

Relativistic Coupled-Cluster Theory of Atomic Parity Nonconservation: Application to $^{137}\text{Ba}^+$

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We report the result of our *ab initio* calculation of the $6s^2S_{1/2} \rightarrow 5d^2D_{3/2}$ parity nonconserving electric dipole transition amplitude in $^{137}\text{Ba}^+$ based on relativistic coupled-cluster theory. Considering single, double, and partial triple excitations, we have achieved an accuracy of less than 1%. If the accuracy of our calculation can be matched by the proposed parity nonconservation experiment in Ba^+ for the above transition, then the combination of the two results would provide an independent nonaccelerator test of the standard model of particle physics.

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Parity nonconservation (PNC) in atoms arising from neutral weak currents has the potential to test the standard model (SM) of particle physics [1,2]. By combining the results of high precision measurements and calculations of atomic PNC observables, it is possible to extract the nuclear weak charge [2] and compare with its corresponding value in the SM. A discrepancy between these two values could reveal the possible existence of new physics beyond the SM. The most accurate data on atomic PNC currently comes from the $6s^2S_{1/2} \rightarrow 7s^2S_{1/2}$ transition in cesium (Cs), where the claimed experimental [3] and theoretical [4] accuracies are 0.35% and 0.5%, respectively, and the deviation from the SM is about 1σ [4]. It would indeed be desirable to consider other candidates which could yield accurate values of the nuclear weak charge. In this context an experiment to observe PNC in the $6s^2S_{1/2} \rightarrow 5d^2D_{3/2}$ transition in Ba^+ using the techniques of ion trapping and laser cooling proposed by Fortson is of special importance [5,6].

This Letter is concerned with a high precision calculation of the amplitude of the above mentioned parity nonconserving electric dipole ($E1_{\text{PNC}}$) transition in Ba^+ using relativistic coupled-cluster (RCC) theory, which is equivalent to all order relativistic many-body perturbation theory [7]. It is the first application of this theory to atomic PNC. Blundell *et al.* had used this theory in the linear approximation to calculate $E1_{\text{PNC}}$ for the $6s^2S_{1/2} \rightarrow 7s^2S_{1/2}$ transition in Cs [8]. Dzuba *et al.* [9] and Geetha [10] have calculated this PNC amplitude for the $6s^2S_{1/2} \rightarrow 5d^2D_{3/2}$ transition in Ba^+ as discussed later.

The parity nonconserving nuclear spin independent (NSI) interaction arises from the nucleon-electron neutral weak interaction and its Hamiltonian is given by

$$H_{\text{PNC}}^{\text{NSI}} = \frac{G_F}{2\sqrt{2}} Q_w \gamma_5 \rho_N(r), \quad (1)$$

where G_F is the Fermi constant, Q_w is the nuclear weak charge which is equal to $[(2Z + N)c_{1u} + (2N + Z)c_{1d}]$ with

c_{1u} and c_{1d} representing electron-up-quark and electron-down-quark coupling constants, respectively, $\rho_N(r)$ is the nuclear density function, and $\gamma_5 (= i\gamma_0\gamma_1\gamma_2\gamma_3)$, which is a pseudoscalar, is the product of the four Dirac matrices.

$H_{\text{PNC}}^{\text{NSI}}$ is responsible for mixing atomic states of opposite parities but with the same angular momentum. Its strength is sufficiently weak for it to be considered as a first-order perturbation. It is therefore possible to write the n th state atomic wave function as

$$|\Psi_n\rangle = |\Psi_n^{(0)}\rangle + G_F |\Psi_n^{(1)}\rangle. \quad (2)$$

In RCC, the atomic wave function $|\Psi_v^{(0)}\rangle$ for a single valence (v) open-shell system is given by [7,11]

$$|\Psi_v^{(0)}\rangle = e^{T^{(0)}} \{1 + S_v^{(0)}\} |\Phi_v\rangle, \quad (3)$$

where we define $|\Phi_v\rangle = a_v^\dagger |\Phi_0\rangle$, with $|\Phi_0\rangle$ as the Dirac-Fock (DF) state for closed-shell system.

