Enhancement of the electron electric dipole moment in gadolinium 3+

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There have been recent suggestions for searching for the electron electric dipole moment, using solid state experiments with compounds containing Gd$^{3+}$ ions. These experiments could improve the sensitivity compared to present atomic and molecular experiments by several orders of magnitude. The analysis of the problem requires a calculation of the enhancement coefficient $K$ for the electron electric dipole moment in the Gd$^{3+}$ ion. In this work we perform this calculation. The result is $K \approx -4.9 \pm 1.6$. Limitations of the accuracy of the calculation are mainly due to the lack of data on Gd$^{3+}$ excitation spectra. We formulate which quantities have to be measured and/or calculated to improve the accuracy.

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I. INTRODUCTION

A violation of the combined symmetry of charge conjugation (C) and parity (P) has been discovered in the decay of the $K^0$ meson about 40 years ago. The exact origin of this symmetry violation remains an enigma, although the standard model of electroweak interactions can describe these processes phenomenologically. It has also been proposed by Sakharov that the matter-antimatter asymmetry observed in our universe could have arisen from a CP-violating interaction active at an early stage of the big bang. The CP-violation implies a time-reversal (T) asymmetry and hence violation of the combined TP-symmetry, because there are strong reasons to believe that the combined CPT-symmetry should not be violated. An electric dipole moment (EDM) of a system in a stationary quantum state indicates a violation of the TP-symmetry; this is why searches for EDMs of elementary particles, atoms and molecules are a very important approach to the studies of violations of fundamental symmetries. In the present work we concentrate on the EDM of the electron $d_e$.

At present the best limitation on $d_e$ comes from the Berkeley experiment with an atomic thallium beam, $d_e < 1.6 \cdot 10^{-27} e$ cm. There are some ideas for improving the sensitivity. One way of improvement is an experiment with metastable levels of PbO molecules. A breakthrough could be achieved in solid state experiments. This idea was suggested by Shapiro in 1968. The application of strong electric fields to electrons bound within a solid would align the electric dipole moments of these electrons. This should lead to a simultaneous alignment of the electron spins; the magnetic field arising from this alignment could be detected experimentally. An experiment of this kind has been performed with nickel-zinc ferrite. However, due to experimental limitations, the result was not very impressive. Interest to the approach has been renewed recently due to suggestions of Lamoreaux and Hunter to perform similar experiments with gadolinium gallium garnet, Gd$_3$Ga$_5$O$_{12}$, and gadolinium iron garnet, Gd$_3$Fe$_5$O$_{12}$, employing new experimental techniques. First estimates of sensitivity promise to improve the current upper limit on the electron EDM by at least three orders of magnitude, depending on the experimental setup. A thorough analysis of the problem requires the calculation of the enhancement coefficient for the electron EDM in the Gd$^{3+}$ ion. We perform this calculation in the present work.

II. SINGLE PARTICLE CONTRIBUTION

The Gd$^{3+}$ ion has a nucleus with charge $Z = 64$ and 61 electrons. The 7 electrons in the outer shell occupy the 4$f$ orbitals. So the shell is half filled, and hence the total orbital angular momentum is zero, $L = 0$, and total spin $S = 7/2$. Let us consider the state with maximum z-projection of the spin. In the representation of second quantization the ground state wave function is of the form

$$|gs\rangle = f_{-3}^{\dagger} f_{-2}^{\dagger} f_{-1}^{\dagger} f_{0}^{\dagger} f_{1}^{\dagger} f_{2}^{\dagger} f_{3}^{\dagger} |0\rangle,$$

where $f_{m}^{\dagger}$ is the creation operator for a 4$f$ electron with spin $\sigma$ and z-projection of orbital angular momentum $l_z = m$. The TP-odd interaction of the electron EDM with the electric field $E$ is of the form, see e.g. Ref. [1]
\[ V_d = -e_d \gamma_0 \mathbf{\Sigma} \cdot \mathbf{E}, \quad (2) \]

