

Lithium isotope shifts as a measure of nuclear size

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The isotope shifts for $2^2P_J-2^2S$ and 3^2S-2^2S transition energies in lithium are calculated variationally in Hylleraas coordinates, including nonrelativistic, relativistic, and QED terms up to $O(\mu/M)$, $O(\mu/M)^2$, $O(\alpha^2\mu/M)$, and $O(\alpha^3\mu/M)$ atomic units, and the lowest-order finite nuclear size correction. With high-precision isotope shift measurements, our results can potentially yield a precise determination of the nuclear charge radius for different isotopes of lithium, and especially for the exotic ^{11}Li ‘‘halo’’ isotope. For the case of $^7\text{Li}-^6\text{Li}$, using the nuclear charge radii from nuclear scattering data, our calculated isotope shifts for the $2^2P_{1/2}-2^2S$, $2^2P_{3/2}-2^2S$, and 3^2S-2^2S transitions are 10 534.31(61)(6) MHz, 10 534.70(61)(6) MHz, and 114 54.31(39)(5) MHz, respectively, where the first brackets indicate the uncertainties due to the nuclear charge radii, and the second brackets indicate the computational uncertainties. The experimental isotope shifts are inconsistent with each other and with theory for these transitions.

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I. INTRODUCTION

The root-mean-square (rms) radius of the nuclear charge distribution is a quantity of fundamental importance in nuclear physics [1]. For lithium, although the rms nuclear radii of the stable species ^6Li and ^7Li are determined at the 1% level [2], for unstable ^8Li , ^9Li , and ^{11}Li the rms charge radii are unknown. Among these isotopes, the study of ^{11}Li is of great current interest [3,4] because this isotope consists of a ^9Li core with a ‘‘halo’’ of two loosely bound neutrons orbiting the nucleus. In addition to the traditional nuclear scattering method, the nuclear charge distribution can be probed by a combination of atomic physics theory and experiment, provided both theory and measurement can be carried out to sufficiently high accuracy. One advantage of such an approach is its nuclear model independence. Although the influence of a finite nuclear charge distribution on the energy level of an atom is well known (see Ref. [5] for a review), it was pointed out by Drake [6] that a high-precision measurement of an isotope shift for a chosen transition might be used to extract the rms nuclear radius. Although the QED terms for light atoms are comparable in size to the nuclear size corrections, they are, to a first approximation, independent of the nuclear mass, and so they largely cancel from the calculated isotope shift. The significance of the method is therefore that the nuclear radius can be determined independently of QED uncertainties. This method was recently applied successfully to the studies of $^4\text{He}-^3\text{He}$ [1,7] and $^7\text{Li}^+-^6\text{Li}^+$ [8] isotope shifts. The determined rms radii for ^3He and $^6\text{Li}^+$ are in good agreement with nuclear scattering data, but with substantially improved accuracy.

For ^6Li and ^7Li , Vadla *et al.* [9] measured the isotope shifts of several transition lines using resonant Doppler-free two-photon laser spectroscopy. For the 3^2S-2^2S transition, the accuracy is 0.17%. Using laser-atomic-beam spectroscopy, Windholz and Umfer [10] measured the isotope shifts for the $2^2P_J-2^2S$ transitions. The precisions they obtained are 28 ppm for the D_1 line ($J=1/2$) and 114 ppm for the D_2 line ($J=3/2$). Later, they improved their experiment [11]

and reduced the uncertainties to 14 ppm for both lines. The third recent measurements for the isotope shifts in the $2^2P_J-2^2S$ transitions were done by Sansonetti *et al.* [12] using Doppler-free frequency-modulation spectroscopy. The precision they achieved is 30 ppm for the D_1 line and 21 ppm for the D_2 line (reduced to one standard deviation). Although for the D_1 line the value of Sansonetti *et al.* and the Windholz *et al.* revised value agree with each other, there is a discrepancy of 160 ppm for the D_2 line.

One purpose of this paper is to report the theoretical results of the $^7\text{Li}-^6\text{Li}$ isotope shifts for the 3^2S-2^2S and $2^2P_J-2^2S$ transitions. A second purpose is to provide a formula for the isotope shift between any two Li isotopes as a function of the assumed rms nuclear radii. This is motivated by the experimental efforts currently under way at GSI, Darmstadt [4], to measure the Li 3^2S-2^2S isotope shift with a proposed accuracy of 200 kHz. The ultimate goal is to determine the rms radii of Li isotopes, particularly the halo nucleus ^{11}Li , with a precision of 10% or better. The calculations are based on our recent advances [13–19] in high-precision variational calculations for lithium and lithiumlike ions using multiple basis sets in Hylleraas coordinates.

The paper is organized as follows. In Sec. II we review the theoretical formulation of the problem and construction of basis sets in Hylleraas coordinates for the lithium atom wave function. Finite nuclear mass effects and the mass polarization operator are taken into account up to second order by perturbation theory. Especially important are relativistic recoil terms of order $\alpha^2\mu/M$ a.u., since it is the accuracy of these terms that limits the accuracy of the final results. Section III presents results for basis sets containing up to 3502 terms, together with a general formula for the determination of rms nuclear radii for any isotope of lithium relative to a chosen reference isotope. Section IV discusses the comparison with experiment for the $^7\text{Li}-^6\text{Li}$ isotope shifts, and Sec. V presents some concluding remarks.

