Role of negative-energy states and Breit interaction in calculation of atomic parity-nonconserving amplitudes.

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It is demonstrated that Breit and negative-energy state contributions reduce the 2.5σ deviation [S.C. Bennett and C.E. Wieman, Phys. Rev. Lett. 82, 2484 (1999)] in the value of the weak charge of 133Cs from the Standard Model prediction to 1.7σ. The corrections are obtained in the relativistic many-body perturbation theory by combining all-order Coulomb and second-order Breit contributions. The corrections to parity-nonconserving amplitudes amount to 0.6% in 133Cs and 1.1% in 223Fr. The relevant magnetic-dipole hyperfine structure constants are modified at the level of 0.3% in Cs, and 0.6% in Fr. Electric-dipole matrix elements are affected at 0.1% level in Cs and a few 0.1% in Fr.

Presently, the PNC effect in atoms was most precisely determined ratio of PNC amplitude \( Q_{\text{E}} \) to the tensor transition polarizability \( \beta \) for 7S\(_{1/2} \) – 6S\(_{1/2} \) transition with a precision of 0.35%. In 1999, Bennett and Wieman accurately measured tensor transition polarizability \( \beta \) and, by combining the previous theoretical determinations of the \( E_{\text{PNC}} \) with their measurements, they have found a value of the weak charge for 133Cs \( Q_{W} = -72.06(28)_{\text{exp}}(34)_{\text{theor}} \) which differed from the prediction of the Standard Model \( Q_{W} = -73.20(13) \) by 2.5 standard deviations. They also reevaluated the precision of the early 1990s atomic structure calculations [4,5], and argued that the uncertainty of the predicted \( E_{\text{PNC}} \) is 0.4%, rather than previously estimated 1%. This conclusion has been based on a much better agreement of calculated and recently accurately measured electric-dipole amplitudes for the resonant transitions in alkali-metal atoms.

In view of the reduced uncertainty, the purpose of this Letter is to evaluate contributions from negative-energy states (NES) and Breit interaction. It will be demonstrated that these contributions correct theoretical \( E_{\text{PNC}} \) and the resultant value of the weak charge by 0.6% in Cs. It is worth noting, that due to the smallness of these contributions at what had been believed to be a 1% theoretical error in Cs, the previous calculations have either omitted [3], or estimated the contributions from Breit interaction only partially [4]. The main focus of the previous \( \textit{ab initio} \) calculations has been correlation contribution from the residual Coulomb interaction (i.e. beyond Dirac-Hartree-Fock level). In both calculations important chains of many-body diagrams have been summed to all orders of perturbation theory.

This Letter also reports correction due to NES and Breit interaction for \( E_{\text{PNC}} \) in francium. The interest in Fr stems from the fact that analogous PNC amplitude is 18 times larger in heavier 223Fr compared to Cs [6]. The measurement of atomic PNC in Fr is pursued by Stony Brook group [8].

The quality of theoretical atomic wave-functions at small radii is usually judged by comparing calculated and experimental hyperfine-structure magnetic-dipole constants \( A \), and in the intermediate region by comparing electric-dipole matrix elements. It will be demonstrated that the corresponding corrections to all-order Coulomb values are at the level of a few 0.1%.

The PNC amplitude of \( nS_{1/2} \rightarrow n'S_{1/2} \) transition can be represented as a sum over intermediate states \( mP_{1/2} \)

\[
E_{\text{PNC}} = \sum_{m} \frac{\langle n'S\vert D\vert mP_{1/2}\rangle \langle mP_{1/2}\vert H_{W}\vert nS\rangle}{E_{mP_{1/2}} - E_{nS}} + \sum_{m} \frac{\langle n'S\vert H_{W}\vert mP_{1/2}\rangle \langle mP_{1/2}\vert D\vert nS\rangle}{E_{mP_{1/2}} - E_{n'S}}.
\]

