Hyperfine structure of the $2^3P$ state in $^9\text{Be}$ and the nuclear quadrupole moment

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We have performed accurate calculations of the hyperfine structure of the $2^3P$ state in the $^9\text{Be}$ atom with the help of highly optimized, explicitly correlated functions, accounting for the leading finite nuclear mass, radiative, nuclear structure and relativistic effects. By comparison with measurements, we have determined the $^9\text{Be}$ nuclear quadrupole moment to be $Q_N = 0.05350(14)$ barns, which is not only the most accurate result, but also disagrees with previous determinations.

The electric quadrupole moment $Q_N$ of nuclei is a measure of the deformation of the nuclear charge distribution with respect to the spherical symmetry. This deformation comes from a strong dependence of nuclear forces on the orientation of the nucleus with respect to the spherical symmetry. This deformation comes from a strong dependence of nuclear forces on the orientation of the nucleon spin and is present in atomic nuclei with a spin equal to or greater than one, as it is for $^9\text{Be}$ with $I = 3/2$. In principle, $Q_N$ could be determined from the nuclear structure theory, but such calculations with controlled accuracy have not yet been feasible. Only very recently and only for such a simple nucleus as the deuteron, theoretical predictions based on the chiral effective field theory have reached the uncer-}

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with which are conventionally used to represent the measured values, while the fine structure is given in terms of differences in the centroid energies.

If the atomic levels had a definite value \( J \), the fine structure would be given by the expectation value of \( H_{\text{eff}} \), namely

\[
E_{\text{fs}}(J) = \begin{cases} 
 c_1 + c_2/6 & \text{for } J = 2 \\
 -c_1 - 5c_2/6 & \text{for } J = 1 \\
 -2c_1 + 5c_2/3 & \text{for } J = 0
\end{cases}
\]

(14)

and the hyperfine structure by

\[
A_J = \begin{cases} 
 a_1/2 + a_2/2 + a_3/6 & \text{for } J = 2 \\
 a_1/2 + a_2/2 - 5a_3/6 & \text{for } J = 1 \\
 b & \text{for } J = 1
\end{cases}
\]

(15)

Because, in general, one cannot assume that the levels have a definite value of \( J \), the effective hfs Hamiltonian in Eq. (12) has to be diagonalized numerically. Nonetheless, the hyperfine structure can still be represented in terms of \( A_J \) and \( B_J \) coefficients, and we use them for comparison of theoretical predictions with experimental results and for the determination of the nuclear quadrupole moment \( Q_{\text{N}} \).

**Relativistic, radiative, and finite nuclear size corrections.—**

The hyperfine Hamiltonian in Eq. (5) represents the leading hyperfine interactions, but there are also many small corrections which are often overlooked in literature. These corrections contain terms with higher powers of the fine structure constant \( \alpha \). Because most of them are proportional to the Fermi contact interaction, we account for them in terms of the following factor

\[
\tilde{a}_1 = a_1(1 + \epsilon).
\]

(16)

Below, we briefly describe contributions included in the \( \epsilon \) term.

The \( O(\alpha) \) correction is analogous to that in hydrogenic systems \([17]\) and consists of two parts. The first part is due to the nuclear recoil

\[
H_{\text{rec}}^{(5)} = \left[ -3\frac{Z\alpha}{\pi} \frac{m}{m_{\text{N}}} \ln\left(\frac{m_{\text{N}}}{m}\right) \right] \sum_a 2 \frac{Z\alpha g_{N}}{3 m_{\text{N}}} \hat{\sigma}_a \pi \delta^3(\vec{r}_a)
\]

(17)

and numerically is very small, almost negligible. The recoil contribution to \( \epsilon \) amounts to \( \epsilon_{\text{rec}} = -0.000011 \). The second part of the \( O(\alpha) \) correction is due to the finite nuclear size and the nuclear polarizability, and is given by \([17][18]\)

\[
H_{\text{fs}}^{(5)} = \left[ -2Z\alpha ma_r \frac{2}{3} \sum_a \frac{Z\alpha g_{N}}{m_{\text{N}}} \hat{\sigma}_a \pi \delta^3(\vec{r}_a) \right]
\]

