

***CP*-Violating Effect of the Th Nuclear Magnetic Quadrupole Moment: Accurate Many-Body Study of ThO**

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Investigations of *CP* violation in the hadron sector may be done using measurements in the ThO molecule. Recent measurements in this molecule improved the limit on the electron electric dipole moment (EDM) by an order of magnitude. Another time-reversal (*T*) and parity (*P*)-violating effect in ²²⁹ThO is induced by the nuclear magnetic quadrupole moment. We perform nuclear and molecular calculations to express this effect in terms of the strength constants of *T*, *P*-odd nuclear forces, neutron EDM, QCD vacuum angle θ , quark EDM, and chromo-EDM.

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Introduction.—The best limits on the electric dipole moment (EDM) of the proton and *T*, *P*-violating nuclear forces have been obtained using measurements of the Hg atom EDM [1,2]. The Hg EDM measurements also give a limit on the neutron EDM, which is only twice weaker than that from the direct neutron EDM measurement. There are also studies in other diamagnetic atoms (Xe, Ra, Rn) [2–5] and the TIF molecule [6,7].

The problem is that the nuclear EDM, d_N , in neutral atoms and molecules is screened by electrons (the Schiff theorem) and can not be measured directly. Therefore, the atomic EDM in heavy diamagnetic atoms is generated by the Schiff moment [8,9]. The Schiff moment is $\sim r_N^2 d_N$, where r_N is a very small nuclear radius [10] on the atomic scale. As a result, the atomic EDM produced by the nuclear Schiff moment is significantly smaller than the nuclear EDM.

The magnetic interaction between the nuclear moments and electrons is not screened. The lowest *T*, *P*-odd magnetic moment is the nuclear magnetic quadrupole moment (MQM). To have MQM working, we should consider paramagnetic atoms and molecules, where the electron angular momentum is not zero, and electrons produce a magnetic field interacting with MQM (MQM interacts with the gradient of the magnetic field). This interaction mixes states of opposite parity and generates atomic or molecular EDM. In molecules, because of the closeness of rotational levels of opposite parity, permanent molecular EDM can be strongly enhanced compared to that in atoms. It was shown in Ref. [8] that in paramagnetic atoms and molecules MQM induces larger EDM than the Schiff moment (see also Refs. [11,12]).

In addition, it was shown in Ref. [13] that in deformed nuclei MQM has a collective nature and is significantly enhanced. The following is a qualitative explanation. The *T*, *P*-odd nuclear forces create a spin hedgehog; i.e., the nucleon spins obtain a small projection on the radial

direction $\langle \mathbf{s} \rangle \propto \mathbf{r}$. Because of the Gauss theorem, the spherically symmetric configuration of the magnetic moments does not produce any magnetic field, as the magnetic monopole is forbidden. However, in deformed nuclei the spin hedgehog produces the higher multipole, MQM. Remarkably, in all molecules of current experimental interest, a heavy atom has a deformed nucleus and this collective enhancement works (in isotopes with nuclear spin $I > 1/2$, where MQM exists). Note that an ordinary electric quadrupole moment is also enhanced in deformed nuclei.

The authors of Refs. [8,14,15] suggested the use of paramagnetic molecules to measure *T*, *P*-violating effects produced by MQM. Heavy diatomic molecules with a ³ Δ_1 electron term look especially promising [15]. There are several reasons. First, the effect of MQM rapidly increases with the nuclear charge Z , as $Z^2 R_M$, where R_M is the relativistic factor slowly increasing with Z [8]. Second, the ³ Δ_1 electron term has Ω -doublet structure with a very small interval between the opposite parity levels. This allows one to polarize the molecule by a weak electric field and cancel some systematic errors since the effect on the doublet components has an opposite sign [16–18]. The magnetic moment of the ³ Δ_1 electron term is very small, and this is another reason for reducing the systematic errors. Finally, a new experimental technique was developed that allows one to improve the limit on the electron electric dipole moment using the ThO molecule by more than an order of magnitude [19].