The overwhelming contribution from parity-violating interactions arises from the Hamiltonian

\[
H_{W} = \frac{G_{F}}{\sqrt{8}} Q_{W} \rho_{\text{nuc}}(r) \gamma_{5},
\]

where \( G_{F} \) is the Fermi constant, \( \gamma_{5} \) is the Dirac matrix, and \( \rho_{\text{nuc}}(r) \) is the nuclear distribution. To be consistent with the previous calculations the \( \rho_{\text{nuc}}(r) \) is taken to be a Fermi distribution with the “skin depth” \( a = 2.3/(4 \ln 3) \) fm and the cutoff radius \( c = 5.6743 \) fm for 133Cs as in Ref. [3], and \( c = 6.671 \) fm for 223Fr as in Ref. [4]. The PNC amplitude is customarily expressed in the units of \( 10^{-11}v(-Q_{W}/N) \), where \( N \) is the number of neutrons in the nucleus (\( N = 78 \) for 133Cs and \( N = 136 \) for 223Fr ). Atomic units are used throughout the Letter. The results of the calculations for 133Cs are \( E_{\text{PNC}} = -0.905 \times 10^{-11}(-Q_{W}/N) \),
Ref. 4 and $E_{\text{PNC}} = -0.908 \times 10^{-11} i(-Q_W/N)$, Ref. 5. The former value includes a partial Breit contribution $+0.002 \times 10^{-11} i(-Q_W/N)$, and the latter does not. Both calculations are in a very close agreement if the Breit contribution is added to the value of Novosibirsk group. The reference many-body Coulomb value

$$E_{\text{PNC}}^C = -0.9075 \times 10^{-11} i(-Q_W/N)$$

is determined as an average of the two, with Breit contribution removed from the value of Notre Dame group. For $^{235}$Fr $E_{\text{PNC}} = 15.9 \times 10^{-11} i(-Q_W/N)$, Ref. 6, this value does not include Breit interaction.

\textit{Ab initio} relativistic many-body calculations of wavefunctions, like coupled-cluster type calculations of 7, to avoid the “continuum dissolution problem” 8, start from the \textit{no-pair} Hamiltonian derived from QED 1. The \textit{no-pair} Hamiltonian excludes virtual electron-positron pairs from the resulting wavefunction. If the \textit{no-pair} wavefunctions are further used to obtain many-body matrix elements, the negative-energy state (NES) contribution is missing already in the second order. Recently, it has been shown that the magnetic-dipole transition amplitude both in He-like ions 2 and alkali-metal atoms 9 can be strongly affected by the NES correction. The enhancement mechanism is due to vanishingly small lowest-order values and also due to mixing of large and small components of a Dirac wavefunction by magnetic-dipole operator. For the NES ($E < -mc^2$) the meaning of large and small components is reversed, i.e., small component is much larger than large component. The mixing of large positive-energy component with small component of NES results in much larger one-particle matrix elements, than in the \textit{no-pair} case. The $2mc^2$ energy denominators lessen the effect, but, for example, the Rb 5$S_{1/2} - 6S_{1/2}$ magnetic-dipole rate is reduced by a factor of 8 from the \textit{no-pair} value by the inclusion of NES 13. The inclusion of Breit interaction also becomes important and the size of the correction is comparable to the Coulomb contribution. Just as in the case of magnetic-dipole operator, the Dirac matrix $\gamma_5$ in the weak Hamiltonian Eq. 2 mixes large and small components of wavefunctions. Similar mixing occurs in the matrix element describing interaction of an electron with nuclear magnetic moment (hyperfine structure constant $A$). As demonstrated below, the relative effect for these operators is not as strong as in the magnetic-dipole transition case, since the lowest order matrix elements are nonzero in the nonrelativistic limit, but is still important. It is worth noting that the problem of NES does not appear explicitly in the Green’s function 10 or mixed-parity 11 approaches; however the correction due to Breit interaction still has to be addressed. The NES Coulomb corrections have to be taken into account explicitly in the “sum-over-states” method 1, employing all-order many-body values obtained with the \textit{no-pair} Hamiltonian.

