Energies, transition rates, and electron electric dipole moment enhancement factors for Ce IV and Pr V

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Energies, transition rates, and electron electric dipole moment (EDM) enhancement factors are calculated for low-lying states of Ce IV and Pr V using relativistic many-body perturbation theory. This study is related to recent investigations of the more complicated Gd IV ion, which is promising for electron EDM experiments. The ions Ce IV and Pr V both have a single valence electron, permitting one to carry out reliable ab-initio calculations of energy levels, transition rates and other atomic properties using well developed computational methods.

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I. INTRODUCTION

Recently, searches for an electron electric dipole moment (EDM) in atoms and molecules have gained considerable interest. Since an electron EDM violates time-reversal symmetry and parity, the discovery of an EDM would have many implications for modern fundamental theories (a general overview is given by Khriplovich and Lamoreaux [1]). The best limit on the value of the electron EDM was obtained by Regan et al. [2] in Tl, $d_e < 1.6 \times 10^{-27}$ e cm. Although there is a large enhancement ($\sim 600$) of the electron EDM in Tl, the density of atoms in a beam is much lower than in a solid-state system. To utilize the advantage of high densities, it has been proposed to use Gadolinium Gallium Garnet Gd$_3$Ga$_5$O$_{12}$ or Gadolinium Iron Garnet Gd$_3$Fe$_5$O$_{12}$ (densities $\sim 10^{22}$/cc) in EDM experiments [3, 4]. There are various features of these materials that make them particularly useful for such experiments. For example, magneto-electric effects are forbidden owing to the FCC symmetry of the crystals, simplifying the exclusion of systematics; the crystal Gd$_3$Fe$_5$O$_{12}$ has a very high resistivity ($\gg 10^{16}$ $\Omega$-cm); and spin alignment is relatively easy. The electron EDM enhancement for the Gd$^{3+}$ ion ($\sim 2 - 3$) is much smaller than for Tl; nevertheless there is a substantial gain in the number of atoms.

Motivated by proposed EDM measurements, calculations of EDM enhancement have been performed recently [5, 6]. Core polarization effects in Gd IV were discussed by Dzuba et al. [6] and found to decrease the size of EDM enhancement factor from -3.3 to -2.2. Thus, there is a strong sensitivity of the EDM enhancement to many-body effects and further investigation is necessary.

The ion Gd$^{3+}$, which has a 4f$^7$ ground-state configuration is extremely complicated and difficult for atomic theory, though some understanding was gained by Dzuba et al. [6] using a relativistic configuration-interaction (RCI) method and the widely used Cowan [8] code. As mentioned above, core polarization was found to be substantial. For example, to match the experimental $4f \rightarrow 5d$ transition energies, scaling factors of 0.8 and 0.85 for Coulomb integrals were used in the RCI and Cowan codes, respectively. In addition, a polarization potential was introduced in RCI code to match experimental energies and a systematic energy shift of $\sim 18,000$ cm$^{-1}$ was made in the Cowan code. This shift can be attributed to core polarization by a single valence electron. An ab-initio investigation of correlation should help to understand these empirical adjustments. The simplest ions (those having a single 4f valence electron) which exhibit core-polarization effects similar to Gd IV are Ce IV and Pr V. To gain a clearer understanding of the role of core polarization and other many-body effects in Ce IV and Pr V, we calculate energies, dipole transition matrix elements, and EDM enhancement factors for low lying states using relativistic many-body perturbation theory (MBPT).

The present calculations of energies are carried out to third order in MBPT using the methods developed earlier to study the Li, Na, and Cu isoelectronic sequences [9, 10, 11]. The calculations of transition matrix elements is also carried out to third order in MBPT using the methods developed in [12] to treat the alkali atoms and alkalilike ions. Here, we use the gauge-independent version of the third-order MBPT code described in [13].
The calculations of electron EDM enhancement factors, which involve a sum over intermediate states, are carried out in the random-phase approximation (RPA) following a procedure similar to that described in [4].

It should be mentioned that Ce\(^{+3}\), which has a 4\(f\)\(_{5/2}\) ground state and an observed magnetic moment 2.3-2.5 \(\mu_B\) can be embedded into garnet crystals and used in EDM experiments of the type proposed in [4]. In those experiments, a strong electric field polarizes ions which in turn produce a small magnetic field that is measured in a sensitive SQUID detector. The magnetic field at low temperatures is proportional to the product of the electric dipole moment of the ion and its magnetic moment. The estimated EDM of Gd\(^{+3}\) as is a factor of about 3 larger than for Ce\(^{+3}\) and the magnetic moment of Gd\(^{+3}\) (7.9-8.0 \(\mu_B\)) is also about three times larger; therefore, there is an overall advantage of nine in Gd\(^{+3}\) compared to Ce\(^{+3}\). Nevertheless, the Ce\(^{+3}\) ion could still compete in setting experimental limits on the electron EDM owing to the fact that its ionic EDM has significantly smaller theoretical uncertainty.

