Appraising nuclear octupole moment contributions to the hyperfine structures in $^{211}$Fr

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I. INTRODUCTION

When the non-central electromagnetic fields of nuclear magnetic moments interact with the atomic electrons, they give rise hyperfine structures to the atomic energy levels [1]. These energy levels are conventionally expressed in terms of the hyperfine structure constants, which are traditionally denoted by $A$, $B$, $C$ etc., that depend on the electron wave functions and nuclear moments. These constants are typically determined experimentally by measuring hyperfine structure energy levels for different angular momentum states of the atomic systems and fitting in a set of equations involving the corresponding angular momentum coefficients [2]. Among all the hyperfine structure interactions, the magnetic dipole hyperfine component typically contributes predominantly for a finite nuclear spin ($I$) atomic systems compared to others followed by the electric quadrupole hyperfine component and so on when $I > 1/2$. Owing to comparable orders of magnitudes of the higher multipole contributions with the systematics in the measurements of hyperfine splittings, it is extremely strenuous to estimate contributions due to these higher nuclear moments, especially from the nuclear octupole moment on wards, to the hyperfine structure. Thus, hyperfine structure constants mostly up to the electric quadrupole interactions are usually reported by the experimental studies. However with the advent of modern technologies, it is now possible to measure hyperfine splittings very precisely from which contributions until the magnetic octupole interaction are segregated in a number of systems like $^{85}$Rb [3], $^{133}$Cs [4], $^{137}$Ba$^+$ [5] and $^{173}$Yb [6].

On the other hand direct measurements of nuclear moments are obscured, since isolating a bare nucleus from the external electromagnetic fields is not a straightforward task. They are obtained either from the nuclear magnetic resonance (NMR), nuclear quadrupole resonance (NQR) etc. measurements or by combining experimental results of $A$, $B$, $C$ etc. with their corresponding atomic and molecular calculations. Nuclear magnetic moment ($\mu$) obtained from an NMR measurement is generally more precise than its value obtained from the hyperfine structure constant due to achievement of high precision results using the NMR spectroscopy technique [7]. But, many of the available nuclear quadrupole moments ($Q$s) for different isotopes are obtained from the measured hyperfine structure constants [7]. Similarly, all the reported nuclear octupole moments ($\Omega$s) for the above investigated atoms are inferred from the high precision measurements on hyperfine splittings [3, 4, 8]. Nevertheless there are no NMR spectroscopy of the Fr isotopes available till date, $\mu$ and $Q$ values of $^{211}$Fr were extracted out by measuring energies of different hyperfine transitions in its ground state using the atomic-beam magnetic resonance (ABMR) method [9] and by combining measured $B$ value of the $7p^22P_{3/2}$ state with an atomic calculation using a lower-order relativistic many-body calculation [10], respectively.

Theoretical studies of hyperfine structure constants are of immense interest because, comparison with their corresponding experimental results are served as bench marks to test the capability of an employed many-body method for their precise determination. Since accuracies in the evaluation of these quantities are very sensitive in the calculations of the atomic wave functions in the nuclear region, their precise estimates are the indications of the potentials of the methods to produce the atomic wave functions appropriately in this region. Thats why their theoretical studies have been drawn a lot of attentions in the context of investigating atomic parity violating (APV) effects and electric dipole moments (EDMs) due to parity and time-reversal symmetry violations in the atomic systems [11, 12]. Fr is under consideration for both the APV and EDM measurements [13, 10]. In our previous work [14], we had applied relativistic many-body methods at various approximations to determine the $A$ and $B$
constants of $^{210}$Fr and $^{212}$Fr. In that work, we had found that the relativistic coupled-cluster (RCC) theory at the truncation level of singles, doubles and important valence triple excitations in the perturbative approach (CCSD$_3$ method) gives rise the results within the reasonable accuracies. Since the aim of the present work is to estimate $\mu$ and $Q$ values of $^{211}$Fr precisely and demonstrate possible way of extracting $\Omega$ value of $^{211}$Fr, we carry out the calculations using the same CCSD$_3$ method for this isotope here.

II. THEORY

The Hamiltonian describing the non-central hyperfine interactions between the electrons and nucleus in an atomic system in terms of the tensor operators is given by \[2\]

\[ H_{hf} = \sum_k M^{(k)} \cdot T^{(k)}, \]

where $M^{(k)}$ and $T^{(k)}$ are the magnitudes of the $M^{(k)}$ and $T^{(k)}$ tensors, respectively, the allowed values of ranks $k_1$ and $k_2$ in the above multipole expansions depend on the selection rules of Sixj angular momentum factors and $E_J$ is the energy of an atomic state with angular moment $J$. In the present interest of study, we restrict the multipole values only to $k_1 = 1, 2, 3$. Definitions of the nuclear moments are given by

\[
\begin{align*}
\langle II|M^{(1)}|II \rangle & = \mu = \mu_N \mu_I, \\
\langle II|M^{(2)}|II \rangle & = \frac{1}{2} Q, \\
\langle II|M^{(3)}|II \rangle & = -\Omega = -\mu_N \Omega_I \text{ etc.},
\end{align*}
\]

where $\mu_I$ and $\Omega_I$ are the dimensionless nuclear dipole and octupole moments respectively, and $\mu_N = \frac{e}{2m_e}$ is the nuclear magneton with the proton mass $M_p$. Nuclear shell-model yields expressions for $\mu_I$, $Q$ and $\Omega_I$ as

\[
\begin{align*}
\mu_I &= -\langle II|r^3C^{(1)}_{00}(\theta, \phi)(\vec{\nabla} \times \vec{D})|II \rangle/\mu_N \\
&= \begin{cases} \\
(0, (g_s - g_I)/2I) & \text{for } I = l + 1/2 \\
(g_I - (g_s - g_I)/(2I + 2) & \text{for } I = l - 1/2,
\end{cases}
\end{align*}
\]

and

\[
\begin{align*}
Q &= \langle II|gr^2C^{(2)}_{00}(\theta, \phi)|II \rangle \\
&= -\frac{2I - 1}{2I + 2}g_I \langle r^2 \rangle
\end{align*}
\]