II. THEORETICAL FORMULATION

After rescaling distances according to $r \rightarrow (m/\mu)r$, the Hamiltonian for a three-electron atomic system is

$$H = H_0 + \lambda H', \quad (1)$$

with

$$H_0 = -\frac{1}{2} \sum_{i=1}^3 \nabla_i^2 - Z \sum_{i=1}^3 \frac{1}{r_i} + \sum_{i>j}^3 \frac{1}{r_{ij}}, \quad (2)$$

and

$$H' = \sum_{i>j}^3 \nabla_i \cdot \nabla_j, \quad (3)$$

in units of $2R_M$, where $R_M = (1 - \mu/M)R_\infty$, $\mu = mM/(m+M)$ is the electron reduced mass, and $\lambda = -\mu/M$, which can be treated as a perturbation parameter. The Schrödinger equation

$$H\Psi = E\Psi \quad (4)$$

can be solved perturbatively by expanding Ψ and E according to

$$\Psi = \Psi_0 + \lambda \Psi_1 + \dots, \quad (5)$$

$$E = \varepsilon_0 + \lambda \varepsilon_1 + \lambda^2 \varepsilon_2 + \dots. \quad (6)$$

Thus Eq. (4) becomes

$$H_0 \Psi_0 = \varepsilon_0 \Psi_0, \quad (7)$$

$$(\varepsilon_0 - H_0) \Psi_1 = (H' - \varepsilon_1) \Psi_0. \quad (8)$$

ε_1 and ε_2 are

$$\varepsilon_1 = \langle \Psi_0 | H' | \Psi_0 \rangle, \quad (9)$$

$$\varepsilon_2 = \langle \Psi_0 | H' | \Psi_1 \rangle - \varepsilon_1 \langle \Psi_0 | \Psi_1 \rangle. \quad (10)$$

Both Ψ_0 and Ψ_1 were solved variationally in multiple basis sets in Hylleraas coordinates containing terms of the form

$$r_1^{j_1} r_2^{j_2} r_3^{j_3} r_{12}^{j_{12}} r_{23}^{j_{23}} r_{31}^{j_{31}} e^{-\alpha r_1 - \beta r_2 - \gamma r_3} \times \mathcal{Y}_{(l_1 l_2) l_{12}, l_3}^{LM}(\hat{\mathbf{r}}_1, \hat{\mathbf{r}}_2, \hat{\mathbf{r}}_3) \chi_1, \quad (11)$$

where $\mathcal{Y}_{(l_1 l_2) l_{12}, l_3}^{LM}$ is a vector-coupled product of spherical harmonics for the three electrons to form a state of total angular momentum L , and χ_1 is a spin function with spin angular momentum $1/2$. As described previously [13,18], all terms from Eq. (11) are nominally included such that

$$j_1 + j_2 + j_3 + j_{12} + j_{23} + j_{31} \leq \Omega, \quad (12)$$

and the convergence of the eigenvalues is studied as Ω is progressively increased. Further details may be found in Ref. [18]. Since Eq. (6) is expressed in units of $(1 + \lambda)2R_\infty$, the explicit mass dependence of E is

$$E = \varepsilon_0 + \lambda(\varepsilon_0 + \varepsilon_1) + \lambda^2(\varepsilon_1 + \varepsilon_2) + O(\lambda^3) \quad \text{in units of } 2R_\infty. \quad (13)$$

The lowest-order relativistic corrections of $O(\alpha^2)$ and the spin-dependent anomalous magnetic moment corrections of $O(\alpha^3)$ can be written in the form [20,21] (in atomic units)

$$\Delta E_{\text{rel}} = \langle \Psi | H_{\text{rel}} | \Psi \rangle_J, \quad (14)$$

where Ψ is a nonrelativistic wave function, and H_{rel} is defined by

$$\begin{aligned} H_{\text{rel}} = & B_1 + B_2 + B_{3e} + B_{3z} + B_5 - \pi \alpha^2 \\ & \times \sum_{i>j}^3 \left(1 + \frac{8}{3} \mathbf{s}_i \cdot \mathbf{s}_j \right) \delta(\mathbf{r}_{ij}) + \frac{1}{2} Z \pi \alpha^2 \sum_{i=1}^3 \delta(\mathbf{r}_i) \\ & + \frac{m}{M} (\tilde{\Delta}_2 + \tilde{\Delta}_{3z}) + \gamma \left(2B_{3z} + \frac{4}{3} B_{3e} + \frac{2}{3} B_{3e}^{(1)} + 2B_5 \right) \\ & + \gamma \frac{m}{M} \tilde{\Delta}_{3z}. \end{aligned} \quad (15)$$

