On the Search for the Electric Dipole Moment of the Electron: $\mathscr{P}$-, $\mathscr{T}$-Odd Faraday Effect on a PbF Molecular Beam

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It has been proposed to measure the electric dipole moment of the electron on the basis of the observation of the $\mathscr{P}$-, $\mathscr{T}$-odd Faraday effect on a molecular beam intersecting a cavity using cavity-enhanced intracavity laser absorption spectroscopy. The effective electric field acting on the electric dipole moment of the electron, as well as the molecular parameter of the scalar–pseudoscalar nucleus–electron interaction, has been calculated for the ground and excited electronic states of the lead monofluoride molecule. The simulation of the experiment has shown that the proposed approach allows improving the current bounds on the electric dipole moment of the electron and the scalar–pseudoscalar nucleus–electron interaction constant by six orders of magnitude.

1. INTRODUCTION

The presence of the electric dipole moment of the electron (*e*EDM) simultaneously violates the space inversion ($\mathscr{P}$) and time reversal ($\mathscr{T}$) symmetries of fundamental interactions [1–3]. The best bound on the *e*EDM $d_e < 1.1 \times 10^{-29}$ *e* cm [4] was obtained on the ThO molecule. The extraction of the *e*EDM from experimental data requires a theoretical calculation of the enhancement factors in the case of atoms and effective electric fields in the case of molecules (see, e.g., [5, 6]).

The *e*EDM should be measured on molecules with open electron shells with a nonzero electron momentum projection on the molecular axis. It is noteworthy that the same experiments can provide bound on the dimensionless scalar–pseudoscalar nucleus–electron $\mathscr{P}$-, $\mathscr{T}$-odd interaction constant [7–9]. To distinguish effects caused by this interaction and effects induced by the *e*EDM, it is necessary to perform a series of experiments with various atoms or molecules [6, 10].

The existing experiments on molecule or molecular ion beams in a trap are based on the measurement of electron spin precession [4, 11, 12] in external electric fields. The bound on the *e*EDM established in these experiments is already at the level of predictions of various extensions of the Standard Model [13]. However, the *e*EDM estimated within the Standard Model is about nine orders of magnitude below this bound [14, 15]. Therefore, it is of particular importance to develop alternative approaches and principles of the measurement of the *e*EDM involving atoms and molecules.

The Faraday effect, i.e., the optical rotation of the plane of polarization of linearly polarized light propagating in a medium in an external magnetic field, is well studied. However, the so-called $\mathscr{P}$-, $\mathscr{T}$-odd Faraday effect can be considered in the presence of the $\mathscr{P}$-, $\mathscr{T}$-odd interaction in atoms or molecules of the medium, where optical rotation is caused by the external electric field [16–19]. We recently proposed to observe this effect on atoms by cavity-enhanced intracavity absorption spectroscopy (ICAS) [20–22]. Theoretical calculations and simulation of this effect for various atoms oriented to the application of ICAS were performed in [23–26]. In this work, we analyze the possibility of such an experiment on the observation of the $\mathscr{P}$-, $\mathscr{T}$-odd Faraday effect on a PbF molecular beam.

We chose this molecule because it is quite well studied. It exhibits the $X1\ ^2\Pi_{1/2} \rightarrow X2\ ^2\Pi_{3/2}$ transition at the wavelength $\lambda = 1210$ nm, which is in the range available for modern lasers. A small width of this transition (collisional broadening can be neglected in experiments with beams) is also an important advantage. To polarize the PbF molecule, one can use an experimentally available external electric field of about 10^4 V/cm, which can be produced at distances of about a centimeter.

We propose the following experiment. Let a molecular beam transversely intersect a laser beam propa-

gating along a cavity. The region of interaction between the molecular and laser beams is placed in the external electric field oriented along the light propagation direction. The typical transverse size of the cavity is 1 m. However, the cavity-enhanced optical path length can reach 100 km [20] or even 70 000 km [21]. Since the region of interaction of molecules with the laser beam has a dimension of about 1 cm, it can be accepted that the effective cavity-enhanced optical path length is approximately 1 km [20] and 700 km [21]. The existing experiments based on the cavity-enhanced scheme demonstrated that the sensitivity of the phase shift in a birefringence experiment limited by shot noise is about 3×10^{-13} rad [22]. These parameters will be used below to estimate the effect under study.