(18)

where \( r_z \) is a kind of effective nuclear radius called the Zemach radius. Disregarding the inelastic effects, this radius can be written down in terms of the electric charge \( \rho_E \) and magnetic-moment \( \rho_M \) densities as

\[
r_z = \int d^3r d^3r' \rho_E(r') \rho_M(r') |\vec{r} - \vec{r}'|.
\]

(19)
Nevertheless, the inelastic, i.e. polarizability, corrections can be significant, but because they are very difficult to calculate, they are usually neglected. In this work we account for possible inelastic effects by employing \( r_{2z} \) from a comparison of very accurate calculations of hfs in \(^{8}\text{Be}^+\) with the experimental value, namely \( r_{2z} = 4.07(5) \) fm \(^{18}\). Because this correction is also proportional to the contact Fermi interaction, we represent it in terms of \( \epsilon_{\text{fs}} = -0.000 615 \).

Next, there are radiative and relativistic corrections of the relative order \( O(\alpha^2) \). The radiative correction, beyond that included by the free electron g-factor, is \(^{17}\)

\[
H_{\text{rad}}^{(6)} = Z \alpha^2 \left( \ln 2 - \frac{5}{2} \right) \sum_n 2 \sum_{\nu} \frac{Z \alpha \beta_{\nu}}{m \nu N} \sigma_\nu \pi \delta(r_n) \tag{20}
\]

and the corresponding \( \epsilon \) factor is \( \epsilon_{\text{rad}} = -0.000 384 \). The \( O(\alpha^2) \) relativistic and higher order corrections are much more complicated. They have been calculated for the ground state of \(^{8}\text{Be}^+\) \(^{18}\). Here we take this result and assume that it is proportional to the Fermi contact interaction, and obtain \( \epsilon_{\text{rel}} = 0.001 664 \). This is the only approximation we assume in this work, and as a consequence we neglect the mixing of the \( 2^3P_1 \) state with the nearby lying \( 2^1P_1 \). Exactly for this reason we will use only the hyperfine splitting of the \( 2^3P_2 \) state, which does not mix with \( 2^1P_1 \) for the determination of \( Q_N \). The resulting total correction is

\[
\epsilon = \epsilon_{\text{rel}} + \epsilon_{\text{fs}} + \epsilon_{\text{rad}} + \epsilon_{\text{rel}} = 0.654 \cdot 10^{-3}. \tag{21}
\]

Some previous works present these multiplicative correction for all individual hyperfine contributions, but in our opinion this cannot be fully correct because higher order relativistic corrections may include additional terms, beyond that in \( H_{\text{eff}} \) in Eq. \((12)\). These corrections are expected to be much smaller than the experimental uncertainty for the \( B_J \) coefficient, and therefore are neglected.

**ECG wave function and expectation values.**—Let us now move to the calculations of the \( a_i, b_i, \) and \( c_i \) coefficients. To obtain sufficiently high accuracy for these parameters, we express the four-electron atomic wave function as a linear combination of properly symmetrized explicitly correlated Gaussian (ECG) functions,

\[
\Psi(\{r_n\}) = \sum_{n=1}^{K} t_n A \left[ \phi_n(\{r_n\}) \chi_{\{a\}} \right], \tag{22}
\]

where \( t_n \) are linear coefficients, \( A \) is the antisymmetrization operator over electronic indices, and \( \{a\} \) and \( \{r_n\} \) denote the sequence of electron indices and coordinates, respectively. The electronic \( P \) symmetry of the states was enforced using the following spatial functions

\[
\phi^{L}(\{r_n\}) = r_n^L \exp \left[ - \sum_{b} \nu_b r_b^2 - \sum_{c<d} u_{cd} r_{cd}^2 \right] \tag{23}
\]

with \( \nu_b \) and \( u_{cd} \) being nonlinear variational parameters, while the spin functions corresponding to different spin \( S = 1 \) projections were, for \( \{a\} = (1, 2, 3, 4) \):

