Calculation of isotope shifts for cesium and francium

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We perform *ab initio* calculations of isotope shifts for isotopes of cesium (from $A=123$ to $A=137$) and francium (from $A=207$ to $A=228$). These calculations start from the relativistic Hartree-Fock method and make use of several techniques to include correlations. The field (volume) isotope shift is calculated by means of an all-order correlation potential method and within the singles-doubles partial triples linearized coupled-cluster approach. Many-body perturbation theory in two different formulations is used to calculate the specific mass shift. We discuss the strong points and shortcomings of the different approaches and implications for parity nonconservation in atoms. Changes in nuclear charge radii are found by comparing the present calculations with experiment.

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I. INTRODUCTION

Accurate calculations of isotope shifts for many-electron atoms are needed to address a number of pressing problems. Although possible changes of isotope abundances in the early universe are important systematic effects in the study of variation of fundamental constants [1, 2, 3], isotope shift data are unavailable for most of the spectral lines of interest. Furthermore, studies of parity nonconservation (PNC) in atoms have come to the point where more information on nuclear structure is needed. The analysis of the most precise measurement of PNC in cesium reveals that at the present level of accuracy (0.35% for experiment, 0.5% for theory, see, e.g. [4]) there is perfect agreement with the standard model of the electroweak interactions. To search for new physics beyond the standard model in low-energy physics, one needs to improve the accuracy of the analysis. While improving the accuracy of measurements is probably feasible, improving the accuracy of calculations is problematic. To avoid problems with the accuracy of calculations, Dzuba et al. [5] suggested that PNC be measured in a chain of isotopes. The electronic structure factor would be canceled in the ratio for different isotopes. However, at the level of accuracy needed to search for new physics, the cancellation is not sufficiently complete. One must account for the change in electronic structure caused by the change in nuclear charge radius. The change in neutron distribution presents a separate problem [6] that will not be discussed further in this work, since the isotope shift is not sensitive to the changes in the neutron distribution. The change in the nuclear charge radius can be obtained by comparing experimental and theoretical values of isotope shifts. Moreover, calculations of the isotope shift provide tests of atomic wave functions at short distances, thereby providing another way of evaluating the accuracy of atomic PNC calculations.

We have chosen cesium and francium for our analysis since both atoms are being used or planed to be used in atomic PNC measurements. Also, both atoms have relatively simple electronic structure with one external electron above closed shells, making it easier to study different theoretical approaches. Accurate experimental values of isotope shifts for a range of isotopes of both atoms are available.

Isotope shifts of cesium, francium, and thallium have been studied theoretically by A.-M. Mårtensson-Pendrill [7, 8, 9]. In this paper, we present several different approaches aimed at improving the accuracy of isotope shift calculations and the corresponding analysis of changes in nuclear charge radii. We demonstrate that, owing to the extremely poor convergence of many-body perturbation theory, all-order techniques are needed to obtain reliable results.

In heavy atoms, the isotope shift is dominated by the field (or volume) shift (FS), which is the shift in energy caused by the change in the nuclear charge radius. In light atoms, the isotope shift is dominated by the mass shift, which is the change in energy due to differences in isotopic masses. Both of these effects are important for cesium. The field shift strongly dominates for francium. The FS is easier to calculate owing to the simpler form of the corresponding operator. We present two different all-order methods, both of which use the finite-field ap-
approach to reduce the calculation of the FS to a calculation of energy. The first method is the all-order correlation potential method and the second is the singles-doubles, partial triples, linearized coupled-cluster method. In order to reduce the calculation of the FS to a calculation of an energy, we incorporate the rescaled operator of the derivative of the nuclear potential with respect to the nuclear radius into the Hamiltonian at each stage of the calculation. The isotope shift is calculated as the derivative of the energy with respect to the scaling parameter. Both methods give very close results. Calculations of energies and hyperfine structures (hfs) are used as additional tests of the accuracy of the calculations. We believe that the uncertainty in our calculation of the field shift does not exceed 1%.

