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Double-core-polarization contribution to atomic parity-nonconservation and electric-dipole-moment calculations

B. M. Roberts, V. A. Dzuba, and V. V. Flambaum

School of Physics, University of New South Wales, Sydney, New South Wales 2052, Australia

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We present a detailed study of the effect of double core polarization (the polarization of core electrons due to the simultaneous action of the electric dipole and parity-violating weak fields) for amplitudes of the *ss* and *sd* parity-nonconserving transitions in Rb, Cs, Ba^+, La^{2+}, Tl, Fr, Ra^+, Ac^{2+}, and Th^{3+} as well as electron electric-dipole-moment enhancement factors for the ground states of the above neutral atoms and Au. This effect is quite large and has the potential to resolve some disagreement between calculations in the literature. It also has significant consequences for the use of experimental data in the accuracy analysis.

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I. INTRODUCTION

Measurements of parity nonconservation (PNC) and atomic electric dipole moments (EDMs) provide important tests of the electroweak theory (see, e.g., the reviews [1,2]). The PNC amplitude of the 6*s*-7*s* transition in cesium is the most precise low-energy test of the Standard Model to date. This precision is a result of highly accurate measurements [3] as well as almost equally accurate atomic calculations [4,5], which are needed for their interpretation (see also [6–8]).

For calculations of PNC in Cs, there is very good agreement between calculations, and the high accuracy is widely accepted. For other systems, however, there is disagreement between various calculations—sometimes by as much as 5%. Due to the potential significance of these calculations for probing physics beyond the Standard Model, it is very important that this disagreement be resolved.

In addition to the well-known experiments for Cs, PNC measurements are under consideration for the Ba^+ ion [9] and are in progress for the Ra^+ ion [10]. The FrPNC collaboration has begun the construction of a laser cooling and trapping apparatus with the purpose of measuring atomic parity nonconservation in microwave and optical transitions of francium [11]. There are also experiments underway at the Cyclotron and Radioisotope Centre (CYRIC) at Tohoku University to use Fr in an electron EDM measurement [12]. For more current and prospective EDM experiments, see, e.g., [1,2].

A reliable interpretation of all these measurements requires accurate atomic calculations. In this paper, we consider a particular aspect of atomic calculations which has received little attention in previous publications. This is the effect of double core polarization, which arises due to the simultaneous action of the electric dipole ($E1$) and parity-violating weak fields. The polarization of the atomic core by the electric field of the laser is affected by the presence of the weak interaction and vice versa. This leads to an additional contribution to the PNC amplitude or atomic EDM, which varies significantly between different atoms and transitions. It is 0.26% for the 6*s*-7*s* PNC transition in Cs but significantly larger for the *sd* PNC transitions—reaching 6% for the $6s$-$5d_{3/2}$ PNC transition in Ba^+. A special case is the thallium atom. If thallium is treated as a monovalence system, then the double-core-polarization contribution is about 40% for the $6p_{1/2}$-$6p_{3/2}$ PNC amplitude and about 60% for the EDM induced in the $6p_{1/2}$ ground state.

The importance of the double-core-polarization contribution is known and is included in many PNC and EDM calculations (see, e.g., [13,14]). However, it was never studied in detail and its importance was never properly emphasized. Perhaps for this reason it may be that some calculations based on the sum-over-states approach have missed this contribution.

In this work, we study the effect of double core polarization for the amplitudes of the *ss* and *sd* parity nonconserving transitions in Rb, Cs, Ba^+, La^{2+}, Tl, Fr, Ra^+, Ac^{2+}, and Th^{3+} as well as electron EDM enhancement factors for the ground states of the above neutral atoms and Au. We show that the effect is large and in some cases can explain the discrepancy between different calculations. We also show that this contribution affects the analysis of the accuracy of the calculations based on the use of experimental data.

