Isotope shift of $3^2S_{1/2} - 2^2S_{1/2}$ transition in lithium and nuclear polarizability

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High precision calculation of the isotope shift of $3^2S_{1/2} - 2^2S_{1/2}$ transition in lithium is presented. The wave function and matrix elements of relativistic operators are obtained by using recursion relations. Apart from the relativistic contribution we obtain the nuclear polarizability correction for ^{11}Li. The resulting difference of squared charge radii ^{11}Li - ^{7}Li based on the measurement by R. Sánchez *et al.* [Phys. Rev. Lett. **96**, 033002 (2006)] is $\delta r^2_{\rm ch} = 0.157(81)$ fm^2 which significantly differs from the previous evaluation.

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The recent advances in precise spectroscopy of atomic systems make possible the determination of nuclear charge radii from isotope shift measurements [1–4]. In spite of the fact that the nuclear size is 5 orders of magnitude smaller than the atomic size, the achieved experimental precision for transition frequencies allows one to determine nuclear charge radii much more accurately than from electron scattering measurements. This, however requires the theoretical calculations to be performed with high relative precision, for example, at least 10^{-6} for lithium isotopes. The accuracy of theoretical predictions achieved for hydrogen [5] leads to the determination of the proton charge radius [5, 6], which is far more accurate than from the electron scattering measurements. A similar experimental accuracy achieved for deuterium allowed one to determine very accurately the deuteron radius [1]. Surprisingly, the atomic measurements lead to a slightly different value from the electron scattering determination, which stimulated a reanalysis of electron scattering data. What makes the deuteron different from other typical nuclei is its low binding energy of about 2.226 MeV. Such a soft nucleus is distorted by a surrounding electron which leads to a shift in atomic transition frequencies. Even smaller is the two-neutron separation energy in ^{11}Li [7], what indicates possible significance of nuclear structure effects on the isotope shift.

In this work we present significantly improved calculations of finite nuclear mass contributions to the isotope shift in the lithium $3^2S_{1/2} - 2^2S_{1/2}$ transition. Such calculations have already been performed by Yan and Drake in [8–10] and were used to determine nuclear charge radii of various lithium isotopes on the basis of recent isotope shift measurements [3, 4]. Our result for the relativistic recoil correction is about 10 times smaller than that reported in [8, 9]. Apart from the known nonrelativistic, leading relativistic and QED corrections we include higher order recoil corrections and nuclear polarizability effect $E_{\rm pol}$, the last being significant for the ^{11}Li nucleus. Finally, we use our combined results to calculate new nuclear charge radii for the lithium isotopes, taking the experimental isotope shift results from [3, 4].

Let us denote $\eta = -\mu/M$. The expansion of an energy level in the fine structure constant α is

$$\begin{aligned} E(\eta,\alpha) &= m\,\alpha^2\,\mathcal{E}^{(2)}(\eta) + m\,\alpha^4\,\mathcal{E}^{(4)}(\eta) + m\,\alpha^5\,\mathcal{E}^{(5)}(\eta) \\ &\quad + m\,\alpha^6\,\mathcal{E}^{(6)}(\eta) + O(\alpha^7) + E_{\rm pol} + E_{\rm fs}, \end{aligned} \tag{1}$$

where $E_{\rm fs}$ is the nuclear finite size correction, m, M, μ is electron, nucleus, and the reduced mass respectively. In the following, we obtain these expansion coefficients. The nonrelativistic Hamiltonian of the lithium atom in atomic units is

$$H = H_\infty + H_M, \tag{2}$$

$$H_\infty = \sum_a \left[\frac{p_a^2}{2} - \frac{Z}{r_a}\right] + \sum_{a>b} \frac{1}{r_{ab}}, \tag{3}$$

$$H_M = \sum_a \frac{p_a^2}{2\,M} + \sum_{a>b} \frac{\vec p_a \cdot \vec p_b}{M}. \tag{4}$$

