6) explain the fact that $\delta\Delta E^{u(l)}$ decreases quadratically with $\mathcal{E}$.

Perturbation by $J=2$ changes the parameters:

$$A_{u(l)}=\Delta E_{ST}+\delta^{1}\Delta E_{ST}^{u(l)}+\delta^{2}\Delta E_{ST}^{u(l)}, \tag{8}$$
$$B_{u(l)}=B+\delta^{1}B_{u(l)}+\delta^{2}B_{u(l)}. \tag{9}$$

$\delta^{1(2)}\Delta E_{ST}$ is the correction to ΔE_{ST} due to shifting down $|J=1,M=\pm1,\Omega\rangle$ ($|J=1,M=0,\Omega\rangle$) levels when

TABLE I. The $\tilde{\delta}d_e$(sys) (in units 10^{-29} e cm) and δd_e(sys) (in units 10^{-34} e cm) calculated for the $J = 1$ $H^3\Delta_1$ state in ^{232}Th^{16}O.

	$\omega_\perp/2\pi = 4$ MHz		$\omega_\perp/2\pi = 1$ MHz		$\omega_\perp/2\pi = 250$ kHz	
$\mathcal{E}$ (V/cm)	$\tilde{\delta}d_e$(sys)	δd_e(sys)	$\tilde{\delta}d_e$(sys)	δd_e(sys)	$\tilde{\delta}d_e$(sys)	δd_e(sys)
20.0	−1299.0	−2144.0	−313.0	−31.0	−78.0	−0.48
30.0	−565.0	−608.0	−139.0	−9.1	−35.0	−0.14
40.0	−315.0	−253.0	−78.0	−3.8	−19.0	−0.059
50.0	−201.0	−128.0	−50.0	−1.9	−12.0	−0.030
60.0	−139.0	−74.0	−35.0	−1.1	−8.6	−0.018
70.0	−102.0	−47.0	−25.0	−0.70	−6.3	−0.011
80.0	−78.0	−31.0	−19.0	−0.47	−4.8	−0.0073
90.0	−62.0	−22.0	−15.0	−0.34	−3.8	−0.0053
100.0	−50.0	−16.0	−12.0	−0.24	−3.1	−0.0038
110.0	−41.0	−12.0	−10.0	−0.18	−2.6	−0.0028
120.0	−35.0	−9.2	−8.6	−0.14	−2.2	−0.0022
130.0	−30.0	−7.3	−7.3	−0.11	−1.8	−0.0017
140.0	−26.0	−5.8	−6.3	−0.088	−1.6	−0.0014
150.0	−22.0	−4.7	−5.5	−0.071	−1.4	−0.0011
160.0	−20.0	−3.9	−4.9	−0.059	−1.2	−0.00093
170.0	−17.0	−3.2	−4.3	−0.050	−1.1	−0.00078
180.0	−15.0	−2.7	−3.8	−0.043	−0.96	−0.00067
190.0	−14.0	−2.3	−3.4	−0.034	−0.86	−0.00053
200.0	−12.0	−2.0	−3.1	−0.030	−0.77	−0.00047

interacting with $J = 2$. $\delta^1\Delta E_{ST}^{u(l)}$ is negative. It decreases (increases absolute value) ΔE_{ST} for upper (lower) Ω-doublet levels. In turn $\delta^2\Delta E_{ST}^{u(l)}$ is positive. It increases (decreases absolute value) ΔE_{ST} for upper (lower) Ω-doublet levels. $\delta^{1(2)}B$ is the correction to B due to the perturbation of the wave function $|J = 1, M = \pm 1, \Omega\rangle$ ($|J = 1, M = 0, \Omega\rangle$) by $|J = 2, M = \pm 1, \Omega\rangle$ ($|J = 2, M = 0, \Omega\rangle$) one. It is shown in the appendix that for $J = 1$ level the corrections $\delta^1\Delta E_{ST}^{u(l)}$, $\delta^2\Delta E_{ST}^{u(l)}$, $\delta^1 B_{u(l)}$, and $\delta^2 B_{u(l)}$ are correlated in such a way that

$$\frac{B_{u(l)}^2}{A_{u(l)}^2} = \frac{B^2}{A^2}. \tag{10}$$

Equation (10) is correct up to the second order in small parameter $\Delta E_{ST}/\Delta E_{\rm rot}$, where $\Delta E_{\rm rot} = E(J = 1) - E(J = 2)$ is energy difference between the first and second rotational levels. Due to Eq. (10) the linear term in the difference $\delta d_e(\text{sys}) = \frac{\delta\Delta E^{\rm l} - \delta\Delta E^{\rm u}}{4\mathcal{E}_{\rm eff}}$ is canceled and δd_e(sys), in the leading