For reasons explained below, the same methods cannot be used at the present time for the calculation of the mass shift. Therefore, we use two different approaches there. The first approach is a third-order perturbation theory calculation (with certain classes of terms included to all orders). The second method which we use in this work is a combination of perturbation theory and the finite-field approach in which only core polarization diagrams are included to all orders. While such a treatment of the specific mass shift allowed us to obtain reasonably good results suitable for the present work, we stress the need to develop a complete all-order technique for the calculation of the mass shift, similar to that used here for the field shift.

We extract values of the change in nuclear charge radius for different isotopes of cesium and francium by comparing our calculations with available experimental data and we discuss the implications of this work for PNC in atoms.

II. METHOD OF CALCULATIONS

The shift of transition frequency of isotope $A'$ compared to isotope $A$ can be written as

$$\delta \nu_{AA'} = (K_{\text{NMS}} + K_{\text{SMS}})(\frac{1}{A} - \frac{1}{A'}) + F \delta \langle r^2 \rangle_{AA'},$$  \hspace{1cm} (1)

where $A$ and $A'$ are mass numbers of the two isotopes and $\langle r^2 \rangle$ is the mean square nuclear radius. The first term on the right represents the mass shift and the second term represents the field shift. The mass shift consists of two parts: a normal mass shift (NMS) and a specific mass shift (SMS). The normal mass shift constant is expressed in terms of the experimental frequency by

$$K_{\text{NMS}} = \frac{\nu_{\text{exp}}}{1822.888},$$ \hspace{1cm} (2)

while SMS and FS constants $K_{\text{SMS}}$ and $F$ are the subjects of the present calculations.

| TABLE I: RPA iterations for the field shift constants $F$ (MHz/m^2) in Cs. |
|---------------------------|---------------------------|---------------------------|
| $6s$ | $6p_{3/2}$ | $6p_{1/2}$ |
| 0 | -1270.24 | -15.551 | -0.0005 |
| 1 | -1191.91 | 82.136 | 95.749 |
| 2 | -1486.35 | 6.227 | 27.535 |
| 3 | -1341.68 | 52.758 | 71.219 |
| 4 | -1436.05 | 25.459 | 46.265 |
| 5 | -1388.48 | 39.612 | 59.382 |
| 6 | -1413.91 | 32.157 | 52.532 |
| 7 | -1400.94 | 35.973 | 56.055 |
| 8 | -1407.65 | 34.003 | 54.241 |
| 9 | -1404.21 | 35.013 | 55.172 |
| 10 | -1405.98 | 34.494 | 54.694 |

A. Field shift

We start with field shift calculations because they are easier to carry out and because they play more important roles in our analysis owing to their implication for PNC in atoms and their dominance for the francium isotope shift.

We use the following form for the FS operator:

$$\delta V_{\text{nuc}}(r) = \frac{dV_{\text{nuc}}(r, R_N)}{dR_N}\delta(R_N),$$  \hspace{1cm} (3)

where $R_N$ is the nuclear charge radius. We assume a Fermi distribution for the nuclear charge. The derivative $dV_{\text{nuc}}(r, R_N)/dR_N$ is calculated numerically.

The change in the nuclear potential $\delta V_{\text{nuc}}(r)$ is a small perturbation which suggests that perturbation theory is probably a proper tool for calculations. However, the convergence of perturbation theory in the residual Coulomb interaction is extremely poor. In Table I we present the dependence of the FS constants for the 6s and 6p states of Cs on the number of iterations (order of perturbation theory) of the linearized Hartree-Fock equations for an atom in external field. Here external field is produced by the change of nuclear radius and corresponding equations are equivalent to the random phase approximation (RPA) (see, e.g. [11]). The convergence for the 6s state is seen to be very poor while perturbation theory completely fails for the 6p states, inasmuch as one needs more than ten iterations to get stable results.

