Ground State Hyperfine Splitting in ^{6,7}Li Atoms and the Nuclear Structure

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(Received 7 October 2013; published 9 December 2013)

Relativistic and QED corrections are calculated for a hyperfine splitting of the $2S_{1/2}$ ground state in ^{6,7}Li atoms with a numerically exact account for electronic correlations. The resulting theoretical predictions achieve such a precision level that, by comparison with experimental values, they enable determination of the nuclear properties. In particular, the obtained results show that the ^{7}Li nucleus, having a charge radius smaller than ^{6}Li, has about a 40% larger Zemach radius. Together with known differences in the electric quadrupole and magnetic dipole moments, this calls for a deeper understanding of the Li nuclear structure.

DOI: 10.1103/PhysRevLett.111.243001 PACS numbers: 31.30.J−, 21.10.Ky, 31.15.ac

Introduction.—Hyperfine splitting (hfs) of atomic energy levels results from the interaction between the magnetic moment of the atomic nucleus and that of the electrons. It has been measured very accurately for many elements, including light ones: H [1], D [2], ^{3}He [3], Li, and Be^+ [4]. Since the hyperfine interaction is singular at small distances, it strongly depends on the nucleus. For example, the nuclear structure contribution in H is -33 ppm, in D it is 138 ppm, and in $^3He^+$ it is -212 ppm [5], while experimental precision is orders of magnitude larger. This means that theoretical predictions for hydrogenic systems can only be as accurate as the uncertainty in the nuclear structure contribution. The situation is different for many electron systems where the limiting factor is the electron correlation, which is difficult to accurately account for using relativistic formalism based on the multielectron Dirac Hamiltonian [6,7].

In this work we overcame this problem by using the nonrelativistic QED approach, where relativistic and QED effects are treated perturbatively. We were able to accurately account for electron correlations by using explicitly correlated basis sets. We derived an exact formula for $O(\alpha^2)$ corrections, and higher orders were treated approximately with the help of hydrogenic results. This enabled us to achieve a few ppm accuracy and clearly identify the nuclear structure contribution. Surprisingly, the obtained results show significantly different magnetic moment distributions in ^{6}Li and ^{7}Li. This calls for a deeper understanding of the Li nuclear structure, or signals the existence of some unknown spin-dependent short-range force between charged hadrons and the lepton.

Effective Hamiltonian.—To calculate the hyperfine splitting in the Li atom we use the nonrelativistic QED approach, which consistently accounts for relativistic and QED effects. In this approach all corrections are treated perturbatively in powers of the fine structure constant and are expressed in terms of an effective Hamiltonian. For example, hyperfine splitting in the S state is given by the Fermi contact interaction

$$H^A_{\rm hfs} = \frac{2g_N Z\alpha}{3mM}\sum_a \vec{I}\cdot\vec{\sigma}_a \pi\delta^3(r_a). \tag{1}$$

The relation of g_N with the magnetic moment μ of the nucleus of charge Z is

$$g_N = \frac{M}{Zm_p}\frac{\mu}{\mu_N}\frac{1}{I}, \tag{2}$$

where μ_N is the nuclear magneton and I is the nuclear spin. Numerical values of the nuclear g factor for Li are presented in Table I. In general, the leading relativistic correction $H^{(4)}_{\rm hfs}$ of order $m\alpha^4$, which depends on nuclear spin I, is

$$H^{(4)}_{\rm hfs} = \frac{g}{2}H^A_{\rm hfs} + H^B_{\rm hfs} + H^C_{\rm hfs}, \tag{3}$$

$$H^B_{\rm hfs} = \varepsilon\frac{Z\alpha}{m^3}\sum_a \vec{I}\cdot\frac{\vec{r}_a\times\vec{p}_a}{r_a^3}, \tag{4}$$

$$H^C_{\rm hfs} = -\varepsilon\frac{Z\alpha}{2m^3}\sum_a \frac{I^i\sigma_a^j}{r_a^3}\left(\delta^{ij} - 3\frac{r_a^i r_a^j}{r_a^2}\right), \tag{5}$$