TABLE II. The $\tilde{\delta}d_e$(sys) (in units 10^{-29} e cm) and δd_e(sys) (in units 10^{-34} e cm) calculated for the $J = 2$ $H^3\Delta_1$ state in ^{232}Th^{16}O.

	$\omega_\perp/2\pi = 4$ MHz		$\omega_\perp/2\pi = 1$ MHz		$\omega_\perp/2\pi = 250$ kHz	
$\mathcal{E}$ (V/cm)	$\tilde{\delta}d_e$(sys)	δd_e(sys)	$\tilde{\delta}d_e$(sys)	δd_e(sys)	$\tilde{\delta}d_e$(sys)	δd_e(sys)
20.0	−1996.0	−213 498.0	−320.0	−1663.0	−78.0	−33.0
30.0	−658.0	−42 436.0	−140.0	−495.0	−35.0	−9.9
40.0	−342.0	−15 657.0	−79.0	−211.0	−20.0	−4.2
50.0	−212.0	−7 546.0	−50.0	−107.0	−13.0	−2.1
60.0	−144.0	−4 229.0	−35.0	−61.0	−8.7	−1.2
70.0	−105.0	−2 613.0	−26.0	−39.0	−6.4	−0.78
80.0	−80.0	−1 728.0	−20.0	−26.0	−4.9	−0.52
90.0	−63.0	−1 204.0	−15.0	−18.0	−3.9	−0.37
100.0	−51.0	−872.0	−13.0	−14.0	−3.1	−0.27
110.0	−42.0	−653.0	−10.0	−10.0	−2.6	−0.20
120.0	−35.0	−501.0	−8.7	−7.7	−2.2	−0.15
130.0	−30.0	−393.0	−7.4	−6.1	−1.9	−0.12
140.0	−26.0	−314.0	−6.4	−4.9	−1.6	−0.098
150.0	−22.0	−255.0	−5.6	−4.0	−1.4	−0.080
160.0	−20.0	−210.0	−4.9	−3.1	−1.2	−0.063
170.0	−17.0	−175.0	−4.3	−2.7	−1.1	−0.055
180.0	−15.0	−147.0	−3.9	−2.2	−0.97	−0.044
190.0	−14.0	−125.0	−3.5	−1.9	−0.87	−0.038
200.0	−13.0	−107.0	−3.1	−1.6	−0.78	−0.032

order, is a cubic function of $\omega_\perp$ for $J=1$. This behavior can be seen from the data in Table I. Dependence of the δd_e(sys) for $J=2$ level on $\omega_\perp$ has also nearly the cubic character.

The calculations confirm that the experiment on ThO $H^3\Delta_1$ state is very robust against systematic errors related to geometric phases. The developed code can be applied for calculation of molecules in an ion trap in the presence of a rotating field [17,18].

The author is grateful to A. V. Titov for useful discussions. Codes for calculations of diatomic molecules were developed with the support of the Russian Science Foundation grant (Project No. 14-31-00022). Calculation of the ThO molecule was performed with the support of Saint Petersburg State University Research Grant No. 0.38.652.2013 and RFBR Grant No. 13-02-0140.

APPENDIX

In the first order in the small parameter $\Delta E_{ST}/\Delta E_{\rm rot} \sim \mathcal{E}d/\Delta E_{\rm rot}$ for $\delta^1\Delta E^{u(l)}_{ST}$, $\delta^2\Delta E^{u(l)}_{ST}$, $\delta^1 B^{u(l)}$, and $\delta^2 B^{u(l)}$ we have

$$\delta^1\Delta E^{u(l)}_{ST} = \mathcal{E}^2/\Delta E_{\rm rot}|\langle J=1,M=1,\Omega|d_z|J'=2,M=1,\Omega\rangle|^2 = \frac{\mathcal{E}^2d^2}{\Delta E_{\rm rot}}\frac{((J+1)^2-M^2)((J+1)^2-\Omega^2)}{(2J+1)(2J+3)(J+1)^2},$$