The analysis is based on the $V_{N-1}$ Dirac-Hartree-Fock potential wavefunctions, with the valence and virtual orbitals $m$ calculated in the “frozen” potential of core orbitals $a$. The second-order correction to a matrix element of one-particle operator $Z$ between two valence states $u$ and $v$ is represented as

$$Z_{uv}^{(2)} = \sum_{i \neq v} \frac{z_{ui}}{\epsilon_u - \epsilon_i} + \sum_{i \neq w} \frac{b_{wi}}{\epsilon_w - \epsilon_i} +$$

$$+ \sum_{ma} \frac{z_{am}}{\epsilon_a + \epsilon_m - \epsilon_w} + \sum_{ma} \frac{d_{a}^{m} + \bar{b}_{awm}}{\epsilon_a + \epsilon_m - \epsilon_w}.$$

This expression takes into account the residual (two-body) Coulomb, $g_{ijkl}$, and two-body $b_{ijkl}$ and one-body $b_{ij} = \sum_{a} \tilde{b}_{iija}$ Breit interaction. Static form of Breit interaction is used in this work. The tilde denotes antisymmetric combination $\tilde{b}_{ijkl} = b_{ijkl} - b_{ikjl}$. Subscript $i$ ranges over both core and excited states. Note that summation over states $i$ and $m$ includes negative-energy states. The NES correction to PNC amplitudes arises in two circumstances, directly from the sum in Eq. 1, and in the values of electric-dipole and weak interaction matrix elements. If the length-gauge of the electric-dipole operator is used, the direct contribution of NES in the amplitude Eq. 1 is a factor of $10^{-13}$ smaller than the total amplitude, and will be disregarded in the following. The numerical summations are done using 100 positive- and 100 negative-energy wavefunctions in a B-spline representation 14 obtained in a cavity with a radius of 75 a.u.

The breakdown of second-order corrections to matrix elements of weak interaction for Cs is given in Table 1. The all-order values from Ref. 1 are also listed in the table to fix the relative phase of the contributions. The matrix elements are each modified at 0.6-0.7% level. Most of the correction arises from positive-energy Breit contribution, negative-energy states contribute at a smaller but comparable level. The contributions from NES due to one-body and two-body Breit interaction are almost equal ($B_{F}^{(3)} \approx B_{F}^{(2)}$) and, in addition, $B_{F}^{(1)} \approx 2B_{F}^{(2)}$. The same relations hold also in francium. The corrections to the relevant length-form matrix elements are overwhelmingly due to the one-body Breit interaction, and are at 0.1% level. For example, the all-order \textit{reduced} matrix element $\langle 6S_{1/2} | D | 6P_{1/2} \rangle = 4.478$ Ref. 1 is increased by 0.005, bringing the total 4.483 into an excellent agreement with experimental value 15 4.4890(65). Generally, the corrections reduce absolute values of the weak interaction matrix elements, and increase absolute values of the dipole matrix elements, therefore, their net contributions to $E_{\text{PNC}}$ have an opposite sign. Matrix elements of weak interaction are affected more strongly, because of the sampling of wave-function in the nucleus, where relativity is important.

As demonstrated in Ref. 1, the four lowest-energy valence $mP_{1/2}$ states contribute 98% of the sum in Eq. 1, and for the purposes of this work, limiting the sums to only these states is sufficient. The corrections to $E_{\text{PNC}}$
are calculated first by replacing the weak interaction matrix elements with the relevant second-order contributions and at the same time using all-order dipole matrix elements, and second by taking all-order $H_W$ matrix elements, and replacing $D$ with the appropriate correction. In both cases the experimental energies are used in the denominators. The needed all-order matrix elements for Cs are tabulated in Ref. [4]. The summary of corrections to $E_{\text{PNC}}$ is presented in Table I. The modifications in the weak interaction matrix element provide a dominant correction. The contribution due to NES in the Coulomb part is insignificant, and is already effectively included in the reference many-body Coulomb value $E_{\text{PNC}}^{C}$, Eq. (3). The reference value $E_{\text{PNC}}^{C}$ is modified by the Breit contributions by 0.6%, almost two times larger than the uncertainty in the Boulder experiment [2]. The modified Breit contribution is 0.003 variation radius

\[ Q_{W}^{(133\text{Cs})} = -72.42(28)_{\text{expt}}(34)_{\text{theor}}. \]