Therefore, instead of using perturbation theory, we use an all-order finite-field approach similar to our early work [11]. Calculations of the FS are done for the reference isotope $A$ with nuclear potential $V_{\text{nuc}}(r, R_N(A))$ replaced by

$$V_{\text{nuc}}(r, R_N(A)) + \lambda \delta V_{\text{nuc}}(r),$$ \hspace{1cm} (4)

where $\lambda$ is scaling parameter. The value of $\lambda$ is chosen in such a way that the corresponding change in the nuclear
TABLE II: Field shift constants $F$ (MHz/fm$^2$) for Cs and Fr in different approximations.

|        | Cs          | Fr          |
|--------|-------------|-------------|
|        | 6s  | $6p_{1/2}$ | $6p_{3/2}$ | $7p_{1/2}$ | $7s$ | $7p_{1/2}$ | $7p_{3/2}$ |
| HF     | -1270 | -15.7       | 0.0        | -5.6       | -14111 | -458       | 0.0        |
| RPA    | -1405 | 34.6        | 54.9       | 12.4       | -15819 | -209       | 510        |
| $\langle \Sigma^{(2)} \rangle$ | -2050 | 17.4        | 45.4       | 8.7        | -22358 | -697       | 313        |
| $\text{BO}(\Sigma^{(2)})$ | -2119 | 17.6        | 46.7       | 10.0       | -22447 | -759       | 301        |
| $\text{BO}(\Sigma^{(\infty)})$ | -1914 | 22.4        | 51.2       | 10.6       | -20463 | -640       | 361        |
| $\text{SD+E3}$ | -1894 | 23.9        | 52.2       | 11.3       | -20188 | -640       | 361        |
| Other  | -2069$^a$ | 42.78$^a$  | 70.53$^a$  | 15.17$^a$  | -20782$^b$ | -696$^b$  | 245$^b$    |

$^a$Hartley and Mårtensson-Pendrill $^b$

$^b$Mårtensson-Pendrill

Potential is much larger than the numerical uncertainty of the calculations but is still sufficiently small for the final energy to be a linear function of $\lambda$. The FS constant for a particular atomic state $v$ is then found as

$$F_v = \frac{dE_v(\lambda)}{d\lambda}$$  \hspace{1cm} (5)

This approach reduces the calculation of the FS to a calculation of energy. We use two different techniques to calculate energies. One is the all-order correlation potential method $^{11,12}$ (also called perturbation theory in the screened Coulomb interaction). Another is the singles-doubles linearized coupled cluster method combined with many-body perturbation theory (MBPT) to account for missing third-order diagrams (SD+E3). We present the results in Table III in order of improving approximations. The first line (HF) gives the average value of the $\delta V_{\text{new}}(r)$ over Hartree-Fock wave functions. The second line (RPA) gives the result of Hartree-Fock iterations with the potential given by Eq. 4. The result of this calculation (linear in the scaling parameter $\lambda$) is equivalent to the RPA. The next line ($\langle \Sigma^{(2)} \rangle$) includes second-order correlation corrections by means of many-body perturbation theory. Since we use “dressed” basis states (states calculated in a potential given by the Eq. 4 with the FS operator included) to calculate these corrections, all third-order terms are included (first in FS and second in Coulomb interaction) as well as chains of higher-order terms corresponding to core polarization. We use the notation $\hat{\Sigma}$ for the correlation correction operator (correlation potential). Details of the use of $\hat{\Sigma}$ in atomic calculations can be found elsewhere $^{10}$. The line labeled $\text{BO}(\Sigma^{(2)})$ presents results obtained by including the operator $\Sigma^{(2)}$ in the Hartree-Fock equations for the valence electron and calculating Brueckner orbitals (BO) and the corresponding energies. These results differ from those in the previous line by higher-order contributions in $\Sigma (\Sigma^2, \Sigma^3, \text{etc.})$. Finally, the line $\text{BO}(\Sigma^{(\infty)})$ presents results obtained with the all-order $\hat{\Sigma}$, in which screening of Coulomb interaction and hole-particle interactions are included in $\Sigma^{\infty}$ to all orders (see, e.g. $^{12}$ for details) and $\Sigma^{\infty}$ is used to calculate BO’s. These are the most accurate results obtained in the all-order correlation potential (CP) method. We compare them with results obtained in the SD+E3 approach presented in next line. We refer the reader to Ref. 13 for the details of the (SD+E3) all-order energy calculation. For the 6s $-$ 6p and 6s $-$ 7p intervals (the only ones important for the analysis of the experimental results), the two methods agree to about 1%.