II. CALCULATIONS

A. PNC and EDM amplitudes

The PNC amplitude, E_{PNC}, of a transition between states of the same parity can be expressed via the sum over all possible intermediate opposite parity states n,

$$E_{\mathrm{PNC}} = \sum_n \left[\frac{\langle b|\hat{d}_{E1}|n\rangle\langle n|\hat{h}_{\mathrm{PNC}}|a\rangle}{\varepsilon_a - \varepsilon_n} + \frac{\langle b|\hat{h}_{\mathrm{PNC}}|n\rangle\langle n|\hat{d}_{E1}|a\rangle}{\varepsilon_b - \varepsilon_n} \right], \quad (1)$$

where a, b, and n are many-electron wave functions of the atom with corresponding energies ε, $\hat{d}_{E1}$ is the electric dipole transition operator, and $\hat{h}_{\mathrm{PNC}}$ is the operator of the weak interaction.

Likewise, the contribution to an atomic EDM induced in the atomic state a by a mixing of opposite parity states n has the form

$$d_{\mathrm{atom}} = 2\sum_n \frac{\langle a|\hat{d}_{E1}|n\rangle\langle n|\hat{h}_{PT}|a\rangle}{\varepsilon_a - \varepsilon_n}, \quad (2)$$

where $\hat{h}_{PT}$ is the P- and T-odd operator that depends on the electron EDM and mixes states of opposite parity.

The amplitudes can then be evaluated via a direct summation of products of matrix elements and energy denominators over the states n. We refer to this method as the direct-summation (DS) method. However, we bypass this technique in favor of a more numerically stable approach based on solving differential equations, the so-called solving-equations (SE) method, or the mixed-states method. This approach, which is outlined in the next section, has many important advantages, not least of which is that it allows the easy inclusion of the important double-core-polarization (DCP) contribution.

B. Atomic structure calculations and core polarization

The above "exact" expressions (1) and (2) can be reduced to approximate formulas containing instead single-electron energies and matrix elements. Then many-body effects are included by modifying the single-electron orbitals and the external field operators. In this section, we use units with $\hbar = 1$.

We begin with the relativistic Hartree-Fock approximation and proceed to include the dominating electron correlation effects using the correlation potential method [7]. The correlation potential is used to construct the so-called Brueckner orbitals (BOs) for the valence electron, which are found by solving the Hartree-Fock-like equations with the extra operator $\hat{\Sigma}$:

$$(\hat{H}_0 + \hat{\Sigma} - E_n)\psi_n^{(\mathrm{BO})} = 0, \tag{3}$$

where $\hat{H}_0$ is the relativistic Hartree-Fock Hamiltonian and the index n denotes valence states. The BO $\psi_n^{(\mathrm{BO})}$ and energy E_n include correlations.

Interactions with the external fields are included via the time-dependent Hartree-Fock (TDHF) approximation (see, e.g., [7,15]). The external fields in question are the electric dipole ($E1$) interaction with the electric field of the photon, d_{E1}, and either the nuclear-spin–independent weak interaction h_{PNC}, or the P- and T-odd weak interaction h_{PT} in the case of atomic EDMs. It is with this method that we also include the important core-polarization effects, which arise from the action of the external fields on the Hartree-Fock V^{N-1} core potential.

Within the framework of the TDHF method, the single-electron wave function in external weak and $E1$ fields is expressed as

$$\psi = \psi_0 + \delta\psi + Xe^{-i\omega t} + Ye^{i\omega t} + \delta Xe^{-i\omega t} + \delta Ye^{i\omega t}, \tag{4}$$

where ψ_0 is the unperturbed state, $\delta\psi$ is the correction due to the weak interaction acting alone, X and Y are corrections due to the photon field acting alone, δX and δY are corrections due to both fields acting simultaneously, and ω is the frequency of the PNC transition. Since the EDM amplitude is a diagonal matrix element with no transition, $\omega = 0$ in the EDM case. This method is equivalent to the well-known random phase approximation (RPA).

The corrections δV to the core potential are found by solving the following system of RPA equations self-consistently for the core states.