\[
\chi_{\{a\}}^{1-1} = \frac{1}{\sqrt{2}} (\alpha_1 \beta_2 - \beta_1 \alpha_2) \beta_3 \beta_4, \tag{24}
\]

\[
\chi_{\{a\}}^{10} = \frac{1}{2} (\alpha_1 \beta_2 - \beta_1 \alpha_2) (\alpha_3 \beta_4 - \beta_4 \alpha_3), \tag{25}
\]

\[
\chi_{\{a\}}^{11} = \frac{1}{\sqrt{2}} (\alpha_1 \beta_2 - \beta_1 \alpha_2) \alpha_3 \alpha_4. \tag{26}
\]

To control the uncertainty of our results we performed the calculations with several basis sets successively increasing their size. The nonlinear parameters were optimized variationally with respect to the nonrelativistic energy \( E \) until the energy reached stability in a desired number of digits. From the analysis of convergence we obtained the extrapolated nonrelativistic energies and mean values used in Table \( I \). The largest wave functions optimized variationally were composed of 6144 terms, leading to the nonrelativistic energy in agreement with the result obtained by Kedziorz et al. \(^{19}\).

The optimized nonrelativistic wave functions were subsequently employed to evaluate the fine and hyperfine parameters from the expectation values of the corresponding quantum-mechanical operators \((2)-(10)\). For this purpose we chose \( J = 2, L = 1 \), and for an arbitrary \( M \)

\[
c_1 = \langle J, M | Q_1 | J, M \rangle, \tag{27}
\]

\[
c_2 = 6 \langle J, M | Q_2 | J, M \rangle, \tag{28}
\]

\[
a_1 = \langle J, M | \vec{Q}_1 \cdot \vec{J} | J, M \rangle / 3, \tag{29}
\]

\[
a_2 = \langle J, M | \vec{Q}_2 \cdot \vec{J} | J, M \rangle / 3, \tag{30}
\]

\[
a_3 = \langle J, M | \vec{Q}_3 \cdot \vec{J} | J, M \rangle, \tag{31}
\]

\[
b = \langle L, M | Q^{(J)} L^i L^j L | L, M \rangle / 5. \tag{32}
\]

The numerical values of these parameters and their convergence with increasing size of the ECG basis are shown in Table \( I \). Their relative accuracy can be estimated collectively as better than \( 6 \cdot 10^{-6} \).

**Fine and hyperfine structure.**—The hyperfine transition frequencies \( \nu_J(F; F+1) \) can be expressed in terms of the \( A_J \) and \( B_J \) parameters and vice versa. For this, one writes

\[
\langle H_{\text{hfs,eff}} \rangle_F = A_J A_{IJF} + B_J B_{IJF} + C_J C_{IJF} \tag{33}
\]

where

\[
A_{IJF} = \frac{1}{2} K, \tag{34}
\]

\[
B_{IJF} = \frac{3}{4} K (K + 1) - I (I + 1) J (J + 1) \tag{35}
\]

with \( K = F(F + 1) - I (I + 1) - J(J+1) \) and the total angular momentum \( \vec{F} = \vec{S} + \vec{L} + \vec{I} \). The octupole term \( C_{IJF} \) is given e.g. by Schwartz \(^{22}\) and Jaccarino \(^{23}\).

For \( J = 1 \) we arrive at

\[
A_1 = - \frac{1}{6} \nu_1 \left( \frac{1}{2} \frac{3}{2} \frac{3}{2} \right) - \frac{10}{3} \nu_1 \left( \frac{3}{2} \frac{3}{2} \frac{3}{2} \right), \tag{36}
\]

\[
B_1 = \frac{1}{6} \nu_1 \left( \frac{1}{2} \frac{3}{2} \frac{3}{2} \right) - \frac{1}{5} \nu_1 \left( \frac{3}{2} \frac{3}{2} \frac{3}{2} \right), \tag{37}
\]

and for \( J = 2 \)

\[
A_2 = - \frac{3}{50} \nu_2 \left( \frac{1}{2} \frac{3}{2} \frac{3}{2} \right) - \frac{7}{50} \nu_2 \left( \frac{3}{2} \frac{3}{2} \frac{3}{2} \right) - \frac{4}{25} \nu_2 \left( \frac{5}{2} \frac{5}{2} \frac{5}{2} \right), \tag{38}
\]
TABLE I. Convergence of the nonrelativistic energy and theoretical fine and hyperfine structure parameters for the $^2P$ state of $^9$Be (in MHz). Mass $m_N = 9.01218307(8)$ u [20] and magnetic moment $\mu/\mu_N = -1.1774(3)$ [21] were used for the $^9$Be nucleus.