In the last line of Table III, we present the FS calculations of Hartley and Mårtensson-Pendrill $^{7,8}$. They use an approximation very similar to that listed on line $\langle \Sigma^{(2)} \rangle$. Naturally, the results are also very close. The larger discrepancy seen for the 6p states of Cs is probably due to larger contributions from structural radiation diagrams (which are called internal diagrams in Ref. 7). In our approach, calculation of the IS is reduced to a calculation of energy and no special treatment for the structural radiation diagrams is needed. In contrast, Hartley and Mårtensson-Pendrill evaluated these diagrams using a procedure based on a modification of the basis. We believe that our calculations are more accurate since (a) the finite-field approach ensures that no important diagrams are missed, (b) we have very good agreement between two very different methods, and (c) we have very good agreement with experiment for both energies and hyperfine structures (see below).

The final result of Hartley and Mårtensson-Pendrill for the 6s state of Cs (-2000 MHz/fm$^2$) was obtained by rescaling the ab initio value using a comparison between theoretical and experimental hyperfine structure (hfs) constants. Their calculated value for the hfs of 6s is larger than the experimental value by 3%. Therefore, they reduced the FS constant by the same ratio. Since we include higher-order correlations which bring calculated values of the hfs constants and the energies into very good agreement with experiment, we can check how well this rescaling works.

Table III presents the data for the second- and all-order energies, hfs constants, and FS constants for Cs and Fr. We see that higher-order correlations reduce the ground state energy by 3%, the hfs constant by 5 to 8% and FS by 9 to 10%. Therefore, energies cannot be used for rescaling of the FS constants, and accuracy of extracting the higher-order correlation correction to the FS by rescaling the second-order correction using hfs data is between 20 and 40%. This corresponds to 2 to 4% accuracy in final results for s-states. Moreover, no reliable rescaling can be done for p-states.

Note that the rescaling of FS is very different from the “fitting of the energy” procedure used in some of our works (see, e.g. 4) and in the next section. In that procedure, we replace $\Sigma$ in equations for valence BO’s by the rescaled operator $\lambda \Sigma$ with $\lambda$ chosen to fit experimental energies exactly. New BO’s are then used to calculate matrix elements. This procedure changes energies and matrix elements at different rates usually
TABLE III: Contributions of the higher-order correlations to the energies, hyperfine constants (hfs), and field shift constants (FS) of Cs and Fr.

|          | BO(\(\Sigma^{(2)}\)) | BO(\(\Sigma^{\infty}\)) | Ratio     | Exp     |
|----------|----------------------|--------------------------|-----------|---------|
| Cesium energies (cm\(^{-1}\)) |                       |                          |           |         |
| 6s       | 32375                | 31470                    | 0.97      | 31407\(^a\) |
| 6p\(_{1/2}\) | 20524                | 20296                    | 0.99      | 20229\(^b\) |
| 6p\(_{3/2}\) | 19926                | 19728                    | 0.99      | 19675\(^c\) |
| 133Cs hfs \((g_I=0.7377208)\) (MHz) |                       |                          |           |         |
| 6s       | 2459                 | 2270                     | 0.92      | 2298.2\(^d\) |
| 6p\(_{1/2}\) | 314                 | 295                      | 0.94      | 291.89\(^e\) |
| 6p\(_{3/2}\) | 51.8                 | 48.7                     | 0.94      | 50.275\(^d\) |
| Francium energies (cm\(^{-1}\)) |                       |                          |           |         |
| 7s       | 34089                | 32899                    | 0.97      | 32849\(^f\) |
| 7p\(_{1/2}\) | 20986                | 20711                    | 0.99      | 20612\(^g\) |
| 7p\(_{3/2}\) | 19164                | 18976                    | 0.99      | 18925\(^h\) |
| 211Fr hfs \((g_I=0.888)\) (MHz) |                       |                          |           |         |
| 7s       | 9269                 | 8769                     | 0.95      | 8713.9(8)\(^i\) |
| 7p\(_{1/2}\) | 1261                | 1193                     | 0.95      | 1142.0(3)\(^g\) |
| 7p\(_{3/2}\) | 98.3                 | 102.4                    | 1.04      | 94.9(3)\(^j\) |
| Francium FS (MHz/fm\(^2\)) |                       |                          |           |         |
| 7s       | -22447               | -20463                   | 0.91      |         |
| 7p\(_{1/2}\) | -759                 | -693                     | 0.91      |         |
| 7p\(_{3/2}\) | 301                  | 303                      | 1.01      |         |