The equations for the $E1$ core polarization,

$$\begin{aligned}(\hat{H}_0 - E_c - \omega)X_c &= -(\hat{d}_{E1} + \delta\hat{V}_{E1})\psi_{0c},\\ (\hat{H}_0 - E_c + \omega)Y_c &= -(\hat{d}^{\dagger}_{E1} + \delta\hat{V}^{\dagger}_{E1})\psi_{0c},\end{aligned} \tag{5}$$

and for the weak core polarization,

$$(\hat{H}_0 - E_c)\delta\psi_c = -(\hat{h}_f + \delta\hat{V}_f)\psi_{0c}, \tag{6}$$

are independent and can be solved separately. Here, the index c denotes core states, $\hat{h}_f$ is the operator of the external weak field, and $\delta\hat{V}_f$ and $\delta\hat{V}_{E1}$ are corrections to the core potential arising from the weak and $E1$ interactions, respectively. Again, ω is the energy of the PNC transition, and is zero in the case of EDMs.

There is also the set of equations corresponding to the double core polarization:

$$\begin{aligned}(\hat{H}_0 - E_c - \omega)\delta X_c &= -\delta\hat{V}_{E1}\delta\psi_c - \delta\hat{V}_f X_c\\ &\quad -\delta\hat{V}_{fE1}\psi_{0c} + \delta E_c\psi_{0c},\\ (\hat{H}_0 - E_c + \omega)\delta Y_c &= -\delta\hat{V}^{\dagger}_{E1}\delta\psi_c - \delta\hat{V}_f Y_c\\ &\quad -\delta\hat{V}^{\dagger}_{fE1}\psi_{0c} + \delta E_c\psi_{0c}.\end{aligned} \tag{7}$$

Here, $\delta\hat{V}_{fE1}$ is the correction to the core potential arising from the simultaneous perturbation of the weak field and the electric field of the laser light, and δE_c is the corresponding correction to the core energy. The correction to the core energy is zero in the case of PNC (since the matrix elements of the weak PNC interaction are imaginary and cannot produce a shift in the energy, even in an electric field), but nonzero for EDMs.

Equations (7) depend on the solutions to Eqs. (5) and (6), and must therefore be iterated after (5) and (6) are solved. In the solving-equations method, the PNC amplitude between valence states a and b is then given by

$$\begin{aligned}E_{\mathrm{PNC}} &= \langle\psi_b|\hat{d}_{E1} + \delta\hat{V}_{E1}|\delta\psi_a\rangle\\ &\quad + \langle\psi_b|\hat{h}_{\mathrm{PNC}} + \delta\hat{V}_W|X_a\rangle + \langle\psi_b|\delta\hat{V}_{fE1}|\psi_a\rangle\\ &= \langle\psi_b|\hat{d}_{E1} + \delta\hat{V}_{E1}|\delta\psi_a\rangle\\ &\quad + \langle\delta\psi_b|\hat{d}_{E1} + \delta\hat{V}_{E1}|\psi_a\rangle + \langle\psi_b|\delta\hat{V}_{fE1}|\psi_a\rangle,\end{aligned} \tag{8}$$

and the corresponding atomic EDM is given by

$$d_{\mathrm{atom}} = 2\langle\psi_a|\hat{d}_{E1} + \delta\hat{V}_{E1}|\delta\psi_a\rangle + \langle\psi_a|\delta\hat{V}_{fE1}|\psi_a\rangle. \tag{9}$$

By using BOs for the valence states ψ_a and ψ_b in (8) and (9), we can include correlations in the calculation of the PNC and EDM amplitudes. The corrections $\delta\psi_a$ and $\delta\psi_b$ to the BOs ψ_a and ψ_b are also found with the use of the correlation potential $\hat{\Sigma}$:

$$(\hat{H}_0 - E_a + \hat{\Sigma})\delta\psi_a = -(\hat{h}_f + \delta\hat{V}_f)\psi_{0a}. \tag{10}$$

The last term in Eqs. (8) and (9) represents the DCP contribution, which is due to the simultaneous action of the two external fields. This term gives an important correction that is often not included in sum-over-states calculations.

