I. INTRODUCTION

In the recent study of the hyperfine structure of the 6p_{3/2} state of 133Cs by Gerginov et al., intervals between hyperfine levels were measured to an accuracy of ≤ 2 kHz, which was sufficient to give, for the first time, a non-zero value for the c hyperfine constant. The value of the nuclear octupole moment of 133Cs obtained from c was Ω = 0.82(10) b × μN, which is about a factor of 40 larger than nuclear shell-model prediction Ω_{n.s.m} = 0.022 b × μN, motivating a re-examination of corrections to the hyperfine constants. One such correction is the second-order hyperfine interaction between the 6p_{3/2} and 6p_{1/2} states. An estimate of this correction, based on an independent particle model of the cesium atom, was used in [1]. In the present work, we carry out a detailed third-order MBPT calculation and obtain corrections to the 6p_{3/2} hyperfine levels that are a factor of 10 smaller than the values used in [1]. Revised values of the hyperfine constants a, b, and c, obtained using the present results for the second-order hyperfine energies agree with those reported in [1] to within the error estimates. However, for future experiments, especially experiments aimed at a precision of better than 1 kHz, it will be important to use the correlated values of the corrections presented here, rather than the larger values given in [1].

II. PERTURBATION EXPANSION

We write the hyperfine interaction in the form

$$H_{hf} = \sum_{k\lambda} (-1)^k T^{(k)}_{-\lambda} M^{(k)}_{\lambda},$$

where $T^{(k)}_{-\lambda}$ is an irreducible tensor operator acting in the electron sector and $M^{(k)}_{\lambda}$ is an irreducible tensor operator acting in the nuclear sector. The first-order hyperfine correction to the energy of a state $|1\rangle$ is

$$W^{(1)}_F = \langle 1 \vert H_{hf} \vert 1 \rangle = \sum_k (-1)^I + J + F \left\{ \begin{array}{ccc} J & I & F \\ I & J & k \end{array} \right\} \frac{1}{\|J\|} \frac{1}{\|I\|} \frac{1}{\|M^{(k)}\|} \langle I \vert M^{(k)} \vert I \rangle.$$

The nuclear matrix elements are given in terms of conventional nuclear moments through

$$\langle JJ \vert M^{(1)}_0 \vert JJ \rangle = \mu,$n
$$\langle JJ \vert M^{(2)}_0 \vert JJ \rangle = \frac{1}{2} Q,$n
$$\langle JJ \vert M^{(3)}_0 \vert JJ \rangle = -\Omega.$n

Here, $\mu$ is the nuclear magnetic dipole moment, $Q$ is the nuclear electric quadrupole moment, and $\Omega$ is the nuclear magnetic octupole moment.

Introducing the conventional hyperfine constants a, b, and c through the relations:

$$a = \frac{\mu}{I J} \langle JJ \vert T^{(1)}_0 \vert JJ \rangle,$n
$$b = 2 Q \langle JJ \vert T^{(2)}_0 \vert JJ \rangle,$n
$$c = -\Omega \langle JJ \vert T^{(3)}_0 \vert JJ \rangle,$n

we get

$$\langle JJ \vert T^{(k)}_0 \vert JJ \rangle = \sum_{\lambda=\pm 1} M^{(k)}_{\lambda} T^{(k)}_{-\lambda}.$n

The nuclear moments are given in terms of conventional nuclear moments through

$$\langle JJ \vert M^{(k)}_0 \vert JJ \rangle = M^{(k)}_{\lambda} T^{(k)}_{-\lambda}.$n

The factor of 10 difference has negligible effect on the conclusions of the recent experiment but will become important for experiments carried out at a precision of better than 1 kHz.
and write the first-order hyperfine energy as

\[
W_F^{(1)} = \frac{1}{2} K a + \frac{3K_+ - 4J_+ I_+}{8(2I - 1)(2J - 1)} b \\
+ \frac{5K^2(K + 4) - 4K[3J_+ I_+ - J_+ - I_+ - 3] - 20J_+ I_+}{I(2I - 1)(2J - 1)(2J - 2)} c,
\]

(4)

where \( I_+ = I(I + 1), J_+ = J(J + 1), F_+ = F(F + 1), K = F_+ - J_+ - I_+, \) and \( K_+ = K(K + 1). \)

The second-order (in the hyperfine interaction) energy of a state is given by

\[
W_F^{(2)} = \sum_{n \neq 1} \frac{\langle 1|H_{hf}|n\rangle \langle n|H_{hf}|1\rangle}{E_1 - E_n}.
\]