We solve the Schrödinger equation for H_∞ and treat H_M perturbatively. Following Ref. [11], the Schrödinger wave function is represented in the Hylleraas basis set

$$\phi = r_{23}^{n_1}\, r_{31}^{n_2}\, r_{12}^{n_3}\, r_1^{n_4}\, r_2^{n_5}\, r_3^{n_6}\, e^{-w_1\,r_1 - w_2\,r_2 - w_3\,r_3}, \tag{5}$$

with all combinations of n_i, such that $\sum_{i=1}^6 n_i \leq \Omega$ and $\Omega = 3, ..., 12$. Details can be found in Ref. [12]. The matrix elements of the nonrelativistic Hamiltonian can all be expressed by a Hylleraas integral $f = f(n_1, n_2, n_3, n_4, n_5, n_6)$

$$\begin{aligned} f &= \int \frac{d^3r_1}{4\,\pi} \int \frac{d^3r_2}{4\,\pi} \int \frac{d^3r_3}{4\,\pi}\, e^{-w_1\,r_1 - w_2\,r_2 - w_3\,r_3} \\ &\quad r_{23}^{n_1-1}\, r_{31}^{n_2-1}\, r_{12}^{n_3-1}\, r_1^{n_4-1}\, r_2^{n_5-1}\, r_3^{n_6-1}, \end{aligned} \tag{6}$$

with integer values of n_i. We calculate f analytically for values of $n_i = 0, 1$ in sextuple precision and use recursion relations for $n_i > 1$ [13]. Results for nonrelativistic energies obtained with the largest number of basis functions, namely 9576 are so far, the lowest ones in the literature

$$\mathcal{E}(2^2S_{1/2}) = -7.478\,060\,323\,890, \tag{7}$$

$$\mathcal{E}(3^2S_{1/2}) = -7.354\,098\,421\,380, \tag{8}$$

and finite mass corrections are presented in Table I. The high quality of the nonrelativistic wave function allows one to obtain accurate results for matrix elements, for example

$$\Big\langle \sum_a \delta^3(r_a) \Big\rangle_{3S} - \Big\langle \sum_a \delta^3(r_a) \Big\rangle_{2S} = -0.106\,108\,0\,. \tag{9}$$

Relativistic corrections are obtained from the Breit-Pauli Hamiltonian, which includes here finite mass corrections [8]

$$\begin{aligned}\mathcal{E}^{(4)} &= \sum_a \biggl\langle -\frac{\vec p_a^{\,4}}{8} + \frac{\pi\,Z}{2}\,\delta^3(r_a) \\ &- \frac{m}{M}\frac{Z}{2}\,p_a^i\left(\frac{\delta^{ij}}{r_a}+\frac{r_a^i\,r_a^j}{r_a^3}\right)\sum_b p_b^j\biggr\rangle_M \\ &+ \sum_{a>b}\biggl\langle \pi\,\delta^3(r_{ab}) - \frac{1}{2}\,p_a^i\left(\frac{\delta^{ij}}{r_{ab}}+\frac{r_{ab}^i\,r_{ab}^j}{r_{ab}^3}\right)p_b^j\biggr\rangle_M,\end{aligned} \tag{10}$$

where $\langle\ldots\rangle_M$ denotes a matrix element, calculated with the wave function that includes the finite nuclear mass. Matrix elements for $\mathcal{E}^{(4)}$ involve extended Hylleraas integrals, where one n_i becomes negative. High precision calculations of Hylleraas integrals with inverse powers of r_a and r_{ab} are quite difficult and are one of the principal achievements of this work. We use the integration by parts identities to express $f(-1,0,0;0,0,0)$ in terms of one-dimensional integrals which are performed with 48 digit accuracy, and $f(-1,n_2,n_3;n_4,n_5,n_6)$ are obtained by recursion relations [14]. Similarly, $f(0,0,0,-1,n_5,n_6)$ are expressed as an one-dimensional integral which is performed numerically, and $f(n_1,n_2,n_3,-1,n_5,n_6)$ are obtained by recursion relations [12]. Results for relativistic recoil corrections are presented in Table I. We note a strong cancellations between reduced mass, mass polarization and the direct electron-nucleus Breit interaction. Our result of 0.038 MHz is more than 10 times smaller than the previous one in [8, 9], but it is in agreement with the recent recalculation by Yan and Drake [15] which gives $0.024(19)$ MHz.