**II. CALCULATION OF ENERGIES**

First-, second- and third-order Coulomb energies \(E^{(n)}\), \(n = 1, 3\), and first- and second-order Breit energies \(B^{(n)}\), \(n = 1, 2\), calculated using methods described in Refs. [4, 5, 11], are presented in Table I along with the resultant theoretical energies \(E_{\text{tot}}\) and predicted energies \(E_{\text{NIST}}\) from the National Institute of Standards and Technology (NIST) given by Martin et al. [15]. We see that second-order corrections are large and improve the accuracy of the first-order Dirac-Hartree-Fock (DHF) energies. Third-order MBPT further improves the ground state energy. However, the third-order correction is relatively large (roughly one-third of the second order) and overshoots the experimental value, which indicates that oscillations in higher orders are likely. Using a geometric progression with \(q = -1/3\) we can extrapolate the second- and third-order values to give a limiting ground-state energy \(-296420 \text{ cm}^{-1}\) for Ce\(^{+3}\), in close agreement with the predicted value from [15]. The corresponding extrapolation for Pr\(^{+4}\) gives \(-468290 \text{ cm}^{-1}\) differing from the predicted ground-state energy given in [13] by about 4000 cm\(^{-1}\). This rather large difference casts doubt on the threshold energy of Pr V predicted in [13] and explains the large differences with the NIST energies seen in lower half of Table I.

In Refs. [4, 5, 11], where the ionic ground-states were \(2s, 3s,\) and \(4s\), respectively, the third-order correction was uniformly much smaller than the second-order correction. The relatively large size of the third-order corrections in Ce IV and Pr V arise because of the double-well feature of the \(4f\) Coulomb potential discussed, for example, by Cheng and Froese-Fischer [14].

| \(nlj\) | \(E^{(1)}\) | \(B^{(1)}\) | \(E^{(2)}\) | \(B^{(2)}\) | \(E^{(3)}\) | \(E_{\text{tot}}\) | \(E_{\text{NIST}}\) | \(\delta E\) |
|-------|---------|---------|---------|---------|---------|---------|---------|-------|
| \(\text{Ce IV}\) | \(-261361\) | 616 | \(-64747\) | \(-2552\) | 15672 | \(-294372\) | \(-296470\) | 2098 |
| \(\text{Pr V}\) | \(-431686\) | 798 | \(-48799\) | \(-3008\) | 16270 | \(-466167\) | \(-466000\) | \(-2167\) |

**TABLE I: First-order (DHF) energies \(E^{(1)}\), second- and third-order Coulomb energies \(E^{(2)}\) and \(E^{(3)}\), first- and second-order Breit corrections \(B^{(1)}\) and \(B^{(2)}\) and totals \(E_{\text{tot}}\) for Ce IV and Pr V are compared with predicted energies \(E_{\text{NIST}}\) given by Martin et al. [13]. Units: \(\text{cm}^{-1}\).**

| \(m_{lj}\) | \(E^{(1)}\) | \(B^{(1)}\) | \(E^{(2)}\) | \(B^{(2)}\) | \(E^{(3)}\) | \(E_{\text{tot}}\) | \(E_{\text{NIST}}\) | \(\delta E\) |
|-------|---------|---------|---------|---------|---------|---------|---------|-------|
| \(\text{Ce IV}\) | \(-261361\) | 616 | \(-64747\) | \(-2552\) | 15672 | \(-294372\) | \(-296470\) | 2098 |
| \(\text{Pr V}\) | \(-431686\) | 798 | \(-48799\) | \(-3008\) | 16270 | \(-466167\) | \(-466000\) | \(-2167\) |