In Eq. (15),

$$B_1 = -\frac{\alpha^2}{8} (\nabla_1^4 + \nabla_2^4 + \nabla_3^4), \quad (16)$$

$$B_2 = \frac{\alpha^2}{2} \sum_{i>j}^3 \left[\frac{1}{r_{ij}} \nabla_i \cdot \nabla_j + \frac{1}{r_{ij}^3} \mathbf{r}_{ij} \cdot (\mathbf{r}_{ij} \cdot \nabla_i) \nabla_j \right], \quad (17)$$

$$B_{3e} = \frac{\alpha^2}{2} \sum_{i \neq j}^3 \frac{1}{r_{ij}^3} \mathbf{r}_{ji} \times \mathbf{p}_i \cdot (\mathbf{s}_i + 2\mathbf{s}_j), \quad (18)$$

$$B_{3z} = \frac{Z\alpha^2}{2} \sum_{i=1}^3 \frac{1}{r_i^3} \mathbf{r}_i \times \mathbf{p}_i \cdot \mathbf{s}_i, \quad (19)$$

$$B_5 = \alpha^2 \sum_{i>j}^3 \left[\frac{1}{r_{ij}^3} (\mathbf{s}_i \cdot \mathbf{s}_j) - \frac{3}{r_{ij}^5} (\mathbf{r}_{ij} \cdot \mathbf{s}_i) (\mathbf{r}_{ij} \cdot \mathbf{s}_j) \right], \quad (20)$$

$$\tilde{\Delta}_2 = \frac{iZ\alpha^2}{2} \sum_{j=1}^3 \left[\frac{1}{r_j} \mathbf{p} \cdot \nabla_j + \frac{1}{r_j^3} \mathbf{r}_j \cdot (\mathbf{r}_j \cdot \mathbf{p}) \nabla_j \right], \quad (21)$$

$$\tilde{\Delta}_{3z} = Z\alpha^2 \sum_{i=1}^3 \frac{1}{r_i^3} \mathbf{r}_i \times \mathbf{p} \cdot \mathbf{s}_i, \quad (22)$$

$$B_{3e}^{(1)} = \frac{\alpha^2}{2} \sum_{i \neq j}^3 \frac{1}{r_{ij}^3} \mathbf{r}_{ji} \times \mathbf{p}_i \cdot (\mathbf{s}_i - \mathbf{s}_j), \quad (23)$$

with $\mathbf{p} = \mathbf{p}_1 + \mathbf{p}_2 + \mathbf{p}_3$, and γ is

$$\gamma = \frac{\alpha}{2\pi} + (-0.32847) \left(\frac{\alpha}{\pi} \right)^2 + \dots \quad (24)$$

For doublet states, the operator $-\pi\alpha^2 \sum_{i>j} (1 + \frac{8}{3} \mathbf{s}_i \cdot \mathbf{s}_j) \delta(\mathbf{r}_{ij})$ can be replaced by $\pi\alpha^2 \sum_{i>j} \delta(\mathbf{r}_{ij})$, and the expectation value of the spin-spin term B_5 vanishes. B_i are the Breit-Pauli terms, the terms proportional to m/M are the

nuclear relativistic recoil corrections, and the terms proportional to γ are the anomalous magnetic moment corrections.

The perturbing effect of mass polarization on the expectation values of Breit operators can be obtained using

$$\Psi = \Psi_0 + \lambda (\Psi_1 - \langle \Psi_1 | \Psi_0 \rangle \Psi_0) + \dots, \quad (25)$$

where the extra term $-\langle \Psi_1 | \Psi_0 \rangle \Psi_0$ is added to Ψ_1 so that the first two terms of the right-hand side are orthogonal to each other [22]. Thus, for a Breit operator A , one has

$$\langle \Psi | A | \Psi \rangle = a_0 + \lambda a_1 + \dots, \quad (26)$$

where

$$a_0 = \langle \Psi_0 | A | \Psi_0 \rangle \quad (27)$$

and

$$a_1 = 2\langle \Psi_0 | A | \Psi_1 \rangle - 2\langle \Psi_0 | \Psi_1 \rangle \langle \Psi_0 | A | \Psi_0 \rangle. \quad (28)$$

Furthermore, due to the use of μ -scaled atomic units in Eq. (1), the units of $\langle \Psi | A | \Psi \rangle$ in Eq. (26) are $(\mu/m)^n 2R_\infty$, where $-n$ is the degree of homogeneity of operator A in three-electron coordinate space such that

$$A(\beta \mathbf{r}_1, \beta \mathbf{r}_2, \beta \mathbf{r}_3) = \beta^{-n} A(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3). \quad (29)$$

Using

$$\left(\frac{\mu}{m}\right)^n = (1 + \lambda)^n \approx 1 + n\lambda, \quad (30)$$

one has the explicit mass-dependent formula

$$\langle \Psi | A | \Psi \rangle = a_0 + \lambda (na_0 + a_1) + O(\lambda^2) \quad \text{in units of } 2R_\infty. \quad (31)$$