The first and the second orders changes in the energies of an atomic energy level due to the hyperfine interactions can be given by \[2\]

\[
W^{(1)}_{F,J} = \sum_{k_1} (-1)^{I + J + F} \left\{ \begin{array}{ccc} J & I \\
I & J \end{array} \right\} F \frac{k_1}{1} \langle II|M^{(k_1)}|II \rangle \langle J||T^{(k_1)}||J \rangle
\]

and

\[
W^{(2)}_{F,J} = \sum_{J'} \frac{1}{E_J - E_{J'}} \sum_{k_1, k_2} \left\{ \begin{array}{ccc} F' & J & I \\
J & I \end{array} \right\} \left\{ \begin{array}{ccc} F' & J' & I \\
J' & I \end{array} \right\} \langle II|M^{(k_1)}||II \rangle \langle F||M^{(k_2)}||I \rangle \langle J'||T^{(k_1)}||J \rangle \langle J'|T^{(k_2)}||J \rangle,
\]

with the nuclear magnetization density $\vec{D}$, $\langle r^2 \rangle$ is the mean square radius of the nucleus and $g_I$ and $g_s$ are the orbital and spin gyromagnetic constants for the odd nucleon of the nucleus (as applicable in case of $^{211}$Fr). Values of the nuclear spin $I = l - 1/2 = 9/2$ \[18\] and moment $\mu_I = 4.00(8)$ \[20\] of $^{211}$Fr indicates that this isotope has an odd-proton in the $2n_{9/2}$ level following the nuclear shell-model. Hence, the nuclear moments of $^{211}$Fr can be estimated using the shell-model by substituting values as $g_I \approx 1.16$ and $g_s \approx 5.5857$ \[19\] and $\langle r^2 \rangle \approx 0.308 b$ \[20\].

Rewriting Eq. \[2\] in terms of contributions from the
individual multipoles, it yields

\[ W^{(1)}_{F,J} = W^{M1}_{F,J} + W^{E2}_{F,J} + W^{M3}_{F,J}, \]

where \( W^{M1}_{F,J}, W^{E2}_{F,J}, \) and \( W^{M3}_{F,J} \) are the contributions due to the magnetic dipole (\( M1, k = 1 \)), electric quadrupole (\( E2, k = 2 \)) and magnetic octupole (\( M3, k = 3 \)) interactions, respectively, which are given by

\[ W^{M1}_{F,J} = AJ \]  
\[ W^{E2}_{F,J} = B \left( \frac{3(IJ)^2 + \frac{3}{2}(IJ) - I(I + 1)J(J + 1)}{2I(2I - 1)J(2J - 1)} \right) \]  
\[ W^{M3}_{F,J} = C \left[ \frac{1}{(IJ)(2I - 1)(J - 1)(2J - 1)} \right] \times \left[ \frac{10(IJ)^3 + 20(IJ)^2 + 2(IJ)}{3I(I + 1)J(J + 1) + I(J + 1)} \right] + 5I(I + 1)J(J + 1) \],

and

\[ T^{(2)}_q = \sum_j i_q^{(2)}(r_j) = \sum_j -ie\mu_N \sqrt{8\pi/3r_j^2} \alpha_j Y_{1q}^{(0)}(r_j), \]

\[ T^{(3)}_q = \sum_j i_q^{(3)}(r_j) = \sum_j -ie\mu_N \sqrt{8\pi/3r_j^2} \alpha_j Y_{1q}^{(0)}(r_j), \]

where single particle matrix elements are

\[ \langle k\ell \mu | T^{(1)}_q | k'\ell' \mu' \rangle = -\mu_N \langle k\ell \mu | C^{(1)}_q | k'\ell' \mu' \rangle, \]  
\[ \langle k\ell \mu | T^{(2)}_q | k'\ell' \mu' \rangle = -\mu_N \langle k\ell \mu | C^{(2)}_q | k'\ell' \mu' \rangle, \]  
\[ \langle k\ell \mu | T^{(3)}_q | k'\ell' \mu' \rangle = -\mu_N \langle k\ell \mu | C^{(3)}_q | k'\ell' \mu' \rangle, \]

and

\[ \langle k\ell \mu | \Psi_n \rangle = e^T \{ 1 + S_n \} | \Phi_n \rangle, \]

where \( T \) and \( S_n \) are the excitation operators involving core and core-valence electrons, respectively, with the reference state \( | \Phi_n \rangle \), which is obtained using the Dirac-Fock (DF) method in the present work. In the CCSD\textsubscript{II3} method, the RCC excitation operators are given by

