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Atomic-level-mixing contribution to the $\mathcal{P}$, $\mathcal{T}$-odd Faraday effect as an enhancement factor in the search for $\mathcal{P}$, $\mathcal{T}$-odd interactions in nature

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Recently, a $\mathcal{P}$, $\mathcal{T}$-odd Faraday effect (PTFE), optical rotation in an external electric field parallel to the light propagation direction, was discussed as a promising tool for observation of the electron electric dipole moment in the intracavity absorption spectroscopy with atoms or molecules. The main mechanism leading to this effect, the linear Stark splitting of atomic levels (PTFE, LS) is well known. This mechanism is similar to the Zeeman splitting mechanism in case of the ordinary Faraday effect, i.e., optical rotation in an external magnetic field. In this paper we analyze the other possible mechanisms of PTFE, in particular, the atomic-level-mixing mechanism (PTFE, LM). In our analysis we perform considering PTFE (as well as the ordinary FE) as a kind of electro- (magneto-) optical circular birefringence. For the ordinary FE the level-mixing mechanism (FE, LM) is known apart from the main Zeeman level splitting mechanism (FE, LS). However, (PTFE, LM) mechanism for the PTFE was never considered in literature. Usually, the (FE, LS) mechanism dominates and the same was expected for (PTFE, LS). We demonstrate, however, that with heavy diatomic molecules, in particular in PbF, the contribution of (PTFE, LM) may essentially exceed the (PTFE, LS). This promises an additional enhancement of $\mathcal{P}$, $\mathcal{T}$-odd effects in experiments with PbF molecule and makes this molecule one of the best candidates for performing such experiments.

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I. INTRODUCTION

A search for the interactions (forces) in the nature violating the fundamental symmetries is the most long-standing fundamental problem in physics. The basic equations of motion in classical and quantum theories are invariant under space-inversion ($\mathcal{P}$), charge-conjugation ($\mathcal{C}$), and time-reflection ($\mathcal{T}$) operations. The $\mathcal{P}$-noninvariant interactions were predicted in [1] and discovered in [2]. In 1964 the violation of combined $\mathcal{CP}$ symmetry was discovered in high-energy physics [3]. Due to the $\mathcal{CPT}$ invariance which is believed to be always true (it is connected to causality) means also the $\mathcal{T}$ violation.

A very promising way to search for the $\mathcal{P}$, $\mathcal{T}$-odd interactions is the experiments with atomic systems. In particular, the existence of the electric dipole moment (EDM) for any particle which is not truly neutral (i.e., does not coincide with itself after charge conjugation) or for the closed system of such particles would mean the violation of both $\mathcal{P}$ and $\mathcal{T}$ symmetries. The examples of truly neutral particles are the photon, Z boson, but the electron, quark, nucleon, nucleus, atom, or molecule may possess an EDM. In 1950 it was first suggested to observe the neutron's EDM by the magnetic resonance method [4]. Later it was found theoretically that the electron EDM (eEDM) should be strongly enhanced in heavy atoms, i.e., the atomic EDM should be much larger than the eEDM [5]. Even stronger enhancement of $\mathcal{P}$, $\mathcal{T}$-odd effects occurs in heavy diatomic heteronuclear molecules. For the molecules with closed electron shells (like TlF) the Schiff moment (connected with nuclear EDM) of the Tl nucleus is enhanced [6], for the molecules with open electronic shells the enhancement occurs for the eEDM [7–10]. In the latter case the enhancement is due to the Λ doubling of the rotational molecular levels, including the ground state. Here Λ is the projection of the total electronic orbital angular momentum on the molecular axis. For heavy molecules Λ should be replaced by Ω, where Ω includes also the projection of the total electron spin. The Λ-doubling enhancement of $\mathcal{P}$-odd effects in heavy diatomic molecules on the basis of neutral currents in the standard model (SM) was first discussed in [11]. The Ω doubling as the origin of enhancement of $\mathcal{P}$, $\mathcal{T}$-odd effects was considered in [7–9]. The larger the value of Ω for the ground (or metastable) electronic state in a certain molecule, the smaller is the splitting between the Ω components with opposite parities and the stronger is the enhancement of the $\mathcal{P}$, $\mathcal{T}$-odd effects. In case of the states with $\Omega = 1$ the enhancement coefficient $\mathcal{K}$ may reach the value $\mathcal{K} = 10^9$.

The most advanced result for the lower bounds for particle's EDM is obtained now by ACME collaboration for eEDM with the ThO molecule [12]. Their result, obtained via the observation of the electron spin precession in an external electric field, reads as $|d_e| < 10^{-29}$ e cm (e is the electron charge). In principle, the $\mathcal{P}$, $\mathcal{T}$-odd interaction of the EDM of the particle with an external electric field is not a unique $\mathcal{P}$, $\mathcal{T}$-odd effect which can be observed in atomic physics. Another such effect is the $\mathcal{P}$, $\mathcal{T}$-odd interaction of an electron with the nucleus in atom or molecule [7,9]. Both effects are indistinguishable in any experiment with any particular atom or molecule, but can be distinguished in a series of experiments with different species [13,14]. It is convenient to express $\mathcal{P}, \mathcal{T}$-odd electron-nucleus interaction via equivalent eEDM $d_e^{\rm eqv}$ which produces the same linear Stark shift of atomic levels in the same external electric field.

Extraction of the information on d_e, $d_e^{\rm eqv}$ from experiments with atoms or molecules is not straightforward: it requires the knowledge of enhancement coefficients for eEDM in atoms (molecules) which can be obtained only theoretically. Evaluation of enhancement coefficients in heavy atoms and especially in heavy diatomic molecules is a very complicated problem which requires the usage of the most powerful modern numerical methods (see a discussion on the subject, for example, in [14,15]).

The latest theoretical evaluation of eEDM on the basis of the SM gives the value $d_e = 10^{-39}$ e cm [16]. In the SM, the scalar-pseudoscalar-type electron-nucleus $\mathcal{P}, \mathcal{T}$-odd interaction yields the leading contribution to paramagnetic atoms [7,9,10]. Estimates with the two-photon model of this interaction give the maximum prediction $d_e^{\rm eqv} \sim 10^{-38}$ e cm [17]. Estimates made with another scalar-pseudoscalar model within the SM (exchange by Higgs boson) [18] also do not give the value $d_e^{\rm eqv}$ larger than predicted in [17]. There exists also a vast literature on extensions of the SM which predict larger values for d_e (see, for example, [19]). However, none of these predictions have been yet confirmed and some of them are ruled out by existing experimental bounds [12].

Since the existing experimental bound is nine orders of magnitude above the theoretical SM predictions, it is desirable to consider another (apart from the electron spin precession in an external electric field employed in [12]) possible way to observe the $\mathcal{P}, \mathcal{T}$-odd effects in atomic physics. One of these ways is the observation of the $\mathcal{P}, \mathcal{T}$-odd Faraday effect, the optical rotation (rotation of the polarization plane of a photon) in an external electric field (PTFE). The ordinary Faraday effect (FE) is the optical rotation in an external magnetic field. The PTFE was first considered in [20] among the other magneto- and electro-optical effects, then mentioned in [8], and studied theoretically and experimentally in [21] (see also the reviews on the subject in [22,23]).