The QED corrections can be calculated according to the formulation of McKenzie and Drake [23]:

$$\Delta E_{\text{QED}} = \Delta E_{L,1} + \Delta E_{L,2}. \quad (32)$$

In Eq. (32), $\Delta E_{L,1}$ is given by

$$\Delta E_{L,1} = \alpha^3 Z \left[\frac{x F(1s_{1/2}) + F(nl_j)/n^3}{x + \delta_{l,0}/n^3} \right] \langle \delta(\mathbf{r}_i) \rangle_{1s^n l}, \quad (33)$$

where x is the number of $1s$ electrons, $F(nl_j)$ is the one-electron QED function defined by Johnson and Soff [24], and $\langle \delta(\mathbf{r}_i) \rangle$ denotes $\langle \sum_{i=1}^3 \delta(\mathbf{r}_i) \rangle$. The two-electron QED shift is

$$\Delta E_{L,2} = \alpha^3 \left(\frac{14}{3} \ln \alpha + \frac{164}{15} \right) \langle \delta(\mathbf{r}_{ij}) \rangle - \frac{14}{3} \alpha^3 Q, \quad (34)$$

where the Q term is defined by

$$Q = (1/4\pi) \lim_{\epsilon \rightarrow 0} \langle r_{ij}^{-3}(\epsilon) + 4\pi(\gamma + \ln \epsilon) \delta(\mathbf{r}_{ij}) \rangle. \quad (35)$$

In Eq. (35), γ is Euler's constant, ϵ is the radius of a sphere about $r_{ij}=0$ excluded from the integration, and a summation over $i > j$ from 1 to 3 is assumed. Finally, the last correction to be included is that due to finite nuclear size. It is given in lowest order by

$$\Delta E_{\text{nuc}} = \frac{2\pi Z r_{\text{rms}}^2}{3} \langle \delta(\mathbf{r}_i) \rangle, \quad (36)$$

where $r_{\text{rms}} = R_{\text{rms}}/a_{\text{Bohr}}$, R_{rms} is the root-mean-square radius of the nuclear charge distribution, and a_{Bohr} is the Bohr radius. A mass scaling factor of $(\mu/m)^3$ is included in the definition of $\langle \delta(\mathbf{r}_i) \rangle$.

In a nonrelativistic approximation, the $\lambda \epsilon_0$ term of Eq. [13] is called the normal isotope shift, and the remaining terms $\lambda \epsilon_1 + \dots$ are sometimes called the specific isotope shift. However, this partition becomes somewhat artificial when relativistic corrections are included because the overall multiplying factor is no longer simply $1 + \lambda$ from the finite-mass Rydberg. The B_1 term scales with mass as $(1 + \lambda)^4$ and the other terms in Eq. [15] scale as $(1 + \lambda)^3$.

III. CALCULATIONS AND RESULTS

Table I shows a convergence study of ϵ_0 for 3^2S as the size of basis set increases progressively up to 3502 terms, together with comparisons with King's result [25] using Hylleraas coordinates, and the result of Wang *et al.* [26] using a full-core-plus-correlation wave function. Our calculation improves theirs by factors of 1.2×10^6 and 2.3×10^4 respectively. A similar convergence study for the 2^2S and 2^2P states can be found in Ref. [18]. Our results for the nonrelativistic energies for the 2^2S , 2^2P , and 3^2S are calculated to a computational accuracy of a few parts in 10^{12} . Table II contains the nonrelativistic energy λ -expansion coefficients ϵ_0 , ϵ_1 , and ϵ_2 for these states.

The expectation values of the Breit operators and the two-electron Q term were evaluated for the 2^2S , 2^2P_J , and 3^2S states of lithium, together with the first-order finite nuclear mass corrections according to Eq. (31). The results are presented in Table III, and a comparison is made with the work of Wang *et al.* [26] for the 3^2S state. For the 2^2S and 2^2P_J states, a comparison with Chung's work [27] can be found in Ref. [19].

Table IV lists the contributions to the 3^2S-2^2S and $2^2P_J-2^2S$ transition energies, from the nonrelativistic, the lowest-order relativistic, and the lowest-order QED terms up to μ/M , $(\mu/M)^2$, $(\mu/M)\alpha^2$, and $(\mu/M)\alpha^3$, as well as the contributions from the finite nuclear size. The leading isotope-independent term is not included because it does not contribute to the isotope shift for the transition energies. Combining all the coefficients of μ/M , we arrive at the following formulas giving the Li isotope shifts for the 3^2S-2^2S , $2^2P_{1/2}-2^2S$, and $2^2P_{3/2}-2^2S$ transitions:

TABLE I. Convergence of the nonrelativistic energy for the $1s^2 3s^2 S$ state of lithium, in atomic units. $R(\Omega)$ is the ratio between two successive differences.

Ω	No. of terms	$\varepsilon_0(\Omega)$	$\varepsilon_0(\Omega) - \varepsilon_0(\Omega - 1)$	$R(\Omega)$
3	51	-7.353 807 225 055		
4	121	-7.354 068 196 845	-0.000 260 971 790	
5	257	-7.354 095 840 826	-0.000 027 643 980	9.440
6	503	-7.354 098 120 924	-0.000 002 280 098	12.124
7	919	-7.354 098 382 175	-0.000 000 261 251	8.727
8	1590	-7.354 098 417 067	-0.000 000 034 892	7.487
9	2626	-7.354 098 420 627	-0.000 000 003 559	9.803
10	3502	-7.354 098 421 082	-0.000 000 000 455	7.821
∞		-7.354 098 421 149(18)		
		-7.354 076 ^a		
		-7.354 098 0 ^b		

^aKing, Ref. [25].