In the singles and doubles approximation we have

$$T^{(0)} = T_1^{(0)} + T_2^{(0)}, \quad S_v^{(0)} = S_{1v}^{(0)} + S_{2v}^{(0)}, \quad (4)$$

where $T_1^{(0)}$ and $T_2^{(0)}$ are the single and double particle-hole excitation operators for core electrons and $S_{1v}^{(0)}$ and $S_{2v}^{(0)}$ are the single and double excitation operators for the valence electron, respectively. The amplitudes corresponding to these operators can be determined by solving the relativistic coupled-cluster singles and doubles equations. The most important triple excitations have been considered by constructing the amplitudes [12,13]

$$S_{vbc}^{pqr(0)} = \frac{V_{es} \hat{T}_2^{(0)} + V_{es} \hat{S}_{v2}^{(0)}}{\epsilon_v + \epsilon_b + \epsilon_c - \epsilon_p - \epsilon_q - \epsilon_r}, \quad (5)$$

where ϵ 's are the orbital energies. The above amplitudes are added to the open-shell coupled-cluster singles and doubles cluster amplitude equations and they are solved self-consistently. The underlying reason for adding the triples amplitudes to the open-shell and not closed-shell amplitudes equation is that the major contributions to the

properties of single valence systems come from the open-shell singles and doubles amplitudes; the contributions of the closed-shell amplitudes being rather small.

Using Eq. (2), the explicit form of $E1_{\text{PNC}}$ is given by

$$E1_{\text{PNC}} = \frac{\langle \Psi_f | D | \Psi_i \rangle}{\sqrt{\langle \Psi_f | \Psi_f \rangle \langle \Psi_i | \Psi_i \rangle}} = \frac{\langle \Psi_f^{(0)} | D | \Psi_i^{(1)} \rangle + \langle \Psi_f^{(1)} | D | \Psi_i^{(0)} \rangle}{\sqrt{\langle \Psi_f^{(0)} | \Psi_f^{(0)} \rangle \langle \Psi_i^{(0)} | \Psi_i^{(0)} \rangle}}, \quad (6)$$

where D is the electric dipole ($E1$) operator, i and f subscripts are used for initial and final valence electrons, respectively. Using the explicit expression for the first-order perturbed wave function, we get

$$E1_{\text{PNC}} = \sum_{I \neq i} \frac{\langle \Psi_f^{(0)} | D | \Psi_I^{(0)} \rangle \langle \Psi_I^{(0)} | H_{\text{PNC}}^{\text{NSI}} | \Psi_i^{(0)} \rangle}{E_i - E_I} + \sum_{I \neq f} \frac{\langle \Psi_f^{(0)} | H_{\text{PNC}}^{\text{NSI}} | \Psi_I^{(0)} \rangle \langle \Psi_I^{(0)} | D | \Psi_i^{(0)} \rangle}{E_f - E_I}, \quad (7)$$

where I represent intermediate states.

It is obvious from the above equation that the accuracy of the calculation of $E1_{\text{PNC}}$ depends on the excitation energies of the different intermediate states, the matrix

elements of $H_{\text{PNC}}^{\text{NSI}}$ and D. Blundell *et al.* have used the above equation to determine $E1_{\text{PNC}}$ for the $6s^2S_{1/2} \rightarrow 7s^2S_{1/2}$ transition in Cs by considering the most important intermediate states [8]. The drawback of this approach is that the summation can be performed only over a finite set of intermediate states which limits the accuracy of the calculation. The method we have used in the present work circumvents this problem by solving the first-order perturbed equation

$$(H^{(0)} - E^{(0)})|\Psi_v^{(1)}\rangle = (E^{(1)} - H_{\text{PNC}}^{\text{NSI}})|\Psi_v^{(0)}\rangle, \quad (8)$$

where $E^{(1)}$ vanishes for the first-order correction.