where $\varepsilon = g_N m^2/(2M)$, and M, m are masses of the nucleus and the electron, respectively. $H^B_{\rm hfs}$ and $H^C_{\rm hfs}$ in principle involve the electron g factor, which is set here to $g = 2$. This is because their expectation values vanish in any S state and they contribute only in the second order of perturbation theory (see below). Higher-order relativistic and QED corrections to hyperfine splitting are also expressed in terms of an effective Hamiltonian, so the expansion in α takes the form

$$E_{\rm hfs} = \langle H^{(4)}_{\rm hfs}\rangle + \langle H^{(5)}_{\rm hfs}\rangle + \langle H^{(6)}_{\rm hfs}\rangle + 2\left\langle H^{(4)}\frac{1}{(E-H)'}H^{(4)}_{\rm hfs}\right\rangle + \langle H^{(6)}_{\rm rad}\rangle + \langle H^{(7)}_{\rm hfs}\rangle, \tag{6}$$

where the prime denotes exclusion of the reference state from the resolvent.

TABLE I. Numerical values for the leading orders of hfs in the Li atom. The results from Ref. [8] are multiplied by 2.

	^{7}Li	^{6}Li
g_N [9]	5.039 274 8(26)	1.635 884 1(12)
$\mu/M \times 10^5$	7.820 202 745 2(50)	9.121 675 279(24)
$A^{(4,0)}$	5.811 937 88(5)	
Reference [8]	5.811 937 888(74)	
$A^{(4,1)}$	16.738 971(4)	
Reference [10]	16.8(1.8)	
$A^{(4)}$	5.810 628 86(5)	5.810 411 01(5)
$A^{(5)}_{\rm rec}$	−0.008 207 110	−0.009 416 884

$H^{(4)}$ is a Breit Hamiltonian in the nonrecoil limit,

$$H^{(4)} = H^A + H^B + H^C, \tag{7}$$

$$H^A = \sum_a \left[-\frac{p_a^4}{8m^3} + \frac{Z\alpha\pi}{2m^2}\delta^3(r_a) \right] + \sum_{a,b;a>b} \left[\frac{\pi\alpha}{m^2}\delta^3(r_{ab}) - \frac{\alpha}{2m^2} p_a^i \left(\frac{\delta^{ij}}{r_{ab}} + \frac{r_{ab}^i r_{ab}^j}{r_{ab}^3} \right) p_b^j \right], \tag{8}$$

$$H^B = \sum_a \frac{Z\alpha}{4m^2} \frac{\vec{r}_a}{r_a^3} \times \vec{p}_a \cdot \vec{\sigma}_a + \sum_{a,b;a\neq b} \frac{\alpha}{4m^2} \frac{\vec{r}_{ab}}{r_{ab}^3} \times (2\vec{p}_b - \vec{p}_a) \cdot \vec{\sigma}_a, \tag{9}$$

$$H^C = \sum_{a,b;a>b} \frac{\alpha}{4m^2} \frac{\sigma_a^i \sigma_b^j}{r_{ab}^3} \left(\delta^{ij} - \frac{3 r_{ab}^i r_{ab}^j}{r_{ab}^2} \right), \tag{10}$$

and $\vec{r}_{ab} = \vec{r}_a - \vec{r}_b$, $r_{ab} = |\vec{r}_{ab}|$.