^bWang *et al.*, Ref. [26].

$$\begin{aligned}
 f_{3^2S-2^2S} = & -0.133\,767\,15(64) (\mu/M) \\
 & + 0.123\,648\,10(29) (\mu/M)^2 \\
 & - 0.666\,646\,3(55) r_{\text{rms}}^2 + 1.980\,2(19) r_{\text{rms}}^2 (\mu/M),
 \end{aligned} \tag{37}$$

$$\begin{aligned}
 f_{3^2P_{1/2}-2^2S} = & -0.122\,993\,78(79) (\mu/M) \\
 & - 0.003\,95(14) (\mu/M)^2 \\
 & - 1.045\,610\,95(89) r_{\text{rms}}^2 \\
 & + 2.136(15) r_{\text{rms}}^2 (\mu/M),
 \end{aligned} \tag{38}$$

$$\begin{aligned}
 f_{2^2P_{3/2}-2^2S} = & -0.122\,998\,37(79) (\mu/M) \\
 & - 0.003\,95(14) (\mu/M)^2 \\
 & - 1.045\,610\,95(89) r_{\text{rms}}^2 \\
 & + 2.136(15) r_{\text{rms}}^2 (\mu/M),
 \end{aligned} \tag{39}$$

in units of $2R_\infty$. Table V summarizes the nuclear data for the various isotopes of lithium, including the values of μ/M (in terms of the atomic mass M_A) and R_{rms} for the isotopes ^7Li and ^6Li . With these values as input, Table VI lists the calculated contributions to the isotope shifts for the $2^2P_J-2^2S$ transitions. The first uncertainties are due to the uncertainties of the rms nuclear radii, and the second uncertainties are from the uncertainties of the computed coefficients. The main uncertainties of the total isotope shifts are due to the

nuclear charge radii, since the terms of order μ/M , $(\mu/M)^2$, and $\alpha^2 \mu/M$ are well established, and the QED terms of order $\alpha^3 \mu/M$ hardly affect the comparison. The contribution from the $r_{\text{rms}}^2 \mu/M$ term is negligible.

Table VII contains the contributions to the isotope shift for the 3^2S-2^2S transition. King [25] also studied this isotope shift using Hylleraas type wave functions. His result is $-0.381\,800\text{ cm}^{-1}$. However, his value for the expectation value of the mass polarization operator for the 3^2S state is only accurate to about 270 ppm, and he did not include the relativistic recoil term of order $\alpha^2 \mu/M$. The accuracy of this latter term is in fact the dominant source of theoretical uncertainty.

Determination of nuclear radii

The principal motivation for this work is to provide a spectroscopic means of determining nuclear radii from the observed isotope shifts. For this purpose, we take the calculated coefficients in Eqs. (37) to (39) as correct and rewrite these equations in the form

$$R_{\text{rms}}^2(^A\text{Li}) = R_{\text{rms}}^2(^6\text{Li}) + \frac{E_{\text{meas}}^A - E_0^A}{C}, \tag{40}$$

for the nuclear radius squared of an arbitrary isotope ^ALi relative to ^6Li . E_{meas}^A is the measured isotope shift for ^ALi relative to ^6Li , and E_0^A contains all the calculated contribu-

TABLE II. Nonrelativistic eigenvalue coefficients ε_0 , ε_1 , and ε_2 for the $1s^2 2s^2 S$, $1s^2 3s^2 S$, and $1s^2 2p^2 P$ states of lithium.

Coefficient	$1s^2 2s^2 S$	$1s^2 3s^2 S$	$1s^2 2p^2 P$
ε_0	-7.478 060 323 650 3(71)	-7.354 098 421 149(18)	-7.410 156 531 763(42)
ε_1	-0.301 842 783 02(25)	-0.292 039 834 10(22)	-0.246 738 887 5(70)
ε_2	-1.499 788 67(17)	-1.385 943 52(23)	-1.558 84(14)

TABLE III. $\lambda = -\mu/M$ expansion coefficients a_0 and $na_0 + a_1$ in Eq. (31) of the expectation values of the Breit operators and the two-electron Q term for the $1s^2 2s^2 S$, $1s^2 3s^2 S$, and $1s^2 2p^2 P_{1/2}$ states of lithium. Units are $2R_\infty$.