Recently it was suggested to use the fast progress in intracavity absorption spectroscopy (ICAS) for observation of the $\mathcal{P}, \mathcal{T}$-odd effects in atoms and molecules [24–28]. During last decades the optical path length of 70 000 km for the light propagating through an absorber within a cavity was reached [29] and a sensitivity of 10^{-12} rad for observation of birefringence in the cavity experiments [30] was reported. In [31,32] the theoretical simulations of the experiments on the observation of $\mathcal{P}$-odd effects in atoms were described. The techniques which were assumed to be employed in such experiments are very close to what is necessary to observe PTFE. The principal scheme of the proposed experiment can be seen in Fig. 1.

In this paper we analyze the $\mathcal{P}, \mathcal{T}$-odd electro-optical effects which may be used for the observation of $\mathcal{P}, \mathcal{T}$-odd interactions in atoms or molecules or may give an additional contribution to PTFE. In principle, all $\mathcal{P}, \mathcal{T}$-even and -odd magneto-, electro-, and magneto-electro-optical effects were considered in a seminal paper [20]. In this work, written more than 40 years ago, all the known to this time as well as the "new" effects were included in the consideration. Some of those "new" effects were discovered afterwards, the others remain "new" up to now, among the latter ones PTFE. It is important to note that the authors of [20] were interested in the symmetry properties of the effects in liquids where the $\mathcal{T}$ noninvariance may arise due to dissipation of energy. Nevertheless, experiments on the search for $\mathcal{T}$ noninvariance were suggested in gases, liquids, and even solids [10]. It is commonly assumed that the dissipation effects, i.e., interaction of particular atom (molecule) with environment can be made sufficiently small or can be taken into account. This can be achieved easier in gaseous phase, especially with the use of atomic (molecular) beams. In this paper we are interested only in the latter case. Moreover, we will consider only chiral-odd effects. The chiral transformation may be defined as the mirror reflection (M) and is connected with the space inversion via $MR(\pi) = \mathcal{P}$, where $R(\pi)$ is the rotation by an angle π. The circular birefringence is the chiral-odd process since $Mn^{\pm} = n^{\mp}$ and $M(n^+ - n^-) = -(n^+ - n^-)$ but the linear birefringence is not. Here $n^{\pm}$ are the refractive indices for the right (left) circularly polarized light. Below we will consider the chiral-odd (M-odd) birefringence effects, connected with the real parts of refractive indices $\mathrm{Re}(n^+ - n^-)$ and ignore the chiral-odd (M-odd) dichroism effects, corresponding to the imaginary parts of the refractive indices. The experimental accuracy of measurement of optical dichroism (difference between the absorption coefficients for the right and left photons) as a rule is lower than the accuracy of measurement of the optical rotation for the right and left photons. We will consider the optical rotation as a kind of circular birefringence as it was done, for example, in [22,23]. The ordinary Faraday effect (FE) is the chiral-odd magneto-optical birefringence caused by an external magnetic field [23]. It is commonly observed as a rotation of polarization plane for linearly polarized light propagating through a medium in the presence of an external magnetic field oriented along the direction of the light propagation. For description of the chiral-odd processes it is convenient to introduce the photon spin

$$\vec{s}_{\rm ph} = i(\vec{e} \times \vec{e}^*), \tag{1}$$

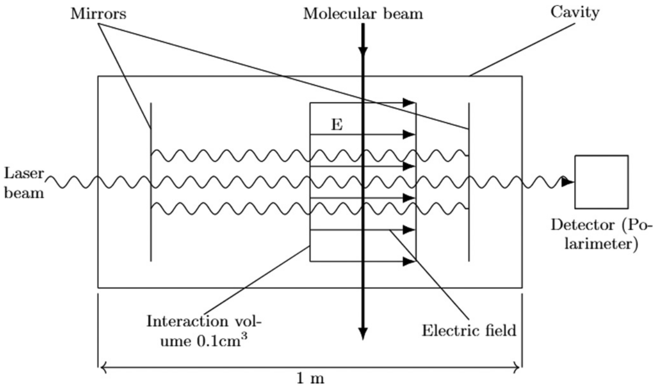

FIG. 1. Principal scheme of the proposed experimental setup. The laser beam travels back and forth within the cavity. In interaction volume there is an electric field oriented along the direction of the laser beam. Through this interaction volume, the molecular beam crosses the laser beam in the transverse direction.

where $\vec{e}$ is the photon polarization vector. For the linear polarization $\vec{s}_{\rm ph}=0$, for the circularly polarized light

$$M\vec{s}^{\pm}_{\rm ph}=\vec{s}^{\mp}_{\rm ph}. \tag{2}$$

The projections $\vec{s}_{\rm ph}\vec{\nu}=\pm 1$ where $\vec{\nu}$ is the unit vector along the light propagation direction are called chiralities. The FE is $\mathcal{P}$, $\mathcal{T}$ even but M odd, the PTFE is $\mathcal{P}$, $\mathcal{T}$ odd and M odd.

Our paper is organized as follows. In Sec. II we give the basic formulas for evaluating the optical rotation in an external electric field ($\mathcal{P}$, $\mathcal{T}$-odd Faraday effect) including both linear Stark level splitting (LS) contribution (PTFE, LS) and level-mixing contribution (PTFE, LM). These formulas are necessary to start more detailed derivations for both contributions in succeeding sections. In Sec. III the detailed derivation of the $\mathcal{P}$, $\mathcal{T}$-odd optical rotation via the linear Stark splitting (PTFE, LS) mechanism is given. The formulas include the relativistic effects since the maximum PTFE rotation is expected for heavy atoms where the relativity is important. Although the (PTFE, LS) mechanism has been already discussed in [24–28], such step-by-step derivation did not yet arrive in the literature. Moreover, all these formulas are necessary to explain the second contribution to PTFE, the $\mathcal{P}$, $\mathcal{T}$-odd level-mixing (PTFE, LM) mechanism. The (PTFE, LM) mechanism never was considered before. The full account for this mechanism is contained in Sec. IV. In Sec. V the comparison of both (PTFE, LS) and (PTFE, LM) mechanisms is made. It appears that in PbF molecule the contribution of (PTFE, LM) to the optical rotation becomes 10 times stronger than the contribution of (PTFE, LS). This gives an order of magnitude additional enhancement to the optical rotation (PTFE). The possible PTFE experiments with PbF were described in [27]. This may be considered as the main result of our studies in this article.

