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Possible experiment on the observation of the $\mathcal{P}$, $\mathcal{T}$-odd Faraday effect in intracavity absorption spectroscopy: Statistics and systematic errors

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A recently proposed experiment on the observation of the $\mathcal{P}$, $\mathcal{T}$-odd ($\mathcal{P}$ is space parity and $\mathcal{T}$ is time reflection) Faraday effect (optical rotation in an external electric field) (PTFE) in intracavity absorption spectroscopy is discussed. Two versions of this experiment are considered: the propagation of a continuous laser beam through an optical cavity filled with molecular gas (ThO vapor) and propagation of a continuous laser beam through an optical cavity crossed by the molecular beam (ThO molecules). With the use of optical cavities of high fineness, the accuracy of the experiment may reach a level exceeding the recently attained one by several orders of magnitude. The statistics of both types of experiment are analyzed. Ways to avoid experimentally the dangerous systematic errors, which can mimic the PTFE, are discussed.

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I. INTRODUCTION

A search for $\mathcal{T}$-noninvariant interactions in nature is one of the most important and most long-standing problems in fundamental physics. Since 1950 it has become clear that it may be easier to observe the $\mathcal{P}$, $\mathcal{T}$-odd effects than pure $\mathcal{T}$-odd ones [1]. In Ref. [1] it was suggested to measure the electric dipole moment (EDM) of a neutron by the magnetic resonance method. The existence of the EDM for any particle that is not strictly neutral, i.e., does not coincide with itself under charge ($\mathcal{C}$) conjugation, violates both $\mathcal{T}$ and $\mathcal{P}$ symmetry. Later it was predicted that the electron EDM (eEDM) is strongly enhanced in heavy atoms and especially in heavy diatomic molecules [2–6]. In Refs. [4,6] it was also shown that apart from the $\mathcal{P}$, $\mathcal{T}$-odd interaction of the eEDM with an external electric field, another $\mathcal{P}$, $\mathcal{T}$-odd effect, namely, the $\mathcal{P}$, $\mathcal{T}$-odd interaction between an electron and the nucleus in an atom (molecule), should exist. The constant for this interaction should be extracted from the experimental data together with the value for the eEDM. Various theoretical models for this interaction will be mentioned later in this section. In an external electric field both interactions lead to the same observable effect: a linear Stark shift (splitting) of atomic energy levels. Both effects can be distinguished only in a series of experiments with different species [7,8]. The results of experiments on the search for the $\mathcal{P}$, $\mathcal{T}$-odd effects can be presented either as an upper bound for the eEDM value d_e or as an upper bound for the constant of the $\mathcal{P}$, $\mathcal{T}$-odd electron-nucleus interaction. It is convenient to present this constant as an equivalent eEDM d_e^{eqv} which produces the same (as d_e) Stark shift of atomic (molecular) levels in the same external electric field. The peculiarities of the choice of strength of external field for diatomic molecules will be discussed in Sec. II.

The most stringent upper bound for the eEDM was most recently obtained in an experiment by the ACME Collaboration [9]: $d_e < 1.1 \times 10^{-29}$ e cm (e is the electron charge). In that experiment the electron spin precession in an external electric field had to be observed. The experiment was performed according to the scheme elaborated earlier in Ref. [10] of the xenon atom on the molecular beam of ThO molecules. Extraction of the d_e value from the experimental data requires theoretical support in the evaluation of the enhancement coefficient. This evaluation is a very complicated procedure which can be performed only with the application of the most powerful modern quantum chemistry methods. For the ThO molecule these calculations were done in Refs. [8,11–13]. Very recently, Ref. [14] reported the results of a new experiment on the search for the eEDM. In that experiment HfF^+ molecular ions trapped in a rotating electric field were employed and the linear Stark shift of molecular levels was observed. This gave a new bound for the eEDM: $d_e < 4.1 \times 10^{-30}$ e cm. The enhancement coefficients were evaluated in Refs. [15,16].

The theoretical predictions for the magnitudes of d_e and d_e^{eqv} within the standard model (SM) are the following. The recent calculation of d_e gives the result $d_e = 10^{-39}$ e cm [17]. As it was shown in Refs. [4,6,18], the $\mathcal{P}$, $\mathcal{T}$-odd electron-nucleus interaction in atoms (molecules) can be only of scalar-pseudoscalar type. In Ref. [19] this interaction was estimated on the basis of a two-photon exchange model with the result $d_e^{\mathrm{eqv}} = 10^{-38}$ e cm. The electron-nucleus interaction on the basis of another model (exchange by a Higgs boson) was estimated in Ref. [20] with the result between 10^{-40} and 10^{-45} e cm. The largest predicted value for the electron-nucleus interaction d_e^{eqv} was reported recently on the basis of the exchange by a neutral K meson: $d_e^{\mathrm{eqv}} = 10^{-35}$ e cm [21]. There exists a vast amount of literature on the various extensions of the SM which predict larger values for d_e and d_e^{eqv} (see, for example, Ref. [22]). None of these predictions have been verified yet and some of them have already been ruled out by existing experiments.

Due to the large gap between the lower experimental results for d_e and d_e^{eqv} and the upper theoretical predictions within the SM, other experimental approaches to the observation of the $\mathcal{P}, \mathcal{T}$-odd effects remain important. One such approach consists of the observation of the $\mathcal{P}, \mathcal{T}$-odd Faraday effect (optical rotation in an external electric field). The $\mathcal{P}, \mathcal{T}$-odd Faraday effect (PTFE) was first discussed between many other magneto- and electro-optical effects in Ref. [23], then mentioned in Ref. [5], and studied theoretically and experimentally in Ref. [24] (see Ref. [25] for further discussion of PTFE experiments in optical cells). In a series of papers [26–32] it was suggested that the PTFE in optical cavities could be observed by taking advantage of the rapid progress in recent decades of intracavity absorption spectroscopy (ICAS). In Ref. [33] an experiment was described where laser light traveled 70 000 km through an absorber within a cavity. In Ref. [34] an accuracy of 10^{-12} rad for the birefringence phase shift measurement was reported. In Ref. [35] a possible experiment on the measurement of the $\mathcal{P}$-odd optical rotation in an optical cavity was suggested and a theoretical simulation of this experiment was made. The technique that was proposed to be used in this experiment is closest to what is necessary for the observation of the PTFE.