Operator	λ^0	λ^1
	$1s^2 2s^2 S$	
B_1	-0.004 183 221 020(30)	-0.017 006 84(50)
B_2	-0.000 023 196 186 8(73)	-0.000 233 449 3(20)
$\tilde{\Delta}_2$	-0.006 971 407 48(15)	-0.023 183 410 7(30)
$\langle \delta(\mathbf{r}_i) \rangle$	13.842 609 642(55)	42.012 420(30)
$\langle \delta(\mathbf{r}_{ij}) \rangle$	0.544 329 79(31)	1.550 973 5(93)
Q	0.021 778(21)	-0.065 3(50)
	$1s^2 3s^2 S$	
B_1	-0.004 146 016 6(18)	-0.016 852 32(40)
B_2	-0.000 022 893 198(20)	-0.000 224 300(30)
	-0.000 023 0 ^a	
$\tilde{\Delta}_2$	-0.006 892 527 026(74)	-0.022 833 349(50)
$\langle \delta(\mathbf{r}_i) \rangle$	13.736 509 59(87)	41.697 27(30)
$\langle \delta(\mathbf{r}_{ij}) \rangle$	0.536 175 15(47)	1.530 145(40)
	0.564 ^a	
Q	0.015 794(96)	-0.084 4(60)
$B_1 + Z\pi\alpha^2 \langle \delta(\mathbf{r}_i) \rangle / 2$	-0.000 698 961 3(18)	
	-0.000 696 8 ^a	
	$1s^2 2p^2 P_{1/2}$	
B_1	-0.004 127 280 433 0(40)	-0.016 819 160(65)
B_2	-0.000 021 110 238 5(48)	-0.000 223 13(10)
B_{3e}	0.000 004 014 992 20(99)	-0.000 001 208(50)
B_{3z}	-0.000 005 030 101 04(65)	-0.000 000 053(30)
$B_{3e}^{(1)}$	0.000 002 478 390 2(69)	0.000 002 624 3(30)
$\tilde{\Delta}_2$	-0.006 848 916 194(77)	-0.022 854 3(20)
$\tilde{\Delta}_{3z}$	0.000 001 799 199(82)	-0.000 006 162(50)
$\langle \delta(\mathbf{r}_i) \rangle$	13.676 195 49(13)	41.672 5(24)
$\langle \delta(\mathbf{r}_{ij}) \rangle$	0.532 281 42(51)	1.530 11(10)
Q	0.022 997 5(88)	-0.079 5(20)

^aWang *et al.*, Ref. [26].

TABLE IV. Expansion coefficients of λ , λ^2 , r_{rms}^2 , and λr_{rms}^2 for the lithium $1s^2 3s^2 S - 1s^2 2s^2 S$ and $1s^2 2p^2 P_{J-1} - 1s^2 2s^2 S$ transition energies, where $\lambda = -\mu/M$. The subscript indicates the value of J . Units are $2R_\infty$.

Term	Source	$3^2 S - 2^2 S$	$2^2 P_{J-2} - 2^2 S$
λ	Nonrelativistic	0.133 764 851 42(33)	0.123 007 687 4(70)
λ^2	Nonrelativistic	0.123 648 10(29)	-0.003 95(14)
λ	Relativistic, α^2	0.000 002 22(64)	-0.000 016 34(79) _{1/2}
			-0.000 011 75(79) _{3/2}
λ	Anomalous magnetic, α^3		-0.000 000 002 05(10) _{1/2}
			0.000 000 001 02(10) _{3/2}
λ	One-electron QED, α^3	-0.000 000 046	0.000 002 312
λ	Two-electron QED, α^3	0.000 000 132(14)	0.000 000 123 2(98)
r_{rms}^2	Finite nuclear size	-0.666 646 3(55)	-1.045 610 95(89)
λr_{rms}^2	Finite nuclear size	-1.980 2(19)	-2.136(15)

TABLE V. Summary of the nuclear spin (S), lifetime ($T_{1/2}$), atomic mass (M_A), magnetic dipole and electric quadrupole nuclear moments (μ_I and Q), hyperfine structure splitting (HFS, in the $2S$ state), rms mass radius $R_{\text{rms}}^{(m)}$, and charge radius $R_{\text{rms}}^{(e)}$ for the isotopes of lithium.

Quantity	${}^6\text{Li}$	${}^7\text{Li}$	${}^8\text{Li}$	${}^9\text{Li}$	${}^{11}\text{Li}$
S	1	3/2	2	3/2	3/2
$T_{1/2}$ (ms) ^a	∞	∞	838(6)	178.3(4)	8.59(14)
M_A (u) ^b	6.015 1223(5)	7.016 0040(5)	8.022 4867(5)	9.026 7891(21)	11.043 796(29)
μ_I (nm) ^c	0.822 0473(6)	3.256 4268(17)	1.653 560(18)	3.439 1(6)	3.667 8(25)
Q (mbarn) ^d	-0.83(8)	-40.0(3)	31.1(5)	-27.4(1.0)	-31.2(4.5)
HFS (MHz) ^e	228.205 259(3)	803.504 0866(10)	382.543(7)	856(16)	920(39)
$R_{\text{rms}}^{(m)}$ (fm) ^f	2.35(3)	2.35(3)	2.38(2)	2.32(2)	3.10(17)
$R_{\text{rms}}^{(e)}$ (fm) ^g	2.55(4)	2.39(3)	?	?	?

^aReference [28].

^bReference [29]. For Li, $\mu/M = 1/[1822.888\,511(43)M_A - 2]$.

^cReference [30].

^dSee Ref. [31] for ${}^6\text{Li}$, Ref. [32] for ${}^7\text{Li}$, and Ref. [33] for ${}^8\text{Li}$, ${}^9\text{Li}$, and ${}^{11}\text{Li}$.

^eSee Ref. [34] for ${}^6\text{Li}$ and ${}^7\text{Li}$, Ref. [35] for ${}^8\text{Li}$, and Ref. [36] for ${}^9\text{Li}$ and ${}^{11}\text{Li}$.