This value differs from the prediction of the Standard Model $Q_{W} = -73.20(13)$ by 1.7σ, versus 2.5σ discussed in Ref. [3], where σ is calculated by taking uncertainties in quadrature.  

The only previous calculation of Breit contribution to PNC amplitude in Cs has been performed by the Notre Dame group [1], using mixed-parity Dirac-Hartree-Fock formalism. The one-body Breit interaction has been included on equal footing with the DHF potential, but the linearized modification to one-body Breit potential due to $H_W$ (\(V_{\text{PNC-\text{HDF}}}\) in notation of Ref. [1]) has been omitted. It is straightforward to demonstrate that because of this omission, the comparable contribution from two-body part of the Breit interaction has been disregarded. In units of $10^{-11}i(-Q_W/N)$, the result of the present calculation for one-body Breit contribution is 0.003 versus 0.002 in Ref. [3]. Such disagreement is most probably caused by different types of correlation contribution included in the two approaches. Treating one-body Breit together with the DHF potential effectively sums the many-body contributions from one-body Breit interaction to all orders, and presents the advantage of the scheme employed in Ref. [1]. However, the dipole matrix elements and energies in the sum Eq. (3) are effectively included at the DHF level in the formulation of Ref. [1], in contrast to high-precision all-order values employed in the present work. The difference between the two values can be considered as a theoretical uncertainty in the value of the Breit correction. Clearly more work needs to be done to resolve the discrepancy. The accuracy of the present analysis can be improved if the one-body Breit interaction is embodied in DHF equations, and the many-body formulation starts from the resulting basis. However, to improve present second-order treatment of the two-body part of the Breit interaction, higher orders of perturbation theory have to be considered. Apparently the most important contribution would arise from terms linearized in the Breit interaction, i.e. diagrams containing one matrix elements of the Breit interaction and the rest of the residual Coulomb interaction.  

The Breit and NES corrections to PNC amplitude in heavier Fr are more pronounced. The $^{233}$Fr PNC amplitude $15.9 \times 10^{-11}i(-Q_W/N)$ from Ref. [3] is reduced by 1.1%. Using all-order dipole matrix elements from Ref. [3], the following corrections due to modifications in the $h_W$ are found (in units of $10^{-11}i(-Q_W/N)$): one-body Breit $B_{1}^{(1)} = -0.131$, two-body Breit $B_{2}^{(2)} = -0.53$, and the C.. contribution, implicitly included in Ref. [3], is $-0.003$. As in the case of Cs, the all-order no-pair Coulomb result [4] for reduced matrix element \(\langle 7p_{1/2}\rangle \langle D||7s_{1/2}\rangle = 4.256\times 10^{-11}\) is increased by inclusion of the Breit interaction and NES by 0.0111, a 0.3% modification, leading to a much better agreement with experimental value 4.277(8) [3]. The modification of the $E_{\text{PNC}}$ due to corrections in the dipole matrix elements is much smaller than in the case of $h_W$. At present there is no tabulation of accurate matrix elements of weak interaction for Fr, and the influence on $E_{\text{PNC}}$ due to the Breit contribution in dipole matrix elements is estimated from average of the modification of individual dipole matrix elements 0.2%. The net result decreases the reference Coulomb value for $^{233}$Fr by 0.18 $\times 10^{-11}i(-Q_W/N)$, and the corrected value is

\[ E_{\text{PNC}}^{C+B}(^{233}\text{Fr}) = 15.7 \times 10^{-11}i(-Q_W/N). \]