It is possible to include a term for the DCP perturbatively directly after solving Eqs. (5) and (6) and without iterating Eqs. (7). For example, by solving Eqs. (7) once without iterating, or by adding the term as a many-body perturbation theory (MBPT) diagram that corresponds to a double core excitation. This contribution corresponds to the lowest-order DCP term, which we refer to as $\delta V_{fE1}^{\mathrm{pert}}$. There is, however, another contribution that comes from further iterations of the pair of Eqs. (7). This effect, which we refer to as the relaxation effect, $\delta V_{fE1}^{\mathrm{relax}}$, has a significant impact on the value of the double core polarization. The relative size of this relaxation effect means it is not enough to simply include the term perturbatively, and the total DCP term must be taken as $\delta V_{fE1} = \delta V_{fE1}^{\mathrm{pert}} + \delta V_{fE1}^{\mathrm{relax}}$.

In these calculations, we did not include corrections such as structure radiation (the correction to the correlation potential Σ due to the $E1$ field, $\delta\Sigma_{E1}$, the weak correlation potential, $\delta\Sigma_W$, and the combined weak and $E1$ fields, $\delta\Sigma_{WE1}$), or other higher-order corrections such as ladder diagrams, and renormalization of states. These corrections are typically small (with perhaps the exception of thallium when treated as a single valence system), although they should be taken into account for accurate calculations.

III. RESULTS AND DISCUSSION

A. PNC amplitudes

We have performed calculations of the double-core-polarization correction to many PNC amplitudes, the results of which are presented in Table I along with several existing PNC calculations for comparison. We present the contributions of the double core polarization that stem from including the term perturbatively, $\delta V_{fE1}^{\text{pert}}$, and the subsequent relaxation effect, arising from further iterations of (7), $\delta V_{fE1}^{\text{relax}}$, separately.

Our results show that the double-core-polarization term is quite large, especially for the *sd* PNC transitions, and also that the relaxation effect is not small and must be included along with the perturbative lower-order term. We also demonstrate that in these cases the majority of the discrepancy between the solving equations (SE) and direct sum-over-states (DS) calculations can be explained by the possible omission of the DCP term.

In Ref. [18], calculations of *sd* PNC transitions were performed for Cs, Ba$^+$, Fr, and Ra$^+$ using both the solving equations and the direct-summation methods. As discussed, the double-core-polarization contribution was included in [18] in the SE calculations only. In that work, there was about a 4% discrepancy between the DS and SE calculations for Cs and Fr, 8% for Ba$^+$, and 7% for Ra$^+$. Here, we calculate the contribution of the double core polarization for these *sd* transitions to be approximately 3% for Cs and Fr, 6% for Ba$^+$, and 5% Ra$^+$—consistently making up for most of the disagreement. The rest of the difference likely comes from the numerical accuracy of the different methods and minor differences in correlation calculations. If the double-core-polarization contribution is removed from the SE calculations, then our SE and DS calculations match perfectly for Ba$^+$ and Fr, and are within 1% for the Cs and Ra$^+$ values from [18].

The *sd* transition in Ra$^+$ is a particularly useful case to study as there are a number of values available for comparison. The total DCP contribution is about −5% (see Table I), which is very close to the difference between the most complete calculations of Ref. [17] and all calculations using the DS approach where this contribution may be missing. The range of values that do not include the double-core-polarization term, including the DS values from Ref. [18], lie within 1% of each other. They also lie within 1% of the value obtained by removing the DCP contribution from the result of Ref. [17].

Another value, calculated by Wansbeek *et al.* [24] using a relativistic coupled-cluster (CC) approach, also agrees with these values, lying within 0.3% of the value calculated in this work without double core polarization and 0.2% of the Pal *et al.* [23] DS value. We are not sure if the DCP contribution was included in Refs. [23,24].

The difference between the 6*s*-7*s* PNC transitions in Cs for the solving-equations value 0.9041(45) of Ref. [6] and the sum-over-states value 0.8906(24) of Ref. [5] is larger than the DCP term—it is mainly due to missed contributions to the core and tail parts of the summation in (1) (see [4] for full detail). It is worth noting, however, that the double-core-polarization contribution of 0.26% is of the same size as the uncertainty quoted in [5] of 0.27%—meaning that this uncertainty can only be claimed if the DCP contribution is included. As we shall discuss in the next section, the double-core-polarization contribution has a particular impact on the accuracy analysis.