The perturbed cluster operators can be written as

$$T = T^{(0)} + G_F T^{(1)}, \quad S_v = S_v^{(0)} + G_F S_v^{(1)}, \quad (9)$$

where $T^{(1)}$ and $S_v^{(1)}$ are the first-order G_F corrections to the cluster operators $T^{(0)}$ and $S^{(0)}$, respectively. The amplitudes of these operators are solved, keeping up to linear in PNC perturbed amplitudes, by the following equations

$$\langle \Phi_a^p | \bar{H}_N^{(0)} T^{(1)} + \bar{H}_{\text{PNC}}^{\text{NSI}} | \Phi_0 \rangle = 0, \quad (10)$$

$$\langle \Phi_{ab}^{pq} | \bar{H}_N^{(0)} T^{(1)} + \bar{H}_{\text{PNC}}^{\text{NSI}} | \Phi_0 \rangle = 0,$$

and

$$\langle \Phi_v^p | \bar{H}_N^{(0)} S_v^{(1)} + (\bar{H}_N^{(0)} T^{(1)} + \bar{H}_{\text{PNC}}^{\text{NSI}})\{1 + S_v^{(0)}\} | \Phi_v \rangle = -\langle \Phi_v^p | S_v^{(1)} | \Phi_v \rangle \text{IP},$$

$$\langle \Phi_{vb}^{pq} | \bar{H}_N^{(0)} S_v^{(1)} + (\bar{H}_N^{(0)} T^{(1)} + \bar{H}_{\text{PNC}}^{\text{NSI}})\{1 + S_v^{(0)}\} | \Phi_v \rangle = -\langle \Phi_{vb}^{pq} | S_v^{(1)} | \Phi_v \rangle \text{IP}, \quad (11)$$

where $H^{(0)}$ is the Dirac-Coulomb (DC) Hamiltonian and \bar{H} is defined as $e^{-T^{(0)}} H e^{T^{(0)}}$, which is computed after determining $T^{(0)}$, IP is the ionization potential corresponding to the valence electron “ v ”, and the subscript N denotes normal form of an operator. We have used $a, b \dots$ and $p, q \dots$ etc. to represent holes and particles, respectively. $|\Phi_v^p\rangle$ and $|\Phi_{vb}^{pq}\rangle$ are the single and double excited states, respectively, with respect to $|\Phi_v\rangle$. Using Eqs. (3), (6), and (9) and only keeping terms linear in G_F , the expression for $E1_{\text{PNC}}$ can be written as

$$E1_{\text{PNC}} = \frac{\langle \Phi_f | \{1 + S_f^{(1)\dagger} + T^{(1)\dagger} S_f^{(0)\dagger} + T^{(1)\dagger}\} e^{T^{(0)\dagger}} D e^{T^{(0)}} \{1 + T^{(1)} + T^{(1)} S_i^{(0)} + S_i^{(1)}\} | \Phi_i \rangle}{\sqrt{(1 + N_f^{(0)})(1 + N_i^{(0)})}} = \frac{\langle \Phi_f | S_f^{(1)\dagger} \bar{D}^{(0)}(1 + S_i^{(0)}) + (1 + S_f^{(0)\dagger}) \bar{D}^{(0)} S_i^{(1)} + S_f^{(0)\dagger} (T^{(1)\dagger} \bar{D}^{(0)} + \bar{D}^{(0)} T^{(1)}) S_i^{(0)} + (T^{(1)\dagger} \bar{D}^{(0)} + \bar{D}^{(0)} T^{(1)}) S_i^{(0)} | \Phi_i \rangle}{\sqrt{(1 + N_f^{(0)})(1 + N_i^{(0)})}}. \quad (12)$$

In the above expression we define $\bar{D}^{(0)} = e^{T^{(0)\dagger}} D e^{T^{(0)}}$ and $N_v^{(0)} = \langle \Phi_v | S_v^{(0)\dagger} e^{T^{(0)\dagger}} e^{T^{(0)}} S_v^{(0)} | \Phi_v \rangle$ for the valence electron v and each term is connected. The above matrix element is evaluated by a method similar to that used in our earlier works of Ba⁺ [14,15].