\[ T = T_1 + T_2 \quad \text{and} \quad S_n = S_{1n} + S_{2n}, \]
where the subscripts 1 and 2 represent for the single and
double excitations.
The hyperfine structure constants are determined by
estimating the differences in our calculations of the hyperfine structure con-
stants with respect to their experimental values. In order to
test the accuracies of our results. Calculations for some of the states
were reported by Safronova

and Dzuba

Using the CCSDA method calculations for A/µ and B/Q and
considering the precise values of µ and Q values of 211Fr from Table I, we evaluate theoretical values of A and
B. These values are given in Table II along with the
results from the previous calculations and available experimental results. Comparison of our calculations with
the experimental results give some indication on the accuracies of our results. Calculations for some of the states
are reported by Safronova et al using a similar method
like ours but keeping only the linear terms in the RCC
theory (SDpT method). Dzuba et al had also employed a restricted Hartree-Fock method in the relativistic
framework and incorporated correlation effects using
many-body perturbation theory to investigate correlation effects in the hyperfine structure constants of a few low-
lying states of 211Fr. Heully and Mårtensson-Pendrill had used a relativistic many-body perturbation method
treating polarization effects to all orders. Our calcula-
tions match well with the calculations of Safronova et al and Dzuba et al, but calculations by Heully and
Mårtensson-Pendrill differ significantly from all the other
calculations. The reason may be owing to predominant contributions from the pair-correlation effects to the hyperfine structure, which are missed out in the method employed by Heully and Mårtensson-Pendrill. In fact, theoretical results of A from the CCSDA method seem to be more accurate among all the calculations. This suggests that our B/Q and C/Ω calculations are also reliable enough to be used for inferring Q and Ω values combined with their corresponding B and C values (may be measured in future). So this justifies why our extracted new Q value is more valid than the previously estimated values. Now combining the new Q value

| Moment | Shell-model | Atomic study | Reference |
|--------|-------------|--------------|-----------|
| µI     | 3.41        | 4.00(8)      | [9]       |
| Q      | −0.26       | −0.19(3)     | [9]       |
|        | −0.24       | −0.21(2)     | [10]      |
| ΩF     | 0.229       | Unavailable  |           |

In Table II we now give the values of nuclear moments
of 211Fr estimated using the shell-model and compare
them against the precise values obtained from the atomic
studies. This comparison demonstrates that the nuclear
shell-model values for µ and Q differ only by 15-25% from
the atomic results. Since till date hyperfine splittings in
211Fr are not yet measured very precisely to be able to
infer Ω (or ΩF) value from those measurements, the pre-
liminary value obtained from the shell-model can be used
to estimate contributions from the octupole component
to the hyperfine splittings by combining with our calcula-
tions so that some ideas about how precisely it is required
to measure the hyperfine splittings to infer the Ω value of
211Fr unambiguously can be gauge.

IV. DISCUSSIONS AND RESULTS

Before perusing in estimating W_{FJ} contributions for
different states, it would be appropriate to test the accu-
racies in our calculations of the hyperfine structure con-
stants with respect to their experimental values. In order
to estimate A and B values theoretically, it also requires
knowledge of µ and Q values. Ekström et al had mea-
sured hyperfine splittings between at least 14 sub-states
in the ground state of 211Fr employing the ABMR tech-
nique and adopted a least-square fit on them to separa-
ate out the electronic and nuclear parts of the hyper-
fine structure from which they had extracted out its µ value as 4.00(8) µN [9]. This value seems to be very pre-
"
TABLE II: Theoretical and experimental results of $A$, $B$ and $C$ in MHz. Results under the head of DF and CCSD$_{t3}$ methods are from the present work. References for the other results are given at the bottom of the table.

| State         | $A$ values       | $B$ values       | $C \times 10^4$ values |
|---------------|------------------|------------------|------------------------|
|               | CCSD$_{t3}$      | Others           | Experiment             | DF         | CCSD$_{t3}$ |
| $7s \ ^2S_{1/2}$ | 8786.20          | 8833.0$^a$       | 8713.9(8)$^b$          |           |             |
|               | 9027.04$^c$      | 8698.2(10.5)$^d$ |                        |           |             |
| $7p \ ^2P_{1/2}$ | 1137.40          | 1162.1$^a$       | 1139.2(14)$^b$         |           |             |
|               | 1125.12$^c$      | 1142.1(2)$^e$    |                        |           |             |
|               | 778.0$^f$        |                  |                        |           |             |
| $7p \ ^2P_{3/2}$ | 92.68            | 91.80$^a$        | 94.9(3)$^b$            |           |             |
|               | 102.34$^c$       | 94.7(2)$^d$      |                        |           |             |
|               | 85.5$^f$         |                  |                        |           |             |
| $6d \ ^2D_{3/2}$ | 75.72            |                  |                        |           |             |
| $6d \ ^2D_{5/2}$ | -52.28           |                  |                        |           |             |
| $8s \ ^2S_{1/2}$ | 1912.64          | 1923.3$^a$       |                        |           |             |
|               | 1971.96$^c$      |                  |                        |           |             |
| $8p \ ^2P_{1/2}$ | 357.36           | 362.91$^a$       | 358.0(4)$^g$           |           |             |
|               | 364.14$^c$       |                  |                        |           |             |
| $8p \ ^2P_{3/2}$ | 31.12            | 30.41$^a$        | 31.47(4)$^g$           |           |             |
|               | 35.24$^c$        |                  |                        |           |             |
| $7d \ ^2D_{3/2}$ | 27.48            |                  | -6.41                  |           |             |
| $7d \ ^2D_{5/2}$ | -14.48           |                  | -8.38                  |           |             |