II. OPTICAL ROTATION IN AN ELECTRIC FIELD

It is well known that there are two different mechanisms that lead to the ordinary Faraday effect in an external magnetic field: Zeeman splitting of atomic levels (FE, LS) (we have to note that here, as throughout the text, LS stands for "level splitting") and atomic level mixing in magnetic field (FE, LM) [10,33]. The first mechanism is usually dominant and is due to the absorption of the right and left photons in transitions between different Zeeman sublevels of two atomic states. This leads to the different absorption frequencies and hence to the different refractive indices $n^{\pm}$. The additional contribution (FE, LM) originates from the mixing of atomic levels with different total angular momentum values F (but with the same projections M_F on the magnetic field direction) by an external magnetic field. Magnetic field mixes the states with the same parities, therefore, the main contribution to (FE, LM) comes from the mixture of hyperfine sublevels JF, JF' having the same parity and lying close to each other [10,33].

The similar two contributions exist also in PTFE. The (PTFE, LS) which arises due to the linear Stark splitting of atomic levels with account for the $\mathcal{P}$, $\mathcal{T}$-odd effects is fully similar to the (FE, LS). The other contribution (PTFE, LM) was not yet considered in the literature. This contribution is similar to (FE, LM) and originates from the mixing of atomic levels in an external electric field. Electric field in the absence of $\mathcal{P}$, $\mathcal{T}$-odd interactions mixes the states with opposite parities but with account for $\mathcal{P}$, $\mathcal{T}$-odd interactions mixes JF and JF' states which as in case of (FE, LM) gives the major contribution to (PTFE, LM).

The optical rotation angle connected with the $\mathcal{P}$, $\mathcal{T}$-odd birefringence due to both (PTFE, LS) (level splitting) and (PTFE, LM) (level mixing) mechanisms is given by the general formula

$$\psi=\pi\frac{l}{\lambda}\mathrm{Re}(n^{+}-n^{-}), \tag{3}$$

where l is the optical path length, λ is the wavelength of the propagating light, and $n^{\pm}$ are the refractive indices for the right and left circularly polarized light. In frames of the linear optics the refractive index $n(\omega)$ for any type of the light polarization is defined via the amplitude of the elastic forward photon scattering by the atomic electron $A(\omega)$,

$$n(\omega)=1+2\pi\rho A(\omega), \tag{4}$$

where ω is the photon frequency and ρ is the atomic (molecular) number density.

An expression for the photon scattering amplitude $A(\omega)$ looks like

$$A(\omega)=\sum_{M_F M'_F}\frac{|\langle\gamma JFM_F|\hat{A}|\gamma' J'F'M'_F\rangle|^2}{E_{\gamma' J'F'M'_F}-E_{\gamma JFM_F}-\omega-\frac{i}{2}\Gamma_{\gamma' J'F'}}. \tag{5}$$

Here γJFM_F are the quantum numbers characterizing the state of atomic (molecular) system: J is the total angular momentum of a system (without nuclear spin), F is the total angular momentum including the nuclear spin, M_F is the projection of the momentum $\vec{F}$ on the electric field direction, and γ is the additional set of quantum numbers necessary to characterize the state. Via $E_{\gamma JFM_F}$ we denote the energies of atomic (molecular) system including the linear Stark splitting components labeled by the index M_F. We assume the case of resonant scattering when the photon frequency is close to the energy difference $E_{\gamma' J'F'M'_F}-E_{\gamma JFM_F}$. The linear Stark splitting components are not necessarily resolved, the state γJF is assumed to be the ground state, $\Gamma_{\gamma' J'F'}$ is the natural width of the excited state. The operator $\hat{A}$ in the matrix element is the operator of interaction for the electromagnetic and electron-positron fields. In the dipole approximation for electromagnetic field this operator looks like

$$\hat{A}=\vec{e}\vec{D}^{\rm E} \tag{6}$$

for E1 transition and

$$\hat{A}=(\vec{e}\times\vec{\nu})\vec{D}^{\rm M} \tag{7}$$

for M1 transition. In Eqs. (6) and (7), $\vec{e}$ is the photon polarization vector. $\vec{D}^{\rm E}$ is the electric dipole moment operator for the electron

$$\vec{D}^{\rm E}=e\vec{r}, \tag{8}$$

$\vec{D}^{\rm M}$ is magnetic dipole moment operator for the electron

$$\vec{D}^{\rm M}=\mu_0(\vec{l}+2\vec{s}), \tag{9}$$

e is the electron charge, $\vec{r}$ is the radius vector for atomic electron, μ_0 is the Bohr magneton, and $\vec{l}$ and $\vec{s}$ are the orbital and spin angular momentum operators for atomic electron.

In Eqs. (4) and (5) the contributions of both (PTFE, LS) and (PTFE, LM) mechanisms are incorporated. To introduce the (PTFE, LS) we should write the energies in the denominator of (5) with account for the linear Stark splitting caused by the $\mathcal{P}, \mathcal{T}$-odd interactions:

$$E_{\gamma JFM_F} = E^{(0)}_{\gamma JF} + \Delta E_{\gamma JFM_F}, \tag{10}$$

where $E^{(0)}_{\gamma JF}$ are the energies in the absence of external electric field and $\Delta E_{\gamma JFM_F}$ represents the linear Stark splitting

$$\Delta E_{\gamma JFM_F} = \langle \gamma JFM_F | \vec{d}_e \vec{E} | \gamma JFM_F \rangle \equiv \langle \gamma JF | S | \gamma JF \rangle d_e E M_F. \tag{11}$$

Here $E = |\vec{E}|$. Inserting (11) in (5) and expanding in terms of external field E we find

$$A(\omega) = A^0(\omega) + \sum_{M_F M'_F} |\langle \gamma JIFM_F | \hat{A} | \gamma' J' IF' M'_F \rangle|^2 \times \left[\frac{\langle \gamma' J' IF' | S | \gamma' J' IF' \rangle M'_F}{E^{(0)}_{\gamma' J' IF'} - E^{(0)}_{\gamma JIF} - \omega - \frac{i}{2}\Gamma_{\gamma' J' IF'}} - \frac{\langle \gamma JIF | S | \gamma JIF \rangle M_F}{E^{(0)}_{\gamma JIF} - E^{(0)}_{\gamma' J' IF'} - \omega - \frac{i}{2}\Gamma_{\gamma' J' IF'}} \right] E d_e. \tag{12}$$

In case of (PTFE, LS) in the sum over M_F, M'_F in (12) only the terms with $M'_F - M_F = \pm 1$ contribute to the effect since they correspond to the scattering of the right (left) circularly polarized photons. As a result, the resonances for the right and left photons occur at the different frequencies and the birefringence effect arises.