In the present paper we reconsider the statistics and study the possible systematic errors in the PTFE ICAS experiment. From a general point of view, there are at least three main sources of systematic errors which can mimic $\mathcal{P}, \mathcal{T}$-odd effects: uncontrolled ellipticity of the laser light, uncontrolled stray magnetic fields (including a motional magnetic field for the particles moving in the electric field), and uncontrolled natural or $\mathcal{P}$-odd optical rotation. We will consider the limitations set by these systematic errors as well as methods to avoid them.

We consider this paper to be the last one in a series of papers [26–32]. We hope that the theoretical studies performed in this series would prompt new experimental studies of the $\mathcal{P}, \mathcal{T}$-odd effects in nature.

II. $\mathcal{P}, \mathcal{T}$-ODD FARADAY EXPERIMENT IN INTRACAVITY ABSORPTION SPECTROSCOPY

The $\mathcal{P}, \mathcal{T}$-odd Faraday effect is the optical rotation of the light (rotation of the polarization plane for the linearly polarized light) propagating through a medium in the presence of an external electric field oriented in the direction of the light propagation. In the case of ICAS techniques, the light of a continuously working laser propagates through a medium located within an optical cavity. We will consider two types of PTFE ICAS experiments. In the first type of experiment the medium (absorber) is represented by the molecular beam crossing the cavity in the direction orthogonal to the direction of the laser beam. The crossing point of the two beams is located in an external electric field. In the experiment of the second type the laser beam travels through an optical cavity filled with molecular gas vapor. In this case the electric field should be applied the entire length of the cavity. We assume the length of the cavity is about 1 m, the diameter of the laser beam is about 1 mm, and the diameter of the molecular beam is about 1 cm. The ThO molecule seems to be the best candidate among all molecules considered for the search of $\mathcal{P}, \mathcal{T}$-odd effects by the PTFE ICAS method. The enhancement coefficient for the eEDM in this case is about $\mathcal{K} \sim 10^9$. The next candidate for the search of $\mathcal{P}, \mathcal{T}$-odd effects by this method is the PbF molecule, first mentioned in this sense in Ref. [6]. For PbF the enhancement coefficient is about $\mathcal{K} \sim 10^6$ [36–38]. Nevertheless, in spite of difference in $\mathcal{K}$ coefficients, both molecules are equally suitable for performing experiments. The enormous values for $\mathcal{K}$ coefficients in diatomic molecules are connected to the Ω doubling of rotational molecular levels. The importance of Λ doubling (Λ is the projection of the total electron orbital angular momentum on the molecular axis; the quantum number Ω includes also the projection of total electron spin and is relevant for heavy molecules) in the search of $\mathcal{P}$-odd effects on the basis of the SM, including neutral currents, was first considered in Ref. [39] and for $\mathcal{P}, \mathcal{T}$-odd effects in Refs. [5,6]. The Ω doublet consists of two components (sublevels) of opposite parity and the larger the Ω value, the smaller the Ω splitting and consequently the larger the $\mathcal{P}, \mathcal{T}$-odd effects due to the mixing of Ω components by the $\mathcal{P}, \mathcal{T}$-odd interactions. This explains the difference in $\mathcal{K}$ coefficients in ThO and PbF: In ThO the metastable level most suitable for performing experiments has $\Omega = 1$ and the ground level in PbF most suitable for the same purpose corresponds to $\Omega = \frac{1}{2}$. However, the sensitivity of the experiment, i.e., the maximum optical rotation angle, is proportional to the effective electric field $\mathcal{E}_{\text{eff}}$ which arises in the molecule after applying an external electric field $\mathcal{E}_{\text{ext}}$: $\mathcal{E}_{\text{eff}} = \mathcal{K}\mathcal{E}_{\text{ext}}$. For molecules, unlike atoms, due to the extreme closeness of Ω doublets, the saturation effect does exist: The $\mathcal{E}_{\text{ext}}$ may become so strong that the levels of opposite parity (Ω components) begin to repulse, i.e., the $\mathcal{P}, \mathcal{T}$-odd effect will stop growing. Then there exists a saturation field $\mathcal{E}_{\text{sat}}$ which $\mathcal{E}_{\text{ext}}$ should not exceed. The condition $\mathcal{E}_{\text{eff}} = \mathcal{K}\mathcal{E}_{\text{sat}}$ should be satisfied [40]. For ThO $\mathcal{E}_{\text{sat}} \approx 10$ V/cm and for PbF $\mathcal{E}_{\text{sat}} \approx 10^4$ V/cm, so $\mathcal{E}_{\text{eff}}$ is approximately of the same order of magnitude. The sensitivity of both molecules in the experiments for the search of $\mathcal{P}, \mathcal{T}$-odd effects is also approximately the same. From these considerations it follows also that it is of no use to search for the molecules with higher Ω values in the ground (metastable) state [40]. For such molecules the coefficients $\mathcal{K}$ would be higher, but the field $\mathcal{E}_{\text{sat}}$ would be lower, so $\mathcal{E}_{\text{eff}}$ would be the same [40]. Experiments with very low $\mathcal{E}_{\text{ext}} = \mathcal{E}_{\text{sat}}$ would be more difficult. Then in the experiment of the first type the maximum external electric field can be 10^4 V/cm, which is enough to satisfy the condition $\mathcal{E}_{\text{ext}} = \mathcal{E}_{\text{sat}}$ for the PbF molecule; for ThO only 10 V/cm will be enough. In the experiment of the second type this condition can be satisfied only for ThO by applying a voltage of 10^4 V across the cavity length. Therefore, in what follows we will concentrate on the ThO molecule as the best candidate for both types of experiment. In both experiments the photons which have interacted with molecules and have acquired an optical rotation angle should be detected outside the cavity. In both types of experiment the statistics are determined by the number of detected photons; the number of photons is equal to the number of molecules (see Sec. III). In what follows we

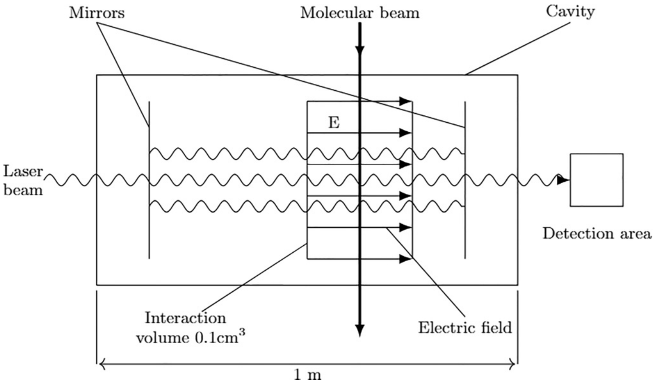

FIG. 1. Schematic of the possible experimental setup for the PTFE-ICAS experiment of type I.

will denote the two types of experiment by the symbols I and II. Schematics of the possible experimental setups for types I and II of the PTFE ICAS experiments are given in Figs. 1 and 2, respectively.