2. SIMULATION OF THE EXPERIMENT ON THE SEARCH FOR THE $\mathcal{P}$-, $\mathcal{T}$-ODD FARADAY EFFECT ON THE PbF MOLECULAR BEAM

The $\mathcal{P}$-, $\mathcal{T}$-odd Faraday effect is manifested as circular birefringence caused by the propagation of light in a medium subjected to an external electric field in the presence of $\mathcal{P}$-, $\mathcal{T}$-odd interactions. In this case, the linear Stark effect occurs. The plane of polarization of light for any type of birefringence is rotated by the angle

$$\psi(\omega) = \pi \frac{l}{\lambda} \mathrm{Re}\left[n^+(\omega) - n^-(\omega) \right], \qquad (1)$$

where l is the optical path length, ω is the transition frequency, λ is the corresponding wavelength, and $n^+(\omega)$ and $n^-(\omega)$ are the refractive indices for left and right circularly polarized photons, respectively. In the case of the $\mathcal{P}$-, $\mathcal{T}$-odd Faraday effect on molecules [24],

$$\mathrm{Re}\left[n^+(\omega) - n^-(\omega) \right] = \frac{d}{d\omega} n(\omega) \left(\omega^+ - \omega^- \right), \qquad (2)$$

where ω^+ and ω^- are the frequencies of left and right circularly polarized photons, respectively. We represent the signal $R(\omega)$ in birefringence experiments in the form

$$R(\omega) = \psi(\omega) T(\omega). \qquad (3)$$

Here, $T(\omega)$ is the transmission function related to the absorption of light inside a cavity and satisfying the Bouguer−Lambert−Beer law

$$T(\omega) = e^{-l/L(\omega)}, \qquad (4)$$

where $L(\omega)$ is the absorption length. Here, we do not include transmission from the mirrors of the cavity, which are used to increase the optical length. The shot noise limit for any polarimeter is proportional to the square root of the number of detected photons. Thus,

the signal-to-noise ratio $(R(\omega)/\sqrt{T(\omega)})$ is optimal when $\frac{d}{dl}\left(l e^{-l/2L(\omega)} \right) = 0$, i.e., when $l = 2L(\omega)$.

In the case of a magnetic dipole ($M1$) transition, the rotation angle and absorption length for the $\mathcal{P}$-, $\mathcal{T}$-odd Faraday effect can be represented in the form

$$\psi(\omega) = \frac{2\pi^2}{3} \frac{l}{\lambda} \rho \mu_0^2 |\langle i|\mathbf{l} - g_S \mathbf{s}|f\rangle|^2 \frac{h(u,v)}{\hbar \Gamma_D} \frac{\left(\omega^+ - \omega^-\right)}{\Gamma_D}, \quad (5)$$

$$\omega = \omega_0 + \Delta\omega, \qquad (6)$$

$$L^{-1}(\omega) = 4\pi\rho \frac{\omega_0}{\Gamma_D} f(u,v) \frac{\mu_0^2 |\langle i|\mathbf{l} - g_S \mathbf{s}|f\rangle|^2}{3\hbar c}. \qquad (7)$$

Here, ρ is the number density of molecules; $|i\rangle$ and $|f\rangle$ are the initial and final states in the resonance transition, respectively; $\mathbf{s}$ and $\mathbf{l}$ are the spin and orbital angular momentum operators of the electron, respectively; $g_S = -2.0023$ is the g-factor of the free electron; μ_0 is the Bohr magneton; Γ_D is the Doppler width; ω_0 is the resonant transition frequency; $\Delta\omega$ is the frequency detuning; $\hbar$ is the reduced Planck constant; and c is the speed of light. The functions $g(u,v)$, $f(u,v)$, and $h(u,v)$, as well as the variables u and v, are related to the Voigt spectral line shape as [1]