where \( \gamma_0 \) and \( \mathbf{\Sigma} = \gamma_0 \gamma_0 \gamma \) are Dirac \( \gamma \)-matrices. Because of Schiff’s theorem [11] it is crucially important to account for very complex many-body screening effects, when working with the Hamiltonian \((2)\). Technically this means that the many-body perturbation theory practically is not convergent. The standard way [6] to avoid this complication is to split the Hamiltonian into two terms: \( V_d = -e_d \gamma_0 \mathbf{\Sigma} \cdot \mathbf{E} = -e_d \mathbf{\Sigma} \cdot \mathbf{E} - e_d (\gamma_0 - 1) \mathbf{\Sigma} \cdot \mathbf{E} \). Then due to Schiff’s theorem [11] the contribution of the first term to the EDM of the ion is identically zero, so one can reduce the interaction

\[ V_d \rightarrow V'_d = -e_d (\gamma_0 - 1) \mathbf{\Sigma} \cdot \mathbf{E}. \quad (3) \]

Perturbation theory with this operator is reasonably convergent.

In leading order of single particle perturbation theory the EDM of the ion \( D_{\text{sp}} \) is given by diagrams shown in Fig. 1.

![Diagram](image)

**FIG. 1.** Leading contribution to the Gd\(^{3+}\) EDM. The dashed line denotes the dipole moment \( e\mathbf{z} = ev \cos \theta \), and the cross denotes the TP-odd reduced interaction \( V'_d \), eq \((4)\).

The corresponding formula reads

\[ D_{\text{sp}} = 2 \sum_{mn} \frac{\langle 4f_m | e\mathbf{r} \cos \theta | n \rangle \langle n | V'_d | 4f_m \rangle}{E_{4f} - E_n} = K_{\text{sp}} e_d, \quad (4) \]

where the EDM enhancement coefficient is given by

\[ K_{\text{sp}} = 2 \sum_{mn} \frac{\langle 4f_m | (r/a_B) \cos \theta | n \rangle \langle n | (e\mathbf{a}_B/d_e) V'_d | 4f_m \rangle}{E_{4f} - E_n}. \quad (5) \]

Here \( e \) is the electron charge and \( a_B \) is the Bohr radius. We first consider 5\( d \) and 5\( g \) intermediate states, as the contribution of other states is much less. The E1 matrix elements are of the form

\[ \langle 4f_m | (r/a_B) \cos \theta | 5d_m \rangle = \frac{9 - m^2}{35} r_{4f,5d}, \quad (6) \]

\[ \langle 4f_m | (r/a_B) \cos \theta | 5g_m \rangle = \frac{16 - m^2}{63} r_{4f,5g}, \quad (7) \]

where \( r_{ik} \) is the E1-transition radial integral expressed in atomic units (Bohr radius),

\[ r_{ik} = \int_0^\infty R_i(r) R_k(r) r^3 \, dr. \quad (8) \]

\( R_i(r) \) is the radial wave function of the corresponding orbital, \( \int_0^\infty R_i^2(r) r^2 \, dr = 1 \).

The matrix element of \( V'_d \) has been calculated earlier, see e.g. Ref. [3]. In jj coupling scheme, and using a semiclassical approximation, it reads

\[ \langle n'j' l \pm 1 | (e\mathbf{a}_B/d_e) V'_d | nj l \rangle = -\frac{4Z^2 \alpha^2}{\gamma (4\gamma^2 - 1)} \cdot \frac{Z_{\text{eff}}^2}{(\nu' \nu)^{3/2}} E_0, \quad (9) \]