$$\delta^2\Delta E^{u(l)}_{ST} = -\mathcal{E}^2/\Delta E_{\rm rot}|\langle J=1,M=0,\Omega|d_z|J'=2,M=0,\Omega\rangle|^2 = -\frac{\mathcal{E}^2d^2}{\Delta E_{\rm rot}}\frac{(J+1)^2((J+1)^2-\Omega^2)}{(2J+1)(2J+3)(J+1)^2},$$

$$\begin{aligned}\delta^1 B_{u(l)} &= \mathcal{E}_\perp/2\langle J'=2,M=1,\Omega|d_+|J=1,M'=0,\Omega\rangle \times \mathcal{E}/\Delta E_{\rm rot}\langle J=1,M=1,\Omega|d_z|J'=2,M=1,\Omega\rangle\\ &= -\frac{\mathcal{E}_\perp\mathcal{E}d^2}{2\Delta E_{\rm rot}}\sqrt{\frac{(J+M)(J+M+1)((J+1)^2-\Omega^2)}{(2J+1)(2J+3)(J+1)^2}}\times\sqrt{\frac{((J+1)^2-M^2)((J+1)^2-\Omega^2)}{(2J+1)(2J+3)(J+1)^2}},\end{aligned}$$

$$\begin{aligned}\delta^2 B_{u(l)} &= \mathcal{E}_\perp/2\langle J=1,M=1,\Omega|d_+|J'=2,M'=0,\Omega\rangle \times \mathcal{E}/\Delta E_{\rm rot}\langle J=1,M'=0,\Omega|d_z|J'=2,M'=0\rangle\\ &= \frac{\mathcal{E}_\perp\mathcal{E}d^2}{2\Delta E_{\rm rot}}\sqrt{\frac{(J-M+2)(J-M+1)((J+1)^2-\Omega^2)}{(2J+1)(2J+3)(J+1)^2}}\times\sqrt{\frac{(J+1)^2((J+1)^2-\Omega^2)}{(2J+1)(2J+3)(J+1)^2}}.\end{aligned}$$

$\Delta E_{\rm rot}$ is negative, therefore $\delta^{(1)}\Delta E^{u(l)}_{ST}<0$ and $\delta^{(2)}\Delta E^{u(l)}_{ST}>0$.

Then retaining terms up to the first order in $\Delta E_{ST}/\Delta E_{\rm rot}$ we have

$$\begin{aligned}\frac{B^2_{u(l)}}{A^2_{u(l)}} &= \frac{(B+\delta^1 B_{u(l)}+\delta^2 B_{u(l)})^2}{\left(\Delta E_{ST}+\delta^1\Delta E^{u(l)}_{ST}+\delta^2\Delta E^{u(l)}_{ST}\right)^2} \approx \frac{B^2+2B\delta^1 B_{u(l)}+2B\delta^2 B_{u(l)}}{\Delta E^2_{ST}+2\Delta E_{ST}\delta^1\Delta E^{u(l)}_{ST}+2\Delta E_{ST}\delta^2\Delta E^{u(l)}_{ST}}\\ &= \frac{\frac{\mathcal{E}^2_\perp d^2\Omega^2}{4}\frac{(J-M+1)(J+M)}{(J(J+1))^2}}{\frac{\mathcal{E}^2d^2M^2\Omega^2}{(J(J+1))^2}}\times\frac{\left(1+\left[(J+M+1)-(J+1)\sqrt{\frac{J-M+2}{J+M}}\right]K(J)/\Omega\right)}{(1+MK(J)/\Omega)}\\ &= \frac{B^2\left(1+\left[(J+M+1)-(J+1)\sqrt{\frac{J-M+2}{J+M}}\right]K(J)/\Omega\right)}{A^2(1+MK(J)/\Omega)},\end{aligned} \tag{A1}$$

where

$$K(J) = 2\frac{\mathcal{E}d}{\Delta E_{\rm rot}}\frac{J(J+1)((J+1)^2-\Omega^2)}{(2J+1)(2J+3)(J+1)^2}.$$