The orbitals are constructed as linear combinations of Gaussian type orbitals (GTOs) of the form [16]

$$F_{i,k}(r) = r^k e^{-\alpha_i r^2}, \quad (13)$$

where $k = 0, 1, \dots$ for s, p, \dots type orbital symmetries, respectively. For the exponents, we have used

$$\alpha_i = \alpha_0 \beta^{i-1}. \quad (14)$$

We have considered $30s_{1/2}$, $25p_{1/2}$, $25p_{3/2}$, $25d_{3/2}$, $25d_{5/2}$, $20f_{5/2}$, $20f_{7/2}$, $20g_{7/2}$, and $20g_{9/2}$ GTOs for the DF calculation and all occupied (active holes) orbitals in the RCC calculations. We have chosen α_0 as 0.00525 and β as 2.73 for all the symmetries. All orbitals are generated on a grid using a two-parameter Fermi nuclear distribution approximation given by

$$\rho = \frac{\rho_0}{1 + e^{(r-c)/a}}, \quad (15)$$

where ρ_0 is the average nuclear density, “ c ” is the half-charge radius, and “ a ” is related to the skin thickness.

TABLE I. Excitation energy (cm^{-1}), $E1$ transition amplitudes (a.u.) and magnetic dipole hyperfine structure constant (MHz) for different low-lying states of Ba^+ .

Initial state	$6s^2S_{1/2}$	$6s^2S_{1/2}$	$5d^2D_{3/2}$	$5d^2D_{3/2}$
\rightarrow Final state	$6p^2P_{1/2}$	$6p^2P_{3/2}$	$6p^2P_{1/2}$	$6p^2P_{3/2}$
Excitation energy	20 410	22 104	15 097	16 795
Experiment [18]	20 262	21 952	15 388	17 079
$E1$ transition amplitude	3.37	4.72	3.08	1.36
Experiment [19]	3.36(0.16)	4.67(0.08)	3.03(0.08)	1.36(0.04)
Atomic state	$6s^2S_{1/2}$	$6p^2P_{1/2}$	$6p^2P_{3/2}$	$5d^2D_{3/2}$
Hyperfine constant (A)	4078.18	740.77	128.27	189.92
Experiment [20–22]	4018.871(2)	743.7(3)	127.2(2)	189.7288(6)

These values are considered using the relations given by Parpia and Mohanty [17].

Our earlier calculations of excitation energies [13], $E1$ transition amplitudes [14], and magnetic dipole hyperfine constants [15] for some of the low-lying states in Ba^+ based on RCC theory suggest that it is in principle possible to perform a calculation of $E1_{\text{PNC}}$ for the $6s^2S_{1/2} \rightarrow 5d^2D_{3/2}$ transition in that ion to an accuracy of better than 1%. We have recalculated these quantities using the same method but with a larger basis and the results are given in Table I. The agreement with experiment of the most important excitation energy ($6p^2P_{1/2}$) for the calculation of $E1_{\text{PNC}}$ is less than 1%. This is also the case for the hyperfine constants of three of the states— $6p^2P_{1/2}$, $6p^2P_{3/2}$ and $5d^2D_{3/2}$, while for the $6s^2S_{1/2}$ state, the agreement is a little over 1%. All the transition amplitudes are within the experimental error bars. The result of our calculation of the electric quadrupole ($E2$) amplitude for the $6s^2S_{1/2} \rightarrow 5d^2D_{3/2}$ transition is 12.61 in a.u. It is in agreement with our earlier calculation [14] and well within the experimental bounds [23]. In Table II, we present the values of the square root of the product of the hyperfine constants. The accuracies of these two quantities give an indication of the accuracies of the PNC matrix elements between $6s^2S_{1/2}$ and $6p^2P_{1/2}$ states as well as $6p^2P_{3/2}$ and $5d^2D_{3/2}$ states. Both of them are in excellent agreement with experiment, suggesting that the two leading PNC matrix elements used in the $E1_{\text{PNC}}$ calculation are very accurate. The contributions from the different terms in $E1_{\text{PNC}}$ are presented in Table III. It is clear that the largest contribution comes from $DS_1^{(1)}$ [Fig. 1(iii)] which represents the DF term and a certain subclass of core polarization as well as pair correlation effects [10]. This is due to the relatively large $(6s_{1/2} - 6p_{1/2}) S_1^{(1)}$ cluster amplitude.