The optical rotation angle is always connected to some kind of birefringence and can be presented as

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

where $n^{+}(\omega)$ and $n^{-}(\omega)$ are the refractive indices for the right and left polarized light, respectively, l is the optical path length, ω is the light frequency, and λ is the light wavelength. In the case of the PTFE [27],

$$\mathrm{Re}[n^{+}(\omega) - n^{-}(\omega)] = \frac{d}{d(\hbar\omega)}n(\omega)Q, \tag{2}$$

where $n(\omega)$ is the refractive index for the linearly polarized light, $\hbar$ is the Planck constant, and Q is the linear Stark shift of atomic (molecular) levels. This shift can be presented as

$$Q = R_d d_e \mathcal{E}_{\mathrm{ext}}, \tag{3}$$

where d_e is the absolute value of the eEDM, R_d is the enhancement coefficient for the eEDM in an atom (molecule), and $\mathcal{E}_{\mathrm{ext}}$ is the external electric-field strength. Instead of R_d, also an effective field $\mathcal{E}_{\mathrm{eff}} = R_d\mathcal{E}_{\mathrm{ext}}$ can be introduced. As mentioned in the Introduction, for molecules it is natural to set $\mathcal{E}_{\mathrm{ext}} = \mathcal{E}_{\mathrm{sat}}$, where $\mathcal{E}_{\mathrm{sat}}$ is the saturation field.

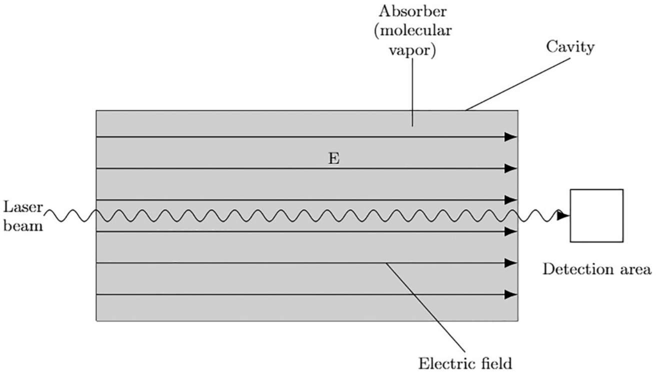

FIG. 2. Schematic of the possible experimental setup for the PTFE-ICAS experiment of type II.

In the case of a resonant PTFE with a resonant $M1$ transition between the initial (i) and final (f) molecular states (which corresponds to the choice of the ThO molecule as a candidate for the experiment) and employing the Voigt parametrization of the optical line profile, we can use the following expression for the optical rotation angle [29]:

$$\psi(\omega) = \frac{\pi}{3}\frac{l}{\lambda}\rho e^{2}|\langle i|\boldsymbol{l} + 2\boldsymbol{s}|f\rangle|^{2}\frac{h(u,v)}{\hbar\Gamma_{\mathrm{D}}}\frac{2d_e\left(\mathcal{E}^{i}_{\mathrm{eff}} + \mathcal{E}^{f}_{\mathrm{eff}}\right)}{\Gamma_{\mathrm{D}}}. \tag{4}$$

In this expression ρ denotes the molecular number density and Γ_{D} is the Doppler width. The function $h(u,v)$ is the Voigt parametrization function [18,28] of two dimensionless variables $u = \frac{\Delta\omega}{\Gamma_{\mathrm{D}}}$ and $v = \frac{\Gamma}{\Gamma_{\mathrm{D}}}$, where $\Delta\omega$ is the detuning $\Delta\omega = \omega - \omega_0$, with $\omega_0 = E_f - E_i$ (E_i and E_f are the energies of the molecular states i and f, respectively). The width Γ is different for the different types of experiment discussed above. In the first type of experiment (molecular beam plus optical cavity) $\Gamma = \Gamma_{\mathrm{nat}}$, where Γ_{nat} is the natural width of the excited molecular state f in the transition $i \to f$ chosen as the basic (resonant) one in the PTFE experiment. The state i is assumed to be the ground or metastable state. For the PTFE experiment the most suitable transition in the ThO molecule is $i \to f = X\,^{1}\Sigma_0 \to H\,^{3}\Delta_1$, where $H\,^{3}\Delta_1$ is the metastable level with $\Gamma_{\mathrm{nat}} = 5\times10^{2}\ \mathrm{s}^{-1}$. The collisional width in the beam experiment is considered negligible. Contrary to this, in the second type of experiment $\Gamma = \Gamma_{\mathrm{col}}$, where Γ_{col} is the collisional width. We assume, however, that for a low enough density of the molecular gas within the cavity, this gas can be cooled to the same level as in molecular beam so that $\Gamma_{\mathrm{col}} = \Gamma_{\mathrm{nat}}$. This suggestion can be also confirmed by the direct estimate [18]

$$\Gamma_{\mathrm{col}} = 4\sqrt{\frac{2}{\pi}}\rho\sigma\sqrt{\frac{2kT}{M}}, \tag{5}$$

where ρ is molecular number density, k is the Boltzmann constant, T is the absolute temperature, M is the mass of the molecule, and σ is the cross section of the scattering molecule on the molecule. For estimates we assume that σ is approximately the same as for heavy atom-atom collisions [18].

We assume that the Doppler width is of the level $\Gamma_{\mathrm{D}} = 10^{8}\ \mathrm{s}^{-1}$ [18]. An important feature of the proposed PTFE experiment is the use of a large detuning. This is necessary to suppress the absorption compared to optical rotation. In Ref. [27] it was found that the optimal detuning is $\Delta\omega \approx 5\Gamma_{\mathrm{D}}$.