The aim of the present Letter is to perform accurate calculations of the MQM effect in ThO, which should allow one to measure nuclear *CP*-violating interactions and nucleon EDM using ThO experiments [19]. These measurements provide a method to search for physics beyond the standard model and test unification theories.

The *T*, *P*-odd electromagnetic interaction of the nuclear magnetic quadrupole moment with electrons is described by the Hamiltonian [11,20,21]

$$H = -\frac{M}{2I(2I-1)} T_{ik} \frac{3}{2} \frac{[\boldsymbol{\alpha} \times \mathbf{r}]_i r_k}{r^5}, \quad (1)$$

where Einstein's summation convention is implied, $\boldsymbol{\alpha}$ are the 4×4 Dirac matrices

$$\boldsymbol{\alpha} = \begin{pmatrix} 0 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & 0 \end{pmatrix},$$

$\boldsymbol{\sigma}$ are the Pauli matrices, \mathbf{r} is the displacement of the electron from the Th nucleus, \mathbf{I} is the nuclear spin, and M is the nuclear MQM,

$$M_{i,k} = \frac{3M}{2I(2I-1)} T_{i,k}, \quad (2)$$

$$T_{i,k} = I_i I_k + I_k I_i - \frac{2}{3} \delta_{i,k} I(I+1). \quad (3)$$

In the subspace of the $\pm\Omega$ states, the Hamiltonian (1) is reduced to the following effective molecular Hamiltonian [8]:

$$H_{\text{eff}} = -\frac{W_M M}{2I(2I-1)} \mathbf{S} \hat{\mathbf{T}} \mathbf{n}. \quad (4)$$

Here, parameter W_M , defined in Eq. (14), will be found from the molecular calculations, \mathbf{S} is the effective electron spin [14], $S = |\Omega| = 1$, \mathbf{n} is a unit vector directed along the molecular axis ζ from Th to O, and $\Omega = \mathbf{J}^e \cdot \mathbf{n}$ is the projection of the total electronic angular momentum \mathbf{J}^e on the molecular axis. Note that contrary to the $|\Omega| = 1/2$ case, Hamiltonians (1) and (4) do not mix $\Omega = \pm 1$ components. Neglecting the interaction between different rotational levels, one can obtain that the MQM energy shift is

$$\delta(J, F) = (-1)^{1/2+\Omega/2+I+F+1} C(J, F) W_M M, \quad (5)$$

$$C(J, F) = \frac{(2J+1)}{2} \frac{\begin{pmatrix} J & 2 & J \\ -\Omega & 0 & \Omega \end{pmatrix}}{\begin{pmatrix} I & 2 & I \\ -I & 0 & I \end{pmatrix}} \begin{Bmatrix} J & I & F \\ I & J & 2 \end{Bmatrix}, \quad (6)$$

where (\dots) means elements with $3j$ and $\{\dots\}$ with $6j$ symbols [22], and F is the total angular momentum. For ^{229}ThO ($I = 5/2$) and ground rotational level $J = 1$, Eq. (5) gives the MQM energy shifts $|\delta(J, F)|$, equal to $0.14W_M M$, $0.16W_M M$, and $0.05W_M M$ for $F = 3/2, 5/2, 7/2$, correspondingly.

Nuclear magnetic quadrupole moment.—Khriplovich calculated a nuclear MQM produced by the valence proton EDM [23]. In Ref. [8] it was shown that a 1–2 orders of magnitude greater MQM is produced by the T , P -odd nuclear forces. An analytical estimate of this MQM may be obtained if the strong potential $U(r)$ is proportional to the nucleon number density $\rho(r)$ (the strong interaction is short

range). In this approximation the T , P -odd nuclear potential $W = (\boldsymbol{\sigma} \cdot \nabla \rho) \eta G / (2^{3/2} m_p) \approx \xi (\boldsymbol{\sigma} \cdot \nabla U)$, where $\xi = [\rho(0)/U(0)] \eta G / (2^{3/2} m_p) \approx -2 \times 10^{-21} \eta$ [cm], η is the dimensionless strength constant of the T , P -odd nuclear potential, m_p is the proton mass, and G is the Fermi coupling constant. The perturbed valence nucleon wave function in the potential $U + \xi (\boldsymbol{\sigma} \cdot \nabla U)$ may be expressed analytically in terms of the unperturbed wave function ψ_0 in the potential U , $\psi = [1 + \xi (\boldsymbol{\sigma} \cdot \nabla)] \psi_0$, and the analytical expression for the valence nucleon MQM may be obtained with [8]