Finally, it is worth discussing Breit and NES contributions to hyperfine-structure magnetic-dipole constants $A$ for the states involved into PNC calculations. The all-order no-pair Coulomb values in the recent work [3] have been corrected using a similar second-order formulation; no details of the calculation have been given. The explicit contributions listed in Table I will be useful for correcting ab initio many-body Coulomb values. The table presents the contributions for two lowest valence $S_{1/2}$ and $P_{1/2}$ states. The calculations are performed using a model of uniformly magnetized nucleus with a magnetization radius $R_m$ given in the table. One finds that the additional terms reduce values calculated in the no-pair Coulomb-correlated approach. For Cs the corrections are of order 0.2% for $6S_{1/2}$, 0.1% for $7S_{1/2}$, and 0.3% for $6P_{1/2}$ and $7P_{1/2}$. The relative contributions to hyperfine constants in heavier Fr are larger, accounting for 0.5% of the total value for $7S_{1/2}$, 0.4% for $8S_{1/2}$, and 0.6% for $7P_{1/2}$ and $8P_{1/2}$.  

This work demonstrates that the Breit and NES contributions are comparable to the remainder of Coulomb correlation corrections unaccounted for in modern relativistic all-order many-body calculations and hence have
to be systematically taken into account. In particular, the Breit interaction contributes 0.6% to parity-nonconserving amplitudes in Cs and 1.1% in Fr. The correction for Cs is almost twice the experimental uncertainty and reduces the recently determined $\pm 2.5\sigma$ deviation in the value of weak charge from the Standard Model prediction to $1.7\sigma$. Both hyperfine constants and electric-dipole matrix elements are affected at a few 0.1%. By including NES and Breit correction, the no-pair Coulomb all-order dipole matrix elements [9] for resonant transitions are brought into an excellent agreement with the accurate experimental values.

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### TABLE I. Contributions to matrix elements of the weak interaction for $^{133}$Cs in units $10^{-11}i(-Q_W/N)$. All-order no-pair values are from Blundell et al. [4]. $C_-$ is the correction from negative-energy states for the residual Coulomb interaction, and $B^{(1)}_\pm$ and $B^{(2)}_\pm$ are positive/negative energy state contributions from one-body and two-body Breit interaction. Notation $x[y] = x \times 10^y$.

| n  | all-order | $C_-$  | $B^{(1)}_+$ | $B^{(1)}_-$ | $B^{(2)}_+$ | $B^{(2)}_-$ |
|----|----------|--------|-------------|-------------|-------------|-------------|
| 6  | 5.62[-2] | -9.05[-6] | -3.10[-4] | 5.64[-5] | -1.54[-4] | 5.73[-5] |
| 7  | 3.19[-2] | -5.41[-6] | -1.82[-4] | 3.37[-5] | -9.21[-5] | 3.43[-5] |
| 8  | 2.15[-2] | -3.71[-6] | -1.23[-4] | 2.31[-5] | -6.32[-5] | 2.35[-5] |
| 9  | 1.62[-2] | -2.86[-6] | -9.27[-5] | 1.79[-5] | -4.87[-5] | 1.81[-5] |

### TABLE II. Summary of corrections to PNC amplitude in $^{133}$Cs due to Breit interaction and negative-energy states. Line $\delta H_W$ lists contributions due to modifications in the weak interaction matrix elements, and $\delta D$ due to corrections in the electric dipole matrix elements. See the Table III caption for the explanation of columns. The units are $10^{-11}i(-Q_W/N)$, and $x[y] = x \times 10^y$.