where \( \alpha = 1/137.036 \) is the fine structure constant, \( \gamma = \sqrt{(j + 1/2)^2 - (Z\alpha)^2} \), \( E_0 = 2 \text{ Ry} = 27.2 \text{ eV} = 2 \times 109,737 \text{ cm}^{-1} \) is the atomic energy unit, \( Z_{\text{eff}} = 4 \) is the effective charge for electron motion at large distances, and finally the effective principal quantum numbers \( \nu \) and \( \nu' \) are defined by the electron energy with respect to the ionization limit, \( E = -Z_{\text{eff}}^2 E_0/(2\nu^2) \). Our numerical calculations in Hartree-Fock approximation agree with the semiclassical result [3] within ten per cent. The ground state wave function [6] is given in LS coupling scheme, therefore we have to rewrite the operator \( V'_d \) in this representation. A simple calculation gives
Combining equations (5), (6), (8), and (9) and performing the summation over \( E \), the single particle contribution to the enhancement coefficient is given by:

\[
\langle 5d_{m/1} | (e \alpha_B / d_e) V_{dd}^f | 4f_{m/1} \rangle = -\sqrt{\frac{9 - m^2}{35}} \langle 5d_{5/2} | (e \alpha_B / d_e) V_{dd}^f | 4f_{5/2} \rangle,
\]

\[
\langle 5g_{m/1} | (e \alpha_B / d_e) V_{dd}^f | 4f_{m/1} \rangle = -\sqrt{\frac{16 - m^2}{35}} \langle 5g_{7/2} | (e \alpha_B / d_e) V_{dd}^f | 4f_{7/2} \rangle.
\]

As a result of the calculation, we obtained the following values of energies and radial integrals:

Note that from here on we skip the atomic energy unit \( E_0 \) in all equations, assuming that all energies are expressed in units of \( E_0 \). To find values of the parameters that appear in this equation, we have performed a Hartree-Fock calculation for the \( \text{Gd}^{3+} \) ion. It is known that a Hartree-Fock calculation for an open shell is not a uniquely defined procedure. In our calculation, we used the following averaging: we assumed that the shell is fully occupied, but the occupation number of each single particle \( f \)-orbital is \( 1/2 \). This is a crude approximation, it gives reasonable values of energy levels and radial integrals, but one cannot rely on this calculation as far as energy splittings are concerned.

As a result of the calculation obtained the following values of energies and radial integrals:

\[
E_{4f} = -1.65 \quad E_{5d} = -1.20 \quad E_{5g} = -0.32
\]

\[
r_{4f,5d} = 0.63 \quad r_{4f,5g} = 0.088
\]

Where available, experimental data should be used, but unfortunately the experimental data on this particular ion is scarce. Only the value of the \( 4f \) energy level (ionization limit) is known, see Ref. [12],

\[
E_{4f} = -355000 \text{ cm}^{-1} = -1.62 E_0.
\]

It agrees well with (11). From (12) and (11) one finds \( \nu_{4f} = 2.22 \), \( \nu_{5d} = 2.58 \) and \( \nu_{5g} = 5.0 \).

The most important is the \( E_{5d} - E_{4f} \) energy splitting. The accuracy of the present Hartree-Fock calculation is not sufficient to determine this splitting. There is reliable experimental data for the energy levels of \( \text{Gd}^{2+} \), \( \text{Gd}^{1+} \), and \( \text{Eu}^{2+} \), see Ref. [14]. Naive extrapolation of the splitting from these ions gives

\[
A : \quad E_{5d} - E_{4f} \approx 40,000 \text{ cm}^{-1} \approx 0.18 E_0.
\]

On the other hand there is experimental data for \( \text{Gd}^{3+} \) [13] that indicates

\[
B : \quad E_{5d} - E_{4f} \approx 100,000 \text{ cm}^{-1} \approx 0.45 E_0.
\]

Unfortunately Ref. [13] does not contain identifications of all the possible levels, therefore we cannot quite rely on the data. For this reason we will present two estimates of \( K \): one for the case (A), see (13), and another for the case (B), see (14). Experimental and/or theoretical determination of the \( E_{5d} - E_{4f} \) splitting would be the most important to improve the accuracy of the calculation of the enhancement coefficient \( K \).