| $K$ | $E$/a.u. | $c_1$ | $c_2$ | $a_1$ | $a_2$ | $a_3$ | $b/Q_N$ (MHz/barn) |
|-----|----------|-------|-------|-------|-------|-------|---------------------|
| 1536 | 14.5663414086 $5$ | 32.986385 | 5.399.032 | -231.116.70 | -22.982.781 | 14.788.494 | 27.150.476 |
| 2048 | 14.5663414417 | 32.897.849 | 5.399.186 | -231.132.66 | -22.989.23 | 14.78.389 | 27.149.896 |
| 3072 | 14.5663413800 | 32.898.401 | 5.399.240 | -231.130.99 | -22.989.86 | 14.788.467 | 27.149.112 |
| 4096 | 14.5663414455 | 32.989.854 | 5.399.190 | -231.129.61 | -22.991.24 | 14.788.579 | 27.148.954 |
| 6144 | 14.5663414660 | 32.990.027 | 5.399.172 | -231.128.99 | -22.998.86 | 14.786.930 | 27.148.988 |
| $\infty$ | 14.566341474(8) | 32.990.2(2) | 5.399.15(2) | -231.128.4(6) | -22.999.24(5) | 14.786.68(5) | 27.148.87(3) |

\[
B_2 = \frac{2}{5} \nu_2 \left( \frac{1}{2} \frac{1}{2} \frac{1}{2} \frac{3}{2} \right) + \frac{2}{5} \nu_2 \left( \frac{3}{2} \frac{5}{2} \frac{5}{2} \frac{3}{2} \right) - \frac{16}{35} \nu_2 \left( \frac{5}{2} \frac{7}{2} \frac{5}{2} \frac{7}{2} \right) \quad (39)
\]

\[
C_2 = -\frac{1}{50} \nu_2 \left( \frac{1}{2} \frac{3}{2} \frac{1}{2} \frac{3}{2} \right) + \frac{1}{50} \nu_2 \left( \frac{3}{2} \frac{5}{2} \frac{3}{2} \frac{5}{2} \right) - \frac{1}{175} \nu_2 \left( \frac{5}{2} \frac{7}{2} \frac{5}{2} \frac{7}{2} \right). \quad (40)
\]

The above listed parameters [36]-[40] were evaluated using both experimental and theoretical transition frequencies $\nu_F (F; F + 1)$, and their numerical values are presented in Table II. The experimental values of $\nu_F$ (in MHz) were taken from Ref. [14] while theoretical ones were found by diagonalization of the effective Hamiltonian of Eq. [12]. Most importantly, the parameter $b$ was fixed by matching theoretical and experimental $B_2$, which holds for

\[
b = 1.4525(40) \text{ MHz}, \quad (41)
\]

where the uncertainty originates from that of the experimental one in $B_2$.

The differences between experimental and theoretical values, shown in Table III, are not discrepancies but result from neglected higher order relativistic and QED effects, which are beyond those proportional to the Fermi contact interaction. For the $A_2$ coefficient these effects are of relative order $0.07 \cdot 10^{-3}$, while for $A_1$ they are $0.33 \cdot 10^{-3}$. They are larger for $A_1$ due to hyperfine mixing of $2^1P_1$ with the nearby $2^1P_0$ state, which we have not taken into account in our calculation. Moreover, the relative difference for the $B_1$ coefficient is $\delta B_1/B_1 = 0.80 \cdot 10^{-3}$, which most probably is also due to this mixing. For this reason, we have chosen $B_2$ and not $B_1$ for the determination of the $b$ coefficient.