\(^a\)Moore \(^{14}\)
\(^b\)Arimondo et al. \(^{12}\)
\(^c\)Rafac and Tanner \(^{16}\)
\(^d\)Tanner and Weiman \(^{17}\)
\(^e\)Bauche et al. \(^{18}\)
\(^f\)Arnold et al. \(^{19}\)
\(^g\)Ekström et al. \(^{20}\)
\(^h\)Grossman et al. \(^{21}\)

The substitution (6) can be easily done in the Hartree-Fock approximation or in perturbation theory calculations. However, the methods used in the previous section to include higher-order correlation corrections in the FS constant are not applicable here. Note that for FS calculations only the Hartree-Fock program needs to be modified to incorporate the change in nuclear potential. By contrast, for SMS calculations every program must be modified. While this is still straightforward in Hartree-Fock and perturbation theory codes, it becomes much more difficult in higher orders. In the correlation potential method, inclusion of higher-order correlations is done by summing the matrix geometric progression (7)

\[ \tilde{Q} = Q + OIQIQ + OIQIQ + \ldots \]

where \(Q\) is the Coulomb interaction \((Q_k(r_1,r_2) = r_1^{-k}/r_2^{k+1})\), \(I\) is polarization operator, and \(\tilde{Q}\) is screened Coulomb interaction (see, e.g., \(^{12}\) for details). To include the SMS operator in this summation, one would need to modify Coulomb interaction in the following way

\[ Q \rightarrow Q + \lambda P, \]

where \(P\) is the coordinate representation of the SMS operator, which would lead to the correct expressions for radial integrals (6) when integrated over the wave functions. It is clear that there is no such representation for the SMS operator. This problem does not appear in the SD+E3 method, since everything there is expressed in terms of Coulomb integrals which can be modified according to Eq. (6). While such modification is as straightforward as for perturbation theory codes, technically it is not an easy task. The problem here is not only with the large number of terms which must be modified, but also with different symmetry properties of the SMS operator. Exchanging indexes \(a\) and \(c\) (or \(b\) and \(d\)) in Eq. (6) leaves the Coulomb part of the equation unchanged while the SMS contribution changes sign! While we stress that it would be extremely useful to have a finite-field program for the SMS, we must leave this for future work.

In the present work, we use two less sophisticated (and less accurate) approaches. The first is perturbation theory (7) and the second is a finite-field approach in which only the second-order correlation operator \(\Sigma^{(2)}\) is used. In the perturbation theory calculation, we express the SMS operator \(P = \sum_{i<j} P_i \cdot P_j\) as a sum of a normally ordered one-particle operator \(S\) and normally ordered two-particle operator \(T\). We carried out an all-order calculation of the matrix element of \(S\); the calculation of the matrix element of \(T\) is complete through third-order (first order in the SMS operator and up to second order in Coulomb interaction). The results of the perturbation theory calculation for the SMS for Cs and Fr are presented in Table IV. The lowest-order values are given in row \(S^{(1)}\), the results of the all-order singles-doubles calculation of the matrix element of \(S\) are given in row \(SD\), and the second- and third-order matrix elements of \(T\) are...
TABLE IV: Perturbation theory contributions to specific mass isotope shift constants (GHZ amu) in Cs and Fr

|       | Cs     | Fr     |
|-------|--------|--------|
|       | $6s$   | $6p_{1/2}$ | $6p_{3/2}$ | $7s$   | $7p_{1/2}$ | $7p_{3/2}$ |
| $S^{(1)}$ | -781.3 | -191.4  | -168.7  | -1359.7 | -260.0 | -187.0 |
| $SD$   | 316.5  | 139.9   | 140.3   | 363.7   | 182.7  | 182.4  |
| $T^{(2)}$ | 286.8  | 58.5    | 51.2    | 499.9   | 78.9   | 57.5   |
| $T^{(3)}$ | -136.2 | -30.1   | -27.8   | -296.0  | -53.7  | -45.0  |
| Total  | -314.2 | -23.1   | -5.0    | -786.1  | -53.0  | 7.9    |

given in rows labeled $T^{(2)}$ and $T^{(3)}$. The total values of the specific mass shift constants are listed in the last row.