Substituting values of the parameters listed above into eq. (10) we find the contribution of the \( 5d \) intermediate state to the EDM enhancement coefficient:

\[
A : \quad K_{5d} = -4.5,
\]

\[
B : \quad K_{5d} = -1.8
\]

The contribution of the \( 5g \) intermediate state is very small. We also estimate the contribution of the higher \( d \)-levels (mainly the continuous spectrum) as

\[
A, B : \quad K_{\text{nd}, n>5} \approx -1.
\]

Altogether this gives the following value of the single particle contribution to the EDM enhancement coefficient:

\[
A : \quad K_{sp} = -5.5,
\]

\[
B : \quad K_{sp} = -2.8
\]
III. MANY-BODY CORRECTIONS

In the situation with cesium or any other atom with valent $s$- or $p$-electrons the single particle estimate is satisfactory. However here we have $f$-electrons that have a very small wave function in the vicinity of the nucleus. As a result the single particle contribution is strongly suppressed. Technically this suppression is reflected in eq. (8); the matrix element is proportional to $1/\gamma^3$, and for $f_{5/2}$-electrons $\gamma \approx 3$, while for $s_{1/2}$- or $p_{1/2}$-electrons $\gamma \approx 0.88$. Therefore it is very important to estimate the many-body corrections. The leading many-body corrections to the EDM enhancement coefficient $K_{mb}$ are shown in diagrams Fig. 2, where the wavy line denotes residual Coulomb interaction

$$V_C = \frac{1}{|r_i - r_j|} = \sum_{k=0}^{\infty} \sum_{q=-k}^{k} \frac{4\pi}{2k+1} \cdot \frac{r_<^k}{r_<^{k+1}} Y_{kq}(\Omega_i) Y_{kq}(\Omega_j).$$

We only account for diagrams with momentum of the Coulomb quantum $k$ not higher than 2. The contribution of each diagram from Fig. 2 must be doubled, because an opposite order of operators is also possible.

![Diagram](https://example.com/diagram.png)

**FIG. 2.** Leading many-body corrections to the Gd$^{3+}$ EDM. The dashed line denotes the dipole moment $e\cos \theta$, the cross denotes the TP-odd reduced interaction $V_d^{r}$, eq (1), and the wavy line denotes residual Coulomb interaction

The formula for the many-body contribution to the enhancement coefficient due to the diagrams in Fig. 2 reads

$$K_{mb} = -2 \sum_m \left( \langle 4f_m| r/a_B \cos \theta| 5d_m \rangle \langle 5p_{1/2}| 5d_m| V_C| 6s \rangle \langle 6s| e a_B/d_e| V_d^{r}| 5p_{1/2} \rangle (E_{5f} - E_{5d}) (E_{6p} - E_{6s}) \right.\left. + \langle 5s| e a_B/d_e| V_d^{r}| 6p_{1/2} \rangle \langle 4f_m| r/a_B \cos \theta| 5d_m \rangle \langle 6p_{1/2}| 5d_m| V_C| 5s \rangle \langle 4f_m \rangle (E_{5s} - E_{6p}) (E_{4f} + E_{5s} - E_{5d} - E_{6p}) \right.\left. + \langle 4f_m| r/a_B \cos \theta| 5d_m \rangle \langle 5s| e a_B/d_e| V_d^{r}| 6p_{1/2} \rangle \langle 6p_{1/2}| 5d_m| V_C| 5s \rangle \langle 4f_m \rangle (E_{4f} - E_{5d}) (E_{4f} + E_{5s} - E_{5d} - E_{6p}) \right),$$

We have only included $s - p_{1/2}$ matrix elements of $V_d^r$ and only the intermediate states involving $5d$ electrons which give the main contribution. All the diagrams in Fig. 2 are exchange ones, this is why the sign in eq. (19) is negative.