References:
$^a$[25], $^b$[23], $^c$[26], $^d$[22], $^e$[27], $^f$[10], $^g$[28].

with the CCSD$_{t3}$ results and calculation by Heully and Mårtensson-Pendrill of $B/Q$, we get the $B$ values for all the considered states of $^{211}$Fr. In fact, we have calculated these quantities for many states as gives freedom to the experimentalists to select suitable states as per their choices to measure the hyperfine splittings within the required precision so that $\Omega$ of $^{211}$Fr can be inferred. Though experimental results of $A$ for many states are not available to verify their accuracies, but good agreements for the states for which measurements are available provide some confidence on their reliabilities. Moreover, consistencies among the calculated results also imply that our calculations are quite accurate. On this basis, we also expect that our calculations for $B/Q$ and $C/\Omega$ are reasonably accurate.

Roles of the correlation trends in the evaluation of the $A/\mu$ and $B/Q$ values both in $^{210}$Fr and $^{212}$Fr isotopes were already demonstrated explicitly by us recently in [17], we also observe the similar trends in the $^{211}$Fr though there are slight changes in the results owing to different nuclear structure. Just to gain some insights into the roles of the correlation effects in the evaluations of the $C/\Omega$ values, we have given the estimated $C$ values from both the DF and CCSD$_{t3}$ methods in Table [11] using $\Omega_t = 0.229b$ from the nuclear shell-model. Differences between these results show contributions from the electron correlation effects captured by the CCSD$_{t3}$ method in the evaluations of $C/\Omega$. We also observe that the trend of the correlation effects in the evaluations of $C$ and $A$ are almost similar. In both the cases, the core-

TABLE III: Important off-diagonal matrix elements in MHz among the fine structure partners obtained using our DF and CCSD$_{t3}$ methods. To determine the conjugate matrix elements between these results show contributions from the electron correlation effects captured by the CCSD$_{t3}$ method in the evaluations of $C/\Omega$. We also observe that the trend of the correlation effects in the evaluations of $C$ and $A$ are almost similar. In both the cases, the core-

| Off-diagonal Matrix | DF | CCSD$_{t3}$ |
|---------------------|----|-------------|
| $\langle 7p_{1/2}|T^{(1)}|7p_{3/2}\rangle$ | -67.42 | -32.78 |
| $\langle 8p_{1/2}|T^{(1)}|8p_{3/2}\rangle$ | -24.35 | -10.44 |
| $\langle 6d_{3/2}|T^{(1)}|6d_{5/2}\rangle$ | -16.12 | -790.87 |
| $\langle 7d_{3/2}|T^{(1)}|7d_{5/2}\rangle$ | 7.93 | 238.11 |
| $\langle 7p_{1/2}|T^{(2)}|7p_{3/2}\rangle$ | -750.86 | -1588.40 |
| $\langle 8p_{1/2}|T^{(2)}|8p_{3/2}\rangle$ | -271.23 | -501.30 |
| $\langle 6d_{3/2}|T^{(2)}|6d_{5/2}\rangle$ | -63.01 | -253.07 |
| $\langle 7d_{3/2}|T^{(2)}|7d_{5/2}\rangle$ | 31.03 | 76.17 |
polarization effects in the $D_{5/2}$ states are found to be extremely large and opposite than their DF results. This may be due to the fact that single particle expressions given in Eqs. (19) and (21) are similar except different powers on $r$. Now from this analysis, it is obvious that the $C$ values are about at least four orders smaller than the $A$ and $B$ values in $211^{\text{Fr}}$. This means that measurements of the hyperfine splittings in the $J > 1/2$ states of this isotope need to be measured till the fifth decimal place accuracies in case we aim to infer $\Omega$ from these measurements. Also comparing to the magnitudes of the estimated $C$ values, we find $C$ values are in particularly large in the $7p \ 2P_{3/2}$, $8p \ 2P_{3/2}$ and $6d \ 2D_{5/2}$ states.

Hyperfine energy level of a state cannot be measured directly, but in practice their differences are being measured. Thus, it is essential to identify suitable hyperfine transitions among as many as sub-states possible to extract out contributions from the individual multipole precisely. Keeping this in mind, we give explicit expressions with appropriate angular momentum coefficients for the energy differences ($\delta W^F_{J} - \delta W^F_{J'}$) between different hyperfine momenta (say $F$ and $F'$) for each $J$ - symmetry states in appendices. Although expressions for the $J = 1/2$ states may not be required in the estimations of the $C$ values, but they can be used to eliminate contributions from the off-diagonal matrix elements appearing through the second-order effects among the fine structure partners. It is also obvious that these off-diagonal elements are one of the the major systematics in the extractions of the $C$ values from the measured hyperfine splittings. Therefore, it is important to determine the off-diagonal matrix elements of the hyperfine interaction operators accurately. For the same purpose, we have also calculated these quantities using the DF and CCSD $\alpha_{3}$ methods and give them in Table III. Large differences between the results from these two methods imply that proper inclusion of the correlation effects are also crucial for their accurate evaluations. Though our derived expressions for the different $J$ states will be useful for estimating the $C$ values provided the hyperfine splittings can be measured very high precisely, however we would like to point out from the associated large angular momentum coefficients from the derivations given in the appendices that the energy differences $\delta W^{6-3}_{np3/2}$ and $\delta W^{7-2}_{nd5/2}$ of the $np \ 2P_{3/2}$ and $nd \ 2D_{5/2}$ hyperfine states seem to be more suitable for the unambiguous extractions of the $C$ values as they are free from the off-diagonal matrix element contributions.