Table V presents results of the finite-field approach. The line HF gives the expectation values of the SMS operator over HF wave function of the valence electron. It is equivalent to the line $S^{(1)}$ of Table IV. The difference in numerical values is due to the fact that the relativistic form of the momentum operator was used in the PT calculations ($S^{(1)}$) while the non-relativistic operator was used in the finite-field calculation. Note that relativistic corrections for $s-p$ intervals do not exceed 4% for Cs and 8% for Fr.

This is a negligible contribution since the mass shift itself is small for heavy atoms owing to the huge suppression by the mass factor (see Eq. (1)). Note, however, that relativistic corrections are probably very important for highly-charged ions.

The line RPA in Table IV presents results of HF iterations with the SMS operator included in the HF potential by redefining of the Coulomb interaction according to Eq. (6). The line $\Sigma^{(1)}$ includes second-order correlation corrections. Finally, the line Brueck presents results for valence electron Brueckner orbitals calculated with using $\Sigma^{(2)}$. We have also included two ways of simulating higher-order correlations to try to further improve the results. Note that Brueckner orbitals with second order $\Sigma$ considerably overestimate the correlation correction to the energy. They probably have the same effect on the SMS. Therefore, we reduce the total correlation correction to the SMS in two different ways.

Firstly, we note that iterations of $\Sigma^{(2)}$ enhance correlation corrections to the SMS by a larger factor than for energies. If we use the energy ratio to determine the enhancement instead, the total correlation correction to the SMS is smaller. The corresponding interpolated results are presented in the line Interp in Table IV. Note that the correction is huge. It can even change sign of the SMS constant. This is very different from the FS constants discussed in previous section. For the FS constants higher-order correction is small and should be treated accurately. This is why rescaling works for $s$-states only.

Secondly, we rescaled the operator $\Sigma^{(2)}$ while calculating Brueckner orbitals to fit the experimental energies. Scaling factors are $\lambda(6s)=0.802, \lambda(6p)=0.85$ for Cs and $\lambda(7s)=0.786, \lambda(7p)=0.85$ for Fr. This procedure also reduces the correlation correction to the SMS. Corresponding results are presented in line Fit of Table IV. It is interesting that the two procedures give close results.

Comparison of results in Table IV and Table V reveals the poor convergence of perturbation theory and the significant difference in final results between the two calculation methods. Note that the two methods are equivalent at second order in the Coulomb interaction. The difference comes from higher orders. Perturbation theory calculations use the SMS matrix elements which are just expectation values of the SMS operator over HF wave functions (HF matrix elements). By contrast, the finite-field approach corresponds to including “dressed” SMS matrix elements in which certain chains of Coulomb diagrams are included to all orders by iterating the HF equations (RPA matrix elements). By comparing lines HF and RPA of Table IV one can see that this indeed must lead to large differences in final results. Note that the Brueckner-orbital calculations are also in better agreement with the calculations of Hartley and Martensson-Pendrill.

To check whether Brueckner orbitals really give better results than PT for the SMS, we have performed calculations for potassium and rubidium. For these atoms, “experimental” values of the SMS can be inferred by subtracting the NMS and FS from known experimental values of the isotope shifts. The NMS is given by Eq. (2) while the field shift is calculated as $F\delta(r^2)$. The field shift constant $F$ can be calculated to high precision as described in the previous section. The corresponding values are presented in Table VI. Values of $\delta(r^2)$ between the most abundant isotopes are also known from experimental studies [23]. Table VII presents the extraction of the “experimental” SMS between $^{39}$K and $^{41}$K for the $4s-4p_{1/2}$ transition and between $^{85}$Rb and $^{87}$Rb for the $5s-5p_{3/2}$ transition. In Table VIII we compare these experimental results with calculations. One can see that interpolated, fitted, and unfitted Brueckner orbital results are closest to experiment while the perturbation theory results are much further away. It is natural, therefore, to take the average value of the most accurate results (Brueck, Interp and Fit) as a final central point of the calculations, while using differences between these results as an estimate of numerical uncertainty.