Another contribution to PTFE comes from the (PTFE, LM) mechanism. To demonstrate this we may note that the external electric field (when $\mathcal{P}, \mathcal{T}$-odd interactions are taken into account) mixes the states with the same space parity, in particular, the components of the hyperfine structure for the ground (or excited) electronic state in case of atoms or the components of the hyperfine structure for the ground (or excited) electronic, vibronic, and rotational states for molecules. The mixing of the hyperfine components gives the major contribution to the (PTFE, LM). To derive the contribution of this type we have to replace the wave functions $\psi_{\gamma JFM_F}$ in the matrix element of the operator $\hat{A}$ in (5) by functions

$$\widetilde{\psi}_{\gamma JFM_F} = \psi_{\gamma JFM_F} + d_e E M_F \sum_{\widetilde{F}} \frac{\langle \gamma JF | S | \gamma J\widetilde{F} \rangle}{E^{(0)}_{\gamma J\widetilde{F}} - E^{(0)}_{\gamma JF}} \psi_{\gamma J\widetilde{F}M_F}. \tag{13}$$

Note that the linear Stark effect in an external homogeneous electric field (which we always assume to be the case in our discussion) mixes only F but not M_F quantum numbers. After insertion of the wave functions (13) into (5) and summation over M_F, M'_F the terms containing the photon spin can be singled out and the (PTFE, LM) contribution proportional to the photon spin

$$i(\vec{e}^* \times \vec{e})\vec{E} = i[(\vec{e}^* \times \vec{\nu}) \times (\vec{e} \times \vec{\nu})]\vec{E} = (\vec{s}_{\text{ph}} \vec{E}) \tag{14}$$

in both cases of E1 and M1 photon arises.

III. LINEAR STARK SPLITTING CONTRIBUTION TO THE $\mathcal{P}, \mathcal{T}$-ODD FARADAY EFFECT

Using (12) and going over from the Lorentz profile to the more realistic Voigt profile we can present the linear Stark contribution to the angle of rotation of polarization plane of the light ψ_{LS} in the following way:

$$\psi_{\text{LS}}(\omega) = 2\pi \frac{l}{\lambda} \rho \sum_{\substack{M_F, M'_F \\ (M_F - M'_F = \pm 1)}} |\langle \gamma JFM_F | \vec{D}^{M,E} | \gamma' J' F' M'_F \rangle|^2 \times \frac{1}{\hbar \Gamma_D} \frac{h(u, v)}{\Gamma_D} [\langle \gamma JFM_F | \vec{d}_e \vec{E} | \gamma JFM_F \rangle + \langle \gamma' J' F' M'_F | \vec{d}_e \vec{E} | \gamma' J' F' M'_F \rangle], \tag{15}$$

where $\vec{E}$ is the external electric field and the vector $\vec{d}_e$ will be defined below. In (15) the Voigt parametrization of the spectral line profile is used [10]:

$$g(u, v) = \text{Im}\mathcal{G}(u, v), \tag{16}$$

$$f(u, v) = \text{Re}\mathcal{G}(u, v), \tag{17}$$

$$h(u, v) = \frac{d}{du} g(u, v), \tag{18}$$

$$\mathcal{G}(u, v) = \sqrt{\pi} e^{-(u+iv)^2} \{1 - \text{Erf}[-i(u + iv)]\}, \tag{19}$$

$$u = \frac{\Delta\omega}{\Gamma_D}, \tag{20}$$

$$v = \frac{\Gamma_n}{2\Gamma_D}. \tag{21}$$

Here $\text{Erf}(x)$ is the error function, $\Delta\omega$ is the detuning, Γ_D is the Doppler width, and Γ_n is the natural width of atomic level. We fully neglect the collisional width as is commonly accepted for the atomic beam experiments [25–28].

The matrix element $\langle \gamma JFM_F | \vec{d}_e \vec{E} | \gamma JFM_F \rangle$ in (15) defines the linear Stark splitting of atomic levels in an external electric field when the $\mathcal{P}, \mathcal{T}$-odd interactions are taken into account. In case of the interaction of the eEDM with an external electric field this matrix element looks like [26,34]

$$\langle \gamma JFM_F | \vec{d}_e \vec{E} | \gamma JFM_F \rangle = -d_e \langle \gamma JFM_F | (\gamma_0 - 1) \vec{E} \vec{\Sigma} | \gamma JFM_F \rangle + d_e e \vec{E} \times \sum_{\gamma'' J'' F'' M''_F} \left[\frac{\langle \gamma JFM_F | \vec{r} | \gamma'' J'' F'' M''_F \rangle \langle \gamma'' J'' F'' M''_F | (\gamma_0 - 1) \vec{E}_c \vec{\Sigma} | \gamma JFM_F \rangle}{E^{(0)}_{\gamma'' J'' F''} - E^{(0)}_{\gamma JF}} + \frac{\langle \gamma JFM_F | (\gamma_0 - 1) \vec{E}_c \vec{\Sigma} | \gamma'' J'' F'' M''_F \rangle \langle \gamma'' J'' F'' M''_F | \vec{r} | \gamma JFM_F \rangle}{E^{(0)}_{\gamma'' J'' F''} - E^{(0)}_{\gamma JF}} \right]. \tag{22}$$

Now we can introduce the vector $\vec{d}_e$:

$$\vec{d}_e = d_e \vec{n}_d, \tag{23}$$

$$\vec{n}_d = (\gamma_0 - 1)\vec{\Sigma} + \sum_{\gamma''J''F''M''_F} \frac{e\vec{r}|\gamma''J''F''M''_F\rangle\langle\gamma''J''F''M''_F|(\gamma_0 - 1)(\vec{E}_c\vec{\Sigma})}{E^{(0)}_{\gamma''J''F''} - E^{(0)}_{\gamma JF}} + \sum_{\gamma''J''F''M''_F} \frac{(\gamma_0 - 1)(\vec{E}_c\vec{\Sigma})|\gamma''J''F''M''_F\rangle\langle\gamma''J''F''M''_F|e\vec{r}}{E^{(0)}_{\gamma''J''F''} - E^{(0)}_{\gamma JF}}, \tag{24}$$

where γ_0, $\vec{\Sigma}$ are the Dirac matrices and $\vec{E}_c$ is the Coulomb field of the nucleus and the other electrons acting on a certain electron in an atom, $\vec{r}$ is the electron radius vector. The factor $(\gamma_0 - 1)$ in the matrix elements in (24) takes into account the Schiff theorem [35] on the screening of eEDM effect in atoms. In the case of the scalar-pseudoscalar electron-nucleus interaction, d_e in (22) should be replaced by d_e^{eqv}.

The vector $\vec{n}_d$ is the pure electronic vector independent on the nuclear variables. Then the dependence on the quantum numbers M_F can be separated out and the expression for ψ_{LS} can be presented in the form

$$\psi_{\text{LS}} = 2\pi \frac{l}{\lambda} \rho \frac{\langle\gamma JF||R||\gamma' J' F'\rangle}{\Gamma_D} \frac{h(u, v)}{\Gamma_D} d_e E, \tag{25}$$

where $E = |\vec{E}|$ and the factor $\langle\gamma JF||R||\gamma' J' F'\rangle$ can be defined as

$$\langle\gamma JF||R||\gamma' J' F'\rangle = \sum_{\substack{M_F, M'_F \\ (M_F - M'_F = \pm 1)}} |\langle\gamma JFM_F|\vec{D}^{M,E}|\gamma' J' F' M'_F\rangle|^2 [\langle\gamma JFM_F|\vec{n}_d\vec{\mathcal{E}}|\gamma JFM_F\rangle + \langle\gamma' J' F' M'_F|\vec{n}_d\vec{\mathcal{E}}|\gamma' J' F' M'_F\rangle]. \tag{26}$$