TABLE II. Square root of the magnetic dipole hyperfine constants (MHz) and their deviations from experimental results.

	Experiment	This work	Deviation (%)
$\sqrt{A_{6s^2S_{1/2}} A_{6p^2P_{1/2}}}$	1728.83	1738.1	0.5
$\sqrt{A_{6p^2P_{3/2}} A_{5d^2D_{3/2}}}$	155.35	156.08	0.5

TABLE III. Contributions to the $E1_{\text{PNC}}$ calculation in $\times 10^{-11} iea_0(-Q_W/N)$ using RCC calculation.

Initial perturbation	$6s^2S_{1/2} \rightarrow$	Final pert.	$6s^2S_{1/2} \rightarrow$
Terms	$5d^2D_{3/2}^{(0)}$	Terms	$5d^2D_{3/2}^{(1)}$
Dirac-Fock contribution			
$DH_{\text{PNC}}^{\text{NSI}}$	2.018	$H_{\text{PNC}}^{\text{NSI}}D$	-0.3×10^{-5}
$DT_1^{(1)}$	0.0003	$T^{(1)\dagger}D$	0.418
$\bar{D}^{(0)}S_{1i}^{(1)}$	2.634	$S_{1f}^{(1)\dagger}\bar{D}^{(0)}$	-0.179
$\bar{D}^{(0)}S_{2i}^{(1)}$	-0.242	$S_{2f}^{(1)\dagger}\bar{D}^{(0)}$	-0.166
$S_{1f}^{(0)\dagger}\bar{D}^{(0)}S_{1i}^{(1)}$	0.149	$S_{1f}^{(1)\dagger}\bar{D}^{(0)}S_{1i}^{(0)}$	0.003
$S_{1f}^{(0)\dagger}\bar{D}^{(0)}S_{2i}^{(1)}$	0.007	$S_{1f}^{(1)\dagger}\bar{D}^{(0)}S_{2i}^{(0)}$	0.008
$S_{2f}^{(0)\dagger}\bar{D}^{(0)}S_{1i}^{(1)}$	-0.116	$S_{2f}^{(1)\dagger}\bar{D}^{(0)}S_{1i}^{(0)}$	-0.009
$S_{2f}^{(0)\dagger}\bar{D}^{(0)}S_{2i}^{(1)}$	-0.001	$S_{2f}^{(1)\dagger}\bar{D}^{(0)}S_{2i}^{(0)}$	0.001
Others	-0.011		0.012
Norm.	-0.046		-0.001
Total	2.375		0.087

Two different types of core polarization effects; $DT_1^{(1)}$ [Fig. 1(i)] and $DS_2^{(1)}$ as well as its conjugate [Fig. 1(v) and (vi)] also make significant contributions. The former is mediated by the neutral weak interaction and involves the $6s$ valence and core electrons. Correlation effects corresponding to $S_{1i}^{(0)\dagger}DS_{1i}^{(1)}$ and $S_{2i}^{(0)\dagger}DS_{1i}^{(1)}$ are non-negligible, but their signs are opposite. Contributions from other terms are comparatively small.