The absorption of the light in the medium as a function of the optical path length is determined by the equation

$$dI(l) = -I(l)\rho_M\sigma\,dl, \tag{6}$$

where $I(l)$ is the light intensity, ρ_M is the medium (molecular) number density, and σ is the absorption cross section. When ρ_M does not depend on l, from Eq. (6) follows the Beer-Lambert law

$$I(l) = I_0 e^{-\rho_M\sigma l} = I_0 e^{-l/L}, \tag{7}$$

where I_0 is the initial intensity and

$$L = (\rho\sigma)^{-1} \tag{8}$$

is the absorption length. The optimal choice of the l value for the PTFE ICAS experiment will be discussed in the next section.

III. STATISTICS OF THE PTFE ICAS EXPERIMENT

The PTFE experiment in any case is a statistical experiment. Now we have to compare the statistics in both types of PTFE experiments considered above.

The signal in the PTFE experiment can be defined as

$$S(\omega) = \psi(\omega)N_{\text{ev}}, \tag{9}$$

where $\psi(\omega)$ is determined by Eq. (4) and N_{ev} is the number of statistical events in a statistical experiment. In the case of the PTFE, the value of N_{ev} was considered to be $N_{\text{ev}} = N_{\text{ph}}$ [26–32], where N_{ph} is the number of photons which have interacted with molecules and were then detected. The statistical (shot) noise in this case is

$$N = 2\pi\sqrt{N_{\text{ev}}} \tag{10}$$

and the signal-to-noise ratio is

$$\frac{S}{N} = \frac{\psi}{2\pi}\sqrt{N_{\text{ev}}}. \tag{11}$$

To pass the shot-noise limit it is necessary to have $\frac{S}{N} = 1$ and the sensitivity of the experiment can be defined as

$$\psi_s = \frac{2\pi}{\sqrt{N_{\text{ev}}}}, \tag{12}$$

where N_{ev} is determined from the condition $\psi_s \leqslant \psi_{\text{expt}}$, where ψ_{expt} is a small rotation angle expected due to the effect being sought. This means that when an observer detects an angle $\psi > \psi_s$, this angle originates from the effect, not from the fluctuation. So it is necessary to have a sufficiently large number of photons in the experiment. The number of photons for a continuously working laser in the PTFE experiment is

$$N_{\text{ph}} = j_{\text{ph}}\sigma_{\text{ph}}t_{\text{meas}} = \frac{I_{\text{tr}}(\omega)}{\hbar\omega}\sigma_{\text{ph}}t_{\text{meas}}, \tag{13}$$

where j_{ph} is the photon flux, σ_{ph} is the cross section of the laser beam, t_{meas} is the measurement time, $I_{\text{tr}}(\omega)$ is the intensity of the light transmitted through the cavity, and $\hbar\omega$ is the energy of the photons. To increase the number of detected photons we can optimize the signal-to-noise ratio inside the cavity with respect to l, finding the optical path length value which corresponds to the maximum $\frac{S}{N}$. For this purpose we set

$$N_{\text{ev}} = \frac{I(l)}{\hbar\omega}t_{\text{meas}}, \tag{14}$$

where $I(l)$ is determined by Eq. (7) and $\hbar\omega$ is the energy of the photon.

However, we should not violate the saturation threshold: When the light intensity becomes too high the medium becomes transparent with respect to the light, the absorption vanishes, and the optical rotation vanishes too. Of course, in the very dense absorber the light of any intensity will be absorbed. The level of the light intensity when the medium becomes transparent determines the saturation threshold. This saturation with respect to the light intensity is different from the saturation with respect to the external electric field, mentioned in the Introduction. In the case of resonance the saturation occurs when the spontaneous decay rate becomes equal to the stimulated decay rate and hence to the absorption rate. The number of absorbed photons becomes equal to the number of emitted photons, so the light stops being absorbed and travels through the medium without a loss of intensity. The optical rotation also vanishes since the populations of the ground and excited states of an atom become equal and the directions of the optical rotation by the atoms in both these states are opposite. From this condition it follows that [29]

$$I_s = \frac{\hbar\omega^3\Gamma_{\text{D}}}{\pi^2c^2f(u, v)}, \tag{15}$$

where I_s is the saturation intensity and $f(u, v)$ is the Voigt parametrization function for absorption. In the case of a large detuning the absorption cross section can be presented as $\sigma = \sigma_0 f(u, v)$, where σ_0 is the absorption cross section at the point of resonance and the function $f(u, v)$ describes the behavior of absorption with the detuning in terms of the Voigt parameters. For detuning $\Delta\omega = 5\Gamma_D$ we can use the asymptotic behavior of $f(u, v)$: $f(u, v) \to \frac{v}{u^2}$ [18]. Then the numerical calculation for the PTFE ICAS experiment with $v = \Gamma_{\text{nat}}$ and $u = \frac{1}{5}$ for the chosen transition in the ThO molecule gives the result of 10^{-19} cm^{-2} for the σ value [31]. With the typical number density for molecular beams $\rho_M \approx 10^{10}$ cm^{-3}, this leads to the very large absorption length $L = 10^9$ cm $= 10^4$ km according to Eq. (8). In the case of the PTFE ICAS experiment I this length corresponds to $N_p = 10^9$ passes of the light through the molecular beam with a diameter of 1 cm, or to $N_p = 10^9$ passes of the light through a cavity of $l_0 = 1$ m length. Such an optical path length was reported only once [33]. It is more realistic to use the cavities with $N_p \leqslant 10^7$. We will use this number in our further estimates. Recalling the behavior of $f(u, v)$ for large detuning $\Delta\omega$, we can employ an estimate

$$I_s = \frac{\hbar\omega^3\Delta\omega^2}{\pi^2c^2v\Gamma_{\text{D}}}. \tag{16}$$

In the case of the type-I experiment, $v = \frac{\Gamma_{\text{nat}}}{\Gamma_{\text{D}}}$ and for the chosen transition in the ThO molecule we find $I_s(I) \approx 3 \times 10^3$ W/cm^2. This corresponds to a photon flux of 3×10^{20} photons per second.