Here the vector $\vec{n}_d$ was defined in (24) and $\vec{\mathcal{E}} = \frac{\vec{E}}{E}$. To evaluate the rotation angle we have to start with consideration of the matrix element $\langle\gamma JFM_F|\vec{D}^{M,E}|\gamma' J' F' M'_F\rangle$ in (15). The wave function $\psi_{\gamma JFM_F}$ is

$$\psi_{\gamma JFM_F} = \sum_{M_J M_I} C^{JI}_{FM_F}(M_J M_I)\psi_{\gamma JM_J}\psi_{IM_I}, \tag{27}$$

where $\psi_{\gamma JM_J}$ is the electron wave function, JM_J are the total electron angular momentum and its projection, ψ_{IM_I} is the angular part of the nuclear wave function, IM_I are the nuclear spin and its projection, FM_F are the total angular momentum of an atom and its projection, and $C^{JI}_{FM_F}(M_J M_I)$ are the Clebsch-Gordan coefficients. We start with the case of many-electron atom in the one-electron approximation. For description of an atomic electron we employ the one-electron Dirac wave functions

$$\psi_{\gamma JM_J}(\vec{r}) = \begin{pmatrix} g_{nl}(r)\Omega_{JlM_J}(\Omega) \\ if_{n\bar{l}}(r)\Omega_{J\bar{l}M_J}(\Omega) \end{pmatrix}. \tag{28}$$

Here $g_{nl}(r)$ and $f_{nl}(r)$ are the radial parts of the Dirac wave function, n is the principal quantum number, quantum number l determines the parity of the atomic state, $\mathcal{P} = (-1)^l$, $\bar{l} = 2J - 1$. The angular part is represented by spherical spinors

$$\Omega_{JM_J}(\Omega) = \sum_{m_l m_s} C^{l\frac{1}{2}}_{JM_J}(m_l, m_s)Y_{lm_l}(\Omega)\eta_{m_s}, \tag{29}$$

$Y_{lm_l}(\Omega)$ is the spherical function and η_{m_s} is the electron spin function. The numbers $l,\ m_l,\ m_s$ denote the electron orbital angular momentum, its projection, and the electron spin projection, respectively. By Ω the spherical coordinates $\Omega = \theta, \varphi$ are denoted. Using spherical components of the vector $\vec{D}^{M,E}$ we can write

$$|\langle\gamma JFM_F|\vec{D}^{M,E}|\gamma' J' F' M'_F\rangle|^2 = \sum_{q=0,\pm 1} (-1)^q \big\langle\gamma JFM_F\big|D^{M,E}_q\big|\gamma' J' F' M'_F\big\rangle^* \big\langle\gamma JFM_F\big|D^{M,E}_{-q}\big|\gamma' J' F' M'_F\big\rangle, \tag{30}$$

where $D^{M,E}_q$ is the component q of the spherical tensor of the rank 1. Inserting the wave functions (29) in the matrix elements in (30) we obtain

$$\begin{aligned}\big\langle\gamma JFM_F\big|D^{M,E}_q\big|\gamma' J' F' M'_F\big\rangle = {} & \sum_{M_J M'_J}\sum_{M_I M'_I}\sum_{mm'}\sum_{m_s m'_s} C^{JI}_{FM_F}(M_J, M_I)C^{J'I}_{F'M'_F}(M'_J M'_I) \\ & \times \Big[C^{l\frac{1}{2}}_{JM_J}(m_l m_s)C^{l'\frac{1}{2}}_{J'M'_J}(m'_l m'_s)\Big\langle nlm_l\frac{1}{2}m_s\Big|D^{M,E}_q\Big|n'l'm'_l\frac{1}{2}m'_s\Big\rangle \\ & + C^{\bar{l}\frac{1}{2}}_{JM_J}(m_l m_s)C^{\bar{l}'\frac{1}{2}}_{J'M'_J}(m'_l m'_s)\Big\langle n\bar{l}m_l\frac{1}{2}m_s\Big|D^{M,E}_q\Big|n'\bar{l}'m'_l\frac{1}{2}m'_s\Big\rangle\Big] \times \langle IM_I|IM'_I\rangle.\end{aligned} \tag{31}$$

Here γ represents the other possible quantum numbers depending on the chosen coupling scheme. For simplicity here we will restrict ourselves with the atomic configurations with one electron in the outer-electron nonclosed shell. Then, introduction of γ is unnecessary. Extension of the results to the arbitrary atomic configurations can be easily done by the standard methods employed in atomic theory.

In (31) we have taken into account that the operator $D_q^{M,E}$ does not depend on the nuclear variables. Then

$$\langle IM_I|IM_I'\rangle = \delta_{M_IM_I'}. \tag{32}$$

We consider first the electric dipole operator, when $D_q^E = e(\vec{r})_q$, $(\vec{r})_q$ are the spherical components of radius vector of the electron. After integration (summation) over spin variables in (31) we obtain

$$\left\langle nlm_l\frac{1}{2}m_s\middle|r_q^1\middle|n'l'm_l'\frac{1}{2}m_s'\right\rangle = \left\langle nlm_l\middle|r_q^1|n'l'm_l'\right\rangle\delta_{m_sm_s'}, \tag{33}$$

$$\left\langle n\bar{l}m_l\frac{1}{2}m_s|r_q^1|n'\bar{l}'m_l'\frac{1}{2}m_s'\right\rangle = \langle n\bar{l}m_l|r_q^1|n'\bar{l}'m_{\bar{l}}'\rangle\delta_{m_sm_s'}. \tag{34}$$

In (33) the radial parts of the Dirac wave functions are $g_{nl}(r)$ and in (34) the radial wave functions are $f_{nl}(r)$. For the evaluation of the matrix elements $\langle nlm_l|r_q^1|n'l'm'\rangle$ in (33) and $\langle n\bar{l}m_l|r_q^1|n'\bar{l}'m'\rangle$ in (34) we can apply the Wigner-Eckart theorem [36].

After summation over all the angular momenta projections but M_F, M_F' this yields

$$\begin{aligned}&\langle\gamma JFM_F|D_q^E|\gamma'J'F'M_F'\rangle\\ &\quad= \begin{pmatrix}F & F' & 1\\ M_F & \overline{M}_F' & q\end{pmatrix}\begin{Bmatrix}J' & J & 1\\ F & F' & I\end{Bmatrix}Q^E_{nJl,n'J'l'},\end{aligned} \tag{35}$$

$$Q^E_{nJl,n'J'l'} = eG^E_{nl,n'l'}\begin{Bmatrix}J & J' & 1\\ l' & l & \frac{1}{2}\end{Bmatrix} + eF^E_{n\bar{l},n'\bar{l}'}\begin{Bmatrix}J & J' & 1\\ \bar{l}' & \bar{l} & \frac{1}{2}\end{Bmatrix}. \tag{36}$$