V. CONCLUSION

We have studied hyperfine structures of many low-lying states in $211^{\text{Fr}}$ using a relativistic coupled-cluster theory in the singles, doubles and partial triples excitations approximation. Its nuclear quadrupole momentum value has been revised by combining our atomic calculations with the measured $B$ value of the $7p \ 2P_{3/2}$ state. We also find our calculated $A$ values are in very good agreement with the available experimental results. By estimating $\Omega$ value from the nuclear shell-model and using our calculations of $C/\Omega$, preliminary values of $C$ up to the $7d \ 2D_{5/2}$ low-lying state in $211^{\text{Fr}}$ are evaluated. We also give the off-diagonal matrix elements to determine the second order energy shifts due to the dipole-dipole and dipole-quadrupole hyperfine interactions, which are typically in the same order of magnitudes compared to the contributions from the $C$ values. From all these analysis, we find that hyperfine energy differences between the $F = 6$ to $F = 3$ hyperfine levels in the $np \ 2P_{3/2}$ states and between the $F = 7$ to $F = 2$ hyperfine levels in the $nd \ 2D_{5/2}$ states are most suitable to infer their corresponding $C$ values unambiguously; thusly $\Omega$ of $211^{\text{Fr}}$. Nevertheless, our derivations on the hyperfine splittings for all the important low-lying states will be very useful when the measurements are carried over to extract out the $A$, $B$ and $C$ constants in the hyperfine transitions of $211^{\text{Fr}}$.

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Appendices

Here, we express hyperfine structure splittings of different atomic states in terms of $A$, $B$, $C$ and the corresponding off-diagonal $\eta$ and $\zeta$ coefficients. Splittings for the states with same $J$ and parity but having different principal quantum numbers, denoted by an index $n$, are given in general forms. For $211^{\text{Fr}}$, hyperfine state angular momenta are determined by considering the nuclear spin $I = 9/2$. Again, we account off-diagonal contributions only from the fine structure splittings and neglect contributions from other higher states owing to large energy denominators associated with those intermediate states.

Appendix A: $[(n-1)p^6] \ ns^2S_{1/2}$ state

For the $ns \ 2S_{1/2}$ states, $J = 1/2$. Thus, it yields $F = 4, 5$ and

$$W^{(1)}_{4,ns} = -\frac{11}{4} A^{ns} \quad \text{and} \quad W^{(1)}_{5,ns} = \frac{9}{4} A^{ns}. \quad (A1)$$

Using these relations, it yields

$$\delta W^{5-4}_{ns} = W^{(1)}_{5,ns} - W^{(1)}_{4,ns} = 5A^{ns}. \quad (A2)$$
Appendix B: \([(n-1)p_6] np^2 P_{1/2} \) state

In this case also, we have \( J = 1/2 \) and \( F = 4, 5 \). Thus, the first order effects are given by

\[
W^{(1)}_{4, np_{1/2}} = -\frac{11}{4} A_{np_{1/2}} \quad \text{and} \quad W^{(1)}_{5, np_{1/2}} = \frac{9}{4} A_{np_{1/2}} \tag{B1}
\]

The \( np_{1/2} \) states can have second order effect owing to small energy differences with their fine structure levels and can be given by

\[
W^{(2)}_{4, np_{1/2}} = (11/3) g^I_1 \left\{ \frac{J_{np_{3/2}} | T^{(1)}_e | J_{np_{1/2}} \rangle^2}{E_{np_{1/2}} - E_{np_{3/2}}} \right\} - \frac{(33/2) \sqrt{1/15} g_1 Q \langle J_{np_{3/2}} | T^{(1)}_e | J_{np_{1/2}} \rangle^2}{E_{np_{1/2}} - E_{np_{3/2}}} \times \frac{\langle J_{np_{1/2}} | T^{(2)}_e | J_{np_{3/2}} \rangle}{E_{np_{1/2}} - E_{np_{3/2}}} \tag{B2}
\]

and

\[
W^{(2)}_{5, np_{1/2}} = (9/2) g^I_1 \left\{ \frac{J_{np_{3/2}} | T^{(1)}_e | J_{np_{1/2}} \rangle^2}{E_{np_{1/2}} - E_{np_{3/2}}} \right\} + \frac{(9/2) \sqrt{3/5} g_1 Q \langle J_{np_{3/2}} | T^{(1)}_e | J_{np_{1/2}} \rangle^2}{E_{np_{1/2}} - E_{np_{3/2}}} \times \frac{\langle J_{np_{1/2}} | T^{(2)}_e | J_{np_{3/2}} \rangle}{E_{np_{1/2}} - E_{np_{3/2}}} \tag{B3}
\]