$$g(u,v) = \mathrm{Im}\mathscr{F}(u,v), \qquad (8)$$

$$f(u,v) = \mathrm{Re}\mathscr{F}(u,v), \qquad (9)$$

$$\mathscr{F}(u,v) = \sqrt{\pi} e^{-(u+iv)^2} \left[1 - \mathrm{Erf}(-i(u+iv)) \right], \qquad (10)$$

where $\mathrm{Erf}(z)$ is the error function,

$$u = \frac{\Delta\omega}{\Gamma_D}, \qquad (11)$$

$$v = \frac{\Gamma_n}{2\Gamma_D}, \qquad (12)$$

where Γ_n is the natural width,

$$h(u,v) = \frac{d}{du} g(u,v). \qquad (13)$$

The function $f(u,v)$ determines the behavior of absorption and has a maximum at the resonance point ω_0, where the function $h(u,v)$ also has a maximum. The function $h(u,v)$ has also a second maximum [24], which allows the observation of the $\mathcal{P}$-, $\mathcal{T}$-odd Faraday effect far from resonance in the region where absorption is very small.

An expression for the optimal column density ρl follows from the optimality condition $l = 2L(\omega)$. In terms of the natural width of the excited level Γ_n instead of the matrix element of the $M1$ transition, this expression has the form

$$\rho l = \frac{2\omega_0^2}{\pi c^2} \frac{\Gamma_D}{f(u,v)\Gamma_n}. \qquad (14)$$

The substitution of Eq. (14) into Eq. (5) yields

$$\psi_{\max}(u) = \frac{h(u,v)}{2f(u,v)} \frac{(\omega^+ - \omega^-)}{\Gamma_D}. \tag{15}$$

At $u \gg 1$, $h(u,v) \approx 1/u^2$ and $f(u,v) \approx v/u^2$. These asymptotic expressions describe well the exact behavior of the functions $h(u,v)$ and $f(u,v)$ in Eqs. (14) and (15) at $u > 4$, i.e., above the point of the second maximum of $h(u,v)$ and

$$\psi_{\max}(u) = \frac{(\omega^+ - \omega^-)}{\Gamma_n}. \tag{16}$$

Below, we accept $u = 5$ and use Eq. (16) to estimate the rotation angle.

We consider the $X1\,{}^2\Pi_{1/2} \rightarrow X2\,{}^2\Pi_{3/2}$ transition ($\lambda = 1210$ nm) in the PbF molecule. The natural width of the $X2$ level is $\Gamma_n = 2.7 \times 10^3$ s^{-1} [27]. The typical transverse temperature for such molecular beams is 1 K (e.g., for the YbF molecular beam in [28]). Then, $\Gamma_D = 4.5 \times 10^7$ s^{-1}. The difference $\omega^+ - \omega^-$ for the eEDM is expressed in terms of effective electric fields of the molecule $\mathscr{E}_{\text{eff}}$, which should be calculated theoretically (see below) for states between which the transition under consideration occurs. In the case of the $X1 \rightarrow X2$ transition in PbF, $\omega^+ - \omega^- = 2d_e\left(\mathscr{E}_{\text{eff}}({}^2\Pi_{+1/2}) - \mathscr{E}_{\text{eff}}({}^2\Pi_{+3/2})\right)$.

The effective electric field is related to the parameter W_d used to interpret the experimental search for the eEDM as $E_{\text{eff}} = W_d|\Omega|$. In turn,

$$W_d = \frac{1}{\Omega}\left\langle \Psi \left| \frac{H_d}{d_e} \right| \Psi \right\rangle, \tag{17}$$

where Ψ is the wavefunction of the considered molecular state and Ω is the projection of the total angular momentum of the electron on the molecular axis. The interaction Hamiltonian has the form [29]

$$H_d = d_e \sum_j \frac{2i}{e\hbar} c\gamma_j^0 \gamma_j^5 \mathbf{p}_j^2, \tag{18}$$

where $\mathbf{p}_j$ is the momentum operator of the jth electron and γ^0 and γ_j^5 are the Dirac matrices.