$H^{(5)}_{\rm hfs}$ is a correction of order $m\alpha^5$. It is a Dirac-delta-like interaction with the coefficient obtained from the two-photon forward scattering amplitude. It has the same form as in hydrogen and depends on the nuclear structure. At the limit of a point spin 1/2 nucleus, it is

$$H^{(5)}_{\rm hfs} = -H^A_{\rm hfs} \frac{3Z\alpha}{\pi} \frac{m}{m_N} \ln\left(\frac{m_N}{m}\right) \equiv H^{(5)}_{\rm rec}, \tag{11}$$

a small nuclear recoil correction. For a finite-size nucleus $H^{(5)}_{\rm hfs}$ does not vanish at the nonrecoil limit. If we use a simple and inaccurate picture of the nucleus as a rigid ball described by the electric $\rho_E(r)$ and the magnetic $\rho_M(r)$ form factors, then $H^{(5)}_{\rm hfs}$ takes the form

$$H^{(5)}_{\rm hfs} = -H^A_{\rm hfs} 2Z\alpha m r_Z, \tag{12}$$

where

$$r_Z = \int d^3r\, d^3r' \rho_E(r) \rho_M(r') |\vec{r} - \vec{r}'|, \tag{13}$$

and the whole correction is encoded into the Zemach radius r_Z. The more accurate formula goes beyond the elastic form factor treatment. It was first found by Low and then much later reanalyzed and applied in calculations for such nuclei as D, T, and ^{3}He by Friar and Payne in Ref. [5],

$$H^{(5)}_{\rm hfs} = \frac{\pi\alpha^2}{2} \sum_a \delta^3(r_a) \int d^3r\, d^3r' \langle \{ \rho(\vec{r}), \vec{\sigma}_a \cdot (\vec{r} - \vec{r}') \times \vec{j}(\vec{r}') \} |\vec{r} - \vec{r}'| \rangle = -H^A_{\rm hfs} 2Z\alpha m \tilde{r}_Z, \tag{14}$$

where ρ and $\vec{j}$ are the nuclear charge and current density operators, respectively, and the last equation is the definition of $\tilde{r}_Z$. Both formulas include the same feature: linear dependence on the average distance of the magnetic moment density from the charge density. We did not attempt to perform nuclear structure calculations to obtain $H^{(5)}_{\rm hfs}$, because they are beyond our range. Instead, we used an experimental hyperfine splitting value to obtain the nuclear structure contribution and we expressed it in terms of an effective Zemach radius $\tilde{r}_Z$ according to Eq. (14). This gives us clues about the structure of Li nuclei.

The next term $H^{(6)}_{\rm hfs}$ includes nuclear spin-dependent operators that contribute at order $m\alpha^6$. This term is not well known in the literature. In hydrogenic systems it leads to the so-called Breit correction. For two-electron atoms it was presented in the work on ^{3}He hyperfine splitting [11], while for three-electron atoms the operators were derived in Ref. [12]. We rederived this result herein to obtain a slightly simplified but equivalent form. This was done as follows: the magnetic field coming from the nuclear magnetic moment is

$$e\vec{A}(\vec{r}) = \frac{e}{4\pi} \vec{\mu} \times \frac{\vec{r}}{r^3} = -Z\alpha \frac{g_N}{2M} \vec{I} \times \frac{\vec{r}}{r^3}. \tag{15}$$

Consider the part $\delta H_{\rm BP}$ of the Breit-Pauli Hamiltonian of the atomic system, which includes the coupling of the electron spin to the magnetic field

$$\delta H_{\rm BP} = \sum_a \Big\{ \frac{\vec{\pi}_a^2}{2m} - \frac{e}{2m}\vec{\sigma}_a\cdot\vec{B}_a + \frac{Z\alpha}{4m^2}\vec{\sigma}_a\cdot\frac{\vec{r}_a}{r_a^3}\times\vec{\pi}_a + \frac{e}{8m^3}(\vec{\sigma}_a\cdot\vec{B}_a\vec{\pi}_a^2 + \vec{\pi}_a^2\vec{\sigma}_a\cdot\vec{B}_a) + \sum_{b,b\neq a}\frac{\alpha}{4m^2 r_{ab}^3}\vec{\sigma}_a\cdot\vec{r}_{ab}\times(2\vec{\pi}_b - \vec{\pi}_a)\Big\}, \quad (16)$$