Leading QED recoil corrections $E^{(5)}$ have been studied in [16] and calculated for lithium in [9, 10]. They can be represented (in atomic units) as:

$$\begin{aligned}\mathcal{E}^{(5)} &= \left[\frac{19}{30} - 2\,\ln\alpha - \ln k_{0M}\right]\frac{4\,Z}{3}\Bigl\langle\sum_a \delta^3(r_a)\Bigr\rangle_M + \left[\frac{164}{15}+\frac{14}{3}\,\ln\alpha\right]\Bigl\langle\sum_{a>b}\delta^3(r_{ab})\Bigr\rangle_M - \frac{7}{6\,\pi}\Bigl\langle\sum_{a>b}\frac{1}{r_{ab}^3}\Bigr\rangle_M \\ &+\frac{m}{M}\left[\left(-\frac{2}{3}\,\ln\alpha+\frac{62}{9}-\frac{8}{3}\,\ln k_0\right)\Bigl\langle\sum_a\delta^3(r_a)\Bigr\rangle - \frac{7\,Z^2}{6\,\pi}\Bigl\langle\sum_a\frac{1}{r_a^3}\Bigr\rangle\right] + O\Bigl(\frac{m}{M}\Bigr)^2,\end{aligned} \tag{11}$$

where $\ln k_0$ is the Bethe logarithm and $1/r^3$ has implicit subtraction of $\ln\varepsilon+\gamma$, with ε being the cutoff and γ the Euler constant. $1/r^3$ terms and $\ln k_0$ including mass polarization corrections have been calculated by Yan and Drake in [9, 10], and we use their results here. The complete $m\,\alpha^5$ contribution is presented in Table I. It is slightly different from that in [9, 10], due to including the additional term from reduced mass scaling of $1/r_{ab}^3$ in Eq. (11).

TABLE I: Contributions to Li isotope shift, $\eta = -\mu/M$

contribution	$3^2S_{1/2} - 2^2S_{1/2}$	^{11}Li$-^7$Li (MHz)
$m\,\alpha^2\,\eta$	0.133 764 842	25 104.489(21)
$m\,\alpha^2\,\eta^2$	0.123 659 8	-2.968
$m\,\alpha^4\,\eta$	0.003 78(3)	0.038
$m\,\alpha^5\,\eta$	-1.43	-0.104
$m\,\alpha^6\,\eta$	$-38.(10)$	$-0.020(5)$
$E_{\rm pol}$		0.039(4)

Considering higher order recoil corrections $m^2/M\,\alpha^6$, they are known exactly only for hydrogenic systems [17–19]. Some of them, are known to be proportional to the Dirac delta function, such as the radiative or radiative recoil, so the extension to lithium is simple. Pure recoil corrections are more complicated. There are state dependent terms which are known for hydrogen [17, 19], but the extension to more than one-electron atoms has not yet been achieved. We neglect them here and associate roughly 25% uncertainty. This correction is already quite small, so the approximate treatment is sufficient. The formula we use for $\mathcal{E}^{(6)}$ is

$$\begin{aligned}\mathcal{E}^{(6)} &\approx \biggl[\left(\frac{\mu}{m}\right)^3 Z^2\left(\frac{427}{96}-2\,\ln 2\right) \\ &+ \frac{m}{M}\,Z^2\left(\frac{35}{36}-\frac{448}{27\,\pi^2}-2\,\ln 2+\frac{6\,\zeta(3)}{\pi^2}\right) \\ &+ \frac{m}{M}\,Z^3\left(4\,\ln 2-\frac{7}{2}\right)\biggr]\,\pi\,\Bigl\langle\sum_a\delta^3(r_a)\Bigr\rangle,\end{aligned} \tag{12}$$

and numerical results using Eq. (9) are presented in Table I, in agreement with the estimation from Ref. [9].