To determine the sensitivity ψ_s for every type of experiment according to Eqs. (12) and (1), we need to know I_{tr} [see Eq. (13)]. In Ref. [29] it was suggested to use such a regime for the light transmission through an optical cavity, when $I_{\text{tr}} = I_0$ (I_0 is the initial intensity of the laser light). However, then the intensity of the light coupled inside the cavity is very high $I_{\text{int}} = \frac{I_{\text{tr}}}{T}$ [41]. Here T is the mirror transmission, which is roughly connected to the possible number of passes of the light along the cavity via $T = N_p^{-1}$. With our choice of N_p this means that the intensity of the light within the cavity will be 10^7 times stronger than the saturation threshold and the medium inside the cavity will be fully "bleached." The only outcome is a 10^7 times diminished input intensity following from the condition $I_0 = I_s$, with the value of I_s defined by Eq. (16). Returning to the optimization problem, the optimization condition follows from

$$\frac{d}{dl}\left[lI_0^{1/2}(l)e^{-l/2L}\right] \approx \frac{d}{dl}\left[l\left(\frac{l_0}{l}\right)^{1/2} e^{-l/2L}\right] = 0. \tag{17}$$

The nonlinear equation (17) has two roots: $l = \infty$ and $l = L$. The physical sense has only the second root $l = L$. The optimization $l = L$ is valid for the optical cavities and is different from the standard optical optimization condition $l = 2L$ [18]. This difference is due to the dependence of I_0 on l, which occurs in optical cavities. The growth of I_0 in optical cavities leads to the growth of I_{int} by an additional factor $N_p = l/l_0$, where l_0 is the size of the cavity. This explains the appearance of the factor $(l_0/l)^{1/2}$ in Eq. (17).

Hence, we have to reduce the input value of the light intensity $I_0 = I_s$ given above and the output value I_{tr} by seven orders of magnitude: $I_0(I) = I_{\text{tr}}(I) \approx 10^{-4}$ W/cm^2. According to Eq. (13) with $\sigma_{\text{ph}} = 10^{-2}$ cm^2 and $N_{\text{ph}} = 10^{13}$ photons/s $\times$ [t_{meas} (s)] [29], the sensitivity is [see Eq. (12)] $\psi_s \approx 10^{-8}[t_{\text{meas}}\ (\text{s})]^{-1/2}$ rad. An experimental constraint for d_e can be obtained from the equality

$$\psi = \psi_s, \tag{18}$$

where ψ is the measured angle of rotation for the ThO molecule. Equation (4) for the angle ψ can be presented as

$$\psi\ (\text{rad}) = \mathcal{K}\ (\text{rad}/e\,\text{cm}) \times d_e\ (e\,\text{cm}). \tag{19}$$

From the calculations in Ref. [29] it follows that

$$\mathcal{K}\ (\text{rad}/e\,\text{cm}) = \frac{10^{-8}\ \text{rad}}{10^{29}\ e\,\text{cm}}. \tag{20}$$

Then Eq. (18) looks like

$$10^{-8}\frac{d_e}{10^{29}\ e\,\text{cm}} = 10^{-8}[t_{\text{meas}}\ (\text{s})]^{-1/2} \tag{21}$$

and with $t_{\text{meas}} \approx 10^6$ s the constraint for d_e appears to be $d_e(I) = 10^{-31}$ $e\,$cm, two orders of magnitude better than the experimental bound obtained in the ThO experiment by the ACME Collaboration [9].

Consider now the situation in the PTFE ICAS experiment II. For a cavity of $l_0 = 10^2$ cm length, the optical path length l for the photons after the same number of passes as we suggested for the PTFE ICAS experiment II will be 100 times longer than in the PTFE ICAS experiment I. This promises a corresponding gain in the eEDM bound observation. However, the total number of cooled ThO molecules with the number density $\rho_{\text{mol}} = 10^{10}$ mol/cm^3 within the volume $V = 10^2 \times 10 \times 10$ cm$^3 = 10^4$ cm^3 can be only 10^{14} molecules, while the total number of photons during the experiment time 10^6 s, as it was estimated above for the PTFE ICAS experiment I, is about 10^{19} photons. Filling the cavity with a molecular beam [molecular number density $\rho_{\text{mol}} = 10^{10}$ molecules/cm^3, molecular velocity $v_{\text{mol}} = 10^4$ cm/s, molecular beam cross section $\sigma_{\text{mol}} = 1$ cm^2, and molecular flux $j_{\text{mol}} = 10^{14}$ molecules/(s cm^2)] will happen within 1 s. Then repeating the process for $t_{\text{meas}} = 10^6$ s, we estimate the total number of molecules participating in the experiment to be 10^{20}, i.e., approximately equal to the total number of photons. Consequently, we can consider the number of detected photons as the number of statistical events in the PTFE ICAS experiment II. This would allow us to reduce the bound for the eEDM to the value 10^{-33} $e\,$cm. From the estimates above it follows that the statistics in the PTFE ICAS experiment II are determined by the number of molecules, as in the experiment by the ACME Collaboration [9]. Both experiments are statistical and the measurement time t_{meas} also can be the same. A question arises of where the gain in the experimental bound from the eEDM in the PTFE ICAS experiment I may arise from. One possible answer is that in the experiment by the ACME Collaboration a serious loss of molecules of the order of 10^{-3} was mentioned: Only one molecule from every thousand in the molecular beam reached the detectors [42]. In the PTFE ICAS experiment II this loss is absent since the molecular beam is used only to fill the cavity. According to our estimates [29–32], no serious photon loss in the PTFE ICAS experiments is expected.

IV. POSSIBLE SYSTEMATIC ERRORS

In this section we will consider the main sources of systematic errors which can mimic the $\mathcal{P},\mathcal{T}$-odd effect in the $\mathcal{P},\mathcal{T}$-odd Faraday experiment. These sources are uncontrolled ellipticity of the laser light, stray magnetic fields including the motional magnetic field, and natural and $\mathcal{P}$-odd optical rotation. We will give estimates for the dangerous levels of all these effects and will discuss ways to eliminate the corresponding errors.

A. Uncontrolled ellipticity

The $\mathcal{P},\mathcal{T}$-odd Faraday experiment should be performed, in principle, with linearly polarized laser light. In an ideal experiment the polarization plane of the light should not rotate when the light in the absence of external fields travels through an absorber which is not naturally optically active and does not possess $\mathcal{P}$-odd optical activity. However, if the polarization of the light is not purely linear but elliptic, an optical rotation will occur which in the case of the $\mathcal{P},\mathcal{T}$-odd Faraday experiment will mimic the $\mathcal{P},\mathcal{T}$-odd effect. For estimates it is convenient to present the uncontrolled ellipticity of the light in the form

$$P = L + aC, \tag{22}$$

where P is the light polarization, L is the linear polarization, C is the circular polarization, and a is the ellipticity coefficient.