^fReference [37].

^gReference [2].

tions to the isotope shift except for the nuclear size contributions. The constant C is given by

$$C = \frac{2\pi Z}{3} [\langle \delta(\mathbf{r}_i) \rangle_i - \langle \delta(\mathbf{r}_i) \rangle_f]$$

$$= -2.4565 \text{ MHz/fm}^2 \quad \text{for } 2^2P_J - 2^2S$$

$$= -1.5661 \text{ MHz/fm}^2 \quad \text{for } 3^2S - 2^2S.$$

It depends on the transition $i \rightarrow f$ in question, but it is nearly independent of the mass number A . Using the atomic masses from Table V, the numerical values for E_0^A are listed in Table VIII. In the case of ${}^{11}\text{Li}$, the uncertainty in E_0^A for the $3^2S - 2^2S$ transition is composed of a ± 0.18 MHz uncertainty from the theoretical coefficients (mainly the $\alpha^2 \mu/M$ term),

and a ± 0.12 MHz uncertainty from the ${}^{11}\text{Li}$ atomic mass. The final uncertainty of ± 0.21 MHz is sufficient to determine R_{rms}^2 for ${}^{11}\text{Li}$ to an accuracy of $\pm 0.13 \text{ fm}^2$. Somewhat higher accuracy in R_{rms}^2 could be obtained from the $2^2P_J - 2^2S$ transitions ($\pm 0.10 \text{ fm}^2$), but the larger linewidth would pose additional experimental difficulties. With further improvements to the theory, a better measurement of M_A for ${}^{11}\text{Li}$ would also be desirable in order to exploit fully the isotope shift method of measuring the nuclear charge radius.

IV. COMPARISONS WITH EXPERIMENT

Our result for the $3^2S - 2^2S$ ${}^7\text{Li} - {}^6\text{Li}$ isotope shift of $11\,454.29(38)(5)$ MHz lies just at the upper edge of the error limits for the value $11\,434(20)$ MHz measured by Vadla

TABLE VI. Contributions to the ${}^7\text{Li} - {}^6\text{Li}$ isotope shifts for the $1s^2 2p^2 P_J - 1s^2 2s^2 S$ transitions and comparison with experiment. Units are MHz.

Contribution	$2^2P_{1/2} - 2^2S$	$2^2P_{3/2} - 2^2S$
	Theory	
μ/M	10 533.501 81(60) ^a	10 533.501 81(60) ^a
$(\mu/M)^2$	0.057 3(20)	0.057 3(20)
$\alpha^2 \mu/M$	-1.397(66)	-1.004(66)
$\alpha^3 \mu/M$, anom. magnetic	-0.000 175 3(84)	0.000 087 5(84)
$\alpha^3 \mu/M$, one-electron	0.198	0.198
$\alpha^3 \mu/M$, two-electron	0.010 55(84)	0.010 55(84)
r_{rms}^2	1.94(61)	1.94(61)
$r_{\text{rms}}^2 \mu/M$	-0.000 73(11)	-0.000 73(11)
Total	10 534.31(61)(7)	10 534.70(61)(7)
	Experiment	
Sansonetti <i>et al.</i> [12]	10 532.9(3)	10 533.3(2)
Windholz and Umfen [10]	10 534.3(3)	10 539.9(1.2)
Scherf <i>et al.</i> [11]	10 533.13(15)	10 534.93(15)

^aThe additional uncertainty from the atomic mass determinations is ± 0.008 MHz.

TABLE VII. Contributions to the ${}^7\text{Li}$ - ${}^6\text{Li}$ isotope shift for the $1s^23s^2S-1s^22s^2S$ transition. Units are MHz.

Contribution	3^2S-2^2S
μ/M	11 454.668 686(29) ^a
$(\mu/M)^2$	-1.793 864 0(41)
$\alpha^2 \mu/M$	0.188(53)
$\alpha^3 \mu/M$, one electron	-0.003 95
$\alpha^3 \mu/M$, two electron	0.011 3(12)
r_{rms}^2	1.24(39)
$r_{\text{rms}}^2 \mu/M$	-0.000 677(98)
Total	11 454.31(39)(5)
King [25]	11 446.1
Vadla <i>et al.</i> (experiment) [9]	11 434(20)

^aThe additional uncertainty from the atomic mass determinations is ± 0.008 MHz.

et al. [9] (see Table VII). Here, the experimental precision of ± 20 MHz is not sufficient to provide a significant test of theory beyond the lowest order μ/M term, and the uncertainty is much larger than the nuclear radius contribution.

However, as illustrated in Fig. 1, the interpretation of the measurements for the $2^2P_J-2^2S$ isotope shifts is much more obscure for several reasons. First, as a preliminary remark, we have verified in parallel calculations for the $1s2p^3P_J-1s2s^3S_1$ isotope shift in Li^+ that theory and experiment are in good agreement. In fact, the Li^+ experiment determines the difference in nuclear radii to be $R_{\text{rms}}({}^6\text{Li}) - R_{\text{rms}}({}^7\text{Li}) = 0.15 \pm 0.01$ fm, in close agreement with the value 0.16 ± 0.05 fm from nuclear scattering data. We therefore take the values for R_{rms} listed in Table V as correct. For the case of neutral lithium, the two sets of measurements by Sansonetti *et al.* [12] and Scherf *et al.* [11] do not agree with each other, and of the four separate measurements, only the $2^2P_{3/2}-2^2S$ result of Scherf *et al.* is in reasonable agreement with theory. In Fig. 1, note that the large outer error bars on the two theoretical data points are due to the nuclear radius uncertainty. These error bars are directly correlated since a change in R_{rms}^2 would shift both theoretical points in the same direction and by the same amount, as further discussed in the following paragraph.