For the state \(|1\rangle = |6p_{3/2}\rangle\) of Cs, the second-order hyperfine energy is dominated by the single state \(|n\rangle = |2\rangle = |6p_{3/2}\rangle\). Moreover, the largest contribution from this state is the one associated with the magnetic dipole term \(k = k' = 1\). After angular reduction, the second-order energy is

\[
W_F^{(2)} = \left\{ \begin{array}{ll}
J_2 & I \\
I & J_1
\end{array} \right\} \frac{2}{E_1 - E_2} \frac{\langle I_2|T^{(1)}|I_1\rangle^2 \langle I|M^{(1)}|I\rangle^2}{E_1 - E_2}.
\]

(5)

Contributions to the second-order energy from the nuclear quadrupole moment have been evaluated in lowest order and found to change the values obtained from Eq. (5) by less than 1%. The fine-structure interval \(E_1 - E_2\) in the denominator is determined as the difference between \(f_{02}\) the centroid of the \(6s^2S_{1/2} \rightarrow 6p^2P_{3/2}\) transition \(2, 3\) and \(f_{01}\) the centroid of the \(6s^2S_{1/2} \rightarrow 6p^2P_{1/2}\) transition \(4\), both of which have been measured to high precision. One obtains \(E_1 - E_2 = f_{02} - f_{01} = 1.660969667(11) \times 10^6\) MHz.

### III. NUMERICAL ESTIMATES

Correlation corrections to hyperfine matrix elements in alkali-metal atoms are large. Thus, for example, a lowest-order Dirac-Hartree-Fock calculation of the hyperfine constant \(a_{3/2}\) for the \(6p_{3/2}\) state of Cs (which is proportional to the diagonal matrix element \(\langle 3/2|T^{(1)}|3/2\rangle\)) leads to a result that is a factor of two smaller than the experimental value. One expects (and indeed finds) corrections of a similar size for the off-diagonal matrix element \(\langle 1/2|T^{(1)}|3/2\rangle\) appearing in the numerator of the expression for the second-order hyperfine energy. If we assume that the relative size of the correlation corrections to the two matrix elements mentioned above are the same, then we can determine the ratio \(\langle 1/2|T^{(1)}|3/2\rangle / \langle 3/2|T^{(1)}|3/2\rangle\) by means of a lowest-order calculation and, using that ratio together with the experimental value of \(a_{3/2}\), obtain an accurate value for \(\langle 1/2|T^{(1)}|3/2\rangle\). That was the strategy used to obtain the values \(W_F^{(2)} = 401\) Hz and \(W_F^{(2)} = 520\) Hz for the \(6p_{3/2}\) state of \(^{133}\)Cs quoted in Ref. [1]. (The ratio of matrix elements was determined in the non-relativistic approximation and did not depend on details of the \(6p\) wave function. The non-relativistic approximation is not a serious problem, however, since the ratio obtained using relativistic Dirac-Hartree-Fock wave functions differs from the non-relativistic ratio by less than 5%).

### IV. CORRELATION CORRECTIONS

The estimates made in the previous section depend on the assumption that correlation corrections to reduced matrix elements of the hyperfine operator \(\langle j|T^{(1)}|j'\rangle\) are independent of the total angular momentum \(j\) of the \(6p_j\) state. To test that assumption, we carried out correlated third-order MBPT calculations of the three \(6p_j\) matrix elements.

In Table I we give a detailed breakdown of contributions to the third-order matrix elements. Formulas for the first-, second-, and third-order matrix element are given in [1]. We use a modified version of these formulas in which: (a) the sum of the second-order matrix element and the third-order contribution to the random-phase approximation (RPA) is replaced by the exact solution to the RPA equations, and (b) all one-electron matrix elements in third-order are replaced by their RPA counterparts. These modifications give dipole transition matrix elements that are gauge invariant in second- and third-order [6]. The third-order hyperfine constants for the \(6p_{1/2}\) and \(6p_{3/2}\) states evaluated in this way are within a few percent of experiment. Since we use the same method to evaluate diagonal and off-diagonal matrix elements, we expect the third order off-diagonal matrix element to be accurate to a few percent.

Substituting the third-order off-diagonal matrix element given in Table I into Eq. (6), we find \(W_F^{(2)} = 37.3\) Hz and \(W_F^{(2)} = 48.3\) Hz for the \(6p_{3/2}\) state. Combining the second-order corrections with the observed \(6p_{3/2}\)
hyperfine intervals (MHz) from [1]

\[
\begin{align*}
W_5 - W_4 &= 251.0916(20) \\
W_4 - W_3 &= 201.2871(11) \\
W_3 - W_2 &= 151.2247(16),
\end{align*}
\]

we obtain the following values for the hyperfine constants (MHz)

\[
\begin{align*}
a &= 50.28825(23) \\
b &= -0.4940(17) \\
c &= 0.00056(7).
\end{align*}
\]

These values agree within error limits with those found in [1].

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