We have performed detailed PNC calculations for these Fr- and Cs-like ions in our recent paper, Ref. [17]. A more complete analysis of the accuracy of these calculations, including calculations of energy levels, lifetimes, and matrix elements, is given in that work.

B. Atomic EDM

In addition to parity nonconservation, calculations for several atomic EDMs induced by the dipole moment of the electron (d_e) have been performed. These calculations, along with several existing calculations for comparison, are presented in Table II.

Our previous calculations of the EDM for Cs [28], as well as Fr and Au [29], do not include the double-core-polarization term. These values, along with one for Rb calculated in this work, are presented in the $d_{\text{atom}}^{(0)}$ column of Table II. They are then corrected by adding the DCP term with the corrected results given in the column $d_{\text{atom}}^{\text{new}}$. We find here also that the double-core-polarization term is quite a large contribution, and that by including this term we can improve the agreement between our previous values and several other calculations.

It is interesting to note that if we include only the perturbative DCP term into the EDM calculations for Cs and Fr and do not include the relaxation term, we reproduce the values from Refs. [25] and [30] almost exactly (see Table II).

The thallium atom represents an interesting case for both PNC and EDM calculations. If we treat Tl as a monovalence system, then the DCP contribution to the PNC amplitude is huge. It contributes 36% to the PNC amplitude of the $6p_{1/2}$-$6p_{3/2}$ transition and about 60% to the EDM of the ground state. The DCP contribution is strongly dominated by the 6*s* electrons. This reflects the well-known fact that the correlations between the three outermost electrons in thallium are strong and should be treated accurately. In our view, the best approach is to treat thallium as a triple-valence-electron system and to use the configuration interaction (CI) technique combined with many-body perturbation theory (MBPT) for including valence electrons core interactions. However, good results can be obtained in other approaches too if correlations between 6*s* and 6*p* electrons, including the DCP contribution, are treated accurately. In our early calculations of the PNC in thallium [13], it was treated as a monovalence system and the DCP contribution was included. Recent calculations of the EDM enhancement factor [28,31] used the CI approach; the calculations of the Tl EDM based on the coupled-cluster approach [14,32] seems to include the DCP contribution too

TABLE I. Double-core-polarization contribution to parity nonconservation amplitudes for transitions in several atoms and ions. We present several of the most complete calculations, and what their value would be if the DCP term was omitted ($E_{\mathrm{PNC}}^{-\delta V_{fE1}}$). Shown separately are the lowest-order perturbative DCP term, $\delta V_{fE1}^{\mathrm{pert}}$, and the relaxation contribution that comes from iterations of Eqs. (7), $\delta V_{fE1}^{\mathrm{relax}}$. Also shown are several available calculations and the methods they used for comparison. SE refers to the solving-equations (or mixed-states) method, which typically includes the DCP term, and DS is the direct-summation method, which may not. Amplitudes are presented in units of $iea_B(-Q_W/N)\times 10^{-11}$.