Apart from relativistic and QED effects there is a finite nuclear size and nuclear polarizability which contribute to the isotope shift. Since one uses the isotope shift measurement to determine nuclear charge radius, one shall estimate the effect of nuclear polarizability, which we find here to be significant for ^{11}Li. For this we assume that the interaction of the nucleus with the electromagnetic field can be described as follows

$$H_{\rm int} = q\,A^0 - \vec d\cdot\vec E - \vec\mu\cdot\vec B + \frac{q}{6}\,\langle r^2\rangle\,\nabla^2 A^0. \tag{13}$$

The dominating nuclear excitations, are $E1$ transitions by the electric dipole coupling $-\vec d\cdot\vec E$ [20]. The energy shift due to the two-photon exchange in the temporal gauge is

$$E_{\rm pol} = i\,e^2\,\psi^2(0)\int\frac{d\,\omega}{2\,\pi}\int\frac{d^3k}{(2\,\pi)^3}\,\omega^2\,\frac{\left(\delta^{ik}-\frac{k^i\,k^k}{\omega^2}\right)}{\omega^2-k^2}\,\frac{\left(\delta^{jl}-\frac{k^j\,k^l}{\omega^2}\right)}{\omega^2-k^2}$$
$$\times{\rm Tr}\left[\left(\gamma^j\,\frac{1}{\not p-\not k-m}\gamma^i+\gamma^i\frac{1}{\not p+\not k-m}\,\gamma^j\right)\frac{(\gamma^0+I)}{4}\right]\left\langle\phi_N\left|d^k\,\frac{1}{E_N-H_N-\omega}\,d^l\right|\phi_N\right\rangle, \tag{14}$$

where $\psi^2(0) = (m\,\alpha)^3\,\langle\sum_a\delta^3(r_a)\rangle$, $p = (m,\vec{0})$, we used plane wave approximation for the electrons, since the characteristic photon momentum k is much larger than the inverse Bohr radius. After performing k integration and replacing $\omega = i\,w$ one obtains

$$E_{\rm pol} = -m\,\alpha^4\left\langle\sum_a\delta^3(r_a)\right\rangle(m^3\,\tilde{\alpha}_{\rm pol}), \tag{15}$$

where $\tilde{\alpha}_{\rm pol}$ is a kind of electric polarizability of the nucleus, which is given by the following double integral

$$\tilde{\alpha}_{\rm pol} = \frac{16\,\alpha}{3}\int_{E_T}^{\infty}dE\,\frac{1}{e^2}\,|\langle\phi_N|\vec{d}|E\rangle|^2\int_0^{\infty}\frac{dw}{w}\,\frac{E}{E^2+w^2}$$
$$\times\frac{1}{(\kappa+\kappa^\star)}\left[1+\frac{1}{(\kappa+1)(\kappa^\star+1)}\left(\frac{1}{\kappa+1}+\frac{1}{\kappa^\star+1}\right)\right] \tag{16}$$

where $\kappa = \sqrt{1+2\,i\,m/w}$, and E is the excitation energy of the nucleus with respect to the ground state with threshold value E_T. This general formula agrees in the limit $E \gg m$ with that derived previously for the nuclear polarizability effect in deuterium [21]. $\tilde{\alpha}_{\rm pol}$ involves a square of the transition dipole moment. This can be related to the $B(E1)$ distribution which was recently accurately measured for ^{11}Li in Ref. [20]

$$|\langle\phi|\vec{d}|E\rangle|^2 = \frac{4\,\pi}{3}\,\frac{dB(E1)}{dE} \tag{17}$$

in units e^2 fm^2 MeV, what explains the presence of e^2 in the denominator in Eq. (16). With the new data from Ref. [20], see Fig. 1, and the two-neutron separation energy $E_T = 0.376(5)$ MeV [7] one obtains

$$\tilde{\alpha}_{\rm pol} = 60.9(6.1)\,{\rm fm}^3 = 1.06(0.11)\,10^{-6}\,m^{-3}, \tag{18}$$

and a polarizability correction to the $3^2S_{1/2} - 2^2S_{1/2}$ transition frequency of $\nu_{\rm pol} = 39(4)\,$kHz. Polarizability correction for ^{9} Li and lighter isotopes is expected to be at least 10 times smaller, and is thus negligible.