The result of $E1_{\text{PNC}}$ for the $6s^2S_{1/2} \rightarrow 5d^2D_{3/2}$ transition in our calculation is $2.46 \times 10^{-11} iea_0(-Q_W/N)$. It is larger in magnitude than those obtained by Dzuba *et al.* [9] and Geetha [10] as shown by Table IV. The former work is based on a variant of all order many-body perturbation theory, but it has some semiempirical features. It is carried out by using two different approaches. One of them is similar to the sum-over-states approach by Blundell *et al.* [8] and the other is known as the mixed approach where the PNC interaction explicitly mixes states of opposite parities. However, both calculations do not include contributions from certain correlation effects; i.e., structural radiation, weak correlation potential, and normalization of states [9] that are included in our calculation. Their $6p^2P_{1/2} \rightarrow 5d^2D_{3/2}$ $E1$ matrix element, which is important for the above mentioned PNC transition amplitude is not as accurate as ours. Furthermore, the accuracies of their PNC matrix elements are not known as they have not performed calculations of the hyperfine constants of the relevant states. The reason for the discrepancy between our calcu-

TABLE IV. Comparison of $E1_{\text{PNC}}$ results from different calculations in $\times 10^{-11} iea_0(-Q_W/N)$.

Dzuba <i>et al.</i>	[9]	Geetha [10]	Present work
(mixed parity)	(sum-over-states)		
2.17	2.34	2.35	2.46 ± 0.02

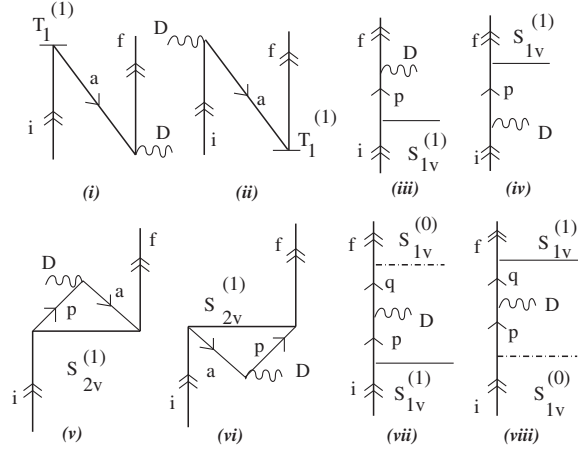


FIG. 1. Important Goldstone diagrams corresponding PNC amplitudes.

lation and Geetha's is that our approach implicitly includes several intermediate states; particularly doubly excited opposite parity states which her sum-over-states approach omits.

The error accrued in our calculation of $E1_{\text{PNC}}$ can be determined from the errors in the excitation energies, $E1$ transition amplitudes and hyperfine constants (see Table I). We have not estimated the errors in the calculated values of these quantities by comparing with measurements, since the error bars in the $E1$ transition amplitudes are rather large. Instead, we have taken the differences of our RCC calculations with single, double, and leading triple excitations and just single and double excitations as the errors. The error in $E1_{\text{PNC}}$ (0.02) has been obtained by adding the errors for the different quantities it depends on in quadrature for the leading intermediate states $6p^2P_{1/2}$ and $6p^2P_{3/2}$ and using a scale factor to estimate the errors from other intermediate states that together make a small contribution.

The contribution of the Breit interaction to $E1_{\text{PNC}}$ at the DF level is 0.1% and the nuclear structure contribution is 0.3%. The latter has been determined more accurately than Blundell *et al.* [8] using relativistic mean field theory. Our calculation of Cs $E1_{\text{PNC}}$ is $-0.902(4) \times 10^{-11} iea_0(-Q_W/N)$ using our present method has an accuracy of about 0.5% and this is a strong indication that our RCC method is capable of high precision PNC calculations for single valence systems.

In conclusion, we have performed a sub 1% calculation of $E1_{\text{PNC}}$ for the $6s^2S_{1/2} \rightarrow 5d^2D_{3/2}$ transition in Ba^+ using RCC. We have included single, double, as well as a leading class of triple excitations and highlighted the importance of various many-body effects. Given the promise that the Ba^+ PNC experiment holds out, it does indeed appear that in the future the result of that experiment combined with our calculation would constitute a new and an important probe of physics beyond the SM.

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