The centroids of the calculated hyperfine energy levels are shown in Table III in the column ‘fs levels’ for all $J$ values of the $^2P$ state. This table contains also experimental [14] and calculated fine-structure transition frequencies as well as their difference. This difference originates again from the higher order relativistic and QED effects. Due to a large experimental uncertainty we cannot conclude much about the magnitude of these effects.

**Determination of $Q_N( ^9$Be).**—Having calculated the value of $b/Q_N = 27.148 \pm 1(3) \text{ MHz/barn}$, see Tab. I and fixed $b$ by matching theoretical and experimental values of $B_2$, we obtain the electric nuclear quadrupole moment $Q_N$ of $^9$Be

\[
Q_N = 0.05358(14) \text{ barn}. \quad (42)
\]

The uncertainty in $Q_N$ comes from the experimental error in the $\nu_2$ hyperfine frequencies. In this context, the uncertainties from the neglected higher order relativistic and QED effects in the $b$ parameter, as well as the numerical uncertainties, are much smaller and are not shown.

Among the literature results listed in Tab. IV and depicted in Fig. 1 only the $Q_N$ reported by Beloy et al. [15], thanks to its large uncertainty, agrees with our quadrupole moment. The lack of agreement with the other previous studies indicates that it is very difficult to estimate theoretical uncertainties using well-established, atomic structure methods such as MCHF or CI+MBPT.

**Conclusions.**—We have determined the electric quadrupole moment $Q_N( ^9$Be) with significantly higher accuracy and in disagreement with previous values. The improvement achieved in this work has several independent sources. The first one is the recalculation of the $A_2$, $B_2$, and $C_2$ coefficients from experimental hyperfine splitting. More precisely, including $C_2$ in the effective Hamiltonian $H_{\text{hf eff}}$ in Eq. (13)
enabled us to decrease the experimental uncertainties by a factor of 2. The second improvement is due to the inclusion of the hyperfine mixing by exact diagonalization of the effective fine- and hyperfine-structure Hamiltonian of Eq. (12). The third improvement comes from the very accurate calculation of the expectation values of the fine- and hyperfine-structure operators using explicitly correlated functions allowing for the complete electron correlations. The fourth one is due to the very accurate calculation of the hyperfine mixing by exact diagonalization of the effective Hamiltonian of Eq. (12). The fifth source of improvement is due to accounting for the relativistic, radiative and nuclear structure corrections by appropriate rescaling of the $\alpha_1$ parameter.

Regarding our theoretical uncertainties, they come exclusively from neglected higher order relativistic and QED contributions, in particular those due to hyperfine mixing of the $2^3P_1$ and $2^3P_0$ states. The calculation of the complete $O(\alpha^2)$ correction is possible but technically difficult. It has been performed for Be$^+$ in [29] due to the possibility to use the exponentially correlated basis functions there. Nevertheless, if a complete $O(\alpha^2)$ correction is known, one can use both $B_1$ and $B_2$ parameters to obtain an even more accurate nuclear quadrupole moment of $^9$Be.

**TABLE IV.** Comparison of the electric quadrupole moment (in barns) of the $^9$Be nucleus obtained using atomic structure calculations.

| Source                        | Q_N($^9$Be) |
|-------------------------------|-------------|
| Blachman and Lurio, 1967      | 0.049(3)    |
| Ray et al., 1973 [24]         | 0.052 5(3)  |
| Sinanoğlu and Beck, 1973 [25]| 0.054 94    |
| Beck and Nicolaides, 1984     | 0.05545     |
| Sundholm and Olsen, 1991      | 0.052 88(38)|
| Jönsson and Fisher, 1993      | 0.052 56    |
| Nemouchi et al., 2003 [9]     | 0.052 77    |
| Beloy et al., 2008 [15]       | 0.053(3)    |
| This work                     | 0.053 50(14)|

**FIG. 1.** Comparison of the electric quadrupole moment (in barns) of the $^9$Be nucleus obtained using atomic structure calculations.

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