Here $G^E_{nl,n'l'}$ and $F^E_{n\bar{l},n'\bar{l}'}$ are

$$G^E_{nl,n'l'} = \int_0^\infty r^3dr\, g_{nl}(r)g_{n'l'}(r), \tag{37}$$

$$F^E_{n\bar{l},n'\bar{l}'} = \int_0^\infty r^3dr\, f_{n\bar{l}}(r)f_{n'\bar{l}'}(r). \tag{38}$$

Similarly, we can consider the magnetic dipole operator $D_q^M = \mu_q^1$, μ_q^1 are the spherical components of the electron magnetic moment:

$$\mu_q^1 = \mu_0\left(l_q^1 + 2s_q^1\right). \tag{39}$$

In (39), l_q^1 are the components of the electron orbital angular momentum and s_q^1 are the components of the electron spin momentum. After using the Wigner-Eckart theorem and summation over all angular momenta projections we obtain

$$\langle\gamma JFM_F|D_q^M|\gamma'J'F'M_F'\rangle = \begin{pmatrix}F & F' & 1\\ M_F & \overline{M}_F' & q\end{pmatrix}\begin{Bmatrix}J' & J & 1\\ F & F' & I\end{Bmatrix}Q^M_{nJl,n'J'l'}, \tag{40}$$

$$\begin{aligned}Q^M_{nJl,n'J'l'} = {}&\mu_0G^M_{nl,n'l'}\left[\begin{Bmatrix}J & J' & 1\\ l' & l & \frac{1}{2}\end{Bmatrix}\delta_{ll'}\sqrt{l(l+1)(2l+1)} + \begin{Bmatrix}J' & J & 1\\ \frac{1}{2} & \frac{1}{2} & l\end{Bmatrix}\delta_{ll'}\sqrt{\frac{3}{2}}\right]\\ &+\mu_0F^M_{n\bar{l},n'\bar{l}'}\left[\begin{Bmatrix}J & J' & 1\\ \bar{l}' & \bar{l} & \frac{1}{2}\end{Bmatrix}\delta_{\bar{l}\bar{l}'}\sqrt{\bar{l}(\bar{l}+1)(2\bar{l}+1)} + \begin{Bmatrix}J' & J & 1\\ \frac{1}{2} & \frac{1}{2} & \bar{l}\end{Bmatrix}\delta_{\bar{l}\bar{l}'}\sqrt{\frac{3}{2}}\right],\end{aligned} \tag{41}$$

$$G^M_{nl,n'l'} = \int_0^\infty r^2dr\, g_{nl}(r)g_{n'l'}(r), \tag{42}$$

$$F^M_{n\bar{l},n'\bar{l}'} = \int_0^\infty r^2dr\, f_{n\bar{l}}(r)f_{n'\bar{l}'}(r), \tag{43}$$

$$G^M_{nl,n'l'} + F^M_{n\bar{l},n'\bar{l}'} = \delta_{nn'}\delta_{ll'}. \tag{44}$$

Inserting (35) or (40) in (26) we get

$$\begin{aligned}\langle\gamma JF||R||\gamma'J'F'\rangle = {}&\sum_{\substack{M_F,M_F'\\ (M_F-M_F'=\pm1)}}\begin{pmatrix}F & \frac{F'}{\overline{M}_F+1} & \frac{1}{1}\\ M_F & & \end{pmatrix}^2\begin{Bmatrix}J' & J & 1\\ F & F' & I\end{Bmatrix}^2\left(Q^{E,M}_{nJl,n'J'l'}\right)^2\\ &\times\left[\begin{Bmatrix}J & J & 1\\ F & F & I\end{Bmatrix}\langle nJl|n^1_{d0}|nJl\rangle + \begin{Bmatrix}J' & J' & 1\\ F' & F' & I\end{Bmatrix}\langle n'J'l'|n^1_{d0}|n'J'l'\rangle\right],\end{aligned} \tag{45}$$

where $\langle nJl|n^1_{d0}|nJl\rangle$ and $\langle n'J'l'|n^1_{d0}|n'J'l'\rangle$ are the enhancement coefficients. These coefficients for various atoms were calculated in [25,26]. In (45) n^1_{d0} is the projection of the vector $\vec{n}_d$ on the direction of electric field $\vec{E}$.

IV. LEVEL-MIXING CONTRIBUTION TO THE $\mathcal{P}, \mathcal{T}$-ODD FARADAY EFFECT

In (15) for the $\psi_{\rm LS}$ rotation angle the dependence on the photon polarization $\vec{e}$ was omitted since the result does not depend on the particular choice of $\vec{e}$. Now we have to retain the dependence on the photon polarizations in the expression for $A(\omega)$ since they are needed for obtaining the birefringence contribution. We will consider only M1 transition; the derivations for E1 transition are fully similar. Then, we come to an expression

$$\begin{aligned} A(\omega) = {} & \sum_{M_F M'_F} \frac{|\langle \gamma J F M_F|(\vec{e}\times\vec{\nu})\vec{D}^M|\gamma' J' F' M'_F\rangle|^2}{\omega^{(0)}_{\gamma' J' F',\gamma J F} - \omega - \frac{i}{2}\Gamma_{\gamma' J' F'}} + \sum_{M_F M'_F} \frac{1}{\omega^{(0)}_{\gamma' J' F',\gamma J F} - \omega - \frac{i}{2}\Gamma_{\gamma' J' F'}} \\ & \times \Bigg\{ 2\,\mathrm{Re}\Bigg[\sum_{\widetilde{F}} \frac{\langle \gamma J F M_F|\vec{d}_e\vec{E}|\gamma J \widetilde{F} M_F\rangle}{E^{(0)}_{\gamma J\widetilde{F}} - E^{(0)}_{\gamma J F}} \langle \gamma J \widetilde{F} M_F|(\vec{e}\times\vec{\nu})\vec{D}^M|\gamma' J' F' M'_F\rangle \langle \gamma J F M_F|(\vec{e}\times\vec{\nu})\vec{D}^M|\gamma' J' F' M'_F\rangle^* \Bigg] \\ & + 2\,\mathrm{Re}\Bigg[\sum_{\widetilde{F}'} \frac{\langle \gamma' J' \widetilde{F}' M'_F|\vec{d}_e\vec{E}|\gamma' J' F' M'_F\rangle}{E^{(0)}_{\gamma' J'\widetilde{F}'} - E^{(0)}_{\gamma' J' F'}} \langle \gamma J \widetilde{F} M_F|(\vec{e}\times\vec{\nu})\vec{D}^M|\gamma' J' F' M'_F\rangle \langle \gamma J F M_F|(\vec{e}\times\vec{\nu})\vec{D}^M|\gamma' J' F' M'_F\rangle^* \Bigg]\Bigg\}. \end{aligned} \tag{46}$$

Here, $\omega^{(0)}_{\gamma' J' F',\gamma J F} = E^{(0)}_{\gamma' J' F'} - E^{(0)}_{\gamma J F}$, $\vec{e}$ is the photon polarization, and $\vec{\nu}$ is the unit vector of its propagation direction which coincides with the direction of the external electric field. The summation over M_F, M'_F is extended over all possible values since the birefringence contribution is not connected with the special choice of Stark sublevels (all of them contribute to the effect).