According to Table I, for the most sensitive experiment II with the ThO molecule the optical rotation angle after 10^7 passes of the light through an absorber of 1 m length becomes rotated by an angle of about 10^{-6} rad. Then the rotation angle per pass is 10^{-13} rad. From the other side the circularly polarized light polarization rotates by an angle of 2π rad after the light covers the distance of one wavelength λ. For $\lambda \approx 10^3$ nm $= 10^{-4}$ cm (as in the ThO case) the rotation angle during one pass of the light through the cavity of 1 m length will be about 10^6 rad. Then from the equality 10^{-13} rad $= a \times 10^6$ rad the uncontrolled ellipticity coefficient should be less than $a \approx 10^{-19}$.

TABLE I. Comparison of experiments I (molecular beam) and II (molecular vapor). The length of the cavity is 1 m, the diameter of the laser beam is 1 mm, and the diameter of the molecular beam is 1 cm.

	Experiment	
Parameter	I	II
molecular number density ρ (cm^{-3})	10^{10}	10^{10}
optical path length l (cm)	10^{7}	10^{9}
ρl (cm^{-2})	10^{17}	10^{19}
laser intensity I_0 ($\mathrm{W/cm^2}$)	10^{-4}	10^{-4}
saturation intensity I_s ($\mathrm{W/cm^2}$)	10^{3}	10^{3}
photon flux at the detector j (photons/s)	10^{13}	10^{13}
mirror transmission$T$	10^{-7}	10^{-7}
number of passes of the light through the cavity (cell)	10^{7}	10^{7}
expected bound for eEDM (e cm)with $t_{\mathrm{meas}} = 10^6$ s	10^{-31}	10^{-33}

B. Stray magnetic fields

To estimate the dangerous level of stray magnetic fields, we have to compare the rotation angle for the ordinary Faraday effect in an external magnetic field $\mathcal{H}$ and the $\mathcal{P}, \mathcal{T}$-odd Faraday effect in an external electric field $\mathcal{E}$. Considering both effects for the same resonant transition in the same molecule, we have to compare the Zeeman splitting and the linear Stark splitting in the ThO molecule. In Ref. [43] it was found that the g factor for the H state in ThO is unusually small: $g \sim 10^{-3}$. The influence of the stray magnetic fields can be limited even more if we take into account the behavior of Ω doublets in the ThO molecule in external fields. This behavior shows that for one Ω component Zeeman and linear Stark shifts add up, but for other component they subtract. The total contribution of the stray magnetic field to the $\mathcal{P}, \mathcal{T}$-odd effect is then determined by the difference of the g factors for both components, which is of the order 10^{-6}. Moreover, in Ref. [43] it was also found that if one were to use for the experiment the next rotational level $J = 2$ of the H state, it would be possible to choose the external field $\boldsymbol{\mathcal{E}}$ in such a way ($\mathcal{E} \approx 50$ V/cm) that the g factor would become practical zero. The enhancement coefficient for the linear Stark shift in a heavy molecule is equal to $\frac{\mathcal{E}_{\mathrm{eff}}}{\mathcal{E}_{\mathrm{ext}}}$ and for the ThO molecule can be taken from Ref. [11]: $\frac{3.8\ \mathrm{GV/cm}}{10\ \mathrm{V/cm}} \approx 10^9$. Then using the Gauss system of electromagnetic units we can write

$$10^{-6}\mu_0\ (e\,\mathrm{cm}) \times \mathcal{H}\ (\mathrm{G}) = 10^9 d_e\ (e\,\mathrm{cm}) \times \mathcal{E}\ (\mathrm{V/cm}), \quad (23)$$

where $\mu_0 = 1.68 \times 10^{-11}$ $e\,$cm. Equation (12) can be rewritten in the form

$$\mathcal{H}\ (\mathrm{G}) = 10^{15}\frac{d_e\ (e\,\mathrm{cm})}{\mu_0\ (e\,\mathrm{cm})}\mathcal{E}\ (\mathrm{V/cm}). \quad (24)$$

From Eq. (21) it follows that for the $\mathcal{P}, \mathcal{T}$-odd Faraday experiment in the ThO molecule with a maximum accuracy $d_e \sim 10^{-33}$ $e\,$cm, a dangerous level of stray magnetic field would be $\mathcal{H}_C \sim 10^{-8}$ G. This is a rather strong limitation which may however be softened by the standard procedure first suggested in Ref. [1]. An external magnetic field should be applied and both ordinary and $\mathcal{P}, \mathcal{T}$-odd Faraday effects should be observed together. The total signal will be $S = S^{\mathcal{H}} + S^{\mathcal{E}}$, where $S^{\mathcal{H}}$ and $S^{\mathcal{E}}$ are the contributions of the ordinary and $\mathcal{P}, \mathcal{T}$-odd Faraday effects, respectively. Then the direction of the electric field should be reversed, $S' = S^{\mathcal{H}} - S^{\mathcal{E}}$, and the results of both measurements should be subtracted, $S - S' = 2S^{\mathcal{E}}$. The cancellation of $S^{\mathcal{H}}$ should absorb also the contributions of all the permanent stray magnetic fields. The contributions of oscillating stray magnetic fields should cancel in a sufficiently large series of measurements.

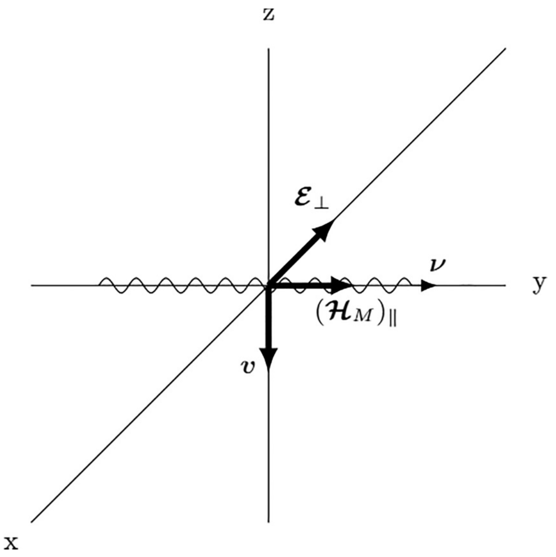

FIG. 3. False eEDM effect arising from the motional magnetic field in the case of the external electric field tilted with respect to the laser beam propagation direction $\boldsymbol{\nu}$. Here $\boldsymbol{\mathcal{E}}_\perp$ is electric-field component orthogonal to $\boldsymbol{v}$ in the yz plane, $\boldsymbol{v}$ denotes the molecular beam velocity, and $(\boldsymbol{\mathcal{H}}_M)_\parallel$ is the component of motional magnetic field parallel to $\boldsymbol{\nu}$.