where $\vec{\pi} = \vec{p} - e\vec{A}$. The leading interaction $H_{\rm hfs}^{(4)}$ between the nuclear $\vec{I}$ and electron spins $\vec{\sigma}_a$ is obtained from the nonrelativistic terms

$$H_{\rm hfs}^{(4)} = -\sum_a \frac{e}{m}\vec{p}_a\cdot\vec{A}(\vec{r}_a) - \frac{e}{2m}\vec{\sigma}_a\cdot\vec{B}(\vec{r}_a), \quad (17)$$

with the magnetic field coming from the nucleus, Eq. (15). The relativistic correction $H_{\rm hfs}^{(6)}$ is similarly obtained from $\delta H_{\rm BP}$,

$$H_{\rm hfs}^{(6)} = \varepsilon\sum_a \vec{\sigma}_a\cdot\vec{I}\Big[\frac{(Z\alpha)^2}{6m^4}\frac{1}{r_a^4} - \frac{Z\alpha}{12m^5}\{p_a^2, 4\pi\delta^3(r_a)\} + \sum_{b;b\neq a}\frac{Z\alpha^2}{6m^4}\frac{\vec{r}_{ab}}{r_{ab}^3}\cdot\Big(2\frac{\vec{r}_b}{r_b^3} - \frac{\vec{r}_a}{r_a^3}\Big)\Big]. \quad (18)$$

However, the resulting operators are singular, and in the next section we briefly describe the cancellation of these singularities with those in the second-order matrix elements.

$H_{\rm rad}^{(6)}$ in Eq. (6) is a QED radiative correction [13,14]

$$H_{\rm rad}^{(6)} = H_{\rm hfs}^{A}\alpha(Z\alpha)\Big(\ln 2 - \frac{5}{2}\Big), \quad (19)$$

which is similar to that in hydrogen. The last term $E_{\rm hfs}^{(7)}$ of order $m\alpha^7$ is calculated approximately using the hydrogenic value for the one-loop correction from Ref. [15] and the two-loop correction from Ref. [14],

$$H_{\rm hfs}^{(7)} = H_{\rm hfs}^{A}\Big[\frac{\alpha}{\pi}(Z\alpha)^2\Big(-\frac{8}{3}\ln^2(Z\alpha) + a_{21}\ln(Z\alpha) + a_{20}\Big) + \frac{\alpha^2}{\pi}(Z\alpha)b_{10}\Big], \quad (20)$$

where $a_{21}(2S) = -1.1675$, $a_{20}(2S) = 11.3522$, and $b_{10} = 0.771\,652$.

We will express the hyperfine splitting in terms of the hyperfine constant A, defined as

$$E_{\rm hfs} = \vec{I}\cdot\vec{J}A, \quad (21)$$

where $\vec{J}$ is the total electronic angular momentum, which, for the ground state of Li, is equal to $1/2$. If we use the notation $H_{\rm hfs} = \vec{I}\cdot\vec{H}_{\rm hfs}$, then

$$A = \frac{1}{J(J+1)}\langle\vec{J}\cdot\vec{H}_{\rm hfs}\rangle. \quad (22)$$

The expansion of A in α takes the form

$$A = \varepsilon\Big[\frac{g}{2}\alpha^4 A^{(4)} + \sum_{n=5}^{\infty}\alpha^n A^{(n)}\Big]. \quad (23)$$

All of the results of the numerical calculations are given here in terms of dimensionless coefficients $A^{(n)}$.

Numerical results.—The matrix elements of all the operators are calculated with the nonrelativistic wave function Ψ expressed in terms of antisymmetrized functions ϕ_i,

$$\Psi = \sum_{i=1}^{N}\lambda_i\mathcal{A}[\phi_i(\vec{r}_1,\vec{r}_2,\vec{r}_3)(|\uparrow\downarrow\uparrow\rangle - |\downarrow\uparrow\uparrow\rangle)], \quad (24)$$

where λ_i are real coefficients and $\mathcal{A}$ denotes antisymmetrization. In this work we used for ϕ the explicitly correlated Hylleraas [8], Slater [16], and Gaussian [17] basis functions for various types of matrix elements. For convenience, in this section we will use atomic units, so all $A^{(i)}$ are dimensionless.