Only the second and the third terms in the right-hand side of (46) contribute to the optical rotation. The matrix elements in the numerator of the second term in the right-hand side of (46) are

$$\begin{aligned} 2\,\mathrm{Re}\sum_{q q' q''} & \big\langle \gamma J F M_F\big|D^M_q(\vec{e}\times\vec{\nu})_{-q}\big|\gamma' J' F' M'_F\big\rangle \big\langle \gamma' J' F' M'_F\big|D^M_{q'}(\vec{e}\times\vec{\nu})_{-q'}\big|\gamma J \widetilde{F} M_{\widetilde{F}}\big\rangle^* \\ & \times \big\langle \gamma J \widetilde{F} M_{\widetilde{F}}\big|d^1_{eq''}E^1_{-q''}\big|\gamma J F M_F\big\rangle. \end{aligned} \tag{47}$$

Employing the Wigner-Eckart theorem and performing the summation over M_F, $M_{\widetilde{F}}$, M'_F in (47) gives

$$\sum_{M_F, M_{\widetilde{F}}, M'_F} \begin{pmatrix} F & F' & 1 \\ M_F & M'_F & q \end{pmatrix} \begin{pmatrix} \widetilde{F} & F' & 1 \\ M_{\widetilde{F}} & M'_F & q' \end{pmatrix} \begin{pmatrix} F & \widetilde{F} & 1 \\ M_F & M_{\widetilde{F}} & q'' \end{pmatrix} = \begin{Bmatrix} 1 & 1 & 1 \\ F' & \widetilde{F} & F \end{Bmatrix} \begin{pmatrix} 1 & 1 & 1 \\ q & q' & q'' \end{pmatrix}. \tag{48}$$

Then, using the definition of the vector product of two vectors $\vec{A}$ and $\vec{B}$ in terms of the irreducible tensors [36]

$$\sum_{q q'} \begin{pmatrix} 1 & 1 & 1 \\ q & q' & q'' \end{pmatrix} A_q B_{q'} = -\frac{i}{2}(-1)^{q''}(\vec{A}\times\vec{B})_{q''} \tag{49}$$

and the same for the scalar product

$$\sum_q (-1)^q A_q B_{-q} = (\vec{A}\vec{B}) \tag{50}$$

and using (40) we get for the numerator in the second term in (46) (note that the denominator is now independent on M_F) an expression

$$\begin{aligned} (\vec{s}_{\rm ph}\vec{E}) & \begin{Bmatrix} 1 & 1 & 1 \\ F' & \widetilde{F} & F \end{Bmatrix} \begin{Bmatrix} J' & J & 1 \\ F & F' & I \end{Bmatrix} \begin{Bmatrix} J' & J & 1 \\ \widetilde{F} & F' & I \end{Bmatrix} \begin{Bmatrix} J' & J & 1 \\ F & \widetilde{F} & I \end{Bmatrix} \\ & \times \big(Q^M_{nJl,n'J'l'}\big)^2 \big\langle nJl\big|n^1_{d0}\big|nJl\big\rangle. \end{aligned} \tag{51}$$

The same operations can be done also for the third term in the right-hand side of (46). Introducing the Voigt profile for the optical rotation angle caused by (PTFE, LM) we can write

$$\psi_{\rm LM}(\omega) = 2\pi\frac{l}{\lambda}\rho\left\{\sum_{\widetilde{F}} \frac{\langle \gamma J F, \gamma J \widetilde{F}||T||\gamma' J' F'\rangle}{E^{(0)}_{\gamma J\widetilde{F}} - E^{(0)}_{\gamma J F}} + \sum_{\widetilde{F}'} \frac{\langle \gamma J F||T||\gamma' J' F', \gamma' J' \widetilde{F}'\rangle}{E^{(0)}_{\gamma' J'\widetilde{F}'} - E^{(0)}_{\gamma' J' F'}}\right\} \frac{g(u,v)}{\Gamma_D} d_e E, \tag{52}$$

where

$$\langle \gamma J F, \gamma J \widetilde{F}||T||\gamma' J' F'\rangle = 2 \begin{Bmatrix} 1 & 1 & 1 \\ F' & \widetilde{F} & F \end{Bmatrix} \begin{Bmatrix} J' & J & 1 \\ F & F' & I \end{Bmatrix} \begin{Bmatrix} J' & J & 1 \\ \widetilde{F} & F' & I \end{Bmatrix} \begin{Bmatrix} J' & J & 1 \\ F & \widetilde{F} & I \end{Bmatrix} \big(Q^M_{nJl,n'J'l'}\big)^2 \big\langle nJl\big|n^1_{d0}\big|nJl\big\rangle, \tag{53}$$

$$\langle \gamma JF||T||\gamma' J' F', \gamma' J' \widetilde{F}'\rangle = 2\begin{Bmatrix} 1 & 1 & 1 \\ F & F' & \widetilde{F}' \end{Bmatrix}\begin{Bmatrix} J' & J & 1 \\ F & F' & I \end{Bmatrix}\begin{Bmatrix} J' & J & 1 \\ F & \widetilde{F}' & I \end{Bmatrix}\begin{Bmatrix} J' & J & 1 \\ F' & \widetilde{F}' & I \end{Bmatrix}\left(Q^{M}_{nJl,n'J'l'}\right)^2 \langle n'J'l'|n^1_{d0}|n'J'l'\rangle. \quad (54)$$

The same result with the replacement $Q^M \rightarrow Q^E$ can be obtained for E1 transitions.

V. RESULTS AND DISCUSSION

In the real situation energy denominator in the expression for the rotation angle due to (PTFE, LS) in the resonance case should be replaced by the Doppler width Γ_D. In the expression for the rotation angle due to (PTFE, LM) the energy denominator will contain the hyperfine structure interval $\Delta_{\text{HFS}} = E^{(0)}_{\gamma J\widetilde{F}} - E^{(0)}_{\gamma JF}$. Assuming that the matrix elements for (PTFE, LS) and (PTFE, LM) are approximately the same apart from the pure angular factor $\mathcal{F}(JF\widetilde{F}, J'F'\widetilde{F}')$ we obtain an estimate for the ratio $\psi_{\text{LM}}/\psi_{\text{LS}}$:

$$\frac{\psi_{\text{LM}}}{\psi_{\text{LS}}} = \xi(JF\widetilde{F}, J'F'\widetilde{F}')\frac{\Gamma_D}{\Delta_{\text{HFS}}}, \quad (55)$$

where

$$\xi(JF\widetilde{F}, J'F'\widetilde{F}') = \frac{\mathcal{F}_{\text{LM}}(JF\widetilde{F}, J'F'\widetilde{F}')}{\mathcal{F}_{\text{LS}}(JF\widetilde{F}, J'F'\widetilde{F}')}. \quad (56)$$

Here $\widetilde{F}$ and $\widetilde{F}'$ are the admixed hyperfine levels for the ground and excited states. In [27] it was found that one of the best candidates for the $\mathcal{P}, \mathcal{T}$-odd Faraday experiment on the search for eEDM is the PbF molecule. The details of calculations in Secs. III and IV refer to the atomic systems. Although the formulas (55) and (56) (obtained by the perturbation theory) strictly speaking are not valid anymore for molecular systems, the size of the effect will have the same value $\frac{\Gamma_D}{\Delta_{\text{HFS}}}$ by order of magnitude.