The $\mathscr{P}$-, $\mathscr{T}$-odd effect can be caused not only by the eEDM but elso by the scalar–pseudoscalar nucleus–electron interaction having the dimensionless coupling constant C_S. The Hamiltonian of this interaction can be represented in the form [2]

$$H_{\text{SP}} = i\frac{G_F}{\sqrt{2}} A C_S \sum_j \gamma_j^0 \gamma_j^5 \rho_N(\mathbf{r}_j), \tag{19}$$

where A is the mass number of the nucleus, G_F is the Fermi constant, and $\rho_N(\mathbf{r})$ is the nuclear density function normalized to unity. The considered interaction is characterized by the molecular parameter

$$W_S = \frac{1}{\Omega}\left\langle \Psi \left| \frac{H_{\text{SP}}}{C_S} \right| \Psi \right\rangle. \tag{20}$$

It is noteworthy that the constant $W_{\text{SP}} = W_S\, Z/A$, where Z is the number of protons in the nucleus, is often used in the literature [30].

In this work, the effective electric fields and constants W_S for the $X1\,{}^2\Pi_{1/2}$ and $X2\,{}^2\Pi_{3/2}$ electronic states of the PbF molecule were calculated by the methods of relativistic coupled clusters with single, double, and noniterative triple cluster amplitudes using the Dirac–Coulomb Hamiltonian [31] within the finite field approach. All electrons were included in the correlation. The AAETZ all-electron basis set [32] was used for the Pb atom [32]. The AETZ all-electron basis set [33–35] was used for the F atom. The error of the calculation of the effective electric fields can be estimated at 5%. In these calculations, we used the DIRAC15 code [36] and codes developed in [37]. The effective electric fields $\mathscr{E}_{\text{eff}}$ for the ground electronic state of PbF are in good agreement with the preceding calculations [38, 39]. Our calculation gives the effective electric fields $\mathscr{E}_{\text{eff}}({}^2\Pi_{+1/2}) = 38.0$ GV/cm and $\mathscr{E}_{\text{eff}}({}^2\Pi_{+3/2}) = 9.3$ GV/cm and the constants $W_S({}^2\Pi_{+1/2}) = 222.3$ kHz and $W_S({}^2\Pi_{+3/2}) = 19.4$ kHz. According to the simulation, the value $\rho l = 2 \times 10^{19}$ cm^{-2} should be used for the optimal signal-to-noise ratio. In this case, the angle corresponding to the current bound on the eEDM [4] is $\psi_{\max} = 4 \times 10^{-7}$ rad. The authors of [40, 41] discussed the achievement of the molecule number densities for molecular beams up to $\rho \sim 10^{12}$–10^{13} cm^{-3}. In this case, the optical length for achieving the maximum effect should be $l \sim 20$ km. Such optical lengths can be obtained for the beam experiment in the cavity [21].

The results of the discussed experiment can also be interpreted in terms of the constant C_S under the assumption that $d_e = 0$, as made, e.g., in [4]. Similar to the case of the eEDM, using the calculated constants W_S and a bound on the constant C_S established in [4], we obtain $\psi_{\max} = 3 \times 10^{-7}$ rad.

3. CONCLUSIONS

The best bound on the eEDM $d_e < 1.1 \times 10^{-29}$ e cm was obtained in the experiment with the ThO molecule [4], where the electron spin precession effect in the external electric field was studied. This effect is proportional to the time for which an individual molecule is subjected to the electric field. In this work, we have proposed another method for observation of such effects based on the ICAS experiment on the PbF

molecular beam aimed at searching for the $\mathscr{P}$-, $\mathscr{T}$-odd Faraday effect. The proposed experiment has been simulated with the parameters corresponding to the current achievements in ICAS. In this work, we have discussed only the principle of the experiment and do not consider its details and possible technical problems. In the proposed experiment, it is no longer necessary to increase the time of residence of an individual molecule in the electric field because the effect is carried by a laser beam interacting with numerous molecules. According to our estimates and taking into account the record sensitivity of 3×10^{-13} rad [22], the ICAS experiment with the PbF beam can in principle improve the bound on the eEDM by six orders of magnitude. This corresponds to probing the scale of masses of new particles three orders of magnitude higher than that in [4], i.e., tens of PeVs [4, 13].

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