The leading $A^{(4)}$ coefficient using Eq. (1),

$$A^{(4)} = \frac{1}{J(J+1)}\frac{4\pi Z}{3}\langle\vec{J}\cdot\vec{\sigma}_a\delta^3(r_a)\rangle, \quad (25)$$

is calculated by using the expansion in the ratio of the reduced electron mass μ to the nuclear mass M,

$$A^{(4)} = A^{(4,0)} - \frac{\mu}{M}A^{(4,1)}. \quad (26)$$

The next-to-leading correction $A_{\rm rec}^{(5)}$ and all others are obtained in the leading order in the mass ratio, so that

$$A_{\rm rec}^{(5)} = -A^{(4)}\frac{3Z}{\pi}\frac{m}{m_N}\ln\Big(\frac{m_N}{m}\Big). \quad (27)$$

The most difficult part of the calculation is $A^{(6)}$, which is expressed in terms of the following matrix elements:

$$A^{(6)} = A_{AN}^{(6)} + A_B^{(6)} + A_C^{(6)} + A_R^{(6)}, \quad (28)$$

where

$$A_{AN}^{(6)} = \frac{2}{J(J+1)}\Big\langle\frac{4\pi Z}{3}\sum_a\vec{J}\cdot\vec{\sigma}_a\delta^3(r_a)\frac{1}{(E-H)'}H^A\Big\rangle + \frac{1}{J(J+1)}\Big\langle\sum_a\vec{J}\cdot\vec{\sigma}_a\Big[\frac{Z^2}{6}\frac{1}{r_a^4} - \frac{2Z}{3}p_a^2\pi\delta^3(r_a) + \sum_{b;b\neq a}\frac{Z}{6}\frac{\vec{r}_{ab}}{r_{ab}^3}\cdot\Big(2\frac{\vec{r}_b}{r_b^3} - \frac{\vec{r}_a}{r_a^3}\Big)\Big]\Big\rangle, \quad (29)$$

$$A_B^{(6)} = \frac{2}{J(J+1)}\Big\langle Z\sum_a\vec{J}\cdot\frac{\vec{r}_a\times\vec{p}_a}{r_a^3}\frac{1}{(E-H)'}H^B\Big\rangle, \quad (30)$$

$$A_C^{(6)} = \frac{2}{J(J+1)}\Big\langle -\frac{Z}{2}\sum_a\frac{J^i\sigma_a^j}{r_a^3}\Big(\delta^{ij} - 3\frac{r_a^i r_a^j}{r_a^2}\Big)\frac{1}{(E-H)'}H^C\Big\rangle, \quad (31)$$

and

TABLE II. Numerical values for relativistic and QED corrections (dimensionless) to the hyperfine splitting. Results from Ref. [6] in terms of G_{M1} are multiplied by 27.

Contribution	Value
$A^{(6)}_{AN}$	102.134(5)
$A^{(6)}_{B}$	0.020 50(3)
$A^{(6)}_{C}$	0.088 89(4)
$A^{(6)}_{R}$	−31.503 95
$A^{(6)}$	70.739(5)
Reference [6]	72.4
Reference [10]	62.(8)
$A^{(7)}$	−381.(48)

$$A^{(6)}_R = A^{(4)}\left(\ln 2 - \frac{5}{2}\right). \tag{32}$$

$\mathcal{A}^{(6)}_{AN}$ consists of two terms, which are separately divergent at small r_a. We obtained a finite expression by transforming operators in the second-order matrix element by

$$H^A \equiv H'^A + \frac{1}{4}\sum_a \left\{\frac{Z}{r_a}, E - H\right\}, \tag{33}$$

$$4\pi\delta^3(r_a) \equiv 4\pi[\delta^3(r_a)]' - \left\{\frac{2}{r_a}, E - H\right\}. \tag{34}$$