The last significant contribution to the isotope shift is due to the finite size of the nucleus

$$E_{\rm fs} = \frac{2\,\pi}{3}\,Z\,\alpha^4\,m^3\,\langle r_{\rm ch}^2\rangle\,\langle\sum_a\delta^3(r_a)\rangle. \tag{19}$$

This nuclear volume effect can now be extracted from the isotope shift measurements, to obtain nuclear charge radii. We additionally account for the leading relativistic correction $\phi^2(0)\,[1-(Z\,\alpha)^2\,\ln(m\,r_{\rm ch}\,Z\,\alpha)]$ to the square of the wave function at the origin. Results are summarized in Table II.

TABLE II: Summary of isotope shift determination of Li charge radii, $r_{\rm ch}(^7{\rm Li})$ = 2.390(30) fm [22], $m(^7{\rm Li})$ = 7.016 003 425 6(45) u [23], the first uncertainty of $\nu_{\rm the}$ comes from unknown higher order terms, the second uncertainty is due to the atomic mass.

	mass(u) [7, 24]	$\nu_{\rm exp}$(MHz) [3, 4]	$\nu_{\rm the}$(MHz)	$\delta r^2_{\rm ch}$(fm^2)	$r_{\rm ch}$(fm)
^{6}Li	6.015 122 794(16)	−11 453.983(20)	−11 452.822(2)(0)	0.738(13)	2.540(30)
^{8}Li	8.022 487 36(10)	8 635.782(44)	8 634.990(1)(1)	−0.503(28)	2.282(32)
^{9}Li	9.026 789 5(21)	15 333.272(39)	15 331.797(3)(13)	−0.938(26)	2.185(33)
^{11}Li	11.043 715 7(54)	25 101.226(125)	25 101.473(9)(21)	0.157(81)	2.423(34)

All results for charge radii differences are significantly improved compared to previous determinations, for example, our result for the difference ^{11}Li - ^{7}Li, $\delta r^2_{\rm ch} = 0.157(81)$ fm^2 can be compared to $\delta r^2_{\rm ch} = 0.374(112)$ fm^2 which is obtained using results presented in Ref. [4]. The result for the difference between ^{11}Li and ^{9}Li, $\delta r^2_{\rm ch} = 1.09(8)$ fm^2 does not agree

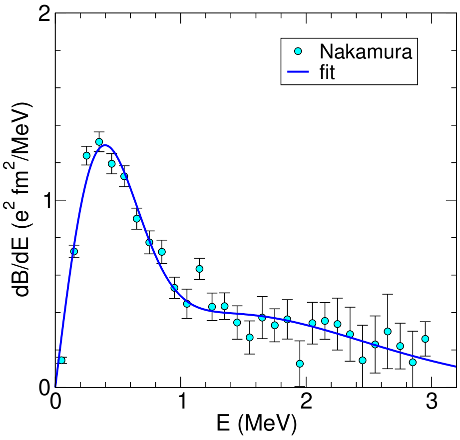

FIG. 1: Electric dipole line strength by Nakamura *et al.* [20] adapted to the new value of E_T from [7].

with that obtained on the basis of $B(E1)$ data, namely

$$\delta r^2 = \frac{4\,\pi}{3}\,\frac{1}{(Z\,e)^2}\int_{E_T}^{\infty} dE\,\frac{dB(E1)}{dE} = 0.74\,\mathrm{fm}^2. \tag{20}$$

δr^2 here is the square of a distance of ^{9}Li core from the mass center. From this, one may conclude, that the ^{9}Li core is significantly perturbed by the presence of the two valence neutrons in ^{11}Li. A similar conclusion has already been drawn in Ref. [4].

The difference in squared charge radii between ^{6}Li and ^{7}Li can also be obtained from the isotope shift measurement of $2^3S - 2^3P_J$ transitions in Li$^+$ by Riis *et al.* [25]. Taking the weighted average value and theoretical isotope shift from [26], one obtains $\delta r^2_{\rm ch} = 0.702(60)$ fm^2, in a moderate agreement with the result obtained here $\delta r^2_{\rm ch} = 0.742(13)$ fm^2.