**III. CALCULATION OF TRANSITION MATRIX ELEMENTS AND TRANSITION RATES**

Transition matrix elements provide another test of quality of atomic-structure calculations and another measure of the size of correlation corrections. Third-order MBPT reduced matrix elements for transitions between low-lying states of Ce\(^{+3}\) and Pr\(^{+4}\) are presented in Table II. The first-order reduced matrix elements \(Z^{(1)}\) are obtained from length-form DHF calculations. Length- and velocity-form matrix elements differ typically by 10%. Second-order matrix elements in the table \(Z^{(2)}\), which include \(Z^{(1)}\), are extended to include all higher-order corrections associated with the random-phase approximation. These second-order calculations are practically gauge independent. In the present calculations, length- and velocity-form matrix elements in the RPA agree to six or more digits. The third-order matrix elements \(Z^{(3)}\) include \(Z^{(2)}\) plus Brueckner-orbital (BO), structural radiation, and normalization corrections described, for example, in [12]. These calculations are carried out in a gauge-independent manner, including appropriate derivative terms, as described in [13]. We truncated our basis set to include only those partial waves.
TABLE II: Reduced matrix elements of the dipole operator in first-, second-, and third-order perturbation theory for transitions in Ce IV and Pr V.

| Transition          | Ce IV | Pr V |
|---------------------|-------|------|
|                     | Z(1)  | Z(2) | Z(3) | Z(1) | Z(2) | Z(3) |
| 4f_{5/2} → 5d_{3/2} | 1.498 | 0.972| 1.172| 1.146| 0.678| 0.706|
| 4f_{5/2} → 5d_{5/2} | 0.396 | 0.264| 0.308| 0.302| 0.186| 0.189|
| 4f_{5/2} → 5d_{7/2} | 1.799 | 1.193| 1.413| 1.370| 0.828| 0.852|
| 5d_{3/2} → 6p_{1/2} | 1.976 | 1.768| 1.682| 1.648| 1.471| 1.396|
| 5d_{3/2} → 6p_{3/2} | 8.363 | 0.756| 0.719| 0.690| 0.625| 0.592|
| 5d_{5/2} → 6p_{3/2} | 2.585 | 2.352| 2.229| 2.140| 1.950| 1.837|
| 6s_{1/2} → 6p_{1/2} | 2.847 | 2.482| 2.402| 2.560| 2.207| 2.136|
| 6s_{1/2} → 6p_{3/2} | 4.012 | 3.512| 3.401| 3.609| 3.125| 3.020|

with \( l \leq 8 \), and found that length- and velocity-form third-order reduced matrix elements agreed to 4 digits.

As can be seen in Table II, RPA corrections are very large, 10-40%, being largest for \( 4f \rightarrow 5d \) transitions, and must be taken into account. Such behavior can be attributed to core shielding which is substantial because valence electrons penetrate deeply into the core.  

Transition rates \( A \) (s\(^{-1}\)), oscillator strengths \( f \), and wavelengths \( \lambda \) (Å) for dipole transitions between low-lying states of Ce IV and Pr V are given in Table III.

These data are calculated using the dipole matrix elements \( Z^{(3)} \) from Table II and predicted NIST transition energies \( E_n \). In the two final columns of Table II, we compare our MBPT wavelengths with the wavelengths from Ref. 15. We also compare our MBPT oscillator strengths with theoretical oscillator strengths obtained by Migdalek and Wyrozumska 17. The data in 17 were obtained using a relativistic model potential (RMP) approach together with a core-polarization (CP) model potential. Our data and that from Ref. 17 agree well for \( 5d - 6p \) and \( 6s - 6p \) transitions but differ for \( 4f - 5d \) transition where \( f \) values are very small.

IV. EDM ENHANCEMENT

A. Basic equations

According to Schiff’s theorem 14, the electric dipole moment of an atom induced by an intrinsic electron EDM vanishes in the nonrelativistic limit; however, as shown by Sandars 19, the atomic EDM is nonvanishing relativistically and can be a large multiple of the intrinsic electron moment for heavy atoms. If we assume that the electron has an intrinsic EDM \( d_e \), then the EDM of a many-electric atom \( D \) may be written 14

\[
D = 2 \sum_n \langle v_{j_n} | e Z | n \rangle \langle n | H_{edm} | v_{j_n} \rangle, 
\]

where \( eZ \) is the dipole electric operator

\[
eZ = \sum_i eZ_i, 
\]

and \( H_{edm} \) is an equivalent EDM interaction 14 given by

\[
H_{edm} = -2i \frac{d_e}{e} \sum_j p_j^2 \beta_j (\gamma_3)_{j} = H_{edm}^I. 
\]

This equivalent interaction, which automatically accounts for Schiff’s theorem, is rotationally invariant and therefore conserves angular momentum; it violates both parity and time-reversal symmetry.

For an atom or ion with one valence electron, one-electron matrix elements of \( H_{edm} \) may be written in lowest order as

\[
\langle nm_n| H_{edm} | v_{j_o} \rangle = \delta_{n_o - n} \delta_{m_o,j_o} \langle n | H_{edm} | v \rangle, 
\]

where the (somewhat unconventional) reduced matrix el-
The MBPT then reduces to with “dressed” RPA matrix elements.