A special problem is the influence of the motional magnetic field that acts on the molecule moving in an external electric field

$$\boldsymbol{\mathcal{H}}_M = \frac{1}{c}(\boldsymbol{v} \times \boldsymbol{\mathcal{E}}). \quad (25)$$

Here $\boldsymbol{v}$ is the velocity of the molecule and $\boldsymbol{\mathcal{E}}$ is an external electric field. The contribution of $\boldsymbol{\mathcal{H}}_M$ (false effect) cannot be eliminated like the contribution of the permanent stray magnetic field by reversing the direction of the electric field since $\boldsymbol{\mathcal{H}}_M$ also changes sign after this reversal. However, this can be done by reversing both the electric field $\boldsymbol{\mathcal{E}}$ and the direction of the molecular beam velocity $\boldsymbol{v}$ in the type-I experiment, since the EDM effect does not depend on the direction of $\boldsymbol{v}$. In experiment II the influence of the motional magnetic field is strongly diminished due to the chaotic motion of the molecules across the laser beam in both directions within the cavity. Moreover, $\boldsymbol{\mathcal{H}}_M$ is always strictly orthogonal to the applied electric field $\boldsymbol{\mathcal{E}}$ and therefore the contribution of $\boldsymbol{\mathcal{H}}_M$ to the $\mathcal{P}, \mathcal{T}$-odd Faraday effect is absent provided the electric field $\boldsymbol{\mathcal{E}}$ is strictly parallel to the laser beam direction. If there is a component $\boldsymbol{\mathcal{E}}_\perp$ orthogonal to the laser beam, the motional magnetic field will have a component $(\boldsymbol{\mathcal{H}}_M)_{||}$ parallel to the laser beam direction and thus able to mimic the $\mathcal{P}, \mathcal{T}$-odd Faraday effect (see Fig. 3). The order of magnitude of the motional magnetic field creating false contributions to the $\mathcal{P},\mathcal{T}$-odd Faraday effect is

$$\mathcal{H}_M\ (\mathrm{G}) = \frac{v}{c}\frac{(\boldsymbol{\mathcal{E}})_\perp}{(\boldsymbol{\mathcal{E}})_{||}}\boldsymbol{\mathcal{E}}\ (\mathrm{V/cm}). \tag{26}$$

Here $(\boldsymbol{\mathcal{E}})_{||}$ is the electric-field component parallel to the laser beam. Setting $\frac{v}{c}\sim 10^{-6}$ in experiment I, with $\frac{(\boldsymbol{\mathcal{E}})_\perp}{(\boldsymbol{\mathcal{E}})_{||}}\sim 10^{-3}$ and $\mathcal{E} = 10$ V/cm, we obtain $\mathcal{H}_M \sim 10^{-10}$ G, which is below the critical value $\mathcal{H}_c$ mentioned earlier in this section.

C. Natural and $\mathcal{P}$-odd optical activity

To estimate the necessary condition for the absence of the naturally optically active molecules, we can present the basic equation (4) for the optical rotation in the form

$$\psi_{\mathcal{P},\mathcal{T}}\ (\mathrm{rad}) = \rho_{\mathrm{mol}}\ (\mathrm{mol/cm}^3) \times l\ (\mathrm{cm}) \times \sigma_{\mathrm{scat}}\ (\mathrm{cm}^2) \times P_{\mathcal{P},\mathcal{T}}\ (\mathrm{rad}), \tag{27}$$

where $\psi_{\mathcal{P},\mathcal{T}}$ is the rotation angle due to the $\mathcal{P},\mathcal{T}$-odd interactions, ρ_{mol} is the number density for the molecules employed in the experiment, l is the optical path length, σ_{scat} is the forward photon scattering cross section, and $P_{\mathcal{P},\mathcal{T}}$ is the rotation coefficient. Here $P_{\mathcal{P},\mathcal{T}}$ can be called the rotation power of $\mathcal{P},\mathcal{T}$-odd effects per molecule per 1 cm in each particular experiment. If we put the value for the rotation angle $\psi_{\mathcal{P},\mathcal{T}} = 10^{-13}$ rad for one pass into Eq. (27), with $\rho_{\mathrm{mol}} = 10^{10}$ mol/cm^3, $l = 100$ cm, and using the typical value $\sigma_{\mathrm{scat}} \approx 10^{-12}$ cm^2 from Eq. (27), we will obtain $P_{\mathcal{P},\mathcal{T}} = 10^{-13}$.

We will assume that all optically active molecules (OAM) in the PTFE ICAS experiment I can arrive only with the molecular beam. Then we can use an equation similar to Eq. (27) to derive the limitation for the density of optically active molecules in the molecular beam:

$$\psi_{\mathrm{OAM}}\ (\mathrm{rad}) = \rho_{\mathrm{OAM}}\ (\mathrm{OAM/cm}^3) \times l\ (\mathrm{cm}) \times \sigma_{\mathrm{scat}}\ (\mathrm{cm}^2) \times P_{\mathrm{OAM}}. \tag{28}$$

Here ψ_{OAM} is the false rotation angle due to the admixture of the optically active molecules, ρ_{OAM} is the number density of such molecules, and we assume l and σ_{scat} to be approximately the same as in Eq. (27). We set the rotation power for the optical active molecules equal to 1: $P_{\mathrm{OAM}} = 1$.

The dangerous level of optically active molecule impurities arises when ψ_{OAM} becomes equal to $\psi_{\mathcal{P},\mathcal{T}}$ for one pass. Comparing the right-hand sides of Eqs. (27) and (28), we find that it will happen when the density ρ_{OAM} reaches the level $\rho_{\mathrm{OAM}} = 10^{-13}$ OAM/cm^3. This is a very restrictive level of impurity, but there is a natural way to avoid it (see below in Sec. IVD).