All singular terms are moved to the first-order matrix elements, which, when combined, form a well-defined and finite expression. The calculation of $A^{(6)}$ is the main result of this work. It agrees well with the former calculations in Refs. [6,10] (see Table II) but is much more accurate. The higher-order term $A^{(7)}$ is obtained directly from Eqs. (20) and (22). Numerical results for all of the expansion coefficients are presented in Table II. Final results are combined together in Table III. The uncertainty of final theoretical predictions for a point nucleus are estimated as 25% of the a_{20} coefficient in Eq. (20), which is calculated approximately using the hydrogenic result. The achieved accuracy is sufficient to obtain precise values of the nuclear structure effect. This is expressed in terms of $\tilde{r}_Z$, the effective Zemach radius, the value of which should not be very different from the charge radius r_E. While our results are in agreement with those of Yerokhin [6] for the point nucleus, the nuclear structure contribution compares strangely to the nuclear calculations performed in Ref. [6]. Namely, they agree well for ^{7}Li and strongly disagree for ^{6}Li, for which we do not have conclusive explanation.

Conclusions.—Until now, only H, D, and ^{3}He nuclei have been studied to a high degree of accuracy, due to the development in hfs theory of one-electron systems [14]. Here we extend the high-accuracy theoretical predictions to three-electron atoms (ions). Namely, we have calculated hyperfine splitting in ^{6,7}Li with an accuracy of a few ppm, which allows the determination of nuclear structure effects, expressed in terms of the effective Zemach radius $\tilde{r}_Z$.

TABLE III. Contributions in MHz to the hyperfine splitting constant A in ^{6,7}Li. Used constants are $g = 2.002\,319\,304\,361\,53(53)$, $\alpha^{-1} = 137.035\,999\,074(44)$, the next-to-last row is a Zemach radius inferred from comparison of experiment (expt) [4] with theoretical (theor) value for the point nucleus.

	^{7}Li	^{6}Li
$\varepsilon \times 10^{-9}$	24.348 067(13)	9.219 580(7)
$\varepsilon\alpha^4 g A^{(4)}/2$	401.654 08(21)	152.083 69(11)
$\varepsilon\alpha^5 A^{(5)}_{\rm rec}$	−0.004 14	−0.001 80
$\varepsilon\alpha^6 A^{(6)}$	0.260 08(2)	0.098 48(1)
$\varepsilon\alpha^7 A^{(7)}$	−0.010 2(13)	−0.003 9(5)
$A_{\rm theor}$ (point nucleus)	401.899 8(13)	152.176 5(5)
Reference [6]	401.903(11)	152.177 8(42)
$A_{\rm expt}$	401.752 043 3(5)	152.136 839(2)
$(A_{\rm expt} - A_{\rm theor})/A_{\rm expt}$	−368(3) ppm	−261(3) ppm
Reference [6] (nuclear calculations)	−369(23) ppm	−368(60) ppm
$\tilde{r}_Z$	3.25(3) fm	2.30(3) fm
r_E	2.390(30) fm	2.540(28) fm

The obtained result for $\tilde{r}_Z(^7\text{Li})$ is about 40% larger than $\tilde{r}_Z(^6\text{Li})$, in spite of the fact that the charge radius is smaller in ^{7}Li; see Table III. This indicates significant differences in the magnetic distribution of ^{7}Li and ^{6}Li nuclei, which shall be confirmed by the nuclear theory. This may also indicate that the standard treatment of finite nuclear size effects in the evaluation of the hyperfine splitting through elastic form factors fails in some cases.

In summary, we have shown that through purely atomic calculations and experiments one can gain valuable information on the structure of the atomic nucleus, in particular, the Zemach radius. Similar calculations can be performed for ^{11}Be where one expects a significant neutron halo [18].

The authors acknowledge support from NCN Grants No. 2012/04/A/ST2/00105 and No. 2011/01/B/ST4/00733.

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