In the above equation, $G_k(r)$ and $F_k(r)$ are the large and small components, respectively, of radial Dirac wave functions. Similarly, we may write

$$\langle v|eZ|n_j\rangle = \sqrt{\frac{j_v}{(2j_v+1)(j_v+1)}} \langle v||eZ||n\rangle$$

with

$$\langle v||eZ||n\rangle = e \langle \kappa_v|C_1| - \kappa_v \rangle \times \int_0^\infty r dr \{G_v(r)G_n(r) + F_v(r)F_n(r)\},$$

where $C_{1q}(\hat{r})$ being a normalized spherical harmonic. The expression for the atomic dipole moment in lowest-order MBPT then reduces to

$$D^{(1)} = 2 \sqrt{\frac{j_v}{(2j_v+1)(j_v+1)}} \sum_i \langle v||eZ||i\rangle \langle i||H_{edm}||v\rangle, \quad \epsilon_v - \epsilon_i$$

where $\epsilon_k$ are eigenvalues of the valence-electron Dirac equation.

### B. RPA correlation corrections

Lowest-order calculations of the induced atomic EDM are carried out in a frozen-core $V^{N-1}$ DHF potential. Such calculations were shown in [14] to be very sensitive to correlation corrections. For that reason, the lowest-order “bare” matrix elements in Eq. (4) are replaced by “dressed” RPA matrix elements.

#### 1. Z-RPA

Thus, we replace the lowest-order dipole matrix element $\langle w||Z||v\rangle$ in Eq. (3) by

$$\langle w||Z_{RPA}||v\rangle = \langle w||Z||v\rangle$$

$$+ \sum_{a,n} (-1)^{a-n+1} \frac{1}{3} \langle a||Z_{RPA}||n\rangle Z_1(wvna)$$

$$+ \sum_{a,n} (-1)^{a-n+1} \frac{1}{3} Z_1(wvna) \langle n||Z_{RPA}||a\rangle,$$

where the index $a$ extends over all core orbitals and the index $n$ extends over all virtual orbitals permitted by angular-momentum selection rules. The quantities $Z_J(ijkl)$ are Coulomb integrals.

$$Z_J(ijkl) = X_J(ijkl) + [J] \sum_L \left\{ \frac{i k J}{l j L} \right\} X_L(ijkl),$$

where $X_J(ijkl)$ are defined by

$$X_J(ijkl) = (-1)^J \langle i||C_J||k\rangle \langle j||C_J||l\rangle R_J(ijkl),$$

$R_J(ijkl)$ being a Slater integral

$$R_J(ijkl) = \int_0^\infty \int_0^\infty drdr' \frac{j_l}{j_l+1} [G_i(r)G_k(r) + F_i(r)F_k(r)]$$

We designate the corresponding approximation to the atomic EDM by $D_Z^{(2)}$. Note that if we replace $\langle n||Z_{RPA}||a\rangle$ by $\langle n||Z||a\rangle$ on the right hand side of Eq. (3), then we obtain the second-order correlation correction to the valence-excited dipole matrix element. The atomic EDM calculated in this approximation is designated by $D_Z^{(2)}$.

#### 2. H-RPA

Similarly, we replace the bare matrix element of the EDM interaction $\langle w||H_{edm}||v\rangle$ by its dressed counterpart

$$\langle w||H_{edm}^{RPA}||v\rangle = \langle w||H_{edm}||v\rangle$$

$$+ \sum_{a,n} \sqrt{\frac{j_a}{j_v}} \langle a||H_{edm}^{RPA}||n\rangle Z_0(wvna)$$

$$+ \sum_{a,n} \sqrt{\frac{j_a}{j_v}} Z_0(wvna) \langle n||H_{edm}^{RPA}||a\rangle,$$

where the index $a$ extends over all core orbitals and the index $n$ extends over all virtual orbitals permitted by angular-momentum selection rules. The quantities $Z_J(ijkl)$ are Coulomb integrals.

$$Z_J(ijkl) = X_J(ijkl) + [J] \sum_L \left\{ \frac{i k J}{l j L} \right\} X_L(ijkl),$$

where $X_J(ijkl)$ are defined by

$$X_J(ijkl) = (-1)^J \langle i||C_J||k\rangle \langle j||C_J||l\rangle R_J(ijkl),$$

$R_J(ijkl)$ being a Slater integral

$$R_J(ijkl) = \int_0^\infty \int_0^\infty drdr' \frac{j_l}{j_l+1} [G_i(r)G_k(r) + F_i(r)F_k(r)]$$

We designate the corresponding approximation to the atomic EDM by $D_H^{(2)}$. Again, if we replace dressed matrix elements by bare matrix elements on the right hand side of Eq. (3), then we obtain a second-order approximation to $\langle w||H_{edm}||v\rangle$. The resulting correction to the atomic EDM is designated by $D_H^{(2)}$.