Another source of systematic errors is the $\mathcal{P}$-odd optical activity which should always be present for atoms or molecules exhibiting $\mathcal{P},\mathcal{T}$-odd effects. Theoretical and experimental studies of $\mathcal{P}$-odd optical activity for heavy atoms were discussed in detail in Ref. [18]. More recently, the same problems were addressed theoretically in connection with ICAS abilities in Ref. [35].

The $\mathcal{P}$- and $\mathcal{P},\mathcal{T}$-odd electron-nucleus interaction Hamiltonians have very similar structures [18–20]

$$H_{\mathcal{P}} = \frac{G_{\mathrm{F}}}{\sqrt{2}}\xi_{\mathcal{P}}(\bar{e}\gamma_\mu\gamma_5 e)(\overline{N}\gamma_\mu N), \tag{29}$$

$$H_{\mathcal{P},\mathcal{T}} = i\frac{G_{\mathrm{F}}}{\sqrt{2}}\xi_{\mathcal{P},\mathcal{T}}(\bar{e}\gamma_5 e)(\overline{N}N), \tag{30}$$

where G_{F} is the Fermi constant, the constant $\xi_{\mathcal{P}}$ for different important types of $\mathcal{P}$-odd interactions [18] is of the order of 1, e and N denote the electron and nucleus wave functions, respectively, and γ_μ and γ_5 are the Dirac matrices. From recent theoretical predictions for the eEDM in frame of the SM [19] it follows that

$$\xi_{\mathcal{P},\mathcal{T}} \approx 10^{-18}\xi_{\mathcal{P}}. \tag{31}$$

Then the requirement to restrict the contribution of $\mathcal{P}$-odd interactions to the PTFE experiment seems very hard. For the solution of this problem, see the following section.

D. Elimination of systematic errors

We begin with the problem of uncontrolled ellipticity, i.e., an imbalance between the right- and left-circularly polarized light. Every reflection of the circularly polarized light in the mirror changes the direction of the optical rotation: Clockwise changes to counterclockwise and vice versa. Therefore, the rotation angle gained in the onward travel of the circularly polarized light between two mirrors is exactly canceled with the angle gained during the backward travel. Then to eliminate the uncontrolled ellipticity contribution it is enough to make sure that the light inside the cavity has done an even number of passes (to and fro) along the same trajectory. For this purpose it is enough to locate the output mirror in the same front wall of the cavity where the input mirror is located.

Exactly the same idea may help to eliminate the contribution of natural optical activity and $\mathcal{P}$-odd interactions. The rotation angle in this case also changes sign after mirror reflection [18]. In principle, the cancellation of systematic errors described in Secs. IVA and IVC, after one pass of the light through the cavity (here we understand "pass" as travel of the light forward and backward), is exact. This happens because the optical rotation changes sign after reflection of the light wave from the wall. In the case of the (ordinary or $\mathcal{P},\mathcal{T}$-odd) Faraday effect, it does not happen since it simultaneously changes the sign of the external field with respect to the direction of the particle's velocity. Therefore, to observe the $\mathcal{P}$-odd optical rotation within the optical cavity it is necessary to insert the quarter waveplates into the front and back walls (mirrors) of the optical cavity [18]. Without these devices the $\mathcal{P}$-odd rotation would be zero. Of course, no such devices are necessary to observe the PTFE. Therefore, no $\mathcal{P}$-odd background exists in the PTFE; the situation is exactly the same as with the uncontrolled ellipticity. The errors in Secs. IVA and IVC may remain in PTFE experiments after cavity cancellations only to the technical defects, mirror impurities, etc. However, this remainder loses its systematic character and becomes a random statistical one. The role of statistical errors was already discussed in Sec. III. As a general method of control we can suggest the parallel measurement of the effect with the electric field reversed. The systematic errors in Secs. IVA and IVC do not change by this reversal, and the PTFE changes sign. Then, after subtraction of the results of two measurements, only the contribution of the PTFE with the factor 2 will survive. Elimination of systematic errors connected to stray magnetic fields was discussed in Sec. IVB. In Ref. [42] several other systematic errors which can mimic the eEDM effect in the experiment by the ACME Collaboration were considered. One of these errors is due to the inhomogeneity of the electric field. According to the Kramers theorem, the energy levels characterized by the half-integer total electron angular momentum of any atomic system remain degenerate with respect to the sign of projection of this momentum in any kind of electric field. For the integer-value levels like $H\,^3\Delta_1$ in ThO this statement holds only in homogeneous electric fields [44]. The inhomogeneity leads to the splitting of the levels by the sign of angular momentum projections, i.e., to the different frequencies of absorption for the photons with different chiralities. This would mimic the eEDM also in the PTFE. The numerical estimates given in Ref. [42] for this false effect are at the level $d_e \sim 10^{-32}$ e cm, i.e., at the level of the bound that may be reached by the present version of the PTFE ICAS experiment. However, in PTFE ICAS experiments the first-order inhomogeneity contributions to the scalar product $\boldsymbol{d}_e\boldsymbol{\mathcal{E}}_{\rm ext}$ cancel during the photons travel forward and backward within the cavity. We should mention that the molecular levels with $\Omega = \frac{1}{2}, \frac{3}{2}$ should be more robust with respect to the field inhomogeneity systematic errors.

Another systematic error also discussed in Ref. [42] for the ACME Collaboration and also applicable to the PTFE is the geometry phase. It concerns the time-dependent magnetic or electric fields (mentioned above in Sec. IVB) that may change at the moment of the electric field switching and mimic therefore the $\mathcal{P}$, $\mathcal{T}$-odd effect. Imagined in the harmonic form, these systematic errors may be reduced to the arrival of the Berry (geometry) phase, which may influence the transition frequencies. These systematic errors were specifically addressed in Ref. [45]. It was shown that in an experiment of the ACME Collaboration type or PTFE ICAS type, if one were to perform the experiments with separate components of the Ω doublet (as mentioned in Sec. IVB), the systematic errors due to the geometry phase can be reduced to the level of 10^{-35} e cm, i.e., lower than the eEDM bound predicted in this paper.

V. CONCLUSION

Comparing the results given in Table I with recent achievements [9,14], we can expect that the PTFE ICAS experiment I could improve these results by an order of magnitude. The PTFE ICAS experiment II could give an improvement of three orders of magnitude. With the employment of ICAS techniques at the level already reached in Ref. [33], the PTFE ICAS experiment II could give another order of magnitude improvement, i.e., it would nearly close the gap between the experiment and theoretical predictions [21].

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