$$M = [d - 2 \times 10^{-21} \eta (\mu - q) (e \cdot \text{cm})] \lambda_p (2I - 1) t_I, \quad (7)$$

where $t_I = 1$ for $I = l + 1/2$ and $t_I = -I/(I+1)$ for $I = l - 1/2$, I and l are the total and orbital angular momenta of a valence nucleon, the nucleon magnetic moments are $\mu_p = 2.79$ for valence proton and $\mu_n = -1.91$ for valence neutron, $q_p = 1$ and $q_n = 0$, $\lambda_p = \hbar/m_p c$, and the contribution of the valence nucleon EDM d is taken from Ref. [23].

In a deformed nucleus, the strong field splits orbitals with different absolute values $|I_z|$ of the projections of the angular momentum on the nuclear symmetry axis. The sum over all I_z gives zero contribution to MQM. However, because of the difference of energies for different $|I_z|$ some of the I_z orbitals are vacant, and the remaining contribution is not zero. As a result, the MQM of a deformed nucleus in the “frozen” frame (rotating together with a nucleus) may be estimated using the following formula [13]:

$$M_{zz}^{\text{nucl}} = \sum M_{zz}^{\text{single}}(I, I_z, l) n(I, I_z, l), \quad (8)$$

where the sum goes over occupied orbitals, $M_{zz}^{\text{single}}(I, I_z, l)$ is given by Eqs. (7) and (2), $T_{zz} = 2I_z^2 - \frac{2}{3} I(I+1)$, and $n(I, I_z, l)$ are the orbital occupation numbers. For the ^{229}Th nucleus, the occupation numbers have been found using the diagrams presented in Ref. [24]: 13 neutrons on the orbitals $g_{9/2}$, $I_z = 5/2, \pm 3/2, \pm 1/2$; $j_{15/2}$, $I_z = \pm 5/2, \pm 3/2, \pm 1/2$; $i_{11/2}$, $I_z = \pm 1/2$, and 6 protons on the orbitals $h_{9/2}$, $I_z = \pm 3/2, \pm 1/2$; $i_{13/2}$, $I_z = \pm 1/2$.

The MQM in the laboratory frame $M \equiv M_{\text{lab}}$ can be expressed via MQM in the rotating frame (8) as

$$M^{\text{lab}} = \frac{I(2I-1)}{(I+1)(2I+3)} M_{zz}^{\text{nucl}} = \quad (9)$$

$$[7 \times 10^{-20} \eta_n \mu_n (e \cdot \text{cm}) - 19 d_n] \lambda_p, \quad (10)$$

where $I = 5/2$ is the ^{229}Th nuclear spin. The proton contribution is small due to an accidental cancellation of the contributions of different orbitals.

The T , P -odd nuclear forces are dominated by the π_0 meson exchange [8,25]. Therefore, we may express the strength constants via strong πNN coupling constant

$g = 13.6$ and T , P -odd πNN coupling constants corresponding to the isospin channels $T = 0, 1, 2$: $\eta_n = 5 \times 10^6 g(\bar{g}_1 + 0.4\bar{g}_2 - 0.2\bar{g}_0)$. The numerical coefficient here was obtained as a product of two factors: $[Gm_\pi^2/2^{1/2}]^{-1} = 6.7 \times 10^6$ from the π -meson exchange in the zero-range limit and the factor of 0.7 corresponding to the zero-range reduction of the finite range interaction due to the π_0 exchange [8,25]. For the charge meson exchange, the reduction factor is about 0.16 (since the exchange interaction contains a small overlap of the proton and neutron wave functions in the π^-pn vertex), and we neglect this contribution. We have also included two additional correction factors for the value of M . More accurate numerical calculations in the Saxon-Woods potential [8,25] give larger values of MQM (the factor ~ 1.2) than the simple analytical solution in Eq. (7). On the other hand, the many-body corrections reduce the effective strength constants of the T , P -odd potential $\eta \sim 1.5$ times [13,26]. The magnitude of these corrections may also serve as an estimate of the accuracy of MQM calculations in spherical nuclei, $\sim 50\%$ (the error in the deformed nuclear case may be larger, and the corresponding accuracy estimate requires numerical calculations of MQM). As a result, we obtain