This gives us

\[
\delta W^{5-4}_{np_{1/2}} = W^{(1)}_{5, np_{1/2}} - W^{(1)}_{4, np_{1/2}} + W^{(2)}_{5, np_{1/2}} - W^{(2)}_{4, np_{1/2}} = 5 A_{np_{1/2}} + (5/6) g^I_1 \left\{ \frac{J_{np_{3/2}} | T^{(1)}_e | J_{np_{1/2}} \rangle^2}{E_{np_{1/2}} - E_{np_{3/2}}} \right\} + 10 \frac{\sqrt{3/5} g_1 Q \langle J_{np_{3/2}} | T^{(1)}_e | J_{np_{1/2}} \rangle^2}{E_{np_{1/2}} - E_{np_{3/2}}} \times \frac{\langle J_{np_{1/2}} | T^{(2)}_e | J_{np_{3/2}} \rangle}{E_{np_{1/2}} - E_{np_{3/2}}} \tag{B4}
\]

Appendix C: \([(n-1)p_6] np^2 P_{3/2} \) state

Considering \( J = 3/2 \) and \( I = 9/2 \), we have \( F = 3, 4, 5, 6 \). This gives us the first order splittings as

\[
W^{(1)}_{3, np_{3/2}} = -(33/4) A_{np_{3/2}} + (11/24) B_{np_{3/2}} - (143/42) C_{np_{3/2}}\),
\[
W^{(1)}_{4, np_{3/2}} = -(17/4) A_{np_{3/2}} - (5/24) B_{np_{3/2}} + (13/2) C_{np_{3/2}}\),
\[
W^{(1)}_{5, np_{3/2}} = (3/4) A_{np_{3/2}} - (5/12) B_{np_{3/2}} - (13/3) C_{np_{3/2}}\)
\[
W^{(1)}_{6, np_{3/2}} = (27/4) A_{np_{3/2}} + (1/4) B_{np_{3/2}} + C_{np_{3/2}} \tag{C1}
\]

and the second order splittings are

\[
W^{(2)}_{3, np_{3/2}} = 0,
\]
\[
W^{(2)}_{4, np_{3/2}} = (11/3) g^I_1 \left\{ \frac{J_{np_{1/2}} | T^{(1)}_e | J_{np_{3/2}} \rangle^2}{E_{np_{3/2}} - E_{np_{1/2}}} \right\} - \frac{(33/2) \sqrt{1/15} g_1 Q \langle J_{np_{1/2}} | T^{(1)}_e | J_{np_{3/2}} \rangle^2}{E_{np_{3/2}} - E_{np_{1/2}}} \times \frac{\langle J_{np_{1/2}} | T^{(2)}_e | J_{np_{3/2}} \rangle}{E_{np_{3/2}} - E_{np_{1/2}}} ,
\]
\[
W^{(2)}_{5, np_{3/2}} = (9/2) g^I_1 \left\{ \frac{J_{np_{1/2}} | T^{(1)}_e | J_{np_{3/2}} \rangle^2}{E_{np_{3/2}} - E_{np_{1/2}}} \right\} + \frac{(9/2) \sqrt{3/5} g_1 Q \langle J_{np_{1/2}} | T^{(1)}_e | J_{np_{3/2}} \rangle^2}{E_{np_{3/2}} - E_{np_{1/2}}} \times \frac{\langle J_{np_{1/2}} | T^{(2)}_e | J_{np_{3/2}} \rangle}{E_{np_{3/2}} - E_{np_{1/2}}} \tag{C2}
\]

Thus, it implies that

\[
\delta W^{5-4}_{np_{3/2}} = W^{(1)}_{5, np_{3/2}} - W^{(1)}_{4, np_{3/2}} + W^{(2)}_{5, np_{3/2}} - W^{(2)}_{4, np_{3/2}} = 5 A_{np_{3/2}} - \frac{(5/24) B_{np_{3/2}} - (65/6) C_{np_{3/2}}}{E_{np_{3/2}} - E_{np_{1/2}}} + \frac{(5/6) g^I_1 \left\{ \frac{J_{np_{1/2}} | T^{(1)}_e | J_{np_{3/2}} \rangle^2}{E_{np_{3/2}} - E_{np_{1/2}}} \right\}}{E_{np_{3/2}} - E_{np_{1/2}}} \\
+ \frac{\sqrt{3/5} g_1 Q \langle J_{np_{1/2}} | T^{(1)}_e | J_{np_{3/2}} \rangle^2}{E_{np_{3/2}} - E_{np_{1/2}}} \times \frac{\langle J_{np_{1/2}} | T^{(2)}_e | J_{np_{3/2}} \rangle}{E_{np_{3/2}} - E_{np_{1/2}}} \tag{C3}
\]

\[
\delta W^{6-5}_{np_{3/2}} = W^{(1)}_{6, np_{3/2}} - W^{(1)}_{5, np_{3/2}} + W^{(2)}_{6, np_{3/2}} - W^{(2)}_{5, np_{3/2}} = 6 A_{np_{3/2}} + \frac{(2/3) B_{np_{3/2}} + (16/3) C_{np_{3/2}}}{E_{np_{3/2}} - E_{np_{1/2}}} - \frac{(9/2) g^I_1 \left\{ \frac{J_{np_{1/2}} | T^{(1)}_e | J_{np_{3/2}} \rangle^2}{E_{np_{3/2}} - E_{np_{1/2}}} \right\}}{E_{np_{3/2}} - E_{np_{1/2}}} \\
- \frac{(9/2) \sqrt{3/5} g_1 Q \langle J_{np_{1/2}} | T^{(1)}_e | J_{np_{3/2}} \rangle^2}{E_{np_{3/2}} - E_{np_{1/2}}} \times \frac{\langle J_{np_{1/2}} | T^{(2)}_e | J_{np_{3/2}} \rangle}{E_{np_{3/2}} - E_{np_{1/2}}} \tag{C3}
\]

and

\[
\delta W^{6-3}_{np_{3/2}} = W^{(1)}_{6, np_{3/2}} - W^{(1)}_{3, np_{3/2}} = 15 A_{np_{3/2}} - \frac{(5/24) B_{np_{3/2}} + (185/42) C_{np_{3/2}}}{E_{np_{3/2}} - E_{np_{1/2}}} \tag{C3}
\]