Core-excited matrix elements $\langle n||Z_{RPA}||a\rangle$ and $\langle n||H_{edm}^{RPA}||a\rangle$ in Eqs. (3) and (5) satisfy sets of coupled equations given explicitly in [23].

### C. Calculations of EDM enhancement factors

The sums over intermediate states in Eqs. (2) and (5) are carried out using basis functions obtained as linear combinations of B-splines as described in [21]. We use 40 splines of order 7 and constrain the ions to lie in a cavity of radius $R = 35$ a.u. for Ce$^{+3}$ and 30 a.u. for Pr$^{+4}$.

A detailed breakdown of the contributions to $D$ for 4f states of Ce IV is given in Table [15] where we list the
TABLE IV: Comparison of first-order, second-order, and RPA calculations of the atomic EDM enhancement factor $D/d_e$ for $4f$ states of Ce IV.

| State | $D^{(1)}$ | $\Delta D^{(2)}_1$ | $\Delta D^{(2)}_2$ | $D_{RPA}^{(1)}$ | $D_{RPA}^{(2)}$ | $D_{RPA}$ |
|-------|-----------|-------------------|-------------------|-----------------|-----------------|--------|
| 4f$_{5/2}$ | -0.382 | -0.383 | 0.332 | -0.438 | -0.785 | 0.387 | -0.780 |
| 4f$_{7/2}$ | -0.0026 | -0.033 | 0.022 | -0.013 | -0.045 | 0.015 | -0.032 |

TABLE V: EDM enhancement factors $D/d_e$ for low-lying states of Ce IV and Pr V.

| State | $D^{(1)}$ | $\Delta D_{RPA}^{(1)}$ | $\Delta D_{RPA}^{(2)}$ | $D_{RPA}$ |
|-------|-----------|-------------------|-------------------|--------|
| Ce IV | 4f$_{5/2}$ | -0.382 | -0.785 | 0.387 | -0.780 |
| | 4f$_{7/2}$ | -0.00225 | -0.0451 | 0.0151 | -0.0323 |
| | 5d$_{3/2}$ | -1.95 | -3.38 | 0.779 | -4.55 |
| | 5d$_{5/2}$ | 0.425 | -0.347 | -0.136 | -0.0628 |
| | 6s$_{1/2}$ | 120 | 27.5 | -19.8 | 128 |
| | 6p$_{1/2}$ | -158 | -30.9 | 19.8 | -169 |
| | 6p$_{3/2}$ | 2.89 | 7.24 | -1.07 | 9.06 |
| Pr V | 4f$_{5/2}$ | -0.142 | -0.0806 | 0.0926 | -0.130 |
| | 4f$_{7/2}$ | -0.00266 | -0.00444 | 0.000912 | -0.00802 |
| | 5d$_{3/2}$ | -1.80 | -2.91 | 0.655 | -4.05 |
| | 5d$_{5/2}$ | 0.174 | -0.827 | 0.125 | -0.528 |
| | 6s$_{1/2}$ | 127 | 27.3 | -22.2 | 132 |
| | 6p$_{1/2}$ | -157 | -30.0 | 21.7 | -166 |
| | 6p$_{3/2}$ | 2.78 | 6.06 | -1.01 | 7.83 |

DHF approximation, $D^{(1)}$, the second-order correction $D^{(2)}$, the RPA approximation, $D_{RPA}^{(1)}$, and the individual contributions to the second-order and RPA corrections from the dipole and weak-interaction matrix elements. One can see from the table that the correlation corrections to the weak-interaction matrix element are comparable to or larger than the lowest order matrix element. Moreover, there are significant changes in these correlation corrections going from second-order MBPT to full RPA calculations.

Finally, in Table V, we present DHF and RPA values of the EDM enhancement factors $D/d_e$ for the low-lying 4f, 5d, 6s, and 6p states of Ce IV and Pr V.

V. CONCLUSION

We have studied energies, transition probabilities, and EDM enhancement factors for Ce$^{+3}$ and Pr$^{+4}$. We found that perturbation theory converges quite slowly and that RPA corrections are the dominant correlation corrections for transitions. We use our third-order MBPT with “dressed” matrix elements to obtain accurate transition rates. The most interesting discovery is that RPA corrections modify lowest-order values of the EDM enhancement factor significantly.

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