Matrix elements of the Coulomb interactions are of the form

$$\langle 5p_{1/2}| 5d_m| V_C| 6s \rangle \langle 4f_m \rangle = -\frac{1}{5} \sqrt{\frac{9 - m^2}{35}} F^{(2)}(5p, 4f; 5d, 6s)$$

$$\langle 6p_{1/2}| 5d_m| V_C| 5s \rangle \langle 4f_m \rangle = -\frac{1}{5} \sqrt{\frac{9 - m^2}{35}} F^{(2)}(6p, 4f; 5d, 5s)$$

where $F^{(k)}(i; j; h, l)$ is the usual Coulomb radial integral,

$$F^{(k)}(i; j; h, l) = \int_0^\infty \int_0^\infty \frac{1}{r_>^k r_<^{k+1}} R_i(r_1) R_j(r_1) R_h(r_2) R_l(r_2) r_1^2 r_2^2 \, dr_1 \, dr_2,$$

expressed in atomic units.

Using matrix elements (18), (19) and (21), and performing the summation over $m$ in (19), we obtain the following expression for the leading many-body correction to the enhancement coefficient

$$K_{mb} = -\frac{8Z^2 a_0^2 Z_{de}^2}{5\gamma_{1/2}^2 (4\gamma_{1/2}^2 - 1)} r_{4f, 5d} \left\{ \frac{F^{(2)}(5p, 4f; 5d, 6s)}{(v_{6s} v_{6p})^{3/2}} \cdot \frac{1}{(E_{4f} - E_{5d})} \cdot \frac{1}{(E_{6p} - E_{6s})} \right.\left. + \frac{F^{(2)}(6p, 4f; 5d, 5s)}{(v_{6s} v_{6p})^{3/2}} \cdot \frac{1}{(E_{4f} - E_{5d} + E_{5s} - E_{6p})} \left( \frac{1}{(E_{4f} - E_{5d})} + \frac{1}{(E_{5s} - E_{6p})} \right) \right\}.$$
Like the parameters presented in (11) the energy levels and radial integrals in this formula have been calculated using a Hartree-Fock method

\[
E_{5s} = -3.15 \quad E_{5p} = -2.26 \quad E_{6s} = -1.03 \quad E_{6p} = -0.84
\]

\[
F^{(2)}(5p,4f;5d,6s) = 0.028 \quad F^{(2)}(6p,4f;5d,5s) = -0.023.
\]

The corresponding effective principal quantum numbers are \(\nu_{5s} = 1.59\), \(\nu_{5p} = 1.88\), \(\nu_{6s} = 2.78\), \(\nu_{6p} = 3.09\). Using the \(E_{5d} - E_{4f}\) energy splitting given in (13) and (14) and substituting all the parameters into eq. (22), we find the many-body correction due to the 6s-, 6p-, and 5d- intermediate states, see Fig.2

\[
A: \quad K_{mb1} \approx -0.7,
B: \quad K_{mb1} \approx -0.3.
\]

One should also perform the summation over higher s-, p-, and d- intermediate states. We estimate this contribution as

\[
A, B: \quad K_{mb2} \approx -0.2
\]

Combining (24) and (23) one finds the leading many-body correction to the enhancement coefficient

\[
A: \quad K_{mb} \approx -0.9,
B: \quad K_{mb} \approx -0.5.
\]

It is substantially smaller than the single particle contribution (17), so the many-body perturbation theory is convergent. The final result for the electron EDM enhancement coefficient, \(K = K_{sp} + K_{mb}\), reads

\[
A: \quad K \approx -6.4,
B: \quad K \approx -3.3.
\]

We recall that the case A corresponds to the energy splitting (13), and the case B corresponds to (14).

IV. CONCLUSION

We have calculated the electron EDM enhancement coefficient in the Gd\(^{3+}\) ion. The single particle contribution as well as the leading many-body corrections have been taken into account. The result is \(K \approx -4.9 \pm 1.6\). The main reason for such a large uncertainty lies in the unknown energy splitting \(E_{4f} - E_{5d}\). Experimental and/or theoretical determination of the splitting in Gd\(^{3+}\) would be the most important step for improving the accuracy of the enhancement coefficient calculation.

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