The uncertainty of numerical calculations for lithium-like atoms is negligible in comparison to the estimation of unknown higher order terms. The pure recoil correction of order $\alpha^6\,m^2/M$ gives the largest theoretical uncertainty of a few kHz for lithium isotope shifts, and will become more significant for heavier lithium-like ions. At present, however, the dominating source of uncertainties comes from measurement of transition frequencies and from the charge radius of the reference nucleus, see Table II. Regarding the direct charge radius determination of the reference nucleus ^{7}Li, calculations of transition frequencies are far more difficult and have been performed with the sufficient precision only for hydrogenic systems. Therefore, the spectroscopic determination of the absolute charge radius can possibly be achieved by measurement of two-photon transitions in the hydrogen-like lithium ion or 2S-2P transition in the muonic lithium.

In summary, we have obtained high precision finite nuclear mass corrections to lithium isotope shifts and nuclear polarizability (for ^{11}Li). The resulting nuclear charge radii are significantly improved over the former determinations. Our approach is based on an analytical calculation of matrix elements with Hylleraas wave functions, which can easily be applied to lithium like ions, and, we think, can be extended to 4-electron systems as well.

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[1] A. Huber, *et al.*, Phys. Rev. Lett. **80**, 468 (1998).
[2] L.-B. Wang, *et al.*, Phys. Rev. Lett. **93**, 142501 (2004).
[3] G. Ewald, *et al.*, Phys. Rev. Lett. **93**, 113002 (2004).
[4] R. Sánchez, *et al.*, Phys. Rev. Lett. **96**, 033002 (2006).
[5] V.A. Yerokhin, P. Indelicato, and V.M. Shabaev, Zh. Exp. Theor. Phys. **128**, 322 (2005), [JETP **101**, 280 (2005)].
[6] T. Udem, *et al*, Phys. Rev. Lett. **79**, 2646 (1997).
[7] C. Bachelet, Eur. Phys. J. A **25**, 31 (2005).
[8] Z-C. Yan and G.W.F. Drake, Phys. Rev. A **61**, 022504 (2000).
[9] Z-C. Yan and G.W.F. Drake, Phys. Rev. A **66**, 042504 (2002).
[10] Z-C. Yan and G.W.F. Drake, Phys. Rev. Lett. **91**, 113004 (2003).
[11] Z.-C. Yan and G.W.F. Drake, Phys. Rev. A **52**, 3711 (1995); Z.-C. Yan, M. Tambasco, and G.W.F. Drake, Phys. Rev. A **57**, 1652 (1998).
[12] M. Puchalski and K. Pachucki, Phys. Rev. A **73**, 022503 (2006).
[13] K.Pachucki, M. Puchalski and E. Remiddi, Phys. Rev. A **70**, 032502 (2004).
[14] K.Pachucki and M. Puchalski, Phys. Rev. A **71**, 032514 (2005).
[15] G.W.F. Drake, *private communication*.
[16] K. Pachucki, J. Phys B **31**, 5123 (1998).
[17] K. Pachucki and H. Grotch, Phys. Rev. A **51**, 1854 (1995).
[18] K. Pachucki Phys. Rev. A **52**, 1079 (1995).
[19] M.I. Eides, H. Grotch, and V.A. Shelyuto, Phys. Rep. **342**, 63 (2001).
[20] T. Nakamura, *et al.*, Phys. Rev. Lett. **96**, 252502, (2006).
[21] K. Pachucki, D. Leibfried, and T.W. Hänsch, Phys. Rev. A **48**, R1 (1993); K. Pachucki, M. Weitz, and T.W. Hänsch, Phys. Rev. A **49**, 2255 (1994).
[22] C.W. de Jager, H. deVries, and C. deVries, At. Data Nucl. Data Tables **14**, 479 (1974).
[23] Sz. Nagy, *et al.*, Phys. Rev. Lett. **96**, 163004 (2006).
[24] G. Audi, A.H. Wapstra, and C. Thibault, Nucl. Phys **A729**, 337 (2003), http://amdc.in2p3.fr/masstables/Ame2003.
[25] E. Riis, A.G. Sinclair, O. Poulsen, G.W.F. Drake, W.R.C. Rowley, and A.P. Levick, Phys. Rev. A **49**, 207 (1994).
[26] G.W.F. Drake, W. Nörtershäuser, and Z.-C. Yan, Can. J. Phys. **83**, 311 (2005).