$$M(g) = -[g(\bar{g}_1 + 0.4\bar{g}_2 - 0.2\bar{g}_0) + d_n/(1.4 \times 10^{-14} e \cdot \text{cm})] \times 6 \times 10^{-27} e \cdot \text{cm}^2. \quad (11)$$

Possible CP violation in the strong interaction sector is described by the CP violation parameter θ . According to Ref. [27] $g\bar{g}_0 = -0.37\theta$. This gives the following value of MQM for ^{229}Th :

$$M(\theta) = -4 \times 10^{-28} \tilde{\theta} e \cdot \text{cm}^2. \quad (12)$$

Finally, we can express MQM in terms of the quark chromo-EDM \tilde{d}_u and \tilde{d}_d using the relations $g\bar{g}_1 = 4 \times 10^{15} (\tilde{d}_u - \tilde{d}_d)/\text{cm}$, $g\bar{g}_0 = 0.8 \times 10^{15} (\tilde{d}_u + \tilde{d}_d)/\text{cm}$ [28]

$$M(\tilde{d}) = -2 \times 10^{-11} (\tilde{d}_u - \tilde{d}_d) e \cdot \text{cm}. \quad (13)$$

The contributions of d_n to MQM in Eqs. (11)–(13) are from 1 to 2 orders of magnitude smaller than the contributions of the nucleon CP -odd interactions.

Electronic structure calculation.—To obtain W_M in the ThO molecule, one can evaluate the following matrix element [15]:

$$W_M = \frac{3}{2} \frac{1}{\Omega} \langle \Psi_{3\Delta_1} | \sum_i \left(\frac{\alpha_i \times \mathbf{r}_i}{r_i^5} \right)_\zeta r_\zeta | \Psi_{3\Delta_1} \rangle, \quad (14)$$

where Ψ is the electronic wave function of the considered ThO state.

In the ionic model, the $^3\Delta$ state of ThO corresponds to the [...] $5s^2 5p^6 5d^{10} 6s^2 6p^6 7s^1 6d^1$ effective electron configuration of Th and $1s^2 2s^2 2p^6$ of O. Internuclear distance between Th and O, 3.511 a.u., for the state is known experimentally [29,30] and is also confirmed theoretically [31].

Similar to the hyperfine interaction between the nucleus magnetic moment and electrons, only the electron spin density in the vicinity of Th nucleus contributes to the matrix element (14). As contributions from core electrons with opposite spins (closed shells) compensate each other, the main contribution comes from open-shell (unpaired) electrons of s and $p_{3/2}$ type due to their largest amplitude near the nucleus (the matrix element between s and $p_{1/2}$ is zero since the operator in Eq. (14) is a second rank tensor). However, as was shown in Ref. [32] the exchange interaction with open-shell $7s^1 6d^1$ electrons can result in a spin polarization of the subvalence closed-shell $5s, 5p, 5d, 6s, 6p$ states of Th and $1s 2s 2p$ of O which, in turn, leads to a non-negligible perturbation of the hyperfine structure interaction and other properties dependent on the effective spin.

It was shown in Refs. [7,33] that the relativistic four-component problem of evaluating matrix elements such as Eq. (14) can be effectively divided into two steps. For this, the space around a given heavy atom (Th) is divided into valence and core regions. At the first step, one calculates the molecular wave function within the effective Hamiltonian [34,35]. It is built in such a way that the corresponding wave function is very accurate in the valence region but has incorrect behavior in the core region. At the second step, the true four-component behavior of a wave function is restored in the core region using the procedure [33] based on a proportionality of valence and virtual (unoccupied in the reference Slater determinant) spinors in the inner-core region of the heavy atom.