This gives us
Appendix D: \([np^6]\) \(nd^2D_{3/2}\) state

Since \(J = 3/2\) and \(I = 9/2\), it follows \(F = 3, 4, 5, 6\) for which we have
\[
W_{3, nd3/2}^{(1)} = -(33/4)A_{nd3/2} + (11/24)B_{nd3/2} - (143/42)C_{nd3/2},
\]
\[
W_{4, nd3/2}^{(1)} = -(17/4)A_{nd3/2} - (5/24)B_{nd3/2} + (13/2)C_{nd3/2},
\]
\[
W_{5, nd3/2}^{(1)} = (3/4)A_{nd3/2} - (5/12)B_{nd3/2} - (13/3)C_{nd3/2},
\]
\[
W_{6, nd3/2}^{(1)} = (27/4)A_{nd3/2} + (1/4)B_{nd3/2} + C_{nd3/2},
\]
and
\[
W_{3, nd3/2}^{(2)} = (11/10)g_J^2\frac{|\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle|^2}{E_{nd3/2} - E_{nd5/2}}
- (99/20)\sqrt{7}g_J Q\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle
\times \frac{\langle J_{nd5/2}|T_e^{(2)}|J_{nd3/2}\rangle}{E_{nd3/2} - E_{nd5/2}},
\]
\[
W_{4, nd3/2}^{(2)} = (21/10)g_J^2\frac{|\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle|^2}{E_{nd3/2} - E_{nd5/2}}
- (3/4)\sqrt{7}g_J Q\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle
\times \frac{\langle J_{nd5/2}|T_e^{(2)}|J_{nd3/2}\rangle}{E_{nd3/2} - E_{nd5/2}},
\]
\[
W_{5, nd3/2}^{(2)} = (13/5)g_J^2\frac{2|\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle|^2}{E_{nd3/2} - E_{nd5/2}}
\times \frac{\langle J_{nd5/2}|T_e^{(2)}|J_{nd3/2}\rangle}{E_{nd3/2} - E_{nd5/2}},
\]
\[
W_{6, nd3/2}^{(2)} = (21/10)g_J^2\frac{|\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle|^2}{E_{nd3/2} - E_{nd5/2}}
+ (9/10)\sqrt{7}g_J Q\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle
\times \frac{\langle J_{nd5/2}|T_e^{(2)}|J_{nd3/2}\rangle}{E_{nd3/2} - E_{nd5/2}}.
\]

This gives us
\[
\delta W_{nd3/2}^{4-3} = W_{4, nd3/2}^{(1)} - W_{3, nd3/2}^{(1)} + W_{4, nd3/2}^{(2)} - W_{3, nd3/2}^{(2)}
= 4A_{nd3/2} - (2/3)B_{nd3/2} + (208/21)C_{nd3/2}
+ g_J^2\frac{2|\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle|^2}{E_{nd3/2} - E_{nd5/2}}
- (3/10)\sqrt{7}g_J Q\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle
\times \frac{\langle J_{nd5/2}|T_e^{(2)}|J_{nd3/2}\rangle}{E_{nd3/2} - E_{nd5/2}},
\]
\[
\delta W_{nd3/2}^{5-4} = W_{5, nd3/2}^{(1)} - W_{4, nd3/2}^{(1)} + W_{5, nd3/2}^{(2)} - W_{4, nd3/2}^{(2)}
= 5A_{nd3/2} - (5/24)B_{nd3/2} - (65/6)C_{nd3/2}
+ (1/2)g_J^2\frac{|\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle|^2}{E_{nd3/2} - E_{nd5/2}}
+ (3/4)\sqrt{7}g_J Q\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle
\times \frac{\langle J_{nd5/2}|T_e^{(2)}|J_{nd3/2}\rangle}{E_{nd3/2} - E_{nd5/2}},
\]
and
\[
\delta W_{nd3/2}^{6-5} = W_{6, nd3/2}^{(1)} - W_{5, nd3/2}^{(1)} + W_{6, nd3/2}^{(2)} - W_{5, nd3/2}^{(2)}
= 6A_{nd3/2} + (2/3)B_{nd3/2} + (16/3)C_{nd3/2}
- (1/2)g_J^2\frac{|\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle|^2}{E_{nd3/2} - E_{nd5/2}}
+ (9/10)\sqrt{7}g_J Q\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle
\times \frac{\langle J_{nd5/2}|T_e^{(2)}|J_{nd3/2}\rangle}{E_{nd3/2} - E_{nd5/2}}.
\]