In order to provide the comparison of contributions and describing the dependence of the optical rotation angles $\psi(\omega)$ on ω with the Voigt parametrization we can rewrite (3) for (PTFE, LS) and (PTFE, LM) in the form

$$\psi_{\text{LS}} = C(\mathcal{A}_{\text{PTFE, LS}} + \mathcal{A}'_{\text{PTFE, LS}})\frac{d_e E}{\Gamma_D}h(u, v), \quad (57)$$

$$\psi_{\text{LM}} = C\left(\frac{\mathcal{A}_{\text{PTFE, LM}}}{\Delta_{\text{HFS}}} + \frac{\mathcal{A}'_{\text{PTFE, LM}}}{\Delta'_{\text{HFS}}}\right)d_e E g(u, v), \quad (58)$$

where $E^{(0)}_{\gamma J\widetilde{F}} - E^{(0)}_{\gamma JF} \equiv \Delta_{\text{HFS}}$, $E^{(0)}_{\gamma' J'\widetilde{F}'} - E^{(0)}_{\gamma' J'F'} \equiv \Delta'_{\text{HFS}}$. Here, Γ_D is the Doppler width and Δ_{HFS} is the hyperfine structure interval. The coefficient C depends on the resonant transition frequency, on the optical path length, on the atomic number density, and on the transition rate and is approximately the same for both contributions to the rotation angle in the $\mathcal{P}, \mathcal{T}$-odd Faraday effects for particular transition in a particular atom (molecule). The coefficients $\mathcal{A}_{\text{PTFE, LS}}$, $\mathcal{A}'_{\text{PTFE, LS}}$, $\mathcal{A}_{\text{PTFE, LM}}$, $\mathcal{A}'_{\text{PTFE, LM}}$ denote angular factors defined in Eqs. (45), (53), and (54). The Voigt parametrization functions $g(u, v)$ and $h(u, v)$ at small detunings are of the same order of magnitude. However, as it was shown in [25], the optimal detuning for performing the $\mathcal{P}, \mathcal{T}$-odd Faraday experiment would be $\Delta\omega \approx 5\Gamma_D$. An asymptotic behavior of g and h functions at $u \rightarrow \infty$ is $g \sim \frac{1}{u}$, $h \sim \frac{1}{u^2}$, so we can consider $\frac{g(u,v)}{h(u,v)} \approx 5$.

We suggest using $X1^2\Pi_{\frac{1}{2}} - X2^2\Pi_{\frac{3}{2}}$ transition in PbF with the nucleus ^{207}Pb. Enhancement coefficients are $\mathcal{K}(X1) = 38\times10^5$ and $\mathcal{K}(X2) = 9.3\times10^5$ [27]. In spite of large values of hyperfine structure $A^{\text{Pb}}_{\parallel} = 10\,146$ MHz, $A^{\text{Pb}}_{\perp} = -7264$ MHz, and Ω doubling $E_p = -0.138\,27$ cm^{-1} constants for the ground state of ^{207}PbF [37,38] it was shown [39–41] that a coincidental near degeneracy of levels of opposite parity in the ground rotational state $J = \frac{1}{2}$ for ^{207}PbF takes place. It is caused by near cancellation of energy shifts due to Ω-type doubling and the ^{207}Pb ^{19}F magnetic hyperfine interaction shifts with account for the nuclear spin of ^{19}F atom. As a result, there are closely spaced Ω-doublet hyperfine states $F^p = \frac{3}{2}^-$, $F^p = \frac{1}{2}^-$, $F^p = \frac{1}{2}^+$, and $F^p = \frac{3}{2}^+$ lying within energy interval of about $\Delta_{\text{HFS}} = 300$ MHz. The levels are the second, third, fourth, and fifth energy levels of the ground rotational state of $X^2\Pi_{1/2}$. The relevant energy levels can be seen in Fig. 1 of Ref. [39]. Formally, the quantum numbers $F^p = \frac{1}{2}, \frac{3}{2}$ can be obtained by introducing the total nuclear spin $I = 1$ ($I = \frac{1}{2}$ for ^{207}Pb and ^{19}F). The effect for $X^2\Pi_{3/2}$ is negligible as the molecule is completely polarized. To estimate the ratio of (PTFE, LS) and (PTFE, LM) contributions we can introduce an "effective atom" with the same quantum numbers: total angular momentum values $F = F' = \frac{1}{2}$, $\widetilde{F} = \widetilde{F}' = \frac{1}{2}$, $M_F = \frac{1}{2}$ and total "electronic" angular momentum values $J = J' = \frac{1}{2}$. All $3j$ symbols in Eqs. (45), (53), and (54) should be the same for such an atom and PbF molecule in the chosen state. For the rough estimate we can assume that the integrals in Eqs. (45), (53), and (54) are of the same order of magnitude for atoms and molecules and also of the same order of magnitude for LS and LM contributions. For the simple integrals in Eqs. (45), (53), and (54) this should be the case, though an accurate molecular calculation of both LS and LM contributions is still required. After substitution, the values $J = J' = \frac{1}{2}$, $F = F' = \frac{1}{2}$, $\widetilde{F} = \widetilde{F}' = \frac{1}{2}$, $M_F = \frac{1}{2}$ into Eqs. (45), (53), and (54) we get

$$\xi = \frac{\psi_{\text{LM}}}{\psi_{\text{LS}}} \approx 10. \quad (59)$$

This number demonstrates that the (PTFE, LM) contribution essentially exceeds the (PTFE, LS) in the PbF molecule which therefore can be considered as a promising candidate for the search for $\mathcal{P}, \mathcal{T}$-odd effects (interactions) in the nature. This result is evident from (55) due to the unusual smallness of Δ_{HFS} in PbF molecule comparing to the ordinary values of Δ_{HFS} in heavy atoms (~10^3 MHz). Summarizing, we can state that the diatomic heavy molecules seem to be the most suitable objects for the search of $\mathcal{P}, \mathcal{T}$-odd effects in atomic physics. PTFE-ICAS experiments with these molecules may become an important tool for this search.

Theoretically, as it was shown in this paper, $\mathcal{P}, \mathcal{T}$-odd effects in such molecules arise due to the two different mechanisms: LS and LM. All earlier theoretical predictions for the magnitude of $\mathcal{P}, \mathcal{T}$-odd effects in PTFE-ICAS experiments with heavy diatomic molecules were based on LS, LM was ignored. We have shown that in some special cases, in particular in PbF molecule, the LM contribution may become even stronger than LS, thus enhancing essentially the total expected experimental result. We should note that preliminary results for the possible observation of (PTFE, LM) in atoms (where these effects are much weaker than in molecules) were reported in [42].

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