| state | Expt | $C_-$ | $B^{(1)}_+$ | $B^{(1)}_-$ | $B^{(2)}_+$ | $B^{(2)}_-$ | $\delta E_{\text{PNC}}$ |
|-------|------|-------|-------------|-------------|-------------|-------------|----------------|
| $^{133}$Cs, $g_I = 0.73789$, $R_m = 5.6748$ fm |       |       |             |             |             |             | 0.0002 | 0.0042 | 0.0019 | 0.0063 |
| $6S_{1/2}$ | 2298.2 | -0.11 | -8.14 | 0.25 | 3.50 | -0.35 | -4.64 |
| $7S_{1/2}$ | 545.90(9) | 0.03 | -1.80 | 0.07 | 0.96 | -0.097 | -0.83 |
| $6P_{1/2}$ | 291.89(9) | -6.1[-4] | -1.58 | 0.25 | 0.73 | -0.27 | -0.87 |
| $7P_{1/2}$ | 94.35 | -2.2[-4] | -5.43 | 0.09 | 0.26 | -0.098 | -0.29 |

$^{211}$Fr, $g_I = 0.888$, $R_m = 6.71$ fm

| state | Expt | $C_-$ | $B^{(1)}_+$ | $B^{(1)}_-$ | $B^{(2)}_+$ | $B^{(2)}_-$ | $\delta E_{\text{PNC}}$ |
|-------|------|-------|-------------|-------------|-------------|-------------|----------------|
| $7S_{1/2}$ | 8713.9(8) | 0.07 | -66.7 | -0.08 | 19.8 | -0.54 | -47.4 |
| $8S_{1/2}$ | 1912.5(1.3) | 0.02 | -12.8 | -0.02 | 5.08 | -0.14 | -7.88 |
| $7P_{1/2}$ | 1142.0(3) | -5.6[-3] | -10.8 | 1.23 | 3.62 | -0.95 | -6.90 |
| $8P_{1/2}$ | 362.91 | -2.0[-3] | -3.61 | 0.44 | 1.29 | -0.34 | -2.23 |

*All-order many-body calculations Ref. 9.*

### TABLE III. Contributions to hyperfine-structure constants in MHz. Column “Expt” lists experimental values, where available, and $\delta A$ gives the total of the contributions from negative-energy states and Breit interaction. See the Table I caption for the explanation of other columns. Notation $x[y] = x \times 10^y$.

| state | Expt | $C_-$ | $B^{(1)}_+$ | $B^{(1)}_-$ | $B^{(2)}_+$ | $B^{(2)}_-$ |
|-------|------|-------|-------------|-------------|-------------|-------------|
| $^{133}$Cs, $g_I = 0.73789$, $R_m = 5.6748$ fm |       |       |             |             |             |             | 0.0002 | 0.0042 | 0.0019 | 0.0063 |
| $6S_{1/2}$ | 2298.2 | -0.11 | -8.14 | 0.25 | 3.50 | -0.35 | -4.64 |
| $7S_{1/2}$ | 545.90(9) | 0.03 | -1.80 | 0.07 | 0.96 | -0.097 | -0.83 |
| $6P_{1/2}$ | 291.89(9) | -6.1[-4] | -1.58 | 0.25 | 0.73 | -0.27 | -0.87 |
| $7P_{1/2}$ | 94.35 | -2.2[-4] | -5.43 | 0.09 | 0.26 | -0.098 | -0.29 |

$^{211}$Fr, $g_I = 0.888$, $R_m = 6.71$ fm

| state | Expt | $C_-$ | $B^{(1)}_+$ | $B^{(1)}_-$ | $B^{(2)}_+$ | $B^{(2)}_-$ | $\delta E_{\text{PNC}}$ |
|-------|------|-------|-------------|-------------|-------------|-------------|----------------|
| $7S_{1/2}$ | 8713.9(8) | 0.07 | -66.7 | -0.08 | 19.8 | -0.54 | -47.4 |
| $8S_{1/2}$ | 1912.5(1.3) | 0.02 | -12.8 | -0.02 | 5.08 | -0.14 | -7.88 |
| $7P_{1/2}$ | 1142.0(3) | -5.6[-3] | -10.8 | 1.23 | 3.62 | -0.95 | -6.90 |
| $8P_{1/2}$ | 362.91 | -2.0[-3] | -3.61 | 0.44 | 1.29 | -0.34 | -2.23 |

*All-order many-body calculations Ref. 9.*
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