Appendix E: \([np^6]\) \(nd^2D_{3/2}\) state

For \(J = 5/2\) and \(I = 9/2\), we now have \(F = 2, 3, 4, 5, 6, 7\). This gives many hyperfine levels for which it yields
\[
W_{2, nd5/2}^{(1)} = -(55/4)A_{nd5/2} + (11/24)B_{nd5/2}
- (143/42)C_{nd5/2},
\]
\[
W_{3, nd5/2}^{(1)} = -(43/4)A_{nd5/2} + (19/120)B_{nd5/2}
+ (221/210)C_{nd5/2},
\]
\[
W_{4, nd5/2}^{(1)} = -(27/4)A_{nd5/2} - (1/8)B_{nd5/2}
+ (559/210)C_{nd5/2},
\]
\[
W_{5, nd5/2}^{(1)} = -(7/4)A_{nd5/2} - (7/24)B_{nd5/2}
+ (119/210)C_{nd5/2},
\]
\[
W_{6, nd5/2}^{(1)} = -(17/4)A_{nd5/2} - (13/60)B_{nd5/2}
- (41/15)C_{nd5/2},
\]
\[
W_{7, nd5/2}^{(1)} = (45/4)A_{nd5/2} + (1/4)B_{nd5/2}
+ C_{nd5/2},
\]
and
\[
W_{2, nd5/2}^{(2)} = 0,
\]
\[
W_{3, nd5/2}^{(2)} = (11/10)g_J^2\frac{2|\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle|^2}{E_{nd3/2} - E_{nd5/2}}
- (99/20)\sqrt{7}g_J Q\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle
\times \frac{\langle J_{nd3/2}|T_e^{(2)}|J_{nd5/2}\rangle}{E_{nd3/2} - E_{nd5/2}},
\]
\[
W_{4, nd5/2}^{(2)} = (21/10)g_J^2\frac{2|\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle|^2}{E_{nd3/2} - E_{nd5/2}}
- (3/4)\sqrt{7}g_J Q\langle J_{nd5/2}\rangle|T_e^{(1)}|J_{nd3/2}\rangle
\times \frac{\langle J_{nd3/2}|T_e^{(2)}|J_{nd5/2}\rangle}{E_{nd3/2} - E_{nd5/2}}.
\]
\[
\begin{align*}
\delta W^{5-6}_{\text{nd}5/2} &= W^{(1)}_{\text{6,nd}5/2} - W^{(1)}_{\text{5,nd}5/2} + W^{(2)}_{\text{5,nd}5/2} - W^{(2)}_{\text{6,nd}5/2} \\
&= 6A_{\text{nd}5/2} + (3/40)B^{\text{nd}5/2} - (33/10)C^{\text{nd}5/2} \\
&\quad - (1/2)g_7^2 \left\langle \left| J_{\text{nd}3/2} \right| T_e^{(1)} \left| J_{\text{nd}5/2} \right\rangle \right\rangle \\
&\quad \times \left\langle J_{\text{nd}3/2} \right| T_e^{(2)} \left| J_{\text{nd}5/2} \right\rangle \\
&\quad \times \left\langle J_{\text{nd}3/2} \right| T_e^{(1)} \left| J_{\text{nd}5/2} \right\rangle \\
\end{align*}
\]

This corresponds to
\[
\begin{align*}
\delta W^{3-2}_{\text{nd}5/2} &= W^{(1)}_{\text{3,nd}5/2} - W^{(1)}_{\text{2,nd}5/2} + W^{(2)}_{\text{2,nd}5/2} - W^{(2)}_{\text{3,nd}5/2} \\
&= 3A_{\text{nd}5/2} - (3/10)B^{\text{nd}5/2} + (156/35)C^{\text{nd}5/2} \\
&\quad + (11/10)g_7^2 \left\langle \left| J_{\text{nd}3/2} \right| T_e^{(1)} \left| J_{\text{nd}5/2} \right\rangle \right\rangle \\
&\quad \times \left\langle J_{\text{nd}3/2} \right| T_e^{(2)} \left| J_{\text{nd}5/2} \right\rangle \\
\delta W^{4-3}_{\text{nd}5/2} &= W^{(1)}_{\text{3,nd}5/2} - W^{(1)}_{\text{4,nd}5/2} + W^{(2)}_{\text{4,nd}5/2} - W^{(2)}_{\text{3,nd}5/2} \\
&= 4A_{\text{nd}5/2} - (17/60)B^{\text{nd}5/2} + (169/105)C^{\text{nd}5/2} \\
&\quad + g_7^2 \left\langle \left| J_{\text{nd}3/2} \right| T_e^{(1)} \left| J_{\text{nd}5/2} \right\rangle \right\rangle \\
&\quad \times \left\langle J_{\text{nd}3/2} \right| T_e^{(2)} \left| J_{\text{nd}5/2} \right\rangle \\
\delta W^{5-4}_{\text{nd}5/2} &= W^{(1)}_{\text{4,nd}5/2} - W^{(1)}_{\text{5,nd}5/2} + W^{(2)}_{\text{5,nd}5/2} - W^{(2)}_{\text{4,nd}5/2} \\
&= 5A_{\text{nd}5/2} - (1/6)B^{\text{nd}5/2} - (44/21)C^{\text{nd}5/2} \\
&\quad + (1/2)g_7^2 \left\langle \left| J_{\text{nd}3/2} \right| T_e^{(1)} \left| J_{\text{nd}5/2} \right\rangle \right\rangle \\
&\quad \times \left\langle J_{\text{nd}3/2} \right| T_e^{(2)} \left| J_{\text{nd}5/2} \right\rangle \\
\end{align*}
\]

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