### III. RESULTS AND DISCUSSION

Final values for the mass and field shift constants are presented in Table IX. These are the BO ($\Sigma^{\infty}$) results for FS (see Table ID) and the average of Brueck, Interp and Fit results for SMS (see Table VII). We use these values to analyze experimental data and to extract the change in nuclear charge radius for a range of isotopes of cesium and francium. The results are presented in Tables X and XI. We present two uncertainties for the $\delta(r^2)$ for Cs. The first one is experimental and the second one is theoretical. The theoretical uncertainty is dominated by
TABLE V: SMS constants for Cs and Fr in different approximations (GHZ amu).

|        | 6s    | 6p_{1/2} | 6p_{3/2} | 7p_{1/2} | 7p_{3/2} | 7s     | 7p_{1/2} | 7p_{3/2} |
|--------|-------|----------|----------|----------|----------|--------|----------|----------|
| HF     | -773.18 | -208.70  | -170.40  | -74.68   | -61.44   | -1330.48 | -317.79  | -190.87  |
| RPA    | -355.55 | -76.96   | -40.31   | -27.59   | -14.56   | -666.71  | -127.90  | -10.48   |
| Σ(2)   | -133.19 | -22.83   | 20.66    | -5.63    | 8.81     | -334.15  | -85.24   | 60.08    |
| Brueck | 6.75   | 6.85     | 47.33    | 0.95     | 14.31    | -110.29  | -45.11   | 89.86    |
| Interp | -99.84 | -14.39   | 29.38    | -4.31    | 10.21    | -288.26  | -77.90   | 69.96    |
| Fit    | -89.31 | -10.26   | 30.17    | -2.94    | 10.47    | -311.96  | -63.83   | 70.35    |
| PT\(^c\) | -314.2 | -23.1    | 9.2\(^b\) | -5.0     | -786.1   | -53.0    | -7.9     |
| Other  | -23.5\(^b\) | -36.6\(^b\) | 9.2\(^b\) | -5.0     | -786.1   | -53.0    | -7.9     |

\(^a\)From Table IV
\(^b\)Hartley and Martenson-Pendrill
\(^c\)Martenson-Pendrill

TABLE VI: Field shift constants for K and Rb (MHz/fm\(^2\))

|        | K 4s | K 4p_{1/2} | Rb 5s  | Rb 5p_{3/2} |
|--------|------|------------|--------|-------------|
| K 4s  | -104.20 | 4.04 | -551.85 | 15.60 |

TABLE VII: Extracting an “experimental” SMS for K and Rb.

| IS(exp) | \(\delta(r^2)^a\) | FS    | NMS   | SMS   |
|---------|------------------|-------|-------|-------|
| MHz     | (fm\(^2\))      | (MHz) | (MHz) | (MHz) |
| 39\(^a\)-41K 4s - 4p_{1/2} | 235.25(75)\(^b\) | 0.105 | -11.33 | 267.11 |
| 85\(^b\)-87Rb 5s - 5p_{3/2} | 77.992(20)\(^c\) | -0.042 | 23.83 | 57.00 |

\(^a\)Angeli
\(^b\)Touchard et al.
\(^c\)Banerjee et al.

the uncertainty in the SMS constant. Strong cancellation between the field shift and the normal mass shift makes these results sensitive to the SMS. However, the very poor convergence of perturbation theory make it difficult to predict the SMS to high accuracy. We stress once more the need for accurate all-order techniques to calculate the SMS.

It has been long known (see, e.g. 27) that the change of nuclear radius along the chain of Cs isotopes is slower than expected from the formula \(R_N = 1.1A^{1/3}\). One possible explanation for this fact is that neutrons and protons have different distributions. According to data deduced from antiprotonic atoms 27, the radius of the neutron distribution for \(^{133}\)Cs is by 0.16 ± 0.06 fm larger than radius of the proton distribution. It is interesting to note that under certain assumptions, the isotope shift data is in very close agreement with the antiprotonic atom data. These assumptions are: (a) the \(A^{1/3}\) law is still valid but for the total nuclear radius (including the “neutron skin”) not just charge radius; (b) the neutron and proton distributions are very close for the neutron poor nuclei. This is one more argument in favor of the “neutron skin” correction to the PNC in Cs 27.