E_{PNC}				ΔE_{PNC}(DCP)—This work				Other values			
Transition		Most complete		$\delta V_{fE1}^{\mathrm{pert}}$	$\delta V_{fE1}^{\mathrm{relax}}$	% $\delta V_{fE1}^{\mathrm{total}}$	$E_{\mathrm{PNC}}^{-\delta V_{fE1}}$	E_{PNC}		Method	
^{85}Rb	5*s*-6*s*	0.1390(7)	[16]	−0.0004	0.0001	−0.24%	0.1393	0.139(2)	[13]	SE	MBPT
	5*s*-4$d_{3/2}$	−0.450	[a]	0.0065	0.0021	−2.0%	−0.459				
^{133}Cs	6*s*-7*s*	0.9041(45)	[6]	−0.0034	0.0010	−0.26%	0.907	0.8977(40)	[4]	DS	MBPT
								0.8906(24)	[5]	DS	CC
	6*s*-5$d_{3/2}$	−3.70(4)	[17]	0.070	0.030	−2.6%	−3.80	−3.76(7)	[18]	DS	MBPT
								−3.62(7)	[18]	SE	MBPT
^{137}Ba$^+$	6*s*-7*s*	0.658(7)	[17]	−0.007	0.001	−0.84%	0.664				
	6*s*-5$d_{3/2}$	−2.20(2)	[17]	0.073	0.067	−6.0%	−2.34	−2.34(9)	[18]	DS	MBPT
								−2.17(9)	[18]	SE	MBPT
								−2.46(2)	[19]	DS	CC
^{139}La^{2+}	6*s*-5$d_{3/2}$	−2.14(2)	[17]	0.051	0.085	−6.0%	−2.28				
^{223}Fr	7*s*-8*s*	15.49(16)	[20]	−0.05	0.05	−0.06%	15.5	15.41	[21]	DS	CC
								15.9(2)	[22]	SE	MBPT
	7*s*-6$d_{3/2}$	−58.0(6)	[17]	1.12	0.40	−2.6%	−59.5	−59.5(24)	[18]	DS	MBPT
								−57.1(23)	[18]	SE	MBPT
^{226}Ra$^+$	7*s*-8*s*	10.9(1)	[17]	−0.10	0.07	−0.28%	10.9				
	7*s*-6$d_{3/2}$	−44.3(4)	[17]	1.29	0.92	−4.8%	−46.5	−45.89	[23]	DS	CC
								−46.4(14)	[24]	DS	CC
								−45.9(19)	[18]	DS	MBPT
								−43.9(18)[b]	[18]	SE	MBPT
^{227}Ac^{2+}	7*s*-6$d_{3/2}$	−42.8(4)	[17]	1.01	1.21	−4.9%	−45.0				
^{232}Th^{3+}	7*s*-6$d_{3/2}$	−43.6(4)	[17]	0.75	1.44	−4.8%	−45.8				

[a] This work.

[b] Rescaled from ^{223}Ra$^+$.

by introducing the perturbed excitation operators T_1 and T_2 (see [14] for details).

IV. IMPLICATIONS TO ACCURACY ANALYSIS

Most of the accuracy analysis in the literature assumes that the PNC and EDM amplitudes can be reduced to a sum of products of matrix elements and energy denominators that are all independent. The $E1$ matrix elements and energies can then be compared with experimental values in order to judge the accuracy of the calculations. The accuracy of the weak matrix elements can similarly be judged by calculating hyperfine structure constants, since both the weak interaction and the HFS rely on the form of the wave functions on short distances. The accuracy of this analysis, however, is limited by the value of the double-core-polarization effect, which is

TABLE II. Double-core-polarization contribution to atomic EDM calculations for several atoms including both the perturbative and relaxation parts. The values $d_{\mathrm{atom}}^{(0)}$ do not include DCP, and the values $d_{\mathrm{atom}}^{\mathrm{new}}$ do. Values are in units of d_e.

			Δd_{atom}(DCP)—This work				Other values	
State		$d_{\mathrm{atom}}^{(0)}$	$\delta V_{fE1}^{\mathrm{pert}}$	$\delta V_{fE1}^{\mathrm{relax}}$	% $\delta V_{fE1}^{\mathrm{both}}$	$d_{\mathrm{atom}}^{\mathrm{new}}$	d_{atom}	Ref.
Rb	5*s*	26.8[a]	−0.59	−0.86	−5.4%	25.4	25.74(26)	[25]
							25.7	[26]
							24.6	[27]
Cs	6*s*	124(4)[b]	−3.0	−2.5	−4.4%	119(4)	120.5(12)	[25]
							114.9	[27]
Au	6*s*	260(39)[c]	−6.7	−3.4	−3.9%	250(39)	249.9	[27]
Fr	7*s*	910(46)[c]	−24.3	−12.1	−4.0%	874(46)	894.93	[30]

[a] This work.

[b] Reference [28].

[c] Reference [29].

by no means negligible. The DCP contribution cannot easily be presented as a product of weak and electric dipole matrix elements which are independent on each other. If the analysis of accuracy ignores this contribution, it does not present the whole picture.

V. CONCLUSION

We have calculated the contribution of the double-core-polarization effect to the PNC and EDM amplitudes of several atomic systems. This is an important contribution that is of the same order as or even larger than the Breit [33], neutron-skin [34], and QED [35] corrections. This term has the potential to restore the agreement between differing calculations.

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