To solve the many-body problem of finding the wave function Ψ , we used the “all-order” method with respect to single and double excitations, in which some of the most important connected triple excitations are also taken into account, i.e., the coupled cluster with single, double, and perturbative treatment of triple cluster amplitudes [36]. To account for spin-polarization effects, we considered excitations from the 38 highest occupied spinors. For the description of electron density in ThO, we used the basis sets centered on Th and O from Refs. [32,37] (see the Supplemental Material for an explicit specification of the basis sets used [38]). For molecular calculations we used codes from Refs. [31,40–44].

The final calculated value of W_M is $1.66(12) \times 10^{33} \text{ Hz}/(e \text{ cm}^2)$. Theoretical uncertainty was estimated with a procedure similar to that given in Ref. [32]. We have found that the effect of spin polarization of subvalence closed shells contributes about $+0.1 \times 10^{33} \text{ Hz}/(e \text{ cm}^2)$ while the spin-orbit interaction of valence and subvalence

electrons contributes $-0.08 \times 10^{33} \text{ Hz}/(e \text{ cm}^2)$. The result of the earlier performed “semiempirical” estimate [15], $1.9 \times 10^{33} \text{ Hz}/(e \text{ cm}^2)$, is in good agreement with the current *ab initio* calculations [45].

One can express the MQM energy shift $(-1)^{1/2+\Omega/2+I+F+1} C(J, F) W_M M$ in terms of the fundamental *CP*-violating physical quantities $\tilde{\theta}$ and $\tilde{d}_{u,d}$ using Eqs. (12) and (13). For the lowest rotational level, for which the coefficient $|C(J=1, F=5/2)| = 0.16$ reaches a maximum value, we have

$$0.16 W_M M = -11 \times 10^{10} \tilde{\theta} \cdot \mu\text{Hz} \quad (15)$$

$$0.16 W_M M = -5 \times \frac{10^{27} (\tilde{d}_u - \tilde{d}_d)}{\text{cm}} \cdot \mu\text{Hz}. \quad (16)$$

The current limits on $|\tilde{\theta}|$ and $|\tilde{d}_u - \tilde{d}_d|$ ($|\tilde{\theta}| < 2.4 \times 10^{-10}$, $|\tilde{d}_u - \tilde{d}_d| < 6 \times 10^{-27} \text{ cm}$, see Ref. [2]) correspond to the shifts $|0.16 W_M M| < 26$ and $30 \mu\text{Hz}$, respectively. The current accuracy in measurements of the energy shift produced by the *e*EDM in ^{232}ThO is $700 \mu\text{Hz}$ [19,46,47]. However, it is anticipated in Refs. [48,49] that it can be improved by at least a factor of 10 over the next 5 years. Therefore, similar measurements on ^{229}ThO can, in principle, result in large enough frequency shifts produced by the nuclear MQMs to compete in the improvement of limits on the $\tilde{\theta}$ term and on the difference of the quark chromo-EDMs ($\tilde{d}_u - \tilde{d}_d$) with other experiments.

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- [45] Such a coincidence is not an accident. Since there were no experimental data obtained for the hyperfine structure constants of the $H^3\Delta_1$ state of ThO up to now, the given “semiempirical” value is obtained on the basis of our previous *ab initio* calculation of an effective electric field [13], and the current and our previous *ab initio* calculations are very close methodologically.
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- [47] $700 \mu\text{Hz}$ is the 1-sigma statistical uncertainty on the energy shift (not splitting) of the single level. It can be obtained if the statistical error of $3.7 \times 10^{-29} e \cdot \text{cm}$ [19] is multiplied by the used effective electric field $2 \times 10^{25} \text{ Hz}/(e \cdot \text{cm})$.
- [48] P. Hess, thesis, Department of Physics, Harvard University, 2014; see <http://laserstorm.harvard.edu/edm/publications.html>.
- [49] B. N. Spaun, thesis, Department of Physics, Harvard University, 2014; see <http://laserstorm.harvard.edu/edm/publications.html>.