Table VIII presents IS data for Fr. The experimental accuracy for the IS is very high. Therefore, only the theoretical error is presented for the \(\delta(r^2)\). The SMS is less than 1% of the total IS for Fr. Also, there is a strong cancellation between the SMS and the NMS, which makes the extracted values of the change in nuclear charge radius insensitive to the SMS. The theoretical uncertainty is therefore quite low and at the level of 1% which is
mostly the uncertainty in the FS constant. Our results for \( \delta(r^2) \) differ from those obtained in \( \text{[S]} \) by about 2.4%. This is due to the difference in the FS constant in the \( 7s - 7p_{3/2} \) transition. Our value is -20.8 GHz amu while a value of -21.0 GHz amu is used in \( \text{[S]} \). Since 2% uncertainty is estimated in \( \text{[S]} \), and we believe that our accuracy is 1%, we can say that the results are in good agreement.

Since nuclear radii change quite significantly along the chain of francium isotopes, it is important to check how PNC matrix elements are affected by this change. We have conducted a numerical test for the change of the matrix element \( W(R_N) \equiv \langle 7s|H^{PNC}|7p_{1/2} \rangle \) with the change of nuclear radius, assuming that proton and neutron distributions remain the same. The numerical results can be presented in a form

\[
\frac{W(R_N)}{W(R_0)} = 1 - 0.21 \left( \frac{R_N}{R_0} - 1 \right),
\]

where \( R_0 \) is the nuclear radius of a reference isotope. Note that total change between the lightest and the heaviest isotopes in Table X is almost 1%. This is a significant change and should be taken into account in any future analysis of the PNC in the chain of Fr isotopes.

Let us now discuss the role that isotope shift calculations may play in the study of PNC in atoms. As it is clear from the results presented above, for atoms in the middle of the periodic table (Cs, Ba, etc.) the information that can be extracted from the IS calculations and used for the PNC analysis is limited by the accuracy of the SMS calculations. Unless adequate methods are developed to significantly improve the accuracy of such calculations it is unlikely that the IS data will provide information of any practical use for PNC analysis.

The situation is very different for heavier atoms, such as Fr, Tl, etc. The mass shift is small and the corresponding uncertainty can be reduced to an acceptable level. On the other hand, calculations of the FS are much easier and can be done with accuracy of about 1% or possibly better. There are several possibilities arising from this fact. To use the IS data to test the electron wave function one needs to know the value \( \delta(r^2) \) from an independent source. Such data can be obtained from electron scattering, analysis of X-rays from muonic atoms, etc. (see, e.g. \( \text{[23]} \)). However, the accuracy of that data is often insufficient for the PNC purposes. One can do a consistency test instead. If the isotope shift is known for several different transitions, one can check whether comparison of the theory with experiment leads to the same value of \( \delta(r^2) \) for all transitions.

There is another possibility for many-electron atoms. If the IS is known for an ion with simple electronic structure (one electron above closed shells), then calculations of the FS are much simpler than in atoms with closed shells (e.g. Fr). However, the accuracy of that data is often insufficient for the PNC purposes. One can do a consistency test instead. If the isotope shift is known for several different transitions, one can check whether comparison of the theory with experiment leads to the same value of \( \delta(r^2) \) for all transitions.

IV. CONCLUSION

We have developed methods to calculate the isotope shift for many-electron atoms with one external electron above closed shells. While methods for the field shift
seem to be adequate and capable of producing results at the 1% or better level of accuracy, methods for the SMS need further consideration. It would be useful to have an all-order technique similar to that used in the FS calculations to address the problem of the very poor convergence of perturbation theory. We use our calculations for cesium and francium to extract the change in nuclear charge radius for chains of isotopes in both atoms. We have demonstrated that, at least for heavy atoms, calculations and measurements of the isotope shifts may provide important information for the analysis of the PNC in atoms.

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