Substituting $M=1$, $J=1$ to Eq. (A1), we obtained Eq. (10). Equation (A1) is obtained for $M=+1$ level. The result is the same for $M=-1$. Taking into account that $|J=1,M=+1,\Omega=+1\rangle$ ($|J=1,M=+1,\Omega=-1\rangle$) corresponds to the upper (lower) Ω-doublet level, for the next term we have

$$\frac{B^2_u}{A^4_u} = \frac{B^2(1+K)}{A^4(1+2K)} = \frac{B^2\left(1+\frac{\mathcal{E}d}{5\Delta E_{\rm rot}}\right)}{A^4\left(1+2\frac{\mathcal{E}d}{5\Delta E_{\rm rot}}\right)},$$

$$\frac{B^2_l}{A^4_l} = \frac{B^2(1-K)}{A^4(1-2K)} = \frac{B^2\left(1-\frac{\mathcal{E}d}{5\Delta E_{\rm rot}}\right)}{A^4\left(1-2\frac{\mathcal{E}d}{5\Delta E_{\rm rot}}\right)}.$$

As an example, below are the parameters calculated for $\mathcal{E}=110$ V/cm:

$$\frac{2\hbar B^2_u}{4\mathcal{E}_{\rm eff}A^2_u} = 10.33057851\times10^{-29}\frac{e\ \text{cm}}{2\pi\ \text{MHz}},\quad \frac{2\hbar B^2_l}{4\mathcal{E}_{\rm eff}A^2_l} = 10.33057851\times10^{-29}\frac{e\ \text{cm}}{2\pi\ \text{MHz}},$$

$$\frac{2\hbar^3 B^2_u}{4\mathcal{E}_{\rm eff}A^4_u} = 0.00080156\times10^{-29}\frac{e\ \text{cm}}{(2\pi\ \text{MHz})^3},\quad \frac{2\hbar^3 B^2_l}{4\mathcal{E}_{\rm eff}A^4_l} = 0.00080343\times10^{-29}\frac{e\ \text{cm}}{(2\pi\ \text{MHz})^3}.$$

[1] I. B. Khriplovich and S. K. Lamoreaux, *CP Violation without Strangeness* (Springer, Berlin, 1997).
[2] M. Pospelov and A. Ritz, Ann. Phys. **318**, 119 (2005).
[3] A. V. Titov, N. S. Mosyagin, A. N. Petrov, T. A. Isaev, and D. P. DeMille, Progr. Theor. Chem. Phys. **15**, 253 (2006).
[4] J. L. Feng, Annu. Rev. Nucl. Part. Sci. **63**, 351 (2013).
[5] J. Baron, W. C. Campbell, D. Demille, J. M. Doyle, G. Gabrielse, Y. V. Gurevich, P. W. Hess, N. R. Hutzler, E. Kirilov, I. Kozyryev *et al.*, Science **343**, 269 (2014).
[6] N. R. Hutzler, H.-I. Lu, and J. M. Doyle, Chem. Rev. **112**, 4803 (2012).
[7] D. Patterson and J. M. Doyle, J. Chem. Phys. **126**, 154307 (2007).
[8] D. DeMille, F. Bay, S. Bickman, D. Kawall, L. Hunter, D. Krause, S. Maxwell, and K. Ulmer, AIP Conf. Proceed. **596**, 72 (2001).
[9] 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).
[10] A. Vutha and D. DeMille, arXiv:0907.5116.
[11] A. N. Petrov, Phys. Rev. A **83**, 024502 (2011).
[12] L. V. Skripnikov, A. N. Petrov, and A. V. Titov, J. Chem. Phys. **139**, 221103 (2013).
[13] 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 **43**, 074007 (2010).
[14] L. V. Skripnikov and A. V. Titov, J. Chem. Phys. **142**, 024301 (2015).
[15] S. Bickman, P. Hamilton, Y. Jiang, and D. DeMille, Phys. Rev. A **80**, 023418 (2009).
[16] J. Lee, J. Chen, L. V. Skripnikov, A. N. Petrov, A. V. Titov, N. S. Mosyagin, and A. E. Leanhardt, Phys. Rev. A **87**, 022516 (2013).
[17] A. Leanhardt, J. Bohn, H. Loh, P. Maletinsky, E. Meyer, L. Sinclair, R. Stutz, and E. Cornell, J. Mol. Spectr. **270**, 1 (2011).
[18] H. Loh, K. C. Cossel, M. C. Grau, K.-K. Ni, E. R. Meyer, J. L. Bohn, J. Ye, and E. A. Cornell, Science **342**, 1220 (2013).