A useful check on the experimental data is provided by the $2^2P_{3/2}-2^2P_{1/2}$ splitting isotope shift (SIS), obtained from the difference between the $2^2P_{3/2}-2^2S$ and $2^2P_{1/2}-2^2S$ isotope shifts. To the necessary accuracy, this quantity

TABLE VIII. Values of E_0^A to determine R_{rms}^2 from the measured isotope shift in various transitions (see Eq. [40]). Units are MHz.

Isotopes	$E_0^A(2^2P_{1/2}-2^2S)$	$E_0^A(2^2P_{3/2}-2^2S)$	$E_0^A(3^2S-2^2S)$
${}^7\text{Li}-{}^6\text{Li}$	10 532.37(7)	10 532.76(7)	11 453.07(6)
${}^8\text{Li}-{}^6\text{Li}$	18 473.18(12)	18 473.87(12)	20 088.23(10)
${}^9\text{Li}-{}^6\text{Li}$	24 631.53(16)	24 632.44(16)	26 785.18(13)
${}^{10}\text{Li}-{}^6\text{Li}$	29 575.97(20)	29 577.07(20)	32 162.12(17)
${}^{11}\text{Li}-{}^6\text{Li}$	33 615.77(24)	33 617.02(24)	36 555.34(21)

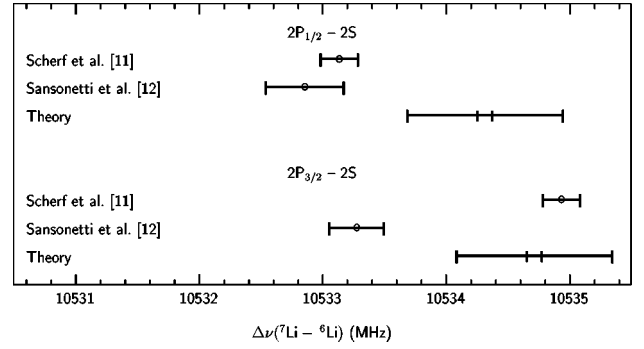


FIG. 1. Comparison between theory and experiment for the ${}^7\text{Li}$ - ${}^6\text{Li}$ $2^2P_J-2^2S$ isotope shift. For the theoretical data points, the inner error bars denote the computational uncertainty, and the outer error bars include the nuclear radius uncertainty. The outer error bars for the two theoretical data points are directly correlated (see the text). Experimental error bars represent one standard deviation.

is determined entirely by the spin-dependent part of the single $\alpha^2 \mu/M$ term in Table VI. QED and finite nuclear volume corrections largely cancel since they are nearly the same for both states. The predicted SIS is 0.393(66) MHz, in excellent agreement with the measured value 0.4(3) MHz from Sansonetti *et al.* It may be that their SIS is more accurate than their error estimates for the full transition frequencies would indicate, but that both measurements are too low by about 1.4(3) MHz. On the other hand, the data of Scherf *et al.* yield a SIS of 1.80(15) MHz, indicating that at least one of their measurements is incorrect by about nine standard deviations. It is clear that additional experimental work is required to resolve these discrepancies.

V. DISCUSSION

In this work, we have obtained the theoretical data necessary to derive nuclear radii from isotope shift measurements in neutral lithium. This work complements earlier work for transitions in Li^+ [8] where theory and experiment were shown to be in good agreement, and consistent with the nuclear radii for ${}^6\text{Li}$ and ${}^7\text{Li}$ derived from nuclear scattering measurements. The results can be applied directly to the ${}^{11}\text{Li}$ ‘‘halo’’ isotope for which the rms nuclear charge radius is difficult to determine by standard methods because of its low abundance. The present theoretical accuracy would allow a measurement of R_{rms}^2 accurate to ± 0.13 fm². Further improvements to the theory would also require an improved atomic mass measurement for ${}^{11}\text{Li}$ in order to exploit to the full the potential accuracy of the isotope shift method.

The comparison between theory and experiment for the ${}^7\text{Li}$ - ${}^6\text{Li}$ isotope shift in the $2^2P_J-2^2S$ transitions is not very satisfactory. The experiments are inconsistent with each other and with theory. A resolution of these inconsistencies would be very valuable in verifying that all contributions to isotope shifts in lithium have been calculated correctly and to sufficient accuracy.

Note added in proof. In recent calculations for helium, Pachucki and Sapirstein [38] discuss additional QED recoil corrections of order $\alpha^3 \mu/M$ which are not included in the present work. Similar corrections should also be included for

lithium, but they are unlikely to change the present results for the isotope shifts by more than a few tenths of a megahertz.

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